# PHONONS 2023

# XVII INTERNATIONAL CONFERENCE ON PHONON SCATTERING IN CONDENSED MATTER

PARIS, July 2<sup>nd</sup> - 7<sup>th</sup>, 2023



**BOOK OF ABSTRACTS** 

# Topics

- Light, neutron and x-ray inelastic scattering
- Nano- and micro-scale phonon transport
- Phonons and phonon-phonon interactions in nanoscale materials and structures
- Coherent phonons and ultrafast acoustics
- Lattice dynamics
- Electron-phonon interactions
- Optomechanics
- Phonons in MEMS and NEMS and optomechanical systems
- Phonons in glasses and disordered materials
- Phononic crystals
- Phonon laser (saser)
- Phonons in biological materials
- Phonons in emerging two-dimensional systems
- Quantum nanoacoustics
- Surface acoustic waves, science and technology
- Topological phononics
- Nonlinear phonon phenomena
- Phonon applications in quantum technologies
- New phonon techniques, materials and phenomena

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#### PHONONS CONFERENCE SERIES

The history of the Phonons Series (Conference on Phonon Scattering in Condensed Matter) is longstanding, since 1972. Over the past 50 years it took place in the following venues:

- Phonons 2018, Nanjing, China. Chaired by Lifa Zhang.
- Phonons 2015, Nottingham, United Kingdom. Chaired by Tony Kent.
- Phonons 2012, Ann Arbor, United States of America. Chaired by Kevin Pipe.
- Phonons 2010, Taipei, Taiwan. Chaired by Chi Kuang Sun.
- Phonons 2007, Paris, France. Chaired by Bernard Perrin.
- Phonons 2004, St. Petersburg, Russia. Chaired by Alexander A. Kaplyanskii.
- Phonons 2001, Hanover, USA. Chaired by Martin Wybourne.
- Phonons 1998, Lancaster, England. Chaired by John Wigmore.
- Phonons 1995, Sapporo, Japan. Chaired by Tsuneyoshi Nakayama.
- Phonons 1992, Ithaca, USA. Chaired by R. O. Pohl.
- Phonons 1989, Heidelberg, Germany. Chaired by Siegfried Hunklinger, Wolfgang Ludwig, Georg Weiss
- Phonons 1986, Urbana, USA. Chaired by A. C. Anderson and J. P. Wolfe.
- Phonons 1983, Stuttgart, Germany. Chaired by W. Eisenmenger.
- Phonons 1979, Providence, USA. Chaired by Humphrey J. Maris.
- Phonons 1975, Nottingham, UK. Chaired by L.J Challis, V. W. Rampton and A.F.G. Wyatt
- Phonons 1972, St. Maxime and Paris, France. Chaired by H. J. Albany.

#### INSTRUCTIONS FOR ORAL AND POSTER PRESENTATIONS

#### **Poster Session Instructions:**

- Two poster sessions will take place, one on Tuesday and the other on Thursday.
- The schedule with the assigned order of participants for each day will be provided soon.
- The poster size must be in the standard A0 format, portrait.

#### **Oral Presentation Instructions:**

- Duration of each talk
  - Plenary talks: 45 minutes, with questions included
  - Invited talks: 30 minutes, with questions included
  - o Contributed talks: 20 minutes, with questions included
- The amphitheaters are equipped with video projectors set to the 16:9 aspect ratio.
- The presentations must be uploaded at the local computer (in .pptx or .pdf format) before the session starts
- Please ensure that you adhere to the designated time slot for your talk, as any delays may disrupt the subsequent sessions.

#### VENUE MAP

The Amphitheaters Jean Fourastié (JF) and Jean Prouvé (JP) at the Conservatoire National de Arts et Métiers (CNAM) will host the Plenary sessions, contributed and invited talks.

For the poster sessions, the Salon d'Honneur (SH) will be the designated location. During the entire poster session, attendees can enjoy cold drinks and appetizers.

Please refer to the CNAM map below for directions to the Amphitheaters and the poster session venue.



ER.

#### SOCIAL EVENTS

#### WELCOME RECEPTION

The Welcome Reception will take place at Le Bateau Phare, a charming boat at the Seine riverside.





Venue: Le Bateau Phare

**Date:** Sunday (02/07), from 19h to 21h

Address: <u>3 Port de la Gare, 75013 Paris</u>

**General instructions:** Le Bateau Phare has easy access via the metro lines M6 and M14, and train line RER C.

6 🚺 <mark>C</mark>

From the conference venue (CNAM) to Le Bateau Phare:

- Take the metro M11 at Arts et Metiers, direction Chatelet
- Change to metro line M14 at Chatelet station, direction Olympiades
- Stop at Bibliothèque François-Mitterrand
- Exit via Av. de France
- Head northeast on Rue Neuve Tolbiac
- Turn left onto Quai François Mauriac

#### **CONFERENCE DINNER**

The Conference Dinner will take place at Le Bouillon Racine, a restaurant situated in the 6th arrondissement, founded in 1906, and listed as a historical monument in 1995.





Venue: Bouillon Racine

**Date:** Monday (03/07), from 20h to 23h

Address: 3 Rue Racine, 75006 Paris

**General instructions:** Le Bouillon Racine has easy access via the metro lines: M4 and M10, and train lines RER B and RER C.



The easiest way to reach the Bouillon Racine from the conference venue (CNAM) is by metro:

- Take the metro M4 at Réaumur - Sébastopol, direction Bagneux
- Stop at Odéon station
- Exit via Rue de L'école de Médecine
- Turn right onto Rue Racine

# PROGRAM

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Phonons 2023

# **Scientific Program**

#### Monday, 3<sup>rd</sup> July

Mo. 9:15 Opening Ceremony

#### **Plenary Session**

Mo. 9:30 Alexander BALANDIN Thermal Conductivity of Graphene – Twists and Turns in Understanding Phonon Transport in Quasi-Two-Dimensional Materials

#### Sesssion MO1: SAW, phononics, nanoacoustics

- Mo. 10:40Claire Prada (invited)Backward modes and local resonances in anisotropic elastic waveguides
- Mo. 11:10 Andrea Bragas Coherent acoustic phonons as elasticity and adhesion local sensors in silica mesoporous thin films
- Mo. 11:30Paulo SantosPolariton-based GHz optomechanics with electrically driven guided phonons
- Mo. 11:50Chushuang XiangGHz acoustic phonon transport in optophononic waveguides

#### Session MO2: New phonon techniques, materials and phenomena

Mo. 10:40	Eduardo Gil-Santos <b>(invited)</b> Optomechanical detection of single bacterium phonons
Mo. 11:10	Hamadou Dicko Coherent Raman scattering microscopy for the study of biological organisms: valuable analytical tool
Mo. 11:30	Yann Chalopin Wave Localization in Disordered Phonon Systems and the Emergence of Basic Biological Functions
Mo. 11:50	Laurent Belliard Correlative Imaging of Motoneuronal Cell Elasticity by Pump and Probe Spectroscopy

#### **Plenary Session**

Mo. 14:00 Eva WEIG Coherent control and nonlinear dynamics with nanomechanical string resonators

#### Session MO3: Optomechanics, nanomechanics, NEMS-MEMS

- Mo. 14:50 Sunil Bhave (invited)
- Mo. 15:20 Julien Claudon Exploring the high-frequency mechanical resonances of a quantum dot–microwire hybrid system
- Mo. 15:40 Andrey Akimov Flexural phononic crystals in the structures with van der Waals nanolayers

#### Session MO4: Time resolved phononics and nonlinear phononics

- Mo. 14:50 Hanyu Zhu (invited) Chiral phonon-induced spin polarization
- Mo. 15:20 Bernard Perrin Acoustic phonon dynamics in Bi<sub>2</sub>Se<sub>3</sub> thin films
- Mo. 15:40 Silvia Bocatto Picosecond acoustic measurements at extreme pressure and temperature conditions

#### Session MO5: Phonon - magnon / exciton / polariton interactions, 2D materials, multifunctional materials

- Mo. 16:30 Andrew Rushforth (invited) Magnon – phonon coupling in magnetostrictive nanostructures
   Mo. 17:00 Youssef El Helou Detection of Surface Phonon Polaritons by Photo-Induced Force Microscopy
- Mo. 17:20 Michal Kobecki Giant Photo-Elasticity of the Superlattice Polaritons for Detection of Coherent Phonons

#### Session MO6: Thermal transport and nanothermal properties

- Mo. 16:30 Riccardo Rurali **(invited)** *From electrophononics to photophononics: controlling heat flux with external fields*
- Mo. 17:00 Aurélien Crut Cooling Dynamics of Single Substrate-Supported Gold Nanodisks

#### Tuesday, 4<sup>th</sup> July

#### Session TU1: Phonon - magnon / exciton / polariton interactions, 2D materials, multifunctional materials

Tu. 14:50	Jouni Ahopelto <b>(invited)</b> Optoelectromechanics Exploiting Nanocrystalline Silicon
Tu. 15:20	Pauline Rovillain Impact of spin waves dispersion on surface acoustic waves and ferromagnetic resonance in iron thin films
Tu. 15:40	Riccardo Cucini Surface acoustic waves seeding for magnon generation in a thin film

#### Session TU2: Thermal transport and nanothermal properties

Tu. 14:50	Zhiting Tian <b>(invited)</b> Thermal Phonon Transport and Phonon-Magnon Interactions
Tu. 15:20	Raja Sen Role of dimensionality, size, and transport-direction in governing the drag Seebeck coefficient of doped silicon nanostructures: A first-principles study
Tu. 15:40	Paola Giura Effect of phonon anharmonicity on MgO lattice dynamics at extreme conditions: Impact on

#### Session TU3: SAW, phononics, nanoacoustics

thermal conductivity

Tu. 16:30	Sarah Benchabane <b>(invited)</b> Sub-wavelength confinement of surface acoustic waves induced by collective coupling of phononic resonators
Tu. 17:00	Kevin Silverman A traveling-wave broadband SAW transducer based on superconducting nanowires
Tu. 17:20	Mingyun Yuan GHz Surface-Acoustic-Wave Resonators Probed by Atomic-Force Microscopy on AlN Layers Grown on 6H-SiC(0001) by Molecular Beam Epitaxy
Tu. 17:40	Aswathi Kanjampurath Sivan Phonon engineering in Heterostructure Superlattice Nanowires

#### Session TU4: New phonon techniques, materials and phenomena

Tu. 16:30	Thomas Dehoux <b>(invited)</b> Probing molecular crowding in cells using Brillouin spectroscopy
Tu. 17:00	Aymeric Ramiere Thermoelectric magnetotransport quantum oscillations in topological semimetal ZrTe5
Tu. 17:20	Mathieu Ducousso Thermo-elastic evaluation of single micrometric carbon fibers by picosecond acoustic pulses
Tu. 17:40	Simon Ayrinhac Thermoelastic properties of liquid metals measured by picosecond acoustics

Tu. 18:00 Poster Session A

#### Wednesday, 5<sup>th</sup> July

#### **Plenary Session**

We. 9:30 Chris BAUERLE Flying electron qubits using sound waves

#### Sesssion WE1: SAW, phononics, nanoacoustics

We. 10:40	Marta de Luca <b>(invited)</b> Phonon spectroscopy and phonon engineering in semiconductor nanowires
We. 11:10	Martin Esmann <b>(invited)</b>
	Topological confinement and robust light-matter interaction of ultra-high frequency phonons
We. 11:40	Habib Rostami
	Strain and Layer-Twist Engineering of Valley Phonons in 2D Materials
We. 12:00	Changxiu Li
	Sub-THz Rayleigh and Bulk Surface Skimming Acoustic Modes on Superlattices

#### Session WE2: Thermal transport and nanothermal properties

We. 10:40	Konstantinos Termentzidis
	Phonons Interference Using Nano-Composites

# We. 11:10Giuseppe RomanoRecent Advances in Solving the Space-Dependent Phonon Boltzmann Transport Equation: from<br/>Materials Prediction to Inverse Design

- We. 11:40Guillaume NatafLarge anisotropic thermal conductivity variations controlled by ferroelastic domain walls
- We. 12:00Xin HuangRapid heat flow driven by collective phonons in 12C enriched sub-Im graphite

#### Session WE3: New phonon techniques, materials and phenomena

We. 14:15	Hubert Krenner <b>(invited)</b> Interfacing quantum dots in coherent elastic waves devices
We. 14:45	laroslav Mogunov Laser-induced phase transitions for ultrafast acoustics
We. 15:05	Monty Clark Apertures for generating spatial superoscillations of coherent acoustic phonons
We. 15:25	Filippo Bencivenga Extreme ultraviolet transient gratings: a new tool for nanoscale thermoelasticity
We. 15:45	Régis Decker Using X-ray emission spectroscopy to measure the electron-phonon scattering rates in the demagnetization transient state of ferromagnets

#### Session WE4: Thermal transport and nanothermal properties

We. 14:15	Jose Ordonez Miranda <b>(invited)</b> Conductive Heat Shuttling Driven by Thermal Waves in VO <sub>2</sub>
We. 14:45	Akash Patil Raman thermometry characterization of GeSbTe based phase change materials
We. 15:05	Xiaokang Li Phonon thermal Hall effect in black phosphorus
We. 15:25	Chaitanya Arya Thermal conductivity of GaAs-GaP superlattice nanowires using the thermal bridge method
We. 15:45	Sebin Varghese Direct Observation of Heat Diffusion in Suspended Transition Metal Dichalcogenides

#### Session WE5: Phonon - magnon / exciton / polariton interactions, 2D materials, multifunctional materials

We. 16:40Ilya Akimov (invited)Magnetic proximity effect in hybrid ferromagnet-semiconductor structures mediated by chiral<br/>phonons

- We. 17:10 Eiji Saitoh (invited) Phonons in spintronics
- We. 17:40Mads C. WeberEmerging spin-phonon coupling through cross-talk of two magnetic sublattices

#### Session WE6: Time resolved phononics and nonlinear phononics

- We. 16:40Ryan Duncan (invited)Ultrafast x-ray scattering reveals composite amplitude collective mode in Weyl charge density<br/>wave material (TaSe4)2l
- We. 17:10Chi Kuang Sun (invited)THz Time-Domain Phonon Spectroscopy for van der Waals Coupling of 2D MoS2 Interfaces
- We. 17:40Danny FainozziHard X-ray transient grating spectroscopy

#### Thursday, 6<sup>th</sup> July

#### **Plenary Session**

Th. 9:30Alex FAINSTEINMetamaterials of fluids of light and sound

#### Sesssion TH1: Optomechanics, nanomechanics, NEMS-MEMS

Th. 10:40	Warwick Bowen <b>(invited)</b> Towards quantum control of a room temperature mechanical resonator
Th. 11:10	Xin Zhou Coupled membrane nanomechanical resonators, for room temperature phonon-cavity electromechanics
Th. 11:30	Kyrylo Gerashchenko Quantum control of an ultracoherent mechanical resonator with a fluxonium qubit
Th. 11:50	Christophe Galland <b>(invited)</b> <i>Molecular Optomechanics</i>

#### Session TH2: Time resolved phononics and nonlinear phononics

Th. 10:40	Vincent Juvé <b>(invited)</b>
	Picoseconds lattice dynamics in multiferroic BiFeO3 probed by time-resolved x-ray diffraction
Th. 11:10	Pascal Ruello
	Polar super-orders and folded acoustic phonons in BiFeO $_3$ /LaFeO $_3$ superlattices

- Th. 11:30Samuel RaetzTime-domain Brillouin scattering for the evaluation of materials interface inclination:<br/>Application to imaging of crystal degradation upon non-hydrostatic compression
- Th. 11:50Grazia Raciti<br/>Exploring hydrodynamic heat transport in 2D materials using ultrafast spectroscopy

#### Session TH3: Brillouin and Raman spectroscopies

Th. 14:00	Bartlomiej Graczykowski Thickness-dependent Elastic Softening of Few-layer Free-standing MoSe2
Th. 14:30	Takuya Satoh Chiral Phonons in Chiral Materials by Circularly Polarized Raman Spectroscopy
Th. 14:50	Butian Zhang Electrochemical Modulation of Resonance Raman Intensities in Quantum Dots
Th. 15:10	Jérémie Margueritat Environmental effects on the inelastic light scattering efficiency by acoustic vibration modes of a single nanocube
Th. 15:30	Philipp Stein

Local Symmetry Breaking and Low-Energy Continuum in K<sub>2</sub>ReCl<sub>6</sub>

#### Session TH4: Thermal transport and nanothermal properties

Th. 14:00	Valentina Giordano <b>(invited)</b> Thermal relaxation and phonon lifetime in a nanophononic SiN suspended membrane
Th. 14:30	Ahmad Zenji Thermal Characterization of Nanostructured Model Materials Through FDTR Broadband Response
Th. 14:50	Jelena Sjakste Electron-phonon coupling and ultrafast dynamics of hot carriers in semiconductors: from interpretation of photoemission experiments to transport simulations in devices
Th. 15:10	Jay Amrit Oscillatory Lévy walk of quasi-ballistic phonons in nanowires induced by surface roughness
Th. 15:30	Wilken Seemann Thermal Conductivity of c-plane GaN Membranes Studied by One- and Two-Laser Raman Thermometry

#### XXII

#### Session TH5: SAW, phononics, nanoacoustics

Th. 16:10	Anthony Kent <b>(invited)</b> High-speed modulation of a THz quantum cascade laser using coherent phonon pulses	
Th. 16:40	Clivia Sotomayor-Torres Generation and modulation of phononic signals based on a slotted phonon waveguide	
Th. 17:00	Alexey Scherbakov Neuromorphic computing with coherent acoustic phonons	
Th. 17:20	Edson Cardozo de Oliveira Design of optophononic devices based on SiO2 and TiO2 mesoporous materials for sensing applications	
Session TH6: Thermal transport and nanothermal properties		
Th. 16:10	Wu Li <b>(invited)</b> Large lattice thermal conductivity in several metals and semimetals	
Th. 16:40	Yashpreet Kaur	

- Telescopic nanowires for thermal rectification
- Th. 17:00Sebastian VolzPhonon Temporal Coherence in Heat Conduction
- Th. 18:00 Poster Session B

#### Friday, 7<sup>th</sup> July

#### **Plenary Session**

Fr. 9:15	Andrea ALU
	Extreme light control mediated by nanoscale phonon-photon interactions

#### **Plenary Session**

Fr. 10:00 Hiroshi YAMAGUCHI

#### **Plenary Session**

Fr. 11:05 Alexei MAZNEV Diffusive-to-ballistic transition and second sound in phonon-mediated thermal transport on micro/nano scale

#### **Plenary Session**

- Fr. 11:50 Lifa ZHANG Chiral phonons and related novel effects
- Fr. 12:35 Farewell

#### Phonons 2023

# ABSTRACTS

XXIII

# Thermal Conductivity of Graphene – Twists and Turns in Understanding Phonon Transport in Quasi-Two-Dimensional Materials

#### Alexander A. Balandin

University of California at Los Angeles, Los Angeles, California, 90095 U.S.A. <u>balandin@seas.ucla.edu</u>

The discovery of unique heat conduction properties of graphene [1] motivated research focused on the thermal conductivity of graphene, few-layer graphene, and composites with graphene fillers [2]. Recent developments suggest that graphene's first large-scale practical applications are appearing in thermal management – thermal interface materials, thermal coatings, and related [3-4]. On the other side, it has been demonstrated that graphene composites are efficient electromagnetic interference shielding materials they can reflect and absorb electromagnetic energy efficiently even at a low loading fraction of graphene, below the percolation limit, while remaining electrically insulating [5]. The dual function of graphene composites and the possibility of independent control of their electrical and thermal properties provide extra benefits for practical applications [5]. In this talk, I reflect on the evolution of the understanding of the phonon thermal conductivity in quasi-two-dimensional (2D) graphene, addressing the issues of divergent thermal conductivity in strictly 2D systems [6-8] and the relative contribution of the longitudinal acoustic (LA), transverse acoustic (TA) and out-of-plane acoustic (ZA) phonons to the thermal conductivity [2, 4, 6-8]. The original Klemens model neglected ZA phonon contributions to the thermal conductivity of graphite and graphene due to ZA phonons' low group velocity and large anharmonicity [9]. Later, it has been suggested theoretically that due to phonon-scattering selection rules, ZA phonons are long-lived and can account for a significant portion of overall heat conduction, at least, in certain temperature ranges [10-11]. However, the recent development in this field was the exact solution to the linearized phonon Boltzmann transport equation, which includes three-phonon and four-phonon scattering processes [12]. It turned out that the reflection symmetry in graphene, which forbids the three-ZA phonon scattering, allows the four-ZA processes. As a result, the large phonon population of the low-energy ZA branch originated from the quadratic phonon dispersion leads to the four-phonon scattering rates, which are higher than the threephonon scattering rates at room temperature [12]. The relative contribution of the ZA phonon branch is correspondingly reduced to lower than those of the LA and TA branches [12]. Recently we witnessed another intriguing development indicating a different possible explanation for the exceptionally high thermal conductivity of graphene and few-layer graphene. It originated from the concept of the phonon hydrodynamic transport and the closely related phenomenon of the second sound [13-14]. I will conclude the talk with an outlook on the practical applications of graphene in thermal management [15-16].

#### References

- [1] A. A. Balandin, et al., Nano Lett., 8 (2008) 902
- [2] A. A. Balandin, Nature Mater., 10 (2011) 569
- [3] Y. Fu et al., 2D Mater., 7 (2019) 012001
- [4] A. A. Balandin, ACS Nano, 14 (2020) 5170
- [5] F. Kargar, et al., Adv. Electron. Mater., 5 (2019) 1800558
- [6] D. L. Nika, E. P. Pokatilov, and A. A. Balandin, phys. status solidi, 248 (2011) 2609
- [7] N. Bonini, et al., Nano Letters, 12 (2012) 2673
- [8] G. Fugallo, et al., Nano Letters, 14 (2014) 6109
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- [10] L. Lindsay, D. Broido and N. Mingo, Phys. Rev. B, 82 (2010) 115427
- [11] L. Lindsay and D. Broido, Phys. Rev. B, 81 (2010) 205441
- [12] T. Feng and X. Ruan, Phys. Rev. B, 97 (2018) 045202
- [13] A. Cepellotti, et al., Nature Communications, 6 (2015) 6400
- [14] Y. Machida, et al., Science, 367 (2020) 309
- [15] J. S. Lewis, et al., Nanotechnology, 31 (2021) 142003
- [16] Z. Ebrahim Nataj, et al., Nature Communications, 14 (2023) 3190

#### **Claire Prada**

Sylvain Mezil, Daniel A. Kiefer Institut Langevin, ESPCI Paris, Université PSL, CNRS, 75005 Paris, France claire.prada@espci.psl.eu

#### Abstract

In elastic waveguides, the coupling between shear and compression waves results in complex dispersion effects. As a consequence, negative phase velocity elastic guided modes are observed in most homogeneous waveguides like plates, tubes or ribbons. In the absence of attenuation or leakage, the minimum frequency of a backward branch corresponds to a Zero-Group-Velocity (ZGV) mode which is associated with a narrow local resonance. These ZGV resonances are very well observed using non-contact laser ultrasonic techniques (LUS) [1-3]. Several studies have demonstrated their usefulness in assessing local properties of materials such as thickness, Poisson's ratio or elastic anisotropy [4][5]. The remarkable properties of these backward and ZGV modes will be reviewed through miscellaneous examples. Special attention will be given to anisotropic plates where the frequency of the ZGV modes depends on the propagation direction, leading to quasi-resonances with energy propagation perpendicular to the wave vector. This effect of the elastic anisotropy will be explained through measurements made with LUS in the MHz frequency range on a silicon plate.

#### References

- [1] Laurent, Royer, Prada, Wave Motion, 51, (2014), 1011-1020
- [2] Laurent, Royer, Prada, J. Acoustical Soc. Am. 147, (2020), 1302–1310
- [3] Xie, Mezil, Otsuka, Tomoda, Laurent, Matsuda, Shen, Wright, Nature Comm. (2019), 10:2228.
- [4] Prada, Clorennec, Murray, Royer, J. Acoust. Soc. Am. 126, (2009), 620-625
- [5] Thelen, Bochud, Brinker, Prada, Huber, Nature Comm., (2021), 12:3597



**Figure 1:** Zero-Group-Velocity Lamb modes measured in a 525µm thick silicon plate after excitation by a ~1 mm diameter and 8 ns duration laser pulse at point (0, 0). The horizontal axis corresponds to the [110] silicon crystallographic axis.

# Coherent acoustic phonons as elasticity and adhesion local sensors in silica mesoporous thin films

#### Andrea V. Bragas\*

Hilario D. Boggiano, Juan I. Ramallo, Lin Nan, Emiliano Cortés, Stefan A. Maier, Gustavo Grinblat, María C. Fuertes, Paula C. Angelomé

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Understanding and measuring the mechanical properties of mesoporous oxide thin films are required to decide whether these nanomaterials are suitable to be integrated into heterogeneous devices, have enough mechanical integrity to withstand manufacturing and integration processes, and have durability and good adhesion in *in-operando* conditions. Various techniques are suitable to characterize the mechanical properties of nanoporous thin films [1], among which the well-established nanoindentation (NI) [2] method is one of the most widely used. In turn, ultrafast optics techniques, such as picosecond acoustics [3] or the recently introduced technique based on the frequency shift of nanoantennas (FRESA) [4], have the advantage of characterizing the dynamical mechanical response of nanomaterials in the GHz range where most devices operate.

In this work, we use FRESA to determine how porosity affects the value of the elastic modulus and the adhesion of nanoporous silica thin films and compare the results with measurements done by NI. FRESA is a femtosecond optical technique that uses coherent acoustic phonons optically generated in plasmonic nanostructures as a sensor of the mechanical properties of their surroundings (see Figure 1). Indeed, the phonon frequency shift produced when these mechanical resonators are buried inside the film allows extracting the local Young's modulus by comparing the experimental results with full FEM (finite element method) simulations. Also, recording the mechanical nanoresonator's quality factor enables the evaluation of the adhesion of the film to the surface of the nanoantenna. Finally, this technique is suitable for determining the elastic modulus of arbitrarily thin nanometer-thickness films since the substrate's presence does not alter the result as it occurs with NI, and provides nanometer spatial accuracy in determining the mechanical moduli.

#### References

- [1] D. F. Lionello et al., J Solgel Sci Technol, 101 (2022), 114.
- [2] J. I. Ramallo *et al.*, J Solgel Sci Technol, 102 (2022), 185.
- [3] C. Mechri et al., Eur Phys J Spec Top, 153 (2008), 211.
- [4] H. D. Boggiano, et al., ACS Photonics, 7 (2020), 1403.

#### Figure



**Figure 1:** (a) Generation and detection of coherent acoustic phonons, using a two-color pump-probe experiment, in single plasmonic nanorods in air (top) and surrounded by a mesoporous oxide (MPO) film (bottom). (b) Frequency shift between the two conditions described in (a). (c) Young's modulus obtained by the frequency shift of nanoantennas method (FRESA) compared with the nanoindentation (NI) technique.



### Polariton-based GHz optomechanics with electrically driven guided phonons

#### Paulo V. Santos

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Electrically controlled hybrid opto-mechanical systems operating at several GHz offer extraordinary opportunities for the coherent control of opto-electronic excitations down to the quantum limit. In this contribution, we introduce a monolithic platform for GHz semiconductor optomechanics based on electrically excited phonons guided along the spacer of a planar (Al,Ga)As microcavity (MC) for exciton-polaritons [1]. In this platform (cf. Fig. 1a), the GHz phonons with frequencies well beyond 6 GHz are generated by bulk acoustic wave resonators (BAWRs) fabricated on a cleaved facet of the MC chip. We introduce a simple process for the fabrication of the piezoelectric BAWRs on the cleaved sample edge. The MC spacer embedding quantum wells and bound by cleaved lateral facets acts as an embedded waveguide (WG) cavity for the phonons. Phonon propagation was investigated at low temperatures (10K) by detecting multiple acoustic echoes using the BAWR: due to the low acoustic absorption, the generated phonons propagate back-and-forth several times along the 3.5 mm-long cavity, leading to a very high-quality factor ( $Q \sim 10^{\circ}$ ) and lifetimes of tens of  $\mu$ s. The strong acoustic fields and enhanced optomechanical coupling mediated by polaritonic resonances induce a huge modulation of the energy (in the meV range) and strength (over 80%) of the QW photoluminescence (PL), which, in turn, becomes a sensitive local phonon probe. As an example, Fig. 1b displays a photoluminescence map of the narrow (100 µm wide) and very long acoustic transport path. Furthermore, we show the coherent coupling of acoustic modes propagating along the WG and along the substrate and present evidence for nonlinear acoustic absorption due to the high acoustic amplitudes stored in the acoustic cavity. In contrast to the polariton modulation by phonons propagating along the MC growth axis [2], the platform for optomechanics introduced here enables much higher acoustic quality factors and opens the way for phonon-mediated coherent control and interconnection of three-dimensional epitaxial nanostructures (this work was carried out with inancial support from the German DFG).

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#### Figures



**Figure 1:** a. Exciton-polariton microcavity (MC) pumped by a piezoelectric bulk acoustic resonator (BAWR) fabricated on a side facet. The 6.5 GHz phonons generated by the BAWR modulate the photoluminescence (PL) of exciton-polaritons along its transport path. b. PL map of the 100  $\mu$ m wide and several mm long acoustic transport acoustic beam.



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During the past decade, confinement of ultrahigh-frequency acoustic phonons in planar structures and micropillars has been achieved [1][2][3]. However, coherent manipulation of propagation and dynamics of phonons remains challenging [4][5]. Here, we aim at characterizing the transport of 20 GHz phonons in acoustic waveguides based on GaAs/AlAs multilayers. We used a reflection-type pump-probe technique to generate and detect acoustic phonons. In order to evaluate the phonon transport, we performed the pump-probe measurements in two configurations: local, when both pump and probe are overlapped, and remote, by physically separating both beams. In the later configuration we analyse the direct time-of-flight of phonons generated in the pump position reaching the probe. Time-dependent reflectivity variations for local and remote measurements are shown at the top and bottom panels in figure 1(a), respectively. In order to analyse the evolution of acoustic phonons in time, we performed a windowed Fourier transform. For the local experiment (figure 1(b), top panel), the acoustic mode at  $\sim$ 20 GHz has an exponential decay in time, as observed in standard resonators. When the probe beam is separated 8 µm from the pump beam, we observe a delayed signal at ~20 GHz reaching maximum amplitude at around 8 ns (figure 1(b), bottom panel). This delayed signal is the first indication of phonon transport in optophononic waveguides. This result is promising for revealing the fundamental properties of phonon dynamics and manipulating phonon propagation in more complex structures.

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#### Figures



**Figure 1(a):** Time-dependent reflectivity for local measurement (top), where pump and probe beams are focused on the same position, and remote measurement, where the probe is separated 8  $\mu$ m from the pump. (b) Windowed Fourier transform results corresponding to the time traces shown in (a).

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Vibration modes of biological particles such as proteins, viruses and bacteria involve coherent structural vibrations at frequencies in the THz and GHz domains. These vibration modes carry information on its structure and mechanical properties that play a pivotal role in many relevant biological processes. Despite the rapid advances of optical spectroscopy techniques, detection of vibration modes of single bioparticles has remained elusive. Here we harness a particular regime in the physics of mechanical resonator sensing that serves for detecting them. By depositing single bacterium on the surface of ultra-high frequency nano-optomechanical disk resonators, we demonstrate that the vibration modes of the nano-optomechanical disk and bacterium hybridize when their associated frequencies are similar (Figure). A general theoretical framework is developed to describe the different regimes that can be found when an analyte adsorbs on a mechanical resonant sensor. The model recovers the classical inertial mass sensing regime as a limit case of a more general problem. Theory, numerical calculations and experiments show excellent agreement, allowing to retrieve the eigenfrequencies and mechanical loss of the bacterium vibration modes. Our method is applied for analysis of the effect of hydration on the vibrational properties of a single bacterium. This work opens the door for a new class of vibrational spectrometry based on the use of high frequency mechanical resonators with the unique capability to obtain information on single biological entities [1].

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Figure: a. Frequency of the first radial breathing mode of a 320 nm thick disk (blue region) and of the fundamental mode of a quasi-spherical bioparticle adsorbed on a rigid support (red region), as a function of their radii. b. Effect of bacterium adsorption on the first radial breathing mode of a gallium arsenide nano-optomechanical disk (Radius=2.5 μm, Thickness=320 nm). The inset shows a scanning electron microscopy image of the nano-optomechanical disk with an attached Staphylococcus epidermidis cell.



# Coherent Raman scattering microscopy for the study of biological organisms: valuable analytical tool

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#### Abstract

In living organisms, calcium carbonate biomineralization combines complex bio-controlled physical and chemical processes to produce crystalline hierarchical hard tissues (usually calcite or aragonite), typically from an amorphous precursor phase [1]. Understanding the nature of the successive transient amorphous phases potentially involved in the amorphous-to-crystalline transition requires characterization tools, which are able to provide a spatial and spectroscopic analysis of the biomineral structure. In this work, we present a highly sensitive coherent Raman microscopy approach, which allows one to image molecular bond concentrations in post mortem shells and living animals, by exploiting the vibrational signature (phonon) of the different carbonates compounds [2]. To this end, we target the  $v_1$  calcium carbonate vibration mode and produce spatially and spectroscopically resolved images of the shell border of a mollusk shell, the *Pinctada margaritifera* pearl oyster. A novel approach is further presented to efficiently compare the amount of amorphous carbonate with respect to its crystalline counterpart. Finally, the whole microscopy method is used to image *in vivo* the shell border and demonstrate the feasibility and the reproducibility of the technique. We use the same technique to characterize *in-situ* the matrix composition of some bacterial biofilms growing in the presence or absence of metals. These findings open chemical imaging perspectives for the study of biogenic and bio-inspired crystals.

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**Figure** (Hamadou Dicko and al., Structural Biology, 2022, 214 (4), pp.107909): a) Experimental SRS spectrum from the region shown in b) (mature prism). The data has been fitted with a double-peak Gaussian profile (dashed line) corresponding to the calcite (Cal) contribution (blue line) and the amorphe calcium carbonate (ACC) contribution (red line). b) The C is calculated from the two image intensities taken at the Cal (1086 cm<sup>-1</sup>) and ACC (1074 cm<sup>-1</sup>) values depicted in a. c) The C<sub>q</sub> is calculated using the amplitudes obtained for Cal and ACC from the fit procedure. d) Ratio between the two different contrast maps, showing less than 30% of variation.



# Wave Localization in Disordered Phonon Systems and the Emergence of Basic Biological Functions

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Phonon localization occurs when lattice vibrations are confined by scattering from structural irregularities in crystal lattices. Despite extensive research on Anderson localization of electrons in disordered electronic solids, the localization of phonons remains poorly understood. In this study, we present a novel phonon localization theory [1] based on the concept of a localization landscape, which tracks the confining domains of phonon modes with a particular focus on its application to proteins. Proteins, among the most structurally disordered systems found in nature and further subjected to evolution, serve as relevant targets for investigating the dynamical properties of living matter. We apply our theory to several classes of proteins [2][3][4], including enzymes, membrane receptors, and viral envelopes, demonstrating that localized phonon modes are correlated with the emergence of basic biological functions, such as catalysis, regulation, and intramolecular communication. Our work highlights the potential of our approach in understanding emerging properties of disordered systems, including biological media, and suggests potential applications in the design of biomimetic materials.

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# Correlative Imaging of Motoneuronal Cell Elasticity by Pump and Probe Spectroscopy

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Abstract:

Because of their role of information transmitter between the spinal cord and the muscle fibers, motor neurons are subject to physical stimulation and mechanical properties modifications. We report on motoneuron elasticity investigated by time resolved pump and probe spectroscopy. A dual picosecond geometry probing simultaneously the acoustic impedance mismatch at the cell/Titanium transducer interface and acoustic wave propagation inside the motoneuron is presented. Such noncontact/non-destructive microscopy, correlated to standard atomic force microscopy or fluorescent labels approach have been carried out on a single cell to address some physical properties as bulk modulus of elasticity, dynamical longitudinal viscosity or adhesion.

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Figure 1: - Image scale 91µm. a) Mapping of the Brillouin life time in the cell. b) Fluorescence mapping of the same cell. The blue zone marked the nucleus material and the green color pointed out the cytoskeleton rich in Actin. c) Mapping of the Brillouin frequency cell response. The values are fixed at zero outside the motoneuron where no Brillouin oscillation are detected.



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#### Abstract

Nanomechanical resonators are freely suspended, vibrating structures with nanoscale dimensions. They show great promise as versatile linking elements in hybrid nanosystems, as sensors or signal transducers both in the classical and in the quantum realm, generating an increasing amount of attention both for fundamental explorations and for practical applications. Here I will focus on doubly-clamped pre-stressed nanostring resonators. As a result of dissipation dilution, they exhibit remarkably large room temperature quality (Q) factors. Dielectric transduction via electically induced gradient fields represents an efficient way not only for actuation and displacment detection, but also for eigenfrequency control over a large frequency range [1]. Dielectrically controlled nanostrings are thus an ideal testbed to explore a variety of dynamical phenomena. I will review recent progress in controlling the coherent as well as the nonlinear dynamics of high Q nanostring resonators. In particular, I will discuss the interplay between the two orthogonal fundamental flexural modes of the string vibrating in- and out-of-plane with respect to the sample surface. These two modes are strongly coupled and reveal a pronounced avoided crossing [2], which can be described as a classical two-mode system mimicking the coherent dynamics of a quantum two-level system [3]. I will further highlight the nonlinear dynamics of the resonantly driven nanostring, which gives rise to thermomechanical squeezing [4], and has recently been shown to lead to the generation of a novel type of frequency comb [5].

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## Exploring the high-frequency mechanical resonances of a quantum dot-microwire hybrid system

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Hybrid mechanical systems are composite devices, which couple a two-level quantum system (TLS) to a mechanical resonator. Besides applications in sensing or in quantum information technologies, these devices have also been proposed to explore the quantum/classical boundaries, by coherently mapping non-classical states of the TLS onto the "macroscopic" resonator. Together with colleagues in Grenoble, we developed a hybrid system based on a semiconductor InAs quantum dot (QD) that is strain-coupled to the fundamental flexural vibration of a GaAs microwire [1]. Although this system has enabled many pioneering contributions to the field [2,3], further developments call for a massive increase of the resonator frequency (from the sub-MHz up to the GHz range). The higher-order mechanical resonances of the microwire are particularly appealing in this context, but investigations have been hindered by the lack of suitable excitation technique. Here, we report a mechanical spectroscopy of these high-frequency modes [4], based on a new actuation

technique [5]. On-chip electrodes allow applying an oscillating electrostatic force on the microwire (Fig. 1(a)). Vibrational resonances are then revealed by a blurring of the QD emission lines, which directly reflects the time-averaged oscillation of their emission wavelength (Fig. 1(b)). Leveraging the broad operation bandwidth of the technique, we detected so far resonances up to 300MHz, an increase by three orders of magnitude compared to the fundamental mode. Surprisingly, large mechanical quality factors (a few 10<sup>3</sup>) are preserved at high frequencies.

Since the radiative limit of the QD emission linewidth is on the order of 160MHz, the resolved-sideband regime is within reach in this system. These results thus open appealing prospects for the – possibly coherent – optical manipulation of the mechanical state and for the realization of photon-phonon interfaces.

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#### Figures



**Figure 1:** (a) Scanning electron microscope image of the device (tilted view). (b) Colour plot of the QDs photoluminescence intensity (blue=low, yellow=high) as a function of the photon energy and of the driving frequency (measured at T=4K). The flexural resonance, here close to 59.5 MHz, is indicated by a blurring of the QD emission lines.

## Flexural phononic crystals in the structures with van der Waals nanolayers

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Two-dimensional van der Waals crystals (2D-vdW) consist of chemically bonded atomic layers held together by weak vdW forces. Coherent THz and sub-THz phonons, which carry dynamical strain, could become an instrument to control classical and quantum phenomena in the unexplored picosecond temporal and nanometer spatial regimes.

The present contribution describes a series of experiments [1] where coherent phonon modes are studied in MoS<sub>2</sub> vdW nanolayers deposited on nanogratings with a period 50-250 nm fabricated from various materials (see the scheme in Fig. 1a). We use the picosecond ultrasonic technique to generate and detect coherent phonons. Figure 1b shows the phonon modes studied in our experiments: flexural A0 Lamb mode; LA-like S0 Lamb mode; and out-of-plane breathing mode. The most interesting results are obtained for flexural phonon modes with frequencies up to 10 GHz. The dependences of phonon frequency on the period of nanograting, i.e. dispersion curves, and on the thickness of vdW nanolayers lead us to conclusions about the hybrid nature of flexural phonon modes, which arises from the periodic modulation of the elastic coupling of the vdW layer at the grooves and ridges of the nanograting as shown in the inset in Fig. 1c. The scheme in the inset in Fig. 1c illustrates the model for theoretical analysis of hybrid (H) phonons and the main panel of Fig. 1c shows the dispersion curves for hybrid modes, where anticrossing between phonon modes are clearly seen. Figure 1d shows the example of the dependence for phonon frequency on the nanolayer thickness. The dotted lines in Fig. 1d correspond to the fundamental and second harmonic flexural (A0) modes for free standing nanolayer while the solid lines are calculated dependences for hybrid phonon modes. It is seen that the experimental points agree well with the concept of hybrid phonons in such nanostructures. Figure 1e shows schematically the layer motion for hybrid phonon modes. The experimental results and theoretical simulations create a new type of a tailorable 2D periodic phononic nanoobject, a *flexural phononic crystal*, offering exciting prospects for the ultrafast manipulation of states in 2D materials.

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**Figure 1: (a)** MoS<sub>2</sub> on a nanograting. **(b)** Studied phonon modes. **(c)** Calculated dispersion curves for first three hybrid phonon modes. Horizontal dotted line corresponds to the frequency of a nanolayer elastically coupled to a plane surface with stiffness  $\eta_z$ . Anticrossing between the pairs H1/H2 and H2/H3 is the results of mode hybridization. **(d)** The dependence of frequencies on the vdW layer thickness measured in FeGa grating with a period *d*=200 nm (symbols) and corresponding theoretical dependences H1 and H2 (lines). The dotted lines are the calculated thickness dependences for A0 modes in a free standing layer. **(e)** The layer motion for H1 and H2 hybrid modes displayed for three layer thicknesses. It is seen how the modes localized above the groove start to penetrate on the grating teeth with the increase of layer thickness *a*.



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#### Abstract:

Controlling spins on very short time scales may enable novel non-equilibrium magnetic physics as well as ultrafast spintronics for energy-efficient information processing. Strongly driven phonons directly influence the interatomic distances, orbital symmetry, and exchange interactions that are closely related to magnetism in materials. Chiral phonons, where atoms rotation unidirectionally around the equilibrium position inside crystalline lattice, break time reversal symmetry and are expected to directly couple with magnetization. We excited chiral phonons using circular-polarized terahertz pulses and observed a prominent transient magneto-optic Kerr effect in rare-earth trihalides. The effective magnetic field needed to polarize the paramagnetic spins is on the order of 1 T under very moderate pump fluence of 0.2 mJ/cm2. The temperature dependence of the spin dynamics indicates the magnetic field indeed is generated by the phonons, as opposed to a pure electromagnetic inverse Faraday effect. Our result may open a new route to investigate spin-phonon interaction in ultrafast magnetism.





**Figure 1:** (a) Transient magnetization generated by circularly polarized terahertz pump-induced chiral phonons and probed by Faraday rotation. The magnetization direction is controlled by the helicity of the pump light. (b) Effective magnetic field of chiral phonons as a function of pump fluence calculated from the Kerr ellipticity and magneto-optic coefficient. Inset: the linear component of the electric field from the narrowband pump pulses, centered at phonon resonance of 10.8 THz.
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#### Abstract

Topological insulators have attracted much interest and time resolved spectroscopy technics have proven to be very useful to investigate different properties of these materials: time- and angle-resolved photoemission spectroscopy (trARPES) to study Dirac surface states [1], optical pump-THz probe spectroscopy [2] and time-resolved X-ray diffraction [3] to study dynamics of photoexcited states, time-resolved reflectivity measurements to study dynamics of photoexcited coherent optical phonons [4]. The same technique used to investigate acoustics phonons in Bi<sub>2</sub>Se<sub>3</sub> revealed a strong anomaly in the temperature dependence of sound velocity in a bulk sample [5] and intriguing features in very thin films (10-40nm) [6]. In order to clarify these points, we performed picosecond acoustic experiments on epitaxial Bi<sub>2</sub>Se<sub>3</sub> films that were of sufficient thickness to obtain well separated acoustic echoes. The measurement of the acoustic echoes time of flight allowed an accurate determination of the speed of sound over a wide temperature range which did not reveal any anomaly in contradiction with experiments performed on bulk samples [6]. Furthermore, the study of the acoustic echoes revealed very interesting features on the phonon generation process and the electronic diffusivity.

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## Picosecond acoustic measurements at extreme pressure and temperature conditions

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Picosecond acoustics is a time-resolved optical pump-probe technique that allows studying the propagation of acoustic echoes in a large variety of samples at different pressure and temperature conditions. This laboratory-technique allows determining the complete elastic properties of crystals as well as phase diagrams, melting curves and longitudinal velocities for polycrystalline samples.

Combined with diamond anvil cell, picosecond acoustics measurements are now routinely performed at ambient temperature at pressures exceeding the Mbar [1-3] and, coupled with resistive heated diamond anvil, several measurements up to 600 K and 10 GPa have been performed to determine sound velocity, melting curves and thermodynamic properties of various solids and liquids [4-7]. Extension to pressures above the Mbar and temperatures of several thousands of K by coupling a laser heating system to picosecond acoustics [8] not only has a fundamental interest in condensed matter physics and materials science, but also in planetary science. This indeed offers the possibility of comparing the measurements of sound velocity and density of planetary materials with the values obtained by reference seismic models and constraints the temperature profile of planetary interiors by direct measurements of melting curves.

Here I will illustrate the basic working principle of the experimental setup and will highlight the importance of these experiments for Earth and planetary science by providing examples of studies including measurements on iron and iron alloys, metallic liquids and MgO. Finally, I will present first test measurements showing the great potentiality of picosecond acoustics coupled to laser heating.

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### Magnon – phonon coupling in magnetostrictive nanostructures

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Coupling excitations in different systems allows scientists and engineers to harness and exploit the beneficial properties of each system and can lead to new ways to investigate fundamental physics. Magnon and phonon modes can be engineered to exist in the low GHz frequency range in nanostructures. Magnons can be tuned by magnetic field and methods exist and are being developed to couple magnons to microwave and optical photons. Phonons can have longer lifetimes than magnons and can be propagated over larger distances, and methods already exist to manipulate and utilise surface acoustic waves in communications technologies. It would be desirable to develop methods to couple magnons and phonons in the same structure. Recent works have reported methods to drive magnons with phonons (and vice versa) and to achieve the strong coupling limit in which the magnon and phonon modes hybridise [1]. Key challenges include developing materials possessing strong magnetostriction, capable of supporting magnon and phonon modes with long lifetimes, and engineering structures in which the coupling between the magnon and phonon modes is optimised. We will discuss the development of a suitable material system – epitaxial thin layers of galfenol (Fe<sub>81</sub>Ga<sub>19</sub>) which possesses favourable properties including strong magnetostriction, large moment and magnetocrystalline anisotropy, and magnon modes with long lifetimes [2]. Experimental progress in developing device concepts for achieving magnetisation precession driven by coherent phonons [3], and hybridisation of magnon and phonon modes [4] will then be described.

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#### Figures



**Figure 1:** Magnon modes in a nanograting fabricated from galfenol. Strong magnon phonon coupling is evidenced by the anticrossing at 13 GHz. Taken from [4].



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In recent years, the study of surface phonon polariton waves SPhP has gained a lot of interest due to their diverse applications, and their energy-transfer abilities in low-dimensional systems, in addition to being low-loss. These SPhP waves result from the coupling of light to the mid-infrared electric field associated with optical phonon modes. The generation and control of SPhP's are crucial for designing novel applications whether in microscopy, photonics, thermal emission or in biosensing.

In this contribution, we experimentally investigate the generation and the detection of SPhP's in different polytypes of Silicon Carbide SiC samples; cubic SiC and 4H-SiC. The study is performed using a non-destructive near-field microscopy technique, the Photo-induced Force Microscopy PiFM, where the photo-induced force measurements are recorded at different laser powers using a tunable mid-infrared quantum cascade laser (QCL), with a spectral range of 770-1860 cm<sup>-1</sup>. Our measurements demonstrate that the illumination of the tip-sample junction in PiFM, with a mid-infrared light source, does not only lead to local heating of the sample, but the latter also excites localized surface modes, namely the SPhP's. The obtained force spectra also provide a strong experimental evidence of the influence of the applied laser powers on the varying intensity of the excited surface mode, which is an aspect that opens the possibility of extracting the local temperature of the sample and eventually determining its thermal conductivity.

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Figure 1: Photo-induced Force (PiF) spectra (left-side) and normalized PiF (right side) measured on 3C-SiC showing how the localized surface mode is varying with the different IR laser powers, from 1% till 7%.



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High-frequency coherent phonons in the GHz range have a potential for application in quantum technologies due to their nanometer wavelengths being comparable with the size of quantum nanodevices. Therefore, increasing strength of photon-photon coupling became of interest to the community [1,2]. In this work, we show that it is possible to reach quantum sensitivity of the phonon detection with a standard pump-probe set-up by exploiting giant photo-elasticity of exciton-polariton resonance in a short period GaAs/AlAs superlattice (SL) [3].

For excitation and detection of phonon wave packet, we use a standard pump-probe experimental set-up with a mechanical delay line and a lock-in amplifier. A coherent acoustic phonon wave packet in a form of the strain pulse is generated by the absorption of the pump laser pulse by a metal transducer at the back of the sample. Strain pulse propagates into GaAs substrate and reaches the GaAs/AlAs SL at the front, where it gets detected (see Fig.1). By sweeping the central wavelength of the probe in the vicinity of the polariton resonance of the SL we obtained signals for several detuning values of the probe photon energy. The amplitude of the measured signal depends strongly on the photon energy of the probe.

Our experiments reveal giant photo-elasticity of polaritons and extremely high sensitivity to propagating coherent phonons. The strong dispersion of the dielectric permittivity in the vicinity of the polariton resonance results in a strong ultrafast response of the optical properties to dynamical strain which accompanies the coherent phonons. This discovery opens new possibilities of ultrasensitive measurements of extremely low-density phonon fluxes.

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Figures



Figure 1: Excitation and detection of coherent phonon wave packet by means of pump-probe experiment.

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Dynamical tuning of the thermal conductivity in crystals,  $\kappa$ , is critical for developing novel phononic devices able to perform logic operations with phonons, as well as for solid-state refrigeration, energy harvesting, and thermoelectrics. Such a desired  $\kappa$  control can be achieved in functional materials that experience large structural and phonon variations as a result of field-induced phase transformations.

Current strategies to design materials with tailored thermal properties typically rely on the creation of structural inhomogeneities that scatter phonons and reduce the thermal conductivity (e.g., lattice defects, grain boundaries and nanostructure surfaces) and the engineering of periodic heterostructures (superlattices) in which wave interference induces the appearance of phonon frequency gaps. These approaches, however, yield changes in the thermal properties of materials that are static and irreversible. The design of *phononic* devices [1], capable of performing logic operations with phonons similarly to what their electronic counterparts do with charge carriers, on the other hand, relies on the possibility of dynamically tuning the thermal conductivity of materials,  $\kappa$ .

A promising dynamical  $\kappa$  tuning approach consists in using electric or magnetic fields to manipulate the lattice of polar and magnetic materials, respectively, ideally exploiting the effects of field-induced phase transitions on the thermal properties. The ensuing *electrophononic* [2-5] and *magnetophononic* [6-7] effects can provide fast and dynamical manipulation of the heat carriers, thus yielding à la carte thermal properties for on-demand applications.

Nevertheless, for these strategies to be practical, the operation conditions need to be close to zero-bias transition points, since otherwise the required driving fields may grow unfeasibly large and the materials performances be seriously compromised owing to the presence of leakage/eddy currents and dielectric/magnetic losses. On the other hand, the possibility of manipulating  $\kappa$  with light thus far has received very little attention. Light-driven control of the thermal conductivity could bypass some of the issues posed by the schemes described above (e.g., application of large driving fields) as well as simplify the design of logic devices (i.e., lack of electrical contacts) [8].

In this talk I will discuss our predictions, based on first-principles calculations, of each of these three effects: *electro-, magneto-,* and *photopononic* couplings as effective way to dynamically manipulate the thermal conductivity of materials with external fields.

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Time-resolved pump-probe optical spectroscopy on metal nano-objects constitutes a powerful tool to investigate the specific modalities of nanoscale heat transfer, which plays a crucial role in many applicative fields such as electronics, thermoelectricity and nanomedicine [1]. This approach relies on the sudden excitation of nano-objects and on the time-domain monitoring of their subsequent cooling dynamics using femtosecond pump and probe pulses. It enables non-contact studies of energy exchange mechanisms between the nano-objects and their environment, whose kinetics generally depends on both the thermal conductance of the interface between the nano-objects and their environment and the thermal properties of the latter. Such experiments can now be performed at the single-particle level, which allows to test the reproducibility of the cooling dynamics from one nano-object to another and to quantitatively measure its induced transient optical response [2,3].

Single-particle time-resolved optical spectroscopy was applied here to the study of the cooling dynamics of gold nanodisks synthesized using colloidal chemistry and deposited on various dielectric substrates differing by their composition, thermal properties and thickness and including nanometric membranes. The cooling dynamics of nanodisks deposited on a thick sapphire substrate were found to be mostly ruled by interfacial heat transfer, with little modification of the gold-sapphire thermal interface conductance as compared to recent experiments performed on gold nanodisks produced using electron beam lithography [2]. In contrast, the cooling of nanodisks supported by thin suspended membranes was seen to be strongly influenced by heat conduction in the membrane and much slower, the time separating two successive pump pulses in our experiments being insufficient for its completion. The detailed analysis of the cooling dynamics of a membrane-supported nanodisk was performed using finite-element simulations in the frequency domain. This modeling approach provided a better understanding of the non-complete nanodisk cooling before each pump pulse in the permanent regime, and allowed us to quantitatively estimate the thermal conductances of nanodisk/membrane interfaces and the thermal conductivities of the used thin membranes.

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Optomechanical nanobeams are typically made on silicon-on-insulator (SOI) substrates, i.e., using the single crystalline device layer for the devices. Surprisingly, nanocrystalline silicon can provide higher Q factors and, e.g., higher frequency self-pulsing than single crystalline silicon [1-4]. Furthermore, the thickness and strain of nanocrystalline silicon layers can be easily controlled, thus offering a flexible platform for optoelectromechanics. In this talk we will present the specific properties and dynamics arising from the nanocrystalline structure and the effects on phonons.

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#### Figures





**Figure 1:** (Left) Bright field TEM image of nanocrystalline silicon. (Right) Schematics of an optoelectromechanical nanobeam structure (courtesy of G. Madiot).



## Impact of spin waves dispersion on surface acoustic waves and ferromagnetic resonance in iron thin films

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Surface Acoustc Waves (SAW) have been proposed to dynamically control Spin Waves (SW) propagation and even to generate SWs, in order to implement reconfigurable and energy efficient magnonic devices. Indeed, SAW technology is mature and widely used in today's sensors, filters and microwave circuitry, notwithstanding the lack of tunability of SAW transducers. For this reason, a multitude of them are currently integrated in modern devices (e.g. mobile phones).

Recently, the so-called SAW induced Ferromagnetic Resonance (SAW-FMR) has been observed by Weiler et al. [1] in Ni thin films by exciting SAWs in the GHz and sub-GHz regime in piezoelectric media. Moreover, SAW-FMR permits to induce spin-pumping in CoPt bilayers [2,3]. The SAW-FMR interaction is often described by taking into account only the uniform FMR mode. However, this approximation is rather crude, and it misses out the wealth of modes that can be excited in a ferromagnetic (FM) material.

Here, we study SAW propagation in a Fe thin film epitaxially grown on a piezoelectric GaAs substrate. The SAW velocity and absorption show a dependence on the external magnetic field in amplitude and direction after coupling with the FMR. [4,5] The observed angle dependence can be described only by taking into account the spin wave dispersion. To interpret this dependence on the magnetic field angle, a phenomenological approach to the relative change in SAW velocity is implemented with the calculated spin wave dispersion curves.

Our study permits to envisage SAW-based magnonic devices where a single IDT provides the energy needed to activate magnetization dynamics and SW propagation in hundreds if not thousands of magnonic waveguides, in a Joule heat free manner.

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#### Figures



**Figure 1:** (left) Sketch of the sample: Fe mesa (54 nm thick) on GaAs (001).  $k_{SAW}$  is parallel to [110],  $B_{ext}$  is the in-plane applied magnetic field. (right) Relative variation of the SAW velocity V measured versus decreasing amplitude of the external field  $B_{ext}$  at  $v_{SAW}$  = 119 MHz and 833 MHz. The magnetic field  $B_{ext}$  is applied in the film plane, parallel (a) to the [110] direction //  $k_{SAW}$ , (b) to the [100] direction, or (c) to the [110] direction.

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The world-wide need for ever-increasing computing performances calls for new technologies for data storage and handling. Most prominently, in new-generation non-volatile magnetic memories the write operation through an external applied magnetic field is no longer possible, due to Moore-ruled scaling down of the magnetic bits. Instead, acoustic control of magnetism, often called magneto-acoustic, appears as an appealing route for low-energy-consumption operation, exploiting the magneto-elastic coupling, driven by inverse magnetostriction at micron-size wavelength [1]. Besides the technological relevance, much attention has been recently paid to fundamental investigation on interplay of spin degrees of freedom and coherent phonon excitation, without the intermediation of electronic excitation [2].

Here we propose an all optical approach for generating surface acoustic waves (SAW) able to seed coherent spin waves via magneto-elastic coupling [3,4]. We used a four-wave mixing approach, called transient grating, in which the elastic modulation, generated by the interference of two laser pump pulses, couples with magnetic degrees of freedom, generating a spin wave. Tuning an external applied magnetic field on magnetic thin films, we can reach the condition of acoustically-driven ferromagnetic resonance.

The combination of a transient grating scheme with time-resolved Faraday polarimetry on a polycrystalline Ni thin film allows the extraction of the effective magnetization value and the Gilbert damping. The results are in full agreement with measurements on the very same samples from standard FMR. Higher-order effects due to parametric modulation of the magnetization dynamics, such as down-conversion, up-conversion, and frequency mixing, are also reported.

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#### Abstract

A solid grasp of thermal phonon transport and phonon-magnon interactions is essential to designing materials and devices for thermal energy conversion, microelectronics, and magnonics. In this talk, I will first share my group's work on inelastic X-ray scattering measurements to probe phonon dynamics of in 3D and 2D hybrid organic-inorganic perovskites using inelastic X-ray scattering, where we gained useful insights into their ultralow thermal conductivity and remarkably weak anisotropy in 2D hybrid. I will then cover our development of the anharmonic atomistic Green's function method that overcame a decade-long challenge for interfacial thermal modeling, which considers inelastic phonon scattering at interfaces for practical temperature ranges. Last but not least, I will introduce our theoretical modeling of topological phonon-magnon hybrid excitations in a 2D ferromagnet.

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#### Figures



Figure 1: Schematic of the inelastic X-ray scattering on hybrid perovskites in the transmission geometry

## Role of dimensionality, size, and transport-direction in governing the drag Seebeck coefficient of doped silicon nanostructures: A first-principles study

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Under the presence of a temperature gradient, the electric and heat currents experience a mutual drag via the interaction between charge carriers and phonons [1]. The effect of phonon-drag on the charge carriers leads to significant enhancement of the Seebeck coefficient of some materials and therefore presents a potential interest for thermoelectric devices [1]. However, the magnitude of phonon-drag, which in turn depends on the strength of carrier-phonon coupling and on the value of out-of-equilibrium phonon populations, is expected to diminish with nanostructuring. To quantify the relative contribution of the phonon-drag Seebeck coefficient on the nanoscale, a detailed understanding of the dependence of the drag Seebeck coefficient on the dimensionality and size of nanostructures is necessary, and this can only be achieved through theory because of the impossibility to separately measure the diffusion and drag contributions to the Seebeck coefficient. In this talk, I will present our recent study of the influence of dimensionality, size reduction, and heat-transport direction in governing the drag Seebeck coefficient of doped silicon nanostructures [2]. Our theoretical approach is based on the solution of the partially coupled Boltzmann transport equations for the charge carriers and phonons, as well as on the fully ab initio description of carrier-phonon [3,4] and phonon-phonon interactions [5]. We have considered various nanostructure dimensionalities (Fig.1, left panel): 3D (grains), 2D (thin films) and 1D (wires). To the best of our knowledge, this is the first study that accounts for the anisotropy of the boundary scattering in different kinds of nanostructures as well as for the spin-orbit coupling for holes. Our results have been found in excellent agreement with the recent experimental findings of Refs. [6a,b], concerning the impact of phononboundary scattering on the Seebeck coefficient of doped silicon nanowires (Fig. 1, right panel).

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Figures



**Figure 1:** (Left panel): Schematic diagram shows the in-plane and out-of-plane direction of heat and current flow in the examined nanostructures. Right panel: Variation of the total Seebeck coefficient as a function of the silicon nanostructure size ( $L^{Cas}$ ) at 300 K. Experimental data are from Refs. [6a,b].

## Effect of phonon anharmonicity on MgO lattice dynamics at extreme conditions: Impact on thermal conductivity

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We present a study of the phonon dynamics of MgO (energies and lifetimes) measured as a function of pressure and temperature by means of infrared spectroscopy and inelastic x-ray scattering and modelled mainly by *ab-initio* calculations based on Density Functional Perturbation Theory (DFPT). Our results show the origin of the phonon anharmonicity in MgO [1] and the limits of the quasi-harmonic model over a large volume range (~20% variation of V) obtained on the one hand by hydrostatic compression (low V) and on the other hand by thermal expansion (high V) [2]. We compare the experimental data with *ab-initio* quasi-harmonic calculations to estimate energies and third-order anharmonic calculations to estimate linewidths. The results obtained in the case of energies show the increasing disagreement between calculations and experiments as the temperature increases. This suggests the need for an energy treatment including three- and four-phonon scattering and the predominance of the latter in the overall balance. In the case of widths, instead, the simulations agree with the experimental observations and help to explain the non-monotonic behavior of the optical transverse mode lifetime at high pressure. This validates the perturbative model used to treat the widths, which neglects the contributions due to terms above third order. We then extend the comparison between calculations and measurements by pushing the simulations towards thermodynamic conditions typical of the Earth's deep mantle. Here, the anharmonic treatment of energies is introduced with the addition of three- and four-phonon scattering. The former is treated by the DFPT and the latter by the Stochastic Self Consistent Harmonic Approximation (SSCHA). This allows to follow the joint effects of temperature and pressure on the collective dynamics of MgO and the evolution of the different anharmonic terms. We provide, in such a way, a quantitative balance of the anharmonic contributions, both in energies and in widths in the Earth's lower mantle. The results show how the cancellation or dominance of one perturbative order over the other depends on the absolute values of pressure and temperature, preventing the generalization of the results obtained for the Earth to other planetary systems where temperature and pressure vary with depth in a different way than expected for our planet. Finally, we calculate the thermal conductivity and compare it with the available experimental measurements. This gives an order of magnitude of agreement between theory and experiment and permits to estimate the weight of intrinsic and extrinsic contributions. In particular, we see how the addition of a minimal number of impurities, treated as oxygen vacancies, allows thermal conductivities to be found in excellent agreement with the measured values. Considering the combined effects of temperature and pressure, despite the progressive hardening of the energies due to the predominance of pressure over temperature, the continuous and progressive broadening of phonon widths contributes to arrest the increase in thermal conductivity, whose value decreases near the core-mantle boundary.

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# Sub-wavelength confinement of surface acoustic waves induced by collective coupling of phononic resonators

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Micro- and nanomechanical cavities form an essential ingredient of modern hybrid opto-electro-mechanical systems. The enhancement of strain-induced interactions deriving from confined vibrations has stimulated sustained interest and found numerous applications, ranging from the investigation of fundamental quantum physics phenomena to radiofrequency signal processing or sensing [1].

In this work, we propose to achieve sub-wavelength confinement of elastic field by exploiting coupling between multiple micron-scale pillar resonators and surface acoustic waves. We investigate a sample made of six pillar cylinders fabricated by ion-beam-induced deposition on a lithium niobate substrate. The pillars form a hexagonal arrangement that define a phononic cavity, as illustrated in Figure 1a. Interdigitated transducers operating around 70 MHz are used to generate a Rayleigh wave that excites the first flexural mode of the pillar resonators. Mapping of the displacement field by laser scanning interferometry reveal a strong confinement of the elastic field within the hexagonal resonator ring. Mode volumes below 10  $\mu$ m, much smaller than the 50- $\mu$ m free-surface wavelength, are observed and the measured elastic displacement is shown to be 6 times higher than the one of the impinging Rayleigh wave. We show that this confinement directly originates from a synchronized, collective state of the resonators, where all six pillars vibrate in-phase and towards the ring's geometric center. A numerical model implemented using the finite element method is in excellent agreement with the experimental observations and further suggests that both the quality factor of the confined mode and the confined field amplitude are linked to the resonator-to-resonator coupling strength. The proposed device is readily scalable and opens a path towards sub-micron scale confinement of GHz elastic waves for enhanced strain-induced multiphysical interactions.

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**Figure 1:** (a) Scanning electron microscope image of a phononic resonator ring grown by ion-beam-induced deposition on a lithium niobate substrate. The resonators are 3.6  $\mu$ m-high and 3  $\mu$ m wide; the gap spacing is equal to 1.5  $\mu$ m. (b) Frequency response of the substrate surface measured at the center of the cavity. A resonance appears at 67.28 MHz. (c) Corresponding numerical simulations. (d-e) Experimental and simulated elastic field maps taken at resonance frequency on the substrate surface, within the cavity. (f-g) Experimental and simulated phase at showing the pillar vibration at resonance.

### A traveling-wave broadband SAW transducer based on superconducting nanowires

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Surface acoustic wave (SAW) devices are crucial tools in RF signal processing and have promising applications in quantum information [1]. To further enable these applications, we developed a novel traveling-wave SAW transducer (TWST) based on superconducting nanowires. Using a prototype device on a GaAs substrate, we were able to convert broadband electrical signals to and from SAWs, and we have characterized the transmission and reception characteristics over a decade of bandwidth (40-600 MHz). The TWST utilizes superconducting-nanowires as slow-wave transmission lines to create a phase-velocity matching condition between the electromagnetic and acoustic modes. The resulting transducer has a number of desirable properties for classical and quantum transduction, including (1) broadband transmission and reception, (2) near-zero electrical dissipation, (3) intrinsic directionality, and (4) tunable coupling either through an applied B-field, temperature, or an injected current.

In a typical traveling-wave configuration, transduction is usually accomplished by matching the phase velocities of two systems, for instance by placing an electrical transmission line on top of a photonic waveguide and ensuring that velocities of the optical and electrical modes match. However, this matching is extremely difficult in the case of electrical-to-acoustic transduction, as the phase velocities differ by a factor of 10<sup>5</sup>. One can overcome this mismatch by meandering the wires perpendicularly so that the electrical phase velocity along the SAW axis is reduced, but with a typical conductor this would require extreme dimensions e.g. 150-nm-wide wires meandered 15 mm. However, superconducting nanowires can have propagation velocities on the order of 0.01c [2], reducing the phase-velocity mismatch to approximately 10<sup>3</sup>. This enables the creation of devices with practical dimensions, e.g. 150-nm-wide wires meandered 150 um. Moreover, these superconducting nanowire transmission lines have near-zero loss at the SAW frequencies of relevance, and so are an ideal transduction platform for quantum transduction processes.

In this presentation, we discuss the design, fabrication, and testing of an initial proof-of-concept TWST. We report the frequency response as well as time-domain transmission and reflection measurements, compare results with predicted performance, and discuss potential improvements for the next generation of devices.

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**Figure 1.** (a) Schematic diagram of traveling-wave electro-acoustic transduction process. The transducers are meandered transmission lines, and sine wave voltages are input to the left terminal of the input transducer. The SAW waves travel across a short delay gap and are transduced back into the electrical domain at the receiving transducer, where they are emitted from the right terminal. (b) Transmission amplitude response versus carrier frequency for a 10-ns-wide gaussian pulse. (c) Time delay versus carrier frequency, red line shows the expected total acoustic delay of 174.5 ns.



## GHz Surface-Acoustic-Wave Resonators Probed by Atomic-Force Microscopy on AlN Layers Grown on 6H-SiC(0001) by Molecular Beam Epitaxy

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High-frequency acoustic wave devices are the key to future telecommunication technologies. They also play important roles in other applications such as gas sensing, biosensing and microfluidics. In this context, AIN/SiC heterostructures are an attractive material combination for several reasons. To begin with, both materials support acoustic waves of high frequencies as a result of their high sound velocities [1-2]. Secondly, they exhibit high thermal conductivities as well as thermal stability, and can operate at high power levels, high temperatures, and in other harsh environments [3]. Moreover, AIN is highly piezoelectric and inherently insulating, facilitating the efficient generation of surface acoustic waves (SAWs). Finally, AIN has a comparatively small lattice mismatch to SiC, allowing the synthesis of single-crystal AIN films by molecular beam epitaxy (MBE).

In this work, we demonstrate the effective operation of SAW resonators in the range of 3–7 GHz on AlN layers grown by MBE on 6H-SiC(0001) substrates. The high-quality MBE layers enable us to measure the SAW propagation loss at different frequencies. In particular, we form circular SAW cavities using curved interdigital transducers and use atomic-force microscopy (AFM) to map the SAW profile in the cavity. The lock-in AFM technique enables fast imaging of SAWs with a spatial resolution down to a fraction of a nanometer [4], as well as simultaneous recording of the height profile. Here, the atomic-force topographs directly reveal a Gaussian-like beam shape, with strongly enhanced intensity as well as a beam waist down to the diffraction limit at the center of the cavity, a much desired condition for coupling the SAW to quantum nanostructures such as defect centers for high-frequency single-photon emissions.

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#### Figures



Figure 1: Surface-acoustic-wave profile inside a cavity, mapped by atomic-force microscopy.



### Phonon engineering in Heterostructure Superlattice Nanowires

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#### Abstract

Phonons are the fundamental quasi-particles responsible for heat and sound transport in materials. The ability to manipulate and control phonons lie at the heart of heat management. [1] However, to control heat, one must engineer materials at the nanoscale. Advancements in nanofabrication techniques allow us to engineer new material systems with tunable phononic spectrum.

Superlattices (SL) are compositionally modulated periodic heterostructures, which can be used to explore novel photonic and phononic properties. [2] In particular, SL structures with nanoscale periodicities have been used to show the crossover from coherent to incoherent phonon transport. [3] One of the main difficulties in growing SL structures of different materials is the lattice mismatch between the constituent materials. Nanowires (NWs) are quasi one-dimensional structures which allow the growth of axial and radial heterostructures otherwise difficult in planar structures. [4] Hence, SL NWs are an ideal system to study phonon engineering.

In this work, we studied GaAs/GaP SL NWs with different periodicities obtained by Au-assisted chemical beam epitaxy on GaAs (111) B substrates. [5] Using inelastic light scattering techniques such as Raman spectroscopy and Brillouin interferometry, along with ab initio calculations, we show that the phononic properties of GaAs/GaP SL NWs can be tuned by controlling the SL periodicity. This work shows that nanowires can serve as a template for combining different materials at the nanoscale for designing new materials systems with tunable phononic spectra.

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#### Abstract (Calibri 11)

Volume regulation plays an essential role in maintaining important tissue functions, such as growth or regeneration. The modulation of the membrane tension and of water content in cells allows regulating the volume, but also impacts on the molecular crowding within the cells.

In this work we induce variations in the volume of individual cells in a controlled manner using osmotic shocks. Thanks to the addition of a sucrose solution, we generate an increase in the osmotic pressure resulting in a decrease of cell volume and an increase an increase in the concentration of species within the cell.

We monitor this process in single cells using Brillouin light scattering (BLS) and we use the elastic properties as a read out.[1] Light from an incident laser beam is backscattered by single cells and produces a slight frequency shift, due to the interaction between the incident light waves and the natural acoustic vibrations in the sample. The features of the backscattered spectrum allow extracting the sound velocity and attenuation at GHz frequencies.

Our results suggest that osmotic compression triggers a non-Newtonian response in the cells, characterized by a non-linear increase in the viscosity. This observation sheds a new light on the behavior of cells during volume changes, and might have important applications for the monitoring neurodegenerative diseases where the regulation of molecular crowding is altered.

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# Thermoelectric magnetotransport quantum oscillations in topological semimetal ZrTe<sub>5</sub>

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ZrTe<sub>5</sub> is a layered transition metal dichalcogenide (TMD) material that has been studied extensively over the past few years for its unique magnetotransport properties. Bulk ZrTe<sub>5</sub> single crystals display clear magnetoresistance Shubnikov de Haas (SdH) quantum oscillations under a magnetic field [1]. Similar SdH oscillations were observed in the thermopower, indicating that the electronic quantum properties can translate into thermal effects [2]. Recently, oscillations were also observed in the thermal conductivity without being linked to the SdH effect [3]. In this work, we measured simultaneously the electrical resistivity, the thermopower and the thermal conductivity on bulk ZrTe<sub>5</sub> single crystals grown by chemical vapor transport within a temperature range from 2 K to 400 K and a magnetic field up to 9 T. Our measurements can reveal the interplay between the electrical and thermal properties of ZrTe<sub>5</sub>. At 2 K, we observe clear oscillations mirror those measured for the electrical resistivity, which could indicate that the thermal oscillations are due to the contribution of electrons. We notice that the magnetoresistance at 9 T reaches 1600% while not exceeding 10% for the thermal conductivity, thus indicating a tiny, yet noticeable, electronic contribution to the thermal transport. Fig. 1 shows that the oscillations are 1/B periodic, following the SdH effect. The amplitude of the oscillations quickly decreases with the temperature, disappearing above a few Kelvins.

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Figure 1: Shubnikov de Haas quantum oscillations of electrical resistance and thermal conductivity in ZrTe<sub>5</sub> at 2 K.

### Thermoelastic properties of liquid metals measured by picosecond acoustics

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Picosecond acoustics (PA) is a time-resolved optical pump-probe technique based on reflectivity measurements used to study the propagation of acoustic waves in a large variety of samples at different pressure and temperature conditions [1, 2]. When associated with a resistively heated diamond anvil cell (RHDAC), it allows the precise determination of structural, thermoelastic and thermodynamic properties of materials through sound velocity measurements including equation of state [3], melting curves [5, 8], phase diagrams, or quantum effects [4, 6, 7].

In such experiments, it is possible to simultaneously measure the external conditions – of pressure and temperature – by recording transit time or surface phonon imaging consequently deducing the physical state and properties of the sample (liquid or solid) with a great accuracy.

The versatility and capabilities of PA combined with RHDAC will be illustrated with some results obtained in various liquid metals : the melting line of Indium [8], the complex variations in sound velocity of liquid alkali (Rubidium [7] and Cesium [6]) related to their "electride" state, or the opportunity for detecting liquid-liquid phase transitions in the stable or metastable liquid state of Gallium [5] and Bismuth [9].

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The control of single electrons has been a subject of intense research for several decades. A typical example is the definition of the electrical current of our SI unit system, which relies on a precise control and measurement of the electron charge over time. Recently, the concept of flying electron qubits has emerged [1], where the charge [2] or spin [3] degree of freedom of an electron are used as qubits that are manipulated and transported through electronic circuits. These qubits are particularly interesting because they can be controlled using simple electromagnetic fields. However, there are still challenges to be overcome in the implementation of flying electron qubits, including high-fidelity control of individual electrons and the design of scalable quantum circuits.

In this talk, I will introduce the different approaches to achieve this ambitious goal and present the latest advances in the field of single-electron transport. In particular, we will present an original method in which a single charge carrier is transported through a quantum electronic circuit by sound [2]. We show that electrons can be transported with an accuracy well above 99% [4], and we demonstrate that it is possible to confine the electron in a single propagating acoustic pulse using clever surface acoustic wave engineering [5]. Furthermore, by implementing synchronized transport of a single electron pair, we have demonstrate the antibunching of two individual electrons colliding on a beam splitter [6]. The presented study sheds light on the electron interaction in flight, paving the way for the implementation of controlled phase gates in sound-driven electron quantum optics experiments.

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#### Figures



**Figure 1:** Sound-assisted HOM interferometry. (a) Scanning-electron-microscopy image of the single-electron circuit of coupled quantum rails. A pair of single-electrons are synchronously sent from the upper (U) and the lower (L) source quantum dots (QD) towards the tunnel-coupled wire (TCW). (b) Single-shot transfer probabilities as a function of the delay between the two electrons in units of SAW period, *T*<sub>SAW</sub>. The employed notation corresponds to P<sub>20</sub> (both electrons at the lower detector), P<sub>11</sub> (one electron at each detector) and P<sub>02</sub> (both electrons at the upper detector).



### Phonon spectroscopy and phonon engineering in semiconductor nanowires

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The quantum-mechanical behavior of phonons can enable the observation of coherent phonon transport via phonon engineering. Nanowires (NWs) are rod-shaped crystals that represent an ideal platform for exploring phonon engineering, since they offer the possibility to design the phononic properties by enabling the formation of different kinds of superlattices (SLs). Besides the conventional SLs based on heterostructures, NWs can indeed host SLs based on crystal-phase homostructures, where the crystal composition is constant and different crystal structures are alternated. We demonstrated by Raman spectroscopy that a special type of SL, obtained introducing periodical twinning in a GaP zincblende lattice, enables a controlled design of the materials' phononic properties [1]. Our findings were corroborated by ab initio calculations, see Figure 1. Interestingly, we could decide à *la carte* the number of phonon modes by tuning the SL period, thus providing the first demonstration of phonon engineering in NWs.

Moreover, we explored the phononic properties of crystal phases that in bulk are hardly formed and can be controllably grown in NWs. This is the case, among others, of hexagonal-phase group IV-semiconductors as Ge [2] and SiGe [3]. The advent of hexagonal Ge and SiGe in NWs promises to overcome some of the limitations that have kept these materials out of the "photonic arena" so far. Indeed, the bandgap of hexagonal Ge is direct, a fact that could lead to the long-standing goal of designing Ge-based light-emitting materials. We probed the crystalline, phononic and electronic properties of hexagonal Ge and hexagonal-cubic Ge homostructures by a comprehensive investigation in NWs by combining polarization-resolved Raman experiments with density-functional-calculations.

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#### Figures



**Figure 1:** (a) TEM image of a GaP TSL NW. (b) Spacing between consecutive twin planes as a function of the position. (c,d) Calculated Raman spectra of GaP TSL (green line), bulk zincblende (black) and wurtzite (red). (e,f) Corresponding experimental Raman spectra [1,4].



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The engineering of acoustic phonon resonators in the GHz-THz frequency range may largely facilitate the manipulation of mechanical systems for ultra-high frequency optoelectronic and quantum applications [1]. To this end, robust confinement mechanisms for phonons with few-nanometer wavelengths and tailored transduction to other excitations in solids are highly desired. While in optics, electronics, and classical acoustics considering topological invariants has enabled the generation of robustly confined states, topological phases to engineer nanophononic devices remain a largely unexplored and promising field.

Here, we introduce the concept of topological invariants to nanoacoustics [2]. We experimentally construct topological nanoacoustic states through band inversion, i.e. by concatenating semiconductor superlattices with inverted spatial mode symmetries around a particular acoustic band gap. By embedding this type of nanoacoustic resonator in an optical Bragg micropillar cavity [3], the resulting monolithic device resonantly confines light and acoustic phonons at frequencies up to 300 GHz with large spatial overlap for efficient optophononic transduction. The topological nanoacoustic mode is evidenced experimentally through high resolution Brillouin spectroscopy [3] and time-resolved pump-probe spectroscopy [4]. We furthermore show that GaAlAs devices even allow perfectly co-localized topologically confined optical and nanoacoustic states [4] (Fig. 1). These states not only feature topological protection of both light and sound, but result in topologically robust light-sound transduction.

The devices introduced here can be readily integrated with fibered architectures and on-chip optical or mechanical waveguides, e.g., for sensing applications or on-chip generation of optical frequency combs.



**Figure 1:** (left) Perfect co-localization of light and ultra-high frequency phonons in a topologically protected interface state. (right) This state enables Brillouin light scattering, which remains orders of magnitude more robust under fluctuations compared to an optophononic Fabry-Perot resonator, from [4].

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### Strain and Layer-Twist Engineering of Valley Phonons in 2D Materials

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The scientific community has been highly interested in the unique properties of graphene, especially in the field of nanoelectronics. One remarkable aspect of graphene, h-BN, and transition metal dichalcogenides (TMDs) is their hexagonal lattice symmetry, which results in each valley having a distinct chiral structure. The chirality of opposite K points corresponds to the Dirac phonon and Dirac electron characteristics. Additionally, the breaking of inversion symmetry causes a gap to form, accompanied by a finite electronic Berry curvature. Recent studies have also revealed that the lattice dynamics of BN and TMDs can support topological chiral phonons with finite orbital angular momentum and Berry curvature.

Unfortunately, due to the constraints of lattice symmetry, these chiral phonon modes are typically limited to the corners of the Brillouin zone with a large wave vector, leaving little room for manipulation of their chiral characteristics by optical means. We demonstrate in a recent study [1] how the application of uniaxial strain can lead to the emergence of new chiral modes in the vicinity of the zone center, see Figure 1 which shows the emergence of new chiral modes at Q-points. Unlike the modes confined to the K points, these strain-induced chiral modes can be efficiently manipulated by adjusting the strain itself, which controls their position in the Brillouin zone. These findings provide a new technique for engineering the quantum properties of phonons in two-dimensional lattices.

Furthermore, twisting bilayer graphene has shown potential in manipulating the electronic Dirac-like properties, resulting in the discovery of flat bands at magic angles with interesting properties. In recent theoretical research [2], we have found that high-frequency lattice vibration modes experience significant band-flattening at these angles, including the valley Dirac phonon, valley optical phonon, and zone-center optical phonon bands, see Figure 2 indicating the phonon-band flattening at small twist angle. While an approximate approach estimates finite magic angles where phonon bandwidth vanishes, more accurate modelling that considers the existence of a restoring potential prohibits the emergence of a magic angle. Overall, the discovery of phonon band-flattening in graphene offers a promising avenue for exploring novel materials and technologies.

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**Figure 1.** Results obtained using a force-content model in TMD with 2% uniaxial strain. (a) Phonon dispersion  $(\omega_{\alpha\lambda})$ , (b) and (c) phonon eigenvector components  $(\epsilon_{\alpha\lambda})$  for phonon band  $\lambda = 3$  and  $\lambda = 4$ , respectively, and (d) phonon orbital angular momentum. The label  $\alpha$  indicates displacement along  $x_i$  and  $y_i$  for atom i in the unit cell.

**Figure 2.** The evolution of phonon dispersion with twist angle in the moiré Brillouin zone for LO/LA and TO modes at K and  $\Gamma$ . The left panels depict  $\theta = 4^{\circ}$  and the central panels show  $\theta$ = 1.05°. Relevant Dirac modes in the twisted cases are highlighted in red in panel (b). The right panels display the band renormalization factor R for each mode as a function of twist angle.



#### Changxiu Li

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#### Abstract

The demand of nanoimaging and nanometrology as well as signal processing in information and communication technologies, requires the progress on the high-frequency surface acoustic waves. Resorting to the semiconductor based nanostructures with atomic level layering precision and interface quality, the requirements for reaching THz frequencies are directly attainable [1]. In the first series of experiments we have observed a weak signature of Rayleigh modes on cleaved AlAs/GaAs superlattices (SLs) [2]. In the latest femtosecond laser spectroscopy experiments, an Al<sub>2</sub>O<sub>3</sub> thin film covers the cleaved Al<sub>0.7</sub>Ga<sub>0.3</sub>As/GaAs SL with gradient periods to prevent the AlGaAs layers from oxidation. Both laser configurations in the visible red/blue and UV/UV regimes are applied to monitor the coherent acoustic phonons with the beam incidence normal to the SL growth direction (Fig. 1 (a)). A frequency triplet at ~160, 196 and 293 GHz is observed at the SL position with a period of ~ 17.8 nm using the UV/UV setup (Fig. 1 (b) and (c)), while only the mode with the highest frequency is monitored by the visible light setup. Those frequencies scale inversely with the SL period, indicating phonons folding due to SL periodicity. In the calculated dispersion relations, the triplet has good correlation with the first-order k=0 Rayleigh, transverse, and longitudinal modes. The simultaneous observation of the surface wave and surface skimming bulk waves above 100 GHz, serves as a milestone towards the success of the THz opto-acoustic/acousto-optic transducers. Monitoring of sub-THz skimming waves opens the opportunities to evaluate the cleaved nanostructures with oxidised and rough surfaces.

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Figures



**Figure 1:** (a) Schematic of pump-probe experiments on the cleaved SL. The dimensions are not in scale. (b) Phononsinduced oscillations in transient optical reflectivity after removing the electronic background. (c) Spectra of the acoustic phonons shown in (b). (d) The frequency changes with SL periods, which vary on the cleaved surface with the positions (A, B, C, D, E) parallel to the SL layers. (e) The short-time Fourier transform of the signal obtained in one of the positions.



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#### Abstract

Both phonons diffraction and interference patterns are observed at the atomic scale in nanocomposite systems containing crystalline silicon and obstacles of nanometric voids or amorphous silicon inclusions with dimensions of the same order as the phonon wavelengths [1]. The phonon diffraction and interference patterns due to these nano-architectured systems are similar to the ones predicted by a simple Fresnel-Kirchhoff integral. These findings give evidence of the wave nature of phonons, and they can increase the comprehension of the phonons interaction with nanoobjects and at long term they can be useful for intelligent thermal management strategies and devices as well as a phonon frequency filtering at the nanoscale.

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**Figure 1:** Visualization of diffraction of a 6 THz longitudinal plane wave by an aperture with width of 3 nm. The color scale represents the kinetic energy per atom in eV.

## Recent Advances in Solving the Space-Dependent Phonon Boltzmann Transport Equation: from Materials Prediction to Inverse Design

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Heat and electrical transport can exhibit hydrodynamics signatures at the nanoscale, such as Poiseuille flux and vortices [1]. To model and optimize this non-diffusive behavior, the Boltzmann transport equation (BTE) must be used. However, unlike its diffusive counterpart, the BTE requires space, time, *and* momentum resolution, making BTE solvers computationally challenging. In this presentation, I will discuss our efforts to develop a framework to perform ab-initio BTE simulations in arbitrary 2D and 3D geometries using common laptops. I will introduce vectorial mean-free-path interpolation to reduce momentum-space degree-offreedoms [2] and iterative approaches to tackle heat transport in 2D materials [3]. The simulation workflow is illustrated in Fig. 1a. An example of calculating a structure's effective thermal conductivity tensor (ECT) will conclude this part (Fig. 1a).

In the second part, I will outline our recent work on the inverse design of nanoscale heat transport [4]. In this study, we identify optimal geometries with prescribed ETC. This talk will discuss several aspects, including the adjoint method for enabling gradient-based optimization, novel material interpolation, and reverse-mode automatic differentiation. Efforts in applying manufacturable constraints will also be addressed. In Fig.1c, an example of the inverse design of a 3D structure is reported, showing both the optimized solid and void phases. Two examples will be detailed: Optimizing thermoelectric materials and an anisotropic thermal spreader. The presentation will conclude with a demonstration of OpenBTE [5], our in-house parallel and GPU-enabled open-source software.

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**Figure 1:** (a) The workflow of OpenBTE. (b) Prediction of the effective thermal conductivity tensor (ECT) of a complex 2D material. (c) An example of inverse design. Both the solid and void phases are displayed.



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Enabling on-demand control of heat flow is key for the development of next generation of electronic devices, solid-state heat pumps and thermal logic [1]. Recently, ferroic oxides have been proposed for obtaining fast and reversible solid-state thermal switches with large switching ratios [2,3]. They rely on interactions between phonons conducting heat, and spontaneously occurring topological defects known as domain walls that move in response to an electric field or uniaxial stress [4]. However, a precise and agile tuning of the microscopic material parameters for adjusting thermal conductivities remains elusive. Here, we show that ferroelastic domain walls in lanthanum aluminate (LaAIO<sub>3</sub>) behave as boundaries that act like efficient controllers to govern thermal conductivity. At low temperature, we demonstrate a fivefold reduction in thermal conductivity induced by domain walls orthogonal to the heat flow and a twofold reduction when they are parallel to the heat flow (Fig. 1). With atomistic calculations, we observe that domain walls reduce the thermal transport moderately when parallel to the heat flow and do so more markedly when orthogonal to it, in good agreement with our experimental results. We account for these results by analyzing the temperature dependence of the measured thermal conductivity within the framework of the Holland model and derive a quantitative relation between thermal conductivity variations and domain wall organization and density.

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#### Figures



**Figure 1:** Thermal conductivity as a function of temperature of selected LaAlO<sub>3</sub> samples displaying distinct domain wall densities and orientations. Fitting lines originate from the Holland model.



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In recent times, hydrodynamic phonon transport has been intensively reviewed owing to its peculiar phonon collective behaviours [1-2], which are very similar to fluid dynamics. Graphitic materials, with their dominant normal scattering in nature, are currently considered as perfect hosts to establish the hydrodynamic flow [3]. However, observation of phonon Poiseuille flow in graphite is rarely claimed due to the experimental difficulties and missing theoretical understanding. In this work [4], we experimentally investigate phonon Poiseuille flow in microscale graphite ribbons with both natural (1.1% <sup>13</sup>C) and purified (0.02% <sup>13</sup>C) carbon isotope concentrations (Fig. 1a). We measure the thermal conductivity ( $\kappa$ ) of the graphite ribbons in a wide temperature range of 10-300 K. At 90 K, isotope scattering plays an important role in phonon transport, resulting in an enormous enhancement of thermal conductivity of purified graphite ribbon compared to that of the natural one with the same width of 5.5  $\mu$ m by 105% (Fig. 1b). We advocate more straightforward and rigorous criteria for ascertaining phonon hydrodynamic flow - a faster rise of  $\kappa$  than the ballistic thermal conductance ( $G_{\text{ballistic}}$ ). As shown in Fig. 1c, with the temperature increases,  $\kappa/G_{\text{ballistic}}$  shows a non-monotonous trend in the isotopically purified sample, indicating a clear transition of ballistic-hydrodynamic-diffusive regime. The value of  $\kappa/G_{\text{ballistic}}$  is enhanced by 51% from 40 to 90 K, this super-ballistic scaling of thermal conductivity with temperature is unambiguous evidence of phonon Poiseuille flow. While it continuously decreases in the natural one with increasing temperature, where the absence of phonon Poiseuille flow results from sufficient momentum-destroying isotope scattering. This work thus provides a deeper understanding of phonon transport in the whole regime of diffusive-hydrodynamic-ballistic and opens innovative possibilities for thermal management in modern micro-and nano-scale electronic devices.

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**Figure 1:** (a) Collective behaviours of phonons driven by temperature difference in a graphite ribbon. (b) Thermal conductivity ( $\kappa$ ) and (c) Ratio of  $\kappa$  over ballistic thermal conductance ( $G_{\text{ballistic}}$ ) as a function of temperature for purified (dark blue) and natural (light blue) graphite ribbons with a designed width of 5.5 µm.

#### Figures



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Elastic waves are an indispensable phononic technology finding applications in diverse fields ranging from the life sciences, signal processing to quantum technologies [1]. In my talk, I will introduce surface acoustic wave (SAW) control of the optical properties of epitaxial semiconductor quantum dots (QDs). On this platform QDs provide a unique ultrafast nanoscale strain sensor to map acoustic fields and an optically active two-level system for frequency transduction of single photons [2,3]. I will show how the underlying optomechanical interaction can be deliberately exploited for three-wave mixing of near infrared photons and gigahertz phonons by the optical dipole of the QD [3,4]. Finally, I will discuss directions to exploit the universal coupling of phonons to both electronic (QD) and photonic degrees of freedom in integrated device platforms [4] for future hybrid information processing technologies.

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Coherent acoustic phonons (CAPs) comprised into wavepackets with durations of several picoseconds are the main tool in ultrafast acoustics for probing [1,2] and exciting [3] the dynamics of lattice, electrons and spins in nanostructures on GHz-THz frequencies. The desire to control and excite materials with CAP wavepackets, also referred to as strain pulses, raises an efficiency problem. The two ways to address it are (i) to utilize processes highly sensitive to strain or (ii) to raise CAP amplitudes. But high-amplitude strain generation results in overheating which may damage delicate samples used in modern ultrafast acoustics such as quasi-2D materials [1] or biological cells [2]. Therefore (iii) a non-thermal mechanism of CAP photogeneration is required. Here we show experimentally that both directions (i), (ii) and (iii) can be achieved with the use of 1<sup>st</sup>-order ultrafast photo-induced phase transitions (PIPTs). Our proof-of-concept works employ phase transition in VO<sub>2</sub> owing to the following properties [4]: technologically attractive near-room temperature (340K); large ~1% lattice change; the PIPT occurs on a femtosecond timescale; insulator-to-metal transition leads to huge optical changes; 1<sup>st</sup>-order transition with significant latent heat. In the first experiment [5] we utilized sharp optical changes during PIPT and its sensitivity to strain. We injected a strain pulse with the amplitude 0.1% into VO<sub>2</sub> nanostructure, which led to ~1% increase or decrease of the transformed VO<sub>2</sub> volume, depending on the sign of the strain at the moment of the PIPT trigger. Such sign dependence allows one to control the effect by moving a delay line. In the second experiment [6] we constructed an optoacoustic transducer of  $VO_2$  to utilize the large ultrafast lattice change for CAP generation. We observed the strain pulses generated during PIPT and propagated away and deduced the highest strain amplitude reaching 1.4% in VO<sub>2</sub>. We found a new contribution to CAP generation saturated at 0.45% which consumed laser energy rather than dissipating it into heat. This contribution lowered the transducer heating by a factor of 3 for the same CAP amplitude generation. To date, only few works on ultrafast acoustics utilized PIPTs [7,8]. With the concepts shown here we hope to encourage research considering various PIPTs such as magnetic [8] or ferroelectric ones.

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Figure 1: The proof-of-concept experiments: 1) Strain pulse affects PIPT in VO<sub>2</sub>; 2) Strain pulse emitted upon PIPT in VO<sub>2</sub>

# Apertures for generating spatial superoscillations of coherent acoustic phonons

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Superoscillation is an interference effect whereby, in specific regions, functions can oscillate faster than their fastest Fourier component. This can be exploited to achieve superresolution, where the diffraction limit may be broken, as has been demonstrated in optical imaging [1]. In this presentation we discuss the approaches we have taken to design superresolving devices for phonons by exciting an opto-acoustic transducer with a spatially modulated femtosecond-pulsed light beam.

Our objective was to design an array of standard apertures (holes, discs, annuli) in an optically opaque mask such that the overall diffraction pattern obtained by illuminating the mask with light of a given wavelength generates a superoscillating optical field at the opto-acoustic transducer thereby generating a superoscillating acoustic strain field. Our route to solving the inverse problem of finding the required positions for the apertures in the mask was by aiming to generate a far-field pattern within the broad class of superoscillations constructed by Aharonov [2] with well-characterized Fourier components. A simulation of the acoustic superoscillations generated by a grating pattern is shown in Fig.1(b).

In the pump-probe experiments, gold shadow masks corresponding to the aperture designs were fabricated by optical lithography on the surface of a 5 GHz Ga(Al)As superlattice transducer, Fig. 1(c). Where the pump light fell on gold it was reflected leaving a spatially modulated light intensity distribution on the transducer. The spatial distribution of the 5 GHz phonon signal intensity on the opposite side of the 450 micron-thick GaAs substrate was measured using a scanning probe beam, Fig. 1(d).

Further work is being done to generalise the solving process by using overlap integrals to resolve both aperture widths and offsets.

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**Figure 1: (a)** Designs for a diffraction grating superoscillating aperture array. **(b)** Simulated results in natural units for the design in **(a)** with a phonon wavelength of approximately 1µm. The orange dashed line shows the simulated intensity distribution of a single slit compared to the solid blue line of the superoscillating aperture array. As can be seen there are features smaller than the highest Fourier component, a superoscillation. **(c)** A gold shadow mask for the opto-acoustic transducer in the shape of the superoscillating concentric annuli aperture array described by Torlado di Francia [3], examined by Cox et al [1] and produced by us using optical lithography. **(d)** Results from an initial investigation of the design in **(c).** Due to noise and acoustic anisotropy, it is difficult to resolve the superoscillation.

#### Filippo Bencivenga

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Phonon dynamics and heat transport processes at the nanoscale are of fundamental relevance in condensed matter physics and play a key role in advancing modern technology. Experimental tools to investigate these processes at sub-100 nm length-scales and on the relevant (ps) timescale are mainly based on the combination between ultrafast laser excitation and sample's nanostructuration.

The recent development of the extreme ultraviolet transient grating (EUV TG) approach, nowadays available at the FERMI free electron laser user facility (Triste, Italy), represents an alternative way to reach sub-100 nm and sub-ps length- and time-scales [1,2]. The method is based on the generation of an ultrafast and nanoscale interference pattern of light intensity, via the interference between two crossed EUV photon pulses, while the thermoelastic response of the sample is read, via transient diffraction, by a third and time-delayed EUV pulse.

We will show how this novel experimental tool can be used for probing: (i) bulk and surface phonons on a previously inaccessible wavelength range and in ultra-thin samples [1-4]; (ii) nanoscale thermal transport in crystalline silicon and amorphous silicon nitride membranes, where heat transport timescale in the crystalline sample shows an order of magnitude deviation with respect to the diffusive regime, and (iii) time-domain stimulated Brillouin scattering with wavevectors as large as  $\approx 1 \text{ nm}^{-1}$  [5].

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# Using X-ray emission spectroscopy to measure the electron-phonon scattering rates in the demagnetization transient state of ferromagnets

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The experimental determination of microscopic processes in magnetic systems is important to better understand their macroscopic properties like the ultrafast demagnetization [1]. In crystalline ferromagnets, one of the main microscopic mechanisms of spin relaxation is the electron-phonon driven spin-flip, or Elliott-Yafet, scattering [2-4]. Therefore, the experimental determination of the electron-phonon scattering rate in solids is of first importance in order to better understand the demagnetization mechanisms. However, while most effort has been dedicated to the measurement of the demagnetization time using pump-probe experiments, experimental studies of the underlying microscopic mechanisms are still scarce. To measure the spin-flip scattering rate, we exploit the stringent atomic symmetry selection rules of X-ray Emission Spectroscopy (XES) and observe the quantifiable change in the decay peak intensities in spectra when changing the temperature, *i.e.* when changing the phonon population. We show a reduction of the decay peak intensity for the Ni model system and its absence in the diamagnetic Cu case [5]. In FeNi alloys, this approach evidences a thresholding of the Elliott-Yafet mechanism by the intra- and intersublattice exchange energies [6]. In Gd, where the magnetic moment is held by the 5d and the 4f electrons, we evidence an Elliott-Yafet mechanism for the itinerant 5d electrons and its absence for the localized 4f electrons [7].

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#### Figures



Figure 1: Temperature-dependent L-edge X-ray emission spectra of nickel [5].



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#### Abstract

The existence of a net thermal current of conductive thermal waves is demonstrated even in the absence of a mean temperature gradient. This effect, which we call heat shuttling, is generated by the temperature dependence of the thermal conductivity of materials excited with a thermal excitation periodically modulated in time (Fig. 1(a)). We show that this modulation gives rise to a thermal current superimposed on the one generated by the mean temperature gradient, which enhances the heat transport when the thermal conductivity increases with temperature. By contrast, if the thermal conductivity decreases as temperature increases, the thermal-wave heat current inverts its direction and reduces the total heat flux (Fig. 1(b)). The reported shutting effect is sensitive to the amplitude of the periodic thermal excitation, which can facilitate its observation and application to harvest energy from the temperature variations of the environment.



**Figure 1:** (a) Scheme of a phase-change material supporting the heat conduction between two thermal baths due to their temperature difference  $\delta T = \Delta T \cos(\omega t)$  periodically modulated in time *t*. (b) Average (net) heat flux as a function of the temperature  $T_h = T_c$ , for three amplitudes of the periodic excitation.



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Phase change materials for embedded memory automotive applications are dynamically evolving with the aim of high temperature data retention [1]. This requires higher crystallization temperature and amorphous phase stability, which was achieved by increasing the Germanium content and doping with Nitrogen in the traditional Ge<sub>2</sub>Sb<sub>2</sub>Te<sub>5</sub> (GST-225) [2]. Here, we study the temperature dependent structural evolution of Ge-rich GST (GGST) and Ge-rich GST N-doped (GGSTN). The spectra of this ternary alloy is composed of Ge-Te vibrations arising from distorted octahedral (o) and tetrahedral units (t), Sb-Te vibrations and Ge-Ge vibrations. We observed an increase in crystallization temperature (T<sub>c</sub>) by 40°C between GGST and GGSTN signified by change of amorphous Ge vibrations at 275 cm<sup>-1</sup> to crystalline Ge vibrations at 300 cm<sup>-1</sup> (Fig (1,2)). The vibrations in amorphous phase displayed thermal stability with no significant structural changes below the crystallization temperature. Further, in-situ measurements revealed the temperature dependence of vibrations showing weak sensitivity of Ge-Te vibrations to temperature compared to Sb-Te and Ge-Ge vibrations. The sensitivity of vibrations to temperature in GGST and GGSTN was found out to be different. For both GGST and GGSTN, SbTe vibrations in amorphous phase and crystalline Ge-Ge vibrations displayed a strong linear dependence on temperature. In amorphous phase, the Sb-Te vibrations sensitivity was -0.018 cm<sup>-1</sup>K<sup>-1</sup> in GGST, whereas in GGSTN was -0.015 cm<sup>-1</sup>K<sup>-1</sup> which can be a consequence of N-doping. Similarly, for crystalline phase, Ge-Ge vibrations sensitivity was extracted to be -0.019 cm<sup>-1</sup>K<sup>-1</sup> in GGST and -0.021 cm<sup>-1</sup>K<sup>-1</sup> in GGSTN. These temperature calibration coefficients are crucial for extracting the power sensitivity of these vibrations and hence, the global thermal conductance of thin films. Finally, we were successful in extraction of temperature dependent thermal conductivity for this ternary alloy aided with finite element simulations.

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Figures









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The origin of phonon thermal Hall Effect (THE) observed in a variety of insulators is yet to be identified. Here, we report on the observation of a thermal Hall conductivity in a non-magnetic elemental insulator, with an amplitude exceeding what has been previously observed. In black phosphorus (BP), the longitudinal ( $\kappa_{ii}$ ), and the transverse,  $\kappa_{ij}$ , thermal conductivities peak at the same temperature and at this peak temperature, the  $\kappa_{ii}/\kappa_{ii}/B$  is ~10<sup>-4</sup>-10<sup>-3</sup> T<sup>-1</sup>. Both these features are shared by other insulators displaying THE, despite an absolute amplitude spreading over three orders of magnitude. The absence of correlation between the thermal Hall angle and the phonon mean-free-path imposes a severe constraint for theoretical scenarios of THE. We show that in BP a longitudinal and a transverse acoustic phonon mode anti-cross, facilitating wave-like transport across modes. The anisotropic charge distribution surrounding atomic bonds can pave the way for coupling between phonons and the magnetic field.

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Figure 1: Phonon thermal Hall conductivity (a-c) and angle (d) in black phosphorus.

# Thermal conductivity of GaAs-GaP superlattice nanowires using the thermal bridge method

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In the last decades, continuous efforts have been made to understand and control phonons with great potential for numerous technological applications [1]. Our objective is to investigate phonons interference effects and different phonon transport regimes, which are crucial for the manipulation of phonons. Nanowires are promising candidates for studying the phonons interference effect because they offer unique possibilities in terms of heterostructuring as well as of branched nanowire junctions. In this respect, a superlattice (SL), i.e. a lattice made by different materials alternated periodically, can be used to investigate the behavior of phonons scattered from interfaces. Phonons scattered from single interfaces lose their phase information, leading to diffusive thermal transport. However, for periodic repetition of interfaces with a period comparable to the phonon mean free path, scattered phonons can interfere before losing their phase information resulting in a modified phonon dispersion and thermal conductivity of the nanowire [2,3].

In this project, we are investigating phonon interferences effect in SL nanowire through thermal conductivity measurements. The thermal conductivity of SL nanowires is measured using the thermal bridge method for which we fabricated microdevices consisting of platinum coils patterned on suspended SiNx membranes to ensure thermal isolation [4,5]. The thermal conductivity of GaAs-GaP superlattice nanowire is measured for various SL periods (Fig1) [6]. We observed that the conductivity values decrease with increasing period length and there is a minimum around a period of 10 nm, after which the conductivity increases for longer periods. The minimum in thermal conductivity decreases due to scattering from increasing number of interfaces. This work is a promising step to use SL nanowires as a powerful platform for exploiting coherent phonons in thermal applications.



Figure 1. (a) Optical and SEM image of suspended microdevice (b) Thermal conductivity as a function of superlattice period.

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Thermal transport plays a crucial role in the performance of electronic and optoelectronic devices. Understanding the underlying mechanisms of heat transfer in two-dimensional (2D) materials, such as transition metal dichalcogenides (TMDs), is of great importance for the development of high-performance devices. However, the currently available techniques for determining the heat transport in 2D materials have significant limitations, such as the requirement for accurate knowledge of material parameters like thickness, optical absorption, and heat capacity, and the need for relatively strong heating ( $\Delta T \sim 100$  K) [1].

In this study, we address these challenges and present a novel spatiotemporal pump-probe microscopy technique [2] that takes advantage of the spatial profile measured at a pre-time-zero pump-probe delay. This means that the probe pulses arrive before the pump pulses, making the probe sensitive to residual heat generated by previous pump pulses from the incident pulse train. In our case, this pre-time-zero corresponds to a time delay of ~13 nanoseconds. We demonstrate the efficacy of our method by quantifying the thermal diffusivity (*D*) of four TMD materials, obtaining,  $0.18 \pm 0.01 \text{ cm}^2/\text{s}$  for MoSe<sub>2</sub>,  $0.20 \pm 0.03 \text{ cm}^2/\text{s}$  for WSe<sub>2</sub>,  $0.35 \pm 0.03 \text{ cm}^2/\text{s}$  for MoS<sub>2</sub> and  $0.59 \pm 0.07 \text{ cm}^2/\text{s}$  for WS<sub>2</sub>, in excellent agreement with reported thermal properties [3]. We also explore the thickness dependence of *D* in MoSe<sub>2</sub>, ranging from 3L to 32L, and observe good agreement with recently reported literature [4], highlighting the accuracy and versatility of our technique in quantifying the thermal properties.

We also predict that our proposed technique could provide a pathway to investigate non-Fourier transport and other unconventional thermal transport phenomena in low-dimensional materials.

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**Figure 1:** (a) Optical microscope image of a suspended TMD. (b, c) Normalized transient reflectivity maps at time delay  $\Delta t = 0$  (time-zero) and  $\Delta t < 0$  (pre-time-zero), respectively. The broader profile in (c) is due to the diffused phonon heat in the sample.

# Magnetic proximity effect in hybrid ferromagnet-semiconductor structures mediated by chiral phonons

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Hybrid ferromagnet-semiconductor systems possess new outstanding properties which emerge when bringing magnetic and semiconductor materials into contact. In such structures the long-range magnetic proximity effect couples the spin systems of the ferromagnet and semiconductor on distances exceeding the carrier wave function overlap as manifested by optical studies [1]. In particular I will focus on effective p-d exchange interaction of acceptor-bound holes in the CdTe quantum well with d-electrons of the ferromagnet (Co, Fe, magnetite) where the coupling persists on the distances of up 30 nm [2]. Such indirect interaction is established via the phononic ac Stark effect mediated by the chiral phonons. The presence of elliptically polarized phonons in the FM layer is associated with the interaction between phonons and magnons, which leads to strong circular dichroism. In contrast to charge carriers phonons do not experience noticeable potential barrier at the interface between the FM layer and the QW. Thus, chiral phonons with polarization defined by the FM magnetization, can penetrate into the quantum well and interact with charge carriers. The spin-phonon interaction with holes removes the Kramers degeneracy of ±3/2 doublet even in a zero external magnetic field. The magnitude of the exchange splitting for the acceptor spin levels in the QW corresponds to 0.1 meV as directly measured using spin-flip Raman scattering [3]. Moreover, low-voltage-control of the exchange coupling in the same type of hybrid structures enables promising routes in the quest for electrically tunable ferromagnetism [4].

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#### Abstract

In this talk, the experimental direct observation of the coherent oscillation between magnons and phonons are presented, together with demonstration of anomalous reflection of magnons owing to magnon-phonon hybridization [1]. We developed time-resolved magneto-optical imaging technique by combining conventional magneto-optical imaging and pump-and-probe spectroscopy, which enables us to obtain snapshots of spin-wave propagation dynamics in real space with a temporal resolution of sub nanoseconds. In a Bi-doped magnetic garnet, Lu 2 Bi 1 Fe 3.4 Ga 1.6 O 12, we observed coherent temporal oscillation between magnons and phonons as a result of hybridization, where magnons and phonons are coherently interconverted to each other during propagation. It is also found that the magnon-phonon hybridized wave exhibits abnormal reflection at the sample edge owing to the mode degree of freedom of phonons [2]. Since phonons have longitudinal and transverse modes, both modes may not be an eigenstate where translational symmetry is broken down, such as a sample edge. Owing to the mode degree of freedom, the hybridized wave may split into two reflected waves with the same frequency, which is not the case for pure magnon propagation. The experimental demonstration of these dynamics of magnon-phonon hybridized waves will be reported in the talk.

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### Emerging spin-phonon coupling through cross-talk of two magnetic sublattices

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Many material properties such as superconductivity, magnetoresistance or magnetoelectricity emerge from the interactions between spins and the crystal lattice/phonons. An in-depth understanding of spin–phonon coupling is therefore at the heart of many properties. When talking about spin-phonon coupling, most examples concern the crosstalk of the lattice vibrations with one magnetic lattice only. Yet, the simultaneous presence of multiple magnetic orderings may yield non-linear spin-phonon interactions and potentially unknown properties. Here, we demonstrate a strong spin–phonon coupling in SmFeO<sub>3</sub> that emerges from the interaction of two spin systems: magnetic order of iron and the one of samarium. We probe this coupling as a significant shift of phonon frequencies and the appearance of new phonon modes when both magnetic orders are activated. The spin–phonon coupling is absent for the magnetic ordering of iron alone. It only emerges with the additional ordering of the samarium spins. Intriguingly, the samarium ordering is not spontaneous. But it is induced by the iron magnetism. This emergent spin-phonon coupling from the non-linear interaction of multiple orders, which do not need to occur spontaneously, this indicates a conceptually different approach in the search for yet unknown properties.

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**Figures** 



Figure 1: Anomalous deviations of lattice vibrations in SmFeO<sub>3</sub>.



# Ultrafast x-ray scattering reveals composite amplitude collective mode in Weyl charge density wave material (TaSe<sub>4</sub>)<sub>2</sub>I

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(TaSe<sub>4</sub>)<sub>2</sub>I is a quasi-1D Weyl semimetal [1] that hosts a low-temperature charge density wave state with a complex lattice distortion along multiple phonon coordinates [2, 3]. It has lately attracted much research interest as a putative dynamical axion insulator due to recent magnetotransport measurements in the CDW phase [4]. We report ultrafast x-ray scattering experiments of  $(TaSe_4)_2I$  following photoexcitation with femtosecond infrared laser pulses, carried out at the LCLS and SACLA hard x-ray free-electron laser facilities [5]. Time-resolved diffraction signals were obtained at the first-order ( $q_{CDW}$ ) and second-order ( $2q_{CDW}$ ) CDW sidebands surrounding the (2 2 4) crystal Bragg peak. Two modes with frequencies of 0.11 and 0.23 THz are observed at the CDW sidebands. Based on analysis of the atomic displacement polarizations we identify the 0.11 THz mode as the amplitude mode derived primarily from the transverse acoustic component of the CDW static distortion. This mode does not exhibit any pump-induced softening, even up to fluences that entirely quench the CDW order, and the excitation of this mode appears to be displacive in nature. We explain these features with a model of an indirect excitation of this mode mediated through a coupling to the optical phonon component associated with a tetramerization of the Ta chains [6], which is predicted to be involved in structural instabilities of the high-symmetry phase [7].

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**Figure 1:** (a) Crystal structure of  $(TaSe_4)_2I$ . (b) Schematic of the ultrafast infrared-pump x-ray-probe experiments performed at LCLS and SACLA. (c) Fluence dependence of the diffraction signal measured at the CDW sidebands, showing the 0.11 THz amplitude mode derived from the transverse acoustic phonon mode of the high temperature phase.



### THz Time-Domain Phonon Spectroscopy for van der Waals Coupling of 2D MoS<sub>2</sub> Interfaces

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Semiconducting 2D materials such as layered-stacking MoS<sub>2</sub> have been widely considered as a versatile platform for electronics design and for novel physics research. Since the van der Waals (vdWs) interfaces are often defective, it is crucial to have a non-destructive detection technique to monitor the interface quality. In addition, the acoustical behavior of 2D-based devices determines the heat transfer pathway, and thus understanding it is important for the design of electronics and optoelectronics. In this work, we first used picosecond ultrasonics technique to observe the actual interlayer vdWs bonding strength of epitaxiallygrown bi-layer and tri-layer MoS<sub>2</sub> [1]. The optical observation of all layer breathing vibrational modes beyond 1 THz reveals both Raman-active and Raman-inactive modes (see Figure 1(a)). We extended the linear chain model by considering the substrate effect and the next nearest neighbor vdWs coupling. We further considered the intralayer stiffness as a correction term to acquire the actual interlayer vdWs coupling. Our results provided the interlayer force constants for the vdWs interfaces. In addition, type-II van der Waals heterojunctions (vdWHs) consisting of transferred monolayer and bi-layer MoS<sub>2</sub> on GaN (0001) substrate were investigated [2]. The generation and coupling of acoustic pulses into the substrate were evidenced by the measured Brillouin oscillation. Using a piezoelectric InGaN single quantum well photoacoustic transducer (see Figure 1(b)), we observed an asymmetric bipolar acoustic strain wave, which describes the surface of a substrate undergoing strong compressive deformation after weak tensile deformation in the out-of-plane direction. We developed a theory to explain the mechanisms responsible for the observed strain waveform in the vdWHs elastic system. Through fitting, we obtained the parameters of the carrier dynamics, which will complement our understanding of the thermal transfer at the 2D/substrate interface. This work is sponsored by National Science and Technology Council under MOST 110-2112-M-002-033-MY3.

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**Figure 1:** (a) The top images show the TEM measurements of our as-grown few-layer MoS<sub>2</sub> samples. The bottom graph shows the spectra obtained by the THz coherent phonon spectroscopy. (b) Schematic of our MoS<sub>2</sub>/GaN vdWs heterostructures cross section.

#### Figures



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Optical-domain transient grating (TG) spectroscopy is a versatile background-free four-wave-mixing technique that is used to probe vibrational, magnetic and electronic degrees of freedom in the time domain. The newly developed coherent X-ray free-electron laser (XFEL) sources allow its extension to the X-ray regime. X-rays offer multiple advantages for TG: their large penetration depth allows probing the bulk properties of materials, their element specificity can address core excited states, and their short wavelengths create excitation gratings with unprecedented momentum transfer and spatial resolution. Here, we demonstrate TG excitation in the hard X-ray range at 7.1 keV using a single XFEL pulse exploiting the Talbot effect, in contrast with the usual crossing beam setup since it becomes very challenging in the hard X-ray range. In bismuth germanate (BGO), the non-resonant TG excitation generates coherent optical phonons detected as a function of time by diffraction of an optical probe pulse. This experiment [1] demonstrates the ability to probe bulk properties of materials and paves the way for ultrafast coherent four-wave-mixing techniques using X-ray probes and involving nanoscale TG spatial periods.

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#### Figures



**Figure 1:** XTG signals from BGO at 7.1 keV with an excitation grating pitch of 770 nm. Panel a) shows the normalized time trace at short times (< 5 ps). The non-oscillatory decay of the signal is fitted (in black) with a single exponential. The fast oscillations are attributed to optical phonon coherence [2]. The Fourier transform of the fast-oscillatory component is presented in panel b) (residuals from the fit in a are shown in the inset).

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Lattices of exciton polariton condensates represent a rich platform for the study and implementation of non-Hermitian non-linear bosonic quantum systems. Actuation with a time dependent drive provides the means, for example, to perform Floquet engineering, Landau-Zenner-Stückelberg state preparation, and to induce resonant inter-level transitions. We introduce polaromechanical metamaterials, two-dimensional arrays of µm-size zero-dimensional traps confining light-matter polariton fluids and GHz phonons. A strong excitonmediated polariton-phonon interaction [1] can be exploited in these metamaterials, both using electrically injected bulk-acoustic waves [2] or self-induced coherent mechanical oscillations [3], to induce a timedependence in the level energies and/or in the inter-site polariton coupling J(t). This has remarkable consequences for the dynamics. For example, when locally perturbed by continuous wave optical excitation, polariton condensates respond by locking the energy detuning between neighbour sites at multiples of the phonon energy [4]. We theoretically describe these observations in terms of synchronization phenomena involving the polariton and phonon fields. We study lattices of closely connected traps (defined by a linear optomechanical coupling), and also well separated traps (characterized by a quadratic optomechanical coupling), and discuss the role of dissipation and non-linearities in the stability of the observed asynchronous locking. The described metamaterials open the path for the study of non-reciprocal transport in dissipative quantum light fluids spatially and temporally modulated by GHz hypersound.

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Figures



**Figure 1:** Left: scheme of the sample and optomechanically-induced phenomena. Right: Asynchronous locking of the energies of polariton condensates in neighbour sites of the lattice.



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#### Abstract

It is now possible to engineer the quantum state of macroscopic mechanical resonators. This opens up the possibility for new quantum technologies, such as quantum-enhanced sensors and quantum memories/interfaces for quantum computers. However, ultrahigh vacuum or cryogenic environments are currently required, limiting the breadth of applications.

In this presentation I will introduce a new approach to control the quantum state of a macroscopic mechanical resonator via measurement and conditioning [1]. This approach is based on continuous position measurement, but – unlike other schemes – operates with a measurement that is faster than the mechanical oscillation. By operating in this regime, beyond the usual rotating wave regime, it is possible to prepare quantum squeezed states of motion. Remarkably, our theory predicts that the experimental requirements are greatly relaxed, even compared to mechanical ground-state cooling, to the point that quantum state preparation is feasible at room temperature with existing technology.

I will present experiments showing a classical version of the predicted squeezing effect in a new engineered double-disk optomechanical device fabricated on a silicon chip [2]. These experiments take advantage of both structural damping – which we show that, compared to the usual viscous damping, can improve quantum state preparation – and arrays of mechanical modes. Specifically, I will present experiments demonstrating that continuous position measurement can prepare thermomechanical squeezed states of motion, and to this for ensembles of structurally damped mechanical resonances.

Together, our results provide a new way to generate nonclassical states of macroscopic mechanical oscillators and open the door to quantum sensing and tests of quantum macroscopicity at room temperature.

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Figure 1: Schematic of experiment. A: illustration of double-disk. B: Photograph of fabricated device. C: Experiment.



### Coupled membrane nanomechanical resonators, for room temperature phononcavity electromechanics

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#### Abstract

Phonon-cavity electromechanics studies the interactions between two mechanical modes by analogy with optomechanics [1]. It is interesting because it can transmit information carried by phonons between mechanical modes and to filter signals in different frequency bands by controlling the energy transfer [2]. However, most previous work has relied on mechanical coupling, which limits the resonant frequency and flexible tunability of the integration. Here, we will present our recent achievements in developing a novel nanoelectromechanical system, consisting of capacitive coupling of two distinct resonators: a silicon nitride drum resonator (with resonance frequency  $\Omega_{m1}$ ) and an aluminum one (with resonance frequency  $\Omega_{m2}$ ) [3-4]. The whole device structure can be viewed as a parallel plate capacitor, where each plate is a membrane drum. Integrated with a versatile platform composed of microwave reflectometry and a microwave cavity, both resonators can be manipulated and detected independently.

With this unique device structure, we first investigated phonon-cavity electromechanics by taking the silicon nitride drum as a phonon cavity and performing double-tone operations, through an analogue of a microwave optomechanical system [4]. Here, it is quite different from conventional optomechanical systems in which the mechanical damping rate is usually much smaller than that of the coupled cavity. Electromechanically induced transparency and amplification of input signals have been demonstrated by sideband pumping the phonon-cavity. This unique coupled system gives access to the observation of the phonon-cavity force affecting the mechanical damping rate of both movable objects. Second, we explore a three-tone operation scheme by pumping the phonon-cavity at its blue and red sideband at the same time and using the third tone to probe either the cavity or the coupled Al drum, in analogy with the back-action evading scheme in optomechanics. We have observed phase sensitive parametric amplification in this double pump scheme, as shown in Fig.1. Besides, destructive interferences between the probe signal and these phonons generated by the double-pump tones have also been observed. These measurement results are in good agreement with our analytical calculations based on electrostatic coupling model. Our results open up new possibilities in the study of phonon-cavity based signal processing in the classical and potentially in the future in the quantum regimes. Besides, this kind of device will offer more degrees of freedom in routing propagation of photons and phonons in microwave optomechanical circuits.

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[4] Alok Pokharel, Hao Xu, Srisaran Venkatachalam, Eddy Collin, Xin Zhou, Nano Lett., 22 (2022), 7351-7357 Figure



**Figure 1:** phase sensitive signal amplifications measured by pumping the phonon-cavity at its red ( $\Omega_{m1}$ - $\Omega_{m2}$ ) and blue ( $\Omega_{m1}$ + $\Omega_{m2}$ ) sideband at the same time and probing signal at  $\Omega_{m1}$ . All measurements are performed at room temperature.



# Quantum control of an ultracoherent mechanical resonator with a fluxonium qubit

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In recent years, chip-scale phononic-crystal membranes have demonstrated impressive lifetimes exceeding 100 seconds and coherence times on the order of seconds, even in thermal environments at 10 mK [1]. These remarkable achievements have been made possible through the utilization of softly-clamped siliconnitride membranes operating in the MHz frequency range. Establishing a strong coupling between these exceptional mechanical resonators and superconducting qubits, which operate in the GHz frequency range and represent a highly promising platform for scalable quantum computers, has long been a sought-after objective. Such a coupling holds the potential to unlock a host of quantum technology applications, including ground-breaking quantum memories, microwave-optical quantum transducers [2], and even fundamental tests of quantum gravity [3] through the creation of superpositions of non-Gaussian states in the membrane. However, the primary challenge lies in bridging the significant frequency disparity between these two quantum devices, often on the order of 10^3. Inspired by recent works [4, 5], our research group has proposed a novel coupling scheme that addresses this long-standing challenge. We have developed a state-of-the-art 2 MHz qubit architecture with exceptional coherence times, allowing us to achieve direct resonant coupling with phononic-crystal membranes MHz phononic crystal membranes [6]. I will present first results on the qubit and mechanical systems.

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The internal vibrational modes of molecules embedded in plasmonic nanogap can be used as ultrahigh frequency (1 – 50 THz) mechanical oscillators and constitute a new form of optomechanical nanocavities [1]. I will first briefly review recent experiments evidencing quantum correlations that are generated between light and collective molecular vibrations in the process of spontaneous off-resonant Raman scattering [2-5], in the absence of any cavity. Then, I will introduce our work on plasmonic gap modes [6,7] and show that coupling molecular vibrations to properly designed dual-resonant plasmonic cavities [8] allows for the observation of coherent optomechanical transduction between mid-infrared (~32 THz) and visible (~450 THz) electromagnetic fields [9]. I will conclude with some perspectives and challenges in the field of molecular cavity optomechanics.

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#### Figures



**Figure 1:** Coherent frequency conversion with a molecular optomechanical cavity. (a,b) Simulated field enhancement factors for mid-IR and visible illumination. The lower panel shows an SEM picture of the nanoparticle-in-groove cavity, while the right panel is an artistic rendering of the device. Estimated enhancement factors result in 13 orders of magnitude improvement in per-molecule upconversion efficiency. (c) Example of high-resolution Raman spectrum (Stokes sideband, laser at 405 THz) with the upconverted signal (colored lines) riding on the spontaneous emission line (grey shade) as the mid-IR laser frequency is tuned around 32.4 THz (or 1080 cm<sup>-1</sup>).



# Picoseconds lattice dynamics in multiferroic BiFeO<sub>3</sub> probed by time-resolved x-ray diffraction

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The understanding of the lattice dynamics in ferroic compounds driven by an ultrashort light pulse is an exciting research direction due to the exceptional non-linear properties (optical, elastic, electric and magnetic) of ferroic and multiferroic materials [1-5]. Photo-induced strain in ferroic materials is driven by a complex interplay between charge, phonon and spin dynamics with microscopic mechanisms that still need to be determined [5-10]. We present recent experiments where ultrafast photoinduced strain is evaluated in BiFeO<sub>3</sub>-based multiferroic materials, with a focus on the description of the ultrafast symmetry change of the unit-cell that appears after an ultrashort laser pulse. A combination of optical and X-ray time-resolved techniques enables to clearly demonstrate how the light excitation can lead to a modulation of the symmetry in ferroic materials. By studying two asymmetric Bragg reflections (i.e. h0l and -h0l for instance) of a BiFeO<sub>3</sub> crystal, we show how it is possible to disentangle at the picosecond time scale the light-induced longitudinal and shear strain in the unit cell. The talk will be split into two parts. After presenting results obtained on a (001)<sub>c</sub> single domain Single crystal of BiFeO<sub>3</sub> [11], I will present the latest results obtained on a well-characterized single domain BiFeO<sub>3</sub> nanofilm. I will show that in this sample we can fully reconstruct the lattice deformation after the ultra-short laser pulse illumination [12]. The unit-cell distortion leads to the modulation of the ferroelectric polarization at picoseconds timescale.

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#### Figures



**Figure 1:** Left: Bragg diffraction peak of the (101)<sub>c</sub> at equilibrium. Right: (101)<sub>c</sub> Bragg diffraction peak intensity variation at 200 ps after ultra-short laser illumination.



# Polar super-orders and folded acoustic phonons in $BiFeO_3/LaFeO_3$ superlattices

#### Pascal Ruello<sup>1</sup>

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A superlattice (SLs) is a periodic structure of layers of two (or more) materials. This periodic stack offers a unique opportunity to control the folding the Brillouin zone. This folding gives rise to additional electronic and phonons bands that enrich the properties of the materials. For instance, this artificial chemical modulation is widely exploited in semiconductors to tailor the electronic structure [1] and has leaded to the development of the quantum cascade laser [2]. Such superlattices are also a playground for phonon physics [3] and several works have demonstrated how to drive picosecond strain pulse and to control the spectrum of THz coherent acoustic phonons in semiconductors [4-8] and oxides [8-11]. Here we explore the role of supplementary periodic polar and structural orders in the multiferroic BiFeO3/LaFeO3 (BFO/LFO) SLs. Using transmission electron microscopy, X-ray diffraction and first-principles calculations, we thus show that in-plane and out-ofplane extra polar and structural orders take place. In particular, the out-of-plane polar and structural orders develop with a period that is about twice the supercell parameter, i.e. about twice the chemical order length. Using ultrafast pump-probe experiment we demonstrate that these additional orders give rise to a new folded coherent acoustic phonon mode at 0.7 THz, in addition to that at 1.2 THz due solely to the chemical modulation [12]. Our work therefore demonstrates that playing with chemically periodic SLs made of multiferroics offers further artificial (polar and structural) orders for tuning in the future, on demand, the spectrum of quasiparticles such as phonons, magnons or electromagnons in functional materials.

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## Time-domain Brillouin scattering for the evaluation of materials interface inclination: Application to imaging of crystal degradation upon non-hydrostatic compression

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Laser ultrasonics is a very handy technique to characterize, evaluate and image materials remotely under extreme (harmful) conditions, such as high pressures. Time-domain Brillouin scattering (TDBS) is such a technique that uses ultrafast laser pulses to generate and detect coherent acoustic pulses (CAPs) as they propagate through a transparent material for the probe laser wavelength [1-2]. In TDBS, the probe light scattered by the propagating CAPs interferes with the probe light reflected on a stationary interface, which leads to a signal with Brillouin oscillations (BOs). If a CAP is reflected from and transmitted through a material interface, the TDBS signal can also feature an echo. Both BOs and echo features can be used for profilometry of (buried) interfaces [3-5] by using information from measurements performed at different lateral positions while scanning cofocused pump and probe beams. A recently developed theory of this type of non-collinear sound-light interaction [6] also provides the opportunity of interface profilometry without the drawback of lateral averaging, i.e., from a local estimation of the angle using a single measurement. The TDBS technique was here applied to monitor the degradation of a single crystal made of lithium niobate ( $LiNbO_3 - LN$ ) upon non-hydrostatic compression in a diamond anvil cell (DAC). The plate-like sample made of a single crystal of LN was prepared from a nominally 20 µm-thick single-crystal disk (x-cut). The nominal geometrical characteristics of the surfaces of the disk provided by the supplier allowed us to estimate the maximum surface inclination to be 0.6° at sub-µm scale and averaged down to 0.02° at µm-scale. One side of this crystal was then coated with a 70 nm-thick Ti layer which served mostly as the optoacoustic generator in our TDBS measurements. The crystal was then loaded in a DAC with water as pressure-transmitting medium and compressed up to 10.4 GPa, where the liquid water transforms to the phase VII of water ice. The 3D TDBS imaging experiments conducted in the DAC reveal modifications of the thickness profile of the crystal with initially plane-parallel surfaces, as well as of one of its face profiles, caused by the non-hydrostatic compression. The map of the echo arrival times reveals at least two orders increase in the inclination angles. Using our developed analytical theory [6], the inclination angle of the Ti-coated surface of the LN relative to the diamond anvil surface has been estimated by fitting the experimental signals. The evaluation performed from each point of measurement allows to locally assess the inclination, which can be revealed either through the modifications of the BOs in the absence of the detectable CAPs, or through the detection of the CAP echoes arrivals at the interface in the absence of BOs, or simultaneously through both, which further complement the usual use of BOs amplitude and frequency variation. The zoology of TDBS signals gathered in this sample is very rich and will necessitate further theoretical investigations to fully explain all the waveforms by taking into account the optical and elastic anisotropy of LN and water ice, for example. Performing TDBS experiments by analyzing probe polarization would also complement our current analysis.

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#### Grazia Raciti

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In macroscopic solids, thermal transport is generally well described by Fourier diffusion theory. However, Fourier's law may break down for a variety of low dimensional materials such as graphene and hexagonal boron nitride. In these materials, the normal phonon scattering is dominant over resistive scattering and, therefore, phonons can flow without any thermal resistance, as in the case of wave propagation in fluids [1]. Because of this similarity, phonon transport is said to occur in the hydrodynamic regime, in which a thermal impulse can propagate within the material as a wave without any significant damping. This phenomenon is known as second sound and was initially observed at cryogenic conditions in solid He [2], NaF [3], Bi [4], and Al<sub>2</sub>O<sub>3</sub> [5]. Recently, the observation of second sound in graphite above 100 K [6], as well as over 200 K [7] has renewed the interest in this field.

However, the experimental observation of second sound in 2D materials is extremely challenging. Ultrafast time-resolved techniques are well-suited to measure the thermal response of a material after impulsive excitation and, thus, detect this elusive hydrodynamic heat transport. In our work, we use a pump-probe setup, which consists of an ultrafast pump laser pulse (< 30 fs), which brings the system out of equilibrium. Then, a second less intense pulse, the probe, monitors the sample response after pump excitation as a function of delay time between the pump and the probe.

Our versatile pump-probe setup, can perform either time-resolved Raman spectroscopy (TRRS) or transient reflectivity (TR) measurements, when monitoring either Raman spectra or change in reflectivity, respectively. In the TRRS scheme, we employ a pulse shaper to stretch out the probe pulse duration, and therefore, achieve higher spectral resolution ( $\approx 10 \text{ cm}^{-1}$ ). A triple spectrometer then collects Raman spectra as a function of delay time, which allows to track the temperature evolution of single phonon modes. In addition, we developed a TR setup that allows us to further study the different energy flow mechanisms in materials.

We have studied the dynamic evolution of phonons in intrinsic semiconductors such as silicon. We observed an important dynamic asymmetry in the shape of the Stokes Raman phonon response, originating from the Fano interference between the optical phonon and the hole continuum population [8]. Moreover, TR measurements provide valuable information on electron-electron, electron-phonon and phonon-phonon interactions. This shows that combining TRRS and TR provides a deeper understanding of the carrier excitation and energy dissipation in solid state material, which is a key to investigate the new properties of the hydrodynamic heat transport regime.

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Few-layer van der Waals (vdW) materials have been extensively investigated in terms of their exceptional electronic, optoelectronic, optical, and thermal properties. However, the impact of nanoconfinement on their mechanical properties remains controversial in the scientific community. Due to the small lateral sizes of samples, and the limitations of experimental approaches, a complete evaluation of their mechanical properties is an undeniable challenge. In particular, there is no systematic experimental study on whether the elastic constants change when reducing the material thickness, with respect to the bulk, and to which extent. In this work, we employ micro-Brillouin light scattering to investigate the anisotropic elastic properties of singlecrystal free-standing 2H-MoSe<sub>2</sub> as a function of thickness, down to the two molecular layers. We report the socalled elastic size effect, i.e., significant and systematic elastic softening of the material with decreasing numbers of layers. Besides, we show that our approach allows for a complete mechanical examination of fewlayer membranes, i.e., their elasticity, residual stress, and thickness. In perspective, this method can be applied to other few-layer vdW materials for their mechanical evaluation. [1] Our findings are highly relevant for related research fields such as nanoscale thermal transport, electronics, or resonators employing vdW materials. Furthermore, the reported nanoscale softening has profound implications in designing and developing nanodevices, where mechanical properties are essential for their durability and robust performance.

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Chirality is the structural property wherein the object is different from its mirror image. Moreover, not only physical objects such as crystals and molecules have chirality but also dynamic motions such as phonons. Recently, a chiral phonon with pseudo-angular (PAM) and angular momenta was proposed [1] and observed [2]. However, chiral phonons that propagate while rotating in three-dimensional material have not been reported. Here, we used circularly polarized Raman spectroscopy and first-principles calculations to identify chiral phonons in chiral bulk crystals (trigonal  $\alpha$ -HgS [3] and Te [4]). In circularly polarized Raman spectroscopy, the peak split of the  $\Gamma_3$  mode was detected along the principal axis of  $\alpha$ -HgS in the backscattering configuration. On the anti-Stokes spectra [Fig. 1(a)], the splitting of observed  $\Gamma_3$  modes are mirror images of those on the Stokes spectra [Fig. 1(b)]. In addition, peak splitting occurs when the PAM of the incident and scattered circularly polarized light are reversed. From the calculation of phonon PAM [Fig. 1(c)], we verified the conservation law of PAM in Raman scattering by taking into account the three-fold rotational symmetry. This approach enabled us to determine the chirality of a crystal in a noncontact and nondestructive manner. We also evaluated the chirality of the phonons using a measure with symmetry similar to that of an electric toroidal monopole.

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**Figure 1:** (a) Anti-Stokes and (b) Stokes spectra of a  $\Gamma_3$  mode in  $\alpha$ -HgS. The blue and red lines denote the  $\Gamma_3$  mode that were experimentally observed with RL- and LR-polarized configurations, respectively. (c) Phonon PAM in the phonon dispersion from to A point.

## Electrochemical Modulation of Resonance Raman Intensities in Quantum Dots

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Resonance Raman intensities are highly dependent on electron-phonon coupling strength and excited state dynamics[1], which are thus promising to be used for probing the electrically induced effects on QDs. In a previous study, we investigated the free carrier charging effects on ZnO QDs by Raman spectroscopy. Upon injection of electrons, a decrease in overtone to fundamental Raman intensity ratios is observed, which is explained by the reduction of electron-phonon coupling strength due to screening or band filling effect[2]. In a more recent study, we further employ Raman spectroscopy to study the effect of electrochemical potential on CdSe QDs[3]. A significant, reversible change in resonance Raman intensities of QDs is observed under ionic liquid-assisted electrochemical modulation (Figure 1a). The modulation amplitude of Raman intensities is found highly dependent on excitation intensity and becomes larger when increasing the fraction of biexcitons to monoexcitons (Figure 1b). Meanwhile, the overtone to fundamental Raman intensity ratios are almost unchanged. Accordingly, we propose a mechanism to associate the Raman intensity changes with the excited state decay dynamics. The proposed mechanism is further supported by a theoretical calculation, attributing the enhanced/weakened Raman intensity to lengthening/shortening of the excited state lifetime. The results of our studies highlight that the electrically induced effects on exciton-phonon interaction and exciton state dynamics of QDs should be considered in understanding the phonon-involved physical processes of QDs and QD-based devices.

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#### Figures



**Figure 1:** Electrochemical modulation of resonance Raman intensities in CdSe QDs[3]. (a) Raman spectra of QD-ionic liquid device at representative potentials. (b) Evolution of resonance Raman intensities during a sweep of bias voltages.

# Environmental effects on the inelastic light scattering efficiency by acoustic vibration modes of a single nanocube

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The measurement of acoustic vibrational modes is fundamental to the full comprehension of the mechanical properties of nano-objects, since they are related to the intrinsic characteristics of the material, such as crystallinity, size, shape, and elasticity.[1] Furthermore, it has been shown that modifications in local environment can induce the appearance of new features in the inelastic light scattering spectra of nanoparticles that do not follow the usual selection rules.[2,3] In this sense, optical spectroscopy techniques represent a powerful tool to study the vibrational modes of nano-objects and address both their intrinsic properties and their interaction with light, which plays a key role for applications in nanophotonics and optomechanics.

In this contribution, we report low-frequency inelastic light scattering measurements on single gold nanocrystals, combined with detailed characterizations of their morphology and environment by transmission electron microscopy and tomography, and numerical simulations of their optical and vibrational responses. We demonstrate that low-frequency Raman spectroscopy of single nanoparticles, a simple to implement and non-destructive technique, can be used to study the coupling between light and vibration at the nanoscale, by monitoring the changes in the internal electric field of the nanoparticle. We show that shape anisotropy and electric field inhomogeneities within a particle, induced by a local change in the particle environment, lead to the appearance of inelastic light scattering by acoustic modes that are not predicted by usual Raman selection rules, which are based on the (here invalid) assumption of a symmetric internal electric field.

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Figu	res

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Using polarisation selective spontaneous Raman scattering, we have investigated the 5d transition metal compound K<sub>2</sub>ReCl<sub>6</sub> which displays a series of structural phase transitions. We observe a violation of the Raman selection rules in the cubic high temperature phase as well as a low-energy scattering continuum persistent throughout the investigated temperature range from 300 down to 5 K. The continuum couples to one of the phonon modes at temperatures above the lowest structural phase transition at 76 K. We propose a common origin of these observations caused by disorder in the orientation of the ReCl<sub>6</sub> octahedra which locally breaks the long-range cubic symmetry. Consistent results from the related non-magnetic compound K<sub>2</sub>SnCl<sub>6</sub> support this interpretation.

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#### Figures



**Figure 1:** Raman spectra of  $K_2 \text{ReCl}_6$  in two different polarisation geometries. Yellow areas indicate the modes that are forbidden by selection rules in the respective geometry. The inset shows a blow-up of the energy region containing the Fano-shaped phonon that couples to the continuum.



**Figure 2:** Temperature dependence of the low-energy part of the Raman spectrum in (xy) polarisation geometry. The scattering continuum peaking at 2.5 meV is visible for all temperatures.

# Thermal relaxation and phonon lifetime in a nanophononic SiN suspended membrane

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Nanophononic materials are at the forefront of current research for thermal management applications.

These are periodic nanostructures, able to generate constructive and destructive phonon interferences that profoundly change phonon dynamics. The periodicity imposes a new Brillouin zone, whose range in wavevector is much shorter than the one "intrinsic" to the material and related to interatomic distances. As such,

phonon dispersions and velocities are strongly modified, leading to an effective thermal conductivity reduction<sup>1-2</sup>.

It is however not clear the role of periodicity on phonon attenuation: boundary scattering depends only on interface density and roughness. The question remains whether another attenuation contribution could arise, related to the coherent interference of the phonons reflected by periodic interfaces.

In this work, we have used the extreme ultraviolet transient grating technique (EUV-TG)<sup>3</sup> at the free electron laser FERMI (Trieste, Italy) to measure for the first time phonon frequencies and lifetimes in a nanoporous phononic membrane at wavelengths between 50 and 100 nm, comparable to the nanostructure lengthscale.

We show that phonon lifetime is strongly reduced and exhibits an opposite frequency dependence with respect to a uniform membrane. Supported by finite element simulations, we interpret this behaviour as the consequence of a coherent mechanism, which dominates phonon lifetime at these wavelengths.

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#### Figures



**Figure 1:** EUV TG spectra collected on a uniform (left column) and a nanostructured (right column) SiN membrane (blue). the individual panels. Red lines are best fit. The phono wavelength (equal to the TG period) is indicated.



### Thermal Characterization of Nanostructured Model Materials Through FDTR Broadband Response

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Nowadays, it is still a challenge to make thermoelectric materials more efficient in energy conversion. For that, nanostructures (superlattices, nanowires, thin films and nanocomposites materials) are developed to improve overall efficiency due to their ability to tune material properties by reducing their size and dimensionality. In this context, crystalline semi-conductors with embedded crystalline nano-inclusions are good model materials [1], since nano-inclusions, depending on their sizes and distribution, are expected to increase phonon scattering (see figure 1). This reduces the lattice thermal conductivity, whereas the crystalline nature of the material would in principle let the electrical properties unaffected.

In this work, we have studied the thermal properties of an innovative nano-inclusions embedded semiconductor, which consists of a crystalline germanium matrix with embedded crystalline spherical Ge<sub>3</sub>Mn<sub>5</sub> nano-inclusions. For that, we have developed a broadband spectral thermal sensor using Frequency-Domain ThermoReflectance measurement (FDTR). This setup allows to measure a wide thermal response of materials with high bandwidth up to 200MHz. The thermal measurement of GeMn showed the effect of inclusions concentration, which strongly modify the films thermal properties. The thermal characterization revealed strong reductions in the thermal conductivity by a factor of 10 for GeMn thin films with different Mn concentrations, compared to the Ge bulk at room temperature.

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*Figure 1*: Thermal phonons transition in a natural semiconductor crystal (A) and in a pure crystal with a tailored distribution of nano-inclusions (B).

# Electron-phonon coupling and ultrafast dynamics of hot carriers in semiconductors: from interpretation of photoemission experiments to transport simulations in devices.

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Electron-phonon coupling determines the charge transport properties of materials as well as the relaxation dynamics of photoexcited carriers. Computational methods based on density functional theory, on the one hand, and time-energy- and momentum- resolved spectroscopy, on the other hand, allow today an unprecedently detailed insight into the role of the electron-phonon coupling [1]. At the same time, hot carriers start to attract attention in the context of emerging concepts for energy conversion.

In this work, we will present the theoretical and experimental results for relaxation dynamics of photoexcited electrons in semiconductors [2-5]. Our computational method based on density functional theory (DFT) and on interpolation of the electron-phonon matrix elements in Wannier space allowed to successfully interpret the dynamics of photoexcited electron relaxation in GaAs, Si, Ge and InSe, in good agreement with two-photon photoemission experiments [2-5]. In particular, recently we have studied, by photoemission spectroscopy and by ab initio calculations, the influence of the two-dimensional (2D) electron gas, created on the InSe surface by Cs deposition, on the relaxation dynamics of photoexcited electrons. We have shown that the remote coupling with the bulk phonons persists even in the case of complete thermalization of the photoexcited electrons with the 2D gas, and that the coupling of electrons to polar optical phonons is not completely screened even at high Cs concentrations [5].

More generally, we will discuss the necessity of time-resolved simulations as well as the problematics related to the coupling of *ab initio* data with device-oriented simulation methods. Indeed, the search for new functional thermoelectric and/or photovoltaic materials requires device-oriented simulation methods of carrier transport, such as stochastic Monte Carlo method, as well as *ad hoc* information on the strength and relative importance of the various scattering channels, which can only be obtained from *ab initio* calculations [6].

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# Oscillatory Lévy walk of quasi-ballistic phonons in nanowires induced by surface roughness

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#### Abstract

The mean free path (MFP,  $\Lambda$ ) of phonons plays a key role in determining the thermal properties of dielectric materials. For quasi-ballistic phonons, thermal diffusion can be described by Lévy statistics [1] with the probability density function  $\Psi(\Lambda) \sim \Lambda^{-\gamma-1}$ , where the Lévy index  $\gamma$  defines the different diffusion transport regimes: ballistic, superdiffusive, and diffusive. In our study, we investigate the MFP distribution of phonons in a square nanowire using spectral Monte Carlo simulations. We show that the Lévy index is controlled by the ratio of the wall surface roughness ( $\sigma_s$ ) to the thermal phonon wavelength ( $\lambda$ ). The behaviour of the Lévy index as a function of  $\sigma_s / \lambda$  is established as shown in Fig. 1 (a), which indicates a smooth transition between the transport regimes. For  $1 < \gamma < 1.8$ , an unconventional superdiffusive regime is put to evidence for the first time that is characterized by sawtooth oscillations in the  $\Psi(\Lambda)$  distribution as shown in Fig. 1 (b) [2]. Further investigations of the angular distribution acquired by phonons during transport were conducted, and salient features are demonstrated, like a distinct "bunching" or spreading of phonons around the long axis of the nanowire depending on the thermal wavelength and the scale of the surface roughness. These results match many features observed in real nanowires, showing the critical role of quasi-ballistic phonons.

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**Figure 1: (a)** Lévy index obtained by fitting the first peaks of the  $\Psi(\Lambda)$  distribution. **(b)** The unconventional super diffusive regime is characterized by sawtooth oscillations in the probability density function when  $0.01 < \sigma_s / \lambda < 0.1$ .

### Thermal Conductivity of c-plane GaN Membranes Studied by One- and Two-Laser Raman Thermometry

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Semiconductor devices, like light-emitting diodes or laser structures, commonly suffer from substantial heating during operation, which can promote the formation of structural defects. This can significantly limit the lifetime or performance of these devices, which is especially relevant for devices on the mirco- and nanoscale [1]. Understanding the dissipation of thermal energy from the active region is therefore vital for any future optimization. We study the in-plane thermal conductivity  $\kappa$  of GaN-based membranes grown on GaN substrates (low dislocation density of  $5 \cdot 10^8$  cm<sup>-2</sup>) by means of Raman thermometry. For this, the temperature of the sample is probed by the shift and broadening of a Raman mode under heating with a UV laser providing above bandgap excitation. Two different methods are employed: For One-Laser Raman Thermometry (1LRT), the UV heating laser also serves as temperature probe and measurements take place at the heat spot (resonant Raman spectroscopy). In Two-Laser Raman Thermometry (2LRT) [2], a 532 nm Raman laser is scanned across the sample yielding a temperature map based on non-resonant Raman spectra. An exemplary map can be seen in figure 1 (a) below. In this contribution, we will compare these two measurement techniques that differ regarding their respective temperature probe volumes, which are about 0.13 μm<sup>3</sup> (1LRT) and 1300 μm<sup>3</sup> (2LRT). Further, by varying the membrane underetching process, we tune the bottom-facet roughness and membrane porosity. Comparing the κ of different membranes therefore lets us asses the influence of surface roughness and porosity on the thermal transport. Finally, we show the possibility to employ 2LRT in the future to analyze the phononic properties of porous and structured membranes. This could pave the way for the preparation of materials for phonon frequency filtering to locally mitigate temperature spikes in optical and electronical devices.

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**Figure 1:** (a) 2LRT temperature map based on the FWHM of the  $E_2^{high}$  mode of a 850nm-thick GaN membrane with an absorbed heating laser power of  $P_{abs}$  = 19 mW. In the bottom left corner a hole in the membrane used in the etching process can be seen. (b) Temperature curve along the marked path in (a), the exponential decay observed here can be used to extract  $\kappa$  from the data.

### Figures



# High-speed modulation of a THz quantum cascade laser using coherent phonon pulses

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We used laser generated coherent acoustic phonon (strain) pulses to modulate the electronic transport and THz emission of a 2.6 THz Ga(AI)As quantum cascade laser (QCL). The modulation amplitude is of the order 10% and the rise time, limited by the measurement system response, is less than 1 nanosecond.

The GaAs/AlGaAs QCL structure, designed to emit at about 2.6 THz, consisted of 88 repeat periods of the injector and active regions. The total thickness of the structure was about 14 microns. A QCL ridge 2mm-long by 150  $\mu$ m-wide was formed by etching and electrical contacts made to the top and back. The GaAs substrate was thinned to 150  $\mu$ m and polished, and a 100 nm aluminium thin film acoustic transducer deposited opposite the QCL ridge. The device was mounted in an optical cryostat, cooled to a temperature in the range 10 – 20K and was electrically pumped by a constant current of up to 1.8A.

Acoustic strain pulses were generated by exciting the aluminium transducer with ~100 fs-duration pulses from an amplified Ti:Sapphire laser, wavelength = 800 nm; repetition rate 1 kHz; and average power in the range 1 - 10 mW. The generated acoustic pulse propagated across the substrate and entered the QCL stack, travelling vertically up through the structure to the top contact, whereupon it was reflected and travelled back down through the QCL structure.

As they propagate through the device, the strain pulses cause transient changes to the device bandstructure, via the deformation potential electron phonon interaction. This modulates the QCL by changing the injection of electrons into the active region. The theoretical limit to the speed is the time it takes for the acoustic wave, travelling at the speed of sound, to travel through the resonant tunneling region, which is ~100s of GHz. The acoustic pulse-induced transient changes in the voltage across the QCL,  $\Delta V(t)$ , were extracted by using a bias tee in the DC pumping line and changes in the intensity of the QCL THz emission,  $\Delta L(t)$ , detected using a THz Schottky diode [1].

Using a new theoretical model for calculating the phonon modes in an arbitrary heterostructure profile, we determine the envelope of the acoustic deformation potential in the QCL and analyse the electronic transport. We find that acoustic modes up to ~200 GHz are capable of significantly perturbing the QCL transport as observed experimentally.

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Figures



**Figure 1:** (a) experimental arrangement; (b) V(t) and L(t) signals



# Generation and modulation of phononic signals based on a slotted phonon waveguide

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In the quest for phononic circuits for possible information processing [1], phonon sources, waveguides and modulation schemes play key roles. Laboratory demonstrators have been realised in silicon-on-insulator (SOI) by electron beam lithography and reactive ion etching [2, 3].

Several types of phonon sources have been investigated [4] and here we explore the generation of phonons by means of a 2D SOI phononic crystal membrane cavity with a guided mode around 6.8 GHz. By incorporating an air-slot flanked by phononic crystal mirrors, we turn the phononic waveguide into an optomechanical platform that exploits photonic modes localised by inherent fabrication variations for the transduction of mechanical modes [5]. Such a platform exhibits fine control of phonons using light and is capable of coherent self-sustained phonon generation via mechanical lasing around 6.8 GHz [6].

While the transmission of signals is essential for information processing by phonons, modulation is a necessary condition. The concept and validation presented here is of a mechanical-optical-mechanical (MOM) configuration and is based on the same structure described in [7] and shown in figure 1, where a combination of both, modulation of the MHz phonon lasing mode and the GHz phonon lasing mode is realised to produce a frequency comb based on a bimodal phonon laser. We show that, in addition to a rich set of dynamic regimes, the design allows to control the modulation frequency and the spectral distribution of the frequency comb.

The slotted waveguide platform operates at room temperature and is fully compatible with silicon technology.

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#### Figures

**Figure 1:** A phonon source based on a slotted waveguide. (a-top) SEM image of the optomechanical cavity waveguide and the central air-slot region. (a-bottom left) Magnified SEM image of the center slot highlighted above in red. (a-bottom right) The electromagnetic energy density. (b) Mechanical mode probed with a tapered fiber loop. Inset: magnified spectrum of the same mode. (c) Radio-frequency spectrum of transmitted light showing the mechanical mode. Dynamical back-action is observed leading to mechanical lasing. Colour scale indicates laser power in mW. Inset: driven optical mode exhibits a thermo-optic shift with increasing power.



**Figure 2:** A frequency comb based on a mechanical-optical-mechanical configuration (MOM). Left: Input-output schematic of a mechanical-optical-mechanical multimodal configuration, where two mechanical modes (M1 in red and M2 in green) couple to a waveguide-coupled single optical mode via their respective optomechanical coupling rates. The optical mode is highlighted in grey. Right: (a) Power spectral density of the output optical field in the simultaneous lasing regime measured over the 0-13.5 GHz frequency range. (b) Up to 10 pairs of sidebands on each side of  $\Omega_2$  are observed, while the second harmonic of this pattern (centred at 22 GHz) is partially visible near the signal analyser limit.





### Neuromorphic computing with coherent acoustic phonons

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Nowadays, acoustic phonons play a crucial role in modern information technologies and are gaining niche in quantum communications. The present work reveals the potential of acoustic phonons in the emerging field of physical neuromorphic computing [1]. For this aim, we develop and test a neuromorphic reservoir based on phonon-magnon interaction. In analogy with the cortea in the human eye, which converts the visual signal into electrical impulses for the following recognition by the brain, our device converts graphic shapes drawn by the laser beam on the surface into the propagating phonon wavepacket, which mixture with magnons helps to recognize the drawn shapes by a simple artificial neural network (ANN).

The developed hybrid structure consists of a semiconductor GaAs/AlAs superlattice, which serves as a phonon waveguide, and a metallic ferromagnetic Fe<sub>0.81</sub>Ga<sub>0.19</sub>-layer hosting magnons. The femtosecond pump pulses (1050 nm, 80 MHz, 150 fs, 3-µm spot diameter) hit the nanograting patterned on the metallic surface and generate coherent phonons with the in-plane wavevector. Among surface waves, it includes a multimode phonon wavepacket guided by the superlattice along the surface on distances up to 100  $\mu$ m [2]. The wavepacket consists of 22 phonon modes with the frequencies in the 16 – 18 GHz range. During propagation, the phonon wavepacket interacts with magnon modes of the ferromagnetic layer. The probe pulses read the phonon-driven precession of the net magnetic moment exploiting the transient polar Kerr rotation [3]. The measured complex waveform is a phase-sensitive superposition of the phonon and magnon modes, which have individual frequencies, spatial shapes, phases, and velocities. As a result, the readout signal is extremely sensitive to the relative position of the pump and probe laser spots. Their relative shift in any direction on a distance much smaller than the pump and probe spot diameters results in noticeable modification of the readout waveform. Such a sensitivity enables efficient selection a "graphic shape" drawn by sequential positioning of the pump beam on the surface and conversion it to the magnon readout for the following recognition by a readout ANN. After a short training process, ANN is able to confidently recognize the randomized and noisy shapes drawn in the area comparable with a single pixel of a digital camera.

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**Figure 1.** (a) Several readout signals (filtered, bandpass filter 15-20 GHz) measured at the distance of 22 µm between the pump and probe spots along the reciprocal wave vector of the nanograting. Their relative position is slightly varied along and perpendicular to the propagation direction ( $\Delta x$ ,  $\Delta y$ , respectively). (b) Readout signal corresponding to letter "L" drawn by the pump laser on the surface: upper curve - as measured and recognized by ANN; lower curve – filtered by the bandpass filter 15-20 GHz. The inset illustrates the simulated spatial distribution of the pump laser intensity on the surface with randomized positioning error.



# Design of optophononic devices based on SiO<sub>2</sub> and TiO<sub>2</sub> mesoporous materials for sensing applications

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Ultrahigh-frequency acoustic-phonon resonators usually require atomically flat interfaces to avoid phonon scattering and dephasing, [1] leading to expensive fabrication processes, such as molecular beam epitaxy, and are bound to fixed frequency operations. Mesoporous materials, with pores at the nanoscale, are good candidates to overcome these limitations. The high surface-to-volume ratio and tailorable mesopores allow the incorporation of chemical functionalization to nanoacoustics. Here, we present multilayered resonators based on mesoporous  $SiO_2$  and  $TiO_2$  (Fig. 1(a)) with acoustic resonances in the 5-100 GHz range. [2,3] We characterize the acoustic response using coherent phonon generation and detection experiments. [4] Fig. 1(b) show experimental results of phononic modes in structures with different mesoporous layer thicknesses. Modes up to 60 GHz are confined within the mesoporous layer, as shown in Fig 1(c). Under liquid infiltration the mesoporous materials are known to present changes in the mechanical properties, and therefore, are suitable for sensing applications. [5] However, the mesoporous layer has to have access to the environment and thus be the outermost layer in a structure. We designed new structures based on surface mesoporous spacer cavity on top of an acoustic distributed Bragg reflector to confine phonons at 100 GHz (inset of Fig. 1(d)). Simulations show strong coherent-phonon signals at the designed frequency (Fig. 1(d)). Our findings unveil a promising platform for nanoacoustic sensing and reconfigurable optoacoustic nanodevices based on soft, inexpensive fabrication methods.

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- Figures



**Figure 1:** (a) Schematic of the samples. (b) Measured spectra of the surface displacement of  $TiO_2$ -based samples A, B and Control, and  $SiO_2$ -based. The low-frequency-shaded areas (up to 50 GHz) indicate the first confined modes in the mesoporous layer. (c) Displacement field of the modes at 13, 25.5 and 38 GHz on TiO2 sample B, respectively indicated as black, red and blue circles in (b). (d) Simulations of the pump-probe spectra for the proposed structure. Inset shows the layer configuration of the device.

### Phonons 2023



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#### Abstract

It is conventionally thought that around and above room temperature the lattice thermal conductivity ( $\kappa_{ph}$ ) in metals is negligible. Recently anomalously weak anharmonic scattering has been revealed in certain metals including tungsten [1] and transition metal carbides (TMCs)[2,3], leading to large and nearly temperature-independent  $\kappa_{ph}$ . These behaviours have the origin in either the crystal structure[4] or the electronic structure[3]. From the relation between the phonon transport and the electronic structure in the TMCs we identified a high thermal conductivity material  $\theta$ -TaN[5] with a room temperature value of ~1000W/m-K. We have also quantified the lattice and electronic thermal conductivities of arsenic, We find that the  $\kappa_{ph}$  of As exhibits strong anisotropy, and the calculated room temperature  $\kappa_{ph}$  reaches as high as 19 W/m-K along the binary direction, larger than those reported for most of other elemental metals.

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#### Figures

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Heat dissipation has become a critical problem in the performance of electronic devices, thus, reducing their lifespans [1]. Heat is transported by phonons, which are the quanta of lattice vibrations. However, phonon manipulation hasn't been easy and remains challenging till today [2]. Therefore, to manipulate and control phonons, electronic analogues like thermal diodes, transistors and thermal logic gates are needed. A thermal diode works as a rectifier allowing flow of heat preferentially in one direction, thus allowing excess heat to escape to the environment, while insulating from heat coming from the environment [4]. In our current research, we have studied telescopic nanowires for their thermal rectification capabilities [3].

A telescopic nanowire is a quasi-one-dimensional structure with a thick and a thin part. The diameters are 300 and 90 nm for thick and thin part respectively, with a transition region in between. The nanowires are grown by MBE technique using VLS mechanism [4]. Suspended platforms [5], with Joule heaters and thermometers, are used to measure thermal conductance properties of the nanowires in forward and reverse bias conditions (figure 1a). Thermal rectification ratios of up to 8% were measured as a function of applied temperature bias (figure 1b). The effect of the mechanical contact resistance on rectification is studied by depositing metal at the contacts on the suspended nanowire. Further, the temperature jump at the contacts is extracted using Raman thermometry while applying different temperature bias. This is the first experimental study on telescopic nanowires indicating rectification and an important contribution towards the development of thermal circuit elements.

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**Figure 1:** (a) Top view of a telescopic nanowire on a suspended platform device (b) Rectification factor as a function of applied thermal bias for several base temperatures  $T_b$ 

#### Figures


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We propose a general heat conduction formalism based on theoretical arguments and direct atomic simulations, which bridges conventional phonon gas model and the wave picture of thermal phonons.

By naturally introducing wavepackets in the fundamental heat flux expression [1], we derive an original thermal conductivity formula where coherence times and life-times appear [2]. We apply the theory to a complex crystal where interatomic potentials are optimized by a Machine Learning procedure (see Figure 1). The simulation reveals an intrinsic and a -previously investigated- mutual coherence appearing in two different temperature ranges.

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**Figure 1:** Time evolution of the phonon number arising on a high-frequency TI3VSe4 mode as a function of temporal coherence time. Average coherence time increases with temperature.

## Extreme light control mediated by nanoscale phonon-photon interactions

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In this talk, I discuss our recent efforts in the context of nano-optics and nanoscale photonics, with a special emphasis on strong light-matter interactions enabled by phonon resonances in polaritonic materials and metasurfaces. I will discuss our recent results in the context of hyperbolic phonon polariton manipulation, focusing, canalization and extreme dispersion engineering in polaritonic metasurfaces. During the talk, we will discuss the exotic light-matter interactions arising by strongly coupling phonon and optical resonances, stemming from the combination of material and nanophotonic engineering, and the various opportunities that these systems enable for wave physics and photonics technology.

# Diffusive-to-ballistic transition and second sound in phonon-mediated thermal transport on micro/nano scale

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Non-diffusive heat conduction by phonons has traditionally been studied at low temperatures, where ballistic transport and second sound propagation occur over macroscopic distances. More recently, the focus of the scientific community has shifted towards heat transport on the micro/nano scale where non-diffusive phenomena can be observed even at room temperature. While in the heat pulse experiments of the last century, ballistic pulses and second sound waves were directly observed to travel from point A to point B, modern studies increasingly rely on indirect methods combined with complicated modelling, with physical effects not immediately discernible in the data. In this talk, we will review recent experimental work performed with a technique approaching the heat pulse method in terms of allowing direct observation of the phenomena of interest, namely, the transient thermal grating (TG) technique, in which two short laser pulses crossed in the sample produce a spatially sinusoidal temperature profile whose decay is monitored via diffraction of a probe laser beam. While in the past the shortest TG half-pitch, which sets the heat transfer distance, was limited by the optical wavelength, the recent progress in coherent extreme ultra-violet (EUV) and x-ray sources such as free electron lasers makes it possible to access deep nanometer scale. As a case in point we will discuss diffusive-to-ballistic transition in the room temperature heat transport in silicon. Owing to the wide spectrum of the mean free paths of heat-carrying phonons, this transition is extremely gradual: while significant deviations from the diffusive regime are seen already on the micron scale, the heat propagation at ballistic velocities is only observed at distances of a few tens of nanometers. Second sound is another peculiar phenomenon encountered in some materials on the way from diffusive to ballistic transport. We will discuss recent observations of second sound in graphite at temperatures up to 200 K as well as reports of room temperature phonon hydrodynamics and second sound in other materials such as silicon and germanium.

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#### Abstract

Phonons were traditionally considered as linearly polarized. Recently we predicted that phonons can be chiral and have angular momentum both in magnetic [1] and nonmagnetic systems [2]. The nondegenerate chiral phonons are also predicted and easily tuned in graphene/hexagonal Boron Nitride heterostructures[3]. The chiral phonons were observed in monolayer tungsten diselenide, where the phonon chirality is confirmed by the infrared circular dichroism arising from pseudoangular momentum conservation [4]. Our further experiments showed that through the emission of a chiral phonon the momentum-dark intervalley exciton is brightened [5] and chiral phonons can have entanglement with photons [6]. Recently, we predicted that the chiral phonons can propagate along high-symmetry axis of 3D materials [7], and will show a diode effect of chiral phonons in chiral systems [8]. We also observed a chiral-phonon-activated spin-Seebeck effect in chiral materials very recently [9].

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## POSTERS

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Plant-based materials offer a unique engineering platform to develop sustainable and multifunctional biocomposites, due, in part, to their natural abundance and en-masse production, their functional diversity, as well as their self-growing capacity. While these bio-based materials have already started to replace inorganic compounds in many existing technologies, including photonics and plasmonics [1], soft electronics [2], and energy harvesting, their phononics' potential remains less explored. In this work, we study the phononic design and characterization of plant-based materials composed of micro-structured films of cellulose, extracted from decellularized onion cell scaffolds [4]. We use laser ultrasonics setups to generate and detect the propagation of MHz to GHz acoustic waves in these biomaterials. By measuring their interaction with surface acoustic waves (SAWs), we reveal a complex and rich dispersion curve that stems, in part, from their structural features and mechanical anisotropy. In addition, we demonstrate the ability to precisely tune their phononic features via chemical intervention. Our results demonstrate the possibility to use biologically-derived thin films and surfaces to manipulate the propagation of high frequency ultrasonic waves, which we anticipate can enable a new class of green ultrasonic devices with tailorable and reconfigurable properties for diverse applications such as RF filtering, sensing, and waveguiding.

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**Figure 1:** (a) Honeycomb-like structure of decellularized plant cells scaffolds with a superimposed illustration of a propagating surface acoustic wave. (b) MHz phononic spectrum of the plant structure showing the presence of a phononic bandgap due to locally resonant cell walls. Taken from Ref. [4]

Figures

## Electrothermal measurements of metal-semiconductor structures by scanning thermal microscopy and $3\omega$ method

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#### Abstract

Heat transfer across the contact between two materials constitutes a research topic of fundamental and technological interest. In particular, metal-semiconductor contacts, or Schottky contacts, are widely present in electronics technology. Understanding the mechanisms of thermal transport, which involves electrons and phonons in the junction region, requires the determination of both thermal conductivities of materials in contact and thermal contact resistance between them; these are key quantities driving various phenomena undergone by energy carriers and leading to the overall sample thermal conductance [1].

We study experimentally Schottky interfaces within layered samples by means of two electrothermal techniques: scanning thermal microscopy (SThM) and 3ω method. SThM is based on a self-heated metallic thermal sensor that is part of a movable AFM tip and enters in contact with the sample surface [2]. This technique allows to perform thermal measurements with a spatial resolution down to the nanometric scale, at sample temperature close to ambient. Figure 1 shows an example of tip temperature map over a sample composed of a silicon substrate on top of wich a titanium and a gold nanolayers were successively deposited. While SThM allows for spatial resolution, it is hardly able to be performed as a function of sample temperature. In contrast, the  $3\omega$  method, which is based on the deposition of a metallic resistive wire on top of the sample and whose electrical resistance is measured as a function of an alternating input power [3], provides access to experiments over a wide temperature range through the use of a cryogenic system, but it does not allow spatially localised measurements. The experimental results are accompanied by a sensitivity analysis and, in the case of the  $3\omega$  method, a comparison to a semi-analytical model [4].

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Figure 1: (a) Tip temperature rise map over a sample that contains three different material structures. The values over each region correspond to the average tip temperature rise. (b) Diagram representing each part of the sample and showing how heat flux conduction inside each of them contributes to cooling the tip.



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# Active mode-locking of a vertical-cavity quantum-cascade saser by pump modulation

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We present experiments on active mode-locking of an electrically pumped vertical-cavity quantum-cascade saser structure by modulation of the cavity gain via the applied pump voltage.

The saser device was based on the structure reported in [1], which consists of a 50-period quantum cascade gain superlattice (SL) in a vertical cavity between two SL phonon mirrors, a 40-period 'high reflector' and a 15-period 'output coupler'. The design frequency of the saser structure was 325 GHz. Phonons emitted into the GaAs substrate via the output coupler were detected on the opposite surface of the substrate using superconducting Al bolometers.

The gain SL was pumped with electrical pulses with voltage amplitude V, which were modulated with a sinewave with peak-to-peak amplitude  $\delta V$  and frequency, f, see Fig. 1. Measurements were made of the temporal dependence of the bolometer signal amplitude as a function of the frequency and amplitude of the modulation. We look for mode-locking of the saser emission, which is evidenced by a train of output phonon pulses observable when f = 1/T, where T is the round-trip time for phonons in the cavity,  $\approx 6.5$  ns.

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#### Figures



**Figure 1:** Schematic diagram of the saser device. The device structure was grown by MBE on a 380 µm-thick semiinsulating GaAs substrate and consisted of a 50-period 'gain' SL, each period consisting of 6 nm of GaAs and 4 nm of AlAs, uniformly n-doped with Si to a density of  $10^{17}$  cm<sup>-3</sup>. Electrical contact to the gain SL was made via 0.5 µm-thick GaAs contact layers, n+-doped with Si to a density  $10^{18}$  cm<sup>-3</sup>. SL acoustic Bragg mirrors were fabricated at both ends of the structure. Each period of the mirrors consisted of 4 nm of GaAs and 4 nm of AlAs, both undoped. The top mirror (high-reflector) has 40 periods, and the bottom mirror (output-coupler) has 15 periods. The total effective cavity length is just over 1.5 µm. The electrical pump pulse has the form of a rectangular voltage pulse, average voltage *V* and overall duration  $\tau$ , modulated by a sinewave of pk-to-pk amplitude  $\delta V$  and frequency *f*. The emitted phonons are detected on the back surface of the substrate using a superconducting Al bolometer. The bolometer signal is amplified and displayed on a high-speed digitizing oscilloscope.



## Towards acoustic microscopy at the nanoscale by coupling atomic force microscopy with picosecond ultrasonics

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Abstract:

Acoustic imaging techniques are powerful, nondestructive tools used to perform elastic imaging of systems. Acoustic imaging techniques have a large variety of applications in both industrial and fundamental research. Acoustic wave generation in the GHz range using picosecond acoustics has enabled the development of elastic images with nanometric in-depth resolution. However, the spatial lateral resolution for this imaging technique is limited by the laser spot size, which cannot be reduced below a couple hundred nanometers due to light diffraction. We report the results from an approach <sup>1-2</sup> in which we couple time-resolved pump-probe spectroscopy and atomic force microscopy. We generate GHz waves directly into an AFM probe from the top of the probe. Acoustic transmission from the probe to the sample is also achieved, which paves the way to perform subsurface imaging. Due to the nanometric radius size of the commercial tip, acoustic imaging with an improved lateral resolution could be achieved. These results are an encouraging step toward the development of a new acoustic microscopy technique.

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#### Figures





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The possibility of creating and manipulating nanostructured materials encouraged the exploration of new strategies to control electromagnetic properties. Among the most intriguing nanostructures are those that respond differently to helical polarization, i.e., exhibit chirality [1,2]. Here, we present a simple structure based on crossed elongated nanobars with the long axis parallel to the polarization plane forming a given angle between them and vertically aligned, as shown in Fig. 1(a). We theoretically demonstrate that, in such structure, light-handedness defines the dominating cross-section absorption or scattering, with a 200% difference from its counterpart (scattering or absorption). The proposed chiral system opens the way to enhanced coherent phonon excitation and detection. In general, the generation of acoustic phonons is optimized by maximizing the absorption, while the detection is enhanced by engineering the scattering properties [3,4]. In chiral structures, this could be achieved by a simple pump-probe experiment using circularly polarized light at the same wavelength. The use of structures sustaining chiral optical modes appears as a natural, yet unexplored strategy to address these conditions. These results constitute one of the first steps towards harvesting chirality effects in the design and optimization of efficient and versatile acoustoplasmonic transducers.

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**Figure 1:** (a) Sketch of the geometrical layout of the chiral structure. Two gold nanobars separated by a distance D = 80 nm, with the long axis parallel to the polarization plane forming a given angle between them and vertically aligned. The bottom bar is embedded in SiO<sub>2</sub> while the top bar sits on the surface of SiO<sub>2</sub>. (b) Absorption and scattering cross-sections for right (RCP) and left (LCP) circularly polarized incident beam.



### Interference of magnons excited by coherent phonons and ultrafast spin torque

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Magnons are quasi-particles that are the result of quantising the spin waves in ferro- and antiferromagnets. Coherent phonons drive magnons in a similar way as electromagnetic waves drive spin excitations [1]. We perform experiments on a 20 nm thick ferromagnetic Fe<sub>0.81</sub>Ga<sub>0.19</sub> film, known as Galfenol, with a nanograting (grooves are 7 nm deep and 48 nm wide; the period is 120 nm) fabricated by focused ion beam on the surface of the Galfenol film. Due to the nature of the milling process the milled areas have a higher Ga concentration than the unmilled areas. We excite the film by femtosecond laser pulse which triggers the excitation of coherent magnons via two mechanisms: (1) coherent Rayleigh phonons with frequency 20.1 GHz excited in the nanograting; (2) ultrafast torque pulse acting on the magnetization vector. As a result, we observe constructive and destructive interference of magnon responses to the joint excitations originating from coherent phonons and ultrafast torque controlled by the same laser pulse.

In the experiments we exploit the ultrafast magnetoacoustic technique [2]. We use two synchronised femtosecond lasers as pump and probe beam sources with wavelengths of 1050 nm and 780 nm respectively. The pump has a maximum fluence of 10 mJ/cm<sup>2</sup>. The ASOPS scheme [3] is used for obtaining temporal signals with 1 ps time resolution. A magnetic field is applied in the plane of the sample at 45° from the normal direction of the grooves in the nanograting. The magnon dynamics is measured by detecting the polarisation rotation (Kerr effect) of the reflected probe beam.

Figure 1(a) shows the measured magnon spectra obtained as fast Fourier transforms from the temporal magnon signals for different magnetic fields. Figure 1(b) shows the same spectra in the form of a colour map. The maximum amplitude of magnons is observed in the vicinity of the magnon-phonon resonance where magnon spectra are asymmetric and possess the features of Fano resonances. The specific features of the measured spectra include the nulling of the magnon amplitude at the low frequency side of the magnon-phonon resonance [marked by a circle in Fig. 1(b)] and the increase of the magnon amplitude for magnon frequencies higher than the magnon-phonon resonance [marked by a rectangle in Fig. 1(b)]. The analysis of the results based on the Landau-Lifshitz-Gilbert equation supports the explanation of the observed spectral features in the magnon spectra as the interference of responses from Rayleigh coherent phonons and ultrafast torque induced by the laser pulse.

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Figures



**Figure 1:** (a) the fast Fourier transform data of magnon signals for different magnetic fields from 200 mT to 400 mT in 10 mT steps. (b) the same data shown as a colour map which better illustrates the destructive and constructive regions of the data.



### Phonon anharmonicity at Earth's lower mantle conditons:

### Consequences on the thermal conductivity of MgO

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MgO is a material important for many technological applications as well as in planetary sciences since it is an archetypal mineral comprising the Earth's lower mantle (LM). Here, we address how the anharmonic effects, due to phonon scatterings, influence phonons' energies and lifetime at the extreme high-temperature high-pressure conditions pertinent to the LM (pressure between 24 and 135GPan temperatures up to 3000K) and their consequences on the lattice thermal conductivity. Where our previous work highlighted the importance of anharmonicity on the lattice dynamics of MgO at ambient conditions, at high-pressure but ambient temperature, and at high temperatures but ambient pressure [1,2], the simultaneous effects of the high-pressure and high temperature were not addressed yet.

In this poster, I will present the results of *ab-initio* calculations of the phonon anharmonicity in two cases: I) ambient pressure and high temperature up to T=1223K, covering the range where calculations can be compared to existing experimental results ii) at Earth's lower mantle conditions with T-P in the range from 1900K-24GPa to 3000K-130GPa. High-temperature effects on the phonon energies and linewidths due to the 3 and 4 phonons scatterings are considered in the framework of perturbation theory. These results are compared with Infrared spectroscopy and Inelastic x-ray scattering measurements carried out by our group. The high-temperature case permits benchmarking the joint DFPT [3] and SSCHA [4] approach in describing the anharmonic lattice dynamics of MgO. Once validated against the experiments, calculations are extended to the lower mantle conditions to discuss the effect of pressure and temperature on the anharmonicity of MgO. Finally, I will show the consequences of the anharmonic terms on the lattice thermal conductivity of MgO at LM conditions in relation to phonons scattering.

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Figures



**Figure 1:** Phonon dispersion of MgO at Earth's lower mantle T-P conditions calculated according to different approximations, Red: Quasi-harmonic (QH), Blue: QH+ 3 phonons scattering contributions, Orange: SSCHA renormalized phonons, Black: combination of the effects of 3 and 4 phonons scattering.



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Disorder can sometimes produce vibrational mode localization as has been shown in previous studies of amorphous Silicon. Two signatures of mode localization are exponential decay of the vibrational eigenvector and participation ratios \$<0.1\$ for eigenmodes above a threshold frequency[1]. Recently, interesting materials called high configurational entropy oxides have been produced which exhibit a mixture of long-range order (oxygen sublattice) and disorder (random cation sublattice). One such material is Mg<sub>0.2</sub>Co<sub>0.2</sub>Ni<sub>0.2</sub>Cu<sub>0.2</sub>Zn<sub>0.2</sub>O (HEO) which, if quenched from 1000°C or above to room temperature, forms a singlephase rocksalt structure. This work addresses the question of whether the cation disorder in HEO is enough to produce localized modes by employing the General Utility Lattice Program (GULP). The first step was to develop new empirical interatomic potentials (EIPs) for the parent binary oxides MgO, CoO, CuO, NiO, and ZnO by fitting to experimental phonon frequencies, dielectric constants, and lattice parameters. The new EIPs were then used in HEO, neglecting cation-cation interactions. Experimental support for the approach is in the agreement between simulated and recently measured infrared and Raman spectra for HEO. It is found that a smaller percentage of modes are localized in HEO than in a cluster of amorphous Si with a similar number of atoms. Thus, mass and potential disorder is not strong enough to induce a large number of localized modes in the rocksalt structure. Additionally, we have explored mode localization in a number of other structures which contain disordered cation sublattices but ordered oxygen sublattices.

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## A Novel Four-Terminal Suspended Device for Nanoscale Thermal Characterization

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The increasing demand for effective thermal management of microelectronic devices and efficient thermoelectric has raised the need for new materials with either extremely high or exceedingly low thermal conductivity, respectively [1]. 2D flakes such as graphene or hexagonal boron nitride (h-BN) exhibit among the highest thermal conductivity in solid state materials. Their small dimension in combination with their interesting charge and heat transport properties make them appealing candidates for heat management for nano-electronics devices. One of the major issues with the integration of 2D materials in industrial semiconductor fabrication processes is the reproducibility of high quality flakes [2]. It is challenging to produce thin homogenous flakes with good mechanical and thermal properties, even more, transferring one flake onto a target remains a critical step that may damage or contaminate the material. To guide these efforts, it is of uttermost importance to characterize the intrinsic properties of the materials with a consistent and reliable method with a high signal-to-noise ratio. To meet these requirements, a novel four-terminal device is proposed which involves fabricating suspended four transducers, which can act as thermometer as well as heater, combined with the suspension of an h-BN flake. The resistive transducers are made of platinum meanders placed on top of under-etched silicon nitride (SiNx) membranes, which are connected to the substrate by SiNx beams. In this way, the sample is suspended and decoupled from the environment while assessing its thermal properties in vacuum-based measurements. During operation, one transducer is electrically heated to generate a heat pulse that raises the temperature on a side of the sample. The remaining transducers may be used as sensors to probe changes in temperature on the other sides of the sample. To address the known issues of microstructure-based thermometry, the device is engineered to enable additional local thermometry using Raman spectroscopy, which is applied to map the temperature distribution of the flakes, so that the thermal conductivity is quantitatively extracted [3]. We demonstrate the capabilities of this novel four-terminal devices through electrical measurements and first results on h-BN flakes.

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**Figure 1:** Suspended hBN flake, four terminal device. The optical micrograph on the right, there is a wide view of the device where the bridged membranes with beams are visible. On the left, there are four suspended membranes of SiNx with an h-BN flake suspended in the middle. The flake is clamped on the side with platinum patches that anchor it to the SiNx layer.

### Phonons 2023



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Surface acoustic waves (SAWs) are used in numerous applications both in technology and in fundamental research [1]. Wide-frequency-band SAWs are especially useful for the evaluation of subsurface inhomogeneity and of on-surface depositions, i.e., for imaging and sensing. The shorter the acoustic wavelength (the larger the frequency), the closer to the material surface the localization of the SAWs, hence the better the sensitivity to inhomogeneities in close vicinity of the surface. Ultrashort laser pulses can generate and detect SAWs at GHz frequencies. SAW pulses with a broad frequency spectrum, approaching 10 GHz frequency bandwidth, were monitored by focusing pump and probe femtosecond laser radiations on the surface of highly-ordered polygraphite to sub-µm size spots [2]. Rayleigh wave packets were all-optically monitored with sub-100 nm width and thickness metallic nanorods, whose acoustic resonances controlled the carrier frequencies at 5 and 10 GHz [3]. Here we report on a new approach for GHz SAW pulses monitoring using nanostructured perovskites. Strontium titanate (SrTiO<sub>3</sub> – STO) is a well-known material with a perovskite crystalline structure. Optically transparent in the visible domain, it was used as a substrate to grow, using pulsed laser deposition, few-tensof-nm-thick layers of materials, which are either transparent ( $Ba_{0,6}Sr_{0,4}TiO_3 - BSTO$ ) or opaque ( $SrRuO_3 - SRO$ ;  $La_{0.7}Sr_{0.3}MnO_3 - LSMO$ ) in the visible domain [4]. SRO and LSMO display optical and acousto-optical properties very different from the STO, while the acoustic impedance mismatch between STO and the thin films remains very low. The opaque films can be used to efficiently generate and detect coherent acoustic pulses (CAPs), which are travelling with negligible reflections at the interfaces between the perovskite materials. For our experiments, cross sectioning of the grown layered samples, with layer thicknesses of few tens to hundreds of nm, has been realized either by focus ion beam or by mechanical polishing (Fig. 1 a) and c), respectively), providing modulation of the material parameters along the surface at deeply sub-optical scale. Experiments revealed the surface propagation of CAPs, with the frequency spectrum of 20-30 GHz bandwidth, between two opaque layers (Pt, SRO) and their reflection at mechanically-free surface (red signals in Fig. 1 b) and d), respectively). The identification of acoustic modes contributing to surface echoes is facilitated by comparison with bulk longitudinal CAPs (LA) detected in the traditional experimental configuration (blue signals in Fig. 1 b) and d), respectively).

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#### Figure



**Figure 1:** a), c): Schematics of the samples with a) two and c) one opaque layers. b), d): Transient optical reflectivity signals measured on the nanostructured surface (upper parts, red) and in the traditional configuration (lower parts, blue). Gray dashed lines indicate the surface propagating echoes which crossed: in b), the BSTO layer once (faster than the bulk LA echo that did it twice (blue signal)); in d), the STO layer twice. LA denotes the arrivals of the longitudinal echoes.



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Modern semiconductor membranes for photonic applications are often characterized by thermal techniques that lack the combination of high spatial resolution and a solid quantification of temperature. Thus, understanding the thermal limitations of such membrane-based devices, like nanobeam lasers (1D) or photonic crystals (2D) is hindered as heat spots, thermal anisotropies, or interface heating phenomena cannot sufficiently be characterized. In this contribution, Raman thermometry employing one laser beam (1LRT) and two laser beams (2LRT) is used to quantify the thermal conductivity κ of 250-nm-thick, state-of-the-art, c-plane photonic GaN membranes as depicted in Fig. 1a and b. The corresponding layer sequence including an AIN interlayer is deposited on silicon to allow a 10-µm-deep under-etch. The 2-nm-thick InGaN quantum well serves as a built-in light source, rendering the structure a photonic membrane used for numerous photonic applications [1]. A top-view SEM image of this membrane is shown in Fig. 1b, including a sketch of the freestanding part of the membrane that will be targeted by the Raman thermometry. A sketch of the fully customized optical setup allowing for 1LRT and 2LRT measurements is shown in Fig. 1c. First, a cw UV laser (266 nm) heats the photonic membrane by over-bandgap excitation and is simultaneously used to probe temperature by resonant Raman spectroscopy (1LRT). Second, an additional cw laser (488 nm) can be scanned over the heat spot of the UV laser to built a temperature map with a spatial resolution of  $\approx 1 \, \mu$ m as shown in Fig. 1d (2LRT). Interestingly, this experimental approach allows a direct access to most photonic structures, without the need of any detrimental substrate removal or additional invasive processing steps. Based on our temperature maps we can directly extract the thermal conductivity k for all in-plane directions and compare our results to state-of-the art DFT calculations. Finally, following the method described in Ref. [2], also the thermomechanical stress induced by the heating laser is computed based on the Raman spectra providing mode shifts and FWHM values. In conclusion, we introduce a novel experimental scheme for Raman thermometry, which is especially suited for photonic structures requiring a spatial resolution on the order of micrometres.

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**Figure 1 (top): a)** Schematic of the MOCVD deposited layers. **b)** Top view SEM image of the whole GaN pad. The scheme on the right helps locating the freestanding part. **c)** Scheme of the optical apparatus needed to perform a mapscan. **d)** Thermal map on the freestanding part of the membrane. The heat laser (325nm, cw) is focused and located  $4\mu$ m away from each edge while the probe laser (532, cw) is scanned over the membrane surface.



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#### Abstract

Accurate prediction of the thermodynamic and transport properties of materials requires an understanding of anharmonicity resulting from phonon-phonon interaction, beyond the quasi-harmonic theory of lattice dynamics. Anharmonic phonon methods seek to address the limitations of the small displacement assumption inherent in the quasi-harmonic approximation that renders it invalid at high temperatures, particularly near a phase transition, or even at low temperatures if the energy profile is shallow. We present a method of computing anharmonic phonons in which the only assumption appears to be the form of the dynamical matrices. Starting from a given crystal geometry and a grid of q-points in the irreducible Brillouin zone, we define a trial dynamical matrices are then optimized by fitting to forces sampled from *ab initio* canonical Langevin or molecular dynamics. Our method gives converged phonons for selected test cases including ferroelectric oxide, SrTiO3, in good agreement with the experiment.

#### Figures



**Figure 1:** Temperature-dependence of phonon dispersion and DOS of cubic SrTiO3, and a zoomed-in region around the antiferrodistortive R-mode

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Robotic missions to extraterrestrial objects in our solar system are nowadays equipped with instruments allowing to explore the geochemistry of the surfaces by e.g. identification of their characteristic vibrational fingerprints. Femtosecond lasers have in recent years been shown to be in principle space qualified, opening up the opportunity to explore the potential of different time-domain techniques as compact, robust alternatives to e.g. Raman spectroscopy. In this contribution the potential of coherent phonon spectroscopy as an emerging in-situ spectroscopic technique to identify solids by their characteristic phonon spectra is discussed based on exemplarily measurements of different planetary materials.

References

Figures

### Surface plasmon-based detection for picosecond ultrasonics in planar geometries

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Picosecond ultrasonics (PU) has been a valuable tool to investigate and, to a certain extent, also control coherent phonons dynamics in various materials and micro-/nanostructures. However, the accompanying changes in optical or geometrical properties are usually small and therefore still challenging to detect. This has resulted in different approaches to tackle this issue and provide improved sensing schemes during the last decades. Since PU is based on all-optical pump-probe spectroscopy the idea to harness plasmonic effects, that have been proven to be a powerful asset in many sensing applications, has also been pursued.

Here, we want to present our results on the combination of asynchronous optical sampling with acoustoplasmonic sensing in planar gold/dielectric layer systems [1] where we explored the potential of this method to access higher order acoustic modes and attempt to unfold the underlying contributions, i.e., changes in the dielectric function's real and imaginary part as well as layer thicknesses, which are caused by the acoustic dynamics. We also highlight open questions and useful aspects in the implementation of this approach.

We achieve a coupling of the optical probe pulse to the surface plasmon by the Kretschmann configuration shown in Fig 1a). Here, gold films are thermally evaporated on semi-cylindrical glass prisms. The necessary angle distribution is achieved by focusing of the optical beam. This implementation requires a slit in front of the detector to select a specific angle distribution at the cost of a slight broadening of the surface plasmon resonance curve shown in Fig. 1b). This configuration was chosen since it does not require a repositioning of the beam for angle-resolved measurements, which we found to be beneficial in our pump-probe measurements.

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#### Figures

**Figure 1:** a) Schematic of the Kretschmann configuration in the detection path of the used pump-probe setup. b) obtained surface plasmon resonance in the static case, i.e., without pump excitation. Adapted from [1].

## Light scattering on acoustic phonons in lead free double perovskite Cs<sub>2</sub>AgBiBr<sub>6</sub> in cubic and tetragonal phases.

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Halide double perovskite  $Cs_2AgBiBr_6$  is a perspective material for photovoltaic and photoelectronic applications as a non-toxic and stable alternative for solution grown lead-based perovskites. Phonons and electron-phonon interaction play important role in its optical properties. In this work we study inelastic light scattering on acoustic phonons in  $Cs_2AgBiBr_6$  perovskite at 78K and 300K.  $Cs_2AgBiBr_6$  possess cubic crystal structure (space group #225) at room temperature and under 120K it undergoes a phase transition to a tetragonal structure (space group #87). Fig. 1 shows Brillouin spectra measured for both phases at the temperatures 78K and 300K. Longitudinal (LA) and transverse (TA) phonons with frequencies at around 23GHz and 11GHz are observed in backscattering geometry along 111 direction at room temperature. Two TA modes are observed in low symmetry tetragonal phase. The spectra measured for variety of facets corresponding to different crystallographic directions allow us to evaluate all 7 elastic constants  $C_{ij}$  for  $Cs_2AgBiBr_6$  in tetragonal phase at 78K. In addition, we evaluate elastic constants  $C_{11}$ ,  $C_{12}$ ,  $C_{44}$  for cubic phase and compare our results with the data, obtained using nanoindentation and contact resonance atomic force microscopy (CR-AFM) techniques obtained by Y. Lun et. Al [1]. In contrast to CR-AFM, Brillouin scattering technique allows us to examine materials non-invasive and in a wide range of temperatures.

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Figures



**Figure 1:** Brillouin scattering spectra measured in backscattering geometry for Cs2AgBiBr6 perovskite at 78K and 300K at [111] facet, excitation is at  $\lambda ex - 542$  nm.

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Pump-probe optical methods made it possible to explore acoustic properties of glasses in the difficult but crucial frequency region reaching the THz frequency range. A universal property of glasses is that a large excess of modes exists, with a maximum density of states near 1 THz, forming the so-called boson peak. The acoustic modes are expected to be strongly affected as their frequency nears the boson peak. Here, we present an experimental study of the onset of this effect in v-SiO<sub>2</sub> as a function of temperature. A coherent hypersonic pulse is photogenerated after absorbing a femtosecond optical pulse (the pump) in a metallic film or an InGaN/GaN quantum well, and the detection of its echoes after propagation through variable-thickness glass layers is performed thanks to a second optical pulse (the probe) delayed in time. We used two setups (figure 1): i) broadband generation with a metallic film and enhanced detection of discrete frequencies using a superlattice consisting of multiple quantum wells, (low temperature) and ii) broadband generation and detection using an InGaN/GaN single quantum well (room temperature). The anticipated transition, from an  $\omega^2$ - to  $\omega^4$ -law for the mean free path of the longitudinal acoustic modes is observed well below 1 THz, along with a negative velocity dispersion.



**Figure 1**: (a) scheme of the pump-probe experiment using GaAs/AlAs superlattices for detection. (b) scheme of the pump-probe experiment using an InGaN/GaN quantum well, with the paths of the acoustic pulses reflected by the GaN/SiO2 interface

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Surface optical modes are commonly used in Surface Plasmon Resonance (SPR) sensor devices. In this technology, target molecules flowing in a fluid sample are captured by molecular probes attached to the surface of a resonant thin film. Gold thin film with a plasmonic surface is usually excited in total internal reflection at a specific angle and wavelength, allowing for optimal surface mode excitation. As the target molecules in the fluid bind with the targets, the refractive index of the surrounding medium near the surface changes, and the resonant angle and wavelength increase. The shift of the resonances scales linearly with the change in the refractive index, providing information about the concentration of the existing target and the probe-target affinity. Thus, the presence of the target can be quantitatively determined in real time by measuring the characteristics of the light reflected from the surface. The typical SPR sensor device has a sensitivity in the µRIU range. With such sensitivity, the detection of nanomolar concentrations can be achieved. However, picomolar concentration detection is necessary for advanced detection applications.

From a theoretical point of view, the sensitivity of the optical technique described above can be significantly increased by moving to the infrared (IR) spectral range, where polar materials can be used instead of gold films and surface phonon polariton (SPhP) modes can be used instead of surface plasmon polariton modes. The SPhP modes can be considered as surface electromagnetic waves resulting from the coupling of electromagnetic modes (photons) with lattice vibration modes (optical phonons) in polar dielectric materials. Relevant research has shown that the lifetime of SPhP modes is several orders of magnitude longer than that of SPP modes. This makes SPhP a promising candidate for enhancing the lifetime of surface-confined electromagnetic energy, contributing to many technological applications in nanophotonics, including optical sensors of viruses, proteins, and macromolecules. In this contribution, we present the results of an ongoing project in which we intend to take advantage of the superior optical properties of SPhP to develop a photothermal IR optical technique for screening the required picomolar concentrations. It is designed to promote the interaction between incident infrared light and SPhP modes and record spectra presenting characteristics indicative of the nature of the examined species. Analysis of the spectra can be performed with classical theories and fast numerical methods to give a quantitative and accurate description of the examined species in record time.

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Using a laser as a local optomechanical detector of superfluid helium-4 surface modes, known as third sound waves, we propose an experiment to probe observer-dependence in detector response. By moving the interaction point along a circular trajectory, the response can be compared with its inertial counterpart. Disagreement between the two is a manifestation of the Unruh effect, predicting that the uniformly accelerated observer will experience a temperature in empty space [1]. Third sound waves propagate in an effective spacetime, wherein the effect of acceleration is enhanced. Incorporating a nonzero ambient temperature, we identify an acceleration-dependent signal in the laser phase, and evaluate a signal-to-noise measure to show that observing this signal is within experimental reach.

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**Figure 1: (a)** Schematic diagram of an experimental setup for optomechanical detection of third sound on a superfluid helium film. The deflector moves the probe laser beam in a circular trajectory corresponding to a uniform acceleration of the observer; **(b)** Signal to noise ratio (SNR) predicted by simulations as a function of the radius of the circular trajectory of the probe, *R*, and the excitation frequency probed by the detector,  $\omega/2\pi$ . Here, *a* is the acceleration of the detector which is rotating at the frequency shown by the black dashed curve in the upper panel.

## Low Frequency Raman Scattering: From Inorganic and Hybrid Materials to Biological Systems

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Halide perovskite materials present extraordinary bulk photovoltaic properties with very high conversion efficiencies when used as absorbers in perovskite solar cells [1]. Low frequency Raman (LFR) spectroscopy is a sensitive probe of temperature and pressure effects on perovskites structure via shifts in the Raman bands frequencies. In this work, LFR results on  $Cs_2Snl_6$  fully inorganic halide perovskites [2,3] will be presented in order to identify the behaviour of Sn-I vibrations and lattice modes vs lowering the temperature and applying high pressure.

Metal organic frameworks (MOFs) are hybrid organic-inorganic porous crystalline materials, some of them demonstrating outstanding  $CO_2$  capture capacity [4]. Raman spectroscopy has been applied to study flexibility of MOF structures as well as binding and dynamics of  $CO_2$  incorporation inside them. The low frequency vibrations in the THz range are of uppermost interest since these vibrations become very sensitive to gas adsorption and to variations of temperature and gas pressure. Recently obtained LFR results in ZIF-68 and Zif-69 [5] as well as on Kaust-7 MOFs will be presented.

Biological systems, such as tissues, are extensively examined by Raman spectroscopy in the conventional fingerprint, 800-1800 cm<sup>-1</sup> and the high-frequency range 2800-3500 cm<sup>-1</sup>, in order to identify molecular bonding [6]. LFR is much less used in such studies despite being able to identify, protein conformation, rotational and liberational modes as well as hydrogen bonding interactions [7]. In this work, LFR scattering on human colorectal tissues will be also presented.

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## Thermal Transport by Long-Range Surface Plasmon-Phonon-Polaritons in Bilayer and Double-Layer Graphene

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Bilayer graphene (BLG) is a stack of two single-layer graphene sheets and it has unique electronic and optical properties that make it suitable for studying two-dimensional (2D) plasmons. In double-layer graphene (DLG), two single layers of graphene form a cladding of an insulating spacer layer with nanothickness. If the insulating spacer is made from a polar material such as silicon carbide or silicon dioxide, 2D plasmons in graphene interact with surface phonon-polaritons and form in general the surface plasmon-phonon-polaritons (SPPPs), in which the interaction of 2D plasmons with optical phonons in the spacer layer influences the dispersion and damping of SPPPs. The unique electronic properties of single and bilayer graphene allow for the engineering of the SPPP dispersion relation, providing a way to tailor the SPPP wave vector, frequency, and confinement. We predict that the long-range (LR) SPPPs should be efficient energy carriers due to their high group velocity and very long propagation length. LR SPPPs can propagate in a symmetric dielectric surrounding [1]. One of the important properties of BLG and DLG, which allows the existence of the LR SPPPs, is the normal-to-layer component of anisotropic dielectric tensor exceeding the dielectric function of the surrounding medium and finite thickness d of BLG or insulating layer in DLG [2]. In such symmetric dielectric surrounding, the propagation length of LR SPPPs is determined by the intrinsic relaxation rate  $\Gamma$  in the limit of  $\omega$ >> $\Gamma$  of the BLG or of two single layers of graphene in DLG and monotonously decreases with the increase of thickness d. BLG presents the 2D material with a smallest thickness, which supports the LR SPPP, because the single-layer graphene has zero physical thickness and supports only one 2D plasmon that is not a LR SPPP. Figure 1 shows the dispersion of three SPPPs, two symmetric and one antisymmetric, including the symmetric LR SPPP (a), antisymmetric optical (OP) and symmetric acoustic (AP) 2D plasmons (b) in DLG with Si spacer with d=100nm.

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**Figure 1:** Dispersion of long-range SPPP (a), optical (OP) and acoustic (AP) plasmons (b) in DLG with Si spacer with thickness d=100nm, suspended in air.

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#### Abstract

The few-layer vdW nanosheets exhibit unique physical properties and are prospective for various applications in optics and electronics including quantum technologies. Coherent phonons in vdW nanolayers are known as an instrument to control physical properties on ultrafast time scale [1]. We report on generation and detection of coherent phonons in suspended  $MoSe_2/WSe_2$  heterobilayers with a wide range of twist angles using picosecond laser ultrasonic techniques. Coherent acoustic phonons, observed as temporal oscillations in probe optical reflectivity, are identified as breathing mode, that modulates the bilayer thickness and mean refractive index of heterobilayer. The phonon frequency of ~0.8THz and a lifetime up to ~5ps are obtained, which have no distinct dependences on the twist angles between two layers. The amplitude of the generated phonons is the lowest at the intermediate twist angles that is explained by a significant role of Coulomb attraction between the layers. Coulomb attraction is a specific feature for phonon generation in heterobilayers and appears instantaneously with the femtosecond pump excitation due to ultrafast spatial separation of photoexcited carriers between the neighbouring layers. The importance of three mechanisms in the phonon generation process, i.e., electrostatic, thermoelastic, and deformation potential, explains the amplitude of phonon induced reflectivity changes of ~10<sup>-5</sup>, which is an order of magnitude higher than in MoSe<sub>2</sub> homobilayers [2].

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#### **Figures**



**Figure 1:** (a) Sketch of the  $MoSe_2/WSe_2$  heterobilayer. (b) Microscopy image of one sample stacked at 5.5° over the patterned holes. (c) Temporal signals  $\Delta R/R$  after subtraction of the background measured in the flakes with various stacking angles. (d) The fast Fourier transforms of the signals shown in panel (c).

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#### Abstract

In contrast to normal diffusion processes, thermal conduction in one-dimensional systems is anomalous. The thermal conductivity is found to vary with the length as  $\kappa \sim L^{\alpha}$  ( $\alpha > 0$ ), while there is a long-standing debate on the value of the exponent  $\alpha$ . Here we present a canonical example of this behaviour in densely packed polymer-grafted spherical nanoparticle (GNP) melts at fixed grafting density and nanoparticle (NP) radius. For long chains (degree of polymerization  $N \ge 1000$ ), the experimental  $\kappa(N)$  of GNP melts decreases with N, i.e. polymer composition. For N < 1000, however,  $\kappa(N)$  unexpectedly increases with N thus displaying a pinning behaviour near  $N \sim 1000$ . For a GNP melt, the extensional free energy per polymer chain attains a maximum at  $N_{max} \approx 940$  for  $\sigma = 0.47$  chains/nm<sup>2</sup> that indicates domination of extended conformation at short N and Gaussian-like conformation for longer N. In the former region, thermal conductivity of extended polymer chains increases with N and follows  $\kappa_p \sim N_{dry}^{0.49 \pm 0.03}$  that provides experimental evidence of the novel class of superdiffusive thermal transport with  $\alpha = 1/2$ .

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#### Figures



**Figure 1:** Left panel: schematic illustration of two poly(methylacrylate) (PMA) grafted silica nanoparticles (GNP); Right panel: Thermal conductivity  $\kappa$  of the GNP melts as a function of the PMA degree of polymerization, N. The representation of the experimental  $\kappa$  by an effective medium model with different prefactor b and exponent  $\alpha$  describing the PMA contribution ( $\kappa_p = bN_{dry}^{\alpha}$ ) is indicated by coloured solid lines. The dashed line denotes the thermal conductivity of the bulk PMA.

## Phonons 2023



# Low frequency Raman spectroscopy as a characterization tool of nanoplatelets surface chemistry

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Cadmium chalcogenides are direct semiconductors that can be synthesized into the shape of nanoplatelets (NPLs) in which the exciton confinement is determined by the number of atomic layers that make up their thickness. This number of layers is precisely controlled during the colloidal synthesis process, resulting in atomically flat nanoplatelets with controlled optical properties. However, colloidal synthesis is performed using surface ligands that can be seen as a part of this hybrid material and as such, they induce changes in their physical properties. The development of a tool to characterize the presence of these ligands and their impact on the NPL structure is thus essential to develop new 2D materials with controlled properties.

Raman spectroscopy is an excellent candidate to characterize the nature and density of the ligands in particular in the low-frequency Range. In this range, the vibration of the nanoplatelets thickness can be measured, and we demonstrated in our previous works that this vibration is sensitive to the presence of surface ligands. Indeed, by increasing the length of the ligands, a down-shift of the vibration frequency of the NPL is observed. This frequency change is mainly induced by the mass change of the ligands [1] but we also observed structural modifications of the nanoplatelets itself as their curvature change depending on the surface ligand size.

We are now using low frequency Raman scattering to monitor these structural changes. Thiol ligands with identical masses and different conformations are used to control the NPLs curvature and rolling. While the steric hindrance of the ligands increases we observe an unrolling of the nanoplatelets and a slight increase of the vibration frequency (figure below). Combination of absorbance spectroscopy, Raman measurements and EDX allowed us to identify the origin of this frequency change induced by a decrease of the ligands surface density as their steric hindrance increased.

We believe that low frequency Raman spectroscopy can then become an essential tool for the nanochemist seeking a fast and precise characterization of its nanoparticles surface ligands nature and density.

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**Figure 1:** TEM images of 3 monolayers CdSe Nanoplatelets with: a) 1-Octanethiol or b) 2-Ethylhexanethiol ligands on their surfaces. c) Corresponding low-frequency Raman spectra.



Figures

## High-order topological state by band inversion

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Topological interface states have been demonstrated for a wide range of excitations (photons, phonons, vibrations, polaritons). In particular, acoustic interface states have been evidenced in acoustic superlattices with frequencies at tens to hundreds of GHz [1,2]. A scheme to generate interface state in one-dimensional superlattices is based on the principle of band inversion, obtained by concatenating two periodic lattices with inverted spatial mode symmetries around the bandgap [3]. Most of the realizations exploit a given bandgap for which there is one inversion of symmetry. In this work, we present high-order topological nanophononic interface states in multilayered structures based on GaAs/AlAs. We achieve band inversion by modifying the internal unit cell structure of the two lattices [4]. We extend the principle of band inversion to create an interface state present for the second bandgap to higher order bandgaps (see Fig.1). By carefully choosing the appropriate material thickness ratio in the two concatenated superlattices we show that we can engineer interface states at the nth bandgap. We can engineer versatile topological devices where interface states are simultaneously created in a large frequency range. In addition, we designed hybrid structures formed by two concatenated superlattices with different order bandgaps centered around the same frequency that support interface states. The topological modes can be experimentally accessed in Brillouin or pump-probe experiments. Furthermore, the ability to explore higher bandgap order acoustic interface states in the GHz range is due to the linear dispersion relation of acoustic phonons, therefore we can investigate regions that are difficult or impossible to study in electronics or optics due to the non-linear dispersion relations.

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Figures



**Figure 1:** Band inversion of the acoustic bandgap. The mode symmetries are indicated with orange (symmetric) and blue (anti-symmetric) lines.

# Full Band Monte Carlo simulation of 2D h-BN nanostructure for phonon transport based on ab initio calculation

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Nanostructures based on two-dimensional (2D) thermoelectric materials have acquired a prominent role in recent research due to their fascinating properties and potential ability to recover lost thermal energy [1]. In other words, effective thermal management becomes a critical issue to increase the energy efficiency of electronic devices [2]. Among them, 2D hexagonal boron nitride (h-BN), a structure very similar to graphene, is gaining importance due to its high chemical and mechanical stability, revealing a wide range of applications [3]. Despite the potentials of this material, only a small fraction of its opportunities has been explored to date, and relatively few theoretical and computational studies have been conducted on it [4]. In this work, we present a Full-Band Monte Carlo approach to solve the Boltzmann's transport equation for phonons using ab initio parameters for both dispersion (shown in Fig. 1 (a)) and scattering rates [5]. Ab initio methods are performed in the framework of the density functional theory (DFT) as implemented in Quantum ESPRESSO [6]. In the temperature range from 0 to 1000 K, phonon-phonon scattering rates taking into account anisotropy and isotope scattering, due to high isotopic disorder of boron are considered and plotted in Fig. 1 (b). Figure 1. (c) shows the calculated thermal conductivity as a function of the length in 2D

h-BN nanofilms compared to semi-analytical (SA) models from a ballistic regime occurring in ultra-short films to a diffusive one in long nanofilms. Static and dynamic performances are studied for different nanostructures. Finally, the presented methodology will allow a better understanding of a wide range of 2D materials and their nanostructures.

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#### Figures



**Figure 1:** (a) Calculated phonon dispersions for 2D h-BN along the high symmetry paths in comparison with experimental data [7]. (b) Average scattering rates as a function of temperature. (c) Thermal conductivity as function of length for 2D h-BN nanofilms at room temperature.

#### Acknowledgment

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## Photoexcitation of longitudinal and shear coherent acoustic phonons in single ferroelectric domain BiFeO3 thin films

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The general demand for smaller, faster, more reliable and increasingly efficient GHz-THz devices (spintronics, data storage, high-frequency transducers, or optoelectronic), leads us to look into the ultrafast processes in materials. Among technologically relevant materials [1], multiferroics are good candidates since they exhibit multifunctional properties based on the coupling between the polar, magnetic and lattice degrees of freedom. The multiferroic compound Bismuth Ferrite (BiFeO<sub>3</sub>) is a unique room temperature multiferroic with a small band gap accessible with a near UV photon energy [2]. Here, we report a study of the photoexcitation of coherent acoustic phonons in a single ferroelectric domain BiFeO<sub>3</sub> thin film. Using a two-color time-resolved pump-probe ultrafast spectroscopy, we report the observation of longitudinal and, for the first time, shear acoustic phonons in such ferroelectric thin film. (Figure 1) A complete pump fluence dependence in the range 0-5 mJ.cm<sup>-2</sup> of the Brillouin mode amplitude is presented. A combination of these experiments with picosecond time-resolved x-ray diffraction measurements [3] enables us to show that we can reach longitudinal and shear strain amplitude as high as 0.6% and 0.04% respectively [4]. With these results, the thermal and nonthermal coherent acoustic phonon generation processes are then discussed and provide a new insight into the complex interaction between charge and lattice dynamics in Bismuth Ferrite at the picosecond time scale.

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Figure 1: Time-resolved pump probe spectroscopy measurement.



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The study of heat transport in nanoscopic heterostructures is crucial for both fundamental research and technological applications such as thermal management in devices and heat-assisted magnetic recording.

As metallic heterostructures become smaller than the mean free path of their conduction electrons, the transport of thermal energy by electrons changes from diffusive to ballistic. Their mean free path is on the nanoscale and determined by electron-phonon coupling, along with the time required for equilibration of electron and lattice temperatures, which is on the order of picoseconds. Trilayer heterostructures combining metals with different electron-phonon coupling strengths provide an interesting case, where heat energy deposited in a few atomic layers of Pt is transported into a nanometric Ni film, which is heated more than a Cu film through which the heat is released. [1]

We investigate the effect of a thin Pt layer with high electron-phonon coupling on thermal transport through a 100 nm Cu/Au layer sandwiched between thin Pt and Ni films on picosecond time scales after femtosecond laser excitation. Using ultrafast x-ray diffraction (UXRD) at the MID end-station of the European XFEL, we track the time dependence of heat transport. A time-dependent diffusive two temperature transport model reproduces our experimental data and thus verifies the efficient transport channel from the laser excited Ptlayer to the Ni-layer via hot electrons.

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#### Figures



**Figure 1:** The ultrafast X-ray diffraction experiments reveal counterintuitive heat transport consistent with heat diffusion including electron-phonon interaction. The energy deposited in few atomic layers of Pt is transported into a Ni film, which is heated more than the 100 nm Cu film through which the heat is conducted. [1]



## Time-domain Brillouin scattering for evaluation of materials interface inclination: Theory

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Time-domain Brillouin scattering (TDBS) is a technique that uses ultrafast laser pulses to generate (pump) coherent acoustic pulses (CAPs) and detect (probe) them as they propagate through a transparent material for the probe laser wavelength [1-2]. The interference between the probe light scattered by the propagating CAPs and the reflection of the probe light on stationary surfaces/interfaces of the sample, called heterodyning light, generates a signal containing Brillouin oscillations (BOs). Upon reflection/transmission, CAPs can also produce echo features in the signal. Typically, TDBS is used to characterize laterally homogeneous materials or layered samples structured along the normal to the sample surface, i.e., where CAPs and probe laser pulses propagate collinearly. The interaction of plane collinear sound and light waves effectively explains the TDBS signals in such cases. However, to analyze recent experiments with buried grain boundaries [3-4] or material interfaces [5-6] inclined relative to the probe beam propagation direction, the theory should be extended to the case of 180° Brillouin backscattering in non-collinear propagation of probe light and CAP beams. Both beams can indeed deviate from collinear propagation and heterodyning directions when they are incident on such boundaries/ interfaces. We have developed an analytical theory that describes TDBS probing of an acoustic Gaussian beam by a Gaussian probe light beam inclined relative to the sound propagation direction within their Rayleigh ranges [6]. The temporal profiles of CAPs and laser pulses are also Gaussian. Recently, we extended the theory to the case where the CAP is incident on the stationary surface not normally and at a shifted position relative to the position where the probe beam is normally reflected to provide the heterodyning probe beam. This analytical theory provides useful insight to appreciate the influence on the BOs and echo features of a TDBS signal of (i) the CAP and probe beams radii, (ii) the CAP length, (iii) the angle at which sound and light waves interact, (iv) the mismatch of spatial overlap at the interface, and (v) the CAP reflection coefficient value. The developed theory can be used for estimating the inclination angle of an interface from a single TDBS signal which allows local estimation of interface inclination without the drawback of lateral averaging when such estimation is done from several measurements at different lateral positions. This application of the theory will be developed in another communication [6].

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## Polarization-controlled Brillouin scattering in elliptical optophononic micropillar resonators

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Brillouin scattering is extensively used for material characterization, biological imaging, optical and optoelectronic devices [1, 2]. In Brillouin scattering processes, the selection rules formally constrain energy, direction and polarization of the scattered photons for a given input state. Usually, these selection rules in crystalline solids are considered as intrinsic material properties, and the signal states depend on the relative polarization of the excitation [3]. Here, we use elliptical optophononic micropillar resonators to control the optical polarization selection rules (Fig. 1(a)). The elliptical cross-section of the micropillars lifts the degeneracy of the optical cavity modes, leading to orthogonally polarized optical cavity modes and splitted in energy, as observed in Fig. 1(b). The polarization state of an optical field will depend on both orthogonal cavity modes and their associated polarization states. Therefore, an incident laser beam equally detuned from both cavity modes, and polarized along the diagonal axis of the elliptical pillar undergoes a rotation of polarization. Furthermore, the rotation of polarization is wavelength dependent, which results in a different polarization state for the Brillouin scattering signal. By choosing the polarization and wavelength of the incident laser, we demonstrate that the polarization state of incident and reflected laser and the Brillouin signal are different [4]. In this way, background-free spontaneous Brillouin scattering spectra can be efficiently measured in a cross-polarization scheme down to 18 GHz, as shown in Fig. 1(c). The same working principle applies to others optical system with localized, polarization sensitive modes, such as plasmonic resonators, photonic crystals, and birefringent micro- and nanostructures.

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**Figure 1:** (a) SEM image of an elliptical pillar. *m* and *n* correspond to the major and minor axis lengths of the cross-section, respectively. (b) Measured cavity reflectivity for horizontally (blue) and vertically (red) polarized incident light as a function of the laser-cavity detuning energy. Inset: Schematic of the pillar cross-section. (c) Brillouin spectrum of an elliptical micropillar.

### Heat transfer in crystalline clathrate compounds at low temperatures

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Clathrates are inclusion compounds in which guest atoms or molecules are placed inside the host framework. The thermal conductivity of crystalline clathrate compounds is still a subject of discussion, as it has certain peculiarities.

The experimental results on the thermal conductivity  $\kappa(T)$  of some ordered and disordered clathrate hydrates, thermoelectric clathrate compounds, and skutterudites are analyzed. For most of them in a wide region of temperatures above 2 K,  $\kappa(T)$  exhibits a behavior typical of disordered solids, which depends weakly on their chemical composition, crystalline structure, and microstructure. [1,2]

An analysis of temperature dependence data of the thermal conductivity of clathrate compounds in the region where phonon-phonon scattering processes predominate was carried out. It is demonstrated that the temperature dependence of all the mentioned crystals can be described in a unified way by the expression  $\kappa(T) = \kappa P + \kappa 0 \exp(-E/T)$ . The first term,  $\kappa P$ , is the standard Peierls contribution to the conductivity and the second term is related to the wave-like tunneling and loss of coherence between different vibrational eigenstates.

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The thermal properties of transition metal dichalcogenide (TMD) crystals are currently not fully understood, particularly regarding the effects of the material's thickness and its environment. This knowledge is crucial to engineer heat transport and develop applications using these materials. In this work, we used an experimentaltheoretical approach to gain a better understanding of the intrinsic lattice thermal conductivity of MoSe<sub>2</sub>, a representative TMD. We measured the thermal conductivity using Raman thermometry on a range of clean, single-crystalline, suspended MoSe<sub>2</sub> flakes with different thicknesses, prepared as in Ref. [2] and compared the experimental results with ab initio simulations using phonons at finite temperature. Our results indicate that while phonon dispersions and lifetimes change significantly with flake thickness, the in-plane thermal conductivity of MoSe<sub>2</sub> remains relatively constant. [3] This is in contrast with the demonstrated reduction of thermal conductivity of 3D-bonded crystals when thickness is decreased due to increased phonon boundary scattering at the surface. [4] In the thinnest TMDs, this increased phonon boundary scattering effect does not seem to play an important role, likely due to the 2D nature of the TMDs. Furthermore, heat-carrying modes emerge around 0.1 THz that compensate for the reduction of available modes for thinner films. The large surface-to-volume ratio of suspended monolayer flakes also results in efficient out-of-plane heat dissipation to air molecules, increasing their apparent thermal conductivity by an order of magnitude. Our findings have important implications for designing TMD-based devices towards thermal management, opto-electronic and high-frequency electronic applications.

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#### Figures



**Figure 1:** Experimental and theoretical in-plane thermal conductivity  $\kappa$  of MoSe<sub>2</sub> (*left*) and its spectral decomposition  $\kappa_{\text{spec}}$  as a function of frequency and flake thickness at 400 K (*right*)
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Based on the same principles as for light focussing, acoustic Fresnel lenses has been proposed and experimentally demonstrated at low frequencies in air and water [1] [2]. In this work, we investigate the use of acoustic Fresnel lenses at much higher acoustic frequencies in pump-probe coherent phonon experiments. We use a 5 GHz GaAs/AlAs superlattice (SL) transducer pumped by a laser pulse which is spatially modulated in the form of a Fresnel zone plate for the generated acoustic wavelength, ~ 1 $\mu$ m. This is achieved by fabricating gold shadow masks corresponding to the Fresnel aperture designs on the surface of the transducer SL by optical lithography, Fig.1(a). The near-IR pump light was efficiently reflected from the gold coated areas, resulting in a spatially modulated light intensity distribution on the SL transducer. The spatial distribution of the 5 GHz phonon signal intensity on the opposite side of the GaAs substrate was measured using a scanning probe beam. Our results, Fig.1(b), show the focussing of the phonons by the lens aperture resulting in an intensity distribution which is much narrower than the original pump laser spot.

The results of our experiment demonstrate the ability to obtain focussed coherent phonons by spatially modulating the pump laser intensity with a Fresnel lens aperture. Using a spatial light modulator based on, e.g. a DMD device, instead of the gold aperture could lead to wider applications of the technique in laser ultrasonics. Potential applications include the generation of very high strain amplitudes at a point while the pump fluence is kept below the damage threshold of the transducer. This provides a promising avenue for the engineering of phononic devices with potential applications in acousto-optics, phononics, and optomechanics.

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**Figures** 

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**Figure 1:** (a) Experimental arrangement: the gold Fresnel aperture pattern is fabricated on top of the 5GHz GaAs/AlAs opto-acoustic transducer; (b) linescans of the phonon signal on an axis through the centre of the pump beam.

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We report generation of ultra-broadband longitudinal acoustic coherent phonon wavepackets in SrTiO<sub>3</sub> (STO) with frequency components extending throughout the first Brillouin zone<sup>1</sup>. The wavepackets are efficiently generated in STO using femtosecond infrared laser excitation of an atomically flat 1.6 nm-thick epitaxial SrRuO<sub>3</sub> opto-acoustic transducer film. We use femtosecond x-ray diffraction at the European X-Ray Free Electron Laser Facility to study the dispersion and damping of phonon wavepackets. The experimentally determined damping constants for multi-THz frequency phonons compare favourably to the extrapolation of a simple ultrasound damping model over several orders of magnitude.

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**Figure 1:** (a) Cubic unit cell of SrTiO<sub>3</sub> with a perovskite structure. (b) Atomic force microscopy topography of the SrRuO<sub>3</sub> surface revealing monoatomic terraces. (c) Measured and calculated static x-ray reflectivity curves from the SrRuO<sub>3</sub>/SrTiO<sub>3</sub> heterostructure. Thickness oscillations correspond to the 1.6nm-thick SrRuO<sub>3</sub> transducer film. The upper x-axis shows Γ and X as the SrTiO<sub>3</sub> Brillouin zone center and boundary, respectively. Dashed vertical arrows indicate positions at which femtosecond time series shown in d) were taken. (d) Femtosecond X-ray reflectivity oscillations observed at different reciprocal lattice points L. Simple analysis of these damping oscillations allows for the determination of coherent longitudinal acoustic phonon dispersion and life time in SrTiO<sub>3</sub>.

### Low Mode-Volume Surface Acoustic Wave Cavities for Coupling to Single Photon Emitters

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Recently, surface acoustic waves (SAWs) have attracted attention in the quantum information field with applications including hosting quantum states and transduction between disparate quantum systems. They have been dubbed a 'universal transducer' [1], as the strain and EM field carried by a SAW can efficiently couple to almost any qubit technology. Paramount to most proposed schemes is SAW cavities with small mode volume and high quality-factor (Q). Small mode volume ensures strong interaction between target systems and single phonons and large Q is required for storage of the phonons for long enough to complete interactions.

Here, we report on design and fabrication of SAW microcavities on GaAs substrates for microwave-tooptical signal transduction with embedded InAs quantum dots. Microwave electrical power is coupled to the SAW cavities with inter-cavity superconducting interdigitated transducers (fig. 1a). We demonstrate curved, focusing cavities at 3.6 GHz with mode volume down to  $6\lambda^3$  and estimated zero-point motion of 1.3 femtometers (fig. 1b). At these sizes, a linewidth of less than one MHz at cryogenic temperature is maintained with careful correction of the anisotropic speed of sound in GaAs and mitigation of bulk scattering from our etched-grove Bragg reflector cavity mirrors [2].

The above microcavities also contained InAs quantum dots that act as single photon emitters and couple to the strain field via the deformation potential [3]. Spectrally resolved emission from these QDs under microwave excitation show strong modulation at very low applied RF powers (fig 1c). From these measurements, a fit to the modulation depth, and careful calibration of the microwave power coupled into the cavity we can estimate a single phonon coupling strength of  $g_0=2\pi$ \*1.2 MHz. The value of this important figure of merit is comparable or better than many of the currently most successful microwave-to-optical transduction platforms [4]. Prospects for transduction at the single phonon level and planned improvements for the next generation of devices will also be discussed.

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#### Figures



**figure 1.** (a) cartoon of QD-SAW transducer. (b) AFM image of the SAW microcavity device used in this work. Bright stripes are ground and signal leads, the narrow fingers made of Niobium connected to the leads are the IDTs, and dark curved features are the etched-groove cavity mirrors. (c) Photoluminescence spectrum of a QD located at the focus of the microcavity shown in (b) at -56 dbm excitation on resonance (red) and off resonance (black).

## Complete thermoelectric characterization of integrated silicon nanowires using TEMcompatible microdevices

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Currently employed materials in Thermoelectric Generators (TEGs) such as bismuth telluride or lead telluride are scarce, expensive, toxic, and environmentally harmful. In recent years, the thermoelectrics paradigm has changed mainly due to the introduction of low-dimensional materials able to reduce the thermal conductivity by phonon scattering. Semiconductor nanowires (NWs) have demonstrated fascinating properties with application in various fields, including energy and information technologies. In particular, increasing attention has been focused on Si and silicon alloy NWs for applications in thermoelectric generation after recent successful implementation in miniaturized devices. Despite this interest, a proper evaluation of such nanostructures' thermoelectrical properties still represents a great challenge, especially when the complete characterization of the device-integrated NW is desired.

In this work, we describe the thermoelectric analysis of epitaxially suspended rough Si NWs fabricated using a bottom-up approach following the Vapour-Liquid-Solid (VLS) method. The thermal analysis is carried out using a technique that simultaneously combines the Joule self-heating of the nanowire with micro-Raman thermometry for their thermal profiling. This sub-micrometer-resolved temperature profiling is achieved using a custom-made set-up for the acquisition of the Raman spectra within a temperature-controlled vacuum chamber. The high resolved signal of the nanostructure can be measured thanks to specifically designed silicon-based micro structures with though-trenches that eliminate the background signal of the substrate. Over the same device, temperature gradients can be built up at each side of the NW. Combined with electrical access to the NW, electrical and Seebeck characterization can be performed on the same NW, thus completing the thermoelectric characterization. This device also enables the analysis of the studied NW with Transmission Electron Microscopy (TEM) for an accurate morphological and structural analysis.

This experiment allows to characterize the electrical and thermal conductivity of individual Si NWs and the effects of a high surface roughness on these properties. The defect-free crystalline structure of the NWs and the nature of their roughness is studied with TEM. Complementary, the shape of the obtained thermal profiling yields information about the absence of electrical or thermal contact resistance between bulk and nanostructure, confirming the double-side epitaxial attachment expected for a VLS grown NW. Finally, the combination of Joule and laser power tests required for the calibration permits to estimate the light absorption coefficient of such rough surfaces.

In conclusion, the combination of this set of techniques enables us to verify the integration of VLS NWs in the very same configuration as those used into micro-thermoelectric generators, yielding accurate values for the thermoelectric properties that can be directly applied on the improvement in the output power of these generators.

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# Surface Acoustic Wave propagation on nanostructured photoresist thin films fabricated by soft lithography

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Surface wave propagation (SAW) is a physical phenomenon which appears in a wide range of different systems, both at the surface of liquids and solids. Excited by optical or electrical means, SAWs present a particular interest when they interact with discontinuities in thin layers<sup>1</sup>. During propagation, SAWs generate strain-induced refractive index changes along the surface of the sample, which locally alter the value of the reflectivity at very short time scales<sup>2</sup>. Due to their nature, it is possible to use non-contact optical techniques in relatively simple geometries to study these reflectivity changes in a fast manner, which allows to non-invasively characterize the mechanical properties of thin films and other SAW-based devices<sup>3</sup>.

Recently, soft lithography (SL) has emerged as a promising fabrication technique for large samples (>1 cm<sup>2</sup>) with features down to the nanometer scale in an inexpensive and fast way, which could pave the way for their use in commercial devices. Here, we explore the temporal evolution over large fields-of-view (100×100  $\mu$ m<sup>2</sup>) of SAWs generated by absorption of ultrafast light pulses on thin films of nanostructured photoresist materials fabricated by using SL. We also study the changes in the SAW spectrum when modifying the geometrical properties of the nanostructures in the sample (lattice period, pillars height and diameter).

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### Figures

**Figure 1: a)** Experimental design of the SAW characterization system. Abbreviations: LS: laser source, PBS: polarizing beam splitter, BS: beam splitter, PD: photodiode, AOM: acousto-optic modulator, SHGC: nonlinear second harmonic generation crystal, L: lens, TL: tube lens, CAM: digital camera, OB: microscope objective,  $\lambda/2$ : half wave plate,  $\lambda/4$ : quarter wave plate, DF: dichroic filter. **b)** Snapshot of the SAW propagation over a 100×100  $\mu$ m<sup>2</sup> region of a glass substrate covered with a 1  $\mu$ m thick Su8 photoresist film.



# Can coherent LO phonon and LO-phonon-plasmon coupled mode be generated in the absence of the photo-Dember effect in a heavily doped *n*-type InSb crystal?

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Terahertz electromagnetic waves, which are emitted from surfaces of narrow gap semiconductors with the use of illumination of femtosecond laser pulses, provide information on the photogenerated carrier diffusion process, the so-called photo-Dember effects. The photo-Dember effects result from the difference in a diffusion coefficient connecting with mobility between electrons and holes [1-2]. The diffusion-coefficient difference builds up transient polarization producing the terahertz wave. In a typical narrow gap semiconductor, InSb, the emission process of the terahertz wave is dominated by the photo-Dember effects. We, here, point the fact that the earlier terahertz time-domain spectroscopic works in InSb was focused on lightly doped samples [3]. It is reasonable to assume that the diffusion process is strongly influenced as the background carrier density is increased. This is because the mobility depends on the carrier density. Accordingly, for verifying the above assumption, it is meaningful to clarify the characteristics of the terahertz wave emitted from the InSb crystals with the relatively high background carrier density. In the present work, we investigated the terahertz-wave emission from undoped and *n*-type InSb single crystals with relatively high major carrier density of  $1 \times 10^{17}$  cm<sup>-3</sup>. We observed the terahertz wave originating from the photo-Dember effects in the undoped InSb crystal as shown in Figs. 1(a), appearing as the strong pulse at the time delay of 0 ps, and 1 (c), appearing as the main frequency band at the frequency of 2 THz. In contrast, we found that the terahertz wave originating from the photo-Dember effects disappears in the *n*-type InSb crystal as shown in Figs. 1(b) and 1(d). According to Ref. 2, the photo-Dember voltage  $V_D$  is proportional to  $\ln[1+\{(b+1)\Delta n/(bn_0+p_0)\}]$ , where  $n_0$  and  $p_0$  is background electron and hole densities, respectively, and b is the ratio of the electron mobility to hole mobility:  $\mu_e/\mu_h \approx 100$ . In addition, the quantity  $\Delta n$  is the photogenerated electron density. It is apparent that, in highly doped InSb crystal,  $bn_0$  is much larger than  $p_0$ , which reduces the photo-Dember voltage  $V_D$ . We, therefore, attribute the origin of the observed difference between the undoped and *n*-type InSb crystals to the fact that the photo-Dember voltage is suppressed in the case where the background electron density is relatively high. We, however, note that the coherent LO phono-plasmon coupled modes are observable in the *n*-type InSb crystal. This means that the ultrafast photo-Dember effect does not necessary generates coherent LO-phonon-plasmon coupled mode. The present finding indicates that only the ultrafast change in the built-in electric filed caused by the photo-Dember effects cannot account for the generation of the coherent LO-phonon-plasmon coupled mode in heavily doped narrow-gap semiconductors.

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Figures



Figure 1: Terahertz waveforms from (a) the undoped InSb crystal and (b) the *n*-type InSb crystal (c) Fourier power spectra of the terahertz waves show in (a). (d) Those of the terahertz waves shown in (b).



## Brillouin Light Scattering investigation of nano-scale soldering in polymer colloidal crystals exposed to hydrostatic gas pressure

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### Abstract

Brillouin light scattering (BLS), as a non-contact optical technique, is very helpful in studying the mechanical and acoustic properties of polymer CCs. Using BLS studies, we found that polystyrene (PS) CCs exposed to supercritical gases showed an increase of particle contacts due to the plasticization effect. Such mechanically reinforced CCs preserved the shape and periodicity of individual particles; BLS allows us to in-situ monitor their vibrational frequencies at the specific gas pressure. In this work, we used high-pressure Brillouin Light Scattering (BLS) as a non-destructive tool to in-situ monitor the nano-scale soldering in polymer colloidal crystals (CCs) when exposed to supercritical fluids[1]. BLS is very sensitive to particle contacts, which allowed us to quantitatively determine the relative changes in particle contact area[2]. We illustrated the dependence of particle size in soldering and the influence of different gases in manipulating the vibrational frequencies of CCs using BLS measurements. This work demonstrates the importance of BLS in the study of PS CCs exposed to supercritical fluids. Also, our results can provide guidelines for designing mechanically robust CCs.

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**Figure 1:** Size-dependent nanoscale soldering of polystyrene colloidal crystals illustrated by the change of effective Elastic modulus ( $C_{eff}$ ) when exposed to Ar and He gas pressure; investigated by in-situ high pressure BLS experiments.

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# Optimal generation and detection of GHz acoustic phonon using elliptical micropillars

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Micropillar structures based on GaAs/AlAs multilayers can confine photons and phonons at ultrahigh frequencies [1][2]. The investigation of phonons in these structures can be performed in the time domain with the pump-probe technique. The intense pump pulse triggers the phonons in the system, and the probe detects the transient changes in reflectivity. However, the optimal generation and detection of acoustic phonons in the resonators occur at the center and the slope of the optical cavity, respectively, which makes simultaneous optimization of generation and detection of acoustic phonons difficult [3][4]. Here, we overcome this issue by designing elliptical micropillars to manipulate the polarization of microcavity modes [5][6]. The elliptical micropillar resonator lifts the degeneracy of the optical cavity modes, leading to orthogonally polarized cavity modes (horizontal and vertical, H/V) with shifted wavelengths. By aligning the polarization of the pump and the probe beams with H and V modes in a cross-polarization scheme, we experimentally achieve the optimization of phonon generation and detection. Figure 1 (a) top panel shows the measured H(blue) and V(red) modes in three pillars with different ellipticities. The mode splitting increases as the minor axis decreases. The bottom panel corresponds to the cross-polarized laser pulses coupling with the corresponding optical modes. The pump is tuned in resonance with V mode to excite the structure while the probe, coupled to the H mode, detects the dynamics of the phonons. For the circular pillar, in which there is no splitting, the probe is at the center of H mode. Therefore, it is insensitive to differential changes in the index of refraction. By changing the ellipticity of the pillar, the modes splitting reach a value in which, when the pump is tuned to the center of the V mode, the probe is at the slope of the H mode or outside it. We compare the phonon spectra in 5 pillars with different ellipticities demonstrating a simultaneous optimization of the generation and detection of acoustic phonons.

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### Figures



**Figure 1:** (a) Top panel: measured optical reflectivity of micropillars with different ellipticities. Bottom panel: pump(red) with V polarization is tuned at the center of the V mode to maximize the generation. Probe(blue) with the same wavelength as the pump is coupled to the H mode. (b) Phonon spectrum power as a function of modes splitting and minor axis.



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### Abstract:

Topological bosonic excitations such as photons, phonons, and magnons potentially enable low-loss transport of information and energy in analogy to electronic topological states. Compared with photons, phonons have smaller mode volume that can increase the coupling strength with spins to reach the quantum coherent regime and enable atomic-scale chiral topological bosons. Van der Waals magnetic materials are promising to realize such states due to their recently discovered strong interactions among the electronic, spin, and lattice degrees of freedom. Here, we report the first observation of coherent hybridization of magnons and phonons in monolayer antiferromagnet FePSe<sub>3</sub> by cavity-enhanced magneto-Raman spectroscopy. The robust magnonphonon cooperativity in the 2D limit, which accidentally occurs at zero magnetic field, originates from the material's large single-ion anisotropy, weak interlayer exchange coupling, and bond-dependent spin-exchange interaction. We then identified nontrivial band inversion between longitudinal and transverse optical phonons caused by the strong coupling with magnons. The spin and lattice symmetry theoretically guarantee magnetic field-controlled topological phase transition, which is also verified by non-zero Chern numbers calculated from coupled spin-lattice model. The 2D topological magnon-phonon hybridization potentially offers a new route toward quantum phononics and magnonics with ultrasmall footprint.

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### Figures



**Figure 1:** (a) Strong coupling of phonons and magnons in monolayer FePSe3 observed by magneto-Raman spectroscopy. (b) The optical image of the monolayer and bilayer samples in optical half cavity. (c) The magnetic field-tunable topological gap in the hybrid magnon-phonon band structure according to the symmetry of the observed modes.



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### Abstract:

Spectroscopy in the "new terahertz gap" between 5 to 15 terahertz (THz), or 166 to 500 cm<sup>-1</sup>, provides additional fingerprints of molecular functional groups and allows the optical manipulation of quantum materials based on vibrational resonances. This spectral range is very challenging to study with conventional optics and photonics. Phonon-polaritonic materials, structures, and devices are promising for photonics in the THz gap due to their low loss, but existing phonon polariton materials are narrowband and mostly serve the mid-infrared frequencies. Here we experimentally demonstrate that crystalline perovskite oxide SrTiO<sub>3</sub> is an excellent material platform for broadband, ultrafast, and high-field THz photonics due to its superior phonon polaritonic properties. As a proof of concept, we designed and realized SrTiO<sub>3</sub>-based phonon-polaritonic THz pulse amplifiers without the need to pattern the crystalline material itself. We have formulated the design rules for devices capable of coherently amplifying transient THz fields by up to 6 times, as well as achieving maximum spectral enhancement of 90, for arbitrary polarizations in an optically resolvable area and with a broad tuning range from 7 to 13 THz. Such a working bandwidth far exceeds any previous phonon-polaritonic materials. We then experimentally verified the performance of the concentrator by measuring THz field-induced second harmonic generation and reported intense, multicycle THz pulses with a peak field of 0.5 GV/m realized by a table-top setup.



