Phonon Transport in the Gigahertz to Terahertz Range: Confinement, Topology and Second Sound

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Transport of heat and hypersound with gigahertz (GHz) to terahertz (THz) phonons, is crucial for heat management in electronics, mediating signal processing with microwave radiation, thermoelectrics, and various types of sensors based on nanomechanical resonators. Efficient control of heat and sound transport requires new materials, novel experimental techniques, and a detailed knowledge of the interaction of phonons with other elementary excitations. Wave-like heat transport, also known as second sound, has recently attracted renewed attention since it provides several opportunities for overcoming some of the limitations imposed by diffusive transport (Fourier's regime). The frequency-domain detection of GHz-to-THz phonons can be carried out in a remote, non-destructive, and all-optical manner. The ongoing development of nanodevices and metamaterials made of low-dimensional nanostructures will require spatially resolved, time-resolved, and anisotropic measurements of phonon-related properties. These tasks can be accomplished with Brillouin light scattering (BLS) and various newly developed variants of this method, such as pumped-BLS. In the near future, pumped BLS is expected to become useful for characterizing GHz topological nanophononics. Finally, second-sound phenomena can be observed with all-optical methods like frequency-domain thermoreflectance.

I. INTRODUCTION

Nowadays, material science and solid-state physics aim to provide devices with: (i) low energy consumption and efficient thermal management, (ii) mechanical robustness, and (iii) closed-loop lifecycle management. The efficient control of heat is particularly challenging as the device components approach nanoscale dimensions[1,2]. In addition, energy efficiency in signal processing can require transforming one type of signal into another, e.g., the transformation of microwaves into acoustic phonons with piezoelectric elements[3]. Mechanical robustness is necessary for wireless devices[3], sensors[4,5] and actuators[6], especially when they operate in harsh environmental conditions. On the other hand, closed-loop lifecycle management often requires trade-offs between devices' performance, durability, and environmental impact of their components.

The mechanical and thermal properties of materials are strongly connected with the physics of acoustic phonon transport in the gigahertz (GHz) to terahertz (THz) regime. This creates a growing demand for the so-called nanophononics, meaning nano- and meta-materials that enable control over phonons. In general, nanophononics offers heat management, efficient heat removal, energy harvesting, vibrational attenuation, sensors based on nanomechanical resonators, and optomechanics for mediating signal-processing with microwaves[7–9]. It goes without saying that the progress in this field requires the development of new sensitive probes of phonons that can be harnessed for contactless, non-destructive, mechanical, and thermal evaluation of functional materials. The generation, transport, and attenuation of GHz-THz phonons can be efficiently studied with various all-optical, inelastic light scattering techniques. In combination with optical microscopy, light-based techniques provide adequate spatial resolution for most practical designs of GHz phononic metamaterials[8–11]. The detection of acoustic phonons with light is a manifestation of optomechanical (OM) coupling. The OM coupling can be sufficiently strong for mechanical evaluation of single nanostructures[12–15]. Light-based techniques can be categorized into frequency (GHz-THz) and time-domain (ns-ps) methods. Frequency-domain methods include Raman thermometry[16,17], Brillouin light scattering (BLS)[18] and frequency-domain thermoreflectance (FD-TR)[19,20]. Time-domain methods include transient grating techniques[21–23], time-resolved thermoreflectance (TD-TR)[24,25], time-domain BLS[26] and the method of asynchronous optical sampling (ASOPS)[27,28]. In this work, we emphasize experimental works and particularly the techniques of BLS, FD-TR and TD-TR. We discuss how the recently developed pumped BLS approach can be used to study acoustic Anderson localization, one-way transport in acoustic diodes, and topological acoustics. In the discussion of BLS we omit its use on the biomedical sector[29–32] and on spintronics (magnonics)[33–36]. Finally, we turn our attention to thermal waves, otherwise known as Second Sound. In the hydrodynamic regime the temperature field is partially transported by thermal waves, as given by the hyperbolic heat equation. We provide a historical overview of the field, emphasizing the recent developments in experiments, theoretical aspects, and numerical experiments.

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II. SPONTANEOUS BRILLOUIN LIGHT SCATTERING FROM THERMAL PHONONS

The development of BLS started in 1922 with the work of Léon Brillouin[37] and continued with early contributions by Mandelstam in 1926[38] and with the discovery of stimulated BLS in 1964[39]. Initially, BLS was mainly applied to study bulk materials such as transparent glasses, polymers, and liquids that give a strong signal of inelastically scattered light. However, the development of high-quality single-mode lasers and multipass tandem Fabry-Perot interferometers[40] paved the way for more demanding BLS measurements of opaque, semiconducting, and spatially confined systems. Spontaneous BLS is a versatile, heavily used, frequency-domain probe of thermally populated GHz phonons[18], while stimulated BLS plays an important role in fiber optics[41]. Spontaneous BLS combined with optical microscopy, termed micro-BLS (μ-BLS), offers a spatial resolution of hundreds of nanometers to few micrometers that can be used to study sub-micrometer thick membranes and phononic metamaterials[11]. Nowadays, BLS is used for characterization of inorganic bulk and nanoscale materials, fiber optics, soft matter and biological systems, and for studying spin waves (magnons). On the one hand, BLS can be used to reveal the elastic properties of nanomaterials like nanospheres[42–45] (OD), nanowires[12] or nanopillars[46,47] (1D), and nanosheets[15,48] (2D). On the other hand, BLS can be used to study thermal transport[49–52].

A. Optomechanical coupling in low-dimensional materials

The OM-coupling driving BLS stems from two phenomena: the photoelastic coupling (PE) and the moving interface (MI) or surface ripple mechanism[53,54]. The PE coupling can be understood as the (bulk) inelastic diffraction of light from acoustic waves. On the other hand, the MI mechanism is inelastic light scattering by surface (interface) motions. Interestingly, the PE and MI effects can give inelastically scattered light of opposite phase leading to self-cancellation of BLS[43,55].

The OM-coupling and BLS signal of nanomaterials can be enhanced through the plasmonic near fields of metallic nanostructures[13,14,56–58]. However, we believe that the enhancement factor for BLS cannot reach that of surface-enhanced Raman scattering (SERS), in which the signal of molecular vibrations is enhanced by over 10^8 times with plasmons[59]. On the one hand, the strong enhancement observed in SERS is currently attributed to atomic-scale lightning rod effects close to the so-called picocavities[60]. Such phenomena are relevant for molecules but cannot be exploited for the confined acoustic modes of entire nanostructures. One the other hand, the BLS signal comes from volume (PE) and surface (MI) integrals that take into account the entire distribution of atomic displacements and their overlap with the electromagnetic fields[58]. This means that if there is a considerable spatial separation between the maximum atomic displacement and the maximum electromagnetic field, then the OM-coupling and the BLS enhancement will be limited. The

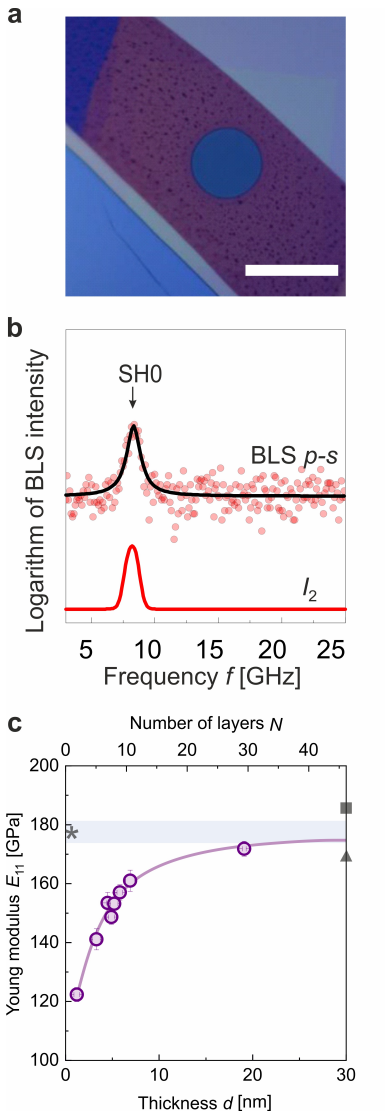


FIG. 1. Optomechanical coupling and elastic size effect in $MoSe_2$. (a) Optical microscopy of free-standing ultrathin $MoSe_2$ membranes suspended over circular holes on a Si_3N_4 substrate. (b) Evidence of ultra-strong OM-coupling in $MoSe_2$: experimental and calculated BLS spectra of shear-horizontal (SH) acoustic waves in a 6.9 nm-thick membrane. (c) The thickness dependent Young modulus of $MoSe_2$ showing significant elastic softening. Figures 1a-c obtained from Babacic *et al.*, Advanced Materials, Vol. 33, 2008614, 2021; licensed under a Creative Commons Attribution (CC BY) license.

experimental efforts for plasmon-enhanced BLS are facilitated by the development of electromagnetic perturbation theories for dissipative systems[61].

Nevertheless, the recent study of Babacic *et al.*[15] on few-nanometer thick free-standing $MoSe_2$ membranes (Fig. 1a) shows that the intrinsic PE coupling of some low-dimensional materials can be exceptionally strong. $MoSe_2$ is a van der Waals (vdW) layered material and a prototypical member of the transition metal dichalcogenides (TMDCs) family. In $MoSe_2$, the OM coupling enables detection of confined acoustic (Lamb) waves (Fig. 1b). As the membranes' thickness de-

creases, the bulk acoustic phonons transform into symmetric (S) and anti-symmetric (A) Lamb waves and shear-horizontal (SH) waves. All these waves can be probed with μ-BLS for selected polarization of the incident and scattered light (p-p for A and S, p-s or s-p for SH). Due to momentum-conservation during light scattering, the acoustic wavevector q of all these waves is parallel to the surface with magnitude $q = 4\pi \sin\theta/\lambda$, where θ is the angle of incidence and λ the wavelength of light. The measured dispersion relationships are sensitively dependent on the elastic properties, the membrane thickness, and the residual stress. Thus, μ-BLS is a new contactless method for elastic evaluation and measurements of thickness and stress of 2D nanomaterials. Interestingly, BLS is capable of detecting SH waves (Fig. 1b) due to PE coupling despite the minimal scattering volume governed by membranes thicknesses down to three molecular layers. This observation calls for further investigations of light-matter interactions and OM-coupling in ultrathin TMDCs.

Regarding bulk homogenous semiconductors, μ-BLS can probe under certain conditions all types of acoustic phonons: longitudinal (L) and transverse (T) bulk acoustic waves (BAWs), Rayleigh surface waves (RSWs), pseudo-RSWs, and the so-called high-frequency pseudo-surface waves (HFPSAWs). The surface-confined RSWs contribute to BLS through the surface-ripple mechanism (MI coupling). The probed RSWs have acoustic wavevector $\mathbf{q}$ parallel to the surface, identical with that of Lamb waves. The acoustic wavevector of HFPSAWs obeys the same relationship for highly opaque materials like TMDCs. In this case, the signal comes from sub-surface PE coupling. The back-scattering BLS from usual bulk phonons is suppressed due to the anisotropic optical properties of TMDCs and particularly the very short penetration depth at optical wavelengths. Sub-surface PE coupling also enables the detection of transverse (SH) bulk waves propagating in the close vicinity of the free surface. In conclusion, the optical properties of TMDCs facilitate momentum-resolved μ-BLS detection of all types of acoustic waves using the simple back-scattering geometry. These non-destructive measurements reveal the complete anisotropic elasticity of low-dimensional materials (elastic constants), which are inaccessible to semi-empirical techniques like atomic force microscopy nano-indentation.

B. Elastic size effect in 2D materials

As all fields of nanotechnology, research in nanophononics was strongly affected by the discovery of graphene. It has even been suggested that the most successful applications of graphene could stem from its phononic rather than electronic properties, and that practical applications will be based on few-layer structures rather than pure single-layers[62]. Graphene remains at the forefront of research in phononics, including exotic phonon-hydrodynamics phenomena[62] and second sound (section IV). However, the study of phonon transport in two dimensions can become particularly challenging for 2D materials beyond graphene, termed Van der Waals materials, which can have complicated compound structures and strong in-plane anisotropies.

Van der Waals materials and particular TMDCs and nanomaterials are expected to be the game-changer for flexible and miniaturized devices due to their unique electronic, optical, optoelectronic, and thermal properties with respect to bulk —see Refs. 15 and 48 and references therein. All real-world applications of TMDCs require mechanical and thermal durability, as well as accurate knowledge of their size-dependent anisotropic mechanical properties. In addition, a long-debated topic on TMDCs has been the effect of thickness on their elastic properties. These questions cannot receive a definite answer with techniques that give a spatial average of the elastic response (e.g., nano-indentation or buckling-based metrology). Instead, it is possible to combine BLS, Raman, and pump-probe measurements to find all the independent components of the elasticity tensor.

Following this approach, it has been shown that the elastic parameters of $MoSe_2$ are decreasing as the thickness gets smaller —see for instance the thickness-dependence of the in-plane Young modulus in Fig. 1c. The observed elastic size effect has two profound implications. First, it affects the performance of nanomechanical devices, such as nanomechanical resonators for sensors. Second, it is expected to affect the thermal transport with GHz-THz acoustic phonons in nanodevices. Thus, these measurements are also valuable for achieving good thermal management in nanodevices. Equipped with the knowledge on prototypical TMDCs like MoS_2 and $MoSe_2$, it is now possible to apply BLS on more complicated materials, such as 2D metamaterials based on TMDCs[63] and 2D materials with strongly anisotropic structures such as black phosphorus[64,65] and orpiment[66] (As_2S_3).

III. BRILLOUIN LIGHT SCATTERING FROM NON-THERMAL PHONONS

BLS of semi-opaque, light-absorbing, and ultrasmall semiconductors is characterized by a poor signal-to-noise ratio and requires exceptionally long exposure times. This obstacle can be overcomed with the recently developed technique called pumped-BLS[69]. This all-optical technique combines femtosecond laser excitation of non-thermal, coherent acoustic waves with continuous wave (CW) lasers for frequency domain detection. Using this technique, the BLS signal of photoexcited gigahertz phonons in semiconducting nanomembranes displays a hundredfold enhancement as compared with thermal equilibrium, without the need of prescribed metallic transducers. Recently, Rolle *et al.*[70] have used a similar approach to inject coherent phonons in opaque phononic crystals and study them with BLS.

Pumped-BLS can also be used in a time-resolved manner using appropriate electronics. Such time-resolved measurements are useful for studying phase transitions, for instance glass transitions[71]. Beyond pulsed lasers, time-resolved BLS can be combined with other external stimuli, such as magnetic fields for studying spin waves[72]. An interesting task for time-resolved BLS will be to examine the elastic properties of 2D materials with interesting topological properties and photoin-

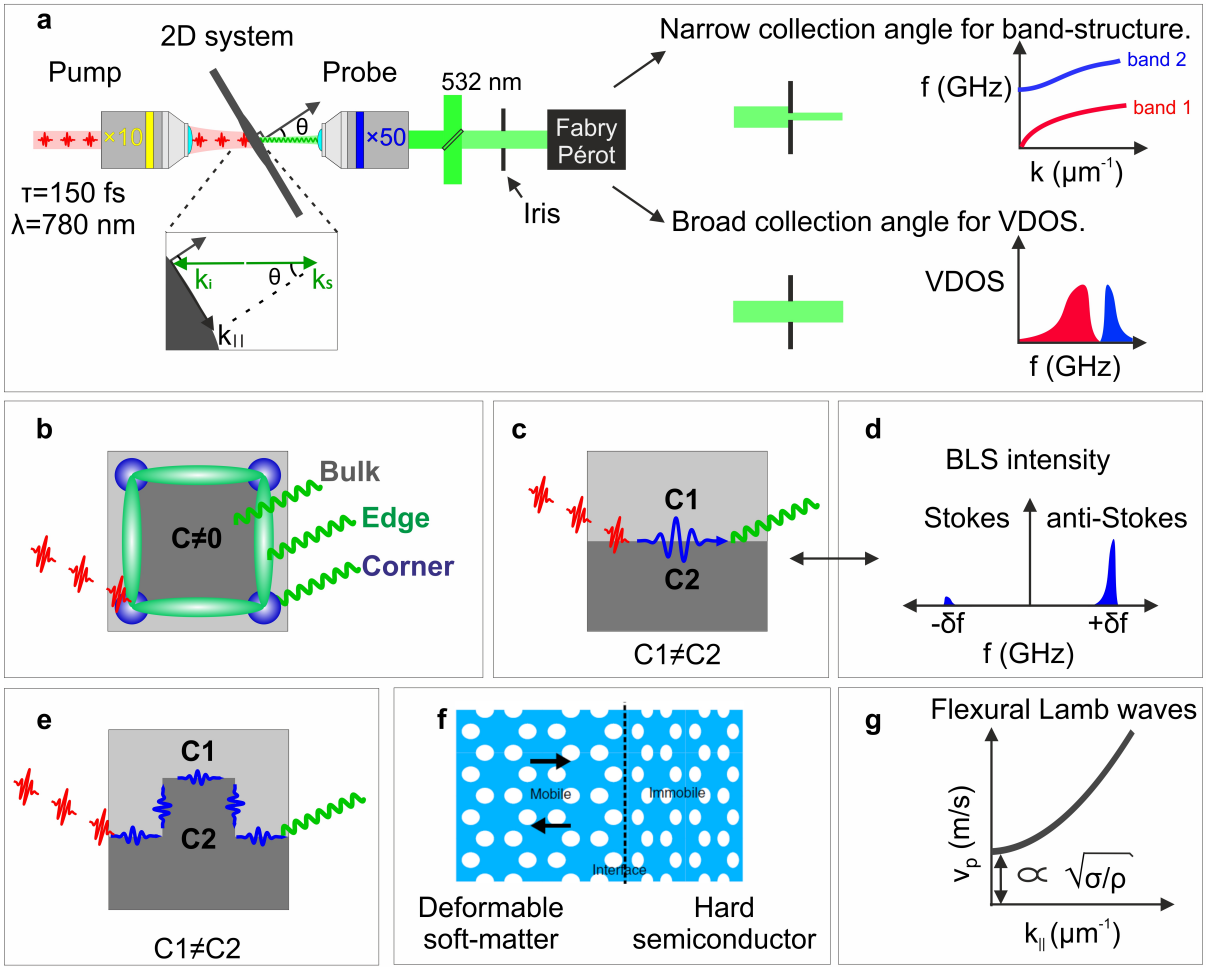


FIG. 2. Pumped Brillouin light scattering and its potential uses in topological phononics. (a) Schematic illustration of the pumped-BLS setup. (b) Pumped-BLS measurements of second-order topological phononic crystals. (c) (d) Excitation and detection of one-way edge states, and the resulting asymmetric Stokes and anti-Stokes intensities, respectively. (e) Potential experiment for verifying topological robustness to disorder and immunity to back-scattering with pumped-BLS. (f) Tunable topological phononics based on heterostructures of deformable and immobile phononic crystals as in the work of Li *et al.* Ref. 67. Reproduced with permission from Nat. Commun. 9, 1370 (2018). Copyright 2018 Springer Nature. (g) Scheme for extracting the residual strain of nanomembranes from measurements of the dispersion of flexural acoustic waves (see Refs. 15, 48, and 68).

duced phase transitions like $MoSe_2$[73], $MoTe_2$[74] and WTe_2[75].

A. Topological nanophononics

The functionalities of phononic devices cannot be entirely explained by their vibrational density of states (VDOS) and a single experimental spectrum. For instance, topological phononics own their properties on reciprocal-space characteristics, e.g., double Dirac cones[76–78] and band-inversion[79,80]. Such features in momentum-space can be revealed through angle-resolved (momentum-resolved) BLS measurements. Moreover, it is not straightforward to conceive the reciprocal-space characteristics of topological phononics from their real-space morphology. Various factors, such as unavoidable structural imperfections, can lead to disorder and incoherent phonon transport[28]. In this aspect, BLS is again a useful tool for the characterization of topological photonics.

Studies of topological acoustics in the sub-GHz range employ loudspeakers to launch acoustic waves and microphones for detection. A similar approach can be adopted for the recently developed field of high frequency (GHz) topological nanophononics[79,81] with pumped-BLS (Fig. 2a). In this case, the "loudspeaker" and the "microphone" will be femtosecond laser pulses and μ-BLS, respectively. The μ-BLS measurements can provide the VDOS or the band structure with a broad or narrow collection angle, respectively (Fig. 3a right). The various phononic bands have a BLS cross-section that varies according to symmetry considerations. Thus, to extract as many bands as possible, it is necessary to employ various scattering geometries and polarizations. Despite being tedious, such measurements reveal the character (symmetry) of the various vibrational modes, which is an important factor for topological phase transitions such as band inversion at

interfaces[80,82].

A particularly interesting class of topological systems are the so-called higher-order topological crystals[82–84]. A 2D second-order topological crystal will host 2D bulk-states, 1D edge-states and 0D corner-states. Owning to their spatial resolution, μ-BLS and pumped-BLS can be used to map the VDOS and to reveal states with 1D and 0D confinement (Fig. 2b). Spatially resolved pumped-BLS can also be useful for studying acoustic Anderson localization[85]. Moreover, BLS can be used to prove one-way transport on 1D edge-states (Fig. 2c). The relative intensities of Stokes and anti-Stokes scattering is a direct manifestation of directional transport of acoustic waves[69]. The scenario depicted in Fig. 2c will lead to an asymmetric BLS spectrum as the one sketched in Fig. 2d (the same approach can be followed for acoustic diodes[86–88]). Additionally, topological edge-states show robustness to disorder and immunity against back-scattering. These aspects can be examined using pumped-BLS as in Fig. 2e.

In view of applications, a highly desired characteristic of topological phononics is tunability[80,89]. Li *et al.*[67] have synthesized tunable sub-GHz topological phononics by creating a heterostructure of immobile hard PnCs and mobile soft PnCs (Fig. 2f). Such functional combinations of hard and soft matter have also been explored in the field of GHz nanophononics. For instance, Wang *et al.*[90] have demonstrated thermally tunable anisotropic thermoelastic properties in hybrid Bragg stacks of mica-type nanosheets and polymer layers. We consider hard-soft heterostructures as a promising class of materials for tunable nanophononics and for the emerging field of topological phononics in the GHz range[91]. Within the spirit of pumped-BLS, the external stimulus for mobilizing the soft part can be focused laser light. The conversion of light into motion is a separate challenge that can be addressed with various types of soft-matter photoactuators[6]. Some of the authors have already worked in this direction. Recently, we demonstrated fast light-driven motion of bioinspired nanomembranes, while simultaneously probing their mechanical and thermal properties with BLS[92]. The combination of softness and flexibility with photothermal and photoactuative properties was made possible using ultrathin polydopamine with a layered lamellar-like microscopic structure[68,92]. In the case of membrane-like systems, the additional merit of BLS is the ability to extract the strain from the dispersion relationship of flexural acoustic waves[15,48,68] (Fig. 2g).

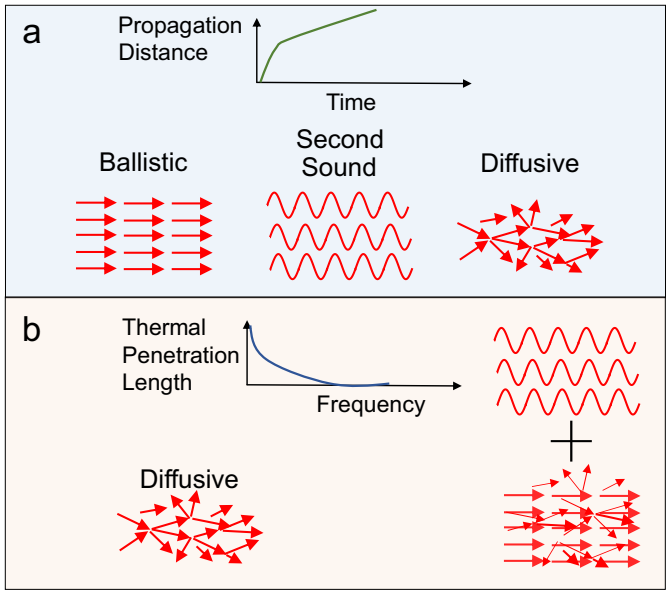
IV. THERMAL WAVES OR "SECOND SOUND"

The study of wave-like heat transport became a subject of mainstream research after the discovery of thermal waves in liquid He. At that time, superconductivity was also a flourishing research field, which triggered many studies to explain the origin of such waves. In fact, the physical similarities between superfluidity and superconductivity led, in some cases, to the development of similar theoretical models, e.g., the popular "two-fluid model". Wave-like heat transport was also later rediscovered in solid He,[93] which encouraged numerous researchers to search for the existence of thermal waves in other solid materials.

The theoretical foundations of second sound were set already in the 1960s by Guyer and Krumhansl,[95] Chester,[96] Hardy,[97] Enz,[98,99] and co-workers, who predicted the existence of different types of wave-like heat transport. Thermal waves of the "drifting" type were first predicted, originating from dominant Normal phonon-phonon scattering events. In addition, the existence of a "driftless" or "high-frequency" type, as well as "other types" of thermal waves, were also envisioned, as predicted by different solutions to the linearized Boltzmann transport equation (BTE). Interestingly, for the case of driftless or high-frequency thermal waves, it was theoretically found that the dominance of Normal scattering events is not a necessary condition. However, the key requirement is instead the slow decay of the energy flux. Figures 3a and 3b display a simplified schematic representation of the different types of thermal waves as observed in the time- or frequency-domain.

The first theoretical framework was recently re-interpreted in terms of hydrodynamic thermal transport through thermal collective excitations named after "relaxons", i.e., the thermal quasiparticles responsible for relaxing a thermal perturbation. In two recent publications,[100,101] Simoncelli, Ceppellotti and Marzari further developed the hydrodynamic formalism based on the framework initially proposed by Guyer and Krumhansl,[95] and Hardy,[97] i.e., redefining the problem in terms of relaxons and solving *ab initio* the linearized BTE. They have provided a valuable platform to study second sound by setting the solution of the BTE in terms of thermal quasiparticles, a linear combination of individual phonons, which describe heat propagation more realistically than through the individual phonon picture. In particular, they have shown that it is possible to derive the damped wave equation (hyperbolic heat equation) for the case of drifting second sound, namely when Normal scattering processes are the leading phonon-phonon scattering mechanism. The more complex case of driftless second sound or high-frequency second sound, i.e., when the slow decay of the heat flux is the key requirement, was recently theoretically studied by Beardo *et al.*[102] (see S6) from a macroscopic perspective. The authors proposed an *ansatz* for the solution of the linearized BTE, which takes into account the presence of a rapidly varying heat flux. Hence, they were able to derive an expression for the relaxation time of high-frequency thermal waves depending only on individual phonon properties, which can be obtained using well-established *ab initio* methods.

The experimental observation of second sound in solid materials is limited to a very small number of materials and experimental methods to date. Few techniques have been proven useful to detect the existence of thermal waves, among which are: (i) impulse experiments (time of flight), (ii) Brillouin scattering, (iii) thermal transient grating (TTG), and (iv) frequency-domain thermoreflectance. From the materials perspective, after thermal waves were experimentally demonstrated in solid He, substantial research efforts were invested to detect them in other solid materials. Disappointingly, the observation of thermal waves was only initially possible for solid He, NaF and Bi. After a gap of almost forty years (1972



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Material	Year	Velocity (m/s)	Method	T_{min} ≈(K)	T_{max} ≈(K)
solid He	1966	160	ToF	1	2
NaF	1969	2000	ToF	7.5	14
Bi	1972	780	ToF	2.5	4
$SrTiO_3$	2007	NA	Brillouin	6.4	40
graphite	2019	3200	TTG	50	150
Ge	2021	250	FDTR	7	300

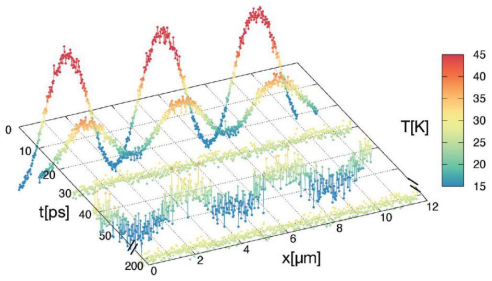

FIG. 3. Illustration of the different heat propagation regimes as observed in the time- and frequency-domain, on a hypothetic material which supports wave-like heat transport (second sound). (a) Time evolution of an instantaneously excited heat pulse. The propagation speed of the initially excited heat wavepacket decreases as time elapses due to the transition between different thermal regimes. After certain amount of time, Normal phonon scattering events (conserving energy and momentum) trigger wave-like heat transport. (b) Frequency response to a broadband thermal harmonic waveform. The thermal penetration depth drastically decreases for the higher frequencies. Wave-like transport is observed in the higher frequency range over a diffusive background. (c) Properties of thermal waves as experimentally observed in different materials, including the year when they were detected, their propagation velocity and detection method, as well as the temperature window where the thermal waves are observed [ToF: time of flight, TTG: thermal transient grating, FDTR: frequency-domain thermoreflectance]. (d) Extracted from Ref. 94. Numerical experiments using NEMD simulation the temporal response of Cumulene under a spatially harmonic-shaped thermal excitation resembling experiments done using TTG. C. Melis, G. Fugallo and L. Colombo, Phys. Chem. Chem. Phys., Vol. 23, 15275, 2021; licensed under a Creative Commons Attribution (CC BY) license.

to 2007) with no new experiments on second sound, in 2007 Koreeda *et al.* suggested the observation of thermal waves in $SrTiO_3$. Their work was based on Brillouin scattering experiments, which exhibited a singular broad band for temperatures below ≈50 K. The origin of this band was suggested to be of wave-like character based on a rather complex lineshape analysis of the Brillouin spectra, hence, providing an indirect demonstration of the existence of second sound in $SrTiO_3$. More recently, second sound was observed almost up to 150 K in highly oriented pyrolytic graphite by Huberman *et al.*[103] The authors have used thermal transient grating experiments to study the temporal evolution of sinusoidal temperature distribution on the surface of the samples. Interestingly, these observations were also theoretically confirmed using *ab initio* calculations by Simoncelli *et al.*[101] Interestingly, second sound has been recently suggested as possible explanation for the exceptionally high thermal conductivity of graphene and few-layer graphene[62,104].

High-frequency wave-like heat transport (so-called high-frequency second sound) was only recently experimentally demonstrated in Ge by Beardo *et al.*[102] The authors have shown that it is possible to unlock the propagation of thermal waves in natural bulk Ge by driving the system out of equilibrium with a rapidly varying harmonic temperature field. As the frequency of the thermal excitation increased towards the hundreds of MHz range, the observation of thermal waves gradually developed. It was shown that this approach is robust enough to expose wave-like heat transport independently, to a certain extent, on the phonon scattering rates of the studied material (Normal or Resistive), as well as on temperature. Figure 3c displays a summary of the materials where second sound was claimed to be observed to date, including the experimental method used for its detection, as well as some of its fundamental properties (velocity and temperature window for its observation).

Several recent numerical experiments have also provided valuable results to address the existence of second sound in conditions that can not be currently experimentally reproduced. To name a few, in a pioneer work MacDonald and Tsai[105] have shown the existence of thermal waves under a strong shock-wave using molecular dynamics (MD) in extreme pressure and temperature conditions. More recently, second sound was studied in low dimensional materials[106,107] and carbon nanotubes.[108] In particular, Melis *et al.*[94] addressed the existence of second sound in cumulene using non-equilibrium molecular dynamics. The authors have simulated a TTG experiment, showing how an initially excited harmonic temperature distribution exhibits a phase shift of π as time elapses. Figure 3d displays the spatio-temporal evolution of the temperature field in a similar fashion as it would be obtained in a TTG experiment. A phase shift of π is observed after ≈ 50 ps, where damping of the thermal wave is evidenced

by the significant signal decrease. Note that a phase shift of π in the temperature field can only be explained through the presence of thermal waves. In the case of a purely diffusive response, a monotonous decay of the initially excited harmonic thermal field towards steady-state equilibrium is expected. Finally, Zhang *et al.*[109] have recently extended the study of second sound to organic materials numerically showing its existence in polyacene $(\text{-}C_4H_2\text{-})_n$, polyacetylene $(\text{-}C_2H_2\text{-})_n$, and polyethylene $(\text{-}C_2H_4\text{-})_n$ bulk crystals.

V. CONCLUDING REMARKS

The mechanics and thermodynamics of materials are ultimately connected with their elastic properties. Basic solid-state physics and the advent of nanotechnology show that these elastic properties are generally anisotropic and size-dependent. All-optical techniques, particularly Brillouin light scattering, can be used to extract the components of the elasticity tensor and their size-dependent evolution in a remote, non-destructive way. Thus, BLS can be used to monitor the engineering of the acoustic properties of materials, for instance with chemical doping[110]. Acoustically engineered materials can then be tailored for various functionalities. As an example, lattice softening can be used to reduce the thermal conductivity and improve thermoelectric performance[111].

Recently, the BLS experimental setup has gained the capability to operate at any wavelength of the visible spectrum so that it can be easily adapted for a large variety of semiconducting and plasmonic nanomaterials. The underlying physics of optomechanical coupling is a separate fertile field with far-reaching implications for light-matter interactions. In addition to a spontaneous scattering of light from thermal waves, it is possible to combine Brillouin light scattering with femtosecond lasers to increase the population of phonons and thus their signal. The femtosecond laser pulses can also be used to prepare phononic wavepackets for testing novel concepts in nanophononics, e.g., one-way transport in topological edge states and acoustic Anderson localization.

Finally, the various activities (theory, experiments, and numerical experiments) on the detection of thermal waves in solid materials have gained substantial momentum in recent years. It is yet to be expected that through novel experimental concepts targeting small length scales, short timescales, or high-frequencies, the existence of second sound will probably be demonstrated in other materials. The question remains to which extent can we profit from this heat transport regime to better control heat propagation as an alternative to the diffusive regime described by Fourier's law.

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DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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