# Anomalous thermal transport under high pressure in boron arsenide

https://doi.org/10.1038/s41586-022-05381-x

Received: 22 October 2021

Accepted: 22 September 2022

Published online: 23 November 2022

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High pressure represents extreme environments and provides opportunities for materials discovery<sup>1-8</sup>. Thermal transport under high hydrostatic pressure has been investigated for more than 100 years and all measurements of crystals so far have indicated a monotonically increasing lattice thermal conductivity. Here we report in situ thermal transport measurements in the newly discovered semiconductor crystal boron arsenide, and observe an anomalous pressure dependence of the thermal conductivity. We use ultrafast optics, Raman spectroscopy and inelastic X-ray scattering measurements to examine the phonon bandstructure evolution of the optical and acoustic branches, as well as thermal conductivity under varied temperatures and pressures up to 32 gigapascals. Using atomistic theory, we attribute the anomalous high-pressure behaviour to competitive heat conduction channels from interactive high-order anharmonicity physics inherent to the unique phonon bandstructure. Our study verifies ab initio theory calculations and we show that the phonon dynamics-resulting from competing three-phonon and four-phonon scattering processes-are beyond those expected from classical models and seen in common materials. This work uses high-pressure spectroscopy combined with atomistic theory as a powerful approach to probe complex phonon physics and provide fundamental insights for understanding microscopic energy transport in materials of extreme properties.

Understanding materials properties under high pressure is critical to many areas, from planetary science, geophysics and condensed-matter theory, to new chemical bonding and superconductivity<sup>1-8</sup>. Over 100 years of literature studies of high pressure thermal transport, from the first experiment<sup>9</sup>, to additional analysis<sup>10-13</sup>, the lattice thermal conductivity of crystals has been found to increase monotonically with hydrostatic pressure, unless a phase transition takes place. From a theoretical perspective<sup>9,14</sup>, the crystal lattice symmetry remains under hydrostatic pressure, and therefore such a universal increase of thermal conductivity can be well understood as a result of volume shrink under pressure that leads to increased atom density and atomic bonding strength. Classical theories<sup>15-19</sup> use parameterized models to link with the Debye temperature, bulk modulus and Grüneisen parameters to predict that thermal conductivity must increase with pressure<sup>20</sup>. Although a quantum mechanical picture for this governing rule is missing, it has shown consistency with all experimental reports in the literature so far.

Here we measured high-pressure thermal transport in a recently discovered semiconductor material, cubic boron arsenide (BAs), and observed anomalous pressure dependence of the thermal conductivity that has not, to our knowledge, been measured in any other materials<sup>9–13,15–30</sup> (see summary in Fig. 1a). On the basis of our experimental characterizations, and in combination with ab initio atomistic

theory, we attribute this anomalous behaviour to competitive heat conduction channels that are inherent and arise from the unique bandstructure and high-order phonon physics. We verified ab initio calculations and the unique competition mechanisms from interactive high-order anharmonic scattering processes that are beyond understanding using classical theories.

### BAs with high thermal conductivity

To experimentally measure the high-pressure transport behaviour, we synthesized high-quality, single-crystal BAs and performed ultrafast pump-probe measurements under high pressure and temperature dependence. Cubic BAs (Fig. 1b) is a semiconductor that was recently discovered to have an ultrahigh thermal conductivity<sup>31-34</sup> beyond most common materials and demonstrates high promise for electronics thermal management<sup>35,36</sup>. Importantly, these recent theoretical studies indicate that BAs can be an exception and showcase material for higher-order phonon anharmonicity. Ab initio theory expects anomalous thermal transport at high pressure<sup>37</sup>; however, this has yet to be experimentally explored. Here we synthesized BAs crystals through chemical vapour deposition in a customized three-zone furnace as described in our recent reports<sup>32,38</sup>. The high crystal quality of BAs was carefully verified with structural characterizations (Fig. 1c) and

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**Fig. 1** | **Anomalous thermal conductivity of BAs under high pressure compared to literature studies. a**, Summary of thermal conductivities of non-metal crystals as a function of hydrostatic pressure (CsCl<sup>21</sup>, CsBr<sup>21</sup>, NaCl<sup>22</sup>, Nal<sup>22</sup>, quartz<sup>23</sup>, mica<sup>24</sup>, ice<sup>25</sup>, MgSiO<sub>3</sub><sup>26</sup>, diamond<sup>27</sup>, MgO<sup>28</sup>, BN<sup>29</sup>, GaAs<sup>30</sup>). Results

from existing literature indicate a ubiquitous increase in lattice thermal conductivity under a single phase, whereas BAs is an exception. **b**, Zinc blende crystal structure of cubic BAs. **c**, High-resolution transmission electron microscopy image showing atomically resolved lattices of BAs. Scale bar, 2 nm.

BAs exhibits two Raman peaks at 20.9 and 21.6 THz, which arise from

measurement of a room-temperature (298 K) thermal conductivity of approximately 1,300 W m<sup>-1</sup>K<sup>-1</sup> (ref. <sup>32</sup>). Our experimental set-up uses a diamond anvil cell (DAC) to provide high pressure, which was installed in a cryostat chamber for temperature control; pump–probe and optical measurements are applied to measure the sample through the optical window (Fig. 2a). The DAC can generate tens of GPa of pressure on the loaded BAs crystals, as shown in Fig. 2b. Importantly, we performed synchrotron X-ray diffraction measurements up to 32.6 GPa and verified that BAs crystal remains stable in the cubic phase and that there is no phase transition under all pressures. We use pump–probe measurements based on ultrafast optics (Fig. 2c) to perform in situ characterizations of the phonon properties under varied pressures and temperatures. The ultrafast set-up is configured for varied measurements, as discussed in the following.

#### Pressure-dependent phonon bandstructure

To understand the anomalous high-pressure behaviour of BAs, we characterize its pressure-dependent evolution of the phonon bandstructure. First, to examine the energy shift of optical phonons, we performed in situ Raman spectroscopy on BAs under pressure up to about 32 GPa. During Raman measurement, the energy of the incident light is absorbed by the crystal lattice and creates lattice vibrations as optical phonons; through energy transfer, the frequency of the scattering light is shifted. The peak position in the Raman spectra corresponds to the optical phonon frequency at the zone centre, owing to energy and momentum conservation. Our measured Raman spectra of BAs crystals under different pressures and the two-dimensional mapping result are shown in Fig. 3a,b, respectively, which clearly verify a shift of the Raman peaks towards higher frequency with increased pressure. Under ambient pressure, the two natural boron isotopes <sup>11</sup>B and <sup>10</sup>B (ref. <sup>32</sup>). When the pressure increases to approximately 32 GPa, the measurement shows that these two peaks shift to 24.8 and 26.3 THz, respectively. For comparison with the experimental results (symbols), we performed ab initio calculations (lines) of the Raman peak (Fig. 3c) and found good consistency for the optical band evolution: with the increase of pressure, the interatomic bonding becomes stronger, owing to the smaller interatomic distance, which pushes the atomic vibrations to higher frequencies. In addition, high pressure increases the energy splitting between longitudinal optical (LO) and transverse optical (TO) branches. As predicted by ab initio theory in Fig. 3c, we attribute this to LO-TO splitting due to the long-range Coulomb interaction that increases the frequencies of long-wavelength LO modes. Under high pressure, a greater number of electrons are attracted towards the boron atoms owing to the reduced interatomic distance, which leads to larger LO-TO splitting. The agreement between experiments (symbols) and ab initio calculations (lines) verifies that the optical phonons shift to higher energy and split, indicating an increased phonon bandgap under high pressure.

Next, to examine the energy shift of the acoustic phonon branches of BAs, we performed picosecond laser ultrasonic (PLU) and Brillouin scattering measurements under high pressure and verified the phonon hardening effect. In our PLU experiment<sup>39</sup> (Fig. 3d), a pump laser generates a picosecond acoustic pulse and the acoustic wave propagates inside the BAs sample at the speed of sound (that is, the acoustic phonon velocity). When the acoustic wave reaches the interfaces of the BAs sample, the wave is partially reflected and creates sound echoes. The echoes are then detected by the following probe laser when returning to the surface. The delay between the pump and probe pulse is the interval time ( $\Delta t_{echo}$ ) for a round trip of the acoustic wave inside the BAs



**Fig. 2** | **Experimental set-up for in situ ultrafast optical measurements under high pressure. a**, Schematic of DAC loaded with the sample for pumpprobe measurements. **b**, BAs crystal in the DAC sample chamber from top view. Scale bar is 100 μm. **c**, Schematic of ultrafast pump-probe optics set-up,

configurable as picosecond laser ultrasonic (PLU), Brillouin scattering, and time-domain thermoreflectance experiments. EOM, electro-optic modulator; BIBO, bismuth borate crystal.

sample with a thickness d. Therefore, the acoustic phonon velocity (v) is calculated by

$$v = 2d/\Delta t_{\rm echo} \tag{1}$$

Figure 3e shows our measurement data of a two-dimensional mapping of sound echoes under varied pressures and ultrafast time dependence with picosecond resolution. The two echoes are represented as a high-amplitude signal (vellow). The first echo corresponds to reflection by the interface between the top surface of BAs crystal and a thin-film metal transducer (deposited on top of BAs)<sup>39</sup>. The second echo corresponds to reflection by the bottom surface of BAs (Fig. 3d);  $\Delta t_{echo}$  represents the time interval between these two echoes. As shown in Fig. 3e, a clear reduction of  $\Delta t_{echo}$ , and thus an increase of the sound velocity, is measured with increased pressure. We have also measured the Brillouin frequencies under pressures that provide consistent sound velocities. The sound velocity is measured as 8,150.0 m s<sup>-1</sup> (longitudinal phonons along the <111> direction) under ambient pressure, and increases to 9,209.4 m s<sup>-1</sup> at 29.3 GPa. Quantitatively, the pressure-dependent sound velocity from our PLU measurement (squares) and Brillouin scattering measurement (triangles) are plotted in Fig. 3f, together with our ab initio calculations (line). The experimental and ab initio theoretical results are in good agreement, verifying the phonon hardening and evolution of acoustic phonon branches under high pressure.

To directly measure the evolution of the phonon bandstructure, we performed an inelastic X-ray scattering (IXS) experiment on BAs under high pressure. The IXS spectrum across the reciprocal vector space is characterized by scattering peaks that determine the corresponding phonon energy. In Fig. 4a, IXS experimental results (circles) of BAs phonon dispersion under hydrostatic pressures (ambient pressure, 18 GPa, and 32 GPa), are plotted together with the Raman measurement at the centre of the Brillouin zone (triangles), which clearly verify the ab initio calculation (lines). Fundamentally, the phonon bandstructure of BAs evolves under increased pressure with several key features (Fig. 4a): (i) Optical phonon branches move up resulting in an increased bandgap. For example, the optical phonon frequency at the centre of the Brillouin zone ( $\Gamma$  point) increases from 20.9 to 24.8 THz, and the energy gap between the acoustic and optical phonon branches (X point) is measured to increase from 9.2 to 12.8 THz with higher pressure. (ii) For acoustic branches, phonon hardening with increased acoustic sound velocity is expected under pressure, thus expanding their total energy space. For example, the longitudinal acoustic velocity has a substantial increase, measured from 8,150.0 to 9,277.1 m s<sup>-1</sup>, consistent with theory, as shown in Fig. 4a. This leads to a larger energy difference between different acoustic branches<sup>31</sup> under higher pressure: the longitudinal acoustic phonon frequency (K point) is measured to increase from 8.8 to 10.1 THz with higher pressure.

#### Competition of three- and four-phonon processes

Such phonon bandstructure evolution under high pressure will affect all heat conduction channels that contribute to the thermal conductivity, but their pressure-dependent effects differ, owing to varied fundamental phonon processes. By modern atomistic theory, thermal energy in semiconductors and insulators is mainly carried by lattice vibrations and thermal conductivity is calculated based on scattering between different phonon modes, that is, anharmonicity<sup>14</sup>. For each scattering event, multiple phonons can participate, and the three-phonon processes represent the lowest-order anharmonicity (Fig. 4b). For the past several decades, the thermal conductivity of solids has been usually considered in terms of the three-phonon scattering processes; the effects of higher-order anharmonicity such as the four-phonon processes were found to be negligible for most materials, and their low scattering probability was assumed by past literature according to the momentum and energy conservation requirements<sup>14,40,41</sup>. However, owing to the unique phonon bandstructure of BAs, both three- and



**Fig. 3** | **Experimental measurements and ab initio calculations for the pressure-dependent phonon bandstructure evolution. a**, Raman spectra of BAs measured at varied pressures from ambient to 32 GPa. **b**, Two-dimensional mapping of the pressure-dependent Raman data of BAs. **c**, Experimental data of the Raman peak frequencies in comparison with ab initio calculations, as a function of pressure. **d**, Schematic of picosecond laser ultrasonic (PLU) measurement. **e**, Two-dimensional mapping of the pressure-dependent ultrasonic data along the <111> direction of BAs. **f**, Experimental data of the longitudinal sound velocity of BAs by PLU and Brillouin scattering measurements, in comparison with ab initio calculations, as a function of pressure.

four-phonon processes can be important at room temperature and make interactive contributions to thermal conductivity<sup>37,42,43</sup>. We also note that BAs has a unique large bandgap (approximately 10 THz at ambient pressure and further increases at higher pressure) between the acoustic (a) and optical (o) phonon branches. Such a large a–o bandgap makes optical phonons inaccessible for most three-phonon processes<sup>31</sup>, but critically involved for most four-phonon processes, as dictated by the energy and momentum conversation laws.

Now a paradox exists in the consideration of high-pressure effects on thermal conductivity, owing to contributions from these competitive conduction channels. On one hand, for the three-phonon processes only: high pressure increases the energy separation (expands the total energy spreading) of the acoustic branches, and so ramps up the scattering phase space and thus increases the probability for three-phonon processes (that is, exclusive aaa processes in BAs)<sup>31</sup>. On the other hand, in consideration of the four-phonon processes only: high pressure increases the a-o bandgap and suppresses four-phonon scattering, which mostly involves optical phonons (for example, aaoo or aaao processes), as energy matching across a larger a-o bandgap becomes more challenging. Together, the effects of high pressure on BAs crystals affect different conduction channels in an interactive and competitive way, which is very different from common materials where only the three-phonon processes dominate (four-phonon processes are negligible) and the a-o bandgap is small. Here we consider high pressure as a tuning knob to study such interactions: in BAs, the strength of these different processes should vary with pressure-that is, for different pressure ranges, the dominant contribution channel to thermal conductivity varies. Therefore, with the unique competition mechanisms between three-phonon and four-phonon processes, ab initio theory serves as a powerful tool with which to capture the details of phonon scattering and predicts that the evolution under pressure can lead to anomalous pressure dependence of thermal conductivity<sup>37,43</sup>.

#### Pressure and temperature dependence

To quantitatively verify such interactive behaviours, we measured the thermal conductivity of BAs crystals as a function of pressure and temperature and compared the results with ab initio theory calculations. For thermal conductivity measurement, the ultrafast set-up is configured as time-domain thermoreflectance (TDTR)<sup>32,35</sup>. TDTR is well suited for measuring samples in a DAC under high pressure as no physical contact with the sample is required and the measurement can provide high spatial resolution, down to the micrometre size. The thermal conductivity of BAs at varied pressures from the ambient to approximately 30 GPa and temperatures were carefully measured. Figure 4c shows typical experimental data in which the ultrafast signal (that is, the time-dependent phase decay) was fitted to the thermal diffusive model to determine the thermal conductivity<sup>32,35</sup>. Our experimental results are presented in Fig. 4d and indeed show an anomalous non-monotonic trend with dependence on both pressure and temperature. At 300 K, the measurement shows that the thermal conductivity first monotonically increases from around 1,300 W m<sup>-1</sup> K<sup>-1</sup> at ambient pressure to approximately 1,500 W m<sup>-1</sup> K<sup>-1</sup> at 16.5 GPa, indicating that in this pressure range, the effect of the increased a-o bandgap on suppressing four-phonon processes dominates over the separation of acoustic branches on increasing three-phonon processes. From 16.5 to 29.3 GPa, the thermal conductivity of BAs is measured to monotonically decrease, indicating that in this pressure range, the four-phonon processes are weak and further suppressed by an increased a-o bandgap, whereas the increased energy separation of different acoustic branches enhances the three-phonon processes to dominate the thermal conductivity trend. We note that such non-monotonic pressure dependence exists for our measurement results at all different temperatures (Fig. 4d), and the transition pressure (that is, for local maximum conductivity) shifts to a lower pressure (from 16.5 to 7.0 GPa) when temperature is reduced from 300 to 200 K. This is because



Fig. 4 | Experimental measurements and ab initio theory for phonon dispersion and thermal conductivity as a function of pressure and temperature. a, Experimentally measured phonon dispersion from inelastic X-ray scattering (IXS; circles) and Raman spectroscopy (triangles) overlaid on the ab initio calculations (lines) under hydrostatic pressures of ambient, 18 GPa and 32 GPa. 3ph and 4ph indicate three-phonon and four-phonon processes, respectively. **b**, Phonon scattering physics: schematic examples for threephonon scattering (combination process) and four-phonon scattering (redistribution process). **c**, Typical time-domain thermoreflectance data:

thermal reflectance phase signal versus time for BAs under ambient pressure (black circles), 8.5 GPa (red circles) and 16.5 GPa (blue circles), fitted by thermal transport model (solid lines). To illustrate the measurement accuracy, the dashed lines indicate  $\pm 10\%$  changes in the thermal conductivity values. **d**, Measurement results of thermal conductivity for varied pressures (0–30 GPa) and temperatures (200–300 K). Dashed lines are theoretical results from ab initio calculations. The shaded background regions represent measurement uncertainty.

at low temperature a greater number of phonons occupy lower energy states, making it less possible to access high-energy optical phonons across the a-o bandgap, which suppresses the four-phonon processes and the trend of the pressure dependence becomes determined by the three-phonon processes. Indeed, the experimental results (symbols) agree well with ab initio theory (dashed line; Fig. 4d), and clearly verify that the atomistic origin of such anomalous pressure-dependent thermal conductivity results from the unique competition between three-phonon and four-phonon high-order anharmonic processes<sup>37</sup>.

#### **Conclusions and outlook**

Our study shows that the general rule of monotonic pressure dependence fails when the lowest-order interactions no longer dominate in energy transport—and these phase spaces could emerge under extreme conditions for many other materials systems. Therefore, we expect this study not only provide a benchmark result to revise current understanding, but also could impact established modelling predictions for extreme conditions such as the Earth's interior when direct measurements are not possible, or standard techniques such as shockwave studies when reliable modelling of the window materials is critical. In addition, the discovery of novel materials with non-monotonic thermal conductivity bandwidth could enable innovative design for pressure-adapted thermal windows or thermal switches to control energy flow under high pressure.

In summary, we have reported the observation of anomalous pressure dependent thermal conductivity that breaks the generic rule for highpressure heat conduction established over the past 100 years. Our study developed high-pressure transport experiments in combination with ab initio theory to provide microscopic understanding over the complex phonon physics. We measured the evolution of optical and acoustic phonon bands and thermal transport of BAs using Raman spectroscopy, PLU, Brillouin scattering, inelastic X-ray scattering and TDTR techniques up to 32 GPa under varied temperatures. Our experimental results show good agreement with ab initio calculations and revealed competing contributions to BAs thermal conductivity from the interactive three-phonon and four-phonon processes. This study provides fundamental insights into understanding microscopic energy transport in materials of extreme properties and opens up new opportunities in designing novel structures.

#### **Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information,

acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-022-05381-x.

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#### Methods

#### Chemical synthesis of cubic BAs crystals

Cubic BAs crystals were synthesized by the chemical vapour transport method. High-purity boron and arsenic powders (99.9999%, Alfa Aesar) were ground using a mortar and pestle with a stoichiometric ratio of 1:2. The reaction sources were introduced into a quartz tube followed by evacuating under high vacuum ( $10^{-5}$  torr) before flame-sealing. The quartz tube was placed into a customized three-zone reaction furnace with a 1,083 K hot zone, 1,058 K centre zone and a 1,033 K cold zone for the growth, before slowly cooling down to room temperature. Further details on the chemical synthesis of BAs can be found in our recent reports<sup>32,38</sup>.

#### Structural characterization of BAs

Transmission electron microscope (TEM) samples were prepared using a focused ion beam (FIB) machine (Nova 600, FEI) before transferred to the TEM holder (PELCO FIB Lift-Out, Ted Pella). The high-angle annular dark-field (HAADF) image was obtained by using aberration-corrected high-resolution scanning TEM (Grand ARM, JEOL, 300 kV). The atomicresolution TEM image was processed with Gatan software. The optical image of BAs sample inside the high-pressure DAC set-up was obtained by the CCD-camera mounted on an optical microscope (Leica DM 4000M) with a 5× objective lens.

#### High-pressure set-up

A piston-cylinder-type DAC was used to generate ultrahigh pressure. BAs samples were loaded into the DAC with 400- $\mu$ m diameter culet diamonds during the study. Rhenium gaskets were pre-indented to a thickness of roughly 60  $\mu$ m followed by an approximately 180- $\mu$ m-diameter aperture drilled at the centre. Silicone oil was used as the pressure-transmitting medium to apply the hydrostatic pressure uniformly to the sample. Ruby spheres were used to carefully calibrate and determine the pressure using its R<sub>1</sub> fluorescence peaks<sup>44</sup> with a pressure uncertainty less than 0.2 GPa. All the measurements including Raman spectroscopies, inelastic X-ray scattering, synchrotron X-ray diffraction, and thermal and phonon transport measurements were performed on samples inside the DAC.

#### Raman spectroscopy

Raman measurements were performed under high pressure using a confocal micro-Raman system (inVia, Renishaw) equipped with 633-nm laser excitation and 1,200 mm<sup>-1</sup> grating. The polarized laser has back-scattering geometry with a Leica DM2500 optical system. A  $\times$ 50/0.75 objective lens was used with lateral spatial resolution of 0.5 µm. For surface-enhanced Raman measurement, gold nanoparticles (-5 nm) were used on the BAs samples to enhance the signal resolution.

#### Synchrotron X-ray experiments

Synchrotron X-ray experiments were performed at the Advanced Light Source (ALS) at Lawrence Berkeley National Laboratory and the Advanced Photon Source (APS) at Argonne National Laboratory to understand the crystal structure and phonon dispersion of BAs under high pressure.

Pressure-dependent X-ray diffraction (XRD) experiments were performed to verify the crystal phase structure of BAs on the beamline 12.2.2 of the Advanced Light Source at Lawrence Berkeley National Laboratory, with a focused monochromatic X-ray beam at 30 keV (ref. <sup>45</sup>). The synchrotron XRD measurements from ambient pressure to 32.6 GPa (Extended Data Fig. 1a) show that the diffraction peaks were identified and indexed to the cubic diffraction facets of the BAs crystal and no additional or disappearing peaks are observed, verifying the single cubic phase for all the pressures. The pressure-dependent lattice constant of BAs was determined from our synchrotron XRD measurements and is shown in Extended Data Fig. 1b (circles), in good agreement with ab initio theory (line). In addition, single-crystal XRD experiments (Extended Data Fig. 2) were conducted at both ALS and APS.

Pressure-dependent IXS measurements were conducted to directly measure the phonon dispersion of BAs. The IXS measurements were performed on beamline 30-ID of the Advanced Photon Source at Argonne National Laboratory<sup>46,47</sup>. The experiment was carried out under a transmission geometry with a high energy resolution at ~1.5 meV by the monochromatic synchrotron-based IXS technique with the wavelength of the incident X-ray at 23.7 keV. The size of the beam was 10 µm in diameter. Helium gas was used as the pressure-transmitting medium<sup>48</sup>. The IXS spectrum is collected at varied O-points characterized by scattering peaks that determine the corresponding phonon energy (Extended Data Fig. 3). The energy gap between the optic and acoustic branches is directly measured and shows a monotonic increase from 9.2 THz to 12.8 THz when the pressure increases from ambient to 32 GPa (Fig. 4a). The IXS measurements verify the ab initio theory prediction of the phonon bandstructure evolution and the single cubic structure of BAs under high pressure.

#### In situ thermal and phonon transport measurements

Thermal and phonon transport experiments were performed in situ under varied high pressures using ultrafast optical techniques, including TDTR, PLU and Brillouin scattering.

The TDTR technique is a non-contact optical method to measure thermal conductivity<sup>32,35,49-53</sup>. A mode-locked Ti:sapphire laser with a repetition rate of 80 MHz generates ultrafast femtosecond pulses and is split into a pump and a probe beam. Using a second harmonic generator, the pump beam is fixed at 400 nm by doubling its frequency, and the probe beam stays at 800 nm. A thin metal film is coated on the sample surface, which absorbs the pump laser energy to heat up the sample and generate an instantaneous temperature rise. A mechanical delay stage is used to change the arrival time of the probe beam. The surface temperature is detected by the probe beam. The transient temperature decay is continuously recorded versus the delay time between pump and probe with a subpicosecond temporal resolution. The temperature decay is fitted with a thermal model to determine the thermal conductivity of the sample. We have used TDTR to measure thermal conductivity of a wide range of materials including BAs<sup>32,35,49-53</sup>.

The PLU technique<sup>39</sup> uses a pump beam to generate acoustic pulses propagating from the surface inside the sample and a probe beam to measure the round-trip time of the acoustic pulses across the sample. Sound echoes are detected and used to obtain sound velocity under varied high pressures.

Ultrafast pump-probe optics was applied to measure the Brillouin frequency following literature settings<sup>54-59</sup>: the pump pulses launch a strain wave that propagates inside BAs, and the probe pulse is partially reflected at the acoustic strain owing to the local change of refractive index (Brillouin scattering) and measured as a function of delay time. The time-dependent probe signal carries Brillouin scattering oscillations (Extended Data Fig. 4) due to constructive or destructive interference with the travelling strain pulse, and the Brillouin frequency is determined through fast Fourier transform analysis.

#### Ab initio theory for lattice dynamics

More calculation details for ab initio phonon theory<sup>27,31,32,39,42,43,60-64</sup> and consistent calculation results<sup>37</sup> can be found in the recent papers. The phonon dispersion, sound velocity and lattice thermal conductivity are calculated using ab initio methods based on density functional theory. The phonon dispersion is determined through diagonalization of the dynamical matrix and the thermal conductivity is determined by solving the phonon Boltzmann transport equation through self-consistent iterations.

The lattice vibrations are determined by the interatomic potential. The interatomic potential U can be expanded with respect to atomic displacement.

$$U = U_{0} + \frac{1}{2} \sum_{\{l,b,\mu\}} \Phi_{\mu_{l}\mu_{2}}(l_{l}b_{1}; l_{2}b_{2})u_{\mu_{1}}(l_{l}b_{1})u_{\mu_{2}}(l_{2}b_{2})$$

$$+ \frac{1}{3!} \sum_{\{l,b,\mu\}} \Phi_{\mu_{1}\mu_{2}\mu_{3}}(l_{l}b_{1}; l_{2}b_{2}; l_{3}b_{3})u_{\mu_{1}}(l_{l}b_{1})u_{\mu_{2}}(l_{2}b_{2})u_{\mu_{3}}(l_{3}b_{3})$$

$$+ \frac{1}{4!} \sum_{\{l,b,\mu\}} \Phi_{\mu_{1}\mu_{2}\mu_{3}\mu_{4}}(l_{l}b_{1}; l_{2}b_{2}; l_{3}b_{3}; l_{4}b_{4})u_{\mu_{1}}(l_{1}b_{1})u_{\mu_{2}}(l_{2}b_{2})$$

$$u_{\mu_{3}}(l_{3}b_{3})u_{\mu_{4}}(l_{4}b_{4})$$

$$+ \dots$$

$$(2)$$

where  $U_0$  is the equilibrium potential and the summation is performed over all numbered indices.  $u_{\mu}(lb)$  denotes the atomic displacement of the *b*th atom in the *l*th unit cell from its equilibrium position along the  $\mu$  direction. The first-order derivatives are zero as they are calculated at equilibrium.  $\phi_{\mu_1\mu_2}(l_1b_1; l_2b_2)$  are the second-order interatomic force constants (IFCs). From the second-order IFCs, the phonon frequency of given wave vectors **q** can be calculated by diagonalizing the dynamical matrix, and the group velocity of the phonon mode  $\lambda$  can be calculated from  $\mathbf{v}_{\lambda} = \partial \omega_{\lambda}/\partial \mathbf{q}$ . The sound velocity is the group velocity at the Brillouin zone centre.  $\phi_{\mu_1\mu_2\mu_3}(l_1b_1; l_2b_2; l_3b_3)$  and  $\phi_{\mu_1\mu_2\mu_3\mu_4}(l_1b_1; l_2b_2; l_3b_3; l_4b_4)$  are the third- and fourth-order IFCs, respectively, which are used to determine the three- and four-phonon scattering rates, respectively.

The lattice thermal conductivity tensor is given as

$$\kappa^{\alpha\beta} = \frac{1}{N} \sum_{\lambda} C_{\lambda} v_{\lambda}^{\alpha} F_{\lambda}^{\beta}$$
(3)

where  $\lambda = (\mathbf{q}, s)$  labels a phonon mode with wavevector  $\mathbf{q}$  and polarization s.  $\alpha$  and  $\beta$  represent the direction in the Cartesian coordinate system. N is the number of **q**-points in the mesh of the Brillouin zone.  $C_{\lambda}$  and  $v_{\lambda}^{\alpha}$ are the volumetric specific heat and the group velocity along the  $\alpha$  direction of phonon mode  $\lambda$ , respectively.  $F_{\lambda}^{\alpha}$  measures the deviation of the phonon distribution  $(n_{\lambda})$  from the equilibrium  $(n_{\lambda}^{0})$  as driven by the temperature gradient  $\nabla T$ ,  $n_{\lambda} = n_{\lambda}^{0} + (-\partial n_{\lambda}^{0}/\partial T)\mathbf{F}_{\lambda} \cdot \nabla T$ . For most materials, the deviation of  $n_{\lambda}$  will not be large, and so the scattering rates  $\tau_{\lambda}^{-1}$ for each individual phonon mode  $\lambda$  can be calculated by keeping background phonons in equilibrium, and in this case  $\mathbf{F}_{\lambda} = \tau_{\lambda} \mathbf{v}_{\lambda}$ . But for high thermal conductivity materials. n<sub>1</sub> is usually driven far away from equilibrium by temperature gradient, so the deviation of  $n_1$  for all the phonon modes should be simultaneously considered by determining  $\mathbf{F}_{\lambda}$ from the Boltzmann transport equation through self-consistent iteration. In the Boltzmann transport equation, a phonon flux is driven by temperature gradient and balanced by phonon scatterings,

$$\mathbf{v}_{\lambda} \cdot \nabla T \frac{\partial n_{\lambda}}{\partial T} = \left(\frac{\partial n_{\lambda}}{\partial t}\right)_{\text{scattering}}$$
(4)

The scattering term on the right-hand side of equation (3) describes the rate of change of  $n_{\lambda}$  owing to phonon scatterings, and is the summation of different types of scattering processes,

$$\left(\frac{\partial n_{\lambda}}{\partial t}\right)_{\text{scattering}} = \left(\frac{\partial n_{\lambda}}{\partial t}\right)_{3\text{ph}} + \left(\frac{\partial n_{\lambda}}{\partial t}\right)_{4\text{ph}} + \left(\frac{\partial n_{\lambda}}{\partial t}\right)_{\text{iso}}$$
(5)

where  $\left(\frac{\partial n_{\lambda}}{\partial t}\right)_{3ph'}$ ,  $\left(\frac{\partial n_{\lambda}}{\partial t}\right)_{4ph}$  and  $\left(\frac{\partial n_{\lambda}}{\partial t}\right)_{iso}$  represent the rate of change of  $n_{\lambda}$  due to three-phonon, four-phonon and isotope scattering, respectively, and can be determined by quantum perturbation theory<sup>65</sup>. The only input of this ab initio approach is the IFCs calculated from density functional theory<sup>66,67</sup>.

The procedure to obtain the IFCs is as follows. First, an irreducible displacement set is generated based on a real-space 256-atom supercell.

For each displacement configuration, the forces acting on atoms were calculated by density functional theory using the Quantum ESPRESSO package<sup>68,69</sup>, and on that basis we build up force–displacement sets. Then the IFCs are extracted by fitting the displacement-force sets using the ALAMODE package<sup>70</sup>.

All the density functional theory calculations in this work are based on the projector-augmented wave approach. We use PZ pseudopotentials (B.pz-n-kjpaw\_psl.0.1.UPF and As.pz-n-kjpaw\_psl.0.2.UPF from http:// www.quantum-espresso.org) under a local density approximation to determine the IFCs and use PBEsol pseudopotentials (B.pbesol-n-kjpaw\_ psl.1.0.0.UPF and As.pbesol-n-kjpaw\_psl.1.0.0.UPF from http://www. quantum-espresso.org) under a generalized gradient approximation to determine the theoretical lattice constants in Extended Data Fig. 1b. The Monkhorst–Pack grids for electronic structure calculations are  $6 \times 6 \times 6$  and  $2 \times 2 \times 2$  for an eight-atom unit cell and 256-atom supercell, respectively. The convergence threshold for self-consistency is  $10^{-11}$ . The kinetic energy cut-offs for electronic wavefunctions are 120 Ry and 100 Ry for the PZ and PBEsol pseudopotentials, respectively.

#### **Data availability**

The data that support the plots within this paper and findings of this study are available from the corresponding author upon request.

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Acknowledgements We thank B. Kalkan, K. Armstrong and B. Lavina for technical help and discussion. Y.H. acknowledges support from a CAREER Award from the National Science Foundation (NSF) under grant no. DMR-1753393, an Alfred P. Sloan Research Fellowship under grant no. FG-2019-11788, and the Vernroy Makoto Watanabe Excellence in Research Award. This work used computational and storage services associated with the Hoffman 2 Shared Cluster provided by UCLA Institute for Digital Research and Education's Research Technology Group, and the Extreme Science and Engineering Discovery Environment (XSEDE), which is

supported by National Science Foundation grant number ACI-1548562. This research used resources at the US Department of Energy (DOE) Office of Science user facility, including the Advanced Photon Source by Argonne National Laboratory under contract no. DE-AC02-06CH11357 and the Advanced Light Source by Lawrence Berkeley National Laboratory under contract no. DE-AC02-05CH1123.

Author contributions Y.H. proposed and directed the research. S.L., Z.Q. and M.L. performed the experiments. H.W. performed the theory calculations. M.K. helped with the XRD study. A.A. helped with the IXS study. A.K. helped with technical discussions. The manuscript was prepared by S.L., Z.Q., H.W., M.L. and Y.H. with input from all co-authors.

Competing interests The authors declare no competing interests.

#### Additional information

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**Extended Data Fig. 1** | **Synchrotron X-ray diffraction (XRD) measurement of BAs and its pressure-dependent lattice constant. a**, Synchrotron XRD patterns of BAs measured at different pressures (-0-32.6 GPa), verifying the

cubic phase of BAs under all high pressures without phase transition. **b**, Experimentally measured pressure-dependent lattice constant of BAs (circles), in comparison with the ab initio calculation (line).



**Extended Data Fig. 2**|**Synchrotron single-crystal X-ray diffraction measurement.** Example diffraction image of BAs in which all peaks are indexed to the cubic phase.



**Extended Data Fig. 3 | Inelastic X-ray scattering (IXS) spectrum measurement of BAs crystal.** Shown in figure are scattering peaks that determine the phonon frequency for scattering vector *Q* at (3.52.52.5).



**Extended Data Fig. 4** | **Brillouin scattering oscillation signals measured on a BAs sample.** Fast Fourier transform result of Brillouin scattering data (inset) determines the Brillouin frequency.