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Advancing Thermal Management Technology for Power Semiconductors through Materials and Interface Engineering

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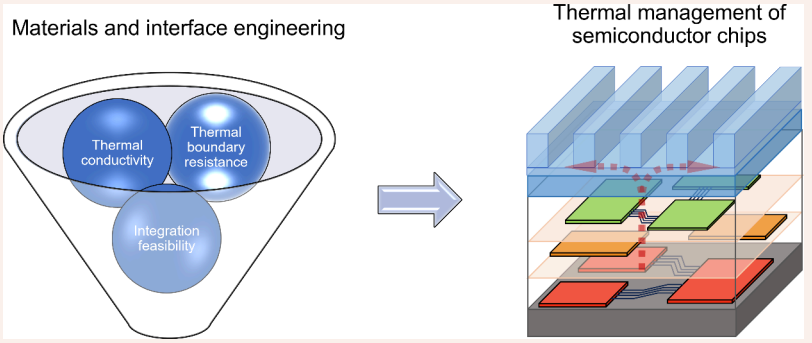

CONSPECTUS: Power semiconductors and chips are essential in modern electronics, driving applications from personal devices and data centers to energy technologies, vehicles, and Internet infrastructure. However, efficient heat dissipation remains a critical challenge, directly affecting their performance, reliability, and lifespan. High-power electronics based on wide- and ultrawide-bandgap semiconductors can exhibit power densities exceeding 10 kW/cm^2, hundreds of times higher than digital electronics, posing significant thermal management challenges. Addressing this issue requires advanced materials and interface engineering, alongside a comprehensive understanding of materials physics, chemistry, transport dynamics, and various electronic, thermal, and mechanical properties.

Despite progress in thermal management solutions, the complex interplay of phonons, electrons, and their interactions with material lattices, defects, boundaries, and interfaces presents persistent challenges. This Account highlights key advancements in thermal management for power semiconductors and chips, with a focus on our group's recent contributions. Our approach addresses several critical issues: (1) developing materials with ultrahigh thermal conductivity for enhanced heat dissipation, (2) reducing thermal boundary resistance between power semiconductors and emerging 2D materials, (3) improving thermal and mechanical contacts between chips and heat sinks, (4) innovating dynamic thermal management solutions, and (5) exploring novel principles of thermal transport and design for future technologies.

Our research philosophy integrates multiscale theoretical predictions with experimental validation to achieve a paradigm shift in thermal management. By leveraging first-principles calculations, the recent studies redefined traditional criteria for high-thermal-conductivity materials. Guided by these insights, we developed boron arsenide and boron phosphide, which exhibit record-high thermal conductivities of up to 1300 W/mK. Through phonon band structure engineering, we reduced TBR in GaN/BAs interfaces by over 8-fold compared to GaN/diamond interfaces. The combination of low TBR and high thermal conductivity significantly reduced hotspot temperatures, setting new benchmarks in thermal design for power electronics.

We further explored the anisotropic TBR properties of two-dimensional materials and Moiré patterns in twisted graphene, expanding the thermal design landscape. To address challenges at device−heat sink interfaces, we developed self-assembled boron arsenide composites with a thermal conductivity of 21 W/mK and exceptional mechanical compliance (∼100 kPa). These composites provide promising solutions for thermal management in flexible electronics and soft robotics.

In dynamic thermal management, we pioneered the concept of solid-state thermal transistors, enabling electrically controlled heat

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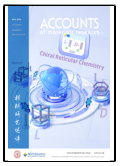

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flow with unparalleled tunability, speed, reliability, and compatibility with integrated circuit fabrication. These innovations not only enhance thermal performance but also enable the exploration of novel transport physics, improving our fundamental understanding of thermal energy transport under extreme conditions.
Looking forward, we reflect on remaining challenges and identify opportunities for further advancements. These include scaling up the production of high-performance materials, integrating thermal solutions with existing manufacturing processes, and uncovering new physics to inspire next-generation power electronics technologies. By addressing these challenges, we aim to inspire future codesign strategies that enable the development of more efficient, reliable, sustainable, and high-performance electronic systems.

1. INTRODUCTION

Efficient thermal management is crucial for electronic devices.[1] Heat accumulation creates hotspots that degrade reliability and performance. Studies have shown that for every 10 K increase in temperature, device lifetime diminishes by ∼50% (inset in Figure 1).[2] The growing power and heat flux density over recent decades highlights the urgent need for better heat dissipation. As depicted in Figure 1, the maximum hot-spot power has surged from less than 10 W/cm^2 in the 1970s to a projected nearly 1 MW/cm^2 in 2030.[1−9] Achieving the performance limit for wide and ultrawide bandgap devices, such as gallium nitride (GaN) systems, requires an 80% reduction in thermal resistance.[10]

Traditional cooling techniques have been widely explored, including fans, fins, heat pipes, and microfluidics.[11] However, a primary challenge remains in the near-junction region, where most Joule heat is generated, as depicted in Figure 2. The thermal resistance stems from (1) thermal conductivity of constituent materials and (2) thermal boundary resistance (TBR) across the interfaces of heterogeneous layers. The former has led research toward the integration of high-thermal-conductivity materials. The latter encompasses myriad factors such as near-interface defects, surface roughness, and the disruption of phonon coherence; these have been extensively detailed in our recent review.[1]

Our research develops new approaches toward advanced thermal management, emphasizing material designs. We begin by providing an overview of the recent development of benchmark high-thermal-conductivity materials, drawing insights from atomistic theories and spectroscopy experiments. Subsequently, we delve into interface engineering aimed at mitigating TBR. We discuss strategies such as phonon band structure alignment, bonding chemistry optimization, and effects from defects, crystal orientations, atomic bonding, etc. Furthermore, we illustrate the greatly enhanced cooling performance attained through our exemplary studies. Key developments include the direct integration of BAs with wide-bandgap materials and GaN high electron mobility transistor (HEMT), flexible thermal interface materials, and the solid-state thermal transistor for dynamical thermal control. Finally, we highlight the persistent challenges in electronic thermal management and identify prospective avenues for future research opportunities.

2. RECENT PROGRESS IN DEVELOPING NEW HIGH-THERMAL-CONDUCTIVITY MATERIALS

Effective heat dissipation in electronic devices relies on high-thermal-conductivity materials. Industrial standards like copper and SiC (∼400 W/mK) are widely used, but materials surpassing this benchmark are rare. Recently, the discovery and development of ultrahigh-thermal-conductivity materials has emerged as a research frontier essential for advancing next-generation thermal management technologies.

2.1. Conventional Models, First-Principles Atomistic Theory, and Machine Learning

Traditionally, diamond, cubic BN, graphite, graphene, and nanotubes were explored for high thermal conductivity. However, challenges like high costs, slow growth rates, additional grains and phases during growth, and fabrication difficulties have limited their adoption.[12−14] The practical utility of graphite, graphene, and nanotubes has been limited by the inherent anisotropy or degradation from ambient scattering.[15,16]

Classical models such as the Slack equation[17] have been used to derive thermal conductivity (κ) from metrics such as atomic mass ($\bar{M}$), lattice structure (n being the number of atoms in the primitive unit cell and δ being the lattice size), the Gruneisen parameter (γ), and other related factors, including the Debye temperature Θ.

$$\kappa = A\frac{\bar{M}\Theta^3\delta n^{1/3}}{\gamma^2 T} \quad (1)$$

Based on the classical models, κ above 400 W/mK is limited to diamond, cubic BN, and silicon carbide.[18] Later on, people noticed that these classical models oversimplify phonon behaviors and overlook the complicity of momentum- and energy-dependent phonon spectra like higher-order phonon anharmonicity. New guidelines for high thermal conductivity searching are needed.

During the recent decades, with the development of density functional theory (DFT) and computational hardware, the calculations of atomic interaction in solids have become feasible. Combining DFT calculations with Fermi's golden rule, the phonon Boltzmann transport equation can be solved without any empirical input. The κ of materials thus can be evaluated within an ab initio framework with an accurate consideration of the momentum- and energy-dependent phonon properties.[19] The use of atomistic theory has not only improved the fundamental understanding of phonon behaviors but also opened up a new era for discovering new materials. The atomistic theory, in combination with recent progress in machine learning,[20] further enables high-throughput thermal property predictions. The new development through a combination of experiments and advanced theory has been exemplified by our recent discovery of boron phosphide (BP), boron arsenide (BAs), and other new materials and properties,[7,21−26] as discussed below.

2.2. Boron Phosphide

Our recent study examined BP by synthesizing its single crystals and measuring its high room-temperature thermal conductivity, which exceeds that of all metals.[24] The high thermal conductivity is a result of its high-symmetry structure, strong interatomic bonding, and low anharmonicity.[22] As shown in Figure 3 a and b, the BP crystal resembles a zinc-blende structure in the $F\bar{4}3m$ space group. The room-temperature thermal conductivity of BP is ∼500 W/mK for natural abundance

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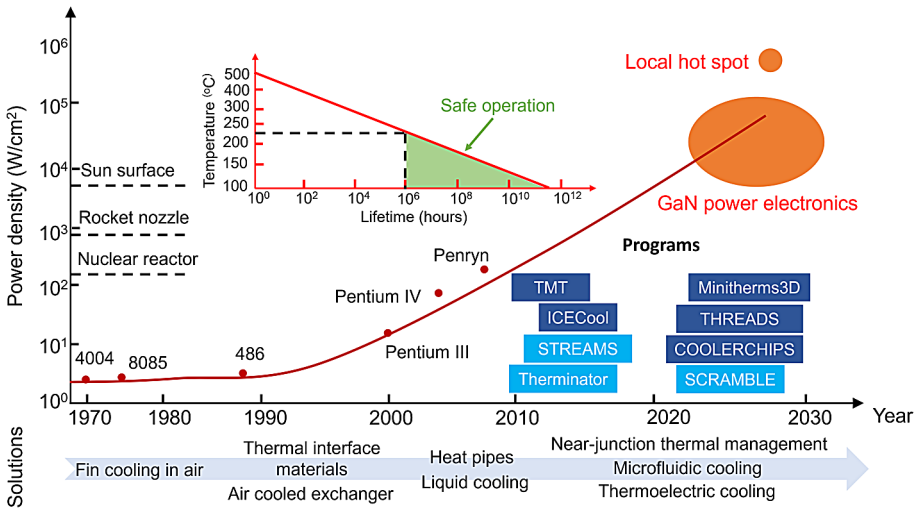

Figure 1. Thermal crisis history of electronics and the technology roadmap for resolving the challenges. The red data points indicate the power density increase from less than 10 W/cm^2 for the earliest central processing units to nearly 1 MW/cm^2 projected for local hot spots in GaN power electronics, which triggered many government-funded attempts, as exemplified by the US programs (dark blue boxes) and European Union programs (light blue boxes). The inset plot shows the strong dependence of GaN device lifetime on temperature derived by a program performer.[10]

samples and ~600 W/mK in isotope-enriched samples, as shown in Figure 3c and d, exceeding the industrial high thermal conductivity standard (copper and SiC). A strong temperature dependence was observed, indicating the dominant role of Umklapp scattering at high temperatures due to the minimum impurity in the samples. Moreover, the variance-reduced Monte Carlo simulation of the three-dimensional and phonon mode-dependent Boltzmann transport equation was developed to fully quantify the phonon transport behaviors, including the mean free path distributions with and without consideration of isotope effects, showing good agreement with our ballistic thermal transport measurements.[24]

2.3. Boron Arsenide

Our recent study discovered BAs, a new semiconductor with record-high thermal conductivity of 1300 W/mK, above those of all common semiconductors and metals.[4,7,27] BAs has a zinc-blende crystal structure, as verified by the high-resolution transmission electron microscopy image and other structural characterization techniques[27] (Figure 4a and b). The room-temperature thermal conductivity of BAs is over three times higher than that of the industrial standard (copper or SiC) and about twice that of cubic BN,[7,27−29] as shown in Figure 4c. To understand the fundamental origin of the high thermal conductivity in BAs, we performed a direct measurement of phonon mean free paths. We probed diffusive to ballistic thermal transport using an ultrafast pump−probe optical technique.[7,30,31] As depicted in Figure 4d, we found a pronounced heating size effect on the thermal conductivity of BAs. The reduction of κ is up to 50% with an excitation size of 2 μm, much larger than the other high-thermal-conductivity materials like diamond and BN, with only ~20% reduction of κ. This observation indicates that phonons with very long mean free paths (1−10 μm) contribute substantially to thermal transport in BAs. These studies established BAs as a benchmark material for thermal management of electronics and revised the high thermal conductivity search criteria by pointing out the importance of higher-order anharmonicity. Importantly, the basic physical propreties of BAs were characterized and reported as an experimental database,[27] such as refractive index, elastic modulus, and specific heat of BAs, to stimulate broad applications of this new material in electronics, optoelectronics, and photonics.[27] These new materials exhibit high thermal conductivity, along with additional advantages, such as isotropy, thermal spectral matching with prototype semiconductors like GaN and high thermal boundary conductance, comparable thermal expansion coefficients, and potential compatibility with semiconductor manufacturing processes, highlighting their promise for integration into chip technologies.[1]

2.4. Theoretical Identification of Future High-Thermal-Conductivity Materials

In addition to experimental efforts, theoretical identification of new high-thermal-conductivity candidate materials is critical to further extending the materials toolbox. Our recent study explored first-principles theory to search for new candidates and examined the lattice vibrational spectra of various materials. Considering the crystal structures and high-order anharmonicity associated with four-phonon processes,[25] the thermal conductivities of ternary compounds such as $BAsC_2$, BPC_2, and BNC_2 are found to be high for their multiple phases, with an exceptionally high room-temperature thermal conductivity reaching 2100 W/mK for isotope-pure crystals, as shown in Figure 5a. The fundamental origin of high thermal conductivity is attributed to reduced scattering phase space from the robust carbon−carbon bonds as well as high phonon group velocity. Due to this origin, the nanostructured forms of BNC_2 are expected to experience less scattering when applied for thermal management in nanoelectronics.

Advances in artificial intelligence (AI) also offer complementary opportunities for progress.[20] For instance, machine learning potentials have been utilized to approximate the accuracy of DFT while operating orders of magnitude faster. This enables large-scale simulations and access to previously inaccessible domains, such as GaN-BAs heterostructure,[5,32] and ballistic thermal transport in amorphous states.[33] Recent developments in thermodynamically consistent data-driven methods demonstrate how deep neural networks can manage uncertainties.[20,34] We expect that these modeling approaches including first-principles theory and AI-driven computation will greatly advance this field.

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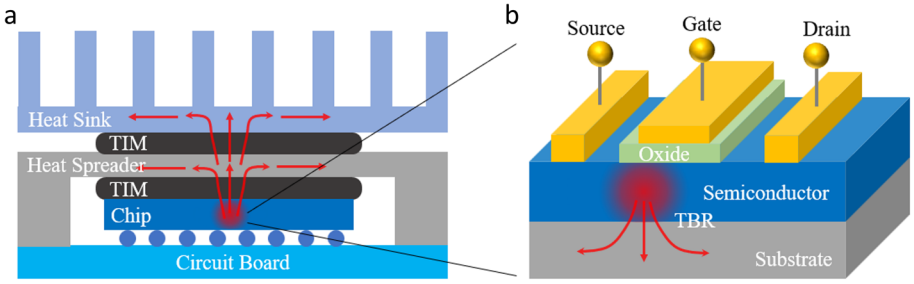

Figure 2. Thermal management design of electronics, including optimization of (a) heat sinks, heat spreaders, thermal interface materials, and others at the device or package level and (b) the thermal boundary resistance and cooling substrate at the near-junction region of transistors.

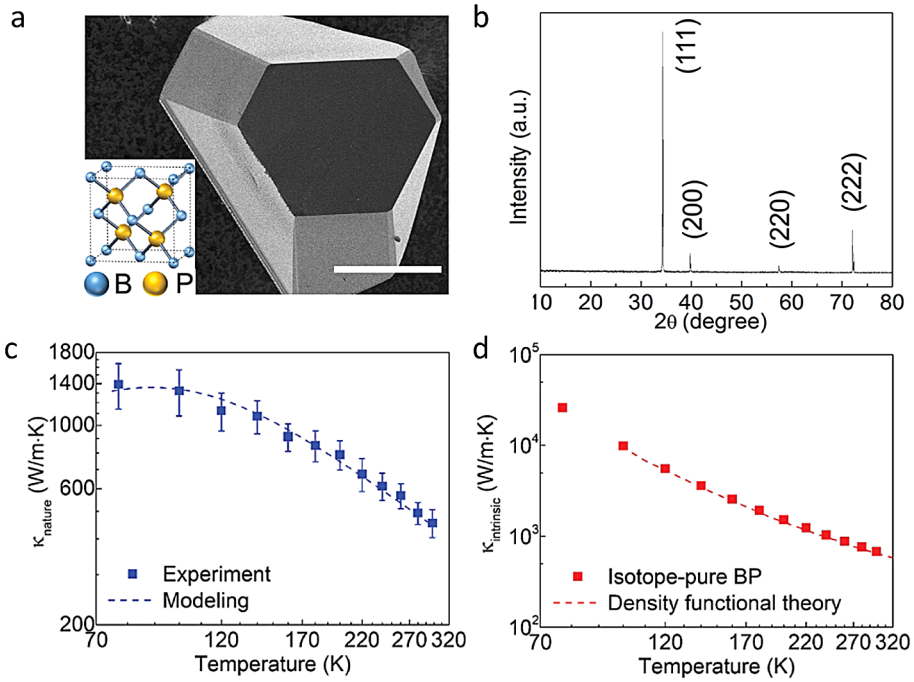

Figure 3. Material synthesis and structural and thermal characterization of the single crystal of boron phosphide (BP). The crystals are characterized with the (a) scanning electron microscope image and (b) powder X-ray diffraction patterns. The scale bar is 50 μm. (c) The experimentally measured temperature-dependent thermal conductivity of BP shows a reduction from ~1400 to 460 W/mK, captured by the modeling fit considering multiple phonon scattering processes. (d) The extracted intrinsic thermal conductivity of isotope-free BP decreases from 26000 to 1400 W/mK over a temperature range from 77 to 298 K, in comparison with the calculation results from DFT. Reproduced with permission from ref 24. Copyright 2017 American Chemical Society.

2.5. Two-Dimensional Materials

Besides bulk materials, 2D materials have attracted attention in thermal management field. Many literature have been focused on a studied available 2D structures such as graphene, MoS2, etc. To look for new 2D candidates, for instance, our recent study employed first-principles theory to determine the thermal properties of 2D boron compounds in varied configurations, like hexagonal form of h-BN, h-BP, h-BAs, and h-BSb. The predicted thermal conductivities span a range from around 160 to 1045 W/mK, as depicted in Figure 5b.[26] For monolayer h-BAs, although it has similar structural and dispersion characteristics to that of its bulk cubic crystals, the 2D form is found to have stronger phonon scattering due to the midfrequency ZO branch coupling with acoustic bunching. Further studies of 2D and 1D materials will help reveal phonon transport physics under confinement and assess their application potential for nanoscale devices and interfaces.

3. INTERFACE ENGINEERING FOR THERMAL BOUNDARY RESISTANCE

Thermal boundary resistance (TBR) significantly affects power dissipation rate and consequently device performance, reliability, and lifetime. TBR generally exists at the heterogeneous interfaces and has both intrinsic and extrinsic contributions, which are discussed in detail in our recent report.[1] Extrinsic factors include incomplete contact, defects, or disorder at the interface. Intrinsic contributions arise from phonon spectra mismatches between materials, leading to the discontinuity of phonon propagation and thus a temperature discontinuity at the interface, forming a TBR (shown in Figure 6a).[1,35] In the following, we will discuss our efforts in the investigation of the fundamental concept of TBR and contributions from key factors such as phonon density of states and crystal directions; further, we will discuss possible strategies for mitigating its impact on electronic systems.

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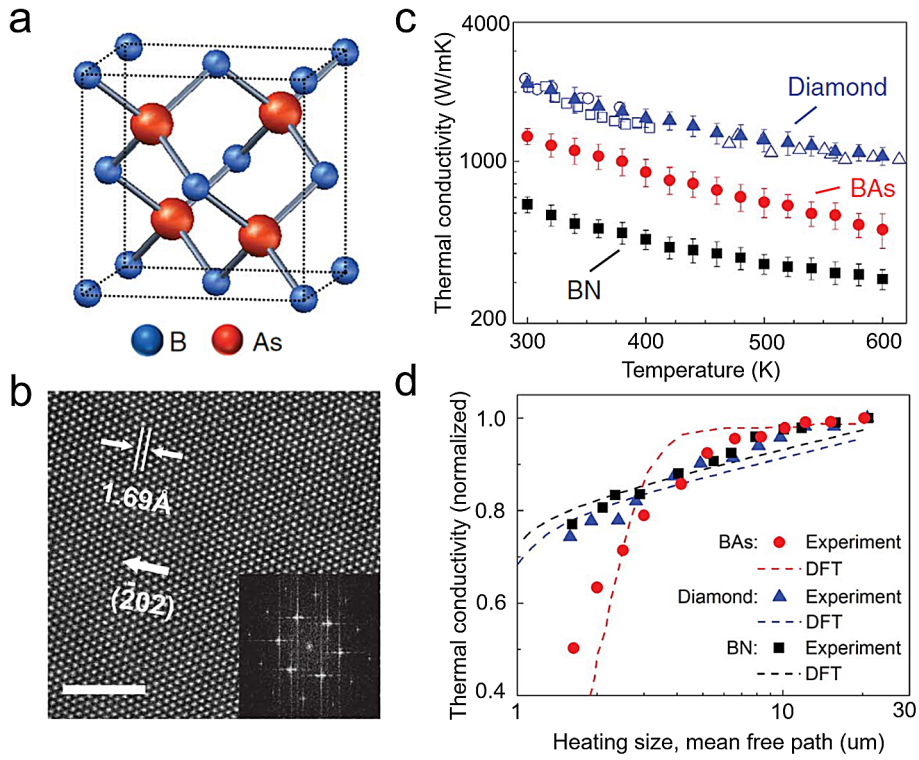

Figure 4. Experimental investigation of exceptional thermal conductivity and unusual phonon transport characteristics in boron arsenide (BAs). (a) Illustration of the zinc-blende crystal structure for cubic BAs. (b) High-resolution transmission electron microscope image displaying the BAs lattice structure and dimensions, with an inset diffraction pattern along the [111] zone axes. Scale bar: 2 nm. (c) Temperature-dependent thermal conductivity of BAs in the range of 300−600 K, benchmarked against diamond and cubic BN. (d) Phonon mean free path spectra for BAs from heater size-dependent thermal experiments, in comparison with theory. Reproduced with permission from ref 7. Copyright 2018 American Association for the Advancement of Science.

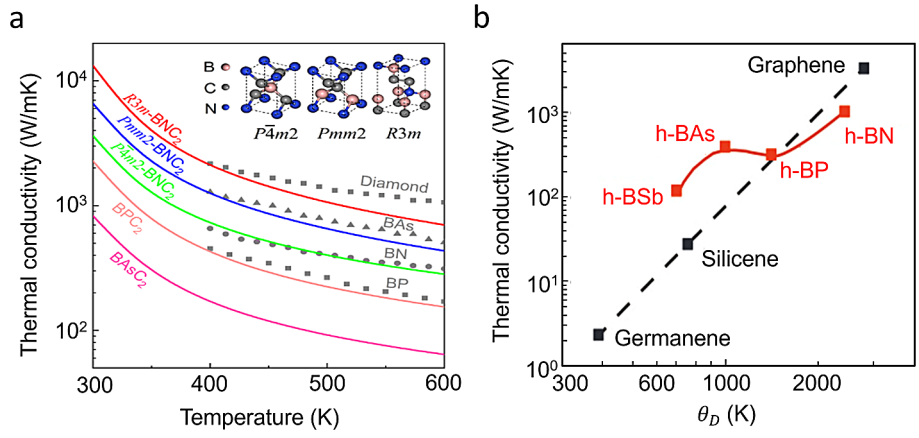

Figure 5. Exploration of high-thermal-conductivity materials among ternary and two-dimensional materials. (a) Temperature-dependent thermal conductivity of three different phases of BNC_2, BPC_2, and $BAsC_2$ in comparison to diamond, BAs, BN, and BP. (b) Thermal conductivity prediction of two-dimensional BN, BP, BAs, and BSb in comparison with graphene, silicene, and germanene. Reproduced with permission from refs 25 and 26. Copyright 2021 and 2019 American Physical Society.

3.1. Reducing TBR by Phonon Band Structure Engineering

Mathematically, TBR characterizes the temperature difference ΔT at the interface induced by heat flux q'' and is described as

$$\mathrm{TBR} = \frac{\Delta T}{q''} \tag{2}$$

A key factor affecting TBR is the overlap between phonon density of states. A useful rule of thumb for estimating TBR is to compare the Debye temperatures of materials, which provide phonon energy limits. Generally, a larger difference in Debye temperatures results in a higher TBR. For instance, as illustrated in Figure 6b,[5] the Debye temperature of diamond (~2000 K) is significantly higher than that of conventional semiconductors, leading to poor TBR and limiting its application in thermal management. In contrast, BAs and BP exhibit lower Debye temperatures than diamond and BN, suggesting a lower TBR. In the following, we use the example of the GaN-BAs interface for more discussions.

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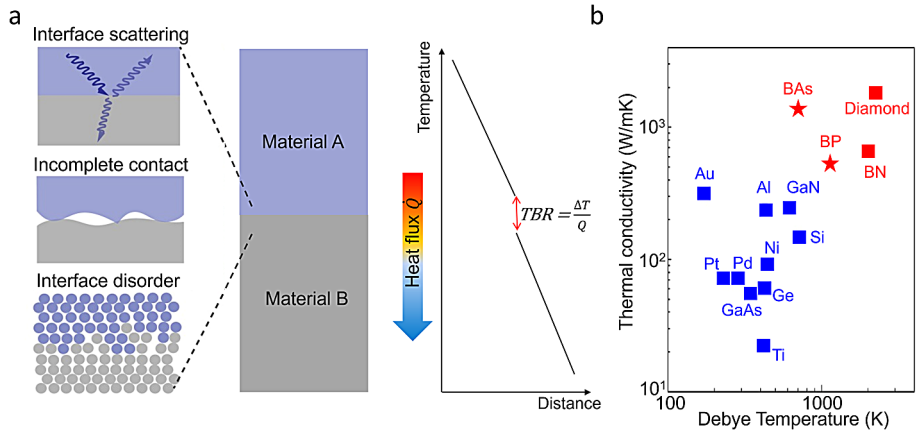

Figure 6. Interface thermal transport and thermal boundary resistance (TBR). (a) TBR, ascertained from the temperature discontinuity at the interface, can result from phonon scattering at the interface, incomplete contact, and interface disorder. (b) Debye temperatures of typical high-thermal-conductivity materials and materials commonly utilized in the semiconductor industry. Reproduced with permission from refs 1 and 5. Copyright 2020 and 2021 Royal Society of Chemistry and The Authors, under exclusive license to Springer Nature Limited.

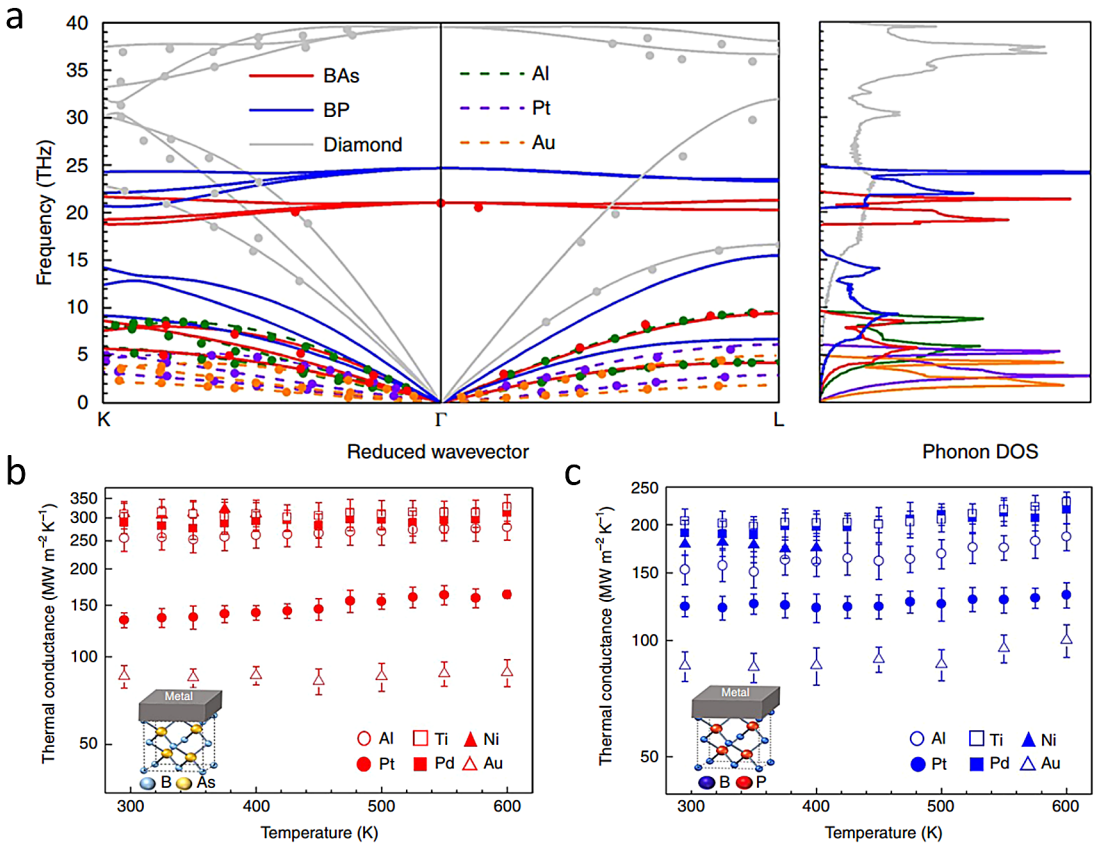

Figure 7. Thermal boundary resistance at interfaces between metals and BAs/BP. (a) Ab initio-derived phonon band structures for BAs, BP, diamond, Al, Au, and Pt. (b, c) Experimentally measured temperature-dependent TBR between metals and (b) BAs and (c) BP. Reproduced with permission from ref 5. Copyright 2021 The Authors, under exclusive license to Springer Nature Limited.

The first-principles-derived phonon band structures show good overlap between BAs/BP and common metals/semiconductors, indicating low TBRs as shown in Figure 7a.[5] The phonon density of states for semiconductors and metals typically ranges from 0 to 10 THz, whereas diamond has phonon frequencies extending up to 40 THz, creating a large spectral mismatch. For the BAs and BP, due to the presence of heavier elements, their phonon density of states aligns better with semiconductors and metals. Multiple modeling approaches were employed to predict TBR values, including radiation limit models without considering interface scattering, diffusive mismatch models assuming completely diffusive scattering at interface, and machine learning-assisted molecular dynamics (MD) simulations,[20] fully considering the harmonic effects and high-order anharmonicity. These approaches consistently indicate that BAs form substantially lower TBRs with metals compared to diamond. Our experimental measurements further confirmed that the TBR between Al and BAs or BP is less than 4 or 6 m^{2}K/GW, respectively, as demonstrated in Figure 7b and c.

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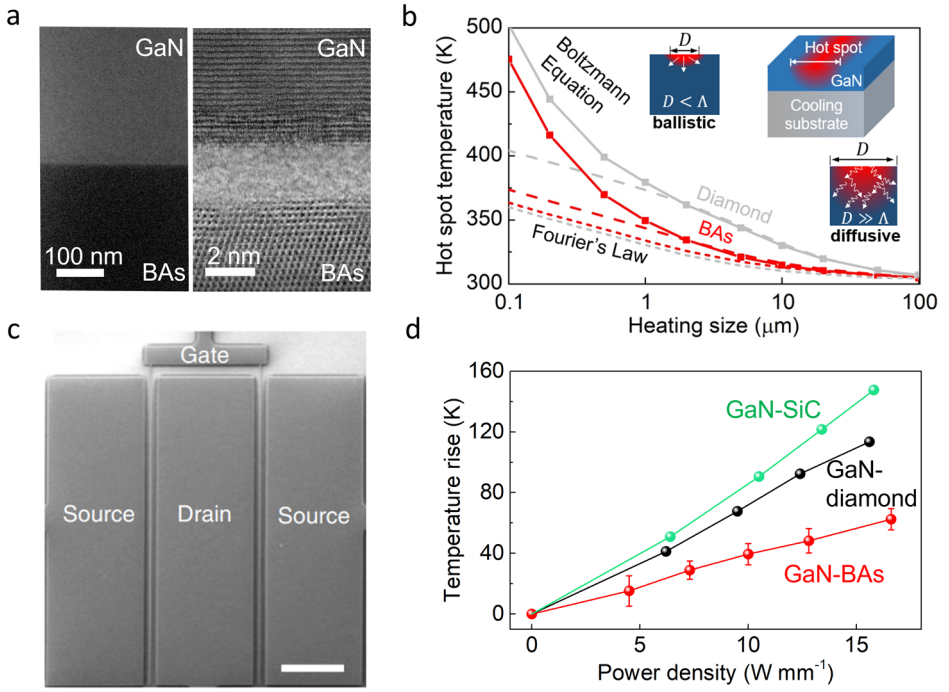

Figure 8. Integration of BAs and GaN for advanced thermal management. (a) Cross-sectional SEM (left) and high-resolution TEM (right) images showcasing the atomic interface between GaN and BAs layers. (b) Simulated hot-spot temperatures for BAs and diamond as a function of heating size, highlighting the transition from diffusive to ballistic thermal transport using a comparison between Fourier's heat conduction law and the spectral-dependent Boltzmann transport equation. (c) SEM image of an AlGaN/GaN high-electron-mobility transistor with two fingers, 100 μm width, and 34 μm gate pitch. Scale bar: 20 μm. (d) Experimental measurements of hot-spot temperature increase in operating devices as a function of transistor power density compared to reference GaN-on-diamond and GaN-on-SiC wafers. Reproduced with permission from ref 5. Copyright 2021 The Authors, under exclusive license to Springer Nature Limited.

These values are approximately five times lower than the TBR for diamond, which is around 20 m^2K/GW.[36]

3.2. Experimental Integration of Novel High Thermal Conductivity Materials with Low TBR into Power Electronics

Device integration of high-thermal-conductivity materials with low TBR is essential yet challenging. The thermal resistance of material layers and interfaces primarily determines the thermal bottleneck. The unique combination of low TBR and high thermal conductivity makes BAs a promising substrate for enhancing high-power cooling.[5] However, the direct integration of BAs with GaN is difficult because of their different crystal structures—zinc blende cubic for BAs and wurtzite for GaN—hindering perfect epitaxial interfaces. To establish a high-quality interface, we introduced a thin oxide layer to serve as an adhesion layer.[5] Following this, an oxygen plasma treatment was applied to activate interface bonds, and the sample was annealed at 773 K for 24 h under vacuum. The SEM and high-resolution TEM images in Figure 8a confirmed the formation of an atomically clean heterogeneous interface. By employing time-domain thermoreflectance (TDTR),[30] the TBR of the high-quality GaN-BAs interface was measured at approximately 4 m^2K/GW, which is over eight times improved compared to that of typical GaN-diamond interfaces.[5,37,38] This experimental result underscores the promise of using new materials such as BAs and BP for the high-performance cooling solution of the next-generation electronic devices.

Due to the nanoscale characteristic size of electronics, phonon transport changes from the diffusive to quasi-ballistic regime.[5,31,39] To consider the transport behavior transition, numerical simulations using both Fourier's law of heat conduction and the spectral-dependent Boltzmann transport equation were performed to study the combinational effects of low TBR and high κ on hot spot temperature in GaN transistors compared to the current state-of-the-art GaN-diamond devices. The hot spot temperature as a function of heating size is plotted in Figure 8b. It was observed that ballistic thermal transport becomes substantial when the heating spot size is less than 5 μm for GaN-BAs and 1 μm for GaN-diamond, which is consistent with the phonon mean free path spectra of these two materials. Under ballistic transport regime, the hot-spot temperature would be much higher than the estimation from diffusive regime. Regardless of the transport regime, for the heating size range from 100 nm to 100 μm, BAs has superior heat dissipation performance compared with diamond due to both the high thermal conductivity and the low TBR. Incorporating the GaN HEMT with a BAs substrate, as depicted in Figure 8c, the enhanced cooling performance was experimentally evaluated. Figure 8d presents the hot spot temperature measured as a function of power density for GaN HEMTs on a cooling substructe, utilizing three of the best thermal conductors: BAs, diamond, and SiC. Under identical device layouts and operating conditions, the cooling performance of BAs surpasses that of diamond and SiC,[40,41] corroborating the predictions from the modeling. For instance, at a transistor power density of approximately 15 W/mm, the hot spot temperature increase for GaN-BAs devices is around 60 K, which is significantly lower than the temperature increase for GaN-diamond and GaN-SiC devices. In addition, our study[42] demonstrates a scalable processing route for wafer-scale bonding with high interface thermal conductance, advancing chip-level packaging. These efforts aim to provide key insights into mitigating near-junction

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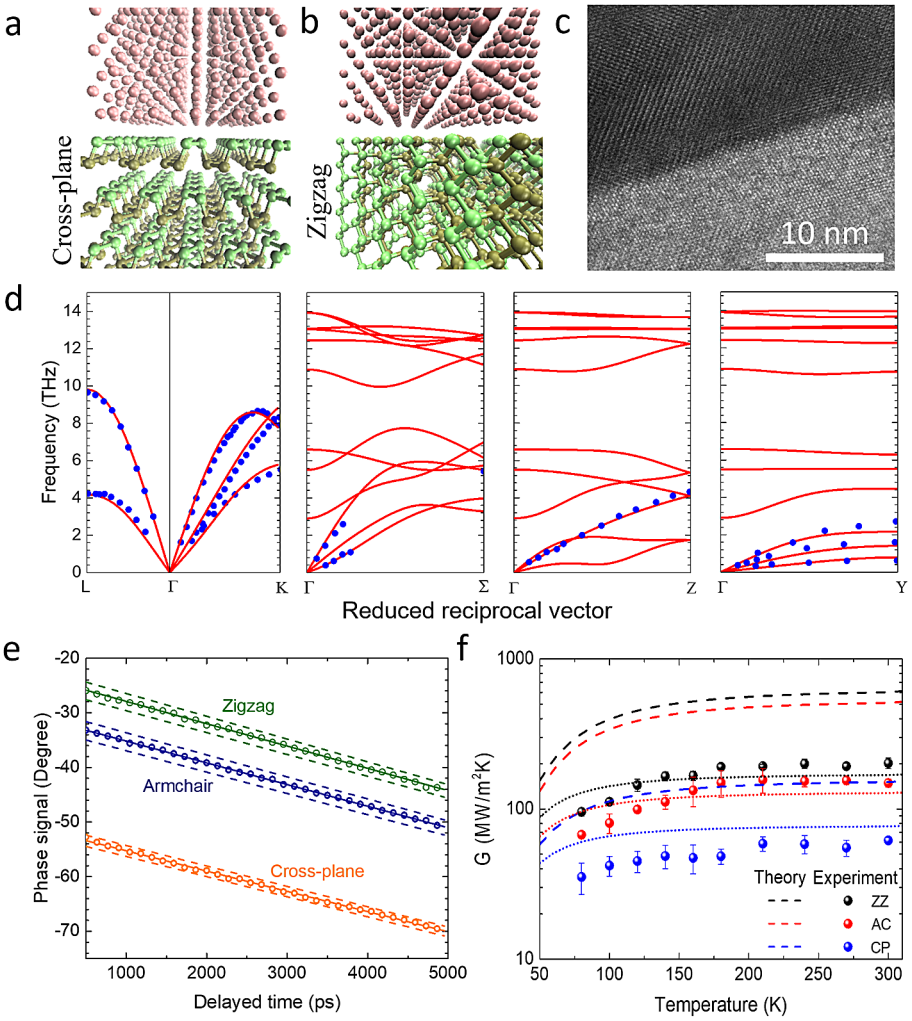

Figure 9. Anisotropic thermal boundary resistance (TBR) across a two-dimensional black phosphorus interface. (a and b) Schematic of interfaces between Al and black phosphorus with zigzag and armchair orientations, respectively. (c) TEM image of the interface between Al and black phosphorus. (d) First-principles-derived phonon dispersion relations along various reciprocal space directions. (e and f) TDTR measurements of temperature-dependent TBR along different directions, in comparison with modeling results. Reproduced with permission from ref 45. Copyright 2019 Wiley-VCH.

thermal resistance and supporting codesign strategies for advanced thermal management technologies.

3.3. TBR Consideration for Thermal Management in Electronics of 2D Materials

Two-dimensional materials have emerged as promising platforms for high-performance electronics and photonics since the discovery of graphene, making studying heat dissipation across their interfaces of high interest.[1,3,26,43−46] Most TBR studies focus on idealized interfaces, but materials with anisotropic structures remain underexplored. 2D materials are the ideal platforms for exploring the structural relationship because of their highly orientation-dependent phonon band structures.[45,46] To study the anisotropic TBR across 2D black phosphorus, we prepared interfaces between Al and black phosphorus with different crystal orientations, such as zigzag, armchair, and cross-plane directions.[45] These interfaces were examined using TEM and Raman spectroscopy, as illustrated in Figure 9a−c. First-principles calculations reveal that the anisotropic behavior originates from the intrinsic anisotropy of the phonon dispersions, as shown in Figure 9d. This anisotropy results in a significant difference in the phonon group velocities along the armchair and zigzag directions. Thermal measurements of temperature-dependent TBRs between a metal film and a 2D black phosphorus sample confirmed the predicted strong anisotropy, with the armchair direction displaying a higher TBR than the zigzag direction, as illustrated in Figure 9e and f. The observed anisotropy of TBR reached up to 3.27, setting a record of the highest anisotropy among all reported TBR values.

To quatify the phonon spectral contributions to TBR, we performed MD simulations to study thermal transport across heterostructure interfaces.[45] The simulations on Al-black phosphorus interface revealed that the anisotropic behavior primarily stems from the anisotropic phonon density of states in black phosphorus, leading to varying degrees of phonon scattering across the interface. Additionally, by comparing the MD results with the diffusive mismatch model, with and without considering phonon mode conversion, it was found that phonon hopping between different branches plays an important role in modulating the interfacial transport process, with directional preferences. The study provides microscopic understanding of anisotropic TBR across 2D materials to guide electronic device design.

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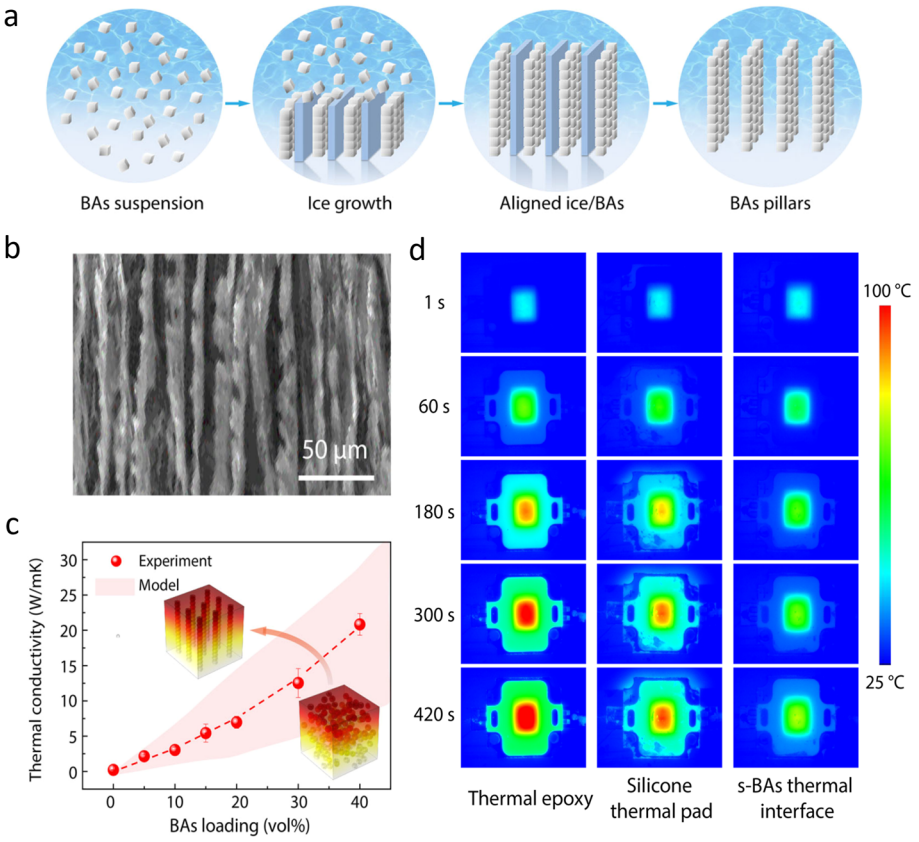

Figure 10. Flexible thermal interface material with high thermal conductivity based on self-assembled BAs. (a) Schematic to illustrate the self-assembly process, where freeze-drying of BAs suspensions leads to the formation of aligned BAs pillars. (b) Cross-section SEM image of the BAs composite confirms the presence of an aligned lamellar structure. (c) Thermal conductivity of BAs composites with different filler loadings in comparison with the modeling results. (d) Time-dependent infrared images of the LED integrated with different materials (thermal epoxy, silicone thermal pad, and BAs composite) display temperature distributions near the hot spot, highlighting the superior cooling capability of BAs composite compared to conventional materials. Reproduced with permission from ref 6. Copyright 2021 The Authors.

In addition, twisted van der Waals materials with Moiré patterns[46] offer new design possibilities and unconventional behaviors in electrical, optical, spintronic, and superconducting properties. However, thermal experiments across Moiré patterns remain limited, despite its critical importance. Our recent work[46] presents the first experimental study of thermal transport across twisted graphene, illustrating a phonon polarizer concept enabled by rotational misalignment between stacked layers. Through direct thermal and acoustic measurements, structural characterizations, and atomistic modeling, it was found that thermal conductance varies with Moiré angles while maintaining high acoustic transmission. By comparing experiments with DFT and MD simulations, we quantify mode-dependent phonon transmissions based on the angle alignment of graphene band structures, which we attribute to coupling among flexural phonon modes. The agreement confirms that tuning mechanisms predominantly adjust phonon transmission in high-frequency thermal modes, with minimal effects on low-frequency acoustic modes near the Brillouin zone center. This study provides essential insights into fundamental thermal transport in Moiré structures, paving the way for quantum thermal devices and novel thermal management design of phonon spectral manipulation.

4. ELECTRONIC PACKAGING WITH EFFICIENT THERMAL INTERFACE MATERIAL

Effective thermal management also relies on minimizing thermal resistance across multiple device layers and their interfaces. Developing thermal interfaces that enhance thermal coupling is the key to addressing this challenge in both academia and industry.[47,48] Various thermal interface materials have been developed from academia and industry, including thermal greases, gels, pads, tapes, conductive adhesives, phase change materials, and metallic solders.[1,6] However, current commercial thermal interfaces often face limitations due to either low thermal conductivity (~1 W/mK) or a high elastic modulus (~1 GPa). Strong-bonding materials provide high thermal conductivity, but their rigid structures can degrade performance through mechanical pump-out, delamination, or cracking.[49] In contrast, soft materials like polymers maintain effective interface contact but are limited by low thermal conductivity.[50] Ideally, high-performance thermal interfaces should possess both high κ value and low elastic modulus. Additionally, properties such as damping, toughness, and adhesion contribute to maintaining stable thermal contact.[1,51,52]

Our recent study made progress in developing a high-performance and flexible thermal interface through structural optimization and self-assembly of BAs.[6] In general, polymer matrixes with fillers enable mechanical compliance, but their low κ values have remained unsatisfactory. When fillers are dispersed randomly, heat transfer paths within polymers can be

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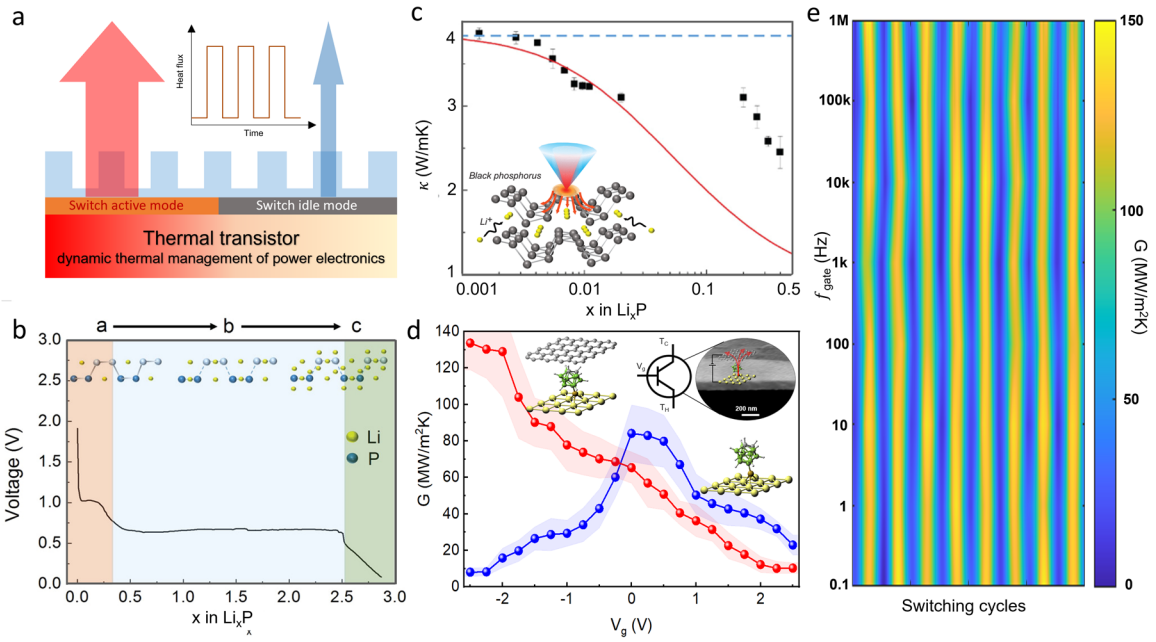

Figure 11. Dynamical thermal management using low-dimensional materials. (a) Schematic illustrating dynamic thermal management with precise heat control across spatial and temporal dimensions. (b) Galvanostatic discharge curve for the Li-black phosphorus device. (c) In situ thermal measurement of cross-plane thermal conductivity of electrochemically intercalated black phosphorus. (d) The thermal conductance switching behavior as a function of the gate voltage (V_g).. Inset: SEM image of an electrically gated molecular thermal switch. (e) Thermal conductance switching cycles versus scanning frequency up to 1 MHz, measured at every decade. Reproduced with permission from refs 55 and 3. Copyright 2017 American Chemical Society and 2023 The Authors, some rights reserved; exclusive licensee American Association for the Advancement of Science.

significantly elongated, limiting the contribution from fillers. In order to achieve optimal alignment of BAs, a self-assembly manufacturing method utilizing an ice-template process was developed, as shown in Figure 10. BAs particles were dispersed to create an aqueous suspension, which was then transferred into a tube mold. A directional temperature gradient was applied across the mold, promoting the growth of ice crystals into aligned lamellar pillars following the temperature gradient direction.[53,54] Simultaneously, the ice template's growth drove BAs crystals to assemble into arrays, replicating the morphology and occupying the spaces between the ice pillars. Once the self-assembly process was complete, the sample underwent freeze-drying to preserve the aligned BAs structures. Next, a polymer melt infiltrated into the BAs assembly and solidified, resulting in a composite with improved mechanical support. Cross-sectional SEM images in Figure 10b verified the resulting structures. The thermal conductivity of BAs composites, measured using the laser flash method, reached a high value up to 21 W/mK. This value is over an order of magnitude greater than those of typical thermal epoxies and greases used as industrial thermal interfaces. Meanwhile, the Young's modulus and shear modulus of the BAs composites remained low at 256 and 148 kPa, respectively, indicating good mechanical compliance.

To evaluate the cooling performance in device applications, three types of thermal interfaces (commercial thermal epoxy, silicone sheet, and BAs composite) were integrated between a 10 W LED chip and a copper heat sink. Infrared images measuring the transient temperature dependence revealed that the chip surface temperature was significantly lower when the BAs composite was employed as the thermal interface. This marked difference in hot spot temperature (Figure 10) highlights the superior cooling capability of the developed BAs composite, establishing it as a promising solution for future thermal management for flexible electronics.

5. DYNAMICAL THERMAL MANAGEMENT

To operate electronic devices at optimal temperature, the ability to retain heat becomes paramount in specific conditions, such as frigid climates and aerospace environments.[3,55] It is crucial to make devices maintain their optimal temperature through dynamical thermal management (Figure 1a). Over past decades, scientists and engineers have explored various methods, like phase changes, mass transfer, ion motions, and carrier scattering, to tackle this challenge.[55] One intensively explored approach is electrochemical interaction in layered materials.[55] Our prior study shows that the thermal conductivity of layered materials can be modulated by electrochemically modulating the ion concentration, as depicted in Figure 11 b and c. By performing in situ thermal characterization of battery-like lithium-ion intercalated black phosphorus, we revealed the precise and reversible control of anisotropic thermal conductivity. The black phosphorus can experience three phases with the increasing Li concentration. The range of κ can be tuned, from 2.45 to 3.86, 62.67 to 85.80, and 21.66 to 27.58 W/mK. This modulation in κ is attributed to phonon scattering induced by ionic intercalation within interspacing layers, showing an anisotropic behavior. Notably, in its fully discharged state ($x \sim 3$ in Li$_x$P), κ can be reduced to nearly a sixth of its original value in the pristine crystal. On the other side, the rely on liquid electrolytes and ion mass movements could result in substantial limitations in device performance and portability. Additionally, structural deformation and degradation have hindered reliability and reversibility. Other strategies, involving ferroelectric materials, phase change materials, thermal radiative systems, microelectromechanical systems, or quantum devices, have shown limited performance (on/off ratio of <5 and frequency of <1 Hz) or contingent requirements for working environments (e.g., below 10 K).[56]

To address these challenges, more recently, we developed a new concept of an electrically gated thermal transistor to

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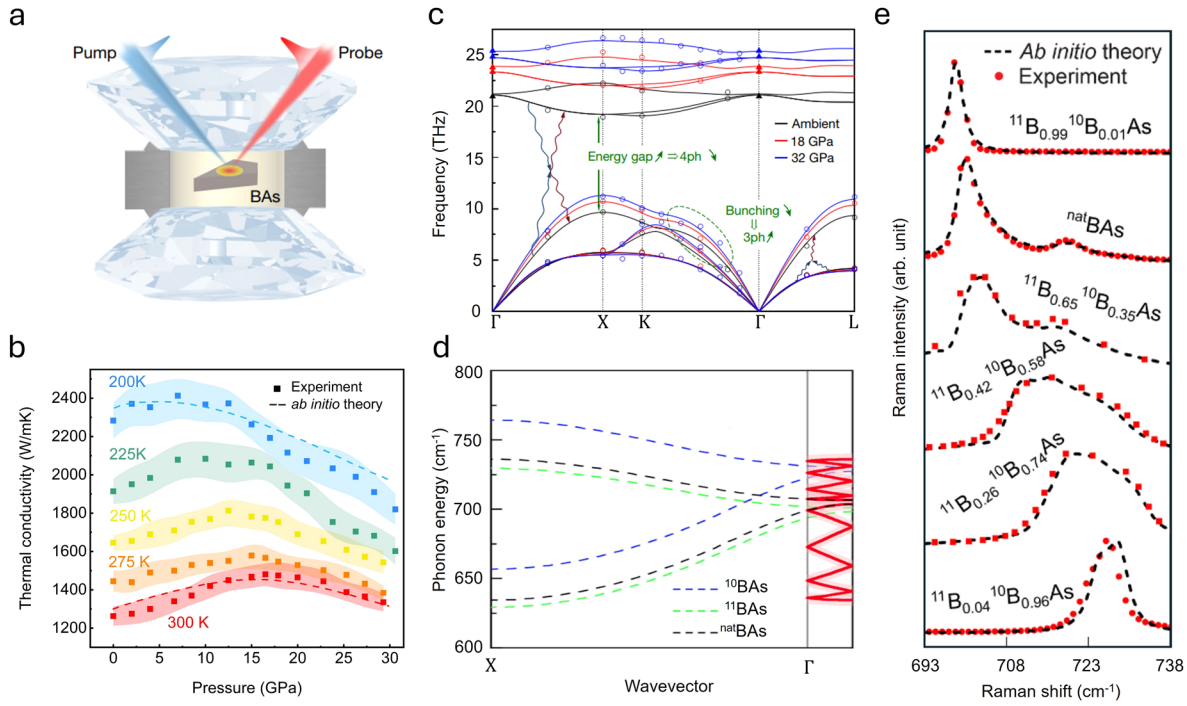

Figure 12. High-order phonon anharmonicity and non-perturbative interactions in BAs and BP. (a) Schematic of in situ spectroscopy measurements at high pressure with a diamond anvil cell. (b) Pressure-dependent thermal conductivity of BAs at various temperature and pressure. (c) Experimentally measured phonon dispersion from inelastic X-ray scattering (circles), Raman spectroscopy (triangles), and ab initio calculations (lines) under three pressures indicates the interplay of three-phonon and four-phonon scatterings. (d) Phonon dispersion (optical branches) for isotopically pure and naturally abundant BAs. The schematic in the right column illustrates the Brillouin zone folding caused by local and coupled isotope effects. (e) Comparison between calculated (black curve) and experimental (circle) of Raman spectra of BAs with different B isotope concentrations. Reproduced with permission from refs 4 and 58. Copyright 2022 and 2023 The Authors, under exclusive license to Springer Nature Limited and American Physical Society.

advance future technologies for dynamic thermal management.[3] We developed solid-state thermal transistor that employs a field effect (the modulation of thermal conductance by the application of an external electric field) and a fully solid state (no moving parts), offering high performance and compatibility with integrated circuits in semiconductor manufacturing processes. Our design, as shown in the Figure 11d inset, incorporates the field effect on charge dynamics at an atomic interface to allow an electrical gate using a negligible power to continuously switch and amplify a heat flux. This device demonstrates a record-high performance with a switching speed of more than 1 megahertz, or 1 million cycles per second, and offers a 1300% tunability in thermal conductance and reliable performance for more than millions of switching cycles (Figure 11 d and e). Comprehensive atomistic modeling, employing DFT and MD, provides insightful explanations for these observed behaviors.[3] This innovation represents a fundamental advancement in dynamic thermal management strategies for semiconductors and 3D-stacked chips, enabling new thermal management technology with precise thermal power control in both time and spatial domains.

6. NEW PHYSICS AND HIGH-ORDER PHONON INTERACTIONS

The developed high-thermal-conductivity materials provide a powerful platform to explore transport physics[57] beyond classical principles. For instance, high-order phonon anharmonicity[4] could refine the design rules and expand research into extreme conditions, including high temperature, pressure, and power density. By examining the thermal properties of BAs under high pressure, the interplay between three-phonon and four-phonon scattering processes was systematically analyzed and experimentally confirmed.[4] Past literature studies of high-pressure thermal transport show that thermal conductivity of materials usually changes monotonically with pressure unless phase transition takes place. In our recent study, we observed an abnormal pressure dependence of κ in BAs.[4] Utilizing a diamond anvil cell, as depicted in Figure 12a, we investigated the evolution of the phonon band structure under pressure through multiple in situ spectroscopy methods like ultrafast optics, Raman spectroscopy, and inelastic X-ray scattering measurements. We measured the κ values across a range of temperatures and pressures, reaching up to 32 GPa. Our observations in Figure 12b show that κ first increases and then decreases with pressure, different from all other materials. Experimental findings combined with ab initio calculations, highlighted in Figure 12c, show an increase in the phonon energy bandgap and separation of closely bunched acoustic phonon branches under pressure. These changes suggest a decline in four-phonon scattering and an enhancement in three-phonon scattering, competitively leading to the nonmonotonic change of thermal conductivity. On the other hand, our study revealed that strong intrinsic anharmonicity can induce phonon renormalization near room temperature, leading to the breakdown of the quasi-harmonic approximation in phonon modeling. These studies provide fundamental insights into microscopic energy transport mechanisms under extreme conditions, guiding the design of materials for various environments.

Understanding the Raman spectra of BAs and BP has been a challenge over the past decade.[58] The anomalous two-mode

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Raman response in BAs is unique and cannot be explained by standard models such as quantum perturbation theory. Initially attributed to TO-LO splitting, this explanation was later found to be inconsistent with experimental energy scales. Our recent theoretical work[58] was able to identify the fundamental origin of this anomalous behavior: the coupling and random distribution of isotope atoms in boron compounds breaks the translational symmetry and folds the Brillouin zone, as illustrated in Figure 12d. To precisely quantify the Raman spectra, we developed a nonperturbative ab initio approach[58] to accurately model the unique vibrational physics in BAs and BP. By combining DFT calculations with a nonperturbative supercell exact diagonalization method, we calculated the Raman spectra of BAs as a function of isotope concentration (Figure 12e), achieving excellent agreement with experiments. We found that the vibrational spectra are notably influenced by local fluctuations and isotope coupling. By decomposing the vibrational spectra based on individual isotope eigenvectors, we identified both positive and negative contributions to Raman intensity from constituent atoms. Additionally, we attributed the LO-TO energy splitting at high pressures to the increased LO mode frequencies from intensified long-range Coulomb interactions. At elevated pressures, the reduced interatomic distances drive more electrons toward boron atoms, amplifying the LO-TO splitting and forming a new resonance Raman peak.

7. CONCLUSIONS AND OUTLOOK

Heat dissipation represents one of the most challenging technical issues in modern electronics and is expected to further escalate for 3D IC packaging and chiplet technology. This Account reviews the efforts in developing high-thermal-conductivity materials, engineering interface thermal boundary resistance, and establishing effective thermal interfaces between devices and heat sinks. More specifically, high-quality BAs and BP single crystals have been successfully synthesized and measured, exhibiting thermal conductivities of up to 1300 and 500 W/mK, respectively. Low TBRs between BAs, BP, and typical materials used in semiconductor devices have been experimentally observed. The integration of GaN HEMTs on BAs has demonstrated superior cooling performance compared to devices on state-of-the-art materials like diamond and SiC. Furthermore, an efficient and flexible thermal interface was formed with structurally aligned BAs in polymer composites, as demonstrated by bonding a light-emitting diode with a heat sink, resulting in a significantly improved heat dissipation rate. The development of high thermal conductivity BAs, coupled with its low TBR and composite form, has the potential to significantly impact electronic cooling applications. The understanding of new phonon physics from these new materials could inspire discovery of next-generation building blocks and novel utilization of phonon transport in energy technologies. In addition, we highlight the latest developments of dynamic thermal management devices with a particular focus on solid-state thermal transistor.

With the increasing power density of electronics, challenges remain in overcoming thermal bottlenecks. First, the discovery of new high-thermal-conductivity materials is an ongoing objective that relies on the understanding of phonon transport, especially the interaction of phonons with other bosons and fermions, such as electrons, magnons, and excitons. The picture becomes even more critical at interfaces, where mode-resolved characterization is needed. Second, while ab initio modeling provides great insights and rigorousness, the integration with machine learning and AI approaches may help accelerate the computational processes.[20] Third, current heat dissipation design of power electronics is primarily focused on the chip level. A bottom-up philosophy, encompassing the transistor, circuit, and device levels, should be adopted to optimize heat dissipation pathways, including considerations at the early stages of electrical design. This becomes increasingly critical with the advent of more complex chip architectures, such as 2.5D and 3D chip packaging. A multiscale modeling incorporating ballistic phonon transport, multiple scattering mechanisms, and diffusive behaviors is essential. Additionally, the development of new characterization methods capable of mapping electric and temperature fields with high spatial and temporal resolutions will be instrumental in device design and testing. Lastly, the mass production of new materials will become a focal point in the industry. The design and manufacturing of thermal management systems, such as fins, heat pipes, and heat exchangers, using these high-performance materials will be crucial for industry adoption and effectiveness.

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Notes

The authors declare no competing financial interest.

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deepen our fundamental understanding of transport phenomena and guide the next generation of thermal management strategies.

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■ REFERENCES

(1) Cui, Y.; Li, M.; Hu, Y. Emerging Interface Materials for Electronics Thermal Management: Experiments, Modeling, and New Opportunities. *J. Mater. Chem. C* **2020**, *8*, 10568.
(2) Ravi, S. *International technology roadmap for semiconductors (ITRS)*, 2015 ed.; Semiconductor Industry Association, 2015. https://www.semiconductors.org/resources/2015-international-technology-roadmap-for-semiconductors-itrs/.
(3) Li, M.; Wu, H.; Avery, E. M.; Qin, Z.; Goronzy, D. P.; Nguyen, H. D.; Liu, T.; Weiss, P. S.; Hu, Y. Electrically Gated Molecular Thermal Switch. *Science* **2023**, *382*, 585−589.
(4) Li, S.; Qin, Z.; Wu, H.; Li, M.; Kunz, M.; Alatas, A.; Kavner, A.; Hu, Y. Anomalous Thermal Transport under High Pressure in Boron Arsenide. *Nature* **2022**, *612*, 459−464.
(5) Kang, J. S.; Li, M.; Wu, H.; Nguyen, H.; Aoki, T.; Hu, Y. Integration of Boron Arsenide Cooling Substrates into Gallium Nitride Devices. *Nat. Electron* **2021**, *4*, 416−423.
(6) Cui, Y.; Qin, Z.; Wu, H.; Li, M.; Hu, Y. Flexible Thermal Interface Based on Self-Assembled Boron Arsenide for High-Performance Thermal Management. *Nat. Commun.* **2021**, *12*, 1284.
(7) Kang, J. S.; Li, M.; Wu, H.; Nguyen, H.; Hu, Y. Experimental Observation of High Thermal Conductivity in Boron Arsenide. *Science* **2018**, *361*, 575−578.
(8) Waldrop, M. M. The Chips Are down for Moore's Law. *Nature* **2016**, *530*, 144.
(9) Ball, P. Computer Engineering: Feeling the Heat. *Nature* **2012**, *492*, 174.
(10) DARPA Microsystems Technology Office. *Technologies for Heat Removal in Electronics at the Device Scale (THREADS)*; U.S. Department of Defense: Arlington County, VA, 2022.
(11) Bar-Cohen, A. *Thermal Packaging Techniques*; Encyclopedia of Thermal Packaging, Set 1; World Scientific, 2012.
(12) Won, Y.; Cho, J.; Agonafer, D.; Asheghi, M.; Goodson, K. E. Fundamental Cooling Limits for High Power Density Gallium Nitride Electronics. *IEEE Trans. Compon. Packag. Manufact. Technol.* **2015**, *5*, 737−744.
(13) Mishima, O.; Tanaka, J.; Yamaoka, S.; Fukunaga, O. High-Temperature Cubic Boron Nitride Pn Junction Diode Made at High Pressure. *Science* **1987**, *238*, 181−183.
(14) Lux, B.; Kalss, W.; Haubner, R.; Taniguchi, T. Nucleation of c-BN on Various Substrate Materials under High-Pressure−High-Temperature Conditions. *Diam. Relat. Mater.* **1999**, *8*, 415−422.
(15) Balandin, A. A. Thermal Properties of Graphene and Nanostructured Carbon Materials. *Nat. Mater.* **2011**, *10*, 569−581.
(16) Marconnet, A. M.; Panzer, M. A.; Goodson, K. E. Thermal Conduction Phenomena in Carbon Nanotubes and Related Nanostructured Materials. *Rev. Mod. Phys.* **2013**, *85*, 1295−1326.
(17) Slack, G. A. Nonmetallic Crystals with High Thermal Conductivity. *J. Phys. Chem. Solids* **1973**, *34*, 321−335.
(18) Slack, G. A.; Tanzilli, R. A.; Pohl, R. O.; Vandersande, J. W. The Intrinsic Thermal Conductivity of AlN. *J. Phys. Chem. Solids* **1987**, *48*, 641−647.
(19) Broido, D. A.; Malorny, M.; Birner, G.; Mingo, N.; Stewart, D. A. Intrinsic Lattice Thermal Conductivity of Semiconductors from First Principles. *Appl. Phys. Lett.* **2007**, *91*, No. 231922.
(20) Li, M.; Dai, L.; Hu, Y. Machine Learning for Harnessing Thermal Energy: From Materials Discovery to System Optimization. *ACS Energy Lett.* **2022**, *7*, 3204−3226.
(21) Lindsay, L.; Broido, D. A.; Reinecke, T. L. First-Principles Determination of Ultrahigh Thermal Conductivity of Boron Arsenide: A Competitor for Diamond? *Phys. Rev. Lett.* **2013**, *111*, No. 025901.
(22) Broido, D. A.; Lindsay, L.; Reinecke, T. L. *Ab Initio* Study of the Unusual Thermal Transport Properties of Boron Arsenide and Related Materials. *Phys. Rev. B* **2013**, *88*, No. 214303.
(23) Feng, T.; Lindsay, L.; Ruan, X. Four-Phonon Scattering Significantly Reduces Intrinsic Thermal Conductivity of Solids. *Phys. Rev. B* **2017**, *96*, No. 161201.
(24) Kang, J. S.; Wu, H.; Hu, Y. Thermal Properties and Phonon Spectral Characterization of Synthetic Boron Phosphide for High Thermal Conductivity Applications. *Nano Lett.* **2017**, *17*, 7507−7514.
(25) Wu, H.; Fan, H.; Hu, Y. *Ab Initio* Determination of Ultrahigh Thermal Conductivity in Ternary Compounds. *Phys. Rev. B* **2021**, *103*, No. L041203.
(26) Fan, H.; Wu, H.; Lindsay, L.; Hu, Y. *Ab Initio* Investigation of Single-Layer High Thermal Conductivity Boron Compounds. *Phys. Rev. B* **2019**, *100*, No. 085420.
(27) Kang, J. S.; Li, M.; Wu, H.; Nguyen, H.; Hu, Y. Basic Physical Properties of Cubic Boron Arsenide. *Appl. Phys. Lett.* **2019**, *115*, No. 122103.
(28) Tian, F.; Song, B.; Chen, X.; Ravichandran, N. K.; Lv, Y.; Chen, K.; Sullivan, S.; Kim, J.; Zhou, Y.; Liu, T. H.; Goni, M.; Ding, Z.; Sun, J.; Gamage, G. A. G. U.; Sun, H.; Ziyaee, H.; Huyan, S.; Deng, L.; Zhou, J.; Schmidt, A. J.; Chen, S.; Chu, C. W.; Huang, P. Y.; Broido, D.; Shi, L.; Chen, G.; Ren, Z. Unusual High Thermal Conductivity in Boron Arsenide Bulk Crystals. *Science* **2018**, *361*, 582−585.
(29) Li, S.; Zheng, Q.; Lv, Y.; Liu, X.; Wang, X.; Huang, P. Y.; Cahill, D. G.; Lv, B. High Thermal Conductivity in Cubic Boron Arsenide Crystals. *Science* **2018**, *361*, 579−581.
(30) Li, M.; Kang, J. S.; Hu, Y. Anisotropic Thermal Conductivity Measurement Using a New Asymmetric-Beam Time-Domain Thermoreflectance (AB-TDTR) Method. *Rev. Sci. Instrum.* **2018**, *89*, No. 084901.

https://doi.org/10.1021/accountsmr.4c00349
Acc. Mater. Res. 2025, 6, 563−576

(31) Hu, Y.; Zeng, L.; Minnich, A. J.; Dresselhaus, M. S.; Chen, G. Spectral Mapping of Thermal Conductivity through Nanoscale Ballistic Transport. *Nat. Nanotechnol.* **2015**, *10*, 701−706.
(32) Wu, J.; Zhou, E.; Huang, A.; Zhang, H.; Hu, M.; Qin, G. Deep-Potential Enabled Multiscale Simulation of Gallium Nitride Devices on Boron Arsenide Cooling Substrates. *Nat. Commun.* **2024**, *15*, 2540.
(33) Li, M.; Dai, L.; Wu, H.; Yan, Y.; Kang, J.; King, S.; McNeil, P. E.; Butts, D.; Galy, T.; Marszewski, M.; Lan, E.; Dunn, B. S.; Tolbert, S. H.; Pilon, L.; Hu, Y. Ballistic Transport from Propagating Vibrational Modes in Amorphous Silicon Dioxide: Thermal Experiments and Atomistic-Machine Learning Modeling. *Mater. Today Phys.* **2025**, *51*, No. 101659.
(34) Samaniego, E.; Anitescu, C.; Goswami, S.; Nguyen-Thanh, V. M.; Guo, H.; Hamdia, K.; Zhuang, X.; Rabczuk, T. An Energy Approach to the Solution of Partial Differential Equations in Computational Mechanics via Machine Learning: Concepts, Implementation and Applications. *Comput. Methods Appl. Mech. Eng.* **2020**, *362*, No. 112790.
(35) Swartz, E. T.; Pohl, R. O. Thermal Boundary Resistance. *Rev. Mod. Phys.* **1989**, *61*, 605.
(36) Stoner, R. J.; Maris, H. J.; Anthony, T. R.; Banholzer, W. F. Measurements of the Kapitza Conductance between Diamond and Several Metals. *Phys. Rev. Lett.* **1992**, *68*, 1563.
(37) Cho, J.; Li, Z.; Asheghi, M.; Goodson, K. E. Near-Junction Thermal Management: Thermal Conduction in Gallium Nitride Composite Substrates. *Annu. Rev. Heat Transfer* **2015**, *18*, 7−45.
(38) Cheng, Z.; Mu, F.; Yates, L.; Suga, T.; Graham, S. Interfacial Thermal Conductance across Room-Temperature-Bonded GaN/Diamond Interfaces for GaN-on-Diamond Devices. *ACS Appl. Mater. Interfaces* **2020**, *12*, 8376−8384.
(39) Shen, Y.; Yang, H.-A.; Cao, B.-Y. Near-Junction Phonon Thermal Spreading in GaN HEMTs: A Comparative Study of Simulation Techniques by Full-Band Phonon Monte Carlo Method. *Int. J. Heat Mass Transfer* **2023**, *211*, No. 124284.
(40) Sun, H.; Simon, R. B.; Pomeroy, J. W.; Francis, D.; Faili, F.; Twitchen, D. J.; Kuball, M. Reducing GaN-on-Diamond Interfacial Thermal Resistance for High Power Transistor Applications. *Appl. Phys. Lett.* **2015**, *106*, No. 111906.
(41) Pomeroy, J. W.; Bernardoni, M.; Dumka, D. C.; Fanning, D. M.; Kuball, M. Low Thermal Resistance GaN-on-Diamond Transistors Characterized by Three-Dimensional Raman Thermography Mapping. *Appl. Phys. Lett.* **2014**, *104*, 083513.
(42) Li, M.; Pan, K.; Ge, Y.; Huynh, K.; Goorsky, M. S.; Fisher, T. S.; Hu, Y. Wafer-Scale Bonded GaN−AlN with High Interface Thermal Conductance. *Appl. Phys. Lett.* **2024**, *125*, No. 032104.
(43) Liu, Y.; Duan, X.; Shin, H.-J.; Park, S.; Huang, Y.; Duan, X. Promises and Prospects of Two-Dimensional Transistors. *Nature* **2021**, *591*, 43−53.
(44) Gu, X.; Wei, Y.; Yin, X.; Li, B.; Yang, R. Colloquium: Phononic Thermal Properties of Two-Dimensional Materials. *Rev. Mod. Phys.* **2018**, *90*, No. 041002.
(45) Li, M.; Kang, J. S.; Nguyen, H. D.; Wu, H.; Aoki, T.; Hu, Y. Anisotropic Thermal Boundary Resistance across 2D Black Phosphorus: Experiment and Atomistic Modeling of Interfacial Energy Transport. *Adv. Mater.* **2019**, *31*, No. 1901021.
(46) Qin, Z.; Dai, L.; Li, M.; Li, S.; Wu, H.; White, K. E.; Gani, G.; Weiss, P. S.; Hu, Y. Moiré Pattern Controlled Phonon Polarizer Based on Twisted Graphene. *Adv. Mater.* **2024**, *36*, No. 2312176.
(47) Larson, L.; Tang, Y.; Durfee, L.; Hale, C.; Plante, D.; Iruvanti, S.; Wagner, R.; Davis, T.; Longworth, H.; Lavoie, A. Engineered Thermal Interface Material. In *Proceedings of the 2014 IEEE 64th Electronic Components and Technology Conference*, Orlando, FL, May 27−30, 2014; IEEE, 2014; pp 236−241.
(48) Chainer, T. J.; Schultz, M. D.; Parida, P. R.; Gaynes, M. A. Improving Data Center Energy Efficiency with Advanced Thermal Management. *IEEE Trans. Compon. Packaging Manuf. Technol.* **2017**, *7*, 1228−1239.
(49) Ma, H.; Suhling, J. C. A Review of Mechanical Properties of Lead-Free Solders for Electronic Packaging. *J. Mater. Sci.* **2009**, *44*, 1141−1158.
(50) Baur, J.; Silverman, E. Challenges and Opportunities in Multifunctional Nanocomposite Structures for Aerospace Applications. *MRS Bull.* **2007**, *32*, 328−334.
(51) Xu, M.; Du, F.; Ganguli, S.; Roy, A.; Dai, L. Carbon Nanotube Dry Adhesives with Temperature-Enhanced Adhesion over a Large Temperature Range. *Nat. Commun.* **2016**, *7*, 13450.
(52) Tan, C.; Dong, Z.; Li, Y.; Zhao, H.; Huang, X.; Zhou, Z.; Jiang, J.-W.; Long, Y.-Z.; Jiang, P.; Zhang, T.-Y.; et al. A High Performance Wearable Strain Sensor with Advanced Thermal Management for Motion Monitoring. *Nat. Commun.* **2020**, *11*, 3530.
(53) Deville, S.; Saiz, E.; Nalla, R. K.; Tomsia, A. P. Freezing as a Path to Build Complex Composites. *Science* **2006**, *311*, 515−518.
(54) Zhang, H.; Hussain, I.; Brust, M.; Butler, M. F.; Rannard, S. P.; Cooper, A. I. Aligned Two- and Three-Dimensional Structures by Directional Freezing of Polymers and Nanoparticles. *Nat. Mater.* **2005**, *4*, 787−793.
(55) Kang, J. S.; Ke, M.; Hu, Y. Ionic Intercalation in Two-Dimensional van Der Waals Materials: *In Situ* Characterization and Electrochemical Control of the Anisotropic Thermal Conductivity of Black Phosphorus. *Nano Lett.* **2017**, *17*, 1431−1438.
(56) Wehmeyer, G.; Yabuki, T.; Monachon, C.; Wu, J.; Dames, C. Thermal Diodes, Regulators, and Switches: Physical Mechanisms and Potential Applications. *Appl. Phys. Rev.* **2017**, *4*, No. 041304.
(57) Su, C.; Wu, H.; Dai, L.; Zhang, Z.; Li, S.; Hu, Y. Non-Classical Heat Transfer and Recent Progress. *ASME J. Heat Mass Transfer* **2025**, *147*, No. 032502.
(58) Wu, H.; Qin, Z.; Li, S.; Lindsay, L.; Hu, Y. Nonperturbative Determination of Isotope-Induced Anomalous Vibrational Physics. *Phys. Rev. B* **2023**, *108*, No. L140302.

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