



Lattice thermal transport in superhard hexagonal diamond and wurtzite boron nitride: A comparative study with cubic diamond and cubic boron nitride

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ABSTRACT

Hexagonal diamond (h-C) and wurtzite boron nitride (w-BN) are two superhard materials recently identified to be comparable to or even harder than their cubic counterparts, cubic diamond (c-C) and cubic boron nitride (c-BN). To understand the effect of lattice structure on thermal transport in these materials, we conduct first-principles calculations to investigate their harmonic and anharmonic lattice properties. Owing to the strong C-C or B-N bonds, h-C and w-BN are found to have a high lattice thermal conductivity (κ_L) exceeding the overall thermal conductivity of metals, albeit lower than that of their cubic counterparts. By analyzing the phonon band structure and volume of the 3-phonon scattering phase space, we attribute the lower κ_L of the hexagonal phases to their larger volume of 3-phonon scattering phase space than the cubic ones. Moreover, we reveal that a high pressure of 125 GPa leads to a two- to three-fold increase in the κ_L of these materials, because the pressure enlarges the optical-acoustic phonon bandgap and thus reduces the volume of the 3-phonon scattering phase space. This work uncovers the significant effect of lattice structure and pressure on phonon scattering and transport, which is crucial for the application of superhard materials.

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1. Introduction

Diamond, of which the lattice can be viewed as a pair of intersecting face-centered cubic lattices, is one of the “supermaterials” that deliver extreme performance across a variety of applications. In particular, it has long been known as one of the hardest and thermally conductive materials on earth. In terms of its thermal performance, theoretical and experimental studies [1–4] have been conducted to investigate thermal transport in both naturally occurring and isotopically pure diamond, revealing that it possesses one of the highest thermal conductivities (κ) reported so far [5]. The ultrahigh κ of diamond primarily stems from its strong sp^3 C-C bonds and the light carbon atoms, which lead to high phonon group velocities that are higher than most materials in nature. Graphene, another allotrope of carbon with a hexagonal lattice, also has very high κ in the in-plane direction owing to the strong sp^2 C-C bonds and restricted phase space for anharmonic phonon

scattering [5–7]. Owing to their outstanding performance in dissipating heat, there is an increasing interest in applying diamond, graphite, and graphene in thermal management applications [5,7,8]. Diamond, as an excellent electrical insulator, is even preferred when electrical insulation is required.

Similar to those allotropes of carbon, various structures of boron nitride have also been found to possess high κ , though lower than that of diamond and graphene. For example, the room-temperature κ in the basal plane of a bulk sample of pyrolytic hexagonal boron nitride (h-BN) was measured to be up to 390 W/m-K [9]. In 2013, Jo et al. measured the κ of an 11-layer h-BN using a thermal bridge approach and found a value of approximately 360 W/m-K at room temperature, comparable to its bulk-limit value [10]. Later, Zhou et al. obtained room-temperature κ 's between 227 W/m-K and 280 W/m-K for a 9-layer h-BN using the confocal micro-Raman method [11]. Another allotrope of BN, cubic boron nitride (c-BN), was also found to have a high κ of 768 W/m-K from an early experiment [12] or 940 W/m-K from a recent first-principles study [13].

Recently, hexagonal diamond (h-C), a much less understood allotrope of carbon also referred to as lonsdaleite, was found to be

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even harder than its cubic counterpart [14]. With a similar structure as h-C, wurtzite boron nitride (w-BN) was also predicted to be ultrahard [14]. As shown in Fig. 1, h-C and w-BN have the same hexagonal lattice structure, which renders it more difficult for lattice planes to glide than their cubic counterparts, c-C and c-BN. Consequently, the hardness of h-C and w-BN are even superior than that of cubic diamond (c-C) and c-BN. Nonetheless, the thermal properties of h-C, c-BN, and w-BN are not well understood yet, inspiring us to investigate their κ and understand how their atomic structures dictate thermal transport properties, which is of both practical and fundamental importance. Moreover, as ultrahard materials are usually used under high-pressure conditions, we will also systematically study how high pressure affects thermal transport in these materials.

2. Methodology

The four materials to be studied in this work are insulators with wide electronic bandgap, in which thermal transport is primarily contributed by phonons. Phonons are quanta of lattice vibrations and are usually described with a quantum number, $\lambda = (\nu, \mathbf{q})$. In this equation, \mathbf{q} denotes wave vector and ν denotes branch index. The advancement in first-principles based calculation of lattice thermal conductivity κ_L [15–18] has enabled an accurate solution to the linearized phonon Boltzmann transport equation, and, correspondingly, the tensor of κ_L can be calculated as [19].

$$\kappa_L^{\alpha\beta} = \frac{1}{k_B T^2 \Omega N} \sum_{\lambda} f_0 (f_0 + 1) (\hbar \omega_{\lambda})^2 v_{\lambda}^{\alpha} \mathbf{F}_{\lambda}^{\beta}, \quad (1)$$

in which α and β are the Cartesian coordinates, k_B is the Boltzmann constant, Ω is the volume of the unit cell, N is the number of discrete \mathbf{q} points of the Γ -centered \mathbf{q} grid for sampling the first Brillouin zone (FBZ), f is the Bose–Einstein distribution function, \hbar is the Planck's constant, ω is phonon frequency, v is phonon group velocity, and \mathbf{F} is usually referred to as “mean free displacement” [19]. The key to obtain κ_L through Eq. (1) is to calculate \mathbf{F}_{λ} using

$$\mathbf{F}_{\lambda} = \tau_{\lambda}^0 (\mathbf{v}_{\lambda} + \Delta_{\lambda}), \quad (2)$$

where τ_0 is the relaxation time, or lifetime, of phonon mode λ and Δ_{λ} is a complicated function of \mathbf{F} [19]. As a result, Eq. (2) has to be solved iteratively. Phonon relaxation time in solids can be limited by various phonon scattering mechanisms, for example, interaction with other phonons, isotopes, impurities, electrons, etc. Under the relaxation time approximation, the 3-phonon anharmonic scattering contribution to the phonon scattering rate γ_{λ} , or the inverse phonon relaxation time, is given by the Fermi's golden rule (FGR) as [20,21].

$$\begin{aligned} \gamma_{\lambda}^{pp} = & \frac{\hbar \pi}{4N} \sum_{\lambda_1 \lambda_2}^+ 2 \frac{f_1 - f_2}{\omega \omega_1 \omega_2} \left| V_{\lambda \lambda_1 \lambda_2}^+ \right|^2 \delta(\omega + \omega_1 - \omega_2) \\ & + \frac{\hbar \pi}{8N} \sum_{\lambda_1 \lambda_2}^- \frac{f_1 + f_2 + 1}{\omega \omega_1 \omega_2} \left| V_{\lambda \lambda_1 \lambda_2}^- \right|^2 \delta(\omega - \omega_1 - \omega_2). \end{aligned} \quad (3)$$

In the above equation, the first term on the right hand side denotes phonon absorption process, in which two phonons combine into one higher energy phonon; while the second term describes phonon emission process, in which one phonon splits into two lower energy phonons. In addition to the conservation of energy, the lattice or phonon quasi-momentum also needs to be conserved as $\mathbf{q}_2 = \mathbf{q} \pm \mathbf{q}_1 + \mathbf{Q}$ during the summation over phonon modes in Eq. (3). Here, \mathbf{Q} is the reciprocal lattice vector with $\mathbf{Q} = 0$ denoting normal process while $\mathbf{Q} \neq 0$ denoting Umklapp process. Besides, the Dirac function δ is approximated by a Gaussian function following the scheme in Ref. [19], even though the feasibility of Lorentzian function was also demonstrated previously [22]. The 3-phonon scattering matrix elements $V_{\lambda \lambda_1 \lambda_2}^{\pm}$ are calculated from the density-functional theory (DFT), of which the details can be found in Refs. [19,21,23,24], while the phonon band structures, or phonon dispersion relations, are calculated through the Fourier transform of the reciprocal-space dynamical matrices obtained from the linear-response theory [25,26]. We only consider three-phonon scattering processes in our calculations, as higher-order phonon scattering processes play an insignificant role in determining the lattice thermal conductivity of the superhard materials studied in this work [27].

In this work, we will consider materials composed of elements (C, B, or N) with natural isotopic distributions as well as isotopically pure materials. The contribution to phonon scattering rates by isotopic disorder can be captured by the well-established model

$$\gamma_{\lambda \lambda_1}^{iso} = \frac{\pi \omega^2}{2} \sum_{i=1}^N g(i) \left| \mathbf{e}_{\lambda}^*(i) \cdot \mathbf{e}_{\lambda_1}(i) \right|^2 \delta(\omega_{\lambda} - \omega_{\lambda_1}), \quad (4)$$

in which i sums over the unit cell. In the above equation, $g(i)$ characterizes the mass disorder among all isotopes as

$$g(i) = \sum_s f_s(i) \left[1 - \frac{m_s(i)}{\bar{m}(i)} \right]^2, \quad (5)$$

in which $f_s(i)$ and $m_s(i)$ denote the frequency of occurrence and the atomic mass of the s 'th isotope of atom i , respectively.

We conduct all the DFT calculations using the Quantum ESPRESSO package [25,26]. The Troullier-Martins type normconserving pseudopotentials [28] and the Perdew–Burke–Ernzerhof generalized gradient approximation (PBE-GGA) are used. Prior to calculating the force constants, we conduct structural relaxation

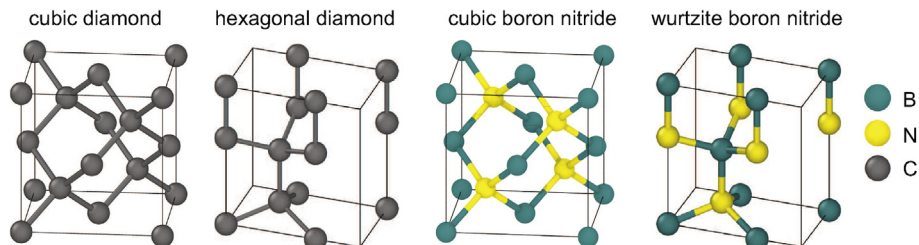


Fig. 1. Lattice structures of the four materials studied in this work. (A colour version of this figure can be viewed online.)

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