

Decoding thermal transport mechanisms induced by short-range order in amorphous system

Zhongwei Zhang,¹ Rui Ma,¹ Shuang Lu, Sebastian Volz,^{1,2} and Jie Chen^{1,*}¹*Center for Phononics and Thermal Energy Science, China-EU Joint Lab for Nanophononics, MOE Key Laboratory of Advanced Micro-structured Materials, School of Physics Science and Engineering, Tongji University, Shanghai 200092, People's Republic of China*²*Laboratory for Integrated Micro and Mechatronic Systems, CNRS-IIS UMI 2820, The University of Tokyo, Tokyo 153-8505, Japan*

(Received 15 March 2025; accepted 10 December 2025; published 7 January 2026)

Despite the absence of long-range structural order, amorphous solids retain an atomic-scale short-range ordered environment. However, the mechanisms through which short-range order influences vibrational modes and its subsequent effects on thermal conductivity remain elusive. In this work, we employ molecular dynamics and lattice dynamics simulations to investigate how short-range order modulates vibrational properties and thermal transport in amorphous silicon. Our simulations reveal that rapid cooling rates during the melt-quench process result in amorphous silicon structures with diminished short-range order. This reduction primarily intensifies the scattering of propagative vibrational modes for both transverse and longitudinal polarizations, particularly near the Ioffe-Regel crossover. Consequently, enhanced scattering weakens the mode correlation of propagative modes, further suppressing thermal conductivity. Additionally, our spatial coherence analysis demonstrates that reduced short-range order diminishes the wavelike behavior of propagative modes. Our findings provide a fundamental interpretation of the prevailing concept of mode correlation with the spatial coherence of thermal vibrations in amorphous systems, as well as physical insights into the role of short-range order in the wavelike nature of vibrational modes and the engineering of thermal transport in amorphous materials.

DOI: [10.1103/k5pw-jnl8](https://doi.org/10.1103/k5pw-jnl8)

I. INTRODUCTION

Amorphous solids are extensively used in various areas such as electronics [1], photovoltaics [2,3], and thermal insulation [4], owing to their unique thermal and electrical properties. Unlike crystals, where the long-range periodicity is essential to determine the thermal transport properties [5,6], amorphous solids lack the long-range order, which results in the breakdown of the phonon picture and thus brings challenges to the establishment of a comprehensive theory of thermal transport suitable for various solids [7–11]. Over the past few decades, numerous attempts have been made in this field [7–9,12–15]. Notable developments by Allen and coworkers [7,8] asserted that in amorphous systems, vibrational modes with closely spaced frequencies correlate with each other, leading to a significant contribution to thermal conductivity from quasilocalized modes. More recently, Isaeva and coworkers [13,16] introduced the quasiharmonic Green-Kubo (QH GK) method within a similar framework of mode correlation. This contribution from mode correlation has been demonstrated as a fundamental characteristic of thermal transport in amorphous or amorphouslike systems [12,17].

In amorphous materials, two kinds of vibrational modes primarily contribute to thermal conductivity: (1) propagons, which are low-frequency propagative modes, and (2)

diffusons, which are high-frequency quasilocalized modes [8,9,18]. Moon *et al.* [19,20] emphasized the predominance of propagative modes in amorphous silicon, uncovering that the elastic scattering of these modes dominates the acoustic collective excitations in this material. The study by Gelin *et al.* [21] on local elastic heterogeneities found the dependence of propagative modes on the local disorder. Moreover, Zhang *et al.* [22–24] examined the coherence characteristics of propagons and diffusons, revealing that diffusons can also exhibit wavelike characteristics and interact with local structural disorder in amorphous silicon. However, these works lack a clear link between the thermal conductivity and structure characteristics in amorphous solids. The underlying mechanism governing wavelike mode correlation in prevailing theories, as well as the dependence of vibrational modes and their correlation on variations in short-range order in amorphous materials, remains an open problem.

On the other hand, due to the absence of long-range order, conventional approaches [25–27] used to modulate the thermal conductivity of crystals through phonon-based models fail for amorphous materials [28–30]. Previous studies [31–34] have attempted to develop alternative methods to tune thermal transport in amorphous materials, such as band-order engineering and ultrafine nanostructuring [33,34]. However, the efficiency and impact of these methods on thermal conductivity remain limited. Unveiling the underlying mechanisms that can link intrinsic short-range order, which is the most crucial structural characteristic of amorphous materials, to thermal transport properties would unlock opportunities for engineering these amorphous materials for various applications.

In this work, we investigate the thermal transport properties of amorphous silicon with varying short-range order

*Contact author: jie@tongji.edu.cn

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

using lattice dynamics (LD) and molecular dynamics (MD) simulations. The thermal and vibrational properties of these amorphous structures are predicted and compared, clarifying how vibrational modes, particularly propagons and diffusons, respond to variations of short-range disorder. Furthermore, we unveil the underlying mechanism by which short-range order influences thermal conductivity through mode correlations, highlighting the importance of wavelike behavior in amorphous materials. Our study offers physical insights into the nature of vibrational modes and their contributions to thermal conductivity of amorphous solids.

II. COMPUTATIONAL APPROACH

A. Molecular dynamics simulations

We employ MD simulations to model the amorphous silicon system with different short-range orders and study their vibrational properties. All MD simulations are performed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) [35] with a time step of 0.5 fs. Periodic boundary conditions are applied in all directions. The Si-Si interaction in the amorphous silicon is described by the empirical Tersoff potential [36]. The melt-quench approach is adopted to generate the amorphous silicon [37,38]. Crystalline silicon structures, respectively, with 1728 atoms and 27 000 atoms, are first heated and melted at 4000 K for 500 ps under the isothermal-isobaric (NPT) ensemble. The system is then annealed to 300 K with different cooling rates, ranging from 0.1 to 130 K/ps, to generate amorphous silicon structures with various short-range orders. Each structure is equilibrated in both NPT and microcanonical (NVE) ensembles for 500 ps before further calculations. Following relaxation, the radial distribution function (RDF) is calculated [39] and averaged over 100 ps in the NVE ensemble. The coordination number is determined by counting neighboring atoms of each atom within a cutoff distance of 2.85 Å. This choice of cutoff is consistent with previous studies on amorphous silicon, capturing the essential nearest-neighbor interactions [38,40].

B. Vibrational properties

The characteristics of vibrational modes are further analyzed through the vibrational density of states (VDOS) and dynamic structure factor (DSF), which describe vibrational and collective excitation properties, respectively. When calculating VDOS and DSF, we use models consisting of 27 000 atoms to ensure high resolution in our analysis. The trajectories of atomic velocity and position are collected in an NVE ensemble every 0.025 ps for a total duration of 500 ps. VDOS is obtained from the Fourier transform of the autocorrelation of atomic velocities [41], while DSF is calculated as [42,43]

$$S_\alpha(\mathbf{q}, \omega) = \frac{|\mathbf{q}|^2}{2\pi\omega^2} \int dt \langle \mathbf{u}_\alpha(\mathbf{q}, t) \cdot \mathbf{u}_\alpha(\mathbf{q}, 0) \rangle e^{i\omega t}, \quad (1)$$

where $\mathbf{u}_\alpha$ is the collective velocity, α denotes the longitudinal (L) or the transverse (T) polarization, $\mathbf{q}$ refers to the quasi-wave vector, and ω is the angular frequency. The

time-dependent collective velocity $\mathbf{u}_\alpha$ can be further expressed as [42,43]

$$\mathbf{u}_L(\mathbf{q}, t) = \frac{1}{N} \sum_i (\mathbf{v}_i(t) \cdot \hat{\mathbf{q}}) \hat{\mathbf{q}} e^{i\mathbf{q} \cdot \mathbf{r}_i(t)}, \quad (2)$$

$$\mathbf{u}_T(\mathbf{q}, t) = \frac{1}{N} \sum_i [\mathbf{v}_i(t) - (\mathbf{v}_i(t) \cdot \hat{\mathbf{q}}) \hat{\mathbf{q}}] e^{i\mathbf{q} \cdot \mathbf{r}_i(t)}, \quad (3)$$

where $\hat{\mathbf{q}} = \mathbf{q}/|\mathbf{q}|$ is the normalized quasiwave vector, N represents the total number of atoms in the system, and $\mathbf{v}_i(t)$ and $\mathbf{r}_i(t)$ are the atomic velocity and position of i th atom, respectively. The linewidth and peak frequency of each vibrational mode at $\mathbf{q}$ is calculated by fitting with a damped harmonic oscillator model as [43,44]

$$S_\alpha(\mathbf{q}, \omega) = I_\alpha(\mathbf{q}) \frac{\Omega_\alpha^2(\mathbf{q}) \Gamma_\alpha(\mathbf{q})}{\omega^2 \Gamma_\alpha^2(\mathbf{q}) + [\omega^2 - \Omega_\alpha^2(\mathbf{q})]^2}, \quad (4)$$

where $I_\alpha(\mathbf{q})$, $\Gamma_\alpha(\mathbf{q})$, and $\Omega_\alpha(\mathbf{q})$ represent the fitted peak intensity, linewidth, and peak frequency, respectively. The linewidth serves as an analog to the inverse of phonon lifetime. When the linewidth becomes comparable to the central frequency, the propagative nature of the mode is lost. Therefore, we can determine the frequency of the Ioffe-Regel crossover when $\Omega_\alpha = \pi \Gamma_\alpha$ [20,43].

Participation ratio (PR) is calculated to examine the degree of localization of vibrational modes. The PR is defined as follows [9,45]:

$$PR(\mu) = \frac{[\sum_i e_i^2(\mu)]^2}{N \sum_i e_i^4(\mu)}, \quad (5)$$

where $\mathbf{e}_i(\mu)$ denotes the eigenvector of mode μ on atom i . This definition implies that extended modes are characterized by a high PR value, whereas localized modes have a low PR value.

C. Thermal transport properties

Thermal conductivities of the amorphous silicon are calculated using the QHGK method, which is a unified approach suitable for both ordered and disordered systems [13]. After extracting the second-order and third-order force constant matrices through the finite displacement method, eigenvectors and eigenvalues are computed within the framework of LD. Thermal conductivity κ is computed by using the QHGK method as [13]

$$\kappa = \frac{1}{3V} \sum_\alpha \sum_{\mu\mu'} c_{\mu\mu'} v_{\mu\mu'}^\alpha v_{\mu\mu'}^\alpha \tau_{\mu\mu'}, \quad (6)$$

where α is the Cartesian coordinate indices and V represents the system volume. The summation is performed over all modes μ and μ' . The terms $c_{\mu\mu'}$, $v_{\mu\mu'}^\alpha$, and $\tau_{\mu\mu'}$ correspond to the generalized heat capacity, α component of the generalized velocity, and generalized lifetime, respectively, as derived from the QHGK theory [13]. The generalized heat capacity $c_{\mu\mu'}$ is expressed as $c_{\mu\mu'} = \frac{\hbar\omega_\mu\omega_{\mu'}}{T} \frac{n_\mu - n_{\mu'}}{\omega_\mu - \omega_{\mu'}}$, where ω_μ is the frequency calculated from harmonic dynamical matrix and n_μ represents the Bose-Einstein distribution governing mode occupation at different temperatures according to quantum statistics. The generalized velocity is defined as

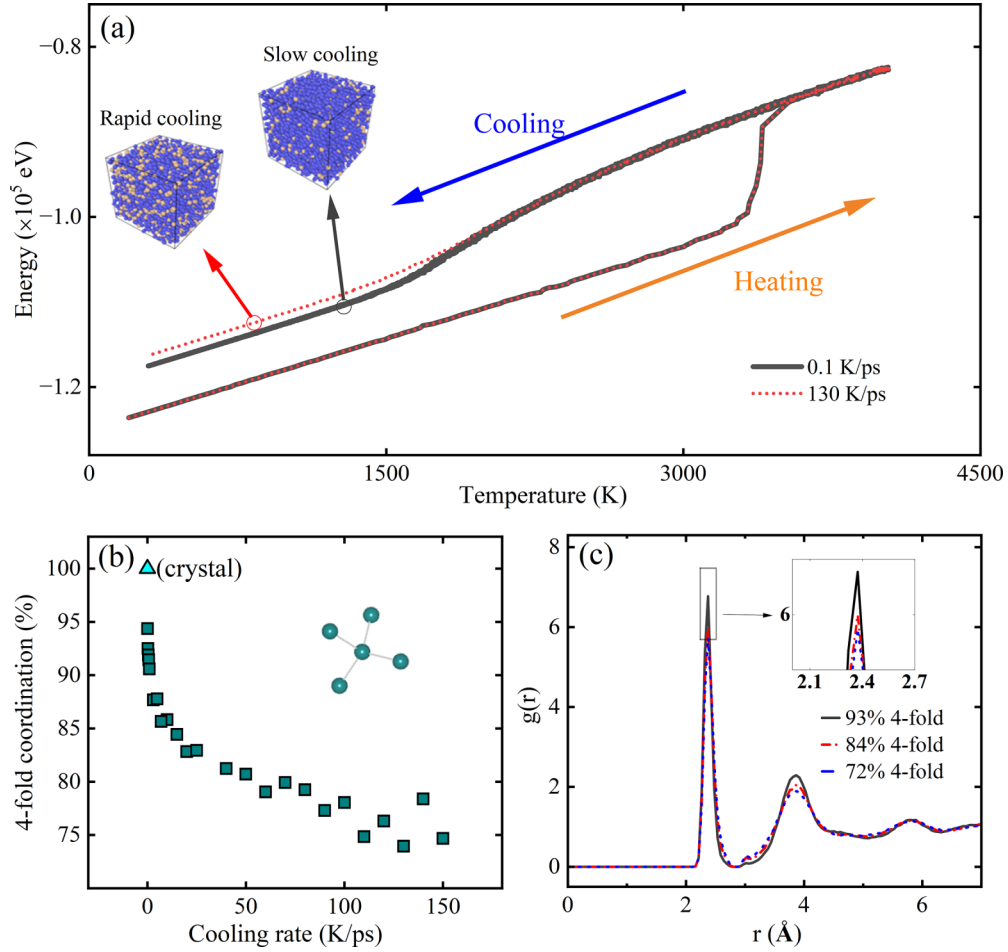


FIG. 1. (a) Schematic graph for the melt-quench process under different cooling rates used to prepare amorphous structures. Each line traces the energy of the structure as it is heated to a fully melted state and then cooled down to form a solid. Solid and dotted lines represent the cooling rates of 0.1 and 130 K/ps, which result in amorphous structures with 93% and 72% fourfold coordination, respectively. The inset shows the corresponding amorphous structures, where blue atoms have a coordination number of 4, and yellow atoms have other coordination numbers. (b) The percentage of fourfold coordinated atoms in different amorphous structures as a function of cooling rate. (c) Radial distribution function $g(r)$ for amorphous structures with 93%, 84%, and 72% percentages of fourfold coordination, respectively. The inset provides enlarged view of region at nearest-neighbor distance.

$v_{\mu\mu'}^\alpha = \frac{1}{2\sqrt{\omega_\mu\omega_{\mu'}}} \sum_{ij} (x_{ia} - x_{ja}) M_{ij}^{\alpha'\beta'} e_{\mu i}^{\alpha'} e_{\mu' j}^{\beta'}$, with the harmonic dynamical matrix entry $M_{ij}^{\alpha'\beta'}$ and its eigenvector component $e_{\mu i}^{\alpha'}$. The generalized lifetime $\tau_{\mu\mu'} = \frac{\Lambda_\mu + \Lambda_{\mu'}}{(\omega_\mu - \omega_{\mu'})^2 + (\Lambda_\mu + \Lambda_{\mu'})^2}$ weights the diffusive processes between vibrational modes with close frequencies, where Λ_μ is the linewidth of mode μ that can be computed using Fermi golden rule based on quantum statistics. This lattice-dynamics-based approach can inherently account for quantum statistics, as it includes both the temperature-dependent heat capacity and anharmonic scattering effects. Similar to the Allen and Feldman model [7,8], the diffusivity in the QHGK method represents the strength of mode correlations. The microscopic definition of mode diffusivity can be written as [7,16]

$$D_{\mu\mu'} = \frac{1}{3V} \sum_{\alpha} v_{\mu\mu'}^\alpha v_{\mu\mu'}^\alpha \tau_{\mu\mu'}. \quad (7)$$

The magnitude of the mode diffusivity describes the strength of correlation between two modes.

III. RESULTS AND DISCUSSION

A. Amorphous silicon with varying short-range order

In this work, the amorphous silicon systems with varying short-range order are obtained by altering the cooling rate during the melt-quench process in MD simulations. The melt-quench process preserves a nearly constant density across different cooling rates and avoids the introduction of artificial microstructures [28,31,32,38]. The full melt-quench process, from crystal to amorphous structures with different short-range orders, is shown in Fig. 1(a). A higher cooling rate results in structures with higher energy. As atoms are cooled down to reach their global energy minimum, rapid cooling causes the structures to retain atomic characteristics similar to those of the disordered liquid, resulting in higher energy [dotted line in Fig. 1(a)].

Due to the nature of covalent bonding in the diamond structure, silicon atoms tend to form bonds with four neighboring atoms, resulting in a fourfold coordinated environment [see the inset of Fig. 1(b)]. As shown in Fig. 1(a), the structure

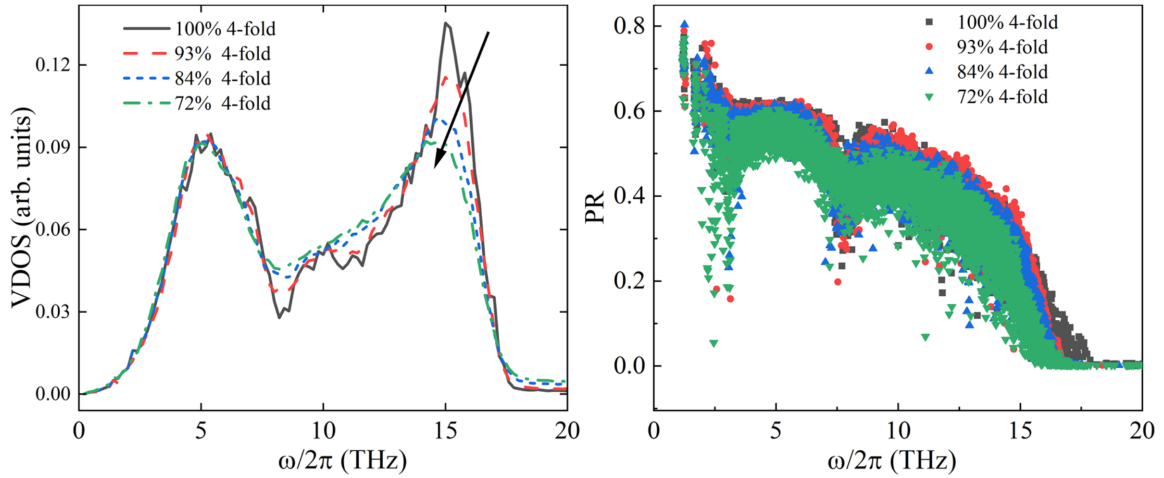


FIG. 2. (a) The VDOS and (b) PR of amorphous silicon with different short-range orders of 100%, 93%, 84%, and 72% fourfold coordination, respectively. The 100% fourfold structure is generated by the WWW algorithm, while the others are generated using the melt-quench process.

obtained with a cooling rate of 0.1 K/ps has lower energy and exhibits a higher proportion of fourfold coordinated atoms compared to the structure obtained with a cooling rate of 130 K/ps. This suggests that a higher cooling rate leads to a structure with reduced short-range order.

In amorphous solids, short-range order is inherently statistical, rather than governed by strict spatial symmetry. To systematically investigate the relationship between cooling rate and short-range order in amorphous silicon, we calculate the statistical ratio of fourfold coordinated atoms across structures with varying cooling rates. In ideal crystalline silicon, each atom is coordinated with four neighbors, yielding a 100% fourfold coordination ratio. Therefore, deviations from this 100% ratio can be used to monitor the decrease in short-range order. Moreover, the local bond order parameters have been employed to characterize the short-range order in various systems [46,47]. However, we find that under the same extent of random variation in amorphous silicon, the fourfold coordination number is a direct and more sensitive indicator for the short-range order compared to the local bond-order parameters. Therefore, the fourfold coordination number is used in this work to measure the variation of short-range order induced by various cooling rates. As shown in Fig. 1(b), the ratio of fourfold coordination exhibits a decreasing trend when the cooling rate increases. At very high cooling rates, this reduction becomes erratic, reflecting the unrelaxed atomic configurations caused by the rapid quenching. This dependence demonstrates that higher cooling rates produce less ordered short-range environments, suggesting that the rapid cooling prevents atoms from settling into more ordered (i.e., crystalline) configurations.

The RDF is also calculated to study the structural properties of these amorphous structures. Figure 1(c) shows the RDF for three structures with different short-range order characterized by 93%, 84%, and 72% fourfold coordination, which are associated with cooling rates of 0.1, 10, and 130 K/ps, respectively. The absence of distinct peaks after the distance of the second nearest neighbor confirms the loss of long-range order across all amorphous structures, indicating no residual

signatures from the initial crystalline state. The inset in Fig. 1(c) further shows the suppression of the first-neighbor peak as the fourfold coordination decreases, which corresponds to the reduction of the short-range order. This is because atoms have sufficient time to settle into their global energy minima with a lower cooling rate, resulting in a more ordered atomic environment.

There are several methods to experimentally control short-range order in amorphous solids, such as annealing amorphous structures at different temperatures [48,49] or adjusting the temperature during the deposition process [50]. These studies share a common principle: they modulate the short-range order by accelerating or decelerating the annealing process in the amorphous material. Our work follows a similar approach, where varying the annealing time (by changing the cooling rate) effectively modulates the short-range order.

B. Modal analysis

To further investigate the vibrational properties under different short-range order, we conduct a modal analysis combining both MD and LD methods. We start with a conventional analysis using the VDOS to provide a comprehensive view of the distribution of vibrational modes, together with the PR to elucidate the nature of vibrational states. Additionally, a model generated using the Wooten-Winer-Weaire algorithm [51], reported to have 100% fourfold coordination, is also included in the calculations.

The VDOS in Fig. 2(a) demonstrates that as the degree of short-range order decreases, low-frequency modes below 6 THz remain largely unchanged, while modes at high frequencies (~ 15 THz) exhibit a noticeable redshift with reduced amplitude [see arrow in Fig. 2(a)], thereby increasing the density of states in the middle-frequency range. This behavior suggests that low-frequency modes, which are typically associated with phononlike behavior and relatively long wavelengths, are not significantly affected by changes in short-range order. In contrast, the modes at higher frequencies, usually corresponding to more localized vibrational

states, shift toward middle frequencies, and the VDOS peak density is also reduced. To further explore the localization characteristics of the vibrational modes, the PR value is also calculated, as shown in Fig. 2(b). Except for those modes near the zone center and edge, the majority of the modes exhibit an overall decrease in PR with decreasing fourfold coordination ratio, indicating an enhanced localization as short-range order diminishes. Figure 2(b) also shows a relatively high PR value for structures with 100% fourfold coordination at high frequencies, particularly near the frequency of localized modes. The combined analysis suggests two key findings: First, long-wavelength modes remain largely unaffected by the reduction in short-range order. Second, despite the redshift of high-frequency modes to middle frequencies, the overall vibrational states tend to become more localized as short-range order decreases.

Due to the absence of lattice periodicity in amorphous materials, the notion of normal modes may lose its validity [10,18,52]. Previous studies [19,22] have shown that collective excitation, a concept borrowed from similar disordered systems like liquids or liquidlike solids [42], provides a more appropriate description of thermal vibrations in amorphous solids. In amorphous silicon, thermal vibrations can be characterized by collective excitations through the DSF [20,53–55] (see method in Sec. II B). In this regard, we analyze DSF spectrum for amorphous silicon structures with different short-range orders.

Our calculation results of the DSF for structures with 72% and 93% fourfold coordination ratios are shown in Fig. 3. The DSF results in Figs. 3(a)–3(e) show that modes with low frequency still maintain a dispersionlike relation between the wave vector and frequency, which suggests that they retain phononlike characteristics. The sound velocities can be obtained by calculating the first derivative of DSF spectrum at very low frequency, yielding a longitudinal sound velocity of 7797 m/s and a transverse sound velocity of 4436 m/s. Both values are comparable to those of crystalline silicon [56], indicating that the modes at low frequencies exhibit propagative characteristics similar to phonons. This phononlike low-frequency range corresponds to the range where the VDOS does not vary significantly with decreasing short-range order [see Fig. 2(a)].

With the increase of wave vector, the vibrational frequency is broadened to a wider range for both polarizations, giving rise to a notable linewidth at each wave vector. When the linewidth increases dramatically to a value comparable with the central frequency (see in methods), this corresponds to the Ioffe-Regel crossover (horizontal lines in Fig. 3) beyond which the dispersionlike feature becomes very blurred as those vibrational states are heavily scattered by structural disorder in amorphous materials, leading to a transition from propagons to diffusons. Figures 3(c) and 3(f) further show that the Ioffe-Regel crossover is associated with a sharp broadening of the linewidth for both longitudinal and transverse modes, as highlighted by the dashed circles. Since the Ioffe-Regel crossover originates from the breakdown of long-range periodicity during the transition from crystalline to amorphous structures [55], this crossover frequency remains nearly unchanged despite variations in short-range order.

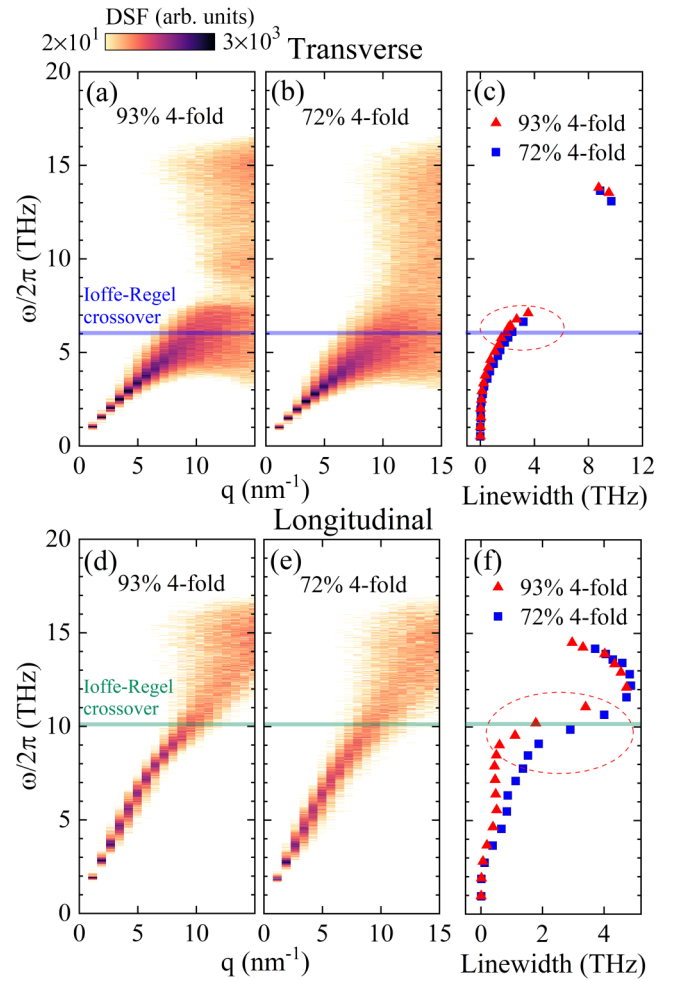


FIG. 3. DSF for transverse polarization of amorphous silicon with (a) 93% and (b) 72% fourfold coordination, respectively. (c) The linewidth of transverse polarization vs frequency for the structures with 93% and 72% fourfold coordination. DSF for longitudinal polarization of (d) 93% and (e) 72% fourfold coordination. (f) The linewidth of longitudinal polarization vs frequency for the structures with 93% and 72% fourfold coordination. The linewidths in panels (c) and (f) are fitted at specific wave vectors in panels (a), (b), (d), and (e). The Ioffe-Regel crossover is indicated by the horizontal lines for structure with 93% fourfold coordination, and the dashed circles highlight the regions where the linewidth changes rapidly.

Furthermore, longitudinal modes in amorphous silicon maintain a clear relation between wave vector and frequency up to 10 THz [Fig. 3(d)], whereas the transverse modes exhibit a clear dispersion only up to 6 THz [Fig. 3(a)], showing a larger linewidth compared to that of longitudinal modes. This difference reveals that the transverse modes are more sensitive to the loss of long-range order, which is consistent with literature results [57,58]. With the reduction in short-range order as indicated by the decrease in fourfold coordination, it leads to the enhanced linewidth for frequencies below the Ioffe-Regel crossover, especially for the longitudinal polarization. Such enhancement in the linewidth is more pronounced for those modes close to the Ioffe-Regel crossover [see dashed circles in Fig. 3(f)]. These findings suggest that the decrease of short-range order in amorphous silicon has a

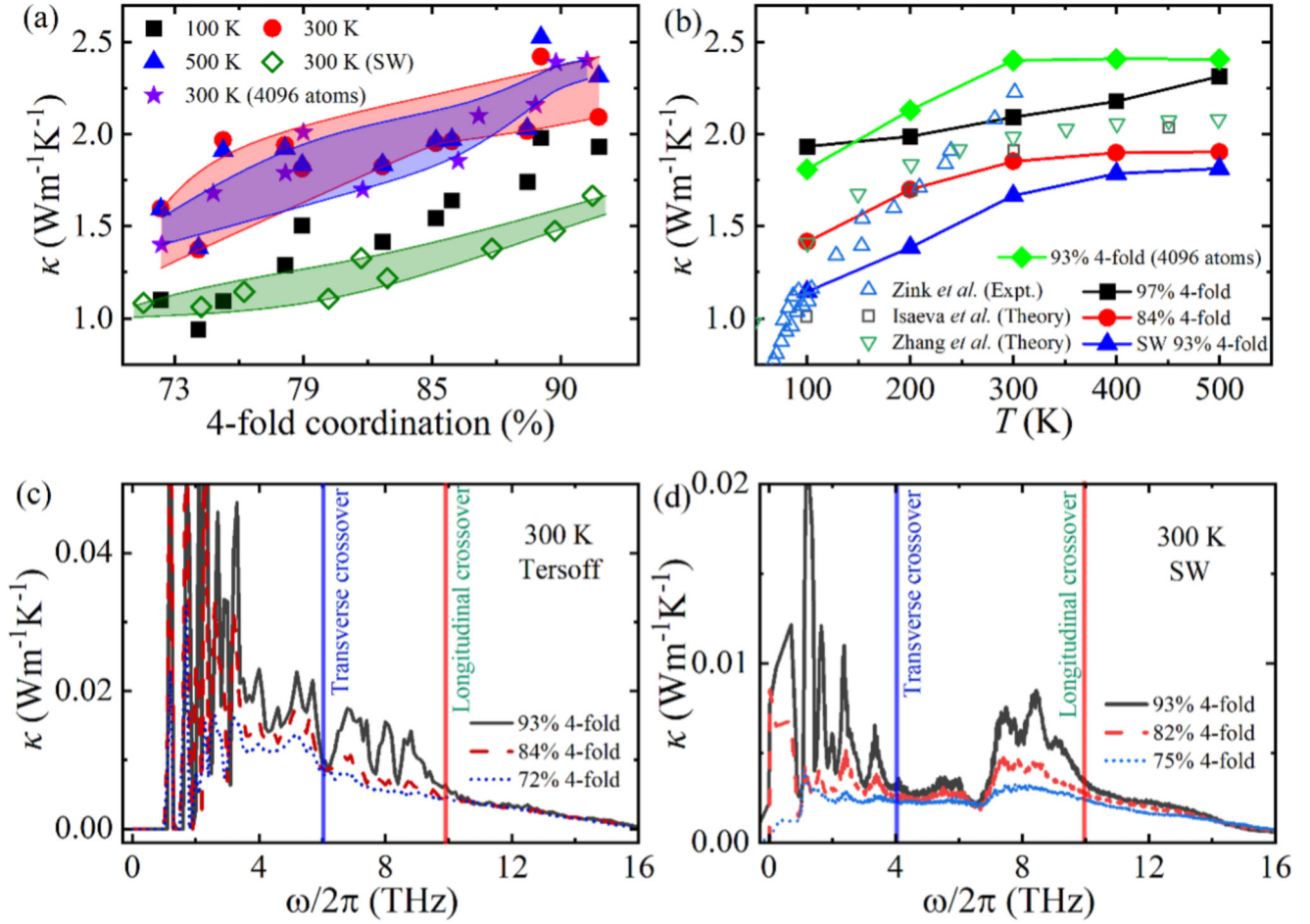


FIG. 4. (a) Thermal conductivity κ of amorphous silicon as a function of fourfold coordination, calculated using the Tersoff potential (filled symbols) and the Stillinger-Weber (SW) potential (open diamonds) for 1728 atoms. Stars denote results obtained with the Tersoff potential for 4096 atoms at 300 K. The shaded region shows the envelope of the data, highlighting the overall increasing trend of κ with fourfold coordination. (b) κ of amorphous silicon under different conditions as a function of temperature. The literature results from experimental [59] (Expt.) and theoretical (Theory) [13,22] works are also shown for comparison. Panels (c) and (d) show the spectral κ for varying fourfold coordination at 300 K computed using the Tersoff and SW potentials, respectively. Vertical lines mark the Ioffe-Regel crossover frequencies for different polarizations.

more pronounced impact on propagons rather than diffusons, and the resultant enhancement in linewidth primarily targets longitudinal polarization.

C. Thermal transport properties

We compute thermal conductivity κ of amorphous silicon at temperature of 100, 300 and 500 K using the QHGK method within the lattice dynamics framework [according to Eq. (6)]. Figure 4(a) illustrates that at all three temperatures, κ of amorphous silicon exhibits an increasing trend with increasing short-range order. For instance, at room temperature, when the short-range order (characterized by the fourfold coordination ratio) varies from 72% to 93%, κ is enhanced by as large as 77%. It should be noted that the intrinsic stochasticity of the MD-based melt-quench process leads to structural variations in the amorphous systems, even at a fixed fourfold coordination, which consequently causes a scattered distribution of κ . To better capture the overall variation trend, we plot the data envelopes as shaded regions in Fig. 4(a). These envelopes

clearly demonstrate a general increasing trend of κ with the increase of fourfold coordination fraction, a tendency that is consistently observed across different interatomic potentials and system sizes. However, this enhanced κ is still much lower than that of crystalline state, indicating the amorphous nature of these structures with different fourfold coordination. This enhancement demonstrates that short-range order can also dramatically influence the thermal transport in amorphous systems, although the loss of long-range order is often considered the primary factor in reducing κ of amorphous solids compared to the crystalline case. To avoid the finite size effect, we perform additional calculations on a larger model containing 4096 atoms at 300 K, and consistent results are obtained [see stars in Fig. 4(a)], indicating that a simulation model of 1728 atoms is sufficiently large. Furthermore, we also validate our findings against the choice of interatomic potentials, and find that the simulation results obtained using Stillinger-Weber (SW) potential [see empty diamonds in Fig. 4(a)] also reveal a similar increasing trend in thermal conductivity with a higher fourfold coordination at 300 K,

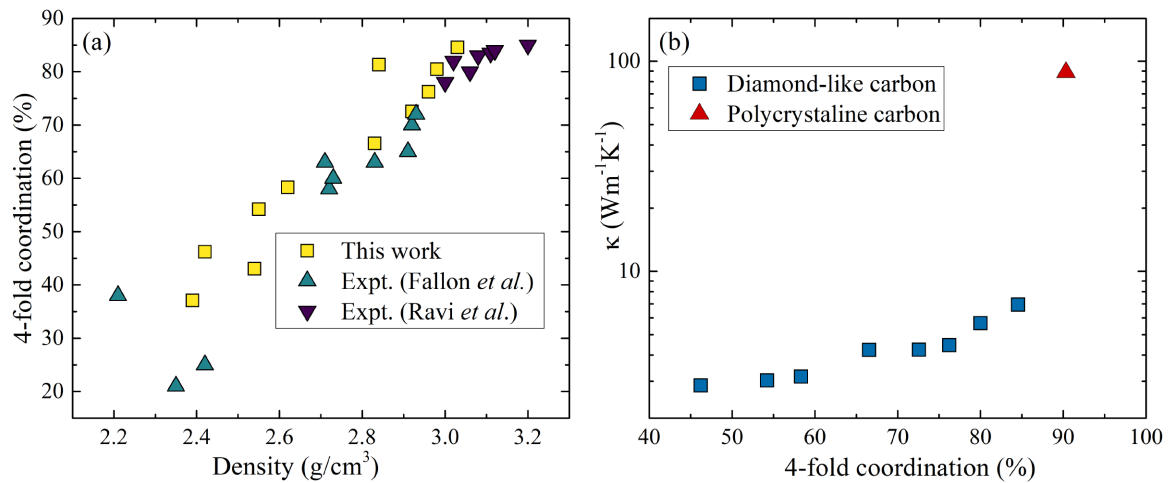


FIG. 5. (a) The fourfold coordination ratio as a function of density for diamondlike amorphous carbon obtained from simulations (squares) in this work and experiments (triangles) by Fallon *et al.* in Ref. [63] and Ravi *et al.* in Ref. [64]. (b) Thermal conductivity of amorphous and polycrystalline carbon as a function of fourfold coordination ratio.

showing its independence on the choice of interatomic potentials.

Figure 4(b) further presents the temperature-dependent κ of amorphous silicon, examining the influence factors of short-range order, system size, and interatomic potential. A general increase in κ with temperature is observed across all conditions, which aligns well with trends reported in literature [13,22,59]. Specifically, κ exhibits a significant increase as the temperature rises from 100 to 300 K for various degrees of short-range order, but its growth becomes weaker at higher temperature (300–500 K). Crucially, κ of the amorphous structure with 84% fourfold coordination exhibits close agreement with reported experimental values, suggesting this particular degree of short-range order might closely resemble the experimental state. Furthermore, the degree of temperature dependence is shown to be contingent upon the fourfold coordination fraction, revealing a unique approach for engineering thermal transport via short-range order. These findings collectively indicate that manipulation of the short-range order, for instance via the control of cooling rate, offers an effective route to modulate κ in amorphous materials. Although the same short-range order can be measured and modulated experimentally [48–50], its effect on the thermal properties of amorphous solids has not yet been rigorously tested. Our approach offers a promising strategy for designing amorphous materials with tailored thermal properties for specific applications, extending beyond the conventional phonon transport framework.

Similar relationships between atomic coordination and κ have also attracted experimental interest in carbon systems [60,61], particularly in diamondlike carbon. To generalize our findings, we further generate diamondlike amorphous carbon structures with sp^3 coordination ratio ranging from 45% to 85%, as well as a polycrystalline carbon structure composed of four grains. In our MD simulations, the sp^3 coordination ratio is determined by counting atoms bonded to four neighbors within the first-neighbor cutoff distance, which corresponds to the definition of fourfold coordination in our work. The diamondlike carbon structures are generated by using a modified

Tersoff potential that accurately captures both the structural and mechanical properties of diamondlike carbon [62]. The ratio of fourfold coordinated atoms is controlled by adjusting the density during the melt-quench process at a cooling rate of 1 K/ps. The relationship between density and coordination ratio in diamondlike amorphous carbon is shown in Fig. 5(a). Our simulation results (squares) agree well with experimental (triangles) trends [63,64]. The polycrystalline carbon structure is constructed by using a Voronoi tessellation algorithm, which enables the realistic grain boundary formation [65]. As shown in Fig. 5(b), the κ of diamondlike amorphous carbon increases with the sp^3 coordination ratio (i.e., the fourfold coordination ratio), consistent with our findings in silicon systems. Compared to the diamondlike amorphous carbon, the polycrystalline carbon exhibits a significantly higher κ , which can be attributed to both the high sp^3 coordination ratio and its partially crystalline structure.

Further insights can be obtained by examining the spectral contributions to the total κ . Figure 4(c) reveals a declining trend in spectral κ as short-range order diminishes, with significant differences observed below 10 THz. Based on the DSF analysis in Fig. 3, modes below 6 THz are identified as propagons for both polarizations, while modes between 6 and 10 THz include propagons for longitudinal polarization and diffusons for transverse polarization. This indicates that it is challenging to disentangle the relative contributions of propagons and diffusons to κ within the QHGK framework. However, our previous work [22] based on a heat conduction theory including coherence [23] demonstrated that in amorphous silicon with a fixed high proportion of fourfold coordination, propagative modes dominate thermal transport, contributing up to 66% of the total κ at 300 K. Notably, the coherence terms—represented by the off-diagonal elements in the QHGK formalism—account for 68% of the total κ at 300 K, highlighting their critical role in the thermal transport process.

Since the reduction in short-range order mainly affects the linewidth of longitudinal propagon [Fig. 3(f)] rather than transverse diffuson [Fig. 3(c)] within the frequency between

6 and 10 THz, the spectral κ analysis highlights an important finding: It is predominantly the propagons that respond to the decrease in short-range order, driving the observed reduction in thermal conductivity. This conclusion is also consistent with negligible variation of spectral κ contributions with varying short-range order for high-frequency modes above 10 THz, which correspond to the diffusons and locons for both polarizations. The consistency of these findings is further validated using the SW potential, as shown in Fig. 4(d). While the SW potential yields different κ values compared to the Tersoff potential, the underlying mechanism by which the short-range order impacts the modal-level thermal properties remains consistent across different degrees of short-range order. In addition, it also highlights the deficiency of VDOS and PR-based analysis: They lack the modal transport information and thus are insufficient to understand the influence of short-range order on spectral contribution, although notable variations in VDOS and PR are observed in Fig. 2 for high-frequency modes above 10 THz.

To further elucidate how different vibrational modes, particularly their correlations, contribute to thermal transport and their dependence on short-range order, we examine the mode diffusivity in our system. Analogous to the Allen-Feldman model [7,8], the mode diffusivity in the QHGK approach describes a unique thermal transport behavior in disordered systems, showing that heat is carried not only by propagative modes but also by diffusive modes through correlations between different modes. Figure 6 illustrates that the intermode correlations, i.e., off-diagonal terms, are predominantly concentrated near the diagonal line, indicating that only modes with close frequencies can correlate with each other in amorphous solids. This mechanism is different from the observations in complex crystals that retain the phonon picture, where mutual correlation between phonon modes can extend to those even with large frequency differences [12,66].

In addition, strong correlations occur in amorphous silicon at frequencies below 4 THz and within the range between 7 and 9 THz, reflecting the significant involvement of propagons in mode correlation. The magnified figures on the right panels of Fig. 6 further reveal that mode correlations within these frequency ranges are significantly suppressed as the short-range order decreases. Similar to phonon coherence in crystals, where coherence is sensitive to the loss of long-range order [67], the coherence between vibrational modes in amorphous materials also strongly depends on short-range order. This reduced coherence, coupled with the increased linewidth of propagative modes, suggests that enhanced structural disorder significantly hampers thermal transport in amorphous systems.

D. Spatial coherence analysis

Previous studies have demonstrated that coherence or mode correlation originates from the unique wavelike nature of thermal vibrations, as extensively studied in crystals and nanophononic crystals [67]. Moreover, as reported by Gelin *et al.* [21] and Shintani and Tanaka [43], the wave behavior of various vibrational modes in amorphous materials can be visualized by projecting the eigenvector field onto the real space. In this regard, we define the spatial wave coherence

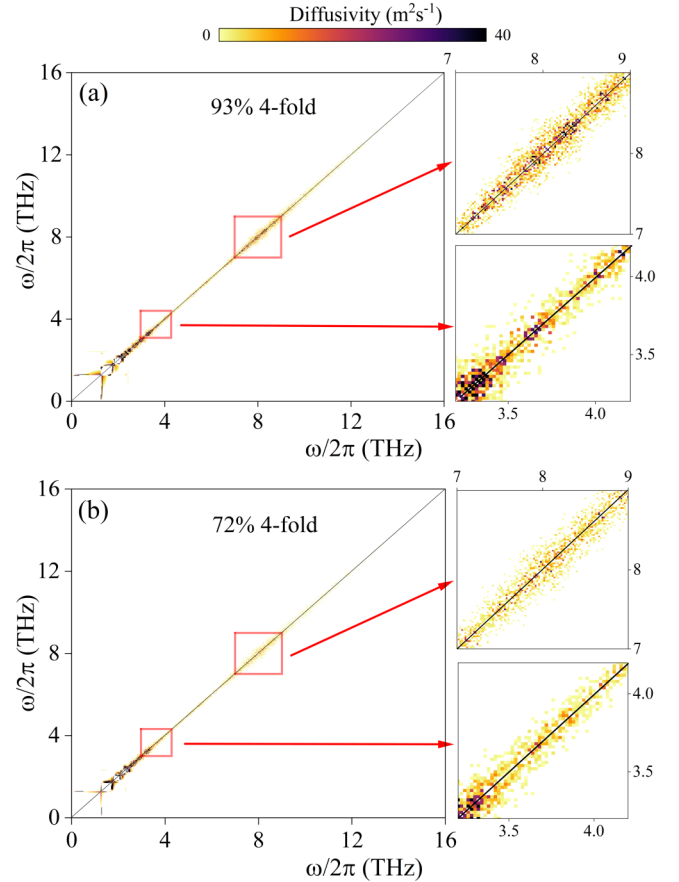


FIG. 6. Mode diffusivity of amorphous silicon with (a) 93% and (b) 72% fourfold coordination. Right panels are magnified views of the square regions.

$\Psi_i(\mu)$ of mode μ on atom i as [57]

$$\Psi_i(\mu) = \frac{1}{N_i} \sum_j^{N_i} \frac{\mathbf{e}_i(\mu) \cdot \mathbf{e}_j(\mu)}{|\mathbf{e}_i(\mu)| |\mathbf{e}_j(\mu)|}, \quad (8)$$

where N_i is the number of nearest neighbors for atom i and $\mathbf{e}_i(\mu)$ refers to eigenvector of mode μ on atom i . The summation is performed over all the nearest neighbors j of atom i . The value of $\Psi_i(\mu)$ ranges between 1 and -1 , with $\Psi_i(\mu) = 1$ denoting that the eigenvector component on an atom has the same direction as its neighboring atoms, while $\Psi_i(\mu) = -1$ indicates the opposite direction (out-of-phase). A $\Psi_i(\mu)$ value near zero suggests incoherent motion between atoms. Therefore, the amplitude of $\Psi_i(\mu)$ indicates the extent to which the component of mode μ on atom i exhibits the wavelike behavior. In this work, we define the degree of spatially continuous and extended vibrations as the spatial “wavelike” behavior, which is reflected by the large positive $\Psi_i(\mu)$ values.

Our DSF results in Fig. 3 reveal that the dispersionlike feature is distinct between the propagons and diffusons. In order to examine their wave behavior, we select three representative frequencies of 3.6, 8, and 11 THz, which correspond to the propagons, mixed vibrons (transverse diffusons and longitudinal propagons), and diffusons, respectively. We exclude the modes above 16 THz (locons) because their vibrations only involve a few atoms, preventing them from exhibiting

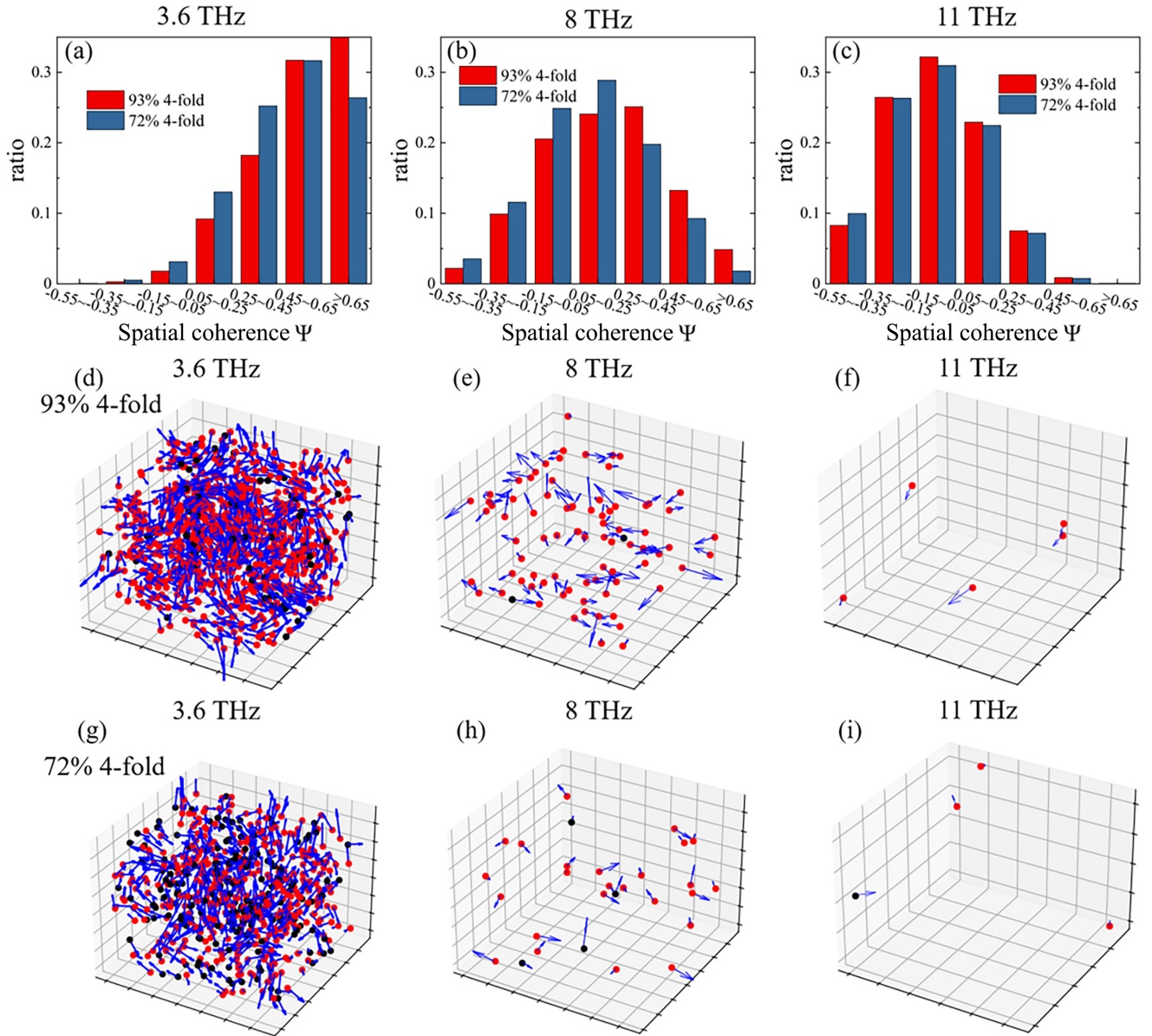


FIG. 7. The spatial coherence Ψ distribution for amorphous structures with 93% and 72% fourfold coordination at frequencies of (a) 3.6 THz, (b) 8 THz, and (c) 11 THz. Eigenvector fields of eigenvectors with $\Psi > 0.65$ for the structure with 93% fourfold coordination at frequencies of (d) 3.6 THz, (e) 8 THz, and (f) 11 THz, and for the structure with 72% fourfold coordination at frequencies of (g) 3.6 THz, (h) 8 THz, and (i) 11 THz. The red and black dots denote atoms with fourfold coordination and other coordination numbers, respectively.

substantial wavelike behavior [8,18]. Figures 7(a)–7(c) compare the spatial coherence distribution for amorphous structures with 93% and 72% fourfold coordination at three representative frequencies. When the frequency varies from 3.6 to 11 THz, a lower spatial coherence is consistently observed, since the peak values of $\Psi_i(\mu)$ distribution progressively shift to the low-value region. This dependence indicates that the wavelike behaviors of thermal vibrations in amorphous materials decline at higher frequencies. In contrast to the standing waves of high-frequency optical modes in crystals that consist of out-of-phase vibrations [with the large negative $\Psi_i(\mu)$], previous studies [22,57,68] demonstrated that structural disorder significantly suppresses the spatial “wavelike” behavior (i.e., spatial coherence) of these modes. Therefore, the modes with the large negative $\Psi_i(\mu)$ values in

Figs. 7(a)–7(c) indicate the loss of spatial coherence in amorphous silicon. Besides, very small $\Psi_i(\mu)$ values correspond to highly localized vibrations with minimal correlation between neighboring atoms.

In order to make more detailed comparisons between different frequencies, we select a relatively large threshold value (e.g., $\Psi = 0.65$), above which indicates a strong wavelike behavior. Figures 7(d)–7(i) depict the eigenvector distribution in real space above this threshold for three representative frequencies, with the red and black dots denoting atoms with fourfold coordination and other coordination numbers, respectively. Obviously, the number of eigenvector components exhibiting high spatial coherence decreases at higher frequencies. Additionally, as short-range order decreases, these components are significantly reduced for propagons at 3.6 THz

[Figs. 7(d) and 7(g)] and mixed vibrons at 8 THz [Figs. 7(e) and 7(h)], while negligible variation is observed for diffusons at 11 THz [Figs. 7(f) and 7(i)]. These results clearly explain that the mode correlation of propagons is more sensitive to short-range order than that of the diffusons from a wavelike perspective. This finding agrees very well with the enhanced linewidth in Fig. 3(f) and reduced κ spectrum in Figs. 4(c) and 4(d) for propagons when the short-range order decreases.

Moreover, the atomic coordination information reveals that wavelike behaviors are primarily associated with atoms having fourfold coordination. This indicates that thermal vibrations become more coherent and wavelike among atoms within higher short-range ordered environments. For high-frequency diffusons in Figs. 7(f) and 7(i), the wavelike component becomes negligible, suggesting that these diffusive vibrations seldom interact through short-range order in amorphous materials.

IV. CONCLUSION

We have constructed amorphous silicon structures with different short-range orders by varying the cooling rates during the melt-quench process in MD simulations. The computed thermal transport properties of these amorphous structures via QHGK method reveal that thermal conductivity of amorphous silicon depends significantly on the short-range structural environment. Our vibrational mode analysis using VDOS and PR demonstrates that modes with middle and high frequencies are particularly sensitive to changes in short-range order. The DSF and spectral thermal conductivity analysis together elucidate the underlying mechanism responsible for the suppressed thermal conductivity with the loss of short-range order. As short-range order diminishes, the linewidth of propagons near the Ioffe-Regel crossover increases notably, leading to the significant suppression of spectral contribution to total thermal conductivity from propagons in both transverse and longitudinal polarizations. Furthermore, eigenvector field analysis

further reveals that wavelike behaviors are predominantly associated with atoms exhibiting fourfold coordination, which are suppressed by the reduced short-range order. This coherence among atoms with similar short-range environments contributes to strong wavelike collective behavior in vibrational modes. These findings suggest that mode correlation in different prevailing theories originates from the picture of spatial phase correlation, and that manipulating short-range order through controlled cooling rates can effectively modulate the thermal conductivity of amorphous systems. Our results also emphasize the importance of collective and wavelike behaviors on investigating vibrational and thermal transport properties in amorphous solids.

ACKNOWLEDGMENTS

This project was supported in part by the grants from the National Natural Science Foundation of China (Grants No. 12205220 and No. 12475037), the Science and Technology Commission of Shanghai Municipality (Grant No. 24520711200), the Basic Research Program of Jiangsu (BK20244002), the Fundamental Research Funds for the Central Universities (Grants No. 22120250138 and No. 22120230212), and the Special Funds of the Tongji University for “Sino-German Cooperation 2.0 Strategy” (Grant No. ZD2023005). J.C. was supported by the Shuguang Program of Shanghai Education Development Foundation and Shanghai Municipal Education Commission (Grant No. 23SG18).

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible, and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

-
- [1] L. Yi, Y. Zhang, K. Nie, B. Li, Y. Yuan, Z. Liu, and W. Huang, Recent advances in the engineering and electrochemical applications of amorphous-based nanomaterials: A comprehensive review, *Coord. Chem. Rev.* **501**, 215569 (2024).
 - [2] S. Sreejith, J. Ajayan, S. Kollem, and B. Sivasankari, A comprehensive review on thin film amorphous silicon solar cells, *Silicon* **14**, 8277 (2022).
 - [3] B. Wang, G. M. Biesold, M. Zhang, and Z. Lin, Amorphous inorganic semiconductors for the development of solar cell, photoelectrocatalytic and photocatalytic applications, *Chem. Soc. Rev.* **50**, 6914 (2021).
 - [4] M. C. Wingert, J. Zheng, S. Kwon, and R. Chen, Thermal transport in amorphous materials: A review, *Semicond. Sci. Technol.* **31**, 113003 (2016).
 - [5] J. Chen, J. He, D. Pan, X. Wang, N. Yang, J. Zhu, S. A. Yang, and G. Zhang, Emerging theory and phenomena in thermal conduction: A selective review, *Sci. China Phys. Mech. Astron.* **65**, 117002 (2022).
 - [6] W. Ren, S. Lu, C. Yu, J. He, Z. Zhang, J. Chen, and G. Zhang, Impact of Moiré superlattice on atomic stress and thermal transport in van der Waals heterostructures, *Appl. Phys. Rev.* **10**, 41404 (2023).
 - [7] P. B. Allen and J. L. Feldman, Thermal conductivity of disordered harmonic solids, *Phys. Rev. B* **48**, 12581 (1993).
 - [8] J. L. Feldman, M. D. Kluge, P. B. Allen, and F. Wooten, Thermal conductivity and localization in glasses: Numerical study of a model of amorphous silicon, *Phys. Rev. B* **48**, 12589 (1993).
 - [9] P. B. Allen, J. L. Feldman, J. Fabian, and F. Wooten, Diffusons, locons, propagons: Character of atomic vibrations in amorphous Si, *Philos. Mag. B* **79**, 1715 (1999).
 - [10] W. Lv and A. Henry, Examining the validity of the phonon gas model in amorphous materials, *Sci. Rep.* **6**, 37675 (2016).
 - [11] W. Lv and A. Henry, Non-negligible contributions to thermal conductivity from localized modes in amorphous silicon dioxide, *Sci. Rep.* **6**, 35720 (2016).
 - [12] M. Simoncelli, N. Marzari, and F. Mauri, Unified theory of thermal transport in crystals and glasses, *Nat. Phys.* **15**, 809 (2019).
 - [13] L. Isaeva, G. Barbalinardo, D. Donadio, and S. Baroni, Modeling heat transport in crystals and glasses from a

- unified lattice-dynamical approach, *Nat. Commun.* **10**, 3853 (2019).
- [14] Q. Xi, Z. Zhang, J. Chen, J. Zhou, T. Nakayama, and B. Li, Hopping processes explain T-linear rise of thermal conductivity in thermoelectric clathrates above the plateau, *Phys. Rev. B* **96**, 064306 (2017).
- [15] Q. Xi, Z. Zhang, T. Nakayama, J. Chen, J. Zhou, and B. Li, Off-center rattling triggers high-temperature thermal transport in thermoelectric clathrates: Nonperturbative approach, *Phys. Rev. B* **97**, 224308 (2018).
- [16] G. Barbalinardo, Z. Chen, N. W. Lundgren, and D. Donadio, Efficient anharmonic lattice dynamics calculations of thermal transport in crystalline and disordered solids, *J. Appl. Phys.* **128**, 135104 (2020).
- [17] M. Simoncelli, N. Marzari, and F. Mauri, Wigner formulation of thermal transport in solids, *Phys. Rev. X* **12**, 041011 (2022).
- [18] H. R. Seyf and A. Henry, A method for distinguishing between propagons, diffusions, and locons, *J. Appl. Phys.* **120**, 25101 (2016).
- [19] J. Moon, B. Latour, and A. J. Minnich, Propagating elastic vibrations dominate thermal conduction in amorphous silicon, *Phys. Rev. B* **97**, 024201 (2018).
- [20] J. Moon, R. P. Hermann, M. E. Manley, A. Alatas, A. H. Said, and A. J. Minnich, Thermal acoustic excitations with atomic-scale wavelengths in amorphous silicon, *Phys. Rev. Mater.* **3**, 065601 (2019).
- [21] S. Gelin, H. Tanaka, and A. Lemaître, Anomalous phonon scattering and elastic correlations in amorphous solids, *Nat. Mater.* **15**, 1177 (2016).
- [22] Z. Zhang, Y. Guo, M. Bescond, J. Chen, M. Nomura, and S. Volz, How coherence is governing diffusion heat transfer in amorphous solids, *npj Comput. Mater.* **8**, 96 (2022).
- [23] Z. Zhang, Y. Guo, M. Bescond, J. Chen, M. Nomura, and S. Volz, Heat conduction theory including phonon coherence, *Phys. Rev. Lett.* **128**, 015901 (2022).
- [24] Z. Zhang, Y. Guo, M. Bescond, J. Chen, M. Nomura, and S. Volz, Generalized decay law for particlelike and wavelike thermal phonons, *Phys. Rev. B* **103**, 184307 (2021).
- [25] C. Yu, Y. Hu, J. He, S. Lu, D. Li, and J. Chen, Strong four-phonon scattering in monolayer and hydrogenated bilayer BAs with horizontal mirror symmetry, *Appl. Phys. Lett.* **120**, 132201 (2022).
- [26] L. Shi, J. Chen, G. Zhang, and B. Li, Thermoelectric figure of merit in Ga-doped [0001] ZnO nanowires, *Phys. Lett. A* **376**, 978 (2012).
- [27] W.-J. Ren, S. Lu, C.-Q. Yu, J. He, and J. Chen, Carbon honeycomb structure with high axial thermal transport and strong robustness, *Rarer Met.* **42**, 2679 (2023).
- [28] A. Hashemi, H. Babaei, and S. Lee, Effects of medium range order on propagon thermal conductivity in amorphous silicon, *J. Appl. Phys.* **127**, 45109 (2020).
- [29] T. Kawasaki, T. Araki, and H. Tanaka, Correlation between dynamic heterogeneity and medium-range order in two-dimensional glass-forming liquids, *Phys. Rev. Lett.* **99**, 215701 (2007).
- [30] Y. Shang, *et al.*, Enhancement of short/medium-range order and thermal conductivity in ultrahard sp^3 amorphous carbon by C70 precursor, *Nat. Commun.* **14**, 7860 (2023).
- [31] A. Giri, C. J. Dionne, and P. E. Hopkins, Atomic coordination dictates vibrational characteristics and thermal conductivity in amorphous carbon, *npj Comput. Mater.* **8**, 55 (2022).
- [32] S. Thakur, C. J. Dionne, P. Karna, S. W. King, W. Lanford, H. Li, S. Banerjee, D. Merrill, P. E. Hopkins, and A. Giri, Density and atomic coordination dictate vibrational characteristics and thermal conductivity of amorphous silicon carbide, *Phys. Rev. Mater.* **6**, 094601 (2022).
- [33] Y. Liao and J. Shiomi, Akhiezer mechanism dominates relaxation of propagons in amorphous material at room temperature, *J. Appl. Phys.* **130**, 035101 (2021).
- [34] N. Tambo, Y. Liao, C. Zhou, E. M. Ashley, K. Takahashi, P. F. Nealey, Y. Naito, and J. Shiomi, Ultimate suppression of thermal transport in amorphous silicon nitride by phononic nanostructure, *Sci. Adv.* **6**, eabc0075 (2020).
- [35] A. P. Thompson, H. M. Aktulga, R. Berger, D. S. Bolintineanu, W. M. Brown, P. S. Crozier, P. J. in't Veld, A. Kohlmeyer, S. G. Moore, T. D. Nguyen, *et al.*, LAMMPS—A flexible simulation tool for particle-based materials modeling at the atomic, meso, and continuum scales, *Comput. Phys. Commun.* **271**, 108171 (2022).
- [36] J. Tersoff, Empirical interatomic potential for silicon with improved elastic properties, *Phys. Rev. B* **38**, 9902 (1988).
- [37] F. H. Stillinger and T. A. Weber, Computer simulation of local order in condensed phases of silicon, *Phys. Rev. B* **31**, 5262 (1985).
- [38] I. Santos, M. Aboy, L. A. Marqués, P. López, and L. Pelaz, Generation of amorphous Si structurally compatible with experimental samples through the quenching process: A systematic molecular dynamics simulation study, *J. Non-Cryst. Solids* **503–504**, 20 (2019).
- [39] Y. Ma, Z. Zhang, J. Chen, K. Sääskilähti, S. Volz, and J. Chen, Ordered water layers by interfacial charge decoration leading to an ultra-low Kapitza resistance between graphene and water, *Carbon* **135**, 263 (2018).
- [40] V. L. Deringer, N. Bernstein, A. P. Bartók, M. J. Cliffe, R. N. Kerber, L. E. Marbella, C. P. Grey, S. R. Elliott, and G. Csányi, Realistic atomistic structure of amorphous silicon from machine-learning-driven molecular dynamics, *J. Phys. Chem. Lett.* **9**, 2879 (2018).
- [41] X. Peng, P. Jiang, Y. Ouyang, S. Lu, W. Ren, and J. Chen, Reducing Kapitza resistance between graphene/water interface via interfacial superlattice structure, *Nanotechnology* **33**, 035707 (2021).
- [42] J. P. Boon, S. Yip, and J. K. Percus, Molecular hydrodynamics, *Phys. Today* **34**, 92 (1981).
- [43] H. Shintani and H. Tanaka, Universal link between the boson peak and transverse phonons in glass, *Nat. Mater.* **7**, 870 (2008).
- [44] H. R. Schober and C. Oligschleger, Low-frequency vibrations in a model glass, *Phys. Rev. B* **53**, 11469 (1996).
- [45] H. R. Seyf, W. Lv, A. Rohskopf, and A. Henry, The importance of phonons with negative phase quotient in disordered solids, *Sci. Rep.* **8**, 2672 (2018).
- [46] W. Lechner and C. Dellago, Accurate determination of crystal structures based on averaged local bond order parameters, *J. Chem. Phys.* **129**, 114707 (2008).
- [47] A. Antidormi, L. Colombo, and S. Roche, Thermal transport in amorphous graphene with varying structural quality, *2D Mater.* **8**, 015028 (2021).
- [48] E. Holmström, B. Haberl, O. H. Pakarinen, K. Nordlund, F. Djurabekova, R. Arenal, J. S. Williams, J. E. Bradby,

- T. C. Petersen, and A. C. Y. Liu, Dependence of short and intermediate-range order on preparation in experimental and modeled pure a-Si, *J. Non-Cryst. Solids* **438**, 26 (2016).
- [49] J. Fortner and J. S. Lannin, Structural relaxation and order in ion-implanted Si and Ge, *Phys. Rev. B* **37**, 10154 (1988).
- [50] H. C. Jacks, M. Molina-Ruiz, M. H. Weber, J. J. Maldonis, P. M. Voyles, M. R. Abernathy, T. H. Metcalf, X. Liu, and F. Hellman, Structural tunability and origin of two-level systems in amorphous silicon, *Phys. Rev. Mater.* **6**, 045604 (2022).
- [51] F. Wooten, K. Winer, and D. Weaire, Computer generation of structural models of amorphous Si and Ge, *Phys. Rev. Lett.* **54**, 1392 (1985).
- [52] J. Moon, Examining normal modes as fundamental heat carriers in amorphous solids: The case of amorphous silicon, *J. Appl. Phys.* **130**, 55101 (2021).
- [53] J. L. Feldman, Calculations of the generalized dynamic structure factor for amorphous silicon, *J. Non-Cryst. Solids* **307–310**, 128 (2002).
- [54] K. Sturm, Dynamic structure factor: An introduction, *Z. Naturforsch. A* **48**, 233 (2014).
- [55] T. Kim, J. Moon, and A. J. Minnich, Origin of micrometer-scale propagation lengths of heat-carrying acoustic excitations in amorphous silicon, *Phys. Rev. Mater.* **5**, 065602 (2021).
- [56] S. Neogi and D. Donadio, Thermal transport in free-standing silicon membranes: Influence of dimensional reduction and surface nanostructures, *Eur. Phys. J. B* **88**, 73 (2015).
- [57] Y.-C. Hu and H. Tanaka, Origin of the boson peak in amorphous solids, *Nat. Phys.* **18**, 669 (2022).
- [58] H. Liu, X. Shi, F. Xu, L. Zhang, W. Zhang, L. Chen, Q. Li, C. Uher, T. Day, and G. J. Snyder, Copper ion liquid-like thermoelectrics, *Nat. Mater.* **11**, 422 (2012).
- [59] B. L. Zink, R. Pietri, and F. Hellman, Thermal conductivity and specific heat of thin-film amorphous silicon, *Phys. Rev. Lett.* **96**, 055902 (2006).
- [60] M. Shamsa, W. L. Liu, A. A. Balandin, C. Casiraghi, W. I. Milne, and A. C. Ferrari, Thermal conductivity of diamond-like carbon films, *Appl. Phys. Lett.* **89**, 161921 (2006).
- [61] J. Robertson, Diamond-like amorphous carbon, *Mater. Sci. Eng. R Rep.* **37**, 129 (2002).
- [62] Z. D. Sha, P. S. Branicio, Q. X. Pei, V. Sorkin, and Y. W. Zhang, A modified Tersoff potential for pure and hydrogenated diamond-like carbon, *Comput. Mater. Sci.* **67**, 146 (2013).
- [63] P. J. Fallon, V. S. Veerasamy, C. A. Davis, J. Robertson, G. A. J. Amaratunga, W. I. Milne, and J. Koskinen, Properties of filtered-ion-beam-deposited diamondlike carbon as a function of ion energy, *Phys. Rev. B* **48**, 4777 (1993).
- [64] S. Ravi, P. Silva, S. Xu, B. X. Tay, H. S. Tan, and W. I. Milne, Nanocrystallites in tetrahedral amorphous carbon films, *Appl. Phys. Lett.* **69**, 491 (1996).
- [65] S. Falco, J. Jiang, F. De Cola, and N. Petrinic, Generation of 3D polycrystalline microstructures with a conditioned La guerre-Voronoi tessellation technique, *Comput. Mater. Sci.* **136**, 20 (2017).
- [66] Q.-Y. Xie, P.-F. Liu, J.-J. Ma, L.-M. Wu, K.-W. Zhang, and B.-T. Wang, Microscopic mechanisms of glasslike lattice thermal conductivity in tetragonal α -CsCu₅Se₃, *Phys. Rev. B* **108**, 014302 (2023).
- [67] S. Hu, Z. Zhang, P. Jiang, W. Ren, C. Yu, J. Shiomi, and J. Chen, Disorder limits the coherent phonon transport in two-dimensional phononic crystal structures, *Nanoscale* **11**, 11839 (2019).
- [68] R. Hu and Z. Tian, Direct observation of phonon Anderson localization in Si/Ge aperiodic superlattices, *Phys. Rev. B* **103**, 045304 (2021).