

Bond-Network Entropy Governs Heat Transport in Coordination-Disordered SolidsKamil Iwanowski^{1,2}, Gábor Csányi³ and Michele Simoncelli^{1,2,*}¹*Department of Applied Physics and Applied Mathematics,
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Understanding how the vibrational and thermal properties of solids are influenced by atomistic structural disorder is of fundamental scientific interest and paramount to designing materials for next-generation energy technologies. While several studies indicate that structural disorder strongly influences the thermal conductivity, the fundamental physics governing the disorder-conductivity relation remains elusive. Here we show that order-of-magnitude, disorder-induced variations of conductivity in network solids can be predicted from a “bond-network” entropy, an atomistic structural descriptor that quantifies heterogeneity in the topology of the atomic-bond network. We employ the Wigner formulation of thermal transport to demonstrate the existence of a relation between the bond-network entropy and observables such as smoothness of the vibrational density of states and macroscopic conductivity. We also show that the smoothing of the vibrational density of states encodes information about the thermal resistance induced by disorder and can be directly related to phenomenological models for phonon-disorder scattering based on the semiclassical Peierls-Boltzmann equation. Our findings rationalize the conductivity variations of disordered carbon polymorphs ranging from nanoporous electrodes to defective graphite used as a moderator in nuclear reactors.

DOI: [10.1103/w4p6-b9mp](https://doi.org/10.1103/w4p6-b9mp)Subject Areas: Computational Physics,
Condensed Matter Physics,
Materials Science**I. INTRODUCTION**

Carbon forms a variety of disordered allotropes useful in several industrial applications: Amorphous carbon is a promising material for next-generation electronic and mechanical technologies due to its disorder-tunable electrical conductivity [1], high elastic modulus, and hardness [2]; nanoporous carbon is employed as structural nanomaterial [3], as well as in devices for energy harvesting [4,5] or water desalination [6,7]; nuclear-grade defective graphite is used as a moderator in nuclear reactors [8–10]. This wide range of applications is made possible by the high variability in the macroscopic properties of carbon, which derive from its versatility at the atomic scale [11], particularly its ability to form chainlike bonds between two atoms, planar (graphitelike) bonds involving three atoms, and tetrahedral (diamondlike) bonds between four atoms. The thermal conductivity is a striking example of such

high variability [12–14], as it can change by more than 4 orders of magnitude—from the ultralow (<1 W/mK) conductivity of nanoporous carbon [15] to the ultrahigh (>2000 W/mK) conductivity of graphene [16–22], graphite [23–31], and diamond [32,33]. Past works studied structural disorder and thermal conductivity in specific classes of carbon polymorphs, which include (i) experiments, e.g., in amorphous carbon [34–40], graphite with variable degree of irradiation-induced defects [41–51], and nanoporous carbon with different pore-size distributions [15], and (ii) molecular dynamics (MD) simulations [52–54], e.g., in amorphous carbon [55–61], electron-irradiated graphite [9], and nanoporous carbon [59,61,62]. However, each of those past studies focused on one (or few) of the aforementioned special classes of carbon polymorphs, without investigating similarities and differences between their atomistic structure and macroscopic conductivity. As a result, the structure-conductivity relation is far from being fully understood.

Here, we address this long-standing question relying on the Wigner formulation of thermal transport [63–65] in conjunction with the machine-learned Gaussian approximation potential (GAP) [66]. We employ the latter to describe with quantitative accuracy structural, vibrational, and thermal properties of 23 disordered carbon polymorphs belonging to five different classes: amorphous, irradiated,

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phase separated, variable porosity, and nanoporous carbide-derived carbon. We find that disorder in the atomic-bond network induces order-of-magnitude variations in the thermal conductivity. To quantify such disorder and its relation with conductivity, we decompose the solid into a collection of local atomic environments (LAEs), and we show that in carbon these can be comprehensively characterized using the H_1 barcode—a ring-based structural descriptor which is formally defined in terms of the first homology group [67]. After discussing how the size (i.e., number of atoms n) in the LAE determines the resolution with which structural heterogeneity can be resolved, we define a bond-network entropy (BNE) that, in the presence of disorder, grows with the LAE's size n . We demonstrate that the BNE's growth rate with n [$\text{BNE}(n)/n$] quantifies disorder in the atomic-bond network, distinguishing the disordered phases of carbon. Most importantly, we rely on these insights to elucidate how disorder affects observables such as the smoothness of the vibrational density of states (VDOS) [68] and the macroscopic thermal conductivity [34,36–38]. Finally, we rationalize the physics underlying the correlation between $\text{BNE}(n)/n$ and conductivity by formally connecting the disorder-induced repulsion between vibrational eigenstates to the thermal resistance caused by disorder. Specifically, from the smoothness of the VDOS and diffusivity, we determine the transport length scales for all vibrational excitations in irradiated graphite, showing that the characterization of transport length scales is not limited to low-frequency modes that approximately display a band structure blurred by disorder—i.e., a dynamical structure factor (DSF), reminiscent of three acoustic bands with disorder-dependent linewidth [57,69–71]. Ultimately, we elucidate the disorder-conductivity relation across a comprehensive range of disordered carbon polymorphs, paving the way for the theory-driven optimization of technologies ranging from nanoporous-carbon-based supercapacitors to nuclear reactors.

II. WIGNER FORMULATION OF THERMAL TRANSPORT

To investigate the atomistic physics underlying the conductivity of disordered carbon, we employ the Wigner transport equation (WTE) [64]. Such an equation accounts for the quantum Bose-Einstein statistics of vibrations, anharmonicity, and disorder; its solution yields a thermal conductivity expression that comprehensively describes solids ranging from crystals to glasses [65,72]. In the following, we employ the regularized WTE conductivity expression [65] to ensure a proper bulk-limit extrapolation in the presence of disorder:

$$\kappa = \frac{1}{\mathcal{V}N_c} \sum_{\mathbf{q},s,s'} \frac{\omega_{qs} + \omega_{qs'}}{4} \left(\frac{C_{qs}}{\omega_{qs}} + \frac{C_{qs'}}{\omega_{qs'}} \right) \frac{\|\mathbf{v}(\mathbf{q})_{s,s'}\|^2}{3} \times \pi \mathcal{F}_{[\eta;\Gamma_{qs}+\Gamma_{qs'}]}(\omega_{qs} - \omega_{qs'}), \quad (1)$$

where $\mathcal{V}$ is the volume of the reference cell of the solid, s is a mode index, and $\mathbf{q}$ is a wave vector that labels vibrations—the necessity to consider $\mathbf{q}$ depends on the size and degree of disorder in the solid; for strongly disordered systems described with a very large ($\gtrsim 5000$ atoms) reference cell, it is sufficient to consider only $\mathbf{q} = \mathbf{0}$ (see Fig. 4 in Ref. [65] and Secs. III A and IV D in Ref. [73]; we discuss size effects also in Fig. 1). ω_{qs} and C_{qs} are the frequency and the quantum specific heat, respectively, of the vibration $\mathbf{q}s$:

$$C_{qs} = C[\omega_{qs}, T] = \frac{\hbar^2 \omega_{qs}^2}{k_B T^2} N_{qs} (N_{qs} + 1), \quad (2)$$

where N_{qs} is the Bose-Einstein distribution at temperature T : $N_{qs} = [\exp(\hbar\omega_{qs}/k_B T) - 1]^{-1}$. $\mathbf{v}(\mathbf{q})_{s,s'}$ is the velocity operator—its diagonal elements are the usual group velocities, while its off-diagonal nondegenerate elements describe couplings between different vibrations. $\mathcal{F}_{[\eta;\Gamma_{qs}+\Gamma_{qs'}]}$ is a two-parameter Voigt distribution, employed to numerically preserve in finite-size models of disordered solids the physical property that neighboring (quasidegenerate) vibrational modes can interact and conduct heat even in the limit of vanishing intrinsic linewidths Γ_{qs} (due to third-order anharmonic interactions and isotope-mass impurities). Specifically, η is a computational parameter that has to be numerically converged [65,73] and is of the order of the average energy-level spacings; see Supplemental Material [74] for details. When $\Gamma_{qs} + \Gamma_{qs'} \gg \eta$, the Voigt distribution reduces to a Lorentzian $L[\omega, \Gamma] = (1/\pi) \{(\Gamma/2)/[\omega^2 + (\Gamma/2)^2]\}$ with full width at half maximum (FWHM) Γ determined by the sum of the intrinsic linewidths of mode $\mathbf{q}s$ and $\mathbf{q}s'$, while in the opposite limit $\Gamma_{qs} + \Gamma_{qs'} \ll \eta$ it reduces to the Gaussian representation of the Dirac delta. In formulas,

$$\mathcal{F}_{[\eta;\Gamma_{qs}+\Gamma_{qs'}]} \rightarrow \begin{cases} L[\omega_{qs} - \omega_{qs'}, \Gamma_{qs} + \Gamma_{qs'}], & \text{for } \Gamma_{qs} + \Gamma_{qs'} \gg \eta, \\ \frac{1}{\pi\eta} \exp\left(-\frac{1}{\pi} \frac{(\omega_{qs} - \omega_{qs'})^2}{\eta^2}\right), & \text{for } \Gamma_{qs} + \Gamma_{qs'} \ll \eta. \end{cases} \quad (3)$$

Equation (3) implies that, for $\Gamma_{qs} + \Gamma_{qs'} \gg \eta$, Eq. (1) yields the standard anharmonic WTE conductivity [64], while in the opposite limit $\Gamma_{qs} + \Gamma_{qs'} \ll \eta$ Eq. (1) numerically reduces to the harmonic Allen-Feldman (AF) [104] conductivity.

III. CONDUCTIVITY OF DISORDERED CARBON POLYMORPHS

Figure 1 shows the room-temperature conductivity of five structurally different classes of carbon polymorphs, obtained evaluating Eq. (1) with the quantum-accurate GAP [66]. In particular, carbide-derived carbon (CDC) is a low-density (≤ 1.0 g/cm³) solid consisting of nanoporous, curved

graphene sheets with tunable amount of coordination defects [105,106]. CDC draws its name from the carbide precursors (TiC or SiC) used in the chlorination-based synthesis procedure [15,75]; its properties depend on the chlorination temperature, which is, therefore, reported in the structure name [75] (e.g., CDC800 and CDC1200 refer to materials synthesized with a chlorination temperature of 800 and 1200 °C, respectively, and ann-CDC1200 denotes a CDC1200 structure further annealed after synthesis; see Supplemental Material [74]). Variable-porosity carbon (VPC) is constituted of curved graphene sheets with variable porosity and rare coordination defects (see Supplemental Material [74] for details on coordination number distributions); their density ranges from 0.9 to 1.9 g/cm³ [59,76,77,107]. Amorphous carbon (AC) is a packed structure with variable density from 1.5 to 3.5 g/cm³, where atoms have coordination number that ranges from two to four and its average increases with density [36,37,55,57,61]. Irradiated graphite (IRG) features interlayer bonding defects (interstitial atoms, dislocations, and point defects) caused by electron or neutron irradiation, in concentration proportional to the irradiation time [9]. Finally, the phase-separated phase (PSP) structure blends a mostly threefold coordinated graphitic phase with a mostly fourfold coordinated amorphous-carbon phase [77].

The room-temperature conductivity of these polymorphs varies by more than one order of magnitude, from 0.5 W/mK for CDC800 to 8.1 W/mK for IRG with the lowest irradiation time (“IRG T2” corresponds to an exposure to irradiation of 2 min). Notably, we find that the thermal conductivity-density relation highly depends on the type of disorder. For AC and VPC, increasing density causes an increase in conductivity, which is faster in VPC compared to AC (the former shows 69% decrease in κ upon decreasing density from 1.9 to 0.9 g/cm³, while the latter shows 56% decrease in κ upon decreasing density from 2.9 to 1.5 g/cm³). In CDC, the structural details—determined by the chlorination temperature used in the synthesis [75]—have a weak (negligible) influence on the density but strongly impact the thermal conductivity, which decreases by 53% upon decreasing the chlorination temperature from 1200 to 800 °C. Similar conductivity variations at nearly constant density are observed in IRG structures, where increasing the irradiation exposure from 2 to 9 min causes a conductivity reduction of 56%. Lastly, we see that annealing the maximum-density AC phase (2.9 g/cm³) yields partial graphitization (PSP phase) and a conductivity increase of 44%.

Figure 1 also shows that our nanometric atomistic models are sufficiently large to describe the bulk limit of the conductivity [65,69,70,73,108,109]; in fact, for all the polymorphs, we studied multiple atomistic models of sizes differing by more than one order of magnitude and always found compatible results for their conductivity. Importantly, we highlight that our conductivity predictions

are broadly compatible in both magnitude and density dependence with several independently performed experiments and simulations. Specifically, for AC our predictions match experiments by Hurler, Pietralla, and Hammerschmidt [35] at densities of 1.8 and approximately 2.0 g/cm³, and MD simulations by Giri, Dionne, and Hopkins [55], after accounting for quantum corrections [110] (see Appendix A for details on the accuracy of the quantum correction applied to the MD conductivity predictions). Our predictions for AC also overlap with the broad density-conductivity relation experimentally observed by Bullen *et al.* [36], Shamsa *et al.* [37], Morath *et al.* [34], Hurler, Pietralla, and Hammerschmidt [35], and Scott *et al.* [38]; we note that the dispersion of the experimental values may originate from several factors, including presence of hydrogen impurities (especially at low density [36,38]). These impurities can increase the effective mass of carbon atoms [34], cause material softening [39], and promote disorder in the bond network [78]—all these factors contribute to decrease in thermal conductivity [111] and explain the tendency of our theoretical predictions for structures made purely of carbon to be an upper bound for the experimental values. For CDC, our predictions are compatible with the experiments by Kern *et al.* [15] at density of approximately 0.68 g/cm³ (samples synthesized from TiC₂ precursor) matching the increase of conductivity with chlorination temperature—specifically, CDC synthesized at 1200 °C (CDC1200) has a higher conductivity than CDC synthesized at 800 °C (CDC800) in both theory and experiments. We also note that the density of the experimental samples by Kern *et al.* [15] is lower than the density of our CDC models, and to extrapolate the predicted conductivity at the experimental density (dashed lines in Fig. 1) we employ a simple density-based extrapolation originating from the inverse volume dependence in Eq. (1), obtaining $\kappa = \rho_{\text{exp}}(\kappa_{\text{ref}}/\rho_{\text{ref}})$, where ρ_{exp} denotes the experimental density, ρ_{ref} the theoretical density, and κ_{ref} the predicted conductivity at the theoretical density. We have validated the accuracy of this density-based conductivity extrapolation procedure for nanoporous carbon (NPC): Applying it to estimate the conductivity of low-density NPC from predictions at high density (both ours and those by Wang *et al.* [61]) yields extrapolations that are consistent with explicit calculations in low-density NPC. We justify this further in the next sections, showing that, for the carbon polymorphs analyzed, the value of κ/ρ is related to the degree of structural disorder. Focusing on our conductivity predictions for the VPC class, for ann-CDC1200 our prediction is in agreement with simulations from Wang *et al.* [61] for densities between 0.75 and 1.0 g/cm³; for VPC(D) 1.9, we are compatible with measurements in irradiated graphite by Barabash *et al.* [47] and Maruyama and Harayama [41]; finally, for VPC(D)/(T) 1.5, we match quantum-corrected predictions from Suarez-Martinez and Marks [59] and

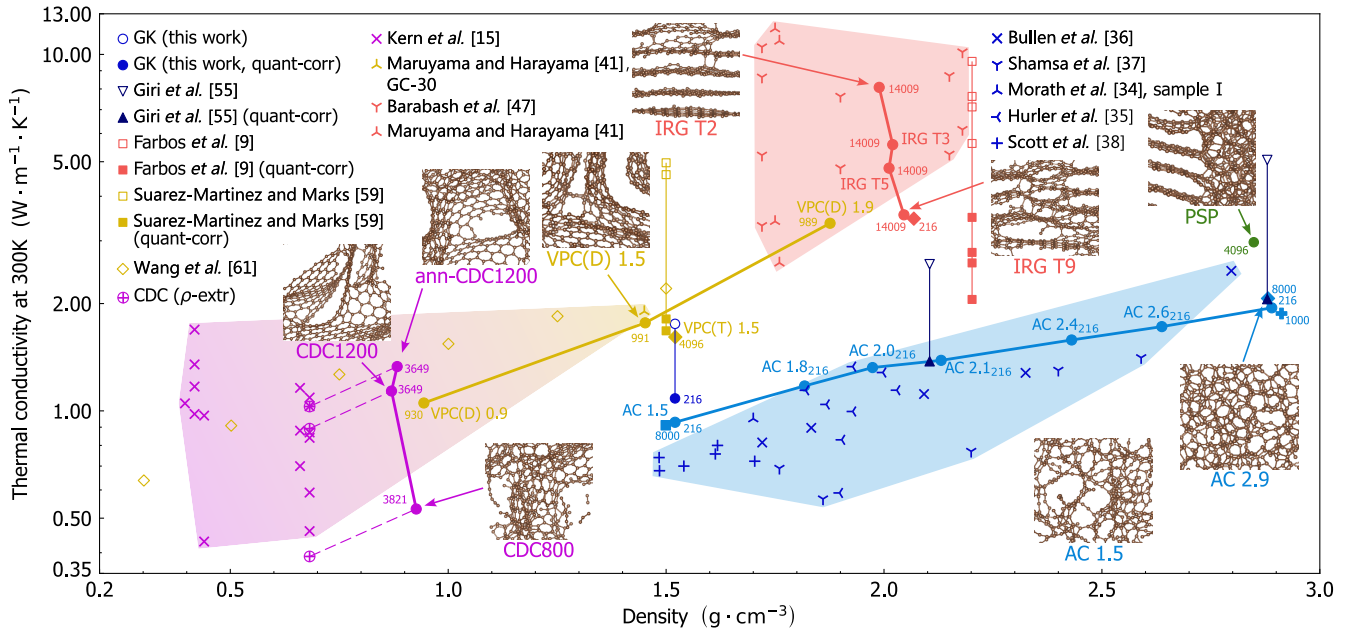


FIG. 1. Room-temperature conductivity as a function of density in various classes of carbon polymorphs. Carbide-derived carbon (CDC) is purple, variable-porosity carbon (VPC) is yellow, amorphous carbon (AC) is blue, irradiated graphite (IRG) is red, and phase-separated phase (PSP) is green. Filled markers are results from this work; empty markers are direct results from MD simulations (irradiated graphite from Farbos *et al.* [9], nanoporous carbon from Wang *et al.* [61] and Suarez-Martinez and Marks [59], and amorphous carbon from Giri, Dionne, and Hopkins [55] and from this work). The filled markers connected by solid lines to empty markers show results postprocessed from classical MD simulations [9,55,59]; each marker represents the conductivity after applying the correction to account for the quantum Bose-Einstein statistics of vibrations discussed in Appendix A. Purple $\oplus$ markers, connected by dashed lines to solid circles, represent extrapolations of CDC conductivities from the simulated density (approximately 0.9 g/cm³) to the experimental density (approximately 0.7 g/cm³) (see main text for details). The unfilled markers $\times$, γ , $\triangle$, ∇ , and $+$ are experiments in carbide-derived carbon [15], irradiated glassy carbon [41], irradiated graphite [41,47], and amorphous carbon [34–38]. Shaded areas show convex hulls of experimental conductivities for different polymorph classes. Each conductivity prediction from this work is labeled with the structure name and the number of atoms in the simulation.

experiments in heavily irradiated glassy carbon (GC-30 [41], which has structural characteristics dominated by graphenelike sheets and, therefore, similar to NPC [112]). For IRG, we find that our density-conductivity predictions overlap with the range of experimental results from Refs. [41,47], and we confirm the inverse trend between irradiation dose and conductivity previously discussed by experiments and MD simulations by Farbos *et al.* [9]. In Supplemental Material [74], we provide details on (i) the negligible electronic contribution to the conductivity for the classes of carbon polymorphs analyzed here; (ii) the impact of irradiation characteristics (e.g., dose, temperature, type) on the conductivity of irradiated graphite; and (iii) how the experimental data shown in Fig. 1 were extracted from the corresponding references. Finally, we note that the analysis above has been limited to room temperature because this is sufficient for our goal of understanding the disorder-conductivity relation; the temperature dependence of the conductivity is discussed in detail in Supplemental Material [74].

IV. THERMAL CONDUCTIVITY AND BOND-NETWORK ENTROPY

To characterize the disorder-conductivity relation, we introduce a descriptor of disorder that quantifies heterogeneity in the atomic-bond network. The salient idea is to represent a solid as a collection of LAEs sampled from a certain probability distribution—for a perfect crystal, this distribution will be peaked only around environments contained in crystal’s primitive cell, while for a disordered solid the number of different LAEs will be higher and, therefore, their distribution broader. We start by defining a LAE around a reference atom by constructing a connection graph that starts from such reference atom and reaches its $n - 1$ closest neighbors, with n denoting the size of the LAE. We consider atoms as graph vertices, which are connected if they are within the carbon bonding distance (1.8 Å). We distinguish between different LAEs by looking at their ring structure; specifically, we employ the H_1 barcode descriptor [67] to characterize the algebraically independent rings in the LAE. For example, the LAE in

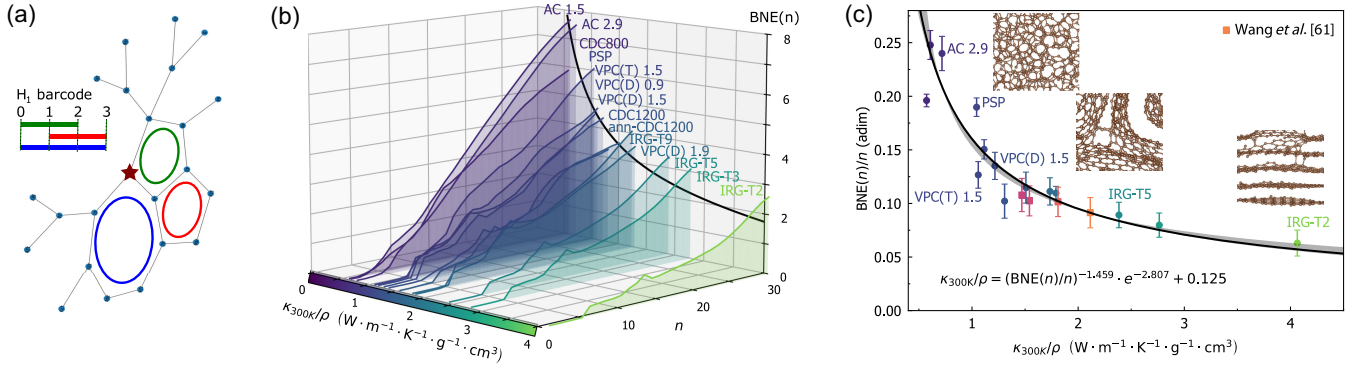


FIG. 2. Relation between bond-network entropy and thermal conductivity in disordered carbon. (a) H_1 barcode of an exemplary LAE around an atom (red star) in amorphous carbon with size $n = 23$. (b) Growth of the bond-network entropy with the size of LAEs ordered by their value of room-temperature conductivity divided by density. (c) Relation between average value of bond-network entropy divided by size of LAEs, $BNE(n)/n$, and room-temperature conductivity divided by density, $\kappa_{300\text{ K}}/\rho$. The vertical error bars show the standard deviation of $BNE(n)/n$ computed for $n \in [14, 30]$. The black line is the best shifted-power-law fit ($\kappa/\rho = [BNE(n)/n]^a \times e^b + c$, where a , b , and c are constants) of the relation between $BNE(n)/n$ and $\kappa_{300\text{ K}}/\rho$ [best-fit parameters are shown in (c)]. The shaded area shows changes in the best-fit power laws when the constant c is changed from 0 to $0.4 \text{ W} \cdot \text{m}^{-1} \cdot \text{K}^{-1} \cdot \text{g}^{-1} \cdot \text{cm}^3$. The square markers (Wang *et al.* [61]) are calculated using $\kappa_{300\text{ K}}$ from Ref. [61], and $BNE(n)/n$ computed from the structures used in that reference.

Fig. 2(a) contains three algebraically independent rings (green, red, and blue), which are classified by H_1 barcode in terms of their minimum and maximum edge distance from the reference-atom vertex (0–2 for green, 1–3 for red, and 0–3 for blue); see Appendix B for technical details. Changing the LAE's size n allows us to change the resolution with which we resolve structural disorder in a solid. For small values of n (e.g., $n \lesssim 4$), the LAE contains information only about connectivity between nearest neighbors and, therefore, describes short-range order (SRO). For $14 \lesssim n \lesssim 30$, the LAEs are large enough to describe structural features in the length scale range 5–20 Å, known as medium-range order (MRO) [113].

To describe disorder through the statistical distribution of LAEs, for each atom in our system we consider an n -sized LAE centered around it; we calculate its H_1 barcode (one for each atom in the system) and construct a distribution of H_1 barcodes:

$$P(H_1, n) = \frac{1}{N_{\text{at}}} \sum_b \delta_{H_1[\text{LAE}(b, n)], H_1}, \quad (4)$$

where N_{at} is the number of atoms in the atomistic model, $H_1[\text{LAE}(b, n)]$ is the H_1 barcode of the LAE having size n and centered at atom b , and $\delta_{H_1[\text{LAE}(b, n)], H_1}$ is an indicator function equal to one if $H_1[\text{LAE}(b, n)]$ is equal to the given barcode H_1 and zero otherwise. The H_1 barcode distribution (4) becomes broader as disorder in the topology of the atomic-bond network increases. Therefore, it is natural to quantify disorder through the information entropy of such a distribution, which we hereafter intuitively refer to as BNE:

$$BNE(n) = - \sum_{H_1} P(H_1, n) \ln P(H_1, n). \quad (5)$$

To understand the information provided by BNE, it is useful to evaluate it in two limiting cases: (i) a perfectly ordered, idealized crystal with one atom per primitive cell; (ii) a strongly disordered bulk glass. In the idealized crystalline case, $BNE(n) = 0 \forall n$, since the crystal order implies that each atom in the system has the same LAE for all n , and, therefore, $P(H_1, n)$ is a Kronecker delta. In contrast, in the second case, the presence of disorder implies that different LAEs are present, so the LAEs' distribution becomes broader and the corresponding information entropy larger.

We note that the choice of an appropriate descriptor to distinguish between LAEs is crucial for quantifying disorder in an accurate and numerically amenable way [67, 114, 115]. The choice of employing the H_1 barcode descriptor here is motivated by the recent work of Schweinhart, Rodney, and Mason [67], which showed that a classification based on the H_1 barcode resolves the problem of the proliferation of equivalence classes [67]. This problem arises when established methods such as graph isomorphism [116, 117] are used to distinguish LAEs, since with these methods nearly every environment is considered unique, making it necessary to sample a prohibitively large number of environments to estimate the probability distribution of LAEs and its information entropy. See also Supplemental Material [74] for a discussion of the advantages of describing LAEs with the H_1 barcode rather than with other descriptors.

Here, we focus on the scaling of $BNE(n)$ with respect to LAE size n , in particular, on the following key findings:

(i) BNE(n) is a size-dependent quantity that grows linearly with n across all carbon polymorphs considered; (ii) the growth rate BNE(n)/ n is a size-independent, intensive quantity determined by the degree of disorder in the topology of the atomic-bond network; (iii) BNE(n)/ n correlates with the macroscopic thermal conductivity and determines the smoothness of the vibrational density of states.

In Fig. 2(b), we show how BNE's growth rate distinguishes different classes of carbon polymorphs. In particular, BNE(n) displays the fastest growth with n in the phases having the lowest conductivity (e.g., AC and CDC800). As disorder decreases, the growth rate becomes lower: A medium growth rate is found for CDC1200, VPC(D) 1.5 or IRG T9, and the lowest growth rate emerges in weakly irradiated graphite (IRG T3 and T2). We note, in passing, that the atomistic models discussed here have a size much larger than the range of LAE's size n reported in Fig. 2; therefore, this analysis is not affected by periodic boundary conditions (see Fig. 8 in Appendix C for details). In Fig. 2(c), we show that BNE's average growth rate [computed averaging BNE(n)/ n for $n \in [14, 30]$] and room-temperature conductivity divided by density are inversely correlated (Spearman's rank-correlation coefficient equal to -0.947). For the materials analyzed here ($0.9 \lesssim \rho \lesssim 2.9$ g/cm³), the correlation is approximately a shifted power law ($\kappa/\rho = [\text{BNE}(n)/n]^a \times e^b + c$). As BNE(n)/ n decreases, the derivative of our fitting function decreases; i.e., fixed changes in BNE(n)/ n correlate with increasingly larger changes in the conductivity. Finally, to evaluate the robustness of our findings, we computed BNE(n)/ n for the atomistic models of nanoporous carbon released by Wang *et al.* [61], using the density and conductivity values reported in that reference to add additional points—independent from our study—to Fig. 2(c). The perfect overlap between these external data and the relation between BNE(n)/ n and κ/ρ established in this work further validates our results.

The correlation in Fig. 2(c) provides insights on the influence of atomic disorder on the conductivities in Fig. 1. Starting from a comparison between AC and VPC at density 1.5 g/cm³, we rationalize the lower conductivity of the former as originating from a higher degree of disorder in the bond network [i.e., higher BNE(n)/ n for $n \in [14, 30]$]. Analogous considerations hold for CDC and IRG, where samples with very similar density show conductivities that are very different and inversely related to BNE(n)/ n . Focusing on the conductivity variations observed within a certain class upon changing density, we see that, in AC, increasing density from 1.5 to 2.9 g/cm³ has negligible effect on BNE(n)/ n ; therefore, the conductivity differences between the various phases of AC can be explained mainly in terms of density—we see later that the higher the density, the higher the number of vibrational modes per unit volume, and in disordered systems this contributes to increasing the conductivity.

In contrast, in VPC increasing density implies also a decrease in disorder [BNE(n)/ n]; therefore, the conductivity increase observed in VPC upon increasing density is stronger than in AC.

Overall, these findings suggest that conductivity and density measurements can be used to quantify the structural heterogeneity of disordered solids and motivate us to investigate further the relationship between the structural-disorder descriptor BNE(n)/ n and both thermal and vibrational properties.

In Supplemental Material [74], we analyze the relations between the descriptor BNE(n)/ n we introduced and other properties, including density, heat capacity, and thermal conductivities at low and high temperature (100 and 700 K, respectively). We also investigate the relation between BNE(n)/ n and conductivity for polycrystalline graphene with different grain sizes [54].

V. ATOMIC VIBRATIONS AND BOND-NETWORK ENTROPY

In this section, we show that a relation between BNE(n)/ n and microscopic vibrational properties exists. We start by characterizing atomic vibrations with the VDOS:

$$g(\omega) = \frac{1}{\mathcal{V}N_c} \sum_{q,s} \delta(\omega - \omega_{qs}). \quad (6)$$

We also resolve how atoms with a certain coordination contribute to the VDOS by decomposing Eq. (6) into coordination-resolved partial VDOS [73,118] (C -PDOS):

$$g_{C_x}(\omega) = \frac{1}{\mathcal{V}N_c} \sum_{q,s} \delta(\omega - \omega_{qs}) \sum_{b,\alpha} |\mathcal{E}_{qs}^{ba}|^2 \delta_{b,C_x}, \quad (7)$$

where δ_{b,C_x} is an indicator function equal to one if atom b has coordination number equal to x and zero otherwise; $\mathcal{E}_{qs}^{ba}$ is the eigendisplacement that describes how atom b oscillates in direction α when the mode qs is excited [64,119]. Equation (7) is directly related to the VDOS via marginalization over the coordination variable: $\sum_{C_x} g_{C_x}(\omega) = g(\omega)$. Importantly, we can go beyond the SRO nearest-neighbor (coordination) analysis and look at how MRO properties influence the VDOS. To this aim, we further decompose the C -PDOS into contributions from different H_1 barcodes (C - H_1 -PDOS):

$$g_{C_x, H_1}(\omega) = \frac{1}{\mathcal{V}N_c} \sum_{q,s} \delta(\omega - \omega_{qs}) \sum_{b,\alpha} |\mathcal{E}_{qs}^{ba}|^2 \delta_{b,C_x} \delta_{H_1[\text{LAE}(b,n)], H_1}, \quad (8)$$

where we choose the size of the LAE n to be equal to 14 for the following reasons: (i) $n = 14$ is sufficiently large to

capture changes in the MRO features related to three graphitic sixfold rings, even in the presence of perturbations in the graphite interlayer distance, or changes in the number of atoms within one of the rings; (ii) the range $n \in [14, 30]$ is sufficient to capture signatures of MRO, and, since Fig. 2 shows that the BNE's growth with n is practically constant for $n \in [14, 30]$, we can choose the lowest value of n . Finally, it can be verified that marginalizing the $C-H_1$ -PDOS (8) with respect to the barcode variable yields the C -PDOS (7): $\sum_{H_1} g_{C,H_1}(\omega) = g_C(\omega)$.

In Fig. 3, we plot the VDOS and its decomposition into C -PDOS and $C-H_1$ -PDOS for all the classes of carbon polymorphs studied. Starting from AC, we see that increasing density yields an increase in magnitude of the VDOS, which trivially follows from the appearance of the system's cell volume V in the denominator of Eq. (6). Moreover, the changes in the shape of the VDOS with density are

determined by having different proportions of C_2 , C_3 , and C_4 coordination environments, and we highlight how the shape of the C -PDOS contributions are almost unchanged across all amorphous carbon structures (see Fig. SF6 in Supplemental Material [74] for details). In particular, at low density (1.5 g/cm³), the VDOS is bimodal with the low-frequency peak stronger than the high-frequency peak—the C -PDOS decomposition shows that these are determined by a superimposition of a monomodal low-frequency C_2 PDOS and a bimodal C_3 PDOS. Upon increasing density, the magnitude of the low-frequency monomodal C_2 contribution is progressively replaced by a high-frequency monomodal C_4 PDOS, resulting in a bimodal distribution with the high-frequency peak stronger than the low-frequency peak. In contrast, in less disordered polymorphs such as CDC, VPC, and IRG, SRO is dominated by the C_3 coordination environment;

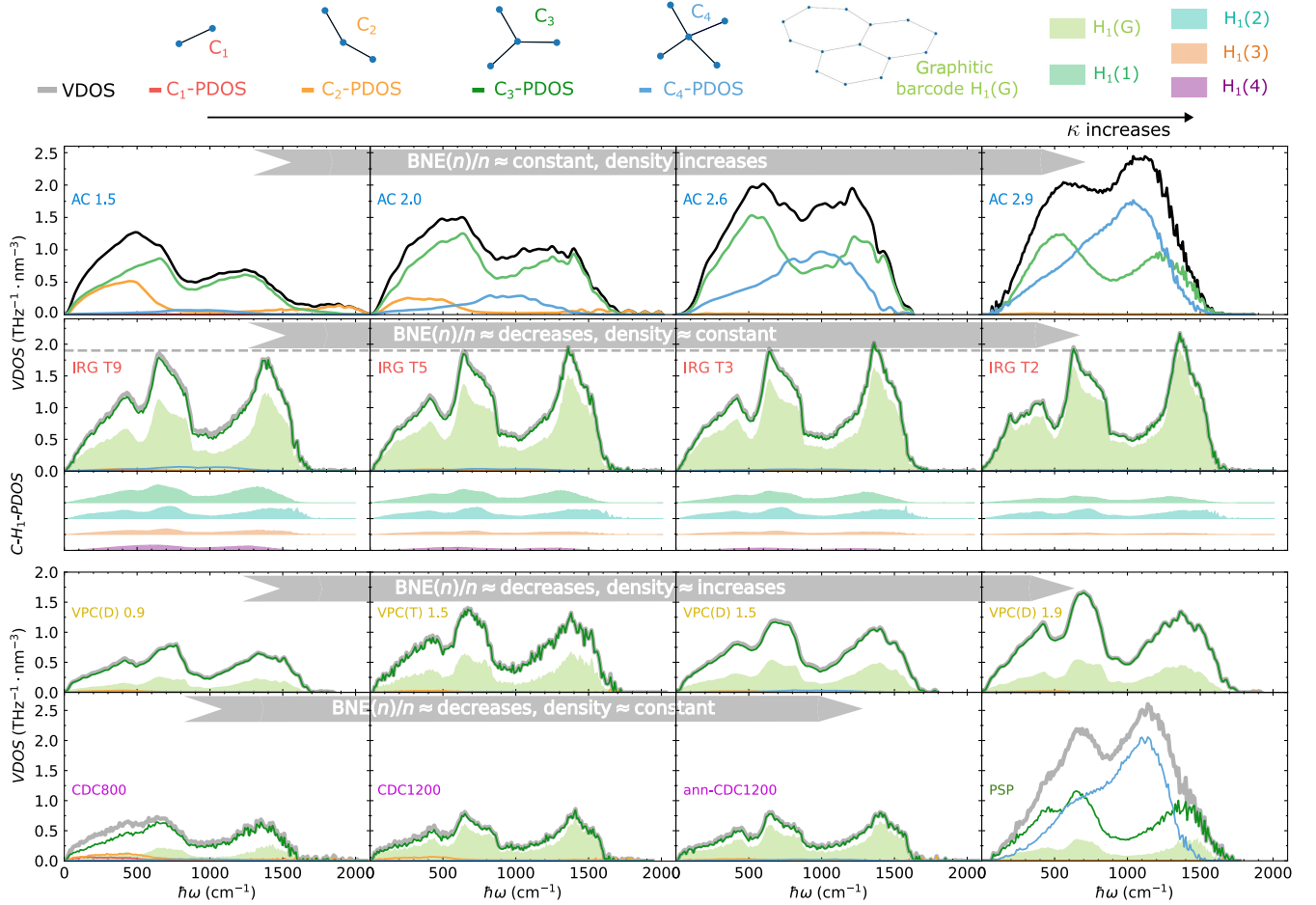


FIG. 3. Dependence of VDOS on short- and medium-range order. The panels are split by different groups of structures: the first row is AC, the second row is IRG, the third row is VPC, and the fourth row shows CDC and PSP. The total VDOS is thick gray, its decomposition into C -PDOS contributions from atoms having different coordination (SRO) is given by thin lines of different colors: red for dangling bonds (present appreciably only in CDC800, albeit practically invisible), while orange, green, and blue show the PDOS for two-, three-, and fourfold coordinated atoms, respectively. The shaded areas show the C_3-H_1 -PDOS: light green corresponds to the graphitic barcode, and the other four colors show other common barcodes in IRG. The dashed gray horizontal line in the IRG panel shows that the VDOS becomes smoother as irradiation-induced structural disorder increases.

hence, $\text{VDOS} \approx C_3\text{-PDOS}$ —this indicates that these solids are ordered over the SRO length scale, and, therefore, the visible VDOS changes must originate from disorder over a larger length scale. Therefore, in Fig. 3, we further decompose the $C_3\text{-PDOS}$ into contributions from different MRO using the $C_3\text{-}H_1\text{-PDOS}$. We show that the “graphitic $H_1(G)$ ” barcode—which describes MRO due to three hexagonal sixfold rings—allows us to resolve whether the C_3 SRO is associated with high density of graphitic rings (e.g., CDC1200 and IRG-T2) or not (e.g., CDC800). We note, in passing, that the $C_3\text{-}H_1(G)\text{-PDOS}$ has a triple-modal shape in all IRG structures, as well as in CDC1200 and in medium-high density VPC—specifically, we show in Supplemental Material Fig. SF7 [74] that, for a finite concentration of graphitic $H_1(G)$ environments, the shape of $C_3\text{-}H_1(G)\text{-PDOS}$ is practically independent from their concentration. More generally, the decomposition of IRG’s $C_3\text{-PDOS}$ into the five most frequent barcodes (shaded areas in Fig. 3) shows that increasing disorder yields an increase in the number of different barcodes at a fixed LAE size that contribute to the VDOS, consistently with $\text{BNE}(n)/n$ ’s increase shown in Fig. 2. Most importantly, we note that different barcode environments have $C_3\text{-}H_1\text{-PDOS}$ with different shapes; their superimposition leads to an overall smoothing of the VDOS and, hence, a decrease in the magnitude of its peaks. These findings generally apply to all carbon polymorphs, as we observe VDOS smoothing when we compare (i) CDC1200 and CDC800; (ii) VPC(D) 1.9, 1.5, and 0.9; and (iii) PSP and AC 2.9. In summary, increasing disorder in the atomic-bond network causes an increase in BNE’s growth rate and in the VDOS’ smoothness; in the next section, we show that the latter can be related to an effective model of a solid, where heat transport is limited by phonon scattering due to structural disorder, whose special limit is Kittel’s empirical interpretation of conductivity in glasses [120].

VI. EXPLICITLY DISORDERED GLASS VS PERTURBATIVELY DISORDERED CRYSTAL

In this section, we elucidate a relation between our WTE-based conductivity predictions and those obtained employing a model that generalizes Kittel’s phenomenological treatment of the thermal resistance induced by disorder [120,121]. We show how this relation allows us to connect $\text{BNE}(n)/n$, smoothness of the VDOS, and conductivity to the length scales of disorder and of the heat-transport mechanisms and, thus, fundamentally rationalize the correlation between $\text{BNE}(n)/n$ and thermal conductivity shown in Fig. 2.

We start by resolving the thermal conductivity with the usual frequency-dependent decomposition [65]

$$\kappa = \int_0^{\omega_{\max}} d\omega g(\omega)C(\omega)D(\omega), \quad (9)$$

where $\omega_{\max}$ is the maximum vibrational frequency of the solid, $g(\omega)$ is the VDOS (6), $C(\omega)$ is the specific heat of a vibration with frequency ω [Eq. (2)], and $D(\omega)$ is its diffusivity, describing the rate at which the heat carried by a vibration spreads [104]. The first description of heat transport in the presence of structural disorder was done relying on the semiclassical Peierls-Boltzmann transport equation (BTE) [122] and phenomenologically considering disorder-induced thermal resistance in a “perturbatively disordered crystal” (PDC). The PDC picture accounts for the frequency dependence of the diffusivity, interpreting it in terms of particlelike excitations having energy $\hbar\omega$ and propagating isotropically with velocities $v_{\text{eff}}(\omega)$ over disorder-limited transport length scales (mean free paths) $\lambda_{\text{eff}}(\omega)$:

$$D_{\text{PDC}}(\omega) = v_{\text{eff}}(\omega)\lambda_{\text{eff}}(\omega) = \frac{v_{\text{eff}}^2(\omega)}{\Gamma_{\text{dis}}(\omega)}, \quad (10)$$

where the last equality follows from the relation $v_{\text{eff}}(\omega)/\lambda_{\text{eff}}(\omega) = \Gamma_{\text{dis}}(\omega)$, with $\Gamma_{\text{dis}}(\omega)$ being the “disorder linewidth” or inverse scattering time $\Gamma_{\text{dis}}(\omega) = [\tau_{\text{dis}}(\omega)]^{-1}$ that describes the thermal resistance encountered by a vibration ω due to the presence of structural disorder.

A well-known special case of Eq. (10) is Kittel’s “phonon liquid” picture [120], which attempted to phenomenologically explain the conductivity of glasses by combining the BTE with Casimir’s model [121] for phonon-interface scattering around its physical lower bound. Specifically, this picture considers (i) the mean free path (MFP) $\lambda_{\text{eff}}(\omega)$ to be a temperature-dependent, but frequency-independent length scale $\lambda_{\text{eff}}(\omega) \rightarrow \Lambda_0$ determined by the type of disorder (e.g., for silica glass [120], Λ_0 is the ring size, approximately 7 Å) and (ii) the propagation velocity $v_{\text{eff}}(\omega)$ as frequency-independent average velocity of sound [$v_{\text{eff}}(\omega) \rightarrow v_{\text{sound}}$]. Kittel’s model is top-down interpretative but not bottom-up predictive; in fact, it allows one to estimate the value of the microscopic Λ_0 from the knowledge of measured values of κ (as well as of v_{sound} and C) but does not provide a rigorous prescription for determining the value of Λ_0 from first principles that can be used for the bottom-up prediction of κ . To address the limitations of Kittel’s phenomenological explanation, Allen and Feldman [104] introduced an alternative definition of diffusivity for harmonic glasses based on a Zener-like tunneling transport mechanism between quasidegenerate vibrational modes. In contrast to considering disorder as originating from perturbations of long-range crystalline order, AF describes heat transport in an explicitly disordered glass (EDG) that does not need a crystalline precursor or a relation to it. As already mentioned, the harmonic AF formalism emerges as a special case of the more general anharmonic WTE framework [64], and in the following we show that the latter allows one to shed light on the connection between atomic disorder,

VDOS smoothing, macroscopic κ , and Kittel's phonon liquid interpretation.

We start by showing that the WTE exposes a proportionality relation between the diffusivity of a generic (EDG or PDC) disordered system, its VDOS, and quasidegenerate velocity operator elements ($\mathbf{v}_{s,s'}$ with $\omega_s \simeq \omega_{s'}$). This is apparent when we consider Eq. (1) in the EDG limit, i.e., (i) we take the bulk-disordered limit (in practice using $\mathcal{V}$ containing thousands of atoms and, thus, considering $\mathbf{q} = \mathbf{0}$ only) and (ii) we consider linewidths slowly varying with frequency, larger than the average energy-level spacing, and far from the overdamped regime. This implies that the Voigt distribution reduces to a Lorentzian with FWHM determined by the sum of intrinsic linewidths $\Gamma_s + \Gamma_{s'}$ of vibrations with practically equal frequencies $\omega_s \sim \omega_{s'}$ and specific heats $C_s \sim C_{s'}$. Under these conditions, the WTE diffusivity [65] reduces to

$$D_{\text{EDG}}(\omega) = \frac{\pi}{\mathcal{V}g(\omega)} \sum_{s,s'} \frac{\|\mathbf{v}_{ss'}\|^2}{3} L[\omega_s - \omega_{s'}, \Gamma_s + \Gamma_{s'}] \times \delta(\omega - \omega_s) \approx \pi \Upsilon^2(\omega) \frac{1}{\rho_n} \left[\int d\omega' L[\omega - \omega', \Gamma(\omega')] g_d(\omega') \right], \quad (11)$$

where the simplification from the first to the second line is justified in strongly disordered systems [65,73] that feature velocity-operator elements negligibly dependent on the frequency difference between the modes s and s' [these are denoted by $\Upsilon^2(\omega_s) \approx N_{\text{at}} \|\mathbf{v}_{ss'}\|^2/3$; see Appendix D]. Moreover, in the second line, we have denoted the atom number density with $\rho_n = (N_{\text{at}}/\mathcal{V})$ and rewritten the mode linewidth Γ_s as a function of frequency $\Gamma(\omega_s)$ using the bijective mapping between frequency and mode arising from lack of symmetries (and, hence, lack of perfectly degenerate frequencies) in disordered systems [123]. Equation (11) shows that in an EDG the diffusivity is practically determined by a convolution between a Lorentzian $L[\omega - \omega', \Gamma(\omega)]$ and the “dressed” VDOS

$$g_d(\omega) = \frac{1}{\mathcal{V}} \sum_s L[\omega - \omega_s, \Gamma(\omega_s)] = \int d\omega' L[\omega - \omega', \Gamma(\omega')] g(\omega'), \quad (12)$$

whose name derives from its differences relative to the “bare” VDOS $g(\omega)$ defined in Eq. (6). Specifically, within the many-body Green's function formalism, the Dirac deltas $\delta(\omega - \omega_s)$ appearing in the bare VDOS (6) can be seen as resulting from the integration of bare noninteracting phonon spectral functions, while in the dressed VDOS (12) we have the integration of Lorentzian spectral functions

$b(\omega)_s = (1/\pi) \{ (\Gamma_s/2) / [(\omega - \omega_s)^2 + (\Gamma_s/2)^2] \}$, i.e., $g_d(\omega) = (1/\mathcal{V}) \sum_s b(\omega)_s$ [124,125], where the broadenings Γ_s are determined by interactions (due to, e.g., disorder or anharmonicity). We highlight that the second equivalence in Eq. (12) shows that the dressed VDOS is related to the bare VDOS $g(\omega)$ via a “dressing” integral, which practically is a convolution with a Lorentzian having frequency-dependent broadening (linewidth) and implies that the dressed VDOS becomes smoother as linewidths (proportional to the interaction strength) become larger [126].

In strongly disordered systems (EDG), the vibrational energy levels significantly repel each other [127], implying that the bare VDOS is already very smooth and practically indistinguishable from the dressed VDOS, $g_{\text{EDG}}(\omega) \approx g_d(\omega)$. This also implies that we can neglect the convolution between dressed VDOS and Lorentzian in the second line in Eq. (11), obtaining that in this (noninteracting, harmonic) disordered limit the diffusivity of an EDG is directly proportional to the bare VDOS:

$$D_{\text{EDG}}(\omega) \approx D_{\text{AF}}(\omega) = \pi \Upsilon^2(\omega) \frac{1}{\rho_n} g_{\text{EDG}}(\omega). \quad (13)$$

Equation (13) shows that the EDG diffusivity arises from a vibration mixing with a dense set of quasidegenerate vibrations, and the strength of this mixing is described by the square of the quasidegenerate velocity operator $\Upsilon^2(\omega)$. Finally, we note that Eq. (13) can be equivalently obtained from Eq. (11) by taking the ordered limit $\mathcal{V} \rightarrow \infty$ and $\Gamma_s \rightarrow \eta \rightarrow 0$ and coincides with the AF diffusivity [104].

VII. DISORDER-INDUCED VDOS SMOOTHING AND THERMAL RESISTANCE

In the BTE picture for conduction in a PDC, κ [Eqs. (9) and (10)] is influenced by structural disorder through a disorder-limited MFP determined phenomenologically. In contrast, in the rigorous WTE treatment of an EDG [Eqs. (9) and (11)], disorder impacts both VDOS and diffusivity. It is, therefore, natural to ask whether it is possible to obtain two compatible physical descriptions of heat transport using the rigorous WTE for an EDG and BTE treatment for a PDC. Here, we demonstrate that this is indeed possible, showing that PDC's phenomenological disorder linewidth can be formally derived from the WTE EDG through algebraic manipulations; then, we use these insights to formally determine disorder-limited MFP.

We start from the PDC picture, which prescribes specific heat and VDOS to be those of an unperturbed ordered crystal (UOC) and the influence of disorder to be accounted by the diffusivity term, which is formally unknown and usually determined by phenomenological arguments. Since the WTE in the EDG limit shows that in the presence of structural disorder the diffusivity is related to the VDOS [Eq. (11)], we hypothesize that the unknown PDC diffusivity also assumes such functional form, which we

highlight contains a convolution between the dressed VDOS and a Lorentzian distribution having FWHM that is unknown (and, in principle, frequency dependent). Importantly, through algebraic manipulations one can recast such convolution to apply on the UOC VDOS appearing at the very beginning of PDC conductivity expression. These manipulations imply that the disorder-mediated interaction that limits thermal transport within PDC model can be formally related to the smoothing of the dressed VDOS. In particular, the unknown disorder linewidth $\Gamma_{\text{dis}}(\omega)$ in Eq. (10) can be determined as the broadening that within the convolution [Eq. (12)] transforms the bare UOC VDOS into a dressed PDC VDOS equal to the bare EDG VDOS. In formulas,

$$\begin{aligned} \kappa_{\text{PDC}} &= \int d\omega g_{\text{UOC}}(\omega) C(\omega) D_{\text{unknown}}(\omega) \\ &= \int d\omega g_{\text{UOC}}(\omega) C(\omega) \pi \Upsilon^2(\omega) \frac{1}{\rho_n} \\ &\quad \times \left[\int d\omega' L[\omega - \omega', \Gamma_{\text{dis}}(\omega)] g_{\text{PDC}}(\omega') \right] \\ &\approx \underbrace{\int d\omega \underbrace{g_{\text{PDC}}(\omega)}_{\approx g_{\text{EDG}}(\omega)} C(\omega)}_{\approx D_{\text{EDG}}(\omega) \approx D_{\text{AF}}(\omega)} \underbrace{\left[\pi \Upsilon^2(\omega) \frac{1}{\rho_n} g_{\text{PDC}}(\omega) \right]}_{\approx D_{\text{EDG}}(\omega) \approx D_{\text{AF}}(\omega)}, \quad (14) \end{aligned}$$

where the last approximated equivalence holds under the assumptions used to obtain Eqs. (11) and (13); see Appendix E for details. From a mathematical viewpoint, Eq. (14) shows that the algebraic manipulations used to rewrite the PDC conductivity in WTE form are self-consistent; i.e., by rearranging the integration of the Lorentzian appearing in the WTE conductivity (1), one obtains an expression in which the dressed PDC VDOS appears twice, consistently with the form of the WTE EDG conductivity (13) (see Appendix E for details). In practice, $\Gamma_{\text{dis}}(\omega)$ is determined starting from the established [111,128] frequency-linewidth relations for phonon-disorder scattering:

$$\Gamma_{\text{dis}}(\omega) = \frac{v(\omega)}{L} + R\omega^2 g_{\text{UOC}}^{\text{DR}}(\omega). \quad (15)$$

The first term weakly depends on frequency (constant for $\omega \rightarrow 0$); it describes wave packets of atomic vibrations having velocity $v(\omega)$ and scattering with structural non-homogeneities of length scale L —e.g., grain boundaries [121], dislocations, or interstitial atoms [129] that have been observed in irradiated graphite [79]. The second term, instead, strongly depends on frequency and in IRG is, in practice, most relevant for $\hbar\omega \gtrsim 600 \text{ cm}^{-1}$; it accounts for pointlike defects [111] or mass impurities [80] having density proportional to the parameter R . Importantly, in such a term we also account for how the density reduction (DR) caused by irradiation disorder (porosity

increase) changes the VDOS: $g_{\text{UOC}}^{\text{DR}}(\omega) = (\rho_{\text{EDG}}/\rho_{\text{UOC}}) \times g_{\text{UOC}}(\omega/a)1/a$, where $a = \sqrt[3]{(\rho_{\text{EDG}}/\rho_{\text{UOC}})}$ accounts for the frequency shift due to density changes through an elementary Debye-like model [130] (see Appendix G for computational details); the factor $1/a$ ensures that the integral of the VDOS is preserved by the shift transformation; finally, the density ratio $\rho_{\text{EDG}}/\rho_{\text{UOC}}$ ensures the proportionality between density and VDOS magnitude [Eq. (6)].

Then, the two parameters (L and R) in the frequency-linewidth relation [Eq. (15)] are fitted to map, through the dressing (convolution) (12) and DR transformations, the bare UOC VDOS into a dressed PDC VDOS that overlaps with the bare EDG VDOS. Figure 4 demonstrates compatibility between the EDG VDOS (IRG T2) and the corresponding PDC VDOS (see Fig. 10 for analogous results in other structures). It is now important to summarize and highlight two insights. First, the equivalence between PDC and EDG VDOS shows that the disorder-induced repulsion between vibrational eigenstates, which yields a smooth bare VDOS in an EDG, can be interpreted in terms of PDC's disorder linewidth $\Gamma_{\text{dis}}(\omega)$ [appearing in Eq. (10)] that yields a smooth dressed VDOS in a PDC. Second, the relation (14) also implies that, when PDC and EDG conductivities are compatible, PDC and EDG diffusivities not only have the same functional form, but also assume the same numerical values.

These two insights allow us to connect and interpret the rigorous WTE predictions with Kittel's intuitive phonon liquid picture. In particular, Eq. (10) shows that the

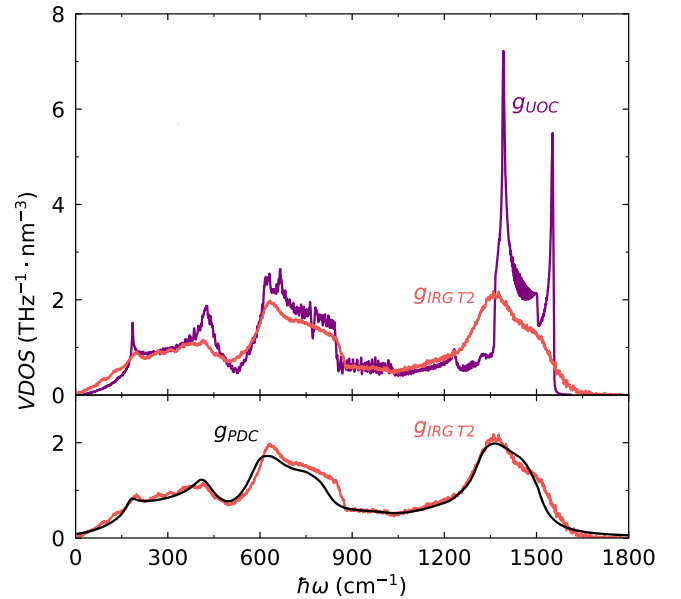


FIG. 4. Equivalence map between PDC and EDG. Top: VDOS of UOC graphite (purple) and of EDG irradiated graphite (IRG T2, red). Bottom: PDC VDOS $g_{\text{PDC}}(\omega)$ obtained perturbing the VDOS of UOC graphite (black) and VDOS of EDG IRG T2 (red).

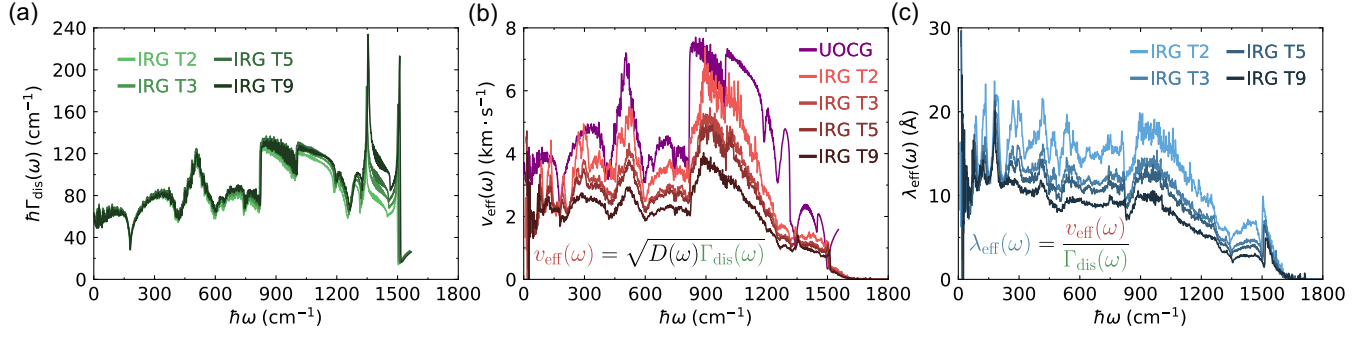


FIG. 5. Influence of structural disorder on (a) linewidths, (b) propagation velocity, and (c) vibrations' mean free paths. Starting from unperturbed ordered crystalline graphite (UOCG), structural disorder is progressively induced by irradiation in samples IRG T2, T3, T5, and T9 (see the text). Increasing disorder yields larger linewidths and lower velocities; consequently, the vibrational mean free paths decrease upon increasing disorder.

knowledge of the diffusivity and of the disorder linewidth $\Gamma_{\text{dis}}(\omega)$ fully determines the disorder-limited MFP:

$$\lambda_{\text{eff}}(\omega) = \sqrt{\frac{D_{\text{EDG}}(\omega)}{\Gamma_{\text{dis}}(\omega)}}, \quad (16)$$

as well as the propagation velocity $v_{\text{eff}}(\omega) = \sqrt{D_{\text{EDG}}(\omega)\Gamma_{\text{dis}}(\omega)}$. In Fig. 5, we show all these quantities, evaluated in IRG using the quantum-accurate GAP machine-learning potential [66] and also discussing how they are impacted by the amount of structural defects. We highlight that the disorder linewidth $\Gamma_{\text{dis}}(\omega)$ generally increases upon increasing structural disorder, with visible variations at high vibrational energy [the parameter $L \approx 20$ Å that practically controls the low-frequency behavior of Eq. (15) is negligibly affected by the increase in disorder from IRG T2 to IRG T9, while the parameter R that controls the high-frequency behavior increases from $R = 9.5 \times 10^{-6}$ in IRG T2 to $R = 17.4 \times 10^{-6}$ THz · cm · nm³ in IRG T9]. This increase of disorder linewidth upon irradiation is consistent with the increase in BNE's growth rate highlighted in Fig. 2, confirming that $\text{BNE}(n)/n$ is an informative descriptor for the defect density in IRG. We also note that the propagation velocity of vibrations in IRG structures follows similar trends with frequency as the average group velocity of UOC graphite and smoothly decreases upon increasing disorder—this quantitatively confirms the intuitive expectation that disorder induces repulsion between phonon bands [127] and, hence, reduces the average phonon group velocity. In Appendix F, we clarify the meaning of propagation velocity in disordered systems and also show how to obtain Kittel's picture [120] as a limiting case of the PDC framework. Finally, the MFP is frequency dependent and generally decreases upon increasing structural disorder.

Overall, these insights improve our understanding of transport beyond Kittel's picture for two reasons. First, they

explain how increasing structural disorder reduces the phonon propagation velocity, through strongly decreasing diffusivity and weakly increasing linewidth. This is an important difference from Kittel's model, in which the sound velocity was phenomenologically set relying on experimental values. Second, they extend Kittel's single temperature-dependent MFP picture by showing explicitly how MFPs depend on both frequency and degree of disorder; specifically, heat-transport length scales at room temperature are not necessarily limited to the upper-bound SRO length scale but can be continuously engineered in the MRO range through control of structural disorder.

VIII. CONCLUSIONS

We have rationalized the fundamental mechanisms governing the thermal-conductivity variations induced by atomistic disorder in a broad density range of structurally diverse disordered carbon polymorphs, solving the WTE [65] with quantum accuracy using a machine-learned interatomic potential [66]. Specifically, we have introduced the bond-network entropy growth rate $\text{BNE}(n)/n$ as a structural descriptor that quantifies heterogeneity in the topology of the atomic-bond network and demonstrated that it predicts variations in the conductivity of coordination-disordered solids. We have shown that $\text{BNE}(n)/n$ determines the smoothness of the VDOS and relied on all these findings to show how $\text{BNE}(n)/n$, VDOS smoothness, and conductivity are related to the length scales of structural disorder and to the thermal resistance they induce. These fundamental insights on the relationship between atomistic structure and macroscopic conductivity address the long-standing problem of formally determining the length scales underlying heat transport in disordered materials. In particular, we demonstrated the existence of a mapping between the WTE's bottom-up predictive theory for the conductivity of an EDG and a generalized version of Kittel's top-down phonon liquid interpretative model for the thermal conductivity of a PDC [120].

We note that the established approaches used to estimate heat-transport length scales in disordered solids—based on the DSF [69] and its vibrational extension [70] or velocity-current correlations [57]—all rely on an approximate (lossy) mapping of the vibrational properties of a glass into the Brillouin zone of a reference crystal, i.e., identification of a blurred band structure. This identification is often (albeit not always [131]) possible at low frequency but, in general, very challenging at high frequency in structurally disordered materials where disorder cannot be directly obtained from a reference crystalline structure [132], limiting the application of these approaches. The VDOS smoothing approach we introduced overcomes these limitations, as it does not require one to identify a reference periodic crystal structure and a Brillouin zone to approximately define bands—it requires one only to know the variations in the smoothness between two (observable) VDOS. Most importantly, the VDOS smoothing approach relies on a formal PDC-EDG mapping to determine from (physically observable) diffusivity and VDOS smoothness the length scales underlying heat transport in disordered solids.

Finally, we note that the established interpretation of transport in glasses—which classifies vibrations into propagons, diffusons, and locons [108]—is based on band blurring and phenomenologically relies on the property that the transition between propagon (inbandlike or propagationlike) transport and diffuson (interbandlike or tunnelinglike) transport is often centered around one single Ioffe-Regel frequency in the terahertz regime [70,108], which can be estimated from the DSF. Within this picture, transport length scales can be attributed exclusively to low-frequency propagon excitations. In contrast, within the VDOS smoothing picture transport length scales can be determined for all excitations in the vibrational spectrum, and we have found that some low-frequency vibrations can have transport length scales similar to those of some high-frequency vibrations.

Overall, we proposed an explanation of transport in disordered solids based on VDOS smoothing that is alternative to the established band-blurring picture and related propagon and diffuson classification. As such, this work calls for future studies to characterize conduction phenomena in solids in terms of heterogeneity of local atomic environments, VDOS smoothing and transport length scales, and the relation between them. From a technological viewpoint, this study shows that it is possible to extract information on atomistic structural properties and heat-transport length scales from VDOS smoothness [68] or conductivity measurements [133] and establishes the bond-network entropy as a fundamental degree of freedom to control and engineer the thermal properties of materials for energy-management applications [134–147]. In particular, the conductivity-BNE relation discussed here may be especially relevant for (i) quantifying atomic disorder in

supercapacitors, which strongly influences capacitance [5,148], and (ii) assessing structural degradation and reliability of ceramic plasma-facing materials exposed to neutron irradiation in nuclear-fusion reactors [149–153].

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G. C. has equity interest of Symmetric Group LLP that licenses force fields commercially and also in Ångström AI. The other authors declare that they have no competing interest.

DATA AVAILABILITY

The data that support the findings of this article are openly available [154,155]; the data consists of the atomistic models of disordered carbon polymorphs analyzed in this work.

APPENDIX A: QUANTUM CORRECTIONS ON CONDUCTIVITY FROM PREVIOUS MOLECULAR DYNAMICS SIMULATIONS

Several previous studies have computed the thermal conductivity of coordination-disordered carbon polymorphs using MD simulations [9,55,59], in which vibrational energy is distributed among vibrational modes according to classical equipartition [110]. To compare our quantum-accurate thermal conductivity predictions with these earlier classical results, we must account for differences in the energy distribution of the microscopic vibrational degrees of freedom. In particular, in quantum-accurate approaches such as the WTE, the atomic vibrational energy is distributed among microscopic vibrational degrees of freedom according to the Bose-Einstein statistics; therefore, the specific heat $C(\omega, T)$ depends on frequency and temperature [Eq. (2)]. In contrast, in MD simulations each vibrational mode has, regardless of its

frequency, a constant specific heat equal to the Boltzmann constant k_B [i.e., the infinite-temperature limit of Eq. (2)]. Consequently, classical MD simulations tend to overestimate the thermal conductivity at low temperature; to correct this and compare with our quantum calculations, we have adopted a phenomenological correction inspired by past work [156].

Specifically, the thermal conductivity analysis reported in Giri, Dionne, and Hopkins [55] allowed us to implement the following frequency-dependent corrections.

- (1) Starting from the frequency-dependent VDOS and AF diffusivity for densities 2.1 and 2.88 g/cm³ [see Figs. 3(a) and 4(a) in Ref. [55]], we transformed VDOS to have density-dependent integral equal to $\int_0^{\omega_{\max}} g(\omega) d\omega = [(3N_{at})/\mathcal{V}]$.
- (2) We computed the conductivity at room temperature using the equation $\kappa(300 \text{ K}) = \int_0^{\omega_{\max}} d\omega g(\omega) \times C(\omega, 300 \text{ K}) D(\omega)$ and its classical limit according to $\kappa_{\text{classical}} = \int_0^{\omega_{\max}} d\omega g(\omega) k_B D(\omega)$.
- (3) From those quantum (room-temperature) and classical values, we obtained a correction factor (CF) for each density, $\text{CF} = [\kappa(300 \text{ K})/\kappa_{\text{classical}}]$.
- (4) We multiplied the classical MD conductivity predictions [which used a modified Tersoff potential; see Fig. 4(b) in Ref. [55]] by the corresponding CF and reported them with filled markers in Fig. 1.

The works by Farbos *et al.* [9] and Suarez-Martinez and Marks [59] report the conductivities of IRG and VPC, respectively. Both these studies focus on the macroscopic conductivity and do not provide frequency-dependent mode diffusivity or VDOS. To implement a frequency-dependent correction as described above, we approximately

treated the structures in those references as having a VDOS equivalent to that of our corresponding (or most similar) structures. For Farbos *et al.* [9] the VDOS of IRG T2, and for Suarez-Martinez and Marks [59] we used VPC(D) 1.5 g/cm³. Then, we implemented a correction based only on specific heat.

- (1) We computed the specific heats according to $C(300 \text{ K}) = \int_0^{\omega_{\max}} d\omega g(\omega) C(\omega, 300 \text{ K})$ and $C_{\text{classical}} = \int_0^{\omega_{\max}} d\omega g(\omega) k_B$, obtaining a correction factor $\text{CF} = [C(300 \text{ K})/C_{\text{classical}}]$.
- (2) We multiplied the conductivities reported in Refs. [9,59] by CF and reported them with filled markers in Fig. 1.

In Fig. 6, we assess the accuracy of the two MD-conductivity correction protocols discussed above. Specifically, in structure of AC 1.5 with 216 atoms, we compare Green-Kubo (GK) equilibrium molecular dynamics conductivity calculations corrected either by the AF conductivity (κ_{AF}) or specific heat (C) correction factors with direct rWTE calculations. For details of the simulation, see Supplemental Material [74]. We find that direct GK predictions at 100 and 300 K are in satisfactory agreement with rWTE and AF conductivity predictions in the classical limit ($T = 2000 \text{ K}$). After corrections, in AC 1.5 the κ_{AF} -corrected GK conductivity slightly overestimates the rWTE predictions and C -corrected GK conductivity slightly underestimates it. At room temperature, the GK-corrected predictions are within 20% of the rWTE value; at 100 K, the κ_{AF} -corrected GK conductivity is within the error bars of rWTE result, and the upper error bar of C -corrected value is within 35% of the rWTE result. A discussion on the agreement between quantum-corrected

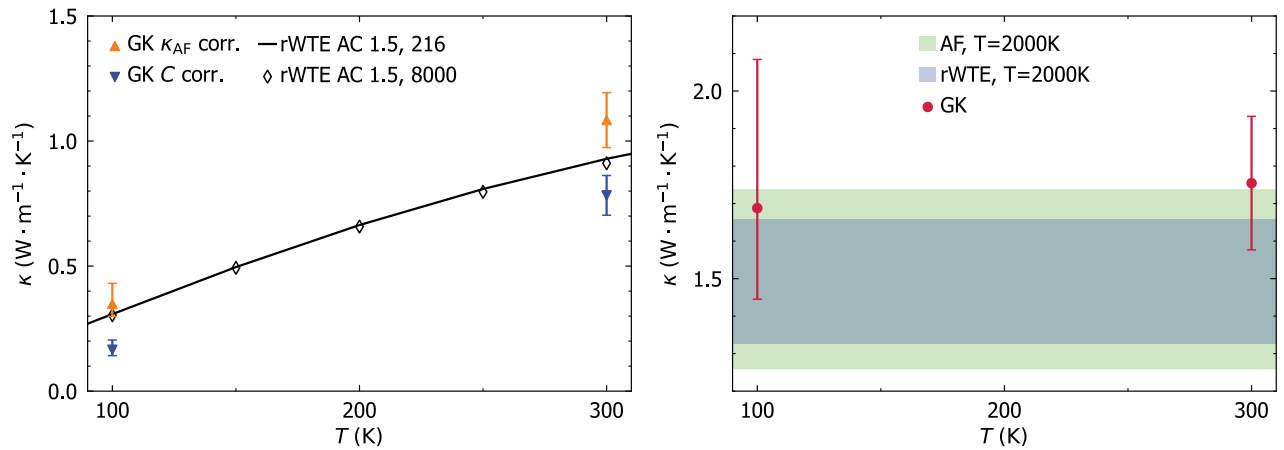


FIG. 6. Comparison between GK and rWTE predictions for amorphous carbon at density 1.5 g/cm³. Left: comparison between rWTE conductivity predictions for AC 1.5 models containing 216 and 8000 atoms (solid line and diamond scatter points, respectively) and GK conductivity for AC 1.5 with 216 atoms with the quantum correction schemes discussed in the text, derived either from Allen-Feldman conductivity (κ_{AF} , orange triangles) or from specific heat (C , blue triangles). Right: comparison in AC 1.5 with 216 atoms between the trace of uncorrected GK conductivity and AF and rWTE predictions at 2000 K. The spread of AF and rWTE predictions denotes the variations in the eigenvalues of the conductivity tensors. The GK error bars in the right panel are derived from both directional uncertainty and from standard deviation of GK conductivity in the last 250 ps of the simulation, and they have been scaled down by the corresponding correction factors in the left panel.

MD methods and rWTE predictions is also reported in Ref. [157], and other correction schemes are discussed in Refs. [57,61].

APPENDIX B: DETERMINATION OF THE H_1 BARCODE

Here, we summarize the algorithm discussed in Ref. [67] to compute the H_1 barcode of the atoms-bonds graph obtained from a certain LAE. We denote with r the root atom (center) of the LAE. Given non-negative integers $c \leq d$, we define the (c, d) -shell annulus $S(c, d)$, which is a subgraph of the LAE composed of all atoms (and bonds connecting them) with edge distance (number of bonds on shortest path) between c and d from r . To characterize the bond network of the subgraph, it is useful to determine the number of algebraically independent rings in $S(c, d)$, given by

$$F[(c, d)] = \text{number of components}[S(c, d)] - \text{number of atoms}[S(c, d)] + \text{number of bonds}[S(c, d)]. \quad (\text{B1})$$

The number of algebraically independent rings is related to the H_1 barcode as [67]

$$F[(c, d)] = \text{rank}\{H_1[S(c, d)]\}; \quad (\text{B2})$$

i.e., it is the size of the set of rings $H_1[S(c, d)]$ that allows one to express all other rings by linear combinations. $F[(c, d)]$ can also be resolved in terms of the number of intervals of the form (a, b) [e.g., shown as bars in Fig. 2(a)] in the H_1 barcode, denoted with $G[(a, b)]$:

$$F[(c, d)] = \sum_{(a,b) \leq (c,d)} G[(a, b)], \quad (\text{B3})$$

where the sum is taken over annuli (a, b) contained within or equal to the (c, d) annulus. As discussed in Ref. [67], the form of Eq. (B3) implies that $G[(c, d)]$ can be obtained from $F[(a, b)]$ using Möbius inversion:

$$G[(c, d)] = \sum_{(a,b) \leq (c,d)} F[(a, b)] \mu[(a, b), (c, d)], \quad (\text{B4})$$

where $\mu[(a, b), (c, d)]$ is a Möbius function defined as

$$\mu[(a, b), (a, b)] = 1, \quad (\text{B5})$$

$$\mu[(a, b), (c, d)] = - \sum_{(a,b) \leq (e,f) < (c,d)} \mu[(a, b), (e, f)], \quad (\text{B6})$$

which can be solved recursively. We note that the sum in Eq. (B6) is computed over $(a, b) \leq (e, f) < (c, d)$ [excluding the case where $(e, f) = (c, d)$, as implemented in the

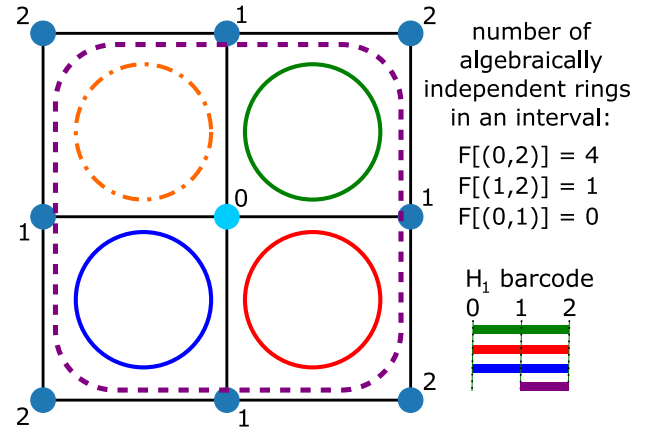


FIG. 7. Algebraically independent vs primitive rings. Example of a LAE formed by blue atoms and black bonds and having root atom cyan. The primitive rings [159] are solid green, red, blue, and dash-dotted orange. In contrast, the algebraically independent rings in the LAE are solid green, red, blue, and dashed purple; each of these corresponds to a bar in the H_1 barcode. The matrix $F[(c, d)]$ (see the text) contains information about the number of algebraically independent rings in the (c, d) -shell annulus $S(c, d)$ of the LAE, where c and d are edge distances of atoms from the root atom, denoted with numbers in the figure. Specifically, the number of algebraically independent rings in the annulus that contains the entire LAE is equal to four ($F[(0, 2)] = 4$, the four primitive rings). The number of rings in the annulus that excludes the root atom is equal to one (only purple dashed ring, $F[(1, 2)] = 1$). There are no rings in the annuli containing atoms with edge distance equal to only 0, 1, or 2 or either 0 or 1 ($F[(0, 0)] = 0$, $F[(1, 1)] = 0$, $F[(2, 2)] = 0$, $F[(0, 1)] = 0$). The H_1 barcode reproduces the number of algebraically independent rings according to Eq. (B3). The dash-dotted orange ring is not contained in the H_1 barcode, since it is not algebraically independent from the solid green, red, blue, and dashed purple rings.

SWATCHES software [158], correcting a typo present in the original paper [67].

We also note that, in order to characterize and compare disorder across structures with different density, in this work we define LAEs in terms of number of atoms; this differs from the convention adopted by Schweinhart, Rodney, and Mason [67], who defined LAEs as a function of number of layers (atoms with the same edge distance to the root). Finally, we note that algebraically independent rings obtained from the intervals $G[(c, d)]$ are not necessarily primitive rings (i.e., rings that cannot be decomposed into smaller rings; see Ref. [159] for details) present in the LAE; see Fig. 7 for an example.

APPENDIX C: BNE AND FINITE-SIZE EFFECTS

In Fig. 8, we show the behavior of BNE as a function of LAE's size n for three models of amorphous carbon that are expected to represent the same physical system, as they all have density of approximately 2.9 g/cm³ and they differ

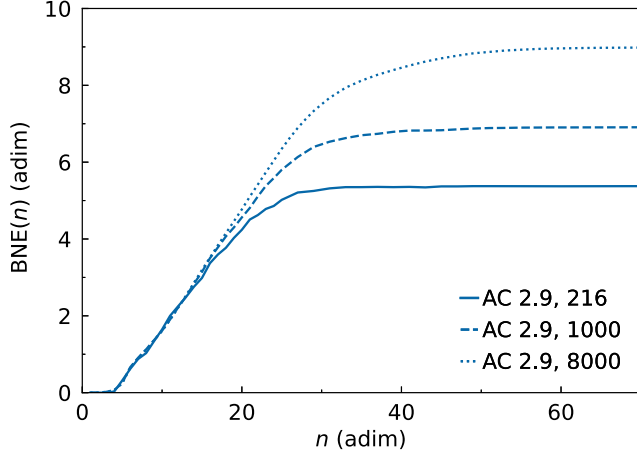


FIG. 8. BNE and finite-size effects. Solid, dashed, and dotted lines correspond to 216-, 1000-, and 8000-atom models of AC with density of approximately 2.9 g/cm³, respectively. n is the number of atoms in the local environments. We note that, for $5 \lesssim n \lesssim 15$, BNE's growth rate is approximately constant and indistinguishable between the three physically equivalent representations of AC considered. We highlight how the finite-size saturation of BNE(n) occurs for larger value of n in larger models. This shows that BNE's growth rate can be used to characterize structural disorder in AC over length scales smaller than the model's size.

only by simulation-cell size. We see that BNE grows with the LAE's size n , as expected from the extensivity property of the entropy and then saturates to a constant value $\text{BNE}_{\text{sat}} = \ln(N_{\text{at}})$ determined by the number of atoms in the simulation cell (N_{at}). From a mathematical viewpoint, the saturation occurs when the H_1 barcode distinguishes all atomic environments, and, therefore, the distribution of barcodes approaches a uniform distribution with value $1/N_{\text{at}}$ for each atom. Most importantly, before the finite-size effect saturation occurs, BNE's growth rate is practically indistinguishable between the three models of AC, suggesting that it can be used to characterize structural disorder in AC over length scales smaller than the model's size. This motivates using BNE's growth rate as a descriptor for disorder in the bond network of solids.

We additionally note that the saturation is the reason we limit range of n in Fig. 2 to $n \leq 30$, as higher n require exponentially bigger models. For example, in amorphous carbon to obtain converged results for $n = 30$ compared with $n = 15$, one requires a model which is approximately 40 times bigger (8000 vs 216 atoms). Hence, the estimated model size to obtain converged results for $n = 45$ in amorphous carbon is around 320 000 atoms. As we

mentioned in Sec. IV, the range of $n \in [14, 30]$ is large enough to capture medium-range order features, and all structures in Fig. 2 have disorder in the short to medium range. The region of $n > 30$ might be a subject of future research analyzing very weakly disordered solids, where conductivity is both significantly affected by structural order as well as intrinsic anharmonicity, going beyond the regime analyzed in this paper.

APPENDIX D: QUASIDEGENERATE VELOCITY OPERATOR

The quasidegenerate velocity operator in the frequency representation $\Upsilon(\omega)$ appearing in Eq. (13) is defined as

$$\Upsilon^2(\omega) = [\mathcal{G}(\omega)]^{-1} \frac{1}{\mathcal{V}} \sum_{s,s'} \frac{\|\mathbf{v}_{ss'}\|^2}{3} \delta\left(\omega - \frac{\omega_s + \omega_{s'}}{2}\right) \times \delta(\omega_s - \omega_{s'}), \quad (\text{D1})$$

where $\mathcal{G}(\omega)$ is a density of states defined as

$$\mathcal{G}(\omega) = \frac{1}{N_{\text{at}}} \frac{1}{\mathcal{V}} \sum_{s,s'} \delta\left(\omega - \frac{\omega_s + \omega_{s'}}{2}\right) \delta(\omega_s - \omega_{s'}). \quad (\text{D2})$$

APPENDIX E: DERIVATION OF THE WTE DIFFUSIVITY IN THE EXPLICITLY DISORDERED GLASS LIMIT

We start from the first line in Eq. (11), and we use Eq. (D1) to approximate the velocity operator elements $\|\mathbf{v}_{ss'}\|^2/3$ by a single-frequency-dependent function $[\Upsilon^2(\omega)]/N_{\text{at}}$:

$$D_{\text{EDG}}(\omega) = \frac{\pi}{\mathcal{V}g(\omega)} \sum_{s,s'} \frac{\|\mathbf{v}_{ss'}\|^2}{3} L[\omega_s - \omega_{s'}, \Gamma_s + \Gamma_{s'}] \delta(\omega - \omega_s) \approx \frac{\pi}{g(\omega)} \frac{\Upsilon^2(\omega)}{N_{\text{at}}} \left[\frac{1}{\mathcal{V}} \sum_{s,s'} L[\omega_s - \omega_{s'}, \Gamma_s + \Gamma_{s'}] \delta(\omega - \omega_s) \right]. \quad (\text{E1})$$

Then, we rewrite the quantity in the square brackets in terms of the convolution:

$$\begin{aligned}
\left[\frac{1}{\mathcal{V}} \sum_{s,s'} L[\omega_s - \omega_{s'}, \Gamma_s + \Gamma_{s'}] \delta(\omega - \omega_s) \right] &= \frac{1}{\mathcal{V}} \sum_{s,s'} \int d(\omega' - \omega_{s'}) L[(\omega_s - \omega_{s'}) - (\omega' - \omega_{s'}), \Gamma_s] \\
&\quad \times L[\omega' - \omega_{s'}, \Gamma_{s'}] \delta(\omega - \omega_s) \\
&= \frac{1}{\mathcal{V}} \sum_{s,s'} \int d\omega' L[\omega_s - \omega', \Gamma_s] L[\omega' - \omega_{s'}, \Gamma_{s'}] \delta(\omega - \omega_s) \\
&= \sum_s \delta(\omega - \omega_s) \int d\omega' L[\omega - \omega', \Gamma(\omega)] \frac{1}{\mathcal{V}} \sum_{s'} L[\omega' - \omega_{s'}, \Gamma_{s'}] \\
&= \mathcal{V}g(\omega) \int d\omega' L[\omega - \omega', \Gamma(\omega)] g_d(\omega') = \mathcal{V}g(\omega) g_{dd}(\omega), \tag{E2}
\end{aligned}$$

where $g_{dd}(\omega)$ denotes the convolution of the dressed VDOS with the Lorentzian with FWHM $\Gamma(\omega)$. In Eq. (E2), to go from the first to the second line, we used the property that a Lorentzian with FWHM $\Gamma_s + \Gamma_{s'}$ can be obtained by convolving two Lorentzians with FWHM Γ_s and $\Gamma_{s'}$, respectively. To transform the second line into the third, we changed the variable of integration from $\omega' - \omega_{s'}$ to ω' . From third to fourth line, we exploited the delta function $\delta(\omega - \omega_s)$ to replace ω_s and Γ_s in the first Lorentzian to ω and $\Gamma(\omega)$. $\Gamma(\omega)$ denotes the linewidth evaluated at frequency ω ; this notation is possible here because the presence of structural disorder forbids degeneracies and, thus, allows a bijective mapping between frequency and mode, i.e., $\Gamma_s = \Gamma(\omega_s) = \Gamma(\omega = \omega_s)$. In the fourth line, we also regrouped the terms, and from the fourth to the fifth line

we used the definitions of VDOS $\mathcal{V}g(\omega) = \sum_s \delta(\omega - \omega_s)$ and dressed VDOS $g_d(\omega')$ [Eq. (12)]. By combining Eqs. (E1) and (E2), we obtain the second line in Eq. (11).

If we neglect the influence of the intrinsic linewidth on the broadening of $g_{dd}(\omega)$, then we can approximate the disordered solid to be harmonic, leading to the alternative expression for AF diffusivity shown by Eq. (13). In Fig. 9, we show the numerical equivalence between Eq. (13) and the AF diffusivity in various IRG models.

Finally, we discuss how to obtain the self-consistent relation discussed in Eq. (14). We start considering the frequency decomposition equation [Eq. (9)], plug in the expression (11) for the EDG diffusivity $D(\omega)$, and perform the following algebraic manipulations:

$$\begin{aligned}
\kappa &= \int d\omega g(\omega) C(\omega) D(\omega) \\
&= \int d\omega g(\omega) C(\omega) \pi \Upsilon^2(\omega) \frac{1}{\rho_n} \left[\int d\omega' L[\omega - \omega', \Gamma(\omega)] g_d(\omega') \right] \\
&= \pi \frac{1}{\rho_n} \iint d\omega d\omega' C(\omega) \Upsilon^2(\omega) g(\omega) L[\omega - \omega', \Gamma(\omega)] g_d(\omega') \\
&\approx \pi \frac{1}{\rho_n} \int d\omega' g_d(\omega') C(\omega') \Upsilon^2(\omega') \int d\omega g(\omega) L[\omega - \omega', \Gamma(\omega)] \\
&= \pi \frac{1}{\rho_n} \int d\omega' g_d(\omega') C(\omega') \Upsilon^2(\omega') g_d(\omega') \\
&= \int d\omega g_d(\omega) C(\omega) \pi \Upsilon^2(\omega) \frac{1}{\rho_n} g_d(\omega) = \int d\omega g_d(\omega) C(\omega) D_d(\omega), \tag{E3}
\end{aligned}$$

where we denote the dressed diffusivity $D_d(\omega) = \pi \Upsilon^2(\omega) (1/\rho_n) g_d(\omega)$ [i.e., having the same form as Eq. (13) but with bare VDOS replaced by the dressed VDOS]. In the third line we reordered the integrals, and in the fourth line we used the fact that in derivation of Eq. (11) we used an approximation that the interacting modes have similar frequency due to value of the linewidth $\Gamma(\omega)$ being much smaller than the value of the frequency ω . We used this approximation again here to set $C(\omega) \sim C(\omega')$ and

$\Upsilon(\omega) \sim \Upsilon(\omega')$ for interacting modes. In the fifth line we identify $\int d\omega g(\omega) L[\omega - \omega', \Gamma(\omega)]$ with the dressed VDOS $g_d(\omega')$. In the sixth line we renamed ω' to ω . The expression in the final line in Eq. (E3) is particularly insightful, since the VDOS in the first term and VDOS in the diffusivity are dressed, preserving the property that the diffusivity in disordered systems is proportional to the VDOS. We conclude by noting that the relation between the PDC and EDG conductivities is summarized in Table I.

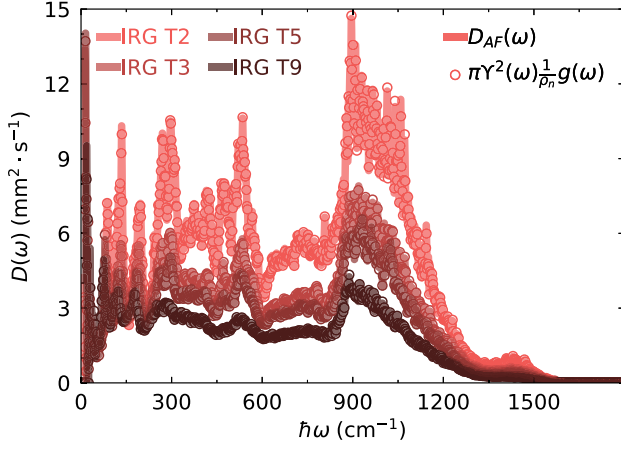


FIG. 9. Equivalence between EDG and AF diffusivity in IRG. Solid lines are AF diffusivities in various IRG samples, and the overlapping empty circles are the corresponding WTE diffusivities in the EDG limit [Eq. (13)].

APPENDIX F: RELATION BETWEEN PDC AND KITTEL'S INTERPRETATION OF CONDUCTIVITY IN GLASSES

We find that the PDC-EDG mapping is a generalization of Kittel's interpretation, since the conductivity of a PDC reduces to the Kittel's expression $\kappa = \frac{1}{4} C_V v_{\text{sound}} \Lambda_0$ [120] by averaging propagation velocity and transport length scale over frequency:

$$\begin{aligned}
 \kappa_{\text{PDC}} &= \int d\omega g_d(\omega) C(\omega) v_{\text{eff}}(\omega) \lambda_{\text{eff}}(\omega) \\
 &= \left[\int d\omega g_d(\omega) C(\omega) \right] \frac{\int d\omega g_d(\omega) C(\omega) v_{\text{eff}}(\omega) \lambda_{\text{eff}}(\omega)}{\int d\omega g_d(\omega) C(\omega)} \\
 &= C_V v_{\text{eff}} \lambda_{\text{eff}} \\
 &\rightarrow \frac{1}{4} C_V v_{\text{sound}} \Lambda \rightarrow \frac{1}{4} C_V v_{\text{sound}} \Lambda_0,
 \end{aligned} \tag{F1}$$

where in the second and third lines $v_{\text{eff}}(\omega)$ and $\lambda_{\text{eff}}(\omega)$ are averaged and become frequency independent; in the fourth line the propagation velocity is changed to velocity of sound and a factor of 4 is absorbed into the product $v_{\text{sound}} \Lambda$ to account for the phenomenological factor $\frac{1}{4}$ in the original Kittel paper [120]. The final arrow represents the limit of heavily disordered glasses at noncryogenic temperatures [120], where Λ is assumed to be independent of temperature and, hence, is redefined as Λ_0 .

We note that increase of Λ with decrease in temperature for a quartz glass observed by Ref. [120] naturally follows from Eq. (F1), since diffusivity can depend on temperature and is averaged over frequency with temperature-dependent weights:

$$\Lambda(T) = \frac{4}{v_{\text{sound}}} \frac{\int d\omega g_d(\omega) C(\omega, T) D(\omega, T)}{\int d\omega g_d(\omega) C(\omega, T)}. \tag{F2}$$

Here, we also note that the name “propagation velocity” for $v_{\text{eff}}(\omega)$ was used to point out the relationship to the BTE, as we use a BTE-like expression for the diffusivity $D(\omega) = v_{\text{eff}}(\omega) \lambda_{\text{eff}}(\omega)$, and propagation is the transport mechanism described by the BTE [64]. Because of the frequency dependence, this quantity generalizes the frequency-independent Debye-like sound velocity v_{sound} used in Kittel's interpretation of conductivity in glasses. We note that defining a frequency-dependent velocity does not require resolving phonon wave vectors or band structures and is effectively determined here from the off-diagonal elements of the velocity operator that assume nonzero values in glasses; interestingly, we note that resolving the velocity in frequency allows one to connect it a vibrational mode in strongly disordered systems where disorder forbids perfect degeneracies and, therefore, one has bijective mapping between frequency and mode. In summary, the meaning of frequency-dependent effective propagation velocity for disordered systems is different from either the sound velocity or from group velocity. The propagation velocity

TABLE I. Comparison between the WTE treatments of PDC and EDG.

	PDC	EDG
Transport mechanism	Particlelike propagation	Wavelike tunneling
VDOS	Not broadened $g(\omega) = g_{\text{UOC}}^{\text{DR}}(\omega)$	Broadened by structural disorder $g(\omega) = g_{\text{EDG}}(\omega)$
Dressed VDOS	Broadened by structural disorder $g_{\text{PDC}}(\omega) = \int d\omega' L[\omega - \omega', \Gamma_{\text{dis}}(\omega')] g_{\text{UOC}}^{\text{DR}}(\omega')$ $g_d(\omega) \equiv g_{\text{PDC}}(\omega) \approx g_{\text{EDG}}(\omega)$	Broadened by disorder and anharmonicity $g_d(\omega) = \int d\omega' L[\omega - \omega', \Gamma(\omega')] g(\omega')$ $g_d(\omega) \approx g(\omega) = g_{\text{EDG}}(\omega)$
Origin of linewidth	Structural disorder	Intrinsic, e.g., due to isotopes or anharmonicity
Diffusivity	Eq. (10), $D_{\text{PDC}}(\omega) = v_{\text{eff}}(\omega) \lambda_{\text{eff}}(\omega) = \frac{v_{\text{eff}}^2(\omega)}{\Gamma_{\text{dis}}(\omega)}$, and Eq. (14), $D_{\text{PDC}}(\omega) = \pi \Upsilon^2(\omega) \frac{1}{\rho_n} g_{\text{PDC}}(\omega)$	Eqs. (11) and (13), $D_{\text{EDG}}(\omega) \approx \pi \Upsilon^2(\omega) \frac{1}{\rho_n} [\int d\omega' L[\omega - \omega', \Gamma(\omega')] g_d(\omega')] \approx \pi \Upsilon^2(\omega) \frac{1}{\rho_n} g_{\text{EDG}}(\omega)$
Compatible observables	Thermal conductivity κ , dressed VDOS $g_d(\omega)$, and diffusivity $D(\omega)$	

$v_{\text{eff}}(\omega)$ and transport length scale $\lambda_{\text{eff}}(\omega)$ are defined based on physical observables of frequency-dependent VDOS and diffusivity. Their product is equal to the diffusivity $D(\omega) = v_{\text{eff}}(\omega)\lambda_{\text{eff}}(\omega)$, and their quotient is the disorder linewidth $\Gamma_{\text{dis}}(\omega) = v_{\text{eff}}(\omega)/\lambda_{\text{eff}}(\omega)$ that determines the smoothness of the dressed PDC VDOS $g_d(\omega) = \int d\omega' L[\omega - \omega', \Gamma_{\text{dis}}(\omega')]g(\omega')$ equal to the VDOS of a disordered solid.

Through PDC-EDG mapping, we can represent the diffusivity of a glass (EDG) in terms of propagation velocities and transport length scales of an effective crystal (PDC), in which vibrations interact in a way determined by the disorder within the EDG. The existence of a formal mapping between PDC and EDG does not imply that the vibrational modes in the actual disordered system (EDG) propagate as in a PDC—this is evident from the fact that the effective velocities and transport length scales emerging from this mapping result from combining several quasidegenerate off-diagonal elements of the EDG's velocity operator.

The usefulness of the EDG-PDC mapping lies in its ability to determine the microscopic physical length scale underlying heat transport mediated by vibrations of frequency ω (albeit without resolving whether they contribute through propagation or tunneling mechanisms). Because the effective frequency-dependent velocities and transport length scales are derived from observables such as the VDOS and diffusivity, they have a physical meaning. Their determination is made possible by the PDC-EDG mapping, and we have shown that one can obtain fundamental insights into structure-conductivity relations by comparing the transport length scales with material-structure length scales (e.g., short- and medium-range order).

APPENDIX G: DETERMINATION OF THE DISORDER LINEWIDTH FROM PDC VDOS

In this appendix, we discuss the numerical details on how to determine the PDC disorder linewidths $\Gamma(\omega')$ that have to be used in Eq. (12) to transform the VDOS of pristine UOC graphite into the VDOS of IRG.

To obtain the density-renormalized VDOS discussed in Sec. VII, $g_{\text{UOC}}^{\text{DR}}(\omega)$, we proceeded as follows. First, we used the GAP [66] to compute second-order interatomic forces in a $8 \times 8 \times 2$ supercell of graphite and then used these and Fourier interpolation [160] to compute vibrational frequencies and group velocities on a dense $128 \times 128 \times 32$ $\mathbf{q}$ mesh. By multiplying the frequencies by the factor a , as discussed in Sec. VII, we take into account the effect that the density reduction induced by irradiation has on the vibrational frequencies. The bare VDOS was computed using a Lorentzian broadening for the delta functions with FWHM equal to $2 \times 0.6 \text{ cm}^{-1}$, where 0.6 cm^{-1} is the value of the convergence plateau parameter η used for thermal conductivity calculations in IRG (see Supplemental Material [74] for details).

After determining the bare $g_{\text{UOC}}^{\text{DR}}(\omega)$, we apply to it the dressing transformation (12) to obtain the dressed VDOS of the PDC. When performing such transformation, we assume that the disorder linewidths appearing in the dressing integral assume the established functional form (15); specifically, we calculated the modulus of the group velocity in the frequency representation, $v(\omega)$, which appears in such an expression as

$$v(\omega) = \frac{\sum_{\mathbf{q}s} \sqrt{\frac{v_{\mathbf{q}s}^2}{3}} \delta(\omega - a\omega_{\mathbf{q}s})}{\sum_{\mathbf{q}s} \delta(\omega - a\omega_{\mathbf{q}s})}, \quad (\text{G1})$$

where sum $\sum_{\mathbf{q}s}$ runs over the aforementioned $128 \times 128 \times 32$ $\mathbf{q}$ mesh, $\omega_{\mathbf{q}s}$ is the frequency of the mode $\mathbf{q},s$ in UOC graphite, and $v_{\mathbf{q}s}$ is the modulus of its group velocity. To evaluate numerically Eq. (G1), the Dirac delta distributions are broadened with a Lorentzian having FWHM equal to twice the aforementioned value of the convergence plateau parameter η in IRG ($2 \times 0.6 \text{ cm}^{-1}$). The values for the parameters L and R appearing in Eq. (15) are determined as the values that minimize the mean squared deviation

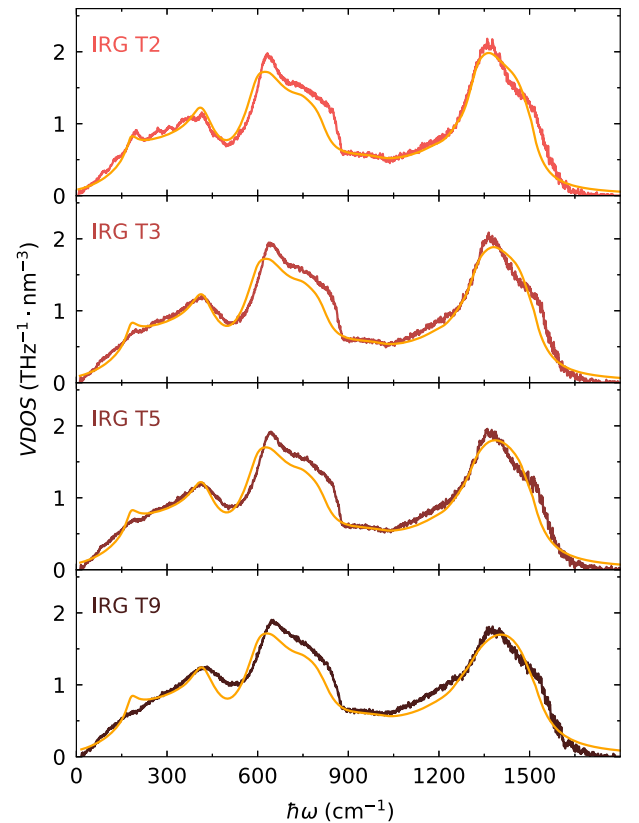


FIG. 10. Bare VDOS of IRG vs PDC VDOS. Yellow solid lines are the PDC VDOS obtained dressing the bare VDOS of pristine UOC graphite, and red-to-black lines are bare EDG VDOS of IRG graphite.

(MSD) between the PDC VDOS and EDG VDOS of irradiated graphite:

$$\text{MSD} = \sum_i (g_{\text{PDC}}(\omega_i) - g_{\text{EDG}}(\omega_i))^2. \quad (\text{G2})$$

This minimization is performed using a stochastic gradient descent optimizer implemented in PYTORCH [161].

We show the results of the fits for disorder linewidth $\Gamma_{\text{dis}}(\omega)$ in Fig. 5(a) and the corresponding predictions for the PDC VDOS in Fig. 10. We find that PDC VDOS reasonably replicates the variation with frequency and irradiation dose of the VDOS for all structures of IRG analyzed in this work. Finally, we note that, after shifting the frequencies of UOC graphite, there are no vibrational modes above approximately 1550 cm^{-1} . These modes have negligible impact on thermal transport; to decompose IRG's diffusivity above approximately 1550 cm^{-1} , we determine the disorder linewidth of frequencies above approximately 1550 cm^{-1} by fitting the linewidth value that matches the tail of disorder linewidth plot in Fig. 5(a), finding a linewidth of 24 cm^{-1} .

APPENDIX H: INFLUENCE OF ANHARMONICITY ON THERMAL CONDUCTIVITY IN IRG

Figure 1 shows that the rWTE yields results in agreement with experiments and previous studies based on molecular dynamics. We recall that the rWTE accounts for the influence of anharmonicity only when these are not spuriously affected by finite-size effects. It has been shown in Refs. [65,73] that the rWTE allows to converge thermal-conductivity calculations faster compared to the bare WTE in “strongly disordered” solids having (i) disorder-induced repulsion between energy levels [127] comparable to the intrinsic linewidth [162] and (ii) average velocity-operator elements nearly constant with respect to the energy difference between two eigenstates [65]. In this strongly disordered regime, the rWTE evaluated in a small atomistic model (containing hundreds of atoms) is expected to yield results in agreement with the bare WTE evaluated in a large model (containing several thousands of atoms). In this section, we provide additional information to strengthen the claim that, in the disordered carbon polymorphs that we studied, structural disorder is the dominant source of thermal resistance and, hence, the rWTE applied to small models yields results compatible with the WTE applied to very large models. We focus on irradiated graphite, recalling that in Fig. 1 we showed that for IRG T9 the room-temperature rWTE conductivity of a 216-atom model is practically indistinguishable from the bare WTE conductivity of a 14009-atom model. To confirm that disorder is the dominant source of thermal resistance, we evaluate the bare WTE on very large 14009-atom atomistic models and artificially rescale the anharmonic linewidths, enlarging or reducing

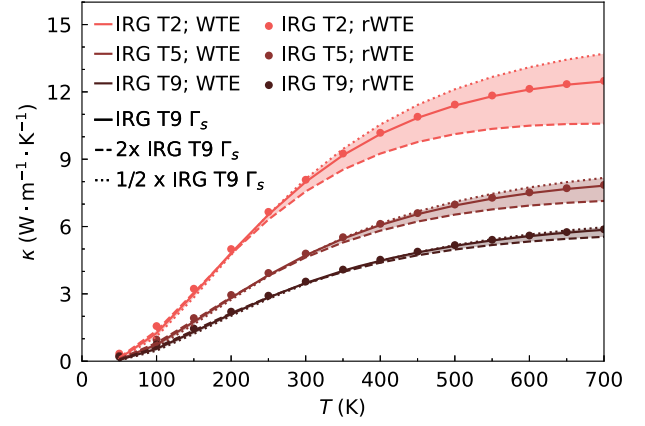


FIG. 11. Influence of anharmonicity on thermal conductivity in IRG. Solid, dashed, and dotted lines show WTE conductivity calculations for IRG with linewidth equal to linewidth derived from 216-atom model of IRG T9, linewidth of twice this value, and linewidth of half this value, respectively. The scatter points are rWTE conductivity calculations with linewidth derived from IRG T9 216. Red, brown, and black colors denote IRG T2, T5, and T9, respectively.

them by a factor 2. Figure 11 shows that the lower the structural disorder in IRG, the higher the conductivity and the more relevant the effects of anharmonicity. Importantly, for all the IRG structures considered, this artificial amplification or reduction of anharmonicity has negligible effect on the conductivity in the temperature range $50 < T \lesssim 300 \text{ K}$. Since IRG graphite is the class of carbon polymorphs with the largest conductivity and, hence, most sensitive to anharmonicity, we infer from Fig. 11 that, for the polymorphs studied, the conductivity in the temperature range $50 < T \lesssim 300 \text{ K}$ is mainly limited by structural disorder. Our findings are in broad agreement with Refs. [70,163], which discussed how compositional disorder can yield damping linewidth stronger than those due to anharmonic effects, resulting in a convergent conductivity in the low-temperature limit where anharmonicity phases out.

Finally, we note that upon increasing temperature anharmonicity becomes more relevant—we see that at 700 K it is important to use the actual physical values of the anharmonic linewidths. We also highlight how in this high-temperature regime the rWTE and WTE yield indistinguishable results, confirming that anharmonicity is strong enough to dominate over the computational broadening η employed in the rWTE (i.e., anharmonic effects are negligibly affected by finite-size effects).

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