

Anharmonicity in Thermal Insulators: An Analysis from First PrinciplesFlorian Knoop^{1,2}, Thomas A. R. Purcell,¹ Matthias Scheffler,¹ and Christian Carbogno¹¹The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft

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The anharmonicity of atomic motion limits the thermal conductivity in crystalline solids. However, a microscopic understanding of the mechanisms active in strong thermal insulators is lacking. In this Letter, we classify 465 experimentally known materials with respect to their anharmonicity and perform fully anharmonic *ab initio* Green-Kubo calculations for 58 of them, finding 28 thermal insulators with $\kappa < 10$ W/mK including 6 with ultralow $\kappa \lesssim 1$ W/mK. Our analysis reveals that the underlying strong anharmonic dynamics is driven by the exploration of metastable intrinsic defect geometries. This is at variance with the frequently applied perturbative approach, in which the dynamics is assumed to evolve around a single stable geometry.

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Efficient energy conversion is one of the key challenges for the 21st century. In this context, the thermal conductivity κ is an important material property that can strongly influence device performance and efficiency. This is the case, for example, in combustion engines, where thermal barrier coatings are used to increase operating temperatures [1,2], or in thermoelectric materials for waste-heat recovery [3]. In both cases, strong thermal insulators are required and the best available materials intrinsically have a low $\kappa < 10$ W/mK. Additional defect and grain boundary engineering can reduce κ even further [4,5]. Since heat transport is limited by effects beyond the harmonic phonon picture [6], elucidating the microscopic anharmonic mechanisms leading to a low intrinsic thermal conductivity is a prerequisite for searching the next generation of thermal insulators. However, our current knowledge about thermal insulators is rather limited. Experimental measurements are hindered by the challenge of growing macroscopic, well-characterized samples [7]. In turn, Springer Materials lists only a little over one thousand measurements for all inorganic solids, and just a few hundred semiconductors and insulators are characterized well at ambient conditions [8,9].

One way to expand on this knowledge is via computational materials discovery based on (high-throughput) screening of chemical space that is accelerated by

artificial-intelligence techniques [10–12]. To this end, reliable simulations of lattice thermal conductivities in the sub-10 W/mK regime are necessary. Typically, Peierls-Boltzmann [13,14] or Wigner-type [15,16] approaches based on a (renormalized [17]) phonon picture are employed for this purpose. However, these methods are only applicable as long as the phonon quasiparticle picture is valid, i.e., when the phonon lifetime exceeds the vibration period, as formulated within the Ioffe-Regel limit [16,18]. In this Letter, we demonstrate that the presence of metastable defect geometries is associated with strong anharmonic effects, which break the Ioffe-Regel limit and result in the exploration of different energy minima. This breakdown of the phonon picture leads to unexpectedly low thermal conductivities.

To this end, we employ the *ab initio* Green-Kubo (aiGK) method which is not affected by the aforementioned limitations since the atomic motion is evaluated via *ab initio* molecular dynamics (aiMD) simulations [19,20]. Here, we benchmark and validate the approach for 24 crystalline materials for which reliable experimental measurements are available, showing that the necessary accuracy to predict materials in the sub-10 W/mK regime can be reached, even when the phonon picture is no longer valid. We subsequently devise a hierarchical screening strategy to search for strongly anharmonic thermal insulators in a chemically and structurally diverse space of materials that covers 465 experimentally known compounds that are stable at ambient conditions, but without experimentally known κ . By that, we identify 28 materials with $\kappa < 10$ W/mK and we discuss the role of structural displacements to metastable geometries for the underlying strongly anharmonic dynamics.

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The aiGK approach evaluates the Green-Kubo (GK) equation [21,22]

$$\kappa(T) = \frac{V}{3k_B T^2} \lim_{t_0 \rightarrow \infty} \int_0^{t_0} \langle \mathbf{J}(t) \cdot \mathbf{J}(0) \rangle_T dt, \quad (1)$$

with the heat flux $\mathbf{J}(t)$, volume V , temperature T , and Boltzmann constant k_B . Here, $\langle \bullet \rangle_T$ denotes an ensemble average. In this Letter, the heat flux $\mathbf{J}(t)$ is computed from first principles along aiMD trajectories using the formalism defined in Ref. [20]. We use the FHI-aims code [23,24] and the PBEsol [25] exchange-correlation functional. By neglecting convective contributions to $\mathbf{J}(t)$, which are usually negligible in solids [26], physically well-founded strategies to overcome finite-size and -time effects can be established. This includes noise filtering of the integrand in Eq. (1), i.e., the heat ux autocorrelation function (HFACF), and accounting for long-wavelength excitations missing in the simulation cell via reciprocal-space interpolation [20] as discussed in detail in a separate work [27] and implemented in FHI-vibes [28]. Computational parameters were chosen to obtain reliable values of κ at room temperature: All aiMD simulations are performed in extended supercells (160–256 atoms) with trajectory lengths of up to 60 ps. Error bars are estimated by ensemble averaging over at least three independent trajectories. Lattice expansion is accounted for by minimizing the thermal pressure at 300 K [29]. Further details are given in the Supplemental Material [23,25,30–32].

To benchmark the aiGK approach, we investigate 24 dielectric compounds, for which experimental values of the bulk thermal conductivity κ at room temperature are reported in the literature [7,33–53]. The reference data covers two orders of magnitude in κ , but also carries an uncertainty which is rooted in differences in sample quality and experimental techniques [9]. For instance, available data for tin selenide (SnSe) varies by up to 50% [7]. As shown in Fig. 1, the aiGK calculations provide a satisfactory agreement with experiment over the whole range of κ : For mildly anharmonic conductors with $\kappa > 20$ W/mK, the quality of the predictions [mean absolute percentage error (MAPE) of 22%] is on par with results from advanced perturbative methods [54,55]. Good accuracy is retained also for strongly insulating materials with $\kappa < 20$ W/mK, which show a comparable MAPE of 28% and a resulting mean absolute error (MAE) of just 1 W/mK. Accounting for strongly anharmonic effects is decisive to reach this accuracy in the sub-10 W/mK regime, as exemplified by the fact that the aiGK method correctly predicts κ to be 1.4 W/mK for copper iodide (CuI), just 16% smaller than in experiment. In contrast, different phonon-based approaches yield values between 5.8 and 6.8 W/mK, overestimating experiment by $\gtrsim 350\%$, cf. Supplemental Material, Sec. IV [56–60]. Even more importantly, the accurate lifetimes obtained from fully anharmonic aiMD

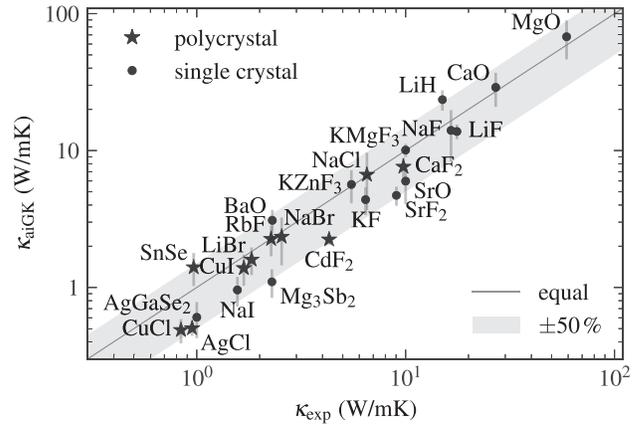


FIG. 1. Correlation between measured (circles: single-crystals; stars: polycrystals, [7,33–53]) and computed (aiGK) thermal conductivities at room temperature on a log-log scale. The gray area indicates 50% deviation in terms of κ from experiment and the error bars denote the standard error between trajectories. Where multiple experimental references were available, the average was taken.

reveal a breakdown of the phonon picture in this compound, cf. Supplemental Material, Sec. VI [30].

For CuI, an analysis of the aiMD dynamics confirms that strong anharmonic effects are at play: Figure 2 shows the time evolution of the anharmonicity measure $\sigma^A(t)$ introduced in Ref. [61], which quantifies the differences between the actual and the harmonic potential-energy surface (PES) and hence would vanish in perfectly harmonic materials [30,61]. The sudden increase of $\sigma^A(t)$ from its value of 0.5 to 1.2 signals that the harmonic approximation becomes qualitatively incapable to describe the dynamics: In this several picoseconds long period, a metastable Frenkel defect is formed [62]. As shown in

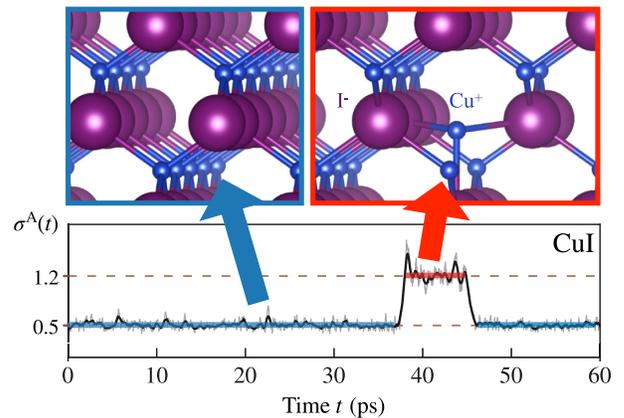


FIG. 2. Anharmonicity measure $\sigma^A(t)$ for KCaF_3 evaluated at each time step during one aiMD trajectory. The upper row shows averaged positions in $(10\bar{1})$ direction for time periods with $\sigma^A(t) \approx 0.5$ (blue), and increased $\sigma^A(t) \approx 1.2$ (red), in which one Cu cation moves to an interstitial site along $(\bar{1}11)$.

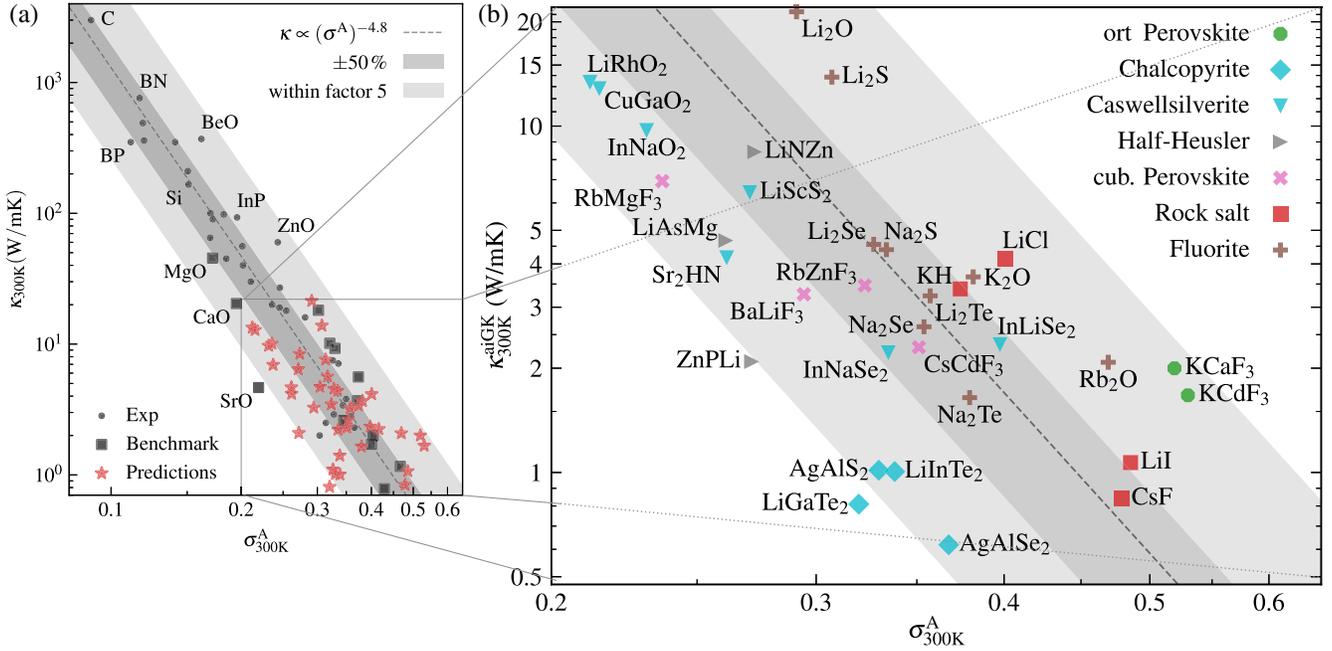


FIG. 3. Thermal conductivity κ vs anharmonicity at 300 K, $\sigma_{300\text{K}}^A$ (tabulated data given in the SM [30]). The dashed trend line and gray areas are shown to guide the eye. (a) The qualitative scaling of κ with $\sigma_{300\text{K}}^A$ suggested in Ref. [61] and used to disregard good thermal conductors during screening is showcased using experimental measurements (black circles) as well as our aiGK calculations for the benchmark materials from Fig. 1 (black squares) and predictions from subplot (b) (red stars). (b) Thermal conductivity aiGK predictions for materials with $\sigma_{300\text{K}}^A > 0.2$. The symbol and color code indicate the lattice type.

Fig. 2, one Cu cation moves to an interstitial site along the $(\bar{1}11)$ direction and vibrates around this metastable defect geometry, before it jumps back to the zinc blende equilibrium configuration associated with $\sigma^A(t) \approx 0.5$. This temporary defect formation may be viewed as a local precursor of the superionic β -phase of CuI, which is known to exhibit a considerable defect concentration $> 10\%$, but only becomes stable at significantly higher temperatures (> 643 K) [63].

In the next step, we employ the aiGK method to screen for thermal insulators via a hierarchical workflow [12,30,31,64,65]: We study 465 experimentally known binary and ternary materials for which κ at room temperature has not been measured. This set of materials covers seven space groups (62, 122, 166, 186, 216, 221, 225), including not only simple rock salt materials with two atoms per unit cell, but also more complex compounds, e.g., orthorhombic perovskites with 20 atoms in the unit cell. Note that only materials with elements lighter than lanthanum are considered to avoid potential artifacts in the dynamics associated with the treatment of spin-orbit coupling. First, we compute the band gap and the anharmonicity measure at 300 K, $\sigma_{300\text{K}}^A$, for all 465 compounds. The results cover the range $\sigma_{300\text{K}}^A = 0.1\text{--}0.7$, cf. Supplemental Material. Over 50% of the materials feature an anharmonicity of $\sigma_{300\text{K}}^A \approx 0.23$ or less, in line with Ref. [61]. Second, we exclude materials with DFT-PBEsol band gaps below 0.2 eV to ensure that electronic

contributions to heat transport are negligible and we discard more harmonic materials with $\sigma_{300\text{K}}^A < 0.2$ to avoid unnecessary and expensive calculations, since those materials can be expected to be good thermal conductors [61] that can be treated with perturbative approaches. The first aspect is substantiated by Fig. 3(a), which shows that the materials with $\sigma_{300\text{K}}^A < 0.2$ feature κ values (much) larger than 10 W/mK. From the remaining $\sigma_{300\text{K}}^A > 0.2$ pool, we generate a diverse test set of materials that uniformly cover the range $\sigma_{300\text{K}}^A = 0.2\text{--}0.6$. Materials with a particularly slow phonon dynamics were excluded to ensure that simulation time convergence can be reached within reasonable trajectory lengths ≤ 60 ps, as detailed in the Supplemental Material [30]. Finally, we obtain thermal conductivities for 34 materials for which κ has *not* been measured yet, see Fig. 3(b).

Although the σ^A criterion is only qualitatively valid for the identification of thermal insulators, no good thermal conductors are found within this set. On the contrary, we find 28 new materials with a thermal conductivity of $\kappa_{\text{aiGK}} < 10$ W/mK, 6 of which with an ultralow conductivity $\lesssim 1$ W/mK. These compounds comprise simple binary, cubic materials such as the rock salts CsF and LiI or the fluorite Na_2Te , but also more complex structures such as the KCdF_3 and KCaF_3 perovskites. Furthermore, we find four chalcopyrites (AgAlSe_2 , AgAlS_2 , LiGaTe_2 , LiInTe_2), a material class that has recently gained interest for thermoelectric applications [66–68]. In the strongly

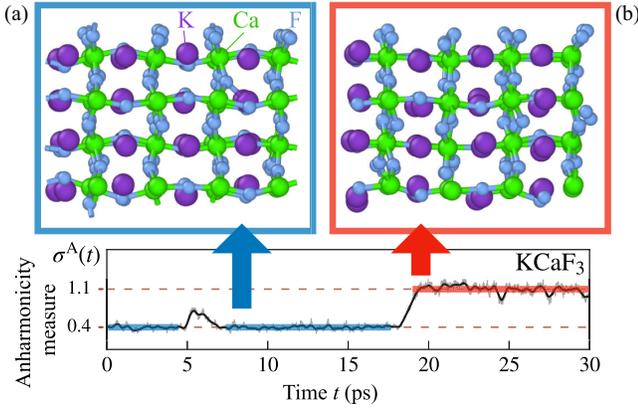


FIG. 4. Anharmonicity measure $\sigma^A(t)$ for KCaF_3 evaluated at each time step during one aiMD trajectory. The upper row shows time-averaged positions in (100) direction for the periods with (a) $\sigma^A(t) \approx 0.4$ (blue), and (b) $\sigma^A(t) \approx 1.1$ (red). Note the rearrangement of Ca-F bonds from zigzag to straight shape when comparing (a) and (b). See Supplemental Material for a video of the tilt rearrangement around 18 ps.

insulating regime shown in Fig. 3(b), the simple power-law correlation between κ and $\sigma_{300\text{K}}^A$ observed in Fig. 3(a) is no longer valid due to the increased variety and complexity of the compounds. This can be explained by comparing two materials (LiGaTe_2 , KCaF_3); more details are given in the Supplemental Material [30]: The strongly insulating character of the chalcopyrite LiGaTe_2 ($\kappa_{\text{aiGK}} = 0.8$ W/mK) can already be rationalized in a harmonic picture, since most phonon branches of this compound, including the acoustic ones, exhibit only an extremely weak dispersion, cf. Supplemental Fig. 1 [30]. Accordingly, only few modes with $\omega < 4$ THz can contribute to thermal transport, and even these feature very low group velocity $\lesssim 1.5$ km/s, cf. Supplemental Fig. 2, [30]. Since this is a common property in chalcopyrites, Plata *et al.* were able to correctly predict thermal insulators in this material class [67], even if the employed perturbative approach might overestimate κ due to the low-order description of anharmonicity. In comparison, KCaF_3 features much more dispersive phonon frequencies: The majority of modes across the whole frequency range contributes to heat transport with considerably larger group velocities that reach 5 km/s, suggesting a moderately good thermal conduction. In this case, the strongly insulating character of KCaF_3 ($\kappa_{\text{aiGK}} = 2$ W/mK) can only be explained by its strong anharmonicity ($\sigma_{300\text{K}}^A = 0.5$), which results in extremely low phonon lifetimes $\lesssim 1$ ps. As shown in Fig. 4, this is reflected in the real-space dynamics that displays temporary rearrangements towards different, metastable configurations. These involve an increase of $\sigma^A(t)$, as discussed above for CuI in Fig. 2. Unlike CuI however, in which these rearrangements correspond to the formation of intrinsic point defects, we observe more extended distortions in KCaF_3 that span multiple primitive unit cells, cf. Fig. 4. As discussed in the

Supplemental Material [30], these configurations are associated to reorientations of the CaF_3 octahedra into different, nearly degenerate octahedral tilting patterns. These metastable configurations are a common feature of perovskite materials and their dynamics often leads to the stabilization of higher-temperature phases [69–71]. For KCaF_3 , the described dynamics is a precursor of the cubic phase, which becomes stable at temperatures > 550 K [72].

Note that these temporary atomic rearrangements into metastable geometries are not observed in all trajectories. Nonetheless, their microscopic origin, i.e., a strongly-anharmonic PES featuring multiple minima, influences the whole dynamics. Even when these minima are not explicitly explored, harmonic and anharmonic contributions are at least of equal importance, as signaled by the high σ^A value ≥ 0.5 , and the fact that the fully anharmonic lifetimes, which violate the Ioffe-Regel limit in both compounds, demonstrate a breakdown of the phonon picture, as shown in Supplemental Fig. 3 and Sec. VI in the Supplemental Material [30]. These observations are direct evidence that the existence of barely accessible metastable states is sufficient to increase the anharmonicity in CuI and KCaF_3 beyond the level at which it can be treated as a perturbation. We also note that these localized effects are different from other local, anharmonic scattering mechanisms such as “rattlers” that do not involve temporal lattice distortions [73].

In summary, we presented a comprehensive set of aiGK calculations to search for thermal insulators. By hierarchically screening over 465 experimentally known materials, we identified 28 new materials with $\kappa < 10$ W/mK including 6 compounds with ultralow $\kappa \lesssim 1$ W/mK. Our analysis reveals that the dynamics in many thermal insulators is not limited to displacements around a *single*, stable equilibrium geometry, as assumed in perturbative approaches like the Boltzmann transport equation (BTE). On the contrary, strongly anharmonic dynamical effects such as point defect formation in CuI or extended rearrangements in KCaF_3 involve multiple metastable geometries that need to be accounted for. Besides providing a microscopic explanation of the insulating character of these materials, our findings suggest a fundamentally different route for reducing κ that is complementary to traditional phonon-based approaches [74,75]: For instance, incorporating defects that promote the formation of additional metastable states [76,77] can strengthen the anharmonic effects leading to low thermal conductivity even at temperatures at which these states are not fully accessible. Our results and the more than 10 ns of available PBEsol-aiMD trajectories can be used to benchmark the various methods available for computing κ in increasingly anharmonic materials. Similarly, this data lends itself to train machine-learning potentials to accelerate κ predictions in the future [78–80] and to develop improved descriptors for κ [10–12]. In this regard, quantitative models that go beyond the qualitative σ^A scaling used in this work to

identify anharmonic materials can accelerate materials' space exploration considerably.

The electronic-structure theory calculations and aiMD trajectories produced in this project are available via the NOMAD repository [81]. Resources for the plots can be found on figshare [82].

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