

Phonon hydrodynamic regimes in sapphire

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When an ideal insulator is cooled, four regimes of thermal conductivity are expected to emerge one after another. Two of these, the Ziman and the Poiseuille, are hydrodynamic regimes in which collision among phonons are mostly normal. It has been difficult to observe them, save for a few insulators with high levels of isotopic and chemical purity. Our thermal transport measurements, covering four decades of temperatures between 0.1 K and 900 K, reveal that sapphire displays all four regimes, despite its isotopic impurity. In the Ziman regime, the thermal conductivity exponentially increases, attaining an amplitude as large as 35 000 W/Km. We show that the peak thermal conductivity of ultrapure, simple insulators, including diamond, silicon, and solid helium, is set by a universal scaling depending on isotopic purity. The thermal conductivity of sapphire is an order of magnitude higher than what is expected by this scaling. We argue that this may be caused by the proximity of optical and acoustic phonon modes, as a consequence of the large number of atoms in the primitive cell.

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I. INTRODUCTION

A remarkable accomplishment of the quantum theory of solids has been a compelling account of heat transport by phonons in insulators. Peierls distinguished between two kinds of phonon-phonon scattering processes: those conserving momentum (normal events) and those which relax it (Umklapp events) [1]. The change in the relative weight of these two kinds of processes with cooling led Guyer and Krumhansl [2] to identify four thermal-transport regimes in an ideal insulator. These four regimes, however, have rarely been observed experimentally all together. Let us recall them one by one.

At temperatures exceeding the Debye temperature, the phonon wave vector k is comparable to the width of the Brillouin zone G . In this regime, called kinetic, scattering between two phonons can give rise to a third phonon with a wave vector larger than G . Such an “Umklapp” event [1] leads to loss of $\hbar G$ momentum and a resistance to the flow of heat. The average number of phonons increases linearly with T and thermal conductivity κ decreases linearly with warming $\propto T^{-1}$ [Fig. 1(a)]. The scattering rate in this limit is bounded by $\tau_p^{-1} = \frac{\hbar}{k_B T}$ [3,4]. As the sample is cooled down, the wave vector of phonons is reduced, following

$k \simeq k_B T / \hbar v_s$. As a consequence, Umklapp processes rarefy, and instead momentum-conserving (normal) collisions become dominant [Fig. 1(b)]. This leads to hydrodynamics, and the two regimes known as Ziman and Poiseuille. An exponential increase of κ in the Ziman hydrodynamic regime is driven by the decay of the phonon population capable of giving rise to Umklapp events. In the Poiseuille hydrodynamic regime, normal collisions dominate and Umklapp collisions become even less frequent than boundary collisions; this boosts heat conduction and leads to a κ varying faster than T^3 . Cooling further, phonons travel across the sample without meeting other phonons. Their wavelength becomes so long that the crystal wall becomes the exclusive scattering center. In this ballistic (or Casimir [5]) regime, the thermal conductivity follows a T^3 temperature dependence. These four regimes [2,6,7] provide a road map in the quest for detecting signatures of phonon hydrodynamics in the two regimes where normal collision between phonons dominate, i.e., the Ziman and Poiseuille regimes [6–8].

The two hydrodynamic regimes are rarely observed in real insulators [9], presumably because they are severely hindered by the presence of defects, and chemical or isotopic impurities. Recent studies, however, have found that exceptional purity is not the necessary condition for detecting a phonon Poiseuille flow. It has been observed in strontium titanate, a quantum paraelectric [10], in layered solids such as graphite and black phosphorus [11,12], and in semimetallic antimony [13].

As for the Ziman heat-transport regime, previous reports of its observation have been restricted to a few ultrapure insulators composed of single-isotope elements, such as solid helium [14,15], CsI [9], and NaF [16]. Surprisingly, the thermal conductivity of naturally available sapphire (Al_2O_3)

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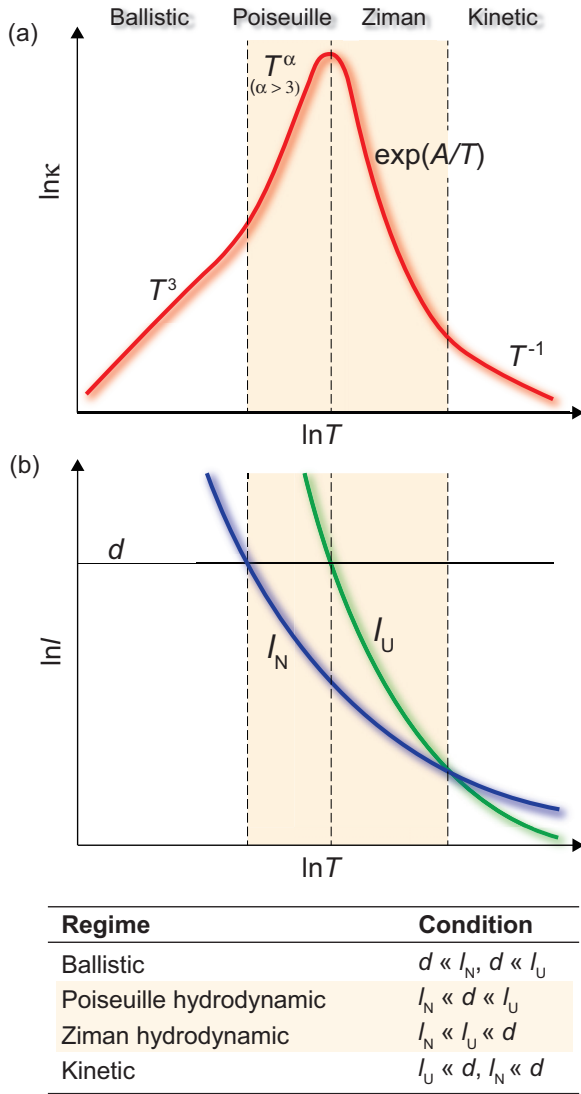


FIG. 1. Four thermal conductivity regimes. (a) Four regimes in temperature dependence of thermal conductivity. (b) The four regimes are distinguished by the hierarchy of the three length scales l_U , l_N , and d . These are the mean-free path of Umklapp and normal scattering, and the sample dimension. Dominant normal collisions lead to two hydrodynamic regimes in between the ballistic and kinetic regimes.

displays the Ziman regime. While aluminum is pure (^{27}Al : 100%), this is not the case of oxygen (^{16}O : 99.76%, ^{17}O : 0.04%, ^{18}O : 0.21%). Nevertheless, in 1951, Berman [17] reported an exponential temperature dependence based on a limited number of data points (see Fig. S3(a) [18]). The issue has not been reexamined since then.

In this paper, we present a study of heat transport in sapphire, documenting its putative Ziman regime and Poiseuille regime. Prior to our study, solid helium has been the only case in which both regimes have been observed. We find that naturally occurring sapphire stands out among insulators by displaying both hydrodynamic regimes and an extreme peak-thermal conductivity at their boundary. The observed conductivity exceeds what is expected from a universal plot of dimensionless thermal conductivity as a function of isotopic

impurities obeyed by other insulators. We argue that the large number of atoms per primitive cell and/or the proximity of the optical and acoustic phonon branches are the key ingredients for this robustness against isotopic disorder.

This remarkable robustness leads to two insights. First, phonon hydrodynamics is a phenomenon more ubiquitous than believed previously. Previous studies on SrTiO_3 [10], black phosphorus [11], and graphite [12] suggested that proximity to a ferroelectric instability and/or a layered crystal structure are crucial for the frequent momentum exchange between phonons. Our present observation implies that none of them is a necessary ingredient for the emergence of phonon hydrodynamics. Second, a promising avenue to find high-thermal conductivity materials opens up. Even in absence of isotopic purification, a robust Ziman transport regime allows achieving a very high thermal conductivity.

II. RESULTS

Sapphire has a layered trigonal structure with the space group $R\bar{3}c$. Each layer consists of edge-shared AlO_6 octahedra arranged in a honeycomb pattern [Fig. 2(a)], and the layers are stacked such that each octahedron shares edges with the layers above and below it [Fig. 2(b)].

Let us quantify the isotopic purity of sapphire. Isotopic disorder can be quantified by a dimensionless parameter [19]

$$g = \sum_i f_i (\Delta M_i / M)^2. \quad (1)$$

Here, f_i is the concentration of the i th isotope, whose mass M_i differs from the average mass $M = \sum_i f_i M_i$ by $\Delta M_i = M_i - M$. In naturally abundant sapphire, that is in absence of any isotopic purification, $g \sim 34 \times 10^{-6}$. This is to be compared with that of the natural abundant diamond (74×10^{-6}) and silicon (200×10^{-6}). Here, to evaluate g , the isotopic mass and percent natural abundance are taken from Refs. [20,21], respectively. Thanks to the maturation of crystal-growth technique, highly pure industrial sapphire single crystals with minimal contamination and defects are nowadays available.

The sapphire samples used in this study were purchased from Kyocera corporation. In order to acquire the thermal conductivity data in a wide temperature range, we employed the 3ω method and the standard steady-state method. The 3ω measurements were performed in the temperature range between 80 K and 900 K by utilizing the N_2 cryostat and the electrical furnace. The measurements, using a one-heater-two-thermometers steady-state method, were performed in the temperature range between 0.1 K and 300 K in a commercial measurement system (Quantum Design PPMS) and in a homemade dilution refrigerator. (See the details of measurement in the Supplemental Material Ref. [18]).

Figure 2(e) shows temperature dependence of thermal conductivity κ of sapphire. As seen in the figure, there is a satisfactory match between the data obtained by the different methods in the different temperature ranges. The four conductivity regimes are all visible. In the highest temperature regime, with decreasing temperature, κ grows following $1/T$. Upon further cooling, the temperature variation evolves into the exponential behavior $\kappa \propto \exp(A/T)$, signaling an entry to the Ziman regime. The result agrees with what was found

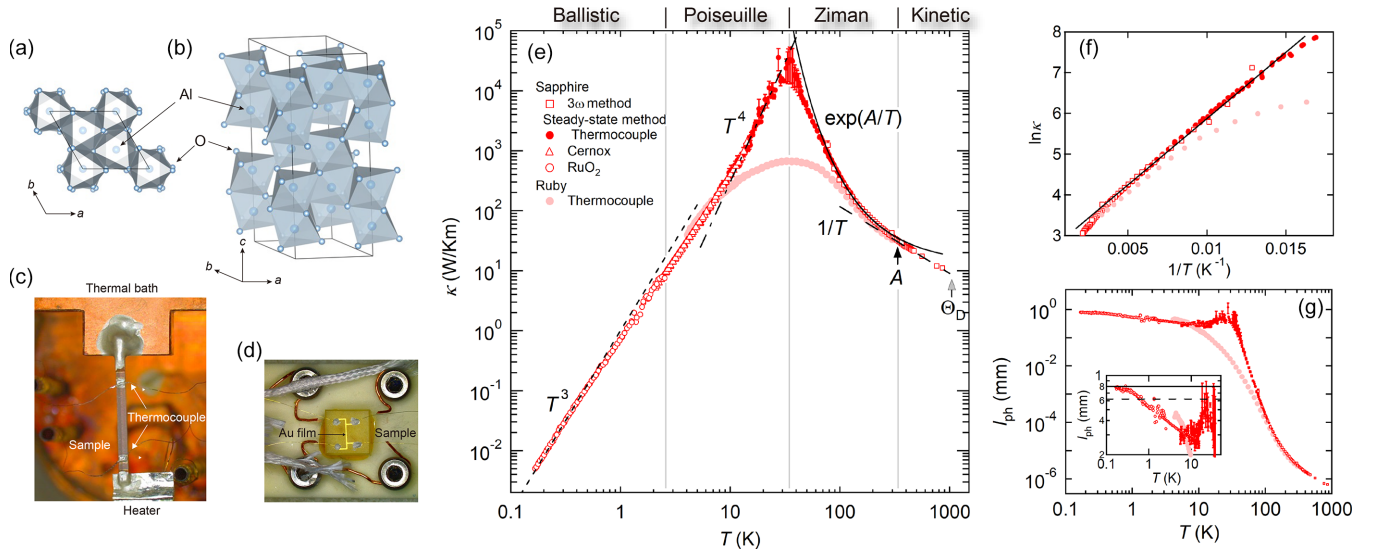


FIG. 2. Thermal conductivity of sapphire. (a) Top view and (b) side view of crystal structure of sapphire. A thermal conductivity-measurement setup for (c) steady-state method and (d) 3ω method. (e) Thermal conductivity κ of sapphire and ruby as a function of temperature. For sapphire, κ displays an exponential increase $\kappa \propto \exp(A/T)$ with $A = 337$ K and forms a prominent peak around 40 K, below which κ falls faster than T^3 as a signature of phonon Poiseuille flow. Temperature variation of κ changes from $1/T$ to the exponential across $T = A$. The characteristic temperature dependence of κ allows to detect the four regimes as denoted on the top of panel (e). An Arrhenius plot shown in the panel (f) clearly demonstrates the presence and absence of exponential behavior in sapphire and ruby, respectively. (g) A pronounced Poiseuille peak is detected in temperature dependence of phonon mean-free path, which increases aiming to attain the crystal dimension at low temperature as shown in the inset of panel (g).

by Berman (see Fig. S3(a) [18]). The exponential increase persists down to 40 K, reaching a magnitude as large as 35 000 W/Km. Such a magnitude has only been attained in isotopically pure diamond [22] and Si [19] [see Figs. 6(a) and 6(b)]. Below the peak, κ falls rapidly following $\propto T^4$. Such a faster than T^3 decrease of κ is a hallmark of the phonon Poiseuille flow [10–12,23–26]. Further cooling the sample below 1 K eventually leads to the ballistic regime and a T^3 behavior. As shown in Fig. 3, our results, including the exponential and $1/T$ evolution of κ , are in perfect agreement with the first-principle calculations [27]. Note that κ becomes size dependent at low temperature, but calculations were conceived for a sample of infinite size. Therefore, theory and experiment do not match in the vicinity of the peak and below it, i.e., in the Poiseuille regime.

The peculiar features of $\kappa(T)$ can be seen by an examination of the phonon mean-free path l_{ph} extracted from the thermal conductivity, the specific heat C , and the phonon velocity v_s ($l_{ph} = \frac{3\kappa}{v_s C}$). Its evolution with temperature is shown in Fig. 2(g). An exponential increase of l_{ph} ends up attaining a magnitude close to the sample dimensions [the horizontal dashed line in the inset of Fig. 2(g)]. At low temperature, l_{ph} displays a local peak (around 30 K) and a local minimum (around 10 K) as commonly observed in systems where hydrodynamic phonon transport becomes dominant [10–13,25]. In the millikelvin temperature, l_{ph} saturates to a length comparable to sample dimensions as expected in the ballistic regime [see the inset of Fig. 2(g)].

We found that the Poiseuille regime is sensitive to the quality of the sample surface. Such a surface sensitivity was detected in samples purchased from two different manufacturers (Kyocera and Orbray). Figure 4(a) and its inset show

the thermal conductivity and the mean-free path, respectively, of two samples with a cross section of 0.5×0.6 mm² ($d = 0.62$ mm) and 0.3×0.34 mm², ($d = 0.36$ mm) purchased from the same company (Kyocera). Here, d is a characteristic sample dimension defined as $d = 2\sqrt{wt}/\pi$, where w and t are sample cross-section dimensions. The data for the sample

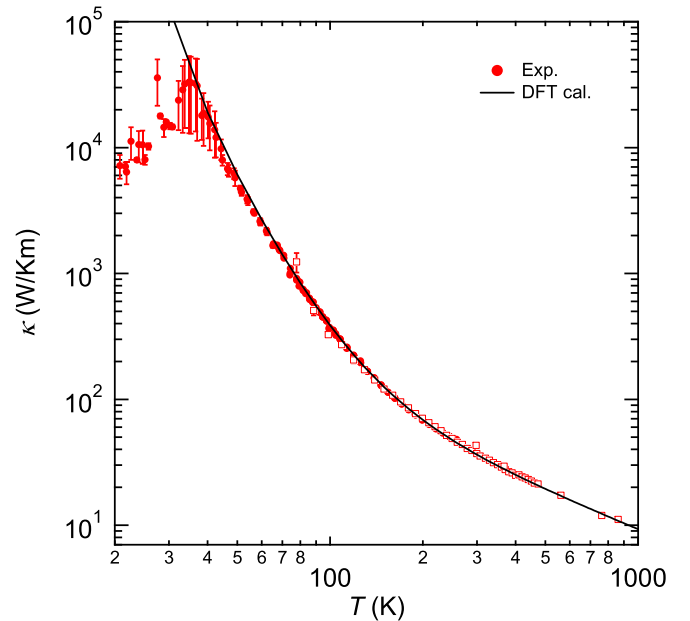


FIG. 3. A comparison between experiment and theory. A comparison of experimental thermal conductivity with DFT calculations [27].

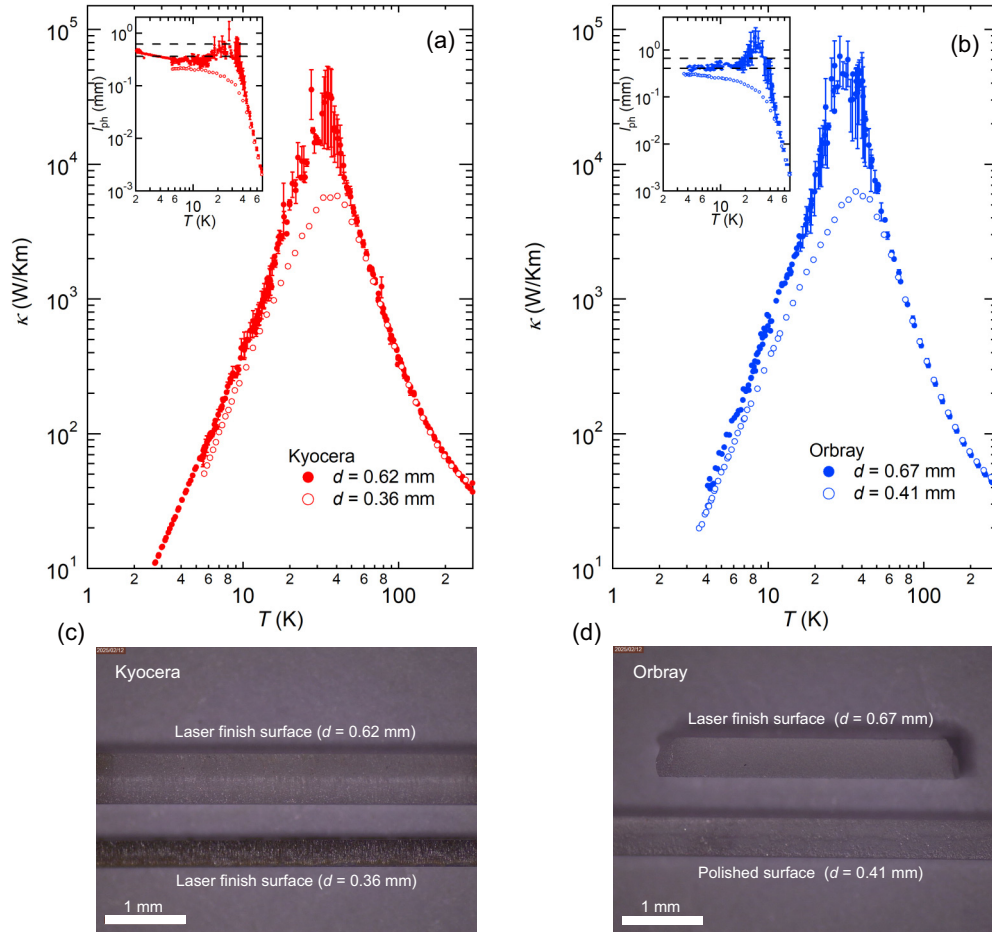


FIG. 4. Presence and absence of the Poiseuille regime. Thermal conductivity of the sapphire samples with different cross-section areas are compared. The samples purchased from Kyocera and Orbray corporation are shown in the panels (a) and (b), respectively. Insets display temperature variations of phonon mean-free path. The Poiseuille regime is absent in samples with a small cross section, which have rough surfaces compared with those showing the Poiseuille flow as shown in the panels (c) and (d).

with a larger cross section is the same as the one shown in Fig. 2(e). One can see that the Poiseuille features are absent in the sample with a smaller cross section. In contrast, the Ziman regime is present in both samples. The two samples were prepared from wafers with a thickness of 0.5 mm and 0.3 mm, by laser cutting with the same laser power. Thus, four out of six surfaces are laser finish and two surfaces are mirror finish. However, the darker color of the laser-finish surface of the smaller sample [Fig. 4(c)] implies rough surface and/or damage near the surface induced by laser heat, suspected to be causing the disappearance of the Poiseuille regime.

A similar suppression of the Poiseuille hydrodynamics is observed by polishing the Orbray sample ($0.5 \times 0.7 \text{ mm}^2$, $d = 0.67 \text{ mm}$) which was also prepared from a wafer by laser cutting. By polishing one of the laser-finish surfaces with a sand paper and reducing the thickness from 0.7 mm to 0.26 mm, the Poiseuille regime disappears [Fig. 4(b)]. Again, the rough surface of the polished sample seen in Fig. 4(d) may cause this suppression.

Thus, we can optically detect a difference between surfaces which show Poiseuille flow and those which do not. This indicates that the surface roughness is crucial for the Poiseuille regime. Let us note that the wavelength of acoustic phonons

at $T \sim 30 \text{ K}$ (where the Poiseuille phenomena occurs) is about $\lambda \sim 2\pi\hbar v_s/k_B T \sim 18 \text{ nm}$. According to Ref. [28], the proportions of specular and diffuse reflection is given by $p(\lambda) = \exp(-16\pi^3\eta^2/\lambda^2)$, where η represents the asperity height of surface. Although the quantitative characterization of the surface roughness is hard to achieve, the phonon scattering at the boundary is expected to be specular only when the surface rugosity is negligible over a length scale smaller than λ (i.e., $\lambda \gg \eta$). Higher rugosity would make it diffusive (i.e., $\lambda \ll \eta$). Our observation of the correlation between the surface smoothness and the Poiseuille regime suggests that the latter requires specular phonon reflection at the boundary. This strict surface-smoothness criterion for the Poiseuille regime may also explain another intriguing feature. In isotopically pure materials such as NaF [16] and CsI [9], the Ziman regime has been detected, but the Poiseuille regime is absent.

Let us put under scrutiny the exponential behavior $\kappa \propto \exp(A/T)$ in the Ziman regime. The Arrhenius plot shown in Fig. 2(f) yields $A = 337 \text{ K}$. The Debye temperature is $\Theta_D \sim 1047 \text{ K}$, and therefore: $A \sim \Theta_D/3$, as predicted by Peierls [1]. One can see from Fig. 2(e) that A and the crossover temperature from the kinetic to Ziman regime are comparable to each other.

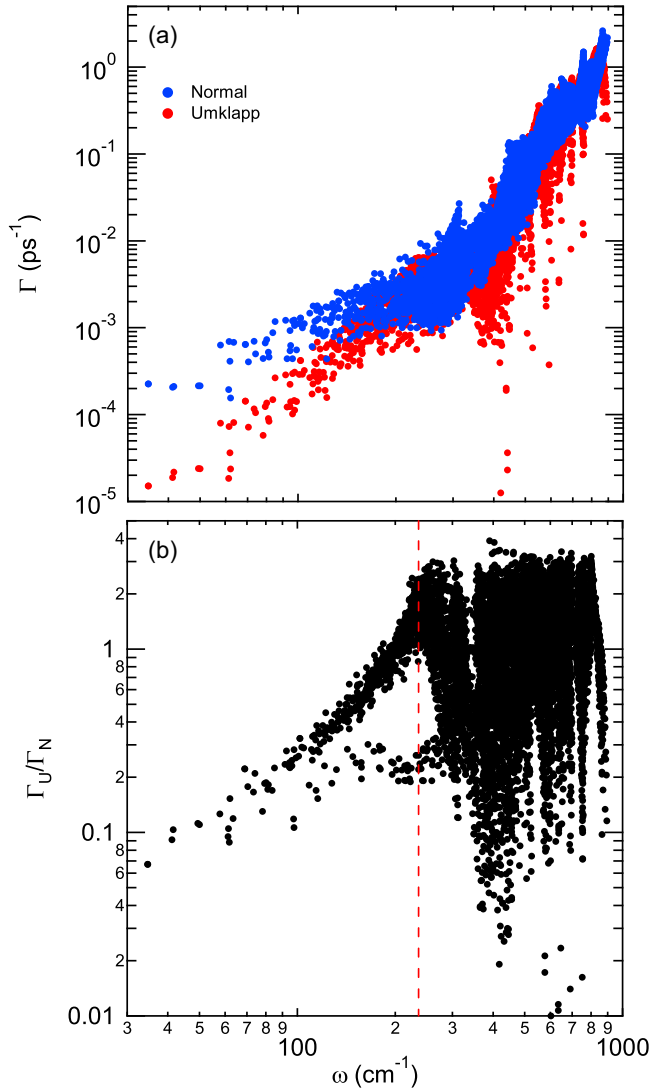


FIG. 5. Normal and Umklapp scattering rate. Scattering rate of (a) the Normal Γ_N and Umklapp Γ_U processes and (b) their ratio Γ_U/Γ_N as a function of frequency. The vertical dashed line at $\omega = 236 \text{ cm}^{-1}$ in the panel (b) denotes below which the normal scattering dominates over the Umklapp scattering.

This experimental observation is compatible with the theoretical calculations. Figure 5(a) displays the scattering rate of normal Γ_N and Umklapp Γ_U processes as a function of frequency (See the details of calculations in the Supplemental Materials [18]). One can see that the rate of normal scattering is higher than Umklapp scattering at low frequencies. Figure 5(b) shows the frequency dependence of Γ_U/Γ_N . The striking feature of this figure is the clear presence of a cutoff frequency of $\omega \sim 236 \text{ cm}^{-1}$. Above this frequency, there is a larger variation between the abundance of normal and Umklapp events for a given frequency. Below this frequency, this is no more the case and normal scattering events become dominant over the Umklapp one. Thus, there is an energy scale associated with the hydrodynamic regime. One should emphasize that $\omega \sim 236 \text{ cm}^{-1}$ ($\sim 336 \text{ K}$) is in perfect agreement with the experimental $A \sim 337 \text{ K}$, supporting the presence of Ziman regime.

Hydrodynamic regimes in sapphire are destroyed in presence of a significant population of pointlike defects. This is shown by comparison with a ruby sample purchased from Orbray corporation, in which about 3% of Al atoms are substituted by Cr atoms. As seen from Figs. 2(e)–2(g), the exponential increase, the faster than T^3 decrease, and the non-monotonic mean-free path are all absent in the ruby sample. As a result, the maximum κ is damped by a factor of 50. The phonon mean-free path l_{ph} smoothly increases all the way down to 3 K, approaching a value close to the sample dimension [represented by the solid horizontal line in the inset of Figs. 2(g)] without showing any trace of the phonon hydrodynamics. In other words, a defect concentration of 3% does not affect the kinetic regime, but wipes out the Ziman and the Poiseuille regimes.

III. DISCUSSION

Let us compare sapphire with other high-purity insulators. Although this has remained unnoticed, isotopically enriched diamond [22], Si [19], and LiF [29] with the isotope compositions summarized in Table 1, display a similar exponential behavior, asymptotically evolving to a $1/T$ dependence at a temperature close to A , which is shown in Figs. 6(a)–6(c). This is also true of the systems composed with one isotope, NaF [16,30] and CsI [9], as seen in Figs. 6(d) and 6(e). In all these cases, κ deviates from the $1/T$ dependence (dashed line) around $T \sim A$, extracted from an Arrhenius fit, and asymptotically follows the exponential temperature dependence. Finally, as seen in Fig. 6(f), this is also the case of the two helium isotopes. ^4He [14] and ^3He [15] both display a similar behavior. However, in both cases, the kinetic regime has not been attained.

In Fig. 7, temperature dependence of thermal conductivity of sapphire is compared with those of ultrapure systems shown in Figs. 6(a)–6(f). One should emphasize that even in the absence of isotope purification, the peak thermal conductivity of sapphire competes with the isotopically purified Si [19] and diamond [22].

Figure 8 reveals a universal feature common to high-purity insulators. The amplitude of A as a function of Θ_D in different systems is shown in Fig. 8(a). One can see that in all of them, it lies between $\Theta_D/5$ and $\Theta_D/2$.

The upper bound to A can be justified considering Umklapp phonon-phonon collisions. They are only possible if the wave vector of each colliding phonon, k , exceeds $G/4$, where $G = 2\pi/a$ (a : the lattice constant) is a reciprocal lattice vector. Since $k = k_B T / \hbar v_s$, the energy of such a phonon is given by $k_B T_U = \pi \hbar v_s / 2a$. Assuming linear dispersion [$\omega_D = k_B \Theta_D \sim v_s \hbar (G/2)$], one finds $T_U \sim \Theta_D/2$. The average number of such a phonon decreases exponentially with temperature, $\langle N \rangle \sim \exp(-T_U/T) \sim \exp(-\Theta_D/2T)$, so that the thermal conductivity is $\kappa \propto \exp(\Theta_D/2T)$. This leads to $A = \Theta_D/2$ in good agreement with what is seen in Fig. 8(a).

It is instructive to consider exceptions to this correlation. For example, in SrTiO_3 , $A = 20 \text{ K}$ ($\sim \Theta_D/25$) [10]. This unusually small value points to a role played by low-energy soft phonons of this quantum paraelectric and their coupling with acoustic modes [31]. The case of bismuth, where $A = 14.3 \text{ K}$

TABLE I. A comparison of various parameters of sapphire with untrapure insulators. Comparison of crystal structure, number of atoms in the unit cell N , isotope composition, dimensionless parameter of isotopic disorder g , Debye temperature Θ_D , and A for materials exhibiting the exponential rise in thermal conductivity. The percent natural-abundance data is from Ref. [21]. Isotope composition of isotopically purified diamond, Si, LiF, and ^3He is from Refs. [15,19,22,29].

	Crystal structure	N	Isotope	Natural abundant sample		Isotopically pure sample		Θ_D (K)	A (K)
				Composition (%)	$g (\times 10^{-6})$	Composition (%)	$g (\times 10^{-6})$		
Sapphire	Trigonal	30	^{27}Al	100	34	—	—	1047	337
			^{16}O	99.76		—			
			^{17}O	0.04		—			
			^{18}O	0.21		—			
Diamond	Cubic (fcc)	8	^{12}C	98.9	74	99.9	7	2220	425
			^{13}C	1.1		0.1			
Si	Cubic (fcc)	8	^{28}Si	92.2	200	99.995	0.08	645	331
			^{29}Si	4.7		0.0046			
			^{30}Si	3.1		0.0004			
LiF	Cubic (Rock salt)	8	^6Li	7.6	1460	0.01	2	735	184
			^7Li	92.4		99.99			
			^{19}F	100		100			
NaF	Cubic (Rock salt)	8	^{23}Na	100	0	—	—	488	128
			^{19}F	100		—			
CsI	Cubic (bcc)	2	^{133}Cs	100	0	—	—	128	34
			^{127}I	100		—			
^4He	Hexagonal	2	^3He	0.0001	0.06	—	—	36.7	7.3
^3He	Cubic (bcc)	2	^4He	99.9999	—	—	—	23	4.5
			^3He	—		99.9998			
			^4He	—		0.0002			

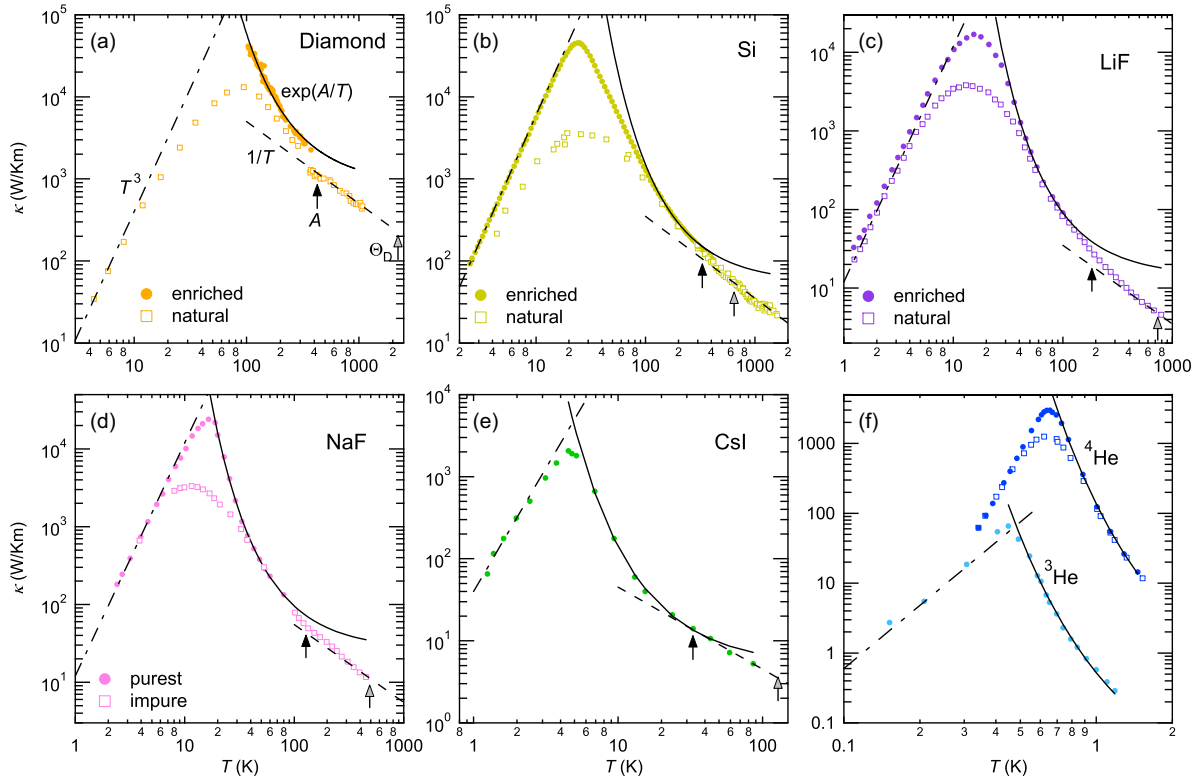


FIG. 6. Thermal conductivity of various insulators showing the Ziman transport. Temperature dependence of thermal conductivity of (a) diamond [22,32,33], (b) Si [19,34], (c) LiF [29,30], (d) NaF [16,30], (e) CsI [9], and (f) ^4He [14] and ^3He [15]. The dot-dashed, solid, and dashed lines represent the T^3 , exponential, and $1/T$ dependence, respectively. The exponential behavior is fragile against the random isotopic mixture in diamond, Si, and LiF, and impurities in NaF as shown in the panels (a)–(d). A and Debye temperature Θ_D of each material are denoted by closed and open arrows, respectively.

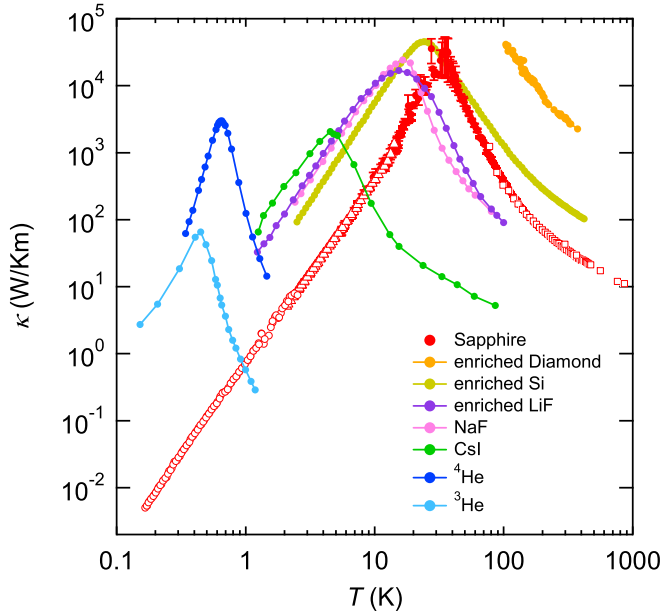


FIG. 7. Thermal conductivity of sapphire and ultra-pure insulators. A comparison of thermal conductivity with the ultrapure insulators [9,14–16,19,22,29] showing the Ziman transport behavior.

($\sim \Theta_D/8$) [35], while less striking, points towards a similar direction.

A more striking similarity between ultrapure insulators is shown in Fig. 8(b). It displays κ , normalized by its value at $T = A$ as a function of T/A . All data fall into a universal curve, which is exponential for $T/A \leq 1$ and evolves towards $1/T$, when $T/A \geq 1$. This figure indicates that in a perfect insulator with no disorder (and no soft phonons) knowing

A (which is a fraction of Θ_D) is sufficient to know thermal conductivity at any temperature above its peak value.

As seen in Figs. 6(a)–6(d), in all these high-purity insulators, the random mixture of isotopes or impurities pulls down κ . Not only the exponential variation but also the prominent peak in κ are considerably suppressed in naturally abundant diamond [32], Si [32], and LiF [29] as well as in impure NaF [16].

The case of sapphire is outstanding, because it exhibits an exponential behavior in absence of isotopic purity. As shown in Table I, the dimensionless parameter quantifying isotopic disorder, g , is exceptionally high for sapphire, compared to other insulators displaying an exponential rise. Moreover, the specific heat of our samples display a Schottky anomaly indicating the presence of magnetic impurities (Fig. S5 [18]). Despite this level of isotopic and chemical impurities in the range of 30 ppm (see Table I), our sapphire samples behave like ultrapure insulators.

The exceptional case of sapphire is emphasized by considering the amplitude of the peak thermal conductivity $\kappa_{\max}$ as a function of g . The magnitude of $\kappa_{\max}$ depends on the sample dimension d and the Debye temperature Θ_D . Increasing them would enhance $\kappa_{\max}$, by allowing the exponential increase to persist down to lower temperatures. Figure 9 is a plot of $\kappa_{\max}$ normalized by a product of $d \times \Theta_D$ as a function of g . The isotopic purification pushes up $\kappa_{\max}/d\Theta_D$ towards higher values (from the open to closed symbol in each material) following $\kappa_{\max}/d\Theta_D \propto g^{-0.3}$ as guided by the solid arrows. As seen in the figure, $\kappa_{\max}/d\Theta_D$ in sapphire stands out. Despite a g 500 times larger, sapphire surpasses its close rival ^4He .

Let us now consider a dimensionless thermal conductivity, the ratio of $\kappa_{\max}/d\Theta_D$ to $(k_B^2/\hbar)G^2$, where G is the width of Brillouin zone. As seen in Fig. 9(b), the available data

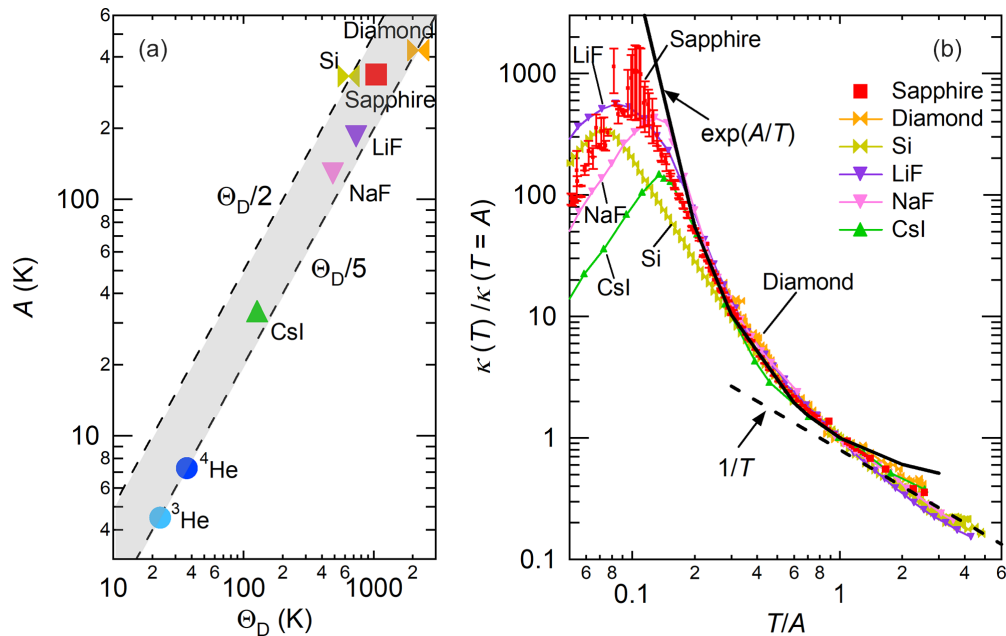


FIG. 8. A correlation between A and Θ_D , and scaling of thermal conductivity as a function of T/A . (a) Comparison of A and Θ_D in insulators showing the exponential behavior in thermal conductivity. Their ratio A/Θ_D remains between $1/5$ and $1/2$. (b) Normalized κ by those values at $T = A$ as a function of T/A .

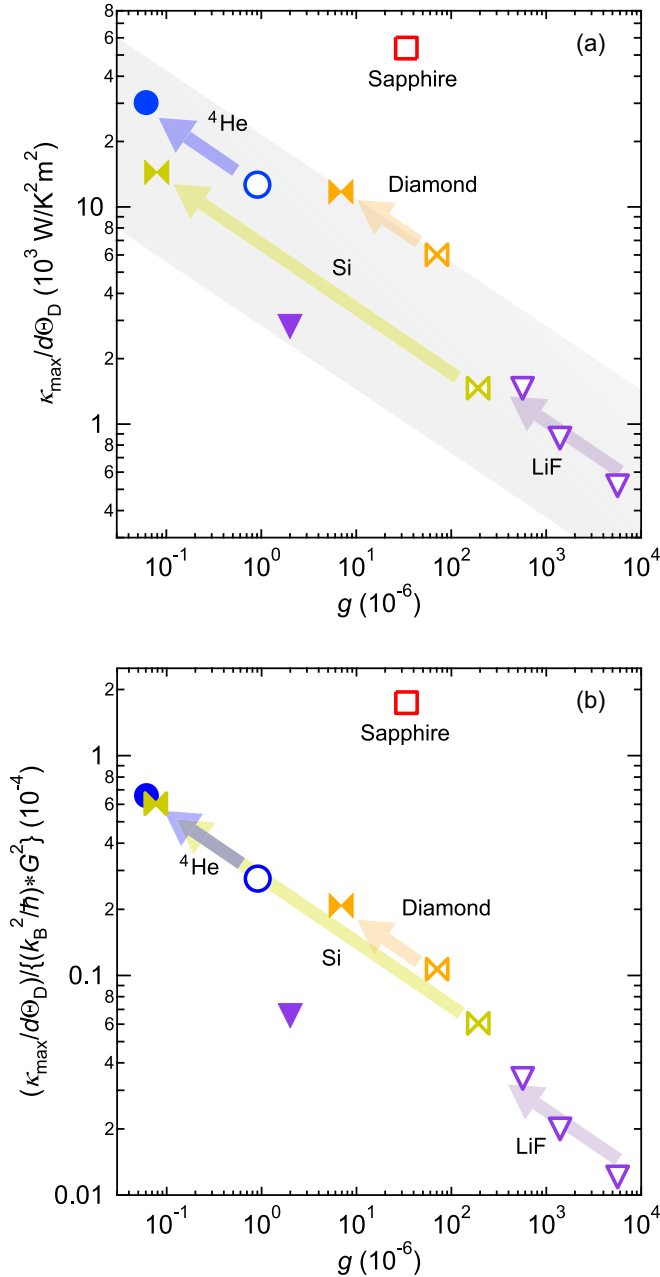


FIG. 9. Scaling of peak thermal conductivity as a function of isotope disorder parameter. (a) A plot of the maximum thermal conductivity $\kappa_{\max}$ divided by the sample dimension d and the Debye temperature Θ_D against the isotope disorder parameter g . In each system, $\kappa_{\max}/d\Theta_D$ for the isotopically pure and the natural abundant samples are presented by solid and open symbols, respectively. For LiF, the data of several variations of isotope compositions are shown by open triangles. (b) A plot of the dimensionless thermal conductivity $(\kappa_{\max}/d\Theta_D)/\{(k_B^2/\hbar)G^2\}$ vs g .

collapse onto a single line, implying that the effect of isotope scattering on the phonon heat transport across a variety of insulating solids is properly accounted by g . One can see again that sapphire does not participate in this scaling, indicating a

puzzling robustness of its high thermal conductivity against the isotopes.

Compared to other insulators discussed here, sapphire has many more atoms, N , in its primitive cell. As listed in Table 1, $N = 30$ for sapphire, $N = 8$ for diamond, Si, and LiF with fcc or rock-salt structure, and $N = 2$ for hexagonal solid ^4H . A large N attenuates mass perturbation by isotopic impurities. Moreover, it leads to an increase in the number of optical phonon branches. As shown in Fig. S6(a) [18], in sapphire the gap between the optical and the acoustic branches is small. This enhances the phase space for scattering between the acoustic and optical phonons and reduces a chance for the acoustic phonons to be scattered by the isotopes. Notably, the fact that the energy range (~ 10 THz) where the high-frequency acoustic phonons can meet the optical phonons is close to the upper bound of Ziman regime, that is A (~ 7 THz), suggests that both phonon modes indeed participate in the normal processes. Because of the proximity of optical and acoustic phonon energies, the thermal conductivity of sapphire, in contrast to other “simple” insulators, is not set by g .

We note that this scenario is compatible with theoretical calculations, suggesting that isotopic purity does not necessarily lead to higher thermal conductivity due to a modification of coupling between acoustic and optical phonons induced by isotope enrichment [36]. It has been demonstrated that isotopic purification of natural LiH by replacing ^7Li with ^6Li leads to a higher frequency of acoustic modes and a smaller phonon gap. This allows for increased coupling of the heat-carrying acoustic phonons with the optical phonons and reduction of their lifetimes. As a result, thermal conductivity of the isotopically pure ^6LiH is lower than that of the natural LiH. The possible correlation between isotope mass variations and normal collision rate emerges as a potential theme for future theoretical studies.

In this context, let us also recall the case of another highly conducting oxide, In_2O_3 , which has forty atoms per primitive cell and numerous optical phonons and its thermal conductivity can surpass that of naturally abundant silicon [37].

To sum up, naturally occurring sapphire displays the two phonon hydrodynamic regimes and a thermal conductivity much higher than what is expected, given its isotopic purity. We identify the large number of atoms per the primitive cell as a possible source of frequent momentum exchange among phonons including optical modes and attenuation of phonon-isotope scattering.

Our findings open up new possibilities for the development of high-thermal conductivity materials exploiting the exponential enhancement offered by the Ziman transport regime. It has been demonstrated that phonon hydrodynamics may find potential applications for thermal rectification [38,39] or for transmitting thermal signals [40].

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