

ENHANCEMENT OF IN-PLANE THERMAL CONDUCTIVITY OF THIN FILMS VIA SURFACE PHONON-POLARITONS

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ABSTRACT

We predict theoretically that the effective in-plane thermal conductivity of crystalline and amorphous thin films can be increased by surface phonon-polaritons significantly beyond their intrinsic bulk values. We show that the thermal conductivity due to surface phonon-polaritons increases with decreasing film thickness. In particular, for a 40 nm thick film of amorphous silicon dioxide, we calculate a total thermal conductivity of $4 \text{ W m}^{-1} \text{ K}^{-1}$ at 500 K, which is an increase of ~100% over the intrinsic phonon thermal conductivity.

Keywords: surface phonon-polariton, in-plane thermal conductivity, thin film

NOMENCLATURE

- A = area of film
- K_s = surface polariton thermal conductivity
- L_x = length of film in x -direction
- L_y = length of film in y -direction
- Q_x = heat flux in x -direction
- $Q_{x, \text{net}}$ = net heat flux in x -direction
- T = temperature
- T_H = high temperature
- T_L = low temperature
- f = distribution function
- f_o = Bose-Einstein distribution function
- g_{2D} = two dimensional density of states
- k_{sp} = complex in-plane wavevector
- k_x = wavevector in x -direction
- k_y = wavevector in y -direction
- k_{1z} = perpendicular component of wavevector in surrounding medium
- k_{2z} = perpendicular component of wavevector in film
- $k_{||}$ = real part of k_{sp}
- t = thickness of film

\bar{v}_g = group velocity

GREEK SYMBOLS

- ϵ_1 = dielectric function of surrounding medium
- ϵ_2 = dielectric function of film
- ϵ_∞ = high frequency dielectric constant
- Γ = damping constant
- \hbar = Planck's constant
- Λ = propagation length
- θ = in-plane angle from x -direction
- ω = angular frequency
- ω_{LO} = longitudinal optical frequency
- ω_{TO} = transverse optical frequency

INTRODUCTION

The ability of a material to conduct heat is phenomenologically described by its macroscopic thermal conductivity, and can be low for various reasons. Amorphous materials generally have inherently low thermal conductivities. For nanostructures such as thin films and nanowires, size effects can also greatly reduce the effective thermal conductivity due to increased interface and surface scattering of the phonons in crystalline materials [1-3]. In both of these situations, there is no established method for increasing heat conduction. We present here an approach which is potentially capable of enhancing the effective in-plane thermal conductivity of polar thin films. In particular, the in-plane thermal conductivity of amorphous, polar thin films can be increased above their bulk values. This is realized by exploiting the energy flux transported by surface polaritons. Surface polaritons are a hybrid mode that consists of the interaction of electromagnetic radiation with a free electron gas (surface plasmon-polaritons) or with transverse optical phonon

vibrations (surface phonon-polaritons). Recently, it has been shown that surface polaritons can be used to increase the radiative heat transfer between two parallel surfaces [4-6] (i.e. in the direction perpendicular to the surfaces). However, to date the thermal energy transport characteristics due to surface waves along a continuous surface have not been explored. We consider this in-plane energy flux because it is well-known that surface polaritons have long propagation lengths [7, 8], particularly on thin films [9], which in turn can lead to large effective thermal conductivities. In addition, in nanostructures, surface effects are more important than volumetric effects due to a high surface area to volume ratio. This also suggests that surface polaritons may play an important role in energy transport along films with nanoscale thickness. Specifically, we show that in amorphous silicon dioxide films tens of nanometers thick, the in-plane heat flux carried by surface phonon-polaritons can exceed the heat flux carried by phonons in the film. We also show that the effective thermal conductivity due to surface polaritons increases with decreasing film thickness, offering a method to potentially offset the reduction in thermal conductivity due to increased interface and surface scattering in crystalline thin films.

MODELING

We consider energy transport along the two surfaces of a thin film as shown in Fig. 1. The thickness of the film is t and the lengths in the x - and y -directions are L_x and L_y respectively. The lengths L_x and L_y are finite, but much longer than the mean free path, and the surface area in the x - y plane, A , is $L_x L_y$. There is an imposed temperature gradient in the x -direction from T_L to T_H , and the temperature is taken to be uniform in the y -direction. The film is assumed thin enough to neglect temperature gradients in the z -direction. The material has a frequency-dependent dielectric function given by $\epsilon_2(\omega)$ and is bounded on either side by a medium with dielectric constant ϵ_1 , which we take to be vacuum. We find that with higher dielectric constant surroundings the thermal conductivity increase still occurs, although the onset of the increase is delayed to smaller film thicknesses. The surface polariton fields on both surfaces of the film decay exponentially into the film as well as into the surroundings.

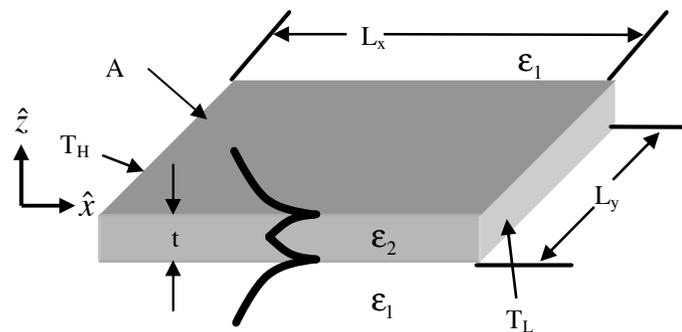


Fig. 1 A thin film of polar material with surface phonon-polaritons propagating on both sides of the thin film. A temperature gradient is imposed in the x -direction from T_L to T_H .

We calculate the contribution of the surface polaritons to the in-plane energy flux using kinetic theory. A diffusion approximation is valid when the characteristic length for heat transfer is at least several times longer than the surface wave in-plane propagation length. In the field of radiative heat transfer, this is called the Rosseland diffusion approximation [10]. The surface wave heat flux crossing a plane normal to the x -direction, per unit length in the y -direction (in units of W m^{-1}), is given by

$$Q_x = \frac{1}{A} \sum_{k_x} \sum_{k_y} \hbar \omega(k_x, k_y) f(\omega, T) \vec{v}_g \cdot \hat{x}, \quad (1)$$

where k_x and k_y are the wavevectors in the x - and y -directions respectively, \hbar is Planck's constant divided by 2π , ω is the angular frequency, f is the distribution function (to be discussed below), T is the temperature, and \vec{v}_g is the group velocity. The relationship between the frequency and the wavevectors is given by the dispersion relation of the surface polariton. In the case of phonons, for example, ω , k_x , and k_y are real quantities, and $k_x = 2\pi n/L_x$ and $k_y = 2\pi m/L_y$ where n and m take on integer values from $-\infty$ to ∞ . In the limit of $\Delta k_x, \Delta k_y \rightarrow 0$, Eq. (1) is transformed into an integral,

$$Q_x = \frac{1}{(2\pi)^2} \iint \hbar \omega(k_x, k_y) f(\omega, T) \vec{v}_g \cdot \hat{x} d^2 k, \quad (2)$$

where $d^2 k = d\theta k dk$, and θ is the in-plane angle from the x -axis.

In the case of surface polaritons, the dielectric function and the dispersion relation are complex. Thus, in solving for these dispersion relations, either the wavevector is chosen to be real, resulting in a complex frequency which gives the lifetime of a mode, or vice versa, yielding the propagation length of a mode [11, 12]. In the present paper we choose a real frequency which results in a complex in-plane wavevector, $k_{sp} = \sqrt{k_x^2 + k_y^2}$, whereby the imaginary part of k_{sp} is then associated with the mean free path as discussed further below. The real part of k_{sp} , denoted here as $k_{||}$, is used with the real frequency in calculating the energy flux, and the identification of the imaginary part of k_{sp} with the mean free path follows the approach of work done for superlattices [13] and electron energy bands [14]. The validity of this approach is further supported by the fact that the imaginary part of k_{sp} is much smaller than $k_{||}$ in the range over which the dominant contribution to the thermal conductivity integral occurs.

From the Boltzmann transport equation under the relaxation time approximation, the surface polariton distribution function under steady-state conditions is

$$f(T) = f_o(T) - \Lambda \cos\theta \frac{\partial f}{\partial x}, \quad (3)$$

since the temperature varies only in the x -direction. Here f_o is the equilibrium Bose-Einstein distribution, and Λ is the mean

free path. Substituting Eq. (3) into Eq. (2), and integrating from $-\frac{\pi}{2} < \theta < \frac{\pi}{2}$ leads to the flux in the positive x -direction, and integrating from $\frac{\pi}{2} < \theta < \frac{3\pi}{2}$ leads to the flux in the negative x -direction. The net x -direction flux is then given by

$$Q_{x,net} = -t \frac{dT}{dx} \left[\frac{1}{4\pi t} \int_0^\infty \hbar \omega \Lambda \frac{df}{dT} \Big|_{\bar{v}_g} \Big|_{k_{\parallel}} dk_{\parallel} \right]. \quad (4)$$

Since this flux is in units of power per unit length in the y -direction, the left-hand side of Eq. (4) must be divided by the film thickness to put it into a form for comparison with the standard definition of thermal conductivity. Thus, the term in brackets is defined as the effective thermal conductivity due to surface polaritons, K_s (in $W m^{-1} K^{-1}$). Equivalently, using the density of states per unit area, $g_{2D}(\omega)$,

$$K_s = \frac{1}{2t} \int_0^\infty \hbar \omega \Lambda \frac{df}{dT} \Big|_{\bar{v}_g} \Big|_{g_{2D}(\omega)} d\omega. \quad (5)$$

DISPERSION RELATIONS

It is well-known that for surface plasmons on thin films, the dispersion relation splits into an anti-symmetric mode and a symmetric mode. This splitting is more pronounced when $k_{2z}t \ll 1$ [9, 15-17], where k_{2z} is the perpendicular component of the wavevector in the film and $k_{2z}^2 + k_{sp}^2 = \epsilon_2 \omega^2 / c^2$. The same splitting also occurs for surface phonon-polaritons, and the anti-symmetric and symmetric modes (with respect to the electric field distributions) are given respectively by [16]

$$\frac{\epsilon_2}{\epsilon_1} = -\frac{k_{2z}}{k_{1z}} \tanh\left(\frac{t}{2i} k_{2z}\right) \quad (6)$$

$$\frac{\epsilon_2}{\epsilon_1} = -\frac{k_{2z}}{k_{1z}} \coth\left(\frac{t}{2i} k_{2z}\right), \quad (7)$$

where i is equal to $\sqrt{-1}$ and k_{1z} is the corresponding perpendicular component of the wavevector in medium 1. Equation (5) and Eq. (6) are transcendental equations that we solve numerically using a Newton-Raphson method to obtain the dispersion relations.

For crystalline materials, the dielectric function is well-approximated by a damped, harmonic oscillator model, $\epsilon_2(\omega) = \epsilon_\infty \left[1 + (\omega_{LO}^2 - \omega_{TO}^2) / (\omega_{TO}^2 - \omega^2 - i\omega\Gamma) \right]$, where ω_{LO} is the longitudinal optical frequency, ω_{TO} is the transverse optical frequency, ϵ_∞ is the high frequency dielectric constant, and Γ is the damping constant. However, for amorphous materials such as silicon dioxide (glass), the actual form of the dielectric function is more complicated than the simple expression used above, and consists of several overlapping resonances as shown in Fig. 2. As such, the frequency dependences of the dielectric

functions used in this paper are obtained from published experimental values of the complex index of refraction [18].

The dispersion relations for a thin film surrounded by vacuum are calculated and plotted in Fig. 3 for various film thicknesses. For reference, the dispersion relation at a single interface between a half-space of the same material and vacuum is also shown. At large values of k_{\parallel} both modes of the thin film dispersion relations approach the single interface curve asymptotically. Furthermore, as the thickness of the film increases, both the symmetric and anti-symmetric dispersion relations approach the single interface curve. This is analogous to the splitting of two degenerate energy levels connected by a coupling potential.

PROPAGATION LENGTH AND SCATTERING

The mean free path of the surface phonon-polaritons is taken to be $\Lambda = 1/2 \text{Im}\{k_{sp}\}$. The propagation lengths calculated in this manner agree well with approximate expressions found in the literature for surface polaritons on thin films [16]. In calculating the mean free path, scattering due to

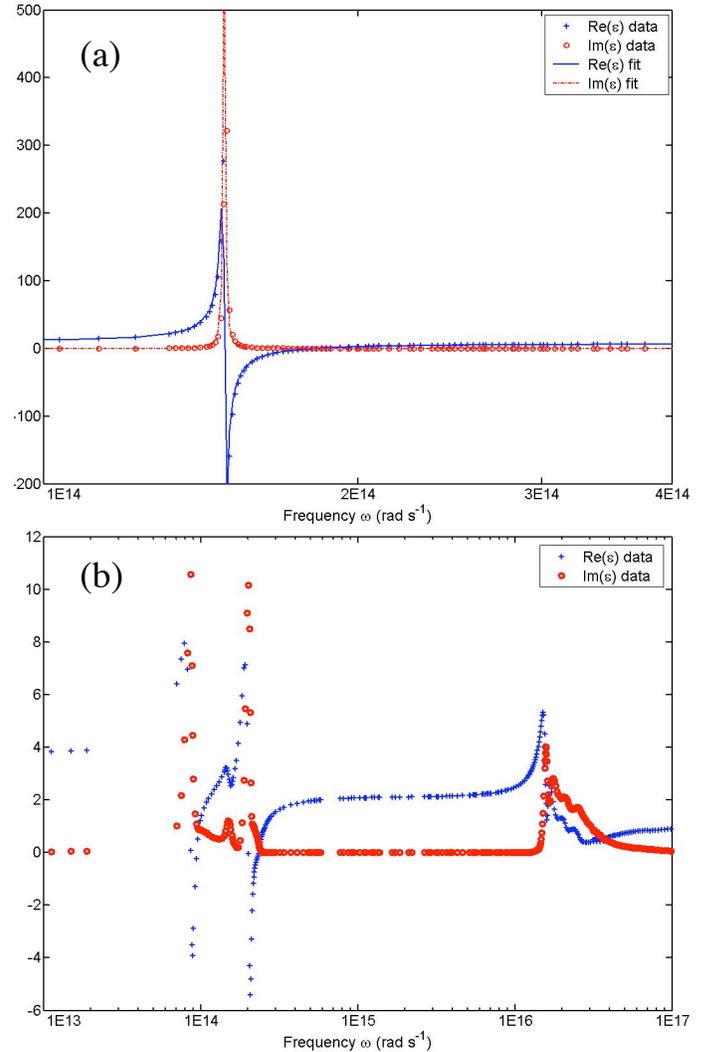


Fig. 2 Real and imaginary parts of the dielectric function of a) crystalline silicon carbide and b) amorphous silicon dioxide.

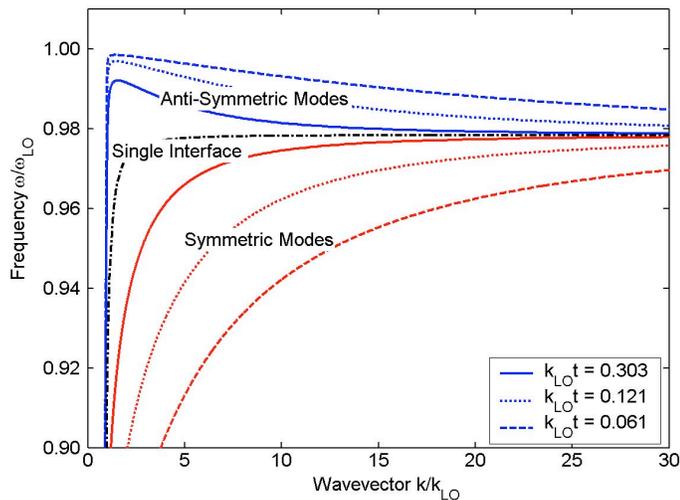


Fig. 3 Anti-symmetric mode and symmetric mode dispersion relations for surface phonon-polaritons on a thin film of silicon carbide surrounded by vacuum.

surface roughness is a potential concern. Following the work of Mills [8] the propagation lengths due to surface roughness scattering into radiative modes and into other surface phonon-polariton modes are calculated. Typical CMOS gate oxides are now less than 2 nm thick, and so a conservative value of 1 nm for the film root-mean-square surface roughness height and average distance between peaks was chosen to determine the propagation lengths due to surface roughness. The propagation lengths due to these two loss mechanisms are found to be several orders of magnitude greater than that due to the surface phonon-polariton attenuation calculated above. Furthermore, an examination of the integrand of Eq. (3) shows that the majority of the surface phonon-polariton contribution to the thermal conductivity occurs for in-plane wavelengths longer than 1 μm . Since these wavelengths are significantly larger than the surface roughness, the surface irregularities will not significantly scatter the surface phonon-polaritons [16]. Thus, the propagation length of the surface wave as determined from $\Lambda = 1/2 \text{Im}\{k_{sp}\}$ is used for the mean free path.

RESULTS AND DISCUSSION

Using Eq. (3) the thermal conductivity due to surface phonon-polaritons for various film thicknesses of amorphous silicon dioxide is calculated. The total thermal conductivity due to both bulk phonons and surface phonon-polaritons are plotted as a function of film thickness in Fig. 4. We see that at large film thicknesses, the thermal conductivity is dominated by the phonon contribution and is essentially constant. However, as the film thickness decreases below about 0.1 μm , the thermal conductivity increases dramatically due to the increasing contribution of the surface phonon-polaritons.

We note that the phonon and surface phonon-polariton contributions to the thermal conductivity follow distinctly different trends. In general, the phonon contribution decreases as the film thickness decreases because of the increasing effects of surface scattering and can be estimated using a Fuchs-Sonderheim model [19]. However, in amorphous thin films such as SiO_2 , phonons have a very short mean free path (on the

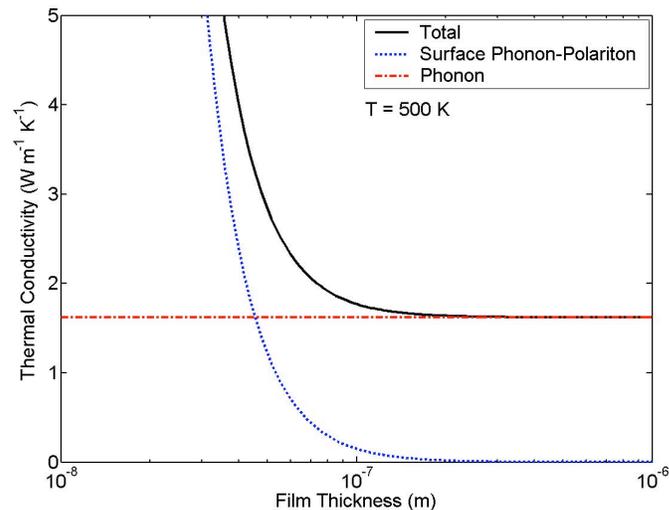


Fig. 4 Amorphous silicon dioxide (glass) thermal conductivity.

order of 3 nm) and thus their contribution to the thermal conductivity is approximately independent of the film thickness in the range considered. In contrast, the thermal conductivity contribution from the surface phonon-polaritons increases with decreasing film thickness. An examination of the heat flux per unit temperature gradient ($Q_{x,net}/(dT/dx)$) and thermal conductivity as a function of film thickness due to the surface modes alone shows two distinct regimes as shown in Fig. 5.

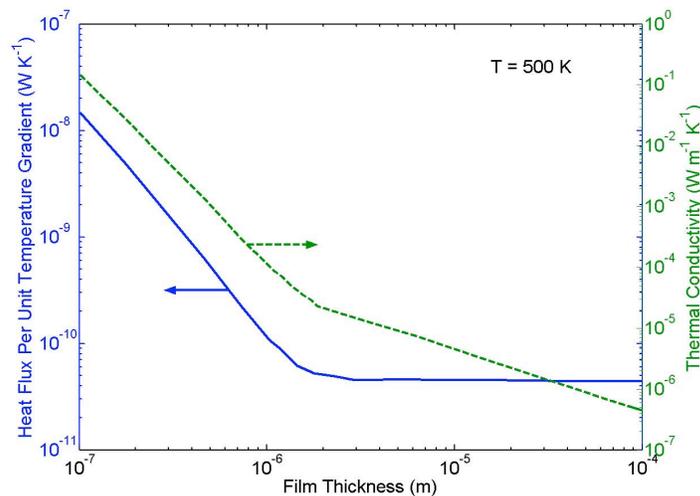


Fig. 5 Heat flux and thermal conductivity due to surface phonon-polaritons as a function of film thickness.

For film thicknesses greater than about 1 μm the heat flux is a constant, and the thermal conductivity simply scales inversely proportional to the film thickness. In this regime the surface phonon-polariton modes at the two surfaces are not coupled, and hence the total energy flux is not affected by the thickness of the film. The increase in the thermal conductivity is due solely to the geometric consideration of transporting the same amount of flux through a smaller cross-sectional area.

However, in the thickness regime below $1 \mu\text{m}$, the slope of the thermal conductivity curve is much larger. This is due to the fact that in addition to the thickness-dependent scaling, the actual amount of heat flux transported by the surface phonon-polaritons is greatly increased. In this regime, the film is thin enough that there is significant interaction between the surface phonon-polaritons on either side of the film.

A more detailed examination of the individual components that constitute the argument of the thermal conductivity integral in Eq. (4) reveals that the mean free path is the source of the increased contribution. The argument of the integral is essentially the product of the mean free path, group velocity, and density of states. These parameters are plotted for the anti-symmetric and symmetric modes of a 50 nanometer thick film of amorphous silicon dioxide in Fig. 6.

The density of states is proportional to the in-plane wavevector and is inversely proportional to the group velocity. As such, the density of states and group velocity effectively cancel each other in the thermal conductivity integral. Thus, the source of the increased surface polariton thermal conductivity is the propagation length. As can be seen from Fig. 6, the

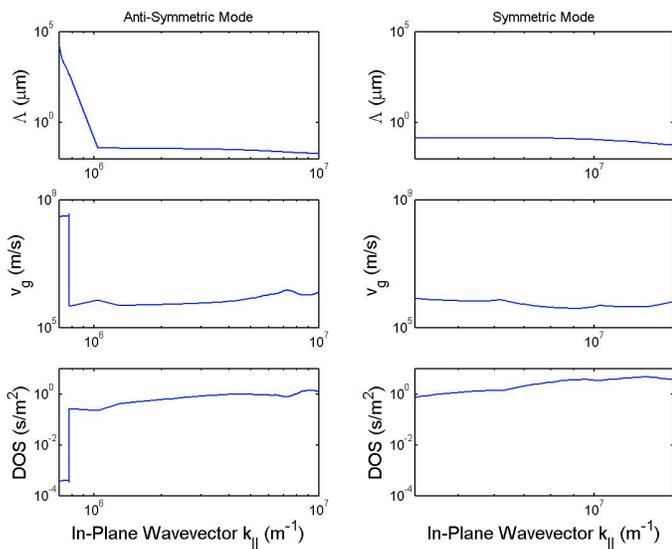


Fig. 6 Propagation length, group velocity, and density of states as a function of in-plane wavevector for a 50 nm thick film of silicon dioxide.

overwhelming majority of the contribution is due to the anti-symmetric branch at short wavevectors where the propagation length is orders of magnitude larger than at large wavevectors. As the film thickness decreases, the propagation length of the anti-symmetric branch continues to increase [11, 17] as a greater fraction of the mode propagates in the lossless surrounding dielectric [11]. This large mean free path then produces the large increase in the thermal conductivity. In particular, it is the light-line portion of the anti-symmetric branch where most of the contribution to the thermal conductivity occurs. As seen in Fig. 3, in this region the anti-symmetric mode is photon-like, with a large group velocity and large propagation length. A typical propagation length in this long wavelength region is 2 cm for a 50 nm thick silicon dioxide film.

For crystalline materials, the resonance of the dielectric function is significantly stronger as can be seen from Fig. 2. As a result, the propagation lengths are even greater than in amorphous materials. For thin films of silicon carbide tens of nanometers thick, the propagation length is calculated to be on the order of meters. However, at practical device dimensions the surface polariton propagation would thus be ballistic, and the above diffusive model would no longer be valid.

CONCLUSIONS

To conclude, we present a kinetic theory-based approach to calculate the heat flux carried by surface polaritons on nanoscale thin films. We find that for amorphous silicon dioxide films, the in-plane heat flux carried by surface phonon-polariton modes can exceed the heat flux carried by phonon modes. As the film thickness decreases, the contribution to the effective thermal conductivity due to surface phonon-polaritons increases. This phenomenon points to a novel method to increase thermal conductivity. Particularly for thin films and amorphous materials, where the phonon thermal conductivity is reduced due to size effects or is intrinsically low, this approach presents the only method, to our knowledge, to counteract that reduction in thermal conductivity and to increase the thermal conductivity of amorphous materials without changing the material structure. This approach promises to find various prospective applications in fields such as microelectronics and optoelectronics.

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