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Quantifying thermal transport in amorphous silicon using mean free path spectroscopy

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The wide application of amorphous materials in solar cells, memristors and optical sensors has stimulated the interest of understanding heat conduction in amorphous system owing to their thermal management issues. Thermal transport in amorphous materials fundamentally differs from their crystalline counterparts due to the lack of long-range order. Despite great progress in understanding the thermal transport in crystalline materials over the past few decades from both first principles computations and thermal transport characterizations, details of heat conduction in amorphous systems remain largely unknown. Here, we quantify different types of heat carriers in amorphous silicon using mean free path spectroscopy, with characteristic sizes down to 50 nm. We show that despite its disordered nature more than half of thermal conductivity is contributed by propagating vibrational waves, which have mean free paths mostly above 100 nm. This provides a direct evidence supporting the diversity of heat carriers in amorphous systems – some modes transport heat as propagating waves while others do not. Our results suggest mean free path spectroscopy as a versatile tool for understanding the thermal transport in disordered systems.

Keywords: Amorphous material, Mean free path spectroscopy, Accumulated thermal conductivity

Recent decades have witnessed wide applications of amorphous materials in microelectronic and energy conversion devices, such as solar cells[1], memristors[2], infrared sensors[3], and transistors[4]. As the thermal management is critical to the device performance, the thermal transport behavior in amorphous systems has received increasing attention recently[5–14]. In crystalline solids, the heat carriers are quantized lattice vibrations with well-defined wave vectors, known as phonons[15]. In amorphous materials, however, the absence of long-range order leads to breakdown of the phonon picture. Instead, a variety of vibrational modes are allowed that can carry heat, including modes behaving as propagating waves, known as "propagons" (similar to phonons but without well-defined wave vector)[16,17], or non-propagating modes that exhibit random vibrations, such as "diffusons" (extended in space) and "locons" (localized in space)[18]. Because the thermal transport involves such a broad range of heat carriers, quantifying the contributions from different heat carriers has become a central topic in understanding the materials' heat conduction behavior[19–21].

Spectroscopy methods such as neutron or Raman scattering have been widely used to categorize the vibrational modes in crystalline and molecular materials. For amorphous systems, the lack of long-range order allows the scattering process to occur from almost all the vibrational modes and consequently spectroscopy tools in general reflect the vibrational density of states but cannot distinguish their difference for thermal transport[22–24]. Alternatively, transport measurements on amorphous samples have been conducted to investigate the size effect in thermal conductivity, using classical electrical resistance thermometer[25,26], 3ω method[7,27], thermo-reflectance[8,28], or thermal-bridge method[9]. Size effect occurs when modes with long propagating lengths experience strong scatterings by sample boundaries. While these experiments have shown strong size effect and suggested propagating modes may contribute significantly to the total thermal conductivity[7–9,28], the methods are indirect as the thermal transport is inferred mostly based on samples prepared with different thicknesses, subject to the uncertainty due to sample variations. The lack of an experimental tool to quantify the contribution of propagating and non-propagating modes in a single sample has impeded efforts to understand the thermal transport in amorphous material-based nanostructures, where the sample-specific structures may have a large impact on their transport properties[7].

The distinction between propagating and non-propagating modes is given by the Ioffe-Regel criterion[18], which states that the propagating length for propagons is larger than their quantum wavelength and thus mean free paths (MFP) are well-defined, while for non-propagating modes MFPs become smaller than their wavelength and are no longer meaningful. Consequently, the mean free path spectroscopy[19], recently developed for understanding nanoscale thermal transport in crystalline materials, presents an ideal tool to quantify different types of heat carriers in an amorphous material. For crystalline materials, Dames and Chen[29,30] have introduced the cumulative thermal conductivity to describe the broad phonon

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spectrum based on their mean free paths,

$$F(\Lambda_{\rm m}) = \int_0^{\Lambda_{\rm m}} \frac{1}{3} C \cdot v \cdot \Lambda \cdot (-\frac{{\rm d}\Lambda}{{\rm d}\omega})^{-1} {\rm d}\Lambda \qquad (1)$$

where C, v and Λ are the phonon mode-specific heat capacity, group velocity and mean free path. Recent developments in first principles computations and mean free path spectroscopy techniques have allowed quantification of the cumulative thermal conductivity and thus the broad mean free path spectrum in crystalline material systems including alloys with certain disorder[19–21,31–33]. In contrast, the mean free path spectrum for the propagating modes in disordered systems, and their relative contribution compared to non-propagating modes, remain largely unknown. In this work we present a comparative study of the thermal transport in amorphous silicon (a-Si) and crystalline silicon (c-Si) with mean free path spectroscopy, by probing the quasi-ballistic thermal transport with varying heater sizes using time-domain thermo-reflectance method. Aluminum metal grating with line widths down to 50 nm allows us to extract the mean free path spectrum of phonons (propagons in the case of a-Si). Furthermore, we provide a quantitative measure of the relative contribution to heat conduction in a-Si from different heat carriers, with more than half contribution to the total thermal conductivity coming from the propagating modes.

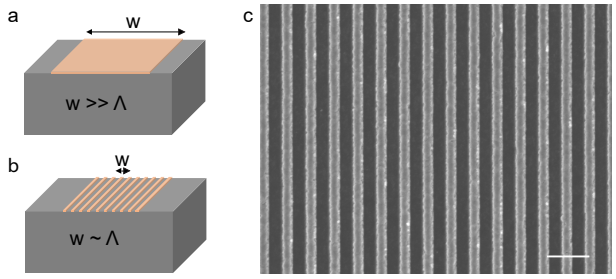

FIG. 1. Illustration of diffusive and quasi-ballistic phonon transport. (a) In diffusive limit, $w >> \Lambda$, phonons experience enough scattering and reach thermal equilibrium. Measured thermal conductivity corresponds to the bulk thermal conductivity of the substrate material. (b) As the characteristic length w is smaller than Λ, phonons propagate ballistically from the heater to the substrate. (c) Scanning electron microscopy (SEM) image of a typical aluminum grating on a-Si sample substrate. Scale bar is 500 nm.

Quasi-ballistic heat conduction occurs when the characteristic lengths of heat sources are comparable to the mean free paths of propagating heat carriers[34]. By varying this characteristic length, one can thus probe the mean free path spectrum of propagating modes contributing to heat conduction[19,33,35]. In our experiments, one-dimensional metal gratings with width w are used as nanoscale heaters, as illustrated in Fig. 1. Fig. 1a shows the diffusive limit ($w >> \Lambda$) scenario, where heat carriers experience strong scatterings and reach thermal equilibrium. In this case the thermal decay from the heat sources can be well described by a diffusion equation with the bulk thermal conductivity $\kappa_{\rm bulk}$. When the size of the heater is comparable to or smaller than Λ, part of the heat carriers will propagate ballistically from the heater to the underlying substrate. By gradually reducing the characteristic length w of heaters (Fig. 1b), the measured thermal conductivity thus represents the contribution to the thermal transport from different portions of the entire spectrum of heat carriers[19,33,35].

The samples include an a-Si film (~ 1.2 μm thickness) and a crystalline silicon. The a-Si film is deposited by low pressure chemical vapor deposition (LPCVD) on a crystal silicon substrate. First, the sample is loaded vertically with its face perpendicular to the axis of the chamber tube. The silane (SiH_4) gas is then turned on to flow into the chamber at a constant flow rate of 30 sccm. The deposition conditions are maintained at 550 ^{o}C in temperature and 250 mTorr in pressure. The deposition rate is approximately 5 nm/min.

It is important to characterize the structure of amorphous materials when comparing between different studies because the degree of disorder could be different depending on the sample preparation. To this end, we have used Raman spectroscopy measurement to characterize the structure of the LPCVD grown a-Si. Compared to the c-Si which has a sharp Raman peak around 520 cm^{-1} coming from its optical phonon mode, a much broader peak is found in a-Si around 480 cmcm^{-1} (Fig. S3a). Following Beeman et al.[36], we define the line width Γ of this broad peak as twice the half width on the high-frequency side at half the maximum height, which gives $\Gamma = 65$ cm^{-1}. It has been understood that this line broadening effect comes as a result of the local disorder of Si-Si bond that deviates from the ideal tetrahedral configuration. One quantity that describes this disorder is the root-mean-square bond-angle distortion $\Delta\theta_b$ (zero in the case of crystalline silicon). Based on the Beeman-Tsu-Thorpe model, our measured line width corresponds to a root-mean-square bond-angle distortion of $\Delta\theta_b \approx 8.3^o$. This value is among the low-end of the bond-angle distortions[36]. Furthermore, Raman spectrum around 2000 cm^{-1} shows that the sample does not have any peak (Fig. S3b). It is known that if the sample is hydrogenated, Si-H bonds would lead to Raman peaks in this spectral range[37]. These results signature that the a-Si sample is not hydrogenated and has a low degree of bond-angle disorder.

After obtaining the a-Si film together with the crystalline silicon as the reference material, we then fabricate metal grating lines onto both of these samples. The nanofabrication procedure is based on a metal lift-off method. Details of the fabrication procedure are provided in the Supplemental Information (SI)[38]. Fig. 1c shows typical scanning electron microscopy (SEM) image of the fabricated metal gratings. Aluminum grating width varies from 400 nm to 50 nm, and the spacing between adjacent lines is fixed at 150 nm.

The thermal conductivity of samples was measured using a polarized two-tint time domain thermo-reflectance (TDTR) set-up (see ref. 21 for more details). In TDTR method[39–41], a pump laser first heats up the sample, and as the heat decays into the substrate a second probe beam is reflected from the sample surface meanwhile the signal is recorded as a function of the delay time. As the reflectance changes with temperature, the signal thus monitors the surface temperature decay, from which the substrate thermal conductivity can be extracted. It is worth noting that the thermal decay as characterized by the peak temperature of the heater is different from that in the transient grating experiments (by either crossing of two beams or diffraction from metallic gratings) which measure the temperature difference between the peak and valley of the grating[35]. A metal layer is typically deposited onto the sample to avoid direct heating of the sample and for larger thermo-reflectance response. Aluminum was chosen due to its high thermo-reflectance coefficient at our probe laser wavelength (∼785 nm)[42].

For TDTR measurements on samples with grating structure, it is necessary that most of the laser energy does not transmit through the metal layer and generates electron-hole pairs that influence the signal. First, a polarizer is added with its polarization direction in parallel with the grating lines, which enhances the absorption of the pump and probe by the grating structure so that the transmission is reduced. Furthermore, we have adopted a two-tint configuration, where long-pass and short-pass filters confine the pump and probe wavelengths to 780 nm and 790 nm, respectively. As the spacing (the gap between adjacent lines) is fixed at 150 nm, the pump and probe beam with wavelength at ∼785 nm will have minimal transmission due to their larger wavelength compared to the spacing.

The optical simulation for aluminum metal grating on c-Si and a-Si shows that less than 10% of the total energy of incoming light will transmit through the metal grating. We have also experimentally verified the insignificant laser transmittance by varying the polarization angle relative to the grating lines (details of modeling and experiment can be found in Section S3 in the SI). The polarizer together with the two-tint configuration thus avoids direct substrate heating from the laser, as have been verified by Zeng et al. in our previous work[21]. Furthermore, based on the simulated optical absorption and ignoring the metal layer, we estimate the steady state temperature rise for the pump and probe beam to be at most 26 and 8 degrees respectively (SI, section S4). While there is a modest temperature rise in our system, we note that the thermal properties of the measured samples (c-Si and a-Si) have small changes within such temperature range[5].

We measured effective thermal conductivity of amorphous silicon with various characteristic length w (metal line width) at room temperature as shown in Fig. 2. Fig. 2a shows plots of phase versus delay time for a-Si with several representative grating widths. The data can be fitted using an effective thermal conductivity based on a diffusive transport model. The interface is treated as a thermal resistance and included in our fitting model. This treatment is acceptable in our system for the following reasons. First, while for metal/non-metal interface studies have proposed electron-phonon coupling across the interface as an additional heat transfer channel[43–45], from simulation it is found that for Al/c-Si interface phonon-phonon coupling is dominant and the spatial span of non-equilibrium region near the interface is usually small[46]. Moreover, compared to phonons in c-Si, vibrational modes in a-Si at similar frequency level have much smaller propagating lengths[13]. These suggest that the spatial span of the non-equilibrium region near the Al/a-Si interface is small such that the thermal transport across the interface can be treated as an effective interfacial resistance. By fitting the thermoreflectance data we extract the interfacial thermal resistance and effective thermal conductivity at different grating widths. The fitted thermal interface conductance shows no apparent size dependence (SI, Fig. S7), consistent with previous experiments[19,21] as well as heat transport modeling[47] that explicitly includes the phonon transmittance across the interface. From the variation of the effective thermal conductivity with the grating width we then infer the non-diffusive thermal transport from the nanoscale heaters to the sample substrate. Fig. 2b shows the fitted effective thermal conductivity of silicon with varying characteristic lengths averaged between two modulation frequencies. No modulation frequency dependence was observed on the samples (Fig. S6). The measured bulk thermal conductivity of a-Si is about 2.2 W/mK, which is in line with previously published experiments[7–9,28].

Over past few decades, the thermal transport property of a-Si has been studied theoretically in detail based on lattice dynamics or molecular dynamics simulations[10,11,13,14,48,49]. The theory predicts the existence of propagating modes (propagons) with long MFP in a-Si. In Fig. 2b, a-Si shows strong size-dependent thermal conductivity. This result is consistent with previous measurements on a-Si samples with different thicknesses[7–9]. One feature of our measurement is that there is still finite contribution to the total thermal conductivity even at the smallest metal line width, indicating there are heat carriers that do not experience much from size effect. These contributions are believed to come from non-propagating modes. However, the boundary between the propagating and non-propagating modes has been under debate. Past studies of thermal conductivity in amorphous silicon thin films (in the cross-plane direction, same as ours) seem to suggest that the thermal conductivity does not decrease further for film thicknesses below 100 nm[8,9]. However, most of these measurements are subject to large uncertainty due to the presence of interfacial resistance and sample-to-sample variations, and therefore the differences below 100 nm are not well resolved.

On the other hand, recent simulations suggest that

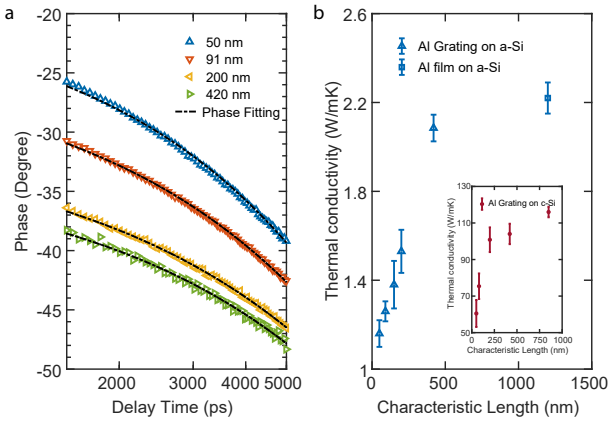

FIG. 2. TDTR experiments reveal a characteristic length-dependent thermal conductivity in a-Si. (a) Measured phase versus delay time for a-Si with aluminum grating and the fitting result using an effective thermal conductivity based on a diffusive transport model. (b) Effective thermal conductivity changes with respect to the characteristic length w in a-Si. For a-Si with aluminum grating the characteristic length is the metal line width, while for aluminum film this length represents the thickness of the a-Si sample. Inset shows measured thermal conductivity of crystalline silicon sample. The uncertainty is calculated based on the standard deviation of the measurement results.

propagating modes may exist with mean free paths well below 100 nm[13,49]. Although the behavior of those extended non-propagating modes may be altered by the periodic boundary condition in the smaller computational cell, these results nonetheless suggest that the boundary between propagating and non-propagating modes may occur at a much smaller length scale. Our measurement supports this picture by showing that the thermal conductivity does not plateau at 100 nm but slightly decreases with decreasing line widths. The previous observation of the seemingly saturating thermal conductivity can be understood because a major portion of the propagating modes still come from those with mean free paths larger than 100 nm, as seen by the large decrease of thermal conductivity from larger line widths down to around 100 nm. Moreover, the large uncertainty in previous reports may also mask the differences in thermal conductivity below 100 nm, as non-propagating modes start to dominate the heat conduction. The recent study by Kwon et al. has reached similar conclusion by studying thermal conductivity in amorphous silicon nanotubes with different shell thicknesses but in the axial direction[9].

The characteristic length-dependent thermal conductivity allows us to reconstruct the cumulative thermal conductivity, which provides a quantitative description of the mean free path spectrum. For this we have also measured the single crystalline silicon as a benchmark material so that a comparative study can be performed. Fig. 3a shows the normalized thermal conductivity as a function of metal line widths for both c-Si and a-Si. Below we use the same reconstruction scheme for both samples, effectively treating propagons in amorphous silicon the same as phonons.

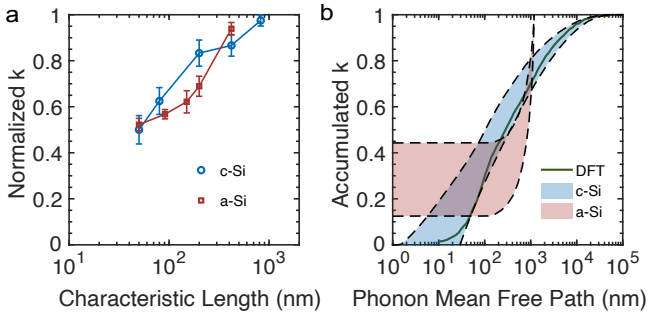

FIG. 3. Thermal conductivities of c-Si and a-Si. (a) Normalized thermal conductivity (to their bulk value) with respect to the characteristic length. (b) Cumulative thermal conductivity. The olive green line represents the density function theory calculation result[32].

The reduction of thermal conductivity with decreasing heater sizes can be described by a suppression function S, which in our case of metal grating heaters depends on both the ratio between mean free path and linewidth, and the filling fraction[21]:

$$\kappa = \int_0^\infty S(\frac{\Lambda}{w}, \frac{w}{L}) \cdot f_p(\Lambda) \cdot \mathrm{d}\Lambda + \kappa_{np} \qquad (2)$$

where f_p is the differential thermal conductivity at given mean free path from propagating modes and κ_{np} is the contribution from non-propagating modes. The suppression function quantifies how the contribution to heat flux from heat carriers with certain MFP are suppressed with respect to the Fourier's law prediction. The key assumption is that the suppression function only depends on the experimental heating geometry, while the material properties influence the measurement through the MFP distribution f_p. In such case, the same suppression function can be found in a model "gray-body" material and calculated by solving the gray-body BTE[47]. Briefly, we solve the transient heat diffusion in the corresponding heating geometry using the phonon Boltzmann transport equation assuming all phonons have the same mean free paths (gray-body assumption). Here the gray-body assumption is used only for obtaining the suppression function at different length scales. The MFP reconstruction from the experimental data to be shown later still considers the entire MFP distribution. For the simulation, an initial temperature rise is applied at the metal grating, and drives the heat flow into the substrate. The free boundaries are modeled assuming specular reflection, and the boundary condition treatment (diffusive or specular) is found to have minimal impact on the simulation results[47]. For given grating width and filling fraction, the surface temperature rise is monitored and fitted with Fourier's diffusion equation to yield an effective thermal conductivity. By varying the grating width and filling

fraction, suppression function with respect to the two non-dimensional numbers ($\frac{\Lambda}{w}$ and $\frac{w}{L}$) can be obtained.

With the suppression function, Eq. 2 can then be rewritten to infer the cumulative thermal conductivity by taking the derivative of the suppression function, denoted as kernel function K (see derivation in SI)

$$\kappa = S^{\infty} + \int_0^{\infty} K(\frac{\Lambda}{w}, \frac{w}{L}) \cdot \frac{1}{L} \cdot F(\Lambda) \cdot \mathrm{d}\Lambda \qquad (3)$$

where S^{∞} is the value of the suppression function at infinitely large mean free path, which arises from the overlapping of ballistic transport between adjacent grating lines[47], and F, the cumulative thermal conductivity, takes into account the contributions from both propagating and non-propagating modes. The suppression and Kernel functions in general depend on the heating geometry and can be computed separately using Monte Carlo methods[47].

To reconstruct the cumulative thermal conductivity $F(\Lambda)$, we need to solve the inverse problem given by Eq. 3. Such inverse problem often does not have unique solution, and Minnich proposed an optimization procedure to reconstruct the function F with certain constraints[50]. For amorphous materials, our a priori assumptions for F are (1) the cumulative curve goes from a positive finite value to one, namely $F(0) \geq 0$ and $F(\infty) = 1$; (2) F should be monotonically increasing; (3) F is smooth. The first condition is different from previous work in that we do not require F to start from zero. This is because in amorphous materials there are non-propagating modes which do not experience appreciable size effect. Therefore there could be significant contribution to the thermal conductivity even at the smallest heater size. The last condition comes from the observation that most cumulative thermal conductivities are averaged over a broad phonon spectrum and seldom have sharp features. We further note that because our a-Si film thickness is 1200 nm, boundary scatterings limit the maximum mean free path of propagating modes in a-Si to approximately 1200 nm, and for a-Si we only reconstruct F up to this length scale.

Fig. 3b shows the reconstructed cumulative thermal conductivity for crystalline and amorphous silicon with a credible interval constructed based on the uncertainty in the measurements (SI, mean free path reconstruction). The reconstructed phonon MFP spectrum for crystalline silicon is consistent with previous first-principle calculations which show that phonon MFPs in c-Si span a broad range from 10 nm to a few μm[32,51]. In comparison, while strong size effect has also been seen in a-Si, the propagating modes that are subject to size effect have mean free paths mostly above 100 nm. This result indicates that while propagating modes with MFPs smaller than 100 nm exist, their contribution to the total thermal conductivity is small compared to those with longer MFPs, which is in agreement with previous work[8,9]. The reconstructed cumulative thermal conductivity has a larger uncertainty than the measured thermal conductivity values due to the non-uniqueness of solving the inverse problem. Nevertheless, the reconstructed cumulative curve shows that propagons contribute significantly (about 70%) to the total thermal conductivity in our a-Si sample. Due to the finite sample size, propagating modes experience strong boundary scatterings. One expects that in an intrinsic amorphous silicon sample without boundaries the relative contribution from propagating modes could be even larger. In comparison, the non-propagating modes contribute to the rest about 30% of the thermal transport. Because the non-propagating modes do not have well-defined mean free paths, they diffuse on a length scale comparable to their wavelength. Correspondingly their contribution to the thermal transport will be minimally affected by the shrinking heater size, as shown by the plateau in the cumulative curve at small mean free path limit. We further note that while a-Si and c-Si have comparable covalent bonds, the mean free path spectrum of propagons in a-Si does not necessarily overlap with that of phonons in c-Si due to the different scattering mechanisms. As pointed out by Moon et al., propagating modes in amorphous silicon may also experience scatterings from the fluctuations of the elastic modulus, which alters their transport behavior compared to phonons in c-Si[13].

In this work, the time-domain thermo-reflectance (TDTR) technique with varying heater size is used to study the thermal transport in crystalline and amorphous silicon. By reconstructing the mean free path cumulative thermal conductivity, it is shown that different types of heat carriers – both propagating and non-propagating – can contribute to the thermal transport in amorphous system, distinct from the heat conduction in crystalline materials where heat is mainly carried by propagating phonons. We directly quantify the contributions from these different heat carriers, with propagating modes, known as propagons, contributing to more than half of the total thermal conductivity and have relatively long mean free paths. This result supports the existence of long mean free path modes in disordered materials. The rest of thermal transport is contributed by non-propagating modes that do not experience strong size effect. With the ability to fabricate nanostructures with further reduced dimensions, the mean free path spectroscopy can be potentially applied to other disordered materials for studying their thermal transport and the contributions from different vibrational modes.

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