Ballistic Phonon Penetration Depth in Amorphous Silicon Dioxide

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ABSTRACT: Thermal transport in amorphous silicon dioxide (a-SiO₂) is traditionally treated as random walks of vibrations owing to its greatly disordered structure, which results in a mean free path (MFP) approximately the same as the interatomic distance. However, this picture has been debated constantly and in view of the ubiquitous existence of thin a-SiO₂ layers in nanoelectronic devices, it is imperative to better understand this issue for precise thermal management of electronic devices. Different from the commonly used cross-plane measurement approaches, here we report on a study that explores the in-plane thermal conductivity of double silicon nanoribbons with a layer of a-SiO₂ sandwiched in-between. Through comparing the thermal conductivity of the double ribbon samples with that of corresponding single ribbons, we show that thermal phonons can ballistically penetrate through a-SiO₂ of up to 5 nm thick even at room temperature. Comprehensive examination of double ribbon samples with various oxide layer thicknesses and van der Waals bonding strengths allows for extraction of the average ballistic phonon penetration depth in a-SiO₂. With solid experimental data demonstrating ballistic phonon transport through a-SiO₂, this work should provide important insight into thermal management of electronic devices.

KEYWORDS: Thermal conductivity, silicon nanoribbon, van der Waals interface, amorphous silicon dioxide, ballistic phonon transport

Amorphous silicon dioxide (a-SiO₂) has been an indispensable component as dielectric materials in silicon-based microelectronic devices. The low thermal conductivity of a-SiO₂ and the thermal boundary resistance at the interfaces between SiO₂ and other thin film layers are of major concerns in device thermal management. In fact, aggressive miniaturization of microelectronic devices has reduced the thickness of gate oxide down to a mere 1.5 nm in metal-oxide-semiconductor field-effect transistors (MOSFETs). As such, thermal transport through thin a-SiO₂ layers is of fundamental importance for cooling of microelectronic devices, yet the detailed transport mechanisms remain an issue of debate.

Classically, according to the amorphous limit model proposed by Einstein, heat conduction in greatly disordered materials, such as a-SiO₂, is treated as random walks of independent vibrators, which corresponds to a mean free path (MFP) approximately the same as the interatomic distance (i.e., < 1 nm). This physical picture has remained challenged and reaffirmed over the past decades in the effort to develop a modern understanding of thermal transport through a-SiO₂.

For example, measurements of the effective cross-plane thermal conductivities of thin a-SiO₂ layers indicated a thickness dependence up to 10 nm at room temperature, which was attributed to contributions from long wavelength phonons that could transport ballistically through the a-SiO₂ films and experience boundary scattering. However, later studies argued that the observed thickness dependence could be due to the thermal boundary resistance between the a-SiO₂ layer and its substrate but not the reduced phonon MFP in a-SiO₂ layers from boundary scattering.

More recently, efforts have been made to distinguish the contributions of different vibrational modes (i.e., propagons, diffusions, and locons) to thermal transport in a-SiO₂. Both experiments and numerical modeling have been conducted, and the results indicate that thermal transport in a-SiO₂ is dominated by diffusions while propagons only make a marginal contribution. For example, using time domain thermoreflectance (TDTR) technique, the measured cross-plane thermal conductivity of a 3 nm thick a-SiO₂ film remains bulklike at room temperature, suggesting negligible contribution from propagons. Meanwhile, based on molecular dynamics (MD) simulations, it has been argued that diffusions are the dominant energy carriers in a-SiO₂ with less than 6% contribution from propagons to its thermal conductivity.
One subtle point that has not been explicitly discussed is the potential difference between energy transport in bulk a-SiO$_2$ and through a-SiO$_2$ thin films in contact with crystalline materials; while the latter is in fact more relevant to cooling of microelectronic devices. Even though the concept of thermal phonons might not be appropriate for describing vibrational modes in bulk a-SiO$_2$, phonons from crystalline materials can propagate through a-SiO$_2$ films in contact with them. This difference, however, is not important if the penetration depth of thermal phonons in a-SiO$_2$ is approximately the same as the interatomic distance. We note that phonon transmission through Si/a-SiO$_2$/Si sandwiched structures has been studied using wave packet simulations; the results suggested that a large portion of phonons with frequencies up to 1.4 THz, which contribute more than 20% to the thermal conductivity of silicon, could still ballistically penetrate through a 10 nm a-SiO$_2$ layer. However, no convincing experimental demonstration of this prediction has been reported.

To date, nearly all measurements of thermal transport through composite interfaces have been carried out in the cross-plane direction, using either the three-omega method or TDTR. One challenge associated with the cross-plane approach is the dual effects of phonon-boundary scattering, which leads to both reduced effective phonon MFP and thermal boundary resistance; decoupling these two factors at a nanometer scale is tricky. Therefore, it could be difficult to draw a solid conclusion on the ballistic phonon penetration depth through a-SiO$_2$.

Measurement of in-plane thermal conductivity, on the other hand, could provide valuable information about ballistic phonon transmission through interfaces. For example, comparison of the in-plane thermal conductivities of single and double boron nanoribbons stuck together via van der Waals (vdW) interactions helped to determine the percentage of phonons that could ballistically pass through the vdW interface. In the regime where intrinsic phonon MFP is much larger than the nanoribbon thickness, if phonons can ballistically transmit through the interface between two ribbons the effective phonon MFP in the double ribbon will be extended, leading to an enhanced thermal conductivity for the double ribbon. Therefore, comparison of the in-plane thermal conductivities of single and double silicon nanoribbons with a layer of a-SiO$_2$ on their surfaces could help to determine the ballistic penetration depth of phonons through a-SiO$_2$.

In this paper, we show that the thermal conductivity of double silicon nanoribbons with a layer of a-SiO$_2$ sandwiched in between can indeed be higher than that of corresponding single ribbons, indicating that thermal phonons can ballistically transmit through composite vdW interfaces with a thin a-SiO$_2$ layer. For an annealed double ribbon sample with high adhesion energy between the two ribbons and an oxide thickness of ∼5.1 nm, we observed ∼11% thermal conductivity enhancement at 300 K, indicating that thermal phonons can ballistically penetrate through an a-SiO$_2$ layer of more than 5 nm.

For this study, the silicon nanoribbons were fabricated from 6" (150 mm) SOI wafers (Simgui Technology Co., Ltd.). To prepare double ribbon bundles, we used a sharp probe to pick up one ribbon and stacked it on top of another one sitting on a polystyrene substrate. Through careful alignment, the two ribbons can overlap with each other almost perfectly (Section I in the Supporting Information). The resulting double ribbon was then transferred to a microdevice for subsequent thermal measurement.

Figure 1a shows a scanning electron microscopy (SEM) micrograph of a double ribbon sample after it was transferred to the measurement device and placed between the two suspended membranes with integrated resistance heaters/thermometers. Figure 1b presents an atomic force microscopy (AFM, Bruker Dimension Icon) image of the denoted section in Figure 1a, which clearly demonstrates the different heights of the overlapped and nonoverlapped ribbon segments, confirming nearly complete overlapping of the two ribbons. On the basis of the AFM and SEM measurements, the dimensions of the silicon core in each ribbon are 18 ± 1 nm thick and 140 ± 2 nm wide after subtracting a 2.5 nm thick a-SiO$_2$ layer on each surface.

To investigate how the a-SiO$_2$ layer affects phonon transport in the Si/a-SiO$_2$/Si sandwiched structure, we prepared multiple double ribbon samples with different a-SiO$_2$ thicknesses at the interface. To achieve this, a well-aligned double ribbon bundle was first transferred from the PDMS substrate to a Petri-dish (polystyrene, Fisher Scientific). The sample was then immersed in a drop of buffered oxide etchant (BOE, 6.38% HF, Sigma-
Aldrich) for 2 min, after which the double ribbon sample was rinsed with reagent alcohol (Fisher Scientific). To minimize the oxidization effects on the HF-etched double ribbons after being transferred to the measurement device, it was either stored in a high vacuum chamber (\(<10^{-6}\) mbar) or immediately loaded into the cryostat for thermal measurement. On the other hand, to achieve thicker interfacial oxide layers, ribbon bundles were left in the ambient environment (AE) for various durations.

For the thermal measurements, we first tested a double ribbon sample with native oxide on the surface (DN#1), that is, a sample prepared without HF treatment. As shown by the HRTEM image in Figure 2a, the interfacial a-SiO₂ layer is about 5.1 nm thick (see Section III in the Supporting Information for details of TEM characterization). The thermal conductivity of all measured ribbon samples is extracted based on the dimension of the silicon core with the nominal contribution of the SiO₂ layers to the measured thermal conductance subtracted. The obtained data (Figure 3a) indicate that the thermal conductivity of this double ribbon is approximately the same as that of a single ribbon, which has the same cross-sectional dimensions as each individual ribbon in the double ribbon sample. This is very different from the results for boron nanoribbons, where the thermal conductivity of double ribbon bundles is \(\sim45\%\) to \(\sim70\%\) higher than that of a single ribbon as temperature drops from 300 to 50 K.²⁴ There are two possible reasons for this difference. One is the different interface morphology, which includes two aspects. (1) The interfacial adhesion energy of DN#1 could be low as no wetting process was involved in the sample preparation process; and (2) the thicker and denser oxide layer between the two silicon nanoribbons could diffusely scatter most phonons so a negligible amount of thermal phonons could penetrate through ballistically. The other possibility is that, because silicon has a much lower Debye temperature (\(\sim645\) K) than boron (\(\sim2025\) K),²⁴ phonons of the same energy have much shorter wavelengths in silicon and are scattered more strongly at the vdW interface between the two ribbons.

Figure 2. (a) An HRTEM micrograph of a double ribbon sample without BOE etching (DN#1), and the interfacial a-SiO₂ layer is measured as 5.1 nm thick. (b) An HRTEM image of a double ribbon after BOE etching and with minimum oxygen exposure (DN#4), where the interfacial a-SiO₂ layer is thinned to 1.7 nm. The insets show diffraction patterns recorded, indicating all ribbons were patterned along the [110] direction.

Figure 3. (a) Measured thermal conductivity of double Si nanoribbon and single ribbon without BOE etching, as well as a nonetched double ribbon annealed at 450 K. The legend indicates that the silicon core is 140 nm wide and 18 nm thick after subtracting the 2.5 nm thick a-SiO₂ layer on each surface. The inset depicts the formation of covalent siloxane bonds upon high-temperature annealing. (b) Measured thermal conductivity of BOE etched double Si nanoribbon samples with various interfacial a-SiO₂ thickness and interfacial adhesion energy. Along with the double ribbons are the measured thermal conductivity of a BOE etched single ribbon as well as a single nanoribbon of double thickness. AE stands for ambient environment, and DN#7 and 8 are double ribbons stored in ambient for 4 and 60 days after BOE etching.
423 K (150 °C), hydrogen bonds will convert into covalent siloxane bonds as depicted by the inset in Figure 3a, which leads to an interfacial bonding energy of up to 1200 mJ/m².30

As shown in Figure 3a, this treatment boosted the thermal conductivity of sample DN#2 to a value significantly higher than that of DN#1 (11% higher at 300 K). Here, the enhancement is defined as \( (\kappa_{\text{double}} - \kappa_{\text{single}}) / \kappa_{\text{single}} \) where \( \kappa_{\text{double}} \) and \( \kappa_{\text{single}} \) represent the measured thermal conductivities of double ribbon bundles and corresponding single ribbons, respectively. This observation is not as expected since it has been argued that the thermal conductivity of a-SiO₂ remains bulklike even as the film thickness is reduced to 3 nm and that propagons only contribute <6% to the thermal conductivity of a-SiO₂.14 Our results, however, clearly demonstrate that thermal phonons can ballistically penetrate through over 5 nm thick a-SiO₂ films, as shown schematically in Figure 1d.

Next we examine how oxide layer thickness affects phonon transport in double ribbon bundles. As described previously, we can vary the a-SiO₂ thickness between the two ribbons through HF etching and oxidation for different periods in ambient environment. For BOE-treated silicon surface, it has been shown that the surface dangling bonds are terminated by hydrogen and fluorine atoms.31−34 The subsequent reagent alcohol rinsing and bonding under high vacuum condition caused the surface remain hydrophobic.35

To better evaluate the thermal conductivity enhancement for the etched double ribbon samples, we also prepared and measured the thermal conductivity of a single Si nanoribbon processed with the same etching procedure. To demonstrate the repeatability, we measured four double ribbon samples (DN#3−6) right after they are processed with HF etch, which yielded essentially identical results and are considerably higher than that of the single ribbon, as shown in Figure 3b. The HRTEM micrograph of the interface of these double ribbon samples is shown in Figure 2b, which displays an a-SiO₂ layer thickness of 1.7 nm, about one-third of the a-SiO₂ thickness in the nonetched sample. The enhanced thermal conductivity of DN#3−6 clearly indicates that a portion of thermal phonons could ballistically penetrate through the vdW interface within 1.7 nm thick oxide layer, which leads to an extended effective phonon MFP in the double ribbon bundles. The excellent repeatability of DN#3−6 also suggests that the extracted thermal conductivity enhancement is well-beyond measurement uncertainty (Section IV in the Supporting Information).

Table 1. Summary of Sample Configurations, Measured Thermal Conductivity and Enhancement, and Extracted Interfacial Parameters for Single and Double Si Nanoribbons at 300 K

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Oxide Thickness (nm)</th>
<th>( \kappa ) at 300 K (W/m·K)</th>
<th>( \kappa ) Enhancement at 300 K (%)</th>
<th>Adhesion Energy (mJ/m²)</th>
<th>Transmission Coefficient at 300 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrophilic Interface</td>
<td>DN#1</td>
<td>5.1</td>
<td>14.6 (±0.92)</td>
<td>0.7</td>
<td>1200 (±100)</td>
</tr>
<tr>
<td>DN#2</td>
<td>5.1</td>
<td>16.1 (±1.14)</td>
<td>11</td>
<td>22</td>
<td>115</td>
</tr>
<tr>
<td>SN</td>
<td>14.5 (±0.95)</td>
<td></td>
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</table>

Hydrophobic Interface (BOE etched)

<table>
<thead>
<tr>
<th>Sample #</th>
<th>Oxide Thickness (nm)</th>
<th>( \kappa ) at 300 K (W/m·K)</th>
<th>( \kappa ) Enhancement at 300 K (%)</th>
<th>Adhesion Energy (mJ/m²)</th>
<th>Transmission Coefficient at 300 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>DN#3−6</td>
<td>1.7</td>
<td>16.3 (±2.17)</td>
<td>22</td>
<td>115</td>
<td>0.38</td>
</tr>
<tr>
<td>DN#7</td>
<td>3</td>
<td>15.3 (±1.15)</td>
<td>14</td>
<td>85</td>
<td>0.25</td>
</tr>
<tr>
<td>DN#8</td>
<td>5</td>
<td>13.7 (±0.89)</td>
<td>2</td>
<td>25</td>
<td>0.04</td>
</tr>
<tr>
<td>DN#9−10</td>
<td>1.7</td>
<td>17.6 (±1.25)</td>
<td>31</td>
<td>360</td>
<td>0.55</td>
</tr>
<tr>
<td>SN</td>
<td>13.4 (±0.88)</td>
<td></td>
<td></td>
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</tbody>
</table>

“DN denotes a double Si nanoribbon, and SN denotes a single Si nanoribbon. The interfacial oxide thickness is measured from high resolution transmission electron microscopy (HRTEM). The uncertainty in \( \kappa \) is determined using the method described in Section IV of the Supporting Information. The interfacial adhesion energy is adopted from the Si wafer bonding experiments under the same processing conditions.30

To further explore the effects of a-SiO₂ thickness on ballistic phonon transmission through the vdW interface, we left double ribbon samples in ambient conditions for various periods for oxide growth. For sample DN#7, which was left in ambient environment for 4 days, the measured oxide thickness is \( \sim 3\) nm, and for sample DN#8, which was left in ambient for 60 days, the oxide thickness is \( 5\) nm, the same as that for the nonetched double ribbon sample. The measured data for these samples are also shown in Figure 3b, and the thermal conductivity for DN#7 is lower than that of DN#3, but still higher than that of the single ribbon. For sample DN#8, the thermal conductivity is essentially the same as that of the single ribbon.

An interesting observation is that if we put a double ribbon sample in high vacuum (\(< 10^{-6} \) mbar) for 24 h, then no further oxidation could occur at the interface even we put the sample back into ambient environment. This is evidenced by the fact that the thermal conductivity of the ribbon bundle, post the high vacuum process, does not show any further decrease even after being left in ambient for \( >40 \) days (Section V in Supporting Information). We attribute this to that under the high vacuum any residual volatile species between the two ribbons will be pumped out of the interface and the vdW interface becomes gas tight.36,37

Annealing at elevated temperature has also been shown as an effective approach to increase the interfacial adhesion energy for silicon wafers with hydrophobic surfaces.30,38 To examine if annealing can influence ballistic phonon transport in ribbon bundles with hydrophobic interfaces, we treated HF-etched double ribbon samples in high vacuum and 430 K for 24 h. The measured thermal conductivity of annealed samples (DN#9−10) is further enhanced compared with the ribbon bundles prepared at 300 K (DN#3−6), as shown in Figure 3b. The further enhanced thermal conductivity for DN#9−10 is because for HF-treated Si surfaces, the surface terminating H is more stable in monohydrides (Si−H) than in dihydrides (Si−H₂); if annealed at a temperature beyond 423 K (150 °C), conversion of Si−H₂ to Si−H results in an increased polarity of the hydride, leading to stronger hydrogen bonding between the surfaces.30,36 The results of all measured ribbon samples are summarized in Table 1 for comparison.

To evaluate the upper limit of the double ribbon thermal conductivity, we also measured the thermal conductivity of a single ribbon of double thickness. In the classical size effect regime, the phonon MFP suppression caused by phonon boundary scattering in thin films and wires are well described.
by the Fuchs–Sondheimer reduction function, and for a ribbon of double thickness the theoretical thermal conductivity enhancement is estimated as ~55% at 300 K (Section VII in the Supporting Information), which is significantly lower than the experimentally observed ~72% as shown in Figure 3b. We attribute this difference to the recently discovered acoustic softening effects in thin Si nanoribbons.

Since the thickness of the measured silicon ribbons is only ~14 nm after HF etching, the effects of a reduced Young’s modulus and acoustic softening have to be considered. Three-point bending test using an atomic force microscope (AFM) yielded Young’s moduli of 103 and 97 GPa for single and double ribbons, respectively, which are very close to each other but much lower than the value of $E_{110} = 170$ GPa for bulk silicon. This reduced Young’s modulus would lead to reduced thermal conductivity as the speed of sound is related to the Young’s modulus according to $v = \sqrt{E/\rho}$, where $E$ and $\rho$ are the Young’s modulus and density, respectively. Meanwhile, for the single ribbon of double thickness, its Young’s modulus is the same as bulk value therefore, other than the classical size effect, the experimentally measured enhancement for the single ribbon of double thickness involves additional contribution from the enhanced elastic stiffness.

To quantitatively understand phonon transport in double ribbon samples prepared under different conditions, we plot the calculated thermal conductivity enhancement in Figure 4a. For all five groups of samples, the enhancement increases as the temperature is reduced, which can be attributed to the higher transmissivity of longer wavelength phonons that are more important at low temperature. For nonannealed samples with 1.7 nm thick oxide (DN#3–6), the thermal conductivity enhancement is ~22% at 300 K. Annealing at 430 K for 24 h boosts the enhancement from 22% to 31%, indicating that a higher adhesion energy at the vdW interface results in a higher transmission coefficient. On the other hand, as the interfacial a-SiO$_2$ layer thickness increases from 1.7 to 5 nm (DN#3, 7, and 8), the thermal conductivity enhancement decreases from 22% to 2% at 300 K, while for sample DN#2 with 5.1 nm thick oxide layer but much enhanced adhesion energy, its room temperature thermal conductivity is still 11% higher than that of a single ribbon. These observations collectively suggest that both the oxide layer thickness and the vdW interfacial adhesion energy play important roles in phonon transmission through the interface.

The spectral phonon transmissivity $t_{\omega j}$, based on the modified acoustic mismatch model (vdW-AMM) can be written as

$$t_{\omega j} = \frac{1}{1 + \left(\frac{\omega_j^2}{K_{\alpha}^2}E_s^2\right)\cos^2 \theta}$$

where $\omega_j$ is the phonon angular frequency; $z_j$ is the acoustic impedance and is equal to the product of density and phonon group velocity; $\rho; j$ represents different phonon modes; $\theta$ is the angle between the interface normal and the phonon propagation direction; and $K_{\alpha}$ is the spring constant per unit area, which is related to the interfacial adhesion energy $E_{ad}$ (Section VIII in the Supporting Information). Equation 1 indicates that the phonon transmission coefficient across the vdW interface decreases as $\omega$ increases.

In addition to disturbance from the vdW force, the interfacial a-SiO$_2$ layer can also diffusely scatter phonons. A simple model for the scattering probability can be written as

$$1 - e^{-(d/\cos \theta)/l_{a-SiO_2}}$$

where $d$ is the oxide thickness, $\theta$ is the angle between the interface normal and the phonon propagation direction in the a-SiO$_2$ layer, and $l_{a-SiO_2}$ is the temperature dependent phonon ballistic penetration depth or MFP in a-SiO$_2$. According to Snell’s law, if the direction of the incident phonon from Si ribbon is $\theta$, the refracted phonon direction inside the oxide layer is determined by $\sin \phi = \frac{\sin \theta}{n_{Si}}$. The speed of sound in Si nanoribbon and a-SiO$_2$, respectively, $v_{SiNR}$ and $v_{a-SiO_2}$ can be calculated as $v_{SiNR} = \sqrt{E_{SiNR}/\rho_{Si}}$, where $E_{SiNR}$ is the measured Young’s modulus, and $\rho_{Si}$ is density of Si. For a-SiO$_2$, previous measurements show that its Young’s modulus remains bulk-like even when the critical dimension reduces to 7.5 nm, and thus we use bulk values to calculate $v_{a-SiO_2}$.

As to the Si/a-SiO$_2$ interfaces, wave packet simulations have shown that for phonons of lower than 1 THz frequencies, their

![Figure 4](image-url)
transmission coefficients at the interface are 0.98 and 0.95 for longitudinal and transverse phonon branches, respectively. These results are comparable with the prediction from the acoustic mismatch model (AMM), which yields transmission coefficients for longitudinal and transverse phonons as 0.98 and 0.94, respectively.48 As such, in our model, we approximate the phonon transmission coefficient at the Si/a-SiO2 interface as

\[ \Gamma_{rj} = \int_0^{\omega_{\max}} \int_0^{\theta_{\max}} C_{v \omega, r, j} \sin \theta \cos \theta \times e^{-d/(\cos(\arcsin(\sin \theta / \sin \theta_{\max})))/l_{\text{SiO}}^2} \, d\theta \, d\omega \]

where \( \omega_{\max} \) is the maximum angular phonon frequency, \( C_{v \omega, r, j} \) is the frequency dependent heat capacity per unit volume, and \( \theta_{\max} \) is the critical angle (which is \( \pi/2 \) as the speed of sound in Si is higher than in a-SiO2). The full dispersion relation of bulk Si is adopted in the model,50 and acoustic softening effect is taken into account by multiplying both the phonon frequency and phonon group velocity using a scaling factor as \( \sqrt{E/E_{\text{bulk}}} \).

Thus, based on eq 2, the thermal conductivity enhancement for double ribbon bundles can be expressed as

\[ \frac{\kappa_{\text{double}} - \kappa_{\text{single}}}{\kappa_{\text{single}}} = \Gamma_2 (\alpha - 1) \]

where \( \Gamma_2 \) is the average of the ballistic transmission coefficients for the three different acoustic phonon branches, and \( \alpha \) is the thermal conductivity ratio of single ribbon of double thickness to that of a single ribbon, which is calculated based on the Fuchs-Sondheimer reduction function (see Section VII in the Supporting Information).

The two key parameters determining the ballistic phonon transmission coefficient through the interface are the spring constant per unit area at the vdW interface, \( K_A \), and the phonon ballistic penetration depth or MFP in a-SiO2, \( l_{\text{SiO}}^2 \). However, for sample DN#2, the high adhesion energy leads to a ballistic phonon transmission coefficient at the vdW interface of approximately unity.45 Thus, the thermal conductivity enhancement for DN#2 is dominated by only one parameter, \( l_{\text{SiO}}^2 \). Therefore, using eqs 2 and 3, we can extract \( l_{\text{SiO}}^2 \) by fitting the thermal conductivity enhancement for sample DN#2, which reduces from 4.9 to 3.7 nm as the temperature rises from 50 to 300 K, as shown in Figure 4b.

As mentioned previously, the Einstein random walk model suggests an MFP of ~0.5 nm for vibrations in a-SiO2.18 This very short MFP is obtained based on a gray model, and if more detailed analysis considering the frequency-dependent transport, a MFP of ~1.1 nm was extracted at 50 K based on bulk a-SiO2 thermal conductivity.19 However, this study clearly shows that phonons can ballistically propagate much longer in a-SiO2, at least in the case when oxide films are in contact with crystalline materials. In fact, it is remarkable to see the 11% thermal conductivity enhancement for the double ribbon with an interfacial oxide thickness of 5.1 nm, which corresponds to a ballistic phonon transmission coefficient of 19%.

We point out that ballistic phonon transport or propagon contribution to thermal conductivity in amorphous materials highly depends on the materials type. For example, it has been recently shown that for amorphous silicon, heat-carrying propagons can transport ballistically up to 10 \( \mu \)m.52 On the other hand, studies of other types of amorphous materials, such as copper phthalocyanine (CnPc)53 and poly(3-hexyloxyphene) (P3HT),54 which are widely used in photovoltaic and thermoelectric applications, indicate that thermal transport are dominated by diffusions. In addition, the phonon MFP of amorphous indium–gallium–zinc oxide (a-IGZO) films,55 a high-performance channel material for thin-film transistors (TFTs) was claimed to be 0.35 nm,66 comparable to the distance of neighboring In–In atoms (0.33 nm).66

With the phonon MFP in a-SiO2 determined, we fit the thermal conductivity enhancement of etched double ribbons with various interfacial conditions. Now the only fitting parameter is \( K_A \) and as shown in Figure 4a, by varying \( K_A \), the fitting result can capture both the trend and magnitude of the thermal conductivity enhancement remarkably well in the whole temperature range. The thickness of the fitting line in Figure 4a represents the range where a ± 0.1 nm uncertainty is considered for the measured oxide thickness. The extracted adhesion energy \( (E_{\text{ad}}) \) at the vdW interfaces is 360, 115, 85, and 25 mJ/m² for the samples that are annealed at 430 K (DN#9 and 10), measured directly after etching (DN#3–6), left 4 days in ambient (DN#7), and left 60 days in ambient (DN#8), respectively (see summary in Table 1). Importantly, the fitted \( E_{\text{ad}} \) are in good quantitative agreement with the reported values from Si wafer bonding experiments.30,35,37 For hydrophobic Si wafers rinsed with alcohol and left in an ambient environment until the interface is fully oxidized, the bonding energy was determined to be 30 mJ/m²,35 which is very close to that of sample DN#8 in this work. In addition, it has been shown that annealing at elevated temperature or vacuum condition could significantly increase the adhesion energy,30,37 especially for alcohol treated Si wafers, where evaporation of alcohol could pull two surfaces into intimate contact.33 Thus, for DN#3–7, 9 and 10, the interfacial adhesion energies are much higher than 25 mJ/m². To illustrate the effects of the adhesion energy and the oxide thickness, we plot a contour map in Figure 5, which presents the calculated room temperature phonon ballistic transmission coefficient as a function of both \( E_{\text{ad}} \) and \( d_{\text{oxide}} \). The circles in the contour denote the fitted \( E_{\text{ad}} \) based on the measured oxide layer thickness \( d_{\text{oxide}} \) and the thermal conductivity enhancement.

In summary, measurements of the in-plane thermal conductivity of double Si nanoribbons stuck together through vdW interfaces with a layer of oxide in-between disclose a ballistic phonon penetration depth that is nearly one order of magnitude higher than the prediction of Einstein’s random walk model. Interesting effects of various interface treatment on phonon transport through the interface have been demonstrated. This study should help to conclude the lasting debate...
of ballistic phonon penetration depth in a-SiO2 and provide important insights into thermal management of nanoelectronic devices. To further understand the phonon transport dynamics in a-SiO2 layers, more strict theoretical analysis with less assumptions could be carried out, and double ribbons of systematically varied thickness could also be studied.

**ASSOCIATED CONTENT**

* Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.nanolett.7b02380.

Double Si nanoribbon sample preparation, AFM thickness characterization, HRTEM characterization of the amorphous layer at the interface, experimental uncertainty, effects of high vacuum treatment on double Si nanoribbons, Young’s modulus measurements, derivation of thermal conductivity ratio $\alpha$, and adhesion energy calculation ($\Gamma_{ad}$).

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Author Contributions

L.Y. conducted thermal conductivity measurements, Young’s modulus measurements, and theoretical modeling. Q.Z. helped with double ribbon sample preparation. Z.C. performed the HRTEM characterization. L.Y. and D.L. prepared the manuscript. D.L. supervised the project. All authors discussed the results and commented on the manuscript.

**Notes**

The authors declare no competing financial interest.

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