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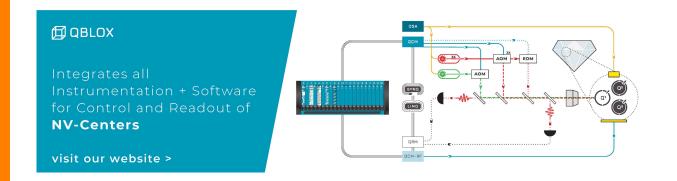


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## Observation of heat transport mediated by the propagation distance of surface phonon-polaritons over hundreds of micrometers

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#### ABSTRACT

Efficient heat dissipation in micro/nano electronics requires long-distance propagation of heat carriers operated above room temperature. However, thermal phonons—the primary heat carriers in dielectric nanomaterials—dissipate the thermal energy after just a few hundred nanometers. Theory predicts that the mean free path of surface phonon-polaritons (SPhPs) can be hundreds of micrometers, which may improve the overall dissipation of heat in nanomaterials. In this work, we experimentally demonstrate such long-distance heat transport by SPhPs. Using the  $3\omega$  technique, we measure the in-plane thermal conductivity of SiN nanomembranes for different heater-sensor distances, membrane thicknesses, and temperatures. We find that thin nanomembranes support heat transport by SPhPs, as evidenced by an increase in the thermal conductivity with temperature. Remarkably, the thermal conductivity measured 200  $\mu$ m away from the heater is consistently higher than that measured 100  $\mu$ m closer. This result suggests that heat conduction by SPhPs is quasi-ballistically over at least hundreds of micrometers. Our findings pave the way for coherent heat manipulations above room temperature over macroscopic distances, which impacts the applications in thermal management and polaritonics.

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Heat conduction in semiconductors is essentially governed by the mean free path of heat carriers. In nanomaterials of size comparable to the mean free path, heat conduction can even happen ballistically,<sup>1–3</sup> without dissipation of thermal energy, which could be used for cooling in microelectronics.<sup>4</sup> However, even in silicon membranes, the mean free path of phonons at room temperature is as short as 30–300 nm,<sup>5,6</sup> which only becomes shorter at higher temperatures.<sup>7</sup> Such a short propagation length makes it unfeasible to attain the ballistic regime in the micro-sized dielectric materials used in modern electronics. Therefore, heat carriers with a longer propagation length are desirable.

Over the past two decades, the propagation, detection, and energy transport of surface electromagnetic waves have attracted attention due to the predominance of surface effects over the volumetric ones in nanostructures with high surface-to-volume ratio.<sup>8</sup> Certain types of surface electromagnetic waves may even carry heat<sup>9-12</sup> and, thus, improve the thermal performance and stability of nanoscale devices.<sup>13-15</sup> One type of such waves is the surface phonon-polaritons (SPhPs)—evanescent surface waves generated by the coupling of photons with optical phonons.<sup>16-21</sup> SPhPs can be excited with a monochromatic light source with given frequency, which correspond to the SPhP resonant frequency. It is reported that the SPhPs generated on SiC are excited and detected optically by using scattering-type near field optical microscopy (s-SNOM).<sup>22</sup> Theoretical works predict that SPhPs can propagate along the surface of polar dielectric materials without dissipation for hundreds of micrometers,<sup>23,24</sup> which is orders of magnitude longer than the typical mean free path of phonons. Moreover, recent experiments demonstrated that in thin dielectric membranes, SPhPs can even carry heat at least as efficiently as phonons.<sup>25,26</sup> However, the quasi-ballistically conduction remains to be experimentally demonstrated.

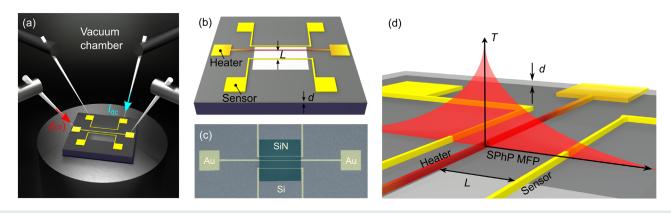
In this work, we aim to demonstrate that the heat transport via SPhPs depends on propagation distance over hundreds of micrometers. Our experiments compare the thermal conductivity of SiN membranes measured 100 and  $200 \,\mu$ m away from the heater. The SPhPs in the SiN film are expected to exist in the frequency range between 125 and 225 Trad/s.<sup>25</sup> The intrinsic propagation length of SPhP depends on the sample's thickness, and their spectra are shown in the supplementary material. The propagation length scales down as the thickness increases. Assuming that phonon transport in amorphous membranes is purely diffusive at these length scales,<sup>27</sup> any difference in the measured thermal conductivity can only be explained by the presence of additional heat conduction channels. Thus, by conducting the measurements for different heater-sensor distances, membrane thicknesses, and temperatures, we demonstrate how heat conduction by SPhPs occurs in thin membranes and remains quasiballistic for hundreds of micrometers.

Samples of amorphous SiN membranes with thicknesses of 50, 100, and 200 nm were suspended in a  $1 \times 1 \text{ mm}^2$  square window on Si substrates, while the thickness of 30 nm was suspended in a 0.5  $\times$  0.5 mm<sup>2</sup> square window. The SiN membranes are commercially available from the NORCADA company and were suspended by back etching the silicon substrate. These high-stress ( $\approx$ 250 MPa) membranes were flat (curvature radius of 4 m) to ensure an ideal measurement for the long-range SPhP propagation. Gold wires of 4  $\mu$ m in width and 100 nm in height are serving as the heater and the sensors. A layer of 10-nm-thick Cr was deposited between Au and SiN to enhance the adhesion.

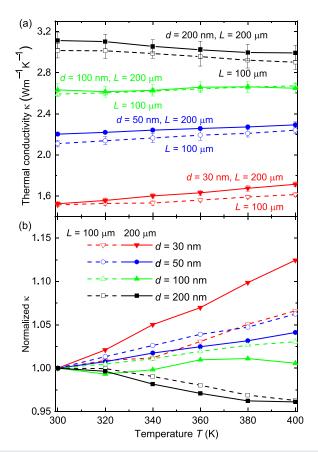
To measure the in-plane thermal conductivity of SiN membranes, we used the  $3\omega$  method.<sup>28,29</sup> Figure 1(a) illustrates the  $3\omega$ setup with four probes coupled to a heating stage and placed in a vacuum chamber. To probe the thermal conductivity at different propagation distances, two sensor wires were, respectively, placed 100 and 200  $\mu$ m away from the heater wire, as shown in Figs. 1(b) and 1(c). Although the SPhPs have intrinsic propagation length up to 1 m,<sup>17</sup> in our designed measurement, the propagation of the SPhPs is truncated by the sensor, where they thermalize with phonons and raise the temperature detected by the sensor.<sup>30</sup> Figure 1(d) illustrates how SPhPs are excited by the Joule effect in the heater wire. The in-plane thermal conductivity was extracted by comparing the  $3\omega$  signal with an analytical model (supplementary material "Heat diffusion model"). Thermal radiation losses were taken into account as explained in Supporting Information "Heat diffusion model."

Figure 2(a) reports the temperature dependence of in-plane thermal conductivity of SiN membranes of different thicknesses. The data measured for the heater-sensor distances of 100 and 200  $\mu$ m are joined by the dashed and solid lines and labeled as  $\kappa_{100}$  and  $\kappa_{200},$  respectively. Longer heater-sensor distance beyond 200  $\mu$ m was probed, yielding at too low signal-to-noise ratio and unworkable data. The thickest membrane (d = 200 nm) at 300 K has  $\kappa_{100} \approx \kappa_{200} \approx 3.1 \text{ W m}^{-1} \text{ K}^{-1}$ , which decreases by about 4% at 400 K. This reduction above room temperature is characteristic of the phonon thermal conductivity driven by the internal scattering processes of phonons. Although the thermal conductivity of 200-nm-thick membranes also shows a difference between L = 100 and 200  $\mu$ m, this difference is statistically insignificant and lays within experimental uncertainty. Likewise, the thermal conductivity of the 100-nm-thick membrane is nearly independent of the heater-sensor distance and temperature. The independence of the heater-sensor distance also indicates that the thermal transport is mainly driven by the phonons and, thus, is diffusive at this scale, as predicted by the theory.<sup>17</sup> Mean free paths of phonons in SiN are short but still extend to the 100 nm range<sup>27</sup> and are larger than the thicknesses of the thinner SiN membranes. Consequently, the phonon mean free path is limited by boundary scattering, leading thermal conductivity to decrease as the membrane thickness reduces.

Interestingly, the thermal conductivity of the two thinner membranes (30 and 50 nm) shows a 5% increase with temperature [shown in Fig. 2(b)], which differs from that of the thicker ones. For the 50-nm-thick membrane,  $\kappa_{100}$  ( $\kappa_{200}$ ) increases from 2.1 W m<sup>-1</sup>K<sup>-1</sup> (2.2 W m<sup>-1</sup>K<sup>-1</sup>) at 300 K to 2.24 W m<sup>-1</sup>K<sup>-1</sup> (2.29 W m<sup>-1</sup>K<sup>-1</sup>) at 400 K. For the 30-nm-thick membrane, this enhancement raises to



**FIG. 1.** Schemes of the  $3\omega$  experimental setup and the SiN sample. (a) Illustration of the SiN sample in a vacuum chamber. The sample is measured with an improved  $3\omega$  setup. AC input current  $i(\omega)$  is supplied to the central heater wire to generate a heat flux by Joule heating. On the sensor located at a distance *L* (100 and 200  $\mu$ m), the temperature raise is calculated from the  $2\omega$  signal measured with a lock-in amplifier on a DC Wheatstone bridge. (b) Schematic of a suspended SiN sample, where *L* is the distance between the heater and the sensor, and *d* is the thickness of suspended SiN. (c) Scanning electron microscope image of the top view of the SiN sample. The yellow color highlights the Au wires, and the green color highlights the suspended SiN. (d) Illustration of a SiN membrane supporting the heater-to-sensor propagation of SPhPs excited by the Joule effect in the heater wire.



**FIG. 2.** Thermal conductivity of SiN membranes. (a) Thermal conductivity as a function of temperature for membranes of different thicknesses. The solid and dashed lines stand for the measurements performed with the sensors placed 100 and 200  $\mu$ m away from the heater, respectively. (b) In-plane thermal conductivity normalized by the values at 300 K.

6.6% and 12.4% for the thermal conductivity values  $\kappa_{100}$  and  $\kappa_{200}$ , respectively. The observed increase in the thermal conductivity with temperature is the sign of the heat conduction by SPhPs. As SPhP contribution becomes stronger at higher temperatures, it compensates for the thermal conductivity reduction due to the phonon–phonon scattering, thus leading to the overall increase in the thermal conductivity with temperature. This explains the difference in trends for thicker and thinner membranes.

Remarkably, the thermal conductivity measured 200  $\mu$ m away from the heater is higher than that measured 100  $\mu$ m away ( $\kappa_{200} > \kappa_{100}$ ). Since the phonon transport is diffusive at such a long length scale,<sup>27</sup> we attribute the observed difference in the thermal conductivity to the ballistic heat conduction by SPhPs. Indeed, lengthdependent thermal conductivity is one of the signs of ballistic heat conduction<sup>1,3,31,32</sup> and implies that heat carriers propagate ballistically at least as far as the length dependence is measured.

Figure 2(a) seems to indicate a similar difference between  $\kappa_{100}$ and  $\kappa_{200}$  in the 30 and 50 nm cases only because the thermal conductivities of the 50 nm membrane are higher. A clearer perspective is, indeed, provided when the thermal conductivity ratio  $\kappa_{200}/\kappa_{100}$  is plotted (supplementary material Fig. S9), where the error bars for this ratio include the unity in the case of the 50 nm thick film. A difference due to heater-sensor distance, therefore, cannot be strictly concluded. Conversely, the lowest value of the error bars clearly increases beyond unity in the 30 nm case. Hence, the difference between  $\kappa_{100}$  and  $\kappa_{200}$  can only be corroborated in the 30 nm thick membrane, which indicates the ballistic SPhP transport occurs in this case.

To better understand the behavior of the measured thermal conductivity, we analyze the SPhP contribution to the heat transport in SiN membranes. According to the Boltzmann transport equation, under the relaxation time approximation, the SPhP contribution to the in-plane thermal conductivity ( $\kappa_{\text{SPhP}}$ ) of a membrane of thickness *d* is given by<sup>16</sup>

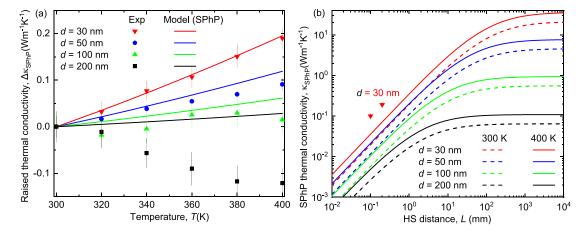
$$\kappa_{\rm SPhP} = \frac{1}{4\pi d} \int_{\omega_{\rm L}}^{\omega_{\rm H}} \hbar \omega \Lambda_{\rm e} \beta_{\rm R} \frac{\partial f_0}{\partial T} \mathrm{d}\omega, \tag{1}$$

where  $\hbar$  is the reduced Planck constant,  $\Lambda_e$  is the effective propagation length of SPhPs propagating along the membrane surface with a complex wavevector  $\beta$ ,  $\beta_R = \text{Re}(\beta)$ ,  $f_0$  is the Bose-Einstein distribution function, T is the average membrane temperature, and  $\omega_H$  am  $\omega_L$ stand for the highest and lowest frequencies supporting the propagation of SPhPs, respectively. Since heat generated by SPhPs inside a material is proportional to the imaginary part of its dielectric function,<sup>12</sup> this material property is strongly linked to these frequencies and hence to  $\kappa_{\text{SPhP}}$ .

To take into account finite lateral size of our SiN membranes, their effective thermal conductivity is determined using Mathiessen's rule  $\Lambda_e^{-1} = \Lambda^{-1} + L^{-1}$ , where  $\Lambda$  is the intrinsic propagation length of polaritons and *L* is the heater-sensor distance (100 and 200  $\mu$ m). Thus, the values of *L* are much shorter than those of  $\Lambda$  and much longer than the intrinsic mean free path of phonons in amorphous materials<sup>33,34</sup> around room temperature. Equation (1), thus, establishes that the values of  $\kappa_{\text{SPhP}}$  are determined by the SPhP dispersion relation  $\beta(\omega)$  driven by the membrane dielectric function that usually changes with temperature and membrane thickness.<sup>35</sup> However, for the thicknesses and temperatures considered in this work, the dielectric function of our SiN membranes is nearly independent of these parameters.<sup>25</sup>

Figure 3 shows the SPhP thermal conductivity predicted by Eq. (1) for different temperatures, heater-sensor distances, and membrane thicknesses. Thinner and hotter membranes with longer heater-sensor distances exhibit higher SPhP thermal conductivity. This behavior is expected as reported for SiO<sub>2</sub> and SiC membranes<sup>16,17</sup> and opposite to the typical behavior of the phonon thermal conductivity. Thus, it signals the SPhP contribution to the heat transport along the membranes.

For the thinnest membrane (d = 30 nm), the measured thermal conductivity enhancement  $\Delta \kappa_{\text{SPhP}} = \kappa(T) - \kappa(300 \text{ K})$  is compared with prediction of the model when the heater-sensor distance is  $200 \ \mu\text{m}$  [Fig. 3(a)]. However, in thicker membranes (d > 30 nm), the phonons make a significant contribution to heat conduction, and the discrepancy between measured (phonons and SPhPs) and theoretical (only SPhPs) values increases with membrane thickness. Remarkably, Fig. 3(b) shows that theoretically, SPhPs can travel ballistically as far as one meter, and the values measured on the 30-nm-thick membrane follow the predicted trend. Since the thermal conductivity enhancement is driven by the SPhPs propagation, which is long (>1 cm) in polar materials, the observed length dependence for SiN films is also expected to be present for other polar films, such as SiC and SiO<sub>2</sub>.<sup>36,37</sup>



**FIG. 3.** Theoretical SPhP thermal conductivity is compared with the experimental values. (a) Enhanced SPhP thermal conductivity as a function of temperature, when sensor is 200  $\mu$ m away from the heater. (b) Theoretical SPhP thermal conductivity as a function of the heater-sensor distance for four SiN membrane thicknesses at 300 and 400 K. The experimental result of 30-nm-thick membrane (red solid triangles) is compared to theoretical values.

In conclusion, we experimentally demonstrated long-distance heat conduction by SPhPs. We measured the in-plane thermal conductivity of SiN membranes and showed that in thin membranes, the heat conduction is mainly driven by SPhPs. Moreover, the thermal conductivity was higher when measured further away from the heater. The observed difference in the thermal conductivity is attributed to ballistic heat conduction by SPhPs and is compared with our theoretical model. Thus, our experimental results suggest that the SPhPs can carry heat ballistically over hundreds of micrometers. Considering that the coherence length is comparable to the propagation length,<sup>19</sup> new engineering possibilities might become available using interference of SPhPs. This work offers an alternative approach to improve the heat dissipation in microelectronics and efficiency in silicon photonics.

See the supplementary material for the detailed information about the theoretical background, the analysis of the experimental results, and the auxiliary measurements. Further discussion is welcomed by contacting our corresponding authors.

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#### AUTHOR DECLARATIONS

#### Conflict of Interest

The authors have no conflicts to disclose.

#### Author Contributions

Yunhui Wu: Investigation (equal); Writing – original draft (equal); Writing – review & editing (equal). Jose Ordonez-Miranda: Formal analysis (equal); Writing – review & editing (equal). Laurent Jalabert: Investigation (equal); Writing – review & editing (equal). Saeko Tachikawa: Investigation (equal); Writing – review & editing (equal). Roman Anufriev: Investigation (equal); Writing – review & editing (equal). Hiroyuki Fujita: Methodology (equal); Writing – review & editing (equal). **Sebastian Volz:** Funding acquisition (equal); Project administration (equal); Supervision (equal); Writing – review & editing (equal). **Masahiro Nomura:** Funding acquisition (equal); Project administration (equal); Supervision (equal); Writing – review & editing (equal).

#### DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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