Blocking phonons via nanoscale geometrical design

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By introducing a serpentine structure in a straight silicon nanowire, we have experimentally achieved a significant reduction in its phonon thermal conductance at low temperature ($T<5$ K.) The magnitude of this effect is one order of magnitude larger than expected based on a thermal resistors picture. With a more refined theoretical model that considers the frequency dependence of phonon transport, we are able to quantitatively account for the experimental results of straight and serpentine nanowires in the whole temperature range. This experimental demonstration of a large, purely geometry induced effect on nanoscale thermal conductance contrasts strikingly with the negligible effects reported on a different nanoscale system in a previous publication.

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A current major challenge in thermal physics is the control and manipulation of phonons, the quantized modes of vibration of the lattice, and hence the heat transfer at small length scale.[1,2] Such control via nanoengineering, also termed nanophononics,[3] may one day enable the development of thermal logic components analogous to those existing in electronics like thermal rectifiers,[4–6] diodes or logic gates and memories.[7] Below 10 K, the modification of phonon transport in nanoscale systems as compared to the bulk has been at the core of rare but significant experiments.[8–13] The effects of surface roughness and contact resistance on phonon transport through nanowires and nanotubes has been studied in numerous publications.[14–19] However, only one work has investigated the effects induced by changes in the conductor's geometrical shape, in the particular case of a carbon nanotube (CNT).[20] That experiment showed that strong bending of the CNT does not affect phonon transport significantly. In contrast, in this letter we show that changes in geometrical shape can significantly affect heat flow through Si nanowires.

By engineering serpentine shaped nanowires, the phonon conductance is reduced by nearly 40% at temperatures below 5 K. We will show that this amount of reduction is strikingly large and cannot be understood by a simple thermal resistors model. We have performed a detailed transmission function analysis, which identifies the presence of a minority of phonons possessing very long mean-free paths as the reason for the large effect observed. This model yields a satisfactory agreement with experimental measurements.

We engineered and measured the phonon thermal conductance of two different monolithic systems: a straight silicon nanowire, and a serpentine nanowire consisting of a silicon nanowire having a serpentine structure, hereafter referenced as serpentine nanowire (see Fig. 1). The nanowires are fabricated from SOI substrate by e-beam lithography. The total length of each structure has been purposely set to 10 μm in order to easily compare the thermal measurement of the two systems. The section of the nanowires is 100 nm by 200 nm, and the serpentine structure is 400 nm long. The central symmetry of the structures is required by the 3ω method employed to measure the thermal conductance.[21] The hottest point is located in the middle of the systems, and heat flows from the center of the nanowire to the heat bath on both sides. Therefore, the nanowire length, L, must be taken as the distance between the center of the structure and the heat bath, i.e., 5 μm. For the measurement of the thermal conductance, a typical power of a few tens of femtowatts is dissipated in the niobium nitride (NbN) transducer deposited on top of the nanowire creating a temperature gradient smaller than 1 mK. The values of the measured thermal conductance are around a few universal quanta of thermal conductance $K_0=\pi^2k_B^2T/3h\sim10^{-12}T\mathrm{W/K}^2$. The theoretical low-temperature thermal conductance of a nanowire with no contact resistance is $4K_0$.[22] The values of thermal conductance are thus presented in units of $4K_0$ [Fig. 2(a)], and correspond to the thermal conductance of 5 μm long nanowires (half of the total length of the structures). The sensitivity of the experiment is much less than one pW/K at 1K and the relative uncertainty is less than few percent.

In Fig. 2(a), we present the thermal conductance measured on the serpentine nanowires (the purple triangles, red squares, and violet dots) as compared to the measurement made on the straight nanowire in blue. The measurement on the three serpentine nanowires are quite reproducible, giving a value at 3 K on the order of $\sim4K_0$, i.e., 12 pW/K. As

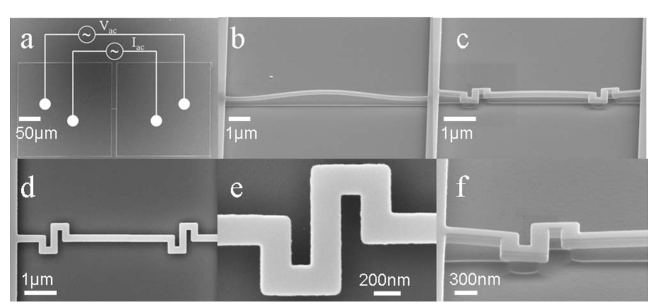

FIG. 1. SEM pictures of the different studied systems. (a) The suspended nanowire linking the two pads where the electrical leads are connected for the 3ω measurement. (b) The straight nanowire used as the reference sample. (c) The serpentine nanowire composed of two serpentine connections to the heat bath. (d) Top view of the serpentine nanowire. (e) Zoom-in top view of the phonon trap. From center to center, the length of the discontinuity of alignment is exactly 400 nm. (f) Side zoom-in view of the suspended serpentine connection.

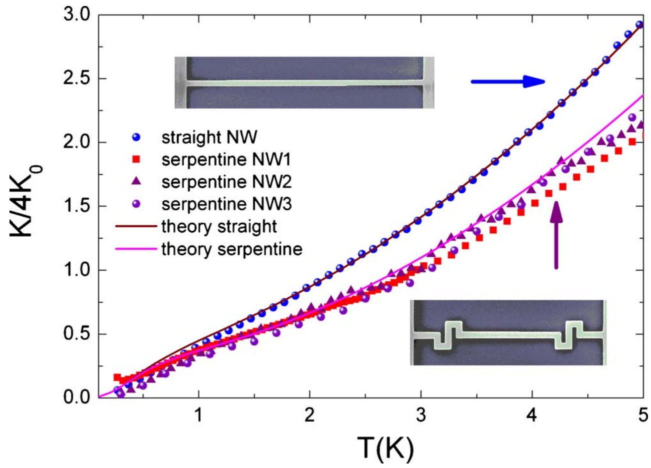

FIG. 2. (Color online) Thermal conductance of 5 μm long nanowires normalized to four times the universal value of thermal conductance versus temperature and the related theoretical fits. The theoretical calculations are in solid lines and the experimental data are presented with symbols.

expected, the thermal conductance of each wire is continuously decreasing with the decrease in temperature. However, major differences can be seen between the straight wire and the serpentine ones. The latters are conducting heat much less than their counterpart. The only difference between the two is the presence of the serpentine. This is the main result of this study: via a purely geometrical effect phonon transport can be strongly blocked, resulting in a reduction in the thermal conductance ranging between 20% and 40%.[23] This amount of reduction is very surprising because the straight wire's thermal resistance is already very high. The measured thermal conductance σ_{str} of the straight wire at 5 K is about 40 times smaller than the ballistic thermal conductance corresponding to its cross section, σ_{ball}.[24] For the serpentine to further reduce the conductance by 40%, the intrinsic resistance of the serpentine part, σ_{db}^{-1}, would have to be such that $\sigma_{str}^{-1}+\sigma_{db}^{-1}\sim\sigma_{str}^{-1}/0.4$, i.e., $\sigma_{db}\sim\sigma_{ball}/60$. In other words, only one in 60 phonons would be allowed to pass through the serpentine structure, which is an unrealistically small transmission probability. In contrast, the classical particle transmission probability value for such a serpentine with mirror-like walls is about 0.43. This implies a thermal resistance associated to the serpentine about 17 times smaller than the total wire resistance, which would yield a minor effect of about 6% on the total thermal conductance. The actual effect we have measured is almost one order of magnitude larger. This means that such oversimplified discussion in terms of resistances is clearly insufficient, and a more sophisticated treatment of the phonon transport process is necessary to understand the striking reduction observed. In what follows, we present a theoretical calculation in terms of frequency-dependent transmission functions, which quantitatively accounts for the experimental results.

In the low-temperature range we are investigating, virtually all the phonon scattering processes are elastic. Therefore, the thermal conductance σ consists of the added independent contributions of different phonon frequencies. At each frequency this is simply the energy of the phonon $\hbar\omega$ times the net propagating number of phonons times their average transmission probability across the structure. It can be expressed in general as[14]

$$\sigma(T) = \int_0^\infty \mathcal{T}(\omega)\hbar\omega\frac{df}{dT}\frac{d\omega}{2\pi}, \tag{1}$$

where $\frac{df}{dT}=\frac{\hbar\omega}{k_BT^2}\frac{e^{\hbar\omega/k_BT}}{(e^{\hbar\omega/k_BT}-1)^2}$, and $\mathcal{T}(\omega)$ is called the transmission function of the system. At frequencies above the confined regime ($\omega>2\pi c/\sqrt{A}\sim0.2$ THz, in our wires, where A is the wire's cross section), the phonon dispersions are practically bulklike, and the transmission is very well approximated by[14]

$$\mathcal{T}=\frac{A}{4\pi^2}\sum_{\alpha=1}^{3}\int_0^{\omega/c_\alpha}\tilde{t}^\alpha(\vec{k}_\perp,\omega)d^2k_\perp \equiv \frac{A}{4\pi}\sum_{\alpha=1}^{3}t^\alpha(\omega)\frac{1}{c_\alpha^2}\omega^2, \tag{2}$$

where $t^\alpha(\omega)\equiv\int_0^{\omega/c_\alpha}\tilde{t}^\alpha(\vec{k}_\perp,\omega)d^2k_\perp/\int_0^{\omega/c_\alpha}d^2k_\perp$ is the average phonon transmission probability across the structure at that frequency. This expression is not correct at very low frequency where confinement becomes apparent ($\omega<\sim0.2$ THz), as we discuss later. Nevertheless, for temperatures above 2 K, the influence of those low frequencies on the total conductance is negligible.

The transmission probabilities $t^\alpha(\omega)$ are the most important magnitudes in the above expression since they account for all the geometry related effects on the thermal conductance. They can be adequately modeled as the sum of the inverse transmission probabilities associated with the different scattering obstacles along the wire.[25] There is a contribution from the nanowire's boundary, proportional to the ratio between the boundary mean-free path $\lambda(\omega)$ and the total nanowire length L; and there is a second contribution, t_{obs}, from the contact and any localized obstacles (such as the serpentine). So

$$t(\omega)\simeq\frac{1}{t_{obs}^{-1}+\dfrac{3L}{4\lambda(\omega)}}. \tag{3}$$

It is easy to verify that this equation yields the Callaway formula for bulk materials[14] when $L\to\infty$, and the ballistic limit when $L\to0$. In the case of a straight wire without contact resistance, t_{obs} becomes 1. If λ is frequency independent the integral in Eq. (1) can be performed analytically, yielding

$$\sigma=A\left\langle\frac{1}{c^2}\right\rangle\frac{1}{1+\dfrac{3L}{4\lambda}}\frac{\pi^2k_B^4}{10\hbar^3}T^3, \tag{4}$$

where $\langle\frac{1}{c^2}\rangle=\frac{1}{3}\Sigma_{i=1}^3 1/c_i^2$. This expression was employed in Ref. 21, where an effective mean-free path l_{mfp} was introduced as $\frac{1}{1+\frac{3L}{4\lambda}}\equiv\frac{4l_{mfp}}{3L}$.[26] The thermal conductance in Ref. 21 was close to the ballistic limit, in strong contrast with the current result. We believe this is related to the larger roughness and longer length of the nanowires investigated here.

It is also straightforward to show that the formula employed in Ref. 12 after a paper by Berman[27] is equivalent to the one above, when the proper numerical values are substituted in the formula, and L in Ref. 12 is the total length of the device (in the present paper L corresponds to half the length of the experimental device).

The boundary mean free path is well described by Casimir's expression, as modified by Ziman[28]

$$\lambda(\omega) \simeq \frac{p(\omega)+1}{p(\omega)-1} 1.12\sqrt{A}. \tag{5}$$

The frequency-dependent specularity $p(\omega)$ can be quantified in terms of the average asperity of the surface, η_0. For a single asperity value η, Ziman gives the approximated expression $p(k,\eta) \sim e^{-\pi 2k^2\eta^2}$ with $k \equiv \omega/c$. If there is not just a single asperity value but a distribution $P(\eta)$, then the specularity is given by $p(k) = \int_0^\infty P(\eta) e^{-\pi 2k^2\eta^2} \simeq \int_0^{1/2k} P(\eta)$. Assuming an exponential asperity distribution of the form[12] $P(\eta) = e^{-\eta/\eta_0}/\eta_0$ yields

$$p(\omega/c) = 1 - e^{-c/2\omega\eta_0}. \tag{6}$$

Up to this point, all the theoretical discussion has relied on modeling the phonon dispersions as a three dimensional bulk system. As already mentioned, this is fully justified provided that the phonon frequencies are well above the confined range. In that situation the ballistic transmission function is, to a very good approximation, quadratic in frequency. In contrast, at very low frequency only a handful of modes exist, resulting in a distinct staircase pattern for the transmission function. This is the confined regime. In the frequency interval between $\omega = 0$ and the onset of the first nonacoustic phonon branches of the nanowire, there are only four phonon branches: two flexural, one torsional, and one longitudinal. This means that in this range the ballistic transmission function of an infinitely long nanowire is constant and equal to 4. The onset of the first nonacoustic phonon branches (i.e., which do not pass through $\omega = 0$), may in principle be calculated for the particular nanowire of interest using the elastic constants of the material. Nevertheless, within a simple model like the one presented here, this frequency can be roughly estimated to take place at a frequency equal to the speed of sound divided by the thickness of the nanowire, namely, $\omega_l \sim 2\pi c/\sqrt{A}$. Here we have arbitrarily chosen the value of c to be 9000 m/s, corresponding to the longitudinal speed of sound in Si, so $\omega_l = 0.4$ THz.

Since the thermal conductance is given as an integral over frequencies, one can simply split it into two contributions: one from the bulklike dispersion frequency range ($\omega > \omega_l$), and one from the confined dispersion frequency range ($0 < \omega < \omega_l$). Then, one can approximate the transmission function for the bulklike contribution by Eqs. (2), (3), (5), and (6).

In the confined dispersion frequency range, $0 < \omega < \omega_l$, the transmission function is the sum of the partial transmissivities of the four acoustic branches of the nanowire: $\mathcal{T} = \sum_{i=1}^4 t_i(\omega)$, where $0 < t_i < 1$ is the transmission probability of each of the four branches. Since the junction is T-shaped, these transmission probabilities are dominated by the contact.[29] As shown by Chang and Geller, the phonon branch with the largest transmission probability across the T-junction is the longitudinal acoustic (LA) one.[29] They analytically derived the transmission probability of this branch at low frequency, which is $t_{contact \leftrightarrow LA} \simeq 0.923 \frac{A}{\pi} \frac{\omega^2}{c_t^2}$, where c_t is the speed of sound of the transverse (T) acoustic modes in the bulk material. As also shown in that article, the transmission probabilities of the other three branches (the rotational one and the two flexural ones) depend more rapidly on frequency, so their contribution to the conductance is smaller. It is then reasonable to approximate the transmission function in the confined dispersion frequency range as having the same frequency dependence as the LA branch transmission probability, with an adjustable parameter on the order of 1, f_{adj}, as

$$\mathcal{T}_{low} \simeq f_{adj} 0.923 \frac{A}{\pi} \frac{\omega^2}{c_t^2}. \tag{7}$$

This form allows us to well match the experimental data below 2 K using only one adjustable parameter. The measurements agree well with the theory for a value of $f_{adj} = 0.5$, which is of order 1 as expected. This confirms the adequacy of Eq. (7) to account for the conductance below 2 K.

We now proceed to understand the curves above 2 K. We first analyze the very low-thermal conductance measured on the straight wire. Afterwards we will explain the surprisingly large conductance reduction reported on the bent nanowires. The conductance above 2 K is nearly independent of f_{adj}, and is instead determined by the dominant surface asperity η_0. The mean asperity value is determined by the sensitivity specification of the resist used in the lithography, which is of a few nanometers. This was also verified by SEM measurements. Therefore, η_0 cannot be considered a free adjustable parameter. A value of $\eta_0 = 4$ nm is found to yield good results in our calculation.

In principle, the smallest conductance one could expect to attain at these temperatures would be that of a completely diffusive surface, with $p = 0$ at all frequencies (the Casimir limit). Using Eqs. (1)–(5), and the fact that the mean-free path in this case is $\lambda = \frac{L}{1.12\sqrt{A}}$, this straightforwardly yields

$$\frac{\sigma}{4K_0} \geq \frac{k_B^2 T^2 A \pi}{5\hbar^2} \left\langle \frac{1}{c^2} \right\rangle \frac{1.12\sqrt{A}}{L}. \tag{8}$$

Here, $\langle \frac{1}{c^2} \rangle \equiv \frac{1}{3} \sum_{i=1}^3 \frac{1}{c_i^2}$ is the average of $1/c^2$ over the three bulk acoustic branches. Taking the experimental value at 5 K, $\frac{\sigma_{5K}}{4K_0} \simeq 3$, implies $\sum_{\alpha=1}^3 \frac{1}{c_\alpha^2} \leq 5.5 \times 10^{-8}$ s^2/m^2. This is evidently not satisfied if one takes the known speeds of sound for Si, $c_l \sim 9000$ m/s, $c_t \sim 5000$ m/s, which yields $c_l^{-2} + 2c_t^{-2} \sim 9.2 \times 10^{-8}$ s^2/m^2. In other words, the measured conductance is smaller than the expected lower limit if all phonon acoustic branches contributed equally.

We note that other works have also reported thermal conductances lower than the Casimir limit, even by two orders of magnitude.[30,31] Although different explanations are possible, we speculate that in the present case the reduced conductance may be associated to a much diminished contribution of the T phonon modes, possibly at the injection level from the heater. To numerically assess this possibility, we

TABLE I. Parameters employed in the calculation. See explanations in main text.

Quantity	Symbol	Value
Nanowire's cross section	A	20 000 nm^2
Mean asperity	η_0	4 nm
Longitudinal speed of sound	c_l	9000 m/s
Transverse speed of sound	c_t	5000 m/s
Confinement frequency cutoff	ω_l	0.4 THz
Low freq. adj. parameter	f_{adj}	0.5
Transverse phonon injection probability	t_{inj}^T	0.017
Serpentine intrinsic transmissivity	t_{obs}	0.2

considered an additional resistive contribution at the interface between the thermometer and the nanowire, in the form of an intrinsic reduction in the transmission probability of the transverse modes. This additional contribution t_{inj}^T is added as in Eq. (3). This implicitly assumes that scattering with asperities does not mix different branches. Using a constant value of t_{inj}^T=0.017, yields a very good match between theoretical and experimental curves (see the brown solid line in Fig. 2.) In contrast, trying to block the longitudinal mode only, or all three modes in the same amount, does not yield a reasonable agreement with experimental data. Here t_{inj}^T is the only true adjustable parameter since the value of the surface asperity is basically determined by the lithographic resolution and thus cannot be chosen freely. The values of all the magnitudes employed in the calculation are listed in Table I.

This strongly diminished contribution from the T-modes could perhaps be happening via a low-temperature phenomenon known as phonon focusing.[32] In crystals such as Ge or Si, the transverse acoustic mode group velocities are not parallel to the phonon wave vector but cluster along particular directions whereas longitudinal phonons are less strongly focused. Consequently T-phonon injection from the heater into the wire will preferentially take place in directions almost normal to the surface. This translates in an increased number of collisions of the T-phonons with the wire surface, and a reduced ability of these phonons to exit the wire, thus diminishing their contribution to the thermal conductance. Further support comes from the fact that the measurements wire with the serpentine also agree very well with the calculated results in the reduced T-branch contribution scenario, with the same parameters as for the straight nanowire (see next paragraph.) Additional investigation of phonon injection from the NbN heater into the Si wire may provide further evidence on how this strong mode selectivity takes place, and whether it is due to phonon focusing. However, this would require a considerable amount of additional experimental techniques beyond the scope of this work.

To model the conductance of the nanowire with the serpentine we use Eq. (3), where the simplest is to assume that t_{obs} is frequency independent as in the classical case. The value t_{obs}=0.2 yields a very good agreement with the experimental curve in the whole temperature range [see the pink solid line in Fig. 2(a)]. We have independently computed the transmission probability of classical particles through a totally specular serpentine via a Monte Carlo simulation, obtaining $t_{obs}\sim 0.4$, which is in the same order as the fitted value. The numerical discrepancy with the best fit result may be due to some of the simplifying assumptions used in the model, to the classical character of the Monte Carlo approach, and/or some degree of experimental uncertainty. The calculation thus allows us to estimate an intrinsic 80% reduction in phonon transmission probability due to the serpentine. This blocking effect due to the serpentine is acting in series with the resistive effect of asperity scattering, which partly masks the former. However, a 40% reduction in thermal conductance is still visible, because asperity scattering is strongly frequency dependent. In other words, although most phonons in the wire scatter diffusively with the boundary, phonons whose wavelengths are longer than η are unaffected by the asperity and reflect in a mostly specular fashion. Those specular phonons give a non-negligible contribution to the thermal conductance, and they are strongly affected by the serpentine. This translates into a large observable effect on the thermal conductance. This is the main conclusion of the theoretical analysis: the observed effect is the result of having strongly frequency-dependent phonon mean-free paths, and it cannot be understood in terms of simpler thermal resistor models.

Our results contrast with the near absence of any observable effects due to bending reported for carbon nanotubes in Ref. 20. However, the results are not in contradiction. The high anisotropy of carbon nanotubes makes them fundamentally different from silicon nanowires, and enables the former to behave like phonon waveguides. In addition, the results in Ref. 20 were obtained at room temperature whereas the ones reported here are at very low temperature.

To conclude, we have demonstrated the efficiency of phonon blocking effect induced by purely geometrical features in a monolithic silicon nanowire. The thermal conductance is reduced by a striking 20–40 % at low temperature. The results cannot be interpreted by the common frequency-independent thermal resistors reasoning. Instead, we presented a theoretical model that takes into account the frequency-dependent character of phonon propagation, correctly accounts for the observed conductance reduction, and yields good agreement with the measured curves in the whole temperature range. The geometrical effect reported could be further enhanced by using nanowires having very smooth surfaces. This kind of phonon blocking system could be used to thermally insulate suspended islands in MEMS and NEMS technology. Furthermore, we speculate that it might be possible to implement the bent structure in smaller nanowires at the scale of 1 nm to be effective at room temperature.

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