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Thermal transport across nanometre gaps: phonon transmission vs air conduction

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Heat transfer between two surfaces separated by a nanometre gap is important for a number of applications ranging from spaced head disk systems, scanning thermal microscopy and thermal transport in aerogels. At these separation distances, near field radiative heat transfer competes with heat transfer mediated by phonons. Here we quantity the contribution of phonon assisted heat transfer using lattice dynamics combined with ab-initio calculations. We clearly demonstrate that phonons dominate by far heat transfer for subnanometre gaps. Strikingly, we conclude that even in the situation where the gap is filled by air molecules, phonons provide the dominant energy channel between the two solids nearly in contact. Our results predict orders of magnitude enhanced phonon heat transfer compared to previous work and bring forward a methodology to analyse phonon transmission across nanoscale vacuum gaps between apolar materials.

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Heat may be transferred by three main mechanisms namely conduction, radiation and convection [1]. Convection occurs when heat is transported by the macroscopic motion of a supporting medium. Heat conduction is also mediated by a supporting medium, and usually not relevant in vacuum : it is of common sense that two bodies in vacuum exchange heat only through radiation. The situation becomes less clear however at the nanoscale, when the separation distance between two bodies is in the nanometre range. At these length scales phonons may be transmitted across the gap between the two solids and compete with radiative heat transfer thus contributing in the heat exchange between the two bodies.

Recent experimental investigations have reported measurements of heat transfer in the extreme near field regime, where the separation distance between two objects is in the nanometer range [2, 3]. While one study shows moderate deviations to near field predictions (Rytov's theory) [4], the other has reported orders of magnitude enhancement. In this Letter, we quantify the phonon contribution to heat transfer in the ultra near field regime on the basis of ab-initio based lattice dynamics calculations. This method enables us to accurately account for phonon dispersion in the two materials nearly in contact. We demonstrate that at the nanometre scale, phonons are the main energy carriers flowing across the gap, wether it is filled or not with air molecules.

Heat transfer between two bodies separated by small distances has attracted the attention of physicists for decades [5–8]. Early studies were motivated by the possibility to observe deviations to Planck's theory of radiation. These pioneering measurements were performed

at low temperatures, when Wien's thermal wavelength is macroscopic hence facilitating the observation of deviations at relative large separation distances [9]. The last decade witnessed intensive effort toward the evidence of extraordinary heat flux in the near field regime [10–19], ever pushing the limits of separation distances.

These fundamental investigations have been fostered by the technological side of the problem. Radiative heat transfer across microscale gaps is at the core of near field thermophotovoltaics, an emerging field of investigation [20]. At shorter separation distances, the applications concern thermal rectifiers [21], heat assisted magnetic recording [22] and scanning thermal microscope [23]. Thermal transport across nanocavities controls also the thermal conductivity of silica aerogels [24].

Despite its technological importance, thermal transport between two bodies nearly in contact still resists a full theoretical treatment. One of the yet unsolved question is how to describe the gradual transition between radiative and conductive heat transfer in the extreme near field regime. Depending on the separation distance, different regimes may be distinguished. Indeed, when two bodies are separated by macroscopic distances, thermal transport is very well described by Planck's blackbody radiation law [1, 25]. When the separation distance becomes comparable to Wien's thermal wavelength $(\lambda \simeq 10 \mu m \text{ at } 300 \text{ K})$, radiation can exceed by orders of magnitude blackbody law. In this regime, photons emitted by one of the two media tunnel across the gap, and the overlapping evanescent waves yield enhanced heat transfer. At the other extreme when two bodies are in contact, heat transfer is mediated by phonon transmission. At nanometre distances, the distinction between radiation and conduction becomes somewhat blurred, and this is precisely this regime that we investigate in this Letter.

Several mechanisms have been invoked to come into play when the separation distance is ultra small [26– 29]. They have in common to involve the coupling between phonons and electric fields, a situation relevant to piezoelectric or polar materials. Here, we consider phonon contribution to heat transfer across *apolar* materials. Our calculations show that phonons are the main energy channel for nanometre gaps, thus correcting by orders of magnitude previous estimates of phonon assisted thermal transport [30].

To calculate the contribution due to phonon across nanoscale gaps, we employ ab-initio based lattice dynamics (aiLD) [31, 32]. The principle of the method is inspired by Zhao and Freund work [33], which considered empirical potentials to describe the atomic lattice dynamics at the interface. We basically extend Zhao and Freund's method obtained from ab-initio first principles calculations. In brief, the system is divided in three regions namely, the left and the right leads and the interface, as illustrated in fig. 1.



FIG. 1. (Color online) Illustration of the configuration considered : two semi-infinite solids separated by a gap of width d.

In the bulk leads, we solve the dynamical equations:

$$(\phi_{\alpha,\beta} - m\omega^2 \delta_{\alpha,\beta}) u_\beta(\omega) = 0 \tag{1}$$

where $\Phi_{\alpha,\beta}$ denotes the harmonic force constant tensor, ω is the phonon frequency and $u_{\beta}(\omega)$ denotes the components of the atomic displacement along the direction β .

In the junction region, the equations to be solved write :

$$(\phi_{\alpha,\beta}^{AA} - m_A \omega^2 \delta_{\alpha,\beta}) u_{\beta}^A(\omega) + \phi_{\alpha,\beta}^{AB} u_{\beta}^B(\omega) = 0 \qquad (2)$$

$$\phi^{BA}_{\alpha,\beta}u^A_{\beta}(\omega) + (\phi^{BB}_{\alpha,\beta} - m_B\omega^2\delta_{\alpha,\beta})u^B_{\beta}(\omega) = 0 \qquad (3)$$

here the subscripts A, B denote the two materials, and we have introduced the corresponding Hessian matrices $\phi^{AA}, \phi^{BB}, \phi^{AB}, \phi^{BA}$ with obvious notations. The displacement u^A in the incoming medium may be split in an incoming wave having a wavevector k_i and n_r reflected waves, each having a wavevector k_r . Similarly, the displacement u^B in the other medium is a sum of n_t waves characterized by their wavevectors k_t . The numbers n_t, n_r and the expressions of the wavevectors k_r, k_t are given by the conservation equations at the interface, see [32, 33] for further details. From the knowledge of the transmitted displacement, one can infer the *mode* dependent transmission coefficient and interface conductance :

$$\mathcal{T}(\mathbf{k}_t, \nu) = \frac{\rho_A}{\rho_B} \frac{v_{g,t}^2 \cdot |A_t|^2}{v_{g,i}^2 \cdot |A_i|^2} \tag{4}$$

where ν is an index denoting the branch index, ρ_A and ρ_B are the mass density characterizing the two materials respectively, $v_{g,t}^z$ is the group velocities of the transmitted phonons projected along the direction perpendicular to the interface, A_t and A_i are the amplitudes of the transmitted and incident wave respectively. The modedependent thermal conductance has the expression :

$$G(\mathbf{k}_t, \nu) = \frac{1}{V} v_{g,t}^z \hbar \omega \frac{\partial f(\omega, T)}{\partial T} \mathcal{T}(\mathbf{k}_t, \nu)$$
(5)

where V is the volume of the system and f denotes the phonon occupation density.

For gold, the bulk harmonic force constants are calculated with the code VASP [34, 35] using the Generalized Gradient Approximation (GGA-PBE) [36, 37]. For silicon, we extracted the values from the literature [38]. In the S1 of the Supplementary material [39], we illustrate the good agreement between the calculated bulk phonon spectra and available experimental data.

For the interaction between the materials across the gap, we consider a Lennard-Jones potential [40]:

$$\Phi_{zz}(z) = 8\pi\rho\epsilon\sigma\left(\left(\frac{\sigma}{z}\right)^{11} - \left(\frac{\sigma}{z}\right)^5\right) \tag{6}$$

where ρ is the material density and z is the distance to the other medium. The Lennard-Jones parameters ϵ, σ are extracted from [41] for metals, and for semiconductors we used the values derived from the knowledge of the material Hamaker's constant [40], as listed in S2 of [39]. The atomistic calculations will be compared with the predictions of a generalized acoustic mismatch model (AMM), describing phonon transport across the solid/vacuum gap/solid interface. In this model, the transmission coefficient is supposed to be given by $\mathcal{T}(\omega) = 1/(1 + \omega)$ $(\omega/2K)^2 z_m^2$) where $z_m = \rho_m c$ is the acoustic impedance of the material with ρ_m and c the mass density and speed of sound respectively, and $K = n_s (\frac{d^2 V}{dz^2})_{z=h}$ is the interface compliance, with n_S the density of surface atoms and $V(z) = 8\pi\rho^2\epsilon\sigma^4 \left(\frac{1}{720}\left(\frac{\sigma}{z}\right)^8 - \frac{1}{24}\left(\frac{\sigma}{z}\right)^2\right)$ is the *total* interaction potential per unit of surface between the two solids. Further details may be found in the S3 [39]. Last, the contribution due to the presence of air molecules in the gap is estimated following [42], as detailed in S4 of [39].

Figure 2 presents the atomistic aiLD calculations results for the thermal transport across the gap between two gold solids. The calculated conductance due to phonons is compared to the contribution due to different energy channels. Clearly, the contribution due to phonons is by far the largest, for the range of gap widths analyzed here. It is orders of magnitude higher than black body conductance $G_{BB} \simeq 6 \text{ W/m}^2/\text{K}$ [1]. Less expectedly, phonon heat transfer beats near field radiative heat transfer also by orders of magnitude. Last, the contribution due to phonons is higher than air conduction for gap widths smaller than 1 nm. This implies that if the gap is filled with air molecules, long range interactions between the two solids contribute much more to the heat transfer than ballistic transport mediated by the molecules. It is remarkable that the ab-initio calculations yield conductance levels comparable to the AMM predictions. We will rationalize this observation through the analysis of the spectral phonon transmission across the gap. Note that the dip present in the AMM conductance for small gaps is related to the non monotonicity of the Lennard-Jones spring constant, driven by short-range repulsive interactions.

Figure 2 demonstrates that the same qualitative conclusions may be drawn for the silicon/gap/silicon system. In this latter case however, the atomistic calculations give conductance levels lower than the AMM predictions. Nevertheless, phonon contribution still dominates air conduction below 1 nm.

The dominance of phonon over air conduction was shown here at room temperature. To appraise the generality of this behavior, the temperature dependence of the conductance characterizing the two main energy channels-phonon transmission and air conductionhas been reported in fig. 3. Clearly over the wide range of temperatures considered, the contribution due to phonons is the highest. Only at cryogenic temperatures, a very limited number of acoustic modes is populated, and air conduction may become the leading channel.

To get insight in the microscopic mechanisms behind thermal conduction in the gap, we discuss now the spectral contribution to the interfacial heat transfer mediated by phonons. Of particular interest we analyze how acoustic models can accurately capture the contribution of heat flow due to phonons. We concentrate first on the case of gold, for which the spectral transmission and conductance are plotted in fig. 4. For all the gap widths analyzed, only low frequency phonons are transmitted across the gap. This behavior is well captured by AMM models at low frequency, while these latter models tend to overestimate slightly phonon transmission for higher frequencies. On the opposite, the AMM predictions underestimate somewhat the conductance, except for the narrowest gap analysed. This conclusion is in full line with the total conductance calculated and presented in fig. 2. The relatively low values of the conductance predicted by AMM may be explained by the underestimation of the density of intermediate frequencies, contributing to undervalue the spectral conductance.

We now turn to the case of silicon, as illustrated in fig. 5. Here a different scenario emerges : the transmission coefficient of long wavelength phonons is low, and only intermediate frequencies phonons can tunnel across the gap. But even the transmission coefficient of these modes is overestimated by AMM, leading to a discrepancy in the spectral conductance. This analysis explains the relatively low conductance of silicon interfaces, as displayed in fig. 2.



FIG. 2. (Color online) Top : Thermal boundary conductance between two gold solids separated by a nanometre gap of width d, calculated at room temperature. Near-field radiative predictions are taken from [43]. Bottom : Thermal boundary conductance between two silicon solids separated by a nanometric gap of width d. Continuous red line is the generalized AMM model. Near-field radiative prediction is taken from [44].



FIG. 3. (Color online) Thermal boundary conductance between two gold solids (top) and two silicon solids (bottom) separated by a nanometre gap d as a function of temperature. The contribution due to air conduction is shown with dashed purple lines.



FIG. 4. (Color online) Phonon transmission coefficient and spectral conductance between two gold solids separated by a nanometre gap d, as a function of frequency. The solid lines display the atomistic calculations while the dashed lines are the AMM predictions.



FIG. 5. (Color online) Same as fig. 4 for Silicon.

Our calculations involve so far phonon transmission, and it is legitimate to wonder wether alternative energy channels may contribute to heat transfer across the gap. As shown experimentally in [3], electron tunneling may occur only when the gap distance becomes smaller than 0.2 nm, thus precluding any direct electronelectron scattering mechanism for larger separation distances. However, metal electrons may play an indirect role in heat transfer through their couplings with phonons. Indeed, electrons couple with phonons in different ways, as summarized in [45]. Of particular interest here, is the situation of weak electron-electron coupling through the gap. Under these conditions, the electronic temperature should obey an adiabatic condition at the boundaries. In the Section S5 of [39], we show that the effect of the electron-phonon coupling is to reduce the effective thermal boundary conductance by an amount : $G_{\text{eff}} \simeq G/(1 + 2G/\sqrt{k_p G_{\text{ep}}})$, where G is the phonon-phonon conductance that we already calculated, k_p is the phononic contribution to the metal conductivity and G_{ep} is the electron-phonon coupling. This latter expression generalizes the expression derived in [46] in the case of metal/semiconductor interface. Considering the values of k_p and G_{ep} for gold [47, 48] $k_p = 2$ W/mK; $G_{ep} = 2.5 \ 10^{16}$ W.m⁻³.K⁻¹ [47, 48]. The resulting values of the conductance is $0.11 \text{ MW}.\text{m}^{-2}.\text{K}^{-1}$, which is higher than the conductance due to the ballistic motion of air molecules. Note that gold has a relatively low electron-phonon coupling constant, so that the electronphonon interfacial conductance is even higher for other

metals (0.62 MW.m⁻¹.K⁻¹ for aluminium). Interfacial electron-phonon transfer will therefore not impede interstitial heat transfer below air conduction level.

In summary, we investigated phonon mediated thermal transport in the extreme near field regime using ab-initio lattice dynamics calculations. We demonstrated that for nanometre gaps, phonons are the main energy carriers exceeding by orders of magnitude near field radiative heat transfer. Strikingly, the contribution of phonons exceeds also largely ballistic transport assisted by gas molecules in the gap. This implies that even in the situation where the gap is filled with air, long range van der Waals interactions provide the dominant heat transfer mechanism. These results open the door to quantify systematically heat transfer mediated by phonons between apolar materials.

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