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Heat transport through nanoscale gaps - A perspective

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Abstract

This perspective describes the key physical concepts at play capturing transport regimes in gaps with sizes ranging from the micron down to the Ångström. The roles of photons, phonons and electrons are highlighted and illustrated across several examples from the literature. Particularly, two situations for which atomistic calculations provide insights in the microscopic mechanisms at play in thermal transport across nanoscale gaps will be expounded, namely thermal transport across silica clusters and in 2D materials. While experimental investigations are highly challenging and a vast field of opportunities still remains open in this matter, several related domains of application such as Thermophotovoltaics, Thermoionics, Thermal management, Imaging and 2D materials are involved.

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1. INTRODUCTION

1.1 Nanogap Energy conversion and management

A nanogap might seem a bit of an odd object and the question of energy transfer across it is a rather fundamental issue. Indeed, this question clearly raises exciting perspectives on how physics may capture those apertures where the "particle" picture fails and electrostatic forces as well as tunneling of photons and electrons are at play. However, a set of rather concrete situations can be considered as in connection with this heterodox heat transfer. The first one is certainly Thermophotovoltaics [1, 2] where the frequency of the slit radiation can be controlled to fit the electron gap and consequently lead to an optimal conversion. The second one might be Thermoionics [3, 4] where the emission of hot electrons through a slit leads to the cooling of the emitting plate. A more expected field would be the one of nanoporous materials where radiation can finally predominate over solid and air conduction [5]. The last and very active area to be cited here is the one of the Van der Waals materials where atomically thin layers introduce sub-nanometer gaps inside bulk matter, as is the case in graphite or bismuth telluride for instance. A broad range of thermoelectric materials does display this type of lattice cells yielding a reduced phonon transport [6].

1.2 Beyond far-field radiation

Radiative heat transfer between two parallel surfaces set at two different temperatures is a matter of bach-

elor textbooks when it comes to describe photon emission, propagation and absorption between non-ideal surfaces, usually characterized by spectral or grey emissivities. When the distance between the two surfaces is larger than Wien's wavelength divided by 2π , photons wavepackets can be defined and their emission, reflection, absorption and eventually transmission are well captured by the Radiative Transfer Equation or ray tracing techniques. A simple analytical formula that includes the effect of multireflections, for instance, provides the heat flux between two surfaces as a function of their emissivities.

However, when the distance between the surfaces reduces below Wien's wavelength divided by 2π , typically below one micron in the vicinity of room temperature, electrostatic direct exchange between the two surfaces -which can also be seen as the overlap of the evanescent tails of the field above the surfaces- rapidly predominates as the gap shrinks down to a few Ångströms [7]. This near-field heat transfer has been the subject of rising interests since twenty years [8, 9], and has more recently yielded new experimental confirmations accessible to micron-scale cleanroom technologies [10–13]. Near-field radiation is still described by classical electromagnetics and either analytical -in elementary geometries- or numerical solutions are available.

In this perspective, we will also reach the point where surface atomic displacements, or phonons, will directly transmit energy by exciting the phonon modes of the opposite side. This phonon tunneling is expected in a very specific range where the gap remains small enough for the dipole-dipole approximation to be dismissed but large enough to preserve the relevance of Maxwell's pre-

dictions.

1.3 Electrons

When reaching aperture widths of a few Ångströms, wave functions of electrons of opposite surfaces may merge and a quantum description of the interaction is required. This interaction is typically of the scope of Van der Waals forces. This regime can also be described as the transition from radiation to conduction or as 'pseudo-conduction'. The electric field in this picture is replaced by the electron density which constitutes the source of direct interatomic forces.

In the following, the approach will remain mostly fundamental with a constant care of revealing the physical mechanisms at play. In a first section, we will start by describing energy transfer in large gaps –a few microns at most– down to a few Ångströms where radiation and finally 'pseudo-conduction' occur. In the second term, Van der Waals materials will be tackled where nanogaps between atomically thin sheets can be chemically controlled to manipulate phonon scattering.

2. NEAR-FIELD RADIATION IN NANOGAPS

The energy exchange in nanoscale gaps plays important roles in modern electronic devices, where the dimensions have been scaled down to a few nanometers. In nanoscale gaps, the evanescent electromagnetic wave emitted by one body can reach the other one and excite atomic vibrations, i.e., phonons, which makes the heat transfer orders of magnitude larger than the one of black-body limit [14, 15]. The mechanism of heat transfer across gaps of a few nanometers is still an open question and sometimes conflicting results have been obtained in the literature [10, 16, 17]. In the mean time, this field is of great importance for new technologies such as heat management in computers, energy harvesting, heat dissipation and so on.

Theoretically, the prediction of near-field radiation has been mostly formulated by the Maxwell equations [18], which characterize all contributions by electromagnetic waves. When the separation between two bodies is comparable to interatomic distances, heat conduction by phonons should take place. So an open question is how the heat transfer regime transits from near-field radiation to conduction in gaps with only a few nanometers, i.e., from evanescent waves to phonons? The conventional theory of fluctuational electrodynamics has proven to be valid down to hundreds of nanometers while it is still questionable for its application in ultra short gaps as it does not capture the phonon contributions, typically from a few nanometers to even Ångströms. However,

other theoretical works indeed demonstrate that phonons can be tunnelled from one body to another in nanoscale gaps [19–21]. The heat transfer due to the phonon tunnelling can be much larger than the value predicted by the Rytov's theory of macroscopic fluctuational electrodynamics [18]. The previous models on phonon assisted heat transfer through nanoscale gaps has been done for acoustic modes, which illustrates the mechanical coupling between the entire bodies through piezoelectric forces [20, 21]. However, for polar materials, the interactions between ionic charges can also promote the tunnelling of optical phonon modes. The optical phonon modes in dielectrical materials correspond to dipole and multipole oscillations, which can emit electromagnetic waves and thus excite the optical phonon modes in the other objects at the corresponding resonant frequency, i.e., coupling through electric field [22, 23]. In such a picture, the coupling of optical phonons between two dielectrical materials is similar to the mechanism of heat transfer by evanescent modes. As a result, there are typically two types of coupling forces for phonon tunneling in the classical limit, i.e., the piezoelectric coupling [21] and the electric field coupling [22, 23]. The piezoelectric coupling originates from the polarization of the entire body due to structure deformation. It treats the objects as a whole and could play an important role in non-ionic materials. While the electric field coupling comes from the oscillations of charged ions, thus allowing for the direct coupling between each atom pairs, which is especially important in ionic and polar materials. To quantify the heat transfer mediated by the ionic charges in dielectric materials, the atomistic Green's function method has been adopted in literatures. The Green's function technique has the advantages of taking into account the interactions between each atom specifically, thus solves the heat transfer problem from the microscopic point of view. Note that compared to the elastic wave model, the phonon Green's function model explicitly considers the heat carried by each mode, including both acoustic and optical ones.

The first work to study heat transfer in nanoscale gaps by implementing phonon Green's function was achieved between Silica clusters (figure 1 top) [22]. In such a cluster system, three different heat transfer behaviors were found with a particle distance below 20 nm. The first regime starts from contact to a particle size independent gap distance of 4 Å, where the conductance decays rapidly with distance through a d^{-12} dependence. Below 4 Å, the electron clouds of the neighboring clusters starts to merge together, forming chemical bonds between the atoms of neighboring clusters. With the increase of gap distance beyond 4 Å, the conductance decays with the power law of d^{-3} until the particle distance reaches 3-5 times the cluster size, which corresponds to the surface charge-charge interactions. In this distance range, the interaction between the ions of two adjacent surfaces is larger than that between other atoms, hence

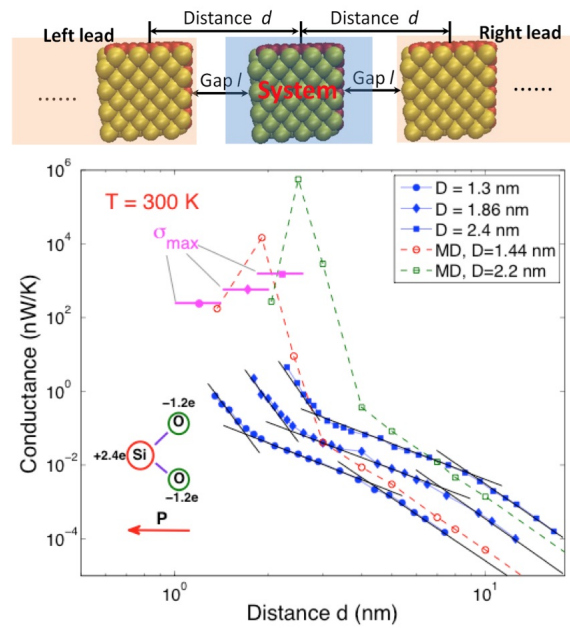


FIG. 1. Schematic illustration of the Green's function calculation set up (up) and the thermal conductance between Silica clusters with different sizes at 300 K as a function of the separation distance d . Reproduced with permission from Phys. Rev. Lett. 112, 114301 (2014). Copyright 2014 American Physical Society.

the heat transfer is predominated by the interactions of surface ions. This mechanism is different from the acoustic tunnelling mechanism considered by other researchers [20, 21], where the couplings take place through the entire particles. Beyond a gap distance 5 times of the cluster size, the conductance follows the trend of d^{-6} . In this range, each cluster can be treated as a dipole formed by ions with positive and negative charges, which eventually promote the heat transfer with the decay law of d^{-6} . The transition from electron overlap to electrostatic interactions in the gap distance of 4 Å thus corresponds to a quantum to classical transition of heat transfer. It is worth to note that the heat transfer behavior observed in Ref [22] is shape dependent. If the geometry of the system changes to nanorods or planes, the conductance power laws, especially at the distances beyond chemical bond formation, will be changed as observed by Chiloyan *et al* [23], where phonon Green's function was also applied to study the heat transfer between two semi-infinite NaCl structures. It is worth to note that electron tunneling might happen beyond 4 Å if there are free electrons as shown recently by Alkurdi *et al* [24], such as in metal systems. However, the heat transfer contribution due to electron tunneling decreases much faster with gap distance compared to phonon contribution and starts to be less important beyond 4 Å.

Experimentally, previous studies of near-field radiation

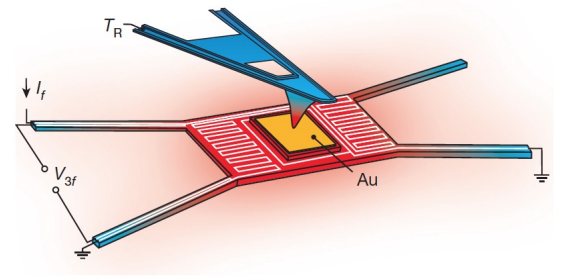


FIG. 2. Schematic of the experimental set-up, in which an SThM probe is in close proximity to a heated substrate. Reproduced with permission from Nature 528, 387 (2015). Copyright 2015 Springer Nature.

has been focused on gap distances larger than hundreds of nanometers due to the technology challenges. With the recent improvement on device fabrications and temperature control, the measurement of heat transport across gaps of a few Ångströms to several nanometers has been realized recently in groups led by Reddy and Meyhofer [10, 16] and Kittel [17]. The measurements were done in customized SThM under high vacuum conditions based on a commercial scanning tunnelling microscope (STM) (figure 2). To hold a stable gap distance of a few nanometers or even Ångströms, the probe tip should possess large stiffness. In Reddy and Meyhofer group's set up, the temperature of the probe tip was measured by the embedded Au-Cr nanoscale thermocouples. The incident laser was applied to measure the deflection of the cantilever, which approaches to the substrate with a constant rate (typically 0.1-0.5 ns/s). The first heat transfer quantification by them [10] was achieved in down to 2-3 nm gaps and extremely large enhancement of radiative heat transfer in such small gaps between both dielectric and metal surfaces was found. Moreover, based on the fluctuational electrodynamics theory, they modelled the heat transfer in such extreme gaps and found good agreement with experimental values, indicating the validity of the traditional fluctuational electrodynamics in predicting the near-field radiation through gaps of a few nanometers. Latter measurements in their group as well as Kittel group pushed the gaps down to 0.2 nm in the Au-Au system with the improvement of tip stiffness up to 10^4 N/m [16]. In the work by Kittel *et al* [17], where the probe temperature was measured by a thermoelectric couple, giant heat transfer was observed when the gaps are smaller than 6 nm. The measured heat flux is more than five orders of magnitude larger than the black body limit and four orders of magnitude larger than the values predicted by conventional fluctuational electrodynamics theory. They ascribed the origin of such large heat flux to phonon tunnelling in those ultra small gaps. However, such a large heat flux might caused by the

contamination of the tip and the substrate [16]. After the cleaning of both probes and substrates by both plasma-cleaning and controlled crashing procedures, Cui *et al* [16] found that the thermal conductance between gaps was largely reduced. However, due to the large noise in their measurement, it is still unclear to which gap sizes the conventional fluctuational electrodynamics fails to describe the radiative heat transfer.

3. NANOGAP CONTROLLED HEAT CONDUCTION IN 2D MATERIALS

When the gap distance between two objects is in the range of the nanometer, the heat transfer between them can also be tuned greatly by bridging them with small molecules, which brings important applications in thermal switches and molecular thermoelectrics. Here we demonstrate that the nanogap between graphene sheets and functionalized graphene oxide (FGO), formed due to the functional intercalated molecules, allows for engineering the inter-layer thermal coupling and the intra-layer heat transfer in a graphene system [25, 26]. Phonon transport on the nanoscale is investigated comprehensively by means of molecular dynamics simulations and atomistic Green's function, considering both the respective contribution of phonons and electrons. These theoretical predictions are corroborated by a set of experimental works including the chemical synthesis of freestanding graphene and graphene oxide with and without functionalization, their composition characterization with Raman spectroscopy and thermal characterization with Fourier transform infrared spectroscopy. Different molecules including 3-Aminopropyltriethoxysilane (APTES) have been tested both in calculations and experiments. This work provides not only insights into the fundamental understanding of heat transport in graphene, but also tackles the key technological challenge of efficient thermal management in the industry of the next-generation integrated circuit.

To explore the effect of functional molecules on the in-plane thermal conductance of the graphene sheets, molecular dynamics simulations are firstly performed to study a nanoscale molecular junction between two stacks of multilayer graphene nanoflakes. The in-plane thermal conductivity κ of the graphene sheet and its interfacial thermal resistance R with the FGO substrate is plotted as a function of the graphene layer number in the film in Fig. 3a and b, respectively. The molecule density is $\rho = 0.081 \text{ nm}^{-2}$. It is firstly shown that the presence of the functional molecule (APTES) results in an unexpected increase both in the graphene film thermal conductivity κ and in R for multilayer graphene. An overall decaying trend of the in-plane thermal conductivity of the graphene film and its resistance R

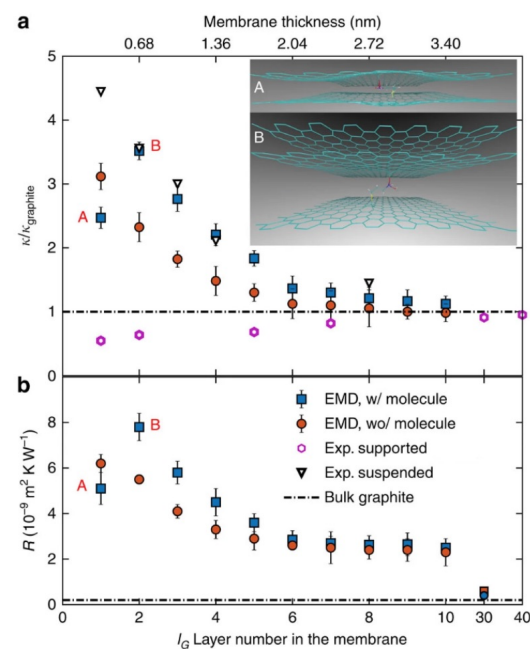


FIG. 3. Thermal resistance and in-plane thermal conductivity of the GBF versus the graphene layer number. (a) Molecular dynamics simulation results of in-plane thermal conductivity of the graphene film and (b) interfacial thermal resistance R between the FGO substrate and the graphene film versus the graphene layer number l_G in the film. Reproduced with permission from Nat. commun. 7, 11281 (2016). Copyright 2016 Springer Nature.

with the substrate versus the layer number is observed until approaching the value of bulk graphite. For a single layer of graphene, κ is reduced by the molecules, which is on the contrary to the case where no molecule links the graphene film and the substrate. This breakdown of the thermal conductivity enhancement is due to a saddle-like surface generated around the molecule's chemical bonds of amino and silano groups connecting the graphene, with the bond center as the saddle point, as shown in the inset A of Fig. 3a. The saddle-like surface strongly scatters all phonon modes, thus decreasing κ of the graphene film. We further study the microscopic origin of the thermal conductivity κ enhancement in the graphene sheets by calculating the phonon relaxation time. The phonon relaxation time τ indicates the time needed by a phonon mode to relax back to equilibrium due to phonon-scattering mechanisms. The phonon dispersion of the supported graphene film for $\rho = 0.081 \text{ nm}^{-2}$ and supported graphene layer number $l_G = 2$, and the calculated relaxation time for all the phonon modes are shown in Fig. 4. By inserting the APTES molecule, the relaxation time of the acoustic flexural modes largely increase at low frequencies, whereas the longitudinal and transverse modes undergo a slight decrease in their relaxation times. The notable increase in accounts for

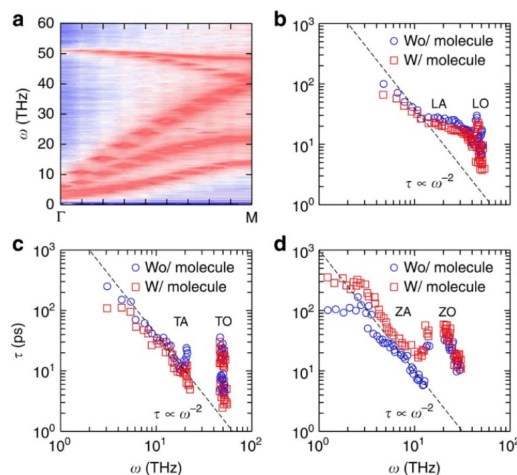


FIG. 4. Phonon dispersion and lifetimes of the graphene film. (a) Phonon dispersion of the graphene film bonded to the APTES-FGO substrate from molecular dynamics simulations, for $\rho = 0.081 \text{ nm}^2$ and $l_G = 2$. (b-d) Mode-wise phonon relaxation time for longitudinal modes including longitudinal acoustic (LA) and optical (LO) branches, for transverse modes including transverse acoustic (TA) and optical (TO) branches, and for flexural modes including flexural acoustic (ZA) and optical (ZO) branches, respectively. Reproduced with permission from Nat. Commun. 7, 11281 (2016). Copyright 2016 Springer Nature.

the enhancement τ of the flexural modes of the graphene sheet bonded to the substrate.

The detailed phononic and electronic transport properties of the APTES molecule to disclose its role in the cross-layer heat conduction have also been investigated. It has been found out that low-energy phonons are mainly responsible for heat conduction due to van der Waals (vdW) interlayer force, whereas high-frequency phonons propagates along the covalent bonds in the molecules.

Finally, the supported graphene is integrated in a micro heater to demonstrate its capacity of improving thermal management. The functionalized graphene oxide introduces alternative heat-escaping channels into a graphene-based sheets bonded to functionalized graphene oxide through amino-silane molecules. Using a resistance temperature probe for in situ monitoring we manage to evidence that the micro heater surface temperature was lowered by $\sim 28^\circ\text{C}$ for a chip operating at $1,300 \text{ W cm}^{-2}$. Thermal resistance probed by pulsed photothermal reflectance measurements demonstrated an improved thermal coupling due to functionalization on the graphene-graphene oxide interface.

4. CHALLENGES AND PERSPECTIVES

4.1 Perspectives on Radiation

Understanding the mechanisms of near-field radiation at the nanoscale is beneficial for improving a variety of technologies ranging from thermal informatics, radiative cooling, energy conversion and non-invasive imaging. Current theoretical and experimental investigations are still behind the essence of extreme near-field heat transfer (gaps below a few nanometer). Theoretically, the validity of the widely used theory based on fluctuational electrodynamics is still questionable to describe the heat transfer behaviors at such small gaps, at which non-locality of the electromagnetic field becomes non-local. The atomistic Green's function method can be helpful to describe the phonon tunnelling phenomenon under nanoscale gaps. It is an ideal method to study the crossover between radiation and heat conduction. However, the atomistic Green's function method relies on the accurate description of interatomic force constants across the gap. The full description of interatomic interactions under different distances is still unclear. Typically, chemical bond interactions is considered for distances comparable to interatomic spacing in crystals [27] while van der Waals forces and/or Coulomb forces are considered for distances from a few nanometers to dozens of nanometers [19, 22]. The interatomic forces decrease rapidly with distances and the relative numerical errors could be large for force constant matrices at large distances, which can bring numerical instabilities in Green's function calculations. Another possible way to study the heat transfer in nanoscale gaps is the atomic molecular dynamics (MD) simulations [28]. Similar to the atomistic Green's function method, the value predicted by MD also relies on the accuracy of interatomic forces. As a result, it is essential to develop unified interatomic forces models at different distance ranges. It is worth to note that the near-field radiation is a classical description of heat transfer. Recently, it was also demonstrated that the Casimir force, which is originated from the vacuum fluctuations and thus is a quantum effect, can also assist phonon tunneling in moderate distances [29, 30]. The quantum effect assisted heat transfer in ultra-short gaps is a new mechanism and is also a new direction.

Although the experimental measurements of near-field radiation at nanoscales has been pushed to the gaps of sub-nanometers with the recent development of fabrication technology, the challenges still remain for accurate measurements with gaps from $\sim 100 \text{ nm}$ to a few Ångströms. First, the large attraction forces between objects separated by subnanometers gaps subject to the residual electrostatic charges makes it difficult to create and robustly maintain such gaps.

Second, the near-field flux is usually in the orders of the pico Watt, which makes it difficult to be detected with high accuracy. Third, both the substrate and probe tip could be easily contaminated by adsorbing unwanted particles, which will largely affect the heat flux and restrict the smallest achievable gap size. The contamination of unwanted particles could also lead to misleading results. Fourth, to fully characterize the heat transfer from near-field radiation to conduction, it is required to measure heat transfer with gaps sizes from hundreds of nanometers to a few Ångströms. For small distances, sharp tips are required to perform well defined measurements. However, it is hard to realize high sensitivity required in measuring the small heat flux at large distances with sharp tips. Consequently, the future experiments on near-field radiation in nanoscale gaps might focus on the solutions of the above challenges, especially the sensitivity and tip shape issues.

4.2 Perspective on Heat Conduction

In so far as heat conduction in nanogaps is in relation with the developments of the several decades old field of energy transfer in molecular systems—which is an above highlighted topic-, the perspectives of expansion are multiple when crossing purposes such as heat exchange, management and conversion with the immense toolbox of available chemical compounds. The second more recent and active field involved is the one of 2D-Materials, which are by a predominant part, constituted of Van der Waals systems. Combining the family of 2D-material polymorphs with the one of existing intercalates is a promising strategy to multiply the range of systems and uncover unexpected thermophysical properties.

5 CONCLUSIONS

This perspective had the ambition to highlight the considerable range of domains from physics involved in the description of energy transfer in a simple gap. At slit sizes larger than a micron, radiative transfer equation is well-known from engineers, but Maxwell equations have to be solved below the micron. At one or two nanometers, Maxwell equations start to become irrelevant due the non-locality of the field -the local dielectric constant is not defined anymore-, and below, the scale of the electron cloud is reached, the classical charge description has to be replaced by the quantum one. At this point, a gap appears as a defect in a quasi-continuous crystal as for instance in 2D-materials or layered bulk materials -among which a large set of thermoelectric materials can be found-. While the fundamental conceptual frames are available, several experimental proofs have still to be im-

plemented, which will certainly be the focus of coming and exciting scientific investigations.

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DATA AVAILABILITY

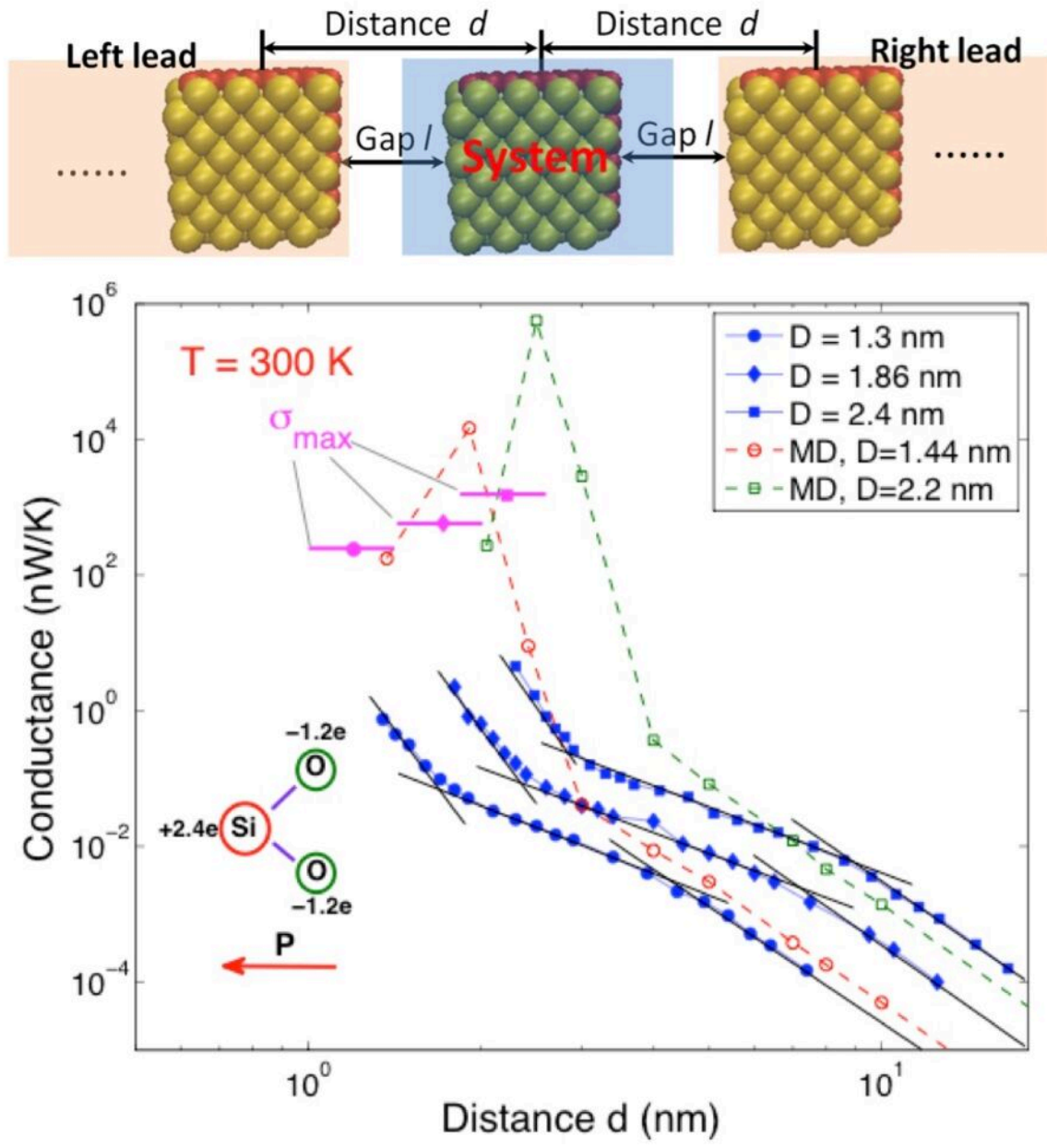
Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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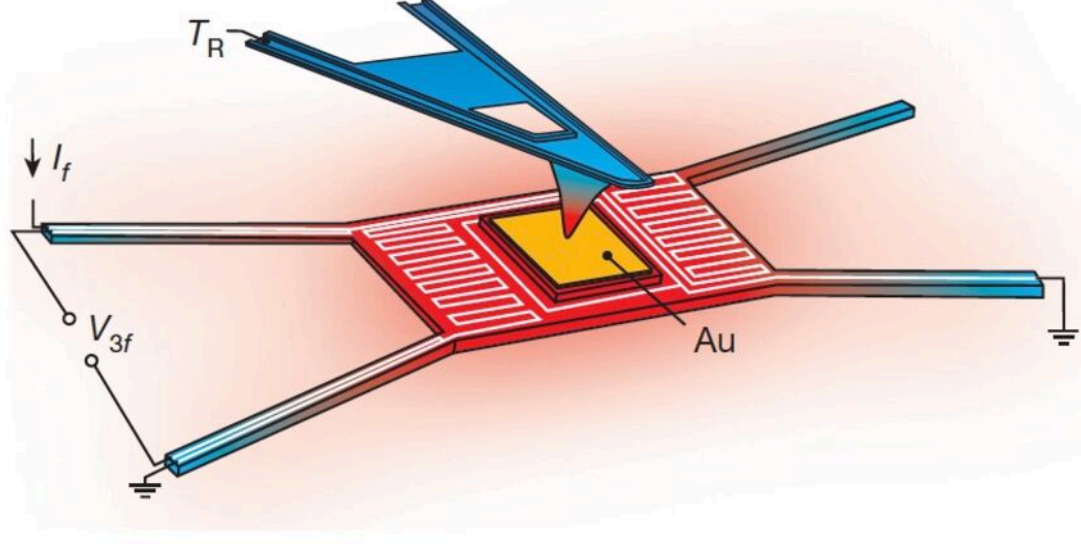
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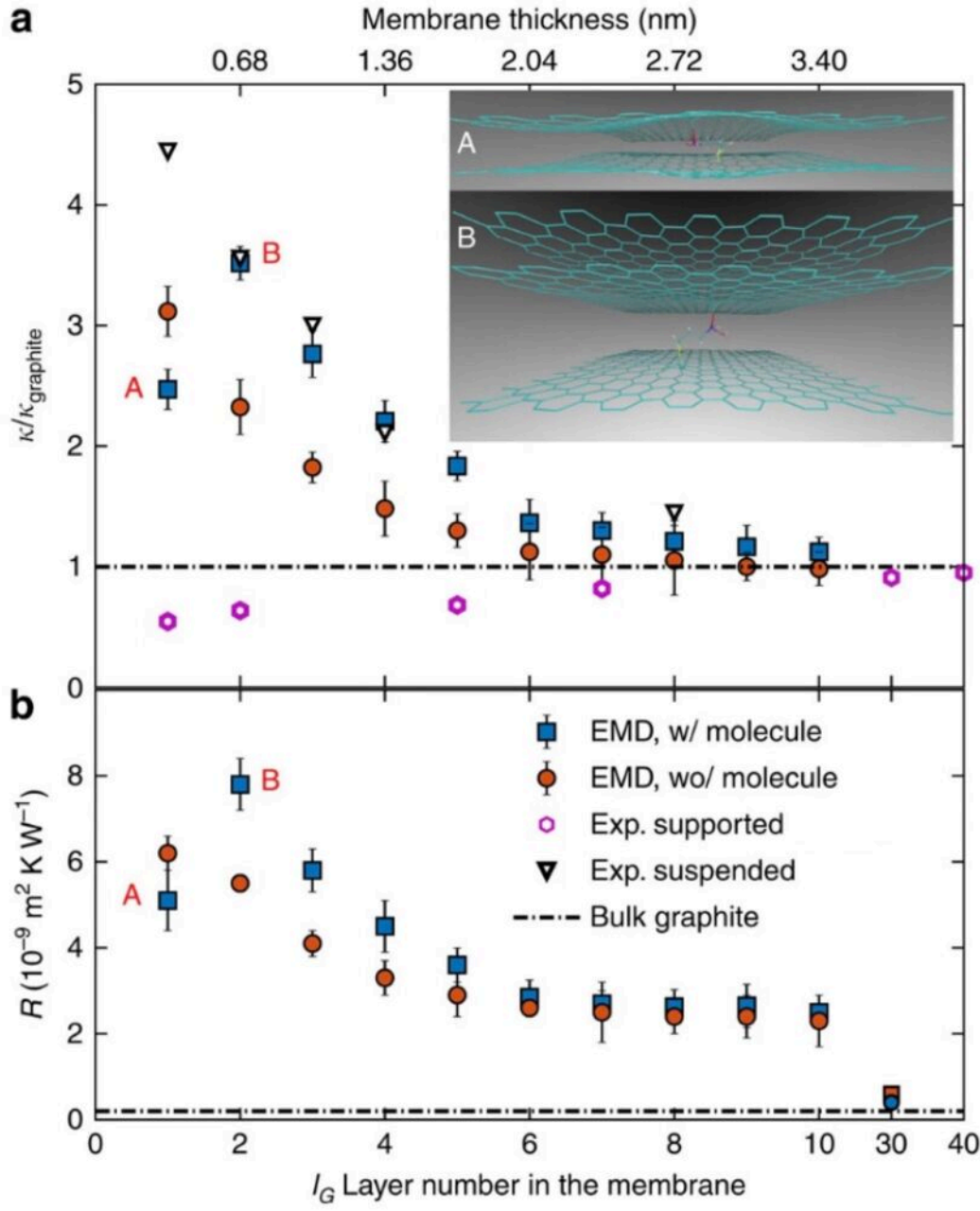
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