Microscopic view of extreme near field heat transfer

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Heat transfer in the extreme near field has shown recently an increased interest driven by the development of scanning probe techniques allowing one to measure thermal transport across a nanoscale gap and across single molecule junctions. On the one hand, recent experimental studies reported giant heat flux transfer between gold surfaces separated by nanometer distances [1,2]. These results showed discrepancies with the conventional theory which are attributed to phonon tunneling neglected in the standard picture. On the other hand, the thermal conductance of single molecule junctions has been recently determined experimentally [3,4] opening the door to confirm the high thermoelectric efficiency displayed by these junctions. All these experiments call for models capable of describing accurately phonon tunneling and phonon thermal transport across single molecules.

In this contribution, we model heat transfer across nanometer gaps and across single-molecule junctions using a combination of molecular dynamics and ab-initio calculations. First, we demonstrate that phonons dominate heat transport at nanometer distances, even in the presence of molecules in the gap [5-7]. We use these results to interpret recent experiments [1-2,6,7]. We also discuss extreme near field heat transfer across gaps between polar materials, and unveil limitations of fluctuational electrodynamics theory [7]. In particular, both acoustic tunneling and non-local dielectric response, not accounted for in fluctuational electrodynamics, are shown to become relatively important for tiny gaps [7].

The second issue to be covered concerns the thermal transport and thermoelectric properties of single molecule junctions. We focus on junctions made of OPE3 derivatives, a single molecule commonly synthesized by experimentalists and which electronic transport properties have been extensively investigated. We first demonstrate that first-principles calculations yield values of figure of merit in good agreement with experiments [8]. We also show that these molecule junctions may realize Peltier cooling by applying a bias voltage in a three-gate terminal device [9]. We discuss how to reach optimal conditions amounting to nanoWatt cooling powers [9]. We eventually show that cross–linking OPE3 is a promising strategy to enhance the figure of merit ZT of the molecule junction. The increase of ZT is shown to be controlled by the broadening and shift of the lowest unoccupied molecular orbital (LUMO) level of the molecule induced by cross-linking [10]. These results unveil an innovative strategy to improve the thermoelectric properties of molecule junctions.

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