FIRST-PRINCIPLES MODELING OF INELASTIC TRANSPORT IN ATOMIC GOLD WIRES

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The rapid evolution in electronics towards increasingly smaller and faster devices will eventually reach the fundamental level set by the atomistic structure of matter. Atomic-size conductors take this development to the extremity of miniaturization [1], and understanding their properties is an important problem in the emerging fields of nanoelectronics and molecular electronics. One relevant aspect is the study of the effects caused by atomic vibrations, since inelastic scattering of traversing electrons and energy dissipation play essential roles for device characteristics, working conditions, and stability. Also, vibrational signals can be used to extract information about the detailed microscopic configuration, since this cannot usually be imaged simultaneously with a transport measurement.

Inelastic effects have in the recent years been observed in a variety of nanoscale systems, including single molecules on surfaces probed with the scanning tunneling microscope (STM) e.g. [2], molecules in break-junctions e.g. [3], and metallic atomic wires [4]. In the latter example, Agraït and co-workers used a cryogenic STM to create a freestanding atomic gold wire between tip and substrate and, further, to perform point-contact spectroscopy measurements. The observed spectra displayed symmetric drops in the conductance at threshold voltages characteristic for phonons, and were found to be very sensitive to the atomic configuration.

In this work we use a method recently developed in our group for including inelastic scattering in a first-principles density-functional theory (DFT) computational scheme for electronic transport, to investigate in detail the inelastic transport through atomic gold wires [5]. We systematically analyze both linear and zig-zag wires of different length, and under different states of strain and contact arrangement. Two generic setups that we consider are depicted in Fig. 1.

The investigation of each structure includes calculations of the vibrational modes and couplings, as well as of the nonlinear differential conductance vs. bias voltage (G-V) under various conditions of external damping of the modes. Typical GV curves calculated for a four-atom wire are shown in Fig. 2a, which displays characteristic features of the experiment, namely the symmetric drop around one particular phonon frequency, as well as a finite slope beyond the threshold. Our theory further explains the experimentally observed selection rule as related to modes with “alternating bond length” character. Fig. 2b shows the non-equilibrium phonon occupation of the dominant scattering mode. As the bias exceeds the energy required for excitations of the vibration, the mode starts to heat up. This population of the mode is found to grow linearly with voltage.

From this study we learn in detail about the relation between microscopic arrangements of the atoms and the phonon spectrum as revealed by a transport measurement. We further extract much quantitative information from the theory, such as conductance changes, frequency shifts with strain, and slope in conductance.

References:

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Figures:

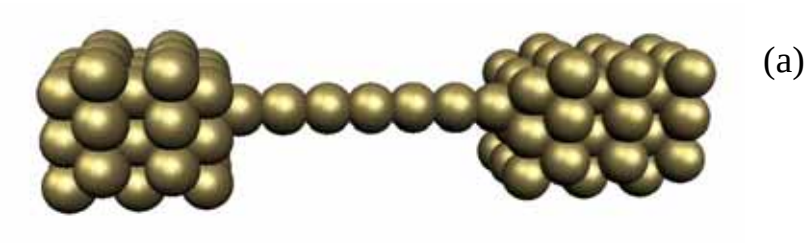

Figure 1: Sample geometries of (a) a linear and (b) a zig-zag gold wire. The system here consists of 7 gold atoms arranged between Au-(100) surfaces in a 3x3 unit cell.

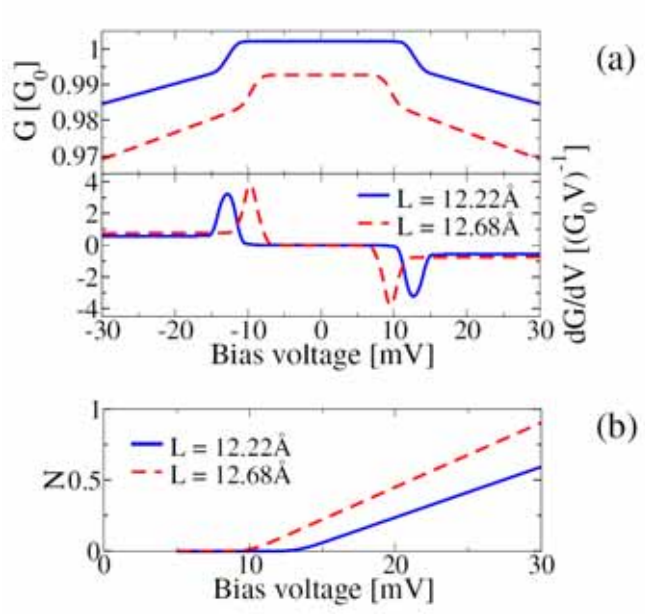

Figure 2: Calculation of (a) differential conductance and its derivative and (b) phonon heating for a four-atom gold wire at two different tensions.