Electron-Vibration Interactions And Transport In Nano-Contacts From Density Functional Theory

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In molecular electronics, the inelastic scattering of traversing electrons and energy dissipation play essential roles for device characteristics, working conditions, and stability. The signatures of interactions with vibrations have been observed in the current-voltage characteristics of atomic-scale conductors [1]. These vibrational signals may be used to extract information about the detailed atomic configuration – information which is usually not accessible simultaneously with transport measurements.

In this talk we discuss theoretical work on inelastic transport due to interaction with atomic vibrations in atomic-scale conductors, which are strongly coupled to electrodes. Our results are obtained by combining density functional theory with non-equilibrium Green's function (NEGF) techniques [2]. The inelastic effects are taken into account using the self-consistent Born approximation (SCBA) [3]. While the full NEGF-SCBA calculations are computationally demanding, we show how these equations can be approximated by a simple lowest order expansion (LOE). The LOE yield excellent agreement with our full SCBA results and may be justified in many cases [4].

To illustrate our methods we present extensive calculations on atomic gold wires with different lengths and strain, where we can compare the calculated inelastic signals in the current-voltage characteristics to results from detailed experiments, see Fig. 1a.
We show how the one-dimensional gold wire systems may react with "impurities" such as hydrogen and discuss how such structures may be revealed by the inelastic signals [5]. We also address the vibrational signatures in the current of organic molecular contacts [6], and discuss 'propensity rules', which may be derived from the electronic scattering states. We calculate the power transferred to the vibrations by the interaction from the electrons and the corresponding damping of the vibrations due to electron-hole pair excitation. We further characterize the corresponding signatures of local heating in the conductance.

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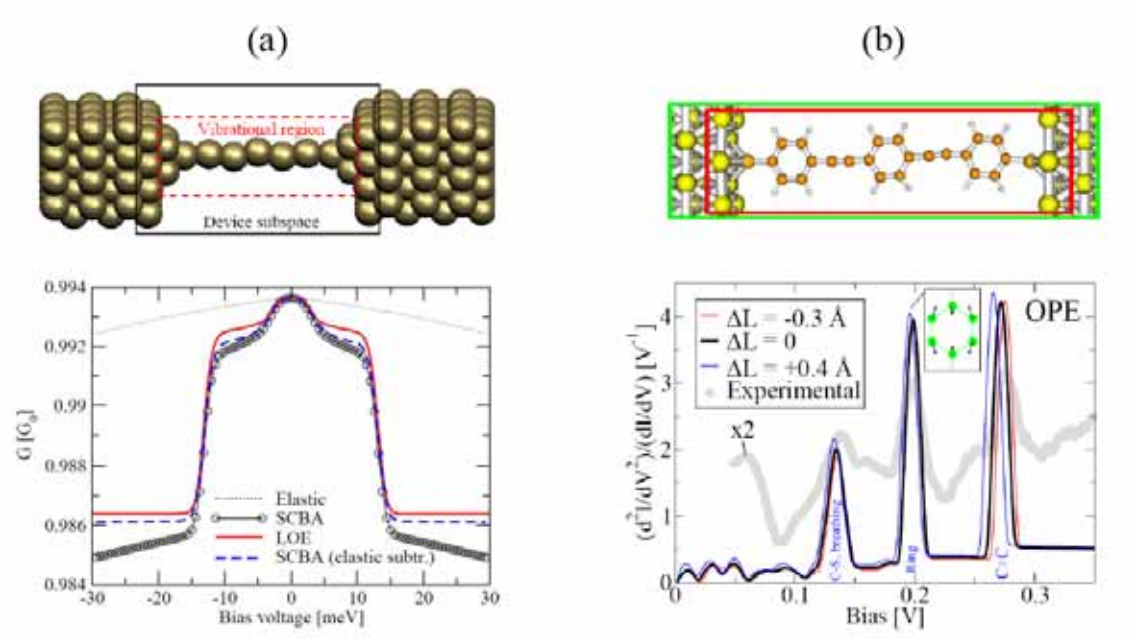

Figure 1a: Calculation of the inelastic signal in conductance vs. bias voltage in a 7-atom long gold chain connected to gold electrodes (top). A decrease in conductance is observed at the on-set of vibrational excitationFigure 1b: The Inelastic Electron Tunnel Spectrum (IETS) calculated for the OPE molecule (top) thiol-bonded to gold electrodes. Only 3 main vibration modes (peaks) are observed in accordance with the experiments (thick curve) [7].