5.6 Thermoelectric and inelastic effects in charge transport through atomic and molecular contacts

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**Goals**

Our research interests are focused on the understanding of charge transport on the molecular scale. Beside elastic conduction, we investigate the inelastic contributions due to electron-vibration coupling and thermoelectric properties.

Our studies include:
- Heat dissipation in the electrodes,
- thermoelectric properties,
- heat conduction by electrons and phonons,
- inelastic electron tunneling spectra (IETS).

**Results**

**Heat dissipation in atomic scale junctions [1]**

Power dissipated in the probe (P) and the substrate (S),
\[ Q_P(V) = \frac{2}{\hbar} \int_{-\infty}^{\infty} dE \left( \mu_P - E \right)^\alpha (E, V) |e - f_s| \]
\[ Q_S(V) = \frac{2}{\hbar} \int_{-\infty}^{\infty} dE \left( \mu_S - E \right)^\alpha (E, V) |e - f_s| \]

A low-temperature, low-bias expansion shows that a heating asymmetry with respect to the bias or the electrodes results from a non-zero thermopower or, more generally, an electron-hole asymmetry.

\[ Q_P(V) - Q_P(-V) \approx 2GTSV + O(V^3) \]
\[ Q_P(V) - Q_S(V) \approx 2GTSV + O(V^3) \]

Using that \( Q_{\text{Total}} = Q_P + Q_S \) and \( Q_{\text{Total}} \approx GV^2 \), one can show that [2]:
\[ Q_P(Q_{\text{Total}}) \approx \frac{1}{2} Q_{\text{Total}} \pm \frac{\sqrt{\pi}}{8} \frac{e^{1/2}}{b \sqrt{G}} \]

where the minus (plus) sign holds for positive (negative) bias voltage. The expression predicts that the voltage-related asymmetry in the power dissipation in the probe \( Q_P(Q_{\text{Total}}) \), expressed through the total dissipated power, should be rather independent of the level alignment, but sensitive to the level broadening.

**Heat transport due to electrons & vibrations**

Our phonon transport scheme uses the harmonic approximation and force constants determined by DFT [3].

\[ H = \frac{1}{2} \sum_i p_i^2 + \frac{1}{2} \sum_i \sum_j q_i K_{ij} q_j \]

Extracting force constants from large clusters, the calculated phonon DOS of bulk gold compares very well with measurements.

The phonon heat conductance is:
\[ \kappa_{\text{ph}} = \frac{1}{\Delta T} \int \frac{dE}{\pi} \frac{\partial P(E, \mu)}{\partial E} \]

Here, \( \partial P(E, \mu) / \partial E \) is the Bose function. Phonon as compared to electron heat transport plays an important role in molecular junctions.

\[ \kappa_{\text{ph}} = \frac{1}{\Delta T} \int \frac{dE}{\pi} \frac{\partial P(E, \mu)}{\partial E} \]

We used the integrals:
\[ K_0 = \int dE \kappa_{\text{ph}}(E) \frac{dE}{\pi} (E - \mu) \]

**Transport through paracyclophane molecules**

We analyzed charge transport properties of paracyclophane molecules. This contributes to the understanding of transport properties through π-stacked systems.

**Findings:**
- Transmission eigenchannels show that the transport is mostly carried by benzene π-orbitals and not by the alkane “clips”.
- Conductance decays exponentially with length.
- Thermopower increases linearly with the molecular length.
- The peaks in the IETS shift for different anchor groups.

**References**

