Transport through a vibrating quantum dot: Polaronic effects

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1 Motivation

Recent progress in nanotechnology allows the fabrication of electronic devices with the active element being a single organic molecule. Molecular electronics may constitute an alternative to conventional semiconductor technology, while their complex structure introduces new electronic transport properties. Such a system can be described as a quantum dot i.e. a system of finite size, weakly coupled to macroscopic charge reservoirs and so small, that quantisation of energy levels becomes important.

The molecule is susceptible to structural changes when being occupied by charge carriers. These deformations can be of substantial energy and may heavily alter the transport properties of the device.

2 Model and method

The deformation of the quantum dot is represented by coupling to a local dispersionless vibrational mode: \[ \Delta H_{\text{QD}} = \Delta \omega \phi^2 - g \phi \nabla \cdot \mathbf{d} \] In it is contact with two macroscopic, non-interacting metallic leads \((e, \ell)\).

\[ \Delta H_{\text{QD}} = \sum_{k} \left( \frac{\epsilon_{k}^{2}}{2} - \frac{\epsilon_{k}^{2}}{2} \right) \phi_{k} + g \phi \nabla \cdot \mathbf{d} \]

We assume semi-infinite leads with \( W \) being the bandwidth of the DOS.

\[ \rho(\omega) = \frac{1}{\pi} \sum_{k} \delta(\omega - \epsilon_{k}) = \frac{1}{\pi \sqrt{W^{2} - \omega^{2} (W - \omega)}^{2}} \]

For sufficiently large electron-phonon (E-phonon) coupling \( g \) and phonon frequencies \( \omega_{0} \), we expect the formation of a polaron-like state at the dot, where the gain in potential energy compensates a kinetic energy loss.

To account for this process, we apply a generalised Landau-Finsler transformation [cf. A. La Magna, I. Doretto, Private communication, 2007] \( \omega \rightarrow \omega_{0} \) in terms of the retarded electronic NEGF of the dot.

\[ \omega_{R} \approx \omega_{0} - \frac{i g}{\pi} \left( \frac{\partial \rho(\omega)}{\partial \omega} \right) \]

As \( \gamma \rightarrow 1 \), the direct E-phonon coupling vanishes at the cost of a phonon-affected transfer. In this way, our ansatz allows the interpolation between weak and strong coupling for moderate to large frequencies \( \omega_{0} \).

We base our calculation on the equations of motion of generalised temperature Green functions [Kadanoff, Baym, Benjamin/Cumming Publishing Co., 1962],

\[ \mathcal{G}_{\text{QD}}(\omega_{1}, \omega_{2}) = \sum_{\lambda} \mathcal{G}_{\lambda}(\omega_{1}, \omega_{2}) \mathcal{G}_{\lambda}^{*}(\omega_{2}, \omega_{1}) \]

where the mean value and the time dependences are deterministic, \( \mathcal{G}_{\lambda}(\omega) \).

In low temperature approximation we obtain

\[ L = \epsilon_{\text{QD}} + \frac{1}{\pi} \int \frac{d\omega}{\sqrt{W^{2} - \omega^{2} (W - \omega)}} \]

The self-consistent determination of the variational parameter is essential, once \( \gamma_{\text{pol}} \) is known, a modified renormalisation scenario gives a good approximation for \( L \).

4.1 Adiabatic regime

If the phononic timescale is much slower than the electronic timescale \( \tau_{\text{pol}} \approx 1 \), the deformation of the dot adjusts quasi-statically to the occupation.

For a repulsive dot \((\Delta < 0)\) and increasing EP-coupling, we find an abrupt transition related to a jump \( \Delta \rightarrow 0 \) at a critical EP-coupling strength \( \gamma_{\text{pol}} \)

\[ \approx \frac{\left| \Delta \right|}{\gamma_{\text{pol}}} \]

- localised states + continuum around \( \omega = 0 \)
- small \( \gamma_{\text{pol}} \) = reduced \( L \)
- quasi-localised polaron-like state + side bands
- strongly reduced \( \Delta \) and renormalised transfer integral \( \Delta \rightarrow L \) vanishes

4.2 Nonadiabatic regime

Here the phononic timescale is much faster than the electronic timescale \( \tau_{\text{pol}} \approx 1 \), so that the deformation adjusts instantaneously to the electronic occupation.

Now a continuous transition occurs, with \( \gamma_{\text{pol}} \rightarrow 1 \).

- for \( \gamma_{\text{pol}} \approx \Delta \), \( \Delta(\gamma_{\text{pol}}) \approx 1 \) DOS
- \( \approx \frac{\left| \Delta \right|}{\gamma_{\text{pol}}} \)
- reduced \( \gamma_{\text{pol}} \) = reduced \( L \)
- polaron-like bound state for \( \gamma_{\text{pol}} > \Delta \)
- \( L \) can be reproduced by non-interacting model using renormalised \( \gamma_{\text{pol}} \)

4.3 Intermediate regime

Our approach allows the investigation of the intermediate case where phononic and electronic timescales become comparable \((\gamma_{\text{pol}} < 1)\).

- jump-like or smooth transition, depending on \( \Delta \)
- few overlapping phononic bands
- no localised state even for large \( \gamma_{\text{pol}} \)
- renormalisation argument with \( \gamma_{\text{pol}} < 1 \) fails to describe transport; better agreement when using \( \gamma_{\text{pol}} \)

5 Conclusions

We followed a variational approach, that extends the description of the polaron problem away from the limit of large phonon frequencies, to derive the basic polaronic effects at the quantum dot

- formation of a polaron-like state
- rapid transition resulting in a sudden conductance drop in the adiabatic regime
- phonon-assisted transport through a tunnelling barrier in the adiabatic regime

![Fig. 4: The quantum dot as a molecular switch](image)

**Outlook**

Let’s keep the EP-coupling fixed and tune the quantum dot level \( \epsilon_{\text{QD}} \approx 0.5 \).