Transport through a vibrating quantum dot: Polaronic effects

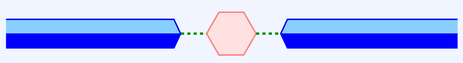
T. Koch[1], J. Loos[2], A. Alvermann[1], A. R. Bishop[3], H. Fehske[1]

[1]Universität Greifswald, [2]Czech Academy of Science Prague, [3]Los Alamos National Laboratory

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

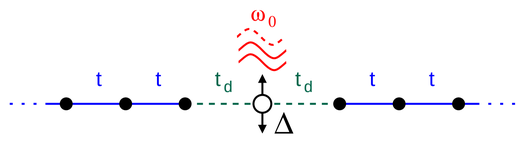
quantum dot



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:

$$H_{QD} = \Delta d^\dagger d \;\; - g\omega_0 d^\dagger d(b^\dagger + b) + \omega_0 b^\dagger b \; .$$

It is in contact with two macroscopic, non-interacting metallic leads ($a = l, r$),

$$H_L = \sum_{k,a} E_k c^\dagger_{ka} c_{ka} - \frac{t_d}{\sqrt{N}} \sum_{k,a} \left(d^\dagger c_{ka} + c^\dagger_{ka} d \right) \; ,$$

We assume semi-infinite leads with $W = 2t$ being half the bandwidth of their DOS:

$$\varrho(\xi) = \frac{1}{N} \sum_k \delta(\xi - E_k) = \frac{2}{\pi W^2} \sqrt{W^2 - \xi^2}\, \Theta(W^2 - \xi^2) \; .$$

For sufficiently large electron-phonon-(EP)-coupling g and phonon frequencies ω_0, we expect the formation of a polaron-like state at the dot, when the gain in potential energy compensates a loss in kinetic energy.

To account for this process, we apply a generalised variational Lang-Firsov transformation (cf. [A. La Magna, I. Deretzis, PRL **99**, 136404 (2007)])

$$U = e^{\widetilde{g}(b^\dagger - b) d^\dagger d} \; , \quad \widetilde{g} = \gamma g \; , \quad \gamma \in [0, 1] \; .$$

We then arrive at $\widetilde{H} = U(H_{QD} + H_L)U^\dagger$ where

$$\widetilde{H} = \sum_{k,a} E_k c^\dagger_{ka} c_{ka} - \sum_{k,a} \left(C_t\, d^\dagger c_{ka} + C_t^\dagger c^\dagger_{ka} d \right) + \widetilde{\Delta}\, d^\dagger d \;\; - C_d d^\dagger d + \omega_0 b^\dagger b \; ,$$

with $\varepsilon_p = g^2\omega_0$, $\widetilde{\Delta} = \Delta - \varepsilon_p\gamma(2 - \gamma)$ and the new interaction coefficients

$$C_t = \frac{t_d}{\sqrt{N}} e^{-\widetilde{g}(b^\dagger - b)} \; , \quad C_d = g\omega_0(1 - \gamma)(b^\dagger + b) \; .$$

As $\gamma \to 1$, the direct EP-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 ω_0.

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

$$G_{dd}(\tau_1, \tau_2; \{V\}) = -\frac{1}{\langle S \rangle} \langle \mathcal{T}_\tau d(\tau_1) d^\dagger(\tau_2) S \rangle \; ,$$

where the mean value and the time dependences are determined by $\widetilde{H} - \mu\hat{N}$. The S-matrix

$$S = \mathcal{T}_\tau \exp\left\{ - \int_0^\beta \mathrm{d}\tau\, V_t(\tau) C_t(\tau) + \bar{V}_t(\tau) C_t^\dagger(\tau) + V_d(\tau) C_d(\tau) \right\}$$

describes the coupling to the components of a fictitious external potential $\{V\}$.

Quantities of interest

- single particle spectrum at the dot
- electronic current through the dot

Both are determined by the polaronic spectral function $A_{dd}(\omega)$

3 Theoretical results

3.1 Spectral function

Based on the equations of motion, e.g.

$$\left[-\frac{\partial}{\partial\tau_1} - (\widetilde{\Delta} - \mu) \right] G_{dd}(\tau_1, \tau_2; \{V\}) = \delta[\tau_1 - \tau_2] + \frac{1}{\langle S \rangle} \langle \mathcal{T}_\tau C_d(\tau_1) d(\tau_1) d^\dagger(\tau_2) S \rangle + \sum_{k,a} \frac{1}{\langle S \rangle} \langle \mathcal{T}_\tau C_t(\tau_1) c_{ka}(\tau_1) d^\dagger(\tau_2) S \rangle \; ,$$

we express the polaronic self energy

$$\Sigma_{dd}(\tau_1, \tau_2; \{V\}) = G^{(0)-1}_{dd}(\tau_1, \tau_2) - G^{-1}_{dd}(\tau_1, \tau_2; \{V\})$$

by its functional derivatives with respect to $\{V\}$. In an iterative scheme we evaluate the self energy up to second order in the interaction coefficients $\langle C_t(\tau) \rangle$.

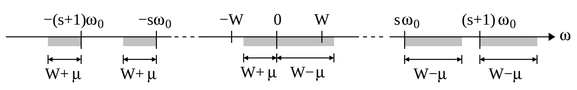
We then let $\{V\} \to 0$ and calculate the correlation functions of the interaction coefficients assuming independent Einstein-oscillators. After Fourier-transformation and summation over bosonic Matsubara frequencies, we arrive in low temperature approximation ($\beta\omega_0 \gg 1$) at

$$\Sigma^{(2)}_{dd}(z) = 2\, t_d^2\, e^{-\widetilde{g}^2} \int_{-W}^{W} \mathrm{d}\xi\, \varrho(\xi) \left[\frac{1}{z - (\xi - \mu)} + \sum_{s \geq 1} \frac{(\widetilde{g}^2)^s}{s!} \left(\frac{n_F(\xi - \mu)}{z - (\xi - \mu) + s\omega_0} + \frac{1 - n_F(\xi - \mu)}{z - (\xi - \mu) - s\omega_0} \right) \right] + [(1 - \gamma) g\omega_0]^2 \int_{-\infty}^{+\infty} \mathrm{d}\omega' A^{(1)}_{dd}(\omega') \left(\frac{n_F(\omega')}{z - \omega' + \omega_0} + \frac{1 - n_F(\omega')}{z - \omega' - \omega_0} \right)$$

with $\Sigma^{(1)}_{dd}(z)$ being the first two terms in $\Sigma^{(2)}_{dd}(z)$. This self energy accounts for multi-phonon processes and finite particle densities. The spectral functions follow, with $\delta \to 0^+$, in an iterative way from

$$A^{(n)}_{dd}(\omega) = \frac{1}{\pi} \frac{-\mathrm{Im}\, \Sigma^{(n)}_{dd}(\omega + \mathrm{i}\delta)}{\left[\omega + \mu - \widetilde{\Delta} - \mathrm{Re}\, \Sigma^{(n)}_{dd}(\omega + \mathrm{i}\delta) \right]^2 + \left[\mathrm{Im}\, \Sigma^{(n)}_{dd}(\omega + \mathrm{i}\delta) \right]^2}$$

For finite EP-coupling, the spectrum contains multiple phonon side bands. They overlap along the whole ω-axis if $\omega_0 < W - |\mu|$. Otherwise a quasiparticle state may exist in the intervals where $\mathrm{Im}\, \Sigma_{dd}(\omega) = 0$.



3.2 Current

In [Meir, Wingreen, PRL **68**, 2512 (1992)], a Landauer-type formula was derived, that expresses the current J for finite voltage $\Phi = -(\mu_l - \mu_r)/\mathrm{e}$ in terms of the retarded electronic NEGF of the dot:

$$J = -\mathrm{e}\, t_d^2 \int_{-W}^{W} \mathrm{d}\xi\, \varrho(\xi)\, \mathrm{Im}\, G^R_{dd}(\xi) \times \left[n_F(\xi - \mu_l) - n_F(\xi - \mu_r) \right] \; .$$

For low source-drain-voltage, i.e. $\mu_{l,r} = \mu \pm \delta\mu/2$, we express the current as $J = -L\delta\mu/\mathrm{e}$, so that the linear conductance $L = \lim_{\delta\mu \to 0} \{-\mathrm{e}J/\delta\mu\}$ is

$$L = \mathrm{e}^2 \pi\, t_d^2 \int_{-W}^{W} \mathrm{d}\xi\, \varrho(\xi) \left[-n'_F(\xi - \mu) \right] A^e_{dd}(\xi - \mu) \; .$$

A relation between the electronic and polaronic spectrum has been established in [Loos, Hohenadler, Fehske, J. Phys.: Cond. Mat. **18**, 2453 (2006)]:

$$A^e_{dd}(\omega) = e^{-\widetilde{g}^2} \sum_{s \geq 0} \frac{(\widetilde{g}^2)^s}{s!} \Big[A_{dd}(\omega - s\omega_0)\Theta(\omega - s\omega_0) + A_{dd}(\omega + s\omega_0)\Theta(-\omega - s\omega_0) \Big] \; .$$

In low temperature approximation we obtain

$$L = \mathrm{e}^2 \pi\, t_d^2\, e^{-\widetilde{g}^2} \varrho(\mu) A_{dd}(0) \; .$$

In addition to the Lang-Firsov-renormalization of the transfer integral, the conductance depends on the accessibility of dot-states at the Fermi-level.

4 Numerical results

We determine the extremal variational parameter γ_{min} by minimising the energy

$$E = 2\, t_d^2\, e^{-\widetilde{g}^2} \int_{-W}^{W} \mathrm{d}\xi\, \varrho(\xi) \int_{-\infty}^{+\infty} \mathrm{d}\omega' A_{dd}(\omega') \left[n_F(\omega') - n_F(\xi - \mu) \right] \times \left[\frac{2}{\omega' - (\xi - \mu)} + \frac{\xi - \mu}{(\omega' - (\xi - \mu))^2} \right] + (\widetilde{\Delta} - \mu) \int_{-\infty}^{0} \mathrm{d}\omega' A_{dd}(\omega') \; .$$

Despite being an approximation, $\Sigma^{(2)}_{dd}$ fulfills the sum rule $\int A_{dd}(\omega)\mathrm{d}\omega = 1$. To avoid an artificial violation of this important property when evaluating it numerically, we keep δ in A_{dd} as a small parameter ($\delta \lesssim 5 \cdot 10^{-3}$). In the following, we set $t = 1$.

4.1 Adiabatic regime

If the phononic timescale is much slower than the electronic timescale ($\omega_0 = 0.1 \ll 1$), the deformation of the dot adjusts quasi-statically to the occupation.

For a repulsive dot ($\Delta = 3$) and increasing EP-coupling, we find an abrupt transition related to a jump in γ_{min} at a critical EP-coupling strength ε_p^c.

$\varepsilon_p < \varepsilon_p^c$, $\gamma_{min} \ll 1$

- localised states + continuum around $\omega = 0$
- small $A_{dd}(0) \rightsquigarrow$ reduced L

$\varepsilon_p > \varepsilon_p^c$, $\gamma_{min} = 1$

- quasi-localised polaron-like state + side bands
- strongly reduced $A_{dd}(0)$ and renormalised transfer integral $t_d \rightsquigarrow L$ vanishes

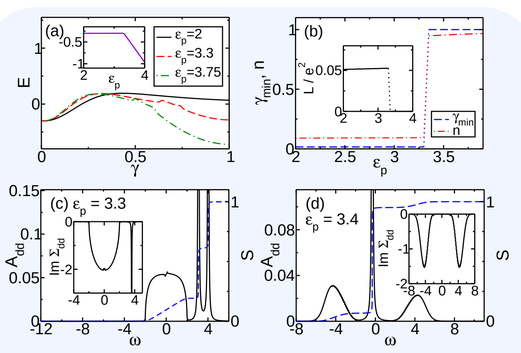


Fig. 1: $\omega_0 = 0.1$, $\Delta = 3$, $t_d = 1$, $\mu = 0$.
(a): Dot energy E as a function of γ. At $\varepsilon_p^c \approx 3.35$ the global minimum jumps. (b): Linear conductance L, minimising γ_{min}, occupation of the dot n as functions of ε_p. (c) and (d): Spectral functions and self energies for ε_p in the vicinity of the transition show the sudden formation of a long-living polaron-like state.

Our variational approach simulates a rapid adiabatic transition – as seen for the one electron case in [Alvermann, Fehske, PRB **77**, 045125 (2008)] – by a sudden change in γ_{min} when a gain in potential energy outweighs the loss in kinetic energy.

4.2 Antiadiabatic regime

Here the phononic timescale is much faster than the electronic timescale ($\omega_0 = 10 \gg 1$), so that the deformation adjusts instantaneously to the electronic occupation.

Now a continuous transition occurs, with $\gamma_{min} \to 1$.

- for $\varepsilon_p = \Delta$: $A_{dd}(\omega) \approx$ 1D DOS $\rightsquigarrow$ phonon-assisted transport, maximum in L
- $\omega_0 > W$, phononic bands do not overlap $\rightsquigarrow$ polaron-like bound state for $\varepsilon_p > \Delta$
- L can be reproduced by non-interacting model using renormalised $\Delta - \varepsilon_p$ and $t_d \exp\{-g^2/2\}$

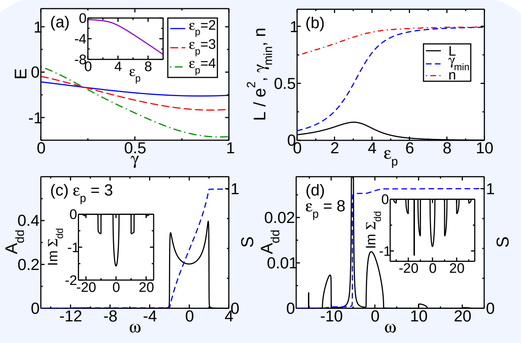


Fig. 2: $\omega_0 = 10$, $\Delta = 3$, $t_d = 1$, $\mu = 0$.
(a): E has a single minimum for all ε_p. Therefore no transition occurs. (b): L exhibits a maximum when $\varepsilon_p = \Delta$ and the spectrum in (c) resembles the DOS of a 1D chain. (d): For $\varepsilon_p > \Delta$, a bound polaron-like state forms where $\mathrm{Im}\, \Sigma_{dd} = 0$. Because $\omega_0 > W$, it can not decay by phonon emission / absorption.

As for the Holstein model, the complete Lang-Firsov transformation is limited to large phonon frequency and strong EP-coupling. By allowing $\gamma_{min} < 1$, our approach accounts for important corrections in the weak coupling domain.

4.3 Intermediate regime

Our approach allows the investigation of the interesting case where phononic and electronic timescales become comparable ($\omega_0 = 1$).

- jumplike or smooth transition, depending on Δ
- few overlapping phononic bands $\rightsquigarrow$ no localised state even for large ε_p
- renormalization argument with $\gamma = 1$ fails to describe transport; better agreement when using γ_{min}

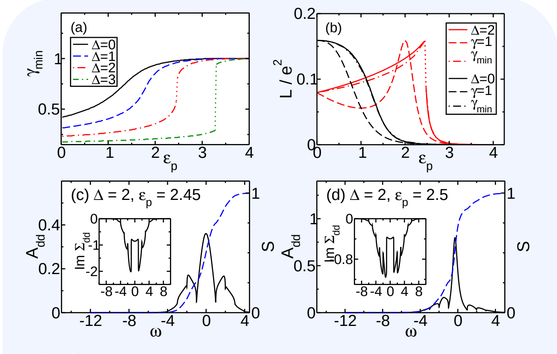


Fig. 4: $\omega_0 = 1$, $\mu = 0$.
(a) When lowering t_d while keeping ω_0 fixed, the disagreement between the numerical results (solid lines) and a simple renormalization scenario (dashed lines) vanishes, indicating a transition to the antiadiabatic regime. (b): Although the transition remains for a repulsive dot, L develops a sharp maximum as $t_d \to 0$, which again signals the antiadiabatic regime. (c) and (d): The strongly suppressed phonon bands give rise to localised states.

The self-consistent determination of the variational parameter is essential; once γ_{min} is known, a modified renormalization scenario gives a good approximation for L.

4.4 Weak dot-lead coupling

The physical picture suggests the adiabaticity of the dot to be determined by the local ratio of ω_0 and t_d. To see this, we reduced t_d by one order of magnitude.

- for $\Delta = 0$: $\gamma_{min} \to 1$ and renormalization picture works $\rightsquigarrow$ signals antiadiabatic regime
- for $\Delta = 3$: still crossover, but pronounced maximum in L at $\varepsilon_p = \Delta \rightsquigarrow$ antiadiabatic regime
- strongly reduced $\mathrm{Im}\, \Sigma_{dd} \propto t_d^2 \rightsquigarrow$ localised states even for overlapping bands

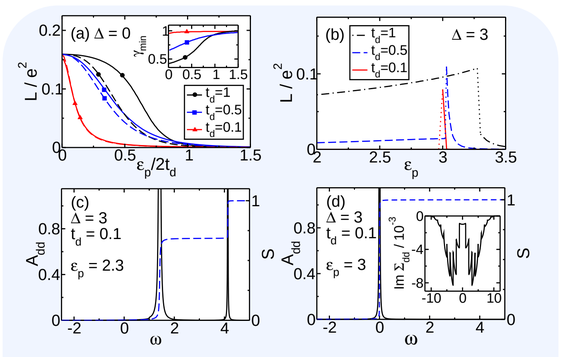


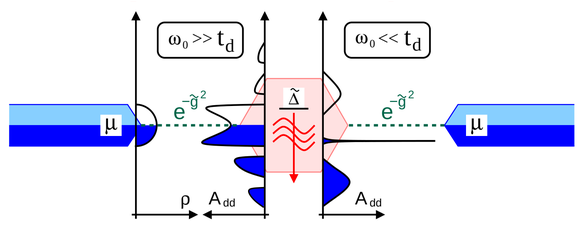
Fig. 4: $\omega_0 = 1$, $\mu = 0$.
(a) When lowering t_d while keeping ω_0 fixed, the disagreement between the numerical results (solid lines) and a simple renormalization scenario (dashed lines) vanishes, indicating a transition to the antiadiabatic regime. (b): Although the transition remains for a repulsive dot, L develops a sharp maximum as $t_d \to 0$, which again signals the antiadiabatic regime. (c) and (d): The strongly suppressed phonon bands give rise to localised states.

The quotient of the phonon frequency and the dot-lead transfer integral distinguishes the adiabatic from the antiadiabatic regime.

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

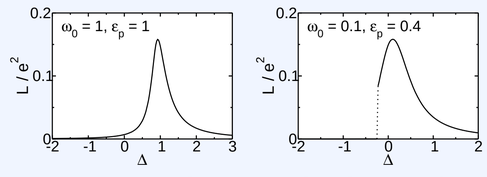
basic polaronic effects at the quantum dot



- 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 antiadiabatic regime

Outlook

Let's keep the EP-coupling fixed and tune the quantum dot level ($t_d = 0.5$, $\mu = 0$):



$\rightsquigarrow$ quantum dot as a molecular switch

[J. Loos, T. Koch, A. Alvermann, A. R. Bishop, H. Fehske, J. Phys.: Cond. Mat. **21**, 395601 (2009)]