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Thermopower for a molecule with vibrational degrees of freedom

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ABSTRACT

We study a simple model to describe resonant tunneling through an organic molecule between two conducting leads, taking into account the vibrational modes of the molecule. We solve the model approximately analytically in the weak coupling limit and give explicit expressions for the thermopower and Seebeck coefficient. The behavior of these two quantities is studied as function of model parameters and temperature. For a certain regime of parameters a rather peculiar variation of the thermopower and Seebeck coefficient is observed.

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1. Introduction

In a recent experiment, Reddy et al. [1] studied transport through an organic molecule (benzenedithiol (BDT)) attached to a scanning-tunneling-microscope (STM) tip and a substrate, generated by a break junction using the STM tip. The authors were able to measure different transport properties, including the thermopower and respectively Seebeck coefficient of this setup, which is is shown schematically in Fig. 1. Quite generally, the idea to contact organic molecules to leads in a controlled fashion is rather fashionable and – if successful – can lead to a variety of interesting applications, ranging from nanoscale computing to quantum computing [2]. First progress of reproducibly and controlled contacting organic molecules has been made using carbon nanotubes [3], but similar attempts with smaller molecules usually suffer from several problems, among them the actual question of how the contact is established and how one can control its quality [4].

It is well known that the physics of nano-objects coupled to conducting leads is highly nontrivial [5], and already concentrating on electronic degrees of freedom only presents an extreme challenge to both experiment and especially theory. The latter is due to the fact that correlations on these nano-structures can usually not be ignored and hence one is plagued with complicated quantum impurity problems [6], which can be solved only numerically and only in equilibrium and for rather restricted model setups. The situation becomes even worse if one wants to take into account vibrational modes present in complex molecules. Here, even advanced numerical tools rather quickly meet their limits [8].

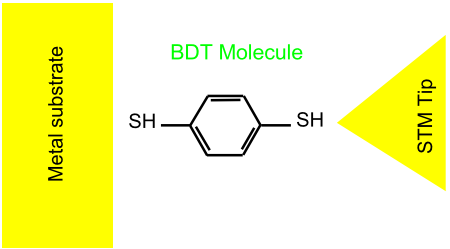

Fig. 1. Schematic experimental setup according to [1]. The STM tip is connected to a reservoir which maintains a constant tip temperature. The substrate, on the other hand, can be heated, thus creating a temperature gradient across the molecule.

And last, but by no means least, the conventional experimental probe used to study nano-systems is transport. To this end one usually applies a bias voltage or, as in the above-mentioned experiment, a temperature gradient across the nano-object. In this situation, the assumption of thermal equilibrium is quickly questionable, and reliable tools to study correlated systems out of thermal equilibrium – even only the stationary state – are not yet existing.

Therefore, setting up simple models, which preferably are solvable analytically and which allow to study the physical properties also off equilibrium in the stationary state, is still a highly relevant approach to obtain at least a qualitative feeling how various degrees of freedom influence the transport properties of nano-objects.

The situation in an experimental setup like the one shown in Fig. 1 has the advantage that one can, for example, by stretching

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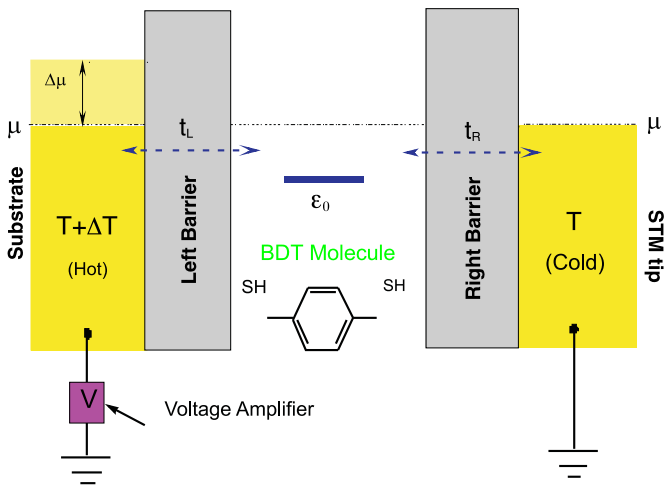

Fig. 2. Schematic setup of the model. We consider only one relevant orbital of the BDT molecule, which we assume to be the HOMO. The coupling between the substrate respectively the STM tip are modeled as tunneling barriers.

the bond through removing the tip, control the coupling between the leads and the molecule very sensitively and actually shift the system into the weak coupling regime. Here, theoretical calculations become much easier. For our purpose, we will therefore assume that the system is in this regime, and suggest to employ a very simple model which will allow us to see how the interplay between electronic and vibrational degrees of freedom influence transport properties.

The model setup and general expressions for thermopower and possible situations will be presented in the next section. The central result, viz. analytical expressions for the thermopower and Seebeck coefficient will be derived in Section 3, and a summary will conclude the paper.

2. Setup and model

We study an organic molecule attached to two metallic leads. The leads are assumed to be in thermal equilibrium, but can have different temperatures and chemical potentials. We will take the temperature and chemical potential of the right leads as reference and write $T_R = T$, $T_L = T + \Delta T$ respectively $\mu_R = \mu$, $\mu_L = \mu + \Delta\mu$. In the following, we use $\mu = 0$ as reference of energy.

Using first-principle approaches, such a setup has been studied before in a quite general way [9], concentrating however on the local properties of the molecule. Here, we intend to investigate transport properties, and how local vibration modes influence them. Calculating transport for a general situation becomes rather quickly a cumbersome task. Therefore, in order to be able to gain some insight into the physics of the problem while at the same time having as little parameters as possible, we concentrate her on a very simple model. To this end we assume that the HOMO of the molecule has a certain energy ϵ_0 below the chemical potential of the leads, but the LUMO well above it, so that we can neglect it. Also, all other electronic states of the molecule are assumed to be well separated, too. The model motivated above is shown schematically in Fig. 2, where we also explicitly included the various model parameters.

Up to now the model is the standard setup to describe resonant transport through a mesoscopic system like molecules or quantum dots. Here, however, we also allow for vibrational modes of the molecule, which we describe in a harmonic approximation, and which couple to the electronic degrees of freedom via a Holstein term. Although the calculation can be performed for several phonon modes, we concentrate here on the case of one single mode with frequency ω_0. This is for example permissible when one studies low temperature, where only the lowest-lying modes play a role anyway. Finally, the molecule is coupled to the leads via tunneling barriers. The resulting model is the well-known Anderson–Holstein model adapted to describe the experimental setup from Fig. 1. Its Hamiltonian thus reads

$$H = H_{\mathrm{L}} + H_{\mathrm{M}} + H_{\mathrm{LM}} \tag{1}$$

$$H_{\mathrm{L}} = \sum_{\mathbf{k},\alpha=L,R} \varepsilon_{\alpha,\mathbf{k}} c^{\dagger}_{\alpha,\mathbf{k}} c_{\alpha,\mathbf{k}} \tag{2}$$

$$H_{\mathrm{M}} = \epsilon_0 d^{\dagger} d + \omega_0 b^{\dagger} b + \lambda\omega_0 d^{\dagger} d\left(b^{\dagger} + b\right) \tag{3}$$

$$H_{\mathrm{LM}} = \sum_{\alpha=L,R} \left(t_\alpha c^{\dagger}_{\alpha,B} d + \mathrm{h.c.}\right) \tag{4}$$

where we use standard second quantized notation. H_{L} – Hamiltonian is describes the electrons on the electrode (source), H_{M} denotes possible interaction in molecule, $b^{\dagger}, b$ – creation and annihilation of phonon, H_{LM} – describing the tunneling electron through the barriers, λ denotes the dimensionless coupling between the electronic and phononic modes, and t_α the tunneling matrix elements to the leads. The operator $c^{(\dagger)}_{\alpha,B}$ denotes the projection of the band states to the barrier.

Note that we did not include the spin degrees of freedom of the electrons in the model (1). Although this is at first glance a rather crude approximation, we want to employ it nevertheless for two reasons. First, including the spin makes already the molecular problem highly nontrivial and would prevent the semi-analytical solution we will discuss in the section about the results. Second, we are presently aiming at a more qualitative insight into the influence of vibrational modes on transport effects. To this end we believe that the additional complications due to additional electronic degrees of freedom will surely alter the results quantitatively, but possibly not strongly on a qualitative level.

The model (4) has of course been introduced in the context of electron–phonon coupling and polaron physics long ago and, in the case of $t_\alpha = 0$, can be solved analytically [12]. It has been studied extensively with various methods [13–24] in many different context [19–21,25–30] including but not exclusively studying transport through nano-structures in and out of equilibrium.

The basic relation in studying transport through nano-structures is the Meir–Wingreen formula [10] to calculate the electric current as

$$J = J_0 \int d\omega \left[f_L(\omega) - f_R(\omega)\right] \rho_M(\omega, \Delta\mu, \Delta T) \tag{5}$$

where $\rho_M(\omega, \Delta\mu, \Delta T)$ denotes the electronic density of states on the molecule in the presence of a potential difference ΔV and a temperature difference ΔT between the leads, and

$$f_\alpha(\omega) = \frac{1}{1 + \exp(\frac{\omega - \mu_\alpha}{k_{\mathrm{B}} T_\alpha})}$$

the Fermi functions of the left and right lead, respectively. The prefactor $J_0 = \frac{2\pi e^2}{\hbar} \cdot \frac{\Gamma_L \Gamma_R}{\Gamma_L + \Gamma_R}$ collects the coupling parameters $(\alpha = L, R)$ $\Gamma_\alpha := \pi N_\alpha(\mu_\alpha)|t_\alpha|^2$, where $N_\alpha(\epsilon)$ denotes the density-of-states of lead α, and the natural constants.

Calculating the DOS in the presence of the leads for arbitrary coupling t_α, $\Delta\mu$ and ΔT is not possible. However, in the case $t_\alpha \to 0$ (weak coupling) for both $\alpha = L$ and R, one can approximate $\rho_M(\omega, \Delta\mu, \Delta T)$ by the result for the isolated molecule, which reads

$$\rho_M(\omega, \Delta\mu, \Delta T) = \frac{2\pi}{e^\lambda} \sum_{l=-\infty}^{\infty} \frac{\lambda^{|l|}}{|l|!} \delta\left(\omega - \epsilon_0 - (l + \lambda)\omega_0\right) \tag{6}$$

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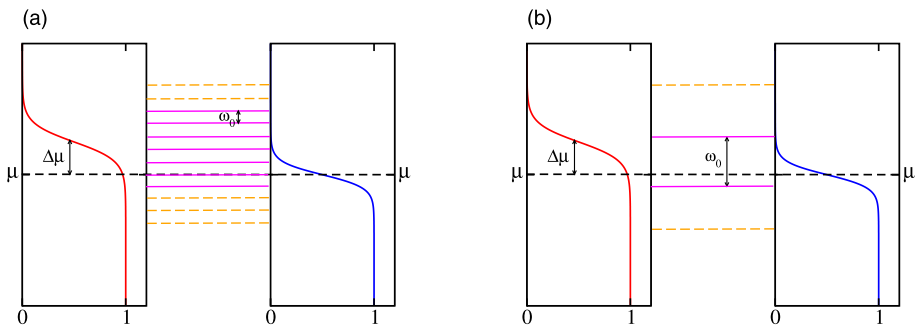

Fig. 3. Schematic representation of the different situations arising from Eq. (7). The left and right boxes denote the left and right leads, with the Fermi functions as full lines. In the left lead, we included some $\Delta\mu$ and ΔT. In (a) there are several polaronic modes which lie in the Fermi window, while in (b) the temperature is much smaller than the level splitting, resulting in effectively only two modes within the Fermi window.

and does not depend on $\Delta\mu$ and ΔT explicitly.

This model has been studied in the context of calculation of thermopower before [19–21], including higher order effects like co-tunneling through rate equations [19] respectively equations of motion for the Keldysh Green's functions [20,21]. This, in principle, allows for a much more elaborate treatment of the present model. However, the equations derived in these approaches are rather complex and cannot be evaluated readily. On the other hand, the present experiment appears to be situated in the extreme weak-coupling limit at rather elevated temperatures, and hence these higher order effects will be of little influence here. Making use of the resulting simplifications lead to a very simple analytical formula where one can extract several generic features analytically and is also able to explain the experiment almost quantitatively with little effort. We therefore believe that our approach, despite its admittedly very simple theoretical basis, represents a valuable contribution.

We should note at this point, that in experimental setups like the one depicted in Fig. 1 one usually has to expect very asymmetric couplings between the molecule and the leads, which will in general lead to a dependence of the physical and transport properties on this asymmetry [31]. However, the results presented in [1] are actually averages over a huge number of similar setups with varying coupling strengths for both molecule-surface and molecule-tip. Since the experiments are rather stable and seem to produce reasonably sharp distributions over a rather large region of tip-molecule distances we are certain that this particular experiment is rather well described by $\Gamma_\alpha \to 0$ for both leads, with little influence by a left–right asymmetry.

When we insert expression (6) into the Meir–Wingreen formula (5), the expression for the current becomes

$$J = \frac{2\pi J_0}{e^\lambda} \sum_{l=-\infty}^{\infty} \frac{g^{|l|}}{|l|!} \big[f_L\big(\epsilon_0 + (l+\lambda)\omega_0\big) - f_R\big(\epsilon_0 + (l+\lambda)\omega_0\big) \big]$$

Since we are interested in the thermopower, the current has to be zero, i.e. for a given temperature difference ΔT between left and right lead we have to adjust $\Delta\mu$ such that

$$\sum_{l=-\infty}^{\infty} \frac{\lambda^{|l|}}{|l|!} \big[f_L\big(\epsilon_0 + (l+\lambda)\omega_0\big) - f_R\big(\epsilon_0 + (l+\lambda)\omega_0\big) \big] = 0 \qquad (7)$$

In general, we are interested in low temperatures, i.e. typically we will have $|\epsilon_0|, \omega_0 \gg k_B T$. In this case, only a certain range of values of $l \in [l_{\min}, l_{\max}]$, which have the property that $\epsilon_0 + (l+\lambda)\omega_0 = \mathcal{O}(k_B T)$, will actually be important, because for the other terms the Fermi functions are either both zero or both one, i.e. cancel in the difference. This situation is schematically depicted in Fig. 3a.

Let us take this idea to the extreme and assume, that T is low enough such that only one vibrational mode with index l_0 will contribute, see the schematic representation in Fig. 3b. The necessary conditions for transport in this case are

$$\epsilon_0 + (l_0 + \lambda)\omega_0 < 0$$
$$0 \leqslant \tilde{\epsilon}_0 := \epsilon_0 + (l_0 + 1 + \lambda)\omega_0 < \omega_0$$

from which l_0 can be determined. Eq. (7) then simplifies to

$$\frac{1}{1+\exp(\frac{\tilde{\epsilon}_0-\omega_0-\Delta\mu}{k_B(T+\Delta T)})} - \frac{1}{1+\exp(\frac{\tilde{\epsilon}_0-\omega_0}{k_B T})} + \frac{\lambda}{l_0+1}\left[\frac{1}{1+\exp(\frac{\tilde{\epsilon}_0-\Delta\mu}{k_B(T+\Delta T)})} - \frac{1}{1+\exp(\frac{\tilde{\epsilon}_0}{k_B T})}\right] = 0 \qquad (8)$$

This equation can be easily solved for $\Delta\mu$ for given model parameters and ΔT. Although this is strictly speaking not the case for the experimental setup in Fig. 1, we want to note that in nano-devices ϵ_0 can often be controlled through a gate voltage, $\epsilon_0 = -eV_G$, and consequently the active polaronic mode respectively l_0 is controllable experimentally.

3. Results

Without vibrational modes, the thermoelectric response of the model (1) has been studied before by Ermakov et al. [11]. Within their model, the authors observed a linear relation between the temperature difference and thermovoltage. In the following we want to try to understand what modifications arise through the presence of polaronic modes in the molecule.

In the present paper we want to concentrate on the situation shown in Fig. 3b. Here we can use the fact that $\omega_0 \gg k_B T$ and try to solve equation (8) approximately analytically and provide explicit expressions for $\Delta\mu$ and the Seebeck coefficient as function of model parameters and temperature.

The limiting cases $\tilde{\epsilon}_0 \to 0$ and $\tilde{\epsilon}_0 \to \omega_0$ turn out to be uninteresting, because in the first case the combination $f_L(\tilde{\epsilon}_0 - \omega_0) - f_R(\tilde{\epsilon}_0 - \omega_0)$ vanishes, while in the second $f_L(\tilde{\epsilon}_0) - f_R(\tilde{\epsilon}_0) \to 0$. In both cases one thus recovers the solution by Ermakov et al. [11] in the limit when the level position is close to the chemical potential.

The more interesting case therefore is $\tilde{\epsilon}_0$ finite and reasonably far away from the chemical potential. Then we can furthermore assume $\tilde{\epsilon}_0 \gg k_B T$ as well as $|\tilde{\epsilon}_0 - \omega_0| \gg k_B T$, and replace the Fermi functions by $f(x) \approx e^{-\beta x}$, if $x > 0$, respectively $f(x) \approx 1 - e^{\beta x}$, if $x < 0$. Inserting these approximations into Eq. (8) one obtains a quadratic equation for $e^{\frac{\Delta\mu}{k_B(T+\Delta T)}}$. From the two solutions one must pick the one that has the correct limit

$$\Delta\mu = (\omega_0 - \epsilon_0)\frac{\Delta T}{T}$$

as $\lambda \to 0$. The solution fulfilling this requirement reads

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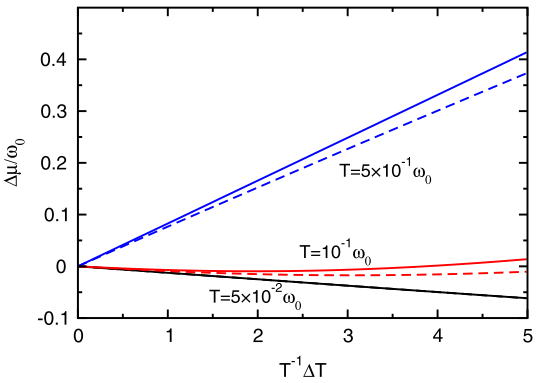

Fig. 4. $\Delta\mu$ vs. $T^{-1}\Delta T$ for various values of T. Other parameters see text. The dashed curves are the results from the approximate formula (9).

$$\Delta\mu \approx (T+\Delta T)\ln\frac{e^{-\frac{\tilde{\epsilon}_0}{k_B T}}}{\frac{2\lambda}{l_0+1}}\left[e^{\frac{\tilde{\epsilon}_0}{k_B(T+\Delta T)}}\left(\frac{\lambda}{l_0+1}-e^{\frac{2\tilde{\epsilon}_0-\omega_0}{k_B T}}\right)\right.$$
$$\left.+\sqrt{e^{\frac{2\tilde{\epsilon}_0}{k_B(T+\Delta T)}}\left(e^{\frac{2\tilde{\epsilon}_0-\omega_0}{k_B T}}-\frac{\lambda}{l_0+1}\right)^2+\frac{4\lambda}{l_0+1}e^{\frac{2\tilde{\epsilon}_0-\omega_0}{k_B(T+\Delta T)}+\frac{2\tilde{\epsilon}_0}{k_B T}}}\right] \tag{9}$$

Since this expression is rather hard to interpret, we inspect it behavior for small λ. Taylor expanding it to first order results in

$$\Delta\mu \approx (\omega_0-\tilde{\epsilon}_0)\frac{\Delta T}{T}-\frac{\lambda(T+\Delta T)}{l_0+1}e^{\frac{\omega_0-2\tilde{\epsilon}_0}{k_B T}}\left[e^{\frac{\omega_0\Delta T}{k_B T(T+\Delta T)}}-1\right]$$

The first term is identical to the result by Ermakov et al. [11]. The correction is negative, i.e. the vibrations tend to initially reduce the thermopower. Note that the strenght of this effect depends on the relation between $\tilde{\epsilon}_0$ and ω_0: If $\omega_0-2\tilde{\epsilon}_0<0$, it will be exponentially suppressed, while it is actually strongly enhanced for $\omega_0-2\tilde{\epsilon}_0>0$.

Let us see in to what extent the approximate solution (9) is reliable. To this end we solve Eq. (8) numerically for $\Delta\mu$. As generic parameter set we chose $\tilde{\epsilon}_0=\omega_0/8$ and $\lambda/(l_0+1)=1/40$. Due to the exponential structure a numerical solution fails for too small T. Using multi-precision arithmetic, one can extend the range of solvability slightly. We checked the stability of the root finding algorithm by comparing the results for $\Delta\mu$ at $\lambda\to 0$ with the analytical result and found that stable solutions can be obtained for $T\geqslant 5\cdot 10^{-2}\omega_0$ using a precision of 100 digits. The resulting function $\Delta\mu(\Delta T)$ for $T=5\cdot 10^{-2}\omega_0$, $10^{-1}\omega_0$ and $\omega_0/2$ is shown in Fig. 4 as full lines, together with the approximate functions as dashed lines. For low T we observe a linear dependence with negative coefficient, while at high T the sign of the thermopower reverses. For intermediate T, however, one can see a rather peculiar sign change of $\Delta\mu$ as function of ΔT. Changing the model parameter does not influence this qualitative behavior as long as $\omega_0-2\tilde{\epsilon}_0>0$. For example, decreasing $\tilde{\epsilon}_0$ moves the zero in $\Delta\mu$ towards smaller values of ΔT, but also decreases the magnitude of the effect. Increasing λ produces a similar behavior. However, when $\omega_0-2\tilde{\epsilon}_0<0$ the features due to the vibrations vanish and one recovers more or less the behavior for $\lambda\to 0$, i.e. linear with slope $(\tilde{\epsilon}_0-\omega_0)/T>0$.

The curves resulting from the approximate formula give, as expected from the assumptions behind this analytical approximation, an accurate description for low temperatures, while for higher temperatures deviations become more pronounced. However, the qualitative behavior is well reproduced in all cases and we conclude that the analytical function represents a reasonable approximation for higher temperatures, too.

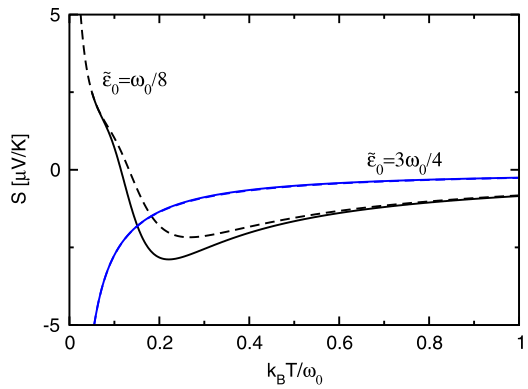

Fig. 5. Seebeck coefficient $S=-\frac{\Delta V}{\Delta T}|_{\Delta T\to 0}$ as function of T for two characteristic values of $\tilde{\epsilon}_0$. Other parameters as in Fig. 4. The dashed curves are the result from the approximate formula (10).

Finally, one can look at the initial slope of $\Delta\mu(\Delta T\to 0)$, which, when divided by the electron charge, is the negative Seebeck coefficient S for the junction. The result as function of temperature for the same model parameters used in Fig. 4 is shown in Fig. 5. Again, we include the result (e_0 denotes the electric charge)

$$S\approx\frac{k_B}{e_0}\frac{1}{k_B T}\left[\tilde{\epsilon}_0-\frac{\omega_0}{1+\frac{\lambda}{l_0+1}e^{\frac{\omega_0-2\tilde{\epsilon}_0}{k_B T}}}\right] \tag{10}$$

derived from the approximate formula (9) as dashed curve in Fig. 5. It apparently describes the overall behavior quite well, becoming asymptotically exact for low T. Interestingly it also provides the correct asymptotic for high T. Note that the unit of the thermopower is, as usual, determined by the universal ratio k_B/e_0 and is hence not model or parameter dependent. As already noted in the case of $\Delta\mu$, we here, too, obtain a sensible dependence of the Seebeck coefficient on $\tilde{\epsilon}_0$. When $\omega_0-2\tilde{\epsilon}_0>0$, it shows strong variation with T due to the denominator in Eq. (10), while for $\omega_0-2\tilde{\epsilon}_0<0$ the latter influence is exponentially suppressed and hence one obtains a temperature dependence which is very similar to the case without molecular vibrations. Since one can possibly control the value of ϵ_0 and hence $\tilde{\epsilon}_0$ experimentally, at least for electrically gated molecules, the described effects and their switching could in principle be observable.

4. Summary

A simple model for an organic molecule attached to two conducting leads via tunnel barriers was introduced. Main emphasis was laid on the inclusion of vibration modes for this molecule and to study their impact on transport properties, here especially the thermopower. We derived an analytical approximate formula for the thermopower $\Delta\mu(\Delta T)$ and the Seebeck coefficient and showed that for a rather broad range of model parameters the influence of the molecular oscillations can strongly influence these two quantities. In particular, the thermopower can develop a pronounced extremum and sign changes as function of temperature gradient across the molecule, and the Seebeck coefficient showed pronounced nonmonotonic behavior in this regime. When one shifted the local electronic level of the molecule to larger values, one can observe a transition into a regime where the vibrational modes have negligible influence and the system behaves similar to the case without any elastic degrees of freedom. Since in some experimental setups the local parameters are accurately controllable via e.g. gate voltage or the like, we are certain that one can test these theoretical predictions to some extent.

Of course, the simplifications introduced to allow for an analytical solution are rather severe: Electron spin as well as additional

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molecular levels were neglected, and the vibration modes were restricted to a single mode. Furthermore, for the calculation of the transport only two of the polaron levels were taken into account. The latter approximation can more easily be relaxed, even within the proposed model (see Fig. 3a). Similarly, including several vibration modes can be done straightforwardly (see for example [12]), too, while taking into account more realistic electronic level structures will actually require more refined analytical approaches [7] or a numerical solution of the resulting model for the molecule [8]; and possibly also a reinspection of the validity of the Meir–Wingreen formula for the current.

In any case, adding these additional and physically important features to the model of the molecule makes the problem harder to treat. At least as long as we stay in the weak-coupling limit, we can expect that one can handle them reasonably within existing numerical approaches, or even, up to the solution of the transport equation, to a large extent analytically. The actual problematic part is hidden in the solution of the transport equation, even in weak coupling. Here, the exponential nature of the Fermi functions renders the numeric root searching extremely awkward and severely limits the accessible parameter and especially temperature regime (see e.g. discussion of Fig. 4). Finally, relaxing the condition of weak coupling to the leads leads to a full-scale many-body problem out of equilibrium, which at present cannot be really attacked with reliable analytical or numerical tools. It will be interesting to examine the asymmetry of barriers on influence the Seebeck effect with the vibrational degrees of freedom, which corresponds to different degrees of contacts in STM experiments.

Thus, to conclude, we believe that even such an extremely simple model will be of some importance to increase our understanding of properties of nano-systems, and can eventually also serve as a benchmark for more advanced methods to be developed in the future.

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