Coherent laser control of the current through molecular junctions

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Abstract – The electron tunneling through a molecular junction modeled by a single site weakly coupled to two leads is studied in the presence of a time-dependent external field using a master equation approach. In the case of small bias voltages and high carrier frequencies of the external field, we observe the phenomenon of coherent destruction of tunneling, i.e. the current through the molecular junction vanishes completely for certain parameters of the external field. In previous studies the tunneling within isolated and open multi-site systems was suppressed; it is shown here that the tunneling between a single site and electronic reservoirs, i.e. the leads, can be suppressed as well. For larger bias voltages the current does not vanish any more since further tunneling channels participate in the electron conduction and we also observe photon-assisted tunneling which leads to steps in the current-voltage characteristics. The described phenomena are demonstrated not only for monochromatic fields but also for laser pulses and therefore could be used for ultrafast optical switching of the current through molecular junctions.

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Introduction. – Electronic transport through molecular wires and junctions has recently attracted much attention experimentally as well as theoretically [1–3]. Under the influence of a bias voltage and because of the coupling to the leads which act as electron source and drain, a current through the molecular junction is established. When an external time-dependent field, such as a laser field or an additional ac voltage is applied to the molecular junction, several interesting effects arise. One phenomenon is the well-known photon-assisted tunneling (PAT) [4]. It was studied already in the early 1960’s experimentally by Dayem and Martin [5] and theoretically by Tien and Gordon using a simple theory which captures already the main physics of PAT [6]. The main idea is that an external field periodic in time with frequency \( \omega \) can induce inelastic tunneling events when the electrons exchange energy quanta \( \hbar \omega \) with the external field. Another important effect is the famous phenomenon of coherent destruction of tunneling (CDT). Grossmann et al. [7–9] first studied this effect and found that tunneling can be quenched in a periodically driven quantum system. In the context of molecular wires, this phenomenon can be explained using Floquet theory in the case of a periodic laser field [10], and CDT occurs for certain amplitudes of the laser field at fixed frequencies [11–15]. Different scenarios of controlling the tunneling in molecular wires and quantum dots have been proposed based on different mechanisms [4,14–18]. Also current-induced light emission in molecular junctions has been studied [19].

In the current paper we focus on the tunneling through a single-site molecular junction. This might be a quantum dot (though the temperatures in the current examples are rather high for quantum dots) or a single molecular level acting as a molecular wire. The theoretical foundation is a density matrix formalism using a perturbative treatment within the molecule-lead coupling to second order. Applying this technique it is possible to calculate the time-dependent population in and the current through the molecular junction under the influence of a time-varying external field [15,20]. Since the effect of the external field on the coupling between molecule and leads is treated exactly and not neglected as for example in Redfield theory, effects based on the influence of the laser on this coupling can be investigated. The tunneling between the molecule and the leads can, for small bias voltages, be suppressed by a monochromatic laser. This result is different from previous studies [13–15,21] in which the current vanished because of the CDT between the sites of
the wire. In ref. [13] the possibility of CDT for a single site was briefly mentioned as a limiting case of a wire with two sites having a large inter site coupling, but it was not further explored. Additionally we demonstrate that not only monochromatic laser fields but also laser pulses can lead to CDT. This opens opportunities for building optical current switches with a time resolution on the femtosecond scale.

Model. – The total system Hamiltonian \( H(t) = H_S(t) + H_R + H_{SR} \) includes three parts: the relevant system \( H_S(t) \) mimicking the molecule, the reservoirs \( H_R \) modeling the two leads and the system-reservoir coupling \( H_{SR} \). The creation and annihilation operators of electrons with spin \( \sigma \) are denoted by \( c_{\sigma}^\dagger \) and \( c_{\sigma} \), respectively, so that the description of a single-site molecule reads

\[
H_S(t) = \sum_{\sigma} (\varepsilon_0 - \mu E(t)) c_{\sigma}^\dagger c_{\sigma} + U c_{\uparrow}^\dagger c_{\uparrow}^\dagger c_{\downarrow} c_{\downarrow},
\]

with spin-independent on-site energy \( \varepsilon_0 \) and electron interaction \( U \) within the doubly occupied states. The time-dependent term \( -\mu E(t) \) describes the effect of the external field \( E(t) \) and for simplicity we assume that the proportionality factor \( -\mu \) equals unity. The external field is of the form \( E(t) = E_0(t) \cos(\omega t) \) with a possibly time-varying amplitude \( E_0(t) \). The two leads coupled to the molecular junction are mimicked as electron reservoirs in thermal equilibrium by \( H_R = \sum_{q\sigma} \omega_{q\sigma} b_{q\sigma}^\dagger b_{q\sigma} \). Here \( b_{q\sigma}^\dagger \) and \( b_{q\sigma} \) denote the creation and annihilation operators of an electron with spin \( \sigma \) in reservoir mode \( \omega_q \). Due to the assumed thermal equilibrium of the electronic leads, the occupation expectation values of the reservoir modes are determined by \( \langle b_{q\sigma}^\dagger b_{q\sigma} \rangle = n_F(\omega_q - E_F) \delta_{q0} \) where \( n_F \) is the Fermi function and \( E_F \) the Fermi energy. In further derivations we will only refer to the left lead but the formalism has to be applied to the right lead coupled to the wire as well. The coupling of the left lead to the molecule is given by

\[
H_{SR} = \sum_{\sigma, \sigma', \lambda = 1,2} K_{\lambda \sigma} \Phi_{\lambda \sigma'} = \sum_{q\sigma} \left( V_q c_{\uparrow}^\dagger b_{q\sigma} + V_q^* b_{q\sigma}^\dagger c_{\downarrow} \right)
\]

with \( \Phi_{\lambda \sigma} = \Phi_{\lambda \sigma'} = \sum_q V_q b_{q\sigma}^\dagger c_{\downarrow} \). \( K_{\lambda \sigma} = K_{\lambda \sigma'} = c_{\lambda \sigma}^\dagger \), and a wire-lead coupling strength \( V_q \). For the coupling of the molecule to the right lead similar equations hold.

The calculations are performed with and without electron interaction in the molecule. Neglecting electron interaction all electron-carrying states of the molecule are degenerate with energy \( \varepsilon_0 \). In the case of electron interaction \( U \), double occupancy leads to a state with energy \( 2\varepsilon_0 + U \). When the bias voltage is small and \( \varepsilon_0 \) is between the Fermi energies \( E_{F\sigma} \) and \( E_{F\uparrow} \) of the right and the left lead, respectively, only the tunneling through the channel with energy \( \varepsilon_0 \) leads a current, \( I_0 \). When the value of the bias voltage is above \( U \), an extra channel opens and there is a shoulder in the current. Thielmann et al. [22] used these energy levels to explain the shoulders in the tunneling current for a quantum dot at low temperature. The excited state in the investigations by Bruder [23,24], Oosterkamp [25–27] and others has the same effect [28,29].

Since in most cases one is only interested in the time evolution of the relevant system, i.e. in this case the molecule, a time-local quantum master equation (QME) based on a second-order perturbation theory in the molecule-lead coupling has been developed for the reduced density matrix of the molecule \( \rho_S(t) \) [15,20]

\[
\frac{\partial \rho_S(t)}{\partial t} = -i \mathcal{L}_S(t) \rho_S(t) - \sum_{s, \sigma} [K_{s\sigma}, \Lambda_{s\sigma}(t) \rho_S(t) - \rho_S(t) \tilde{\Lambda}_{s\sigma}(t)]
\]

with auxiliary operators for the molecule-lead coupling

\[
\Lambda_{s\sigma}(t) = \int_0^t dt' C_{s\sigma}(t-t') U_S(t',t') K_{s\sigma},
\]

\[
\tilde{\Lambda}_{s\sigma}(t) = \int_0^t dt' C_{s\sigma}(t-t') U_S(t',t') K_{s\sigma}.
\]

Here \( \mathcal{L}_S(t) = [H_S(t), \cdot] \) is the Liouville operator, \( U_S(t,t') = T_e \exp[-i \int_t^{t'} dt' \mathcal{L}_S(t')] \) the time evolution operator and \( C_{s\sigma}(t) = \text{tr}_R \{ e^{iH_St} \Phi_s e^{-iH_St} \rho_R \} \) the reservoir correlation functions with spin-independent reservoir density matrix \( \rho_R \). \( T_e \) is the time-ordering operator.

The properties of the Fermionic reservoirs are described by a single quantity, the spectral density \( J_R(\omega) \). For a dense spectrum, \( J_R(\omega) \) is a smooth function and one can approximate it by a numerical decomposition into few Lorentzian functions [15]

\[
J_R(\omega) = \sum_{k=1}^{m} \frac{p_k}{4\Omega_k} \frac{1}{(\omega - \Omega_k)^2 + \Gamma_k^2},
\]

with real fitting parameters \( p_k \), \( \Omega_k \) and \( \Gamma_k \). Using the theorem of residues and denoting the Fermi function as \( n_F \) yields

\[
C_{12}(t) = \sum_{k=1}^{m} \frac{p_k}{4\Omega_k \Gamma_k} \left( n_F(\Omega_k^+ - E_F)e^{-i\Omega_k t} \right)
\]

\[
-2i \frac{m}{\beta} \sum_{k=1}^{m} J_R(\nu_k) e^{-i\nu_k t} \sum_{k=1}^{m} a_{12}^k e^{i\nu_k t},
\]

\[
C_{21}(t) = \sum_{k=1}^{m} \frac{p_k}{4\Omega_k \Gamma_k} \left( n_F(\Omega_k^+ - E_F)e^{i\Omega_k t} \right)
\]

\[
-2i \frac{m}{\beta} \sum_{k=1}^{m} J_R(\nu_k) e^{i\nu_k t} \sum_{k=1}^{m} a_{21}^k e^{i\nu_k t},
\]

with the abbreviation \( \Omega_k^+ = \Omega_k \pm i \Gamma_k \) and the Matsubara frequencies \( \nu_k = \frac{2\pi k + \pi}{\beta} + E_F \). The infinite sums over the
\( \nu_k \) can be truncated at a finite value depending on the temperature \( T \) and the spectral width of \( J_B(\omega) \). With these forms of \( C_{12} \) and \( C_{21} \) one can obtain a set of differential equations for the auxiliary density operators

\[
\frac{\partial}{\partial t} \hat{\Lambda}_{xx'\sigma}^k(t) = a_{xx',K_{xx'} - i[H_S(t), \hat{\Lambda}_{xx'\sigma}^k(t)]} + \gamma_{xx', \hat{\Lambda}_{xx'\sigma}^k(t)},
\]

\( (9) \)

\[
\frac{\partial}{\partial t} \hat{\Lambda}_{xx'\sigma}^k(t) = (a_{xx',K_{xx'}} - i[H_S(t), \hat{\Lambda}_{xx'\sigma}^k(t)] + (\gamma_{xx'} \hat{\Lambda}_{xx'\sigma}^k(t)),
\]

\( (10) \)

with \( \hat{\Lambda}_{xx'\sigma}^k(t) = \sum_{k=1}^{m+m'} \hat{\Lambda}_{xx'\sigma}^k(t) \) and \( \hat{\Lambda}_{xx'\sigma}^k(t) = \sum_{k=1}^{m+m'} \hat{\Lambda}_{xx'\sigma}^k(t) \). As it has been detailed previously [15], the coupled differential equations (3), (9), (10) can now be numerically integrated applying, e.g., the Runge-Kutta scheme.

Using the electron number operator of the left lead with the summation performed over the reservoir degrees of freedom \( N_l = \sum_{\sigma} b_{\sigma}^\dagger b_{\sigma} \), the expression for the current is given by [15]

\[
I_l(t) = \frac{e}{d} \frac{d}{dt} \text{tr}(N_l \rho_S(t)) = -ie \text{tr} \{ [N_l, H(t)] \rho_S(t) \}
\]

\[
= 2e \text{Re} \left( \text{tr}_S \left( \sum_{\sigma} \{ c_{1\sigma} \Lambda_{12\sigma}(t) \rho_S(t) - c_{1\sigma}^\dagger \rho_S(t) \Lambda_{12\sigma}^\dagger(t) \} \right) \right).
\]

\( (11) \)

Here \( e \) denotes the elementary charge. This equation describes the current \( I_l(t) \) from the lead into the molecule. A similar expression holds for \( I_r(t) \) from the right lead into the molecule. In a steady state and after averaging over one period of the driving field, \( I_l \) and \( I_r \) have the same magnitude but opposite signs and therefore a total transient current through the molecular junction can be defined as \( I(t) = (I_l(t) - I_r(t)) \)/2. The time-dependent average current \( \overline{I} \) will be determined below by averaging \( I(t) \) over five periods of the highly oscillating carrier field.

A simple spectral density with only one Lorentzian \( (m=1) \) was chosen. With \( \Omega_1 = \varepsilon_0 \) we locate the maximum of the coupling at the site energy. By using \( \Gamma_1 = 5\omega \), the coupling between the leads and the system is almost in the wide-band limit. Choosing \( p_1 = 0.04\omega\Omega_1\Gamma_1^2 \) we obtain a maximum coupling strength of \( 0.01\omega \) which is smaller than the thermal energy \( k_B T = 0.025\omega \) and much smaller than the external field energy \( \omega \). So the results below are all within the high-frequency limit.

According to the approximate Tien-Gordon model [4,6] for monochromatic external fields which is based on a simplified scattering picture, the rectified dc currents through ac-driven molecular junctions are determined as [4]

\[
I_{TG} = \sum_{n=-\infty}^{\infty} J_n^2 \left( \frac{E_0}{\omega} \right) I_{dc}(\varepsilon_0 + n\omega) = \sum_{n=0}^{\infty} I_n, \quad (12)
\]

where the current in the driven system is expressed by a sum over contributions of the current \( J_n^2 \left( \varepsilon_0 + n\omega \right) \) in the un-driven case but evaluated at side-band energies \( \varepsilon_0 + n\omega \) shifted by integer multiples of the photon quantum and weighted with squares of Bessel functions. Note that the partial currents \( I_n \) contain contributions from \( \pm n \). The term \( J_n(E_0/\omega) \) denotes the \( n \)-th order Bessel function of the first kind. The photon absorption \((n > 0)\) and emission \((n < 0)\) processes can be viewed as creating effective electron densities at energies \( \varepsilon_0 \pm n\omega \) with probability \( J_n^2(E_0/\omega) \). Already Dayem and Martin [5] showed clear steps in the tunneling current induced by these two processes. Although “a great deal of information can be extracted from simple models like the Tien-Gordon model” [4] in many cases one needs more sophisticated approaches. In double quantum dot theories, for example, the Tien-Gordon formula has been recovered in the limit of weak inter-dot coupling [4,30–32].

In the following we use a bias symmetric with respect to \( \varepsilon_0 \), i.e. \( E_{F,1} = \varepsilon_0 + V_b/2 \) and \( E_{F,2} = \varepsilon_0 - V_b/2 \). The current \( I_{TG} \) obtained with the help of the Tien-Gordon theory is compared to the average current \( \overline{I} \) obtained from the QME described above. The partial currents \( I_n \) defined in eq. (12) are used to qualitatively explain the steps in the \( I-V \) characteristics. In the results shown below we first restrict ourselves to a monochromatic laser field with constant field amplitude of \( 2.405\omega \), i.e. a zero of the zeroth-order Bessel function \( J_0(\omega/\omega = 2.405) = 0 \).

**Monochromatic laser, \( U = 0 \).** – In this case, energy levels are equal to \( \varepsilon_0 \) regardless whether there is already an electron with opposite spin on the site or not, and lead to the partial current \( I_0 \). The PAT-induced states have also equal energies \( \varepsilon_0 \pm n\omega \) leading to the inelastic current contributions \( I_n \). In fig. 1 the average current \( \overline{I} \)
shows a step at $V_b/2 = \omega$. When $V_b/2$ is smaller than $\omega$, the current vanishes. At these low bias voltages channels with $n \neq 0$ cannot participate in the conduction since the energies of the PAT-induced states are not in the window between the Fermi energies of the right and left contacts. Because $E_0/\omega = 2.405$, $I_0$ also vanishes due to CDT. For other ratios of $E_0/\omega$ it is non-zero. When $V_b/2 = \omega$, the left Fermi energy $E_{F,L} = \varepsilon_0 + \omega$ equals the PAT-induced energy and $E_{F,R} = \varepsilon_0 - \omega$. Therefore the current $I_1$ jumps to a finite value, so do $I_{TG}$ and $I$. A comparison of $I_{TG}$ and $I$ in fig. 1 shows that each step in these $I$-$V$ curves is related to an inelastic current $I_n$. The slope of the steps is of course directly connected to the temperature. The sum $I_{TG}$ of all partial currents for $n = 0, \ldots, 3$ agrees reasonably with the current $I$ from the QME, especially for small bias voltages.

Figure 2: Same as fig. 1 but with electron interaction $U = 0.4\omega$. Shoulders in the current are at $V_b/(2\omega) = 0.6, 1.0, 1.4, 1.6, 2.0$, etc.

Fig. 2: Same as fig. 1 but with electron interaction $U = 0.4\omega$. Shoulders in the current are at $V_b/(2\omega) = 0.6, 1.0, 1.4, 1.6, 2.0$, etc.

Concentrate on this case of small Coulomb interaction. Because $E_0/\omega = 2.405$, $I_0$ also vanishes due to CDT. For other ratios of $E_0/\omega$ it is non-zero. When $V_b/2 = \omega$, the left Fermi energy $E_{F,L} = \varepsilon_0 + \omega$ equals the PAT-induced energy and $E_{F,R} = \varepsilon_0 - \omega$. Therefore the current $I_1$ jumps to a finite value, so do $I_{TG}$ and $I$. A comparison of $I_{TG}$ and $I$ in fig. 1 shows that each step in these $I$-$V$ curves is related to an inelastic current $I_n$. The slope of the steps is of course directly connected to the temperature. The sum $I_{TG}$ of all partial currents for $n = 0, \ldots, 3$ agrees reasonably with the current $I$ from the QME, especially for small bias voltages.

For the current through two coupled quantum dots the Tien-Gordon result is perturbative in the interdot coupling, as mentioned above [30–32]. For that case, as in our study, the Tien-Gordon approximation overestimates the current $I_n$. The deviations between the average current $\bar{I}$ and the Tien-Gordon current $I_{TG}$ become larger with increasing bias. In the present study and for a small bias voltage the average current stems only from the main contribution $I_0$ and for this case the Tien-Gordon results match the present results for different laser amplitudes $E_0$ (not shown). For larger bias voltages, the energetic positions of the steps are equal though the Tien-Gordon approach seems to overestimate the contributions from the PAT-induced states $\varepsilon_0 \pm n\omega$.

At first sight it might be astonishing that CDT also works for one site coupled to reservoirs since the infinitely large reservoirs could be expected to destroy any coherent effect. Let us go back to the two-level system [7,9] in which it does not matter whether both or just one level is periodically driven to be able to observe CDT. It is also not important if the static level is replaced by a continuum of states, since the driving only modifies the coupling. As can be seen from the present results the situation is even not changed if the continuum of states is replaced by a reservoir or if a second one is added. In all these situations the coupling between the sites can be suppressed if the correct amplitude of the external field is applied.

Monochromatic laser, $0 < U < \omega$. – In this case the current-voltage curve behaves similar to that for $0 < U < \omega$. Current steps still appear at $V_b/2$ values $n\omega - U$, $n\omega$ and $n\omega + U$. In this case the bias is increased $\omega$. The energy levels $\varepsilon_0 + U - n\omega$ of some conduction channels can be larger than $\varepsilon_0$ and might coincide with the higher Fermi level, but the states with $\varepsilon_0 - n\omega$ have again to be compared to the lower (right) Fermi energy. As shown in fig. 3 for $U = 1.4\omega$ one observes the first shoulder at $V_b/(2\omega) = 0.6, 1.0, 1.4, 1.6, 2.0$, etc. The CDT phenomenon occurs again for small bias voltages and disappears at $V_b/2 = 0.6\omega$. The partial current $I_0$ has, in principle, a shoulder at $V_b/2 = 0.4\omega$ but it is not visible in fig. 2 because $I_0 = 0$ due to CDT.

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Laser pulse. – Above we investigated laser fields with constant amplitude $E_0 = 2.405\omega$, i.e. equal to a zero
of the Bessel function $J_0$. One can also use other external fields with time-dependent envelope function, e.g. with a Gaussian envelope $E_0(t) = 2.405\omega \exp\left(-\frac{(t-t_0)^2}{2\sigma^2}\right)$ 
[20]. For the following example we set $t_0 = 400$ fs and $\sigma = 80$ fs. Since the amplitude of this laser is time-dependent, it induces many more inelastic channel states. During the pulse the laser amplitude changes $E_0$ from 0 to 2.405\omega following the Gaussian shape. At each moment in time there is one resonance state $\varepsilon_0$ and many PAT-induced energy states $\varepsilon_0 \pm n\omega$ with probability $J_0^2(\varepsilon_0/\omega)$. For a small bias voltage only the partial current $I_0$ is non-zero and all other partial currents vanish. So in this case the fulfillment of the CDT condition leads to a complete suppression of the current for $t = t_0$. For larger bias, the CDT condition just suppresses the partial current $I_0$ but not the total current. In fig. 4, when the bias is small, CDT can be seen clearly with and without electron interaction. When the bias is big, CDT is only visible as a small dip in the current. For the case with electron interaction $U = 0.4\omega$, the energy level $\varepsilon_0 + U$ just lines up with the Fermi level for the bias 0.8$\omega$, and the current tunneling through this channel is smaller than the resonance current. This is the reason why the current is somewhat smaller than without electron interaction.

As mentioned above, the average current $\bar{I}$ was obtained by averaging $I(t)$ over five periods of the highly oscillating carrier field. For the laser pulse with time-dependent envelope the current $\bar{I}$ is of course depending on the number $n_{av}$ of periods used in the averaging procedure. In these cases any averaging procedure will not only average over the highly oscillating carrier field but also smooth the envelope function. This effect will become more pronounced for large $n_{av}$. For a Gaussian shape of the laser pulse the averaged current will still be close to a Gaussian form although slightly shifted and unsymmetric. For the example in fig. 4 there are only minor changes when changing $n_{av}$ between 2 and 10. The averaging process does not depend much on $n_{av}$ for monochromatic laser pulses.

**Conclusions.** – To conclude, we have shown that the phenomenon of CDT already appears in molecular junctions which can be modeled by one site coupled to two contacts and not only in systems in which the tunneling is quenched within the molecule itself. Since the total current consists of several partial currents additional channels might be opened by the external field. A complete suppression of the current occurs only for small bias voltages when only the channel for $I_0$ is open and if the amplitude of the external field fulfills the CDT condition. In the present letter we concentrated on the case of a high carrier frequency but as in the case of the two-level system similar effects should be possible for smaller frequencies [33]. The density matrix formalism used here restricts the present results to weak coupling between the leads and the molecular junction. In the case of CDT between two sites the phenomenon does not depend on the size of the coupling. Therefore we believe that the effect of CDT between a single site coupled to one or more leads will also survive for stronger coupling between site and leads. Although the present model calculations certainly can only be a very rough description of real systems, the hope is that the basic physics survives in more realistic and complex systems. Then the effect of CDT with short laser pulses might allow for the construction of fast optoelectronic switches if one finds materials with long enough coherence times.

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