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# Controlling currents through molecular wires

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## Abstract

We investigate the possibility of controlling by means of external laser fields both the current through molecular wires and the corresponding noise properties. It is found that an appropriate off-resonant driving field reduces the coherent transport across the molecule resulting in a strong current suppression. The relative level of transport noise, characterized by a Fano factor, exhibits characteristic maxima and minima near regimes of current suppression. In a three-terminal configuration, the field allows one to steer the current selectively towards one terminal.

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## 1. Introduction

The ongoing experimental progress in the field of molecular electronics [1–4] has revived theoretical interest in the transport properties of molecules which are placed between metallic leads [5]. Indeed during the last few years, the field has progressed rapidly and at present enjoys intensive activity [6, 7]. Tight-binding models for the wire have been used to compute current–voltage characteristics, within a scattering approach [8] and from electron transfer theory [5]. The two approaches have the same essentials [9]. On the other hand, in the limit where the wire electrons lose their quantum coherence, the transport is dominated by incoherent hopping between neighbouring sites [10, 11].

Typical electronic excitation energies in molecules are in the range up to an eV and, thus, correspond to light quanta from the optical and the infrared spectral regime where most of today's lasers work. It is therefore natural to use such coherent light sources to

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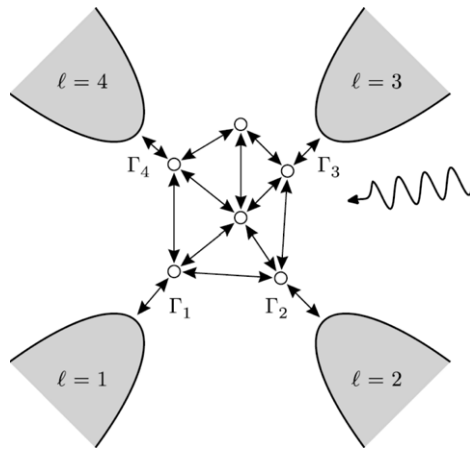


Fig. 1. The molecular circuit consisting of  $N = 6$  sites of which the sites  $1, \dots, L$  are coupled to  $L = 4$  leads.

excite molecules and to study their influence on the transport properties with the goal of selectively manipulating both the current and its noise properties. On the other hand, since such frequencies lie below typical plasma frequencies of metals, the laser light will be reflected at the metal surface, i.e. it does not penetrate the leads. Consequently, we do not expect major changes of the leads' bulk properties—in particular each lead remains close to equilibrium. Moreover, we assume that hot electrons, which might be generated by the impinging laser, do not influence the transport properties significantly. Thus, we consider herein the influence of the driving solely in the molecular Hamiltonian. In addition, the energy of infrared light quanta is far smaller than the work function of a common metal, which is of the order of 5 eV. This prevents the generation of a photocurrent, which otherwise would dominate the effects discussed below.

One particularly prominent example of quantum control is the so-called coherent destruction of tunnelling (CDT), i.e. the suppression of the tunnelling dynamics in an *isolated* bistable potential by the purely coherent influence of an oscillating bias [12, 13]. The crucial point there is that the long-time dynamics in a periodically driven quantum system is no longer dominated by the energies, but rather by the so-called quasienergies [13–15]. The latter may be degenerate for properly chosen driving parameters yielding a divergent timescale. Inspired by these results, we address here the question of controlling by the use of properly tailored laser fields the transport through time-dependent *open* systems, i.e. systems that allow particle exchange with external leads.

Alternatively, the model discussed below can be realized with coherently coupled quantum dots [16, 17]. For these so-called artificial molecules, the relevant excitation frequencies lie in the microwave regime.

## 2. The wire–lead model

We start out by specifying the model Hamiltonian of the entire system as sketched in Fig. 1. This consists of the molecule in the laser field, ideal leads and the molecule–leads

coupling Hamiltonian:  $H(t) = H_{\text{molecule}}(t) + H_{\text{leads}} + H_{\text{molecule-leads}}$ . The irradiated molecule is modelled by a tight-binding description taking into account  $N$  molecular orbitals  $|n\rangle$ , which are relevant for the transport. Disregarding the electron–electron interaction, the most general form of the Hamiltonian reads

$$H_{\text{molecule}}(t) = \sum_{n,n'} H_{nn'}(t) c_n^\dagger c_{n'}, \quad (1)$$

where the fermionic operators  $c_n$  and  $c_n^\dagger$  destroy and create, respectively, an electron in the molecular orbital  $|n\rangle$ . The sums extend over all tight-binding orbitals. The  $\mathcal{T}$ -periodic time dependence of the single-particle Hamiltonian  $H_{nn'}(t) = H_{nn'}(t + \mathcal{T})$ , reflects the influence of the laser field with frequency  $\Omega = 2\pi/\mathcal{T}$ . As discussed above, we assume that the  $L$  leads remain close to equilibrium and hence can be described by grand-canonical ensembles of electrons at temperature  $T$  and electrochemical potential  $\mu_\ell$ ,  $\ell = 1, \dots, L$ . Thus, the lead Hamiltonian reads  $H_{\text{leads}} = \sum_{q\ell} \epsilon_{q\ell} c_{q\ell}^\dagger c_{q\ell}$ , where  $c_{q\ell}$  destroys an electron in state  $q$  in lead  $\ell$ . All expectation values of lead operators can be traced back to  $\langle c_{q\ell}^\dagger c_{q'\ell'} \rangle = \delta_{qq'} \delta_{\ell\ell'} f(\epsilon_{q\ell} - \mu_\ell)$ , where  $f(\epsilon) = (1 + e^{\epsilon/k_B T})^{-1}$  denotes the Fermi function. The model is completed by the molecule–leads tunnelling Hamiltonian

$$H_{\text{molecule-leads}} = \sum_{q\ell} V_{q\ell} c_{q\ell}^\dagger c_\ell + \text{h.c.}, \quad (2)$$

that connects each lead directly to one of the suitably labelled molecular orbitals. Since we are not interested here in the effects that arise from the microscopic details of the molecule–lead coupling, we restrict our analysis in the following to energy-independent couplings, i.e.  $\Gamma_\ell = 2\pi \sum_q |V_{q\ell}|^2 \delta(\epsilon - \epsilon_{q\ell}) = \text{const.}$

### 3. Floquet transport theory

For a two-terminal configuration, one can eliminate the leads to find for the retarded Green function of the wire electrons the equation of motion

$$\left[ \mathcal{H}(t) - i\Sigma - i\hbar \frac{d}{dt} \right] G(t, t') = \delta(t - t'), \quad (3)$$

where  $\mathcal{H}(t) = \sum_{n,n'} |n\rangle H_{nn'}(t) \langle n'|$  and  $2\Sigma = |1\rangle \Gamma_L \langle 1| + |N\rangle \Gamma_R \langle N|$  is the self-energy. For the current defined as the change of the charge in the, e.g., left lead,  $I_L = -e(d/dt) \langle N_L \rangle$ , we find after some algebra that it assumes the commonly expected “scattering form” [18] but with periodically *time-dependent* transmission probabilities and, as well, an additional contribution that accounts for a  $\mathcal{T}$ -periodic charging/discharging of the wire [19]. Only the former contributes to the time-averaged current

$$\bar{I} = \frac{e}{2\pi\hbar} \sum_{k=-\infty}^{\infty} \int d\epsilon \left\{ T_{LR}^{(k)}(\epsilon) f_R(\epsilon) - T_{RL}^{(k)}(\epsilon) f_L(\epsilon) \right\}, \quad (4)$$

where  $T_{LR}^{(k)}(\epsilon) = \Gamma_L \Gamma_R |G_{1N}^{(k)}(\epsilon)|^2$  is the transmission of an electron with energy  $\epsilon$  from the right lead to the left lead under the absorption (emission) of  $|k|$  photons if  $k > 0$  ( $k < 0$ )

and  $T_{RL}^{(k)}(\epsilon)$  is defined accordingly.  $G_{1N}^{(k)}(\epsilon)$  denotes the relevant matrix elements of the Fourier transform

$$G^{(k)}(\epsilon) = \int_0^T \frac{dt}{T} e^{ik\Omega t} \int_0^\infty d\tau e^{i\epsilon\tau/\hbar} G(t, t - \tau) \tag{5}$$

of the retarded Green function. Since the coefficients of the equation of motion (3) are  $T$ -periodic, a complete solution can be constructed with the help of the Floquet ansatz  $|\psi_\alpha(t)\rangle = \exp[-(i\epsilon_\alpha/\hbar - \gamma_\alpha)t] |\Phi_\alpha(t)\rangle$ . The Floquet states  $|\Phi_\alpha(t)\rangle = \sum_k |\Phi_{\alpha k}\rangle \exp(-ik\Omega t)$  obey the time periodicity of the differential equations and fulfil, in a Hilbert space that is extended by a periodic time coordinate, the eigenvalue equation

$$\left[ \mathcal{H}(t) - i\Sigma - i\hbar \frac{d}{dt} \right] |\Phi_\alpha(t)\rangle = (\epsilon_\alpha - i\hbar\gamma_\alpha) |\Phi_\alpha(t)\rangle. \tag{6}$$

Since the eigenvalue equation (6) is non-Hermitian, its eigenvalues  $\epsilon_\alpha - i\hbar\gamma_\alpha$  are generally complex valued and the (right) eigenvectors are not mutually orthogonal. Therefore, we need to solve also the adjoint Floquet equation yielding again the same eigenvalues but providing the adjoint eigenvectors  $|\Phi_\alpha^+(t)\rangle$ . Thus, we find the retarded Green function

$$G(t, t - \tau) = -\frac{i}{\hbar} \sum_\alpha |\psi_\alpha(t)\rangle \langle \psi_\alpha^+(t - \tau)| \Theta(\tau) = G(t + T, t + T - \tau) \tag{7}$$

and, consequently,

$$G_{nn'}^{(k)}(\epsilon) = \sum_{\alpha, k'} \frac{\langle n | \Phi_{\alpha, k'+k} \rangle \langle \Phi_{\alpha, k'}^+ | n' \rangle}{\epsilon - (\epsilon_\alpha + k'\hbar\Omega - i\hbar\gamma_\alpha)}. \tag{8}$$

The current noise is given by the auto-correlation function  $S_L(t, \tau) = (1/2) \langle [\Delta I_L(t), \Delta I_L(t + \tau)]_+ \rangle$  of the current fluctuation operator  $\Delta I_L(t) = I_L(t) - \langle I_L(t) \rangle$ . It can be shown that  $S_L(t, \tau) = S_L(t + T, \tau)$  shares the time periodicity of the driving. Therefore, it is possible to characterize the noise level by the time-averaged noise strength at zero frequency,  $\bar{S}_L = \int d\tau \int_0^T dt S_L(t, \tau) / T$ . Since the total charge is conserved, we find  $\bar{S}_L = \bar{S}_R = \bar{S}$ , where [19]

$$\begin{aligned} \bar{S} = & \frac{e^2 \Gamma_L \Gamma_R}{2\pi \hbar} \sum_k \int d\epsilon \left\{ \Gamma_L \Gamma_R \left| \sum_{k'} G_{N1}^{(k'-k)}(\epsilon_k) \left[ G_{N1}^{(k')}(\epsilon) \right]^* \right|^2 f_L(\epsilon) \bar{f}_L(\epsilon_k) \right. \\ & \left. + \left| \Gamma_L \sum_{k'} G_{1N}^{(k'-k)}(\epsilon_k) \left[ G_{11}^{(k')}(\epsilon) \right]^* - i G_{1N}^{(-k)}(\epsilon_k) \right|^2 f_L(\epsilon) \bar{f}_R(\epsilon_k) \right\} \tag{9} \end{aligned}$$

+ same terms with the replacement  $(L, 1) \leftrightarrow (R, N)$ ,

with  $\bar{f}_{L/R} = 1 - f_{L/R}$  and  $\epsilon_k = \epsilon + k\hbar\Omega$ . Expressions (4) and (9) contain as special cases prior findings: in the absence of any driving, the Floquet eigenvalues  $\epsilon_\alpha - i\hbar\gamma_\alpha$  reduce to the complex-valued eigenenergies; this implies  $G_{nn'}^{(k)} = 0$  for all  $k \neq 0$ , yielding the transmission probability for an electron with energy  $E$  of  $T(E) = \Gamma_L \Gamma_R |G_{N1}^{(0)}(E)|^2$ .

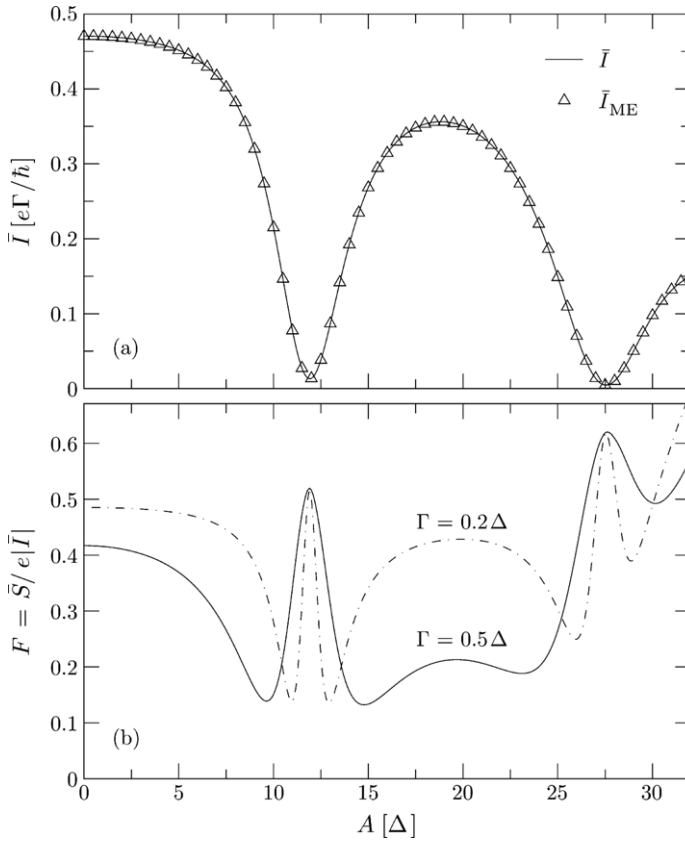


Fig. 2. Time-averaged current  $\bar{I}$  (a) as a function of the driving amplitude  $A$  for a wire with  $N = 3$  sites with on-site energies  $E_n = 0$  and chemical potentials  $\mu_R = -\mu_L = 25\Delta$ . The other parameters are  $\Omega = 5\Delta/\hbar$  and  $\Gamma = 0.5\Delta$ . The triangles mark the results obtained within the master equation approach of [22]. Panel (b) displays the Fano  $F$  factor for these parameters (full curve) and for a smaller wire–lead coupling (dash-dotted curve).

Thus, the quantities  $\bar{I}$  and  $\bar{S}$  agree with the well-known expressions obtained within the time-independent, non-driven scattering approach [20]. For a system for which the ac potential is spatially uniform in the driven region, the average current and the noise strength follow in the weak tunnelling limit already from the static conduction properties [21].

Especially for long wires, the master equation approach presented in [22, 23] allows a more efficient numerical computation. The central idea behind this approach is to solve the coherent dynamics by means of a transformation into the basis given by the coherent Floquet states, i.e. the solution of the eigenvalue problem (6) for  $\Sigma = 0$ . In this basis, a master equation for the occupation of the Floquet states can be derived. Its solution describes the state of the wire at asymptotic times and thus, in particular, the corresponding current. In the situation considered below (cf. Fig. 2(a)), the two results are in perfect agreement.

## 4. Laser-controlled current

### 4.1. Current and noise suppression in two-terminal devices

As a simple, yet nontrivial application, we consider a wire composed of  $N = 3$  sites. Each orbital is coupled to its nearest neighbours by a hopping matrix element  $\Delta$ . The on-site energies are modulated by the influence of the ac dipole field,  $H_{nn}(t) = E_n - A \cos(\Omega t)(2 - n)$ ,  $n = 1, 2, 3$ . The energy  $A$  equals the electric field strength multiplied by the electron charge  $-e$  and the distance between two neighbouring sites. The wire is assumed to couple equally to the two leads,  $\Gamma_L = \Gamma_R = \Gamma$ , and the laser frequency is far off resonance,  $\Omega = 5\Delta/\hbar$ . For a molecular wire, a typical value for the hopping matrix element  $\Delta$  and the coupling strength  $\Gamma$  is 0.1 eV, leading to a current unit  $e\Gamma/\hbar \simeq 25 \mu\text{A}$ , while the laser frequency lies in the optical regime. For a distance of  $2 \text{ \AA}$  between two neighbouring sites, a driving amplitude  $A = \Delta$  corresponds to an electric field strength of roughly  $5 \times 10^6 \text{ V/cm}$ . For the evaluation of the current  $\bar{I}$  and the noise  $\bar{S}$ , we restrict ourselves to zero temperature. Then, the Fermi functions turn into step functions and the energy integrations in equations Eqs. (4) and (9) can be carried out analytically. This limit is physically well justified for molecular wires at room temperature and for quantum dots at helium temperature since, in both cases, thermal electron excitations do not play a significant role.

Fig. 2(a) depicts the dc current and the zero-frequency noise for a wire with equal on-site energies  $E_n = 0$  and a relatively large applied voltage. As a remarkable feature, we find that for certain values of the field amplitude  $A$ , the current drops to a value of some percent of the current in the absence of the field [24]. A perturbation theory for the Floquet equation (6) for  $\Delta, \Gamma \ll \hbar\Omega$  yields that the driving results in a renormalized hopping matrix element  $\Delta \rightarrow \Delta_{\text{eff}} = J_0(A/\hbar\Omega)\Delta$ , where  $J_0$  denotes the zeroth-order Bessel function. Then,  $G_{1N}$  and  $G_{N1}$  vanish if the condition  $J_0(A/\hbar\Omega) = 0$  is fulfilled [25, 26]. Consequently, the dc current (4) and the zero-frequency noise (9) also vanish.

The relative noise strength can be characterized by the so-called Fano factor  $F = \bar{S}/e|\bar{I}|$  depicted in Fig. 2(b). Interestingly enough, we find that the Fano factor exhibits, as a function of the driving amplitude  $A$ , both a sharp maximum at current suppression and two pronounced minima nearby. For a sufficiently large voltage, the Fano factor assumes at the maximum the value  $F \approx 1/2$ . Once the driving amplitude is of the order of the applied voltage, however, the Fano factor becomes much larger. The relative noise minima are distinct and provide a typical Fano factor of  $F \approx 0.15$ . Reducing the coupling to the leads renders these phenomena even more pronounced, since then the suppressions occur in a smaller interval of the driving amplitude; cf. Fig. 2(b). The overall behaviour is robust in the sense that approximately the same values for the minima and the maximum are also found for larger wires, different driving frequencies, different coupling strengths and slightly modified on-site energies, provided that  $\Delta, \Gamma, E_n \ll \hbar\Omega$  and that the applied voltage is sufficiently large. The qualitative behaviour can again be understood within a perturbative approach. With increasing driving amplitude, a crossover from  $\Delta_{\text{eff}} \gg \Gamma$  to  $\Delta_{\text{eff}} \ll \Gamma$  at the current suppression occurs. Both limits correspond to the transport through a symmetric double barrier and, therefore, are characterized by  $F \approx 1/2$  [20].

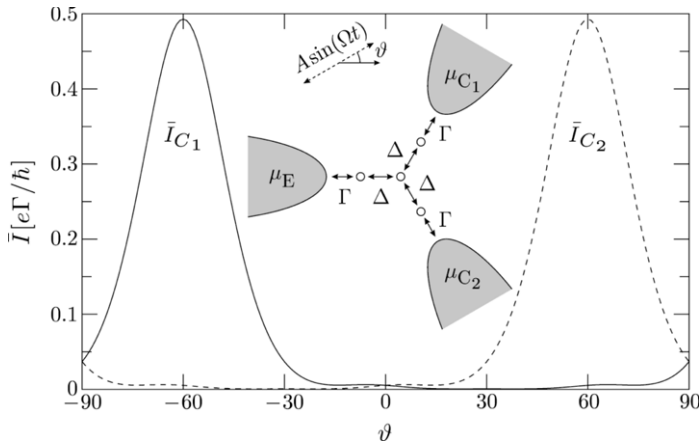


Fig. 3. Average currents through contacts  $C_1$  (solid) and  $C_2$  (broken) as a function of the polarization angle  $\vartheta$  for the three-terminal device depicted in the inset. The chemical potentials are  $\mu_E = -\mu_{C_1} = -\mu_{C_2} = 50\Delta$ ; the on-site energies  $E_n = 0$ . The driving field is specified by the strength  $A = 25\Delta$  and the angular frequency  $\Omega = 10\Delta/\hbar$ ; the effective coupling is  $\Gamma = 0.1\Delta$  and the temperature  $k_B T = 0.25\Delta$ .

At the crossover  $\Delta_{\text{eff}} \approx \Gamma$  the effective barriers vanish and, consequently, the Fano factor assumes its minimum.

#### 4.2. Current router

An experimentally more ambitious configuration consists in a planar three-terminal configuration with  $N = 4$  sites. We borrow from electrical engineering the designations E,  $C_1$  and  $C_2$ . Here, an external voltage is always applied such that  $C_1$  and  $C_2$  have equal electrochemical potential, i.e.  $\mu_{C_1} = \mu_{C_2} \neq \mu_E$ . In a perfectly symmetric molecule, where all on-site energies are equal to each other, reflection symmetry at the horizontal axis ensures that any current which enters at E is equally distributed among  $C_{1,2}$ ; thus  $I_{C_1} = I_{C_2} = -I_E/2$ .

The fact that this structure is essentially two-dimensional brings about a new degree of freedom, the polarization of the laser field. We assume it to be linear with a polarization angle  $\vartheta$  as sketched in the inset of Fig. 3. The effective driving amplitudes of the orbitals that are attached to the leads acquire now a geometric factor which is only the same for the two orbitals  $C_1$  and  $C_2$  when  $\vartheta = 0$ . For any other polarization angle, the above mentioned symmetry is broken and the outgoing currents may be different from each other. The difference may be huge, as depicted in Fig. 3. Their ratio varies from unity for  $\vartheta = 0^\circ$  up to the order of 100 for  $\vartheta = 60^\circ$ . Thus, adapting the polarization angle enables one to route the current towards one or the other drain.

For a qualitative explanation of the mechanism behind this effect, it is instructive to look at the time averages of the overlaps  $|\langle n | \Phi_\alpha(t) \rangle|^2$  of the Floquet states and the terminal sites  $n = E, C_1, C_2$ . Fig. 4 shows these overlaps for three different polarization angles  $\vartheta$ . Let us consider, for instance, the current across contact  $C_1$ . It is plausible that only Floquet modes which have substantial overlap with both the site  $C_1$  and also the site E contribute to the current through these terminals. For a polarization angle  $\vartheta = -60^\circ$ , we

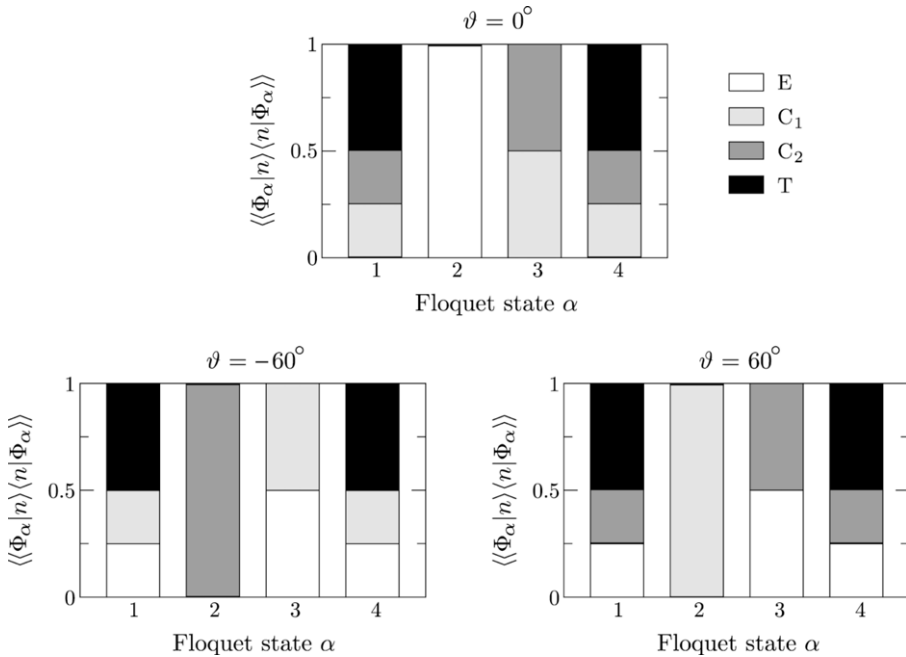


Fig. 4. The time average  $\langle\langle \Phi_\alpha(t) | n \rangle \langle n | \Phi_\alpha(t) \rangle\rangle$  of the overlaps  $| \langle n | \Phi_\alpha(t) \rangle |^2$  of the sites  $n = E, C_1, C_2$  and T (central site) with a Floquet state  $|\Phi_\alpha(t)\rangle$  for three different polarization angles  $\vartheta$ . All parameters are as in Fig. 3.

can infer from Fig. 4 that the Floquet states with indices  $\alpha = 1, 3$  and 4 fulfil this condition and, consequently, a current flows from lead E into lead  $C_1$ . By contrast, for  $\vartheta = 0^\circ$  and  $\vartheta = 60^\circ$  such current carrying states do not exist and the respective current vanishes.

## 5. Conclusions

The transport properties of molecular wires and coherently coupled quantum dots can be influenced significantly by ac fields. In particular, such an external driving can be used to selectively suppress both the current and its noise. Investigating the *relative* noise level characterized by the Fano factor has revealed that the current suppression is accompanied by a noise maximum and two remarkably low minima. The underlying physics is dominated by *coherent destruction of tunnelling* for the electrons on the molecule. These phenomena can be used to devise novel current sources with controllable noise levels. In a three-terminal device, the polarization of the laser field can be adjusted such that single exits can be selectively closed. Then, the current is routed through the respective other terminals.

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## References

- [1] M.A. Reed, C. Zhou, C.J. Muller, T.P. Burgin, J.M. Tour, *Science* 278 (1997) 252.
- [2] X.D. Cui, A. Primak, X. Zarate, J. Tomfohr, O.F. Sankey, A.L. Moore, T.A. Moore, D. Gust, G. Harris, S.M. Lindsay, *Science* 294 (2001) 571.
- [3] C. Joachim, J.K. Gimzewski, A. Aviram, *Nature* 408 (2000) 541.
- [4] J. Reichert, R. Ochs, D. Beckmann, H.B. Weber, M. Mayor, H. von Löhneysen, *Phys. Rev. Lett.* 88 (2002) 176804.
- [5] A. Nitzan, *Annu. Rev. Phys. Chem.* 52 (2001) 681.
- [6] P. Hänggi, M. Ratner, S. Yaliraki (Eds.), *Processes in Molecular Wires*, *Chem. Phys.* 281 (2002) 111–502 (special issue).
- [7] A. Nitzan, M.A. Ratner, *Science* 300 (2003) 1384.
- [8] V. Mujica, M. Kemp, M.A. Ratner, *J. Chem. Phys.* 101 (1994) 6849.
- [9] A. Nitzan, *J. Phys. Chem. A* 105 (2001) 2677.
- [10] E.G. Petrov, P. Hänggi, *Phys. Rev. Lett.* 86 (2001) 2862.
- [11] J. Lehmann, G.-L. Ingold, P. Hänggi, *Chem. Phys.* 281 (2002) 199.
- [12] F. Grossmann, T. Dittrich, P. Jung, P. Hänggi, *Phys. Rev. Lett.* 67 (1991) 516.
- [13] M. Grifoni, P. Hänggi, *Phys. Rep.* 304 (1998) 229.
- [14] J.H. Shirley, *Phys. Rev.* 138 (1965) B979.
- [15] H. Sambe, *Phys. Rev. A* 7 (1973) 2203.
- [16] R.H. Blick, R.J. Haug, J. Weis, D. Pfannkuche, K. von Klitzing, K. Eberl, *Phys. Rev. B* 53 (1996) 7899.
- [17] T.H. Oosterkamp, T. Fujisawa, W.G. van der Wiel, K. Ishibashi, R.V. Hijman, S. Tarucha, L.P. Kouwenhoven, *Nature* 395 (1998) 873.
- [18] S. Datta, *Electronic Transport in Mesoscopic Systems*, Cambridge University Press, Cambridge, 1995.
- [19] S. Camalet, J. Lehmann, S. Kohler, P. Hänggi, *Phys. Rev. Lett.* 90 (2003) 210602.
- [20] Ya.M. Blanter, M. Büttiker, *Phys. Rep.* 336 (2000) 1.
- [21] J.R. Tucker, *IEEE J. Quantum Electron.* QE-15 (1979) 1234.
- [22] J. Lehmann, S. Kohler, P. Hänggi, A. Nitzan, *Phys. Rev. Lett.* 88 (2002) 228305.
- [23] J. Lehmann, S. Kohler, P. Hänggi, A. Nitzan, *J. Chem. Phys.* 118 (2003) 3283.
- [24] J. Lehmann, S. Camalet, S. Kohler, P. Hänggi, *Chem. Phys. Lett.* 368 (2003) 282.
- [25] F. Großmann, P. Jung, T. Dittrich, P. Hänggi, *Z. Phys. B* 84 (1991) 315.
- [26] F. Großmann, P. Hänggi, *Europhys. Lett.* 18 (1992) 571.