



# Laser controlled molecular switches and transistors

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Received 27 May 2002; in final form 7 November 2002

## Abstract

We investigate the possibility of optical current control through single molecules which are weakly coupled to leads. A master equation approach for the transport through a molecule is combined with a Floquet theory for the time-dependent molecule. This yields an efficient numerical approach to the evaluation of the current through time-dependent nano-structures in the presence of a finite external voltage. We propose tunable optical current switching in two- and three-terminal molecular electronic devices driven by properly adjusted laser fields, i.e., a novel class of molecular transistors.

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## 1. Introduction

Spurred by the ongoing experimental progress in the field of molecular electronics [1–4], theoretical interest in the transport properties of molecules which are placed between lead electrodes has revived [5]. Indeed, since the pioneering work on molecular rectifiers by Aviram and Ratner [6], the field has progressed rapidly and presently enjoys intense activity [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]. Both approaches bear the same essentials [9]. For high temperatures, the wire electrons lose their quan-

tum coherence and 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 excite molecules and to study their influence on the transport properties with the goal to selectively manipulate electronic currents. 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

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properties significantly. Thus, we consider herein the influence of the driving solely in the molecule Hamiltonian. In addition, the energy of infrared light quanta is by 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 [12–16]. 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 quasi-energies [16–18]. The latter may be degenerate for properly chosen driving parameters yielding a divergent time-scale. Inspired by these results, we address in this Letter the question of controlling by use of properly tailored laser fields the transport through time-dependent *open* systems, i.e., systems that allow particle exchange with external leads.

For the computation of electrical currents through wires exposed to strong laser fields, we put forward a Floquet approach [19,20]. The central idea of this method lies in a non-perturbative solution of the Schrödinger equation of the isolated time-dependent wire plus laser field, while the wire-lead coupling is treated perturbatively. This corresponds to the physical assumption that the molecular orbitals that are relevant for the transport are weakly coupled to the leads. The resulting density operator equation is decomposed into a time-dependent Floquet basis permitting a numerically efficient treatment. We generalise here this method to the analysis of networks with an arbitrary number of contacts to leads. Subsequently, we apply the so obtained formalism to the investigation of optical current switching in two- and three-terminal devices as prototypical examples of a new class of molecular transistors.

A heuristic approach to the problem of transport through driven molecular wires has recently been introduced in [21], where the assumption is made that each sideband of a Floquet state forms an independent conductance channel. This

approximation neglects coherences between different Floquet states and between different sidebands of a Floquet state. These coherences are, however, crucial for the effects discussed in the present work.

## 2. Model for wire and leads

We embark by specifying the model Hamiltonian of the entire system as sketched in Fig. 1. It consists of the molecule in the laser field, ideal leads, and the molecule-leads coupling Hamiltonian,

$$H(t) = H_{\text{molecule}}(t) + H_{\text{leads}} + H_{\text{molecule-leads}}. \quad (1)$$

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

$$H_{\text{molecule}}(t) = \sum_{n,n'} H_{nn'}(t) c_n^\dagger c_{n'}, \quad (2)$$

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

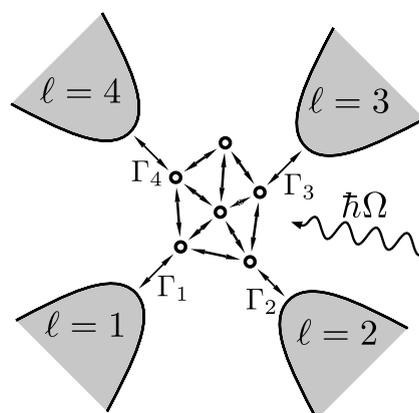


Fig. 1. Molecular circuit consisting of  $N = 6$  sites of which the sites  $1, \dots, L$  are coupled to  $L = 4$  leads.

assume that the  $L$  leads remain close to equilibrium and hence can be described by grand-canonical ensembles of electrons at temperature  $T$  and electro-chemical potential  $\mu_l$ ,  $l = 1, \dots, L$ . Thus, the lead Hamiltonian reads  $H_{\text{leads}} = \sum_{ql} \epsilon_{ql} c_{ql}^\dagger c_{ql}$ , where  $c_{ql}$  destroys an electron in state  $q$  in lead  $l$ . All expectation values of lead operators can be traced back to  $\langle c_{ql}^\dagger c_{ql'} \rangle = \delta_{ql'} \delta_{ll'} f(\epsilon_{ql} - \mu_l)$ , where  $f(\epsilon) = (1 + e^{\epsilon/k_B T})^{-1}$  denotes the Fermi function. The model is completed by the molecule-leads tunnelling Hamiltonian

$$H_{\text{molecule-leads}} = \sum_{ql} V_{ql} c_{ql}^\dagger c_l + \text{h.c.} \quad (3)$$

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 [22], we restrict our analysis in the following to energy-independent couplings, i.e.,  $\Gamma_l = (2\pi/\hbar) \sum_q |V_{ql}|^2 \delta(\epsilon - \epsilon_{ql})$ .

### 3. Perturbative description and Floquet ansatz

Let us assume that the dynamics of the driven wire is dominated by the time-dependent wire Hamiltonian so that the coupling to the leads can be taken into account as a perturbation. This allows to derive by use of standard methods the approximate equation of motion for the total density operator  $\varrho(t)$ ,

$$\begin{aligned} \dot{\varrho}(t) = & -\frac{i}{\hbar} [H_{\text{molecule}}(t) + H_{\text{leads}}, \varrho(t)] \\ & -\frac{1}{\hbar^2} \int_0^\infty d\tau \left[ H_{\text{molecule-leads}}, \right. \\ & \left. [\tilde{H}_{\text{molecule-leads}}(t - \tau, t), \varrho(t)] \right]. \end{aligned} \quad (4)$$

We have omitted a transient term that depends purely on the initial preparation. The tilde denotes operators in the interaction picture with respect to the molecule plus the lead Hamiltonian,  $\tilde{O}(t, t') = U_0^\dagger(t, t') O(t) U_0(t, t')$ , where  $U_0(t, t')$  is the time-evolution operator without the coupling. The first term describes the coherent dynamics of the electrons on the wire, while the second term represents incoherent hopping of electrons between the leads and the wire.

The net (incoming minus outgoing) electrical current that flows from lead  $l$  into the molecule is then given by the rate at which the electron number in the corresponding lead decreases multiplied by the electron charge  $-e$ ,

$$I_l(t) = e \frac{d}{dt} \langle N_l \rangle. \quad (5)$$

Note that this expectation value is time-dependent through the non-equilibrium density operator  $\varrho(t)$ . To evaluate the right-hand side of Eq. (5), we employ Eq. (4) to derive after some algebra the result

$$\begin{aligned} I_l(t) = & -e\Gamma_l \left\{ \text{Re} \int_0^\infty \frac{d\tau}{\hbar} \int \frac{d\epsilon}{\pi} e^{i\epsilon\tau/\hbar} f(\epsilon - \mu_l) \right. \\ & \left. \times \left[ c_l, \tilde{c}_l^\dagger(t - \tau, t) \right]_+ - \langle c_l^\dagger c_l \rangle \right\}. \end{aligned} \quad (6)$$

Because of the time-periodicity of the wire Hamiltonian  $H_{\text{mw}}(t)$ , there exists a complete set of solutions of the corresponding one-particle Schrödinger equation of the form  $|\Psi_\alpha(t)\rangle = \exp(-i\epsilon_\alpha t/\hbar) |\Phi_\alpha(t)\rangle$ . The so-called quasi-energies  $\epsilon_\alpha$  take over the role of the energy eigenvalues in static systems and govern the long-time dynamics. The Floquet modes  $|\Phi_\alpha(t)\rangle$  obey the time-periodicity of the driving field which allows to express them as a Fourier series,  $|\Phi_\alpha(t)\rangle = \sum_{k=-\infty}^\infty \exp(-ik\Omega t) |\Phi_{\alpha,k}\rangle$ . They can be obtained from the eigenvalue equation [16–18]

$$\left( \sum_{n,n'} |n\rangle H_{nn'}(t) \langle n'| - i\hbar \frac{d}{dt} \right) |\Phi_\alpha(t)\rangle = \epsilon_\alpha |\Phi_\alpha(t)\rangle. \quad (7)$$

Moreover, the Floquet modes define the complete set of operators

$$c_\alpha(t) = \sum_n \langle \Phi_\alpha(t) | n \rangle c_n, \quad (8)$$

whose time-evolution assumes the convenient form  $\tilde{c}_\alpha(t - \tau, t) = \exp(i\epsilon_\alpha \tau/\hbar) c_\alpha(t)$ . The orthogonality of the Floquet states at equal times [16–18] yields the back-transformation  $c_n = \sum_\alpha \langle n | \Phi_\alpha(t) \rangle c_\alpha(t)$  and thus results in the required spectral decomposition. Using (8) and performing the energy and

the  $\tau$ -integration in Eq. (6), we obtain for the time-averaged current the main result

$$\bar{I}_l = -\frac{e\Gamma_l}{\hbar} \sum_{\alpha k} \left[ \langle \Phi_{\alpha,k} | l \rangle \langle l | \Phi_{\alpha,k} \rangle f(\epsilon_\alpha + k\hbar\Omega - \mu_l) - \sum_{\beta k'} \langle \Phi_{\alpha,k'+k} | l \rangle \langle l | \Phi_{\beta k'} \rangle R_{\alpha\beta,k} \right]. \quad (9)$$

Here, we have introduced the expectation values  $R_{\alpha\beta}(t) = \langle c_\alpha^\dagger(t)c_\beta(t) \rangle$ , which assume in the long-time limit the time-periodicity of the driving force and thus can be decomposed into the Fourier series  $R_{\alpha\beta}(t) = \sum_k \exp(-ik\Omega t) R_{\alpha\beta,k}$ . It is straightforward to derive for the  $R_{\alpha\beta,k}$  from the density operator equation (4) the following set of inhomogeneous linear equations:

$$\begin{aligned} & \frac{i}{\hbar} (\epsilon_\alpha - \epsilon_\beta + k\hbar\Omega) R_{\alpha\beta,k} \\ &= \frac{1}{2} \sum_{lk'} \Gamma_l \left\{ \sum_{\beta'k''} \langle \Phi_{\beta,k''+k'} | l \rangle \langle l | \Phi_{\beta',k''+k} \rangle R_{\alpha\beta',k'} \right. \\ &+ \sum_{\alpha'k''} \langle \Phi_{\alpha',k''+k'} | l \rangle \langle l | \Phi_{\alpha,k''+k} \rangle R_{\alpha'\beta,k'} \\ &- \langle \Phi_{\beta,k'-k} | l \rangle \langle l | \Phi_{\alpha,k'} \rangle f(\epsilon_\alpha + k'\Omega - \mu_l) \\ &\left. - \langle \Phi_{\beta,k'} | l \rangle \langle l | \Phi_{\alpha,k'+k} \rangle f(\epsilon_\beta + k'\Omega - \mu_l) \right\}, \quad (10) \end{aligned}$$

which will be solved numerically. We have found that even in the case of strong driving where the Floquet states comprise many sidebands, a few Fourier coefficients  $R_{\alpha\beta,k}$  are in fact sufficient to obtain numerical convergence. This justifies a posteriori the use of the Floquet states as a basis set.

#### 4. Optical current gate

As a first setup that may be suitable as a current control device, we investigate the transport through a two-level system, i.e., a wire that consists of  $N = 2$  sites – one of them is coupled to the left lead and the other to the right lead. Then the time-dependent wire Hamiltonian reads in the basis of the molecular orbitals

$$H_{\text{molecule}}(t) = \begin{pmatrix} E_L & -\Delta \\ -\Delta & E_R \end{pmatrix} + \frac{A}{2} \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} \cos(\Omega t), \quad (11)$$

where  $\Delta$  denotes the tunnel matrix element between the two sites and  $E_L$  and  $E_R$  are the corresponding on-site energies. The laser field contributes to the Hamiltonian (11) a time-dependent bias with amplitude  $A = -e\mathcal{E}d$ , i.e., charge times electrical field strength times the site-to-site distance. Note that the electrical field may be drastically enhanced due to the presence of the metallic tips [23]. The effective coupling to each lead is assumed to be equal,  $\Gamma_l = \Gamma$ , and an external voltage  $V$  is taken into account by a difference in the electro-chemical potentials,  $\mu_L - \mu_R = -eV$ .

We use in all numerical calculations the tunnel matrix element  $\Delta$  as the energy unit and assume that the effective couplings to the leads are by one order of magnitude smaller,  $\hbar\Gamma = 0.1\Delta$ . This corresponds to a large contact resistance and ensures the applicability of a perturbational approach. A realistic value is  $\Delta = 0.1$  eV, resulting in a current unit  $e\Gamma = 0.256$   $\mu\text{A}$ . For a site-to-site distance of 10 Å and a laser frequency  $\Omega = 10\Delta/\hbar$ , the driving amplitudes considered below correspond to an electrical field amplitude of 10<sup>6</sup> V/cm at 1  $\mu\text{m}$  wavelength.

The time-averaged current  $\bar{I} = \bar{I}_L = -\bar{I}_R$  through the molecule in a case where both on-site energies are equal is depicted in Fig. 2. As a striking feature, we find that at certain values of the driving amplitude, the current collapses to less than 1% of its maximal value reached in the absence of the driving. Closer inspection (not shown) reveals that the width of this depression is proportional to the molecule-lead coupling  $\Gamma$ . Comparison with the quasi-energy spectrum in the lower panel demonstrates that the current break downs occur at quasi-energy crossings. This relates the present phenomenon to the CDT, i.e., the standstill of the tunnel dynamics in a driven bistable potential at quasi-energy crossings [12]. For the *isolated* two-level system (11) with  $\Delta \ll \hbar\Omega, A$ , the CDT condition reads  $J_0(A/\hbar\Omega) = 0$  [13], i.e., the suppression of the tunnelling dynamics is related to the zeros of the Bessel function  $J_0$ . As our analysis reveals, the

same condition results in a suppression of the transport through the *open* system.

An external voltage may be of peculiar influence to the on-site energies of a molecular wire [24] and cause an effective bias  $E_L \neq E_R$  in originally symmetric molecules. Thus, a crucial question is whether the above current suppression is stable against such a modification. The broken lines in Fig. 2 demonstrate that this is indeed the case. Although the quasi-energies now form an avoided crossing, the current breakdowns do survive; they are even more pronounced, but slightly shifted towards larger driving amplitudes. This robustness of CDT based current control combined with the huge on/off ratio suggests the presented setup as a promising alternative to structural chemistry-based switching devices [25,26].

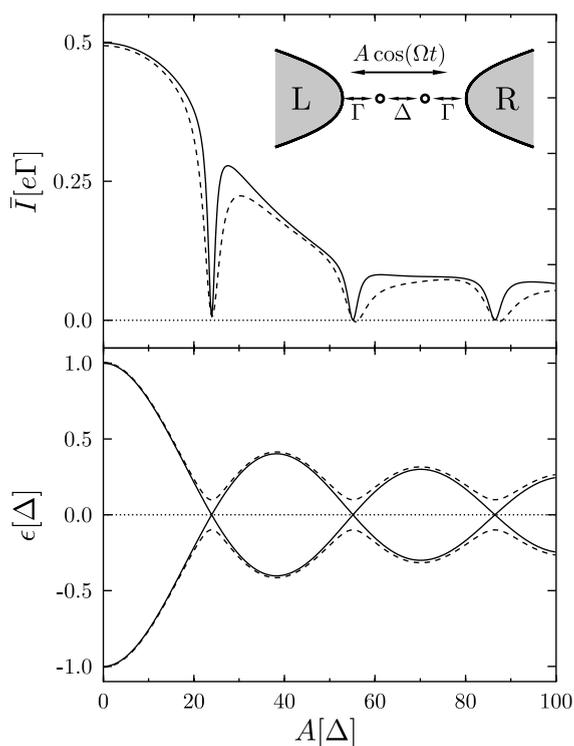


Fig. 2. Average current and quasi-energy spectrum versus driving amplitude for a wire which consists of two sites between two electrodes (cf. inset) for unbiased ( $E_R = E_L = 0$ , solid lines) and biased ( $E_R = -E_L = 0.1A$ , dashed lines) on-site energies. The leads' chemical potentials are  $\mu_R = -\mu_L = 10A$ ; the other parameters read  $\hbar\Omega = 10A$ ,  $k_B T = 0.25A$ ,  $\hbar\Gamma = 0.1A$ .

## 5. Optical current router

An experimentally more ambitious configuration consists in a planar molecule with  $N = 4$  sites, three of which are coupled to a central site and are directly connected to leads (cf. inset of Fig. 3). We borrow from electrical engineering the designation E,  $C_1$ , and  $C_2$ . Here, an external voltage is always applied such that  $C_1$  and  $C_2$  have equal electro-chemical potential, i.e.,  $\mu_{C_1} = \mu_{C_2} \neq \mu_E$ . In a perfectly symmetric molecule, where all on-site energies equal each other, reflection symmetry at the horizontal axis ensures that any current which enters at E, is equally distributed among  $C_{1,2}$ , thus  $I_{C_1} = I_{C_2} = -I_E/2$ . Since this structure in Fig. 3 is essentially two-dimensional, we have to take also the polarisation of the laser field into account. We assume it to be linear with a polarisation angle  $\phi$  as sketched in the inset of Fig. 3. The effective driving amplitudes of the orbitals which are attached to the leads acquire now a geometric factor which is only the same for both orbitals  $C_1$  and  $C_2$  when  $\phi = 0$ . For any other polarisation angle, the 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

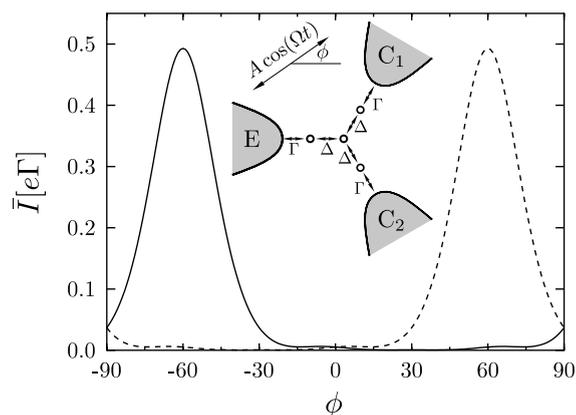


Fig. 3. Average currents through contacts  $C_1$  (solid) and  $C_2$  (broken) as a function of the polarisation angle  $\phi$  for the three-terminal device depicted in the inset. The chemical potentials are  $\mu_E = -\mu_{C_1} = -\mu_{C_2} = 50A$ ; the on-site energies  $E_n = 0$ . The driving field is specified by the strength  $A = 25A$  and the angular frequency  $\Omega = 10A/\hbar$ ; the effective coupling is  $\hbar\Gamma = 0.1A$  and the temperature  $k_B T = 0.25A$ . The maximal value of the current ratio  $I_{C_1}/I_{C_2} \approx 100$  is assumed at  $\phi = 60^\circ$ .

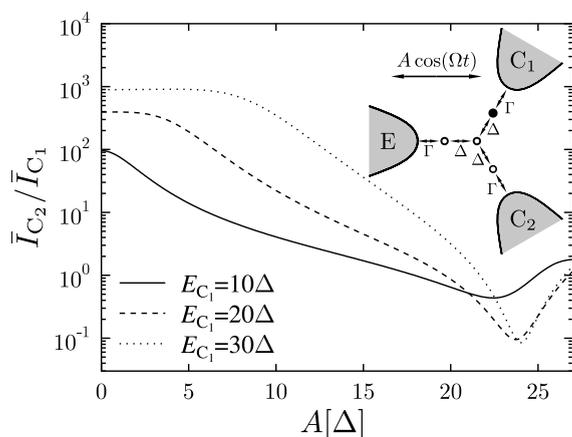


Fig. 4. Ratio of the outgoing average currents versus driving strength  $A$  for the three-terminal device at a polarisation angle  $\phi = 0$ . The filled circle in the inset depicts a site with an on-site energy  $E_{C_1}$  that differs from the others. All other on-site energies and parameters as in Fig. 3.

ratio varies from unity for  $\phi = 0$  up to the order of 100 for  $\phi = 60^\circ$ . Thus, adapting the polarisation angle enables one to route the current towards the one or the other drain.

Alternatively, one can keep the polarisation angle at  $\phi = 0$  and break the reflection symmetry by using an intrinsically asymmetric molecule, as sketched in the inset of Fig. 4. This allows to control sensitively the ratio of the outgoing currents by the strength  $A$  of the external field, cf. Fig. 4. The switching range comprises up to four orders of magnitude with an exponential sensitivity.

## 6. Concluding remarks

We have presented a method for the efficient numerical computation of currents through periodically time-dependent networks with two or more contacts to external leads. The application to two types of setups substantiated that external fields bear a wealth of possibilities for the manipulation of electrical currents: in a molecular wire the current can be suppressed by proper time-dependent fields. In a three-terminal device, it is possible to route by tailored optical fields the current that enters from a source towards the one or the other drain.

The authors hope that their proposals will motivate experimentalists to accept the challenge of implementing the proposed molecular wire schemes in the laboratory. The two-terminal current gate can possibly be realised using break junctions exposed to a laser field. Alternatively, one could use a self-assembled, laser-irradiated maze-like layer of sparsely distributed conducting molecules on a conducting surface. Then by positioning a scanning tunnelling microscope tip directly over one such molecule, it should be possible to measure the features of the predicted gating behaviour. Experimentally more ambitious is the realisation of the arrangement in Fig. 3 with a planar molecule contacted to three electrodes. Here again, laser-irradiated self-assemblies of molecules such as carbon nanotube complexes or of biomolecules like metalised DNA [27], or the use of cationic lipid–DNA complexes [28] as DNA-nanocables, with the centre-molecule covalently bound to such planar structures, might make the experiment feasible.

A completely different realisation of our findings should be possible in semiconductor heterostructures. There, instead of a molecule, coherently coupled quantum dots [29] form the central system. Furthermore, owing to the lower level spacings, the suitable frequency of the coherent radiation source is then in the microwave spectral range.

## Acknowledgements

This work has been supported by the Deutsche Forschungsgemeinschaft through SFB 486 and by the Volkswagenstiftung under Grant No. I/77 217. One of us (S.C.) has been supported by a European Community Marie Curie Fellowship.

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