

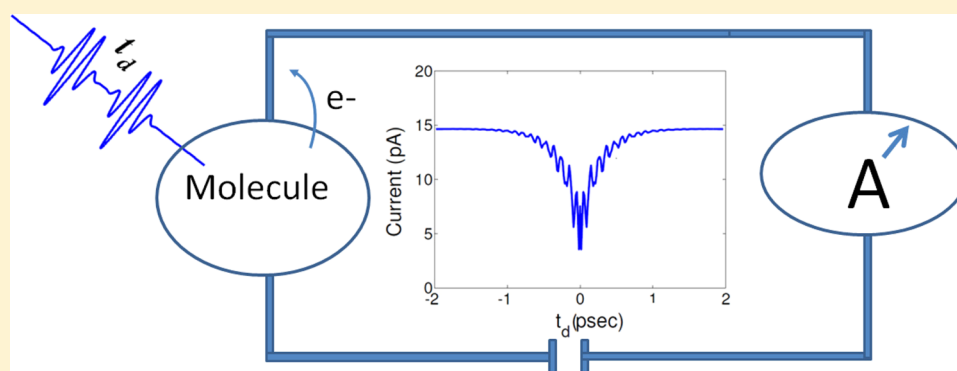
Transient Dynamics in Molecular Junctions: Picosecond Resolution from dc Measurements by a Laser Pulse Pair Sequence Excitation

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ABSTRACT: Molecular electronic devices are a promising future alternative to standard electronic switches due to their fast response on the pico-second time scale. An experimental scheme is proposed in order to correlate pico-second dynamics in molecular junctions to steady state current measurements. The time resolution is obtained using a sequence of ultrafast pulse pair excitations with a controlled time delay between the two pulses in each pair. The dependence of the steady state current on the delay time reveals the periods of molecular dynamics on the sub-pico-second time scale. The approach is demonstrated theoretically for a generic model of a single molecule based coherent electron pump. Theoretical analysis enables to correlate the steady state current to the underlying intramolecular dynamics.

I. INTRODUCTION

Electronic transport through molecular wires and junctions has been attracting much attention in recent years due to remarkable experimental and theoretical advances.^{1–13} The molecular junction set up, in which a small quantum mechanical system is at the bottleneck of charge and energy transport between macroscopic reservoirs, reveals the unique physics of quantum systems out of equilibrium. The field has by now matured to a level where statistically reliable measurements of single molecule junctions can be performed in order to explore the effect of various parameters^{9,14–19} on their conductance properties. Yet, our current understanding of molecular junctions is based on steady state measurements, with quite limited accessibility to the underlying transient phenomena which control the steady state observables.

An essential motivation to understand the transient behavior of molecular junctions is technological. While standard ultrafast electronic components are operating in the Gigahertz regime, molecular components with their characteristic electronic and vibronic response times are expected to be switchable within picoseconds, that is, in the Terahertz regime, suggesting an

apparent 3 orders of magnitude improvement in the speed of operation. These estimates of intramolecular dynamics time scales were not yet tested under nonequilibrium transport conditions, with its unique statistical and dynamical characteristics.^{20–26} Obviously, experimental detection on these time scales is a major challenge of the field, because the state-of-the-art electronic components are much too slow to measure directly dynamics with pico-second resolution.

Insight into the dynamical response of molecular junctions was gained by attempting to control the transport with external laser fields. The effect of radiation on current through molecular junctions was investigated experimentally and studied theoretically,^{27,28} suggesting dependence of the steady state (dc) current on the external field parameters (frequency, intensity, and pulse duration). Because quantum mechanics dominates the intrinsic molecular time scales, special attention

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was given to phase preserving excitations (coherent control) of the molecule by the field, and theoretical propositions of coherently controlled molecular rectifiers, switches, and electron pumps were discussed.^{29–37} Most of the analysis relates to periodic (ac) fields,^{27–29,38–42} which directly excite the molecule, the leads, or both, leading to a steady-state response. A smaller number of studies^{32–37,43–50} relates to pulse excitations and to the transient field-induced phenomena.

In this work a new scheme is proposed for targeting the intramolecular pico-second dynamics time scale in molecular junctions. The scheme is based on a coherent excitation of the molecular system using a periodic Laser Pulse Pairs Sequence (LPPS) with a controlled time delay between the two pulses in each pulse pair, t_d . The induced steady state current, I_{dc} , is used as a probe, and the intramolecular time scale is expected to be revealed in correlation plots, $I_{dc}(t_d)$, circumventing the limited time resolution of state of the art electronics. The LPPS method is closely related to various spectroscopic methods to analyze molecular dynamics by sequences of pump–probe laser pulses.⁵¹ The difference, however, is in the probing scheme. While in all other methods probing is based on the *spectral content* of the probing pulse, in the LPPS, the measured signal is electrical current, that is, direct probing of the kinetics. In this sense, the LPPS is also closely related to two-photon photoemission studies of surfaces,⁵² in which current is the measured output. The difference in this case is that the LPPS probes *molecular* dynamics, while photoemission studies probe surface dynamics of charge carriers within conductors.

The new scheme is demonstrated theoretically for a model molecular junction, in which an asymmetric bichromophoric molecule is coupled to two leads. In the absence of bias, a coherent excitation of one of the chromophores can induce charge pumping between the leads. The system response to a single pulse excitation and to a periodic monochromatic field was analyzed before.^{36,37} In this work we simulate its dc response to the LPPS excitation and demonstrate the appearance of the molecular dynamics time-scale in the correlation plot, $I_{dc}(t_d)$.

The paper is organized as follows: The theoretical formulation linking between transient observables and the steady state current in a molecular junction is introduced in section II. In section III we briefly introduce the model system of a pulse driven bichromophoric molecular electron pump. Illustrative numerical results for this model are given in section IV, and conclusions are given in section V.

II. THEORETICAL FORMULATION

Our purpose is to account simultaneously for picosecond intramolecular dynamics induced by a time-dependent excitation (in particular, a pulse pair with a controlled time delay) and for the steady state current generated by a periodic sequence of such excitations. For this purpose, we consider the generic Hamiltonian of a field driven molecule in a junction's architecture,

$$\begin{aligned}\hat{H}(t) &= \hat{H}_M + \hat{V}_{M,f}(t) + \hat{H}_{\text{leads}} + \hat{H}_{M,\text{leads}} \\ \hat{H}_{\text{leads}} &= \hat{H}_l + \hat{H}_r \\ \hat{H}_{M,\text{leads}} &= \hat{V}_{M,l}\hat{D}_l + \hat{V}_{M,r}\hat{D}_r + \text{h.c.}\end{aligned}\quad (1)$$

\hat{H}_M is the field-free molecular Hamiltonian, $\hat{V}_{M,f}(t)$ is the molecular dipole coupling to the field, and \hat{H}_{leads} is the leads

Hamiltonian, where $\{\hat{H}_l, \hat{H}_r\}$ are free electron reservoirs Hamiltonians. $\hat{H}_{M,\text{leads}}$ is the molecule–leads interaction, where $\{\hat{V}_{M,j}\}$ are electron hopping terms from the molecule to the respective leads, and $\{\hat{D}_j\}$ are the respective lead terms.⁵³

To account for the system dynamics, we invoke in this work a reduced density matrix approach^{27–29,34,36,44–50,53–56} valid to second order in the molecule–leads coupling. The Markovian equation of motion for the reduced molecular density operator reads⁵³

$$\begin{aligned}\frac{d\hat{\rho}_M(t)}{dt} &= \frac{i}{\hbar} [\hat{H}_M + \hat{V}_{M,f}(t), \hat{\rho}_M(t)] \\ &+ \sum_{j=r,l} \{ [\hat{V}_j^\dagger, \hat{H}_j(t)\hat{\rho}_M(t)] + [\hat{V}_j^\dagger, \hat{H}_j^\dagger(t)\hat{\rho}_M(t)] \\ &+ \text{h.c.} \}\end{aligned}\quad (2)$$

where

$$\begin{aligned}\hat{H}_j(t) &\equiv \frac{-1}{\hbar^2} \int_0^t dt' C_j(t-t') \hat{U}_M(t) \hat{U}_M^\dagger(t') \\ &\quad \hat{V}_j^\dagger \hat{U}_M(t') \hat{U}_M^\dagger(t) \\ \hat{H}_j^\dagger(t) &\equiv \frac{-1}{\hbar^2} \int_0^t dt' \tilde{C}_j(t-t') \hat{U}_M(t) \hat{U}_M^\dagger(t') \\ &\quad \hat{V}_j \hat{U}_M(t') \hat{U}_M^\dagger(t)\end{aligned}$$

$$C_j(t-t') = \text{tr}_j[\hat{D}_j e^{-i\hat{H}_j(t-t')/\hbar} \hat{D}_j^\dagger e^{i\hat{H}_j(t-t')/\hbar} \hat{\rho}_j]$$

$$\tilde{C}_j(t-t') = \text{tr}_j[\hat{D}_j^\dagger e^{-i\hat{H}_j(t-t')/\hbar} \hat{D}_j e^{i\hat{H}_j(t-t')/\hbar} \hat{\rho}_j]$$

$\{\hat{\rho}_j\}$ are the equilibrium density operator of the uncoupled leads, where $C_j(t-t')$ and $\tilde{C}_j(t-t')$ are lead correlation functions, and $\hat{U}_M(\tau)$ is the time evolution operator of the field-driven molecular system.

Equation 2 enables to account accurately for intramolecular coherent excitations and dynamics (the first term on the right-hand side) and to the relaxation processes induced by the leads (the second term). It is a valid approximation in the limit of weak electronic coupling between the molecular conductor and the macroscopic leads, which is the typical situation, for example, when alkyl residues are used as spacers between the molecular wire and the leads. Nevertheless, level broadening effects on the relaxation (typical to strong molecule-lead coupling³⁷) are not accounted for. Additional field-induced phenomena, such as Plasmon excitations in the leads^{57–59} and photoinduced tunneling^{60–62} are also excluded in eqs 1 and 2. The latter can be formally mapped on time-dependent molecule-leads coupling terms, which are beyond our present scope, and their effect will be included elsewhere.

In the absence of a time-dependent field, maintaining the two leads under a constant bias potential would yield a steady state current through the molecule. If, however, a transient field is switched on, charge flow into the molecular conductor from the leads would generally be time-dependent. Using eqs 1 and 2, one obtains the following equations for the charge flow into the molecule,⁵³

$$\frac{d}{dt} Q_M(t) = J_r(t) + J_l(t) \quad (3)$$

$J_j(t)$ is the transient current from the j th lead into the molecule,

$$J_j(t) = e \cdot \text{tr}_M[(\hat{L}_j \hat{\rho}_M(t)) \hat{N}_M] \quad (4)$$

\hat{N}_M is the molecular electronic number operator, and \hat{L}_j is the dissipative Liouville space operator associated with the j th lead,

$$\hat{L}_j \hat{\rho}_M(t) = \{[\hat{V}_j, \hat{F}_j(t) \hat{\rho}_M(t)] + [\hat{V}_j^\dagger, \hat{F}_j^\dagger(t) \hat{\rho}_M(t)] + \text{h.c.}\} \quad (5)$$

The transient currents can therefore be extracted from the dynamics of the reduced molecular density, $\hat{\rho}_M(t)$, as obtained by eq 2. In parallel, any intramolecular observable of interest can be calculated, as $O(t) = \text{tr}_M[\hat{O} \hat{\rho}_M(t)]$.

Our purpose is to relate the intramolecular dynamics and the respective transient currents to steady state (direct current, “dc”) measurements. Let us consider a typical experimental scenario in which the laser excitation time is 0.01–0.1 psec, and the relaxation time of charge into the leads is ~ 1 psec. The laser induced charge transfer event is therefore much shorter than the typical response time of standard current measurement devices (typically in the nsec regime). Moreover, the amount of charge transferred to the macroscopic leads following a single excitation is vanishingly small and the process can not be followed directly. However, employing a periodic sequence of excitations (the LPPS scheme) with the typical repetition period of 10 ns, the laser induced charge transport is inherently averaged and translates into a measured dc current. For example, the left-to-right current can be measured at the right lead, as the rate of charge transfer from the molecule to that lead

$$I_{\text{dc}} \equiv \frac{-1}{t_{\text{rep}}} \int_{-t_{\text{rep}}/2}^{t_{\text{rep}}/2} J_r(t) dt \quad (6)$$

The integral accumulates the charge transferred during a single laser pulse pair, and t_{rep} is the laser repetition period. Notice that, typically, the integral convergence time is 10^3 – 10^4 times shorter than the laser repetition period, which reduces the required computational effort for numerical time integration. Below we demonstrate how intramolecular time scales are reflected in the calculated dc current I_{dc} .

III. THE MODEL

As a model system, we choose the bichromophoric electron pump operating at zero bias potential, studied in refs 36 and 37. The molecular junction is based on an asymmetric donor–bridge–acceptor (DBA) molecule, in which the donor and acceptor chromophores have nearly degenerate lowest unoccupied molecular orbital (LUMO) energies but different highest occupied molecular orbital (HOMO) energies (see Figure 1). The laser carrier frequency is tuned to be in resonance with the HOMO–LUMO excitation at the donor site. In the absence of donor–acceptor coupling, any electron hopping from the donor’s LUMO to the left lead is compensated by hole hopping from that lead to the donor’s HOMO, with no net current. In the presence of donor–acceptor coupling, however, the excited electron can tunnel through the bridge into the acceptor LUMO (provided that the intramolecular tunneling period is larger than the hopping time), while similar tunneling of the hole to the acceptor is excluded by the molecular asymmetry. The result is a possibility for electron hopping into the right lead, which is not compensated by hole hopping to that lead and a net current in the left to right direction, even at zero bias potential.

This model was studied and analyzed with sudden initial excitation,³⁶ as well with periodic and finite pulse excitations using the nonperturbative nonequilibrium Green function

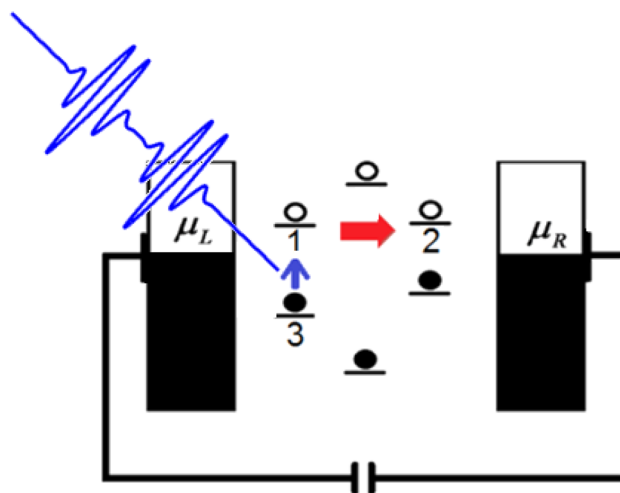


Figure 1. A schematic model for a pulse driven coherently controlled bichromophoric molecular electron pump.

formalism.³⁷ For weak molecule–electrode coupling, the results obtained by the density matrix approach were found to be in quantitative agreement with the NEGF calculations.

The model for the molecular DBA can be further simplified considering that the HOMOs of the bridge and acceptor groups remain populated at all times and need not be accounted for explicitly in the dynamics calculations. Additionally, rather than treating the bridge LUMO explicitly, we account for the through-bridge tunneling between the donor and the acceptor LUMOs in terms of an effective tunneling matrix element.⁶³ This leads to the following minimal model Hamiltonian for the three active DBA orbitals (see the numbered sites in Figure 1),

$$\hat{H}_M = \sum_{n=1}^3 E_n \hat{d}_n^\dagger \hat{d}_n + \beta \hat{d}_1^\dagger \hat{d}_2 + \beta \hat{d}_2^\dagger \hat{d}_1 \quad (7)$$

\hat{d}_n^\dagger (\hat{d}_n) is a creation (annihilation) operator of an electron at the n th local orbital. States “1” and “3” correspond, respectively, to the LUMO and HOMO of the donor chromophore, while state “2” represents the LUMO of the acceptor. β is the effective tunneling matrix element corresponding to the electronic coupling between two chromophores via the molecular bridge.

The leads Hamiltonians (see eq 1) correspond to non-interacting electron reservoirs, $\hat{H}_j = \sum_k \varepsilon_k \hat{b}_k^\dagger \hat{b}_k$, where \hat{b}_k^\dagger (\hat{b}_k) is a creation (annihilation) operator of an electron at the k th orbital of the j th lead. The molecule–leads coupling operators take the form

$$\begin{aligned} \hat{V}_{M,r} &= \hat{d}_2^\dagger & \hat{V}_{M,l} &= \hat{d}_1^\dagger + \hat{d}_3^\dagger \\ \hat{U}_r &= \sum_k u_k \hat{b}_k^\dagger & \hat{U}_l &= \sum_{kl} u_{kl} \hat{b}_k \end{aligned} \quad (8)$$

where the state-to-state coupling parameters $\{u_k\}$ reflect the electrodes spectral densities. Invoking, for example, a model of a half-filled tight binding chain⁵³ for each electrode, with a bandwidth $|4\gamma_j|$, and a chemical potential, μ_j , one has $\varepsilon_{kj} = \mu_j + 2\gamma_j \cos(k_j\pi/(N+1))$ and $u_k = \xi_j (2/(N+1))^{1/2} \sin(k_j\pi/(N+1))$, where ξ_j is the coupling (hopping) parameter between the first electrode site and the respective MO of the DBA.

The DBA is coupled to the time-dependent (semiclassical) field via the donor dipole transition

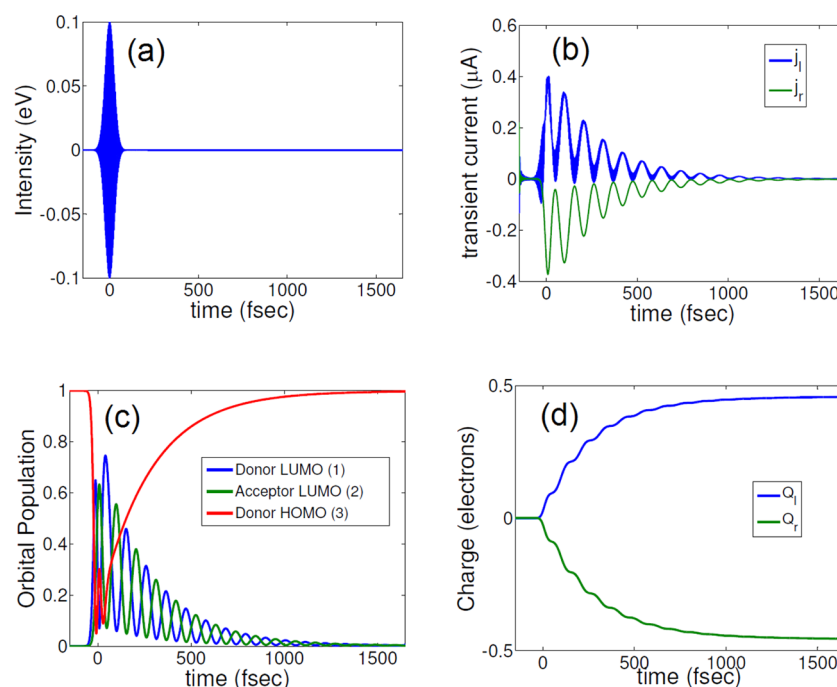


Figure 2. Transient observables associated with a single pulse excitation; (a) the pulse intensity; (b) transient currents into the molecule from the left and the right leads (negative values correspond to charge transfer out of the molecule); (c) local orbitals populations; and (d) integrated currents. The model parameters are $E_3 = -0.5$, $E_1 = E_2 = 1$, $\beta = -0.02$. The leads parameters are $\mu_L = \mu_R = 0$, $\gamma_l = \gamma_r = -2$, $\xi_l = \xi_r = -0.05$, $N = 16000$, and the pulse parameters are $A_1 = 0.1$, $A_2 = 0$, $\Omega = 1.5$, and $I_2 = 0$ (all energy values are in eV). The Gaussian pulse width parameter is $\tau = 25$ corresponding to ~ 60 fsec FWHM.

$$\hat{V}_{M,j}(t) = f(t)\hat{d}_1^\dagger\hat{d}_3 + f^*(t)\hat{d}_3^\dagger\hat{d}_1 \quad (9)$$

where $f(t)$ is the field intensity profile. For a pulse pair excitation the following profile was used

$$f(t) = A_1 \cos(\Omega t) e^{-t^2/(2\tau^2)} + A_2 \cos(\Omega[t - t_d]) e^{-(t-t_d)^2/(2\tau^2)} \quad (10)$$

The laser frequency is set to $\Omega = (E_1 - E_3)/\hbar$, τ is the pulse width parameter, A_1 and A_2 are the amplitudes of the first and second pulse, respectively, and t_d is the delay time between the pulses.

IV. ILLUSTRATIVE NUMERICAL RESULTS

The molecular junction model was studied under the influence of different laser pulses. The bias potential was set to zero in all cases, $\mu_l = \mu_r = 0$, implying that any current is due to the molecule–laser interaction. The leads densities were set to their grand canonical equilibrium states, $\hat{\rho}_j = e^{-(\hat{H}_j - \mu_j \hat{N}_j)/(k_B T)} / \text{tr}_j[e^{-(\hat{H}_j - \mu_j \hat{N}_j)/(k_B T)}]$, where $\hat{\rho}_{\text{leads}} = \hat{\rho}_r \otimes \hat{\rho}_l$. The initial molecular density was set to its approximated equilibrium state, corresponding to electronically populated donor HOMO and empty LUMOs of the donor and acceptor groups,

$$\hat{\rho}_M(0) = \hat{d}_1 \hat{d}_1^\dagger \otimes \hat{d}_2 \hat{d}_2^\dagger \otimes \hat{d}_3^\dagger \hat{d}_3 \quad (11)$$

Equation 2 was solved numerically using a sixth order Runge Kutta propagator. For the calculation of the dissipative term the time evolution operator of the molecular system was replaced by its field free version, that is, $\hat{U}_M(\tau) \approx e^{-i\hat{H}_M\tau/\hbar}$, which significantly simplifies the calculation. The induced error is insignificant as long as the decay of bath correlations is sufficiently fast on the system dynamics time scale (e.g., in the wide-band limit), which is the case for a wide range of model

parameters, including the ones used below. The system was propagated for ~ 100 fsec to reach its correct equilibrium state prior to the pulse excitation and then the pulse was turned on. Transient currents were calculated using eqs 4 and 5.

First, let us consider excitation by a single ultrafast (sub psec) pulse. In Figure 2, the results are presented for this case. The field (Figure 2a) is shown to induce bursts of current in which charge is transported from the left lead into the molecule and from the molecule to the right lead (Figure 2b). Examination of the corresponding dynamics of the intramolecular orbital populations (Figure 2c) reveals that the pulse induces a HOMO–LUMO transition at the donor, followed by intramolecular tunneling oscillations between the donor and the acceptor LUMOs. While the hole decay is restricted to the left electrode, the electron decay is oscillating between the two electrodes, resulting in net left-to-right transient currents within the relaxation time, $\hbar|\gamma|/(2\xi_j^2) \approx 0.27$ psec, set by the (weak) molecule–lead coupling. Notice that both the left and the right currents are synchronized with the motion of the electronic population toward the acceptor. Indeed, the left current is obtained when the donor LUMO population declines, and hole hopping to the left lead is not compensated by electron hopping to that lead. At the same time, the acceptor LUMO population increases and electron hopping to the right (not compensated by hole hopping to that lead) produces a right current. Time integration of the transient currents (eq 6) reveals a net transfer of <0.5 electron per single pulse (Figure 2d). Considering now a sequence of pulse excitations with a typical repetition period, $t_{\text{rep}} = 10^{-8}$ s, the resulting steady state current (per spin state) is $I_{\text{dc}}^0 < 0.5e/t_{\text{rep}} = 8$ pA. However, such an average measurement can not reveal the details of the intramolecular dynamics in this case.

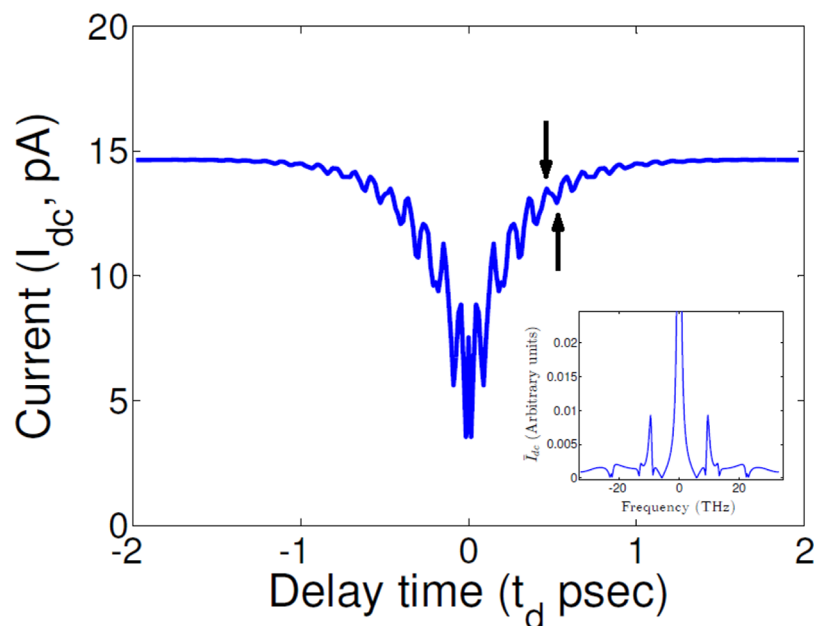


Figure 3. Direct current during a laser pulse pair sequence (LPPS) excitation, plotted vs delay time. The arrows indicate currents that are specifically discussed in Figure 4. Inset: absolute values of the Fourier transformed LPPS signal. The model parameters are the same as in Figure 2.

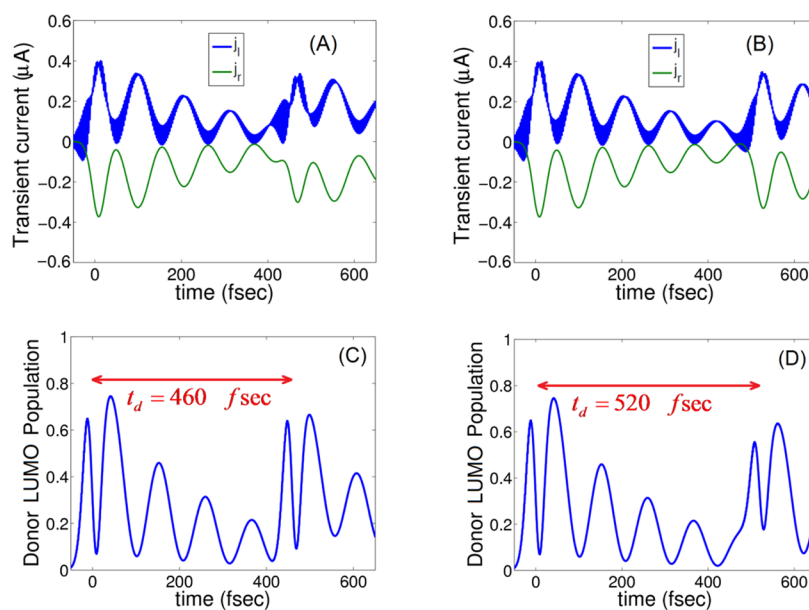


Figure 4. Transient currents (top) and donor LUMO populations (bottom) induced by two different pulse pair excitations. The corresponding steady-state currents are marked by arrows in Figure 3. (A, C) The delay time is synchronized with an integer number of intramolecular oscillation periods. (B, D) The delay time is not synchronized with the period of intramolecular oscillations. The model parameters are the same as in Figure 2.

Consider now a pair of two identical excitation pulses with a delay time, t_d , between them. If t_d exceeds by far the time of relaxation following a single excitation, the net effect of doubling the pulse would be to double the measured current, $I_{dc} = 2I_{dc}^0$. If, however, t_d is of the same order as the induced dynamics time, one expects a change in the measured current, that is, $I_{dc} \neq 2I_{dc}^0$. Moreover, one may expect the dependence of the current on the delay time to reveal the intramolecular dynamics in this case. This is demonstrated below for the present model system. The current was calculated according to eqs 4–6, propagating eq 2 for 3450 fsec, which was sufficient for convergence. I_{dc} is plotted in Figure 3 as a function of the delay time between two identical pulses ($A_1 = A_2$ in eq 10). As

one can see for $|t_d| > 1$ psec the direct current approaches an asymptotic value, which corresponds to an additive contribution of the two pulses to the current, that is, $I_{dc} = 2I_{dc}^0$. For shorter delay times the current is different, and drops significantly as the delay time approaches zero. The latter condition corresponds to a single pulse with a doubled intensity, and the drop reflects a change in the field induced Rabi frequency, which reduces the excitation efficiency within the finite pulse time.

The LPPS signal, $I_{dc}(t_d)$, is dominated by oscillations with a characteristic period ~ 0.1 psec, apparent also as two peaks at the frequencies ± 10 THz in the Fourier transformed signal (see inset). This frequency is observed also in Figure 2, and

corresponds to the electronic tunneling oscillations between the donor and acceptor LUMOs. The direct current measurements are therefore shown to be sensitive to the intramolecular dynamics and reveal the molecular frequency. A more detailed analysis shows that local maxima in the $I_{dc}(t_d)$ signal are obtained when the second pulse excitation is synchronized with the intramolecular tunneling oscillations. In particular, the efficiency of the excitation is maximized when the population of the donor LUMO is minimal (near its equilibrium value) right before the second pulse. This is demonstrated in Figure 4, where transient currents and the donor LUMO population are plotted for two different delay times. Figure 4(A,C) represent a synchronized double pulse excitation. The donor LUMO population is nearly zero prior to the second pulse operation, which implies that the HOMO–LUMO pumping by the second pulse is near its peak efficiency. This translates to an increase in the transient currents associated with the HOMO–LUMO excitation. Figure 4(B,D) represent an a-synchronized double pulse excitation, in which the donor LUMO is already excited to some extent when the second pulse operates. This leads to less efficient donor excitation and a respectively smaller current.

V. CONCLUSIONS

A new approach was introduced for measuring the dynamical response of molecular junctions to ultrafast external excitations. The approach correlates the sub picoseconds intramolecular dynamics to steady state currents, measurable with conventional electronic devices. The high time-resolution is obtained by using a sequence of fsec laser pulse pair excitations (LPPS) with a controlled delay time between the two pulses in each pair. When the delay times are short with respect to lead induced relaxation processes, and with respect to intramolecular oscillation periods, the transient currents excitation are sensitive to the time delay. This sensitivity is reflected also in the averaged direct current, which enables to relate the intimate intramolecular dynamics to macroscopic steady state measurements.

The method was demonstrated for a generic model of a bichromophoric electron pump operating at zero bias. The system was previously proposed as a coherently controlled molecular junction device in which intramolecular dynamics produces transient currents in the leads. In this work a reduced density matrix approach was used to demonstrate that not only transient currents, but also integrated steady state current can reveal the transient molecular dynamics, when the proposed LPPS scheme is applied.

While the demonstration here was limited to a generic model system and to zero bias potential, the proposed approach is expected to be valid within the realm of molecular junction devices. It is expected to provide a new direct probe into the full complexity of non equilibrium transport through these systems. The present results indeed call for more detailed simulations of LPPS signals in molecular junctions, which will account for their inherent complications, including spin and many-electron correlations, inelastic transport and vibronic coupling, strong molecule–leads coupling, level broadening and lead induced interference effects. The modeling of the laser interaction with the molecular junction need also be generalized to account for lead plasmons formation^{57–62} and to the precise nature of their coupling to the molecular excitation.^{64,65} These extensions are beyond the scope of the present work, but we believe that a future combined

experimental and theoretical effort to realize and analyze the LPPS scheme for realistic systems would promote significantly the fundamental understanding of transport in molecular junctions. Moreover, reliable experimental detection of sub psec dynamics in these systems as proposed here could pave the way to utilizing molecular devices as fast (THz) switches for future electronics.

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Notes

The authors declare no competing financial interest.

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