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Nonlinear organic plasmonics: Applications to optical control of Coulomb blocking in nanojunctions

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Purely organic materials with negative and near-zero dielectric permittivity can be easily fabricated. Here, we develop a theory of nonlinear non-steady-state organic plasmonics with strong laser pulses that enable us to obtain near-zero dielectric permittivity during a short time. Our consideration is based on the model of the interaction of strong (phase modulated) laser pulse with organic molecules developed by one of the authors before, extended to the dipole-dipole intermolecular interactions in the condensed matter. We have proposed to use non-steady-state organic plasmonics for the enhancement of intersite dipolar energy-transfer interaction in the quantum dot wire that influences on electron transport through nanojunctions. Such interactions can compensate Coulomb repulsions for particular conditions. We propose the exciton control of Coulomb blocking in the quantum dot wire based on the non-steady-state near-zero dielectric permittivity of the organic host medium. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4928181]

Plasmonics and metamaterials provide great scope for concentrating and manipulating the electromagnetic field on the subwavelength scale to achieve dramatic enhancement of optical processes and to develop super-resolution imaging, optical cloaking, etc.[1–4] However, metallic inclusions in metamaterials are sources of strong absorption loss. This hinders many applications of metamaterials and plasmonics and motivates to search for efficient solutions to the loss problem.[5] Highly doped semiconductors[5,6] and doped graphene[7–9] can in principle solve the loss problem. However, the plasmonic frequency in these materials is an order of magnitude lower than that in metals making the former most useful at mid-IR and THz regions. In this relation, the question arises whether metal-free metamaterials and plasmonic systems, which do not suffer from excessive damping loss, can be realized in the visible range? With no doubts, inexpensive materials with such advanced properties can impact whole technological fields of nanoplasmonics and metamaterials.

Recently, Noginov *et al.* showed that purely organic materials characterized by low losses with negative, near-zero, and smaller than unity dielectric permittivities can be easily fabricated, and propagation of a surface plasmon polariton at the material/air interface was demonstrated.[10] And even non-steady-state organic plasmonics with strong laser pulses may be realized[11] that can enable us to obtain near-zero dielectric permittivity during a short time only. Approach[10] was explained in simple terms of the Lorentz model for linear spectra of dielectric permittivities of thin film dyes. However, the experiments with strong laser pulses[11] challenge theory.

Here, we develop a theory of nonlinear non-steady-state organic plasmonics with strong laser pulse excitation. Our consideration is based on the model of the interaction of strong (phase modulated) laser pulse with organic molecules,[12] extended to the dipole-dipole intermolecular interactions in the condensed matter. We propose the exciton compensation of Coulomb blocking (ECCB)[13] in the quantum dot wire based on the non-steady-state near-zero dielectric permittivity of the organic host medium using chirped laser pulses.

Let us extend our picture of "moving" potentials of Ref. 12 to a condensed matter. We consider a molecule with two electronic states $n = 1$ (ground) and 2 (excited) in a solvent described by the Hamiltonian $H_0 = \sum_{n=1}^{2} |n\rangle[E_n + W_n(\mathbf{Q})]\langle n|$, where $E_2 > E_1, E_n$ is the energy of state n, and $W_n(\mathbf{Q})$ is the adiabatic Hamiltonian of reservoir R (the vibrational subsystems of a molecule and a solvent interacting with the two-level electron system under consideration in state n). The molecule is affected by a (phase modulated) pulse $\mathbf{E}(t) = (1/2)\mathbf{e}\mathcal{E}(t)\exp(-i\omega t + i\varphi(t)) + \text{c.c.}$, the frequency of which is close to that of the transition $1 \to 2$. Here, $\mathcal{E}(t)$ and $\varphi(t)$ describe the change of the pulse amplitude and phase in time, $\mathbf{e}$ is unit polarization vectors, and the instantaneous pulse frequency is $\omega(t) = \omega - d\varphi(t)/dt$.

One can describe the influence of the vibrational subsystems of a molecule and a solvent on the electronic transition within the range of definite vibronic transition related to a high frequency optically active (OA) vibration as a modulation of this transition by low frequency (LF) OA vibrations $\{\omega_s\}$.[14] In accordance with the Franck-Condon principle, an optical electronic transition takes place at a fixed nuclear configuration. Therefore, the quantity $u_1(\mathbf{Q}) = W_2(\mathbf{Q}) - W_1(\mathbf{Q}) - \langle W_2(\mathbf{Q}) - W_1(\mathbf{Q})\rangle_1$ representing electron-vibration coupling is the disturbance of nuclear motion under electronic transition, where $\langle\rangle_n$ stands for the trace operation over the reservoir variables in the electronic state n. Electronic transition relaxation stimulated by LFOA vibrations is described by the correlation function $K(t) = \langle\alpha(0)\alpha(t)\rangle$ of the corresponding vibrational disturbance with characteristic attenuation time τ_s,[15–17] where $\alpha \equiv -u_1/\hbar$. The analytic solution of the problem under consideration has been obtained due to the presence of a small parameter. For broad vibronic spectra satisfying the "slow modulation" limit, we have $\sigma_{2s}\tau_s^2 \gg 1$ where $\sigma_{2s} = K(0)$ is the LFOA vibration contribution to a second central moment of an absorption spectrum, the half bandwidth of which is related to σ_{2s} as $\Delta\omega_{abs} = 2\sqrt{2\sigma_{2s}\ln 2}$. According to Refs. 16 and 17, the following times are characteristic for the time evolution of the system under consideration: $\sigma_{2s}^{-1/2} < T' \ll \tau_s$, where $\sigma_{2s}^{-1/2}$ and $T' = (\tau_s/\sigma_{2s})^{1/3}$ are the times of reversible and irreversible dephasing of the electronic transition, respectively. The characteristic frequency range of changing the optical transition probability can be evaluated as the inverse T', i.e., $(T')^{-1}$. Thus, one can consider T' as a time of the optical electronic transition. Therefore, the inequality $\tau_s \gg T'$ implies that the optical transition is instantaneous where relation $T'/\tau_s \ll 1$ plays the role of a small parameter. This made it possible to describe vibrationally nonequilibrium populations in electronic states 1 and 2 by balance equations for the intense pulse excitation (pulse duration $t_p > T'$) and solve the problem.[12,18]

Equations of Ref. 12 describing vibrationally nonequilibrium populations in electronic states $j = 1$, 2 for the exponential correlation function $K(t)/K(0) \equiv S(t) = \exp(-|t|/\tau_s)$ and the intense chirped pulse excitation, extended to the dipole-dipole intermolecular interactions in the condensed matter, take the following form:[26]

$$\frac{\partial \rho_{jj}(\alpha,t)}{\partial t} = \frac{(-1)^j \pi}{2\hbar^2}\delta[\omega_{21} - p\Delta n - \omega(t) - \alpha]\eta^2 \times |\mathbf{D}_{21}\vec{\mathcal{E}}(t)|^2 \Delta'(\alpha,t) + L_{jj}\rho_{jj}(\alpha,t), \tag{1}$$

where $\eta \equiv (\varepsilon_b + 2)/3$, $\Delta'(\alpha,t) = \rho_{11}(\alpha,t) - \rho_{22}(\alpha,t)$, $\Delta n = n_1 - n_2$, $\mathbf{D}_{21}$ is the electronic matrix element of the dipole moment operator. Here, ρ_{jj} are the diagonal elements of the density matrix, ω_{21} is the frequency of Franck-Condon transition $1 \to 2$, operator $L_{jj} = \tau_s^{-1}\left[1 + \left(\alpha - \delta_{j2}\omega_{st}\right)\frac{\partial}{\partial\left(\alpha - \delta_{j2}\omega_{st}\right)} + \sigma_{2s}\frac{\partial^2}{\partial\left(\alpha - \delta_{j2}\omega_{st}\right)^2}\right]$ describes the diffusion with respect to the coordinate α in the corresponding effective parabolic potential, $U_j(\alpha)$, δ_{ij} is the Kronecker delta, and $\omega_{st} = \hbar\sigma_{2s}/(k_B T)$ is the Stokes shift of the equilibrium absorption and luminescence spectra. The partial density matrix of the system $\rho_{jj}(\alpha,t)$ describes the system distribution in states 1 and 2 with a given value of α at time t. The complete density matrix averaged over the stochastic process, which modulates the system energy levels, is obtained by integration of $\rho_{jj}(\alpha,t)$ over α, $\langle\rho\rangle_{jj}(t) = \int \rho_{jj}(\alpha,t)d\alpha$, where quantities $\langle\rho\rangle_{jj}(t)$ are the normalized populations of the corresponding electronic states: $\langle\rho\rangle_{jj}(t) \equiv n_j$, $n_1 + n_2 = 1$. Here, ε_b is the "bulk" relative permittivity (which can be due distant high-frequency resonances of the same absorbing molecules or a host medium), $p = \frac{4\pi}{3\hbar}|D_{12}|^2 N$ is the strength of the near dipole-dipole interaction,[19] N is the density of molecules.

Knowing $\rho_{jj}(\alpha,t)$, one can calculate the susceptibility $\chi(\Omega,t)$[12] that enables us to obtain the dielectric function ε due to relation $\varepsilon(\Omega,t) = 1 + 4\pi\chi(\Omega,t)$

$$\varepsilon(\Omega,t) = 1 + ip\eta\left\{\sqrt{\frac{\pi}{2\sigma_{2s}}}w\left[\frac{\Omega - (\omega_{21} - p\Delta n(t))}{\sqrt{2\sigma_{2s}}}\right] - \pi\sigma_a\eta^2\int_0^t dt'\Delta'\left(\omega_{21} - p\Delta n(t') - \omega(t'),t'\right) \times \tilde{J}(t')\sum_{j=1}^{2}\sqrt{\frac{\sigma_{2s}}{\sigma(t-t')}}w\left[\frac{\Omega + p\Delta n(t) - \omega_j(t,t')}{\sqrt{2\sigma(t-t')}}\right]\right\}, \tag{2}$$

where $\tilde{J}(t)$ is the power density of the exciting radiation, σ_a is the cross section at the maximum of the absorption band, $\sigma(t-t') = \sigma_{2s}[1 - S^2(t-t')]$, $\omega_j(t,t') = \omega_{21} - \delta_{j2}\omega_{st} + [\omega(t') - \omega_{21} + p\Delta n(t') + \delta_{j2}\omega_{st}]S(t-t')$ are the first moments of the transient absorption ($j = 1$) and the emission ($j = 2$) spectra, and $w(z) = \exp(-z^2)[1 + i\frac{2}{\sqrt{\pi}}\int_0^z \exp(y^2)dy]$ is the probability integral of a complex argument.[20] It is worthy to note that magnitude $\varepsilon(\Omega,t)$ does make sense, since it changes in time slowly with respect to dephasing. In other words, $\varepsilon(\Omega,t)$ changes in time slowly with respect to the reciprocal characteristic frequency domain of changing $\varepsilon(\Omega)$.

Let us consider the particular case of fast vibrational relaxation when one can put the correlation function $S(t-t')$ equal to zero. Physically, it means that the equilibrium distributions into the electronic states have had time to be set during changing the pulse parameters. Using Eq. (1), one obtains the equations for the populations of electronic states $n_{1,2}$ in the case under consideration, which represents extending Eq. (25) of Ref. 12 to the interacting medium

$$\frac{dn_{1,2}}{dt} = \pm\sigma_a\eta^2\exp\left\{-\frac{[\omega_{21} - p\Delta n - \omega(t) - \omega_{st}]^2}{2\sigma_{2s}}\right\} \times\tilde{J}\left\{n_2 - n_1\exp\left[\frac{\left(\omega(t) + p\Delta n - \omega_{21} + \frac{\omega_{st}}{2}\right)}{k_B T/\hbar}\right]\right\} \pm \frac{n_2}{T_1}, \tag{3}$$

where we added term "$\pm n_2/T_1$" taking lifetime T_1 of the excited state into account. In case of fast vibrational relaxation, Eq. (2) becomes

$$\varepsilon(\Omega,t) = 1 + ip\eta\sqrt{\frac{\pi}{2\sigma_{2s}}}\sum_{j=1,2}(-1)^j \times n_j(t)w\left[\frac{\Omega - \omega_{21} + p\Delta n(t) + \delta_{2j}\omega_{st}}{\sqrt{2\sigma_{2s}}}\right]. \tag{4}$$

Eqs. (1) and (3) for populations are nonlinear equations where the transition frequencies are functions of the electronic states populations. However, one can use pulses that are suitably chirped to compensate for a change of frequency of the optical transition in time induced by the pulses themselves. This idea was proposed in studies of a two-state system in relation to Rabi oscillations in inter-subband transitions in quantum wells[21] and for obtaining efficient stimulated Raman adiabatic passage in molecules in a dense medium.[19] Let us suppose that we use suitably chirped pulses compensating the

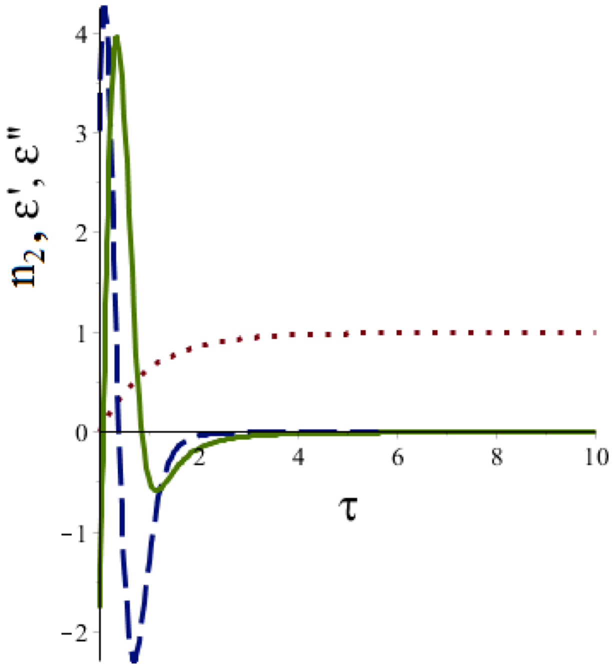

FIG. 1. Population of the excited state (dots), and real (solid line), and imaginary (dashed line) parts of the dielectric function as functions of time for $WT_1 = 1000$. For a typical value of the lifetime of the excited state $T_1 = 10^{-9}$ s, this gives $W = 10^{12}$ s^{-1}. Corresponding times $t = \tau/W = \tau \times 10^{-12}$ s lie in the picosecond time range.

"local field" detuning $p\Delta n$ that enables us to keep the value of $\omega_{21} - p\Delta n(t) - \omega(t) \equiv \Delta\omega$ as a constant. In that case, one can obtain an integral equation for non-equilibrium population difference $\Delta'(\Delta\omega, t)$.[26] We also use Eqs. (3) and (4) in the case under consideration to demonstrate obtaining near-zero dielectric function in non-steady-state regime. Consider a dense collection of molecules ($N \sim 10^{21}$ cm^{-3} (Ref. 10)) with parameters close to those of molecule LD690:[12] $\sqrt{\sigma_{2s}} = 546\,\text{cm}^{-1}$, $D_{12} \sim 10^{-17}$ CGSE that gives $\omega_{st} = 1420\,\text{cm}^{-1}$, $p = 2107.2\,\text{cm}^{-1}$. We put $\varepsilon_b = 1$ (Ref. 10) and $\Delta\omega = -420\,\text{cm}^{-1}$. Fig. 1 shows the population of excited electronic state n_2 and the real $\varepsilon'(\Omega, t)$ and imaginary $\varepsilon''(\Omega, t)$ parts of $\varepsilon(\Omega, t)$ for $\Omega - \omega_{21} = -2.0405\sqrt{2\sigma_{2s}}$, during the action of a rectangular light pulse of power density $\tilde{J}$ that begins at $t = 0$. Here, we denoted the probability of the optical transitions induced by external field as $W = \sigma_a \eta^2 \exp[-\Delta\omega^2/(2\sigma_{2s}))]\tilde{J}$, and $\tau = Wt$—dimensionless time. Fig. 1 illustrates non-steady-state near-zero dielectric permittivity. As population n_2 approaches to 1, dielectric permittivity approaches to zero. This may be applied to ECCB in nanojunctions. It is worthy to note that $W = 10^{12}$ s^{-1} (see the figure caption of Fig. 1) is achieved at power densities of the exciting radiation, $\tilde{J} \sim 10^{28} \frac{phot}{\text{cm}^2\text{s}}$, for the absorption cross section $\sigma_a \sim 10^{-16}\text{cm}^2$ that is typical for dyes.

In Ref. 13, we studied the influence of both exciton effects and Coulomb repulsion on current in nanojunctions. We showed that dipolar energy-transfer interactions between the sites in the wire can at high voltage compensate Coulomb blocking for particular relationships between their values. Although in free exciton systems dipolar interactions J ($\lesssim 0.01 - 0.1$eV (Ref. 22)) are considerably smaller than on-site Coulomb interaction U (characteristically $U \sim 1$ eV (Ref. 23)) the former may still have strong effects under some circumstances, e.g., in the vicinity of metallic structures in or near the nanojunctions. In such cases, dipolar interactions may be enhanced. The enhancement of the dipole-dipole interaction for the dimer of silver spheres and for a single sphere reached the value of 0.13 eV for nanosphere-shaped metallic contacts,[13] which was smaller than U. In addition, this enhancement was accompanied by metal induced damping of excitation energy. Here, we show that purely organic materials characterized by low losses with near-zero dielectric permittivities will enable us to obtain $\hbar J \sim 1$ eV $\sim U$. Let us consider a nanojunction consisting of a two site quantum dot wire (each represented by its ground, $|g\rangle$, and excited, $|e\rangle$, states) positioned between two leads with applied voltage bias $eV_{bs} = \mu_L - \mu_R$, Fig. 2. The junction is found in an organic material with dielectric permittivity ε. The quantum dots of the wire posses transition dipole moments $\mathbf{D}_1$ and $\mathbf{D}_2$. The point dipoles are positioned at points $\mathbf{r}_1$ and $\mathbf{r}_2$, respectively, and oscillate with frequency Ω. The interaction energy between the dipoles can be written as $J_{12} + J_{21}$ where $J_{ij} = -(1/2)\mathbf{D}_i \cdot \mathbf{E}_j(\mathbf{r}_i, \Omega, t) \quad (i, j = 1, 2; \quad i \neq j)$, $\mathbf{E}_j(\mathbf{r}_i, \Omega, t)$ is the electric field at a point $\mathbf{r}_i$ induced by the dipole $\mathbf{D}_j$ and is given by Coulomb's law $\mathbf{E}(\mathbf{r}, \Omega, t) = \varepsilon^{-1}(\Omega, t) \int \rho_j(\mathbf{r}')(\mathbf{r} - \mathbf{r}')|\mathbf{r} - \mathbf{r}'|^{-3} d^3\mathbf{r}'$ corresponding to the electrostatic approximation. Such extension of the electrostatic formula is possible due slow changes in time of $\varepsilon(\Omega, t)$ (see above). Here, $\rho_j(\mathbf{r}')$ is the external charge transition density created by dipole $\mathbf{D}_j$. One can show that $J_{12} = J_{21} \equiv \frac{\hbar}{2} J(\Omega, t)$.[24] If the dipoles are oriented parallel to the symmetry axis of the junction, $J(\Omega, t)$ is given by $J(\Omega, t) = J_{vac}/\varepsilon(\Omega, t)$, where $J_{vac} = -2\hbar^{-1} D_1 D_2 |\mathbf{r}_1 - \mathbf{r}_2|^{-3}$ is the dipole-dipole interaction in vacuum. The bottom of Fig. 4 shows J as a function of time for a medium with dielectric function given by Fig. 1. Putting $D_1 = D_2 = 25D$ and $|\mathbf{r}_1 - \mathbf{r}_2| = 5\,\text{nm}$, one gets $\hbar|J_{vac}| = 0.00625\,\text{eV}$, and the value of $\hbar|J(\Omega, t)| = 1.6602\,\text{eV}$ for $\tau = 10$.

Let us calculate current through the two site quantum dot nanojunction described above using approach of Ref. 13. The corresponding Hamiltonian is $H = H_{wire} + H_{leads} + H_{int}$ where $H_{leads} = \sum_{k \in \{L,R\}} E_k c_k^\dagger c_k$

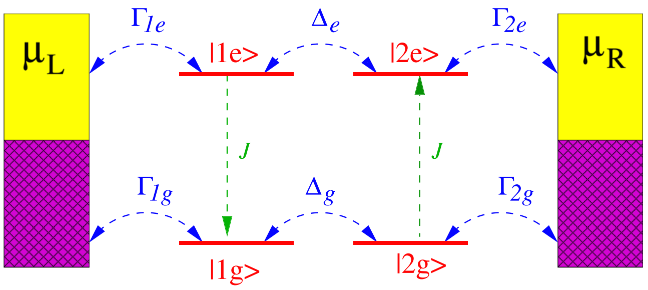

FIG. 2. A model for energy-transfer induced effects in conduction nanojunction consisting of a two site quantum dot wire between two metal leads with applied voltage bias. Each site is represented by its ground, $|1g\rangle$ and $|2g\rangle$, and excited, $|1e\rangle$ and $|2e\rangle$, levels with the nearest neighbor site coupling Δ_g and Δ_e. The two metal leads characterized by electrochemical potentials μ_R and μ_L are coupled with their nearest site by H_{int}, Eq. (6), with the transfer rates Γ_{2e}, Γ_{2g}, Γ_{1e}, and Γ_{1g} that are calculated using Eq. (6) of Ref. 13. J indicates exciton hopping.

$$H_{wire} = \sum_{\substack{m=1,2\\ f=g,e}} E_{mf} c^{\dagger}_{mf} c_{mf} - \sum_{f=g,e} \Delta_f \left(c^{\dagger}_{2f} c_{1f} + c^{\dagger}_{1f} c_{2f} \right)$$

$$+ \hbar \left(J b^{\dagger}_1 b_2 + H.c. \right) + \frac{U}{2} \sum_{m=1,2} N_m (N_m - 1), \qquad (5)$$

$$H_{int} = \sum_{mf,k \in K_m} V^{(mf)}_k c^{\dagger}_k c_{mf} + H.c., \qquad (6)$$

where $c^{\dagger}_{mf}$ (c_{mf}) ($m = 1, 2, f = g, e$, see Fig. 2) are creation (annihilation) operators for electrons in the different site states of energies E_{mf}, while $c^{\dagger}_k$ (c_k) ($k \in L, R$) are creation (annihilation) operators for free electrons (energies E_k) in the leads L and R. $n_{mf} = c^{\dagger}_{mf} c_{mf}$ are the occupation operators for the different site states, and site occupation operators are given by $N_m = n_{mg} + n_{me}$. The operators $b^{\dagger}_m = c^{\dagger}_{me} c_{mg}$ and $b_m = c^{\dagger}_{mg} c_{me}$ are exciton creation and annihilation operators on the molecular sites $m = 1$ and 2. In Eq. (5), Δ_f represents electron tunneling between site states of similar energies (i.e., between $|g\rangle$ levels of sites 1 and 2 and between $|e\rangle$ levels on these sites), the J terms represent exciton hopping (energy transfer) between molecular sites and the U terms correspond to on-site Coulomb interactions. The molecule-leads interaction H_{int} describes electron transfer between the molecular bridge and the leads that give rise to net current in the biased junction. In (6), K_m is the lead closer the molecular site m ($K_1 = L$, $K_2 = R$) and $H.c$. denotes Hermitian conjugate. Since in the medium with near-zero dielectric permittivities, both exciton-exciton interaction J and on-site Coulomb interaction U can achieve the value of about 1 eV (see above), we account and add the additional two off-resonance terms to H_{wire}, Eq. (5),

$$H_{off-ex-ex} = \hbar J(\Omega, t) b^{\dagger}_1 b^{\dagger}_2 + H.c.\,, \qquad (7)$$

$$H_{off-el-el} = - \sum_{f,f'=g,e;f \neq f'} \Delta_{ff'} (\hat{c}^{\dagger}_{2f} \hat{c}_{1f'} + \hat{c}^{\dagger}_{1f'} \hat{c}_{2f}). \qquad (8)$$

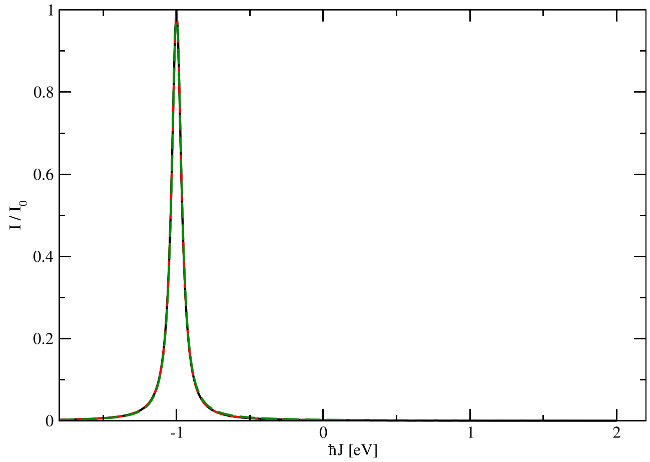

FIG. 3. Current I displayed as function of the energy-transfer coupling J for $U = 1$ eV and $\Delta_f = \Delta_{ff'} \equiv \Delta = 0.01$ eV. The current shows a maximum at $U = -\hbar J$. Solid black line—calculations using Eq. (5) for the wire Hamiltonian; red dashed line—calculations using Eqs. (5) and (7) for the wire Hamiltonian; green dashed line—calculations using Eqs. (5), (7), and (8) for the wire Hamiltonian. Comparison of these lines displays small effects of non-resonance contributions.

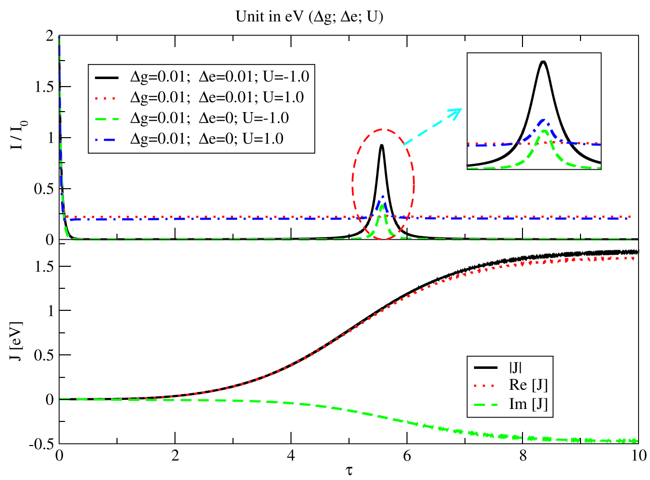

FIG. 4. Laser induced current I (top) and the dipole-dipole interaction in an organic material J (bottom) displayed as functions of τ. Other parameters are identical to those of Fig. 1. Times $t = \tau / W = \tau \times 10^{-12}$ s lie in the picosecond time range.

Eq. (8) takes into consideration the possibility of the charge transfer between $|g\rangle$ state of one site and $|e\rangle$ state of another that may be in resonance due to the reconstruction of the wire energetic spectrum by large J and U. Eq. (7) is so called non-Heitler-London term[25] taking into account the creation and annihilation for excitation simultaneously at two sites (quantum dots) when $\hbar J \sim E_{me} - E_{mg}$. In this relation the following question arises: "Does the effect of ECCB survive for such large values of $\hbar J \sim 1$ eV $\sim U$?" Fig. 3 shows that the ECCB does survive for large values of $J \sim 1$ eV. We put the bias voltage $V_{bs} = 8$ eV and the rate of charge transfer from a quantum dot to the corresponding lead $\Gamma = 0.02$ eV in our simulations, and denoted the unit of current as $I_0 = e\Gamma/\hbar$ (e is the charge of one electron). Fig. 4 shows current through the nanojunction during the action of the rectangular laser pulse with parameters given above on the host organic material. One can see dramatic increase in the current when $\hbar J$ approaches to $-U$ for $\Delta_g = \Delta_e$, and to $\pm U$ for $\Delta_g \neq 0$ and $\Delta_e = 0$. After this moment, the current decreases in spite of increasing J, since its value exceeds that of U. So, current exists during the time that is much shorter than the pulse duration. As a matter of fact, Fig. 4 illustrates optical switches based on the effect of ECCB-ECCB switches.

Furthermore, if one does not use suitably chirped pulses (see above), Eqs. (1) and (3) for populations become nonlinear equations and can demonstrate a bistable behavior that will be considered elsewhere.

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[26] See supplementary material at http://dx.doi.org/10.1063/1.4928181 for derivation of Eq. (1) and for integral equation for non-equilibrium population difference $\Delta'(\Delta\omega, t)$.