Rabi oscillations and photocurrent in quantum-dot tunnelling junctions

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Motivated by the experiments by Zrenner et al. [Nature 418, 612 (2002)], we study the influence of relaxation processes on converting Rabi oscillations in a strongly biased single-quantum-dot photodiode into deterministic photocurrents. We show that the behavior of a quantum dot with different tunnel rates for electron and holes is qualitatively different from that with the equal tunnel rates: in the latter case the current shows attenuating oscillations with the Rabi frequency \( \Omega \). In contrast, for different electrons and holes tunnelling rates, the frequency of these oscillations diminishes, and they disappear beyond a definite asymmetry threshold. We give an analytical solution of the problem and a numerical example showing a different behaviour of the transferred charge in the small attenuation limit for equal and different tunnel rates for electrons and holes.

1 Introduction

Quantum-dot (QD) and molecular conduction nanojunctions have been under intense study for some time [1-3]. The possible characterization and control of such systems using light has been recently discussed [4-7]. In an earlier work we have developed a theory for light-induced current by strong optical pulses in tunnelling nanojunctions [7]. We considered a molecular bridge represented by its highest occupied and lowest unoccupied states. We took into account two types of couplings between the bridge and the metal leads: electron transfer coupling that gives rise to net current in the biased junction and energy transfer interaction between excitations on the bridge and electron-hole formation in the leads.

We have proposed an optical control method based on the adiabatic rapid passage for enhancing charge transfer in unbiased junctions where the bridging molecule is characterized by a strong charge-transfer transition. The method is robust, being insensitive to pulse area and the precise location of resonance that makes it suitable for a molecular bridge even for inhomogeneously broadened optical transition. In the absence of inhomogeneous broadening another procedure based on the \( \pi \)-pulse excitation can be applied. Recently Zrenner et al. [1] have demonstrated that Rabi oscillations between two excitonic energy levels of an InGaAs QD placed in a photodiode can be converted into deterministic photocurrents [Fig. 1]. This device can function as an optically triggered single-electron turnstile. However, its efficiency for converting Rabi flopping into photocurrent is reduced at low bias voltage [10] when dephasing in the main is related to electron-phonon interaction [8,9]. In this mechanism, any \( \pi \)-pulse can induce a passage of one elementary charge \( e \) provided that the subsequent tunneling process occurs with probability 1. Strong enough bias is needed to overcome the electron-hole attraction. At the same time the yield of photocurrent generation will be re-
duced by competing processes such as radiative and non-
radiative recombination of electrons and holes.

In the experiment of Ref. [1] on strongly biased single-
QD photodiode the tunnelling time $\tau_{\text{tunneled}}$ (< 10 ps) is much
shorter than low-temperature dephasing times in self-
assembled InGaAs QDs ($\tau_{\text{dephase}}$ > 500 ps). This makes
photocurrent generation by Rabi oscillations relatively ef-

cient. Below we study the influence of relaxation proc-

cesses on this process at rather large bias voltages, where
electron/ hole tunnelling to the leads is the main relaxation

mechanism.

2 Equations of motion for the pseudospin vector and their solution
We consider a bridge characterized by two single electron orbitals $|1\rangle$ and $|2\rangle$ that are posi-
tioned below and above the equilibrium Fermi-level, re-
spectively. The bridge interacts with the external radiation
field $(1/2)E(t)\exp(-i\omega t)$ and characterized by the pulse
envelope $E(t)$ and carrier frequency $\omega$. Bearing in mind
the situation shown in Fig.1, Eqs. (41)-(43) and (45) of Ref.
[7] can be written as

$$
dn_m/dt = -(1)^n \text{Im}(\Gamma^\dagger(t)\vec{p}_M) + \Gamma_{Mn}(\delta_{Im} - n_m) \quad (1)
$$

$$
\frac{d\vec{p}_M}{dt} = -i\Delta \vec{p}_M + \frac{i}{2}(\Gamma_M^+ + \Gamma_M^-) \vec{p}_M
$$

$$
I_L = e\pi\Gamma_M \quad (3a)
$$

$$
I_R = e(1-n)\Gamma_M \quad (3b)
$$

where $n_\mu$ is the electron population in state $m$ ($m=1,2$),
$\vec{p}_M$ is the slowly varying amplitude of the QD polariza-
tion,

$$
\Gamma_{Mn} = \frac{2\pi}{\hbar} \sum_l \left| V^{(N)}_{lm} \right|^2 \delta(\varepsilon_l - \varepsilon_m) \quad (4)
$$

is the corresponding tunnelling rate for the bias induced
asymmetry shown in Fig.1 (we assume that hole tunnelling
with rate $\Gamma_M$ takes place to the right electrode
and electron tunnelling with rate $\Gamma_M$ is to the left elec-
trode), $\varepsilon_l$ is the energy of state $l$ and states $|k\rangle$ are single
electron states of the reservoir (i-GaAs). $\Omega(t)=dE(t)/\hbar$ is
the Rabi frequency, $d$ is the transition dipole moment char-
acterizing the optical $|1\rangle \leftrightarrow |2\rangle$ transition,
$\Delta\equiv(\varepsilon_2 - \varepsilon_1)/\hbar - \omega$ is the detuning of the pulse frequency
from the bridge transition frequency, $I_L$ and $I_R$ are respec-
tively the electronic current due to the coupling of state $|2\rangle$
with the left electrode and the hole current due to the cou-
pling of state $|1\rangle$ with the right electrode. Finally, $\delta_{Im}$ is
the Kronecker delta. In writing Eqs.(1)-(2) we have disre-
garded energy transfer terms with the rate parameter

$$
B_\nu(\varepsilon_j - \varepsilon_k, \mu) = \frac{2\pi}{\hbar} \sum_l \left| V^{(N)}_{lm} \right|^2 \delta(\varepsilon_l - \varepsilon_k, + \varepsilon_j - \varepsilon_k) \times f(\varepsilon_l) [1 - f(\varepsilon_k)] \quad (5)
$$

which is associated with electron-hole excitations in the
electrode. (Here $f(\varepsilon_k) = 1/[\exp((\varepsilon_k - \mu)/k_B T) + 1]$ is the
Fermi function and $\mu$ is the chemical potential). In doing
so we have assumed that $\varepsilon_2 - \varepsilon_1$ is smaller than the band-
gap of (intrinsic) i-GaAs (electrodes) as depicted in Fig.1.
(Note that at sufficiently large bias, energy transfer can
take place with the electron and hole created on opposite
sides of the QD bridge, however, we expect that for such
states the matrix elements $V^{(N)}_{lm}$ are small, and disregard
this possibility as well.)

For the following analysis it is convenient to write
down Eqs. (1)-(3) in terms of the Bloch vector components
[11,12] $r_1 = 2\text{Re} \vec{p}_M$, $r_2 = -2\text{Im} \vec{p}_M$ and $n_\mu = n_\mu - n_\mu$, and the
variable $\lambda = n_1 + n_2$. This basis has the advantage of reveal-
ing the symmetry properties of the Lie group SU(2). The
following analysis is made for a square pulse of duration $t_\text{p}$ and height $E_0$ starting at $t=0$. Using the unitary
transformation

$$
\begin{pmatrix}
  R_1 \\
  R_2 \\
  R_3
\end{pmatrix} =
\begin{pmatrix}
  \cos 2\theta & 0 & -\sin 2\theta \\
  0 & 1 & 0 \\
  \sin 2\theta & 0 & \cos 2\theta
\end{pmatrix}
\begin{pmatrix}
  r_1 \\
  r_2 \\
  r_3
\end{pmatrix}
$$

where $\cos 2\theta = \frac{\Delta}{\sqrt{\Delta^2 + \Omega^2}}$, $\sin 2\theta = \frac{-\Omega}{\sqrt{\Delta^2 + \Omega^2}}$, we get
the Bloch equations in the basis of dressed states. They
have exact solution given by the roots of a quaternary
equation corresponding to the system of differential equa-
tions (1)-(2). An interesting case is when the pulse is in
resonance with the QD transition energy ($\Delta=0$). In this
situation we obtain

$$
\frac{dR_1}{dt} = -[\Omega R_1 - \frac{1}{2}(\Gamma_{M2} - \Gamma_{M1})] R_1 - \frac{1}{2}(\Gamma_{M2} + \Gamma_{M1}) R_2
$$

$$
\frac{dR_2}{dt} = [\Omega R_1 - \frac{1}{2}(\Gamma_{M2} + \Gamma_{M1})] R_2
$$

$$
\frac{dR_3}{dt} = \Gamma_{M1} - \frac{1}{2}(\Gamma_{M2} - \Gamma_{M1}) \frac{\Omega}{|\Omega|^2} R_1 - \frac{1}{2}(\Gamma_{M2} + \Gamma_{M1}) R_3
$$

$$
I_L = e\frac{e}{2}(\Gamma_{M2} + \Omega) R_1
$$

where $\lambda$ is invariant under unitary transformation (6). If we
transform the Bloch vector components $R_1, R_2, R_3$ to new magnitudes $\tilde{R}_1, \tilde{R}_2, \tilde{R}_3$ defined by

$$
(R_1, R_2, R_3) = (\tilde{R}_1, \tilde{R}_2, \tilde{R}_3) \exp[-\frac{1}{2}(\Gamma_{M2} + \Gamma_{M1}) t],
$$

Eqs.(7)-(9) are reduced to the second order differential
equation
\[
\frac{d^2 \tilde{r}_1}{dt^2} + i \Omega^2 - \frac{1}{4} (\Gamma M_2 - \Gamma M_1)^2 \tilde{r}_1 = -\frac{\Omega}{|\Omega|} \Gamma M_1 \Gamma M_2 \exp\left[\frac{1}{2} (\Gamma M_2 + \Gamma M_1) \nu \right] \]
\]
(12)

The particular solutions of the homogeneous equation corresponding to Eq.(12) are \(\exp(\pm i t \alpha)\) where
\[
\alpha = \frac{1}{2} \sqrt{(\Gamma M_2 - \Gamma M_1)^2 - 4 \Omega^2} = \begin{cases} \pm \nu, & 4 \Omega^2 > (\Gamma M_2 - \Gamma M_1)^2 \\ 0, & 4 \Omega^2 = (\Gamma M_2 - \Gamma M_1)^2 \end{cases} \quad (13)
\]
which defines the positive numbers \(\nu\) and \(\gamma\). We see that the behavior of a QD with \(\Gamma M_1 \neq \Gamma M_2\) is qualitatively different from that of a QD with \(\Gamma M_1 = \Gamma M_2\) (including the case \(\Gamma M_1 = \Gamma M_2 = 0\) which is realized in the absence of charge transfer). When \(\Gamma M_1 = \Gamma M_2\), \(R_1\) and the current \(I\) show attenuating oscillations with Rabi frequency \(\Omega\). In contrast, if \(\Gamma M_1 \neq \Gamma M_2\), the frequency of these oscillations diminishes, and they disappear when \(4 \Omega^2 < (\Gamma M_2 - \Gamma M_1)^2\).

In general we expect that tunnelling rates differ between different bridge levels, \(\Gamma M_1 \neq \Gamma M_2\).

Solving inhomogeneous Eq. (12) with the initial conditions
\[
R_1(0) = -r_2(0) = 0, \quad \lambda(0) = 0,
\]
and using Eqs. (10), (11) and (13), we get for the electronic current in the underdamped case \(4 \Omega^2 > (\Gamma M_2 - \Gamma M_1)^2\)
\[
I_e = e \frac{e}{2 \Gamma M_1 + \Gamma M_2} \left[ \frac{2 \Gamma M_1 \Omega^2}{\Gamma M_1 + \Gamma M_2} + \left( \Gamma M_2 - \Gamma M_1 \right) \right]
\]
\[
\times \left[ \frac{\Gamma M_1}{\Gamma M_1 + \Gamma M_2} + \Omega^2 \left( \frac{1}{\nu} + \frac{1}{\nu_1} \right) \exp(\nu t) \right] 
\]
\[
+ 2 \nu \left[ \frac{\Omega^2}{\nu_1} + \frac{\Gamma M_1 - \Gamma M_2}{\Gamma M_1 + \Gamma M_2} \right] \exp(\nu t) \right] 
\]
\[
\times \exp\left[ -\frac{1}{2} (\Gamma M_1 + \Gamma M_2) t \right] \right]
\]
(15)

If \(\Omega \gg \Gamma M_1, \Gamma M_2\), we obtain
\[
I_e = e \frac{e}{2 \Gamma M_2} \left[ \frac{2 \Gamma M_1 \Omega^2}{\Gamma M_1 + \Gamma M_2} + \left( \Gamma M_2 - \Gamma M_1 \right) \right]
\]
\[
- \cos(\nu t) \exp\left[ -\frac{1}{2} (\Gamma M_1 + \Gamma M_2) t \right] \right]
\]
(16)

that gives
\[
I_e = e \frac{e}{2 \Gamma M_2} \left[ 1 - \cos(\nu t) \exp(-\Gamma M_2 t) \right].
\]
(16a)

for \(\Gamma M_1 = \Gamma M_2 = \Gamma M = \frac{1}{2} (\Gamma M_1 + \Gamma M_2)\).

The beating phenomenon is seen to be seating on an underlying background. The corresponding expressions for the hole current \(I_h\) can be obtained from Eqs.(15) and (16) by the replacement \(\Gamma M_1 \rightarrow \Gamma M_2\) and \(\Gamma M_2 \rightarrow \Gamma M_1\).
Figure 2 The charge transferred after the completion of the pulse action as a function of its duration. $q=2Q/e$, $x=T_1/T_2$, with $T_1=40$, $T_2=40$ (solid line) and $T_2/T_1=1.9$ (dashed line). The value of $T_2=(T_1+T_2)/2$ is the same for both curves.

Indeed, for a given $T_2$ the product $T_1T_2$ is largest when $T_1=T_2$.

3 Conclusion In this work we have applied a theory developed by us for the light-induced current in tunnelling nanojunctions [7] to the experiments by Zrenner et al. [1] on converting Rabi oscillations in a strongly biased single-QD photodiode into deterministic photocurrents. We have shown that the behavior of a QD with different tunnel rates for electron and holes $T_1$ and $T_2$, respectively, is qualitatively different from that of a quantum dot with equal $T_1$ and $T_2$. In the latter case the current shows attenuating oscillations with the Rabi frequency. In contrast, for different tunnel rates, the frequency of these oscillations diminishes, and they disappear when $4T_1^2 \leq (T_1-T_2)^2$. We have obtained an analytical solution of the problem. Fig. 2 shows a somewhat different behavior of the transferred charge in the small attenuation limit for equal and different tunnelling rates for electrons and holes.

The method considered here, and the method for enhancing charge transfer based on the adiabatic rapid passage, which has been proposed in Ref. [7], taken together enable us to realize an optically triggered single-electron turnstile based on a bridge, which is characterized by both an homogeneously or inhomogeneously broadened optical transitions.

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