Nonlinear current response of a many-level tunneling system: Generation of higher harmonics

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The fully nonlinear response of a many-level noninteracting tunneling system to a strong alternating field of high frequency ω is studied in terms of the Schwinger-Keldysh nonequilibrium Green's functions. The nonlinear time dependent tunneling current $I(t)$ is calculated exactly and its resonance structure is elucidated. In particular, it is shown that under certain reasonable conditions on the physical parameters, the Fourier component I_n is sharply peaked at $n=(\Delta E/\hbar\omega)$, where ΔE is the spacing between two levels. This frequency multiplication results from the highly nonlinear process of n photon absorption (or emission) by the tunneling system. It is also conjectured that this effect (which so far is studied mainly in the context of nonlinear optics) might be experimentally feasible. [S0163-1829(97)04924-2]

I. INTRODUCTION

The physics of resonant tunneling through a single quantum well has been at the center of theoretical and experimental activity for more than two decades. Electron (or hole) confinement between penetrable barriers in a semiconductor enables the investigation of numerous important phenomena such as negative differential resistance, Coulomb blockade, single electron tunneling, single electron pump, and many others. The pertinent physics is very attractive both because of its richness and its potential device oriented nature.

At the core of the phenomena of resonance tunneling lies the relatively simple picture of a small cavity connected by tunneling barriers to two reservoirs of particles (also termed as leads). The dimensions of the cavity are small enough so that the energy levels inside it are well separated. These levels might be either single-particle levels whose spacing is determined solely by geometrical considerations or a few particle levels determined by interactions (as in the Coulomb blockade systems). If there is a difference between the chemical potential of the left lead (μ_L) and that of the right lead, (μ_R) a tunneling current occurs between the two leads. If an energy level of electrons in the cavity occurs between μ_L and μ_R this current displays a resonance structure. Evidently, the pertinent physics is time independent, namely, one speaks here of a dc. Moreover, in many cases, resonance tunneling through a single level can be treated within the formalism of the linear response.

Recently, interest is directed toward nonlinear time-dependent transport phenomena in double-barrier resonance tunneling systems. Experimentally, the investigation of the ac in mesoscopic devices proves to be feasible.[1] (Yet, theoretical analysis of the above experiment remains in the realm of linear response.[2–4])

The relevant physics inevitably becomes richer and more difficult to analyze. It touches upon qualitatively new phenomena which depend on how space- and time-dependent electronic states interfere. Among the effects which are inherently based on nonlinear response one might consider electron pumps,[5–7] lasers,[8] photon-assisted tunneling,[9–14] and others.

In the present work we concentrate on a relatively new effect, namely, frequency multiplication of the current response. As a theoretical model one may consider a double-barrier resonance tunneling system containing at least two quantum levels whose spacing ΔE introduces a new energy scale into the problem. The system is then subject to a strong monochromatic ac voltage of strength W and frequency ω, which results in a nonlinear current response $I(t)$, the main object of our study. The combination of strong ac electric field and level interaction with the reservoirs might lead to transitions between these levels which are assisted by many-photon emission (or absorption), a highly nonlinear effect. It can be analyzed in terms of the Fourier components I_n of the current $I(t)$. In linear response, one expects I_0 and I_1 to be the only nonzero components. Here, however, higher components may be significant. As we show below, I_n as a function of n is peaked at $n=\Delta E/\hbar\omega$, namely, there is a resonance when n photons are absorbed or emitted following the transition between the two levels. The transition occurs due to resonant tunneling from one of the levels to a lead and then to the other level. Frequency multiplication is familiar in nonlinear optics but, to the best of our knowledge has not yet been investigated in microelectronics. We give below some realistic estimates of this effect.

As for the theoretical treatment of the above model, we start from the familiar tunneling Hamiltonian and employ the Schwinger-Keldysh nonequilibrium Green's functions, through which the current is calculated in a straightforward way. In order to avoid complications while stressing the important role of two-level quantum wells we restrict ourselves in this work to noninteracting particles. As we explain in the next section we believe it to be a good model for the calculation of ac in quantum wells. We also argue there that the current response of a Coulomb blockade system in which the interaction is reflected through the charging energy is likely to posses footprints of one-particle motion. In particular, we think that the high freqency resonance leading to frequency multiplication which we describe in the present paper will show also in quantum dots. Of course, quantative treatment of the nonlinear ac response of a general interacting system in a resonance tunneling device goes beyond the scope of the present work.

In the following section the problem is formulated, and

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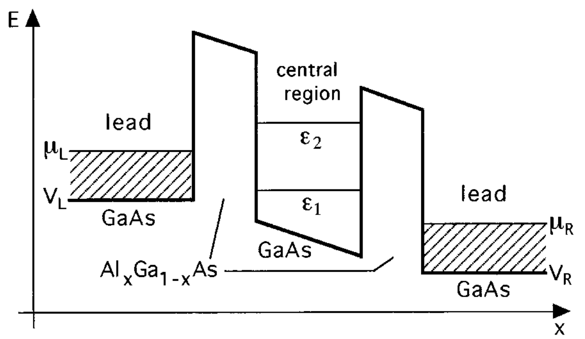

FIG. 1. Schematic energy diagram of the conduction band for an $Al_xGa_{1-x}As$ quantum well in the absence of an ac field. x is the axis perpendicular to the layers, μ_L and μ_R are the left and the right chemical potentials correspondently, V_L and V_R are the energies of the conduction band bottoms of the leads, ϵ_1 and ϵ_2 are the energies of the levels in the central region.

the various parts of the pertinent Hamiltonians are defined and justified. Then, in Sec. III the method of solution in terms of the Keldysh Green's function is introduced. In particular, the free particle Green's functions are written down and the Dyson equation for the ''lesser'' Green's function is derived. Section IV is devoted to the discussion of the tunneling current. An exact expression for the current is derived in terms of Keldysh Green's functions and the origin of resonances in the nonlinear response is explicitly elucidated. Analysis of higher harmonic generation is carried out in Sec. V, where numerical results are presented and the conditions for obtaining peaks at higher harmonics are discussed. The paper is then concluded with a short summary. In Appendix A there is a proof that a pure time dependent potential has no physically observable effect, while some technical point related to the derivation of Dyson equation in the Keldysh formalism is explained in Appendix B.

II. FORMULATION OF THE PROBLEM

We consider a structure where a charge carrier has two barriers on its way like the one drawn in Fig. 1. This structure is analogous to the Fabry-Perot resonator — the motion of a carrier is almost quantized in the central region but it still can escape into the leads. Thus the energy levels of the central region provide resonances for the transmission of the charge carries from one lead to another. Hereafter we will speak about electrons. The same type of structure can be made for holes but the degeneracy of the valence band can complicate the calculation. An arbitrary combination of dc and ac potentials is applied to the above structure (the potential differences are still required to be small compared with the Fermi energies). Our main goal is to calculate the time-dependent currents in the system.

Before doing this let us mention possible experimental realizations. Practically a two-barrier structure can be fabricated in several ways, e.g., (1) by putting several layers of different semiconductors having matching lattices and different gap widths one on top of the other. [15–27] The electrons then move in the direction perpendicular to the layers. As an example, the profile of the conduction band for a particular case of a GaAs-$Al_xGa_{1-x}As$ structure is shown in Fig. 1. (2) By application of an external gate potential.[10,11,28–37,40] The electrons then move along the layers but their movement is restricted by the external repulsive potential. The gate is shaped such that the electrons move in one direction and two barriers are formed in their way. (3) By placing an impurity in a tunnel barrier.[38,39,41]

In order to describe the dynamics of the system we use the tunneling Hamiltonian formalism suggested a long time ago.[42,43] The total Hamiltonian is

$$H(t)=H_f+H_{ac}(t)+H_T, \tag{1}$$

where

$$H_f=\sum_k \epsilon_k a_k^\dagger a_k+\sum_n \epsilon_n c_n^\dagger c_n+\sum_p \epsilon_p b_p^\dagger b_p \tag{2}$$

is the Hamiltonian of free particles with neither ac field nor coupling between the leads and the central region. Here ϵ_k, ϵ_n, and ϵ_p are free-particle energies in the left lead, the central region, and the right lead, respectively. The energy is taken with respect to the bottom of the conduction band in the central region. The operators $a_k^\dagger$, a_k, $b_p^\dagger$, b_p, $c_n^\dagger$, c_n are creation and annihilation operators in the leads and in the central region. Furthermore, k (p) are momenta in the direction perpendicular to the layers, and n counts the levels in the central region [hereafter k (p) refer to the left (right) lead, n, m, n', m' — to the central region]. The summation over n in Eq. (2) expresses the presence of more than one energy level. The third term,

$$H_T=\sum_{k,n} (T_{nk}^L c_n^\dagger a_k+T_{nk}^{L*} a_k^\dagger c_n)+\sum_{p,n} (T_{pn}^R b_p^\dagger c_n+T_{pn}^{R*} c_n^\dagger b_p) \tag{3}$$

is the part responsible for the tunneling through the barriers. $T_{nk(p)}^{L(R)}$ are transfer matrix elements between the leads and the central region. Finally, the time dependent part is

$$H_{ac}(t)=W_L\sin(\omega t)\sum_k a_k^\dagger a_k+W_R\sin(\omega t)\sum_p b_p^\dagger b_p, \tag{4}$$

where $W_{L(R)}\sin(\omega t)$ are potential shifts of the leads with respect to the central region caused by an external ac potential. dc-potential shifts are included into the energies $\epsilon_{k(n,p)}$. The arrangement in which W_L and W_R have opposite signs corresponds to an application of an ac bias as it was done in a few experiments.[16,22,34] The choice $W_L=W_R$ describes an application of an ac voltage to the gate electrode superimposed on the central region (see Refs. 10 and 11). We notice here that shifting both leads together (having the potential of the central region fixed) is equivalent to shifting the central region (having the potentials of the leads fixed) since an application of a uniform (time-dependent) potential is not observable even if it is arbitrary strong and arbitrary fast (see Appendix A). The situation $W_L=0$, $W_R\neq 0$ corresponds to an application of an ac voltage only to one barrier as it was done in the experiment reported in Ref. 10.

Our choice of H_{ac} is based on the following assumptions:

(1) The electrons in the leads respond to an applied field very fast since we deal with the frequencies much less than the plasma frequency. It means that any change of the external potential causes an immediate rearrangement of the elec-

trons. In other words, the internal potential responds very quickly to an external field.[2,44]

(2) The concentration of the electrons in the leads is high enough to screen an external field.[45–47] Therefore the potential is uniform in the leads and drops in the barriers.[9,44,45,47–55]

(3) We used a widespread assumption[44,49,54,56] that the probability of direct transitions between the energy levels in the central region due to the ac field is small and can be neglected.

We do not restrict our consideration to the case of small W_L, W_R (linear response). They can be arbitrary large. A strong ac field ($W_L \gg \omega$ or $W_R \gg \omega$) leads to a nonlinearity. One of its signatures is the generation of current harmonics with frequencies much larger than ω.

The Hamiltonian (1) does not include Coulomb interaction. We think it is a good approximation for quantum wells. Noninteracting model has been widely used for their treatment (Refs. 44, 45, 47, 49–51, 57–64 and others). The Coulomb interaction can be included in the framework of the mean-field approximation[16,65–67] because the number of electrons in quantum wells is very large. Then its influence is expressed in concentration-dependent corrections to the one-particle energies ϵ_n. It can lead to some changes in the dependence of the current on the dc bias (such as the appearance of a hysteresis[16,65–67]) or, may be, on the magnitude of the frequency of the ac field. Nevertheless, if the population of the energy levels does not change drastically during the period of the ac field the physics of the electron-photon interaction remains essentially within the independent-particle model.

In quantum dots competition of Coulomb interaction with positive gate potential restricts the number of possible charge states. If the gate voltage is less than the Coulomb interaction energy this number is two, namely, one of them has N electrons and the other one has $N+1$ electrons[68] (typically N is about 10–100). Tunneling of an electron then blocks the tunneling of any other one. In the framework of the classical Coulomb blockade model, where the charging energy does not depend on the population of different energy levels but only on the total amount of electrons in the dot, this is the main effect of the Coulomb interaction (at least at temperatures higher than the Kondo temperature). Therefore, if the number of electrons in the dot is fixed, the motion of every electron is largely determined by geometrical considerations as in the noninteracting model. The Coulomb interaction of an electron with the other ones just provides it with the potential energy to overcome the gate potential. This statement is clearly seen from inspection of the Coulomb blockade Hamiltonian (H_d): $H_d = \Sigma_i(\epsilon_i - eV)n_i + \frac{1}{2}\Sigma_{i\neq j} U n_i n_j = \Sigma_i(\epsilon_i - eV + UN)n_i$, where V is the gate potential, U is the Coulomb energy, i,j are the numbers of energy levels of the dot, and n_i are their occupations. In the systems we speak about here tunneling through the barriers is small. Although it provides all the current the electrons are kept in the central region (or in the leads) most of the time. Within the master equation approach we would say that the probability of one of the electrons to tunnel from the dot to a lead during the time Δt is $P = (1 - e^{-\Gamma \Delta t})e^{-N\Gamma\Delta t} \approx \Gamma \Delta t$. In the lowest order approximation we can consider the motion of every electron in the ac field neglecting the possibility that one of the other electrons will escape into the leads. Then it is roughly described by the noniteracting model. Of course, the full quantative treatment of a quantum dot in an external ac field is beyond the capabilities of this model. But we think that the features of the one-particle motion found in the present work, in particular the high freqency resonance leading to frequency multiplication, will show in the interacting system too.

Strictly speaking, the Hamiltonian (1) is one dimensional. In realistic quantum wells there are, of course lateral degrees of freedom provided by the motion along the layers. But at every particular value of the lateral momentum the problem is one dimensional. Even if the dependence of electron energy on the lateral momentum is important the total current can be found by the simple integration over it. The lateral degrees of freedom can be removed by a magnetic field perpendicular to the layers.

III. SOLUTION IN TERMS OF KELDYSH GREEN'S FUNCTIONS

In order to work out the current we first have to analyze the electron propagation. To this end we employ the nonequilibrium Green's function technique suggested by Schwinger,[69] Kadanoff and Baym,[70] and Keldysh[71] (for a review see Refs. 52,72–76). Since no equilibrium is required one can rigorously consider large perturbations and high frequencies drawing the system far from its steady state. The method uses a time variable defined on two sides of the real axis. It is equivalent to the introduction of two *independent* Green's functions, one of which characterizes the dynamical properties of the particles the other one describes their distribution.[71]

It is convenient to use the following two Green's functions (the others can be expressed through them):

$$G^r_{i,j}(t_1,t) = -i\,\theta(t_1-t)\langle \varphi_i(t_1)\varphi_j^\dagger(t) + \varphi_j^\dagger(t)\varphi_i(t_1)\rangle, \tag{5}$$

$$G^<_{i,j}(t_1,t) = i\langle \varphi_j^\dagger(t)\varphi_i(t_1)\rangle,$$

where $\varphi_i(t_1)(\varphi_j^\dagger(t))$ are operators in the Heisenberg picture representing $a_k(t_1)$, $c_n(t_1)$, $b_p(t_1)$ ($a_k^\dagger(t)$, $c_n^\dagger(t)$, $b_p^\dagger(t)$) in correspondence with what values the indexes i and j take (k, n, or p). The first Green's function is the usual retarded Green's function. The advanced Green's function (G^a_{ij}) is its Hermitian conjugate that is $G^a_{i,j}(t_1,t) = [G^r_{ji}(t,t_1)]^*$. The other one contains information about the distribution of electrons and their correlations. At $t_1 = t$ it gives the one-particle density matrix (ρ):[72] $G^<_{ij}(t,t) = iN\rho_{ij}(t)$, where N is the number of particles in the system. The advantage of the method is that it treats G^r and $G^<$ in a unified manner. Both of them can be found through the Dyson's equation:[74]

$$G^r = g^r[1 + \Sigma^r G^r], \tag{6}$$

$$G^< = [1 + G^r\Sigma^r]g^<[1 + \Sigma^a G^a] + G^r\Sigma^< G^a. \tag{7}$$

Here multiplication implies the summation (or integration) over space variables and integration over time, g^r and $g^<$ are Green's functions defined as Eq. (5) but for an unperturbed Hamiltonian, Σ^r, Σ^a, and $\Sigma^<$ are proper irreducible self-energies.

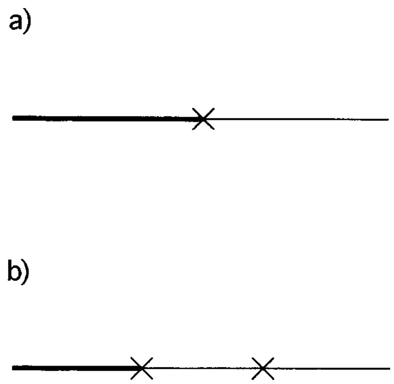

FIG. 2. Tunneling diagrams. A cross denotes a tunneling event, a thin line denotes a free propagator, and a thick line denotes a full propagator. (a) Dyson's equation. Only a single tunneling event contributes to the self-energy. (b) Any other diagram is reducible.

An application of the formalism to tunneling systems is especially powerful if one chooses H_T as a perturbation.[77] Then the self-energies assume a very simple form:

$$\begin{aligned}
&\Sigma^{r(a)}_{kk'}=\Sigma^{r(a)}_{pp'}=\Sigma^{r(a)}_{kp}=\Sigma^{r(a)}_{pk}=0,\\
&\Sigma^{r}_{nk}=\Sigma^{a}_{nk}=T^{L}_{nk},\\
&\Sigma^{r}_{kn}=\Sigma^{a}_{kn}=T^{L}_{kn}=T^{L*}_{nk},\\
&\Sigma^{r}_{np}=\Sigma^{a}_{np}=T^{R}_{np},\\
&\Sigma^{r}_{pn}=\Sigma^{a}_{pn}=T^{R}_{pn}=T^{R*}_{np},\\
&\Sigma^{<}\equiv 0.
\end{aligned}\tag{8}$$

This is due to the fact that any vertex caused by H_T has only two entries. The only diagram contributing to an irreducible self-energy is the simplest one drawn in Fig. 2(a). The next one drawn in Fig. 2(b) is already reducible.

In the next section we will show that tunneling currents can be expressed solely in terms of the Green's functions in the central region. To find these functions we iterate Eq. (6) and get

$$G^{r}_{nm}(t_1,t)=g^{r}_{nm}(t_1,t)+\sum_{n'}\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}dt_2dt_3g^{r}_{nn}(t_1,t_2)\times X_{nn'}(t_2,t_3)G^{r}_{n'm}(t_3,t),\tag{9}$$

$$X_{nn'}(t_2,t_3)\equiv X^{L}_{nn'}(t_2,t_3)+X^{R}_{nn'}(t_2,t_3),\tag{10}$$

$$X^{L}_{nn'}(t_2,t_3)\equiv\sum_{k}T^{L}_{nk}g^{r}_{kk}(t_2,t_3)T^{L}_{kn'},$$

$$X^{R}_{nn'}(t_2,t_3)\equiv\sum_{p}T^{R}_{np}g^{r}_{pp}(t_2,t_3)T^{R}_{pn'}.$$

Equations (7) and (8) establish an algorithm for calculating $G^{<}_{nm}$ without an iteration but nevertheless it contains eleven terms on the right hand side. We showed (see Appendix B) that only two of them remain after a long enough period of time has passed from the moment the tunneling was switched on

$$G^{<}_{nm}(t_1,t)=\sum_{n'm'}\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty}dt_2dt_3G^{r}_{nn'}(t_1,t_2)\times Y_{n'm'}(t_2,t_3)G^{a}_{m'm}(t_3,t),\tag{11}$$

where

$$Y_{n'm'}(t_2,t_3)\equiv Y^{L}_{n'm'}(t_2,t_3)+Y^{R}_{n'm'}(t_2,t_3),\tag{12}$$

$$Y^{L}_{n'm'}(t_2,t_3)\equiv\sum_{k}T^{L}_{n'k}g^{<}_{kk}(t_2,t_3)T^{L}_{km'},$$

$$Y^{R}_{n'm'}(t_2,t_3)\equiv\sum_{p}T^{R}_{n'p}g^{<}_{pp}(t_2,t_3)T^{R}_{pm'}.$$

We notice that $G^{<}_{nm}$ does not depend on the initial distribution of electrons in the central region [the reason is that $g^{<}_{nm}$ does not appear in formula (11)]. It depends only on their distributions in the leads through $g^{<}_{kk}$ and $g^{<}_{pp}$.

It is easy to show (see Appendix A) that even under a time-dependent potential of an arbitrary large amplitude and an arbitrary high frequency the Green's functions for electrons in isolated leads are given by ''adiabaticlike'' expressions:

$$\begin{aligned}
g^{r}_{kk'}(t_2,t_3)&=-i\delta_{kk'}\theta(t_2-t_3)e-i\int_{t_3}^{t_2}[H_f+H_{\mathrm{ac}}(t)]_{kk}dt\\
&=-i\delta_{kk'}\theta(t_2-t_3)\\
&\quad\times e^{-i\epsilon_k(t_2-t_3)+i(W_L/\omega)[\cos(\omega t_2)-\cos(\omega t_3)]},
\end{aligned}\tag{13}$$

$$\begin{aligned}
g^{<}_{kk'}(t_2,t_3)&=i\delta_{kk'}f_L(\epsilon_k)e-i\int_{t_3}^{t_2}[H_f+H_{\mathrm{ac}}(t)]_{kk}dt\\
&=i\delta_{kk'}f_L(\epsilon_k)e^{-i\epsilon_k(t_2-t_3)+i(W_L/\omega)[\cos(\omega t_2)-\cos(\omega t_3)]},
\end{aligned}$$

provided that the time-dependent perturbing potential (H_{ac}) is uniform in every lead. Here $[\,]_{kk}$ denotes matrix element, $f_L(\epsilon_k)=1/\{\exp[(\epsilon_k-\mu)/k\Theta]+1\}$ is the Fermi function for the left lead, μ_L is its chemical potential, and Θ is the temperature. Formulas for $g^{r}_{pp'}$, $g^{<}_{pp'}$ can be obtained from Eq. (13) by replacements $k,k'\rightarrow p,p'$, $W_L\rightarrow W_R$, $\mu_L\rightarrow\mu_R$.

Now we substitute Eq. (13) into Eqs. (10) and (12) and replace the sum over k, p by an integral over energy. Further, we assume wide energy bands in the leads and approximate the elastic couplings to the leads ($\Gamma^{L(R)}_{nn'}(\epsilon_{k(p)})$) by Lorentzian functions:[78]

$$\Gamma^{L}_{nn'}(\epsilon_k)\equiv 2\pi\rho(\epsilon_k)T^{L}_{nk}T^{L}_{kn'}\equiv\Gamma^{L}_{nn'}\frac{D^2}{\epsilon_k^2+D^2},\tag{14}$$

$$\Gamma^{R}_{nn'}(\epsilon_p)\equiv 2\pi\rho(\epsilon_p)T^{R}_{np}T^{R}_{pn'}\equiv\Gamma^{R}_{nn'}\frac{D^2}{\epsilon_p^2+D^2}.$$

Since in the real experimental systems the bandwidths are usually much larger than other relevant energy scales (W,

ω, dc - bias, Θ, Γ, etc.), we take the limit $D\to\infty$ where it is possible. Substituting (13) and (14) into (10) we obtain

$$\begin{aligned}X^L_{nn'}(t_2,t_3)&=-i\Gamma^L_{nn'}\theta(t_2-t_3)e^{i(W_L/\omega)[\cos(\omega t_2)-\cos(\omega t_3)]}\\&\quad\times\int\frac{d\epsilon_k}{2\pi}\frac{D^2}{\epsilon_k^2+D^2}e^{-i\epsilon_k(t_2-t_3)}\\&=-i\Gamma^L_{nn'}\theta(t_2-t_3)e^{i(W_L/\omega)[\cos(\omega t_2)-\cos(\omega t_3)]}\\&\quad\times\frac{D}{2}e^{-D|t_2-t_3|}.\end{aligned}$$

At this stage let us recall that $X^L_{nn'}(t_2,t_3)$ can be considered as a generalized function [it appears only under integration with another function in Eq. (10)]. It is easy to see that in the limit $D\to\infty$,

$$X_{nn'}(t_2,t_3)=-\frac{i}{2}\Gamma_{nn'}\delta(t_2-t_3). \tag{15}$$

The factor $\frac{1}{2}$ appears due to the presence of the step function $\theta(t_2-t_3)$. Substituting Eqs. (13) and (14) into (12) we obtain

$$\begin{aligned}Y_{n'm'}(t_2,t_3)&=i\Gamma^L_{n'm'}e^{i(W_L/\omega)[\cos(\omega t_2)-\cos(\omega t_3)]}\\&\quad\times\int\frac{d\epsilon_k}{2\pi}\frac{D^2}{\epsilon_k^2+D^2}f_L(\epsilon_k)e^{-i\epsilon_k(t_2-t_3)}\\&\quad+i\Gamma^R_{n'm'}e^{i(W_R/\omega)[\cos(\omega t_2)-\cos(\omega t_3)]}\\&\quad\times\int\frac{d\epsilon_p}{2\pi}\frac{D^2}{\epsilon_p^2+D^2}f_R(\epsilon_p)e^{-i\epsilon_p(t_2-t_3)},\end{aligned} \tag{16}$$

where $\Gamma_{nn'}\equiv\Gamma^L_{nn'}+\Gamma^R_{nn'}$. We cannot put here $D\to\infty$ because it would lead to a logarithmic divergence in the ac.

We want to emphasize that substituting Eq. (15) into (9) yields a time-independent equation. The problem then reduces to the task of finding the usual retarded Green's function for a time-independent potential well with finite barriers. This is because in the limit $D\to\infty$ the bands are uniform in energy. The ac field then emerges solely within a time- dependent shift of the chemical potentials. The function G^r_{nm} (unlike $G^<_{nm}$) is not sensitive to it (see discussion at the end of the section). The Fourier transform of the resulting equation is

$$G^r_{nm}(\epsilon)+\frac{i}{2}g^r_{nn}(\epsilon)\sum_{n'}\Gamma_{nn'}G^r_{n'm}(\epsilon)=g^r_{nm}(\epsilon). \tag{17}$$

A rigorous solution for a two-level system shows that the mixing terms G^r_{nm}, $n\neq m$ give a contribution to the current of the order of $\Gamma/|\epsilon_n-\epsilon_m|$. We assume $\Gamma\ll|\epsilon_n-\epsilon_m|$ and drop them out hereafter. With the same accuracy G^r_{nn} are given by

$$G^r_{nn}(\epsilon)=\frac{1}{\epsilon-\epsilon_n+i\frac{\Gamma_n}{2}} \tag{18}$$

where $\Gamma_n\equiv\Gamma_{nn}+\Gamma^{\rm in}_n$, $\Gamma^{\rm in}_n$ are intrinsic level widths due to inelastic interactions, leakage of electrons into lateral directions, etc. This is equivalent to the widely used assumption that the quasilevels of a potential well with finite barriers have complex energies $\epsilon_n-i(\Gamma_n/2)$. We see that the main contribution of tunneling to the *dynamical* properties is contained in level broadening.

Substituting (16) into (11) gives an exact expression for $G^<$ in terms of G^r:

$$\begin{aligned}G^<_{nm}(t_1,t)&=\int\frac{d\epsilon_k}{2\pi}\frac{D^2f_L(\epsilon_k)}{\epsilon_k^2+D^2}e^{-i\epsilon_k(t_1-t)}\\&\quad\times\sum_{s,q=-\infty}^{\infty}i^{s-q+1}J_s\left(\frac{W_L}{\omega}\right)J_q\left(\frac{W_L}{\omega}\right)e^{-is\omega t_1+iq\omega t}\\&\quad\times\sum_{n'm'}\Gamma^L_{n'm'}G^r_{nn'}(\epsilon_k+s\omega)G^{r*}_{mm'}(\epsilon_k+q\omega)\\&\quad+\int\frac{d\epsilon_p}{2\pi}\frac{D^2f_R(\epsilon_p)}{\epsilon_p^2+D^2}e^{-i\epsilon_p(t_1-t)}\\&\quad\times\sum_{s,q=-\infty}^{\infty}i^{s-q+1}J_s\left(\frac{W_R}{\omega}\right)J_q\left(\frac{W_R}{\omega}\right)e^{-is\omega t_1+iq\omega t}\\&\quad\times\sum_{n'm'}\Gamma^R_{n'm'}G^r_{nn'}(\epsilon_p+s\omega)G^{r*}_{mm'}(\epsilon_p+q\omega).\end{aligned} \tag{19}$$

We notice that the transition amplitudes $G^<_{nm}, n\neq m$ are important as we show in the next section.

This difference between G^r_{nm} and $G^<_{nm}$, $n\neq m$ arises from the difference between X and Y in the formulas (15), (16). The quantity X has such a simple form because $f(\epsilon_{k(p)})$ does not appear in g^r_{kk}, g^r_{pp} so that the summation over $k(p)$ [equivalent to integration over $\epsilon_k(\epsilon_p)$] in formula (10) gives $\frac{1}{2}\delta(t_2-t_3)$ in the limit $D\to\infty$. The occurrence of $f(\epsilon_{k(p)})$ in $g^<_{kk}$ ($g^<_{pp}$) leads to an upper limit of integration in Eq. (16) around $\mu_{L(R)}$. If $\mu_{L(R)}$ is large enough $\frac{1}{2}\delta(t_2-t_3)$ is recovered. Then the whole solution becomes invariant under time translation and the transition amplitudes $G^<_{nm}$, $n\neq m$ are small, of the order of $\Gamma/|\epsilon_n-\epsilon_m|$.

IV. CALCULATION OF THE CURRENT

A double-barrier structure is integrated within a circuit. The measured current is determined by its influence on the circuit. If the barriers are modeled as capacitive-resistant elements[81] the currents in the leads are given by[12]

$$\begin{aligned}I_L(t)&=\frac{C_R+C_g}{C}C_L\omega W_L\cos(\omega t)-\frac{C_L}{C}C_R\omega W_R\cos(\omega t)\\&\quad+\frac{C_R+C_g}{C}I^T_L(t)-\frac{C_L}{C}I^T_R(t),\end{aligned} \tag{20}$$

where I_L is the total current in the left lead $I^T_L(t)$ and $I^T_R(t)$ are tunneling currents through the left and right barriers, respectively, $C_{L(R)}$ and C_g are capacitances of the central region relative to the left (right) lead and a gate electrode (or another background), $C\equiv C_L+C_R+C_g$. The current in the right lead is obtained by replacing L by R. The first two terms describe the contribution of capacitive currents (due to the presence of accumulation and depletion layers). They have the frequency of the input ac voltage. But the tunneling currents contain also higher harmonics as we show below.

The tunneling current from the left lead into the central region is defined by the change in the number of electrons in that lead:

$$I_L^T(t)=e\left\langle \frac{d\hat{N}_L}{dt}\right\rangle = -\frac{ie}{\hbar}\sum_{km}\left(T_{mk}^L\langle c_m^\dagger a_k\rangle - T_{mk}^{L*}\langle a_k^\dagger c_m\rangle\right)$$

$$= -\frac{2e}{\hbar}\,\mathrm{Re}\left[\sum_{km} T_{mk}^L G_{km}^<(t,t)\right], \tag{21}$$

where $\hat{N}_L \equiv \Sigma_k a_k^\dagger a_k$. The tunneling current from the right lead into the central region (I_R^T) is obtained from the above expression by a change $L\to R$, $k\to p$, $a_k \to b_p$.

Using Dyson's equation for $G_{kn}^<$ allows one to express the tunneling currents through Green's functions in the central region (which we found in the previous section):

$$I_{L(R)}^T(t) = -\frac{2e}{\hbar}\,\mathrm{Re}\left\{\sum_{nm}\int_{-\infty}^{+\infty} dt_1 [X_{mn}^{L(R)}(t,t_1) G_{nm}^<(t_1,t)\right.$$

$$\left. + Y_{mn}^{L(R)}(t,t_1) G_{nm}^a(t_1,t)]\right\}. \tag{22}$$

Assuming wide bands as we did in the previous section we can use the formulas (15), (16), (18), and (19). Dropping out the terms that are always small of the order of $\Gamma/|\epsilon_j-\epsilon_{j'}|$ in comparison with the others we obtain after some algebra

$$I_L^T(t) = \frac{1}{2}I_L^0 + \sum_{n=1}^{+\infty} |I_L^n|\cos(n\omega t+\phi_L^n), \quad \text{where } \phi_L^n = \arg I_L^n, \tag{23}$$

$$I_L^n = \frac{2e}{\hbar}\sum_j [A_j(n)+A_j^*(-n)+B_j(n)+B_j^*(-n)],$$

$$A_j(n) = i^{-n}\left[i\Gamma_{jj}^L + \sum_{j'} \frac{|\Gamma_{jj'}^L|^2}{\epsilon_{j'}-\epsilon_j - n\omega + i\dfrac{\Gamma_j+\Gamma_{j'}}{2}}\right]$$

$$\times \sum_{s=-\infty}^{+\infty} J_s\left(\frac{W_L}{\omega}\right) J_{s+n}\left(\frac{W_L}{\omega}\right)$$

$$\times F\left(\frac{\epsilon_j-\mu_L}{\omega} - s, \frac{\Gamma_j}{\omega}, \frac{k\Theta}{\omega}, D\right),$$

$$B_j(n) = i^{-n}\sum_{j'} \frac{\Gamma_{jj'}^L \Gamma_{j'j}^R}{\epsilon_{j'}-\epsilon_j - n\omega + i\dfrac{\Gamma_j+\Gamma_{j'}}{2}}$$

$$\times \sum_{s=-\infty}^{+\infty} J_s\left(\frac{W_R}{\omega}\right) J_{s+n}\left(\frac{W_R}{\omega}\right)$$

$$\times F\left(\frac{\epsilon_j-\mu_R}{\omega} - s, \frac{\Gamma_j}{\omega}, \frac{k\Theta}{\omega}, D\right),$$

$$F\left(\frac{\epsilon_j-\mu}{\omega} - s, \frac{\Gamma_j}{\omega}, \frac{k\Theta}{\omega}, D\right)$$

$$= \frac{1}{2\pi}\int \frac{D^2 f(\epsilon)}{\epsilon^2+D^2} \frac{d\epsilon}{\epsilon + s\omega - \epsilon_j + i\dfrac{\Gamma_j}{2}},$$

$$f(\epsilon) = \frac{1}{[e^{[(\epsilon-\mu)/k\Theta]}+1}.$$

Analogous expressions hold for $I_R^T(t)$ as well. For the rest of our discussion we choose to study I_L^T. We point out that the current consists of many harmonics with frequencies $n\omega$. The sums over j,j' express the summation over all energy levels in the central region. We notice that the terms $A_j(n)$ and $B_j(n)$ are quite similar in structure. The first one includes the currents due to direct photon-assisted transitions between the left lead and the central region and due to the effect of the left lead on the population of the central region. The second one describes the current through the left barrier due to the influence of the right lead on the electrons in the central region. The main difference between them (beside a few replacements of L into R) is that the first term in the brackets in A (i.e., $i\Gamma_{jj}^L$) is absent in $B_j(n)$. At zero temperature the integral in the expression for F can be easily computed. For $D \gg W,|\mu|,|\epsilon_j|,\Gamma|$ the result is very simple:

$$F\left(\frac{\epsilon_j-\mu}{\omega} - s, \frac{\Gamma_j}{\omega}, 0, D\right) = \frac{1}{2\pi}\ln\frac{\epsilon_j-\mu-s\omega - i\dfrac{\Gamma_j}{2}}{D}.$$

The rest of our calculation is done at zero temperature.

The term in A containing Γ_{jj} gives an additive contribution of different energy levels to the current. The same is true for the terms of the sums over j' [in $A_j(n)$ and $B_j(n)$] with $j'=j$. The other terms in these sums are caused by the presence of different energy levels (the presence of at least two different levels is necessary). Each contribution is caused by a certain *pair* of energy levels. It is obvious that they have a resonance nature. The j'th term can be large when $|\epsilon_{j'}-\epsilon_j-n\omega|<(\Gamma_{j'}+\Gamma_j)/2$. Thus it can give a large ac at $n_{\mathrm{res}}\approx|\epsilon_{j'}-\epsilon_j|/\omega$ (under condition $W>|\epsilon_{j'}-\epsilon_j|$). If $\omega \ll |\epsilon_{j'}-\epsilon_j|$ one has $n_{\mathrm{res}}\gg 1$, i.e., the frequency in the output is much larger than in the input. We notice that this term generates the n_{res}th harmonic exclusively. The others are small, of the order of $[(\Gamma_j+\Gamma_{j'})/2\omega(n-n_{\mathrm{res}})]$ (we suppose high frequency $\Gamma/\omega \ll 1$).

The resonance is caused by many-photon absorption due to the strength ($W\gg\omega$) of the ac field. The formula (21) expresses the current at the moment of time t through the probability to create an electron in the state m annihilating it in the state k as it is drawn in Fig. 3(a). To do that it is necessary to have an electron in the state k and a hole in the state m. The formula (22) shows that it is provided by the creation of an electron in the state k and a hole in the state n at an earlier moment of time $t_1, t_1 \leq t$ [see Fig. 3(b)]. In the presence of a strong ac field the process in which $n\neq m$ should be considered. The hole then has to propagate from n to m. The formula (11) expresses the transition from n to m through the kind of process drawn in Fig. 3(c) for

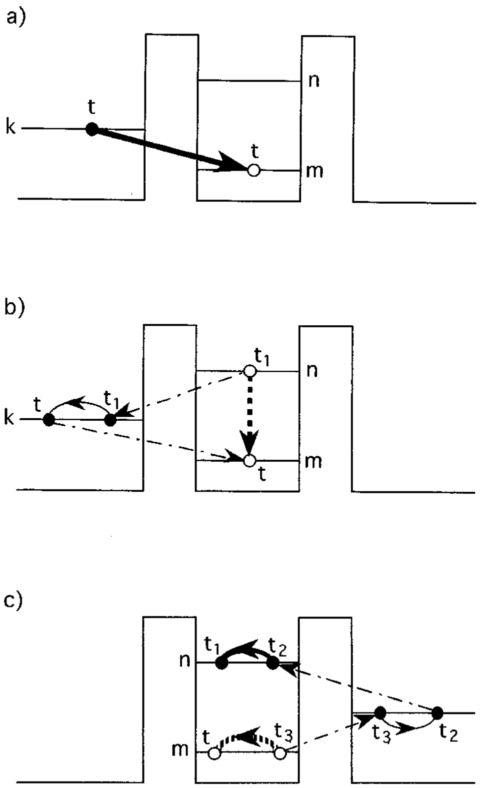

FIG. 3. Tunneling processes leading to the high harmonic resonance. A thick black line denotes a full electron propagator; a thick dashed line denotes a full hole propagator; a thin black line denotes free electron propagator; a thin dash-dotted line denotes tunneling event. (a) The tunneling current is produced by electrons going from the lead to a level in the central region and back. (b) This process can be assisted by other levels if the ac field is strong. (c) Resonance transfer of an electron from the level m to the level n possible in a strong ac field.

$t_3<t_2$. An electron is transferred at the moment t_3, $t_3\leqslant t$ from the state m to the state p in a lead leaving a hole in the state m. At the moment t_2, $t_2\leqslant t_1$ the electron tunnels to the state n. Eventually an electron is found in the state n at t_1 and a hole is found in the state m at t. In the absence of a time-dependent potential this process is impossible because the transitions from m to p and from p to n have to conserve energy (after integration over t_2, t_3). Thus m has to be equal to n. The presence of time-dependent part $H_{ac}(t)$ in the Hamiltonian results in a multiplication of the wave function of the state p by the phase factor $e^{i(W_R/\omega)\cos(\omega t)}$ [see Appendix A and formula (13) in the main text]. In the energy representation the wave function becomes dispersed over all energies $\epsilon_p\pm s\omega$ (where s is an integer), with weights $J_s(W_R/\omega)$. In other words the spectral function of the state p has extra peaks with energies $\epsilon_p\pm s\omega$. Their magnitudes are proportional to $J_s^2(W_R/\omega)$ (see Ref. 9). Thus it is possible to have transitions from m to p and from p to n when

$$\epsilon_m-\epsilon_p=q\omega,\quad \epsilon_n-\epsilon_p=s\omega. \tag{24}$$

This is the origin of J_s and J_q in the formulas (19), (23). These transitions are accompanied by the emission (absorption) of q (s) photons. The total number of absorbed photons

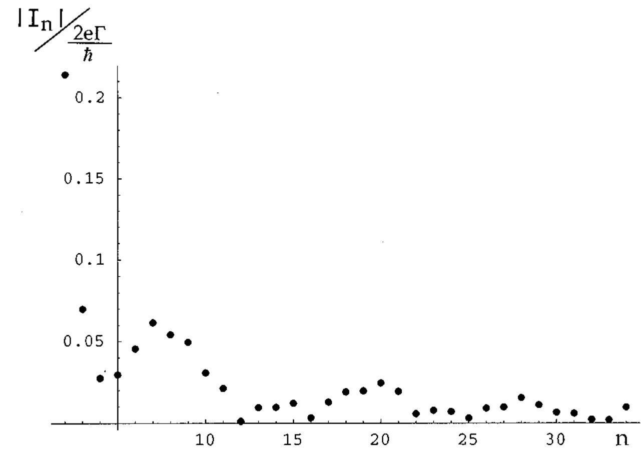

FIG. 4. Spectrum of the tunneling current (amplitude of the harmonics via their number). General situation. $W_L=W_R=30$, $\epsilon_1=11.5$, $\epsilon_2=30.5$, $\mu_R=20$, $\mu_L=5$, $D=70$, $\Gamma\equiv\Gamma_{11}^{L(R)}=\Gamma_{22}^{L(R)}=\Gamma_{12}^{L}=-\Gamma_{12}^{R}=0.05$, $\omega=1$. The dc and the first harmonic are not shown.

is $s-q=n$. When the ac field is strong ($W_R\gg\omega$) $J_s(W_R/\omega)$ and $J_q(W_R/\omega)$ can be large for $s,q\gg1$. This leads to a possibility of many - photon absorption (emission). The resonance conditions (24) give $n_{\rm res}=s-q=[(\epsilon_m-\epsilon_n)/\omega]$. It is easy to show that, in fact, $W_R>n_{\rm res}\omega$ is required to obtain a large $n_{\rm res}$th harmonic. The physical content of Fig. 3(c) expresses the contribution of $Y_{nm}^R(t_2,t_3)$ into Eq. (11). The same process but through the left lead expresses the contribution of $Y_{nm}^L(t_2,t_3)$. At $t_2<t_3$ the picture is very similar. The process just starts from a transfer of an electron from p to n at t_2. Then the hole tunnels from p to m at t_3.

If the lead energy bands are not wide then the main change is that $G_{nm}^{r(a)}$, $n\neq m$ is not negligible. These nondiagonal elements of the retarded Green's functions could make a positive or negative contribution to the process drawn in Fig. 3(c). Yet, we argue that the resonance would persist also in that case.

Every pair of energy levels in the central region gives an independent contribution to the current (23). In the next section we consider one pair of energy levels and explore the dependence of the current it produces on the parameters of the system.

V. HIGH HARMONICS GENERATION

In this section we consider a strong ac field ($W_L\gg\omega$ or $W_R\gg\omega$) and show the dependence of the current on the parameters of the system. Since every pair of energy levels gives a separate contribution to the current it is enough to consider only one pair.

Generally a strong ac field leads to the generation of high harmonics. In Fig. 4 we plot the amplitude of the harmonics ($|I_L^n|$) versus their number n for (as an example) $W_L=W_R=30$, $\epsilon_1=11.5$, $\epsilon_2=30.5$, $\mu_R=20$, $\mu_L=5$, $D=70$, $\Gamma_{11}^{L(R)}=\Gamma_{22}^{L(R)}=\Gamma_{12}^{L}=-\Gamma_{12}^{R}=0.05$, $\omega=1$. We do not show the dc component (because its magnitude is much larger), nor the first harmonic (whose amplitude is also a few times larger than that of the others) since, anyhow, it is only a part

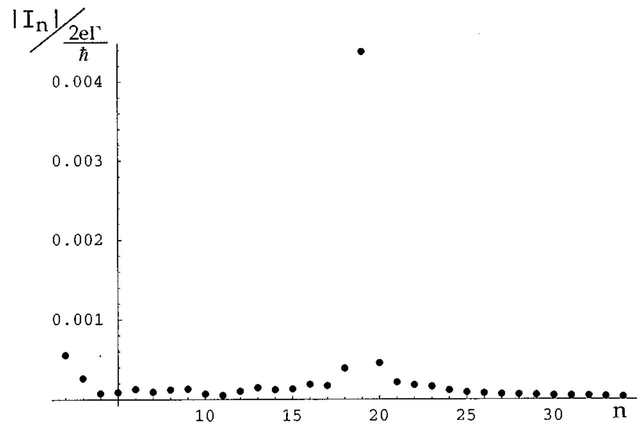

FIG. 5. Spectrum of the tunneling current (amplitude of the harmonics via their number) when the alternating field is applied on the right barrier: $W_L=0$, $W_R=30$, $\epsilon_1=11.5$, $\epsilon_2=30.5$, $\mu_R=20$, $\mu_L=5$, $D=70$, $\Gamma\equiv\Gamma_{11}^{L(R)}=\Gamma_{22}^{L(R)}=\Gamma_{12}^{L}=-\Gamma_{12}^{R}=0.05$, $\omega=1$. The dc and the first harmonic are not shown.

of the current response with the frequency ω. The other part is given by the capacitive currents [first terms in the formula (20)]. The main contribution to the broadened spectrum plotted in Fig. 4 is due to the direct photon-assisted transitions from the left lead to the central region and back described by the first term within the brackets in $A_j(n)$ [see formula (23)]. The terms with $j\neq j'$ arising from the presence of two or more levels together can lead to the generation of one solitary (and high) harmonic (see Fig. 5). In the rest of this section we describe the conditions required to observe one high harmonic alone and show its dependence on the parameters of the system.

Since the first term in $A_j(n)$ generates many harmonics it must be eliminated. The easiest way to do it is to apply the ac voltage only to the right barrier: $W_L=0$ (in fact it is enough to have $W_L\ll|\epsilon_2-\epsilon_1|$). Then $A_j(n)=0$, $n\neq 0$ because $J_s(W_L/\omega)=J_s(0)=0$, $s\neq 0$.

In Fig. 5 we plot the amplitude of the harmonics ($|I_L^n|$) versus their number n for $W_L=0$, $W_R=30$, $\epsilon_1=11.5$, $\epsilon_2=30.5$, $\mu_R=20$, $\mu_L=5$, $D=70$, $\Gamma_{11}^{L(R)}=\Gamma_{22}^{L(R)}=\Gamma_{12}^{L}=-\Gamma_{12}^{R}=0.05$, $\omega=1$. As before we do not plot here the dc current and the first harmonic. They are, of course, large compared with higher harmonics. We predict however that if W_R is made larger than the bandwidth in the right lead D they are reduced, and the amplitude of the resonant harmonic might be comparable to the magnitude of the dc current. It is clearly seen in this figure that among the high harmonics only the 19th one $\{n_{\rm res}=[(\epsilon_2-\epsilon_1)/\omega]=19\}$ is generated.

At $W_L=0$ and $\Gamma_1\approx\Gamma_2$ a simpler expression is easily obtained for the resonant harmonic:

$$I_L^{n_{\rm res}}\approx i^{-n_{\rm res}}\frac{2e}{\hbar}\frac{2\Gamma_{12}^{L}\Gamma_{21}^{R}}{\Gamma_1+\Gamma_2}\sum_{s=[(\epsilon_1-\mu_R)/\omega]}^{\infty}J_s\left(\frac{W_R}{\omega}\right)J_{s+n_{\rm res}}\left(\frac{W_R}{\omega}\right). \tag{25}$$

Only the terms with s so that the energy $\epsilon_1-s\omega$ is inside the Fermi sea in the right band, $\epsilon_1-s\omega<\mu_R$, contribute to the sum. We notice that $J_s(W_R/\omega)$ tends to zero when $|s|$ becomes larger than W_R/ω if $(W_R/\omega)\gg 1$.

In Fig. 6 we draw the dependence of the resonant harmonic ($|I_L^{n_{\rm res}}|$) on both (W_R/ω) (i.e., ac voltage) and $[(\epsilon_1-\mu_R)/\omega]$ (determined by the dc bias) for $n_{\rm res}=[(\epsilon_2-\epsilon_1)/\omega]=8$, $\Gamma_{11}^{L(R)}/\omega=\Gamma_{22}^{L(R)}/\omega=\Gamma_{12}^{L}/\omega=-\Gamma_{21}^{R}/\omega=0.05$, $\mu_R/\omega=20$. There is no generation ($I_L^{n_{\rm res}}=0$) if $W_R<n_{\rm res}\omega/2=(\epsilon_2-\epsilon_1)/2$ because the conditions $|s|<(W_R/\omega)$ and $|s+n_{\rm res}|<(W_R/\omega)$ cannot be satisfied together so one of $J_s(W_R/\omega)$, $J_{s+n_{\rm res}}(W_R/\omega)$ is small. The generation becomes significant for $W_R>\epsilon_2-\epsilon_1$. It is especially important if there are more than two energy levels: distant pairs of levels do not generate harmonics.

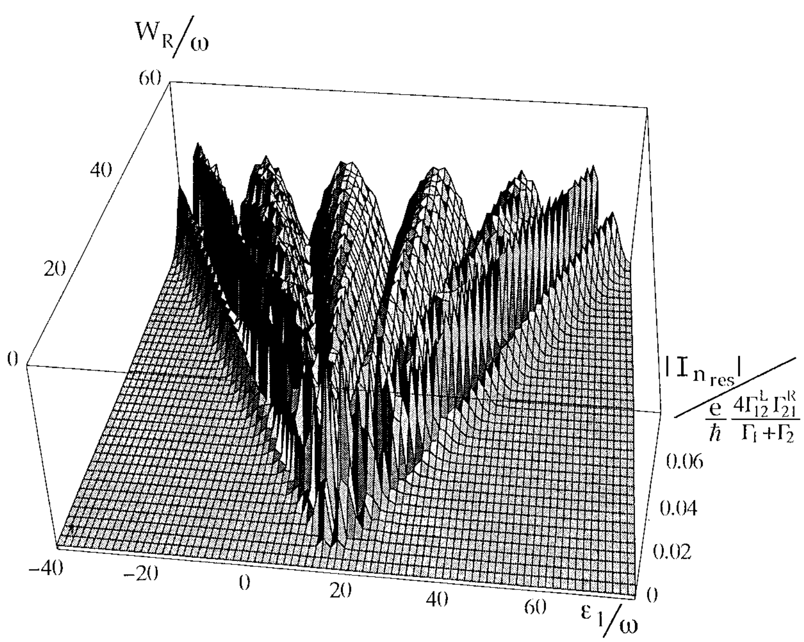

FIG. 6. Dependence of the resonant harmonic ($|I_L^{n_{\rm res}}|$) on the dc bias (i.e., $\epsilon_1-\mu_R$) and the amplitude of the ac voltage W_R. $n_{\rm res}=8$, $W_L=0$, $\epsilon_2=\epsilon_1+8$, $\mu_R=20$, $\Gamma_{11}^{L(R)}=\Gamma_{22}^{L(R)}=\Gamma_{12}^{L}=-\Gamma_{12}^{R}=0.05$, $\omega=1$.

The dependence on the bias (i.e., $[(\epsilon_1-\mu_R)/\omega]$) is oscillating. The reason is the interference of different components of the wave function (remember that in a strong ac field it is spread over a set of energies $\epsilon_p \pm s\omega$ with different s). It shows in the formulas as oscillating behavior of $J_s(W_R/\omega)$ via s. Let us consider the sum in Eq. (25) at $|\epsilon_1-\mu_R| \ll W_R$. It is useful to divide it into two parts:

$$\sum_{s=s_1}^{\infty} J_s\left(\frac{W_R}{\omega}\right) J_{s+n_{\mathrm{res}}}\left(\frac{W_R}{\omega}\right) = \sum_{s=s_1}^{s^*} J_s\left(\frac{W_R}{\omega}\right) J_{s+n_{\mathrm{res}}}\left(\frac{W_R}{\omega}\right) + \sum_{s=s_*+1}^{\infty} J_s\left(\frac{W_R}{\omega}\right) J_{s+n_{\mathrm{res}}}\left(\frac{W_R}{\omega}\right),$$

where $s_1 \equiv [(\epsilon_1-\mu_R)/\omega]$ and $|s^*| \ll W_R$. The second part does not depend on $\epsilon_1-\mu_R$. The Bessel's functions in the first part can be approximated by a trigonometric function:

$$J_s\left(\frac{W_R}{\omega}\right) = \sqrt{\frac{2}{\pi\nu}} \cos\left(\nu - s \arcsin\frac{\nu}{W_R/\omega} - \frac{\pi}{4}\right),$$

$$\nu \equiv \sqrt{\left(\frac{W_R}{\omega}\right)^2 - s^2},$$

since $|s| \ll W_R/\omega$. Then

$$I_L^{n_{\mathrm{res}}} \approx i^{-n_{\mathrm{res}}} \frac{2e}{\hbar} \frac{2\Gamma_{12}^L \Gamma_{21}^R}{\Gamma_1+\Gamma_2} \frac{1}{\pi W_R/\omega} \sum_{s=s_1}^{s=s^*} \left[\cos\left(\alpha - \frac{n_{\mathrm{res}}\omega}{W_R} s\right) + \cos(\beta - \pi s)\right] + \text{const}$$
$$\approx i^{-n_{\mathrm{res}}} \frac{2e}{\hbar} \frac{2\Gamma_{12}^L \Gamma_{21}^R}{\Gamma_1+\Gamma_2} \frac{1}{\pi n_{\mathrm{res}}} \sin\left(\alpha - n_{\mathrm{res}} \frac{\epsilon_1-\mu_R}{W_R}\right) (+\text{ const}), \tag{26}$$

where $\alpha \approx \omega n_{\mathrm{res}}^2/2W_R + \pi n_{\mathrm{res}}/2$ and $\beta \approx 2W_R/\omega - [\pi(n_{\mathrm{res}}+1)/2]$. When $\epsilon_1-\mu_R$ changes from $-W_R$ to W_R the value $I_L^{n_{\mathrm{res}}}$ oscillates about n_{res}/π times. The amplitude of the current $|I_L^{n_{\mathrm{res}}}|$ has about $2n_{\mathrm{res}}/\pi$ maxima (in Fig. 6 the number of maxima is even larger because the frequency of oscillations is larger at $\epsilon_1-\mu_R \approx \pm W_R$). Notice that the current is not generated if $|\epsilon_1-\mu_R| > W_R$. It is significant for systems with many energy levels: only those pairs of levels that are in the energy range from μ_R-W_R to μ_R+W_R generate harmonics.

The dependence of the current on the transparency of the barriers is clear from formulas (23), (25). In Fig. 5 we display the spectrum of the tunneling current at $\Gamma/\omega = 1/20$. If Γ increases two sets of harmonics grow in the vicinity of the 0th and the n_{res}th harmonics. The ratio of the amplitudes of the side harmonics to the amplitude of the leading (0th or n_{res}th) one is about $\Gamma/n\omega$ and $\Gamma/(n-n_{\mathrm{res}})\omega$, respectively. On the other hand, the magnitude of the resonant harmonic is proportional to $2\Gamma_{12}^L\Gamma_{21}^R/\Gamma_1+\Gamma_2$.

The dependence of the current on frequency is oscillatory. If a rectifying device like a diode is placed in the output the harmonics contribute to the dc. In Fig. 7 we draw the value $\langle I\rangle \equiv \frac{1}{2}|I_L^0| + (2/\pi)\Sigma_{n=1}^{+\infty}|I_L^n|$ coming from the time averaging of $I_L^T(t)$ as a function of the input frequency ω at $W_L=0$, $W_R=30$, $\epsilon_1=11.5$, $\epsilon_2=31.5$, $\mu_R=20$, $\mu_L=5$, $D=210$, $\Gamma_{11}^{L(R)}=\Gamma_{22}^{L(R)}=\Gamma_{12}^L=-\Gamma_{12}^R=0.05$. The peaks correspond to different harmonics ($n=22,21,20,19,18$) satisfying the resonant condition $n=[(\epsilon_2-\epsilon_1)/\omega]$.

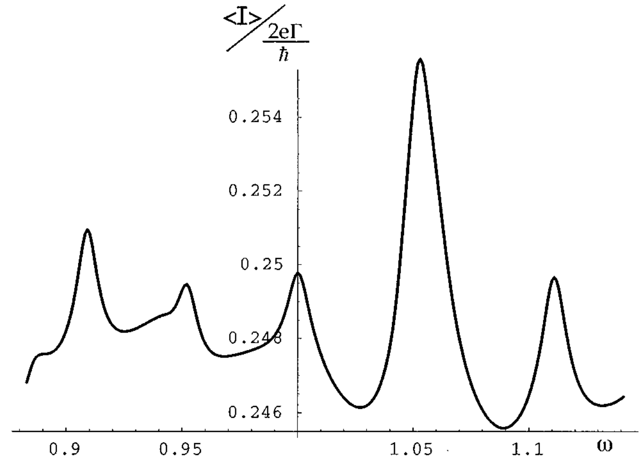

FIG. 7. The output current rectified by a diode versus the frequency ω of the input ac voltage. $W_L=0$, $W_R=30$, $\epsilon_1=11.5$, $\epsilon_2=31.5$, $\mu_R=20$, $\mu_L=5$, $D=210$, $\Gamma \equiv \Gamma_{11}^{L(R)}=\Gamma_{22}^{L(R)}=\Gamma_{12}^L=-\Gamma_{12}^R=0.05$. The peaks correspond to different harmonics ($n=22,21,20,19,18$) satisfying the resonant condition $n=[(\epsilon_2-\epsilon_1)/\omega]$.

We have discussed the high harmonic resonance in the tunneling current through the left barrier. To observe it the measuring device must be made sensitive to the tunneling current only through this barrier. Otherwise other harmonics will mask the resonance (like in Fig. 4). If the device measures the current in the leads we propose two ways to obtain it based on the formula (20).

(1) Making the gate capacitance small: $C_g \ll C_L$, C_R. The capacitance of the right barrier should be much larger than the capacitance of the left one: $C_L \ll C_R$. It can be achieved by making the left barrier thicker than the right one. Notice that the right barrier must be made slightly higher so that Γ_L and Γ_R are of the same order (then I_L^T and I_R^T are of the same order). Thus the high harmonic currents in the leads are approximately given by

$$I_L(t) \approx I_L^T(t) - \frac{C_L}{C_R} I_R^T(t) \approx I_L^T(t),$$

$$I_R(t) \approx -I_L^T(t) + \frac{C_L}{C_R} I_R^T(t) \approx -I_L^T(t).$$

We omitted here the contribution of the capacitive currents having frequency ω. The high harmonic currents in both leads are determined by I_L^T.

(2) Making the gate capacitance large. $C_g \gg C_L$ (the ratio of barrier capacitances C_L and C_R is arbitrary, there is no need to make one of the barriers higher than the other one). Then (dropping out the capacitive currents) we obtain from Eq. (20),

$$I_L(t)\approx I_L^T(t)-\frac{C_L}{C_R+C_g}I_R^T(t),$$

$$I_R(t)\approx -\frac{C_R}{C_R+C_g}I_L^T(t)+\frac{C_g}{C_R+C_g}I_R^T(t).$$

We notice that in this case high harmonics in the left lead are produced by I_L^T but in the right lead by both I_L^T and I_R^T (unless $C_R \gg C_g$). If $W_L=0$, $W_R\neq 0$ the term A generating a wide spectrum of harmonics vanishes in $I_L^T(t)$ but it is not zero in the expression for $I_R^T(t)$. Then the current in the right lead $I_R(t)$ consists of many harmonics. To detect a single harmonic the current in the left lead should be measured.

VI. SUMMARY

The basic physical problem addressed in this work is concerned with a nonlinear response of a two- (or more) level system to a high frequency external field. This makes the formalism developed above particularly attractive since two-level systems are an important model for many realistic physical situations. The most familiar one is evidently a two-level atomic system, whose response to a laser field is one of the hallmarks of nonlinear quantum optics, one of whose signatures is a higher frequency generation. Here we have focused on an electronic analog, where the response is a tunneling current instead of an emitted light. Generation of higher harmonics in this system occurs solely due to tunneling from the central region to the leads and back without direct transitions between the levels. Note that, unlike the optical analog, the theoretical formulation requires a computation of nonequilibrium (Schwinger-Keldysh) Green's functions. As far as the experimental situation is concerned, we see no real obstacle in the road for the actual observation of this effect.

We think that the next step should be a consideration of many-body physics in a strong time-dependent field. When the size of the dot is small enough it can be considered as an Anderson impurity. At voltages and frequencies less than the Coulomb interaction energy U (which is frequently the case) it can be described by the infinite-U Anderson model which is enough to exibite a Kondo type effect when the resonance level is deep below the Fermi level and the temperature is low enough. Hence, the first problem which comes into mind in this context is a nonlinear response of a magnetic impurity to a time-dependent field. The Kondo effect out of equilibrium has been studied recently by several authors, but this is done primarily in the noncrossing approximation, which is valid much above the Kondo temperature. At temperatures closer to the Kondo one, crossed diagrams should also be included (the first one appears when a sixth order term in the tunneling matrix elements is computed). Another interesting problem would be the study of interfierence between a strong external ac field and the Coulomb blockade effects. If the external voltages or frequencies are comparable with the Coulomb interaction energy mutual time-dependent resonant tunneling of electrons might show new physics. We suppose that this problem can be handled using the finite-U Anderson model.

ACKNOWLEDGMENTS

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APPENDIX A

In this appendix we show that an arbitrary strong time-dependent potential has no effect if it is uniform in space. We also calculate here Green's functions for an isolated lead.

If the alternating potential $(H_{\rm ac}(t))$ is uniform the Hamiltonian can be written in the following form:

$$H(t)=H_f+H_{\rm ac}(t), \tag{A1}$$

where H_f is time independent, $H_{\rm ac}(t)$ is space independent. The solutions of the Schrödinger's equation are

$$\Psi_k(t)=\varphi_k(t)e^{-(i/\hbar)\int^t H_{ac}(t_1)dt_1}, \tag{A2}$$

where $\varphi_k(t)$ are eigenstates of H_f. The time-dependent part $H_{\rm ac}(t)$ influences only the phases giving all the solutions *the same* phase factor $e^{-(i/\hbar)\int^t H_{\rm ac}(t_1)dt_1}$. The phase differences are determined by H_f only. Thus $H_{\rm ac}(t)$ has no physical effect. This simple result is the time-dependent analog of the fact that the (time-independent) reference point of energy can be arbitrarily chosen.

We used this result to write the ac part of the Hamiltonian of a double-barrier structure in the form (4). The ac shift of the central region is ignored.

In an isolated lead the ac potential is uniform [see Eq. (4)]. Then the evolution operator $(U(t,t'))$ is obtained from the equation $i\hbar(\partial/\partial t)U(t,t')=H(t)U(t,t')$ with $H(t)$ given by Eq. (A1). Its matrix elements are

$$U_{kk'}(t,t')=\delta_{kk'}e^{-(i/\hbar)\int_{t'}^{t}H_{kk}(t_1)dt_1}. \tag{A3}$$

$\delta_{kk'}$ appears because $H_{kk'}=0, k\neq k'$ (i.e., a uniform potential does not cause transitions). Using the definition of Green's functions in the Schrödinger representation with the evolution operator (A3) it is easy to obtain the formulas (13).

APPENDIX B

In this appendix we analyze the Dyson's equation for $G_{nm}^<$ and obtain the formula (11). The Dyson's equation (7) holds for $G_{nm}^<$:

$$G_{nm}^<=F_{nm}^1(g_{jj}^<)+F_{nm}^2(g_{kk}^<,g_{pp}^<), \tag{B1}$$

$$\begin{aligned}F_{nm}^1(g_{jj}^<)\equiv{}& g_{nm}^<+G_{nk}^rT_{km}g_{mm}^<+G_{np}^rT_{pm}g_{mm}^<+g_{nn}^<T_{nk}G_{km}^a\\&+g_{nn}^<T_{np}G_{pm}^a+G_{nk}^rT_{ki}g_{ii}^<T_{ik'}G_{k'm}^a\\&+G_{np}^rT_{pi}g_{ii}^<T_{ip'}G_{p'm}^a+G_{nk}^rT_{ki}g_{ii}^<T_{ip}G_{pm}^a\\&+G_{np}^rT_{pi}g_{ii}^<T_{ik}G_{km}^a,\end{aligned}$$

$$F^2_{nm}(g^<_{kk},g^<_{pp}) \equiv G^r_{nn'}T_{n'k}g^<_{kk}T_{km'}G^a_{m'm} + G^r_{nn'}T_{n'p}g^<_{pp}T_{pm'}G^a_{m'm}.$$

Here multiplication implies integration over time and the summation over repeated indexes; $j=m,n,i$ belong to the central region, k,k' belong to the left lead, p,p' belong to the right lead.

$F^1_{nm}(g^<_{jj})$ depends on the initial state of the central region. It does not depend on $g^<_{kk},g^<_{pp}$ [G^r, G^a are determined by Eq. (6) which does not depend on $g^<_{kk},g^<_{pp}$]. It describes the behavior of the system with empty leads. Indeed, if $g^<_{kk}=0$, $g^<_{pp}=0$ we have $F^2_{nm}=0$. Then $G^<_{nm}=F^1_{nm}$. When the leads are empty the central region empties with time. F^1_{nm} tends to zero. After a long enough period of time has passed from the moment the tunneling was switched on it can be neglected. Then $G^<_{nm}=F^2_{nm}$. This is the formula (11). We can say that F^1_{nm} describes the transient processes while F^2_{nm} gives some kind of a quasistationary (but fully time-dependent) solution.

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