

Nonlinear dynamical response of a double-barrier resonant-tunneling structure

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The dynamical response of a double-barrier resonant-tunneling structure (DBRTS) on external ac bias $V(t)$ is studied. The rate equation for the occupation number of the level in the DBRTS well as well as the conditions required for this equation are derived in the case of coherent tunneling. The analysis of the equation in question provides the possibility to obtain easily the analytic results for the current through the structure in all limiting cases of interest.

I. INTRODUCTION

There is a great deal of current interest in electron transport through double-barrier resonant-tunneling structures (DBRTS's). Besides other applications such structures have been used as generators¹ and detectors² of microwave radiation, which have stimulated theoretical investigations of DBRTS behavior under time-dependent perturbations.

An attempt to study the dynamical properties of DBRTS's has been reported in the paper by Stone *et al.*³ The authors of Refs. 4 and 5 have discussed the DBRTS interaction with infrared radiation, when the perturbation frequency is comparable with the characteristic interlevel spacing in the well $\xi \sim 10$ meV, while in Refs. 6 and 7 the linear dynamical response of DBRTS to an external bias has been studied (note that applicability of the linear response approximation in such a case requires the condition $eV_0 \ll \hbar/\tau_{\text{well}}$, where V_0 is the characteristic voltage amplitude, and τ_{well} is the characteristic electron lifetime in the well). We should also mention the S -matrix approach to time-dependent resonant tunneling due to Coon and Liu.⁸

At the same time there exist a number of papers⁹⁻¹¹ where nonstationary tunneling through a DBRTS has been discussed with the main attention to the nonlinear regime. In these papers calculation of the change in the electron transmission probability through the structure in question resulting from an ac-voltage component across the structure is presented. In the papers by Sokolovski^{9,10} the time- and space-dependent Schrödinger equation has been analyzed, while in Johansson's treatment¹¹ the tunneling Hamiltonian approach has been used. It was shown that the physical picture of resonant tunneling significantly depends on the value of the ratio $eV_0/\hbar\omega$, where ω is the characteristic frequency of the external ac voltage. If $eV_0/\hbar\omega \gg 1$, the DBRTS response has been shown to be strongly nonlinear and to have a Fourier spectrum of multiple-line form, while in the opposite case $eV_0/\hbar\omega \ll 1$ the transmission probability has been shown to exhibit peaks at integer values of $(\epsilon_{\text{in}} - \epsilon_{\text{w}})/\hbar\omega$, where ϵ_{in} and ϵ_{w} are the initial electronic energy and the energy of the level in the well, respectively.

Unfortunately, direct usage of these results appears to have serious difficulties mainly due to the fact that the exact formulas obtained for the transmission probability are rather complicated. Evaluation of the limiting cases of interest must be done with numerical calculations⁹⁻¹¹ even in such an intuitively simple case as the adiabatic one ($eV_0/\hbar\omega \gg 1, \omega\tau_{\text{well}} \ll 1$). In the present paper it will be shown that under some not too restrictive conditions the semiclassical rate equation for the distribution function of electrons in the well can be derived. This equation can be solved analytically for most of the limiting cases of interest, which provides us with the possibility to obtain rather simple expressions for physical observables like current, etc., instead of the complicated expressions obtained in Refs. 9-11, requiring numerical analysis.

In the next section, a more detailed description of the system in question will be given. Then, the rate equation for particles in the well of a DBRTS in the presence of a time-dependent external signal will be discussed. The full derivation of the rate equation in question using the Keldysh nonequilibrium Green's function technique is performed in the Appendix. In Sec. III a detailed study of the DBRTS nonlinear response to an external signal will be presented for the semiclassical case $eV_0/\hbar\omega \gg 1$. Simplified equations for the electron distribution function and current will be derived and analyzed for adiabatic ($\omega\tau_{\text{well}} \ll 1$) and antiadiabatic ($\omega\tau_{\text{well}} \gg 1$) cases separately, while in Sec. IV the multiphoton case $eV_0/\hbar\omega \ll 1$ will be studied. In Sec. V final remarks on the problem in question will be given.

II. GENERAL DESCRIPTION AND RATE EQUATION FOR PARTICLES IN THE WELL

We will derive the rate equation for electrons in the well using the tunneling Hamiltonian method, which provides a sufficiently accurate and rather transparent description of the tunneling process. We will restrict ourselves to the condition $\hbar\omega/\xi \ll 1$, which allows us to consider the levels in the well independently; for simplicity we will take into account only one level of lateral quantization inside the well. Electron-electron interaction of the particles in the well¹² will be neglected as well. In the absence of external bias, the Hamiltonian for our system

takes the form:

$$H = H_{L,R}^0 + H_W^0 + \sum_{pq} T_{pq} (c_{pL}^\dagger a_q + a_q^\dagger c_{pL} + c_{pR}^\dagger a_q + a_q^\dagger c_{pR}), \quad (1)$$

where

$$H_{L,R}^0 = \sum_p (\varepsilon_{pL} c_{pL}^\dagger c_{pL} + \varepsilon_{pR} c_{pR}^\dagger c_{pR}), \quad (2)$$

$$H_W^0 = \sum_q \varepsilon_{qW} a_q^\dagger a_q.$$

Here the subscripts L, R indicate electrons in left and right electrodes respectively; c_p^\dagger, c_p denote the creation and annihilation operators for electrons in the electrodes; a_q^\dagger, a_q are the same operators for electrons in the well; T_{pq} is the matrix element of the tunneling Hamiltonian ($T_{pq} = T_{pq}^*$).

If the period of the ac signal is larger than the characteristic time scale of onset of electron equilibrium in the electrodes, one can describe the bias $V(t)$ as the difference of electron chemical potentials for the left and right electrodes (all energies and corrections to the chemical potentials will be related to the value of the unperturbed electrode chemical potential μ_0):

$$\mu_{L(R)}(t) = +(-) \frac{eV_0}{2} \cos(\omega t). \quad (3)$$

In what follows we will assume that

$$(eV_0)^2 \ll \frac{U_0 \hbar^2}{m_e d^2},$$

which provides the possibility to neglect the direct effect of the electric field on the barrier (here U_0 is the characteristic height of the barrier, d the barrier width, m_e and e the electron mass and charge, respectively). So the bias can be accounted for by introducing the term

$$H'(t) = \sum_p [\mu_L(t) c_{pL}^\dagger c_{pL} + \mu_R(t) c_{pR}^\dagger c_{pR}] \quad (4)$$

into the Hamiltonian (1).

At first it seems to be useful to emphasize the impor-

tant role of the parameter $eV_0/\hbar\omega$ (see, e.g., Ref. 10). Let us write the Schrödinger equation for the electron wave function $f_W(\mathbf{q}, t)$ in the well, which follows directly from Eqs. (1),(2),(4):

$$\left[i\hbar \frac{\partial}{\partial t} - H_W^0(\mathbf{q}) \right] f_W(\mathbf{q}, t) = \sum_p T_{pq} [f_R(\mathbf{p}, t) + f_L(\mathbf{p}, t)]. \quad (5)$$

In the lowest perturbation-theory approximation one can consider wave functions of electrons in the electrodes f_R and f_L [defined by equations analogous to (5) but without the right-hand side] to be proportional to

$$f_{L(R)}(\mathbf{p}, t) \propto \exp \left[-\frac{i}{\hbar} \varepsilon_{pL(R)} t + \frac{i}{\hbar} \int_0^t \mu_{L(R)}(t_1) dt_1 \right]$$

$$= \exp \left[-\frac{i}{\hbar} \varepsilon_{pL(R)} t + (-) i \frac{eV_0}{2\hbar\omega} \sin(\omega t) \right].$$

One can easily see that the physical picture of resonant tunneling depends significantly on the value of $eV_0/\hbar\omega$. In the case $eV_0/\hbar\omega \ll 1$, the influence of the external ac voltage results in only small time-periodic corrections to the phase of $f_{L(R)}(\mathbf{p}, t)$, while in the case $eV_0/\hbar\omega \gg 1$, the phase exhibits strong rapid oscillations, which leads to a nonlinear behavior. In this case, in order to solve Eq. (5) one might use a procedure similar to that used in semiclassical mechanics.

In view of the fact that the distribution function of particles in the well is far from its equilibrium Fermi form, in order to derive the rate equation for electrons in the well it is convenient to use the nonequilibrium Green's function technique developed by Keldysh¹³ for strongly nonequilibrium processes (for a review see, e.g., Refs. 14 and 15; one should also mention the paper by Caroli *et al.*¹⁶ where the *tunneling* problem has been considered in the nonequilibrium Green's function approach). The full derivation of the rate equation in question is given in the Appendix; here we will put down the resulting equation for the distribution function n_W of electrons in the well:

$$\frac{\partial n_W(t^\dagger, \mathbf{q})}{\partial t^\dagger} = \sum_{n=-\infty}^{+\infty} \left[J_n \left[\frac{2\mu_L(t^\dagger)}{\hbar\omega} \right] + J_n \left[\frac{2\mu_R(t^\dagger)}{\hbar\omega} \right] \right] \frac{2\pi}{\hbar} \sum_p |T_{pq}|^2 [n(\varepsilon_p) - n_W(t^\dagger, \mathbf{q})] \delta(\varepsilon_p - \varepsilon_{qW} - n\omega/2). \quad (6)$$

This equation can be used under the semiclassical conditions discussed in detail at the end of the Appendix.

Now we will consider separately the semiclassical ($eV_0/\hbar\omega \gg 1$) and multiphoton ($eV_0/\hbar\omega \ll 1$) cases.

III. THE SEMICLASSICAL CASE

In this case ($eV_0/\hbar\omega \gg 1, \varepsilon_{qW}/\hbar\omega \gg 1$), the direct evaluation of Eq. (6) appears to be extremely complicated

and requires numerical analysis. However, as is shown in the Appendix, the rate equation can be substantially reduced by taking into account the semiclassical form of the electron Green's function in the leads [see Eq. (A22)]. The resulting reduced rate equation appears to be rather simple:

$$\frac{\partial n_W(t, \mathbf{q})}{\partial t} = \Gamma_L(t, \mathbf{q}) [n_L(t, \varepsilon_{qW}) - n_W(t, \mathbf{q})] + \Gamma_R(t, \mathbf{q}) [n_R(t, \varepsilon_{qW}) - n_W(t, \mathbf{q})]. \quad (7)$$

Here $\Gamma_{L(R)}(t, \mathbf{q})$ is determined by the equation,

$$\Gamma_{L(R)}(t, \mathbf{q}) = \frac{2\pi}{\hbar} \sum_{\mathbf{p}} |T_{\mathbf{pq}}|^2 \delta(\varepsilon_{\mathbf{q}W} - \varepsilon_{\mathbf{p}L(R)} + \mu_{L(R)}(t)), \quad (8)$$

n_L and n_R being the electron distribution functions in the electrodes.

Note that Eq. (7) can be easily shown to be valid not only for the case of a harmonic ac voltage $V(t) \propto \cos(\omega t)$ but for a time-dependent signal $V(t)$ of any type [the general equation (6) is *not* valid for nonharmonic external signal]. The only constraint is that the characteristic amplitude of this signal should be much greater than the level broadening Γ [see Eq. (A21)].

Equation (7) was derived in the absence of any additional intrawell scattering and thus in the case of coherent tunneling. At the same time the analogous equation can obviously be obtained in the case of sequential tunneling as well,¹⁷ using simple probability considerations. Thus we have shown that within the constraints of the semiclassical rate-equation approach there is *no difference between coherent and sequential tunneling processes*, at least for such experimentally measurable quantities as level distribution function and current through the DBRTS. (In the static case the same result has been pointed out by Weil and Vinter.¹⁹) The origin of such an effect appears to be clear if we remember that the semiclassical conditions used imply the classical character of $L \leftrightarrow W$, $R \leftrightarrow W$ transition, which are determined only by particle transition probabilities (not phases). This can be understood from (7).

A. Static case

At first we will rederive well-known results for the stationary situation, when the chemical potentials

$$\mu_L = \frac{eV_0}{2}, \quad \mu_R = -\frac{eV_0}{2} \quad (9)$$

do not depend on time (see, e.g., Ref. 18). In this case the rate equation takes the form,

$$n_W(\mathbf{q}) = \frac{n_R(\varepsilon_{\mathbf{q}W})\Gamma_R(\mathbf{q}) + n_L(\varepsilon_{\mathbf{q}W})\Gamma_L(\mathbf{q})}{\Gamma_R(\mathbf{q}) + \Gamma_L(\mathbf{q})}. \quad (10)$$

$$n_W(\varphi) = \frac{\Gamma}{\omega} \exp\left[-\frac{2\Gamma}{\omega}\varphi\right] \int_0^\varphi d\varphi_1 \exp\left[\frac{2\Gamma}{\omega}\varphi_1\right] \left[\theta\left[\cos\varphi_1 - \frac{2\varepsilon_W}{eV_0}\right] + \theta\left[-\cos\varphi_1 - \frac{2\varepsilon_W}{eV_0}\right] \right]. \quad (17)$$

Here $\theta(x)$ is the theta function.

1. Adiabatic case $\Gamma \gg \omega$

Let us consider the case of fast tunneling, when the characteristic electron lifetime in the well $\tau_{\text{well}} \sim \Gamma^{-1}$ is much smaller than ω^{-1} . For convenience we will introduce two characteristic phases:

$$\varphi_L = \arccos\left[\frac{2\varepsilon_W}{eV_0}\right], \quad (18)$$

$$\varphi_R = \arccos\left[-\frac{2\varepsilon_W}{eV_0}\right]. \quad (19)$$

An expression for the current through the structure can be obtained in a similar way; the result is

$$j_R = \sum_{\mathbf{q}} e\Gamma_R(\mathbf{q})[n_R(\varepsilon_{\mathbf{q}W}) - n_W(\mathbf{q})] \quad (11)$$

$$j_L = \sum_{\mathbf{q}} e\Gamma_L(\mathbf{q})[n_W(\mathbf{q}) - n_L(\varepsilon_{\mathbf{q}W})]. \quad (12)$$

In the static case,

$$j_R = j_L = \sum_{\mathbf{q}} e \frac{\Gamma_R(\mathbf{q})\Gamma_L(\mathbf{q})}{\Gamma_R(\mathbf{q}) + \Gamma_L(\mathbf{q})} [n_R(\varepsilon_{\mathbf{q}W}) - n_L(\varepsilon_{\mathbf{q}W})]. \quad (13)$$

B. Nonstatic case

In our further analysis we will consider resonant tunneling through a quantum dot (or two-dimensional quantum well in the presence of a strong perpendicular magnetic field), so we will not take into account electron motion in the plane of the barrier. In this case, electrons inside the well are characterized by the same energy ε_W . Neglecting the $\Gamma_{L(R)}$ dependence on \mathbf{q} and assuming $\Gamma_L = \Gamma_R \equiv \Gamma$, we obtain equations for the level occupation number $n_W(t)$ and currents in the form,

$$\dot{n}_W(t) + 2\Gamma n_W(t) = \Gamma[n_L(\varepsilon_W, t) + n_R(\varepsilon_W, t)], \quad (14)$$

$$j_R(t) = e\Gamma[n_R(\varepsilon_W, t) - n_W(t)], \quad (15)$$

$$j_L(t) = e\Gamma[n_W(t) - n_L(\varepsilon_W, t)]. \quad (16)$$

We would like to note that in order to clarify the analysis we will study the equations for j_L and j_R separately, despite the fact that according to the Ramo-Scheckley theorem the experimentally measurable current is $j = (j_L + j_R)/2$.

Introducing the phase $\varphi = \omega t$ and considering for simplicity the case of zero temperature, one obtains the solution of Eq. (14), remembering the initial condition $n_W(\varphi=0) = 0$,

Having in mind the initial condition $n_W(\varphi=0)=0$, we obtain the following results for the dynamical DBRTS response in this case (compare with Ref. 7):

	$0 \leq \varphi \leq \varphi_L$	$\varphi_L \leq \varphi \leq \varphi_R$
$n_W(\varphi)$	$\frac{1}{2} \left[1 - \exp \left[-\frac{2\Gamma}{\omega} \varphi \right] \right]$	$\frac{1}{2} \exp \left[-\frac{2\Gamma}{\omega} \varphi \right] \left[\exp \left[\frac{2\Gamma}{\omega} \varphi_L \right] \right]^{-1}$
$j_L(\varphi)$	$\frac{e\Gamma}{2} \left[1 + \exp \left[-\frac{2\Gamma}{\omega} \varphi \right] \right]$	$-e\Gamma n_W(\varphi)$
$j_R(\varphi)$	$e\Gamma n_W(\varphi)$	$e\Gamma n_W(\varphi)$

The physical meaning of this is very transparent. At the time $t=0$, the chemical potential in the left electrode becomes larger than ε_W , while the level in the well is empty. This causes a current through the left electrode $j_L(t=0)$ equal to $e\Gamma$, while $j_R(t=0)=0$. On the time scale $t \sim \Gamma^{-1}$, the level occupation number approaches the "constant" value $n_W \sim [n_R(\varepsilon_W) + n_L(\varepsilon_W)]/2 \sim 1/2$, while j_R becomes close to j_L , $|j_R| \approx |j_L| \approx e\Gamma/2 \equiv j^s$. After the time $t = \varphi_L/\omega$, both μ_L and μ_R are lower than ε_W , and the level occupation starts to relax. At the time $t = \varphi_R/\omega$, the chemical potential μ_R crosses ε_W and the cycle of response is repeated. The time-dependent current behavior is shown in Fig. 1. The presence of

discontinuities in the $j_{L(R)}(t)$ dependence is related to the fact that in our approach we have omitted terms proportional to $\hbar\Gamma/eV_0$. The discontinuities can be smeared by including $\hbar\Gamma/eV_0$ terms or by considering the case of finite temperatures.

2. Antiadiabatic case $\Gamma \ll \omega$

In this case the bias time dependence is much more rapid than the characteristic time of the tunneling process, so during one period the level occupation number n_W changes by only a small fraction, while on a time scale $\sim \Gamma^{-1}$ it begins to fluctuate near some plateau value \bar{n}_W :

$$\bar{n}_W = \lim_{\varphi \rightarrow \infty} \left[\frac{2\Gamma}{\omega} e^{-(2\Gamma/\omega)\varphi} \sum_{n=1}^{I(\varphi/2\pi)} \int_{-\varphi_L+2\pi n}^{\varphi_L+2\pi n} d\varphi_1 \exp \left[\frac{2\Gamma}{\omega} \varphi_1 \right] \right] = \frac{\varphi_L}{\pi}, \quad (20)$$

where $I(x)$ means the integer part of x . Having in mind that $\varphi_L \leq \pi/2$ one can obviously see that \bar{n}_W is always smaller than its "constant" value $1/2$.

At the same time, the currents are determined by the equation,

$$j_{L(R)}(t) \approx e\Gamma [n_{L(R)}(\varepsilon_W, t) - \bar{n}_W]. \quad (21)$$

It is useful also to put down here the expressions for the Fourier spectrum of the DBRTS frequency response.

$$n_W(k) = \frac{2}{\pi k} \sin(k\varphi_L) \frac{\Gamma}{2\Gamma - i\omega k} \quad \text{for even } k \text{ values,}$$

$$n_W(k) = 0 \quad \text{for odd } k \text{ values,}$$

$$j_L(k) = \frac{e\Gamma}{\pi k} \sin(k\varphi_L), \quad j_R(k) = j_L(k) \quad \text{for odd } k \text{ values,} \quad (22)$$

$$j_L(k) = \frac{-i\omega k}{2\Gamma - i\omega k} \frac{e\Gamma}{\pi k} \sin(k\varphi_L), \quad j_R(k) = -j_L(k) \quad \text{for even } k \text{ values.}$$

It is clear that if $\varphi_L \ll 1$, odd- k current harmonics have approximately the same amplitudes up to $k \sim \varphi_L^{-1}$, while even- k harmonics do not vanish up to $k \sim \Gamma/\omega$.

In principle another experimental situation may be of interest, when the level in the well is perturbed by the external signal

$$\varepsilon_W(t) = \varepsilon_W^{(0)} + eV_0 \cos(\omega t),$$

while the difference between electron chemical potentials in the electrodes is held to be independent of time,

$$\mu_L - \mu_R = eV_1 \gg eV_0.$$

In such situation one can easily find the Fourier spectrum of the DBRTS response also:

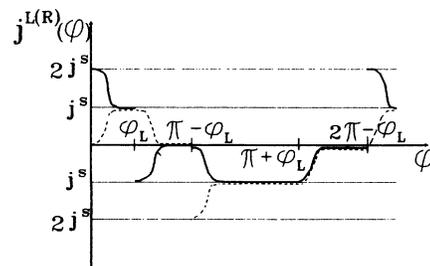


FIG. 1. Time dependence of currents j_L (solid line) and j_R (dashed line) in the adiabatic case.

$$\begin{aligned}
n_W(k) &= \frac{\Gamma}{2\Gamma - i\omega k} \frac{\sin(k\varphi_L)}{\pi k}, \\
j_L(k) &= \frac{e\Gamma}{\pi k} \sin(k\varphi_L) \frac{\Gamma - i\omega k}{2\Gamma - i\omega k}, \\
j_R(k) &= e\Gamma n_W(k).
\end{aligned} \tag{23}$$

IV. MULTIPHOTON CASE

Let us consider the case $\hbar\omega \gg eV_0$. In this case the equations for the distribution functions and currents can be reduced to (ε_W is assumed to be less than zero):

$$\dot{n}_W + 2\Gamma n_W(t) = \Gamma \sum_{n=-\infty}^{n_{\max}} \left[J_n \left(\frac{2\mu_L(t)}{\hbar\omega} \right) + J_n \left(\frac{2\mu_R(t)}{\hbar\omega} \right) \right] n(\varepsilon_W + n\omega/2), \tag{24}$$

$$j_{R(L)}(t) = +(-)e\Gamma \left[\sum_{n=-\infty}^{n_{\max}} J_n \left(\frac{2\mu_{R(L)}(t)}{\hbar\omega} \right) n(\varepsilon_W + n\omega/2) - n_W(t) \right], \tag{25}$$

where $n_{\max} = -I(2|\varepsilon_W|/\hbar\omega)$. This equation requires the condition $\omega \gg \Gamma$ [see Eq. (49)]. Now we will discuss two limiting cases.

(1) $|\varepsilon_W| \ll \omega$. Taking into account only the terms proportional to J_0 and J_1 one can easily see that the $L \rightarrow W$ and $W \rightarrow R$ transition probabilities are equal to each other, so

$$n_W \approx 1 \tag{26}$$

$$j_L \approx j_R \approx \frac{e^2\Gamma}{2\hbar\omega} V_0 \cos(\omega t), \tag{27}$$

and we obtain the linear-response picture. One should have in mind that this is not a trivial linear response; it is a linear response using dressed states responding at ω . In this case the linearity is of another type than that discussed in Refs. 6 and 7. Here it occurs due to a frequency large with respect to the bias amplitude, while in Refs. 6 and 7 it occurs due to a large level broadening \hbar/τ_{well} .

(2) $|\varepsilon_W| \gg \hbar\omega$. Having in mind the well-known relations for the Bessel functions:

$$\sum_{n=-\infty}^{+\infty} J_n(z) = 1, \tag{28}$$

we obtain

$$\begin{aligned}
\sum_{n=-\infty}^{n_{\max}} J_n \left(\frac{2\mu_{L(R)}(t)}{\hbar\omega} \right) &\approx 1 + \left[\frac{\hbar\omega}{|\varepsilon_W|} \right]^{1/2} \left[\frac{|\mu_{L(R)}(t)|}{|\varepsilon_W|} \right]^{n_{\max}} \\
&= 1 + \alpha,
\end{aligned} \tag{29}$$

so the level occupation number $n_W \approx 1$, while the currents are very small, $j_{L(R)} \sim e\Gamma\alpha \ll e\Gamma$.

V. CONCLUDING REMARKS

In summary, the nonequilibrium Green's function technique has been exploited in order to analyze the time-dependent behavior of a double-barrier resonant-tunneling structure. The semiclassical rate equation for the particles in the well has been derived in the case of small level broadening; the conditions which are required

for the equation in question have been evaluated. It has been shown that within the constraints of the semiclassical rate-equation approach there is no difference between coherent and sequential tunneling processes, at least for such experimentally measurable quantities as level distribution function and current through the DBRTS (in the static case a similar effect has been pointed out by Weil and Vinter¹⁹). A further detailed analysis of the rate equation in question has been presented.

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APPENDIX: DERIVATION OF THE RATE EQUATION

We will follow the notation and Feynman rules introduced in the review paper, Ref. 14. In the Appendix we are working with units where $\hbar=1$. In order to derive the rate equation we shall use at first the time-coordinate representation for operators. One should mention that such coordinates do not correspond to physical DBRTS coordinates. Final equations will be formulated in terms of physical observables. Let us take $\psi_q^{W(L,R)}(t, \mathbf{r})$ to be the exact solutions of the Schrödinger equation with the Hamiltonian $H_{W(L,R)}^0$, which are extended into the whole space (for definiteness one can extend $\psi_q(\mathbf{r})$ in a periodic or antiperiodic manner). Thus the Green's functions $G_W^{\alpha\beta}(t_1, \mathbf{r}_1, t_2, \mathbf{r}_2)$ of electrons in the well take the form:

$$\begin{aligned}
G_W^{\alpha\beta}(t_1, \mathbf{r}_1, t_2, \mathbf{r}_2) &= \sum_{p_1, p_2} G_W^{\alpha\beta}(t_1, \mathbf{p}_1, t_2, \mathbf{p}_2) \\
&\times \psi_{p_1}^{*W}(t_1, \mathbf{r}_1) \psi_{p_2}^W(t_2, \mathbf{r}_2);
\end{aligned}$$

the equations for the other Green's functions can be obtained in an analogous way. Note that we do not assume homogeneity of the electronic density in the well.

Let us write the Dyson's equation for the exact Green's function $G_W^{-+}(X_1, X_2)$ of electrons in the well:

$$G_W^{-+}(X_1, X_2) = G_W^{(0)-+}(X_1, X_2) + \int d^4 X_3 d^4 X_4 \\ \times \sum_{\alpha, \beta = \{+, -\}} G_W^{(0)-\alpha}(X_1, X_3) \\ \times \Sigma^{\alpha\beta}(X_3, X_4) G_W^{\beta+}(X_4, X_2). \quad (\text{A1})$$

Here $G_W^{(0)-+}(X_i, X_j)$ is the Green's function of electrons in the well in the absence of tunneling; $X_i \equiv (t_i, \mathbf{r}_i)$; $\Sigma^{\alpha\beta}(X_i, X_j)$ is the self-energy term:

$$\Sigma^{\alpha\beta}(X_i, X_j) = -T(X_i) [G_L^{\alpha\beta}(X_i, X_j) \\ + G_R^{\alpha\beta}(X_i, X_j)] T^*(X_j). \quad (\text{A2})$$

Note that Eq. (A1) can be written in an alternative way:

$$G_W^{-+}(X_1, X_2) = G_W^{(0)-+}(X_1, X_2) + \int d^4 X_3 d^4 X_4 \\ \times \sum_{\alpha, \beta = \{+, -\}} G_W^{-\alpha}(X_1, X_3) \\ \times \Sigma^{\alpha\beta}(X_3, X_4) G_W^{(0)\beta+}(X_4, X_2). \quad (\text{A3})$$

Acting on Eqs. (A1) and (A3) with the operators $[i\partial/\partial t_1 - H_W^0(\mathbf{r}_1)]$ and $[-i\partial/\partial t_2 - H_W^0(\mathbf{r}_2)]$, respectively, and subtracting the second resulting equations from the first one, we obtain

$$\left[i \frac{\partial}{\partial t_1} + i \frac{\partial}{\partial t_2} \right] G_W^{-+}(X_1, X_2) + [H_W^0(\mathbf{r}_2) - H_W^0(\mathbf{r}_1)] G_W^{-+}(X_1, X_2) \\ = \int d^4 X_3 \sum_{\alpha = \{+, -\}} [\Sigma^{-\alpha}(X_1, X_3) G_W^{\alpha+}(X_3, X_2) + G_W^{-\alpha}(X_1, X_3) \Sigma^{\alpha+}(X_3, X_2)]. \quad (\text{A4})$$

Here we have used the well-known relation for the free-particle Green's functions:¹⁴

$$\left[i \frac{\partial}{\partial t_1} - H_W^0(\mathbf{r}_1) \right] G_W^{(0)-+}(X_1, X_2) = \delta(X_1 - X_2). \quad (\text{A5})$$

Now we will examine the energy representation in Eq. (A4), taking into account the semiclassical character of the time dependence of the Green's functions. Let us introduce the new variable $t^+ \equiv (t_1 + t_2)/2$ and consider the typical term from the rhs of Eq. (A4), taking $t^- \equiv t_1 - t_2 = 0$,

$$\int dt_3 G_W^{\alpha\beta} \left[\mathbf{r}_1, \mathbf{r}_2, \frac{t^+ + t_3}{2}, t_3 - t^+ \right] \Sigma^{\beta\gamma} \left[\mathbf{r}_1, \mathbf{r}_2, \frac{t^+ + t_3}{2}, t_3 - t^+ \right] \approx \int dt_3 G_W^{\alpha\beta}(\mathbf{r}_1, \mathbf{r}_2, t^+, t_3 - t^+) \Sigma^{\beta\gamma}(\mathbf{r}_1, \mathbf{r}_2, t^+, t_3 - t^+). \quad (\text{A6})$$

The latter transformation is valid under the condition $|t_3 - t^+| \sim 1/\bar{\epsilon} \ll \Gamma^{-1}$, where $\bar{\epsilon} \sim \epsilon_W$ is the characteristic electronic energy. In the cases where such a condition is not met, it appears to be impossible to derive a simple semiclassical rate equation for the DBRTS dynamical response (see, e.g., Ref. 7) and the equation for the distribution function n_W becomes an integral-differential equation with respect to energy. Note that for different relations between the frequency and amplitude of the external signal, the semiclassical condition in question corresponds to different necessary relations between them and the level broadening Γ [see Eqs. (A20) and (A21)].

Now one can examine the representation of quantum numbers \mathbf{p} and \mathbf{q} of the Hamiltonian (2). Having in mind that in the absence of any processes except tunneling all Green's functions are diagonal in this representation, we obtain as a result:

$$-\frac{\partial n_W(t^+, \mathbf{q})}{\partial t^+} = \int \frac{d\epsilon}{2\pi} [G_W^{+-}(t^+, \mathbf{q}, \epsilon) \Sigma^{-+}(t^+, \mathbf{q}, \epsilon) \\ - G_W^{-+}(t^+, \mathbf{q}, \epsilon) \Sigma^{+-}(t^+, \mathbf{q}, \epsilon)], \quad (\text{A7})$$

where

$$n_W(t^+, \mathbf{q}) = -i \int \frac{d\epsilon}{2\pi} G_W^{+-}(t^+, \mathbf{q}, \epsilon) \quad (\text{A8})$$

is the semiclassical distribution function of electrons in the well. Here we have used the well-known relations for the exact Green's functions and self-energies (see, e.g., Ref. 14):

$$\Sigma^{++} + \Sigma^{--} = -(\Sigma^{-+} + \Sigma^{+-}), \quad (\text{A9})$$

$$G_W^{++} + G_W^{--} = G_W^{-+} + G_W^{+-}. \quad (\text{A10})$$

Let us calculate now the retarded Green's functions $G_L^{L(R)}$ for the electrons in the electrodes in Wigner's representation. Making use of the expressions for the wave functions of electrons in the leads (see discussion of the Schrödinger equation (5) in Sec. II, we have

$$G_{L(R)}^r(t^+, \mathbf{p}, \varepsilon) = i \int_0^\infty dt^- \exp \left[i(\varepsilon - \varepsilon_{pL(R)} + i\eta)t^- + \frac{2i\mu_{L(R)}(t^+)}{\omega} \times \sin(\omega t^- / 2) \right], \quad (\text{A11})$$

where η is a positive infinitesimal.

Having in mind the well-known relation

$$e^{iz \sin \rho} = \sum_{n=-\infty}^{+\infty} J_n(z) e^{in\rho}, \quad (\text{A12})$$

where $J_n(z)$ is the Bessel function, we obtain simultaneously for the retarded Green's functions:

$$G_{L(R)}^r(t^+, \mathbf{p}, \varepsilon) = \sum_{n=-\infty}^{+\infty} J_n \left[\frac{2\mu_{L(R)}(t^+)}{\omega} \right] \times \frac{1}{\varepsilon - \varepsilon_{pL(R)} + n\omega/2 + i\eta}, \quad (\text{A13})$$

whereas for the kinetic Green's functions we have

$$G_{L(R)}^{-+}(t^+, \mathbf{p}, \varepsilon) = 2\pi i \sum_{n=-\infty}^{+\infty} n_{L(R)}(t^+, \mathbf{p}) \times J_n \left[\frac{2\mu_{L(R)}(t^+)}{\omega} \right] \times \delta(\varepsilon - \varepsilon_{pL(R)} + n\omega/2), \quad (\text{A14})$$

$$G_{L(R)}^{+-}(t^+, \mathbf{p}, \varepsilon) = 2\pi i \sum_{n=-\infty}^{+\infty} [n_{L(R)}(t^+, \mathbf{p}) - 1] \times J_n \left[\frac{2\mu_{L(R)}(t^+)}{\omega} \right] \times \delta(\varepsilon - \varepsilon_{pL(R)} + n\omega/2), \quad (\text{A15})$$

where n_L and n_R are the electron distribution functions in the electrodes:

$$n_{L(R)}(\varepsilon) = \frac{1}{1 + e^{(\varepsilon - \mu_{L(R)})\Theta}}. \quad (\text{A16})$$

Here, Θ^{-1} is the lattice temperature.

In what follows we will work in the lowest perturbation-theory approximation, which allows us to use free-particle expressions for the electron Green's

functions in the electrodes in calculating the self-energy term:

$$\Sigma^{\alpha\beta}(t^+, \mathbf{q}, \varepsilon) = \Sigma_R^{\alpha\beta}(t^+, \mathbf{q}, \varepsilon) + \Sigma_L^{\alpha\beta}(t^+, \mathbf{q}, \varepsilon) = - \left[\sum_{\mathbf{p}} |T_{\mathbf{p}\mathbf{q}}|^2 G_R^{\alpha\beta}(t^+, \mathbf{q}, \varepsilon) + \sum_{\mathbf{p}} |T_{\mathbf{p}\mathbf{q}}|^2 G_L^{\alpha\beta}(t^+, \mathbf{q}, \varepsilon) \right], \quad (\text{A17})$$

and also to take in the rhs of Eq. (A7) the expressions for the Green's function in the well in the absence of tunneling. In the absence of tunneling, the electron wave function in the well is determined by the stationary Schrödinger equation with the Hamiltonian H_W^0 [see Eq. (2)], which leads us to the corresponding expressions for the Green's functions:¹⁴

$$G_W^{(0)-+}(t^+, \mathbf{q}, \varepsilon) = 2\pi i n_W(t^+, \mathbf{q}) \delta(\varepsilon - \varepsilon_{qW}), \quad (\text{A18})$$

$$G_W^{(0)+-}(t^+, \mathbf{q}, \varepsilon) = 2\pi i [n_W(t^+, \mathbf{q}) - 1] \delta(\varepsilon - \varepsilon_{qW}). \quad (\text{A19})$$

Here one should note that the use of delta functions in Eqs. (A18) and (A19) requires the additional condition that the characteristic energy $\varepsilon - \varepsilon_{pL(R)}$ (for those values close to the energy spectrum be much larger than Γ . In the case $\omega/eV(t) \gg 1$ such a condition is equivalent to

$$\varepsilon - \varepsilon_{pL(R)} \sim \omega \gg \Gamma, \quad (\text{A20})$$

[see Eqs. (A14),(A15)], while in the case $\omega/eV(t) \ll 1$ such a condition is equivalent to

$$\varepsilon - \varepsilon_{pL(R)} \sim |eV(t)| \gg \Gamma. \quad (\text{A21})$$

[see Eq. (A22)]. Now one can easily obtain the rate equation in the final form (6), assuming the electron dispersion laws in left and right electrodes to be the same ($\varepsilon_{pL} = \varepsilon_{pR} \equiv \varepsilon_p$).

In the semiclassical case ($eV_0/\omega \gg 1, \varepsilon_{qW}/\omega \gg 1$), in order to simplify the further analysis, we will reduce the rate equation (6) by taking into account the semiclassical slow character of the time dependence of the external signal. One can show that, neglecting terms of the order of $\sqrt{\omega/eV_0}$, the integral over all possible values of the momentum \mathbf{p} used in (A17) is determined by the usual semiclassical Green's functions with a time-dependent shift of the chemical potential. So one can take the retarded Green's function of the electron in the electrode in the form

$$G_{L(R)}^r(t^+, \mathbf{p}, \varepsilon) = \frac{1}{\varepsilon - \varepsilon_p + \mu_{L(R)}(t^+) + i\eta}, \quad (\text{A22})$$

(instead of A13) while the reduced rate equation takes the form (7).

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