Linear and nonlinear resonant tunneling through a minimal-size resonant structure in quantum molecular wires

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Electron tunneling in a molecular wire with defects of the type $\cdots AAABAAA\cdots$, where electronic states of molecules, host (A) and guest, at different sites are coupled via resonant interaction, is investigated. The model described by a simple Hamiltonian in the tight-binding representation bears at the same time a close resemblance to a double-barrier resonant heterostructure. Resonant tunneling is considered in the presence of an external electric field with the Hubbard electron-electron interaction at the guest site included. A closed set of equations, which accounts for the feedback between the current density in the wire and the resonance-state energy, is derived and used for calculations of the transmission coefficient of tunneling through defect. The latter determines the wire conductance of a completely degenerate Fermi gas. In the linear version of the theory presented, the Sautet-Joachim result is rederived. With nonlinear effects taken into account, we come to a self-consistent equation coinciding formally with that of the Davydov-Ermakov phenomenological model of bistable tunneling in double-barrier structures. In both studies the external field was absent, and thus, with its inclusion, an important generalization of the previous results is made. In the linear case (small current densities), an analytic expression of the wire-conductance field dependence is obtained and investigated. In the nonlinear case, the bistable regime of tunneling is shown to be operative, resulting in a specific behavior of the current response to the applied field. This behavior, i.e., the place of the hysteresis loop in the I-V curve, its form, and the loop number, correlates strongly with microscopic details of the resonant structure. An analysis of possible manifestations of bistability under varying magnitude of the applied bias constitutes the main result of the paper.

I. INTRODUCTION

Fast progress in the fabrication of submicrometer electronic devices has given a powerful impulse for studies of fundamental properties of electron transport in ultrasmall structures, where the quantum nature of charge carriers is of principal importance. Among the phenomena that gained renewed interest from both experimentalists and theoreticians is resonant tunneling, the wide spectrum of applications of which is nearly impossible to cover.

A fascinating property of this phenomenon is its intrinsic bistability predicted first[1,2] and then confirmed experimentally.[3,4] The origin of the bistable behavior of the electron flux transmitted through a resonant structure lies in the interaction of electrons stored in a quantum well with outgoing electrons. As a result of this interaction, feedback between the current through the resonance structure and the resonance-state energy arises, resulting in the bistable current-voltage characteristics observed in experiments.

Different approaches have been proposed to describe the resonant tunneling intrinsic bistability.[2,5-8] The Davydov-Ermakov theory[2] is based on the assumption that electrons entering the well are acted on by the average charge accumulated between the barriers. The structure of the charge buildup is ignored and, as a consequence of this simplification, the energy of an electron in the well turns out to be linearly dependent on the square amplitude of the wave function of the incident electrons. This effect provides the following bistable characteristics: transmitted flux intensity versus incident flux intensity. Note that the external electric field, which enlarges considerably the difficulties of an analytical treatment, was not taken into account (and therefore, many questions remain unanswered).

Bistable current-voltage characteristics were obtained by Sheard and Toombs[5] and then, in a somewhat more accurate manner, by Rahman and Davies[6] using the "sequential" model of resonant tunneling. A serious disadvantage of the otherwise reasonable model cited is that one of two stable values of the charge in the well is always zero, which in reality cannot be true.

Simulations of quantum transport in resonant structures[6-8] are of course a proper way to solve the problem but they give little insight into the nature of intrinsic bistability. So, we believe that further development of the microscopic theory of nonlinear resonant tunneling is a current problem.

In this paper we consider a model of the electron tunneling in a resonant structure of minimal size conceived on the molecular-level scale. Specifically, it deals with a quantum molecular wire, composed of identical host molecules (or other subunits), with a single guest molecule regarded usually as a traplike or barrierlike defect. The guest molecule, under certain conditions, acts as a double-barrier resonant structure for an electron flux propagating along the wire. Such a model was recently proposed and investigated as a prospective switching element of molecular size by Sautet and Joachim,[9] who con-

sidered the linear regime of tunneling of neutral particles (no electromagnetic-field effects were included). We extend their model to study the influence of the Coulomb electron-electron repulsion at the guest site and of the external electric field.

Intrinsic bistability of the resonance tunneling process is another problem that falls into focus here. There have been some attempts to treat it theoretically but, as mentioned, within the phenomenological or computational approach only. Therefore, the mechanism of the formation of the bistable state at the microscopic level remains, so far, hidden. Our model of bistable resonance tunneling allows us, with minimal simplifications, to investigate the phenomenon at a microscopic level and to estimate the significance of various parameters that determine the current-voltage characteristics when the system is found in a bistable or in a near-bistable state. This is evidently important for design of digital molecular electronics devices.

The remainder of the paper is organized as follows: In Sec. II we introduce our model and the method of calculation, in Sec. III we present and discuss our results, and Sec. IV contains our summary and conclusions.

II. STATEMENT OF THE MODEL AND METHOD OF CALCULATION

The physical reasons why a specific defect of a molecular chain, otherwise ideal, may behave like a resonant structure, are as follows. Let an electron move along a chain due to the exchange electron-electron interaction (with its energy denoted below by L) between the host molecules. The electron transfer energy between host and guest molecules is $\bar{L}$ and the energy of an electron on the host (guest) site is zero ($\mathcal{E}_0$). If $|\bar{L}|<|L|$, an electron is acted on by the regions between host and guest molecules as potential barriers and by the host molecular site, with $\mathcal{E}_0<0$, as by a potential well. Therefore, it is not surprising that such molecular structure can play a role similar to the well-known double-barrier resonant heterostructures.

The analogy between the two models mentioned becomes visible in Fig. 1, where a molecular wire with the "one-molecular" resonant structure and a semiconductor quantum wire with a single quantum dot sandwiched between two tunneling barriers are shown. In Fig. 1(b), the left and the right parts of the wire are regarded usually as the emitter and the collector leads, respectively. Using this terminology in treating the tunneling process seems to be appropriate also for Fig. 1(a). In what follows we refer to the latter only, bearing in mind that the results obtained are, to some extent, relevant to Fig. 1(b).

With the extension of the simple model of the quantum wire formulated above to include Coulomb electron-electron interaction at the guest site in the Hubbard representation, the electronic Hamiltonian of the system considered reads as

$$H=\sum_{n,m}[L_{n,m}-\Theta(n-1)\delta_{n,m}\Phi]a^{\dagger}_{n,\sigma}a_{m,\sigma}+\mathcal{E}_0 a^{\dagger}_{0,\sigma}a_{0,\sigma}+\mathcal{E}_H a^{\dagger}_{0\uparrow}a_{0\uparrow}a^{\dagger}_{0\downarrow}a_{0\downarrow}\ , \tag{1}$$

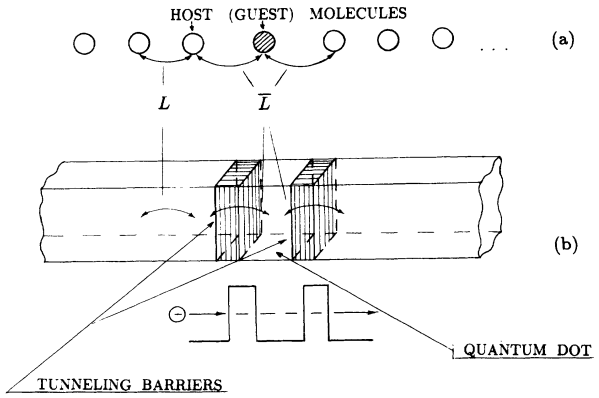

FIG. 1. Schematic representation of molecular (a) and semiconductor (b) quantum wires with tunneling resonant structures. For the molecular wire the meaning of L and $\bar{L}$ is explained in the text. For the semiconductor, quantum wire L has the same meaning as in the tight-binding representation of the electronic Hamiltonian in the wire. $\bar{L}$ is an effective energy of electron transfer between the wire and one of the bound states of the quantum dot. The usual two-barrier energy profile shown below is related to the barrier-dot-barrier resonant structure only.

where

$$L_{n,m}=[L+(\bar{L}-L)(\delta_{n,0}+\delta_{m,0})](\delta_{n,m+1}+\delta_{n,m-1})\ ,$$

$$\Theta(n)=\begin{cases}1, & n\geq 0\\ 0, & n<0\end{cases},$$

$\Theta(n)$ is the Heaviside step function, $a^{\dagger}_{n,\sigma}$, $a_{n,\sigma}$ are the Fermi operators of creation and annihilation of an electron at the site n ($|n|=0,1,\ldots$), with the projection of spin $\sigma=\uparrow,\downarrow$. The shift of the energy of an electron in the right ($n>0$) semichain, Φ, is introduced to study the external electric-field effect on the electron motion in the molecular wire. The last (Hubbard) term of Eq. (1) describes the Coulomb repulsion of electrons at the zeroth (guest) site. The energy of this interaction is denoted by $\mathcal{E}_H$.

Note that the assumption concerning the emitter-to-collector electric potential difference without changes of the resonant structure is somewhat artificial. We concentrate on this aspect of the electric-field effect (i.e., on the influence of the asymmetry of leads in the field) disregarding the shift of the defect-state energy. The latter can easily be accounted for by adding to $\mathcal{E}_0$ a term proportional to Φ.

Among other shortcomings of the model introduced, perhaps the most serious is that the electric-potential difference is assumed to be fixed. In reality, the potential drop across the defect site is dependent on the current intensity through the defect (due to an extra charge accumulated in the guest site) and, therefore, should be treated self-consistently. Instead, we use the Hubbard term, which accounts for the potential-to-current dependence very roughly. A wide discussion of the problem (which can be traced to the problem of the treatment of the

emitter and of the collector leads) with the relevant literature references is given in Ref. 6.

The most general description of the system is given by the matrix elements of the nonequilibrium density matrix operator $\rho(t)$,

$$\rho^{\sigma}_{n,n'}(t)=\mathrm{Tr}\rho(t)a^{\dagger}_{n,\sigma}a_{n',\sigma} ,$$

$$\rho^{\sigma\sigma'}_{n,n';m,m'}(t)=\mathrm{Tr}\rho(t)a^{\dagger}_{n,\sigma}a_{n',\sigma}a^{\dagger}_{m,\sigma'}a_{m',\sigma'} ,\ldots .$$

With the Hamiltonian (1) the quantities $\rho^{\sigma}_{n,n'}(t)$ are subjected to the equations

$$i\hbar\frac{d}{dt}\rho^{\uparrow}_{n,n'}(t)=-\sum_m [L_{mn}\rho^{\uparrow}_{m,n'}(t)-L_{mn'}\rho^{\uparrow}_{n,m}(t)]$$
$$+[\Theta(n-1)-\Theta(n'-1)]\Phi\rho^{\uparrow}_{n,n'}(t)[\mathcal{E}_0\rho^{\uparrow}_{n,n'}(t)+\mathcal{E}_H\rho^{\uparrow\downarrow}_{n,n';0,0}(t)](\delta_{n,0}-\delta_{n',0}) . \quad (2)$$

For the two-particle density matrix in Eq. (2) one can write equations which contain the three-particle density matrix and so on. To get a closed set of equations we make use of the mean-field approximation

$$\rho^{\uparrow\downarrow}_{n,n';0,0}(t)\approx\rho^{\uparrow}_{n,n'}(t)\rho^{\downarrow}_{0;0}(t) . \quad (3)$$

With Eq. (3) and in the absence of a magnetic field,

$$\rho^{\uparrow}_{n,n'}(t)=\rho^{\downarrow}_{n,n'}(t)=\rho_{n,n'}(t) ,$$

Eq. (2) can be rewritten as follows:

$$i\hbar\frac{d}{dt}\rho_{n,n'}(t)=-\sum_m [L_{mn}\rho_{m,n'}(t)-L_{mn'}\rho_{n,m}(t)]$$
$$+[\Theta(n-1)-\Theta(n'-1)]\Phi\rho_{n,n'}(t)-[\mathcal{E}_0+\mathcal{E}_H\rho_{0,0}(t)](\delta_{n,0}-\delta_{n',0})\rho_{n,n'}(t) . \quad (4)$$

The solution of Eq. (4) is readily found in the form[10]

$$\rho_{n,n'}(t)=\psi^{*}_{n}(t)\psi_{n'}(t) , \quad (5)$$

where the wave function $\psi_n(t)$ satisfies

$$i\hbar\frac{d}{dt}\psi_n(t)=\sum_m L_{mn}\psi_m(t)$$
$$+[\mathcal{E}_0-\Theta(n-1)\Phi+\mathcal{E}_H|\psi_0(t)|^2]\delta_{n,0}\psi_n(t) . \quad (6)$$

Here we are interested in a stationary solution of Eq. (6),

$$\psi_n(t)=\exp\left[-\frac{iE_k}{\hbar}t\right]A_n , \quad (7)$$

$$A_n=\begin{cases} I\exp(ikn)+R\exp(-ikn) , & n\le -1 \\ T\exp(ik'n) , & n\ge 1 , \end{cases}$$

which describes the transmitted (T) and reflected (R) electronic waves under the condition of a stationary incident flux of electrons with the given amplitude I and the (dimensionless) wave vector k or, equivalently, with the fixed energy of electrons incoming from $-\infty$. For the model of the molecular wire at hand this energy is

$$E_k=2L\cos k .$$

Then, the wave vector k' of the outgoing electrons in the right semichain is defined by the equation

$$E_{k'}=E_k+\Phi \quad (7a)$$

and amplitudes R, T, and A_0 are subjected to the equations

$$E_kL^{-1}A_0=\alpha(A_1+A_{-1})+(V_0+V_H|A_0|^2)A_0 ,$$
$$E_kL^{-1}A_{-1}=\alpha A_0+A_{-2} , \quad (8)$$
$$(E_kL^{-1}+\Phi/L)A_1=\alpha A_0+A_2 ,$$

where $\alpha=\bar{L}/L$ and $V_{0(H)}=\mathcal{E}_{0(H)}/L$.

Thus, under the assumption that approximation (3) is valid, we obtain the equations describing the equilibrium tunneling of electrons through defects from the emitter lead, which is under zero potential, to the collector lead, which is under potential Φ. The nonlinear term in these equations accounts, in a self-consistent manner, for the feedback between the defect-state energy and the incident-flux intensity.

The solution of Eqs. (8) gives

$$T=\frac{iI}{\mathcal{A}+\frac{i}{2}\left[1+\frac{\sin k'}{\sin k}\right]} , \quad (9)$$

$$R=-iT\left[\mathcal{A}+\frac{i}{2}\left[1-\frac{\sin k'}{\sin k}\right]\right] , \quad (10)$$

$$A_0=\alpha^{-1}T , \quad (11)$$

where

$$\mathcal{A}=\frac{2(\alpha^2V_0+|T|^2V_H)+\alpha^4\Phi/L}{4\alpha^4\sin k}-(\alpha^{-2}-1)\cot k . \quad (12)$$

In accordance with Eqs. (9) and (10), to find the

transmission coefficients

$$\beta_T=(|T|^2/|I|^2)(\sin k'/\sin k)\ ,$$
$$\beta_R=|R|^2/|I|^2\ , \tag{13}$$

we have to solve the equation

$$X_I(k)=\mathcal{F}(X_T(k,\Phi))\ , \tag{14}$$

where

$$\mathcal{F}(X_T)=X_T\{[a(k,\Phi)-X_T]^2+b(k,\Phi)\}\ , \tag{15}$$

$$\begin{Bmatrix} X_I(k) \\ X_T(k,\Phi) \end{Bmatrix}=\frac{|V_H|}{2\alpha^4\sin k}\begin{Bmatrix} |I|^2 \\ |T|^2 \end{Bmatrix}\ , \tag{16}$$

$$a(k,\Phi)=\ \text{sgn}\ (-L)\left[\frac{2\alpha^2V_0+\alpha^4\frac{\Phi}{L}}{4\alpha^4\sin k}-(\alpha^{-2}-1)\cot k\right]\ , \tag{17}$$

$$b(k,\Phi)=\frac{1}{4}\left[1+\left[1-\frac{\Phi}{L}\frac{4\cos k+\frac{\Phi}{L}}{4\sin^2 k}\right]^{1/2}\right]^2\ . \tag{18}$$

A similar equation was obtained by Davydov and Ermakov[2] in their study of nonlinear tunneling based on a phenomenological treatment of charge accumulation in the well of a double-barrier resonant structure in the absence of the external field.

There is only one solution of Eq. (14), when

$$a\le a(k_{\rm cr})=\sqrt{3b(k_{\rm cr})} \tag{19}$$

and, if $a>a(k_{\rm cr})$, there are three solutions in the range of the incident flux amplitude defined by

$$\mathcal{F}(X_T^+)=X_I^{\min}<X_I(k)<X_I^{\max}=\mathcal{F}(X_T^-)\ , \tag{20}$$

where

$$X_T^{\pm}=\tfrac{2}{3}a\pm\sqrt{a^2-3b} \tag{21}$$

and, again, one solution when X_I is out of the intensity range indicated. From the three possible solutions, which are given by the intersections of the two functions $Y(X_T)=\mathcal{F}(X_T)$ and $Y(X_T)=$const, only two (where $\partial\mathcal{F}/\partial X_T>0$) are stable, giving bistable behavior of the transmitted flux. We proceed with a detailed discussion of this effect in the next section.

III. RESULTS AND DISCUSSION

A. Linear tunneling

Before analyzing the nonlinear dependencies of the transmission coefficient, let us consider tunneling in the absence of electron-electron interaction at the guest site. When $V_H=0$, β_T and β_R are completely defined by Eqs. (9) and (10), which for $\Phi=0$ reduce to

$$\beta_T=\frac{1}{\left[(1-\alpha^{-2})\cot k+\frac{V_0}{2\alpha^2\sin k}\right]^2+1}\ ,$$
$$\beta_R=\beta_T\left[(1-\alpha^{-2})\cot k+\frac{V_0}{2\alpha^2\sin k}\right]^2\ , \tag{22}$$

and coincide with the result obtained by Suatet and Joachim[9] (the inter-relation between their and our notations is $X\Longleftrightarrow V_0$, $Y=W\Longleftrightarrow\alpha$, $Z=1, q\Longleftrightarrow 2\cos k$).

In the effective-mass approximation, $k\to 0$, and for $\alpha=1$ Eqs. (22) are the handbook formulas for the transmission and reflection coefficients for the case when a particle is tunneling through a δ-function barrier $U\delta(x)$,

$$\beta_T=\frac{1}{1+\left[\frac{m^*U}{\hbar^2 q}\right]^2}\ ,$$
$$\beta_R=\beta_T\left[\frac{m^*U}{\hbar^2 q}\right]^2\ . \tag{23}$$

In Eqs. (23) $U=\mathcal{E}_0 a$ is the barrier intensity (height $\times$ width), the effective mass of the tunneling particle is

$$m^*=-\frac{\hbar^2}{2La^2}\ ,$$

and $q=k/a$ is its wave vector with k being any possible rational number; a is the lattice constant of the molecular wire.

The most important difference between Eqs. (22) and (23) is that in the former case the condition of resonant tunneling, $\beta_R=0$, can be fulfilled. The resonant value of the wave vector is defined by the equation

$$\cos k_r=\frac{V_0}{2(1-\alpha^2)}\ . \tag{24}$$

We mention here that a guest molecule is associated with a trap (well) for an electron when $\mathcal{E}_0<0$, and with a scattering center (barrier) when $\mathcal{E}_0>0$, and that Eq. (24) may have solutions for both signs of $\mathcal{E}_0$.

Since the k (energy) dependence of β_T was already discussed in detail,[9] we restrict ourselves to a few additional remarks.

The position of the resonant peak depends on V_0 and α but its half-width depends on α only, see Fig. 2. The peak is very sharp, when $|\alpha|\ll 1$, which is intuitively expected. For example, for $k\ll 1$ we have near the resonance

$$\beta_T(k)=\frac{1}{1+\alpha^{-4}(k-k_r)^2} \tag{25}$$

(V_0 is close to 2 in this case). When $|\alpha|>1$, the resonance, if it exists, is very wide. Thus, to get small values of α in a chemical realization of the molecular wire considered is the main necessary condition for providing a molecular size structure that can act effectively as a

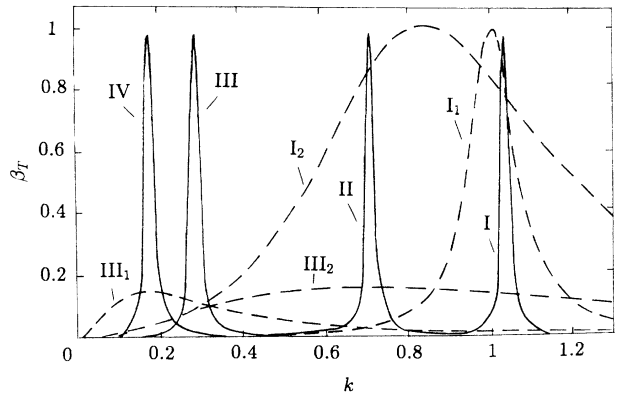

FIG. 2. Transmission coefficient $\beta_T(\Phi=0)$ as a function of the wave vector k for different values of α and V_0: curves I, II, III, IV correspond to $V_0=1,1.5,1.9,1.95$, and $\alpha=0.1$ ($k_r=1.04,0.711,0.285,0.174$, respectively). The subscripts 1,2 refer to the values $\alpha=0.25$ and 0.5. The dashed curves illustrate that there is quite a difference in dependence on the guest-host coupling parameter α between the resonances near and distant to the band edge.

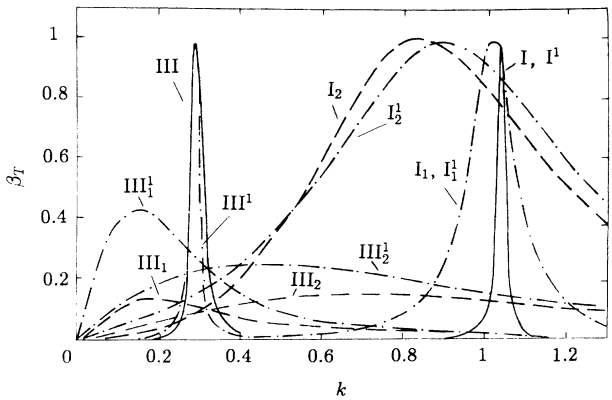

FIG. 3. Transmission coefficient β_T for the electrostatic potential $\Phi=0$, $\Phi/|L|=0.5$; $L<0$. The $\beta_T(k)$ curves are the same as on Fig. 2 except for the numbers with the superscript 1, which refer to $\Phi/|L|=0.5$. Curves I, I^1 and I_1, I_1^1 are undistinguished in the scale given.

switching element. Note that the resonance peak is much more sensitive to the value of α near the band edges: the resonance disappears faster with an increase of α for larger values of V_0; compare curves that correspond to $V_0=1$ and $V_0=1.9$ on Fig. 2. This difference in the resonance peak behavior with variation of α (which can be important for designing a switching molecular device) is directly connected with the conditions of the existing in-band state. At larger values of V_0 these conditions break down at smaller values of $|\alpha|$.[9] Further discussion of this point as well as a comprehensive analysis of $\beta_T(k,\alpha,V_0)$ dependencies can be found in Ref. 9.

The appropriately averaged electronic transmission coefficient through the defect is directly related to the one-dimensional conductance controlled by isolated defects.[11–13] Therefore, the dependence of this coefficient on the electric potential is of principal interest and will now be considered.

Note at first that, in accordance with our model, the electric field affects the system by shifting the defect site energy and making the right and the left leads asymmetric. Since the former effect expressed in the $\beta_T(V_0)$ dependence has, as mentioned above, already been studied, we focus our attention on the latter effect, i.e., on the symmetry loss, assuming hereafter that $\mathcal{E}_0$ is constant.

The electric field destroys the symmetry of the system and, when $\Phi\neq 0$, the resonance does not exist, i.e., $R\neq 0$ at any energy of the incident electrons, see Eq. (10). This is a well-known field effect on the resonant tunneling through a double-barrier structure[14] but, contrary to that case, the dependence of the maximum and of the half-width of the transmission coefficient on the electrostatic potential is rather weak here, see Figs. 3 and 4. In a sense, this result is not surprising because the electric field does not affect the resonant structure itself (the parameters V_0 and α remain unchanged). On the other hand, it was not evident *a priori* that the violation of the symmetry caused by the electric-potential difference between the left and the right semichains (i.e., between the emitter and the collector leads) would have as small effect, as turned out to be the case. Moreover, due to a δ-like dependence of β_T on k for $\Phi=0$, Eq. (25), one could expect that even a small effective shift of k_r in the

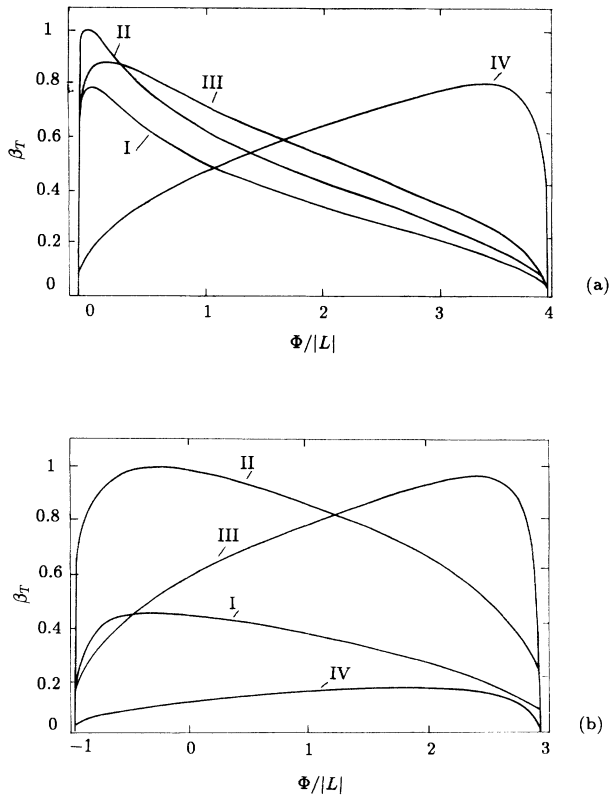

FIG. 4. The dependencies of transmission coefficient $\beta_T(\Phi)$ in and near the resonances $k_r=0.285$ (a) and $k_r=1.04$ (b). Curves I, II, III, IV correspond to $k=0.28$, 0.285, 0.29, 0.31 (a) and $k=1.03,1.04,1.05,1.07$ (b).

electric field would result in a considerable change of the transmission coefficient, at least near the resonance. But our calculations show that this is not the case.

As is seen on Fig. 4, β_T has, as a function of Φ, a maximum but, contrary to the k dependence (Fig. 3), the function $\beta_T(\Phi)$ is smooth for almost all values of Φ except those close to the limiting ones,

$$\Phi_{\pm}=2L\,(1\pm\cos k)\ . \tag{26}$$

The latter correspond to the maximum field-induced shifts, down (+) and up (−), of the right-semichain band relative to the left one, at which Eq. (7a) can still be fulfilled.

The precise form of the $\beta_T(\Phi)$ curves is determined by an interplay of the two main factors. The first one is the density of final states in the vicinity of k', which provides that $\beta_T(\Phi)$ goes to zero whenever Φ goes to Φ_+ or to Φ_-. This factor is evidently dominant when k corresponds to the energy near the band edges [as in the case of the curves in Fig. 4(a), which drop rapidly with an increase of the potential of the negative sign]. The second factor of importance depends on whether the electric field enhances or suppresses tuning between the tunneling electron and the defect-induced resonant state. Its role can be especially pronounced when the wave vector k is far from the resonant value. Taking as an example curves IV [Fig. 4(a)] and III [Fig. 4(b)], for which $k>k_r$, one can see a noticeable increase in β_T with an increase of Φ, when Φ is still far from Φ_-. This can be explained in terms of tuning improvement due to the effective decrease of the tunneling-electron energy under the condition that the resonant level remains unchanged. Contrary to this, curve I [Fig. 4(b)] demonstrates the opposite dependence on Φ, because, when $k<k_r$, the electric potential of the positive sign suppresses tuning for the same reason.

Note that the tendency in the dependence of $\beta_T(\Phi)$ outlined contradicts intuitive expectations. Indeed, for the double-barrier resonant structure shown in Fig. 1(b) the positive potential difference between the left and the right leads will result in a lowering of the resonant energy and, therefore, suppress tuning in the case $k>k_r$. However, this effect should be compared with that just discussed above. The net result cannot be predicted in advance but what can obviously be stated is that accounting for the electric field through the shift of the resonant energy only, as is often done, is not sufficient for the relevant estimate of its role in the resonant tunneling.

The above discussion relates to the energy-dependent transmission coefficient, which, under certain conditions, determines the wire conductance[15,16]

$$\sigma\sim\beta_T(E_F,\Phi)\ , \tag{27}$$

where E_F is the energy of the Fermi level. One of the necessary conditions for relation (27) to be valid is $\Phi\ll E_F$, but the fulfillment of the inequality indicated does not guarantee that the conductance of the system is field independent. If, for instance, E_F turns out to be near the band edges, then the conductance may vary drastically even in the response of small changes of the electric potential applied. This is especially the case when the resonant level is near the band bottom, i.e., when $E_F\approx E_{k_r}\ll 1$ and the effective mass is positive. Under such conditions the one-molecular resonant structure can work effectively as a diode, see Fig. 4(a). At the same time it is evident that this model cannot work as a switching element, the "on" or "out" state being controlled by an external electric field applied along the chain.

B. Nonlinear tunneling

In the discussion of nonlinear effects, just as in all our model calculations, the positive effective mass ($L<0$) is implied. In this case the system may be bistable for $k>k_{\rm cr}>k_r$ only, with $k_{\rm cr}$ defined by Eq. (10). Bistability is more pronounced, in the sense that the difference $X_I^{\max}-X_I^{\min}$ is larger, for larger values of k; compare curves I, I′, I″ of Fig. 5.

The role of the guest-host coupling parameter is different depending on the resonance position in the band, as seen from the calculations presented on the same

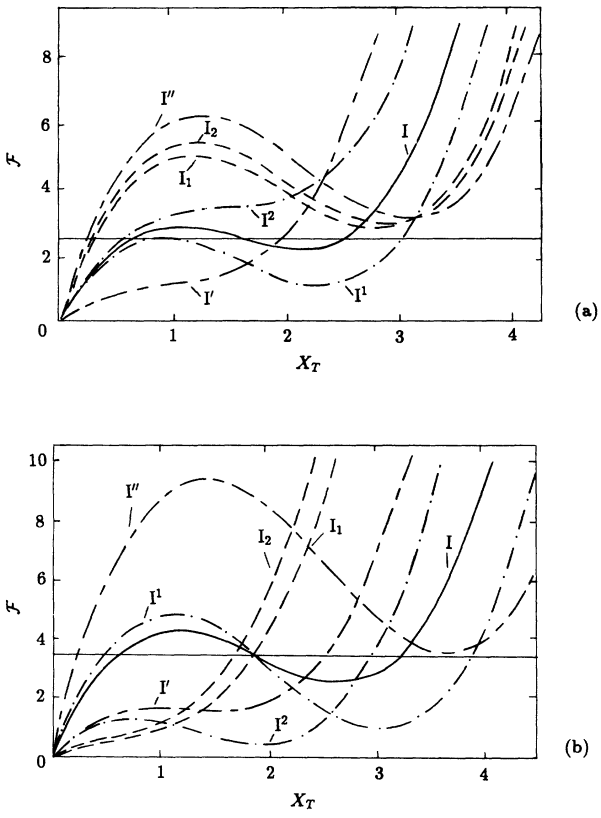

FIG. 5. The dependencies $\mathcal{F}(X_T)$ giving at intersections with lines $X_I(X_T)=$const (horizontal line) solutions of Eq. (14). (a) Near the resonance $k_r=0.285$, $V_0=1.9$, $\alpha=0.1$. Curve I corresponds to $k=0.31$ ($a^2=5.55$). The changes of these curves with k, α, and Φ are represented by long-dashed short-dashed, dashed, and dashed-dotted lines: I′, I″, $k=0.3$, 0.32 ($a^2=2.03$, 10.6); I_1, I_2, $\alpha=0.25$, 0.5; I^1, I^2, $\Phi/|L|=-0.08$, 0.2. (b) Near the resonance $k_r=1.04$, $V_0=1$, $\alpha=0.1$. Curve I, $k=1.07$ ($a^2=7.9$). I′, I″, $k=1.05$, 1.08 ($a^2=0.73$, 14.33); I_1, I_2, $\alpha=0.25$, 0.5; I^1, I^2, $\Phi/|L|=-1.296$.

figure. For the near-band-bottom resonance, Fig. 5(a), the range of bistability becomes larger for larger values of α. In contrast, bistability disappears for the same changes in α, when it deals with a resonance, which is distant from the band bottom, as in Fig. 5(b). The difference is connected with the type of dependence a on α, namely whether it is rising or falling with α. The latter depends on a precise role of the coupling parameter, an increase of which may lead to a disappearance [as in case (a)] or to a broadening of the resonance.

Since in a practical realization of the model discussed its parameters are fixed, a possibility to control the bistability with some outer parameter is of prime interest. Variations of the incident-flux intensity for this purpose, as was proposed previously,[2] are not the best choice, since the relevant experimental setup is questionable. Here we examine another possibility, namely, using the electric potential to change the bistable state of the resonant structure. In other words, we are interested mainly in the bistable current-voltage characteristics. As follows from Eqs. (17) and (18) a and b are field dependent (in the theory of Davydov and Ermakov $b=1$ and $a=$const for the structure given). Changes in these quantities with the field, the most important of which is the linear dependence a on Φ, provoke transformations of the curve $\mathcal{F}(X_T, Y)$, see Fig. 5, and may lead to essential changes in the bistable-state properties. In general, an increase of the right-semichain potential facilitates manifestation of the bistability, as is seen in Figs. 5 and 6. A decrease of the potential results in the opposite effect. Physically, it is quite natural to account for the role of Φ as "tuning parameter."

The graphic solutions presented are helpful for a qualitative understanding of the electric-field influence on the bistable state and of the relationship between different parameters of the system. A quantitative description of this influence is given by numerical solution of Eq. (14) shown as functions $X_T(X_I)$, $X_T(\Phi)$ in Figs. 6 and 7.

While dependence X_T (X_I) is always S-shaped, when the condition $a > a(k_{cr})$, Fig. 6, the function $X_T(\Phi)$ has a rich variety of forms, which correlate strongly with the position of the resonance in the band, compare solid curves of Figs. 7(a) and 7(b), and with the parameter X_I. (When speaking about the electric current through the defect, the latter associates with the density of free charge carriers in the chain.) The transmission coefficient (dashed and dashed-dotted lines in the same figure), and thus, the conductance [relation (27) is assumed to be true] may demonstrate bistable behavior in several ways depending on the two factors mentioned. These are now put into focus.

The range of the bistability is generally asymmetric with respect to the sign of the applied potential except for the special case when the resonance occurs in the middle of the band. Due to this asymmetry the bistability can manifest for one direction of the current and it can be absent for the opposite one, as is illustrated by curves I and 1 in Fig. 7(a). For this example the bistable state is available in a certain region of the potential magnitude. Consequently, changes in potential inside the interval between Φ_{cr} indicated on the figure and the limiting positive value of Φ will be accompanied by changes in β_T very similar to those in the linear regime of tunneling. But going to the left of the critical point, two different values of β_T can be observed, which correspond to a strong (high-transparent state) and to a weak (low-transparent state) amplitude of the current. Obviously, if being in the bistable region of the applied voltage, further rising of the potential causes, independently of the state of the system, a smooth change in the current up to the point it disappears completely. By contrast, lowering the potential from the same starting point causes an abrupt weakening in the current at $\Phi=\Phi_{cr}$, where the system was previously found to be in the high-transparent state. This possibility is illustrated by arrows.

The picture of the transmission-coefficient–voltage dependence is quite different from that described above, if the function $X_T(-\Phi)$ is S-shaped. Then, as is demonstrated by curve 2 [Fig. 7(a)], varying the potential in both sides reveals bistability but its manifestation will be different depending on the history of the system, in particular, on the sign of the potential, which is applied first.

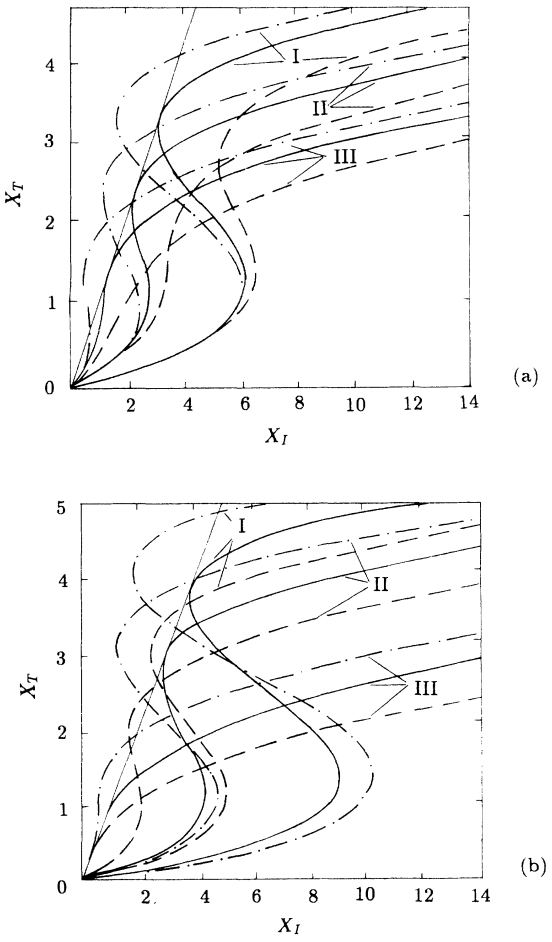

FIG. 6. Solutions of Eq. (14) as functions of the incident-flux intensity. (a) $V_0=1.9$, $\alpha=0.1$, $k=0.32$ (I), 0.31 (II), 0.3 (III), $\Phi/|L|=0$ (——), 0.2 (— — —), −0.8 (—·—·—·). (b) $V_0=1, \alpha=0.1, k=1.08$ (I), 1.07 (II), 1.05 (III), $\Phi/|L|=0$ (——), 2.5 (— — —), −1 (—·—·—·).

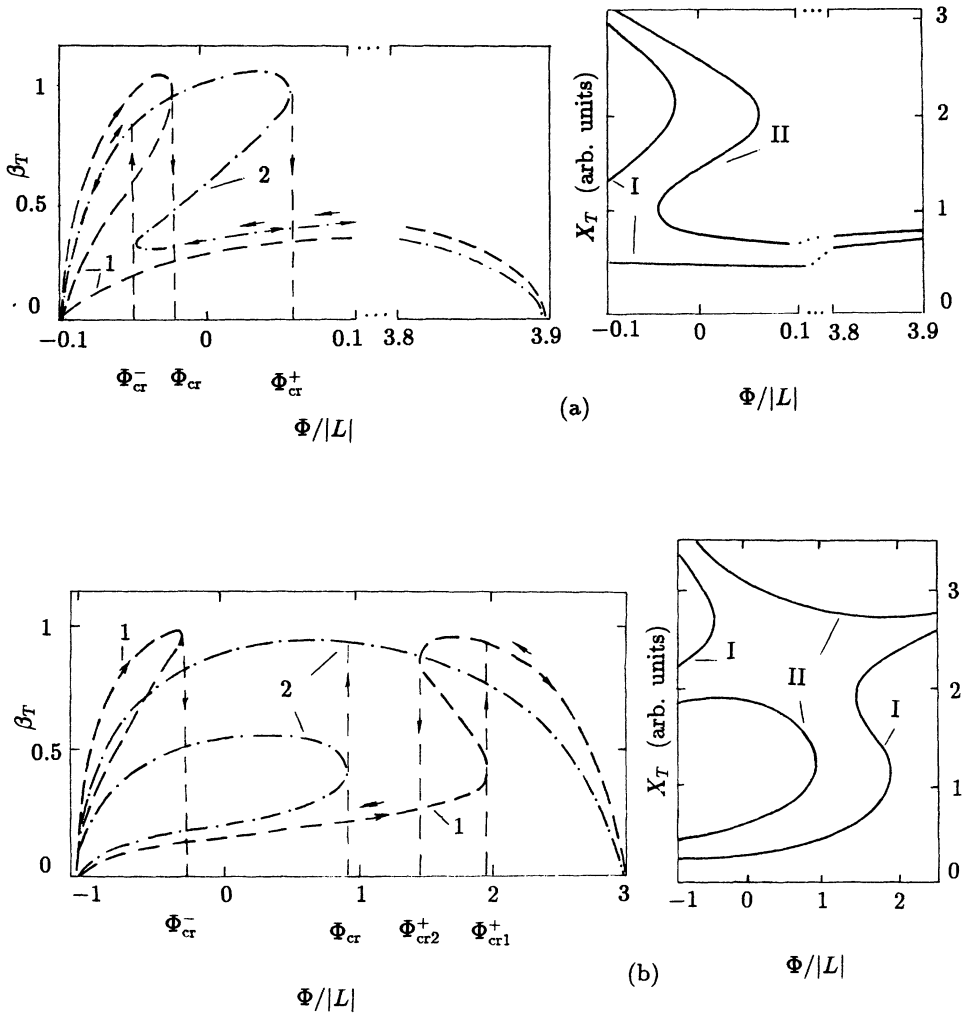

FIG. 7. Solutions of Eq. (14) (solid lines) and transmission coefficient (dashed and dashed-dotted lines) as functions of the applied potential. (a) $V_0=1.9, \alpha=0.1, k=0.31, X_I=2$, (I,1), 2.65 (II,2). (b) $V_0=1, \alpha=0.1, k=1.07, X_I=2.5$ (I,1), 3.5 (II,2).

For instance, let the system be in the low-transparent state at $\Phi=0$. In this case, changing the potential from zero to $\Phi>\Phi_{cr}^+$, then back to $\Phi=0$, then further to $\Phi<\Phi_{cr}^-$, and again back to zero, results in a hysteresis loop, which is to the left of the zero-point potential. Correspondingly, the current-voltage characteristic has also a loop in the negative potential region. But making the same cycle in Φ in the opposite direction, i.e., going first to the left along the Φ axis then to the right, and then back to zero, one obtains the hysteresis loop around the point $\Phi=0$ or, equivalently, two current-voltage loops on both sides from the zero voltage. Evidently, when the high-transparent state is the initial state, analogous cyclic changes of potential in the opposite directions result in current-voltage dependencies, that differ from each other as described above, within the accuracy of the size of the exchange between the one- and two-loop cases. The I-V (current-voltage) curves which correspond to this particular voltage dependence of the transmission coefficient are shown on Fig. 8.

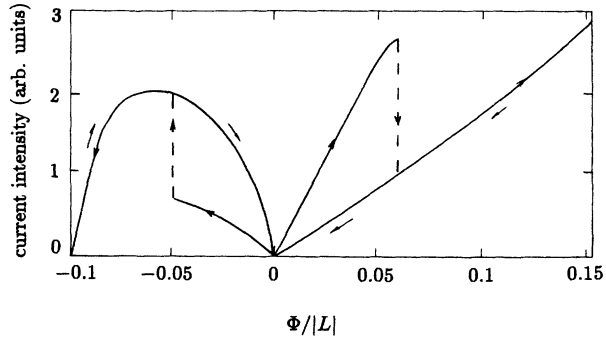

FIG. 8. The current response to changes $0\rightarrow\Phi_-\rightarrow\Phi_+\rightarrow 0$ (the on-line arrows correspond to the first passage in Φ, the out-of-line arrows to the second one). If the same cycled changes in Φ are performed in the opposite direction, the right hysteresis loop is not "observed." In calculations the parameters $V_0=1.9, \alpha=0.1, k=0.31$ are used.

The new interesting feature of the next example, curves I and 1 in Fig. 7(b), is that the bistability does not exist in the region $\Phi_{cr}^{-} < \Phi < \Phi_{cr2}^{+}$ and can be switched on by a sufficiently large field of both $\Phi > \Phi_{cr2}^{+}$ and $\Phi < \Phi_{cr}^{-}$. For negative values of the applied potential the transmission-coefficient–voltage dependence is analogous to that presented by curve 1 [Fig. 7(a)]. For $\Phi > 0$ an increase of Φ results in an abrupt increase of β_T at $\Phi = \Phi_{cr1}^{+}$. The reverse change in Φ leads to an abrupt decrease at $\Phi = \Phi_{cr2}^{+}$. Thus, in this case the picture of hysteresis in the current-voltage characteristic (outlined by arrows) is similar to the dependence of the transmitted flux on the incident-flux intensity obtained by Davydov and Ermakov.

In the last example presented, the high-transparent state, the upper branch of curves II, 2 [Fig. 7(b)] is available for the system at any magnitude of the applied potential. If in this state, the transmission coefficient depends on Φ much like the linear case; compare curves IV and 2 in Figs. 4 and 7(b). At the same time, due to the shift up of the resonant-state energy, the value of β_T is several times larger, when the nonlinear regime of tunneling is operative, than it is in the linear regime of tunneling (i.e., when the charge-carrier density is small) or when the system is found in the low-transparent state. In the latter case, changing Φ from zero to $\Phi > \Phi_{cr}$ and then back to zero, as shown by arrows in curve 2 [Fig. 7(b)], gives rise to hysteresis in the current-voltage characteristics. Note that the model predicts an irreversible transfer at the point Φ_{cr} from the low- to the high-transparent state, from which the system cannot return. The nontrivial behavior of the current response just discussed can be expected for $\Phi > 0$. In the region of negative potentials the dependence $\beta_T(\Phi)$ [and $\sigma(\Phi)$] is smooth up to the limiting values of Φ, for which β_T becomes zero.

The calculations presented by no means exhaust all possible ways the current behavior responds to variations of the applied potential, when the system is in a bistable or in a near-bistable state. Despite the simplicity of the system, the current-voltage characteristics and their dependence on the parameters turn out to be rather complex and could hardly be predicted on phenomenological grounds. This proves the importance of the microscopic approach to the problem of bistability in resonance tunneling.

IV. CONCLUSION

Resonance tunneling in a molecular wire with a single guest molecule has been studied starting from a microscopic Hamiltonian, which includes Coulomb electron-electron interaction at the guest site and is the one-particle Hamiltonian elsewhere. To account for the external electric-field effects the potential difference between the emitter and the collector leads was introduced but the defect state assumed to remain unchanged in the field. The model formulated is a simplified version of the double-barrier resonant structure composed of a quantum dot connected with a quantum wire by tunneling barriers.

In the mean-field approximation a closed set of self-consistent nonlinear equations has been obtained to define the wave function of tunneling electrons for a standard scattering picture of the stationary tunneling process. The solution of these equations has been used for studying the transmission-coefficient properties. Among them the dependence of the tramsitted flux on the applied electric potential received the most attention, since for a degenerate Fermi gas it determines the conductance of the wire under the assumption that the potential is much smaller than the Fermi energy.

Both the linear and nonlinear regimes of tunneling have been considered. In the former case (and in the absence of the electric field) the result of Sautet and Joachim[9] obtained with the transfer-matrix technique was rederived, using the density-matrix formalism. The expression for the transmission coefficient for neutral species has been generalized with an account of the external field and examined in some detail. An analytic formula prescribing nonlinear behavior of the wire conductance in response to the variation of the applied potential is the main result of this part of the paper.

When the energy of the tunneling electrons approaches the resonance, the electronic density in the resonant structure increases substantially, becoming much larger than in the emitter and the collector leads. Therefore, considering tunneling near the resonance, the electron-electron interaction has been taken into account at the guest site. As a consequence of this interaction, the feedback between the incident-flux intensity determined by the charge-carrier density in the wire and the resonance-state energy arises, resulting in a bistable behavior of the transmission coefficient. The necessary and sufficient conditions for the bistable regime of tunneling to exist have been established and this state was investigated using graphic and numerical solutions of the nonlinear equations for the wave function. Some typical examples of the transmission-coefficient–voltage dependencies have been demonstrated to show how the switching on or out of the bistable regime of tunneling by the field affects the current response. Probably the most unexpected result predicted for the model is that the current-voltage characteristics can have one hysteresis loop, if the changes in the potential are of the type $0 \rightarrow \Phi \rightarrow -\Phi \rightarrow 0$, whereas there are two loops if the same changes are made in the opposite direction, i.e, $0 \rightarrow -\Phi \rightarrow \Phi \rightarrow 0$.

The relationship between the model parameters and the form of I-V curves investigated in the present paper clarifies the microscopic picture of the intrinsic bistability formation, yet its experimental verification as just the possibility of practical utilization of the predicted effects in electronic devices is still in question. Work in this direction is now in progress.

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