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Coulomb blockade of resonant tunneling

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Ultrasmall double-barrier semiconductor structures are investigated in terms of the semiclassical sequential theory of resonant tunneling. The quantization of the charge buildup in the quantum well is taken into account. A peaked *I-V* characteristic is obtained, with each peak corresponding to an integer number of electrons in the well. A new explanation of the experiment of Reed *et al.* [Phys. Rev. Lett. **60**, 535 (1988)] is proposed. The large value of the charging energy in their experiment $E_c = e^2/2C = 43$ meV makes semiconductor tunneling structures more convenient for observation of charging effects than the usual ones.

As a result of recent advances in lithography and crystal-growth technology, the possibility for an experimental investigation of two new phenomena appear. The first phenomenon is Coloumb blockade of single-charge-carrier tunneling in ultrasmall normal or superconducting junctions.[1] Modern nanolithography forms the background for experimental investigations of this effect.[2] The second phenomenon is resonant tunneling in double-barrier semiconductor structures.[3] These structures are made with the high technology of molecular-beam epitaxy. Combining these two techniques one can make a new device—ultrasmall resonance tunneling structure[4] (see Fig. 1). In such structures the effects of charge quantization become noticeable.[5]

Recently Reed *et al.*[4] investigated electronic transport through a three-dimensionally confined quantum well (see Fig. 1). Their structure consisted of a 500-nm n^+-type GaAs layer (Si doped at 2×10^{18} cm^{-3}, graded to approximately 10^{16} cm^{-3} over 20 nm, followed by a 10-nm undoped GaAs spacer layer), a 4-nm $Al_{0.25}Ga_{0.75}As$ tunnel barrier, and a 5-nm undoped $In_xGa_{1-x}As$ quantum well. Symmetrically about the central plane of the well corresponding layers were grown. Electron-beam lithography and reactive ion etching were used to define isolated columns. Reed *et al.* measured the current-voltage characteristics of large-area mesas (with lateral dimensions ≥ 2000 nm) and detected two resonance peaks: a ground state at 50 meV and an excited state at 700 meV.

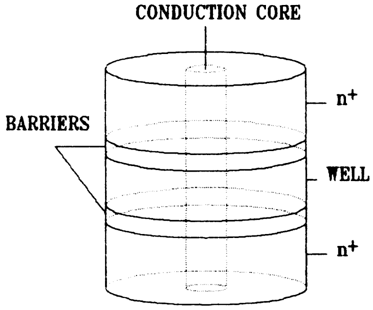

FIG. 1. Ultrasmall double-barrier structure. The conduction core is about five times smaller than the physical dimensions of the structure.

They also measured small-area mesas (quantum-box resonance-tunneling structure) with 100 nm diameter. At low temperature (≤ 10 K) they observed additional fine structure emerging superimposed on the excited-state resonance peak (see the inset of Fig. 2).

There are two theories attempting to explain the position of the fine-structure peaks. Reed *et al.* made estimates for the lateral confinement due to the sidewall depletion, supposing harmonic-oscillator radial potential. They obtained the equidistant harmonic-oscillator splitting of 25 meV which they considered to be consistent with the splitting of the upper four peaks. They could not explain the position of the lowest peak, however. To account for this fact, Luban *et al.*[6] proposed a one-parameter family of anharmonic oscillator potentials which differs from the harmonic one only in a zone of radius ≤ 15 nm around the axis of the device.

Bryant[7] proposed another explanation, accounting for variable lateral confinement in the emitter (collector) and the well. Starting from the harmonic-oscillator splitting of $\Delta E_{box} \simeq 25$ meV in the quantum box and of $\Delta E_{contact} \sim 1$ meV in the contacts, he concluded that the

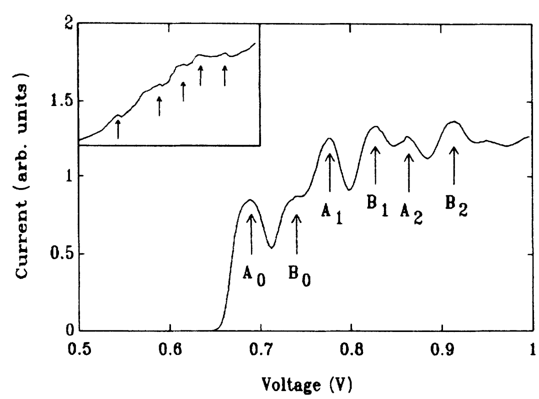

FIG. 2. Current-voltage characteristics of the device obtained with $E_c = 43$ meV, $\Delta E_{box} = 25$ meV, $\sigma = 5$ meV, $\alpha = 0.007$, and $\tau_c/\tau_e = 10$. In the inset schematic low-temperature current-voltage characteristics are shown, obtained by Read *et al.* (Ref. 4). The peaks are grouped in two sets A_n and B_n, $n = 0,1,2$. The number n of the peak denotes the number of the electrons in the well.

lowest peak is one from the oscillator set and there is a missing peak in the middle between the first and second peak. He explained its absence with a large subband coupling at the contact barrier interfaces leading to a multichannel tunneling. Trying to describe the positions of the peaks of the experiment of Reed *et al.* using Bryant's theory, one is forced to use a special form for the lateral potential again—a harmonic oscillator.

But there are some important facts that raise questions about the applicability of the above-mentioned theories.

(i) The first fact is that the lateral splitting of the excited resonance state can produce only one or two resonance levels. We take the effective masses of the structure[7] $m_{\mathrm{GaAs}}=0.067m_e$, $m_{\mathrm{Al\text{-}Ga\text{-}As}}=0.087m_e$, $m_{\mathrm{In\text{-}Ga\text{-}As}}=0.063m_e$, the conduction-band offsets above the Fermi level of n^+-type doped GaAs $V_{\mathrm{Al\text{-}Ga\text{-}As}}=V_{\mathrm{GaAs}}+203$ meV, $V_{\mathrm{In\text{-}Ga\text{-}As}}=V_{\mathrm{GaAs}}-72$ meV (V_{GaAs} is the offset of the conduction band of the pure GaAs). If we take the 5-nm In-Ga-As well we find that the excited resonance state has energy $V_{\mathrm{GaAs}}+200$ meV, i.e., at the very top of the well. Taking into account that the lateral splitting is about[6,7] $\Delta E_{\mathrm{box}}\approx 25$ meV and the fact that lateral confinement can only raise the levels we see that in the well we have only one resonance state. If we take a 6-nm well like Briant,[7] we find for the resonance state $V_{\mathrm{GaAs}}+173$ meV, so there are two states. We shall consider below only the latter case and at the end of the work we will briefly discuss the other possibility. We will denote the lower level with the letter A and the upper one with the letter B.

(ii) Another common feature of the above-mentioned theories is that they did not take into account the electrostatic feedback effects due to the accumulation of the charge in the well. An estimation of their importance can be made by modeling the double-barrier structure with two capacitors C connected in series.[8] The effects have to be of the order of the charging energy $e^2/C=2E_c$ of the single electron (e is the electron charge). If we take the value of Reed *et al.* for the conduction core diameter (13 nm) we can calculate the capacitance of a planar capacitor with dielectric constant $\epsilon=13.2$ and $10+4+2.5$ nm separation between plates. We conclude that the charging energy is of the order of a tenth of an electron volt, i.e., of the order of peaks' separation. This is a clear indication that we cannot neglect the charging effects in this case.

(iii) The third observation is that both theories predict equidistant peaks, but if we look closely at the data of Reed *et al.* we see that the upper four peaks are not *exactly* equidistant. We see that there are two sets of equally spaced peaks at a distances of 87 mV: the set A_n and the set B_n, $n=0, 1, 2$. The set B_n is shifted from A_n by a bias of 50 mV. At the position of peak B_0 there is a weak shoulder on the data of Reed *et al.*

(iv) The last common feature of the above-mentioned theories is that they are based on the supposition of some special kind of lateral potential shapes. It is hard to believe that their predictions will not be greatly altered by irregularities of the wall shapes, imperfections, defects, etc.

Our theory attempts to take these facts into account.

In what follows we will not use a special kind of lateral potential, but we will take as given fact that, whatever it is, there is lateral splitting between the state A and the state B of the order of $\Delta E_{\mathrm{box}}\approx 25$ meV. Because this splitting is defined mainly by the geometrical parameters of the system, this is a more reliable supposition than guessing the exact potential shape. We will also take into account the electrostatic feedback modeling the double-barrier structure with two capacitors connected in series.[8] We will suppose that the shape of the potential well is not altered by the trapped charge, but is only shifted at energy nE_c (see below), where n is the number of trapped electrons in the well. This shift leads to corresponding shifts of levels A and B providing a natural explanation of the experiment of Reed *et al.*

We will consider the tunneling events in the framework of sequential theory of resonance tunneling.[6,9,10] This can be done in terms of the semiclassical model of tunneling.[11]

The semiclassical model assumes that the state of the system can be completely described by classical variables N_l and N_u, the number of the electrons on the lower and upper resonance state, respectively. Taking into account the Pauli principle we find that $N_l,N_u=0,1,2$. Electrostatically, this structure is equivalent to two identical capacitors C connected in series, with charge $Q=(N_l+N_u)e$ on the common plate. The potential drop across the emitter and collector capacitors is

$$V_e(N_l,N_u)=[V-(N_l+N_u)e/C]/2\,,$$
$$V_c(N_l,N_u)=[V+(N_l+N_u)e/C]/2\,, \qquad (1)$$

where $L_u(N_l,N_u)$ and $L_l(N_l,N_u)$ are the transition rates through the emitter barrier into the upper and lower state, respectively, and $R_u(N_l,N_u)$ and $R_l(N_l,N_u)$ are the transition rates through the collector barrier. If $D_e(E)$ is the energy distribution of the emitter electrons hopping into the well and $D_l(E)[D_u(E)]$ is the density of states of the lower (upper) state in the well, we can write

$$L_l(N_l,N_u)=(2-N_l)\tau_e^{-1}\int D_e(E-eV)D_l(E-eV_c(N_l+1,N_u))dE\,, \qquad (2)$$

where τ_e is the elastic tunneling time of the electron through emitter barrier, $2=N_l$ is the number of empty states in the lower level, and $eV_c(N_l+1,N_u)$ is the shift of the position of the resonance levels *after* the electron has tunneled. Similar expressions can be written for L_u,R_l,R_u. If E^0_{res} is the height of the lower resonance level according to the Fermi level at zero voltage (including the shift of conduction band of pure GaAs above the Fermi level of doped GaAs) we can write

$$D_l(E)=\delta(E-E^0_{\mathrm{res}}),\quad D_u=\delta(E-E^0_{\mathrm{res}}-\Delta E_{\mathrm{box}})\,, \qquad (3)$$

where δ is Dirac's δ function. Using (1)-(3) leads to

$$L_l(N_l,N_u)$$
$$=(2-N_l)\tau_e^{-1}D_e(E^0_{\mathrm{res}}-e[V-e(N_l+N_u+1)/C]/2)\,.$$

A similar expression can be derived for L_u. Taking into account that in the collector all states are free, we can write

$$R_l(N_l,N_u)=N_l/\tau_c,\ \ R_u(N_l,N_u)=N_u/\tau_c\ .$$

The time evolution of N_l,N_u can be expressed as a stochastic process; given N_l,N_u at time t, then at time $t+\Delta t$ N_l changes into N_l+1 with probability $L_l(N_l,N_u)\Delta t$; into N_l-1, with probability $R_l(N_l,N_u)\Delta t$; and stays unchanged with probability $1-[R_l(N_l,N_u)+L_l(N_l,N_u)]\Delta t$ and an analogous picture for N_u. The probability distribution $\rho(N_l,N_u,t)$ for having N_l charges on the lower level and N_u on the upper level at time t for fixed V can be determined by solving the master equation, which has the following form:[11,12]

$$\frac{d\rho(N_l,N_u,t)}{dt}=L_l(N_l-1,N_u)\rho(N_l-1,N_u,t)+L_u(N_l,N_u-1)\rho(N_l,N_u-1,t)+R_l(N_l+1,N_u)\rho(N_l+1,N_u,t)$$
$$+R_u(N_l,N_u+1)\rho(N_l,N_u+1,t)-[L_u(N_l,N_u)+L_l(N_l,N_u)+R_u(N_l,N_u)+R_l(N_l,N_u)]\rho(N_l,N_u,t)\ .$$

Solving these equations in the stationary case we can obtain the current using the formula

$$I=e\sum_{N_l=0}^{2}\sum_{N_u=1}^{2}\rho(N_l,N_u)[R_l(N_l,N_u)+R_u(N_l,N_u)]\ .$$

To do this we have to choose the function of the tunneling electrons' energy distribution. Though the concrete shape of the energy distribution is irrelevant,[13] for the position of the peaks we have used the Gaussian shape with characteristic width of the dispersion, which have to be about the Fermi energy corresponding to concentration of $2\times10^{16}\ \mathrm{cm}^{-3}$, i.e., about 5 meV. In the above consideration we have accounted for only elastic (i.e., energy-conserving) tunneling of the closest to the barrier electrons. To account for the scattering events (leading to the loss of energy of tunneling electrons) and the possibility to hop from far from the barrier regions (i.e., with larger Fermi energy) we add a small low energetic "tail" of the energy distribution so that it becomes

$$D_e(E)=\frac{1}{\sigma\sqrt{2\pi}}e^{-E^2/2\sigma^2}+\alpha\theta(-E)\ ,$$

where σ is the dispersion width, and $\theta(x)=1$ for $x>0$ and $\theta(x)=0$ for $x<0$ is the unit step function and α is a phenomenological parameter.

In Fig. 2 the current-voltage characteristics are given, obtained with the following parameters: $\tau_c/\tau_e=10$, $\alpha=0.007$, $\sigma=5$ meV, $E_c=43$ meV, and $\Delta E_{\rm box}=25$ meV.

This curve has to be compared with those obtained by Reed *et al.* (inset of Fig. 2). In fact, we have two sets of peaks produced by the two laterally split resonance states. Each set is labeled with the number of electrons that are trapped in the well *before* the electron tunnels. The shift between the sets is $2\Delta E_{\rm box}$ and the spacing within a given set is e/C. The peak B_0 is masked by the adjacent peak A_1 and there is a shoulder at this position—a fact mentioned by Bryant. At last we have to mention that although the position of peaks did not depend upon concrete distribution of the emitter electrons, and the values of the parameters $\alpha,\tau_e/\tau_c$, the shape of the current-voltage characteristic is rather sensitive to them. Thus, further improvements of the theory are needed.[13]

In order to verify the explanations of the effect one has to study the dependence of the distance between the peaks on the physical radius R of the structure. The explanation of Reed *et al.*[4] is based upon the formula $\Delta E_{\rm box}=(2\Phi_T/m^*)^{1/2}\hbar/R$, where Φ_T is the height of the lateral potential determined by the Fermi-level pinning (fixed at 700 meV) and leads to a $1/R$ dependence. In our case, however, if we suppose that the depletion depth is the same, the area of the effective capacitor changes proportionally to R^2, so we suggest that the distances between the peaks within one set (A_n or B_n) should vary as R^{-2}. The shift between two sets, however, will depend upon the shape of the lateral potential. For example, if we accept the model of Reed *et al.*, it should vary as R^{-1} but if we accept the infinite-square-well model for the lateral potential we have to expect R^{-2} dependence. The other way to experimentally test our picture is based upon the fact that the current is carried by single electron-tunneling events that are correlated in space and time. Thus we expect the current to be oscillating with fundamental frequency $f=I/e$ (see the work of Delsing *et al.*[1]).

At last we will briefly discuss the case in which there is only one level at the top of the well. In this case the only possibility is to suppose that the shift of two sets is given by the optical-phonon emission. This explanation will be sound only if because of unhomogeneous structure there exist phonon modes with energy ~25 meV because the bulk optical phonon of the pure GaAs has 35 meV energy.

In conclusion, we have proposed a new explanation of the experiment of Reed *et al.* We believe that they had observed charging effects in double-barrier structures. The large value of the charging energy supports the conclusion that all charging effects can be more easily investigated on semiconductor structures than on small normal-metal-insulator-normal-metal ($N/I/N$) or superconductor-insulator-superconductor ($S/I/S$) tunneling junctions, small particles, and scanning-tunnel-microscope measurements.[1]

Note added in proof. Very recently[14] Reed and collaborators have performed R-dependence experiments suggested by us. They have shown that charging effects cannot be detected in such structures. This fact is due to bias caused weakness of the collector barrier, i.e., $\tau_c\ll\tau_e$. The theory presented above is applicable to structures with strong enough collector barrier (for example, in field effect quantum dots).

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