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Single-electron tunneling and Coulomb charging effects in asymmetric double-barrier resonant-tunneling diodes

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Resonant tunneling is studied in an ultrasmall asymmetric GaAs-$Al_xGa_{1-x}As$ double-barrier diode at low temperatures. In reverse bias, spikelike current-voltage characteristics are observed and assigned to electrons tunneling from zero-dimensional (0D) states in the accumulation layer to 0D states in the well. The 0D-0D tunneling reflects the single-electron spectrum without Coulomb charging effects. In forward bias, steplike current-voltage characteristics are observed and ascribed to tunneling from one-dimensional subbands in the emitter contacts through 0D states in the well, accompanied by Coulomb charging effects. A moderate magnetic field ($B \approx 4$ T) parallel to the current improves the flatness of the plateaus.

Fine structure in the negative-differential-conductance (NDC) peak of ultrasmall diameter (≈ 100 nm) resonant tunneling diodes (RTDs) has been observed by several groups.[1-6] Two mechanisms have been proposed to explain the fine structure: (i) The strong confinement due to sidewall depletion quantizes the electron motion parallel to the barrier interfaces (i.e., the lateral motion) with corresponding lateral single-particle energies E_n. Quasi-one-dimensional (1D) subbands are formed in the contacts and zero-dimensional (0D) states are formed in the quantum dot.[1-4,6,7] Steplike fine structure appears in the current-voltage (I-V) characteristics when the energies of the 0D states in the quantum dot E_n align with the electrochemical potential in the emitter. Potential fluctuations or single impurities in the well could also be responsible for the lateral confinement.[5] (ii) The steplike I-V fine structure appears when the bias is high enough to overcome the electrostatic energy necessary to charge the dot with one additional electron.[8,9] In analogy to the charging energy of a capacitor, the Coulomb energy is usually taken as $E_C(N) \approx N^2e^2/2C$, where N is the number of electrons in the dot, e is the electron charge, and C is the effective capacitance.[8-12] In metals, the quantization energies are negligible, and a model based on just the Coulomb term can reproduce successfully the I-V characteristics of nanometer scale metal-electrode tunnel junctions.[12] However, in semiconductors, where both the electron effective mass and the Fermi energy are small, the quantization energies and the Coulomb charging energies are comparable. This has been demonstrated in conductance studies of quantum dots formed in a high mobility two-dimensional electron gas.[13]

In this paper we report tunneling experiments in an ultrasmall asymmetric RTD, where charging effects are known to be important only in one bias polarity.[11,14] We observe differences between the fine structures in forward and reverse bias that we attribute to Coulomb charging effects. The experimental data also illustrate the difference between tunneling through single 0D states (1D-0D tunneling) and through two 0D boxes in series (0D-0D tunneling).

The asymmetric double-barrier structure was grown by molecular-beam epitaxy (MBE) on an n^+-type (100) GaAs substrate. The undoped central layer consists of a $d_1 = 10$ nm $Al_{0.33}Ga_{0.67}As$ barrier (top side), a $w = 14$ nm GaAs well, and a $d_2 = 7$ nm $Al_{0.33}Ga_{0.67}As$ barrier (substrate side). Top and bottom contact layers consist of a $d_0 = 7$ nm undoped GaAs spacer (at the barrier interfaces), followed by a 350-nm GaAs layer doped to 2×10^{16} cm^{-3}, a 28-nm GaAs region with the doping graded from 2×10^{16} to 1.4×10^{18} cm^{-3}, and a 350-nm GaAs layer, heavily doped to 1.4×10^{18} cm^{-3}. The dopant used was silicon. With electron-beam lithography and CH_4-H_2 metalorganic reactive-ion etching,[15] free-standing single RTDs with diameters between 0.1 and 10 μm were fabricated.[3] The diode presented here has a nominal diameter of $D = 1$ μm. The sidewall depletion width is $W \approx 0.3$ μm (estimated from the contact doping level) and therefore the diode is not pinched off, but the conducting diameter d_{cond} is small enough to exhibit lateral quantization. The remaining background impurity concentration of $N_D \approx 3\times10^{14}$ cm^{-3} corresponds to an average of less than one impurity in the well.

Figure 1 shows a schematic band diagram to illustrate tunneling in the laterally confined asymmetric RTD. Under reverse bias [Fig. 1(a)], 0D states are formed in the accumulation layer at the interface of the thick barrier (box 1) and in the well (box 2). When in resonance the energies of the 0D states in box 1 and box 2 line up, electrons can tunnel between the 0D states, leading to spikes in the I-V characteristics.[16,17] The tunneling rate Γ_1 through the thick emitter barrier is much lower than the tunneling rate Γ_2 through the thin collector barrier (i.e., $\Gamma_1 \ll \Gamma_2$), and there is at most one electron at a time in box 2. The spacing of adjacent single-particle states $\Delta E_{n,n+1}$ is related to the measured spike spacing in bias ΔV_S through the relation $\Delta V_S = \Delta E_{n,n+1}/e\eta_{rev}$, where $\eta_{rev} = (d_1 + 0.5w)/(d_1 + w + d_2 + d_0)$ is the fraction of the reverse bias voltage dropped between the emitter and well (see Fig. 1). Thus the resonance spacings in bias are related to the 0D single-electron spectrum E_n in box 2.

Tunneling in forward bias is depicted in Fig. 1(b).

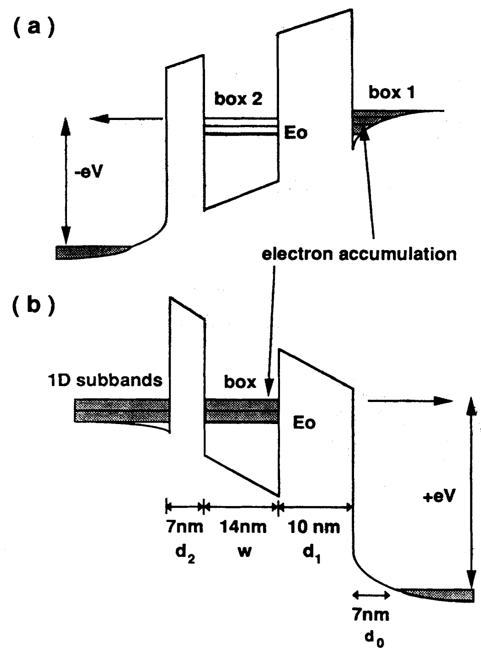

FIG. 1. Band diagram of the asymmetric GaAs-$Al_xGa_{1-x}As$ double-barrier structure, illustrating the tunneling mechanism. 1D subbands are formed in the contact regions, and 0D states are created in the well. (a) In reverse bias electrons tunnel from the 0D states in the accumulation layer (box 1) into the 0D states in the well (box 2). (b) In forward bias, electrons tunnel from the 1D subbands into the 0D states in the well.

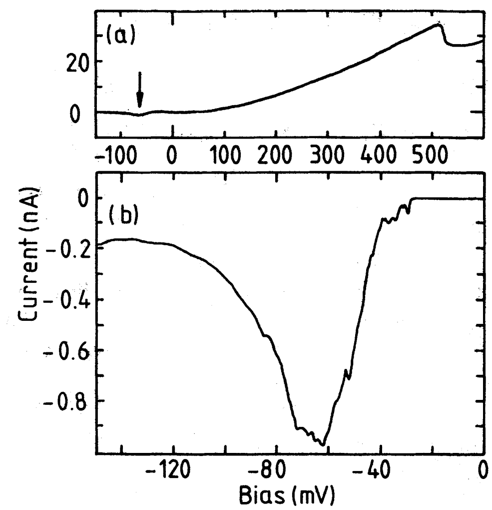

FIG. 2. (a) Current-voltage (I-V) characteristics of a 1-μm-diam asymmetric GaAs-$Al_xGa_{1-x}As$ resonant tunneling diode. The strong asymmetry of the I-V is due to charge accumulation in forward bias. The arrow indicates the small ground-state resonance in reverse bias. (b) Expansion of the reverse bias ground-state resonance, showing fine structure over the whole resonance peak.

When the 0D states in the well fall below the electrochemical potential of the emitter contact, electrons can tunnel from the 1D subbands in the emitter through the thin barrier (with tunneling rate Γ_2), which is too transparent to form an accumulation layer. If the quantum dot contains N electrons, a current step will occur when an additional electron tunnels into the quantum dot, and the energy of the dot will increase by $\Delta\tilde{E}_{n,n+1} = \Delta E_{n,n+1} + \Delta E_C$, where $\Delta E_C = e^2/C$. Thus the plateau widths in bias, $\Delta V_P = \Delta\tilde{E}_{n,n+1}/e\eta_{\text{for}}$, may be larger than the spike-spacing ΔV_S in reverse bias, where only the single-electron spectrum E_n is probed. The value η_{for} is the fraction of voltage dropped between emitter and well in the forward bias configuration. Note that any degeneracy of the single-electron states E_n is lifted by the Coulomb charging energy ΔE_C.

Figure 2 shows the I-V characteristics of the RTD as measured in a top loading dilution refrigerator operating at a base temperature of $T \approx 20$ mK. We know from previous experiments[1–6] that the fine structure is superimposed upon the two-dimensional NDC peaks. Such NDC resonance peaks are present in both bias polarities, but there is a strong asymmetry. The ratio of the NDC peak currents in the two directions is 36:1, and the corresponding ratio of the peak biases is 8:1. The ratios are consistent with experimental data on large area asymmetric double-barrier RTDs and demonstrates that in forward bias there is a charge accumulation in the quantum well.[14] By comparing the peak current of the ultrasmall RTD with that of a 10-μm-diam reference diode, we estimate that $d_{\text{cond}} \approx 360$ nm. The first NDC peak in reverse bias shows rich fine structure that extends over the whole resonance peak. The fine structure is not limited to the low bias regime and therefore it is probably not due to impurity states in the well.[5]

Figure 3 shows an expansion of Fig. 2 in the low bias region. In reverse bias the I-V curves show spikelike fine structure reminiscent of 0D-0D tunneling as discussed previously.[16,17] The peak spacing in bias (indicated in Fig. 3 by the arrows) is $\Delta V_S \approx 0.5$–1 mV, with peak amplitudes of about 50 pA. In forward bias, a clear I-V

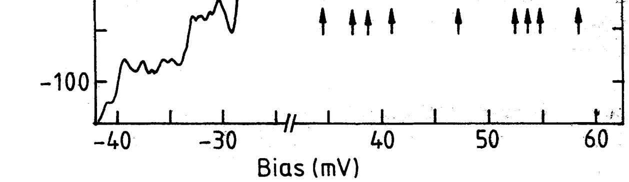

FIG. 3. Details of the I-V characteristics in Fig. 2, showing the steps with large plateau widths in forward bias. In reverse bias the resonance spikes show tunneling between two quantum boxes in series (0D-0D tunneling) and the small spike spacing in bias is assigned to the single-electron spectrum without electron-correlation effects.

TABLE I. Current step heights ΔI and corresponding plateau widths in bias ΔV, as the electron occupation number in the dot increases. Values are obtained in zero magnetic field and at $B=4$ T.

	$B=0$		$B=4$ T	
Number of electrons	ΔI (pA)	ΔV (mV)	ΔI (pA)	ΔV (mV)
1	5.0	3.5	5.0	4.0
2	14.0	1.0	24.0	6.5
3	10.0	3.3		
4	12.0	5.0	16.0	5.5
5	14.0	5.0	15.0	5.0
6	13.0	1.0	25.0	2.0
7	10.0	1.0	15.0	1.0
8	10.0	1.0	18.0	2.5

staircase is observed, with current step heights of $\Delta I \approx 12$ pA. Note, however, that the step height $\Delta I \approx 5$ pA of the first plateau is much smaller than the adjacent plateaus. This effect has also been observed in the Coulomb staircase of ultrasmall metal junctions and was explained in the classical model for Coulomb charging.[12] The plateau widths in bias ΔV_P range between 1 and 6 mV, i.e., they are up to an order of magnitude larger than the peak spacing in reverse bias. In Table I we list the current step heights ΔI and plateau widths ΔV, as measured in zero magnetic field for each electron occupancy.

Figure 4 shows the forward bias I-V as a function of various magnetic fields applied parallel to the current. It can be seen that the "flatness" of the plateaus is improved at 4 T. The magnetic length of $l_B \approx 12$ nm at 4 T is small enough that electrons will not sample any fluctuations in the sidewall confining potential. For even higher fields, the plateaus are quenched; this occurs when l_B is sufficiently small that the electrons may sample fluctuations associated with the barrier interfaces. In Table I the step heights ΔI and plateau widths ΔV are listed from data obtained at $B=4$ T (Fig. 4).

The steplike I-V in forward bias can be assigned to single-electron tunneling. In a model of single-electron tunneling through an asymmetric double-barrier structure, the current levels for each step are given by $\Delta I = e\{\Gamma_1\Gamma_2/(\Gamma_1+\Gamma_2)\}$, where the bias-dependent tunneling rates Γ_1 and Γ_2 can be calculated using the WKB approximation. Using the barrier, well, and spacer layer thicknesses in Fig. 1 we calculate a step height $\Delta I_{\rm WKB} \approx 8$ pA at $V=+30$ mV, which is in good agreement with the experimental value $\Delta I_{\rm expt}=5$–14 pA. To assign our data to Coulomb charging effects, we have to estimate the typical charging energy of the quantum dot. The total capacitance of the double-barrier structure is estimated to be $C \approx C_1 + C_2 \approx 1.36\times 10^{-16}$ F, using $d_{\rm cond} \approx 360$ nm, the dielectric constant $\epsilon_{\rm GaAs}=13.2$, and the layer thicknesses specified in Fig. 1. From this we estimate that the Coulomb charging energy is $\Delta E_C \approx e^2/C \approx 1.2$ meV. The energy spacing between the 0D states due to lateral quantization is estimated to be $\Delta E_{n,n+1} \approx (2\hbar/d_{\rm cond}) \times (2E_F/m^*)^{1/2} \approx 0.6$ meV (for the Fermi energy $E_F=6$ meV, $d_{\rm cond}=360$ nm, and $m^*=0.067m_e$). Therefore, the spike spacing in reverse bias is estimated to be $\Delta V_S = \Delta E_{n,n+1}/e\eta_{\rm rev} = 1.3$ mV, in reasonable agreement with the data (0.5–1 mV). The calculated plateau widths in forward bias are $\Delta V_p = (\Delta E_{n,n+1} + \Delta E_C)/e\eta_{\rm for} = 4.88$ mV and $\Delta V_p = \Delta E_C/e\eta_{\rm for} = 3.2$ mV, in good agreement with the experimental data (1–6 mV).

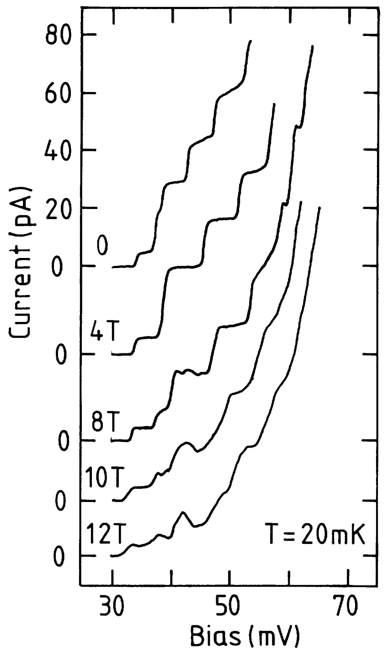

FIG. 4. Detailed I-V characteristics for low forward biases, as a function of a magnetic field applied parallel to the current.

The fact that the first I-V plateau width in forward bias $\Delta V_1 = 3.5$ mV is larger than the second plateau width $\Delta V_2 = 1$ mV is unexpected. Because of spin degeneracy the second electron that tunnels into the dot (i.e., when the second step arises) has the same single-particle energy as the first, and the width $\Delta V_1 = \Delta E_C/e\eta_{\rm for}$ is related only to the charging energy. However, the third electron tunnels into the dot at an additional bias $\Delta V_2 = (\Delta E_C + \Delta E_{2,3})/e\eta_{\rm for}$, because the first excited single-particle state in the dot is already occupied. Therefore, the corresponding plateau width ΔV_2 is expected to be larger than ΔV_1. A theory to explain the experimental fine structure should allow for $\Delta V_1 > \Delta V_2$, as observed in our data.

In conclusion, we have investigated fine structure in the NDC peaks of an ultrasmall asymmetric RTD. The fine structure is attributed to tunneling of individual electrons, since the current step heights ΔI in forward bias are in quantitative agreement with calculations using the tunneling rates for single electrons. We also understand qualitatively the tunneling between 0D states, as observed in the spikelike I-V in reverse bias. However, the plateau widths in forward bias cannot be modeled with present theories for single-electron tunneling that allow for lateral quantization and Coulomb charging effects.[10,11]

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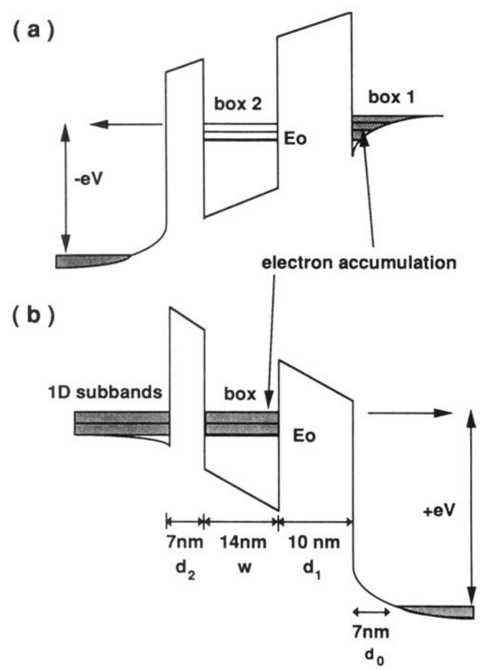

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