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Conductance oscillations in Ge/Si heterostructures containing quantum dots

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Abstract. The conductance of the Si/p^+-Ge/Si epitaxial heterostructure associated with hole tunnelling into isolated ultrasmall (~ 10 nm) quantum dots p^+-Ge has been studied. Quantum dots have been obtained after islanding of 1.3 nm Ge film during MBE growth by the Stransky–Krastanov mode method. Conductance oscillations as a function of bias voltage were observed. The experimental data are analysed in terms of a model that involves the interplay of single-electron charging effects and resonant tunnelling through individual energy levels.

A number of the investigations of Ge/Si heteroepitaxial growth have demonstrated the formation of Ge islands in the size range 10–100 nm on the Si surface [1–6] (the Stranski–Krastanov growth mechanism) if the effective Ge film thickness is more than 1 nm. Such islands must represent so-called 'quantum dots' (a system with zero-dimensional (0D) electron gas) because the spectrum of electrons or holes is quantized in all three dimensions.

It is known that single-electron tunnelling can cause periodic oscillations in the conductance of metallis grains as a function of gate voltage (the Fermi level position in a metallic particle) [7–9]. This so-called Coulomb-blockade effect is due to the discreteness of the electron charge. Recently this phenomenon was found in the Ge/Si structure [6]. The discrete spectrum of a low-dimensional semiconductor causes such a transport phenomenon as resonant tunnelling [10]. Theoretical investigations [11] have predicted that the transport properties of quantum dots are determined by the interplay between size and charge quantization effects, which is not feasible in metallic grains because of the small Fermi wavelength in a metal.

Usually quantum dots are fabricated by lateral confinement of an electron in a 2D electron gas using split-gate techniques [12, 13]. The scale of such dots exceeds a hundred nanometres due to the size limitation of lithography, so the level spacing ΔE in the dot is about one order less than the charging energy e^2/C (C being the capacitance of the islands) [11] which makes the analysis of experimental data difficult. Recently, resonant tunnelling in silicon microcrystallites of size 10 nm, with a-SiO_2 barriers has been reported by Qiu-yi Ye *et al* [14]. The μc-Si layer was produced by crystallization from the amorphous phase. In this paper, we suggest another method that allows the fabrication of a layer of Ge quantum dots 10–20 nm in diameter, buried in Si. In these structures we have observed the differential conductance oscillation as a function of bias voltage. Because of the small scale of the Ge islands, the level spacing and the charging energy are of comparable magnitude, and the interplay of resonant tunnelling and the Coulomb blockade becomes more pronounced. Moreover, the behaviour of conductance in a device with a large number of quantum dots distributed in size turns out to be the same as that in a single-dot structure.

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Heterostructures for tunnelling experiments have been grown on p^+-Si (001) substrates by MBE. Thermal cleaning at $T_S = 1300\,°C$ follows preliminary chemical processing of the substrate. The growth rate and layer structure have been controlled by using RHEED oscillations. Only four or five oscillations have been observed during Ge layer deposition. Then oscillations have disappeared, the specular beam intensity has decreased and the bulk diffraction pattern has been observed. These observations are evidence for morphological transformation of the flat strained Ge film into the droplet film. The density of the islands was found to decrease with increasing Ge temperature of growth. Figure 1 shows a plan-view transmission electron microscopy (TEM) sample of a layer on which $d_1 = 1.3$ nm of Ge has been deposited at $T_S = 300\,°C$ (*a*) and $400\,°C$ (*b*) and covered by 2.0 nm of amorphous silicon. The density of the Ge dots in the plane of growth was calculated to be $n = 3 \times 10^{11}$ cm^{-2} for $T_S = 300\,°C$ and $n = 1.4 \times 10^{11}$ cm^{-2} for $T_S = 400\,°C$. The islands can be varied in size from tens to hundreds of nanometres by controlling the substrate temperature T_S and the thickness of the Ge film during deposition.

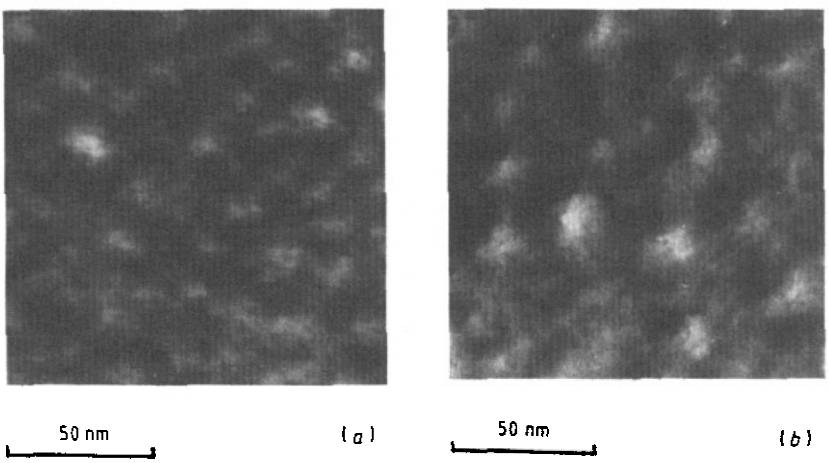

Figure 1. Plan-view TEM images of Ge islands formed after a 1.3 nm Ge deposition on a Si (001) surface at temperatures $T_S = 300\,°C$ (*a*) and $T_S = 400\,°C$ (*b*). The islands are covered by 2 nm of a-Si.

The average height of the islands estimated from cross section TEM images (see figure 2(*a*)) is about $d_2 = 3.0$ nm. Unfortunately it is difficult to determine the size of islands in the plane of growth from figure 1 because of the very illegible images. But we can use the effect of conservation of Ge volume before and after island formation to estimate a mean diameter for the island D. Assuming cylindrical Ge dots we have

$$d_1 = n(\pi D^2/4)d_2 \tag{1}$$

and

$$D = \left(\frac{4}{\pi}\frac{d_1}{d_2}\frac{1}{n}\right)^{1/2}. \tag{2}$$

This implies $D = 1.9$ nm for $T_S = 400\,°C$ and $D = 1.2$ nm for $T_S = 300\,°C$.

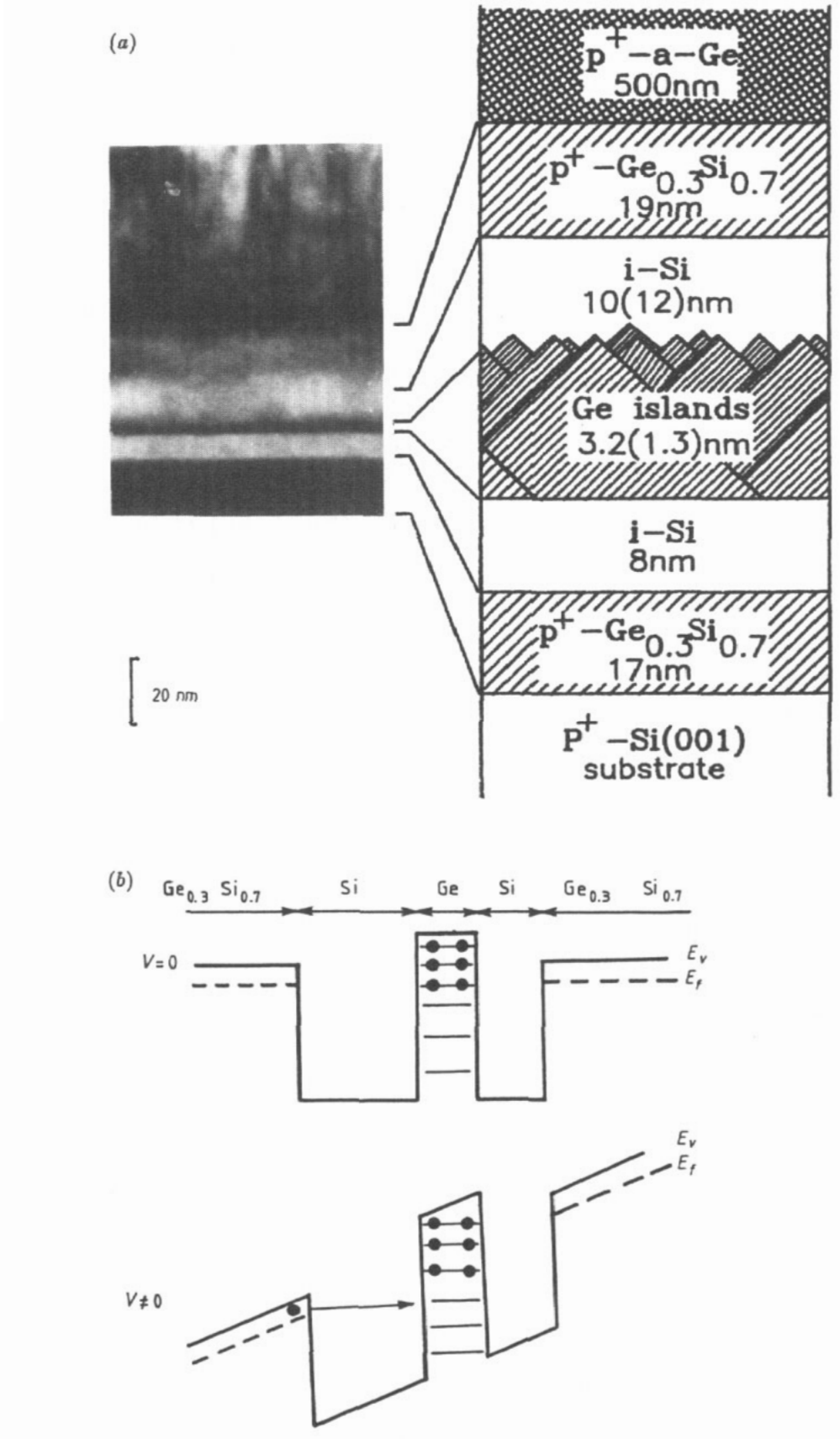

Figure 2. (*a*) A bright-field cross sectional transmission electron micrograph of one of the structures, with a schematic layer sequence; (*b*) a valence band diagram for holes under flatband (above) and biased (below) conditions. The arrow shows the hole tunnelling current.

To be convinced that the Ge film grown on Si is totally broken into islands we have fabricated the structure p^+-Ge (degenerated) on i-Si and measured the temperature dependence of conductivity in the plane of the layers. The conductivity proves not to be 'metallic', but thermally activated. This result demonstrates that there is no continuous film of Ge and we have a system with only 3D quantum confinement.

The results described in this paper were obtained for the structures p^+-$Si_{0.7}Ge_{0.3}$/Si/p^+-Ge/Si/p^+-$Si_{0.7}Ge_{0.3}$ (figure 2(*a*)). The structures consist of the following layers, in order of growth from the substrate:

(i) 17 nm $Si_{0.7}Ge_{0.3}$ base electrode doped to 10^{19} cm^{-3};
(ii) Si tunnel barrier, 10^{15} cm^{-3} (the thickness of Si B_1 has been varied from 6 to 15 nm);
(iii) layer of isolated Ge quantum dots doped to 2×10^{19} cm^{-3};
(iv) Si tunnel barrier ($B_2 = 10$–15 nm), 10^{15} cm^{-3};
(v) 19 nm $Si_{0.7}Ge_{0.3}$ counter electrode, 10^{19} cm^{-3};
(vi) amorphous Ge, 2×10^{19} cm^{-3}, as protection.

The layers were processed into mesas of diameter 200–500 μm. Indium ohmic contacts were made to the substrate and top contacts. The transverse valence band profiles for holes under flatband (above) and biased (below) conditions are presented in figure 2(*b*). The arrow shows the hole tunnelling current.

The differential transversal conductance $G = \mathrm{d}I/\mathrm{d}V$ was measured using a two-probe standard lock-in technique at a modulation amplitude ~ 1 mV and a frequency of 333 Hz.

The charge transport in these structures is associated with hole tunnelling from one p^+-$Si_{0.7}Ge_{0.3}$ electrode to the other via Ge islands as the intermediate state. In figure 3 the conductance traces are shown for three samples with different barrier thicknesses B_1 and B_2. The layer of Ge dots was grown at $T_S = 300\,^\circ$C. The measurement temperature is 4.2 K. The positive-bias direction is defined as that with holes moving from the top contact to the substrate. A sample with thinner barriers (figure 3(*a*)) exhibits conductance oscillations, which are periodic in voltage, of period $\Delta V = 80$ mV. The period of oscillation proves to be quite insensitive to the magnetic field up to 7 T.

Note that the magnitude of conductance in figure 3(*a*) approaches e^2/h. Using a single-channel two-terminal Landauer formula $G = (e^2/h)T^*$ (where T^* is the transmission coefficient) it is easy to show [11] that if

$$G \geqslant e^2/h \tag{3}$$

then

$$h\Gamma \geqslant \Delta E \tag{4}$$

where $h\Gamma$ is the width of the energy levels in quantum dots. Thus in a structure with thin potential barriers the energy levels are not sharply defined. Therefore the spectrum of dots can be considered as continuous, as in a metal. Then the periodicity of the series of conductance peaks shown in figure 3(*a*) can be understood within the Coulomb-blockade model in the classical regime [8, 11, 15] which ignores the discreteness of the electron (hole) spectrum. According to this model, changing the occupancy of the dot requires a finite charging energy. In this case the condition for the conductance peak has the form [11]

$$eV = (N - \tfrac{1}{2})e^2/C \tag{5}$$

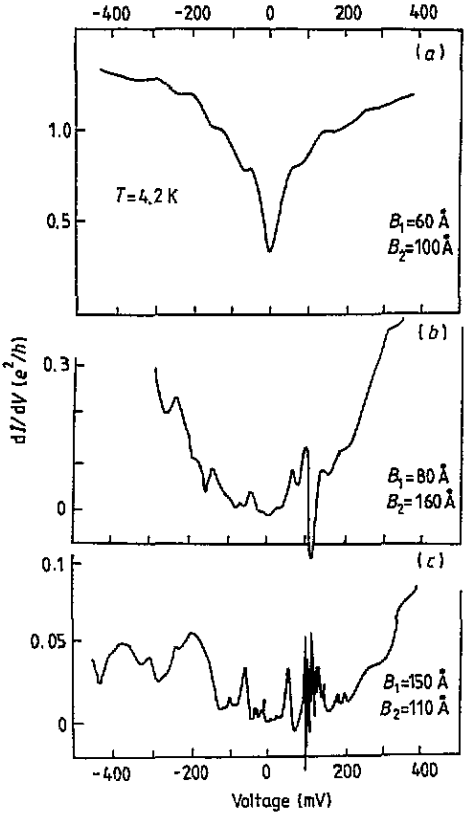

Figure 3. Transversal differential conductance versus bias voltage for samples grown at $T_S = 300\,^{\circ}C$ with different barrier thicknesses B_1 and B_2. The measurement temperature is 4.2 K. (*a*) $B_1 = 6$ nm, $B_2 = 10$ nm; (*b*) $B_1 = 8$ nm, $B_2 = 16$ nm; (*c*) $B_1 = 15$ nm, $B_2 = 11$ nm.

where N is the number of holes on a particle. The periodicity of oscillation in the classical regime ΔV follows from equation (5)

$$\Delta V = e/C. \tag{6}$$

Assuming the tunnel junction between the dots and electrode to be a circular plate capacitor of diameter $D = 1.2$ nm and thickness $B \simeq 10$ nm, and the dielectric constant for Si, $\epsilon = 11.7$, we can estimate C and hence ΔV. The capacitance per dot with a correction due to the fringing effect is given by [16]

$$C = \epsilon\epsilon_0(D/2)[(\pi/2)(D/B) + \ln(8\pi D/B) - 1]. \tag{7}$$

This yields $C = 2.5 \times 10^{-18}$ F and $\Delta V = 65$ mV, compared with the observed peak spacing of 80 mV. The level of agreement here is fine enough because of some uncertainty in the actual effective capacitance of the junctions. Thus the structure observed in figure 3(*a*) is a manifestation of the 'Coulomb staircase' effect associated with the occupation of ultrasmall capacitance by a countable number of holes. This conclusion has been supported by experiments on the temperature dependence of oscillations [6].

If the barrier thickness is larger than 10 nm, the conductance magnitude becomes much less than e^2/h (figures 3(*b*) and 3(*c*)). The traces consist of sharp peaks with irregular spacing. The distinct negative differential resistance (NDR) region appears to be near 100 mV bias. The effect of NDR is a typical feature for resonant tunnelling of carriers into quasibound states in double-barrier structures. Since the tunnel resistance is large compared with the resistance quantum h/e^2 for samples with thick barriers, the energy levels in islands are sharply defined. Therefore transport through the dots proceeds by resonant tunnelling. In the resonant tunnelling regime the energy E_N of the single-particle state affects the position and period of the Coulomb-blockade oscillations [11]. To account for the periodicity of oscillations it is necessary to add E_N into equation (5):

$$eV = (N - \tfrac{1}{2})e^2/C + E_N. \tag{8}$$

Thus

$$\Delta V_N = \Delta E_N/e + e/C. \tag{9}$$

Because of the irregular spacing ΔE_N of the single-electron levels, the conduction oscillations are irregular too. One can see from equation (9) that the charging energy lifts the spin degeneracy (put $\Delta E_N = 0$). In a plot of dI/dV versus bias voltage this must lead to a doublet structure of oscillations, with a spacing alternating between e/C and $E_N/e + e/C$. These doublets are clearly presented in a conductance trace of the sample grown at $T_S = 400\,°C$ with barrier thicknesses $B_1 = 15$ nm and $B_2 = 11$ nm (see figure 4). At a magnetic field of $B = 7$ T the positions of the conductance peaks are shifted up to ~ 2 mV and correspond to the Zeeman splitting of the heavy-hole levels in Ge quantum dots (see inset of figure 4).

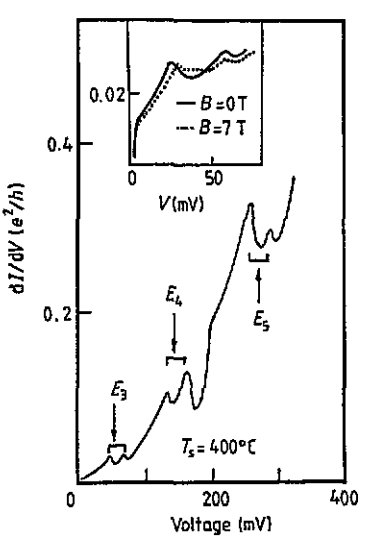

Figure 4. Transversal differential conductance versus bias voltage for a sample grown at $T_S = 400\,°C$, with $B_1 = 15$ nm and $B_2 = 11$ nm. The arrows indicate the calculated peak positions which correspond to the E_3, E_4, and E_5 bound states. Inset: conductance versus V for the device at magnetic fields $B = 0$ and $B = 7$ T.

To make not only a qualitative but also a quantitative analysis of the experimental data we have calculated the hole spectrum in a quantum dot. For the calculation we have used a simple model developed in [17], which describes resonant tunnelling in a 3D (emitter)–0D (confined well)–3D (collector) structure. Assuming a cylindrical slab form for the Ge island we can write the Schrödinger equation and the boundary condition for a hole in cylindrical coordinates (x, r, ϕ) in the hard wall approximation:

$$(-\hbar^2/2m^*)\Delta^2\psi = E\psi \tag{10}$$

and

$$\psi(r = D/2) = 0 \tag{11}$$

where ψ is the hole wave function and m^* is its effective mass in Ge. The solution of equations (10) and (11) has the form

$$E_N = E_z + (\hbar^2/2m^*)(2x_{m,l}/D)^2 \tag{12}$$

where E_z are the energy levels in the z direction, $x_{m,l}$ is the mth root of $J_l(x) = 0$ (J_l is a Bessel function), and m and l are the principal and orbital quantum numbers. Assuming a valence-band offset of 0.3 eV, we have six levels of heavy holes ($m^* = 0.34m$) E_0/E_5 for the structure grown at $T_S = 400\,°C$ ($D = 19$ nm). Three of the levels are occupied by holes from impurities and three are empty. The lowest light-hole state appears to be above the top of the barrier. Therefore, tunnelling holes injected from the $Si_{0.7}Ge_{0.3}$ region are expected to be heavy, which confirms the magnetotunnelling data. The arrows in figure 4 indicate the positions of the peaks calculated from equations (7), (8) and (12). A general consistency between the simple theory and experimental data is apparent.

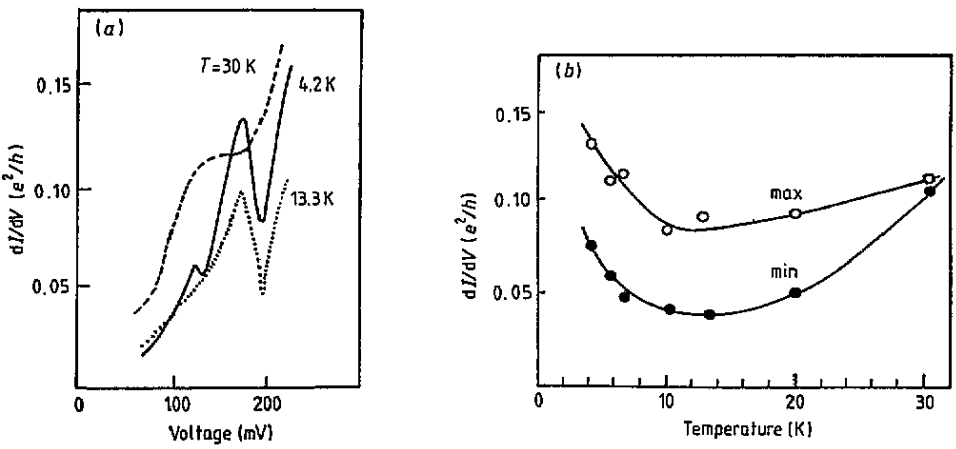

Figure 5. (*a*) The evaluation of the conductance oscillation doublet E_4 from figure 4 with temperature. (*b*) The temperature dependence of the maxima and minima of an oscillation.

The conductance oscillations with bias voltage seen in figures 3 and 4 are observed only at low temperatures. In figure 5(*a*) for doublet E_4 we show how the oscillations gradually disappear as the temperature increases. We found that the width of the peak does not

depend on the temperature below 30 K. This effect has been observed in [18] and explained by a process of electron heating by residual electromagnetic fields. The temperature dependence of the maxima and minima of oscillation is depicted in figure 5(*b*). The behaviour of the peak height at $T < 13$ K is in agreement with theoretical prediction [11]: in the resonant tunnelling regime the peak height increases as the temperature is lowered, due to the diminished thermal broadening of the resonance. Note that in the classical regime of Coulomb-blockade oscillation the peak maxima are temperature independent. The temperature dependence of dI/dV in minimum is very unusual. The conductance at the minima results from the exponential tails of the derivative of the Fermi function, and hence must be activated. Increasing the peak minima as the temperature is reduced at low temperatures is surprising, in the light of theoretical and experimental data in quantum wires [11, 18]. This large deviation from the prediction of the Coulomb-blockade model is still not understood.

In the samples with asymmetric tunnelling barriers under bias conditions, corresponding holes tunnel into the quantum well through the thinner of two barriers, current instability was observed in the region of NDR. One can see an example of such instability in figure 3(*c*) at a bias ~ 100 mV. A possible explanation for current instability is space–charge formation in the well. A large build-up of hole charge is expected at resonance when holes are injected through the thinner emitter and are inhibited from tunnelling out due to the lower transition coefficient of the thicker collector. Current through the device becomes unstable and breaks into high-frequency oscillations. Similar effects have bee observed in double-barrier structures with a 2D electron gas [19, 20]. A more detailed discussion of the current instability observed here will be published elsewhere.

In conclusion, it is necessary to notice one surprising circumstance that has been implied above. The number of experimental details, such as the magnitude of the conductance corresponding to the transition from the classical to the quantum regime of Coulomb-blockade oscillations, the peak width, and sharp conductance traces in the presence of island size variation, indicate that in spite of the large number of Ge islands in the active region of the device, the tunnel current is determined only by a single channel and flows through one 'elite' quantum dot.

Let us discuss the effect of the distribution of island size on the observed conductance structure. One can see in figure 1 that Ge particles have a very large magnitude of variation of particle size. The question is why do such clear structures, as seen in figures 3 and 4, persist in the presence of this variation. Although this problem is not fully understood at present, we suggest that the transfer of a single hole proceeds only through the electrically favourable current path. We believe that one of the possible mechanisms that selects the favourable islands is non-uniformity of the top tunnel barrier, due to a spread in particle size (see figure 6(*b*)). Let us consider a layer of particles of height h covered by Si with a thickness of B_2 (see figure 6). If Si covers the Ge islands uniformly (figure 6(*a*)) then the tunnelling distance $B = B_2$. But the situation presented in figure 6(*b*) is expected in experiment rather than in figure 6(*a*). In this case the thickness of the tunnel barrier $B = B_2 - h$ fluctuates from one particle to another. The tunnel probability is proportional to $\exp(-B\sqrt{8m^*V_0}/\hbar)$, where V_0 is the barrier height, so the current selects only islands of size large enough to provide a low resistance.

Let us assume that the island size is distributed according to the Gauss law

$$g(h) = [1/\sqrt{2}\pi(\sigma d_2)]\exp[-(h - d_2)^2/2(\sigma d_2)^2].$$

The resulting tunnel current can be determined by integrating partial currents through all the islands:

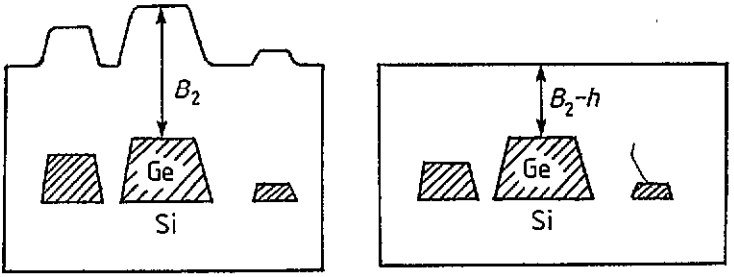

Figure 6. Two possible configurations for the Si top barrier in a structure with Ge island size variation. B_2 is the barrier thickness, and h is the particle height.

$$I \sim \int_0^\infty g(h) \exp[-A(B_2 - h)]\, \mathrm{d}h \tag{13}$$

where $A = (8m^*V_0/\hbar^2)^{1/2}$. The function which is integrated in (13) has a sharp maximum at $h^* = d_2 + \mathrm{A}\sigma^2 d_2^2$ of width $\delta = 2\sigma d_2$. Thus the main contribution to the conductance is made by islands of height h^*. The number of such 'optimal' dots in the structure of area S can be estimated from the expression

$$N^* \simeq Sng(h^*)\delta = \sqrt{2/\pi}\, Sn \exp[-(A\sigma d_2)^2/2]. \tag{14}$$

For $S = 0.02 \times 0.02$ cm^2, $n = 1.4 \times 10^{11}$ cm^{-2}, $V_0 = 0.3$ eV, $m^* = 0.34m$, $d_2 = 3$ nm, and $\sigma = 50\%$, equation (14) yields $N^* = 20$. If we take $\sigma = 60\%$, then $N^* = 0.03$. Therefore if the variation of island height exceeds 50% there are no 'optimal' islands in the sample, and its conductance is actually determined by dots whose number is about unity.

Acknowledgments

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References

[1] Pintus S M, Stenin S I, Toropov A I, Trukhanov E M and Karasev V V 1987 *Thin Solid Films* **151** 275
[2] Eaglesham D J and Cerullo M 1990 *Phys. Rev. Lett.* **64** 1943
[3] Mo Y -W, Savage D E, Swartzentruber B S and Lagally M G 1990 *Phys. Rev. Lett.* **65** 1020
[4] Aleksandrov L N, Lovyagin R N, Pchelyakov O P and Stenin S I 1974 *J. Cryst. Growth* **24/25** 298
[5] Cullis A G and Booker G R 1971 *J. Cryst. Growth* **9** 132
[6] Yakimov A I, Markov V A, Dvurechenskii A V and Pchelyakov O P 1992 *Phil. Mag.* B **65** 701
[7] Kuz'min L S and Likharev K K 1987 *Pis'ma Zh. Eksp. Teor. Fiz.* **45** 389 (Engl. Transl. 1987 *JETP Lett.* **45** 495)
[8] Mullen K, Ben-Jakob E, Jaklevic R C and Schuss Z 1988 *Phys. Rev.* B **37** 98
[9] Barner J B and Ruggiero S T 1987 *Phys. Rev. Lett.* **59** 807
[10] Reed M A, Randall J N, Aggarwal R J, Matyi R J, Moore T M and Wetsel A E 1988 *Phys. Rev. Lett.* **60** 535

[11] van Houten H, Beenakker C W J and Staring A M 1991 *Single Charge Tunneling* ed H Grabert and M H Devorted *(NATO ASI Ser. B)* (New York: Plenum)
[12] McEuen P L, Foxman E B, Meirav U, Kastner M A, Meir Yigal and Wingreen Ned S 1991 *Phys. Rev. Lett.* **66** 1926
[13] Beenakker C W J and van Houten H 1991 *Solid State Physics* vol 44 (New York: Academic) p 1
[14] Ye Qiu-yi, Tsu R and Nicollian E H 1991 *Phys. Rev.* B **44** 1806
[15] Glazman L I and Shekhter R I 1989 *J. Phys.: Condens. Matter* **1** 5811
[16] Landau L D and Lifshitz E M 1960 *Electrodynamics of Continuous Media* (New York: Pergamon)
[17] Su Bo and Goldman V J 1991 *Appl. Phys. Lett.* **58** 747
[18] Meirav U, Kastner M A, Heiblum M and Wind S J 1989-I *Phys. Rev.* B **40** 5871
[19] Goldman V J, Tsui D C and Cunningham J E 1987 *Phys. Rev. Lett.* **58** 1623
[20] Leadbeater M L, Alves E S, Sheard F W, Eaves L, Henini M, Hughes O H and Toombs G A 1989 *J. Phys.: Condens. Matter* **1** 10 605