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Voltage–capacitance and admittance investigations of electron states in self-organized InAs/GaAs quantum dots

V I Zubkov[1,3], C M A Kapteyn[2], A V Solomonov[1] and D Bimberg[2]

[1] St Petersburg Electrotechnical University 'LETI', Professor Popov street 5, 197376, St Petersburg, Russia
[2] Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstraße 36, D-10623 Berlin, Germany

E-mail: vizubkov@mail.eltech.ru

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Abstract

Emission of electrons from localized electron states in InAs/GaAs self-organized quantum dots (QDs) grown by MOCVD has been studied by a combination of steady-state voltage–capacitance and admittance techniques. We have found a fine structure of carrier concentration profile in the area of QDs, which may be attributed to ground and excited energy states of electrons in the QDs. The total range of activation energies detected in the admittance investigations extends from 20 meV up to 140 meV and testifies to the significant inhomogeneous broadening of the density of state function due to the QD scattering in geometric sizes.

1. Introduction

There is considerable interest at present in studying electronic properties of InAs/GaAs quantum dot (QD) heterostructures, suitable, for instance, for fabricating laser diodes emitting at 1.3 μm [1, 2]. Optical methods (photoluminescence, photoluminescence excitation) have commonly been used to obtain information about the optical transients between localized energy levels inside the quantum dots [1, 3, 4], but these methods do involve both electron and hole subsystems, so one cannot obtain the absolute values of quantized energy levels. Capacitance spectroscopy (and admittance spectroscopy as its modification) makes it possible to test separately either the electron or the hole emission mechanism, as well as precisely obtaining the carrier concentration profile and energy band discontinuities [5]. In this work we present the results of admittance and capacitance measurements of p–n heterostructures with self-organized InAs/GaAs quantum dots, which directly indicate the emission of carriers

[3] Author to whom any correspondence should be addressed.

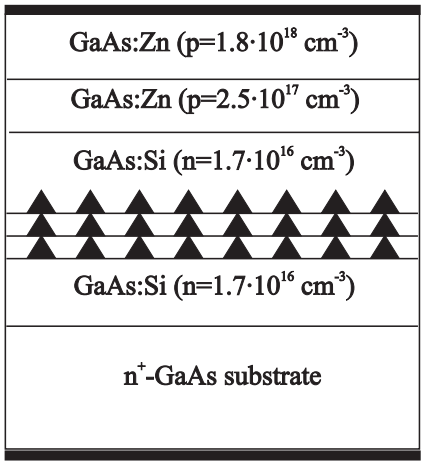

Figure 1. Layer sequence of p–n structure with self-organized InAs/GaAs quantum dots.

from ground and excited electron levels in quantum dots. In contrast with the previous DLTS investigations on the same samples [6], covered with a strong tunnelling phenomenon, the present admittance investigations in a wide range of temperatures, frequencies and biases gave the possibility to obtain accurately the activation energies for the thermionic emission from the QDs. Then the numerical integration of concentration profiles was used for deriving the total charge in the QD system.

2. Experiment

The structures under investigation were grown by metal–organic chemical vapour deposition (MOCVD) on highly n^+-doped GaAs substrate, followed by an Si-doped buffer layer. The quantum dot array consisted of three sheets of vertically coupled InAs QDs, separated by 2.8 nm thick layers of undoped GaAs. The QD sheet density was 1×10^{10} cm^{-2} as obtained from plan view TEM images. The structures were capped with a 448 nm GaAs layer, and then a 650 nm thick p-GaAs layer was deposited to create a p–n junction (figure 1). Details of growth and device fabrication can be seen in [6].

The capacitance and admittance measurements were performed using an HP4284A RLC meter in the range of frequencies 1 kHz–1 MHz and temperatures 10 K–300 K. The amplitude of the measuring signal was 10 or 50 mV.

Capacitance–voltage (C–V) characteristics of the sample exhibit a pronounced plateau in the range of reverse biases 1.0–2.5 V (figure 2) due to charge accumulation in the QDs and InAs wetting layer. The width of the plateau increases permanently when the temperature goes down to 10 K. This behaviour indicates the growth of electron charge inside the QDs with the temperature lowering because of the enhancement of energy barriers and shifting of the Fermi level. We did not observe the suppression of the plateau for temperatures below 80 K, in contrast with the investigations [7, 3] carried out on Schottky diodes with InAs/GaAs quantum dots grown by molecular beam epitaxy (MBE) and sandwiched between undoped GaAs spacers.

After ending the plateau (approximately at $|U| > 2.2$ V) a step in C–V characteristics was observed. This step corresponds to the significant energy band bending near the QDs in applied electric field, when the conditions for thermionic emission of electrons from the QDs are fulfilled. It can be quantitatively registered with the help of admittance spectroscopy during

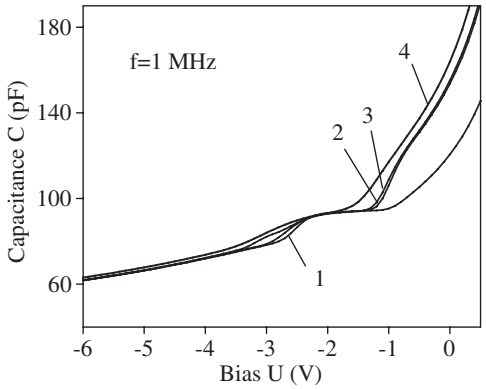

Figure 2. *C–V* characteristics of the structure with InAs/GaAs QDs at the temperatures 1–10 K; 2–45 K; 3–80 K; 4–200 K.

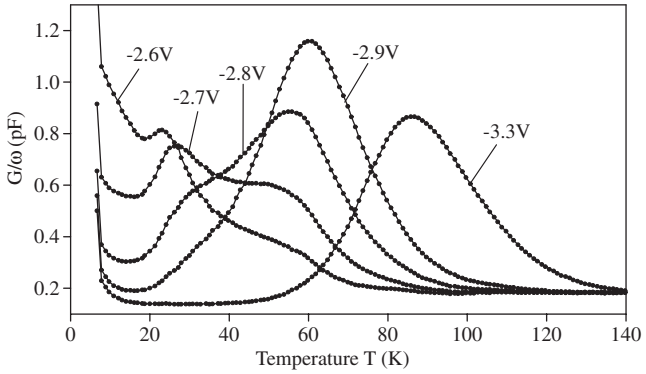

Figure 3. Temperature spectra of conductance for the p–n diode with InAs/GaAs QDs at different applied biases. Testing frequency is 1 MHz.

temperature scanning. The admittance spectra have been measured at frequencies from 1 kHz up to 1 MHz. We have detected the QD response in the range of reverse biases 2.2–3.7 V. A set of spectra for different applied biases is shown in figure 3. At high biases ($|U| > 3$ V) a single peak L1 was noticed in the conductance spectra. Its amplitude and temperature maximum had strong dependences on frequency and bias. On decreasing the reverse bias the amplitude of the conductance peak passed through a maximum at $U = -3.1$ V. At $|U| < 2.9$ V a second, additional, peak appeared at the low temperature side of the spectra, with its own frequency and bias behaviours. The amplitudes of the conductance spectra at 1 MHz are built as functions of applied bias in figure 4. One can see that the maximum of the L1 curve is four times greater than that for L2. At $U = -2.7$ V both peaks are about equal to each other.

According to [3, 8], the conductance spectrum for QDs with broadened DOS function has a maximum when the condition

$$\omega/e_{\mathrm{n}} = 2, \tag{1}$$

is fulfilled, where ω is the testing frequency and e_{n} is the thermionic emission rate of electrons from the QD

$$e_{\mathrm{n}} = e_{\infty} \exp(-E_{\mathrm{a}}/kT), \tag{2}$$

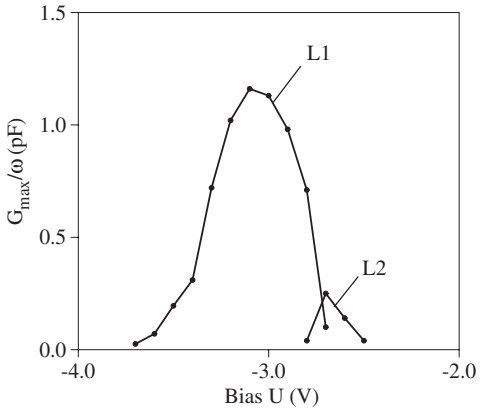

Figure 4. The magnitude of the conductance peak as a function of applied reverse bias. Frequency $f = 1$ MHz.

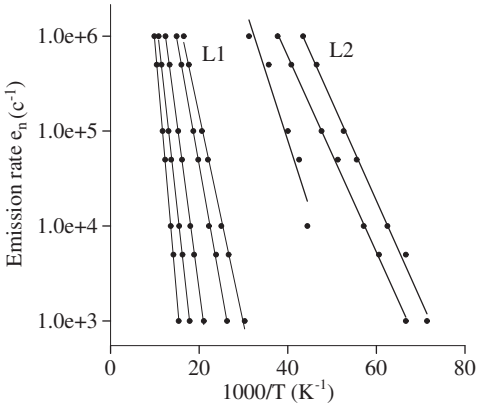

Figure 5. Arrhenius plots of the emission rates at different reverse biases.

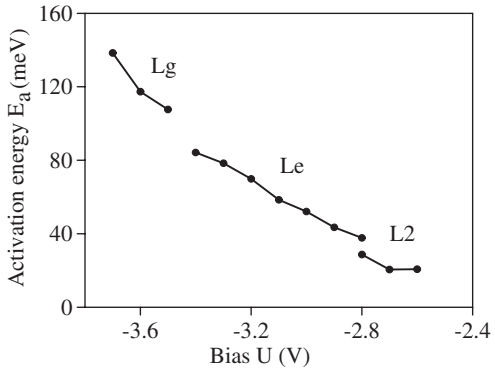

Figure 6. Dependence of 'apparent' activation energy of detected levels in the sample on reverse bias.

which depends on the activation energy E_a. The family of Arrhenius plots necessary for deriving the activation energy is depicted in figure 5.

Determined from the Arrhenius plot the 'apparent' activation energy corresponding to the peaks L1 and L2 depended considerably on U_{rev} (figure 6). The total range of detected activation energies extends from 20 meV up to 140 meV. This phenomenon is well known for DLTS (deep level transient spectroscopy) and admittance measurements of structures with quantum wells and quantum dots under electric field. The reason for this is the following. In moderately doped structures (as in the sample under consideration) in equilibrium the quantized energy levels in the QDs lie below the Fermi level because of the great magnitude of the conduction band offset in the InAs/GaAs heterosystem and a very strong 3D quantum confinement in the quantum dot. With the increase of reverse bias the penetrating external electric field bends the conduction band bottom and lifts the energy levels up, and the Fermi level crosses them one after another (figure 7). In this sense, one may interpret figure 6 as the current energy level position tested by admittance spectroscopy at given U_{rev}. Thus, in fact, the shape of L1 and L2 curves in figure 4 reflects the electron density of states (DOS) associated with the QD layer.

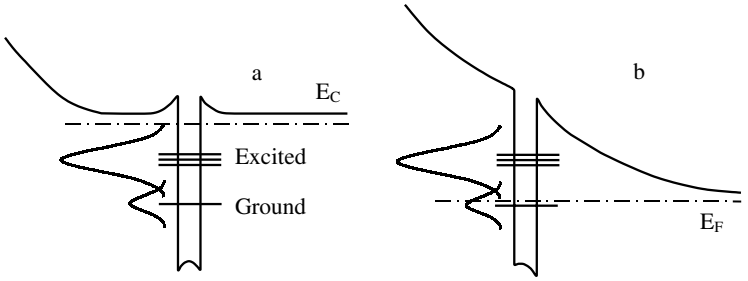

Figure 7. Scheme of the energy band structure for different biases: (a) a low reverse bias; (b) the reverse bias of about 3.5 V is applied. The dash–dotted line is the position of the Fermi quasi-level. The bound level for the quantum well is not shown.

The second reason for the field dependence of activation energy, as was pointed out by Lang [9], may be the competing mechanisms of thermal activation and thermally independent tunnelling from the level. Actually, the tunnelling mechanism did dominate in DLTS spectra of the same samples at low temperatures in [6, 10]. One should note that the activation energy E_a at the bias $U = -3.5$ V appeared to be about 105 meV in quite good agreement with the activation energy 94 meV obtained by DLTS for nearly the same bias condition [6].

Based on the physical description and interpretation, reported in previous publications [6, 10], we suggest that peak L1 actually reflects the carrier emission from the QD ground (Lg) and excited (Le) levels, depending on the external bias. The blending of responses from the ground and excited states due to the QD ensemble fluctuations in the energy levels makes it impossible to resolve these peaks clearly in the conductance spectra, but the step from 110 meV towards 80 meV in the apparent activation energies near -3.5 V as well as unordinary behaviour of the left-hand side of the L1 peak (see figure 4) do testify to the benefit of such a statement.

We guess, furthermore, that the L2 peak appears because of the carrier emission from the InAs wetting layer. The activation energy of about 20 meV for this process is in good agreement with photoluminescence studies [11].

Numerical calculation of the energy level scheme for the present QD geometry was performed in [12] on the basis of eight-band **k·p** theory [13]. By this method, a ground state with energy $E_0 = -195$ meV relative to the GaAs conduction band edge, and a set of three almost degenerate excited states at $E_{1,2} = -115$ meV and $E_3 = -111$ meV, were obtained. In addition, the energy of the first sub-band of the triple quantum well was determined by one-dimensional self-consistent calculation of the Schrödinger equation including strain and was equal -59 meV [14]. The effects of size and shape fluctuations were not taken into consideration during the calculations.

Despite the significant difference of the experimental values from the calculated ones, on the whole the above mentioned level scheme matches unambiguously the three parts of the curve in figure 6. We guess that the ground state has an average activation energy $E_0 = 120$ meV and is inhomogeneously broadened within ± 20 meV due to quantum dot size fluctuations. Further, the broad line Le in figure 6 reflects the emission from three excited levels E_1, E_2 and E_3 (and even more, bearing in mind that QDs with bigger lateral sizes have more closely positioned energy levels). Each state is inhomogeneously broadened in the QD array, resulting in the spreading of Le activation energies from 40 to 80 meV. From this, one may roughly estimate the average energy gap between the ground and first excited states as 60 meV, which is in reasonable agreement with the calculated energy level scheme.

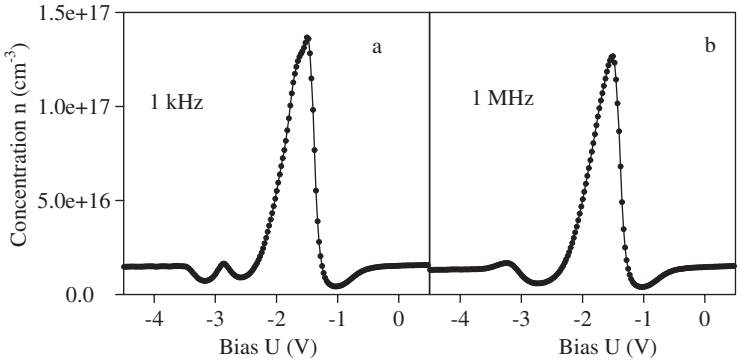

Figure 8. N–U characteristics of the structure with InAs/GaAs QDs at different frequencies. $T = 45$ K.

At low temperatures (<40 K), in addition, we have determined the dopant-related admittance signal. This peak has been detected in the range of small biases $+0.8 \cdots -1.5$ V and its intensity was approximately 20 times greater than from the QD-related peak. The origin of this peak is, obviously, the major carrier freeze-out effect [15] resulting in great enhancement of substrate resistance and, hence, in changing the resonance conditions for the equivalent circuit of the RLC meter.

One more point to be discussed is the presence of three layers of vertically stacked QDs in the sample. As was shown in [16], the ground state of a double-QD system should have a 'red shift' in energy under electric field, which would result in the increasing of its activation energy. The comparison of our data with those presented in [3], where the heterostructure with only one QD layer was investigated, demonstrates the practical similarity in behaviour of activation energies against bias; that is, one should conclude that the above mentioned effect is less pronounced than the thermionic activation phenomenon in the admittance spectroscopy of self-organized quantum dots with broadened density of states.

When calculating the apparent concentration profile $n = f(w)$ using conventional differentiation of C–V characteristics we found a fine structure of the concentration peak in the vicinity of the QD layer (figure 8(a)). (The plots are built in coordinates $n - U$, which is more revealing in this case due to the nonlinear dependence $w = f(U)$.) The fine structure was detected only at low frequencies in the range of temperatures $40 \cdots 80$ K (in contrast see figure 8(b), where no fine structure was observed). The ability to observe a peak depends on the correlation between the carrier emission rate from the corresponding level (2) and the frequency of the probing signal. When the testing frequency becomes much greater than the emission rate e_{n}, the carriers cannot follow the measurement signal. The origin of the weak peak, positioned near $U = -2.8$ V at low frequencies (figure 8(a)), and the large peak we attribute to the ground and excited energy states in the QD array, as established in [17].

The area under the curve $n = f(w)$ qualitatively reflects the total charge accumulated in the QDs. Because of the essentially indirect procedure of deriving the concentration profile from C–V-measurements, the apparent profile near a quantum well or quantum dot layer differs from the true majority carrier profile in the same structure [5] (figure 9). Nevertheless, as was shown analytically by Kroemer [18] for the case of a heterojunction, the total amount of charge is conserved in the smearing process. So, by numerically integrating the area under the curve $n = f(w)$ or $n = f(U)$ in figure 8 one can obtain the total charge accumulated in the QDs. The boundaries for the integration should be the coordinates where the concentration

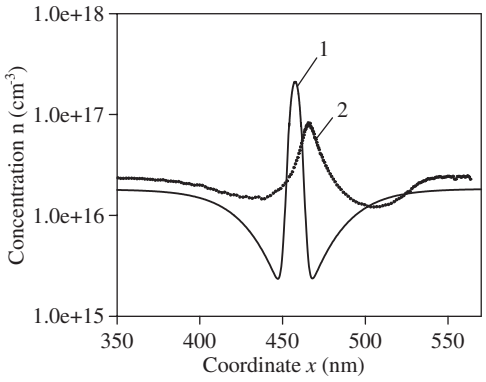

Figure 9. 'True' (1) and 'apparent' (2) carrier concentration profile near a QD. The true majority carrier profile is calculated in a 1D approximation [5].

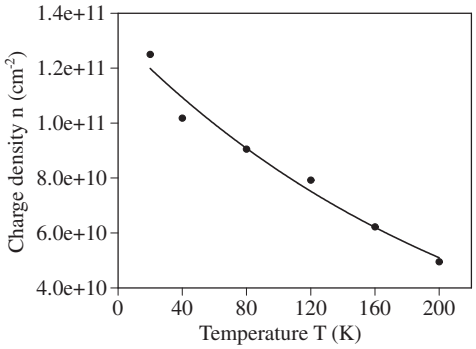

Figure 10. The sheet density of charge accumulated in the QD + wetting layer system at different temperatures. Testing frequency $f = 100$ kHz.

reaches minima. The curve plotted in figure 10 exhibits the increasing amount of charge in the wetting layer + QD system with decreasing temperature in accordance with the permanent growth of the plateau width in C–V characteristics (see figure 2). At 200 K the sheet density of charge is about 5.0×10^{10} cm^{-2} and attains 1.3×10^{11} cm^{-2} at 20 K.

Concerning the correlation of the sheet density of charge with respect to the QD density one should say the following. The total amount of charge in the three-layered system QD + QW is the sum accumulated at the ground and excited states of the QDs and stored in the triple quantum well. According to the quoted numerical simulation, there exist a ground state and three excited states (and a few more bound excited states with considerably smaller localization energies). Each energy state is twice degenerated. Hence, at low temperatures we can expect the value of charge formed by eight electrons (actually even more) of the triple QD plus charge stored in the QWs. The measurements carried out earlier on the reference sample having only three wetting layers without any quantum dots [12] found a QW charge density of about 7×10^{10} cm^{-2} at 75 K. In the sample where both QD and QW exist, as follows from our present study, the role of the wetting layer as a reservoir for storing charge is somewhat lower (the amplitude of the L2 curve is one-quarter that of L1; see figure 4). Therefore, the sheet density of charge obtained in the experiment is in reasonable accordance with the QD density (1×10^{10} cm^{-2}) and does not contradict the above mentioned energy level scheme.

3. Conclusions

The emission of electrons from ground and excited energy levels in InAs/GaAs self-organized quantum dots grown by MOCVD has been studied by means of the steady-state capacitance versus voltage technique and admittance spectroscopy. Two peaks have been obtained in the carrier concentration profile and in conductance spectra, which are attributed to the ground and excited energy states of electrons in the QDs. They are distinguished from the peak in the conductance spectra related to the emission from the InAs wetting layer. The total range of activation energies detected in the admittance investigations extends from 20 meV up to 140 meV and testifies to the significant inhomogeneous broadening of density of states due to the QD scattering in geometric sizes [19]. The dependence of conductance spectra magnitude on applied bias observed in the measurements can be treated as a measure of the broadened density of energy state function for self-organized quantum dots. The total charge accumulated in the wetting layer + QD system was derived by numerical integration of the area under the carrier concentration curve. At 200 K the sheet density of charge is approximately 5.0×10^{10} cm^{-2} and increases about two-and-a-half-fold at 20 K.

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