Electrically modulated photoluminescence in self-organized InGaAs/GaAs quantum dots

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Results of photoluminescence (PL) study of the self-organized InGaAs/GaAs quantum dots (QDs) in a field-effect structure grown by metalorganic vapor phase epitaxy are presented. It has been found that the PL from the QDs strongly depends on the bias voltage. No PL from the QDs ground state can be observed from the reverse biased structure, whereas the PL signal recovers in the forward biased structure. It is proposed that the bias dependence of the PL signal results from the QDs electron occupancy changes driven by the electric field within the structure. Due to a long thermalization time, the photogenerated electrons are swept out of the QDs by the electric field before radiative recombination. The electrically modulated PL (e-m PL), making use of the bias dependence of PL signal, is proposed as a tool for QD investigation. The e-m PL spectra at $T=300$ and $T=4.2$ K are analyzed and discussed. [S0003-6951(98)02345-6]

There is a growing interest in the quasizero dimensional structures—so-called quantum dots (QDs). Confining the electronic motion in all three dimensions results in a discreet, atomic-like electronic density of states.[1–3] The physics of QDs provides an ultimate test for the quantum mechanics. The QDs proved their usefulness in many optoelectronic applications.[4–6] High electric field in those structures influences quantum states within the QD and can induce changes in the QD's electron occupancy.[7] Charging of QDs with electrons can be controlled in field-effect structures by applied bias voltage. The field-effect structures with large arrays of self-organized QDs have been successfully used for investigation of the QD's basic properties by means of capacitance[8] and absorption measurements.[9] Quite recently the effect of the QD's occupancy on the photoluminescence (PL) from the InAs QDs has also been reported.[10] The study of this effect is a main topic of the present letter.

The sample investigated in this work was grown on [100] semi-insulating (SI) GaAs substrate using a low pressure metal organic vapor phase epitaxy (MOVPE). The QDs were grown in the Stranski–Krastanow mode. An epitaxial $In_{0.6}Ga_{0.4}As$ layer was deposited at the growth temperature of 550 °C and a V/III ratio of 540. The details of the growth procedure were previously published.[11] The surface topography of a control sample with $In_{0.6}Ga_{0.4}As$ layer grown at the top of a structure was examined using an atomic force microscopy (AFM). It was found that the lens shaped islands of average basal diameter 34 ± 5 mm were formed with a surface density equal to 7.1×10^{9} cm^{-2}. The $In_{0.6}Ga_{0.4}As$ layer within the structure has been grown under the same conditions. The sample structure is schematically shown in Fig. 1. It consisted of 200 nm of Si doped ($n=2\times10^{18}$ cm^{-3}) GaAs acting as a back contact, followed by 20 nm of undoped

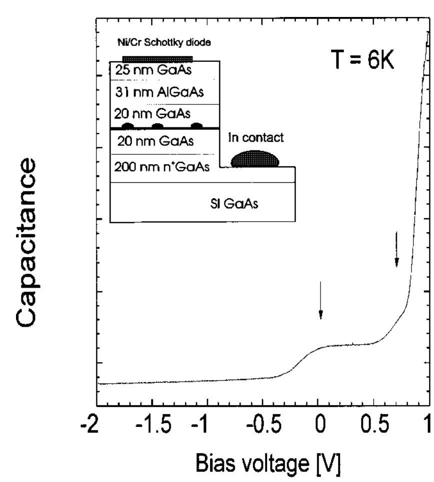

FIG. 1. The capacitance–voltage characteristics of the investigated sample at $T=6$ K (filling of the ground and the excited states within the QDs is marked with arrows). The schematic sample structure is presented in the inset.

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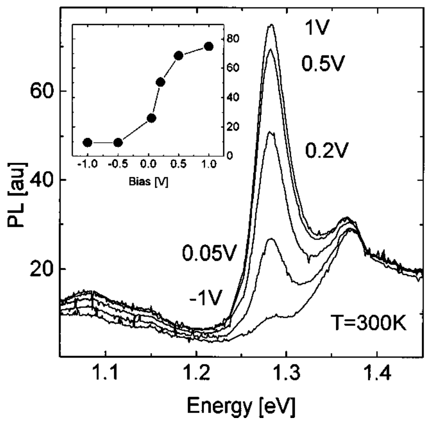

FIG. 2. The photoluminescence spectrum of the biased structure with InGaAs/GaAs quantum dots measured at $T=300$ K. The bias dependence of PL intensity at 1.28 eV is shown in the inset.

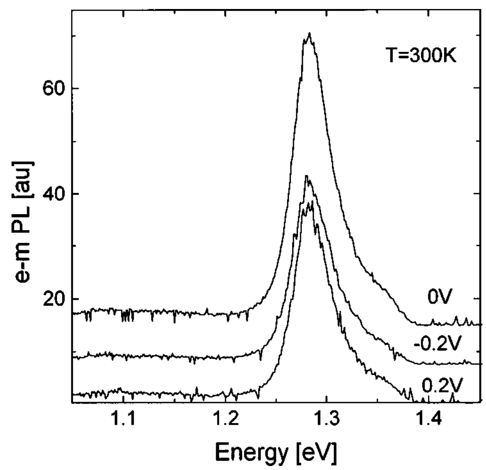

FIG. 3. The electrically modulated photoluminescence spectra at $T=300$ K as a function of bias. The amplitude of modulation signal was equal to 100 mV (for clarity offset added).

GaAs tunneling barrier, the $In_{0.6}Ga_{0.4}As$ layer, the 20 nm undoped GaAs spacer, 31 nm $Al_{0.2}Ga_{0.8}As$ barrier, and 25 nm GaAs cap. The actual layer thicknesses have been found by cross-sectional transmission electron microscopy (TEM) imaging. At the top of the sample, circular (1.5 mm diameter) semitransparent (30 nm thick) Ni/Cr Schottky contact was evaporated through a shadow mask. The back gate was formed by alloying an In contact at the top of n^+ GaAs layer after part of the sample had been electrochemically etched off. The PL measurements have been performed with the sample cooled in a continuous flow Oxford CF-1204 cryostat. Bias dependent PL measurements were taken with a laser illumination from the top of a sample through a Schottky contact. A semiconductor laser ($\lambda=780$ nm) has been used for PL excitation. PL spectra were dispersed by a double-grating SPEX spectrometer and detected with a liquid nitrogen-cooled Ge p-i-n diode.

The capacitance–voltage $(C-V)$ characteristic of the investigated sample measured at low temperature is shown in Fig. 1. The constant capacitance for bias lower than $U=-0.5$ V reflects a geometrical capacitance between the top gate and the n^+ GaAs layer. With increasing bias voltage the QD's ground state can be charged with electrons and the threshold of the capacitance at $U=-0.2$ V (see Fig. 1) is attributed to this effect. A charging energy e^2/C of a disk-shaped dot of $d=34$ nm$(C=4\epsilon\epsilon_0 d)$ embedded within bulk GaAs can be estimated as 10 meV. The charging energy induced splitting of the QD's ground state cannot be observed most likely due to low ''lever arm'' factor[7] and the size distribution of the dots. The next feature in the $C-V$ characteristic seen at $U=0.7$ V is probably due to the filling of the QD's excited state or the formation of a two-dimensional electronic gas (2DEG) in the $In_{0.6}Ga_{0.4}As$ wetting layer (WL). Large capacitance increase at $U=0.8$ V is attributed to the 2DEG formation at the AlGaAs/GaAs interface.

Two characteristic features can be seen in the PL spectra from the biased structure at room temperature (see Fig. 2). The main peak at $E=1.28$ eV is attributed to the PL from the ground state of the QDs. The origin of the feature at 1.37 eV is not fully clear. The applied bias has a different effect on both peaks. The PL signal due to the QDs can be almost completely quenched under reverse bias (see inset to Fig. 2).

The bias dependence of the PL signal can be used for electrically modulated (e-m) experiment. In that experiment the structure is continuously illuminated with a laser beam, and it is biased with a sum of dc bias U_0 and ac modulation signal ΔU. The latter ac voltage ($f=18$ Hz, $\Delta U=0.1$ V) is also used as a reference for a lock-in amplifier. The e-m PL signal depends on the radiative recombination processes sensitive to the applied bias and it can be regarded as a derivative of the PL signal on the bias. The use of e-m PL can clearly reveal the bias dependence of the PL spectrum.

The e-m PL spectra measured at room temperature at several bias voltages are presented in Fig. 3. The main PL peak at 1.28 eV can be identified in the e-m PL spectrum. The maximum intensity of this peak was found at $U_0=0$ V, which corresponds to the largest slope of the PL dependence on the bias (see inset to Fig. 2). No energy shift of the QD's PL peak with an applied bias can be observed between $U_0=-0.2$ V and $U_0=0.2$ V. The e-m PL peak's high energy shoulder can be due to recombination from excited states within the QDs, however its origin is not fully clear. The feature at 1.37 eV seen in the PL from biased structure is no longer observed in the e-m PL spectrum. This reflects the independence of this luminescence on the bias.

On the contrary, the e-m PL spectra recorded at $T=4.2$ K strongly depend on the bias voltage (see Fig. 4). The bias driven changes of PL intensity at $T=4.2$ K are observed between $U=-0.6$ V to $U=0.1$ V, which is the bias range of the QDs charging, seen in the $C-V$ characteristic (see Fig. 1). It can be seen that the higher bias voltage the higher the e-m PL peak energy. Moreover, at the highest bias the lower energy part of e-m PL spectrum is negative. This reflects the decrease of PL signal with increasing bias at this spectral region.

An explanation to the bias induced quenching of the QD's PL signal would be the spatial separation of electron and hole wave functions due to the quantum confined stark effect (QCSE).[12,13] The QCSE could also be responsible for a systematic blueshift of the e-m PL peak as a function of applied bias observed at low temperature (see Fig. 4). However, it is difficult to explain why no PL energy shift can be

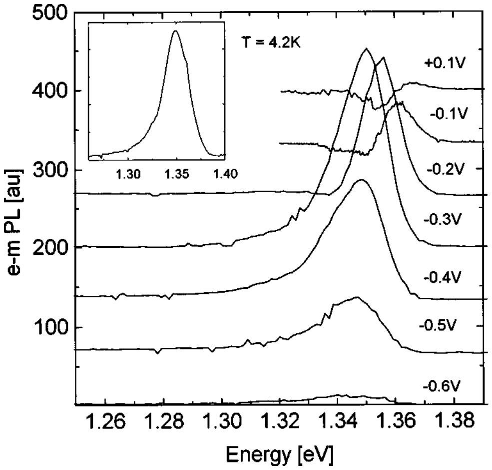

FIG. 4. The electrically modulated photoluminescence at $T=4.2$ K as a function of sample bias. The amplitude of modulation signal was equal to 100 mV (for clarity offset added). The PL spectrum from unbiased structure is shown in the inset.

seen at room temperature (see Fig. 2). We propose that the bias dependence of the QD's PL peak results from the electron occupancy changes within the structure driven by the bias voltage. At reverse bias the QD's ground state is above the Fermi energy and no electrons occupy the QDs. Both photogenerated electron and hole must be trapped onto the QD in order to recombine radiatively. However, due to relatively weak phonon scattering within the QD,[14,15] photogenerated electrons are removed from the dots by the electric field before thermalization to the ground state. As a result, no radiative recombination within the QDs is possible. In a positively biased structure the QDs are filled with electrons. Radiative recombination of an exciton can take place after a photoexcited hole is trapped onto the QD. The model proposed can be also applied to our e-m PL results. The systematic blueshift of e-m PL peak as a function of increasing bias observed at low temperature results in our opinion from the statistical distribution of dot sizes. At large negative bias only the QDs, which have the lowest electron energy levels (''biggest sizes'') are occupied with electrons and the e-m PL from those dots can be observed. Increasing the Fermi energy in the QDs plane with a bias voltage results in population of ''smaller dots,'' of higher energy levels. In such a way the radiative recombination at higher energies can take place and the blueshift of the e-m PL signal can be observed. A negative e-m PL signal at the highest positive bias voltage reflects the decrease of PL intensity with an applied bias. Decrease of the PL intensity from the QDs of lowest energy levels may be due to an increase of recombination efficiency from the excited states within the QDs. The attribution of PL bias dependence to the steady electron occupancy of the QDs is in agreement with the $C-V$ characteristics of investigated sample. Results of e-m PL measurements at room temperature can also be explained in terms of QD size distribution. Due to a thermal broadening of the Fermi distribution, the electron occupancy of QDs is less sensitive to the bias applied and no shift of the e-m PL can be seen.

The PL quenching investigated in our experiment has been explained as an effect of electric field on photogenerated electrons. We believe that in undoped structures, the built-in electric field is substantially lower and the carrier relaxation into the QDs is not affected. The PL from the QDs in such structures can be observed, which was experimentally confirmed for the InGaAs/GaAs QDs similar to those investigated in this work.[16]

In conclusion, the measurements of the QD PL from the field-effect structure have been reported. It has been found that PL intensity strongly depends on the QD electron population controlled by the applied bias. The PL signal can be completely quenched in QDs with no steady electron population and it recovers after filling of QDs with electrons. The e-m PL technique has been proposed as a method of QD investigation.

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[1] R. C. Ashoori, Nature (London) **379**, 413 (1996).
[2] S. Fafard, R. Leon, D. Leonard, J. L. Merz, and P. M. Petroff, Phys. Rev. B **50**, 8086 (1994).
[3] U. Meirav and E. B. Foxman, Semicond. Sci. Technol. **10**, 255 (1996).
[4] N. Kirstaedter, N. N. Ledentsov, M. Grundmann, D. Bimberg, U. Richter, S. S. Ruvimov, P. Werner, J. Heydenreich, V. M. Ustinov, M. V. Maximov, P. S. Kop'ev, and Zh. I. Alferov, Electron. Lett. **30**, 1416 (1994).
[5] D. Klotzkin, K. Kamath, and P. Bhattacharya, IEEE Photonics Technol. Lett. **9**, 1301 (1997).
[6] S. Fafard, K. Hinzer, S. Raymond, M. Dion, J. McCaffrey, Y. Feng, and S. Charbonneau, Science **274**, 1350 (1996).
[7] H. Drexler, D. Leonard, W. Hansen, J. P. Kotthaus, and P. M. Petroff, Phys. Rev. Lett. **73**, 2252 (1994).
[8] G. Medeiros-Ribeiro, D. Leonard, and P. M. Petroff, Appl. Phys. Lett. **66**, 1767 (1995); K. H. Schmidt, G. Medeiros-Ribeiro, J. Garcia, and P. M. Petroff, *ibid.* **70**, 1727 (1997); G. Medeiros-Ribeiro, F. G. Pikus, P. M. Petroff, and A. L. Efros, Phys. Rev. B **55**, 1586 (1997).
[9] R. J. Warburton, C. S. Durr, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro, and P. M. Petroff, Phys. Rev. Lett. **79**, 5282 (1997).
[10] K. H. Schmidt, G. Medeiros-Ribeiro, and P. M. Petroff, Phys. Rev. B **58**, 3597 (1998).
[11] C. Lobo and R. Leon, J. Appl. Phys. **83**, 4168 (1998); R. Leon, T. J. Senden, Y. Kim, C. Jagadish, and A. Clark, Phys. Rev. Lett. **78**, 4942 (1997).
[12] M.-E. Pistol, D. Hessman, J. Lindahl, L. Montelius, and L. Samuelson, Mater. Sci. Eng., B **42**, 82 (1996).
[13] S. A. Empedocles and M. G. Bawendi, Science **278**, 2114 (1997).
[14] H. Benisty, C. M. Sotomayor-Torres, and C. Weisbuch, Phys. Rev. B **44**, 10945 (1991).
[15] I. Vurgaftman, Y. Lam, and J. Singh, Phys. Rev. B **50**, 14309 (1994).
[16] R. Leon, C. Lobo, R. Bozek, A. Wysmolek, A. Kurpiewski, M. Kaminska, T. P. Chin, and J. M. Woodall (unpublished).