Observation of built-in electric field in InP self-assembled quantum dot systems

V. Davydov,[a] I. Ignatiev,[b] H.-W. Ren, S. Sugou,[c] and Y. Masumoto[d]
Single Quantum Dot Project, ERATO, JST, Tsukuba Research Consortium, Tsukuba 300-2635, Japan

(Received 4 May 1998; accepted for publication 16 March 1999)

Strong Franz–Keldysh oscillations were observed in the nonlinear reflection spectra of heterostructures with InP self-assembled quantum dots. These oscillations manifest a built-in electric field of about 30 kV/cm. We propose that this field originates from electric charge captured by the intrinsic defects on the dot interface. The presence of acceptor-like intrinsic defect states is found to be a general feature of the InP/InGaP interface but was not observed in other structures with quantum dots such as InAs/GaAs.
[S0003-6951(99)02920-4]

Self-assembled quantum dots (QDs) fabricated by epitaxial growth in the Stranski–Krastanov mode have been recently a subject of extensive studies.[1–4] The fabrication of the QDs in a single growth process prevents the QD surface from strong contamination. Nevertheless, the structure and properties of the interface between the QDs and the barrier layer are still poorly understood. The lattice mismatch between the materials of the QDs and the barrier layers (which itself governs the formation of the QDs) gives rise to strain around the QDs. For InAs QDs, this problem was investigated by Grundmann *et al.*[4] and for InP QDs by Pryor *et al.*[5] The strain can generate local intrinsic defects around the QD/barrier layer interface. Due to the small size of the dots, the fraction of the surface atoms to the core atoms is relatively large so that the interface defects can affect considerably the properties of the QDs. However, up to now there are no experimental data about these defects and their influence on the properties of the QDs.

In this letter, we present the experimental evidence of a built-in electric field in the self-assembled InP QD system. We found that a large amount of presumably negative electric charge is trapped near the QDs. We suppose that the charge is captured by intrinsic defects around the interfaces between the InP QD layer and the InGaP barrier layers. This process leads to a built-in electric field that causes intense Franz–Keldysh oscillations in the photoreflection (PR) and electroreflection (ER) spectra. For the detailed study of PR, we used the pump-probe method because of its high sensitivity and the possibility to study the time evolution of processes.

The studied heterostructures were grown on n^+ GaAs (100) substrates by the gas source molecular beam epitaxy. The 300 nm GaAs buffer layer with a short period AlAs/GaAs superlattice in the middle was grown (at 600 °C) to suppress dislocations. The 2 nm AlAs layer grown on the buffer layer prevents the compositional interfusion between GaAs and the $In_{0.51}Ga_{0.49}P$ barrier layer. One layer of InP QDs with nominal thickness of 4 monolayers (ML) was grown between the $In_{0.51}Ga_{0.49}P$ barrier layers. The growth rate was 0.5 ML/s for InGaP and 0.25 ML/s for InP. The interruption times used before and after the InP growth were 2 and 20 s, respectively. The QDs sizes were controlled by choosing the growth temperature in the range 480–500 °C. The structure of the samples is schematically drawn in the inset of Fig. 1.

Three heterostructures were studied most thoroughly. The average sizes of QDs (mean diameter d and height h) and the distance between them were determined by atomic force microscopy in the reference structures, without the top barrier layer, grown in the same conditions. They are $d = 40$ nm, $h = 10$ nm for QDO1504; $d = 60$ nm, $h = 5$ nm for QDO1505; $d = 50$ nm, $h = 10$ nm for QDP1779. The average distance between the QDs is about 100 nm for all the three samples. The InGaP barrier layer thicknesses are 100 nm for the sample QDP1779 and 150 nm for the samples QDO1504 and QDO1505.

The sample QDO1505 was studied by a high resolution transmission electron microscopy.[6] The cross-sectional image clearly displays a regular crystal structure of the QDs

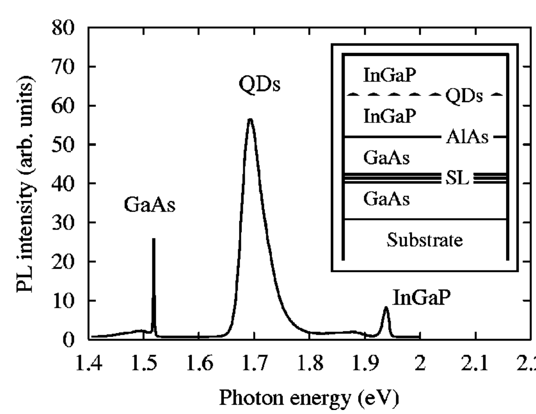

FIG. 1. PL spectrum of the sample QDP1779. ''QDs,'' ''InGaP,'' and ''GaAs'' mark PL of the QDs and of the excitons of the barrier and buffer layers, respectively. Inset: structure of the studied samples.

[a]On leave from S. I. Vavilov State Optical Institute, St. Petersburg, Russia; electronic mail: val@sqdp.trc-net.co.jp
[b]On leave from St. Petersburg State University, St. Petersburg, Russia.
[c]Opto-Electronics Research Laboratories, NEC Corporation, Tsukuba, 305-0841, Japan.
[d]Institute of Physics, University of Tsukuba, Tsukuba, 305-8571, Japan.

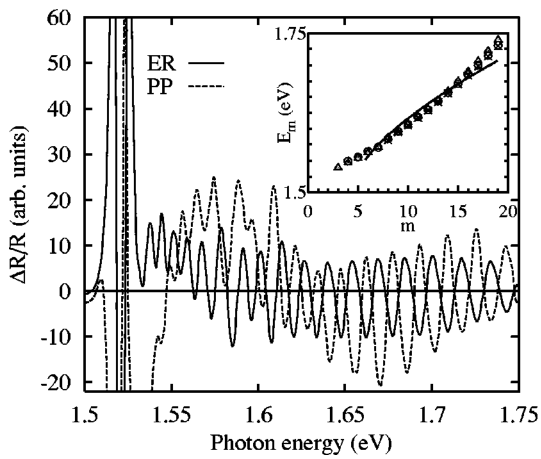

FIG. 2. Pump-probe (PP) and electroreflection (ER) spectra of the sample QDP1779. Inset: the energy position of FKO maxima vs their numbers for the samples QDO1504 (○), QDO1505 (×), and QDP1779 (△). Solid line is fitting by formula (1) with a parameter $F=30$ kV/cm.

and the barrier layers that confirms a coherent growth of the heterostructure.

The photoluminescence (PL) spectra were measured at the temperature 1.6 K under a cw laser excitation ($\lambda=532$ nm). A typical PL spectrum is shown in Fig. 1. The spectral band of the QDs is most intense in the spectra for all the structures studied, and this indicates the high quality of the samples. The spectral position of the band maximum which depends on the QDs size is 718 nm for QDO1504, 742 nm for QDO1505, and 733 nm for QDP1779. The band width at the level of 10% is about 40 nm. The QDs PL intensity depends linearly on the pump power up to 10 W/cm^2.

Pump-probe experiments were performed at 1.6 K in the reflection geometry. The setup includes a femtosecond Ti:sapphire laser (frequency 82 MHz, pulse duration 0.1–1 ps) which is tunable from 720 nm to 850 nm. The amplitude modulation of the pump and probe beams at different frequencies (100 and 2 kHz) and a double lock-in detection of the signal modulated at the differential frequency allows us to avoid noises from the scattered light and to detect fractional reflection changes as low as 10^{-7}. To eliminate the interference between the pump and the probe beams the optical frequency of the pump beam was shifted by an acousto-optical modulator. Time dependence of the pump-probe signal was measured by scanning the optical delay of the probe pulses. For measuring the spectral dependence of the signal, the wavelength of the Ti:sapphire laser was continuously tuned and measured by a wavelength meter.

The time dependence of the measured signal is the subject of a separate study.[7,8] Here we discuss only the spectral dependence of the signal in detail.

The spectral dependence of the pump-probe (PP) signal of the sample QDP1779 for 60 ps delay between the pump and the probe pulses is presented in Fig. 2. This dependence shows strong oscillations in a wide spectral region. The spectra of the other samples reveal a similar behavior with approximately the same period of oscillations. The energy positions of the oscillation maxima in the PP spectra of all the structures studied are shown in the inset of Fig. 2. All experiments led us to the conclusion that we observed the

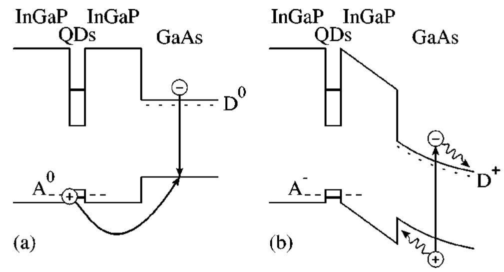

FIG. 3. Charge transfer during the structure growth (a) and the optical pumping (b).

Franz–Keldysh oscillations (FKO). To confirm this conclusion, we supplied the sample QDP1779 with electric contacts and recorded the ER spectrum with an electric field modulation. It is also presented in Fig. 2. One can see oscillations with the same period as is seen in the PP spectrum.

FKO provide clear evidence for the built-in electric field in the heterostructures with InP QDs. We studied the PP spectra of structures without QDs (a sample with a 600 nm InGaP layer on a GaAs buffer, another one with a GaAs quantum well between InGaP barrier layers) and also structures with InAs QDs between GaAs barrier layers. Spectra of all these structures do not have any regular oscillations. Therefore, the existence of the built-in electric field is caused by the presence of InP QDs in the structure.

We offer the following model of the energy structure and physical processes which give rise to the built-in electric field. The model is schematically shown in Fig. 3. Due to strain, the region around QD/InGaP interface contains a number of intrinsic defects which act as the carrier traps. For the explanation of the experimental data, we should suppose that these traps are predominantly acceptors (A^0). During the growth process, the high temperature ($\approx 500\,°$C) provides enough energy to the holes in the QDs layer to jump over the potential barrier and to reach the GaAs layer. The GaAs buffer layers of the investigated structures contain donors (D^0) whose density is of the order of $10^{15}\,\mathrm{cm}^{-3}$. The holes recombine with the electrons produced by these donors. This process is shown in Fig. 3(a) by arrows. The charge transfer can occur also through the thermal activation of the donor electrons from GaAs to the QDs where they recombine with acceptors. As a result of both processes, an excess negative charge (A^-) occurs in the QDs layer and an excess opposite charge (D^+) in the buffer layer. This results in a double electric layer with an electric field inside. The electric field penetrates into the GaAs buffer layer due to the small donor concentration in it.

This model is capable of explaining the main features of the signal. Pump pulses produce free carriers in the GaAs buffer layer. Their motion shown by arrows in Fig. 3(b) changes the built-in electric field inside this layer. The change of the electric field causes the change of the optical properties of the heterostructure. The probe pulse detects mainly the reflectivity of the GaAs buffer layer because its optical thickness is much larger than that of the QD layer. That is why the observed regular oscillations start from the spectral position of the GaAs bulk exciton.

In order to determine the value of the built-in electric field, we utilized the approximation

$$E_m = E_g + \left(\frac{e^2\hbar^2F^2}{2\mu}\right)^{1/3}\left[\frac{3}{4}(m\pi-\varphi)\right]^{2/3} \quad (1)$$

given by Aspens and Studna.[9] Here E_m is the energy position of the mth FKO maximum, $E_g = 1.52$ eV is the GaAs band gap, φ is a fitting parameter, F is the electric field, and $\mu = 0.058m_e$ is the reduced mass, where m_e is the electron mass.

The calculated curve E_m is shown in the inset of Fig. 2. There is a discrepancy between the experimental data and the fitting curve E_m. Therefore, we can only approximately estimate the magnitude of the field to be about 30 kV/cm. We suppose that this discrepancy is caused by the inhomogeneity of the electric field in the GaAs layer.

The obtained magnitude of the electric field allows us to estimate the areal surface charge density by the plane capacitor formula $\sigma = \varepsilon\varepsilon_0 F$. For the studied structures, it yields an areal charge density of about $2\times10^{11}\,\mathrm{cm}^{-2}$. These charges cannot be captured by impurities because their concentration in our samples is less than required to hold these charges by three orders of magnitude. Most probably, they are captured by the intrinsic defects around the interface between the QDs and the barrier layers.

The charging of the QDs changes their electrostatic potential relative to the matrix. According to the calculations of Pryor *et al.*,[5] the band offset for the valence band of the InGaP barrier layers and the InP QDs is small. So charging of the QDs is able to change drastically the localization potential for holes and therefore the optical properties of the QDs.

The presence of the electric charge in the InP QD layer was also observed by Anand *et al.* by the deep level transient spectroscopy.[10] However, they studied samples with *n*-doped InGaP barrier layers and the electric charges in QDs were caused by the electron transfer to the potential well from the barriers. This charging is natural and does not necessarily need the existence of defects around the strained InGaP/InP interface.

The question remains open about the structure and energy spectrum of the interface caused defects which capture the electric charge. Identification of the exact nature of these defects is a challenging problem in itself and calls for considerable further studies. Here we only make a few general comments. These defects are not PL quenchers because of the high quantum efficiency of the QDs luminescence. This is generally the case when the energy levels of the traps are deep enough. Another possibility is that the energy levels of the acceptors are shallow and close to the hole levels. In this case, it is not possible to distinguish the luminescence due to the electron-acceptor transitions from that due to the electron-hole transitions.

In summary, our results show that in the heterostructures with InP QDs there is a built-in electric field of 30 kV/cm. The electric field causes strong FKO in the photo- and electroreflection spectra. We present a model that assumes that the interface between the QDs and InGaP barrier layers contains a number of defects which behave like acceptors. During the growth process at high temperature they capture electrons from the neighboring layers of the structure. This process gives rise to the build-in electric field. In the investigated structures, the QDs layer holds a negative charge with areal density of $\approx 2\times10^{11}\,\mathrm{cm}^{-2}$. The electric charge around the QDs can essentially affect their physical properties and should be taken into consideration.

The authors acknowledge Dr. E. Tokunaga and Dr. S. Nair for fruitful discussions and a critical reading of the manuscript.

[1] N. Carlsson, W. Seifert, A. Petersson, P. Castrillo, M.-E. Pistol, and L. Samuelson, Appl. Phys. Lett. **65**, 3093 (1994).
[2] A. Kurtenbach, K. Eberl, and T. Shitara, Appl. Phys. Lett. **66**, 361 (1995).
[3] S. Raymond, S. Fafard, P. J. Poole, A. Wojs, P. Hawrylak, C. Gould, A. Sachrajda, S. Charbonneau, D. Leonard, R. Leon, P. M. Petroff, and J. L. Merz, Superlattices Microstruct. **21**, 541 (1997).
[4] M. Grundmann, R. Heitz, N. Ledentsov, O. Stier, D. Bimberg, V. M. Ustinov, P. S. Kop'ev, Zh. I. Alferov, S. S. Ruvimov, P. Werner, U. Gösele, and J. Heydenreich, Superlattices Microstruct. **19**, 81 (1996).
[5] C. Pryor, M.-E. Pistol, and L. Samuelson, Phys. Rev. B **56**, 10404 (1997).
[6] H.-W. Ren, M. Sugisaki, K. Nishi, S. Sugou, and Y. Masumoto, Jpn. J. Appl. Phys., Part 1 **38**, (1999).
[7] V. Davydov, I. Ignat'ev, H.-W. Ren, S. Sugou, and Y. Masumoto, *Proceedings of the 6th International Symposium ''Nanostructures: Physics and Technology,''* St. Petersburg, Russia, 1998, p. 200.
[8] V. Davydov, I. Ignat'ev, H.-W. Ren, S. Sugou, and Y. Masumoto, *Proceedings of the 24th International Conference on the Physics of Semiconductors*, Jerusalem, Israel, 1998 (to be published).
[9] D. E. Aspnes and A. Studna, Phys. Rev. B **7**, 4605 (1973).
[10] S. Anand, N. Carlsson, M.-E. Pistol, L. Samuelson, and W. Seifert, Appl. Phys. Lett. **67**, 3016 (1995).