FRANZ-KELDYSH OSCILLATIONS IN PUMP-PROBE SPECTRA OF InP SELF-ASSEMBLED QUANTUM DOTS

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Heterostructures with InP self-assembled quantum dots were studied. Strong Franz-Keldysh oscillations were found in their nonlinear reflection spectra measured by pump-probe method. These oscillations manifest built-in electric field of about 30 kV/cm. We suppose that this field originates from electric charge captured by the structural defects on the dots interface. An estimated areal density of electric charge is about 2×10^{11} cm^{-2}.

Self-assembled quantum dots (QDs) fabricated by epitaxial growth in the Stranski-Krastanov mode have been recently a subject of the extensive study. In this method, QDs are fabricated in a single growth process that prevents a QD surface from being contaminated. Nevertheless, the structure and properties of the interface between QDs and the barrier layer are still in question. The lattice mismatch between materials of QDs and barrier layers gives rise to stresses and strains around the QDs. For InAs and InP QDs, this problem was extensively investigated previously [1,2]. The strains can be sufficiently essential to generate local intrinsic defects at the QD/barrier layer interface.

In this paper we present the first experimental evidence of the existence of local defects in epitaxially grown QDs. We found that InP QDs possess a large amount of presumably negative electric charge. We suppose that the charge is captured by the acceptors located in InP QDs or in InGaP barrier layers near QDs. This process leads to the built-in electric field that causes the intense Franz-Keldysh oscillations in the photoreflection spectra. For the detailed study of these spectra we used pump-probe method because of its high sensitivity and a possibility to study the time evolution of processes.

The heterostructures studied were grown on (100) n^+-GaAs substrates by gas source molecular beam epitaxy. Their simplified structure is shown in the inset of Fig. 1. The average diameter of QDs is 40–60 nm, the average height is 5–10 nm, and the average distance between QDs is about 100 nm, which were determined by an atomic force microscopy in the reference structures. The

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photoluminescence (PL) spectra of the studied structures reveal QDs emission as a most intensive peak (Fig. 1) that indicates high quality of samples.

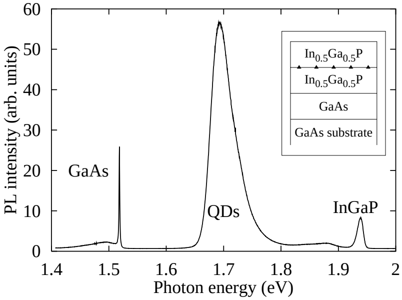

Figure 1. PL spectrum of the sample QDP1779. “QDs”, “InGaP”, and “GaAs” mark accordingly PL of QDs and barrier and buffer layers excitons. Inset: structure of the studied samples.

Pump-probe experiments were performed on the setup which includes a femtosecond Ti:sapphire laser tunable from 720 nm to 850 nm. The amplitude modulation of the pump and probe beams and a double lock-in detection of the signal allowed us to detect the reflection changes as low as 10^{-7} of the reflectance. Their time and pump power dependences are a subject of the other paper [3]. We discuss here only the spectral dependence of the signal.

The spectral dependence of the nonlinear reflection of the sample QDP1779 for the delay between pump and probe pulses of 60 ps is presented in Fig. 2. This dependence looks like intense oscillations in a wide spectral region. The spectra of the other samples reveal a similar behavior with approximately the same period of oscillations. All experiments led us to conclusion that we observe the Franz-Keldysh oscillations (FKO). To confirm this conclusion, the sample QDP1779 was supplied with electric contacts and the electroreflection spectrum was recorded. It is also presented in Fig. 2. One

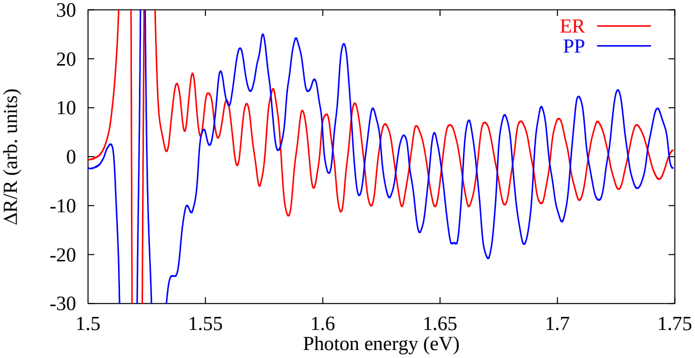

Figure 2. Pump-probe (PP) and electroreflection (ER) spectra of the sample QDP1779.

can see the oscillations with the same period as in the pump-probe spectrum.

FKO are an evidence of the built-in electric field in structures with InP QDs. We investigated pump-probe spectra of the structures without QDs, and also the structures with InAs QDs. The spectra of all these samples 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 physical processes which give rise to the built-in electric field. The model is schematically drawn in Fig. 3. Due to the strains, QDs and their surroundings contain a number of intrinsic defects. For the explanation of the experimental data, we should suppose that these defects are predominantly acceptors (A^0). During the growth process at a high temperature ($\approx 500\,^\circ$C) the holes in the QDs layer jump over the potential barrier and reach the GaAs buffer layer where they recombine with electrons produced by donors D^0. As a result, an excess of negative charge (A^-) occurs in the QDs layer and of opposite charge (D^+) in the buffer layer. This process forms a double electric layer with an electric field inside. The electric field penetrates into the GaAs buffer layer due to a small donor concentration (about a few units of 10^{15} cm^{-3}).

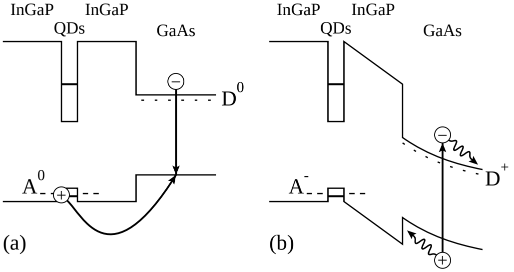

Figure 3. Charge transfer during the structure growth (a) and optical pumping (b).

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. This is followed by the change of the optical properties of heterostructure. Probe pulses detect mainly the reflectivity of the GaAs buffer layer because its optical thickness is much greater than that of the QD layer. That is why the observed regular oscillations start from the GaAs bulk exciton spectral position.

In order to determine the value of the built-in electric field from FKO spectra, we utilized an approximated formula of Aspnes and Studna [4]

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

Here E_m is the energy position of the m-th maximum, $E_g = 1.52$ eV is the GaAs band gap, F is the electric field, φ is the fitting parameter, and

$\mu = 0.058m_e$ is the reduced mass, where m_e is the electron mass.

The energy positions of the oscillation maxima in the spectra of pump-probe signal for all the studied structures and the fit are shown in the Fig. 4. There is a considerable discrepancy between the experimental and fitting curves which allow us to estimate only approximately 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 buffer layer due to the distributed charge of the ionized donors.

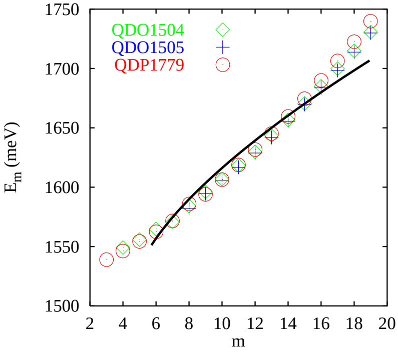

Figure 4. Energy position of FKO maxima versus their numbers. Solid line is the least squares fit by formula (1) with $F = 30$ kV/cm.

The obtained magnitude of the electric field allows us to estimate the average areal charge density by the plane capacitor formula $\sigma = \varepsilon\varepsilon_0 F$. For the studied structures it yields the areal charge density of $\sim 2 \times 10^{11}$ cm^{-2}. Most probably, these charges are captured by the structural defects on the interface between QDs and barrier layers.

In conclusion, the research performed shows that in the heterostructures with InP QDs the interface between QDs and InGaP barrier layers contains a number of defects which behave like acceptors. During the growth procedure at a high temperature they capture electrons from other layers of the structure. This process gives rise to the built-in electric field which causes strong FKO in the photo- and electroreflection spectra. We found that in the investigated structures the layer of QDs carries a negative charge with a surface density of about 2×10^{11} cm^{-2}. This electric charge essentially affects the physical properties of the QDs and should be taken into consideration.

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