GIANT OPTICAL NONLINEARITY OF HETEROSTRUCTURES WITH InP SELF-ASSEMBLED QUANTUM DOTS

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A nonlinear reflection of the heterostructures with InP self-assembled quantum dots is studied by pump-probe technique. A saturation of pump-probe signal in the spectral region of the absorption of quantum dots is found at extremely low pump power density of about 1 W/cm^2. This value together with estimation of the absorption coefficient leads to the conclusion that saturation of the nonlinear reflection occurs when quantum dot absorbs only single quantum of the light. This is a real evidence of the giant optical nonlinearity of the quantum dots.

Optical nonlinearity of the semiconductor heterostructures attracts considerable attention, either in light of the fundamental researches, and also for the important applications[1,2]. The main processes which cause nonlinear photoresponse are related to the screening of a Coulomb interaction and to a renormalization of the energy spectrum. For the quantum dots (QDs) the renormalization of the spectrum due to the photoexcitation is able to lead to a drastic change of their optical properties because of their δ-like density of states. As far as we know, there is no experimental data about nonlinear behavior of the self assembled QDs.

In this work we studied a nonlinear reflection of the InP self-assembled QDs by the pump-probe method. The heterostructures with QDs were grown by the gas source molecular beam epitaxy on (100) n^+-GaAs substrates. They contain one layer of QDs formed from the InP layer with nominal thickness of 4 monolayers (ML) between $In_{0.5}Ga_{0.5}P$ barrier layers on the GaAs buffer layer. The transmitted electron microscopy and scanning atomic force microscopy show that QDs have a pyramidal-like shape with an average base diameter of 40–60 nm, and an average height of 5–10 nm in the structures studied. An average distance between QDs is about 100 nm. A photoluminescence (PL) of QDs is observed in the PL spectra of all of the samples as most intensive

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band in the region 700–750 nm.

In the structures studied a built-in electric field is present [3]. It spreads over the both QDs and buffer GaAs layers. Photoexcitation of the free carriers leads to the change of this field, which is accomplished by the change of the reflection. The main idea of the extraction of the small signal from the one layer of QDs is based on the huge difference between the characteristic recovery times of the electric field after the pulse excitation of the QDs and GaAs. This times fall within subnanosecond scale for QDs and submillisecond[4] scale for bulk GaAs.

Against the conventional photoreflection, we utilized the pump-probe method which is capable of detecting peak reflection changes rather than integral ones. An amplitude modulation of the pump and probe beams at the different frequencies and a double phase detection of the signal allowed us to increase the sensitivity of the setup up to a detection of the reflection changes as low as 10^{-7} of the total reflectance. The modulation of the pump beam at high frequency (up to 5 MHz) let us to exclude the most part of the slow component of the transient reflection.

We studied the kinetics of the nonlinear reflection decay under various pump power densities and in wide spectral range of 710–830 nm, which includes areas of resonance excitation of both QDs and exciton of the GaAs buffer layer. Here we focus only on the pump power dependence of the signal.

The data measured on the samples QDP1779 and QDO1504 are presented in Figs. 1 and 2. The dependencies of the signal on the pump power density P are fitted by the function

$$I = I_0\left(1 - \exp(-\frac{P}{P_0})\right). \quad (1)$$

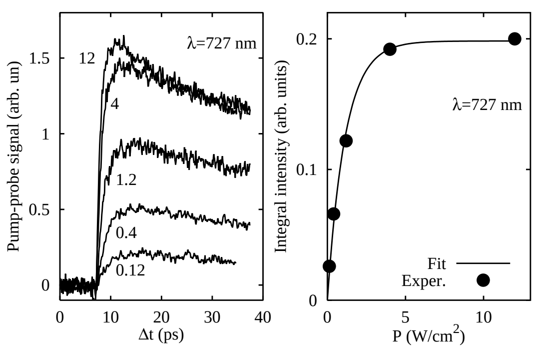

Figure 1. Left panel: time dependencies of the pump-probe signal from the sample QDO1504 for different pump power densities (in W/cm^2) at temperature of 1.6 K. Right panel: fitting of the integrated data by the equation (1) with P_0=1.15 W/cm^2.

All of the obtained data show that in the spectral region of the QDs absorption, the fast saturation of the signal is observed at the pump power density of the order of 1 W/cm^2. Beyond the QDs absorption band ($\lambda >$ 750 nm), the signal decreases (at the same pump power) and saturation power increases. Lower values of the P_0 are observed again at the wavelengths close to the GaAs bulk exciton line.

We have studied also the sample without QDs. It contains only the buffer GaAs layer covered by the thick InGaP barrier layer. The magnitude of the pump probe signal from this sample is much less than from samples with QDs and no saturation is observed except the wavelengths in the vicinity of the GaAs exciton spectral line.

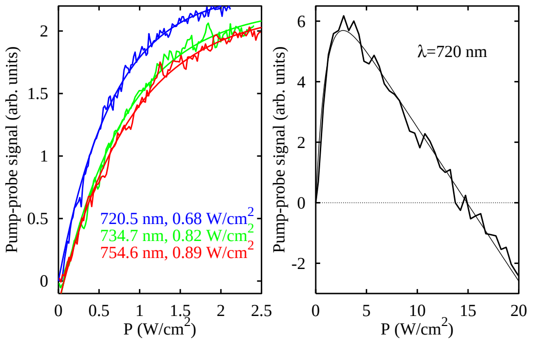

Figure 2. Left panel: pump power dependencies of the signal from sample QDP1779 at different wavelengths fitted by the formula (1). Obtained values of P_0 are shown. Right panel: pump power dependence of the signal from the sample QDO1504. Fit is made by modified formula $S = S_0\big(1 - \exp(-P/P_0)\big) + S_1 P$, where linear term accounts for the absorbtion in GaAs.

We offer the following explanation of the observed phenomena which is illustrated by Fig. 3. The light absorption in the GaAs layer leads to the generation of the free carriers. Their drift *decreases* the electric field. This absorption is not saturated up to the high power densities of the excitation light because of the large number of states in the bulk material. Only near the band gap edge it is possible to observe saturation under moderate power densities [5].

Photoexcitation of the QDs is also accompanied by a charge transfer within the thickness of the QD layer (about 10 nm) which leads to the *increase* of the field in GaAs. The later effect is integrally much smaller than effect of GaAs excitation. However, efforts undertaken were succeeded in it's reliable detection. An easy saturation of the QDs absorption is caused by their relatively low density of states.

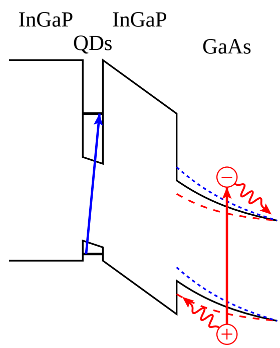

Figure 3. Simplified band diagram of heterostructure. Arrows show photoexcitation of QDs and GaAs buffer. Dotted and dashed lines indicate a change of potential due to both of these processes accordingly.

Probe beam mainly detects the changes in the reflection coefficient of the GaAs, because the latter has much more optical density than QDs. In fact, GaAs layer performs the rôle of the built-in signal amplifier in our experiments. In the framework of the above-described model, it is easy to understand the dependence of the pump-probe signal in the wide range of the excitation power. For example, the initial part of the curve in Fig.2 (right panel) reflects the *excitation of QDs* and *increase* of the electric field. The signal of *negative* sign at the higher power values is caused by *excitation of GaAs* and reflects *decrease* of the electric field.

The saturation power in the QDs spectral region, $P_0 = 1$ W/cm^2, corresponds to about 50 quanta of light per dot per pulse, taking into account inhomogeneous broadening of the QDs line which lets only a part of them be in resonance with the laser light. From the data of the reference book [6], the absorption coefficient of InP with 4 ML thickness at wavelengths around 730 nm can be estimated as about 0.6% per round trip. This value can be somewhat changed due to the modification of the spectrum during QDs formation. Nevertheless the increase of the absorption can not be considerable because it is not observed in a linear reflection. Therefore, we can suppose that the observed saturating power density corresponds to the absorption of *only single light quantum* by QD. The saturation mechanism includes so large renormalization of the QD spectrum that the dot is completely "turned off" after absorbing of the single quantum.

Summary. The research performed shows that utilizing of the pump-probe technique together with the fast modulation of the pump beam and phase detection of the signal allows us to extract the nonlinear part of the reflection signal caused by the excitation of QDs. Measured value of the saturating power density together with estimations of the absorption coefficient lead to the conclusion that saturation of the nonlinear reflection occurs, when QD absorbs only one quantum of the light. This is a real evidence of the giant optical nonlinearity of the quantum dots.

References

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