

Transient Absorption and Linear Dichroism in Self-Assembled InAs/GaAs Quantum Dots

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Time-resolved and polarization-resolved measurements are performed on InAs/GaAs quantum dots by means of time-resolved differential reflection spectroscopy. With this technique, the polarization anisotropy of the absorption bleaching in a quantum dot is measured. The excitation density dependent measurements of the bleaching magnitude, show a strong polarization dependence, which corresponds to linear dichroism. Dichroism with a magnitude of 1.3 is observed, governed by strain within the sample.

Introduction

The time-response and the polarization properties of self-assembled quantum dots (QDs) are of interest from fundamental and device point of views. Applications include low power all-optical switching [1], and also spintronics [2], i.e. processing of quantum information exploiting the spin-dependent properties of a QD. The optical transitions in a QD are very sensitive to the symmetry of the confinement structure. This means that shape anisotropy [3], composition gradients [4], strain, etc, can induce polarization asymmetry [5]. The polarization anisotropy as observed in QD-photoluminescence, has been investigated theoretically as well as experimentally by many groups. However, contrary to luminescence studies, the polarization dependence of the QD absorption spectrum is seldom investigated.

We report results obtained by polarization-resolved pump-probe time-resolved differential reflection spectroscopy (TRDR) [6] on InAs/GaAs self-assembled quantum dots. Using TRDR, we are able to probe the QD absorption spectrum directly. Analyses of the QD-shape, luminescence and TRDR-results, show anisotropy with a common preferential orientation along the [1-10] crystal axis. We observe that the absorption anisotropy, expressed by the degree of polarization, decreases with increasing pump excitation density in analogy with the photoluminescence (PL) observations of Noda *et al.* [7], for linear polarized probe fields. The strong dependence of the polarization degree on the excitation density is due to the difference in the absorption cross section, which indicates linear dichroism of the QDs. We attribute this anisotropy to piezoelectric effects governed by strain within the sample.

Sample and Experimental Details

The time-resolved measurements are performed on a 5 layer self-assembled InAs/GaAs QD sample grown by molecular beam epitaxy on GaAs (100). After deposition of a 295 nm GaAs buffer layer at 580 °C, the temperature was lowered to 490 °C for the growth of the multiple QD layers. A 30 nm GaAs layer was deposited before the growth of the 5

QD-layers: Each QD-layer consists of 2.1 ML of InAs followed by 30 nm GaAs. The QD layers can thus be considered as electronically uncoupled. Finally, the sample is capped by 137 nm GaAs grown at 580 °C. Atomic force microscopy images show that the QDs are formed with a density of approximately $2.8 \cdot 10^{10} \text{ cm}^{-2}$.

To determine the optical time-response of the QDs, the sample is investigated by two-color time-resolved pump-probe differential reflection spectroscopy [6, 8]. In this configuration, a 76 MHz mode-locked Ti:sapphire laser is used as the pump source, and is mechanically chopped with a frequency of 4 kHz. The pump pulses are focused on the sample with a spot size of 55 μm , exciting carriers above the bandgap in the GaAs barrier layers. The pump-induced modulation is probed by 200 fs and 2.5 ps pulses, generated by a tunable optical parametric oscillator (1050-1330 nm). The probe is tuned into resonance with the QD transition. The probe light is focused on the sample with a spot size of 25 μm using a graded index lens. The probe light reflected from the sample is collected by the same lens and is measured using a balanced photodetector.

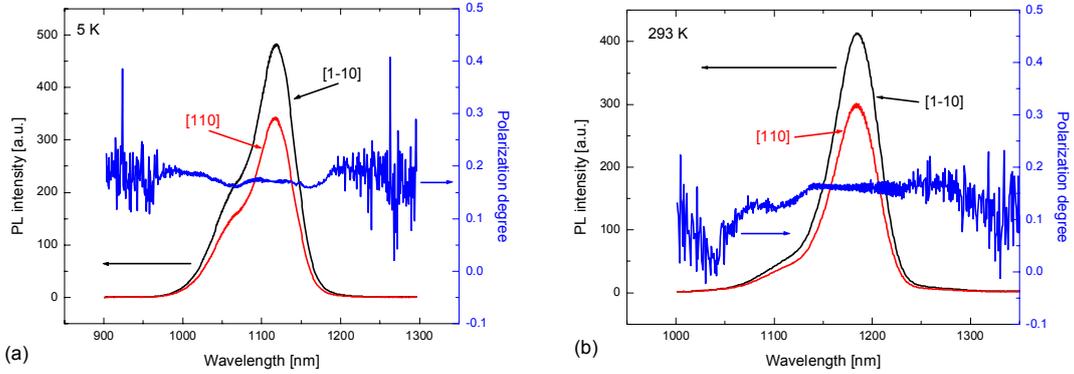


Figure 1: Polarization-resolved PL in the backscatter configuration at 5 K (a) and 293 K (b), including the degree of polarization, as derived by Eq. (1).

Results

The sample has first been examined by atomic force microscopy (AFM). Analysis of the AFM images shows that the QDs are elongated in the [1-10] direction. An average elongation of 2.7 % is observed, which corresponds to a shape anisotropy with a degree of $\rho_{\text{shape}} = 0.013$, as derived by Eq. (1). The degree of anisotropy is given by:

$$\rho = \frac{I_{\alpha} - I_{\beta}}{I_{\alpha} + I_{\beta}} \quad (1)$$

Here, I_i is the size or the signal magnitude in one of the mutually perpendicular directions α , β . As a result of the shape anisotropy, a difference in the confinement along the in-plane directions is induced. Therefore, optical anisotropy is expected, which is verified by polarization-resolved PL measurements, as is depicted in Fig. 1. An inhomogeneous broadening of the spectrum is observed, which is due to the QD size distribution. The PL-spectra show that the QD luminescence has a preferential orientation in the [1-10] direction, in agreement with the elongation direction of the QDs. From the PL-spectra, a linear polarization degree of $\rho_{\text{PL}} = 0.16$ is derived, and is nearly constant over the whole

energy spectrum, indicating a homogeneous or nearly constant shape, orientation and strain distribution. Randomly oriented QDs are expected to have a zero or near zero luminescence anisotropy. Notice, that no temperature dependence of the polarization degree is observed, as is indicated by comparing Fig. 1a and 1b.

Although, no temperature dependence of the luminescence polarization is observed, all TRDR measurements are performed at $T = 77$ K in order to avoid thermally activated processes, e.g. non-radiative recombination. The time-evolution of the differential signal of the Π_x (E // [1-10]) and Π_y (E // [110]) probe polarizations are presented in Fig. 2. The probe wavelength is tuned into resonance with the peak of the ground-state PL-spectrum. The inset of Fig. 2 presents the orientation dependence of the differential signal, $\Delta R/R_0$. A preferred polarization, i.e. Π_x , of the QD optical absorption is clearly observed, consistent with the PL measurements. We observe that the rise times of the bleaching signal, i.e. carrier capture into the dots, as well as the decay times, i.e. the carrier recombination, are independent of probe polarization, with characteristic times of $\Gamma_{\text{cap}}^{-1} = 17$ ps and $\Gamma_{\text{rec}}^{-1} = 1500$ ps, respectively.

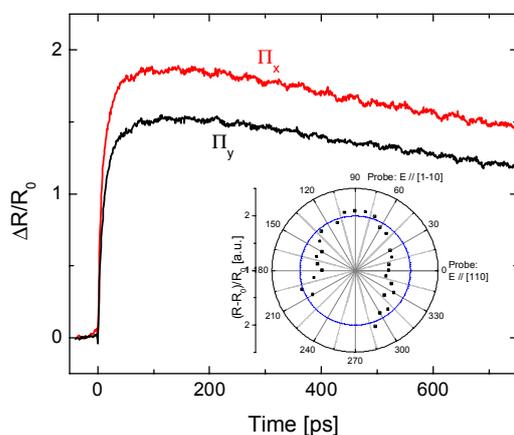


Figure 2: Time-evolution of the TRDR signal probed with a polarized probe beam. The inset depicts the bleaching magnitude with respect to the [110] crystal axis.

Although the preferred orientation direction is the same for the shape, luminescence and absorption anisotropy, the magnitude of the anisotropy is different. Since the

luminescence anisotropy has been reported to depend on the excitation density [6], we also investigate the pump power dependence of the QD absorption anisotropy. The QD absorption bleaching signal increases with the pump power due to an increasing state-filling and can be described by

$$\frac{\Delta R}{R_0}(I_{\text{pump}}) = \frac{\Delta R}{R_0} \Big|_{\text{sat}} (1 - e^{-k_{\text{abs}} I_{\text{pump}}}), \quad (2)$$

in which $\Delta R/R_{0,\text{sat}}$ is the saturated absorption bleaching when $I_{\text{pump}} \rightarrow \infty$, k_{abs} is a absorption cross section proportional to the bleaching of the QD probe absorption and I_{pump} is the pump photon density per pulse. From a fit of Eq. (2) to the results plotted in Fig. 3, we deduce values of $k_x = 6.8 \cdot 10^{-13} \text{ cm}^2$ and $k_y = 5.2 \cdot 10^{-13} \text{ cm}^2$ per QD-layer for Π_x and Π_y probe pulses, respectively. From the two absorption cross sections, a *linear dichroism* of $k_x/k_y = 1.3$ is explicitly derived.

In the inset of Fig. 3, the absorption bleaching anisotropy as calculated using Eq. (1), is depicted as function of the pump power. With identical k -values for Π_x and Π_y polarization, i.e. the dichroism would be zero, the absorption bleaching anisotropy shown in the inset of Fig. 3 would also be constant. The pump power dependence of the

bleaching anisotropy can be well approximated by an exponential fit, shown in the inset of Fig. 3. We observe a saturated absorption bleaching of, $\rho_\infty = 0.066$.

The observed QD-absorption anisotropy can be partly explained by the shape anisotropy, although, atomistic pseudopotential calculations [9, 10] show that even in the presence of QDs with a symmetric geometry, QDs have a preferred polarization direction [10, 11] due to internal piezoelectric fields. We ascribe the measured QD absorption anisotropy to internal piezoelectric fields, which occur in crystals with a lack of inversion symmetry. Hereby, the crystal responds to shear strain, for instance due to composition gradients [4], with a separation of the electron and the hole envelope wavefunctions, resulting in a charge polarization within the QD. Due to this piezoelectric effect, the wavefunctions of the electrons and holes are stretched along the [110] and [1-10] direction, respectively, resulting into a polarization of the optical transitions in the [1-10] direction [3, 12].

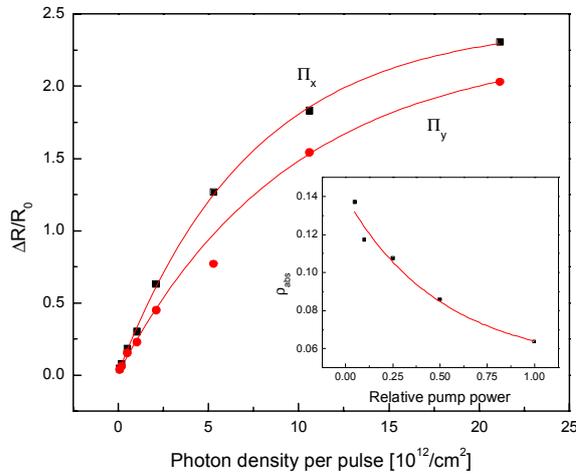


Figure 3: Magnitude of the TRDR signal for two mutually orthogonal linearly polarized probe fields, as function of excitation density. A fit as described by Eq. (2) is included. The inset depicts the polarization degree of the QD bleaching, using Eq. (1).

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