

# Polarization Anisotropy in Self-Assembled Quantum Dots within Transient Absorption Bleaching

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*Abstract*— Excitation dependent optical anisotropy in InAs/GaAs quantum dots is studied by means of time-resolved differential reflection spectroscopy, probing the linear polarization eigenstates. The measured dependence of the bleaching is due to linear dichroism. A dichroism of 1.3 is observed, which is determined by strain.

*Keywords*— quantum dots, time-resolved, pump-probe, absorption, anisotropy

## I. INTRODUCTION

Polarization properties of self-assembled quantum dots (QDs) are of interest from fundamental and device point of views. Not only regarding to waveguide structures [1], but also for the application of QDs in quantum spintronics [2], i.e. processing of quantum information with spin states in QDs. The optical transitions 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 and non-parabolic bands [5]. The polarization anisotropy in 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 seldomly investigated.

We report on the results which are 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, presented by the degree of polarization, decreases with increasing pump excitation density in analogy with the 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 due to strain.

## II. SAMPLE & 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 layers of QDs: 2.1 ML of InAs followed by 30 nm GaAs. Hereby, the QD layers can be considered as electronically uncoupled. Finally, the sample is capped by 137 nm GaAs at a temperature of 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 (Ti:S) 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  $\mu\text{m}$ , exciting carriers in the GaAs barrier layers. The pump-induced modulation is probed by 200 fs and 2.5 ps pulses which are generated from an optical parametric oscillator. The

probe is tuned to the QD optical resonance. The probe light is focused on the sample with a spot size of 25  $\mu\text{m}$  using a graded index lens. Probe light reflected from the sample is collected by the same lens and is focused on a balanced photodetector.

### III. 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 the perpendicular directions  $i = \alpha, \beta$ . As a result of the shape anisotropy a difference in the confinement along the in-plane directions is induced. Therefore, optical anisotropy is to be expected, which is verified by polarization-resolved PL measurements, as is depicted in figure 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, namely the [1-10] direction, in agreement with the elongation direction of the QDs. From these PL-spectra a linear polarization degree of  $\rho_{\text{PL}} = 0.16$  is derived. The anisotropy 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 figure 1a and b.

Contrary to quantum well structures, LH-transitions may contribute to the PL of QDs. Although, HH-transitions are only allowed for the in-plane polarization, LH-transitions are allowed for both in-plane and out-of-plane polarizations. To emphasize the result of HH-LH mixing in the valence band, the cleaved-side luminescence has been measured and the obtained spectra are depicted in figure 1c. We observe strong polarization of the luminescence within the plane perpendicular to the growth direction, from which an anisotropy of  $\rho_{\text{cleaved}} = 0.55$  is derived. This indicates a strong HH-LH mixing, i.e. the higher the contribution of the LH states the lower the optical anisotropy.

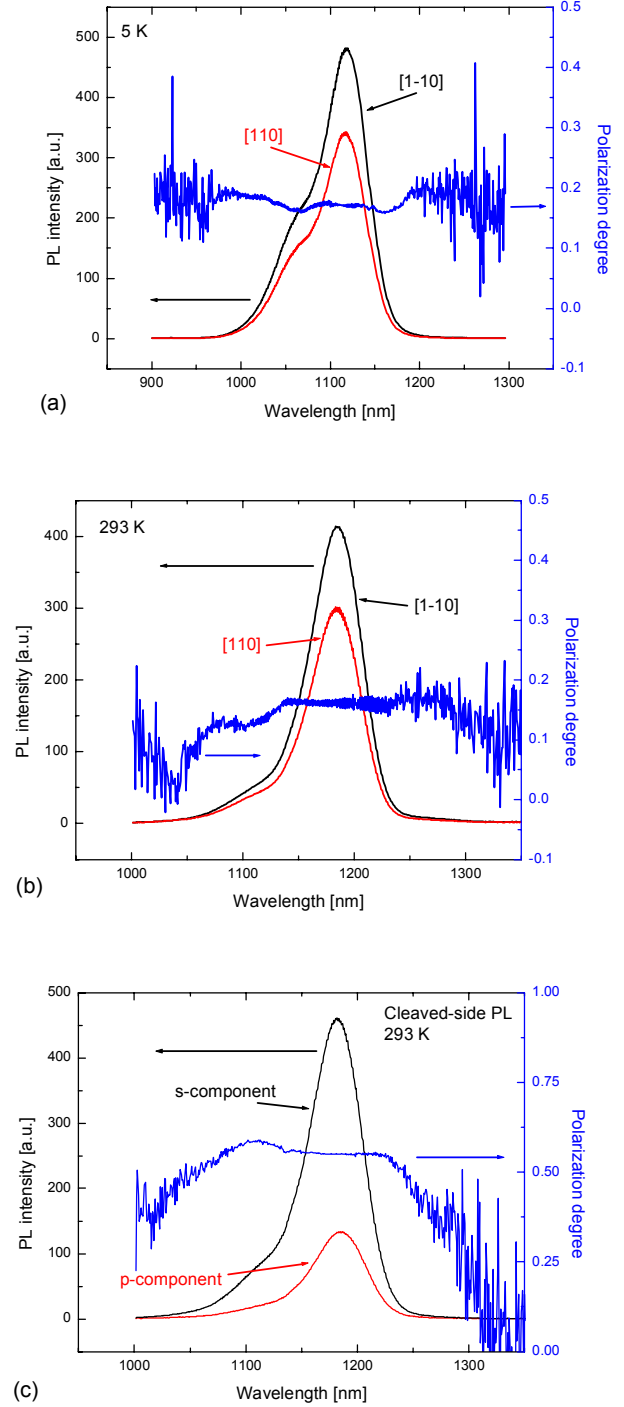


Figure 1 Polarization-resolved PL for the backscatter, (a) and (b), and cleaved-side configuration (c), at 5 K and 293 K, including the degree of polarization, eq. (1).

Although, no temperature dependence of the luminescence polarization is observed, all TRDR measurements are performed at  $T = 77$  K in order to avoid thermal activated processes, e.g. non-radiative recombination. The time-evolution of the differential signal of the  $\Pi_x$  ( $E \parallel [1-10]$ ) and  $\Pi_y$  ( $E \parallel [110]$ ) probe

pulses are presented in figure 2. The photon energy of the probe pulses is tuned into resonance with the peak energy of the ground-state PL-spectrum. The inset of figure 2 presents the magnitude of the differential signal,  $\Delta R/R_0$ , as function of the polarization orientation of the probe field with respect to the [110] crystal axis. A preferred polarization, i.e.  $\Pi_x$ , of the QD optical absorption is clearly observed, which is consistent with the PL measurements.

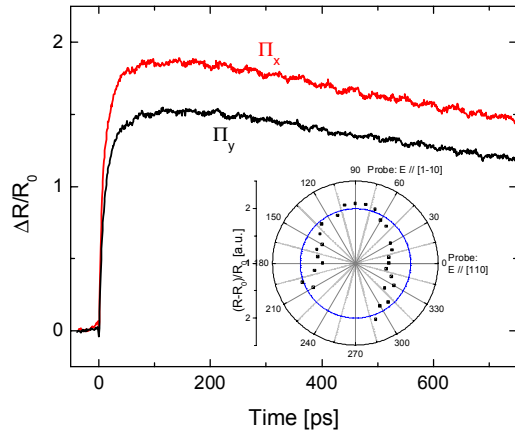


Figure 2 Time-evolution of the TRDR signal with the polarized probe fields. The inset depicts the bleaching magnitude with respect to the [110] crystal axis.

From the time-resolved measurements, we observe that the rise times of the bleaching signal, i.e. carrier capture in the dots, as well as the decay times, i.e. carrier recombination, are independent of the probe polarization direction, with characteristic times of  $\Gamma_{\text{cap}}^{-1} = 17$  ps and  $\Gamma_{\text{rec}}^{-1} = 1500$  ps for the carrier capture and carrier recombination, respectively.

Although, shape, luminescence and absorption anisotropy are consistent in case of the preferred direction, for the magnitude of the anisotropy no consistence is obtained. However, the degree of luminescence anisotropy depends on the excitation density [6], and in order to investigate this dependence in the case of absorption, the differential signal as function of pump excitation power has been investigated for linear polarization. With the increase of the pump power the differential reflectivity signal increases, i.e. more QD eigenstates are occupied by pump-generated carriers and hence an increase of the QD absorption bleaching. The dependence of the bleaching signal as function of excitation power is well described by

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

with  $\Delta R/R_{0,\text{sat}}$  the saturation bleaching as  $I_{\text{pump}} \rightarrow \infty$ ,  $k_{\text{abs}}$  the absorption cross section which includes the absorption probability and is a direct measure of the QD absorption.  $I_{\text{pump}}$  is the photon density per pump pulse. A fit of eq. (2) yields the absorption cross section of the linear fields. We deduce values of  $k_x = 6.8 \cdot 10^{-13}$  cm<sup>2</sup> and  $k_y = 5.2 \cdot 10^{-13}$  cm<sup>2</sup> per layer for  $\Pi_x$  and  $\Pi_y$  probe pulses, respectively. From these  $k$ -values, a ratios of  $k_x/k_y = 1.3$  is derived, explicitly, indicating a difference in the absorption of the QD eigenstates of the linear polarization, i.e. linear dichroism.

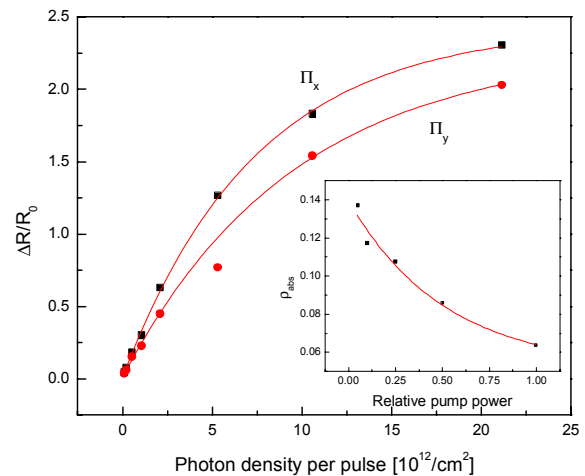


Figure 3 The TRDR magnitude for linear polarized probe fields, as function of excitation density. A fit as described by eq. (2) is included. The inset depicts the polarization degree of the QD bleaching, using eq. (1).

In the inset of figure 3, the degree of absorption anisotropy, eq. (1), is depicted as function of excitation power. The excitation dependence is a direct result of the QD dichroism, i.e. different  $k$ -values (eq. (2)), and can be well approximated in the first order by an exponential fit, as is shown in figure 3. We observe a non-vanishing degree for  $I_{\text{pump}} \rightarrow \infty$  with a value of,  $\rho_{\infty} = 0.066$ .

The observed anisotropy can be partly explained by the shape anisotropy, although, atomistic pseudopotential calculations [9, 10] show that even with the presence of geometrical symmetry, QDs have a preferred polarization direction [10, 11]. Therefore, we ascribe the measured anisotropy of the absorption to the internal piezoelectric fields, which originates from the lack of inversion symmetry of III-V semiconductors. Hereby, the crystal

responds to shear strain, for instance due to composition gradients [4], leading to a separation between 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, hence, polarization of the optical transitions in the [1-10] direction [3, 12].

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