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Dichroic reflection of InAs/GaAs quantum dots

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Polarization-resolved reflection measurements are performed on nearly circular InAs/GaAs quantum dots (QDs) by means of time-resolved differential reflection spectroscopy. We observe linear polarization anisotropy of the differential absorption, revealing the dichroic character of the QD reflection. The observed magnitude of the dichroism is $\Theta_{[1\bar{1}0]}/\Theta_{[110]}=1.07$. The polarization has a preferential direction orientated along the $[1\bar{1}0]$ crystal axis, which is confirmed by polarization-resolved photoluminescence. We observe that the polarization anisotropy of the reflectivity is strongly dependent on the pump excitation density, decreasing from $\rho=0.14$ at low excitation to $\rho=0.06$ at high excitation. The pump power dependence is described by a binomial model taking into account the statistics of carrier capture into a limited number of QDs. © 2005 American Institute of Physics. [DOI: 10.1063/1.2073971]

I. INTRODUCTION

Anisotropy in the photoluminescence (PL) of self-assembled quantum dots (QDs) has been investigated by many groups, experimentally^{1–3} as well as theoretically.^{1,4–7} However, the polarization properties of the QD absorption and reflection have seldom been investigated. Polarization-dependent absorption measurements on QDs have been reported recently by Tribollet *et al.*⁸ They report on linear anisotropy in the absorption bleaching, which reveals the dichroic character of QDs. Their measurements have been done by a time-resolved pump-probe differential transmission technique at a single excitation density. A polarization rate of 30% is observed which is explained as due to size and shape anisotropies. Theoretically, it has been shown that circularly symmetric QDs can have a preferred optical polarization direction^{7,9} due to composition gradients^{7,10} and strain distribution^{2,9,11} within the QDs.

The goal of this study is to experimentally determine the dichroic strength of (nearly) circular QDs. We will show in this report, that the dichroism in QDs (Ref. 8) can also be determined by differential reflectivity, i.e., by using in-plane polarization-resolved two-color pump-probe time-resolved differential reflection spectroscopy (TRDR).^{12–14} The carriers generated by the pump pulses are captured in the QD energy states which leads to the bleaching of the QD transition. The bleaching results in a decrease of the probe pulse absorption and thus also to a change of the QD reflection due to the carrier-induced change in the QD refractive index. The pump-induced reflection changes can be monitored by tuning the probe energy into resonance with the QD transition.

Using nonresonant excitation, TRDR allows us to measure both the carrier capture and relaxation time as well as the carrier lifetime within the QD, because TRDR is directly related to the changes of the occupation of the QD eigenstates. Moreover, reflectance measurements have the prefer-

ence with respect to transmission measurements, because the sample substrate does not have to be removed. Hence, TRDR is a nondestructive experimental technique and allows for a direct comparison with different optical techniques. In addition, this experimental technique can even be used to investigate structures with a low luminescence efficiency such as low-temperature-grown QDs.^{15,16}

Using TRDR at various excitation densities, we observe that the amplitude of the differential reflection signal strongly depends on the pump-induced carrier density η . The signal increases with increasing carrier density due to an increase of state filling, i.e., the TRDR signal is proportional to the population of the QD energy states. In this report, we investigate the polarization anisotropy of the amplitude of the TRDR signal. In particular, we will discuss the decrease of the polarization anisotropy with increasing carrier density.

II. SAMPLE DETAILS AND EXPERIMENTAL SETUP

A. Sample growth

Polarization-resolved reflectivity measurements are performed on a five-layer self-assembled InAs/GaAs QD sample grown by molecular-beam epitaxy on GaAs (100).¹⁷ After the 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 five layers of QDs. Each QD layer consists of 2.1 ML (monolayer) of InAs followed by a 30 nm GaAs spacer layer. 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 (AFM) images of uncovered dots show that the QDs are formed with a density of approximately $2.8 \times 10^{10} \text{ cm}^{-2}$, see Fig. 1. Analysis of the AFM images, obtained under various sample orientations, show that the QDs are elongated in the $[1\bar{1}0]$ direction with an average

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FIG. 1. AFM image of uncovered InAs/GaAs QDs. The scan field is $1 \times 1 \mu\text{m}^2$ and the black-to-white height contrast is 15 nm.

elongation of 2.7%. The elongation corresponds to a shape anisotropy with a degree of $\rho_{\text{shape}}=0.013$, as defined by

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

Here, I_i is the QD length or, in the remainder of this report, the signal amplitude in the orthogonal directions $i=\alpha, \beta$. Hence, the QDs in this study are nearly circular.

B. Experimental details

We study the carrier-induced reflection change by two-color time-resolved pump-probe differential reflection spectroscopy^{12,13} at 77 and 293 K. In this configuration, a 76.6 MHz mode-locked Ti:sapphire (Ti:S) laser is used as the pump source, which 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 in which they diffuse towards the QDs. The capture of the carriers into the QDs leads to a change of the QD reflection $\partial R_{\text{QD}}/\partial \eta$. The carrier-induced reflection changes of the QD layers are probed by 2 ps pulses, generated by a tunable optical parametric oscillator, which is synchronously pumped by the same Ti:S laser. The probe pulses are tuned into resonance with the main energy of the QD ground-state transition. For the polarization-resolved TRDR measurements, the probe field is linearly polarized in the plane perpendicular to the growth direction, i.e., the [001] crystal axis. $\Pi_x(\Pi_y)$ represents the probe field polarized along the $[1\bar{1}0]$ ($[110]$) crystal axis. The probe light is focused on the sample with a spot size of $25 \mu\text{m}$ using a graded index lens. The light reflected from the sample is collected by the same lens and is measured using a balanced photodetector. For all the measurements, the field of the pump pulses is horizontally polarized, i.e., along the $[1\bar{1}0]$ crystal axis. However, the polarization of the pump field has no influence on the results due to the loss of polarization memory during the energy relaxation from the GaAs barrier layer into the QD, which has been verified experimentally.

FIG. 2. (a) Time evolution of the QD differential reflection signal $\Delta R/R_0$ for probe polarization Π_x and Π_y at 293 K. The inset depicts the TRDR amplitude and (b) the TRDR rise and decay times as functions of the probe polarization orientation with respect to the $[110]$ crystal axis.

The QD size is small compared to the wavelength of the probe pulses, such that the QDs are assumed to be electronically small.¹⁸ Hence, the QD layers can be approximated by a thin layer with an effective refractive index. In this case we can use the approach of Tassone *et al.*¹⁹ to derive the QD reflectivity.^{18,20,21} A full derivation of the QD differential reflectivity will be published elsewhere.¹⁴ The bleaching of the QD ground-state transition due to carrier capture within the QD results directly in a change of the probe pulse reflection. $\Delta R = \partial R / \partial \eta$.^{13,14,22} We observe that the carrier-induced reflection change is linear proportional to the QD density of states (DOS), $\Delta R \sim \text{DOS}$, which has been shown in Ref. 17. In addition, as has been discussed in Ref. 12, the TRDR signal can be interpreted as an absorption bleaching signal since the differential reflection signal originates from the occupied QD energy states which are in resonance with the probe photon energy.

III. EXPERIMENTAL RESULTS

A. Time-resolved differential reflection

The time evolution of the reflection signals with Π_x and Π_y -polarized probe pulses, measured at 293 K, are depicted in Fig. 2(a). It is observed that the amplitude of the differential signal $(R - R_0)/R_0 = \Delta R/R_0$, with R_0 the unperturbed reflection, is larger for the Π_x polarization, revealing optical anisotropy.⁸ Since the reflection spectrum directly represents the QD DOS, a linear anisotropy of the QD DOS is observed. The anisotropic reflectance is emphasized by the inset of Fig. 2(a), in which the amplitude of the differential signal is depicted as a function of the probe orientation with respect to the $[110]$ crystal axis.

FIG. 3. Amplitude of the QD differential reflection as a function of the pump-induced carrier density per pulse for probe polarization Π_x and Π_y at 77 K. The fits are calculated by using Eq. (5). The inset depicts the degree of polarization anisotropy of the reflectivity $\rho_{\Delta R}$ as a function of the carrier density.

The initial differential reflectivity rise time τ_{rise} which is governed by carrier capture and relaxation within the QD, as well as the signal decay time τ_{dec} which is governed by the carrier lifetime, are independent of the probe polarization with values of approximately $\tau_{\text{rise}} = 13$ ps and $\tau_{\text{dec}} = 384$ ps, as depicted in Fig. 2(a). Although, a small variation of the carrier dynamics as function of the orientation is observed, as is shown in Fig. 2(b), the consistent carrier lifetimes for the Π_x and Π_y polarization indicates that there is no net transfer of orientation during the carrier relaxation and recombination processes. Thus, the reflection anisotropy is only determined by the anisotropy in the DOS, at all time scales. At 293 K, characteristic times of $\tau_{\text{rise}} = 12.7 \pm 0.3$ ps and $\tau_{\text{dec}} = 382 \pm 5$ ps are observed.

B. Excitation density dependence

Although, a clear anisotropy for the differential reflection is observed, the degree of dichroism is still unknown. Moreover, the reflection anisotropy strongly depends on the carrier density, as will be shown next. Figure 3 depicts the amplitude of the TRDR signal as a function of the pump-induced carrier density for Π_x and Π_y probe polarization. At carrier densities up to 2×10^{12} carriers/cm², the differential signal increases linearly with the carrier density due to an increase of state filling by the pump laser, i.e., the TRDR signal is proportional to the population of the QD energy states. With increasing carrier density above 2×10^{12} carriers/cm², the signal amplitude starts to deviate from the linear behavior and begins to saturate. Since the TRDR signal monitors the population of the QD energy states,¹⁷ we ascribe the onset of the saturation to the finite DOS of the QD ensemble. A strong carrier-density-dependent degree of the polarization anisotropy of the reflectivity, $\rho_{\Delta R}$, is observed, by using Eq. (1). As is shown by the inset of Fig. 3, $\rho_{\Delta R}$ decreases from approximately 0.14 down to 0.06 with increasing excitation density. Hence, the amount of dichroism⁸ can not simply be deduced from a single TRDR measurement.

To derive the dichroism magnitude, the carrier density dependence of the TRDR signals, as are depicted in Fig. 3, has to be examined. We assume that the pump-generated carriers are randomly distributed over the energy states within the QD ensemble.²³ For our derivation we only take the lowest energy state, i.e., the QD ground state, into account. With the increase of the number of occupied energy states, due to the increase of the carrier density, the probability of subsequent carrier capture decreases proportionally.²⁴ If we assume that the carrier distribution, i.e., carrier capture, remains random, even at high excitation densities, the density of occupied QD states N_{oc} can be expressed as

$$N_{\text{oc}}(\eta) = \eta - \frac{\eta(\eta-1)}{2! N_{\text{tot}}} + \frac{\eta(\eta-1)(\eta-2)}{3! N_{\text{tot}}} - \dots \quad (2)$$

The density N_{oc} Eq. (2) can be written in the form of a binomial function, such that

$$N_{\text{oc}}(\eta) = \frac{[1 - (1 - \beta)^\eta]}{\beta}, \quad \beta = \frac{1}{N_{\text{tot}}} \quad (3)$$

Here, N_{tot} is the total number of QD energy states (ground state) within the ensemble. However, due to the inhomogeneous QD size distribution and due to the fact that the probe pulses have a finite width (approximately 1 meV), a selective portion of the QD ensemble is monitored during the measurements. In our case, where the probe is tuned in resonance with the main QD transition energy $\hbar\omega_c$, the fraction of probed QDs is

$$\frac{N_{\text{probed}}(\omega_c)}{N_{\text{tot}}} = \frac{\int_{\omega_c - \Delta\omega}^{\omega_c + \Delta\omega} D(\omega) d\omega}{\int_{-\infty}^{\infty} D(\omega) d\omega} \approx 0.017, \quad (4)$$

where $D(\omega)$ denotes the QD DOS. This means that we should only consider N_{probed} as the number of QDs relevant for the reflection at $\hbar\omega_c$. As a consequence β in Eq. (3) should be substituted by β_{probed} taking into account the probed QD ensemble. However, this is equivalent to replacing η by $(N_{\text{probed}}/N_{\text{tot}})\eta = \gamma\eta$.

As is mentioned above, the TRDR signal depends on the number of occupied QD states which are in resonance with the probe photon energy, i.e., $(\Delta R/R_0)(\omega) \propto N_{\text{oc}}(\omega)$. Thus, we can use Eq. (3) to express the carrier density dependence of the differential signal. We obtain

$$\frac{\Delta R}{R_0}(\eta) = \Theta N_{\text{oc}}(\eta) = \Theta \frac{[1 - (1 - \beta)^{\gamma\eta}]}{\beta}, \quad (5)$$

with Θ the differential reflection per QD. Notice, Θ has the dimension of a cross section. Due to the fact that the TRDR signal can be interpreted as a bleaching signal, Θ can be interpreted as an absorption cross section as is defined by Blood,²⁵ i.e., $\alpha_{\text{QD}} = \sigma N$ with α_{QD} the QD absorption and σ the absorption cross section per dot.

Figure 3 depicts the differential reflection amplitude as function of the excitation density, including the best fit using Eq. (5). From the fitting procedure with $N_{\text{tot}} = 2.8 \times 10^{11}$ cm⁻² (five layers) as the number of spin-up and spin-

FIG. 4. Polarization-resolved PL spectra of the linear polarizations Π_x and Π_y and the degree of linear polarization anisotropy at 5 K (a) and 293 K (b).

down ground states, we obtain $\Theta_x = 2.33 \times 10^{-15} \text{ cm}^2$ and $\Theta_y = 2.17 \times 10^{-15} \text{ cm}^2$. The different absorption cross sections indicate a polarization anisotropy in the optical reflection of the QDs, with a ratio of $\Theta_x/\Theta_y = 1.07$, i.e., the amount of linear dichroism. Hence, a degree of polarization anisotropy of $\rho_{\Delta R} = 3.6\%$ is deduced in the limit of complete saturation, i.e., $\eta \rightarrow \infty$. If the degree of reflection anisotropy is calculated directly from the data as depicted in Fig. 3, we observe a decrease of the degree of polarization anisotropy, which is presented in the inset of Fig. 3.

C. Polarization-resolved PL

For further characterization of our QD sample, polarization-resolved PL measurements are performed with an excitation density of 250 mW/cm^2 at 532 nm . The results are depicted in Fig. 4. Consistent with the previous results, the PL spectra show that the QD luminescence has a preferential orientation, i.e., the $[1\bar{1}0]$ direction, in agreement with the TRDR results. We observe that the degree of linear polarization is nearly constant over the whole energy spectrum, with $\rho_{\text{PL}} = 0.17$ and 0.16 for temperatures of 5 and 293 K , respectively. Only a minor temperature dependence of the polarization degree is observed.

IV. DISCUSSION

In order to fit the data for the $\Pi_{\parallel}$ polarization, we have to modify Eq. (5) with $\gamma\eta \rightarrow \gamma\eta + \mu N_{\text{tot}}$, with $\mu = 0.016$. Notice, from Eq. (4) and from the data fit we observe that, $\mu \approx N_{\text{probed}}/N_{\text{tot}} = \gamma$, which suggests that the $[1\bar{1}0]$ direction has initially the preferential orientation. Due to the offset in the occupation probability, taken into account by μ , instead of a constant value of $\rho_{\Delta R}$, a strong dependence of the polarization anisotropy on the excitation density is expected. An excitation density dependence of the polarization anisotropy is indeed observed, as is shown by the inset of Fig. 3. A similar decrease of the polarization anisotropy with increasing excitation density has also been observed in PL by Noda *et al.*¹ The degree of linear polarization in PL (low excitation) $\rho_{\text{PL}} = 0.17$ is well consistent with the results obtained by TRDR at low excitation $\rho_{\Delta R} = 0.14$. In addition, the observation of a consistent preferred orientation direction of the optical anisotropy, i.e., $[1\bar{1}0]$, and the nearly equal strength suggests a common origin.

The difference between the optical and the geometrical anisotropies has previously been observed experimentally by several groups, e.g., Refs. 1, 2, and 26, showing that the optical anisotropy is not unambiguously determined by the QD shape anisotropy. This is confirmed by our photoluminescence results on nearly circular symmetric QDs. As is mentioned previously the asymmetric strain distribution^{2,9,11} and composition gradients^{7,10} within the QDs strongly influence the preferential polarization direction. The strain distribution within the QD does not only affect the heavy- and light-hole admixture but also induces a piezoelectric field within the QD.^{5,6} Hereby, the rotational symmetry of the confinement is broken altering the QD electronic structure and thus, the envelope wave functions. Thus, the alignment of the electron and the hole envelope wave functions along the $[110]$ and $[1\bar{1}0]$ directions, respectively, is primarily due to the strain-induced piezoelectric effect.^{4,5,7} This directly induces an absorption anisotropy within the QDs (Ref. 8) and is reflected in a dichroic behavior for the reflection of the sample.

V. SUMMARY

The polarization dependence of the QD differential reflectivity has been studied using polarization-resolved TRDR. The QD differential reflection is asymmetric within the plane perpendicular to the growth direction, i.e., $\Theta_{[1\bar{1}0]}/\Theta_{[110]} = 1.07$, revealing the dichroic character of the QDs. Moreover, the reflection anisotropy strongly depends on the carrier density and decreases from $\rho = 0.14$ to $\rho = 0.06$. The optical anisotropy is confirmed by polarization-resolved PL, which shows $\rho = 0.17$ at low excitation density, and can be explained by the strain-induced asymmetry of the carrier wave functions within the QDs. Due to the reduced symmetry, dichroic reflection of the QD sample is observed.

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