Circular dichroism in InAs/GaAs quantum dots: Confinement-induced magnetism

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Self-assembled InAs/GaAs quantum dots (QDs) are studied by means of circular polarization-resolved photoluminescence and pump-probe differential reflectivity as a function of the excitation density using a linear polarized pump laser. We observe an unexpected large circular polarization anisotropy, which decreases with increasing excitation density for both experimental techniques. Comparison of the respective polarization degrees reveals a ratio of approximately 0.2. Moreover, the reflection measurements reveal circular dichroism with a ratio of $\Theta^+/\Theta^-=1.16$. Circular dichroism in semiconductor QDs can be explained by the induced magnetism as a result of light confinement in QDs, providing an anisotropic coupling between the pump-induced QD exciton and the circularly polarized probe field.

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The realization of high-quality semiconductor quantum structures, e.g., quantum dots (QDs), is one of the major challenges in solid-state physics. Semiconductor QDs have the potential to form building blocks of electronic and optoelectronic devices[1] such as quantum processors.[2,3] In order to apply QDs in new generation devices, the physical properties of these nanostructures must be known. Especially for the realization of spin quantum bits by means of QDs, the knowledge of the spin polarization of confined carriers within the QDs (Refs. 2–5) is of crucial importance. The spin state of the carriers within the QDs can be manipulated by applying magnetic fields or by using circular polarized light as the excitation source. To reveal the carrier-spin polarization, QD nanostructures are usually investigated by means of polarization-resolved photoluminescence (PL) spectroscopy. This because the light emitted by the spin polarized carriers is circularly polarized. More direct approaches for studying the spin polarization of the confined carriers are measurement techniques based on polarization selective absorption. An experimentally more convenient approach is provided by differential reflection spectroscopy,[6] which monitors directly the polarization of the carriers within the QD eigenstates.

In many experimental studies,[7–13] small residual circular polarization anisotropies of the order of a few percent have been observed for Stranski-Krastanow grown quantum dots in the absence of an applied magnetic field. In these experiments, the spin state was initially set by a circular polarized laser. However after spin relaxation, a net polarization is still observed which suggests a residual spin polarization of the confined carriers. A straightforward explanation for such a residual circular polarization anisotropy has not been given. But it is generally believed that it should be due to the strain distribution inside and around the QDs. Although many studies observe a small residual anisotropy, only a few studies explicitly state that these anisotropies are larger than the measurement error.

Pryor and Flatté[14] reported that the observed spin polarization in a photoluminescence experiment, is not necessarily directly connected with the carrier spin polarization within the QD of a similar magnitude. In particular, Pryor and Flatté[14] have shown theoretically that a 100% spin polarization along the [110] direction is converted into a small circular polarization of the emitted PL of the order of 5% due to the QD geometry. Hence, it is convenient to define a more general conversion efficiency ξ between the spin polarization ρ_s and the polarization ρ_{PL} of the emitted luminescence by $\rho_{\mathrm{PL}}=\xi\cdot\rho_s$. With $\rho_{\mathrm{PL},s}$ defined as

$$\rho_{\mathrm{PL}}=(I^+-I^-/I^++I^-) \quad \text{and} \quad \rho_s=(I^\uparrow-I^\downarrow/I^\uparrow+I^\downarrow), \qquad (1)$$

where $I^{+(-)}$ denote the amplitudes of the circular polarization components $\sigma^{+(-)}$, and $I^{\uparrow(\downarrow)}$ the amount of spin-up and spin-down polarized carriers. Although Pryor and Flatté[14] also predict a conversion efficiency near unity ($\xi=0.98$) for light propagating along the [001] axis of the QD, which is the usual PL collection configuration, it is not clear whether or not the conversion efficiency remains large for realistic QDs with an anisotropic strain distribution[15] and with the usual compositional gradients.[16,17] Because the conversion efficiency is not equal to unity and strongly depends on the QD properties, PL is not a proper technique to determine the actual carrier-spin polarization.

Time-resolved differential reflection spectroscopy (TRDR) is expected to measure the QD density of states directly.[18] This means that circular polarized TRDR is able to measure the spin polarization in a more direct way. In this report, we compare the polarization degree obtained by TRDR with the residual circular polarization anisotropy of the emitted photoluminescence of the QD nanostructure. For both experimental techniques (PL and TRDR), we perform polarization-resolved measurements as a function of the excitation density in the absence of an applied magnetic field. We observe an unexpected large circular polarization anisotropy in the TRDR signal with the same sign and a similar excitation density dependence as the observed small circular polarization anisotropy of the PL.

The self-assembled QDs are grown by molecular beam epitaxy on a (100) GaAs substrate.[18] Initially a 295 nm GaAs buffer layer is deposited at 580 °C, followed by a 30 nm GaAs layer grown at 490 °C. Five layers of QDs are grown by the sequential deposition of 2.1 MLs InAs followed by 30 nm GaAs. After the QD growth, the sample temperature is increased up to 580 °C, in order to cap the structure with 137 nm GaAs. The 30 nm GaAs spacer layers are inserted to prevent electronic coupling of the QDs. Atomic force microscopy images of uncovered QDs show the formation of nearly circular quantum dots[19] with a density of 2.8

$\times 10^{10}$ cm^{-2} per layer. We emphasize that the sample is undoped.

Photoluminescence spectra of the QD sample are obtained by using a linear polarized cw-Ti:sapphire (Ti:S) laser as the excitation source, with the photon-energy tuned above the GaAs band gap energy (E_{ex}=1.55 eV). Hereby, carriers with spin polarizations ↑ and ↓ are generated with equal probability. That is, linear excitation yields no preferential spin orientation of the photogenerated carriers. Using a quarter wave plate combined with a linear polarizer in the luminescence channel, we are able to discriminate between σ^+ and σ^- polarized luminescence emitted by the QDs.

Polarization-resolved differential reflectivity measurements are performed using 2 ps probe pulses generated from an optical parametric oscillator synchronically pumped by a mode-locked Ti:S laser with a repetition rate of 76.6 MHz.[18] The linear polarized Ti:S laser is used as the pump source and is mechanically chopped at a frequency of 4 kHz. The pump pulses have a photon-energy higher than the GaAs band gap energy. Hence, the pump laser excites carriers within the GaAs barrier layers. The free carriers diffuse in the GaAs toward the QDs where they are captured and subsequently relax down toward the QD ground state. The population of the QD energy levels changes the dielectric function of the QDs, and hereby, inducing a change of the QD reflectivity.[18] The carrier-induced reflection changes are monitored by tuning the probe laser into resonance with the ground state transition energy of the QD ensemble. The population probability of the QD spin eigenstates ↑ and ↓, are expected to be equal for nonresonant excitation. That is, the photogenerated carriers populating the QD ground state are not expected to have an initial spin polarization or spin memory due to the pump laser. For the polarization-resolved TRDR measurements, the probe field is circularly polarized with polarization σ^+ or σ^-, such that we are able to individually monitor the population of the degenerated eigenstates. Initially, the probe light is linearly polarized. To obtain circular polarized light, a quarter wave plate is used.

Figure 1(a) depicts (circular) polarization-resolved PL spectra of the QD nanostructure for various excitation densities. The PL spectra clearly reveal the QD ground and first excited state, and show a weak circular polarization dependence. Using Eq. (1) the circular polarization anisotropy of the luminescence (ρ_{PL}) is derived and is depicted as a function of the pump intensity in Fig. 1(b). The anisotropy decreases with increasing excitation density[7,20] and has a value of approximately ρ_{PL}=1.5% for $P_{\mathrm{exc}} \rightarrow \infty$, as depicted in Fig. 1(b).

A typical differential reflectivity time trace is depicted in the inset of Fig. 2(a), with the probe laser tuned into resonance with the QD ground state, E_{probe}=1.12 eV. The rise of the signal is governed by the carrier capture and relaxation within the QD (Ref. 18) and has a characteristic time of 15 ps. The decay of the signal is due to carrier recombination with an effective lifetime of 1330 ps. It is important to note that we do not observe any polarization dynamics. That is, no time dependence of the circular polarization degree is observed.

Figure 2(a) depicts the amplitude of the TRDR signal $\left(\frac{\Delta R}{R_0}\right)$ as a function of pump-induced carrier density, using σ^+ and σ^- polarized probe pulses. Initially, the differential reflectivity amplitude increases linearly with the carrier density due to an increasing ground state occupation within the QD ensemble.[18] A deviation from the linear behavior is observed, as the carrier density is increased and the average QD occupation begins to saturate. A similar behavior is observed for TRDR measurements in which linear polarized probe light is used, see Ref. 19. Also here, we ascribe the onset of the saturation to the finite density of states of the QD ensemble. From Fig. 2(a), a strong polarization dependence of the TRDR amplitude is observed with σ^+ as the preferential polarization. This illustrates circular polarization anisotropy of the QD reflectivity, which suggests circular dichroism of the QD ground state.

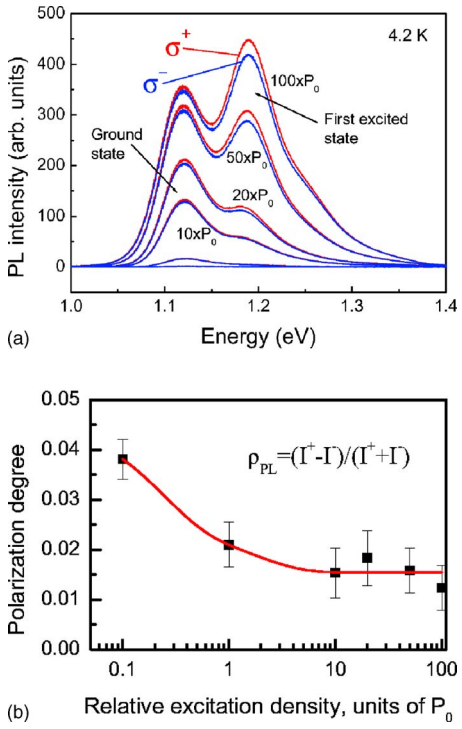

FIG. 1. (Color online) (a) Polarization-resolved PL spectra of the circular polarizations σ^+ and σ^- for various excitation densities in units of P_0. (P_0=20 W/cm^2) (b) Degree of circular polarization anisotropy of the QD ground state transition (ρ_{PL}) versus excitation density in units of P_0. The line is a guide to the eye.

In Ref. 19, a binomial model is developed which describes the level population as a function of the excitation density. The differential signal as a function of the carrier density η can be expressed by a binomial series:

$$\frac{\Delta R}{R_0}(\eta) = \Theta N_{oc}(\eta) = \Theta \frac{[1-(1-\beta)^{\gamma\eta}]}{\beta} \text{ with } \beta = \frac{1}{N_{\mathrm{tot}}}. \qquad (2)$$

Here, Θ denotes the differential reflection per QD and has the dimension of a cross section.[19,21] In Eq. (2), N_{oc} and N_{tot} denote the number of occupied and the total number of QD states, respectively. The fraction of probed QD energy states, which is determined by the probe laser, is taken into account by γ. In Fig. 2(a), we have applied the binomial function [Eq. (2)] to the polarization-resolved TRDR measurement

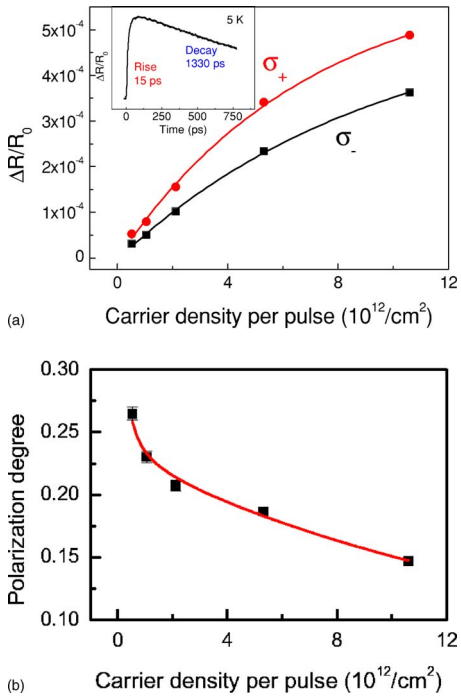

FIG. 2. (Color online) (a) Amplitude of the QD differential reflection, as illustrated by the inset, versus the pump-induced carrier density per pulse for probe polarization σ^+ and σ^-, including the experimental fits by using Eq. (2). (b) The degree of circular polarization anisotropy of the QD reflectivity ($\rho_{\Delta R}$) versus the carrier density.

results. From the experimental fits with $N_{\mathrm{tot}}=2.8\times10^{11}\ \mathrm{cm}^{-2}$ for the five layers, we obtain $\Theta^+=2.23\times10^{-15}\ \mathrm{cm}^2$ and $\Theta^-=1.93\times10^{-15}\ \mathrm{cm}^2$. The different cross sections reveal a circular polarization anisotropy with a ratio of $\frac{\Theta^+}{\Theta^-}=1.16$. This ratio is by definition also equal to the circular dichroism. In addition, our analysis indicate that the QD ensemble has an initially preferential polarization which induces the excitation power dependence.[19]

As is shown in Fig. 2(b), the polarization anisotropy of the TRDR signal ($\rho_{\Delta R}$)—analogous to Eq. (1)—depends strongly on the pump-induced carrier density and decreases from 0.27 down to 0.15 with increasing carrier density within our range of pump excitation. We note that the excitation power dependence in PL and TRDR are very similar, providing a strong indication that the polarization dependence of the QD PL and reflection are correlated. TRDR is able to probe the occupation of the QD density of states and the polarization of the carrier wave functions,[18] and because the emission of circular polarized light is directly related to the carrier-spin polarization within the QD, we suggest that the expression $\rho_{\mathrm{PL}}=\xi\cdot\rho_s=\zeta\cdot\rho_{\Delta R}$ is justified. Here ξ and ζ are constants. The fit included in Fig. 2(b) is derived from the experimental fits of Fig. 2(a) by using Eq. (1). In the limit $\eta\rightarrow\infty$, i.e., for complete QD level saturation, a degree of polarization anisotropy is deduced which has a value of approximately $\rho_{\Delta R}=7.2\%$. A comparison of our results obtained by luminescence and differential reflectivity measurements, Figs. 1 and 2, respectively, reveals a nonunity conversion efficiency of $\zeta\approx0.2$.

As is mentioned above, circular polarized PL is not expected for linear polarized excitation in the absence of a magnetic field. However, a small net polarization is observed for the QD ground state and it is even more pronounced for the first excited state (Fig. 1). Misalignment of the quarter wave plate might cause the observed polarization, however, we have carefully eliminated this in the measurement setup. The observed anisotropy of 1.5% at high pump powers is in the same order as our measurement error. Therefore, it is rather difficult to make a statement with respect to the absolute value of the polarization degree. On the other hand, the measurement error cannot explain the excitation density dependence of the anisotropy.

Quantum coherence of the polarized eigenstates[11,13,22,23] might be a possible origin of the circular dichroism as is deduced from Fig. 2. In this case, quantum beats would arise in the time traces of the differential reflection measurements. However, this phenomenal effect is not observed in our TRDR experiments. To monitor quantum beats, a coherent superposition of the linear polarized eigenstates must be created by means of (quasi)resonant excitation. We use highly nonresonant excitation pulses, and in addition, we do not expect to observe a dynamical dichroism.[11,23] Instead of a transient dichroism, we measure the steady-state circular dichroism. From this we conclude that the polarization anisotropy is not due to quantum coherence, but the results imply that the observed circular dichroism is an intrinsic property of self-assembled QDs.

In order to explain the observed anisotropy, two mechanisms are briefly discussed which are based on carrier and light confinement, respectively. Although InAs/GaAs QDs are nonmagnetic, it has been reported[24] that a deviation of the QD confinement from a circular shape, that is a change of the QD geometry or a reduction of the QD circular symmetry, may result in a piezomagnetic behavior.[25] This means that shape, strain and/or composition gradients can induce a magnetization. Hence, the net spin of the carriers is governed by the spatial confinement of the carriers within the QDs.[14,24]

An other mechanism which plays a role in the interaction of the applied electromagnetic field and the QDs, is the light confinement-induced magnetism.[25–27] Let us approximate a self-assembled QD by a dielectric sphere with radius R_{QD}, which is much smaller than the wavelength of the electromagnetic wave in vacuum ($R_{QD}\ll\lambda_0$). In addition, we assume nonmagnetic QDs (i.e., $\mu_{\mathrm{QD}}=\mu_0$). The QD permittivity can be written in the form of a Lorentzian,[28] which has at resonance a value much larger than one, $|\varepsilon_{\mathrm{QD}}|\gg1$. In this case, the electric part of the absorption is reduced, but the magnetic part of the absorption is enhanced and is not negligible.[25] State filling of the QD ground state by the pump-generated carriers changes the local QD permittivity ($\varepsilon_{\mathrm{QD,peak}}\sim10^4$),[26] and in turn changes the diffraction of the electromagnetic field by the QDs. Thus, it is expected that both the electric and the magnetic polarizability of QDs are strongly changed due to the confined carriers.[29,30] A result of the large carrier induced changes of the local permittivity is that the wavelength of the resonant light inside a QD becomes comparable with the QD size, $\lambda_{\mathrm{QD}}\sim R_{\mathrm{QD}}$. This ultimately leads to light confinement inside a QD,[26,31] and subsequently, the photon strongly couples with the QD exciton.

A QD polariton is formed.[31,32] In this QD polariton picture, electromagnetic field scattering due to the magnetic polarizability of the QDs—by the induced nonzero magnetic momentum of the QDs—predicts induced magnetism.[26,27] The theoretical model[26,27] predicts that the lifetime of the induced magnetism is in the order of 10^{-5} s and exceeds the exciton lifetime. We note that the magnetic resonance lifetime should not be confused with the exciton lifetime. In our TRDR measurements, we observe a circular dichroism which does not decay at the time scale of the experiment. Thus, our observation of a steady-state dichroism is in agreement with the predicted long lifetime of confinement-induced magnetism.

The observation of circular polarization anisotropy in QD reflectivity suggests that electromagnetic fields with σ^+ or σ^- polarization of the electric part couple to the QD exciton with different strengths. Although a homogeneous and spherical QD will not discriminate between the two polarizations, we have to remark that realistic QDs are highly nonspherical and inhomogeneous. This does not only manifest in the linear polarization anisotropy,[19] but also in the spin-to-PL conversion as reported by Pryor and Flatté.[14] Therefore, we expect that the observed circular dichroism monitors the coupling strength of the QD exciton with the polarized photon. This suggests left- and right-handed QD polaritons.

In summary, we have observed circular polarization anisotropy of the QD luminescence and differential reflectivity. From the comparison of the polarized TRDR signal with polarized PL as a function of excitation density, we deduce a ratio of approximately 0.2. Polarization-resolved reflectivity measurements reveal circular dichroism of the QD ground state with a magnitude of 1.16. The observation of circular dichroism in InAs/GaAs QDs can be explained by the induced magnetism due to light confinement which results in the anisotropic coupling between the QD exciton (generated by the pump laser) and the circularly polarized electromagnetic field of the probe laser. Hence, left- and right-handed QD polaritons are created.

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