Circular-to-Linear and Linear-to-Circular Conversion of Optical Polarization by Semiconductor Quantum Dots

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We report circular-to-linear and linear-to-circular conversion of optical polarization by semiconductor quantum dots. The polarization conversion occurs under continuous wave excitation in the absence of any magnetic field. The effect originates from quantum interference of linearly and circularly polarized photon states, induced by the natural anisotropic shape of the self-assembled dots. The behavior can be qualitatively explained in terms of a pseudospin formalism.

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Quantum dots (QDs) are essentially zero-dimensional semiconductor nanostructures that exhibit an atomiclike line spectrum in the optical frequency range and are, therefore, often referred to as artificial atoms. Their small (nanometer-scale) size in combination with their strong interaction with light has led to speculations about possible applications of QDs in optical quantum computation [1,2].

While most device concepts assume highly symmetric dots, it is experimentally well established that self-assembled semiconductor QDs often grow in a highly anisotropic manner, reducing the point-group symmetry of a single QD to C_{2v} or still further. This natural shape anisotropy [3] is reputed to be unwanted, and its consequences for the physics of the system have attracted only limited interest. In this Letter, we demonstrate novel physics that is the direct result of the low in-plane symmetry of the QDs. We observe conversion of the polarization of optical radiation from circular to linear (and *vice versa*) mediated by QDs. The low symmetry of the dots naturally induces quantum interference between linear and circular-polarized photon states. Time resolved experiments would result in quantum beats in the polarization, while, under the steady-state conditions that we examine, a net conversion results. The cw effect has strong analogies with the Hanle effect, be it that our results are all obtained without an external magnetic field.

The CdSe/ZnSe QDs used in our experiments are grown by molecular beam epitaxy [4]. A 0.3 nm-thick CdSe layer is deposited on top of a 50 nm-thick ZnSe buffer at a substrate temperature of 300 °C. A growth interrupt of 10 seconds prior to capping by 25 nm ZnSe results in the formation of the CdSe dots by self-assembly. Typically, these dots are 1 nm high and sub-10 nm in lateral dimensions but with a high areal density (above 10^{11} cm^{-2}). In order to image the QDs using atomic force microscopy (AFM), an uncapped sample has also been grown. The AFM image of this sample, presented in Fig. 1(a), shows distinct elongated islands. The dots are preferentially oriented along the [110] direction, in agreement with the optical characterization discussed below. This is quite similar to earlier studies on monolayer-fluctuation QDs [5]. The preferential orientation implies that the ensemble of dots has a net spatial anisotropy which, as we will show in the following, is essential for the polarization conversion. The average symmetry of the ensemble of dots is reduced to C_{2v}, as compared with the full T_d symmetry of the zincblende bulk lattice and the D_{2d} group of the corresponding quantum well.

For optical excitation, we use a stilbene-3 dye laser, pumped by the ultraviolet lines of an Ar-ion laser. For nonresonant excitation, the laser energy is tuned to $E_{\mathrm{exc}} = 2.83$ eV, exceeding the band gap of the ZnSe barrier. A typical photoluminescence (PL) spectrum of the QDs under nonresonant excitation is shown in Fig. 1(b) as the filled area. The PL band of 30 meV width has a maximum at $E_0 = 2.665$ eV. For the angle dependent polarization

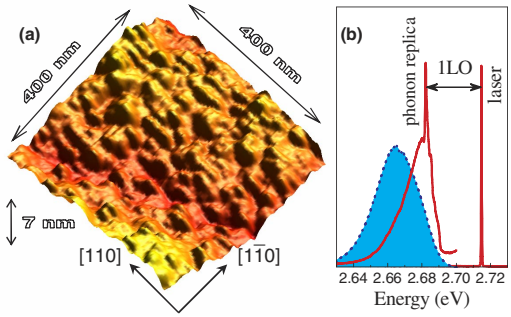

FIG. 1 (color online). Characterization of CdSe/ZnSe quantum dots. (a) Atomic force microscope image of a CdSe/ZnSe quantum dot layer. The QDs are alongated along the [110] axis. (b) PL spectra for nonresonant (filled area under dotted curve) and resonant (solid curve) excitation, respectively. The phonon replica is well resolved in the PL spectrum as a narrow peak separated from the laser line by the LO-phonon energy, which is 32 meV in ZnSe.

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data we discuss below, the polarization is detected at the maximum of the PL band, but we have verified that the degree of polarization does not vary strongly over the band. Resonant excitation of the CdSe QDs is obtained for $E_{\mathrm{exc}} = 2.714$ eV. In this case, the polarization of the PL is detected at the phonon replica, which now can be clearly resolved in the emission spectrum [solid curve in Fig. 1(b)]. Since the QDs are nominally undoped, most of them will be uncharged at these excitation conditions.

In order to investigate the in-plane optical anisotropy of the QDs, the sample is mounted on a rotating holder. Its orientation is controlled using a stepping motor with an accuracy better than 1°. Rotation-angle dependent scans of the PL polarization in the laboratory frame are carried out using fixed analyzers (Glan-Thompson prisms) and a conventional optical setup consisting of a photoelastic modulator operating at frequency $f = 50$ kHz and a two-channel photon counter. The circular polarization $\rho_{\mathrm{circ}}^{\mathrm{lab}}$ is detected at f, and the linear polarization $\rho_{\mathrm{lin}}^{\mathrm{lab}}$ is detected at the double frequency $2f$. The measured polarization degrees in the lab frame are linked to those in the sample frame $[\rho_{l'}, \rho_l, \rho_c]$ as follows: $\rho_{\mathrm{circ}}^{\mathrm{lab}} = \rho_c$; $\rho_{\mathrm{lin}}^{\mathrm{lab}} = \rho_{l'} \times \cos 2\alpha - \rho_l \sin 2\alpha$. Here $\rho_{l'} = (I_{[110]} - I_{[1\bar{1}0]})/(I_{[110]} + I_{[1\bar{1}0]})$ and $\rho_l = (I_{[100]} - I_{[010]})/(I_{[100]} + I_{[010]})$, and α is the angle between the sample and laboratory coordinate frames. For noise reduction, all optical experiments are performed at a temperature of 1.6 K. No magnetic fields are applied.

The absorption of photons by the QDs results in the formation of excitons, where the polarization of the photons is linked to the spin states of the exciton. The confinement of an exciton in the small volume of a QD leads to an enhancement of the electron-hole exchange interaction. Because of the low symmetry of our QDs, this results in an anisotropic exchange splitting $\hbar\Omega$. Typically, for CdSe/ZnSe QDs, $\hbar\Omega \sim 0$–0.5 meV [6,7]. This splitting can be directly observed in the photoluminescence spectrum of a single QD through the occurrence of doublet emission lines. When an ensemble of QDs is probed, the exchange splitting is buried in the much larger ($\sim$ 30 meV) inhomogeneous broadening of the PL band [Fig. 1(b)]. However, for nonresonant excitation, above the ZnSe barrier ($E_{\mathrm{exc}} = 2.83$ eV), the anisotropic exchange splitting manifests itself as a built-in linear polarization. Figure 2(a) shows the degree of linear polarization measured in a fixed coordinate basis while the sample is rotated by an angle α. The polarization oscillates as $\cos 2\alpha$, just as would be observed for a linear polarizer. As can clearly be seen from the polar plot in the inset in Fig. 2(a), the polarization axis is linked to the [110] crystallographic direction, and it does not depend on the handedness of the polarization of the exciting light. This behavior is what one intuitively expects from the shape of the QDs found in Fig. 1(a).

However, more counterintuitive results are obtained under quasiresonant excitation ($E_{\mathrm{exc}} = 2.714$ eV). The

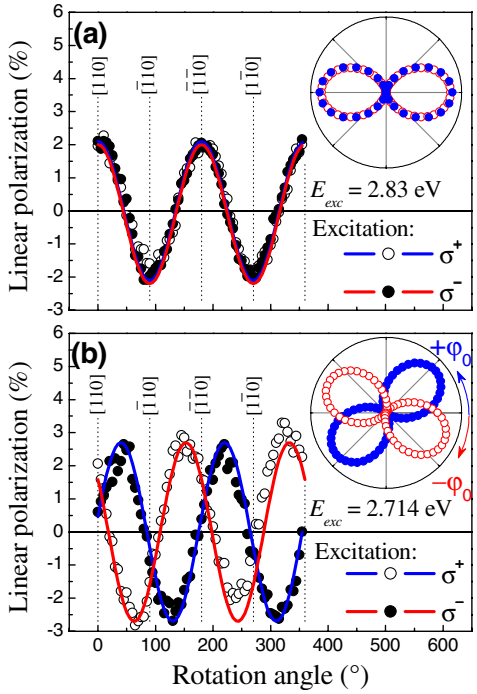

FIG. 2 (color online). Circular-to-linear polarization conversion by CdSe/ZnSe QDs. (a) Angle scans of the linear polarization detected at the PL maximum under nonresonant excitation above the ZnSe barrier ($E_{\mathrm{exc}} = 2.83$ eV) with σ^+ (open symbols) and σ^- (solid symbols) circularly polarized light. The solid curves are fits assuming $\rho_0 \cos 2\alpha$. (b) Angle scans of linear polarization detected at the phonon replica under σ^+ (open symbols) and σ^- (solid symbols) circularly polarized resonant excitation ($E_{\mathrm{exc}} = 2.714$ eV). The solid curves are fits assuming $\rho_0 \cos(2\alpha \mp 2\varphi_0)$, $2\varphi_0 = 67°$. The insets in all panels show the same data (but shifted by a constant of ρ_0 to positive values) in polar coordinates. Zero rotation angle in all panels means that the linear analyzer is orientated parallel to the [110] crystallographic direction. The magnetic field for all data is zero.

PL spectrum of the QDs is now dominated by a narrow peak that we attribute to a phonon replica of the laser line [Fig. 1(b)]. It appears due to fast excitonic recombination combined with the emission of an LO phonon. Under these conditions, the polarization axis is no longer fixed to the [110] crystalline direction. As shown in Fig. 2(b), the angle dependence of the linear polarization now varies as $\cos(2\alpha \mp 2\varphi_0)$, where the sign depends on the handedness of the circularly polarized excitation light and $2\varphi_0 = 67°$. This behavior is ever so more clearly apparent from the polar plot in the inset in Fig. 2(b). The polarization axis is rotated away from [110] by an angle φ_0, counterclockwise towards the [010] direction for σ^+, and clockwise towards the [100] direction for σ^- polarization of the incoming light. Such a behavior implies, indeed, circular-to-linear polarization conversion.

In order to estimate the conversion value under $\sigma^\pm$ circular-polarized excitation, denoted by $P_c = \pm 1$, we describe the total polarization of the emitted light by a vector $[\rho_{l'}, \rho_l, \rho_c]$ inside a Poincaré sphere defining a

novel type of quasispin or a two-level system. Here $\rho_{l'}$ is the linear polarization along [110], ρ_l is the linear polarization along [100], and ρ_c is the circular polarization. These Stokes coordinates satisfy $\sqrt{\rho_{l'}^2 + \rho_l^2 + \rho_c^2} \leq 1$. As efficient conversion, we define the condition $\rho_l > \rho_{l'}$ and $\rho_l > \rho_c$. According to Fig. 2(b), the maximum amplitude of the linear polarization is $\rho_0 = \sqrt{\rho_{l'}^2 + \rho_l^2} = 2.7\%$, so we have $\rho_l = \rho_0 \sin 2\varphi_0 = 2.5\%$ and $\rho_{l'} = \rho_0 \cos 2\varphi_0 = 1.0\%$. We have also measured the optical orientation [8], i.e., the degree of circular polarization of the emitted light under circularly polarized excitation, and obtained $\rho_c \approx 1\%$. For the experimental values, the above condition of efficient conversion is obviously fulfilled.

Polarization conversion in low-dimensional systems has been predicted by Ivchenko *et al.* [9]. In the presence of a preferential direction for the excitonic states in QDs, the circularly and linearly polarized contributions to the emission can show quantum interference (e.g., quantum beats in the time domain). Obviously, an external magnetic field can induce this preferential direction. Meanwhile, magnetic field-induced polarization conversion has been demonstrated experimentally in superlattices [10] and QDs [11]. However, using the anisotropic exchange interaction to define the preferential direction induces a beating of the circular and the [100] linear polarizations even in zero magnetic field. In the simplest case, the time evolution after circularly polarized excitation P_c at $t = 0$ can be expressed as $\rho_c(t) = P_c \cos(\Omega t) \exp(-t/\tau_s)$ and $\rho_l(t) = P_c \sin(\Omega t) \exp(-t/\tau_s)$. The circular and linear polarizations thus beat in antiphase, decaying with spin coherence time τ_s to zero. This has been partly verified previously in quantum beat experiments [12–14], where precession of the linear (circular) polarization component excited with linearly (circularly) polarized light at Larmor frequency Ω was observed.

Within the pseudospin formalism [10,15], the Stokes coordinates in the Poincaré sphere are linked to a pseudospin $\mathbf{S} = [S_1, S_2, S_3]$ by the simple relation

$$\rho_{l'} = S_1, \qquad \rho_l = S_2, \qquad \rho_c = S_3. \tag{1}$$

The $S_1/2$, $S_2/2$, and $S_3/2$ behave as x, y, and z projections of a spin in real space. In zero magnetic field, the pseudospin Hamiltonian can be written in the form

$$\mathcal{H} = \frac{\hbar}{2} \Omega \sigma_x, \tag{2}$$

where σ_x is the Pauli matrix. The dynamics of the polarization of the PL described by the vector $\mathbf{S}$ after $\mathbf{P_{ex}}$-polarized excitation is given by [8]:

$$\frac{\partial \mathbf{S}}{\partial t} = \mathbf{\Omega} \times \mathbf{S} - \frac{\mathbf{S} - \mathbf{P_{eq}}}{\tau_s} - \frac{\mathbf{S} - \mathbf{P_{ex}}}{\tau_0}. \tag{3}$$

Here τ_0 is the exciton lifetime and $\mathbf{P_{eq}}$ is equilibrium polarization of the emission. According to our Hamiltonian (2), $\mathbf{\Omega} = [\Omega, 0, 0]$, and $\mathbf{P_{eq}} = [\Upsilon_{\text{lin}}, 0, 0]$, where the built-in linear polarization Υ_{lin} originates from the linear dichroism of the QDs and thermal population of the exchange-split states. Equation (3) can be solved for steady-state conditions (i.e., under cw excitation) when the PL is excited by arbitrarily polarized light $\mathbf{P_{ex}} = [P_{l'}, P_l, P_c]$, yielding

$$\begin{aligned} \rho_{l'} &= \frac{T}{\tau_0} P_{l'} + \frac{T}{\tau_s} \Upsilon_{\text{lin}}, \\ \rho_l &= \frac{T}{\tau_0} \left[-\frac{\Omega T}{1 + (\Omega T)^2} P_c + \frac{1}{1 + (\Omega T)^2} P_l \right], \\ \rho_c &= \frac{T}{\tau_0} \left[+\frac{1}{1 + (\Omega T)^2} P_c + \frac{\Omega T}{1 + (\Omega T)^2} P_l \right], \end{aligned} \tag{4}$$

where $T^{-1} = \tau_s^{-1} + \tau_0^{-1}$. The second identity describes, indeed, circular-to-linear polarization conversion for the circularly polarized excitation ($P_{l'}, P_l = 0$ and $P_c = \pm 1$).

Equations (4) are simplified, since some factors (such as dark exciton states, heavy and light hole mixing, etc.) cannot be considered within the framework of a pseudospin formalism. Nevertheless, they give a qualitative description for the QD conversion mechanism. The third identity in Eqs. (4) with $P_l = 0$ is very similar to the Hanle effect, with the Zeeman splitting induced by a magnetic field replaced by the zero-field anisotropic exchange splitting. In quantum dots, the anisotropic exchange splitting $\hbar\Omega$ is an order of magnitude larger than in superlattices [10]. As a result, the conversion efficiency under cw excitation is significant. The conversion efficiency is characterized by the factor $K = \rho_l/\rho_c = \langle\Omega\rangle T$, which differs from the conversion value ρ_l/P_c (we deal with $P_c = \pm 1$). The efficiency with $K > 1$ implies the converted polarization of the emission is larger than the polarization of the emission in the initial excitation configuration. For quasiresonant excitation, $K \approx 3$ is found.

We note that the QD ensemble is inhomogeneous; i.e., the anisotropic exchange splitting varies from dot to dot. This is important, as about 10^8 dots are probed simultaneously. The conversion depends nonmonotonically on Ω: $\rho_l \to 0$ for $\Omega = 0$ or $\Omega \to \infty$. So only a fraction of the QDs contributes significantly to the conversion. By comparing Eqs. (4) with the experiment in Fig. 2(b), we deduce a typical value $\sqrt{\langle\Omega^2\rangle} T \approx 10$. This corresponds to an anisotropic exchange splitting $\hbar\Omega$ on the order of tens of μeV with the exciton spin lifetime T being on the order of a hundred picoseconds [16]. It also follows from Eqs. (4) that, for $\Omega T = 1$ and $\tau_0 \ll \tau_s$, the polarization reaches $\rho_c = \rho_l = 50\%$. However, such an effectual QD converter requires a very small anisotropic exchange splitting (on the order of μeV) and additional technological efforts are probably needed for the realization.

Equations (4) successfully explain the polarization behavior presented in Fig. 2. According to Eqs. (4), the observation of non-neglible conversion in Fig. 2(b) implies $\tau_0 \approx T$. This condition is in agreement with the generally expected long spin coherence time in the QD ground state

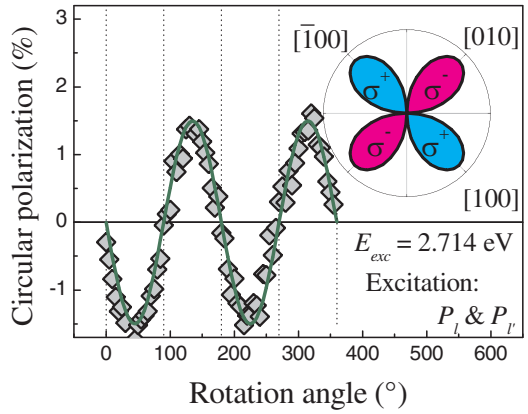

FIG. 3 (color online). Linear-to-circular polarization conversion by CdSe/ZnSe QDs. It reveals itself in an angle scan of circular polarization detected at the phonon replica under linearly polarized resonant excitation ($E_{\rm exc} = 2.714$ eV). The curve is a fit, assuming $\rho_0 \sin 2\alpha$. The inset shows the absolute value of the same data $|\rho_0 \sin 2\alpha|$ in polar coordinates. Zero rotation angle means that the linear polarizer is orientated parallel to the [110] crystallographic direction. The magnetic field is zero.

under quasiresonant excitation [17,18]. For example, the spin relaxation time of a single hole was found to be about 10 ns [19]. We note that the spin coherence time τ_s decreases notably with increasing temperature, leading to a reduction of the conversion value and efficiency. In the case of excitation above the ZnSe barrier ($E_{\rm exc} = 2.83$ eV), an electron and a hole are trapped by a QD independently, so they do not form a coherent spin state. As a consequence, Fig. 2(a) shows no conversion at all, $2\varphi_0 = 0°$.

The most intriguing effect is the counterconversion, i.e., conversion from linear-to-circular polarization, which can occur for our dots due to time reversal symmetry. Indeed, we observe this effect, as shown in Fig. 3. With linear polarized excitation along [100] ($P_{l'} = 0$ and $P_l = 1$), σ^+ polarized emission appears. The effect changes sign to σ^- when excited along [010] ($P_{l'} = 0$ and $P_l = -1$). No conversion is observed when the linear polarizer at the excitation was oriented along [110] or [1$\bar{1}$0] directions ($P_{l'} \neq 0$ and $P_l = 0$). This behavior is in good qualitative agreement with Eqs. (4) and obeys similar formulas as circular-to-linear polarization conversion upon interchange of the indices $l \leftrightarrow c$ and reversing the sign.

All experiments discussed above were obtained for quantum dots containing no electrons. In negatively charged QDs, containing a single extra electron, the anisotropic exchange splitting is modified drastically. With a photocreated electron, the extra electron forms the energetically favorable singlet state with zero total electron spin. Since the electron-hole exchange interaction is proportional to the spins [20] of electrons and holes, the anisotropic exchange splitting in a charged QD equals exactly zero ($\hbar\Omega = 0$). By applying a bias voltage, additional electrons can be pushed into or out of the QDs [21]. This may provide extra functionality to the QD converter and may provide a flexible approach for spin-based electro-optical devices.

In summary, we have demonstrated efficient circular-to-linear and linear-to-circular light polarization conversion by quantum dots. The conversion occurs in zero magnetic field and is induced by anisotropic exchange splitting. For optimized QD dimensions, conversion values up to 50% can be achieved. An important advantage of the QD converter is the possibility of control of the optical activity by charging the dots by application of a bias voltage. Our findings may have obvious practical applications in information processing, as the dots can easily be integrated in semiconductor circuits.

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