Monitoring electrically driven cancellation of exciton fine structure in a semiconductor quantum dot by optical orientation

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We use optical orientation technique to monitor the degeneracy control of exciton states in a single InAs/GaAs quantum dot, achieved by applying an in-plane electric field. Under circularly polarized quasiresonant excitation, the exciton photoluminescence shows a pronounced maximum of circular polarization at electric field corresponding to zero fine structure splitting. By analyzing the width of this maximum we are able to determine the homogeneous linewidth of the excitonic transition. This experimental method is shown to be very efficient to test and possibly tune the photonic properties of an individual quantum dot for the emission of entangled photon pairs. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2805025]

Semiconductor quantum dots (QDs) attract a great attention for their possible applications in quantum cryptography and quantum information processing. In particular, emission of polarization-entangled photon pairs has been a highly competitive field, since Benson *et al.* proposed in 2000 to use the biexciton-exciton cascade in a single quantum dot.[1] However, in this pioneering work, they could prove only classical polarization correlation but no quantum entanglement. This is due to the neutral exciton (X_0) fine structure splitting (FSS) caused by the QD anisotropy.[2–5] The value of the FSS (denoted by δ_1) usually amounts to a few tens of μeV, which largely exceeds the homogeneous linewidth of the transition, introducing thus distinguishability between both paths of the cascade. In 2006, entanglement was achieved by canceling the FSS of a dot thanks to special growth techniques in combination with an in-plane magnetic field.[6] However, the two-photon state tomography, routinely used in such experiments, is very time consuming. Therefore an experimental technique must be developed to select and prepare specific QDs with the required photonic properties.

In this letter, we show that optical orientation under quasiresonant excitation represents an efficient tool to evidence exciton degeneracy in a given QD, achieved here by tuning the native FSS with an in-plane electric field. The exact cancellation of the FSS is characterized by the conservation of any light polarization used to prepare an exciton in the QD, provided no decoherence mechanism due to the environment destroys the exciton polarization in the timescale of the radiative recombination. By introducing the formalism of 1/2 pseudospin to describe the two-level bright exciton states ($|x\rangle$, $|y\rangle$ Ref. 7) this condition reads $\tau_s \gg \tau_r$, where τ_s (τ_r) is the exciton spin (radiative) lifetime, respectively. Measuring the circular polarization of exciton PL is of high interest, as it provides a straightforward means to verify the values of FSS and effective polarization relaxation, which are both detrimental to polarization entanglement. Furthermore, measurements of the optical orientation of excitons versus electric field give in principle access to the homogeneous excitonic linewidth ($\hbar/\tau_r \sim 1$ μeV) which is more than one order of magnitude smaller than the resolution of typical microphotoluminescence (μ-PL) setup used for single QD spectroscopy.

For the optical orientation experiments, the exciton spin state was prepared by circularly polarized (σ^+) 2LO-phonon-assisted excitation, namely, 70 meV above the ground state transition [see Fig. 1(a)]. This quasiresonant excitation scheme provides in principle an efficient transfer of light polarization into the exciton polarization because the electron-phonon interaction is spin independent. When the exciton recombines, its spin polarization is transferred into the polarization of the emitted photon. Such optical orientation was clearly observed for neutral excitons in a longitudinal magnetic field when the Zeeman splitting exceeds δ_1,[8,9] and for charged excitons which have intrinsically zero FSS due to Kramers degeneracy.[10–12] For neutral excitons in zero magnetic field, circularly polarized excitation produces a coherent superposition of the linearly polarized eigenstates ($|x\rangle$, $|y\rangle$) separated by δ_1, as depicted in Fig. 1(a). In cw experiments, one therefore cannot obtain a strong degree of circular polarization because these two components dephase before recombination producing quantum beats at the angular frequency $\delta_1/\hbar$.[13–15] These beatings represent a clock which measures the effective lifetime $T=(\tau_r^{-1}+\tau_s^{-1})^{-1}$ of the exciton polarization. Indeed, when neglecting exciton thermalization during the exciton lifetime (here, $\delta_1/k_BT<0.01$ and $\tau_s>\tau_r$), the time-averaged degree of PL circular polarization is theoretically given by[7,16]

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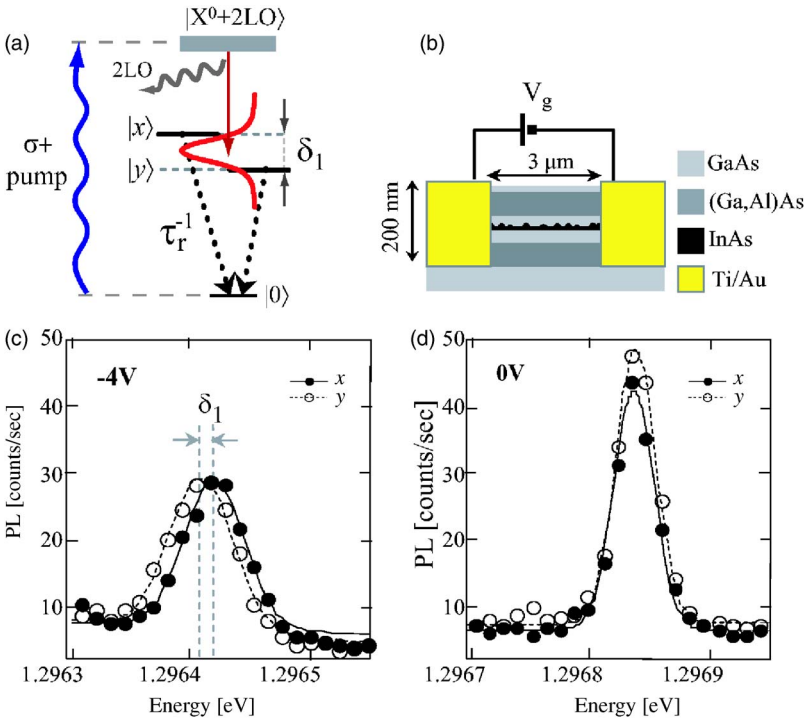

FIG. 1. (Color online) (a) Scheme of 2LO-phonon assisted quasiresonant excitation. (b) Scheme of the sample structure processed to apply an in-plane electric field. [(c) and (d)] Excitonic line measured for two different biases: (c) −4 V and (d) 0 V for two orthogonal linear polarizations corresponding to the QD eigenstates $|x\rangle$ and $|y\rangle$. Points (open and closed)—experimental data and lines (dashed and solid)—Gaussian fits.

$$P_{\mathrm{circ}} = \frac{\eta P^0_{\mathrm{circ}}}{1 + (\delta_1 \eta \tau_r/\hbar)^2}, \tag{1}$$

where $\eta = \tau_s/(\tau_s + \tau_r)$ and P^0_{circ} is the effective initial circular polarization determined by the laser polarization, but possibly reduced during the secondary emission of LO phonons. Let us notice a direct correspondence of this formula to the well-known Hanle effect,[16,17] with δ_1 playing the role of the Zeeman splitting in the transverse magnetic field. By tuning δ_1 with an electric field and monitoring the PL circular polarization we are thus able to detect the restoration of the exciton degeneracy with the precision corresponding to the homogeneous linewidth $\hbar/T$. This accuracy is precisely that required for applicability of quantum dot to emission of entangled photon pairs. Like in Hanle experiments, by fitting with Eq. (1) the polarization dependence on δ_1 one can determine the effective exciton polarization lifetime which is masked by inhomogeneous broadening and spectral resolution in μ-PL spectroscopy.

To achieve an *in situ* control of the FSS we applied an in-plane electric field, a technique which has proven efficient by several groups.[18–20] A sample containing a single layer of InAs QDs grown by molecular beam epitaxy was processed with lateral electrical gates (see Ref. 18 for details). A scheme of the structure is shown in Fig. 1(b). Polarization-resolved μ-PL measurements were performed on single InAs/GaAs quantum dots at low temperature ($T \sim 10$ K).[21] We found a specific QD with a small FSS that moreover could be tuned through zero.[20,22] The FSS was measured using linearly polarized detection along the two components of the excitonic doublet (under excitation linearly polarized at 45°) as illustrated in Figs. 1(c) and 1(d). Figure 2(a) shows the changes of FSS induced by the in-plane electric field. Although the typical line width amounts to ~35 μeV (due to the setup spectral resolution), we can determine the central position of excitonic lines with a much higher precision, which mostly depends on the signal to noise ratio of the PL intensity. In practice, the standard deviation of FSS measurements in Fig. 2(a) reduces to only ~3 μeV. This precision is still not sufficient to determine when the splitting is effectively equal to zero for a given bias, but the quadratic fit of the FSS evolution indicates that the splitting should vanish at −1.7 V. Remarkably, this conclusion is confirmed in a second series of measurements by monitoring the exciton optical orientation as discussed below. Note the strength of the applied field was limited by the reduction of luminescence intensity [by a factor of 2 at −4 V, see Fig. 1(c)], which is accompanied by a broadening of the PL lines [see Fig. 2(b)]. The field-induced escape of electrons from the quantum dot likely explains the loss of PL signal, but not the strong broadening of the PL lines, which is most probably due to the spectral diffusion caused by charge fluctuations in the QD vicinity.[22] The coincident increase of both FSS and line width below −3 V is certainly accidental as it was not observed for other QDs.

Under quasiresonant pumping of the exciton level with σ^+ light, we observed a clear enhancement of the degree of circular polarization for the applied voltage corresponding to zero crossing, with a maximum value of about 70%. Remarkably, the circular polarization resonance plotted as a function of fine structure splitting (δ_1) can be fitted with the Lorentzian function in Eq. (1) as shown in Fig. 3. Note we

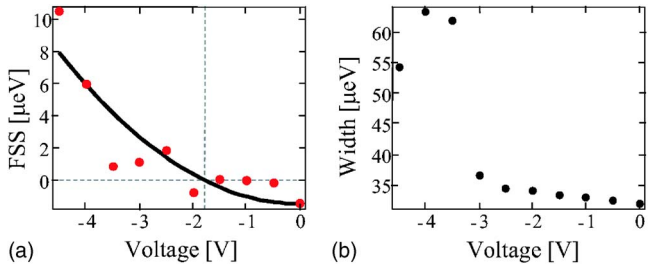

FIG. 2. (Color online) (a) Fine structure splitting as a function of applied bias (measured as the difference in the excitonic line position for two orthogonal linear polarizations of detection): points—experimental data and solid line—quadratic fit. (b) Linewidth vs applied voltage.

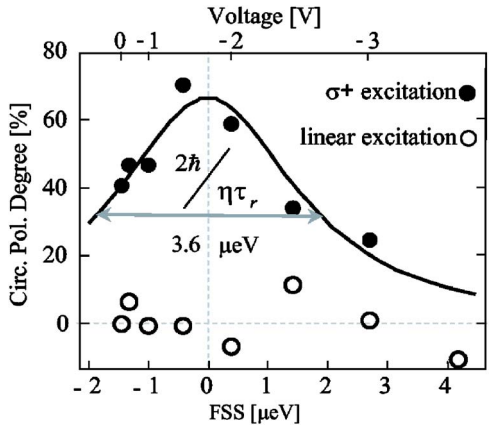

FIG. 3. Degree of circular polarization vs voltage (upper scale) FSS [from the quadratic fit of Fig. 2(a)] under circularly polarized $\sigma+$ and linearly polarized at 45° excitation, respectively. Points—experimental data and solid lines—Lorentzian fit.

limited ourselves to the range of bias for which neither the line intensity nor the linewidth are dramatically affected by the electric field. This guarantees that only the splitting δ_1 experiences appreciable variations while other parameters in Eq. (1) remain essentially constant. Within our model, two parameters are determined from the fit: the maximum polarization degree $\eta P^0_{\mathrm{circ}}=0.66\pm0.05$ and the resonance full width at half maximum $2\hbar/\eta\tau_r=3.6\pm0.3$ μeV (with errors of the fitting procedure). Neglecting any polarization loss in the two phonon quasiresonant excitation ($P^0_{\mathrm{circ}}=1$) would lead to estimates of $\tau_r=0.6$ ns and $\tau_s=0.97$ ns. The τ_r value reasonably compares with data available from time-resolved measurements[10,23] or transmission spectroscopy on similar QDs.[24] The value of τ_s may actually be larger if other broadening mechanisms are taken into account, e.g., the effective exciton lifetime may be reduced due to the in-plane electric field and possible charge fluctuations in the vicinity of the QD. As an improvement of the measurement presented here,[25] using an excitation at 1LO phonon above the exciton ground state should increase the initial polarization state close to 100%, enabling the absolute determination of both lifetimes. Note that similar effects were observed on a few other QDs. When the FSS cancellation was achieved, a strong enhancement of circular polarization was found, but when it was only reduced, (to ~2 μeV) we solely measured a small increase of circular polarization without clear resonance.

In conclusion, optical orientation was used to probe the spin degeneracy of the QD excitonic levels, which was controlled by in-plane electric field. The experiments were performed under quasiresonant excitation at 2LO phonons above the ground exciton state. The presented method provides an efficient test of preparation of the excitonic states for emission of polarization entangled photon pairs in the biexciton-exciton cascade. Indeed, it represents a fast and effective way to select experimental conditions for time consuming polarization-dependent photon correlation measurements. Remarkably, when the FSS can be tuned through zero, we can extract from this relatively simple measurement a good estimate of the natural linewidth of the QD ground state exciton transition.

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