Optically induced energy and spin transfer in nonresonantly coupled pairs of self-assembled CdTe/ZnTe quantum dots

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(Received 7 November 2008; published 2 April 2009)

Asymmetrical horizontal interdot coupling was demonstrated to exist in a system of a single plane of self-assembled CdTe/ZnTe quantum dots. Photoluminescence excitation (PLE), second-order photon correlation, and optical orientation were used as main experimental tools. Each individual absorbing dot was identified by a sharp PLE resonance assigned to the neutral exciton transition, while the corresponding emission contained several excitonic transitions of different charge states in another single quantum dot different from the absorbing one. Energy and spin transfer dynamics were studied. A high efficiency of spin transfer was found from the optical orientation in a vertical magnetic field (70%) as well as without the magnetic field (40%), in spite of a significant anisotropic exchange splitting of the absorbing dot. Coherent mechanism of linear-to-circular polarization conversion was identified, with an efficiency (43%) close to the theoretical limit of 50%.

DOI: 10.1103/PhysRevB.79.153301 PACS number(s): 78.67.Hc, 78.55.−m

One of the reasons of growing interest in semiconductor quantum dots is related to the perspectives of their application in quantum information.[1] The potential of quantum dots (QDs) to generate single photons[2] and entangled photon pairs[3,4] has been demonstrated, opening the way to use the QDs to produce optical qubits. Polarization control of the emitted photons, a prerequisite for quantum information processing, has become an important challenge, in view of anisotropic QD properties.[5] On the other hand, the electronic states of the QDs, especially their spin degrees of freedom, can be directly used as qubits.[6] In this respect, the coherent manipulation of QD states[7] and coupling between qubits located in different QDs (Ref. 8) became an attractive field of investigations. Both vertically[9] and horizontally[10] coupled QD pairs have been studied. The research has been focused mainly on the resonant coupling of identical quantum dots, while the studies of coupling between QDs with strongly different ground-state energy have been less frequent.[11] In all cases, the quantum dot pairs have been prepared by a suitable growth procedure aiming at positioning the corresponding dots at a controlled distance from one another. In this work, we demonstrate that even in a single plane of self-assembled quantum dots there are randomly created pairs of coupled dots. We demonstrate the energy and spin transfer between such dots. Some preliminary results of this study can be found in Ref. 12.

The control of polarization of photons emitted by quantum dots may involve such processes as circular-to-linear and linear-to-circular polarization conversion. Two different mechanisms may lead to such conversion, both related to the QD anisotropy. The first one—population based—takes place in a magnetic field oriented along the growth axis. Its occurrence has been confirmed experimentally, e.g., on GaAs/AlAs superlattices[13] or individual InAs/GaAs QDs.[14] The second conversion mechanism is based on the coherent evolution of two linearly polarized QD eigenstates between the excitation and the emission events. It does not require a magnetic field and does not work with the light linearly polarized along either of the QD principal axes. The optimal light polarization for this mechanism is at 45° with respect to the principal axes and the maximum possible conversion efficiency is 50%. This coherent mechanism of circular-to-linear and linear-to-circular conversion has been observed experimentally on CdSe/ZnSe QD ensembles.[15] As the conversion has been so far observed on QD ensembles only, the resulting polarization degree is reduced due to averaging—the reported values do not exceed a few per cent. In this work, we employed an efficient spin-conserving transfer between coupled quantum dots in order to prepare and to probe specific well-defined quantum states of the quantum dot.

The studied self-assembled CdTe/ZnTe QDs were grown by the tellurium desorption method.[16] Surface QD density of $5\times10^9/\mathrm{cm}^2$ was obtained from atomic force microscopy (AFM) images of similar samples.[17] The AFM images also suggest that the QD spatial distribution is not completely uniform.

The samples were excited by a cw tunable dye laser via a microscope objective. Spectra of individual QDs were identified in the low-energy tail of the inhomogeneously broadened QD photoluminescence (PL) band. The photoluminescence excitation (PLE) was scanned over the maximum of the band. Weak above-barrier illumination was used in some of the experiments to change the occupation of different charge states. The most interesting features of PLE spectra of individual QDs are strong sharp [full width at half maximum (FWHM) down to 90 μeV] resonances as presented in Fig. 1(a). Typical PL spectrum of a single QD consists of a few distinct lines [Fig. 1(b)]. The pattern of the lines is unambiguously attributed to the recombination of exciton (X), biexciton (XX), and charged excitons (X^+ and X^-). It was observed before in similar QD samples. Several of investigated QDs exhibited an additional transition identified as

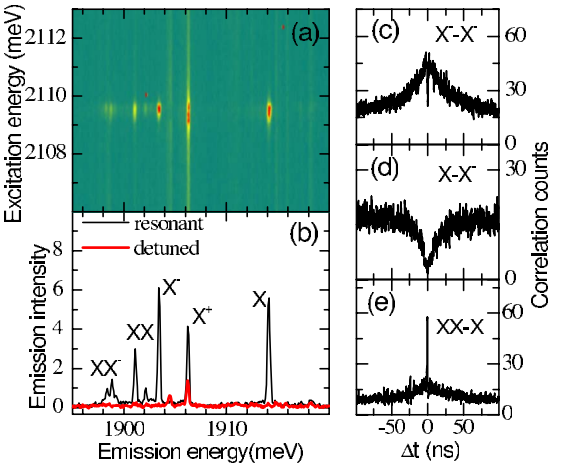

FIG. 1. (Color online) (a) PLE and (b) PL of a single QD measured under resonant excitation (black line) and after detuning by 1 meV (red - gray in print - line). Correlation histograms: (c) X^--X^- autocorrelation, (d) anticorrelation between different charge states (X-X^-), and (e) XX-X cascade.

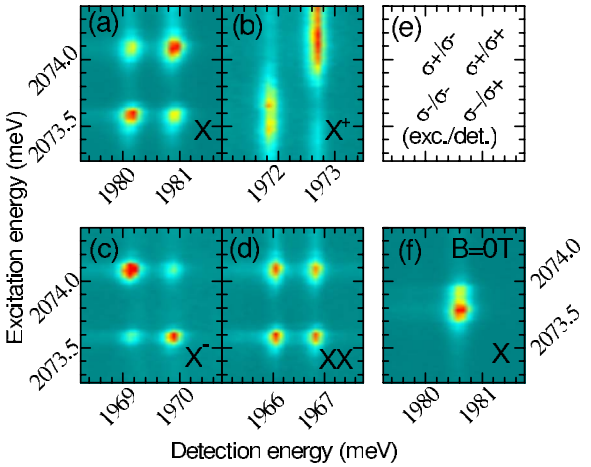

FIG. 2. (Color online) PLE spectra at $B=4$ T of transitions: a-X, b-X^+, c-X^-, and d-XX. Each maximum corresponds to different combination of excitation and detection circular polarizations according to scheme e. Map f presents PLE spectrum of X in $B=0$ T for comparison.

originating from charged biexciton (XX^-). The energies of the absorbing resonances are distributed in the range of maximum PL intensity of the QD emission band between 100 and 250 meV above the corresponding emission energies. No correlation between absorption and emission energies was detected, as reported in Ref. 12. All the lines originating from each emitting QD exhibit similar resonant behavior—corresponding PLE resonance energies are equal within accuracy of tens of μeV. These facts allow us to formulate a hypothesis that there are two different coupled quantum dots involved: an absorbing and an emitting one (it is very unlikely that the resonance condition is fulfilled at the same energy for three different charge states).

The assignment of the PL lines was supported by measurements of QD anisotropy. As expected, X and XX emission lines exhibited a fine-structure splitting (FSS), forming doublets with two orthogonally linearly polarized components. No FSS was detected for the charged exciton emission. The PLE resonances exhibited always the FSS doublet structure; therefore we assigned them to neutral exciton transitions in the absorbing dots. An unambiguous confirmation of the two coupled QDs model required the elimination of the possibility that the resonant absorption occurs in the same (emitting) quantum dot in one of its charge states. If this had been the case, the observation of the two other ones would require neutralization/charging of the QD during the lifetime of the excited state. To test this possibility, we performed photon correlation measurements. We used a Hanbury-Brown and Twiss setup described in detail in Ref. 18. The sample was excited resonantly. As presented in Figs. 1(c)–1(e), the measured histograms prove single-(or paired-in case of XX-X and XX^--X^- cascades) photon emission. Widths of sharp features visible in autocorrelations [Fig. 1(c)] and cross correlations between cascade transitions [Fig. 1(e)] are about 300 ps and are governed by the lifetime of the emitting states involved. In addition, we observed an enhancement or a suppression of the signal in the time range of tens of nanoseconds around zero delay. The enhancement means that the two correlated transitions occur in the same charge state, while for different charge states a suppression is observed. Moreover, the characteristic time scale of the charge state variation in X^+-X^- correlation is about twice longer than that in X-X^- or X-X^+. All measured correlation histograms can be described by rate-equation model calculations, using capture rates of electron-hole pairs and of single carriers as fitting parameters. The obtained values are 0.2 ns^{-1}, 0.02 ns^{-1}, and 0.03 ns^{-1} for pairs, electrons, and holes, respectively, which is consistent with the hypothesis of neighboring QD being an effective supply channel of resonantly created electron-hole pairs. The other mentioned scenario of the resonant absorption in the same (emitting) QD in one of its charge states would have led to qualitatively different results. Only one autocorrelation histogram (i.e., in resonant charge state) would have exhibited nanosecond-scale signal enhancement while the others would have had a wide dip. However, all lines related to different charge states studied in several QDs evidenced the enhancement, which clearly eliminates the mechanism of absorption in one of the charge states of a solitary QD.

In view of the presented arguments, we conclude at this point that the QDs are not isolated but can interact with one another. Excitons created by the resonant absorption in one QD relax to emitting states in a neighboring one. Interdot spin transfer was studied first in magnetic field up to 6 T in the Faraday configuration. In such a field Zeeman effect, observed both for absorbing (g-factor 1.78) and emitting (g-factor 2.81) QD dominates over FSS (130 and 140 μeV, respectively) and S_z projection of the exciton spin is a good quantum number. As a result, four combinations of circular polarizations of excitation and detection were encoded as peak positions in PLE maps measured with no polarization resolution [Fig. 2(e)]. We established that the resonant PL intensity depends on the polarization of both excitation and detection as presented in Figs. 2(a)–2(d). Efficient spin transfer is visible for all excitonic transitions, except XX, which is a singlet state. An interesting case of the negative optical

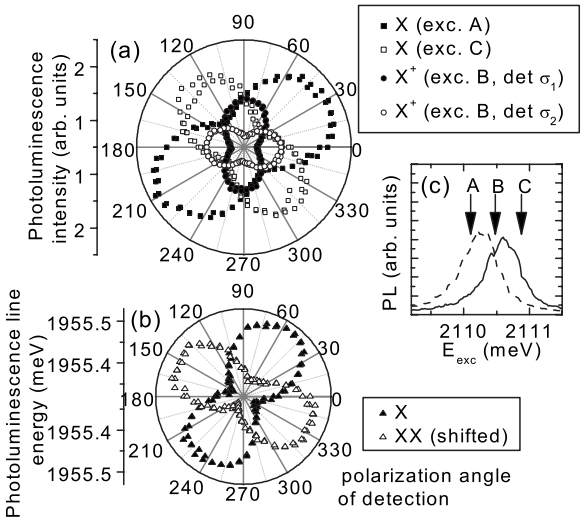

FIG. 3. Orientations of the anisotropy of the (a) absorbing and (b) emitting dot in a single QD pair. (a) Photoluminescence intensity at different fixed detection energies detected with no polarization resolution (squares) or circular polarization (circles) excited with rotating linear polarization. (b) Averaged PL energy versus the angle of rotating linear polarizer at detection. (c) PLE spectra of the pair (solid and dashed lines: linear polarization along QD eigenaxes). A, B, and C: excitation energies used for measurements presented in panel (a).

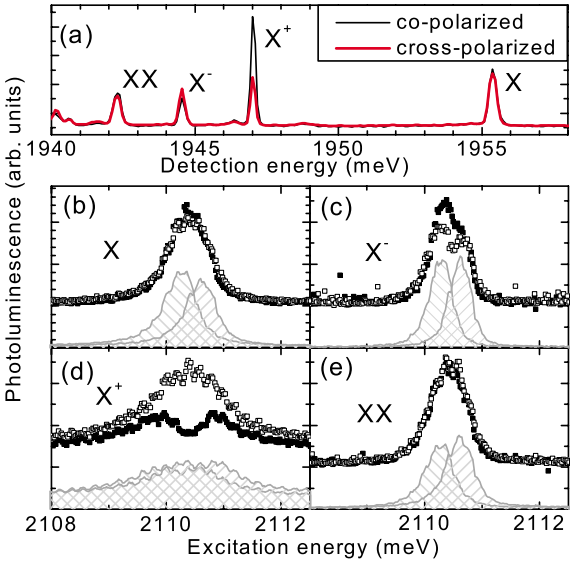

FIG. 4. (Color online) (a) Optical orientation of a QD with relatively broad resonance in circularly polarized excitation and detection. [(b)–(e)] PLE spectra of various transitions with the excitation polarized linearly at 45° and different circular polarizations of detection (points). Grey lines correspond to the excitation light polarized along principal axes of the absorbing QD (lines). The spectra were vertically shifted for clarity.

orientation occurs for X^- transition. We interpret it in terms of electron-hole spin *flip-flop* processes in the emitting QD, following Ref. 19, where similar effect has been observed before for negatively charged excitons in quantum dots pumped via the wetting layer. The negative optical orientation would not be possible without relatively fast spin relaxation of holes. In the experiments of Ref. 19, the relaxation occurred in the wetting layer; while in our case the hole spin may be depolarized during the energy relaxation process after the tunneling of the photocreated exciton to the emitting dot. The observed high degree of negative orientation (50%) proves that the *flip-flop* process can be very efficient.

Spin transfer in the absence of magnetic field is more complex because of the FSS of neutral exciton states. Figure 3 presents resulting anisotropy effects in the absorbing and emitting QDs. As expected, the splitting of the same value but opposite in sign is visible for X and XX emissions [Fig. 3(b)]. No splitting of charged exciton transitions was observed. The orientation of measured QD anisotropy varied from dot to dot, which is typical in II-VI compounds.[20]

The relative anisotropy orientation of the two dots in the pair can be determined from Figs. 3(a) and 3(b). Measured over many QD pairs, it exhibits a significant scatter but some correlation is also present, probably due to the local strain. We did not observe the transfer of linear polarization between dots. On the other hand, under certain conditions spin conservation leads to the circular polarization transfer. These facts favor tunneling mechanism of the excitation transfer, as opposed to Foerster electromagnetic coupling.

For the study of spin conservation in the absence of the field, we selected a QD pair in which the resonance width was larger than the fine splitting in the absorbing QD. In such a case, it is possible to excite a superposition of two linear eigenstates, e.g., $(|x\rangle + i|y\rangle)/\sqrt{2}$. The results are shown in Figs. 3(a) and 4. We found a significant circular polarization transfer to the two trion lines [Fig. 4(a)].

On the other hand, the two excited eigenstates of the absorbing QD have slightly different energies due to the in-plane anisotropy. Therefore, the relative phase φ varies with time. As a consequence, the system undergoes oscillations between circular and 45° linear polarization leading to polarization conversion.[15] We observed a considerable degree of circular polarization of emitted light after excitation with the linearly polarized light. Again, the effect was observed only for trions as the anisotropy of the emitting QD enforces linear emission for X and XX lines. Maximum efficiency was obtained if excitation was polarized at 45° to the principal axes of the absorbing QD as presented by circles in Fig. 3(a). The effect was more pronounced for X^+ than X^- (Fig. 4) due to a larger overlap of the two linearly polarized absorption lines. Difference in the sign of the conversion for the two trions is consistent with previously discussed experiments of optical orientation.

High efficiency of polarization conversion exceeding 40% allowed us to estimate transfer rate to be close to $\hbar/\delta_{\mathrm{FSS}}$, where δ_{FSS} is FSS value of the exciton in the absorbing dot. An efficiency of at least 40% corresponds to the transfer rate between 0.8 and 3.4 ps. This estimation remains in agreement with those based on the width of the resonance, which yields 0.7, 3.1, and 2.2 ps for positive, neutral, and negative charge states, respectively. The latter values may be underestimated because of neglecting inhomogeneous broadening. Similar value of the tunneling time was observed in experiments on coupled CdSe/ZnSe quantum well and quantum dot system separated by 10 nm barrier.[21] The distance between

coupled QDs may be larger if the dots are superimposed on two-dimensional (2D) platelets as reported in Ref. 22. We must stress here that the observed efficient polarization conversion is possible due to the comparable values of the FSS precession time and the lifetime of the exciton in the absorbing dot. This lifetime is governed by the interdot tunneling rate.

Concluding, we have shown the evidence for interdot coupling in a single plane of self-assembled CdTe/ZnTe quantum dots. The coupling was observed as the energy transfers between a neutral exciton state of the absorbing dot and various excitonic states of the emitting dot. This transfer occurs via tunneling of excitons between dots. High (at least 70%) interdot spin transfer efficiency has been demonstrated directly in vertical magnetic field, while at zero field it was observed directly and evidenced through high (above 40%) efficiency of linear-to-circular polarization conversion. Characteristic times of the system dynamics have been determined, ranging from a few picoseconds (FSS precession and interdot tunneling) to tens of nanoseconds (charge state variation).

This work was partially supported by the Polish Ministry of Science and Higher Education as research grants in years 2006-2010 and by the European Commission under Project No. MTKD-CT-2005-029671.

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