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Phonon-assisted biexciton generation in a single quantum dot

F. Findeis,* A. Zrenner, G. Böhm, and G. Abstreiter
Walter Schottky Institut, Technische Universität München, D-85748 Garching, Germany

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Phonon-assisted absorption of a single InGaAs/GaAs quantum dot is investigated by multichannel photoluminescence excitation spectroscopy as a function of magnetic field. GaAs LO phonon-assisted generation of exciton and biexciton states is observed. Magnetic field induced detuning of the resonance conditions results in a twofold diagonal splitting of the exciton and a symmetric fourfold splitting of the biexciton resonances. Those findings are explained in terms of sequential phonon-assisted biexciton generation followed by sequential biexciton decay.

Semiconductor quantum dots (QD's) are often described as artificial atoms due to their δ-function-like density of states. This analogy is nicely reflected in the fact that both free atoms and QD's exhibit optical line spectra with narrow line width. There are, however, also important differences between free atoms and QD's. Those originate mainly from the fact that semiconductor QD's typically consist of thousands of atoms arranged in a finite size lattice, which again is contained in a host crystal with different composition. In contrast to free atoms, QD's are therefore subjected to the specific elementary excitations of such composite solids. Interactions with phonons in particular have been shown to leave their fingerprints in optical spectra of single QD's.[1–3] In the field of inhomogeneously broadened ensembles of self-assembled QD's broad (multi-) LO phonon resonances have been observed by near resonance excited photoluminescence (PL) as well as PL excitation (PLE) experiments.[4–6] Those resonances may be caused partly by photon-assisted absorption and by absorption via excited states followed by resonant interlevel relaxation. In single self-assembled QD's, absorption by excited states and sharp phonon resonances can be observed independently.[2]

In this paper, we discuss phonon resonances of the exciton (1X) and biexciton (2X) line in a single QD as a function of laser energy and magnetic field. Such phonon resonances generally can occur due to phonon-assisted absorption or resonant Raman scattering. Via multichannel PL excitation spectroscopy on an isolated QD, we are able to obtain insight into the phonon-assisted processes and the generation of particular quantum states in the QD. We find a magnetic field induced splitting and double resonance of the 2X line, which is explained by sequential phonon-assisted biexciton generation, followed by sequential biexciton decay. Hence we find a clear dominance of phonon-assisted absorption versus resonant Raman scattering. In view of the new field of coherent optical spectroscopy of individual QD's,[7] knowledge about such processes is of increasing importance. For possible applications, such as quantum computing, coherent control over single QD's is required and systematic studies of all intrinsic sources of decoherence are expected to become key issues.

In our experiments, we use self-assembled $\mathrm{In}_{0.4}\mathrm{Ga}_{0.6}\mathrm{As}$ QD's embedded in GaAs and aluminum shadow masks with $0.2-1.0$ μm apertures. Earlier power dependent PL studies of such QD's have shown an s-p shell splitting of 22 meV and a biexciton binding energy of 2.6 meV (see Ref. 8 for details). For magneto-optical spectroscopy the sample was mounted in a confocal low-temperature microscope which can be operated in a 15 T magnet system at 700 mK (see Ref. 9 for details). The PL has been excited by a linear polarized cw Ti:Sapphire laser. In contrast to conventional PLE measurements with single channel detection, we used a cooled CCD camera for parallel detection of the complete PL data as a function of laser energy. In this paper we call this multichannel PLE (MPLE) spectroscopy.

In Fig. 1, a MPLE overview spectrum of a single QD is shown for a condition of high excitation power of $P_{\mathrm{exc}} = 3$ mW (corresponding to about 3×10^5 W cm^{-2}). Because of the small absorption volume given by a single QD, power densities in such a range are required in order to get detectable light output for excitation energies below the wetting layer (WL) absorption edge. In the displayed MPLE spectrum, PL data are contained along the horizontal direction for the whole range of excitation energies displayed on the vertical axis. PLE data for different detection energies can be inferred along the vertical direction. PL emission from the single exciton ground state appears at 1320 meV (1X), whereas sequential biexciton decay leads to the appearance of the biexciton line at 1317 meV (2X). Clear evidence for the assignment of the biexciton line comes from power dependence of the biexciton line intensity in PL experi-

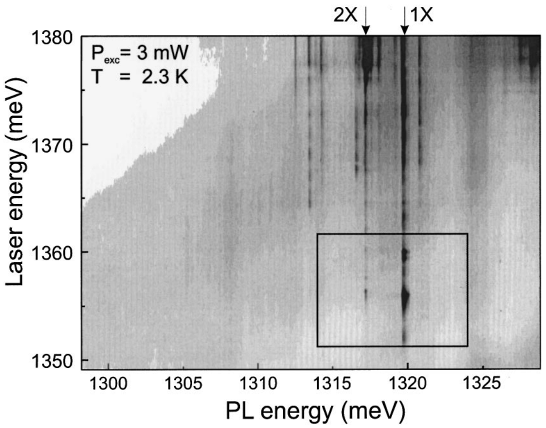

FIG. 1. MPLE overview spectrum of a single self-assembled $\mathrm{In}_{0.4}\mathrm{Ga}_{0.6}\mathrm{As}$ QD. It covers excitation energies between the region of the phonon resonances and the 2D wetting layer. The exciton and biexciton lines are marked as 1X and 2X.

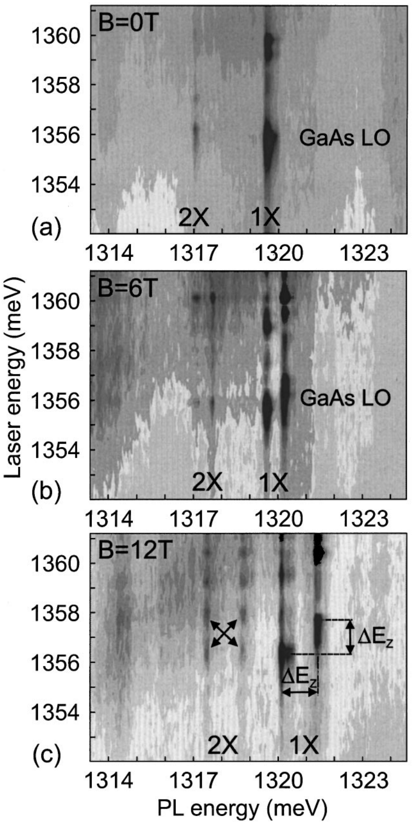

FIG. 2. Details of the MPLE spectra as a function of magnetic field for $B=0$ T (a), $B=6$ T (b), and $B=12$ T (c). The single exciton resonance (1X) shows diamagnetic shift and Zeeman splitting resulting in a twofold diagonal splitting. The weaker biexciton resonance (2X) shows symmetric fourfold splitting [marked in Fig. 2(c)].

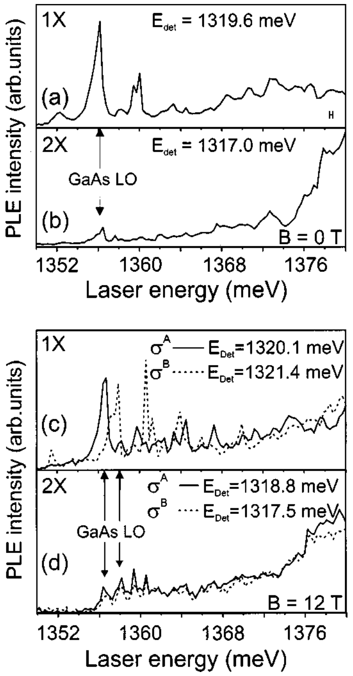

FIG. 3. PLE data at $B=0$ T detected on the 1X (a) and 2X line (b). PLE data at $B=12$ T for both Zeeman components (σ^A, σ^B) of the 1X (c) and 2X line (d). Phonon resonances on the 2X lines are observed for both 1X resonances independent of the 1X polarization.

ments.[8] The biexciton line shows quadratic increase with excitation power in contrast to a linear increase of the exciton line. The exciton line is in fact a doublet as can be seen in Fig. 2 which might be caused by a slightly asymmetric QD shape.[10] Additional, weaker emission lines are caused by the decay of higher neutral and charged *e-h* complexes and are not discussed further in this paper. Here we concentrate on the intensities of the exciton and biexciton line as a function of the laser energy and magnetic field shown in Fig. 2. In Figs. 3(a) and 3(b) we compare PLE data for the 1X and 2X line at zero magnetic field. The applied step size for laser energy in the PLE measurement is shown in Fig. 3(a). At an excitation energy of $E_{\rm exc}=1380$ meV, close to the onset of WL absorption, most of the PL intensity is concentrated in the 2X line. With decreasing laser energy, the absorption rate first goes down. This results in a rapid reduction of the 2X intensity, whereas the 1X intensity first remains approximately constant (1380→1370 meV) and then slowly decreases (1370→1360 meV). The most important features in the MPLE spectrum are, however, the sharp absorption resonances [marked in Fig. 1 and enlarged in Fig. 2(a)], which appear on further decrease of the laser energy (1360→1350 meV). Those resonances appear at laser energies of 1352, 1356, and 1360 meV, above the PL energy of the single exciton ground state $E_{\rm PL,1X}$ of the investigated QD at 1320 meV. With respect to the ground state the corresponding resonance energies ($E_{\rm exc}$–$E_{\rm PL,1X}$) appear at 32, 36, and 40 meV. The strongest peak is detected at a resonance energy of 36 meV, matching nicely the GaAs LO phonon energy for the accuracy of our measurement. Minor deviations could eventually be caused by strain fields in the vicinity of the QD. A weaker resonance at 32 meV is assigned to an InAs phonon mode related to the QD, also observed in ensemble PLE experiments.[5] The resonance around 40 meV falls into the region of the QD *d* shell and shows a doublet fine structure which would be expected for a slightly asymmetric QD shape. As shown in Fig. 2 and discussed in more detail later in this paper, those resonances are observed for both the 1X and 2X line.

Between the resonances, a weak continuum like absorption is observed as a function of excitation energy. Such a background absorption has been reported also by other groups for single QD's formed by interface fluctuations[11] and for self-assembled QD's.[1] The exact origin for this background is still under discussion. Absorption in WL tail states or QD continuum states has been suggested so far. As we see strong phonon resonances in our data, phonon-assisted absorption with a broad band phonon spectrum could also contribute here. This can be due to composition variations in and around the QD, phonon dispersions and multiphonon processes, also including the contributions of acoustic phonons.[12]

In Fig. 2 we display MPLE data of the exciton and biexciton GaAs LO phonon resonances peaks as a function of magnetic field for 0 T (a), 6 T (b), and 12 T (c). With increasing magnetic field, we observe a pure diamagnetic shift of 7.8 ± 0.5 μeV/T^2 for the exciton line and 7.5 ± 0.5 μeV/T^2 for the biexciton line, as expected for *s*-shell states of a QD with zero angular momentum. The observed Zeeman splitting is 1.08 ± 5 μeV/T for the exciton and 113 ± 5 μeV/T for

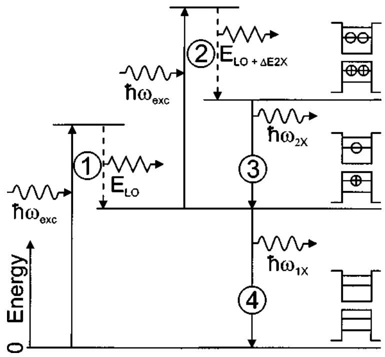

FIG. 4. Model for sequential LO phonon-assisted generation of a biexciton (1),(2) and the following sequential decay (3),(4) as described in the text.

the biexciton line. Phonon resonances generally lead to a constant offset between absorption and emission energy independent from magnetic field. For the GaAs LO phonon resonance, we get a variation in energy offset of less than 0.2 meV for five different magnetic fields. Since the GaAs LO energy is constant as a function of magnetic field, but the energies of both Zeeman components change, the resonance conditions for the laser excitation energies have to change as well. This magnetic field induced detuning of the resonance condition results in a twofold, diagonal splitting of the 1X GaAs LO phonon resonance with exactly the amount of the Zeeman energy ΔE_z [see Fig. 2(c)].

So far, we have only discussed the resonances of the 1X line, where one exciton at a time is generated and annihilated in the QD. However, there are additional features contained in the MPLE data shown Figs. 2(a)–2(c), which clearly go beyond this scenario. In particular, we find that for all laser energies which lead to strong exciton absorption, weaker resonances also appear at the position of the biexciton line. For weak exciton resonances (such as the one at 1352 meV for zero field) we cannot resolve corresponding biexciton resonances. As discussed above for nonresonant excitation, the power dependence in the MPLE data also justifies the assignment of the exciton and biexciton line. A reduction in laser power (not shown here) of a factor of 5 leads to the disappearance of the 2X resonances, whereas the 1X resonances still remain in MPLE data. The above described twofold, diagonal splitting of the exciton resonance at finite magnetic field leads, therefore, to a fourfold, symmetrical splitting of the biexciton resonance.

A model for sequential phonon assisted generation of a biexciton and the following sequential decay is shown in Fig. 4. At the resonant laser energy ($E = \hbar\omega_{\mathrm{exc}}$) an exciton in the QD and a phonon with energy E_{LO} are generated at a fixed rate (1). As long as the dot is occupied with one exciton, this transition is renormalized by the amount of the biexciton binding energy and hence blocked for further absorption at the given excitation energy. However, for unchanged excitation energy a second exciton (with opposite sign) can be generated by absorption under participation of a different phonon with energy $E_{\mathrm{LO}+\Delta E2\mathrm{X}}$ (2). This second absorption process, generating the biexciton in the QD, is made possible by the previous discussed weak background absorption. Each absorption process leading from the single to the two exciton state can contribute here and does not have to be phonon assisted in general. The biexciton decays sequentially and we expect first the emission of a photon at the position of the 2X line at $E = \hbar\omega_{2X}$ (3) followed by the single exciton emission at $E = \hbar\omega_{1\mathrm{X}}$ (4).

Similar correspondence in 1X and 2X PLE has been reported in mesoscopic quantum discs with much lower interlevel spacing of only a few meV.[13] Kamada *et al.* explained this by participation of excited exciton and biexciton states. For the present case this seems unlikely, because the lowest excited exciton state is 20 meV higher in energy. Also, resonant biexciton generation in a four particle process (two photons and two GaAs LO phonons) can be ruled out here. Such a process would be expected to be quite unlikely and appear half the biexciton binding energy below the GaAs LO phonon resonance (e.g., $E_{\mathrm{exc}} = 1.354.7$ meV for $B = 0$ T).

As a function of magnetic field, the biexciton resonance performs a fourfold, symmetric splitting [shown in Fig. 2(c)] in contrast to the twofold, diagonal splitting of the exciton resonances. This behavior nicely confirms the above discussed model for sequential phonon-assisted generation of a biexciton and the following decay. Each Zeeman component of a resonance can be a starting point for biexciton generation. In Figs. 3(c) and 3(d), we show by means of PLE spectra the coincidence of strong exciton resonances with the appearance of biexciton peaks for $B = 12$ T. A biexciton can be generated independently of which 1X Zeeman component shows strong absorption. Starting with a σ^A (σ^B) exciton, the next created exciton must be a σ^B (σ^A) due to the Pauli exclusion principle. Now two excitons with opposite spin orientations are in the QD whose probability of decay is the same. Depending on which decays first, a σ^A or σ^B photon is observed at the position of the corresponding biexciton line. This means that independently of the first absorbed spin orientation, both spin orientations are observed in the sequential decay of the biexciton with the same probability, and we obtain a fourfold, symmetric splitting for a biexciton resonance in the MPLE data.

Previously phonon-related features in QD PLE spectra have been discussed also in terms of resonant Raman scattering. Toda *et al.*[11] observed several additional resonance peaks in near field PLE experiments interpreted as resonant Raman scattering with localized phonons. Sequential biexciton generation in a QD, as observed in our present work, can only happen via an intermediate state with a real single exciton occupancy. In the resonant Raman scattering process, only a virtual exciton is involved. The QD remains, therefore, unoccupied and sequential biexciton generation is not possible. Hence, from our data, resonant Raman scattering can be ruled out as the dominant interaction process.

In conclusion, we have shown the importance of phonon-assisted absorption processes in a single QD. Those additional channels for exciton and biexciton generation are expected to be a major source of decoherence. A model for sequential phonon-assisted biexciton generation and the following sequential decay explains the different behavior of exciton and biexciton resonances as a function of magnetic field.

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*Author to whom correspondence should be addressed. FAX: +49-289-320-6620. Electronic address: findeis@wsi.tum.de

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