Sharp-Line Photoluminescence and Two-Photon Absorption of Zero-Dimensional Biexcitons in a GaAs/AlGaAs Structure

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Using a micron-sized photoluminescence (PL) probe enables us to study single islandlike interface defects of a thin GaAs/AlGaAs quantum well. The bound exciton ground state locally emits a distinct sharp line. With increasing excitation of this quantum dot level additional transition lines emerge at lower energy. They are attributed to localized biexciton states. The biexciton correlation energy is about 4 meV. A distinct two-photon resonant absorption peak of the biexciton ground state is observed in PL excitation spectroscopy. Its linewidth is only about 30 μeV. The spectra and their polarization properties are discussed on the basis of a discrete level scheme and the Pauli exclusion principle.

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For semiconductor quantum dot structures the linear and nonlinear optical effects are expected to be strongly enhanced compared to systems of higher dimensionality. This especially applies to a band gap modulated system like a GaAs/AlGaAs structure, in which electron-hole transitions are direct in momentum and real space. The discrete levels of electrons (e) and holes (h) localized in all three directions result in linear photoluminescence (PL) and absorption spectra with distinct strong peaks. The inherent e-h Coulomb interaction mainly results in a redshift and slight modification of optical transitions [1–3]. In practice, the strong transitions expected for such zero-dimensional (0D) structures are often weakened by structural imperfections mainly caused by lateral patterning. Inhomogeneous broadening is dominant, if the spectra are averaged over an ensemble of structures with fluctuations in composition and size. With increasing excitation density transition lines also broaden homogeneously [4–6]. Some nonlinear optical properties of 0D structures, like absorption bleaching and saturation of PL intensity, can be associated with the finite number of 0D states available. Localization of excitons is also expected to result in a strong enhancement of the biexcitonic correlation energy, with respect to quantum well (QW) and bulk samples [7–11]. For small GaAs/AlGaAs quantum dot structures the calculated values reach about 5 meV [7], compared to about 1 meV observed in QWs [10] and about 0.2 meV in bulk GaAs [10,11]. In this Letter we report on microscopic PL and PL excitation (PLE) spectroscopy from excitons and biexcitons localized in a single GaAs/AlGaAs quantum dot which is embedded within a GaAs/AlGaAs quantum well structure. It forms a 0D system with a few low-lying levels and a 2D quasicontinuum of states at higher energies.

The sample studied here is an as-grown undoped GaAs/$Al_{0.35}Ga_{0.65}As$ quantum well structure fabricated by molecular beam epitaxy (MBE) on (001)-GaAs with growth interruptions (GI) of 30 s without As-stabilization at each interface. The investigated GaAs QW of L_z = 34 Å thickness is embedded in between $Al_{0.35}Ga_{0.65}As$ barriers. The detailed layer sequence is described in Ref. [12]. For PL and PLE spectroscopy we use a DILOR triple-grating Raman spectrometer with an intensified Si-diode multichannel detection system. A cold-finger He-flow cryostat is mounted on an xyz-translation stage with a positioning accuracy of about 50 nm. The sample is cooled to about $T = 5$ K. It is excited by a tunable Ti:sapphire laser beam which is depolarized to the detected PL. The beam is focused by a microscope objective and the PL light passes a pinhole at an image plane which results in a PL probe size of about a micron.

Typical microscopic PL spectra from the GaAs QW with GI are shown in Fig. 1. The main PL line at energy $E = 1671$ meV corresponds to the GaAs QW with a layer thickness of $L_z = 34$ Å. The linewidth of only about 4 meV shows the high quality of the QW structure. Microscopic fluctuations in confining potential, however, are indicated by the peaks and shoulders the main line is consisting of and by the distinct strong PL peaks at up to about 15 meV lower energy. This fine structure and these peaks change in number and in energy with the lateral position of the PL probe at the QW, as illustrated in Fig. 1 and discussed in detail in Ref. [12]. They are attributed to excitons localized at fluctuations of the effective QW thickness. At very low excitation power the measured PL linewidth of 0.1 meV is close to the optimum spectrometer resolution of 0.07 meV. Increasing the power to $P_{exc} = 5$ μW, at each spatial position additional peaks appear lying up to about 5 meV below the exciton transition peaks. An inhomogeneous set of exciton peaks, as observed at Pos. 3, results in an increased number of nonlinear transition peaks at low energy. A remarkable feature of spatially resolved PL from the QW with GI, however, is that there are lateral regions like Pos. 1 and 2 where we collect PL at energies below $E = 1662$ meV which originates only from a single islandlike region of increased QW width. This has been verified in a detailed characterization of the band gap fluctuations in the surrounding QW region by scanning the PL probe, as described in Ref. [12]. In the following,

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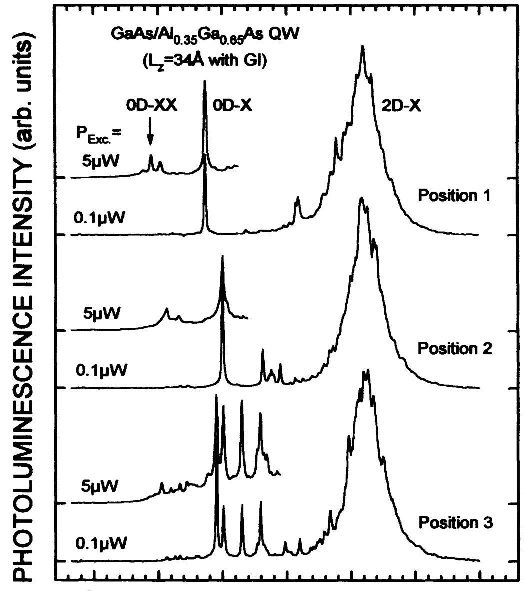

FIG. 1. Microscopic PL spectra from three different locations at a 34 Å thick GaAs/Al$_{0.35}$Ga$_{0.65}$As QW made by MBE with GI. For each position PL spectra excited at laser power $P_{\text{exc}} = 0.1\ \mu$W and $P_{\text{exc}} = 5\ \mu$W at energy $E_{\text{exc}} = 1705$ meV are shown. The spectral resolution is about 0.2 meV.

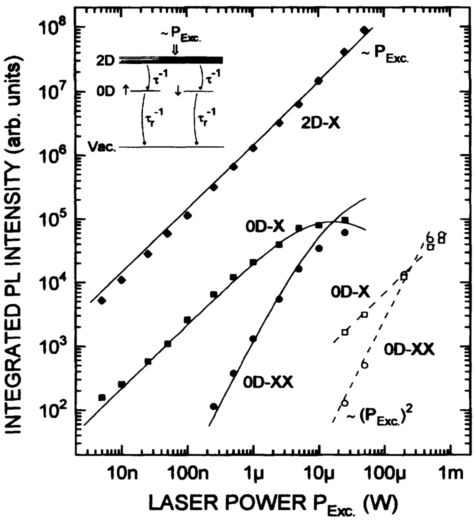

FIG. 2. Excitation power dependence of the integrated PL intensity for the broad quantum well line (2D-X), the sharp line of localized excitons (0D-X) at $E = 1658.6$ meV and the biexciton line (0D-XX) at $E = 1654.4$ meV originating from Pos. 1. The laser energy is 1705 meV and the probe size is $2r_e \approx 1.5\ \mu$m. Open symbols correspond to resonant two-photon absorption of the biexciton ground state. A relaxation scheme is shown in the inset.

we will describe the properties of the low-energy PL originating from Pos. 1. It is considered as a quantum dot structure with 0D levels at low energy and with a 2D-like onset of a quasicontinuous density of states at about 15 meV higher energies.

In Fig. 2 the integrated intensity of the main PL lines are shown in dependence on the power of the exciting focused laser beam. The broad QW emission at energy $E \approx 1671$ meV (2D-X) increases linearly with excitation power in the investigated range $P_{\text{exc}} = 5$ nW to 50 μW. The integrated intensity of the localized exciton transition (0D-X) at $E = 1658.6$ meV is about 2% of the total intensity. It increases also linearly at $P_{\text{exc}} \leq 1\ \mu$W. With stronger excitation the intensity saturates at a value which is about 3 orders of magnitude above our detection limit. At a laser power exceeding 250 nW the additional PL lines at lower energy are observed. The predominant transition at $E = 1654.4$ meV, assigned 0D-XX, gains intensity proportional to $(P_{\text{exc}})^2$ until a less pronounced saturation behavior is observed at similar power level, compared to the 0D-X transition. The linear increase of the 2D PL intensity indicates moderate optical excitation conditions of the QW even at the maximum power density of about 2 kW/cm^2 for $P_{\text{exc}} = 50\ \mu$W. A 2D exciton density of 1×10^{10} cm^{-2} is estimated for the spot center assuming a QW absorption efficiency of 1% and an exciton lifetime of 250 ps. The intensity ratio of the 0D-X and the QW lines in microscopic PL is strongly influenced by the size of the exciting laser spot and by the rates of radiative recombination, energy relaxation and migration of photogenerated carriers [3,12,13]. The low-energy peak 0D-XX is observable already at a laser power well below the onset of 0D-X saturation. The solid lines in Fig. 2 represent the solution of the rate equations for excitonic PL in a four-level system, which is sketched in the inset. The time-averaged occupation probabilities $N_\uparrow$ and $N_\downarrow$ have been calculated for the two degenerate radiative exciton ground states of the 0D system with electron spin up and down, respectively. Electron-hole pairs are generated in the 2D continuum forming 2D excitons with an effective occupation $N_2 = bP_{\text{exc}}$. These relax into the 0D ground states with a rate $1/\tau$ and finally decay radiatively with a rate $1/\tau_r$. Nonradiative recombination is neglected. Assuming rates independent from spin and from 0D excitation results in simple relations of the probability of 0D level occupation, single exciton ($I_{\text{0D-}X}$), and biexciton PL emission ($I_{\text{0D-}XX}$):

$$N = N_\uparrow = N_\downarrow = \frac{N_2\tau_r/\tau}{N_2\tau_r/\tau + 1}, \tag{1}$$

$$I_{\text{0D-}X} = c\,\frac{2}{\tau_r}\,N(1 - N)\,, \tag{2}$$

$$I_{\text{0D-}XX} = c\,\frac{2}{\tau_r}\,N^2\,. \tag{3}$$

Taking adequate constants b and c we get excellent agreement of the measured and calculated intensity behavior. At a laser power $P_{\text{exc}} = 25\ \mu$W both PL lines are about equal in strength $I_{\text{0D-}X} = I_{\text{0D-}XX}$. A dot occupation $N_\uparrow = N_\downarrow = \frac{1}{2}$ can be attributed to this case by comparing Eqs. (2) and (3), even though the rates $1/\tau$ and

$1/\tau_r$ are not known in detail. Using this for calibration, values $N_\uparrow = N_\downarrow = 4 \times 10^{-4}$ and 2×10^{-2} are estimated for our detection limit of 0D-*X* and 0D-*XX* PL, respectively. The well described nonlinear intensity increase of both the lines strongly supports the biexcitonic origin of the 0D-*XX* transition. It is attributed to recombination of one of the two excitons which are populating the 0D ground state at a time. The observed PL energy shift of 4.2 meV agrees well with the biexciton correlation energy expected for 0D systems with dimensions of a few exciton Bohr radii $a_B \approx 100$ Å [7–9]. The satellite peaks close to 0D-*XX* might be attributed to excited biexciton states or slightly modified eigenenergies caused by carriers transiently populating excited states.

PLE spectra of the 0D system at Pos. 1 are shown in Fig. 3. The intensity of the 0D-*XX* PL line at $E = 1654.4$ meV (DET 1) is detected in PLE 1. Absorption is negligible at low energy. At $E = 1664.8$ meV a distinct strong absorption peak is observed. This peak as well as the closely spaced peaks observed at $E > 1668$ meV coincide in energy with the peaks of spectrum PLE 2 recorded for the 0D-*X* PL line (DET 2). All the peaks are present in both spectra. The height and width of peaks differ because of the increased laser power in PLE 1 and the nonlinear properties. Observation of the same PLE peak structure detecting the two lines is further proof that both lines are emitted from the very same QW region of decreased band gap, representing a well defined single dot structure. Using a single particle picture, the 0D-*X* line corresponds to the 0D ground state recombination of electrons and heavy holes $e0 \rightarrow hh0$ and the PLE peak at 1664.8 meV may be attributed to the excited state transition $hh1 \rightarrow e1$. Only slight changes in the energy separation of transitions are expected when Coulomb interaction is taken into account [2,3]. The observed peak separation $\Delta E = 6.2$ meV can be explained, for example, by the 0D level scheme of a circular Gaussian-shaped band edge modulation in the QW plane (x-y) as drawn in the inset of Fig. 3. It is caused by a 4 Å deep protrusion of the GaAs QW into the barrier layer and results in an effective dot size of about 400 Å [12,14]. Short range fluctuations of Al content may lift the orbital degeneracy and the optical selection rules of localized levels. This may explain the multitude of PLE peaks merging to a continuum at higher energy [12]. A unique feature of spectrum PLE 1 is the absence of an absorption peak at the energy of the 0D-*X* line. Because of biexcitonic binding, energy conservation prevents generation of two excitons at the energy of the single exciton transition. Generation of two spatially separated uncorrelated excitons and subsequent biexcitonic binding is possible only in systems with at least one direction of free motion, like in bulk, quantum well, and quantum wire structures [15].

The PL and PLE properties in the energy range of the 0D-*X* and 0D-*XX* line have also been studied at increased laser power. Figure 4(a) shows PL of the dot resonantly excited at the $hh1 \rightarrow e1$ transition at $E = 1664.8$ meV.

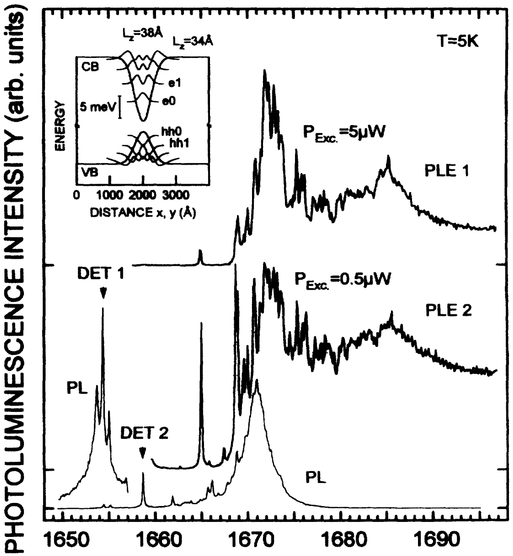

FIG. 3. PL and PLE spectra of the 0D exciton (PLE 2) and the main 0D biexciton recombination line (PLE 1). The excitation power was $P_{\text{exc}} = 0.5$ μW and $P_{\text{exc}} = 5$ μW, respectively. On the left hand side biexciton PL excited at energy $E = 1672$ meV is shown. Calculated 0D single particle levels of electrons and heavy holes are sketched in the inset for a Gaussian-shaped lateral modulation of effective QW band gap caused by a local increase of QW width of about 4 Å.

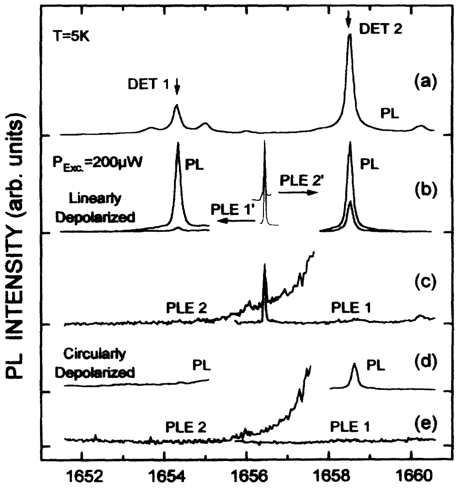

FIG. 4. In (a) PL of the biexciton (Det 1) and the exciton (DET 2) is resonantly excited at the 0D exciton level at $E_{\text{exc}} = 1664.8$ meV with $P_{\text{exc}} = 10$ μW. (b) shows PL spectra excited by a laser beam of power $P_{\text{exc}} = 200$ μW, which is linearly depolarized to the detected PL and is in a resonance at $E = 1656.5$ meV, or at a detuning of 0.2 meV. The corresponding excitation spectra PLE 1′ and PLE 2′ have been recorded with a spectral step size of 10 μeV. Equivalent spectra PLE 1 and PLE 2 are shown in (c) for a larger energy range. The spectra in (d) and (e) are equivalent to that in (b) and (c), but the laser beam and PL detection are circularly depolarized.

The PL spectra of Fig. 4(b) have been excited by a linearly depolarized laser spot of power $P_{\mathrm{exc}} = 200\ \mu\mathrm{W}$ at energy $E = 1656.5$ meV in between the two lines, or at a detuning of 0.2 meV from the exact resonance. The corresponding PLE spectra assigned PLE 1′ and PLE 2′ show the resonant behavior of the 0D-*XX* and 0D-*X* line intensity. The full width at half maximum of the resonance is only about 30 μeV, which is already limited by the linewidth of the exciting Ti:sapphire laser beam. Excitation spectra recorded in an increased energy range PLE 1 and PLE 2 are given in Fig. 4(c). Detecting the biexciton line (DET 1) the sharp peak at 1656.5 meV is the only absorption feature in this energy range. The single exciton line (DET 2) is resonantly excited at the same energy. A broad absorption tail reveals a nearly monotonic raise of intensity with decreasing energy separation of the detected 0D-*X* line and the exciting laser line at lower energy. Figure 4(d) and 4(e) represent PL and PLE spectra which have been observed at the same experimental conditions as in 4(b) and 4(c), but with circularly depolarized excitation (σ^+) and detection (σ^-). With circularly polarized as well as depolarized excitation no resonance peak at 1656.5 meV is observed. Adjustment of the laser focus and dot position has been checked carefully. It is also verified by observing a 0D-*X* absorption tail which is very similar to the linearly depolarized case.

The sharp PLE peaks in Fig. 4(b) and 4(c) are of about the same height for the 0D-*X* and 0D-*XX* line. They are attributed to the two-photon absorption of the biexciton ground state [15,16]. Two photons of the same energy $E = 1656.5$ meV generate two correlated excitons of energy $E = (1654.4 + 1658.6)$ meV via a virtual intermediate state. Resonant excitation of the discrete biexciton ground state in a single dot without population of excited states enables observation of linewidths which are considerably smaller than known from common PL, PLE, and two-photon spectroscopy. The intensity of the exciton and biexciton line are given as open symbols in Fig. 2 for different power levels of resonant two-photon excitation. At $P_{\mathrm{exc}} \approx 1$ mW they reach about the maximum value observed for linear excitation. The biexciton line intensity increases quadratically in P_{exc}, as excepted for a two-photon absorption process [8,16]. The exciton line, however, shows a more linear behavior. This is attributed to the additional nonresonant excitation of excitons predominant at low P_{exc}. This absorption process also causes the increasing intensity in PLE 2 with the laser energy approaching the 0D-*X* transition energy. It might be related to phonon-assisted absorption of the exciton ground state. Phonon broadening of transitions is expected to dominate the linewidth observed from quantum dots, if inhomogeneous as well as homogeneous contributions are negligibly small [1]. The two-photon absorption peak is not observed at circular laser light polarization. Photons of σ^+ (σ^-) polarization excite excitons with a well defined electron and heavy hole spin orientation $e0$, $m_j = {}^{-}_{(+)}\frac{1}{2}$ and $hh0$, $= {}^{+}_{(-)}\frac{3}{2}$, respectively [15,17]. Although energy conservation is fulfilled, two *e-h* pairs of the same spin orientation and the same orbital quantum numbers cannot be generated by the two photons of the same circular polarization. This directly reflects the Pauli exclusion principle applied to the spin-degenerate 0D ground state. In other words, the 0D biexciton ground state is nondegenerate and two-photon absorption requires a σ^+- and a σ^--polarized photon which are offered by linearly polarized light.

In conclusion, we have presented results about the linear and nonlinear optical response of a GaAs/AlGaAs single quantum dot structure. Microscopic photoluminescence spectroscopy of weakly excited single dot structures offers a novel approach to studying well defined exciton states and their interaction with other carriers, photons, and phonons in semiconductor heterostructures.

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