Magnetooptical studies of a single quantum dot: Excited states and spin flip of excitons

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(Received 7 November 1996)

We report on microphotoluminescence experiments in magnetic fields on single quantum dots formed by width fluctuations in a narrow GaAs/$Al_xGa_{1-x}As$ quantum well. The ground state as well as two excited states are discussed. We find a different diamagnetic shift and Zeeman spin splitting for these states. Resonantly exciting zero-dimensional excitons, we observe a strong magnetic-field dependence of the spin flip rates. [S0163-1829(97)51308-7]

An attractive model system for optical experiments on zero-dimensional (0D) semiconductor structures is excitons localized by width fluctuations in quantum wells.[1–4] Especially for narrow quantum wells, the energy of the photoluminescence line strongly changes when the thickness is varied by 1–2 monolayers. Therefore in a real quantum well (QW), deviations from a perfectly smooth interface always lead to potential minima where excitons can be trapped before they recombine. These confined excitons are fully quantized, which is reflected by small spectral linewidths below 0.1 meV. With spatially resolved spectroscopy it is now possible to resolve such narrow lines because inhomogeneous broadening is avoided when a single quantum dot structure is investigated. In a 3.5 nm wide GaAs QW, Brunner *et al.*[1] found such localized exciton lines and even observed excited states of these quantum dots. Recently Gammon *et al.*[4] observed a fine-structure splitting of the order of 50 μeV in the optical spectra of such an artificial atom, which they attributed to asymmetries in the shape of the lateral confinement. The lateral extension of the corresponding 0D states has not yet been resolved, not even in the scanning near-field microscopy experiment (spatial resolution of 100 nm) carried out by Hess *et al.*[3] However, from the energy separation between the 0D states of a single dot the lateral size can be estimated. For the ground state of our structure we thus estimate a size of about 40 nm.

The application of a magnetic field to such a single artificial atom is very interesting. For real atoms the magnetic fields accessible in a laboratory are only a weak perturbation compared to the Coulomb binding energy. In our 0D structures, however, it is possible to reach a regime where the magnetic confinement is as important as the size quantization. For example, this could give rise to qualitatively different magnetic-field dependences of the ground state and the excited states. In this paper we present photoluminescence (PL) and photoluminescence excitation (PLE) spectroscopy measurements on a single quantum dot in magnetic fields. The sample under investigation is a 3.5 nm wide GaAs/$Al_{0.35}Ga_{0.65}As$ single QW structure fabricated by MBE with growth interruptions of 30 s at both interfaces. The PL is excited by a Ti:sapphire laser beam, which is focused by a microscope objective to a spot size of 1.5 μm at the sample surface. The sample is mounted in a continuous flow cryostat in the center of a superconducting magnet, which provides magnetic fields up to 5 T at a temperature of 4 K. The PL signal is collected with the same microscope objective and dispersed by a triple grating Raman spectrometer. A pinhole which is placed at an image plane defines the detection area of 3 μm in diameter. With quarter wave plates we can excite and detect with defined circular polarization. A high precision *xyz* translation stage is used to position the sample and to address single structures.

Throughout this paper we describe the properties of one single quantum dot. Similar results, however not presented in this paper, were found on a different position, which was also examined in detail. The lowest curve in Fig. 1 shows a PL spectrum without magnetic field. The excitation power is as low as 1.5 μW, which means that there is less than one exciton in the dot on the time average. The QW luminescence is observed at 1671 meV while the PL from a single localized exciton is seen at 1657.5 meV. The full width at half maximum of the latter line is about 0.1 meV, the lateral extension of this 0D state is below the spatial resolution. We

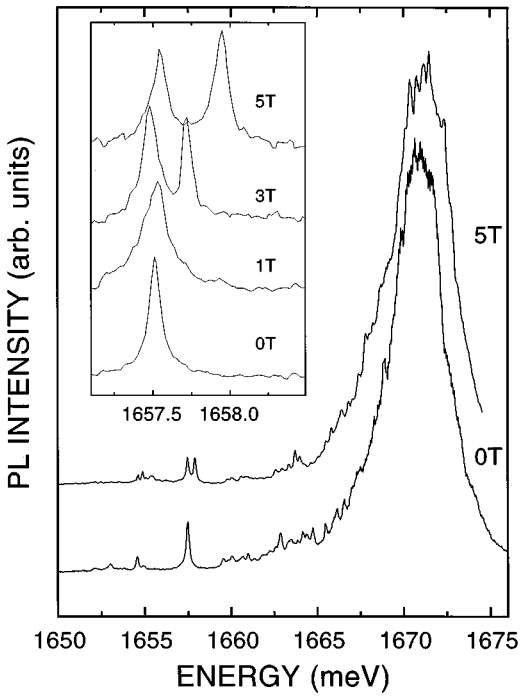

FIG. 1. Unpolarized photoluminescence spectra at various magnetic fields at 4 K. The excitation is done at 1700 meV with a power of 1.5 μW. The PL of the single dot is observed at 1657.5 meV.

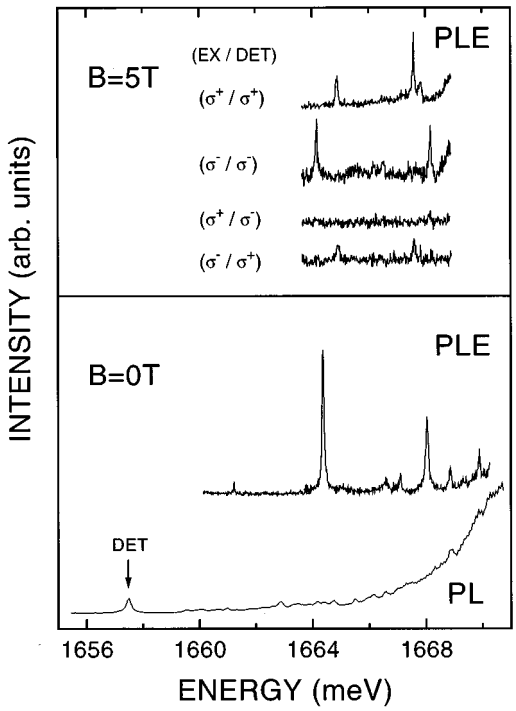

FIG. 2. Photoluminescence excitation spectra at $B=0$ T and 5 T. The excitation power is 4 μW. In the upper part ($B=5$ T) spectra are shown which were recorded in different circularly polarized geometries. The first label gives the polarization of the excitation, the second label corresponds to the polarization of the detection.

do not observe a zero field splitting as reported by Gammon *et al.*[4] within our spectral resolution of 70 μeV. Typically there are several dots in the detection area. Scanning the sample it is, however, possible to find positions where only one line emerges. We perform investigations on such positions because physical effects can then be attributed to a single quantum dot.

When we apply a magnetic field parallel to the growth direction of the QW, the PL line exhibits a diamagnetic shift and a Zeeman spin splitting. The inset shows spectra recorded with the maximum spectral resolution of about 70 μeV. At $B=5$ T the two spin split lines are separated by 0.4 meV; the diamagnetic shift is 0.25 meV. We have also investigated different positions and find that there is no systematic dependence of the splitting on the localization energy or on the diamagnetic shift which is a measure of the lateral confinement. The diamagnetic shift of the ground state is quadratic in B with a constant of 10 μeV/T^2. The linear spin splitting of 82 μeV/T corresponds to an exciton g factor of 1.42. With a quarter wave plate in front of the spectrometer and linearly polarized excitation we find that the lower line is σ^- polarized, the upper line σ^+. Although kT is only of the order of 0.3 meV, both lines exhibit almost the same intensity. This already shows that in high magnetic field these states are not in thermal equilibrium and that spin flip transitions are rather improbable on the time scale of radiative recombination.

In the lower part of Fig. 2 a PLE spectrum is shown with the detection set to the 0D PL line as indicated. We observe two strong peaks in the excitation spectrum below the QW continuum with linewidths of only 50 μeV (at 1664.4 meV) and 120 μeV (at 1668.1 meV), respectively. Gammon *et al.*[4] found values of 30–50 μeV. We can resolve these linewidths because in a PLE experiment the spectral resolution is given by the linewidth of the laser beam. The higher energy lines are not seen in the PL spectra, which means that the exciton relaxation to the ground state is efficient on the scale of the radiative lifetime and that these peaks do not stem from neighbored potential minima, which might contribute to the luminescence of the ground state by migration of carriers after resonant absorption. Therefore these lines are attributed to excited states of the quantum dot. This is also supported by the magnetic-field dependence of these lines, which strongly differs from the ground state as shown below. For strong lateral confinement it is expected that the relaxation becomes inefficient compared to the radiative recombination. In the present dot, however, the level separation is still small enough to allow for efficient relaxation.[5] We have also investigated other positions and two further QW's of width 3 nm and 5 nm grown in another MBE chamber. In all samples we find sharp PL lines below the QW peak and all of them have a spatial extension below the resolution limit. However, we observe a strong variation in the separation between the 0D states and there are even positions without any excited state in the PLE spectrum below the QW continuum.

In a magnetic field the two excited levels also split up according to spin. In the upper part of Fig. 2, PLE spectra for different polarization geometries are shown. The curve denoted by (σ^+/σ^+) was obtained with σ^+ polarization for both excitation and detection, while for the second curve we used σ^- polarization. Each of the two spectra shows two sharp peaks which correspond to the spin split states of the two excited levels at $B=0$ T. The lowest two curves (σ^+/σ^-) and (σ^-/σ^+) were recorded in circularly depolarized geometry. The peak heights in these spectra are much smaller which means that at $B=5$ T the spin is conserved to a large extent during the relaxation process within the single quantum dot.

The magnitude of the polarization $|I^+-I^-|/(I^++I^-)$, where $I^{+,-}$ are the intensities for $\sigma^{+,-}$ detection, strongly depends on the magnetic field. This is shown in Fig. 3. The dot is excited resonantly into the first or second excited level with circularly polarized light and the resulting polarization of the ground state is measured by turning the quarter wave plate. This method is more accurate than the comparison of the absolute peak intensities in the PLE spectra because even a very small drift during the measurement may change the intensities by about a factor of 2. Due to the noncomplete polarization of the experimental setup a ratio of 1 for the polarization cannot be reached. We have separately measured the degree of circular polarization $\eta=0.72$ of the setup and extracted the polarization P of the sample emission by dividing the measured intensity ratio by η. At zero field, where each pair of spin states is degenerate, almost no polarization is observed. When the field is increased, the spin relaxation is reduced because the spin states split up and an energy exchange with the lattice is required. This qualitatively explains the increase of the polarization with magnetic field.

For a more quantitative description we consider the rate equation introduced by Maialle *et al.* [Eq. (2.3) in Ref. 6] in

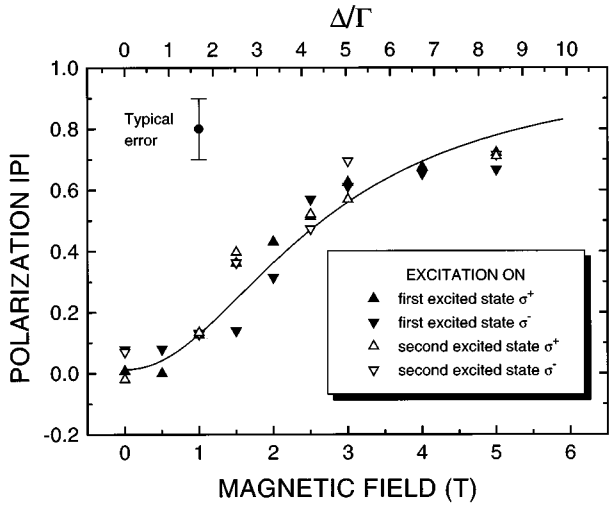

FIG. 3. Dependence of the polarization on the magnetic field. Up- and down-triangles are measured points, the solid line is a fit curve obtained by the described two stage relaxation model. The upper x axis shows Δ/Γ for the ground state.

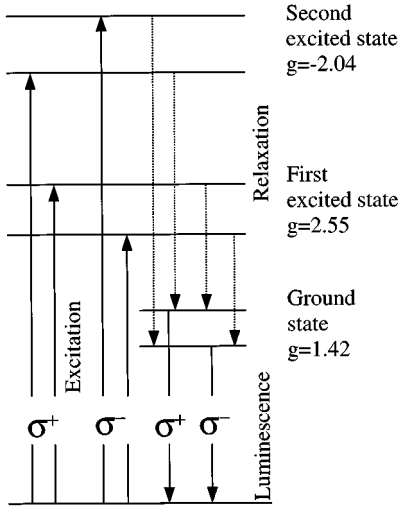

FIG. 4. Summary of the observed transitions within the investigated quantum dot.

the steady state case. For the polarization P_i of a given state i it follows

$$P_i^{-1} = 1 + 2\,\nu_{\mathrm{SF}}\tau_L\,, \tag{1}$$

where ν_{SF} is an effective spin flip rate between the two spin split radiative levels (including direct transitions and transitions via dark exciton states). τ_L is the lifetime of state i with respect to relaxation (only for the excited states) and radiative recombination. To our knowledge no theory for the exciton spin flip in 0D systems is available. Spin flip mechanisms based on motional narrowing[6,7] rely on translational invariance, which is not present in the quantum dot case. This drawback in mind, we nevertheless tried to fit our data using the magnetic field dependence of the 2D exciton spin flip rate according to Eq. (A12) of Ref. 6:

$$\nu_{\mathrm{SF}}(B) = \nu_{\mathrm{SF}}(0)\left[1 + \left(\frac{\Delta}{\Gamma}\right)^2\right]^{-1}. \tag{2}$$

Δ is the magnetic field splitting and Γ is the linewidth of state i. Equation (1) becomes

$$P_i(B) = \frac{1 + \left(\frac{\Delta}{\Gamma}\right)^2}{P_i^{-1}(0) + \left(\frac{\Delta}{\Gamma}\right)^2}. \tag{3}$$

Now, we assume that the total polarization P is given by the product $P = P_i P_j$, where i and j are the optically excited state and the emitting ground state. The same ν_{SF} is used for both states; the lifetime τ_L of the ground state is assumed to be ten times longer than that of the excited state. This is consistent with the absence of higher PL lines. The last two constrictions relate $P_i(0)$ and $P_j(0)$ via Eq. (1). Since we cannot resolve the linewidth of the ground state experimentally, we assume a width of 50 μeV as measured for the first excited state. In Fig. 3 a fit curve is presented which was obtained in the described model. A reasonable fit is obtained for $P_j(0) = 0.05$, which fixes the product of ν_{SF} and τ_L via Eq. (1). With measured PL decay times for excitons in a quantum dot of several hundred picoseconds[8] this corresponds to spin flip times for $B = 0$ T in the range of some 10 ps. The first excited state contributes to the spin relaxation at low field but becomes less important at higher magnetic field due to its larger g factor. That almost the same magnetic field dependence is found when the second excited state is pumped resonantly indicates that the spin relaxation mostly takes place in the ground state. Within the experimental precision, there is no significant difference for σ^+ or σ^- excitation in agreement with the assumption of symmetrical spin flip rates.

The upper two curves of Fig. 2 show another interesting feature, namely that the splitting of the second excited level is of opposite sign to the splittings of the first excited level and the ground state. The size of the splitting is larger for the

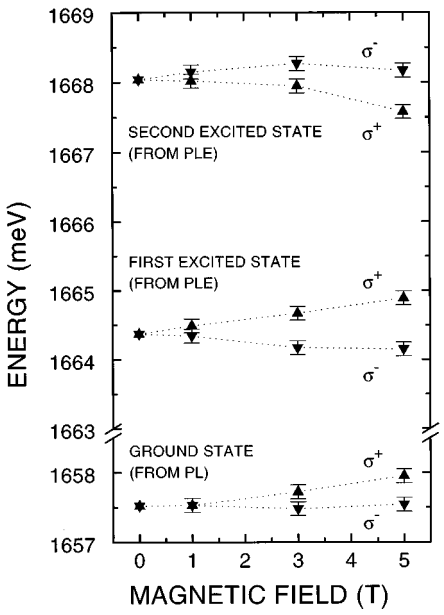

FIG. 5. Peak positions from PL and PLE data as a function of the magnetic field.

excited states than for the ground state. At a magnetic field of 5 T the excited state splittings are 0.74 meV and -0.59 meV, which correspond to exciton g factors of 2.55 and -2.04, respectively. A similar behavior has been observed for magnetoexcitons in a 10 nm wide QW.[9] Theoretically the reversal of the g factor was attributed to the heavy-hole–light-hole mixing in the valence band.[10] In our sample the valence-band mixing is weaker due to the smaller well width. On the other hand, the contribution of the hole to the exciton g factor strongly increases with decreasing well width.[11] For a quantitative description of this result calculations of quantum dot excitons including valence-band mixing are required because in the present case the exciton binding energy and the lateral confinement energy are of comparable size. Figure 4 summarizes the observed transitions for excitation, relaxation, and luminescence within the quantum dot at $B \neq 0$ T.

In Fig. 5 the experimental peak positions of the ground state and the two excited levels are plotted versus magnetic field. We observe a small change in the splitting of the ground state (about 60 μeV at 5 T) when the excitation is changed from σ^+ to σ^- which, however, is close to the spectral resolution of our setup. The diamagnetic shifts of the states strongly differ. While the ground state is blueshifted, the first excited level almost remains unchanged and the second excited state is redshifted. Such a behavior has been obtained theoretically by Halonen *et al.*[12] and Bockelmann[13] for magnetoexcitons in quantum dots with parabolic lateral confinement. In both cases the first excited level is mainly a center-of-mass excitation and the second excited level an excitation of the relative coordinate of electron and hole. This again agrees well with the calculations of Bauer *et al.*,[10] because an excitation of the relative coordinate corresponds to an $h(2s)$ state in their notation and just for this state they predicted a reversal of the exciton g factor.

In conclusion, we have performed photoluminescence and photoluminescence excitation spectroscopy on an individual quantum dot in a magnetic field. We observed a ground state and two excited states which in a magnetic field split into doublets. The ground state shows a smaller spin splitting than the excited levels; for the second excited state the sign of the splitting is reversed. The three states show qualitatively different diamagnetic shifts. Increasing the magnetic field from 0 to 5 T, the degree of spin polarization induced by optical pumping changes from below 0.1 to values above 0.7. It is possible to fit the measured magnetic-field dependence of the polarization of the 0D system with a 2D theory. For related measurements on quantum wells this means that data consistent with the 2D theory of spin relaxation do not necessarily imply extended exciton states.

We thank G. Abstreiter, G. Bastard, R. Ferreira, and U. Rössler for helpful discussions and G. Böhm, G. Weimann, M. Bichler, and W. Wegscheider for providing the samples. This work has been supported financially by the Deutsche Forschungsgemeinschaft (SFB 348).

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