Magneto-optical properties of ring-shaped self-assembled InGaAs quantum dots


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Abstract

We report on measurements of the magneto-optical properties of excitons confined in ring-shaped self-assembled semiconductor quantum dots. The rings are embedded in a field-effect structure that allows the number of confined electrons to be set electrostatically. In addition, electron-hole pairs are generated optically. The resulting photoluminescence spectra of neutral, singly and doubly charged excitons were measured at 4.2 K as a function of the applied magnetic field (0–9 T). The emission energy shows a diamagnetic shift as well as a Zeeman splitting. We measured the emission of different charge states of the exciton in many individual dots. In a few of the measured rings, a new behavior was observed, namely a clear departure from the low field diamagnetic dispersion for fields larger than 6 T.

PACS: 78.67.Bf; 73.21.2b, 73.40.Rw; 78.66.Fd

Keywords: Spectroscopy; Self-assembled systems; Quantum dots
diamagnetic energy shift of the photo-luminescence (PL) of excitons confined in ring-shaped quantum dots.

The samples were grown by solid-source molecular beam epitaxy. InAs self-assembled dots are grown on a thick GaAs epitaxial film on a GaAs substrate. A thin layer (1 nm) of GaAs is then grown on top of the self-assembled dot layer and annealed at a temperature of 520°C for 1 min [3,4]. The shape of the nanostructures changes during this annealing step from a lens shape (about 20 nm in diameter and 6 nm high) to a 2 nm high dot with a crater of 20 nm diameter in its center. At this stage, the structures resemble volcanoes of roughly 60–140 nm in outer diameter [3,4]. Further capping with a 150 nm barrier material (GaAs and then AlAs/GaAs short period superlattice) is necessary so that the nanostructures become optically active. The dot layer is grown on a 25 nm undoped GaAs layer that separates it from a degenerately Si-doped GaAs back contact. With a semi-transparent NiCr front gate an electric field can be applied which makes it possible to control electrostatically the number of electrons present per ring from none to a few. We have taken low temperature (4.2 K) photoluminescence (PL) spectra on tens of individual rings as a function of the front gate voltage in the range between −0.8 and 0 V. Each time an individual electron tunnels into the ring from the back contact, an abrupt red-shift of the emission energy is observed as a result of the charging of the nanostructure [5–8]. Due to Coulomb blocking, the number of electrons present in the ring can be kept stable and the emission shows characteristic charging plateaus as a function of \( V_g \) [5]. Such gate sweep PL measurements are systematically recorded before measuring the magnetic field dependence of the emission in order to define unambiguously the charge state of the investigated ring. For \( V_g \) smaller than −0.8 V electrons tunnel out of the quantum dot at a rate fast enough such that no excitonic emission can be detected. For \( V_g \) larger than +0.1 V, the thin InAs wetting layer surrounding the rings fills with a two-dimensional electron system.

The PL was obtained by optically pumping the InAs wetting layer with \( \lambda = 820 \text{ nm} \) just below the band gap of GaAs. The collected PL was spectrally dispersed with a 300 mm focal length monochromator and detected with a liquid nitrogen cooled Si CCD camera, a system with spectral resolution of 0.3 meV. In this work the polarization of the PL was not analyzed. Both the single ring PL and the ensemble measurements were performed with low enough pumping intensity such that the emission of the higher excited excitonic levels or bi-excitonic emission was not possible. The emission of the wetting layer is detected at 1.425 eV (870 nm) and the ring emission was found in a spectral range spanning 1.26–1.36 eV. The PL of a few tens of individual rings was measured using a confocal microscope setup together with a sample preparation outlined in Ref. [5]. Fig. 1 shows the evolution of the PL spectrum of a single ring as a function of the applied magnetic field. The PL emission splits into a lower and an upper branch. Such a characteristic Zeeman splitting of the excitonic emission was observed on all the measured dots independent of their charge. Within our spectral resolution, the Zeeman splitting was found to be linear in \( B \) with an average slope of 120 µeV/T varying from dot to dot with a standard deviation of 30 µeV/T. Fig. 1 also shows that the PL peak positions shift with applied field \( B \) to higher energies. In order to analyze such a diamagnetic shift, the peak position of the lower (upper) branch of the PL peak is plotted against negative (positive) field values. The choice of sign convention for \( B \) is arbitrary. For the majority of measured rings, the PL energy dependence with the applied magnetic field was found to follow a quadratic function of the type

\[
E_{\text{PL}}(B) = E_0 + (\frac{1}{2})g_{\text{ex}}\mu_B B + xB^2.
\]

Here \( g_{\text{ex}} \) is the effective exciton Landé factor and \( \mu_B \) is the Bohr magneton. The curvature \( x \) in this expression was found in all measured cases to be positive, characteristic of a diamagnetic behavior. We have analyzed the magnetic field of the PL for about 20 different rings. Whenever it was possible, we measured the excitonic emission of each ring as a function of charge, namely the neutral excitonic emission \( X^0 \), the singly and doubly charged exciton \( X^{1+} \) and \( X^{2-} \). In many cases charging with one excess electron could only be done in an energy level degenerate with the two-dimensional continuum in the InAs wetting layer. In the present analysis we choose to ignore such rings.

We found that we can classify the behavior of individual rings into two categories according to their diamagnetic-shift. Half of the measured rings have an excitonic diamagnetic shift of 10 µeV/T² independent of their state of charge as shown in Fig. 2a. We label these rings as type A. Remarkably, for this type of ring
Fig. 1. Left: gray scale of the PL emission intensity as a function of B and energy. White level is the background signal, black level = 100 counts/30 s above background. Right: the position of the PL peak against negative (positive) B for the lower (upper) spin-split branch. The solid line is the best quadratic fit with \( z = 11.0 \mu \text{eV}/\text{T}^2 \) and \( (\frac{1}{2}) \mu_{\text{eff}} B = 86 \mu \text{eV}/\text{T} \).

Fig. 2. Diamagnetic shift of the neutral and charged exciton in three different types of rings. Left panel, rings of type A with \( z = 10 \mu \text{eV}/\text{T}^2 \) and \( (\frac{1}{2}) \mu_{\text{eff}} B = 45 \mu \text{eV}/\text{T} \). The three different symbols represent \( X^0 \), \( X^{-1} \), and \( X^2^{-} \). Center: rings of type B. The neutral exciton \( (X^0) \) has \( z = 16.5 \mu \text{eV}/\text{T}^2 \) and \( (\frac{1}{2}) \mu_{\text{eff}} B = 48 \mu \text{eV}/\text{T} \), the charged exciton has \( z = 8.7 \mu \text{eV}/\text{T}^2 \) and \( (\frac{1}{2}) \mu_{\text{eff}} B = 56 \mu \text{eV}/\text{T} \). The right panel represents the type of ring that shows a clear drop of \( z \) (from 20 \( \mu \text{eV}/\text{T}^2 \) down to 13 \( \mu \text{eV}/\text{T}^2 \)) for \( B > 6 \text{T} \).

The diamagnetic shift was found to vary by only 10% from ring to ring. Another class of ring (type B) has the property that the diamagnetic shift reduces with the addition of one excess electron. A characteristic example of this type B behavior is shown in Fig. 2b. There the diamagnetic shift of the \( X^0 \) exciton is found to be 16.6 \( \mu \text{eV}/\text{T}^2 \) while that of \( X^{-1} \) is 8.7 \( \mu \text{eV}/\text{T}^2 \). We observed that all the type A rings emit in a band centered around 1.31 eV while the type B rings emit in a band centered around 1.34 eV. Correspondingly, a measurement of the PL emission of an ensemble of \( 10^6 \) rings also showed that there are two emission peaks at 1.31 and 1.34 eV.

We define a phenomenological excitonic diamagnetic shift \( \Delta E_{\text{ex}} = (e^2 \rho^2/8 \mu)B^2 \) where the characteristic confinement length \( \rho \), and the mass \( \mu \) are to be determined using specific theoretical models. Recently, Chaplik derived the diamagnetic shift of neutral and charged excitons confined in a ring with a parabolic radial potential [9]. Such a potential, which departs from the one-dimensional ideal ring, allows for a radial motion of the electron and hole. The noninteracting electron and hole ground states are characterized by the zero-point lengths \( l_e \) and \( l_h \), across the ring width. This model [9] predicts that for a neutral exciton in a ring the characteristic length \( \rho \)
is to be identified with \( \rho^2 = (l_e^2 + l_h^2) \) and the mass to be used is \( \mu = (m_e + m_h)/2 \). In this formulation, the diamagnetic shift of the neutral exciton does not relate to the ring radius \( R_0 \), instead it gives the characteristic width of the ring. Using a measured mass \[ 4 \] \( m_e = 0.07 \) and an assumed mass \( m_h = 0.3 \) for the hole, we obtain \((l_e^2 + l_h^2)^{1/2} = 9.5 \) nm for a type A ring. If instead of using a ring potential one assumes that the exciton experiences a normal central parabolic confinement, the characteristic measured length \( \rho \) is now to be identified with the exciton radius and the reduced mass \( \mu = (m_e^{-1} + m_h^{-1})^{-1} \). The diamagnetic shift of the type A rings would correspond within this model to an exciton radius of 4.8 nm. Since both diamagnetic lengths are consistent with the ring-sizes estimated from scanning force microscopy measurements, we see that at this point, using the magnetic behavior of the neutral exciton PL alone it is not possible to differentiate experimentally between a ring and central parabolic confinement. The magnetic property of charged rings is expected to be more informative. The model of Ref. \[ 9 \] predicts that in a ring, the PL of charged excitons shows a paramagnetic behavior (i.e. \( \chi < 0 \)). The change in sign for \( \chi \) is simple to understand; it arises from the electron left after exciton recombination. Since there is only an electron in the final state, its diamagnetic shift is sizably larger than that of the bound excitonic complex in the initial state. The differential shift between the initial and final states leads therefore to a negative curvature. A very similar prediction is also made for two-dimensional free trions \[ 10 \] but this was never clearly measured. Such a predicted negative curvature in the exciton emission was not observed in this experiment. Alternative to the ring-shaped potential, we also considered a parabolic central potential model. Two limits are discussed. First, in the strong confinement limit for which the zero-motion length of the harmonic potential is much smaller than the free exciton radius (\( \sim 12 \) nm), the prediction we make is that the diamagnetic curvature \( \chi \) of the PL emission increases slightly for singly and doubly charged excitons compared to neutral excitons. Including the Coulomb interaction as a weak perturbation, we find that \( \chi \) becomes more sensitive to electron–electron interactions with increasing electron number and increasing confinement length. Second, in the opposite limit of weak confinement, we predict that charged excitons should generally show a paramagnetic behavior (i.e. a negative \( \chi \)) while the neutral exciton has diamagnetic energy dispersion (i.e. a positive \( \chi \)). In the case of the singly charged exciton, the origin of this behavior is very similar to the prediction made for free trions in a magnetic field \[ 10 \]. For doubly and triply charged excitons, the correlated motion of the electrons and the hole is more complex and the PL spectrum in the weak confinement limit can include strong shakeup lines with even stronger paramagnetic dispersion. Since the limiting cases of strong and weak confinements result in opposite behaviors we expect that within a parabolic central potential model the dependence of \( \chi \) on the charge of the exciton should be a sensitive measurement of the confinement strength. Using such an argumentation, the data for the type A rings could then be interpreted using a parabolic potential in the strong confinement limit while the exciton of type B would correspond to intermediate confinement strength.

So far on grounds of the diamagnetic shift data alone it is presently not possible to differentiate between a ring-shaped and parabolic central confining potential. However, we expect to detect a signature of a ring-shaped confining potential when a flux quantum threads the nanostructure \[ 4 \]. When this happens, the orbital symmetry of the neutral exciton is expected to change by one quantum number. This is assuming that the exciton has nonzero polarization in the radial direction and so resembles a rotating dipole \[ 11 \]. By calculating single-particle wave functions we found that indeed electron and hole have different averaged radii due to different effective masses and thus a nonzero dipole moment in the exciton can exist. Due to optical selection rules for such an exciton, we expect a complete extinction of the PL emission \[ 11 \]. Such magnetic field induced quenching of the PL was not detected in our experiments. It is possible that the magnetic fields used here are not high enough to fulfill the condition for flux quantization. We have measured in three individual rings an unexpected sudden change in the diamagnetic curvature \( \chi \) at fields above 6 T. The PL peak position of the neutral exciton of such a ring is shown in Fig. 2c. For this particular set of data, the low-field diamagnetic quadratic term was found to be \( \chi = 20 \mu eV/T^2 \) while above 6 T this curvature reduced to 13 \( \mu eV/T^2 \). In contrast, the linear term (i.e. Zeeman effect) remained constant.
The sudden reduction at 6 T of the diamagnetic-shift is so far not understood but could be related to the threading of a flux quantum [12]. Such rings would have a hole diameter of 30 nm.

This work was supported by the DFG under SFB348 and EPSRC. One of us, A.O.G., acknowledges financial support from the Volkswagen-Foundation.

References