Emission from a highly excited single InAs-GaAs quantum dot in magnetic fields: An excitonic Fock-Darwin diagram

A. Babinski,[1,2] M. Potemski,[2] S. Raymond,[3] J. Lapointe,[3] and Z. R. Wasilewski[3]
[1]*Institute of Experimental Physics, Warsaw University, Hoza 69, 00-681 Warsaw, Poland*
[2]*Grenoble High Magnetic Field Laboratory, CNRS, Boîte Postal-166, 38042 Grenoble Cedex 9, France*
[3]*Institute for Microstructural Sciences, NRC, Ottawa, K1A 0R6, Canada*
(Received 25 July 2006; published 4 October 2006)

Magnetospectroscopy of a highly excited single InAs-GaAs quantum dot is performed. The multiexcitonic emission related to the *s*, *p*, and *d* shells of a zero-dimensional system is identified. Orbital Zeeman effects for the *p* and *d* energy shells are observed. The overall pattern of the magnetic field evolution of the emission lines resembles a single-particle Fock-Darwin diagram. Effects of interaction between the multiexcitonic configurations and the asymmetry of localizing potential are clearly visible when single-particle states become nearly degenerate: at $B=0$ and at a level crossing induced by the magnetic field.

DOI: 10.1103/PhysRevB.74.155301 PACS number(s): 78.67.Hc, 73.21.La, 78.55.Cr

The interesting phenomena which result from the interplay between quantum confinement and electron-electron interactions stimulate fundamental research on semiconductor quantum dot (QD) structures.[1] The "analytical" example of a semiconductor dot is a laterally confined two-dimensional (2D) system, with a parabolic lateral confinement potential. The discrete and energetically equidistant single-particle levels of such a dot are obtained as a simple solution of the 2D harmonic oscillator. These levels are referred to as $s, p, d, \ldots,$ (atomiclike) shells with their characteristic twofold, fourfold, sixfold, …, degeneracy, respectively. The quantum confinement and electron-electron interactions in such a dot can be effectively modified by the application of a magnetic field. A magnetic field applied perpendicularly to the initial 2D plane lifts the orbital degeneracy of a single harmonic oscillator. The resulting Fock-Darwin (FD) diagram[2] in the energy versus magnetic field coordinates displays a characteristic diamondlike level crossing pattern, which implies the reconstruction of the 2D Landau-level structure in the high field limit. The physics becomes more appealing when dots are filled with many particles and electron-electron interactions become important. Characteristically for a quantum system the interactions are expected to be most apparent when single-particle levels are nearly degenerate: at $B=0$ and/or in the vicinity of the crossing points at $B\neq 0$. The structures whose properties best resemble those of "ideal zero-dimensional objects" are relatively large dots, electrostatically shaped from the 2D electron gas. A number of interesting phase transitions tuned by the number of electrons and/or the magnetic field in low-temperature magnetotunneling experiments have been convincingly shown in such structures.[3] The few-electron/hole ground states have also been investigated in smaller, self-assembled InAs-GaAs QDs.[4] In optical measurements on such QDs this picture becomes more complicated, as both electrons and holes are involved in observed transitions. The shell structure of a single "optical dot" is often observed in zero-field photoluminescence (PL) experiments and a number of works have been focused on the evolution of the ground state *s*-like transitions as a function of the magnetic field.[1] Until now, however, the magneto-PL from the excited $p, d, \ldots,$ shells has been only investigated on ensembles of QDs.[5–7] These measurements were unable to reveal details of the magnetic field evolution of multiexcitonic spectra due to inhomogeneous broadening of emission from millions of QDs.

In this paper we report the observation of magnetic field evolution of the *s*-, *p*-, and *d*-like emission due to multiexcitonic complexes in a single QD. We show that a single-particle approach to describe the evolution of multiexciton levels observed for a highly excited dot is rather robust. An overall "landscape" of the emission spectra in *E*-*B* coordinates resembles a simple FD diagram quite closely. More subtle effects are revealed when single lines, related to *s*, *p*, and *d* levels are investigated. Electron-electron interactions[8] in combination with possible effects of asymmetric confinement potential[9] are observed to lift the degeneracy of single-particle states at zero magnetic field as well as at the crossing point induced by the magnetic field. In the experiment, this is manifested as the effect of avoided crossing of transitions which can be modeled in terms of repulsion between single-particle FD levels.

The sample investigated in this work was grown by molecular beam epitaxy on an n^+-GaAs substrate. It contains a single layer of InAs QDs In flushed at 5 nm (Ref. 10) grown on a 600 nm undoped GaAs buffer layer and capped with a 100 nm GaAs layer. The structure was annealed after growth (30 s at 850 °C) to shift the emission into the sensitivity range of a CCD camera and to decrease the confining potential in the QDs.[11] A set of mesa structures was prepared on the sample to limit the number of dots addressed optically. The sample was immersed in liquid helium ($T=4.2$ K) and magnetic field was applied perpendicularly to the sample surface. The excitation laser light ($\lambda=514.5$ nm) was coupled to a single-mode fiber, delivered to the sample and focused by two aspheric microlenses. The obtained spot size was of the order of 10 μm, which assured excitation of a single mesa. The PL was collected by a multimode fiber (600 μm core).[12] The relative position of the sample with respect to the optical setup was controlled by piezodriven positioners. The PL was transmitted to a 1 m double-grating monochromator and detected by a liquid nitrogen cooled CCD camera. Several mesas have been inspected and one of them has been chosen for more extensive studies presented in this paper.

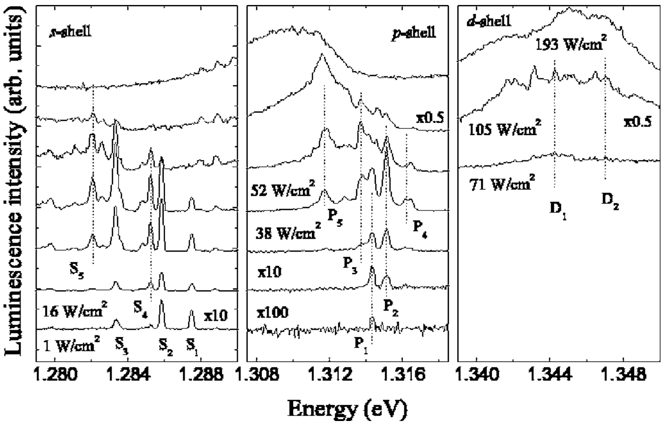

FIG. 1. The emission from the s, p, and d shells of a single dot as a function of excitation power at $T=4.2$ K in zero magnetic field. The spectra are offset for clarity.

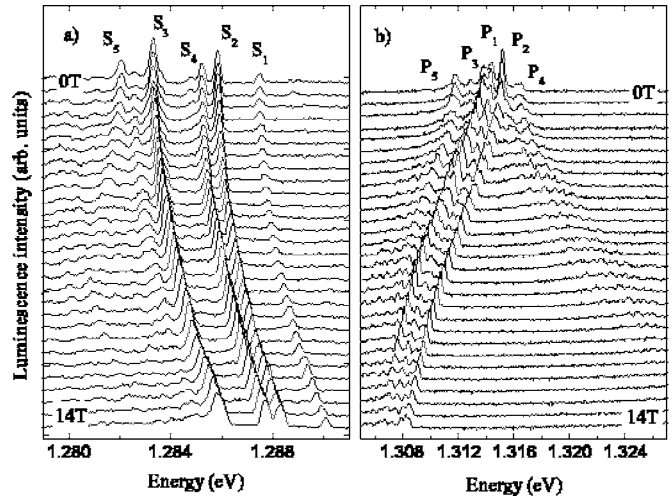

FIG. 2. Luminescence spectra from the s shell (a) and the p shell (b) of a single InAs-GaAs quantum dot in magnetic field up to 14 T (excitation power density ~ 55 W/cm^2). The spectra are offset for clarity. Note different energy scales in both panels.

The evolution of the PL spectra from a submicron-size mesa as a function of the excitation power is shown in Fig. 1. The spectrum obtained at the lowest excitation power accessible in our experiments (~ 1 W/cm^2) is dominated by two emission lines labeled S_1 and S_2, accompanied by weaker emission lines S_3 and S_4. At higher energy (approximately 30 meV) a weak emission line P_1 can also be observed. The multiline pattern observed in the low-energy range was found in the spectra for several different mesas in the investigated sample and in our opinion it is characteristic for the emission from the s shell of an individual dot in the structure.[13] We have found that the relative intensity of the S_1 and S_2 lines measured at low excitation changes from dot to dot, and moreover when observed from a single mesa (dot) these emission lines change their relative intensities as a result of thermal cycling. With this observation it is tempting to relate the S_1 and S_2 lines to the ground state emission from different charge states of a single QD and the S_3 and S_4, which gain intensity with increasing excitation power, to biexcitonic complexes. The effects of "fluctuating charge" are typical for optical spectra of single dots and in our experiments we believe to effectively probe at least two objects: a neutral and a charged dot. Whereas S_1 is assigned to a neutral exciton, the S_2 line is likely due to a negatively charged exciton.[14] Some ambiguity in the assignment of these lines does not limit the general character of our conclusions, as the attribution of observed emission lines to particular shells, which is reflected in our labeling, is confirmed by their characteristic behavior in magnetic field. With increasing excitation power new s-shell-related (S_5) and p-shell-related (P_2 and P_3) emission lines emerge. These must involve NX excitonic complexes with $N \geq 3$, which should give rise to emission lines associated with the s as well as the p shell.[15] Further increase of excitation power (52 W/cm^2) results in new emission lines in both s-related and p-related energy range and in particular emission lines marked as P_4 and P_5. Examining the power dependence we can see that the energy positions of individual emission lines do not depend on excitation power. This is an obvious result of their relation to particular multiexcitonic configurations, but this also confirms that the sample temperature does not change during the experiment. The highest excitation power results in further change of the s-related and p-related emission, as well as in a new emission band at higher energy. The latter emission results from the d shell of the investigated dot. Two d-related emission lines (which can be followed in magnetic field) are labeled D_1 and D_2.

The attribution of the emission lines to particular shells of the dot is unambiguously supported by their behavior in magnetic field. The magnetic-field evolution of the s- and p-related emission is shown in Fig. 2. The S_1, S_2, S_3, and S_4 lines shift diamagnetically in magnetic field. The energies of those lines can be well described with a parabolic dependence. This is in agreement with our attribution, since the energies of a neutral and a singly charged excitons as well as biexcitons in QDs are known to depend quadratically on magnetic field.[16] The P_1, P_2, P_3, and P_5 emission lines redshift, whereas those labeled P_4 blueshift in magnetic field.

Although the orbital Zeeman splitting of the p-shell emission lines is clearly visible in our experiment, no spin splitting of the s-shell (nor p-shell) emission lines can be observed in magnetic field up to 14 T.[17] We have found this behavior for all dots in the investigated structure.[13] In our opinion this is due to the complicated character of hole states in our relatively tall dots. It has recently been proposed that the ground state of the hole in similar InGaAs/GaAs dots is only 73% heavy-hole-like.[18] A vanishing low-field effective g^* factor of excitons in tall dots has also been postulated theoretically.[19]

To analyze the evolution of spectra in magnetic field we start with the FD spectrum of single-particle electron and hole levels. The energy E_{nl} of an electronic (hole) state with a radial quantum number n $(=0,1,2,\ldots)$, and an angular quantum number $l(=0,\pm 1,\pm 2)$ confined in a parabolic potential $\hbar\varpi_0^e(\hbar\varpi_0^h)$ in the presence of a perpendicular magnetic field B is given by $E_{nl}^{\beta}=(2n+|l|+1)\hbar\Omega_{\beta}+\frac{1}{2}l\hbar\varpi_c^{\beta}$, with $\varpi_c^{\beta}=\frac{eB}{m_{\beta}}$ and $\Omega_{\beta}=\sqrt{(\varpi_0^{\beta})^2+\frac{1}{4}(\varpi_c^{\beta})^2}$, where β stands for elec-

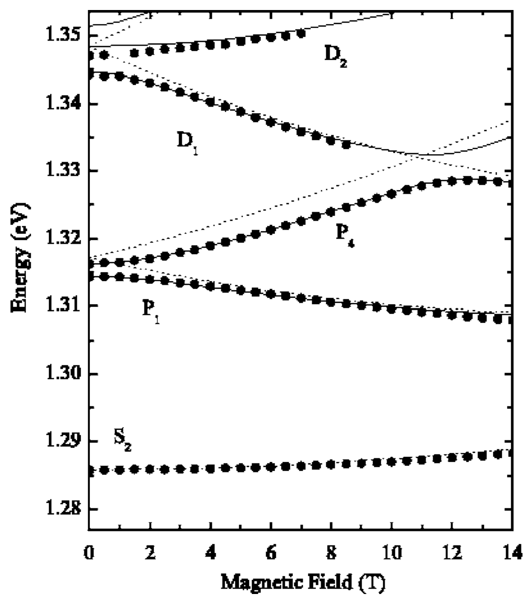

FIG. 3. Energies of the S_2, P_1, P_4, D_1, and D_2 emission lines plotted against magnetic field. Symbols give the experimental data. The calculated excitonic FD spectrum with E_g=1.25455 eV, $\hbar\omega_0$ =31.3 meV, and $\mu^*=0.057m_0$ is shown with dotted lines. Solid lines represent a simulation based on the FD diagram, which includes the zero-field splitting of the p and d shells and the high-field interaction between the p_+ and d_- levels (see text).

tron or hole. Let us assume that the condition of a "hidden symmetry": $m_e\varpi_0^e = m_h\varpi_0^h$ is fulfilled,[8] which implies an identical form of electronic and hole wave functions on their corresponding s, p, d,…, shells. The energy difference between the electronic and hole single-particle levels of the same sets of quantum numbers ΔE_{nl} (which are involved in dipole-allowed optical transitions) can then also be expressed in terms of an "excitonic" (electron-hole pair) FD diagram $\Delta E_{nl} = E_g + (2n + |l| + 1)\hbar\Omega + \frac{1}{2}l\hbar\varpi_c$ with $\varpi_0 = \varpi_0^e + \varpi_0^h$ and $\varpi_c = \frac{eB}{\mu^*} = \left(\frac{1}{m_e} + \frac{1}{m_h}\right)eB$. The effective energy gap E_g in this model accounts for the energy difference between the top of the valence band and the bottom of the conduction band and effectively for the Coulomb interactions within a particular excitonic configuration.

To examine the validity of the FD model in the description of our results we will focus our attention on a few emission lines related to subsequent shells of the dot, which can be followed in magnetic field (see Fig. 3). The data shown in Fig. 3 have been extracted from spectra excited with several values of laser power (from 55 to 83 W/cm^2). Dashed lines in Fig. 3 show the excitonic FD diagram obtained with the following parameters, which will be justified later: reduced electron-hole mass $\mu^*=0.057m_0$, confinement energy $\hbar\omega_0$ =31.3 meV, and the energy gap E_g=1.25455 eV. The evolution of the S_2 emission line in magnetic field is well described by the FD model except for the highest magnetic fields. This high-field discrepancy must result from the changes of excitonic binding energy in magnetic field,[5] which is not accounted for in our model. The evolution of the P_1 emission line deviates from the behavior expected from the FD model both in high and low magnetic fields. The high-field discrepancy is similar to that observed for the S_2 line. The low-field deviation, which we think is partly due to the asymmetry of the lateral potential localizing carriers in the dot, will be analyzed in the following. In the FD model of a symmetric lateral potential, the p-shell states with angular quantum numbers $l=-1$ and $l=+1$ (referred to as the p_- and p_+ levels, respectively) are degenerate in zero magnetic field. Interaction of magnetic moments of recombining excitonic configurations with the magnetic field removes the degeneracy. The orbital Zeeman splitting of single-particle p levels is expected to be linear in magnetic field $\Delta E(B) = \hbar\varpi_c$. Our experimental results indicate that in real QDs this simple model does not apply. The p_- and p_+ levels are mixed as a result of the asymmetry of lateral potential and electron-electron interactions.[8,9,20] This results in the p_x and p_y levels, which in high magnetic field evolve into the p_- and p_+ levels. Using a simple perturbation theory we can describe the energy difference between the emission lines related to the p_x (P_1,P_2,P_3) and p_y (P_4) levels by the following equation:

$$\Delta E'(B) = E_\Delta + \sqrt{\delta^2 + (\hbar\varpi_c)^2}. \quad (1)$$

The energy splitting $\Delta E'(B)$ accounts for possible effects of electron-electron interactions (E_Δ) and the zero-field splitting of the single-particle p levels (δ) due to asymmetry of the lateral confining potential. We have found that the splitting can be reproduced with a single reduced electron-hole effective mass $\mu^*=0.057\pm0.0005m_0$, the zero-field splitting δ=5.5±0.5 meV, and the energy E_Δ ranging from −4.35 meV (P_4-P_2) to −2.6 meV (P_4-P_3). The magnitude of the zero-field splitting agrees with the values of electronic p-shell splitting found in far-infrared experiments, being of the order of 5 meV (Ref. 21) or theoretical value (3 meV) for a similar lens-shaped dot.[8] Using the obtained values of reduced mass μ^* and the zero-field splitting δ, as well as the confinement energy $\hbar\omega_0$=31.3 meV (related to the energy difference between the P_1 and S_2 emission lines) we can describe the magnetic-field evolution of the P_1 emission line (see full line in Fig. 3).[22] A similar approach has been applied to account for the low-field evolution of the d_x (d_-) related D_1 emission line (with the respective zero-field splitting of the d shell equal to δ_1=6.9 meV). The evolution of the D_2 emission line related to the d_0 shell can be reasonably reproduced by the respective unperturbed FD level.

To explain the magnetic-field evolution of the P_4 emission line, which is related to the p_y (p_+) shell, both the zero-field splitting of the p shell and its apparent anticrossing with the emission lines related to the d_- levels in high magnetic field[7] must be taken into account. The latter interaction is an inherent property of excitonic systems. Five (and more) exciton systems, which are related to the p_+ (before crossing) and d_- (after crossing) shells involve electron-hole pairs with zero total angular momentum and they hybridize in the vicinity of the crossing.[23] Phenomenologically, this behavior can be reproduced with a simple perturbation approach, by introducing a phenomenological interaction parameter ε=2 meV, which mixes both configurations. The obtained theoretical dependence describes the evolution of the P_4 emission line

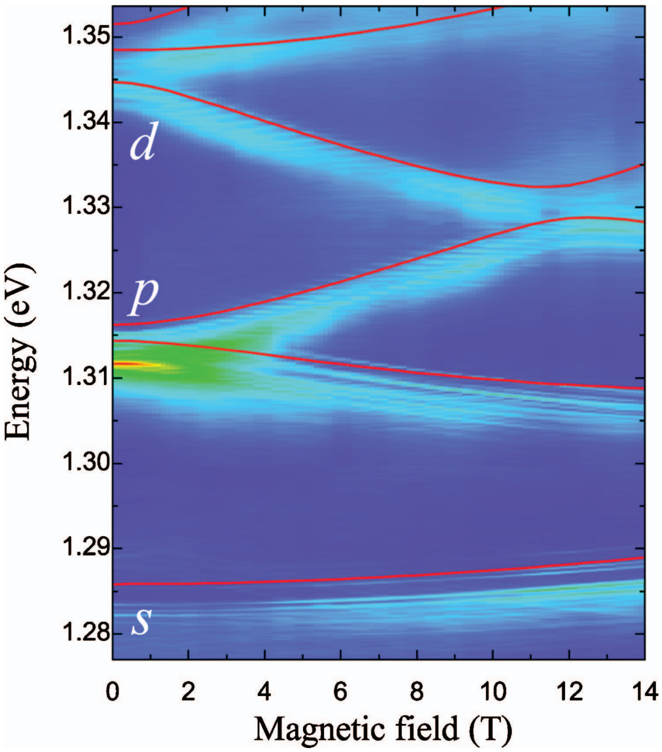

FIG. 4. (Color) Luminescence from the s, p, and d shells of a single InAs-GaAs quantum dot in magnetic field up to 14 T. The spectra were excited with a power density of 83 W/cm^2. The simulation based on the FD diagram, which includes the zero-field splitting of the p and d levels and the high-field interaction between the p_+ and d_- levels (see text) is shown in red.

well, which can be seen in Fig. 3. The PL spectra excited with high power are summarized in the form of a surface plot in Fig. 4. The previously described theoretical dependences are also shown in Fig. 4. It can be seen that the emission related to the s, p, and d shells of the dot follow the results of our simulations. Energy shifts of particular emission lines with respect to thetheoretical dependences reflect differences in electron-electron interactions for different multiexcitonic configuration.

So far, the single-dot magneto-optics was limited to investigations of the emission related to the ground, s-shell states. The diamagnetic shift and Zeeman splitting are two focal effects observed for these lines which are relatively easy to interpret and which permit to extract certain information about the electronic properties of the investigated structures.[19,24] Notably, more input can be expected from the analysis of the magnetic field evolution of excited states. However, in high excitation luminescence experiments, these excited states are probed via emission due to multiexcitonic complexes which involve strong effects of electron correlations and may imply the need for a description far beyond the simplistic (one-particle) approach.[8] In spite of this expectation, the magneto-luminescence spectra of highly excited ensembles of dots have been reported to reflect simple one-particle FD diagrams, but observation of averaged spectra prevented us from drawing firm conclusions. Here we have extended this model with phenomenological corrections accounting for the effects of a lateral potential asymmetry and electron-electron interactions in terms of appropriate energy shifts and level repulsions. We have shown that this simple approach is satisfactory to describe a series of the individual multi-magneto-excitons resonances observed in experiments on single QDs. Thus, we conclude that the results of the magnetoluminescence spectroscopy of highly excited single QDs may be effectively used to investigate the pronounced changes of QD excited states as a function of the magnetic field. This makes such a spectroscopy a powerful tool to study the electronic properties of these systems. We believe that our results will stimulate further work and, in particular, theoretical studies. The latter, likely including the realistic many-body calculations, could be especially valuable to firmly identify the specific emission lines observed in the experiment and to elucidate the exact physical meaning of parameters introduced here only on phenomenological grounds.

Summarizing, we have presented the spectroscopic measurements of a highly excited single InAs-GaAs QD. Sharp emission lines due to multiexcitonic complexes related to the s, p, and d shells of the dot have been identified and investigated in magnetic field. The orbital Zeeman effects for the p and d shells have been observed. Effects of the electron-electron interactions as well as the asymmetry of the lateral potential have been observed. It has been shown that the magnetic field evolution of the emission can be well described by the modified single-particle excitonic Fock-Darwin model.

Valuable discussions with P. Hawrylak and W. Sheng are kindly acknowledged. The EC (Grant Nos. RITA-CT-2003-505474 and ICA1-CT-2002-70010) and the Polish Ministry of Education and Science (Grant No. 1 P03B 014 29) are acknowledged for support.

[1] *Optics of Quantum Dots and Wires*, edited by G. W. Bryant and G. S. Solomon (Artech House, Boston, 2005).

[2] V. Fock, Z. Phys. **47**, 446 (1928); C. G. Darwin, Proc. Cambridge Philos. Soc. **27**, 86 (1930).

[3] S. Tarucha, D. G. Austing, T. Honda, R. J. van der Hage, and L. P. Kouwenhoven, Phys. Rev. Lett. **77**, 3613 (1996); L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, Rep. Prog. Phys. **64**, 701 (2001); M. Ciorga, A. Wensauer, M. Pioro-Ladriere, M. Korkusinski, J. Kyriakidis, A. S. Sachrajda, and P. Hawrylak, Phys. Rev. Lett. **88**, 256804 (2002); *Electron Transport in Quantum Dots*, edited by J. P. Bird (Kluwer Academic Publishers, Boston, 2003).

[4] B. T. Miller, W. Hansen, S. Manus, R. J. Luyken, A. Lorke, J. P. Kotthaus, S. Huant, G. Medeiros-Ribeiro, and P. M. Petroff,

Phys. Rev. B **56**, 6764 (1997); R. J. Warburton, B. T. Miller, C. S. Dürr, C. Bödefeld, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro, P. M. Petroff, and S. Huant, Phys. Rev. B **58**, 16221 (1998); D. Reuter, P. Kailuweit, A. D. Wieck, U. Zeitler, O. Wibbelhoff, C. Meier, A. Lorke, and J. C. Maan, Phys. Rev. Lett. **94**, 026808 (2005).

[5] R. Rinaldi, P. V. Giugno, R. Cingolani, H. Lipsanen, M. Sopanen, J. Tulkki, and J. Ahopelto, Phys. Rev. Lett. **77**, 342 (1996).

[6] P. P. Paskov, P. O. Holtz, B. Monemar, J. M. Garcia, W. V. Schoenfeld, and P. M. Petroff, Phys. Rev. B **62**, 7344 (2000).

[7] S. Raymond, S. Studenikin, A. Sachrajda, Z. Wasilewski, S. J. Cheng, W. Sheng, P. Hawrylak, A. Babinski, M. Potemski, G. Ortner, and M. Bayer, Phys. Rev. Lett. **92**, 187402 (2004).

[8] P. Hawrylak, Phys. Rev. B **60**, 5597 (1999).

[9] G. Bester and A. Zunger, Phys. Rev. B **71**, 045318 (2005).

[10] Z. R. Wasilewski, S. Fafard, and J. P. McCaffrey, J. Cryst. Growth **201/202**, 1131 (1999).

[11] S. Fafard and C. Ni. Allen, Appl. Phys. Lett. **75**, 2374 (1999).

[12] A. Babinski, S. Awirothananon, J. Lapointe, Z. Wasilewski, S. Raymond, and M. Potemski, Physica E (Amsterdam) **26**, 190 (2005).

[13] A. Babinski, G. Ortner, S. Raymond, M. Potemski, M. Bayer, W. Sheng, P. Hawrylak, Z. Wasilewski, S. Fafard, and A. Forchel, Phys. Rev. B **74**, 075310 (2006).

[14] Although we are unable to precisely determine if the extra charge of charged excitons is positive or negative, we believe that the latter attribution is correct, as the sample was grown on an *n*-type substrate. Relative energies of the S_1 and S_2 agree with theoretical values obtained for a neutral and a singly charged exciton in similar dots.

[15] M. Bayer, O. Stern, P. Hawrylak, S. Fafard, and A. Forchel, Nature (London) **405**, 923 (2000).

[16] C. Schulhauser, D. Haft, R. J. Warburton, K. Karrai, A. O. Govorov, A. V. Kalameitsev, A. Chaplik, W. Schoenfeld, J. M. Garica, and P. M. Petroff, Phys. Rev. B **66**, 193303 (2002).

[17] The experiments performed at higher magnetic fields show that the Zeeman spin-splitting of the *s*-shell related emission lines becomes apparent in the spectra at fields above ~20 T.

[18] G. Bester, S. Nair, and A. Zunger, Phys. Rev. B **67**, 161306(R) (2003).

[19] W. Sheng *et al.* (unpublished).

[20] S. Hameau, Y. Guldner, O. Verzelen, R. Ferreira, G. Bastard, J. Zeman, A. Lemaître, and J. M. Gérard, Phys. Rev. Lett. **83**, 4152 (1999).

[21] E. A. Zibik, L. R. Wilson, R. P. Green, G. Bastard, R. Ferreira, P. J. Phillips, D. A. Carder, J-P. R. Wells, J. W. Cockburn, M. S. Skolnick, M. J. Steer, and M. Hopkinson, Phys. Rev. B **70**, 161305(R) (2004).

[22] The confinement energy $\hbar\omega_0 = 31.3$ meV and the reduced mass $\mu^* = 0.057m_0$ are also used to obtain the FD diagram shown in Fig. 3 with dashed lines.

[23] S.-J. Cheng, W. Sheng, and P. Hawrylak, Phys. Rev. B **68**, 235330 (2003).

[24] M. Bayer, G. Ortner, O. Stern, A. Kuther, A. A. Gorbunov, A. Forchel, P. Hawrylak, S. Fafard, K. Hinzer, T. L. Reinecke, S. N. Walck, J. P. Reithmaier, F. Klopf, and F. Schäfer, Phys. Rev. B **65**, 195315 (2002).