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Ground-state emission from a single InAs/GaAs self-assembled quantum dot structure in ultrahigh magnetic fields

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We report on the magnetic field dispersion of the exciton spin-splitting and diamagnetic shift in single InAs/GaAs quantum dots up to $B=28$ T. We have found substantial differences between field evolution of the emission from tall (4 nm height) and flat (2 nm height) dots. Strongly nonlinear spin-splitting is observed in the former case, while in the latter case the nonlinearity is much weaker. The diamagnetic shift of the ground-state emission can be explained in terms of geometric size of the dots. While it can be approximated by a quadratic field dependence in dots of large confining potential, a substantial linear contribution must be included to account for the diamagnetic shift in dots of weaker confinement.

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I. INTRODUCTION

Due to its high level of component miniaturization and integration, semiconductor nanotechnology appears to be highly attractive for scalable quantum information processing.[1] Semiconductor quantum dots (QDs) offer charge and spin excitations for usage as quantum bits.[2–4] Lately, proposals have been made to combine the advantages that both of them offer for these purposes: spins may provide long coherence times, and charges may offer easy coherent manipulation. Thus, electron spins could be used for information storage, and for processing they may be swapped into charge by optically injecting electron-hole pairs through laser pulses, for example.[5] Proposals for quantum bit and quantum gate operation along these lines rely heavily on well-defined optical selection rules for electron-hole excitation. It is widely accepted that in self-assembled QDs, which are often considered for the experimental realization of the proposed schemes, the intrinsic strain induces a large splitting between heavy and light holes, in addition to the confinement-induced splitting. Therefore, for the exciton ground state, "clean" selection rules might be expected. In fact, it has been found that in flat self-assembled $In_xGa_{1-x}As/GaAs$ QDs, the valence band ground state is at least 95% heavy hole.[6] However, theoretical calculations pointed out that the heavy-hole light-hole mixing cannot be neglected in tall dots.[7] This should be reflected by the Zeeman splitting of QD excitons in magnetic field. In this manuscript, we present experimental results that may confirm those predictions. To this end, we use optical studies of single QD structures of different morphology up to very high magnetic fields. We also investigate the diamagnetic shifts of those excitons and relate them to geometrical sizes of investigated dots.

II. EXPERIMENTAL DETAILS

We report photoluminescence (PL) measurements of *single* QDs in magnetic fields up to 28 T applied in the Faraday configuration. The technique to address single quantum systems in such high magnetic fields has only very recently been accomplished.[8] For that purpose, samples in which single quantum objects were geometrically isolated in submicron-size mesas were placed into the liquid helium insert of a cryostat at $T=4.2$ K. The cryostat is located in a Bitter-magnet. As this setup precludes optical access through windows, a fiber system was used for the studies. The laser excitation light (Ar^+ laser at $\lambda=514.5$ nm) is led to the sample via a single-mode fiber and focused by a combination of two aspheric microlenses. The obtained spot size is of the order of 10 μm. The emission signal is collected by a large, 600 μm-core multimode fiber. To address specific positions on the sample, it was mounted on piezo-driven stages. By using such a high stability stage which can be operated also in magnetic field, single quantum structures can be positioned precisely below the spatially fixed fiber optics. The PL has been analyzed by a 1 m double grating monochromator and detected by a liquid nitrogen cooled charge-coupled device (CCD) camera.

The QDs were fabricated by self-assembly extended by an In flush: After its deposition, the dot sheet is covered by a GaAs protection layer with a few nm thickness. Then uncovered In atoms from the upper part of dots are removed, resulting in a dot geometry which can be approximately described by a disk.[9] Two kinds of structures were grown: in the tall (flat) dots the thickness of the protection layer was 5 (3) nm, resulting in dots with a height of about 4 (2) nm after the flush. The sample with tall dots has been additionally annealed after the growth to shift the ground-state emission into the sensitivity range of a Si-CCD camera and to further increase the effective dot size.

The characteristic emission from the *s* shell of several single QDs in the sample with tall dots observed with excitation power density of the order of 1 W/cm^2 is shown in Fig. 1. The spectra comprise few emission lines with a well-reproducible pattern. The most pronounced features are

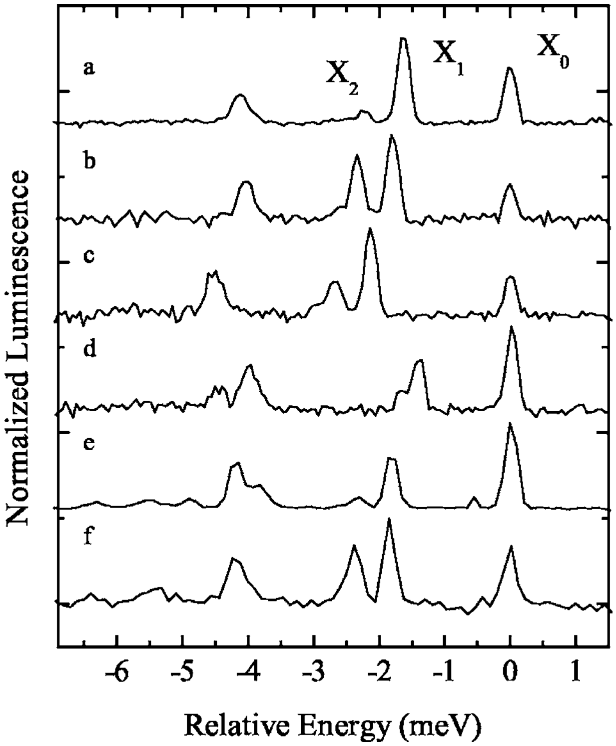

FIG. 1. The normalized luminescence spectra (T=4.2 K) from the s-shell of several single tall dots with the energy of the X_0 emission line equal to 1.2875 eV (a), 1.3121 eV (b), 1.2745 eV (c), 1.2710 eV (d), 1.2700 (e) and (f). Note the effect of thermal cycling on the emission from the same dot (e) and (f). The excitation power density is approximately 1 W/cm^2.

labeled in Fig. 1 with X_0, X_1, and X_2 [Figs. 1(a)–1(e)]. The relative intensities of the features vary from dot to dot. Moreover, for a particular dot the relative intensities change with thermal cycling [Figs. 1(e) and 1(f)], which has not been observed for the flat dots. In our opinion the emission lines originate from different charge states (or different exciton occupations) of a single dot. Simultaneous emission from different charge states was previously observed in QD field-effect structures[10,11] or in unintentionally doped QDs.[12,13] The effect has been attributed to the statistical nature of photoexcited carrier capture into a dot. In the present experiment with nominal flat-band condition, the effect may also be pronounced as both strongly doped substrate and fluctuating charges on the mesa walls may contribute to the actual potential environment of the investigated dot. The unambiguous attribution of the observed emission lines to particular charge states of a single dot is not straightforward, as the charge state cannot be externally controlled in our experiment. The lack of unambiguous identification of the emission lines does not, however, limit the general character of conclusions drawn in the following.

III. EXPERIMENTAL RESULTS

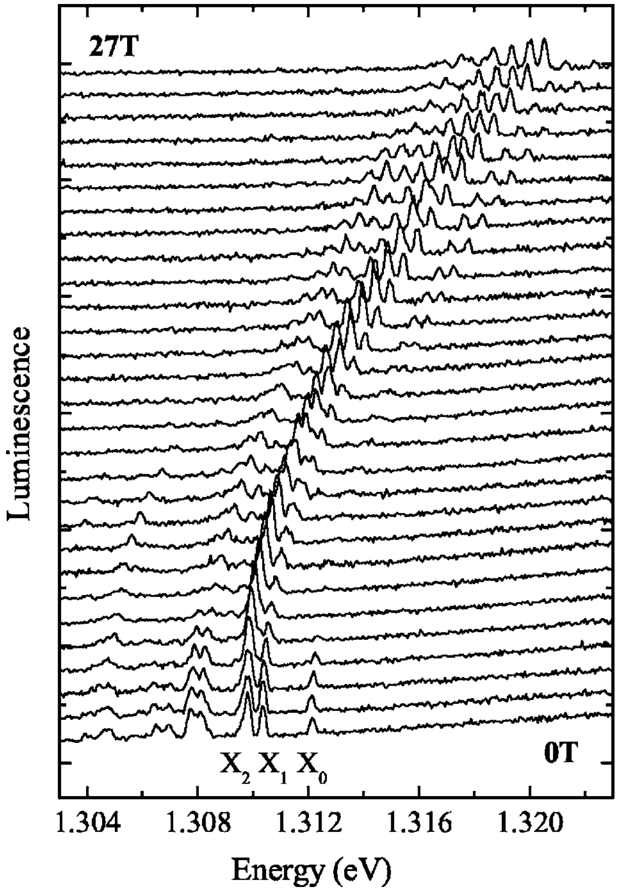

FIG. 2. The luminescence (T=4.2 K) from the s-shell of a single tall QD in magnetic field. The multiline spectrum with X_0, X_1, and X_2 emission lines as well as emission lines related to the multiexciton occupation of a single dot are presented.

The magnetic-field evolution of the photoluminescence spectra from the s-shell of a single tall dot (compare spectrum b in Fig. 1) is shown in Fig. 2. The X_0, X_1, and X_2, as well as other emission lines due to multiexcitonic configurations in a single dot, can be identified. The emission lines blueshift in magnetic field. A striking behavior of the X_0 emission line in magnetic field is noticed: The emission line disappears from the spectrum in magnetic field around 4 T and recovers at ~13 T. The disappearance is accompanied by an increase of the X_2 emission line intensity. This change of intensities may support our attribution of the observed lines to different charge states of a single dot. The intensity of the X_1 emission line does not change considerably between 4 T and 13 T, indicating that the excitation power density stays roughly constant. Most likely the light-induced charging efficiency changes in that field range and the investigated dot stays longer in one of its possible charge configurations (X_2). As a result the emission from the other charge configuration (X_0) becomes weaker.

More surprisingly, no splitting can be resolved below about 10 T for all the emission lines. Only for higher fields are splittings observed, which scale linearly with magnetic field, as can be appreciated in Fig. 3. Both diamagnetic shift and Zeeman splitting of all emission lines are identical within the experimental error. This behavior agrees with our attribution of the observed lines to different charge states of a single dot. The diamagnetic shift of neutral, singly charged,

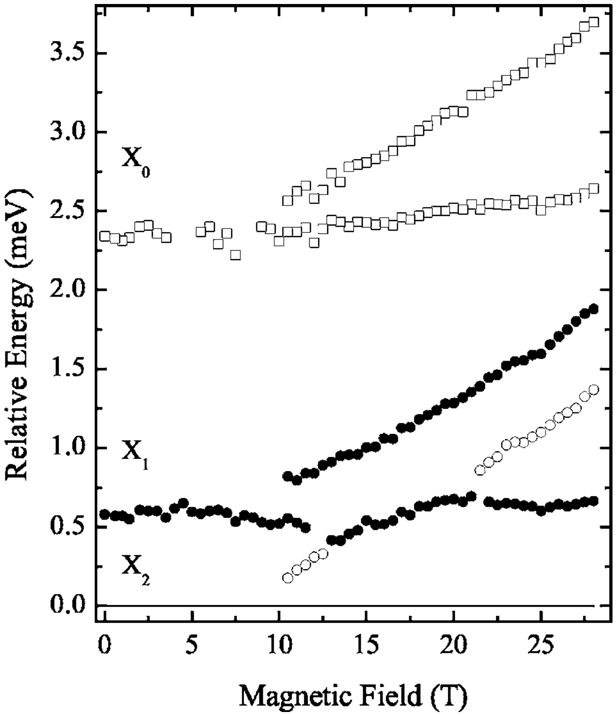

FIG. 3. The energies of the X_0, X_1, and X_2 emission lines with respect to the energy of the lower-energy branch of the X_2 emission line in magnetic field.

and doubly charged excitons in a strong confinement limit (which is the case of our dots) for similar InAs/GaAs QDs has been shown to differ by less than 10%, which is comparable to our experimental resolution.[14] Also the Zeeman splitting has been found to be the same for the three charged states.[15,16]

The spectroscopic data obtained for the tall dots can be compared with those from the flat dots. The two panels of Fig. 4 show photoluminescence spectra of single QDs from two different sets of flat structures for varying magnetic fields. Nominally the two QD sets in Fig. 4 have the same material composition, but the ground-state exciton energies are separated by about 50 meV (also in the ensemble). In the left panel, the zero-field emission spectrum is dominated by a single line at ~1.26 eV, which in magnetic field splits mostly into a doublet and can be attributed to recombination of bright excitons with angular momentum $|M|=1$.[17] Also a few other features with considerably weaker intensities appear: They can be traced to other QDs from which emission is collected by the fiber (at high B, for example, the features on the low-energy side) or may originate from recombination of predominantly dark excitons with angular momentum $|M|=2$, which are confined in the same QD.[17] Since the precise origin of these faint lines is hard to assess, we focus in the following only on the two split features of strong intensity. In the right panel a pair of spectral lines is observed at $B=0$, which arise from exciton recombination at ~1.31 eV. The multiline spectrum is similar to the observed one in tall dots. The same mechanism of charge fluctuations is proposed to explain the doublet emission with the same B dispersion of the two features. Again, only the doublet splittings of the two strong emission features can be uniquely analyzed, so that we restrict our analysis to them.

From the data in Figs. 2 and 4, we have extracted the spin-splitting of the emission lines as well as the diamagnetic shift of the centers of the split lines. Figure 5 shows the magnetic field dependencies of these quantities.

IV. DISCUSSION

Magnetic-field dispersions of the Zeeman splitting and the emission energy can be analyzed based on the results of pre-

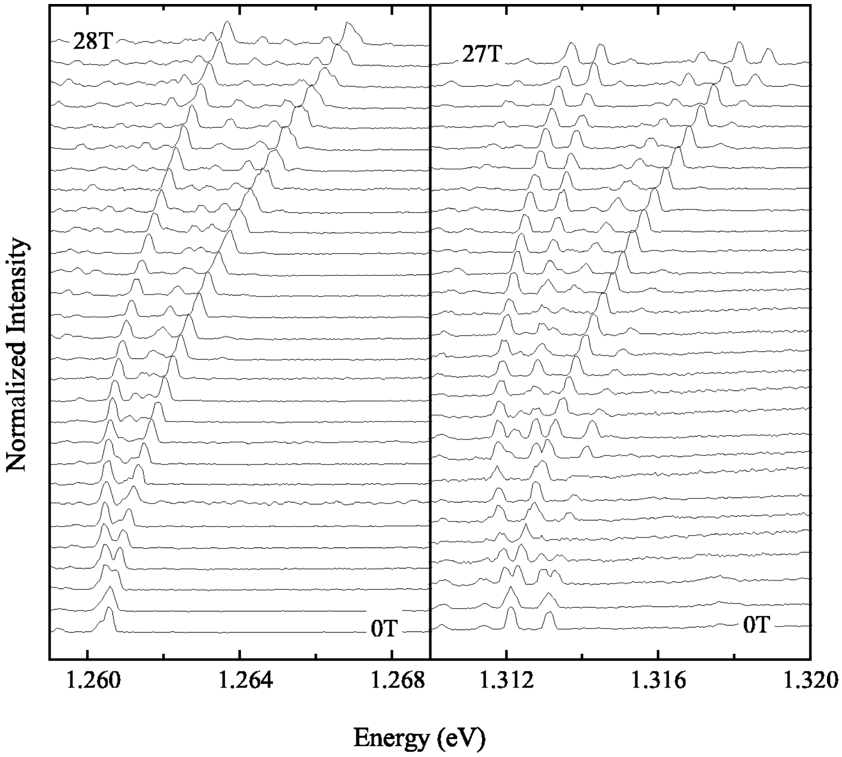

FIG. 4. Luminescence spectra of single InAs/GaAs flat QDs in magnetic field up to 28 (27) T. In the left panel, spectra from a single dot emitting around 1.26 eV are shown; in the right panel, the emission occurs at about 1.31 eV.

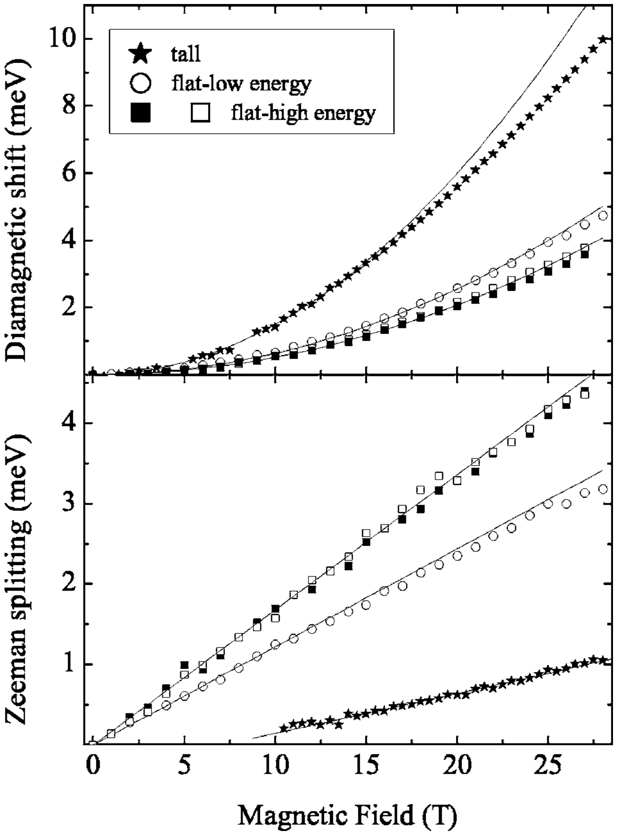

FIG. 5. Upper panel: the diamagnetic shift of the X_0 emission line from a tall dot and of the three emission lines from flat dots vs magnetic field. Here the solid lines are fits to the low-field data below 10 T following B^2 dependencies. Lower panel: spin-splitting of the X_0 emission line from a tall dot and of the three emission lines from flat dots vs magnetic field. Solid lines represent respective linear field dependences (for flat dots fits to low-field data below 10 T).

vious single QD PL studies up to about 10 T.[18] Two features are expected for these dispersions: (a) the exciton spin-splitting should vary linearly with magnetic field, $\Delta_{\text{spin}} = (g_e + 3g_h)\ \mu_B B$, where the g_i are the g factors of electron and hole, and (b) the diamagnetic shift should depend quadratically on magnetic field

$$\Delta_{\text{diamag}} = \frac{e^2}{8}\left(\frac{\langle x_e^2 + y_e^2\rangle}{m_e} + \frac{\langle x_h^2 + y_h^2\rangle}{m_h}\right)B^2,$$

as we address quantum structures in the strong confinement regime, in which the quantization energy is considerably larger than the electron-hole interaction energy. Here we assume that the magnetic field points along the z direction.[19]

A. Diamagnetic shift

First, we have examined the diamagnetic shift of emission lines from tall and flat dots to test whether they are consistent with their sizes. The expectations of quadratic field dependencies are fulfilled in all cases in the range of low B fields (<10 T). For the flat dot emitting at higher energy, the quadratic field dispersion of Δ_{diamag} even extends towards highest magnetic fields, as a corresponding fit to the data shows (the solid lines in Fig. 5). Except for this case, deviations from such a simple behavior are observed in the highest magnetic fields. The diamagnetic shift of the low-energy emission from flat QDs and the emission from tall QDs can no longer be described by a pure B^2 form, but a satisfactory fit can be obtained only by inclusion of a contribution that goes linearly with magnetic field. To discuss those differences, it must be kept in mind that the expression for Δ_{diamag} has been derived by assuming that the magnetic field-induced confinement is clearly weaker than the geometric one, so that the magnetic-field effects can be treated by perturbation theory.[20] The confinement strengths can be characterized by two energy scales: The energy splitting between the confined QD shells is a measure for the lateral geometric confinement and the magnetic confinement is given by the cyclotron energy. For the flat dots under study, the splitting between the p- and s-shell emission is about 50 meV for the low-energy QD, while it is ~70 meV for the high-energy QDs, in accord with their different dot diameters.[21] These values have been taken from high-excitation state-filling spectroscopy. For the tall dots, the splitting found from the difference between the p- and the s-shell emission is found to be about 25 meV. These energies have to be compared to the cyclotron energy $\hbar\varpi_c = eB/\mu$, with μ the reduced electron-hole effective mass. Assuming $\mu = 0.06m_0$, estimated from the orbital Zeeman splitting of the p-shell emission from single tall dots, the corresponding cyclotron energy exceeds 50 meV around 25 T. This demonstrates that the perturbation theory can no longer be used in this field range for the low-energy-emitting flat dots, as the magnetic confinement becomes dominant. The effect is even more pronounced in the case of tall dots, when the magnetic confinement energy equals the localization potential around 15 T. This will naturally lead to linear field contributions in the energy dispersion, because the level structure approaches Landau-level-like behavior. In contrast, for the high-energy flat dots, a pure quadratic field dependence still provides a reasonable description of the data.

Comparing the data for the different dots, we find that the diamagnetic shift for the tall dots is ~10 meV at 28 T, is much larger than the shifts for the flat dots, as expected from their different sizes. We also expect a larger diamagnetic shift for the low-energy flat dot than for the high-energy flat dot, in good accord with experiment (5 and 4 meV at the highest fields, respectively).

B. Zeeman splitting

Let us now examine the exciton spin-splitting as a function of magnetic field. In the case of flat dots, only small deviations from the expected linear field dependence of Δ_{spin} can be observed above 20 T. The linear Zeeman splitting observed in flat dots is in contrast to the results obtained from tall dots, as in the latter case virtually no splitting can be observed in low magnetic field. In high fields, the emission lines split linearly, with the high-field effective g^* factor being considerably smaller than the g^* factor of excitons in flat dots.

From $\mathbf{k} \cdot \mathbf{p}$ theory, it has been established that the g factors of carriers are determined by band-mixing effects. Band mixing can be changed by a magnetic field, as has been reported for structures of higher dimensionality such as quantum wells or bulk. For these systems, nonlinearities of the field dependence of the spin-splitting from heavy-hole-light-hole mixing induced by B are well known.[22] This mixing may therefore also be the origin of spin-splittings nonlinearity in B for QDs.

In Faraday configuration (as used here), nonlinearities of the QD exciton Zeeman splitting can be observed in low magnetic fields, which are related to the exchange electron-hole interaction. At zero field, the bright excitons with momenta $|M|=1$ show a splitting which arises from the in-plane QD asymmetry, often described by a Hamiltonian derived by the methods of invariants. The zero-field splitting arises from mixing of the two bright exciton states by anisotropic exchange, due to which their emission is also linearly polarized. This can only occur in the case of mixing of heavy with light hole states.[23,24]

We have performed polarization-resolved measurements of single dot structures in magnetic fields up to 8 T in Faraday configuration, using an optical cryostat. Within the experimental accuracy, the emission of the dots is circularly polarized for field strengths above 2–3 T, but for lower fields it contains both polarization components due to the bright exciton mixing. This is a clear hint for the importance of light-hole states already without external field[17] and is supported by two further observations: (a) Between the two bright excitons weak lines are observed in the spectra of Fig. 4 which may be associated with dark excitons with $|M|=2$. In that case, their appearance in Faraday configuration (which for pure heavy-hole states does not allow for a mixing of bright and dark excitons) also supports the influence of light-hole states. Further, the intensities of the two Zeeman split emission lines are different in the PL spectra in Fig. 2 with the high-energy line being more prominent than the low-energy line. This can also be explained by mixing of light holes to the QD valence band ground state.[17,23]

Another technique for band mixing is polarization-resolved absorption spectroscopy in Voigt configuration. Such measurements have been performed on ensembles (as mentioned above),[6] for which inhomogeneous broadening limits the accuracy. However, obviously measurements of that type on *single* quantum dots which are isolated by mesa preparation, as in our case, are technically complicated. However, we have recently studied ensembles of tall InAs/GaAs QDs by pump-and-probe Faraday rotation with the magnetic field applied in the QD plane. From these measurements, the in-plane hole g factor can be determined with high accuracy. For a pure heavy-hole state, we would expect a hole g factor very close to zero. However, in the studies a considerable magnitude of 0.15 was found for the hole g factor, which can also be explained with the importance of light-hole states only.[25]

If the band mixing at zero field is not changed by the magnetic field, one obtains a B linear spin-splitting, as is apparently the case for the flat dots. If the band mixing is modified by the magnetic field, the splitting would show a nonlinear dependence on B. Thus heavy-hole-light-hole mixing therefore might indeed offer an explanation for the observed field dispersions of the spin-splitting.

This is tentatively supported by recent model calculations, which have shown that the g factors in III–V QDs may have a rich structure resulting from strain, geometry, and confinement and further may be modified by the In/As intermixing due to the thermal procedure.[26,27] Moreover, a possible effect of the nonzero orbital momenta of light holes together with their stronger mixture with the heavy holes, which takes place in tall dots, has also been proposed to account for the low excitonic g^* factor in low magnetic fields.[28] For higher fields the ground-state exciton in tall dots obtains considerable light-hole admixtures by which the spin-splitting then becomes resolvable, but still is much smaller than for the flat QDs.[29] The proper treatment of the effect demands realistic theoretical calculations, which is beyond the scope of this experimental paper.

The difference in the band structure of the two presented classes of QDs has to be related with the In flush done at different thickness of the GaAs protection layer. In general, two factors affect the heavy-hole-light-hole splitting in low-dimensional structures: confinement and strain. Confinement opens an energy gap and shifts the light hole above the heavy hole. This energy gap decreases with the dot height. In fact, it has been observed earlier[29] that the ground-state emission from In-flushed InAs QDs redshifts with increasing dot height up to 8.5 nm. Surprisingly, the emission from dots In flushed at 11 nm occurs at higher energy than the emission from dots In flushed at 8.5 nm. In our opinion, it is the compressive strain along the growth axis that shifts the light-hole band towards the heavy-hole band, resulting in their mixing in high magnetic field. Further, the strain in the tall dots most likely is considerably reduced by the thermal annealing leading to a diffusion-induced reduction of the In content in the QDs.

We have argued that the valence-band ground state in quantum structures with comparatively weak confinement gains light-hole admixtures in magnetic field. However, by proper tailoring of the quantum confinement, valence-band mixing can be reduced to an extent that it appears to be no longer relevant for coherent manipulation of pure angular momentum states. On the other hand, increase of the confinement might lead to other problems an example of which is exciton dephasing through acoustic phonon assisted transitions, leading to a broad background in the dot spectrum.[30]

V. CONCLUSIONS

In summary, we have performed spectroscopic measurements on single InAs/GaAs QDs in high magnetic field. We have found an intriguing dependence of the Zeeman spin-splitting of the excitons in rather large sized dots, which was in contrast to almost linear field dependence of the splitting found in smaller dots. We propose to explain the nonlinear dependence by the effect of heavy-hole-light-hole mixing in high magnetic fields. Diamagnetic shift of the excitonic emission lines has also been examined. It has been found that the larger is a dot the more important is a linear component in the excitonic energy dispersion.

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[19] In many works on confined geometries, the valence-band ground state is treated as pure heavy-hole state, for which an effective spin formalism with $S=1/2$ is used, which neglects the orbital angular momentum. The exciton g factor is then given by $g_e + g^{*}_{h}$ with $g^{*}_{h}=3g_h$ in this model.

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[21] As the two types of flat dots have nominally the same material compositions, we assume the same carrier masses for them. The difference in their emission energies has to be attributed then to different lateral dot sizes with the size of the high-energy-emitting dots being smaller, leading to larger quantization energies.

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