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Exciton *g* factor of type-II InP/GaAs single quantum dots

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We investigated the magneto-optical properties of type-II InP/GaAs quantum dots using single-dot spectroscopy. The emission energy from individual dots presents a quadratic diamagnetic shift and a linear Zeeman splitting as a function of magnetic fields up to 10 *T*, as previously observed for type-I systems. We analyzed the in-plane localization of the carriers using the diamagnetic shift results. The values for the exciton *g* factor obtained for a large number of a InP/GaAs dots are mainly constant, independent of the emission energy, and therefore, of the quantum dot dimensions. The result is attributed to the weak confinement of the holes in type-II InP/GaAs quantum dots.

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During the last decade, there has been a strong interest in understanding and exploiting the zero-dimensional properties of quantum dots (QDs). Recently, the spin manipulation in those systems has also become a promising field for spintronics applications. Single-dot spectroscopy is a powerful tool to investigate the magneto properties of such systems, providing information on both the *g* factor and the diamagnetic shift. One of the points of interest is the dependence of the *g* factor on the transition energy of the dot. The *g* factors measured from single self-assembled QDs (SAQDs), such as InAs/GaAs,[1] InGaAs/GaAs,[2] and InP/InGaP,[3] do not show a clear correlation with the QD transition energy. In contrast, Kotlyar *et al.*[4] showed a definitive dependence of the *g* factor of single InGaAs/GaAs QDs, obtained by lithography and chemical etching, with the lateral size of the dots and therefore with their emission energy.

We investigated the magneto-optical properties of InP/GaAs SAQDs. This system presents a type-II band lineup in such a way that only electrons are confined in the InP dots. The spatial separation between electrons and holes leads to a smaller exciton binding energy as compared with type-I QDs. The effect of this special configuration on the magneto-optical properties of the SAQDs is a mostly unexplored question. We present here the results of the analysis of the diamagnetic shift and the Zeeman splitting of a series of dots. A remarkable result is that the InP/GaAs QDs present an essentially constant *g* factor, independent of the emission energy, and consequently, of their size.

The InP/GaAs SAQDs were grown by chemical beam epitaxy on a GaAs(001) substrate at 500 °C and capped with a 50 nm GaAs layer. Atomic force microscopy (AFM) measurements were performed in an uncapped sample grown in identical growth conditions. Microphotoluminescence (μ-PL) measurements were performed at $T=6$ K in a magneto-cryostat using an objective lens to focalize the Ar^+ laser beam (514.5 nm line). The luminescence was analyzed using a 0.75 m triple monochromator and a charge-coupled device (CCD) camera (resolution of ~ 200 μeV). A magnetic field *B* was applied along the sample growth direction. The emission intensity of the right (I_+) and left (I_-) components of the circularly polarized light were selected with appropriate optics.

The investigated structure has an ensemble of QDs with a rather large dispersion of height (H) and radius (R), with mean values of $\langle H\rangle \sim 2.1 \pm 1.0$ nm and $\langle R\rangle \sim 26 \pm 5$ nm. A plot of *H* vs *R*, obtained by using AFM measurements, is shown in Fig. 1(a).

Figure 1(b) shows the PL spectrum obtained for our InP/GaAs structure. The band centered at 1.46 eV is attributed to the wetting layer (WL) emission while the emission of the QDs corresponds to the low energy shoulder (see the dotted lines obtained by fitting the PL emission using two Gaussian bands). Figure 2(a) shows a μ-PL spectrum measured for the same structure. It presents clearly resolved sharp emission lines, which are attributed to the recombination of excitons from individual dots. The peaks have a line width of ~ 200 μeV, which is of the order of our experimen-

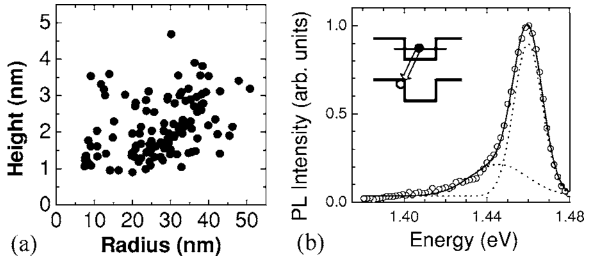

FIG. 1. (a) Height vs radius of the InP/GaAs SAQDs obtained from the AFM image in 1×1 $\mu\mathrm{m}^2$. (b) PL spectrum measured at 6 K. The solid line corresponds to the fitting using two Gaussian bands (dotted lines) and the inset shows a schematic diagram of the type II QD or QW potential profile along the growth direction.

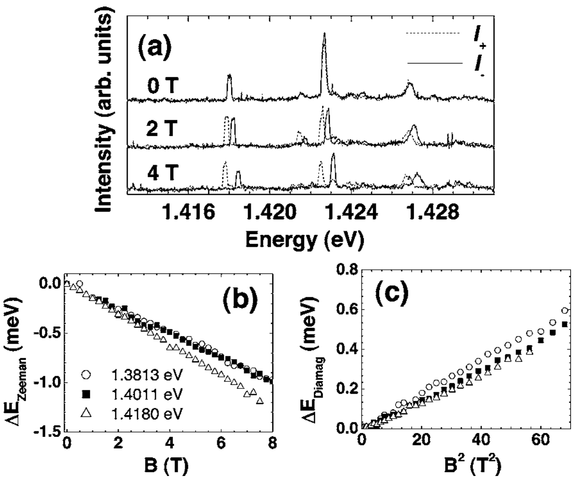

FIG. 2. (a) μ-PL spectra of type-II InP/GaAs SAQDs for different magnetic fields applied along the growth direction, (b) Zeeman splitting energy vs B, and (c) Diamagnetic shift energy vs B^2 obtained for three single QDs.

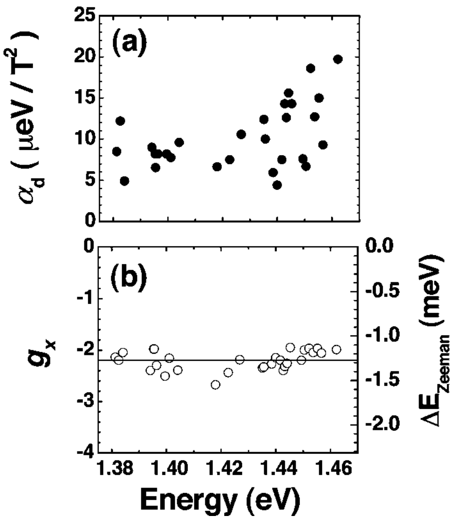

FIG. 3. (a) Diamagnetic coefficient α_d and (b) exciton g factor g_x (left) and Zeeman splitting for $B=10$ T (right) vs emission energy for a series of single QDs.

tal resolution. The μ-PL spectra were measured using a low excitation intensity in order to avoid emission from complexes of excitons, such as trions and multiexcitons.

Figure 2(a) also shows how the μ-PL spectra from the single SAQDs evolve when an external magnetic field B is applied along the growth direction. With increasing B, the emission lines shift and split in two lines that are well described by the simple model given by the equation

$$E_{\pm} = E_0 \pm \tfrac{1}{2} g_x \mu_B B + \alpha_d B^2, \tag{1}$$

where E_0 is the transition energy for $B=0$; the $\pm$ signal corresponds to the optically active excitons with momenta ± 1; μ_B, the Bohr magneton; α_d, the diamagnetic coefficient; and g_x, the exciton g factor. The experimental results for the Zeeman splitting, ΔE_{Zeeman} (second term of the equation), and the diamagnetic shift, ΔE_{diamag} (third term of the equation), for three distinct emission lines are presented in Figs. 2(b) and 2(c), respectively. In agreement with Eq. (1), ΔE_{Zeeman} presents a clear linear behavior and ΔE_{diamag} a quadratic behavior with B.

A previous report[5] on the magneto-exciton emission from a large ensemble of type-II InP/GaAs SAQDs grown by metal-organic chemical vapor deposition, under rather different conditions, has observed an oscillation of the emission band energy *versus* B that was attributed to the effect of a ringlike hole confinement around the type-II QD.[6,7] In contrast, we have not observed any signal of periodic oscillations in the emission of single QDs with B. The ringlike confinement of the hole around the InP QD is, however, a very particular condition that must be extremely sensitive to the QD shape and dimensions,[6] and therefore on the sample details.

The values of the diamagnetic coefficient, α_d, obtained for a large number of QDs ($\sim$30) are presented in Fig. 3(a) as a function of the QD emission energy. The results do not show an apparent correlation between α_d and the emission energy. We observe, however, that the dispersion of α_d increases, embracing larger α_d as the energy emission increases. Previous reports on type-I InP/InGaP SAQDs,[3,8] have not observed a clear correlation of α_d with the QD emission energy either, but only a weak tendency that the diamagnetic shift gets smaller as the emission energy increases. This tendency was observed in a large set of data, including dots from different samples and presenting a relatively large dispersion of values. The values of α_d obtained from our dots range from 5 to 20 μeV/T^2, as compared to a range from 2 to 8 μeV/T^2 observed by Sugisaki *et al.*[8] (for InP/InGaP QDs with $R\sim 15$ nm) and 40 μeV/T^2 for bulk InP.[9]

A detailed analysis[10] for type-I QDs has shown that in the limit of strong confinement and, therefore, weak electron-hole spatial correlation, the diamagnetic coefficient of a QD exciton reflects the mean square of the in-plane electron and hole wavefunction extensions, $\langle \rho_e^2 \rangle$ and $\langle \rho_h^2 \rangle$, respectively. On the other hand, in the limit of weak confinement and thus, strongly correlated electron-hole pairs, the diamagnetic shift reflects the mean square of the in-plane electron-hole separation ($\langle \rho_{eh}^2 \rangle$). Considering that this analysis is valid for type-II QDs, the large dispersion of our QD radius, which ranges from slightly smaller to much larger than the Bohr radius, leads to a condition that varies from weak ($R < r_B$) to strong correlation ($R \gg r_B$) between electrons and holes.[11] We hence base our analysis of the diamagnetic coefficient on those two extreme limits.

For $R < r_B$, the diamagnetic coefficient can be expressed as[10]

$$\alpha_d = \frac{e^2}{8}\left(\frac{\langle \rho_e^2 \rangle}{m_e} + \frac{\langle \rho_h^2 \rangle}{m_h} \right), \tag{2}$$

where e is the electronic charge and $m_{e(h)}$ is the effective mass of the electron (heavy hole). Calculations for the electronic structure of our QDs showed that for $R < r_B$, the in-plane extensions of the ground state wave functions are typically related to the QD radius by $\sqrt{\langle \rho_e^2 \rangle} \sim R/2$ and $\sqrt{\langle \rho_h^2 \rangle} \sim R$. Using those approximations, we can estimate α_d for the smallest QD radius observed by using AFM ($R \sim 7$ nm), and we obtain $\alpha_d \sim 5.8$ μeV/T^2. This is in good agreement with the smallest values of α_d obtained from our magneto-optical results.

For large QDs, the strong confinement regime is no longer appropriate.[10] We thus analyze the opposite limit of weak confinement and strong electron-hole correlation ($R \gg r_B$). In this limit, α_d can be expressed by[10]

$$\alpha_d = \frac{e^2}{8\mu} \langle \rho_{e\text{-}h}^2 \rangle, \tag{3}$$

where μ is the reduced effective mass. For the largest measured $\alpha_d \sim 20$ μeV/T^2, we obtain $\sqrt{\langle \rho_{e\text{-}h}^2 \rangle} = 7.8$ nm, which is a little smaller than the InP bulk excitonic Bohr radius (~ 11 nm). For type-I structures the confinement along the growth direction leads to a decrease of $\sqrt{\rho_{e\text{-}h}^2}$ due to the enhanced electron-hole Coulomb interaction as compared to the bulk,[12] in each case the parameter should correspond to the Bohr radius. This point has not been addressed for type-II QWs. We speculate, however, that this effect should be weaker for type-II QWs due to the significant reduction of excitonic effects on those structures as compared to type-I systems. This may explain the relatively large values of α_d observed for some of our QDs as compared to the typical values of α_d smaller than 8 μeV/T^2 reported on previous works for type-I InP/InGaP QDs.[3,8]

In general view, the QD emission energy is mainly determined by H, whereas α_d is basically related to R. Therefore, the large dispersion of α_d can be attributed to the fact that H and R are rather uncorrelated in our ensemble of QDs. For instance, the AFM results show QDs with H varying from ~ 1 to ~ 4 nm to the whole range of R [see Fig. 1(a)]. The intriguing result is the lack of points with large α_d (> 12 μeV/T^2) and small emission energies in Fig. 3(a). We may reason that, these parameters should correspond to QDs with large H (small emission energy) and large R (large α_d). For this condition, it is likely that QDs become uncoherent, creating defects and thus nonradiative recombination centers.

The g-factor values obtained from the same set of single QDs, for which the diamagnetic shift was analyzed, are presented in Fig. 3(b). The remarkable result is that, in contrast to the α_d, the g factor is practically constant with an average value of -2.2 (horizontal line in the Fig. 3(b)) and a dispersion of only of ±10%. A similar dispersion was previously reported for the g factor of In$_{0.6}$Ga$_{0.4}$As/GaAs SAQDs, but in a much smaller energy range.[2] Most of the reported results on the g factor from single QDs show markedly large dispersions[1,3,8,13] and only few works show a certain correlation between this parameter and the QD emission energy, using controlled lateral size of the dot[4] or systematic composition variation.[13] There are no reported results on the g factor from InP/GaAs SAQDs, but we may compare our results to a system with some similarities, which is the case of InP/InGaP systems. The mean value of the exciton g factor observed for InP/InGaP SAQDs is slightly smaller, $|g_X| \sim 1.2$,[3,14] and presents a much larger dispersion: ±30%. We remark that the excitonic g factor for bulk InP, $g_X \sim -2.1$, and for bulk GaAs, $g_X \sim -1.9$,[15] are both very close to the values obtained for our dots.

Analyzing the relation between exciton g factor and α_d obtained for a given QD, we observe that the relatively large dispersion of α_d, which would imply a variation of the in-plane exciton localization, does not result in a variation of the g factor, which remains practically constant. This result suggests a weak contribution of the lateral confinement on the g factor. The calculation of the g factor for a type-II QD is a rather complex problem involving the electron-hole attraction and the 3D potential and no results have been reported so far concerning this problem. Therefore, as a first approximation we use a simple model based on a type-II QW to analyze the role of the confinement effect along the growth direction on the g factor of our QDs.

The calculation is based on a Weiler-Kane $k \cdot p$ Hamiltonian model[16] for six bands including the electron-hole Coulomb interaction treated separately by a variational method. We calculated the Zeeman splitting for QWs with thicknesses (L_z) ranging from 2.8 to 4.8 nm, which are equivalent to QDs with H varying from 1.6 to 3.6 nm (considering that they are on top of the 1.3-nm-thick InP WL). The calculated results show a slightly quadratic behavior for the excitonic Zeeman splitting versus B, which is an expected result for QWs.[4] We thus compared the calculated values of ΔE_{Zeeman} for $B = 10$ T with the experimental data instead of the g factor. We obtained ΔE_{Zeeman} ranging from -1.18 meV to -1.27 meV for L_z going from 2.8 to 4.8 nm, in a fairly good agreement with the experimental data [see the right scale of the Fig. 3(b)]. The variation of the calculated values is also comparable with the experimental dispersion. This result suggests that the confinement along the growth direction does not play a significant role on the Zeeman splitting in type-II InP/GaAs systems.

Usually the valence band mixing is the dominant effect on the exciton g factor, since it is very sensitive to confinement variations.[4,13] For type-I QDs the hole is strongly confined in the dot and small variations of the dot dimensions imply large changes of the g factor, as observed in previous reports.[1,3,4,8,13] The mostly constant exciton g factor observed in our QDs for a large range emission energies is, thus attributed to the weak dependence of the hole potential profile on the QD dimensions, since for our type-II QDs, the holes are not confined at the InP layer and are, instead, only localized at the GaAs layer due solely to the Coulombic attraction.

In summary, we investigated the magneto-exciton of type-II single InP/GaAs self-assembled quantum dots. The results of the diamagnetic shift show a large dispersion of diamagnetic coefficients with no clear correlation with the emission energy. The relatively large values obtained for the diamagnetic coefficient as compared to type-I InP/InGaP quantum dots, suggests that the reduction of the electron-hole interaction may result in an increase of the in-plane exciton radius. We observed that the excitonic g factor is mainly constant, independent of quantum dot size and its emission energy. This result is attributed to the weak spatial confinement of the hole in type-II InP/GaAs systems. The *g*-factor invariance for a large dispersion of dimensions for those type-II quantum dots gives important information that may be actually used as an advantage for spintronic applications.

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