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Diamagnetic shift as a measure of the penetration of a quasi-two-dimensional exciton into quantum-well barriers

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The diamagnetic shift of the exciton recombination in $In_xGa_{1-x}As/GaAs$ quantum wells goes through a minimum for a well thickness of 40 Å and increases for a thickness of 20 Å. We develop a theory of the exciton diamagnetic shift for intermediate and high fields (5–12 T) that takes into account the three-dimensional electron-hole Coulomb interaction. The theory shows a sensitivity of the diamagnetic shift to the quasi-two-dimensional exciton thickness and describes the experimentally found minimum in the diamagnetic shift. The exciton binding energy as a function of quantum-well thickness is also obtained. A correction term to the average distance of electrons and holes, which takes into account the underestimated theoretical penetration of electron and hole wave functions into the barrier, produces a close agreement with the experimental data.

The shift of free or bound exciton recombination lines in a static magnetic field (diamagnetic shift) can be used to determine details of the dispersion relation $E(k)$ of the conduction and the valence bands. In contrast to cyclotron resonance experiments (in the presence of carriers) or to interband magnetoabsorption, it is an indirect technique that requires knowledge of the physical origin of the luminescence and a theory describing recombination in an applied magnetic field. For three-dimensional excitons, i.e., in bulk semiconductors, this problem has been solved both experimentally and theoretically. However, for the case of two-dimensional excitons in a quantum well there is still no consensus about either experimental or theoretical results.

In general, the perturbation caused by a strong magnetic field should allow the study of the dimensionality of weakly bound electron-hole (exciton) states. For a pure two-dimensional (2D) exciton in an infinitely high potential well, when the binding energy E_B increases by a factor of 4 the diamagnetic shift decreases by a factor of 3/16 of the 3D value. Actually, the pure 2D (i.e., $4\times E_B$) limit is never reached. In narrow quantum wells with finite barriers the exciton wave function penetrates into the barriers and hence exhibits 3D-like properties. For very narrow quantum-well thicknesses the exciton binding energy should reach the 3D value of the barrier material (i.e., $Al_xGa_{1-x}As$ with GaAs quantum wells or InP with $In_xGa_{1-x}As$ quantum wells). The diamagnetic shift behaves in a very similar way. It should decrease as the quantum-well thicknesses decrease, eventually pass through a minimum, and then increase again to the 3D value.

For $In_xGa_{1-x}As$ heterostructures there are only a few papers on the diamagnetic shift of quantum-well excitons.[1,2] However, they are either limited to the low field case (<8 T) or the shift of localized excitons was observed. The theory of Nash *et al.*[2] predicts a decrease in the diamagnetic shift for the $In_xGa_{1-x}As/InP$ system even for very narrow quantum-well thicknesses. However, the experimentally observed diamagnetic shifts were even smaller than the theoretically predicted ones and were claimed to arise from localized excitons. In this investigation, done for the case of strong magnetic fields, we present results of magnetoluminescence experiments for $In_xGa_{1-x}As/GaAs$ single multiple quantum wells of thicknesses between 200 Å and 22.6 Å, in magnetic fields up to 12 T. We compare experiment and theory, and find the expected minimum in the diamagnetic shift of the quasi-2D exciton. We also obtain the dependence of the exciton binding energy on the quantum-well thickness.

The photoluminescence was excited by an argon ion laser. The sample was placed in the center of a solenoid magnet (Magnex) inside a He cryostat. The maximum magnetic field was 12 T, and the sample temperature approximately 6 K. The emitted light from the sample was dispersed by a 22 cm single monochromator (Spex). A mechanical chopper in the excitation light beam was used for lock-in detection. The emitted light was resolved with a resolution of 0.7 meV using a grating blazed at 1000 nm and was detected by a liquid N_2 cooled Ge detector.

The sample was grown by molecular-beam epitaxy at the Daimler Benz research center. It contains four undoped $In_xGa_{1-x}As$ single quantum wells, with thicknesses 22.6 Å, 45.2 Å, 90.4 Å and 200 Å, respectively, buried in GaAs barrier material. The indium concentration in the well is $x=0.1$.

The steady-state luminescence of the sample is shown in Fig. 1 for selected magnetic fields. The four transitions observed at 1.4965 eV, 1.4732 eV, 1.4463 eV, and 1.428 eV at 0 T are due to the four quantum wells with thicknesses, d, of 22.6 Å, 45.2 Å, 90.4 Å, and 200 Å, respectively. As can be seen in Fig. 1 they all show a diamagnetic shift with magnetic field B. Spin splitting was observed for the widest well at the highest magnetic field. This has been described in detail in a recent publication[3] and will not be further dis-

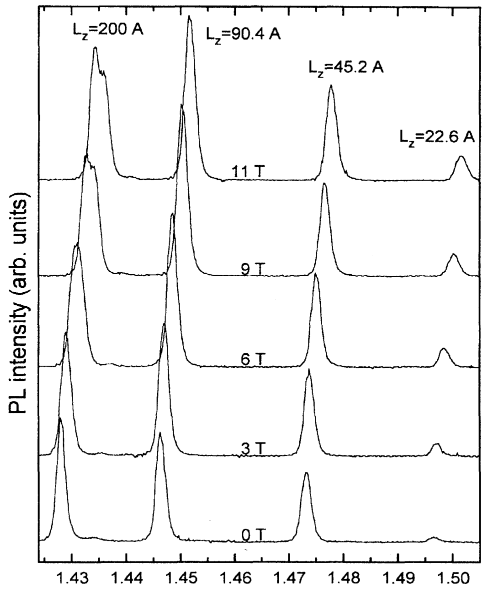

FIG. 1. Photoluminescence spectra for different magnetic fields. For clarity the specrta are arbitrarily shifted along the y axis. The single quantum-well thicknesses are labeled by L_z.

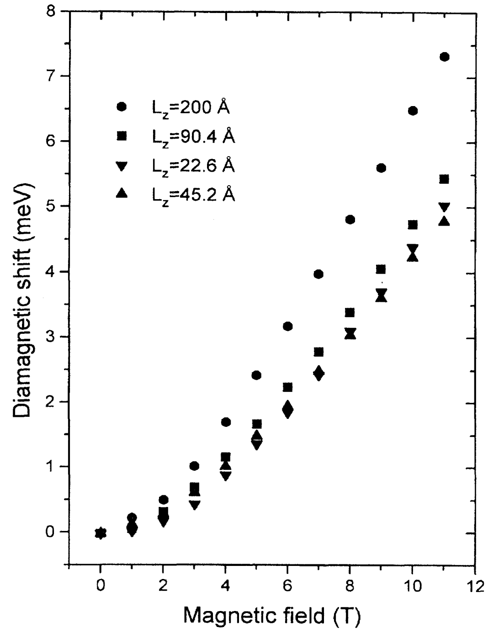

FIG. 2. Experimental diamagnetic shifts relative to the zero magnetic field transition energy for each quantum well. For the 200 Å quantum well, the value at the average position between the two peaks was choosen.

cussed here. For small magnetic fields the shift to higher energy exhibits a B^2 dependence as expected. First order perturbation theory gives the following expression for the diamagnetic shift:[2]

$$\Delta E = \tfrac{1}{2} R^* \gamma^2 w(d^*/a_B), \qquad (1)$$

where $R^* = \mu e^4/(2\hbar^2\kappa^2)$ is the effective exciton Rydberg energy, μ is the reduced effective mass ($1/\mu = 1/m_e + 1/m_h$), where m_e and m_h are the electron and hole effective masses, respectively, and κ is the static dielectric function; $\gamma = \hbar\omega_c/(2R^*)$ where $\omega_c = eB/\mu c$ is the cyclotron frequency. The function $w(d^*/a_B)$, calculated by Bugajski *et al.*,[4] describes the dependence of the diamagnetic shift on the ratio of the effective quantum-well thickness d^*, to the exciton Bohr radius $a_B = \kappa\hbar^2/\mu e^2$. For bulk 3D excitons ($d^*/a_B \gg 1$), $w = 1$. In the case of a pure 2D exciton ($d^*/a_B \ll 1$), $w = 3/16$.

Numerical calculation shows that Eq. (1) describes the diamagnetic shift of the exciton line only in weak magnetic fields, when $\gamma < 0.4$ ($\gamma = 1$ corresponds to $B = 2.8$ T). At high fields ($\gamma > 1$) the shift changes to a linear dependence on B. This can be attributed to the shift in the transition energy between the first electron and hole Landau levels (LL) ($n = 0$). The transition energy between the nth electron and hole Landau levels, neglecting the electron-hole Coulomb interaction, can be written

$$\Delta E_{\mathrm{LL}} = E_g + \hbar\omega_c(n + 1/2), \qquad (2)$$

where E_g is the forbidden gap energy.

We want to focus on the diamagnetic shift at intermediate and high magnetic fields. There is a clear trend in the diamagnetic shift as a function of quantum-well thickness (see Fig. 2). The largest shift is found for the 200 Å thick quantum well. It decreases for the wells with thickness of 90.4 Å and 45.2 Å, but it increases again for the well with thickness 22.6 Å. In order to explain this behavior, we will examine the case of a strong magnetic field ($B \geq 4$ T) where the exciton binding energy is much smaller than the cyclotron energy of the electron and hole.

In a strong magnetic field we can easily obtain the spectrum of the interband transition energies considering the electron-hole Coulomb interaction as a perturbation:

$$\hbar\omega = E_g + \hbar\omega_e(n + \tfrac{1}{2}) + \hbar\omega_h(n + \tfrac{1}{2}) - (e^2/\kappa)\,\langle 1/\rho\rangle_n + \Delta E, \qquad (3)$$

where ω_e and ω_h are the electron and hole cyclotron frequencies, respectively, $\omega_{e,h} = eB/m_{e,h}c$, $(e^2/\kappa)\langle 1/\rho\rangle_n$ is the average pure 2D electron-hole Coulomb potential in the nth Landau level which for the first ($n = 0$) Landau level can be written

$$\frac{e^2}{\kappa}\left\langle \frac{1}{\rho}\right\rangle_0 = \frac{e^2}{\kappa}\int d^2\rho\, \frac{\varphi_0^2(\rho)}{\rho} = \frac{e^2}{\kappa L}\sqrt{\frac{\pi}{2}}, \qquad (4)$$

where $L = \sqrt{c\hbar/eB}$ is the magnetic length and $\varphi_0(\rho) = (1/\sqrt{2\pi}L)\exp(-\rho^2/4L^2)$ is the 2D exciton wave function in the first Landau level. One can see that the 2D-Coulomb term obtained above increases as the square root of the magnetic field B. It does not depend on the energy band parameters or on the quantum-well thickness.

ΔE in Eq. (3) is the 3D correction (see below) to the 2D-Coulomb term. In narrow quantum wells the difference between the 3D and 2D Coulomb potential is small and its contribution to the transition energy can be considered within the framework of perturbation theory:

$$\Delta E = \frac{e^2}{\kappa}\int d^2\rho\, dz_e\, dz_h\, \varphi_0^2(\rho)\,\psi_e^2(z_e)\,\psi_h^2(z_h) \times \left(1/\rho - 1/\sqrt{\rho^2 + |z_e - z_h|^2}\right), \qquad (5)$$

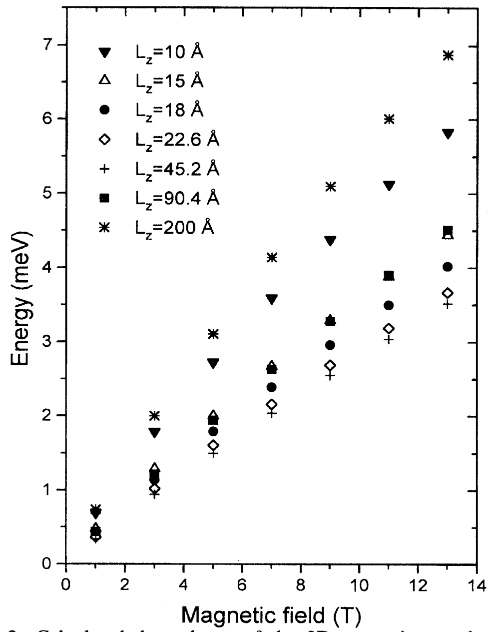

FIG. 3. Calculated dependence of the 3D correction to the pure 2D Coulomb interaction [Eq. (5)].

where $\psi_e(z_e)$ and $\psi_h(z_h)$ are the wave functions for the 1D motion of the electron and hole in the direction perpendicular to the quantum-well plane (z direction). ΔE is a measure of the average distance $\overline{|z_e-z_h|}$ between the electron and hole in the z direction, in other words the thickness of a quasi-2D exciton. For the case of $L \gg \overline{|z_e-z_h|}$ it reduces to

$$\Delta E=\frac{e^2}{\kappa L^2}\overline{|z_e-z_h|}=\frac{e^2}{\kappa L^2}\int dz_e dz_h |z_e-z_h| \psi_e^2(z_e)\psi_h^2(z_h) \ . \tag{6}$$

Thus for narrow wells, ΔE is proportional to the magnetic field and increases with the average distance between electron and hole. This gives an additional contribution to the diamagnetic shift of the transition energy. For wells with infinity high potential barriers it is equal to

$$\Delta E=(\tfrac{1}{3}-5/4\pi)\, e^2 d/\kappa L^2, \tag{7}$$

where d is the quantum-well thickness.

This result is valid if there is no penetration of the electron and hole wave functions into the barriers. However, electron and hole wave functions will penetrate into the barriers in thin quantum wells and the average distance between the particles will no longer be limited by the quantum-well thickness. As the well thickness decreases further $\overline{|z_e-z_h|}$ grows and the diamagnetic shift increases. The calculated dependence of ΔE on magnetic field for several quantum-well thicknesses, using Eq. (5), is presented in Fig. 3. One can see that the diamagnetic shift goes through a minimum for the 45 Å quantum well. It is almost a linear function on the magnetic field. In quantum wells with a thickness smaller than 45 Å, Eq. (6) is valid within an accuracy of 10% up to 8 T. The linear dependence on the magnetic field of the 3D Coulomb correction, ΔE, allows it to be interpreted effectively as an addition to the electron and hole cyclotron energy and thus can lead to misinterpretation of the electron and hole effective masses obtained from interband magneto-optic measurements.[5]

Equation (3) describes the energy of the optical transitions only in a strong magnetic field and does not give the position of the exciton line at zero field. In order to describe the experimental diamagnetic shift in a strong magnetic field we should add to Eq. (3) (as a fitting parameter) the binding energy of the exciton at zero field and find the energy band parameters that describe the dependence on magnetic field of the diamagnetic shift. For example, the result of this fitting procedure for the 45.2 Å thick quantum well is presented in Fig. 4. The effective masses we have used for electrons and holes are listed below. The electron effective mass is assumed to be unaffected by the strain ($x=0.1$) but includes nonparabolicity, i.e.,[6] $m_e^*(E)=m_e^*(0)(E_g+2E)/E_g$. We took the values: $d=200$ Å, $m_e^*=0.062\,91m_0$; $d=90.4$ Å, $m_e^*=0.064\,21m_0$; $d=45.2$ Å, $m_e^*=0.065\,69m_0$; and $d=22.6$ Å, $m_e^*=0.066\,65m_0$. For the hole masses we have to consider that the wells are under compressive strain and thus the heavy hole state is the topmost valence band state and that the in-plane effective mass values are small. We took for $d=200$ Å, $m_{\rm hh}^*=0.122m_0$, and for $d=90.4$ Å, $d=45.2$ Å, and $d=22.6$ Å, $m_{\rm hh}^*=0.14m_0$.[7–10]

In Fig. 5 we present the experimental values of the diamagnetic shift at 11 T (see Fig. 2) for several quantum wells (open squares) together with the results of the theoretical calculations (solid line). Inset (b) shows the exciton binding energy at zero magnetic field obtained as a result of the fitting procedure (closed circles). The open squares in the inset (b) are the results of analytical extrapolation of these data to the binding energy of an exciton in quantum wells with vanishing thickness, i.e., the binding energy of an exciton in GaAs. This extrapolation allows us to theoretically extend the curve into the region of very thin quantum wells.

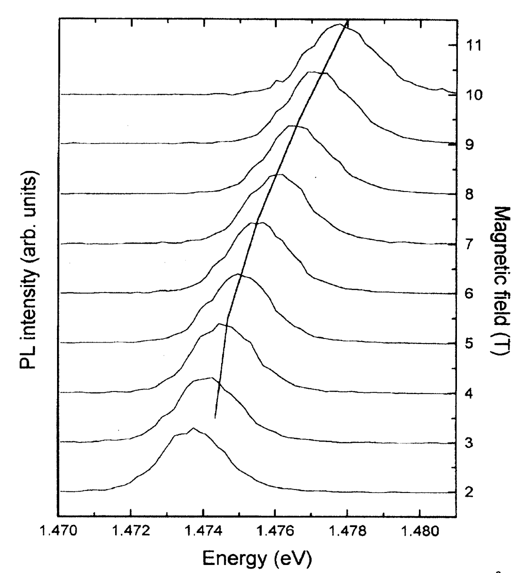

FIG. 4. Magnetophotoluminescence spectra from the 45.2 Å quantum well for different magnetic fields. The spectra are normalized and the zero intensity level at 1.47 eV is adjusted to the corresponding magnetic field on the right y axis. The calculated dependence (solid line) is accordingly also shifted in the y direction in order to compare the peak maximum with the theory.

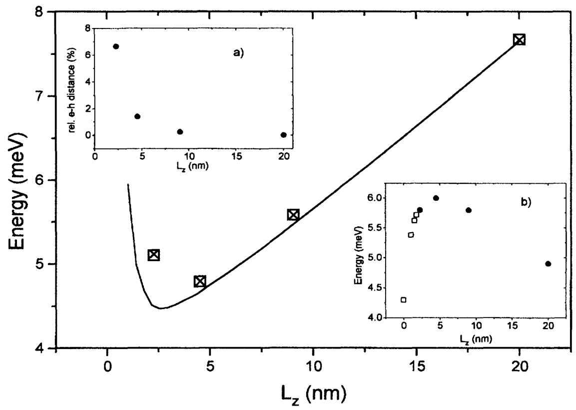

FIG. 5. Diamagnetic shift at 11 T for samples with different quantum-well thicknesses. Experimental data are shown by open squares, the solid line is the theoretical calculation, and the crosses represent fitted theoretical values that include correction to the average electron-hole distance. Inset (a) shows the relative correction to the average electron-hole distance $c_d/\overline{|z_e-z_h|}$[see Eq. (8)]. Inset (b) shows the exciton binding energy. Closed circles represent the result of the fitting procedure. Closed squares are the result of an analytical extrapolation to the binding energy of an exciton in GaAs.

One can see that the theory gives diamagnetic shifts smaller than the experiment for the thin quantum wells. From our point of view it is connected with the theoretical underestimation of the electron and hole wave function penetration into the barrier. To better describe the experimental results, we have used the following *empirical* expression for the 3D Coulomb correction in the narrow quantum wells:

$$\Delta E=(e^2/\kappa L^2)(\overline{|z_e-z_h|}+c_d) \ , \tag{8}$$

where c_d takes into account the additional penetration. This term enables us to get the very good agreement between the theory and experiment shown in Fig. 5 by crosses. The relative value of the correction $c_d/\overline{|z_e-z_h|}$ obtained by fitting Eq. (8) to the data is shown in Fig. 5 inset (a). One can see that it is almost negligible for the 90.4 Å quantum well, and only 6.5% for the 22.6 Å thick quantum well.

The exciton binding energies which we obtain for narrow quantum wells are considerably smaller than those obtained in magneto-optical experiments.[11,12] This disagreement arises from the neglect of the exciton binding energy in the transition energy between Landau levels. For Landau levels with quantum number n, the binding energy is proportional to $e^2/(\kappa L\sqrt{n})$. As a result a linear extrapolation of the transition energy to zero magnetic field does not give the correct energy gap even for Landau levels with $n=4$ and 5. The nonlinear Coulomb correction, considered within the linear magnetic field approximation, leads to an overestimation of the band edge energy which can be as large as 3 meV.

The theoretical estimation of the electron and hole penetration into the barriers is sensitive to the modeling of the boundary between the barrier and the well and to its actual shape and structure (often uncertain). We have encountered this problem already when we compared experimental data on the in-plane electron effective mass in In_xGaAs_{1-x}/InP quantum wells with theoretical calculations.[13] However, the dependence of the diamagnetic shift on the dimensionality of the exciton is even more sensitive to this effect. The additional penetration discussed above can be due to technological aspects of the well fabrication as well as to theoretical reasons. In our considerations we assumed that the quantum wells have a rectangular shape and we used the Ben Daniel–Duke boundary conditions[14] in describing the quantum-well energy levels. In narrow quantum wells, when the penetration into the barrier becomes significant, both assumptions should be examined more critically. First of all the mixing of the anions (As and P) of the interface layers leads to a softening of the barrier and to a deviation of the quantum well from a rectangular shape. Secondly, the Ben Daniel–Duke boundary conditions are obtained neglecting the difference between the Bloch functions in the quantum well and in barrier material. Both effects become more important for narrow quantum wells and lead to additional penetration into the barrier. However they are not included in our present theoretical analysis.

We have developed a theory for the diamagnetic shift dependence on quantum-well thickness as well as on magnetic fields in the regime where the exciton binding energy is much smaller than the cyclotron energy. It has been shown that the diamagnetic shift can measure the thickness of the quasi-2D exciton. We extracted the exciton binding energy by fitting the theoretical model to the experimental data. To obtain good agreement we had to assume a larger penetration of the wave functions into the wall than is given by an rectangular barrier. Two possible reasons are the smoothing of the quantum-well potential due to monolayer fluctuations and the invalidity of the Ben Daniel–Duke boundary conditions for narrow quantum wells.

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[1] M. Sugawara *et al.*, Phys. Rev. B **48**, 8848 (1993).
[2] K. J. Nash *et al.*, Phys. Rev. B **39**, 10 943 (1989).
[3] Th. Wimbauer *et al.*, Phys. Rev. B **50**, 8889 (1994).
[4] M. Bugajski *et al.*, Solid State Commun. **60**, 669 (1989).
[5] K.-S. Lee and E.-H. Lee, J. Appl. Phys. **76**, 5778 (1994).
[6] S. I. Kokhanovskii *et al.*, Fiz. Tekh. Poluprovodn. **25**, 493 (1991) [Sov. Phys. Semicond. **25**, 298 (1991)].
[7] R. J. Warburton *et al.*, Semicond. Sci. Technol. **6**, 359 (1991).
[8] M. Jaffe *et al.*, Appl. Phys. Lett. **51**, 1943 (1987).
[9] I. E. Schirber *et al.*, Appl. Phys. Lett. **46**, 187 (1985).
[10] I. J. Fritz *et al.*, Appl. Phys. Lett. **48**, 1678 (1986).
[11] M. J. L. S. Haines *et al.*, Phys. Rev. B **43**, 11 944 (1991).
[12] G. Duggan *et al.*, Surf. Sci. **228**, 310 (1990).
[13] C. Wetzel *et al.*, Phys. Rev. B **45**, 14 052 (1992).
[14] D.J. Ben Daniel and C.B. Duke, Phys. Rev. **152**, 683 (1966).