

Magneto-excitons in (411)A and (100)-oriented GaAs/AlGaAs multiple quantum well structures

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ABSTRACT

We report magneto-exciton spectroscopy studies of (411)A and (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As multi-quantum well structures. The samples consisted of seven GaAs quantum wells with widths varying between 0.6 and 12 nm, were grown on (411)A and (100)-oriented GaAs substrates. The exciton diamagnetic energy shifts and linewidths were measured between 0 and 14 T at 1.4 K. The dependence of the exciton diamagnetic shifts with magnetic field were calculated using a variational approach and good agreement with experiment for both substrate orientations was found.

Keywords: photoluminescence, magneto-excitons, interface roughness.

1. INTRODUCTION

The relationship between role of interface roughness and semiconductor device performance has been studied since the advent of quantum well structures. There have been several theoretic considerations of the contributions from interface roughness to the exciton photoluminescence (PL) linewidth, quantum well energy levels, etc.¹⁻⁴ A review article by Herman et. al.⁵ presents a summary and overview for these kinds of exciton perturbations. Recently, Hiyamizu and his colleagues have reported⁶⁻¹¹ improved exciton PL linewidths and line shapes by reducing the GaAs/AlGaAs interface roughness when growing GaAs/Al_{0.3}Ga_{0.7}As quantum wells on (411)A-oriented GaAs substrates. They have shown that the resulting low temperature PL full-width-half-maximum (FWHM) exciton linewidths are less for the (411)A structures than found for (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As quantum wells grown under similar conditions. An atomic model of a supposed surface structure of a (411)A-oriented GaAs plane is shown in Fig. 1. It consists of (100) terraces with three As-dimer rows in the [011] direction and a (111)A step with a two-monolayer height as described in Ref. 6. The pitch of the surface corrugation is 1.69 nm which is much smaller than the GaAs exciton radius of about 240 Å. Thus for (411)A-oriented interfaces, it is expected that the "roughness" due to the corrugated nature of the surface will have minimum effect on the exciton PL linewidth, line shape, and quantum well energies. Figure 2 shows a simplified cartoon for interface roughness and the corresponding exciton energies in the quantum wells. The upper part of the figure, Fig. 2a, shows monolayer fluctuation about an average number of monolayers N . Here we show three different PL peaks at energies E_{N-1} , E_N , and E_{N+1} from three quantum-well widths $N-1$, N , and $N+1$. For fluctuation amplitudes much less than the average quantum-well width N , the three PL-peak energies will be nearly the same, giving rise to an apparent increased exciton PL linewidth. On the other hand, it was anticipated by the Osaka University group⁶⁻¹¹ that the small corrugation size of the (411)A-oriented structures will represent a minimal perturbation on the quan-

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tum well energies, resulting in a single, narrow, PL exciton peak as depicted in Fig. 2b.

Perturbations by applied magnetic fields on the spectrum of quantum well excitons have been studied for some time.¹²⁻¹⁴ The purpose of this paper is to present results of studying the changes to the magneto-exciton PL spectrum and compare the results for (411)A and (100)-oriented quantum wells in the presence of large applied magnetic fields. Another important perturbation parameter is the quantum well width, where monolayer fluctuations will have large effects on the narrowest of wells and minimum consequences for quantum-well widths large compared to the fluctuation amplitude. In order to minimize the number of samples required for a large number of quantum well widths, the samples studied here have multiple quantum wells (MQW) ranging between 0.6 and 12 nm located in a single sample.

As will be shown in this paper, the previous⁶⁻¹¹ observations and conclusions about the "flatness" of the (411)A-oriented are further corroborated by our magnetic field dependent studies. Another result of our study is that the exciton PL linewidth for both the (411)A and (100)-oriented sample are nearly independent of magnetic field strength, a result which is also consistent that with a free exciton (delocalized state) transition and not due to PL from "bound" or "pinned" excitons localized by quantum-well-width induced energy fluctuations. Because of slightly different exciton binding energies, the diamagnetic shifts with magnetic field would be different each PL peak shown in Fig. 2a, leading to an apparent line broadening^{3,15} which is not observed for our samples between 0 and 14T. In order to further study PL linewidth dependencies with magnetic field for these particular samples, extremely large magnetic fields are required, where the difference in the diamagnetic shifts may be sufficiently large enough to resolve any structure to the exciton PL line shape.

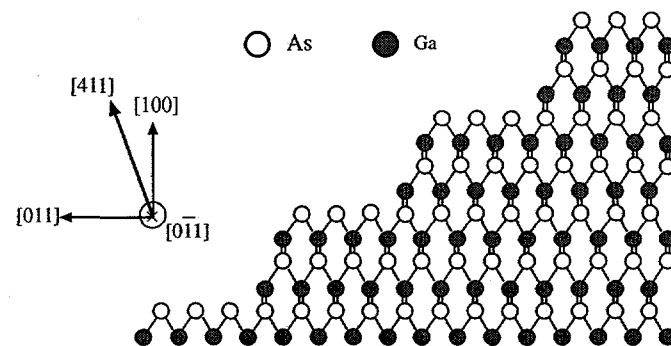


Figure 1. Schematic diagram of a supposed surface structure for a (411)A-oriented GaAs plane

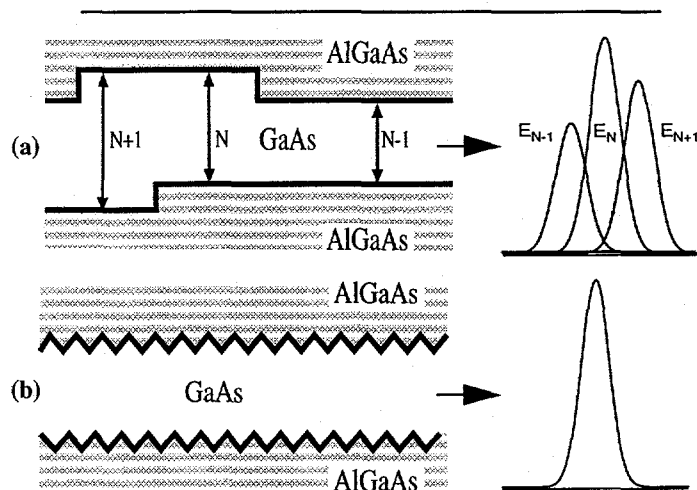


Figure 2. Schematic comparing interface roughness for (100) and (411)A-orientations. Part (a) shows a monolayer fluctuation about an average well width N and the resulting three PL peaks for a (100)-oriented quantum well. Part (b) shows the reduced fluctuation amplitude for (411)A-oriented structure and the resulting single, narrow, PL peak.

2. EXPERIMENTAL

The (411)A and (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW structures were grown using molecular beam epitaxy growth techniques and conditions for this study have been previously reported.⁶⁻¹¹ The individual GaAs quantum-well-widths varied between 0.6 and 12 nm. The width of each Al_{0.3}Ga_{0.7}As quantum well barrier was fixed at 20 nm. The narrowest GaAs quantum well (0.6 nm) was repeated twice in order to increase the PL emission intensity. A schematic representation of the structures is shown in Fig. 3. The magnetoluminescence measurements were made in the temperature range of 1.4 and 76 K, and the magnetic fields varied between 0 and 14 T. The sample was attached to the end of a 100 μm -core-diameter optical fiber. The luminescence measurements were made with an Argon-ion laser operating at 514.5 nm. The laser was injected into the

optical fiber by means of an optical beam-splitter and the returning photoluminescence signal was directed to the optical monochromator and IEEE488-based photon counting data acquisition system from the same single fiber.¹⁶ Typical laser power densities to the sample were less than 1 W/cm^2 , therefore the photo-induced two-dimensional density of electrons and holes are very small, less than $1\text{-}3 \times 10^{10} \text{ cm}^{-2}$. The photomultiplier tube used during the course of these experiments was an InGaAs-enhanced Hamamatsu R3310-02. The direction of the applied magnetic field is parallel to the growth direction, i.e., the resulting magneto-exciton orbits are in the plane of the quantum well.

3. DISCUSSION

Figures 4 and 5 show the low-temperature PL spectrum for the (411)A and (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW. These two spectra are nearly the same as previously published⁶⁻¹¹ for these with the main difference in the ratio of the peak intensities being due to the experimental configuration, optical response factors, laser power density and wavelength, etc. In both Figs. 4 and 5, the lower energy PL-peaks to the left of the 12-nm PL-peak are from the GaAs substrate and or the GaAs cap layer. As can be seen, the spectrum for the (411)A-oriented sample does not have the structure to the exciton PL energies that the (100)-oriented sample exhibits. It should be mentioned here that not all (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW samples exhibit the structure to the exciton PL spectrum shown in Fig. 5. Very good exciton PL spectra, showing no extra PL-peaks due to fluctuations of the quantum well width, have been observed by many others in the literature and also in by ourselves. However, the growth conditions for these structures have to be well understood and controlled if perfect, or nearly perfect, interfaces are to be achieved. For the purposes of this paper, the (100)-oriented sample, discussed in this paper, is used as an illustrative example showing the effect on the exciton spectrum when there are fluctuations to the quantum well width.

Figure 6 shows the PL-peak energy data as a function of the GaAs quantum-well-width for the (411)A-oriented structure at 1.4K. The solid line drawn through the data is a result of our calculation for the diamagnetic shifts in a magnetic field and is discussed below. The results for the (100)-oriented PL-peak energy versus quantum-well-width data will not be discussed because the results are similar. The increasing exciton energy with decreasing quantum-well width is well known to result from quantum confinement.

Figure 7 shows the dependence of the FWHM as a function of the quantum-well width for both samples. The open squares are for the (100)-oriented sample while the closed circles are the data for the (411)A structure. The difference between the two samples is largest for the narrower quantum wells where the effects of quantum-well width fluctuation are the most important. Similar results and conclusions have been previously reported⁶⁻¹¹ in detail. The decreasing FWHM linewidth (or rollover) for the smallest quantum-well width may be an indication that the exciton wavefunction has extended into the

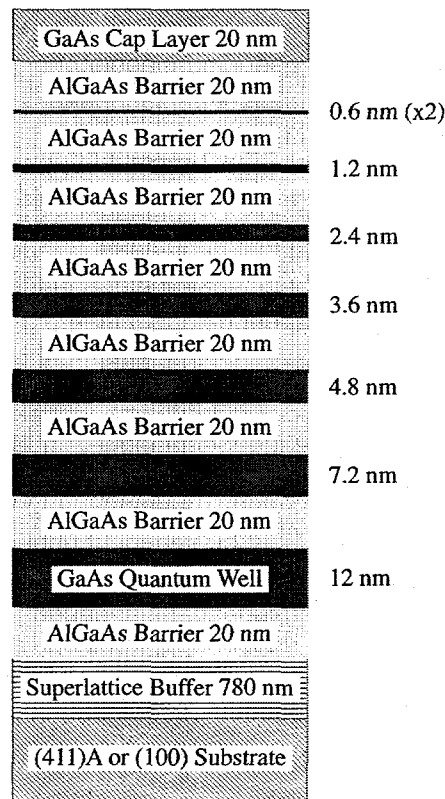


Figure 3. Schematic representation of the structure for both the (100) and (411)A GaAs/Al_{0.3}Ga_{0.7}As MQW. The well-widths for each GaAs layer are indicated in the figure.

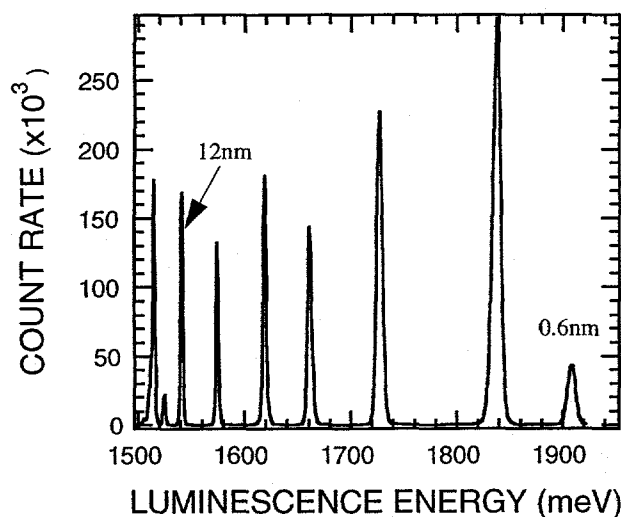


Figure 4. Low temperature (1.4K) PL spectrum for the (411)A-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW. The PL-peaks from the 12 and 0.6-nm-wide-wells are labeled. See Fig. 3 for the other the PL-peak well-widths.

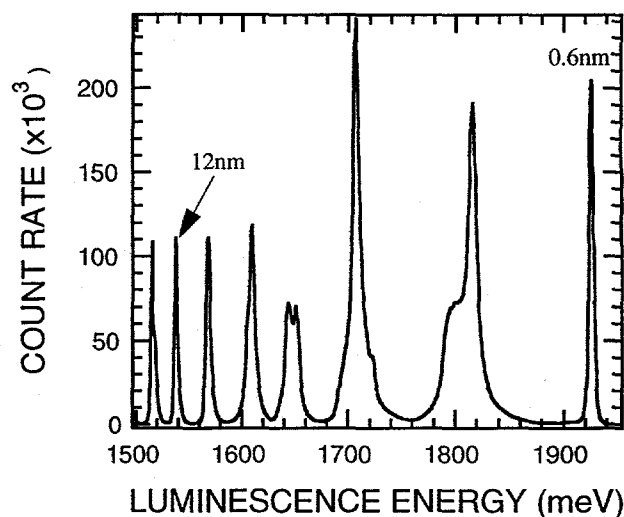


Figure 5. Low temperature (1.4K) PL spectrum for the (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW. The PL-peaks from the 12 and 0.6-nm-wide wells are labeled. See Fig. 3 for the other the PL-peak well-widths.

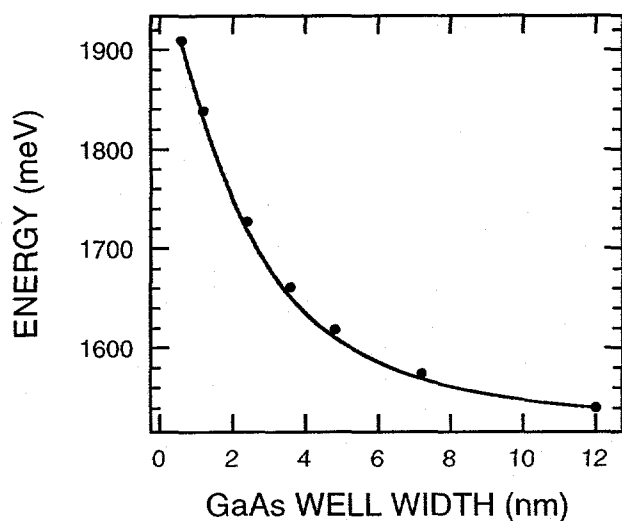


Figure 6. Quantum-well width dependence for the PL peak energy at 1.4K for the (411)A-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW. The solid line is calculated (see text for discussion.)

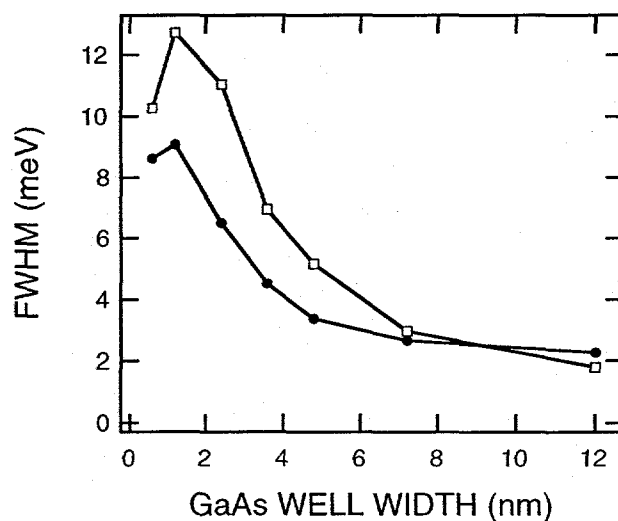


Figure 7. Quantum-well width dependence for the PL line-widths at 1.4K for the (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW (open squares) and the (411)A-oriented MQW (circles).

Al_{0.3}Ga_{0.7}As barriers, and hence transforming the 2D-exciton into a nearly 3D-exciton (sometimes referred to as quasi-2D) reducing the effects from a fluctuating quantum-well width.

The magneto-exciton diamagnetic shift dependence on magnetic field for each (411)A-oriented quantum well is shown in Fig. 8. For each quantum well, the diamagnetic shift was calculated by subtracting the zero-field exciton energy shown in Fig. 6 from the magnetic field dependent PL-peak energy. In order to calculate theoretical diamagnetic shifts, we employed the

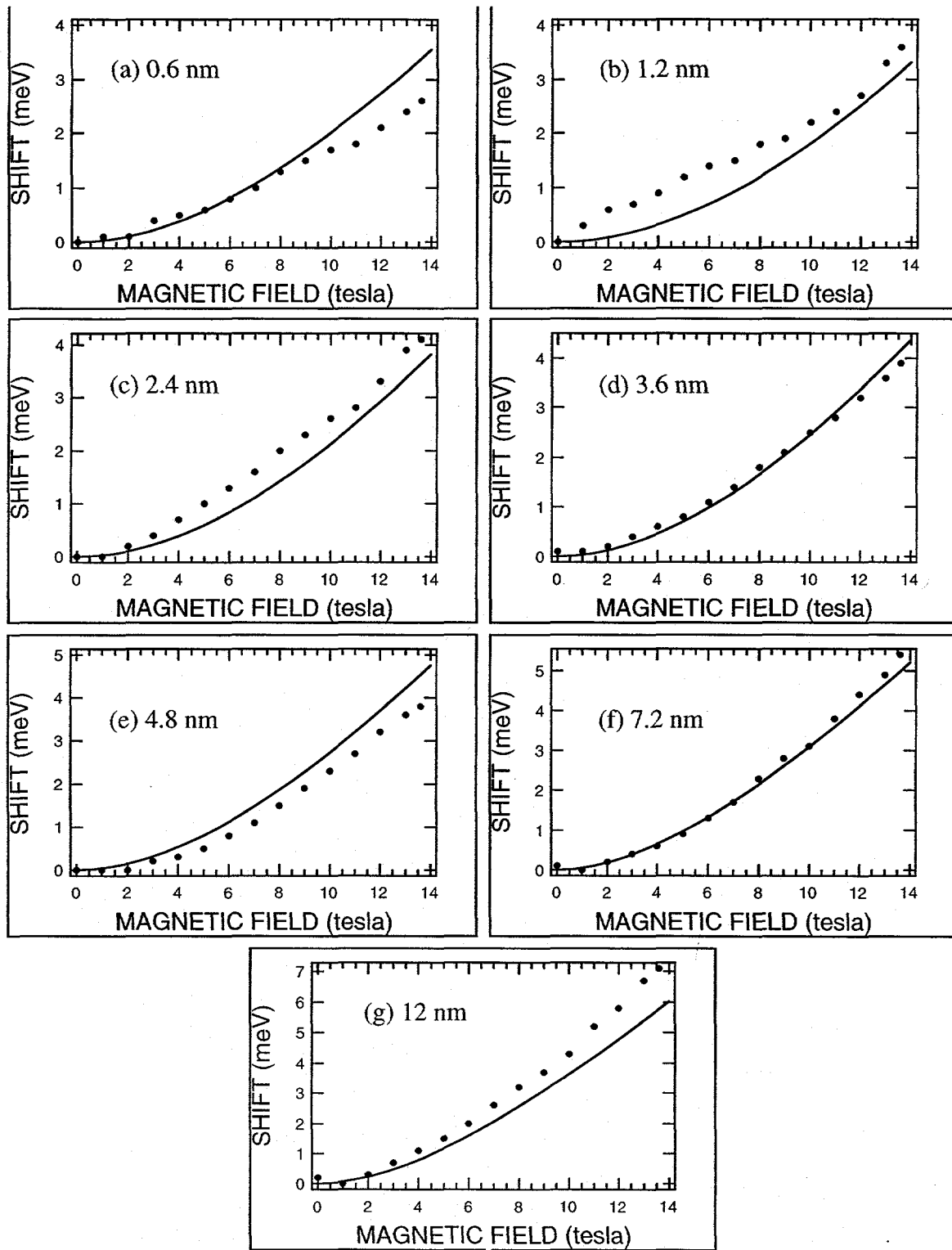


Figure 8. Magnetic field dependence of the exciton diamagnetic shifts for the (411)A-oriented GaAs/Al_{0.3}Ga_{0.7}As quantum wells at 1.4K. The GaAs well-width is indicated in each graph. The solid curve drawn through the data points is a result of a variational calculation of the quantum well exciton wavefunction and energy in an external magnetic field. One fixed set of parameters are used for all seven curves, with the only variable being the quantum-well width. See text for discussion.

variational approach as described by Greene and Bajaj.^{12,13} The trial wavefunctions are expressed in terms of a Gaussian basis set with the magnetic field perpendicular to the growth direction, i.e., the exciton orbits are in the plane of the quantum well. For each quantum well width, the exciton binding energies are calculated for finite values of the height of the GaAs-Al_{0.3}Ga_{0.7}As potential barrier. The envelope function method is also employed to account for the finite quantum-well width and height. Besides the bulk GaAs bandgap energy at 1.4 K, the magnetic field strength, and the quantum-well width, some of the relevant physical parameters include: (1) Conduction and valence-band offsets between GaAs and Al_{0.3}Ga_{0.7}As. (2) The Luttinger parameters γ_1 and γ_2 for the GaAs quantum well and Al_{0.3}Ga_{0.7}As barriers. (3) Conduction and valence-band masses for the GaAs quantum well and Al_{0.3}Ga_{0.7}As barrier. And (4), low frequency dielectric constants ϵ_0 for the quantum well and barrier materials.

Because of the relatively narrow quantum wells, the large quantum-well energy shifts shown in Fig. 6 compared to the bulk GaAs bandgap energy $E_{\text{gap}} \sim 1515$ meV at 1.4K indicates that $k \cdot p$ corrections to the conduction-band mass and thus non-parabolic mass effects must be taken into account. An excellent discussion of the conduction-band mass dependence on the bandgap energy has been presented in Refs. 17 and 18. The conduction-band mass corrections were also included in our variational approach. Furthermore, we added valence-band energy effects, and hence valence-band mass nonparabolicities, due to GaAs heavy-hole light-hole interactions (anti-crossing) away from the Γ -point in the Brillouin zone, i.e., away from $k = 0$.¹⁹ In order to test the accuracy of our calculation, and the validity of all of the aforementioned approximations, we calculated the zero-field energy of the exciton transition as a function of quantum-well width. The solid curve drawn through the data points in Fig. 6 shows the result and accuracy of our variational approach. And as can be seen in Fig. 6, the agreement between experimental exciton transition energies and theory is excellent. Similar results and conclusions are obtained for the (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As MQW sample for which the data is not presented here.

Thus with the confidence gained from comparing theory and experiment shown in Fig. 6, we now discuss the exciton diamagnetic shift dependence on magnetic field as calculated by the variational approach. The solid curves in Fig. 8 are the results of our calculation with varying magnetic field. As can be seen, the agreement with experiment is excellent for the 3.6 and 7.2-nm-wide-wells, Figs. 8d and 8f, but is only fair for the other quantum wells. However, because only one parameter, the GaAs quantum-well width, was varied and no particular attempts for optimization of the other parameters were made, the overall agreement between theory and experiment is good. The erratic behavior of the exciton diamagnetic shifts for the narrower quantum wells shown in Fig. 8 was also observed for the (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As structure. There are several possible explanations for the deviation from non-quadratic behavior of the exciton diamagnetic shift with magnetic field and three are as follows. For the narrow wells, the quantized Γ -point quantum-well energies for both the conduction and valence-bands are not only near the top of their barriers at zero-field, but approach the top of the barriers with magnetic field. The effect of pinning of this energy level to the barrier height may not be adequately treated in our variational approach. Secondly, the valence-band nonparabolicities from heavy-hole light-hole interactions are not as simple as we assumed under these top-of-the-well conditions because of energy pinning of both valence-band states. Finally, interactions between the GaAs Γ -point with the Al_{0.3}Ga_{0.7}As X-point are not considered and maybe important as the energy difference between these two energies decreases with decreasing GaAs quantum-well width. Of course, in this discussion about comparing the results of the variational calculation with experiment assumed that each quantum well was grown perfectly and that the "strange" behavior of the diamagnetic shifts with magnetic field are extrinsic in nature.

In conclusion, we have shown the magnetic field studies are useful for characterizing quantum well structure. In particular, we found that the data presented here collaborates the previous conclusions⁶⁻¹¹ about the "flatness" of the (411)A-oriented GaAs/Al_{0.3}Ga_{0.7}As heterojunctions compared to (100)-oriented GaAs/Al_{0.3}Ga_{0.7}As structures. We also discussed a theoretic-

cal model, based the variational technique, for an exciton in a magnetic field and calculated the dependence of the exciton energy with quantum-well width as well as the dependence of exciton diamagnetic shift with magnetic field. The agreement between theory and experiment for the dependence of quantum-well exciton energies on the well width was excellent. With the same parameters used for the quantum-well width comparisons, an overall good agreement with the magnetic field dependence of the diamagnetic shift was found.

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REFERENCES

1. J. Singh, K. K. Bajaj, and S. Chaudhuri, "Theory of excitonic photoluminescence linewidth in semiconductor alloys," *Appl. Phys. Lett.* **44**, pp. 1075-1077, 1984.
2. J. Christen and D. Bimberg, "Line shapes of interband and excitonic recombinations in quantum wells: Influence of final-state interaction, statistical broadening, and momentum conservation," *Phys. Rev. B* **42**, pp. 7213-7219, 1990.
3. J. Lee, G. D. Sanders, and K. K. Bajaj, "Excitonic photoluminescence line shape due to interfacial quality in quantum well structures in a magnetic field," *J. Appl. Phys.* **69**, pp. 4056-4059, 1991.
4. L. E. Golub, "Exciton localization at islands in quantum-well structures," *Phys. Solid State* **39**, pp. 1673-1676, 1997.
5. M. A. Herman, D. Bimberg, and J. J. Christen, "Heterointerfaces in quantum-wells and epitaxial-growth processes: Evaluation by luminescence techniques," *J. Appl. Phys.* **70**, pp. R1-R51, 1991.
6. S. Shimomura, A. Wakejima, A. Adachi, Y. Okamoto, N. Sano, K. Murase, and S. Hiyamizu, "Extremely flat interfaces in GaAs/AlGaAs quantum wells grown on GaAs (411)A substrates by molecular beam epitaxy," *Jpn. J. Appl. Phys.* **32**, pp. L1728 - L1731, 1993.
7. S. Shimomura, K. Shinohara, T. Kitada, S. Hiyamizu, Y. Tsuda, N. Sano, A. Adachi, and Y. Okamoto, "Extremely high uniformity of interfaces in GaAs/AlGaAs quantum wells grown on (411)A GaAs substrates by MBE," *J. Vac. Sci. Technol. B* **12**, pp. 1043-1046, 1994.
8. S. Shimomura, S. Kaneko, T. Motokawa, K. Shinohara, A. Adachi, Y. Okamoto, N. Sano, K. Murase, and S. Hiyamizu, "Extremely flat interfaces in GaAs/AlGaAs quantum wells with high Al content (0.7) grown on GaAs (411)A substrates by molecular beam epitaxy," *J. Cryst. Growth* **150**, pp. 409-414, 1995.
9. S. Shimomura, K. Shinohara, T. Kitada, S. Hiyamizu, Y. Tsuda, N. Sano, A. Adachi, and Y. Okamoto, "Much improved interfaces in GaAs/AlGaAs quantum wells grown on (411)A GaAs substrates by molecular-beam epitaxy," *J. Vac. Sci. Technol. B* **13**, pp. 696-698, 1995.
10. K. Shinohara, K. Kasahara, S. Shimomura, A. Adachi, Y. Okamoto, N. Sano, and S. Hiyamizu, "As₄ pressure dependence of the interface flatness of GaAs/Al_{0.03}Ga_{0.07}As quantum wells grown on (411)A GaAs substrates by MBE," *Appl. Surf. Sci.* **113**, pp. 73-78, 1997.
11. K. Shinohara, Y. Shimizu, S. Shimomura, Y. Okamoto, N. Sano, and S. Hiyamizu, "GaAs/AlGaAs super-flat interfaces in GaAs/AlAs and GaAs/(GaAs)₂(AlAs)₂ quantum wells grown on (411)A substrates by MBE, *Physica E* **2**, pp. 166-170, 1998.
12. R. L. Greene and K. K. Bajaj, "Effect of a magnetic field on the energy levels of a hydrogenic impurity center in GaAs/Ga_{1-x}Al_xAs quantum-well structures," *Phys. Rev. B* **31**, pp. 913 - 918, 1985.
13. R. L. Greene and K. K. Bajaj, "Binding energies of Wannier excitons in GaAs-Ga_{1-x}Al_xAs quantum-well structures in a magnetic field," *Phys. Rev. B* **31**, pp. 6498 - 6502, 1985.
14. D. C. Reynolds, K. K. Bajaj, C. W. Litton, R. L. Greene, P. W. Yu, C. K. Peng, and H. Morkoc, "Magneto-optical studies of GaAs-Al_xGa_{1-x}As multi-quantum-well structures grown by molecular-beam epitaxy," *Phys. Rev. B* **35**, pp. 4515-4518, 1987.
15. E. D. Jones, R. P. Schneider, Jr., S. M. Lee, and K. K. Bajaj, "Magnetic-field-dependent excitonic photoluminescence linewidth in In_{0.48}Ga_{0.52}P semiconductor alloys," *Phys. Rev. B* **46**, pp. 7225-7228, 1992.
16. H. Ackermann, E. D. Jones, J. E. Schirber, and D. L. Overmyer, "Apparatus for optical studies of materials at hydrostatic pressures and low temperatures in magnetic fields," *Cryogenics* **25**, pp. 496-498, 1985.
17. J. Singleton, R. J. Nicholas, D. C. Rogers, and C. T. B. Foxen, "Cyclotron resonance of electrons in a narrow GaAs/(Ga,Al)As quantum well: Polaron effects and non-parabolicity," *Surface Science* **196**, pp. 429-436, 1989.

18. F. A. P. Osório, M. H. Degani, and O. Hipólito, "Cyclotron resonance of electrons in GaAs-Ga_{1-x}Al_xAs heterojunctions," *Superlattices and Microstructures* **6**, pp. 107-110, 1989.
19. E. D. Jones, "Band structure parameters for quantum wells: Magnetoluminescence determinations," in the *Proceedings of the 191st Meeting of the Electrochemical Society: Twenty-Sixth State-of-the-Art Program on Compound Semiconductors*, Edited by D. N. Buckley, S. N. G. Chu, H. Q. Hou, R. E. Sah, J. P. Vilot, and M. J. Deen (Electrochemical Society, Pennington, NJ 1997) pp. 127-137.