Magnetophotoluminescence of quantum confined states in ordered $Ga_xIn_{1-x}P$ with 200 nanometer resolution

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We observe the diamagnetic shift and Zeeman splitting of ultra-sharp photoluminescence lines which appear in an ordered-GaInP epilayer using a micro-photoluminescence technique in fields up to 5 T. These transitions show many signatures of a fully quantum-confined state, however, a detailed description of the origin of these states is not known. Under a magnetic field, these states show behavior similar to other III-V quantum dot systems, with large variations in diamagnetic shift and exciton *g* factor. By noting their variation with energy, one can make inferences about possible confinement geometries. These results are compared to previous work.

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Spontaneous $CuPt_B$-type ordering in the ternary alloy $Ga_xIn_{1-x}P$ has been intensively studied over the past decade (for a review see Ref. 1). Upon ordering, a broad below-gap emission band appears at 30–40 meV below the band-edge excitonic peak in photoluminescence (PL) spectra. Decreasing the probed volume with microphotoluminescence techniques, ultrasharp lines showing signatures of quantum confinement have been observed in this (bulk) material.[2,3] Growth instabilities, such as spontaneous ordering and composition modulation have potential for synthesizing high density arrays of quantum confined states in thin epitaxial films. Important to both the fundamental understanding of the electronic properties of the material and any possible technological applications, is a better understanding of the origin and properties of these transitions.

We present here a microphotoluminescence study of these energetically narrow transitions in an ordered-$Ga_xIn_{1-x}P$ epilayer under varying magnetic fields. The magneto-optical behavior of these lines appear to follow well-known quantum dot models.[4,5] However, a model of the microscopic origin of these states requires a detailed knowledge of their microstructure, which is complicated by the presence of alloy fluctuations and the ordering-induced anisotropy in these materials.[6] By surveying the magneto-optical properties of these transitions, one can place constraints on their origin. We observe variations in the diamagnetic shift and Zeeman splitting both for select lines which appear within a single aperture and from one aperture to the next. By noting the variation of these parameters with energy, inferences about possible confinement geometries can be made. These observations have important implications for any theory describing the origin of these states.

The sample studied was a 10 μm thick partially-ordered $Ga_{0.52}In_{0.48}P$ epilayer (order parameter $\eta \sim 0.42$) grown on a semi-insulating GaAs substrate miscut 6° toward $[111]_B$. This sample was patterned with an array of 200-nm apertures in an otherwise opaque aluminum mask. All measurements were made at low temperatures (nominally 4.2 K) in a continuous flow cold-finger microscope cryostat equipped with a 5 T superconducting magnet.[7] The photoluminescence was dispersed by a 0.32 m single-grating spectrometer (2400 lines/mm) onto a CCD array detector. Circular polarization selectivity was achieved with a linear polarizer and a quarter-wave plate. The continuous wave excitation ($\lambda = 532$ nm) power was held constant at ~ 0.04 μW for all measurements unless otherwise stated.

As mentioned in Ref. 2, the number of sharp lines which appear within a single aperture varies widely between zero and more than ten, a broad band emission is also observed in every aperture at sufficiently high powers. Selected apertures at lower powers show well-resolved sharp PL lines with linewidths between 0.5 meV and less than 100 μeV. In previous studies of this and other similarly prepared samples, these lines typically show minimal thermal broadening and multiple energetically narrow absorption peaks,[8] both signatures of quantum confinement. In the magneto-PL measurements that will be discussed here, the diamagnetic shifts for these lines are significantly less than 15 $\mu eV/T^2$ measured for the bulk band-edge excitonic states,[6] giving further evidence of quantum confinement. However, no clear evidence for excited state emission was found in the study of Ref. 8. Rather, a model of spatially neighboring confined states coupled via tunneling was proposed to explain the randomly correlated (uncorrelated) absorption and emission spectra. This model is also supported by resonantly excited time-resolved PL measurements, which show evidence of exciton transfer between sharp-line states on timescales of ~ 500 ps.[9] In light of these facts, this study concentrates on select apertures which exhibit well-resolved energetically narrow PL lines and examines their behavior under an applied magnetic field.

Figure 1 shows the high-resolution low-temperature PL spectra collected from a single aperture at $B = 0$ (solid line) and $B = 5$ T (dashed line). The narrowest linewidths in these spectra are less than 100 μeV and approach the resolution limit of the system. The spectra at $B = 5$ T exhibits a positive diamagnetic shift and Zeeman splitting for all peaks. This is typical of all the sharp lines observed whose linewidth was less than the observed splitting, typically less than 400 μeV. This splitting is more clearly observed by selecting out the right and left circularly polarized components of the emission, as shown in Fig. 2 (designated as σ^+ and σ^- in the figure). This behavior is repeated for all such lines observed in every aperture examined, and is similar to what has been observed in III-V quantum dot systems, for instance, quan-

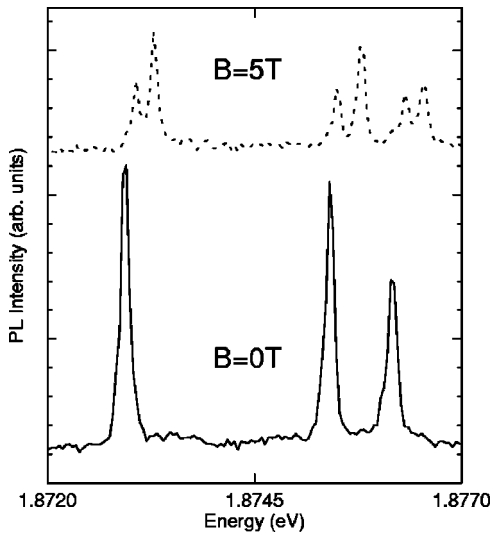

FIG. 1. Spectra at $B=0$ and 5 T of ultrasharp PL lines isolated to 200-nm aperture. 5 T spectra (dashed line, offset vertically for clarity) clearly shows Zeeman splitting and diamagnetic shift.

tum dots formed by monolayer thickness fluctuations in narrow GaAs/AlGaAs quantum wells.[10]

Such systems are often modeled by strong vertical confinement and weaker lateral confinement. Under these conditions, the magnetic field dependence of electrons or holes in a quantum dot can be cast in the form of a two-dimensional (2D) harmonic oscillator in the presence of a perpendicular magnetic field.[5,11] General treatments of semiconductor nanostructures with this symmetry show that for strongly confined heavy-hole excitons, a diamagnetic shift[12] of the form $\Delta E^X_{Dia}=(e^2/8)[\langle\rho_e^2\rangle/m_e+\langle\rho_h^2\rangle/m_h]B^2$ and Zeeman

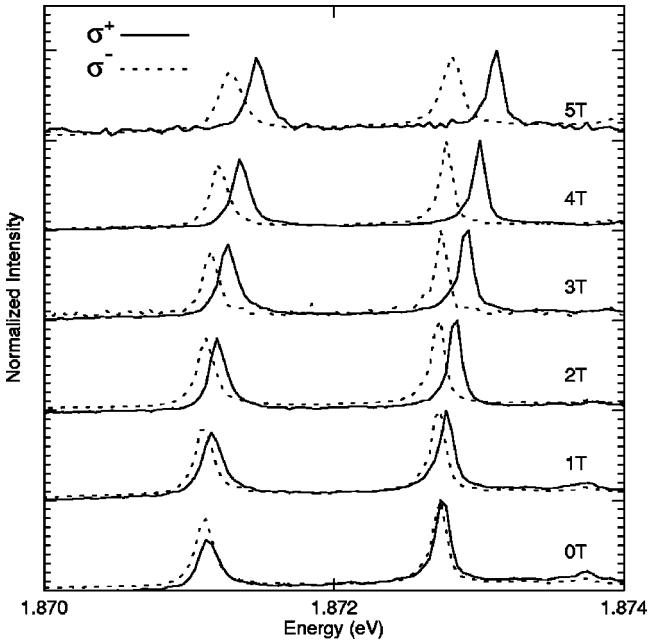

FIG. 2. Spin-polarized PL at varying magnetic field (0–5 T). Plots normalized and offset for clarity, solid line σ^+, dashed line σ^-.

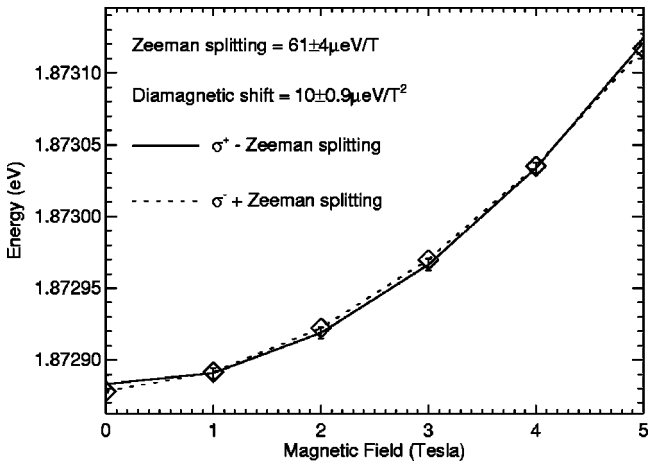

FIG. 3. Least squares fit to data for right-most peaks in Fig. 2. Diamond markers: data; solid line: σ^+ peak position minus linear Zeeman splitting; dashed line: σ^- peak position plus linear Zeeman splitting.

splitting[13] $\Delta E^X_{Zeeman}=\pm\frac{1}{2}g^*\mu_B B$ are predicted, where e, ρ_e, ρ_h, m_e, m_h are the electron charge, the radial extent, and effective mass of the electron (hole) wavefunctions, g^* is the exciton effective g factor and μ_B is the Bohr magneton. From the expression for the diamagnetic shift, one expects a decrease in the diamagnetic shift for increasing quantum confinement or decreasing dot size. According to Ref. 13, a strong increase in $|g^*|$ and hence the Zeeman splitting is also expected for decreasing dot size.

A similar microstructure has been proposed as the origin of the sharp lines studied here, namely, an isolated In-rich double layer embedded in an ordered-GaInP matrix.[3,14] This proposed microstructure may appear at the antiphase boundaries (APB's) of an ordered GaInP alloy, if the APB is aligned perpendicular to the ordering direction and is of the In-rich variety. It should be noted, however, that the orientation and composition of the APB's in ordered GaInP can vary widely. Within the context of this model, this type of APB would form an ultranarrow quantum well, oriented perpendicular to $[111]_B$. Lateral confinement would be provided by alloy fluctuations within the layer, forming so-called quantum discs. The magnetic field behavior of quantum confined states in this system has not been investigated theoretically, and therefore quantitative comparisons to theory cannot be made. However, we do not expect the effects of confinement on the magneto-optical behavior to be qualitatively different from other, well-studied III-V quantum dot systems.

The energetic peak position versus magnetic field for the doublet shown on the right-hand side of Fig. 2 is plotted in Fig. 3, fit to a quadratic diamagnetic shift combined with a linear Zeeman splitting. This fitting procedure was used for the select, sharp-line PL lines labeled a–e in Fig. 4. The derived values of the Zeeman splitting (effective g factor) and diamagnetic shift are shown in Figs. 5(a) and 5(b). Examining Fig. 5(a), an initial increase in exciton g factor is observed with increasing energy for peaks labeled a–c. As the confinement energy should increase with decreasing dot

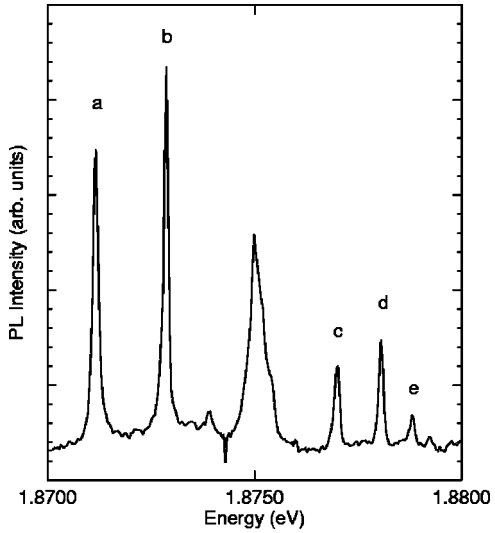

FIG. 4. Select lines $a-e$ referenced in Fig. 5 (the central peak's asymmetric broadening obfuscated an accurate fit).

size, this trend is consistent with the results of Ref. 13. At higher energies (peaks labeled $c-e$), this trend is reversed. In Fig. 5(b), a decrease in the diamagnetic shift is observed for peaks labeled $a-c$. Again, consistent with the above discussion and increasing confinement energy or decreasing dot size. However, this trend is also reversed at higher energies (peaks labeled $c-e$).

If the energy of the localized state is increased by decreasing the localization length, the behaviors of peaks $a-c$ are explained in terms of increased confinement. If the energy of the localized state is increased by raising the band-gap of the material within the confined volume (due to alloy fluctuations), the effects of confinement are decreased, reversing the trend. As the sample is only partially ordered, both effects should be observable. The fact that the Zeeman splitting and diamagnetic shifts for sharp lines with nearly the same energy but located in different apertures varied by up to a factor of two further supports this explanation. An alternate explanation is that the confinement potential is anisotropic, but oriented randomly. In that case, the observed diamagnetic shift and Zeeman splitting would depend on the orientation of the anisotropic confined volume.

The spectrum of diamagnetic shifts reported here ranges from 6 to 12 μeV/T^2, this nearly spans the range of diamagnetic shifts for $B\|(1\bar{1}1)$ and $B\perp(1\bar{1}1)$ ($4-11$ μeV/T^2) for sample *B* reported in Ref. 3. This anisotropy with respect to the ordering direction was cited as evidence to support the quantum-disc model. We report only measurements made with $B \,||\, (001)$, thus our values should fall between the values in Ref. 3. The multiplicative effect of the effective mass anisotropy,[6] which gives a ratio of roughly 1.6 for $\eta \sim 0.42$, would easily extend the range of our measurements to encompass the range reported in Ref. 3. Thus it would be necessary to measure the anisotropy for the *same transition*, in order to attribute any anisotropy due to a preferred orientation of the confinement potential. As this type of measurement has never been done, no strong evidence of any preferred orientation of the confinement potential of these states exists.

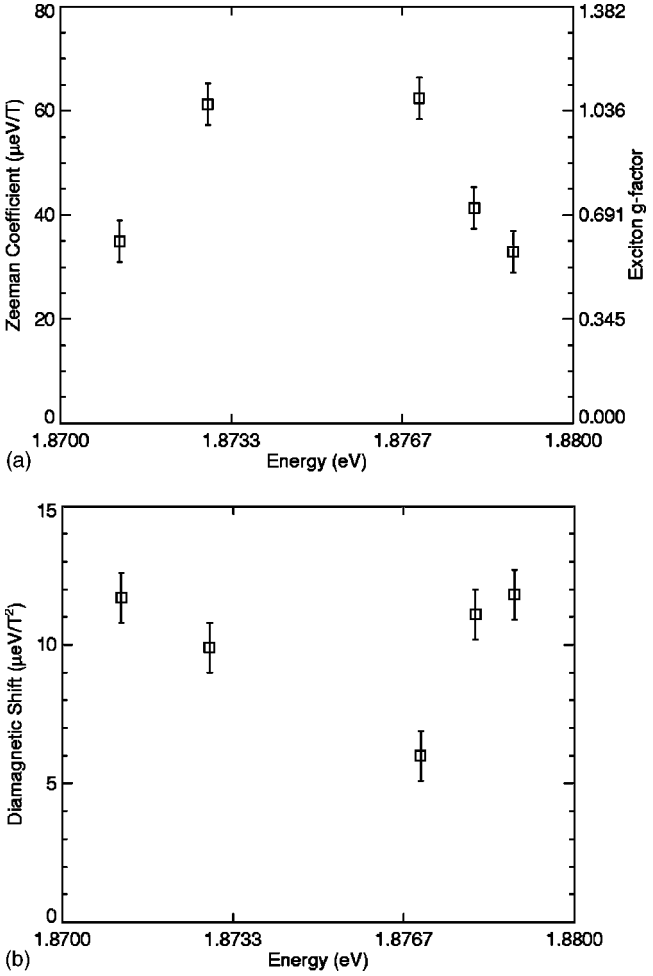

FIG. 5. (a) Zeeman coefficient and apparent exciton *g* factor and (b) diamagnetic shift, for select lines $a-e$ shown in Fig. 4.

Biexciton and multiexciton intensity dependence of sharp line states in ordered GaInP have been reported by previous authors.[3,8] In light of this, it is reasonable to ask if excited state or multiexciton effects can account for the variations in magneto-optical properties discussed above. Figure 6 shows the low-temperature photoluminescence spectra collected from a single aperture at varying excitation power, each spectrum is normalized to its own peak intensity. At the lowest-excitation power, three sharp lines appear (labeled *b*, *c*, and *d*). As the power is increased, new lines appear (labeled *a*, *e*, and *f* with vertical dashed lines through each peak). The peak intensity of these new lines show a weak superlinear power dependence. Lines *b* and *c* increase sublinearly, while line *d* increases linearly. Further increases in power result in the sharp lines being overcome by the overlapping broad band emission. A nonlinear power dependence can be interpreted as evidence that excited state or multiexciton states are being populated. However, it is also consistent with the intersite exciton transfer previously mentioned and discussed below.

As was stated earlier, the PLE measurements in this

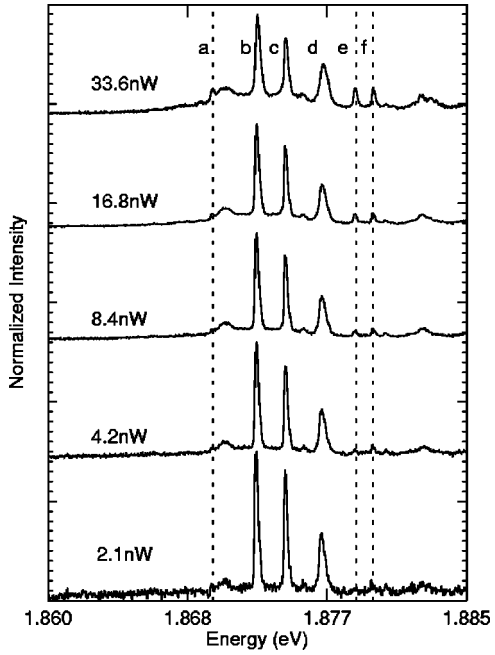

FIG. 6. Series of normalized spectra at varying excitation power (2.1–33.6 nW).

sample failed to find systematic evidence for excited state emission, and proposed a model of multiple spatially neighboring localized states coupled via intersite tunneling.[8] Such coupling would inhibit excited state emission. In the GaAs/AlGaAs quantum dot system of Heller and Bockelmann,[10] both the diamagnetic shift and Zeeman splitting of excited states differ strongly from those of the ground state, where both undergo a sign change. Though this likely depends on the details of the particular system, we observe only positive diamagnetic shifts and no sign reversal of Zeeman splittings.

Line *a* of Fig. 6 appears similar to a biexciton, lines *e* and *f* resemble excited state emission. These lines show no anomalous magnetic field behavior, however. All show positive diamagnetic shift and Zeeman splittings. The magnetic field behavior of the biexciton is expected to follow the exciton, as the biexciton state is a spin singlet which decays into the spin-split exciton state.[15] The magnetic field behavior of multiexciton states in a quantum dot, to our knowledge, are not well characterized. The preceding arguments suggest that the existence or nonexistence of these types of emission should not be important in the context of this discussion. The previously mentioned fact that large variations in both diamagnetic shift and *g* factor for transitions of similar energy, but located in different apertures, further confirms this assertion.

We examined the effect of an externally applied magnetic field on the micro photoluminescence of quantum confined sharp-line states in ordered GaInP. These transitions exhibit magneto-optic behavior similar to other well-studied III-V quantum dot systems. A large variation in both the diamagnetic shift and Zeeman splitting was observed, both for select lines which appear in a single 200-nm aperture, and for lines of similar energy but appearing in different apertures. These variations are likely due to a combination of fluctuations in alloy composition within the confined volume and variations in confinement geometry. The dispersion of diamagnetic shifts for $B\|(001)$ are comparable to those reported for $B\perp(1\bar{1}1)$ and $B\|(1\bar{1}1)$, and therefore weaken previous assertions regarding anisotropy of the confinement potential registered to the ordering direction $(1\bar{1}1)$. In light of these facts, there is little evidence to support the notion that these states are associated with quantum-discs formed at the antiphase boundaries in this material, as proposed by previous authors, and suggest that any new model: (i) need not be tied to this geometry and (ii) should incorporate the effects of these variations.

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