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Exciton Diamagnetic Shifts and Magnetic Field Dependent Linewidths in Ordered and Disordered InGaP Alloys

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ABSTRACT

We have measured the diamagnetic shifts and photoluminescence linewidths of excitonic transitions in ordered and disordered $In_{0.48}Ga_{0.52}P$ alloys, lattice matched to GaAs, in pulsed magnetic fields at 4 and 76K. The pulsed magnetic field ranged between 0 and 50T. The variations diamagnetic shifts with magnetic field in disordered and weakly ordered samples are considerably smaller than those calculated using a free exciton model. For a given magnetic field, the value of the diamagnetic shifts are found to increase with increasing order parameter. Furthermore, for all samples, the diamagnetic shifts at 76K are larger than at 4K suggesting that the excitons are strongly localized.

INTRODUCTION

An understanding and appreciation of the material parameters for the InGaP/GaAs system are important for laser and solar cell applications. In particular, this material system's bandgap energy can be tuned by the phenomenon known as spontaneous ordering, where, depending on the growth conditions, the InGaP crystal structure can be a randomly disordered alloy with the zinc-blende structure or can form a long-ranged ordered monolayer $(AC)_1/(BC)_1$ superlattice in the (111) orientation, known as the $CuPt_2$-like crystal structure. The degree of ordering has been cast in terms of an order parameter η. For $\eta = 0$, i.e., disordered zincblende structure, the 4-K bandgap energy is nearly 2 eV while for $\eta \approx 0.5$ ($CuPt_2$-like) the 4-K bandgap energy is smaller, ~1.9 eV. An excellent review describing this phenomena can be found in the article by Zunger and Mahajan [1].

Low-temperature photoluminescence (PL) is routinely used to asses the quality of semiconductor alloys. At liquid helium temperatures, the PL full-width-at-half-maximum (FWHM) linewidth of an excitonic transition is generally larger than those observed in their binary components. This broadening is proportional to the amount of compositional disorder [2-9] which

is inevitably present in these systems. Because alloy fluctuations are reduced in ordered structures, the PL FWHM linewidth decreases with increasing order parameter η [10].

In this paper we present PL data at 4 and 76K for the diamagnetic shifts of the excitonic transition and FWHM as a function of magnetic field in two InGaP samples, a weakly ordered and disordered $In_{0.48}Ga_{0.52}P$ alloy. For both the weakly ordered and disordered samples, we find that the observed variations of the FWHM with magnetic field are smaller than those calculated by Lee and Bajaj[7] using a free exciton model. The value of the exciton diamagnetic shift, for a given magnetic field, is also dependent on η. The values of the diamagnetic shifts are found to increase with increasing order parameter. Furthermore, for all samples, the diamagnetic shifts at 76K are larger than at 4K suggesting that the excitons are strongly localized. Previously reported magnetic field studies [11, 12] for InGaP alloys were restricted to the disordered samples.

EXPERIMENTAL

The InGaP samples were grown by metal-organic vapor-phase-deposition (MOCVD.) Substrates used were n^+-type GaAs with a misorientation of 6° to the nearest <111>A axis. A structure consisting of a epilayer of GaAs grown on top of the substrate followed by a 1-μm-thick-layer of InGaP. The order parameters were determined using polarized PL techniques described in Ref. 10. The samples are loaded directly into liquid helium in the tail of a cryostat which itself slips into the 15-mm-diameter bore of a 0.5-sec-duration 50T pulsed magnet. Light is coupled to

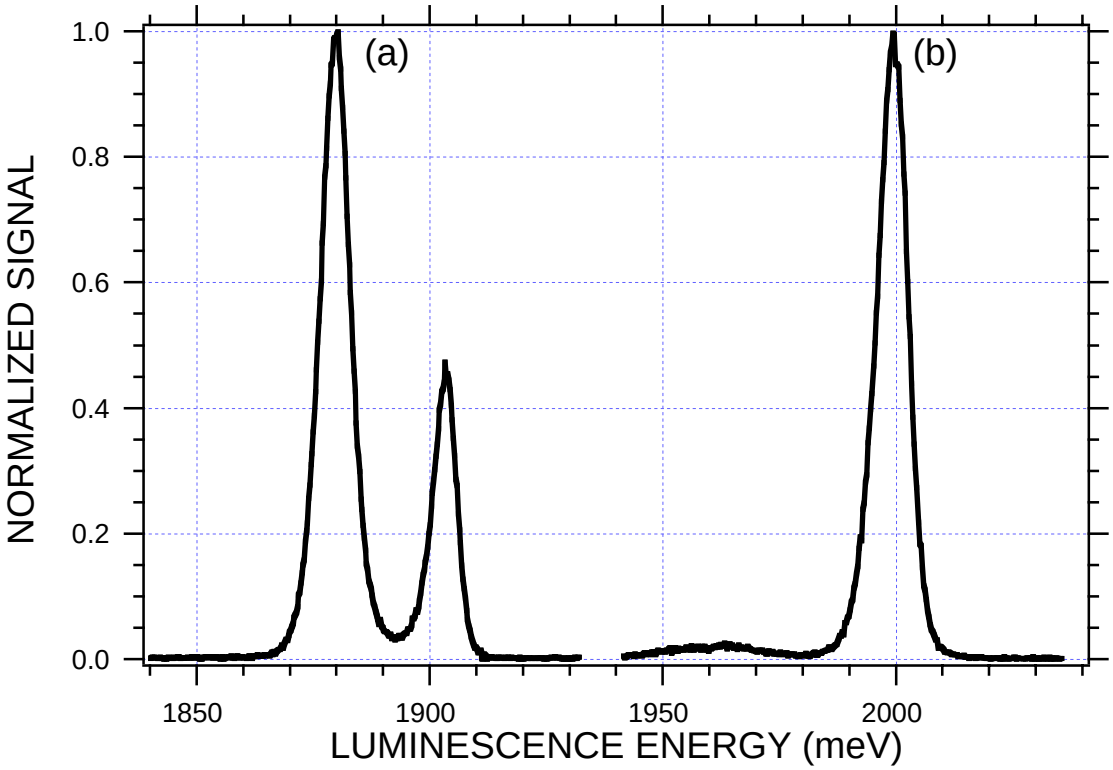

Figure 1. Low temperature (T=4K) photoluminescence spectra for a weakly ordered, $\eta \approx 0.5$, InGaP sample (a) and a disordered, $\eta \approx 0$, InGaP sample (b). As explained in the text, the lower energy peak in spectrum (a) is from an impurity bound exciton.

the sample, and PL is collected from the sample, via a single 600-μm-diameter optical fiber. The PL signal is dispersed by a 1/3M spectrometer and detected by a back-thinned CCD detector. The CCD detector collects high-resolution spectra continuously throughout the entire magnet pulse at a rate of 600Hz. In this way the entire magnetic field dependence of the PL is acquired in a single 50T magnet pulse. The magnetic field orientation was along the <100> crystallographic axis. Currently, it is not possible to align the magnetic field along the <111> direction, one of the principal axis for the $CuPt_2$-like structures, in the small-bore pulsed-field magnet solenoid.

Figure 1 shows 4-K spectra for the two InGaP samples discussed here. Spectrum (a) is from the weakly ordered ($\eta \approx 0.5$) sample #MA407, while (b) is from a disordered ($\eta \approx 0$) sample #MA565. For these two spectra, the laser power density was about 1 mW/cm^2. A study of the intensity of the two peaks as a function of laser power density (and temperature) indicates that the lower energy peak of (a) is a result of unknown impurity bound exciton luminescence.

RESULTS AND DISCUSSION

Figure 2 shows the magnetic field dependencies for the diamagnetic shifts at T = 4K. Also shown in Fig. 2 are calculated diamagnetic shifts for two cases: (1) The diamagnetic shift for an InGaP free exciton, reduced exciton mass $\mu_{AV} = 0.08$, and (2) the diamagnetic shift for a InGaP infinite valence-band mass exciton, $\mu_{INF} = 0.13$. The diamagnetic shift for the disordered sample (b) and the calculated diamagnetic shift for the infinite valence-band mass exciton μ_{INF} are nearly identical strongly suggesting that the valence-band holes are strongly localized in this disordered InGaP sample. On the other hand, the diamagnetic shift for the weakly ordered sample (a) falls between the two theoretical cases suggesting that the valence-band holes are less localized when compared with results from the disordered sample. Not shown here are diamagnetic shifts for InGaP samples with varying order parameters η. However, the same general conclusions are reached, i.e., for a given magnetic field, with increasing order parameter η, the value of the diamagnetic shift also increases.

Figure 3 shows the dependence of the 4-K FWHM for the weakly ordered and disordered InGaP samples. As can be seen, the value of the zero-field FWHM of about 6 meV for the weakly ordered sample, trace (a), is much less than the 8 meV zero-field FWHM disordered sample, trace (b) and as mentioned previously, the reduction in the FWHM in going from a disordered to an ordered sample is a result of reduced alloy fluctuations in ordered structures [10]. The theoretical FWHM magnetic field dependence in a semiconductor alloy has been calculated by Mena et al., [13]. Currently, a comparison between theory and experiment for the data shown in Fig. 3 are being performed and the results will be reported in detail at a later date.

The 76-K diamagnetic shift data is shown in Fig. 4. The calculated diamagnetic shifts for the two excitons with reduced masses μ_{AV} and μ_{INF} are the same as shown in Fig. 2. It is obvious from Fig. 4 that the diamagnetic shifts for both the weakly ordered and disordered samples are larger at 76K compared to the 4-K data shown in Fig.2. One possible explanation is that the exciton center-of-mass thermal energy has overcome, somewhat, the valence-band mass confinement potential leading to an apparent "*lighter*" exciton reduced mass and hence a larger diamagnetic shift. In order to clarify these speculations, further experiments are needed before definite conclusions can be reached.

For completeness we show the 76-K FWHM magnetic field dependence for the two samples in Fig. 5. The increased FWHM at 76K is a result of broadening due to the higher temperatures. Because of thermal broadening, the FWHM for both samples are nearly the same.

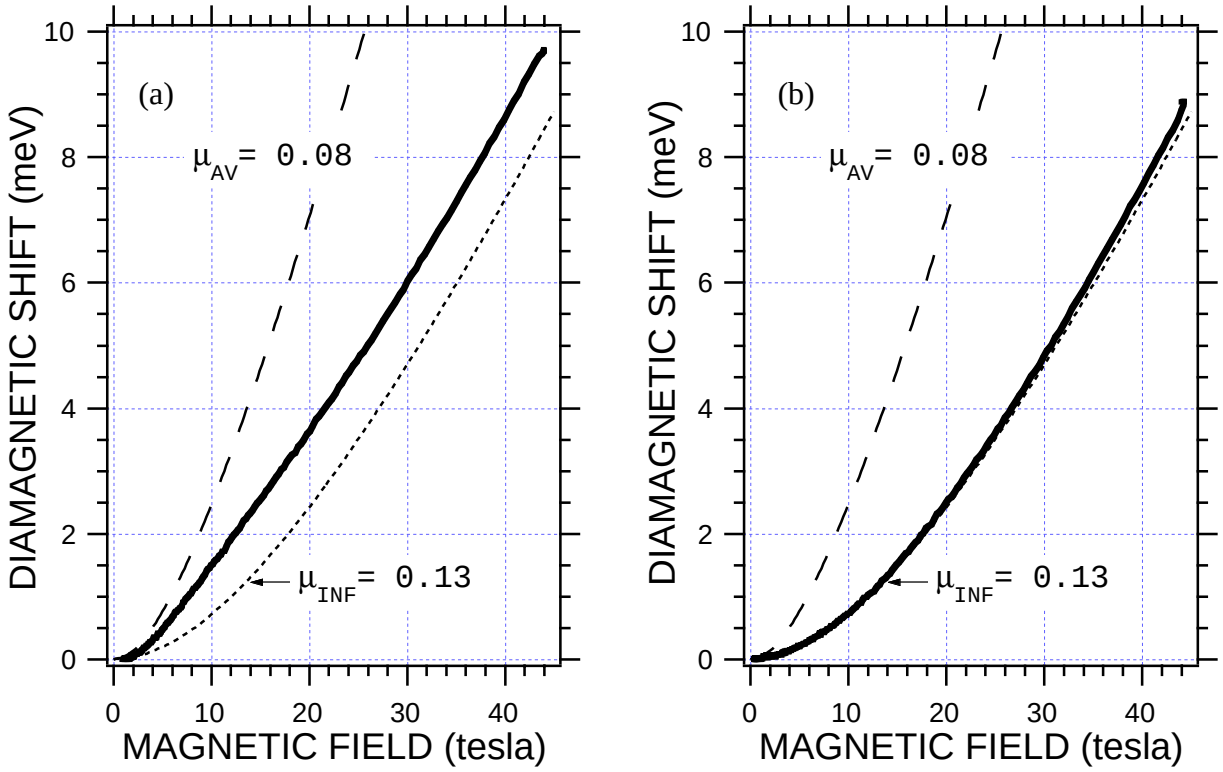

Figure 2. Magnetic field dependence of the exciton diamagnetic shift at 4K. The left graph (a) is for the partially ordered InGaP, E_{gap} = 1903.2 meV. The right graph (b) is for the disordered sam ple, E_{gap} = 1999.2 meV. The theoretical diamagnetic shifts for a free exciton with reduced excitor mass, μ_{AV} = 0.08, and an infinite-valence band mass exciton, μ_{INF} = 0.13 are shown.

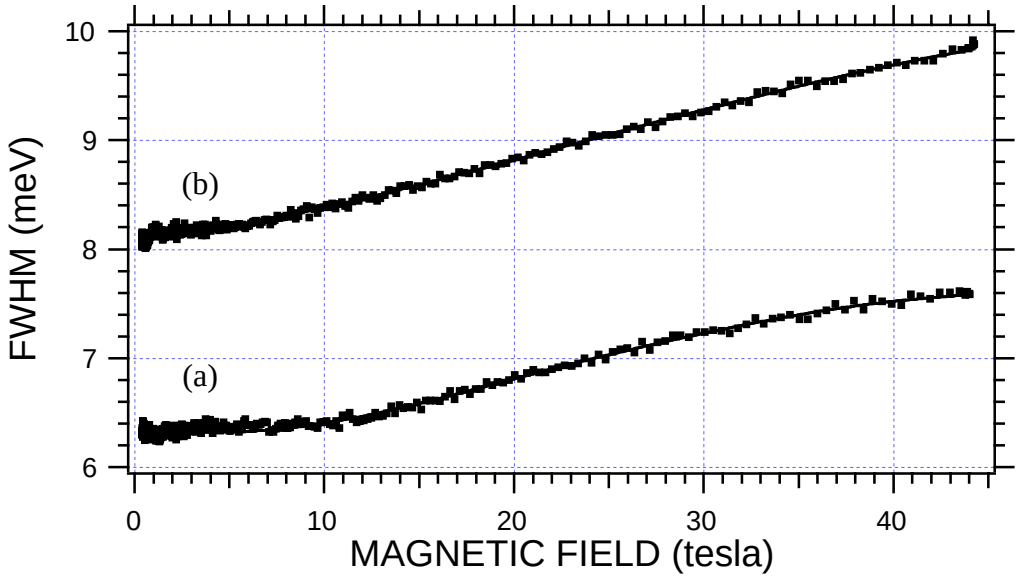

Figure 3. Magnetic field dependence of the FWHM at 4K. The upper trace (b) is for the disor- dered InGaP and the lower trace (a) is for the partially ordered InGaP sample.

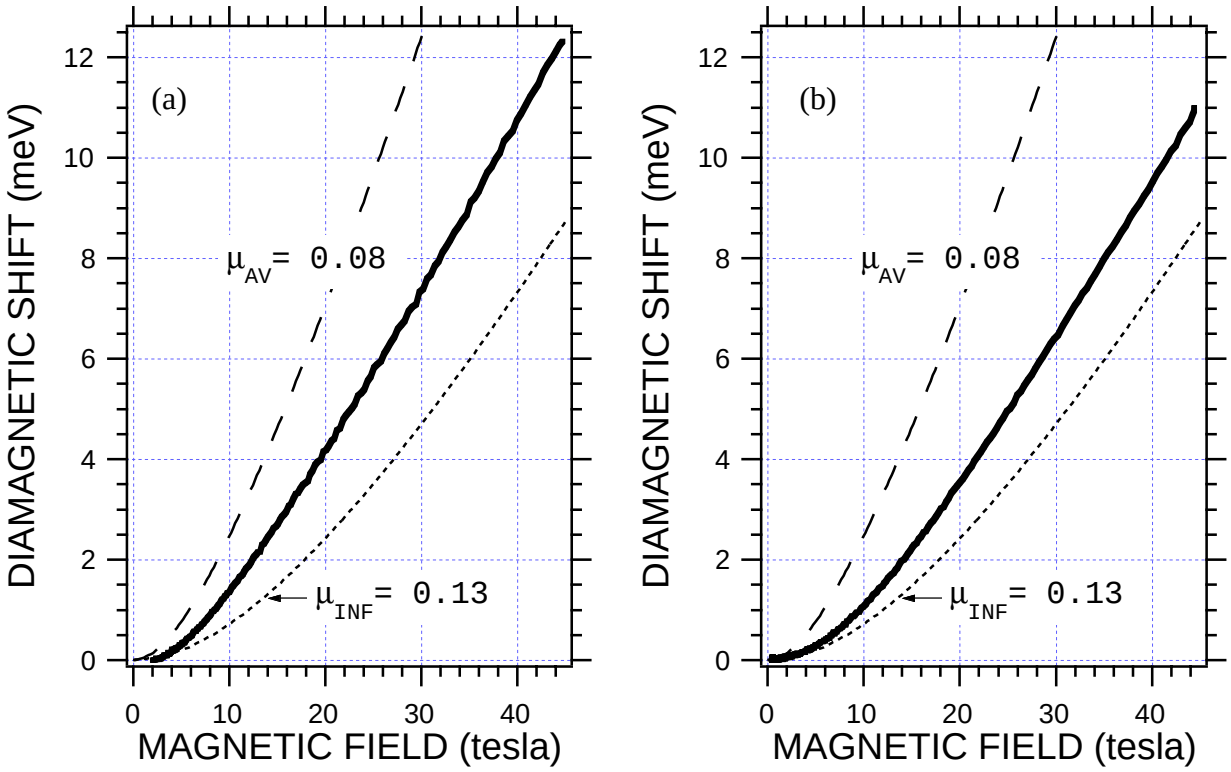

Figure 4. Magnetic field dependence of the exciton diamagnetic shift at 76K. Graph (a) is for partially ordered InGaP and graph (b) is for disordered InGaP. The 76K bandgap energies are respectively E_{gap} = 1900 meV and E_{gap} = 2004.8 meV for graphs (a) and (b). The theoretical shifts labeled μ_{AV} and μ_{INF} are the same as shown in Fig. 2.

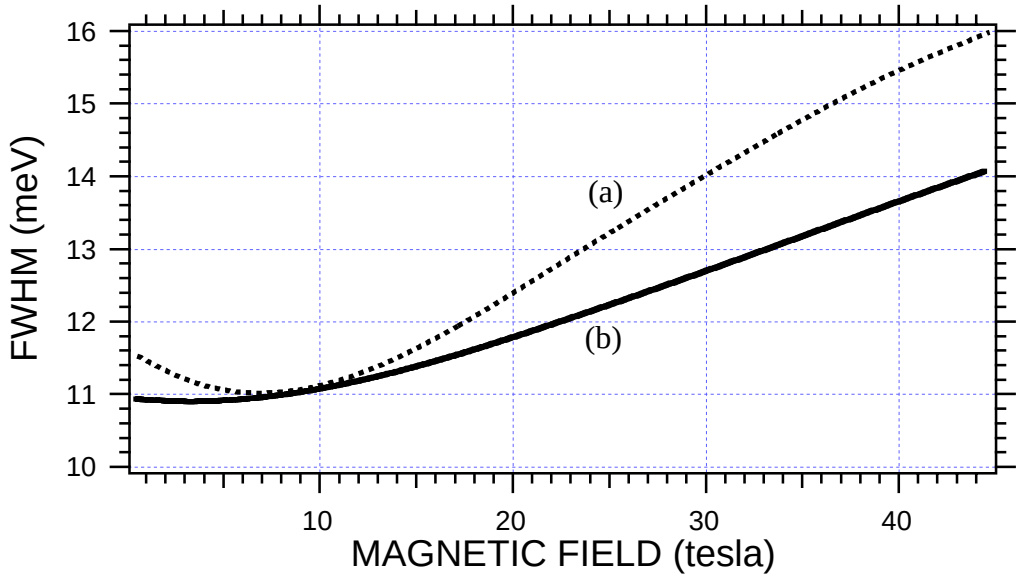

Figure 5. Magnetic field dependence of the FWHM at 76K. The dashed trace (a) is for the partially ordered InGaP and the solid trace (b) is for the disordered sample.

CONCLUSION

In summary, we have measured both the diamagnetic shifts and the linewidths of excitonic transitions in a series of weakly ordered and disordered $In_{0.48}Ga_{0.52}P$ alloys, lattice matched to GaAs, as a function of magnetic field at 4 and 76K using photoluminescence spectroscopy. The magnetic field was varied from 0 to 50T. The value of the exciton diamagnetic shift, for a given magnetic field, is also dependent on the order parameter η. The value of the diamagnetic shifts are found to increase with increasing order parameter. Furthermore, for all samples, the diamagnetic shifts at 76K are larger than at 4K suggesting that the excitons are more localized at 4K than at 76K.

ACKNOWLEDGEMENTS

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