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MAGNETOLUMINESCENCE STUDIES IN ORDERED $InGaP_2$

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ABSTRACT

Photoluminescence measurements on ordered $InGaP_2$ were studied as a function of temperature, laser power density, and magnetic field. The temperature varied between 1.4 and 300 K, the laser power densities ranged from 10 nW/cm^2 to 20 W/cm^2, and the maximum magnetic field was 13.6 T. The data show both excitonic and band-to-band behavior, depending upon the incident laser power density. A consistent interpretation of all data leads to a type-II valence-band offset between the ordered domains. Finally, a charge transfer model between domains which provides an explanation for intensity dependent energy shifts is discussed.

INTRODUCTION

The $In_xGa_{1-x}P$ alloy system exhibits many interesting physical and crystallographic properties. For concentrations x ~ 0.5, there are several crystallographic phases, varying from a random (disordered) alloy with the zincblende structure, to an ordered CuPt-type structure. With the different crystallographic forms is a changing bandgap energy. The reader is referred to two review articles [1,2] (and the references contained therein) describing some of the physical properties of this alloy system. The lattice constant for $InGaP_2$ is nearly equal to that of GaAs and thus there have been many studies describing the differences in the optical properties, bandgap energies, etc., between the disordered and CuPt-type ordered phases of $InGaP_2$. Because of the large numbers of reported photoluminescence (PL) studies only a few recent publications describing these observations [3-7] are referenced here and is not meant to be complete.

In this paper, we report on temperature and magnetic field dependent PL studies of an $In_{0.48}Ga_{0.52}P$ alloy lattice-matched to GaAs which exhibits well behaved ordering properties, i.e., a narrow range of order parameters.

EXPERIMENTAL

The sample was grown at a temperature of 675 C by low-pressure metal-organic chemical vapor deposition on a (100) GaAs substrate, tilted 2° towards <110>. Sample characterization techniques consisted of double-crystal x-ray scattering measurements, transmission electron microscopy (TEM) measurements, polarized photomodulated reflectance spectroscopy, ellipsometry, polarized photoluminescence excitation (PLE) spectroscopy, and low temperature PL. The double-crystal x-ray scattering data indicate that the $InGaP_2$ lattice constant is within 0.1% of the GaAs-substrate lattice constant. The results from the TEM measurements show that there is almost a single <111> variant for the CuPt-type ordering. Analyses of the polarized photomodu-

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lated reflectance spectroscopy indicate that the energy difference between the crystal field split valence-bands is about 30 meV. Figure 1 shows the polarized PLE spectra obtained at 10 K. The two polarizations are indicated in the figure and the energy difference in their respective peaks is about 30 meV in agreement with the polarized photomodulated reflectance spectroscopy data. The energy difference between the PL peak and the PLE peaks lead to an exciton binding energy of about 15 meV.Temperature dependent PL measurements were made between 1.4 and 300 K using a 3/4-meter double monochromator and standard photon counting apparatus.

RESULTS

Figure 2 shows 4-K spectra taken at two different laser power densities, 60 nW/cm^2 and 4 W/cm^2. The peak of the 4 W/cm^2 spectrum is near 1900 meV while the 60 nW/cm^2 spectrum has two peaks, a smaller amplitude still at 1900 meV and the other (largest) near 1840 meV. The peak-amplitude of the of the 4 W/cm^2 data is about 1000 times larger than that for the 60 nW/cm^2 spectrum. As can be seen in Fig. 2, there is still evidence of a peak near 1900 meV for the low power density data and a detailed examination of the high power density data shows a small-amplitude peak near 1850 meV. Thus, depending on the incident power density of the exciting laser, the peak appears to move between 1840 and 1900 meV. Power dependent shifts for ordered $InGaP_2$ alloys the have been previously studied by DeLong et al [3] and they presented their results in terms of a spatially indirect conduction electron and a valence-band hole. We performed low-temperature PL lifetime time measurements as a function of energy on our sample and find, in agreement with others [3,6], that for the low energy portion of the spectrum, the PL decay time τ is very long, τ ~

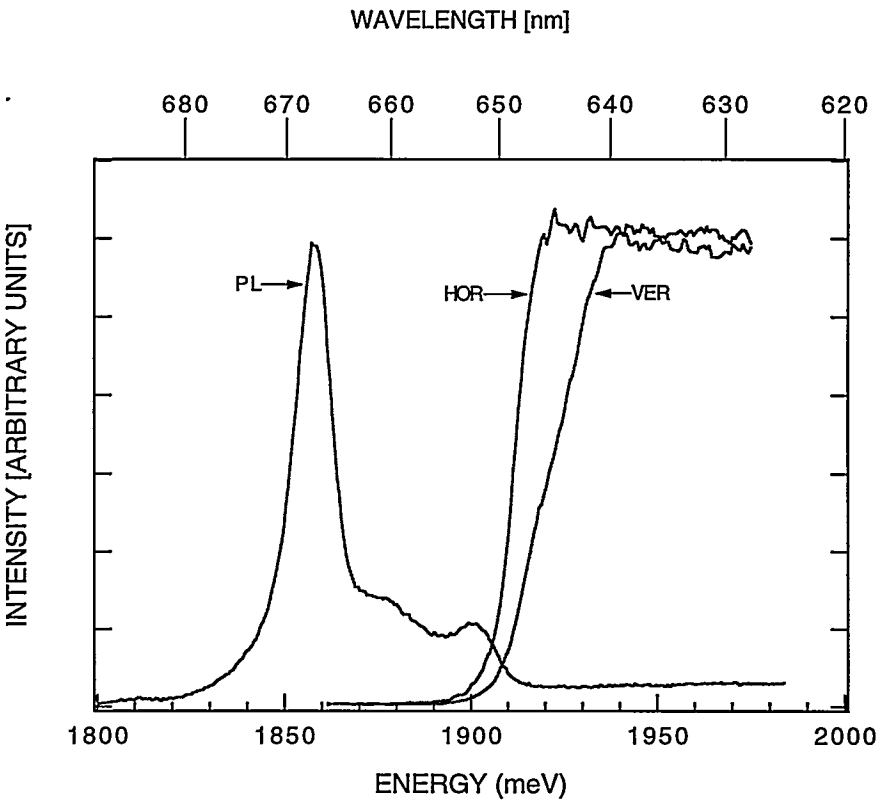

Figure 1. Photoluminescence excitation spectroscopy at 10K for two polarizations. The PL spectrum is also indicated. Note the 30 meV difference between the two PLE peaks.

50 μsec, while for the high energy side of the spectrum, e.g., E ~ 1900 meV, $\tau < 100$ nsec, the resolution of our measurement.

The temperature dependencies of the spectra shown in Fig. 2 were also studied between 1.4 and 300 K and again there are observed spectral peak shifts and our observations are in substantial agreement with similar studies [3,5]. The main result of these studies is that as the temperature is raised from 4 K, the double peak structure disappears leaving only the single higher energy peak identical to that in the higher power density data.

In order to gather new information about the mechanisms giving rise to the spectra shown in Fig. 2, we performed magnetoluminescence measurements between 0 and 14 tesla. The orientation of the applied magnetic field was along the growth direction, e.g., (100) and hence the field is at an angle to the (111) ordering planes. From the magnetic field dependence of the PL shift, one can distinguish between excitonic behavior, band-to-band transitions, or donor-acceptor pair recombination. The exciton diamagnetic energy shift is given by

$$\Delta E_{ex}^{dia} = \frac{\hbar^4 \kappa^2}{4\mu^3 e^2 c^2} B^2, \qquad (1)$$

where B is the magnetic field, μ is the reduced mass ($\mu^{-1} = m_c^{-1} + m_v^{-1}$) where m_c and m_v are respectively the conduction-band and valence-band effective masses in terms of the free electron mass m_0, and κ is the dielectric constant.

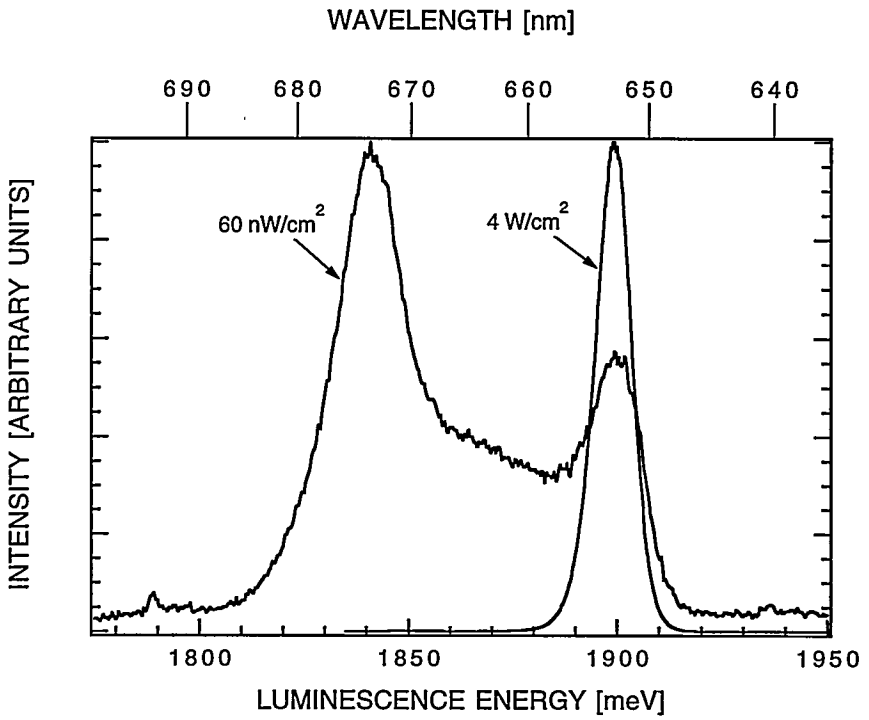

Figure 2. Low temperature (4 K) photoluminescence spectra at two laser power densities, 60 nW/cm^2 and 4 W/cm^2. The intensity of the 4 W/cm^2 peak at 1900 meV is about 1000 times larger than the intensity of the 60 nW/cm^2 peak at 1845 meV.

The magnetic field dependence of the higher laser power density peak of Fig. 2 is shown as the lower data set in Fig. 3. The solid line drawn through the data is quadratic, i.e., $\Delta E \sim B^2$, indicating excitonic behavior. Using a conduction band mass $m_c \sim 0.1m_0$ and a valence band mass $m_v \sim 0.2m_0$ yields reasonable agreement between Eq. 1 and the measured energy shift. It should be noted here that for disordered $InGaP_2$ alloys, an analysis of the magnetic field induced shifts also indicated [4] excitonic behavior, i.e., $\Delta E \sim B^2$. The low-temperature PLE measurements (Fig. 1) made on this sample show a binding energy of about 15 meV. For excitons in $InGaP_2$ the expected Rydberg is in the same range (15 meV) which along with the quadratic field dependence and short PL decay times confirm the excitonic identification for this PL peak.

In contrast to the quadratic field dependence of Eq. (1), band-to-band luminescence energy shifts are linear in the magnetic field and given in terms of the Landau index n = 0, 1, 2, 3 ... by

$$\Delta E_{bb} = \left(n + \frac{1}{2}\right)\frac{e\hbar}{\mu c}B. \qquad (2)$$

Because of the linear field dependence, the low power density data (upper data set of Fig. 3) is interpreted in terms of band-to-band behavior. The maximum energy shift of 12 meV is fairly large and is also indicative of the band-to-band nature of this peak. Magnetic field induced energy shifts were also taken for a range of other power densities between those shown in Fig. 3 with the result that as the power-dependent peak-intensity neared the exciton peak, the magnitude of the maximum shifts were reduced and the magnetic dependence went from linear behavior at low power densities to quadratic at high power densities.

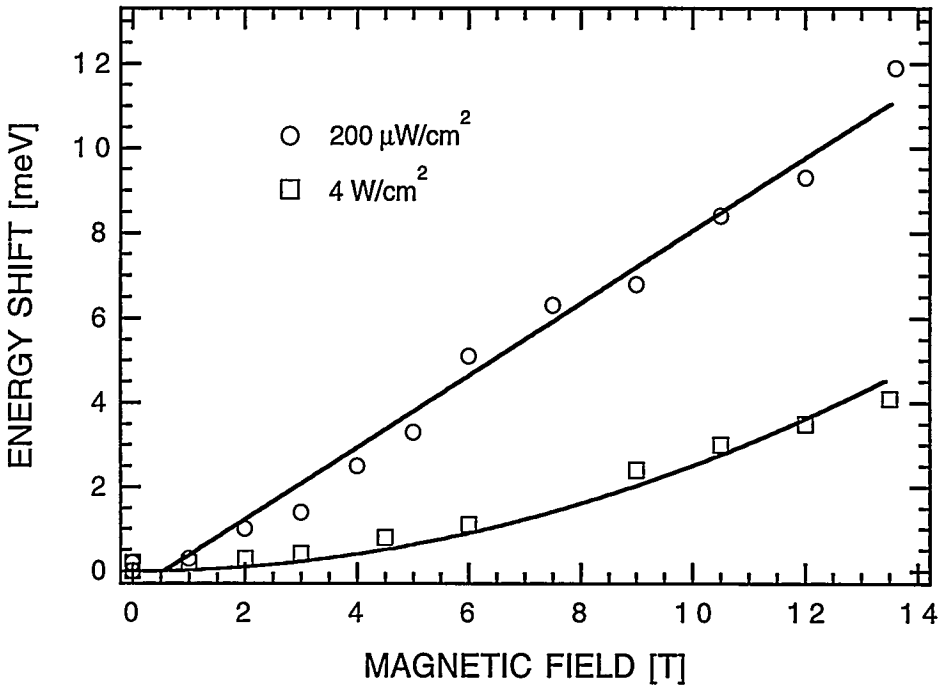

Figure 3. Magnetic field dependent PL shifts for two laser power densities, 200 $\mu W/cm^2$ ($E_{peak} \sim 1850$ meV) and 4 W/cm^2 ($E_{peak} \sim 1900$ meV) and 1.4 K. The straight line through the 200 $\mu W/cm^2$ data has a slope of 0.87 meV/T. The line drawn through the high density (4 W/cm^2) data is quadratic in the magnetic field.

DISCUSSION

From the linear magnetic field behavior, we believe that the PL peak found for low power densities represents a band-to-band transition. Using Eq. 2 and the measured slope of 0.87 meV/T, one can calculate the reduced mass μ. The conduction-band mass in ordered $InGaP_2$ was recently measured [8] to be $m_c \sim 0.09m_0$ by cyclotron resonance techniques. With a knowledge of the reduced mass μ and the conduction-band mass m_c, the valence-band mass m_v can be calculated to be $m_v \sim 0.25m_0$. Using the measured crystal-field split valence-band energy of 30 meV, we have performed a $k \cdot p$ calculation for ordered $InGaP_2$ and find that the valence-band is highly anisotropic, with $m_v$varying from $0.16m_0$ in the (111) plane to $0.33m_0$ for a direction perpendicular to (111). For a magnetic field along the (100) direction, the $k \cdot p$ calculation yields a valence-band mass $m_v \sim 0.2m_0$ which is in good agreement with that inferred ($0.25m_0$) from the magnetoluminescence data. Because of the linear magnetic field shift for the low power density PL peak and the agreement between the calculated and inferred valence-band masses, we assign these transitions as band-to-band.

The long lifetimes and linear magnetic field dependent energy shifts for the low power density data suggest a spatially indirect electron-hole recombination process. We rule out donor to acceptor recombination for two reasons; (1) the magnetic field data yielded a valence band mass of $m_v \sim 0.25m_0$ is in agreement with theoretical expectations, and (2) for donor-to-acceptor transitions the acceptor masses are very heavy and hence, the inferred mass would be $m_v > 0.7m_0$.

Both DeLong et al [3] and Fouquet, Minsky, and Rosner [6] suggest that spatially indirect electron-hole recombination is a result of radiative transitions in a disordered matrix of $InGaP_2$ containing the CuPt-type ordered $InGaP_2$, i.e., "concrete." On the other hand, Horner et al [7], present a picture where there is no disordered phase in the sample, but where all observations can be explained by considering a distribution function for the order parameter.

All of our data resulting from PL, PLE, photomodulated spectroscopy, or ellipsometry measurements show no indication of bandgap absorption near 2 eV, where the disordered phase of $InGaP_2$ should appear. In order to present a consistent interpretation of the data in terms of the model presented by Horner et al [7], we have to postulate a type-II valence-band offset. A type-II valence-band offset has been recently observed by F. A. J. M. Driessen [9].

The question of the power dependent position of the lower energy peak may be explained in terms of a charge transfer process between energy minima due to the random nature of the ordered domains as recently proposed by D. S. Kim, et al [10]. These authors have reported similar behavior in their PL studies of coupled GaAs/AlGaAs quantum wells. They found that as the population density of the two wells is altered by increasing laser power, the resulting coupled quantum well energy state is larger than either of the two isolated quantum wells resulting in a power dependent PL energy peak. The charge transfer process was attributed to the conduction-band electrons. Because of the heavy valence-band states, the tunneling probability is small and hence the resulting magnetic filed shifts for the PL energies would be linear for those carriers that have transferred. At very high excitation densities, both the conduction- and valence-band states will be equilibrated and thus act as a simple exciton, e.g. quadratic magnetic field behavior. While these interpretations are preliminary, this model is consistent with the observations of the magnetoluminescence data for ordered $InGaP_2$ alloys.

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