PHYSICAL REVIEW B **67**, 233304 (2003)

Magnetophotoluminescence studies of (InGa)(AsN)/GaAs heterostructures

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(Received 13 September 2002; revised manuscript received 18 December 2002; published 17 June 2003)

Photoluminescence (PL) measurements under a magnetic field ($B=0-12$ T) have been performed on $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y/\mathrm{GaAs}$ heterostructures for a wide range of N concentration ($0\leqslant y\leqslant 0.052$). We find that the value of the diamagnetic shift depends on temperature as long as localized carrier recombination contributes sizably to the PL spectra. Consequently, magneto-PL data have been taken at sufficiently high temperature so as to eliminate such contribution. The diamagnetic shift of the free exciton energy has been analyzed using a theoretical model developed in two dimensions for arbitrary strengths of the magnetic field. From this analysis we derive values of the electron effective mass, which are in good agreement with those reported in the literature from different experimental techniques and measuring different physical quantities. Finally, our work suggests that the concept of effective mass based on the envelope-function approximation holds also in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y/\mathrm{GaAs}$ system, although the crystal translational symmetry is strongly perturbed by the nitrogen atoms.

DOI: 10.1103/PhysRevB.67.233304 PACS number(s): 78.66.Fd, 71.35.Ji, 78.55.Cr, 71.55.Eq

I. INTRODUCTION

Recently, $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y/\mathrm{GaAs}$ heterostructures have attracted much interest owing to the strong modifications exerted by N incorporation into the band structure of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ host lattice. These include a giant band gap reduction[1] and a decrease in the rate at which the band gap varies upon hydrostatic pressure[2,3] and temperature increase.[4,5] Moreover, a strong dependence of the electron effective mass m_e on the nitrogen concentration has been found by a variety of experimental techniques.[6–13] The introduction of N in the host lattice is also a source of a high degree of disorder, which manifests itself on the optical properties of the material, leading to a sizable inhomogeneous broadening of the radiative transitions,[4] to a large Stokes shift between absorption and emission,[14] and to the presence of localized states, which at low temperature dominate often the emission spectra.[4,5] Moreover, from a more fundamental standpoint, the highly disordered potential arising from the large fluctuations induced by N might undermine a representation of the crystal electronic states in terms of coherent Bloch states with a well-defined **k** vector.[15] In turn, this could invalidate an effective mass scheme for describing the electronic properties of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y/\mathrm{GaAs}$ heterostructures, also an issue of great importance for the design of optoelectronic devices.[16–18]

In this work, we report on magneto-photoluminescence (magneto-PL) measurements in $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y/\mathrm{GaAs}$ heterostructures for a wide range of N concentrations. We show that the diamagnetic shift of the PL peak varies with temperature, T, being higher at lower T. This result indicates that the low-T PL emission is determined by the radiative recombination of a loosely bound electron-hole pair in which one carrier is localized by N-induced potential fluctuations and the other carrier is delocalized as suggested recently.[19] Therefore, in the analysis of the shift (ΔE_d) of the PL peak energy induced by B, we consider magneto-PL spectra at $T\geqslant 100$ K in order to avoid localized state contributions. We find that ΔE_d decreases with increasing y, for $x\sim 0.3$. By the dependence of ΔE_d on B for samples with different N concentration, we infer that the electron effective mass rapidly increases when y changes from 0 to 0.01 and saturates to $\sim 0.08m_0$ (m_0 is the bare electron mass) for higher y's (and $x\sim 0.3$). A comparison with data reported in the literature shows a good agreement between the values of m_e here estimated by the diamagnetic shift of the free exciton energy and those found by different experimental techniques for $y<0.02$ and measuring different physical quantities.[11–13] In turn, this allows one also to define a carrier effective mass concept in the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y/\mathrm{GaAs}$ system, where the virtual crystal approximation breaks down.

II. EXPERIMENT

We investigated $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y/\mathrm{GaAs}$ single quantum wells (QWs) ($x=0-0.38$, $y=0.007-0.052$, QW thickness $L=6-8$ nm), the corresponding N-free reference samples, and $\mathrm{GaAs}_{1-y}\mathrm{N}_y$ epilayers and QWs ($y\sim 0.01-0.02$, $L=9.0$ nm). All samples were grown by solid source molecular beam epitaxy. Photoluminescence was excited by the 515-nm line of an Ar^+ laser, dispersed by a $\frac{3}{4}$-m double monochromator, and detected by a liquid nitrogen cooled Ge detector. The measurements have been carried out in a liquid He optical cryostat between $T=10$ and 200 K. The magnetic field ($B=0-12$ T) was applied parallel to the growth axis of the samples.

III. RESULTS AND DISCUSSION

First we describe the effect of temperature on the magneto-PL spectra of N-containing samples. Figure 1

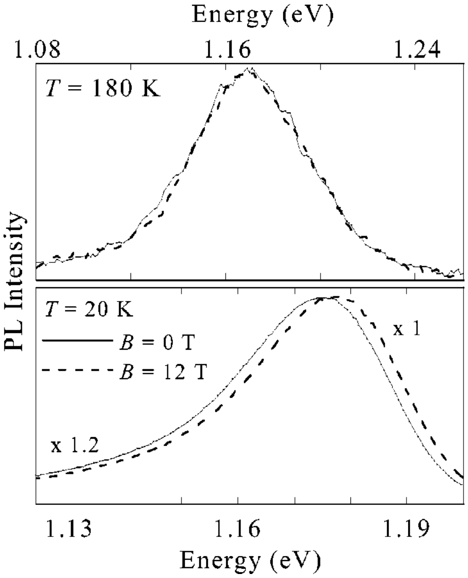

FIG. 1. Peak normalized PL spectra at $T=20$ K (lower panel) and 180 K (upper panel) of a 310-nm-thick $GaAs_{0.98}N_{0.016}$ epilayer at $B=0$ T (continuous line) and $B=12$ T (dashed line). Multiplication factors are given. Laser power density $P=20$ W/cm^2.

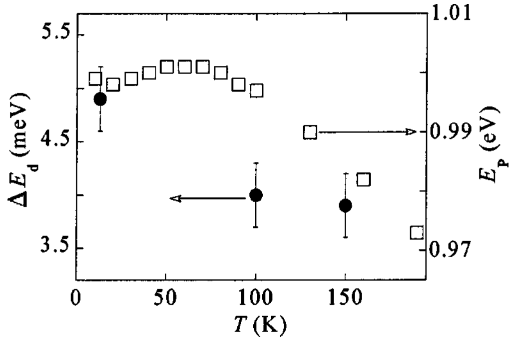

FIG. 2. Left axis: Dependence of the diamagnetic shift on temperature between $B=0$ and 12 T of the PL peak position of an $In_{0.32}Ga_{0.68}As_{0.973}N_{0.027}$ QW with $L=6.0$ nm. Right axis: Dependence on temperature of the PL peak position for the same sample.

shows the PL spectra of a 310-nm-thick $GaAs_{0.984}N_{0.016}$ epilayer taken at 20 K (lower panel) and 180 K (upper panel) for different values of the magnetic field. The B-induced shift of the PL peak energy between 0 and 12 T is zero at high temperature, whereas a 3.4-meV blueshift of the PL band is observed in the same B interval at $T=20$ K. Similar behaviors have been found in $GaAs_{1-y}N_y$ QWs, too (not shown here). The temperature dependence of ΔE_d is consistent with data on carrier dynamics in $GaAs_{1-y}N_y$ published recently.[19] In that work, the fast rise time ($\sim$25 ps) of the PL signal suggests that the radiative recombination in $GaAs_{1-y}N_y$ occurs between localized electrons and delocalized holes at low T, implying that the recombining electron-hole pair is more loosely bound than a localized or free *exciton*. In turn, the interaction of these electron-hole pairs with a magnetic field results in a stronger perturbation and, therefore, in a greater diamagnetic shift with respect to the exciton case. Since localized carriers are detrapped and free excitons dominate the PL spectrum as T increases, ΔE_d decreases with increasing temperature until it saturates to the diamagnetic shift of the free exciton. This behavior is exemplified in Fig. 2, where the diamagnetic shift (left ordinate axis) and the peak energy position (right ordinate axis) of the PL of an $In_{0.32}Ga_{0.68}As_{0.973}N_{0.027}$ QW are plotted vs T. As reported previously,[4,5] the nonmonotonic dependence of the PL peak energy vs T is a signature of carrier recombination from localized states. As carriers are detrapped from localized states, the band gap thermal variation of the sample loses its sigmoidal dependence and ΔE_d saturates ($T \geqslant 80$ K in Fig. 2). In order to ensure that only the free-exciton-related states contribute to the diamagnetic shift, in the following we will consider magneto-PL data recorded at high temperature.

For the range of magnetic field values employed in this work a sizeable diamagnetic shift can be measured in In-containing QWs, only. Two factors concur to give in $In_xGa_{1-x}As_{1-y}N_y$ a diamagnetic shift larger, namely, an electron effective mass smaller, than in $GaAs_{1-y}N_y$. First, the electron effective mass of the $In_xGa_{1-x}As$ host lattice has a smaller value than that of GaAs. Second, in terms of the phenomelogical interaction model of Ref. 3, the interaction between the N-induced and conduction band levels decreases when decreasing the host band gap (i.e., increasing In concentration).

The inset of Fig. 3 shows the PL spectra recorded on an $In_{0.32}Ga_{0.68}As_{0.973}N_{0.027}$ QW and an $In_{0.32}Ga_{0.68}As$ reference QW for representative B values. The PL spectra have been taken at a relatively high temperature ($T=100$ K) for the reason discussed above. In both samples the PL intensity increases with increasing B due to a squeezing of the exciton

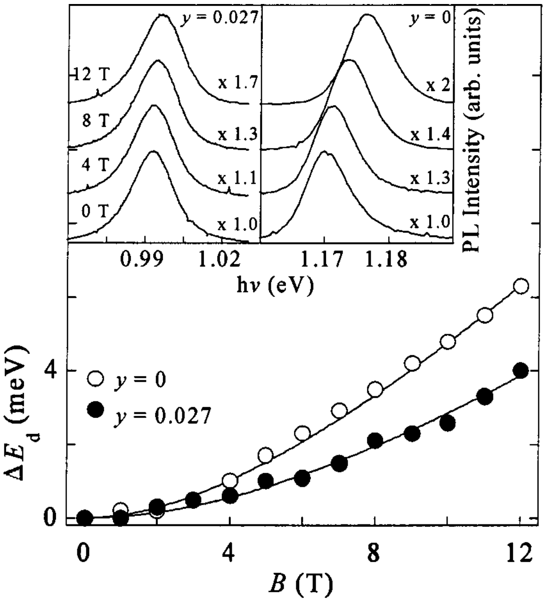

FIG. 3. Diamagnetic shift, ΔE_d, measured at $T=100$ K in an $In_{0.32}Ga_{0.68}As_{0.973}N_{0.027}$ QW (full dots) and in an $In_{0.32}Ga_{0.68}As$ reference QW (open dots) vs magnetic field, B. The continuous lines are fits of Eq. (1) to the data (the values of the only fitting parameter $\mu_{\parallel}$ are given in Table I). The inset shows the PL spectra of these two samples at representative magnetic field values. Multiplication factors are given. Laser power density $P=20$ W/cm^2.

TABLE I. List of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y$/GaAs samples investigated. x and y are the indium and nitrogen concentrations, respectively. L is the thickness of $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y$ quantum wells. $\mu_\parallel$ is the in-plane exciton reduced effective mass (in units of the bare electron mass, m_0), as determined by fitting Eq. (1) to the experimental diamagnetic shifts. The uncertainty in the estimate of $\mu_\parallel$ is also given. The last column gives the percentage variation in $\mu_\parallel$ between N-containing and N-free samples.

x	y	L (nm)	$\mu_\parallel$ (m_0)	$[\mu_\parallel(y)-\mu_\parallel(0)]/\mu_\parallel(0)$
0.34	0	7.0	0.038±0.001	0
0.34	0.007	7.0	0.045±0.001	0.18
0.25	0	6.0	0.042±0.001	0
0.25	0.011	6.0	0.052±0.001	0.24
0.32	0	6.4	0.039±0.001	0
0.32	0.027	6.0	0.049±0.001	0.26
0.38	0	8.0	0.038±0.001	0
0.38	0.052	8.2	0.048±0.001	0.26

wave function in the plane perpendicular to the field direction.[20] The diamagnetic shift $\Delta E_d = E_p(B) - E_p(0)$ of the PL peak energy E_p is shown in Fig. 3 for the two samples as a function of B. The presence of a few percent of N atoms leads to ΔE_d values quite smaller than those measured in the reference QW.

In our samples, the magnetic energy $\hbar\omega_c$ ($=\hbar eB/\mu_\parallel$, $\mu_\parallel$ being the exciton effective mass in the plane perpendicular to B) for $\mathrm{In}_{0.3}\mathrm{Ga}_{0.7}\mathrm{As}$ is about 18 meV at 6 T, namely, of the order of the expected exciton binding energy (~10 meV). Therefore, the usual linear B (Landau levels) or quadratic B^2 (perturbation theory) approximations do not hold in the whole 0–12-T field interval investigated here. Then, we analyze the diamagnetic shift data by using a numerical method proposed by MacDonald and Ritchie for two-dimensional Coulombic systems at arbitrary magnetic fields.[21–23] On the basis of this model the diamagnetic shift can be written in terms of a two-point Padé approximant,

$$\Delta E_d(B;z) = \sum_{i=0}^{7} p_i z^i \Bigg/ \sum_{k=0}^{5} q_k z^k, \tag{1}$$

where p_i and q_k are numerical coefficients whose values can be found in Ref. 21, $z = \sqrt{(\varepsilon^2\hbar^3 B)/(\mu_\parallel^2 e^3 c)}$, and ε is the material dielectric constant. A very good agreement is found between the experimental data and the theoretical fits (continuous lines) for both samples shown in Fig. 3—as well as for all the other $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y$ samples investigated here. The values of $\mu_\parallel$, which is the only fitting parameter, are reported in Table I together with the relative change in $\mu_\parallel$ between the N-containing and N-free QWs. Note that the smaller value of $\mu_\parallel$ found in the $y=0.052$ sample with respect to the to $y=0.011$ sample is due to the different In concentrations of the two $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ host matrices.

One might wonder if an envelope function approach can be used to derive an electron (and exciton) effective mass when the introduction of N atoms in the (InGa)As lattice strongly perturbs the periodic potential with an ensuing mixing of different **k** vectors in the electron wave function. In fact, Mattila *et al.*[24] have shown that the electron wave function at the conduction band edge, E_-, has a 42% non-Γ character, which results in a sizable wave function localization and in intrinsically short electron diffusion lengths. However, the conduction band edge state E_- has an extended character far away from nitrogen, and allows those authors to discuss the dependence of the E_- state effective mass on the nitrogen concentration.[24,25]

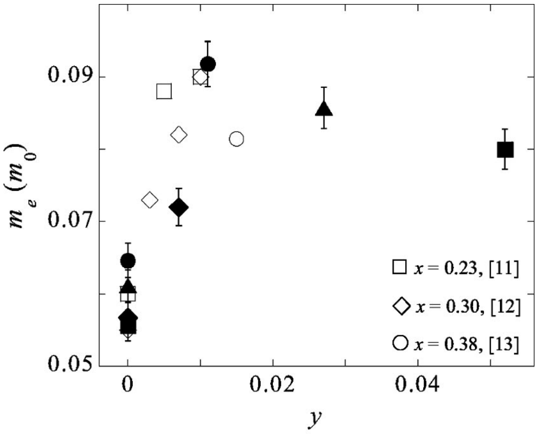

FIG. 4. N concentration dependence of the electron effective mass m_e for the samples studied in this work [full symbols: $x=0.25$ (●), $x=0.32$ (▲), $x=0.34$ (◆), and $x=0.38$ (■)] and taken from the literature (open symbols). PL data have been taken at $T=180$ K ($y=0.007$ and 0.011), and at $T=150$ K ($y=0.027$ and 0.052).

In order to verify the validity of an envelope function approximation in these disordered systems, from our data we estimate an electron effective mass to be compared with the experimental results reported in the literature for $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y$ with high x (>0.2).[11–13] To this purpose we set the in-plane heavy-hole effective mass equal to $0.12m_0$, as reported in Ref. 26 for samples similar to ours. Then, we get the m_e values shown as a function of y by full symbols in Fig. 4. Data reported by other authors for $y<0.02$ (and $x\sim0.3$) are shown in the same figure by open symbols. For $y\leqslant0.01$, m_e increases rapidly with y, as found in Refs. 11–13 on the ground of different experimental techniques, and seems to saturate at a high N concentration. The agreement between all these m_e values further justifies the use of an effective mass concept in $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}_{1-y}\mathrm{N}_y$. In Refs. 24 and 25, the increase in m_e has been attributed to a steady increase in the localized character of the conduction band edge up to y of the order of a few percent. In a phenomenological model,[3,7] the increase in m_e with the N concentration has been accounted for by a repulsive interaction between the conduction band minimum of the $\mathrm{In}_x\mathrm{Ga}_{1-x}\mathrm{As}$ host matrix and a strongly localized N level resonant with the conduction band states. Finally, tight-binding calculations within a band anticrossing framework account for the saturation of the electron effective mass upon increasing y, similarly to what is shown in Fig. 4.[27]

IV. CONCLUSIONS

In summary, we have shown that a measure of the diamagnetic shift of the free exciton PL needs to be done at a temperature high enough to get rid of localized carrier contributions. We have estimated by magneto-PL the electron effective mass in $In_xGa_{1-x}As_{1-y}N_y$/GaAs heterostructures up to N concentrations almost three times higher than those reported until now in the literature, to our knowledge. We show that N incorporation leads to an increase in the exciton reduced effective mass, which is consistent with theoretical predictions,[3,24,25,27] and to its saturation for high values of N concentration and $x \sim 0.3$.[27] The values of the electron effective mass derived here are in good agreement with those reported in the literature and derived by different experimental techniques. This supports the applicability of an effective mass scheme for describing the electronic properties of the (InGa)(AsN)/GaAs highly disordered system.

ACKNOWLEDGMENTS

The authors thank Y. Zhang for useful comments, and A. Miriametro and L. Ruggieri for technical assistance. This work was partially funded by Ministero dell'Universita' e della Ricerca Scientifica e Tecnologica (MIUR-COFIN 2001), and Progetto Giovani Ricercatori.

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