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To cite this article: D H Rodrigues *et al* 2012 *J. Phys. D: Appl. Phys.* **45** 215301

IOP PUBLISHING JOURNAL OF PHYSICS D: APPLIED PHYSICS

J. Phys. D: Appl. Phys. **45** (2012) 215301 (5pp) doi:10.1088/0022-3727/45/21/215301

Anomalous optical properties of GaMnAs/AlAs quantum wells grown by molecular beam epitaxy

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Received 6 February 2012
Published 9 May 2012
Online at stacks.iop.org/JPhysD/45/215301

Abstract
We have investigated the optical properties of GaMnAs/AlAs quantum wells (QWs) grown by molecular beam epitaxy under relatively high substrate temperatures (400 and 450 °C) and low Mn concentrations ($\leqslant$0.1%). We have studied the time- and polarized-resolved photoluminescence emission as a function of the laser power and an applied magnetic field. Several anomalous results have been observed including long decay times, enhancement of the diamagnetic shift and reduction in the polarization degree with increasing Mn concentration.

(Some figures may appear in colour only in the online journal)

1. Introduction

Dilute magnetic semiconductors have been investigated intensively in the last years due to their large potential for spintronic applications [1–3]. The combination of the electronic structure of a semiconductor with magnetic properties makes it possible to develop a distinct class of devices based not only on the charge but also on the spin of the carriers. Among the new materials that have been explored, GaMnAs alloys are especially interesting as they are based on a traditional semiconductor system which has already reached a high developed stage of fabrication. Even though GaMnAs is one of the most extensively investigated dilute magnetic semiconductors, some fundamental questions concerning this material still remain relative to its electronic structure. One of the fundamental questions is wheter the Mn doping generates or not an impurity band merged with the GaAs valence band, as this would have a profound impact on the origin of ferromagnetism on such alloys. On the other hand, from the point of view technology, a major problem for developing GaMnAs alloy devices is the incorporation of Mn atoms with concentration levels that are sufficiently large to attain ferromagnetic properties while maintaining good optical properties. Usually, in order to overcome the low Mn solubility in GaAs, GaMnAs growth is performed at relatively low temperatures (~250 °C), that significantly increases the Mn incorporation on the GaAs host. Unfortunately, optical studies [4–8] have shown that the low growth temperature also generates a large density of defects that quenches the optical emission. Therefore, the optical properties of GaMnAs alloys remain mainly unexplored.

In this work, a detailed optical analysis of GaMnAs/AlAs quantum wells (QWs) with low Mn concentrations ($x_{Mn} \leqslant$ 0.1%) has been performed. For those concentrations, the growth can be performed at relatively high substrate temperatures (400–450 °C), so that the resulting structures preserve a high degree of the optical quality. The optical properties of 6 nm thick QWs were investigated by means of photoluminescence (PL) and time-resolved PL measurements, and varying several experimental parameters such as the excitation intensity and external magnetic field.

2. Experimental details

The samples were grown on semi-insulating GaAs substrates in a Veeco Modular Gen II molecular beam epitaxy (MBE)

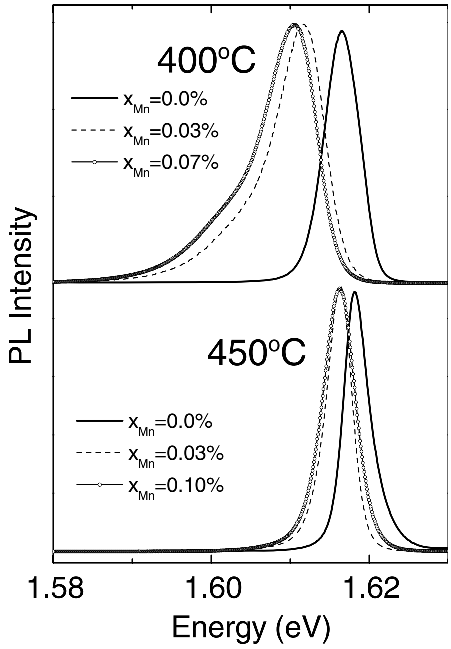

Figure 1. Normalized PL spectra for a set of 6 nm QWs with different Mn concentrations and growth temperatures.

reactor equipped with a real-time wafer temperature sensor, which allows accurate monitoring of the substrate temperature during growth. The structures were grown using As_4 species on In-free sample holders and consist of a GaAs buffer layer and an AlAs/GaAs/$Ga_{1-x}Mn_xAs$ QW. During the GaMnAs QW growth, the As_4/(Ga+Mn) ratio was kept close to 1.The QW was surrounded by 50 nm thick AlAs barriers. The total QW thickness is 6 nm, which comprises two 1 nm layers of GaAs adjacent to the AlAs interfaces and a 4 nm thick GaMnAs layer. The structures were capped with a 10 nm GaAs layer. This configuration was used with the intent to reduce the effect of interface segregation of Mn that might degrade the optical properties of the QW. The nominal Mn concentration was varied from 0% up to 0.07%. The GaAs buffer layer was grown at 580 °C and the rest of the structure was grown at 400 or 450 °C.

PL measurements were performed using either an Ar laser for continuous excitation (CW) or a picosecond Ti-Sa laser for time-resolved measurements (TR). The detection was performed using a Si CCD (CW) or a streak-camera system (TR). The sample was maintained at 4 K and a magnetic field (up to 15 T) was applied perpendicular to the sample surface (Faraday geometry).

3. Results

The CW-PL emission from a set of GaMnAs/AlAs samples is presented in figure 1. On the whole, the introduction of Mn results in a broadening of the PL band, a decreasing of optical efficiency and a red-shift of the PL peak as expected for GaMnAs QWs with low Mn concentrations. An additional broad band was also observed at lower energy side of the main emission when the Mn concentration is increased. The PL intensity of this band shows a clear correlation with the Mn concentration. This indicates that this recombination should involve a Mn-related level. Similar results were previously obtained for GaMnAs/GaAlAs QWs grown at 400 °C [6]. It was shown that lowering the substrate temperatures during the GaMnAs/GaAlAs QW growth resulted on sharper Mn profiles and higher Mn concentrations [6]. Furthermore, similar to our results, an increased Mn incorporation was followed by the appearance of a low-energy shoulder on the main QW PL peak. This additional band was attributed to a shallow donor associated with interstitial Mn (Mn_i) [6]. This attribution is supported by the theoretical prediction concerning the impurity level created by Mn_i in GaAs [9]. In bulk GaAs, Mn_i act as a double donor and their states remain inside the GaAs conduction band. Calculations for the electronic structure of Mn_i in a GaAs QW show, however, that the QW confinement should result in a shift of the Mn_i states to lower energies, so that they are located to a position within the GaAs band gap [9]. This finding justifies why transitions related to Mn_i donors are never observed in bulk GaAs, but there is experimental evidence of such level in GaAs QWs. This shoulder was not observed in the PL spectra of our samples grown at 450 °C, probably because the amount of incorporated Mn is smaller in this case, as it should depend on the substrate temperature. We have also observed that for equal amounts of nominal Mn, the red shift of the main PL band is larger for samples grown at lower temperatures (400 °C) as compared with those grown at 450 °C, which is consistent with increasing Mn incorporation efficiency at lower growth temperatures.

Figure 2 presents the TR-PL results for two samples grown at 400 °C at 15 mW laser power. From the presenting transients we obtained decay time of ~15 ns for the reference sample, and ~3 ns for the sample with $x_{Mn} = 0.03\%$, for both the main QW PL peak and for the low-energy shoulder that we associated with a shallow Mn donor level. The long PL decay time for the reference sample is a rather anomalous result for a single QW, whose origin is still under investigation. A possible explanation may be related to the QW interfaces with the indirect-AlAs barriers. In particular, the observed reduction in the PL decay time of the sample with Mn is consistent with the creation of additional decay channels associated with the Mn incorporation.

Magneto-optical measurements were performed at 4 K for magnetic fields up to 15 T under the Faraday configuration. Figure 3 shows the resulting magneto-shift of the QW emission bands for all the samples under a laser power excitation of 20 mW. We observe an overall increase in the magneto-shift with the Mn concentration and a clear correlation with the results from figure 1. The two samples with the largest Mn concentrations grown at 400 °C present a distinctly larger magneto-shift, and they are precisely the samples for which the QW PL peak appears at lower energies. Furthermore, the low-energy shoulder associated with Mn_i is also larger for those two samples. Considering that the magneto-shift basically reflects the localization of the carriers' wave-functions along the QW plane, it should depend mainly on parameters such as the QW thickness and the interface roughness. Concerning the QW thickness, it is not expected that a systematic variation of the QW thickness with increasing Mn concentration can

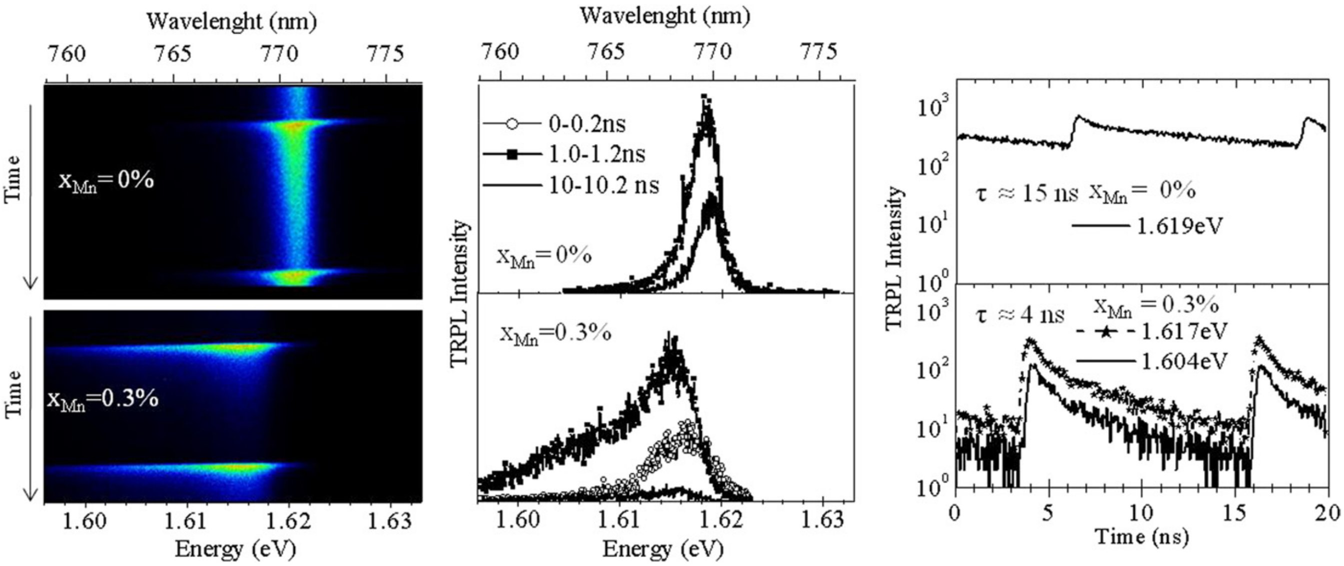

Figure 2. Time-resolved PL results for samples grown at 400 °C with $x_{\mathrm{Mn}} = 0\%$ and 0.03%. First column: streak-camera images, where the horizontal and vertical axes correspond to the emission wavelength and the time delay, respectively. The total time window of the images is 20 ns. PL intensity is represented by a colour scale normalized for each image. Second column: time-integrated PL spectra for three distinct 0.2 ns time-windows. Third column: PL transient obtained by integrating the PL signal for different 0.2 nm wavelength windows.

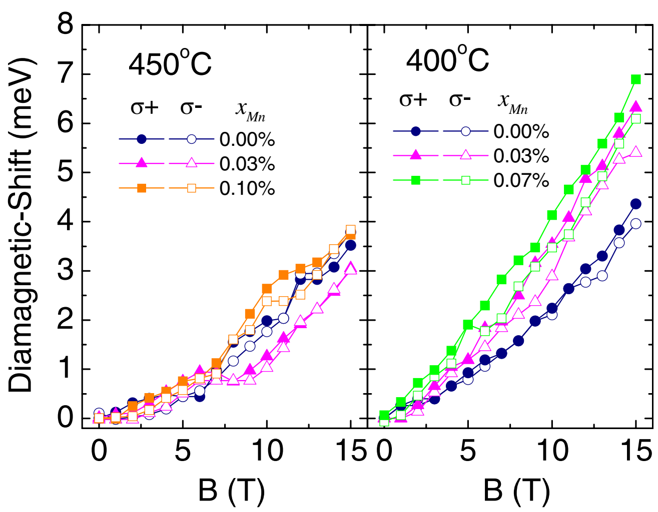

Figure 3. Diamagnetic shift versus magnetic field for the set of samples with different Mn concentrations and growth temperatures.

occur. On the other hand, the incorporation of Mn could increase the interface roughness. But this would result in an inverted effect that does not explain our results. In fact, the samples with larger Mn concentrations show an enhanced diamagnetic shift which implies less localized carriers, and should therefore correspond to a reduced interface roughness. In contrast, Mn incorporation should cause an increase in the density of defects in the QWs and this should be reflected in a stronger localization of the carriers. An alternative explanation for this unexpected increase in the magneto-shift with the Mn concentration in the QW is that the magnetic moment of the Mn ions may be aligned by the external magnetic field, acting as an effective additional component of the total magnetic field experienced by the carriers. We also observe that the Zeeman-splitting energy is relatively small, but this parameter tends to increase with the Mn concentration, going from ∼0 for the samples without Mn to ∼0.7 meV for the sample with the largest effective Mn concentration ($x_{\mathrm{Mn}} = 0.07\%$ grown at 400 °C) at 15 T, revealing a Mn effect on the effective g-factor of the GaMnAs alloy. In order to investigate the effect of Mn incorporation on the QWs, we focus on the results of the sample that has the largest Mn concentration: the sample grown at 400 °C with $x_{\mathrm{Mn}} = 0.07\%$. The sample grown at 400 °C with $x_{\mathrm{Mn}} = 0$ will be used as a reference sample. The sample grown at 450 °C with $x_{\mathrm{Mn}} = 0$ was also investigated because it was considered as a better reference sample with relatively much less background Mn impurities. This is due to the fact that any residual Mn due to memory effects in the MBE system will be incorporated less significantly at this higher growth temperature. Figure 4 shows the circularly polarized PL emission from those three samples under 15 T and different excitation intensities. The spectra of the sample without Mn grown at 450 °C comprise two peaks that are separated by ∼2.5–3.0 meV. Even though the peaks are unresolved, the fact that the band comprises two distinct transitions becomes evident because their relative intensities vary significantly with the excitation intensity and they present different circular polarizations. The low-energy peak dominates the spectra at high excitation intensities and presents a positive polarization, while the high-energy peak is relatively stronger at low excitation intensities and presents a negative polarization. Based on their energy separation, a possible explanation for those peaks could be excitonic complexes, such as excitons and trions, with distinct effective g-factors. It is noticed that the low-energy peaks dominate under high excitation intensities. This is a rather unusual, and therefore the detailed identification of those peaks becomes more difficult.

For the sample with $x_{\mathrm{Mn}} = 0.0\%$ grown at 400 °C, we no longer discern the presence of the two peaks observed in the sample grown at 450 °C and the QW emission

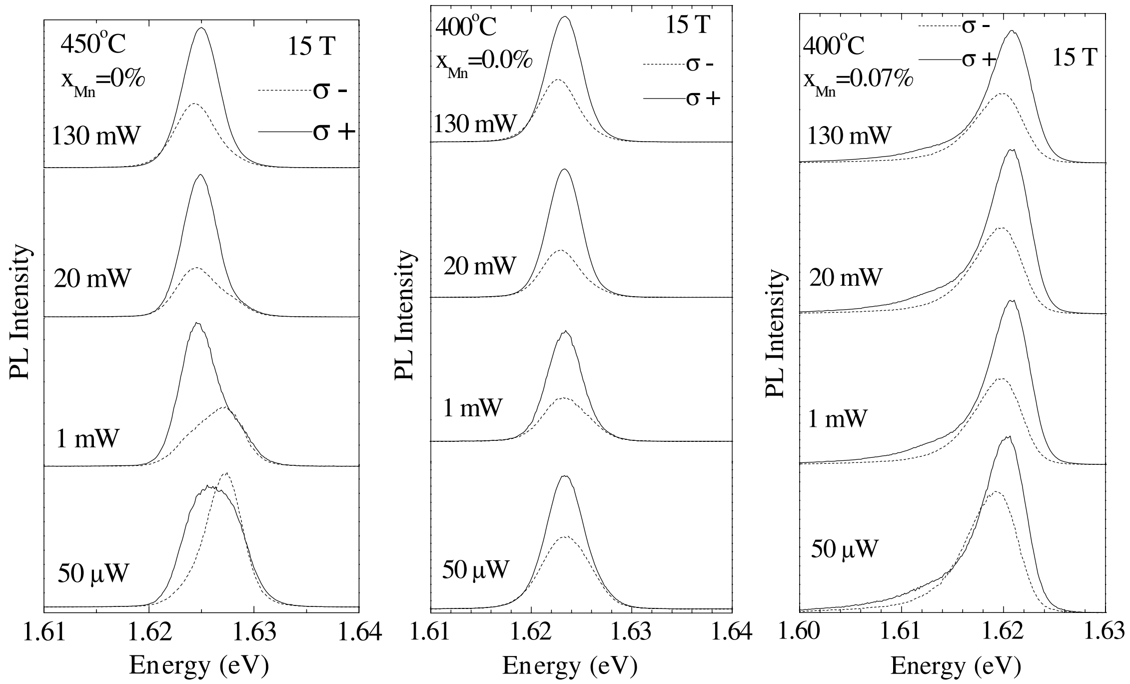

Figure 4. Typical PL spectra as a function of laser power excitation for $B = 15$ T for different Mn concentrations and growth temperatures.

becomes dominated by a single band with positive polarization, suggesting that the emission is equivalent to the low-energy peak observed on the sample grown at 450 °C. This result may be related to a higher residual Mn contamination which could alter the carrier concentration on the QW since the Mn atoms are expected to act preferentially as acceptors creating a hole concentration. This may affect the formation of excitonic complexes reducing the intensity of the high-energy peak. For the sample with $x_{Mn} = 0.07\%$ grown at 400 °C, we observe, in contrast to the samples without Mn, the additional low-energy shoulder that was attributed to a shallow donor associated with interstitial Mn_i [6]. This emission seems to be positively polarized, but as this band is not very well described by a simple Gaussian or Loretzian function, the tentative of extracting a qualitative number for its polarization is prevented by a relative large uncertainty.

Figure 5 presents the polarization degree of the total integrated PL emission as a function of the laser power for the same samples analysed in figure 4. The obvious stronger dependence of the polarization degree under the low-excitation regime for the sample grown at 450 °C without Mn is probably associated with the transition between the two peaks separated by 2.5–3.0 meV with distinct circular polarization degrees. As the laser power is decreased, the high-energy peak that shows a negative polarization becomes relatively stronger; this effect must contribute to a decreased polarization degree of the total PL emission for this sample. The other samples, for which these two peaks are not evident, present a smoother dependence of the polarization degree with the laser power. We note that based on the PL spectra from figure 4, the relation between the PL intensity of the QW peak and the Mn shoulder remains mainly constant with the laser power and they seem to present similar polarization degrees. Therefore the presence of the shoulder does not seem to significantly affect the total

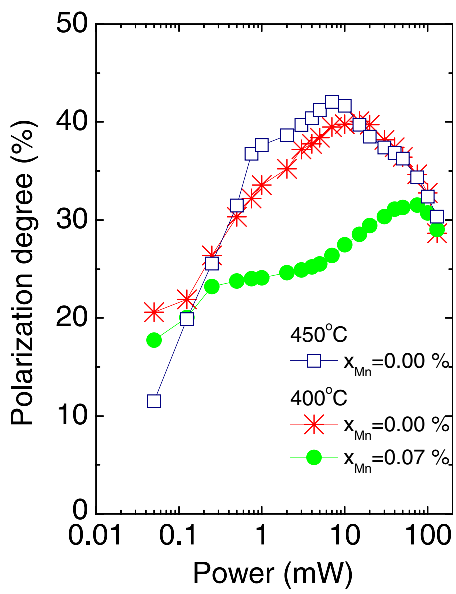

Figure 5. Polarization degree as a function of laser power for $B = 15$ T for different Mn concentrations.

polarization presented in figure 5. In general, it was observed that all the samples present a polarization weakening under higher excitation intensities, which is expected due to a band filling effect. We note that for intermediate intensities the sample with Mn shows a markedly smaller polarization degree. In the analysis of this result we must also consider the fact that this sample presents a slightly larger Zeeman splitting as compared with the samples without Mn (see figure 3). It is worth pointing that the basically zero excitonic g-factor observed for the samples without Mn do not result in a null polarization degree. This can be understood as a result of g-factors for electrons and holes with similar modulus and

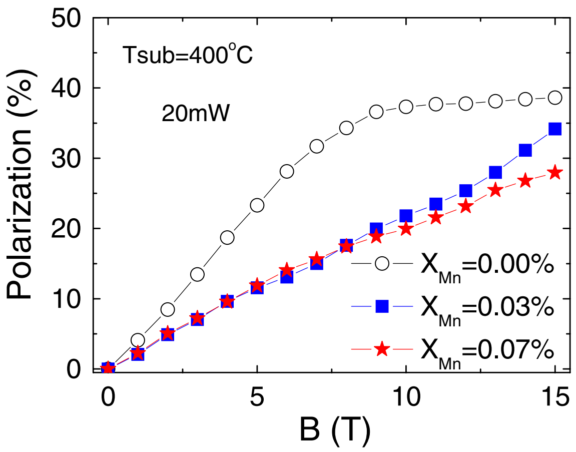

Figure 6. Polarization degree as a function of magnetic field for different Mn concentrations.

opposite signs and the presence of a majority density of a given carrier in the QW. The majority carriers are probably holes, as they are generated by the Mn acceptors that are expected to be present as a residual impurity even on the samples nominally without Mn. Therefore, the smaller polarization degree observed for the sample with Mn seems to indicate a reduction in the electronic g-factor in such a way as the excitonic g-factor is no longer null (different modulus for electron and hole g-factors). This model can explain the observation of a reduced polarization degree (due to the reduced splitting of the minority carriers) associated with an increased excitonic g-factor.

The QW polarization degree as a function of the magnetic field has also been investigated, as shown in figure 6. The results are consistent with the reduced polarization degree observed for the samples with Mn at high magnetic fields (15 T). It is expected that due to the magnetic properties of the GaMnAs, the polarization degree should present a Brillouin-function dependence with the magnetic field. Surprisingly, an almost linear behaviour is observed for the samples with Mn, in contrast to the sample without Mn that does present a Brillouin-like behaviour. This result shows that the polarization-degree dependence on the magnetic field is not a simple reflection of the Mn incorporation and it must be dominated by other effects such as the complex valence band structure. The origin of these anomalies is still under investigation.

4. Conclusion

In summary, the optical properties of GaMnAs QWs with low Mn concentrations grown at high substrate temperatures have been investigated. The results show that these samples present an anomalous PL behaviour. Particularly, an unusual long PL time decay is observed for the samples without Mn. This decay time is reduced by the Mn incorporation, probably due to the increasing density of non-radiative centres introduced by the Mn impurities. We also observed an increase in both the diamagnetic shift and the excitonic g-factor and a reduction in the QW polarization degree for the samples with increasing Mn concentration at high magnetic fields. In addition, the QW polarization degree has showed a linear behaviour for the samples with higher Mn content as a function of magnetic field although a Brillouin-function field dependence was expected.

Acknowledgments

The financial support from the Brazilian Agencies FAPESP, CAPES, CNPq, the UK Engineering and Physical Sciences Research Council are gratefully acknowledged. We thank M Cukr and J Oswald (Institute of Physics of ASCR, Prague, Czech Republic) for useful discussions and sample growth.

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