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T. Lebihen, E. Deleporte, M. Voos, S. Guha, H. Munekata. Optical study of a ZnSe/Zn_{0.73}Mn_{0.27}Se heterostructure. Journal de Physique IV Proceedings, 1993, 03 (C5), pp.C5-425-C5-428. 10.1051/jp4:1993591 . jpa-00251678

HAL Id: jpa-00251678

<https://hal.science/jpa-00251678>

Submitted on 1 Jan 1993

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Optical study of a $\text{ZnSe}/\text{Zn}_{0.73}\text{Mn}_{0.27}\text{Se}$ heterostructure

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Abstract : Photoluminescence, excitation spectroscopy and magneto-photoluminescence experiments are performed up to 5.5T at low temperature (1.7K) in a $\text{ZnSe}/\text{Zn}_{0.73}\text{Mn}_{0.27}\text{Se}$ double quantum well. The experimental results are compared with calculations of the excitonic transitions, available for a type-I and a type-II band structure. These calculations include the strain effects, the giant Zeeman effect in the $(\text{Zn},\text{Mn})\text{Se}$ layers under a magnetic field, the diamagnetic shift of the exciton. We determine the strain state of the heterostructure and find a quite unusual configuration of the band structure: the fundamental optical transitions of the two wells are type-I light hole transitions, while the excited optical transitions are type-II heavy hole transitions.

Among the various II-VI compounds, the ZnSe materials require a particular attention because of their blue emission. As a matter of fact, the first blue laser has been obtained with ZnSe based compounds [1]. The $\text{ZnSe}/(\text{Zn},\text{Mn})\text{Se}$ semimagnetic heterostructures are of particular interest because of their great magneto-optical properties, due to the exchange interaction between the spin of the carriers and the spin of the paramagnetic Mn^{2+} ions located in $(\text{Zn},\text{Mn})\text{Se}$ layers [2]. The so-called giant Zeeman effect can lead to the formation of a spin superlattice structure in which the carriers can be separated and spatially localized according to their spin [3,4]. Moreover, large strains, due to the great lattice mismatch between the binary and the ternary alloys, are present in the structure and strongly modify its band profile. From a comparison between photoluminescence and magneto-photoluminescence experiments and calculations, we demonstrate here that these strains can lead to a quite unusual configuration: the fundamental optical transition is a type-I light hole exciton (electrons and light holes are localized in the same layers), the heavy hole transitions are excited states and correspond to a type-II situation (electrons and heavy holes are localized in adjacent layers).

The heterostructure has been grown by molecular beam epitaxy along the (100) axis and consists in two ZnSe quantum wells whose thicknesses are 53Å and 106Å respectively, separated by a 350Å thick $\text{Zn}_{0.73}\text{Mn}_{0.27}\text{Se}$ layer. The double quantum well is grown on a 1200Å $\text{Zn}_{0.73}\text{Mn}_{0.27}\text{Se}$ layer deposited on a 9700Å thick ZnSe buffer layer. The cap layer is constituted with a 350Å $\text{Zn}_{0.73}\text{Mn}_{0.27}\text{Se}$ layer. The substrate is GaAs.

Figure 1 shows the photoluminescence (the excitation wavelength is 4000Å) and excitation spectra at low temperature (1.7K), performed with a high pressure Xe lamp, followed by a HR320 Jobin-Yvon monochromator. Because the Xe lamp spectrum is not completely flat between 4600Å and 4000Å, the intensity of the excitation spectra shown in figure 1 is normalized, by making the ratio between the photoluminescence signal and the lamp intensity. Two strong photoluminescence lines are clearly seen at 2775 meV (PL_W) and at 2805 meV (PL_T). The excitation spectrum of (PL_W) (fig 1a) exhibits a strong line (X_W) at 2778 meV and a much weaker one (Y_W) at 2817 meV. The excitation spectrum of (PL_T) (fig 1b) shows the same features: a strong line (X_T) at 2816 meV and a much weaker one (Y_T) at 2840 meV. (PL_W) and (X_W) are attributed to the fundamental state of the wide well and (PL_T) and (X_T) are attributed to the

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Figure 1: Photoluminescence spectrum (solid curve, excitation energy: 3100 meV), photoluminescence excitation spectra (dashed curves) (a) of the PL_W line (detection energy: 2769 meV), (b) of the PL_T line (detection energy: 2805 meV), for a 106Å/350Å/53Å ZnSe/Zn_{0.73}Mn_{0.27}Se double quantum well. The spectra have been performed with a high pressure Xe lamp, without magnetic field, at low temperature (1.7K).

Figure 2: Photoluminescence spectrum of the double quantum well, performed with the 363nm line of an Ar⁺ laser, without magnetic field, at low temperature (1.7K).

fundamental state of the thinner well. The small Stokes-shift between the (PL_W) and (X_W) or (PL_T) and (X_T), is an indication of the relatively good quality of the sample.

Figure 2 shows the photoluminescence spectrum obtained with the 363nm line of an Ar⁺ laser. The incident beam power is much larger in this case. The two strong lines (PL_W) and (PL_T) are still present at the same energies, but at higher energy, two weak peaks appear: (PL_B) at 2942 meV corresponds to the Zn_{0.73}Mn_{0.27}Se photoluminescence and (PL'_T) is at the same energy than (Y_T) in figure 1b. In fact, it is possible to see excited transitions in the photoluminescence spectrum because of the large electronic temperature.

To interpret these experimental data, we have performed a calculation of the transition energies, which takes into account the strain effects resulting from the lattice mismatch and the excitonic effects.

The influence of the strain on the band structure has been calculated, using the elastic constants and deformation potentiels given in ref. [5,6]. If the 1200Å Zn_{0.73}Mn_{0.27}Se layer, grown on the ZnSe buffer layer, is not fully relaxed, the barrier layers will be strained. We take as a parameter the residual strain in the barrier: $\epsilon_{\text{bar}} = \frac{a - a^0(x)}{a}$ where a is the mean parameter in the strained layer, and $a^0(x) = 5.666 + 0.268 \cdot x$ Å [7] is the strain-free lattice constant of Zn_{1-x}Mn_xSe. We assume an elastic accommodation of the lattice parameter of the two ZnSe wells with the barrier one.

As an example we have represented in figure 3 the band extrema structure for two extreme situations: (a) the barrier layer fully relaxed, (b) the barrier layer fully strained by the ZnSe buffer. The percentage of the strain-free gap difference which is in the valence band, called the relative valence band offset q_v^0 , is assumed to be 10%. In these two cases, the band structure is type-I for the light holes, type-II for the heavy holes.

Figure 3: Potential profile along the growth axis without strain (solid line) and with strain for the conduction band (dotted line), the valence band for the heavy holes (dashed-dotted line) and for the light holes (dashed line). The assumed relative strain-free valence band offset is $q_v^0 = 10\%$. Two cases are represented: (a) the barrier layer is fully relaxed $\epsilon_{\text{bar}} = 0$, (b) the barrier layer is fully strained by the ZnSe buffer: $\epsilon_{\text{bar}} = -0.0128$.

Knowing the band profile, a variational calculation of the excitonic transitions, which takes into account the possible type-II nature of the band profile has been performed [8]. The effective masses and the dielectric constant are taken from ref. [9], the energy gap of $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ is taken from ref. [10]. For a type-II band structure, the coulombic interaction creates two interface excitons [8]. The thinner is the ZnSe well, the stronger is the interface exciton, because of the larger penetration of the electron wavefunction in the barrier. For a type-I band structure, we have checked that the $350\text{\AA}$ $\text{Zn}_{0.73}\text{Mn}_{0.27}\text{Se}$ barrier was large enough to avoid the coupling between the two wells. The light hole transitions are found to be very sensitive on the value of ϵ_{bar} , as it can be understood in figure 3, and independent of q_v^0 . On the contrary, the heavy hole transitions are not very sensitive to ϵ_{bar} but depends strongly on q_v^0 (at least for $q_v^0 < 15\%$, for which the band structure is type-II).

From the comparison between the experimental and theoretical results, we deduce that the fundamental states (X_W) and (X_T) of the two quantum wells are light hole transitions and that the excited transitions (Y_W) and (Y_T) are heavy hole transitions. From the energy position of (X_W) and (X_T), we find that the (Zn,Mn)Se layers are compressed: $\epsilon_{\text{bar}} = -5.5 \cdot 10^{-3}$. As the light hole transitions are not sensitive to the relative band offset, this value of the strain is not dependant on q_v^0 . The experimental energy position of the heavy hole transitions is consistent with $q_v^0 = 10\%$.

The magneto-photoluminescence experiments can provide an affirmation of our precedent findings, because these experiments allow us to identify the nature of the transitions. In fact, the magnetic field induced variation of the energy position of the lines originates from the giant Zeeman effect present in the $\text{Zn}_{0.73}\text{Mn}_{0.27}\text{Se}$ layers [2], which is three times larger for the heavy holes than for the light holes. The calculation of the excitonic transitions, taking into account the Zeeman splitting of the barrier band edges and the diamagnetic shift of the exciton like in ref [8] shows that the magnetic field induced (up to 5T) redshift of the light hole transitions is a few meV at the most, while heavy hole transitions redshift is at least 10 meV.

The magneto-photoluminescence experiments have been performed at low temperature (1.7K) with a superconducting magnet up to 5.5T, in the Faraday configuration, the laser (363 nm line of the Ar^+ laser) ingoing and the photoluminescence outgoing lights were collected by a multi-fiber-optic guide. Figure 4 shows the energy position of the (PL_W), (PL_T), (PL_T'), (PL_B) lines versus the magnetic field. The (PL_T') line is visible only up to 1.5T, above this value, (PL_T') disappears in the strong (PL_T) line.

Figure 4: Magnetic field induced variation of the (PL_W), (PL_T), (PL_T') and (PL_B) photoluminescence lines, up to 5.5T, at low temperature 1.7K

Firstly, the magnetic field induced redshift of the barrier photoluminescence (PL_B), which is 30 meV for 5.5T and has the look of a Brillouin function [2], is consistent with the redshift of a heavy hole transition. This confirms the heavy hole character of the fundamental transition in the barrier and thus the negative sign of ϵ_{bar} . Secondly, the (PL_W) line does not move at all with the magnetic field and (PL_T) line exhibits a small redshift of 5 meV up to 5.5T. The small observed redshifts are consistent with the light hole character of the fundamental states of the two wells. The (PL_T) variation is larger than the (PL_W) variation, because the hole wavefunction of the thinner well has a larger probability density in the barrier than the one of the wide well. Finally, the fact that the (PL_T') line redshift is quite parallel to the barrier photoluminescence redshift, at least up to 1.5T, confirms the heavy hole character of this transition and the large penetration of the hole wavefunction in the (Zn,Mn)Se layer, indicating the type-II character of the band structure for the heavy holes. Moreover the weak luminescence intensity of the heavy hole transitions (Y_W) and (Y_T) in figure 1 is an additional proof of the type-II character of the heavy hole band structure.

In conclusion, magneto-photoluminescence experiments have allowed us to identify the nature of the optical transitions observed in the photoluminescence and excitation spectra at $B=0T$. From the comparison between the experimental results and the calculations, an estimation of the strain effects in the structure has been performed: the $1200\text{\AA}$ $Zn_{0.73}Mn_{0.27}Se$ layer is partially relaxed on ZnSe buffer layer, the (Zn,Mn)Se barrier layers are compressed. We have found that the fundamental optical transitions of our two uncoupled wells are type-I light hole excitons. The heavy hole transitions are excited transitions and correspond to a type-II band structure. From these results, we have obtained an estimation of the relative valence band offset: $q_v^0 = 10\%$. This value of q_v^0 is consistent with the small values usually found in the literature [3,4]. As the magnetic field induced redshift is much larger for the heavy hole transitions than for the light hole transitions, we expect that the heavy hole transitions will become the fundamental excitonic states for a certain magnetic field. We have to perform magneto-photoluminescence up to a higher field, in order to confirm this phenomenon.

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