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Magnetic field-induced type-I type-II transition in (ZnMn)Se/(ZnBe)Se spin superlattices

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Abstract. We have performed a magneto-optical study on spin superlattice structures fabricated on the basis of (ZnMn)Se/(ZnBe)Se. The band alignment of a $Zn_{0.91}Mn_{0.09}Se$/ $Zn_{0.972}Be_{0.028}Se$ superlattice was found to undergo a type-I to type-II transition for one of the exciton spin component. This spin superlattice formation manfests itself in an asymmetric Zeeman splitting of a spatially direct exciton resulting from a spin dependent change of the exciton binding energy. A pronounced broadening of the spatially direct exciton in reflectance and photoluminescence excitation spectra was observed when scattering in the (ZnBe)Se barriers becomes possible.

Introduction

The use of dilluted magnetic semiconductors (DMS) in nanostructures has brought a lot of new and interesting phenomena in the physics of low dimensional systems. In particular the ablity of tuning confining potentials for carriers by applying external magnetic fields which can lead to the formation of a spin superlattice has contributed in understanding the effect of quantum confinement on exciton states both in experiment and theory. The peculiar magnetic properties of DMSs stem from a strong $s, p{-}d$ exchange interaction between the electron and hole states and the localized $3d$-states of the magnetic ions.

Here we have applicated (ZnBe)Se as a new non-magnetic barrier material for (ZnMn)Se and present first results of optical studies on (ZnMn)Se/(ZnBe)Se multi quantum wells (MQWs) as a novel representative for spin superlattices.

1 Experimentals

The sample under investigation was grown by molecular beam epitaxy (MBE) in a Riber system on (001)-oriented GaAs substrates. The multi quantum well (MQW) consists of 20 periods of alternating 100 Å-thick layers of $Zn_{0.91}Mn_{0.09}Se$ and $Zn_{0.972}Be_{0.028}Se$.

Optical spectra were taken at sample temperatures of 1.7 K (pumped liquid helium) and 8 K. Magnetic fields up to 7.5 T, generated by a superconducting split-coil solenoid, were applied parallel to the growth axis (Faraday geometry). An Ar-ion laser served as the pump source for a tunable dye-laser (Stilben 3) used in photo luminescence excitation (PLE) experiments. For reflectance measurements a halogen lamp was applied. The circular polarized signal was analyzed by a 1-m monochromator and detected with a charged-coupled-device (CCD) or a cooled photomultiplier tube followed by a photon counting system.

2 Results and discussion

2.1 Photoluminescence studies

In Fig. 1(a) we present PLE spectra of the $Zn_{0.91}Mn_{0.09}Se$/ $Zn_{0.972}Be_{0.028}Se$ MQW taken at 1.7 K for different magentic field strengths detected under σ^--polarized excitation. The strongest feature observed in the zero field spectrum at 2.83 eV is attributed to the 1s-state of the e1−hh1 exciton (denoted by X_{hh1}) in the $Zn_{0.91}Mn_{0.09}Se$ wells. By increasing the magnetic field the high energy spin-state of the X_{hh1}-exciton energetically meets with the barrier exciton at a critical magnetic field of $B_0 = 1.5\,\mathrm{T}$. In the field range $B > B_0$ for which the ground state is taken over by the barrier exciton the further energy shift is accompanied by a rapid decrease of the absorption-maximum intensity. Fig. 1(b) shows the PLE peak positions of the various interband transitions as a function of magnetic field. We may point out that a perturbative approach which uses fixed single-particle wave functions to determine exciton binding energies is not suitable for this MQW with small band offsets. In particular the weak carrier confinement which is in the order of the exciton binding energy does not allow to treat excitonic effects as a perturbation and requires a gerneralized variational exciton wave function for an iterative numerical solution. Thus we calculated the Zeeman splitting of free electron and heavy hole (hh) states and used the constant exciton binding energy of 20 meV for all magnetic fields to meet experimental energies [solid lines in Fig. 1(b)].

The total zero-field band offset for this structure has been calculated to 34 meV using the alloy dependence of the $Zn_{1-x}Mn_xSe$ and $Zn_{1-x}Be_xSe$ band gap. Whereas the zero-field type-I band alignment is maintained for the $(-1/2, -3/2)$ exciton transition (σ^+-polarized) the effect of a magnetic field for $m_s = +1/2$-electrons and $m_j = +3/2$-holes is to decrease

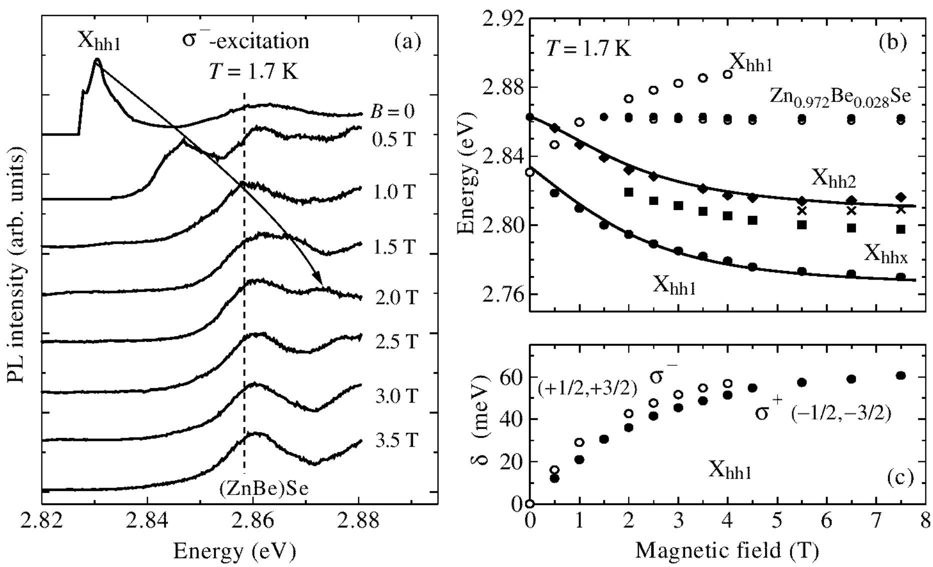

Fig. 1. (a) PLE spectra of a $Zn_{0.91}Mn_{0.09}Se/Zn_{0.972}Be_{0.028}Se$ MQW measured under σ^--excitation at 1.7 K. (b) Exciton transition energies as a function of magnetic field (σ^+: closed symbols, σ^-: open symbols). (c) Relative X_{hh1} spin splitting $\delta E = |E(0) - E(B)|$.

the respective barrier heights (σ^--polarized). At sufficient magnetic fields in the range $0 < B < B_0$ (about 0.3 T) the small valence band discontinuity is overcompensated by the heavy hole Zeeman splitting. However for a small negative valence band offset the Coulomb potential generated by the confined electrons keep the $m_j = +3/2$ holes localized in the (ZnMn)Se layers. By further increasing the magnetic field the band alignment becomes effectively type-II if the hole spin-splitting in addition compensates the exciton binding energy. It has been shown both experimentally [1, 2] and theoretically [3] that a spatially direct exciton state can be formed besides of an indirect one which is composed of an electron localized in the (ZnMn)Se layer and a hole state in the non-magnetic barrier. It is appropriate to regard the direct exciton as a metastable state as its energy lies above the energy of the indirect one. The oszillator strength (i. e. absorption coefficient) of the indirect state is expected to be not sufficient to give a signal in our PLE measurements. A clear indication for the spin dependent confinement is given by the asymmetric spin splitting $\delta E = |E(0) - E(B)|$ of the metastable X_{hh1}-exciton which differs for the $(-1/2, -3/2)$-and for the $(+1/2, +3/2)$-transition shown in Fig. 1(c). Since the Zeeman splitting of band edges is symmetric the difference in ΔE of about 7 meV (3 T) results from a magnetic field dependent exciton binding energy which increases for the lower and decreases for the upper Zeeman branch mainly caused by changes of the hole confinement.

We explain the experimentally observed weakening in the PLE maximum intensity of the (+1/2,+3/2)-transition by the broadening of the spatially direct exciton state. In the regime of a type-II band alignment the hole from the (ZnMn)Se layer can be scattered into the (ZnBe)Se barrier by the emission of acoustical phonons. The mechanism corresponds to the scattering of a metastable exciton into a stable exciton. Additional scattering channels become available when the energy of the metastable exciton is shifted above the energy of the barrier exciton ($B > B_0$) and hence the well exciton as a whole can scatter into exciton states of the non-magnetic barriers. For the MQW under study both scattering channes are expected to get involved in the same magnetic field range. By accounting 20 meV for the Coulomb potential (about the exciton binding energy) we estimated the band alignment to get effectively type-II at $B = 1.3$ T. This field almost coincides with $B_0 = 1.5$ T for which the energies of the barrier and well exciton are equal. The influence of scattering on the metastable exciton is further investigated by reflectance measurement. Moreover by means of reflectance we exclude possible contributions of relaxation dynamics that may differ PLE spectra from absorption spectra.

2.2 *Reflectivity studies*

According to the temperature dependence of the Zeeman splitting it is expected with raised sample temperature to shift the critical field B_0 to higher values. We made use of this dependence to clearly identify the situation of resonant barrier and well energy in reflectance measurements. Two series of σ^--polarized spectra taken at 1.7 K and 8 K are given in Fig. 2(a) and (b). The temperature of 8 K was chosen low enough to avoid changes of the band gap or thermal broadening by exciton-phonon-interaction. In accordance with the energetic position in PLE spectra the sharp feature marked in the low temperature reflectance spectra ($B = 0$) in Fig. 2(a) is the X_{hh1}-exciton resonance. The magnetic field of 1.5 T beyond which the spectra start to be drastically altered coinsides with the critical field B_0. As previously argued the similar behaviour is observed for the 8 K-spectra shown in Fig. 2(b) at a higher critical field of $B_0(8\,\mathrm{K}) \approx 4.5$ T. In fact in the range of X_{hh1}-exciton energies taken from PLE which are labeled by arrows in Fig. 2(a) no significant resonance is observable for $B > B_0$. For both temperatures the disappearence of the X_{hh1}-resonance

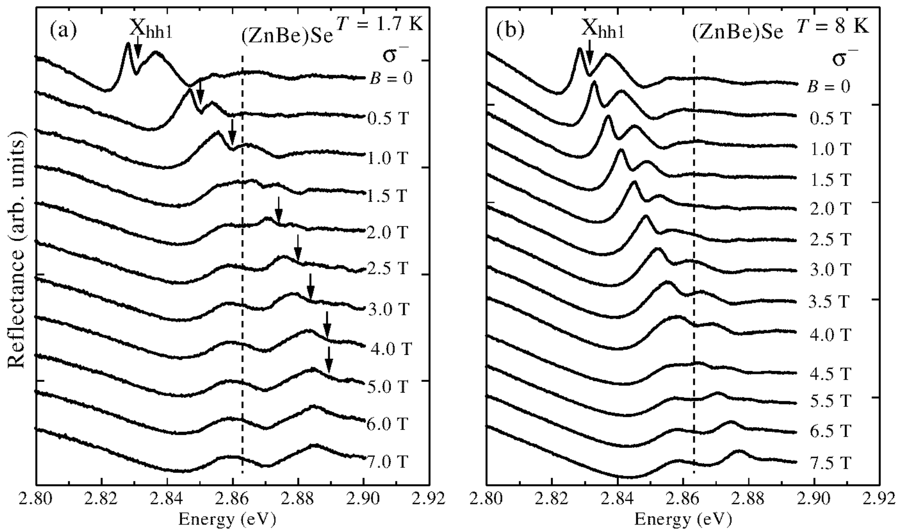

Fig. 2. σ^--polarized reflectance spectra of a $Zn_{0.91}Mn_{0.09}Se/Zn_{0.972}Be_{0.028}Se$ MQW taken a temperatures of 1.7 K (a) and 8 K (b). Arrows indicate X_{hh1}-energies determined from PLE spectra.

at $B > B_0$ is obvious due to exciton scattering into the (ZnBe)Se barriers. However the resonance keeps pronounced in σ^+-polarized spectra (not discussed here).

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