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Interface exciton magnetic polaron in ZnSe/$Zn_{1-x}Mn_xSe$ quantum-well structures

V. V. Rossin,* J. Puls, and F. Henneberger
Humboldt-Universität zu Berlin, Institut für Physik, Invalidenstrasse 110, Berlin 10115, Germany

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A study of cw photoluminescence and photoluminescence excitation spectra of ZnSe/$Zn_{1-x}Mn_xSe$ quantum wells with semimagnetic barriers in a magnetic field clarifies the scenario of magnetic polaron formation for the situation when the light-hole exciton is the electronic ground state at zero magnetic field. It is shown that magnetic polarons are formed by heavy-hole excitons when they become the lowest exciton state at a certain magnetic field due to the giant Zeeman splitting. The magnetic polaron is concluded to be localized at the interface which is favored by a magnetic-field-induced type-I–type-II transition for heavy holes.

In semimagnetic semiconductors, the strong exchange interaction between carriers and magnetic ions gives rise to unique magneto-optical properties.[1] One of the interesting features is the formation of a magnetic polaron (MP)—an alignment of the magnetic ion spins caused by the presence of a carrier which, in turn, lowers the energy of the latter.[2] In principle, reduction of the dimensionality favors the MP formation.[3,4] A system of particular recent interest is a nonmagnetic quantum well (QW) between magnetic barriers. Here, a carrier excited in the QW may lower its energy by reorganizing its wave function in such a way that the exchange with the barrier ions becomes larger. First, in thin QW's, the in-plane motion undergoes autolocalization within the polaron radius (quasi-two-dimensional MP).[4,5] Second, in sufficiently thick QW's with low barriers, the wave function for the motion along the QW axis shifts towards the heterointerface, increasing the penetration into the barrier (interface MP).[6-8] Both cases were experimentally evidenced in CdTe/$Cd_{1-x}Mn_xTe$ QW's.[9,10] However, a recent self-consistent calculation[4] provides that the criteria required for stable free MP's are not met by the $Cd_{1-x}Mn_xTe$ system. This contradiction between experiment and theory is removed when the necessary in-plane primary localization is produced by a mechanism other than the exchange coupling.[5-7] In a QW, in-plane localization occurs inherently as a result of potential fluctuations (interface roughness, alloy disorder, etc.).

The present paper is addressed to the formation of excitonic MP's in ZnSe/$Zn_{1-x}Mn_xSe$ QW's. These structures have smaller band offsets than the $Cd_{1-x}Mn_xTe$ system and thus favor interface localization. Recently,[11] the observation of a MP was reported on ZnSe/$Zn_{1-x}Mn_xSe$ superlattices with small Mn concentration and thus small confinement. In this work we study QW's with thick barriers and high Mn concentration. Moreover, as a specific feature, the tensile in-plane strain causes the light-hole state of $J_h = \pm\frac{1}{2}$ to be that of lowest energy. Unlike the heavy-hole state ($J_h = \pm\frac{3}{2}$), which is magnetically active only along the QW axis, the light hole exhibits a magnetic moment in arbitrary direction with a maximum along the heterointerface. Thus the MP formation process and the related spin alignment are expected to be quite different for these states, particularly when an external magnetic field is applied.[12]

The ZnSe/$Zn_{1-x}Mn_xSe$ QW structures were grown by molecular-beam epitaxy (MBE) on (001) GaAs substrates. The structure studied here consisted of a 0.7-μm-thick $Zn_{0.8}Mn_{0.2}Se$ layer and five ZnSe QW's of 5 nm thickness separated by 85-nm-wide $Zn_{0.8}Mn_{0.2}Se$ barriers. The circularly polarized cw photoluminescence (PL) and PL excitation (PLE) spectra were studied at temperatures from 1.6 to 20 K in magnetic fields up to 9 T oriented parallel and perpendicular to the growth direction (respectively, Faraday or Voigt geometry).

PL spectra at above-barrier excitation and PLE spectra are shown in Fig. 1 for different magnetic fields. At $T = 1.6$ K and zero magnetic field, the PL spectrum exhibits the light-

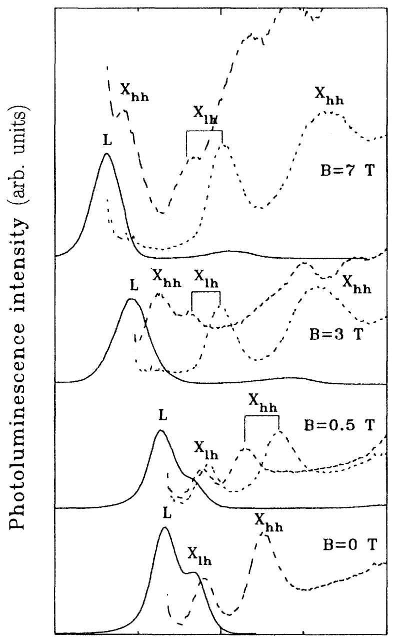

FIG. 1. PL (solid lines) and circular polarized PLE spectra (dashed lines, σ^+; dotted lines, σ^-) detected at the L line at different magnetic fields.

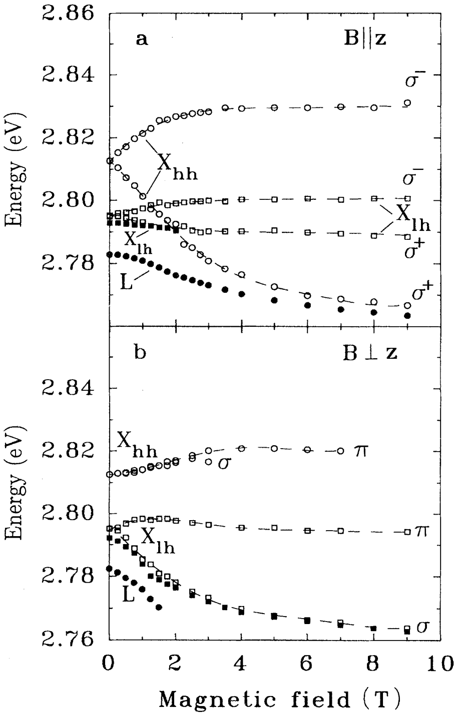

FIG. 2. Energy shifts of PLE (open symbols) and PL (filled symbols) peaks in a magnetic field in Faraday (a) and Voigt (b) geometries. Lines are drawn to guide the eye.

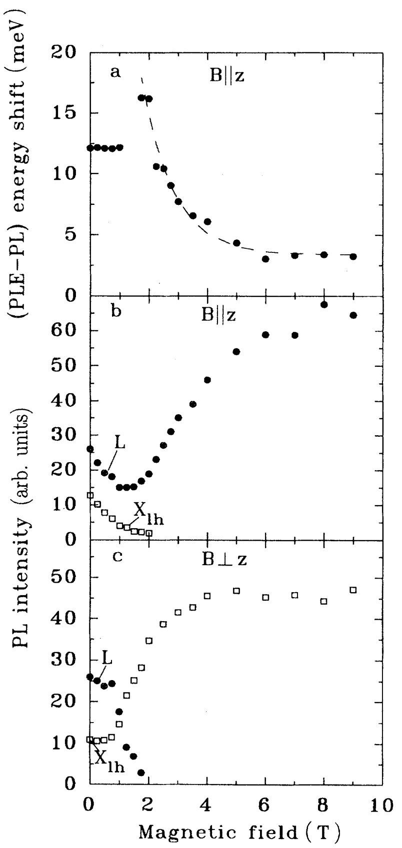

FIG. 3. Magnetic field dependencies of the energy separation between the L line and lowest-energy PLE peak (a), PL intensities of the L line (filled circles), and X_{lh} (squares) (b) and (c). Faraday geometry (a) and (b); Voigt geometry (c). Line is drawn to guide the eye.

hole exciton line (X_{lh}) and a second structure labeled L about 10 meV below X_{lh} (Fig. 1, lowest spectra). The PLE spectrum detected at the L line displays light- and heavy-hole exciton lines (X_{lh} and X_{hh}) with an energy separation of 18 meV. The Stokes shift between PL and PLE for X_{lh} is 2.3 meV. No indication of the L line is seen in the PLE spectrum. Similar low-energy features were previously observed and attributed to MP (Ref. 13) and strain effects.[14]

When a magnetic field in Faraday geometry is applied [Figs. 1 and 2(a)], the PLE spectrum reveals a large Zeeman splitting[15] of the X_{hh} state into σ^+- and σ^--polarized components. The splitting of X_{lh} is markedly less pronounced. Eventually, at a critical field B_c of about 2 T, the σ^+ component of X_{hh} crosses the X_{lh} resonance so that the lowest excitation in absorption is now that with $J_h=\frac{3}{2}$. In PL, the low-energy shift of the L line is initially equal to that of X_{lh} but becomes much stronger (19 meV at 9 T) at higher magnetic fields. The shift remains, however, always smaller than that of X_{hh} (σ^+ component) so that the energy separation between the lowest states in PL (L) and PLE (X_{hh}) decreases from 16 meV at 2 T to 3 meV at 9 T [Fig. 3(a)]. The latter value is close to the PL-PLE Stokes shift of the X_{lh} exciton seen in the zero-field spectrum. In the PL spectrum, the X_{lh} emission becomes weaker and disappears at $B>B_c$ [Fig. 3(b)]. The emission yield of the L line behaves nonmonotonically. At first, it becomes weaker similar to X_{lh} but increases again above B_c. Additionally, around B_c an increase of the L-line width is observed. The same overall behavior of the spectra was found at higher temperatures but with less pronounced splittings and an increase of B_c from 2 T at 1.6 K to 6.5 T at 15.3 K.

In Voigt geometry, the characteristic anticrossing behavior of the components emerging from X_{lh} and X_{hh} is observed[15] [Fig. 2(b)]. This indicates the field-induced mixing of those states.[16] Since the observed mixing is small, one can assign in good approximation the energetically lower-lying σ and π components to X_{lh} and very slightly split high-energy components to X_{hh}. In PL, the L line is shifted parallel to the σ component of X_{lh}. However, the dependence of the line intensities on magnetic field is now opposite: the L line is quenched already at $B=2$ T while the exciton line gains intensity until it runs into saturation [Fig. 3(c)]. We emphasize that in both configurations only one line remains in the PL spectrum at fields $B>2$ T. However, the energy shift between the lowest features of PL and PLE in Faraday geometry is much larger than in the Voigt case.

We start the discussion with the situation at zero magnetic field. Though the low-energy L line definitely involves a light hole (it was checked by a PLE measurement using circularly polarized excitation and detection) its assignment to a MP stemming from X_{lh} is ruled out. First, there is a coexistence of L and X_{lh} in PL which would require a fairly complex formation kinetics. Second and more rigorous, we did not observe any decrease of the L-X_{lh} separation for increasing temperature and/or magnetic field. Therefore, we conclude that for the present QW design there is no polaron contribution in the X_{lh} emission that can be separated from the disorder-induced Stokes shift. Indeed, formation of quasi-two-dimensional MP's is suppressed in thick QW's.[9] The criterion for the formation of an interface MP is that the exchange energy at the interface must be larger than the potential barrier.[8] The strain-enhanced offset of the X_{lh} state in the present structure is about 100 meV with a 65-meV portion seen by the hole.[17] An upper limit for the exchange energy is given by the field-induced shift of the PLE lines. It is obvious from the data in Fig. 2 that the interface MP criterion is not fulfilled for X_{lh}. On the other hand, the heavy-hole offset is merely 30 meV (Ref. 17) and, indeed, smaller than the shift of the respective low-energy PLE feature. At zero magnetic field, however, formation of an X_{hh} MP is prevented by the heavy- to light-hole conversion which was demonstrated to occur on a 1-ps time scale,[17] i.e., considerably shorter than MP formation time.[18]

If a magnetic field is applied, the yield of the L line drops down in both field geometries for $B<B_c$. That behavior is typical for a bound exciton state with oppositely oriented spins of the identical particles.[19] As a result, the spin alignment caused by the external field suppresses formation of the complex. On the other hand, the saturation of L observed under increased excitation intensity is much less pronounced than expected for a reasonable density of centers. Therefore, the origin of L for $B<B_c$ is still somewhat vague. In the present context, it is only of importance that L is definitely not related to a MP. The intensity decrease of X_{lh} in Faraday geometry in the same field range is caused by depopulation of the exciton state involved in the σ^+ transition. In this geometry, the X_{lh} ground state ($J_c=-\frac{1}{2}, J_h=\frac{1}{2}$) emits in the direction perpendicular to the QW axis and is not detected in our experimental arrangement. This is changed in Voigt geometry, where the emission along this axis becomes allowed and, consistently, no quenching is observed here.

Beyond the heavy- to light-hole crossover in Faraday geometry, the L line regains intensity. The physical origin behind the line is, however, entirely different from the zero-field transition. In what follows, we argue that, in accordance with the above estimates, formation of a MP related to X_{hh} occurs. First, we recall that only one PL line is seen for $B>B_c$. At $B\approx B_c$, the energy separation of this line from the PLE ground state is distinctly larger than the L-X_{lh} separation at zero field [Fig. 2(a)]. Further increase of B decreases the separation again and, eventually, the zero-field PL-PLE Stokes shift is recovered at about $B=7$ T. That is exactly the behavior anticipated for a MP. At low fields, its formation is promoted by the reduced barrier potential,[8] whereas saturation of the Mn spin alignment at large external fields causes the MP to disappear, since the photoexcited exciton can no longer contribute to the spin polarization. Further evidence for the MP nature of L above B_c is presented in Fig. 4. Characteristically, the energy shift associated with the MP formation must decrease with growing temperature as the available magnetization is reduced. To figure this out, we have plotted in Fig. 4 the energy spacing between the PLE and PL low-energy features versus the Zeeman shift of X_{hh} in PLE (σ^+) for various temperatures. The comparison of data at fixed Zeeman shift cancels out the temperature dependence of the external field action and the expected shrinking of the MP shift is clearly demonstrated.

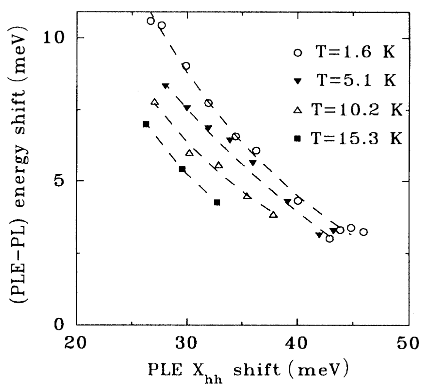

FIG. 4. Dependence of the energy separation between the L line and the σ^+ X_{hh} PLE peak on the Zeeman shift of the σ^+ X_{hh} PLE peak in magnetic field at different temperatures. Lines are drawn to guide the eye.

A standard exchange calculation reproduces the observed Zeeman shifts reasonably well, except the X_{hh} low-energy component, for which the experimental shift is much larger. We correlate this with a type-I–type-II transition for the heavy hole, which should occur at $B\leqslant B_c$. At higher Mn barrier content ($x=27\%$), the type-II heavy-hole offset is even proposed at zero magnetic field for the ZnSe/Zn$_{1-x}$Mn$_x$Se system.[20] Undoubtedly, that transition favors the formation of an interface MP. We believe that, unlike in Ref. 11, the electron-hole Coulomb attraction plays a crucial role and ensures the wave-function overlap required to see clear PLE resonances even in this situation.

The exchange coupling of the light hole is maximal in the plane perpendicular to the QW axis where, in first order, the heavy hole is not affected. Thus the formation of an X_{lh}-related interface MP is at best in Voigt geometry. However, no indication for that is seen in Fig. 2(b). This emphasizes the importance of sufficiently small band offsets for the MP formation.

In conclusion, it has been shown that heavy-hole excitons form MP's in ZnSe/Zn$_{1-x}$Mn$_x$Se QW's. The formation occurs only when an external magnetic field is applied in the direction of the QW axis so that the heavy-hole state becomes that of lowest energy. The formation of the interface MP is promoted by a type-I–type-II transition of the heavy hole. It will be interesting to study other QW designs in this context.

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[*] Permanent address: A. F. Ioffe Physico-Technical Institute, St. Petersburg 194021, Russia.

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