RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 33, NUMBER 4 15 FEBRUARY 1986

Magnetic polaron effects for excitons in narrow CdTe-(Cd,Mn)Te quantum wells

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(Received 25 November 1985)

We present a variational calculation of the energy levels of an exciton confined to a CdTe quantum well by semimagnetic (Cd, Mn)Te potential barriers. The degeneracy of the top of the valence band is lifted by strain and quantum confinement effects, introducing a preferential axis of quantization for the heavy mass $|m_j|=\frac{3}{2}$ doublet. This results in strongly anisotropic magnetic field shifts of the levels. The shifts are greatly enhanced due to the penetration of the wave functions into the barriers, where they are subject to the exchange potential of the polarized Mn spins. Our results are in good qualitative agreement with recent photoluminescence data on CdTe-(Cd,Mn)Te superlattices.

The growth of superlattices of CdTe-(Cd,Mn)Te (Refs. 1-4) and, more recently, ZnSe-(Zn,Mn)Se (Ref. 5) and the investigation of their magneto-optical properties[6-10] has revealed in these materials a variety of magnetic field and temperature-dependent phenomena to match those observed in bulk semimagnetic semiconductors.[11] One of the striking differences between the properties of CdTe-(Cd,Mn)Te superlattices grown along the [111] axis and bulk alloys of (Cd,Mn)Te is the observation of a very strong anisotropy in the red shift of the exciton recombination photoluminescence line.[7-10] At low temperatures the shift depends not only on the intensity of the applied field, but also on its direction with respect to the superlattice growth axis. The effect is much stronger for fields parallel than for fields perpendicular to this axis. Zhang and coworkers[7-10] have argued that strain effects, alloy-concentration fluctuations, and magnetic polaron effects on the almost two-dimensional exciton formed in narrow quantum wells ($r_{\text{exc}} > L_z$) or confined near the interface in wider wells ($r_{\text{exc}} < L_z$) may be responsible for the observed shifts. To date, however, there has been no published theoretical analysis of the behavior of excitons in these structures. Recently, we examined the problem of bound polarons in semimagnetic quantum wells and showed that small valence-band offsets lead to strong polaronic effects.[12]

In this Rapid Communication we examine how the existence of a preferential axis of quantization for the hole spin, a consequence of strain and quantum confinement effects, leads to striking anisotropic properties of the excitons. Excitons in quantum wells have been discussed previously,[13] but without incorporating this feature, which is essential for the understanding of the properties of semimagnetic superlattices. In these materials, the exchange interaction between hole spin and Mn ion spins greatly enhances the magnetic anisotropy, which should also exist, albeit on a much reduced scale, in nonmagnetic superlattices. We formulate a model sufficiently simple to be tractable almost analytically and, at the same time, realistic enough to shed light on the interpretation of the experimental data. Below, we summarize the elements which go into our calculation with brief comments about their limitations wherever appropriate.

(a) We consider a narrow quantum well of CdTe, $L_z = 50$ Å, with perfect interfaces. The band offsets are taken to be $\Delta E_v/\Delta E_c = 0.1/0.9$ and $\Delta E_g = 1.5x$ eV,[14] where x is the Mn concentration.

(b) Strain effects are important due to the lattice mismatch $\Delta a/a = 0.00285x$.[3] Using standard elasticity theory[15] and the deformation potentials measured by Gavini and Cardona,[16] we can estimate the splitting of the top of the valence band. The fourfold degeneracy is lifted, with the $|m_j|=\frac{1}{2}$ doublet lying below the $|m_j|=\frac{3}{2}$ doublet. For $x = 0.25$, assuming that the strain is accommodated elastically, the energy splitting is a sizable 30 meV.

(c) The effective masses are calculated from the valence-band parameters of Lawaetz.[17] We obtain them by neglecting the kinetic coupling between the two strain split doublets. We find the following for $|m_j|=\frac{3}{2}$: $m_y = 0.13m_0$, $m_z = 2.7m_0$; for $|m_j|=\frac{1}{2}$ we find the following: $m_y = 0.35m_0$, $m_z = 0.10m_0$, where y and z are, respectively, orientations parallel and normal to the plane of the interface.

(d) The Mn ions are assumed noninteracting. This is not true, but we know, qualitatively, what to expect from the introduction of Mn-Mn interactions.[18]

(e) We neglect orbital effects of the applied field, i.e., Landau quantization, since these are quantitatively less important than the exchange energy for the description of the energy levels. The latter is treated in the mean-field approximation, as in Ref. 12, with the parameters given in Ref. 19.

(f) We neglect time-dependent polaronic effects by considering an infinitely lived exciton. In light of the time-resolved photoluminescence data,[10] we know that the exciton lifetime and the time needed to establish the polaron are comparable. However, polaronic effects are detected, which indicates that a fair amount of polarization of the Mn spins has been achieved by the time the exciton recombines.

The exciton ground state can be described by a four spinor $(f_3, f_1, f_{-1}, f_{-3})$, which satisfies the following Schrödinger equation:

$$
\begin{aligned}
&(h_e + h_3 + V_c - W_e - 3W_{hz})f_3 + (S - \sqrt{3}W_{hy})f_1 + Tf_{-1} = Ef_3 \;, \\
&(\bar{S} - \sqrt{3}W_{hy})f_3 + (h_e + h_1 + V_c - W_e - W_{hz})f_1 - 2W_{hy}f_{-1} + Tf_{-3} = Ef_1 \;, \\
&\bar{T}f_3 - 2W_{hy}f_1 + (h_e + h_1 + V_c - W_e + W_{hz})f_{-1} - (S + \sqrt{3}W_{hy})f_{-3} = Ef_{-1} \;, \\
&\bar{T}f_1 - (\bar{S} + \sqrt{3}W_{hy})f_{-1} + (h_e + h_3 + V_c - W_e + 3W_{hz})f_{-3} = Ef_{-3} \;.
\end{aligned}
\tag{1}
$$

In Eqs. (1), h_e, h_3, and h_1 describe the kinetic plus barrier potential plus strain energies of the electron, $|m_j| = \frac{3}{2}$ holes and $|m_j| = \frac{1}{2}$ holes, respectively; S and T describe the kinetic coupling between the holes subbands,[19] and are neglected in the following; V_c is the Coulomb potential; W_e, W_{hz}, and W_{hy} are the exchange potentials acting, respectively, on the electron and on the hole, perpendicular and parallel to the interface (the electron spin follows the applied field). The spinor components f_m give the amplitude of the $m/2$ hole state contribution to the wave function. We recall that the axis of quantization is the growth axis of the superlattice.

An examination of Eq. (1) shows that when the external field is applied perpendicular to the interface (i.e., $W_{hy}=0$) it is a good first approximation to set $f_1 = f_{-1} = f_{-3} = 0$. This leads to the single equation

$$(h_e + h_3 + V_c - W_e - 3W_{hz})f = Ef \quad . \tag{2}$$

When the external field is applied parallel to the interface, (i.e., $W_{hy}=0$) we can easily see that, for the ground state, $f_3 = f_{-3}$ and $f_1 = f_{-1}$, which leads to

$$\begin{aligned}(h_e + h_3 + V_c - W_e)f_3 - \sqrt{3}\,W_{hy}f_1 &= Ef_3 \ ,\\ -\sqrt{3}\,W_{hy}f_3 + (h_e + h_1 + V_c - W_e - 2W_{hy})f_1 &= Ef_1 \ .\end{aligned} \tag{3}$$

We remark that, to first order, the $|m_j| = \frac{3}{2}$ doublet is unaffected by the exchange field, although it is perturbed through the coupling to the isotropic $|m_j| = \frac{1}{2}$ doublet.

We now discuss in some detail the variational solution of Eq. (2), taking as trial wave function

$$\begin{aligned}f(\mathbf{r}_e, \mathbf{r}_h) = {} & N\exp(-R/\eta)\exp(-\rho/a)\psi_e(z_e)\psi_h(z_h)\\ & \times \{\cosh[\xi(z_e - z_h)]\}^{-1} \ ,\end{aligned} \tag{4}$$

where N is the normalization factor. In Eq. (4), η and a are, respectively, the exciton center of mass localization radius and relative motion radius in the plane of the interface; ψ_e and ψ_h are, respectively, the electron and hole envelope functions in the direction perpendicular to the interface; ξ measures the perpendicular correlation of the electron and hole motions due to the Coulomb interaction. For trial functions in the perpendicular direction we take

$$\psi(z) = (\sqrt{\alpha}/\sqrt{\pi})\exp[-0.5\alpha^2(z - z_0)^2] \ , \tag{5}$$

with different parameters for electrons and holes.

Before presenting our results, we comment on Eq. (4). It has eight variational parameters and its minimization can be fairly involved. We have made one major assumption in dealing with the $|m_j| = \frac{3}{2}$ case: We take the hole wave function to be "frozen" with $\alpha_h = 0.0688$ Å^{-1} (a value appropriate for the nonmagnetic quantum well) and take z_{0h} as a parameter not subject to minimization. Although this cannot be rigorously justified we take the value $m_z = 2.7m_0$ as a strong indication that the hole is easily localized in the z direction. We have not included in Eq. (1) any explicit localization potentials, but we introduce them implicitly through this assumption. We take $\xi = 0$, which is justifiable for a narrow well. Numerical tests indicate that we may take the electron wave function as independent of the applied field with $\alpha_e = 0.0572$ Å^{-1} and $z_{0e} = 0$ (center of the well). This is so because the barrier potential energy is the dominant term for the electron.

With the above values for α_h and α_e, the total energy of the noninteracting electron-hole pair is 66.5 meV, measured from the gap energy. For comparison, the exact solution of the corresponding quantum-well problems for the electron and the hole yields 63.9 meV. The exciton binding energy, not including polaronic effects, is 19.3 meV, for a dielectric constant $\epsilon = 10$.

In Fig. 1 we show the exciton energy obtained for different values of z_{0h}. In order to interpret this figure, the reader must keep in mind the picture of a hole, created by photon excitation, that binds an electron and recombines before it has time to travel an appreciable distance along the z axis but after it has polarized the Mn spins in the barrier. Excitons formed around holes centered at different distances from the interface have different energies due to the barrier and exchange potentials. Curve (a) corresponds to $B=0$ and $T=1$ K. The behavior of the exciton energy is very similar to that calculated for bound polarons in Ref. 12. The exciton center-of-mass localization radius is essentially infinite ($\gtrsim 200$ Å) for $z_{0h} < 15$ Å and then falls rapidly to about 30 Å for $z_{0h} = 25$ Å (hole at the interface). This is a self-trapped exciton in the plane on the interface. This shows that a "free" magnetic polarion, i.e., not bound to any impurity or defect, may exist in a narrow quantum well. Curve (b) corresponds to $B_z = 20$ kG and $T=1$ K. This is roughly the saturation field in our model at this temperature. There is a rapid shift of the exciton energy which varies from about 6 meV for $z_{0h}=0$ to more than 100 meV for $z_{0h} = 25$ Å, accompanied by a progressive delocalization of the exciton in the plane of the interface. The red shift is strongly temperature dependent, as shown by curve (c), for $B_z = 20$ kG and $T=10$ K. The dashed curve presents the result for a transverse field $B_y = 20$ kG and $T=1$ K. The

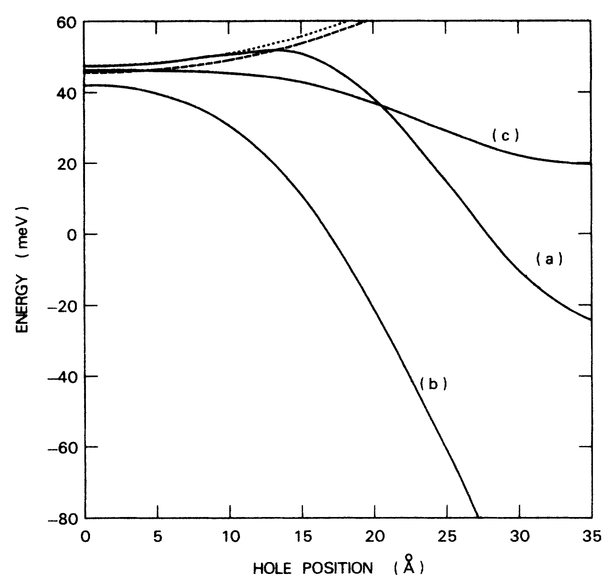

FIG. 1. Calculated exciton–magnetic-polaron energy as a function of the position z_{0h} of the center of the hole wave function in the quantum well. The well center is at $z=0$ and the interface is at $z=25$ Å. The zero of the energy corresponds to the band-gap energy of bulk CdTe in the same strained state as the CdTe layer. Curve (a): $B=0$ and $T=1$ K; curve (b): $B_z=20$ kG and $T=1$ K; curve (c): $B_z=20$ kG and $T=10$ K. Dashed line: $B_y=20$ kG and $T=1$ K. Dotted line: exciton energy with exchange interaction set equal to zero. For details of the parameters, see text.

dotted curve is the result for an exciton in a nonmagnetic quantum well (or infinite spin temperature).

To make qualitative contact with the experimental results, we remark that the exciton recombination probability depends on the overlap of the hole and electron wave functions, which decreases very rapidly as the hole moves away from the center of the well. Thus, only the "dispersion curves" near the center of the well are observable in photoluminescence. A precise calculation of the line shape depends on a knowledge of the hole distribution (in energy and in space), on density-of-states considerations in the sense of Bastard's definition,[20] on the effect of magnetization fluctuations,[21] and on the time dependence of the magnetic polaron formation.

Since W_e is small (about 3 meV at full Mn spin polarization) the exciton ground state is a doublet split by the applied field, with no recombination allowed from the upper state—obtained from Eq. (2) by reversing the sign of W_e. This means that its lifetime τ is strongly affected by applied field (decreases τ) and temperature (increases τ).

We can see from Eq. (3) that the $|m_J|=\frac{1}{2}$ doublet is effectively isotropic and has a red shift which varies rapidly with the applied field. A transverse magnetic field brings it into interaction with the $|m_J|=\frac{3}{2}$ doublet, mixing the states, and pushing the latter down in energy. This is the most likely explanation of the small red shift detected near saturation for B parallel to the interface.[10] If the $|m_J|=\frac{1}{2}$ doublet is the ground state, as we have reasons to expect it to be in ZnSe-(Zn,Mn)Se semimagnetic superlattices,[5] where the strain is of the opposite sign to that in CdTe-(Cd,Mn)Te, the magnetic anisotropy discussed in this work is much reduced. A more complete analysis of Eq. (3) will be presented elsewhere.

It can also be easily seen, within this model, that light emitted along the growth axis is σ_+ and π polarized for applied magnetic fields, respectively, parallel and perpendicular to the same axis.

In conclusion, we have shown that, in semimagnetic quantum wells, a description of the exciton states must include the hole spin anisotropy induced by strain and quantum confinement effects. We can then understand the substantial anisotropy observed with respect to magnetic field orientation in CdTe-(Cd,Mn)Te superlattices in photoluminescence experiments.[7-10]

The author would like to thank Dr. F. Stern and Dr. S. von Molnar for a critical reading of the manuscript and helpful discussions, and Professor A. V. Nurmikko for communicating to him results of his work previous to publication. The partial financial support of the Conselho Nacional de Desenvolvimento Cientifico e Tecnologico (Brasil) is gratefully acknowledged.

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