PHYSICAL REVIEW B VOLUME 52, NUMBER 16 15 OCTOBER 1995-II

Exciton magnetic polarons in short-period CdTe/$Cd_{1-x}Mn_xTe$ superlattices

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(Received 19 June 1995)

The formation of magnetic polarons in short-period semimagnetic CdTe/$Cd_{1-x}Mn_xTe$ superlattices is studied by low-temperature cw and time-resolved photoluminescence and compared to the polaron formation in single quantum wells with the equivalent well width and manganese content. The dependence of the polaron energy on the superlattice period, i.e., quantum-well width, is identical to the dependence on well width in the corresponding single quantum wells. This implies that in the superlattice structures the polaron formation process does not lead to a spatial shift of the center of the hole wave function into the semimagnetic barrier. Temperature and magnetic-field-dependent measurements of the polaron energy reveal a more efficient polaron suppression in superlattices than in single quantum wells. This is attributed to the modification of the magnetic properties in the thin semimagnetic barrier layers of the superlattices. The different mechanisms of magnetic-field-induced polaron suppression in Faraday and Voigt geometries are discussed.

I. INTRODUCTION

The unique magnetic properties of semimagnetic semiconductors, such as $Cd_{1-x}Mn_xTe$, stem from the strong exchange interaction between the carrier spin and the spins of the magnetic Mn ions.[1] For example, localized carriers or excitons ferromagnetically align the spins of the magnetic ions in their vicinity; this is referred to as magnetic polaron (MP) formation.[2] Theoretically, it has been shown that the conditions of primary localization of excitons (or carriers) play a crucial role in this formation process.[3,4] In the case of bound magnetic polarons (BMP's), which are formed from excitons (or carriers) bound to shallow impurities, the Coulomb potential of the impurity center provides the initial localization and MP's are detectable at low Mn content.[5] The situation is considerably different for excitons not bound to impurities. Free MP's are not stable in $Cd_{1-x}Mn_xTe$ and primary localization on the potential of alloy and/or magnetic fluctuations is required. Experimentally, it has been shown that the conditions for the formation of such localized magnetic polarons (LMP's) in thick $Cd_{1-x}Mn_xTe$ epilayers, i.e., in the three-dimensional (3D) case, are fulfilled for Mn content higher than 0.1.[6,7] However, a reduction of the dimensionality of the carrier system from 3D to quasi-2D enhances the stability of LMPs, as theoretically predicted[3,4] and experimentally shown for $Cd_{1-y}Mn_yTe/Cd_{1-x}Mg_xTe$ single quantum well (SQW) structures.[8]

CdTe/$Cd_{1-x}Mn_xTe$ quantum wells (QW's) exhibit type-I band alignment, i.e., both electron and hole wave functions are centered in the CdTe well layers.[9] The valence-band offset (VBO) in these structures has been determined to be about 30% of the total band-gap discontinuity.[10] In superlattices (SL's) the formation of minibands leads to a delocalization of carriers. However, in semimagnetic SL's the exchange interaction may result in the formation of MP's, i.e., magnetically induced localization. A theoretical analysis of the energy spectrum of MP's in CdTe/$Cd_{1-x}Mn_xTe$ SL's has been made in Ref. 11 assuming zero VBO. It was found that the polaron ground state corresponds to a hole wave function centered in the middle of the semimagnetic barrier layer. This was associated with the large magnetic exchange energy gained by polaron formation, which overcomes the strain-induced valence-band offset. The question remains open whether the polaron formation in SL's could overwhelm a considerable VBO of 30%.

Apart from the interest in the intrinsic properties of MP's in SL's, the polaron can be used as a sensitive tool to investigate the modifications of the magnetic properties of the Mn ion system in semiconductor quantum structures. Evidence for such modifications at the interfaces between magnetic and nonmagnetic semiconductors has been reported for single quantum wells (SQW's), where nearest-neighbor Mn spins at the interfaces are missing,[12,13] and SL's, where the formation of large Mn clusters is limited by the barrier thickness.[14]

In this paper we report on cw and time-resolved photoluminescence studies of exciton magnetic polaron formation in CdTe/$Cd_{1-x}Mn_xTe$ short-period superlattices. The dependence of the MP energy on SL period, crystal temperature, and external magnetic field is investigated and compared to the polaron properties in SQW's. Different mechanisms for the magnetic-field suppression of the MP are discussed.

II. EXPERIMENTAL DETAILS

The CdTe/$Cd_{1-x}Mn_xTe$ structures were grown by molecular-beam epitaxy on (100)-oriented CdTe substrates after a 0.6-μm-thick buffer layer of $Cd_{0.76}Mn_{0.24}Te$. SL's with periods (d) of 24, 48, and 60 Å and with a Mn content of 0.24 in the barrier layers have been studied. All SL's have equal thicknesses of the CdTe well (L_z) and the $Cd_{0.76}Mn_{0.24}Te$ barrier (L_B); thus the SL period $d=L_z+L_B=2L_z$. In previous work we have confirmed the formation of minibands in these short-period superlattices through the observation of Bloch-like perpendicular transport.[15] The 24-Å-period SL (20 periods) and the 48-Å-period SL (10 periods) were grown in one structure separated by a 500-Å-thick $Cd_{0.76}Mn_{0.24}Te$ barrier layer in order to keep similar growth conditions for both SL's. All SL's are overgrown by a 500-Å-thick $Cd_{0.76}Mn_{0.24}Te$ cladding layer in order to exclude surface recombination. A 12-Å-thick $CdTe/Cd_{0.74}Mn_{0.26}Te$ SQW was used for comparison with the 24-Å-period SL. For the cw photoluminescence (PL) measurements the samples were inserted in pumped liquid helium in the core of a superconducting split-coil magnet. PL spectra were taken in the Faraday and Voigt geometries, i.e., with magnetic fields applied parallel and perpendicular to the structure growth axis, respectively. Excitation was provided by the 514.5 nm line of an argon-ion laser, a cw DCM dye laser, or, for the time-resolved measurements, by a synchronously pumped mode-locked dye laser system producing 7-ps pulses at 76 MHz. The PL was detected either by a cooled photomultiplier with associated photon-coupling system (combined with a 1-m spectrometer) or by a synchroscan streak camera (combined with a 0.64-m spectrometer) with a time resolution of 30 ps.

III. RESULTS AND DISCUSSION

Magnetic polaron energies were determined from the Stokes shift between the laser energy and the luminescence maximum under selective excitation of localized excitons (details of this method are published in Ref. 9). In Fig. 1(a) PL (excited nonselectively at 2.41 eV) and PL excitation (PLE) spectra of the structure with the 24-Å- and 48-Å-period SL's are shown. The maxima of the PLE spectra correspond to the energy of the free heavy-hole (hh) exciton. The luminescence of both SL's is shifted to lower energies with respect to the free-exciton energy, implying that the recombination is dominated by localized excitons. Under selective excitation of the localized excitons the PL is shifted to lower energies with respect to the excitation energy by $\Delta E=3$ and 12 meV for the 48-Å- and 24-Å-period SL's, respectively [see Fig. 1(b)]. As these additional energy shifts are independent of the energy of selective excitation we conclude, in analogy with the experimental findings in SQW's,[9] that ΔE is determined by magnetic polaron formation only and consider ΔE as the MP energy. This is confirmed by the typical suppression of ΔE by temperature and external magnetic fields as shown for the 24-Å-period SL in Figs. 2 and 3, respectively.

As a result of the alignment of the Mn spins along the direction of an external magnetic field, the polaron shift in the 24-Å-period SL decreases with increasing field applied in both the Faraday (closed circles) and Voigt geometry (open circles) and vanishes for fields above 3 T [Fig. 2(a)]. The suppression, however, is weaker in the Voigt geometry as compared to the Faraday geometry. This difference is even more pronounced in the case of a 12-Å-thick SQW which is shown for comparison in Fig. 2(b).[12] Note that, although the MP energies at zero field are comparable in the SQW and the 24-Å-period SL (13 and 12 meV, respectively), the magnetic-field suppression of the polaron shift in Faraday configuration is twice as efficient in the SL as compared to the SQW. In addition,

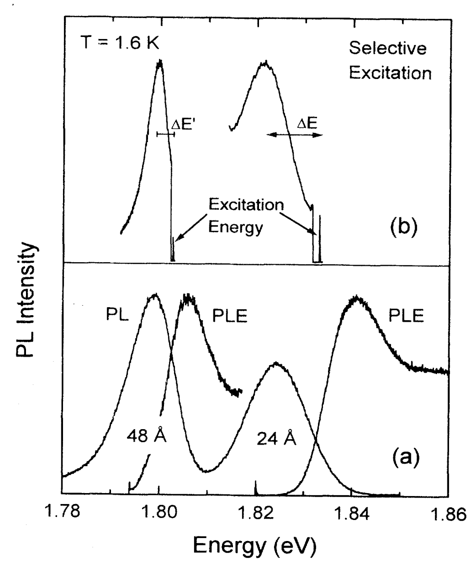

FIG. 1. Photoluminescence and photoluminescence excitation spectra of $CdTe/Cd_{0.76}Mn_{0.24}Te$ SL's with periods of 24 and 48 Å at $T=1.6$ K. In (a) PL is excited nonresonantly at $\hbar\omega_{exc}=2.41$ eV. (b) shows PL spectra taken under selective excitation at $\hbar\omega_{exc}=1.833$ (1.803) eV for the 24-Å- (48-Å-) period SL.

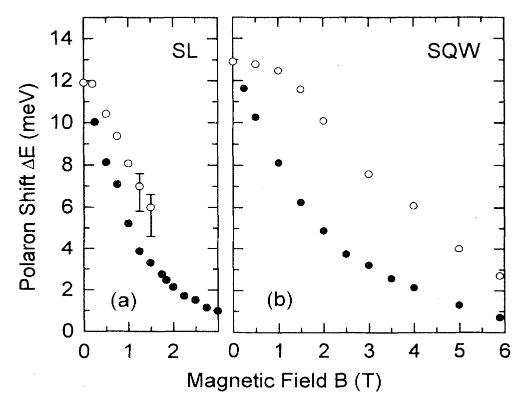

FIG. 2. Polaron shift as a function of external magnetic fields applied parallel (closed circles) and perpendicular (open circles) to the structure growth axis for a 24-Å-period $CdTe/Cd_{0.76}Mn_{0.24}Te$ superlattice (a) and a 12-Å-thick $CdTe/Cd_{0.74}Mn_{0.26}Te$ single quantum well (b). $T=1.6$ K.

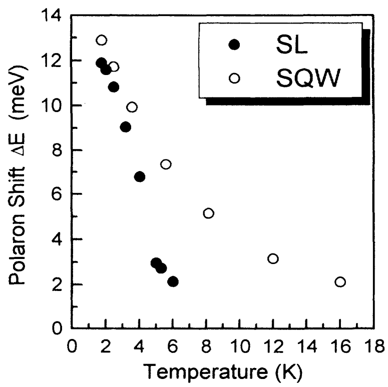

FIG. 3. Polaron shift as a function of temperature in a 24-Å-period $CdTe/Cd_{0.76}Mn_{0.24}Te$ SL (closed circles) and in a 12-Å-thick $CdTe/Cd_{0.74}Mn_{0.26}Te$ SQW (open circles).

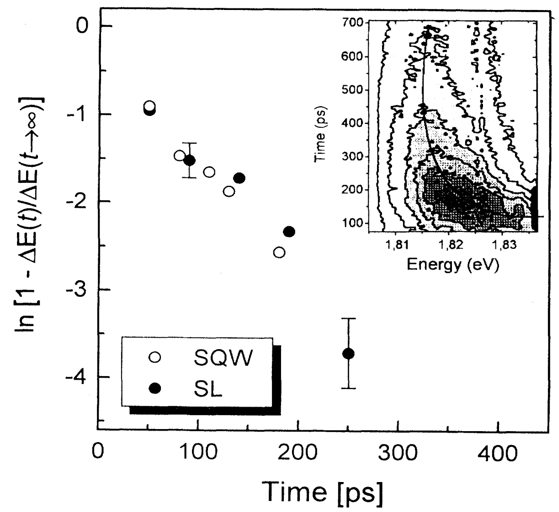

FIG. 4. Dynamics of the magnetic polaron formation in a 24-Å-period $CdTe/Cd_{0.76}Mn_{0.24}Te$ SL (closed circles) and in a 12-Å-thick $CdTe/Cd_{0.74}Mn_{0.26}Te$ SQW (open circles). The inset shows a contour plot of the spectrally and time-resolved PL in the SL excited selectively at 1.836 eV.

the anisotropy between Faraday and Voigt geometries is considerably stronger in the case of the SQW. Similarly to the magnetic-field behavior the MP energy in the SL is suppressed more effectively with increasing temperature (shown in Fig. 3 by closed circles) than in the SQW (Fig. 3, open circles). While the polaron shift in the SL disappears at temperatures as low as 6 K, it vanishes in the SQW for temperatures above 16 K.[16] Mechanisms responsible for these differences between the SL and the SQW will be discussed below. Here we only show the suppression of ΔE with magnetic field and temperature in order to confirm the magnetic origin of the low-energy shift of the PL line under selective excitation.

For a further comparison of the MP energies determined under cw excitation in different structures we made sure that these energies are not underestimated due to the finite lifetime of the exciton. It has been shown that, if the polaron formation time exceeds the polaron lifetime, i.e., the formation process is interrupted by recombination, the MP energy determined under cw excitation is smaller than the polaron equilibrium energy.[17,7] The time evolution of the polaron shift ΔE is measured by time-resolved photoluminescence under selective excitation. The inset of Fig. 4 shows a contour plot of the spectrally and time-resolved PL in the 24-Å-period SL. Here the different gray levels correspond to different luminescence intensities (dark gray, high intensity; light gray, low intensity). The solid line schematically tracks the energy shift of the PL maximum in time. This formation process is described by an exponential law:

$$\Delta E(t)=\Delta E(t\to\infty)\left\{1-\exp\left[-\frac{t}{\tau_f}\right]\right\}, \quad (1)$$

where τ_f is the polaron formation time, and $\Delta E(t\to\infty)$ is the saturation energy shift, which corresponds to the equilibrium MP energy. Figure 4 shows a comparison of the time evolution of the MP shift in the 24-Å-period SL (closed circles) and the 12-Å-thick SQW (open circles) in a logarithmic scale according to Eq. (1). Linear fits to the experimental data yield formation times of 80 ps for both the SL and SQW. Polaron lifetimes of 150 and 170 ps for the SL and the SQW, respectively, are determined from the decay of the spectrally integrated PL intensity. As the SL and SQW under study display similar polaron life- and formation times a comparison of the polaron energies as measured under cw excitation at 1.6 K is valid.

In order to study the MP ground state in $CdTe/Cd_{1-x}Mn_xTe$ SL's in comparison to the case in SQW's we will, in the following, compare the MP energies at 1.6 K in SL's (closed circles) and SQW's (open circles, data are taken from Ref. 9) of different periods and well widths, respectively (Fig. 5). The polaron energy in the SL's decreases with increasing period and is not detectable for $d \geq 60$ Å, i.e., $L_z \geq 30$ Å, which is identical

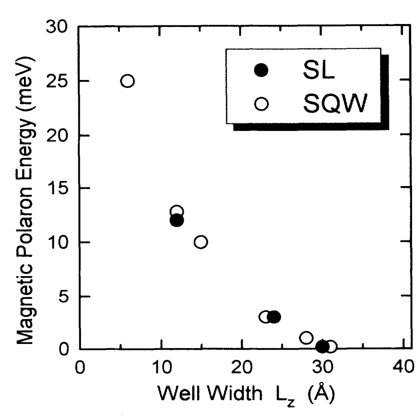

FIG. 5. Energy of magnetic polarons in $CdTe/Cd_{0.76}Mn_{0.24}Te$ superlattices (closed circles) and in $CdTe/Cd_{0.74}Mn_{0.26}Te$ SQW's (open circles) as a function of well width. For SL's $L_z=d/2$. $T=1.6$ K.

to the findings in SQW's. Hence we conclude that in the SL's during the MP formation process the hole wave function does not shift into the semimagnetic barrier layer and remains centered in the nonmagnetic well.[18] This is in contrast to theoretical calculations in which the polaron ground state in a 40-Å-period $CdTe/Cd_{1-x}Mn_xTe$ SL has been found to be composed of a heavy hole centered in the middle of the semimagnetic barrier.[11] In the numerical calculations, however, the strain-free valence-band offset was assumed to be zero and, as a consequence, the exchange potential of the hh leads to the energetically favorable ground state located in the barrier. Yet recent determinations of the strain-free valence-band offset in $CdTe/Cd_{1-x}Mn_xTe$ QW's yield a value of 30% of the total band-gap discontinuity between CdTe and $Cd_{1-x}Mn_xTe$,[10] which in turn cannot be overcome by the exchange energy and thus stabilizes the center of the heavy-hole wave function in the nonmagnetic CdTe layer. In SQW's hole localization in the center of the well layer arises from the combined action of the Coulomb potential of strongly confined electrons[19] and the potential well induced by the valence-band offset.[9] In the short-period SL's studied in this work electrons have wide minibands[15] and are mobile in the structure growth direction. As a consequence, the mobile electrons do not induce the localizing Coulomb potential for the holes and the heavy-hole localization in the structure growth direction is determined by the valence-band offset only. But even in this case, according to our experimental results, the center of the hole wave function is still located in the CdTe well layers. Therefore the Coulomb potential induced by electrons localized in QW's is a conducive but not a necessary condition for the stabilization of the center of the hole wave function in the process of magnetic polaron formation in QW's.

Now we turn to the discussion of the different anisotropic magnetic-field suppressions of the polaron shift as observed in the SL's and SQW's (Fig. 2). In $CdTe/Cd_{1-x}Mn_xTe$ QW's and SL's the moments of hh excitons are oriented along the structure growth axis (z axis) since quantum confinement and strain effects lift the light-hole (lh) states to higher energies and thereby preclude hh-lh mixing at zero field.[20] During the MP formation process the manganese spins inside the polaron orbit are aligned via their exchange interaction with the exciton moment and, finally, the total magnetic moment of the polaron is oriented along the z axis. Therefore an external magnetic field applied in the Faraday geometry does not change the orientation of the Mn spins inside the polaron, but aligns those outside the polaron. This reduces the energy gain of an exciton in the polaron formation process and changes the conditions of MP formation. As a result, the polaron shift is suppressed.

In the Voigt geometry, however, the polaron suppression is more complex. Two cases might be distinguished. (a) Magnetic fields which are sufficiently strong to provide a magnetic energy of the Mn spins exceeding the value of the exchange interaction of these spins with the hh spin in the MP. In this case the Mn spins are oriented by the external field and the anisotropy of the MP suppression is determined by the strength of the exchange field inside the polaron. (b) Stronger magnetic fields which induce a sufficient mixing of hh and lh states to reorientate the exciton moment from the initial quantization axis to the field direction. Thereafter the MP suppression follows the scenario corresponding to the suppression in the Faraday geometry. The field strength required for such a reorientation of the hh spin is determined by the ratio between the Zeeman and hh-lh splitting. Hence the relative contributions of the mechanisms (a) and (b) depend on the properties of the particular structure under investigation and can be studied by a quantitative analysis of the polaron energy, the Zeeman pattern, and hh-lh splitting.

We have measured the exciton Zeeman pattern of the lowest 1s light- and heavy-hole exciton states in Faraday and Voigt geometry for both the 24-Å-period SL and the 12-Å-thick SQW. The exciton energies shown have been determined from the PLE spectra and are shown in Figs. 6 and 7 for the SL and the SQW, respectively. According to the selection rules of the optical transitions we used different circularly polarized light (σ^+ and σ^- in Faraday geometry) and linearly polarized light (π and σ in Voigt geometry) to distinguish the Zeeman components corresponding to states with different spin structures.[21] With respect to the above discussion of the MP suppression, we note that for the SQW, which exhibits a splitting between the light-hole and heavy-hole exciton states (Δ_{HL}^{SQW}) of 32 meV, the Zeeman splitting itself is strongly anisotropic. By contrast, it is nearly isotropic for the SL with $\Delta_{HL}^{SL}=5$ meV. These results are in accordance with previous experimental and theoretical findings,[20–22] in which the anisotropy of the Zeeman splitting was associated with a change of the quantization axis of the hole

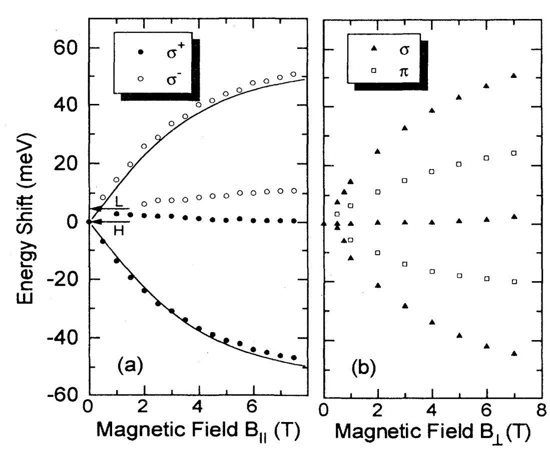

FIG. 6. Exciton Zeeman splittings in a 24-Å-period $CdTe/Cd_{0.76}Mn_{0.24}Te$ SL taken in the Faraday (a) and Voigt (b) geometries. $T=1.6$ K. Zero of the energy shift corresponds to energy of the heavy-hole excitation of 1.840 eV. Light- and heavy-hole exciton energies at zero field are shown by arrows and labeled L and H, respectively. Solid line shows a fit with parameters given in text.

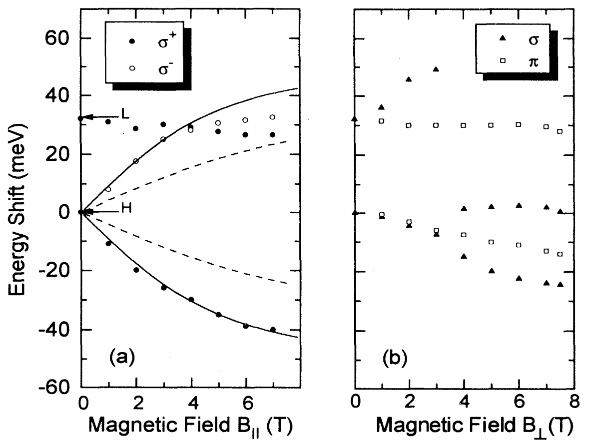

FIG. 7. Exciton Zeeman splittings in a 12-Å-thick $CdTe/Cd_{0.74}Mn_{0.26}Te$ SQW taken in the Faraday (a) and Voigt (b) geometries. $T=1.6$ K. Zero of the energy shift corresponds to energy of the heavy-hole exciton of 1.900 eV. Light- and heavy-hole exciton energies at zero field are shown by arrows and labeled L and H, respectively. Dashed and solid lines show results of calculations with parameters discussed in text.

spin in quasi-two-dimensional layers induced by magnetic fields in the Voigt geometry. In was shown that if Δ_{HL} is smaller than the Zeeman splitting of the lh state in the Voigt geometry a strong mixing of the valence subbands occurs at low fields and the anisotropy is weak (as observed in the SL in Fig. 6). On the other hand, a large Δ_{HL} as compared to the lh Zeeman splitting in Voigt geometry results in a strong anisotropy of the Zeeman pattern (as observed in the SQW in Fig. 7).

Thus for an identification of the mechanism underlying the magnetic-field suppression of the magnetic polaron in the SL and the SQW the hh-lh splitting and the Zeeman splitting of the lh in Voigt geometry have to be compared. As the lh Zeeman splitting in Voigt geometry is similar in both SL and SQW, the magnetic field necessary to provide a sufficient mixing of light- and heavy-hole states is determined by Δ_{HL} only. Experimentally, the magnetic field above which this mixing is effective can be determined from the hh Zeeman patterns in Figs. 6(b) and 7(b). The hh states start to split at 0.3 T in the SL (with $\Delta_{HL}^{SL}=5$ meV) and at 1.7 T in the SQW ($\Delta_{HL}^{SQW}=32$ meV). According to Refs. 20–22 a mixing of the hh and lh states, which occurs at these magnetic fields, leads to a reorientation of the hole spin towards the magnetic-field direction. In turn, these values are in reasonable agreement with the magnetic fields above which the suppression of the magnetic polaron in Voigt geometry starts (0.3 and 1.2 T for the SL and the SQW, respectively; see Fig. 2). On the other hand, model calculations of the MP energy suppression in the Voigt geometry without taking into account mixing of the hole states have shown that the MP suppression due to mechanism (a) becomes effective in fields of 1–2 T only.[23] We therefore conclude that the small anisotropy and low threshold field of the MP suppression in the SL are in accordance with mechanism (b) only. In the SQW both proposed mechanisms contribute. A quantitative distinction, however, is not possible on the base of these experimental findings.

Finally, we turn to the discussion of the stronger magnetic-field (Faraday geometry) and temperature suppression of the MP in the SL as compared to the SQW, despite their polaron energies at 1.6 K being similar (Figs. 2 and 3). This behavior evidences stronger paramagnetic properties of the Mn spins participating in the MP formation in the SL in comparison to the SQW. This interpretation is confirmed by conclusions based on a fit of the Zeeman pattern of the heavy-hole excitons shown in Figs. 6(a) and 7(a). For bulk $Cd_{1-x}Mn_xTe$ the Zeeman splitting of the heavy-hole exciton is well described by the mean-field approximation:[21]

$$E_z=(\alpha-\beta)N_0xS_0B_{5/2}\left\{\frac{5g\mu_BB}{2k_B(T+T_0)}\right\}, \qquad (2)$$

where $N_0\alpha=0.22$ eV and $N_0\beta=-0.88$ eV are the exchange intergrals for conduction and valence band, respectively, x is the manganese content, $B_{5/2}$ is the modified Brillouin function, $g=2$ is the g factor of the Mn ion, μ_B is the Bohr magneton, B is the external magnetic field, and T is the lattice temperature. The effective spin S_0 and the effective temperature T_0 are the parameters representing antiferromagnetic interactions between the Mn ions: $S_0=\frac{5}{2}$ and $T_0=0$ in the limit of noninteracting ions. An increase of the Mn concentration leads to a stronger interaction inside the Mn system, which in turn requires smaller S_0 and larger T_0 values in order to fit the Zeeman pattern with Eq. (2).

In $CdTe/Cd_{1-x}Mn_xTe$ heterostructures the Zeeman splitting is proportional to the part of the wave function which overlaps with the semimagnetic barriers. Taking this penetration of the wave function into account, we have calculated the Zeeman splitting for the SQW with parameters $T_0=11.5$ K and $S_0=0.5$, which describe the Zeeman pattern in the thick $Cd_{0.75}Mn_{0.25}Te$ barrier layers. The calculated splitting [shown by the dashed line in Fig. 7(a)] is much smaller than that found in the experiment. The best fit of the experimental splitting is shown by a solid line and requires $T_0=6.7$ K and $S_0=0.7$ (for details see Ref. 20). These parameters evidence a stronger paramagnetic behavior of the Mn spins located at the interfaces, which give the main contribution to the Zeeman splitting of the SQW exciton. Such a modification of the magnetic properties of the Mn system is due to a reduction of the number of nearest neighbors for Mn spins located at the interface between nonmagnetic and semimagnetic materials. A fit of the SL Zeeman pattern [see the solid line in Fig. 6(a)] requires the parameters $T_0=5.3$ K and $S_0=0.85$, which correspond to a more paramagnetic behavior than in the SQW. In the 24-Å-period SL the barrier layers are 12 Å wide, i.e., four monolayers only. Hence the majority of the Mn ions are located in or next to the interface layers. Therefore high-ordered clusters, which determine the hardness of the magnetic properties,[1] are restricted in their size by the barrier thickness. As a result, the modification of the magnetic properties of the Mn system in the quasi-two-dimensional barrier layers of the SL is even stronger than at the heterointerfaces of the SQW. Evidence for such a modification of the magnetic properties at the interfaces between magnetic and nonmagnetic layers has been reported for $CdTe/Cd_{1-x}Mn_xTe$ QW's (Refs. 12, 13, and 24) and for thin $Cd_{1-x}Mn_xTe$ (Refs. 14 and 25) and MnSe (Ref. 26) layers.

In conclusion, exciton magnetic polarons formed in $CdTe/Cd_{1-x}Mn_xTe$ short-period superlattices have been studied by low-temperature cw and time-resolved photoluminescence for different SL periods, temperatures, and external magnetic fields. A comparison to the polaron formation in single quantum wells shows that the polaron ground state in superlattices is composed of a heavy hole centered in the nonmagnetic well. Temperature- and magnetic-field-dependent measurements of the polaron energy reveal a more efficient polaron suppression in superlattices than in single quantum wells. This is attributed to the modification of the magnetic properties of the Mn system in the thin semimagnetic barrier layers of the superlattices. The anisotropy of the magnetic-field-induced polaron suppression is significantly smaller in SL's in comparison with SQW's. Different mechanisms of polaron suppression in Faraday and Voigt geometries are discussed and it is shown that their contributions differ considerably for the polarons formed in SQW's and in SL's.

ACKNOWLEDGMENTS

The authors are thankful to K. V. Kavokin, I. A. Merkulov, and V. P. Kochereshko for helpful discussions. Support by the Graduiertenkolleg of the Deutsche Forschungsgemeinschaft and the Volkswagen Foundation is acknowledged. R.H. and E.O.G. gratefully acknowledge support by the DFG through the Sonderforschungsbereich SFB 383.

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[16]Similar to the observations in the 24-Å-period SL, in the 48-Å-period SL the MP is suppressed at magnetic fields above 3 T and at temperatures above 6 K.

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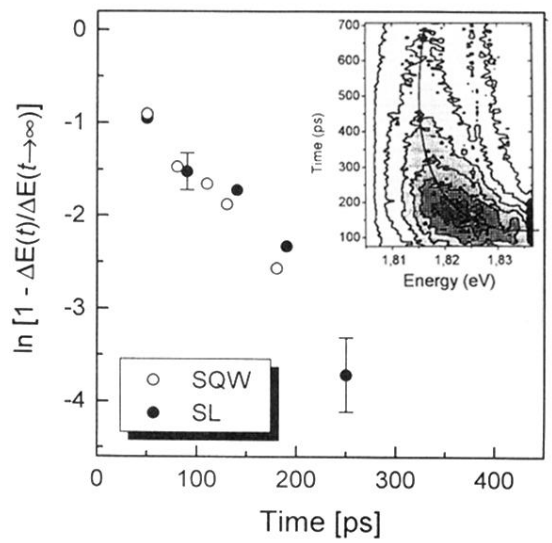

FIG. 4. Dynamics of the magnetic polaron formation in a 24-Å-period CdTe/$Cd_{0.76}Mn_{0.24}Te$ SL (closed circles) and in a 12-Å-thick CdTe/$Cd_{0.74}Mn_{0.26}Te$ SQW (open circles). The inset shows a contour plot of the spectrally and time-resolved PL in the SL excited selectively at 1.836 eV.