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Localized exciton magnetic polarons in $Cd_{1-x}Mn_xTe$

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Using the method of selective excitation of the exciton luminescence in $Cd_{1-x}Mn_xTe$ epilayers we have measured energies of localized magnetic polarons (LMP's) for alloys with manganese mole fractions $x \leq 0.34$. The suppression of the LMP energy has been studied in external magnetic fields and with temperature increase. Polaron formation times and exciton lifetimes have been measured by time-resolved photoluminescence. We have found that in alloys with $x < 0.17$ the polaron formation process is interrupted by exciton recombination and, as a result, the LMP does not reach its equilibrium energy. This dynamical effect on the polaron energy together with the strong sensitivity of the LMP formation to the conditions of primary exciton localization causes the absence of the LMP formation in layers with $x < 0.1$. Antiferromagnetic clustering of Mn ions, which leads to the spin-glass phase formation at low temperatures, affects the polaron energy and results in the increasing stability of LMP's against suppression by temperature increase and magnetic fields. In $Cd_{1-x}Mn_xTe$ with $x > 0.20$ a considerable part of the polaron energy is controlled by the input of clusters of antiferromagnetically coupled Mn spins located in the nonuniform molecular field of localized excitons. The comparison of the exciton Zeeman splitting and the LMP magnetic-field suppression provides insight into the internal structure of LMP's.

I. INTRODUCTION

Unique magnetic properties of semimagnetic semiconductors, such as giant Zeeman splitting of the band states, large Faraday rotation, and magnetic polaron formation result from the strong exchange interaction between carrier spins and magnetic Mn^{2+} ions.[1] Carrier and ion spins interact ferromagnetically. The magnetic polaron formation is caused by the fact that the localization of a carrier leads to the formation of an aligned spin arrangement in the vicinity of the carrier, resulting in a decrease of its energy.[2] Two types of magnetic polarons (MP's) with respect to the conditions of primary localization of carriers have been considered: (i) bound magnetic polarons (BMP's) if carriers are initially bound to a Coulomb impurity (donor or acceptor); and (ii) free magnetic polarons (FMP's) formed from carriers (or excitons) not bound to Coulomb impurities. The stability of FMP's has been investigated by Kasuya, Yanase, and Takeda.[3] Their analysis shows that the polaron energy is determined by a balance of two terms of opposite sign: a positive kinetic energy due to the carrier localization, and a negative energy due to the exchange potential well. Theoretical estimations predict that the stability conditions for pure FMP's are hardly realized in semimagnetic semiconductors, and that the primary localization of carriers has a key importance for the MP stability.[4-6] That is why the term "localized magnetic polaron" (LMP) is used for a FMP affected by the primary localization of carriers. The mechanism of primary localization is not well understood as yet. Golnik, Ginter, and Gaj[7] suggested that compositional fluctuations and fluctuations of magnetization provide the primary localization.

The acceptor bound magnetic polaron (ABMP) has been widely studied both experimentally[8-10] and theoretically[10-13] during the last decade. In contrast to that, there are few experimental investigations concerning the LMP[7,14-16] and a detailed theory of LMP's has not yet been developed. Thus a detailed experimental study of LMP formation is presented in this paper.

LMP's and BMP's differ in the following distinctive properties: (i) BMP's are formed from carriers bound to impurities with relatively long lifetimes. Therefore, the dynamics of the polaron formation is not expected to affect the polaron energy, whereas in LMP's the polaron formation process is limited in time by the exciton recombination, and that can affect the polaron energy. (ii) For BMP's the wave functions are controlled by the Coulomb interaction of the carriers with the impurity center, and are not modified significantly by the MP formation, but in the case of LMP's the wave functions of excitons may shrink considerably in the process of polaron formation, which could be considered as an autolocalization of excitons in an exchange-induced potential.

The LMP was first reported by Golnik, Ginter, and Gaj.[7] The observation of the $L2$ line in p-type $Cd_{1-x}Mn_xTe$ was interpreted as a self-trapped magnetic polaron.[7] The fast decrease and final disappearance of the difference between exciton absorption and luminescence with increasing temperature and in magnetic field were taken as arguments for the identification of the magnetic polaron. The decrease of this difference includes changes in the polaron energy as well as in the nonmagnetic localization energy induced by alloy fluctuations. A separation of both parts is impossible, since raising the temperature or the magnetic field affects not only the po-

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laron energy but also the localization energy and the threshold between nonlocalized and localized states. A direct way to obtain the LMP energy is the method of selective excitation of excitons in the band of localized states, where the nonmagnetic spectral diffusion is suppressed.[17,18] Excitation in the band of localized states generates excitons which are trapped in potential fluctuations around the energy of excitation. Spatial migration for these excitons is suppressed. Therefore, the exciton energy is reduced by the polaron formation process only. In this case the LMP energy could be measured as a shift between the energy of selective excitation and the luminescence peak. The method of selective excitation has been used to determine the LMP energies for contents $0.2 \leq x \leq 0.4$,[14,15,19] and recently down to $x = 0.07$.[16]

The study of two-dimensional exciton magnetic polarons (2D EMP) has been reported in Ref. 18. In this case, the exciton sits in a nonmagnetic CdTe quantum well localized at monolayer fluctuations of the quantum-well width, and forms a polaron due to the penetration of the exciton wave function into the semimagnetic barriers.

In this paper, we present results of selective excitation studies of the localized exciton magnetic polaron formation over a wide range of manganese contents ($0.07 \leq x \leq 0.34$). We show that the shift of the luminescence line under selective excitation provides the polaron energy of LMP's. The dependence of the polaron energy on manganese content, magnetic field, and temperature was studied. We discuss the main differences between BMP and LMP behavior and conclude that the experimentally observable polaron energy is strongly influenced by dynamical effects of the polaron formation in the case of LMP's. The strong influence of the ratio between polaron formation time and exciton lifetime on the polaron energy is demonstrated by picosecond time-resolved photoluminescence experiments. Our experimental results evidence that magnetic hardening of the material at higher x content is another important parameter determining the LMP energy.

II. EXPERIMENTAL DETAILS

The experimental results were obtained on epilayers grown by molecular-beam epitaxy (MBE). The $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ layers were grown on (100)-oriented CdTe substrates at a temperature of 230 °C after 0.2-μm CdTe buffer layers. The epilayer thicknesses were between 1.2 and 3.0 μm, and the manganese concentration varied between 0.07 and 0.34. The samples were mounted in the Faraday configuration in the bore of a superconducting split-coil magnet (fields up to $B = 7.5$ T), immersed in superfluid helium at 1.8 K. Photoluminescence (PL) was excited by the 514-nm line of an argon-ion laser. Selective excitation and PL excitation spectra were carried out using tunable Rhodamine 6G, DCM, or Pyridine 2 dye lasers. The spectra were measured with a single-grating 1-m spectrometer and a cooled photomultiplier. For the time-resolved experiments, excitation was provided by a synchronously pumped mode-locked dye laser producing 5-ps pulses at 76 MHz. A 0.64-m spectrometer and a synchroscan streak camera were used for time-resolved detection of the PL spectra. The resolution of the total system in time and photon energy were 20 ps and 0.4 meV, respectively.

III. RESULTS AND DISCUSSION

A. Determination of localized magnetic polaron energy

In Fig. 1 a typical photoluminescence spectrum (a) and a photoluminescence excitation (PLE) spectrum b of a sample with $x = 0.15$ are shown. The PL spectrum a was taken under nonselective excitation, i.e., with an excitation energy exceeding the free exciton energy. The PL spectrum is dominated by a single broad line. The main contribution to this line is due to the recombination of localized excitons. The exciton localization takes place on alloy and magnetization fluctuations. Donor-acceptor-pair recombination and (e, A^0) recombination are not observed. Our PL lines are of the $L2$ type (i.e., a recombination of self-trapped excitons[7]) in the whole range of contents we have studied. Even for $x < 0.1$, where in p-type bulk $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$ the $L1$ line (acceptor bound excitons) usually dominates the spectrum, we observe the $L2$ line. This is manifested in a very small shift between PLE and PL maximum (e.g., 2.9 meV at $x = 0.07$), that is less than the binding energy of an exciton to a donor or acceptor.

The PLE spectrum b was obtained by detecting on the low-energy side of the PL exciton line. The intensity of the PLE spectrum reflects the density of states which are available to free excitons. The maximum of the PLE spectrum gives a free exciton energy of 1.828 eV. The

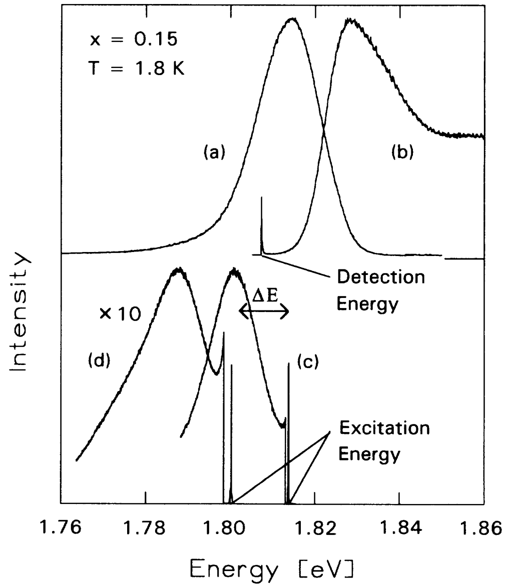

FIG. 1. Spectra of a $\mathrm{Cd}_{0.85}\mathrm{Mn}_{0.15}\mathrm{Te}$ epilayer: photoluminescence spectrum excited at $\hbar\omega_{\mathrm{exc}} = 2.41$ eV (a), photoluminescence excitation spectrum (b), and luminescence spectra under selective excitation at $\hbar\omega_{\mathrm{exc}} = 1.814$ (c) and 1.800 eV (d).

shift between the maximum of the PL line *a* and the PLE spectrum *b* is due to the localization and the magnetic polaron effect. We used the method of selective excitation in order to study the magnetic polaron formation.[17,18] For that we chose excitation energies which are well below the energy of the free exciton to directly excite excitons in the band of localized states. Spectra *c* and *d* in Fig. 1 correspond to excitation within the localized exciton regime. We found that the shift $\Delta E = 12.8$ meV between the excitation energy and the luminescence maximum is independent of the energy of selective excitation. The linewidths of spectra *c* and *d* of 15 meV are smaller than the linewidth of spectrum *a* of 17.5 meV taken under nonselective excitation. The PL intensity of spectrum *d* is smaller by a factor of 10 than the intensity of spectrum *c*, which just reflects the decrease in the density of states for localized excitons.

In order to highlight the constancy of the shift ΔE under selective excitation, we present in Fig. 2 experimental data for layers with two manganese contents $x = 0.15$ and 0.34. Figure 2 displays the Stokes shift between the excitation energy ($\hbar\omega_{\text{exc}}$) and the PL peak taken under this excitation versus the energy distance between the excitation energy and the maximum of the PL line (E_{PL}) taken under nonselective excitation. The dashed line shows the Stokes shift of a line which would not shift with the excitation energy. Decreasing the excitation energy, we proceed from the regime of nonselective excitation, where the PL maximum is independent of $\hbar\omega_{\text{exc}}$ and follows the dashed line in Fig. 2, to the regime of selective excitation of localized excitons, where the PL line shifts together with $\hbar\omega_{\text{exc}}$ at an energy distance ΔE. In the selective excitation regime the Stokes shift becomes independent of $\hbar\omega_{\text{exc}}$ and gives us the value ΔE. The saturation value of the Stokes shift corresponding to ΔE is marked by arrows in Fig. 2, and gives ΔE values of 12.8 and 28 meV for $x = 0.15$ and 0.34, respectively.

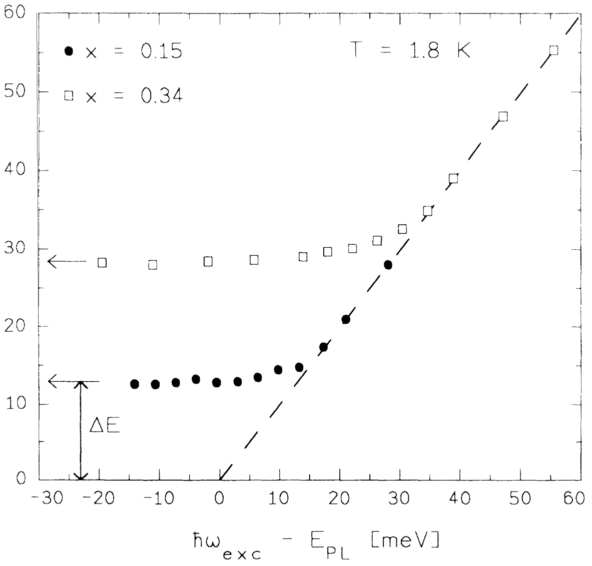

FIG. 2. Stokes shift vs difference between excitation energy and PL maximum taken under nonselective excitation for the manganese contents $x = 0.34$ (squares) and 0.15 (filled circles). The dashed line shows the Stokes shift of a line not shifting with the excitation energy, which corresponds to the regime of nonselective excitation.

The threshold energy separating the two regimes corresponds to the transition from delocalized to localized exciton states and is considered an "effective" mobility edge introduced by Cohen and Sturge.[20] Above the effective mobility edge excitons are mobile within their lifetime, below the effective mobility edge they are localized. For excitons excited selectively below the effective mobility edge the condition of primary localization is satisfied.[5] These excitons form localized exciton magnetic polarons, which appear in the spectra as the shift ΔE. Exciting deeper in the band of localized states reduces the chances for further energy relaxation by tunneling even more. Hence, if there were some contributions to ΔE from this kind of energy relaxation, we should find ΔE becoming smaller with a reduction of the excitation energy. The independence of ΔE from the excitation energy indicates that a contribution from spectra diffusion caused by energy relaxation in the nonmagnetic alloy potential is negligible. Thus the whole value ΔE is due to the magnetic localization via the LMP formation.

Crossing the effective mobility edge is accompanied by a narrowing of the luminescence line (compare spectra *a* and *c* in Fig. 1). This just reflects the fact that the PL linewidth associated with excitons excited above the mobility edge is contributed by fluctuations of both nonmagnetic and magnetic potentials. Contrary, the linewidth of selectively excited excitons is controlled by magnetic fluctuations only.

As one can see in Fig. 2, the transition from delocalized to localized exciton states is not sharp, and the broadening of this transition increases for larger Mn contents. Local variations of the potential (nonmagnetic and magnetic) make it possible that exciton states of the same energy are localized in one part of the crystal, and delocalized in another. This causes a macroscopic broadening of the effective mobility edge which is stronger in samples with higher x values.

For deeper insight into the nature of the shift ΔE we measured the temperature and magnetic field dependences. In Fig. 3(a) the dependence of ΔE on temperature increase for $\text{Cd}_{0.85}\text{Mn}_{0.15}\text{Te}$ is displayed. The energy of selective excitation was kept constant at the maximum of the PL line when the temperature was raised up to $T = 30$ K. Since the band-gap energy remains nearly constant in this temperature range, the condition of selective excitation was not changed. The magnitude ΔE decreases with temperature increase and finally approaches zero at $T = 30$ K. The change of ΔE is much larger than the thermal energy ($kT \sim 2$ meV at $T = 25$ K) and could not be explained by the activation process. The change of ΔE is due to the changes of the magnetic susceptibility, which is a strongly decreasing function of temperature.[1]

The magnetic-field suppression of the shift ΔE at $T = 1.8$ K is illustrated in Fig. 3(b). External magnetic fields were applied along the growth axis of the epilayer in the Faraday geometry. The PL line undergoes a

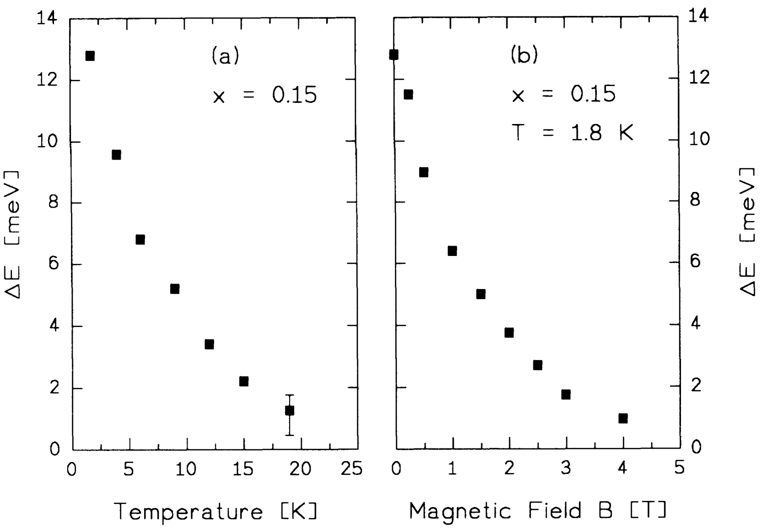

FIG. 3. Suppression of the shift ΔE by temperature increase (a), and in external magnetic fields applied in Faraday geometry (b) for a $Cd_{0.85}Mn_{0.15}Te$ epilayer.

strong low-energy shift due to the giant Zeeman splitting of the exciton states. In order to maintain the conditions of selective excitation in the presence of a magnetic field we tuned the excitation energy to the energy of the PL peak taken under nonselective excitation in this field. We found that an external magnetic field of $B = 5$ T is able to suppress the shift ΔE completely. External magnetic fields align the Mn spins along the field direction. This reduces the energy gain in the process of magnetic polaron formation. The exciton localization energy in the nonmagnetic fluctuation potential is only slightly affected by magnetic fields. Hence the effective decrease of the shift ΔE in magnetic fields proves the magnetic polaron nature of ΔE.

The PL linewidths are observed to shrink strongly with increasing magnetic field. For $x = 0.15$ the nonselectively excited PL line narrows from 17.5 meV at $B = 0$ T to 12 meV at $B = 3$ T, and the line taken under selective excitation narrows from 15 meV at $B = 0$ T to 7 meV at $B = 3$ T. Magnetic fields influence the magnetic part of the fluctuation potential. As a result, magnetic fields suppress the linewidth of the selectively excited PL line more strongly than the nonselectively excited line, since the former one is mainly affected by magnetic fluctuations.

The constancy of the shift ΔE at different energies of selective excitation (see Fig. 2) together with the full suppression of this shift by temperature raise and in magnetic fields (see Fig. 3) gives us direct arguments for the identification of the shift ΔE as the energy of localized exciton magnetic polarons.

As the next step we measured the energy of localized magnetic polarons in $Cd_{1-x}Mn_xTe$ epilayers in the range of Mn content from 0.07 to 0.34 at $T = 1.8$ K. The results are displayed in Fig. 4 by filled circles. We found no detectable polaron shift ΔE in layers with $x < 0.1$. In the range $0.10 \leq x \leq 0.25$ the LMP energy increases with the manganese content and approaches a saturation value of about 28 meV at $x > 0.25$. The open circles in Fig. 4 show the data of Kusrayev and Zakharchenya[15] for the LMP polaron energies determined under selective excitation on Bridgman grown bulk $Cd_{1-x}Mn_xTe$. Aguekian and co-workers[14,21] reported the LMP energies for Bridgman grown samples which coincide with the data of Kusrayev and Zakharchenya. We suppose that the small difference in the LMP energies at high Mn contents is due to the different conditions for Mn clustering in MBE- and Bridgman-grown samples. This could take place because the MBE layers were grown at low temperatures in nonequilibrium conditions. For comparison we show measurements of Bugajski *et al.*[9] representing values of the acceptor-bound magnetic polaron (ABMP) determined from the electron to neutral acceptor recombination. These polaron energies are larger than the energies of the LMP.

In $Cd_{1-x}Mn_xTe$, holes have considerably larger exchange constants[22] and more compact wave functions than electrons. As a result, the hole exchange energy

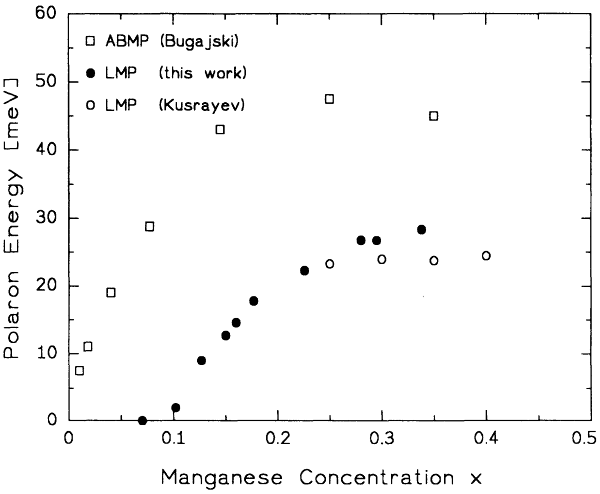

FIG. 4. Energies of LMP's as a function of the x value in $Cd_{1-x}Mn_xTe$ epilayers (filled circles), and bulk data [open circles (Ref. 15)] at $T = 1.8$ K. The ABMP energies (at $T = 5$ K) reported in Ref. 9 are shown by open squares.

dominates in the polaron formation both in LMP's and ABMP's, and the electron input into the polaron energy can be neglected. But ABMP's and LMP's differ in the polaron formation process.

First, they have different conditions for the primary localization. In the case of ABMP's it is the Coulomb binding of a hole to an acceptor which provides a strong initial localization of the hole and controls the size of the polaron orbit. This localization is nearly independent of the x value, and the increase of x leads to a linear increase of the polaron energy, which is proportional to the number of Mn spins included in the polaron orbit. But at $x > 0.12$ the antiferromagnetic coupling between Mn ions exclude some spins from the polaron formation, which leads to the saturation and even decrease of the polaron energy with increasing x (see Fig. 4). In the LMP the initial localization is due to potential fluctuations (nonmagnetic and/or magnetic), which depend strongly on the Mn content. A decrease of x results in an increase of the localization radius, which has a strong effect on the polaron formation. It has been shown by theoretical considerations that the conditions of primary localization control the polaron energy, and the free magnetic polarons are not stable in $Cd_{1-x}Mn_xTe$.[5]

Second, ABMP's and LMP's differ in the dynamics of polaron formation. The polaron shift ΔE reflects the equilibrium polaron energy if the polaron formation time is considerably shorter than the polaron lifetime. If this is not the case, the polaron energy measured in steady-state conditions will be reduced from its equilibrium value, as the process of polaron formation will be interrupted by the polaron recombination. We note that this mechanism does not contribute to the energies of ABMP's. ABMP's are formed from holes which live long enough to reach the equilibrium energy. The finite time period given for the LMP formation and limited by the exciton lifetime could cause the enhancement of the $\Delta E(x)$ dependence at low x.

In order to study the LMP formation process and to understand the qualitatively different dependences of the polaron energies on x for LMP's and ABMP's at low contents, we measured the evolution of the polaron formation in time-resolved experiments with ps resolution.

B. Dynamics of localized magnetic polaron formation

We examined the time evolution of the polaron shift ΔE under 5-ps-pulse selective excitation at $T = 1.8$ K. Under these conditions spectral diffusion is suppressed and $\Delta E(t)$, where t is time delay after the excitation pulse, reflects the process of LMP formation. Figure 5 shows typical data on the evolution of ΔE with time for $x = 0.15$. Two dependences are displayed. The experimental data (open circles) show that $\Delta E(t)$ shifts very fast initially and saturates at larger delays. The saturation value $\Delta E_\infty \equiv \Delta E(t \rightarrow \infty) = 15.5$ meV corresponds to the equilibrium energy of the LMP. The filled circles in Fig. 5 represent the data on a semilogarithmic scale as $\ln[1-\Delta E(t)/\Delta E_\infty]$. A good linear dependence of this function (dashed line) shows that the polaron formation process is satisfactorily described by the exponential law

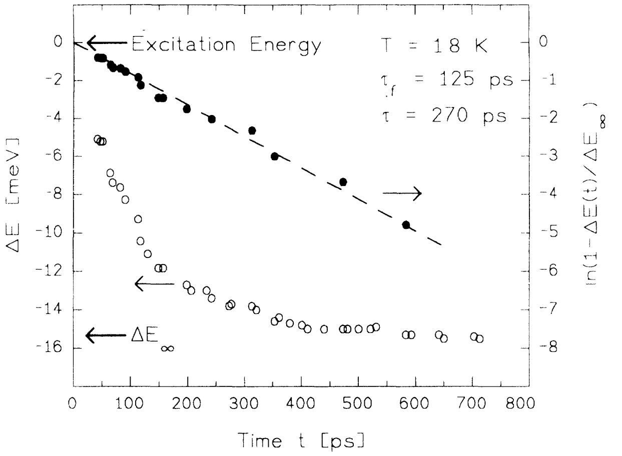

FIG. 5. Time evolution of the polaron shift ΔE for a $Cd_{0.85}Mn_{0.15}Te$ epilayer at $T = 1.8$ K: original data (open circles) and a logarithmical display (filled circles) are presented in order to determine the magnetic polaron formation time τ_f. The fitted slope is shown by a dashed line. τ is the LMP lifetime.

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

where τ_f is the LMP formation time. For $x = 0.15$ a formation time $\tau_f = 125$ ps was determined. We also measured the decay time of the integrated PL intensity under selective excitation in order to determine the LMP lifetime (τ) under the same experimental conditions. During the lifetime of $\tau = 270$ ps the LMP gains about 90% of its equilibrium energy. As a consequence, the LMP energies determined from cw experiments (ΔE_{cw}) are expected to be smaller than the saturation energies. This in fact is the case for the $Cd_{0.85}Mn_{0.15}Te$ sample, where $\Delta E_{cw} = 12.8$ meV on the one hand, but $\Delta E_\infty = 15.5$ meV on the other hand. This difference is due to a dynamical effect on the polaron energy.

The polaron formation times and lifetimes measured for alloys with different content x are summarized in Fig. 6. The formation time decreases with increasing content from 130 ps at $x = 0.13$ down to 55 ps at $x = 0.34$. That reflects the dependence of the exchange interaction on the number of Mn spins participating in the LMP formation. Furthermore, in the alloys of larger x content the conditions of initial localization are better fulfilled due to the stronger localization on magnetic and nonmagnetic fluctuations. Itoh and Komatsu reported a formation time of 40 ps in bulk $Cd_{0.8}Mn_{0.2}Te$,[19] which is close to our results. Our measurements of the decay times of the PL intensity under selective excitation provide LMP lifetimes between 180 and 480 ps for the samples investigated. In contrast to the polaron formation time, the lifetime increases with increasing x. We found that for $x = 0.34$ the lifetime is much longer than the polaron formation time ($\tau_f/\tau = 0.125$). Consequently, the LMP's

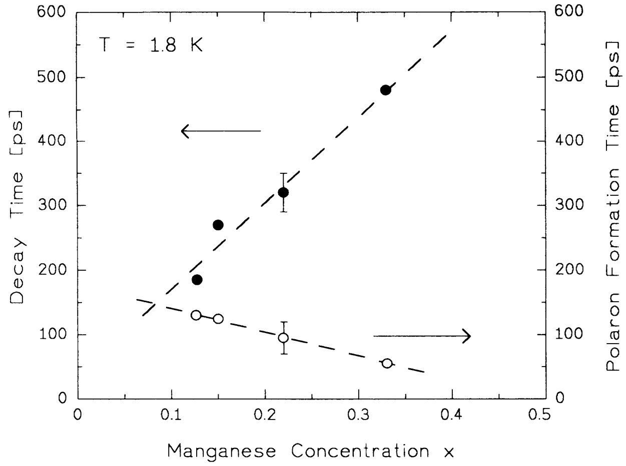

FIG. 6. Decay time of integral photoluminescence (filled circles), and magnetic polaron formation time (open circles) measured under selective excitation as a function of manganese content. Dashed lines are drawn as a guide to the eye.

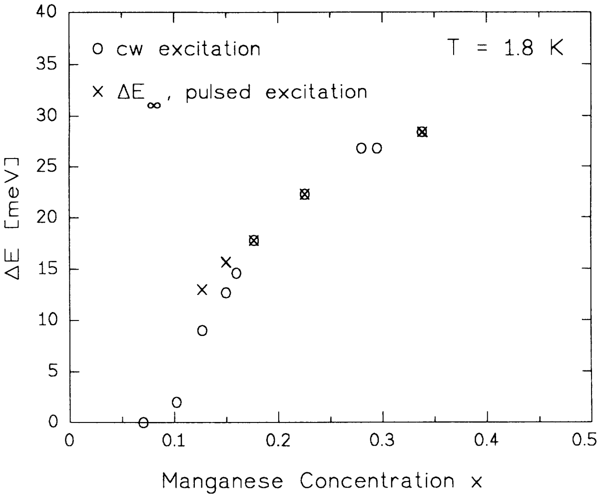

FIG. 7. LMP energies measured under cw selective excitation (circles) and taken as the saturation value of the polaron shift under pulsed selective excitation (crosses) as a function of the manganese content in $Cd_{1-x}Mn_xTe$.

can reach their equilibrium energy, and the value of the polaron energy obtained from cw expansions should correspond to the saturation value ΔE_∞. That has been found experimentally for layers with $x > 0.15$. For alloys with $x \leq 0.15$ the condition under which the LMP can reach its equilibrium energy during the lifetime ($\tau_f/\tau \ll 1$) is not fulfilled. In the case of $x = 0.13$ the ratio between formation time and lifetime is $\tau_f/\tau = 0.7$, and the saturation value of $\Delta E_\infty = 12.8$ meV considerably exceeds the polaron shift of 8.6 meV determined under cw conditions. Comparing polaron formation time and polaron lifetime in Fig. 6, we conclude that for $x < 0.1$ the polaron formation time is longer than the polaron lifetime. In this case the dynamical effect is accompanied by a weak exciton localization at small manganese contents. There are not enough fluctuations of sufficient depth, which are able to provide a considerable degree of exciton localization and, as a consequence, the mobile excitons cannot form LMP's. The criteria of initial localization of excitons preceding the LMP formation[5] are not satisfied for $x < 0.10$.

The results for LMP energies determined by two methods are summarized in Fig. 7. The LMP energies taken from the shift under cw excitation are shown by circles, and the values for equilibrium polaron energies extracted from the saturation of $\Delta E(t)$ in time-resolved experiments are shown by crosses. Figure 7 demonstrates that the polaron energies determined in cw experiments are equal to the equilibrium polaron energies for $x \geq 0.17$. These energies differ for lower x contents, and this difference is quite considerable for $x = 0.13$. It shows that in alloys with $x < 0.17$ the polaron shift taken under cw conditions no longer reflects the equilibrium polaron energy, and the difference between the polaron shift obtained under cw conditions and the saturation energy ΔE_∞ increases with decreasing x. As a result, we conclude that the strong suppression of ΔE for $x < 0.15$ under cw conditions is due to the decrease of the LMP equilibrium energy and is enhanced by the dynamical effect. We were not able to resolve in time-resolved measurements the polaron shift for $x \leq 0.1$. We suppose that for this range of contents a low probability of exciton localization is mainly responsible for the nonformation of the LMP. We have demonstrated the role of the dynamical effect on the LMP energy, which is important for alloys with contents below $x = 0.17$. The recombination of excitons interrupts the process of LMP formation, and polarons do not reach their equilibrium energy. Contrary to the LMP, the observed shift of the ABMP reflects the equilibrium polaron energy in the whole range of contents, as polarons are formed involving holes which are bound to acceptors, and live long enough to reach the equilibrium energy.

Figure 8 shows the decay times of the spectrally in-

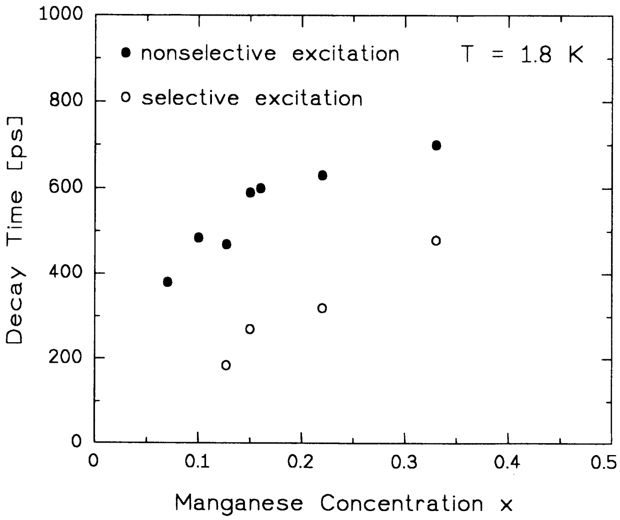

FIG. 8. Decay time of the integral photoluminescence taken under nonselective (filled circles) and under selective excitation (open circles) as a function of manganese content in $Cd_{1-x}Mn_xTe$ epilayers.

tegrated PL measured under selective and nonselective excitation conditions. Both decay times determined under selective and nonselective excitation increase strongly with increasing x. We found much shorter lifetimes for selective excitation. The difference in lifetimes is between 250 and 300 ps for all contents studied. We suppose that longer decays under nonselective excitation are due to the spectral diffusion process preceding the exciton localization. The spectral diffusion process is suppressed under selective excitation. It was shown recently that in GaAs heterostructures the relaxation of excitons with large-**K** vectors into radiating $\mathbf{K}=0$ excitons is predominantly responsible for the long decay times under nonselective excitation conditions.[23,24] The relaxation of large-**K** excitons to $\mathbf{K}=0$ excitons takes place via exciton-exciton collisions and exciton–acoustic-phonon collisions. We note that the influence of nonradiative mechanisms should be excluded, as they should be stronger for free excitons (i.e., under nonselective excitation) and should have the opposite effect on lifetimes.

C. Effect of antiferromagnetic clusters on localized magnetic polarons

In this section we present an experimental study of the LMP in alloys with high-Mn contents, where the antiferromagnetic coupling of neighboring Mn ions leads to the hardening of the magnetic system of Mn ions. In $Cd_{1-x}Mn_xTe$ semimagnetic semiconductors at low temperatures the nearest-neighbor Mn ions couple in antiferromagnetic (AF) pairs and clusters of higher order and give no or only small input into the magnetic susceptibility χ.[25] The number of Mn spins involved in clusters increases rapidly with increasing x content and, as a consequence, the function $\chi(x)$ decreases for $x \geq 0.12$ at $T=1$ K.[26] For $Cd_{1-x}Mn_xTe$ with high-x content the decrease of temperature leads to the increase of interacting Mn spins and, finally, to the formation of an infinite cluster, which is considered as the spin-glass phase formation. The temperature of the phase transition from a high-temperature paramagnetic phase to a low-temperature spin-glass phase has been determined from a cusp on the dependence $\chi(T)$[1] and mirrors the magnetic hardening of the system. The increase of the magnetic susceptibility with temperature decrease $\chi \propto T^{-1}$ is interrupted at the temperature of the spin-glass transition (T_{SG}), and χ becomes a weak function of temperature below T_{SG}. The temperature of the spin-glass transition and the magnetic hardness of the Mn system at a given temperature increase for higher Mn contents.

It has been suggested by Janiszewski that the response of the clusters of antiferromagnetically coupled ions to a strongly nonuniform molecular field induced by a localized carrier can govern the MP formation and give input into the polaron energy.[27] Recently Kavokin and Merkulov considered a quadrupole magnetic susceptibility of AF pairs, which are the smallest clusters, and demonstrated their role for the LMP energy.[28] Hence the LMP energy is contributed by two terms which are attributed to the isolated paramagnetic ions (E_P) and to the clusters of antiferromagnetically coupled ions (E_{AF}). In E_{AF} we include the polaron energy which is due to the exchange interaction of the heavy-hole spin with the total moment of clusters, and with the higher-order magnetic moments starting from the quadrupole moment. Strictly speaking, the total polaron energy could not be considered as a sum of two independent terms, as the value of E_{AF} depends on the gradient of the molecular field, i.e. on the degree of carrier localization, and, as a consequence, depends on the value E_P. Nevertheless, the introduction of these terms is convenient for the qualitative consideration of the LMP formation in the spin-glass phase.

The increase of the Mn content for $x \geq 0.10$ is accompanied by the reduction of the number of paraspins and, as a result, by the decrease of E_P. On the other hand, the number of spins participating in AF clusters increases and E_{AF} grows upward. The increase of E_{AF} contributes to maintain the LMP energy in the wide range of x content from 0.25 to 0.40 (see Fig. 4) in spite of the strong decrease of the magnetic susceptibility $\chi(x)$ and, respectively, the decrease of E_P.[26] External magnetic fields affect the two terms of the LMP energy in a different way. Magnetic fields suppress the input of the paraspins via their alignment along the field direction, and have no influence on the input of AF clusters up to very high fields ($B > 10$ T), which break the antiferromagnetic coupling of nearest-neighbor pairs.[29] With a temperature increase E_P should decrease following the $\chi(T)$ dependence. But, the input of E_P on the LMP is reduced for larger x contents. Hence the temperature suppression of the LMP energy should occur at higher temperatures for larger x contents.

In order to experimentally study the effect of the AF clusters on the LMP formation we measured the temperature and magnetic-field dependences of the LMP energy in a wide range of Mn content varying from 0.13 to 0.34. The LMP energies shown in Figs. 9 and 10 are normalized to their values at zero field and at 1.8 K for convenience of comparison.

In Fig. 9 the decrease of the LMP energy with increas-

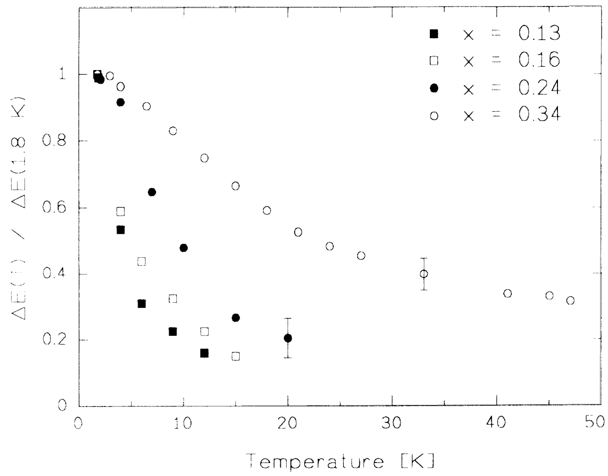

FIG. 9. The polaron shift ΔE as a function of temperature for $Cd_{1-x}Mn_xTe$ layers with $x=0.13$, 0.16, 0.24, and 0.34. Energies are normalized to their values at $T=1.8$ K.

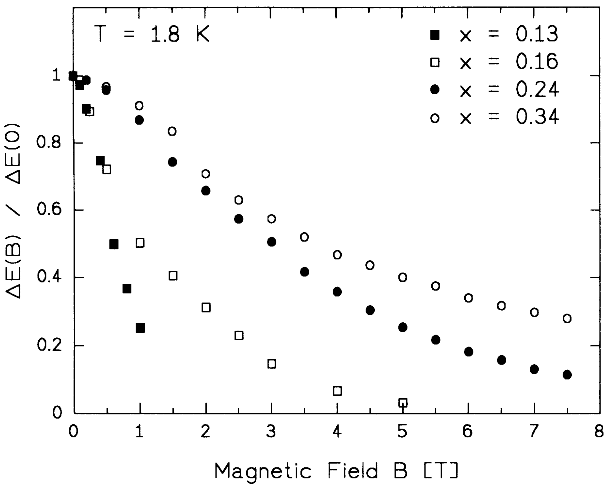

FIG. 10. The polaron shift ΔE as a function of external magnetic fields for $Cd_{1-x}Mn_xTe$ layers with $x=0.13$, 0.16, 0.24, and 0.34. Energies are normalized to their values at zero magnetic field.

ing temperature is presented for four manganese contents (0.13, 0.16, 0.24, and 0.34). The polaron energy is found to decrease dramatically for all contents. For $x \leq 0.24$ the full suppression of the LMP energy is observed at $T=30$ K. We note that the suppressions for $x=0.24$ and 0.34 differ strongly, despite the fact that they have very close values of the LMP energies at 1.8 K (see Fig. 4).

The magnetic-field suppression of LMP's is illustrated in Fig. 10, where the shift ΔE (normalized to the value at $B=0$ T) taken under selective excitation is plotted as a function of magnetic field at $T=1.8$ K for four different contents. Magnetic fields were applied parallel to the growth axis of the epilayers (Faraday geometry). The polaron energy is found to decrease dramatically with increasing magnetic field for all contents, but with different suppression effectiveness. An external magnetic field of $B=7.5$ T is able to fully suppress the LMP formation in samples with $x \leq 0.2$. For samples with larger Mn content this is not the case, though we still observe a slow decrease of the polaron energy within this field range. The similar results for the magnetic-field suppression of the polaron shift in $Cd_{1-x}Mn_xTe$ with $x \geq 0.25$ have been reported recently.[21] The polaron suppression behavior is governed by the spin alignment in the system of Mn ions. The field strength required to suppress the polaron energy to 50% increases with increasing manganese concentration. The comparison of the suppression behavior for different Mn concentrations suggests two different features: (i) For $x \leq 0.24$ the curves differ widely in their slopes at low magnetic fields. We attribute this to the increase of the internal exchange interaction in the LMP with increasing x values. Therefore, only larger magnetic fields are able to cause a degree of Mn^{2+}-spin alignment in the crystal that approaches the spin alignment in the polaron orbit due to the internal exchange interaction. For contents of 0.13 and 0.16, the suppression with temperature increases, and in magnetic fields could be enhanced by the dynamical effect (see Sec. III B), as the polaron formation time increases with temperature[19] and in external magnetic fields.[8] (ii) The suppression curves of the $x=0.24$ and 0.34 samples show a significant difference of the polaron energy at $B > 4$ T. This behavior is attributed to the magnetic hardening at higher manganese concentrations.

In order to highlight the effect of AF clusters on the LMP formation, we compare the experimental data for the two contents 0.24 and 0.34 presented in Figs. 9 and 10. For $x=0.24$ and 0.34 the condition $\tau_f/\tau \ll 1$ is satisfied, and experimental data from Figs. 9 and 10 measured under cw excitation reflect the changes of the equilibrium polaron energy. In line with considerations presented above in this section, the strong distinction in the temperature suppressions of 0.24 and 0.34 contents (see Fig. 9) is due to the different hardness of the Mn system, which could be characterized by different temperature of the spin-glass phase transition, which are 4 and 9 K for $x=0.24$ and 0.34, respectively.[25] In magnetic fields up to 3 T the dependences for $x=0.24$ and 0.34 are very similar (see Fig. 10). At this field the paramagnetic term E_P of the LMP energy is suppressed. At fields higher than 4 T the polaron energies for the two contents differ considerably, which reflects the larger contribution of the E_{AF} term in $x=0.34$ than in $x=0.24$. An estimation of the E_{AF} terms could be done from the values of polaron energies, which are not suppressed by the field of 7.5 T. They are 2.5 meV for $x=0.24$, and 8 meV for $x=0.34$.

As we have shown experimentally, the magnetic hardening of the Mn system accompanied by the spin-glass phase formation at low temperatures contributes to maintaining the LMP energy from 0.25 to 0.40, and increases the stability of LMP's against temperature and magnetic field suppression. In $Cd_{1-x}Mn_xTe$ the part of the polaron energy controlled by AF clusters increases with increasing Mn content.

In Fig. 11 two dependences for each of the contents $x=0.16$ and 0.34 are compared: (i) the polaron energy versus strength of magnetic field (normalized to its value at zero magnetic field), and (ii) the exciton Zeeman splitting. The Zeeman splitting is normalized to its saturation value at $B \rightarrow \infty$ determined from the extrapolation of the modified Brillouin function, with corresponding parameters of effective temperature and effective spin.[22] The saturation value of the Zeeman splitting corresponds to the full alignment of paramagnetic spins along the field direction. Hence the Zeeman splitting taken at a given magnetic field could be used as a measure of the degree of spin alignment in this field. The AF clusters are not effectively aligned by uniform external fields because of their small total moment, and the contribution of paramagnetic spins dominates the Zeeman splitting. On the other hand, the LMP energy includes inputs from both paraspins and AF clusters. While a field of $B=2.5$ T is necessary to obtain half of the total Zeeman splitting at $x=0.16$, the polaron energy has decreased to half of its value already at $B=1$ T and it is fully suppressed when the Zeeman splitting reaches 80% of its saturation value. This significant difference between Zeeman split-

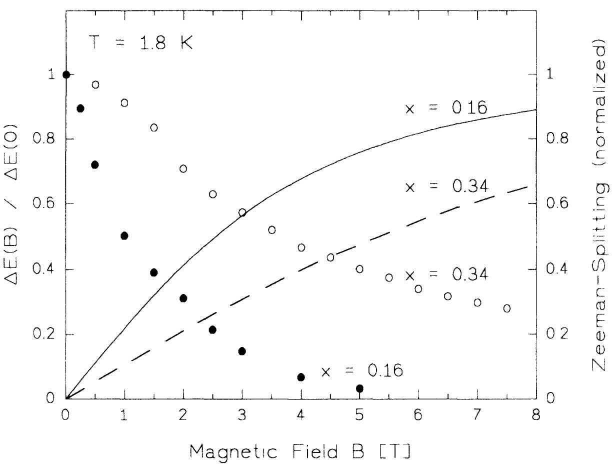

FIG. 11. Comparison between exciton Zeeman splitting (lines) and polaron energy suppression (circles) in magnetic fields for $x = 0.16$ (closed circles and solid line) and 0.34 (open circles and dashed line). The Zeeman splitting determined from PLE measurements is normalized to its saturation value at $B \rightarrow \infty$, and the LMP energies are normalized to their values at zero magnetic field.

ting and polaron suppression is reduced at larger Mn contents.

A comparison between Zeeman splitting and suppression of the LMP in high magnetic fields allows us to estimate the degree of spin alignment within the orbit of a magnetic polaron. The Zeeman splitting reflects the degree of spin alignment in the entire Mn system since the effect is caused by free carriers reacting to the total average spin alignment in the sample. The LMP energy is only affected by the spins inside the LMP orbit. As can be seen from Fig. 11, the polaron suppression is a nonlinear function of the spin alignment in the entire system. The LMP formation is completely suppressed by external magnetic fields when the degree of spin alignment inside and outside the polaron orbit approach each other. Therefore, the fact that the polaron energy at $x = 0.16$ is suppressed at 80% of the total Zeeman splitting reflects that the degree of spin alignment inside the polaron orbit is not less than 80%. We note that the increase of the polaron formation time in external magnetic fields[8] could enhance the suppression effectiveness and thus lead to an underestimation of the degree of spin alignment in the method we use. For $x = 0.13$ the difference between Zeeman splitting and polaron suppression is even more pronounced than for $x = 0.16$. But in this case the polaron dynamics restricts the formation of polarons with equilibrium energies, and also has a stronger influence on the polaron suppression in magnetic fields, which screens the real degree of spin alignment in the magnetic polaron orbit. We have shown that the comparison of the LMP suppression and the Zeeman splitting allows us to obtain information about the internal magnetic structure of the LMP, i.e., about the degree of spin alignment inside the polaron orbit, which exceeds 80% in the case of $x = 0.16$.

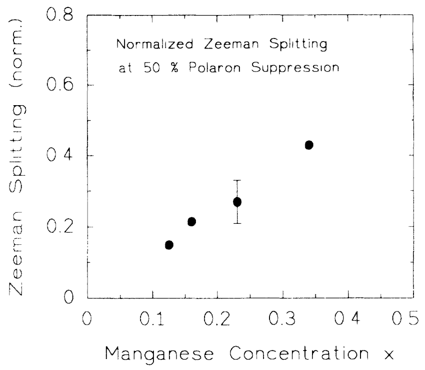

FIG. 12. Normalized exciton Zeeman splitting at the magnetic field, which suppresses the magnetic polaron energy to 50% of its zero-field value, as a function of manganese content. $T = 1.8$ K.

We now analyze how the degree of Mn spin alignment induced by external fields affects the LMP suppression. The results are summarized in Fig. 12, where the value of the normalized Zeeman splitting at the field which suppresses the LMP to 50% if its zero-field value is shown as a function of Mn content. The degree of the spin alignment required for the polaron suppression strongly increases with increasing x content. We note that the data for $x = 0.13$ and 0.16 could be slightly underestimated because of the dynamical effect on the polaron energy. But even the comparison of the contents $x = 0.24$ and 0.34, where the dynamical effect has no input, shows that the suppression of the LMP in the deep spin-glass phase ($x = 0.34$) requires considerably stronger spin alignment (42% of the Zeeman splitting) than at $x = 0.24$ (28% of the Zeeman splitting only). This result demonstrates that in spite of the approximate constancy of the polaron energy at $x \geq 0.25$ the properties of the LMP's change strongly with increasing Mn content accompanied by the hardening of the spin-glass phase.

D. Final remarks

Finally, we wish to discuss the polaron effect on the value of the Stokes shift between the PL line taken under nonselective excitation and the maximum of the PLE spectra, which corresponds to the energy of the free exciton. This Stokes shift just reflects the characteristic energy of localization reached by the excitons within their lifetime. In semimagnetic semiconductors the magnetic fluctuations and the magnetic polaron formation participate in the exciton localization in addition to the nonmagnetic compositional fluctuations, which control the exciton localization in nonmagnetic materials. As we have shown in this paper, the magnetic polaron formation is extremely sensitive to temperature and external magnetic fields, and can be effectively suppressed. This causes the strong dependence of the Stokes shift on temperature and magnetic field. We found that in a $Cd_{0.76}Mn_{0.24}Te$ epilayer the Stokes shift decreases from 23 to 14 meV with a temperature increase from 1.8 to 25 K, and decreases from 23 meV at zero magnetic field to 11 meV at $B = 7.5$ T. The LMP energy determined by the selective excitation technique gives 23 meV, and exceeds considerably the part of the Stokes shift suppressed by temperature (9 meV) or magnetic field (12 meV). This comparison shows that the Stokes shift behavior gives no reliable value for the LMP energy, and could not be used for the determination of the polaron energy. There are several reasons which make it difficult to extract the LMP energies from the Stokes shift behavior: (i) External magnetic fields smooth the fluctuations of magnetization, and could induce changes of the effective mobility edge. (ii) Magnetic fields affect the lifetime, which could interrupt the localization process at earlier stages. (iii) It remains undetermined at what field the polaron is fully suppressed, resulting in an uncertainty in the determination of the localization energy. We found that the polaron energy is not fully suppressed at $B=7$ T for $x>0.2$.

We have shown that in semimagnetic semiconductors the Stokes shift of the luminescence line of localized excitons is strongly affected by the polaron formation process, but the changes of the Stokes shift with temperature and in magnetic field give no reliable data for the LMP energy determination.

IV. CONCLUSIONS

We have studied the formation of localized exciton magnetic polarons in $Cd_{1-x}Mn_xTe$ over a wide range of manganese contents ($0.07 \leq x \leq 0.34$). We have shown that the shift ΔE appearing under selective excitation in the band of localized exciton states represents the energy of the localized exciton magnetic polarons. The dependences of the polaron energy on manganese content, temperature, and magnetic field have been studied. The polaron formation can be fully suppressed by an external magnetic field of 7.5 T or/and by temperature increase in epilayers with $x \leq 0.2$. The dynamics of the LMP formation has been studied by use of time-resolved spectroscopy. The polaron formation time decreases from 130 to 55 ps with increasing Mn content, whereas the lifetime increases from 180 to 480 ps. The ratio between the polaron formation time and the polaron lifetime controls the dynamical effect which inhibits the formation of polarons with equilibrium energies for $x<0.17$. Antiferromagnetic clustering of Mn ions, which leads to the spin-glass phase formation at low temperatures, affects the polaron energy and results in the increasing stability of LMP's against temperature and magnetic-field suppression. In $Cd_{1-x}Mn_xTe$ with $x>0.20$ a considerable part of the polaron energy is controlled by the input of clusters of antiferromagnetically coupled Mn spins located in the nonuniform molecular field of localized excitons. The comparison of the exciton Zeeman splitting and the LMP suppression in magnetic fields provides insight into the internal structure of LMP's. Contrary to the BMP's, a theoretical description of the LMP's is not well developed as yet, especially for the situation where dynamical processes should be taken into account. We hope that our detailed study of the properties of LMP's will stimulate further theoretical considerations in this field.

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