Optical properties and dynamical behavior of localized and bound excitons in $Cd_{1-x}Mn_xTe$ ($x=4\%$) grown by molecular-beam epitaxy

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An epitaxial $Cd_{1-x}Mn_xTe$ ($x=4\%$) layer has been grown on a CdTe buffer layer on a GaAs substrate by the molecular-beam-epitaxy method. Comparing the photoluminescence (PL) spectra of $Cd_{1-x}Mn_xTe$ with those of CdTe, two peaks in the $Cd_{1-x}Mn_xTe$ PL domain were assigned to localized and bound exciton emissions. The results obtained by PL measurements in magnetic fields up to 40 T support this assignment. The observed temporal energy shifts of the localized and bound exciton emissions can be attributed to localized and bound magnetic polaron formations, respectively. Comparing the temporal behavior of bound excitons in magnetic fields with that of localized excitons, a possible process of the bound magnetic polaron formation is discussed.

I. INTRODUCTION

$Cd_{1-x}Mn_xTe$ is a typical semimagnetic semiconductor and in this system spins of carriers interact strongly with spins of Mn^{2+} ions through the *sp-d* exchange interaction. As a result, unusual magneto-optical properties have been observed in this compound.[1-3] A magnetic polaron state is also formed due to the *sp-d* interaction.[4] A time-resolved photoluminescence (PL) study can provide real-time information of the dynamics of the magnetic polaron formation. Previous studies have shown that the localization of an electron (hole) or exciton leads to a Mn^{2+} spin alignment, which is called a magnetic polaron.[5,6]

In $Cd_{1-x}Mn_xTe$ samples grown by conventional growth methods, it is reported that a strong emission of excitons bound to neutral acceptors, (A^0,X), is usually observed for low-Mn concentrations ($x \lesssim 5\%$).[4,7] In this system, the magnetic polaron is formed mainly by the hole localization; we will refer to magnetic polarons formed by this process as bound magnetic polarons (BMP's). In $Cd_{1-x}Mn_xTe$ samples with high-Mn concentrations ($5\% \lesssim x \lesssim 40\%$) exciton localization due to fluctuating potentials arise from compositional inhomogeneities becomes dominant.[4,8] The localization effect in the fluctuating potentials is a well-known cause of inhomogeneous broadening of the exciton emission.[9,10] The free exciton captured by the fluctuating potential is called a localized exciton. In high-Mn concentrations, the excitonic magnetic polaron is formed mainly by this type of localization. Itoh and Komatsu[11] reported that the time constant for excitonic magnetic polaron formation induced by localization was 40 ps in $Cd_{0.8}Mn_{0.2}Te$ at 2 K. Since this type of magnetic polaron is formed only after the exciton localization, it is called a localized magnetic polaron (LMP). Therefore, in this paper, the LMP is distinguished from the BMP by the origin of the localization; the localized carrier in the (A^0,X) state and the alloy fluctuation for the BMP and the LMP, respectively.

The comparison of the results of time-resolved localized and bound exciton emissions enables one to elucidate the magnetic polaron formation mechanism, since the conditions for the localization are different. For example, the number of the localized holes in the BMP of (A^0,X) and the LMP are two and one, respectively. Since the dynamics of excitons are strongly affected by a nonradiative decay, which depends strongly on alloy fluctuations and the impurity profile, it is important to investigate the dynamics of localized and bound excitons in the same sample. To our knowledge such an investigation has not been done well so far, because clear emission lines due to these two different origins cannot be observed simultaneously in previously reported samples. It was difficult in the previous experiments to prepare the samples which showed such behaviors, because these two lines can be observed only in a small range of Mn concentrations ($x=4-5\%$).

In the present study, a $Cd_{1-x}Mn_xTe$ epitaxial film, with a Mn concentration of 4%, was grown on a CdTe buffer layer on a GaAs substrate by molecular-beam epitaxy (MBE). By adjusting the growth conditions we could grow a sample in which the emissions from both localized and bound excitons are observed clearly in the same sample, although the bound exciton emission is expected to dominate in this sample according to previous reports.[4,7] In previous studies either the BMP or the LMP was observed in each sample, but there have been no reports of the simultaneous observation of both magnetic polarons in one sample. In the present study we

succeeded in growing a sample in which the dynamical behaviors of these emission lines can be investigated simultaneously.

The assignment of the time-integrated PL spectra was made using the spectral-decomposition method. The stacking structure of the sample is such that it enables one to observe the PL spectra arising from both the $Cd_{1-x}Mn_xTe$ epitaxial film and the CdTe buffer layer. In this situation, the decomposed $Cd_{1-x}Mn_xTe$ PL spectra can be assigned by comparing the spectra with that of CdTe grown under the same condition as $Cd_{1-x}Mn_xTe$. CdTe spectra are well assigned from previous literature.[12–14] In addition, the time-integrated PL spectra in magnetic fields up to 40 T and the time-resolved PL spectra at 4.2 K have been also measured for this sample. These measurements support the assignment of PL spectra done by the spectral-decomposition method and have elucidated the physical and optical properties of excitons in this sample, such as the effective mass of the excitons and the effective moment of the Mn^{2+} spins. The dynamics of excitonic emissions have been measured by the time-resolved PL spectra in a magnetic field at 4.2 K. New information on the magnetic polaron formation process is expected to be obtained from time-resolved PL spectra of localized and bound excitons in the same sample.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

A sample was grown by the MBE method, as illustrated in Fig. 1. The substrate temperature during the MBE growth process was 350 °C and the growth orientation of the epitaxial film was (100). In order to avoid structural defect in the $Cd_{1-x}Mn_xTe$ layer due to the large lattice mismatch of 14.6% between CdTe and GaAs, a sufficiently thick CdTe buffer layer (about 4.5 μm) was grown on top of the GaAs substrate. Then a $Cd_{1-x}Mn_xTe$ epitaxial film with a thickness of about 5000 Å was grown on the CdTe buffer layer. Three CdTe, Te, and Mn Knudsen cells were operated at about 490, 300, and 750 °C, respectively. Under this condition, the growth rate of the epitaxial film was about 3 Å/s.

The time-integrated PL spectra were measured at 4.2 K using the 5145-Å line of an Ar^+ ion laser. The emission collected in the backscattering geometry was focused on the entrance slit of a SPEX double monochrometer equipped with a GaAs photomultiplier tube and photon-counting detecting system. The time-integrated PL spectra in pulsed magnetic fields up to 40 T were measured at 4.2 K. The sample was mounted in the [**H**||**z**||**k**] Faraday configuration, where **H**, **z**, and **k** express the applied magnetic field, the normal vector to the sample surface, and the wave vector of the excitation/luminescence lights, respectively. The 5145-Å line of an Ar^+ ion laser was used for the excitation. Light from the laser was transferred to the sample holder by an optical fiber and the luminescence light was transferred to a spectrometer with an optical multichannel analyzer (OMA) through another fiber.

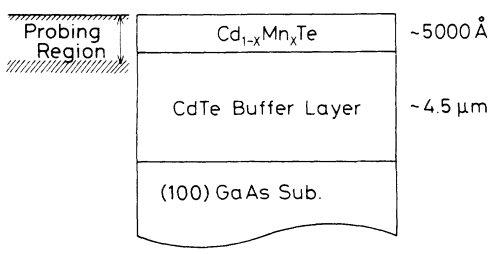

FIG. 1. The stacking structure of the MBE-grown sample.

The time-resolved PL spectra were measured at 4.2 K. In this case, the optical excitation was provided by a R6G dye laser synchronously pumped by a frequency-doubled mode-locked Nd:YAG (yttrium aluminum garnet) laser. Luminescence was collected in the back-scattering geometry, passed through a spectrometer, and finally time resolved using a synchroscan optical streak camera. The time-resolved PL spectra were measured in the Faraday geometry in a magnetic field using a superconducting magnet. The time-resolved PL σ^+ component was collected. The time resolution of the experimental system in the present setup is 55 ps. In all above-mentioned PL measurements, the excitation light power was kept less than 100 mW/cm^2.

The energy position of the free-exciton peak E_{exc} in $Cd_{1-x}Mn_xTe$ can be expressed as a function of composition x, as follows:

$$E_{exc}=E_{exc_0}+1.592x \quad (\text{eV}) , \tag{1}$$

according to a piezomodulation measurement of reflectively reported by Lee and Ramdas,[15] where E_{exc_0} is the peak position of free exciton in CdTe. The composition of $Cd_{1-x}Mn_xTe$ was determined from the position of localized exciton emission in time-integrated PL measurement, assuming that the energy position of localized exciton emission changes as a function of x following this expression. As a result, the Mn concentration in this sample is determined to be 4%.

III. PL PEAK ASSIGNMENT AND SAMPLE CHARACTERIZATION

As has been described above, the $Cd_{1-x}Mn_xTe$ epitaxial film was adjusted to allow the simultaneous observation of PL from both the $Cd_{1-x}Mn_xTe$ epitaxial film and the CdTe buffer layer. In Fig. 2, the time-integrated PL spectra are shown by the broken line. The solid lines show the result of the spectral-decomposition analysis. As the energy gap of CdTe is 1.606 eV (Ref. 16) (about 12 950 cm^{-1}) and it increases with increasing x, it is obvious that the excitonic luminescences above 12 950 cm^{-1} are those from $Cd_{1-x}Mn_xTe$ while the luminescences below 12 950 cm^{-1} are arising from the CdTe buffer layer. In the curve-fitting analysis CdTe PL spectra were decomposed by Lorentzian curves, while for $Cd_{1-x}Mn_xTe$ the spectra were decomposed by Gaussian curves, because the $Cd_{1-x}Mn_xTe$ spectra were dominated by the inhomogeneous broadening.

First, we will describe the origin of the lines from the

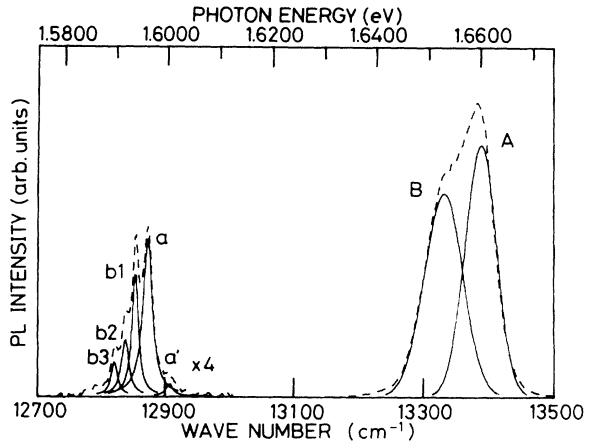

FIG. 2. The time-integrated photoluminescence spectra at 4.2 K of $Cd_{1-x}Mn_xTe$ ($x=4\%$) ($>12\,950$ cm^{-1}) and CdTe buffer layer ($\leq 12\,950$ cm^{-1}) are shown by the broken lines. The spectra shown by the solid lines are the results of the spectral-decomposition analysis.

CdTe buffer layer. The assignment can be done by comparing the present lines with those observed in previous works.[12–14,17]

(i) Lines a and a'; emissions of two kinds of free excitons. Lattice mismatch between the CdTe buffer layer and the GaAs substrate induces a biaxial stress perpendicular to the growth direction. This stress is partially relaxed by misfit dislocation, but still remains in the surface region.[17] The free-exciton emission line splits into heavy-hole-related (line a) and light-hole-related (line a') lines due to the uniaxial component induced by this biaxial stress. The splitting is about 4.3 meV and comparable with the result reported by Tatsuoka *et al.*[17]

(ii) Line b; emissions of bound excitons. Possible assignment can be done on the basis of the previous work as follows: line $b1$ (1.5933 eV) is assigned to the emission of an exciton bound to a neutral donor (D^0,X);[12,14] line $b2$ (1.5914 eV) is assigned to the emission of an exciton bound to either a neutral acceptor (A^0,X) (Ref. 12) or an ionized donor (D^+,X);[14] and line $b3$ (1.5893 eV) is assigned to the emission of an exciton bound to a neutral acceptor (A^0,X).[13,14]

In $Cd_{1-x}Mn_xTe$ PL spectra, because of the inhomogeneous broadening, the splitting between lines a and a' and that among lines $b1$, $b2$, and $b3$ was smeared out. However, a reasonable assignment of the $Cd_{1-x}Mn_xTe$ PL spectra can be done by comparing the observed intensities with those of PL lines in CdTe as follows.

(i) Line A; an emission of a free exciton. The line is affected by the fluctuating potentials, so it can be called a localized exciton.

(ii) Line B; an emission of a bound exciton. These spectra in $Cd_{1-x}Mn_xTe$ are similar to those of $Cd_{1-x}Mn_xTe$ ($x=5\%$) reported by Golnik, Ginter, and Gaj.[4] They denoted line A and line B as $L2$ and $L1$, respectively. The intensity ratio of line A to line B in the present spectra is opposite to their result, but it is consistent with that of line a to line b in the CdTe PL domain of the present sample. Bulk samples with stronger emissions from localized excitons than from bound excitons in $Cd_{1-x}Mn_xTe$ with $x \lesssim 5\%$ have not yet been grown. Using the epitaxial $Cd_{1-x}Mn_xTe$ film discussed in this paper, the dynamical behavior of both the localized and the bound excitons in $Cd_{1-x}Mn_xTe$ can be investigated in the same sample.

The intensity ratio of line A to line B is plotted against temperature in Fig. 3. Above 20 K, this ratio decreases rapidly. The decrease of this ratio with temperature can be interpreted as the thermal activation of excitons from bound states, so this temperature dependence supports the assignment of line B as being due to bound excitons.

In Fig. 4, the magnetic-field dependence of the PL spectra in $Cd_{1-x}Mn_xTe$ is shown by broken lines. Furthermore, the decomposed spectra lines are shown by solid lines. A pronounced redshift due to the enhanced Zeeman effect was observed. It is also clearly seen that the intensity ratio of line B to line A decreases with increasing magnetic field. The decrease of the intensity of line B can be attributed to magnetic-field-induced exciton transitions as described by Heiman *et al.*[3] According to them, the (D^0,X) complex has two weakly bound electrons around a positive impurity center. The electron spins are paired antiparallel in the absence of external magnetic field, but one of the electrons can lower its energy by going to a larger orbit by flipping its spin when in the presence of an applied magnetic field. Therefore, although the (D^0,X) state is stable in zero magnetic field, it is expected to be unstable in nonzero magnetic fields. The (A^0,X) state is also unstable in a magnetic field for almost the same reason as for the (D^0,X) state. The field-induced behavior of the decomposed line intensity is consistent with that observed by Heiman *et al.*, and thus supports the assignment of the spectra as described above.

Figure 5 shows a comparison of the magnetic-field dependence of line A with that of line a. They are plotted by open and closed circles, respectively. According to Gaj, Planel, and Fishman, the photoluminescence peak position of free excitons (localized exciton in the present case) in $Cd_{1-x}Mn_xTe$, E_{EX}, is expressed as follows:[1]

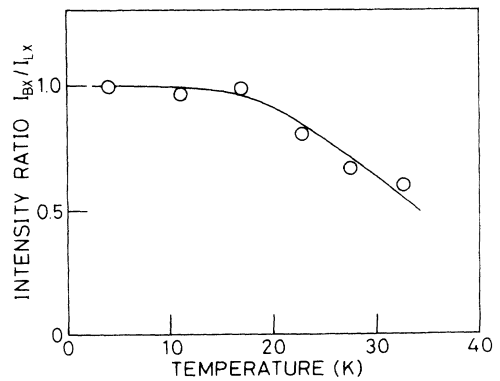

FIG. 3. The temperature dependence of the intensity ratio of line B to line A. Line A and line B are assigned in Fig. 2 to the emissions from localized and bound excitons. Therefore, the ordinate of the figure shows the intensity ratio of the bound exciton emission (I_{BX}) to the localized exciton emission (I_{LX}).

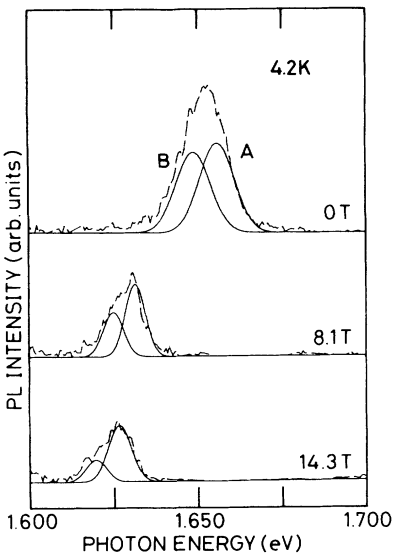

FIG. 4. The magnetic-field dependence of the time-integrated photoluminescence spectra at 4.2 K are shown by the broken lines. The spectral-decomposed spectra are shown by the solid lines.

$$E_{EX}=E_0-E_Z \ , \tag{2}$$

$$E_Z=(\alpha-\beta)N_0x\langle S_z\rangle_{Mn}/2 \ , \tag{3}$$

where E_0 is the exciton energy without a magnetic field, α and β are the exchange integrals for the conduction- and valence-band electrons, respectively. N_0 is the number of unit cells per unit volume, x is the Mn concentrations, and $\langle S_z\rangle_{Mn}$ is the thermal average of the Mn^{2+} spin component along the magnetic field. According to their measurements, $N_0\alpha$ and $N_0\beta$ are 0.22 and -0.88 eV, respectively. The recombination responsible for the photoluminescence takes place between the conduction band $|\frac{1}{2},-\frac{1}{2}\rangle$ and the valence band $|\frac{3}{2},-\frac{3}{2}\rangle$. It is noted that Eqs. (2) and (3) are correct only under the condition that the Zeeman shift is large enough to neglect other effects, such as the diamagnetic effect of excitons.

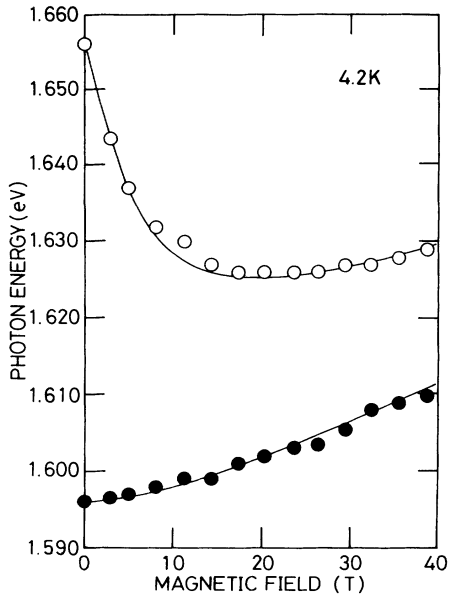

FIG. 5. The magnetic-field dependence of the peak position of line A at 4.2 K (open circles) and that of line a (closed circles).

In the present result, because the diamagnetic shift cannot be neglected in the higher magnetic-field region, we have compensated for the effect of the diamagnetic shift through the following procedure. The energy level of a hydrogenlike system in a constant magnetic field of arbitrary strength was calculated by Makado and McGill.[18] Using their result, the luminescence peak position is expressed as the revised expression

$$E_{EX}=E_0+E_1-E_Z \ , \tag{4}$$

where E_1 is exciton energy in a nonmagnetic semiconductor, calculated by Makado and McGill. Using the experimentally determined values[1,19] of $\varepsilon_0=10.2$, $N_0\alpha=0.22$ eV, and $N_0\beta=-0.88$ eV, the magnetic-field dependence of the PL position can be calculated from Eq. (4) using a parameter-fitting method, as shown by the solid lines in Fig. 5. In this analysis, we assume that $\langle S_z\rangle_{Mn}$ can be expressed by the empirical equation[20]

$$\langle S_z\rangle_{Mn}=SB_{5/2}[5\mu_BH/k_B(T+T_0)] \ , \tag{5}$$

where $B_{5/2}$ is the Brillouin function for spin, $\frac{5}{2}$, μ_B is the Bohr magneton, k_B is the Boltzmann constant, H is the magnetic field, and T is the temperature. S and T_0 are treated as the fitting parameters. S is expected to be less than $\frac{5}{2}$ due to strong antiferromagnetic coupling between Mn^{2+} ions, and T_0 is expected to be positive due to more-distant-neighbor coupling of Mn ions.

Fitting parameters are shown as follows: (i) For line A (localized exciton of $Cd_{1-x}Mn_xTe$),

$$S=1.6, \quad T_0=8\,\text{K}, \quad \mu=0.11m_e \ ;$$

(ii) For line a (heavy-hole-related free exciton of CdTe),

$$S=0, \quad \mu=0.08m_e \ ,$$

where μ is the effective mass of exciton and m_e is the free-electron mass. The value of μ in CdTe is in good agreement with the result reported in a previous study.[16] Furthermore, the value of T_0 is also comparable with the result obtained in a magnetization measurement.[20]

IV. TIME-RESOLVED PL STUDY IN MAGNETIC FIELDS

The time-resolved PL spectra in magnetic fields have been measured at 4.2 K. The decomposed peak positions of localized and bound excitons for $Cd_{1-x}Mn_xTe$ are plotted versus time in Fig. 6 for various magnetic fields. In this figure, the time dependences of localized and bound excitons are shown by open and closed symbols, respectively. The horizontal axis denotes the time measured from that at which the intensity of excitation laser pulse is maximum.

In an applied magnetic field, Mn spins are polarized.

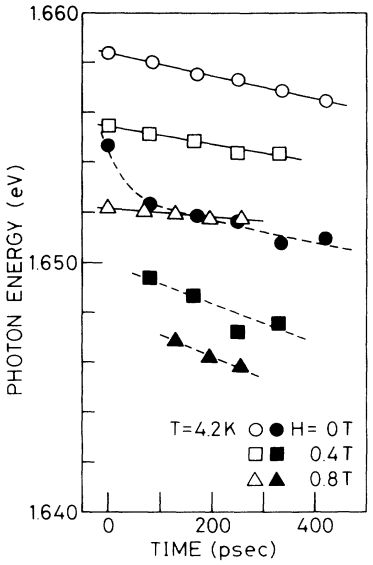

FIG. 6. The time dependence of the peak positions of line A (localized exciton emission) and line B (bound exciton emission) at 4.2 K in various magnetic fields are plotted by open and closed symbols, respectively.

Therefore, the temporal energy shift due to the magnetic polaron formation is expected to become smaller with the increase of magnetic field.[7,21] On the basis of this interpretation, the temporal energy shift at zero field and its suppression in a magnetic field are evidence of excitonic magnetic polaron formation. The temporal energy shift of the localized exciton was observed as about 5 meV/ns at 0 T and it was suppressed to about 1.5 meV/ns at 0.8 T as shown in Fig. 6. Therefore, the temporal energy shift observed during 400 ps is considered to arise from the LMP formation. The magnetic polaron formation time for the LMP state in the present sample is longer than the results in previous studies in $Cd_{0.8}Mn_{0.2}Te$.[11,21] This difference may stem from the difference in the Mn concentration of the investigated samples. The fact that the decay time increases with decreasing x can be explained by magnetic dilution[22–24] or spin diffusion.[25]

For the bound exciton emission, a fast energy shift within about 100 ps and a slower energy shift after about 100 ps were observed, as shown by the closed circles. It is possible that the faster energy shift is due to the magnetic polaron formation arising from the same two carriers in the bound exciton state [electrons in the (D^0,X) state and holes in the (A^0,X) state].[25] Since the carriers are expected to be rather localized, a carrier-induced magnetic polaron state is expected to form rapidly. On the other hand, the slower energy shift is considered to be due to the magnetic polaron formation in the expanded wave function of the other carrier.

In an applied magnetic field, the faster energy shift disappears. We attribute this disappearance to the instability of the same two carriers in a bound exciton in the presence of a magnetic field.[3] The instability is explained by the same scheme as described in the magnetic-field dependence of the bound exciton in Fig. 4. On the basis of this concept, the slower energy shift, which was enhanced by the applied magnetic field, can also be interpreted as follows: The bound excitons are released from the impurities by the magnetic field, and then relax to lower energy states arising from the inhomogeneous broadening. In a magnetic field, the enhanced energy shift is dominated by this process.

V. CONCLUSION

Detailed studies on the excitonic emission of MBE-grown $Cd_{1-x}Mn_xTe$ sample with $x=4\%$ have been performed. The localized and bound exciton emission lines were observed in the photoluminescence (PL) spectra, and the magnetic-field dependent and the time-dependent behavior of these excitonic emissions have been investigated. The former measurement has elucidated the exciton properties in the sample; the effective mass of the localized exciton of $Cd_{0.96}Mn_{0.04}Te$ was measured to be $0.11m_e$. In the time-resolved PL spectra, the temporal energy shift and its suppression in a magnetic field were observed for localized exciton emissions during about 400 ps. These facts suggest that LMP's are formed from the localized excitons. On the other hand, the faster temporal energy shift within about 100 ps and the slower energy shift following the fast shift are observed in the bound exciton emission at 0 T. The faster and slower energy shifts can be assigned to those due to the magnetic polaron formation by the same kind of trapped carriers and another kind of expanded carrier in the bound exciton state, respectively. Since the fast energy shift disappears in nonzero magnetic fields, it is considered that the BMP state is more unstable than the LMP state in the presence of a magnetic field, because the spins of the same kind of carriers are paired antiparallel in the bound exciton state. On the other hand, the slower energy shift is enhanced in an external magnetic field. The enhanced energy shift may be due to excitons released from the bound state by the applied magnetic field.

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