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High-pressure and high-magnetic-field study of energy transfer from excitons into local d electrons in a CdTe/(Cd, Mn)Te quantum well structure

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Abstract

Photoluminescence measurements have been conducted for a CdTe/Cd$_{1-x}$Mn$_x$Te ($x = 0.4$) single-quantum-well structure at low temperatures under pressures to 0.49 GPa and magnetic fields to 60 T. At the ambient pressure, a new emission was induced by the application of a magnetic field. The emission has been assigned to exciton emission from the barrier layer, which is suppressed below 9 T due to the energy transfer from the exciton to local d electrons. At 0.49 GPa, the emission recovered at 44 T. In the field region where the energy transfer occurs, an anomalous red-shift of the exciton energy was observed clearly for the case of the ambient pressure. The alloy potential fluctuation effect and the magnetopolaron effect are examined as candidates for the mechanism to cause this phenomenon.

1. Introduction

In (II, Mn)VI semiconductors, the so-called diluted magnetic semiconductors, energy transfer could occur between an extended electron and a local d electron. It has been reported that the exciton emission becomes smaller in bulk Cd$_{1-x}$Mn$_x$Te with increasing Mn concentration, and this occurs together with an enhancement of the Mn^{2+} $^4T_1 \rightarrow {}^6A_1$ luminescence [1]. It was suggested that the energy transfer occurs when the exciton energy overlaps with the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation band as the Mn concentration is increased.

Instead of changing the Mn concentration, the exciton energy and the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation band can be adjusted by the application of hydrostatic pressures and magnetic fields [2]. To apply pulsed high magnetic fields to a sample under high hydrostatic pressures had been difficult, because increase of the sample temperature cannot be neglected when using conventional metallic pressure cells under pulsed fields. This problem is resolved by using recently developed diamond anvil cells (DACs) with plastic bodies [3]. Taking advantage of high hydrostatic pressures and high magnetic fields, one can adjust the exciton energy and the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation band continuously and investigate the energy transfer in diluted magnetic semiconductors in more detail.

We have already reported a novel phenomenon: an exciton emission is extinguished by the application of high pressures and the emission is recovered by applying magnetic fields in photoluminescence (PL) measurements at low temperatures for a 1.0 nm thick CdTe/$Cd_{1-x}Mn_xTe$ ($x = 0.4$) single quantum well (SQW); this is attributed to the energy-level crossing of the exciton energy and the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation band induced by pressures and magnetic fields [2]. In the present work, we have observed a similar phenomenon for the barrier layer of the same sample. Moreover, we have found that the Zeeman shift of the exciton emission energy for the barrier shows anomalous nonlinear behaviour. We examine the mechanism of the anomalous Zeeman shift through a high-pressure and high-magnetic-field study.

2. Experimental details

The sample that we used for this study has a SQW structure which consists of 1.0, 1.9 and 3.8 nm thick CdTe quantum wells (QWs) separated by 48 nm thick $Cd_{1-x}Mn_xTe$ ($x = 0.4$) barriers. The Mn mole fraction was estimated from the composition of the raw materials. Hybrid buffer layers between the QWs and a (100) GaAs substrate were constructed.

External magnetic fields to 60 T were generated by a long-pulsed magnet with a 32 mm bore and typical pulse duration of 2 s. The magnet was charged with a 1430 MV A/600 MJ inertial energy storage generator. Field profiles could be modified flexibly by controlling converters that drive three parts of the magnet independently.

A plastic DAC was employed in order to avoid increase of sample temperature due to the eddy current during application of the pulsed fields [3]. The column-shaped DAC was 8.9 mm in diameter and 15.5 mm in height. There was no metallic component except for a small gasket in the DAC. No significant increase in sample temperature was observed during the field pulses. A methanol:ethanol:water 16:3:1 mixture was used as the pressure-transmitting medium. Pressure was calibrated by measuring the shift of the R_1 fluorescence line of ruby. The DAC was immersed in liquid helium for the measurements at the temperature 4.0 K.

Excitation light of 442 nm from a He–Cd laser was transferred to the DAC through a single optical fibre. PL signals were transferred back through the same fibre to a spectrometer equipped with a liquid-nitrogen-cooled back-illuminated charge-coupled device (CCD) (Princeton Instruments, LN/CCD-100EB). Each spectrum was recorded at a rate of one per 2.1 ms. The two-second-long pulse of the magnet allowed us to take very clean PL spectra at intervals smaller than 0.3 T.

3. Results and discussion

At the ambient pressure, we observed PL signals assigned to the exciton emissions from the three QWs, and the Mn^{2+} $^4T_1 \rightarrow {}^6A_1$ transition at low temperatures, whereas there was no signal for an exciton emission from the barrier layer [2]. At about 9 T, a new emission peak emerged around 2.187 eV. The peak was observed to increase its intensity and shift toward lower energies with increasing magnetic field as shown in figure 1(a). At 0.49 GPa, a new

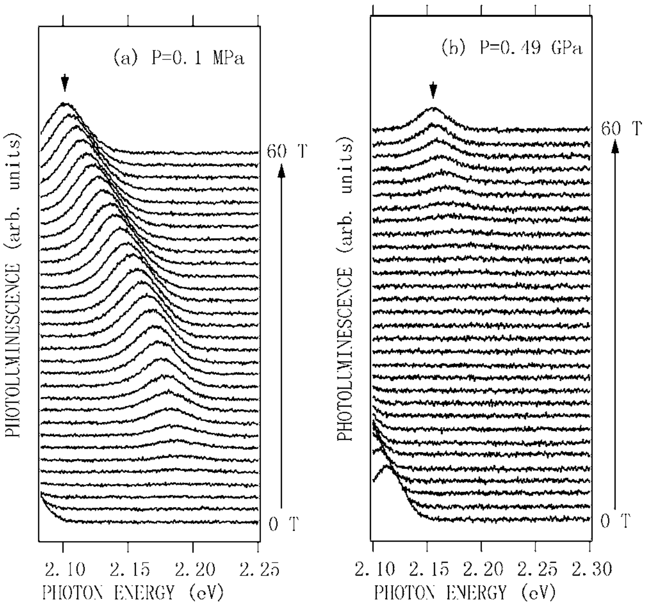

Figure 1. Magnetic field expansion of PL spectra at intervals of 2 T for a CdTe/$Cd_{1-x}Mn_xTe$ ($x = 0.4$) SQW structure at 4.0 K and the pressures of (a) 0.1 MPa and (b) 0.49 GPa, in the energy range (a) between 2.083 and 2.25 eV and (b) between 2.10 and 2.30 eV. A new emission peak indicated by an arrow is assigned to the exciton PL from the barrier layer (see the text).

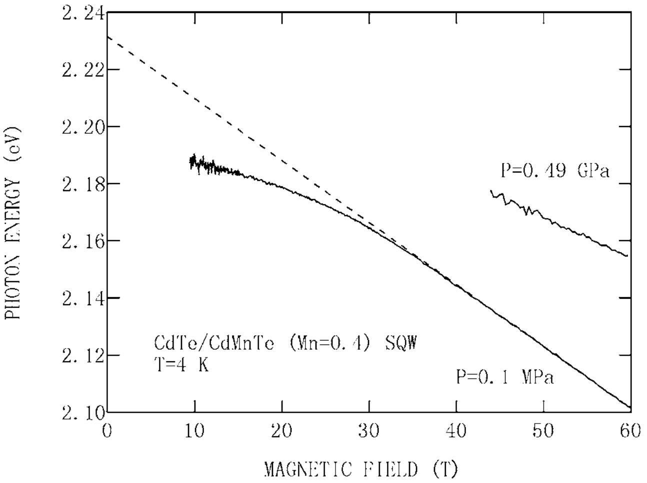

Figure 2. The magnetic field dependence of the emission peak energy for excitons in the barrier at the pressures of 0.1 MPa and 0.49 GPa. The broken line represents a linear function fitted to the data for 0.1 MPa between 40 and 60 T.

peak appeared at higher fields as shown in figure 1(b). Figure 2 exhibits the magnetic field dependence of the emission peak energy of the new peaks.

We have assigned the new peak to an exciton emission from the barrier as explained in the following. The exciton energy was observed to depend linearly on the magnetic field in the region of 40 and 60 T. We fitted a linear function to the experimental data in this region by the

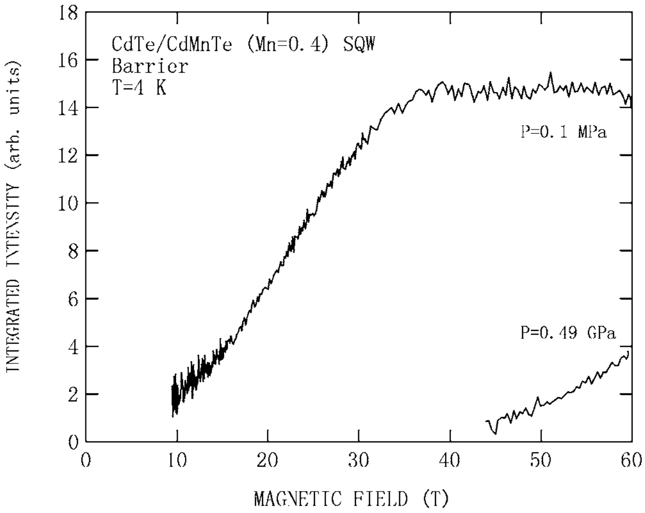

Figure 3. The magnetic field dependence of the exciton emission intensity for the barrier layer at 0.1 MPa and 0.49 GPa.

least-squares method as shown with a broken line in figure 2, and have found the value at zero field and the slope of the function to be 2.2314 eV (E_1) and −2.1693 meV T^{-1} (k_1), respectively. According to a photoreflectivity study [4], the energy of a free exciton is 2.2318 eV in bulk $Cd_{1-x}Mn_xTe$ ($x = 0.4$). This value is quite close to the zero-field value E_1 mentioned above. In addition, the magnetization curve of bulk $Cd_{1-x}Mn_xTe$ ($0.1 \leqslant x \leqslant 0.5$) [5] and the Zeeman shift of the exciton energy for a $Cd_{1-x}Mn_xTe$ ($x = 0.24$) barrier in a CdTe/$Cd_{1-x}Mn_xTe$ ($x = 0.24$) QW structure [6] have been determined in a field region up to 50 T already. As the sp–d coupling constant is independent of the Mn mole fraction in (Cd, Mn)Te [7], one can estimate the slope of the Zeeman shift of the exciton energy for the $Cd_{1-x}Mn_xTe$ ($x = 0.4$) barrier in the vicinity of 50 T, where the Zeeman shift shows a linear field dependence due to the spin–spin coupling in small Mn clusters and the random network [5]. We have thus estimated it at -2.3690 ± 0.4738 meV T^{-1}, which agrees well with the value k_1 obtained for the new peak. Judging from the agreement in these comparisons, we have assigned the new peak to an exciton emission from the $Cd_{1-x}Mn_xTe$ ($x = 0.4$) barrier layer.

The mechanism by which the exciton emission recovers on the application of magnetic fields is thought to be same as that which we proposed for the case of the exciton emission from the 1.0 nm thick QW [2]. In the situation where the exciton energy is higher than the threshold of the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation band, energy transfer from excitons to local Mn d electrons is induced, and the exciton emission is quenched. On the application of magnetic fields, the exciton energy decreases due to the sp–d coupling. The exciton emission recovers when the exciton energy becomes lower than the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation band. The energy where the exciton emission recovered at the ambient pressure was 2.187 eV, which agrees well with the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation threshold energy introduced by Leinen [1]. When the pressure was increased to 0.49 GPa, the peak recovered at lower energy (2.177 eV) than at the ambient pressure. This pressure dependence of the recovery energy agrees qualitatively with our observation for the exciton emission from the QW. We have attributed this behaviour to pressure-induced decrease of the Mn^{2+} $^6A_1 \rightarrow {}^4T_1$ excitation threshold energy, which is suggested on the analogy with the negative pressure dependence of the Mn^{2+} $^4T_1 \rightarrow {}^6A_1$ transition.

The magnetic field dependence of the exciton energy at the ambient pressure in the lower-field region was observed to be smaller by 64% than in the higher region. As the Zeeman shift of the exciton energy is generally almost linear above 15 T, this behaviour is quite peculiar. Judging from the Zeeman shift of the exciton energy for the QW, the magnetic field dependence for both the conduction and the valence band edges is deduced to be linear above 15 T. The deviation of the exciton energy from the linear dependence toward lower energies seems to occur in the process of the exciton formation. The deviation was observed below about 40 T. Figure 3 shows the magnetic field dependence of the integrated intensity of the exciton emission. One notes that the intensity increases monotonically up to 40 T and saturates above that. Energy transfer is thought to occur in the former region. Viewing the magnetic field dependences of the exciton energy and the intensity comprehensively, one sees that the deviation of the exciton energy from the linear field dependence is observed in the region where the energy transfer occurs. This leads to a possibility that the exciton binding energy is enhanced in the situation where the energy transfer is induced.

If we assume the Zeeman shift of the exciton energy to be linear like the broken line in figure 2, the deviation of the exciton energy from the line at 10 T is 22.2 meV. Such a large deviation (anomalous red-shift) was, however, not observed clearly for the pressure of 0.49 GPa and for the QW. We tentatively explain the anomalous red-shift in terms of the alloy potential fluctuation [8]. As the exciton energy fluctuates in position, some excitons with smaller energies can emit radiation by the recombination process even if the average exciton energy is higher than the excitation threshold. With increasing magnetic field, the alloy fluctuation effect would become smaller because of the following.

(1) The Zeeman shift of the exciton energy in a domain of higher Mn mole fraction is larger than that in a domain of lower Mn mole fraction, which would reduce the potential fluctuation, as the former exciton energy is higher at zero field than the latter.
(2) The diamagnetic energy of excitons increases with increasing magnetic field, which would be unfavourable to the localization of the exciton.

The fact that the Zeeman shift of the exciton energy did not show the anomalous red-shift at 0.49 GPa could be attributed to this reduction of the alloy potential fluctuation effect. However, it is difficult to explain the lack of the anomalous red shift in the Zeeman shift for the QW from the viewpoint of the alloy potential fluctuation effect. There is another possibility—the magnetopolaron effect [9]—for the mechanism which causes the anomalous red-shift of the exciton energy. However, the possibility of this would be very slight, because the ratio of isolated local spins to the cation sites is as small as about 0.002 for the Mn mole fraction of 0.4.

4. Conclusions

It is suggested that the recovery of the exciton emission on the application of magnetic fields and the increase of the recovery field by the application of a pressure in the barrier layer of the CdTe/$Cd_{1-x}Mn_xTe$ ($x = 0.4$) SQW structure are caused by energy transfer from excitons to local Mn d electrons. The anomalous red-shift of the exciton energy observed for the barrier at the ambient pressure was not observed for the QW in the SQW structure. The alloy potential fluctuation effect could explain the pressure dependence of the anomalous red shift. However, further examination is required to explain this phenomenon comprehensively, including the lack of the anomalous red shift in the case of quantum effect.

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