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PRESSURE EFFECT ON MAGNETO-OPTICAL PROPERTIES IN CdTe/(Cd,Mn)Te SINGLE QUANTUM WELLS WITH HIGH Mn CONCENTRATION

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The pressure effect on the magnetic field induced type I-type II transition is studied in a CdTe/$Cd_{1-x}Mn_xTe$ (x=0.24) single quantum well (SQW). Photoluminescence (PL) measurements under hydrostatic pressures up to 1.07 GPa and long pulsed magnetic fields up to 60 T with a pulse duration of 2 sec are reported. The pressures were generated in a plastic diamond anvil cell (DAC). A bend toward lower energies (additional red shift) is observed above 28.5 T in magnetic field dependence of the exciton energy for a 13 Å thick quantum well. We attribute this red shift to a phenomenon preceding the type I-type II transition after a comparison with a simple quantum mechanical calculation. The onset field of the additional red shift increases by 3.4 T by applying a pressure of 1.07 GPa. Spin-spin coupling between the exciton and the Mn ion in the interface region is also investigated and found to be enhanced by pressure.

In $CdTe/Cd_{1-x}Mn_xTe$ SQWs and multiple quantum wells (MQWs), the magnetic field induced transition of band alignment from type I to type II has been investigated intensively. The heavy-hole band with $m_j = -3/2$ of the $Cd_{1-x}Mn_xTe$ barrier layers exhibits a large Zeeman shift due to *sp-d* hybridization by the application of magnetic fields. This Zeeman shift could be large enough to exceed a band offset with a sufficiently large field. In MQWs with small x, this type I-type II transition is expected to occur at moderate field strengths. However, no conclusive magneto-optical evidence for the type I-type II transition has been reported in spite of intensive work on MQWs with x of 5-10%.[1-3] For larger x, the type I-type II transition is expected at very high fields. Kuroda *et al.* performed magneto-absorption measurements of MQWs with x of about 30% and found features associated with the type I-type II transition in strong magnetic fields above 80 T.[4] The features include a reduction in the absorption intensity and the onset of a red-shift of the exciton energy, which were in line with results of a variational calculation. In the present work, we have studied the pressure effect on the type I-type II transition in SQWs with x of 24% using PL measurements to monitor the exciton energy shift to fields of 60 T and hydrostatic pressure of 1.07 GPa at T=2 K. In our observation, the onset of a red shift of the exciton energy occurs at much smaller fields than that reported by Kuroda *et al.*[4] The observed feature is compared with a simple quantum mechanical calculation. Contribution of Mn spins at the interface to the exciton states in a quantum well is presented. Its pressure dependence is also discussed.

Pressure is expected to affect the type I-type II transition via *p-d* exchange interaction. It is difficult, however, to perform this study under pressure because pulsed magnets are necessary to obtain the required fields and the large dB/dt associated with these magnets cause eddy current heating in the typical pressure cell which is made of metal. A plastic diamond anvil cell has been developed to address this problem.[5] The columnar shaped DAC is 8.9 mm in diameter and 15.5 mm in height. There was no metallic component except for the small gasket in the DAC. No increase in sample temperature due to eddy current heating 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 sample that we used for this study has a SQW structure which consists of 13, 19 and 38 Å thick CdTe wells separated by 480 Å thick $Cd_{1-x}Mn_xTe$ (x=0.24) barriers. Hybrid buffer layers between the quantum wells and a (100) GaAs substrate were constructed.

A recently commissioned motor-generator driven long pulsed magnet with a 32 mm bore and typical pulse duration of 2 sec was employed. The magnet was charged by a 1430 MVA/600 MJ inertial energy storage generator. Field profiles could be modified flexibly by controlling converters that drive three parts of the magnet independently.

Excitation light of 442 nm from a He-Cd laser was transferred to the DAC through a single optical fiber. Excitation power was about three milliwatts at the top surface of the diamond anvil. PL signals were transferred back through the same fiber to a spectrometer equipped with a liquid-nitrogen-cooled back-illuminated charge-coupled-device (CCD) (Princeton Instruments, LN/CCD-100EB). Each frame recorded on the CCD was transferred to a computer at a rate of one per 2.1 msec. The two second long pulse of the magnet allowed us to take very clean PL spectra every 0.05 T below 15 T and every 0.3 T between 30 and 60 T. The variation in the field during acquisitions in these two regions did not exceed 0.08 T and 0.5 T, respectively.

We observed four luminescence signals assigned to exciton PLs from the three wells and the barrier. Each peak showed a red shift (Zeeman shift) by the application of magnetic fields. We measured the Zeeman shifts for the barrier and the 13 Å thick well at the ambient pressure and 1.07 GPa (Fig. 1). The shift for the barrier shows a Brillouin function like behavior below 15 T and a linear behavior with respect to magnetic fields between 15 T and 60 T. The shift is suppressed by pressure. For the 13 Å thick well, the shift also shows a Brillouin function like behavior below 15 T and a linear behavior between 15 T and 28 T. In contrast to the result for the barrier, an upward bend of the shift (additional red shift) is observed above 28.5 T. By applying pressure of 1.07 GPa, the whole Zeeman shift is suppressed and the onset field of the additional red shift increases by 3.4 T.

The observed features for the barrier is the same as those which we have observed in the magnetic field region up to 30 T in our previous work[6]. The linear field dependence of the Zeeman shift is interpreted as a consequence of the spin-spin coupling in small isolated clusters and a random network.[7] We propose that the suppression of the Zeeman shift is caused by the attenuation of the *p-d* hybridization due to a pressure induced relaxation of the Mn-Te-Mn bond distortion.[6]

The red shift in the magnetic field dependence of the exciton energy is thought to be evidence for the type I-type II transition.[4] The onset has been observed above 80 T in magneto-absorption measurement for MQWs with x of 27 to 32%. In our SQWs with x of 24%, however, the onset is observed around 30 T. We have compared our experimental results with a calculation for a free electron confined in a rectangular finite potential. We made the following assumptions, i) the hole in the system has an effective mass of $0.8m_0$ where m_0 is an electron mass; the value of $0.8m_0$ is generally accepted for the heavy hole mass in CdTe[8], ii) the valence-band offset is 100 meV at 0 T; we have used the valence-band offset ratio of 30% which was reported by Kuhn-Heinrich *et al.*,[9] iii) the offset decreases by 4/5 of the Zeeman shift of the barrier with increasing a magnetic field; the value of 4/5 is derived from $\beta/(\alpha+\beta)$ where α and β are *s-d* and *p-d* exchange constants and their values are reported to be 0.22 and 0.88, respectively,[10] iv) We have neglected the difference of these assumed values between at the ambient pressure and at 1.07 GPa because their changes by a few percents at most do not affect the result of our calculation significantly.

After the calculation, we obtain the shift of the lowest electronic level for the heavy hole as a function of magnetic field (Fig. 2). One can see the shift showing a superlinear behavior above about 30 T, where the lowest heavy hole level decreases acceleratively with decreasing the band offset below 50 meV. By subtracting the shift from the observed

Zeeman shift for the 13 Å thick well, the red shift is nearly canceled. This result means that the additional red shift indicates the beginning of the type I-type II transition. We attribute our success of this observation to better resolutions of magnetic fields and PL spectra in our measurement with the 60 T-long pulsed magnet than those in the previous magneto-absorption measurement. The increase of the onset with increasing pressure would bring the first experimental suggestion for pressure effect on the type I-type II transition. We estimate the type transition fields at about 65 T for the ambient pressure which agrees well with the previous result[4], and about 76 T for 1.07 GPa, judging from the Zeeman shift for the barrier. Further investigation with higher magnetic fields is required in order to clarify the pressure dependence of the transition field.

The remainder of the above subtraction enables us to discuss qualitatively the contribution of spins of Mn ions in the interface region to the exciton Zeeman shift. The probability that a Mn ion finds another in the nearest cation sites is much smaller in the interface region than that in the inside of the barrier layer. The remainder shows a Brillouin function like behavior, which suggests that isolated spins in the interface region seem to contribute to the remaining magnetic shift mainly. The magnetization of isolated spins is saturated above 20 T at 2 K. One can see increase of the shift around 20 T with increasing pressure. This tendency is in good agreement with pressure dependence of a Zeeman shift reported for bulk (Cd,Mn)Se with Mn concentration smaller than 5%[11] and could be attributed to pressure caused enhancement of the *p-d* interaction in the interface region. We interpret that the decrease of the Mn-Te bond length by pressure affects on the *p-d* hybridization more significantly in this region than the relaxation of the distortion in the Mn-Te-Mn bond because the Mn concentration is practically low in the region.

In conclusion, a phenomenon associated with the onset of a type I-type II transition induced by a magnetic field in a CdTe/ $Cd_{1-x}Mn_xTe$ (x=0.24) SQWs is observed in the PL measurement of the exciton energy. Using the 60 T long pulsed magnet and the plastic diamond anvil cell, the onset is shown clearly to move toward higher magnetic field with increasing pressure. This pressure dependence is attributed mainly to the suppression of Zeeman shift in the barrier layers. In addition, the enhancement of the spin-spin coupling between the exciton and the Mn ion in the interface region by pressure is shown qualitatively.

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List of Figure Captions

Figure 1 Zeeman shifts of the exciton energies for the 13 Å thick quantum (labeled as QW4ML, left axis) and the barrier (right axis) in the $CdTe/Cd_{1-x}Mn_xTe$ ($x=0.24$) SQWs at ambient pressure and 1.07 GPa and T=2 K as a function of magnetic field. Dots are experimental data taken during one field pulse. The arrows show onset of the bend in the Zeeman shifts.

Figure 2 Experimental data for the Zeeman shifts of the exciton energies (Z_0 and Z_1), shifts of the lowest electronic level for the heavy hole (S_0 and S_1) and the corrected Zeeman shifts (R_0 and R_1) in the 13 Å well at the ambient pressure and 1.07 GPa, respectively, as a function of magnetic field.

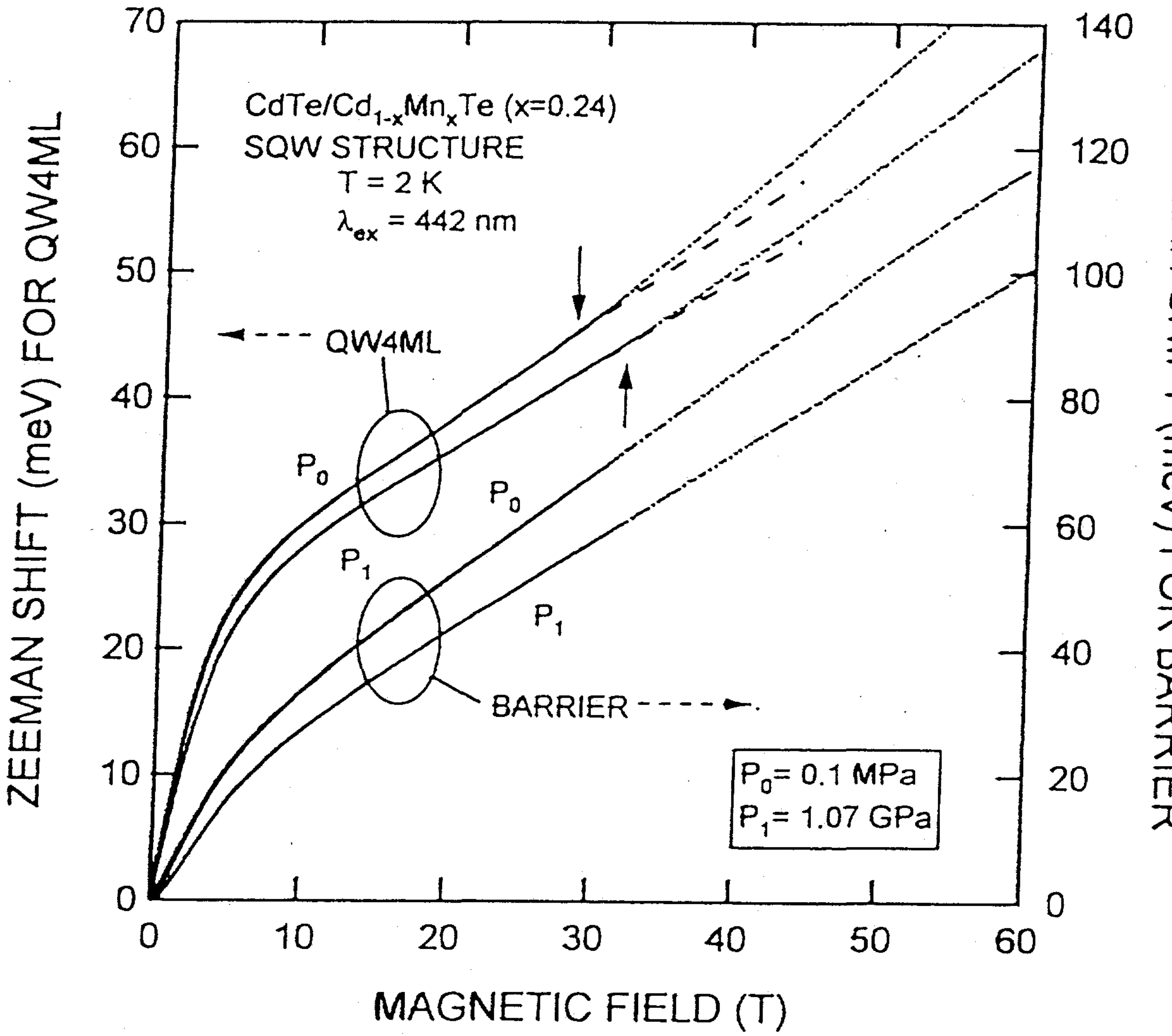

Figure 1, Hiroyuki Yokoi, J. Appl. Phys.

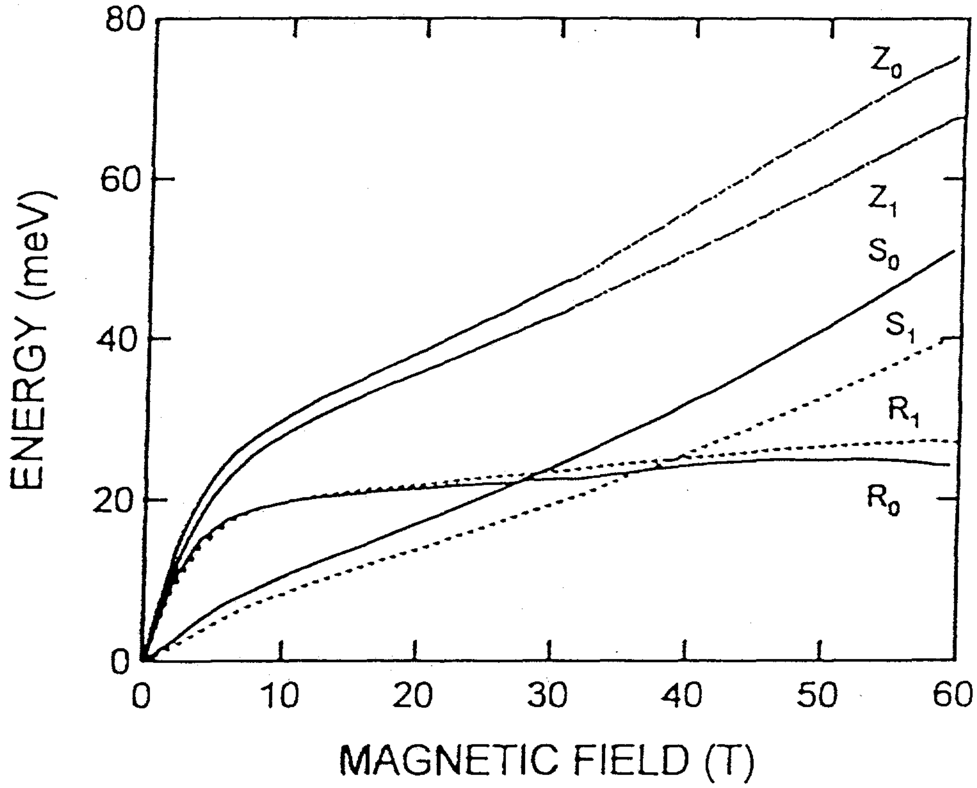

Figure 2, Hiroyuki Yokoi, J. Appl. Phys.