Magneto-Optical Properties and Exciton Dynamics in Nanostructure Diluted Magnetic Semiconductors: $Cd_{1-x}Mn_xSe$ Quantum Dots and $Cd_{1-x}Mn_xTe/ZnTe$ Quantum Wells

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Abstract

Magneto-optical effects and dynamics of excitons are reported in the quantum-dot (QD) and quantum-well (QW) structures of diluted magnetic semiconductors. QDs of $Cd_{1-x}Mn_xSe$ show the excitonic magnetic polaron effects with a increased binding energy of 20 meV. In the $Cd_{1-x}Mn_xTe/ZnTe$ QW system (x=0-0.2), the dephasing process of the free QW exciton is measured by the transient four wave mixing, where the measured dephasing time T_2 of 360-450 fs indicates the fast dephasing process influenced by the strains existing in the QWs. In the asymmetric double QWs, the tunneling time of carriers between the QWs through the barrier is determined as 0.01-10^6 ps. The enhanced excitonic magnetic polaron binding energy, the rapid dephasing of free excitons and the magnetically affected tunneling of carriers are the particular dynamics in the DMS nanostructures.

1. Introduction

Nanometer-scale structures of diluted magnetic semiconductors (DMSs) show marked magneto-optical properties owing to the confinement effect of band-electronic states. Magnetic interaction, that is the exchange interaction, takes place between the electrons (holes) and the involved magnetic ions in the nanostructures. In the quantum dots (QDs) of $Cd_{1-x}Mn_xSe$, the zero-dimensional structures induces highly confined states for electrons and excitons. A quantum well (QW) composed of $Cd_{1-x}Mn_xTe$ and ZnTe is a strained QW in which the $Cd_{1-x}Mn_xTe$ well layer is compressed by the ZnTe barrier layers owing to the mismatch (~6%) of the lattice constants. The carriers and the excitons in the $Cd_{1-x}Mn_xTe/ZnTe$ QW system are affected by both the strain and magnetic effects. In this paper we report the magneto-optical properties and the exciton dynamics in the QDs, the multiquantum wells (MQW) and the asymmetric double quantum wells (ADQW) of DMSs.[1,2]

2. Experimental Details

$Cd_{1-x}Mn_xSe$ QDs have been grown in the SiO_2 matrices by rf sputtering and subsequent heat treatment. Typical diameter of the QDs is 20-400 Å and the Mn concentration x is 0.05-0.2. MQWs and ADQWs of $Cd_{1-x}Mn_xTe/ZnTe$ (x=0-0.2) were prepared by the hot wall epitaxy method. The MQWs consist of the well layers with the width of 10-124 Å and the barriers of 8-35 Å. The period of the MQWs is 35-100. The effect of the strain in the $Cd_{1-x}Mn_xTe$ well layer due to the lattice mismatch exists in these MQWs. The ADQW samples of $Cd_{1-x}Mn_xTe$ /ZnTe (x = 0.1) were fabricated with the width of the narrow well (NW) of $Cd_{1-x}Mn_xTe$, L_{NW} = 20 Å and that of the wide well (WW), L_{WW} = 30 Å, which are separated by the ZnTe barrier of 40-320 Å thickness.

Magneto-optical effects and the exciton dynamics ware studied by the time resolved photoluminescence (TRPL), the degenerate four wave mixing (DFWM) and the pump-and-probe differential absorption spectroscopy (PPAS). The excitation light sources in these optical measurements were 130 fs pulses of a mode-locked Ti-sapphire laser and the amplification system.

3. Experimental Results and Discussion

Transient characteristics of the exciton luminescence in $Cd_{1-x}Mn_xSe$ QDs (the average diameter is 220 Å) is shown in Fig. 1 as a contour map of the luminescence intensity.[3]

The exciton luminescence at 0 T decays with the time constant of 100 ps in the higher energy region at 2.03 eV, while in the lower energy region around 1.94 eV the luminescence decay is much slower. At 5 T the exciton luminescence shows much faster decays. The lifetime of the QD excitons deduced from the luminescence decay is shown in Fig. 2 as a function of the QD diameter, where the lifetime is distributed in the range of 100 ps - 2 ns. The variation is caused by the difference in the exciton lifetime in the size-distributed QDs and shows the dynamical process of the excitonic magnetic polaron annihilation.[4] In magnetic field of 5 T, the exciton lifetime decreases markedly due to the increase of the radiative recombination rate, which is caused by the level crossing of the singlet and triplet exciton states in magnetic field. Therefore the marked decrease of the exciton lifetime by the magnetic field also displays the evidence of the exchange interaction of the exciton with the Mn ions in the QDs. The interaction enables the exciton in the QDs to form the excitonic magnetic polaron.

The average energy of the excitons in the QDs is given by the gravity center of the time-resolved spectra in the contour maps. At 0 T, the decay time for the average energy of the excitons is 900 ps, where the average energy decreases by 40 meV. On the other hand, at 5 T, the increased radiative recombination rate induces a fast decay of the exciton luminescence and the time constant for the decay of the average energy becomes 440 ps, where the energy shift of the average energy to the lower energy increases to 60 meV. The observed variation of the exciton luminescence shown in Fig. 1 displays the formation process of the excitonic magnetic polaron in the individual QDs and the radiative energy transfer of the exciton energy from the smaller QDs to the larger ones as well as the radiative decay of the excitonic magnetic polaron in the QDs. Recently the magnetic polaron binding energy of the exciton state in DMS QDs has been calculated.[5] The fairly good agreement between the experimental results of $Cd_{1-X}Mn_XSe$ QDs with the calculation shows marked increase of the exchange interaction in the QDs due to the confinement effect.

Exciton dynamics in the DMS QWs is studied by the transient nonlinear spectroscopies. Fig. 3 shows the DFWM signals in the MQWs (CZ1: x = 0, L_W = 124 Å, L_b = 8 Å, the periods P = 35) at the resonance excitation on the exciton state, with parallel and perpendicular polarization configurations for the two incident beams[6]. In the perpendicular polarization configuration, the DFWM signal is mostly decayed with T_2 of 360 fs. A doubly-decayed signal is observed in the parallel configuration,

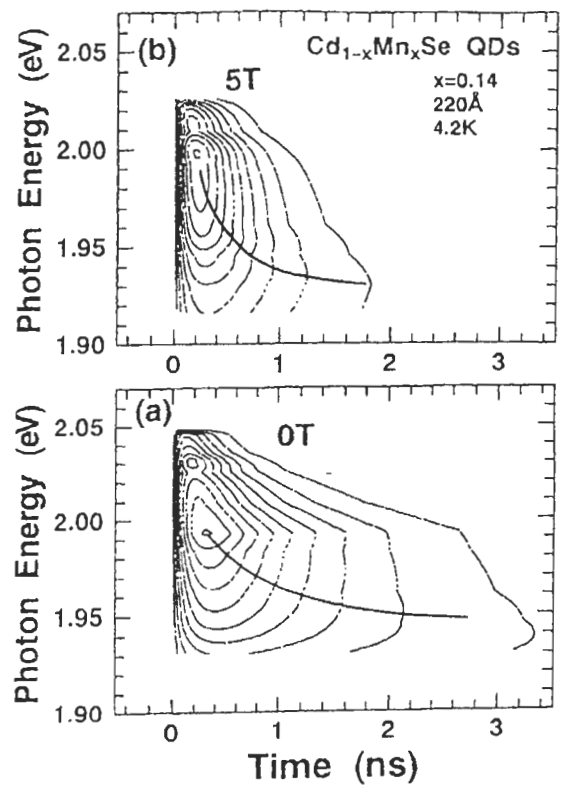

Fig. 1 Time variation of luminescence in the $Cd_{1-X}Mn_XSe$ QDs at H=0 T (a) and 5 T (b).

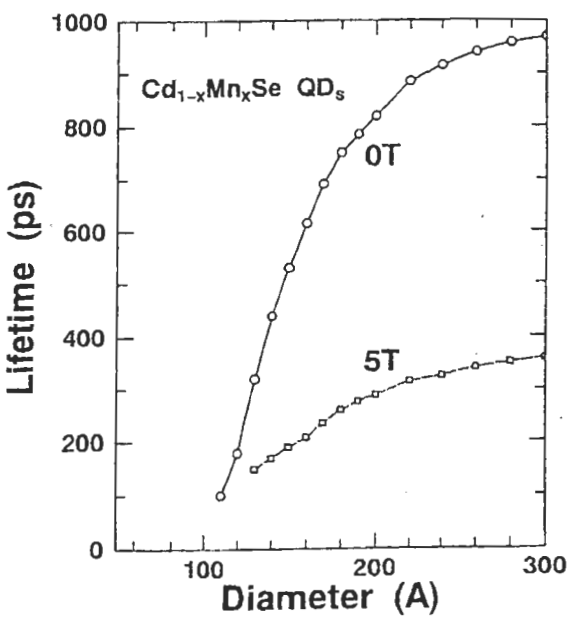

Fig. 2 Lifetime of the exciton in the QDs as a function of the diameter.

where the fast decay component corresponds to the dephasing time T_2 = 450 fs. This fast dephasing is caused for the free exciton in the QWs, which is confirmed by the resonance behavior of the component at the exciton absorption peak as shown in Fig. 4. The signal intensity ratio for the perpendicular to parallel configurations in Fig. 3 is 1/5, which is much higher than that observed in GaAs/GaAlAs QWs (1/100 in this case). Therefore the origin of the signal in the perpendicular configuration is different from the biexciton model of DFWM.[7,8]. The slow decay component in the parallel configuration is due to the dephasing process of the localized excitons. The slow decay of 6.4 ps partly involves the longitudinal relaxation rate of the exciton in the QW by the effective pump and probe measurement.

The observed short T_2 of the QW exciton is independent of the lattice temperature, which indicates a less affection of acoustic and optical phonon relaxations than in the GaAs system. The fast dephasing of the exciton in the CdTe/ZnTe system is caused by the relaxation due to the lattice inhomogeneity involved in the present strained QWs because the free exciton is much affected by the inhomogeneous relaxation of the lattice. The DFWM signal in the perpendicular polarization is also induced by the lattice inhomogeneity by partial breaking of the polarization selection rule for the QW exciton transition. Present experimental results display the dynamics and the dephasing process of the excitons in the strained quantum wells.

Fig. 5 shows the PPAS spectra of the $Cd_{0.84}Mn_{0.16}Te/ZnTe$ MQWs with L_W = 23 Å and L_b = 33 Å.[9] The spectra shown in the top are the steady state absorption and luminescence spectra of the exciton. The exciton in this MQW locates at 2.13 eV, while the exciton luminescence peak is shifted by 100

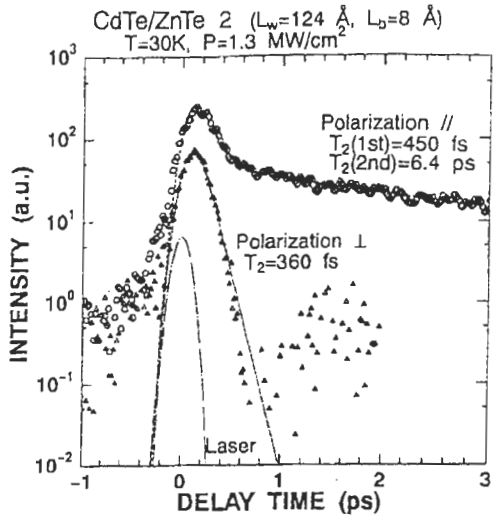

Fig. 3 DFWM signals in the sample CZ1 for the parallel and perpendicular polarization configurations.

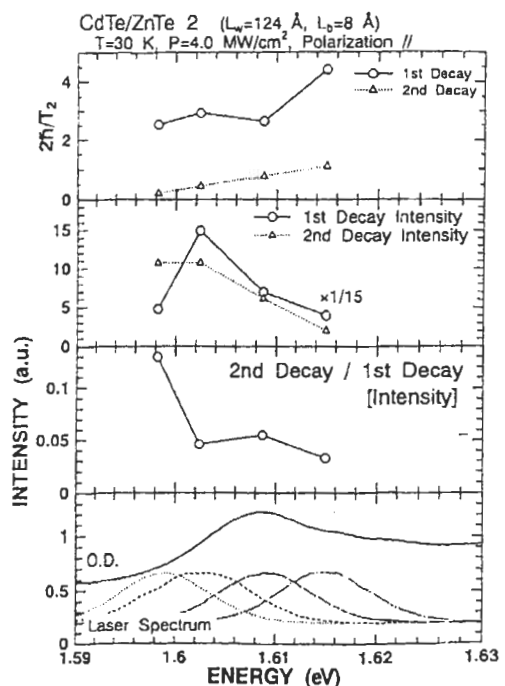

Fig. 4 Resonant behavior of DFWM signals at the QW exciton energy.

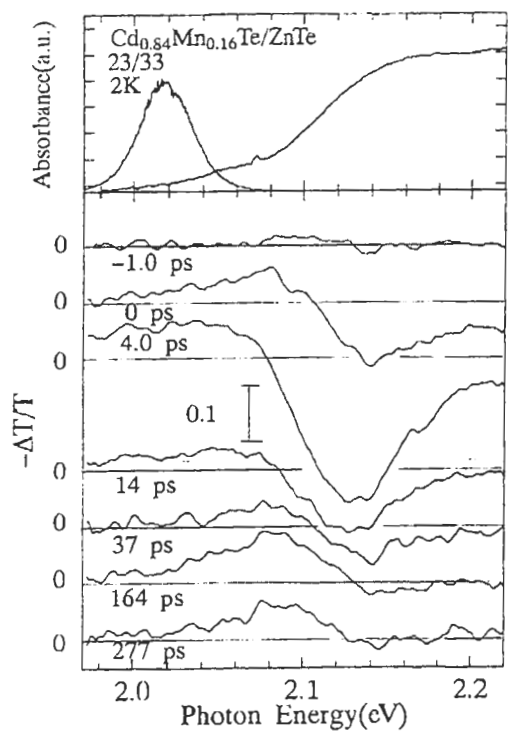

Fig. 5 Transient absorption spectra in the $Cd_{0.84}Mn_{0.16}Te/ZnTe$ MQWs. The top spectra are the absorption and luminescence spectra of the QW exciton in steady state.

meV to the lower energy side. The large Stokes shift of the exciton luminescence shows the existence of the fluctuation of the well thickness in the MQWs. The lower part of Fig. 5 is the transient absorption spectra, which show the increase of the absorption-saturation intensity at 2.23 eV until 4 ps after the pump excitation and the decrease of the saturation intensity with the time constant of 20 ps. The fast decay of the exciton is due to the energy transfer of the exciton energy to the Mn d-electrons, where the exciton energy in this QW is higher than the Mn d-d transition energy. An additional induced absorption arises at 2.08 eV, which can be seen in 37-277 ps region. The difference of the energy of the induced absorption to the exciton peak energy is 40 meV. The origin of the induced absorption peak is considered as the excitonic magnetic polaron absorption in which the Mn ion spins are polarized by the formerly created and annihilated excitons.

The result of PPAS in the CdTe/ZnTe ADQWs (L_{NW}=10 Å, L_{WW}=20 Å) is shown in Fig. 6. In case of L_b=300 Å, the saturation of absorption in NW and WW decays with same time constant of 160 ps, where the tunneling time of NW exciton is the order of 10^6 ps. In the ADQW of L_b=75 Å, the saturation of absorption of NW decays faster than that of WW, which is interpreted by the fast tunneling

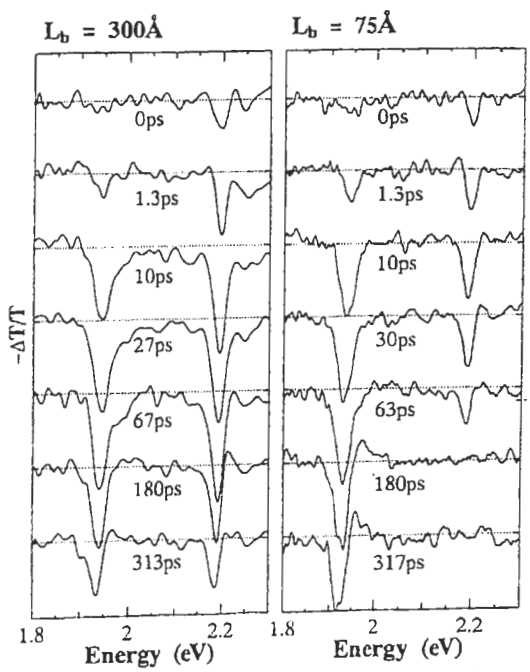

Fig. 6 Transient absorption spectra in the CdTe/ZnTe ADQWs.

time (1 ps) of the electrons from NW to WW. Numerical analysis of the PPAS spectra shows the individual tunneling process of electrons and holes through the barrier in the ADQW of L_b < 38 Å.

The exciton luminescence in NW and WW of the $Cd_{1-X}Mn_XTe$/ZnTe ADQWs also shows the tunneling effect of the carriers from NW to WW. An external magnetic field of 7 T induces the faster tunneling rate of the carriers than that at zero magnetic field. This fast tunneling induced by the magnetic field is closely correlated with the exciton state in the magnetic QW: At zero magnetic field the exciton is coupled with the Mn ions by the exchange interaction and the magnetic polaron state of the exciton is formed as a bound state in the QW. Therefore the exciton is bound in the QWs with the magnetic polaron binding energy. On the other hand, in the high magnetic field, the magnetic polaron effect decreases due to the external alignment of the Mn ion spins by the magnetic field. In this case the exciton can tunnel easily from NW to WW owing to the decrease of the magnetic polaron effect. The tunneling process is analyzed by using the semiclassical carrier tunneling model.

In conclusion the present ultrafast spectroscopic study clarified the energy and phase relaxations of the excitons as well as the carrier tunneling in the DMS nanostructures, where both the confinement effect and the exchange interaction play significant roles.

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