Excitonic effects in diluted magnetic semiconductor nanostructures

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Excitonic properties and the dynamics are reported in quantum dots (QDs) and quantum wells (QW) of diluted magnetic semiconductors. Transient spectroscopies of photoluminescence and nonlinear-optical absorption and emission have been made on these quantum nanostructures. The $Cd_{1-x}Mn_xSe$ QDs show the excitonic magnetic polaron effect with an increased binding energy. The quantum wells of the $Cd_{1-x}Mn_xTe/ZnTe$ system display fast energy and dephasing relaxations of the free and localized excitons as well as the tunneling process of carriers and excitons in the QWs depending on the barrier widths. The observed dynamics and the enhanced excitonic effects are the inherent properties of the diluted magnetic nanostructures.

1. Introduction

Nanometer-scale confinements of the band electrons in semiconducting materials provide varieties of quantum phenomena such as low dimensional electronic states, increased exciton binding energies and dynamics of carriers in the systems. Diluted magnetic semiconductors (DMSs), which involve magnetic ions in the cation sites, show marked magneto-optical properties due to the exchange interaction of the band electrons (holes) with the magnetic ions. Therefore, nanosturcture DMSs are expected to show particular magneto-optical effects owing to the confinement of both the electronic and magnetic states. In this paper, we report properties of the quantum dots (QDs) of $Cd_{1-x}Mn_xSe$ and the quantum wells (QWs) of the $Cd_{1-x}Mn_xTe$ and ZnTe layers and show the dynamics of low-dimensional excitons in the confined magnetic nanostructures [1–7].

2. Experimental

Fabrication of $Cd_{1-x}Mn_xSe$ QDs in the SiO_2 matrices was made by an rf-sputtering and subsequent heat treatments. Diameter of the QDs are 20–400 Å and the Mn concentration x is 0.05–0.2. Multiquantum wells (MQWs) and asymmetric quantum wells (ADQWs) of $Cd_{1-x}Mn_xTe/ZnTe$ ($x = 0$–0.2) were prepared by the hot-wall epitaxy method. Excitonic properties and the dynamics have been studied by the transient spectroscopies of photoluminescence, degenerate four wave mixing (DFWM) and pump-probe differential absorption, where the optical excitations were made by a mode-locked Ti-sapphire laser and amplification system.

3. Excitonic effects in DMS quantum dots

Microcrystals of $Cd_{1-x}Mn_xSe$ grown in SiO_2 amorphous matrices show the lattice image as displayed in Fig. 1, where the microcrystal of 150 Å diameter $Cd_{1-x}Mn_xSe$ in the SiO_2 matrix appears with the lattice fringe of the wurtzite sturcture [1,3]. The crystal structure is the same as that of the bulk crystal and the lattice constant is 4.3 Å. The absorption spectra of the $Cd_{1-x}Mn_xSe$ microcrystals show humps of the band-edge absorption in the 2.0–2.5 eV region, which are the evidence of the exciton absorption in the $Cd_{1-x}Mn_xSe$ microcrystals. The averaged diameter D of the microcrystals in the samples in determined from the x-ray diffraction.

The microcrystal-size dependence of the exciton energy measured from the absorption edge agrees with the calculation of the exciton confinement energy, which indicates that these $Cd_{1-x}Mn_xSe$ microcrystals can be described by the QDs for the excitonic state. Transient characteristics of the luminescence in $Cd_{1-x}Mn_xSe$ QDs (the average diameter is 220 Å) is shown in Fig. 2, where the exciton luminescence at 0 T decays with the decay 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 decay is much slower. At 5 T the exciton luminescence decays with a faster decay time constant [4]. The lifetime of the QD excitons deduced from the luminescence decay is distributed in the range of 100 ps–1 ns. The variation is caused by the difference in the

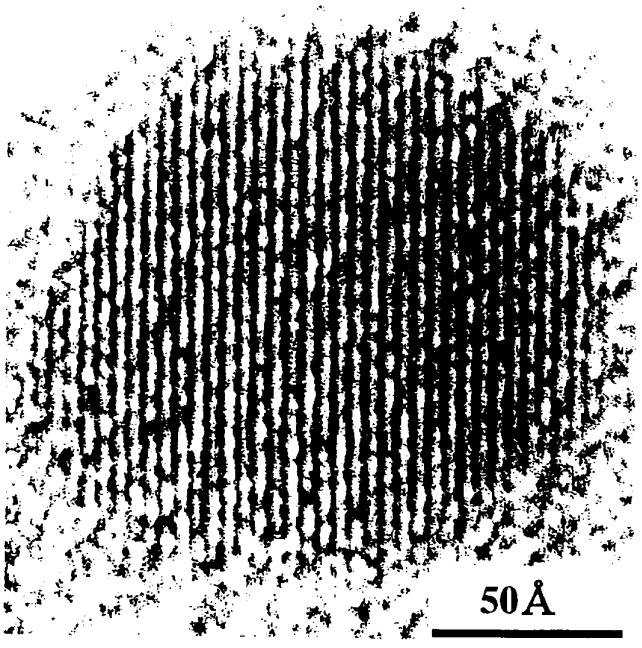

Figure 1. Lattice image of the $Cd_{1-x}Mn_xSe$ ($x = 0.15$) QD grown in the SiO_2 amorphous matrix.

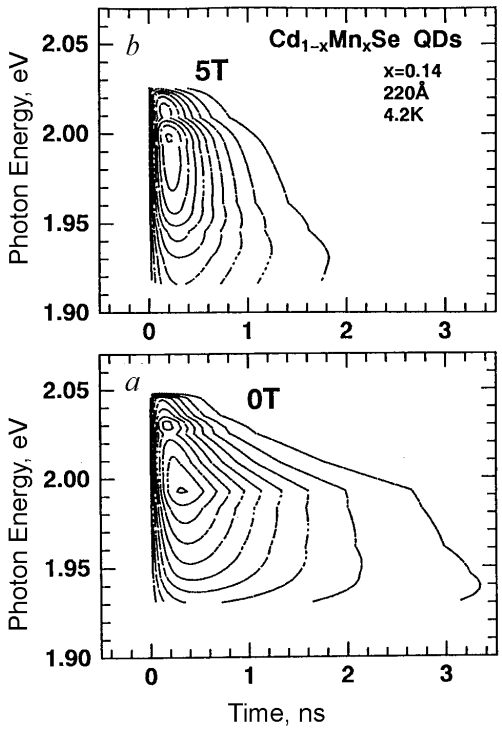

Figure 2. Transient exciton luminescence in the $Cd_{1-x}Mn_xSe$ QDs at 0 and 5 T.

exciton lifetime in the size-distributed QDs. In magnetic field of 5 T, the exciton lifetime decreases markedly due to the increase of the radiative recombination rate. The decrease of the exciton lifetime by the magnetic field also indicated the evidence of the exchange interaction of the exciton with the Mn ions in the QDs, since this decrease is caused by the level crossing of the singlet and triplet exciton states in magnetic field.

The exction average-energy in the QDs, which is given by the gravity center of the time-resolved luminescence spectra, decreases by 40 meV at 0 T for the increase of time with the time constant of 900 ps. At 5 T, the increased radiative recombination rate induces a fast decay of the exciton luminescence. The observed variation of the exciton luminescence displays the formation process and the radiative decay of the excitonic magnetic polaron in the QDs.

The magneto-luminescence of the $Cd_{1-x}Mn_xSe$ QDs for the selective energy excitation in the exciton absorption region shows decrease of the Stokes shift energy ΔE of the luminescence wtih increasing the magnetic lield. ΔE varies from 80 meV to 53 meV with increasing H from 0 T to 7 T for a fixed exctiation energy of $E_{exc} = 1.905$ eV. The Stokes shift ΔE in the selective excitation is related to the magnetic polaron formation energy. The formation energy of the excitonic magnetic polaron is suppressed by the presence of strong magnetic fields, since the external magnetic field aligns the Mn spins along the field direction. Therefore the reduction of the Stokes shift due to the magnetic field corresponds to the binding energy of the excitonic magnetic polaron. The existence of the excitonic magnetic polaron in the DMS QD of 160 Å diameter is confirmed in the present work with the magnetic polaron binding energy of 27 meV. Recently the magnetic polaron binding energy of the exciton state in DMS QDs has been calculated [8]. The fairly good agreement between the experimental results of $Cd_{1-x}Mn_xSe$ QDs with the calculation indicates marked increase of the exchange interaction in the QDs due to the confinement effect.

4. Excitonic effects in DMS quantum wells

The DMS QWs are studied by the transient nonlinear optical spectroscopies [6,7]. The DFWM measurements have been made on the $Cd_{1-x}Mn_xTe/ZnTe$ QWs. In the $Cd_{1-x}Mn_xTe$ MQWs (the well width $L_x = 124$ Å, the barrier width $L_b = 8$ Å, the periods $P = 35$) at the resonance excitation on the exciton state, a double-exponential-decay signal is observed, where the fast decay component corresponds to the dephasing time $T_2 = 450$ fs. This fast dephasing is caused by 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. 3. The slow decay component can be attributed to the dephasing process of the localized excitons, where the slow

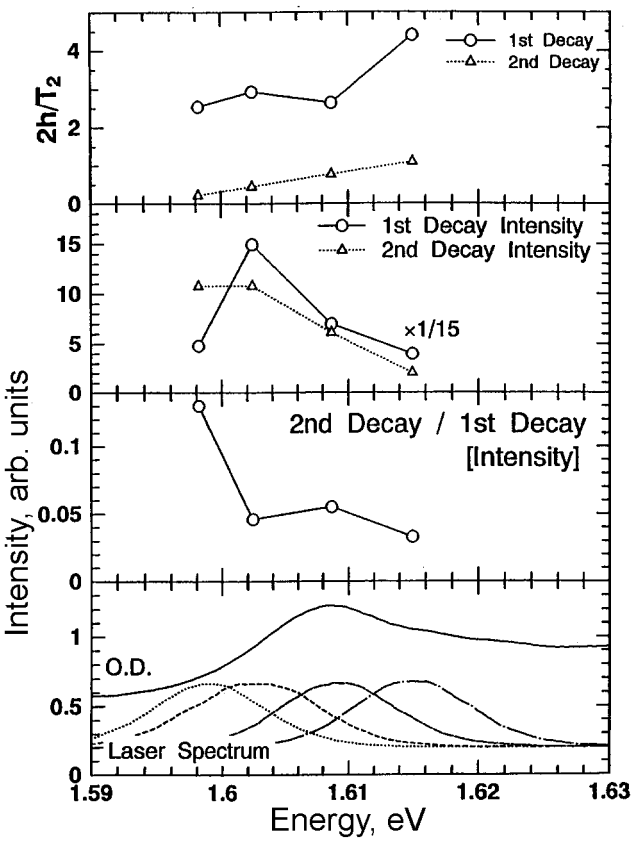

Figure 3. DFWM signal intensities in the CdTe/ZnTe MQWs, which show resonant behavior of DFWM signals at the QW exciton energy.

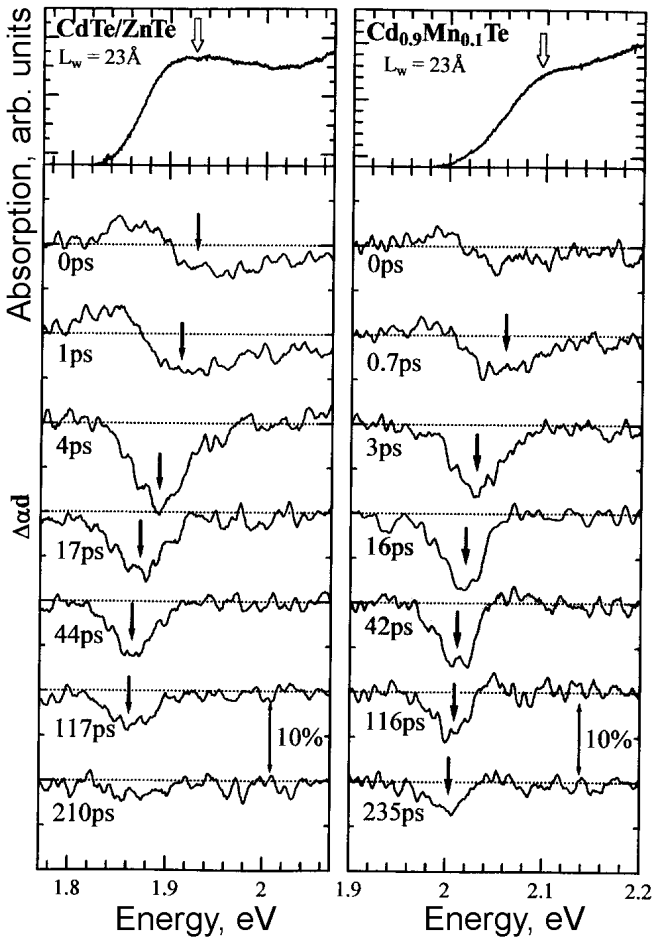

Figure 4. Pump-probe differential absorption spectra in the $Cd_{1-x}Mn_xTe/ZnTe$ MQWs ($x = 0$ and 0.1).

decay of 6.4 ps partly involves the longitudinal relaxation rate of the free exciton to the localized state in the QW. The observed short T_2 of the QW exciton is independent of the lattice temperature, indicating a less affection of acoustic and optical phonon relaxations. The fast dephasing of the exciton in the $Cd_{1-x}Mn_xTe/ZnTe$ QW system is caused by the relaxation due to the lattice inhomogeneity involved in the strained QWs because the free exciton is much affected by the inhomogeneous relaxation of the lattice. Present experimental results display the dynamics and the dephasing process of the excitons in the strained quantum wells.

The pump-probe differential absorption spectra of the $Cd_{1-x}Mn_xTe/ZnTe$ MQWs ($x = 0$ and 0.1) with L_w=23 Å are displayed in Fig. 4. The exciton absorption in these MQWs locates at 1.92 and 2.10 eV. In the $Cd_{0.9}Mn_{0.1}Te/ZnTe$ MQWs the absorption saturation peak shows a fast low-energy shift of 70 meV within 10 ps and also shows a subsequent gradual low-energy shift of 20 meV until 235 ps after the pump excitaion. On the other hand, the CdTe/ZnTe MQWs exhibits the gradual low energy shift of 60 meV until 210 ps after the pumping. The dominant fast-energy-relaxation of the exciton in the $Cd_{0.9}Mn_{0.1}Te/ZnTe$ MQWs indicates the formation process of the excitonic magnetic polarons in the two dimensional QWs, while the gradual energy relaxations seen in both of the MQWs are due to the localication process of the excitons in the inhomogeneous MQWs. Exction luminescence peaks are shifted by 60–100 meV to the lower energy side from the exciton absorption peak in the steady state, where the large Stokes shift of the exciton luminescence indicates the existence of the fluctuation of the well thickness in the MQWs.

The result of the pump-probe differential absorption in the $Cd_{1-x}Mn_xTe/ZnTe$ ADQWs (the narrow well width $L_{NW} = 10$ Å, the wide well width $L_{WW} = 20$ Å) shows, in the case of $L_b = 300$ Å, that the saturation of absorption in NW and WW decays with a same time constant of 160 ps, where the tunneling time on 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 time (1 ps) of the electrons from NW to WW. Numerical analysis of the pump-probe absorption spectra shows the tunneling process of electrons and holes through the barrier in the ADQW of $L_b > 38$ Å.

In summary, ultrafast spectorscopies on the QDs and QWs of MDSs in magnetic fields have clarified the energy and phase relaxations and the tunneling processes of the excitons and carriers. The magneto-optical properties of the DMS nanosturctures are significantly affected by both the confinement effect and the exchange interaction.

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