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Excitation spectrum of infrared photoluminescence in CdMnTe

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In semimagnetic semiconductor $Cd_{1-x}Mn_xTe$, we investigate excitation spectra of infrared photoluminescence (~1.2eV) which is related to internal d-d transitions of Mn^{2+} for various Mn concentrations. In the low concentration region (x<0.4) where band gap is smaller than the internal d-d transition, the peak position of excitation spectra shifts linearly to x. And in the high concentration region (x>0.4) where band gap exceeds Mn absorption energy, the peak position no longer shifts and stays near Mn absorption energy. We discuss the direct energy transfer from extended states to localized d-states with lattice distortion.

1. INTRODUCTION

The ternary compound $\operatorname{Cd}_{1-x}\operatorname{Mn}_x\operatorname{Te}$ is semimagnetic semiconductor. And its crystal structure is zincblende type up to $x\sim0.7$. This material has two properties as semiconducting and magnetic material. Interesting optical and transport properties such as giant Faraday rotation [1], large Zeeman effect [2], magnetic polaron [3] etc. are observed. These effects are interpreted as a result of the exchange interaction between the localized 3d electrons of Mn and Bloch electrons in conduction and valence bands. And the band gap energy increases linearly to Mn concentration x as in the usual mixed semiconductor.

The optical properties of $Cd_{1-x}Mn_x$ Te have been investigated for many years [4]-[9]. In this material there are three kinds of intrinsic photoluminescence(PL) named as exciton PL(EXPL), manganese PL (MnPL), and infrared PL(IRPL). It is considered that MnPL and IRPL relate to internal d-d transitions of $Mn^{2+}[5, 8]$. For these PL, it is predicted that the lattice distortion is important. However there are few reports on lattice distortion in their excited states of MnPL and IRPL. About MnPL and IRPL the following results are proposed: (i) the differences in position of minima of ground states and excited states in configuration coordinate model are large for both MnPL and IRPL, (ii) the lattice distortion of the excited states of IRPL is larger than that of MnPL

under the assumption that only Mn^{2+} moves and the meta-stable position of excited Mn^{2+} as the excited states of IRPL is similar to DX center and/or EL2 in AlGaAs and GaAs. From the analyses, the thermal barrier from the lowest relaxed excited states of MnPL to the excited states of IRPL is estimated to be 36meV[8].

In this paper we will present the excitation spectra of IRPL(IRPLE) for various Mn concentrations of $Cd_{1-x}Mn_x$ Te, and discuss the energy transfer between localized states and extended states.

2. EXPERIMENTS

We observed three kinds of PL and IRPLE spectra at 140K in single crystals of $Cd_{1-x}Mn_xTe$ grown by a Bridgman technique. We obtain PL spectra using Ar⁺ ion laser of 2.54eV and IRPLE spectra using a tunable dye laser pumped by a Nd:YAG laser. Lights emitted from samples are detected by a photomultiplier tube. And its outputs are analyzed by phase sensitive detector and by digital oscilloscope, respectively.

3. RESULTS AND DISCUSSIONS

Typical IRPLE spectra in $Cd_{1-x}Mn_x$ Te are shown in Figs.1 and 2 by closed circles for vari-



Fig. 1: Normalized excitation spectra of IRPL (closed circles) and PL spectra (solid lines) in $Cd_{1-x}Mn_x$ Te at 140K when the band gap energy is smaller than Mn^{2+} d-d absorption energy. Figure(i) x=0.3, and (ii) x=0.4. Arrows (a) indicate the peak energy of band gap absorption energy for corresponding Mn concentration x, which are determined by reflectivity measurement[4]. And arrows (b) indicate that of estimated Mn^{2+} d-d transition absorption energy. Photoluminescence near the arrows (a) is assighed as EXPL.

ous x at 140K. And solid lines show PL spectra of $Cd_{1-x}Mn_xTe$. Arrows (a) indicate the position of exciton absorption peak energy obtained by reflectance measurements and arrows (b) indicate that of Mn^{2+} d-d absorption energy[4].

In Figs. 1(i) and 1(ii) the spectra of PL and IR-PLE are shown for x=0.3 and 0.4, respectively. The band gap energy of x=0.3 is smaller than Mn²⁺ d-d transition absorption energy estimated by extrapolating its concentration dependence in the region of x > 0.4. The peak of PL spectra shown by solid lines near band absorption energy is EXPL, and MnPL is not observed in this concentration. The peak energy of IRPLE is lower than the band gap energy by \sim 30 meV. In the case of x=0.4 the energy for band absorption and dd absorption at 140K are same. The features of EXPL and IRPLE spectra in Figs.1(i) and 1(ii) are similar and the peaks are near the corresponding band gap energy. This result indicates that the effective excitation is band gap excitation.



Fig. 2 : Normalized excitation spectra of IRPL (closed circles) and PL spectra(solid lines) in $Cd_{1-x}Mn_xTe$ at 140K when the band gap energy is larger than Mn^{2+} d-d absorption energy. Figure(i) x=0.46, and (ii) x=0.6. Arrows (a) indicate the peak energy of band gap absorption energy. And arrows (b) indicate that of $Mn^{2+}d$ -d transition absorption energy. Photoluminescence at 2.0eV is assigned as MnPL (d-d transition from ${}^{4}T_{1}$ to ${}^{6}A_{1}$) and PL at 2.2eV in figure (i) is assigned as EXPL.

In Figs.2(i) and 2(ii), IRPLE and PL spectra are shown for x=0.46 and for x=0.6, respectively. In this case d-d transition absorption energy is smaller than exciton absorption energy. Photoluminescence at 2.0eV is assigned as MnPL, the internal transition from ${}^{4}T_{1}$ to ${}^{6}A_{1}$. In Fig.2(i) the sharp PL at 2.2eV is assigned as EXPL. For x=0.46 the peak of IRPLE is not observed near band gap absorption energy, but at 2.15eV of Mn^{2+} d-d transition absorption peak. In Fig.2(ii) for the case of x=0.6 the peak of IRPLE is observed at 2.15eV as in the case of x=0.46. In this case EXPL is not shown, but observed at 2.4eV. In Fig.2 there is no peak near the band gap energy and the threshold of IRPLE is at the lower energy than the band gap energy. This result indicate that the band gap excitation is not effective to IRPL, but the internal transition is effective.

In Fig.3 the various peak positions are shown as a function of Mn concentration x. Closed circles show the peak energy of IRPLE. And closed triangles, reversed triangles and diamonds show the peak energy of EXPL, MnPL and IRPL, respectively. Solid line (a) shows the exciton absorption peak energy and solid line (b) shows Mn^{2+} absorption peak energy, which are determined by reflectivity measurement[4]. Here EXPL shifts linearly to higher energy increasing x. Further MnPL and IRPL do not shift with x in comparison with the large shift of EXPL. Manganese PL is not observed in the region of x < 0.4, but IRPL is observed even in small x as x=0.2. At 140K full widths at half maximum are \sim 30meV for EXPL, about \sim 160meV for MnPL, and about \sim 140meV for IRPL. The line shapes of IRPL spectra are almost same for the samples of all x we measured.

As shown in Fig.3 the concentration dependence of the peak energy of IRPLE has change around x=0.4. When the band gap is smaller than Mn^{2+} d-d absorption energy, the peak of IRPLE shifts with band gap. When the band gap energy crosses over Mn^{2+} d-d absorption energy, peak energy of IRPLE becomes constant at 2.15eV.

In $Cd_{1-x}Mn_x$ Te, roughly, there are two types of energy states. One is band states like conduction band and valence band. The other is localized states such as 3d multiplet states. In the latter case, excitation and relaxation process are discussed using configuration coordinate model. In high Mn concentration region, the proposed configuration coordinate model for explaining the temperature dependence of MnPL and IRPL[8] and time profile of the intensity of IRPL[9] is applicable. However in low Mn concentration region, there is no relaxation process from the excited states of MnPL to the excited states of IRPL. Because there is no peak at 2.15eV in IRPLE for x < 0.4, but at the band gap energy. The character of effective excitation of IRPL changes from band gap excitation to internal d-d transitions. Thus the energy transfer from band gap excitation to internal d-d transitions is realized.

3. CONCLUSIONS

We measure the excitation spectra of infrared photoluminescence for various Mn concentration in semimagnetic semiconductor $Cd_{1-x}Mn_x$ Te. We find the peak of the excitation spectra of IRPL shifts with Mn concentration for x < 0.4, and is



Fig.3 : The peak positions at 140K are shown as a function of Mn concentration x. Closed circles show the peak energy of IRPLE. And closed triangles, reversed triangles and diamonds show the peak energy of EXPL, MnPL and IRPL, respectively. Solid line (a) shows the band gap absorption energy, solid line (b) shows Mn^{2+} absorption peak energy, which are measured by photo reflectivity[4], and its extrapolation is shown by dots.

constant at d-d absorption energy 2.15eV for x>0.4. From these experimental results we suggest the energy transfer from band gap excitation to internal d-d transitions.

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