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High-density optical spin injection in self-assembled semiconductor quantum dots

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ABSTRACT

High-density optical spin injection and the dynamics from a diluted magnetic semiconductor (DMS) of $Zn_{0.95}Mn_{0.05}Se$ as a spin injector into self-assembled quantum dots of CdSe have been studied by means of time- and spin-resolved photoluminescence. Spin relaxation during the injection becomes significant with increasing the density of spin-polarized exciton in the DMS. This spin relaxation can be caused by the diffusive transfer of spin-polarized electrons through the barrier, when the energy of spin-polarized electron generated in the DMS exceeds the barrier energy due to a filling effect for localized-exciton states in the DMS.

1. Introduction

Spin injection in semiconductor quantum dots (QDs) has been extensively studied in several systems of self-assembled QDs coupled with spin generators of diluted magnetic semiconductors (DMS) or metallic ferromagnetic films [1–4]. We have studied the spin-injection dynamics in the former system using DMS by means of circularly polarized, thus spin-resolved, transient photoluminescence (PL). Self-assembled CdSe QDs with high optical qualities were epitaxially grown on the DMS layer of ZnMnSe with a barrier layer of ZnSe. This epitaxial system allowed us to observe the spin-injection dynamics of spin-polarized excitons in the picosecond time domain [5]. Spin relaxation during the injection was also elucidated [6]. In this paper, we report on the excitation-spin-density dependence of the spin-injection dynamics for the purpose of establishing high-density optical spin injection into QDs. The high-density spin injection into QDs will be strongly motivated for future developments of spin-functional optical devices based on semiconductor QDs. The spin density was controlled by changing the power of light pulse used for selective excitation for a spin-split exciton level in the DMS.

2. Experimental procedure

A self-assembled QD layer was epitaxially grown on a DMS layer of $Zn_{0.95}Mn_{0.05}Se$ with a 2 nm-thick barrier layer of ZnSe by molecular beam epitaxy. Time-resolved PL spectra discriminated by circular polarization were measured by using second-harmonic excitation pulses of a mode-locked Ti:sapphire laser and a streak camera. Selective excitation with linearly polarized pulses was employed for a lower-energy spin-split exciton level in the DMS under magnetic fields [7]. This selective excitation with the linearly polarized light prevented optical spin orientation in the QDs. The excitation power (P) was varied from 0.1 to 20 mW. The sample was set at 2 K and magnetic fields up to 5 T in the Faraday geometry.

3. Results and discussion

Fig. 1(a) shows a schematic illustration of exciton-spin injection from the DMS layer of $Zn_{0.95}Mn_{0.05}Se$ into the self-assembled QD of CdSe. σ^+-polarized PL in the QD means exciton-spin injection from the DMS layer since spin-polarized excitons generated in the DMS are σ^+-active, while excitonic PL in the CdSe QDs shows σ^--polarization under magnetic fields. Circular polarization degree (ρ) of exciton PL in the CdSe QDs with the detection energy of 2.60–2.61 eV at 5 T and 2 K is shown in (b) as a function of excitation power, where an example of circularly polarized PL spectra in the QDs are shown as an inset. A positive value of ρ indicates spin injection from the DMS layer to the QDs, as described above. The ρ value decreases significantly with increasing P at $P \geq 2$ mW. We ruled out a filling effect for the QDs when high-power excitations up to 20 mW were performed, because a mobility edge of this QD ensemble ($\cong 2.59$ eV) was not affected by such high-power excitations [8].

We have observed time-resolved circularly polarized PL in the QDs, reflecting the spin-injection dynamics into the QDs. Fig. 2 (a) shows a model of rate equation analysis. Spin-polarized excitons are injected from the lower-energy spin-split exciton level of the DMS into the exciton levels of the QD. In this injection process, we introduce a spin-conservation factor η [6]. A value

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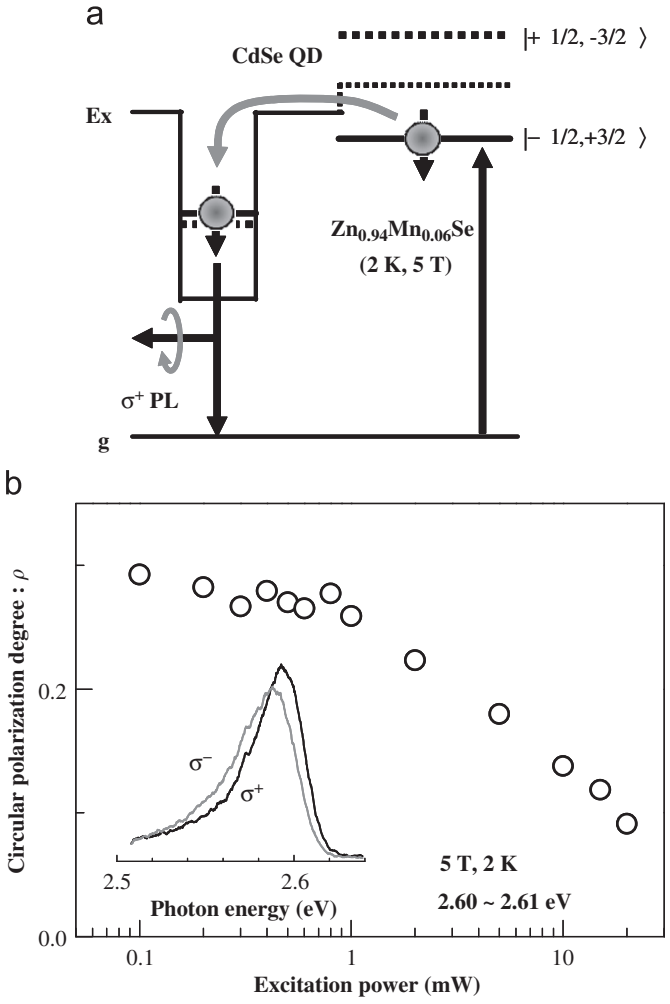

Fig. 1. A schematic illustration of the exciton-spin injection process from a DMS layer of $Zn_{0.95}Mn_{0.05}Se$ into a self-assembled QD of CdSe (a). Magnetic quantum numbers of the spin-split exciton states are indicated in the DMS under magnetic fields. Spin-polarized excitons are selectively excited by linearly polarized light pulses (excitonic-spin generation). Circular polarization degree ρ of exciton PL in the CdSe QDs with the photon energy of 2.60–2.61 eV at 5 T and 2 K, as a function of excitation power (b). The ρ was defined as $\rho \equiv (I_{\sigma+} - I_{\sigma-})/(I_{\sigma+} + I_{\sigma-})$, where I was a circularly polarized PL intensity. The inset shows an example of the circularly polarized PL spectra in the QDs.

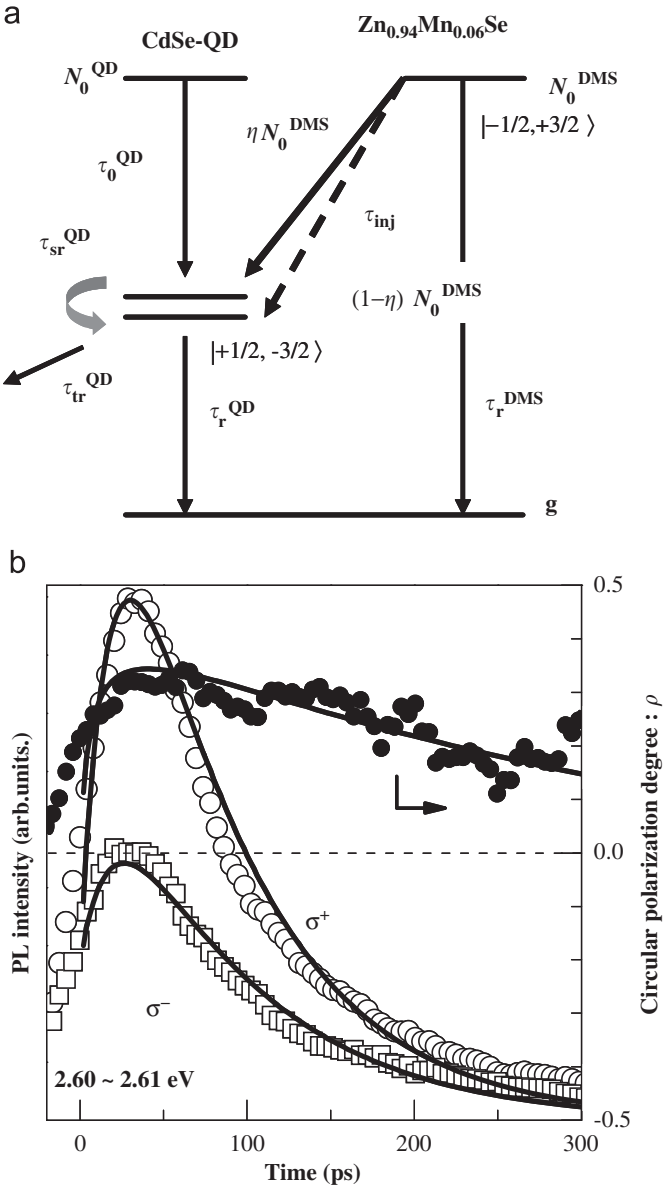

Fig. 2. A model of rate equation analysis (a). Spin-polarized excitons are injected from $Zn_{0.95}Mn_{0.05}Se$ to the QD, where a spin-conservation factor η is introduced. Transient circularly polarized PL (open circles; σ^+ and squares; σ^-) and the circularly polarization degree (closed circles) at 5 T and 2 K in the QDs (b). Solid lines are rate-equation calculations.

of η can change from 0.5 to 1, corresponding to complete spin-relaxation and spin-conservation, respectively. Transient circularly polarized PL and the corresponding circular polarization degree as a function of time in the QDs are shown in Fig. 2 (b). A steep increase in the σ^+-polarized PL intensity immediately after the pulsed excitation indicates the spin-injection dynamics. Then, the circular polarization degree decreases gradually due to exciton-spin relaxation inside the QDs, where the σ^--polarized exciton state is energetically stable. We calculated the circularly polarized PL time profiles using the rate equations described above. From the fitting, we obtained parameters responsible for the spin-injection dynamics as a function of excitation power P. Fig. 3 shows spin-conservation factor η as a function of P. A value of η decreases as the excitation power increases, reflecting spin relaxation during the spin injection. This tendency coincides well with the decrease in the ρ value as shown in Fig. 1(b). This P-dependent decrease in the η value indicates spin relaxation, i.e., spin loss during the exciton-spin injection.

To explore the origin of this spin loss at high excitation powers, we examined the power-dependence of PL spectrum in the DMS. The DMS-PL spectra show a typical filling effect for localized-exciton states in the DMS (an inset in Fig. 4(b)). Potential fluctuations due to compositional fluctuations were essential in DMS materials because of the existence of magnetic ions. This filling effect induces diffusive transfer of spin-polarized electrons through the barrier at high excitation powers, which is schematically illustrated in Fig. 4 (a). We calculated averaged energy differences as a function of P between carriers (an electron and heavy hole (hh)) in the DMS and those in the barrier of ZnSe from the peak energy of the PL spectrum in the DMS. Percentage of diffusive electrons (the number ratio of the diffusive electrons to totally generated ones) in the DMS was then calculated as a function of P, as shown in Fig. 4 (b). The difference of exciton binding energy between the DMS and ZnSe was not taken into consideration because the chemical composition of Mn was only 5% and the resultant energy band gap and band offset were almost identical at 0 T. Energies of giant Zeeman shifts are different between the electron and hh, when we apply magnetic