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Exciton Spin Stability in InP Quantum Dots at the Elevated Temperatures

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1. Introduction

The lowest-energy exciton in III-V quantum dots (QDs) is composed of an electron with $s_{ez} = \pm 1/2$ and a heavy hole with $J_{hz} = \pm 3/2$. Combination of 2x2 spin states gives quartet excitons simply denoted by projection of the total angular momentum of the exciton $J=s_e+J_h$ upon the quantization axis, $|m>$, (m=+1,-1,+2,-2), respectively. Bright excitons, |-1>, |+1>, can be directly excited by light and can luminesce. They decay at the radiative decay rate plus the nonradiative decay rate. Dark excitons, |+2>, |-2>, cannot be directly excited by light nor can luminesce. Dark excitons decay at the nonradiative decay rate and, therefore, usually have long lifetime.

Spin flip of either an electron or a hole is needed for the conversion between bright excitons and dark excitons. If bright and dark excitons are equally formed and if the spin flip rate is not much faster than the lifetime of exciton, dark excitons are superior in existing probability to bright excitons. Then dark excitons can work as the carrier and spin reservoir, because of their long lifetime.

Spin lifetime of excitons in QDs becomes much longer than the exciton lifetime at low temperatures [1]. Stable exciton spins in QDs are candidates for quantum information storage. Spin stability of excitons is thought to be reduced at the elevated temperatures, but the stability at the elevated temperatures is not understood. Spin stability is well reflected by the resonant spin orientation of excitons in QDs we reported very recently [2], because the resonant spin orientation of excitons arises from inefficient conversion between bright excitons and dark excitons. The resonant spin orientation of excitons becomes the measure of inefficient spin flip of either an electron or a hole converting between bright excitons and dark excitons. In this work, spin stability of excitons in InP QDs is investigated at the elevated temperatures by monitoring resonant spin orientation of excitons.

2. Experimental Procedures

The sample studied was neutral InP self-assembled QDs (areal density $\approx 10^{10}$ cm^{-2}) grown on an n$^+$-GaAs substrate. A single layer of InP QDs 40 nm in average diameter and 5 nm in height was grown between the 100 nm $In_{0.5}Ga_{0.5}P$ barrier layers. Under the above-barrier excitation, the sample shows the photoluminescence (PL) band peaked at 1.73 eV, as shown in Fig.1(a).

A HeNe laser was used for the linearly polarized above-barrier excitation of InP QDs at 1.959 eV. The sample was set in a superconducting magneto-optic cryostat. The magnetic field was parallel to the optical excitation axis. The PL polarization was measured at 1.722 eV in the reflection geometry with a photoelastic modulator and a Glan-Thompson linear polarizer as a circular polarization analyzer, and was detected by a GaAs photomultiplier together with a double monochromator and a 2-channnel gated photon counter. The degree of circular polarization, P_c, is defined by, $P_c = (I^+ - I^-)/(I^+ + I^-)$, where I^+ and I^- are the intensities of the right (σ^+) and left (σ^-) circularly polarized PL under linearly polarized pumping, respectively.

3. Exciton Fine Structure and Resonant Spin Orientation

In the longitudinal magnetic field (Faraday configuration), bright excitons and dark excitons are described by $(1/2)[\delta_0 \pm\{\delta_1^2+(g_{h,z}-g_{e,z})^2\mu_B^2B^2\}^{1/2}]$, and $(1/2)[-\delta_0 \pm\{\delta_1^2+(g_{h,z}+g_{e,z})^2\mu_B^2B^2\}^{1/2}]$, respectively, where elec-

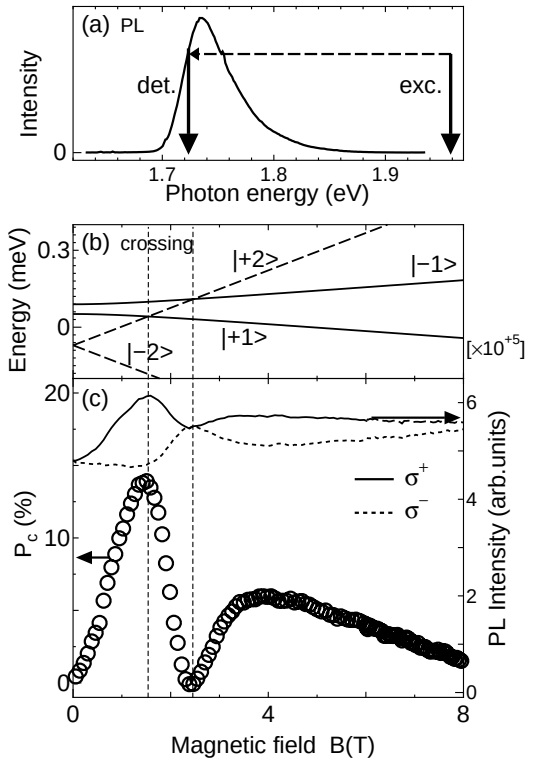

Fig. 1 (a) Photoluminescence (PL) spectrum of the sample under HeNe laser excitation at 1.959 eV. (b) Calculated exciton eigen energies for InP quantum dots (QDs) studied. (c) Circular polarization (○) of PL detected at 1.722 eV under the longitudinal magnetic field at 5 K. The σ^+ and σ^- polarized PL intensities are shown as a function of the longitudinal magnetic field.

tron-hole exchange interactions, δ_0=0.141 meV, δ_1=0.038 meV and the electron (hole) g-factor, $g_{e,z}$=1.5 ($g_{h,z}$=1.02) are known from our preceding studies [2]. Zeeman energy of dark excitons is larger than that of bright excitons and hence causes energy crossing. Calculated energies are plotted in Fig.1(b). A dark exciton |+2> crosses with bright excitons |-1> and |+1> at 1.5T and 2.5T, respectively. Small off-diagonal elements in the spin Hamiltonian change crossing into anticrossing. Resonant spin orientation of excitons is observed under linearly polarized above-barrier excitation around magnetic-field-induced two (anti)crossings of bright and dark excitons in InP QDs. Under the above-barrier excitation, the observed resonant spin orientation of excitons is enhanced compared with that observed under the quasi-resonant excitation [2]. The enhancement is explained by the increased generation of dark excitons as a result of spin relaxation. At the (anti) crossings, bright and dark excitons mix with each other and the wavefunction mixing induces the resonant spin orientation.

4. Exciton Spin Stability at the Elevated Temperatures

Temperature rise reduces the resonant spin orientation structures, as is shown in Fig. 2(a). Resonant spin orientation structures disappear at 65K. Resonant spin orientation structures depend on spin relaxation time, τ_s, and lifetime, τ_r, of excitons and decrease with the increase of $(1/\tau_s)/(1/\tau_r)=\tau_r/\tau_s$. They are roughly proportional to $\tau_s/(\tau_r+\tau_s)$. The lifetime, τ_r, of excitons is about 260ps and depends on temperature little. Simulation reproduces the reduction of the resonant spin orientation structures by changing the ratio, τ_s/τ_r as is shown in Figs. 2(b) and 2(c) [3]. Comparing the experimental resonant spin orientation structures and those simulated, we obtained the temperature-dependent spin relaxation time, as is shown in Fig. 2(d). Spin relaxation time becomes shorter with the increase of temperature. It is 6 times longer than the lifetime at 5K but is comparable with the lifetime at 40K. Temperature-dependent spin relaxation can be interpreted by the spin-phonon interactions consisting of the direct process and the two-phonon Orbach process [4]. In fact, the spin relaxation rate $1/\tau_s$ can be fitted by the sum of the direct process proportional to T and the two-phonon Orbach process proportional to $1/[\exp(\Delta E/kT)-1]$, where ΔE is the activation energy to the excited state.

4. Conclusions

Exciton spin stability in InP QDs at the elevated temperatures is studied by means of resonant spin orientation of excitons. With the increase of temperature, spin relaxation time becomes shorter and resonant spin orientation of excitons is reduced. It is 6 times longer than the lifetime of 260ps at 5K but is comparable with the lifetime at 40K.

Acknowledgements

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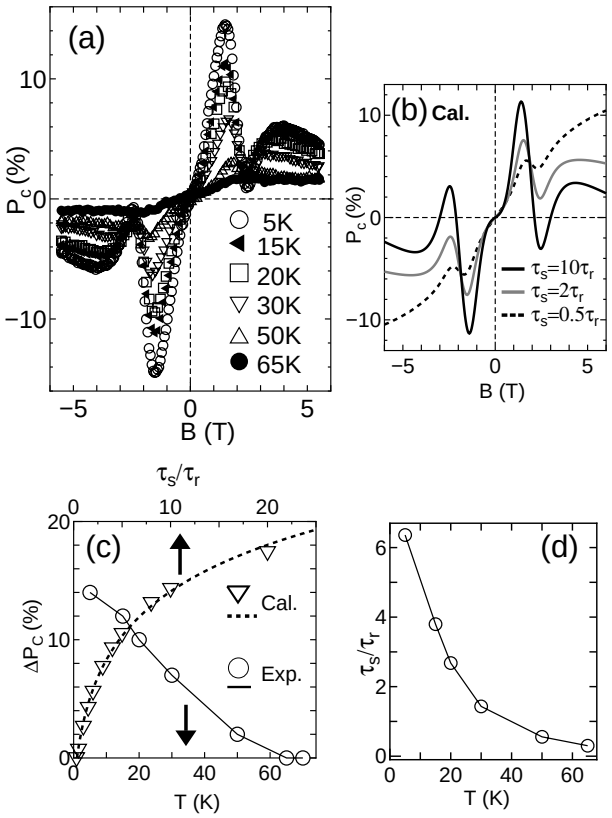

Fig. 2 (a) Temperature-dependent resonant spin orientation coming from the exciton level anticrossings. (b) Temperature dependent resonant circular polarization amplitude $\Delta P_c=P_c(1.5\text{T})-P_c(2.5\text{T})$. (c) Simulated circular polarization amplitude as a function of spin relaxation time, τ_s, over lifetime, τ_r. (d) Temperature-dependent τ_s/τ_r.

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