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Polarization conversion of excitonic photoluminescence under zero and nonzero magnetic fields in single InAlAs quantum dots

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Abstract

We demonstrated the circular to linear polarization conversion in single InAlAs/AlGaAs QDs. Since the anisotropic exchange interaction acts on the photo-created exciton spin as an in-plane effective magnetic field, the polarization of emissions can be converted from circular to linear and vice versa even under a zero external magnetic field. By using the quasi-resonant excitation, high conversion ratio up to ~50% was obtained under nonzero longitudinal magnetic field. Additionally, exciton spin relaxation time and built-in linear dichroism were estimated by exciton spin dynamics in the 3D pseudospin precession model.

Key words: quantum dot, optical alignment, optical orientation, InAlAs, fine structure

1. Introduction

Dynamics of localized spins in the semiconductor quantum dots (QDs) are attractive from both fundamental and practical points of view. For the application to quantum information processing [1], as well as QD lasers and single-photon sources, the polarization of the emitted photon associated with the exciton annihilation is one of the key quantities. For ideal QDs, the eigenstates are bright excitons formed by the circularly polarized photon with the angular momentum +1, and the polarization of the emission is detected as circularly polarized one (σ+) via the selection rule of optical transitions. However, actual QDs contain the anisotropy of the shape and strain distribution originated from the QD formation process, and as a result, the confinement potential symmetry is reduced from $D_{2d}$ to $C_2$ or lower. Since the anisotropic exchange interaction (AEI) caused by the symmetry reduction works as an in-plane magnetic field effectively, the precession of exciton spin with frequency $\Omega_{exc} = \delta_0 / \hbar$ can be observed even under a zero external magnetic field. Here, $\delta_0$ is the exciton fine structure splitting (FSS) and is of several tens $\mu$eV for typical InAlAs QDs. Consequently, AEI modifies the exciton eigenstates from $|\pm 1\rangle$ to $|+1\rangle \pm |-1\rangle$, and at the recombination lifetime, the polarizations are converted to linear ones ($x$ or $y$). In the sense that an exciton created with circularly polarized photon tends to emit the linearly polarized photon, this phenomenon can be interpreted as “polarization conversion”. A lot of works related to similar conversion in the QD ensembles have already been reported so far (for example, for InAlAs/AlGaAs QDs [2] and CdSe/ZnSe QDs [3]). Yet, it is difficult to deduce the intrinsic parameters from ensemble average over QDs containing the inhomogeneous shape and strain distribution.

In this work, we demonstrate the polarization conversion for single InAlAs/AlGaAs QDs. The quasi-resonant excitation brings the high conversion efficiency under zero and nonzero longitudinal magnetic fields. Additionally, considering the exciton spin dynamics based on a simple coherent precession model permits us to estimate the exciton spin relaxation time and built-in linear dichroism.

2. Experiments

We used self-assembled In$_{0.75}$Al$_{0.25}$As/Al$_{0.3}$Ga$_{0.7}$As QDs which were grown on a (100) GaAs substrate by molecular beam epitaxy [4]. Atomic force microscopy measurements on a reference, i.e. uncapped QD with the same growth conditions revealed that the average height and diameter of QDs are ~4 and ~20 nm with the areal QD density of ~5 x 10$^{10}$ cm$^{-2}$. To perform the single QD spectroscopy, small mesa structures with typical diameter ~150 nm were fabricated by wet etching and electron beam lithography.

A cw-Ti:Sapphire laser (~1.664 eV) was used to illuminate the QD sample. As we shall see later, this photon energy corresponds to the quasi-resonant excitation for a target InAlAs QD. The photoluminescence (PL) signals were collected by an objective lens, and were detected with the triple-grating spectrometer (f=0.64 m) and the liquid nitrogen cooled Si-charge coupled device array. Though the energy resolution of our setup is ~12 $\mu$eV, it can be improved to 5 $\mu$eV by the spectral fitting. The measurement temperature was kept to ~5 K, and a magnetic field up to 5 T was applied in Faraday geometry ($\mathbf{B} \parallel \mathbf{k}$). The excitation polarization was fixed to $\sigma_+$ or $\sigma_-$ by a quarter-wave plate and the polarization of PL spectra were analyzed with a half-wave plate (HWP) and a polarizer inserted in the detection path before the spectrometer.

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3. Results and Discussion

3.1. Without magnetic field

To study the circular to linear polarization conversion, efficient generation and injection of bright excitons into their ground states are required. For this purpose, the excitation energy was tuned to 1LO resonance of the exciton ground state. As well known for InAlAs/AlGaAs QDs [2], CdSe/ZnSe QDs [3], InAs/GaAs QD [5], LO phonon-assisted excitation causes the fast energy relaxation of photo-created excitons with preserving the degree of spin polarization, and correspondingly is feasible for effective injection of the exciton with high spin polarization.

Fig. 1 (a) shows the time-integrated PL detected at the PL energy of neutral exciton (X\(^0\)), neutral biexciton (XX\(^0\)) and positively charged exciton (X\(^+\)) varying the excitation energy (E\(_{ex}\)) under a zero magnetic field. In the case of the wetting layer excitation (E\(_{ex}\) ~1.70 eV), as shown by the gray line in the inset, not only X\(^0\) PL but XX\(^0\) and X\(^+\) can be observed simultaneously, and they decreased gradually with decrease of E\(_{ex}\). At E\(_{ex}\) =1.664 eV (~745 nm), only X\(^0\) indicates the abrupt increase (black line of the inset), which is identified as the LO phonon-assisted excitation. All of the experimental data shown hereafter were obtained with this excitation energy.

When the polarization of X\(^0\) emissions was analyzed with a rotating HWP and a polarizer, not only the PL energy (Fig. 1 (c) open circles) but the intensity (Fig. 1 (b)) show the periodical changes under a circularly quasi-resonant excitation. In those figures, the detection angle \(\phi\) was varied. The peak-to-peak amplitude of energy shift of ~50 \(\mu\)eV corresponds to the exciton fine structure splitting (\(\delta_0\)), and the intensity modulation suggests the observation of the circular to linear (C\(\rightarrow\)L) polarization conversion. In this paper, the degree of linear polarization (DLP) is used as a key index of the conversion efficiency, and defined as \(\rho = (I_\varphi - I_{\varphi + \pi/2})/(I_\varphi + I_{\varphi + \pi/2})\). Here, \(I_\varphi\) and \(I_{\varphi + \pi/2}\) is the PL intensity in the \((\varphi + \pi/2)\)-frame. As shown in Fig. 1 (c), we observed the DLP of about 20% for X\(^0\) PL under a zero magnetic field.

3.2. Model calculation

For more detailed analysis, we introduce the exciton pseudospin description. If we treat the bright exciton doublet (±1) as an effective 3D pseudospin with S=1/2, the exciton spin state can be depicted as a vector \(S\) in Bloch space. Additionally, the mutual conversion was considered between \(S\) and \(\rho\) which is a vector in Poincare sphere representing the light polarization. In this picture, the dynamics of the exciton spin (to be more exactly “pseudospin”) is described by the following rate equation [3, 6],

\[
\frac{dS}{dt} = \Omega \times S - \frac{1}{\tau_s} (S - P_{eq}) - \frac{1}{\tau_R} (S - P_{ex})
\]

where \(\tau_s\) and \(\tau_R\) are the exciton decay times caused by the spin relaxation and recombination, respectively. The vector \(P_{ex} = (P_t, P_r, P_c)\) is the exciton polarization vector in Poincare sphere, and is copied as the initial spin state in Bloch space. Here \(P_t\) and \(P_r\) are the DLP along [110] and [100] crystallographic axes, and \(P_c\) is the degree of circular polarization (DCP: projection to [001]). The first term of the rhs of Eq. 1 represents the coherent spin precession induced by the effective field \(\Omega\), and second (third) term explains the decay of spin vector due to the exciton spin relaxation (recombination). Under a zero external field, the torque vector \(\Omega\) can be written as \(\Omega = (\Omega_{xyz}, 0, 0)\), and lies in the equatorial plane of the sphere. It is noteworthy that the polarization equilibrium \(P_{eq} = (Y, 0, 0)\) is included in this model. \(Y\) leads to the convergence direction of \(S\) in xy-plane. Here, \(Y\) is used as a parameter of built-in linear dichroism of QD, and originates from the effects of valence-band mixing and thermalization. Although the main origin of \(Y\) has not been identified yet, this treatment brings a flexible way to the calculation. For simplicity, we assume that \(Y\) has the same sign with \(\Omega_{xyz}\) and is in the [110] axis. The information of steady state \(S\) is transferred to \(\rho\) when the exciton is recombined, and gives the polarization of the emission. By using the Stokes parameters \(\rho_t\), \(\rho_r\) and \(\rho_c\), which describe the DLP along [110], [100] and the DCP, \(\rho\) can be written as \(\rho = (\rho_t, \rho_r, \rho_c)\) where they satisfy the following relation: \(|\rho| = \sqrt{\rho^2_t + \rho^2_r + \rho^2_c} \leq 1\).

From Eq. 1, the steady states of the polarizations can be derived as follows;

\[
\rho_t = \left(\frac{Y_t}{\tau_s} + \frac{P_t}{\tau_R}\right)T_s
\]

\[
\rho_r = \frac{T_s}{\tau_R} \left[ \frac{P_r}{\tau_s + (\Omega_{exc}T_s)^2} - \frac{\Omega_{exc}P_r}{1 + (\Omega_{exc}T_s)^2} \right]
\]

\[
\rho_c = \frac{T_s}{\tau_R} \left[ \frac{P_c}{\tau_s + (\Omega_{exc}T_s)^2} - \frac{\Omega_{exc}P_c}{1 + (\Omega_{exc}T_s)^2} \right]
\]

Here, \(T_s\) is the exciton spin lifetime and is represented as \(T_s = (\tau^{-1}_s + \tau^{-1}_r + \tau^{-1}_c)^{-1}\). As shown by Eqs. 3-4, the polarization...
As a result, $\Delta \theta$ comes close to a number of InAlAs QDs with the calculated curves are depicted by changing $\delta$ as a function of $\tau$ by using the recombination lifetime $\tau$ respectively. From both figures, it is shown that $\Omega/\rho$ing technique for the same InAlAs QD ensemble under a zero magnetic field ($\tau_0 = 0.01$ by $\tau_0 \sim 5\tau_R$, $\tau_3 \sim 3$ ns, $\tau_1 = 0.5$, respectively. (c) Cross-section curve at the dashed line in (b). At the external field $B_x = \delta_0/g_\rho \mu_B$, the maximum value of DLP was observed.

As seen in this section, the spin precession model and the introduction of $Y_l$ offer a helpful way for the intuitive understanding of the exciton spin dynamics. However, high DLP under a zero magnetic field as shown in Fig. 1 (c) cannot be obtained only by the effect of $Y_l$. In order to explain this high DLP, we shall consider the effect of optically created nuclear field ($B_N$). As widely known, a continuous circular polarized excitation brings a non-zero magnitude of $B_N$, which works as an additional magnetic field along $z$-direction. With an effective formation of $B_N$ via a positively charged exciton state by wetting layer excitation (i.e. non-resonant excitation), Belhadj et al. demonstrated the efficient polarization conversions under a zero magnetic field for single InAs QDs [9]. Although our experimental conditions (zero-magnetic field, quasi-resonant excitation, even a small $B_N$ corresponding to the energy shift of $\sim 5 \mu$eV induces the observed high DLP [10]. More quantitative discussion will be seen elsewhere.

3.3. Under longitudinal magnetic field

In order to investigate the exciton spin dynamics more in detail and to achieve more efficient polarization conversion, similar experiments (that is, $C \rightarrow L$ conversion) under the longitudinal magnetic field $B_z$ were performed for a typical single QD. $B_z$ lifts the torque vector off the $xy$-plane $\Omega = (\Omega_{\text{exc}}, 0, \Omega_z)$ as shown in Fig. 3 (a). $\Omega_z$ is the precessional frequency due to the Zeeman splitting, and written as $\Omega_z = g_X \mu_B B_z/\hbar$, where $g_X$ is the exciton g factor in growth direction and $\mu_B$ is Bohr magneton. For this InAlAs QD, $g_X$ is found to be $2.20 \pm 0.01$ by magneto-PL measurement under linearly polarized excitation. Fig. 3 (b) shows 2D-plot of DLP of $X^0$-PL as functions of $B_z$ and $\varphi$ (upper panel). Not only the DLP amplitude but its pattern is reproduced very well by the calculated shown in the lower panel. Here the following parameters were used; $\tau_R = 0.75$ ns, $\tau_3 = 3$ ns, and $Y_l = 0.5$. 

conversion from circular to linear ($P_x \rightarrow \rho_x$) and vice versa ($P_y \rightarrow \rho_y$) can be predicted. It should be noted that the observed DLP can be written as $\rho(\varphi) = \rho_1 \cos(\varphi) + \rho_2 \sin(\varphi)$ in the laboratory frame.

Let us consider the phase difference of DLP depending on the excitation orientation. The initial spin states created by circularly polarized photons ($\sigma_+$, $\sigma_-$) are positioned at the poles of Bloch sphere. Though each vector starts the precession around $\Omega$, the axes of observed DLPs are also symmetrical as for [110] because both $\Omega$ and $Y_l$ distributions are directed to [110] in our assumption. If the variation of DLP axis from [110] is defined as $\Delta \theta$, the angle difference of $2\Delta \theta$ is observed from the $\sigma_+$ and $\sigma_-$ excited DLP curves (Fig. 2 (a)). We measured the $\Delta \theta$ for a number of InAlAs QDs with the different FSS and plotted as a function of $\delta_0$ in Fig. 2 (b) and (c). In those figures, the calculated curves are depicted by changing $\tau_x/\tau_R$ and $Y_l$, respectively. From both figures, it is shown that $\Delta \theta$ decreases with increasing $\delta_0$. If a QD has a large $\delta_0$, the spin vector precesses with a large angular frequency, and the steady state value comes close to $Y_l$ regardless of the excitation polarization $P_{\text{exc}}$.

As a result, $\Delta \theta$ reduces with increasing $\delta_0$. Compared with the calculation curves, the spin relaxation time of neutral exciton $\tau_x$ and the polarization equilibrium $Y_l$ can be estimated roughly by using the recombination lifetime $\tau_R = 0.75$ ns, which is obtained by time-resolved PL measurements via a streak camera for a single InAlAs QD in the same sample. Since $Y_l$, $\tau_x$, and $\tau_R$ may be different for individual QDs, dispersion from calculated curves is observed. However, many QDs are around the curve with $\tau_x \sim 4\tau_R$, $Y_l = 0.5$, which are typical for average In$_{0.75}$As$_{0.25}$QDs. The obtained spin relaxation time agrees well with our previous results evaluated by the four-wave mixing technique for the same InAlAs QD ensemble under a zero magnetic field ($\tau_x \sim 5\tau_R$) [7], and the photon correlation method for a single InAlAs QD ($\tau_x \sim 3.6\tau_R$) [8].
In addition, we shall consider the slant of DLP pattern observed in Fig. 3 (b). Compared with the calculated results, the angle at which maximum (or minimum) of DLP is detected is slightly tilted with respect to $B_z$. At the present stage, the effectively inclined magnetic field seems plausible candidate for the origin of this slant. If the sample growth axis and an external field $B$ are not parallel for some technological reasons, in-plane component of the torque vector ($\Omega$) changes its magnitude and direction depending on $B$. Since the direction of $\Omega$ decides the polarization axis, the observed DLP pattern may be distorted by an effectively changing in-plane magnetic field. Though this slant pattern could be explained in the same model, more detailed discussion needs other experimental verification.

Let us focus on the profile (Fig. 3 (c)) at the dashed line in the 2D plot (Fig. 3 (b)). At $|B| \sim 0.35$ T, the DLP shows the maximum (minimum) when $B_z$ was positive (negative). Zeman splitting induced by this field has just the same energy of FSS (i.e. $\delta_0 = g\mu_B B_z$), and the conversion efficiency reaches $\sim 50\%$, which is the maximum value predicted by the above model (Eq. 1). Further increase of $|B|$ reduces the DLP amplitude, and this indicates the polarization of emitted light is close to circular. Since the magnetic confinement corrects the potential asymmetry, the effect of AEI will be diminished under a large magnetic field (several Teslas). In addition, the amplitude of DLP is the asymmetric about $B_z = 0$ line. This can be explained by the effect of spin equilibrium $Y$. In the case of $B_z > 0$, the observed DLP (strictly speaking, $\rho_\parallel$) increases from the initial value because $\Omega$ (or $S$) and $Y$ are in the same half sphere $(x \geq 0)$. However, in the case $B_z < 0$, since $\Omega$ directs oppositely and $S$ precesses in the half sphere $(x \leq 0)$ which does not contain $Y$, $S$ is relaxed gradually into $Y$ during the precession. Consequently, DLP becomes small. Though the precise evaluation of its magnitude is difficult, this fact implies $Y$ has some finite value and can be estimated as $\approx -0.5$ for this target QD. The calculated results using $Y_l = 0.5$ and $\tau_R = 0.75$ ns, were obtained as shown by the solid curve in Fig. 3 (c), and $\tau_s$ is roughly estimated to be 3 ns. This is quite consistent with the estimation of $\tau_s = 4 \tau_R$ obtained under a zero magnetic field.

4. Summary

In summary, the circular to linear polarization conversion was investigated for single InAlAs QDs under zero and nonzero magnetic fields. Quasi-resonant excitation made possible to inject the highly polarized exciton spin, and consequently the polarization conversion with high efficiency $\sim 50\%$ was achieved under a longitudinal magnetic field. Regarding the bright exciton doublets as two-level system, the dynamics was very well described by a simple rate equation which includes the spin equilibrium due to built-in linear dichroism. Measuring the angle difference of DLP or the conversion efficiency, $Y_l$ and $\tau_s/\tau_R$ were estimated. By using the recombination lifetime $\tau_R = 0.75$ ns which obtained by other independent measurement, we evaluated $\tau_s$ and $Y_l$ as $\sim 3$ ns and 0.5, respectively for the target QD. The obtained $\tau_s$ under the magnetic fields was in good agreement with our results for the same InAlAs QD without a magnetic field. This long spin relaxation, exceeding the recombin-