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Polarization-dependent shift in excitonic Zeeman splitting of self-assembled $In_{0.75}Al_{0.25}As/Al_{0.3}Ga_{0.7}As$ quantum dots

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We report optical spectroscopic results of a single self-assembled $In_{0.75}Al_{0.25}As/Al_{0.3}Ga_{0.7}As$ quantum dot. The polarization-dependent shift of the Zeeman splitting in a single InAlAs quantum dot (QD) has been observed. The induced Overhauser field is estimated to be ~0.16 T in this InAlAs QD and the magnitude is shown to be controllable by the degree of circular polarization of excitation light.

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Semiconductor self-assembled quantum dots (QDs) exhibit a variety of confinement-related optical and electronic properties useful for optoelectronic device applications such as QD lasers and detectors. In particular, broad efforts are currently underway to develop new techniques for controlling spin degrees of freedom in QDs. These efforts are stimulated in part by some proposals to use the spin systems as quantum bits (qubits) in quantum information processing.[1–3] While the rapid spin relaxation in solid-state surroundings was suggested to be the main obstacle for the realization of coherent control of spins, the exciton spin relaxation is getting recognized to far exceed the exciton lifetime and lasts up to several nanoseconds if excitons are excited and detected resonantly.[4,5] As another problem, the influence of a nuclear-spin-induced magnetic field on electronic energy states is pointed out. Since the relaxation time of the nuclear spin is extremely long, the induced electronic energy shifts due to nuclear-spin polarization will cause errors in quantum gate operations using magnetic fields.[6] On the other hand, it can be possible to apply this nuclear-spin polarization for long-lived quantum memory in a quantum information network.[7] Therefore, the magnitude of the nuclear-spin-induced magnetic field and its controllability in a self-assembled QD should be studied experimentally.

In this work, we report the magnetic-field studies of a single self-assembled InAlAs/AlGaAs QD. The polarization-dependent energy shift of excitonic emission, commonly known as the Overhauser shift, is clearly observed and the magnitude is shown to be controllable. The observation of the Overhauser shift in a naturally formed QD using monolayer fluctuation of a quantum well (QW)[8,9] has been reported earlier. However, there have as yet been no observations of the Overhauser shift in a self-assembled QD that is suitable for formation of vertically coupled QDs. Further, this work provides valuable information on the red-emitting InAlAs/AlGaAs QDs which have been reported in only a few studies.[10–13] Most of the single QD measurements have been performed thus far in combinations such as InAs/GaAs (Refs. 14,15) and InGaAs/GaAs (Ref. 16) with infrared emission.

The QD samples grown by molecular-beam epitaxy have two QD layers ($In_{0.75}Al_{0.25}As$ and $In_{0.7}Ga_{0.3}As$) separated by an 11-nm-thick $Al_{0.3}Ga_{0.7}As$ layer. The QDs are formed using the spontaneous island formation in Stranski-Krastanow growth mode during the epitaxy of strained InAlAs (or InGaAs) on AlGaAs layers grown on CrO-doped (100) GaAs substrates. A GaAs cap terminates the heterostructure. In this study, we concentrate on the single QD emission from the InAlAs QDs in this sample. The details of the sample are seen in Refs. 17 and 18.

Figure 1(a) shows the time-integrated photoluminescence

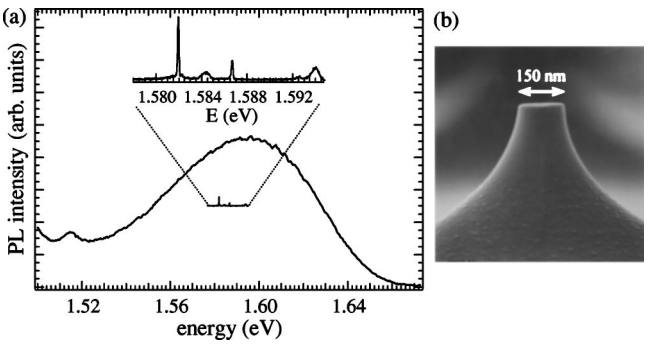

FIG. 1. (a) Ensemble PL spectra and single QD emissions (inset) from InAlAs QDs. (b) TEM image of a mesa structure.

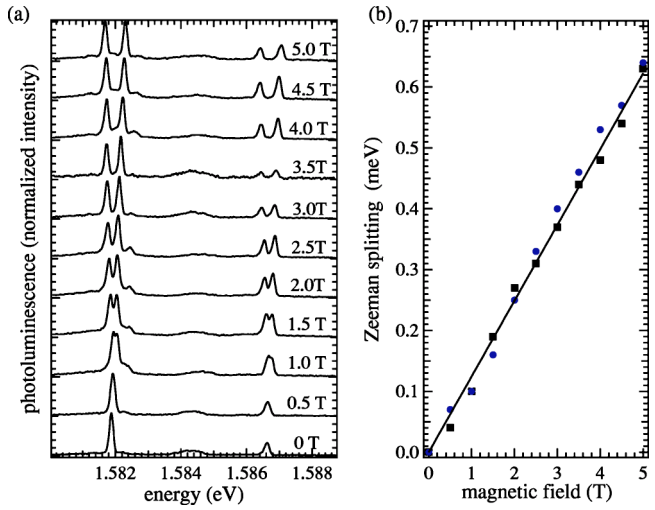

FIG. 2. (a) Photoluminescence spectra from the lowest exciton states of two different InAlAs QDs recorded at different magnetic fields. The spectra are normalized by the magnitude of the exciton emission around 1.5818 eV. (b) Magnetic-field dependence of the energy splitting of two exciton lines in InAlAs QDs shown in (a). Solid squares (for higher energy exciton) and circles (for lower energy exciton) are experimental data and the line is the fitting result using the forms obtained by diagonalizing the exciton fine-structure Hamiltonian.

(PL) spectra at 10 K from the excitation spot with a diameter of $\sim 150\ \mu$m. The excitation has been carried out with a HeNe laser on an $Al_{0.3}Ga_{0.7}As$ barrier. At the lowest excitation intensity, the peak centered around ~ 1.59 eV for InAlAs QDs is observed. The PL spectra have the linewidth of ~ 120 meV due to inhomogeneous QD size distribution. The emission from the wetting layer (WL) was observed at 1.689 eV for larger excitation intensity (not shown here).

In order to isolate a single QD, small mesa structures were fabricated by electron-beam lithography and wet chemical etching as shown in Fig. 1(b). The typical top lateral size of the mesa structure is 150 nm. Since the QD density is estimated as $\sim 5\times 10^{10}\ \mathrm{cm}^{-2}$, a mesa contains several QDs on an average and some mesas have one or a few QDs, from which well-separated sharp emissions appear by conventional far-field spectroscopy as shown in the inset of Fig. 1(a). For the single QD spectroscopy, the sample was held in a LHe cryostat and was kept at 4.2 K. All data shown here were taken under the HeNe laser excitation (632.8 nm). The QD emissions were dispersed by a triple grating spectrometer ($f=0.64$ m) and were detected with a LN_2-cooled Si-charge-coupled device (CCD) camera. The system resolution was 13 μeV and the energies of the emission peaks can be determined to be of the order of 5 μeV by spectral fitting. A magnetic field up to 5 T was applied to the sample along the growth direction. The polarization of the PL emissions was analyzed with a quarter-wave plate (QWP) and a linear polarizer in front of the spectrometer.

Figure 2(a) shows the PL spectra obtained from the lowest exciton states of two different InAlAs QDs by varying the magnetic field up to 5 T at 4.2 K. The magnetic field was aligned parallel to the heterostructure growth direction z and the sample was excited in Faraday configuration. The excitation was linearly polarized and its power was decreased to a level at which the biexciton and excited states disappear in the spectra. In the energy range of the figure, the exciton recombination of two different QDs at $B=0$ T is located at 1.5818 eV and 1.5866 eV. Under the application of a low magnetic field ($\leqslant 1.5$ T) where Zeeman splitting of the exciton line is not clearly observed, the emission has been analyzed with respect to its circular polarization. The zero-field emission at 1.5818 eV has a full width at half maximum (FWHM) of $\sim 90\ \mu$eV and the linewidth varies from dot to dot within 30–200 μeV. While a very small energy splitting ($\sim 14\ \mu$eV) was detected at $B=0$ T depending on the observed QDs, they showed no significant linear polarization.[16] In $B\neq 0$ T, the emissions split into a doublet due to Zeeman interaction of the exciton spin with the magnetic field. The low-energy portion of the spectrum was found to be σ_+ polarized and the high-energy portion was σ_- polarized. Further, in these QDs, a single emission was the result of the recombination of the degenerate $m_j=\pm 1$ excitons and the application of magnetic field results in the splitting of $m_j=+1$ and $m_j=-1$ states. A diamagnetic shift Δ_{diamag} to higher energies, which is the energy shift of the center of the exciton doublet, is expressed well by $\Delta_{\mathrm{diamag}}=\alpha B^2$. With respect to QDs in Fig. 2, the coefficient α is found to be very small ($3.4\ \mu\mathrm{eV/T}^2$). Thus far, the value of α has been reported as $8.6\pm 0.9\ \mu\mathrm{eV/T}^2$ for $In_{0.55}Al_{0.45}As/Al_{0.35}Ga_{0.65}As$ QDs (Ref. 11) and $0.8\pm 0.3\ \mu\mathrm{eV/T}^2$ for $In_{0.64}Al_{0.36}As/Al_{0.33}Ga_{0.67}As$ QDs.[12] Since the diamagnetic shift is proportional to the squared average of the lateral extension of the exciton wave function, the small value of α indicates a strong confinement. Moreover, it is natural to observe the different value of α, depending on the lateral dot size, particularly for the self-assembled QDs. In fact, in this sample, some of the QDs exhibited larger diamagnetic shifts.

In Fig. 2(b), Zeeman splitting is plotted against the external magnetic field. The exciton energies in Faraday configuration are given by the following Hamiltonian using the exciton states $|m_j\rangle=|+1\rangle,|-1\rangle,|+2\rangle,|-2\rangle$ as the basis:

$$H = H_{\mathrm{exchange}} + H_{\mathrm{Zeeman}} = \frac{1}{2}\begin{pmatrix} \delta_0 & \delta_b & 0 & \\ \delta_b & \delta_0 & & \\ 0 & -\delta_0 & \delta_d & \\ & \delta_d & -\delta_0 & \end{pmatrix} + \frac{\mu_B B}{2}\begin{pmatrix} g_{bx} & 0 & 0 & 0 \\ 0 & -g_{bx} & 0 & 0 \\ 0 & 0 & -g_{dx} & 0 \\ 0 & 0 & 0 & g_{dx} \end{pmatrix}. \quad (1)$$

In the above equation, $\delta_0, \delta_b, \delta_d$ are the exchange energy between bright ($|\pm 1\rangle$) and dark ($|\pm 2\rangle$) excitons, the splitting energy between bright excitons, and the splitting energy between dark excitons, respectively. The μ_B is the Bohr magneton and $g_{bx}(g_{dx})$ is the g factor of bright (dark) excitons, given by $g_e^z+g_h^z(g_e^z-g_h^z)$ using electron and hole g factors in the growth direction.

By fitting using the forms obtained from diagonalizing the above exciton Hamiltonian, we obtain an exciton g factor $g_{bx}=2.10\pm 0.03$. A number of single InAlAs QDs were studied in this sample, and the Zeeman splitting changed slightly from dot to dot within 0.1 meV.

In this section, the results of the excitation of circular polarization are reported. In the case of circularly polarized excitation, the formation of the nuclear-spin polarization via hyperfine interaction with spin-polarized electrons is expected. As previously mentioned, the polarization-dependent shift (Overhauser shift) of a single QD emission was observed in a natural GaAs/AlGaAs QD where excitons are trapped in the monolayer fluctuation of QW width;[8,9] however, the energy shift has not yet been observed in the case of a self-assembled QD.

Nuclear-spin polarization is formed by a two-step process in optical pumping. The formation of electron-spin polarization is achieved by circularly polarized optical excitation in a longitudinal external magnetic field. Next, the electron-spin polarization is transferred to the nuclear system via hyperfine interaction,[19] the Hamiltonian for which is given by

$$H_{HF} = v_0 \sum_j A^j |\psi(\mathbf{R}_j)|^2 \left(I_z^j S_z + \frac{I_+^j S_- + I_-^j S_+}{2} \right). \qquad (2)$$

In Eq. (2), v_0 is the unit-cell volume, A^j is the hyperfine constant, and $|\psi(\mathbf{R}_j)|^2$ is the electron density at the jth nuclear site $\mathbf{R}_j$. The interaction consists of two terms: a term proportional to the electronic S_z and nuclear I_z spin polarizations along the direction of the external magnetic field, and a term including electron and jth nuclear raising and lowering operators $S_{+/-}$ and $I_{+/-}^j$, respectively. The second term describes the dynamic part of hyperfine interaction, i.e., the mutual electron-nuclear spin flips. Through the second term, electron-spin polarization is transferred to a nuclear-spin system. The resultant nuclear-spin polarization then generates a static effective nuclear magnetic field B_N, via the first term, inducing the electronic energy shift. This energy shift is known as the Overhauser shift.[19] The hole in the valence band has a p-like wave function that vanishes at the position of the nucleus. Thus, only the electron-spin polarization contributes to the formation of the nuclear-spin polarization. The Overhauser field B_N is given by the following:

$$\langle H_{HF} \rangle_N = A \langle I_z \rangle S_z = \mu_B g_x S_z B_N, \qquad (3)$$

where A is the summation of A^j over all the nuclei in a unit cell and $\langle I_z \rangle$ is the average nuclear-spin polarization that is determined by the balance of the nuclear-spin polarization rate and its depolarization rate. From the above, the Overhauser shift ΔE_{OH}in QDs should be observed as $g_x \mu_B B_N$ in the excitonic Zeeman splitting for circularly polarized excitation.

Figure 3(a) shows the evolution of the exciton emission in an external magnetic field when exciting a single QD with circularly polarized light. In the case of excitation with σ_+-polarized light, the energy levels are resolvably split at 0.5 T into two levels. As the magnetic field increases up to 5 T, the splitting of the two levels increases. A similar splitting is observed by increasing the magnetic field in the opposite direction (up to −5 T); however, the splitting energy is found to be slightly different from that for 0–5 T except in the range $|B| \leqslant 0.5$ T as shown in Fig. 3(b). While this difference is marginal (38 μeV on an average), an identical result was found in conversion from σ_+ to σ_- excitation under the magnetic field in the same direction. These observations can be explained by the aforementioned Overhauser field B_N. Under σ_+ excitation in our system, the induced B_N decreased the external magnetic field B (i.e., $B-B_N$), while B_N increased the external magnetic field in the opposite direction (i.e., $-B-B_N$). Further, the direction of B_N is determined by the direction of the electron spin, which, in turn, is determined by the polarization of the exciting light. In fact, the observed Zeeman splitting ΔE changed clearly depending on the degree of circular polarization of the excitation light as shown in Fig. 4. In Fig. 4, ΔE is plotted as a function of the rotational angle of the QWP. As expected, very good agreement with the $\cos 2(\theta - 45°)$ (solid curve) was obtained for B_N. Figure 4 also shows the controllability of the B_N by using the degree of circular polarization. The energy difference in Fig. 3(b) is given as $\Delta E_{OH} = g_{bx} \mu_B B_N$ by Eq. (2) and B_N is calcu-

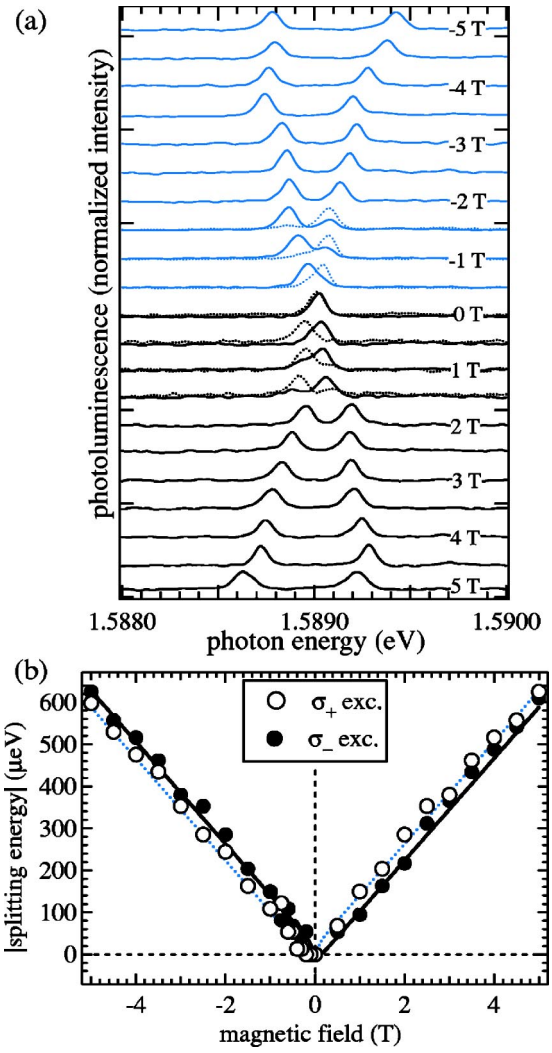

FIG. 3. (a) Excitonic emission energies for σ_+ excitation. In $|B| \leqslant 1.5$ T, where the splitting is not clear, the σ_+ emissions (dotted lines) and σ_- emissions (solid lines) were detected separately by using a quarter-wave plate and a linear polarizer. (b) Absolute values of the splitting energy are plotted for σ_+ excitation (open circles) and σ_- excitation (black circles). The lines are the fitting curves by Eq. (1), and including Eq. (3), for σ_- (solid) and σ_+ (dotted) excitations, respectively.

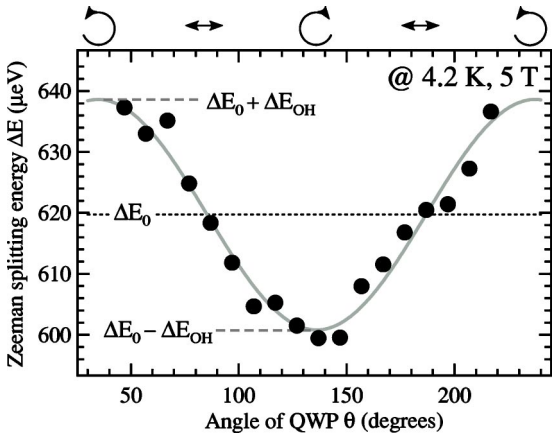

FIG. 4. Energy splitting ΔE at 5 T is plotted as a function of the angle θ of the quarter-wave plate used to change the excitation polarization. The polarization of the excitation light is illustrated above the top axis. In the figure, ΔE_0 is the Zeeman splitting for linearly polarized excitation that is indicated by the dotted line. The solid curve is a fitting line.

lated to be 0.16 ± 0.01 T for g_{bx} of 2.1 and ΔE_{OH} of 19 μeV.

The large nuclear spins of In ($I=9/2$) and Al ($I=5/2$) strongly affect the magnitude of the hyperfine interaction in InAlAs QDs as compared to GaAs QDs, where Ga and As have the same nuclear spins ($I=3/2$). Since the estimation of hyperfine constant A of $In_{0.75}Al_{0.25}As$ is presently difficult, assuming that the value of AI is similar to the value for InAs (Ref. 20) and that the nuclear polarization rate $\langle I \rangle / I$ is the same for all nuclear species, $\langle I \rangle / I (=\Delta E_{OH} / \Sigma_\alpha A_\alpha I_\alpha)$ is approximately 19 μeV/323 μeV=6%. Brown and Gammon observed that $\Delta E_{OH} \sim 90$ μeV and $\langle I \rangle / I \sim 65\%$ for a natural GaAs QD in a 4.2-nm-wide QW.[8,9] The possible physical origin of the smaller nuclear polarization as compared to GaAs QDs is supposed to be due to the low spin-polarization rate of the optically injected electrons and due to the magnetic spin disorder of InAlAs lattice that consists of nuclei with different spins. Further research on nuclear-spin-polarization dynamics is in progress.

In summary, we have observed the polarization-dependent shift of the Zeeman splitting in a single InAlAs QD. While the energy difference (19 μeV) and the induced hyperfine field (0.16 T) were small in the InAlAs QD, the magnitude was shown to be controllable by the degree of circular polarization of excitation light.

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