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Luminescence quantum beats of strain-induced GaAs quantum dots

Kazuhiro Nishibayashi, Tsuyoshi Okuno, Yasuaki Masumoto, and Hong-Wen Ren

Institute of Physics, University of Tsukuba, Tsukuba, Ibaraki 305-8571, Japan

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Quantum beats of the strain-induced GaAs quantum dots were observed in the time-resolved photoluminescence in the magnetic field parallel and perpendicular to the growth direction. Quantum beats observed under the longitudinal magnetic field are caused by quantum interference of bright excitons showing Zeeman splitting. The oscillation period depends on the angle between the growth direction of the crystal and the magnetic field. Analysis based on the spin Hamiltonian for excitons explains the observed data and gives $g$ factors 0.51, 0.17, and 0.34 to the exciton, electron, and heavy hole, respectively. Quantum beats coming from electron Larmor precession were observed under the transverse magnetic field. The isotropic electron $g$ factor is observed in contrast to the anisotropic electron $g$ factor for the corresponding quantum well and is ascribed to the strain-induced opposite energy shift of heavy- and light-hole bands.

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I. INTRODUCTION

The semiconductor quantum dots (QDs), artificial atoms, have quantized energy levels characterized by orbital and spin angular momenta similar to atomic levels. Long optical coherence of exciton and spin in semiconductor quantum dots is expected to apply to quantum computation and quantum information processing. Spin coherence time is known to be much longer than exciton coherence time in bulk and quantum structures. Further, carrier spin relaxation is expected to be greatly suppressed by quantum confinement. Nevertheless, study of spin-relaxation time and spin structures in quantum dots is still in the elementary stage. Therefore, new knowledge on the spin structure and the spin dynamics becomes important. Static and transient optical orientation, magneto-optic spectroscopy, spin-flip Raman scattering, and quantum beats have given us valuable information on spin-dependent energy levels of bulk and quantum well (QW) semiconductors. Especially, quantum beats are known to be an efficient method for studying a fine energy structure and spin dynamics of carriers in semiconductors because they are not limited by the spectral resolution and hence give us finest energy splitting and because they also give us lower limit time of coherence between spin-split sublevels.

Quantum beats in semiconductor quantum structures under the magnetic field have been reported. Time-resolved pump-and-probe measurement revealed the electron and hole $g$ factors in quantum wells. In the time-resolved luminescence measurement, quantum beats were observed under the resonant excitation of quantum wells. Time-resolved luminescence revealed hole spin quantum beats. Although many observations of quantum beats have been reported in quantum wells, the observations of quantum beats for quantum dots are limited. This is ascribed to the large inhomogeneous broadening in the ensemble of quantum dots.

The utilization of stress caused by lattice mismatch between different semiconductors is an ideal method for fabrication of well-characterized quantum dots. Quantum dots are formed in a single quantum well by the stress modulation from the self-assembled islands fabricated on the surface. Self-assembled islands cause tensile strain perpendicular to the growth direction and therefore, the potential energies of the conduction band and valence band form additional harmonic potential wells laterally in the quantum well layer. Almost equally spaced quantum energy levels are observed and the formed quantum structures are called strain-induced quantum dots. The strain-induced quantum dots have homogeneous size distribution in the growth direction and no defect at the interface. They have well-characterized quantum energy levels which are suitable for the precise optical study. In this paper we report the first, to the best of our knowledge, observation of two kinds of quantum beats coming from the Zeeman splitting of bright excitons and electron Larmor precession in strain-induced quantum dots.

II. SAMPLES

The samples were grown by metal organic vapor phase epitaxy at 60 Torr on semi-insulating GaAs (001) substrates. Single GaAs-Al$_{0.3}$Ga$_{0.7}$As quantum wells were grown. The thickness of the GaAs quantum well is 3.9 nm in one sample and 4.8 nm in another sample. The Al$_{0.3}$Ga$_{0.7}$As top barrier layer was 9.6-nm thick. After the growth of the single quantum well, a 2.4-nm GaAs cap layer was grown. Finally, InP islands were formed by depositing four monolayer InP as stressors. The islands were ~90-nm wide and ~16-nm high. The areal density of the islands was $\approx 3 \times 10^9$ cm$^{-2}$. An additional sample consisting of single quantum wells 2.4-nm, 4.8-nm, and 9.6-nm thick without InP stressors was also grown for the measurement of the well-width dependent $g$ factor of the electron.

III. PHOTOLUMINESCENCE AND PHOTOLUMINESCENCE EXCITATION SPECTRA

Photoluminescence (PL) spectrum of the sample whose GaAs well width is 3.9 nm, excited at 2.33 eV by using a continuous-wave (cw) Nd$^{3+}$:YVO$_4$ laser is shown in the bottom panel of Fig. 1. The luminescence located at 1.64 eV...
in the middle panel and QD3 denotes the absorption by the third excited state of the dots. There is a sharp peak at 1.632 eV. The energy difference between this peak and the detected energy position is 35 meV which is equal to the longitudinal optical (LO) phonon energy of GaAs. Therefore, the origin of this peak is the luminescence through one LO-phonon relaxation. The peaks at 1.653 eV and 1.680 eV correspond to heavy-hole (HH) exciton and light-hole (LH) exciton in a quantum well, respectively. Clear difference is detected for \( \sigma^+ \) and \( \sigma^- \) luminescence. The degree of circular polarization \( \rho_c = (I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-}) \) is as much as 0.4 at the LO-phonon peak, as is shown in the top panel of Fig. 1. When the HH state was excited, \( \rho_c \) is 0.5, the highest value. When the LH state was excited, \( \rho_c \) is negative and is \(-0.05\). Based on the selection rule for the absorption and luminescence transitions by circularly polarized light,\(^{18} \) negative polarization at the LH exciton of the quantum well and positive polarization at the HH exciton of quantum well show that quantum dot state is composed of the HH state while decrease of \( \rho_c \) from 0.5 to 0.4 at the quantum dot state may contain the LH state of the quantum well.

**IV. EXCITON QUANTUM BEATS IN THE LONGITUDINAL MAGNETIC FIELD**

The sample was mounted in a cryostat with a superconducting magnet at 5 K for the magneto-optic study. The direction of an external magnetic field is along the epitaxial growth direction. For the measurement of time-resolved photoluminescence, 2-ps pulses from a mode-locked Ti:sapphire laser were used for the excitation at the repetition rate of 82 MHz. The excitation and detection paths were along the magnetic field (Faraday configuration) and in opposite directions. The excitation pulses were linearly polarized. The luminescence components with linear polarization parallel (\( || \)) or perpendicular (\( \perp \)) to that of the excitation laser were selected by a half wave plate and a linear polarizer. The luminescence was dispersed by a 25-cm subtractive-dispersion double monochromator and detected by a synchroscan streak camera. The typical time resolution of the system was \( \sim 10 \) ps.

The excitation energy was carefully selected based on the PLE spectrum in Fig. 1. When the excitation energy is above the quantum well, the luminescence intensity of the QD1 band is high and most of quantum dots show luminescence, as is shown by a solid line in the inset of Fig. 2. This condition is not favorable for observation of the quantum beats because luminescence of quantum dots is inhomogeneously broadened. When the excitation energy is below the quantum well, the luminescence intensity of QD1 is reduced, because the light absorption by a selected ensemble of quantum dots is much weaker than the light absorption either by a quantum well or by a thicker barrier layer. However, in this case, narrow-band picosecond pulses site selectively excite an ensemble of quantum dots of almost the same size. The fast LO-phonon-mediated relaxation makes sharp sideband luminescence of site-selectively excited quantum dots. In the Faraday configuration, luminescence was detected at \( E_{\text{det}} \) under

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FIG. 1. Bottom panel: Photoluminescence (PL) spectrum of strain-induced GaAs quantum dots excited at 2 K with a cw Nd\(^{3+}:\)YVO\(_4\) laser. The GaAs well width is 3.9 nm. Middle panel: Circularly polarized photoluminescence excitation spectra. The detected energy is set to the first excited state of the quantum dots, which is obtained from the luminescence measurement. Vertical scale is expanded by ten times between 1.60 eV and 1.64 eV. The closed (open) circles show the same (opposite) circular polarization as that of the excitation laser. Top panel: Degree of circular polarization \( \rho_c \) of photoluminescence excitation spectrum.

is attributed to heavy-hole exciton recombination in the GaAs quantum well without modulation by stress. In the lower-energy region around 1.60 eV, the luminescence from strain-induced quantum dots is observed. In the figure, QD1 and QD2 denote the PL bands of the first and second excited states of dots, respectively, whose luminescence nonlinearity was reported previously.\(^{17} \) The energy separation between the first and second excited states of dots is 16 meV which was also seen clearly in the nonlinear luminescence.\(^{17} \) Circularly polarized photoluminescence excitation (PLE) spectrum was measured to specify the energy states of the dots more precisely. A tunable cw Ti:sapphire laser was used as an excitation source and its intensity was kept constant by an acousto-optic modulator. Circularly polarized light was made by a linear polarizer and a quarter-wave plate. The luminescence components of the same circular polarization as the excitation laser (\( \sigma^+ \)) and its opposite (\( \sigma^- \)) were selectively measured by using another quarter-wave plate and a linear polarizer placed in front of a 1-m double monochromator. The luminescence intensity was measured by a liquid-nitrogen-cooled charge-coupled-device camera by changing the wavelength of the excitation laser.

The circularly polarized PLE spectra are shown at the middle panel of Fig. 1. The detected energy is set to the peak of the QD1 band (1.597 eV). Several structures are observed. The energy of the second excited state of the dots, QD2 in the bottom panel, agrees with the absorption structure QD2

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the excitation at the QD1 energy plus the LO-phonon energy, $E_{\text{ex}} = E_{\text{det}} + \hbar \omega_{\text{LO}}$ (see the inset in Fig. 2).

Time traces of luminescence under magnetic field of $B = 0$ T and $B = 6$ T are shown in Fig. 2. The solid trace was measured at parallel linear polarization. The trace plotted by circles was measured at perpendicular polarization. At $B = 0$ T, the polarization decay time is about 15 ps. The photoexcited electron-hole pairs immediately relax to the QD1 state by emitting LO phonon, and then they recombine. The intensity of parallel linear polarization component corresponds to the number of carriers that conserve the polarization memory in the LO-phonon relaxation. The difference of two curves shows that the initial degree of linear polarization $\rho_0 = (I_1 - I_2)/(I_1 + I_2)$ is $\sim 0.2$. When a magnetic field is applied, a damping oscillation structure appears upon the decay profile. The parallel component at $B = 6$ T decreases faster than that at $B = 0$ T and is followed by damped oscillations. Still more dramatic changes were observed on the perpendicular linear component. After the quick rise around $t \sim 12$ ps, the remarkable oscillation appeared. The parallel and perpendicular linear polarization traces are antiphase and they coincide after 60 ps. Excitation by circular polarization did not induce the oscillation of luminescence. These polarization selection rules for the observation of the oscillation in the Faraday geometry suggest the quantum beat of bright excitons. Linearly polarized light which is the superposition of right-handed and left-handed circularly polarized light excites both of the Zeeman-split bright excitons coherently and the quantum interference of the bright excitons shows quantum beat. Another excitonic quantum beat between bright exciton and dark exciton is observed for co-linear and co-circular polarization under the magnetic field tilted from the longitudinal direction and does not hold true in the observed quantum beat here.

The degree of linear polarization was analyzed by varying the magnetic field. For the analysis, degree of linear polarization was fitted by formula $\rho_t = \exp(-t/\tau)\sin(\omega t)$, where $\omega$ means the angular frequency of the oscillation and $\tau$ means its damping time constant. The fitted result is displayed in the inset of the upper panel of Fig. 3 for the magnetic field of $B = 6$ T. The solid curve is the fitted function with $2\pi/\omega = 24$ ps and $\tau = 20$ ps. The period of the beats decreased with increasing magnetic field. The energy splitting corresponding to inverse of the beat period is proportional to the magnetic field, as is shown in the upper panel of Fig. 3. The obtained splitting is much smaller than the laser linewidth (0.7 meV). From the absolute value of splitting and its linear dependence on the magnetic field, the beat is attributed to the quantum interference of bright excitons caused by the Zeeman splitting and the $g$ factor of bright excitons in the quantum dots is determined to be 0.51.

Further, the beat period was investigated in the Faraday configuration as a function of the angle between the magnetic field and the growth direction, as is shown at the lower panel of Fig. 3. If the electron-hole exchange energy is much smaller than the electron Zeeman splitting, the spin Hamiltonian of the electron-heavy-hole pair in the GaAs quantum structures under a magnetic field gives the simple expression for the en-
energy splitting of bright excitons, \( E_1 - E_2 = \Delta E_{12} \), represented by

\[
\Delta E_{12} = \mu_B B (g_{hi} \cos \theta + \sqrt{g_{c||}^2 \cos^2 \theta + g_{c\perp}^2 \sin^2 \theta}),(1)
\]

where \( g_{hi} \) (\( g_{c||} \)) is the \( g \) factor of the hole (electron) for the magnetic field parallel to growth direction, \( g_{c\perp} \) is the \( g \) factor of the electron for the magnetic field perpendicular to the growth direction, and \( \theta \) is the angle between the direction of magnetic field \( B \) and the crystal growth axis, under the assumption that \( g_{hi} \) is much smaller than \( g_{hi} \). The assumption is verified for the GaAs quantum well,\(^8\) where \( |g_{hi}| \) is measured to be 0.04 and is comparable to 1/50 of \( g_{hi} \). Because anisotropy is very large in GaAs quantum well, relation \( g_{hi} \ll g_{hi} \) is expected to hold for the strain-induced GaAs quantum dots where the lateral size is much larger than the height. As is shown in the following independent experiment in the Voigt configuration, electrons in the strain-induced GaAs quantum dots have isotropic \( g \) factor \( g_e \) and Eq. (1) is simplified to be \( \Delta E_{12} = \mu_B B (g_{hi} \cos \theta + g_e) \). By using the expression, the angle dependence of the energy splitting is well fitted, as is shown in the lower panel of Fig. 3. Here, we assumed that the lowest QD1 band mainly comes from heavy-hole exciton luminescence. The \( g \) factors for exciton, electron, and hole obtained are 0.51, 0.17, and 0.34, respectively.

V. ELECTRON QUANTUM BEATS IN THE TRANSVERSE MAGNETIC FIELD

Another type of quantum beats of the strain-induced GaAs dots coming from the electron Larmor precession was observed not only for the site-selective excitation of quantum dots but also for the excitation at the quantum well absorption in the Voigt configuration. The co- and cross-circular polarization components were selected by using a quarter-wave plate and were detected by the same experimental system that was used in the Faraday configuration. Under the high magnetic field above \( B = 5 \) \( \text{T} \), the periodic oscillation appeared in the time trace, as is shown by the bottom traces (QD 0°) for strain-induced GaAs quantum dots (well width = 3.9 nm) in Fig. 4. Here, the circularly polarized laser pulse excited the quantum dots and the LO-phonon sideband luminescence in the QD1 band was observed. The crystal-growth axis is perpendicular to the direction of the magnetic field and is parallel to the direction of the incident laser light. In the Voigt configuration, \( \theta' \) denotes the angle between the crystal-growth axis and the direction of the incident laser light. Because of the circular polarization selection rule for the observation of the oscillation in the Voigt configuration, the periodic oscillation was thought to be originated from the quantum beat of electron Larmor precession. The electron Zeeman splitting \( \Delta E_{e0°} \) is given by

\[
\Delta E_{e0°} = g_{c\perp} \mu_B B.
\]

We estimated \( g_{c\perp} \) to be 0.17 by equating the oscillation period to \( 2 \pi h / g_{c\perp} \mu_B B \). The positive sign of this value is determined by knowing that the well-width dependent \( g \) factor of electrons discussed in the following and that the electron \( g \) factor monotonously increases with the decrease of the volume ratio of GaAs in GaAs-AlGaAs quantum structures from the electron \( g \) factor of -0.44 in bulk GaAs.\(^{21–25}\) Value \( g_{c\perp} \) nicely agrees with the electron \( g \) factor obtained in the Faraday configuration. The periodic oscillations were also observed in the luminescence time trace of the quantum well, as is typically shown by the second top traces (QW 0°) for a GaAs quantum well (well width = 2.4 nm) in Fig. 4. The oscillation period for the quantum well was found to be shorter than that for the quantum dots in the same sample, although the quantum dots are formed in the quantum well by the strain. These results clearly show that \( g_{c\perp} = 0.17 \) for the quantum dots is smaller than \( g_{c\perp} = 0.225 \) for the quantum well in the same sample whose GaAs well width is 3.9 nm.

For the measurement of the electron \( g \) factor parallel to the sample growth direction (\( g_{hi} \)), the crystal was rotated by 45°. The luminescence oscillations observed in this geometry are shown by the top traces (QW 45°) and the second bottom traces (QD 45°) in Fig. 4. Compared with the oscillation period observed at \( \theta = 0° \), the oscillation period observed at \( \theta = 45° \) is longer for the quantum well. On the other hand, the oscillation period does not change between \( \theta = 45° \) and \( \theta = 0° \) for the quantum dots. When the crystal is rotated to \( \theta = 45° \), the electron Zeeman splitting is given by

\[
\Delta E_{e45°} = \mu_B B \left( g_{c||}^2 + g_{c\perp}^2 \right)^{1/2}. \tag{3}
\]
anisotropic $g$ factor of conduction electron comes from the different optical selection rule for heavy hole to electron and light hole to electron transitions. Anisotropic $g$ factors of conduction electron in GaAs quantum wells are described by:

$$g_{e\perp} - g_{e\parallel} = \frac{2p_{ce}^2}{m_0} \left[ \frac{1}{E_g + E_{e\perp} + E_{hh1}} - \frac{1}{E_g + E_{e\parallel} + E_{hh1}} \right],$$

(4)

where $p_{ce}$ is the transition matrix element between the valence band and the conduction band, $m_0$ is the bare electron mass, $E_g$ is the band-gap energy, and $E_{e\parallel}$, $E_{hh1}$, and $E_{lh1}$ are size-quantized energies of electron, heavy hole, and light hole, respectively. The calculated anisotropic $g$ factor for electrons in quantum wells nicely explains the observed anisotropy. The anisotropy is reduced if quantized energy of heavy hole and that of light hole approach each other in Eq. (4).

Strain effect on quantum wells can be analyzed by using the deformation potentials. The tensile strain formed by InP stressors on the Al$_{0.5}$Ga$_{0.5}$As surface causes hydrostatic deformation potential (2/3) $\delta E_{sh}$, to the conduction electron, hydrostatic deformation potential (1/3) $\delta E_{hy}$, minus shear deformation potential (1/2) $\delta E_{sh}$ to the heavy hole, and hydrostatic deformation potential (1/3) $\delta E_{hy}$ plus shear deformation potential (1/2) $\delta E_{sh}$ to the light hole. As a result, two-dimensional parabolic potential well is formed laterally in the well for electron, heavy hole, and light hole, and potential wells for light hole and heavy hole approach each other. Based on parameters listed in Ref. 26, ratio $\delta E_{hy}/\delta E_{sh}$ is calculated to be 2.18. From the observed red-shift of the energy of strain-induced quantum dots from the energy of the heavy-hole exciton peak of the quantum well, $\delta E_{hy} - (1/2) \delta E_{sh}$ is evaluated to be 43 meV. Therefore, $\delta E_{hy}$ and $\delta E_{sh}$ are obtained to be 55.8 meV and 25.6 meV, respectively. Although the energy splitting between heavy hole and light hole in the quantum well is 27 meV which is seen at the middle panel of Fig. 1, the reduction in the energy splitting in strain-induced quantum dots is estimated to be $\delta E_{sh} = 25.6$ meV. We believe the isotropic $g$ factor for the electron in the strain-induced GaAs quantum dots comes from the partial overlapping of the heavy-hole band and the light-hole band split once in the quantum well by the quantum confinement effect. Further study may be necessary to confirm the partial overlapping of the heavy-hole band and the light-hole band in the strain-induced GaAs quantum dots. In a spherical GaAs quantum dots, theory predicted the isotropic $g$ factor for the electron. This is another possible explanation of the isotropic $g$ factor for the electron in the strain-induced GaAs quantum dots. However, we cannot simply use this theory because the shape of strain-induced GaAs quantum dots is far from the sphere and because the vertical confinement is much stronger than the lateral confinement. We consider this is a possible explanation but strain effect is much plausible in the strain-induced GaAs quantum dots. This problem needs further study.

From the measured oscillation period at $\theta = 45^\circ$, $g_{e\parallel}$'s of quantum dots and a quantum well in the sample were derived by using Eq. (3) and the $g_{e\perp}$'s of electrons in quantum dots and a quantum well from the measured oscillation period at $\theta = 0^\circ$. The $g_{e\parallel}$'s of quantum dots and a quantum well in the sample were 0.17 and 0.09, respectively, for the sample whose well width is 3.9 nm. The $g_{e\parallel}$ is equal to the $g_{e\perp}$ in quantum dots, although $g_{e\parallel}$ and $g_{e\perp}$ of a quantum well were different from each other. This indicates the isotropic $g$ factor for electrons in dots, in spite of anisotropic $g$ factor for electrons in the well in the same sample. Isotropic $g$ factor for electrons in dots was also observed in another strain-induced GaAs quantum dots formed in the GaAs well 4.8-nm thick. In two kinds of strain-induced GaAs quantum dots 3.9-nm or 4.8-nm high, electron $g$ factors in dots are average values of $g_{e\parallel}$ and $g_{e\perp}$ of electron in the same quantum wells, although $g_{e\parallel}$ and $g_{e\perp}$ of electron in the quantum wells strongly depend on the well width, as is shown in Fig. 5. Electron $g$ factors of GaAs quantum wells increase from a negative value to positive ones with the decrease in the well width. The observed well-width dependence agrees well with the previous reports. The observed well-width dependence is well interpreted by the $k\cdot p$ perturbation theory.

Anisotropic $g$ factor of electrons in quantum wells has been investigated extensively. Its expression is given by the $k\cdot p$ perturbation theory and the physical origin for the

FIG. 5. Well-width dependence of the electron $g$ factor in strain-induced GaAs quantum dots and GaAs quantum wells. Electron $g$ factors $g_{\parallel}$ and $g_{\perp}$ were derived from quantum beats of electron Larmor precession for two angles between the magnetic-field and the crystal-growth direction, $\theta = 0^\circ$ and $\theta = 45^\circ$, in the Voigt configuration. Circles and triangles represent $g_{\parallel}$ and $g_{\perp}$ of electrons in quantum wells, respectively. Samples having two kinds of dots and three kinds of single quantum wells are used for the experiments. Diamonds represent $g$ factor of electrons in quantum dots. Solid lines show calculated $g_{\parallel}$ and $g_{\perp}$ of electrons in a single quantum well in the Kane model (Ref. 21).
VI. CONCLUSIONS

In summary, two kinds of luminescence quantum beats of the strain-induced GaAs quantum dots were observed in the magnetic field parallel and perpendicular to the growth direction. Quantum beat of bright excitons showing Zeeman splitting and quantum beat coming from electron Larmor precession were observed. The oscillation period in the quantum beat of bright excitons observed under the longitudinal magnetic field depends on the angle between the growth direction of the crystal and the magnetic field. Analysis based on the spin Hamiltonian for excitons explains the observed correction of the crystal and the magnetic field. Analysis based on the longitudinal magnetic-field-dependent electron Zeeman splitting, we found that the quantum beats coming from electron Larmor precession were observed under the transverse magnetic field. The isotropic electron g factor is observed in the dots in contrast to the anisotropic electron g factor for the corresponding quantum well and is ascribed to the strain-induced opposite energy shift of heavy- and light-hole bands. The quantum beats give us unique and fine information on not only spin structure but also energy structure of quantum dots.

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