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Spin relaxation mechanism of strain-induced GaAs quantum dots studied by time-resolved Kerr rotation

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We observed electron spin precession under magnetic field in single-layer quantum dots (QDs) by highly sensitive time-resolved Kerr rotation measurement. The spin lifetime is longer than that for the quantum well (QW). This is a result of the additional spatial confinement of electrons in QDs. Below 60 K, the spin lifetime is almost constant, and is 7 times shorter than the carrier lifetime. This suggests that the strong electron-hole exchange interaction dominates over the electron spin lifetime in QDs at low temperature.

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An electron spin in a semiconductor is one of the most hopeful candidates for a quantum bit in quantum information processing.[1] Especially in a semiconductor quantum dot, an electron spin may have a long spin coherence time due to suppression of spin relaxation by three-dimensional confinement of electrons.[2] Spin relaxation measurements by using various technique have been performed in bulk semiconductors, quantum wells, and quantum dots. In quantum dots, for example, the spin relaxation measurements by using optical orientation in steady-state and time-resolved photoluminescence have been reported.[3–5] These measurements are made, however, under nonresonant and quasiresonant excitations because the photoluminescence spectroscopy is difficult to be done under resonant excitation. However, the measurement under resonant excitation is important not only for obtaining direct information on spin relaxation but also for coherent processing of spins. In quantum wells and bulk semiconductors, many time-resolved magneto-optic measurements such as Kerr and Faraday effects have been performed in recent years.[6,7] The methods are suitable to observe electron spin dynamics under resonant excitation. In the single-layer quantum dots, however, such measurements have never been done because of the weak signal.[8,9] In this work, we constructed a new system for highly sensitive time-resolved Kerr rotation (TRKR) measurement. The angle resolution of the system was 5×10^{-6} deg., which is highest among those reported so far. As a result, we could observe the carrier spin dynamics in single-layer strain-induced GaAs quantum dots under resonant excitation.

The sample was fabricated by molecular beam epitaxy on a semi-insulating (001) GaAs substrate. An $Al_{0.3}Ga_{0.7}As$ buffer layer was deposited on the substrate. On the buffer layer, a quantum well layer 4-nm thick, an $Al_{0.3}Ga_{0.7}As$ barrier 14-nm thick, and a GaAs cap layer 2.3-nm thick were deposited in order. Stranski-Krastanov grown self-assembled InP stressor dots were fabricated on the surface of the cap layer. Since these stressor dots put the local strain into a 4 -nm quantum well layer, the strain-induced quantum dots were formed in the quantum well layer. The diameter, the hight and the areal density of the InP stressor dots are about 60 nm, 15 nm and 4×10^{9} cm^{-2}, respectively. By using the strain-induced quantum dots, we can compare the electron spin relaxation for the quantum dots and the quantum well directly because the quantum dots are formed in the quantum well.[10,11] The optical density of the single-layer quantum dots is about 3×10^{-3}, which is estimated by the absorption coefficient for the GaAs quantum well and the coverage ratio of 11%.[12] The optical density of GaAs quantum dots in our sample is 10^{-2} times smaller than that of about 0.5 for the chemically synthesized CdSe quantum dots studied in Ref. 8, which is a unique report for the quantum dots studied by the time-resolved Faraday rotation measurement.

For the TRKR measurements, the 80 fs pump and probe pulses were generated at a repetition rate of 80 MHz by a mode-locked Ti:sapphire laser. The circularly polarized pump pulse was made by passing through a Glan-laser prism and a quarter waveplate. An optical chopper modulated the pump beam. The probe pulse, temporally delayed with respect to the pump pulse by using an optional delay, was spectrally narrowed by passing through a filter unit made of a grating and a slit. The filter unit was also used to tune the photon energy of the probe beam. Then, the polarization of the probe beam was modulated by a photoelastic modulator. The pump and probe beams were focused by a lens on the sample kept in a superconducting magneto-optic cryostat in the transverse magnetic field geometry. The reflected probe beam from the sample passed through a Wollaston prism serving for an optical bridge and was detected by a balanced photodetector. The detected signal was amplified by a lock-in amplifier at twice the trigger frequency of the photoelastic modulator for the direct detection of the Kerr rotation angle.[13] The output signal of the first lock-in amplifier was amplified again by the second lock-in amplifier at the chopping frequency of the pump beam for the reduction in the noise caused by the scattered light. The excitation densities of the pump and probe beams were about 30 W/cm^2 and 3 W/cm^2, respectively. The pump pulse generated about one electron-hole pair per quantum dot.

Figure 1 shows the photoluminescence spectrum for the sample. The peaks originated from the GaAs quantum well, the strain-induced quantum dots, and the bulk GaAs substrate are clearly observed. The peaks originating from the excited state of the quantum dots are observed under high excitation density. The probe energy dependence of the TRKR signal without the magnetic field are also shown. The peaks of the TRKR signals are in agreement with those in the

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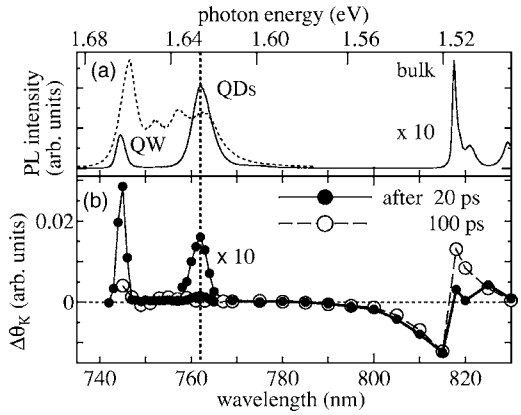

FIG. 1. (a) Photoluminescence spectrum at 10 K for excitation by the frequency-doubled output of a Ti:sapphire laser. The dotted line is under high excitation density. (b) Probe wavelength dependence of the TRKR signal without the transverse magnetic field at 10 K. Closed and open circles indicate the TRKR signals at the delay time of 20 and 100 ps, respectively.

photoluminescence spectrum. The ratio between the TRKR signal intensity for the quantum well and the quantum dots is about 20:1. As is estimated from the areal density and the diameter of the stressor quantum dots, the coverage ratio 11% of the InP quantum dots is obtained. It roughly agrees with the intensity ratio between the TRKR signals for the quantum well and the quantum dots.

The TRKR signals under the transverse magnetic field $B=6$ T at 10 K are shown in Fig. 2. Under the magnetic field, the spin of the photogenerated electrons precesses around the axis of the magnetic field. Thus, the TRKR signal is described by $\Delta\theta_K(\Delta t)\propto\exp(-\Delta t/\tau_s)\cos(\omega\Delta t)$, where τ_s is the spin lifetime, Δt is the temporal separation between the pump and probe pulses, and ω is the Larmor precession frequency. Because of the reflection geometry in the TRKR measurement, the oscillatory component for the bulk GaAs substrate was observed in our experiment as the probe energy is higher than the GaAs band gap energy. The absolute value of the g factor estimated from the observed oscillation frequency is about 0.43. This is consistent with the electron g factor of the GaAs. Therefore, we need to divide the experimental data into the individual signals by the formula described by $\Delta\theta_K(\Delta t)\propto\exp(-\Delta t/\tau_s)\cos(\omega\Delta t)+\exp(-\Delta t/\tau_{s\mathrm{GaAs}})\cos(\omega_{\mathrm{GaAs}}\Delta t)$, where $\tau_{s\mathrm{GaAs}}(\omega_{\mathrm{GaAs}})$ is the spin lifetime (Larmor precession frequency) of the bulk GaAs. In case of resonance of the quantum well [Figs. 2(d)–2(f)], the oscillatory signal for the bulk component is seen but is almost negligible because the signal is 10 times weaker than the dominant signal for the quantum well. In Fig. 2(f), the fitting errors are seen from 0 to 100 ps because of the weak signal component. On the other hand, in case of resonance of the quantum dots [Fig. 2(a)], a modulation in the amplitude of the oscillatory signal is observed due to the large contribution of the bulk component, which is 3 times larger than that for the quantum dots. The observed frequencies for the quantum dots, the quantum well and the bulk substrate are different from each other. In the latter part, we try to state the observed g factors in detail.

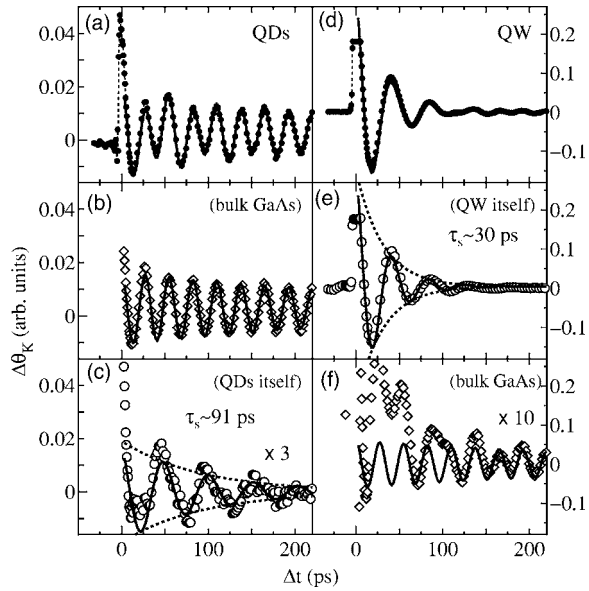

FIG. 2. Component analysis of TRKR signals taken under the magnetic field $B=6$ T at 10 K. The experimental data (closed circles) for the probe photon energy of 1.622 eV resonant to QDs (a) and those for the probe photon energy of 1.664 eV resonant to QW (d) and calculated signals (solid lines) by sum of two components are shown. The dominant components are shown in (b), (e). The minor component in resonance to the QDs is originated from the QDs (c). The dominant component (b) comes from the bulk GaAs substrate. The contribution of the bulk GaAs signal in resonant to the QW is shown in (f).

The obtained spin lifetime of about 90 ps for the quantum dots is longer than that of about 30 ps for the quantum well at 10 K (Fig. 2). The D'yakonov-Perel' (DP) mechanism due to the spin-orbit interaction dominates over electron spin relaxation in the undoped (100) GaAs quantum well and shortens the spin lifetime because the moving electron feels the effective magnetic field due to the lattice.[14] In fact, the spin lifetime reported is below 70 ps at low temperature.[15,16] Our experimental finding suggests that the spin relaxation due to the DP mechanism is suppressed as a result of the additional spatial confinement of electrons in quantum dots.

The spin precession at the frequency ω is interpreted by the quantum interference between the Zeeman levels separated by $\Delta E=\hbar\omega$ to each other.[19] The magnetic field dependence of the observed Zeeman splitting is shown in Fig. 3. These energies show a linear relationship described by $\hbar\omega=g\mu_B B$ with the magnetic field B, where g is the g factor of the electron. Thus, we can estimate the effective g factors by using the above relation. The observed g factors for the quantum well and the quantum dots are 0.26 and 0.22, respectively. The effective g factor is influenced by not only the structure but also the surrounding matrix into which the wave function is penetrating. The g factor for the quantum well is assigned to the perpendicular component $g_\perp$ because the magnetic field applied in perpendicular to the crystal growth axis. The inset of the figure shows the well width dependence of the g factors. The solid lines show the calculated electron g factor on the Kane model in an $Al_{0.3}Ga_{0.7}As/GaAs$ single quantum well,[18] and the filled

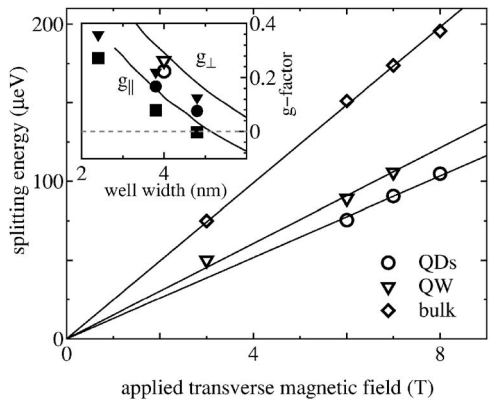

FIG. 3. The magnetic field dependence of the estimated Zeeman splitting energy. Open circles, triangles, and diamonds are splittings in the quantum dots, the quantum well, and the bulk GaAs substrate, respectively. The straight lines are the least square fittings. Inset: Quantum well width dependence of the electron g factor. The filled squares, triangles, and circles are, respectively, the g factor values for the parallel $g_\parallel$ and perpendicular $g_\perp$ components for the QW and that for the QDs taken from Ref. 17. Open circle and triangles are our experimental results for the QDs and the QW. The solid lines show the calculated result for the electron g factors for the single quantum well.[18]

marks are the experimental results by the time-resolved photoluminescence quantum beat measurements in the strain-induced GaAs quantum dots.[17] The present report gave slightly larger g factors than the previous work, because the stressor-induced strain in our sample is weaker than the sample used in Ref. 17. This is because the distance between the surface and the quantum well is longer, and because the diameter of the stressor dots is smaller in the sample used in this work. Thus, the quantum well layer feels smaller strain than that in the sample of the previous work, and approaches the *bare* quantum well. Therefore, the observed g factor is in good agreement with the calculated result because of the small strain. Because the wave function of the electron in the quantum dot penetrates into the AlGaAs barrier in the similar way as the GaAs quantum well, the effective g factor for the quantum dots is considered to be close to that for the quantum well. The consistent g factors shown in the inset of Fig. 3 confirm that we could observe the electron spin precession of the single-layer quantum dots under resonant excitation.

In the strain-induced GaAs quantum dots, the spin lifetime in the dots is lengthened by the suppression of the DP mechanism. Both electron-hole exchange interaction and hyperfine interaction were suggested as the possible electron spin relaxation processes in the quantum dots.[2,20,21] To identify the dominant spin relaxation mechanism for the strain-induced quantum dots, we performed the TRKR measurements at elevated temperatures. Figure 4 shows the temperature dependence of the measured spin lifetime and the carrier lifetime for the quantum dots under $B=6$ T. The latter was measured by the time-resolved photoluminescence spectroscopy with a streak camera under the quasiresonant excitation, where the excitation and detection energies were separated by the LO phonon energy of InP (42.5 meV). Above 80 K, the spin and carrier lifetimes showed the similar decrease. This means that the spin lifetime is limited by

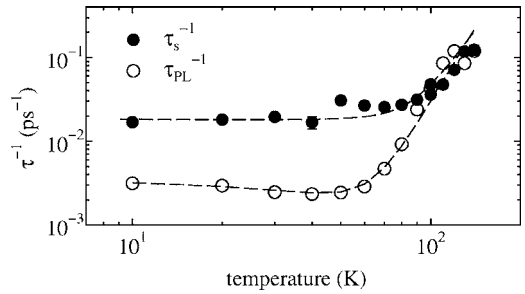

FIG. 4. Temperature dependence of the inverse of spin lifetime and carrier lifetime for the quantum dots under the magnetic field $B=6$ T. Open and closed circles show the carrier and spin lifetimes, respectively. Dashed lines are guide lines for the eyes.

the carrier lifetime above 80 K. The observed temperature independence of the spin lifetime at lower temperature indicates that the spin relaxation mechanism does not involve phonons. We evaluated the possibility that the g factor inhomogeneity might shorten the decay of the observed spin precession at lower temperature. However, the inhomogeneous shortening of the observed spin precession lifetime was found negligible. Because the spin lifetime is shorter than the carrier lifetime, some spin relaxation mechanisms are considered to dominate over the electron spin relaxation. In the neutral quantum dots, the electron-hole exchange interaction plays a dominant role in the electron spin relaxation.[22,23] The effective magnetic field due to the exchange interaction is considered to be larger than that due to the hyperfine interaction through the fluctuating nuclear spin orientations, although the spin relaxation due to the hyperfine interaction is important in the quantum dots.[21] It is because the exchange energy reached to an order of 100 μeV due to the three-dimensional confinement as is compared with the energy of the fluctuating hyperfine field of an order of 1 μeV.[24,25] Therefore, it is considered that the electron-hole exchange interaction is the predominant mechanism for the electron spin relaxation in the strain-induced quantum dots, which is referred to as the Bir-Aronov-Pikus mechanism.[26] The exchange interaction is considered to be independent of the temperature and reduce the spin lifetime with increasing electron-hole density because the overlap of the wave functions of the carriers increases. We cannot confirm, however, the excitation power dependence of the spin lifetime because of the weak power of the laser. On the other hand, for the quantum well, it is considered that the electron spin is relaxed by the DP mechanism mentioned above. The carrier lifetime limits the spin lifetime above 80 K as is the case for the quantum dots. The spin lifetime is longest at 80 K, but becomes shorter with decreasing temperature. The reason is not clear, but the similar results were observed under the magnetic field for the bulk semiconductors.[27,28]

In summary, we could observe the electron spin dynamics for the single-layer quantum dots under resonant excitation. The spin lifetime is longer than that for the quantum well at low temperature as a result of the additional spatial confinement of electrons. This suggests that the DP mechanism, which causes the electron spin relaxation in the quantum well, is suppressed in the quantum dots. The spin lifetime

at lower temperature is almost constant, and is shorter than the carrier lifetime. This indicates that the spin relaxation mechanism due to the electron-hole exchange interaction works on the electron spins in the quantum dots.

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