Spin dynamics of two-dimensional electrons in a quantum Hall system probed by time-resolved Kerr rotation spectroscopy

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Time-resolved Kerr rotation spectroscopy under the radio frequency field to depolarize dynamic nuclear polarization reveals the intrinsic spin-relaxation time (T_2^*) and g factor of two-dimensional electrons in a quantum Hall system. Out-of-plane magnetic field increases the spin coherence drastically through the Landau level quantization. T_2^* is enhanced strongly around odd filling factors where a quantum Hall ferromagnet is formed. Collapse of spin coherence and appearance of an anomalous Kerr signal observed around $\nu=1$ are discussed in the relation to the formation of Skyrmions.

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Recent interest in spintronic device and quantum information processing has spurred the research on the dynamics of electron spins in semiconductor nanostructures.¹ It is vital to understand the intriguing spin dynamics in strongly correlated electron systems. Quantum Hall system presents a variety of spin related phenomena and provides an excellent field to study the fundamental physics of electron-electron and electron-nuclear spin interactions. So far, many studies have been devoted to the investigation of spin effects in quantum Hall systems. Nuclear magnetic resonance (NMR), optical, and magnetotransport experiments have revealed various spin related excitation.²⁻⁶ It is well accepted that the elementary spin excitations in the quantum Hall ferromagnet (QHF) at $\nu = 1$ are the "spin exciton" (spin wave) with the dispersion relation derived by Bychkov et al.⁷ and Kallin et al.8 and spin-textured quasiparticle called Skyrmion proposed by Sondhi *et al.*⁹ in the case of low Zeeman energy.

Concerning the spin dynamics in a quantum Hall system, an increase in spin-relaxation time due to Landau level (LL) quantization was theoretically studied by Burkov et al.¹⁰ Relaxation of the spin exciton in the QHF (ν =1) was discussed by Frenkel¹¹ and Apel et al.¹² based on an inelastic phonon scattering,¹³ and by Dickmann¹⁴ based on a smooth disorder potential scattering. However, few experimental works have been reported for the spin dynamics in the quantum Hall regime. Recently, time-resolved Kerr rotation (TRKR) measurements were employed to investigate spin dynamics of two-dimensional (2D) electrons, but most of them were carried out in Voigt geometry¹⁵⁻¹⁸ where magnetic field is applied parallel to the 2D plane. Effects of LL quantization on the spin dynamics were examined in an In_{0.2}Ga_{0.8}As/GaAs quantum well (QW) (Ref. 16), where the mobility is low and the quantum Hall effect is not well developed.

In this Rapid Communication, we apply TRKR spectroscopy for the first time to high mobility 2D electrons in the quantum Hall regime in order to investigate how the spinrelaxation time depends on the filling factor and how is the dynamics of spin excitation in the QHF. We use a single 17-nm GaAs/Al_{0.3}Ga_{0.7}As QW with one-side δ -doped barrier layer grown on a (100) GaAs substrate.¹⁹ The 2D electrons density decreases appreciably under photoexcitation as reported in the Rapid Communication.²⁰ The change occurs in the millisecond time scale, so 2D electron density is constant under the light modulation of 50 kHz in the experiment. The density and mobility under the experimental condition are monitored by the magnetoconductivity measurement and estimated to be 1.6×10^{11} cm⁻² and 1×10^{6} cm²/Vs, respectively. A mode-locked Ti:sapphire laser with a pulse width of 120 fs and a repetition rate of 76 MHz is tuned to the absorption edge of the QW (1.567 eV). Kerr rotation is measured with the sensitivity of 1 μ rad by means of a balanced homodyne detection and a helicity modulation ($\sigma^+ - \sigma^-$) of a circularly polarized pump beam. A folded optical delay unit enables TRKR measurement at a delay time in the range of -1.3 to 9.3 ns.

TRKR measurements are carried out in a cryostat with a Helmholtz-coil superconducting magnet. The sample is set in three geometries: Voigt geometry ($\alpha = 0^{\circ}$), 53° tilted-field geometry ($\alpha = 53^{\circ}$), and 65° tilted-field geometry ($\alpha = 65^{\circ}$), where α is defined as the angle between **B** and the sample plane. Figure 1(a) shows the measurement geometry in the tilted field (α =53°). **B** is applied in the [110]x-[001]z plane and the incident direction of the pump beam is perpendicular to **B** in the same x-z plane. Probe beam is tilted from pump beam by 2.5° (in the same plane) and is linearly polarized in the incident plane. Low-power pump (0.4 mW) and probe (0.2 mW) beams are focused on the sample with spot sizes of 400 and 300 μ m, respectively, to suppress the optical disturbance and the pump inhomogeneity. Each pump pulse is considered to generate $\sim 4 \times 10^8$ cm⁻² electron-hole pairs, which corresponds to $\sim 0.3\%$ of the electron density. Since the photoexcited holes loose the spin coherence very rapidly and then recombine with excess electrons in the OW on a subnanosecond time scale,^{21,22} only the photoexcited electron spin S is transferred to the 2D electron gas.

In the tilted-field geometry, the *g* factor of electron spin in a GaAs quantum well is anisotropic and substantially different from the bulk value -0.44 because of the quantum confinement.¹⁵ In the present experiment since **B** is applied in (*x*-*z*) plane, the effective *g* factor is given by



FIG. 1. Schematic illustration of (a) the tilted-field geometry for TRKR measurement and (b) the split-coil geometry for the rf field to depolarize DNP. (c) TRKR signals and (d) their Fourier-transform spectra obtained with and without rf field in the 53° geometry at 1.5 K and 1 T.

$$|g| = \sqrt{g_x^2 \cos^2 \alpha + g_z^2 \sin^2 \alpha}, \qquad (1)$$

where g_x and g_z are the respective components of the $\hat{\mathbf{g}}$ tensor. Photoinduced spins in the 2D electron gas precess around $\boldsymbol{\Omega}$ in a tilted-field geometry. The precession vector $\boldsymbol{\Omega}$ is defined by the equation, $\boldsymbol{\Omega} = \hat{\mathbf{g}} \mathbf{B} \mu_B / \hbar$, where $\hat{\mathbf{g}}$ is a tensor of *g* factor. $\boldsymbol{\Omega}$ is tilted away from **B** in the (*x*-*z*) plane and the angle γ from *x* [110] axis is given by the equation, $\tan \gamma = g_z/g_x \tan \alpha$ in an anisotropic spin system. Kerr rotation angle θ_K as a function of the time delay Δt can be expressed by

$$\theta_K(\Delta t) \propto A_1 e^{-\Delta t/T_1} + A_2 e^{-\Delta t/T_2^*} \cos \Omega_L \Delta t, \qquad (2)$$

where $T_1(T_2^*)$ is the longitudinal (transverse) spin-relaxation time and Ω_L is the Larmor frequency. The coefficient $A_1(A_2)$ is proportional to nonprecessing component S_{\parallel} (precessing component S_{\perp}) of **S** in polar Kerr effect scheme.

In the quantum Hall regime, dynamic nuclear-spin polarization (DNP) happens under the optical pumping and strongly modifies the electron-spin dynamics. To eliminate the unintentional DNP, radio frequency (rf) magnetic field is applied perpendicular to **B** by a split coil [Fig. 1(b)]. The rf field is swept by 40 kHz every 0.75 s across the nuclear resonance frequencies for the constituent elements: ⁷⁵As, ⁷¹Ga, and ⁶⁹Ga. Figure 1(c) shows TRKR signal at B=1 T in the 53° tilted-field geometry. Top and bottom traces are obtained without and with rf field, respectively. Fouriertransform spectra for these traces are shown in Fig. 1(d). The change in the precession frequency and decay are induced by the effective nuclear field and the dephasing due to the inhomogeneous nuclear field, respectively.

The spin-flip energy is obtained from the peak of the Fourier-transform spectrum. The spin-relaxation time (T_2^*) is obtained from the amplitude decay of the oscillating Kerr signal. Figure 2(a) shows the spin-flip energies in the 53° tilted-field geometry at 1.5 K. Crosses and open circles are

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FIG. 2. (a) Magnetic-field dependence of the spin-flip energy measured at 1.5 K in the 53° geometry with rf field (crosses) and without rf field (open circles). (b) g factor converted from the spin-flip energy under rf field.

for the data obtained with and without applying rf field, respectively. Even under the $\sigma^+ - \sigma^-$ helicity modulation, the spin-flip energy without rf field is considerably larger than value under rf field by DNP over a whole experimental range. The bare g factor and the intrinsic spin relaxation of 2D electrons are obtained under nuclear depolarizing rf field in the saturation regime.

Magnetic-field dependence of the *g* factor under the rf field is plotted in Fig. 2(b) for the 53° tilted-field geometry. The *g* factor strongly depends on the position of the Fermi energy and jumps at even filling factors, reflecting the band nonparabolicity in the QW (Ref. 23). The data fit of the expression, $|g(B,N)| = g_0 - c(N+1/2)$ with the parameters; $g_0=0.235$ and c=0.0055, where N is the Landau index. From α dependence of g_0 and the helicity dependence of the Kerr signal the *g* tensor components are obtained to be $|g_x| = 0.297$ and $|g_z| = 0.150$ based on the Eq. (1). The small and anisotropic *g* factor may be ascribed to the asymmetric wave-function profile in the one-side doped QW.

A circularly polarized pump pulse generates a coherent spin wave with a wave number $q = (\Omega_L/c) \sin \alpha$ in the tiltedfield geometry, where c is the velocity of light. Since this value is negligibly small compared to the inverse of magnetic length l_0 , TRKR measurement in the tilted-field geometry detects the spin-flip energy of bare electrons $(q \approx 0)$ as electrically detected electron spin resonance (ED-ESR) (Ref. 23). In contrast to ED-ESR, TRKR measurement allows us to measure the spin-flip energy (g factor) in the magneticfield region where a quantum Hall plateau is observed. The transition between the g factors corresponding to different LL indices occurs very sharply at even filling factors. This contrasts with the result in the InGaAs QW by Sih et al.¹⁶ In a high mobility 2D electron system, the spin precession is totally supported by the electrons in the LL where the Fermi level locates. The oscillation of g factor disappears for lower field at elevated temperature without a significant change in the relaxation time. This behavior is well understood as a motional narrowing effect.

Figure 3(a) shows the magnetic-field dependence of T_2^*



FIG. 3. (a) Spin-relaxation times as a function of the magnetic field measured in the 53° geometry at 1.5 K (closed circles) and \sim 5 K (crosses). Open circles present T_2^* around ν =1 measured in the 65° geometry at 1.5 K. (b) A linearly polarized pump light induced Kerr rotation signal under the pump beam intensity of 0.4 mW (solid line) and 0.2 mW (dotted line).

measured in the 53° geometry at 1.5 K (closed circles) and ~5 K (crosses). Some additional data around 6 T observed for the 65° geometry at 1.5 K are plotted by open circles. T_2^* increases quadratically with increasing magnetic field below 1 T and then shows sharp peaks at the magnetic fields corresponding to ν =7 and 5. Around ν =3 and 1, T_2^* shows a dip at the center of the broad peak. There is a wide deep valley around ν =2. The peak/dip structure gradually disappears with increasing temperature.

Spin-relaxation time, T_2^* in the Voigt geometry is about 100 ps and scarcely depends on magnetic field. The spinrelaxation mechanism of 2D electrons without perpendicular magnetic field is well studied^{17,18} and the dominant decoherence mechanism in high mobility 2D electrons is believed to be the D'yakonov, Perel', and Kachorovskii (DPK) mechanism.¹⁷ Therefore, the increase in the spin coherence in the tilted-field geometry is ascribed to the suppression of the DPK mechanism by the LL quantization. The quadratic increase in T_2^* for lower field below 1 T agrees with the theoretical prediction.¹⁰ However, for higher field above 1 T, the spin-relaxation time undergoes sharp peaks at the magnetic fields corresponding to higher odd filling factors $(\nu=7,5)$ rather than the predicted Shubnikov-de Haas-like oscillation.¹⁰ Furthermore, on the contrary to the theoretical prediction, dips appear at the even filling factors, where the spin-relaxation time is expected to be long due to the long electron-scattering time.

The enhancement of the spin-relaxation time at odd filling factor was theoretically predicted.¹²⁻¹⁴ Apel, Bychkov, and Khaetskii^{12,13} discussed the spin relaxation of the Goldstone mode in a QHF by the phonon scattering mechanism and estimated the relaxation time at ν =1 to be $1 \sim 10 \ \mu$ s, which is 3 orders of magnitude larger than the observed values. Dickmann¹⁴ discussed the spin relaxation at ν =1 by a smooth disorder potential and the decay time was estimated to be $10 \sim 100$ ns, which is close to our results. Though he did not discuss the filling factor dependence of the relaxation time explicitly, the spin-relaxation process is analyzed as "coalescence" of condensed spin excitons (q=0), which



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FIG. 4. Typical TRKR signals for several values of the filling factor.

strongly depends on the spin exciton dispersion. The relaxation rate expressed by Eq. (10) in Ref. 14 is proportional to the square of the spin exciton mass. So, we believe the observed peak at odd filling factor corresponds to a Goldstone mode in the QHF. The origin of center dip of T_2^* at $\nu = 1$ and 3 is discussed later.

Figure 3(b) shows a Kerr rotation signal for the intensity modulation of a linearly polarized pump light at a time delay of 13 ns in the 53° geometry. Sharp peaks are ascribed to the photoinduced spin depolarization of ferromagnetic spin states at odd filling factors. The peaks at 1.19, 1.66, and 2.77 T exactly coincide with the fields for $\nu = 7, 5, \text{ and } 3$, respectively, estimated by using the carrier density of 1.6 $\times 10^{11}$ cm⁻² obtained by the magnetotransport measurement. However, the peak at 7.4 T is appreciably smaller than the field for $\nu = 1$ (8.3 T) calculated for the same carrier density. This deviation increases with the increasing pump intensity as seen in Fig. 3(b). Judging from the systematic shift of photoinduced Kerr rotation peak, the deviation is ascribed to the decrease in the electron density in the high magnetic field. Therefore, the collapse of spin coherence (dip of T_2^*) is considered to occur at $\nu = 1$. The decrease in electron density in the high magnetic field ($\nu < 2$) is unusual, but it can be explained as follows: The absorption edge (Fermi level) undergo a large jump to low-energy side at $(\nu=2)$ (Ref. 6) and the consequent absorption change in the laser beam [full width at half maximum (FWHM): $\sim 15 \text{ meV}$] may introduce additional decrease in the quasiequilibrium 2D electron density under photoillumination.²⁰

Figure 4 shows the filling factor dependence of TRKR signals measured at 1.5 K in the 53° tilted-field geometry. TRKR signal for lower field below 2 T are well approximated by Eq. (2) with the relation $T_2^*=2T_1$, for the homogeneous classical spin system. The profile of the TRKR signal strongly depends on the filling factor in the quantum Hall regime. The amplitude and the decay time of the oscillating Kerr component decrease when the filling factor approaches even integers (e.g., ν =2), while the nonoscillating Kerr com-

ponent keeps the amplitude and decay time. In particular, the TRKR signal changes drastically around $\nu = 1$. A large negative nonoscillating Kerr signal appears in the vicinity of $\nu = 1$ in addition to the normal oscillating Kerr signal. The amplitude of oscillating signal decays rather fast at $\nu = 1$.

The origin of the unusual negative Kerr signal around $\nu = 1$ was examined by the TRKR measurements by the intensity modulation of the σ^+ (σ^-) and linearly polarized pump light. The unusual nonoscillating Kerr component has the same sign for σ^+ and σ^- polarized pump lights, while the oscillating component changes the sign. The amplitude of the unusual Kerr signal obtained by the light intensity modulation is several times larger than that for the helicity modulation. This behavior is well explained by the formation of large size Skyrmions and anti-Skyrmions under the photoexcitation. The injection of an electron-hole pair generates a pair of Skyrmion and anti-Skyrmion in the QHF (ν =1), which reduces the spin polarization. This is the scenario for the appearance of the unusual negative Kerr signal. This scenario is confirmed by the sharp peak at $\nu = 1$ or 3 of the photoinduced spin depolarization shown in Fig. 3(b). In the above model, the first decrease in the Kerr signal corresponds to the formation process of Skyrmion and anti-Skyrmion and the recovery of the Kerr signal corresponds to the annihilation process by the mutual coalescence. The characteristic times are estimated to be 1 and 10 ns, respectively.

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The collapse of the spin-relaxation time at $\nu = 1$ in Fig. 3(a) strongly correlates with the appearance of the unusual Kerr signal. Furthermore, this unusual behavior is observed in a wider filling factor region around $\nu = 1$ for the 65° tilted-field geometry where the lower Zeeman energy favors the formation of Skyrmions.⁹ Therefore, it is natural to consider that the collapse of the spin coherence is induced by the formation of Skyrmions and anti-Skyrmions under the photoexcitation. This effect might be related to the softening of the spin wave near $\nu = 1$ due to the magnetic instability in the Skyrmion system, recently observed by Gallais *et al.*²⁴

The dip of T_2^* and sharp peak of the photoinduced spin depolarization at $\nu=3$ shown in Fig. 3(b) would be ascribed to a similar effect. The formation of Skyrmion at higher odd filling factor will be reported somewhere.

TRKR spectroscopy has revealed intrinsic spin relaxation in the quantum Hall system. This technique can be a versatile tool to investigate dynamics of spin related excitations in the quantum Hall regime.

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