Optically Induced Nuclear Spin Polarization in the Quantum Hall Regime: The Effect of Electron Spin Polarization through Exciton and Trion Excitations

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We study nuclear spin polarization in the quantum Hall regime through the optically pumped electron spin polarization in the lowest Landau level. The nuclear spin polarization is measured as a nuclear magnetic field B_N by means of the sensitive resistive detection. We find the dependence of B_N on the filling factor nonmonotonic. The comprehensive measurements of B_N with the help of the circularly polarized photoluminescence measurements indicate the participation of the photoexcited complexes, i.e., the exciton and trion (charged exciton), in nuclear spin polarization. On the basis of a novel estimation method of the equilibrium electron spin polarization, we analyze the experimental data and conclude that the filling factor dependence of B_N is understood by the effect of electron spin polarization through excitons and trions.

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The coupling between electron and nuclear spins through the contact hyperfine interaction realizes the dynamic nuclear spin polarization and the detection of a small ensemble of nuclear spins. This allows us to perform nuclear magnetic resonance (NMR) in a microscopic region through electrical or optical manipulation of electron spins [1]. This new type of NMR technique is a powerful tool to probe electronic properties and also has a potential to implement quantum information processing by using nuclear spins as qubits. Indeed, its intriguing electronic properties have been revealed in the quantum Hall system [2–6], and multiple quantum coherences of nuclear spins have been controlled in a nanometer-scale region [7]. In these experiments, electrical pumping and resistive detection of nuclear spins play an important role, while optical pumping of nuclear spins has also been achieved [8–12]. The condition for the electrical pumping is restricted to a special electronic state such as spin phase transition in the 2/3 fractional quantum Hall state and this limits the application of the NMR technique. However, the optical pumping does not bring about this restriction. When one combines electrical and optical means, the fascinating electronic states in the quantum Hall system will be widely investigated and rich quantum information processing can be demonstrated.

The research on the optical pumping conditions for the dynamical nuclear polarization has been performed in the quantum Hall regime [8,13]. However, how the quantum Hall electronic states affect its polarization has not been fully investigated. In this Letter, we study the dependence of the optically induced nuclear spin polarization on the electric state in the quantum Hall regime, i.e., the Landau level filling factor ν. We find a correlation between the nuclear polarization and the photoluminescence (PL). Our experimental data are analyzed by use of the estimation of electron spin polarization that we constructed. We understand the ν dependence of the optical nuclear polarization as the effect of the electron spin polarization through excitons and trions in the quantum Hall regime.

Experiments were carried out on a single 18-nm $GaAs/Al_{0.33}Ga_{0.67}As$ quantum well with single-side doping, which was processed to a 100-μm-long and 30-μm-wide Hall bar. The electron density n_s of the two-dimensional electron system can be tuned by applying a voltage to the n-type GaAs substrate (back gate). The sample was cooled in a cryogen free ^{3}He refrigerator down to 0.3 K and pumped by a mode-locked Ti:sapphire laser (pulse width: ~2 ps, pulse repetition: 76 MHz). The electron mobility is $185\ m^2/(V\,s)$ for $n_s = 1.2 \times 10^{15}\ m^{-2}$. A laser beam (diameter:230 μm) irradiated the whole Hall bar structure through an optical window on the bottom of the cryostat. The propagation direction of the laser beam was parallel to the external magnetic field $B = 7.15$ T, which was perpendicular to the quantum well. We can vary $\nu = hn_s/(eB)$ using the back gate, where h is the Planck constant and e is the elementary charge.

The optical pumping was performed as follows. First, the nuclear spin polarization was fully destroyed by setting the electronic state to the Skyrmion region [2]. Second, right or left circularly polarized light (σ^+ or σ^-) illuminated the sample, where the electronic state was set to ν during illumination. The pumping time was 250 s, which was long enough to saturate the optical nuclear polarization. The pumping photon energy E_{laser} and the average power density P are specified below. The laser illumination increased the temperature of the sample holder up to

0.4 K and also disguised the sample resistance. Third, after optical pumping, ν was set to 1 for 70 s so that the resistance returned to the value before illumination, where the relaxation of nuclear polarization at $\nu = 1$ is the smallest within the available ν. This relaxation time was over 1.6×10^3 s. Therefore, the nuclear spin polarization generated by optical pumping was not destroyed during the waiting time at $\nu = 1$.

The optically induced nuclear polarization was measured by the resistive detection method using a peak shift of the spin phase transition at $\nu = 2/3$ [9,13]. This method is a highly sensitive detection of nuclear polarization. Here, we recorded the nuclear magnetic field B_N induced by the nuclear polarization of the relevant three nuclides (^{69}Ga, ^{71}Ga, and ^{75}As) at the electric-current-flowing region. We used a standard low-frequency (83 Hz) and low-current (30 nA) lock-in technique to measure the resistance. Thus, the resistive detection method we used (see our previous paper [13] for the experimental details) enables us to probe only a small ensemble of nuclear spins interacting with the two-dimensional electron system.

Figure 1 shows the ν dependence of optically induced B_N for $\sigma^+(\sigma^-)$ excitation with $E_{\rm laser} = 1.5328(1.5321)$ eV. Here, $\sigma^\pm$ excitation is associated with the interband transition from a heavy hole band with angular momentum $J_z = \mp 3/2$ to the lowest electron Landau level with spin $S_z = \mp 1/2$. B_N induced by σ^+ and σ^- excitations show the opposite direction because the conduction electrons with down and up spins are created by σ^+ and σ^- excitations, respectively [13]. When ν increases from the lower side, the magnitude of B_N decreases for both excitations. In the ν range from 0.4 to 0.9, relatively small values of B_N are observed for σ^+ and the apparent nuclear spin polarization is not observed for σ^-. Around $\nu = 1$, nuclear spins are not polarized for either excitation. The stronger excitation power exhibits the larger magnitude of B_N, which is explained by the increased pumping rate. However, the nonmonotonic behavior of the ν dependence remains unchanged. There are three regions for optical nuclear spin polarization in the lowest Landau level: (I) $\nu < 0.4$, (II) $0.4 < \nu < 0.9$ and (III) $\nu > 0.9$.

To investigate these behaviors, we measured ν dependence of optically induced B_N by changing $E_{\rm laser}$ with $P = 1.2$ W/cm^2. The optical pumping rate is expected to depend on the photon absorption rate, and ν dependence with constant $E_{\rm laser}$ should be modified when the absorption spectrum is varied by changing ν. The optical transitions in the quantum Hall system (both absorption and luminescence) are determined by the strong Coulomb interaction between the valence hole and the surrounding electrons, resulting in the existence of bound electron-hole complexes, e.g., neutral and charged (trions) excitons in the lowest Landau level [14]. The configuration of our sample is not suitable for absorption measurements. Although the peak positions of the absorption and luminescence are not completely coincident, the luminescence peak can be used as the indicator of the absorption peak due to the relatively small energy difference [15]. Therefore, we also measured the circularly polarized PL with a spectral resolution better than 0.2 meV, where we used the linearly polarized light [16] with the excitation energy of 1.58 eV and the power density of 1.2 W/cm^2.

Figure 2(a) [(b)] shows the color map of B_N for σ^+ (σ^-) excitation. The transverse and longitudinal axes indicate ν

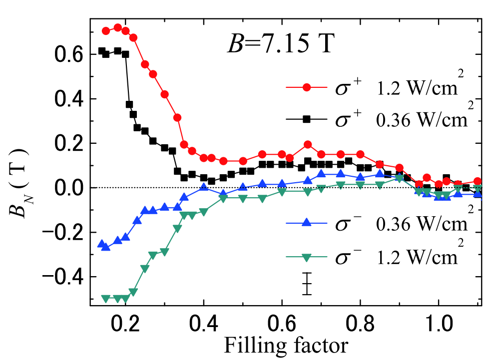

FIG. 1 (color online). The ν dependence of optical nuclear spin polarization at $B = 7.15T$. The error bar shows typical errors in B_N.

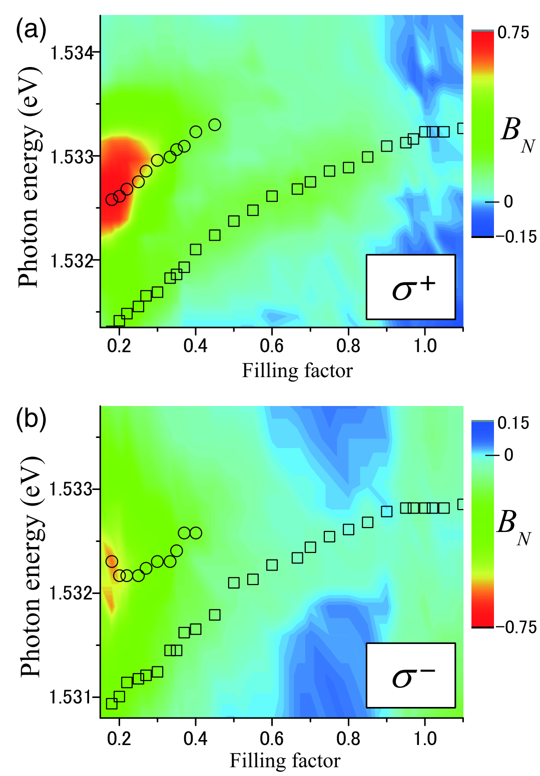

FIG. 2 (color online). The 2D color map of B_N for (a) σ^+ and (b) σ^- excitations. The scale of the color bars is linear. The directions of the bars for (a) and (b) are reversed for clarity. The circles (squares) show the photoluminescence peak positions of the triplet (singlet) trion.

and photon energy, respectively. The σ^+ (σ^-) PL peak positions of triplet (circles) and singlet (squares) trions are overlaid in (a) [(b)], where the peaks were assigned by the B and n_s developments [17] and the triplet and singlet mean the spin alignments of two electrons in the trion. The increase of the peak energies as ν increases is understood by the quantum confined Stark effect because we controlled ν using the gate voltage. Taking into consideration the spectral width of the pumping laser (full width at half maximum ~0.6 meV), we find that the nuclear polarization basically occurs at the PL peak positions. The nuclear polarization at the triplet trion peak is larger than that at the singlet trion peak. Thus, in terms of the PL peak as the indicator of the absorption peak (the details will be discussed in the next paragraph.), we observe the correlation between the nuclear polarization and the photoexcited complex absorption. This accounts for the difference between regions (I) and (II) in Fig. 1.

We here consider what information for the photon absorption is elicited from the observed PL since the absorption is important to polarize nuclear spins as mentioned above. Although we assigned the upper PL peak to the triplet trion, the neutral exciton peak is expected to be merged into (or have slightly higher energy than) the triplet trion peak [15,17] under our experimental conditions. The neutral exciton has greater oscillator strength than the triplet trion in the absorption measurement [15] and in the numerical calculations in high B field [18]. Therefore, we can attribute the nuclear polarization at the triplet PL peak to the absorption of the neutral exciton [19]. In contrast, we consider the nuclear polarization at the singlet PL peak as the consequence of the absorption of the singlet trion [20]. Indeed, in the absorption experiment (under conditions similar to ours) performed by Groshaus *et al.* [21], two peaks were assigned to the neutral exciton X and the singlet trion T [22].

Next, we discuss how the optically excited complexes affect the nuclear spin polarization. Our experimental results indicate that the photoexcitation of X leads to higher nuclear polarization than that of T. The photon absorption rate is proportional to the number of the injected electron spins, which subsequently polarizes the nuclear spin. X in high B field or at low n_s is expected to have larger absorption than T [15,18]. This can explain our results simply. However, the absorption measurement does not always show such behavior under the experimental conditions similar to ours [21]. To polarize nuclear spins, primarily, the electron spin polarization under optical pumping $\langle S_z\rangle$ is more crucial than the number of injected electron spins. The effective nuclear magnetic field after long pumping time is given by $B_N = -A(\langle S_z\rangle - \langle S_z\rangle_{\rm eq})$, where $A(>0)$ is a constant and $\langle S_z\rangle_{\rm eq}$ is the equilibrium electron spin polarization [13,23]. This fact and the experimental results indicate that the photoexcitation of X generates higher $\langle S_z\rangle$ than that of T in the quantum Hall regime. Indeed, this can be expected from the study of optical spin pumping in the II-VI quantum well [24]. Moreover, there is a possibility that X directly and indirectly polarizes the nuclear spins, because the electron in X has s-type symmetry, which considerably contributes to the contact hyperfine interaction, and because X forms T by capturing the resident electron.

We also take into consideration $\langle S_z\rangle_{\rm eq}$ to understand the ν dependence of B_N. To know the electron spin polarization P_e, the optical dichroism calculated from the trion absorption is available [21]. Although we cannot measure the absorption of our sample, the PL polarization P_L has a contribution of P_e and has been utilized to extract the P_e characteristics [25,26]. We develop the P_e estimation from P_L. The $\sigma^+(\sigma^-)$ PL intensity $I_{\sigma^+(\sigma^-)}$ is proportional to the number of photoexcited particles multiplied by its oscillator strength. We define P_L as $(I_{\sigma^+} - I_{\sigma^-})/(I_{\sigma^+} + I_{\sigma^-})$ and here consider this formulation for T. Since the photocreated electron needs to pair with an opposite spin, the oscillator strength of each T can be modeled as proportional to the number of unpaired electrons with opposite spin [21]. Consequently, the calculation of P_L for T gives $P_e = (P_L - P_h)/(1 - P_L P_h)$ for $\nu \le 1$ and $P_e = [(2-\nu)/\nu](P_L - P_h)/(1 - P_L P_h)$ for $\nu > 1$, where P_h is the hole polarization due to the singlet nature of T [27]. While P_h increases with B [29], our experiments were performed under constant B, and P_h should be constant. Figures 3(a) and 3(b), respectively, show the measured P_L for T and the P_e calculated with the P_h values of −0.9, −0.7, and −0.5. Since the optical pumping was performed with the strong illumination, the P_e obtained here (under

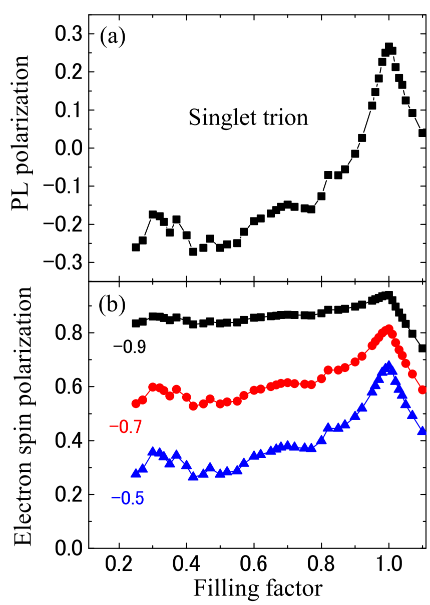

FIG. 3 (color online). (a) The PL polarization obtained from the singlet-trion peak intensities with the 1.2 W/cm² linearly polarized excitation. (b) The electron spin polarizations estimated from (a) with $P_h = -0.9$, −0.7, and −0.5.

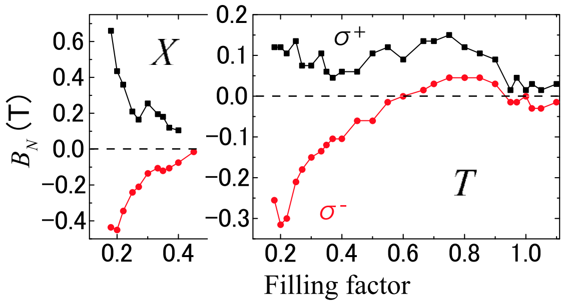

FIG. 4 (color online). The ν dependence of B_N along the PL peak positions in Fig. 2.

the strong photoexcitation) is treated as $\langle S_z\rangle_{eq}$ [30]. Thus, we obtain the trend of $\langle S_z\rangle_{eq}$ although the correct values are uncertain due to the lack of P_h information.

To consider how $\langle S_z\rangle_{eq}$ affects B_N, we should exclude the pronounced difference of $\langle S_z\rangle$ between X and T resonant excitations. To this end, we extract the values of B_N at the PL peak positions from Fig. 2. The data are shown in Fig. 4 [31]. The slight B_N increase in the ν range from 0.4 to 0.8 can be understood from the obtained trend of $\langle S_z\rangle_{eq}$, which is an almost monotonic increase. In $\nu < 0.4$, B_N does not obey the trend of $\langle S_z\rangle_{eq}$. The lower the n_s, the higher the nuclear polarization obtained. This is attributed to the increase of $\langle S_z\rangle$ for both X and T excitations. Increasing the number of the injected electrons relative to n_s can enhance $\langle S_z\rangle$. Although a theoretical study on the electron spin pumping in the quantum Hall regime is required for a complete explanation, it should be noted that diminishing the doping enhances $\langle S_z\rangle$ in B parallel to the well [32].

Finally, we consider the optical nuclear polarization in region (III). In this region, the Skyrmion exists under our experimental conditions. The low-frequency spin fluctuations associated with the Skyrmion destroy the nuclear polarization. We measured the nuclear spin relaxation by changing the waiting time at temporal ν after optical pumping. The time decay of B_N is fitted by the exponential function. The observed nuclear spin relaxation rates $1/T_{1N}$ are displayed in Fig. 5 [33]. The relatively small values of $1/T_{1N}$ at $\nu = 2/3$ and 1 are due to the energy gap of the quantum Hall state. We clearly observe the strong nuclear spin relaxation around $\nu = 1$ [34]. This diminishes the nuclear spin polarization. At $\nu = 1$, the up spin sublevel of the lowest Landau level is expected to be fully occupied. Therefore, the up spin cannot be excited and the photo-excited down spin cannot relax to the up spin. This can inhibit the nuclear spin polarization.

In conclusion, we studied nuclear spin polarization in the quantum Hall regime through the optically pumped electron spin polarization in the lowest Landau level. We found the obvious ν dependence of the optically induced B_N. To understand this behavior, we constructed a novel estimation method of $\langle S_z\rangle_{eq}$ from the photoluminescence polarization.

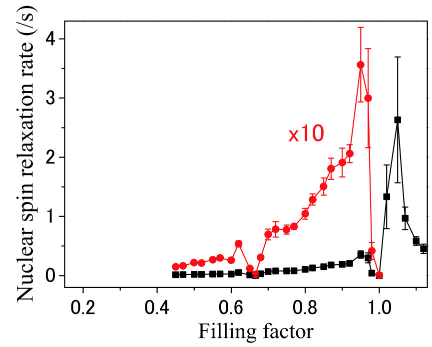

FIG. 5 (color online). The ν dependence of the nuclear spin relaxation rate. The red curve shows the magnification of $\nu \leq 1$.

This method is based on the fact that B_N is proportional to the electron spin polarization difference between the optical pumping and equilibrium conditions. On the basis of this estimation method, we obtained the trend of $\langle S_z\rangle_{eq}$ and thus analyzed the experimental data. Finally, we concluded that ν dependence of B_N is understood by not fractional quantum Hall states but the effect of electron spin polarization through excitons and trions. The obtained understanding of the optical nuclear spin polarization leads to nuclear spins being effectively manipulated by combining optical and electrical means.

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