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 filling factor nonmonotonous. The comprehensive measurements of \( B_N \) with the help of the circularly polarized photoluminescence measurements indicate the participation of the photo-excited complexes i.e., the exciton and trion (charged exciton), for nuclear spin polarization. On the basis of a novel estimation of the equilibrium electron spin polarization, we 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 [2–5], and multiple quantum coherences of nuclear spins have been controlled in a nanometer-scale region [6]. 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 [7, 8]. The condition for the electrical pumping is restricted to a special electronic state such as spin 3/2. 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 \( \nu \). 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 \( \nu \)-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-\( \mu \)m-long and 30-\( \mu \)m-wide Hall bar. The electron density \( n_s \) can be tuned by applying the voltage to a \( n \)-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: \( \sim 2 \) ps, pulse repetition: 76 MHz). The electron mobility is 185 m\(^2\)/Vs for \( n_s = 1.2 \times 10^{11} \) m\(^{-2}\). A laser beam irradiated the whole Hall bar structure (beam diameter: 230 \( \mu \)m) 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.

The optical pumping was performed as follows. First, the nuclear spin polarization was fully destroyed by setting the electronic state to the skyrmion region \( \nu \). Second, circularly polarized light was injected to the electronic state with \( \nu \). The pumping time was 250 s. The pumping photon energy \( E_{\text{las}} \) and the average power density \( P \) is specified below. The laser illumination increased the temperature of the sample holder up to 0.4 K and also disguised the sample resistance. Third, \( \nu \) 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 \( \nu \). This relaxation time we estimated is over \( 1.6 \times 10^3 \) s.

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

The optically induced nuclear spin polarization was measured by the resistive detection method using a peak shift of the spin phase transition at $\nu = 2/3$ [8, 9], because the resistive detection provides the high sensitivity of nuclear polarization. Here, we recorded the nuclear magnetic field $B_N$ induced by the nuclear spin 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.

Figure 1 shows the $\nu$-dependence of optically induced $B_N$ for $\sigma^+$ ($\sigma^-$) excitation with $E_{\text{laser}} = 1.5328 \ (1.5321) \text{ eV}$. 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 $\sigma^+$ and $\sigma^-$ excitations show the opposite direction because the conduction electrons with down and up spins are created by $\sigma^+$ and $\sigma^-$ excitations, respectively [9]. When $\nu$ increases from the lower-side, the magnitude of $B_N$ decreases for both excitations. In the $\nu$-range from 0.4 to 0.9, the relatively small value of $B_N$ is observed for $\sigma^+$ and the apparent nuclear spin polarization is not observed for $\sigma^-$. Around $\nu = 1$, nuclear spins are not polarized for either excitations. The stronger excitation power exhibits the larger magnitude of $B_N$, which is explained by the increased pumping rate. However, the non-monotonic behavior of the $\nu$-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 $\nu$-dependence of optically induced $B_N$ by changing $E_{\text{laser}}$ with $P = 1.2 \text{ W/cm}^2$. The optical pumping rate is expected to depend on the photon absorption rate, and $\nu$-dependence with the constant $E_{\text{laser}}$ should be modified when the absorption spectrum is varied with $\nu$. 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 [10]. The use of our sample exhibits the difficulty of the absorption measurement. 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 [11]. Therefore, we also measured the circularly polarized PL with a linear polarized laser excitation energy of 1.58 eV, the power density of 1.2 W/cm$^2$, and a spectral resolution better than 0.2 meV.

Figure 2 (a) ((b)) shows the color map of $B_N$ for $\sigma^+$ and $\sigma^-$ excitations. The scale of 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.

FIG. 2. (Color online) The 2D color map of $B_N$ for (a) $\sigma^+$ and (b) $\sigma^-$ excitations. The scale of 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.
triplet trion peak is larger than that at the singlet trion peak. This accounts for the difference between (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 [11, 12] under our experimental conditions. The neutral exciton has greater oscillator strength than the triplet trion in the absorption measurement [11] and in the numerical calculations in high B-field [13]. Therefore, we can attribute the nuclear polarization at the triplet PL peak to the absorption of the neutral exciton. In contrast, we consider the nuclear polarization at the singlet PL peak as the consequence of the absorption of the singlet trion [14]. Indeed, in the absorption experiment (under conditions similar to ours) performed by Groshaus et al. [8], two peaks were assigned to the neutral exciton X and the singlet trion T [16].

We discuss how the optically excited complexes affect the nuclear spin polarization. Our experimental results indicate that the photo-excitation 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 [11, 12]. This can explain our results simply. However, the absorption measurement does not always show such behavior under the experimental conditions similar to ours [1]. To polarize nuclear spins, primarily, the electron spin polarization under optical pumping $\langle S_z \rangle$ is more crucial than the number of injecting electron spins. The effective nuclear magnetic field after long pumping time is given by $B_N = -A \langle (S_z - S_z^{eq}) \rangle$, where $A(>0)$ is a constant and $\langle S_z^{eq} \rangle$ is the equilibrium electron spin polarization [8, 17]. This fact and the experimental results indicate that the photo-excitation 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 [18]. 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 $\langle S_z^{eq} \rangle$ to understand the $\nu$-dependence of $B_N$. To know the electron spin polarization $P_e$, the optical dichroism calculated from the trion absorption is available [1]. 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 [19, 20]. We develop the $P_e$ estimation from $P_L$. The right (left) circular polarized PL intensity $I_{+(-)}$ is proportional to the number of photo-excited particles multiplied by its oscillator strength. We define $P_L$ as $\langle I_{+} - I_{-} \rangle / \langle I_{+} + I_{-} \rangle$ and here consider this formulation for $T$. Since the photo-creating 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 [1]. Consequently, the calculation of $P_L$ for $T$ gives $P_L = (P_L - P_h) / (1 - P_h P_h)$ for $\nu \leq 1$ and $P_L = (2 - \nu) / \nu \cdot (1 - P_L - P_h) / (1 - P_L P_h)$ for $\nu > 1$, where $P_h$ is the hole polarization due to the singlet nature of $T$ [21]. While $P_h$ increases with $B$ [2], our experiments were performed under constant $B$, and $P_h$ should be constant. Figure 3 (a) and (b) respectively show the measured $P_L$ for $T$ and the calculated $P_L$ with the constant $P_h$ of $-0.9$, $-0.7$ and $-0.5$. Since the optical pumping was performed with the strong illumination, the $P_e$ obtained here (under the strong photo-excitation) is treated as $\langle S_z^{eq} \rangle$ [22]. Thus, we obtain the trend of $\langle S_z^{eq} \rangle$ although the correct values are uncertain due to the lack of $P_h$ information.

To consider how $\langle S_z^{eq} \rangle$ 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. The slight $B_N$ increase in the $\nu$-range from 0.4 to 0.8 can be understood by the obtained trend of $\langle S_z^{eq} \rangle$. In $\nu < 0.4$, $B_N$ does not obey the trend of $\langle S_z^{eq} \rangle$. 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 injecting electron 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 complete explanation, it should be noted that diminishing the
doping enhances \( \langle S_z \rangle \) in \( B \) parallel to the well [24].

Finally, we consider the optical nuclear polarization in (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 \( \nu \) after optical pumping. The time decay of \( B_N \) is fitted by the simple exponential function. The observed nuclear spin relaxation rates \( 1/T_{1N} \) are displayed in Fig. 5. 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 \( \nu = 1 \) [25]. 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 \( \nu \) dependence of the optically induced \( B_N \). To understand this behavior, we constructed a novel estimation of the equilibrium electron spin polarization from the photoluminescence polarization because \( B_N \) is proportional to the electron spin polarization difference between the optical pumping and equilibrium conditions. On the basis of this estimation, we concluded that \( \nu \) 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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the $T$ transition strength always exists.

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Supplemental Material for

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In this supplemental material, we describe the details of our estimation of electron spin polarization from the singlet trion photoluminescence (PL) polarization. We construct the relationship between the electron spin polarization and the PL polarization by modeling the oscillator strength (OS) of a singlet trion as proportional to the number of unpaired electrons with the spin opposite to the photo-created electron spin. By using the constructed relationship, we show an example of the estimation of electron spin polarization from the experimentally obtained PL polarization.

I. SINGLET TRION PHOTOLUMINESCENCE POLARIZATION

A singlet trion is two conduction band electrons with opposite spin coupling with a valence band hole due to the Coulomb interaction. The PL of the singlet trion shows the right and left circular polarization ($\sigma^+$ and $\sigma^-$) corresponding to the two opposite directions of the hole spin. Since the PL intensity is generally proportional to the number of the photo-excited particles and their OSs, the $\sigma^+$ and $\sigma^-$ PL intensity are given by

$$I_{\sigma^+} = c N_\uparrow/\tau_\uparrow, \quad I_{\sigma^-} = c N_\downarrow/\tau_\downarrow,$$

where $c$ is a constant, $N_\uparrow$ ($N_\downarrow$) is the number of singlet trion with the up (down) spin $\uparrow$ ($\downarrow$) hole, and $1/\tau_\uparrow$ ($1/\tau_\downarrow$) is its OS.

We define the PL polarization $P_L$ for singlet trion as

$$P_L = I_{\sigma^+} - I_{\sigma^-}/I_{\sigma^+} + I_{\sigma^-}.$$  \hspace{1cm} (S2)

Note that the energies of $\sigma^+$ and $\sigma^-$ are different.

According to the study of singlet trion absorption $^1$, since the photo-created electron needs to pair with an opposite spin, the OS of each trion is proportional to the number of unpaired electrons with opposite spin. Thus, we obtain

$$1/\tau_\uparrow = C f_\uparrow N_\uparrow, \quad 1/\tau_\downarrow = C f_\downarrow N_\downarrow,$$  \hspace{1cm} (S3)

where $C$ is a constant, $f_\uparrow$ ($f_\downarrow$) is the fraction of spin up (down) $\uparrow$ ($\downarrow$) electrons that are unpaired, and $N_\uparrow$ ($N_\downarrow$) is the number of $\uparrow$ ($\downarrow$) electrons.

Assuming $f_\uparrow = 1$ for $\nu \leq 1$ $^1$, we substitute Eqs. (S1) and (S3) in Eq. (S2) and obtain

$$P_L = P_h + P_e/1 + P_e P_h,$$  \hspace{1cm} (S4)

where $P_h = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$ is the trion spin polarization and $P_e = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$ is the electron spin polarization. The trion spin polarization $P_h$ is identical to the hole spin polarization, because two electrons in trion form the spin singlet state, which has zero resultant spin.

Accordingly, Eq. (S4) can be transformed into

$$P_e = P_L - P_h/1 - P_L P_h,$$  \hspace{1cm} (S5)

and we can estimate $P_e$ from $P_L$ when we attain the information of $P_h$.

For $\nu > 1$, we assume $f_\uparrow = 1 - [(N_\uparrow + N_\downarrow) - N_\phi]/N_\phi$, where $N_\phi$ is the degeneracy factor of the spin split Landau level $^1$. The similar calculation with $(N_\uparrow + N_\downarrow)/N_\phi = \nu$ gives

$$P_e = P_L - P_h (2 - \nu)/(1 - P_L P_h) \nu,$$  \hspace{1cm} (S6)

where $\nu$ is filling factor.
II. ESTIMATION OF ELECTRON SPIN POLARIZATION

We measured the PL under experimental conditions almost the same as those in the main manuscript. Therefore, the sample we used was a single 18-nm GaAs/Al_{0.33}Ga_{0.67}As quantum well that was cooled in a cryogen free He refrigerator down to 0.3 K. The linearly polarized excitation laser beam with photon energy of 1.58 eV was injected through an optical window on the bottom of the cryostat and the left circularly polarized PL was collected through the same window with a spectral resolution better than 0.2 meV under the external magnetic field $B$ of ±7.15 T. Negative $B$ was used to avoid the optical loss difference caused by the different optical system. The left circular polarization under negative $B$ corresponds to the right circular polarization under positive $B$. The difference from the main manuscript is the much smaller laser power of 2.4 mW/cm$^2$.

We recorded the dependence of the PL on $\nu$ by using the back gate. The obtained singlet trion PL polarization $P_L$ from Eq. (S2) is shown in Fig. S1. We clearly observe the peak at $\nu = 1/3$, $2/3$, and 1. In the main manuscript, we only observe the peak at $\nu = 1$ in Fig. 3 (a). This difference can be understood by the weaker laser heating effect because only the excitation laser power is different between Fig. S1 and Fig. 3 (a).

![FIG. S1](image1.png)

**FIG. S1.** The $\nu$-dependence of the single trion PL polarization obtained from the PL measurement with the excitation laser power of 2.4 mW/cm$^2$.

![FIG. S2](image2.png)

**FIG. S2.** (Color online) The calculated $P_e$ with $P_h = -0.9, -0.7$ and $-0.5$ using Eqs. (S5) and (S6).

To calculate $P_e$ from Eqs. (S5) and (S6), we have to acquire $P_h$. The value of $P_h$ generally depends on $B$ [2]. In our experiment, $\nu$ is tuned by using the back gate and $B$ is a constant. Therefore, $P_h$ is expected to be constant in all ranges of $\nu$. If one has the knowledge of a certain value of $\nu$ with $P_e = 0$ (e.g. $\nu = 2$), $P_e$ is equal to $P_h$ and one can obtain the value of $P_h$ from the PL polarization experiment. However, we cannot extract the correct value of $P_h$ from our experimental results. The fact that the g-factor of GaAs is negative indicates that $P_e$ ranges from 0 to 1 in quantum Hall regime. We assume the constant value of $P_h$ is given by the resultant value of $P_e$ satisfied with this range. Figure S2 shows the calculated $P_e$ with $P_h = -0.9, -0.7$ and $-0.5$ using Eqs. (S5) and (S6). The obtained behavior of $P_e$ is consistent with the previous studies [1, 3]. This can validate our estimation method.

Although we cannot obtain the correct value of $P_e$ due to the lack of information of $P_h$ in our experiments, we can at least conclude that the trend of $P_e$ reflects that of $P_L$ under constant $B$ from the comparison between Figs. S1 and S2.
Furthermore, once one obtains the correct value of $P_h$ or determines the $P_e$ value at a certain $\nu$ from the experiment such as the nuclear magnetic resonance, the estimation of $P_e$ from $P_L$ we constructed here becomes fairly useful method.

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