Simultaneous Measurement of Resistively and Optically Detected Nuclear Magnetic Resonance in the $\nu = 2/3$ Fractional Quantum Hall Regime

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We observe nuclear magnetic resonance (NMR) in the fractional quantum Hall regime at Landau level filling factor $\nu = 2/3$ from simultaneous measurement of longitudinal resistance and photoluminescence (PL). The dynamic nuclear spin polarization is induced by applying a huge electronic current at the spin phase transition point of $\nu = 2/3$. The NMR spectra obtained from changes in resistance and PL intensity are qualitatively the same; that is, the Knight shift (spin polarized region) and zero-shift (spin unpolarized region) resonances are observed in both. The observed change in PL intensity is interpreted as a consequence of the trion scattering induced by polarized nuclear spins. We conclude that both detection methods probe almost the same local phenomena.

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Quantum Hall effect has attracted a lot of physical interest since its discovery, and it has been investigated by using various kinds of experimental methods as well as other phenomena of condensed matter physics. Different experimental techniques can usually offer a good understanding of physics. However, considerable discrepancies between them occasionally arise in the quantum Hall system, even when sample preparations and experimental conditions are almost the same. For instance, the different size of skyrmion has been observed and argued [1–4]. The optical nuclear polarization observed in optical method is much larger than that in conventional and resistive methods [4–9]. The case of the electron spin polarization at Landau level filling factor $\nu = 5/2$ is more complicated; the different experimental methods show the fully polarized state, unpolarized system, and partially polarized domains [10–17]. In these studies, the results obtained with optical methods especially exhibited controversial disagreements with other experimental methods. The possible explanation of such disagreements can be expected as follows: optically accessible phenomena can occur in the spatially limited region and/or photoexcited holes can considerably affect the system due to the Coulomb interaction.

To investigate such different experimentally-observed results, the simultaneous measurement by different experimental methods is effective. In this paper, we measure the resistance and photoluminescence (PL) in the $\nu = 2/3$ fractional quantum Hall regime simultaneously. At $\nu = 2/3$, an electron spin phase transition (SPT) can occur due to competition between Coulomb and Zeeman energies [18], and this SPT has been observed from the resistance and PL so far [18, 20]. Associated with the two electron spin phases (i.e., spin polarized and unpolarized states), nuclear spins are polarized when a huge electronic current is applied [18]. In the present study, the target phenomena to be simultaneously measured are this current-induced nuclear spin polarization and its nuclear magnetic resonance (NMR). This is because NMR provides local information from its spectrum [21], which has a possibility to identify the essential difference between resistive and optical detections. In addition, the still-ambiguous details of the nuclear spin polarization at $\nu = 2/3$ SPT, which are crucial for a future application to quantum information technology [22], can be investigated.

We demonstrate simultaneous measurement of resistively and optically detected NMR with the current-induced nuclear spin polarization at $\nu = 2/3$ SPT. Accompanying the dynamic nuclear spin polarization, the previously-reported enhancement of longitudinal resistance [18] and the variation of PL intensity occurred at the same time. Subsequently, we obtained NMR spectra from changes in both resistance and PL intensity. The simultaneously-measured spectra are qualitatively the same. The resistively-detected spectrum is consistent with that of a previous study [23], excluding the influence of optical illumination. The optically-detected spectra enable us to interpret that the variation of PL intensity due to nuclear spin polarization is caused by the trion (photoexcited particle) scattering. It is thus concluded that the proposed simultaneous measurement (namely, resistive and optical detection methods) probe almost the same local phenomenon.

Experiments were carried out on a single 18-nm GaAs/Al0.33Ga0.67As quantum well, which was processed to a 100-μm long and 30-μm wide Hall bar. The electron density $n_s$ can be controlled by applying a voltage to a n-type GaAs substrate (back gate). The electron mobility is $185 \, \text{m}^2/\text{Vs}$ for $n_s = 1.2 \times 10^{15} \, \text{m}^{-2}$. This sample was cooled down to 0.3 K in a cryogen free $^3\text{He}$ refrigerator. A longitudinal resistance was mea-
FIG. 1. (Color online) Photoluminescence spectra at $\nu = 2/3$ before (solid line) and after (dotted line) current-induced nuclear spin polarization.

sured by using a lock-in technique with a low-frequency (83 Hz) constant current. Luminescence was excited by a linearly-polarized output of a mode-locked Ti:sapphire laser (pulse width: $\sim 2$ ps, pulse repetition: 76 MHz) with wavelength of 784 nm and average power density of 2 mW/cm$^2$. A laser beam (diameter: $\sim 230 \mu m$) continuously irradiated the whole Hall bar through an optical window on the bottom of the cryostat. The propagation direction of the laser beam was parallel to an external magnetic field of 7.15 T, which was perpendicular to the quantum well. The left circularly polarized ($\sigma^{-}$) PL was collected from the entire laser-excitation area through the same optical window, where the PL collection time was 265 s. The details of this experimental setup are the same as those stated in our previous work [24].

The solid line in Fig. 1 shows the PL spectrum at $\nu = 2/3$. We observe two peaks: an apparent peak at 1.5324 eV, and a tiny peak at 1.5333 eV. The optical spectrum in the fractional quantum Hall regime has been understood by existing bound electron-hole complexes, e.g., neutral and charged (trions) excitons [26]. Here, the two electrons in a trion form singlet and triplet spin states. The lower and higher energy peaks are respectively assigned to singlet and triplet trion peaks [20, 25].

Figure 2 shows the simultaneously measured longitudinal resistance $R_{xx}$ and PL around $\nu = 2/3$. The applied current was 30 nA, which was low enough not to polarize nuclear spins, and $\nu$ was tuned by using the back gate in the fixed magnetic field. The large dip in $R_{xx}$ in (a) is associated with the fractional quantum Hall state at $\nu = 2/3$ and the $R_{xx}$ peak at $\nu \sim 0.67$ is caused by the spin phase transition [13]. The spin polarized (unpolarized) state is known to be formed on the lower (higher) $\nu$ side of this peak. As shown in Fig. 1, the singlet and triplet peaks appear in the PL spectrum. The integral intensity around these peaks in (b) and (c) was recorded. As $\nu$ increases, the singlet intensity starts to increase around the $R_{xx}$ peak. This increase in singlet intensity accompanies with a decrease in triplet intensity. These changes in PL intensity are consistent with the SPT; the triplet (singlet) trion mainly should reside in the spin polarized (unpolarized) region because the two electron spins in a triplet (singlet) trion are aligned in parallel (anti-parallel). Indeed, our observation of the SPT from PL is in qualitative agreement with a previous investigation on optical detection of the spin phase transition [20, 27].

In order to induce dynamic nuclear spin polarization, we applied a huge current of 240 nA at the $\nu = 2/3$ SPT point (the broken line in Fig. 2). After the current-induced nuclear spin polarization (polarization time: 8000 s), singlet intensity decreases and triplet intensity increases as shown by the dotted line in Fig. 1. The temporal development of these PL intensity changes is presented in Fig. 3 where $R_{xx}$ was measured simultaneously. As already reported [18], the resistance enhancement occurs due to the nuclear spin polarization. The time scale of the change in PL integral intensity (i.e., the decrease in singlet integral intensity and the increase in triplet integral intensity) is almost the same as that of the resistance enhancement (see Fig. 3). This strongly suggests that the PL intensity changes are caused by the current-induced nuclear spin polarization.

As mentioned above, the triplet (singlet) trion mainly should reside in the spin polarized (unpolarized) region. Therefore, the increase in triplet intensity accompanying
the decrease in singlet intensity in Fig. 3 seems to mean both spreading of the spin polarized region and shrinking of the spin unpolarized region. However, the saturated value of triplet integral intensity is larger than the maximum value of that in Fig. 2 (c); that is, triplet intensity after nuclear spin polarization is larger than that in the spin polarized state. The total amount of PL intensity with dynamic nuclear spin polarization is attributed to not only the size of the spin region but also another factor. In a later section, we will discuss the origin of the change in PL intensity.

Next, we performed the NMR experiment. To obtain the NMR spectra, we used the following procedures. First, a huge current (240 nA) was applied to the sample at \( \nu = 2/3 \) for long enough to saturate the resistance change (over 3 h), where an off-resonant radio frequency (RF) magnetic field was irradiated using a handmade split coil in order to incorporate the influence of the RF irradiation (e.g. electron temperature increase). Second, we only changed the RF frequency and then waited for 250 s so that the system reached a stationary state. Third, the PL spectra were collected for 265 s and \( R_{xx} \) was measured 100 times during the PL collection time. As a result, we acquired an averaged \( R_{xx} \) and singlet and triplet integral intensities at a certain RF. By repeating the second and third procedures, we obtained the NMR spectra from the changes in resistance and PL intensity simultaneously.

Figure 4 shows the NMR spectra for \(^{75}\text{As}\) nuclear spin \( I = 3/2 \), where 40 spectra obtained by each detection method were averaged, since the signal-to-noise ratio was too low for PL detection [28, 29]. As shown in (a), the resistively-detected NMR spectrum clearly exhibits two relatively sharp resonance lines at 52.536 and 52.549 MHz and one broadened resonance peak on the lower-frequency side. These features are also observed in the (b) singlet and (c) triplet PL detections, where the signal in (b) varies in the opposite direction because the change in singlet intensity associated with the nuclear spin polarization is opposite to the others (see Fig. 3). Although the signals obtained from the PL were influenced by the PL intensity fluctuation, all spectra (a)–(c) are qualitatively the same [30]. Note that we demonstrate for the first time not only the simultaneous measurement of the resistive and optical NMR but also the optical NMR from trion intensity.

The sharp and broad resonances are respectively attributed to the spin unpolarized and polarized regions. The energy of nuclear spin resonance is shifted by the electron spin polarization \( P_e \) due to hyperfine interaction, which is known as the Knight shift [18]. The spin unpolarized region (\( P_e = 0 \)) brings about no energy shift and the spin polarized region (\( P_e = 1 \)) causes a negative energy shift. This energy shift depends on electron density; the electron distribution in the growth direction, which is formed by confinement of the quantum well, broadens the total Knight shift [31]. We interpret two of
the sharp resonance lines as a consequence of quadrupole splitting and the population difference among fourfold-nuclear-spin levels. Although the quadrupole splitting of $^{75}$As nuclear spins causes three resonance lines, the population distribution among four levels changes the relative strengths of these lines. The fact that two Knight shift resonances were not observed is due to the broadening. The frequency scales of the broadening and the splitting (i.e., the total shape of the NMR spectrum) are consistent with the previous studies on resistively-detected NMR of $\nu = 2/3$ [23, 32, 34]. Moreover, the consistency of the previous reports means that the laser illumination does not influence resistively-detected results in spite of the existence of photoexcited carriers.

On the basis of our interpretation of the NMR spectra, we consider physics behind the optical detection as follows.

The NMR spectra obtained by both the singlet [Fig. 4(b)] and triplet [Fig. 4(c)] PLs show the contributions from the spin unpolarized and polarized regions. This fact indicates that the singlet and triplet trions co-exist in the spin polarized and also unpolarized regions after the nuclear spin polarization.

We, here, consider the strength of the NMR signal. In Figs. 4(b) and (c), neither the spin polarized nor unpolarized signal is striking. This observation is not expected and might seem to be a contradiction; that is, although the singlet (triplet) trions reside in both the spin polarized and unpolarized regions, the singlet (triplet) trion preferably exists in the spin unpolarized (polarized) region and the spin unpolarized (polarized) signal should be pronounced in (b) [(c)]. However, we only recorded the deviations from the change in PL intensity accompanying the nuclear spin polarization in (b) [(c)]. Therefore, when the nuclear spin polarization simply decreases singlet intensity and simultaneously increases triplet intensity in both the spin polarized and unpolarized regions, we can understand the seemingly contradicting result even though the spin unpolarized (polarized) region mainly radiates singlet (triplet) trion light. This means that the change in singlet (triplet) PL intensity due to nuclear spin polarization does not solely depend on the size of the spin unpolarized (polarized) region.

We interpret the change in PL intensity as trion scattering induced by the nuclear spin polarization. After the current-induced nuclear spin polarization, the observed $R_{xx}$ peak around $\nu = 2/3$ becomes larger and broader, indicating that the obtained nuclear spin polarization is spatially inhomogeneous and that both positive and negative polarizations exist [18, 55]. Therefore, the current-induced nuclear spin polarization creates a spatial modulation of the electron Zeeman energy through the hyperfine interaction. This potential fluctuation can enhance the trion-scattering process. The scattering usually suppresses radiative recombination, which accounts for the change in singlet PL intensity. In the case of the triplet trion, the scattering should enhance PL intensity in order to explain the experimental results. This is understood by the existence of the dark triplet trion. The triplet PL intensity is contributed by the bright and dark triplet trions [36, 37]. The dark triplet state can recombine through a scattering process that changes its total angular momentum. Indeed, the increase in triplet PL intensity due to a random potential induced by remote ionized donors has been observed [20]. Therefore, we claim that the similar scenario occurs owing to the potential fluctuation induced by polarized nuclear spins.

In conclusion, we demonstrated the simultaneous measurement of resistively and optically detected NMR at $\nu = 2/3$. The simultaneously measured NMR spectra qualitatively showed the same features, which are broad and sharp resonances respectively associated with spin polarized and unpolarized regions. From the unexpected optical NMR spectra, we interpreted the optically detected signal as a consequence of trion scattering induced by polarized nuclear spins. Thus, both detection methods probe almost the same local phenomena. This means that optical accessible phenomena do not occur in the spatially limited region. Even though our identical observations of the NMR spectra with different methods are not surprising naively, considering how and where to detect the phenomena by each method provides a new insight into the nuclear spin phenomena at $\nu = 2/3$, which can lead to a future application to quantum information technology. We believe that simultaneous measurement by different methods and the consideration of their detection details are important and give a way to understand physics behind controversial disagreements between different detection methods.

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