Strain-induced enhancement of electric quadrupole splitting in resistively detected nuclear magnetic resonance spectrum in quantum Hall systems

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We show intentional enhancement of electric quadrupole splitting in nuclear spin energy levels is intentionally enhanced by applying an external stress to the heterostructure. Nuclear magnetic resonance spectra with clearly separated triple peaks are obtained, and Rabi oscillations are observed between the nuclear spin energy levels. The decay of the spin-echo signal is compared between the cases before and after the enhancement of quadrupole splitting.

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Nuclear spins in semiconductors have recently attracted considerable attention because their extremely long coherence time is suitable for the implementation of quantum bits/memories. In order to manipulate nuclear spin quantum states coherently, all-electrical/optical nuclear magnetic resonance (NMR) techniques have been developed on the basis of the hyperfine interaction between nuclear spins and electron spins. However, all of these new NMR techniques have been successfully applied in GaAs. Since all the constituent atoms in GaAs have nuclear spins I = 3/2, the nuclear spin states split into four-level J_z eigen states |±3/2⟩ and |±1/2⟩ in a magnetic field as shown on the left-hand side of Fig. 1(a). Such a four-level system can be regarded as coupled quantum bits if transitions between any pairs of the levels are controlled selectively.

In the presence of a local electric field gradient, the electric quadrupole interaction produces non-equidistant nuclear spin energy levels as shown on the right-hand side of Fig. 1(a). Although the electric quadrupole splitting energy ∆Q is zero in GaAs because of the cubic symmetry of the GaAs crystal, it is possible to increase the amplitude of ∆Q by applying an external stress to the crystal, because ∆Q is proportional to the local electric field gradient. Such an external stress can be applied using pressure cells or piezoelectric devices or by coating the surface of GaAs with different materials.

In this paper, we show intentional enhancement of electric quadrupole splitting and selective control of a four-level nuclear spin system. Using the breakdown phenomenon of quantum Hall effect, nuclear spins are polarized and NMR are detected. We apply an external stress to a Hall-bar device by coating its surface with a polyimide film. NMR spectra with clearly separated triple peaks are obtained in the polyimide-coated devices. Splitting of NMR spectra enables us to show the selective and coherent manipulation of a four-level nuclear spin system using pulsed NMR techniques. Furthermore, the decay of spin-echo signal is compared between the cases before and after the enhancement of quadrupole-splitting.

The experiments were performed using a two-dimensional electron gas (2DEG) in a GaAs/Al_{0.3}Ga_{0.7}As single heterostructure wafer grown by molecular beam epitaxy on a (001) oriented GaAs substrate. The 2DEG is located at 230 nm below the surface. The mobility and density of the 2DEG are 220 m^{2}/Vs and 1.6 × 10^{15} m^{-2}, respectively. Figure 1(b) shows an optical micrograph of the Hall-bar device used in the present study. A 10-µm-wide Ti/Au Schottky gate electrode that was used for tuning electron density also functioned as a local coil for generating radio-frequency (rf) magnetic fields B_{rf} parallel to the
As an initialization process, nuclear spins are dynam-
ization (DNP) and resistive detection (RD) techniques
dilution refrigerator to make a good thermal contact.

The manipulated nuclear spin state is read out by mea-
suring the longitudinal voltage $V_{xx}$ of the Hall-bar de-
vice. The read-out procedure is based on the fact that
the positively polarized nuclear spins ($\langle I_z \rangle > 0$) reduce
the Zeeman splitting energy of electrons, which increases
$V_{xx}$.

First, we measured the NMR spectrum using an un-
coated Hall-bar device. In Figs. 2(a), (b), and (c), the
changes in the longitudinal voltage $\Delta V_{xx}$ induced by
the $B_{rf}$ irradiation are plotted as a function of the $B_{rf}$
frequency. Each curve corresponds to the NMR spectrum
for $^{75}$As, $^{69}$Ga, and $^{71}$Ga. A single-peak spectrum is
observed for all the three nuclear species. These single-
peak spectra indicate that the nuclear spin levels are dis-
buted almost equidistantly as illustrated in the left-
hand panel in Fig. 1(a).

Next, the Hall-bar device was warmed up to room
temperature; at this temperature, a droplet of polyimide so-
lution was dropped onto the surface of the device. Then,
the polyimide coating was baked in $N_2$ atmosphere at 180
°C for 15 min. The polyimide-coated device was cooled
down again for the NMR measurements. Since the ther-
mal shrinkage rate of the polyimide film is considerably
higher than that of the GaAs film, the subsequent cool-
ing of the polyimide-coated device is expected to induce
a large strain in the device.

Figure 2(d) shows the NMR spectrum of $^{75}$As after
it was coated with the polyimide film. The NMR spec-
trum is split into three peaks. These peaks correspond to
transitions A, B, and C shown on the right-hand side of
Fig. 1(a). The NMR spectra of $^{69}$Ga and $^{71}$Ga are also
split as shown in Figs. 2(e) and (f), respectively. Ampli-
ditudes of the splitting are $\Delta f = 36$ kHz, 18 kHz, and 11
kHz for $^{75}$As, $^{69}$Ga, and $^{71}$Ga, respectively. The ratio of
$\Delta f$ is in good agreement with the ratio of the quadrupole
moment $Q$: $\Delta f^{(69)Ga}/\Delta f^{(71)Ga} = 1.6$ agrees with
$Q^{(69)Ga}/Q^{(71)Ga} = 0.19 \times 10^{-28} \text{ m}^2/0.12 \times 10^{-28} \text{ m}^2 =
1.6$. This indicates that the splitting of the NMR spectra is
attributed to the electric quadrupole interaction. We
observed the splitting in the spectrum of $^{75}$As in another
device; the single-peak spectrum before the polyimide
coating is split to three peaks ($\Delta f = 16$ kHz) after the
polyimide coating. The NMR peak splitting of 7.5 kHz
for $^{75}$As was also observed in yet another device after
coating its surface with PMMA electron-beam resist.

We consider that the polyimide film produces a strain
in the GaAs/AlGaAs heterostructure, resulting in the gen-
eration of a large electric field gradient at nuclear spin
sites, and the induced electric field gradient enhances $\Delta Q$.
Comparing the observed splitting of 36 kHz in the NMR
spectrum of $^{75}$As [Fig. 2(d)] with the earlier measure-
ments in GaAs quantum wells, the strain in our device
is estimated as $1.7 \times 10^{-4}$. The estimated value of the
strain seems consistent with the results of electron trans-
port measurements of 2DEG under a strain-induced pe-
riodic potential modulation. From the FWHM of the
NMR spectrum of $^{75}$As [Fig. 2(a)], the strain in the de-
vice before the polyimide film coating is estimated to be

2DEG. External static magnetic field $B$ was applied
perpendicular to the 2DEG, hence parallel to the [001]
direction of the GaAs crystal. All the measurements
were performed at 50 mK using a $^3$He-$^4$He dilution
refrigerator. The sample chip (1 mm $\times$ 1 mm $\times$ 0.5 mm)
was glued backside to a ceramic chip carrier using silver
paste. After wiring to the Hall-bar device, the ceramic
package was held against the cold finger plate of the
dilution refrigerator to make a good thermal contact.

NMR signals were obtained by dynamic nuclear polar-
ization (DNP) and resistive detection (RD) techniques
in a breakdown regime of integer quantum Hall effect
(QHE), as already demonstrated in our earlier studies. As
an initialization process, nuclear spins are dynam-
ically polarized through the hyperfine interaction be-
tween nuclear spins and electron spins under a breakdown
regime of a quantum Hall state with the Landau level fill-
ing factor $\nu = 1$. By applying a bias current larger than the
critical current of the QHE breakdown, electrons are
excited to the upper Landau subband, accompanied by
flips of electron spins. The flips of electron spins cause
flips of nuclear spins via the hyperfine interaction, re-
sulting in positive nuclear polarization ($\langle I_z \rangle > 0$). Then,
the nuclear spin states are manipulated by applying $B_{rf}$.

![NMR spectra](image-url)
The frequencies of $B_1$ are 41.952 MHz (A), 41.987 MHz (B), and 42.022 MHz (C) in (a), and 41.969 MHz (D) and 42.007 MHz (E) in (b). The curves are offset for clarity. The inset in (a) shows schematic energy diagram for single- and two-photon absorption/emission. The inset in (b) shows the NMR spectrum of $^{75}$As with the input rf-voltage $V_{rf}$ = 4.8 V.

not larger than $3.9 \times 10^{-5}$, even if the broadening of the spectrum is attributed to $\Delta Q$. Therefore, contribution of the other sources of strain, such as Ti/Au Schottky gate or the silver paste on the backside of the sample chip, is small compared to that of the polyimide film.

Figure 3(a) shows the changes in $V_{xx}$ induced by applying a pulse of $B_{rf}$ with various pulse durations $\tau_{\text{pulse}}$ at $B = 5.82$ T ($\nu = 1$). We note that the amplitude of $B_{rf}$ in the pulsed NMR measurements (Fig. 3) is 12 times larger than that used to obtain the continuous-wave NMR spectra (Fig. 2). The $B_{rf}$ frequencies for the curves A, B, and C are 41.952 MHz, 41.987 MHz, and 42.022 MHz, respectively, as indicated in the inset in Fig. 3(b). The oscillatory changes in $\Delta V_{xx}$ denoted A, B, and C correspond to the Rabi oscillations of $^{75}$As for transitions A ($|+3/2\rangle \leftrightarrow |+1/2\rangle$), B ($|+1/2\rangle \leftrightarrow |-1/2\rangle$), and C ($|-1/2\rangle \leftrightarrow |-3/2\rangle$), respectively. These results clearly show that the intentional enhancement of $\Delta Q$ enables the selective and coherent control of the four-level nuclear spin system. Additional two peaks (D and E) are seen in the spectrum at the middle frequencies between the peaks A and B, and B and C as similar to the work by Yusa et al.20. These additional peaks correspond to the two-photon absorption/emission processes ($|+3/2\rangle \leftrightarrow |-1/2\rangle$ and $|+1/2\rangle \leftrightarrow |-3/2\rangle$) induced by the irradiation of $B_{rf}$ with a large amplitude. The oscillations D and E in Fig. 3(b) correspond to the two-photon Rabi oscillations taken at the $B_{rf}$ frequencies of 41.969 MHz and 42.007 MHz, respectively. The observed frequency of the two-photon Rabi oscillation $\Omega_{\Delta m=2} = 3.8$ kHz nearly agrees with the calculated value $\Omega_{\Delta m=2} = 3.8$ kHz.

We verify the effect of electric quadrupole splitting on the nuclear spin coherence time by performing spin-echo experiments. We applied a sequence of $\pi/2$-rf pulses, as shown in the inset of Fig. 3(a). The inset of Fig. 4(b) shows a representative spin-echo signal in the device after the polyimide film coating obtained by changing the second waiting time $\tau_2$ with a fixed first waiting time $\tau_1 = 100 \mu s$ with the $B_{rf}$ frequency of 42.022 MHz. The coherence time $T_2$ is estimated from the decay of the spin-echo signal by changing the total waiting time $\tau_1 + \tau_2$ under the condition $\tau_1 = \tau_2$. Figure 4(a) shows the decay of the spin-echo signals for $^{75}$As in the device before the polyimide film coating. The $B_{rf}$ frequency was tuned to 41.963 MHz, the peak frequency in Fig. 4(a), where all the three NMR transitions occur simultaneously. The value of $T_2$ is estimated to be no longer than 0.2 ms, and the signal decays non-monotonically. In contrast, after coating the Hall-bar device with the polyimide film, the spin-echo signal decays exponentially as shown in Fig. 4(b). The $B_{rf}$ frequency was tuned to 42.022 MHz, the peak C in the inset of Fig. 4(b). The value of $T_2$ is estimated as 0.42 ms, which is almost twice longer than that obtained before the polyimide film coating. The decay time of the Rabi oscillations is also increased after the polyimide film coating (not shown). In addition, as shown in Fig. 4(c), the value of $T_2$ is further increased to 1.1 ms by decoupling the nuclear system from the electron system during nuclear-spin manipulation.22,23; electrons are depleted by applying negative dc voltage to the

FIG. 3: Changes in $V_{xx}$ induced by applying a pulse of $B_{rf}$ with various pulse durations $\tau_{\text{pulse}}$ at $B = 5.82$ T ($\nu = 1$). The frequencies of $B_{1}$ are 41.952 MHz (A), 41.987 MHz (B), and 42.022 MHz (C) in (a), and 41.969 MHz (D) and 42.007 MHz (E) in (b). The curves are offset for clarity. The inset in (a) shows schematic energy diagram for single- and two-photon absorption/emission. The inset in (b) shows the NMR spectrum of $^{75}$As with the input rf-voltage $V_{rf}$ = 4.8 V.

FIG. 4: (a) Decay of spin-echo signal obtained in the Hall-bar device before the polyimide coating. $B = 4.92$ T ($\nu = 1.08$). (b)-(c) Decays of spin-echo signals obtained in the Hall-bar device after the polyimide coating. $B = 5.82$ T ($\nu = 1.00$). In the case of (c), the electrons were depleted during the rf pulse irradiation. The inset of (a) shows a schematic of the pulse sequence for the spin-echo measurements. The inset of (b) shows a representative spin-echo signal obtained by changing $\tau_2$ with a fixed $\tau_1 = 100 \mu s$. 

We verify the effect of electric quadrupole splitting on the nuclear spin coherence time by performing spin-echo experiments. We applied a sequence of $\pi/2$-rf pulses, as shown in the inset of Fig. 3(a). The inset of Fig. 4(b) shows a representative spin-echo signal in the device after the polyimide film coating obtained by changing the second waiting time $\tau_2$ with a fixed first waiting time $\tau_1 = 100 \mu s$ with the $B_{rf}$ frequency of 42.022 MHz. The coherence time $T_2$ is estimated from the decay of the spin-echo signal by changing the total waiting time $\tau_1 + \tau_2$ under the condition $\tau_1 = \tau_2$. Figure 4(a) shows the decay of the spin-echo signals for $^{75}$As in the device before the polyimide film coating. The $B_{rf}$ frequency was tuned to 41.963 MHz, the peak frequency in Fig. 4(a), where all the three NMR transitions occur simultaneously. The value of $T_2$ is estimated to be no longer than 0.2 ms, and the signal decays non-monotonically. In contrast, after coating the Hall-bar device with the polyimide film, the spin-echo signal decays exponentially as shown in Fig. 4(b). The $B_{rf}$ frequency was tuned to 42.022 MHz, the peak C in the inset of Fig. 4(b). The value of $T_2$ is estimated as 0.42 ms, which is almost twice longer than that obtained before the polyimide film coating. The decay time of the Rabi oscillations is also increased after the polyimide film coating (not shown). In addition, as shown in Fig. 4(c), the value of $T_2$ is further increased to 1.1 ms by decoupling the nuclear system from the electron system during nuclear-spin manipulation; electrons are depleted by applying negative dc voltage to the
Schottky gate electrode during the rf-pulse irradiation. In summary, we have demonstrated strain-induced enhancement of the electric quadrupole splitting and electrical coherent manipulation in $I = 3/2$ nuclear spin energy levels in GaAs/GaAs heterostructure. The DNP and RD techniques used in the present study can be employed at temperatures higher than 1 K and even in a 2DEG with a relatively low electron mobility\textsuperscript{6,15}, because the techniques are based on the breakdown phenomena of QHE.

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