Resistive detection of nuclear spins in a single quantum dot under Kondo effect regime

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We study dynamic polarization and resistive detection of nuclear spins in a semiconductor quantum dot (QD) under the Kondo effect regime. We find that the differential conductance spectra of the QD exhibit hysteresis under the Kondo effect regime in magnetic fields. Relevance of nuclear spins to the hysteresis is confirmed by the detection of nuclear magnetic resonance signals by monitoring the differential conductance. We attribute the origin of the hysteresis to the dynamic nuclear spin polarization (DNP) induced in the QD. Using the DNP, we demonstrate nuclear spin relaxation rate measurements in the QD under the Kondo effect regime.

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The Kondo effect, arising from the interaction between a localized spin and itinerant spins at the Fermi energy, is one of the most fundamental many-body phenomena observed in semiconductor quantum dot (QD) systems.1–4 The differential conductance (dI/dVsd) spectrum of the QD exhibits a sharp zero-bias conductance peak (ZBCP), which reflects the Kondo resonance at the Fermi energy. The electronic state in the QD under the Kondo effect regime has a potential to open a way for studying electron spin polarization in a quantum dot through nuclear magnetic resonance (NMR) measurements.5

In this paper, we report dynamic polarization and resistive detection of nuclear spins in a single QD under the Kondo effect regime. We find that the dI/dVsd spectra exhibit hysteresis under the Kondo effect regime in magnetic fields. We also find that the value of dI/dVsd evolves slowly at the bias voltages between the split dI/dVsd peaks. Relevance of nuclear spins to the hysteresis and the slow evolution of dI/dVsd is confirmed by the detection of NMR signals by monitoring dI/dVsd. We attribute the origin of the hysteresis to the dynamic nuclear spin relaxation rate measurements in the QD under the Kondo effect regime.

We prepared QDs using a wafer of GaAs/Al0.3Ga0.7As single heterostructure with a two-dimensional electron gas (2DEG) at the interface. The carrier density and
mobility of the 2DEG are $n = 2.3 \times 10^{15} \text{ m}^{-2}$ and $\mu = 17 \text{ m}^2/\text{Vs}$, respectively. The QDs were made using split-gate devices [inset of Fig. 2(e)], following the approach in Refs. 2 and 3. QDs form occasionally in split-gate devices near the pinch-off conditions in the presence of disorder, and some of them exhibit the Kondo effect.7,8 The devices were cooled using a dilution refrigerator and magnetic fields were applied parallel to the plane of the 2DEG. A standard lock-in technique with an excitation voltage of 10 $\mu$V and a frequency of 18 Hz was used to measure $dI/dV_{sd}$ applying a bias voltage $V_{sd}$ between the source and drain electrodes.

Figure 2(a) shows the gate voltage $V_g$ dependence of the conductance of one of the devices. The conductance decreases with decreasing $V_g$, and several conductance peaks appear before the pinch-off, indicating formation of a QD at the constriction. The transport properties of the device exhibit behavior typical of the Kondo effect in a QD. (1) The conductance shows an increase with decreasing temperature $T$ at a conductance minimum in the $dI/dV_{sd}$ curve and the opposite temperature dependence at the neighboring conductance minimum [Fig. 2(a)]. (2) A pronounced ZBCP in the $dI/dV_{sd}$ curve develops with decreasing temperature below 700 mK [Fig. 2(b)]. The ZBCPs appear as a bright colored ridge in the gray-scale plot of $dI/dV_{sd}$ as a function of $V_g$ and $V_{sd}$ [Fig. 2(c)]. (3) The $dI/dV_{sd}$ spectrum splits into two peaks under in-plane magnetic fields $B$ [Fig. 2(e)], accompanied by the splitting of the $dI/dV_{sd}$ ridge [Fig. 2(d)]. The peak-to-peak voltage $V_{p-p}$ between the split $dI/dV_{sd}$ peaks is almost independent of $V_g$ as expected from the theory of the Kondo effect in a QD. The value of $V_{p-p}$ increases with increasing $B$, showing an agreement with theoretically expected values of $V_{p-p} = 2|g^*|\mu B/e$ with $|g^*| = 0.44$ for GaAs, as marked by triangles in Fig. 2(e).

We find that remarkable hysteresis appears in the $dI/dV_{sd}$ spectra under magnetic fields; the $dI/dV_{sd}$ curve of the positive scan ($dV_{sd}/dt > 0$) appears above that of the negative scan ($dV_{sd}/dt < 0$) as shown in Fig. 2(c). The $dI/dV_{sd}$ curves shown in Fig. 2(e) are obtained by scanning $V_{sd}$ between $-500 \mu$V and $+500 \mu$V in positive and negative directions at a rate of 5 $\mu$V/s. The hysteresis appears in a particular range of $V_{sd}$ between the $dI/dV_{sd}$ peaks at each magnetic field. The hysteresis in the $dI/dV_{sd}$ spectrum indicates involvement of a phenomenon with a long relaxation time in the mechanism of electron transport.

Figure 3(a) shows representative data describing the temporal change in $dI/dV_{sd}$ at $B = 2.88$ T after a rapid change of $V_g$ and $V_{sd}$ from the pinch-off condition to ($V_g$, $V_{sd}$) = (−0.65 V, −40 $\mu$V) on the slope of the $dI/dV_{sd}$ peak. The value of $dI/dV_{sd}$ increases continuously over a period of 150 s. The long time required to change $dI/dV_{sd}$ in Fig. 3(a) is in accordance with the typical time scale for building up a DNP in GaAs, suggesting the occurrence of the DNP in the present QD.

Relevance of the nuclear spins to the hysteresis and the slow evolution of $dI/dV_{sd}$ is confirmed by the following NMR spectroscopy measurement. We apply continuous waves of radio-frequency (rf) magnetic fields using a single-turn coil wound around the device. The frequency $f$ of the rf-magnetic field is scanned after the saturation of $dI/dV_{sd}$ at ($V_g$, $V_{sd}$) = (−0.65 V, −40 $\mu$V). As shown in Fig. 3(b), the value of $dI/dV_{sd}$ decreases when the frequency matches the NMR frequencies of $^{75}$As and $^{69}$Ga and $^{71}$Ga are also obtained (not...
shown). The observed decreases in $dI/dV_{sd}$ correspond

to the decreases in the nuclear spin polarization due to

the absorption of rf-magnetic fields at the NMR frequen-

2cies. Thus, the changes in the nuclear spin polarization

are detected by measuring $dI/dV_{sd}$ under the Kondo ef-

tect regime as expected [Fig. 1(b)].

Because $dI/dV_{sd}$ reflects changes in the nuclear spin

polarization, the slow increase of $dI/dV_{sd}$ in Fig. 3(a)

is interpreted as the development of the DNP in the

present QD. The possibilities of the electron heating or

the drift of the gate voltages are ruled out from the or-

igin of the hysteresis and the slow evolution of $dI/dV_{sd}$

because of the absence of the hysteresis at $B = 0.00 \text{T}$.

The induced DNP modifies the $dI/dV_{sd}$ spectra through

the Zeeman energy, changing the peak-to-peak voltage to

$V_{n-p} = 2|g^*|\mu_B(B + B_N)/e$. Because the DNP de-

velops during the scans of $V_{sd}$ in the $dI/dV_{sd}$-$V_{sd}$ mea-

surements in Fig. 2(e), the resultant difference in the nuclear

spin polarization at the same $V_{sd}$ causes the hysteresis in

the $dI/dV_{sd}$-$V_{sd}$ curves. We think that coexistence of

the DNP and the Kondo effect is allowed because the

electron-nuclear flip-flop process of the DNP does not

interfere with the formation of the Kondo singlet signif-

icantly due to the small characteristic energy for the

flip-flop process ($\sim 1 \text{~meV}$) compared to that for the

Kondo effect ($k_B T_K \sim 80 \mu \text{eV}$).

The above described mechanism of the hysteresis is

based on the sensitivity of the $dI/dV_{sd}$ spectra to the

changes in the Zeeman energy. Therefore the conditions

for the hysteresis to occur is expected to be related to

the appearance of the Kondo effect. Figure 4(a) shows

the hysteresis $\Delta$, defined as the difference in $dI/dV_{sd}$ be-

tween the positive and negative scans of $V_{sd}$ shown in

Fig. 2(e). To discuss the conditions for the hysteresis to

occur, we introduce the area $S$ of the hysteresis by inte-

grating the hysteresis $\Delta$ over a range $-200 \mu \text{V} \leq V_{sd} \leq 200 \mu \text{V}$. The area $S$, which is zero at $B = 0.00 \text{T}$, in-

creases with increasing $B$ and becomes saturated above $B = 2 \text{T}$ [Fig. 4(b)]. The dependence of $S$ on $V_g$ [Fig. 4(c)]

reveals that the hysteresis is remarkable in a range of

$V_g$ where the Kondo effect appears ($-0.67 \text{~V} < V_g <

-0.64 \text{~V}$). The values of $S$ are almost zero at the other
gate voltages. With increasing temperature $T$ under a

fixed magnetic field, the hysteresis becomes small and

the value of $S$ decreases as shown in Figs. 4(d) and 4(e).
The hysteresis almost disappears at around $T = 360$ mK, which is considerably lower than the onset temperature of the Kondo effect (about 700 mK). The temperature where the hysteresis disappears is close to the temperature where the splitting of the $dI/dV_{sd}$ spectrum becomes unclear [Fig. 4(d)]. These observations show that the hysteresis is one of the unique features of the Kondo effect in the magnetic fields and that the differential conductance under the Kondo effect regime is sensitive to the changes in the effective magnetic field $B + B_N$.

The effect of the DNP on the $dI/dV_{sd}$ spectra can be seen more directly in the following pump-probe measurement. The dashed and solid curves in Fig. 5(a) show the $dI/dV_{sd}$-$V_{sd}$ curves at the same $V_g$. The dashed curve is obtained immediately after relaxing the nuclear spin polarization under the pinch-off condition and the solid curve is obtained after the DNP pumping at $(V_{g,pump}, V_{sd,pump}) = (-0.650 \, V, -320 \, \mu V)$ for $t_{pump} = 600$ s. The peak-to-peak voltage $V_{p-p}$ after the DNP pumping is smaller than that without the DNP pumping. With increasing the period of the DNP pumping $t_{pump}$, $V_{p-p}$ decreases gradually, as shown in Fig. 5(b). The gradual change in $V_{p-p}$ with increasing $t_{pump}$ gives an additional evidence for the occurrence of the DNP. The shift $\Delta V_{p-p}$ measured from the value of $V_{p-p}$ without the DNP pumping allows us to evaluate the effective magnetic field $B_N$ using a proportional relation $\Delta V_{p-p}/V_{p-p} = B_N/(B + B_N)$, which is yielded based on an assumption $V_{p-p} = 2|g^*|\mu_B(B + B_N)/e$. A representative value of $\Delta V_{p-p} = -17.9 \, \mu V$ obtained from the difference between the dashed and solid curves in Fig. 5(a) corresponds to $B_N = -0.26$ T. This value is equivalent to the nuclear spin polarization of 4.9 % using the expression for $B_N$ in GaAs obtained in Ref. [21].

Figures 5(c)-5(e) show the dependence of $\Delta V_{p-p}$ on the DNP pumping voltages $(V_{g,pump}, V_{sd,pump})$. The values of $\Delta V_{p-p}$, which are proportional to $B_N$, are almost zero at around $V_{sd,pump} = 0$, while large values of $|\Delta V_{p-p}|$ are obtained at $|V_{sd}| > 100$ $\mu V$. In addition, the sign of $\Delta V_{p-p}$, which indicates the DNP polarity, strongly depends on the DNP pumping voltage. The negative $\Delta V_{p-p}$ $(B_N < 0)$ is remarkable at $V_{g,pump} = -0.650 \, V$ with $V_{sd,pump} < 0$ [Fig. 5(c)], while the positive $\Delta V_{p-p}$ $(B_N > 0)$ is remarkable at $V_{g,pump} = -0.650 \, V$ with $V_{sd,pump} > 0$ [Fig. 5(e)]. At the gate voltage between them $(V_{g,pump} = -0.655 \, V)$, both polarities appear depending on the sign of $V_{sd,pump}$ [Fig. 5(d)].

The optimum values of $|V_{sd,pump}|$ for the DNP pumping are larger than the $|V_{sd}|$ values for the $dI/dV_{sd}$ peaks. Moreover, the $V_{sd,pump}$ range where the DNP is achieved does not coincide with the $V_{sd}$ range where the hysteresis is observed. Both facts suggest that the mechanism for the DNP pumping is not directly related to that of the Kondo effect. In addition, the dependence of the DNP polarity on the sign of $V_{sd,pump}$ indicates that the present QD is asymmetric and that the asymmetry plays an important role in the DNP pumping mechanism. We think that the positive $B_N$ could be attributed to the electron spin relaxation in the QD. When adding an electron to the empty QD, both up-spin and down-spin states are allowed. If the added electron is in the down-spin state, it can relax to the up-spin state in the QD before going to the electrode. The down-to-up flips of electron spins cause to decrease the nuclear spin polarization $\langle I_z \rangle$ via the electron-nuclear flip-flop process of the hyperfine interaction, building up the positive $B_N$ in Fig. 5(c). Because of the asymmetric couplings between the QD and the electrodes, the added electron probably goes through the QD before relaxing its spin when $V_{sd,pump} < 0$. The negative $B_N$ in Fig. 5(c) indicates involvement of the up-to-down flips of electron spins in the QD. At the present stage, we do not have a comprehensive explanation why the up-to-down flips become dominant as the gate voltage is increased. Further studies will be needed to fully understand the DNP pumping mechanism.
the QD can be studied through the electron spin fluctuation, the electron spin dynamics in the pinch-off regime are emphasized by the following nuclear spin relaxation rate $1/T_1$ measurements. Because $1/T_1$ is enhanced by the electron spin fluctuation, the electron spin dynamics in the QD can be studied through the $1/T_1$ measurements. The decay curve of nuclear spin polarization is obtained by the following procedure: First, the DNP is pumped at $(V_g, V_{sd}) = (-0.650 \text{ V}, -320 \mu\text{V})$. Then, the nuclear spin polarization is relaxed under a certain voltage condition $(V_{g_{\text{temp}}}, V_{sd_{\text{temp}}})$ for a given time $t_{\text{relax}}$. Finally, the change in the nuclear spin polarization is read out by monitoring $dI/dV_{sd}$ at $(-0.650 \text{ V}, 0 \mu\text{V})$. By repeating this procedure with different periods $t_{\text{relax}}$, we obtain the decay curve of the nuclear spin polarization as shown in the inset of Fig. 6(a). The value of $1/T_1$ is evaluated by fitting the decay curve to the form $dI/dV_{sd} = a + b \exp(-t_{\text{relax}}/T_1)$.

Figure 6(a) shows the dependence of $1/T_1$ on $V_{g_{\text{temp}}}$ at $B = 2.88 \text{ T}$ for $V_{sd_{\text{temp}}} = 0 \mu\text{V}$. The relaxation rate $1/T_1 = 4.4 \times 10^{-3} \text{ s}^{-1}$ for $V_{g_{\text{temp}}} = -0.70 \text{ V}$ (the QD is depleted during the relaxation) is in the same order as the reported $1/T_1$ values in GaAs. The relaxation rate increases as $V_{g_{\text{temp}}}$ approaches the conductance peaks (Fig. 6(b)). We think that electron spin fluctuations introduced by the electron tunneling cause the increase in $1/T_1$. The small values of $1/T_1$ at around $V_{g_{\text{temp}}} = -0.66 \text{ V}$ can be attributed to the suppression of the electron spin fluctuation due to the Zeeman effect. When a bias voltage $V_{sd_{\text{temp}}}$ is applied during the relaxation (Fig. 6(c)), the values of $1/T_1$ do not increase largely at $|V_{sd_{\text{temp}}}| < 40 \mu\text{V}$. $1/T_1$ becomes large at $|V_{sd_{\text{temp}}}| > 40 \mu\text{V}$ associated with the increase in $dI/dV_{sd}$ (Fig. 6(d)). The increase in $1/T_1$ is probably related to the electron transport through the Kondo resonance state. We think that the $1/T_1$ measurement using the DNP technique is useful to understand spin properties of the Kondo effect under the non-equilibrium regime.

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FIG. 6: (color online). (a) Dependence of $1/T_1$ on $V_{g_{\text{temp}}}$ for $V_{sd_{\text{temp}}} = 0 \mu\text{V}$ and $B = 2.88 \text{ T}$. Inset shows a representative decay curve of the nuclear spin polarization at the pinch-off condition $(V_{g_{\text{temp}}}, V_{sd_{\text{temp}}}) = (-0.70 \text{ V}, 0 \mu\text{V})$. (b) $dI/dV_{sd}$- $V_g$ curve at $V_{sd} = 0 \mu\text{V}$, $V_{sd_{\text{temp}}} = -0.66 \text{ V}$, and $B = 2.88 \text{ T}$. (c) Dependence of $1/T_1$ on $V_{sd_{\text{temp}}}$ for $V_{temp} = -0.66 \text{ V}$. (d) $dI/dV_{sd}$- $V_{sd_{\text{temp}}}$ curve at $V_{sd} = -0.66 \text{ V}$. 

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