Nuclear-Spin-Induced Oscillatory Current in Spin-Blockaded Quantum Dots

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We show experimentally that electron transport through GaAs-based double quantum dots can be affected by ambient nuclear spin states in a certain regime where transport is blocked in the absence of electron spin flip. Current through the dots oscillates in time with a period up to 200 s depending on magnetic field. Oscillation is quenched by application of a continuous wave ac magnetic field which can induce nuclear magnetic resonance in ^{71}Ga or ^{69}Ga. A possible mechanism for dynamically polarizing the nuclear spins is proposed.

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The prospect of spintronic and quantum computing applications have made the nuclear spin degree of freedom in semiconductor nanostructures the subject of intensive studies [1]. Hyperfine coupling between electron and nuclear spins, for instance, are instrumental in the implementation of nuclear spin qubits within a semiconductor system. Heretofore, experimental examination of hyperfine coupling between heterostructure embedded electrons and nuclei has been mostly restricted to the quantum Hall systems [2]. Recently, hyperfine interaction involving electrons in quantum dots has received considerable attention, with the emergence of numerous theoretical works, concerning, for example, electron spin decoherence [3] and coherent coupling of multiple nuclear spins via interaction with dot electrons [4]. However, experimental results on hyperfine interaction in quantum dots are quite limited [5]. In this Letter we present for the first time clear experimental evidence based on an NMR study of hyperfine interactions in a double quantum dot with a well-defined electron configuration. We observe unexpected time dependent oscillation of a "leakage current" in the spin-blockade (SB) regime, and crucial contributions of nuclear interaction to this current are confirmed by application of an rf magnetic field in resonance with ^{71}Ga and ^{69}Ga nuclear spin splittings. This is thus the first demonstration of electrically detected NMR in quantum dots. We propose a tentative model, based on the electronic structure, wherein leakage current proceeds via spin flip coupling to the nuclei and dynamically polarized nuclear spins are accumulated in a certain external magnetic field range.

We have recently observed a novel SB effect due to Pauli exclusion using a device such that two quantum dots are weakly connected in series between source and drain contacts. The source-drain voltage V_S and gate voltage V_G are tuned so that four accessible electronic configurations are coupled via *irreversible* single electron tunneling as indicated in Fig. 1(a), A—D [6]. Beginning with A, where one electron with either spin up or spin down is already trapped in the lowest orbital state of the right dot [7], electron injection to the left dot from the left lead produces a two-electron spin triplet B or singlet C state with comparable probability. If the singlet state is formed, A is recovered after the second electron tunnels out of the double dot via D; i.e., the spin singlet where the lowest orbital state of the right dot is doubly occupied. However, once the triplet B is formed, A cannot be recovered without a spin flip or cotunneling transition. The spin lifetime is much larger than the tunneling time [8]. Thus, since B will sooner or later be formed, electron transport is blocked. Second order cotunneling processes and/or the spin scattering from B to C can give the leakage current [6].

These previous experiments, and those discussed herein, are based on a vertical double dot device consisting of a gated submicron pillar of a triple barrier remnant tunneling structure composed of two 8 nm thick $Al_{0.22}Ga_{0.78}As$ outer barriers, a 6 nm thick $Al_{0.22}Ga_{0.78}As$ center barrier, and two 12 nm thick $In_{0.05}Ga_{0.95}As$ wells, as schematically shown in Fig. 1(b) inset [6,9]. For comparison, a similar device, but having a 7.5 nm thick center barrier, is also prepared. A black line in Fig. 1(b) shows the current, I, flowing through the double dot versus V_S, measured at 1.8 K. V_G is fixed at the Coulomb peak at $V_G = 0.05$ V (lower right inset). Then electrons transit through the two-electron states in the double dot. The SB region appears in the V_S range 2–6 mV, where a small leakage current, $I \sim 1$ pA, is observed. This implies that the triplet B has a lifetime of $e/I \sim 100$ ns (e, elementary charge), which is much longer than the inelastic electron tunneling time ($\sim$ a few ns).

We set V_S in the SB region of Fig. 1 and measure the leakage current as a function of dc magnetic field B_{dc} applied horizontally. In this field direction the field-induced shift of the orbital energy can be safely neglected. Indeed the I-V_S characteristic in Fig. 1(b) does

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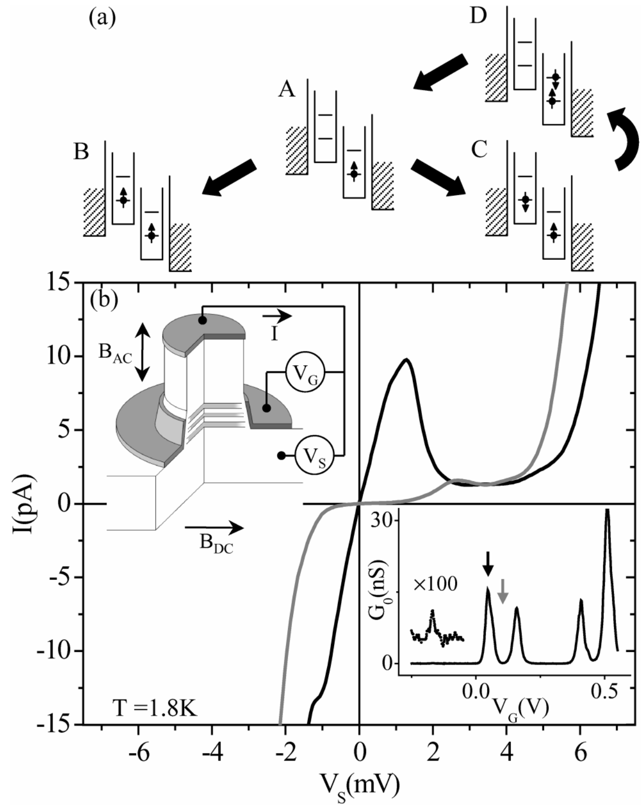

FIG. 1. (a) Accessible electric configuration A–D in spin-blockade region. (b) Current (I) voltage (V_S) characteristic measured at $T = 1.8$ K, zero magnetic field. Black and gray lines at V_G's are indicated by black and gray arrows in the right inset. In the spin blockade region for $2 < V_S < 6$ mV the system is in a spin triplet state B. Left inset: schematic of vertical double dot devices. Directions of dc and ac magnetic fields are indicated. Note that, for the black line, for small V_S, transitions between A–D become *reversible*, hence the system does not show spin blockade. For the gray line, the two-electron Coulomb blockade is present for $|V_S| < 1$ mV, and spin blockade for $V_S > 1$ mV.

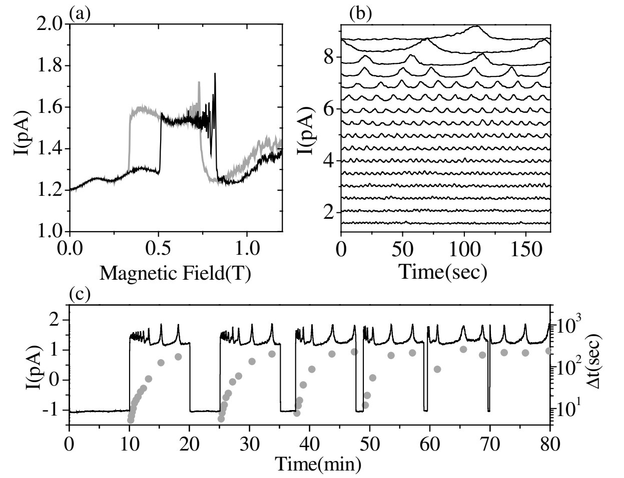

FIG. 2. (a) Magnetic field (B_{dc}) dependence of the leakage current at $V_S = 3.0$ mV, in the middle of the spin-blockade region, as a function of in-plane magnetic field for sweep up (black) and sweep down (gray). Detailed positions of the step and largest fluctuations depend on the B_{dc} sweep rate and values of V_S and V_G. (b) Leakage current evolving with time measured for fixed magnetic fields of $B_{\mathrm{dc}} = 0.70$ to 0.85 T with 0.01 T step for the curves from bottom to top. Each curve is vertically offset by 0.5 pA for clarity. (c) Transient behavior of the oscillatory current. V_G set at the gray arrow in Fig. 1(b), right inset. V_S switched from 3.0 to -1 mV, where the Coulomb blockade is almost lifted and a small current of ~ -1 pA flows. After dwelling for 300, 150, 75, 36, and 18 s outside the SB region we switch V_S back to 3.0 mV. The peak spacing Δt (right axis) is plotted.

not change significantly up to $B_{\mathrm{dc}} = 8$ T and the leakage current is in the 1 pA range. This contrasts with the ease of *vertically* applied field as discussed in Ref. [6] where the shift of the second lowest orbital eventually lifts the SB. Figure 2(a) shows the leakage current taken at $V_S = 3.0$ mV with a constant field sweep rate of 2 min/T. As the magnetic field initially increases, the current is nearly constant for $B_{\mathrm{dc}} < 0.5$ T, but then rises sharply at $B_{\mathrm{dc}} \sim 0.5$ T. Fluctuations emerge and increase in amplitude with increasing B_{dc} up to $\sim$0.87 T, and then leakage suddenly collapses for $B_{\mathrm{dc}} > 0.9$ T. A similar characteristic, but shifted to lower field, is observed when sweeping the field downward. The width of this hysteresis loop, $\sim$0.2 T in Fig. 2(a), shrinks for slower B_{dc} sweep rate and saturates at $\sim$0.15 T for sweep rates below 1 h/T. Similar characteristics, i.e., a step followed by fluctuations and hysteresis, are observed at different (V_S, V_G) within the SB region. For any B_{dc} field in the current fluctuation regime ($0.6 \sim 0.87$ T), the current shows periodic oscillations as a function of time [Fig. 2(b)]. Oscillation is in fact depending only on time and both the period and amplitude of the current oscillations increase with B_{dc}, reaching maximal period, $\sim$200 s, and amplitude, $\sim$0.4 pA, near 0.87 T. These oscillations last with no definite damping for 15 h or longer. We observe no clear periodic oscillations after the current decreases to the low level for $B_{\mathrm{dc}} > 0.87$ T. Note that in Fig. 2(b) only variations of the current slower than our measurement time constant of $\sim$1 s can be detected. We observe no significant thermal dependence of this oscillatory behavior over temperatures from 1.8 to 0.3 K, provided that V_S and V_G remain well within the SB region.

These characteristics are observed not merely in one sample. I-B_{dc} characteristics similar to that shown in Fig. 2(a) are observed for four double dot samples showing spin blockade: three samples with a 6 nm center barrier and one sample with a 7.5 nm center barrier. A leakage current step, oscillations, and hysteresis are observed in a smaller B_{dc} range (step at $\sim$0.3 T and maximal oscillations at $\sim$0.6 T) in the device with a 7.5 nm center barrier. The other two 6 nm center-barrier samples show a similar step although in some samples the oscillations are less clear.

To investigate how the current oscillations initially evolve with time in the SB region, we have performed the transient measurements as shown in Fig. 2(c). First we set $B_{dc} = 0.87$ T and $V_S = 3.0$ mV, where the current oscillations have a nearly maximal period ($\sim$200 s) and amplitude ($\sim$0.4 pA). Then we temporarily move V_S out of the SB region and back again after calibrated dwell times. This measurement clearly shows that a time scale of $\sim$5 min is needed for both establishing and erasing the oscillatory behavior. Such a slow response is exactly what we would associate with the nuclear spin system, which has an unusually long longitudinal decay time constant, $>$10 min at low temperatures [2,5].

As is well known from studies employing NMR techniques, nuclear spin effects can be investigated with an ac magnetic field. We use a three-turn coil of 3 mm diameter located 0.5 mm above the device to apply a vertical ac magnetic field, B_{ac}, to the double dot [see Fig. 1(b) inset], and measure the change in the oscillatory current. Figure 3(a) shows the data measured for various frequencies of B_{ac} at $B_{dc} = 0.85$ T. A strong reduction in both the oscillation period and amplitude is observed when the B_{ac} frequency coincides with the ^{71}Ga nuclear spin resonance [Fig. 3(b), [10]]. The resonance frequency changes linearly with B_{dc} [inset of Fig. 3(b)]. It is interesting to note that the oscillations are consistently described if we phenomenologically assume that larger nuclear polarization leads to larger period and amplitude of the current oscillations. Overall, nuclear polarization grows with increasing magnetic field [Fig. 2(b)], gradually grows (decays) by turning on (off) the SB [Fig. 2(c)], and resonantly decays under the NMR condition [Fig. 3(a)].

For a horizontally applied magnetic field, Zeeman splitting of the triplet state B [Fig. 1(a)] will be the only

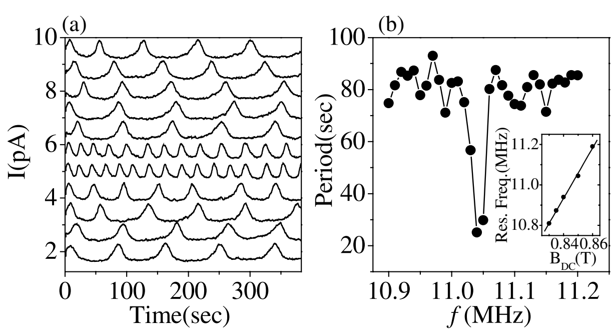

FIG. 3. (a) Current oscillations under ac magnetic field in the frequency range of $f = 11.00$ to 11.10 MHz with 0.01 MHz step for the curves from bottom to top. Each curve is vertically offset by 0.75 pA for clarity. Amplitude of the ac voltage applied to the coil is adjusted so that the effects of neither the heating nor "pumping current" due to the stray electrical coupling to the coil are reasonably small. (b) Time period of the current oscillations measured for various frequencies of the ac magnetic field (f). Inset: dc magnetic field dependence of the resonance frequency observed in the oscillation period.

change in the electronic states. In the field range discussed here ($<$1 T) the Zeeman shift, as well as a small zero-field exchange splitting between the triplet B and the singlet C are of order 10 μeV [11]. This is much smaller than typical energy scales, $\sim$5 meV, in Fig. 1(a), such as on-site Coulomb energy, orbital level spacing, and detuning of those levels from the Fermi levels of the leads. Thus first order tunneling processes described in Fig. 1(a) are not modified. Any change in the leakage current must be due to changes in the cotunneling rates and/or the spin scattering rate.

We can exclude most possible spurious effects that may account for the observed step, hysteresis, and oscillations since these are never observed outside the SB regime in any sample. For instance, the current-voltage characteristic in Fig. 1(b) shows negative differential conductance (NDC) and it is well known that spontaneous oscillations of current may occur depending on circuit parameters of the system such as capacitance and conductance. However, the oscillations in Fig. 2 are not confined to the NDC region but also occur in the other SB regions, where differential conductance is positive. On the other hand, no current oscillations are observed for V_S and/or V_G outside the SB region even where NDC is seen due to, for example, the resonance between the lowest and second lowest orbital [6]. Bolometer detection of NMR, where absorption of the resonant ac field by the nuclei is measured via an increase in temperature, is a conventional technique [12]. Quantum dots can be a sensitive bolometer because of their large temperature dependence in certain circumstances. However, the NMR response we observe is not due to the bolometer effect because we never observe the NMR response outside the SB region, where the current might be more temperature sensitive. Finally, thermal nuclear polarization due to the nuclear Zeeman energy is probably negligible in our measurement (10^{-4} at 1.8 K and 0.85 T). For these reasons, it seems reasonable that the nuclei (number of order of 10^5) in *quantum dots* are *dynamically polarized* at a certain B_{dc} field in the SB region [2].

Oscillation of the nuclear spin demands consideration of complex feedback mechanisms of the nuclear spin on the electronic state, which we discuss below. Here we discuss a tentative model for only the B_{dc} field dependent dynamic polarization of nuclei in the SB double dot. Transition from the triplet B [Fig. 1(a)] to the singlet state C can be induced by hyperfine flip-flop scattering with the nuclei in the quantum dots. However, this process is significantly suppressed due to the discreteness of electron energy in quantum dots [3]. A small but finite tunnel coupling and exchange interaction between dots lifts the degeneracy of the singlet and triplet states. The energy separation between these states is calculated to be a few tens of μeV for the 6 nm center-barrier sample [11]. Zeeman splitting of the triplet states reduces this separation and ultimately induces degeneracy of the $S_Z = +1$

triplet and the singlet states. Around this field, the hyperfine flip-flop scattering from $S_Z = +1$ triplet to the singlet states should be substantially favored over those from $S_Z = 0$ or -1. Thus a nuclear spin will be flopped from "down" to "up" rather than "up" to "down." The long relaxation time of nuclear spins at low temperatures causes the flopped nuclear spins to steadily accumulate, leading eventually to dynamic polarization of the nuclei. Assuming an electron g factor of -0.44 the degeneracy of the $S_Z = +1$ triplet and singlet states occurs at $B_{dc} = 0.4$–2 T. This agrees with the experimental B_{dc} field where a step and oscillations are observed. A sample with wider center barrier has a smaller tunnel coupling and correspondingly smaller zero-field splitting between triplet and singlet. Thus the degeneracy is induced at lower B_{dc}, quantitatively consistent with our observation of the 7.5 nm center-barrier sample.

Regarding feedback from the polarized nuclei to the electron, the Overhauser effect due to dynamic nuclear polarization is known to affect electron spin and the transport characteristics [2]. However, this effect simply detunes the singlet-triplet crossover and cannot cause the oscillatory behavior. The results suggest the presence of more complicated feedback from the polarized nuclei to the electron spin [13,14]. One recent calculation, incorporating the evolution of the parallel and transverse components of the nuclear spin, has shown that the coupled electron-nuclear spin system can exhibit instability near the singlet-triplet crossover [13].

In conclusion, we have studied magnetic field effects on a small leakage current of order 1 pA in the spin-blockaded vertical double quantum dot system. In the presence of a dc in-plane magnetic field of 0.7–0.87 T we have observed oscillations of the leakage current with a period as long as 200 s. Application of an NMR rf field significantly quenches the oscillatory behavior, indicating the presence of the hyperfine flip-flop scattering and polarized nuclear spin state in the quantum dots.

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[11] Y. Tokura (unpublished). The energy separation is calculated to be 10–40 μeV depending on V_S. The triplet state B [Fig. 1(a)] lies below the singlet B. The *ground state* is singlet state D. The level broadening due to dots-lead couplings is estimated to be ~ 2 μV from the current negative V_S in Fig. 1(b), and is smaller than the energy separation.

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