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To cite this article: T Kodera et al 2009 J. Phys.: Conf. Ser. 150 022043

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Pauli-Spin Blockade in a Vertical Double Quantum Dot
Holding Two to Five Electrons

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Abstract. We use a vertical double quantum dot (QD) to study spin blockade (SB) for the two- to five-electron states. SB observed for the two- and four-electron states is both assigned to Pauli exclusion with formation of a spin triplet state, and lifted by singlet-triplet admixing due to fluctuating nuclear field. SB observed for the five-electron state is caused by combined Pauli effect and Hund’s rule. We observe a hysteretic behavior of the SB leakage current for up and down sweep of magnetic field, and argue that SB and its lifting by hyperfine interaction are subtle with the spin configuration and modified depending on the inter-dot detuning and number of electrons.

1. Introduction
Pauli-spin blockade (P-SB) is one of the most striking spin-related tunneling phenomena observed for coupled double quantum dots (QDs) [1]. P-SB occurs when a spin triplet state is formed with one electron in the lowest energy state of each QD, because subsequent electron transfer from one QD to the other is prohibited by Pauli exclusion. Finding of P-SB in double QDs has opened up a new approach to probe the spin effect such as hyperfine interaction (HF) with lattice nuclei, because P-SB can be lifted by spin flip-flop interactions [2] and singlet-triplet admixing by statistical fluctuation of nuclear field [3]. In addition, P-SB has been utilized to detect coherent rotation of single electron spin [4] and prepare SWAP operation of coupled electron spins [5]. All of the experiments on P-SB have been performed on a genuine system of two-electron states in coupled QDs to date.

Here we study the effect of P-SB in a vertical double QD holding not only two electrons but also four and five electrons. The electronic configuration is well defined in vertical QDs [6], and this allows us to discuss the states involved in P-SB in a rigorous way. In general, the effect of P-SB is expected to appear for a system of more electrons, although the states involved can be more...
complicated. In the two-electron P-SB, we observe lifting of P-SB due to fluctuating nuclear magnetic field [3, 7, 8]. We observe P-SB for the four- and five-electron states as well, and assign the four-electron P-SB to standard Pauli exclusion and the five-electron P-SB to combined Pauli exclusion and Hund’s rule. For the five-electron P-SB, we observe a hysteretic behavior of the leakage current for sweeping up and down the magnetic field. This behavior is a signature of flip-flop HF, and depends on the source-drain voltage ($V_{sd}$). We finally argue that P-SB and its lifting by HF for the double QD are subtle with the spin configuration and modified, depending on the inter-dot detuning and number of electrons.

2. Experimental
The weakly coupled vertical double QD studied here is embedded in a ~400nm diameter circular mesa composed of a Al$_{0.22}$Ga$_{0.78}$As /In$_{0.05}$Ga$_{0.95}$As /Al$_{0.22}$Ga$_{0.78}$As / In$_{0.05}$Ga$_{0.95}$As /Al$_{0.22}$Ga$_{0.78}$As triple barrier substructure (TBS) (see Inset to Figure 1) [9]. The thickness of the AlGaAs barrier and InGaAs well is 8nm and 12nm, respectively. Two QDs are coupled in series, surrounded by a single Schottky gate electrode, which is capacitively coupled to two QDs and can tune their electrochemical potential almost equally. The electrochemical potential of the upper QD (QD1) is higher than that of the lower QD (QD2) due to the slightly trapezoidal shape of the mesa as shown in the inset to Fig. 1. The difference of the electrochemical potential between the two QDs is called initial offset ($\delta$). The intra-dot Coulomb energy of QD1 ($U_1$) is larger than that of QD2 ($U_2$) due to the same reason. The confinement potential of each QD is well approximated as a two-dimensional harmonic potential and the eigenstates are described as the single-particle Fock-Darwin states [10]. We call the lowest (second) eigenstate 1s (2p) state. We measure the current ($I_{sd}$) between the source and drain contact by applying a source-drain voltage ($V_{sd}$) and gate voltage ($V_g$) at 0.3 K. The detuning ($\Delta$) between the two QDs is varied with $V_{sd}$. The conversion factor ($x$) between the increment of $V_{sd}$ and that of $\Delta$ is 0.25 in this device, which is estimated from experiments of photon assisted tunneling with application of microwave. Magnetic field ($B_p$) is applied perpendicular to the QD plane.

3. Results and discussions
Figure 1 (a), and right inset show the color-scale plot of the Coulomb diamond plot or measured, and simulated $|I_{sd}|$ vs. $V_{sd}$ - $V_g$, respectively. The Coulomb blockade (CB), and transport region are shown in white and blue, respectively. The total electron number $N$ in the double QD is indicated in the figure. The CB diamond is not well closed because there are no available states for tunneling through both QDs due to the inter-dot detuning $\Delta (=\delta$ at $V_{sd} = 0$ V). ▲ shows the vertical line, which appears when the energy level is aligned between the two QDs and ● shows the kink, which occurs when an electron escapes from one QD due to the rise in the electrostatic potential by addition of an extra electron onto the other QD. The positions of ▲ and ● in Fig. 1 are well reproduced by the simulation using constant interaction model. At the positions of A-C, on the other hand, the experiment shows blockade of $I_{sd}$ but the simulation does not. In the simulation, we only consider the electrostatic effect such as intra-dot Coulomb energy $U_1$ and $U_2$, inter-dot Coulomb energy $V_s$, and $\delta$, with no spin effect. Therefore, the deviation between the experiment and the simulation at A-C are assumed to be due to the spin effect or P-SB for the two or more ($N$ = 4 and 5) electron states. Note P-SB has been exclusively discussed for the two-electron state.

Main features marked by A-C are explained using the potential diagrams A-C in the insets. The region A located to the right of the $N$=2 CB is well assigned to the $N$=2 P-SB [1]. The leakage current is 0.4 pA in the P-SB region. The blue line parallel to the $V_g$ axis located just to the right of the region A indicates resonance between a triplet state $T(1s, 1s)$ having one electron in the lowest $1s$ state of each QD and a triplet state $T(0, 1s2p^+)$ having two electron in the $1s$ and $2p^+$ states of QD2 [11]. The $2p$ states in QD2 are split by ~0.8 meV into the $2p^+$ and $2p^-$ states. The energy separation is derived from the two resonance peaks as shown in Fig. 1, and arises from lifting of $2p$ state degeneracy by the anisotropy and anharmonicity of the confinement potential.
Figure 1. (a) Color scale plot of $|I_{sd}|$ vs. $V_g$ and $V_{sd}$. White indicates low current, $|I_{sd}| < 0.5$ pA. The potential diagrams of A-C regions are shown in the insets A-C. Right inset: Simulated current $|I_{sd}|$ plotted in the $V_g$ vs $V_{sd}$. Here we use the realistic values of $U_1 = 5.15$ meV, $U_2 = 4.85$ meV, $V = 2.50$ meV, $\delta = 2.80$ meV, and $x = 0.25$. Left inset: Schematic diagram of the vertical double QD. (b) $S_{2p^+2p^-}$ and $T_{2p^+2p^-}$ states of QD2 for the $(N_1,N_2)=(1,4)$ regime are schematically shown.

Figure 2. (a)-(c) Magnetic field $B_p$ dependence of the leakage current in the P-SB region A-C. (c) shows the $V_{sd}$ dependence of $I_{sd}$ vs $B_p$ from $V_{sd} = -1.0$ mV (bottom) to $-3.5$ mV (top) with a $-0.25$ mV step. Each curve is offset by 0.1 pA to the top.

For the $N=4$ state in the region B, the transition from $(N_1, N_2) = (1, 3)$ to $(2, 2)$ is suppressed because of P-SB, as schematically shown in the inset B. $N_1$ ($N_2$) is the number of electrons in QD1 (QD2). This blockade is similar to that of $N=2$ P-SB, because we can consider two electrons in the 1s state of QD2 as a closed core shell. Then, we assume that transition from the $(n_1, n_2) = (1, 1)$ to $(2, 0)$ is suppressed, where $n_1$ ($n_2$) is the electron number in the valence shell of QD1 (QD2). For the $N=5$ state in the region C, the transition from $(N_1, N_2) = (1, 4)$ to $(2, 3)$ is prohibited due to the combination of Hund’s rule [6] and Pauli exclusion, as shown in the inset C. Here we also concentrate on the valence shells, and discuss the transition from $(n_1, n_2) = (1, 2)$ to $(2, 1)$. The energy state of QD2 is then apparently either a triplet state $T_{2p^+2p^-}$ (QD2) having one electron in each 2p± state or a singlet state $S_{2p^+2p^+}$ (QD2) having two electrons in the 2p+ state as schematically shown in Fig. 1 (b). Exactly speaking, we should consider the three-electron state in the double QD, but just for simplicity we refer the singlet and triplet states in QD2 (or QD1), because the inter-dot exchange energy is very small as described in the next paragraph. In our device, $T_{2p^+2p^-}$ (QD2) state is thought to be almost degenerate with $S_{2p^+2p^+}$ (QD2) state because the 2p+ - 2p- splitting of ~0.8meV is comparable to the exchange energy, which is previously reported for vertical single QDs [6]. In this case, either $S_{2p^+2p^+}$ (QD2) or $T_{2p^+2p^-}$ (QD2) can be populated by tunneling of an electron onto QD2 from the right lead. If the $S_{2p^+2p^+}$
(QD2) is populated, an electron tunnels from QD2 to QD1 to form $S_{1s1s}$ in QD1. On the other hand, once $T_{2p+2p}$ (QD2) is populated, subsequent electron transfer from QD1 to QD2 is blocked by Pauli exclusion.

Figure 2 (a)-(c) show the magnetic field $B_p$ dependence of the leakage current $I_{sd}$ measured in almost the center of region A-C in Fig. 1, respectively. In Fig. 2 (a) measured for region A, as the $B_p$ field is initially increased, the current gradually decreases and finally becomes saturated above $\approx 40$ mT. This current reduction is due to the lifting of admixed $S(1s,1s)$ and $T(1s,1s)$ by the fluctuating nuclear field [3,7,8]. Figure 2 (b) for region B looks similar to Fig. 2 (a). This is reasonable, because the blockade mechanism is basically the same. In Fig. 2 (c) for region C, we observe a small but definite hysteric loop for sweeping up and down $B_p$. As $B_p$ increases, $I_{sd}$ is initially constant and gradually increases, and then has a sharp decrease (arrow) to the initial current level. A similar but somewhat shifted hysteretic loop is observed when the measurement is performed at various $V_{sd}$ points. The hysteretic behavior is a signature of P-SB lifting mediated by HF as previously revealed the detuning dependence of $B_{sw}$ for two-electron vertical double QDs [8]. The mechanism is yet unclear but these results indicate that P-SB and its lifting by HF are subtle with spin configurations and also tunable with various parameters including inter-dot detuning and number of electrons.

4. Conclusion
We study lifting of P-SB in a vertical double QD holding not only two electrons but also four and five electrons. Magnetic field dependence of the leakage current in the P-SB region shows the singlet-triplet mixing effect for the two and four electron states. We observe a hysteretic behavior of the P-SB leakage current for the five-electron state for sweeping up and down the magnetic field. The mechanism of this hysteretic behavior is not yet clear but P-SB and its lifting by HF are subtle with the spin configurations, number of electrons in the double QD, and inter-dot detuning.

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