Direct observation of the orbital spin Kondo effect in gallium arsenide quantum dots

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Besides the spin Kondo effect, other degrees of freedom can give rise to the pseudospin Kondo effect. We report a direct observation of the orbital spin Kondo effect in a series-coupled gallium arsenide (GaAs) double quantum dot device where orbital degrees act as pseudospin. Electron occupation in both dots induces a pseudospin Kondo effect. In a region of one net spin impurity, complete spectra with three resonance peaks are observed. Furthermore, we observe a pseudo-Zeeman effect and demonstrate its electrical controllability for the artificial pseudospin in this orbital spin Kondo process via gate voltage control. The fourfold degeneracy point is realized at a specific value supplemented by spin degeneracy, indicating a transition from the SU(2) to the SU(4) Kondo effect.

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The Kondo effect is an essential many-body physics process that describes the screening of a spin impurity by a sea of conduction electron spins. This is actively investigated at the frontier of condensed matter physics; for example, in the Kondo topological insulator samarium hexaboride [1,2], heavy fermion superconductivity [3,4], and metallic islands [5]. An electron confined in a quantum dot (QD) can be taken as the smallest magnetic impurity with spin $1/2$ [6–8]. Many-body interactions between this impurity and conduction electrons in the reservoir yield a spin Kondo singlet formed at the Fermi energy through virtual spin-flip processes. Besides spin, a pseudospin Kondo effect might also occur depending on the pseudospin-flip process [9–11], especially in specific materials with an intrinsic pseudospin degree of freedom, for example, a valley in silicon field-effect transistors [12] or orbital in a carbon nanotube [13,14]. Specifically, if pseudospin can be tuned to degeneracy as well as spin, a higher symmetric SU(4) Kondo effect is expected, which has attracted much theoretical attention [10,15,16].

GaAs semiconductor QDs, with strong parameter controllability, provide an extraordinary platform to study these problems. Nevertheless, corresponding experimental studies are still relatively few. This is partially due to the fact that an intrinsic pseudospin is absent in GaAs so that the exploration should be carried out in multiple dots with artificial pseudospin. Keller *et al.* [16] performed an experimental exploration in a lateral GaAs double QD (DQD) structure. Detailed agreement between experimental data at the orbital spin fourfold degeneracy point and numerical renormalization group calculations suggests an emergent SU(4) symmetry. However, there is a more intuitive method to demonstrate the spin-orbital and SU(4) Kondo effect avoiding sophisticated theory background: (1) complete three resonance spectra with definite one net spin impurity as predicted in theory [9]; (2) continual evolution of spin and pseudospin resonances with eventual fourfold degeneracy under an electric field; (3) Kondo-type behavior when varying the temperature and magnetic field.

Here, we study an orbital spin Kondo effect in a series-coupled GaAs DQD device and obtained a series of experiment results which increases our knowledge about Kondo effect in QDs system. It contains the following: (1) In a region where only one net electron is involved, three Kondo resonances with degenerated spin and undegenerated pseudospin are observed firstly in source-drain bias spectra. (2) We demonstrated a pseudo-Zeeman effect under electrical field by gate control. (3) We realized a controllable convergence of three Kondo resonances with a higher Kondo temperature, which is a striking feature that indicates a transition from the SU(2) to the SU(4) Kondo effect.

The artificial designable and electrical tunable pseudospin impurities in GaAs QDs overcome the reliance on specific materials and make GaAs QDs a promising candidate to explore the exotic Kondo effect based on both spin and pseudospin impurities, like the non-Fermi liquid state, in multidots structure [17–19].

The series-coupled GaAs DQDs device was fabricated on a $GaAs/Al_{0.3}Ga_{0.7}As$ heterostructure with a two-dimensional electron gas (2DEG) lying 100 nm below the surface with an electron density of 2.3×10^{11} cm^{-2} and mobility of 1.5×10^5 $cm^2\,V^{-1}\,s^{-1}$. It was placed in a dilution refrigerator with a base temperature of 40 mK and parallel magnetic field. Differential conductances were measured at the source terminal "s" and drain terminal "d" [Fig. 1(f)].

Spin configuration in DQDs. An precondition for the SU(4) Kondo effect is that only one net spin impurity is involved. In Fig. 1(e), N electrons in the left QD and M electrons in the right QD are represented as $(N, M)x$ (x = I, II, III, IV stands for the four charging regions). One arrow represents a single spin impurity (odd N or M) in a QD, while double arrows represent antiferromagnetically coupled electrons [even N or M; Fig. 1(e)]. It is convenient to determine the charging number

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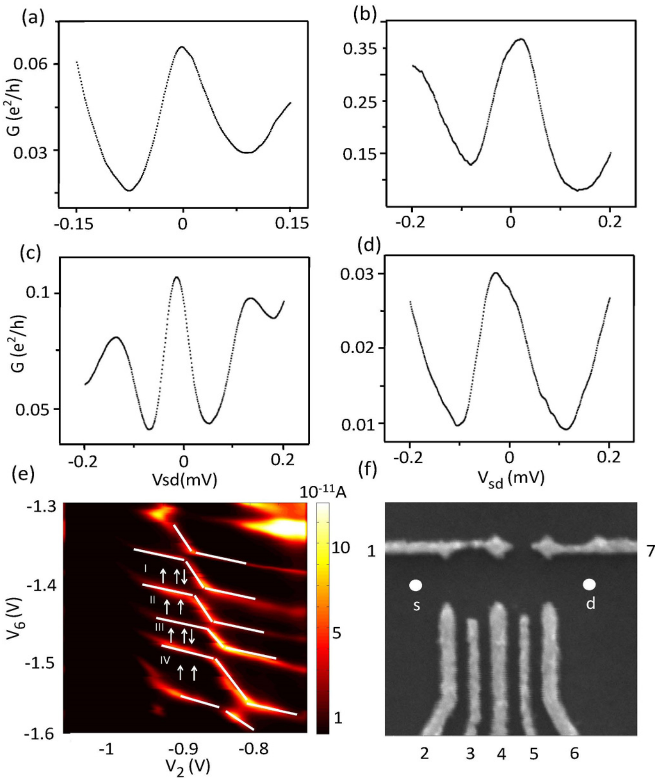

FIG. 1. (a–d) dI/dV curves in the region I–IV with obvious ZBP at $V_{sd} = 0$ [three peaks in (c)]. (e) Honeycomb diagram with the corresponding spin states. The single arrow represents a single-spin impurity (net spin = 1/2) and double arrows represent double antiferromagnetically coupled spins (net spin = 0). (f) SEM image of our typical series-coupled DQD device defined by gates 1–7. White dots labeled "s" ("d") represent the source (drain).

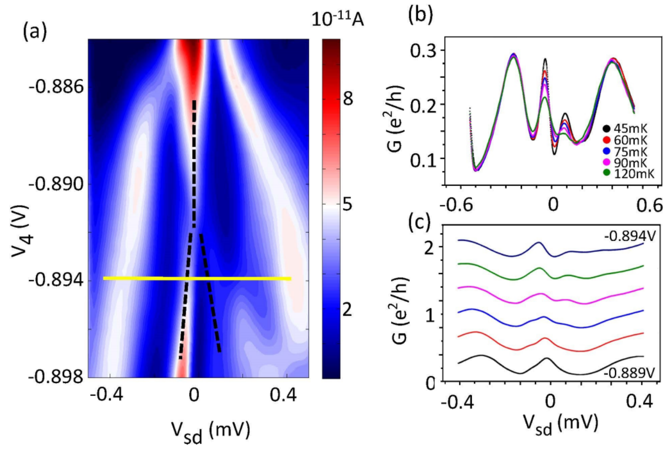

FIG. 2. (a) dI/dV curves while sweeping V_4 in region II. A single peak clearly splits into two peaks (indicated by black dashed lines), suggesting that a QPT occurs at about −0.892 V. (b) Temperature dependence of the Kondo peaks [as the yellow solid line indicates in (a)] at 45, 60, 75, 90, and 120 mK. (c) dI/dV curves extracted from (a) with different V_4 (–0.889 to –0.894 V with step of 1 mV). A splitting from a single Kondo peak to two peaks occurs by varying V_4.

by counting the Coulomb peaks but the corresponding spin configuration is not trivial [20]. We identified the spin status by two steps: Firstly, we demonstrated that N is odd; secondly, we demonstrated that $(N,M)_{\rm II}$ is (odd, odd).

A zero-bias peak (ZBP) is the most well-known signature for the Kondo effect if N or M is odd [Figs. 1(a)–1(d)]. In Figs. 1(a)–1(d), V_1, V_3, V_4, V_5, and V_7 were kept as –1.1, –0.06, –0.9, –0.32, and –1.1 V while $(V_2, V_6)_x$ are $(-0.883, -1.407\ {\rm V})_{\rm I}$, $(-0.869, -1.449\ {\rm V})_{\rm II}$, $(-0.859, -1.485\ {\rm V})_{\rm III}$, and $(-0.836, -1.5543\ {\rm V})_{\rm IV}$ for four continuous regions with ZBP at 40 mK and zero magnetic field. This demonstrated that N is odd while M is still unknown.

Then we demonstrate that $(N,M)_{\rm II}$ are both odd. Figure 2(a) is the differential conduction in region II with V_4 and source-drain voltage V_{sd} at 40 mK and zero magnetic field (here $V_3 = 0$ and other gate voltages are unchanged). By carefully tuning the gate voltages, the ZBP occurs when $V_4 = -0.888$ V (illustrated by the black dashed line). If both N and M are odd, V_4 changes. In other words, when the coupling between QDs is tuned, a quantum phase transition (QPT) [21–23] with ZBP splitting into two peaks may occur. It is observed at about $V_4 = -0.89$ V. The dI/dV curves with bias voltage from $V_4 = -0.889$ to –0.894 V with steps of 1 mV are drawn in Fig. 2(c) for more obvious illustration. Furthermore, the two peaks' heights decrease with temperature in Kondo behavior, as illustrated in Fig. 2(b) [at $V_4 = -0.892$ V, yellow line in Fig. 2(a)].

The above evidence supports our hypothesis that $(N,M)_{\rm II}$ are both odd (more discussion and data under magnetic field are included in the Supplemental Material [24]). Therefore, we reach the conclusion that region III has a net spin of 1/2 ($N = $ odd, $M = $ even). In this case, the spin and orbital are independent, leading to a fourfold degeneracy SU(4), where the Kondo effect is theoretically predicted to occur.

Orbital spin Kondo effect in the single-spin impurity region. A degenerated spin impurity with undegenerated (gap of Δ) orbital impurity will induce a hybrid orbital spin Kondo effect with signature of multiple conductance enhancement peaks, such as in carbon nanotubes [14] or silicon nanowires [12]. In region III ($V_1 - V_3 = -1.1, -0.929, -0.12$ V; $V_5 - V_7 = -0.6, -1.439, -1.1$ V), a three-peaks pattern [indicated by white dashed lines in Fig. 3(a)] is observed under 40 mK and zero magnetic field. One peak is fixed at $V_{sd} = 0$ and the other two appear symmetrically at V_{sd} of about $\Delta = \pm 0.1$ mV. The ZBP corresponds to a pure SU(2) spin flip. Peaks at $\Delta = \pm 0.1$ mV are typically associated with a transition where an electron enters one orbital state in one QD and an electron jumps out from another orbital state [Fig. 3(b)]. By the finite element analysis we evaluated the electrical field at the DQD geometric center in the 2DEG layer obtaining $E = \alpha V_4 + \beta$, where $\alpha = 8.0 \times 10^5\ {\rm m}^{-1}$ and $\beta = 13.9 \times 10^5\ {\rm V\,m}^{-1}$. As V_4 moves to a more negative voltage, Δ becomes small. The dI/dV curves with bias voltage from $V_4 = -0.872$ to –0.88 V with steps of 2 mV are drawn in Fig. 3(c) for more obvious illustration under electric field. This result is consistent with orbital Kondo states because Δ can be modulated by tuning the coupling strength between QDs so that the orbital pseudo-Zeeman effect is demonstrated.

Next, we investigated the temperature and magnetic field dependence in region III to better understand this process. Figures 3(b)–3(e) show the temperature dependence of the

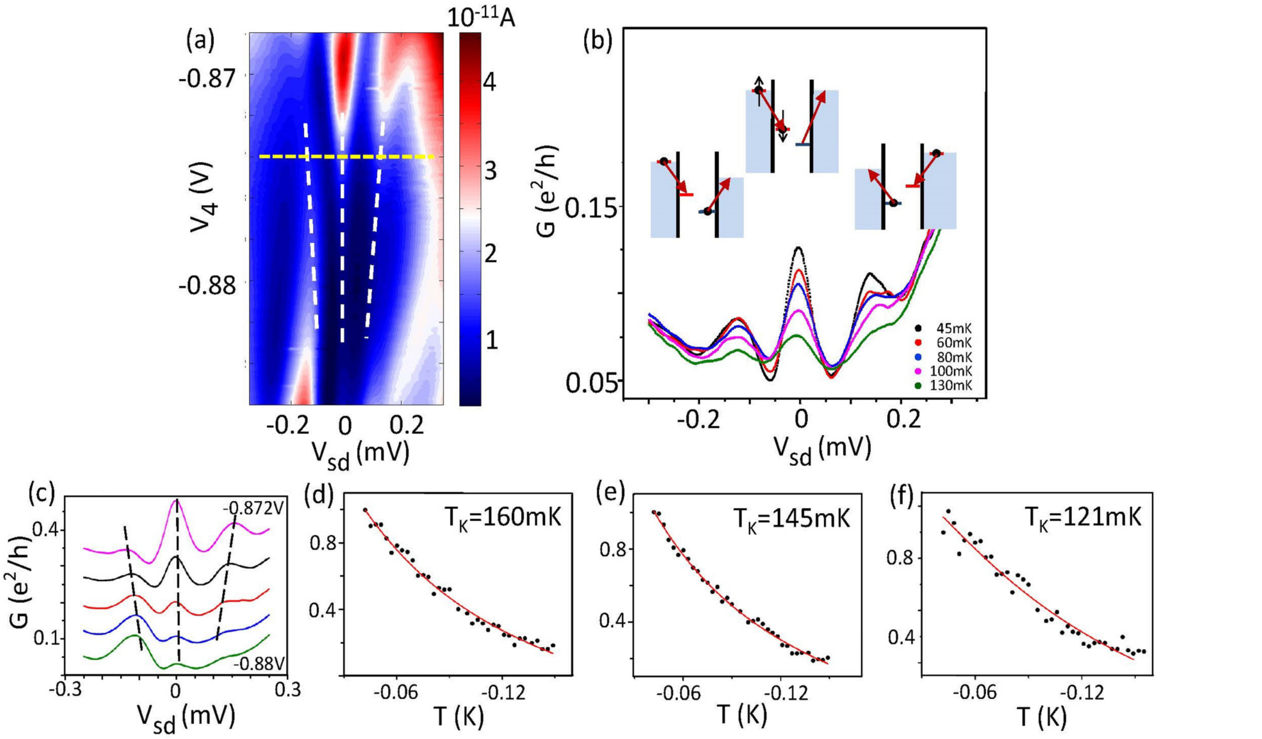

FIG. 3. (a) Three peaks were observed in the Coulomb blockade region, as indicated by dashed white lines. A pseudo-Zeeman effect is observed as a convergence of the outer two peaks when V_4 changes (electrical field $E = \alpha V_4 + \beta$, where $\alpha = 8.0 \times 10^5\,\mathrm{m}^{-1}$ and $\beta = 13.9 \times 10^5\,\mathrm{V\,m}^{-1}$). (b) dI/dV curves extracted from the dashed yellow lines at different temperatures from 45 to 130 mK. The three insets illustrate the corresponding processes. For the left peak, one electron (without spin) is transported to a QD with an orbital state change (from the red to the blue state). In the red state, an electron in the source tunnels into the QD. Then the former blue-state electron in the QD tunnels into the drain, where an orbital flip occurs. The same process also occurs for the other two peaks. (c) dI/dV curves extracted from (a) with different V_4 (–0.872 to –0.88 V with steps of 2 mV). The dashed lines indicate the converging of the three Kondo peaks. (d–f) Fitting curves for three peaks with corresponding Kondo temperature.

three Kondo peaks [indicated by the yellow horizontal dashed line in Fig. 3(a) at $V_4 = -0.874$ V]. We clearly observed a conductance suppression with increasing base temperature from about 40 to 200 mK. The conductance can be described by the following phenomenological relationship [12,25]:

$$G(T) = G_0\left(\frac{T_K^{*\,2}}{T_K^{*\,2} + T^2}\right)^s + G_1, \qquad (1)$$

where $T_K^* = T_K/(2^{1/s} - 1)^{1/2}$, $G(T)$ is the conductance normalized to the respective $(dI/dV)_{\max}$ at about 40 mK, G_1 is a constant and G_0 is the conductance at zero temperature, and $s = 0.22$. The fitting results for the three peaks are plotted in Figs. 3(d)–3(f) with corresponding T_K of 160, 145, and 121 mK, respectively. The uniform temperature scaling demonstrates that both the orbital-state flip and non-orbital-state flip are caused by the Kondo effect.

We then investigated the resonance dependence of this device under a magnetic field. Figure 4 shows a color map plot of dI/dV under different V_{sd} and B at 40 mK. At about $B = 1.5$ T, the central peak splits into two peaks, as indicated by the black lines closest to $V_{\mathrm{sd}} = 0$, with a Zeeman splitting gap of $2g^*\mu_B B$. Here, the g^* factor is about 0.59 (compare with 0.44 for bulk GaAs). Furthermore, we observed that the orbital Kondo resonances do not split and the bias voltage is nearly unchanged (right peak nearly vanished under 1.5 T), at least

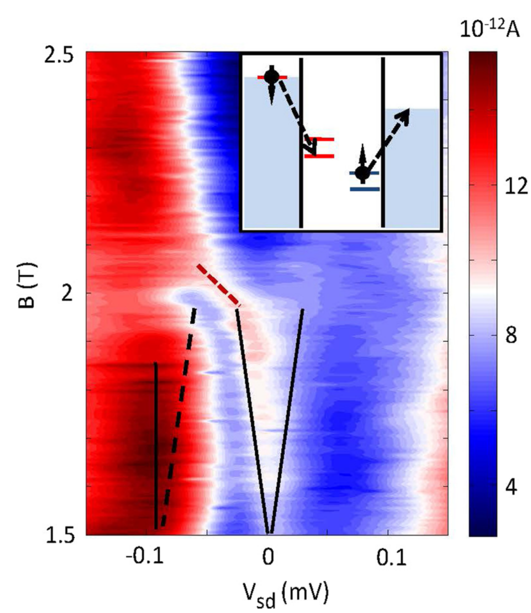

FIG. 4. Black lines indicate the spin-flip and orbital-flip evolution. The dashed black line depicts an invisible but possible process with an energy of $-\Delta + g^*\mu_B B$. An unusual conductance enhancement region with a different slope with B, which is expected theoretically (as illustrated in the inset), is indicated by the brown dashed line.

when $B < 2\,\mathrm{T}$, as indicated by the black lines at $V_{\mathrm{sd}} = -\Delta$. For $B > 2.5\,\mathrm{T}$, the splitting of the middle two peaks is strongly suppressed by the magnetic field. Under higher $B(>3\,\mathrm{T})$, all peaks disappeared.

Besides a pure spin-state flip, the orbital-state flip resonances at the bias $\pm\Delta$ are associated with different processes based on their evolution under magnetic field. The processes involving both an orbital and a spin flip induce an electron transition where an electron with spin up (down) enters one orbital state in one QD and an electron with spin down (up) jumps out from another orbital state (see Fig. 4 inset). In theory, these twofold flip processes can be recognized by the fact that the orbital resonances at $\pm\Delta$ split and shift to $\pm\Delta \pm g^*\mu_B B$ [13,26]. However, there are few experiments, partially because this higher-order process involves spin and pseudospin simultaneously. The black dashed line in Fig. 4 indicates the resonance peak corresponding to $-\Delta + g^*\mu_B B$ (not observed). Because Δ is relatively small (only about 0.1 meV) in our experiment, the two processes labeled by their V_{sd} position as $-\Delta + g^*\mu_B B$ and $0 - g^*\mu_B B$ can be close enough in energy such that state mixing and conductance enhancement are probable and expected around $V_{\mathrm{sd}} = -\Delta/2$. The black dashed line in Fig. 4 indicates the resonance peak corresponding to $-\Delta + g^*\mu_B B$ (not observed). This predicted unusual conductance enhancement was definitely observed, with different slopes with respect to B around $B = 2\,\mathrm{T}$, at $V_{\mathrm{sd}} = -\Delta/2$ indicated by the brown dotted line in Fig. 4.

The SU(2) to SU(4) Kondo effect transition. If the spin and the orbital states are set into degeneracy, a SU(4) Kondo effect involving two kinds of impurities is expected to occur. In experiment, this process presents directly as convergence of the three peaks into one peak, as illustrated in Fig. 5(a) by white dashed lines, which is a fingerprint corresponding to the transition from SU(2) to SU(4) symmetry [9,27]. Tuning V_3 from -0.12 to -0.06 V while scanning V_4 caused the three peaks to converge at $V_4 = -0.902\,\mathrm{V}$ [indicated by the yellow dashed lines in Fig. 5(a)] with a sudden conductance enhancement.

Theory predicts a higher T_K for the SU(4) Kondo effect [6,28]. In the SU(2) region in Fig. 5(a), the extracted T_K is 145 mK, which is in agreement with our claim that T_K is uniform in the SU(2) Kondo region, even though here we change V_3 only slightly compared to the change of T_K in Fig. 3. At the converged single peak, T_K ($V_4 = -0.902\,\mathrm{V}$) is 256 mK [Fig. 5(b)]. It is 76% larger than 145 mK, which agrees with the theory predicted enhancement.

One aspect that should be discussed is that region III is a charge region with even/odd electrons which in principle induces that only one net electron is evolved into this Kondo process and denies other possible explanations, like spin singlet and $S = 1$ Kondo via triplet state [29]. The other aspect is that though the Kondo temperature above is smaller than tunneling coupling strength (a rough estimated value of $\sim$100 μeV; see discussion in the Supplemental Material [24]), when V_4 is varied, the coupling strength could be smaller which promises that the SU(4) Kondo effect emerging becomes possible.

The magnetic field dependence of the single peak is complicated [Fig. 5(c)]. Compared with the rapid suppression of

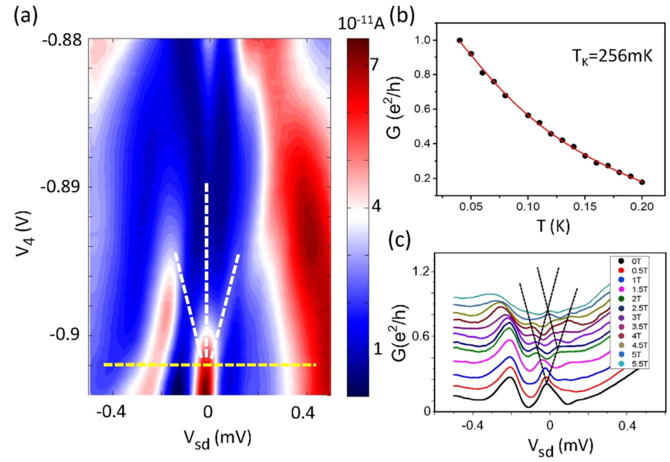

FIG. 5. (a) Slight tuning of V_3 causes the orbital states to converge at about $V_4 = -0.902\,\mathrm{V}$ (electrical field $E = \alpha V_4 + \beta$, where $\alpha = 8.4 \times 10^5\,\mathrm{m}^{-1}$ and $\beta = 14.1 \times 10^5\,\mathrm{V\,m}^{-1}$). As a result of the degeneration of the spin and orbital degrees of freedom, an enhanced single Kondo peak emerges, which indicates an SU(4) symmetry. (b) A higher Kondo temperature fitting result of 256 mK at $V_4 = -0.902\,\mathrm{V}$. (c) Kondo peaks splitting under different magnetic fields of 0–5.5 T in steps of 0.5 T. Peaks are offset to aid visualization. Unexpected complicated twofold splitting, as illustrated by black dashed lines, occurs when the magnetic field is larger than 2.5 T. This multiple peak splitting directly suggests higher symmetry than SU(2).

the three peaks with increasing magnetic field, the resonances derived from the SU(4) peak can survive under a field as high as 5.5 T. Considering the parity of the spin Zeeman splitting and orbital pseudo-Zeeman splitting, three peaks are expected to occur, with the outer two corresponding to Zeeman splitting and the central one to an orbital SU(2) process when the spin degeneracy is entirely broken. However, in our experiment, a Zeeman-like splitting was only observed when $B < 3\,\mathrm{T}$. Moreover, when $B > 3\,\mathrm{T}$, unique behavior was observed as each branch splits again in a Zeeman-like way with the same Landé g factor, which should in principle induce a pattern of four peaks [illustrated by the dashed lines in Fig. 5(c), along with three of the four peaks observed].

In other systems, like carbon nanotubes [14], the Zeeman energy or pseudo-Zeeman energy provides relatively large splitting so that degeneracies are lifted entirely. The above data indicate that the Zeeman energy in GaAs is small while the pseudo-Zeeman energy is tuned by electric field so the orbital and spin index mixing may not be entirely prevented by a magnetic field of several teslas. This may be essential information to explain our unique results which exhibit higher symmetry than the usual SU(2).

In summary, we demonstrated an electrically tunable orbital-spin Kondo effect in a GaAs QD system by directly observing the predicted three Kondo peaks in a dI/dV spectrum. Furthermore, upon converging the orbital states, a single Kondo peak with higher T_K was observed, revealing an emerging SU(4) symmetry. We believe that our work lays a solid foundation for future study of complex Kondo states in multiple QD systems.

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