# Magnetic field dependence of the spin-$\frac{1}{2}$ and spin-1 Kondo effects in a quantum dot

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We study the magnetic field evolution of the spin-$\frac{1}{2}$ and spin-1 Kondo effects in a quantum dot formed from a single-walled carbon nanotube. In the spin-$\frac{1}{2}$ case, the energy of spin-resolved Kondo conductance peaks is proportional to magnetic field at high fields, contrary to recent reports. At lower fields, the energy falls below this linear dependence, in qualitative agreement with theoretical expectations. For even electron occupancy, we observe a zero-bias Kondo peak due to the degeneracy of the spin-1 triplet ground states. Tuning gate voltage within the same Coulomb diamond drives a transition to a singlet ground state. We also independently tune the energy difference between singlet and triplet states with a magnetic field. The Zeeman splitting thus measured confirms the value of the $g$ factor deduced from the high-field behavior of spin-$\frac{1}{2}$ Kondo.

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## I. INTRODUCTION

The Kondo effect—the screening of a local moment by a reservoir of conduction electrons—is perhaps the simplest many-body effect and hence holds a special place in the study of correlated-electron systems.[1] Although the Kondo effect has traditionally been studied in bulk metals containing magnetic impurities,[2] a recent resurgence of interest is spurred by the ability to measure transport through a single magnetic site: a quantum dot with an unpaired spin.[3] The tunability of this novel system has enabled tests of long-standing theoretical predictions, but behavior at finite magnetic field remains controversial and ripe for quantitative experimental study (as explicated below). Magnetic field is a particularly important tool for studying the Kondo effect: As in many correlated-electron systems, the many-body physics in this system stems from local spin fluctuations, to which magnetic field couples strongly.

Quantum dots formed from carbon nanotubes are particularly well suited to studying the Kondo effect in magnetic field. Advantages of nanotube-based dots relative to the gated GaAs-based dots in which Kondo effect has been more thoroughly studied include the following.

*High Kondo temperature*. The strength of the Kondo effect is characterized by the Kondo temperature $T_K$. Within a simple model, $T_K \sim \sqrt{\Gamma U} e^{\pi \epsilon_0(\epsilon_0+U)/\Gamma U}$, where $\epsilon_0$ is the energy of a localized state relative to the Fermi level, $U$ the charging energy, and $\Gamma$ the coupling to the leads.[4] The underlying energy scales, and hence the maximum achievable $T_K$, grow with decreasing dot size, with $T_K$ in the middle of the Kondo valley reaching several Kelvin for nanotube dots,[5] an order of magnitude higher than for gate-defined GaAs dots.[6,7]

*Large g factor*. The Landé $g$ factor—the strength of magnetic field coupling to the spin of the local site—is large and well defined in nanotubes ($g \approx 2$) (Refs. 8 and 9) compared to GaAs-based dots ($|g|=0.1$ to 0.44).[10] Thus, the Kondo temperature can be much larger than accessible temperatures but still smaller than Zeeman splitting at accessible magnetic fields.

*Pure Zeeman coupling*. The large $g$ factor and small area of nanotube quantum dots render orbital effects of magnetic field insignificant for magnetic field perpendicular to the tube axis. Magnetic field parallel to the tube axis does strongly affect orbital states,[11] so we apply field perpendicular rather than parallel to the tube axis to ensure that only the Zeeman effect is important.

In this paper, we present data on the magnetic field evolution of Kondo effect beginning from zero-field ground states with spins 0, $\frac{1}{2}$, and 1 on the local site, all in the same device. For spin-$\frac{1}{2}$ Kondo, we summarize the extensive theoretical literature and compare our data to representative models, showing that they agree qualitatively but not quantitatively. We also note previous experimental works which foreshadow our results. For integer spin, where less theoretical work has been done, we present our data together with a simple model which explains the main features of the data. We hope this will serve as a spur for further theoretical efforts.

## II. EXPERIMENT

Our device is a carbon nanotube connected to palladium source and drain electrodes.[12] It lies on a 500 nm thick surface oxide layer atop a highly doped Si substrate which acts as a gate. The source and drain are fabricated 250 nm apart using electron beam lithography, and the nanotube is 1 nm in diameter according to atomic force microscopy measurements. The differential conductance $G$ across the device is measured as a function of the bias voltage $V_b$, gate voltage $V_g$, and magnetic field $B$ in a $^3$He refrigerator using standard ac lock-in techniques.

## III. SPIN-$\frac{1}{2}$ KONDO

Measurements of $G(V_g, V_b)$ display Coulomb diamonds with striking horizontal features—peaks in $G$ at $V_b=0$—which signal the presence of the Kondo effect [Fig.

 

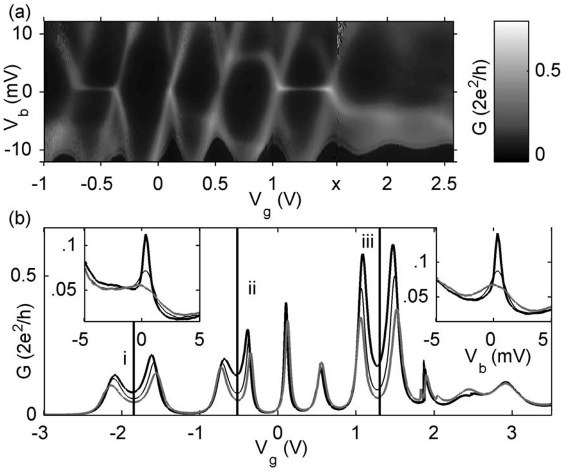


FIG. 1. (a) Differential conductance $G$ as a function of bias voltage $V_b$, showing zero-bias features for odd electron occupancy. Data to the right of the location marked "×" have been shifted to account for a random charging event. (b) Linear conductance as a function of voltage $V_g$ on a back gate, at $T \approx 317$ mK (black line), 1.8 K (fine black line), and 6 K (gray line). (Insets) Zero-bias features in valleys ii and iii at the same temperatures.

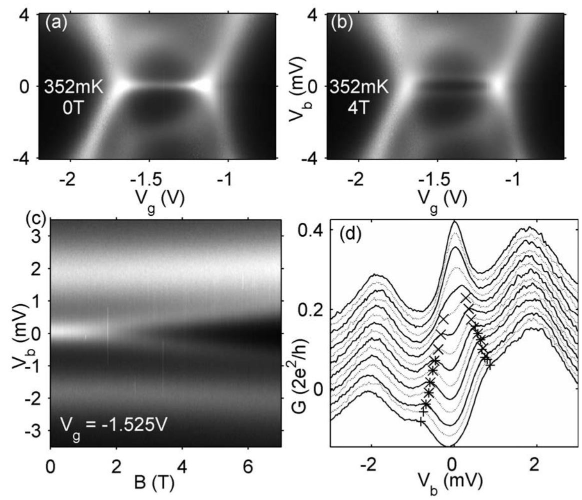


FIG. 2. (a) Kondo diamond at zero magnetic field. $T \approx 352$ mK. Black is low conductance and white high. (b) The same diamond at 4 T. (c) Evolution of the features in the middle of the valley in (a) and (b) with magnetic field. (d) Slices of the data in (a) in 0.5 T steps. (Our data are 20 times denser.) Successive curves are offset downward by $0.02 \times 2e^2/h$. Peak locations for the central Kondo peaks were obtained by fitting the data from (c) with two Lorentzians plus a field-independent background (+ symbols). A slightly different background is assumed for a similar fit at intermediate field points (× symbols). Between 4 and 6 T, the two versions of our fitting procedure produce nearly indistinguishable results [see also Figs. 3(a)–3(d)].

1(a)].[13] These zero-bias anomalies are more prominent at lower temperatures [Fig. 1(b), insets], as expected, and from their widths, we estimate the Kondo temperature to be 1.7, 1.8, and 1.5 K in the middle of valleys (i), (ii), and (iii).[14,15]

Upon application of a magnetic field, a Kondo zero-bias conductance peak with $T_K = 2$ K splits into the sum of two peaks at energies $\pm\delta$ corresponding to peaks in the densities of spin-up and spin-down states (Fig. 2), as predicted[16] and previously observed.[8,10,17,18] Naïvely, one might expect these peaks to occur at the Zeeman energy $\pm g\mu_B B \equiv \pm\Delta$, and indeed this is what we observe at high fields (see supplementary information[19] for details of how we extract peak energies and compare with theory). From $B = 4-7$ T $[g\mu_B B = (2.9-5.1)kT_K]$, we find linear splitting $2\delta[\mu V] = 240B[T] - 5 \pm 14$, corresponding to $g = 2.07 \pm 0.02$ (95% confidence) [Fig. 3(b)].[20] This linearity at high fields has been predicted theoretically,[16,21] and our value for the $g$ factor agrees well with previous work on nanotubes.[8,9]

At low fields, however, we find that the extracted peak energy falls below linear [Figs. 3(b) and 3(c)]. This is in qualitative agreement with the theoretical literature which predicts $\delta < \Delta$,[21–23] with $\delta$ reduced to $(2/3)\Delta$ as $B \to 0$ (Ref. 24) due to the attraction of the Kondo resonance to the Fermi level.[23]

The field at which this reduction disappears should therefore be related to the strength of the Kondo interaction. Despite broad theoretical agreement on the high- and low-field limits, descriptions of the crossover region differ. Here, we consider two representative theories due to Moore and Wen (MW)[21] and Costi.[23,25] MW's Bethe-ansatz calculation converges to the high-field result extremely slowly (logarithmically), predicting, for example, $\delta \approx 0.9\Delta$ for $g\mu_B B = 1000kT_K$ (Ref. 21) (this slow convergence may be an artifact of the single-spinon approximation for the electronic spectral function.) Costi, using a density matrix numerical renormalization group approach, predicts a more rapid crossover, with convergence to the high-field result around $g\mu_B B = 20kT_K$.[23] In contrast, our measured peak splitting crosses over to linearity by $g\mu_B B = 2.8kT_K$, a much lower field than predicted by either theory[26] but also a more intuitive result.

This can be seen in Fig. 3(c) where we compare our data to the Moore-Wen and Costi predictions: Our data have already saturated to the linear splitting value at 4 T, yet both theories predict this saturation only at much higher fields, off the scale of the plot (assuming $g = 2$). Another way to look at the contrast between theory and experiment is to normalize the predicted splittings to our data. (This is equivalent to assuming, unrealistically, $g = 2.9$ and 2.35 for MW and Costi, respectively.) One then sees that both theories predict a more gradual evolution to high-field behavior than we observe [Fig. 3(d)].

The splitting of the spin-$\frac{1}{2}$ Kondo peak in transport through a quantum dot has been measured in detail on two previous occasions. Cronenwett *et al.*[17] measure $\delta \sim g\mu_B B$ over a broad range of fields, but their assignment of peak positions is not precise enough to identify a 10% deviation such as we report in this paper. In measurements by Kogan *et al.*,[10] we note an apparent suppression of $\delta$ for $g\mu_B B < 1.1T_K$, though this was not how the data were interpreted by the authors. In both these measurements, $T \sim T_K/3$—compared to $T \sim T_K/6$ in the present work—and the higher relative temperature may have obscured the low-field suppression of $\delta$ that we can now observe.[21]

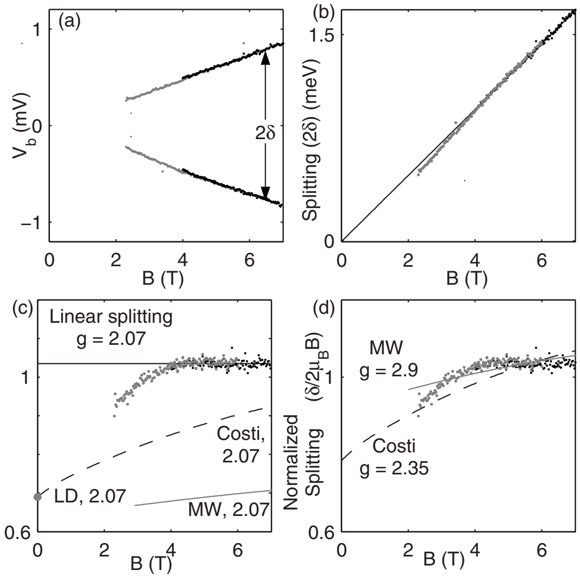


FIG. 3. (a) Peak positions in Fig. 2(c) obtained as described in Fig. 2(d). Black (gray) dots mark results from the high (intermediate) field fit. (b) Energy difference between peaks from (a). The line is a fit to the high-field points. (c) Splitting from (a) normalized by the naïve Zeeman energy—$\delta/2\mu_B B$ (black and gray dots). The gray line is the Moore-Wen prediction, the dashed line is the Costi prediction, the solid black line is the predicted high-field limit, and the gray dot marks the Logan-Dickens low-field prediction, all for $g=2.07$. (d) The data from (b) are reproduced (black and gray dots). The gray line is the Moore-Wen prediction and the dashed line is the Costi prediction, now using unrealistically large Zeeman coupling $g=2.9$ and 2.35, respectively, to attempt to match the data.

Another question of interest concerns the field at which splitting between the two spin-resolved resonances first becomes visible: Although these resonances, in principle, move away from zero energy even at very low fields, they initially overlap so that the measured Kondo peak does not visibly split into two until a critical field $B_c$. Costi had predicted[25] that at low temperatures ($T < T_K/2$, satisfied in our case), $g\mu_B B_c \approx kT_K$. We observe this splitting first at $B=2.3$ T, modestly higher than the predicted $B_c^{\mathrm{Costi}}=1.5$ T, and roughly where we can start to robustly fit peak positions. Previous work on GaAs dots also found the splitting occurring roughly at $B_c^{\mathrm{Costi}}$.[7]

## IV. SPIN-1 KONDO

We now turn our attention to Kondo effects with an even number of electrons on the dot (Fig. 4). Empirically, in quantum dots with even electron occupancy, the ground state is usually a singlet ($S=0$), particularly in the presence of strong tunnel coupling to leads.[27] However, when the exchange energy gained from aligning spins is larger than the energy difference between singlet and triplet states, the ground state will be a triplet ($S=1$).

In our dot, we observe for one particular even occupancy a zero-bias conductance peak which splits into peaks at finite bias as a function of gate voltage [Figs. 4(a) and 4(b)]. These

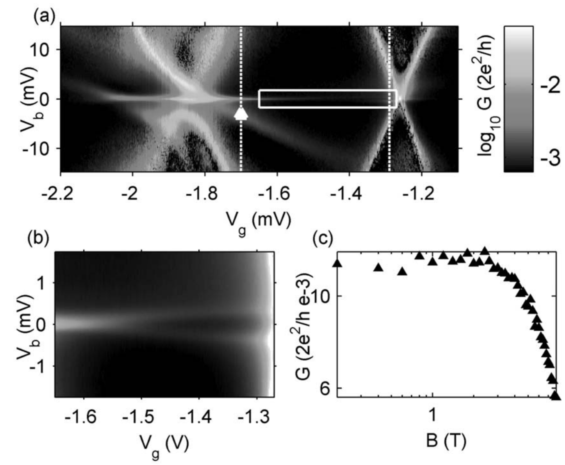


FIG. 4. (a) Zero-bias feature in an even Coulomb diamond (parity determined by careful study of ten diamonds on either side). $T \lesssim 250$ mK. (b) Higher resolution scan of the region in (a) bounded by the rectangular box. The data indicate a gate-induced transition between singlet and triplet ground states for the dot. (c) Magnetic field dependence of the conductance at the point marked by the white triangle in (a).

features can be understood as a transition between triplet and singlet ground states,[28] respectively, as follows.

On the left-hand side of the Coulomb diamond in question, the triplet is the ground state and correlated interactions with the leads arise from the degeneracy of the three triplet states. As the total spin on the dot is 1, interactions with two channels in the leads are required to fully screen this spin.[19,29] When couplings to the two leads are comparable, previous experiment and theory have shown two peaks in conductance at finite bias, flanking a zero-bias conductance dip: the "two-stage" Kondo effect.[30–33] Our contrasting observation of a simple zero-bias peak indicates strong coupling to only one channel so that the spin is underscreened.[28,34–36]

On the right-hand side of the Coulomb diamond, the ground state is a nondegenerate singlet and so conductance is low at zero bias; however, peaks appear at finite bias corresponding to the energy difference between singlet and triplet states.

This sort of gate-driven Kondo singlet-triplet transition at zero magnetic field has been reported only once before, in a GaAs dot.[28] The mechanism proposed there was the Stark effect, which is small for quantum dots in nanotubes. A different gate-driven spin transition has been observed in nanotubes[37] and was thought to be due to electron-electron correlations, which could play a role here too.

Magnetic-field-driven transitions of ground state spin in a quantum dot are more common than gate-driven transitions (cf. Refs. 36 and 38), but they usually stem from orbital physics. In contrast, we can apply a pure Zeeman field, allowing us to study the magnetic field evolution of the Kondo features on both sides of the gate-driven transition in a simple manner.

On the triplet (left-hand) side of the diamond, a magnetic field linearly splits the zero-bias peak into two finite-bias

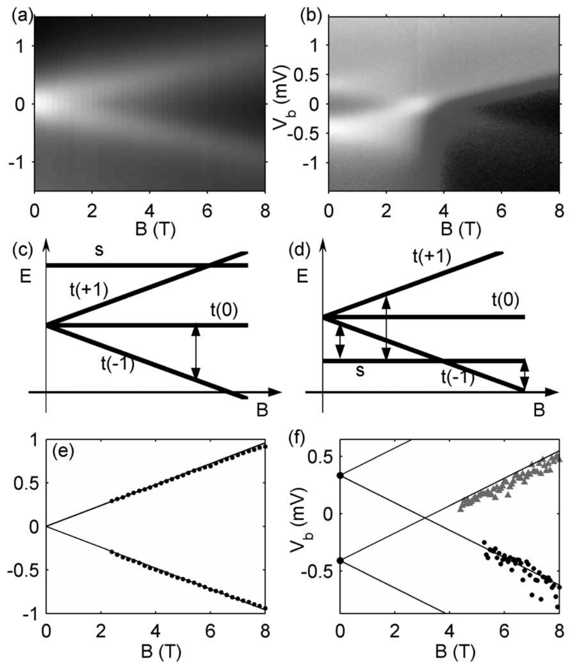


FIG. 5. [(a) and (b)] Magnetic field evolution of conductance versus bias voltage on the left (triplet) and right (singlet) sides of the diamond in Fig. 4(a). The gate voltages chosen have been marked with dashed lines in Fig. 4(a). [(c) and (d)] Schematic of singlet ($s$) and triplet ($t$) energy levels as a function of magnetic field in the configurations corresponding to (a) and (b), respectively. Double-headed arrows indicate transitions seen in our data. (e) Peak positions in (a) obtained by fitting two Lorentzians, corresponding to the transition shown in (c). The lines are $\pm g\mu_B B$, with $g=2.07$. (f) Analysis of data in (b). The black circles are peaks obtained using a simple peak-finding function. The gray triangles are the locations of steepest slope for the step. The lines are guides to the eye and have slope $\pm g\mu_B B$, with $g=2.07$, and mark the transitions shown in (d).

peaks [Figs. 5(a) and 5(e)]. The voltage of these peaks corresponds to the energy difference between the two lowest triplet states [Fig. 5(c)]. As with the spin-$\frac{1}{2}$ Kondo peak, the splitting here first occurs at slightly larger-than-expected field $B=1.8$ T $> B_c^{\text{Costi}}=1.1$ T.[25,39]

On the singlet (right-hand) side, the two finite-bias peaks each split with increasing field, with two of the resulting features meeting at zero bias at finite field and then splitting apart again [Fig. 5(b)].

This field evolution can be understood by considering finite-energy transitions between the singlet ($s$) and the three Zeeman-split triplet states $t(-1)$, $t(0)$, and $t(1)$, labeled according to their spin in the direction of the field [Fig. 5(d), cf. Ref. 40, Fig. 2(b)].

*$s \leftrightarrow t(-1)$*. The features moving toward zero bias with increasing field correspond to this transition, so the zero-bias crossing is when $t(-1)$ becomes degenerate with $s$. Beyond this crossing field, $t(-1)$ is the ground state, and we continue to see transitions between $t(-1)$ and $s$, but not the those between $t(-1)$ and $t(0)$ or $t(1)$. The latter is forbidden at lowest order because of the large change in spin; the former should be possible but would occur at very high bias.

*$s \leftrightarrow t(1)$*. The features starting at zero field that move to higher bias with increasing field mark this transition. These features are less pronounced, probably because of stronger decoherence at higher bias, and disappear around when $t(-1)$ becomes the ground state.

*$s \leftrightarrow t(0)$*. We do not clearly discern this transition, likely because the three overlapping peaks associated with singlet-triplet transitions form a plateau, in which only the edges of the plateau, associated with $t(-1)$ and $t(1)$, can be easily identified, while the middle peak remains hidden.

The transformation in Fig. 5(b) of the upper peak into a step after the crossing point can be understood as being caused by asymmetric coupling to the leads.[40]

We observe several additional strongly gate-sensitive features in our diamond plots [Fig. 4(a)], which we investigate in less detail. At least one of them appears to be Kondo-related—the conductance decays logarithmically with $B$ at high field and saturates to a constant value at low field [Fig. 4(c), see also supplementary information[19]].

## V. CONCLUSIONS

In conclusion, we have studied the magnetic field evolution of integer-spin and spin-$\frac{1}{2}$ Kondo effects in a quantum dot formed from a carbon nanotube. We carefully track a spin-$\frac{1}{2}$ Kondo zero-bias peak and find that the energy of its spin-resolved constituents is linear with field at high field, and sublinear at low field, qualitatively matching predictions. However, the crossover from sublinear to linear splitting is sharper than predicted by any existing theory and occurs at lower-than-expected field. We also demonstrate the independent gate and magnetic field tuning of integer-spin Kondo effects and find good quantitative agreement with a simple model using the $g$ factor previously extracted.

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[19] See EPAPS Document No. E-PRBMDO-76-065744 for details. For more information on EPAPS, see http://www.aip.org/pubservs/epaps.html.

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