Zero-field splitting of Kondo resonances in a carbon nanotube quantum dot

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We present low-temperature electron transport measurements on a single-wall carbon nanotube quantum dot exhibiting Kondo resonances at low temperature. Contrary to the usual behavior for the spin-1/2 Kondo effect we find that the temperature dependence of the zero bias conductance is nonmonotonic. In nonlinear transport measurements low-energy splittings of the Kondo resonances are observed at zero magnetic field. We suggest that these anomalies reflect interactions between the nanotube and a magnetic (catalyst) particle. The nanotube device may effectively act as a ferromagnetically contacted Kondo dot.

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Transport measurements on individual single-wall carbon nanotubes have demonstrated their potential for nanoscale electronics ranging from high performance field-effect transistors to ideal one-dimensional quantum dots with well-defined spin structure. The latter devices may even have prospects for solid-state quantum computing based on the electronic spins although fundamental issues regarding spin relaxation and decoherence still need further investigations in addition to the requisite progress in device processing.

A prominent example of quantum coherence and spin physics in quantum dots is the Kondo effect. Below the Kondo temperature, \( T_K \), an extended many-body state arises from the interactions between a localized electron spin on the dot and the conduction electrons in the leads. Kondo effects have been found in quantum dots based on carbon nanotubes and in single molecule transistors. In contrast to semiconductor dots, these systems allow for studies of Kondo effects in devices with different types of contacts, e.g. superconducting or magnetic electrodes. A nanotube Kondo dot coupled to superconducting electrodes has been realised and magnetically contacted quantum dots in the Kondo regime were reported recently in molecular transistors. We present here experimental data on a nanotube quantum dot, where the Kondo resonances are split at zero field. We interpret this as evidence for coupling of the dot to a ferromagnetic impurity (in the form of a catalyst nanoparticle).

Our devices incorporate single-wall carbon nanotubes grown by chemical vapor deposition (CVD) on an SiO\(_2\) substrate. Ferric iron nitrate nanoparticles deposited from a solution in isopropyl alcohol acted as catalyst for the CVD process, which was carried out in a tube furnace by flowing methane and hydrogen over the sample at 900°C. This process yielded mostly individual nanotubes with diameters in the range 1-3 nm as determined by atomic force microscopy. The nanotubes were contacted by thermally evaporated metal electrodes (35 nm Au on 4 nm Cr), spaced by 250 nm and patterned by electron beam lithography. Highly doped silicon below the 400 nm SiO\(_2\) cap layer acted as a gate electrode, see Fig. 1(a).

Electron transport measurements were carried out in a dilution refrigerator with a base electron temperature of \( T_a \sim 80\) mK as estimated from the device characteristics. The two-terminal conductance was measured using standard lock-in techniques with \( \sim 5\) mV ac excitation and voltage bias \( V \) applied to the source with the drain grounded through a low-impedance current amplifier.

In the appropriate range of back-gate voltage, \( V_g \), the room temperature conductance is around \( 1.8\ e^2/h\) and only weakly dependent on gate voltage \( V_g \) (not shown), indicating that the conducting nanotube is metallic. At low temperature, the differential conductance, \( dI/dV \), of the tube in the same range of back-gate voltage shows a strong dependence on \( V_g \), as seen in Fig. 1(b). The overall characteristics of the device are more clearly seen from the 2D plot of \( dI/dV \) as a function of source-drain voltage \( V \) and gate voltage \( V_g \) in Fig. 1(c). The dominant dark regions of low conductance are caused by Coulomb
blockade (CB) while the sloping bright lines are edges of the CB diamonds, where the blockade is overcome by the finite source-drain bias.

Moreover, faint light horizontal ridges of high conductance around zero bias are seen in Fig. 1(c). These ridges occur in an alternating manner, for every second electron added to the nanotube dot. They are consistent with Kondo resonances induced by the finite electron spin and the resonances are thus expected to appear as thin narrow peaks in $dI/dV$ as a function of $V$. The zero bias resonances are absent for the other regions (with even $N$) where the ground state spin is $S = 0$. A finite source-drain bias $eV \sim kT_K$ suppresses the Kondo effect and the resonances are thus expected to appear as thin horizontal lines at $V = 0$ in Fig. 1(c), or equivalently as narrow peaks in $dI/dV$ as a function of $V$. However, we note in Fig. 1(b) that in fact a fine splitting of the Kondo peak is observed.

Fig. 2 shows the temperature dependence of the zero bias conductance. We find a nonmonotonic temperature dependence in contrast to the usual Kondo resonances where the linear conductance, $G = dI/dV|_{V=0}$, scales as $G \sim -\log(T)$ for $T \gg T_K$ and decreases monotonically with increasing $T$. In the present device the conductance increases with $T$ up to around 200 mK, where $G$ saturates at 0.05 $e^2/h$. For higher $T$ (above ~500 mK) $G$ decreases as expected. The accompanying $dI/dV$ plots in Figs. 2(b)-(d) show the peak profiles at various temperatures. We note that the splitting is only observed at the lowest temperatures, corresponding to the range where $G(T)$ is increasing. Similar behavior is found for the other Kondo resonances in Fig. 1(c), although the magnitude of the splittings vary between the different resonances which involve different orbitals.

In a previous study of a semiconductor quantum dot in the Kondo regime double resonance peaks were found for an $S = 1/2$ 'two-stage' Kondo effect, where two different energy scales result in the appearance of a dip in the peak. However, this scenario does not apply to our case since the Kondo resonances here only exist for $S = 1/2$ as seen from the regular even-odd alternations in Fig. 1(c).

Application of an external magnetic field will normally cause the $S = 1/2$ Kondo resonance to split into two components with a peak spacing $\Delta V = 2E_Z/e$, where $E_Z = (1/2)g\mu_BB$ is the electron Zeeman energy. For the resonance in Fig. 1(b) the splitting is $\Delta V \approx 0.12$ mV, which would correspond to an external magnetic field of 0.51 T, since $g = 2.0$ for the conduction electrons in metallic carbon nanotubes. Such a large external field offset cannot exist in our experiment.

The magnetic field dependence for the resonance in Fig. 2(b) is shown in Fig. 3(a). The resonance is split for all fields. At large field the splitting grows as expected for the Zeeman splitting of a Kondo resonance. However, the minimal splitting is in fact achieved at a small finite field $B \sim 0.15$ mT, see Fig. 3(b). The splitting and asymmetry in field are significant and cannot be explained based on a model of a purely non-magnetic quantum dot in a weak external field.

Nanotubes can couple electrically to metal nanoparticles. For example, Ref demonstrates that a nonmagnetic gold nanoparticle placed in the gap between two nanotube segments can form a single-electron transistor with the nanotubes acting as leads. The sensitivity of the electronic properties of nanotubes to the presence of magnetic particles adsorbed on the tube walls has been proven by low-temperature STM measurements on tubes with Co clusters. We interpret the splitting of the Kondo peak as resulting from contact with a ferromagnetic particle, presumably from the iron-containing catalyst material used to grow the nanotubes. Theoretical work has shown the Kondo resonances persist when coupling a quantum dot to two ferromagnetic leads, however, the Kondo peaks in $dI/dV$ may split, even in the absence of an external magnetic field. It is anticipated that this result would also apply for dots coupled to just one magnetic lead. It has recently been shown using numerical renormalization group (NRG) theory that the conductance for such a one-magnetic-lead quantum dot would exhibit a nonmonotonic temperature dependence. These results support our interpretation.

FIG. 2: (a) Temperature dependence of the zero-bias conductance $G$ for the resonance shown in Fig. 1b ($V_0 = -5.10$ V). (b)-(e) $dI/dV$ as a function of $V$ for the same resonance, at temperatures 80 (b), 145 (c), 220 (d), and 550 mK (e).

FIG. 3: (a) Magnetic field dependence of $dI/dV$ as a function of source-drain voltage $V$ for the peak in Fig. 2(b) at base $T_d = 80$ mK. The applied magnetic field $B$ was increased from $-0.5$ T (dashed) over 0 T (thick) to 0.5 T (dotted) in steps of 0.1 T. Subsequent curves are offset by 0.01 $e^2/h$ for clarity. (b) Peak splitting $\Delta V$ as a function of applied magnetic field $B$ for the data in (a) (solid) and another series where $B$ was swept from 0.5 T to $-0.3$ T (open).
that the splitting of the Kondo resonances in our device reflects interactions of the quantum dot with a magnetic impurity.

In conclusion we have observed clear splittings of the Kondo resonances in a nanotube dot at zero field. Interestingly, gaps below 1 meV were recently observed in another nanotube quantum dot but $T$ or $B$ dependences were not reported, precluding detailed comparison to the present data. We propose that the Kondo resonances in this study has probed the effect of a magnetic impurity on electron transport in a carbon nanotube device. Future experiments allowing gate-controlled interaction with a ferromagnetic particle will provide important further information on the effects of ferromagnetic particles on quantum transport in nanotubes. Until now all published studies on the effects of defects on nanotube transport have considered nonmagnetic impurities or defects, for instance charge traps in gate oxide, atomic defects, contaminant particles, kinks, and normal metal particles. Meanwhile, in most fabrication methods for single-wall nanotubes magnetic particles from the catalyst remain in the material after growth, although a catalyst free route for single-wall nanotube synthesis was devised recently. It should be feasible to probe nanotubes coupled deliberately to individual magnetic particles, eg, by moving them into contact by AFM manipulation. Likewise, it would be desirable to achieve high transparency contacts to lithographically defined ferromagnetic electrodes. Nanotubes have already been contacted by magnetic electrodes but in all reported studies the transmission was too low to allow for Kondo resonances to form. Notably, controlling or eliminating unwanted interactions with magnetic materials will be crucial for experiments on spintronics as well as electron spin resonance and quantum computing in nanotubes.

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1 P. L. McEuen, M. Fuhrer, and H. Park, IEEE Trans. on Nanotech. 1, 78 (2002).
2 D. H. Cobden et al., Phys. Rev. Lett. 81, 681 (1998).
3 P. Jarillo-Herrero et al., Nature 429, 389 (2004).
4 D. Loss and D.P. DiVincenzo, Phys. Rev. A 57, 120 (1998).
5 D. Goldhaber-Gordon et al., Nature 391, 156 (1998).
6 L. Kouwenhoven and L. Glazman, Physics World 14, 33 (2001).
7 J. Nygård, D. H. Cobden, and P. E. Lindelof, Nature 408, 342 (2000).
8 J. Park et al., Nature 417, 722 (2002), W. J. Liang et al., Nature 417, 725 (2002).
9 M. Buitelaar et al., Phys. Rev. Lett. 89, 256801 (2002).
10 A. N. Pasupathy et al., Science 306, 86 (2004).
11 J. H. Hafler et al., J. Phys. Chem. B 105, 743 (2001).
12 See, e.g., L. P. Kouwenhoven, C. M. Marcus, P. L. McEuen, S. Tarucha, R. M. Westervelt, and N. S. Wingreen, in Mesoscopic Electron Transport, edited by L. P. Kouwenhoven, G. Schön, and L. L. Sohn (Kluwer, Dordrecht, The Netherlands, 1997).
13 W.G. van der Wiel et al., Phys. Rev. Lett. 88, 126803 (2002).
14 From the superconducting coil of our cryostat an offset field of no more than 10 mT can be expected as shown in other experiments. Moreover, care has been taken to avoid any magnetic material in the setup (chip carrier, holder, leads, filters, cryostat part etc.).
15 C. Thelander et al., Appl. Phys. Lett. 79, 2106 (2001).
16 T. Odom et al., Science 290, 1549 (2000).
17 G. A. Fiete et al., Phys. Rev. B 66, 24431 (2002).
18 The overall diamond pattern of the dot was stable for over a month while the device was kept at subkelvin temperatures, i.e. the charge states did not change. Details in the excited state spectrum and Kondo peak splittings changed occasionally, possibly reflecting varying interactions with an impurity particle.
19 J. Martinek et al., Phys. Rev. Lett. 91, 127203 (2003), ibid. 91, 247202 (2003).
20 M.-S. Choi, D. Sanchez and R. Lopez, Phys. Rev. Lett. 92, 056601 (2004).
21 M. Sindel and J. von Delft (unpublished).
22 B. Babic, T. Kontos, and C. Schönenberger, condmat/0407193.
23 Even strong coupling to a ferromagnetic oxide such as Fe$_2$O$_4$, which is half-metallic and believed to be fully spin polarized, may influence the electronic properties like for coupling to a magnetic metal.
24 F. Chen et al., Appl. Phys. Lett. 83, 4601 (2003).
25 V. Derycke et al., Nano Lett. 2, 1046 (2002).
26 K. Tsukagoshi, B.W. Alphenaar, and H. Ago, Nature 401, 573 (1999).