Kondo Effect in the Presence of Magnetic Impurities

H. B. Heersche,* Z. de Groot, J. A. Folk,† L. P. Kouwenhoven, and H. S. J. van der Zant
Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, 2628 CJ Delft, The Netherlands

A. A. Houck, J. Labaziewicz, and I. L. Chuang
Media Lab, Massachusetts Institute of Technology, Cambridge, Massachusetts, 02139, USA
(Received 24 July 2005; published 10 January 2006)

We measure transport through gold grain quantum dots fabricated using electromigration, with magnetic impurities in the leads. A Kondo interaction is observed between dot and leads, but the presence of magnetic impurities results in a gate-dependent zero-bias conductance peak that is split due to a RKKY interaction between the spin of the dot and the static spins of the impurities. A magnetic field restores the single Kondo peak in the case of an antiferromagnetic RKKY interaction. This system provides a new platform to study Kondo and RKKY interactions in metals at the level of a single spin.

DOI: 10.1103/PhysRevLett.96.017205 PACS numbers: 75.30.Hx, 72.15.Qm, 73.23.−b, 73.63.Kv

The observation of the Kondo effect in quantum dot systems has generated renewed experimental and theoretical interest in this many-body effect. The Kondo effect is the screening of a localized spin by surrounding conduction electrons. The localized spin can take the form of a magnetic atom, or the net spin in a quantum dot (QD). The Kondo effect has been studied extensively in quantum dot systems such as semiconductor quantum dots [1,2], carbon nanotubes [3], and single molecules contacted by metal leads [4–7].

The Kondo effect in a quantum dot can be used to probe interactions of a local spin with other magnetic moments. Whereas the Kondo effect enhances the zero-bias conductance through spin-flip processes, exchange interactions tend to freeze the spin of the QD. This competition results in a suppression and splitting of the Kondo resonance. The Kondo effect has been used to study the direct interaction between spins on a double dot [8,9], the exchange interaction with ferromagnetic leads [10], and the indirect Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction of two QDs separated by a larger dot [11]. In bulk metals with embedded magnetic impurities, the competition between the Kondo effect and RKKY coupling between impurities gives rise to complex magnetic states such as spin glasses [12].

In this Letter, we use the Kondo effect to study the RKKY interaction between the net spin of a quantum dot and magnetic impurities in the leads of an all-metal device. The system consists of a small gold grain in the vicinity of magnetic cobalt impurities [Fig. 2(a)]. By itself, the Kondo interaction with the net spin on such a grain induces a zero-bias peak in conductance. This feature is regularly observed in samples without impurities [13]. In the present experiment, cobalt impurities deposited intentionally cause the zero-bias peak to split. The splitting is explained by the RKKY interaction between the impurities and the spin of the grain. Temperature and magnetic-field dependence of the split zero-bias peak (SZBP) confirm this interpretation.

Measurements are performed on gold wires that have been broken by a controlled electromigration process, which is tailored to produce narrow gaps. Two substantially different procedures were followed, in two laboratories, but yielded similar results. Both procedures begin with a 12 nm gold bridge on top of an Al/Al_2O_3 gate electrode; see Fig. 1(a). A submonolayer of cobalt (Co) is evaporated on the sample before electromigration. For the first method, we monitor the change in resistance during electromigration (at room temperature) and adjust the applied voltage to maintain a constant break rate [13,14]. For the second, the junctions are broken by ramping the voltage across the circuit at $T = 4.2$ K and a series resistor is used to control the final gap size. The series resistance in our measurements was typically 50 Ω.

The differential conductance of the junctions is measured after breaking as a function of gate and bias voltage. As in samples without Co [13], Coulomb blockade and/or the Kondo effect were observed in 30% of the junctions that showed any conductance (this percentage depends on the precise electromigration procedure). Both effects are attributed to transport through ultrasmall gold grains, small enough to act as quantum dots with discrete energy levels [13,15]. This explanation is supported by the observation of electroluminescence from 18–22 atom gold grains in samples prepared in a similar manner [16].

An example of a gate-dependent Kondo resonance in a gold grain *without* Co is shown in Fig. 1(b). The Kondo effect enhances the differential conductance $G \equiv dI/dV_b$, around zero bias (dotted line) left of the charge degeneracy point (crossing point of Coulomb diamond edges, dashed lines). The zero-bias peak in G is suppressed with increasing temperature [Figs. 1(c) and 1(d)]. The height of the peak fits closely to the predicted functional form, $G(T) = G(0)/[1 + (2^{1/s} - 1)(T/T_K)^2]^s$ [17,18] with $s = 0.22$ for a spin-$\frac{1}{2}$ dot, yielding a Kondo temperature $T_K \approx 60$ K.

When magnetic impurities are scattered on the surface of the wire before breaking, over 10% of the samples [19]

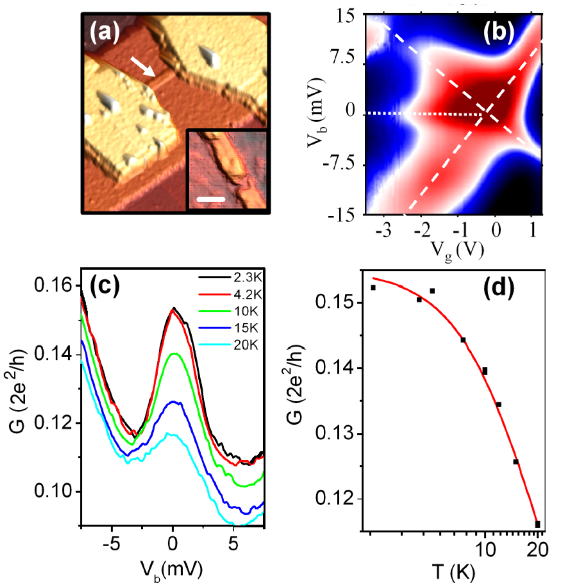

FIG. 1 (color online). Kondo effect in a gold grain quantum dot without magnetic impurities. (a) Atomic force microscopy picture of the device. A thin (12 nm) Au wire, connected to thick leads, lies on top of an oxidized Al gate (width 1 μm). Inset: After electromigration, a small gap ($\lesssim$1 nm, too small to resolve) is created containing small grains. (Scale bar corresponds to 100 nm.) (b) Differential conductance as a function of bias (V_b) and gate voltage (V_g). At $V_g \sim -0.2$ V, four diamond edges (peaks in $G = dI/dV_b$) come together in a charge degeneracy point. At the left hand side of the degeneracy point a conductance enhancement around $V_b = 0$ V is observed due to the Kondo effect. The dashed (diamond edges) and dotted (Kondo effect) lines are drawn as guides to the eye. Color scale ranges from 2 μS (black, dark blue online) to 22 μS (dark gray, dark red online). $T = 2.3$ K. (c) The height of the Kondo peak (at $V_g = -2$ V) decreases as a function of temperature. (d) Fit (red curve online) of the peak height to the expected temperature dependence suggests $T_K \approx 60$ K.

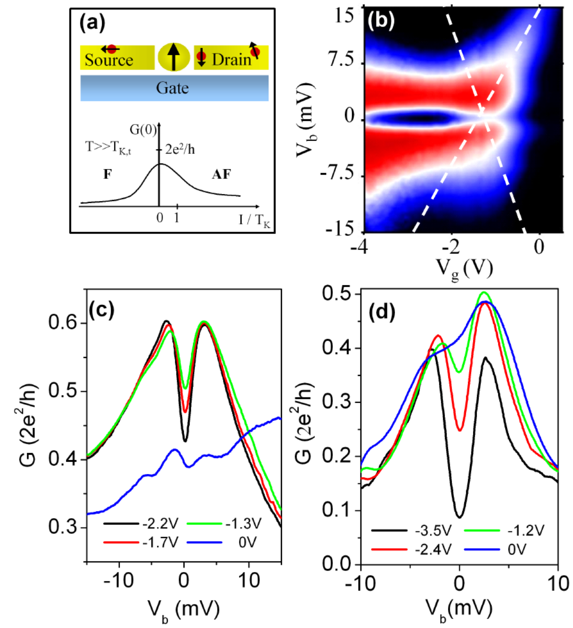

FIG. 2 (color online). SZBP of a gold grain quantum dot in the presence of magnetic impurities. (a) Top: Schematic of the device. Bottom: Sketch of the expected dependence of the zero-bias conductance $G(0)$ on the scaled RKKY coupling strength (I/T_k), at temperatures higher than the triplet Kondo temperature T_{K-t} [26]. $G(0)$ is suppressed both for strong FM and AFM interactions. (b) Differential conductance G as a function of bias (V_b) and gate voltage (V_g). The split zero-bias anomaly vanishes when an extra electron is added to the quantum dot ($V_g = -1$ V). Dashed lines (diamond edges) are a guide to the eye. Color scale from 28 μS (black, dark blue online) to 55 μS (dark gray, dark red online). $T = 2.3$ K. (c) Line plots from (a), showing suppression of zero-bias dip near charge degeneracy. (d) $G \equiv dI/dV_b$ versus bias for several gate voltages from a different device. Here the Kondo peak can be nearly restored with the gate. The depth of the zero-bias suppression is strongly gate dependent, but the peak separation remains constant.

show a split peak around zero bias rather than the single peak described above [20]. In Fig. 2(b), the differential conductance of one such device is plotted as a function of gate and bias voltage. Left from $V_g = -1$ V, a split zero-bias peak is observed; no SZBP is present at the right hand side. The onset of the SZBP coincides with a change in the number of electrons on the gold grain, as indicated by the diamond edge that intersects at $V_g \approx -1$ V (the fact that not all four diamond edges can be resolved is typical for these strongly coupled dots [7]). The parity effect observed in Fig. 2(b), like that in Fig. 1(b), is explained by a change of the net spin of the dot on the addition of an extra electron.

The SZBP can be explained by a competition between the Kondo effect and the RKKY coupling of the spin on the dot to one or more magnetic impurities in its vicinity [see Fig. 2(a) for a schematic of the system]. The relevant energy scales are T_K and the RKKY interaction strength I. An RKKY interaction suppresses elastic spin-flip processes and therefore suppresses the Kondo effect for low bias. Recently, the competition between RKKY interaction and the Kondo effect was studied theoretically by Vavilov *et al.* [21] and Simon *et al.* [22].

Peaks in conductance at $eV_b \simeq \pm I$ correspond to the voltage above which inelastic spin-flip processes are energetically allowed. The devices measured in Fig. 2 both give peak separations of 6 ± 1 meV, yielding $I = 3$ meV. Most devices that were measured fell in the range 1 meV $\lesssim I \lesssim$ 3 meV. The Kondo temperature, estimated from the total width of the SZBP, is found to be of the same order as I/k_B.

The temperature dependence of the zero-bias conductance is expected to be nonmonotonic due to the competition between the Kondo effect and RKKY interaction [23–25]. With increasing temperature, conductance increases due to thermal broadening of the peaks at $eV_b = \pm I$. The temperature of maximum zero-bias conductance is $T_m \sim I/k_B$, where both peaks have come together to form a single peak around zero bias. For $T > T_m$, the zero-bias conductance decreases for increasing temperature, similar to the Kondo effect without interactions. This behavior is also observed experimentally. The temperature dependence of the SZBP in Fig. 2(b) is shown in Fig. 3. Here $T_m = 25 \pm 5\ \mathrm{K} \simeq 0.7I$, with I extracted from the peak separation.

The sign of I is determined by the phase ϕ of the RKKY interaction, which is periodic in distance with the Fermi wavelength. Depending on the sign of I, the RKKY interaction is ferromagnetic ($I < 0$) or antiferromagnetic ($I > 0$). Both ferromagnetic (FM) and antiferromagnetic (AFM) interactions suppress the $S = \frac{1}{2}$ Kondo effect when $|I| \gtrsim T_K$ and $|eV_b| < |I|$. For an AFM interaction, the dot and impurity spins form an unscreened singlet ($S = 0$) state. In this case, the single peak due to an $S = \frac{1}{2}$ Kondo effect is replaced by a SZBP with peaks at $eV_b = \pm I$, at which bias the singlet-triplet transition becomes energetically available. For a FM interaction, the spins form a triplet ($S = 1$) state. The Kondo temperature associated with the triplet state, T_{K-t}, is much smaller than T_K [21,22]. At temperatures larger than T_{K-t}, a SZBP is also observed for a ferromagnetic I. As a result, the zero-bias conductance $G(0)$ as a function of the RKKY interaction I is maximum at $I \simeq 0$ (assuming $T > T_{K-t}$) [26] [see Fig. 2(a)].

The magnetic-field dependence of the SZBP depends on the sign of I, and is therefore an important tool to determine whether the interaction is FM or AFM. An external field can restore the Kondo effect if the RKKY interaction is AFM [21,22]. This is because the energy between the singlet ground state and the $|S = 1, m = -1\rangle$ triplet state decreases with $|B|$, Fig. 4(a). A Kondo state is restored at $B = I/(g\mu_B)$, where singlet and triplet states are degenerate and the external field compensates the AFM interaction. For a FM interaction, on the other hand, the peak spacing is expected to increase monotonically with $|B|$ because the splitting between the triplet $|S = 1, m = -1\rangle$ ground state and the singlet state also increases.

A characteristic field dependence for the AFM case is shown in Fig. 4(b). Upon increasing the field, the dip in the SZBP gradually diminishes until the Kondo peak is fully restored at 4.5 T [27]. Above 4.5 T the Kondo peak splits again. Because the g factors of the dot spin and the magnetic impurity may be different, it is difficult to compare I with the Zeeman energy at 4.5 T. In two of the devices showing a SZBP at zero magnetic field, the splitting increased with $|B|$ as is expected for an FM interaction [see Fig. 4(c)].

Both AFM and FM interactions are observed in different devices because the sign of I depends on the exact device geometry. The surprising fact that more AFM interactions are observed compared to FM interactions may result from experimental temperatures below T_{K-t}. In that case a triplet Kondo peak could be confused with an $S = \frac{1}{2}$ Kondo peak. Interactions with several cobalt impurities at varying distances may contribute to the imbalance as well.

A characteristic feature of these samples is that the dip around zero bias becomes more pronounced away from the charge degeneracy point, whereas the peak positions are insensitive to the gate [see Figs. 2(c) and 2(d)]. A gate

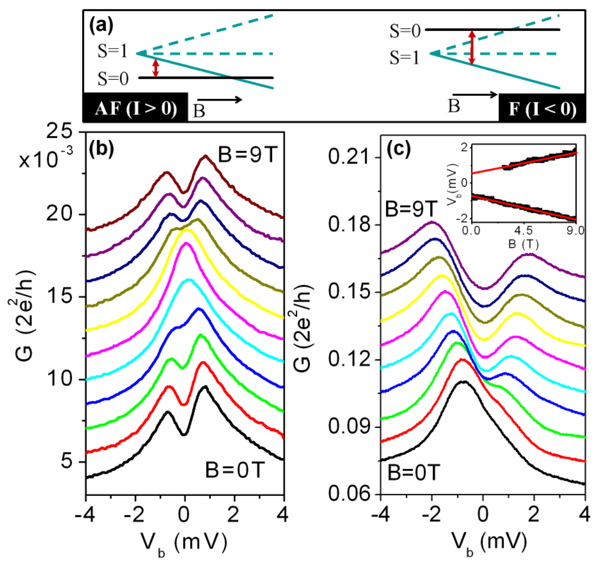

FIG. 4 (color online). Magnetic-field dependence of the split zero-bias anomaly. (a) For AFM interaction, the singlet-triplet transition energy (vertical arrow) decreases, then increases with B field. For FM interaction, the triplet-singlet energy always increases with field. (b), (c) Line plots are taken at different values of the external magnetic field, increasing from $B = 0$ T (bottom line) to $B = 9$ T (top line) in steps of 0.9 T. Data taken at $T = 250$ mK (b) Restoration of the Kondo effect at finite field, typical for AFM interaction between two spins. Line plots offset by $1.5 \times 10^{-3}(2e^2/h)$, for clarity. (c) For a FM interaction the peak separation increases linearly with $|B|$. The peak separation at $B = 0$ T is determined by extrapolating from peak positions to zero field (inset) and yields 1.4 ± 0.3 meV. Line plots offset by $1 \times 10^{-2}(2e^2/h)$.

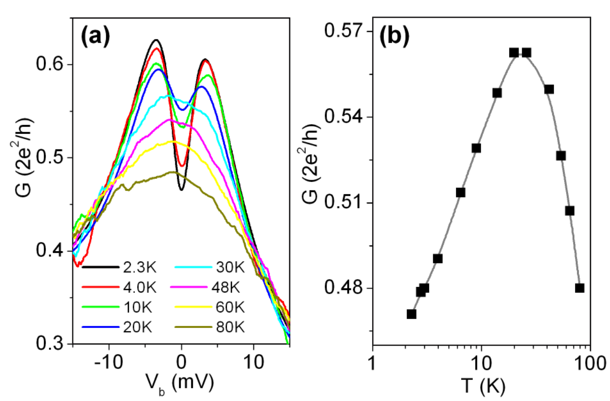

FIG. 3 (color online). Temperature dependence of the split zero-bias peak [same device as in Fig. 2(b)]. (a) $G \equiv dI/dV_b$ as a function of bias for different temperatures. (b) Nonmonotonic temperature dependence of the conductance at $V_b = 0$ V. Data points are measurements; the line is a guide to the eye.

voltage can change the coupling strength J_1 between the spin of a QD and the conduction electrons in the leads, $J_1 \propto 1/V_g$. The Kondo temperature depends exponentially on J_1, $T_K \propto \exp(-1/\rho|J_1|)$, so T_K rapidly decreases away from the degeneracy point [18]. Compared to T_K, the RKKY interaction energy $I \propto J_1 J_2 \cos\phi$ depends less strongly on J_1, so the ratio I/T_K increases away from the degeneracy point (J_2 is the coupling of the spin of the magnetic impurity to the free electrons in the leads). A quantum phase transition has been predicted between Kondo and RKKY phases as a function of I/T_K, which is replaced by a smooth crossover at higher temperatures or when particle-hole symmetry is broken [21,22,28,29] [Fig. 2(a)]. The transition from SZBP to Kondo peak in Fig. 2(d) may indicate a gate induced transition between RKKY and Kondo phases.

Other mechanisms that can lead to a SZBP have also been considered, but can be ruled out for several reasons. First, nearly degenerate singlet-triplet states *within* the dot may result in a SZBP [30–32]. However, this option is disregarded since it does not explain the observed dependence on the presence of magnetic impurities. Second, a SZBP at zero magnetic field was recently observed in a single C_{60} molecule QD with ferromagnetic leads [10]. The (gate-independent) SZBP in that work was attributed to exchange splitting of the Kondo peak by the ferromagnetic leads. Evidence for this explanation was provided by the dependence of the splitting on the relative orientation of the ferromagnetic electrodes. The absence of hysteresis with magnetic field in any of our measurements, together with the relatively low ($\lesssim 1\%$) Co concentration, make this an unlikely mechanism to explain our results.

In conclusion, we have observed a gate-dependent SZBP in electromigrated gold break junctions in the presence of magnetic impurities. These observations are consistent with a RKKY interaction between the local spin of a small gold grain and magnetic Co impurities. Magnetic-field dependence distinguishes between FM and AFM interactions. This system is a flexible platform to study the interaction between static magnetic impurities and the spin on a tunable quantum dot in an all-metal system. It bridges the gap between studies of the RKKY and Kondo interactions in bulk metals, and measurements of the two effects in semiconductor quantum dots.

We thank R. Lopez, J. Martinek, P. Simon, and M. G. Vavilov for useful discussions. Financial support was obtained from the Dutch organization for Fundamental Research on Matter (FOM), and from the RTN Spintronics Network. The work at MIT was funded by an HP-MIT alliance through the Quantum Science Research Group, AFOSR MURI Grant No. F49620-03-1-0420, and the NSF Center for Bits and Atoms. A. A. H. acknowledges support from the Hertz Foundation.

*Electronic address: hubert@qt.tn.tudelft.nl
†Present address: Department of Physics, University of British Columbia, Vancouver, Canada.

[1] D. Goldhaber-Gordon *et al.*, Nature (London) **391**, 156 (1998).
[2] T. H. O. S. M. Cronenwett and L. P. Kouwenhoven, Science **281**, 540 (1998).
[3] J. N. Nygård, D. H. Gobden, and P. E. Lindehof, Nature (London) **408**, 342 (2000).
[4] W. Liang *et al.*, Nature (London) **417**, 725 (2002).
[5] J. Park *et al.*, Nature (London) **417**, 722 (2002).
[6] L. H. Yu and D. Natelson, Nano Lett. **4**, 79 (2004).
[7] L. H. Yu and D. Natelson, cond-mat/0505683 [Phys. Rev. Lett. (to be published)].
[8] A. M. C. H. Jeong and M. R. Melloch, Science **293**, 2221 (2001).
[9] J. C. Chen, A. M. Chang, and M. R. Melloch, Phys. Rev. Lett. **92**, 176801 (2004).
[10] A. N. Pasupathy *et al.*, Science **306**, 86 (2004).
[11] N. J. Craig *et al.*, Science **304**, 565 (2004).
[12] A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, Cambridge, 1993).
[13] A. A. Houck *et al.*, Nano Lett. **5**, 1685 (2005).
[14] D. R. Strachan *et al.*, Appl. Phys. Lett. **86**, 043109 (2005).
[15] R. Sordan *et al.*, Appl. Phys. Lett. **87**, 013106 (2005).
[16] J. I. Gonzalez *et al.*, Phys. Rev. Lett. **93**, 147402 (2004).
[17] T. A. Costi, A. C. Hewson, and V. Zlatić, J. Phys. Condens. Matter **6**, 2519 (1994).
[18] D. Goldhaber-Gordon *et al.*, Phys. Rev. Lett. **81**, 5225 (1998).
[19] For the devices broken with feedback and measured at $T = 250$ mK, split zero-bias peaks were observed in 18/62 devices and unsplit Kondo peaks in 13/62 devices. At $T = 1.6$ K and no feedback, these numbers were 9/120 and 17/120, respectively.
[20] An SZBP was also observed in 2 samples without Co, most likely due to the presence of unintended magnetic impurities in the gold.
[21] M. G. Vavilov and L. I. Glazman, Phys. Rev. Lett. **94**, 086805 (2005).
[22] P. Simon *et al.*, Phys. Rev. Lett. **94**, 086602 (2005).
[23] M. Pustilnik and L. I. Glazman, Phys. Rev. Lett. **87**, 216601 (2001).
[24] R. López, R. Aguado, and G. Platero, Phys. Rev. Lett. **89**, 136802 (2002).
[25] R. Aguado and D. C. Langreth, Phys. Rev. B **67**, 245307 (2003).
[26] M. Pustilnik and L. I. Glazman, Phys. Rev. B **64**, 045328 (2001).
[27] Transitions between singlet and triplet excited states [21,22] are not observed. This is probably due to their low intensity and/or smoothing by temperature.
[28] B. A. Jones and C. M. Varma, Phys. Rev. Lett. **58**, 843 (1987).
[29] B. A. Jones and C. M. Varma, Phys. Rev. B **40**, 324 (1989).
[30] W. G. van der Wiel *et al.*, Phys. Rev. Lett. **88**, 126803 (2002).
[31] S. Sasaki *et al.*, Nature (London) **405**, 764 (2000).
[32] A. Kogan *et al.*, Phys. Rev. B **67**, 113309 (2003).