# The Kondo effect in the presence of magnetic impurities

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We measure transport through gold grain quantum dots that are fabricated by electromigration, with magnetic impurities in the leads. A Kondo interaction is observed between the gold grain dot and the leads, but the presence of magnetic impurities results in a split zero-bias peak in differential conductance. We attribute this splitting to an RKKY interaction between the tunable spin of the quantum dot and the static spins of surrounding magnetic impurities. The split peaks are gate dependent, show a parity effect, and a non-monotonic temperature dependence. A magnetic field restores the Kondo peak in the case of an antiferromagnetic interaction, whereas the splitting increases for a ferromagnetic interaction. This system provides a new platform to study the competition between Kondo and RKKY interactions in metals at the level of a single spin.

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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, alternatively, the net spin in a quantum dot (QD). When electron transport is mediated by tunneling through such a localized spin site, the signature of the Kondo effect is an enhancement of conductance around zero-bias in the Coulomb blockade regime. The Kondo effect has been studied extensively in many different 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 interaction tends to freeze the spin of the QD. This competition results in a suppression and splitting of the Kondo resonance. Experimentally, 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 an all-metal device. The system consists of a small gold grain in the vicinity of magnetic cobalt impurities, Fig. 2(a). 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 net spin on the gold grain. Temperature and magnetic field dependence of the split zero-bias peak (SZBP) confirm this interpretation. Gold grain quantum dots are particularly well-suited to the study of the competition between Kondo and RKKY interactions due to their high Kondo temperature ($\sim 5 - 100$ K) compared to semiconducting quantum dots.

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 different laboratories, but yielded similar results. Both procedures begin with a thin (±12 nm) gold bridge on top of an $Al/Al_2O_3$ gate electrode, see Fig. 1. Prior to electromigration, a sub-monolayer of cobalt (Co) is evaporated on top of the gold bridge. 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]. For the second, a series resistor is used to control the final gap size at $T = 4.2$ K. We find that when the junctions are broken by ramping the voltage across the circuit, the total series resistance (external resistor plus on-chip wires) is an important parameter for 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 the samples without Co [13], Coulomb blockade and/or the Kondo effect were observed in 30 percent of the junctions that showed a finite conductance (this

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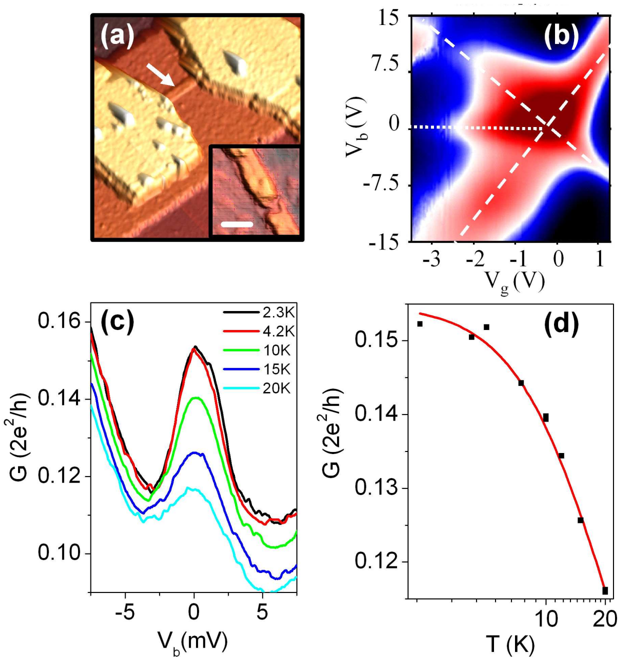

FIG. 1: Kondo effect in a gold grain quantum dot without magnetic impurities. a) Atomic force microscopy picture of the device. A thin (12 nm) Au wire, connected to thick leads, lies on top of an oxidized Al gate (width 1 $\mu$m). *Inset* After electromigration, a small gap ($\lesssim$1 nm, too small to resolve) is created containing small grains. (Scale bar corresponds to 100nm). b) Differential conductance as a function of bias ($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 $\mu$S (dark blue) to 22 $\mu$S (dark red). $T = 2.3$ K c) The height of the Kondo peak (at $V_g = -2$ V) decreases as a function of temperature. d) Fit (red curve) of the peak height to the expected temperature dependence suggests $T_K \approx 60$ K.

percentage depends on the precise electromigration procedure). Both effects are attributed to transport through ultra-small gold grains, small enough to act as quantum dots with discrete energy levels [13, 14]. This explanation is supported by the observation of electroluminescence in 18-22 atom gold grains in samples that were prepared in a similar manner [15].

A typical example of a gate dependent Kondo resonance in a gold grain *without* Co is shown in Fig. 1(b). A single degeneracy point is visible at the crossing of the edges of Coulomb blockade diamonds ($V_g = -0.2$ V). These edges appear as peaks in differential conductance $G = dI/dV_b$ and are broadened due to the strong coupling to the leads. On the left hand side of the degeneracy point, an enhancement of the differential conductance around zero-bias is visible (dotted line). On the right hand, no zero-bias anomaly is present due to the change of the net spin upon adding an electron to the dot.

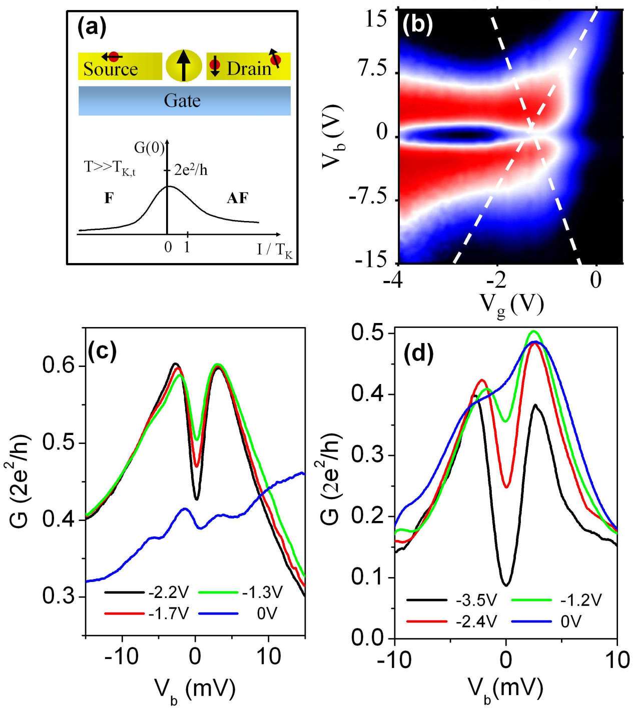

FIG. 2: Split zero-bias peak (SZBP) of a gold grain quantum dot in the presence of magnetic impurities. a) *Top*: Schematics of the device. *Bottom*: Sketch of the expected dependence of the zero bias conductance $G(0)$ on the relative RKKY coupling strength $(I/T_k)$ for temperatures higher than the triplet Kondo temperature $T_{K,t}$ [18]. $G(0)$ is suppressed both for strong ferromagnetic (F) and anti-ferromagnetic (AF) interaction. 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 at $V_g = -1$ V. Dashed lines (diamond edges) are a guide to the eye. Color scale from 28 $\mu$S (dark blue) to 55 $\mu$S (dark red). $T = 2.3$ K c) Line plots from (a) for several gate voltages. d) $G = dI/dV_b$ versus bias for several gate voltages from a different device. For this device the Kondo peak can be almost completely restored with the gate. Although the depth of the zero-bias suppression is strongly gate dependent, the peak separation is insensitive to the gate.

The zero-bias conductance peak is suppressed with increasing temperature. A line trace at a gate voltage of $V_g = -2$ V is shown in Fig. 1(c) for several temperatures. The peak height follows the predicted functional form: $G(T) = G(0)/[1 + (2^{1/s} - 1)(T/T_K)^2]^s$ [16, 17], with $s = 0.22$ for a spin $\frac{1}{2}$ dot, as illustrated in Fig. 1(d). A fit to this equation yields the Kondo temperature $T_K \approx 60$ K.

When magnetic impurities are scattered on the surface of the wire before breaking, over ten percent of the samples [19] show a split peak around zero bias rather than the single peak described above [20]. The origin of this split peak is the main topic of this Letter. In Fig. 2(b), the differential conductance of one such device is plotted as a function of gate and bias voltage. Left from $V_g \approx -1$ V, a split zero-bias peak (SZBP) 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.

In Fig. 2(c), the differential conductance is plotted as a function of bias for different gate voltages. A characteristic feature of these samples is that the dip around zero bias becomes more pronounced away from the degeneracy point, whereas the peak positions are insensitive to the gate. A strong gate dependence of the SZBP is shown in Fig. 2(d), where the dip can be almost completely suppressed with the gate. This gate dependence will be discussed later.

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 given by $T_K$ and the RKKY interaction strength $I$. Recently, this competition was studied theoretically by Vavilov *et al.* [21] and Simon *et al.* [22]. Depending on the sign of $I$, the RKKY interaction is ferromagnetic ($I < 0$) or antiferromagnetic ($I > 0$). Both ferromagnetic (F) and antiferromagnetic (AF) interactions are expected to suppress the $S = \frac{1}{2}$ Kondo effect when $eV < |I|$. For a ferromagnetic interaction, the net spin of the quantum dot and the spin of the magnetic impurity form a triplet ($S = 1$) state. The Kondo temperature of the triplet state $T_{K-t}$ is, however, much smaller than $T_K$. When the RKKY interaction is anti-ferromagnetic, the two spins form a singlet ($S = 0$) state, and no Kondo effect is possible. The zero bias conductance $G(0)$ as a function of the RKKY interaction $I$ has a maximum around $I/T_K = 0$ for temperatures above $T_{K-t}$ [18] [Fig. 2(a)]. In both the F (with $T > T_{K-t}$) and AF cases, the single Kondo peak is replaced by an SZBP.

Peaks in conductance at $|eV| \simeq I$ correspond to the bias voltage above which the triplet-singlet (F) or singlet-triplet (AF) transitions are energetically allowed. The peak separation for both devices in Fig. 2, $6 \pm 1$ meV, yields $I = 3$ meV. Most devices that were measured fell in the range 1 meV $\lesssim I \lesssim$ 3 meV. The Kondo temperature can be estimated from the total width of the SZBA, and is usually found to be of the same order as $I/k_B$.

The temperature dependence of the zero-bias conductance is predicted to be non-monotonic [23–25] in the presence of a competition between Kondo effect and RKKY interaction. For increasing temperature, conductance is expected to increase as long as $T < T_m \sim I/k_B$, due to thermal broadening of the peaks at $eV = \pm I$. When the temperature is increased above $T_m$, however, the conductance decreases, similar to the ordinary Kondo effect. A measurement of the temperature dependence of the SZBP in Fig. 2(b) is shown in Fig. 3. Upon increasing the temperature, the zero-bias conductance first increases until, at $T_m = 25 \pm 5$ K, the dip vanishes completely and a single zero-bias peak remains. For even higher temperatures the zero-bias conductance decreases with increasing temperature as expected. In this device, $T_m = 25 \pm 5$ K$\simeq 0.7I$ with $I$ extracted from the peak separation.

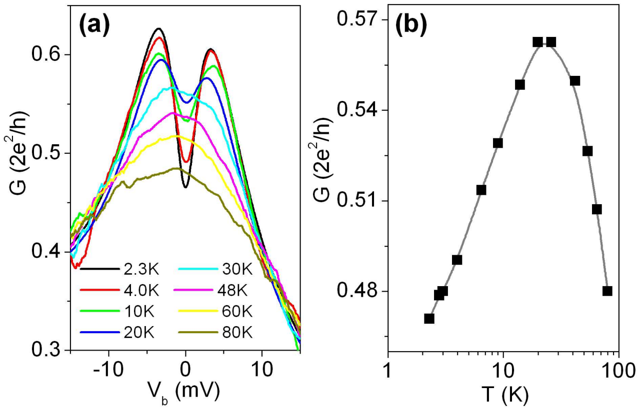

FIG. 3: Temperature dependence of the split zero-bias peak [same device as in Fig. 2(b)]. a) $G = dI/dV_b$ as a function of bias for different temperatures. b) Non-monotonic temperature dependence of the conductance at $V_b = 0$ V. Data points correspond to measurements, the line is a guide to the eye.

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 F (with $T > T_{K-t}$) or AF. The sign of $I$ is determined by the phase $\phi$ of the RKKY interaction, which is periodic in distance with the Fermi wavelength. Therefore the sign of $I$ is expected to be random. An external magnetic field can restore the Kondo effect if the interaction is AF [21, 22]. This is because the energy difference between the singlet ground state and the $|S{=}1, m{=}{-1}\rangle$ triplet state decreases linearly with $|B|$, Fig. 4(a). The Kondo state is restored at $B = I/(g\mu_B)$, where singlet and triplet states are degenerate and the external field compensates the AF interaction. For higher magnetic fields, the Kondo peak splits with the Zeeman energy. For an F interaction, the peak spacing is expected to increase linearly with $|B|$. This is due to the fact that the energy difference between the triplet $|S{=}1, m{=}{-1}\rangle$ ground state and the excited singlet state increases with $|B|$.

A characteristic field dependence for the AF case is shown in Fig. 4(b). Here the splitting $1.6 \pm 0.3$ meV ($I = 0.8$ meV) is relatively small at zero field. Upon increasing the magnetic field, the dip in the SZBP gradually diminishes until the Kondo peak is fully restored at

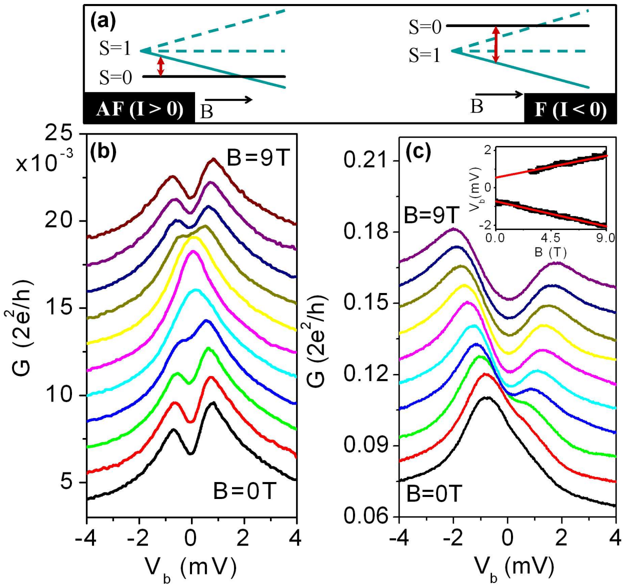

FIG. 4: Magnetic field dependence of the split zero-bias anomaly. a) For AF interaction, the singlet-triplet transition energy (red arrow) decreases, then increases with B-field. For F interaction, the triplet-singlet energy always increases with field. b,c) Line plots are taken at different values of 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 anti-ferromagnetic interaction between to spins. The plots are offset by $1.5 \times 10^{-3}(2e^2/h)$, for clarity. c) Peak separation increases linearly with $|B|$. This behavior corresponds to ferromagnetic interaction. The peak separation at $B = 0$ T is determined by extrapolating from peak positions at finite field (inset) and yields $1.4 \pm 0.3$meV. The plots are offset by $1 \times 10^{-2}(2e^2/h)$.

4.5 T [26]. When the magnetic field is increased even further, the Kondo peak splits again. Because the g-factors of the dot spin and the magnetic impurity may be different, it is difficult to extract the g-factor from these measurements.

In two of the devices showing an SZBP at zero magnetic field, the splitting increases with increasing $|B|$, as is expected for an F interaction. An example is shown in Fig. 4(c). The zero field splitting is determined to be 1.4 ± 0.3 meV by extrapolating the field dependent peak positions (inset). We find more AF interactions compared to F interactions. This imbalance may be due to the fact that in some cases the experimental temperature may have been below $T_k - t$ resulting in a triplet Kondo peak. In addition, interactions with several cobalt impurities at varying distances cannot be excluded. Further theoretical work will be required to understand the consequences of multiple interactions.

We are not aware of any theoretical predictions for the gate dependence of an SZBP that results from competition between Kondo effect and RKKY interaction. In the case of the ordinary Kondo effect, the gate changes the coupling strength, $J_1$, between the spin of the 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. The RKKY interaction energy $I \propto J_1 J_2 \cos\phi$ is proportional to both $J_1$ and the coupling of the spin of the magnetic impurity to the free electrons in the lead, $J_2$. Because the Kondo temperature depends more strongly on $J_1$ (and therefore on the gate voltage) than $I$, the ratio $I/T_K$ also depends on gate voltage, and is larger away from a charge degeneracy point. This may explain the gate dependence seen, for example, in Figs. 2(c,d).

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 and when particle-hole symmetry is broken [21, 22, 27, 28] [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 an SZBP at zero magnetic field have also been considered, but can be ruled out for several reasons. First, an SZBP at zero magnetic field has been observed recently in electromigrated ferromagnetic break junctions with $C_{60}$ molecules deposited on top [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.

Nearly degenerate singlet and triplet states *within* the quantum dot can result in a two-stage Kondo effect [29–31] with experimental signatures very similar to those observed here: a sharp dip superimposed on a broader Kondo resonance. We rule out the possibility of a two-stage Kondo effect in our measurements for two reasons. First, this explanation is not consistent with the observed dependence on the presence of magnetic impurities. Second, the observed parity effect is not expected for two-stage Kondo.

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 an RKKY interaction between the local spin of a small gold grain and magnetic Co impurities. Magnetic field dependence distinguishes between F and AF interactions. This system is a flexible platform to study the interaction between static magnetic impurities and the spin on a tunable quantum dot in a metal system. It bridges the gap between studies of the RKKY and Kondo interactions in bulk metals and the single-impurity Kondo effect measured in quantum dots.

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