Current-Induced Kondo Splitting in a Single-Atom Contact

D.-J. Choi,¹ M. V. Rastei,¹ J. S. Lim,² R. López,² P. Simon,³ and L. Limot¹

¹Institut de Physique et Chimie des Matériaux de Strasbourg, Université de Strasbourg, CNRS, 67034 Strasbourg, France
²Departament de Física, Universitat de les Illes Balears, 07122 Palma de Mallorca, Spain
³Laboratoire de Physique des Solides, CNRS, Université Paris-Sud, 91405 Orsay, France

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We use a low-temperature scanning tunneling microscope to split apart the Abrikosov-Suhl-Kondo (ASK) resonance by means of a high-density current. To this end, a copper-coated nickel tip is brought into contact with a single Co atom adsorbed on a Cu(100) surface. We show that the splitting is produced by the spin-polarized nature of the current in the copper spacer and, moreover, that the spin polarization and therefore the ASK resonance can be altered through the chemical properties of the contact.

Controlling a spin-polarized current by electrical means is central to the giant magnetoresistance effect [1, 2], and more generally to spintronics. A simple realization of a spintronic device consists in a layered structure comprising two ferromagnets separated by a non-magnetic region [2]. A non-equilibrium spin population is generated with the highest efficiency possible in one of the ferromagnets (the injector) and driven by the electron current into the nonmagnet [3, 4]. The spin population is then probed through the second ferromagnet by measuring the resistance of the entire device. A fundamental question is the limiting case where one of the layers would be downsized to a single magnetic atom. In this case, quantum electron transport occurs through conductance channels mainly determined by the chemical environment and valence orbitals of the atom bridging the two electrodes [5]. The conductance is of order 2e²/h as predicted by Landauer theory [6]. Despite notable advances in probing spin-polarized electron tunneling into single atoms with a scanning tunneling microscope (STM) [9, 11]—the conductance in this case does not exceed 0.01 × 2e²/h due to the vacuum barrier—the spin-polarized current in the contact regime remains little explored [12, 13]. This may provide alternative routes [14, 15] for monitoring—and eventually switching, magnetic bits in the quantum regime.

STM offers the possibility of customizing the contact measurement by choosing the material of the tip, surface and bridging atom [12, 18, 19], which is then appealing for studying the Kondo effect of a magnetic atom coupled to a non-magnetic tip and surface [20, 22]. Here we use a cryogenic (4.6 K) STM operated in ultrahigh vacuum to build a well-calibrated junction comprising a single Co atom on a Cu(100) surface. At variance with previous studies however, we employ a bulk ferromagnetic tip (nickel) coated by a thick layer of copper. Upon contact with the Co atom, a splitting of the Abrikosov-Suhl-Kondo (ASK) resonance is observed and assigned to the presence of a spin-polarized current into the copper coating. Our findings, supported by a theoretical description based on the Anderson model, show that the Kondo splitting is produced by a non-equilibrium effect known as spin accumulation [6] and, therefore, is fundamentally different from previous work of this kind [16]. We also show that spin transport can be altered by the tip-apex chemistry, as the ASK resonance is “preserved” with a pristine Ni tip. A single Kondo atom can therefore fulfill one of the major objectives recently envisioned in the domain of spintronics, i. e. the detection of a non-equilibrium spin current at the atomic-scale.

Figure 1(a) presents the evolution of the conductance (G = I/V₀) for two given Cu-coated tips and for two pristine Ni tips (solid blue lines) above a Co atom on Cu(100) (V₀ = −160 mV). The vertical dashed line is positioned at z = 0 where G = G₀. Inset: Constant-current image of Co and Cu atoms on Cu(100) (13 × 13 Å², −200 mV, 1 nA). Apparent heights are 1.1 and 0.8 Å for Co and Cu, respectively. (b)—(d) Histogram of G₀ = I₀/V₀ for Co/Cu tips (b), for W/Cu tips (c), and for Ni tips (d).
is influenced by atomic-scale relaxations [18, 24]; these vanish once $z \gtrsim 1$ Å. The $z = 0$ boundary is deduced through the contact conductance ($G_0 = I_0/V_0$), which is determined following a geometrical approach described elsewhere [12]. The contact formation is stabilised by charge transfer between the Co atom and the surface [18], and is highly reproducible and reversible. The average contact conductance is $\langle G_0 \rangle_{\text{Ni/Cu}} = 1.04 \pm 0.09$ (in units of $2e^2/h$) for Ni tips coated with copper (noted Ni/Cu), while it is $\langle G_0 \rangle_{\text{W/Cu}} = 1.05 \pm 0.09$ for W tips coated with copper (noted W/Cu). The similar values found for $G_0$ indicate that the conductance is governed by the bottleneck structure comprising the Co atom and the Cu apex atom of the tip. The contact of a Ni tip with Co produced a lower value of $\langle G_0 \rangle_{\text{Ni}} = 0.91 \pm 0.05$ compared to copper-coated tips, in agreement with previous work [12]. The weaker conductance observed with Ni tips is assigned to a smearing of the minority $d$ states of Co due to the hybridization with the minority $d$ states of Ni [27].

Figure 2 presents the evolution of the Kondo effect of Co/Cu(100) with non-magnetic tips (W/Cu) and bulk nickel tips covered with copper (Ni/Cu). The $dI/dV$ spectra are acquired by freezing the geometry of the junction at a selected conductance or, equivalently, current (the bias was $-160$ mV). A tip excursion of about 5.5 Å was covered by varying the conductance over several orders of magnitude. In Fig. 2 we focus on the contact region where substantial spectral changes are evidenced. With W/Cu tips (Fig. 2a), an ASK resonance is detected at the Fermi level, which is carried by about 10% of the electrons in the junction [22]. Along with Co, we also monitored the $dI/dV$ spectra on non-magnetic Cu and Au atoms, where, as expected, no ASK resonance is present (see orange line in Fig. 3 and Figs. 2a). To closely match the exact solution of the Numerical Renormalization Group theory, we reproduce the peak-like shape by a Frota function (solid orange lines in Fig. 2a) [28] and extract a Kondo temperature ($T_K$) from the line width [23, 29]. As shown in Fig. 2b, $T_K$ increases with current [22], reflecting the existence of tip-induced atomic relaxations in the environment of the Co atom [20]. The Kondo temperature varies typically from 100 K in the transition regime up to 220 K in the contact regime. The spectral signature of the ASK resonance changes dramatically when Ni/Cu (Figs. 2a and 2b) tips are brought into contact with the Co atom. Instead of a single peak, two peaks are detected in the $dI/dV$, approximately symmetric about the Fermi level. A total of fifty tips (see histogram of Fig. 1b) were employed and all showed a similar trend (copper-coated Fe tips also showed a similar behavior [23]). The ASK resonance splits apart as the tip excursion increases, reaching a value close to 25 mV at the highest tip excursions explored ($z \approx -0.7$ Å). Depending on the tip employed, the splitting is observed at

FIG. 2. (a) $dI/dV$ spectra for a W/Cu tip (transition regime: light-colored lines; contact: dark-colored lines). The solid orange lines correspond to fits using a Frota function [23]. All data are fully reversible with tip displacement. (b) and (c) present $dI/dV$ spectra acquired with two different Ni/Cu tips (the dashed lines are guide to the eye). The $\pm 5\%$ difference in the peak heights reflects the imbalance of the spin populations (majority and minority) in the electrodes [23]. (d) $T_K$ versus current for various W/Cu tips. The Kondo temperature is extracted from Frota fits [see (a)]. The vertical dashed line is positioned at $I = I_0$. The solid orange line corresponds to an exponential fit [22]. (e) $\Delta$ versus current for two typical Ni/Cu tips (solid circle: tip 1, open circle: tip 2). The lines are a guide to the eye. The solid red line corresponds to the exponential fit of panel (d) scaled by a factor 0.8 (see text), while the solid squares correspond to the smallest splittings observed with various tips. Below these values, the resonance splitting is lost.
most up to $z = 0.8 \text{ Å}$ away from contact [25]: it is lost above this value, i.e., when the tunneling barrier is fully restored [26]. Hereafter we therefore exclusively focus onto the contact regime.

A splitting of the ASK resonance is known to occur when a Kondo impurity is magnetically coupled with a ferromagnetic electrode [10, 24, 30, 32]. Since in our case the Co atom is separated from the bulk nickel by a copper spacer several nanometers thick [25], the coupling must necessarily be indirect. The splitting observed here is too large to be associated with the stray fields of Ni, as the Zeeman splitting amounts only to $2g\mu_B H \approx 0.2 \text{ meV}$. Another possibility to consider is a Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction mediated by the electrons of the copper spacer. Given the thickness of the spacer, the RKKY coupling constant is at most 1 meV [34, 35] or lower due to the roughness of the ferromagnet/nonmagnet interface [36]. The RKKY interaction therefore cannot explain our findings. Such a conclusion is confirmed by recalling that the smallest splittings observed for a collection of tips [33]. The critical value $\Delta_c > 0.8k_B T_K$, which is close to predictions [33]. The critical value $\Delta_c$ is plotted versus the conductance for various Ni tips. The Kondo temperatures are extracted from Frota fits [23]. The solid orange line is the fit of Fig. 2d.

Our findings rather indicate the existence of a spin-polarized current at the Co site. In macroscopic spin devices, it is well-known that when a current is injected from a ferromagnet into a non-magnetic metal, a magnetization builds up in the nonmagnet as a result of a balance among spin injection, spin accumulation and spin relaxation [3–6, 39]. Assuming the existence of two spin populations of currents $I_\uparrow$ and $I_\downarrow$, an effective magnetic field $H_{sd} \propto (I_\uparrow - I_\downarrow) \exp(-r/l_{sd})$ is generated at a distance $r$ away from the interface [40], where $l_{sd}$ represents the thickness of the copper spacer. Since at low temperature the spin diffusion length is $l_{sd} = 500 \text{ nm} \gg r$ for copper [40], we propose that the ASK resonance is spin split by the effective field $H_{sd} \propto (I_\uparrow - I_\downarrow)$.

The splitting of the ASK resonance ($\Delta$) is quantified in Fig. 2 for two typical tips. As shown, $\Delta$ increases with current, which within a spintronics paradigm reflects a higher spin injection occurring at the nickel/copper interface with increasing current [2]. Figure 2 also shows that the behavior of $\Delta$ is tip dependent. This is not surprising as some tip-related structure cannot be totally controlled within our setup. Firstly, the Ni/Cu interface, and therefore the polarization of the current injected into copper, may change with the tip employed. Secondly, structural relaxations induced by the tip-atom contact depend on structural details of the tip apices employed. These modify the conduction band of the contact [3, 41] and consequently the current flowing across the Co atom at a given tip excursion (see Fig. 1). Similarly, we expect structural relaxations to modify the current polarization across Co based on Eq. 1 [see here below]. Despite these limiting factors, practically all the Ni/Cu tips produced a splitting $\Delta > 20 \text{ mV}$ at the highest currents employed.

A higher current across the cobalt atom has a mixed effect on $\Delta$. It favors a stronger spin-polarized current through the spin injection at the ferromagnet/copper interface, but it also favors a higher Kondo temperature $T_K$, which tends instead to restore a single Kondo line [32]. This competition can result in the disappearance of the splitting in the contact regime (Fig. 2). To quantify this competition, we have plotted in Fig. 2 the smallest splittings observed for a collection of tips (solid squares). Based on this data, we then estimate that the resonance is approximately observable whenever $\Delta = 2g\mu_B H_{sd} > \Delta_c = 0.8k_B T_K$, which is close to predictions [33]. The critical value $\Delta_c$ is plotted versus the current in Fig. 2 and for this purpose we use the fit to $T_K$ in Fig. 2 scaled by a factor 0.8.

We also probed the impact of tip material on the spin-polarized current by performing contacts with a pristine Ni tip (Fig. 3). Compared to the previous setup, a direct ferromagnetic coupling is now present between the cobalt atom and the tip-apex atom. The ferromagnetic exchange field $H_{\text{FM}}$ adds up to the field $H_{sd}$ to split the ASK resonance apart. Supposing the Ni-Co exchange

![FIG. 3. (a) $dI/dV$ spectra from transition (light-colored lines) to contact (dark-colored lines) acquired with a pristine Ni tip. (b) $dI/dV$ spectra acquired in the contact regime with three different tips labeled 1, 2, 3 in the panel. For clarity, spectra 2 and 3 are displayed downward by 0.03 and 0.06, respectively. The bottom spectrum was acquired with the tip into contact with a non-magnetic Cu atom. (c) $T_K$ versus conductance for various Ni tips. The Kondo temperatures are extracted from Frota fits [23]. The solid orange line is the fit of Fig. 2d.](image)
FIG. 4. Theoretical calculation of the ASK resonance in the presence of a (a) spin-polarized current only, and (b) of a spin-polarized current and ferromagnetism with $P = 0.1$. For these calculations, $Q$ is constant with bias. We have chosen the following numerical values for the parameters of the Anderson model: $\epsilon = -3.5\Gamma$, $U \rightarrow \infty$, $\Gamma_{t} = 0.3\Gamma$, $\Gamma_{x} = 0.7\Gamma$ (the label $s$ refers to the surface), $\mu_{t} = 0.20\Gamma$, $\mu_{x} = 0$ and the Kondo temperature is determined by $T_{K} = D \exp(\pi\epsilon/\Gamma)$, where $D = 50\Gamma$ is the bandwidth. The differential conductance $dI/dV$ is plotted as a function of the bias in units of $eV/\Gamma$ where $\Gamma = \Gamma_{t} + \Gamma_{x}$. Panels (c) and (d) sketch how the ASK resonance changes in the experimental setup when using a copper-coated Ni tip and a pristine Ni tip, respectively. Note that spin injection causes an imbalance in the spin population in copper [39]. (e) ASK resonance in the presence of a spin-polarized current ($P = 0$) accounted for by a bias-dependent $Q$ [Eq. (2)]. The resonances were computed for various values of $eV$, with $Q_{0} = 0.2$. They are shifted vertically for clarity.

The ferromagnetism of the tip is accounted for through a spin-dependent hybridization function $\Gamma_{t,\uparrow} - \Gamma_{t,\downarrow}/(\Gamma_{t,\uparrow} + \Gamma_{t,\downarrow})$ defined at the Fermi energy. With these notations $H_{\text{fm}} \propto P\Gamma_{t}$ where $\Gamma_{t} = \Gamma_{t,\uparrow} + \Gamma_{t,\downarrow}$. To model instead the spin-polarized current, we recall that spin injection entails non-equilibrium spin-dependent chemical potentials in the copper covering the tip, which we note $\mu_{t,\uparrow}$ and $\mu_{t,\downarrow}$ [40, 41, 47] (we assume, without loss of generality, that this effect is solely carried by the tip). The difference $I_{t,\uparrow} - I_{t,\downarrow}$ is then proportional to $Q(V) = (\mu_{t,\uparrow} - \mu_{t,\downarrow})/(\mu_{t,\uparrow} + \mu_{t,\downarrow})$. Although $Q$ is generically a function of the voltage bias, we show later that the bias dependency may be neglected in the calculations so that $Q = Q_{0}$, where $Q_{0}$ is a constant. Under this assumption, the differential conductance can be computed through the truncated equation of motion techniques. As shown in Fig. 4(a), a non-equilibrium spin-polarized current encoded by $Q \neq 0$ splits the Kondo resonance into two peaks centered at $(\mu_{t,\uparrow} - \mu_{t,\downarrow})/2$ and $-(\mu_{t,\uparrow} - \mu_{t,\downarrow})/2$ (Fig. 4). The spin-polarized current at the Co site is therefore equivalent to a local magnetic field $H_{sd} \propto Q\Gamma_{t}$ [29], which decreases exponentially with $\Gamma_{t}$ as the tip is pulled away from the atom. This agrees with the experimental findings indicating the absence of a splitting in the tunneling regime. The more general situation of a finite equilibrium polarization where $P \neq 0$ and $Q \neq 0$ is presented in Fig. 4(b). The two effective fields add up so that the predicted splitting of the ASK resonance is then larger compared to Fig. 4(a) as now $P = 0.1$. Such a reinforcement of the resonance splitting is however not observed for the Ni tips (Fig. 4). This confirms that $H_{sd}$ and therefore $Q$, must decrease with these tips and, moreover, that $H_{\text{fm}}$ is not sufficient alone to split apart the Kondo resonance of cobalt (Fig. 4).

Finally, we discuss the realistic case of a bias-dependent $Q$. A general expression of $Q(V)$ can be obtained through the M"{e}ir-Wingreen formula, but requires a self-consistent calculation not technically at hand. As a first step towards a full description of the problem, a...
phenomenological expression of $Q(V)$ can be given on the basis of the following considerations. In the absence of bias, the equilibrium regime imposes the spin-polarized current to be zero and therefore $Q(0) = 0$. At low bias, where the linear response applies, the spin-polarized current is linear in $V$ and therefore also $Q(V)$. As bias increases, non-linear terms come into play, but beyond a certain bias $Q(V)$ saturates. To demonstrate this last assertion, we provide a microscopic description of the spin injection into an atom by remarking within our Anderson model that (for simplicity we set $P = 0$)

$$I_{\uparrow}(V) - I_{\downarrow}(V) \approx \frac{2\pi e}{\hbar} \Gamma_\uparrow \Gamma_\downarrow [n_\uparrow(V) - n_\downarrow(V)],$$

where $\Gamma_\sigma$ is the hybridization function of the surface. Equation (1) shows that the spin-polarized current is carried by the non-equilibrium local density of states (LDOS) of the atom $\langle \rho_\sigma \rangle$ through the $d$ orbital occupation numbers $n_\sigma(V) = \int d\omega \rho_\sigma(\omega, V)$ ($\sigma = \uparrow, \downarrow$). The spin-dependent LDOS is in turn affected by the choice of the atom, but also by the tip (surface) material and structure as corroborated experimentally. Most importantly, since $n_\uparrow + n_\downarrow \approx 1$ in the Kondo regime $[13]$, the difference between the occupation numbers is upper bounded ($n_\uparrow - n_\downarrow \leq 1$). A saturation can then be expected at large bias for the spin-polarized current and consequently also for $Q(V)$. In order to reconcile both the small and large bias limits, we have then adopted the following phenomenological form

$$Q(V) = Q_0 (1 - e^{-|V|/V_c}),$$

where $V_c$ is a phenomenological parameter. From the aforementioned arguments, $V_c$ is related to the non-equilibrium polarization of the $d$ orbital, the larger $V_c$, the less polarizable the atom being. In Fig. 4 we present the differential conductance computed by means of the equation of motion techniques for different values of $V_c$. These are compared to $k_B T_K$, which constitutes the main energy scale entering in the Kondo effect. Only when $Q(V)$ is linear in $V$ over an extremely large bias range ($eV_c \gg 10k_B T_K$), a splitting is not found. Otherwise, the phenomenological expression of $Q(V)$ of Eq. (2) provides results similar to the case $Q = Q_0$. The current-induced splitting of the ASK resonance is therefore a generic property (see also [14]).

In conclusion, the point contact reduced to a single Kondo atom constitutes a useful benchmark for investigating the interplay between Kondo correlations and a spin-polarized current. In this regards, our findings provide a fresh insight into the Kondo effect of ferromagnetic nanoconstrictions by clearly indicating the necessity of considering a spin-polarized current and its potential impact on the Kondo signature [13]. Although our work focused onto single atoms, studies of this kind can be extended to molecules and therefore to the blossoming research area of molecular spintronics. This, we believe, definitely gives a new twist to the Kondo effect.

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