

A tunable two-impurity Kondo system in an atomic point contact

J. Bork, Y. Zhang, P. Wahl and K. Kern

If a magnetic impurity is introduced into a non-magnetic metallic environment, its spin interacts with the conduction electrons of the host. Below a characteristic temperature T_K^0 , this leads due to the Kondo effect to a non-magnetic singlet ground state, where the spin of the magnetic impurity is completely screened [1]. As soon as two or more magnetic impurities are in proximity to each other, a competition between Kondo screening and magnetic interaction among the spins sets in and the ground state depends sensitively on their respective magnitude. Many of the peculiar properties of correlated electron materials are attributed to this competition between screening of local spins and magnetic interaction of neighbouring spins. Depending on which interaction dominates, the properties can vary, e.g., between antiferromagnetic ordering and heavy fermion behaviour, between being an insulator or a metal. Certain materials can be tuned continuously between the aforementioned phases through a quantum phase transition (QPT) as a function of an external control parameter, such as doping, pressure or magnetic field. The physics close to the quantum critical point is often described based on models in which the formation of dimers governs the physics. Thus the study of a model system consisting of a dimer, as presented here, with magnetically coupled spins can serve as a test ground for these theories.

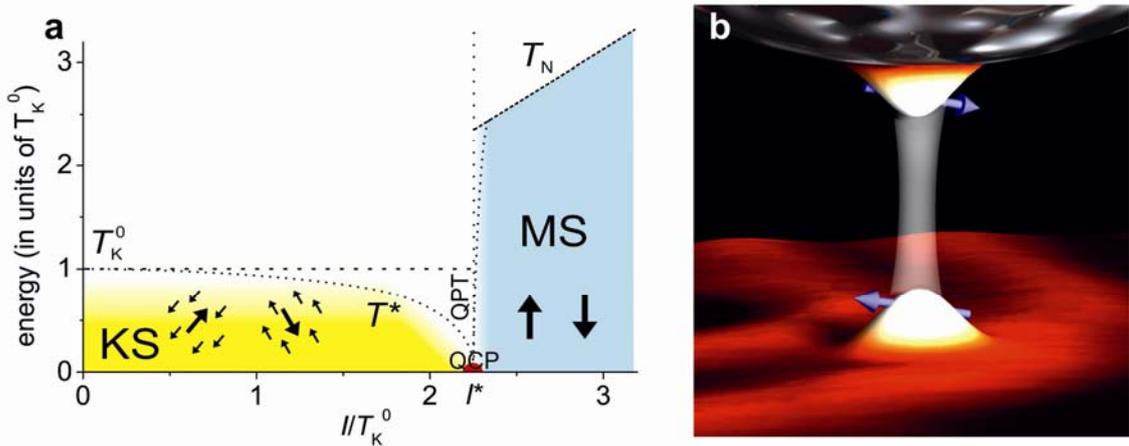


Figure 1: Phase diagram and experimental realization of the two-impurity Kondo Problem. (a) Relevant energy scales and ground states as function of coupling between the spins: KS – single-impurity Kondo screening dominates; AF – due to antiferromagnetic coupling, the two spins lock into a molecular singlet. The vertical dotted line indicates the critical coupling I^* where the quantum phase transition (QPT) occurs. (b) Illustration of measurement setup with one cobalt atom on the tip and one on the surface. The coupling between tip and sample results in an antiferromagnetic interaction I between the two spins. The strength of the interaction is varied by changing the tip-sample distance.

The physics of two Kondo impurities coupled by some interaction has been studied theoretically in great detail. The relative strengths of an antiferromagnetic exchange interaction I and of the single impurity Kondo temperature T_K^0 determine whether the ground state of the two-impurity system is governed by the formation of a singlet state between the two spins due to antiferromagnetic coupling or a Kondo singlet of each spin due to screening by the conduction electrons. A schematic phase diagram of the two-impurity Kondo problem with antiferromagnetic coupling I is displayed in Fig. 1(a). If the transition between the two ground states is governed by quantum fluctuations it constitutes a QPT.

We have realized a two-impurity Kondo system consisting of two magnetic atoms, one attached to the tip of a Scanning Tunnelling Microscope (STM) and one adsorbed on a metal surface, each constituting a Kondo system [2]. This configuration has been proposed as one of the simplest conceivable systems potentially exhibiting quantum critical behavior [3]. The coupling between the two atoms can be tuned continuously by varying the tip-sample separation, such that the system can be driven through the QPT (Fig. 1(a)). Electronic transport measurements on this two-impurity Kondo system reveal a rich physical scenario which is governed by a crossover from local Kondo

screening to non-local singlet formation due to antiferromagnetic coupling as a function of separation of the cobalt atoms. At the QPT, an additional resonance is expected to appear in the differential conductance at zero bias whose width is only determined by the temperature of the experiment [3].

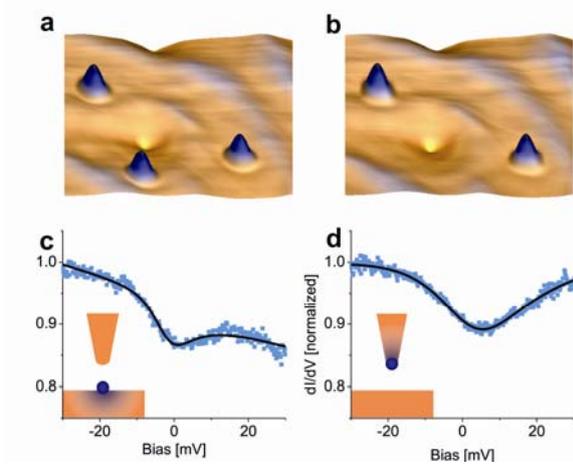


Figure 2: Preparation of the tip Kondo system. (a) STM image of three cobalt adatoms on Au(111) taken with a spectroscopically featureless tip, (b) after applying a voltage pulse with the tip on top of a cobalt adatom, the atom is transferred to the tip and has disappeared from the surface. (c) Spectra taken on cobalt adatoms show a Kondo resonance. (d) After picking up a cobalt atom, spectra taken on the clean surface show a resonance at the Fermi level (0V), too.

Following sample preparation, in STM images measured at a temperature of 6.7K we find single cobalt adatoms on the Au(111) surface (Fig. 2(a)), which can be identified by their characteristic Kondo resonance. In tunnelling spectra, the Kondo resonance is detected as a Fano line shape due to interference between electrons tunnelling into the conduction band of the substrate and into the Kondo state (Fig. 2(c)). Depending on the relative strengths of the two channels, the line shape of the Kondo resonance can be a dip, an asymmetric line shape or a peak. We have attached single cobalt atoms to the STM tip by positioning the tip on top of an adatom and applying a voltage pulse. After application of a successful pulse the cobalt atom disappears from the surface (see Fig. 2(b)) and the spectrum taken over a clean spot exhibits a similar resonance compared to the one found for cobalt atoms on the surface (see Fig. 2(d)).

While approaching the two cobalt atoms towards each other, the junction makes a transition from the tunnelling to the point contact regime (Fig. 3(a)). Spectra acquired in the tunnelling regime for $z > 0$ Å (see Fig. 3(b)) show a resonance similar to the one found for a single cobalt atom in the junction (compare figs. 2(c), (d)) however with a larger amplitude as it is a superposition of the Kondo resonances of tip and sample. Shape and width of the feature change only little for rather large tip-sample distances $z > 2$ Å. Once the two atoms are moved within 2 Å of point contact the width of the resonance is reduced. Close to the transition to the point contact regime, the resonance changes its shape from a dip to a peak of similar width (see Fig. 3(b) for $z \sim -30$ pm). On pushing the tip further in, the peak splits into two resonances at almost symmetric positions with respect to zero bias. The splitting increases with decreasing tip-sample distance (Fig. 3(b), (c)). We note that all data shown in this work is obtained from reversible, non-destructive measurements, where STM images taken before and after formation of the point contact show no changes of the tip or the cobalt atom itself. The conductance traces during approach and withdrawal are apart from some vertical drift coincident with each other. Occasionally we observe lateral hops of the cobalt atom which can however be directly detected in the conductance trace as well as from the STM images taken before and after the approach curve is acquired.

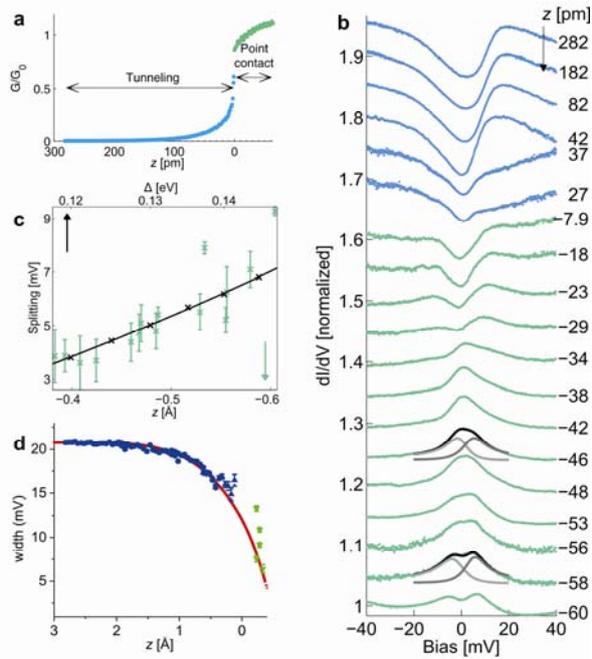


Figure 3: Measurements on two-impurity Kondo system in tunnelling and transport. (a) Approach curve acquired while moving the tip with a cobalt atom towards a cobalt atom on the surface. Shown is the conductance in units of the spin-degenerate conductance quantum $G_0=12.9k\Omega$ as function of tip-sample distance (relative to the transition from tunnelling (blue) to point contact (green)) at fixed bias voltage ($V_{\text{bias}} = 40\text{mV}$). (b) Spectra as a function of tip-sample distance (as indicated on the right) in the tunnelling and point contact regime. (c) Splitting of peaks seen in (b) as function of tip-sample distance. Black crosses have been obtained from NRG calculations. The solid line is a linear fit to NRG calculations. (d) width of the resonance extracted from spectra as in (b) as function of tip-sample distance, circles and triangles indicate separate sets of measurements following different sample preparations with different tips (colours as in (a)). The solid line shows the calculated width of the resonance, where the line shape parameter q is assumed to depend on the tip-sample distance z as $q \sim \exp(-z/\kappa d)$. Spectra in (b) are shifted vertically.

We can rationalise the reduction by the superposition of the Kondo resonances of tip and sample. In the spectra, only one resonance is observed because the resonances of the two cobalt atoms are both at the Fermi level. To assess the influence of their superposition on the overall spectrum, we have performed a model calculation assuming that the tip and sample spectra can be described by Fano resonances (with only one cobalt atom in the junction, see Fig. 2(c), (d)). We assume that only the line shape of the individual resonances changes from a dip to a peak while approaching the two cobalt atoms towards each other, keeping the parameters of the Kondo resonances (i.e. width, amplitude and position) constant as found in NRG calculation. With this model, we can recover the behaviour of the tunnelling spectra and of the overall width of the resonance, as can be seen from fig. 3(d).

We can clearly identify two coupling regimes for the cobalt dimer. Far out in the tunnelling regime, the two cobalt atoms behave as weakly interacting Kondo impurities, each with its own Kondo resonance. Approaching the two cobalt atoms towards each other, the interaction rises and the resonance splits due to exchange interaction. Further reduction of the tip-sample distance increases the interaction and hence the separation of the two peaks. For much stronger exchange interactions, the spectra are expected to be governed by inelastic spin excitations [4] breaking the singlet rather than by a split Kondo resonance.

Instead of showing a QPT, the cobalt dimer undergoes a crossover between the two regimes, Kondo screening and antiferromagnetism. By comparison with NRG calculations, the crossover can be attributed to the strong coupling between the electrodes. For the system to reveal quantum critical behaviour, the coupling between the electrodes needs to be small compared to the single impurity Kondo temperature. One way to come closer to a system exhibiting critical behaviour is to use atoms as Kondo systems which exhibit a significantly lower Kondo temperature while still having substantial coupling to the electrodes (by which the magnetic coupling is mediated). This can be achieved for Kondo systems which have a Coulomb repulsion about a factor of two higher than cobalt atoms, possible candidates could be rare earth atoms. Another way would be to increase the direct coupling between the orbitals which form the Kondo state. Direct charge coupling also renders the QCP unstable, so it will only be observable for the right balance of direct hopping and exchange interaction.

The atomically precise engineering of a two-atom Kondo system provides a unique playground to probe electron correlations, complementing studies on semiconductor quantum dots. Our experiment provides a model system for studying the competition between electron correlation and local moment antiferromagnetism which eventually might allow studying quantum criticality on a two-impurity system. It also opens up new perspectives for atomic scale magnetometry. The tip Kondo system can be used as a probe for magnetic interactions, yielding quantitative information about the magnetic

interaction. This offers the opportunity to study magnetic properties of atoms, magnetic molecules, clusters and islands with the high lateral resolution of STM.

References:

- [1] *Hewson, A. The Kondo Problem to Heavy Fermions*, Cambridge University Press (1993)
- [2] *Bork, J., Y. Zhang, L. Diekhöner, L. Borda, P. Simon, J. Kroha, P. Wahl and K. Kern. A tunable two-impurity Kondo system in an atomic point contact*, Nat. Phys. **7**, 901 (2011).
- [3] *Sela, E. and I. Affleck*. Phys. Rev. Lett. **102**, 047201 (2009).
- [4] *Heinrich, A.J., J.A. Gupta, C.P. Lutz and D.M. Eigler*. Science **306**, 466-469 (2004).

In collaboration with:

- L. Diekhöner (Aalborg Universitet)
- L. Borda (Budapest University of Technology and Economics)
- P. Simon (Université Paris-Sud)
- J. Kroha (Universität Bonn)