Two-impurity Kondo effect in potassium-doped single-layer p-sexiphenyl films

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Herein, we show that a self-assembled phase of potassium (K)-doped single-layer para-sexiphenyl (PSP) film on a gold substrate is an excellent platform for studying the two-impurity Kondo model. On K-doped PSP molecules well separated from others, we observe a Kondo resonance peak close to $E_F$ with a Kondo temperature of 30 K. The Kondo resonance peak splits when another K-doped PSP molecule is present in the vicinity, and the splitting gradually increases with the decrease in intermolecular distance without signs of phase transition. Our data demonstrate how a Kondo singlet state gradually evolves into an antiferromagnetic singlet state due to the competition between Kondo screening and antiferromagnetic Ruderman-Kittel-Kasuya-Yosida coupling, as described in the two-impurity Kondo model. Intriguingly, the antiferromagnetic singlet is quickly destroyed on increasing temperature and transforms back to a Kondo singlet below the Kondo temperature. Our data provide a comprehensive picture and quantitative constraints on related theories and calculations of the two-impurity Kondo model.

two-impurity Kondo effect, Kondo resonance, antiferromagnetic singlet, scanning tunneling microscopy

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1 Introduction

Kondo effect arises from interactions between an isolated magnetic impurity and surrounding conduction electrons in a metal. The spin of the magnetic impurity is completely screened by conduction electrons, and a Kondo singlet state forms below the Kondo temperature [1]. It is commonly present in condensed matter systems such as individual magnetic atoms [2-8] or molecules [9-15] on metals and quantum dots [16-25]. However, when the magnetic atoms or ions form an ordered Kondo lattice, as in various heavy-fermion crystals, magnetic sites could interact with each other via the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which is mediated through conduction electrons. The competition between Kondo screening and RKKY exchange interaction ($J$) in a Kondo lattice gives rise to a variety of complicated and interesting phenomena such as ferromagnetic or antiferromagnetic orders, heavy fermions, and quantum criticality [26-28].

Both the single-impurity Kondo model and Kondo lattice
model have been extensively explored in theory and experiment; however, the intermediate regime between these two cases has been addressed to a much less extent, particularly on the experimental side, despite its importance for the comprehensive understanding of Kondo physics. In this regime, the two-impurity Kondo model (2IKM) is a typical case in which two Kondo impurities are coupled via RKKY interactions [23-25,29-37]. Theoretically, the ground state of the two-impurity Kondo system is determined by the relative strengths of inter-impurity magnetic interaction and single-impurity Kondo temperature. It was predicted that for systems with particle-hole symmetry, a quantum phase transition would occur from the Kondo resonance state to an AFM singlet state as a function of intermolecular distance (which controls the exchange interaction strength) and the temperature. Our results provide a comprehensive phase diagram of the 2IKM, which deepens our understanding of this important many-body model system.

2 Experimental details

The experiments were performed in a low-temperature scanning tunneling microscope (STM) with an ultra-high vacuum and a base temperature of 5 K. Au(111) substrates were cleaned via cycles of Ar$^+$ sputtering and thermal annealing at 650°C. The PSP films of one monolayer (ML)-thick were prepared by depositing PSP molecules (95% powder from Alfa Aesar) on the Au(111) substrate via cycles of Ar$^+$ sputtering and thermal annealing at 650°C. The pristine PSP film is insulating as discussed in sect. 1 of Supplementary Materials (SM). Subsequently, K atoms were then doped into the films using alkali metal dispensers, and the deposition rate of K was calibrated in advance by directly counting K atoms deposited on Au(111) under the same condition. The sample quality was improved by annealing at room temperature for ~1 h. STM topography was taken in a constant current mode, and the $dI/dV$ spectra were collected using a standard lock-in technique with a modulation frequency $f=943$ Hz. PtIr tips were used for all STM measurements after being treated on a clean Au(111) surface. Except for temperature-dependent experiments, all experimental data were collected at ~5 K.

3 Results and discussion

Figure 1(a), (b) show the STM images of a single-layer PSP film grown on the Au(111) substrate (labeled as PSP/Au). The PSP molecules (C$_{36}$H$_{26}$), consisting of six benzene rings linked by a C–C single bond in the para-position, self-assemble in a stripe-like structure, similar to other phenyl-based molecules adsorbed on metal substrates [42-48]. The stripe-like structure exhibits a periodicity of (2.95±0.05) nm, while the intermolecular distance along the stripe direction is (0.70±0.05) nm. An individual PSP molecule can be clearly resolved in a magnified STM image, as shown in Figure 1(b).

Figure 1(c) with $K_c = 3.39$, is the focus of this paper, while other ordered phases are discussed in sect. 2 of SM. Figure 1(c) and Figure S3 in SM display the detailed lattice structure of phase I. The unit cell of this phase is indexed by the red rectangle with a size of ~3.98±0.05 nm × 3.37±0.05 nm. Assuming the same areal density of PSP molecules in single-layer PSP/Au film (~5.14×10$^{13}$ cm$^{-2}$), one of these phases, labeled as phase I in Figure 1(c) with $K_c = 3.39$, is the focus of this paper, while other ordered phases are discussed in sect. 2 of SM.

Figure 1(d) shows a zero-bias conductance (ZBC) map taken in the same region like that shown in Figure 1(c). Most PSP molecules in a unit cell show the featureless density of states (DOS) near $E_F$ except the two on the edges as marked with the blue color in Figure 1(d) (see sect. 4 of SM for
For the two edge PSP molecules, we observed either a high DOS (bright yellow areas in Figure 1(e)) or a low DOS (dark blue areas in Figure 1(e)), depending on the details of the arrangements between two edge molecules. Large-energy-scale $dI/dV$ spectra significantly differ for molecules on the edge and in the center, as shown in Figure 1(f). Such a difference in electronic states suggests that the electron filling or the couplings between the molecules and Au substrate strongly vary.

Figure 2 displays four representative molecular dimers (referring to the two neighbor blue PSP molecules in Figure 1(d)) with varying intermolecular distance $d$ (as defined in Figure 2(a)) and their corresponding $dI/dV$ spectra. For molecular dimer #1, the two PSP molecules are far apart ($d = 2.34$ nm), and sharp zero-bias peaks (ZBP) are observed on both of them, as shown in Figure 2(a). ZBP is the strongest right on the PSP molecules, and it gradually weakens as the tip moves away. However, when two PSP molecules are closer than those in dimer #1, such as in dimers #2-#4 shown in Figure 2(b)-(d), the ZBP symmetrically splits into two peaks with respect to $E_F$. The splitting magnitude strongly depends on the relative positions of two edge molecules. For dimer #4 shown in Figure 2(d), where the two PSP molecules are extremely close and side by side, the splitting of the ZBP is the largest, thus reaching 8-10 meV. In terms of dimers #2 and #3, where the two PSP molecules are staggered, the splitting is weaker than it is for dimer #4.

A ZBP observed in dilute magnetic metals [2-15] or in quantum dots [16-25] has been attributed to the Kondo effect. Here we examine if the ZBP spectrum observed in our work fits the Kondo singlet scenario. As shown in Figure 3(a), the ZBP could be well fitted by a Fano lineshape, with a form factor $q$ of $\sim 43$. Such a large $q$ indicates that the tunneling in the possible Kondo resonance state dominates over directly tunneling in the metal substrate, thus facilitating an easy extraction of spectroscopic properties. As shown in Figure 3(b),
with increasing temperatures, the ZBP broadens and its intensity decreases quickly. The peak width $\Gamma$ defined as the half-width at half-maximum (HWHM) of the ZBP, linearly grows with increasing temperature at higher temperatures but tends to saturate at low temperatures, as shown in Figure 3(c). The linewidth broadening with temperature can be well fitted via an approximated expression developed in the framework of Fermi-liquid theory for a Kondo singlet:

$$I(T) = \sqrt{(ak_BT)^2 + (2k_BT_K)^2},$$

where $k_B$ is Boltzmann’s constant, $a$ is an empirical parameter, and $T_K$ is the Kondo temperature [8,14,49,50]. $\Gamma$ at zero temperature defines the Kondo temperature $T_{K0} = 2\Gamma/k_B$; thus, $T_K$ is determined as $(30\pm 2)$ K for dimer #1. Moreover, the temperature dependence of the conductance of a Kondo-induced peak can be fitted by an empirical expression that reproduces the numerical renormalization group results for spin-1/2 systems:

$$h(T) = h_0 \left( \frac{T_K^2}{(2^{1/s} - 1)^2 + T_K^2} \right)^s,$$

where $h_0$ is the $h$ value at zero temperature and $s$ is an empirical parameter of $0.22$ for the spin-1/2 system [14,51]. As shown in Figure 3(d), our data can be well fitted by this formula, which yields $T_K$ of $(30\pm 3)$ K and $s$ of $-0.21\pm 0.02$. Thus, the temperature dependence of ZBP is consistent with the predicted behavior for a spin-1/2 Kondo effect. Based on these quantitative analyses, our data clearly indicate that the observed ZBP can be attributed to the emergence of the Kondo effect. As the PSP molecule itself is nonmagnetic, the local moment possibly arises from an unpaired electron localized in an orbital of the PSP molecule due to K doping. In fact, the two step-like peaks at $\pm 34$ meV in the spectrum obtained at 5 K in Figure 3(b) have been identified as a phonon-assisted inelastic tunneling feature of the Kondo effect in molecular systems such as that of pure organic TTF-TCNQ (where TTF and TCNQ denote tetrathiafulvalene and tetracyanoquinodimethane, respectively) molecules on the Au(111) surface [13]. Please see sect. 5 of SM for more discussions on the origin of the molecular spin of PSP molecules.

The Kondo resonance of a K-doped PSP molecule makes it promising for studying how two Kondo singlets interact. As shown in Figure 4(a), when the intermolecular distance $d > 1.49$ nm, the Kondo peak remains intact with similar height and width (Figure 4(d), (f)). As $d$ decreases to below 1.49 nm, the peak starts to broaden with reduced height (Figure 4(b), (d)). Finally, peak splitting can be clearly resolved when $d < 1.3$ nm (Figure 4(c)). The smallest $d$ we can observe is $-0.76$ nm, likely limited by the finite size of the PSP molecule. Figure 4(d) plots the peak height (ZBC) as a function of $d$. The decrease of ZBC at $d < 1.49$ nm indicates that the peak starts to split. We therefore fitted the spectra of $1.30 < d < 1.43$ nm (Figure 4(b)) by two peaks with their respective HWHM fixed to a constant value similar to those in Figure 4(f). Figure 4(e) shows the splitting magnitude ($E_S$, energy separation of the two split peaks or fitted double peaks), which linearly decreases with increasing $d$. A linear fit of $E_S$ suggests that the splitting begins at $1.5$ nm.

In previous studies of 2IKM, the competition between Kondo screening and antiferromagnetic RKKY interaction $J$ was reported to cause the splitting of the Kondo resonance in the absence of a magnetic field [29-41]. With the increase of antiferromagnetic $J/T_K$, the two single-impurity Kondo resonances gradually transform into an antiferromagnetic singlet state, manifesting as the splitting of ZBP. The splitting magnitude $E_S$ is found to be $2J$ [29-41]. In our case, when two PSP molecules are far apart, they independently behave as Kondo impurities screened by itinerant electrons and demonstrate the usual Kondo resonance. When $d < 1.5$ nm, the intermolecular interaction becomes obvious; the exchange interaction between localized molecule spins, likely RKKY antiferromagnetic coupling mediated by the Au(111) substrate, causes the Kondo resonance peak to split. The well-formed double-peak spectra at small $d$ indicate the formation of an AFM singlet, although there is still certain Kondo screening in the small $J$ regime, as described by the 2IKM.

Figure 5(a) shows the temperature dependence of the AFM singlet state of dimer #4, whose $d = 0.82$ nm (Figure 2(d)).
As temperature increases, the splitting is quickly suppressed; the spectra at 18 and 20 K still exhibit a double-peak line-shape, whereas the spectra obtained above 25 K are almost identical to the Kondo peaks of a stand-alone molecule, which are overlaid on them. Furthermore, ZBC is a non-monotonic function of temperature with a maximum of ~25 K (Figure 5(b)). These signature peaks suggest that 25 K is the crossover temperature between Kondo and AFM singlets. We conducted a double-Fano-function fit to those data obtained below 25 K (see Figure S6 in SM), which yields the temperature dependence of $E_S$ in Figure 5(c). The extrapolation in Figure 5(c) suggests that $E_S$ diminishes at ~26 K. Therefore, our data indicate that the AFM singlet state evolves back to the Kondo singlet state above 26 K, and there is a transition from AFM to paramagnetic state at about 26 K for these two local spins. This can be understood when considering that Kondo screening exhibits a crossover behavior in temperature that persists above $T_K$. In addition, an intriguing feature in Figure 5(d) is that the peak width obtained from the double-Fano-function fit of the double-peak spectra and those from the fit to the single Kondo resonance spectra in Figure 3(c) are essentially the same and follow the same temperature dependence, which suggests the Kondo coupling is independent of intermolecular distance. For dimer #2 with a smaller $E_S$ (Figure 2(b)), a similar temperature dependence is observed, but the AFM singlet state transforms back to a Kondo singlet at a lower temperature of ~15 K (see Figure S7 in SM).

The RKKY coupling $J$ for a pair of molecular spins follows $J = J_0 \cos(2k_Fd)/d^3$, where $J_0$ is a constant, $k_F$ is the Fermi wave vector of conductance electrons, and $d$ is the distance between molecules. Because the decreased $d$ monotonously corresponds to the increased $E_S$, or $2J$, the intermolecular
interaction should be antiferromagnetic in the examined range of 0.8-1.5 nm (Figure 6(a)). We then fit the experimental $J$ values to the abovementioned formula and obtained $k_F \approx 1.49 \text{ nm}^{-1}$, close to the previously reported values of $k_F = 1.62$ and $1.85 \text{ nm}^{-1}$ considering the Rashba splitting of Au(111) surface states [52]. As per our fit, values of $J$ at $d > 1.5 \text{ nm}$ are ferromagnetic and negligibly small; thus, it would not affect the Kondo singlet behavior for molecules farther apart than 1.5 nm, consistent with the experiment.

The phase diagram presented in Figure 6(b) summarizes our findings in this two-Kondo-impurity system. With increased AFM RKKY interactions, we observed a crossover from a Kondo singlet state to an AFM singlet—the Kondo singlets start to interact when there is only a small amount of $J$, while the largest observed $J$ is about 5 meV or $1.95 k_B T_K$. We cannot determine if the AFM singlet state is partially formed (meaning there is still certain Kondo screening as shown in Figure 6(b)) or is completely developed with the largest achievable experimental $J$ here, which requires to be further studied in the future. Note that, in theory, a quantum phase transition would occur at $J^* \approx 2.2-2.5 k_B T_K$, when there is particle-hole symmetry; this is not the case here.

$\#4$ can be fitted with a line going through the zero point. This line defines the boundary between the Kondo singlet regime and AFM singlet regime (including the crossover regime with a partially formed AFM singlet and the regime with a completely developed AFM singlet). At first sight, this is hard to rationalize in the context of the standard 2IKM [53]. As illustrated in Figure 5, although the RKKY interaction $J$ is $1.95 k_B T_K$ at 5 K for dimer $\#4$, the AFM coupling of the two spins diminishes at $\approx 0.85 T_K$. However, this result may be understood by considering a quickly diminishing RKKY interaction with increasing temperature and the persistence of Kondo screening above $T_K$.

Therefore, our experimental findings are relevant to investigate not only the artificial nanostructures but also strongly correlated bulk systems. The complex phase diagram of the 2IKM in our study is reminiscent of multiple magnetic heavy fermion systems in which both magnetism from the RKKY interaction and heavy quasiparticles from the Kondo effect are prominent even deep inside the magnetically ordered phase, as a result of the dual character of 4f electrons [26-28,54]. 2IKM is the minimum model system and starting point to study the magnetic Kondo lattice and heavy fermion systems, which in principle possess the basic physics of the Kondo lattice. Moreover, being able to mea-
sure the properties of two magnetic molecular impurities is a prerequisite for understanding and designing complex nanostructures such as complex Kondo clusters or even a 1D or 2D engineered Kondo lattice.

4 Conclusion

We find that one of the self-assembled ordered phases of K-doped single-layer PSP/Au films is unexpectedly an excellent platform to study the 2IKM. One can study the transformation between a Kondo singlet and an AFM singlet as a function of temperature and RKKY interaction strength. Our findings depict a comprehensive experimental phase diagram of the 2IKM, thus highlighting the subtle interplay between RKKY interaction and Kondo screening. This, in turn, may provide critical clues for understanding magnetic Kondo lattice and heavy fermion systems. The revealed experimental energy and temperature scales put constraints on and deserve further investigations of theories and calculations of the 2IKM. Moreover, our findings suggest that complex Kondo clusters and even 1D or 2D Kondo lattices may be developed using such molecular systems.

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Supporting Information

The supporting information is available online at phys.scichina.com and http://link.springer.com/journal/11433. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

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