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Demonstrating Kondo behavior by temperature-dependent scanning tunneling spectroscopy

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The Kondo effect describes the scattering of conduction electrons by magnetic impurities, manifesting as an electronic resonance at the Fermi energy with a distinctive temperature evolution. In this Letter, we present a critical evaluation of the current methodology employed to demonstrate Kondo behavior in transport measurements, underscoring the limitations of established theoretical frameworks and the influence of extrinsic broadening. We introduce an approach for analyzing spectroscopic indicators of the Kondo effect, employing the Hurwitz-Fano lineshape as a model for the Kondo resonance in the presence of extrinsic broadening. Through precise scanning tunneling spectroscopy measurements on an exemplary spin-1/2 Kondo system, phenalenyl on Au(111), we demonstrate the efficacy of our proposed protocol in extracting accurate intrinsic Kondo linewidths from finite-temperature measurements. The extracted linewidths exhibit a robust fit with a recently derived expression for the temperature-dependent intrinsic Kondo linewidth, providing compelling evidence for the validity of the underlying theory.

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The Kondo effect is one of the most enigmatic phenomena in condensed matter physics and one of the hallmarks of strong electronic correlations [1–4]. It occurs when a magnetic impurity interacts with the conduction electrons in a metallic host. Below a characteristic temperature, called the Kondo temperature, $T_K$, the impurity spin is effectively screened by the formation of a total spin-singlet state with the surrounding conduction electrons. As the low-temperature dynamics of individual Kondo impurities are governed by the energy scale $k_B T_K$, its magnitude has a major influence on the ground states and quantum phase diagrams of strongly correlated materials, such as Kondo lattices and heavy fermion systems [5–9].

The Kondo effect is signaled by the appearance of a sharp resonance at the Fermi level in the density of states of the magnetic impurity. This can be exploited for the detection of magnetic moments in atoms and molecules adsorbed on metallic substrates by scanning tunneling spectroscopy (STS), where it shows up as a zero-bias anomaly in the conductance ($dI/dV$) spectra [10–14]. However, a clear-cut proof of Kondo behavior requires to discriminate the Kondo resonance from other zero-bias anomalies. This can be achieved, for example, by measuring the temperature evolution of the resonance’s linewidth [15–19], which shows a characteristic universal behavior in the Kondo regime [20,21].

Attempts to derive an analytic expression for the temperature dependence of the Kondo peak from Fermi liquid theory [15] were shown to be problematic due to the limitation to very low temperatures $T \ll T_K$ and energies $\omega \ll \Gamma_K$ [22,23]. As a consequence, empirical expressions for the temperature-dependent Kondo linewidth are often used to fit experimental data, e.g., $\Gamma_{\text{emp}}(T) = \sqrt{(\alpha k_B T)^2 + 2(k_B T_K )^2}$ where $\alpha$ was introduced as an additional fitting parameter instead of $\alpha = \pi$ as originally defined in Ref. [15]. Due to the free parameter $\alpha$ and different definitions for the Kondo temperature (e.g., $T_{K,N} = 2.77 T_K$) [24], various forms for $\Gamma_{\text{emp}}(T)$ can be found in the literature [16,17,19,25–27]. It has also been noted that simple square-root expressions such as $\Gamma_{\text{emp}}(T)$ cannot capture the universal scaling behavior obtained in accurate numerical renormalization group calculations [28].

Recently, an analytic equation for the temperature-dependent linewidth of the Kondo peak was derived from a novel theoretical Ansatz for the renormalized self-energy that...
molecules deposited on an Au(111) surface [24]. The unpaired
system, and provide an efficient protocol to experimentally
π measurements and analysis of a prototypical spin-1
reported in Ref. [23] by accurate temperature-dependent mea-
proved the agreement. Therefore, a clear experimental proof
in the STS data [30]. Post-hoc removal of extrinsic broadening
likely due to the presence of extrinsic broadening mechanisms
in the STS measurement [30]. Thus, in order to
obtain a larger temperature range. Fitting Eq. (1) to the
merged data set yields a relatively poor fit as shown by the red
line in Fig. 1(b), although somewhat better than the empirical expression \( \Gamma_{\mathrm{emp}}(T) \) with fixed temperature coefficient \( \alpha = \pi \)
(gray solid line), similar to the finding in Ref. [23].

We will now see that the mismatch with Eq. (1) is caused by
extrinsic broadening mechanisms. In STS, the conductance
spectra \( (dI/dV) \) are measured at finite temperatures \( T \)
where the Kondo peak is broadened (i) due to the intrinsic
temperature dependence of the Kondo peak [15,21,23], and
(ii) due to the presence of different extrinsic broadening
mechanisms in the STS measurement [30]. Thus, in order to
obtain the actual intrinsic halfwidth \( \Gamma_{\mathrm{K}} \) of the Kondo peak
and demonstrate Kondo behavior, one has to first remove the
extrinsic contributions from the measured HWHM. Assuming
a noise-optimized (electronic and mechanical) experimental
setup, the two main sources of extrinsic broadening are caused
by Fermi-Dirac (FD) broadening of the tip and the voltage
modulation for the lock-in detection. A good fit with \( \Gamma_{\mathrm{emp}}(T) \)
can only be obtained by using \( \alpha \) as a free fitting parameter
dashed gray line).

In order to properly incorporate the most important broad-
ealing mechanisms into the analysis of the STS data we now
resort to theory. The experimental situation of STS is depicted
schematically in Fig. 2(a): A molecule (M) on a metallic
substrate (S) is probed by an STM tip (T). Application of a voltage \( V \) to the sample drives a current \( I \) from the sample
electrode via the molecule to the tip electrode. Typically, in
STS the coupling of M to T is much weaker than the coupling
of M to S. In this situation, also called ideal STM limit [34],
M is effectively in equilibrium with S, and the \( dI/dV \) of the
current from the tip to the molecule at the substrate is given by the convolution
\[
G(V) \equiv \frac{dI}{dV} \propto \int d\omega \{ -f'(\omega) \} A(\omega + eV).
\] (2)
where \( f'(\omega) = -\beta/(4\cosh^2(\beta\omega/2)) \) is the derivative of the tip’s FD distribution, \( \beta = 1/k_BT \) with \( T \) the temperature at
the tip, and \( A(\omega) \) the spectral function of the molecule [35].
We assume a fully thermalized system so that tip and sample
have the same temperature \( T \).

The convolution of the spectral function \( A(\omega) \) with the FD
derivative \( f'(\omega) \) leads to broadening of the Kondo resonance,
to which we now simply refer to as FD broadening. This is

FIG. 1. (a) \( dI/dV \) spectrum of phenalenyl from DS2 at \( T = 1.56 \) K (black dots) showing the Kondo resonance, fitted by Frota-
Fano lineshape (green line). Insets I and II show the theoretical and experimental spatial distribution of the unpaired electron [19].
(b) Kondo linewidths extracted from Frota fits to \( dI/dV \) spectra of DS1 (blue circles) and DS2 (green circles) versus temperature, and fits to Eq. (1) (red solid line), and to \( \Gamma_{\mathrm{emp}}(T) \) with fixed \( \alpha = \pi \) (gray solid line) and with \( \alpha \) as fit parameter (gray dashed line, \( \alpha = 7.45 \)). Error bars show the estimated standard deviation.

extends the temperature and energy range beyond the Fermi
liquid regime [23]:
\[
\Gamma(T) = \Delta_K \cdot \sqrt{a + b \cdot \left[1 + \left(\frac{\tau}{\Delta_K}\right)^2\right] + c \cdot \left(\frac{\tau}{\Delta_K}\right)^2},
\] (1)
where \( a = 1 + \sqrt{3} \approx 2.732, \ b = 2 + \sqrt{3} \approx 3.732, \) and \( c = \sqrt{3}/2 \approx 0.866 \) are constants, \( \tau = \pi k_B T \) is the temperature
parameter, and \( \Delta_K \) the width parameter of the T = 0 Kondo
peak, related to the halfwidth via \( \Gamma_K = 2.542 \Delta_K \) and to the
Kondo temperature via \( \Delta_K = 1.542 k_B T_K \) [29]. As shown in
Ref. [23], Eq. (1) is in excellent agreement with numerical renormalization group calculations [21]. On the other hand,
experimental data of Kondo linewidths versus temperature measured by STS [16] could not be fitted very well, most likely
due to the presence of extrinsic linewidths only slightly
improved the agreement. Therefore, a clear experimental proof
of the theory of Ref. [23] is still lacking.

In this work, we demonstrate the validity of the theory
reported in Ref. [23] by accurate temperature-dependent mea-
measurements and analysis of a prototypical spin-1/2 Kondo
system, and provide an efficient protocol to experimentally
prove the Kondo nature of a zero-bias peak. To this end, we
conducted low-temperature STM experiments on phenalenyl
molecules deposited on an Au(111) surface [24]. The unpaired
\( \pi_e \) electron of phenalenyl forms an \( S = 1/2 \) ground state
and is uniformly delocalized over six equivalent positions, as
shown in the two insets of Fig. 1(a) where the spin density
plot (inset I) and the experimental constant-height map of the
Kondo resonance (inset II) are reported (more details in Ref.
[19]). The high-resolution STS spectra shown in this work
were measured with a metal STM tip and acquired as point
spectra on one of the six equivalent Kondo lobes, as depicted
in inset II of Fig. 1(a).

Two distinct data sets of temperature-dependent \( dI/dV \)
spectra were acquired on two different molecules, which are
simply adsorbed on the face centered cubic (fcc) region of the
Au(111) herringbone reconstruction. We refer to the
lower temperature data set as DS1, while DS2 is the data
set from 1.56 K to 7.5 K. For each temperature the Kondo
peak was fitted with the Frota-Fano lineshape [example shown
in Fig. 1(a)], taking quantum interference into account via
the Fano phase \( \phi \) [31–33], \( F(V) = F_0 \cdot \text{Re}[e^{i\phi}/\sqrt{1 + (\pi V/\Delta/K)^2}] \)
where \( F_0 \) is the amplitude, and \( \Delta \) the Fano width parameter
related to the halfwidth of the Frota-Fano lineshape by \( \Gamma = \sqrt{3 + \pi^2}/\Delta = 2.542 \Delta \). The Frota-Fano lineshape yields
good fits for all temperatures [24]. The obtained halfwidths of
the Frota fits are shown in Fig. 1(b) for both data sets. The
two data sets overlap neatly in the temperature range where
both molecules have been measured, i.e., between 1.5 K and
3 K. This justifies the merging of both data sets in order
to obtain a larger temperature range. Fitting Eq. (1) to the
merged data set yields a relatively poor fit as shown by the red
line in Fig. 1(b), although somewhat better than the empirical expression \( \Gamma_{\mathrm{emp}}(T) \) with fixed temperature coefficient \( \alpha = \pi \)
(gray solid line), similar to the finding in Ref. [23].
FIG. 2. (a) Schematic STM setup for measuring $dI/dV$ spectra of a molecule (M) on a surface (S) by an STM tip (T). (b) Simulated $dI/dV$ spectra assuming a Frota-Fano lineshape in the spectral function at different temperatures. (c) HF lineshapes (red solid lines) according to Eq. (3) at finite temperature $T = 0.4 \Gamma_K/k_B$ and corresponding Frota-Fano lineshapes (blue dashed lines) in underlying spectral function for different values of $\phi$ and fixed halfwidth $\Gamma_K = 2.542 \Delta K$.

FIG. 3. (a) $dI/dV$ spectra of DS1 and DS2 (black dots) fitted with HF lineshapes according to Eq. (3) (blue and green solid lines). (b) Intrinsic Kondo linewidths obtained from HF fits (transparent blue and green circles) versus temperature, fitted with Eq. (1) (red solid line). Blue and green circles show intrinsic linewidths obtained from fitting with Eq. (4), taking into account both FD and lock-in broadening, fitted with Eq. (1) (blue and green solid lines). Blue and green circles show intrinsic linewidths obtained from fitting with Eq. (4) with constant intrinsic linewidth. Also shown are temperature corrected Frota linewidths (blue circles) using $\Gamma_{corr}$ (see text). Gray lines show how the experiment data slightly better than the corresponding Frota fits [24]. Fitting the spectra with the HF lineshape yields smaller mean squared errors and the obtained width parameter $\Delta$ is also more consistent with varying the fit range [24]. Importantly, the Frota width parameter $\Delta$ resulting from each fit now yields the intrinsic halfwidth $\Gamma(T) = 2.542 \Delta$ of the Kondo resonance for a given temperature $T$. As a result, the extracted halfwidths do follow the predicted intrinsic temperature broadening and can be fitted well by Eq. (1), as shown by the transparent red line in Fig. 3(b).

Hurwitz-Fano (HF) lineshapes according to (3), as red solid lines corresponding to three different Frota-Fano lineshapes (shown as blue dashed lines) in the underlying spectral function $A(\omega)$ with different Fano phases $\phi$ as indicated in the plots. The HF lineshape (3) can now be fitted [36] directly to the Kondo resonances in the finite-temperature $dI/dV$ spectra recorded by STS, as shown in Fig. 3(a). We find that it fits the experimental data slightly better than the corresponding Frota fits [24]. Fitting the spectra with the HF lineshape yields smaller mean squared errors and the obtained width parameter $\Delta$ is also more consistent with varying the fit range [24]. Importantly, the Frota width parameter $\Delta$ resulting from each fit now yields the intrinsic halfwidth $\Gamma(T) = 2.542 \Delta$ of the Kondo resonance for a given temperature $T$. As a result, the extracted halfwidths do follow the predicted intrinsic temperature broadening and can be fitted well by Eq. (1), as shown by the transparent red line in Fig. 3(b). This is in stark contrast to the halfwidths extracted from the corresponding Frota fits in Fig. 1(b).
Another important contribution to extrinsic broadening in STS experiments comes from the lock-in modulation of the bias voltage. The effect on the spectra can be described by a further convolution of the differential conductance $G(V)$ [27,30] as

$$G(V) = \int dV' \chi_m(V') G(V + V'),$$

where the lock-in function $\chi_m$ is given by $\chi_m(V) = 2\sqrt{V_m^2 - V^2}/\pi V_m^2$ for $V \leq V_m$, and $\chi_m(V) = 0$ otherwise. $V_m = \sqrt{2} V_{ms}$ is the amplitude of the bias modulation. Thanks to the analytic solution (3) of the first convolution (2), this second convolution can be computed numerically efficiently enough to be used in the fitting procedure [36]. The resulting Frota width parameter $\Delta$ yields the Kondo width without the extrinsic broadening due to FD smearing or lock-in modulation. Thus, $\Gamma = 2.524\Delta$ now essentially yields the intrinsic halfwidth of the Kondo resonance at a given temperature $T$ since the two most important STM inherent contributions of extrinsic broadening have been removed. Figure 3(b) shows in blue and green circles the intrinsic Kondo halfwidths extracted in this way, which can be fitted even better by Eq. (1), resulting in a mean square error of MSEHL = 1870 $\mu$V$^2$ compared to MSEHL = 2474 $\mu$V$^2$ for the fits using just the HF lineshape. In order to visualize the effect of FD broadening in our experimental dataset (DS1 and DS2), in the Supplemental Material [24] we compare the linewidths from Fig. 1(b) (Frota fit) with the linewidths from Fig. 3(b) (Hurwitz fit), emphasizing the importance of removing extrinsic FD broadening.

So far we have shown the advantage of using the HF lineshape in order to directly obtain the intrinsic halfwidth of a Kondo resonance, as well as the validity of Eq. 1 to describe the correct temperature-dependent broadening of the intrinsic halfwidth. The combination of these two expressions is crucial for the verification of a Kondo resonance by its temperature dependence, as illustrated in Fig 3(c) for a simulated data set where a Frota lineshape of constant halfwidth ($\Gamma = 3$ mV) is broadened by FD and lock-in modulation ($V_m = 0.4$ mV). Fitting this data set with a Frota-Fano lineshape (blue dots) [24] yields an increasing halfwidth, whose temperature dependence can be fitted well with the empirical expression $\Gamma_{emp}(T)$, thus giving the false impression of Kondo behavior. Also the often employed temperature correction of the linewidth $\Gamma_{corr} = \sqrt{\Gamma^2 - (1.75k_B T)^2}$ [16,19,25] does not properly compensate for the FD broadening, as shown by the blue circles in Fig 3(c). Using the HF lineshape, on the other hand, yields the correct constant linewidth of the underlying peak (red circles), revealing its non-Kondo nature. Similar results are obtained for a FD-broadened Lorentzian peak with a constant linewidth [24]. The empirical expression $\Gamma_{emp}(T)$ is therefore not a suitable proof for Kondo resonances and can only be used as an empirical way to extrapolate the Kondo temperature from such data sets if the presence of the Kondo effect can already be assumed.

On the other hand, if no proof of Kondo behavior is required, there is no need to perform temperature-dependent measurements in order to determine $T_K$. According to (1) the intrinsic halfwidth $\Gamma$ at a certain temperature depends only on $\Delta_K$ and the (known) experimental temperature. The relation

$$\Delta_K = \sqrt{\frac{1}{\tau^2} \Gamma^4 + \frac{1}{4} \Gamma^2 \tau^4 + \frac{1}{3} \Gamma^2 \tau^4 - \frac{1}{4} \beta \tau^4 - \gamma T^2},$$

where $\alpha \equiv (2 + \sqrt{3})/6 \approx 0.622$, $\beta \equiv 1 - 1/\sqrt{12} \approx 0.711$ and $\gamma \equiv 1 - 1/\sqrt{3} \approx 0.423$ are constants and $\tau \equiv \pi k_B T$ the temperature parameter. Using this relation, the Kondo temperature $T_K = \Delta_K/1.542k_B \equiv \Gamma_K/3.92k_B$ [29] can be determined accurately from the intrinsic halfwidth of a single spectrum, taken at finite temperature. Figure 4(a) displays the $T_K$ obtained from the individual spectra of data sets DS1 and DS2 in dark blue and green circles, respectively. There is less than 3% of variation between the individually determined Kondo temperatures and the $T_K$ obtained before by fitting Eq. (1) to the linewidths as a function of temperature. Similarly consistent Kondo temperatures are found by applying this method to temperature-dependent data sets reported elsewhere [24]. The slight deviations from the expected constant behavior of the extracted $T_K$ versus temperature may be attributed to experimental reasons, such as incomplete thermalization at higher temperatures [30].

We now apply the developed methodology to investigate the dependence of the Kondo temperature on the adsorption site of phenalenyl on the herringbone reconstruction of the Au(111) surface. Along with data sets DS1 and DS2, acquired on two molecules in the fcc region of the Au(111) reconstruction close to the herringbone ridge (adsorption site labeled as FCC*), we also carried out $dV/dI$ measurements for phenalenyl adsorbed in the middle of the fcc (light blue) and hcp (orange) regions. By fitting each spectrum with a lock-in broadened HF lineshape and calculating $T_K$ using (5), we find that different adsorption geometries result in different Kondo temperatures, as clearly shown in Fig. 4(a). This observation can be rationalized by slight changes in the hybridization between molecule and metal substrate and

![FIG. 4. (a) Kondo temperatures $T_K(T)$ determined for each $dV/dI$ spectrum individually by HF + Lock -- in fits [Eq. (4)] and using the resulting intrinsic linewidth $\Gamma$ in Eq. (5). Error bars represent the estimated standard deviations. Red dashed lines indicate $T_K$ obtained by fitting the temperature dependence of the Kondo linewidth for each adsorption site with Eq. (1) [24]. (b) STM images of phenalenyl molecules on Au(111) with boxes highlighting the investigated molecules. Scanning parameters for top image are $V = -2$ V, $I = 100$ pA and $V = -0.1$ V, $I = 50$ pA for the bottom image. Ridges of the herringbone reconstruction are marked with dashed white lines.](Image)
the exponential dependence of the Kondo temperature on the hybridization [37]. The robustness of the single-point $T_K$ determination, here shown for different adsorption sites and on an entire temperature series, justifies the use of this method as a fast and reliable route to estimate $T_K$ of a Kondo system [38].

To conclude, we have performed accurate STS measurements of a pure spin-1/2 Kondo system, and developed tools for an efficient and accurate data analysis that properly takes into account extrinsic broadening of $dI/dV$ spectra in STS. A key element of the methodology is an analytic expression for the Fermi-Dirac broadened Kondo lineshape in terms of the Hurwitz $\zeta$ function. Fitting the spectra with this Hurwitz-Fano lineshape yields the intrinsic width of the Kondo peak which fits very well with the recently derived expression (1) for the Kondo width as a function of temperature, proving the validity of the theory [23]. The procedure developed here allows to unequivocally prove Kondo behavior of a system probed by STS [36]. In contrast the established methodology of fitting the empirical expression $\Gamma_{\text{emp}}(T)$ to linewidths extracted from Frota fits to STS data may give a false impression of Kondo behavior. Finally, our methodology also allows to obtain the intrinsic Kondo width at $T = 0$ and corresponding Kondo temperature $T_K$ from a single spectrum at finite temperature.

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E.T. and M.A. contributed equally to this work.

[28] See Supplemental Note 12 of Ref. [39].
[29] Here as in Ref. [23] we use Wilson’s thermodynamic definition of $T_K$ [40], corrected by Wiegman and Tsvelick [41]. As shown in Ref. [23] $T_K$ is related to the halfwidth $\Gamma_K$ of the Kondo peak by $T_K \sim \Gamma_K/3.92$.
[35] For noninteracting electrons, Eq. (2) can be derived from Bardeen tunneling theory, see, e.g., Ch. 21.8 in Ref. [42]. For interacting electrons it can be derived from the Meir-Wingreen equation [43] assuming the ideal STM limit, see Ref. [34].
[36] Python and Igor programs for fitting Hurwitz and Hurwitz*Lock – in line shapes are publically available on GitHub at https://github.com/david-jacob/HurwitzFanoFit.
[38] An alternative route for determining $T_K$ from STS at $T \sim T_K$, recently proposed in Ref. [44], requires additional control parameters in the experiment such as magnetic field or mechanical gating.