

Measuring the out-of-equilibrium splitting of the Kondo resonance

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An experiment is proposed to measure the out-of-equilibrium splitting of the Kondo resonance in an ultrasmall quantum dot, by adding a third, weakly coupled lead to the standard two-lead quantum-dot system, and sweeping the chemical potential of that lead. Fixing the voltage bias between the source and drain leads, we show that the differential conductance for the current through the third lead traces the out-of-equilibrium dot density of states (DOS) for the two-lead system. This enables one to measure the dot DOS in the presence of an applied voltage bias. We show that this method is robust, and extends also to the case where the coupling to the third lead is no longer weak.

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The understanding of strong electronic correlations far from thermal equilibrium is one of the challenging problems in contemporary mesoscopic physics. A primary example is the out-of-equilibrium Kondo effect, recently measured in ultrasmall quantum dots.¹⁻⁴ In the Kondo effect, an impurity moment undergoes a many-body screening by the conduction electrons, producing a narrow low-energy resonance in the impurity density of states (DOS) at low temperatures.⁵ This Kondo resonance is responsible for the well-known enhancement of the low-temperature resistivity in dilute magnetic alloys, and for the enhancement of the low-temperature conductance in tunneling through small quantum dots with an odd number of electrons. One of the striking predictions for the out-of-equilibrium Kondo effect in quantum dots is the splitting of the Kondo resonance in the dot DOS for a finite applied bias.⁶

Although well-established theoretically, it remains unclear whether one can measure the above splitting in the dot DOS. The splitting of the Kondo resonance does not show up in the differential conductance for the current through the dot since, unlike in conventional noninteracting systems, the DOS itself is strongly voltage dependent. Photoemission, in addition to being limited in resolution, measures the “occupied” DOS, i.e., the product of the DOS and the effective distribution function. Since the latter distribution is unknown away from thermal equilibrium, one cannot reliably extract the underlying DOS using photoemission. A key assumption in conventional tunneling measurements of the DOS is that the weakly coupled sample and probe are each effectively in equilibrium. Clearly this assumption breaks down for the quantum dot, which raises the fundamental question: Can one actually measure the DOS of an interacting mesoscopic system far from thermal equilibrium?

In this paper we show that the out-of-equilibrium splitting of the Kondo resonance can indeed be measured, by adding a third, weakly coupled lead to the standard two-lead quantum-dot system.⁷ Fixing the voltage bias between the source and drain leads and sweeping the chemical potential of the third lead, we show that the differential conductance for the current through that lead traces the out-of-equilibrium two-lead dot DOS, much in the same way as in conventional tunneling measurements of the equilibrium DOS. In this manner, one can measure the dot DOS in the presence of an applied volt-

age bias. Furthermore, we find that the splitting of the Kondo resonance can be observed also at temperatures above the Kondo temperature, and that the same qualitative features are obtained when the coupling to the third lead is no longer weak. Contrary to a previous proposal to probe the splitting of the Kondo resonance using two capacitively coupled quantum dots,⁸ our approach allows for a direct measurement of the nonequilibrium DOS.

Figure 1 shows the proposed experimental setting. It consists of an ultrasmall quantum dot, coupled to three metallic leads. Each lead is kept at a separate chemical potential μ_α ($\alpha=1,2,3$), and is coupled to the dot via a tunneling matrix element t_α . The energy level on the dot E_D is controlled by applying a gate voltage. Modeling the charging energy on the

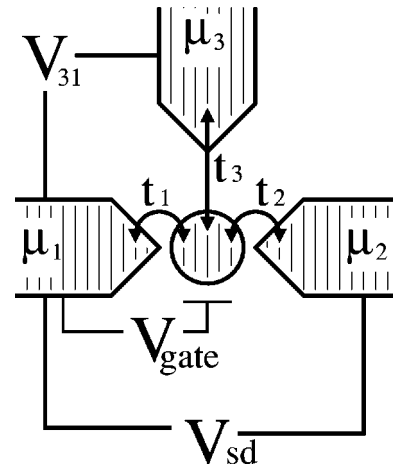


FIG. 1. A schematic sketch of the proposed apparatus. An ultrasmall quantum dot is coupled by tunneling to three metallic leads, each of which is kept at a separate chemical potential. The corresponding tunneling matrix elements are controlled by varying the potential barriers, while the dot energy level is adjusted by applying a gate voltage. The dot level and the couplings are to be tuned such that the dot is in the Kondo regime, and the coupling to the third lead is much weaker than the other two leads. Fixing the source-drain voltage bias at $\mu_2 - \mu_1 = eV_{sd}$ and sweeping the chemical potential μ_3 by varying V_{31} , one measures the current $I_3(V_{31})$ between the dot and the third lead. Up to thermal broadening and rescaling, the differential conductance $G_3 = dI_3/dV_{31}$ traces the out-of-equilibrium two-lead dot DOS.

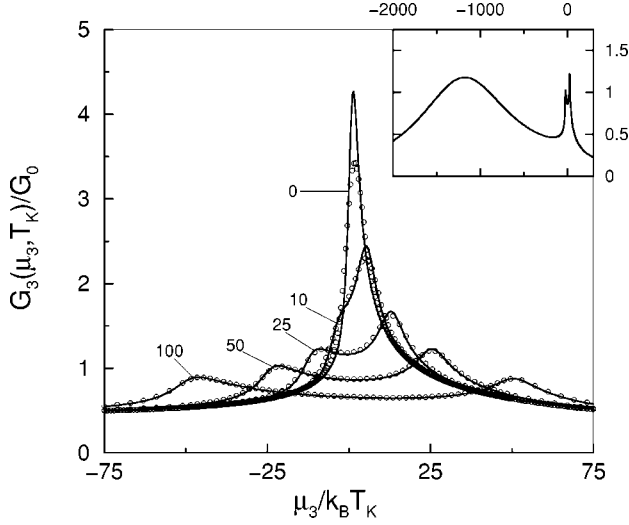


FIG. 2. Comparison of the out-of-equilibrium dot DOS for a two-lead system (full lines) with the differential conductance $G_3(\mu_3)$ in the proposed three-lead setting (empty circles). All calculations were performed using the NCA. Here $\Gamma_1 = \Gamma_2 = D/30$ (symmetric coupling), $E_d/D = -0.278$, $\Gamma_3/\Gamma_1 = 0.01$, $T = T_K$, and $G_0 = 4e^2\Gamma_3(\Gamma_1 + \Gamma_2)/D\hbar\Gamma$. The two-lead dot DOS is scaled by the bandwidth D . The source-drain voltage bias, $\mu_2 - \mu_1 = eV_{sd}$, takes the values $eV_{sd}/k_B T_K = 0, 10, 25, 50$, and 100 , as indicated by the attached labels. Up to thermal broadening, the scaled differential conductance coincides with the out-of-equilibrium dot DOS, in accordance with Eq. (6). Inset: An extended image of the dot DOS for a two-lead system with $T = T_K$ and $eV_{sd}/k_B T_K = 50$.

dot by a Hubbard on-site repulsion U , the system is described by a generalized three-lead Anderson impurity model

$$\mathcal{H} = \sum_{\alpha=1}^3 \sum_{k,\sigma} [(\epsilon_k + \mu_\alpha) c_{\alpha k \sigma}^\dagger c_{\alpha k \sigma} + t_\alpha (c_{\alpha k \sigma}^\dagger d_\sigma + \text{H.c.})] + E_D \sum_{\sigma} d_\sigma^\dagger d_\sigma + U d_\uparrow^\dagger d_\uparrow d_\downarrow^\dagger d_\downarrow. \quad (1)$$

Here $c_{\alpha k \sigma}^\dagger$ creates an electron with wave number k and spin projection σ in lead α , while d_σ^\dagger creates a dot electron with spin projection σ . In the following we assume that the tunneling matrix elements and the energy level of the dot are adjusted in such a way that a stable local moment is formed on the dot. This corresponds to the condition $\Gamma \equiv \sum_{\alpha=1}^3 \Gamma_\alpha \ll -E_D, E_D + U$, where $\Gamma_\alpha = \rho_\alpha \pi t_\alpha^2$ is the hybridization width associated with lead α (ρ_α is the conduction-electron DOS at the Fermi level of lead α). We shall generally regard Γ_3 as much smaller than Γ_1 and Γ_2 , though comparable couplings will also be considered.

The main quantities of interest are the electrical currents flowing from the dot to each of the three leads. The electrical current flowing into lead α is given by the nonequilibrium average of the current operator $\hat{I}_\alpha = it_\alpha (e/\hbar) \sum_{k\sigma} \{c_{\alpha k \sigma}^\dagger d_\sigma - \text{H.c.}\}$ ($-e$ is the electron charge). This average is conveniently expressed via the lesser and retarded Keldysh Green functions for the dot electrons:

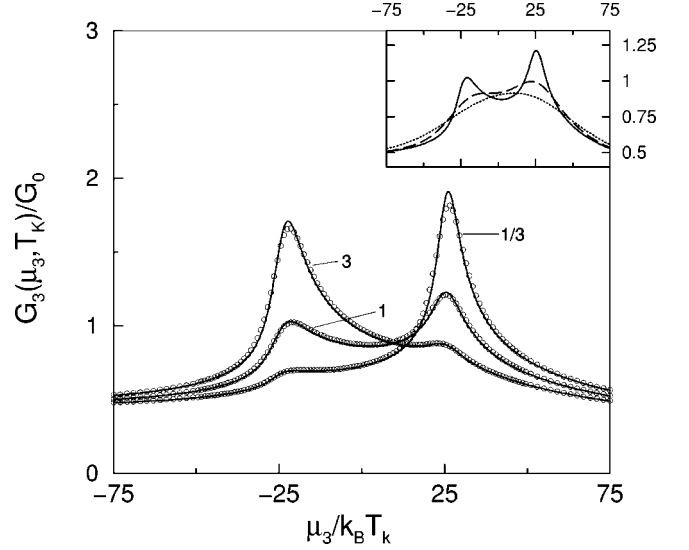


FIG. 3. The out-of-equilibrium two-lead dot DOS (full lines) versus the three-lead differential conductance $G_3(\mu_3)$ (empty circles), for a fixed source-drain voltage bias of $eV_{sd}/k_B T_K = 50$, and different ratios of Γ_1 to Γ_2 . Here $(\Gamma_1 + \Gamma_2)/D = 0.067$ is kept fixed as to maintain the same Kondo temperature, $k_B T_K/D = 2.5 \times 10^{-4}$. The remaining parameters are $E_d/D = -0.278$, $\Gamma_3/(\Gamma_1 + \Gamma_2) = 0.005$, $T = T_K$, and $G_0 = 4e^2\Gamma_3(\Gamma_1 + \Gamma_2)/D\hbar\Gamma$. The ratio Γ_1/Γ_2 takes the values $1/3, 1$, and 3 , according to the labels attached. The effect of an asymmetry in Γ_1 and Γ_2 is to enhance (reduce) the peak at the chemical potential of the more strongly (weakly) coupled lead. Inset: Temperature dependence of $G_3(\mu_3)$, for $eV_{sd}/k_B T_K = 50$. Full, dashed and dotted lines correspond to $T/T_K = 1, 5$, and 10 , respectively. The effect of a temperature is to smear the peak structure of $G_3(\mu_3)$.

$$G_{d\sigma}^<(\epsilon) = \int_{-\infty}^{\infty} \langle d_\sigma^\dagger d_\sigma(t) \rangle e^{i\epsilon t} dt, \quad (2)$$

$$G_{d\sigma}^r(\epsilon) = -i \int_0^{\infty} \langle \{d_\sigma(t), d_\sigma^\dagger\} \rangle e^{i\epsilon t} dt. \quad (3)$$

Specifically, generalizing the expression of Meir and Wingreen⁹ from two to three leads one obtains

$$I_\alpha = \frac{4e}{h} \Gamma_\alpha \int_{-\infty}^{\infty} [2\pi f(\epsilon - \mu_\alpha) A_d(\epsilon) - G_d^<(\epsilon)] \nu_\alpha(\epsilon - \mu_\alpha) d\epsilon, \quad (4)$$

where $A_d(\epsilon) = -(1/\pi) \text{Im}\{G_d^r(\epsilon)\}$ is the dot spectral function (i.e., DOS); $f(\epsilon)$ is the Fermi-Dirac distribution function; and $\nu_\alpha(\epsilon) = \rho_\alpha(\epsilon)/\rho_\alpha$ is the reduced conduction-electron DOS in lead α . Here we use the notation by which the arguments of $\rho_\alpha(\epsilon)$ and $\nu_\alpha(\epsilon)$ are measured relative to the corresponding chemical potential, μ_α . Note that we have restricted ourselves in Eq. (4) to a zero magnetic field, in which case all spin dependences drop.

The integration over energy in Eq. (4) contains a natural cutoff, namely, the maximal chemical-potential difference between the leads, broadened by the temperature. To the extent that one can neglect the energy dependence of $\nu_\alpha(\epsilon)$ on

that scale, it is possible to eliminate $G_d^<$ from the expression for I_3 using the identity $I_3 = I_3(\Gamma_1 + \Gamma_2)/\Gamma - (I_1 + I_2)\Gamma_3/\Gamma$:

$$I_3 = \frac{4e\Gamma_3}{\hbar\Gamma} \int_{-\infty}^{\infty} A_d(\epsilon) [(\Gamma_1 + \Gamma_2)f(\epsilon - \mu_3) - \Gamma_1 f(\epsilon - \mu_1) - \Gamma_2 f(\epsilon - \mu_2)] d\epsilon. \quad (5)$$

Analogous expressions apply to each of I_1 and I_2 .

Equation (5) generalizes the standard two-lead expression for tunneling through an Anderson impurity.⁹ It retains the familiar form of an integral of a spectral function times a weighted difference of Fermi functions. If $A_d(\epsilon)$ were insensitive to variations in the chemical potentials (as is the case for conventional noninteracting systems), then the derivative of I_3 with respect to any of the μ_α 's would have given the thermally broadened dot DOS at μ_α . Clearly, this is not the case for a quantum dot in the Kondo regime. Nevertheless, for $\Gamma_3 \ll \Gamma_1 + \Gamma_2$ one can still fix the chemical potentials μ_1 and μ_2 and differentiate the current I_3 with respect to μ_3 , to obtain the out-of-equilibrium two-lead dot DOS for a source-drain voltage bias of $\mu_2 - \mu_1 = eV_{sd}$. To see this we note that, for $\Gamma_3 \ll \Gamma_1 + \Gamma_2$, the impurity Green functions only weakly depend on the coupling to the third lead. Hence $A_d(\epsilon)$ and $G_d^<(\epsilon)$ are insensitive to variations in μ_3 , despite the strong dependence on μ_1 and μ_2 . Fixing μ_1 and μ_2 and sweeping the bias V_{31} (see Fig. 1), the differential conductance $G_3(\mu_3) = dI_3/dV_{31}$ then reduces to

$$G_3(\mu_3) \approx -\frac{4e^2\Gamma_3}{\hbar\Gamma} (\Gamma_1 + \Gamma_2) \int_{-\infty}^{\infty} A_d(\epsilon + \mu_3) \frac{\partial f(\epsilon)}{\partial \epsilon} d\epsilon. \quad (6)$$

Hence, up to rescaling, $G_3(\mu_3)$ is just the thermally broadened spectral function $A_d(\mu_3)$, which in effect is the two-lead dot DOS for the chemical potentials μ_1 and μ_2 . This means that the weakly coupled third lead acts as a conventional tunneling probe for the dot DOS, even though the dot itself is far from thermal equilibrium.

To test the above result, we have computed the differential conductance $G_3(\mu_3)$ by direct numerical differentiation of the current I_3 of Eq. (4) with respect to V_{31} , without resorting to Eqs. (5) and (6). Focusing for convenience on the limit $U \rightarrow \infty$ (double occupancy is forbidden on the dot), we evaluated the impurity Keldysh Green functions using the noncrossing approximation (NCA).¹⁰ The NCA is a self-consistent perturbation theory about the atomic limit, which is known to provide a good quantitative description of the temperature range $T \geq T_K$. It has been extensively used to study dilute magnetic alloys,¹⁰ and has been successfully applied to the out-of-equilibrium Kondo effect for both single-channel^{6,11,12} and two-channel scatterers.¹³ In the present context it has the crucial advantage that it can be easily generalized to a multilead setting. As a large- N theory, though, the NCA fails to recover Fermi-liquid behavior at low temperatures,¹⁰ and it overshoots the unitary limit for $T < T_K$ in the $N=2$, nondegenerate case. To avoid these NCA pathologies, we restrict attention hereafter to the range $T \geq T_K$.

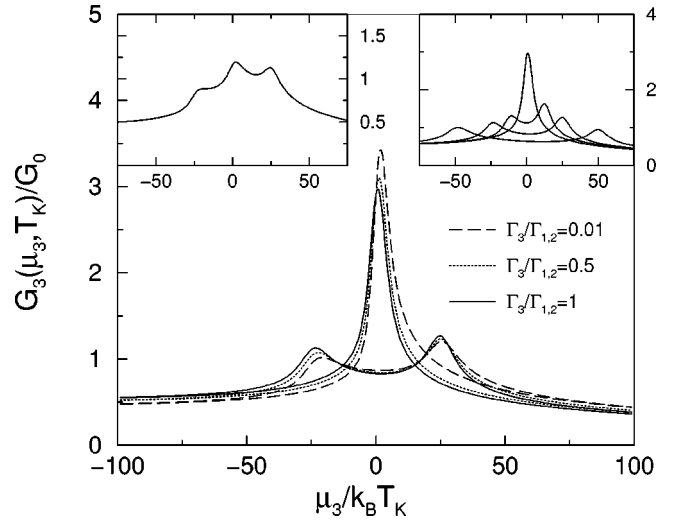


FIG. 4. The three-lead differential conductance $G_3(\mu_3)$, for $\Gamma_1 = \Gamma_2$ and different ratios of Γ_3 to Γ_1 . The source-drain voltage bias $eV_{sd}/k_B T_K$ is equal to zero for the three single-peak curves, and 50 for the three split-peak curves. $\Gamma/D = 0.067$ is kept fixed in all curves as to maintain the same Kondo temperature. Here $E_d/D = -0.278$, $T = T_K$, and $G_0 = D\hbar\Gamma/8e^2\Gamma_3(\Gamma_1 + \Gamma_2)$. The overall peak structure of $G_3(\mu_3)$ is mostly unchanged when Γ_3/Γ is increased. Right inset: The evolution of $G_3(\mu_3)$ with increasing source-drain voltage bias, for $\Gamma_1 = \Gamma_2 = \Gamma_3$ and $T = T_K$. Here $eV_{sd}/k_B T_K$ equals 0, 25, 50, and 100. Left inset: The dot DOS, $DA_d(\epsilon)$, for $eV_{sd}/k_B T_K = 50$, $\mu_3/k_B T_K = 0$, and $\Gamma_1 = \Gamma_2 = \Gamma_3$. There are three separate peaks in the dot DOS, one peak at the chemical potential of each of the three leads.

Figure 2 compares the the out-of-equilibrium dot DOS for a symmetrically coupled two-lead system with the differential conductance $G_3(\mu_3)$ in the proposed three-lead setting. Here and throughout the paper we set $(\mu_1 + \mu_2)/2$ as our reference energy, and use a semi-circular conduction-electron density of states with half-width D : $\nu_\alpha(\epsilon) = \sqrt{1 - (\epsilon/D)^2}$. The impurity model parameters are $E_d/D = -0.278$ and $\Gamma_1 = \Gamma_2 = D/30$, corresponding to a Kondo temperature of $k_B T_K/D = 2.5 \times 10^{-4}$.¹⁴ The third-lead coupling, $\Gamma_3/\Gamma_1 = 0.01$, is sufficiently weak as not to affect T_K .

Up to thermal broadening, the scaled differential conductance of Fig. 2 coincides with the out-of-equilibrium two-lead dot DOS, in excellent agreement with Eq. (6). For zero source-drain voltage bias $eV_{sd} = 0$, there is a single Kondo peak in $G_3(\mu_3)$, corresponding to the thermally broadened Abrikosov-Suhl resonance in $A_d(\epsilon)$. [Note that the height of the resonance in $A_d(\epsilon)$ is somewhat lower than $1/\pi\Gamma$, indicating that the Kondo effect is not yet fully developed.] As the source-drain bias is increased, a two-peak structure gradually develops in $G_3(\mu_3)$. Two well-resolved peaks are finally observed once eV_{sd} exceeds $k_B T_K$ and $k_B T$ by an order of magnitude or more. For $eV_{sd} \gg k_B T, k_B T_K$, the effect of thermal broadening is quite negligible, as the widths of the two Kondo peaks are governed by the dissipative lifetime induced by V_{sd} ,⁶ rather than by $k_B T$ or $k_B T_K$. The asymmetric Kondo line shapes, particularly for small to intermediate values of eV_{sd} , stem from the absence of particle-hole symmetry in the infinite- U Anderson model.

Figure 3 depicts the out-of-equilibrium two-lead dot DOS versus $G_3(\mu_3)$, for $eV_{sd}/k_B T_K = 50$, $T = T_K$, and different ratios of Γ_1 to Γ_2 . The couplings $(\Gamma_1 + \Gamma_2)$ and $\Gamma_3/(\Gamma_1 + \Gamma_2)$ are kept fixed as to maintain the same T_K . As before, excellent agreement is obtained between $G_3(\mu_3)$ and the two-lead dot DOS, in accordance with Eq. (6). The effect of an asymmetry in the couplings to the source and drain leads is to enhance the peak at the chemical potential of the more strongly coupled lead, at the expense of the peak at the chemical potential of the more weakly coupled lead. As seen in Fig. 3, the effect is quite dramatic. Only a shallow peak is left at the chemical potential of the more weakly coupled lead when the ratio between the two couplings is three. Accordingly, the dot DOS increasingly resembles that for sole coupling to just a single lead. The inequivalent line shapes for $\Gamma_1/\Gamma_2 = 3$ and $\Gamma_2/\Gamma_1 = 3$ are again the result of the absence of particle-hole symmetry in our model.

The effect of a temperature is to smear the peak structure of $G_3(\mu_3)$, as seen in the inset of Fig. 3. This smearing stems both from the thermally broadened Kondo peaks in the dot DOS, and from the convolution with the derivative of the Fermi-Dirac function in Eq. (6). The two-peak structure of the Kondo resonance remains visible, though, up to $k_B T$ about an order of magnitude smaller than eV_{sd} (see also Fig. 2). For large source-drain bias, as in Fig. 3, the two-peak structure thus extends up to temperatures well above T_K .

Thus far our discussion has been restricted to weak coupling between the dot and third lead. One may ask how does the qualitative picture change once this coupling is no longer

weak, when the reasoning leading to Eq. (6) no longer holds. In Fig. 4 we have plotted $G_3(\mu_3)$ for $\Gamma_1 = \Gamma_2$ and different ratios of Γ_3 to Γ_1 . Here Γ is kept fixed as to maintain the same T_K . Despite the notable changes in the dot DOS, $G_3(\mu_3)$ remains mostly unchanged even for Γ_3 as large as Γ_1 . Indeed, while $A_d(\epsilon)$ acquires a three-peak structure for $\Gamma_1 = \Gamma_2 = \Gamma_3$ (see Fig. 4, left inset), $G_3(\mu_3)$ remains closely related to the two-lead dot DOS for the chemical potentials μ_1 and μ_2 . This clearly shows the robustness of the proposed method for measuring the splitting of the Kondo resonance.

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Note added. After completion of this manuscript, we learned of a similar proposal by Sun and Guo¹⁵ to measure the out-of-equilibrium dot DOS using a multilead setting. While the basic idea and conclusions are the same, there are two main points that distinguish our work from that of Sun and Guo: (i) Sun and Guo utilize the equations-of-motion approach, which, unlike the NCA, is only qualitatively correct. (ii) We show in Fig. 4 that weak coupling to the third lead is not a prerequisite for measuring the out-of-equilibrium splitting of the Kondo resonance. Rather, the two-peak structure persists in $G_3(\mu_3)$ even when the coupling to the third lead is no longer weak.

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¹⁴ We define T_K as the temperature at which $R(T) = \{-\int_{-\infty}^{\infty} A_d^{-1}(\epsilon) [\partial f(\epsilon)/\partial \epsilon] d\epsilon\}^{-1}$ reduces to 50% of its extrapolated zero-temperature limit. This definition of T_K is common in dilute magnetic alloys, where $R(T)$ corresponds to the electrical resistivity due to scattering off the magnetic impurities. With T_K so defined, the half-width of the Abrikosov-Suhl resonance in the NCA impurity spectral function is about $1.7k_B T_K$.

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