Theory of the non-equilibrium quasiparticle distribution induced by Kondo defects

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It is shown that in resistive nanowires out of equilibrium containing either single- or two-channel Kondo impurities the distribution function \(f(E, U)\) obeys scaling behavior in terms of the quasiparticle energy \(E\) and the bias voltage \(U\). The numerically calculated \(f(E, U)\) curves explain quantitatively recent experiments on Cu and Au nanowires. The systematics of the impurity concentration \(c_{\text{imp}}\) extracted from the comparison between theory and results on various Cu and Au samples strongly suggests that in these systems the scaling arises from magnetic Kondo impurities.

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Electronic interactions in solids are usually investigated by means of linear response or by spectroscopic measurements which essentially probe the system in thermodynamic equilibrium. In an important series of experiments [1,2] the Saclay group demonstrated that unique information about the energy dependence of the interband density of states in a mesoscopic wire can be extracted from the measured \(\theta\) curves [1,2], with energy \(E\) and, hence, should be of local origin. Moreover, in the perturbative treatment it implies a logarithmic divergence of the energy relaxation rate at the Fermi energy \(E_F\). The latter has generated substantial interest because of the possible relation to the problem of dephasing saturation [4] in mesoscopic systems.

Anomalous low-energy behavior of local origin can be induced by the Fermi surface singularities characteristic for Kondo type systems [5]. Inelastic scattering by Kondo impurities was discussed in [6]. Based on these considerations, the single-channel Kondo (1CK) [5] and the two-channel Kondo (2CK) effect [5], possibly produced by degenerate dynamical defects [4], have been proposed as the origin of the anomalous energy relaxation. In this Letter we show that a very small concentration \(c_{\text{imp}}\) of either 1CK or 2CK impurities leads to the observed scaling behavior of \(f_x(E, U)\), when \(eU\) exceeds an intrinsic energy scale \(eU^*\) which is essentially equal to the Kondo temperature \(T_K\). The numerical results are in excellent quantitative agreement with the experimental curves [5], where \(c_{\text{imp}}\) is the only adjustable parameter of the theory. A detailed analysis suggests that the scaling behavior in Cu and Au wires is due to magnetic Kondo impurities.

Let us first set up the general formalism for calculating \(f_x(E, U)\) in a resistive nanowire of length \(L\), subject to the boundary conditions that the left \((x = 0)\) and the right \((x = L)\) leads are in equilibrium at their respective chemical potentials, i.e. \(f_{x=0}(E, U) = f^0(E)\), \(f_{x=L}(E, U) = f^0(E + eU)\), where \(f^0(E) = 1/(e^{E/T} + 1)\) is the Fermi distribution \((k_B = 1)\). The lesser \((\text{\textless})\) and the greater \((\text{\textgreater})\) conduction electron Keldysh Green’s function \(G_{\text{K}}^\pm(\vec{p}, E)\) and \(G_{\text{K}}^\pm(\vec{p}, E)\) respectively, where \(E\) and \(\vec{p}\) denote energy and momentum of the quasiparticles in a small volume centered around \(x\), in which the external fields may be considered constant. A superscript \(r\) indicates here and in the following a retarded propagator. In a disordered electron system with diffusion constant \(D\) the stationary quantum Boltzmann equation for the distribution as function of the qp energy \(E\) takes the diffusive form [4],

\[-DN^2f_x(E, U) = C\{f_x(E, U)\}, \tag{1}\]

The collision integral \(C\) is expressed in terms of the self-energies \(\Sigma\) for scattering into \((\text{\textless})\) and out of \((\text{\textgreater})\) states with given energy \(E\) \((N_o = \text{density of states per spin})\) as

\[C = \frac{1}{2\pi N_o} \int |\Sigma_x(E)|^2 E_x^r(\vec{p}, E) - \Sigma_x(E)|^2 E_x^< (\vec{p}, E)| \tag{2}\]

In the absence of any interactions \((C \equiv 0 \text{ in Eq. (1)})\) the distribution function has the double–step shape,

\[f_x(E, U) = \frac{E}{T}f^0(E + eU) + \left(1 - \frac{E}{T}\right)f^0(E). \tag{3}\]

For a small concentration of Kondo defects \(c_{\text{imp}}\), in addition to the static impurities, the conduction electron selfenergy is given in terms of the single-particle t–matrix
of the defect, \( t_\xi^E (E) \), as \( \Sigma_\xi^E = c_{imp} t_\xi^E \). \( t_\xi^E (E) \) mediates energy transfer between electrons in that it couples the dynamical defect both to the in or outgoing electron and to intermediate particle–hole pairs. The elastic scattering parts of \( t_\xi^E (E) \) cancel each other exactly in \( C \). We emphasize that, apart from the assumption of small \( c_{imp} \), the present formulation, Eqs. (1), (2), contains no approximations, once the \( t \)-matrix is known.

As pointed out in Ref. [1], the precise energy dependence of the electron-electron coupling is crucial for whether or not \( f_\xi (E, U) \) obeys a non-equilibrium scaling property, but has been notoriously difficult to calculate for the Kondo problem. While 4th order (unrenormalized) perturbation theory yields the correct power law for scaling [1], partial summations of logarithmic terms give corrections violating scaling [14]. However, such summations are valid only for \( T, E \gg T_K \) [3], Chapt. 3, while the non-equilibrium situation \( (eU > T_K, T \ll T_K) \) may involve all energies \( T \leq E \lesssim eU \).

Therefore, we use the slave boson formalism, where certain exact properties of the auxiliary particle propagators are known [11–13]. To describe 1CK as well as 2CK impurities we use the SU(N)×SU(M) Anderson impurity model in the Kondo limit, denoting the spin degeneracy by \( N \) and the number of identical, conserved conduction electron channels by \( M \). Following the notation of Ref. [13], the hamiltonian reads

\[
H = H_o + \varepsilon_d \sum_\sigma f_{\sigma}^\dagger f_\sigma + V \sum_{p,m,\sigma} (f_{p,m,\sigma}^\dagger c_{p,m,\sigma} + h.c.) ,
\]

subject to the operator constraint \( \hat{Q} = \sum_\sigma f_{\sigma}^\dagger f_\sigma + \sum_m b_{m}^\dagger b_m = 1 \). \( H_o = \sum_{p,m,\sigma} \varepsilon_{p,m,\sigma} c_{p,m,\sigma}^\dagger c_{p,m,\sigma} \) describes the conduction band. The auxiliary fermion and boson operators, \( f_{\sigma}^\dagger, b_m^\dagger, \) create the local defect in its quantum state \( \sigma \) or in the unoccupied state, respectively. In the 1CK case \( (N = 2, M = 1) \) of a magnetic Anderson impurity \( \sigma \) denotes spin, and \( m = 1 \) has no relevance. For a 2CK defect \( (N = 2, M = 2) \), \( \sigma \) is identified with a pseudospin, e.g. the parity of the local defect wave function, and \( m = 1, 2 \) is the conduction electron spin, acting as the channel degree of freedom. The equilibrium Kondo temperature of the model is \( T_K^{(0)} \approx E_F (N N_o J) (M/N) o^{-1/2} (N N_o J) \), with \( J = |V|^2 / |e_d| \) the effective spin exchange coupling. The bare auxiliary particle propagators read \( G^{(0)}_f (\omega) = 1/(\omega + i0) \) and \( G^{(0)}_b (\omega) = 1/(\omega + \varepsilon_d + i0) \). Here we have gauged the zero of the slave particle energy such that the pole of \( G^{(0)}_f \) is at \( \omega = 0 \).

The numerical evaluations of physical quantities will be done within the non–crossing approximation (NCA) which is shown diagrammatically in Fig. 1 a). The corresponding equations for the auxiliary fermion and boson selfenergies \( \Sigma^F, \Sigma^B \) in non-equilibrium read [14],

\[
\Sigma^F = \frac{G^F (\omega)}{|G^F (\omega)|^2} = - \frac{\Gamma}{N_o} \int \frac{d\varepsilon}{2\pi i} G^F (\varepsilon) G^F (\omega + \varepsilon) ,
\]

\[
\Sigma^B = \frac{G^B (\omega)}{|G^B (\omega)|^2} = + \frac{\Gamma}{N_o} \int \frac{d\varepsilon}{2\pi i} G^B (\varepsilon) G^B (\omega + \varepsilon) ,
\]

where \( \Gamma = \pi N_o |V|^2 \) is the effective hybridization, and \( G^F_b (\varepsilon) = \sum_\sigma G^F_x (\bar{p}, \varepsilon) \). This set of selfconsistent, non–linear equations is closed by the Kramers-Kroenig relations, \( G^F_x (\omega) = - \int d\varepsilon / (2\pi i) G^F (\omega + \varepsilon) \varepsilon / (\omega + \varepsilon + i0) \), which follow from causality and the fact that the auxiliary particle Green’s functions have only forward in time propagating parts. Within NCA the single–electron \( t \)-matrix due to the Kondo impurity is

\[
t^F_x = - \frac{\Gamma}{\pi N_o} \int \frac{d\varepsilon}{2\pi i} G^F_x (E + \varepsilon) G^F_b (\varepsilon) .
\]

By writing the renormalized auxiliary propagators as the bare ones with selfenergy insertions and using \( |V|^2 G^{r_{(b)} (\omega \approx 0)} (\omega) = J \), it is seen that the present formulation includes the Kondo induced electron-electron vertex of \( O(J^3) \) and \( O(J^4) \), with the points 3 and 4 connected (Fig. 1 c)). The exchange diagrams (points 3 and 2 connected) are not included. However, this does not change the scaling properties (see below). By self-consistency, NCA goes beyond this two-particle scattering approximation considered in Ref. [3].

The NCA in non-equilibrium includes both an inelastic spin relaxation rate and scaling behavior in terms of the applied bias. It is instructive to investigate these properties for a single Kondo impurity before we present the numerical solutions. The Kondo scale \( T_K \) is influenced by the step heights in \( f_{\xi} (E, U) \), Eq. (3), and the size of the corresponding logarithmic terms in the Kondo vertex, which is reduced compared to equilibrium. This leads to a suppression of \( T_K \), e.g. in the middle of the wire \((x/L = 1/2)\),

\[
T_K = \sqrt{(eU/2)^2 + T_K^{(0)} - eU/2} (x/L \gg 1/2) \approx T_K^{(0)} (eU/2) \frac{T_K^{(0)}}{eU} .
\]
At an arbitrary position \( x/L \), for \( eU \gg T_K^{(0)} \) we have 
\[ T_K = T_K^{(0)} \beta^{1/\eta/(eU)^{(1/\eta)}}, \] 
where \( \eta = \max[x/L, 1 - x/L] \). At the same time, there is an inelastic spin relaxation rate \( 1/\tau_s \), since in the non-equilibrium electron sea (Eq. (3)) there is finite phase space available for scattering even at \( T = 0 \). Technically, this relaxation rate appears as the imaginary part of the pseudofermion self-energy, \( \Sigma_f(\omega = 0) \), which carries the local spin degree of freedom. To leading order in \( J \) it is obtained by inserting the bare propagators \( G_{f,b}(0) \) in the diagram Fig. 1b, 
\[ \frac{1}{\tau_s} = 2\pi MN \frac{x}{L} (1 - x/L)(N_0 J)^2 eU. \] 
This is analogous to the well-known Korrinng spin relaxation rate [3], with \( T \) replaced by \( eU \). Solving Eqs. (3), 4 selfconsistently in the complete range of validity of NCA, \( T_K \lesssim eU \ll E_F \), we find that beyond perturbation theory \( 1/\tau_s \) depends on \( eU \) and \( T_K \) only, 
\[ \frac{1}{\tau_s} = \frac{x}{L} (1 - x/L) H_{M,N} (eU/T_K)eU, \] 
where the universal function \( H_{M,N}(y) = \pi M/[2N \ln^2(y)] \) for \( y \gg 1 \) [13], in accordance with Ref. [6]. Inserting \( 1/\tau_s \) into the pseudoparticle propagators, it cuts off all logarithmic contributions of perturbation theory. Thus, the low-\( T \) scale of the non-equilibrium Kondo system is \( T_K = \max[T_K(eU), 1/2\tau_s(eU)] \). The crossover from the Kondo (Eq. (3)) or \( T \) limited life time to the inelastic scale (Eq. (10)) occurs as function of \( eU \) at a bias \( eU^* \). It follows from the universality of \( 1/\tau_s \) that \( eU^* \) is only a function of \( T_K^{(0)} \), i.e., for dimensional reasons, \( eU^* = A M_N T_K^{(0)} \). Numerically we find \( A_{1,2} = 1.48 \pm 0.08 \) and \( A_{2,1} = 1.39 \pm 0.05 \). For \( eU > eU^* \) \( T_K \) has lost its relevance, and for \( eU \geq 10 eU^* \) one has with good accuracy \( 1/\tau_s \propto eU \), when \( eU \) is varied by a factor of ~4, wherein the \( eU \) dependence of the log terms is weak.

To investigate scaling of \( f_x(E, U) \) we must consider the energy dependence of the exact pseudoparticle Green’s functions, \( G_{f,b}(\omega) \), from which all other physical quantities are derived. It is known that in equilibrium at \( T = 0 \) it is determined by an infinite logarithmic series which results in power law behavior, 
\[ G_{f,b}(\omega) \propto i\Theta(\pm \omega)|\omega|^{-\alpha f, b}, \]
\[ G_{f,b}(\omega) \propto i\Theta(\pm \omega)|\omega|^{-\alpha f, b} \] 
for \( \omega \lesssim T_K^{(0)} \). The exponents \( \alpha_f, \alpha_b \) are due to an orthogonality catastrophe in the auxiliary propagators and have characteristic values \( \alpha_f = \alpha_b = 1/2 \) for the 1CK and \( \alpha_f = M/(M + N), \alpha_b = N/(M + N) \) for the 2CK fixed point of the model Eq. (1) (Kondo limit) [12,13]. We can exploit this knowledge to determine the frequency dependence of \( G_{f,b}(\omega) \) away from equilibrium without explicitly summing up the logarithmic series. At finite bias \( eU \gg T_K \) this series consists of similar terms as in equilibrium, however with three modifications: (i) Because of the inelastic relaxation rate \( 1/\tau_s = 2\gamma \) all frequency arguments are shifted, \( \omega = \omega + i\gamma \). (ii) \( G_f(\omega) \) has a singularity at \( \omega = 0 + i\gamma \), but there are two singularities in \( G_{b}^S(\omega) \) at \( \omega = 0 + ib\gamma \) and at \( \omega = eU + ib\gamma \), where \( b \) is a numerical factor. (iii) Each frequency integral involving \( G_{b}^S(\varepsilon + \omega) \), like, e.g., in Eq. (3), carries a prefactor \( M \), and each of the two singularities in \( G_{b}^S \) gives a singular contribution of equal weight at the external frequency \( \omega = 0 \). This can be seen as an effective doubling of \( M \). Points (i)–(iii) can be verified by iterating Eqs. (3), (4), starting from the bare propagators \( G_{f,b}^{(0)} \). As a result, we obtain at \( x/L = 1/2 \) damped power law behavior for the auxiliary propagators in non–equilibrium, 
\[ G_{f}^{(0)}(\omega) \propto \frac{1 - f_{1/2}(E, U)}{\omega + i\gamma} + \frac{1 - f_{1/2}(E, U)}{\omega - eU + ib\gamma} \] 
\[ G_{b}^{(0)}(\omega) \propto \frac{1 - f_{1/2}(E, U)}{\omega + ib\gamma} + \frac{1 - f_{1/2}(E, U)}{\omega - eU + ib\gamma}. \] 
The exact exponents [13] in the non-equilibrium situation (with \( M = 2M \) in the logarithmic series) are \( \alpha_f = 2M/(2M + N), \alpha_b = N/(2M + N) \). The \( \omega \) dependence Eq. (11) should extend from \( \omega = 0 \) up to the smallest energy scale of the model, i.e. for \( eU > eU^* \) up to \( \omega = eU \), since in this case the Kondo scale has disappeared. The behavior described above is confirmed by our numerical NCA solutions. For \( x/L \rightarrow 0 \) or \( x/L \rightarrow 1 \) the solution crosses over to the equilibrium one, as expected. The modification of the exponents \( \alpha_f, \alpha_b \) compared to their equilibrium values is reminiscent of a doubling of the channel number due to the two Fermi edges.

It remains to be seen whether a strong coupling region \( (T_K^{(0)} < eU < eU^*) \) can be realized where such behavior can be observed in the presence of \( 1/\tau_s \approx O(eU) \). The latter was neglected in Ref. [14]. Here we are interested in scaling at large bias \( (eU \gg eU^*) \). Inserting the power law forms Eq. (11) into Eqs. (3), (4), (7), dividing Eq. (3) by \( (eU)^{\alpha_f} \) and Eq. (4) by \( (eU)^{\alpha_b} \), and using the exact result \( \alpha_f' + \alpha_b' = 1 \), it is seen that the NCA equations contain only dimensionless energies, \( \varepsilon/eU \) etc. Power counting arguments [13] show that this is reproduced in arbitrary selfconsistent order in \( \Gamma \) beyond NCA. In the presence of a finite concentration \( \varepsilon_{\text{imp}} \), \( f_x(E, U) \) is determined by the selfconsistent coupled set of equations (4), (5) and (6), (7). It follows that the solution obeys scaling, \( f_x(E, U) = f_x(eU/eU^*) \) for \( eU > eU^* \). Our numerical solutions show scaling within a factor of 4 to 9 in \( eU \), depending on parameters, wherein log corrections to \( 1/\tau_s \propto eU \). Eq. (14), are small. Note that the power law behavior Eq. (13) and the fact that the low-energy cutoff \( 1/\tau_s \) itself is proportional to \( eU \) (up to small log corrections) cooperate to produce scaling. For \( eU \lesssim 10 eU^* \) we find deviations from scaling, because then the latter condition is no longer fulfilled. This provides for \( T \ll T_K^{(0)} \) a rough estimate, and for \( T > T_K^{(0)} \) an upper bound on \( T_K^{(0)} \); in the experiments [13] \( T \lesssim T_K^{(0)} \ll eU \).

For the numerical evaluations we assume magnetic
(1CK) impurities (2CK impurities give very similar results) and take $T_K^{(0)} \approx 0.1 \mathrm{K}$ in Cu and $T_K^{(0)} \approx 0.5 \mathrm{K}$ in Au wires (corresponding to $N_o J = 0.041$ and $N_o J = 0.048$, respectively), consistent with the above estimate and with independent estimates of $T_K^{(0)}$ for these samples. After $T_K^{(0)}$ is fixed, $c_{\text{imp}}$ is the only adjustable parameter of the theory. The results for $f_x(E, U)$, as measured by a tunnel junction attached to the wire, are shown in Fig. 2. Excellent quantitative agreement with experiments is obtained for all samples. In Au wires the fitted values of $c_{\text{imp}}$ are consistent with (although somewhat higher than) independent estimates of the magnetic impurity concentration, considering the roughness of both estimates. This suggests that the scaling behavior of $f_x(E, U)$ in the Au samples is due to magnetic (1CK) impurities. Furthermore, in all Cu samples the fitted $c_{\text{imp}}$ is $\sim 10^2$ times smaller than in Au. This systematics is in accordance with $c_{\text{imp}}$ estimated from the plateau in the $T$ dependence of the dephasing time $\tau_\varphi$ in similarly prepared samples.

In conclusion we have shown that single- or two-channel Kondo impurities in quantum nanowires induce scaling behavior of the non-equilibrium distribution function $f_x(E, U)$ at a bias $eU$ exceeding an energy scale $eU^* \approx T_K^{(0)}$. The results give a detailed explanation of related experiments. In the small bias or strong coupling regime ($T_K > eU$), 1CK and 2CK impurities must show qualitatively different behavior, as the former become potential scatterers with frozen spin dynamics, contrary to the latter with (ideally) non-zero entropy at $T = 0$. The quantitative comparison between the present theory and experiments suggests that in Au and at least partially in Cu nanowires both the scaling of $f_x(E, U)$ and the plateau in the low–$T$ dephasing time $\tau_\varphi$ are due to magnetic Kondo impurities. A unique test for magnetic impurities will be measuring $f_x(E, U)$ in a magnetic field.

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