Nonequilibrium 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 $c_{\rm imp}$ 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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While electronic interactions in solids are usually investigated by equilibrium probes like linear response or spectroscopic measurements, the shape of the nonequilibrium distribution function $f_x(E,U)$ of quasiparticles (qp) with energy E at a position x in a mesoscopic wire is sensitive to extremely weak interaction effects. This is so because $f_x(E,U)$ is not influenced by the large elastic scattering background, and has been demonstrated by the Saclay group in an important series of experiments [1,2], where controlled nonequilibrium was established by a finite transport voltage U. In resistive Cu and Au wires the theoretically expected double-step form of $f_x(E,U)$ [3] was found to be rounded such that it obeys the scaling property $f_x(E,U) = f_x(E/eU)$, when U *exceeds* a certain energy scale [1,2]. By a phenomenological analysis, the origin of the scaling was traced back to an anomalous electron-electron interaction $v(\omega)$ which scales with the energy transfer ω as $v(\omega) \propto 1/\omega$ [1]. It means that in 2nd order perturbation theory (PT) the resulting qp relaxation rate would not vanish at the Fermi energy E_F [1]. One can, therefore, conjecture that the anomalous scaling form $f_x(E/eU)$ and the apparent low-temperature saturation of the dephasing time observed [4] in the magnetoresistance of nanowires might have the same microscopic origin. To substantiate this speculation, a quantitative calculation in nonequilibrium is needed, and the perturbative infrared singularity of $v(\omega)$ signals that one has to go beyond finite order PT.

The scaling of the interaction $v(\omega) \propto 1/\omega$ implies, in particular, that it has no essential momentum dependence, i.e., should be of local origin. Anomalous low-energy behavior of local origin can be induced by the Fermi surface singularities characteristic for Kondo type systems [5,6]. Based on such considerations, the single-channel Kondo (1CK) [7] and the two-channel Kondo (2CK) effect [8], possibly produced by degenerate dynamical defects [6], have been proposed as the origin of the anomalous energy relaxation. Inelastic scattering by Kondo impurities was considered before in Ref. [9]. In this Letter we show that a very small concentration $c_{\rm imp}$ of Kondo impurities leads to the observed scaling behavior of $f_x(E,U)$, when eU exceeds an intrinsic energy scale eU_c which is essentially equal to the Kondo temperature T_K. In contrast to the treatment of Ref. [10], we include both vertex renormalization and finite lifetime effects in nonequilibrium. The numerical results are in excellent quantitative agreement with the experimental curves [1,2], with $c_{\rm imp}$ the only adjustable parameter of the theory. A detailed analysis suggests that the scaling behavior in Cu and Au wires is due to magnetic (1CK) impurities.

Model and formalism.—We consider $f_x(E,U)$ 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., $f_{x=0}(E,U) = f^0(E)$, $f_{x=L}(E,U) = f^0(E + eU)$, with $f^0(E) = 1/(e^{E/T} + 1)$ the Fermi distribution ($k_B = 1$). The lesser ($<$) and the greater ($>$) conduction electron Keldysh Green's functions read $G_x^<(\vec{p},E) = -2\pi i f_x(\vec{p}){\rm Im}G_x^r(\vec{p},E)$ and $G_x^>(\vec{p},E) = 2\pi i[1 - f_x(\vec{p})]{\rm Im}G_x^r(\vec{p},E)$, respectively, where E, $\vec{p}$ denote *qp* energy and momentum. A superscript r indicates a retarded propagator. In a disordered electron system with diffusion constant D the stationary quantum Boltzmann equation for the distribution as function of E takes the diffusive form [3]

$$-D\nabla_x^2 f_x(E,U) = C\{f_x(E,U)\}. \tag{1}$$

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

$$C = \frac{1}{2\pi N_o}\sum_p [\Sigma_x^<(E)G_x^>(\vec{p},E) - \Sigma_x^>(E)G_x^<(\vec{p},E)]. \tag{2}$$

In the absence of any interactions [$C \equiv 0$ in Eq. (1)] the distribution function has the double-step shape,

$$f_x(E,U) = \frac{x}{L}f^0(E + eU) + \left(1 - \frac{x}{L}\right)f^0(E). \tag{3}$$

For a small concentration of Kondo defects $c_{\rm imp}$, in addition to the static impurities, the conduction electron self-energy is given in terms of the single-particle t matrix of the defect, $t_x^{\gtrless}(E)$, as $\Sigma_x^{\gtrless} = c_{\rm imp} t_x^{\gtrless}$. The elastic scattering parts of $t_x^{\gtrless}(E)$ cancel each other exactly in C. We emphasize that, apart from the assumption of small $c_{\rm imp}$, the present formulation, Eqs. (1) and (2), contains no approximations, once the t matrix is known.

As pointed out in Ref. [1], the precise energy dependence of the kernel of the collision integral [and hence of $t_x^{\gtrless}(E)$] is crucial for whether or not $f_x(E,U)$ obeys a nonequilibrium scaling property, but has been notoriously difficult to calculate for the Kondo problem. Therefore we use the slave boson (SB) formalism, where certain exact properties of the auxiliary particle propagators are known [11–13]. The magnetic impurity is described by the Anderson model in the Kondo limit, i.e., by a low-lying local level ε_d with infinite on-site Coulomb repulsion, coupled to the conduction electron sea via a hybridization V,

$$H = H_o + \varepsilon_d \sum_\sigma f_\sigma^\dagger f_\sigma + V \sum_{p,\sigma} (f_\sigma^\dagger b c_{p\sigma} + \text{H.c.}). \tag{4}$$

$H_o = \sum_{\vec{p},\sigma} \varepsilon_p c_{\vec{p}\sigma}^\dagger c_{\vec{p}\sigma}$ describes the conduction band. The auxiliary fermion and boson operators, $f_\sigma^\dagger$, $b^\dagger$, create the impurity in its quantum state with spin $\sigma = \pm 1/2$ (spin degeneracy $N = 2$) or in the unoccupied state, respectively. Their dynamics are subject to the operator constraint $\hat{Q} = \sum_\sigma f_\sigma^\dagger f_\sigma + b^\dagger b \equiv 1$. The equilibrium Kondo temperature of the model is $T_K \simeq E_F \sqrt{(NN_oJ)}\, e^{-1/(NN_oJ)}$, with $J = |V|^2/|\varepsilon_d|$ the effective spin exchange coupling. The bare auxiliary particle propagators read $G_f^{r(0)}(\omega) = 1/(\omega + i0)$ and $G_b^{r(0)}(\omega) = 1/(\omega + \varepsilon_d + i0)$. Here we have gauged the zero of the slave particle energy such that the pole of $G_f^r(\omega)$ is at $\omega = 0$. The numerical evaluations will be done within the noncrossing approximation (NCA) which is shown diagrammatically in Fig. 1(a). The

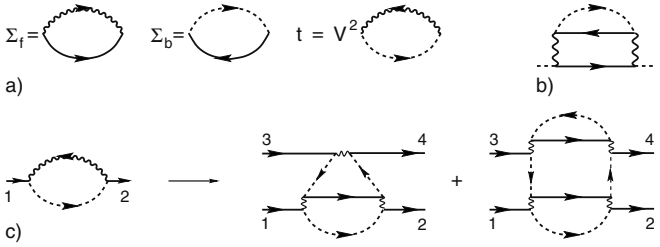

FIG. 1. (a) Diagrammatic representation of the NCA Eqs. (5)–(7). Solid, dashed, and wiggly lines denote the conduction electron, the renormalized local spin and auxiliary boson propagators, respectively. (b) Leading contribution to the inelastic spin relaxation rate $1/\tau_s$. (c) Expansion of the NCA single-particle t-matrix t into bare PT contributions (points 3 and 4 connected) to show that it includes the electron-electron vertex of $O(J^3)$ and $O(J^4)$.

corresponding equations for the auxiliary fermion and boson self-energies $\Sigma_f^{\gtrless}$, $\Sigma_b^{\gtrless}$ in nonequilibrium read [14]

$$\Sigma_f^{\gtrless} \equiv \frac{G_f^{\gtrless}(\omega)}{|G_f^r(\omega)|^2} = \mp \frac{\Gamma}{N_o} \int \frac{d\varepsilon}{2\pi i} G_x^{\gtrless}(-\varepsilon) G_b^{\gtrless}(\omega + \varepsilon), \tag{5}$$

$$\Sigma_b^{\gtrless} \equiv \frac{G_b^{\gtrless}(\omega)}{|G_b^r(\omega)|^2} = \pm \frac{N\Gamma}{N_o} \int \frac{d\varepsilon}{2\pi i} G_x^{\lessgtr}(\varepsilon) G_f^{\gtrless}(\omega + \varepsilon), \tag{6}$$

where $\Gamma = \pi N_o |V|^2$ is the effective hybridization, and $G_x^{\gtrless}(\varepsilon) = \sum_p G_x^{\gtrless}(\vec{p}, \varepsilon)$. This set of self-consistent, nonlinear equations is closed by the Kramers-Kroenig relations, $G_{f,b}^r(\omega) = -\int d\varepsilon/(2\pi i) G_{f,b}^<(\omega)/(\omega - \varepsilon + i0)$, 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

$$t_x^{\gtrless}(E) = \pm \frac{\Gamma}{\pi N_o} \int \frac{d\varepsilon}{2\pi i} G_f^{\gtrless}(E + \varepsilon) G_b^{\lessgtr}(\varepsilon). \tag{7}$$

Analytical analysis.— In order to understand how scaling of $f_x(E,U)$ in nonequilibrium arises, it is instructive to discuss the solutions of Eqs. (5)–(7) analytically for a single impurity. The PT for the Kondo model can be developed economically by identifying $|V|^2 G_b^{r(0)}(\omega \approx 0) = J$ within the SB representation, where Wick's theorem is preserved.

We first discuss the crossover bias eU_c above which the system becomes dominated by incoherent processes as well as the corresponding nonequilibrium spin relaxation rate $1/\tau_s$. As seen below, the energy dependence of $1/\tau_s$ is crucial for the scaling behavior of $f_x(E,U)$, while eU_c sets a lower bound for the voltage range in which scaling is obeyed. At finite bias the breakdown scale of PT, T^*, definied in analogy to the equilibrium T_K, is suppressed compared to T_K, e.g., for $x/L = 1/2$,

$$T^* = \sqrt{(eU/2)^2 + T_K^2} - eU/2 \overset{eU \gg T_K}{\simeq} \frac{T_K^2}{eU}. \tag{8}$$

At an arbitrary position x/L, for $eU \gg T_K$, we have $T^* = T_K^{1/\eta}/(eU)^{(1/\eta)-1}$, where $\eta = \max[x/L, 1 - x/L]$. The inelastic spin relaxation rate $1/\tau_s$ arises, because in the nonequilibrium 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, $\Sigma_f^r(\omega = 0)$, which carries the local spin degree of freedom. To leading order in J it is obtained by inserting the bare propagators $G_{f,b}^{\gtrless(0)}$ in the diagram Fig. 1(b),

$$\frac{1}{\tau_s} = 2\pi MN \frac{x}{L}\left(1 - \frac{x}{L}\right)(N_oJ)^2 eU. \tag{9}$$

This is analogous to the well-known Korringa spin relaxation rate [5], with T replaced by eU. However, the log terms appearing in higher order PT must be resummed, as is done in NCA through self-consistency. Solving Eqs. (5) and (6) in the complete range of validity of NCA,

$T_K \lesssim eU \ll E_F$, we find that due to the resummation $1/\tau_s$ depends on eU and T_K only,

$$\frac{1}{\tau_s} = \frac{x}{L}\left(1 - \frac{x}{L}\right) H_N\left(\frac{eU}{T_K}\right) eU\,, \tag{10}$$

where H_N is a universal function with $H_N(y) \to \pi/[2N\ln^2(y)]$ for $y \gg 1$ [15], in accordance with Ref. [7]. Inserting $1/\tau_s$ into the pseudoparticle propagators, it cuts off all logarithmic contributions of PT. Thus, the low-T scale of the nonequilibrium Kondo system is $T_o = \max[T^*(eU), 1/2\tau_s(eU)]$. The crossover from the Kondo [Eq. (8)] limited lifetime of the local spin to the inelastic time [Eq. (10)] occurs as a function of eU at a bias eU_c. It follows from the universality of $1/\tau_s$, Eq. (10), and of T^*, Eq. (8), that eU_c is only a function of T_K and, hence, for dimensional reasons, $eU_c = AT_K$. We find numerically that $A = 1.48 \pm 0.08$, i.e., the finite bias crossover scale is $eU_c \approx T_K$ itself, and nonequilibrium does not introduce a new energy scale. For $eU \gtrsim 10T_K$ one finds with good accuracy $1/\tau_s \propto eU$, when eU is varied by a factor of $\sim$4, wherein the eU dependence of the log terms in Eq. (10) is weak. To investigate scaling of $f_x(E, U)$ one must incorporate inelastic processes in the conduction electron t-matrix $t_x^{\gtrless}(E)$. To lowest order, these enter in $t_x^{\gtrless}(E)$ in $O(J^4)$ through the imaginary part of the $O(J^2)$ self-energy insertion for the pseudofermion (local spin) propagator [Fig. 1(c), third diagram, with points 3 and 4 connected]. This term is included in NCA, as seen by expanding the self-consistent NCA term in bare PT [Fig. 1(c)]. Its evaluation yields the $1/E^2$ energy dependence of the electron-electron vertex required for scaling [1], in agreement with Ref. [7]. However, in higher order PT lifetime corrections as well as vertex renormalizations occur. In order to capture these beyond finite-order PT, we must consider the exact energy dependence of the pseudoparticle Green's functions $G_{f,b}^{\gtrless}(\omega)$. It is known that in equilibrium at $T = 0$ it is determined by an exponential series of logarithms which results in power law behavior, $G_f^{\gtrless}(\omega) \propto \mp i\Theta(\pm\omega)|\omega|^{-\alpha_f}$, $G_b^{\gtrless}(\omega) \propto \mp i\Theta(\pm\omega)|\omega|^{-\alpha_b}$, for $\omega \lesssim T_K$. The exponents α_f, α_b are due to an orthogonality catastrophe in the auxiliary propagators and have characteristic values, with $\alpha_f = \alpha_b = 1/2$ in the Kondo limit [12,13]. We can exploit this knowledge to determine the frequency dependence of $G_{f,b}^{\gtrless}(\omega)$ away from equilibrium. 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, $\omega \to \omega + i\gamma$. (ii) $G_f(\omega)$ has a singularity at $\omega = 0 + i\gamma$, but there are two singularities in $G_b^{\gtrless}(\omega)$ at $\omega = 0 + ib\gamma$ and at $\omega = eU + ib\gamma$, where b is a numerical factor. (iii) In any frequency integral involving $G_b^{\gtrless}(\varepsilon + \omega)$, as, e.g., in Eq. (5), each of the two singularities in $G_b^{\gtrless}$ gives a singular contribution at the external frequency $\omega = 0$. Points (i)–(iii) can, e.g., be verified by iterating Eqs. (5) and (6), starting from the bare propagators $G_{f,b}^{\gtrless(0)}$. Hence, we obtain damped power law behavior,

$$\begin{aligned}
\frac{G_f^{>}(\omega)}{1 - f_x(\omega, U)} &\propto \frac{1}{i}\,\mathrm{Im}\,\frac{i}{(\omega + i\gamma)^{\alpha_f'}} \\
\frac{G_b^{>}(\omega)}{1 - f_x(\omega, U)} &\propto \frac{1}{i}\,\mathrm{Im}\left[\frac{i(1 - x/L)}{(\omega + ib\gamma)^{\alpha_b'}} + \frac{i(x/L)}{(\omega - eU + ib\gamma)^{\alpha_b'}}\right],
\end{aligned} \tag{11}$$

with log corrections whose size depends on x/L, and which vanish in the limit $eU \gg T_K$. The exponents

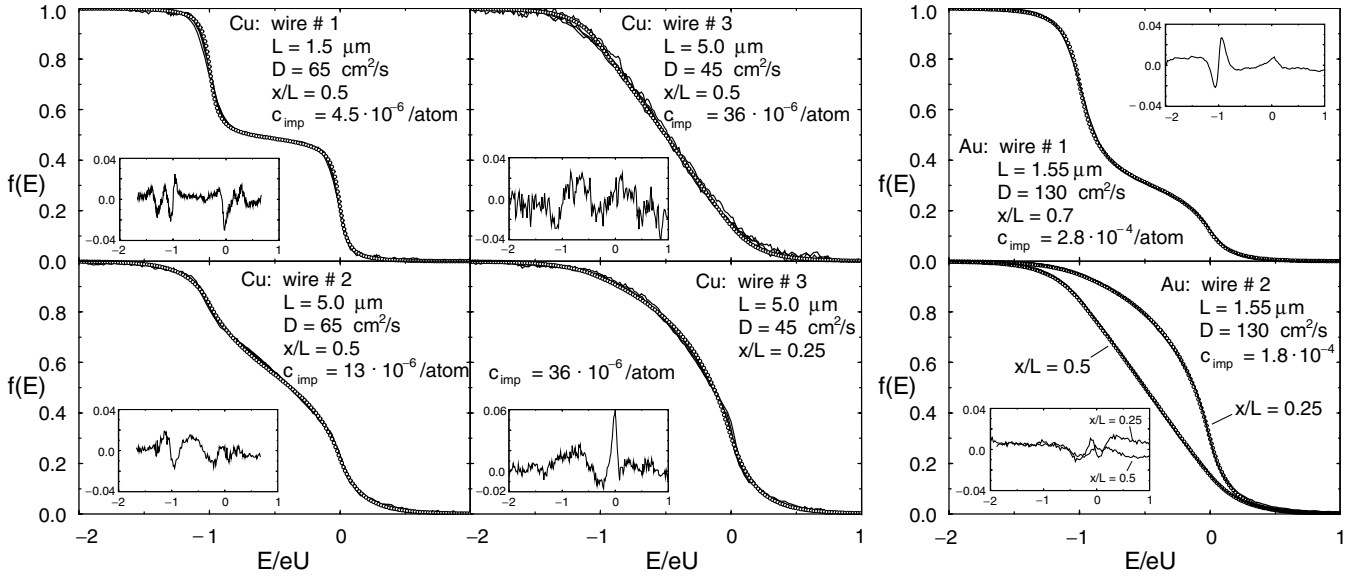

FIG. 2. Nonequilibrium distribution functions for various Cu and Au samples. Black lines: experimental results; Cu: [1], Au: [2]. Open circles: theory for $eU \gg T_K$. Deviations from scaling at smaller eU [1] are also reproduced by the theory (not shown). Fitted c_{imp} values are indicated. The insets show the difference between the experimental and the theoretical curves.

resulting from the NCA resummation [13] are in the nonequilibrium situation $\alpha_f' = 2/(2+N)$, $\alpha_b' = N/(2+N)$. Note that these values are reproduced in arbitrary (finite) order of self-consistent PT beyond NCA [13], and that for $eU > T_K$ a summation to infinite order of self-consistent PT cannot introduce new singularities because of the inelastic rate $1/\tau_s$. The ω dependence Eq. (11) extends from $\omega = 0$ up to the smallest energy scale of the model, i.e., for $eU > T_K$ up to $\omega = eU$, since in this case T_K is irrelevant. For $x/L \to 0$ or $x/L \to 1$ the solution crosses over to the equilibrium one. This behavior is confirmed by our numerical NCA solutions. We note in passing that the nonequilibrium exponents α_f', α_b' are reminiscent of the 2CK problem [6,13]. 2CK strong coupling behavior induced by finite bias has been suggested in Ref. [16]; see, however, [17]. In contrast, the NCA resummation, leading to α_f', α_b' above, is valid only in the region $eU \gtrsim T_K$ where strong coupling behavior is not realized because of the nonequilibrium spin decoherence rate $1/\tau_s \approx O(eU)$ [15]. We are interested in scaling at large bias ($eU \gg T_K$). Inserting the power law forms Eq. (11) into Eqs. (5)–(7), dividing Eq. (5) by $(eU)^{\alpha_f'}$ and Eq. (6) 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, ε/eU, etc. Power counting arguments [13] show that this is reproduced in arbitrary self-consistent order in Γ beyond NCA. In the presence of a finite concentration $c_{\rm imp}$, $f_x(E,U)$ is determined by the self-consistent coupled set of Eqs. (1), (2), and (5)–(7). It follows that the solution obeys scaling, $f_x(E,U) = f_x(E/eU)$, when the log corrections to Eqs. (11), as well as to (9), are small, i.e., for $eU \gg T_K$.

Comparison with experiment.—Our numerical solutions show scaling within a factor of 4 to 9 in eU, depending on parameters. Note that the power law behavior Eq. (11) and the fact that the low-energy cutoff $1/\tau_s$ itself is proportional to eU, i.e., both vertex renormalizations and self-energy corrections, cooperate to produce scaling. For $eU \lesssim 10T_K$ we find deviations from scaling. This provides for $T \ll T_K$ a rough estimate, and for $T > T_K$ an upper bound on T_K; in the experiments [1,2] $T \lesssim T_K \ll eU$. For the numerical evaluations we assume magnetic (1CK) impurities (for $eU \gtrsim T_K$, 2CK impurities give very similar results) and take $T_K \approx 0.1$ K in Cu and $T_K \approx 0.5$ K in Au wires, consistent with the above estimate and with independent estimates of T_K for these samples [2]. After T_K is fixed, $c_{\rm imp}$ 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 [1,2] is obtained for all samples. In Au wires the fitted values of $c_{\rm imp}$ are consistent with (although somewhat higher than) independent estimates of the magnetic impurity concentration [2], considering the roughness of both estimates. This suggets 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 $c_{\rm imp}$ is systematically about 10^2 times smaller than in Au. This systematics is in accordance with $c_{\rm imp}$ estimated from the plateau in the T dependence of the dephasing time τ_φ in similarly prepared samples [2,18].

Conclusion.—We have shown that single- or two-channel Kondo impurities in quantum nanowires induce scaling behavior of the nonequilibrium distribution function $f_x(E,U)$ at a bias eU exceeding an energy scale $eU_c \approx T_K$. The results give a detailed explanation of related experiments. In the small bias or strong coupling regime ($eU < T_K$), 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) nonzero 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 $f_x(E,U)$ [1,2] and the plateau in the low-T dephasing time τ_φ [18] are due to magnetic Kondo impurities. A unique test for magnetic impurities is measuring $f_x(E,U)$ in a magnetic field [19].

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