

## Nonequilibrium electron distribution in the presence of Kondo impurities

Georg Göppert and Hermann Grabert

Fakultät für Physik, Albert-Ludwigs-Universität, Hermann-Herder-Straße 3, D-79104 Freiburg, Germany

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We study the energy relaxation of quasiparticles in voltage biased mesoscopic wires in the presence of magnetic impurities. The renormalization of the exchange interaction of Kondo impurities coupled to conduction electrons is extended to the case of a nonequilibrium electron distribution, which is determined self-consistently from a Boltzmann equation with a collision term due to Kondo impurity mediated electron-electron scattering. The approach leads to predictions in quantitative agreement with recent experiments by Pothier *et al.* [Phys. Rev. Lett. **79**, 3490 (1997)].

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In 1997 the mesoscopic physics community was puzzled by two experimental findings. On the one hand Mohanty, Jariwala, and Webb<sup>1</sup> extracted the electron dephasing time from weak localization measurements of one-dimensional gold wires and found larger dephasing rates than predicted by the standard theory of Altshuler and Aronov (AA).<sup>2</sup> In the same year, Pothier *et al.*<sup>3</sup> published data on the nonequilibrium electron distribution in mesoscopic copper wires in the presence of an applied voltage. The electron-electron scattering rate gained from these data was shown to exceed predictions based on the AA theory. It was intuitively clear that these two observations are very likely due to a common origin, and in the last three years a large body of work has proposed all kinds of mechanisms ranging from interaction with two-level systems,<sup>4</sup> heating by radiation,<sup>5</sup> and  $1/f$  noise,<sup>6</sup> to intrinsic dephasing at zero temperature.<sup>7</sup> None of these predictions could give a quantitative description of the experiments, or they were ruled out by subsequent experimental studies. Finally, a scenario based on the two-channel Kondo effect of symmetrical two-level systems was recently proposed by Kroha.<sup>8</sup> For a rather complete discussion and analysis of available experimental data on both the weak localization and nonequilibrium electron distribution measurements in various metals we refer to the thesis by Pierre.<sup>9</sup>

Already Mohanty *et al.*<sup>1</sup> have demonstrated that iron impurities added to the gold wires can lead to similar effects to those observed in nominally pure wires. In fact, equating the pair breaking rate in superconductors containing magnetic impurities<sup>10</sup> with the magnetic contribution to the dephasing rate in normal metals,<sup>11</sup> one obtains a satisfactory explanation of the weak localization data near and above the Kondo temperature, where the theory is valid.<sup>9</sup> On the other hand, the effect of Kondo impurities on electron-electron scattering has been addressed only very recently. Kaminski and Glazman<sup>12</sup> (KG) have determined the two-particle  $t$  matrix mediated by magnetic impurities for an equilibrium system. The renormalization of the exchange coupling was studied with poor man's scaling using the applied voltage  $V$  as a low-energy cutoff. While the theory can explain the order of magnitude of the observed electron-electron scattering rate, it does not reproduce the correct voltage dependence but leads to more than an order of magnitude deviation from the data over the range of parameters investigated experimentally.<sup>13</sup> In this work we demonstrate that it is es-

sential to go beyond poor man's scaling since deviations from AA predictions are pronounced only below the Kondo temperature. Most importantly, the renormalization flow has to be determined in the presence of a *nonequilibrium* electron distribution calculated self-consistently from the Boltzmann equation. The collision term then depends on the distribution function not only through the occupation probabilities of in- and outgoing electrons but also via the renormalized interaction kernel.

We start by describing briefly the experimental situation. A mesoscopic wire of length  $L$  with diffusion constant  $D$  is attached to leads biased by a voltage  $V$ . (For details see Refs. 3 and 13.) The nonequilibrium electron distribution function  $f(\epsilon, x)$  in the presence of a steady state current can be determined from the Boltzmann equation for a diffusive mesoscopic wire,<sup>14</sup>

$$\frac{1}{\tau_D} \frac{\partial^2 f(\epsilon, x)}{\partial x^2} = I_{\text{coll}}, \quad (1)$$

with the boundary conditions  $f(\epsilon, 0) = f_F(\epsilon - eV/2)$  and  $f(\epsilon, 1) = f_F(\epsilon + eV/2)$  imposed by the leads. Here  $x$  is the position within the wire measured in units of  $L$ , and  $\tau_D = L^2/D$ . In the simple case of vanishing interaction  $I_{\text{coll}} = 0$  we obtain a double step function

$$f_0(\epsilon, x) = (1-x)f_F(\epsilon - eV/2) + xf_F(\epsilon + eV/2). \quad (2)$$

The interaction smears these steps, and in the limit of strong electron-electron interaction leads to a Fermi function with an effective temperature of the electrons.<sup>15</sup> The distribution function  $f(\epsilon, x)$  is determined in the experiment at various locations by tunneling spectroscopy.

Assuming a local interaction, the collision integral reads

$$\begin{aligned} I_{\text{coll}}(x, \epsilon, \{f\}) = & \int d\omega \int d\epsilon' K(\omega, \epsilon, \epsilon') \\ & \times \{f(\epsilon)f(\epsilon')[1-f(\epsilon-\omega)][1-f(\epsilon'+\omega)] \\ & - [1-f(\epsilon)][1-f(\epsilon')]f(\epsilon-\omega)f(\epsilon'+\omega)\} \end{aligned} \quad (3)$$

with an interaction kernel  $K(\omega, \epsilon, \epsilon')$ . Not to overload the notation, we suppress here and in the following the spatial

dependence of the kernel and the distribution function. To determine the kernel  $K(\omega, \epsilon, \epsilon')$ , we follow KG and start from the  $s$ - $d$  exchange Hamiltonian

$$H = H_0 + H_I, \quad (4)$$

where

$$H_0 = \sum_{k\sigma} \epsilon_k C_{k\sigma}^\dagger C_{k\sigma} \quad (5)$$

describes free quasiparticles with one-particle energies  $\epsilon_k$  and creation (annihilation) operators  $C_{k\sigma}^\dagger$  ( $C_{k\sigma}$ ) of states  $k\sigma$ . We assume that the density of impurities is small enough to treat the interaction with each impurity independently. Then, for a single impurity

$$H_I = J \sum_{kk'\sigma\sigma'} \mathbf{S} \cdot \mathbf{s}_{\sigma\sigma'} C_{k\sigma}^\dagger C_{k'\sigma'}, \quad (6)$$

where  $\mathbf{S}$  is the impurity spin operator and  $\mathbf{s}$  the vector of Pauli matrices. Further,  $J$  is the exchange interaction.

Let us first address the electron-electron interaction mediated by Kondo impurities in an equilibrium metal. We rewrite the most singular parts of the interaction in terms of single-particle  $t$  matrices and include renormalization effects for lower temperatures by an approach due to Zittartz and Müller-Hartmann<sup>16,17</sup> based on the Nagaoka equations.<sup>18</sup> This theory, though not able to describe the zero temperature limit correctly, leads to meaningful results at higher temperatures down to temperatures well below the Kondo temperature.<sup>19</sup>

For an effective two-particle interaction, the kernel  $K(\omega, \epsilon, \epsilon')$  is essentially given by the modulus squared of the on-shell two-particle  $t$  matrix. Further, one has to sum over all final electron spins  $\sigma_f, \sigma'_f$  and the initial spin  $\sigma'$  of the second electron and average over the initial spin  $\sigma$  of the first electron and the impurity spin  $S$ . For impurities with density  $C_{\text{imp}}$  the kernel then takes the form

$$K(\omega, \epsilon, \epsilon') = C_{\text{imp}} \rho^3 \frac{\pi}{2\hbar} \sum_{\sigma\sigma_f\sigma'_f\sigma'_S} \langle S | |T_{k\sigma, k'\sigma' \rightarrow k_f\sigma_f, k'_f\sigma'_f}|^2 | S \rangle, \quad (7)$$

where  $\rho$  is the electronic density of states at the Fermi level. The two-particle  $t$  matrix is defined by

$$T_{k\sigma, k'\sigma' \rightarrow k_f\sigma_f, k'_f\sigma'_f} = \langle k_f\sigma_f, k'_f\sigma'_f | T | k\sigma, k'\sigma' \rangle \quad (8)$$

and does not depend on the directions of the incoming and outgoing electrons. Hence, the wave numbers  $k, k'$  and  $k_f, k'_f$  are characterized by the energies  $\epsilon_k = \epsilon$ ,  $\epsilon_{k'} = \epsilon'$ , and  $\epsilon_{k_f} = \epsilon - \omega$ ,  $\epsilon_{k'_f} = \epsilon' + \omega$ , respectively (see Fig. 1). Unlike an effective potential mediated by the impurity spin,<sup>20</sup> the  $t$  matrix here is an operator in impurity spin space. As usual, the operator  $T$  is defined by the series

$$T(\epsilon) = H_I \sum_{n=0}^{\infty} \left( \frac{1}{\epsilon - H_0} H_I \right)^n. \quad (9)$$

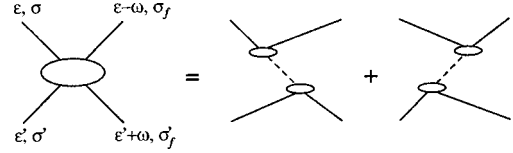


FIG. 1. Decomposition of the two-particle  $t$  matrix into two single-particle  $t$  matrices.

Our interest is in the retarded  $t$  matrix where the energy  $\epsilon$  is determined by the outgoing electrons, i.e.,  $\epsilon = \epsilon_{k_f} + \epsilon_{k'_f} + i\delta$ . Performing perturbation theory up to fifth order in the coupling  $J$ , we get for the inelastic processes

$$K(\omega, \epsilon, \epsilon') = \frac{1}{\omega^2} \frac{\pi}{2\hbar} \frac{C_{\text{imp}}}{\rho} S(S+1) (\rho J)^4 \times \{1 + \rho J [g(\epsilon) + g(\epsilon') + g^*(\epsilon - \omega) + g^*(\epsilon' + \omega)]\} + \text{l.s.} + O(\rho J)^6, \quad (10)$$

where l.s. means less singular terms in  $1/\omega$ . To leading order in  $J$ , this result agrees with the findings of KG. The less singular terms omitted arise from energy denominators that include an intermediate electronic energy to be summed over, leading at most to a logarithmic singularity for  $\omega \rightarrow 0$ . Further, the auxiliary function  $g(\epsilon)$  is given by

$$g(\epsilon) = \int d\epsilon' \left[ f(\epsilon') - \frac{1}{2} \right] \frac{\tilde{\rho}(\epsilon')}{\epsilon - \epsilon' + i\delta}, \quad (11)$$

where  $\tilde{\rho}(\epsilon)$  is the normalized density of states of the electrons with  $\tilde{\rho}(0) = 1$ .

To go beyond perturbation theory we consider the  $t$  matrix in operator form (9). We are interested in the most divergent terms for  $\omega \rightarrow 0$  and therefore search for energy denominators of the form  $1/(\epsilon - H_0) = \pm 1/\omega$ . Since  $\epsilon = \epsilon_{k_f} + \epsilon_{k'_f} = \epsilon_k - \omega + \epsilon_{k'} + \omega = \epsilon_k + \epsilon_{k'}$ , the intermediate energy must include both  $\epsilon_{k_f}$  and  $\epsilon_{k'}$  or  $\epsilon_{k'_f}$  and  $\epsilon_k$ , i.e., two unperturbed electron lines must lead to this intermediate state. Therefore, in leading order in  $1/\omega$  we may decompose the two-particle  $t$  matrix into two single-particle  $t$  matrices, one acting on the  $k$  quasiparticle and the other on the  $k'$  quasiparticle (see Fig. 1),

$$T_{k\sigma, k'\sigma' \rightarrow k_f\sigma_f, k'_f\sigma'_f} = T_{k\sigma \rightarrow k_f\sigma_f} \frac{1}{-\omega} T_{k'\sigma' \rightarrow k'_f\sigma'_f} + T_{k'\sigma' \rightarrow k'_f\sigma'_f} \frac{1}{\omega} T_{k\sigma \rightarrow k_f\sigma_f} + \text{l.s.} \quad (12)$$

Here, the operator character of the  $t$  matrices in impurity spin space plays an essential role since all commuting terms cancel.

To proceed we follow Zittartz<sup>17</sup> and decompose the single-particle  $t$  matrix into a non-spin-flip and a spin-flip amplitude,

$$T_{k\sigma \rightarrow k_f \sigma_f} = t_{kk_f} \delta_{\sigma\sigma_f} I_S + \tau_{kk_f} \mathbf{S} \cdot \mathbf{s}_{\sigma\sigma_f}. \quad (13)$$

Here,  $I_S$  means the identity in impurity spin space, and  $t_{kk_f}$  and  $\tau_{kk_f}$  are the  $t$  matrices in the non-spin-flip and spin-flip channels, respectively. Since the identity commutes with other spin matrices, it does not lead to a leading order contribution in  $1/\omega$ . Inserting the two-particle  $t$  matrix (12) with (13) into Eq. (7), we get

$$K(\omega, \epsilon, \epsilon') = \frac{1}{\omega^2} \frac{\pi}{2\hbar} C_{\text{imp}} S(S+1) \rho^3 |\tau(\epsilon)\tau(\epsilon')|^2 + 1.s. \quad (14)$$

Comparing this expression with the perturbative result (10), we see that the bare coupling  $J$  is replaced by the renormalized quantity  $\tau_{kk_f} = \tau(\epsilon_{k_f})$ . This result (14) is of central importance for the analysis below. Note that the expression (14) displays an explicit  $1/\omega^2$  dependence which is crucial for the experimentally observed scaling behavior.<sup>3</sup> The kernel for the effective electron-electron scattering depends on the off-shell spin-flip part of the  $t$  matrix, which has a different behavior from the on-shell non-spin-flip part of the  $t$  matrix responsible for the temperature dependence of the resistance. In our approach the  $1/\omega^2$  algebraic factor emerges analytically from this representation of  $K(\omega, \epsilon, \epsilon')$ .<sup>22</sup>

Zittartz<sup>17</sup> has shown that the Nagaoka equations lead to a single-particle  $t$  matrix in the spin-flip channel of the form

$$\tau(\epsilon) = \frac{J}{\phi(\epsilon)}. \quad (15)$$

With Hamann's solution<sup>21</sup> of the Nagaoka equations, the denominator reads

$$\phi(\epsilon) = [X(\epsilon)^2 + S(S+1)(\pi\rho J)^2]^{1/2}. \quad (16)$$

Here

$$X(\epsilon) = 1 - S(S+1)(\pi\rho J)^2/4 - \rho J R(\epsilon) \quad (17)$$

is temperature dependent via the function

$$R(\epsilon) = -\ln\left(\frac{\omega + iT}{i\Lambda}\right), \quad (18)$$

where  $\Lambda$  is the electronic bandwidth.  $R(\epsilon)$  is an equilibrium approximation of the auxiliary function  $g(\epsilon)$  introduced in Eq. (11). By inserting the solution (15)–(18) into the kernel (14) and expanding in  $J$ , we can indeed recover the perturbative result (10).

Let us now turn to a nonequilibrium situation. Since in equilibrium and nonequilibrium the  $t$ -matrix expansions differ only in the occupation probabilities of intermediate states, we can use the result for the equilibrium  $t$  matrix, replacing the distribution of intermediate states by its (unknown) nonequilibrium form. In the solution (15)–(17) occupation probabilities affect only the function  $R(\epsilon)$ , which in a nonequilibrium system is no longer of the form (18) but has to be replaced by  $g(\epsilon)$ . Of course, then the nonequilibrium single-particle  $t$  matrix in the spin-flip channel depends via  $g(\epsilon)$  on the nonequilibrium distribution function  $f(\epsilon)$ . The resulting

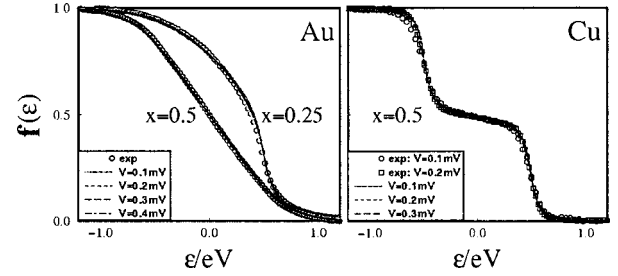


FIG. 2. Nonequilibrium distribution function for various voltages  $V$  and positions  $x$  compared with experimental data. Due to the scaling behavior of  $f(\epsilon, V)$ , the results fall on a single line at each position  $x$ .

form of the collision kernel is fully consistent with perturbation theory up to fifth order in  $J$  and it includes correctly the leading logarithmic terms.

To proceed, the  $t$  matrix and the distribution function need to be determined self-consistently. Both quantities depend on the position  $x$  within the wire. We start with the initial nonequilibrium distribution function (2), and determine the  $t$  matrix from Eqs. (15)–(17) and the collision kernel from Eq. (14). An improved distribution function is then obtained from the Boltzmann equation (1), which gives rise to an improved kernel. This procedure converges after about 20 iterations.

First, we compared our results with recent experimental data<sup>13</sup> on the energy relaxation in Au wires in the presence of Fe impurities. In these experiments all parameters were determined by independent measurements. For instance, for sample 2 measured at  $T=33$  mK the diffusion time  $\tau_D = 1.8$  ns. The impurity density was determined from the temperature dependence of the resistance as  $C_{\text{imp}} \approx 55$  ppm. We set the impurity spin to  $S=1/2$  and the Kondo temperature to  $T_K=1$  K which is a typical value for Fe in Au.<sup>19</sup> Further, we used a density of states of  $\rho=0.25/\text{site eV}$ .<sup>23</sup>

Pothier *et al.*<sup>3</sup> has noticed that the distribution functions  $f(\epsilon, V)$  exhibit scaling behavior so that as a function of  $\epsilon/eV$  all data at a given position  $x$  fall on a single line. In Fig. 2 we show our findings for the distribution function using the parameters of Pierre *et al.*<sup>13</sup> for various voltages  $V = 0.1$ – $0.4$  mV and two positions  $x=0.25, 0.5$ . They are compared with the experimental data points<sup>13</sup> and we find excellent agreement without adjustable parameters. We emphasize that the result is independent of the choice of the bandwidth and is insensitive to the Kondo temperature as long as  $T \ll T_K$ . For other values of  $x$  the curves coincide likewise.

On the other hand, for higher temperatures  $T > T_K$  where poor man's scaling holds, the kernel varies logarithmically with the applied voltage  $V$ ,<sup>12</sup> and the distribution function cannot be written as a function of  $\epsilon/eV$  only. We have also analyzed the data by Pothier *et al.*<sup>3</sup> on Cu wires and find again good agreement assuming a density  $C_{\text{imp}}=14$  ppm of  $S=1/2$  impurities with a Kondo temperature above 200 mK (see Fig. 2). Small deviations between the theoretical and experimental results are likely due to heating in this first experiment.<sup>24</sup>

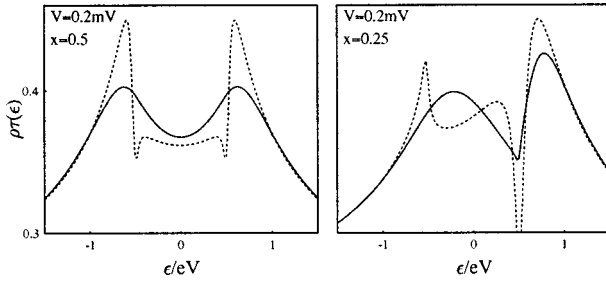


FIG. 3. Real part of the  $t$  matrix in the spin-flip channel. The solid line shows the self-consistent solution while the dotted line gives the result using the initial distribution function  $f_0$ .

To see the effect of the nonequilibrium electronic distribution on the renormalization of the exchange interaction, we show in Fig. 3 for sample 2 of Pierre *et al.*<sup>13</sup> the real part of the single-particle  $t$  matrix in the spin-flip channel  $\rho\tau(E)$  for  $V=0.2$  mV at  $x=0.5$  (left panel) and  $x=0.25$  (right panel). The solid line gives the self-consistent solution obtained from the iteration explained above, while the dotted line depicts the result obtained for the distribution (2) in the absence of energy relaxation. We see that the coupling changes significantly with the distribution function. Using only the initial  $t$  matrix does not suffice to explain the experimental data. Comparing  $t$  matrices for various voltages,

we find weak dependence between the Fermi points  $\epsilon = \pm eV/2$ . This gives rise to the scaling behavior of the distribution function, while poor man's scaling<sup>12</sup> implies a significant voltage dependence of the scaled data, in conflict with experiments.<sup>13</sup>

To complete our discussion we should have included spin dynamics which cuts off the  $1/\omega^2$  divergence of  $K(\omega, \epsilon, \epsilon')$  for small frequencies.<sup>12</sup> This is crucial for effects that depend strongly on the low-frequency limit, such as dephasing. However, the distribution function  $f(\epsilon)$  determined from the Boltzmann equation (1) is almost insensitive to this low-frequency cutoff since the collision integral remains finite even in the absence of a cutoff. Hence, for the problem considered here, one can disregard the impurity spin relaxation.

In summary we have determined the effective electron-electron collision kernel mediated by magnetic impurities for nonequilibrium wires at temperatures below the Kondo temperature. We found excellent agreement with recent experimental findings.<sup>3,13</sup> In particular, we have demonstrated that the distribution function displays scaling only in the regime below the Kondo temperature.

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