Energy and phase relaxation in non-equilibrium diffusive nano-wires with two-level systems

Johann Kroha

Institut für Theorie der Kondensierten Materie
Universität Karlsruhe, Postfach 6980, D-76128 Karlsruhe, Germany

Summary: In recent experiments the non-equilibrium distribution function $f(E,U)$ in diffusive Cu and Au quantum wires at a transport voltage $U$ shows scaling behavior, $f(E,U) = f(E/eU)$, indicating a non-Fermi liquid interaction with non-vanishing $T=0$ scattering rate. The two-channel Kondo (2CK) effect, possibly produced by degenerate two-level systems, is known to exhibit such behavior. Generalizing the auxiliary boson method to non-equilibrium, we calculate $f(E,U)$ in the presence of 2CK impurities. We show that the 2CK equations reproduce the scaling form $f(E/eU)$. For all measured samples the theoretical, scaled distribution functions coincide quantitatively with the experimental results, the impurity concentration being the only adjustable parameter. This provides a microscopic explanation for the experiments and, considering that no other mechanism producing the scaling form is known to date, lends strong evidence for the presence of degenerate two-level defects in these systems. The relevance of these results for the problem of dephasing in mesoscopic wires is discussed.

1 Introduction

Two-level systems (TLS) have been known to exist in disordered solids for a long time. Their signatures have been observed in the anomalous thermodynamic properties of glasses, which may be explained by a flat distribution of level splittings. In metals, slow two-level fluctuators are evidenced by telegraph noise in the conductance [1]. A new physical phenomenon, the two-channel Kondo (2CK) effect [2, 3], arises when a fast, energetically (nearly) degenerate TLS is embedded in a metal, where the local impurity degree of freedom couples to the continuum of conduction electrons via an exchange interaction. The channel degree of freedom, conserved by this interaction, is the magnetic conduction electron spin, which is always degenerate in the presence of time reversal symmetry.
The 2CK effect exhibits striking non-Fermi liquid behavior with a non-vanishing single-particle scattering rate at the Fermi energy $\varepsilon_F$ for temperature $T = 0$, logarithmic behavior of the linear specific heat coefficient and the susceptibility, and a non-analytic, universal correction to the electronic density of states, $\Delta N(E)/(B\sqrt{T}) = h(x)$, where $B$ is a non-universal constant, $x := |E - \varepsilon_F|/T$ and $h(x)$ is a universal scaling function with $h(x) = \sqrt{x}$ for $x \gg 1$. Signatures of the latter have been observed as zero bias conductance anomalies (ZBA) of nano point contacts. As suggested in Ref. [12], the finite single-particle scattering rate at the Fermi energy would provide a natural explanation for the saturation of the dephasing time $\tau_\phi$ at low temperatures, which has recently been observed in magnetotransport measurements of weak localization in disordered wires.

However, the actual existence of the 2CK effect in nature has remained a controversial issue, partly because the ZBA could also be explained qualitatively, by the Al'tshuler-Aronov diffusion-enhanced Coulomb interaction, partly because the physical realization of degenerate TLS is poorly understood. Therefore, it is essential to develop unambiguous methods for detecting 2CK physics in mesoscopic systems.

Recently, it has been demonstrated in a landmark experiment performed by the Saclay “Quantronics” group that unique information about the electronic interactions in a metal may be extracted from the distribution function $f(E, U)$ of quasiparticles with energy $E$ when the system is driven far away from equilibrium by a transport voltage $U$: The shape of $f(E, U)$ is determined by the energy dependence of relaxation processes which tend to equilibrate the system. It has been found that in diffusive Cu and Au nano-wires the distribution function obeys a scaling form, $f(E, U) = f(E/eU)$, implying a non-Fermi liquid interaction.

In this article we discuss the close relation between this peculiar scaling property, which we will call “$E/eU$ scaling” for brevity, and non-Fermi liquid behavior. It is then shown that the 2CK effect obeys $E/eU$ scaling and reproduces quantitatively the measured distribution functions, while any other type of interaction is ruled out. This provides the strongest case to date for the physical realization of the 2CK effect in Cu and Au nano-wires induced by TLS. We briefly discuss its relation to the problem of the dephasing time saturation at low temperatures.

2 Experiment and non-Fermi liquid signature

The distribution function $f(E, U)$ was measured in an experimental setup where a non-equilibrium current was driven through a Cu nano-wire contacted by two reservoirs at chemical potentials 0 and $eU$, respectively (Fig. 1). In addition,
a superconducting Al tunneling junction was attached at a position \( x \) along the wire, the Al slab being in equilibrium with itself. For a voltage \( V \) across the junction the tunneling current is given by

\[
j_{\text{tunnel}} = \frac{e}{\hbar} |t|^2 \sum_\sigma \int dE \left[ f(E) - f^o(E + eV) \right] N_{Cu,\sigma}(E) N_{sc}(E + eV) ,
\]

(2.1)

where \( t \) is the (energy independent) tunneling matrix element, \( f^o(E) \) is the Fermi distribution function in the superconductor, and \( N_{Cu}, N_{sc} \) denote the density of states in the wire and in the superconductor, respectively. Since for the voltages used in the experiment \( N_{Cu} \) is flat and the BCS density of states \( N_{sc} \) is measured independently, the non-equilibrium distribution \( f(E) \) in the Cu wire can be extracted from this expression. The electronic transport in the wire is diffusive with diffusion coefficient \( D \). Length \( L \) and thickness \( d \) of the wire are such that the Fermi surface is three dimensional and the Coulomb and phonon scattering times are large compared to the electronic diffusion time \( \tau_D = L^2 / D \) through the wire, so that equilibration due to these processes can be neglected [18]. In this situation, assuming purely elastic scattering, one expects the distribution function at a given position \( x \) along the wire to be a linear superposition of the Fermi functions in the reservoirs,

\[
f_{elast}^x(E, U) = (1 - \frac{x}{L}) f^o(E + eU) + \frac{x}{L} f^o(E) ,
\]

(2.2)

since there is no energy exchange within the electron system or between the electron system and the lattice. Eq. (2.2) is a solution of the diffusive Boltzmann equation (3.6) [21], when the collision integral vanishes (see section 3). This situation is to be distinguished from the hot electron regime, where local equilibration occurs due to inelastic processes [22].

The measured distribution functions showed rounding of the Fermi steps as compared to Eq. (2.2) and obeyed scale invariance with respect to the transport voltage \( U \), \( f(E, U) = f(E/eU) \), when \( U \) exceeded a certain low energy scale, \( eU \gtrsim 0.1meV \) [18]. Deviations from scaling were observed again for voltages larger than a high energy scale \( E_o \approx 0.5meV \). The latter may be explained by reservoir heating effects or by the electrons coupling to additional degrees of freedom at high energies. We now deduce the non-Fermi liquid signature from the scaling property. The latter implies that the equation of motion for \( f(E) \), the Boltzmann equation and, as a consequence, the inelastic single-particle collision rate \( 1/\tau(E) \) are scale invariant. Assuming a (yet to be determined) two-particle potential \( \tilde{V}(\varepsilon) \) with energy transfer \( \varepsilon \), \( 1/\tau \) is given in 2nd order perturbation theory as [23]

\[
\frac{1}{\tau(E)} = \frac{1}{\tau(E/eU)} \approx N_{Cu}(0)^3 \int d\varepsilon \int d\varepsilon' |\tilde{V}(\varepsilon)|^2 \tilde{F} \left( \frac{E}{eU}, \frac{\varepsilon}{eU}, \frac{\varepsilon'}{eU} \right) .
\]

(2.3)
Here $\tilde{F}$ is a combination of distribution functions $f$ guaranteeing that there is only scattering from an occupied into an unoccupied state. Therefore, the experimental results about the scaling property of $f$ imply that $\tilde{F}$ depends only on the dimensionless energies as displayed in Eq. (2.3). Demanding scale invariance for $1/\tau$ with respect to $eU$, i.e. making the frequency integrals dimensionless, implies a characteristic energy dependence of the interaction and the single particle scattering rate, $\tilde{V}(\varepsilon) \propto 1/\varepsilon$ and $1/\tau(E) \propto -\ln(E/E_0)$, for energies less than $E_0$. These infrared singularities indicate a breakdown of Fermi liquid theory within the 2nd order perturbation theory argument applied here.

3 Two-channel Kondo effect and scaling in non-equilibrium

The strong infrared divergence of the two-particle potential, $\tilde{V}(\varepsilon) \propto 1/\varepsilon$, deduced above, is not explained by any conventional interaction, including the Al'tshuler-Aronov interaction [17]. It must, therefore, be generated by an infinite resummation of logarithmic terms obtained in perturbation theory due to the presence of a Fermi edge. In this section the 2CK effect is briefly reviewed. We then show that the effective electron-electron vertex, mediated by a 2CK impurity, has a $1/\varepsilon$ divergence and calculate the resulting distribution functions away from equilibrium.

The 2CK effect arises whenever a local, energetically degenerate two-level degree of freedom (pseudospin $\tau = \pm 1/2$) is coupled to a system of two identical conduction electron bands or channels via a pseudospin exchange interaction, which, however, conserves the channel degree of freedom. As for the physical realization of the pseudospin, it has been suggested [8] that it might arise from...
Figure 2 Schematic snapshot of the Kondo screening cloud. The bold arrows represent the impurity (pseudo)spin. a) Single-channel Kondo effect. Conduction electrons and local impurity spin form a collective singlet ground state with entropy $S(T = 0) = 0$. b) Two-channel Kondo effect. Each of the two identical conduction electron bands form a screening cloud (black and grey), so that the net pseudospin of the combined clouds is not 0; pseudospin singlet formation is frustrated. The ground state is not unique, resulting in a non-vanishing zero-point entropy $S(T = 0) = k_B \ln \sqrt{2}$.

the degenerate positions of an atom in a symmetrical double well potential, from a rotational degree of freedom of a lattice defect or group of atoms, or from sliding kinks on screw dislocations in a lattice [24]. The channel degree of freedom is then the magnetic electron spin $\sigma = \pm 1/2$, which is necessarily degenerate because of time reversal symmetry (Kramers degeneracy).

In order to describe non-equilibrium situations, it is useful to represent this system in terms of the low-energy physics of a SU(2)$_{\text{pseudospin}} \times$SU(2)$_{\text{channel}}$ Anderson impurity model in the Kondo limit, i.e. in terms of a doubly degenerate local level $E_d < 0$, coupled via a hybridization $v$ to two identical conduction channels, with conduction electron operators $c_{k\tau\sigma}, c_{k\tau\sigma}^\dagger$. The dynamics of the two-level degree of freedom (pseudospin) is described by a fermionic field operator $d_{\tau\sigma}$, with the restriction that the local level must not be doubly occupied at any time. The latter is implemented exactly by decomposing the local operator into auxiliary fermion $f_{\tau}^\dagger$ and boson $b_{\bar{\sigma}}$ operators, $d_{\tau\sigma}^\dagger = f_{\tau}^\dagger b_{\bar{\sigma}}$, supplemented by the operator constraint $\hat{Q} = \sum_{\tau} f_{\tau}^\dagger f_{\tau} + \sum_{\bar{\sigma}} b_{\bar{\sigma}}^\dagger b_{\bar{\sigma}} = 1$ [25]. Thus, we describe the 2CK system by the Anderson Hamiltonian in auxiliary particle representation,

$$H = \sum_{k\sigma\tau} \varepsilon_k c_{k\tau\sigma}^\dagger c_{k\tau\sigma} + E_d \sum_{\tau} f_{\tau}^\dagger f_{\tau} + v \sum_{k\sigma\tau} (f_{\tau}^\dagger b_{\bar{\sigma}} c_{k\tau\sigma} + h.c.) . \quad (3.4)$$

Note that the boson field $b_{\bar{\sigma}}$ transforms with respect to the adjoint representation of SU(2)$_{\text{channel}}$ (denoted by the index $\bar{\sigma}$). By integrating out $b_{\bar{\sigma}}$ in the Kondo regime ($\sum_{\tau} f_{\tau}^\dagger f_{\tau} \approx 1, \ |E| \ll |E_d|)$, $H$ is reduced to a 2CK model where the exchange coupling $J = |v|^2 / |E_d|$ is mediated by the boson propagator and, as
required, conserves the channel degree of freedom $\sigma$. The above auxiliary particle representation is known to describe the infrared behavior of the 2CK effect correctly \[26\] already on the level of a leading-order selfconsistent perturbation theory in the hybridization $v$, the non-crossing approximation (NCA) \[27\]. It also allows for the application of standard Green’s function methods and, in particular, is readily generalized \[8\] to non-equilibrium by means of the Keldysh technique.

Physically, the presence of two identical conduction electron channels leads to an overscreening of the impurity pseudospin below the Kondo temperature $T_K = \frac{2\Gamma}{\pi} \exp\left(-\pi|E_d|/2\Gamma\right)$, $\Gamma = \pi|v|^2 N C_{\mu}(0)$, as elucidated in Fig. 2. This results in a non-vanishing zero-point entropy $S(T=0) = k_B \ln\sqrt{2}$ \[4\] and is closely related to the finite single-particle decay rate $1/\tau(E=0,T=0) > 0$ predicted by the 2CK effect.

Below the Kondo scale $T_K$, the auxiliary particle propagators $G_f, G_b$ are known to exhibit infrared power-law behavior in equilibrium, $G_f(\omega) \propto \omega^{-\alpha_f}$, $G_b(\omega) \propto \omega^{-\alpha_b}$ for \(\omega \ll T_K\) \[28\], where the exact \[26\] relation $\alpha_f + \alpha_b = 1$ is characteristic for the multichannel Kondo effect ($\alpha_f = M/(M+N)$, $\alpha_b = N/(M+N)$ for pseudospin degeneracy $N$ and channel number $M$). Using power counting arguments, it follows that each fermion (boson) propagator contributes a power $-\alpha_f$ ($-\alpha_b$) and each frequency integral contributes a power 1 to the frequency dependence of the effective electron-electron vertex $\gamma(\omega)$, shown in Fig. 3. Thus, the 2CK electron-electron vertex depends on the frequency transfer $\omega$ as $\gamma(\omega) \propto \omega^{1-2(\alpha_f+\alpha_b)} = \omega^{-1}$ \[29\]. On the perturbative level, this implies scale invariance of the inelastic scattering rate $1/\tau(E)$ with respect to the transport voltage $U$, as discussed in section 2. A self-consistent resummation of these terms in non-equilibrium shows that the scale invariance persists up to single-particle energies $E$ of the order of $eU$ \[30\]. It may be shown that the range of $eU$ values for which scaling behavior is obeyed is bounded from below by the Kondo temperature $T_K$. For large bias voltage, deviations from scaling occur when $eU$ becomes comparable to the band cutoff or to the local level $|E_d|$. 

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**Figure 3** Leading contributions to the effective electron-electron vertex induced by a 2CK impurity. Solid, dashed, and wiggly lines correspond to conduction electron, pseudofermion, and slave boson propagators, respectively.
Figure 4  Scaled non-equilibrium distribution functions for a short ($L = 1.5\,\mu m$, diffusion const. $D = 65\,cm^2/s$) and a long ($L = 5.0\,\mu m$, diffusion const. $D = 45\,cm^2/s$) Cu nano-wire. Black curves: Experimental data for $U$ in a range between 0.05mV and 0.3mV in steps of 0.05mV, taken from Ref. [18]. The two curves showing deviations from scaling are at small voltages, 0.05 mV and 0.1mV, respectively. Light curves: Theory in the scaling regime, $eU \gg T_K$. The 2CK impurity concentration $c_{imp}$ obtained from the fit is approximately $8 \cdot 10^{-6}$/(lattice unit cell).
Deviations from scaling. Experimental data are the same as in Fig. 4, upper panel. Open circles represent the fit of the theory to the first experimental distribution function which shows deviations from scaling at small voltage, $eU=0.1\text{meV}$. Since the 2CK impurity density $c_{\text{imp}}$ has been determined as the only adjustable parameter in the scaling regime (Fig. 4), for given transport voltage $U$, the Kondo temperature $T_K$ (or the temperature $T$, whichever is larger), is the only parameter that controls deviations from scaling at small voltage. It may be determined from the fit as $T_K \approx 1K$.

The remaining task is to calculate the non-equilibrium distribution function $f_x(E,U)$ at an arbitrary position $x$ along the wire (compare Fig. 1) in the presence of 2CK defects. In the regime where the inelastic mean free path $\ell_{\text{inel}}$, approximately given by the average spacing $a$ between 2CK impurities, is small compared to the wire length, but large compared to the elastic mean free path $\ell$, $L \gg \ell_{\text{inel}} \approx a \gg \ell$, the dynamics of a given 2CK impurity is determined by interactions with electrons probing many different spatial regions in the wire. It is, therefore, appropriate to first calculate the averaged quasiparticle distribution $f_x(E,U)$ as the solution of the impurity ensemble averaged Boltzmann equation, subject to the boundary conditions $f_{x=0}(E,U) = f^\alpha(E+eU)$, $f_{x=L}(E,U) = f^\alpha(E)$. The equation of motion for $f$ is obtained by summing over all local quasiparticle momenta $p$ and exploiting the fact that the current in the disordered system is diffusive,
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\[ \vec{j}_x(E) = \sum_p \frac{p}{m} f_x(E,U) = -D \nabla_x \rho_x(E) = -D \nabla_x \sum_p f_x(E,U). \]  

(3.5)

Here, \( \nabla_x \) denotes the component of the gradient in the direction along the wire. In the stationary case, the resulting diffusive Boltzmann equation reads \[ \nabla_x^2 f_x(E,U) = -\frac{1}{D} C_{2CK} \left[ f_x(E,U), c_{imp} \right], \]  

(3.6)

where \( C_{2CK} \) is the collision integral due to 2CK scattering. For small impurity concentration \( c_{imp} \) is proportional to \( eU \). Since the diffusion constant \( D \) is measured experimentally, \( c_{imp} \) is the only adjustable parameter of the theory in the scaling regime.

In the experiment, the tunneling current Eq. (2.1) measures the local (unaveraged) distribution function of electrons in the vicinity of the junction. It is determined by the local stationarity condition that it must be equal to the distribution function of the 2CK impurity states. The results are shown in Fig. 4 and display scaling behavior for \( eU \gg T_K \). It is seen that there is excellent quantitative agreement between theory and experiment for all samples. Considering the fact that no other interaction producing \( E/eU \) scaling is known to date, this provides strong evidence for the presence of 2CK impurities in evaporated Cu nano-wires. From the fit of the theory to that experimental curve which for the first time shows deviations from scaling as the voltage is decreased, i.e. for \( eU = 0.1 \text{meV} \) (Fig. 5), the corresponding experimental low energy scale may be identified with the low energy scale of the theory, the Kondo temperature \( T_K \).

Thus we have \( T_K \approx 1K \), which is in rough agreement with earlier experimental results on TLS in Cu point contacts \[ 9, 10 \].

4 Relation to dephasing

In equilibrium, the non-vanishing 2CK quasiparticle scattering rate \( 1/\tau(E) \) crosses over to a pure dephasing rate \( 1/\tau_\phi \), as the quasiparticle energy approaches the Fermi surface, \( E, T \to 0 \), since at the Fermi energy no energy exchange is possible. One might, therefore, conjecture that degenerate TLS could be the origin of the dephasing time saturation observed in magnetotransport measurements of weak localization. This assumption is indeed supported by several coincidences between the dephasing time measurements and the results on the non-equilibrium distribution function: (1) The dephasing time \( \tau_\phi \) extracted from magnetotransport experiments \[ 24, 13 \] is strongly material, sample, and preparation dependent. This suggests a non-universal dephasing mechanism, like TLS, which is not inherent to the electron gas. (2) The dephasing time in Au wires is generically shorter than in Cu wires \[ 19 \]. This is consistent with the fact that the
estimates for the TLS concentration $c_{imp}$, obtained from the fit of the present theory to the experimental distribution functions, is much higher in Au than in Cu wires \[30\]. (3) In Ag wires one observes neither dephasing saturation nor $E/eU$ scaling of the distribution function. This is consistent with the assumption that there are no 2CK defects present in the Ag samples \[31\].

5 Concluding remarks

We have calculated the quasiparticle distribution as a function of the excitation energy $E$ in diffusive nano-wires in the presence of 2CK impurities, when the system is driven far away from equilibrium by a finite transport voltage $U$. The present theory reproduces the experimental finding that in Cu and Au wires the nonequilibrium distribution function displays the scaling property $f(E,U) = f(E/eU)$ for $eU$ above a low-energy scale $T_o$. Within the theory, $T_o$ is given by the Kondo temperature $T_K$ or by $T$, whichever is larger. The 2CK impurity density is the only adjustable parameter of the theory in the scaling regime. The quantitative agreement between the present theory and experiment and the fact that the experimental scaling property is not even qualitatively explained by any other type of interaction, provide strong evidence for the existence of 2CK impurities in Cu and Au wires, while there seem to be no such defects present in Ag wires. We have not attempted here to give a microscopic model for the physical realization of 2CK defects. For that purpose, it should be useful to perform numerical simulations \[24\] of dislocations in the respective materials.

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