Enhancement of the Kondo temperature of magnetic impurities in metallic point contacts due to the fluctuations of the local density of states

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Abstract

The effect of local density of states (LDOS) fluctuations on the dynamics of a Kondo impurities in a small metallic point contact (PC) is studied. To estimate the spatial and energy dependent LDOS fluctuations we investigate a model PC by means of a transfer matrix formalism. For small PC’s in the nanometer scale we find that near to the orifice strong LDOS fluctuations develop. These fluctuations may shift the Kondo temperature by several orders of magnitude, and result in a strong broadening of the PC Kondo peak in agreement with the results of recent measurements.

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Magnetic impurities have been studied in the past few years very successfully using ballistic point contacts (PC’s) [1,2,3,4]. Very recently a thorough reinvestigation of the Kondo effect has been carried out by Yanson et al. [5] using small metallic PC’s with the diameters in the mesoscopic range, \(d \sim 20\text{Å}\). Surprisingly, in contrast to measurements in thin metallic films and quantum wires, where a suppression of the Kondo effect has been found [6], Yanson et al. observed a strong broadening of the zero bias Kondo peak which has been explained by the increase of the Kondo temperature of the impurities in the contact region [5].

In this Letter we report on the first study of the local density of states (LDOS) and their effect on the Kondo resonance in small ballistic PC’s. We find surprisingly strong LDOS fluctuations in the contact region which are generated by scattering of the conduction electrons at the surface of the PC and give a natural explanation to the broadening of the Kondo peak in very small PC’s [5].

The effect of LDOS fluctuations on the Kondo resonance has already been studied in the case of disordered metals [7]. However, Dobrosavljević et al. calculate the effect of the fluctuating LDOS only in the leading logarithmic order and their results are only valid in the case where the characteristic energy scale of the fluctuations, \(\epsilon_c\) is of the same order of magnitude as the bandwidth cutoff, \(D \sim \epsilon_c\). As we shall see in a PC \(\epsilon_c \ll D\) thus we had to treat the fluctuations with more care. Furthermore, we also constructed the next to leading logarithmic scaling equations which goes beyond the theory of Ref. [7].

To study the effects of the LDOS fluctuations on a Kondo impurity we investigate the dynamics of a magnetic impurity in the contact region. For this purpose we use a simplified Hamiltonian. The first part of our Hamiltonian describes the conduction electrons: \(H_0 = \sum_{n,\sigma} \epsilon_n a_{n\sigma}^+ a_{n\sigma}\), where the operator \(a_{n\sigma}^+\) (\(a_{n\sigma}\)) creates (annihilates) a conduction electron with spin \(\sigma\), wave function \(\varphi_n\), and energy \(\epsilon_n\). The states \(\varphi_n\) are not momentum eigenstates but rather appropriately chosen scattering states of the PC. The interaction part of the Hamiltonian can be written as
where the impurity is at the position \( \mathbf{r} \), \( J \) is the strength of the local exchange interaction, and \( \sigma \) and \( S \) denote the spin of the conduction electrons and the impurity, respectively.

Far from the orifice the LDOS must approach its bulk value: \( \tilde{\varrho}(\mathbf{r}, \epsilon) \rightarrow \tilde{\varrho}_{\text{bulk}}(\epsilon) \). For the sake of simplicity we assume that the bulk DOS is constant between the high- and low-energy cutoffs, \( \tilde{\varrho}_{\text{bulk}}(\epsilon) = \tilde{\varrho}_0 \). However, approaching the contact region \( \tilde{\varrho}(\mathbf{r}, \epsilon) \) deviates from its bulk value and random spatial and energy dependent LDOS fluctuations appear.

To estimate the amplitude of these fluctuations we carried out numerical calculations for a free electron model PC consisting of two infinite half-spaces connected by a cylinder of radius \( R \) and length \( L \) [9]. To simplify the calculations we used angular momentum eigenstates propagating along the axis \( z \) of the PC of the form:

\[
\varphi_{\pm, \epsilon \lambda m}(r, z) = e^{i\varphi m} J_m(\lambda r) e^{\pm ik_z(\lambda) z},
\]

where \( m \) is the angular momentum around the axis \( z \) and cylindrical coordinates have been used. The signs \( \pm \) correspond to right- and left-going states, respectively, \( \epsilon \) denotes the energy of the conduction electron, and \( J_m \) stands for the \( m \)'th Bessel function. The \( z \) component of the momentum in Eq.(2), \( k_z \), can be expressed by the energy \( \epsilon \) and the radial momentum \( \lambda \) of the electron as \( k_z = (2m_\epsilon \epsilon - \lambda^2)^{1/2} \) and \( k_z = i(\lambda^2 - 2m_\epsilon \epsilon)^{1/2} \) for \( \sqrt{2m_\epsilon \epsilon} \lambda < \lambda \) and \( \sqrt{2m_\epsilon \epsilon} > \lambda \), respectively. \( m_\epsilon \) denotes the electron mass. Since we require the vanishing of the wave functions at the boundaries, \( \lambda \) is a continuous parameter in the infinite half-spaces while it takes discrete values inside the tube.

The scattering matrices of the problem can be constructed by matching the wave functions and their derivatives at the two ends of the cylinder \([9,10]\). Having obtained the scattering matrices one can proceed by constructing all the scattering states and evaluating the different physical quantities like the LDOS or the conductance of the PC \([9]\).

Fig. 1.a shows the calculated LDOS fluctuations inside the tube for a PC with \( R = 15\AA \) and \( L = 15\AA \) at the point \( r = 7\AA \) and \( z = 7\AA \), where the coordinate \( z \) is measured from the left wall of the PC. Each time a new conduction channel is opening a peak appears in
the function $g(\epsilon, r, z)$. As one can see in Fig. 1b the strong interference effects cause strong fluctuations even for a fixed energy if the spatial coordinate is varied.

In order to explore the effect of these LDOS fluctuations on the Kondo impurity we construct the next to leading logarithmic scaling equations for an arbitrary LDOS using the multiplicative renormalization group technique \[ 13 \]. Following Abrikosov \[ 12 \] we describe the impurity spin dynamics by means of a pseudofermion field, $S^i \rightarrow b^+_s S^i_{s'} b_s$, where the operators $b^+_s (b_s)$ create (annihilate) a pseudofermion corresponding to the spin state $S^z = s$.

The pseudofermion Green’s function, $G(\omega)$, and the vertex function, $\Gamma_{nn'}(\omega)$ can be introduced in the usual way \[ 12 \]. Due to the special structure of the interaction the self-energy of the conduction electrons [the vertex function] factorizes: $\Sigma_{nn'}(\omega) = \varphi^*_n(r)\Sigma(\omega)\varphi_{n'}(r)$ $[\Gamma_{nn'}(\omega_i) = \varphi^*_n(r)\Gamma(\omega_i)\varphi_{n'}(r)]$, where $\Sigma(\omega)$ and $\Gamma(\omega_i)$ depend only on the LDOS at the position of the impurity. Then the multiplicative renormalization group equations can be written in the form \[ 13 \]:

\begin{align}
G(\omega, j, D) &= Z(j, D/D') \, G(\omega, j, D) , \\
\Gamma(\omega_i, j, D') &= Z(j, D/D')^{-1} \, \Gamma(\omega_i, j, D) ,
\end{align}

(3)

where $D'$ is the scaled bandwidth, $Z$ is the pseudofermion wave function renormalization factor, and $j'$ denotes the dimensionless scaled coupling, $j' = J(D')g_{\text{bulk}}(\epsilon_F)$, $\epsilon_F$ being the Fermi energy.

Similarly to the case of Kondo impurities or two level systems in a smooth LDOS \[ 11,13,17 \] the leading logarithmic terms in the scaling equation arise from the second order vertex corrections while the next to leading logarithmic terms are generated by the second order pseudofermion self energy corrections and the third order vertex corrections. Having calculated these diagrams one can easily generate the scaling equations using Eq. (3) and after some lengthy algebra one obtains:

\begin{align}
\frac{dj}{dx} &= j^2 (R(D) + R(-D)) - 2j^3 \int_0^D \frac{d\xi}{D} \frac{1}{(1 + \xi/D)^2} \\
& \quad \times \left( R(\xi)R(-D) + R(-\xi)R(D) \right) ,
\end{align}

(4)
where \( R(\xi) = \varrho(\xi)/\varrho_{\text{bulk}}(\epsilon_F) \), \( \xi \) being the energy of an electron measured from the Fermi energy, \( \xi = \epsilon - \epsilon_F \) and we introduced the scaling variable \( x = \ln(D_0/D) \), \( D_0 \approx \epsilon_F \) being the initial bandwidth cutoff. Naturally, for a system without LDOS fluctuations \( R = 1 \) and Eq.(\( \text{I} \)) reduces to the usual next to leading logarithmic scaling equation [11].

To see how the LDOS fluctuations modify the Kondo temperature we drop the next to leading logarithmic term in Eq.(\( \text{I} \)) and calculate the leading logarithmic Kondo temperature, which is associated to the divergence of the scaled coupling: \( j(D = T_K) = \infty \). Assuming that both the bulk and the fluctuation induced Kondo temperatures, \( T_K^* \) and \( T_K \) are small enough to satisfy \( \varrho(\xi = T_K) \approx \varrho(\xi = T_K^*) \approx \varrho(\epsilon_F) \) one easily obtains the following expression for the ratio of the two Kondo temperatures

\[
\frac{T_K}{T_K^*} = \exp \left\{ \int_{T_K^*}^{D} \frac{d\xi}{2\xi} (\delta R(\xi) + \delta R(-\xi)) \right\}, \tag{5}
\]

where \( \delta R(\xi) = R(\xi) - R_{\text{bulk}}(\xi) \). The important feature of Formula (5) is the appearance of the weight \( 1/\xi \) in the exponent. The physical meaning of this factor is that the Kondo resonance is mainly formed by the low-energy electron-hole excitations. The contribution of high-energy electron-hole excitations is also significant but it is suppressed compared to that of the low-energy ones. Roughly speaking most of the sites with \( \delta \varrho(\epsilon_F) < 0 \) will have an increased \( T_K \) while impurities for which \( \delta \varrho(\epsilon_F) > 0 \) will tend to have a \( T_K \) decreased.

The relative Kondo temperature \( T_K/T_K^* \) can be easily estimated by substituting the ratio \( \delta R(\xi) \) of the free electron calculations into Eq.(5). The calculated average ratio \( < T_K/T_K^* \) as a function of the radius \( R \) of the PC for fixed length \( L = 5\text{Å} \) is shown in Fig. 2(a). The increased average Kondo temperature, \( < T_K \sim 1 - 10 K \), is orders of magnitude larger than bulk Kondo temperature, \( T_K^* = 0.01 K \), and is in reasonable agreement with the experiments (see the diamonds in the Figure) [19]. A rough fitting of our data with the experimentally found power law dependence \( < T_K \sim d^{-\alpha} \) gives an exponent \( \alpha = 2.2 \pm 0.5 \) which agrees qualitatively with the experimental exponent, \( \alpha = 2 \). (Unfortunately, the available PC diameters were limited by our computer capacity and only a rough estimation of \( \alpha \) could have been carried out.) As a comparison we also show the average \( < T_K/T_K^* \) calculated
by integrating the next to leading logarithmic scaling equations, Eq.(4). As one can see in Fig. 2(a) there is no significant difference between the next to leading logarithmic and the leading logarithmic results.

Determining the resistivity contribution of a magnetic impurity at a site $r$ in an ultrasmall PC is a very complicated task because at the length scales involved in the problem the usual Boltzman equation description \[14\] breaks down, and presently there is no theory available which could take into account both the geometrical effects (i.e., the strong quantum interference) and the strongly correlated behavior of a Kondo impurity in the PC. Therefore we proceed in a semiquantitative way and we estimate the scattering rate $1/\tau(\omega)$ of a conduction electron passing through the orifice. This is connected to the imaginary part of the conduction electrons’ self energy, $1/\tau(\omega) = -2\, Im\, \Sigma(\omega)$, and can be measured directly in a PC experiment \[3][14]. Since the conduction electrons’ Green’s function remains unrenormalized in the dilute impurity limit this scattering rate is simply proportional to $1/\tau(\omega) \sim 2\pi S(S+1)(\varrho(\epsilon_F)J(\omega))^2\epsilon_F$, where $J(\omega)$ denotes the scaled coupling at the energy $D = \omega$. As in the unitary limit the contribution of an impurity in the contact region to the conductance of the PC is approximately $\sim -e^2/h$ in the $\omega = eV \to 0$ limit, and $\varrho(\epsilon_F)J(\omega) \to 1$ for $\omega \to 0$ (see Eq.(4)), the change in the conductance of the PC due to magnetic scattering can be estimated as

$$\Delta G(eV) = -\frac{e^2}{h} \Omega \varrho \varrho^2(\epsilon_F) < J^2(\max\{eV,T\}) \ , \quad (6)$$

where $c$ is the concentration of the impurities, $< ...$ denotes the average over the contact region and $\Omega = 8R^3/3$ is the effective volume of the PC \[3\].

The calculated amplitudes of the impurity contributions to the zero voltage conductance of the PC as a function of the system size are compared to the experimental data in Fig. 2(b). The average in Eq. (6) has been carried out over 40 randomly chosen impurity positions. Both the amplitude of $\Delta G(R)$ and its sample to sample fluctuations are in very good agreement with the experiments. We stress at this point that there is no free parameter in Eq. (6) except for the length of the PC which hardly influenced our results. To show that there is a
striking size effect in Fig. 2(b) we also plotted the calculated amplitude of the Kondo signal with impurities having the bulk Kondo temperature. Clearly, both the amplitude and the size dependence ($\Delta G \sim d^{2.2}$) of the experimental and our calculated fluctuation dependent Kondo conductances are quite different from the one we obtained by assuming the bulk Kondo temperature ($\Delta G \sim d^3$).

The size dependence of the Kondo resonance can be understood as follows. Decreasing the contact size strong LDOS fluctuations evolve in the contact region. The smaller the contact size the larger the fluctuations become, and thus for small contact sizes the measured few impurities (i.e., the impurities in the contact region) have a very broad distribution of Kondo temperatures. Since in the temperature range $\sim 1 K$ where the measurements were performed magnetic impurities with a small Kondo temperature, $T_K \approx T_K^* \sim 0.01K$, give no significant contribution to Eq.(6) the differential conductance is always dominated by the impurities with the largest Kondo temperatures. These arguments suggest that the sum in Eq.(6) is dominated by the magnetic impurities having large Kondo temperatures. Therefore our theory gives a natural explanation why the applied magnetic field could not destroy the zero bias Kondo anomaly for small samples.

One can also show analyzing formula (5) that for a fixed PC shape the relative increase of the Kondo temperature, $T_K/T_K^*$, depends essentially on the bulk Kondo temperature $T_K^*$, and for alloys with very small $T_K^*$ the ratio $T_K/T_K^*$ can be much larger than for alloys with large $T_K^*$. This explains why no significant broadening of the Kondo peak has been observed in $CuFe$ PC’s ($T_K^* \approx 20K$) [4], while an enormous broadening was found in $AuMn$ and $CuMn$ ($T_K^* < 0.01K$) [3,5].

According to the picture proposed in this Letter one expects that any physical quantity which is sensitive on the LDOS will have a distribution in the contact region due to the presence of strong fluctuations. As an example we mention the PC spectrum of fast two level systems (TLS) [15,16]. Since the dimensionless TLS – conduction electron couplings are sensitive on the LDOS [7,8] and $T_K^*$ is in the range of $\sim 1K$ [13] we expect a slight broadening of the zero bias TLS peak, which has indeed been observed in metallic break
In our simple model the LDOS fluctuations were basically connected to the openings of new conductance channels through the PC [10]. Such LDOS fluctuations, however, may also be generated by random scattering at the boundary of the PC. Effectively, it has been found recently that random scattering at the surface of a PC is able to produce huge LDOS fluctuations in the contact region [20]. Therefore we think that our conclusions are qualitatively independent of the special model considered.

The case of ultrasmall PC’s with magnetic impurities [5] should be contrasted to the experiments performed on thin metallic films and wires [11]. In the latter case only alloys with high Kondo temperatures (CuFe, AuFe) were investigated, where the LDOS fluctuations do not modify the Kondo temperature very much. Instead, it seems to be that spin orbit interaction induced spin anisotropy develops which leads to the blocking of the spin flip processes [21] and a suppression of the Kondo signal. On the contrary for thin AuMn films, where $T_K^\ast$ is very small, we expect that the Kondo temperature can be increased due to the surface induced LDOS fluctuations by several orders of magnitude similarly to the case of PC's.

In summary, we have proposed that the recently measured anomalous Kondo temperatures in small CuMn PC’s are due to the strong LDOS fluctuations in the contact region generated by the scattering of the conduction electrons at the surface of the PC. Carrying out a model calculation we found that these fluctuations are really able to increase the Kondo temperature by several orders of magnitude in agreement with the measurements. Our results also suggest that the Boltzman equation approach becomes inadequate for ultrasmall PC spectrometry, where quantum interference and fluctuation effects may become extremely important.

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FIGURES

FIG. 1. (a): Fluctuations of the LDOS $\rho(\epsilon, z, r)$ inside the tube for a PC with $R = 15\text{Å}$ and $L = 15\text{Å}$ at the point $r = 7\text{Å}$ and $z = 7\text{Å}$. The fluctuating LDOS does not integrate exactly to its bulk value (dashed line) because the hard wall of the PC pushes the conduction electrons in the inside region of the tube. (b): $\rho(\epsilon, z, r)$ for the same PC at $z = 7\text{Å}$ and energy $\epsilon = 7eV$ as a function of the radius $r$.

FIG. 2. (a) The average relative Kondo temperatures $< T_K/T^*_K$ as a function of the radius $R$ of a $CuMn$ PC with $L = 5\text{Å}$. The cutoff and the bare couplings have been chosen to be $D_0 = 6.8eV$ and $j_0 = 0.032$. Crosses and diamonds denote the results obtained in the leading and in the next to leading logarithmic orders, while data points indicated by boxes have been obtained from the results of Ref. [5]. The large fluctuations are due to the sensitivity of the interference pattern to the geometry of the PC. (b) Size dependence of the amplitude of the dimensionless Kondo conductance $\Delta g = \Delta G h/e^2$ for the same contact. Diamonds denote the experimental data taken from Ref. [5] while our results are indicated by crosses. The impurity concentration and the temperature were $c = 0.1\%$ and $T = 0.05K$, respectively. The dashed line indicates the results without LDOS fluctuations ($g \sim R^3$) while the continuous line corresponds to the best fit to the data of Ref. [5]: $g \sim R^{2.17}$.
FIG. 1. G. Zaránd and L. Udvardi: "Enhancement of the Kondo temperature of magnetic impurities in metallic point contacts..."
FIG. 2. G. Zaránd and L. Udvardi: "Enhancement of the Kondo temperature of magnetic impurities in metallic point contacts..."