Size dependence of Kondo scattering in point contacts: Fe impurities in Cu

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The size dependence of electron-spin scattering is investigated by means of point-contact spectroscopy of the Kondo alloy *Cu*Fe(0.1 at. %). We can study junctions of various sizes ($d \simeq 1.5$–35 nm) using mechanically controllable break junctions (MCB's). In a previous MCB study of the Kondo alloy *Cu*Mn the Kondo temperature T_K was observed to increase by many orders of magnitude in this range of contact sizes. For *Cu*Fe we also find an increase of T_K, but only by a factor ~ 2. This observation is in qualitative agreement with the theory recently proposed by Zárand and Udvardi.

Recent developments in producing three-dimensional (3D) metallic point contacts by the mechanically controllable break-junction (MCB) technique[1] have opened new opportunities for detailed study of the size dependences in many interesting phenomena, such as conductance quantization,[2] superconductivity of weak links,[3] and magnetic impurity scattering (Kondo effect).[4,5] Via MCB various ranges of contact sizes, previously inaccessible, become continuously available from several tens of nm down to the one-atom scale.

In view of previous measurements of size effects in low-dimensional *Cu*Fe, *Au*Fe, and *Cu*Cr Kondo alloys,[6–8] it was surprising to find that the Kondo scattering in *Cu*Mn and *Au*Mn point contacts increases with decreasing contact size.[4,5] Using standard theory[9] for the analysis of the data, a huge increase of the effective Kondo temperature was obtained. Three salient features appear in the low-temperature differential resistance versus voltage characteristics. The first is a weaker dependence of the zero-bias Kondo-peak intensity on the contact size, compared to the intensity of the phonon structure. The second feature manifests itself through a noticeable broadening of the Kondo maximum for ultrasmall contacts, showing explicitly that a larger energy scale (or, equivalently, Kondo temperature T_K) is involved. Finally, the third concerns the magnetic-field behavior of the zero-bias Kondo peak. While for contacts larger than about several tens of nanometers the expected Zeeman splitting is observed,[4,5] it disappears for contact sizes smaller than 10 nm, illustrating the more robust screening of the impurity magnetic moment.

Zárand and Udvardi[10] recently proposed a theory which predicts an enhancement in T_K due to large fluctuations in the local electronic density of states (LDOS) forming in narrow constrictions. The effect should be more prominent for low bulk Kondo temperatures. Stimulated by this work we undertook a detailed study of the size dependence of Kondo scattering by Fe impurities in *Cu* point contacts. The bulk T_K of *Cu*Fe is much greater (≈ 30 K) than that of *Cu*Mn (≈ 0.01 K) and according to theory should be much less enhanced in small contacts. Our measurements have found only a very small size dependence of the Kondo effect in contacts with sizes down to 1.5 nm, in qualitative agreement with the theoretical predictions.

Three samples in the shape of wires with 0.25 mm diameters were prepared from Cu alloys with 0.1 at. % of Fe. Note that this alloy has nominally the same impurity concentration as the *Cu*Mn alloy which we have systematically studied before.[4,5] The wires are broken at low temperatures in the MCB setup and microscopic contacts were formed using piezoelectric control. The contact size d is estimated from R_0, the contact resistance which we take equal to the minimum value of the differential resistance (see Fig. 1), using the Wexler formula.[11] Because the bulk mean free path due to magnetic scattering is comparable with our larger contact diameters, the Wexler corrections to the Sharvin formula are necessary. Similar to the *Cu*Mn alloys,[4] we define the relative intensities of the Kondo peak,[12] $\delta R_K / R_0$, and the increase of differential resistance due to phonon scattering, $\delta R_{\rm ph}/R_0$, as shown in Fig. 1. The steep rise of $dV/dI(V)$ at the characteristic Cu phonon energies, 15–20 mV,[11] evidences that we are in the spectroscopic regime of charge

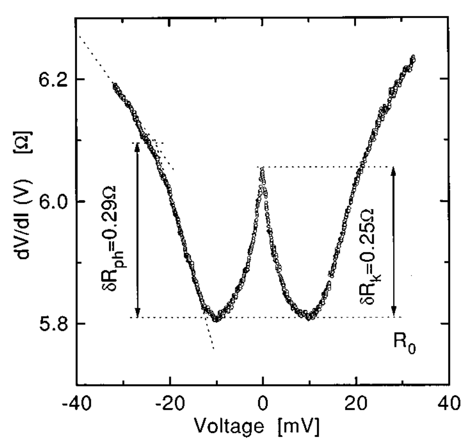

FIG. 1. Differential resistance dV/dI as a function of voltage bias recorded for a *Cu*Fe(0.1 at. %) junction with $R_0 = 5.8\ \Omega$ ($d = 14.6$ nm) and $T = 0.5$ K, $H = 0$.

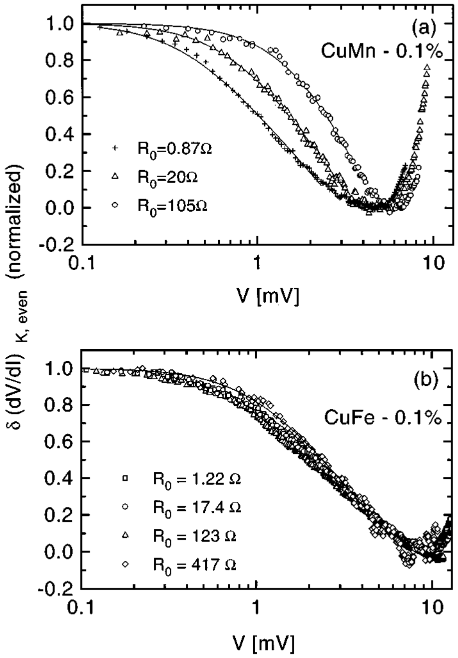

FIG. 2. (a) The normalized even parts of the Kondo maximum for *Cu*Mn(0.1 at. %). These data are taken from Fig. 2 of Ref. 4. The height of the Kondo peak δR_K is 0.003, 0.08, and 0.32 Ω for the 0.87, 20, and 105 Ω samples, and the fitted T_K are 22, 42, and 100 K, respectively. (b) Similar curves recorded for *Cu*Fe(0.1 at. %) with resistances (diameters): 1.22 Ω (33.8 nm, $\delta R_K=0.06$ Ω), 17.4 Ω (8.23 nm, $\delta R_K=0.40$ Ω), 123 Ω (3.0 nm, $\delta R_K=2.8$ Ω), and 417 Ω (1.6 nm, $\delta R_K=4.0$ Ω). The solid lines represent a fit to the data using Eq. (1) and scatter between 31 and 54 K in (b).

transport through the point contact. Since in our experiments the temperature was about 0.5 K and modulation voltage did not exceed 40–50 μV, the experimental resolution is better than 0.17 meV.

The low-bias portions of the point-contact spectra, normalized as $(dV/dI)/\delta R_K$, are plotted on a semilogarithmic scale for *Cu*Fe, and by way of comparison for *Cu*Mn, in Fig. 2. All the *Cu*Fe spectra for resistances R_0 from 1 Ω to 417 Ω converge essentially into a single curve, slightly shifted to higher energies for the smallest contact (largest R_0). In contrast, the *Cu*Mn(0.1 at. %) spectra, taken from Ref. 4 and plotted in the same manner, show a clear increase of the energy scale, determined by the Kondo temperature T_K, for large resistances.

In Fig. 3 the Kondo peak is shown for the contact of $R_0=2.74$ Ω [$d=21.7$ nm, $\delta R_K(0\ \mathrm{T})=0.12$ Ω], measured in magnetic fields up to 12 T. The data are normalized to the zero-bias Kondo peak at $H=0$. Similar measurements were carried out for several other contacts with different resistances in order to investigate the size dependence of the magnetic-field behavior of the Kondo scattering. Opposite to the *Cu*Mn contacts of about the same size, there is *no* Zeeman splitting of the Kondo maxima by the magnetic field, since the bulk Kondo temperature for Fe impurities in *Cu* ($T_K \simeq 30$ K) is significantly larger than the temperature of our measurements, and thus the impurity magnetic moment is more effectively screened by conduction electrons. Never-

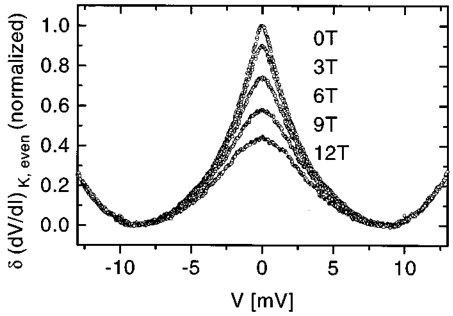

FIG. 3. The magnetic-field dependence of the normalized even parts of the Kondo structure for a *Cu*Fe(0.1 at. %) contact with $R_0=2.74$ Ω [$d=21.7$ nm, $\delta R_K(0\ \mathrm{T})\ =\ 0.12$ Ω].

theless the magnetic field strongly suppresses the intensity of the Kondo peak acting in a fashion analogous to raising the temperature or voltage bias.[13]

To compare the magnetic-field suppression of the Kondo peak in contacts of different size we plot in Fig. 4 the normalized intensities of the zero-bias Kondo maximum, as a function of field for contacts with resistances R_0 differing more than an order of magnitude, corresponding to a sixfold change in the contact size. Unfortunately, contacts with smaller sizes become unstable in high magnetic fields and this hinders the measurement. Together with voltage bias dependences presented in Fig. 2, Fig. 4 represents the central result of our work: *Although the size dependence of the Kondo scattering in CuFe alloys is considerably smaller than in CuMn alloys, a weak size dependence is still clearly observed.* The smaller the contact size, the less is the magnetic-field suppression of the Kondo peak, and this can be qualitatively interpreted as the result of a corresponding increase of the effective Kondo temperature. We will discuss this property in greater detail below.

A comparison of our experimental results with the theoretical predictions is not a simple task, since the perturbation approach breaks down for temperatures and energies comparable to and smaller than the Kondo temperature, which is quite high in the case of *Cu*Fe alloys. There is no rigorous theory for point-contact transport properties of Kondo alloys

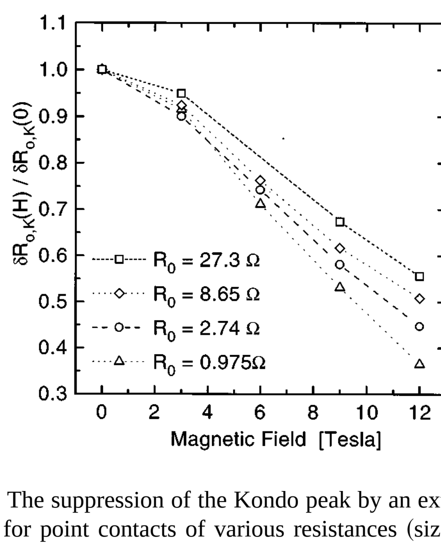

FIG. 4. The suppression of the Kondo peak by an external magnetic field for point contacts of various resistances (sizes). δR_K is 0.90, 0.21, 0.12, and 0.057 Ω for decreasing R_0.

in our energy range of interest. The available approximations do not fully describe the temperature dependence of the bulk resistivity at temperatures around and below T_K.[13] The best we can do for temperatures below the Kondo temperature is to use the empirical relation[7,13] for the resistivity as a function of temperature

$$\rho_m = A - B \ln[1+(T/\theta)^2], \tag{1}$$

where $\ln(\theta/T_K) = -\pi[S(S+1)]^{1/2}$, S being the impurity spin, which for the case of Fe should be taken equal to 1/2 and leads to the relation $\theta \simeq 0.066 T_K$.

The voltage dependence of the point-contact resistance mimics the temperature dependence of the bulk resistivity. To make a direct comparison one has to know the coefficient α in the relation $eV \simeq \alpha k_B T$. Here we adopt a simplified approach by taking the relation between T and V to be the same as for the thermal limit, i.e., $eV \simeq 3.63 k_B T$.[14] Using this relationship and Eq. (1) we can attempt to quantify the data shown in Fig. 2 with T_K as our parameter. The fit of the curves (solid line) yields reasonable accordance with the bulk value of $T_K \approx 34$ K for *Cu*Fe, spreading between 31 and 54 K for various contact sizes. For the sake of clarity only one fit curve is shown. Note how the effective Kondo temperature of *Cu*Mn dramatically increases over the same range of contact diameters [see the solid curves and corresponding T_K's in Fig. 2(a)].

A small trend towards larger Kondo temperatures for decreasing contact sizes can be recognized for *Cu*Fe, even though the scatter is large. As an alternative approach to the size dependence, we consider the clear suppression of the Kondo peak with magnetic field (Fig. 3). In order to accomplish this we adopt the reasonable view,[13,15] supported by experimental results for $T < T_K$ (Ref. 7) and theoretical prediction for the perturbative regime,[15] that a magnetic field can be incorporated in the effective temperature through the relation

$$T_{\text{eff}} = \sqrt{T^2 + \beta^2 H^2}. \tag{2}$$

Here the units of T and H are matched via $\mu_B H = k_B T$ and $\beta \approx 1$ in units of K/T.

Replacing T in (1) by the effective temperature defined in (2) we can fit the effect of the magnetic field on the zero-bias resistance of the junctions, and obtain T_K in an independent way. Doing this, T_K increases from 30 K at $R_0 \simeq 1\ \Omega$ to about 50 K at a contact resistance of $R_0 \simeq 30\ \Omega$. The curves of dV/dI as a function of V (Fig. 3 at $H=0$) and dV/dI at $V=0$ as a function of H (Fig. 4) are consistent in shape and absolute values and give indeed similar values for T_K. However, the dependence on contact size is more clearly seen in the field dependences.

The Kondo effect in nanosized 3D point contacts of *Cu*Fe dilute alloys appears to be size dependent qualitatively in the same way as it was found for *Cu*Mn material: as the contact size decreases the Kondo temperature increases. However, quantitatively a dramatic difference exists between these two kinds of Kondo alloys. While for *Cu*Mn an increase of the Kondo temperature can be observed up to many orders of magnitude, from the bulk value of 0.01 K to tens and hundreds of K in contacts of several nanometers in diameter, for the *Cu*Fe contacts of the same size the increase does not exceed several tens of K, i.e., the bulk Kondo temperature of 30 K increases by only a factor of $\simeq 2$. Qualitatively, such a difference can be explained by the LDOS fluctuation theory of Zárand and Udvardi.[10] They consider the position- and energy-dependent variations in the electron density of states which appear as a result of scattering at the boundary of the point contact. The density of states N_F enters in the exponent of the expression for the Kondo temperature $T_K = T_F \exp(-1/JN_F)$, where T_F is the Fermi temperature and J is the exchange coupling constant. Magnetic impurities sitting at a maximum in the LDOS will have a high T_K, and the high-T_K impurities will dominate the Kondo scattering in the contacts. It is clear that the size effect should be most pronounced for materials with a low bulk Kondo temperature (low J). For higher T_K values the size effect is further limited by the energy scale E_c of the LDOS fluctuations,[16] which can be expressed as $E_c = E_F/N_c$, with N_c the number of conduction channels. For the contacts in Fig. 4 E_c is less than $\simeq 50$ K, which agrees with the fact that we observe the enhancement of T_K in our experiment to be limited to $\simeq 50$ K. For a quantitative comparison, a more exact theory is required to relate T_K to the LDOS fluctuations in the case where $k_B T_K$ is comparable to E_c.

It is interesting to compare our results to the absence of a size dependence in the Kondo temperature of thin films and nanofabricated wires.[6,7] This absence appeared to contradict the point-contact experiments on *Cu*Mn.[4,5] The present results on *Cu*Fe, however, clarify the discrepancy with those experiments. Following Zárand and Udvardi[10] we suggest that the effect of LDOS fluctuations was not observed in previous experiments since these were performed on materials with a high bulk Kondo temperature (*Cu*Fe, *Cu*Cr, and *Au*Fe) and at rather large sizes. We propose that in the recent study of thin films and wires of *Cu*Mn 0.1 at. %,[17] the anomalous temperature dependence of resistivity which was observed could be due to the effective Kondo temperature increase induced by the LDOS fluctuations in those mesoscopic samples. Such local-density-of-states fluctuations have been the subject of previous theoretical attention especially near the metal-insulator transition,[18] where they were predicted to lead to non-Fermi-liquid behavior. The fluctuations may also lead to a number of other interesting effects, such as size dependence of the Ruderman-Kittel-Kasuya-Yosida interaction in spin glasses,[5] a modification of the heavy-fermion state with decreasing contact size, and a gradual metal-insulator transition for contacts with decreasing size and increasing disorder in atomic positions. In addition the superconducting properties of ultrasmall weak links can be strongly affected and we are presently employing our MCB technique to explore these areas.

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