Size Dependence of Kondo Scattering in Point Contacts

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By utilizing a mechanically controllable break junction, point contact spectra dV/dI were determined for various noble metals (Cu;Au) doped with magnetic impurities (Mn). The contact diameter d could be varied continuously between ca. 2 and 50 nm. The zero bias maximum in dV/dI due to the Kondo scattering exhibits a large broadening and *increase* of relative amplitude as d is decreased. By using the theoretical expression for the logarithmic slope of dV/dI vs V we find a significant enhancement of exchange coupling parameter J, which leads to an enormous *increase* in the apparent Kondo temperature T_K for small contact diameters.

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More than a decade ago, the first studies of point contact spectroscopy (PCS) in metals with magnetic impurities [1–3] resulted in the observation of many features analogous to the well-known phenomena in the electrical resistance versus temperature of bulk alloys. In PCS the current-voltage (I-V) characteristics are measured, and for spin scattering processes voltage plays the role of temperature in this analogy. The features observed include the Kondo dependence $dV/dI \sim \ln(V)$ for $eV \gg k_BT_K$, where T_K is the Kondo temperature and the saturation of the contact resistance for $eV \ll k_BT_K$. In all these studies the dependence of the magnetic-impurity scattering on the contact diameter d was not investigated due to the limited range of contact resistances (or equivalently contact sizes) which were then accessible.

In recent years, size effects in Kondo phenomena have attracted much attention and have become a highly controversial issue [4–6]. The experiments mainly employ thin, narrow films to test the theoretical conjecture that, for length scales below a certain Kondo radius $R_c = \hbar v_F/2\pi k_BT_K$ ($\sim$1 μm for $T_K = 1$ K), the Kondo scattering contribution to the resistivity will be reduced. At present there is no overall experimental agreement regarding this conjecture.

We have decided to attack this size problem via PCS using a novel technique: a mechanically controllable break junction (MCB). From the anomalous behavior of the zero bias peak in the differential resistance we find that the apparent Kondo temperature increases many orders of magnitude for contact diameters decreasing from 50 to 2 nm, in contrast to the previously reported behavior for thin films [6].

The MCB technique, described in Ref. [7], enables us to make point contacts with characteristic dimensions down to one atom size [8]. The alloy to be studied, in the form of a wire of 0.1 mm diam, is glued on top of a substrate. At low T, under UHV conditions, the substrate is bent, causing the wire to break. The two ends are brought back into contact by ultrafine piezoelectric control. The great advantage of this technique, apart from producing contacts with exceptionally high stability, is the possibility to continuously change the contact sizes for a single sample over a wide range, without polluting the contact region with additional impurities. The main results presented here have been obtained for a *Cu*Mn (0.1 at.%) alloy and were repeated for three different samples. In addition, experiments have been performed on 0.3 and 1.0 at.% *Cu*Mn and on 0.15 and 0.05 at.% *Au*Mn [9]. Both the first and second derivatives of I-V characteristics were recorded, using a standard modulation technique. The spectroscopic resolution was taken to be equal to $\sqrt{(5.44k_BT)^2 + (1.22eV_{1,0})^2}$, which is standard for PCS and inelastic tunneling spectroscopy [10]. Here, $V_{1,0}$ is the applied voltage modulation amplitude. The temperature for the measurements was between 0.5 and 1.5 K.

Figure 1 displays the typical I-V derivatives for a contact resistance of about 20 Ω in a *Cu*Mn (0.1 at.%) alloy. In a similar experiment on pure Cu, without magnetic impurities, the differential resistance is flat up to ±10 mV, followed by a steep increase as a result of phonon scattering [11]. The contact resistance R_0 is defined by the horizontal tangent to the curve. The diameter of the contacts can be obtained from the resistance through the Sharvin relation for ballistic contacts [11], $R_0 = 8(h/e^2)(k_Fd)^{-2}$, which, using the values for Cu, gives $d = 30/\sqrt{R_0}$, where R_0 is expressed in ohms and d in nm. The full electron-phonon-interaction (EPI) function can be directly obtained from the second derivative, dR/dV (Fig. 1), which agrees well with published results [11]. The maximum M of dR/dV is used to calculate the maximum value of the EPI function for this point contact. In most cases this value is close to the tabulated value of 0.2 [11] which clearly proves the ballistic character of the current flow in the contacts. The background signal at high bias in Fig. 1(b) is fairly large, but also spectra with $\sim$3 times lower background have been obtained with comparable parameters for the Kondo anomaly. From the resolution of the spectra at high bias, we conclude that Joule heating of the sample is negligible.

The magnetic impurity scattering produces a maximum in dV/dI at zero bias as shown in the inset of Fig. 1(a).

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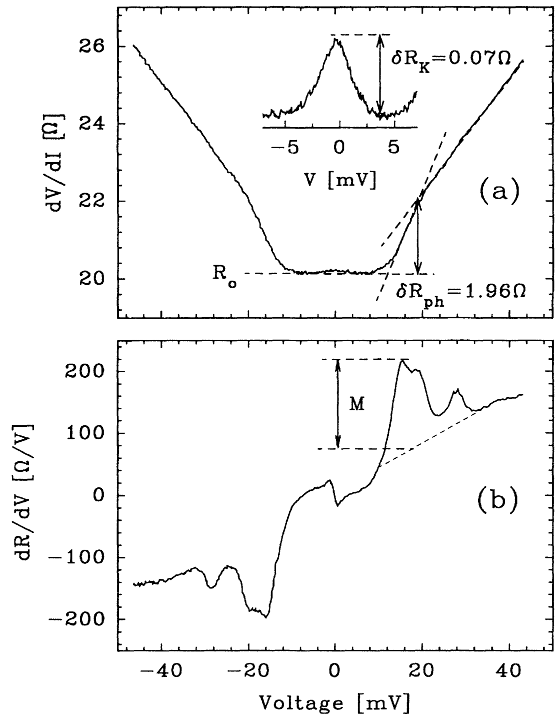

FIG. 1. (a) Differential resistance dV/dI as a function of voltage bias recorded for a *Cu*Mn (0.1 at.%) junction with $R_0 \simeq 20\ \Omega$ and $T = 0.5$ K. The inset shows the Kondo peak structure on an expanded scale. (b) Second derivative $dR/dV = -R^2(d^2I/dV^2)$ of the *I-V* curve for the same junction showing the EPI spectrum beyond ± 10 mV and the Kondo anomaly at zero bias.

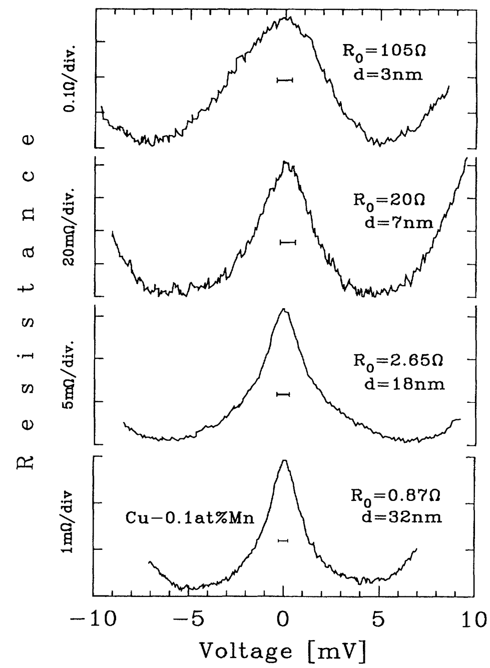

FIG. 2. The Kondo maximum in the differential resistance for point contacts of *Cu*Mn (0.1 at.%) with resistances R_0 between 0.87 and 105 Ω. The horizontal bars indicate the experimental resolution.

This zero bias Kondo peak is plotted in Fig. 2 for several contact resistances of the same alloy. The main result of our experiment is immediately apparent from this figure. The width of the Kondo peak should be determined by a unique energy scale: the Kondo temperature T_K, which for bulk *Cu*Mn is about 0.01 K. The experimental width of the Kondo peak increases strongly as the contact diameter decreases from 32 to 3 nm.

As is further seen in Fig. 2, the amplitude of the zero bias peak has an anomalous dependence on d. For any scattering mechanism it is expected that $\delta R/R_0 \sim d/l$, with the mean free path l independent of d. Indeed, for the phonon spectrum we find an initial dependence $\delta R_{\mathrm{ph}}/R_0 \sim d$, with a tendency to saturation at higher d, when we go from the ballistic limit to diffusive point contacts. For the Kondo peak we find, in a series of over 30 contacts with d ranging from 2 to 50 nm, that the relative amplitude $\delta R_K/R_0$ is nearly *constant,* at a value of about 3×10^{-3}. (δR_K and δR_{ph} are defined in Fig. 1.) Thus, δR_K is seen to *increase* relative to δR_{ph} with decreasing contact size. This suggests an increase in the effective scattering cross section of the Kondo impurity and proves that the larger width of the Kondo peak is not the result of a spurious degradation of the spectral resolution. In some of the PC spectra we observe a well-resolved fine structure which is controlled by the amount of disorder in the contact. We leave this behavior for future study and limit our discussion to data without such structure. For contact resistances above about 200 Ω the average number of impurities in the contact is of order unity, and as a result *both* anomalously large or small values for δR_K have been observed.

In the unitary limit, $k_BT, eV \ll k_BT_K$, each impurity is expected to reduce the conductance by an amount of order e^2/h. Figure 3 plots $\delta G_K = \delta R_K/R_0^2$ divided by e^2/h as a function of d for the 0.1% alloy. The solid line gives the unitary limit for the average number of impurities in the volume $\pi d^3/6$ of the contact. The dependence $\delta R_K/R_0 \sim$ const results in $\delta G_K \sim d^2$ rather than $\delta G_K \sim d^3$. Since the bath temperature is about 100 times larger than the bulk Kondo temperature, this should result in a δG_K 1 order of magnitude below the unitary value. Thus, the Kondo signal for small contacts is larger than expected, but it is consistent with the enhanced T_K for small contacts derived below.

For quantitative analysis of our measurements we turn to the standard theory for Kondo scattering in point contacts [2,3,12]. In second-order Born approximation the magnetic impurity contribution to the PC spectrum is given by [12]

$$-\frac{d^2I}{dV^2} = Q\frac{1}{V} \qquad (eV \gg k_BT)\,. \tag{1}$$

The parameter Q (evaluated for a clean circular contact and taking the free ion spin value $S = 5/2$ for Mn) is

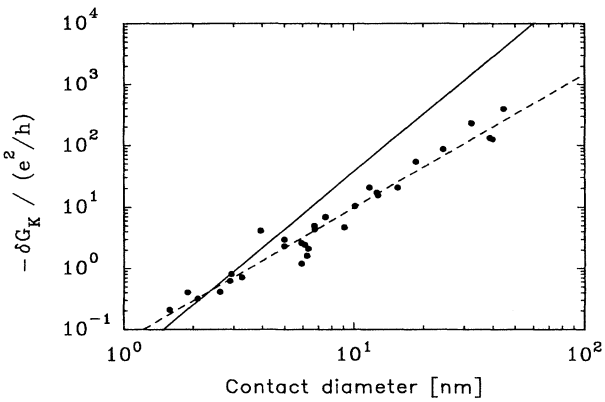

FIG. 3. The amplitude of the zero bias minimum in the conductance in units of e^2/h plotted as a function of the contact diameter d. The solid line is the unitary limit for the average number of impurities in the contact volume (proportional to d^3). The broken line is a least-squares fit with a power $d^{2.17}$.

given by $Q = (105/8)c(dk_F/R_0)(J/E_F)^3$, where J denotes the exchange interaction constant and c the impurity concentration. Using the free-electron values appropriate for Cu we obtain for J/E_F

$$\frac{J}{E_F} = -0.044\left(\frac{1}{c\sqrt{R_0}}\frac{dR}{d\log_{10}V}\right)^{1/3}, \tag{2}$$

which involves the logarithmic derivative of the differential resistance R at low voltages, with R and R_0 expressed in ohms.

By plotting the differential resistance as a function of the logarithm of the voltage, we can extract via (2) values for J/E_F from the Kondo peaks. These values are plotted as a function of the contact diameter d in Fig. 4(a) for 0.1% and 1.0% *Cu*Mn. Similar behavior was obtained for other concentrations and for the *Au*Mn alloys. An example of the $R(V)$ dependence on a semilogarithmic scale is shown in the inset of Fig. 4(a). We also include two points measured by Ralph and Buhrman [13] on a nanofabricated point contact of *Cu*Mn (0.02 at.%).

The increase of interaction strength J/E_F for decreasing d is consistent with our finding that $\delta R_K/R_0$ is nearly d independent. This can be seen from the fact that $dR/d\log_{10}V$ in (2) is about $\delta R_K/2$ and with $\delta R_K/R_0 =$ const and $R_0 \sim d^{-2}$, we obtain $J/E_F \sim d^{-1/3}$ in agreement with Fig. 4(a).

It is common practice to express the interaction of the conduction electrons with the impurity spins in terms of a Kondo temperature T_K. The latter can be calculated from J/E_F in a standard way [14]:

$$k_B T_K = E_F \exp\left(-\frac{2}{3J/E_F}\right). \tag{3}$$

Using $E_F = 7$ eV for Cu we can determine the values of the apparent T_K for our contacts; these are plotted in Fig. 4(b). We obtain the approximate relation $T_K \sim d^{-2}$ (or, equivalently, $T_K \sim R_0$), which appears to be quite general, for all our concentrations and host metals. The

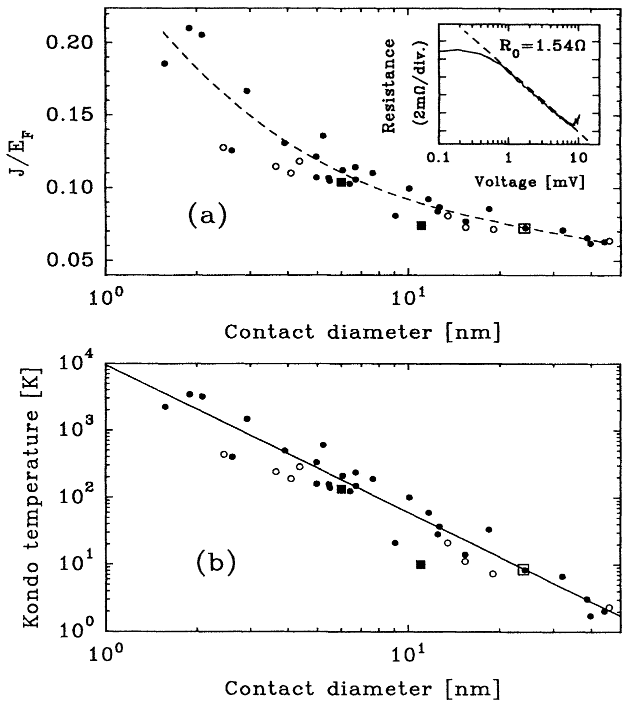

FIG. 4. The exchange interaction parameter J/E_F (a) and the corresponding Kondo temperature (b) plotted as a function of the contact diameter for *Cu*Mn (0.1 at.%) (•) and (1.0 at.%) (∘). The solid squares show the points derived from [13] on a nanofabricated point contact of a 0.02% alloy. The inset illustrates, for the point enclosed in a small square, the raw data on a semilogarithmic scale permitting the determination of $dR/d\log_{10}V$.

contacts for the 1% alloy are no longer purely ballistic; therefore, corrections to the estimates for d were applied using Wexler's formula [11]. The fact that we find consistent values for concentrations ranging from 0.02% [13] up to 1% suggests that impurity-impurity interactions do not influence the effect.

For small contacts there is a more direct way to determine T_K. At low voltages the resistance follows a quadratic dependence $R(V) = R(0)[1 - (V/V^*)^2]$, in analogy to the usual temperature dependence of the resistivity in the unitarity limit. With $eV^* \simeq 0.1 k_B T_K$ [14], we obtain values for T_K of the same order of magnitude.

Further support for a size dependent Kondo energy scale comes from the magnetic field dependence. Figure 5(a) shows a Kondo peak for a 1.2 Ω junction, which develops a minimum at zero bias under the influence of an external field, with a width of the expected size $2g\mu_B H$. For the smaller contact 10.2 Ω junction in Fig. 5(b) the same field only reduces the Kondo peak intensity, but no splitting is observed. For $R_0 = 1.2$ Ω we see from Fig. 4 that $T_K \simeq 6$ K, thus fields of a few tesla are comparable in magnitude to the Kondo temperature. However, for $R_0 = 10.2$ Ω, we have $T_K \simeq 70$ K, so that an 8 T field is only a weak perturbation on the Kondo interaction. For

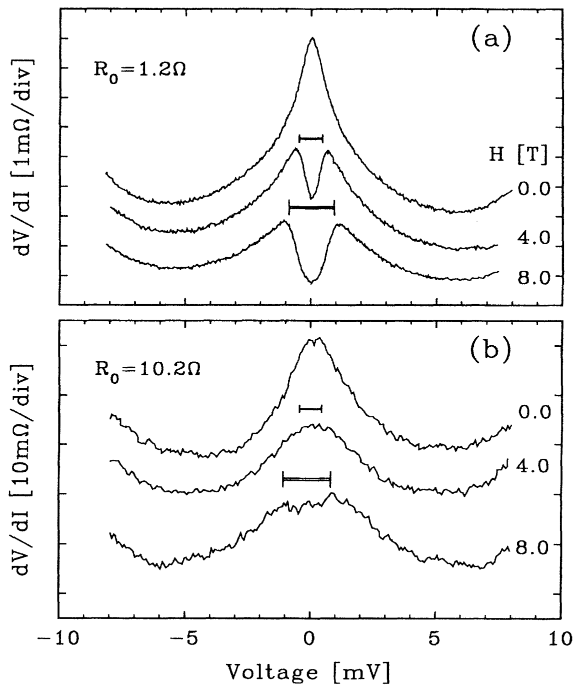

FIG. 5. Effect of a magnetic field on the Kondo peak (a) for a 1.2 Ω junction and (b) a 10.2 Ω junction of *Cu*Mn (0.1 at.%). The magnetic field suppresses the Kondo peak and induces a gap at zero bias for the low resistance junction, but is unable to do this for the high resistance one. The horizontal bar denotes the expected splitting $2g\mu_B H$.

resistances below 1 Ω the Kondo peak is eliminated at a few tesla, while for R_0 greater than 10 Ω the field merely broadens and reduces the peak. By contrast, the one point measured by Ralph for 0.02% Mn at $T = 0.05$ K [13] and $R_0 = 25$ Ω, still shows splitting in a 4 T field. Whether this difference is due to impurity concentration, temperature, contact geometry, or fabrication method remains to be investigated.

The apparent Kondo temperature measured here is a transport property, and the relation with the thermodynamic Kondo temperature requires further study, since it is difficult to imagine how the true Kondo temperature can be so dramatically affected by the contact size. At this point it is interesting to note the following:

(1) The bulk value of T_K for *Cu*Mn is usually taken to be of order 0.01 K. Extrapolating the dependence in Fig. 4(b) towards this value we obtain a characteristic length scale of $d_c \sim 0.5$ μm. Because the Kondo radius R_c for *Cu*Mn (0.1 at.%) is much larger than the bulk mean free path l, we use a diffusive Kondo radius [6] $\xi_K = \sqrt{lR_c}$. Taking $l \simeq 100$ nm we obtain $\xi_K \simeq 3$ μm, which is somewhat larger than our extrapolated length scale. Unfortunately, it is difficult to extend our experiments to contacts of this large size.

(2) The 2D confinement in the contact results in a finite number of conduction channels which are separated in energy by an amount $\Delta \simeq 8E_F/d^2k_F^2$. If the resolution with which we can measure the Kondo resonance is limited by this energy [5], then the identification $k_BT_K \sim \Delta$ produces the correct size dependence and the correct order of magnitude.

In summary, from the anomalous dependence of the width, the amplitude, and the field dependence of the zero bias peak we observe an increase in the Kondo scattering for point contacts with decreasing contact diameters. The complete description of these results requires an extension of the theory of Kondo scattering to include this mesoscopic regime.

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