Finite-Size Effects in the Low-Temperature Resistivity of CuCr Films

J. F. DiTusa, K. Lin, M. Park, M. S. Isaacson, and J. M. Parpia

Laboratory of Atomic and Solid State Physics and School of Applied and Engineering Physics, Cornell University,
Ithaca, New York 14853

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The effect of finite sample size on the Kondo resistance anomaly has been explored in the CuCr system. The resistivity of thin CuCr films was studied as a function of width from 0.5 to 20 K. The magnitude of the Kondo anomaly is significantly depressed as the film width is reduced below 10 μm. This length scale is consistent with the radius of the electron-spin correlation cloud, suggesting a crossover from two- to one-dimensional behavior. A concurrent decrease in the temperature of the resistivity maximum suggests a diminished interimpurity interaction strength.

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The effect of finite film thickness on dilute magnetic alloys has been studied only recently [1-4]. Measurements of the low-temperature resistivity of thin films sought to characterize the electron-magnetic-impurity interaction lengths. In one of these experiments, the Kondo resistance anomaly in AuFe films was depressed as the film thickness was reduced below 250 nm [1]. The authors conclude that a dimensionality crossover from three to two dimensions was a consequence of a film thickness smaller than the conduction-electron magnetic-screening cloud. However, the mean free path is also a function of film thickness and it is not clear what effect this has on the interaction length. At higher concentrations, a study of the resistivity maximum associated with the spin-glass transition as a function of film thickness did not reveal any information on the interaction lengths [2]. A reduction in the temperature of the resistivity maximum (T_m) could be explained by either a shortened elastic mean free path or an increased contribution to the resistivity due to a disorder-enhanced Coulomb repulsion.

This paper presents the results of an experiment which probes the conduction-electron-magnetic-impurity interaction in the CuCr system as a function of film width. In this study the mean free path is held nearly constant. The additional resistivity due to weak localization and interaction effects can be ignored. We conclude that the measured reduction in the magnitude of the Kondo effect as well as T_m are due to a crossover from two- to one-dimensional behavior with respect to the conduction-electron-magnetic-impurity screening lengths.

The Kondo effect is a thoroughly studied resistance anomaly in bulk systems [5]. A metal with magnetic impurities shows a resistivity that is well described by

$$\rho = A_d + \tfrac{1}{2}B\left\{1 - \frac{\ln(T/T_K)}{[\ln^2(T/T_K)+\pi^2 S(S+1)]^{1/2}}\right\}$$

for temperatures at or above the Kondo temperature, and by

$$\rho = A_d + \tfrac{1}{2}B - \frac{1}{2\pi}B\left\{\frac{1}{[S(S+1)]^{1/2}}\ln\left[\frac{\theta^2+T^2}{T_K^2}\right]^{1/2}\right\}$$

for temperatures below the Kondo temperature [6]. Here A_d is the temperature-independent part of the resistivity, B the step height between the low- and high-temperature plateaus of the Kondo resistivity, T the temperature, T_K the Kondo temperature, S the local magnetic moment of the magnetic impurity, and $\ln(\theta/T_K) = -\pi[S(S+1)]^{1/2}$. The parameter B is dependent on the host metal, the impurity species, and concentration c. B is proportional to c for concentrations small enough (≤ 100 ppm for CuCr) that interimpurity interactions are unimportant.

The aim of this experiment was to determine the length scale relevant to the interaction of conduction electrons with magnetic impurities. This length should manifest itself in the slope of the logarithmic temperature dependence near T_K,

$$d\rho/d\lambda = -B/2\pi[S(S+1)]^{1/2}$$

[$\lambda = \ln(T/T_K)$], when measured as a function of sample size. The slope, $d\rho/d\lambda$, is perhaps the most experimentally accessible of the parameters associated with the Kondo effect since at low temperatures its measurement is not dependent on the subtraction of the phonon contribution to the resistivity [6]. We measured $d\rho/d\lambda$ as a function of both film width and film thickness.

The thin films prepared for this experiment were deposited by thermal evaporation from a single CuCr source of known Cr concentration [7]. The substrate is a silicon nitride film grown on a silicon wafer. Thin film strips as well as current and voltage pads were defined by electron lithography and photolithography using a lift-off process. The lithographic techniques allowed the fabrication of thin films of width varying between 0.14 and 35 μm deposited on a single substrate in the same evaporation. The film thickness was measured with a crystal thickness monitor.

The advantages of a study of the effect of sample width rather than film thickness on the Kondo effect are transparent. By preparing several films on the same substrate in a single evaporation one can ensure sample consistency. Furthermore, by studying films where the mean free path (l) is thickness limited we can vary the size of the system without affecting l. The first set of samples are films with a nominal 1000 ppm Cr in Cu concentration that have a thickness of 21.2 nm and width that ranged from 0.14 to 35 μm. Figure 1(a) shows a continuous decrease in the parameter $d\rho/d\lambda$ from 3.8 to 1.3 nΩ cm as the film width is decreased below 9 μm. The decrease in $d\rho/d\lambda$ is large, a factor of 3 over the width and temperature range studied. The method of sample preparation assures a constant magnetic-impurity concentration and degree of impurity clustering for the six films. It is surprising that for such a thin film (21.2 nm thick) we still witness a depression of the Kondo effect at widths of a few microns.

The data in Fig. 1(b) show the resistivity for a sequence of three widths of a 98.0-nm-thick film evaporated from the same source material. In these thicker films, $d\rho/d\lambda$ is larger than in the 21.2-nm films by a factor of $\simeq 9$. This indicates a suppression of $d\rho/d\lambda$ with film thickness which is comparable in magnitude to the suppression due to film width measured in the 21.2-nm film [1]. The depression of $d\rho/d\lambda$ as the width decreases in this thicker film is apparent (a change from 20 to 15 nΩ cm) although it is not as dramatic as in the thinner films.

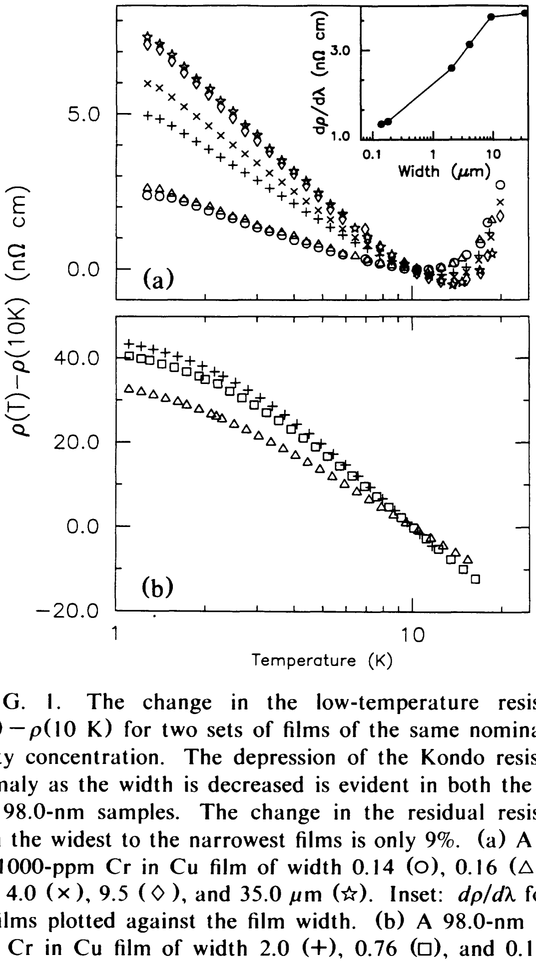

FIG. 1. The change in the low-temperature resistivity $\rho(T)-\rho(10\ \mathrm{K})$ for two sets of films of the same nominal impurity concentration. The depression of the Kondo resistance anomaly as the width is decreased is evident in both the 21.2- and 98.0-nm samples. The change in the residual resistivity from the widest to the narrowest films is only 9%. (a) A 21.2-nm 1000-ppm Cr in Cu film of width 0.14 (○), 0.16 (△), 2.0 (+), 4.0 (×), 9.5 (◇), and 35.0 μm (☆). Inset: $d\rho/d\lambda$ for the six films plotted against the film width. (b) A 98.0-nm 1000-ppm Cr in Cu film of width 2.0 (+), 0.76 (□), and 0.16 μm (△). The depression of the Kondo anomaly with film thickness is evident from the difference in $d\rho/d\lambda$ in the films of the same width and impurity concentration.

For concentrations exceeding 100 ppm in the Cr in Cu system, interimpurity interactions play a role in determining the low-temperature resistivity. In this range of impurity concentration, the total Kondo resistivity contribution B will no longer be proportional to c and therefore $c^{-1}d\rho/d\lambda$ decreases as c increases. Previous studies of this system in the very dilute limit [6] show that for bulk samples $c^{-1}d\rho/d\lambda \simeq 0.23$ nΩ cm/ppm. For our 1000-ppm Cr in Cu, 98.0-nm-thick, 2.0-μm-wide film we measure a $c^{-1}d\rho/d\lambda$ of 0.020 nΩ cm/ppm. The transition from the 3D to 2D limit as the film is thinned can reduce the Kondo effect by an order of magnitude over the range of thickness that we have explored [see Figs. 1(a) and 1(b)]. The results shown in Fig. 1(a) illustrate that for a relatively thin film, well into the 2D regime, a reduction of the width of the film into the 1D regime results in a similar reduction of the Kondo contribution to the resistivity. We cannot ascribe all of the factor of 11 reduction in $c^{-1}d\rho/d\lambda$ discussed above to a 3D to 2D size effect since, at the concentration, interimpurity interactions are known to reduce $c^{-1}d\rho/d\lambda$ to below the unitarity limit [6]. The width study shows that the length scale associated with the reduction of the Kondo effect is of order ~ 1 μm. Therefore, a film of 98.0-nm thickness should not show the bulk Kondo effect. Thus we infer that the interimpurity interactions decrease $c^{-1}d\rho/d\lambda$ by less than a factor of 10 at this concentration.

It was suggested by Chen and Giordano that a decreased Kondo anomaly resulted from the spatial confinement of the electron screening cloud [1,8,9]. In their study of the Kondo resistance anomaly, a decreased $d\rho/d\lambda$ was attributed to a film thickness smaller than R_K, the radius of the electron screening cloud [10]. An estimate for $R_K \sim \hbar v_f/k_B T_K$ in the CuCr system [1,11,12] yields a value of $R_K = 1.0$ μm ($T_K \sim 2$ K). Our results of the width study are compatible with this length scale. While R_K is the calculated value of the spatial extent of the spin correlation density, this quantity is not easily connected with observables [8] such as $d\rho/d\lambda$. It is therefore not possible to make a more quantitative evaluation of our results.

There may be other factors affecting $d\rho/d\lambda$. One can argue that a finite mean free path would have the effect of decreasing the size of the electron screening cloud. For $l < R_K$ the motion is diffusive on the length scale over which the screening of the magnetic impurity will occur. If R_K is considered the length over which the electron spins are randomized then this length is modified [1,8,9] to be $(R_K l)^{1/2}$. In the study of 21.2-nm-thick samples, l is thickness limited and is calculated to be roughly 15 nm [13]. The diffusion-limited screening length is calculated to be 0.12 μm, a much smaller length than the width at which appreciable depression of $d\rho/d\lambda$ is measured. A mean-free-path–dependent screening length may be used to explain the decrease in $d\rho/d\lambda$ as a function of film thickness where the suppression of l is significant. However, the resistivity of our thin films changes by only 9% over the width range studied. The related change in the mean free path would have little effect on the screening lengths. Thus it does not seem reasonable that this decrease in the mean free path could explain our results.

Weak-localization effects and electron-electron interaction effects contribute a small logarithmic resistance rise to the resistivity of a thin film at low temperatures [14]. However, the expected additional resistance from these mechanisms is at most 2% of the resistance rise measured. Furthermore, the reduction in film width increases the sheet resistance and therefore would increase the $d\rho/d\lambda$ which is an opposing effect. We have ignored these contributions to the resistivity in our discussion of the depression of the Kondo effect.

Impurity clustering can have a large effect on the Kondo resistance anomaly [3,6]. Areas of higher impurity concentration will result in regions of the thin film in which interimpurity interactions are stronger. This will decrease the number of impurity spins which are considered isolated and thus decrease the Kondo effect. However, the material characterization using STEM sets the maximum size of the Cr clusters at 1 to 2 nm [15]. Thus decreasing the width of a thin film from 35 to 0.14 μm will not have an effect on the impurity clustering in these films.

An additional manifestation of a size effect is seen in the lower-temperature resistivity. The resistivity around 1 K of a 20.0-nm 2000-ppm Cr in Cu film is shown in Fig. 2 for three film widths. It is clear that the temperature of the resistivity maximum (T_m) decreases as the sample width is decreased from 2.0 to 0.17 μm [2]. In fact, it is not apparent that the narrowest film will show a resistance maximum at any temperature. These same films have a $d\rho/d\lambda$ at 6 K that decreases from 31.2 to 26.9 nΩ cm. For the impurity concentrations studied, interimpurity interactions are important and are dominated by the conduction-electron-mediated RKKY interaction [6]. The effect of interimpurity interactions on the resistivity is twofold. First, as was noted previously, $c^{-1} d\rho/d\lambda$ decreases as c increases. Second, for this impurity concentration, the resistivity has a broad maximum at a temperature (T_m) above the spin-glass temperature (T_{sg}) [6]. This is related to the smaller spin-flip scattering rate as the impurity spins freeze into a random configuration [16]. Since the impurity concentration is held fixed in this study, we interpret the smaller value of T_m as a *decreased* interimpurity interaction strength. Similarly we interpret the decreased $d\rho/d\lambda$ as a diminished conduction-electron–isolated-impurity interaction. We conclude that both the Kondo effect and the interimpurity interactions are suppressed as a function of film width. Since these two effects are depressed in the same range of film width it is possible that they are manifestations of the same phenomena.

We point out that the contributions to $d\rho/d\lambda$ from interimpurity interactions and conduction-electron–impurity interactions compete with each other. Our observation that $d\rho/d\lambda$ decreases with size implies that the size effect is dominated by the reduction of the conduction-electron–isolated-impurity interaction.

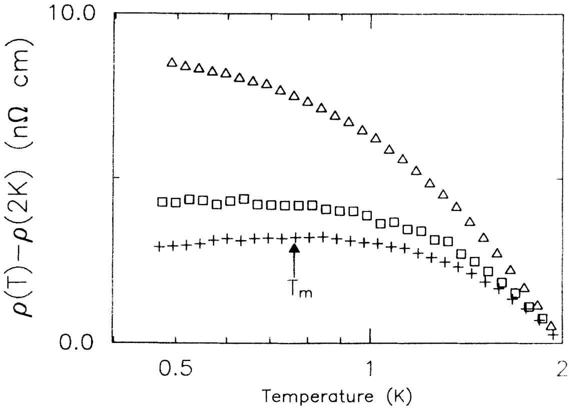

FIG. 2. The change in the low-temperature resistivity $\rho(T)-\rho(2\text{ K})$ for three 2000-ppm Cr in Cu films of thickness 20.0 nm and width 2.0 (+), 0.76 (□), and 0.17 μm (△). The maximum in the resistivity occurs at 0.75 ± 0.02 K for 2.0 μm, at ~0.5 K for 0.76 μm, and we see no evidence for T_m for 0.17 μm. This decrease in the temperature of the resistivity maximum is evidence for a decrease in the interimpurity interaction strength in the narrow films.

It has been proposed that a diminished mean free path would be a mechanism for decreasing the RKKY interaction [2,17]. However, the present understanding considers the RKKY interaction to be insensitive to impurity scattering [18,19]. In either case we do not believe that this is the dominant mechanism for the decreased impurity-impurity interaction in our narrow films. The resistivity changes by only 0.2% in the 2000-ppm, 20.0-nm film over a width variation from 2.0 to 0.17 μm. Any associated change in the RKKY interaction would lead to a much smaller change in T_m than the measured decrease of > 23%.

One may argue that the decrease in both $d\rho/d\lambda$ and T_m for the narrower films is consistent with a lower impurity concentration. The value of T_m increases linearly with impurity concentration and $d\rho/d\lambda$ is dependent on c at this concentration. However, we do not believe that the Cr concentration is smaller in our narrow films. Oxidation of the impurities might be cited as one way of decreasing the magnetic-impurity concentration. In the 1000-ppm Cr in Cu, 21.2-nm-thick films the ratio of volume to surface area changes by only 14% from the widest to the narrowest films. Such a small change cannot account for over a 60% decrease in the apparent impurity concentration in these films. Diffusion of magnetic impurities to the film surfaces during thermal evaporation would be another possible explanation for an apparent decreased impurity concentration [4]. In the study presented here, the film thickness is much smaller than the film width. Therefore a smaller width will not result in an apparent decreased Cr concentration in these films since the ratio of volume to surface area changes relatively little.

In summary, we find that the Kondo resistance anomaly is depressed as the film width is decreased below a few microns in the CuCr system. This depression occurs even though the film thickness is much smaller than R_K. The size of the electron screening cloud is compatible with our observation of the length scale over which the depression of the anomaly occurs, provided that R_K is not reduced by a finite mean free path. An accompanying decrease in T_m is evidence for a diminished interimpurity interaction strength as the sample size is made smaller. The associated reduction in the effective conduction-electron magnetic screening apparent in this experiment is interpreted as a dimensional crossover from two to one dimension. A full understanding of these results is dependent on further theoretical study of the effect of spatial confinement on the Kondo resistance anomaly.

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