Kondo effect in Cu(Fe) films

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Recent experiments have shown that the Kondo effect is suppressed in thin films, a result which has so far eluded theoretical explanation. The previous experiments have been limited to the regime $T \gtrsim T_K$, where T_K is the Kondo temperature. In this paper we present results for $T \lesssim T_K$ which demonstrate that even when the Kondo contribution to the resistivity, $\Delta\rho_K$, is greatly suppressed, the Kondo temperature itself is *not* affected. We also present the results of experiments involving bilayer samples, i.e., Kondo films covered by nonmagnetic layers, which demonstrate the existence of a Kondo "proximity effect."

I. INTRODUCTION AND BACKGROUND

The Kondo effect involves the interaction of a localized magnetic moment with an electron gas,[1,2] and is relevant to the behavior of dilute magnetic alloys, as well as concentrated systems such as the heavy-fermion materials, and perhaps also high-T_c superconductors. The Kondo problem has attracted a great deal of theoretical interest for nearly 30 years, and much has been learned about many-body physics from this work. The single-impurity Kondo problem, i.e., the behavior in the limit of a very low concentration of local moments, is currently believed to be well understood, from both theoretical and experimental viewpoints.[2–4]

Nearly all of the work on the single-impurity Kondo problem has involved bulk, i.e., three-dimensional systems. However, in the past few years several experiments have studied the behavior in lower-dimensional systems, and found some surprising results. Most notably, the Kondo contribution to the resistivity, $\Delta\rho_K$, is greatly suppressed when the sample size is made sufficiently small.[5–8] The "critical" length scale which governs this suppression has been found to be approximately 1500 Å in Au(Fe),[5,7] and somewhat larger (1 μm) in Cu(Cr).[6] This has proved to be a puzzling result, since theoretical arguments[9,10] imply that dimensionality should not play a role in the behavior; that is, it is predicted that $\Delta\rho_K$ should *not* depend on dimensionality, in sharp contrast to the experiments. It thus appears that an important aspect of the Kondo problem is not understood. In this paper we present experimental results concerning the Kondo behavior in two dimensions. Past experiments[5–7] have studied the dimensionality dependence of the Kondo behavior only for temperatures near and above the Kondo temperature T_K. Here we describe experiments to probe the dimensionality dependence of the Kondo effect in the low-temperature limit, $T < T_K$. In addition, we describe bilayer experiments which demonstrate the existence of what might be termed a Kondo "proximity effect."

Kondo[1] was the first to show that the interaction between a local moment and an electron gas leads to an anomalous contribution to the resistivity. At high temperatures this contribution has the now well known form $\Delta\rho_K \sim \log(T)$; this logarithmic dependence persists to temperatures of order T_K, below which $\Delta\rho_K$ approaches a constant value. An attractive qualitative picture of this phenomenon is that the (antiferromagnetic) exchange interaction between the local moment and the conduction-electrons leads to the formation of a screening cloud in which the conduction-electron spin density screens out the local moment.[2] At high temperatures there is only partial screening, but as T is lowered the screening improves to the point that the moment is fully screened for $T \lesssim T_K$. Given this intuitive picture of the Kondo effect, it is natural to consider the size of the screening cloud. Dimensional analysis and also quantitative calculations[11–15] yield a Kondo length scale

$$R_K \approx \hbar v_F / 2\pi k_B T_K \ . \tag{1}$$

For the material considered below, Cu(Fe), $T_K \sim 20$ K,[16] which leads to $R_K \sim 1000$ Å. One might then expect to see a change in $\Delta\rho_K$ in Cu(Fe) films when the thickness is less than this value.

Studies of the Kondo alloy Au(Fe) (Refs. 5, 7) have indeed observed this sort of behavior, but the length scale which governs the suppression was found to be more than an order of magnitude smaller than R_K (1) [as evaluated for Au(Fe)]. There are a number of other length scales which could conceivably play an important role; one which has been mentioned a great deal is an extension of (1) to disordered systems. The result (1) can be derived in various ways;[11–15] one intuitive "derivation" follows by noting that two electrons near the Fermi level whose energies differ by $k_B T_K$ will become "out of phase" after they have traveled a length of order R_K (1). This result assumes ballistic motion of the electrons. If the elastic mean free path ℓ_e is less than R_K, the motion will be diffusive, and the distance traveled will then be

$$R_K^* = \sqrt{R_K \ell_e} \ . \tag{2}$$

For Au(Fe) the experimentally observed crossover length scale is within a factor of 2 of R_K^*, hence this would

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appear to be a possible explanation of the results for Au(Fe). We hasten to add, however, that these are only qualitative arguments; a proper quantitative calculation would be required to settle the issue, and to date such calculations have not lent support to this picture of diffusive corrections to R_K (1).

In any event, the question of whether or not R_K^* is important is one which can be addressed experimentally, and that has been one goal of the work presented in this paper. As we will show below, our experiments with Cu(Fe), when taken together with those obtained in previous work on Au(Fe), suggest that *neither* R_K nor R_K^* is correct as far as the experiments are concerned. Another key issue we address here concerns the behavior of T_K as the dimensionality is reduced. Previous work has been limited to the case $T > T_K$; the results reported below probe the dimensionality dependence in the low-temperature regime $T \leq T_K$. Taken as a whole, our results suggest that physics which is not included in the simplest Kondo model dominates the dimensionality dependence of the Kondo effect.

II. EXPERIMENTAL METHOD

In the present experiments we have studied thin films of Cu(Fe) which were produced by sputtering onto glass substrates at room temperature. The sputtering target was 99.999% pure Cu, onto which were placed several small pieces of Fe (typically 1 mm or less in diameter) in a symmetric arrangement to ensure homogeneity of the Fe concentration.[17] From a consideration of the geometry of the sputtering target we estimate that the Fe concentration in our Cu(Fe) films was approximately 300 ppm. We have obtained an independent estimate of the concentration by comparing our results for very thick (essentially three-dimensional) films with published data for bulk Cu(Fe),[18] and the result is consistent with a value of 300 ppm. Previous work on bulk Cu(Fe) has demonstrated that this is sufficiently low that the effect of interactions between Fe moments on $\Delta\rho_K$ is negligible[18] (at least for bulk systems). Our data for other concentrations support this conclusion. To check for homogeneity we made several test batches, in which a number of different samples with the same thickness were produced. These results showed that the Fe concentration was the same for all of the films prepared in a given deposition. It was more difficult to obtain the same Fe concentration in different depositions, so in the following we will be making direct comparisons *only* between samples prepared in the *same* deposition. By varying the angle between the substrate and the sputtering beam, a wide range of film thicknesses could be obtained in a single deposition.[19] The resistivities of these films varied with thickness, as expected since boundary scattering made a significant contribution. The low-temperature resistivities ρ_0 of a typical batch were 0.89, 0.92, 2.6, and 4.9 $\mu\Omega$ cm for thicknesses of 2000, 1500, 1000, and 500 Å. Thus, the variation of ρ_0 for the thickest films was small, but became larger as the film thickness was reduced. Possible effects of this variation of ρ_0 will be discussed below. As emphasized above, all samples in a given batch were prepared in the same deposition, guaranteeing that they all had the same Fe concentration. The films were patterned photolithographically into strips of width $\sim 150\ \mu$m and length ~ 60 cm, and the resistance was measured as a function of T using standard techniques.

We also studied bilayer samples which consisted of Cu(Fe) bottom layers (prepared as described above) with a layer of nominally pure Cu (99.999%) on top. By using a rotating mask system it was possible to make a collection of samples in which the Cu(Fe) layer of all of the samples was deposited at the same time, with the Cu layer then deposited separately and independently, without breaking vacuum.

III. RESULTS

Figure 1 shows some typical results for the Kondo contribution to the resistivity, $\Delta\rho_K$, as a function of T for Cu(Fe) films. From measurements with pure Cu films, we found that the contribution of electron–phonon scattering to the resistivity becomes important only above about 15 K. We will therefore restrict our analysis to temperatures below 15 K, where only the Kondo effect contributes significantly to the temperature dependence of ρ. We should also add that the contributions of both weak-localization and electron-electron interactions were small and could be neglected, essentially because of the low sheet resistances of our samples. On the scale of Fig. 1 the resistivity of our pure Cu films is temperature independent. Hence, the variation seen in Fig. 1 is due solely to the Kondo effect. For films thicker than about $t = 2000$ Å the Kondo contribution was independent of t, but at smaller thicknesses $\Delta\rho_K$ was suppressed significantly. The inset to Fig. 1 gives the relative magnitude of $\Delta\rho_K$ over the range 1.5–10 K as a function of film thickness, and shows the suppression very clearly.

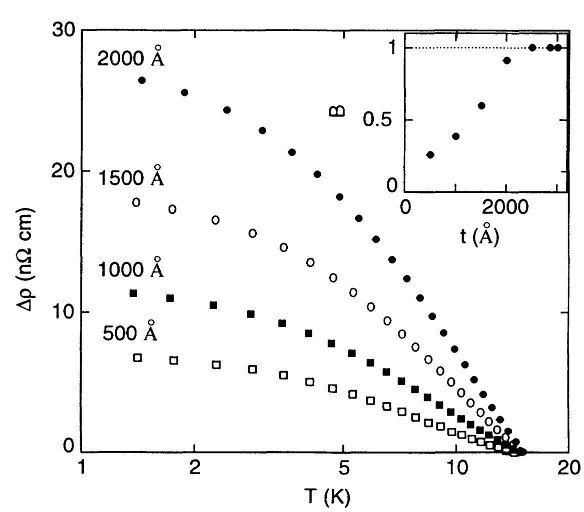

FIG. 1. The Kondo contribution to the resistivity, $\Delta\rho_K$, as a function of T for different sample thicknesses t; the values of t are given in the figure. The inset shows the relative magnitude of $\Delta\rho_K$ at 4 K as a function of t.

The crossover length scale which governs this suppression, i.e., the value of t at which $\Delta\rho_K$ is approximately half the value found in very thick films, is ≈ 1500 Å.

It is natural to compare this with the Kondo length scales R_K and R_K^*, discussed above. For Cu(Fe) the theory predicts $R_K \sim 1000$ Å and $R_K^* \sim 550$ Å (for our films $\ell_e \sim 300$ Å). Thus the experimentally observed crossover length scale is not far from the value predicted by the theory for R_K. It is therefore tempting to conclude that there is agreement between theory and experiment. However, we believe that this conclusion is premature, for the following reason. Previous studies of Au(Fe) *also* found a crossover length scale of 1500 Å. The Kondo temperature of Au(Fe) is approximately 0.2 K, which is two orders of magnitude smaller than that of Cu(Fe). Hence, the theoretical predictions discussed above imply that the Kondo length scale in Au(Fe) should be *much* larger than in Cu(Fe) [note that the Au(Fe) and Cu(Fe) films had similar values of ℓ_e]. R_K for Au(Fe) is longer by a factor of 100, while R_K^* is a factor of 10 larger. Our observation that the experimental length scale does *not* change significantly with T_K is thus in serious disagreement with the theory.

Based on our results for Au(Fe) and Cu(Fe) one might thus conclude that the crossover length scale is the same in all materials. However, studies of the size dependence of the Kondo effect in Cu(Cr),[6] for which $T_K \approx 2$ K, found a length scale of approximately 1 μm. Thus the experimental crossover length scale and T_K do not appear to be correlated in any simple way. The theoretical arguments mentioned above all predict that T_K is the only parameter relevant for determining the Kondo crossover length. Based on our results it seems clear that this is not the case; some other physics, in addition to the Kondo effect, must be controlling, or at least contributing to, the Kondo crossover behavior we have observed. We will return to this point below.[20]

The results shown in Fig. 1, and also those reported previously, clearly show that $\Delta\rho_K$ is suppressed in thin films. It is natural to also consider how T_K is affected by film thickness, a question which was not addressed in previous experiments (which were all limited to temperatures near or above T_K). From Fig. 1 we see that $\Delta\rho_K$ is approximately logarithmic in T for $T > 5$ K, and that the temperature dependence becomes weaker at lower temperatures. This is in good accord with the expected Kondo behavior discussed above, which has been observed in many bulk Kondo systems [including Cu(Fe)].[2,18,21] T_K is, roughly speaking, the temperature at which $\Delta\rho_K$ begins to "roll over." More precisely, it is predicted that $\Delta\rho_K$ is a universal function of T/T_K; thus by fitting measurements like those in Fig. 1 to the theory one could in principle determine T_K. However, so far as we know, none of the available theoretical forms are in terribly good agreement with experiment for bulk systems. Since we are primarily interested in *changes* in T_K we have taken the following approach. Assuming that $\Delta\rho_K$ follows a universal "function" $f(T/T_K)$, one can ask if different samples can be described by the same function with the same value of T_K. With this in mind, we have taken the data in Fig. 1 and scaled the results, i.e., the values of $\Delta\rho_K$, for each film thickness by a constant, temperature-independent factor (this factor is essentially just inversely proportional to the quantity plotted in the inset to Fig. 1). The results are given in Fig. 2, where we see that to within the uncertainties, the curves for each thickness have the same *shape*. Since the temperature axis has *not* been adjusted in any way, all of the data sets can be described by the same function of T, apart from an overall scale factor for the magnitude of the Kondo contribution. Hence, T_K is the *same* for all of the samples. We have obtained a similar result for Au(Fe) films; i.e., T_K is also independent of film thickness for that material.

The results presented above demonstrate that the magnitude of $\Delta\rho_K$ becomes smaller as the film thickness is reduced. We have also studied what might be viewed as the "reverse" effect, in bilayer samples. The sample geometry is sketched in the inset of Fig. 3. The bottom layer is Cu(Fe), deposited in the same manner as for the samples in Figs. 1 and 2. Then, without breaking vacuum, a layer of pure Cu of thickness d_{Cu} was deposited (also by sputtering) on top. The aim of this measurement was to determine how (or if) the Kondo contribution to the Cu(Fe) layer (the bottom film) is affected by the presence of the Cu layer on top. Some typical results for the Kondo contribution to the resistivity are shown in Fig. 3 where we plot $\Delta\rho_K$(bilayer) as a function of T. These samples were all from the same batch; the only parameter which varied was the thickness of the top layer, i.e., the Cu. It is seen that as d_{Cu} is increased, the Kondo contribution to the bilayer resistivity is systematically reduced.

While the quantity plotted in Fig. 3 involves the *entire* bilayer [i.e., the Cu(Fe) and Cu layers in parallel], we are more interested in the behavior of the bottom layer alone, i.e., just the Cu(Fe). We will extract its behavior from these data shortly. However, it is useful to first make a few qualitative observations from the raw data in Fig.

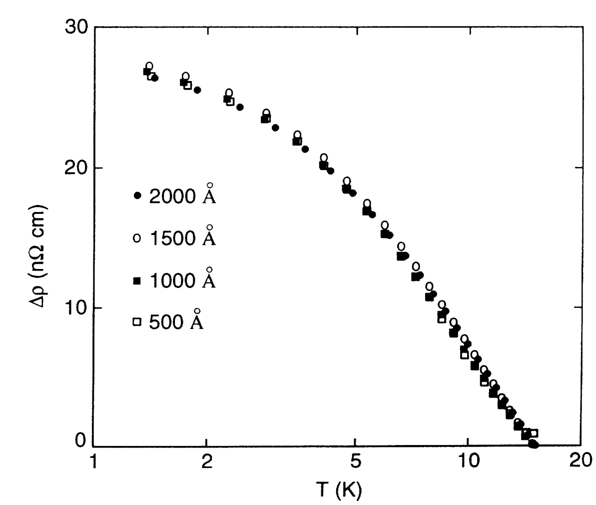

FIG. 2. Scaling plot of the results in Fig. 1. The values of $\Delta\rho_K$ for each sample from Fig. 1 have been scaled by a temperature-independent factor.

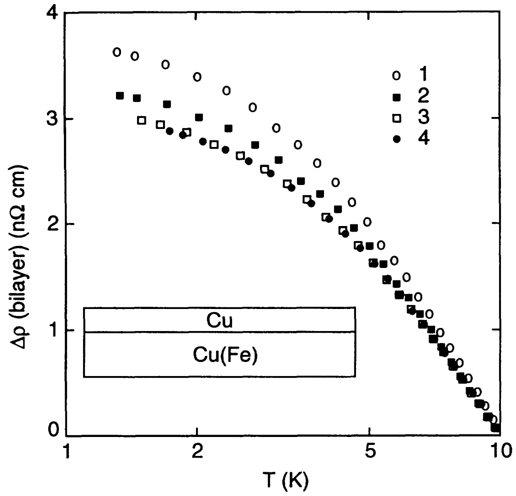

FIG. 3. Kondo contribution to the resistivity of several Cu(Fe)/Cu bilayer samples. The Cu(Fe) layer was 415 Å thick, while the thicknesses of the top (Cu) layers were as follows: sample 1: $d_{\mathrm{Cu}} = 0$; sample 2: $d_{\mathrm{Cu}} = 100$ Å; sample 3: $d_{\mathrm{Cu}} = 200$ Å; sample 4: $d_{\mathrm{Cu}} = 300$ Å. The resistivity of the Cu(Fe) film was 2.0 $\mu\Omega$ cm, while that of the Cu was 0.7 $\mu\Omega$ cm. The inset shows the geometry of the bilayer samples.

3. For simplicity, we will begin by considering a sample for which both the resistivities and the thicknesses of the Cu(Fe) and Cu films are equal; this is convenient for getting an intuitive understanding of what the results in Fig. 3 imply, and is realized approximately by sample 4 in Fig. 3. For such a sample it is easy to see that adding the resistances of the two layers in parallel yields $\Delta\rho(\text{bilayer}) = (\Delta\rho_b + \Delta\rho_t)/2$, where the subscripts refer to the bottom and top films. Since the top film is pure Cu, there is no Kondo contribution to its resistivity and $\Delta\rho_t = 0$. If we further *assume* that the Kondo contribution to the bottom layer [the Cu(Fe)], $\Delta\rho_b$, is unaffected by the presence of the Cu layer, this argument leads to the result that in this case the change in resistivity of the bilayer, $\Delta\rho$(bilayer), will be exactly half of that found for the bottom layer; i.e., the Cu(Fe) alone. This result is what one would expect intuitively, since adding the pure Cu layer effectively just dilutes the Fe, reducing its concentration in this case by a factor of 2. Indeed, based on this line of reasoning one would generally expect that the addition of a Cu layer on top of a layer of Cu(Fe) will always reduce the Kondo contribution to the resistivity, and this is in agreement with the qualitative trend seen in Fig. 3. As noted above, this argument *assumes* that $\Delta\rho_b$ is independent of the thickness of the top layer. We will now show that that assumption is *not* correct.

Sample 1 in Fig. 3 was just the bottom layer alone, while sample 4 had a Cu thickness which was approximately the same as that of the bottom layer, sample 1. We see that for sample 4 the change in resistivity is much greater that half that of the bottom layer alone. This implies that the presence of the top layer enhances the Kondo contribution to the bottom layer; i.e., that there is a sort of proximity effect.[22] This result would appear to be consistent with the simple picture of a Kondo screening cloud which is restricted by the dimensions of the sample. One might then imagine that the addition of the top layer gives the screening cloud room to "expand" thereby "undoing" the suppression of $\Delta\rho_K$ caused by the reduced thickness of the Cu(Fe) film.

Since all of the sample parameters such as the film thicknesses, etc., are known, it is straightforward to extract quantitatively the resistivity of the bottom layer alone from the data in Fig. 3. The results are shown in Fig. 4. As anticipated in the discussion given above, we see that $\Delta\rho_K$ of the bottom layer increases quite significantly as the thickness of the top layer is made larger. The largest enhancement of $\Delta\rho_K$ for this batch is seen to be ≈ 7; similar results have been found for other batches.[23]

IV. DISCUSSION

In summary, we have found that the Kondo effect in Cu(Fe) films is strongly suppressed when the thickness is reduced below about 1500 Å. At the same time, the Kondo temperature is unchanged. At first sight it is tempting to attribute this result to oxidation effects. Oxidation of the Fe could render it nonmagnetic, and since oxidation might be more severe in the thinner samples, it could make the Kondo contribution appear to be smaller in the thinner films. However, we have performed a number of tests which demonstrate that oxidation did not have a significant effect. First, we covered some of the films with a layer of photoresist a few minutes after removing them from the deposition chamber, in order to protect them from exposure to air. We also covered some samples with a protective layer of SiO *before* breaking vacuum. These two types of samples exhibited behavior which was the same as that of samples which were uncoated. Second, none of the samples (coated or un-

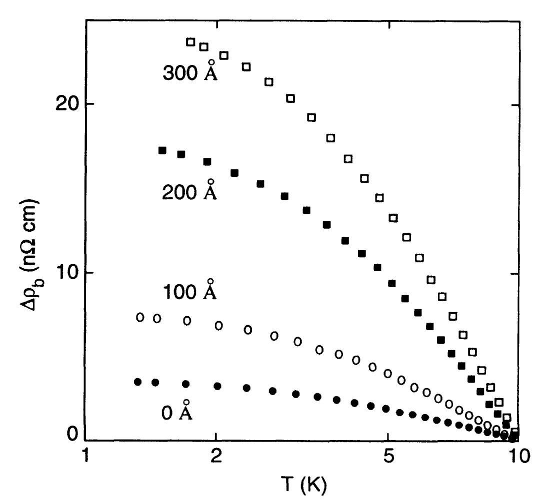

FIG. 4. Kondo contribution to the bottom layer [i.e., the Cu(Fe)] of the bilayer samples in Fig. 3; the thickness of the Cu layer is given in the figure. These results were derived from the data in Fig. 3, as discussed in the text.

coated) exhibited any significant "aging" after exposure to air. That is, the Kondo contribution remained the same even after exposure to air over the course of several months. Finally, the Kondo proximity effect was also observed in samples in which the Cu layer was deposited *first* with the Cu(Fe) on top, and the result was the same as that found with the Cu(Fe) layer on the bottom (as it was in the samples considered in Figs. 3 and 4). Since the proximity-effect experiments involved comparisons of samples which had Cu(Fe) films of the *same* thickness, it is very hard to see how oxidation effects could be responsible for the behavior we have observed. All of these results imply that oxidation was not a significant factor in our experiments.

In all of our discussions we have referred to the "thickness dependence" of the Kondo effect. However, as mentioned earlier, the low-temperature resistivity ρ_0 was also thickness dependent, so it is conceivable that it is the resistivity rather than the thickness that is the key variable. However, we believe that the Kondo behavior is indeed thickness dependent (it may also depend on ρ_0, but that is a separate issue which cannot be adequately addressed on the basis of the present results). First, ρ_0 varies relatively little (only a few percent) in the thickness range 1500–2000 Å, while in Fig. 1 we see that the Kondo contribution varies by about 30% over the same range. Second, in our previous studies of very narrow Au(Fe) strips we observed a similar size dependence of the Kondo behavior.[7] By the nature of the fabrication method used to make those samples, they all had the same ρ_0, implying again that the Kondo contribution does depend on sample size. Since we have found that the Kondo behavior of Au(Fe) and and Cu(Fe) films is very similar, we believe that this result also applies to Cu(Fe).

The results presented in this paper, taken together with those reported earlier for Au(Fe) and Cu(Cr), imply that explanations in terms of the Kondo length scales R_K (1) or R_K^* (2) are not correct. It therefore appears that some physics in addition to the Kondo effect must be playing an important role. Among the length scales (and associated processes) we have in mind are the phase-breaking length, the thermal length scale (which enters in the theory of electron-electron interactions), and the spin-orbit scattering length.[24] While there have been several theoretical discussions of how these length scales and processes might interact with the Kondo effect,[20] it does not appear that these theories, at least in their present forms, provide an explanation of our experiments.

The Kondo problem remains a problem.

ACKNOWLEDGMENTS

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[20] There have been a number of theoretical treatments of the

Kondo effect in systems of reduced dimensionality and in systems which contain disorder [see, for example, F. J. Ohkawa, H. Fukuyama, and K. Yosida, J. Phys. Soc. Jpn. **52**, 1701 (1983); K. Vladár and G. T. Zimányi, J. Phys. C **18**, 3739 (1985); S. Suga, H. Kasai, and A. Okiji, J. Phys. Soc. Jpn. **55**, 2515 (1986); **56**, 863 (1987); H. Fukuyama, *ibid.* **55**, 2118 (1986); Z. Tešanović, J. Phys. C **20**, L829 (1987)]. However, it does not appear that these theories are able to predict behavior like that observed in the present work.

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