Kondo behavior of multilayers: Local-moment physics near surfaces

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(Received 6 May 2000)

We report experimental results for the Kondo behavior of multilayers composed of Au and Au(Fe) films. The results show that the strength of the Kondo behavior produced by the local moments associated with the Fe depends on the distance of the Fe from the free surfaces of the multilayer. This dependence is in good quantitative agreement with recent theoretical predictions.

I. INTRODUCTION AND BACKGROUND

In its simplest version, the Kondo problem involves the effect of a magnetic impurity (i.e., a local magnetic moment) on the properties of a sea of conduction electrons.[1] Prototypical realizations include Fe in Au and Mn in Cu.[2] If the concentration of local moments (here Fe or Mn) is sufficiently small, the effect of interactions between the impurities is negligible and it is then appropriate to consider how a single local moment affects the properties of an electron gas which surrounds it. In many respects, this single-impurity Kondo effect is a ''solved'' problem. The development of this solution began with the work of Kondo, continued with the insights of many others including Anderson and Wilson, and in many ways continues to the present.[3,4,1] While exact results for all aspects of the Kondo problem are not available, a good qualitative and in most cases quantitative understanding has been attained. Even so, we believe that it is worthwhile to explore the Kondo effect in new situations. Indeed, recent studies of the Kondo behavior of thin metal films and narrow wires yielded results which were not explicable in terms of the accepted theories of the Kondo effect as applied to such systems. Specifically, experiments by our group[5–7] and by others[8] showed that the Kondo contribution to the resistivity, $\Delta\rho_K$, is a pronounced function of system size (i.e., film thickness or wire width).

It was initially proposed[6] that this size dependence might be associated with the Kondo screening length, which has been hypothesized to have the form $L_K \sim \hbar v_F / k_B T_K$, where T_K is the Kondo temperature.[9] This proposal could not be dismissed out of hand, since there are relatively few exact theoretical results for spatial correlations, etc., in which L_K might play a role. Nevertheless, a number of theoretical arguments[10,11] were quickly advanced which showed that the physics associated with L_K should not affect $\Delta\rho_K$. At the same time, further experiments[12] demonstrated that the size dependence of $\Delta\rho_K$ does not depend on T_K, and hence not on L_K (since L_K is a function of the Kondo temperature). It was thus clear that we must look elsewhere to understand the experimentally observed size dependence of $\Delta\rho_K$.

A promising direction in which to look was suggested by Újsághy *et al.*,[13] who focused attention on the local moments responsible for the Kondo effect. They showed that when a local moment is situated near a surface, multiple scattering of a conduction electron from the local moment and the surface can give rise to an effective uniaxial anisotropy at the local moment. This anisotropy may be described by the spin Hamiltonian

$$\mathcal{H} = K S_z^2, \tag{1}$$

where the z direction is normal to the surface and K is a parameter which depends on many things, including distance from the surface. This effect arises from the spin-orbit interaction between the conduction electrons and the host atoms, and is predicted to occur whenever the spin-orbit scattering strength changes abruptly. This may happen at a free surface or at an interface between two materials with different spin-orbit scattering strengths. Only the first case will be of interest in this paper.

The anisotropy (1) affects the Kondo behavior in the following way. The parameter K is a function of distance to the surface, and becomes larger as the local moment is brought closer to the surface. For a magnetic species such as Fe, which has an effective spin $S=2$, this anisotropy will split the magnetic sublevels into a singlet plus a series of doublets. The magnitude of this splitting is proportional to K. It is difficult for the theory to estimate this parameter with precision, as it depends sensitively on factors such as the density of states, the scattering amplitudes, etc. (although the best estimates are that the magnitude of K is compatible with the experiments[13]). However, the theory does predict that K must be positive, so that the singlet level will be always be the ground state, and the lowest excited states (a doublet) will be higher in energy by an amount K. If the thermal energy is much smaller than this, only the singlet ground state will be occupied and the ''local moment'' will be nonmagnetic. Since K is a function of distance from the surface, all impurities within some distance from the surface will be rendered nonmagnetic, and the effective concentration of impurities which are actually magnetic, and can thus contribute to the Kondo effect, will be a function of the thickness of the system. The length scale which governs this behavior thus arises from local moment physics and the parameter K.

This theoretical picture makes a number of predictions which were not addressed in the initial round of experiments. One concerns the choice of impurity. The first experiments[5–8] all involved Fe or Cr impurities, both of which have integer spin values. However, impurities such as Mn have half-integral spin, and in such cases the ground level will always be a doublet. Hence, the theory predicts[14] that the Kondo effect in a system such as Cu(Mn) will not

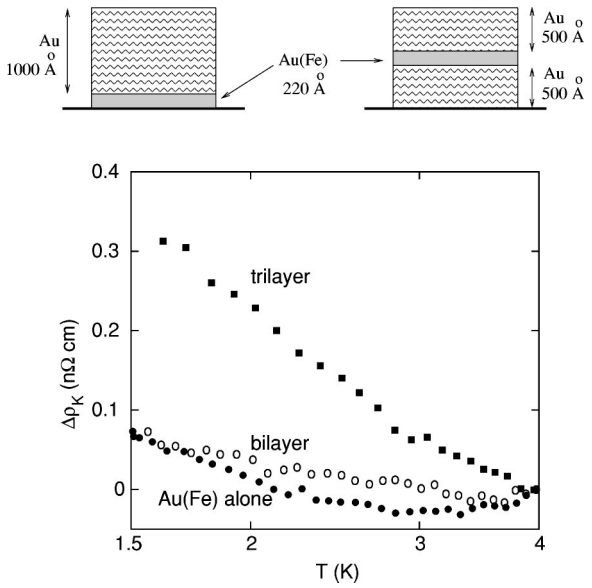

FIG. 1. Variation of the resistivity with temperature for the three samples from batch No. 1. Solid circles: a 220-Å-thick Au(Fe) film. Open circles: a bilayer sample consisting of a 220-Å-thick Au(Fe) film covered with 1000 Å of Au (see diagram at top left). Solid squares: a trilayer sample consisting of a 220-Å-thick Au(Fe) film sandwiched between two 500-Å-thick Au films (see diagram at top right).

vanish as the system size approaches zero. New experiments[15] have confirmed this prediction, although there are complications which seem to arise from the lifting of the degeneracy of the ground state.[16]

In this paper we consider a different way to test the theory. The parameter K and how it varies with distance from the surface, d, are central to the model of Újsághy *et al.* In experiments on homogeneous samples, the impurities are distributed throughout the sample, so one effectively measures an average over the sample volume and the variation of K with d is thus somewhat obscured. Moreover, most samples contain several surfaces, so it is necessary to consider a contribution to the anisotropy from each surface.[17] One would clearly like to probe the manner in which K varies with distance from the surface in a more direct manner.

In this paper we consider an experiment in which all of the impurities are located approximately the same distance from the surface and, moreover, this distance is under experimental control. The sample design is shown at the top of Fig. 1. These two composite samples both contain a Au(Fe) film, along with some thickness of pure Au. However, in one case the Au(Fe) is at one surface of the sample (here against the substrate), with all of the Au on the other side. In the other sample the Au(Fe) is sandwiched between two layers of Au. These Au(Fe) layers are deposited at the same time, so they have the same Fe concentration and thickness. The total thickness of Au in the two samples is also the same. The only difference is in the placement of the Au(Fe). Since the thickness of the Au(Fe) layer is small compared to the total thickness, one can (to a first approximation) imagine that in each of these multilayer samples all of the Fe is at a particular distance from the surface(s). In one case (the bilayer) the Fe is very close to one surface, while in the other (the trilayer) it is as far as possible from both surfaces. These samples enable us to probe directly how the local moment behavior of the Fe varies with distance from the surface(s).[18]

While the subject of this paper is the Kondo size dependence, we should note that experiments have also demonstrated that the Kondo behavior depends on the level of disorder (i.e., the elastic mean free path).[19,20] This has been addressed theoretically by Martin *et al.*,[21] who have explained the disorder dependence with a mechanism which is quite different from the one proposed by Újsághy *et al.* to account for size effects. The experiments described in this paper will be concerned only with the size dependence of the Kondo resistivity in clean (long-mean-free-path) samples.

II. SAMPLE PREPARATION

The samples were prepared by successive flash evaporation of layers of pure (99.999%) Au and Au(Fe). The source material for the Au(Fe) evaporations was prepared by evaporating a thin layer of Fe onto pure Au wire, as described elsewhere.[6,22] Au and Fe evaporate at essentially the same temperature, so flash evaporation of Fe-coated Au produces uniform alloy films.[23] This has been confirmed by previous experiments,[6,22,20] which show no evidence for clustering of the Fe.[24] In addition, estimates based on the known diffusion constant of Fe in Au, along with previous studies of bilayers,[6,19] have indicated that diffusion of the Fe into the pure Au layer is not significant.

The Fe concentration in the Au(Fe) layers was approximately 100 ppm. Previous work[25,6,26] has shown that for the temperatures of interest to us here (>1 K), this concentration is sufficiently small that interactions between Fe impurities have a negligible effect on the Kondo resistivity. The Kondo temperature of Au(Fe) is near 0.1 K, so the results below were all in the regime where the Kondo resistivity varies logarithmically with temperature.

In the next section we will give results from four separate sample batches. Each batch consisted of three individual samples, a bilayer, a trilayer, and a single layer of Au(Fe). All three of these individual samples contained a layer of Au(Fe), and all three of these layers were made in the *same* evaporation. Hence, in all three samples the Au(Fe) layers had the same thickness and Fe concentration. Examples of the structure of the bilayer and trilayer samples are shown at the top of Fig. 1. An arrangement of shutters in the deposition system enabled us to deposit the Au and Au(Fe) layers in succession while maintaining a vacuum of $\sim 5 \times 10^{-7}$ Torr.

The samples were patterned using photolithography and liftoff into a meander-type patter, with a width of 150 μm and a length of 60 cm. The resistance was measured with a four-probe dc method.

III. RESULTS

Results for the samples from batch No. 1 are shown in Fig. 1, which gives the change of resistivity with temperature for the bilayer, trilayer, and Au(Fe) layer. The structures of the bilayer and trilayer samples are shown at the top of the figure, and are also given in the table. The change of resistivity observed here is dominated by the Kondo contribution. Other effects, such as electron-electron interaction effects,

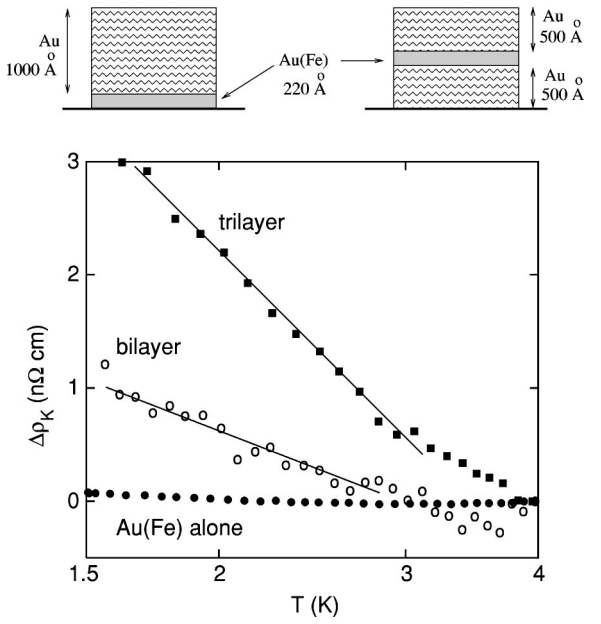

FIG. 2. Results for the behavior of the Au(Fe) layers *alone* (calculated as explained in the text) for the three samples from batch No. 1 and Fig. 1. Solid circles: a 220-Å-thick Au(Fe) film. Open circles: a bilayer sample consisting of a 220-Å-thick Au(Fe) film covered with 1000 Å of Au (see diagram at top left). Solid squares: a trilayer sample consisting of a 220-Å-thick Au(Fe) film sandwiched between two 500-Å-thick Au films (see diagram at top right).

are negligible on this scale.[27] Figure 1 shows the total resistivity of each sample. It is clear that the Kondo resistivity of the trilayer is much larger than that of the bilayer, as predicted by the theory of Ujsághy *et al.*

Another way to view these results is to consider the behavior of just the Au(Fe) layer which is contained in each sample. This can be inferred for the bilayer and trilayer samples if we assume that the resistivities of the different layers add as simple resistors in parallel. The resistivities and thicknesses of the Au and Au(Fe) layers are known from independent measurements,[28] and we also know that in isolation the resistivity of a Au layer is independent of temperature on this scale. Combining these observations with the measured behavior of the multilayer samples (as in Fig. 1), we can then calculate how the resistivity of the Au(Fe) layer must vary with temperature so as to yield the measured behavior.[19,20] The results of this analysis are shown in Fig. 2; here we show the inferred behavior of the Au(Fe) layer *alone* for the three samples from Fig. 1.

It is seen that placing the Au(Fe) layer in either a bilayer or a trilayer significantly enhances the Kondo behavior of the Au(Fe), relative to what is found in an isolated film, with the larger effect being found in the trilayer. This is in accord with the model of Ujsághy *et al.*, since in the trilayer geometry the Fe is located farthest from the surface(s). We should also note that our result for the relative behavior of the bilayer and trilayer does not depend on the ''resistors in parallel'' assumption used in connection with Fig. 2. Since the bilayer and trilayer contain the same total thicknesses of Au and Au(Fe), their different behaviors can only be due to the different location of the Fe within each sample.

As noted in the opening section, a quantitative comparison with the theory is difficult, since factors such as the anisotropy strength K cannot be calculated with much accuracy. However, by comparing the results for bilayer and trilayer samples, we can overcome some of these uncertainties by considering the ratio of the Kondo resistivities for the two geometries. For the samples considered in Fig. 2, the Kondo resistivity of the Au(Fe) in the trilayer is larger than that of the bilayer by a factor of 1.5 ± 0.1.

We have evaluated this ratio using the theory in Ref. 13. According to the theory, for an impurity near one surface K should vary as $K=\alpha/d$ where d is distance from the surface. For our thin film geometry we will consider two surfaces (and ignore the edges), so we have

$$K=\frac{\alpha}{d}+\frac{\alpha}{t-d}, \tag{2}$$

where t is the film thickness. The parameter α depends on many factors and we do not want to rely on a purely theoretical estimate of its value. An analysis of the measured size dependence of the Kondo resistivity in Au(Fe) in terms of the theory of Ujsághy *et al.* found $\alpha\approx60$ Å K.[13] We will use this value in our calculations below. However, we have also used other values, and have found that a 100% change in the (assumed) value of α causes only a 10% change in the trilayer/bilayer ratio of the Kondo resistivities. Hence, by focusing on this ratio we can, in large measure, isolate and test the prediction for the d dependence of the anisotropy in Eq. (2). Using this numerical value of α in Eq. (2), we have used the theoretical prediction for the Kondo resistivity $\Delta\rho_K(K_d)$ as a function of the anisotropy splitting.[13] This function $\Delta\rho_K(K_d)$ was integrated over the sample thickness

TABLE I. Parameters and results for several batches of multilayer Kondo samples. The sample structures are listed with the film farthest from the substrate (sub) given first. The layer denoted ''S'' is the substrate, and indicates how the bilayer samples were configured.

	Batch 1	Batch 2	Batch 3	Batch 4
Trilayer structure	Au/Au(Fe)/Au	Au/Au(Fe)/Au	Au/Au(Fe)/Au	Au/Au(Fe)/Au
Trilayer thicknesses (Å)	500/220/500	500/135/500	250/150/250	500/880/500
Bilayer structure	Au/Au(Fe)/S	Au/Au(Fe)/S	Au(Fe)/Au/S	Au/Au(Fe)/S
Bilayer thicknesses (Å)	1000/220	1000/135	150/500	1000/880
$\Delta\rho_K$(trilayer)/$\Delta\rho_K$(bilayer) measured	1.5 ± 0.1	1.8 ± 0.2	1.4 ± 0.2	1.06 ± 0.1
$\Delta\rho_K$(trilayer)/$\Delta\rho_K$(bilayer) theory	1.4	1.8	1.6	1.05

using the distribution of magnetic impurities as appropriate for the bilayer and trilayer cases. Further details of this evaluation are given in Ref. 26. For the samples considered in Fig. 2 we find a theoretical value of 1.4, in good agreement with the experimental value quoted above.

We have repeated this experiment with other similar multilayer geometries, and the results are listed in Table I. The variations we have studied include changes in the thicknesses of the Au and Au(Fe) layers. We have also examined bilayers in which the Au(Fe) was on the surface away from the substrate. As can be seen in the table, the ratio of the trilayer and bilayer Kondo resistivities was found to agree with the theory in all of the cases we have explored. For three of the sample batches listed in the table, the trilayer exhibited a significantly larger Kondo resistivity than the corresponding bilayer. For the other batch (No. 4) the tri- and bilayers exhibited the essentially the same Kondo resistivity (to within experimental error). This shows that the trilayer/bilayer difference is indeed a sensitive function of the layer thicknesses (i.e., the trilayer Kondo behavior is not always larger than that of the bilayer), as predicted by the theory.

IV. CONCLUSIONS

We have presented results for the Kondo behavior of multilayer samples composed of Au and Au(Fe). The multilayers have been designed to directly probe a key prediction of the theory of Újsághy *et al.* This prediction involves the importance of the location of the local moment, in our case Fe, relative to the surfaces of the sample. By comparing the behavior of two samples, a bilayer and a trilayer, we have shown that moving the Fe from near the surface to a location near the center of the sample significantly enhances the Kondo resistivity. The magnitude of this enhancement is in good agreement with the theory.

ACKNOWLEDGMENTS

We thank A. Zawadowski, P. Phillips, I. Martin, and P. F. Muzikar for many helpful discussions. This work was supported by NSF Grant No. DMR-9970708.

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[27] For the Au(Fe) sample in Fig. 1, electron-electron interaction effects contribute 30% to the change in resistivity in this temperature range, and only a few percent for the other two samples. Effects such as weak localization are even smaller.
[28] The Au layers had typical resistivities of $\sim 1-2\ \mu\Omega$ cm, while for the Au(Fe) layers it was $\sim 4\ \mu\Omega$ cm.