Magnetoresistance of small Kondo systems

N. Giordano
Department of Physics, Purdue University, West Lafayette, Indiana 47907-1396
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Results for the low-temperature magnetoresistance of thin Au films containing $\sim$30 ppm Fe are reported. As expected, we observe contributions from both the classical magnetoresistance and the local magnetic moments (i.e., the Fe). The local-moment magnetoresistance is a strong function of film thickness, and is suppressed as the thickness is reduced. This finding is compared with previous results for the Kondo effect in small systems, and with recent theoretical predictions.

I. INTRODUCTION

The Kondo effect concerns the behavior of a localized magnetic moment which interacts with a sea of conduction electrons.[1–3] A typical system of experimental interest is Au(Fe), i.e., Au containing a small amount of Fe, which is the material studied in the present work; other examples include Cu(Cr) and Cu(Fe). Understanding of the behavior of such a local moment, and its effects on the conduction electrons, has been a challenging problem, but there now exists an accurate and reasonably complete theoretical description for bulk systems. However, recent experiments by several groups have revealed unanticipated behavior in small, quasi-two- and quasi-one-dimensional Kondo systems.[4–10] A key result is that the Kondo contribution to the resistivity, $\Delta\rho_K$, becomes smaller as the size of the system is reduced. For example, in Au(Fe) films, $\Delta\rho_K$ is suppressed by a factor of $\sim$3 as the film thickness is reduced from 2000 to 700 Å.[5,10]

These observations have proved difficult to explain with the existing theory, although very recent calculations,[11] which will be described below, may provide a proper theoretical explanation. The situation is further confused by apparently contradictory experimental results from one group.[12] For these reasons, the Kondo behavior of small systems is currently far from understood. This is unfortunate, since the associated issues are of potentially wide interest for several reasons. First, the Kondo effect is a model many body problem, so any surprises or wrinkles may lead to new insights into many body physics and the electron gas. Second, an understanding of the behavior of isolated local moments would seem to be a prerequisite to formulating a theory of concentrated Kondo systems, which are currently of interest as models of highly correlated electronic materials. Third, the behavior of local magnetic moments in small systems is of general interest to the mesoscopic community, and may eventually be relevant to the function of small devices.

Most of the experimental studies of the Kondo behavior of small systems have involved the Kondo contribution to the resistivity, $\Delta\rho_K$, and its dependence on temperature and system size. In the present work we have instead investigated the magnetoresistance associated with the local moment. We find that this magnetoresistance is suppressed as the size of the system is reduced. Interestingly, the magnitude of this suppression is *different* from that exhibited by the temperature dependence of $\Delta\rho_K$ in zero field.

II. BACKGROUND

The Kondo experiments in our laboratory have recently been reviewed and compared with the work of other groups in Ref. 10. Here we will concentrate on those aspects which are directly relevant to our magnetoresistance measurements. A number of studies of films composed of Kondo alloys have found that the Kondo contribution to the resistivity is affected by the thickness of the sample. For example, work on Au(Fe) above 1 K, which is well above the Kondo temperature ($T_K \sim 0.3$ K), has shown that in this range, and in the absence of a magnetic field, $\Delta\rho_K = -B \log_{10} T$, where B is a positive factor. This behavior has, of course, been known for many years, and was first explained theoretically by Kondo;[1] we will refer to it as the "Kondo logarithm." Work by our group on Au(Fe) films has shown that the magnitude of the Kondo logarithm, i.e., the value of B, becomes smaller as the film is made thinner. Indeed, B appears to vanish in the limit that the film thickness $d \rightarrow 0$. Experiments with other sample geometries, and with other materials, appear to give a consistent picture; namely that the magnitude of the Kondo logarithm is suppressed and becomes vanishingly small, as one or more of the sample dimensions approaches zero.

Proposed explanations of these results initially focused on the Kondo "screening cloud." This term refers to a distribution of conduction electron spins centered on a local moment. A useful intuitive picture of the Kondo effect is that this cloud[13–15] screens the local moment, leading to a spin singlet state at temperatures well below T_K, a central feature of the Kondo effect. Some estimates are that this cloud is relatively large in size [a few μm in Au(Fe)], and it was suggested[5] that $\Delta\rho_K$ might be altered in a film whose thickness is comparable to or smaller than the size of the cloud. However, there are a number of theoretical arguments against this proposed explanation.[16,10] Moreover, subsequent experiments[8,10] have shown that the length scale associated with the suppression of the Kondo logarithm is not directly connected with T_K, as would be expected from the screening cloud picture. It thus appears that the suppression is due to some phenomena which acts to modify the Kondo behavior, but which itself is not contained in the Kondo physics. Just such a theory has recently been proposed,[11] according to

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which a local moment near a surface can experience an anisotropy energy, E_a, of the form

$$E_a = DS_z^2, \quad (1)$$

where S_z is the component of the local moment spin in the direction perpendicular to the surface, and D is a positive constant which depends on film thickness, the strength of the spin orbit scattering, and other parameters. The size of the anisotropy parameter D is hard to estimate with precision, but qualitatively this effect seems to account for the experiments. In particular, since, for integer spin [as appropriate for Fe in Au(Fe)], the $S_z=0$ level would be lowest in energy (D is predicted to always be positive), this anisotropy would lead to a singlet ground state, and hence a vanishing of the Kondo logarithm. The anisotropy is produced by electrons which scatter successively from the local moment and then the surface, so the value of D for a typical local moment becomes larger as the thickness is reduced, leading to a larger suppression in thinner films.

It is not (currently) possible to calculate D with precision, so it is difficult to test this theory critically through studies of the Kondo logarithm alone. In the present work we report measurements of both the Kondo logarithm and the magnetoresistance associated with the local moments. While the model in Ref. 11 has not yet been extended to deal with the magnetoresistance, it is our hope that these experiments will lead to a critical test of the theory, perhaps by providing a second, and independent estimate for factors such as D.

III. EXPERIMENTAL METHOD

The samples were Au(Fe) films deposited onto glass substrates by flash evaporation.[10] It turns out that Au and Fe evaporate at essentially the same temperature,[17] so this method yields homogeneous films (as confirmed in a number of previous experiments[17,10]). The evaporation source consisted of measured amounts of 0.0001% pure Au and Au-Fe wire (7-at. % Fe)[18] in a standard W boat. The Fe concentration in the films was approximately 30 ppm, which is well below the value at which interactions between the local moments become important in bulk Au(Fe),[19] so we believe that the results reported here are indicative of the dilute limit.

Photolithography and liftoff were used to produce "meander" patterns. The Au(Fe) films formed a serpentine pattern, as shown schematically in the inset of Fig. 1; the long sections were approximately 1 cm in length and 120 μm wide. Since the connections between these sections were fairly short ($\sim 100\ \mu$m), it makes sense to consider three different directions of a magnetic field: perpendicular to plane of the substrate, parallel to the plane of the substrate and perpendicular to the direction of the current, and parallel to the direction of the current. Results for all three of these cases will be compared.

The Au(Fe) samples considered below were all prepared in the same evaporation, and hence all had the same Fe concentration. Sample thickness was varied by positioning the substrates at different distances from, and angles with respect to, the evaporation source. The same method was used to make pure Au samples, whose behavior was compared to that of the Au(Fe). Thicknesses were estimated assuming Matthiessen's rule, which has previously[10] been found to yield reasonably accurate values (10% or better) for similarly prepared Au(Fe) films. Deviations from Matthiessen's rule would not affect the *relative* thicknesses, which is the most important aspect in comparing different samples.

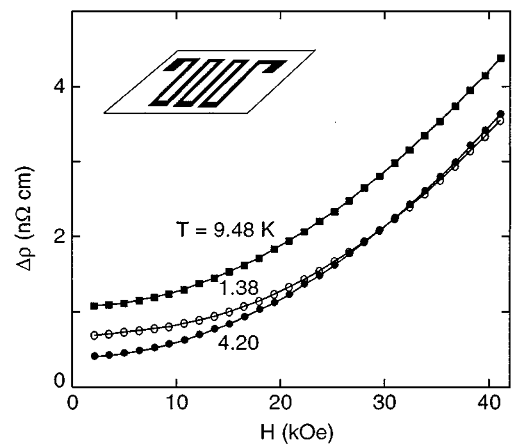

FIG. 1. Change of ρ with field for a 625 Å thick Au(Fe) film, at several temperatures, with the field perpendicular to the plane of the substrate. Note that the zero of the vertical scale is arbitrary; here we wish to emphasize the field dependence of ρ. For this sample $\rho \approx 0.58 \mu\Omega$ cm. The smooth curves are guides to the eye. The inset shows the (serpentine) sample geometry.

The measurements were performed in a ^{4}He cryostat of standard design. The sample resistances were measured with a four-lead, dc method.

IV. RESULTS

Results for the field dependence of the resistivity of a Au(Fe) film at several temperatures are shown in Fig. 1. Here the field was directed perpendicular to the plane of the substrate. The temperature dependence of ρ in zero field was similar to that reported previously. Above about 5 K, $\rho(H=0)$ increased with increasing T due to ordinary electron-phonon scattering, while at lower temperatures ρ became larger as T was reduced due to the Kondo logarithm. These two effects account for the variation of ρ seen in zero field in Fig. 1. The corresponding behavior of a pure Au sample is shown in Fig. 2. Here $\rho(H=0)$ was essentially independent of temperature below about 5 K, as there were no local moments present, and thus there was no Kondo logarithm in this case.

Both Au(Fe) and Au are seen to exhibit a positive, and approximately parabolic ($\Delta\rho \sim H^2$) magnetoresistance. The dominant effect here is just the classical magnetoresistance,[20] and its magnitude was consistent with our estimates for the elastic mean free path.[21] In Au(Fe) there is also a contribution to the magnetoresistance from the local moments. This can be extracted most simply by considering the temperature dependence of $\Delta\rho(H)$. At high temperatures we would expect a magnetic field to have very little effect on the alignment of the local moments, and hence that there would be no contribution from the Fe to the *change* in ρ with field. At sufficiently high temperatures the magnetoresistance should arise solely from the classical magnetoresistance, and thus be independent of temperature (to the extent that the total mean

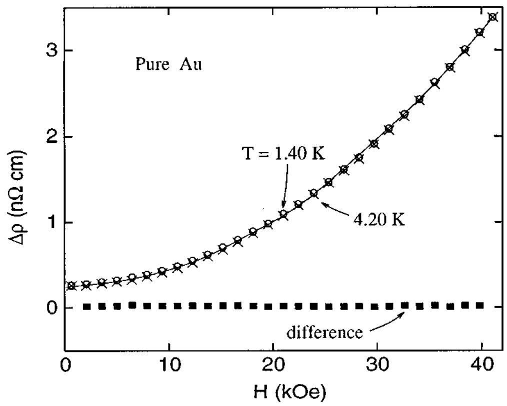

FIG. 2. Change of resistivity with field for a 610 Å thick pure Au sample, as a function of field applied perpendicular to the plane of the substrate. The filled squares show the difference between the results at 4.20 and 1.40 K. For this sample $\rho \approx 0.45$ $\mu\Omega$ cm. The smooth curve is a guide to the eye.

free path, from both elastic and inelastic processes, is temperature independent, which was the case in our experiments). At the fields employed in the present work, there was no discernible change in the field dependence of ρ above about 6 K. We therefore believe that the magnetoresistance at 9.48 K in Fig. 1 was a measure of just the classical magnetoresistance.

At sufficiently low temperatures, or sufficiently high fields, or both, we expect a field to align the local moments, thereby leading to a contribution to the magnetoresistance. In low to moderate fields the alignment will be only partial, and we expect this to yield a contribution to the magnetoresistance which is temperature dependent. It can be extracted from the results in Fig. 1 by subtracting the results at high temperatures, which contain only the classical magnetoresistance, from the behavior at a low temperature, which contains both the classical and local moment magnetoresistances. The results of such subtractions are shown in Figs. 2 and 3. For these samples, and at these temperatures, the classical magnetoresistance should be temperature independent, so our subtraction procedure should yield the magnetoresistance due to the local moment (in addition to a field independent offset due to electron-phonon scattering). The difference curve in Fig. 2 is seen to be field independent, and essentially zero. This implies that for pure Au the classical contribution is the only source of magnetoresistance, as expected.[22,23] However, we see from Fig. 3 that there was a substantial contribution from the local moments in Au(Fe), again, as expected. This contribution was much larger at 1.4 K than at 4.2 K, because for a given field the alignment of the Fe moments was larger at the lower temperature.

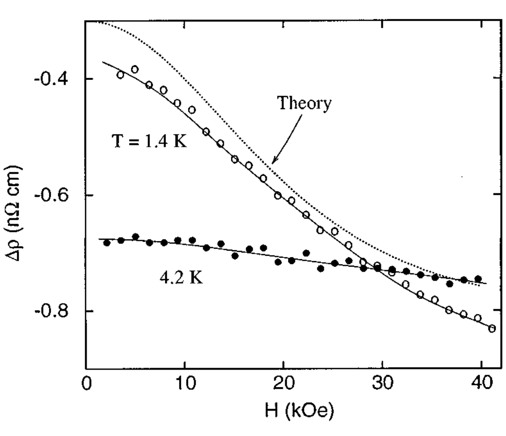

FIG. 3. Difference between $\rho(H)$ at a low temperature and $\rho(H)$ at 9.48 K, for the Au(Fe) sample considered in Fig. 1. The dotted curve, labeled "theory," is drawn proportional to the calculated value of $-\langle S_z \rangle^2$ at 1.4 K, as described in the text. The solid curves are guides to the eye.

The magnetoresistance associated with local moments in a metal has been much studied, both theoretically and experimentally (see, for example, Refs. 24 and 25 and references therein). It is predicted that at temperatures well above T_K, the limit appropriate for our experiments, this magnetoresistance is approximately proportional to $-\langle S_z \rangle^2$; i.e., to the (negative) of the square of the thermal average of the local moment. This relationship is only approximate, as there are also a number of higher order terms associated with the Kondo effect, etc. Under the conditions of our experiment, the $-\langle S_z \rangle^2$ term should be dominant, so we will neglect these terms (they are also hard to estimate with precision). In any case, our main reason for mentioning this prediction is to compare it with the results in Fig. 3. The dotted curve in that figure is drawn proportional to $-\langle S_z \rangle^2$, with $\langle S_z \rangle$ estimated using the Brillouin function with $g=2$ at 1.4 K. The theoretically expected magnitude of this magnetoresistance cannot be predicted with certainty, since the relevant exchange constant, and other required parameters are not accurately known. We did *not* attempt to fit to the measured magnetoresistance, although we believe that with only modest adjustments of the parameters such a fit could be made very accurate. Rather, our point is that this functional form has a *shape* which is *quite* similar to the experimental results, and this shape (i.e., the field dependence) has been obtained without any parameter adjustments. It thus appears that our results are in at least qualitative agreement with the form predicted by the theory, and is thus also consistent with previous experiments in bulk systems.[26]

As mentioned in a previous section, a key result of recent work on Au(Fe) films is that the magnitude of the Kondo logarithm, B, is a function of film thickness, d. For d less than about 2000 Å, B becomes smaller as the thickness is reduced. Unfortunately it was not possible in the present experiments to measure the magnetoresistance for an extremely wide range of d, for the following reasons. For thick films the elastic mean free path became quite long, making the classical magnetoresistance relatively large. This made it hard to extract the much smaller magnetoresistance due solely to the local moments. For thin films the local moment magnetoresistance became very small, and could not be resolved. For these reasons we will only present results for films in the range 300 Å$<d<$650 Å. Even with this restriction, some interesting trends will be apparent.

Figure 4(a) shows the variation of the local moment magnetoresistance with thickness. In this figure $\Delta\rho_H$ is the difference in the resistivities at $H=0$ and $H=40$ kOe, both measured at 1.4 K, with the classical contribution subtracted as in Fig. 3. We see that $\Delta\rho_H$ was a strong function of thick-

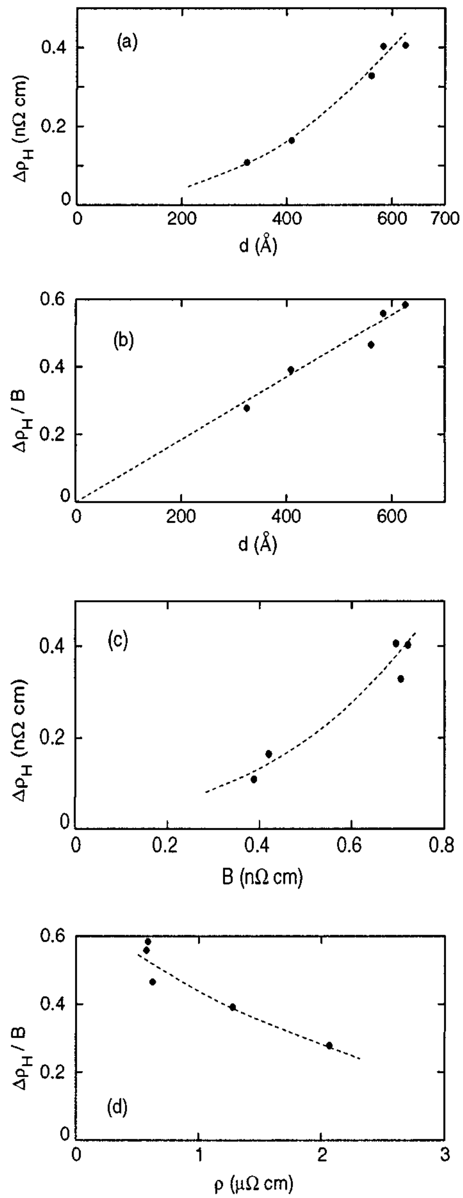

FIG. 4. (a) Thickness dependence of the magnetoresistance of a series of Au(Fe) samples at 1.4 K. Here $\Delta \rho_H$ is the difference in the resistivities at $H=0$ and $H=40$ kOe, with the classical contribution removed. (b) Ratio $\Delta \rho_H / B$ as a function of thickness. (c) $\Delta \rho_H$ as a function of B. (d) $\Delta \rho_H / B$ as a function of the residual resistivity, ρ_0. The dashed curves are guides to the eye; the one in (b) is a straight line drawn through the origin.

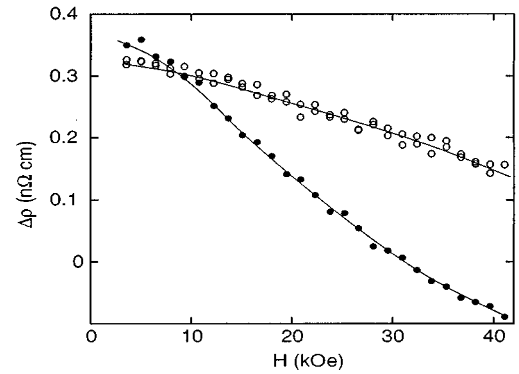

FIG. 5. $\Delta \rho$ as a function of field for a 625 Å (filled circles) and a 410 Å sample (open circles) of Au(Fe). The field was applied perpendicular to the plane of the substrate, the temperature was 1.4 K, and the classical magnetoresistance (which was measured at 9.5 K) has been subtracted. The solid curves are guides to the eye.

ness, and appeared to vanish in the limit $d \rightarrow 0$. This suppression is extremely reminiscent of the suppression of the Kondo logarithm mentioned above. It is therefore interesting to ask if $\Delta \rho_H$ and B are similar functions of thickness. Figure 4(b) shows the *ratio* $\Delta \rho_H / B$ as a function of d, and we see that it also becomes smaller as d is reduced. This means that $\Delta \rho_H$ is suppressed *more rapidly* than B. Whatever is suppressing the Kondo effect impacts on the field dependence of ρ more strongly than on the temperature dependence. This is also evident from Fig. 4(c), which shows the relationship between $\Delta \rho_H$ and B.

To this point we have been referring only to the thickness dependence of quantities such as B and $\Delta \rho_H$. However, as has been shown recently,[9,10] B is a function of both d *and* the degree of disorder. It is therefore worthwhile to ask if $\Delta \rho_H$ also depends on the amount of disorder. A convenient measure of the level of disorder is the elastic mean free path, or equivalently, the residual (i.e., low-temperature) resistivity, ρ_0. In Fig. 4(d) we show $\Delta \rho_H$ as a function of ρ_0. Unfortunately, we were not able to study the effects of d and ρ_0 separately, so we cannot say which effect, thickness or disorder, is more important. In any event, it is clear that the magnetoresistance is suppressed more strongly than the Kondo logarithm, B.

While the theory in Ref. 11 can account, at least semiqualitatively, for the suppression of B, it has not yet been extended to deal with the magnetoresistance. However, *if* the effect of the surface is to *only* introduce a DS_z^2 anisotropy, one would expect there to be an effect on the magnetoresistance. Qualitatively, this anisotropy will split the (otherwise degenerate) levels, and thus require a larger field to produce saturation of $\Delta \rho(H)$. This would lead to a change in the field scale, i.e., a change in the *shape* of $\Delta \rho(H)$, as discussed in connection with Fig. 3. With this in mind, results for two samples are compared in Fig. 5. For the thicker sample an inflection is evident at approximately 25 kOe, as $\Delta \rho$ began to saturate above that field. While the relative uncertainties are greater for the thinner sample, there does not appear to be an inflection at a similar field. Hence, we *tentatively* conclude that $\Delta \rho$ saturates at a higher field for the thinner sample, which is consistent with the proposition that there is a larger anisotropy in that case.

According to the theory, the anisotropy is normal to the surface, so one might also expect the behavior to depend on the field direction. Figure 6 shows the behavior of one sample for all three field directions: perpendicular to the substrate, parallel to the substrate and perpendicular to the current, and parallel to the current. To within the uncertainties, there does not appear to be any dependence on the direction of H. Similar results were found for all samples. At first sight this seems hard to reconcile with (1), but we can think of several ways around this difficulty. First, the film surfaces are not perfectly smooth, and this could make the anisotropy direction vary in a complicated manner with position within

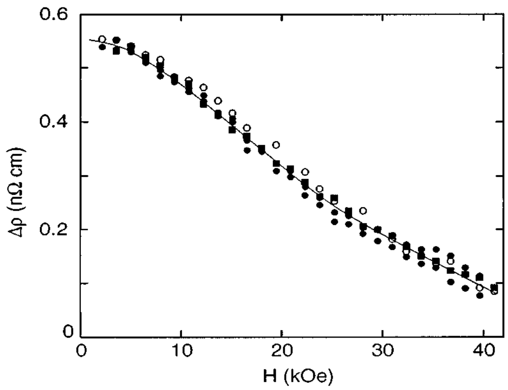

FIG. 6. Change of resistivity with field of a 625 Å thick Au(Fe) sample at 1.4 K, for different field directions. Filled squares: H perpendicular to the substrate and perpendicular to the current. Filled circles: H parallel to the substrate and perpendicular to the current. Open circles: H parallel to the current. The solid curve is a guide to the eye.

the sample. While the long elastic mean free paths (as compared to the film thicknesses) seem to imply that the surfaces are fairly flat, further work is needed to investigate the surface roughness, and its effect on D.[27] Second, the effect of the surface on the magnetoresistance may not be describable simply in terms of a DS_z^2 anisotropy. More work will be needed to determine if either of these speculations is appropriate.

V. SUMMARY AND CONCLUSIONS

We have studied the contribution of the local moments to the magnetoresistance of thin Au(Fe) films, and find that this magnetoresistance is suppressed as the films are made thinner. This suppression is qualitatively similar to, but more pronounced than, the suppression of the Kondo logarithm. Certain aspects of our results are consistent with recent theoretical predictions concerning an effective anisotropy field for local moments positioned near the surface.[11] An appealing aspect of this theory is that the length scale associated with the suppression is determined not by the Kondo physics, but by the elastic mean free path, and this seems to be demanded by the experiments.[10]

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[21] For the Au(Fe) sample considered in Fig. 1, the elastic mean free path was $\ell \sim 1000$ Å, as estimated from the approximate $\rho\ell$ product for Au, $6\times10^{-12}\Omega$ cm (Ref. 20). While, as can be seen from Fig. 6, the local moment magnetoresistance was independent of field direction, the "classical" magnetoresistance was not. This background magnetoresistance observed in pure Au, and in Au(Fe) at high temperatures, was largest with the field perpendicular to the plane of the substrate, and smallest with the field parallel to the current.
[22] The samples considered in this paper all had very low sheet resistances, so the magnetoresistance associated with weak localization (see Ref. 23) was negligible on this scale.
[23] See, for example, B. L. Altshuler, A. G. Aronov, and D. E. Khmel'nitskii, and A. I. Larkin, in *Quantum Theory of Solids*, edited by I. M. Lifshitz (Mir Publishers, Moscow, 1982), p. 130; G. Bergmann, Phys. Rep. **107**, 1 (1984); P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
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[26] The magnitude of the magnetoresistance we observe is also consistent with an extrapolation of the results of H. Rohrer [Phys. Rev. **174**, 583 (1968)] to our concentration.
[27] Scanning tunneling microscope examination of somewhat thinner (~ 125 Å) Au films deposited in a similar manner revealed a surface roughness of order $\sim 10-20$ Å, but the topology of thicker films, and precisely how this would affect D, is not clear to us at present.