Magnetic Screening of Fe Impurities in Mg

Gerd Bergmann
Department of Physics, University of Southern California, Los Angeles, California 90089
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The magnetic and the spin-orbit scattering by magnetic Fe impurities in Mg films are measured by means of weak localization and a Kondo-type behavior is found. The measurements suggest a magnetic screening of the Fe which depends on the distance between the Fe atoms but not on the film thickness of Mg. The spin-orbit scattering rate is an order of magnitude larger than the effective magnetic scattering rate.

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The properties of magnetic impurities in metal hosts and their anomalous behavior, the Kondo effect, have attracted considerable interest over many decades and in recent years this interest is again growing. Although the Kondo problem has been solved for a single impurity (see, for example, Andrei, Furuya, and Lowenstein[1]) the "heavy fermion" systems, often viewed as "periodic Kondo lattice," raise a number of new questions about magnetic screening of the magnetic impurities. For a single Kondo impurity in a pure bulk metal it is believed that the conduction electrons screen the moment of the magnetic impurity (at temperature $T=0$) within a sphere of radius $\eta_K=v_F\tau_K$, where τ_K is a time characteristic for the Kondo temperature T_K: $\tau_K=\hbar/2\pi k_B T_K$. This polarized electron cloud is superimposed on a large oscillation of the polarization, the Rudermann-Kittel-Kasuya-Yosida oscillation with the period of the Fermi wavelength. The critical question in the case of concentrated Kondo systems is, How is the screening modified if the magnetic atoms are closer together than the Kondo screening length η_K? There are different theoretical approaches to this question. On one hand there is a large effort to try to understand the properties of two Kondo impurities as a function of their separation. On the other hand, one tries to solve the other extreme, the periodic Kondo lattice. In this paper I report an experimental investigation of this question using two different approaches. (i) I put the magnetic impurity into a thin film with a thickness much smaller than the Kondo length and change its thickness. If the magnetic impurity needs the whole sphere with radius η_K for its screening then the screening will be incomplete for the thin film and will improve with increasing film thickness. (ii) I condense a two-dimensional arrangement of dilute magnetic impurities in a constant depth of a thin film and investigate their magnetic screening as a function of their separation or (two-dimensional) concentration.

The experimental method of investigating the magnetic impurities is the measurement of the magnetic scattering time by its influence on weak localization.[2-5] Magnetoresistance measurements of disordered thin films correspond to a time-of-flight experiment with conduction electrons[6] and yield their inelastic lifetime and spin-orbit and magnetic scattering times. (The magnetic scattering increases the width of the magnetoresistance curves because the magnetic scattering destroys the coherence of the conduction electrons roughly after the magnetic scattering time τ_s.) The magnetoresistance in the presence of magnetic impurities has been studied only in a few examples,[7,8] but it represents a unique method of exploring magnetic impurities in thin films. Therefore, it is now possible to investigate a number of problems concerning magnetic impurities which have heretofore eluded solution.

In this paper I explore the properties of Fe impurities in Mg. First I investigate a sandwich consisting of Mg/Fe/Mg/Au (see Table I). In this experiment I cover a Mg film ($d=9.6$ nm, $R=99\ \Omega$) with 0.003 atomic layer of Fe and 4.6 layers of Mg. In a last step I condense 0.37 atomic layer of Au. After each evaporation the magnetoresistance of the resulting film is measured in the field range -7 T to $+7$ T. In Fig. 1 the results of this experiment are plotted. The upper curve shows the experimental points of a pure Mg film at 4.5 K. The left ordinate gives the change of the resistance in ohms. Since the theory yields the change of the conductance in units of $L_{00}=e^2/2\pi^2\hbar$ I have added on the right-hand side a conductance scale in units of L_{00}. The points represent the experimental results. The second curve is obtained after a superposition of about 0.003 layer of Fe and 4.6 layers of Mg. The small coverage of the Fe could not be measured directly. To achieve this coverage a Fe wire is heated so that evaporation rates between 1 and 0.1 atomic layer (atola) per minute are obtained. The logarithm of the evaporation rate as a function of the heating power of the Fe wire is plotted and extrapolated to a rate of 0.03 atola/min. With the resulting power, the Fe is evaporated for 6 sec which corresponds roughly to a coverage of 0.003 atola Fe. This Fe coverage yields a considerable broadening of the magnetoresistance curves. Therefore, the scale of the magnetic is changed. In the next step the Fe impurities are

TABLE I. The resistance, thickness, $H_{s.o.}$, and H_s of the sandwich consisting of 34 atola Mg, 0.003 atola Fe, 4.6 atola Mg, and 0.37 atola Au.

Metal	R_n (Ω)	d (nm)	$H_{s.o.}$ (T)	H_s (T)
Mg	99	9.6	0.005	0.000
Fe	98.4	0.0007	0.0245	0.007
Mg	82.8	1.32	0.023	0.0014
Au	81.0	0.10	0.52	0.0014

covered by 4.6 atola of Mg. This transforms the Fe surface impurity into a bulk Fe impurity. The magnetoresistance curve shows an interesting substructure. The first question that arises is whether this minimum is caused by weak localization. The Fe might introduce some spin-orbit scattering which could yield the substructure. However, since the Fe is a rather light atom one would expect a relatively small spin-orbit scattering due to the Fe. For a light host such as Mg this question can be investigated in a rather elegant way. One covers the sandwich with some additional Au. This introduces a strong spin-orbit scattering and the contribution of weak localization changes to weak antilocalization. This means that the negative magnetoresistance changes to a positive one. Other anomalies of the resistance should change much less or not at all with the Au coverage.

The lower curve in Fig. 1 is obtained by covering the sandwich with 0.37 layer of Au. Of course, the magnetoresistance becomes positive because the film is now in the strong spin-orbit scattering region. There is no indication that the substructure still exists. However, quite surprisingly the magnetoresistance curve becomes narrow again so that we have to use the old scale of the magnetic field. This proves that the broad magnetoresistance curve of the Mg/Fe/Mg sandwich with its minimum at zero field is caused by a finite spin-orbit scattering. Now it is possible to evaluate the magnetoresistance curves.

I evaluate the experimental curves with the theory of Hikami, Larkin, and Nagaoka[9] (using the formula as given in Bergmann[10]). It requires essentially the adjustment of three characteristic fields H_i, $H_{s.o.}$, and H_s, which correspond to the inelastic lifetime τ_i, the spin-orbit scattering time $\tau_{s.o.}$, and the magnetic scattering time τ_s. The relation between the time τ_n and the field H_n is given by

$$H_n\tau_n=\hbar e\rho N/4, \quad (1)$$

where ρ is the resistivity of the film and N is the density of states at the Fermi level (for both spins). The evaluation is performed in the following steps:

(i) The single Mg film. Its properties in the quench-condensed state have been intensively studied

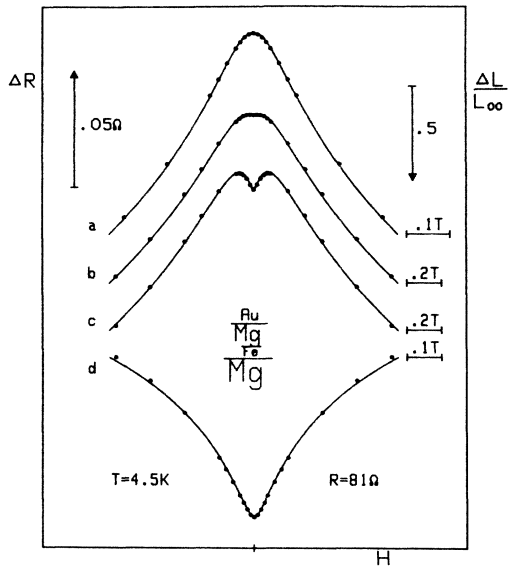

FIG. 1. The magnetoresistance of a Mg sandwich with Fe impurities. The top curve shows the data of the pure Mg film. The second curve is obtained after a superposition with about 0.003 layer of Fe and 4.6 layers of Mg. Finally 0.37 layer of Au are condensed. The points represent the experimental data. The full curves are calculated with the theory and yield the strength of the spin-orbit and the magnetic scattering.

and its characteristic fields are well known.[10] The theoretical curve is calculated with $H_i=0.0095$ T, $H_{s.o.}=0.005$ T, and $H_s=0$.

(ii) The Mg/Fe/Mg sandwich. The structure of the magnetoresistance curve allows us to determine $H_{s.o.}$ and H_s independently for known H_i. The inelastic field H_i is essentially the same as for the pure Mg film. The plotted full curve is obtained with $H_{s.o.}=0.023$ T and $H_s=0.0014$ T. The pure Mg film has a spin-orbit scattering field of 0.005 T which can be subtracted. As a result the bulk Fe impurities introduce a spin-orbit scattering field $H_{s.o.}=0.018$ T which is 12 times as large as H_s.

(iii) The Mg/Fe/Mg/Au sandwich. This sandwich is in the large spin-orbit scattering limit; the exact value of $H_{s.o.}$ is not required but from the magnetoresistance curves at 14 and 20 K we can determine $H_{s.o.}=0.52$ T. Taking $H_i=0.0093$ T I determine $H_s=0.0014$ T without any ambiguity. This value is in excellent agreement with the evaluation of the Mg/Fe/Mg sandwich.

(iv) The Mg/Fe sandwich. The magnetoresistance curve of this sandwich has a very weak minimum at $H=0$. This means that the ratio of $H_{s.o.}$ to the phase-breaking field H_ϕ (which is a sum of H_i and H_s) is reduced compared with the sandwich Mg/Fe/Mg. Fortunately the very weak minimum at zero field allows a (rough) determination of H_s and $H_{s.o.}$. The full curve

is obtained with $H_s = 0.007$ T and $H_{s.o.} = 0.0245$ T.

The weak-localization measurements yield the following results for 0.003 atola of Fe impurities on the surface and in the Mg sandwich. (i) Bulk Fe impurities in Mg: The spin-orbit scattering rate is a factor of $H_{s.o.}/H_s \approx 12$ larger than the magnetic scattering rate. (ii) Bulk to surface Fe impurities: The magnetic scattering rate of Fe on the surface of Mg is a factor of 4 larger than for Fe in bulk Mg. (To obtain this factor one has to form the ratio of the two H_s values divided by the ratio of the corresponding resistances.)

One might suggest that the change of magnetic scattering rate as we go from the surface of Mg to the bulk is just a crystal-field effect. This will be disproven below by a change of the Fe concentration in the middle layer. In addition I performed the first measurements of the temperature dependence of $1/\tau_s$ which will be published elsewhere. The magnetic scattering rate increases strongly between 4.5 and 20 K. I suppose that the Fe on the surface of Mg has a different Kondo temperature than in the bulk. The T_K of the latter should be much larger. Therefore, the magnetic screening is considerably more complete in the bulk.

The strong reduction of the magnetic scattering of the Fe atoms going from the surface to the bulk occurs only for very dilute Fe coverages. For Fe coverages such as 0.01 atola Fe the reduction is only a factor of 2. In the following we investigate Mg/Fe/Mg sandwiches with different Fe concentrations between the two Mg layers whose thickness is kept constant. We obtain the following results: (i) The spin-orbit scattering rate (or the corresponding field $H_{s.o.}$) increases linearly with the Fe coverage (nicely confirming the calibration of the Fe coverage). (ii) The magnetic scattering rate (i.e., H_s) increases within the experimental error as the square of the Fe coverage. The latter is demonstrated in Fig. 2 where the ratio of the magnetic scattering to the spin-orbit scattering $r = (1/\tau_s)/(1/\tau_{s.o.}) = H_s/H_{s.o.}$ is plotted versus the Fe coverage (in atola). This plot shows the ratio of the two cross sections of a Fe atom. Since the experimental points lie within the experimental error on a straight line through the origin and since $H_{s.o.}$ is proportional to the Fe coverage, one finds that the Fe atoms show less and less magnetic scattering when they are diluted. On the other hand, the linear increase of $1/\tau_{s.o.}$ with Fe concentration suggests that the electronic properties of the Fe ions themselves are unchanged by the increasing coverage.

This experimental result suggests that the conduction electrons need a certain volume to screen the magnetic moment of the Fe atoms. When the Fe atoms have only a finite separation then the screening volume is restricted.

However, the volume that is available for the screening is in any case restricted since the upper Mg film has only a thickness of about 5 atola. Therefore the screening can only take place roughly within half a sphere. By superimposing additional Mg we can complete this sphere. If the magnetic scattering cross section per Fe atom does not change with increasing Mg thickness then we expect that d_{Mg}/τ_s remains constant (d_{Mg} is the Mg thickness) since the chance of hitting the impurity decreases as $1/d_{Mg}$. In Fig. 3 I have plotted $H_s^* = H_s \times (100\ \Omega)/R = (\hbar eN/4)\, d_{Mg}/\tau_s$ as a function of the thickness of the second Mg layer. For vanishing Mg thickness we find, of course, a very large value because here the Fe lies on the surface of the Mg. It drops already for a small Mg thickness and beyond 3 atola Mg H_s^* changes only slightly. The experimental increase of the screening volume from half a sphere to a full sphere has very little influence on the magnetic scattering.

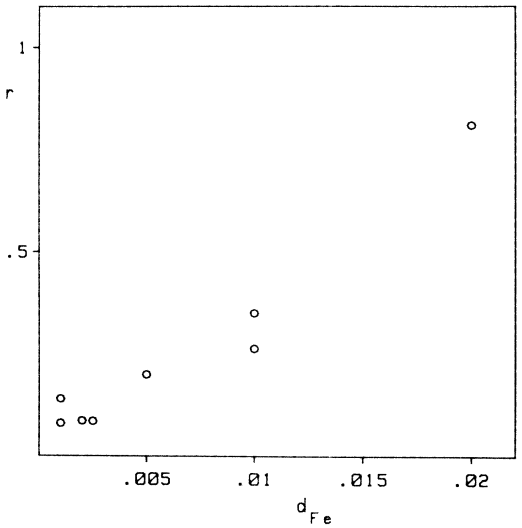

FIG. 2. The ratio of the magnetic scattering to the spin-orbit scattering $r = (1/\tau_s)/(1/\tau_{s.o.}) = H_s/H_{s.o.}$ for the sandwich Mg/Fe/Mg is plotted vs the Fe coverage in atola. This plot shows the ratio of the two cross sections of a Fe atom. Since the spin-orbit scattering rate is proportional to the Fe coverage, the magnetic rate is proportional to the square of the Fe concentration.

This last experiment suggests that the geometrical size and the shape of the host have no or little effect on the magnetic screening. It is likely that the screening is more concentrated in the smaller-volume samples.

Alternative explanations of the observed behavior appear to be less consistent. These are discussed in turn:

(a) The change of the magnetic scattering time might be due to crystal-field effects which are stronger and less isotropic at the surface. This supposition is disproven by the fact that the change of $1/\tau_s$ depends

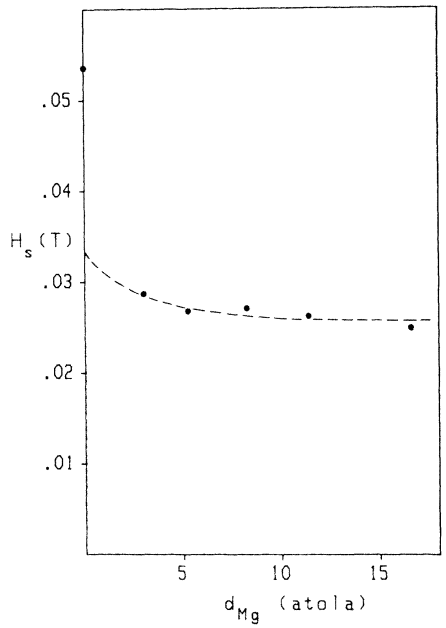

FIG. 3. The normalized magnetic scattering of the Fe impurities in the sandwich Mg/Fe/Mg for different thicknesses of the second Mg film. For constant cross section of the magnetic scattering $H_s^* \propto d_{Mg}/\tau_s$ is constant. The plot demonstrates that already a small coverage with Mg is sufficient to change the strength of the magnetic scattering dramatically. Further coverage with Mg has only a negligible effect showing that the magnetic screening does not critically depend on the screening volume.

on the (small) concentration of the Fe atoms.

(b) The Fe atoms perhaps form clusters with a larger magnetic moment. This possibility is excluded by the quenched condensation. Even at much higher concentration between 5% and 10% coverage (of Fe on Pb) I found that the number of Fe singles and pairs corresponded to the statistical expectation.

(c) The interaction between the (statistical) Fe moments may lock them together and they still form a magnetic cluster. This would be a surprisingly strong interaction which would contradict our former observation that Fe coverages of 5% on different surfaces show free-moment behavior.

Therefore my measurements allow the following conclusions: (i) In passing from the surface to the bulk impurity we find a strong reduction of the magnetic scattering which we attribute to the screening of the magnetic impurities. (ii) The screening does not depend on the volume of the host even when the screening volume is restricted. (iii) The finite distance between the magnetic impurities reduces the screening. (iv) The spin-orbit scattering is much larger than the effective magnetic scattering.

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