Quenching of Interacting Moments and Anomalous Fermi-Liquid Behavior in Disordered Kondo Alloys at Low Temperatures

R. P. Peters and G. Bergmann
Physics Department, University of Southern California, Los Angeles, California 90089

and

R. M. Mueller
Institut für Festkörperforschung der Kernforschungsanlage, D-5170 Jülich, West Germany
(Received 21 September 1987)

The magnetic scattering rate of Kondo alloys with a finite concentration of magnetic impurities is measured by means of weak localization. The three investigated systems—Fe in Mg and Co on the surface of Mg and Cu—show magnetic interactions at liquid-helium temperatures. At much lower temperatures the magnetic scattering rate decreases drastically with a $T^{1/2}$ temperature dependence in the experimental range. This suggests a cooperative effect between the different impurities which appears to suppress the magnetic moments, although the condition for a Landau Fermi liquid is not fulfilled.

PACS numbers: 75.20.Hr, 71.55.Jv, 72.15.Qm

Magnetic impurities in metals interact strongly with the conduction electrons.[1] One of the interesting questions is their low-temperature behavior. Do the magnetic moments remain active at low temperature or do they lose their magnetic character? The latter case is denoted as the "Fermi-liquid" behavior of the system. Two extreme examples are well known: the single Kondo impurity and the periodic Anderson problem, also called the heavy-fermion system. In our present investigation we are interested in dilute Kondo systems. The single Kondo impurity forms at low temperature a singlet state with the conduction electrons. At zero temperature the magnetic moment is suppressed and neither yields spin-flip scattering nor does it act as a random field. At finite temperature $T \ll T_K$, the Fermi-liquid theory[2] yields a T^2 law for the magnetic scattering rate. Recent experiments[3] on thin disordered Mg/Fe/Mg sandwiches with about 300 ppm Fe showed in the temperature range between 4.5 and 20 K qualitatively a T^2 behavior on top of a residual scattering. At He temperatures (which is $\frac{1}{10}$ of the Kondo temperature of[4] about 45 K) the magnetic scattering appeared to saturate at a finite rate. It could be shown that this saturation is due to interaction between the impurities.[5]

In this Letter we try to answer the question, What is the ground state of interacting impurities? Do they form a spin-glass or do they approach a Fermi-liquid behavior? We extend the investigation to temperatures of about 80 mK, far below the Kondo temperature.

A rather dramatic indication for the suppression of the magnetic behavior is the occurrence of superconductivity in some heavy-fermion systems, which means that the pair breaking due to the magnetic moments is suppressed. The magnetic moments no longer dephase the Cooper pairs. However, this detection of the nonmagnetic character is restricted to potential superconductors. We use a rather similar method which detects the dephasing of the electrons by the magnetic impurities —weak localization. By this method, which has been described in several review articles,[6–9] one can quantitatively measure the dephasing rate due to magnetic impurities.

When one is using weak localization, it is very favorable to use disordered thin films. Our films are quench condensed *in situ* in an especially built dilution refrigerator where the substrate is maintained at 4.2 K during the evaporation. This has the very desirable feature that one can produce sandwiches step by step and analyze them after each evaporation step. Here it is possible to separate the additional magnetic scattering from the inelastic background of the host metal.

Since the refrigerator operates without the use of exchange gas, it works as a cryopump and provides an ultrahigh vacuum for the film preparation; this avoids oxidation of the magnetic impurities. The magnetoresistance measurements are performed in the temperature range between 80 mK and 4 K in fields up to 2.2 T. The excitation to the film is low enough that the electron temperature does not exceed the measured phonon temperature. This has been verified by measurements of the logarithmic temperature dependence of the film resistance. Our films are 0.5 mm wide and have an aspect ratio of 52.

For the intended investigation we require Kondo systems with high Kondo temperatures T_K. We investigated three different systems. For each system we describe a characteristic sandwich which has been stepwise evaporated and investigated. The investigated systems are the following:

(i) Fe in Mg as a bulk impurity with a Kondo temperature of about 45 K. We investigated stepwise a sandwich consisting of 31.2 atomic layers (atola) of Mg, about 0.008 atola of Fe, and 5.2 atola of Mg.

(ii) Fe at the surface of Mg which behaves differently than in the bulk. Its Kondo temperature is unknown but our experimental results suggest that T_K lies clearly above 20 K. We studied a sandwich consisting of a Mg film of 30.7 atola and about 0.0007 atola of Fe; and in a third step, an additional 0.0015 atola of Fe was later added.

(iii) Co at the surface of Cu. The latter system has unusual properties: Co in Cu has no magnetic moment (below about 500 K). However, Co at the surface of Cu has been recently investigated by means of weak localization.[10] It shows clearly a magnetic scattering which has a maximum at 23 K corresponding to a Kondo temperature of $T_K = 23$ K. We investigated a sandwich consisting of a Cu film of 33 atola with a coverage of approximately 0.0025 atola of Co; an additional 0.0045 atola of Co was later added. Superimposing a few atola of Cu resulted in an almost complete suppression of the Co moments in agreement with bulk measurements (see for example the review by Daybell[11]).

Figure 1 shows some typical magnetoresistance curves for the Mg/Fe sandwich. Each curve has its separate field scale that is indicated at its right-hand side. The left ordinate gives the change of the resistance in ohms. Since the theory yields the change of the conductance in units of $L_{00} = e^2/2\pi^2\hbar$, we have added on the right-hand side a conductance scale in these units. The points represent the experimental results. The full curves are the theoretical fits resulting from the Hikami-Larkin-Nagaoka theory.[12] This theory has been intensively discussed in the above-cited review articles and many preceding papers. Therefore we recall here only the essential physics. By means of magnetoresistance measurements on thin films one determines the phase-coherence time τ_ϕ of the conduction electrons. This phase coherence is limited by two processes, the inelastic scattering of the electrons and scattering by magnetic impurities. The total dephasing rate is essentially the sum of these two rates. (More precisely one obtains the dephasing rate of singlet and triplet electron pairs.) To avoid confusion it should be emphasized that the dephased scattering caused by the magnetic impurities is denoted by "magnetic scattering." This magnetic scattering can be either scattering of the conduction electrons according to the exchange interaction $JS\sigma$ or, at low temperature, an induced inelastic scattering due to the magnetic impurity. Since we can measure the inelastic background of the pure Mg or Cu film before doping with Fe or Co, the change in the dephasing rate corresponds to the additional scattering by the magnetic impurities.

The temperature dependence of the magnetic scattering in the Mg/Fe/Mg film is shown in Fig. 2. The magnetic scattering appears to be almost constant between 2 and 4 K and drops to about half its value as the temperature is lowered to 80 mK. The observed plateau around 3 K fits very well to the temperature dependence ob-

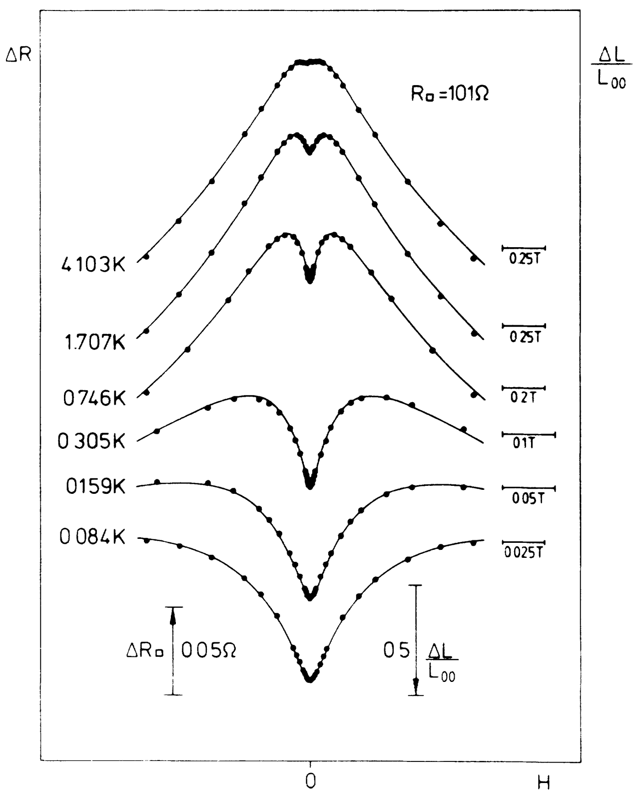

FIG. 1. Typical magnetoresistance curves of the Mg film covered by approximately 0.0022 atola of Fe. The field units are indicated at the right-hand side of each curve; the arrows give the scale for resistance and conductance changes. The points represent the experimental result; the curves are calculated with the Hikami-Larkin-Nagaoka theory.

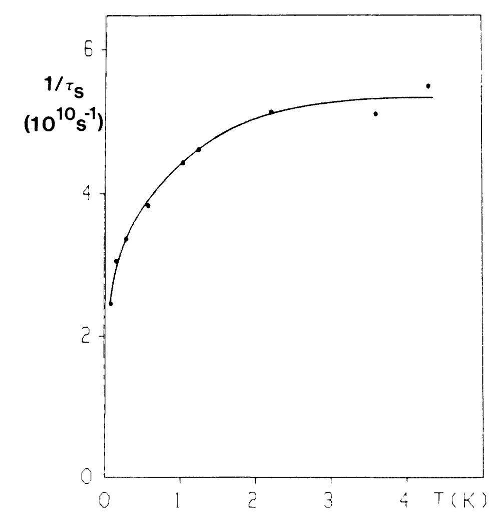

FIG. 2. Temperature dependence of the magnetic scattering rate in the Mg/Fe/Mg sandwich. The points show the measurements; the line is a guide for the eye.

served in similar films at higher temperatures.[3] What is new is the fact that there is a second decrease of $1/\tau_s$ below 2 K indicating a cooperative effect between the magnetic moments (and the conduction electrons). It appears to introduce a new energy scale of about 2 K into the system.

For Fe at the surface of Mg, or Co at the surface of Cu, we observe a strong decrease of the magnetic scattering to lower temperatures in the whole temperature range. It decreases by a factor of about 7 (14) for the higher (lower) Fe concentration on Mg and by factors of 7 and 9 for Co on Cu. In Fig. 3 the magnetic scattering rate is shown for the Fe/Mg and Cu/Cu systems as a function of the temperature in a log-log plot. The full straight lines have a slope of 0.50 which means that the magnetic scattering time varies at low temperatures at $T^{1/2}$.

We do not believe that this reduction of the magnetic scattering can be explained by the formation of a spin-glass. If we assume a spin-glass freezing where the axes of the moments are distributed isotropically (Heisenberg spin-glass) then the magnetic moments act as random fields which violate time-reversal invariance and yield a constant magnetic scattering rate. In fact, recent calculations[13] show that the magnetic scattering decreases only by a factor of $S/(S+1)$ when free spins freeze into a Heisenberg spin-glass. This cannot explain our observed reduction of $1/\tau_s$ by factors of 7 and more. If, on the other hand, the magnetic moments freeze either parallel or antiparallel to each other, we have an Ising spin-glass. Its effect on weak localization is more complicated and will be published elsewhere. However, in the case of dominating spin-orbit scattering, i.e., if we have a positive magnetoresistance curve, the effect of the Ising spin-glass is essentially the same as in a Heisenberg spin-glass. Our experimental data are essentially taken in the range of strong spin-orbit scattering and therefore we believe that the observed reduction of $1/\tau_s$ cannot be explained by the formation of a spin-glass.

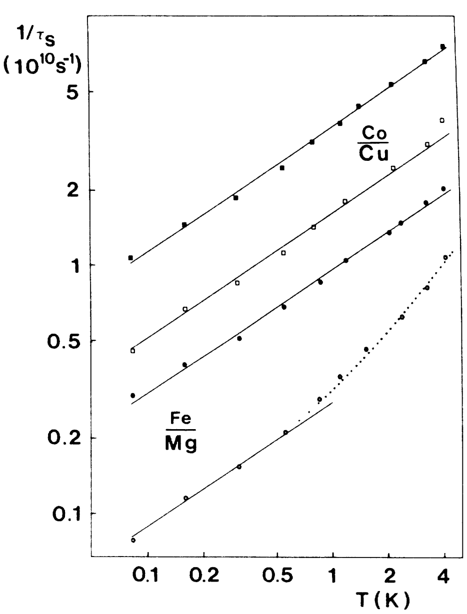

FIG. 3. Double-logarithmic plot of the temperature dependence of the magnetic scattering rates in the Mg/Fe films with about 0.0007 atola of Fe (open circles) and 0.0022 atola of Fe (filled circles), and the CuCo films with about 0.0025 atola of Co (open squares) and 0.007 atola of Co (filled squares), respectively. The slope of the solid lines is 0.50.

Nozières[2] derived on the basis of Wilson's "renormalization approach"[14] a Fermi-liquid theory for the single Kondo impurity. At low temperature he found no spin-flip scattering due to the magnetic impurity but an inelastic scattering which depends as T^2 on the temperature. The low-temperature inelastic scattering arises from an indirect electron interaction. However, at low temperatures, our experimental dephasing rate shows no quadratic behavior. Furthermore, our former experiments showed interaction effects in the Mg/Fe/Mg system already above 4.5 K. We therefore conclude that our observed magnetic scattering rate is not a single-Kondo-impurity behavior but intimately connected with impurity interactions.

The simplest (but not simple) case of interacting magnetic impurities is the problem of two impurities. This problem has been recently investigated by Jones and Varma for two Kondo impurities[15] and by Fye, Hirsch, and Scalapino for two Anderson impurities.[16] Jones and Varma have applied Wilson's renormalization-group methods and found for ferromagnetic interaction that the impurity spins are strongly correlated. Even for a small interaction energy they obtain a ferromagnetic alignment of the two Kondo spins. The quantum Monte Carlo calculations by Fye, Hirsch, and Scalapino show mutual polarization of the magnetic impurities in analogy to the electrical polarization of two atoms (which yield the van der Waals forces). The spin-spin correlation $\langle\sigma_1^z\sigma_2^z\rangle$ is essentially given by $J(r)/4T_K$ where $J(r)$ is the interaction energy between the magnetic impurities for the distance r. The spin-spin correlation does not change very much below the Kondo temperature, which remains the characteristic energy of the system. On the other hand, the experimental decrease of $1/\tau_s$ in Mg/Fe/Mg below 2 K suggests that the interaction in the alloy does introduce a new (low) energy into the system. Therefore the theory of single and pairs of magnetic impurities cannot explain our data.

A $T^{1/2}$ power law of the dephasing scattering rate represents a very unusual and rather challenging temperature dependence which demands a theoretical understanding. It does not strictly fulfill the condition for a Landau-Fermi-liquid behavior even when the magnetic moments become suppressed because it means that for sufficiently small energies the smearing of the quasiparticle energy due to inelastic effects is larger than the excitation energy itself. The Fermi-liquid theory requires the opposite condition: that at least at low energies (and temperatures) the quasi-particle energy is well defined. A square-root temperature dependence of the magnetic scattering makes it, of course, desirable to extend the measurements to lower temperature to see whether there is a change in the power law.

Summarizing our results we find that (i) there is a new universal behavior proportional to $T^{1/2}$ for the scattering by interacting magnetic impurities, (ii) the data suggest the disappearance of the magnetic moments at zero temperature, (iii) the data violate the Fermi-liquid behavior, and (iv) a new temperature, different from T_K, is seen in the interacting system.

The research was supported by National Science Foundation Grant No. DMR-8521662.

[1]J. Kondo, Prog. Theor. Phys. **32**, 37 (1964).

[2]P. Nozières, J. Low Temp. Phys. **17**, 31 (1974).

[3]G. Bergmann, Phys. Rev. Lett. **58**, 1236 (1987).

[4]R. Blanc and M. Belzons, Solid State Commun. **27**, 1137 (1978).

[5]G. Bergmann, Phys. Rev. Lett. **57**, 1460 (1986).

[6]B. L. Altshuler, A. G. Aronov, D. E. Khmelnitskii, and A. I. Larkin, in *Quantum Theory of Solids,* edited by I. M. Lifshits (MIR Publishers, Moscow, 1982).

[7]H. Fukuyama, Surf. Sci. **113**, 489 (1982).

[8]G. Bergmann, Phys. Rep. **107**, 1 (1984).

[9]P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).

[10]Wei Wei and G. Bergmann, Phys. Rev. B (to be published).

[11]M. D. Daybell, in *Magnetism,* edited by G. T. Rado and H. Suhl (Academic, New York, 1972), Vol. 5, p. 121.

[12]S. Hikami, A. I. Larkin, and Y. Nagaoka, Prog. Theor. Phys. **63**, 707 (1980).

[13]Wei Wei, G. Bergmann, and R. P. Peters, unpublished.

[14]K. G. Wilson, Rev. Mod. Phys. **47**, 773 (1975).

[15]B. A. Jones and C. M. Varma, Phys. Rev. Lett. **58**, 843 (1987).

[16]R. M. Fye, J. E. Hirsch, and D. J. Scalapino, Phys. Rev. B **35**, 4901 (1987).