3-28-1994

Absence of Size Dependence of the Kondo Resistivity

V. Chandrasekhar

P. Santhanam

N. A. Penebre

Richard A. Webb
University of South Carolina - Columbia, webbra@mailbox.sc.edu

H. Vloeberghs

See next page for additional authors

Publication Info

Published in *Physical Review Letters*, Volume 72, Issue 13, 1994, pages 2053-2056.
Chandrasekhar, V., Santhanam, P., Penebre, N.A., Webb, R.A., Vloeberghs, H., Van Haesendonck, C., and Bruynseraede, Y. (1994). Absence of Size Dependence of the Kondo Resistivity. *Physical Review Letters*, *72*(13), 2053-2056. doi: 10.1103/PhysRevLett.72.2053

Absence of Size Dependence of the Kondo Resistivity

V. Chandrasekhar,[1] P. Santhanam,[2] N. A. Penebre,[2] R. A. Webb,[2] H. Vloeberghs,[3]
C. Van Haesendonck,[3] and Y. Bruynseraede[3]

[1]*Department of Physics and Astronomy, Northwestern University, 2145 Sheridan Road, Evanston, Illinois 60208*
[2]*IBM Research Division, Thomas J. Watson Research Center, Yorktown Heights, New York 10598*
[3]*Laboratorium voor Vaste Stof-Fysika en Magnetisme, Katholieke Universiteit Leuven, B-3001 Leuven, Belgium*
(Received 11 August 1992; revised manuscript received 17 September 1993)

We have measured the low temperature resistivity of AuFe wires in the dilute magnetic impurity limit as a function of wire width, temperature, and magnetic field. When the width dependence of the electron-electron interaction contribution to the resistivity is taken into account, the temperature dependence of the remaining Kondo contribution to the resistivity of all samples with the same impurity concentration is identical. Similar behavior is observed for the magnetic field dependent resistivity. Thus, the Kondo contribution to the resistivity is independent of width down to 38 nm, much smaller than the Kondo length $\xi_K = \hbar v_F / k_B T_K \approx 10$ μm.

PACS numbers: 72.15.Qm, 72.10.Fk, 75.20.Hr

For nearly thirty years, the Kondo effect in metals with dilute magnetic impurities has remained a subject of continuing experimental and theoretical interest [1]. Its best known manifestation is a contribution to the low temperature resistivity ρ which is associated with the screening of the magnetic moments of single isolated impurities by the conduction electrons of the metal. As the temperature is lowered, more conduction electrons participate in the screening process; enhanced scattering of electrons by the screened impurity gives rise to a logarithmic increase in $\rho(T)$. Below a characteristic Kondo temperature T_K, the screening becomes increasingly efficient, so that eventually, at $T \ll T_K$, the impurity spin is entirely compensated, and $\rho(T)$ saturates. A popular physical picture of this screening process is that the electrons form a "cloud" of opposite spin polarity around the impurity [2]. As the temperature is lowered through T_K, more electrons are added to the cloud, so that finally at $T \ll T_K$ the total spin moment of the electron cloud exactly compensates the spin of the impurity. However, experimental evidence for the existence of such a temperature dependent spin polarization cloud from NMR, electron spin resonance, neutron diffraction, and Mössbauer studies has been far from conclusive [3].

The spatial extent of the screening cloud is believed to be on the order of the Kondo length $\xi_K = \hbar v_F / k_B T_K$, where v_F is the Fermi velocity of the host metal [2]. ξ_K can be quite large. For example, in AuFe, with $T_K \approx 1$ K, $\xi_K \approx 10$ μm. With current lithography capabilities, one can fabricate samples which have one or more dimensions smaller than ξ_K, making it possible to experimentally probe the spatial extent of the spin cloud by investigating the size dependence of the Kondo effect. A number of recent papers have reported performing such experiments. Chen and Giordano [4] measured $\rho(T)$ of dilute AuFe films in the temperature range 1.7 to 4 K as a function of film thickness. They observed a decrease in the logarithmic Kondo slope as the film thickness was decreased below ≈ 300 nm. Blachly and Giordano [5] found a similar reduction in the Kondo slope in AuFe wires as the width of the wires was reduced below ~ 200 nm. DiTusa *et al.* [6] measured $\rho(T)$ of 1000 and 2000 ppm CuCr wires as a function of wire width. They also observed a reduction in the logarithmic Kondo slope when the width of the wires was reduced below 10 μm. These authors attributed their observations to the effects of restricting the screening cloud by reducing one of the sample dimensions below ξ_K. However, earlier work [7,8] on CuCr and AuFe thin films found a spin-flip impurity scattering rate which corresponded to the expected scattering rate in the bulk, independent of film thickness down to 5 nm. In the work of Ref. [6], the concentration of magnetic impurities was high enough so that the samples were in the spin-glass regime, where interactions between impurities cannot be ignored. Furthermore, recent work [9] on AuFe spin-glass samples has shown that the spin-glass resistivity is extraordinarily sensitive to sample size and small variations in the elastic mean free path.

In this Letter, we present measurements of the temperature and field dependence of ρ in dilute AuFe wires with widths W ranging from 38 nm to 105 μm. The samples were prepared by two different techniques: flash evaporation from a master AuFe alloy, and ion implantation of Fe into Au films. The Fe concentrations were 60 ppm and 50 ppm, respectively, so that impurity interactions were not expected to be dominant [10]. In contrast to the studies mentioned above [4–6], we find that the Kondo contribution to $\rho(T)$ is essentially identical for samples of all widths with the same Fe concentration, once the electron-electron interaction contribution is taken into account. Thus, there appears to be no size dependence of the Kondo contribution down to length scales much smaller than ξ_K.

The ion-implanted samples for this study were patterned onto oxidized Si substrates using e-beam lithography, and fabricated by thermal evaporation of 99.999% pure Au followed by lift-off. The thickness of the films was 30 nm. The sheet resistance $R_\square$ of samples wider

0031-9007/94/72(13)/2053(4)$06.00

than ≈ 500 nm was $\approx 0.55\ \Omega$; below this, $R_\square$ increased monotonically with decreasing width to $\approx 1.0\ \Omega$ for the 38 nm wire. To facilitate measurement, the samples were designed so that the total number of squares in each sample was at least 200, the lengths of the samples ranging from $\sim 87\ \mu$m for the narrowest wire to ~ 23 mm for the widest wire. Using weak localization [11] measurements on the samples prior to ion implantation, and also on additional "pure" Au control samples, the electron phase coherence length was determined to be in excess of 5 μm at 40 mK, indicating that the concentration of magnetic impurities in the "pure" Au films was small. Ion implantation of all the films was carried out in one run at 60 keV at a dose of 8.85×10^{12} ions/cm^2, to give an expected concentration of 50 ppm Fe in Au. Standard statistical studies [12] of the implantation process indicate that the Fe ion profile is approximately Gaussian with a peak 14.7 nm below the Au surface and a standard deviation of 13.7 nm. The flash-evaporated samples were fabricated in a single run on stencils defined by e-beam lithography using evaporation techniques that have been described in detail elsewhere [8]. The starting source was a ≈ 100 ppm AuFe alloy, and the Fe concentration in the samples was estimated to be 60 ppm, based on the slope of $\rho(T)$ for the wider films [13]. The film thickness of these samples was also 30 nm. Dimensions of both ion-implanted and flash-evaporated samples were determined after measurement by scanning electron microscopy.

The samples were measured in a top-loading dilution refrigerator with a high-field magnet at temperatures down to ≈ 40 mK, using four-terminal resistance bridges. During the course of this study, the narrower samples were found to be extraordinarily sensitive to heating. Excitation currents as low as 1 nA, much smaller than expected from our previous experience measuring narrow wires, were necessary in order to avoid current dependent behavior at the lowest temperatures.

Figure 1(a) shows the excess resistivity $\Delta\rho(T)$ both before and after ion implantation for a 105 μm wide wire at zero magnetic field. Before ion implantation, there are three dominant contributions to $\Delta\rho(T)$: the phonon contribution at high temperatures, which leads to a rapid increase in $\Delta\rho(T)$ above 8 K, and the electron-electron (EE) interaction and weak localization (WL) contributions at low temperatures [11]. The EE and WL contributions are negligible on the scale of the plot: $\Delta\rho$ is essentially temperature independent below 4 K. For narrower samples, however, both contributions can be appreciable. After ion implantation, the sample shows an additional low temperature contribution to $\Delta\rho(T)$ which increases with decreasing temperature. This can be attributed solely to the presence of the magnetic impurities. From the logarithmic slope of this curve, we estimate the impurity concentration to be 52 ppm [13]. The solid curve is a fit by the empirical form [10]

$$\Delta\rho(T) = A - B\ln[1+(T/\theta)^2], \qquad (1)$$

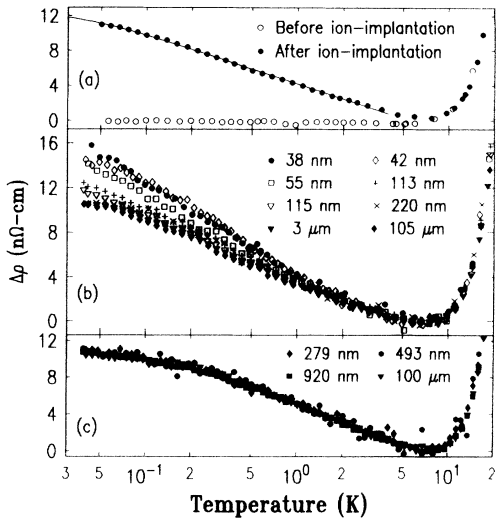

FIG. 1. (a) $\Delta\rho(T)$ for the 105 μm wide sample at $H=0$. Solid line is a fit by Eq. (1) with $A=12.9$, $B=0.1$, and $\theta=0.038$ K. (b) $\Delta\rho(T)$ for eight ion-implanted samples at $H=0$. (c) $\Delta\rho(T)$ for flash-evaporated samples at $H=0$.

where $\ln(\theta/T_K) = -\pi[S(S+1)]^{1/2}$, S being the spin of the impurity. Equation (1) has the expected quadratic dependence [14] on T for $T \ll T_K$ and the standard logarithmic dependence for $T \geq T_K$. Assuming $S=\frac{1}{2}$ [10], we obtain a value of $T_K=0.52$ K from this fit, in agreement with the range of 0.3–0.8 K quoted in the literature for AuFe [10,13].

Figure 1(b) shows $\Delta\rho(T)$ for eight of the ion-implanted samples at zero magnetic field. At high temperatures (8–20 K), data for all samples fall on the same curve due to electron-phonon scattering. At low temperatures, data for the two widest samples also fall on the same curve. For the six narrowest samples, $\Delta\rho$ increases more rapidly with decreasing temperature at low temperatures, the increase being larger the narrower the sample. Figure 1(c) shows $\Delta\rho(T)$ for the four flash-evaporated samples. Here the data for all samples fall essentially on one curve.

As noted above, the EE and WL contributions may be appreciable for the narrower samples. Because of the presence of magnetic impurities, the WL contribution is expected to have a very weak temperature dependence, and can be ignored [7,8,11,15]. Thus, the difference between $\rho(T)$ for the narrowest and the widest wires should contain only the EE contribution and any contribution due to the size dependence of the Kondo effect. Figure 2(a) shows the difference between $\rho(T)$ for the 38 nm and the 105 μm AuFe wires as a function of temperature, as well as data for a 38 nm wide "pure" Au wire with the same value of $R_\square$ at a magnetic field of 0.1 T. The 38 nm "pure" Au wire is expected to have contributions only from electron-electron interactions, since the application of the field eliminates the WL contribution. The two

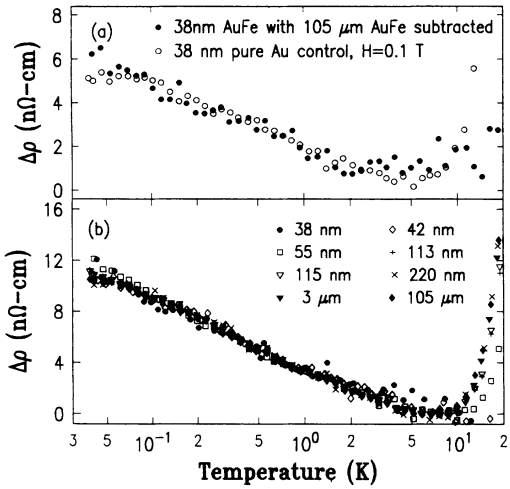

FIG. 2. (a) Data for the 38 nm AuFe wire of Fig. 1(b), with the data of the 105 μm AuFe data subtracted. Also shown are the data for the 38 nm "pure" Au wire at $H=0.1$ T. (b) Data of Fig. 1(b), with the EE contribution subtracted for the six narrowest samples as described in the text.

traces are almost identical, showing that the difference between the narrow and wide AuFe wires is not due to the presence of magnetic impurities.

In one dimension, the EE contribution is expected to have the form [11]

$$\Delta\rho_{ee}(T)=\alpha\frac{R_{\square}^2 t}{\pi\hbar/e^2}\frac{L_T}{W}, \tag{2}$$

where the thermal diffusion length $L_T=(\hbar D/k_B T)^{1/2}$ determines the dimensionality of the EE contribution, t is the film thickness, $D=(1/3)v_F l$ is the diffusion constant (l being the elastic mean free path), and α is a constant of order unity [11]. From Eq. (2), the EE contribution scales with W and $R_{\square}$ as $R_{\square}^{3/2}/W$; we have experimentally confirmed this scaling in "pure" Au samples of different widths above 100 mK [16]. Consequently, we can subtract the EE contribution for the six narrowest samples of Fig. 1(b) by subtracting the data for the 38 nm "pure" Au wire of Fig. 2(a) scaled by the appropriate value of $R_{\square}^{3/2}/W$ for each sample. The resulting plot is shown in Fig. 2(b). The sole remaining contribution is the Kondo contribution, and it can be seen that, within the noise, this is the same for all the samples. No subtraction is required for the flash-evaporated samples: Since they are all wider than 250 nm, the electron-electron contribution is not appreciable.

Further evidence of the size independence of the Kondo effect can be seen by measuring $\Delta\rho$ as a function of magnetic field. $\Delta\rho(H)$ in the Kondo regime is expected to have the same form as $\Delta\rho(T)$ [10]: quadratic in H at low values of field, and logarithmic at higher values of H. Figure 3(a) shows the $\Delta\rho(H)$ for eight ion-implanted samples on a semilogarithmic plot. Note that $\Delta\rho(H)$ is

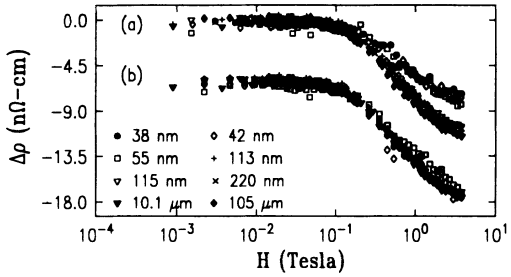

FIG. 3. (a) Magnetic field dependence of the resistivity for eight ion-implanted samples at 40 mK. (b) Data of (a), with $\Delta\rho_{ee}(H)$ subtracted for the six narrowest wires, as described in the text. The ordinate has been shifted for clarity.

indeed similar in form to $\Delta\rho(T)$, and that the change in ρ over this field range is comparable to the change in ρ as a function of temperature for the wider samples. The difference between the narrower samples and the wider samples can again be attributed to the EE contribution. In one dimension, the field dependence of this contribution $\Delta\rho_{ee}(H)$ at high fields ($g\mu_B H\gg k_B T$, $g=2$) is given [11] by an equation similar to Eq. (2), with α replaced by another constant α' and L_T replaced by the spin diffusion length $L_s=(\hbar D/g\mu_B H)^{1/2}$. Because of the presence of the field dependent WL contribution, one cannot simply subtract $\Delta\rho_{ee}(H)$ for the 38 nm pure Au wire to eliminate the EE contribution. Assuming that the difference between $\Delta\rho_{ee}(H)$ for the 38 nm and 105 μm wires is due to the EE contribution, however, one can subtract this difference scaled by $R_{\square}^{3/2}/W$ for the next five narrowest samples in Fig. 3(a). The resulting curves are shown in Fig. 3(b). The curves are essentially identical. Similar results are obtained for the flash-evaporated samples. *Thus, the Kondo contribution to the resistivity appears to be independent of the width of the wires down to widths of 38 nm, a size scale much smaller than ξ_K.*

There could be a number of reasons that we do not observe a finite-size effect. First, the interpretation of finite-size effects in terms of a Kondo screening cloud of radius ξ_K may have validity only in the temperature range $T\ll T_K$, where the screening cloud is fully formed. The experiments of Refs. [4–6] were performed at temperatures comparable to or larger than T_K. Even our measurements, although they go to temperatures $T\approx 0.1T_K$, may not go low enough in temperature. Second, there is some question as to whether one should observe size effects in a transport measurement, where, by the necessity of having measurement probes, only the shape but not the total volume of the screening cloud is restricted. Size effects would then only be observable when the total volume of the sample is less than $\approx\xi_K^3$. Third, it is possible that the size of the screening cloud is not ξ_K, but some smaller length scale. Bergmann [17] has recently presented a calculation that suggests the mean number of electrons needed to screen a single im-

purity is $\approx 10\epsilon_F/k_BT_K$. Using the average volume per electron for Au, this gives a length scale of ≈ 10 nm, much smaller than the width of our narrowest sample. Another view is that no size effects should be seen until the electronic density of states is modified [18]. Finally, the ubiquitous presence of nonmagnetic impurities may also change the Kondo length scale by making the motion of the conduction electron diffusive rather than ballistic [19]. The distance that an electron travels during the lifetime of the Kondo resonance would then not be the ballistic length ξ_K but the diffusive length $(\hbar D/k_BT_K)^{1/2}$, which is much shorter (≈ 350 nm for our samples). However, the fact that there appears to be no size dependence in our samples even below this length would argue that this last argument is not the correct explanation for our observations. Further experiments are required before these questions can be resolved.

We thank A. Verbruggen for the *e*-beam lithography and P. Holweg for making SEM micrographs of the flash-evaporated samples. Work at Leuven was supported by the Belgian Concerted Action (GOA) and Inter-University Attraction Poles (IUAP) programs. H.V. and C.V.H. have been supported financially by the Belgian National Fund for Scientific Research (NFWO). V.C. was supported in part by the NSF under DMR-9357506, and the MRL program of the NSF, under DMR-9120521.

[1] For review articles, see G. Gruner and A. Zawadowski, Rep. Prog. Phys. **37**, 1497 (1974); N. Andrei, K. Furiya, and J. H. Lowenstein, Rev. Mod. Phys. **55**, 331 (1983). See also Refs. [10] and [13].

[2] Y. Nagaoka, Phys. Rev. **138**, A1112 (1965); Prog. Theor. Phys. (Kyoto) **37**, 13 (1967); H. Ishii, J. Low Temp. Phys. **32**, 457 (1978); J. E. Gubernatis, J. E. Hirsch, and D. J. Scalapino, Phys. Rev. B **35**, 8478 (1987).

[3] D. C. Golibersuch and A. J. Heeger, Phys. Rev. **182**, 584 (1969); A. J. Heeger, L. B. Welsh, M. A. Jensen, and G. Gladstone, Phys. Rev. **172**, 302 (1968); J. B. Boyce and C. P. Slichter, Phys. Rev. Lett. **32**, 61 (1974), and references therein.

[4] G. Chen and N. Giordano, Phys. Rev. Lett. **66**, 209 (1991).

[5] M. A. Blachly and N. Giordano, Phys. Rev. B **46**, 2951 (1992).

[6] J. F. DiTusa, K. Lin, M. Park, M. S. Isaacson, and J. M. Parpia, Phys. Rev. Lett. **68**, 678 (1992).

[7] R. P. Peters, G. Bergmann, and R. M. Mueller, Phys. Rev. Lett. **58**, 1964 (1987).

[8] C. Van Haesendonck, J. Vranken, and Y. Bruynseraede, Phys. Rev. Lett. **58**, 1968 (1987).

[9] H. Vloeberghs, Ph.D. thesis, Leuven (unpublished).

[10] M. D. Daybell, in *Magnetism*, edited by G. Rado and H. Suhl (Academic, New York, 1973), Vol. 5.

[11] For a review article discussing both weak localization and electron-electron interactions, see B. L. Altshuler and A. G. Aronov, in *Electron-Electron Interactions in Disordered Systems*, edited by A. L. Efros and M. Pollak (Elsevier, Amsterdam, 1985).

[12] J. F. Ziegler and J. P. Biersak, computer program TRIM, in *The Stopping and Range of Ions in Solids* (Pergamon, New York, 1992).

[13] This and all other estimates of Fe concentration in this paper are determined from a slope of 0.11 nΩ cm/ppm decade K based on the AuFe data of Loram, Whall, and Ford shown in A. J. Heeger, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic Press, New York, 1969), Vol. 23, p. 283. However, data from another publication by the same group [J. W. Loram *et al.*, Phys. Rev. B **2**, 857 (1970)] gives a slope of 0.17 nΩ cm/ppm decade, giving a $\approx 50\%$ reduction in our estimates. This uncertainty does not affect our conclusions.

[14] P. Nozières, J. Low Temp. Phys. **17**, 31 (1974); P. D. Sacramento and P. Schlottman, Solid State Commun. **73**, 747 (1990).

[15] The interplay between weak localization and the Kondo effect has been discussed by many authors. See, for example, F. J. Okhawa and H. Fukuyama, J. Phys. Soc. Jpn. **53**, 2640 (1984).

[16] Data for the "pure" Au control samples above 100 mK are well described by Eq. (2), with a value of $\alpha \sim 1.3$, in good agreement with theory and previous experiments (see Ref. [11]). To our knowledge, the $1/\sqrt{T}$ dependence predicted by Eq. (2) has not been experimentally verified at very low temperatures.

[17] G. Bergmann, Phys. Rev. Lett. **67**, 2545 (1991), and references therein.

[18] P. W. Anderson, P. A. Lee, and B. L. Altshuler (private communications).

[19] Although there has been theoretical work on the effect of nonmagnetic impurities on the RKKY interaction [see, for example, A. Jagannathan *et al.*, Phys. Rev. B **37**, 436 (1988)], we are not aware of any published work which discusses their influence on the Kondo effect. The ballistic-to-diffusive argument given here is similar to the one given for the problem of electron transport in weakly disordered metals.