

Asymmetric Nonlinear Differential Resistance of Mesoscopic AuFe Spin-Glass Wires

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We have measured the differential resistance $R = dV/dI$ of mesoscopic AuFe wires as a function of temperature T and dc current bias I . $R(T)$ shows a maximum at a temperature $T_m \sim 1-4$ K, consistent with the onset of spin-glass order in these films. At temperatures $T < T_m$, $R(I)$ also shows a maximum; however, $R(I)$ is asymmetric in I , the asymmetry increasing with decreasing temperature. The asymmetry is sample specific, sensitive to the four terminal measurement configuration, and is associated with the presence of magnetic impurities in the samples. [S0031-9007(96)01168-4]

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The placement of a transition metal impurity in a noble metal host induces a collective response in the electron gas which attempts to screen the magnetic moment of the impurity. At low impurity concentration, enhanced scattering of electrons from the screened impurity leads to a characteristic logarithmic increase of the resistance with decreasing temperature, the well-known Kondo effect [1]. If the concentration of impurities is high, the interaction of the screening electrons around one impurity with those around other impurities leads to an effective impurity-impurity interaction, the RKKY (Ruderman-Kittel-Kasuya-Yosida) interaction [2]. At high temperatures, the thermal energy of the impurity spins is sufficient to overcome the RKKY interaction, and each individual spin is free to rotate independently. As the temperature is reduced, however, the impurity spins are increasingly fixed in random orientations by the RKKY interaction. The onset of this spin-glass order is signaled by a drop in the resistance of the host metal due to the reduced magnetic scattering of the conduction electrons. In combination with the Kondo effect at higher temperatures, this gives rise to a maximum in the resistance as a function of temperature which is characteristic of spin glasses [2].

Earlier work on magnetic impurities in metals concentrated on the properties of bulk materials; recently, with the opportunities presented by nanolithography, interest has focused on the properties of samples whose dimensions are comparable to relevant microscopic length scales. A number of such microscopic length scales have been proposed for both the Kondo effect [3] and spin glasses [4]. The hope is that measurements on mesoscopic samples would allow one to verify directly the existence of these microscopic length scales. However, the experimental evidence in both Kondo systems and spin glasses has so far been inconclusive. For example, measurements by some groups [5,6] of the Kondo effect in thin films, wires, and small point contacts defined by break junctions show a definite size dependence, but on vastly different length scales, while mea-

surements by other groups [7] on AuFe wires found no size dependence on the Kondo effect. The situation is similar for samples in the spin-glass regime [8-11]. Thus the issue of the existence of fundamental length scales in both the spin-glass and the Kondo regimes remains open.

In this Letter, we report on measurements of the low temperature differential resistance $R(I) = dV/dI$ of AuFe wires as a function of dc current bias I . As reported previously by other groups in measurements on CuCr wires [9] and point contact break-junction devices [10], we find that the shape of $R(I)$ reflects the behavior of the temperature dependent resistance $R(T)$, in that it has a maximum at a particular current I_m . In addition, however, we find that $R(I)$ is *asymmetric* in I , even in zero magnetic field. The asymmetry is small at high temperatures, but grows by more than an order of magnitude as the temperature is lowered, indicating that it is associated with enhanced spin scattering at low temperatures. The degree of asymmetry is sample specific, being larger in some samples and smaller in other nominally identical samples. Furthermore, we find that the asymmetric component of $R(I)$ is sensitive to the particular configuration of contact leads used in a four terminal measurement. We also find that the asymmetry depends on the size of the wire, being generally larger for shorter and narrower wires and disappearing entirely for very long samples. These observations are indicative of the mesoscopic nature of this phenomenon.

The samples in this experiment were patterned onto oxidized silicon substrates by conventional e -beam lithography techniques. After thermal deposition of 99.999% Au, the samples were implanted with Fe ions at energies and dosages calculated to give impurity concentrations of 0.2 and 0.4 at. % [12]. All samples of one concentration were fabricated and ion-implanted at the same time to ensure uniform film properties. The inset of Fig. 1(a) shows a schematic of one of the samples. The thickness of the films was 33 nm, and the sheet resistance R_{\square} was $\sim 1 \Omega$ at 4.2 K after ion implantation. The samples were measured in ^4He and ^3He cryostats (for the 0.4 at. % samples),

and a dilution refrigerator (for the 0.2 at. % samples). For the $R(T)$ measurements, a homemade four terminal ac bridge with a PAR 124 lock-in amplifier was used with current drive low enough to avoid self-heating effects. For the $R(I)$ measurements, a dc current was summed with the ac drive in the same bridge. Although we have measured a number of different samples, we shall discuss below the data for only a few representative samples.

Figure 1(a) shows $\Delta R(T) = R(T) - R(T = 13 \text{ K})$ for two samples: a $\sim 1.8 \mu\text{m}$ long, 0.2 at. % AuFe wire and a $\sim 3.5 \mu\text{m}$ long, 0.4 at. % AuFe wire. Both samples show the temperature dependence expected for a spin-glass sample discussed above with a maximum at a characteristic temperature T_m which depends on the concentration of impurities [2]. T_m is larger for a higher impurity concentration, as can be seen from the data in Fig. 1(a). Figure 1(b) shows $\Delta R(I) = R(I) - R(I = 0)$ for the same two samples of Fig. 1(a), along with data for a $\sim 1.8 \mu\text{m}$ long Au wire of the same geometry without any implanted Fe ions. $\Delta R(I)$ for both spin-glass samples is similar to the form of $\Delta R(T)$. In contrast, $\Delta R(I)$ for the pure Au sample shows only an approximately quadratic increase with increasing current. Such nonlinear current-voltage (IV) characteristics in spin-glass samples have also been reported recently by Lane *et al.* in CuCr wires

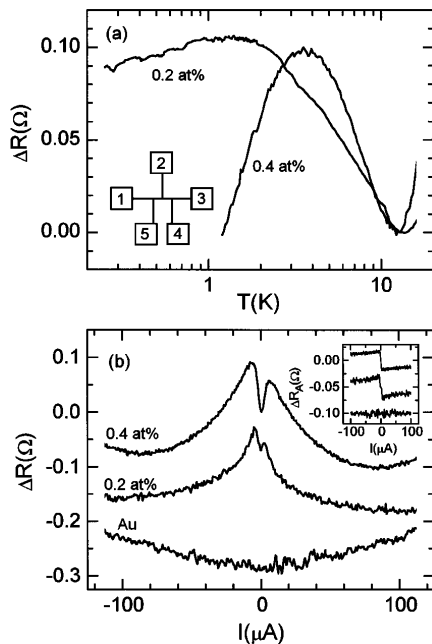


FIG. 1. (a) $\Delta R(T) = R(T) - R(13 \text{ K})$ of a 0.2 at. % AuFe wire (width $w = 135 \text{ nm}$, length $l = 1.8 \mu\text{m}$) and a 0.4 at. % AuFe wire ($w = 85 \text{ nm}$, $l = 3.5 \mu\text{m}$). Inset: A schematic of the samples. (b) $\Delta R(I) = R(I) - R(0 \mu\text{A})$ of the 0.4 at. % AuFe wire at $T = 1.705 \text{ K}$ and the 0.2 at. % AuFe wire at $T = 0.051 \text{ K}$, and a pure Au wire ($w = 117 \text{ nm}$, $l = 1.8 \mu\text{m}$) at $T = 1.36 \text{ K}$. Inset: The antisymmetric component $R_A(I)$ of the three samples of (b). The plots are offset for clarity. The resistance at 4.2 K of the 0.2 at. % sample is 17.7Ω , the 0.4 at. % sample 57.5Ω , and the pure Au wire 28.5Ω .

[13], Grabecki *et al.* in a magnetic heterostructure device [14], and van der Post *et al.* in a break-junction point contact [10]. Nonlinear IV curves have been reported also by many groups on thin film samples without magnetic impurities [15]. In most of these cases, the nonlinearities have been associated with heating of the conduction electrons by the dc bias to a temperature T_{eff} , which is higher than that of the substrate or phonon bath.

Although electron heating is the dominant contribution to the nonlinear behavior in our samples, it is not the only contribution. This can be seen by noting the symmetry of the $\Delta R(I)$ curves as a function of I in Fig. 1(b). The curves for both spin-glass samples are quite clearly *asymmetric* with respect to I . This asymmetry can be seen more clearly in the inset to Fig. 1(b), which shows the antisymmetric component $R_A(I)$ of the three traces in Fig. 1(b). For electron heating, one would expect $\Delta R(I)$ to be symmetric with respect to I since the heating is independent of the current direction. For example, $\Delta R(I)$ for the pure Au wire shows no asymmetry when measured on the same scale. Asymmetries in IV curves in mesoscopic samples have been reported earlier [15] and have been attributed to nonclassical physics; for example, asymmetric IV curves due to electron quantum interference in disordered metals. Comparing the data of the AuFe wires to that of the pure Au wire, it is clear that the asymmetries we observe are associated with the presence of magnetic impurities.

Figure 2(a) shows $\Delta R(I)$ for the 0.2 at. % sample of Fig. 1 at a few different temperatures in zero magnetic field. $R_A(I)$ increases as the temperature is reduced, as can be seen in Fig. 2(b), eventually becoming temperature independent below $\sim 1 \text{ K}$. To obtain a more quantitative

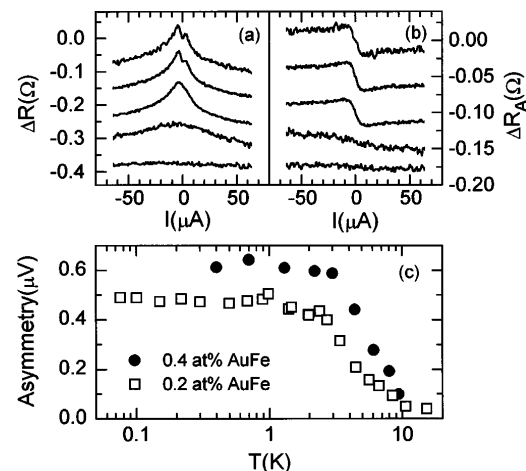


FIG. 2. (a) $\Delta R(I)$ and (b) $R_A(I)$ for the 0.2 at. % AuFe wire of Fig. 1 at five different temperatures. The temperatures are 0.10, 0.68, 1.47, 5.61, and 10.6 K from top to bottom. The plots are offset for clarity. (c) Integrated amplitude of $R_A(I)$ from 0 to $30 \mu\text{A}$ for the 0.2 at. % sample of Fig. 1, and a 0.4 at. % AuFe wire ($l = 1.85 \mu\text{m}$, $w = 85 \text{ nm}$) as a function of T .

estimate of the asymmetry, we have analyzed $R_A(I)$ as a function of temperature by integrating the magnitude of $R_A(I)$ from 0 to $30 \mu\text{A}$. The result of this integration is plotted in Fig. 2(c) for the 0.2 at. % sample of Fig. 1(a) and a $\sim 1.85 \mu\text{m}$ long 0.4 at. % sample. For both spin-glass samples, the antisymmetric component is small at high temperatures, but grows by more than an order of magnitude as the temperature is lowered, eventually saturating at low temperatures. This saturation occurs at $\sim 3 \text{ K}$ for the 0.4 at. % sample and $\sim 1 \text{ K}$ for the 0.2 at. % sample. Comparing these temperatures to the values of T_m for the two concentrations shown in Fig. 1(a), it is tempting to associate this saturation with the onset of the spin-glass transition. However, the saturation temperature obtained by this analysis is dependent on the range of integration, presumably due to electron heating at high current bias. Reducing the integration range lowers the saturation temperature by about (10–20)%, but the increased scatter in the data also increases the uncertainty in determining the saturation temperature by approximately the same amount. Consequently, we have chosen the integration range small enough to minimize the effect of heating but large enough to reduce the scatter in the data to an acceptable level.

The application of a magnetic field is known to strongly influence the properties of dilute magnetic alloys [1,2], and we find that it affects the asymmetry as well. Figure 3(a) shows $\Delta R(I)$ for the 0.4 at. % sample at three different magnetic fields at a temperature below the

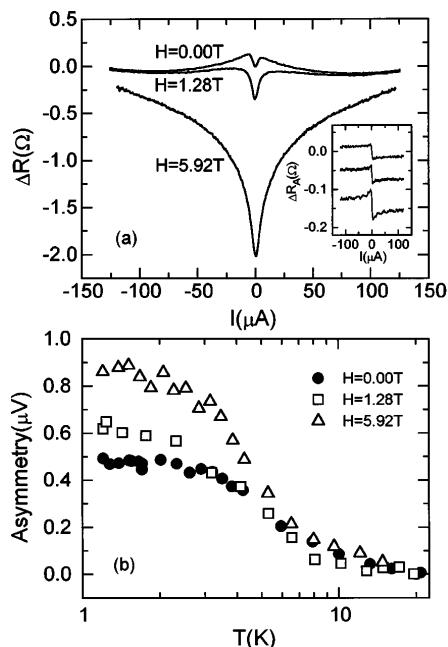


FIG. 3. (a) $\Delta R(I)$ of the 0.4 at. % AuFe wire of Fig. 1 at $T = 1.28 \text{ K}$ at three different magnetic fields. Inset: The antisymmetric component $R_A(I)$ of the traces in (a). The plots are offset for clarity. (b) Integrated amplitude of $\Delta R_A(I)$ for the 0.4 at. % sample of (a) as a function of T at three different magnetic fields.

zero field maximum temperature T_m . The corresponding antisymmetric components are shown in the inset. $R(I = 0)$ decreases in a magnetic field due to the large negative magnetoresistance in a spin glass [2]. Although $\Delta R(I)$ appears to become more symmetric with increasing field, the antisymmetric component $R_A(I)$ actually grows with magnetic field to 6 T. This can be seen in Fig. 3(b), which shows the integrated magnitude of $R_A(I)$ at three different magnetic fields. It should be noted, however, that we see no measurable difference between the field cooled and zero field cooled cases.

A number of further experimental aspects of this phenomenon point to the mesoscopic nature of the asymmetry. First, the effect is size dependent, in that longer and wider samples in general have a smaller degree of asymmetry than shorter and narrower samples, although the present geometry of our samples precludes the possibility of experimentally identifying the relevant mesoscopic length scale. Second, in measurements on a number of different samples, we have found that the degree of asymmetry is sample specific. Samples which are nominally identical in length, width, and concentration can have antisymmetric components which vary by factors of 2–3, even though their $\Delta R(T)$ curves may be similar. Such sample-specific behavior is reminiscent of small disordered metallic and semiconducting devices, and is a hallmark of mesoscopic samples [16]. Finally, both the amplitude and sign of $R_A(I)$ are sensitive to the measurement configuration in a four terminal measurement. Figure 4 shows four representative traces to illustrate this for the 0.4 at. % sample of Fig. 1. In order to identify the measurement probe configuration, we shall use the notation $R_{ij,kl}$, where the first two indices denote the current contacts and the last two the voltage contacts; the numbers refer to the contacts shown in the inset to Fig. 1(a). In all four traces shown in Fig. 4, the dc current flows in the same direction, and classically the same sample is being measured. However, switching current and voltage sometimes reverses the

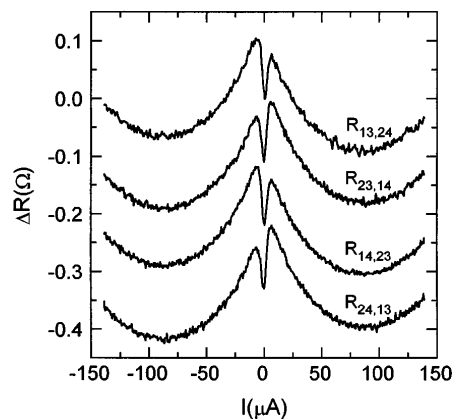


FIG. 4. $\Delta R(I)$ for the 0.4 at. % sample of Fig. 1 at $T = 1.56 \text{ K}$, measured with different probe configurations. The plots are offset for clarity.

sign of $R_A(I)$ and sometimes leaves it unaffected. We have tried all possible combinations of these four probes and have not been able to discern any pattern to this switching. It should be noted that only the *antisymmetric* part of $\Delta R(I)$ depends on the probe configuration; the *symmetric* part of $\Delta R(I)$ is independent of the arrangement of lead contacts.

Such unusual symmetry properties have been observed before in mesoscopic samples. Benoit *et al.* [17] noted that the four terminal magnetic field dependent conductance $G(B)$ of small metallic wires and loops was sensitive to the configuration of current and voltage contacts, even though classically the same sample was being measured. Buttiker [18] explained this as a consequence of mixing of the diagonal and off-diagonal components of the conductance in a four terminal phase coherent sample. Our experiment, however, is quite different from the situation discussed by Buttiker and Benoit *et al.* Because of the presence of magnetic impurities, we expect that the electron phase coherence length is much shorter than the length of the sample [19]. We have no external magnetic field applied to the sample; more important, the asymmetry we observe is with respect to dc current bias rather than magnetic field. In addition, unlike previous experiments which measured electron quantum interference effects in mesoscopic spin glasses [19], the asymmetry does not appear to depend on the relative microscopic orientation of magnetic impurity moments, although it may depend on the spatial distribution of the magnetic impurities. The evidence for this is that the sign and magnitude of the asymmetry in a single sample is completely reproducible, even after repeated thermal cycling to room temperature, a process which is expected to completely randomize the impurity spin orientations. Consequently, we believe that the asymmetry is not associated with electron quantum interference effects in our samples.

What is the origin of the asymmetry in these mesoscopic spin-glass devices? We have shown that it is clearly associated with the presence of magnetic impurities in the samples. One possibility is that this phenomenon is related to the enhanced thermopower in magnetic alloys [1,2] arising from local hot spots in the samples and leads. This mechanism will give a contribution to $R(I)$ antisymmetric in I and may account for the temperature dependence and sensitivity to measurement configuration. Another possibility is that the asymmetry is associated with changes in the electron density of states due to the magnetic impurities, similar to the zero bias Kondo anomalies observed in point contact devices [6,10,20]. Further experimental work needs to be performed before the microscopic origin of this effect can be determined.

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