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Low Temperature Anomaly in Mesoscopic Kondo Wires

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We report the observation of an anomalous magnetoresistance in extremely dilute quasi-one-dimensional AuFe wires at low temperatures, along with a hysteretic background at low fields. The Kondo resistivity does not show the unitarity limit down to the lowest temperature, implying uncompensated spin states. We suggest that the anomalous magnetoresistance may be understood as the interference correction from the accumulation of geometric phase in the conduction electron wave function around the localized impurity spin.

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A localized magnetic moment interacts with the conduction electrons in a metal resulting in a logarithmic increase of the resistivity as the temperature is lowered. This is known as the Kondo effect [1]. Below the Kondo temperature, T_K , an electron cloud begins to screen the impurity until its spin is completely compensated, forming a singlet state at low temperature. The nature of this state and the extent of the screening cloud has been studied for decades. Recently this effect has been explored in mesoscopic systems in an attempt to understand whether the screening is affected by the finite sample size [2–4], including high temperature large-concentration experiments on layered Kondo systems [5], and 2D films [3,6]. Interference effects in mesoscopic Kondo systems containing impurity concentrations $c > 50$ ppm do not generally contribute significantly to the measured magnetoresistance or resistivity because of the strong suppression of long range phase coherence due to spin-flip scattering. In spite of its relevance to mesoscopic systems, a complete study of the low temperature magnetoresistance in very dilute alloys ($c < 10$ ppm), where the Kondo screening length is comparable to the phase coherence length L_ϕ , has not been done. In this regime, an interference experiment which can reveal new information on the development of the Kondo screening cloud is possible. The three-dimensional character of the local dipolar magnetic field from the impurity spin coupled with an externally applied field should provide an additional interference contribution to the electron wave function. This is analogous to the Berry phase effect predicted for coherent electrons in a ring [7] traversing in an externally applied 3D magnetic field texture concentric with the ring.

In this paper, we report the magnetoresistance and the temperature dependence of the resistivity down to 38 mK for five quasi-1D AuFe wires in the concentration range of $3 < c < 10$ ppm. We determine both the spin-flip scattering rate and the phase decoherence rate by fitting the low field magnetoresistance to standard weak localization (WL) theory [8]. We find that the unitarity limit corresponding to the formation of the singlet state is not yet

reached at our lowest temperature [9] in spite of the fact that AuFe Kondo systems are known to have a Kondo temperature of 1 K [10,11]. At intermediate fields we observe a negative magnetoresistance that is characteristic in temperature dependence and shape of an interference correction, and different from the expected standard Kondo magnetoresistance. At low temperatures this magnetoresistance shows hysteresis which vanishes if the magnetic field is swept to a larger value or if the temperature is increased. We argue that our data is not consistent with a spin glass model but rather with a new interference correction similar to a Berry phase effect [7].

Our studies are done on pure (99.9995%) samples of gold (Au) before, and after, the ion implantation of 3–10 ppm of iron (Fe) impurities. This provides a clear advantage over earlier works on layered or flash-evaporated samples in that the contribution to the magnetoresistance at various field scales coming solely from the magnetic impurities could be easily identified. Sample dimensions, diffusion constant D , and L_ϕ measured after implantation are given in Table I. These samples are quasi-1D, since $w, t \ll L_T, L_\phi$, where $L_T = \sqrt{\hbar D/k_B T}$ is the thermal diffusion length. The Kondo contribution to the resistivity $\Delta\rho(T)$ is found to have the expected logarithmic increase [1]: $\Delta\rho(T) = A - B \ln(T)$ (see Fig. 1), after the subtraction of the electron-electron interaction contribution [4] measured before the ion implantation, which has the expected theoretical value [8], $\Delta\rho_{ee} \approx (2e^2 R^2 w t / \hbar L^2) L_T$.

The total scattering rate $1/\tau$ relevant for resistance is $1/\tau = 1/\tau_n + 1/\tau_s$; $1/\tau_n$ is the nonmagnetic scattering rate. The phase-breaking rate $1/\tau_\phi$ in the presence of magnetic scattering is given by $\tau_\phi^{-1} = 2\tau_s^{-1} + \tau_{\phi(\text{nonmag})}^{-1}$. Figure 2 displays the temperature dependence of the magnetic scattering rate $1/\tau_s$ obtained from WL measurements [5,6] for samples AuFe1 and AuFe2. $1/\tau_s$ is obtained from WL after subtracting the inelastic rate $1/\tau_{\phi(\text{nonmag})}$ due to nonmagnetic sources, measured in the same Au wires before ion implantation. The $1/\tau_\phi$ correction term does not produce the observed behavior seen in Fig. 2 because $1/\tau_s$

TABLE I. Sample parameters shown in Figs. 1–5.

Sample	w (nm)	t (nm)	L (μm)	R (Ω)	D m^2/s	L_ϕ (μm)	c (ppm)	B
AuFe1	180	40	155	393	0.02	1.9	10.9	0.52
AuFe2	120	40	155	599	0.02	2.2	7.1	0.29
AuFe3	100	35	155	803	0.02	1.7	6.0	0.24
AuFe4	210	135	4120	783	0.07	5.0	3.3	0.16
AuFe5	120	135	2750	1300	0.05	3.0	10.1	0.46

is much larger than $1/\tau_\phi$ in the corresponding clean system. The maxima near 0.2–0.4 K represent the previously observed resonant spin-flip scattering processes [6,12].

As shown in Fig. 1, the unitarity limit is not reached down to 40 mK, even in the presence of disorder and a finite magnetic field required to quench WL, both of which should help form the singlet state. This is consistent with earlier observations [4,11]. The impurity spin is thus not completely screened. However, at a larger magnetic field, a resistivity plateau is observed corresponding perhaps to the unitarity limit [see Fig. 3(a)]. The plateau shifted to higher temperatures with increasing magnetic field. Additionally, we observed a maximum around T_K [see Fig. 3(b)]. This observation is consistent with earlier experiments on $(\text{LaCe})\text{Al}_2$ and $(\text{LaCe})\text{B}_6$ [13], consequently explained by a wave description of the spin density [14]. This implies that there is a substantial spin polarization around the impurities with a potential $V(r) = V_0 \cos(2k_F r)/r^3$. The local magnetic field of the spin polarization can be on the order of a Tesla within a couple of nanometers from the impurity, though it is negligible on the scale of the typical interimpurity distance of ~ 10 nm. The strength of this potential V_0 is experimentally known to be very large for AuFe, decreasing exponentially with increasing concentration c [15]. Thus, there are strong local magnetic fields for purer samples with longer L_ϕ . NMR measurements of the conduction-electron spin density around Fe atoms in a Cu matrix also find a nonvanishing radial component above and below T_K [16].

That there exists a distribution of local magnetic fields from the impurity spins is further confirmed by the observation of hysteresis in the low field MR. As shown in Fig. 4, the background of the WL curve is asymmetric with a positive or negative slope depending on the field history. Hysteresis disappears at high temperatures, typically between 0.4 and 1.5 K, depending on the sample. In contrast to what is observed in a spin glass, we find this hysteresis to be stronger for systems with longer L_ϕ (hence for lower concentration samples). Hysteresis is expected for a spin glass system below T_g ; so if it were a spin glass, we would have observed stronger hysteresis for higher concentration samples, contrary to our data. Our experiment suggests that hysteresis arises because of different realizations of the three-dimensional local field distribution. As the sample gets cold, impurity spins freeze out in random orientations, providing a particular configuration for the local field distribution. This distribution is modified by a magnetic field due to spin alignment. Magnetic field cycling between ± 1 T removes the hysteresis and flattens the background of the low field MR, while cycling between ± 0.05 T does not.

All of our samples are in the single-impurity regime and the logarithmic increase of resistivity scales with concentration. It is unlikely that these systems behave like a spin glass for a number of reasons: (a) In AuFe, spin glass behavior is not observed for $c \ll 100$ ppm, as is well known [17]; (b) second, spin glass temperature T_g for a system with 3–10 ppm Fe in Au would be 1 mK or lower; (c) the

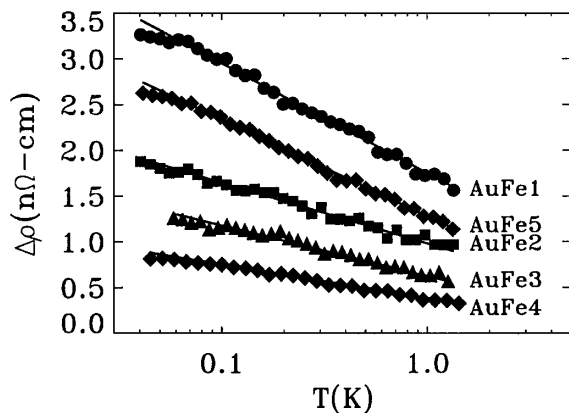


FIG. 1. $\Delta\rho(T)$ for samples AuFe1–AuFe5 at a finite field. The solid lines are fits to $\ln T$.

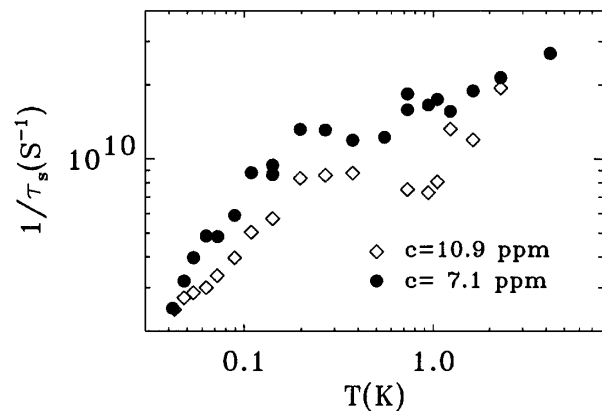


FIG. 2. Spin scattering rate $1/\tau_s$ for samples AuFe1 (diamonds) and AuFe2 (solid circles).

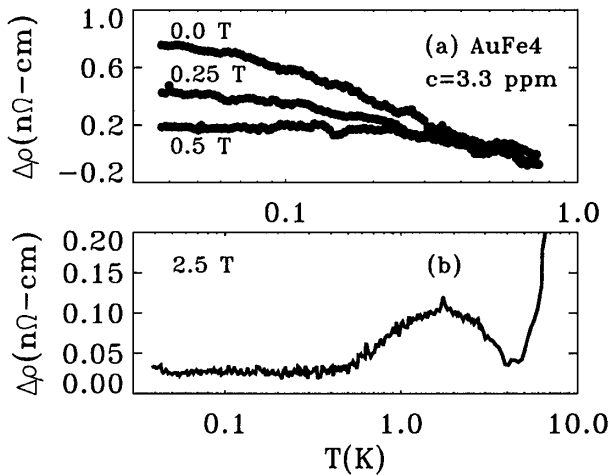


FIG. 3. (a) Resistivity saturation, and (b) the maximum in the Kondo resistivity at 2.5 T.

resistivity maximum expected for a spin glass is also not seen in Fig. 1; (d) the possibility of inhomogeneous pockets of impurities, or clustering, is ruled out by measuring different segments in a sample. The observed behavior is found to be independent of the choice of the segment, suggesting a homogeneous mechanism. For these reasons, the spin glass formation can be ruled out.

High field magnetoresistance of a representative sample, AuFe4, is shown in Fig. 5(a). WL is observed at a field scale of $B < 0.03$ T. At high fields, due to the cyclotron orbits of the electrons, a classical MR is expected: $\Delta R_c/R \sim (\omega\tau_e)^2$, with $\omega \sim eB/m$, and τ_e being the electron mean free path. This classical B^2 dependence is displayed in Fig. 5(b), which is subtracted out in Fig. 5(a). At the intermediate field scale (~ 1 T), we observe a negative magnetoresistance in all of our samples at $T < T_K$ that is very sensitive to temperature. Theoretically, in the standard Kondo model, one expects a negative MR due to the suppression of the spin-flip scattering by the alignment of the spins with the field: $\Delta R_2/R \sim (gS\beta)^2(H/T)^2$, where β is the Bohr magneton. The data is not described by this

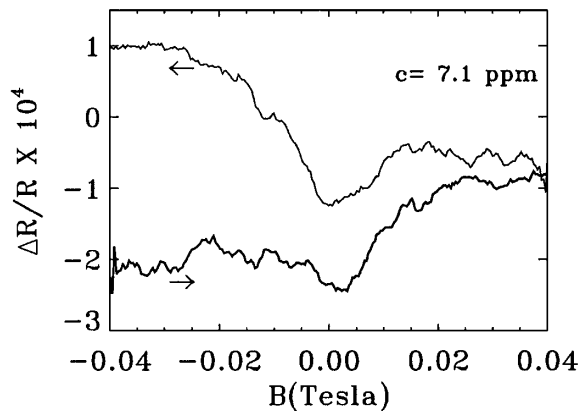


FIG. 4. Typical hysteresis observed in AuFe2 at 140 mK. Field sweep directions are marked.

contribution, as evident in the shape of the MR at various temperatures. We have observed this anomalous MR in all of our samples along with the WL dip at zero field. At 40 mK, the conductance change, $\Delta G = \Delta R/R^2$, for all of our samples in units of e^2/h is ~ 0.001 , ~ 0.002 , 0.018 , 0.028 , and 0.004 for samples AuFe1 through AuFe5, respectively.

Earlier experiments on higher concentration AuFe samples [3,4] revealed a behavior compatible with the standard expected form, and different from what we observe. Above T_K , the standard high field magnetoresistance is essentially a function of the thermal average of the local moment in the field direction $\langle S_z \rangle$. As the temperature is increased, the field scale increases with the height of the MR decreasing, ultimately becoming flat at a very high temperature due to thermal fluctuations of the localized spin. This behavior is observed in 2D Kondo films of AuFe at 1.4 and 4.2 K [3]. However, in another experiment on AuFe wires [4] with a much higher concentration of Fe impurities (~ 50 ppm), temperature dependence of the MR was not studied. There are two important characteristics of our low temperature MR, different from the bulk Kondo behavior. First, the magnetoresistance as a function of temperature cannot be explained by $\langle S_z \rangle$, since the field scale is expected to grow with increasing temperature while conserving area under the curve. Second, $\langle S_z \rangle$ as a function of temperature is

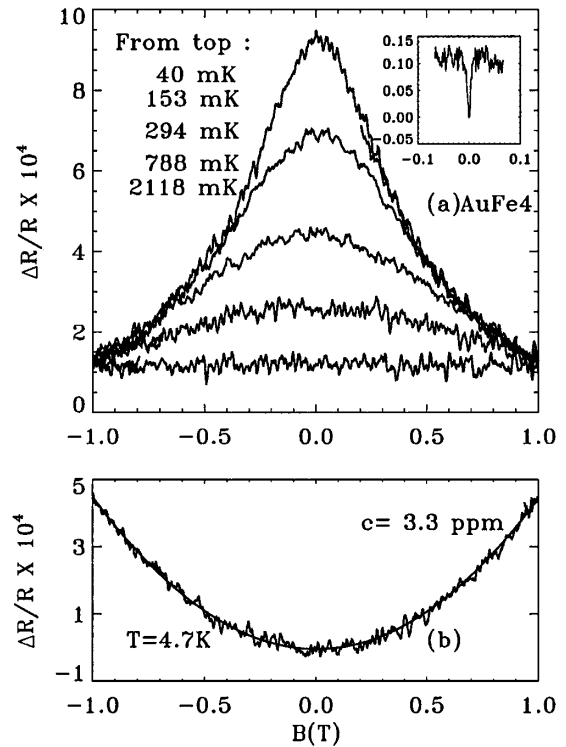


FIG. 5. (a) Magnetoresistance of a representative sample, AuFe4, after the subtraction of the classical magnetoresistance [shown in (b)]. The inset of (a) shows the weak antilocalization contribution at 520 mK.

expected to increase with decreasing temperature, becoming flat at low temperatures, whereas the dependence shown in Fig. 5(a) displays no saturation down to 40 mK. High concentrations of impurities in the earlier experiment on AuFe wires [4] and high temperature range in the experiments on 2D AuFe films [3] imply a very short τ_ϕ in these systems, yielding nonmesoscopic bulk behavior. The local magnetic field due to polarization in these high concentration samples is expected to be extremely weak, in contrast to our samples.

It is clear from our resistivity and scattering rate measurements that the long range polarization of the conduction electrons around the localized spin is effective at low temperatures for our low-concentration mesoscopic systems. From our observation of hysteresis, we believe that this polarization or the local magnetic field causes the anomalous high field magnetoresistance. Furthermore, the shape of the magnetoresistance and its temperature dependence are very much similar to what is expected from a quasi-1D interference effect [8], which suggests a similarity to weak localization. These effects were seen in long L_ϕ samples, implying an essential role played by the phase coherence of electrons. Considering all this, we propose a connection of this new interference correction to Berry phase.

It is possible for the phase coherent mesoscopic Kondo wires to show a weak-localizationlike magnetoresistance driven by a geometric phase $\Gamma = \int_{t_i}^{t_f} \mathbf{A}_g \cdot d\mathbf{R}$, similar to the standard weak localization driven by the Aharonov-Bohm phase [7]. \mathbf{A}_g is the geometric gauge potential, and \mathbf{R} is the position vector describing the tip of the spin. The spin part of the wave function of the phase coherent electron picks up a geometric phase as it aligns along the local magnetic field of the uncompensated spin. This is further helped by disorder in the sample [7], since the electron spends more time around the spin than it would in a ballistic sample. The corresponding geometric phase is equal to half of the solid angle subtended by the area enclosed by the tip of the electron spin vector due to its evolution in a closed loop. A complementary path, going in the opposite direction, will contribute an opposite phase shift. Interference of two such paths around the local field results in a correction to conductivity, analogous to the anticipated Berry phase correction in a ring structure. There are no oscillations as in the Aharonov-Bohm effect, but just half a period in resistivity, because the maximum Berry phase acquired is π , half of the maximum solid angle of 2π . An externally applied perpendicular field aligns the electron spin. If the spin is completely aligned along the external field, the solid angle subtended by the tip of the spin is zero, resulting in the complete suppression of the Berry phase correction.

Berry phase changes sign under time reversal. This leads to a contribution similar to the Cooperon propagator in

WL. Correction to the resistance contains the disorder average of all possible loops acquiring Berry phase. As temperature is increased, L_ϕ (which includes spin fluctuations) reduces greatly, thus reducing the magnetoresistance correction as seen in Fig. 5(a). This dependence is similar to that of WL. In the spirit of WL, a geometric length L_B can be introduced, which is the length scale over which the net accumulated geometric phase is on the order of π . L_B may be defined by $\Delta R_g/R^2 \sim (e^2/\hbar)L_B/L$. For the data from the sample AuFe4 [shown in Fig. 5(a)] at 40 mK, the geometric length $L_B \sim 18 \mu\text{m}$ ($L_\phi \sim 3 \mu\text{m}$ at 40 mK), implying that within L_ϕ the acquired (disorder-averaged) geometric phase is on the order of $\pi L_\phi/L_B \sim \pi/6$ for this sample.

In summary, we have observed an unusual temperature dependence of the magnetoresistance along with hysteresis in quasi-one-dimensional disordered Kondo systems at $T < T_K$. We believe that this arises from the adiabatic evolution of the phase coherent electron around the impurity spin, which results in a Berry phase effect.

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