Electron Coherence in Mesoscopic Kondo Wires

F. Schopfer, C. Bäuerle[*], W. Rabaud[†] and L. Saminadayar[‡]

Low Temperature Research Laboratory, CRTBT-CNRS, B.P. 166 X, 38042
Grenoble Cedex 09, France

Abstract. We present measurements of the magnetoresistance of long and narrow quasi one-dimensional gold wires containing magnetic iron impurities. The electron phase coherence time extracted from the weak antilocalisation shows a pronounced plateau in a temperature region of 300 mK - 800 mK, associated with the phase breaking due to the Kondo effect. Below the Kondo temperature, the phase coherence time increases, as expected in the framework of Kondo physics. At much lower temperatures, the phase coherence time saturates again, in contradiction with standard Fermi liquid theory. In the same temperature regime, the resistivity curve displays a characteristic maximum at zero magnetic field, associated with the formation of a spin glass state. We argue that the interactions between the magnetic moments are responsible for the low temperature saturation of the phase coherence time.

1 Introduction

The understanding of the ground state of an electron gas at zero temperature is one of the major challenges in Solid State Physics. For a long time it has been known that such a ground state is well described by Landau's theory of Fermi liquids [1]. In this description, the lifetime of quasiparticles is infinite at zero temperature, as the coupling to the environment tends to zero.

Alternatively, in mesoscopic physics, one key physical concept is the phase coherence time, *i.e.* the time an electron can travel in a solid before it looses its phase coherence and thus its quantum, wave like behaviour. Such a decoherence is due to inelastic processes, like electron-phonon, electron-electron or electron-photon collisions. It has been shown by Altshuler and coworkers [2] that the phase coherence time diverges at zero temperature as electron-phonon, electron-electron and electron-photon interactions all go to zero at zero temperature.

However, recent experiments on metallic as well as semiconductor wires suggest that the phase coherence time saturates at very low temperature [3]. Following this work, it has been argued that the observed saturation is indeed universal and intrinsic, and due to electron-electron interactions in the ground state of the Fermi liquid [4]. Contrary to this, other interpretations argue that this saturation is extrinsic and due to the coupling to other degrees of freedom, like two level systems [5]. On the other hand, some experimental results suggest that the dephasing depends on the dimensions of the samples [6], whereas another group argues that some of their experimental results agree with standard theory [7], at least down to 50 mK. It should be noted, however, that the problem of relaxation at zero temperature is not subject of debate in the mesoscopic community only. Recent experiments in spin polarized helium, a textbook example of a Fermi liquid, show a saturation of the transverse spin-diffusion coefficient at zero temperature [8]. This equally raises key questions about the applicability of conventional Fermi liquid theory.

Recent experiments invoke the coupling to magnetic impurities as a possible source of the frequently observed low temperature saturation of the phase coherence time [9,10,11]. It is well known that in metals the interaction of conduction electrons with magnetic impurities gives rise to the Kondo effect [12]. Concerning transport properties of metals, the best known feature of this effect is the existence of a minimum and a subsequent logarithmic increase of the resistivity with decreasing temperature below the Kondo temperature T_K. The influence of Kondo impurities on the dephasing rate, on the other hand, is by far more subtle.

Finally, it is well known that above a certain amount of impurities, and below a certain temperature, RKKY interactions between magnetic moments lead to the formation of a spin glass [13]. This regime has basically not been explored so far and may contain a great deal of new physical phenomena.

All this physics related to magnetic impurities leads to new energy scales: the Kondo temperature T_K and the spin glass transition temperature T_g. Both energy scales have to be considered when dealing with the "zero" temperature limit, and have also to be introduced in the theoretical description of dephasing in mesoscopic wires.

2 Historic

Already in the early days of weak localisation, many experimentalists observed a systematic saturation of the electron phase coherence at low temperatures, when extracted from low field magnetoresistance [14,15]. This saturation has often been attributed to the presence of some residual magnetic impurities [16].

To our knowledge, the first measurements which clearly demonstrated the strong influence of magnetic impurities on the phase coherence, even in the presence of extremely dilute magnetic impurities (below the ppm level) has been carried out by Pannetier and coworkers in the 80^{ths} [17,18]. These measurements have been performed on extremely pure Au samples and coherence lengths of several micrometers have been obtained at low temperatures. Again in these experiments, the phase coherence time was almost temperature independent below 1 Kelvin. By annealing the samples, the authors could show that the phase coherence time increases substantially. The annealing process oxidizes magnetic impurities and hence suppresses decoherence due to the

Kondo effect. These experiments therefore clearly show that the presence of an extremely small amount of magnetic impurities can lead to substantial electron decoherence at low temperatures.

A different method to suppress the effect of magnetic impurities can be achieved by applying a sufficiently high magnetic field in order to fully polarise the magnetic impurity spins. In this case, weak localisation measurements are not possible to extract the phase coherence time. On the other hand, measurements of Aharonov Bohm (AB) oscillations and universal conduction fluctuations (UCF) are possible. Pioneering work on both, UCF and AB oscillations in quasi 1D quantum conductors containing a small amount of magnetic impurities (down to 40ppm) has been performed by Benoît and coworkers in the late 80^{ths} [19]. In this work the authors could clearly show that UCF as well as AB oscillations increase considerably at fields larger than 1 Tesla, showing the suppression of the Kondo effect due to the polarization of the magnetic impurity spins. In the context of the present debate on the low temperature saturation of τ_ϕ, these measurements have been repeated recently on metallic samples containing more dilute magnetic impurities [9].

In this article we point out another effect which leads to a saturation of τ_ϕ at low temperatures when measured by weak localisation, namely the formation of a spin glass state. We show that, even in the presence of very dilute magnetic impurities, the impurities cannot be regarded as independent (single impurity limit) at low temperatures and interactions between the magnetic impurities have to be taken into account. It is well known that RKKY interactions between magnetic impurities lead to the formation of a frozen spin configuration at a characteristic temperature T_g. In systems containing very dilute magnetic impurities, this temperature lies well below the Kondo temperature, hence sets another energy scale, which can be of the order of the lowest temperatures presently accessible in experiments.

3 Experimental

In this article we report on measurements of the temperature dependence of the low field magnetoresistance and resistivity of quasi one-dimensional (1D) long and narrow Au/Fe Kondo wires down to temperatures below 0.1 T_K.

Sample fabrication is done using electron beam lithography on silicon substrate. The metal is deposited with a Joule evaporator and standard lift-off technique. In order to improve adhesion to the substrate a 1 nm thin titanium layer is evaporated prior to the gold evaporation. Two sources of 99.99% purity with different iron impurity concentrations are employed for the gold evaporation. The actual iron impurity concentration is determined *via* the resistance variation at low temperature due to the Kondo effect. Such a method directly characterises the purity of the samples, which may be quite different from the purity of the sources.

The samples (A and B) have the same geometrical parameters: their lengths, widths and thicknesses are $L = 450\mu m$, $w = 150nm$ and $t = 45nm$. The 1 K resistance value for sample A (B) is 4654Ω (2235Ω). The samples are quasi 1D with respect to both, the phase-breaking length $l_\phi = \sqrt{D\tau_\phi}$ and the thermal length $l_T = \sqrt{\hbar D/k_B T}$, D being the diffusion constant. From the relation $D = 1/3\, v_F\, l_e$, we obtain a diffusion coefficient of $5.6\, 10^{-3}\, m^2/s$ and $11.5\, 10^{-3}\, m^2/s$ for sample A and B, respectively.

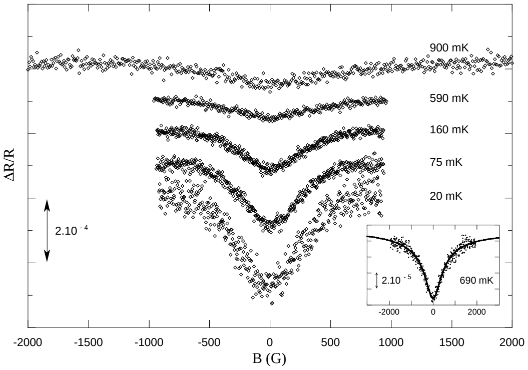

Fig. 1. Low field magnetoresistance of sample A at different temperatures. Inset shows a fit of weak localisation theory to the data

We have studied very carefully the temperature dependence of the low field magnetoresistance as well as the electrical resistivity over a temperature range extending from 4.2 K down to 15 mK. Figures 1 and 2 display the magnetoresistance at different temperatures for two samples containing different amounts of magnetic iron impurities. From fits to weak localisation theory for quasi 1D conductors (see insets) we extract the phase coherence length l_ϕ. For the fitting procedure, we first determine the spin-orbit scattering length at temperatures above 0.5 K with magnetoresistance curves covering a field span of ± 2000 G. We then fix the spin-orbit scattering length to the obtained value of 50 nm for all weak localisation fits [20]. Using the measured geometrical and electrical parameters of the samples, the only fitting parameter is hence the phase coherence length l_ϕ. From the relation $\tau_\phi = l_\phi^2/D$, we then obtain the phase coherence time τ_ϕ as shown in Fig. 3.

Three distinct temperature regimes are clearly distinguishable. At high temperatures (above 1K), the phase coherence time decreases rapidly with increasing temperature due to electron-phonon coupling. This temperature dependence is well described by a T^{-3} power law, as expected from theory. At temperatures between 0.3 K and 1 K the phase coherence time shows a pronounced plateau. Here the temperature independence of τ_ϕ is caused by dephasing due to the Kondo effect as we will see later, when we discuss the temperature variation of the resistivity. Below 0.3 K, the phase coherence time increases again, because of the partial screening of the magnetic impurities [21]. At lower temperatures, however, we again observe an apparent saturation of τ_ϕ. In order to understand this rather unusual temperature dependence of τ_ϕ, it is important to analyse the temperature dependence of the electrical resistivity.

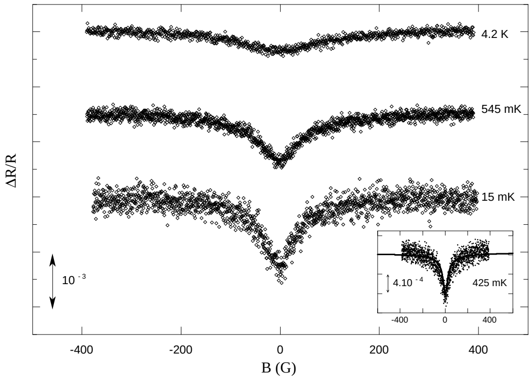

Fig. 2. Low field magnetoresistance of sample B at different temperatures. Inset shows a fit of weak localisation theory to the data

Figure 4 displays the temperature variation of the resistivity for both samples in zero magnetic field. The total contribution to the resistivity is given by three different contributions: electron-electron interaction, weak localisation and the contribution due to magnetic impurities. Below 1 K one observes an increase of the resistivity for both samples. This increase is due to the Kondo contribution and electron-electron interaction. At the lowest temperatures, one observes a clear maximum in the resistivity for both samples. Since the amplitude of the weak localisation curves is basically temperature independent at these temperatures and since the electron-electron contribution increases monotonically with decreasing temperature $\sim (1/\sqrt{T})$, the maximum in the resistivity has to be due to some other phenomenon related to the presence of magnetic impurities.

The maximum in the resistivity is a common feature for Kondo systems [22]. At low temperature, magnetic impurities interact *via* the RKKY interaction. In systems with high Kondo temperatures, and very low impurity concentration, a complete Kondo screening of the magnetic impurities can be obtained. This case is often referred to as the unitary limit, where RKKY interactions are suppressed. However, if the concentration is high enough, and the Kondo temperature low enough, the screening length may be very large as it varies like $1/T_K$. RKKY interactions are then important and the magnetic impurities cannot be treated in the single impurity limit. The unitary limit is hence never reached, and the system transits into a spin glass state at a temperature T_g. The most common features of this transition is a maximum in the resistivity curve as well as an anomaly in the magnetic susceptibility which appear roughly at T_g [23]. Both phenomena have been extensively studied in the past: the dependence of the temperature of the resistivity maximum [24,25,26] and of the susceptibility anomaly [27] as a function of the impurity concentration in Au/Fe systems as well as many others. However, the effect of such a peculiar spin configuration on the phase coherence time has not been explored so far [28]. To our knowledge, this is the first time that weak localisation measurements are accessible in this spin glass regime.

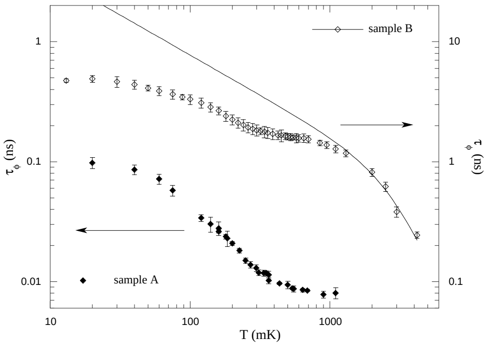

Fig. 3. Phase coherence time as a function of temperature. The solid line is the theoretical prediction from [2] for sample B

In order to determine the impurity concentration of our samples, we subtract the measured contribution due to weak localisation and fit the temperature variation of the resistivity at zero magnetic field to the following expression:

$$\rho(T) - \rho_0 = \frac{\alpha}{\sqrt{T}} + \beta\{0.743 + 0.332\,[1 - \frac{ln(T/T_K)}{ln^2 T/T_K + \pi^2 S(S+1)}]\} \qquad (1)$$

where the first term corresponds to the electron-electron contribution and the second term to the Hamann expression [29] for the Kondo contribution, with β being the impurity concentration in ppm, S the impurity spin and ρ_0 the residual resistivity at 1 K. Taking S = 3/2, $T_K = 300$ mK [24] and fitting both data sets over the same temperature range, we obtain an impurity concentration of approximately 60 ppm (15 ppm) and a coefficient $\alpha = 1.4\,\mathrm{n}\Omega\cdot\mathrm{cm}\cdot\mathrm{mK}^{-1/2}$ ($9.5\,\mathrm{n}\Omega\cdot\mathrm{cm}\cdot\mathrm{mK}^{-1/2}$) for sample A (B), compared to the theoretically expected value of $36.4\,\mathrm{n}\Omega\cdot\mathrm{cm}\cdot\mathrm{mK}^{-1/2}$ ($12.1\,\mathrm{n}\Omega\cdot\mathrm{cm}\cdot\mathrm{mK}^{-1/2}$). The poor agreement between experimental and theoretical values for coefficient α for sample A is due to our choice of fitting both sets of data over exactly the same temperature range. If we fit the data of sample A over a limited temperature range (> 100mK), we then recover the theoretically expected value for α. This proves again that in sample A, where the impurity concentration is higher than in sample B, RKKY interactions between magnetic impurities are already present at these temperatures. As a consequence, the resistivity deviates strongly from the Kondo model [30].

The saturation of τ_ϕ and the subsequent desaturation at lower temperatures can be well understood in terms of the Kondo effect. Spin flip scattering due to the presence of magnetic impurities causes very efficient dephasing at temperatures around T_K. At lower temperatures the magnetic impurities become screened by the surrounding conduction electrons and the spin flip scattering process is attenuated. As a consequence, τ_ϕ increases with decreasing temperatures [21]. At low enough temperatures standard Fermi liquid theory [31] should again describe the temperature dependence of τ_ϕ. It should therefore follow a power law $T^{-2/3}$ [2] as shown by the solid line in Fig. 3. This is clearly not the case for our experimental data.

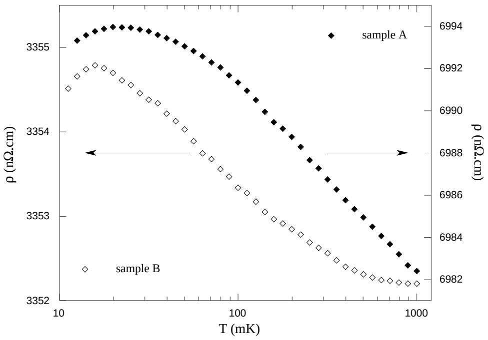

Fig. 4. Resistivity variation as a function of temperature, measured at zero magnetic field

What can be the origin of the observed low temperature saturation of τ_ϕ? One explanation, however presently very controversial, is the possible existence of zero temperature dephasing due to electron-electron interactions [4]. The agreement of the experimental data with this theory is reasonable. For a detailed comparison of our data with this theory, we refer the reader to ref. [10].

Another possibility for the low temperature saturation of τ_ϕ is the presence of another type of magnetic impurity with a Kondo temperature below the measuring temperature (*e. g.* Mn). In this case one would expect again a plateau for τ_ϕ at temperatures around T'_K, in qualitative agreement with the data on the phase coherence. If so, this additional Kondo contribution should also lead to an increase of the resistivity at low temperatures.

None of these two possibilities does explain the maximum in the resistance curve. As already mentioned above, the maximum in the resistance curve is a well known feature which is attributed to freezing of the magnetic impurities into a spin glass state. It is thus clear that RKKY interactions between magnetic impurities are important in our samples and have to be taken into account in the interpretation of our experimental data.

For this purpose, we extract the spin scattering rate from the measurement of the phase coherence time. The total dephasing time is given by [32]

$$\frac{1}{\tau_\phi} = \frac{1}{\tau_{nm}} + \frac{2}{\tau_s} \tag{2}$$

where τ_s is the spin scattering rate and τ_{nm} is the non-magnetic scattering rate given by the usual formula

$$\frac{1}{\tau_{nm}} = AT^{2/3} + BT^3 \tag{3}$$

Coefficient $\mathrm{A} = 0.8\,(0.6)\,\mathrm{ns}^{-1}\,\mathrm{K}^{-2/3}$ is calculated using the parameters of sample A (B) and coefficient $\mathrm{B} = 0.04\,(0.04)\,\mathrm{ns}^{-1}\,\mathrm{K}^{-3}$ is obtained by fitting the data at high temperature. The fit for sample B is diplayed in Fig. 3. This non-magnetic part of the dephasing time is then subtracted from our data, and we obtain the spin scattering time as a function of temperature. This is displayed in Fig. 5.

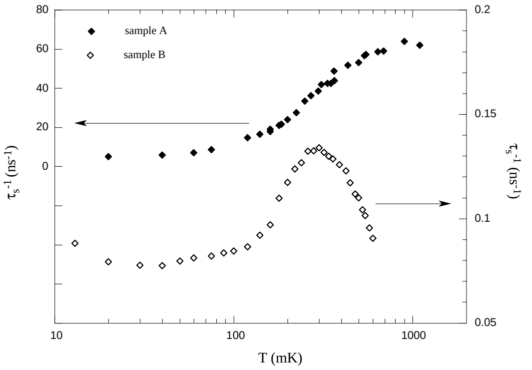

Fig. 5. Magnetic scattering rate for sample A and B obtained by subtraction of the standard dephasing rate from the data of fig. 3

Both curves exhibit a clear maximum around T_K [32,33], where the dephasing mechanism due to the Kondo effect is the most efficient. This is associated with the plateau observed around T_K in the $\tau_\phi(T)$ curve [34]. Below T_K, magnetic impurities get screened, and one expects a decrease of the dephasing time. This is indeed what is observed. Theoretical predictions lead to a T^2 behaviour in Nozières's Fermi liquid theory [31], whereas recent work leads to a $1/\ln^2(T_K/T)$ dependence for partially screened impurities [35]. None of these predictions is observed. The key point is that for both samples the spin-scattering rate saturates and is basically constant down to the lowest temperatures. This is not surprising: it is well known that spin-spin correlations have a strong influence on the measured dephasing time. Freezing of magnetic moments into a spin glass state violates the time reversal symmetry, hence leading to a very efficient dephasing mechanism. When comparing with the resistivity curves, it is obvious that this new regime appears around T_g. This saturation in the spin scattering rate can thus be associated with the formation of a spin-glass due to the RKKY interactions between magnetic impurities.

4 Conclusions

Our results clearly show that RKKY interactions, associated with the spin glass freezing, lead to a constant spin scattering rate, and hence yield a finite phase coherence time at very low temperatures. It is thus important to consider both energy scales, T_K as well as T_g when dealing with metallic systems containing even a very small amount of magnetic impurities. The understanding of electron dephasing in the temperature range below T_K is certainly a challenge for theory, but is probably the key point to interpret properly the experiments carried out on metals as they often contain magnetic impurities on the ppm level at best, with Kondo temperatures in the mK range. Measurements at lower temperatures on samples with very low concentrations of magnetic impurities, well in the unitary limit, would also be of great interest. In this case all magnetic impurities are completely screened, and the standard Fermi liquid behaviour should be recovered. This would be the key test to discriminate between intrinsic dephasing and dephasing due to Kondo impurities.

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Note added: A recent paper by Vavilov et al.[36] predicts a saturation of τ_ϕ in the presence of RKKY interactions, in agreement with our measurements. The disappearance of the resistivity maximum for low impurity concentrations ($T_{sg} \ll T_K$) as predicted by this theory, however, seems to be in contradiction with the experimental data and calls for a systematic study of the resistivity maximum as a function of the impurity concentration.

References

∗. Mail to: bauerle@grenoble.cnrs.fr
‡. Mail to: saminadayar@grenoble.cnrs.fr
†. Present address: Department of Physics, University of Maryland, College Park, MD 20742-4111, USA.

1. D. Pines and P. Nozières, *The Theory of Quantum Liquids*, W.A. Benjamin (1966).
2. B.L. Altshuler, A.G. Aronov and D.E. Khemelnitskii, J. Phys. C **15**, 7367 (1982); B.L. Altshuler and A.G. Aronov, in *Electron-Electron Interactions in Disordered Conductors*, eds. A.L. Efros and M. Pollak, North Holland, Amsterdam (1985).
3. P. Mohanty, E.M.Q. Jariwala and R.A. Webb, Phys. Rev. Lett. **78**, 3366 (1997).
4. D.S. Golubev and A.D. Zaikin, Phys. Rev. Lett. **81**, 1074 (1998); D.S. Golubev, A.D. Zaikin and G. Schön, J. of Low Temp. Phys. **126**, 1355 (2002).
5. Y. Imry, H. Fukuyama and P. Schwab, Europhys. Lett. **47**, 608 (1999); A. Zawadowski, J. van Delft and D.C. Ralph, Phys. Rev. Lett. **83**, 2632 (1999); V.V. Afonin, J. Bergli, Y.M. Galperin, V.L. Gurevich and V.I. Kozub, Phys. Rev. B. **66**, 165326 (2002).
6. D. Natelson, R.L. Willett, K.W. West and L.N. Pfeiffer, Phys. Rev. Lett. **86**, 1821 (2001).
7. F. Pierre, H. Pothier, D. Estève, M.H. Devoret, A.B. Gougam and N.O. Birge, in *Kondo Effect and Dephasing in Low-Dimensional Metallic Systems*, V. Chandrasekhar, C. van Haesendonck and A. Zawadowski eds., 119, Kluwer, Dodrecht, 2001.
8. H. Akimoto, D. Candela, J.S. Xia, W.J. Mullin, E.D. Adams and N.S. Sullivan, Phys. Rev. Lett. **90**, 105301 (2003).
9. F. Pierre and N.O. Birge, Phys. Rev. Lett. **89**, 206804 (2002).
10. F. Schopfer, C. Bäuerle, W. Rabaud and L. Saminadayar, Phys. Rev. Lett. **90**, 056801 (2003).
11. A. Anthore, F. Pierre, H. Pothier and D. Esteve, Phys. Rev. Lett. **90**, 076806 (2003).
12. J. Kondo, Prog. Theor. Phys. **32**, 37 (1964).
13. J.A. Mydosh, *Spin Glasses: An experimental introduction*, Taylor and Francis, London, 1993.
14. M.E. Gershenson, V.N. Gubankov and J.E. Juravlev, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 201 (1982).
15. D. Abraham and R. Rosenbaum, Phys. Rev. B **27**, 33 (1983).
16. G. Bergmann, Physics Reports **107**, 1 (1984) and references therein.
17. B. Pannetier, J. Chaussy, R. Rammal and P. Gandit, Phys. Rev. Lett. **53**, 718 (1984); B. Pannetier, J. Chaussy, R. Rammal and P. Gandit, Phys. Rev. B. **31**, R3209 (1985).
18. B. Pannetier, J. Chaussy and R. Rammal, Physica Scripta **T13**, 245 (1986).
19. A. Benoît, S. Washburn, C.P. Umbach, R.A. Webb, D. Mailly and L. Dumoulin, in *Anderson Localisation*, T. Ando and H. Fukuyama eds., Springer Verlag (1988); A. Benoît, D. Mailly, P. Perrier and P. Nedellec, Superlattices and Microstructures **11**, 3 (1992).

20. This is somewhat oversimplified, since the spin-orbit scattering length may vary slightly with temperature.
21. P. Mohanthy and R.A. Webb, Phys. Rev. Lett. **84**, 4481 (2000).
22. U. Larsen, Phys. Rev. B. **14**, 4356 (1976).
23. The maximum in the resistance curve is a precursor of the spin glass transition. The actual transition is situated at a slightly lower temperature than this maximum.
24. O. Laborde and P. Radhakrishna, Solid State Commun. **9**, 701 (1971); O. Laborde, PhD Thesis, Université Joseph Fourier (1977), unpublished.
25. G. Neuttiens, J. Eom, C. Strunk, V. Chandrasekhar, C. Van Hasendonck and Y. Bruynseraede, Europhys. Lett. **34**, 617 (1996).
26. J. Eom, J. Aumentado, V. Chandrasekhar, P.M. Baldo and L.E. Rehn, cond-mat/0302198.
27. G. Frossati, J.L. Tholence, D. Thoulouze and R. Tournier, Physica **84B**, 33 (1976) and references therein.
28. P.G.N. de Vegvar, L.P. Lévy and T.A. Fulton, Phys. Rev. Lett. **66**, 2380 (1991).
29. D.R. Hamann, Phys. Rev. **158**, 570 (1967).
30. If we subtract the theoretically expected contribution for e-e interactions from the total resistivity, one obtains a maximum in the resistivity at a temperature around 60 mK.
31. P. Nozières, J. of Low Temp. Phys. **17**, 31 (1974).
32. C. van Haesendonck, J. Vranken and Y. Bruynseraede, Phys. Rev. Lett. **58**, 1968 (1987).
33. R.P. Peters, G. Bergmann and R.M. Mueller, Phys. Rev. Lett. **58**, 1964 (1987).
34. The saturation value of τ_ϕ around the Kondo temperature differs very much for the two samples. This is somewhat surprising since the impurity concentration is only by a factor of 4 different when extracted from the resistivity. The determination of the impurity concentration via the phase coherence time at around T_K [32] would lead to an impurity concentration of less than 1ppm for sample B. This is clearly incompatible with the observed resistance variation as well as the position of the maximum in the R(T) curve, which is in relatively good agreement with data on the bulk Au/Fe Kondo system (see ref. [24]). This discrepancy is presently not understood.
35. M.G. Vavilov and L.I. Glazman, Phys. Rev. B. **67**, 115310 (2003).
36. M.G. Vavilov, L.I. Glazman and A.I. Larkin, condmat/0305240.