

Experimental Test of the Numerical Renormalization-Group Theory for Inelastic Scattering from Magnetic Impurities

Christopher Bäuerle,¹ François Mallet,¹ Félicien Schopfer,^{1,*} Dominique Mailly,²
Georg Eska,³ and Laurent Saminadayar¹

¹Center for Low Temperature Research, CRTBT-CNRS, BP 166 X, 38042 Grenoble Cedex 9, France

²Laboratoire de Photonique et Nanostructures, route de Nozay, 91460 Marcoussis, France

³Physikalisches Institut, Universität Bayreuth, Universitätsstrasse 30, 95440 Bayreuth, Germany

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We present measurements of the phase coherence time τ_ϕ in quasi-one-dimensional Au/Fe Kondo wires and compare the temperature dependence of τ_ϕ with a recent theory of inelastic scattering from magnetic impurities [Phys. Rev. Lett. **93**, 107204 (2004)]. A very good agreement is obtained for temperatures down to $0.2T_K$. Below the Kondo temperature T_K , the inverse of the phase coherence time varies linearly with temperature over almost one decade in temperature.

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The Kondo problem has been fascinating physicists for more than 30 years. Such interest is due to the fact that this model, first introduced to describe transport properties of metals containing magnetic impurities, is a generic model for the description of many solid state physics phenomena [1]. Concerning magnetic alloys, the “historical” Kondo solution first allowed one to describe the transport properties at temperatures larger than the characteristic temperature of the model, the Kondo temperature T_K [2]. The major breakthrough has been the pioneering work of Wilson, who was able to calculate the ground state of the Kondo problem at all temperatures using his numerical renormalization group (NRG) [3]. Another important contribution was due to Nozières, who showed that the zero temperature limit of the Kondo model can be described within Landau’s Fermi liquid theory [4], which made precise predictions for transport as well as thermodynamic properties. Since then, many experiments have confirmed the relevance of this approach.

Recently, it has been suggested that scattering by magnetic impurities [5,6] is responsible for the experimentally observed saturation of the phase coherence time τ_ϕ of electrons in metals at low temperatures [7], renewing the interest in Kondo physics. Though the Altshuler-Aronov-Khmelnitzky (AAK) theory describes the temperature dependence of the phase coherence time of electrons in pure metals [8], no exact solution is available for the phase coherence time in the presence of Kondo impurities. Only a high temperature expansion, the Nagaoka-Suhl (NS) expression, was able to describe the experimental data at temperatures $T \gg T_K$ [9]. In the opposite limit ($T \ll T_K$), Fermi liquid theory predicts a T^2 dependence of the inelastic scattering rate [4]. This, however, has never been observed experimentally. Only very recently, Zaránd and co-workers have been able to obtain an exact solution for the inelastic scattering time in Kondo metals using Wilson’s NRG [10]. This calculation constitutes a major breakthrough for the decoherence problem, as it allows one

to compare experimental data with theoretical results for all temperatures, ranging from well above T_K down to zero temperature. It is, thus, of crucial importance to check whether this theory can describe correctly experimental data, as it could give new insights into the problem of the low temperature decoherence in mesoscopic metallic wires.

In this Letter, we compare this recent theory with experimental results and show that the NRG calculation describes very well the experimental data at temperatures around and below the Kondo temperature. In the first part of this Letter, we analyze the phase coherence time of quasi-one-dimensional gold wires containing magnetic iron impurities of Ref. [11], denoted as AuFe1, and Ref. [12], denoted as AuFe2, whereas in the second part, we use the NRG calculation to interpret the phase coherence time as a function of temperature of an extremely pure gold sample (Au1). In Table I, we list the geometrical and electrical parameters of these samples.

The phase coherence time of sample AuFe1 and AuFe2, as extracted from standard weak localization measurements, is shown in Fig. 1. Both samples display a distinct plateau at a temperature above $T \geq 0.3$ K. This plateau is caused by Kondo spin flip scattering due to the presence of iron impurities which leads to dephasing. Decreasing the temperature, the magnetic impurity spin is screened, and, as a result, the phase coherence time increases again.

TABLE I. Sample characteristics: w , t , l , R , correspond to the width, thickness, length, and electrical resistance, respectively. D is the diffusion coefficient and c_{imp} is the impurity concentration extracted from the NRG fits.

Sample	w (nm)	t (nm)	l (μm)	R (Ω)	D (cm^2/s)	c_{imp} (ppm)
AuFe1	180	40	155	393	200	3.3
AuFe2	150	45	450	4662	56	45
Au1	120	50	450	1218	241	<0.015

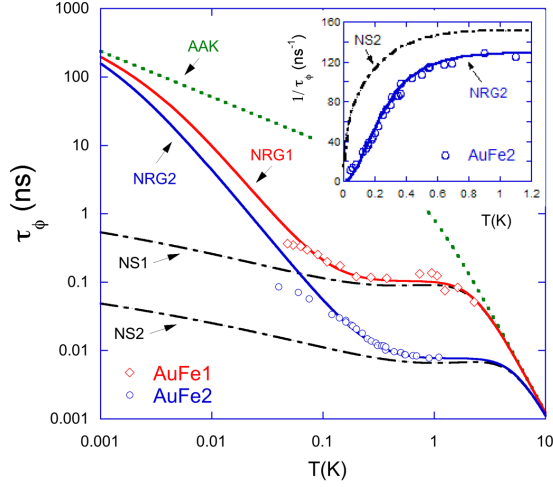


FIG. 1 (color online). Phase coherence time as a function of temperature for two Au/Fe Kondo wires: \diamond sample AuFe1 of Ref. [11], \circ sample AuFe2 (sample B) of Ref. [12]. The green dotted line (denoted AAK) corresponds to the assumption that only electron-electron and electron-phonon interactions contribute to dephasing. The black dashed-dotted lines (NS1 and NS2) take, in addition, account for the magnetic scattering using the NS expression, whereas for the red (NRG1) and blue (NRG2) solid lines, the NRG calculation has been employed for the contribution of the magnetic impurities. The inset shows $1/\tau_\phi$ versus T on a linear scale for sample AuFe2 in order to emphasize the *linear* regime.

The temperature dependence of the measured phase coherence time τ_ϕ in the presence of magnetic impurities can be described in the following way:

$$\frac{1}{\tau_\phi} = \frac{1}{\tau_{e-e}} + \frac{1}{\tau_{e-ph}} + \frac{2}{\tau_{\text{mag}}}, \quad (1)$$

where $1/\tau_{e-e} = a_{\text{theo}} T^{2/3} = [e^2 R \sqrt{D} k_B / 2^{3/2} \hbar^2 L]^2 T^{2/3}$ corresponds to the electron-electron interaction term [13], $1/\tau_{e-ph} = b T^3$ to the electron-phonon interaction, while $1/\tau_{\text{mag}}$ correspond to the contribution due to magnetic impurities. To account for the scattering off magnetic impurities, we use, on one hand, the common Nagaoka-Suhl expression [9,14]

$$\frac{1}{\tau_{\text{mag}}} = \frac{1}{\tau_{\text{NS}}} = \frac{c_{\text{imp}} [\text{ppm}]}{0.6 [\text{ns}] (\pi^2 S(S+1) + \ln^2(T/T_K))}, \quad (2)$$

where S is the impurity spin, c_{imp} the impurity concentration expressed in parts per million (ppm), and the prefactor of 0.6 [ns] has been calculated taking the electron density of gold [6]. On the other hand, we use the inelastic scattering rate calculated by NRG [10]

$$\frac{1}{\tau_{\text{mag}}} = \frac{1}{\tau_{\text{inel(NRG)}}} = A \frac{\sigma(w)_{\text{inel}}}{\sigma_0} c_{\text{imp}}, \quad (3)$$

where $\sigma(w)_{\text{inel}}$ is the inelastic scattering cross section at finite energy w , $\sigma_0 = 4\pi/k_F^2$ the elastic scattering cross section at zero temperature, and A a numerical constant in

units of $[\text{s}^{-1}]$ to express the impurity concentration c_{imp} in ppm.

The green dotted line in Fig. 1, denoted as AAK, corresponds to the assumption that only electron-electron and electron-phonon interactions contribute to the electron dephasing and that there is no other mechanism for decoherence at low temperatures. For the simulation, we have used $a_{\text{theo}} = 0.41 \text{ ns}^{-1} \text{ K}^{-2/3}$ and $b = 0.8 \text{ ns}^{-1} \text{ K}^{-3}$. Keeping these values fixed, and adding the contribution due to magnetic impurities using the NS expression for $S = 1/2$, we obtain the black dashed-dotted lines for samples AuFe1 and AuFe2, denoted as NS1 and NS2. The NS expression describes relatively well the observed temperature dependence of τ_ϕ at temperatures above T_K but fails to describe the desaturation of τ_ϕ due to the screening of the magnetic impurities at temperatures $\leq T_K$. This is not at all surprising, since the NS expression results from a perturbative expansion in (T/T_K) and breaks down for $T \rightarrow T_K$. Instead, taking the inelastic scattering rate obtained by NRG, we obtain a very good agreement with the experimental data in basically the entire temperature range as shown by the red (NRG1) and blue (NRG2) solid lines for samples AuFe1 and AuFe2, respectively. For the fitting procedure, we have adjusted the magnetic impurity concentration such that the NS expression and the NRG calculation coincide at high temperatures ($T > 10T_K$). One clearly sees that the NS expression deviates already from the NRG data at relatively high temperatures ($T \sim 5 \text{ K}$) as already pointed out in Ref. [10], and fitting of experimental data with this expression at temperatures $T < 10T_K$ should, therefore, be avoided. From the NRG fitting procedure, we obtain an impurity concentration of approximately 3.3 ppm and $T_K = 0.4 \text{ K} \pm 0.05 \text{ K}$ for sample AuFe1 and 45 ppm and $T_K = 0.9 \text{ K} \pm 0.05 \text{ K}$ for sample AuFe2 [15], in good agreement with T_K values for Au/Fe found in the literature [17]. It is worth mentioning that the measurement of the phase coherence time is a very precise method to determine the Kondo temperature as well as the magnetic impurity concentration. In resistance measurements, the determination of the Kondo temperature is not straightforward, since the entire temperature range down to the unitary limit ($T \ll T_K$) is necessary to extract the Kondo temperature with a satisfactory precision. The NRG fitting procedure of τ_ϕ on the contrary allows one to extract the Kondo temperature with high precision if temperatures only slightly lower than T_K are attained.

At the lowest temperatures, we observe deviations from the NRG theory. A probable explanation for the deviation is the fact that the spin of Fe in Au is not exactly 1/2 and the impurity spin might not be completely screened. This eventually leads to interactions of magnetic impurities and a saturation of τ_ϕ [12]. Another possibility is the presence of another magnetic impurity with a much lower Kondo temperature. This latter issue will be discussed in the last part of this Letter.

The temperature evolution of τ_ϕ below T_K deserves several comments: After a slow increase of τ_ϕ , the temperature dependence is almost *linear* in temperature over almost one decade in temperature, as emphasized in the inset in Fig. 1. This relatively weak temperature dependence explains why the pioneering experiments [14,16] have not succeeded in observing the Fermi liquid regime. Comparing the experimental results with the NRG calculation, we see clearly that the Fermi liquid regime can be reached only for temperatures typically below $0.01T_K$. Moreover, the AAK behavior is recovered only at extremely low temperatures ($T < 0.001$ K).

It is noteworthy that the calculated quantity by NRG is σ_{inel} and not the phase coherence time τ_ϕ measured in a weak localization experiment. In fact, σ_{inel} has been calculated in the limit of zero temperature and finite energy $\sigma_{\text{inel}}(w, T = 0)$, whereas in a transport experiment one measures $\sigma_{\text{inel}}(w = 0, T)$. The fact that the numerical results describe well the experimental data lets us conclude that these two quantities are not very different, at least for $k_B T \ll \epsilon_F$, ϵ_F being the Fermi energy.

Having now a theory which satisfactorily describes the temperature dependence of τ_ϕ in the presence of magnetic impurities, let us reexamine the temperature dependence of τ_ϕ in extremely pure gold wires. In a recent article [6], the deviation of τ_ϕ from the AAK prediction at very low temperatures has been assigned to the presence of an extremely small amount of magnetic impurities (typically on the order of 0.01 ppm). For this purpose, we have fabricated an extremely pure gold wire (Au1) as shown in the inset in Fig. 2. The fabrication procedure is essentially the same as for the Au/Fe wires, with the only difference that the wire has been evaporated in an evaporator which is exclusively used for the evaporation of extremely pure gold. The gold of purity 5N5 has been evaporated directly on a silicon wafer *without* a sticking layer. In addition, special care has

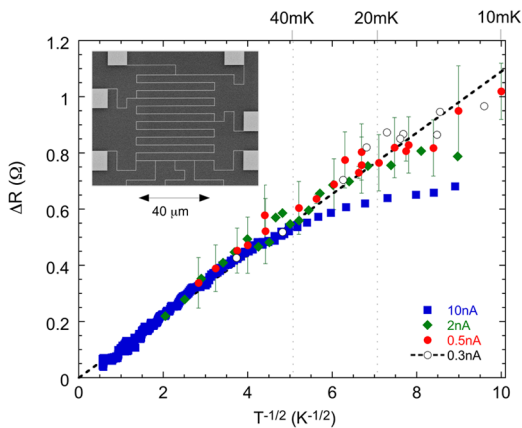


FIG. 2 (color online). Resistance variation of sample Au1 plotted as a function of $1/\sqrt{T}$ for different bias currents. The dotted line corresponds to the theoretical expectation for the resistance correction. The inset shows a scanning electron microscope photograph of the gold wire.

been taken for the sample design such that there is no influence on the phase coherence due to the two-dimensional contact pads (see inset in Fig. 2). For this wire, the phase coherence length at the lowest temperatures is more than $20 \mu\text{m}$. To our knowledge, this is the largest coherence length ever obtained in a metallic wire and confirms the high purity of the sample.

To determine the effective electron temperature of this sample, we have measured the Altshuler-Aronov correction to the resistivity at very low temperatures. A magnetic field of 40 mT has been applied in order to suppress weak localization correction to the resistivity. In Fig. 2, we plot the resistance correction as a function of $1/\sqrt{T}$. For measuring currents below 0.7 nA, the sample is in thermal equilibrium ($eV < kT$) in the entire temperature range, and the resistance correction follows the expected $1/\sqrt{T}$ temperature dependence down to 10 mK. This shows clearly that the electrons of a mesoscopic sample can be cooled to such low temperatures. Fitting the temperature dependence of the resistance correction to $\Delta R(T) = \alpha_{\text{exp}}/\sqrt{T}$ (dotted line in Fig. 2), we determine α_{exp} and compare it to the predicted value [13] of $\Delta R(T) = 2R^2/R_K L_T/L = \alpha_{\text{theo}}/\sqrt{T}$, where $L_T = \sqrt{\hbar D/k_B T}$ is the thermal length and $R_K = h/e^2$. We obtain a value $\alpha_{\text{exp}} = 0.11 \Omega/\text{K}^{1/2}$, which is in very good agreement with the theoretical value of $\alpha_{\text{theo}} = 0.109 \Omega/\text{K}^{1/2}$.

The phase coherence time τ_ϕ is then measured via standard weak localization (see inset in Fig. 3) and the phase coherence length l_ϕ is extracted via the Hikami-Larkin-Nagaoka formula [18]. From the relation $\tau_\phi = l_\phi^2/D$, we then calculate the phase coherence time as displayed in Fig. 3. We fit the experimental data with the AAK expression such that an almost perfect agreement is

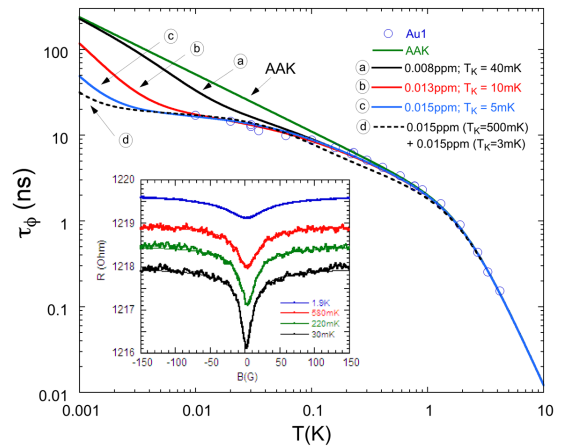


FIG. 3 (color online). Phase coherence time as a function of temperature for sample Au1 (\circ). The solid green line corresponds to the AAK prediction, the black (a), red (b), and blue (c) solid lines correspond to the NRG calculation assuming $T_K = 40$ mK, $T_K = 10$ mK, and $T_K = 5$ mK, respectively. The inset shows typical magnetoresistance curves at different temperatures.

obtained at high temperatures ($T > 100$ mK), as shown by the green solid line. The prefactor we extract from this fit $a_{\text{fit}} = 0.42 \text{ ns}^{-1} \text{ K}^{-2/3}$ is in very good agreement with the theoretical prediction of $a_{\text{theo}} = 0.41 \text{ ns}^{-1} \text{ K}^{-2/3}$. At temperatures below 100 mK, our data deviate substantially from the AAK prediction. To see whether these deviations can be explained by the presence of a very small amount of magnetic impurities, we simulate the temperature dependence of τ_ϕ for the presence of a small amount of magnetic impurities using the NRG calculations. The black (a), red (b), and blue (c) solid lines correspond to a simulation assuming $T_K = 40$ mK, $T_K = 10$ mK, and $T_K = 5$ mK, with an impurity concentration of $c_{\text{imp}} = 0.008$ ppm, $c_{\text{imp}} = 0.013$ ppm, and $c_{\text{imp}} = 0.015$ ppm, respectively. It is clear from our simulations that only magnetic impurities with a Kondo temperature $T_K \leq 10$ mK and with a concentration smaller than 0.015 ppm describe satisfactorily the experimental data. A possible magnetic impurity with a Kondo temperature in this temperature range is Mn ($T_K \simeq 3$ mK) [19].

For the sake of objectiveness, let us point out, however, the following: Assuming magnetic impurities with a Kondo temperature below the measuring temperature leads to an almost temperature independent scattering rate for $T \geq T_K$. Any experimentally observed saturation of τ_ϕ can, therefore, always be assigned to magnetic impurities with an unmeasurably low Kondo temperature. One could also argue that it is curious that, for the case of gold wires, the observed temperature dependence of τ_ϕ can be described satisfactorily only by assuming the presence of one specific magnetic impurity with a Kondo temperature below the measuring temperature $T \leq 10$ mK, whereas it is known that the dominant magnetic impurity in gold is iron. If we assume an additional iron concentration (0.015 ppm; $T_K = 500$ mK) of the same order as, for instance, Mn (0.015 ppm; $T_K = 3$ mK), the temperature dependence of τ_ϕ does not satisfactorily describe the experimental data as displayed by the dotted black line (d) in Fig. 3. A possible explanation of these facts, might be the presence of a distribution of Kondo temperatures, which could be significant for very diluted Kondo impurities [20]. Such a distribution of Kondo temperatures has already been seen in point contact experiments [21]. This, however, can be verified only with phase coherent measurements at high magnetic fields [22]. We also note that our present results do not allow us to rule out the predictions of Ref. [23].

In conclusion, we have shown that the NRG calculation of the inelastic scattering rate describes very well the experimentally observed temperature dependence of τ_ϕ caused by the presence of magnetic impurities. Below T_K , the inverse of the phase coherence time varies basically linearly with temperature over almost one decade in temperature. The T^2 temperature dependence predicted by the Fermi liquid theory, on the other hand, can be reached only for temperatures smaller than $0.01T_K$ and remains an experimental challenge.

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Note added in proof.—An exact calculation of τ_ϕ in the presence of disorder supports the above findings [24].

*Present address: Bureau National de Métrologie, Laboratoire National d’Essais, 29 avenue Roger Hennequin, 78197 Trappes, France.

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