

## Magnetic-Field-Dependent Quasiparticle Energy Relaxation in Mesoscopic Wires

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In order to find out if magnetic impurities can mediate interactions between quasiparticles in metals, we have measured the effect of a magnetic field  $B$  on the energy distribution function  $f(E)$  of quasiparticles in two silver wires driven out of equilibrium by a bias voltage  $U$ . In a sample showing sharp distributions at  $B = 0$ , no magnetic field effect is found, whereas, in the other sample, rounded distributions at low magnetic field get sharper as  $B$  is increased, with a characteristic field proportional to  $U$ . Comparison is made with recent calculations of the effect of magnetic-impurities-mediated interactions taking into account Kondo physics.

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The understanding of the phenomena which, at low temperature, limit the extent of quantum coherence in electronic transport and allow the quasiparticles to exchange energy is presently an important issue in mesoscopic physics. There is indeed a discrepancy between the theory [1], which predicts that Coulomb interactions provide the dominant mechanism for decoherence and for energy exchange, and measurements of the coherence time [2,3] or of energy exchange rates [4–7] in numerous metallic samples. This discrepancy has been attributed either to a flaw in the theory [2], or to the presence in these samples of other mechanisms involving the scattering of electrons by undetected two-level systems or magnetic impurities. It has been indeed recently predicted that even a minute concentration of such scatterers would result in sizable energy exchange if the Kondo effect occurs [8–10]. Whereas the limitation of quantum coherence by the Kondo effect is widely known [11], its efficiency for mediating energy exchange between quasiparticles had not been anticipated. In the case of magnetic impurities, a significant weakening of this effective electron-electron interaction is furthermore predicted when a large magnetic field is applied [12]. In order to test these new predictions and more generally to understand inelastic processes in mesoscopic conductors, we have investigated the magnetic field dependence of the energy exchange rate in mesoscopic wires.

The samples are wires connected to reservoirs biased at potentials 0 and  $U$  (see Fig. 1). The energy distribution function in the middle of the wire,  $f(E)$ , depends on the ratio of the typical interaction time  $\tau_{\text{int}}$  and the diffusion time of quasiparticles  $\tau_D = L^2/D$ . If  $\tau_{\text{int}} \gg \tau_D$ , interactions can be neglected and  $f(E)$  is the average of the Fermi functions in both reservoirs, which have electrochemical potentials shifted by  $eU$ . In the experimental situation where  $k_B T \ll eU$ ,  $f(E)$  is then a two-step function. In the opposite limit  $\tau_{\text{int}} \ll \tau_D$ , local equilibrium is achieved at each coordinate along the wire, and  $f(E)$  is a Fermi function at a temperature given by the balance between Joule heating and electronic heat conductivity

to the reservoirs: This is the “hot-electron” regime [13]. The intermediate regime is of interest for experiments because the precise shape of  $f(E)$  and its dependence on  $U$  are characteristic of the interaction rate and of its energy dependence [4].

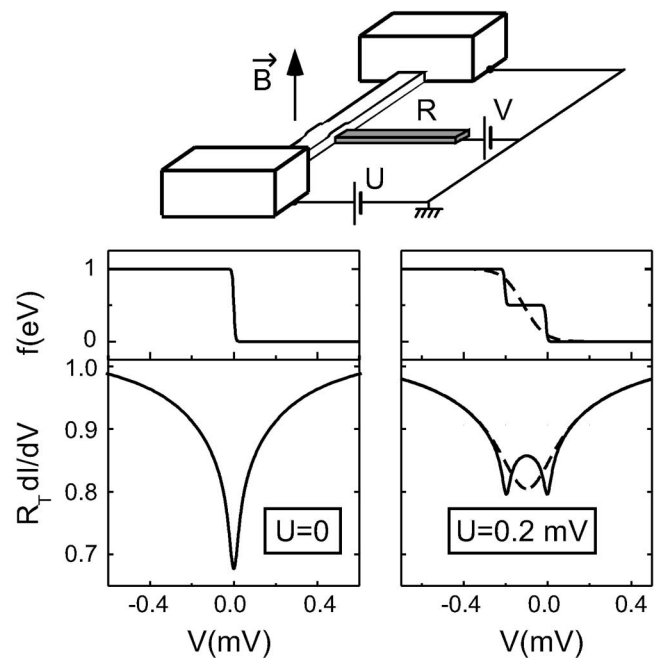


FIG. 1. Top: Layout of the experiment: A wire is connected to two large electrodes biased at potentials 0 and  $U$ . A resistive probe electrode (in grey) forms a tunnel junction with the wire. At equilibrium ( $U = 0$ ), the differential conductance  $dI/dV(V)$  of this junction displays a dip at zero bias, due to Coulomb blockade of tunneling (left). When the quasiparticles of the wire are driven out of equilibrium by a finite voltage  $U$  (right), their energy distribution function  $f(E)$  depends on the interaction rate between quasiparticles. In the absence of interactions,  $f(E)$  is a two-step function and  $dI/dV(V)$  presents two dips (solid lines). With strong interactions,  $f(E)$  is rounded, and  $dI/dV(V)$  presents a broad dip (dashed lines).

At zero magnetic field, the distribution function  $f(E)$  can be inferred from the differential conductance  $dI/dV(V)$  of a tunnel junction between the central part of the wire and a superconducting (aluminum) probe electrode biased at potential  $V$  [4]. In magnetic fields larger than the critical field  $B_c \sim 0.1$  T of the superconducting electrode, another method is required. Here, we have taken advantage of the nonlinearity of the current-voltage characteristic of a tunnel junction placed in series with a resistance  $R$ . When both electrodes of the junction are in the normal state and at thermal equilibrium, the differential conductance  $dI/dV(V)$  displays a dip at  $V = 0$  (see Fig. 1), due to the Coulomb blockade of tunneling [14]. Assuming that the two electrodes have different distribution functions  $f$  and  $f_{\text{ref}}$ , the differential conductance reads

$$\begin{aligned} \frac{dI}{dV}(V) = & \frac{1}{R_T} \int dE f(E) \int d\varepsilon P(\varepsilon) \\ & \times \frac{\partial}{\partial E} [f_{\text{ref}}(E + eV + \varepsilon) - f_{\text{ref}}(E + eV - \varepsilon)], \end{aligned} \quad (1)$$

where  $R_T$  is the tunnel resistance of the junction, and  $P(\varepsilon) = \int \frac{dI}{2\pi\hbar} e^{J(t)+i\varepsilon t/\hbar}$  the probability for an electron to tunnel through the barrier while releasing to the environment an energy  $\varepsilon$ ,  $J(t) = \int \frac{d\omega}{\omega} \{2\text{Re}[Z(\omega)]/R_K\} [(e^{-i\omega t} - 1)/(1 - e^{-\hbar\omega/k_B T})]$  with  $Z(\omega) = 1/(1/R + jC\omega)$ ,  $C$  the junction capacitance,  $R_K = h/e^2 \approx 25.8$  k $\Omega$  the resistance quantum, and  $T$  the environment temperature. In the case where the distribution function  $f(E)$  presents two steps, as in Fig. 1, and  $f_{\text{ref}}$  is a Fermi function at temperature  $T$ , one obtains, by linearity, two dips in  $dI/dV(V)$  at  $V = 0$  and  $V = -U$ . In contrast, in the hot electron regime,  $dI/dV(V)$  displays a broad dip centered at  $V = -U/2$  (see Fig. 1). In the experiments, a large series impedance at the relevant frequencies (up to about 50 GHz) was obtained by designing the probe electrode as a long, narrow, and thin aluminum electrode (25  $\mu\text{m} \times 150$  nm  $\times$  12 nm), which presents a resistance  $R \sim 1.5$  k $\Omega$  in the normal state.

We present here the results obtained on two silver samples in which the distribution functions found at  $B = 0$  were extremely different. The samples were obtained from nominally five-nines purity (99.999%, sample No. 1) and six-nines purity (99.9999%, sample No. 2) source material. For both wires, the length and cross-section area are  $L = 20$   $\mu\text{m}$ ,  $S = 100$  nm  $\times$  48 nm. The diffusion constants  $D = 196$  and 215 cm<sup>2</sup>/s, respectively, were deduced from the low temperature resistance. The tunnel resistances  $R_T$  (167 and 102 k $\Omega$ ) and the capacitances  $C$  (0.8 and 0.9 fF) of the junctions, as well as the environment resistances  $R$  (1.34 and 1.65 k $\Omega$ ), were obtained from fits with Eq. (1) of  $dI/dV(V)$  measured at  $B = 0.3$  T and  $U = 0$ . We have checked that these curves do not change with  $B$  when  $B > B_c$ .

At low magnetic field and low temperature, the probe electrode is superconducting. Its impedance is purely imaginary at frequencies lower than  $2\Delta/h$  [15]. It results that for  $eV \in [-3\Delta + U, 3\Delta]$  Coulomb blockade leads only to a reduction of the differential conductance, which is multiplied by a factor  $\exp(-\int_0^\infty \frac{d\omega}{\omega} \{2\text{Re}[Z(\omega)]/R_K\}) \sim 0.9$ . Numerical deconvolution of  $dI/dV(V)$  is therefore possible, and the distribution functions obtained at  $U = 0.15$  mV are presented in the top of Fig. 2 for both samples. Whereas  $f(E)$  is close to a double-step function in sample No. 2, it is much more rounded in sample No. 1, indicating that the energy exchange rate is much larger in the latter, since the diffusion times are very similar ( $\tau_D = L^2/D \approx 20$  ns). In the bottom of Fig. 2, we plot the calculated  $R_T dI/dV(V)$  using formula (1) with  $f(E)$  the distribution function measured at  $B = 0$  (dashed curves), and present the measured curves for  $B = 0.3$  T and  $B = 1.2$  T (symbols) [16]. In sample No. 2, the magnetic field has no visible effect. Note, however, that the distribution functions are so close to a double step that the experiment is not sensitive enough to detect a possible slight reduction of the energy exchange rate with  $B$ . In contrast, in sample No. 1, the rounded dip at zero field is replaced at 1.2 T by a double dip, showing that the energy exchange rate has been reduced. Figure 3 shows the evolution of  $dI/dV(V)$  with magnetic field, from 0.3 to 1.5 T by steps of 0.3 T, for  $U = 0.1, 0.2,$  and 0.3 mV. A similar behavior is observed at all values of  $U$ : The low-field broad conductance dip at  $B = 0.3$  T tends to be

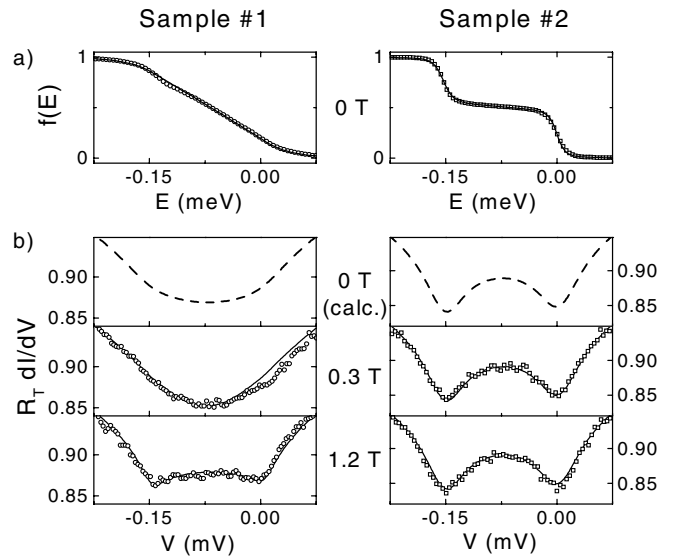


FIG. 2. (a) Symbols: Distribution functions  $f(E)$  at  $U = 0.15$  mV and zero magnetic field in samples No. 1 and No. 2, obtained by deconvolution of  $dI/dV(V)$  with the probe electrode in the superconducting state. Solid lines: Fits with theory including the effect of Kondo impurities (see text). (b) Dashed line: Calculated Coulomb blockade signal  $dI/dV(V)$  using the measured  $f(E)$  at  $B = 0$ . Symbols: Measured  $dI/dV(V)$  at  $U = 0.15$  mV, with  $B = 0.3$  and 1.2 T, the probe electrode being in the resistive state. Solid line: Fits with theory.

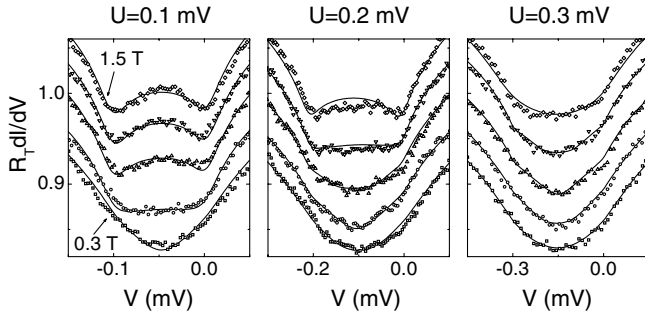


FIG. 3. Magnetic field effect in sample No. 1: differential conductance  $dI/dV(V)$  at  $U = 0.1, 0.2$  and  $0.3$  mV, for  $B$  ranging (from bottom to top) from  $0.3$  to  $1.5$  T by steps of  $0.3$  T. Successive curves have been vertically offset by steps of  $0.033$ , for clarity.

replaced at large fields by a double-dip structure. In particular, the crossover field at which  $dI/dV(V)$  is nearly constant over a broad voltage range is  $0.6$  T at  $U = 0.1$  mV,  $0.9$  T at  $U = 0.15$  mV (not shown),  $1.2$  T at  $U = 0.2$  mV, and  $1.5$  T at  $U = 0.25$  mV (not shown), hence, presenting a linear increase with  $U$ . The comparison of the raw data on sample No. 1 and sample No. 2 in Fig. 2 already allows one to conclude that sample No. 1 presents an extra interaction which can be strongly reduced by applying a magnetic field.

We now compare the experimental data with theoretical predictions. The distribution function is calculated by solving the stationary Boltzmann equation in the diffusive regime [18,19]:

$$\frac{1}{\tau_D} \frac{\partial^2 f(x, E)}{\partial x^2} = -I_{\text{coll}}^{\text{in}}(x, E, \{f\}) + I_{\text{coll}}^{\text{out}}(x, E, \{f\}), \quad (2)$$

where  $I_{\text{coll}}^{\text{in}}$  and  $I_{\text{coll}}^{\text{out}}$  are the rates at which quasiparticles are scattered in and out of a state at energy  $E$  by inelastic processes. Assuming that the dominant inelastic process is a two-quasiparticle interaction which is local on the scale of variations of the distribution function,

$$I_{\text{coll}}^{\text{in}} = \int d\varepsilon dE' K(\varepsilon) f_{E+\varepsilon}^x (1 - f_E^x) f_{E'}^x (1 - f_{E'-\varepsilon}^x), \quad (3)$$

where the shorthand  $f_E^x$  stands for  $f(x, E)$ . The out-collision term  $I_{\text{coll}}^{\text{out}}$  has a similar form. The kernel function  $K(\varepsilon)$  is proportional to the averaged squared interaction between two quasiparticles exchanging an energy  $\varepsilon$ . Coulomb interactions lead, in diffusive wires, to  $K(\varepsilon) = \kappa/\varepsilon^{3/2}$  [1], where  $\kappa = (\pi\sqrt{D}/2\hbar^{3/2}\nu_F S)^{-1}$  with  $\nu_F$  the density of states at the Fermi level [20]. The  $B = 0$  data for sample No. 2 can be well fit using this term with  $\kappa = 0.12$  ns $^{-1}$  meV $^{-1/2}$ , of the same order of magnitude as the theoretical value  $0.07$  ns $^{-1}$  meV $^{-1/2}$  [21], and a term of lesser importance describing phonon emission [22]. The  $B = 0$  data for sample No. 1 can be fit similarly, with  $\kappa = 2.4$  ns $^{-1}$  meV $^{-1/2}$ ; however, the reduction of the energy exchange rate with  $B$  indicates that an extra process is present at  $B = 0$ . We have in the

following fixed  $\kappa$  to the best fit value obtained for the large field, low  $U$  data, where the  $B$  dependent interaction has essentially vanished:  $\kappa = 0.5$  ns $^{-1}$  meV $^{-1/2}$  [21]. The remaining part of the energy exchange rate was fit with the theory of Göppert *et al.* [12,23], which accounts for the effective interaction in the presence of a concentration  $c$  of spin- $\frac{1}{2}$  impurities, with Kondo temperature  $T_K$ , gyromagnetic factor  $g$ , and coupling constant  $J$  between quasiparticles and magnetic impurities. The Kondo effect is included in this calculation, under the assumption that the distribution functions are not too sharp, leading to a renormalization of  $J$  depending on the distribution function itself. The corresponding inelastic integral can also be written in the form of Eq. (3), but with a  $K(\varepsilon)$  function depending on the energies  $E$  and  $E'$  and on  $f$ . At zero magnetic field, the effect of this term on  $f(E)$  is similar to that of a phenomenological kernel  $K(\varepsilon) \propto 1/\varepsilon^2$  as found in [4]. For compatibility with phase-coherence time measurements (see below), the Kondo temperature was fixed at  $T_K = 40$  mK, which is the Kondo temperature of Mn in Ag. As shown by solid lines in Figs. 2 and 3, the data can be accurately reproduced using  $c = 17$  ppm,  $g = 2.9$ , and  $\nu J = 0.08$  [24]. Note, however, that according to material analysis of the silver batch used to fabricate sample No. 1, no magnetic impurity was present in the source at the level of 1 ppm. Since in some samples made out of the same batch the intensity of the energy exchange rate measured at  $B = 0$  was found to be up to 4 times smaller, pollution of the sample during fabrication might, at least partly, explain this large impurity concentration.

The impurity concentration deduced from the fits of  $f(E)$  must be further compared with the one obtained from the analysis of measurements of the phase-coherence time in long wires fabricated previously with the same source materials. We have extracted the phase-coherence time  $\tau_\varphi$  from the magnetoresistance of wires much longer than the phase-coherence length, using the weak localization theory. In samples made of 6N purity Ag,  $\tau_\varphi(T) = A^{-1}T^{-2/3}$  from 1 K down to 40 mK, with  $A = 0.36$  ns $^{-1}$  K $^{-2/3}$ , in reasonable agreement with the theory of Coulomb interactions in disordered wires ( $A_{\text{theory}} = 0.31$  ns $^{-1}$  K $^{-2/3}$ ). At  $T = 40$  mK,  $\tau_\varphi = 21$  ns. In samples made of 5N silver,  $\tau_\varphi(T)$  does not vary between  $T = 200$  mK and 40 mK, where we find  $\tau_\varphi = 3.5$  ns. This behavior can be attributed to the presence of magnetic impurities, with concentration  $c$ , spin  $s$ , and Kondo temperature  $T_K$ , which lead to a spin-flip rate described by [11,25]  $\gamma_{sf}(T) = (c/\pi\hbar\nu)\pi^2 s(s+1)/[\pi^2 s(s+1) + \ln^2(T/T_K)]$ . The resulting phase-coherence time  $\tau_\varphi(T) = 1/[AT^{2/3} + \gamma_{sf}(T)]$  shows very little variation between 40 and 200 mK and describes precisely the experimental data for  $c = 0.13$  ppm,  $T_K = 40$  mK,  $s = 1/2$  and  $A = A_{\text{theory}} = 0.4$  ns $^{-1}$  K $^{-2/3}$ . This value of  $c$ , compatible with the nominal source purity, is smaller by 2 orders of magnitude than the value obtained from the fits of energy exchange data on sample No. 1. A similar set of results was also obtained with Cu samples, a

material in which the oxide at the surface of the films was found to cause dephasing at low temperature [26]. Data on energy exchange [17] could also be fit with the theory of Göppert *et al.* [12], using  $T_K = 300$  mK,  $c = 4.8$  ppm,  $g = 2.3$ ,  $\nu J = 0.1$ , on top of a Coulombic term with intensity  $\kappa = 0.5 \text{ ns}^{-1} \text{ meV}^{-1/2}$  [23]. This result gives evidence that the anomalous interactions observed in many Cu wires at  $B = 0$  [4,7] are also due to magnetic impurities. Here also, measurements of the phase-coherence time [7] are explained by significantly smaller impurity concentrations ( $\sim 0.3$  ppm). This repeated discrepancy on the concentrations deduced from the two types of measurements remains an open problem. From an experimental point of view, a more quantitative test of theory could be obtained in samples with added, identified magnetic impurities at a known concentration [27].

In conclusion, we have found that anomalous energy exchange rates between quasiparticles were strongly reduced by the application of a magnetic field. Moreover, the energy and magnetic field dependence of the exchange rate can be accurately accounted for by the presence of a small concentration of Kondo magnetic impurities [12]. It is worthwhile to compare this result with recent measurements on Aharonov-Bohm rings, which show that the small phase-coherence times found at  $B = 0$  were increased in a finite magnetic field [28]. All these measurements indicate that the presence of very dilute magnetic impurities is a very plausible candidate to explain both extra dephasing and extra energy exchange observed in many mesoscopic samples.

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