Effect of Magnetic Impurities on Energy Exchange between Electrons

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In order to probe quantitatively the effect of Kondo impurities on energy exchange between electrons in metals, we have compared measurements on two silver wires with dilute magnetic impurities (manganese) introduced in one of them. The measurement of the temperature dependence of the electron phase coherence time on the wires provides an independent determination of the impurity concentration. Quantitative agreement on the energy exchange rate is found with a theory by Göppert et al. that accounts for Kondo scattering of electrons on spin-1/2 impurities.

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In diffusive metals, it is expected that the dominant inelastic electron scattering process at low temperature is the Coulomb interaction [1,2], leading to a power law increase of the electron phase coherence time \( \tau_\varphi \) with decreasing temperature \( T \). However, in the presence of a small concentration of magnetic impurities with low Kondo temperature, \( \tau_\varphi \) can be limited by spin-flip scattering, resulting in a nearly temperature independent phase coherence time over a broad temperature range [3]. As shown in Ref. [3], this mechanism could explain the apparent low-temperature saturation of \( \tau_\varphi \) observed in many experiments, which caused a controversy in recent years [4,5]. It was recently proposed that magnetic impurities also affect the energy exchange rate between electrons [6], which could explain the anomalous interaction rate observed in a series of experiments [7,8]. A first hint that this proposal is relevant was the observation of a magnetic field dependence of the rate [9,10], in a manner consistent with a theory taking into account the Kondo effect [11]. In those experiments, however, the nature and amount of magnetic impurities were not controlled. Assuming that the impurities were Mn, the concentrations needed to explain energy exchange experiments in silver wires were up to 2 orders of magnitude larger than the concentrations deduced from \( \tau_\varphi \) measurements on similar samples [9,10]. It was proposed that the samples for energy exchange rate measurements could have been contaminated during fabrication [9,10]. Another hypothesis is that impurities other than Mn, which affect energy exchange rates more drastically than phase coherence, were present [12,13]. Comparison of these proposals with existing experimental results is difficult because it requires dealing with more involved theories (large spin, surface anisotropy, large Kondo temperature), and pointless because it requires uncontrolled extra parameters. In order to overcome these difficulties and investigate quantitatively the mechanism proposed by Ref. [6], we have performed a comparative experiment described in this Letter, in which we probe the specific effect of the addition of 0.7 ppm (parts per million) of Mn atoms on the energy exchange rate between electrons. We measured the temperature dependence of \( \tau_\varphi \) on the same samples, accessing interactions in a complementary manner.

The scattering of electrons by magnetic impurities in metals is a many-body problem known as the Kondo effect: electrons tend to screen the spin of the impurity, leading to a renormalization of the scattering rate. The characteristic energy scale for this process is the Kondo temperature \( T_K \). At \( T \approx T_K \), screening is incomplete, and spin-flip scattering takes place, whereas, at \( T < T_K \), the impurity and the electrons form a singlet state, leading to potential scattering only. As far as electron dephasing is concerned, the Kondo effect results in a maximal dephasing rate at \( T_K \) [14]. The Kondo effect also provides a channel for efficient energy exchange between electrons scattering from the same magnetic impurity [6,15,16]. The rate of such a process depends on the energy of the states of the magnetic impurity, and is therefore sensitive to the magnetic field because of the Zeeman effect [11]. The spin states of the magnetic impurities can furthermore be split in the presence of spin-orbit scattering near an interface [17], which also modifies the rate. Further complications arise when the concentration of magnetic impurities is so high that the RKKY interaction between magnetic impurities constrains the spin dynamics [18,19].

In order to test quantitatively the impact of magnetic impurities on the energy exchange between electrons, we have compared the energy exchange rate and \( \tau_\varphi(T) \) in two wires that differ only by the intentional addition of manganese impurities in one of them, with concentrations low enough so that interactions between Mn impurities can be neglected [18]. To observe specifically the influence of the Mn impurities, the two samples were fabricated simultaneously on the same wafer. In a first step, a set of wires and their contact pads were patterned by e-beam lithography and evaporation of silver from a nominally 6N-purity source (99.9999% Ag from Alfa Aesar®). Mn\( ^{2+} \) ions were implanted at 70 kV in half of them, using the ion implanter IRMA at CSNSM Orsay. The neutralization current from the sample holder to ground was monitored during the implantation, leading to a direct measurement of the number of implanted atoms. Monte Carlo simulations
[20] yield the concentration of Mn atoms that stop inside the silver wire, $c = 0.7 \pm 0.1$ ppm. In order to measure the energy exchange between electrons [9], a long and thin electrode forming a tunnel junction with the middle of the wire is used as a probe. This electrode was patterned on individual chips in a second lithography step followed by evaporation of 3.5 nm of aluminum, oxidation, and evaporation of 16 nm of aluminum. We focus here on the results obtained on two wires, one without manganese added (labeled “bare” in the following), one with manganese added (“implanted”). For both samples, the wire length and cross-section area are added (“implanted”). For both samples, the wire length and cross-section area are $L = 40 \mu$m and $S_c = 230$ nm $\times$ 42 nm. The samples were measured in a dilution refrigerator with base temperature of 20 mK. The lower temperature wire resistance ($R = 55 \, \Omega$) was identical for both wires, which yields the diffusion constant of electrons $D = 0.029 \, m^2/s$.

For each wire, we have first measured the magnetoresistance at temperatures ranging from 20 mK to 7 K. Following Refs. [3,21], magnetoresistance curves are fit using the theory of weak localization, resulting in evaluations of the phase coherence time $\tau_\varphi$. In the bare wire, it was important to take into account finite length corrections because $\tau_\varphi$ is comparable to the diffusion time $\tau_D = L^2/D = 56$ ns below 1 K [22], leading to a reduction of the predicted magnetoresistance by $\approx 30\%$ below 1 K. Reproducible conductance fluctuations were visible, so that the uncertainty in the determination of $\tau_\varphi$ becomes large below 60 mK in the bare sample. The spin-orbit time $\tau_{so} = 8$ ps was extracted from the data above 1 K. The temperature dependence of $\tau_\varphi$ is shown in Fig. 1 for both wires. Below 1 K, $\tau_\varphi$ is smaller by nearly 1 order of magnitude in the implanted wire than in the bare one. In none of the samples does $\tau_\varphi$ increase as $T^{-2/3}$ when temperature is lowered, as would be expected if the electron-electron interaction was the dominant dephasing process (solid line labeled “pure” in Fig. 1). The apparent saturation of $\tau_\varphi$ is attributed to the presence of magnetic impurities [3]. This effect is quantified by a fit of the data with a sum of three terms:

$$\frac{1}{\tau_\varphi} = \mathcal{A} T^{2/3} + B T^3 + \gamma_{sf}(T),$$

(1)

with $\mathcal{A} = \frac{1}{\hbar} \left( \frac{\pi e^2}{4\pi^2 \hbar S S_c} \frac{R}{R_K} \right)^{1/3}$ describing Coulomb interaction [21], $B$ the electron-phonon interaction [5], and

$$\gamma_{sf}(T) = \frac{c}{\pi h \nu_F} \frac{\pi^2 S(S + 1)}{\pi^2 S(S + 1) + \ln(T/T_K)^2}$$

(2)

the spin-flip (sf) scattering rate, according to the Nagaoka-Suhl formula [3,23]. The density of states in silver is $\nu_F = 1.03 \times 10^{14}$ s$^{-1}$ m$^{-2}$ (2 spin states, the resistance quantum $R_K = h/e^2$, and the spin of the magnetic impurities $S$. Assuming that the only magnetic impurities present are Mn atoms, with $S = 5/2$ and $T_K = 40$ mK [24] and that $\mathcal{A}$ is fixed at its theoretical value $\mathcal{A} = 0.19$ ns$^{-1}$ K$^{-2/3}$, the best fits are obtained for $c_b = 0.10 \pm 0.01$ ppm (bare wire) and $c_i = 0.95 \pm 0.1$ ppm (implanted wire). The upper line is the prediction without spin-flip scattering ($c = 0$). Inset: layout of the circuit.

The switch is open for magnetoresistance measurements, closed for energy exchange measurements.

We have then measured the energy exchange rate between electrons and its dependence on magnetic field $B$ on the same two wires. The principle of the experiment is to drive electrons out of equilibrium with a bias voltage $U \gg k_B T/e$. The distribution function $f(E)$ of the electrons in the middle of the wire depends crucially on energy exchange between electrons [7]. The differential conductance $dI/dV(V)$ of the tunnel junction between the wire and the probe electrode (inset of Fig. 1, switch closed; see also Ref. [9]) is a convolution product of $f(E)$ with a function $q(E)$ describing inelastic tunneling [9]:

$$R_t \frac{dI}{dV}(V) = 1 - \int f(E)q(eV - E)dE$$

(3)

where $R_t$ is the resistance of the tunnel junction. The information on $f(E)$ is therefore contained in $dI/dV(V)$ via the $q$ function. The experiment is performed at $B \geq 0.3$ T, and the aluminum probe electrode is in its normal state. The $q$ function is obtained from $dI/dV(V)$ at $U = 0$, where $f(E)$ is a Fermi function. In this situation, $dI/dV(V)$ displays a sharp minimum at zero voltage (sometimes called “zero bias anomaly”), due to dynamical Coulomb blockade of tunneling [26]. The environmental impedance...
The coupling between electrons and magnetic impurities can be described by an exchange Hamiltonian, characterized by a coupling constant $J$. At zero magnetic field, this description leads to energy exchange in second order perturbation theory, as described in Ref. [6]. At finite magnetic field, the spin states of the impurities are split by the Zeeman energy $E_Z = g\mu_B B$. The energy $E_Z$ can then be exchanged at the lowest order in perturbation theory between electrons and impurities. This approach is sufficient to understand qualitatively the magnetic field behavior: the rate of interaction decays rapidly when $E_Z > eU$, because very few electrons can excite the impurities. The magnetic fields $eU/g\mu_B$ (using $g = 2$ for Mn) are 0.86, 1.7, and 2.6 T for $U = 0.1$, 0.2, and 0.3 mV, which correspond in the implanted wire to the fields at which the curvature of $dI/dV(V)$ near $V = U/2$ changes sign.

In the bare sample, the double dip also gets sharper when $B$ is increased. This is an indication that, as inferred from $\tau_{\phi}(T)$ measurements, this sample also contained some magnetic impurities. However, the corresponding energy exchange rate is always smaller than $1/\tau_D$, and $dI/dV(V)$ displays a double dip.

In order to compare quantitatively the measurements with theory, the renormalization of the coupling constant $J$ by the Kondo effect needs to be considered. Very roughly, this renormalization amounts to [6] $J_{\text{eff}}/J = \left[\nu_J J \ln(eU/k_B T_K)\right]^{-1} = 3$. More precisely, $J_{\text{eff}}$ depends on the distribution function $f(E)$, and only the full theory of Ref. [11] is able to quantify this effect and to treat the exchange Hamiltonian at all orders on the same footing. We have therefore solved the Boltzmann equation for $f(E)$ self-consistently, taking into account Coulomb interaction, electron-phonon interaction [28], and the effect of magnetic impurities in a magnetic field following the full theory of Ref. [11]. The concentration of magnetic impurities and the electron-phonon coupling were fixed at the values determined from the fit of $\tau_{\phi}(T)$ [28]. We used $T_K = 40$ mK [24] and $g = 2.0$ [29]. Note that this theory assumes $S = 1/2$ whereas $S = 5/2$ for Mn atoms, but it is not expected that this difference has a large influence on the energy exchange [13]. The intensity of the Coulomb
interaction alone could not be determined accurately from $\tau_{\text{c}}(T)$, and since it was found that theory underestimates the intensity $\kappa_{\text{c}}$ of the Coulomb interaction [30], $\kappa_{\text{c}}$ was used as a free parameter, common to both samples. A slight increase in temperature of the contact pads of the wire with $U$ (0.76 K/mV) was taken into account [28]. We also included in the calculation a slight heating of the electrons in the probe electrode at the junction interface, due to the fact that $R_0$ is not negligible compared to $R_\text{p}$. The corresponding temperature $T_{\text{p}}/U(V)$ of the electrons in the probe electrode is $T_{\text{p}} = 0.34$ K in the bare and 0.16 K in the implanted sample at the dips ($V = 0$ or $U$), at $U = 0.3$ mV where $T_{\text{p}}$ is expected to be the largest. The differential conductance $dI/dV(V)$ was then computed using Eq. (3). The resulting curves are displayed as solid lines in Fig. 2. The best agreement between theory and all the data was found for $\kappa_{\text{c}} = 0.05$ ns$^{-1}$ meV$^{-1/2}$. This value is larger than the Altshuler-Aronov-Khmelnitsky prediction $\kappa_{\text{c,AK}} = 0.016$ ns$^{-1}$ meV$^{-1/2}$ [1], as was repeatedly found in previous experiments [30]. A good overall agreement is found for both data sets, but some discrepancy appears for the implanted sample at $U = 0.3$ mV. We evaluated the sensitivity of the fits of the data on the implanted wire to the concentration $c_i$ of the impurities, and found that the best agreement is obtained at $c_i = 0.9 \pm 0.3$ ppm, in good agreement with the value 0.95 ppm deduced from the data of Fig. 1.

In conclusion, in this comparative experiment, the observed effect of well-identified magnetic impurities on the energy exchange is found to be in good quantitative agreement with the theory of Ref. [11], the concentration of impurities being fixed to the value deduced from the temperature dependence of the phase coherence time, which is also compatible with the expected value from implantation. This well-controlled experiment shows that the interaction mediated by dilute, low Kondo temperature magnetic impurities is well understood. However, it remains that, in this experiment as in all previous ones, the Coulomb interaction seems to be more efficient for energy exchange than predicted [30]. Open questions remain also on the contribution of the Kondo effect to dephasing and energy exchange at energies below $T_K$ [14], on the effect of the interactions between impurities at larger concentrations [18,19], and on finite size effects [12].

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