Dephasing by Extremely Dilute Magnetic Impurities Revealed by Aharonov-Bohm Oscillations

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We have probed the magnetic field dependence of the electron phase coherence time τ_ϕ by measuring the Aharonov-Bohm conductance oscillations of mesoscopic Cu rings. Whereas τ_ϕ determined from the low-field magnetoresistance saturates below 1 K, the amplitude of Aharonov-Bohm h/e oscillations increases strongly on a magnetic field scale proportional to the temperature. This provides strong evidence that a likely explanation for the frequently observed saturation of τ_ϕ at low temperature in weakly disordered metallic thin films is the presence of extremely dilute magnetic impurities.

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Understanding the sources of decoherence is of fundamental importance in all quantum systems that interact with their environment. In mesoscopic physics, quantum phase coherence of conduction electrons in disordered metals and semiconductors gives rise to a wide variety of novel transport phenomena observed at low temperature [1]; decoherence sets the limit on the length scale over which such effects are observable. In metallic thin films below about 1 K, the dominant source of decoherence is thought to be electron-electron interactions. For narrow wires, the phase coherence time for that process was calculated by Altshuler, Aronov, and Khmelnitskii [2] in 1982, and diverges at low temperature as $\tau_\phi \propto T^{-2/3}$.

This theoretical picture was deeply shaken in 1997 when two different experiments suggested that electrons in mesoscopic metallic wires interact with each other much more strongly than predicted by theory. One experiment [3] showed that τ_ϕ determined from the low-field magnetoresistance of mesoscopic Au wires, systematically saturates at low temperature, rather than continuing to increase as a power law. The other experiment [4] showed that the energy exchange rate between electrons measured in several Cu wires exceeded the prediction for electron-electron interactions [2] and obeyed a different energy dependence.

More recently, it was found that the measured energy exchange rate and dephasing rate are sample dependent and tightly correlated to the source material purity: samples fabricated using a very pure (99.9999%) Ag or Au source agreed with the theoretical prediction for electron-electron interactions, whereas samples fabricated with a source of lesser purity, or with Cu, showed a smaller phase coherence time and a larger energy exchange rate [5–9].

There have been several theoretical suggestions regarding the source of the excess dephasing. Several of those suggestions, such as the possibility of electromagnetic interference coming from outside the cryostat [10], can be ruled out purely on experimental grounds, since different samples measured in the same cryostat show different behaviors [6]. The controversial theory by Golubev, Zaikin, and Schön of zero temperature dephasing by high energy electromagnetic modes [11] is able to account for only a subset of the experimental results published in Refs. [3,6,7,12], using the overall prefactor of the dephasing rate as an adjustable parameter. In our opinion, the only two explanations that have not been ruled out by experiment are dephasing by two-level systems [13] and dephasing by paramagnetic impurities [14]. We aim to demonstrate in this Letter that all the data showing τ_ϕ saturation in weakly disordered metallic thin films published to date might be explainable by the presence of paramagnetic impurities at extremely low concentration [15].

How can we determine whether a sample which exhibits a saturation of τ_ϕ indeed contains unexpected magnetic impurities and that τ_ϕ is really limited by spin-flip scattering from these extrinsic degrees of freedom?

Probing the presence of unknown magnetic impurities at a concentration of 1 part per million (ppm) or smaller is a difficult challenge. For instance, the logarithmic contribution to the resistance by Kondo impurities [16] at such small concentrations cannot be reliably detected through the measurement of $R(T)$: it is very small and often hidden by the $1/\sqrt{T}$ contribution due to electron-electron interaction [2,17]. Since the dephasing rate is so sensitive to magnetic impurities, it is natural to use the dephasing rate itself to detect extremely small amounts of such impurities. In the presence of a sufficiently large magnetic field spin-flip collisions are frozen out, hence τ_ϕ should return to the value expected from electron-electron interaction. One method is to measure the weak-localization contribution to the perpendicular magnetoresistance in the presence of a parallel field large enough to freeze the spins. However, this requires an accurate alignment of the parallel field with the wire's axis, and the residual magnetic flux through the wire's cross section complicates the data analysis [18]. Another possibility is to measure the universal conductance fluctuations (UCF) of a metallic wire as a function of magnetic field B. But the weak dependence on τ_ϕ and broad width in B of UCF are not well suited to probe the presence of very dilute magnetic impurities at low

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temperature. We have chosen a third method. We fabricate a ring-shaped sample with connecting leads, and measure the Aharonov-Bohm (AB) oscillations in the magnetoconductance of the ring. Such measurements were pioneered by Webb and co-workers in the 1980s, and observed by several other groups at about the same time [19]. To obtain the largest possible AB oscillations, the rings measured at this time had a very small diameter, thereby reducing their sensitivity to the value of τ_ϕ. Hence, it is not surprising that the AB oscillations were found independent of the magnetic field except on a few samples purposely contaminated with a relatively very large ($\sim$ 100 ppm of Mn in Au) concentration of magnetic impurities [20].

We applied this technique, with carefully designed rings, to probe the magnetic field dependence of τ_ϕ in two Cu samples, labeled Cu3 and Cu4 (see Table I). The choice of copper followed from three observations. First, as illustrated in Fig. 1, all of the samples we have made with this material show a saturation of $\tau_\phi(B \approx 0)$ at low temperature, regardless of the source purity (see also [6,9]). Second, whereas it is possible to attain an apparent saturation of τ_ϕ over a limited temperature range from a combination of electron-electron interaction and spin-flip scattering [9], the flat and small τ_ϕ in samples Cu3 and Cu4 is difficult to reproduce in this manner. Third, no trace of Kondo magnetic impurities could be detected in the temperature dependence of the resistance, shown for sample Cu4 in the inset of Fig. 1. These three observations cast doubt on our proposal that the saturation of τ_ϕ at low temperature in Cu could be explained by magnetic impurities, hence Cu is an ideal candidate for this experiment.

We measured the magnetoconductance of two rings of radius $r = 0.5$ and $r = 0.75$ μm, located, respectively, on samples Cu3 and Cu4. A scanning electron microscope picture of the ring in sample Cu4 is shown in Fig. 2. In addition to the ring, the samples Cu3 and Cu4 contain a long meander line used to extract $\tau_\phi(B \approx 0)$ by fitting the low-field magnetoresistance with the prediction of weak-localization theory [17]. The fit procedure is detailed in [21]. All the Cu samples were deposited on a silicon substrate, through a suspended mask fabricated using standard e-beam lithography, in a thermal evaporator used only for nonmagnetic metals Ag, Al, Au, Cu, Ti. The evaporation rate ranges between 0.2 and 0.5 nm/s, under a pressure that stays below 10^{-6} mbar. The source material and boat were replaced before each evaporation and manipulated using plastic tweezers. Before the actual deposition on the Si substrate we melt the Cu source, evaporate 10–20 nm onto a shutter, and pump the chamber down again for about 15 mn. This preevaporation covers the walls of the evaporator with a clean layer of Cu and makes it possible to maintain a very low pressure during the sample fabrication. The samples were immersed in the mixing chamber of a dilution refrigerator and measured through filtered electrical lines. Resistance measurements were performed using a standard ac four-terminal technique with a lock-in amplifier. The ac voltage excitation V_{ac} across the sample satisfies $eV_{\mathrm{ac}} \lesssim k_B T$ to avoid heating of electrons.

TABLE I. Geometrical and electrical characteristics of the measured copper samples. The diffusion constant D is obtained using Einstein's relation with the density of states in copper $\nu_F = 1.56 \times 10^{47}$ J^{-1} m^{-3} and the resistivity ρ extracted from the thickness t, length $L = 285$ μm, width w, and resistance R of the long wire. In samples Cu3 and Cu4, the ring's radius and linewidth are, respectively, r and w_{AB}.

Sample	t (nm)	w (nm)	R (Ω)	D (cm^2/s)	r (μm)	w_{AB} (nm)
Cu1	45	155	700	146	n.a.	n.a.
Cu2	20	70	7980	61	n.a.	n.a.
Cu3	33	75	4370	65	0.5	67
Cu4	20	80	8500	52	0.75	73

The most important result of this article is visually striking in the raw magnetoconductance data, as illustrated on Fig. 3 for sample Cu4 at $T = 100$ mK. Whereas the Aharonov-Bohm oscillations can hardly be seen at small magnetic field B (left inset in Fig. 3), they are

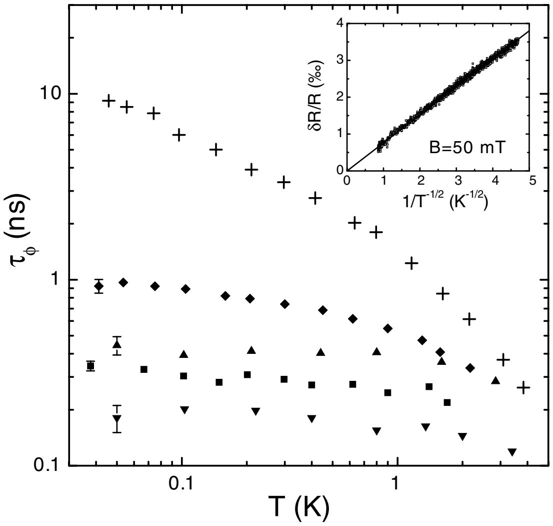

FIG. 1. Phase coherence time $\tau_\phi(B \approx 0)$ vs temperature determined by fitting the low field magnetoresistance of long wires to weak localization theory. The uncertainties are indicated by error bars at the lowest temperature where they are usually the largest. The samples are made of silver Ag1 (+) (data taken from [6]) and copper Cu1 (▲), Cu2 (◆), Cu3 (▼), and Cu4 (■). $\tau_\phi(B \approx 0)$ increases continuously with decreasing temperatures in the silver sample, whereas in all Cu samples $\tau_\phi(B \approx 0)$ shows a saturation at low temperatures. Inset: symbols represent the relative variation of the resistance of sample Cu4 in a small magnetic field $B = 50$ mT, plotted as a function of $1/\sqrt{T}$. The continuous line is a fit using the functional form $A/\sqrt{T}$ predicted by the theory of electron-electron interaction in diffusive metallic wires [17]. The best fit is obtained with $A = 7.6 \times 10^{-4}$ K$^{1/2}$ in close agreement with the theoretical prediction 7.2×10^{-4} K$^{1/2}$.

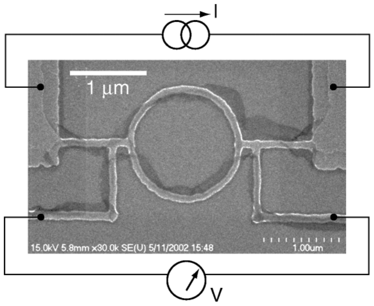

FIG. 2. Photograph of the Aharonov-Bohm ring in sample Cu4. We obtain the resistance by a four-lead measurement. The bottom left voltage lead is a very long wire used to extract $\tau_\phi(B \approx 0)$ from the low field magnetoresistance.

obvious at large B (right inset in Fig. 3). For a given geometry and temperature, the amplitude of AB oscillations depends only on τ_ϕ, which means, without further analysis, that τ_ϕ increases with magnetic field in this sample. A similar behavior was observed in sample Cu3.

To put this qualitative observation on solid ground we analyzed the data by taking the Fourier transforms of magnetoconductance data segments of width 0.2 T, using a Welch window. We define the amplitude of the AB h/e oscillations $\Delta G_{h/e}$ as the square root of the power integrated over a narrow interval surrounding the frequency $1/\Delta B \simeq \pi r^2/(h/e)$. Figure 4 shows the results for sample Cu4 at $T = 40$ and 100 mK, as a function of the reduced magnetic field $2\mu_B B/k_B T$. Apart from confirming quantitatively the increase of AB oscillations with the magnetic field, Fig. 4 shows that the magnetic field scale on which the AB h/e oscillations increase is proportional to the temperature. This proves that the increase of AB oscillations at large field results from the freezing of paramagnetic degrees of freedom in the sample.

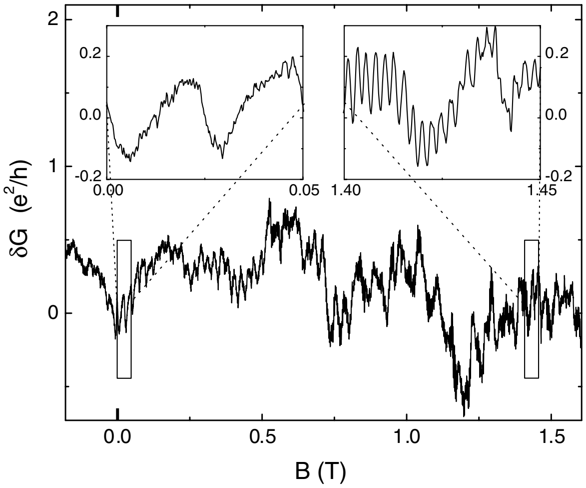

FIG. 3. Measured conductance of the ring in sample Cu4, in units of e^2/h, as a function of magnetic field at a temperature $T = 100$ mK. The narrow Aharonov-Bohm oscillations ($\Delta B \simeq 2.5$ mT) are superimposed on the larger and much broader universal conductance fluctuations. Left inset: blowup of the data near zero field. The AB oscillations are hardly visible. Right inset: blowup of the data at large magnetic field. The AB oscillations are much larger.

We can compare the measured amplitude of AB h/e oscillations with the prediction [22]

$$\Delta G_{h/e} = C\frac{e^2}{h}\frac{L_T}{\pi r}\sqrt{\frac{L_\phi}{\pi r}}\exp[-\pi r/L_\phi], \tag{1}$$

where $L_T = \sqrt{\hbar D/k_B T}$ is the thermal length, D is the diffusion coefficient, T is the temperature, $L_\phi = \sqrt{D\tau_\phi}$, r is the ring radius, and C is a factor of order 1 that depends on the sample geometry in the vicinity of the ring. The total dephasing rate $1/\tau_\phi$ is the sum of contributions from electron-electron [17] and spin-flip scattering:

$$\frac{1}{\tau_\phi} = \frac{1}{\tau_{ee}} + \frac{1}{\tau_{\mathrm{sf}}}. \tag{2}$$

Whereas $1/\tau_{ee}$ is independent of B, the spin-flip scattering rate vanishes at large field as [23]

$$\frac{\tau_{\mathrm{sf}}(B=0)}{\tau_{\mathrm{sf}}(B)} = \frac{g\mu_B B/k_B T}{\sinh(g\mu_B B/k_B T)}, \tag{3}$$

where g is the renormalized gyromagnetic factor of the magnetic impurities.

The continuous line in Fig. 4 is a fit of $\Delta G_{h/e}(B)$ at $T = 100$ mK using Eqs. (1)–(3) with the gyromagnetic factor $g = 1.05$ and the overall multiplicative constant $C = 0.13$ as the only fit parameters. The dotted line is the calculated $\Delta G_{h/e}$ at $T = 40$ mK using the same values for g and C, and taking into account the non negligible noise level of the measurement at this temperature. We used the predicted contribution for electron-electron interactions [17]: $\tau_{ee} = 7.4$ ns and 5.4 ns, respectively, at $T = 40$ and 100 mK, and we used the values

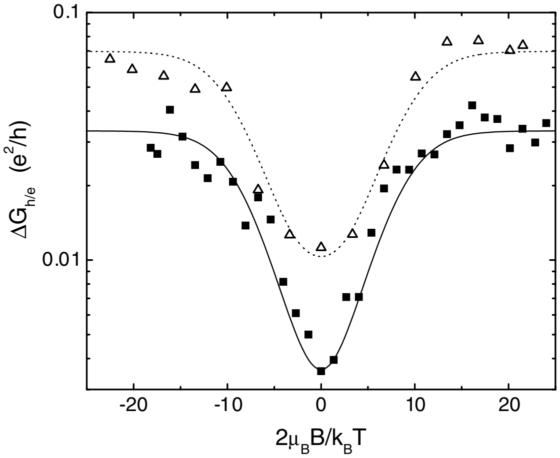

FIG. 4. Symbols: mean amplitude of the AB h/e oscillations ($\Delta G_{h/e}$) in units of e^2/h in sample Cu4 at $T = 40$ (△) and 100 mK (■), plotted as a function of the reduced magnetic field $2\mu_B B/k_B T$. Lines: fits to the two data sets using Eqs. (1)–(3) with C and g as fit parameters (see text).

$\tau_{\mathrm{sf}}(B = 0) \simeq \tau_\phi(B \approx 0)$ [24] obtained from the low-field magnetoresistance of the long wire.

What can be these paramagnetic "impurities" in our samples? It is important to emphasize that the sample purity is not automatically identical to the source material purity. Indeed, the residual pressure in the evaporator is not small enough to rule out extra contamination at the level of 1 ppm during evaporation. The nature of such contaminants depends on the evaporator history. However, our best candidate is the surface oxide of Cu [25]. It was already pointed out in the late 1980s that the surface of Cu may cause dephasing [26]. By comparing the value of $\tau_\phi(B \approx 0)$ at low temperature with the unitary limit ($T = T_K$) of spin-flip scattering in the Kondo regime [9,27], we estimate the concentration of Kondo magnetic impurities in samples Cu1, Cu2, Cu3, and Cu4 to be, respectively, 0.75, 0.3, 1.5, and 1 ppm. Note that these would be lower bounds on the concentrations if the paramagnetic impurities are on the surface, with a distribution of Kondo temperatures.

To conclude, we have measured the Aharonov-Bohm h/e oscillations to probe the presence of very dilute, low Kondo temperature, magnetic impurities. By applying this technique to copper wires, we showed that the saturation of the low-field phase coherence time is due to spin-flip scattering by magnetic impurities. Recent measurements of energy exchange in a magnetic field demonstrate that magnetic impurities are also responsible for the anomalous energy exchange previously observed in mesoscopic wires [28].

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