

# Electron dephasing in metallic thin films at low temperatures

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(Received April 1, 2003)

The low temperature behavior of the electron phase coherence time in metals,  $\tau_\phi$ , is currently a subject of controversy. While theory predicts that  $\tau_\phi$  should increase on cooling as an inverse power law of temperature, many samples exhibit a saturation of  $\tau_\phi$  below a temperature between 0.1 and 1 K. In this paper we present evidence that the saturation of  $\tau_\phi$  often observed in weakly-disordered polycrystalline metals (Au, Ag, and Cu) is most likely due to a dilute concentration of paramagnetic impurities, either on the surface or in the bulk.

**KEYWORDS:** electron phase coherence, mesoscopic physics, saturation of dephasing time, magnetic impurities

## 1. Introduction

At low temperatures, conduction electrons in disordered metals maintain their phase coherence over times often exceeding one nanosecond – several orders of magnitude longer than the time between elastic collisions. The resulting quantum interference of electrons gives rise to many phenomena of mesoscopic physics, such as the weak-localization corrections to the conductance, universal conductance fluctuations, persistent currents and Aharonov-Bohm conductance oscillations in ring-shaped samples, and the proximity effect near a superconducting/normal metal interface.

There is currently experimental and theoretical controversy concerning the very low temperature behavior of the phase coherence time  $\tau_\phi$ . The standard theories of electron-electron and electron-phonon scattering predict that  $\tau_\phi$  should diverge as a power law of temperature in the limit of zero temperature.<sup>1)</sup> Many samples, however, exhibit a saturation of  $\tau_\phi$  at temperatures below 1 K.<sup>2)</sup> It is important to ascertain whether those experimental observations reflect a fundamental, intrinsic decoherence mechanism as suggested by some,<sup>2,3)</sup> or an extrinsic, sample-dependent source of decoherence.

In this paper we focus on the simplest metallic systems, namely polycrystalline thin films of the noble metals Au, Ag, and Cu. The samples we have measured are in the weak-disorder limit ( $k_F l \gg 1$ ), where the standard theory should be valid.<sup>1)</sup> Other systems, such as highly-disordered metallic alloys, may behave differently regarding electron decoherence.<sup>4)</sup> As evidence for the variety of behavior in different classes of materials, we note that the maximum value of  $\tau_\phi$  found by Mohanty et al. in their Au wires<sup>2)</sup> tended to decrease with increasing disorder (i.e. with decreasing diffusion constant  $D$ ), whereas the opposite trend was observed by Lin and coworkers in 3D alloys.<sup>4)</sup>

## 2. Experimental Methods and Prior Results

Over the past several years we have measured  $\tau_\phi$  in 18 samples made of Ag, Cu or Au. All of the samples measured were long, narrow wires, evaporated onto Si substrates through a suspended mask patterned using standard electron-beam lithography. Typical sample dimensions are lengths of several hundred  $\mu m$ , widths of 70-120 nm, and thicknesses of 20-45 nm. Samples are measured in a dilution refrigerator with filtered lines to minimize sample heating or electron dephasing by external rf interference. We measure the low-field magnetoresistance of the samples using standard low-frequency lock-in techniques, and determine  $\tau_\phi$  by fitting the magnetoresistance data to 1D weak-localization (WL) theory. (The fitting function is discussed in<sup>5)</sup>). The fitting procedure involves only two free parameters, namely the phase coherence length  $L_\phi = \sqrt{D\tau_\phi}$  and the spin-orbit scattering length  $L_{so}$ . We extract the diffusion coefficient  $D$  from the measured sample dimensions and resistance, using the Einstein relation  $\sigma = \nu e^2 D$ , where  $\nu$  is the density of states at the Fermi energy. In Au, the spin-orbit scattering is very strong, hence  $L_{so}$  can be ignored and  $L_\phi$  is the only fit parameter for each data set. In Ag and Cu,  $L_{so}$  is determined from the fits at high temperature (near and above 1 K), and is then held fixed at all lower temperatures so that again  $L_\phi$  is the only fit parameter. This procedure ensures that the determination of  $L_\phi$  at low temperature is not influenced by systematic errors in the determination of  $L_{so}$ .

Figure 1 shows a representative subset of our measurements of  $\tau_\phi$  in several samples of Ag, Cu, and Au.<sup>5,6)</sup> In several Ag samples (one of which is shown in the figure), and in one very pure Au sample,  $\tau_\phi$  is observed to increase without saturation down to 40 mK, the lowest temperature measured. These data demonstrate immediately that saturation of  $\tau_\phi$  is not a universal phenomenon in weakly-disordered metals. We note that the dimensions and resistance per unit length of these samples are similar to samples Au-3 and Au-6 in ref.,<sup>2)</sup> hence the claim in that paper that the maximum value of  $\tau_\phi$  depends only on these parameters is not correct.

Fig. 1 also shows a Au sample in which  $\tau_\phi$  is independent of temperature starting well above 1 K and contin-

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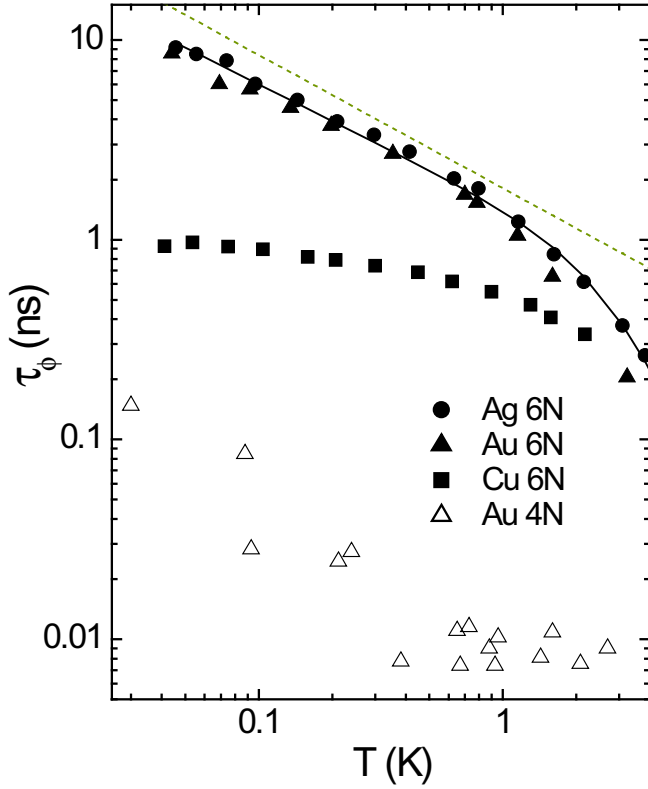


Fig. 1. Electron decoherence time,  $\tau_\phi$ , vs temperature for four samples of various materials and source purity: Ag6N (●), Au6N (▲), Cu6N (■), and Au4N (△).<sup>5,6</sup> The solid line is a fit to the Ag6N data with a function of the form  $\tau_\phi^{-1} = A*T^{2/3} + B*T^3$ , where A and B represent the strength of electron-electron and electron-phonon scattering, respectively.<sup>5</sup> The dashed line shows the predicted  $T^{-2/3}$  dependence of  $\tau_\phi$  at low temperature.

uing down to about 0.3 K, but then increases on further cooling. That sample was fabricated from Au source material of only 4N (99.99%) purity, whereas all the other samples we measured were fabricated from source material of 5N or 6N purity. From measurements of the resistance vs. temperature,  $R(T)$ , as well as the high-field magnetoresistance, we determined that the less pure Au samples contained a very high concentration (30-50ppm) of Fe impurities.<sup>6</sup> Those magnetic impurities limit  $\tau_\phi$  due to spin-flip scattering with a rate that peaks at the Kondo temperature ( $T_K = 0.3$  K for Fe in Au) and then decreases at lower temperature. As a historical note, we mention that Mohanty, Jariwala and Webb (MJW) measured  $\tau_\phi$  in a Au sample intentionally doped with 2.8ppm or Fe to compare the behavior with what they observed in their nominally pure Au samples.<sup>2</sup> Because  $\tau_\phi$  in the doped sample increased below 0.3 K, and because the presence of Fe could be detected in  $R(T)$ , MJW concluded that magnetic impurities could not be the source of the  $\tau_\phi$  saturation observed in the nominally pure samples. That conclusion is erroneous, however, because different magnetic impurities in the same host have different Kondo temperatures, and magnetic impurities on the surface of a metal are likely to have a distribution of Kondo temperatures. While MJW showed convincingly that Fe impurities in the bulk could not cause the saturation of  $\tau_\phi$  they observed, their conclusions do not extend to magnetic impurities with much lower Kondo temperatures, and at concentrations too low to detect in  $R(T)$ .

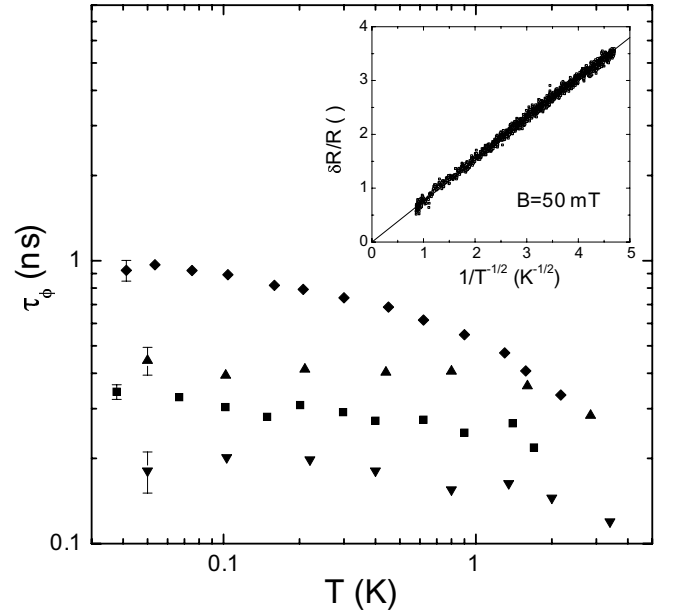


Fig. 2. Decoherence time,  $\tau_\phi$ , vs temperature for four Cu samples all fabricated from 6N purity source material. Inset: Temperature dependence of the resistance of sample Cu4 (■ in figure), showing only the  $A/\sqrt{T}$  behavior predicted by the theory of electron-electron interaction in diffusive metallic wires.<sup>10</sup> The data shown in Figs. 4-6 were obtained from sample Cu3 (▼ in figure).

Fig. 1 also shows data for a Cu sample made from 6N purity source material, in which  $\tau_\phi$  saturates below about 1 K. We have found that  $\tau_\phi$  is nearly independent

of temperature below 1 K in all of our Cu samples, regardless of the purity of the source material. Data for four such samples, all fabricated from 6N purity source material, are shown in Fig. 2. The odd behavior of  $\tau_\phi$  in Cu will be the subject of discussion in Section 4 below.

### 3. Correlation between electron decoherence and energy exchange

Measurements of  $\tau_\phi$  are not the only way to obtain information about electronic inelastic scattering processes. An alternative approach, pioneered by the Saclay group, is to measure energy exchange between quasiparticles in mesoscopic wires driven far from equilibrium by an applied bias voltage.<sup>7)</sup> The first measurements by that group of energy exchange in Cu wires showed that the rate of energy exchange was larger in the experiments than predicted by theory, and the energy dependence of the electron-electron interaction matrix element was different from that predicted by theory.<sup>10)</sup> Since then the Saclay group has performed energy exchange measurements on a large number of samples of Cu, Ag, and Au.<sup>8,9)</sup> They find that in Ag samples made from the highest purity (6N) material, the energy exchange obeys the theoretical prediction. In contrast, in Cu samples, or Ag or Au samples made from lower purity material, the energy exchange rate is too large and has the wrong energy dependence, as in the original measurements on a Cu sample. In a collaboration with the Saclay group, we have measured  $\tau_\phi$  in samples fabricated in the same e-beam evaporation system as the samples used in the energy exchange measurements, and from the same source material. Comparison of the  $\tau_\phi$  data and the energy exchange data on similarly-prepared samples has revealed a strong correlation between these two different probes of electron dynamics, suggesting that a single mechanism is responsible for both anomalies.<sup>5,6)</sup> In particular, Au samples with large concentrations of Fe impurities exhibit strongly enhanced energy exchange as well as strongly enhanced decoherence. Since the decoherence is known to be caused by spin-flip scattering on magnetic impurities, circumstantial evidence points to these same magnetic impurities as the source of the energy exchange also. But the Zeeman states of a paramagnetic impurity are degenerate in the absence of an applied magnetic field, so how can such impurities contribute efficiently to energy exchange between electrons?

The answer to this question was provided by Kaminisky and Glazman, who showed that electrons can exchange energy via a second-order process involving a virtual state of the magnetic impurity.<sup>11)</sup> Although second-order processes are generally much weaker than first-order, renormalization of the interaction between electrons and magnetic impurities by the Kondo effect makes this process more efficient than one might first expect.

### 4. Detection of extremely dilute magnetic impurities

The experimental results summarized in the previous two sections strongly suggest that dilute magnetic impurities are the cause of both excess dephasing and ex-

cess energy exchange between electrons observed in some samples. Samples without any magnetic impurities obey the standard theoretical predictions<sup>1,10)</sup> in both experiments. But how can we be sure? When the impurity concentration is large, as in the case of the 4N purity Au samples discussed earlier, the impurities are easily detectable through the logarithmic temperature dependence of the resistance associated with the Kondo effect, or from the large-field magnetoresistance. When the magnetic impurity concentration is well below 1ppm, however, detection by these means is not possible.

Since  $\tau_\phi$  is extremely sensitive to even a minute concentration of magnetic impurities, it makes sense to use  $\tau_\phi$  itself to detect them. In the presence of a sufficiently large magnetic field, the spin-flip scattering process is frozen out, hence  $\tau_\phi$  should return to the value determined by electron-electron interactions alone. Hence we need a way to measure  $\tau_\phi$  as a function of magnetic field. The weak localization contribution to the magnetoresistance disappears once the transverse field exceeds a few tens of Gauss – enough to put one flux quantum through an area equal to  $L_\phi$  times the sample width. Measuring the transverse magnetoresistance in the presence of a field parallel to the sample is a possibility, but requires extremely accurate alignment of the field with the sample axis, and the residual field flux through the wire's cross section complicates the analysis.<sup>12)</sup> Measurement of the universal conductance fluctuations requires a very broad field range to achieve sufficient ensemble averaging. While this is possible at high field, it is not possible at low field, hence a direct comparison of  $\tau_\phi$  at low and high field is not possible.<sup>13)</sup>

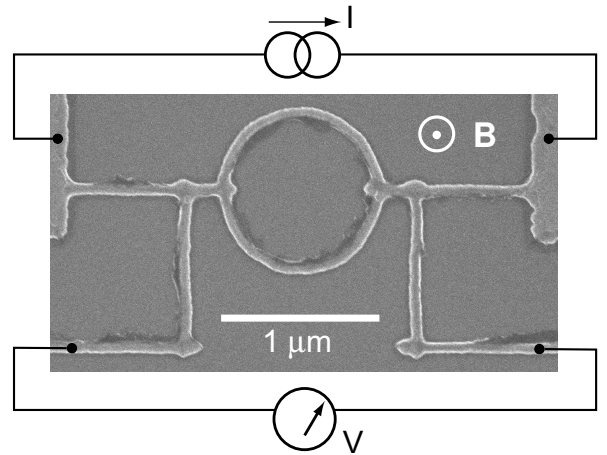


Fig. 3. Electron microscope picture of the ring-shaped part of sample Cu3, with diameter 1.0  $\mu\text{m}$ . Current and voltage measurement leads are labelled.

We have chosen to measure the magnetic-field dependence of  $\tau_\phi$  from the amplitude of the Aharonov-Bohm (AB) conductance oscillations of mesoscopic rings.<sup>14)</sup> Such measurements were pioneered by Webb and coworkers in the mid-1980's,<sup>15)</sup> who also measured the effect of large concentrations (40 and 120 ppm) of magnetic impurities on the amplitude of Aharonov-Bohm oscillations.<sup>16)</sup> We are interested here in extremely low concentrations – below 1 ppm – hence we must optimize the

sample design to achieve the highest possible sensitivity to changes in  $\tau_\phi$ . To get the highest sensitivity to  $\tau_\phi$ , one should make the ring as large as possible, so that the AB oscillations are exponentially suppressed by the ratio of half the ring circumference to  $L_\phi$ .<sup>17)</sup> The problem is that if the ring is too large, the AB oscillations are too small to measure. The compromise for the Cu rings we measured was to fabricate rings of diameter 1.0 and 1.5  $\mu\text{m}$ . The choice of Cu followed from the observations discussed in section 2. First, as shown in Fig. 2,  $\tau_\phi$  in Cu samples always saturates at a rather high temperature, near 1 K. Second, the nearly flat behavior of  $\tau_\phi(T)$  over a broad temperature range does not appear to be due to magnetic impurities: if  $T_K$  were high, then we should see  $\tau_\phi$  increase below  $T_K$ , whereas if  $T_K$  were low, then we should see  $\tau_\phi$  decrease as we approach  $T_K$  from above. Third, as shown in the inset of Fig. 2, no trace of magnetic impurities is visible in  $R(T)$  for our Cu samples.<sup>18)</sup>

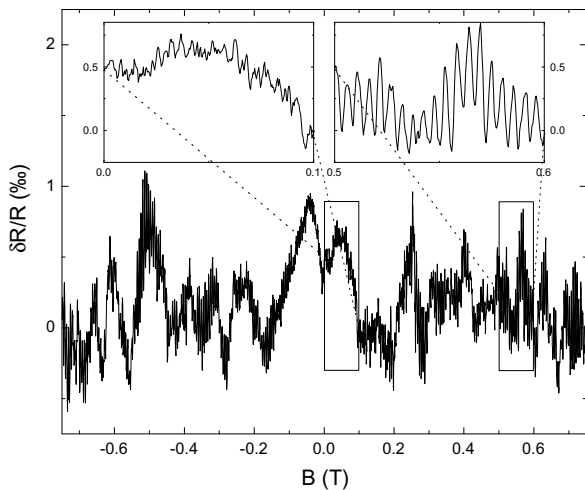


Fig. 4. Conductance of the ring of sample Cu3 shown in Fig. 4, in units of  $e^2/h$ , as a function of magnetic field at a temperature  $T = 56$  mK. The narrow Aharonov-Bohm oscillations ( $\Delta B \simeq 5$  mT) are superimposed on the larger and much broader universal conductance fluctuations. Left 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.

Each of the Cu samples in this study consisted of a ring to measure AB oscillations and a long wire to measure magnetoresistance. Fig. 3 shows a picture of the ring part of sample Cu3, with a diameter of 1.0  $\mu\text{m}$ . Fig. 4 shows the conductance of the ring as a function of magnetic field, at  $T = 56$  mK. The main part of the figure shows mainly the universal conductance fluctuations, since the AB oscillations are on a field scale too small to see clearly. The insets, however, show what is happening. Near zero field, there are UCF, but hardly any AB oscillations. At high field, the AB oscillations are much larger. Hence these raw data already support our suspicion that the small value of  $\tau_\phi$  at low field is due to spin-flip scattering by magnetic impurities, a process that freezes out in a large field. To analyze these data quantitatively, we Fourier transform the data in overlapping magnetic field intervals of width 0.2 T. The results

are shown in Fig. 5 for two field intervals, one centered at zero field and the other centered at  $B=0.6$  T. The  $h/e$  AB oscillations appear as a peak in the power spectrum near the central frequency  $1/\Delta B \simeq \pi r^2/(h/e)$ . To obtain the AB oscillation amplitude from the power spectrum, we take the square root of the integrated power over the frequency interval shown in the figure, which spans the frequency range determined by the inner and outer areas of the ring. (We do not subtract the noise, so as to keep the analysis as straightforward as possible.)

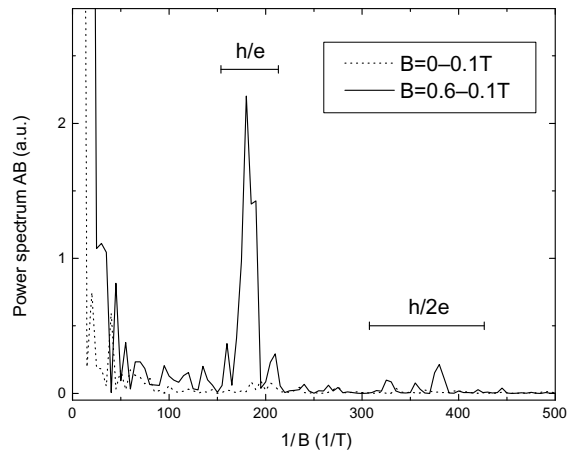


Fig. 5. Power spectrum (squared Fourier transform) of the data in Fig. 4, shown for two different magnetic field intervals. As was apparent in the raw data, the  $h/e$  AB oscillations are quite large for  $B$  near 0.6 T, whereas they are hardly visible near  $B=0$ .

Fig. 6 shows the complete magnetic field dependence of the AB oscillation amplitude, for  $T = 56$  and 100 mK. The figure shows that the characteristic field scale at which the AB oscillations increase is proportional to the temperature. This demonstrates unequivocally that the increase in the AB oscillations is due to freezing out of the spin-flip scattering process, and the concomitant increase of  $\tau_\phi$ .

To go further in the analysis requires fitting the data in Fig. 6 to a theoretical formula for the AB oscillation amplitude. This has been done elsewhere for the 1.5  $\mu\text{m}$ -diameter ring of sample Cu4.<sup>14)</sup> The data agree well with a fit based on the assumption that at high field  $\tau_\phi$  returns to the value determined solely by electron-electron scattering, and given by the theory.<sup>1)</sup> The  $g$ -factor of the magnetic impurities was a fit parameter, and the value obtained from the fit was 1.05.

We mention that the Saclay group has also developed a method to detect magnetic impurities in the energy exchange experiments.<sup>19)</sup> They have pursued measurements of Cu and  $^{51}\text{N}$  Ag samples that exhibit anomalous energy exchange at zero magnetic field. In both cases, the energy exchange rates decrease dramatically at high field, indicating that the anomalous energy exchange was associated with spin degrees of freedom.

## 5. Conclusions

We can draw several conclusions from the work presented here. First, neither the saturation of  $\tau_\phi$  nor the

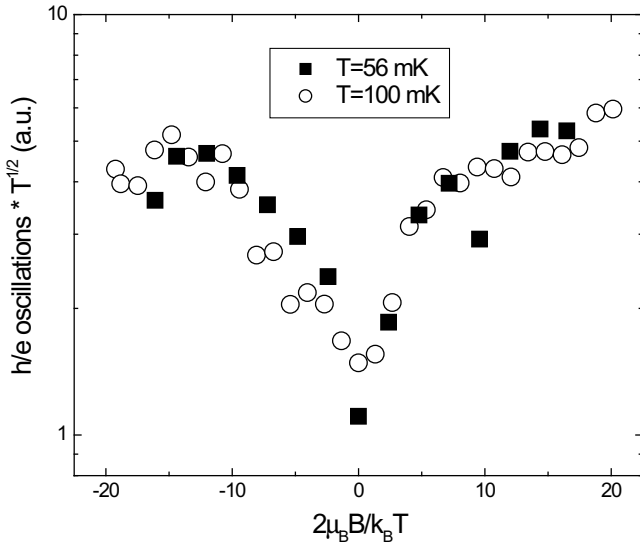


Fig. 6. Symbols: mean amplitude of the AB  $h/e$  oscillations ( $\Delta G_{h/e}$ ) in arbitrary units, normalized so as to remove the temperature dependence due to energy averaging. The two temperatures are  $T = 40\text{ mK}$  ( $\Delta$ ) and  $100\text{ mK}$  ( $\blacksquare$ ). The horizontal axis is the reduced magnetic field  $2\mu_B B/k_B T$ .

anomalous energy exchange we sometimes observe is a universal phenomenon. Indeed we have measured several samples of similar geometry and resistance that show neither effect, but rather obey the theoretical predictions.<sup>1,10</sup> Second, both anomalies, which are often observed in weakly-disordered metal samples, are not due to a measurement artifact such as heating by an external source of electromagnetic radiation. This follows from the observation that samples with similar geometry and electrical resistance measured in the same cryostat can nevertheless behave very differently. Third, we noticed a systematic correlation between observation of a short decoherence time  $\tau_\phi$  and of anomalous energy exchange, suggesting that a single physical mechanism is responsible for both observations. Fourth, whenever we have applied a sufficiently large magnetic field to samples exhibiting either one of the anomalies, the large decoherence and energy exchange rate at small field decreased on a magnetic field scale proportional to the temperature. This last observation strongly suggests that the anomalous decoherence and energy exchange observed in many samples result from the presence of very dilute magnetic impurities. We believe that all of the anomalous decoherence and energy exchange data observed to date in weakly-disordered metals could be explained by the presence of very dilute magnetic impurities.

What are these magnetic impurities? In mesoscopic Cu samples, it was shown long ago that  $\tau_\phi$  depended on the surface treatment, and it was hypothesized that the culprit was a paramagnetic oxide.<sup>20</sup> Our Aharonov-Bohm measurements strongly support that hypothesis. In Ag samples of 5N purity,<sup>21</sup> and in the Au samples measured by MJW,<sup>2</sup> we suspect that dilute Mn or Cr impurities are responsible, since their Kondo temperature is sufficiently low to preclude observation of the desaturation of  $\tau_\phi$  below  $T_K$ . But it is not unlikely that the

culprit is something more subtle, such as surface contamination from the electron lithography process. Of course the only way to prove that MJW's observations are due to magnetic impurities would be to detect them directly, as we have done using the AB effect in Cu.

Saturation of  $\tau_\phi$  has also been observed in some semiconductor systems. There the situation is less clear, but our experience with metals leads us to believe that eventually an extrinsic mechanism will be found.

### Acknowledgements

We thank our many collaborators, especially A. Anthore, A. Gougam, H. Pothier, and D. Esteve. This work was supported by NSF grants DMR-9801841 and 0104178, and by the Keck Microfabrication Facility supported by NSF DMR-9809688.

- 1) B.L. Altshuler, A.G. Aronov and D.E. Khmelnitskii, J. Phys. C: Solid State Physics **15**, 7367 (1982).
- 2) P. Mohanty, E.M.Q. Jariwala and R.A. Webb, Phys. Rev. Lett. **78**, 3366 (1997).
- 3) D.S. Golubev and A.D. Zaikin, Phys. Rev. Lett. **81**, 1074 (1998).
- 4) For a review, see J.J. Lin and J.P. Bird, J. Phys.: Condens. Matter **14**, R501 (2002), or J.J. Lin, T.J. Li, and Y.L. Zhong, this volume.
- 5) A.B. Gougam, F. Pierre, H. Pothier, D. Esteve, and N.O. Birge, J. Low Temp. Phys. **118**, 447 (2000).
- 6) F. Pierre, H. Pothier, D. Esteve, M.H. Devoret, A.B. Gougam, and N.O. Birge, in Kondo Effect and Dephasing in Low-Dimensional Systems, ed. by V. Chandrasekhar, C. Van Haesendonck and A. Zawadowski (Kluwer, Netherlands, 2001)
- 7) H. Pothier, S. Gueron, N.O. Birge, D. Esteve, and M.H. Devoret, Phys. Rev. Lett. **79**, 3490 (1997).
- 8) F. Pierre, H. Pothier, D. Esteve, and M.H. Devoret, J. Low Temp. Phys. **118**, 437 (2000).
- 9) F. Pierre, Ann. Phys. Fr. **26** N4 (2001).
- 10) For a review, see B.L. Altshuler and A.G. Aronov, in *Electron-Electron Interactions in Disordered Systems*, edited by A.L. Efros and M. Pollak (Elsevier Science Publishers B.V., 1985).
- 11) A. Kaminski and L.I. Glazman, Phys. Rev. Lett. **86**, 2400 (2001).
- 12) N. Giordano and M.A. Pennington, Phys. Rev. B **47**, 9693 (1993).
- 13) Relying on a comparison between the value of  $\tau_\phi$  deduced from WL at low field and UCF at high field is not recommended. See D. Hoadley, P. McConville, and N.O. Birge, Phys. Rev. B **60**, 5617 (1999). These authors analyzed the magnetic-field dependence of  $1/f$  noise, which is a probe of UCF at low temperature, and found values of  $\tau_\phi$  not in agreement with those obtained from WL fits to the magnetoresistance of the same samples. Contrary to what is stated in that paper, these results are not understood. See I.L. Aleiner and Ya. M. Blanter, Phys. Rev. B **65**, 115317 (2002).
- 14) F. Pierre and N.O. Birge, submitted to Phys. Rev. Lett.
- 15) S. Washburn and R.A. Webb, Rep. Prog. Phys. **55**, 1311 (1992), and references therein.
- 16) A.D. Benoit, S. Washburn, R.A. Webb, D. Mailly, and L. Dumoulin, *Anderson Localization*, edited by Heidelberg (Springer, 1988).
- 17) A.G. Aronov and Y.V. Sharvin, Rev. Mod. Phys. **59**, 755 (1987); V. Chandrasekhar, Ph.D. thesis, Yale University (1989).
- 18) The resistance versus temperature of sample Cu3 does not follow perfectly the  $T^{-1/2}$  law. Nevertheless,  $R(T)$  in that sample is independent of magnetic field within our experimental resolution, i.e. no sign of magnetic impurities is visible.
- 19) A. Anthore, F. Pierre, H. Pothier, and D. Esteve, submitted to Phys. Rev. Lett.; A. Anthore, F. Pierre, H. Pothier, D. Esteve, and M.H. Devoret, *Proceedings of the XXXVth Rencontres*

- de Moriond* “*Electronic Correlations: From Meso- to Nano-Physics*”, Les Arcs, France, 2001.
- 20) J. Vranken, C. Van Haesendonck, and Y. Bruynseraede, Phys. Rev. B **37**, 8502 (1988).
- 21) F. Pierre, A. Gougam, N.O. Birge, A. Anthore, H. Pothier, and D. Esteve, in preparation.