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Magnetoresistance of small, quasi-one-dimensional, normal-metal rings and lines

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The magnetoresistance of sub-0.4-μm-diam Au and $Au_{60}Pd_{40}$ rings was measured in a perpendicular magnetic field at temperatures as low as 5 mK in search of simple, periodic resistance oscillations that would be evidence of flux quantization in normal-metal rings. However, instead of simple oscillations, a very complex structure developed in the magnetoresistance at low temperatures. Fourier analysis of all the data did not reveal convincing evidence for flux quantization in the rings. Complex structure similar to that observed in the rings was also found in the magnetoresistance of short, narrow, Au and $Au_{60}Pd_{40}$ lines. This structure appears to be associated with the small size of the devices.

The transport properties of electrical devices are expected to change considerably as the size of the devices approaches atomic dimensions. As the number of atoms forming a device becomes very small, the spacing between energy levels will become large enough to influence the device characteristics. In addition, nonequilibrium, ballistic transport phenomena will become increasingly important as device dimensions become smaller than electron scattering lengths. Recently, a great deal of interest was generated by the theoretical prediction[1] and experimental observation[2–5] of flux periodic resistance oscillations with respect to a flux quantum of $h/2e$ in hollow normal-metal cylinders roughly 1 μm in diameter (normal-metal flux "quantization"). These periodic oscillations are a consequence of the phase dependence of electrons on the magnetic vector potential encircled by the cylinder[6] and of the nonequilibrium nature of the transport in normal-metal cylinders with diameters smaller than the inelastic scattering length of electrons.[1] The case of small, one-dimensional rings has also been considered theoretically, and oscillations periodic with respect to h/e,[7] as well as $h/2e$,[1,7] are predicted. In this Rapid Communication, we will describe measurements of the magnetoresistance of small Au and $Au_{60}Pd_{40}$ rings made in search of flux quantization effects in normal-metal rings.

The fine-line devices studied in this experiment were fabricated using contamination nanolithography from 38-nm-thick Au and $Au_{60}Pd_{40}$ films evaporated onto a 200-nm-thick Si_3N_4 membrane. The process has been described in detail elsewhere.[8] The Si_3N_4 membrane was transparent to electrons and permitted the morphology of the fine-line devices to be monitored using high-resolution transmission electron microscopy during sample processing. The fine-line devices were measured in a ^{3}He-^{4}He dilution refrigerator which reached temperatures as low as 5 mK. The four-probe resistance of the samples was measured with an ac bridge consisting of two PAR-124 lock-in amplifiers, a room-temperature standard resistor, and a HP-3456A digital voltmeter. The low-temperature conductivity of the Au samples was limited by surface scattering, while that of the $Au_{60}Pd_{40}$ samples was limited by bulk scattering, with an elastic mean free path of 2.5 nm.

Figure 1 is a transmission electron micrograph of one of the Au rings measured in this experiment. The average diameter of the ring was 320 nm and the diameter of the lines forming the ring was approximately 45 nm. The grain size of the Au film from which the rings were made was roughly 100 nm, so that an electron typically encountered 4–5 grain boundaries as it traversed one arm of the ring.

The magnetoresistance of the ring in Fig. 1 is shown at several temperatures in Fig. 2. Below $T=1$ K, a complex structure appeared in the magnetoresistance that increased in amplitude to a few parts in 10^3 of the sample resistance below $T=0.1$ K. The structure did not shift in field as the temperature was varied, only the amplitude changed. In addition, it showed no sign of attenuation out to a field of 8 T. Similar results were also observed in the magnetoresistance of three other Au rings with the same average diameter, but with linewidths between 40 and 130 nm. The sharp drop in resistance near zero field in Fig. 2 is thought to be due to a superconducting proximity effect induced in the four-terminal device by Al wires bonded to Au pads 0.5 mm from the device. This effect disappeared at fields above 90 G, the critical field of the Al wire, and was not present when the Au sample was measured at a temperature of 1.5 K and above. The positive magnetoresistance above 90 G

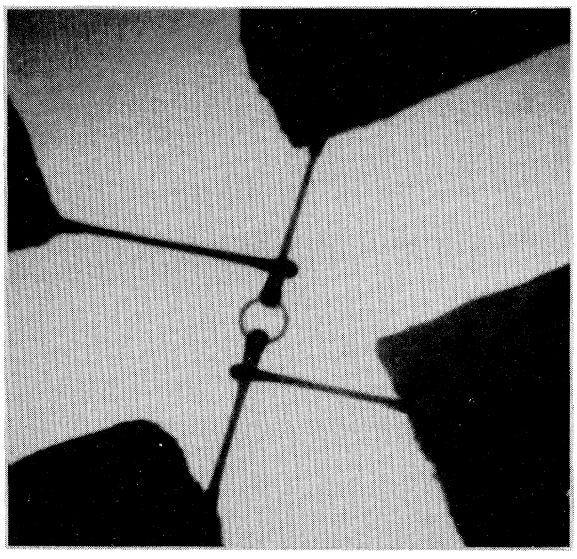

FIG. 1. Transmission electron micrograph of one of the Au rings measured in this experiment. The inside diameter of the ring was 280 nm and the width of the lines forming the ring was roughly 45 nm.

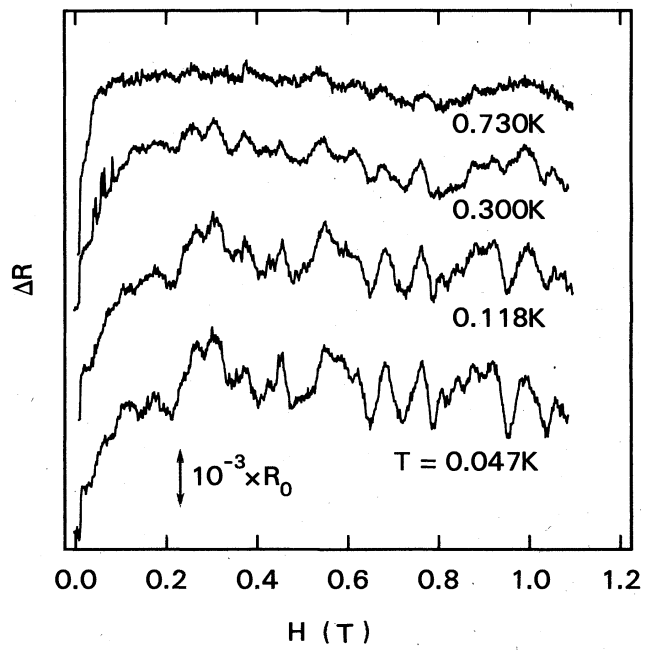

FIG. 2. Temperature dependence of the magnetoresistance from 0–1.2 T of the Au ring shown in Fig. 1. The zero-field resistance of the ring, R_0, was 7.7 Ω.

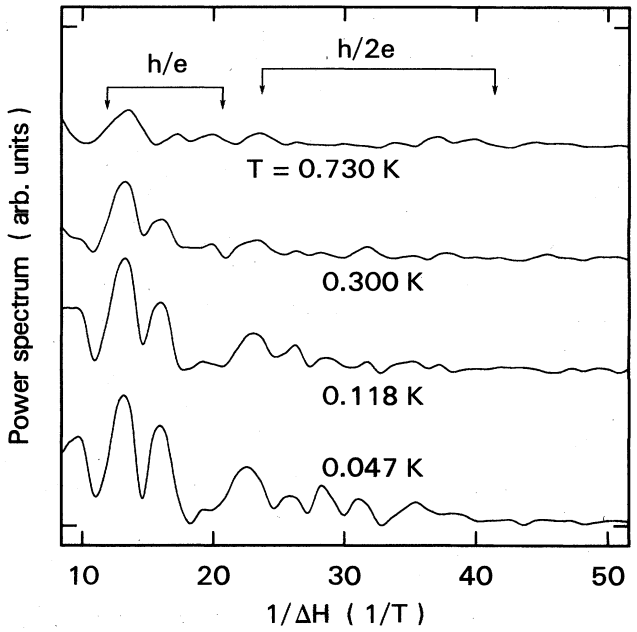

FIG. 3. Fourier power spectra of the magnetoresistance data shown in Fig. 2. The arrows at the top of the graph show where peaks produced by flux quantization with respect to h/e and $h/2e$ would occur based on the (from left to right) inside and average areas of the ring. The average area is one-half of the sum of the areas enclosed by the inside and outside perimeters of the ring. The uncertainty in the determination of the areas was roughly 15%.

may be the result of spin-orbit interactions in the Au sample, with a characteristic cutoff field of 0.2–0.3 T.[9] Above 0.3 T, the background magnetoresistance was slightly negative out to 8 T. (The background magnetoresistance was found, however, to vary from sample to sample.)

Flux quantization, with respect to $h/2e$, is predicted[1] to produce resistance oscillations of the amplitude seen in Fig. 2 if the inelastic scattering length is approximately 2 μm. Attempts to determine the actual inelastic scattering length in the samples were unsuccessful. At $H=0$, sample resistances generally rose logarithmically by a part in 10^4 per decade of decreasing temperature. This logarithmic temperature dependence implies that these samples were either two dimensional, with respect to localization,[10] or electron interaction[11] effects, or that there was a contribution to the resistance from magnetic impurities (possibly a Kondo effect[12]). Since the cross section of the wires comprising the samples was roughly square, it is very unlikely that the samples were ever two dimensional as far as electron diffusion is concerned. Qualitative secondary ion mass spectroscopy (SIMS) revealed that trace amounts of magnetic Fe and Cr were, in fact, present in the Au rings. The presence of magnetic impurities made a reliable estimate of the inelastic scattering length from localization in these samples impossible. A rough estimate for the inelastic length in the Au ring shown Fig. 1 can be obtained, however, by using the inelastic scattering times obtained by Bergmann[13] for 2D Au films, along with the mean free path of approximately 24 nm in the Au ring. The inelastic length $\{l_{\text{in}}=[(1/3)v_f l_{\text{el}} t_{\text{in}}]^{1/2}\}$ is calculated to be 0.47 μm at 4.5 K, slightly larger than the diameter of the ring.

In contrast to experiments on Mg and Li cylinders,[2–5] there was no unique period of oscillation in the magnetoresistance of the Au rings. Fourier power spectra of the magnetoresistance data in Fig. 2 are shown in Fig. 3. A number of peaks appear in the power spectra within the range of periods that could result from flux quantization with respect to h/e or $h/2e$ in the ring. Peaks at $1/\Delta H = 20.7\ \text{T}^{-1}$ and $41.4\ \text{T}^{-1}$ are expected using the average area encircled by the ring in Fig. 1 to define flux quanta of h/e and $h/2e$. The data show a peak near the value for h/e, but nothing near the value for $h/2e$. If the inside area of the ring is the relevant dimension for normal-metal flux quantization, there are two very suggestive peaks in the data. The peak at $13\ \text{T}^{-1}$ is near the value expected for a flux quantum of h/e, and the weaker peak near $23\ \text{T}^{-1}$ is near the value for $h/2e$. A strong h/e oscillation with a weaker $h/2e$ component is predicted by Ref. 7. In spite of this evidence for flux quantization, there are additional factors that must be considered. For example, there is a strong, reproducible peak at $16\ \text{T}^{-1}$ that is not included within either of the schemes mentioned above. In addition, it was found that the positions of the Fourier peaks could be changed by disconnecting and reconnecting the leads to the samples. This change was most likely caused by slight physical changes in the delicate fine-line samples induced by current transients. Under carefully maintained experimental conditions, however, the structure was reproducible, and its amplitude showed a $T^{-0.7}$ dependence above 100 mK. These two features of the data indicate that the structure did, in fact, have its origins in the transport properties of the sample, and was not the result of spurious instrumental effects. Curiously, when the magnetic field direction was reversed under carefuly maintained experimental conditions, both the complex structure in the magnetoresistance and the power spectra were found to be asymmetric with respect to zero field. As expected, the background magnetoresis-

tance, including the "spin-orbit" and "superconducting proximity" effects, was symmetric about $H=0$. The asymmetry with respect to zero field and the experimentally induced variability of the complex structure mentioned above indicate that the data displayed in Fig. 3 should probably not be interpreted as evidence for flux quantization in the Au rings.

Since theory[1] predicts that the amplitude of flux quantization resistance oscillations can increase with increasing sample resistivity (provided that the inelastic scattering length remains the same), $Au_{60}Pd_{40}$ rings with resistivities an order of magnitude higher than the Au rings were also measured. Nothing, however, was observed in the low-field magnetoresistance of these rings, although a complex structure did develop at higher fields. The "superconducting proximity effect" seen in the low-field magnetoresistance of the Au devices was also not observed in the $Au_{60}Pd_{40}$ samples. This is consistent with the much shorter mean free path in the $Au_{60}Pd_{40}$ samples.

Short, narrow, Au and $Au_{60}Pd_{40}$ lines, made from the same films as the rings, were also measured to provide a comparison to the results obtained from the rings. Figure 4 shows the magnetoresistance from 0–8 T of a 60-nm diam by 790-nm-long $Au_{60}Pd_{40}$ line at a variety of temperatures. Again, as the temperature decreased below $T=1$ K, apparently random, but reproducible, structure appeared in the magnetoresistance. The structure from the Au and $Au_{60}Pd_{40}$ lines was qualitatively the same as that observed in the rings, in that the density, the amplitude, and the temperature dependence of the structure were similar in devices made from the same material. In all samples, this structure persisted to magnetic fields of more than 8 T. The structure in the $Au_{60}Pd_{40}$ devices was somewhat less dense than it was in the Au devices. A further comparison to the results obtained from the rings and lines came from a measurement of the magnetoresistance of 2D samples (1400 μm$\times$10 μm) made from the same Au and $Au_{60}Pd_{40}$ films as the quasi-1D samples. At $T=0.3$ K the 2D magnetoresistance varied smoothly with field, with none of the complex structure seen in the rings and lines.

These results suggest that the complex structure observed in the Au and $Au_{60}Pd_{40}$ rings was primarily due to effects instrinsic to the lines making up the branches of the rings, and that any oscillatory contributions to the magnetoresistance arising from changes in the flux encircled by the entire ring were subordinate to them. In addition, the presence of the structure in small, quasi-1D devices that was not present in much larger 2D samples, indicates that it is related to the size of the small devices. If the structure is related to quasi-1D transport, then its growth out of a smooth background magnetoresistance as the rings and lines were cooled below $T=1$ K suggests that the inelastic scattering length became at least comparable to the width of the lines below $T=1$ K.

The origin of the complex structure seen in the small rings and lines measured in this experiment is a matter of conjecture at this time. Shubnikov–de Haas resistance oscillations will not occur in the samples at magnetic fields of less than a couple of hundred tesla (where $\omega_c\tau=1$). Dingle[14] has suggested that the finite spacing between electron energy levels occurring in systems consisting of a small

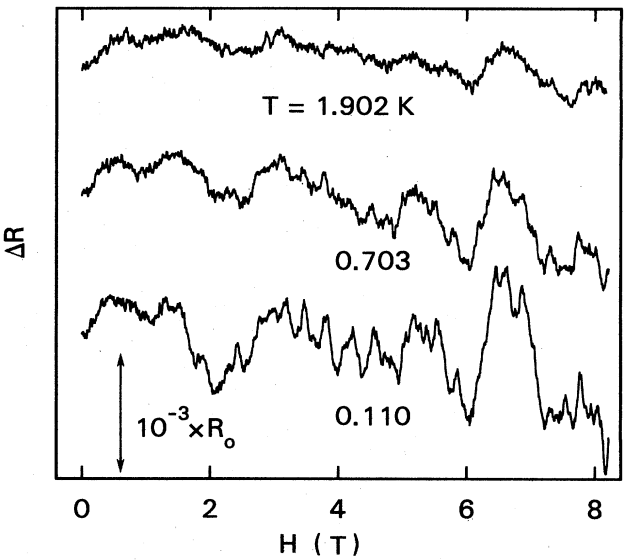

FIG. 4. Temperature dependence of the magnetoresistance from 0–8 T of a 60-nm-diam by 790-nm-long $Au_{60}Pd_{40}$ line. The zero-field resistance of the line, R_0, was 101.7 Ω.

number of atoms can lead to structure in the magnetoresistance; however, this theory must be refined to include the effects of electron scattering. In this case, the interpretation might be more naturally phrased in the language of transmission resonances[15] or of statistical fluctuations in small systems.[16] The large variations recently observed in the conductance of one-dimensional MOSFET's as the gate voltage was varied slightly may be the result of related processes.[17,18] Since the mean free path in the Au devices was limited by the diameter of the lines, the structure seen in the low-field magnetoresistance of the Au samples may also be related to the scattering of ballistic electrons at the surfaces and the finite number of grain boundaries in the samples. The $Au_{60}Pd_{40}$ devices did not exhibit this low-field structure, and, given the large scattering rate in these samples, would not be expected to, if the structure was the result of ballistic processes.

In summary, the magnetoresistance of very small Au and $Au_{60}Pd_{40}$ rings and lines has been measured at low temperatures. Below approximately $T=1$ K, a complex structure developed in the magnetoresistance of the devices. Suggestive, but not convincing, evidence for flux quantization in normal-metal rings was found. The structure seen in the magnetoresistance of the rings and lines seems to be related primarily to the small size of the devices. Recently, similar structure has also been observed in flux quantization experiments carried out using GaAs rings and lines.[19] Further study is required to elucidate the origin of this structure.

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[1]B. L. Al'tshuler, A. G. Aronov, and B. Z. Spivak, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 101 (1981) [JETP Lett. **33**, 94 (1981)].

[2]D. Yu. Sharvin and Yu. V. Sharvin, Pis'ma Zh. Eksp. Teor. Fiz. **34**, 285 (1981) [JETP Lett. **34**, 272 (1981)].

[3]B. L. Al'tshuler, A. G. Aronov, B. Z. Spivak, D. Yu. Sharvin, and Yu. V. Sharvin, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 476 (1982) [JETP Lett. **35**, 588 (1982)].

[4]F. Ladan and J. Maurer, C. R. Acad. Sci. Ser. B **297**, 227 (1983).

[5]M. Gijs, C. van Haesendonck, and Y. Bruynseraede, Phys. Rev. Lett. **52**, 2069 (1984).

[6]Y. Aharonov and D. Bohm, Phys. Rev. **115**, 485 (1959).

[7]Y. Gefen, Y. Imry, and M. Ya. Azbel, Phys. Rev. Lett. **52**, 12 (1984); in *Proceedings of the Fifth International Conference on the Electronic Properties of Two-Dimensional Systems, Oxford, England —1983* [Surf. Sci. **41**, 203 (1984)], p. 294; M. Buettiker, Y. Imry, and R. Landauer, Phys. Lett. **96A**, 365 (1983).

[8]R. B. Laibowitz and A. N. Broers, in *Treatise on Materials Science, Vol. 24,* edited by K. N. Tu and R. Rosenberg (Academic, New York, 1982), p. 285; W. W. Molzen, A. N. Broers, J. J. Cuomo, J. M. E. Harper, and R. B. Laibowitz, J. Vac. Sci. Technol. **16**, 269 (1979).

[9]S. Hikami, A. I. Larkin, and Y. Nagaoka, Prog. Theor. Phys. **63**, 707 (1980).

[10]E. Abrahams, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).

[11]B. L. Al'tshuler, A. G. Aronov, and P. A. Lee, Phys. Rev. Lett. **44**, 1288 (1980).

[12]See, for example, M. D. Daybell, in *Magnetism, Vol. V,* edited by H. Suhl (Academic, New York, 1973), p. 121.

[13]G. Bergmann, Phys. Rev. B **28**, 515 (1983).

[14]R. Dingle, Proc. R. Soc. London, Ser. A **212**, 47 (1952).

[15]M. Ya. Azbel and P. Soven, Phys. Rev. B **27**, 831 (1983).

[16]P. A. Lee (private communication).

[17]A. B. Fowler, A. Hartstein, and R. A. Webb, Phys. Rev. Lett. **48**, 196 (1982).

[18]R. F. Kwasnick, M. A. Kastner, J. Melngailis, and P. A. Lee, Phys. Rev. Lett. **52**, 224 (1984).

[18]G. Blonder, Bull. Am. Phys. Soc. **29**, 535 (1984).