Magnetic Flux Quantization in the Weak-Localization Regime of a Nonsuperconducting Metal

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The magnetic flux quantization effect, with period $\phi_0 = hc/2e$, is observed with high accuracy in the resistance of a Mg honeycomb network at temperatures 50 mK $\leq T \leq$ 6 K. As expected, the phase of the oscillations and the sign of the magnetoresistance are dominated by the spin-orbit interaction. Our results confirm, for a new geometry, the reality of the Bohm-Aharonov effect for a nonsuperconducting metal in the weak-localization regime.

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As a by-product of Anderson localization studies, it was shown by Altshuler, Aronov, and Spivak[1] (referred to as AAS hereafter) that the conductivity of a thin-walled cylinder made of dirty metal must be an oscillatory function of the magnetic flux ϕ which penetrates the cylinder. According to AAS, such oscillations of period $\phi_0 = hc/2e$ (the superconducting flux quantum), associated with twice $(2e)$ the electronic charge, can occur in a system of noninteracting electrons that undergo multiple random elastic collisions, as they move along the trajectories within the cylindrical cavity. This Bohm-Aharonov effect, analogous to the magnetic-flux quantization in superconductors, is expected to occur even when the electrons collide often with defects, i.e., when $l_e << L$ ($L =$ perimeter of the cylinder, $l_e =$ elastic mean free path). The low sensitivity to the degree of disorder and the period ϕ_0 of the predicted oscillations distinguishes this phenomena from Dingle oscillations, which could occur only if $l_e >> L$ and with a period $2\phi_0$. The condition for observing the effect is a sufficiently long diffusion length L_ϕ compared to L. L_ϕ can be expressed as $L_\phi = (l_e l_\phi)^{1/2}$ where l_ϕ is the mean free path for processes that destroy the phase coherence (inelastic processes, scattering by magnetic impurities).

Since the publication of Ref. 1, only two groups were able to observe the AAS effect, in spite of various attempts in different laboratories and serious doubts about the reality of this phenomenon. These measurements were performed on the resistance of long hollow cylinders (length ~ 1 cm, diameter $\sim 1\ \mu$m, thickness ~ 1000 Å) under longitudinal magnetic field at liquid helium temperature. Oscillations of the magnetoresistance were observed in both Mg[2] and Li[3,4] cylinders.

In this Letter we present new evidence for the reality of the AAS effect, using a new geometry and performing measurements over a large temperature range. Before going to the experimental results, let us discuss the physical origin of the AAS effect. According to quantum mechanics, the wave function and the energy spectrum of charged particles depends on the vector potential $\vec{A}$ of the magnetic field. The origin of the AAS effect is the interference of the quantum corrections to the classical conductivity of a normal metal. The interference of most of the amplitudes is not important since the lengths of the trajectories, and hence their phases, are very different, leading to a vanishing interference term. The exception are the trajectories which intersect themselves. Each such broken trajectory can be associated with *two* electronic states, corresponding to passages around the closed loop in opposite directions. These two amplitudes are mutually coherent, so that their interference cannot be neglected. Therefore, the quantum correction is given essentially by the probability of self-intersection for classical diffusion and then a small but finite correction is expected. If a magnetic field is switched on, the phase length of the trajectory is different for different alternative paths. However, the phase difference for all trajectories in a thin-walled cylinder is the same. This produces oscillations of the sample resistance with the period $\phi_0 = hc/2e$: For more extensive discussion on these interference effects, we direct the reader to the illuminating recent paper of Bergmann.[5]

In order to observe this effect, magnesium was chosen for various reasons, particularly because the relevant scattering rates (elastic, inelastic, spin-orbit, . . .) have already been the object of quantitative studies by different groups.[6,7] The samples were prepared by thermal evaporation of very pure Mg films onto the polished surface of a microchannel glass plate. The sample contains about 2.7×10^6 identical hexagonal loops which form a two dimensional (2D) regular honeycomb network whose parameters are determined by the characteristic increases sharply with field. $R(H)$ turns around at perimeter $\sim 9\ \mu$m, loop area $6.0\ \mu m^2$, strand width $\sim 0.5\ \mu$m. The fabrication procedure is similar to that described in a previous paper.[8] In order to make the diffusion length as large as possible compared to the loop perimeter ($9\ \mu$m), the greatest care was taken to optimize sample purity, in particular with respect to magnetic impurities. For that, the initial material was purified by vacuum distillation[9] until the residual concentration of magnetic impurities (Fe, Co, Mn) became less than 1 ppm. On the other hand, the evaporated film was made thick enough (800 Å) to make boundary scattering unimportant. The obtained film had a sheet resistance of order of 0.15 $\Omega/\square$ at 4.2 K and a residual resistivity ratio $R_{300}/R_{4.2}=3.1$, from which we deduce the elastic mean free path $l_e \cong 520$ Å. This value corresponds to $k_F l_e \cong 700$ which is characteristic of the (very) weak-localization regime (k_F= Fermi wave vector). Although the relative amplitude of the AAS oscillations is expected to increase with disorder, it must be realized that the most severe limitation on observation of this effect comes from the exponential damping: $\exp(-L/L_\phi)$ as the loop perimeter L becomes larger than L_ϕ. The results shown below demonstrate that L_ϕ is large enough ($>> 1\ \mu$m) to minimize this damping. Compared to L_ϕ, the lateral size of the Mg strands is small. Therefore, with respect to the localization problem, the sample can be considered as a network of purely one-dimensional Mg wires.

The sample was attached to a copper block thermally linked to the mixing chamber of a dilution refrigerator. A thermal regulation using a calibrated Ge thermometer sensor allows control of the temperature from 50 mK to 7.5 K with an accuracy better than 1 mK. Resistance measurements were carried out by use of a four-terminal bridge with typical bias currents ranging from 20 μA to 1 mA. Optimum matching of the bridge to the sample resistance (0.791 Ω) was realized by a transformer of ratio 326 held in the liquid helium bath. Ultimate rms noise of our system is $\sim 3\times 10^{-11}$ V/Hz$^{1/2}$, which is close to the Johnson noise of the sample resistance. A further improvement by one order of magnitude in the signal-to-noise ratio was obtained by multichannel averaging of the signal, using a microcomputer.

Figures 1 and 2 show, for several temperatures T, the dependence of the resistance on the strength H of the magnetic field perpendicular to the plane of the network. First let us discuss the global features of the resistance as function of T and H. At low temperatures (see $T=0.16$ K), the resistance first

dimensions of the microchannel substrate: loop $H=H^*=12.6$ Oe and then decreases more slowly. This dip at zero field, which is due to spin-orbit scattering, becomes less pronounced as the temperature increases and eventually disappears around 6 K as the inelastic scattering rate $1/\tau_\phi$ becomes greater than the spin-orbit scattering rate $1/\tau_{so}$. These features are characteristic[6,7] of the crossover from the regime dominated by spin-orbit scattering $\tau_{so} << \tau_\phi$ (low temperature, positive magnetoresistance) to the regime dominated by inelastic scattering $\tau_\phi << \tau_{so}$ (high temperature, negative magnetoresistance) as the temperature is increased. The temperature dependence of the magnetoresistance will not be discussed further here. A more detailed

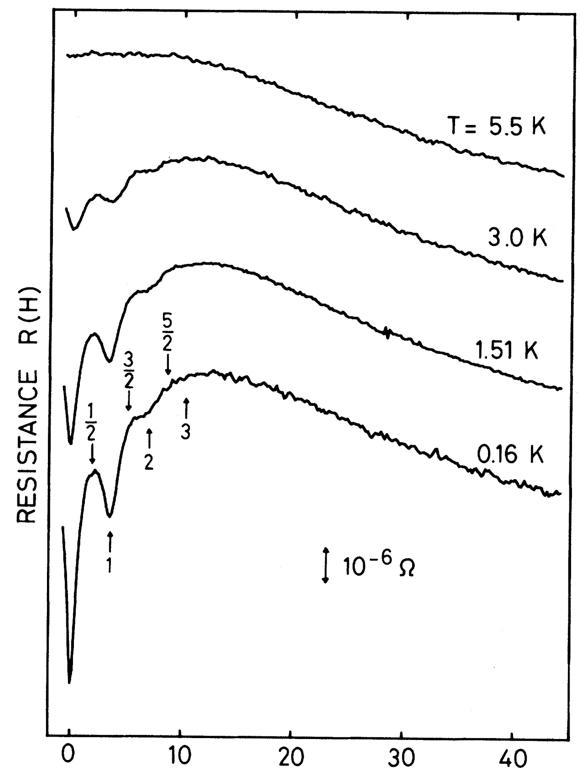

FIG. 1. Magnetoresistance $R(H)$ of a Mg honeycomb network containing about 2.7×10^6 hexagonal elementary cells (of side $a \cong 1.5\ \mu$m) at various temperatures. In the low-field region, $R(H)$ exhibits oscillations with a period corresponding exactly to one flux quantum (ϕ_0) per unit cell. Arrows show the values of the magnetic field corresponding to the reduced flux $\phi/\phi_0=\frac{1}{2}$, 1, $\frac{3}{2}$, 2, $\frac{5}{2}$, and 3, as determined from measurements on a superconducting network of identical geometry (see text). The zero-field resistances at 0.16, 1.51, 3.0, and 5.5 K are respectively 0.791 369 Ω, 0.791 363 Ω, 0.791 370 Ω, and 0.791 434 Ω. Note that the vertical scale is the same for all temperatures.

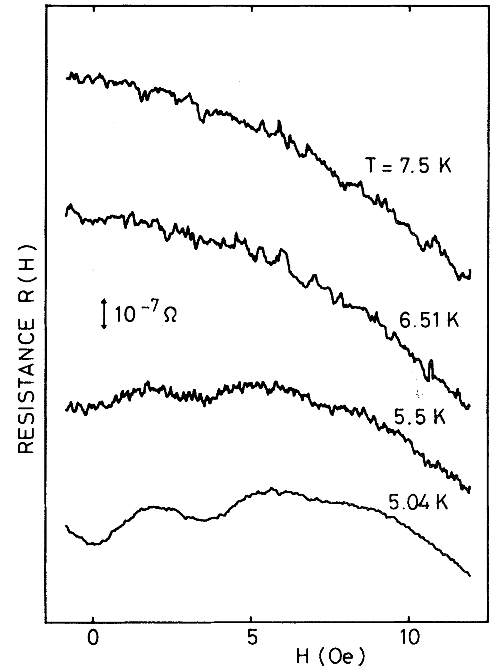

FIG. 2. The same plot as in Fig. 1, in the region of high temperatures $T \geqslant 5$ K. For $T \gtrsim 6.5$ K, a negative magnetoresistance is recovered. In this regime, the oscillations are small and experimentally invisible. Note for that the change of the ordinate scale.

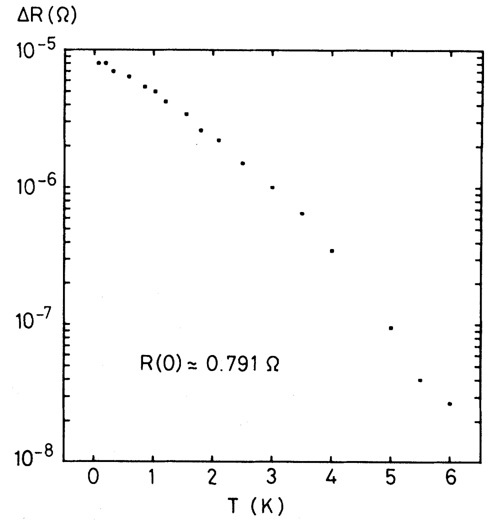

FIG. 3. Temperature dependence of the magnitude associated with the first oscillation $\Delta R(H) = R(H) - R(0)$ at $\phi/\phi_0 = \frac{1}{2}$ in a semilog plot. At $T \gtrsim 6$ K, the oscillations become outside our experimental accuracy.

discussion of features such as antilocalization and electron-electron effects will be reported in a forthcoming paper.[10]

More interesting, in addition to the above features, Figs. 1 and 2 show the apparition of large magnetoresistance oscillations at low fields and low temperatures. These oscillations are observed for both polarities of the field and their phase corresponds to the resistance being minimum at $H = 0$. We interpret this modulation as the manifestation of the AAS effect. The magnetic period is equal to 3.53 Oe and does not depend on the temperature. Given the measured mean loop area $S = 6.0 \pm 0.3$ μm^2, the magnetic period corresponding to exactly one flux quantum per unit loop should be 3.45 ± 0.2 Oe, which is very close to the measured value. A definitive check is provided by a direct comparison of the magnetic period observed in the magnesium sample to that measured for a superconducting sample having *exactly* the same geometry. This was easily realized by evaporating a superconducting In film on the microchannel substrate instead of the Mg. A very accurate determination of the superconducting quantum flux has so been obtained. We find for the present case 3.59 Oe which compares very well to the Mg case. Therefore we claim that, within our experimental accuracy $\sim 2\%$, the flux quantum associated with the AAS effect is the same as the superconducting flux quantum $\phi_0 = hc/2e$ and therefore involves twice the electronic charge.

The oscillations are clearly observed up to $H = H^*$ and they are rapidly smoothed out with further increase of the field. The maximum amplitude is observed at the lowest temperatures: $\Delta R \cong 8 \times 10^{-6}$ Ω at 50 mK. This corresponds to a change in conductance $\Delta R/R^2 \sim 1.3 \times 10^{-5}$ Ω^{-1}, value which must be compared to the conductance scale $e^2/h = 3.9 \times 10^{-5}$ Ω^{-1} of the quantum correction associated with weak localization. Figure 3 shows the peak-to-peak amplitude $[\Delta R = R(\phi_0/2) - R(0)]$ of the oscillation as function of the temperature. The rapid decrease at high temperature is mainly due to the temperature dependence of the diffusion length L_ϕ. Above 6 K, the oscillations become unobservable.

Complete quantitative analysis of these results requires the development of a full theory of the AAS effect in a 2D network. However, a rough analysis of the present data allows some preliminary estimations: The spin-orbit diffusion length is directly related to the crossover field H^* at low temperature. We find $H^* = 12.6$ Oe at $T = 0.16$ K leading to $(D\tau_{so})^{1/2} \simeq 0.97$ μm and then $\tau_{so} = 3.4 \times 10^{-11}$ sec. In the temperature range where positive magnetoresistance is observed ($T \lesssim 6$ K), τ_ϕ is larger than τ_{so}. This means that $L_\phi = (D\tau_\phi)^{1/2}$ is much greater than 0.97 μm at low temperature, which is consistent with the observation of the AAS effect, given the loop perimeter 9 μm. Here D ($=280$ cm^2/sec) denotes the free-electron diffusion constant of our Mg sample.

A direct comparison with flux-quantization phenomena in a superconducting network is provided by the results reported in Ref. 8. It is worth noticing that, in the superconducting case, flux quantization causes the sample resistance to oscillate between zero and the normal-state value as the field is increased. This effect, which is observed close to the transition temperature T_c, is found to vanish far inside the superconducting phase as well as above T_c, in the region of superconducting fluctuations. In contrast, the effect reported in this paper is very small and is observed over a very large range of temperature, from 6 K down to our lowest controllable temperature, 50 mK. We think that superconducting fluctuations are unlikely to produce quantization effects over such a large temperature range (2 orders of magnitude). Moreover, no trace of superconductivity was observed in our Mg samples down to 50 mK.

In summary, we have confirmed experimentally the existence of the magnetic flux quantization effect, as predicted by AAS for normal metals, on a honeycomb Mg network. The observed oscillations of the resistance at low temperatures 50 mK $\leq T \leq 6$ K and low magnetic field ($H \lesssim 12$ Oe) correspond exactly to a change in the magnetic flux in the elementary unit cell of the network by the amount of a flux quantum $\phi_0 = hc/2e$. The observed phenomenon results from a weak localization effect and therefore must be contrasted with the analog flux quantization in superconductors. Presently there is no theory for a quantitative analysis of the AAS effect in an extended network. The extension of the theoretical results of the single-loop problem[9] to the network case is now under active consideration.[10]

After this work was submitted we learned that Gijs, Van Haesendonck, and Bruynseraede[11] have observed resistance oscillations of hollow Mg cylinders at liquid helium temperatures. In the meantime we have observed oscillations similar to those reported here in Cu and Au samples and this will be described elsewhere.

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