# Magnetization of Mesoscopic Copper Rings: Evidence for Persistent Currents

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We have measured the low-temperature magnetization response of $10^7$ *isolated* mescoscopic copper rings to a slowly varying magnetic flux. At sufficiently low temperature, the total magnetization response oscillates as a function of the enclosed magnetic flux on the scale of *half a flux quantum*. The amplitude of the oscillatory moment is $\approx 1.2\times10^{-15}$ A m$^2$ and decreases exponentially with increasing temperature on the scale of the correlation energy $E_c = hD/(2L)^2 \approx 80$ mK. This is evidence for a flux-periodic persistent current in each ring of average value $3\times10^{-3} ev_F/L$.

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An *isolated* normal-metal ring threaded by a magnetic flux $\phi$ is thought to carry an equilibrium current at finite temperature as long as the electron phase coherence is preserved.[1] In a ring geometry, the potential produced by random impurities on each electron repeats periodically after each revolution around the ring. The Bloch wave vector $k_n$ associated to this periodicity depends on the enclosed flux. Using gauge invariance $[A \rightarrow A+\nabla f, \psi_n \rightarrow \psi_n \exp(2i\pi f/\phi_0)]$, the vector potential can be eliminated from the Schrödinger equation and introduced as a modification of the boundary condition,

$$\psi_n(x+L) = \exp\left(2i\pi\frac{\phi}{\phi_0}\right)\psi_n(x)\,, \tag{1}$$

where $\phi_0 = h/e$ is the flux quantum and $L$ is the ring circumference. Then, the Bloch wave vector $k_n$ is determined by this flux-dependent boundary condition: $k_n = (2\pi/L)(n+\phi/\phi_0)$.[2] In turn this imposes a periodicity of the energy spectrum $\epsilon_n$ and related quantities with the flux quantum. For example, state $n$ has a velocity $v_n = \partial\epsilon_n/\hbar\partial k_n = (L/e)\partial\epsilon_n/\partial\phi$ and therefore carries a current $j_n = -ev_n/L = -\partial\epsilon_n/\partial\phi$, periodic with the flux quantum. This implies a change $E_c$ of the total energy $E$ as the flux is increased from 0 to $\phi_0/2$ and a net current $j = -\partial E/\partial\phi$. The correlation energy $E_c$ represents the sensitivity to a change between periodic and antiperiodic boundary conditions [Eq. (1)].[3] When the thermal energy $k_BT$ is less than $E_c$, it is plausible that wave functions may have sufficient "rigidity" to maintain a nonvanishing current $j = -\partial F/\partial\phi$ in thermal equilibrium ($F$ is the free energy).[4]

In a metal the electrons diffuse among impurities and the persistent current fluctuates in sign and magnitude depending on the realization of disorder and the number and electrons.[5] Furthermore, isolated rings have a fixed number of particles: This constraint induces a subtle dependence of the chemical potential on magnetic flux which dominates the ensemble-averaged properties of the current $\langle j\rangle$.[6] On the other hand, the magnitude of the current[5,6] $\langle j^2\rangle^{1/2}$ is insensitive to averaging issues and in the diffusive regime is of the order of the current carried by one electron,[7] $\approx e/\tau_D$, where $\tau_D \approx L^2/\lambda_e v_F$ is the time it takes the electron to diffuse around the ring ($\lambda_e$ is the elastic mean free path). We can estimate the energy sensitivity $E_c$ to a change in boundary condition with the uncertainty-principle argument,[3] $E_c = h/\tau_D$. This gives the same estimate $\delta E/\delta\phi = 2E_c/\phi_0 = 2e/\tau_D$ for the one-electron current. For a copper ring with $L \approx 2.2$ $\mu$m and $\lambda_e \approx 200$ Å, this current is of the order of $10^{-2} ev_F/L \approx 1.2$ nA and induces a magnetic moment $\mu_j = 4.5\times10^{-22}$ A m$^2$ $\approx 50$ Bohr magnetons.

In this Letter, we report a magnetization measurement on $N = 10^7$ disconnected copper rings showing evidence for a flux-periodic persistent current. We stress the main features of the data: The moment oscillates on a scale of $\phi_0/2$ (*not* $\phi_0$). The total oscillatory moment $\mu$ is of the order of $0.3N\mu_j$ and not $(N\mu_j^2)^{1/2}$; i.e., there is a nonzero average moment $\langle\mu_j\rangle$.

The "rings" studied are actually squares which are more convenient for fabrication with the electron-beam techniques employed.[8] The ring dimensions (side, $\approx 0.55$ $\mu$m; circumference, $L \approx 2.2$ $\mu$m; area, $\approx 0.3$ $\mu$m$^2$) were chosen to have phase coherence, on one hand ($L_\phi > L$) and well-defined area (corresponding to one flux quantum every 130 G), on the other hand. $L_\phi$ was measured to be in excess of 2 $\mu$m at 1.5 K by observing the Aharonov-Bohm oscillation in the resistivity of a network made with the same copper. The $10^7$ rings occupy a 7-mm$^2$ area on the sapphire substrate which is mounted at the end of a 3-cm-long single-crystal quartz rod linked to the dilution refrigerator through a 60-n$\Omega$ annealed silver structure. The magnetization is measured with an ultrasensitive SQUID magnetometer which was calibrated against a well-characterized spin-glass film (CuMn, $\chi = 2\times10^{-4}$ emu/g). The 3-mm-diam pickup coil is situated 0.3 mm from the rings. A slowly varying (0.3-Hz) magnetic field is applied by a Ni-Ti superconducting solenoid. A center tap on this modulation coil allows nulling of the linear response of the magnetometer *in situ*. A static magnetic field can be trapped in a cylindrical Ni-Ti superconducting shield which surrounds all components. The SQUID flux transformer, the modulation coil, and the shield are thermally linked with sapphire and silver. This structure is thermally decoupled from the refrigerator by an SP22 vespel structure; in this way, all superconducting parts of the apparatus can be held at a fixed temperature while the sample temperature is varied (from 7 to 400 mK).

The total moment induced by the persistent currents can be distinguished from that due to paramagnetic impurities through its periodic dependence on applied magnetic flux. If a slowly[9] time-varying flux $\phi=\phi_{dc}+\phi_{ac}\times\sin(\Omega t)$ is applied, the Fourier expansion of the induced moment $\mu(t)$ is

$$\mu(t)=\sum_{n=0}\{\mu_{2n}(T)\cos(2n\Omega t)+\mu_{2n+1}(T)\sin[(2n+1)\Omega t]\}\,. \quad (2)$$

If the average moment is assumed to be a sine function of the flux with period $\phi_p$ and amplitude $\mu_a(T)$, the Fourier amplitudes $\mu_n$ can be computed as

$$\mu_{2n}(T)=2\mu_a(T)J_{2n}(\delta)\sin\theta\,,$$
$$\mu_{2n+1}(T)=2\mu_a(T)J_{2n+1}(\delta)\cos\theta\,, \quad (3)$$

where $\delta=2\pi\phi_{ac}/\phi_p$ and $\theta=2\pi\phi_{dc}/\phi_p$. The Fourier amplitudes $\mu_n$ in the frequency domain are not to be confused with the Fourier components of the magnetic moment in harmonics of the flux. When the ac amplitude is sufficiently large ($\approx\phi_p/4$), higher harmonics ($3\Omega,\ldots$) appear as modulation sidebands of the measured signal as illustrated in Fig. 1.[10] On the other hand, paramagnetic impurities contribute predominantly to the fundamental ($\Omega$) response provided that the Zeeman splitting $\mu_B B_{ac}$ remains small compared to the temperature $k_B T$. This provides a very effective background rejection. Phase-sensitive detection is achieved by synchronous digitization of the signal during an integral number of periods

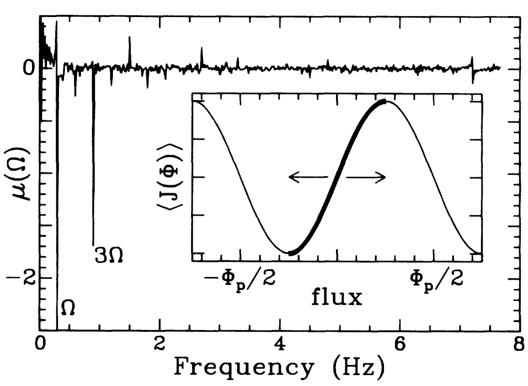


FIG. 1. Fourier transform of the in-phase magnetization signal modulated at 0.3 Hz by a sine wave of 15-G amplitude. The sample temperature is $T=25$ mK, while the magnetometer is held at $T=450$ mK. Inset: Schematic dependence of the average current with flux. A flux modulation of $\pm\phi_p/4$ (arrows and highlighted region of the curve) produces the desired nonlinear response.

($\sim 20$) followed by Fourier analysis. The odd harmonics are in phase with the applied flux while even harmonics are 90° out of phase because of the dissipation-free nature of the persistent currents.

In Fig. 1, the Fourier transform of the in-phase response to an ac magnetic field of amplitude 15 G $\approx\phi_0/8$ shows a clear third-harmonic signal which is independent of the measuring frequency from $10^3$ to 10 Hz. In the course of this study, two spurious backgrounds had to be eliminated. (a) A sizable nonlinear paramagnetism ($\propto 1/T^3$) originated from the Formvar insulation of the Ni-Ti pickup coil. (b) Small ($\approx 1000$ Å) titanium ($T_c=0.39$ K) inclusions on the surface of the Ni-Ti wire used in the flux transformer gave rise to a nonlinear response (incomplete Meissner effect). By temperature regulating the magnetometer at $T\sim 450$ mK above $T_c$(Ti) both effects could be reduced to a sufficiently small level. That is demonstrated in trace (a) of Fig. 2: Without a sample the residual second and third harmonics show no temperature dependence.[11] Thus, no residual nonlinear paramagnetic background ($\propto 1/T^3$) is detected at our level of sensitivity. This is contrasted with the clear temperature dependence of the third harmonic observed when a sample is loaded [Fig. 2, trace (b)]. Furthermore, the same signal is observed when the magnetometer is regulated at 450 and 700 mK (circles and triangles) and extrapolates to the same zero-temperature limit.

Before we analyze the decrease of the signal with increasing temperature, we qualitatively describe the oscillatory behavior of the second and third harmonics at $T=0$ with static magnetic field. Traces (c), (d), and (e) of Fig. 2 show the temperature dependence of the second and third harmonics when a dc field of 15 G ($\phi_0/8$), 30 G ($\phi_0/4$), and 60 G ($\phi_0/2$) is trapped in the superconducting shield. At $\phi_0/8$, the $T=0$ amplitude of the third-harmonic signal goes through zero, while the second harmonic reaches its maximum negative value. This behavior is accounted for by Eqs. (2) and (3) provided that $\theta=\pi/2$ when $\phi_{dc}=\phi_0/8$, i.e., $\phi_p=\phi_0/2$. At $\phi_0/4$, the $T=0$ third harmonic has changed sign ($\theta=\pi$) while the zero-field signal repeats at $\phi_0/2$ ($\theta=2\pi$). This oscillatory behavior on the scale of half a flux quantum is indicative of persistent currents. We now turn to the observed temperature dependence.

At $T\neq 0$, electron wave functions are wave packets of finite width which spread by diffusion. Sensitivity to a change in boundary condition is expected to be lost when the spread in the electron phase during a path around the ring exceeds the total phase acquired. This takes place when the diffusion length $L_T=(hD/kT)^{1/2}$ ($D$ is the diffusion coefficient) is of the order of $2L$, since a quasiclassical wave packet must enclose the flux twice to have a $\phi_0/2$ periodicity. Persistent currents are therefore expected to decrease exponentially ($\exp[-(2L/L_T^2)]$ for a diffusive process) with temperature on a scale $E_c(\phi_0/2)$

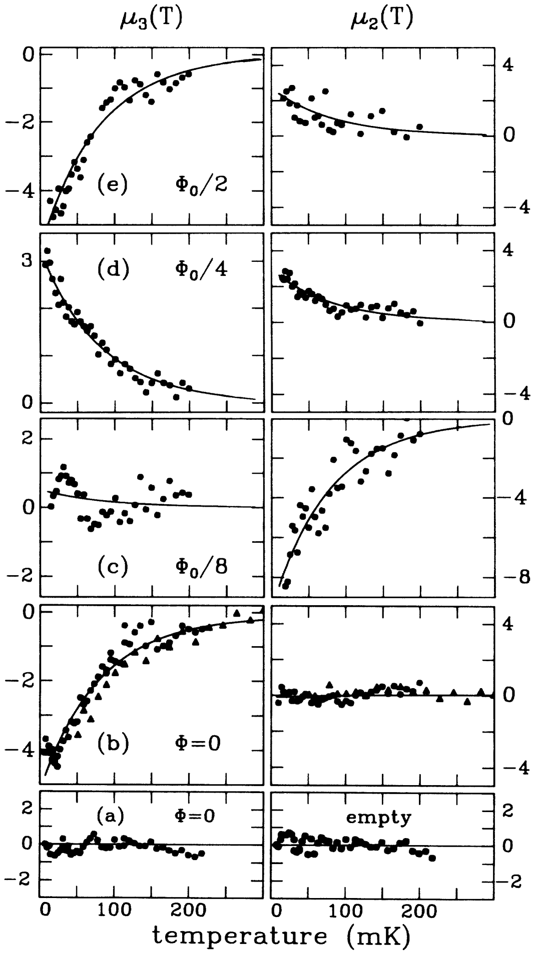


FIG. 2. Temperature dependence of the second ($\mu_2$, right column) and third ($\mu_3$, left column) harmonics (plotted in arbitrary units) of the magnetization response using a 15-G ac drive. (a) Empty magnetometer, (b) sample with no dc field, (c) 15-G dc field, (d) 30-G dc field, and (e) 60-G dc field. The solid lines are one-parameter fits by $\mu_{2,3}(0)\exp(-kT/E_c)$ adjusting only $\mu_{2,3}(0)$.

$=h/4\tau_D=hv_F\lambda_e/(2L)^2$. From resistance measurements on wires of similar aspect ratio as the ring sample, we infer $\lambda_e\approx 200$ Å and $E_c(\phi_0/2)/k_B\approx 80$ mK. Once $E_c$ is known, the only parameter to be fitted becomes the zero-temperature amplitude of the second and third harmonics, $\mu_{2,3}(T)=\mu_{2,3}(0)\exp(-k_BT/E_c)$. For each value of the dc magnetic flux, this one-parameter fit is plotted in Fig. 2 as solid lines. Within the experimental noise, this exponential behavior is nicely verified.[12] This sharp dependence gives the most direct determination of a correlation energy.

With the fitting procedure just described, we can quantitatively study the dependence of the fitted zero-temperature signal as a function of the applied dc field. The measured dependences of the second and third harmonics are plotted in Fig. 3 over a full flux quantum.

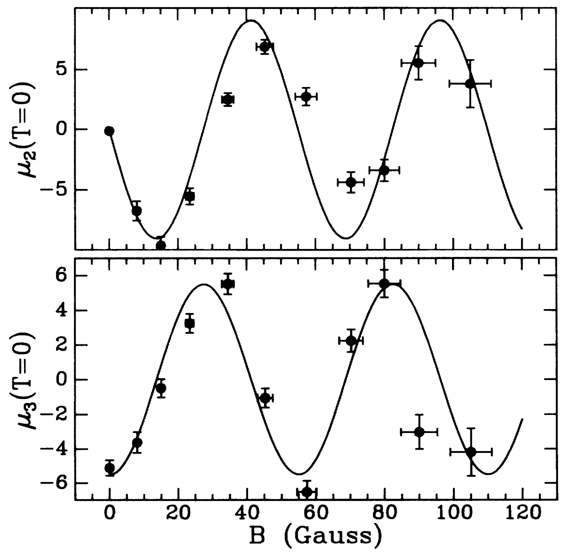


FIG. 3. Dependence of the fitted second- and third-harmonic amplitudes at $T=0$ with dc field as detected with a 15-G amplitude. 130 G correspond to one $\phi_0$. The horizontal error bars arise from the uncertainties in the field compression induced by the superconducting shield. Vertical error bars are statistical.

Both harmonics oscillate with magnetic field on a half-flux-quantum scale. Furthermore, as we hinted earlier, their field dependences are 90° out of phase; i.e., the second harmonic is maximum when the third harmonic goes through zero [Eq. (3)]. The ratio to the second-to-third-harmonic amplitude is $\sim 2$, close to the ratio of Bessel functions $J_2(\pi/2)/J_3(\pi/2)\approx 2.8$ expected from Eq. (3). The magnitude of the oscillatory moment is estimated (within a factor of 2) to be $\sim 1.2\times 10^{-15}$ A m$^2$ corresponding to an ensemble-averaged current of 0.4 nA $\approx 3\times 10^{-3}ev_F/L$ per ring. These are quite compelling facts in favor of persistent currents. With one and a half oscillations, we can only state that the Fourier amplitude of the oscillatory moment at half flux quantum is sizable but other harmonics may be present. To relate the sign of the third harmonic to the sign of the average current requires making specific assumptions about the functional dependence of the average moment with magnetic field. A sine function with $\phi_0/2$ periodicity [cf. Eq. (3)] implies a negative moment $\mu_a(T)$ and leads to a *diamagnetism* at low fields. Other assumptions can lead to other sign assignments.

The sign of the current is *a priori* a random quantity depending on the realization of the disorder and the number of electrons. One would expect the net moment of $N$-independent rings to be dominated by fluctuations $\propto (N\langle j^2\rangle)^{1/2}$ and retain a periodicity of $\phi_0$. In a mesoscopic system, this is *only* true if the chemical potential is held constant by a particle reservoir.[5] In our experiment the number of particles in each ring is fixed. It has been demonstrated in *exact* one-dimensional models[4] and in large-scale numerical simulations[6] that the ensemble average scales in this instance with the numbers of rings and acquires a $\phi_0/2$ periodicity. While clearly established, the physical origin of this period halving remains unclear. Experimentally, the amplitude of the observed oscillatory moment $\mu \approx 0.3N\mu_j$ is too large to be attributed to fluctuation. The measured average current of $0.3\langle j^2\rangle^{1/2}$ per ring seems to agree reasonably well with the canonical ensemble average in the diffusive regime of $0.1\langle j^2\rangle^{1/2}$ obtained in Ref. 6. On the other hand, simulations show a positive average current (*paramagnetism*). This sign is opposite to our tentative assignment based on Eq. (3) (*diamagnetism*). We now propose a possible explanation for this sign difference.

Our weak-localization measurements indicate that the spin-orbit scattering length $\approx 3000$ Å is less than the perimeter. In this limit, the spin-orbit interaction induces a rotation of the spin as the electron goes around the ring which must also be periodic. A spin rotation of $2\pi$ around an arbitrary axis changes the phase of the wave function by a factor $\exp(i\nu\sigma)$, where $\sigma = \pm\frac{1}{2}$ is the initial spin orientation and $\nu$ depends on the actual spin motion. It has recently been shown[13] that after averaging over all possible spin rotations the first harmonic of the current ($\phi_0$) averages out to zero, while the second harmonic ($\phi_0/2$) is multiplied by a factor of $-\frac{1}{2}$ (assuming it is nonzero after an ensemble average). The weak-localization corrections to the conductivity change sign for the same reason that the spin-orbit scattering length decreases.[14]

The physical understanding of the phenomena we have described remains incomplete. The symmetries of the Hamiltonian unbroken by disorder (time reversal) are likely to play a central role in explaining the physics we have described.

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*Note added.*—After this paper was completed, an interesting interpretation of our experiment relying on the electron-electron interaction along a time-reversed path was proposed.[15]

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[1]M. Büttiker, Y. Imry, and R. Landauer, Phys. Lett. **96A**, 365 (1983); R. Landauer and M. Büttiker, Phys. Rev. Lett. **54**, 2049 (1985); F. Bloch, Phys. Rev. **137**, A787 (1965).

[2]This argument is related to the Aharonov-Bohm effect and leads to the quantization of the fluxoid in superconductors; Y. Aharonov and D. Bohm, Phys. Rev. **115**, 485 (1959); N. Byers and C. N. Yang, Phys. Rev. **7**, 46 (1961).

[3]J. T. Edwards and D. J. Thouless, J. Phys. C **5**, 807 (1972); D. J. Thouless, Phys. Rev. Lett. **18**, 1167 (1977).

[4]H. F. Cheung, Y. Gefen, E. K. Riedel, and W. H. Shih, Phys. Rev. B **37**, 6050 (1988); N. Trivedi and D. Browne, Phys. Rev. B **38**, 9581 (1988).

[5]H. F. Cheung, E. Riedel, and Y. Gefen, Phys. Rev. Lett. **62**, 587 (1989); O. Entin-Wohlman and Y. Gefen, Europhys. Lett. **8**, 477 (1989).

[6]H. Bouchiat and G. Montambaux, J. Phys. (Paris) **50**, 2695 (1989); G. Montambaux, H. Bouchiat, D. Sigeti, and R. Friesner (to be published).

[7]In the diffusive regime, $\langle j^2\rangle^{1/2}$ is independent of the number of channels.

[8]G. J. Dolan and J. Dunsmuir, Physica (Amsterdam) **152B**, 7 (1988). The electron does not "see" the corners as long as $\lambda_e$ is less than the transverse dimension ($0.06\times0.03\ \mu\text{m}^2$).

[9]At 0.3 Hz the induced "eddy" current $i = \Omega\phi_{ac}/R \approx 10^{-17}$ A is 7 orders of magnitude smaller than the persistent current.

[10]This technique is described in L. P. Lévy and A. T. Ogielski, Phys. Rev. Lett. **57**, 3288 (1986); see also R. Landauer, IBM J. Res. Dev. **32**, 306 (1988).

[11]When a dc field is trapped in the superconducting shield, the background is likewise temperature independent.

[12]Another exponential fit (Ref. 5), $\exp[-(kT/E_c)^{1/2}]$, also fits the data.

[13]Y. Meir, Y. Gefen, and O. Entin-Wohlman, Phys. Rev. Lett. **63**, 798 (1989).

[14]G. Bergmann, Phys. Rep. **107**, 1 (1984).

[15]V. Ambegaokar and U. Eckern (to be published).