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Measurements have been made of the low-temperature magnetic response of single, isolated, micron-size Au loops. The magnetic response is found to contain a component which oscillates with the applied magnetic flux with a fundamental period of $\Phi_0 = h/e$. The amplitude of the oscillatory component corresponds to a persistent current of $\simeq (0.3\text{--}2.0)ev_F/L$, 1 to 2 orders of magnitude larger than predicted by current theories.

In the presence of a static magnetic field, a single isolated normal-metal loop is predicted to carry an equilibrium current [1] which is periodic in the magnetic flux Φ threading the loop. This current arises due to the boundary conditions [2] imposed by the doubly connected nature of the loop. As a consequence of these boundary conditions, the free energy F and the thermodynamic current $I(\Phi) = \partial F/\partial \Phi$ are periodic in Φ, with a fundamental period $\Phi_0 = h/e$. For a metallic loop without impurities at $T = 0$, the magnitude of this current is expected [2] to be $\simeq ev_F/L$, where L is the perimeter of the loop and v_F is the Fermi velocity. In the presence of electrostatic impurities, this current is reduced [3,4] to $\simeq (ev_F/L)l/L = e/\tau_D$, where $\tau_D = L^2/D$ is the time required for an electron to diffuse around a loop of perimeter L ($D = v_F l$ is the diffusion constant [5] and l the elastic mean free path). At finite temperatures [4,6], this current is further attenuated if the electron phase-coherence length l_ϕ or the thermal diffusion length $l_T = (\hbar D/k_B T)^{1/2}$ becomes comparable to or smaller than L. Both phase-breaking and thermal processes are expected to exponentially reduce the current [4].

Recently, Levy *et al.* [7] measured the magnetization of an ensemble of 10^7 Cu loops. They observed an oscillatory response which was consistent with a $T = 0$ persistent current of $\simeq 3 \times 10^{-3} ev_F/L$ per loop. However, the fundamental period they observed was not h/e, but the first harmonic $h/2e$. This surprising result is believed to be due to the large number of loops in the sample [8–12]. The h/e contribution has a random sign for each loop of the sample [3], so that the total h/e contribution should average to zero. The $h/2e$ contribution, however, is expected to survive this ensemble averaging. In a single isolated loop, presumably both periods would be observed.

In this Letter, we present measurements of the magnetization of single, isolated Au loops. We observe an oscillatory component in the magnetic response which oscillates with a fundamental period of h/e. The amplitude of this oscillatory component corresponds to a persistent current at $T = 0$ of $(0.3\text{--}2.0)ev_F/L$, more than an order of magnitude larger than the predicted value of e/τ_D.

The Au loops in this experiment were fabricated on ox-idized Si substrates using a standard bilayer electron-beam lithography process, followed by thermal evaporation of Au and liftoff. As thermal and phase-breaking processes are expected to rapidly attenuate the persistent currents [4], much attention was paid to obtaining clean Au films. By controlling the evaporation rate, pressure, and linewidths of the loops, we were able to obtain values for the electron phase-coherence length l_ϕ in excess of 12 μm at 40 mK, as inferred from weak localization measurements on long (104 μm) narrow wires. The values of l_ϕ so inferred were essentially temperature independent below $\simeq 300$ mK. The $R_\square$ of the films was $\simeq 0.2$ Ω, from which we infer $l \simeq 70$ nm and $l_T = 0.87/T^{1/2}$ (μm/K$^{1/2}$). Measurements on three different loops are reported here. Two of the loops were rings of diameter 2.4 and 4.0 μm, respectively. The third loop was a rectangle of dimensions 1.4 μm $\times$ 2.6 μm. The linewidths of all loops was $\simeq 90$ nm and the thickness of the Au films was $\simeq 60$ nm. Although we were not able to measure l_ϕ directly in our isolated loops, the identical fabrication process of the 1D wires and loops would imply that the electrons in our loops are able to go at least once around the loop without losing phase memory. At $T = 10$ mK, $l_T \simeq 8.7$ μm, so that any attenuation of the persistent current due to thermal processes at our lowest temperatures is expected to be small, at least in the 2.4-μm ring and the 1.4-μm $\times$ 2.6-μm loop.

The magnetization measurements were made using a thin-film miniature dc-SQUID magnetometer [13]. This device consists of two separate chips: the sample chip, on which the pickup coils and field coils are lithographed, and the SQUID chip itself. Figure 1(a) shows a schematic of the sample chip. To maximize the coupling of the sample to the detection coils, the Au loop is written directly by e-beam lithography into one of a pair of counterwound Nb pickup coils. The counterwinding ensures that sensitivity to any background signal from the substrate, or to stray magnetic fields, is minimal. Figure 1(b) shows a micrograph of the 2.4-μm loop in one corner of the 9-μm-inner-diam Nb pickup loop. Around both pickup coils is a single-turn Nb field coil. This coil is center tapped to enable nulling of any signal due to mismatch of the two pickup coils arising from the fabri-
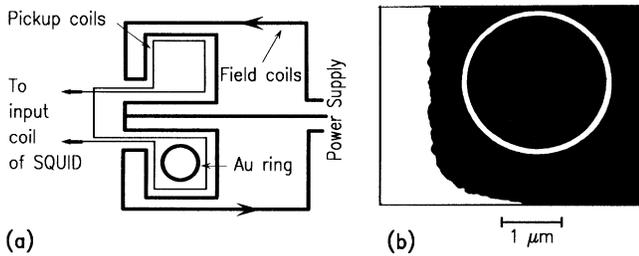
(a)   (b)   1 μm

FIG. 1. (a) Schematic diagram of the pickup coil chip, illustrating the counterwound Nb pickup coils and the on-chip magnetic-field coils. (b) Micrograph of the 2.4-μm-diam Au ring in one corner of the 9-μm-inner-diam Nb pickup coil. The loop linewidth is 90 nm and the thickness is 60 nm.

cation process. The pickup coils are connected to the SQUID chip by Al wires; these wires are kept as short as possible to maximize the flux transfer to the SQUID, and the SQUID chip itself is enclosed in a separate Pb enclosure for shielding. Both chips are mounted on a ceramic holder, enclosed in a cylindrical Nb shield, connected to the room-temperature electronics, and cooled inside the mixing chamber of a dilution refrigerator. The SQUID is current biased and operated in a standard flux-locked loop mode. After signal averaging, the measurement sensitivity (referred to the SQUID) at frequencies above a few Hz is $\simeq 6 \times 10^{-8} \Phi_s$, where $\Phi_s = 2.07 \times 10^{-7}$ G cm^2 is the superconducting flux quantum.

The magnetic response of the loops as a function of field is measured by sweeping the dc current continuously through the on-chip field coils. The maximum magnetic field is determined by the critical current of the field coils and the placement of the loop in relation to them. For this experiment, this field was in the range of 24–35 G. (The magnetic field due to the current in the field coils is calculated from their geometry.) Superposed on top of the dc field is an ac field of frequency $f \simeq 2$–12 Hz [14] whose amplitude is set to maximize the periodic component of the signal [7,15]. This enables us to measure both fundamental (f) and first harmonic ($2f$) signals from the magnetometer using a lock-in amplifier. To calibrate the signal from the Au loop, we calculate the mutual inductance $\mathcal{M}$ between the Au loop and the pickup coils. This is $\simeq 1$ pH for our samples. We then experimentally determine the flux transfer ratio $\mathcal{F}$, which is defined as the ratio of the flux coupled to the SQUID to the flux coupled to the pickup coils. $\mathcal{F}$ is a function of the inductances in the flux transformer circuit and is $\simeq \frac{1}{19}$ for our apparatus. The current in the Au loop is obtained by dividing the SQUID output (in units of Φ_s) by $\mathcal{M}\mathcal{F}$. We estimate the errors in this calibration to be $\simeq 25\%$, arising from the calculation of the magnetic-field profile, $\mathcal{M}$, and $\mathcal{F}$.

Figure 2(a) shows the fundamental response of the magnetometer (in units of Φ_s coupled to the SQUID) as a function of magnetic field for the 1.4-μm×2.6-μm Au
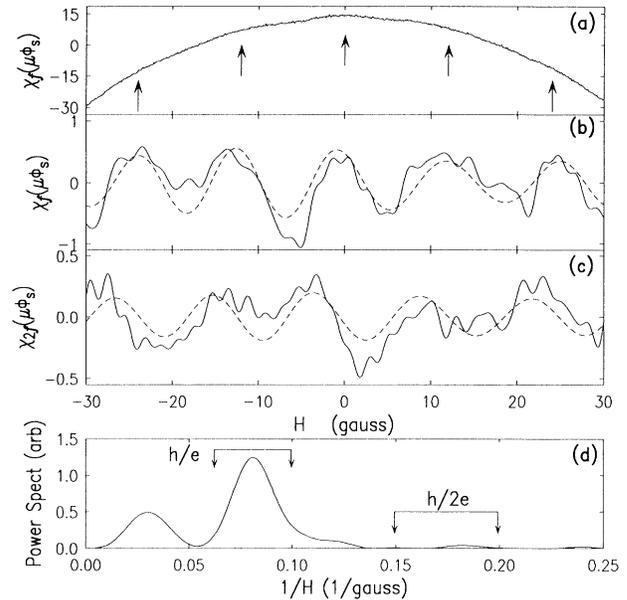
FIG. 2. The magnetic-field dependence of the amplitude of the f and $2f$ signals at 7.6 mK for the 1.4-μm×2.6-μm loop. (a) f response with no signal processing. The arrows point to the maxima of the h/e periodic signal. (b) Data of (a), with the quadratic background subtracted. (c) $2f$ response, after subtraction of a linear background signal. The amplitude of the 4-Hz ac drive field was 4.12 G. (d) Power spectrum for the data displayed in (b). The h/e arrows show the region, centered about the expected frequency for h/e oscillations, over which we bandpassed the data in (b) and (c) to produce the dashed curves. The region where an $h/2e$ signal is expected to appear based upon the inside and outside area of the sample is also shown. The data in (b) and (c) have been digitally filtered to eliminate high-frequency contributions above 0.50 G^{-1}.

loop at 7.6 mK. To first order, this signal is quadratic in field. This quadratic background is present even in the absence of the Au loop. Consequently, only a second-order polynomial has been fitted and subtracted from the raw data in Fig. 2(a) to obtain Fig. 2(b). With this subtraction, clear periodic oscillations as a function of magnetic field are observed, with a fundamental period corresponding to a flux Φ_0 through the loop. The arrows in Fig. 2(a) point to the maxima of the h/e periodic signal in the raw data from the magnetometer. This h/e periodicity is also reflected in the $2f$ response, which is shown in Fig. 2(c) with only a linear contribution subtracted to eliminate the background magnetometer response. Figure 2(d) shows the power spectrum of the data shown in Fig. 2(b). The h/e size bar in Fig. 2(d) shows the region over which we digitally filter the data to produce the dashed curves in Figs. 2(b) and 2(c). The size bar is wider than expected based on the geometry of the loop, due to linewidth broadening resulting from the limited field scale of the raw data. The peak at $\simeq 0.03$ G^{-1} is associated with remanent long field-range fluctua-

tions in the background, and is not a subharmonic of the h/e signal. The ac drive amplitude used (4.12 G zero to peak) for the data displayed in Fig. 2 does not maximize the $h/2e$ contribution. We have also performed measurements at lower drive amplitudes appropriate for maximizing the $h/2e$ contribution. We always observe an h/e contribution, but find the maximum magnitude of the $h/2e$ signal is a factor of 2–3 smaller than the h/e signal at our lowest temperature, as would be expected for a path length of $2L$.

The h/e periodic signal shown in Fig. 2 corresponds to a magnetic moment which is paramagnetic at $H=0$. In general, it is expected that the direction of the persistent current is random, dependent on the total number of electrons N in the loop and the specific realization of the random potential [3,4]. For both the 2.4-μm-diam ring and the 1.4-μm×2.6-μm loop, the two loops for which we made the determination, the sign of the current, which does not change after thermal cycling, corresponds to a moment which is paramagnetic. Quite obviously, one cannot tell from a sampling of only two rings whether this is purely a coincidence, or whether all single rings will yield a paramagnetic response.

In measurements on over ten different sample chips, some with and some without Au loops, we have always found a reproducible, aperiodic background signal in the f and $2f$ responses. The background varies from sample to sample, and can be different for the same sample chip on subsequent cooldowns. Ideally, as we have seen for the rectangular loop of Fig. 2, this background is smooth, and can be eliminated by subtracting a linear or quadratic contribution. More often, as we found for the 2.4- and 4.0-μm loops, the background signal has fluctuations on a field scale comparable to the expected signal. The field dependence of the fluctuations is sample specific, and we believe that they are associated with the dynamics of flux motion in the Nb pickup coils. In the presence of these fluctuations, it becomes a problem to distinguish the signal from the background. In order to determine the error introduced by these fluctuations, we remeasured the 2.4-μm ring sample chip after the ring had been removed. In this experiment, above 12 mK, the empty magnetometer gave a signal a factor of 2 smaller than when the sample was present. At lower temperatures, however, a very rapidly growing background signal was measured, so that, at 5 mK, the signal in the "h/e" bandpass was actually larger than the corresponding signal measured with the Au loop. Nevertheless, we have confidence that the signal we are measuring comes from the Au loops and not from the magnetometer, because, for the 2.4- and 4.0-μm loops, there was always a peak in the power spectrum at a frequency expected for the h/e oscillations. The position of this peak remained unchanged after warming to room temperature and recooling, although the fluctuations in the background signal may have shifted. For the 1.4-μm×2.6-μm Au loop, there were no background fluctua-

tions, and the response of the empty coil, after appropriate background subtraction, was essentially at the level of our noise. We stress, however, that the presence of a fluctuating background for the 2.4- and 4.0-μm loops may introduce systematic errors in estimating the amplitude of the periodic component of the magnetic response.

At finite temperature, the amplitude of the h/e periodic persistent current is expected to be exponentially attenuated [4] with l_ϕ and l_T. For our Au loops, l_ϕ is temperature independent below $\simeq300$ mK, so that the only temperature dependence is through l_T. Thus, we expect the amplitude of the persistent current to go as $I_{h/e} = C(e/\tau_D)e^{-L/l_T}$, where C is a constant of order unity [16]. Then, if we plot $I_{h/e}$ as a function of $T^{1/2}$ on a semilogarithmic scale, we should obtain a straight line whose slope depends only on the dimensions of the sample and the material properties of the Au film. Figure 3(a) shows the zero-to-peak amplitude of the persistent current determined from the f response as a function of $T^{1/2}$ for the 2.4-μm ring and the 1.4-μm×2.6-μm loop. $I_{h/e}$ is determined by integrating the power in the Fourier transform over the range of frequency for the h/e oscillations shown by the size bar in Fig. 2(d), and applying the Bessel function normalization [7] $[2J_n(2.16)]$ appropriate for the ac drive amplitude of 4.12 G. The f and $2f$ data differ by only $\simeq30\%$ at the lowest temperatures after this normalization. The straight line shown in Fig. 3(a) is the function $\exp(-\alpha T^{1/2})$. The value of $\alpha T^{1/2}$ shown is $14.4T^{1/2}$, close to the value of $L/l_T=9.3T^{1/2}$ we compute for both loops. Plotted in this manner, the data appear to
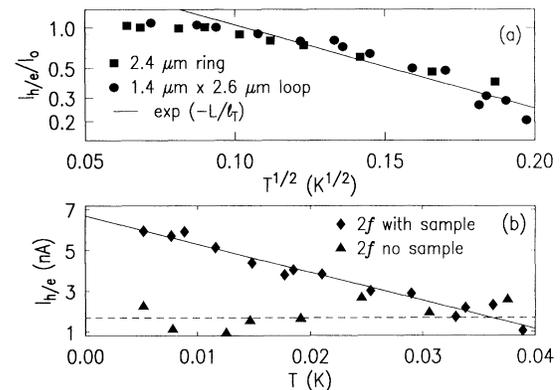

FIG. 3. (a) The amplitude, on a logarithmic scale, of the persistent current in the 2.4-μm ring and the 1.4-μm×2.6-μm loop as a function of $T^{1/2}$, obtained from the fundamental response. I_0 is 36 nA for the 2.4-μm ring and 8 nA for the 1.4-μm×2.6-μm loop. The solid line is the function $\exp(-L/l_T)$, with $L/l_T=14.4T^{1/2}$. (b) The $2f$ response of the 1.4-μm×2.6-μm loop plotted on a linear scale to show that a power law is also consistent with the data. The solid line is a guide to the eye. The solid triangles are the data obtained after removing the Au loop from the magnetometer, using the same measurement parameters.

saturate below 10 mK, the temperature which corresponds to the correlation energy $E_c = \hbar/\tau_D$. Clearly, however, the data are over a very limited temperature range, so that we cannot exclude other functional forms. Indeed, it appears that a linear temperature dependence describes our data equally well, as demonstrated by the $2f$ data for the 1.4-μm×2.6-μm loop shown in Fig. 3(b). Also shown in Fig. 3(b) is the response of the magnetometer with the 1.4-μm×2.6-μm-diam loop removed. This signal is essentially at the level of our noise.

Given the uncertainty in the functional form of the temperature dependence, we have chosen to compare the theoretical predictions to the measured value of $I_{h/e}$ at our lowest temperatures (4.5 mK), rather than attempting to extrapolate the data to $T=0$. With $C=1$ and $l=0.07$ μm at $T=0$, the expected magnitudes of $I_{h/e}$ for the 4-μm, 2.4-μm, and 1.4-μm×2.6-μm loops are 0.09, 0.27, and 0.25 nA, respectively, while the measured values from the $2f$ response are 3 ± 2, 30 ± 15, and 6 ± 2 nA. Clearly, our measured signals are a factor of 30 to 150 times larger than the theoretical estimates. We have no definite explanation for this discrepancy, although a few possibilities suggest themselves. First, note that the value of $I_{h/e}$ measured is on the order of ev_F/L, the amplitude expected if there were no diffusive correction factor of l/L. This factor reduces the theoretical estimate by 125–200 for our samples. Second, there may be an additional correction factor for the finite number of transverse channels [3,4]. For our samples, this factor could be between 12 and 140. Finally, recent theoretical work suggests that the prefactor C in the equation for $I_{h/e}$ may be larger than unity [17]. We point out that the most recent theoretical estimates of the ensemble-averaged $h/2e$ signal are also smaller by about a factor of 10 than the value reported by Levy *et al.* [7].

In conclusion, we have observed periodic h/e oscillations in the magnetic response of three different single Au loops. The amplitude of our signals is much larger than current theoretical estimates, emphasizing the need for more experimental and theoretical work before the origin of this effect is fully understood.

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