Diamagnetic orbital response of mesoscopic silver rings

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We report measurements of the flux-dependent orbital magnetic susceptibility of an ensemble of 105 disconnected silver rings at 217 MHz. Because of the strong spin-orbit scattering rate in silver this experiment is a test of existing theories on ensemble averaged persistent currents. Below 100 mK the rings exhibit a magnetic signal with a flux periodicity of $\hbar/e$ consistent with averaged persistent currents, whose amplitude is estimated to be of the order of 0.3 nA. The sign of the oscillations indicates unambiguously diamagnetism in the vicinity of zero magnetic field. This sign is a priori not consistent with theoretical predictions for average persistent currents. We discuss several possible explanations of this result.

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At low temperature, electrons in metallic mesoscopic samples keep their phase coherence on a length $L_\Phi$ which is larger than the sample size. The transport and thermodynamic properties of the system are then sensitive to interference between electronic wave functions and present spectacular signatures of this phase coherence. To measure these effects the ring geometry is particularly suitable. Indeed in the presence of a magnetic flux $\Phi$ through the ring the periodic boundary conditions for electronic wave functions acquire a phase factor $2\pi\Phi/\Phi_0$ with $\Phi_0 = \hbar/e$ the flux quantum [1]. As a consequence the free energy $F$ of the system is flux dependent which leads to the existence of a non-dissipative current $I = -\partial F/\partial \Phi$, the persistent current, which is a periodic function of $\Phi$ [2] and can be detected by measuring the induced magnetic moment.

Persistent currents in mesoscopic rings have been studied for over 10 years theoretically [3, 4, 5, 6, 7, 8, 9] and experimentally addressing the question of either the typical current (measured on a single or a small number of rings) [10, 11, 12, 13], or of the ensemble averaged persistent current (measured on arrays of rings) [14, 15, 16, 17]. However theory and experiment still do not agree. One important subject of disagreement is the sign of the average value of these persistent currents. Recent experiments on GaAs rings [16] indicate a sign corresponding to low field diamagnetism. In contrast theoretical calculations which include repulsive field interactions predict paramagnetism [3, 4, 17]. A recent theory [18] shows that additional currents may be generated in rings due to the rectification of a high frequency non equilibrium noise. These currents, as also suggested in reference [18], are not experimentally distinguishable from persistent currents. This mechanism yields a diamagnetic sign and gives the right amplitude for the average current measured in GaAs rings. An important feature is that the sign of this current should change from diamagnetic to paramagnetic depending on the strength of spin-orbit scattering. In contrast, the thermodynamic equilibrium persistent currents are insensitive to spin-orbit scattering. This motivates the comparison between the orbital magnetism of mesoscopic rings fabricated in different materials having various strength of spin-orbit scattering. The experiments on GaAs rings dealt with the case of weak spin-orbit interactions. Previous experiments on Au rings [19] addressed the case of strong spin-orbit but were however not conclusive concerning this sign because the current was not averaged on a sufficient number of rings. The present experiment was designed to resolve this question: using Ag (a material with strong spin-orbit interactions) and a large number of rings.

We have investigated the magnetic response at low temperature of a collection of silver rings, with spin-orbit scattering length much shorter than their circumference in contrast with the GaAs case. The magnetic response of the rings is detected by a resonant method: the rings are coupled to the inductive part of a superconducting microresonator made of niobium deposited on sapphire. This resonator has a resonance frequency of 217 MHz with a quality factor of 2105 below 1 K. A schematic picture of the resonator, composed of an inductance (meander line) and a capacitance (comb-like structure), with the rings on the inductive part is shown on Fig. 1(c). The detailed fabrication and characterization of this type of resonator has been described elsewhere [16].

By measuring the shift of the resonance frequency $f$ of the resonator induced by the presence of the rings, we have access to $\chi$ the averaged magnetic susceptibility of the silver rings, according to:

$$\frac{\delta f}{f} = -\frac{1}{2}Nk_m\chi \quad (1)$$

where $N$ is the number of rings coupled to the resonator, $k_m$ is a coefficient characterizing the coupling of one ring to the inductive part of the resonator [16]. This quantity is recorded as a function of the amplitude of a DC magnetic field produced by a superconducting coil. The sample is placed in a dilution refrigerator with a base
temperature of 30 mK.

The fabrication involved two steps of lithography. First the superconducting resonator is fabricated by optical lithography and tested at low temperature. PMMA is then spin-coated on top of the resonator. A high resolution e-beam writer is used to pattern rows of rings aligned between the meanders of the inductive part of the resonator. After metal deposition and lift-off we obtain around $1.5 \times 10^5$ silver rings aligned with the inductance of the resonator, on the same substrate. With this configuration the quality factor of the bare resonator is preserved in contrast with previously measured GaAs/AlGaAs rings [16]. Moreover thanks to the alignment of the rings with the inductance the coupling coefficient $k_m$ is the same for all the rings. As a consequence the rings experience the same AC field, another improvement over the experiments on GaAs rings. Images of the resonator and the rings are shown on Fig. 1. During the lift-off process some of the rings are cut or damaged so that one can estimate the number of rings producing a signal to be between $10^5$ and $1.4 \times 10^5$. This very large number of rings insures that the measurement is dominated by the ensemble average of the magnetic susceptibility of the rings (which was not the case on previous experiments on an array of 30 Au rings [12]). The different characteristic lengths of the silver rings are given in Table I. They verify $L_0 > L > L_{SO}$. This experiment is in the strong spin-orbit scattering regime at magnetic field below 100 Gauss. In this low field regime the magnetic length defined as: $L_H^2 = \Phi_0 / H$ is larger than the spin-orbit scattering length. Data were taken within this field range during slow DC magnetic field sweeps ($0.02 \, \text{G/s}$). Moreover restricted ourselves to magnetic field less than 30 G in order to prevent vortex trapping in the superconducting resonator which give rise to inhomogeneities of DC magnetic field on the different rings. In addition all data was acquired while decreasing the absolute value of the magnetic field to minimize hysteresis effects. We also had to operate the resonator in the over-coupled regime with respect to the RF generator and strongly attenuate the RF power injected to less than 0.1 pW. This was done to prevent electron heating by the RF radiation and minimize spurious signal originating from defects in the meander line.

The resonance frequency shift versus magnetic field has two parts: the first one is due to the magnetic field dependence of the magnetic penetration length in the resonator. This part has been checked to be the same with or without the rings. In the small magnetic field range explored here, it is well approximated by a parabola (see inset of Fig. 2 and Ref. [16]). Periodic oscillations are superimposed on top of this signal. To focus on these oscillations the contribution of the bare resonator which give rise to inhomogeneities of DC magnetic field on the different rings. In addition all data was acquired while decreasing the absolute value of the magnetic field to minimize hysteresis effects. We also had to operate the resonator in the over-coupled regime with respect to the RF generator and strongly attenuate the RF power injected to less than 0.1 pW. This was done to prevent electron heating by the RF radiation and minimize spurious signal originating from defects in the meander line.

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TABLE I: Characteristics of the silver rings. The mean free path $l_c$ is deduced from the resistance of silver wires fabricated together with the rings and with the same width. The spin-orbit length $L_{so}$ and the phase coherence length $L_{\phi}$ are extracted from previous weak-localisation measurements on silver wires also fabricated in the same evaporator and using the same bulk silver [20].

| perimeter $L$ | width $w$ | thickness $h$ | $l_c$ | $D = v_F l_c / 3$ | $E_c = h D / L^2$ | $L_{so}$ | $L_{\phi} (T = 40 \text{mK})$ |
|--------------|-----------|--------------|-------|-----------------|------------------|---------|------------------|
| 4 \mu m     | 130 nm    | 70 nm        | 40 ± 5 nm | 0.018 m$^2$.s$^{-1}$ | 54 mK            | 575 nm  | 13 \mu m         |

FIG. 3: Fourier transform of the frequency shift due to the rings at 40 mK and 140 mK. Inset: temperature dependance of the frequency shift due to the rings. The data are consistent with an exponential decay with a temperature scale of 40 mK.

previous curves (Fig. 3). At low temperature the periodicity measured is consistent with half a flux quantum in the area enclosed in a ring, as expected for the average susceptibility [1, 21]. From the value of the maximum frequency shift $\delta f = f(\Phi_0/4) - f(\Phi = 0)$ and given the coupling coefficient $k_m$ [10] we deduce the amplitude of the variation with magnetic field of the magnetic susceptibility $\delta \chi(\omega) = 5.3 \pm 0.9 \times 10^{-24} \text{m}^3$. In order to compare this value to existing theories on orbital magnetism we make the assumption that the signal measured is due to persistent currents:

$$\frac{\partial I(\Phi)}{\partial \Phi} = -\frac{\delta f(\Phi)}{2f} \frac{L}{N \mathcal{M}^2}$$

(2)

with $L = 0.05 \mu \text{H}$ the estimated inductance of the resonator and $\mathcal{M} = 0.14 \text{pH}$ the calculated mutual inductance between one ring and the inductive part of the resonator. It is then possible to deduce the flux dependence of the average persistent current depicted in Fig. 3 which oscillates with a periodicity $\Phi_0/2$ and an amplitude $|I_0| = 0.33 \pm 0.05 \text{nA}$. The current is diamagnetic at low field. Note that the sign of the susceptibility oscillations can be determined in a completely unambiguous way in this experiment, since it is directly related to the sign of the resonance frequency shift measured. Another check comes from the field dependence of the diamagnetic signal of the bare resonator (see inset of Fig. 2). The temperature dependence of the oscillations is shown on inset of Fig. 3. The signal cannot be detected above 100 mK. From the small number of data points, we can only say that this temperature decrease of the signal is consistent with an exponential behavior with a temperature scale of the order of 40 mK, which is close to the value of the Thouless energy $E_c = h D / L^2$ where $D$ is the diffusion coefficient.

We now compare our result with theoretical predictions. Due to the very small value of the mean level spacing $\Delta$ in metal rings, theoretical predictions based on non-interacting electrons [18], which lead to a current of amplitude $\Delta / \Phi_0$, are not able to explain the value of the current measured. In the case of interacting electrons [10], the expected value of the persistent current is of the order of $0.2 E_c / \Phi_0 = 0.04 \text{nA}$, which is within an order of magnitude of the experimental value. However theory predicts in this case a paramagnetic current for repulsive interactions, with or without strong spin-orbit coupling [5, 6]. Considering instead attractive interactions, superconducting fluctuations in silver with a critical temperature of the order of 50 mK, would lead to a diamagnetic current (therefore in agreement with our experiment) with the same amplitude as the repulsive case. Moreover it has been recently suggested [22] that superconducting fluctuations could give rise to a much larger

FIG. 4: Average persistent currents through the rings reconstituted from the field dependence of the resonance frequency in Fig. 2 according to expression (2) after high pass filtering at 0.025 $\text{Gauss}^{-1}$ and integration of the signal.
current than the initial predictions of reference \[1\]. If one now considers the possibility of non equilibrium noise induced currents, according to ref. \[1\], it is clear that this mechanism cannot explain the present experimental results since a paramagnetic average current is predicted in the strong spin-orbit limit.

All the previous analyses are valid in the limit of zero frequency. Our experiment however is performed at the resonance frequency of the resonator, i.e. 217 MHz. This frequency is larger than \(1/\tau_\Phi\) and not negligible compared to \(1/\tau_D\) (\(1/\tau_\Phi = 100\) MHz and \(1/\tau_D = 1\) GHz). In this frequency range it is reasonable to expect extra contributions to the magnetic response of Aharonov-Bohm rings. This point is already contained in the early work of Altshuler, Aronov and Spivak describing the \(\Phi_0/2\) oscillations in the average conductance \(\Delta G(\omega)\) of a ring of perimeter \(L = 2\pi R\) excited by an electromotive force \[23, 24\]:

\[
\Delta G(\omega) = - \frac{e^2}{\pi \hbar L} \sum_{l=-\infty}^{\infty} \frac{1}{\pi} \frac{R L_\Phi(\omega)}{R_l(\omega)} \left( \frac{L_l(\omega)}{2} \right)^2 \tag{3}
\]

with \(L_\Phi(\omega)^2 = L_\Phi^2 / (1 + i \omega \tau_\Phi)\). At finite frequency the imaginary part \(G''(\omega)\) of \(\Delta G(\omega)\) is non zero and one can associate to this imaginary conductance a non dissipative current which response function reads:

\[
\frac{\partial I}{\partial \Phi} = \omega G''(\omega) \tag{4}
\]

The first harmonics is of the order of \(E_c/\Phi_0\) and changes sign with spin-orbit. More precisely for parameters close to the experimental ones \(\omega \tau_\Phi = 2\) and \(L_\Phi/R = 10\), one finds numerically a diamagnetic current of amplitude 0.1 \(E_c/\Phi_0 = 0.02 nA\) of the order of the thermodynamic current. Note that weak-localisation experiments on silver wires at finite frequency are in agreement with this analysis \[23\]. However it was pointed out by Efetov et al. that this result is in principle only valid for connected geometries \[23\]. Finally we would like to point out that the contribution of electron-electron interactions, which plays an essential role in the thermodynamic current, has not been examined at finite frequency.

To conclude we have demonstrated in this experiment the existence of \(\Phi_0/2\) periodic orbital magnetism in silver rings at 217 MHz and low temperature. The signal is consistent with diamagnetic averaged persistent current \[\Phi_0\] which does not agree with theoretical predictions in the presence of repulsive interactions. This sign can be explained either by the existence of very weak superconducting fluctuations or by the value of the measurement frequency which is larger than the inverse phase coherence time.

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