Magnetoresistance oscillations and the half-flux-quantum state in spin-triplet superconductor Sr$_2$RuO$_4$

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We report results of our low-temperature magnetoelectric transport measurements on micron-sized short cylinders of odd-parity, spin-triplet superconductor Sr$_2$RuO$_4$ with the cylinder axis along the $c$ axis. The in-plane magnetic field and measurement current dependent magnetoresistance oscillations were found to feature an amplitude much larger than that expected from the conventional Little-Parks effect, suggesting that the magnetoresistance oscillations originate from vortex crossing. The free-energy barrier that controls the vortex crossing was modulated by the magnetic flux enclosed in the cylinder, the in-plane field, measurement current, and structural factors. Distinct features on magnetoresistance peaks were found, which we argue to be related to the emergence of half-flux quantum states, but only in samples for which the vortex crossing is confined at specific parts of the sample.

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Fluxoid quantization in a doubly connected conventional superconductor in the unit of a full flux-quantum, \( \Phi_0 = \frac{hc}{2e} \) (where $h$ is the Plank constant and $e$ the electron charge), is a direct consequence of pairing of the electrons and the emergence of long-range phase coherence among the paired electrons. [1] Deaver and Fairbank [2] and Doll and Nábaue [3] measured the magnetic flux trapped in a superconducting hollow cylinder and the torque on the circulating supercurrent, respectively, to demonstrate this remarkable effect. The physics of this effect was further clarified by the Little and Parks (LP) experiment [4], demonstrating the oscillations in the superconducting transition temperature, $T_c$, and the periodic variation of the free energy with the applied magnetic flux in a doubly connected superconducting cylinder. Therefore, after the initial experimental evidence for the half-flux-quantum vortices [5] was found in the odd-parity, spin-triplet superconductor Sr$_2$RuO$_4$, [6-10] in the torque magnetometry experiment [11], the LP measurement has been highly desirable so as to obtain insights into the physics of the half-flux-quantum state in this unconventional superconductor.

The half-flux-quantum state is allowed in a spin-triplet superconductor along with the conventional full-flux-quantum state because of the presence of spin and orbital parts of the order parameter. A possible scenario is that a phase winding of $\pi$ is formed separately in each part of the order parameter around a doubly connected sample or a vortex core to maintain the singlevaluedness of the order parameter. Only the circulating supercurrent due to the orbital phase winding features a magnetic flux, giving rise to a vortex state featuring a half-flux-quantum of $\Phi_0/2$. The free energy of the half-flux-quantum state is usually higher than the conventional full-flux quantum one, making its physical realization difficult. For a doubly connected, micron-sized crystal of Sr$_2$RuO$_4$, the free energy of the half-flux-quantum state appeared to be lowered near applied half-flux quanta by the application of an in-plane magnetic field [11] as proposed theoretically [12].

Recently we carried out magnetoresistance oscillation measurements on micron-sized, single crystal rings of Sr$_2$RuO$_4$, [13], in which a large number of pronounced resistance oscillations with an amplitude much larger than that expected from the LP effect were observed down to very low temperatures. The observed magnetoresistance oscillations were attributed to $c$-axis vortices moving across the sample that leads to a finite transverse voltage according to the Josephson relation [4]. The oscillation amplitude is controlled by the barrier potential for vortex crossing, which is a function of applied flux due to fluxoid quantization. No features associated with half-flux-quantum states were found over a wide range of the out-of-plane field without the application of an in-plane field. Interestingly, a fit of our data to the Ambegaokar-Halperin (AH) model of thermally activated vortex crossing over a free energy barrier [14], [15] yielded values of zero-temperature penetration depth much larger than that of the bulk [16], which is consistent with the small magnetic moments observed in the torque magnetometry experiment compared to the numerical calculation [17]. Here we present magnetoresistance oscillation measurements in the presence of an in-plane field with varying measurement currents and magnetoresistance signatures of the half-flux-quantum state, thereby providing insights into conditions favoring experimental observation of this
exotic state.

To prepare our samples, thin crystal plates of Sr$_2$RuO$_4$ were made on a Si/SiO$_2$ substrate by mechanical exfoliation from a bulk single crystal. Four- or six-point electrical leads, made of 200 nm Au and an underlay of 10 nm Ti, were prepared on the crystal plates by photolithography. A doubly connected cylinder with four leads was cut from the crystal plates using a focused ion beam (FIB), with Sr$_2$RuO$_4$ leads extending from the cylinder to the Ti/Au contacts. Fig. 1a shows a scanning electron microscopy (SEM) image of a typical cylinder, which has a height of about 0.74 µm and a wall thickness increasing from top to bottom due to the profile of the ion beam. This hollow cylinder has a mean wall-thickness ($w$) of 0.26 µm and a mean radius ($r$) of 0.58 µm as indicated in Fig. 1b. More details in sample fabrication and the estimation of sample dimensions can be found in reference [13]. Our samples were measured in a dilution refrigerator with a base temperature of 20 mK, using a dc technique. Since both the out-of- and the in-plane fields, $H_{||c}$ and $H_{||ab}$, are needed, a homemade superconducting Helmholtz coil was incorporated inside a large superconducting solenoid.

Almost all Sr$_2$RuO$_4$ samples prepared this way were found to be superconducting with an onset $T_c$ sample dependent. In Fig. 1c, the temperature dependent resistance data revealed a broad transition with a zero-resistance $T_c$ dependent on the measurement current. A resistance peak was found around 1.6 K, an anomaly observed previously in superconducting nanowires and attributed to charge imbalance encountered when superconducting voltage leads were used. [18] The onset $T_c$ of these samples is higher than the bulk phase, 1.5 K, likely due to the presence of dislocations in the thin crystal plates of Sr$_2$RuO$_4$. [19]

We measured the sample resistance as a function of the in-plane field, $H_{||ab}$, aligned perpendicular to the current leads (Fig. 1b). Hysteresis as well as fluctuations were observed (Fig. 1d), especially for the curves measured at a low temperature, 0.6 K, possibly due to the depinning of parallel vortices. Even though the in-plane field could be misaligned from the $ab$-plane of the cylinders by a small angle, the vortices in the sample should still be aligned in the in-plane direction because of the strong anisotropy of Sr$_2$RuO$_4$. The critical field, at which the sample resistance becomes non-zero, was found to decrease with the increasing measurement current. It should be noted that the $c$-axis vortices can appear simultaneously in Sr$_2$RuO$_4$ crystals. [20] [21]

Pronounced magnetoresistance oscillations were observed at fixed temperatures as the out-of-plane field, $H_{||c}$, was ramped up (Fig. 2). The oscillation period is $\Phi_0$ based on the sample dimensions. A large measurement current was used in order for the magnetoresistance oscillations to be measured at such low temperatures. The vortex crossing origin of the magnetoresistance oscillations is confirmed by a quantitative comparison between the experimental values of the oscillation amplitude and that expected from the conventional LP effect. No obvious feature corresponding to the transition between half- and full-flux quantum states in the resistance oscillations was seen in the presence of a constant in-plane magnetic field, $H_{||ab}$, up to 800 Oe. At such a high in-plane field, the magnetoresistance oscillations showed significant irregularities and instability (Fig. 2d).

To understand the data presented above, the magnetoresistance oscillation signature of the half-flux quantum states in our samples needs to be analyzed. For simplicity, our sample is modeled as a thin-wall hollow cylinder without leads (Fig. 3a). Starting with a two-component order parameter, the Gibbs free energy per unit length of a cylinder with a wall thickness of $w$ and a radius of $r$ in the thin-wall limit ($w \ll r, \lambda$) as a function

![Diagram](https://via.placeholder.com/150)
of an external field, $H_{||c}$, has the following form. $\Delta G$, the kinetic part of the Gibbs free energy, $\Delta G$, as a function of $\Phi$ calculated from equation (1) for a doubly connected cylinder with $r = 0.58 \text{ \mu m}$ and $w = 0.26 \text{ \mu m}$, as shown in (a), at a temperature close to $T_c$ (b) and 0.6 K (c); Near $T_c$, we assume $\rho_{sp}/\rho_s \rightarrow 1$ and $\lambda \rightarrow \infty$; Values of $\rho_{sp}/\rho_s = 0.25$ and $\lambda \sim 100 \text{ nm}$ are used for $T=0.6 \text{ K}$. Values of $\Delta G$ for the full- and half-flux-quantum states are plotted in blue and red, respectively. The light red shadings present the stability regions of the half-flux-quantum state; (d) When the width of the stability region is $\Phi_0/4$, the energy difference between the full- and the half-flux-quantum states at $\Phi = \Phi_0/2$ is 0.31, and the depth of the free energy dip is only 0.04. The scale for $\Delta G$ is $0.4(\Phi_0/8\pi^2 r^2)\beta/(1 + \beta)$.

The value of $\rho_{sp}/\rho_s$ is not known for Sr$_2$RuO$_4$. Numerical studies on the stability regions of half-flux-quantum states based on the torque magnetometry data suggested a value of $\rho_{sp}/\rho_s = 0.25$ at 0.6 K. The ground state free energy shown in Fig. 3 is calculated using parameters of bulk Sr$_2$RuO$_4$. Using the AH model to fit our data we obtain a large penetration depth, and therefore a $\beta$ value much smaller than expected from that using parameters of the bulk, which favors the presence of the half-flux-quantum state; however, a large value of the penetration depth also suggests a suppression of superconductivity which indicates that $\rho_{sp}/\rho_s$ is likely to be much larger than that used in the numerical study, close to 1. The free energy contribution from in-plane field is given as $\Delta F = -g\mu_B(\rho_\uparrow - \rho_\downarrow)|B_{||ab}|$. Assuming
that $\rho_{t\perp}$ is independent of $\Phi$, the free energy lowering of the half-flux-quantum state is proportional to the magnitude of the in-plane field. Fig. 3d is obtained by directly shifting the free-energy curve of half-flux-quantum state vertically to match the width of the stability region observed in the torque magnetometry measurements[11]. The free energy variation in the half-flux-quantum state is tiny compared to the free energy difference between the full- and half-flux-quantum states. The sample resistance tracks the free-energy monotonically even though the precise form that is associated with vortex crossing is not known.

With the emergence of the half-flux-quantum state we expect the resistance peaks of $\Phi_0$ oscillations to develop a dip around applied half-flux quanta, especially as the stability regime becomes substantial. For a sample used in our experiment, vortex trapping and pinning within the sample, the free energy contribution from the electrical leads, and the spatial variation of the superconducting order parameter will all complicate the free-energy barrier for the vortex crossing, making sample resistance as a function of the magnetic flux difficult to track. In the torque magnetometry experiment, the entry of parallel vortices was found to be accompanied by the field shift of the transitions between the adjacent fluxoid states and a reduction in the stability region of the half-flux-quantum states. In our experiment, the resistance peaks seen in high in-plane fields (Fig. 2d) appear to be flatter than those seen in the smaller in-plane fields (Fig. 2a-c), consistent with theoretical expectations. However, no sharp enough feature tied uniquely to the existence of the half-flux-quantum state was found. Irregularities in magnetoresistance due to random vortex motion make the observation more difficult.

The stability condition described above suggests that thinning the cylinder wall, which is limited by FIB damage that could make the sample nonsuperconducting, favors the emergence of the half-flux-quantum state. We observed unexpected features in Sample 3 with a mean radius $r \approx 545$ nm, mean wall thickness $w \approx 244$ nm and out-of-plane thickness $t \approx 482$ nm (Fig. 4a). The resistance oscillations are smooth and show little hysteresis even under a substantial in-plane field. The sample resistance was found to rise gradually only at one side of the resistance peaks, with a nearly vertical resistance rise on the other side when the measurement current was relatively small (Fig. 4b–4d). A gradual resistance rise started to appear on the other side when the measurement current was further increased to a critical value, suggesting the critical currents of the two arms of the cylinder are different. The critical value was found to be around 60 $\mu$A for $H_{||ab}=400$ Oe. We estimate that the difference in the critical currents of two arms of the cylinder is on the order of 10 $\mu$A. The measured oscillation period is about 23.7 Oe, which corresponds to an effective radius of 527 nm based on $\Delta H = \Phi_0/(\pi r^2)$, smaller than the value obtained from the SEM image. Dip features in the magnetoresistance oscillations were observed at rela-

![FIG. 4: (a) A SEM image for a Sr$_2$RuO$_4$ cylinder, Sample 3. Magnetoresistance oscillation measurement, $R$ vs. $H_{||ab}$, with an in-plane field of (b) 0 Oe at $T=0.6$ K, and (c) 400 Oe and (d) 600 Oe at $T=0.3$ K for the sample in (a). The measurement currents are indicated starting from the bottom to the top curves. The dash and solid curves in (b) indicate results for positive and negative measurement currents, respectively. (d) A SEM image for a cylinder sample with two constrictions. $R$ vs. $H_{||ab}$ measured at $T=0.3$ K for the sample in (e) with an in-plane field of (f) 0 Oe, (g) 400 Oe and (h) 1000 Oe. The top curve shifted vertically by 15 m$\Omega$ for clarity in (h).]
restrictions were shown. The sample has a mean radius of the useful magnetoresistance signals.\[24\] The uneven critical currents and the smooth magnetoresistance oscillations found in Sample 3 suggest that this sample may possess a weak link in one arm of the hollow cylinder due to a sample specific reason. Theoretical studies suggest that adding physical constrictions in the superconducting hollow cylinder, the kinetic energy of the spin current relative to that of the charge supercurrent is determined by the narrowest part of the superconducting loop, $\beta = r w_{\text{min}}/2\lambda^2$.\[24\] An additional advantage of the sample with the weak link is that the vortex crossing can be directed to weak link and thereby increasing the amplitude of the useful magnetoresistance signals.\[24\]

In Fig. 4e-4h, results from a sample featuring two constrictions were shown. The sample has a mean radius $r \approx 510$ nm, a wall thickness of 340 nm, and the width of the constriction roughly 250 nm. Since the width of the constrictions in this sample is comparable to the wall thickness of Samples 1 and 2, based on the free-energy consideration, the half-flux-quantum state should appear at a similar in-plane field. Consistently, no feature was seen in the magnetoresistance oscillations at an in-plane field of 400 Oe (Fig. 4g). After an in-plane field of 1000 Oe was applied was a dip found in resistance peaks (Fig. 4h), with features similar to that seen in Sample 3, consistent with expectations from the above analysis.

To conclude, results from the magnetoresistance oscillation measurements suggest that, even though the half-flux-quantum state may indeed exist in micron-sized, doubly connected cylinders of single crystal Sr$_2$RuO$_4$, the free energy barely vary as a function of applied flux enclosed in the cylinder within the stability regime. In the torque magnetometry measurements, a plateau in the magnetization can be detected as long as the free energy of the half-flux-quantum state is lower than that of the full-flux-quantum state. The difference of the free energy between them is difficult to determine through the analysis of the measured magnetoresistance oscillations because the connection between the rate of the vortex crossing and the free energy barrier is complicated. Our measurements show that the kinetic energy part of the free energy, which is tuned by the applied magnetic flux, is only a small part of the total free energy. The spin-orbital coupling to the free energy, which should be large in Sr$_2$RuO$_4$,\[5\] may have made the kinetic part of the free energy even smaller, and the detection of half-flux-quantum through magnetoresistance oscillation measurements significantly more difficult than the torque magnetometry measurements.

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