



## Parity violation in a single domain of spin-triplet Sr<sub>2</sub>RuO<sub>4</sub> superconductors

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### ABSTRACT

We observed an unconventional parity-violating vortex in single domain Sr<sub>2</sub>RuO<sub>4</sub> single crystals using a transport measurement. The current–voltage characteristics of submicron Sr<sub>2</sub>RuO<sub>4</sub> show that the induced voltage has anomalous components which are *even* functions of the bias current. The results may suggest that the vortex itself has a helical internal structure characterized by a Hopf invariant (a topological invariant). We also discuss that the hydrodynamics of such a helical vortex causes the parity violation to retain the topological invariant.

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A quantized vortex is a topological matter in superconductors and superfluids. In metallic superconductors, the Abrikosov vortex is characterized only by an integer winding number of phase. In unconventional superconductors, the superfluid of <sup>3</sup>He and spinor cold atoms, internal degrees of freedom of the order parameter enrich the variety of vortices [1–3]. Although a number of theoretical studies have predicted the existence of such unconventional vortices, experimental confirmation of them is still limited to several studies such as NMR spectroscopy of <sup>3</sup>He and imaging of spinor cold atoms [4,5]. Here a Cooper pair in spin-triplet superconductors has electric charge 2e. Thus one can resolve the dynamics of unconventional vortices through electric transport. We will address this issue in the present Letter.

Sr<sub>2</sub>RuO<sub>4</sub> [6] is a promising spin-triplet chiral *p*-wave superconductor candidate (i.e., spin *S* = 1 and orbital angular momentum *L* = 1). Since two states with different chirality degenerate in the ground state, bulk Sr<sub>2</sub>RuO<sub>4</sub> is considered to have chiral domain structures. The transport properties have been studied in relation to Josephson interferometry using bulk Sr<sub>2</sub>RuO<sub>4</sub>

crystals to determine the symmetry of Cooper pairs and measure the dynamics of the chiral domains [7,8]. These experimental data on bulk Sr<sub>2</sub>RuO<sub>4</sub> should be considered as a result of ensemble averaging over possible chiral domain configurations. Thus we need a small enough sample of Sr<sub>2</sub>RuO<sub>4</sub> rather than the domain size to study phenomena that are peculiar to a single chiral domain such as dynamics of a single chiral domain, spin supercurrent, and unconventional vortices [9–11]. Transport measurements, however, have never been carried out yet in a single domain because it is also difficult to attach electrical contacts to submicron Sr<sub>2</sub>RuO<sub>4</sub> crystals.

In this Letter, we will report an anomalous property of the current–voltage (*I*–*V*) characteristics in a single domain of Sr<sub>2</sub>RuO<sub>4</sub>. The creation of vortices gives a finite resistivity even when the temperature is well below the superconducting transition temperature. In four-terminal measurements, the induced voltage *V* is usually an *odd* function of the bias current *I*. Namely, *V* changes its sign when we flip the direction of current to the opposite direction, which implies parity conservation [12]. However, we find in submicron Sr<sub>2</sub>RuO<sub>4</sub> samples that *V* has anomalous components which are *even* functions of *I*. The existence of the anomalous components means that positive voltage is detected regardless of the current direction and suggests the violation of parity [12]. To understand the nature of the anomalous *I*–*V* characteristics, we consider a simple

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model of a vortex which has a helical internal structure characterized by a Hopf invariant. We also show that hydrodynamics of such a helical vortex violates the parity to retain the topological invariant.

To obtain submicron  $\text{Sr}_2\text{RuO}_4$  single crystals, we synthesized  $\text{Sr}_2\text{RuO}_4$  crystals with a solid phase reaction and then determined the crystal structure of  $\text{Sr}_2\text{RuO}_4$  and the concentration of impurities. We prepared  $\text{SrCO}_3$  and  $\text{RuO}_2$  (both 99.9%, Kojundo Chem.) powders. The mixed powder was then heated at 990 °C for 60 h. The mixture was cooled gradually from 990 °C to 450 °C over 6 h. The samples were kept at 450 °C for 12 h to introduce oxide into the crystals and then cooled down slowly at room temperature. The structure of the  $\text{Sr}_2\text{RuO}_4$  crystals was analyzed by using X-ray power diffraction (Rigaku Diffractometer RINT 2200HK) with  $\text{Cu K}\alpha$  radiation. The observed peaks fitted a body-centered tetragonal unit cell of the  $\text{K}_2\text{NiF}_4$  type with lattice constants  $a = b = 3.867 (\pm 0.004)$  Å and  $c = 12.73 (\pm 0.01)$  Å [13]. The result of secondary ion-microprobe mass spectrometry (SIMS) shows that the concentration of Al in the sample is less than 100 ppm, while the superconductivity of  $\text{Sr}_2\text{RuO}_4$  is destroyed by nonmagnetic impurities [14].

We selected submicron  $\text{Sr}_2\text{RuO}_4$  single crystals from the observations of chemical composition and crystallinity. The samples were dispersed in dichloroethane by sonication and deposited on an oxidized Si substrate. We found typical samples in diameter of about 50 nm–500 nm. Energy dispersion spectroscopy (EDS; EX-64175 JMU, JEOL) was used to determine the components of the submicron samples on the substrate. The molar fraction of the Sr and Ru elements was 2:1. We also confirmed that the dispersed crystals had neither boundaries nor ruthenium inclusions on the sample surface by observing the crystal orientation using the electron backscatter diffraction pattern (EBSP; OIM TSL [15]).

On the analyzed  $\text{Sr}_2\text{RuO}_4$ , we fabricated gold electrodes using overlay electron beam lithography. Inset (a) in Fig. 1 shows a micrograph of our samples. The sample size is 2.50 mm  $\times$  1.88 mm  $\times$  0.10 mm. The sample electrode spacing is 0.63 mm. Since the fabricated sample surface may have the insulator surface of the layer crystals and the residual resist between the sample and the gold electrodes, it is difficult to form an electrical contact. Therefore we performed a welding using electron beam irradiation [16]. We heated each electrode on the sample for 15 s with a beam current irradiation of  $2 \times 10^{-7}$  A. As the result, we succeeded in greatly reducing the contact resistance below  $10^{-4}$   $\Omega$  at room temperature.

The measurements were carried out in a dilution refrigerator (Kelvinox, Oxford) with a base temperature of 60 mK. All measurement leads were shielded. The lead lines were equipped with low pass RC filters ( $R = 1 \text{ k}\Omega$ ,  $C = 22 \text{ nF}$ ). In the DC measurements, a bias current was supplied by a precise current source (6220, Keithley) and the voltage was measured with a nanovoltmeter (182, Keithley) using four-terminal measurements.

We measured the temperature dependence of the resistivity in the submicron  $\text{Sr}_2\text{RuO}_4$ . Inset (b) of Fig. 1 shows the temperature dependence of the resistivity in the  $ab$  plane from room temperature down to 4.2 K. Fig. 1 shows that the resistivity  $\rho_{ab} = 4 \text{ K}/6.0 \text{ m}\Omega \text{ cm}$ . This value is larger than the bulk resistivity by about three times [14]. We estimated the resistivity  $\rho_{ab}$  from the sample size. Since  $\text{Sr}_2\text{RuO}_4$  has anisotropic resistivity  $\rho_{ab} \approx \rho_c \times 10^{-3} \text{ m}\Omega \text{ cm}$ , the resistivity  $\rho_{ab}$  may actually be smaller than the estimation. Here the ratio  $\rho_{ab} = 300 \text{ K}/\rho_{ab} = 4 \text{ K} \sim 40$  is comparable to that of the bulk used in Ref. [6]. Hence we consider there is no degradation of the sample by the solvent. Fig. 1 also shows a transition temperature of  $T_c = 1.69 \text{ K}$  and a broader transition temperature width of  $1 T \approx 200 \text{ mK}$ . There was no decrease of the resistivity when a magnetic field of 3000 G was applied

Fig. 1. Temperature dependence of resistivity of submicron  $\text{Sr}_2\text{RuO}_4$  in zero magnetic field (0 G) and in a magnetic field ( $H = 3000 \text{ G}$ ) applied parallel to the  $c$  axis. Flat tail resistivity can be seen at low temperatures below  $T_c = 1.69 \text{ K}$ . Inset (a) shows a micrograph of a submicron  $\text{Sr}_2\text{RuO}_4$  single crystal connected to gold electrodes. Inset (b) displays the temperature dependence of the resistivity in the  $ab$  plane from room temperature down to 4.2 K.

parallel to the  $c$  axis. Our sample shows neither suppression of  $T_c$  nor enhancement to 3 K [14,17]. Here the resistivity retained its flat tail below  $T_c$ . The result shows that the flow of vortices can occur by quantum fluctuations of the superconducting phase [18]. The results show transport properties of the submicron  $\text{Sr}_2\text{RuO}_4$  single crystals because a broader transition temperature width and quantum fluctuations of the phase are characteristic of mesoscopic superconductors [19].

We observed anomalous  $I$ - $V$  characteristics in zero magnetic field. Fig. 2(a) shows  $I$ - $V$  curves at several temperatures with typical flat tail resistances of  $R^* \approx 0.16 \text{ }\Omega$ . In general, the voltage in  $I$ - $V$  curves for metals, quantum Hall systems and Josephson junction is always an *odd* function of the bias current, which is a result of parity conservation. Surprisingly,  $V$  is not an *odd* function of  $I$  at all. In what follows, we define anomalous nonlinear voltage (ANV) as the component of measured voltage given by an *even* function of  $I$ . An ANV implies the violation of parity. The amplitude of the ANV increases with decreasing temperature in zero magnetic field and shows a maximum below 200 mK. In order to eliminate completely the possibility of instrument malfunction in the DC measurements,  $I$ - $V$  curves were measured with a microvoltmeter (AM 1001, Ohkura Electric Co.) with a battery-powered current source. Furthermore, in the AC measurements, we also measured the differential resistance  $dV/dI$  as a function of the bias current using lock-in techniques. Fig. 2(b) clearly shows that  $dV/dI$  has an *odd* component of  $I$ . The parity violation in the  $I$ - $V$  characteristics is confirmed in both the DC and AC measurements. Moreover, we confirmed that the anomalous effect was reproduced in several samples.

To analyze the ANV in more detail, we subtract the linear part (ohmic contribution to voltage) from the  $I$ - $V$  curves in Fig. 2(a). The results are shown in Fig. 3. We clearly find that the ANV is symmetric with respect to the zero bias current, (i.e.,  $V_1 + |I| = V_1 - |I|$ ). Here the voltage  $V_1$  represents the ANV of the induced voltage  $V = R^*I + V_1$ . These curves are described well by a Lorentzian curve, as shown by the lines.

We discuss the physical difference between parity-violating  $I$ - $V$  characteristics and negative resistance. Negative resistance itself is an unusual phenomenon. The phenomenon, however, is possible. It is instructive to compare our result to the negative resistance of mesoscopic charge density waves (CDWs) reported



