Probing chiral superconductivity in \(\text{Sr}_2\text{RuO}_4\) \textit{underneath} the surface by point contact measurements

He Wang 1,2, Jiawei Luo 1,2, Weijian Lou 1,2, J E Ortmann 3, Z Q Mao 4, Y Liu 4,5 and Jian Wei 1,2

1 International Center for Quantum Materials, School of Physics, Peking University, Beijing 100871, People’s Republic of China
2 Collaborative Innovation Center of Quantum Matter, Beijing, People’s Republic of China
3 Department of Physics, Tulane University, New Orleans, LA 70118, United States of America
4 Department of Physics and Materials Research Institute, The Pennsylvania State University, University Park, PA 16802, United States of America
5 Department of Physics and Astronomy and Key Laboratory of Artificial Structures and Quantum Control (Ministry of Education), Shanghai Jiao Tong University, Shanghai 200240, People’s Republic of China

E-mail: weijian6791@pku.edu.cn

Keywords: p-wave superconductor, chiral domains, point contact measurement, strontium ruthenate, ferromagnetic superconductor, \(\text{Sr}_2\text{RuO}_4\)

Supplementary material for this article is available online

Abstract

\(\text{Sr}_2\text{RuO}_4\) (SRO) is the prime candidate for a chiral p-wave superconductor with critical temperature \(T_c\) \(\sim\) 1.5 K. Chiral domains with opposite chiralities \(p_x \pm ip_y\) have been proposed, but are yet to be confirmed. We measure the field dependence of the point contact (PC) resistance between a tungsten tip and an SRO–Ru eutectic crystal, where micrometer-sized Ru inclusions are embedded in SRO with an atomically sharp interface. Ruthenium is an s-wave superconductor with \(T_c\) \(\sim\) 0.5 K; flux pinned near the Ru inclusions can suppress its superconductivity, as reflected in the PC resistance and spectra. This flux pinning effect originates from SRO \textit{underneath} the surface and is very strong once flux is introduced. To fully remove flux pinning, one needs to thermally cycle the sample above \(T_c\) (SRO) or apply alternating fields with decreasing amplitude. With alternating fields, the observed hysteresis in magnetoresistance can be explained by domain dynamics, providing support for the existence of chiral domains. The origin of the strong pinning could be the chiral domains themselves.

1. Introduction

The mechanism of superconductivity (SC) in the layered perovskite ruthenate \(\text{Sr}_2\text{RuO}_4\) (SRO), the prime candidate for topological chiral p-wave superconductivity, is not clear after over 20 years investigation [1–5]. Results from experiments on muon spin relaxation [6] and the polar Kerr effect [7] suggest that the superconducting order parameter (OP) breaks the time-reversal symmetry and forms chiral domains with two different chiralities \((p_x \pm ip_y)\), similar to the domains in a ferromagnet. The existence of such chiral domains has not been conclusively confirmed, although there is indirect evidence: domain wall pinning was assumed in order to interpret the strong flux pinning (zero flux creep at lower temperatures) in measurements of \textit{bulk} magnetization relaxation [8–10]; and domain dynamics was also assumed to explain the field modulation of critical currents for corner junctions [11]. However, there is no direct evidence of the chiral domains. For example, an edge current around domain walls and sample edges, which should lead to measurable magnetic fields, has not been observed by \textit{local} field imaging methods on etched mesoscopic disks [12–15], nor by early micro-Hall probe studies near the edge of an SRO crystal [16].

Nevertheless, indirect evidence of p-wave superconductivity was reported in local transport measurements, including (scanning) tunnel junction spectroscopy [17–20] and point contact spectroscopy (PCS) [21, 22],

\[\text{DOI} \quad 10.1088/1367-2630/aa65c5\]
where the symmetry of the order parameter may be inferred by fitting the conductance spectra [19, 20], but this method may not distinguish the chiral edge states from helical states [23].

Unlike the above-mentioned local transport measurements, here we conduct PC measurements on the SRO–Ru eutectic system [24]. In the eutectic system, micrometer-sized Ru inclusions are embedded in SRO with an atomically sharp interface. This well-defined interface, between an s-wave elemental superconductor and a presumed p-wave superconductor, may lead to a spontaneous flux distribution [25], similar to the edge current at domain walls.

The most interesting and unexpected outcome of our PC measurements is that point contacts on Ru inclusions can be used for probing local flux: since ruthenium is a superconductor with a low critical temperature, $T_c$ (Ru) $\sim$ 0.5 K, its superconducting transition can be used to probe local flux. This approach differs in several respects from a Hall effect sensor in field imaging [12–14]: (1) Ru inclusions are embedded in an SRO matrix and largely underneath the surface, so we can probe the flux without any influence from surface degradation; (2) field imaging is usually done with field cooling and a close-to-equilibrium distribution of vortices, field sweeping is avoided, while here for PC measurements we focus on field sweeps and hysteresis; (3) the field range probed here is much larger than in field imaging, which is usually in the low-field limit due to the resolution of vortices. As will be shown, although the spontaneous flux [25] cannot be confirmed, we observed strong flux pinning (hysteresis of magnetoresistance) and clear domain dynamics, which are consistent with the proposal of chiral domains. Additionally, the measured PC spectra help us understand better the interaction of the order parameters in crystalline Ru and SRO with an atomically sharp interface, which is an interesting topic by itself [24, 26–28].

2. Experimental methods

Rod-like Ru inclusions on the surface of the cleaved SRO crystal can be seen clearly in the optical microscope image in figure 1(a), and more details are revealed in the scanning electron microscope image in figure 1(b), where surface degradation can be observed for the crystal that was exposed in ambient conditions for an extended period of time (water and carbon dioxide may react with SRO). Before the PC experiment, the surface is scratched with a ceramic knife to expose a relatively fresh surface.

The SRO single crystals are grown by floating-zone methods with Ru as self-flux [29] and with an excess amount of Ru, so that a eutectic phase is formed with embedded lamellar inclusions of Ru [24]. The resulting SRO–Ru crystal has an extended critical temperature for the superconducting transition ($T_c$) from the intrinsic 1.5 K to about 3 K, and this enhancement in $T_c$ is believed to originate from the interface between Ru inclusions and SRO, possibly due to lattice distortions and strain although the exact mechanism is not confirmed [24, 26–28]. In fact, it has been found that uniaxial pressure on pure SRO can enhance SC as well [22, 30–32]. Regarding the symmetry of the OP for this enhanced SC, a non-monotonic temperature dependence of the critical current (kinks near 1.5 K) and critical current switchings in so-called topological junctions suggested that the OP symmetry is different from that of pure SRO [33, 34].

In figure 1(c) a schematic of the tungsten tip and the eutectic SRO–Ru crystal is shown. The point contact is made between a tungsten tip and the SRO crystal, which is fixed on a silicon chip and mounted on the attoCube nanopositioner stack. The tip and the nanopositioner stack are both secured on a metal housing that is suspended by springs from the end of a cold-insertable probe for a Leiden cryogen-free dilution fridge. The base temperature for the sample stage is higher than the temperature of the probe at the mixing chamber stage is about 0.1 K ($\sim$ 0.2 mK) with (without) the point contact setup attached, while the base temperature for the sample stage is higher ($\sim$ 0.3 K) due to thermal loads from the wirings for the sample and nanopositioners, as well as Joule heating during the measurements. Differential resistance (dV/dI) is measured with a standard lock-in technique with home-made battery-powered electronics to reduce external noise. For more details please refer to our previous study on pure SRO [22].

Whether the surface property correlates with that of the bulk underneath the surface is always a concern for surface probes. This is particularly critical for SRO since its surface undergoes reconstruction after cleaving and the SC may be destroyed; also, surface contamination may result in a dead layer. One way to circumvent this problem is to make PCs with hard tips, so the surface layer may be penetrated by the tip [35, 36]. Using hard tungsten tips and our home-built point contact setup, we previously obtained a reproducible result for the SC gap ($\sim$0.2 mV) for pure SRO [22], consistent with that estimated by weak coupling theory.

Since Ru is much softer than SRO and has no dead layer on it, it is not necessary to push the tip to penetrate the surface layer in order to make PCs on Ru inclusions, as plotted schematically in figure 1(d). When the tip is sharp and can penetrate the surface dead layer at the beginning of the experiment, the interface can be made directly between the tungsten tip and the SRO, with Ru inclusions nearby (this type is referred to as W/SRO–Ru).

In figure 1(c) we also plot a cross-sectional view of the PC and the distance dependence of the superconducting OPs for SC in Ru and SRO, without considering intermixing of the two OPs. Results for a PC of
this kind showing both OPs, labelled as PC-1, are presented later in figure 4 due to the complexity. Later in the experiment, the tip gets flattened and the interface is normally between the flattened tip and the bulging Ru inclusions (W/Ru–SRO), as shown in figures 1(d) and (e). Measurement results for two such PCs, labelled PC-2 (in the same run as PC-1) and PC-3 (in a later run), are presented in figures 2 and 3 respectively. For PC-1 we focus on the hysteresis of magnetoresistance (MR) and order parameters shown in point contact spectra; for PC-2 and PC-3 we focus on how to remove the pinned flux by processes similar to thermal demagnetization and field demagnetization for a ferromagnet (see figure 1(f)).

In previous PC studies, flux pinning in a conventional superconductor was only considered in a few cases [37, 38]. MR hysteresis observed here was not reported in those previous PC measurements. It was usually assumed that making the point contact causes some damage and locally the SC is suppressed, thus vortices are trapped near the PC since the energy cost is lower. However, this is irrelevant here since for PCs on Ru inclusions we find that the origin of the flux pinning is not at the PC interface but within the SRO, i.e., flux pinning is not directly affected by any damage caused by making point contacts on Ru inclusions.

3. Results

For simplicity, we first describe the results for PC-2 and PC-3 where the tip gets flattened and the interface is normally between the flattened tip and the bulging Ru inclusions (W/Ru–SRO). The flux pinning effect and the difference between zero-field cooling (ZFC) and field cooling (FC) are presented, then methods to remove the flux pinning effect are introduced and compared with those in ferromagnetism.

3.1. PC-2 and PC-3, on Ru inclusions

During ZFC, as shown in figure 2(c) for PC-2 (figure 3(a) for PC-3), there is only one quick drop around 0.44 K (0.42 K), which is due to the superconductivity in Ru. The deviation from $T_C (\text{Ru}) \sim 0.5 \text{ K}$ could be due to uncertainty of the PC temperature: the local temperature at the point contact may be different from that of the substrate, on which the thermometer is attached near the sample but not in direct contact.

Figure 1. Images of Ru inclusions on the SRO surface and schematics for the point contacts. (a) Optical image showing Ru inclusions on the surface, which are about 1 µm wide of various lengths. The depth of these inclusions can be tens of microns [24] and it is undetermined here. (b) Scanning electron microscope image of the region within the red box. The Ru inclusions clearly bulge from the SRO surface, and the dendritic patterns indicate surface degradation. The scale bar in both images is 5 µm. After imaging, the surface is scratched with a ceramic knife to expose a relatively fresh surface before the PC experiments. (c) Schematic illustration of the PC between a tungsten tip and SRO surface (W/SRO–Ru), used to explain the two superconducting transitions shown in figure 4(a). Also shown is the assumed distance dependence of the order parameters of SC in Ru and SRO, without considering the coupling between them. The thick green line on the surface indicates the dead layer. (d) Blunted tip on a Ru inclusion (W/Ru–SRO). The chirality of the domains, $p_x \pm i p_y$, is shown by the clockwise or counterclockwise arrows, and is not fully developed for zero-field cooling (ZFC). With randomized chirality, the effective $M$ is close to zero. (e) For field cooling, as well as for ZFC but with field history, the polarization of the chiral domains is induced and remains when the field is reduced to zero. The magnetization states in (d) and (e) are indicated by black dots in the hysteresis loops in (f). Also in (f), we show (i) the initial magnetization curve (in green), which can be used for figure 2(e), (ii) the reversal curve (in brown) when we restart the field sweep routine at zero field in the reverse direction of the hysteresis loop, which can be used for figure 3(e), and (iii) the field demagnetization curve for figure 3(d). The two horizontal dashed lines in blue denote the critical fields ($\pm H_c$) for Ru inclusions, within which the PC resistance shows a dip due to the recovered superconductivity.
For PC-2, conductance enhancement at small bias corresponds to SC in Ru, which can be suppressed by raising the temperature above 0.5 K or by ramping the field above 200 Oe (see figures 2(a) and (b), as well as the illustration of initial magnetization in figure 1(f)), suggesting that this conductance enhancement is indeed sustained by SC in Ru inclusions. Note that after the field is ramped from 200 Oe back to zero, the conductance enhancement is still suppressed, as shown in the inset of figure 2(b), suggesting remnant flux or flux pinning in SRO (see figures 1(e) and (f)).

Alternatively, flux pinning is demonstrated by the hysteresis behaviour in MR at zero bias, as shown in figure 2(e). This hysteresis reminds us of the magnetization curve of a ferromagnet (see, e.g., the textbook [39], especially chapter 16 where the same terminology can be used for ferromagnetism and superconductivity). For simplicity of the model, we assume a soft ferromagnet with a reasonably large susceptibility, thus $M \approx B$. Near the coercive field $H_{\text{coer}} \sim 200$ Oe, both $M$ and $B$ pass through zero, and $B < H_{\text{coer}}$ (Ru) leads to the dip in resistance. Similar $H_{\text{coer}}$ were observed for another two PCs in the same run; see supplementary figure S2, and also for PC-1 in figure 3(e).

Figure 2. Point contact measurements for PC-2 (W/Ru–SRO). (a) Normalized PC spectra at different temperatures from 0.38 to 0.45 K after ZFC. (b) Normalized PC spectra at 0.4 K under different $H_{\perp}$. Notice the quick change from 150 Oe. The inset shows the difference in the PC spectra without field history (blue) and those with field history (green), both at $T = 0.38$ K and zero field. (c) Thermal demagnetization: after SC was suppressed by ramping $H_{\perp}$ at low temperature, the sample was warmed at zero field to different cycling temperatures $T_{\text{cycle}}$, then zero-field cooled. $T_{\text{c}}$ is shifted with different $T_{\text{cycle}}$. Inset: $R(T)$ curves up to 1.5 K. (d) $R(T)$ curves during field cooling from 1.5 K with opposite polarities. Inset: $R(T)$ curves up to 1.5 K. (e) MR curves for $H_{\perp}$ at $T = 0.38$ K. (f) MR curves for $H_{||}$ at $T = 0.38$ K.
Another feature of the MR curves is that after ZFC there is a small drop in resistance at around 100 Oe and then a quick increase to the normal state at a field close to 200 Oe, indicating first enhancement and then suppression of SC in Ru. The initial enhancement with increasing field could be due to some proximity effect from SRO, which may reduce the flux in the Ru (figure 1(d)), or is possibly related to the spontaneous flux proposed at the interface between an s-wave superconductor and a p-wave superconductor [25]. By further increasing the field close to $H_{\text{cusp}}$, the chiral domains get polarized since each chirality prefers a certain polarity of the field, and the resulting large $\mathbf{M}$ leads to total suppression of SC in Ru inclusions, even after the field is reduced to zero (figure 1(e)).

With a small field sweeping amplitude the dips in resistance near $\pm H_{\text{cusp}}$ are observed, as shown in figure 2(e) by the green line from 0.2 to $-0.2$ kOe, and the red line from $-0.2$ to 0.5 kOe. But with increasing field sweeping amplitude, the resistance dips were not observed for PC-2. This is probably because the growth of chiral domains is affected by the rate and amplitude of the sweeping field, and the bigger domains (or stronger
pining force) induced by higher fields cause a switching of the polarization that is too fast to resolve (the magnetization curve is still there but the probe is too slow to follow). Disappearance of the dip was also observed for another two PCs in the same run; see supplementary figure S2. Above \( H_{\text{coer}} \) there is almost no change in resistance (in figure 2(e)), indicating no influence of SRO on PC resistance.

**Resemblance to thermal demagnetization.** We further describe how such flux pinning can be removed. It was proposed that chiral domains behave like ferromagnetic domains, thus it is natural to consider how the polarization (magnetization) of the chiral (ferromagnetic) domains may be randomized. A ferromagnet can be demagnetized by thermal or cyclic field methods [39]. For thermal demagnetization, the ferromagnetic domains are randomized after the sample is heated above its Curie point and cooled in the absence of a field. Similarly, the suppressed SC at lower temperatures (due to a large \( M \), see figures 1(e) and (f)) can indeed be recovered by thermal cycling to \( T_c \) (SRO) \( \sim 1.5 \) K and then cooling in the absence of a field. As shown in

---

**Figure 4.** Results for PC-1 (W/SRO–Ru). (a) The temperature dependence of the resistance in the zero-field cooling process shows two drops, indicating the superconducting transitions of the SRO eutectic phase (from 2.3 K) and Ru inclusions (from 0.57 K) respectively. The transitions are totally suppressed with a perpendicular (out-of-plane) magnetic field of 10 kOe. Normalized differential conductance with field ramping history (b) at different temperatures from 0.4 to 2.5 K at zero field, (c) at 0.4 K under different perpendicular magnetic fields \( (H_\perp) \), and (d) compared with that without field history. The highest conductance at zero bias (lowest resistance) is marked in (b)–(d). (e) Hysteresis of the magnetoresistance \( \text{MR} \) at \( T = 0.4 \) K. Inset: \( \text{MR} \) with a larger sweeping amplitude; the dashed box denotes the field range in the main panel. (f) \( \text{MR} \) with in-plane field \( (H) \) at \( T = 0.4 \) K. In (e) and (f) different colors of the curves indicate the order of the field ramping.
amplitude of sweeping field. Also we minimize the waiting time between the measurements of two consecutive data points, and reduce the amplitude, which makes the domains smaller and thus has a negligible influence on the PC; secondly the OP of the 3 K phase may not be chiral p-wave [24] so there is no intrinsic vortex pinning mechanism.

This flux pinning effect is very symmetric to external fields. Starting from the same $T_{\text{cycle}}$ of 1.5 K, cooling with opposite fields (± 50 Oe, ± 75 Oe, ± 100 Oe etc, as shown in figure 2(d)) gives similar R(T) curves. These FC curves are also similar to those with different $T_{\text{cycle}}$ (compare figure 2(c)). Such a resemblance corroborates the existence of remnant magnetization when $T_{\text{cycle}} < 1.5$ K.

**Resemblance to field demagnetization.** The other option to revert to the magnetic ‘virgin’ state for the ferromagnet is to follow the field demagnetization procedure [39], by applying alternating fields of decreasing amplitude, which makes the domains smaller and/or randomly aligned. For PC-2, we only tried with increasing amplitude of sweeping fields. Later, in another run with the same crystal, we made PCs on Ru inclusions similar to PC-2, and we label one of them as PC-3. We start from large alternating fields and then reduce the amplitude; also we minimize the waiting time between the measurements of two consecutive data points, and reduce the step of field ramping, so the resistance drop is not too fast to register. Then the dips in resistance can be observed repeatedly, as shown in figure 3(c). The dip position now changed to around ±270 Oe, higher than ±200 Oe for PC-1 and PC-2, and the dip is much narrower, suggesting faster domain dynamics (narrow resistance dips were also reported for a magnetic insulator, reflecting magnetic domain dynamics [40]). Moreover, when the amplitude is further decreased from 0.4 → −0.3 → 0.2 → −0.1 → 0 kOe, as shown in figure 3(d), the resistance decreases and reaches a similar value to that at the bottom of the resistance dip near $H_{\text{coer}}$, suggesting that the local remnant field $M$ is minimized. As illustrated in figure 1(f), here there are no smaller loops (this is different from the analogy of a ferromagnet) and the only field to observe resistance dips is near $H_{\text{coer}}$.

For comparison, thermal demagnetization and field cooling with different fields are also shown for PC-3 in figures 3(a) and (b) respectively. Similar to that for PC-2 (figures 2(c) and (d)), here the remnant magnetization can be completely removed after thermal cycling to 1.5 K, and for field cooling there is excellent symmetry for both field polarities.

**Parallel field MR.** The parallel field MR data are less well understood, but still are briefly presented here for completeness (additional results of tilted field and in-plane anisotropic field MR are not included here, for simplicity). SRO is a layered superconductor with very less in-plane $H_{\text{c2}}(0) \approx 1.50$ T and out-of-plane $H_{\text{c2}}(0) \approx 0.075$ T [2], thus $H_{\text{c1}}$ with amplitude comparable to $H_{\text{c2}}(0)$ is not expected to affect SC in SRO. However, for PC-2 on Ru inclusions (W/Ru–SRO) we do observe resistance dips in the parallel field MR (see figure 2(f)), with even smaller $H_{\text{coer}}$, and the dip is also broader, about 200 Oe. We note that this is larger than $H_{\text{c1}}$ of the elemental superconductor Ru, which is about 25 Oe at 0.4 K (70 Oe at zero temperature) [41, 42]. Another difference compared with perpendicular field MR is that with smaller amplitudes of the sweeping field the resistance dip appears on the opposite side after sweeping across zero field (to depolarize the remnant field at $H_{\text{coer}}$), whereas with larger sweeping fields the resistance dip appears on the same side towards zero field, e.g., the dip appears near −40 Oe for −0.7 → 0.7 kOe, and at around 60 Oe for 0.7 → −1.4 kOe.

Parallel field MR for PC-3 is also shown in figures 3(e) and (f) for decreasing and increasing amplitude respectively. The resistance minimum near zero field gets lower for consecutive sweeps with decreasing amplitude, which bears some similarity to the MR with $H_{\perp}$ (figure 3(d)). For field sweeping with increasing amplitude, similar to that for PC-2 (figure 2(f)), at smaller field amplitudes the resistance minimum appears on the opposite side after crossing zero field, whereas at larger amplitude it moves towards zero field. Additionally, for field cooling, $H_{\text{c1}}$ has a similar but smaller suppression effect on SC of Ru inclusions (as shown in supplementary figure S3). One may think that there is finite $H_{\perp}$ component at the interface due to the inclined Ru/SRO interface, since the Ru inclusions are not aligned with any crystal orientation.

### 3.2. PC-1, near the SRO–Ru interface
For PC-1, the sharp tip penetrates through the dead layer (see figure 1(c) for illustration), so SC in both SRO and Ru may be probed. The SC in SRO is indicated by the gradual drop in resistance starting from about 2.3 K to 0.57 K (figure 4(a), and the quick drop below 0.57 K is due to SC of Ru, so the contact type is W/SRO–Ru. Note that the SRO/Ru interface does not contribute to the PC resistance, since the interface resistance is usually in the mΩ range (the interface area is usually of the order of 10 μm² [33, 34]), much smaller than the PC resistance.

With two OPs involved, we expect to see something different in the point contact spectra at finite bias. As can be inferred from the temperature and field dependence of the point contact spectra (figures 4(b)–(d)), the dips in conductance at around ±0.5 mV reveal information on the OP of SRO (see supplementary figures S4 and S5 for a similar feature for PC-2 and PC-3). The dips in conductance at ±0.5 mV and the broad zero-bias conductance hump are suppressed when temperature is raised to 2.5 K or the out-of-plane field ($H_{\perp}$) is increased to 10 kOe.
This is in striking contrast to the case for PC-2, where a similar feature is suppressed at 0.45 K and 200 Oe (the local field becomes larger than $H_c$ (Ru)). This suggests that for W/Ru–SRO type contacts, (PC-2 and PC-3) while the PC spectra are influenced by SC in SRO (large gap feature), this feature is still sustained by the conventional SC in Ru. Note that the gap value for elemental Ru is about 0.07 mV using the mean-field estimation with $T_c = 0.5$ K, which is much smaller than 0.5 mV observed here.

The most noticeable feature in addition to the spectra of PC-2 is a deflecting point at around $\pm$0.125 mV, as marked by the blue vertical dashed line, for the blue curve in figure 4(d). This feature is observed for the PC spectra of PC-1 at zero field with field history (finite M). When there is no field history (ZFC), the spectrum is different. As shown by the green curve in figure 4(d), an additional conductance dip evolves at $\pm0.185$ mV, as marked by the green vertical dashed line. The highest conductance without field history (6.47 $\Omega$) is close to that shown in the ZFC $R(T)$ curve (6.5 $\Omega$, figure 4(a)), as this differential conductance was measured right after ZFC. After field ramping, the highest conductance is reduced (7.06 $\Omega$), and the double dips at around $\pm0.185$ mV and $\pm0.5$ mV reduce to dips at $\pm0.5$ mV only, indicating that the OP in Ru is suppressed. This is again consistent with Ru inclusions serving as a local probe for M.

**Hysteresis in magnetoresistance.** For PC-1 there is clearly a MR hysteresis in the perpendicular applied field (figure 4(e)), similar to what we observed previously for PC on pure SRO [22]. The difference is that here there are additional dips in resistance at $H_{Coe}$ $\pm$ 200 Oe due to Ru inclusions, instead of a rounded valley for pure SRO (see supplementary figure S1 for comparison; for pure SRO, there are even Barkhausen-type jumps similar to the real magnetic domain dynamics). Such similar hysteresis confirms that the strong flux pinning is not due to the additional Ru inclusions but originates from SRO itself.

The presence of the resistance dip depends on the field history. Here is a good example: if the sweeping direction is reversed at zero field, as can be inferred from the hysteresis loops in figure 1(f), then there is no $M = 0$ point in that positive ramping direction until the field sweeps back again and crosses zero. As shown in figure 4(e), the field sweep follows the same routine, 0 → 1 → −1 → 1 → 0. So the last sweep is 1 → 0 kOe. Now we restart the field sweep at zero field, 0 → 1 kOe, opposite to the last sweeping direction; then there is no resistance dip at 200 Oe. When we sweep downward, 1 → −1 kOe, the resistance dip appears at $-200$ Oe, both consistent with our model.

The lowest resistance at the position of the resistance dip is around 6.9 $\Omega$, still larger than the resistance observed during ZFC (6.5 $\Omega$, see figure 4(a)). This may be understood if the suppression of SC in Ru is only partially reduced. One possible scenario is that, although the total M for multiple domains is zero at $H_{Coe}$, there could locally be inhomogeneous flux where local M is nonzero. For ZFC, domains were not magnetized (or not trained, as indicated by the dashed arrows in figure 1(d)) so there is no net flux.

For parallel field MR, as shown in figure 4(f), the data are less well understood than in the case of PC-2 and PC-3. There is no hysteresis, nor resistance dips at $H_{Coe}$. Instead, the resistance shows a broad minimum near zero field, and it is even possible to induce a sharp resistance drop near zero field. The broad minimum is consistent with the field dependence of a conventional superconductor. But if compared with the resistance in the perpendicular field MR (figure 4(f)), this change in resistance mostly corresponds to that for SRO, and only the sudden drop in resistance may be related to the Ru inclusion. In the scenario of domain dynamics, this may be interpreted as the chiral domains with out-of-plane polarization being randomized by $H_0$.

### 4. Discussion

Clearly there is strong vortex pinning in SRO, but its origin is still not certain. There are a few theoretical proposals available to understand the vortex state in SRO. First, Sigrist and Ageterberg studied the role of chiral domain walls in the vortex creep dynamics [43], which was used to explain the zero flux creep observed by Mota’s group [8–10]. In this picture the domain walls are pinned at impurities and lattice defects so they do not move easily (this is how the domain picture in figures 1(d) and (e) is derived). Second, Garaud et al [44] consider SRO as a type-1.5 superconductor with long-range attractive and short-range repulsive intervortex interaction. This is used to explain the vortex coalescence observed by scanning Hall probe microscopy [14] and possible clusters of vortices nucleating within a Meissner–like state implied by muon spin rotation ($\mu$SR) measurements [45]. Third, Ichioka et al [46] used the time-dependent Ginzburg–Landau theory to study the magnetization process and found that with increasing magnetic fields, the domain walls move so that the unstable domains shrink to vanishing size, and the single-domain structure is realized at higher fields. Along these lines there are theories of doubly quantized vortices and other exotic behaviours that may lead to a broken field with nonzero chirality degeneracy [47, 48]. Note that compared to the first proposal, chiral domain wall pinning is not emphasized, which to some extent suggests pinning by domains themselves and is probably more relevant to our observations here.
The proposal of chiral domain wall pinning was developed to understand the systematic experimental results on \textit{bulk} magnetization relaxation obtained by Mota’s group \cite{8-10}, where a novel strong flux pinning (even zero flux creep at the lowest temperatures) was found, and the higher the cycling magnetic field, the stronger the pinning effect. This is considered as indirect evidence of chiral domains. In our work the MR hysteresis also suggests strong pinning, but there are a few differences: (1) The previous scenario of chiral domain wall pinning seems inconsistent with the cycling field effect, since with higher cycling field, ‘polarization’ of the chiral domains in SRO is enhanced and domain walls are reduced, resulting in less pinning. Here we propose that the domains themselves can provide strong pinning (once they are formed with field history), and we compare the chiral domain dynamics to that of a ferromagnet; by assuming a local ‘magnetization’ (M) due to chirality polarization, the H_{core} is a natural explanation for the necessity of a high cycling field. (2) Previously the relaxation could not be measured at zero field, but here we can measure the point contact spectra in the ZFC situation, which probes the ‘virgin’ state without magnetization. (3) Previously the focus was the regime of zero flux creep at the lowest temperatures (50 mK and lower), while here the focus is the domain dynamics at higher temperatures (but still much lower than T_c (SRO)). (4) Previously, strong pinning for both H_{lab} and H_{all} was observed, but here only strong hysteresis for H_{all} is observed. The chiral domain walls should exist only in the \textit{ab} plane if the 2d $\gamma$ band is the active superconducting band, thus it is not clear whether the strong pinning for H_{lab} is due to the same mechanism. A further experiment, e.g., a study of ac susceptibility on the in-plane metastable vortex state \cite{49}, maybe helpful to investigate this issue.

The second model proposed to understand vortex clustering on the SRO surface is type-1.5 superconductivity, which was first named after the observation of vortex clustering on the surface of MgB$_2$ \cite{50, 51}, a two-band superconductor that has two weakly coupled order parameters with $\kappa_1 < 1/\sqrt{2}$ and $\kappa_2 > 1/\sqrt{2}$. In fact, for single-band superconductors with $\kappa \sim 1/\sqrt{2}$, there was also such a long-range attractive force \cite{52}. For SRO, the in-plane $\kappa_{\text{lab}} = 2.3$ and out-of-plane $\kappa_2 = 46$ are both in the type II regime \cite{2}. So to apply this theoretical model, Garaud \textit{et al} \cite{44} assume there are several coherence lengths in multicomponent superconductors \cite{44}, and find that type-1.5 behavior can occur in multiband chiral Ginzburg–Landau theories for SRO. This may explain the clustering of vortices imaged by a scanning field probe at low fields \cite{13, 14} and the bulk Meissner–like state implied by the $\mu$SR measurements \cite{45}. However, type-1.5 superconductivity alone cannot explain the MR hysteresis observed here for several reasons: (1) Similar MR hysteresis has not been found in the point contact measurements for MgB$_2$ \cite{53}. (2) The difference between types 1.5 and 2 is usually in the low-field region, but here the temperature and field ranges are outside the typical regions for Ginzburg–Landau theories. The dynamics here may involve only fully penetrated vortex domains instead of vortex domains mixed with Meissner-like domains in the low-field regime. (3) For a system close to type I, one does not expect to see hysteresis in $M(H)$.

The last model is pinning by chiral domains themselves. This model can explain the striking similarity to the ferromagnetism here. After ZFC, the applied field leads to the formation of chiral domains similar to ferromagnetic domains, which themselves become high-energy barriers for flux, instead of resorting to domain wall pinning. This is also consistent with the $\mu$SR experiment, in which a large fraction of the volume is vortex-free until the field is ramped to above 100 Oe. By assigning the quite large observed H_{core} to be the flip field for chiral domains, we have to abandon the previous belief that chiral domains flip easily \cite{16, 54}. In fact, H_{core} is much larger than H_{c1} measured by local magnetization hysteresis loops with a Hall probe \cite{16}, and is close to the thermal dynamic critical field \cite{2}.

The proposed chiral domains should not be mixed with conventional vortex domains, since it is possible to push conventional vortices of different vorticity into the chiral domains with a preferred vorticity but with a different energy cost. And the strong vortex pinning by chiral domains is absent for conventional vortex domain pinning by domain walls. From the MR results here, there is another feature that probably points to unconventional vortex pinning, i.e., for regular vortex pinning the MR usually does not show exact symmetry with respect to zero field \cite{11}, but here it does. The scenario of chiral domain wall pinning was also suggested for UPt$_3$ \cite{10}, but in later experiments it seems that a single domain without domain walls was inferred \cite{55, 56}. This can be explained if pinning is by domains themselves, and not by domain walls. There was also a disparity regarding the size of chiral domains \cite{57}, which was estimated to be around 100 $\mu$m or larger in measurements of the polar Kerr effect \cite{7}, and about 1 $\mu$m in measurements of the critical current for corner junctions \cite{11}. This can be reconciled if the domain size is determined by the internal defects, which are very sample-specific because of sample growth parameters, and then it may also be determined by the alignment in a multiple domain assembly, as drawn in the schematic illustration in figure 1.

As an additional note, it seems difficult to distinguish by the MR hysteresis alone the effect of strong flux pinning from the possible coexistence of ferromagnetism and superconductivity; the latter was proposed for the interface superconductivity between LaAlO$_3$ and SrTiO$_3$ \cite{38}, also an intriguing subject. From the aspect of the breaking of time-reversal symmetry, the difference between ferromagnetism and equal-spin triplet pairing is probably that the former has a static ferromagnetic order parameter while the latter does not.
5. Conclusion

By considering the lamellar inclusions of Ru embedded in single-crystal SRO as a local magnetization sensor, we found a new method to probe the local flux underneath the surface. The observed strong MR hysteresis with applied field perpendicular to the ab plane, and various field dependences (thermal demagnetization, field demagnetization, coercive field etc) indicate a striking similarity to ferromagnetic domains. Such a similarity provides indirect evidence of chiral domains and domain dynamics. We also discussed possible intrinsic pinning mechanisms, including chiral domain wall pinning [43] and type-1.5 superconductivity [50, 51], though both seem to have some difficulties in explaining the hysteresis. One remaining proposal is pinning by chiral domains themselves, which is new and needs further investigation. Besides the zero-bias point contact resistance, the point contact spectra at finite bias manifest the order parameters of both Ru and SRO, and thus might be helpful for understanding the interaction between order parameters of different symmetries, although more investigation is needed. Additional experimental investigation in this direction includes possibly scanning point contact measurement to check the proximity effect near Ru inclusions, and PCs with ferromagnetic or s-wave superconducting tips.

Acknowledgments

Jian Wei thanks Yoshiteru Maeno, Venkat Chandrasekhar, Jim Sauls, William P Halperin, Zhili Xiao, Haihu Wen, Peng Xiong and Laura Greene for discussions, and especially Weida Wu for the suggestion of finding a new method to probe the local flux underneath the surface. Jian Wei also thanks Egor Babaev, Dario Daghero and Simon J Bending for helpful email correspondence. Work at Peking University is supported by National Natural Science Foundation of China [NSFC, Grant No. 11474008] and National Basic Research Program of China (973 Program) through Grant No. 2012CB927400. The work at Tulane is supported by the US Department of Energy under EPSCoR Grant No. DE-SC0012432 with additional support from the Louisiana Board of Regents (support for material synthesis). The work done at SITU is supported by MOST of China (2012CB927403) and that at Penn State by DOE under DE-FG02-04ER46159.

References

[1] Maeno Y, Hashimoto H, Yoshida K, Nishizaki S, Fujita T, Bednorz J G and Lichtenberg F 1994 Nature 372 532
[2] Mackenzie A P and Maeno Y 2003 Rev. Mod. Phys. 75 657
[3] Maeno Y, Kettle S, Nomura T, Yonezawa S and Ishida K 2012 J. Phys. Soc. Japan 81 011009
[4] Kallin C 2012 Rep. Prog. Phys. 75 042501
[5] Liu Y and Mao Z-Q 2015 Physica C 513 339
[6] Luke G M et al 1998 Nature 394 558
[7] Xia J, Maeno Y, Beyersdorf P T, Fejer M M and Kapitulnik A 2006 Phys. Rev. Lett. 97 167002
[8] Mota A C, Dumont E, Amann A and Maeno Y 1999 Physica B 295 934
[9] Mota A C, Dumont E, Smith J L and Maeno Y 2000 Physica C 332 272
[10] Dumont E and Mota A C 2002 Phys. Rev. B 65 144519
[11] Kudwingira F, Strand J D, Van Harlingen D J and Maeno Y 2006 Science 314 1267
[12] Kirtley J R, Kallin C, Hicks C W, Kim E-A, Liu Y, Moler K A, Maeno Y and Nelson K D 2007 Phys. Rev. B 76 014526
[13] Hicks C W, Kirtley J R, Lippman T M, Koshnick N C, Huber M E, Maeno Y, Yuhasz W M, Maple M B and Moler K A 2010 Phys. Rev. B 81 214501
[14] Curran P J, Bending S J, Desoky W M, Gibbs A S, Lee S L and Mackenzie A P 2014 Phys. Rev. B 89 144504
[15] Lederer S, Huang W, Taylor E, Raghu S and Kallin C 2014 Phys. Rev. B 90 134521
[16] Yamazaki K, Tokunaga M, Mao Z and Maeno Y 2003 Physica C 388 499
[17] Upward M D, Kouwenhoven L P, Morpurgo A F, Kikugawa N, Mao Z Q and Maeno Y 2002 Phys. Rev. B 65 220512
[18] Firmo I A, Lederer S, Lapiren C, Mackenzie A P, Davis J C and Kivelson S A 2013 Phys. Rev. B 88 134521
[19] Kashiwaya S, Kashiwaya H, Karnbara H, Furuta T, Yaguchi H, Tanaka Y and Maeno Y 2011 Phys. Rev. Lett. 107 077003
[20] Kashiwaya S, Kashiwaya H, Saito K, Mawatari Y and Tanaka Y 2014 Physica E 55 25
[21] Laube F, Goll G, Löhnisen H V, Fogelström M and Lichtenberg F 2000 Phys. Rev. Lett. 84 1595
[22] Wang H, Lou W-J, Luo J-W, Wei J, Liu Y, Ortmann J E and Mao Z Q 2015 Phys. Rev. B 91 184514
[23] Scaffidi T, Romers J C and Simon S H 2014 Phys. Rev. B 89 220510
[24] Maeno Y, Ando T, Mori Y, Ohmichi E, Ikeda S, Nishizaki S and Nakatsuji S 1998 Phys. Rev. Lett. 81 3765
[25] Kaneyasu H and Sigrist M 2010 J. Phys. Soc. Japan 79 053706
[26] Yaguchi H, Wada M, Akima T, Maeno Y and Ishiguro T 2005 Phys. Rev. B 67 214519
[27] Ying Y A et al 2009 Phys. Rev. Lett. 103 247004
[28] Ying Y A, Staley N E, Xin Y, Sun K, Cai X, Fobes D, Liu T J, Mao Z Q and Liu Y 2013 Nat. Commun. 4 2596
[29] Mao Z, Macnab Y and Fukazawa H 2000 Mater. Res. Bull. 35 1813
[30] Kettaka S, Taniguchi H, Yonezawa S, Yaguchi H and Maeno Y 2010 Phys. Rev. B 81 180510
[31] Hicks C W et al 2014 Science 344 283
[32] Steppe A et al 2017 Science 355 148 http://science.sciencemag.org/content/355/6321/eaafl398.full.pdf
[33] Nakamura T, Nakagawa R, Yamagishi T, Terashima T, Yonezawa S, Sigrist M and Maeno Y 2011 Phys. Rev. B 84 060512
[34] Anwar M S, Nakamura T, Yonezawa S, Yakabe M, Ishiguro R, Takayanagi H and Maeno Y 2013 Sci. Rep. 3 2480
[35] Gloos K et al 1996 J. Low Temp. Phys. 105 37
[36] Gonnelli R S, Calzolari A, Daghero D, Ummarino G A, Stepanov V A, Fino P, Giunchi G, Ceresara S and Ripamonti G 2002 J. Phys. Chem. Solids 63 2319
[37] Shan L, Huang Y, Ren C and Wen H H 2006 Phys. Rev. B 73 134508
[38] Martínez-Samper P, Rodrigo J G, Agrait N, Grande R and Vieira S 2000 Physica C 332 450
[39] Cullity B D and Graham C D 2009 Introduction to Magnetic Materials 2nd edn (Hoboken, NJ: Wiley)
[40] Ma E Y, Cui Y T, Ueda K, Yang S, Chen K, Tamura N, Wu P M, Fujioka J, Tokura Y and Shen Z X 2015 Science 350 538
[41] Gella T H, Matthias B T, Hull G W and Corenzwit E 1961 Phys. Rev. Lett. 6 275
[42] Gibson J W and Hein R A 1966 Phys. Rev. 141 407
[43] Sigrist M and Agterberg D F 1999 Prog. Theor. Phys. 102 965
[44] Garaud J, Agterberg D F and Babaev E 2012 Phys. Rev. B 86 060513
[45] Ray S J, Gibbs A S, Bending S J, Curran P J, Babaev E, Baines C, Mackenzie A P and Lee S L 2014 Phys. Rev. B 89 094504
[46] Ichiioka M, Matsunaga Y and Machida K 2005 Phys. Rev. B 71 172510
[47] Garaud J, Babaev E, Bojesen T A and Sudbø A 2016 Phys. Rev. B 94 104509
[48] Sauls J and Eschrig M 2009 New J. Phys. 11 075008
[49] Shihtaka D, Tanaka H, Yonezawa S, Nojima T and Maeno Y 2015 Phys. Rev. B 91 104514
[50] Babaev E and Speight M 2005 Phys. Rev. B 72 180502
[51] Moshchalkov V, Menghini M, Nishio T, Chen Q H, Silhanek A V, Dao V H, Chibotaru L F, Zhigadlo N D and Karpinski J 2009 Phys. Rev. Lett. 102 117001
[52] Auer J and Ullmaier H 1973 Phys. Rev. B 7 136
[53] Daghero D and Gonnelli R S 2010 Supercond. Sci. Technol. 23 043001
[54] Curran P J, Khotkeyvych V V, Bending S J, Gibbs A S, Lee S L and Mackenzie A P 2011 Phys. Rev. B 84 104507
[55] Strand J D, Bahr D R, Van Harlingen D J, Davis J P, Gannon W J and Halperin W P 2010 Science 328 1368
[56] Schernn E R, Gannon W J, Wishne C M, Halperin W P and Kapitulnik A 2014 Science 345 190
[57] Kallin C and Berlingsky A J 2009 J. Phys.: Condens. Matter 21 164210
[58] Dikin D A, Mehta M, Bark C W, Folkman C M, Eom C B and Chandrasekhar V 2011 Phys. Rev. Lett. 107 056802