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Ferromagnetic SrRuO$_3$ thin-film deposition on a spin-triplet superconductor Sr$_2$RuO$_4$ with a highly conducting interface

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Ferromagnetic SrRuO$_3$ thin films are deposited on the ab surface of single crystals of the spin-triplet superconductor (TSC) Sr$_2$RuO$_4$ as substrates using pulsed laser deposition. The films are under a severe in-plane compressive strain. Nevertheless, the films exhibit ferromagnetic order with the easy axis along the c-direction below the Curie temperature of 158 K. The electrical transport reveals that the SrRuO$_3$/Sr$_2$RuO$_4$ interface is highly conducting, in contrast with the interface between other normal metals and the ab surface of Sr$_2$RuO$_4$. Our results stimulate investigations on proximity effects between a ferromagnet and a TSC. © 2015 The Japan Society of Applied Physics
of 500–700 °C and an oxygen partial pressure of 100 mTorr were employed with a base pressure of <3 × 10⁻⁷ Torr. It takes about 20–50 s to grow one unit cell at a laser intensity of 1.5 J cm⁻² and a repetition rate of 1–3 Hz. We also grew an SRO113 thin film on an insulating SrTiO₃ substrate as a reference sample, whose residual resistivity ratio (RRR) is ≈7, manifesting good performance of our growth facilities. The magnetization was measured down to 4 K with a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS-XL).

To measure the resistivity down to 4 K, we made a 1 × 0.7 mm² pad of SRO113 film and used a 4He cryostat (Quantum Design PPMS). The cleaved ab-surface of SRO214 usually consists of regions of typical size of 100 × 1000 µm² that look flat when viewed by an optical microscope. Indeed, within such flat regions, we can easily find atomically flat areas of the size of 10 × 10 µm² under an atomic force microscope (AFM). However, we also find several steps higher than the thickness of the film (50 nm) with a scanning electron microscope (SEM). If silver paste touches such a higher step, a direct contact between the silver paste and SRO214 through the ac-surface would be created. To avoid such a scenario, we carefully place contacts only at most flat surfaces under an optical microscope, and absence of direct contact was confirmed under SEM after measurements.

Figure 1(a) shows the X-ray diffraction (XRD) spectrum of a 15-nm-thick SRO113 film grown on SRO214. It shows no impurity peaks but only the (00l) peaks for SRO113 and SRO214, indicating that the c-axis of the SRO113 film lies in the same direction as that of SRO214 substrate. It is noted that the SRO113 (00l) peaks are located at smaller angles than those of the bulk (vertical red solid lines), indicating that our film has an elongated lattice along the out-of-plane direction. The out-of-plane lattice constant is estimated to be 4.00 Å in pseudo-cubic notation, which is 1.8% larger than the corresponding bulk value. AFM topographs of the film shown in the inset of Fig. 1(a) reveal that the film has atomically flat terraces with a step height equal to the lattice constant. Around the (001) and (002) peaks of SRO113, we observe that atomically flat top and bottom surfaces of SRO113 are formed.

In addition, the reflection high-energy electron diffraction (RHEED) patterns of the SRO113 film and the SRO214 substrate with the electron beam aligned along the [100] azimuthal direction are shown in the insets of Fig. 1(b). The orientation and distance of the first-order peaks with respect to the central peaks are invariant before and after the SRO113 film deposition. This invariance indicates that the in-plane crystalline axes of the film and substrate are aligned and thus provides evidence for the epitaxial nature of the film. It is noted that RHEED oscillations taken in situ during the film growth, as shown in Fig. 1(b), demonstrate the change-over of the film-growth modes from layer-by-layer to step flow at 500 s.²⁵ We also study an SRO113/SRO214 hybrid using high-resolution transmission electron microscopy (HR-TEM), which reveals that SRO113 films are grown epitaxially [see Fig. 1(c)].

Epitaxial growth of SRO113 on various perovskite substrates has been extensively studied:²⁵,²⁶ Under compressive strain along the in-plane direction, SRO113 films exhibit the c-axis elongation. We found that our SRO113 film has essentially the same peak positions as the one deposited on an NdGaO₃ substrate,²⁶ which has an in-plane lattice constant (3.86 Å in pseudo-cubic notation) very similar to that of SRO214.

Anticipating that the strong compressive strain could modify the magnetic behavior of the film, we measured its magnetization. Figure 2(a) shows the temperature dependence of the magnetization of a 50-nm-thick SRO113 film with a field of 1 mT along both the c-axis (out-of-plane) and the a-axis (in-plane) directions on field cooling. Surprisingly, we observe that T_{Curie} is 158 K, which is almost equal to the values for the bulk.²⁷,²⁸ In contrast, it is reported that high-quality SRO113 thin films deposited on SrTiO₃ substrates show a reduction of T_{Curie} to 150 K, although the a-axis mismatch between SRO113 and SrTiO₃ is only ~0.45%.²⁹ After careful subtraction of the background signals, we obtain the remanent magnetization of the film at 4 K to be 2.8 μ_B/Ru along the c-axis and 2 μ_B/Ru along the a-axis, which are substantially larger than the expected values.²⁹ The magnetization loops for both the c- and a-axis directions are presented in Fig. 2(b). These data show that our films exhibit a strong magnetic anisotropy with larger remanent magnetization along the c-axis.³⁰ We obtained essentially the same T_{Curie}, magnetization values, and anisotropy for a 15-nm-thick SRO113 film.

Figure 3(a) presents the temperature-dependent resistance R(T) between 300 and 4 K for a 50-nm-thick SRO113 film.

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**Fig. 1.** (a) XRD spectrum on a logarithmic scale of a c-axis oriented 15-nm-thick SRO113 film deposited on a SRO214 substrate. Vertical lines indicate the positions of the corresponding peaks of the bulk materials. Insets show an AFM topographic image giving an average roughness of ~0.5 nm and an atomic-step structure along the line given in the AFM image. (b) RHEED oscillations during the film growth. Insets show the RHEED patterns before and after the film growth. (c) HR-TEM image of a SrRuO₃/ Sr₂RuO₄ hybrid, taken from the [120] Sr₂RuO₄ zone axis.
We measured $R(T)$ using a dc four-probe technique with two different channel configurations: first with four contacts on the SRO113 film, $R_{12,34}(T)$, and second with two electrodes on the SRO113 film and the other two electrodes on the side of the SRO214 substrate, $R_{12,56}(T)$. A schematic of the sample with the electrode arrangements is shown in Fig. 3(b). Note that the electrodes were placed in a flat area, avoiding steps larger than the film thickness. The characteristic slope change at 120 K for both data sets suggests that the $c$-axis resistivity $\rho_c$ of SRO214 contributes significantly to transport for both configurations. It is obvious that $\rho_c$ contributes more for $R_{12,56}(T)$, as expected from the electrode configuration. Thus, $R(T)$ mainly reflects the behavior of SRO214 in the whole temperature range down to 4 K. This fact suggests that the SRO113 film has good electrical contact with the SRO214 substrate. A linear current–voltage ($I$–$V$) curve measured at 4 K with the 12,56 configuration of the electrodes also reveals that we obtain an Ohmic contact between SRO113 and SRO214. Note that the residual resistance ratio (RRR) for the 12,34 configuration is 60, which is many times larger than that of SRO113 thin films and much smaller than that of SRO214. This RRR value suggests a small but non-negligible resistance contribution of the interface in particular at low temperatures.

To further discuss the resistance of SRO113/SRO214, we use the simplified model circuit shown in Fig. 3(c). In this circuit, $R_{113}$ and $R_{\text{int}}$ are the resistances of the SRO113 film parallel and perpendicular, respectively, to the $ab$-surface, $R_{\text{int}}$ is the interface resistance, and $R_c$ and $R_a$ are the resistances of SRO214 along the $c$-axis and along the $ab$-plane, respectively. We should note that this simplified model estimates the upper limits for the measured resistances. Thus, the measured values should be lower than the estimated values. To estimate each resistance, we refer to existing resistivity data of SRO113 and SRO214. At 300 K, SRO113 thin films exhibit nearly isotropic resistivity $\rho_{113} \approx 250 \mu\Omega\text{cm}$, depending on the substrate. For any kind of substrate, $\rho_{113}$ decreases linearly with decreasing temperature down to $T_{\text{Curie}}$ and exhibits a sharp kink at $T_{\text{Curie}}$ owing to the reduction of scattering by spin fluctuations. The resistivity of SRO214 along the $c$-axis is $\rho_c \approx 15 \mu\Omega\text{cm}$ at 300 K, which is two orders of magnitude larger than the resistivity along the $ab$-surface, $\rho_{ab} \approx 120 \mu\Omega\text{cm}$. Whereas $\rho_{ab}$ decreases monotonically with decreasing temperature, $\rho_c$ exhibits a broad maximum at $\approx 120$ K. Using these resistivity values and the geometrical factors of the present hybrid system, we performed an order estimate of each contribution of the present system at 300 K, obtaining $R_{113} \approx 70 \Omega$, $R_{\text{int}} \approx 0.2 \mu\Omega$, $R_c \approx 10 \mu\Omega$, and $R_{ab} \approx 3 \mu\Omega$. We also estimated each contribution at 4 K using the values of $\rho_{113} \approx 10 \mu\Omega\text{cm}$, $\rho_c \approx 1 \mu\Omega\text{cm}$, and $\rho_{ab} \approx 1 \mu\Omega\text{cm}$, obtaining $R_{113} \approx 3 \Omega$, $R_{\text{int}} \approx 10 \Omega$, $R_c \approx 1 \mu\Omega$, and $R_{ab} \approx 20 \mu\Omega$.

In the present model, $R_{12,34}$ should satisfy the relation $1/R_{12,34} = 1/R_{113} + 1/(2R_{113} + 2R_{\text{int}} + 2R_c + R_{ab})$. Since $R_{113}$ at 300 K is estimated to be four orders of magnitude larger than the observed value of $R_{12,34} = 6 \mu\Omega$, the first term on the right-hand side should be negligible. In the second term, because $R_{113}$ is estimated to be very small compared with $R_c$ or $R_{ab}$, the $R_{113}$ term has little contribution. Thus, the contributions of the SRO113 film to $R_{12,34}$ should be
negligible. This is indeed consistent with the fact that the resistance anomaly at $T_{\text{Curie}}$ is absent in our results. Now, the relation can be reduced to $R_{12,34} \approx 2R_{\text{int}} + 2R_c + R_{ab}$. We can then notice that the estimated values of $2R_c + R_{ab} \approx 20\, \text{m}\Omega$ at 300 K and 2 m\Omega at 4 K and the observed $R_{12,34}$ value ($\approx 6\, \text{m}\Omega$) are on the same order. The observed temperature dependence of $R_{12,34}$ is also understood as a certain combination of $R_c$ and $R_{ab}$. Therefore, $R_{\text{int}}$ should have only a very small contribution. For $R(T)_{12,56}$, because contributions of $R_{113}^{11}$ and $R_{113}^{12}$ are again ignorable, we obtain $R_{12,56} \approx R_{\text{int}} + R_c + R_{ab} + R_c' + R_{ab}'$ (where $R_c$ and $R_{ab}$ are additional bulk resistances), and the same conclusion is deduced.

Our conclusion of the highly conducting SRO113/SRO214 interface apparently contradicts those of previous experimental works, indicating that the SRO214 $ab$-surface is not a good metal because of surface reconstruction accompanied by RuO$_6$ octahedral rotation. 1) Indeed, we usually cannot achieve good electrical contact between normal metals and the SRO214 $ab$-surface. In addition, such a surface reconstruction is thought to destroy the superconductivity in the $ab$-surface region of SRO214. Thus, the highly conducting interface in the present hybrid system is surprising. This observation indicates that the surface reconstructions might be suppressed under expansive surface strain on SRO214 caused by the epitaxial growth of SRO113 films. Detailed interface investigations would be interesting topics to address in the future.

In summary, we grew ferromagnetic epitaxial SRO113 thin films by PLD using cleaved single crystals of superconducting SRO214 substrates. The films were under severe compressive strain but with very small reduction in $T_{\text{Curie}}$ compared to the bulk. Resistivity measurements revealed that the interface between SRO113 and SRO214 is highly conducting. The epitaxial growth might relax SRO214 surface reconstructions and make the interface rather conducting. The SRO113/SRO214 hybrid system opens up the possibility of studying FM/TSC junctions in the future.

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