Growth of epitaxial $a$-axis and $c$-axis oriented Sr$_2$RuO$_4$ films

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Epitaxial films of Sr$_2$RuO$_4$ have been grown in situ by pulsed laser deposition on (100) LaAlO$_3$ and (100) LaSrGaO$_4$ substrates. X-ray diffraction results show that the films are single domain and grow $c$-axis oriented on (100) LaAlO$_3$ and $a$-axis oriented on (100) LaSrGaO$_4$ substrates. X-ray $\phi$ scans indicate epitaxial alignment of the film and substrate in-plane axes in both cases. Resistivity versus temperature measurements reveal that the as-grown $c$-axis oriented films are semiconducting and the $a$-axis oriented films are metallic. © 1996 American Institute of Physics. [S0003-6951(96)03504-1]

The compound Sr$_2$RuO$_4$ has been known for some time. It is the $n=1$ member of the Ruddlesden–Popper homologous series of the general formula $A_nB_2O_{3n+1}$ ($A=$Sr, $B=$Ru). Interest in this compound was recently renewed due to its low resistivity and excellent lattice match with YBa$_2$Cu$_3$O$_{7-x}$. These properties make it an attractive candidate for use as conductive electrodes or a normal metal barrier layer in device applications of high $T_c$ superconducting films. Lichtenberg et al. synthesized single crystals of this material and used it as a substrate for growing YBa$_2$Cu$_3$O$_{7-x}$. Further interest in this compound has been stimulated by the recent discovery of superconductivity in single crystals of Sr$_2$RuO$_4$ ($T_c=0.93$ K).

Sr$_2$RuO$_4$ is the only known layered perovskite that is free of copper, yet superconducting. In fact, it is isostructural with (La$_{1-x}$Ba$_x$)$_2$CuO$_4$, which was the compound in which high transition temperature ($T_c$) superconductivity was first discovered. By now, it is universally accepted in order to understand the physics of high $T_c$ superconductivity, one needs to understand the physics of CuO$_2$ planes found in all high $T_c$ superconductors, and the effect of carrier doping by the charge reservoir layers adjacent to the CuO$_2$ planes. It has been emphasized recently that while Sr$_2$RuO$_4$ shares many common features with high $T_c$ superconductors, important differences exist. In particular, the Cu$^{2+} (3d^9)$ valence state in CuO$_2$ planes has spin $\frac{1}{2}$ but Ru$^{4+} (4d^8)$ in RuO$_2$ planes has spin 1. This, together with the fact that Sr$_2$RuO$_4$ becomes superconducting without any extrinsic doping, suggests that mechanisms of superconductivity based on doping a 2D antiferromagnetic state of $\frac{1}{2}$ spins are not relevant for the occurrence of superconductivity in this compound. Furthermore, recent band calculations indicated that the Sr$_2$RuO$_4$ may be a much less strongly correlated electron system than high $T_c$ superconductors. This underscores the importance of a strongly correlated normal state in achieving high superconducting transition temperatur

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A Sr$_2$RuO$_4$ target for thin film deposition was fabricated by solid state processing methods. Stoichiometric proportions of SrCO$_3$ (Alfa Aesar, 99%) and RuO$_2$ (Alfa Aesar, 99.9%) powders were weighed out and ballmilled in isopropanol for 24 h. The ground powder was dried and then calcined at 1000 °C for 24 h in air. After regrinding, the powder was pressed into a disk of 2.5 cm diameter with a uniaxial press (10 MPa). This pellet was then further densified by pressing in a cold isostatic press (275 MPa). The pressed pellet was sintered in an Al$_2$O$_3$ crucible at 1350 °C for 24 h in air. X-ray diffraction analysis indicated that the target was a single-phase polycrystalline Sr$_2$RuO$_4$.

The films were grown in situ by pulsed laser deposition (PLD) using a KrF excimer laser (248 nm, Lambda Physik EMG103MSC) on single crystal (100) LaAlO$_3$ [indexed based on the pseudocubic perovskite subcell, or equivalently (012) of the rhombohedral unit cell] and (100) LaSrGaO$_3$ substrates. Deposition was done using an on-axis geometry, with the substrate placed normal to the ablated plume. The substrate was placed at a distance of 7.5 cm from the target and was radiatively heated. Oxygen pressures of 1–2 mTorr, substrate temperatures of 950–1050 °C, pulse energies of 90–130 mJ, and laser energy densities of 3–4 J/cm$^2$ were the deposition conditions used. The films discussed here were 1500–2500 Å thick. After growth, the films were cooled down to room temperature in vacuum. Films grown at significantly lower temperatures and higher oxygen pressures contained mainly SrRuO$_3$, consistent with previous observations.

A 4-circle x-ray diffractometer, using Cu $K\alpha$ radiation and a graphite monochromator, was used to characterize the films. A $\theta$–$2\theta$ scan of a Sr$_2$RuO$_4$ film on LaAlO$_3$ showed that the film is predominantly c-axis oriented with a c-axis length of 12.70±0.01 Å [see Fig. 1(a)]. The full width at half-maximum (FWHM) of the 006 peak in the $\theta$–$2\theta$ scan was 0.5°. The rocking curve FWHM of the 006 Sr$_2$RuO$_4$ peak was 0.4°. Figure 1(b) shows a $\phi$ scan of the 103 Sr$_2$RuO$_4$ reflection with sharp peaks (FWHM of 1°) at $\phi$=0°, 90°, 180°, and 270°, clearly indicating that the film is single domain with the (100) directions of the film being aligned with the (100) directions of the substrate. From the position of the 103 Sr$_2$RuO$_4$ peak, the in-plane lattice parameter was calculated to be $3.90\pm0.05$ Å.
measure the anisotropy in the film resistivity along these axes).

Resistivity versus temperature measurements of c-axis oriented Sr$_2$RuO$_4$ films grown on LaAlO$_3$ indicated higher resistivities (20 mΩ cm at room temperature) and semiconducting behavior at low temperatures. A minimum in the resistivity occurred in the range 70–160 K, depending on growth conditions. The mechanism for the observed semiconducting behavior is unclear at the present time. We note that Sr$_2$RuO$_4$ films grown from a stoichiometric SrRuO$_3$ target at low pressures (e.g., 20 mTorr) also exhibit higher resistivity and semiconducting transport behavior in contrast to SrRuO$_3$ films grown at higher pressure.\textsuperscript{17} More detailed transport measurements down to lower temperatures in both zero and finite magnetic fields are currently underway.

In conclusion, a-axis and c-axis oriented epitaxial films of Sr$_2$RuO$_4$ films have been prepared using PLD. X-ray diffraction scans showed that the films are single domain with excellent epitaxial in-plane alignment. Electrical transport measurements indicate that the a-axis oriented films are metallic while the c-axis oriented ones are semiconducting. Measurements to lower temperatures are needed to see if these films ultimately become superconducting. The successful preparation of epitaxial Sr$_2$RuO$_4$ films will likely provide new opportunities in studying superconducting and normal state properties of this interesting material. It may also help the effort of fabricating high quality oxide superconductor SNS junctions.

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