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COMMUNICATION – ELECTRONIC SUPPLEMENTARY INFORMATION (ESI)

First Complex Oxide Superconductor by Atomic Layer Deposition

Electronic Supplementary Information – Experimental Details

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Experimental Details

Thin films were deposited in an F-120 Sat reactor (ASM Microchemistry). A reactor temperature of 250 °C was maintained throughout all depositions. Nitrogen purging gas was supplied from gas cylinders (Praxair, 99.999 %), run through a Mykrolis purifier and maintained at a 300 cm^3 min^{-1} primary flow rate. Reactor operating pressure was stable at 2.8 mbar, with an increase to approximately 3.5 mbar during ozone pulses.

All depositions were carried out using $La(thd)_3$ (Volatec, 99 %), $Sr(thd)_2$ (Volatec, 99 %) and $Cu(acac)_2$ (Sigma-Aldrich, 97 %) (thd = 2,2,6,6-tetramethyl-3,5-heptanedionato, acac = acetylacetonato) as metal cation precursors. O_3 was used as the oxidizing agent, made from O_2 gas (Praxair, 99.996 %) using an In USA ozone generator (AC-2505) producing 15 mass % O_3 in O_2. Pulse durations were 4, 4, 4 and 2 s for $La(thd)_3$, $Sr(thd)_2$, $Cu(acac)_2$, and O_3 respectively. All purge durations were set to 3 s. Metal cation precursors were supplied from open boats in the reactor, kept at 185, 188 and 135 °C for $La(thd)_3$, $Sr(thd)_2$ and $Cu(acac)_2$ respectively.

The films were deposited on 3 x 3 cm^2 Si (100) substrates for characterization of thickness, conformality, uniformity and cation composition. Selected depositions were carried out on $LaAlO_3$ (100) (LAO, Crystal GmbH) substrates for facilitation of epitaxial growth.

Thin film thickness was measured using a J. A. Woollam alpha-SE spectroscopic ellipsometer in the range of 390-900 nm. A Cauchy function was successfully used to model the collected data.

Cation compositional analysis was carried out using a Panalytical Axios Max Minerals x-ray fluorescence (XRF) system, equipped with a 4 kW Rh-tube. The system is running with Omnian and Stratos options for standardless measurements of thin films.

X-ray diffraction measurements were performed on a Bruker AXS D8 Discover diffractometer equipped with a LynxEye strip detector and a Ge (111) focusing monochromator, providing $CuK\alpha_1$-radiation.

Scanning transmission electron microscopy was carried out using a FEI Titan 60-300 STEM at 300 kV. Sample preparation was carried out by mechanical polishing and ion milling on a Gatan PIPS II, cooled by liquid nitrogen. Collected high angle annular dark field (HAADF) images was treated and Fourier filtered using the Gatan Micsoscopy Suite software.

Room temperature resistivity measurements were performed using a 4-point probe and a Keithley model 2400 SourceMeter. The sheet resistivity was recorded by measuring resistance in 10 points from 1 to 10 µA, and averaged over 5 measurements on different spots close to the center of the single crystal samples.

Low-temperature resistivity measurements were carried out on a Quantum Design Model 4000 physical property measurement system (PPMS). The sample was mounted on a puck and contacted with silver paste and gold wire. Resistivity was collected in a 4-point setup, while the temperature was swept from 100 K to 4 K.

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