Structural and electronic properties of electron-doped Sr_{1-x}La_xCuO_2 epitaxial thin films grown by sputtering

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Abstract

We report on the synthesis of electron-doped Sr_{1-x}La_xCuO_2 thin films by a sputtering technique. Single phase c-axis oriented thin films were epitaxially grown on various oxide substrates, with better results on DyScO_3 and KTaO_3. Our preliminary results on the structural and electronic properties of the thin films are presented.

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The Sr_{1-x}La_xCuO_2 compound belongs to the family of electron-doped infinite layer (IL) superconducting cuprates (T_{c,max} = 43 K), composed of stacked CuO_2 planes, separated by Sr, La atoms. The synthesis of bulk ceramic material is very difficult, requiring very high pressure (3 GPa) and no single crystal has been prepared so far. Epitaxy allows to synthesize thin films [1,2] and offers the unique opportunity to obtain oriented samples needed for physical studies. We report our preliminary results on the synthesis, characterization and the superconducting properties of Sr_{1-x}La_xCuO_2 thin films (x = 0.08, 0.1, 0.15).

Epitaxial c-axis oriented thin films, 360–1500 Å thick, were prepared by on-axis, single target, rf magnetron sputtering on heated (650–690°C) single crystal substrate. Above 700°C, the electrical properties of the films were very poor especially for the films deposited on KTaO_3 substrates. In order to study the major role of epitaxy to obtain the IL phase, the following substrates, with increasing in-plane parameters, were used: (100)SrTiO_3, (100)KTaO_3, (110)DyScO_3. During the deposition the sputtering gas was a mixture of Ar/O_2 (0.5–1) and the total pressure was fixed at 200 mTorr. The distance between the target and the substrate was varied from 30 to 50 mm, allowing us to modulate the rate of deposition (0.45–0.15 Å/s). After deposition, the samples were cooled in pure Ar down to 500–650°C and kept at these temperatures in vacuum (about 3 × 10^{-6} Torr) for 10–60 min. To show evidence of the importance of the cooling conditions, several samples were cooled down in gas process directly to room temperature.

The crystal structure properties of the deposits were studied by X-ray diffraction (XRD). The morphology and roughness of samples surface were also observed by atomic force microscopy and interferential optical microscopy. A standard dc four-probe method, with sputtered gold pads, was used for resistance vs. temperature measurements and the results were compared with ac susceptibility measurements performed with a SQUID magnetometer. Compared to bulk samples, the signal in susceptibility measurements was very small (several 10^{-6} emu or less). So the signal arising from substrates or silver paste were carefully examined.

Fig. 1 shows a typical XRD pattern of a Sr_{0.9}La_{0.1}CuO_2 film, 360 Å thick, on a (110)DyScO_3 substrate. Only (00l) reflections of the IL phase and peaks of the substrates were detected, indicating that the sample is single phase and c-axis oriented.

Oscillations around the main reflections are clearly seen, meaning that the layers are very flat. Such oscillations were
much less visible for the films deposited on the other substrates. This is not surprising because of the small mismatch between the film and the substrate parameter, very close to that of the bulk material. The lattice constant $c$ was determined from (002) reflection, checked by substrate peak position, and, knowing $c$, parameter $a$ was determined from (202) reflection, out of $ab$ plane. Our results show that the lattice parameter $c$ decreases and $a$ increases with increasing substrate parameter or increasing La content. The substrate parameter values are 3.905, 3.944 and 3.988 Å for SrTiO$_3$, DyScO$_3$, KTaO$_3$, respectively, and the values of parameter $a$ of the thin films deposited on these substrates were about 3.935(5), 3.945(5) and 3.968(5) Å. Concerning parameter $c$, it actually depends also on the cooling process, in which the reduction of oxygen content was done. For instance, $c$ values were about 3.410(3), 3.415(3) Å, respectively for films on KTaO$_3$ which were kept at 620 °C and 550 °C in vacuum for 10 min and 3.444(4) Å for the sample directly cooled down to room temperature under gas process. So parameter $c$ is quite a good indicator to estimate the presence of excess oxygen. It should be noted that for the films with La = 0.15, the so-called long-$c$ phase [1] was present when they were oxygen reduced at high temperature.

Concerning resistance vs. temperature measurements, the results of most of our samples on DyScO$_3$ and KTaO$_3$ substrates were quite similar to the ones shown in Ref. [1], for a sample treated in $1 \times 10^{-5}$ mbar. At high temperature (above 150 K) the films show metallic behaviour followed by a semiconducting-like one down to the onset of the superconducting transition. The values of $T_c$ (onset) and $T_c$ (offset) were scattered from 9 to 20 K and 5 to 8 K, respectively. The $T_c$ ($R = 0$) values decrease below 4 K, when the samples were re-measured after 3 months. For the samples deposited on SrTiO$_3$, they generally show semiconducting behaviour, and only few samples had an onset of superconducting transition around 5 K. In contrast, in some samples on KTaO$_3$ substrates, a decrease of the resistance started from above 40 K with a low $T_c$ (offset) value as shown in Fig. 2.

Increasing measuring currents (see insert Fig. 2) reveals non-ohmic behaviour, which might be attributed to the presence of high $T_c$ superconducting islands, and restores a semiconducting-like curve. Interestingly, the angular dependence of the magnetoresistance (MR) measured at low $T$ (4.2 K) exhibits a deep minimum in perpendicular magnetic field. This result is similar to the angular MR behaviour in weakly anisotropic superconducting films with linear defects parallel to $c$ [3]. It shows that our IL films are weakly anisotropic as expected for this phase. It remains to identify the linear or planar defects responsible of this behaviour such as normal junctions between superconducting islands. It also remains to determine if this apparent “granular” behaviour is due to structural or doping inhomogeneities.

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References