Characterization of Microstructure in Ag-doped La_{2/3}Sr_{1/3}MnO₃ Films

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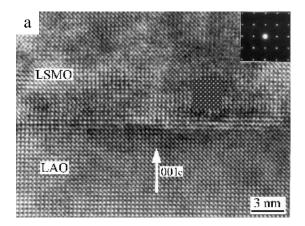
Epitaxial La_{1-x}A_xMnO₃ (A, alkaline metal) thin film is a kind of typical colossal magnetoresistance (CMR) material. Because the elements doping and microstructure may strongly influence magnetic and electrical properties of this material, the microstructure of epitaxial La_{2/3}Sr_{1/3}MnO₃ (LSMO) films and LSMO films doped with Ag were characterized using HREM.

Samples were prepared by co-deposition of LSMO and Ag on (100)-LaAlO₃ single crystal substrates using a dual-beam pulsed laser ablation system. The specimens were examined in a JEM 2010 HREM and a HF 2000 FEG AEM.

When the substrate temperature was 750 °C, the LSMO films were grown epitaxially not only along c-axis but also in the ab-plane. Interestingly, a reversible structural transformation from rhombohedral $R\overline{3}c$ to monoclinic I12/a1 in an epitaxial LSMO film, via electron beam irradiation, was observed by means of *in situ* HREM and electron diffraction, as shown in Fig. 1 and 2. Based on the simulations of HREM image and electron diffraction pattern (EDP), this transformation can be attributed to the cooperating effects of the mismatch stress and the irradiation-induced thermal-stress.

Ag-doped LSMO thin films were also systematically investigated in cross-section and plan-view by HREM. All films deposited at 750 °C were perfectly epitaxial for different doping level of Ag. No Ag was detected by EDS. As T_s decreased to 400 °C, texture is the distinguishing feature in LSMO films. With increasing the doping level of Ag, the size of grains was increased gradually and large clusters consisted of small grains were found in the doped films as indicated by arrows in Fig. 3. More importantly, Ag particle exists at the LSMO grain boundaries as shown in Fig. 4. The nanometer beam EDS analysis in LSMO grains was showed that Ag cannot substitute into LSMO lattice. However, Ag peaks were clearly visible in EDS spectra of large clusters, as shown in Fig 5. Quantitative analysis revealed that the concentration of Ag was varied in different clusters. The LSMO and AgO grains were assembled together to form the polycrystalline clusters. A growth mechanism of Ag-doped LSMO film was then proposed based on the experimental results. Nonmagnetic metal Ag existing at the grain boundaries and the polycrystalline clusters would weaken the ferromagnetic interaction between grains thus lead to a higher inter-grain MR value.

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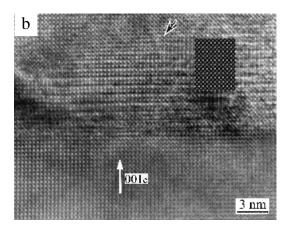


Fig. 1 HREM images of the LSMO film before (a) and after (b) electron beam irradiation. The insets are corresponding EDP and simulated HREM images with structures of $R\overline{3}c$ and I12/a1, respectively.

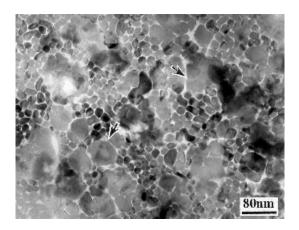


Fig. 3 Plan-view TEM image of the Ag-doped LSMO film.

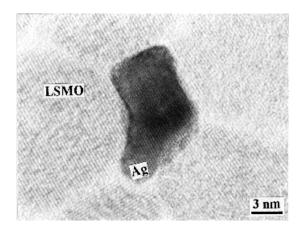


Fig. 4 HREM image of the Ag-doped LSMO film, showing a Ag particle exists at the grain boundaries.

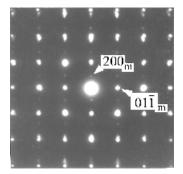


Fig. 2 The corresponding EDP of Fig. 1b.

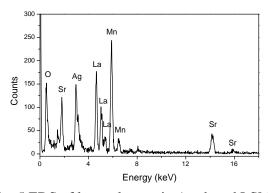


Fig. 5 EDS of large clusters in Ag-doped LSMO films.