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Disordered Phases in Magnetoresistance Sr_2FeMoO_6 Induced by Cation Deficiency

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The double perovskite phase Sr_2FeMoO_6 (SFMO) has potential magnetic device applications at near room temperature. The important low-field magnetoresistance (LFMR) of SFMO at ≤1T has been reported to be very sensitive to its microstructure, including grain size, grain boundaries, and antisite defects. In this study, we applied transmission electron microscopy (TEM) techniques to investigate two SFMO samples with very different magneto-transport properties. We found that cation deficiency, which was introduced by typical reducing environments, results in striped intergrowth phases with cation disorder in SFMO and therefore has deleterious effect on the LFMR of SFMO.

SFMO samples were fabricated by calcining at 950°C ball-milled stoichiometric amounts of $SrCO_3$, Fe_2O_3, and MoO_3 and then sintering in flowing Ar-1% H_2 at either 1300°C for 5h (sample A) or 1200°C for 24 h (sample B). TEM investigation was carried out in JEOL 2010F TEM/STEM.

Sample A presented much better LFMR values than sample B. Structural investigation suggested that the major structural difference between the two samples is the amount of stripped intergrowth phases seen in both samples and that the stripped phases deteriorate the LFMR of SFMO. Figure 1 shows a low magnification Z-contrast image of a SFMO crystal in which stripes appear darker than their surrounding normal area.

Comprehensive study suggested that the stripped phases are cation deficient $SrFe_{1-x-y}Mo_{x-z}O_{3-\delta}$, promoted by typical reducing formation conditions for SFMO in Ar-1%H_2 gas. Electron diffraction patterns (EDPs) (assuming the SFMO is of an fcc structure) suggest that the stripped phases are of superlatice structures with different superlattice periods along [100], which is perpendicular to the stripes. Figure 2 shows [001] EDPs taken from (a) a normal area and (b) and (c) striped areas.

High-resolution Z-contrast images show that the superlattice periods increase with local image brightness (see an example in Figure 3 (a)), indicating local composition dependence of the periods. More importantly, while elemental arrangement in normal areas is very well ordered and matches the ordered fcc SFMO structure as seen in Fig.3 (b) and (c), cation elements in stripped phases are disordered as evidenced by the random atomic column intensity distribution in Fig. 3 (d) and (e). The disordered elemental distribution in the striped phases is the source that reduces the LFMR.

Detailed analysis on (1) the structures of the stripped phases based on high-resolution Z-contrast images and their Fourier transformation and (2) the synthesis conditions that can reduce the amount of the striped phases and therefore improve the LFMR of SFMO will be presented.

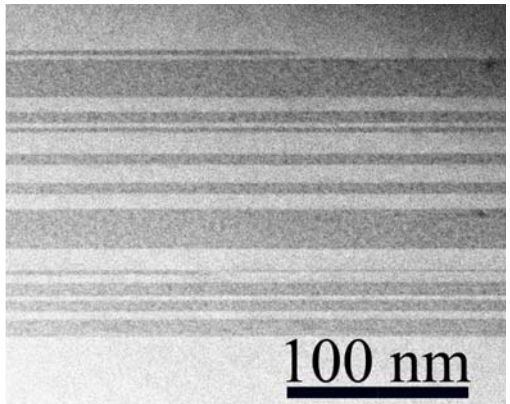

Fig. 1. A low magnification Z-contrast image of the striped phases, which show dark contrast, intergrown in a SFMO crystal.

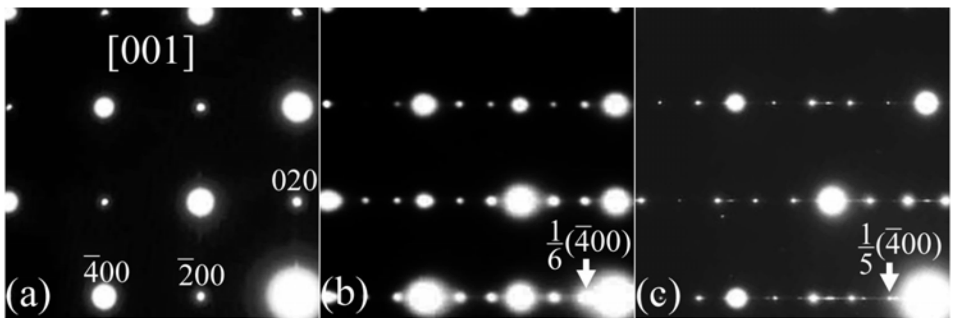

Fig. 2. [001] EDPs taken (a) from a normal area and (b) and (c) from stripped phases. The pattern in (a) can be indexed based on an fcc structure. Extra superlattice spots along [100]* with periods of $\frac{1}{6}[400]^*$ and $\frac{1}{5}[400]^*$ are seen in (b) and (c), respectively.

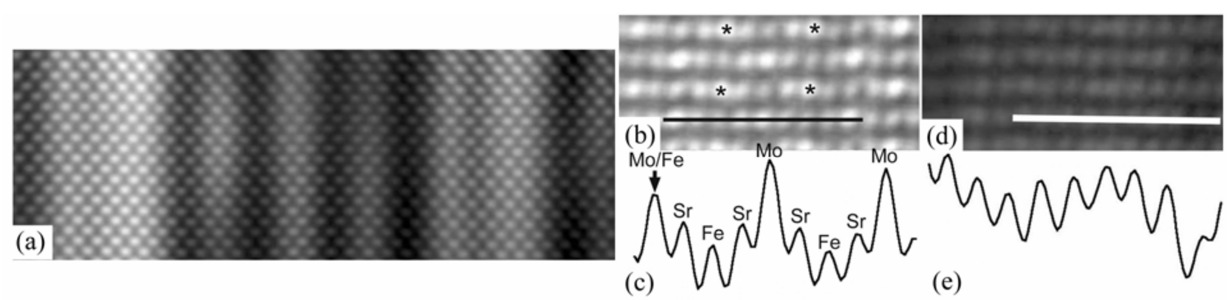

Fig. 3. (a) A [001] Z-contrast image of a stripped area showing varying superlattice periods; (b) a [011] Z-contrast image of a normal area. Four asterisks mark a unit cell of the ordered fcc SFMO structure; (c) the intensity profile along the black line in (b), indicating ordered elemental distribution. The left end peak shows a reduced Mo peak caused by the disordering of Fe onto the Mo site, as is usual in SFMO; (d) a [011] Z-contrast image of a striped area; and (e) the intensity profile along the white line in (d), showing random intensity distribution.

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