

# High Field and Low Temperature X-ray Study on Phase Segregation for $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ Powder and Single Crystal

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## Abstract

It has been found that the coexistence of the high temperature ferromagnetic (FM) and the low temperature antiferromagnetic (AFM) phases was observed below the charge order transition temperature  $T_{co}$  for the  $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  powder sample. In order to investigate details of the phase segregation, the X-ray diffraction was carried out for the  $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  single crystal in magnetic fields up to 5 T. It was found that the phase segregation is strongly depends on the wave length of X-ray. These results suggest that the phase segregation of FM and AF mainly occurs near the surface within a few  $\mu\text{m}$  in depth.

*Key words:* manganite; charge order; phase segregation; X-ray diffraction; high magnetic field

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It is well known that the structure, charge, orbital and magnetic order transitions occur at the same time for the perovskite manganite [1]. Moreover, the phase segregation is observed by many experiments. In the absence of a magnetic field, electron and X-ray diffraction experiments have exhibited the coexistence of ferromagnetism and incommensurate charge-ordering in a narrow temperature width [2]. Moreover, a recent high resolution lattice image study shows that a fine mixture of the antiferromagnetic incommensurate charge-ordered and the ferromagnetic microdomains exist, and this proves the phase separation for  $\text{La}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$  [3]. In the high magnetic field, Allodi et al. reported that the two phases of the ferromagnetic and antiferromagnetic phases are observed at 7T by NMR technique [4]. The phase separation phenomena, however, is still unclear in the presence of a magnetic field. In this paper, we study on the phase segregation both of the powder and single crystal  $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  by using a high magnetic field and low temperature X-ray diffraction [5].

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X-ray diffraction was performed in high magnetic fields up to 5 T and low temperature ranging from 300 to 10 K for both single crystal and powder  $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$ . In particular, various kinds of X-ray sources of Cu, Ag and Mo were utilized in order to investigate surface effects. The  $\text{Nd}_{0.5}\text{Sr}_{0.5}\text{MnO}_3$  single crystal used in this study was grown by a traveling solvent floating zone (TSFZ) method and it was annealed for 48 hours in  $\text{O}_2$  atmosphere. The powder sample was made by grinding the obtained single crystal.

Figures 1 (a)-(c) show the X-ray diffraction pattern for the powder sample on cooling at 3 T. At high temperature, only the reflections of the high temperature FM phase are observed. Other peaks appear on cooling across the charge ordered temperature  $T_{co} \approx 113\text{K}$ . These results indicate that the discontinuous jump of the lattice constant at  $T_{co}$  and are in good agreement with the previous papers [1]. However, high temperature phase still remained at 9 K as shown in Figs. 1 (a)-(c). Hence, this corresponds to the coexistence of the high temperature FM and the low temperature AFM phases. At zero field, the tendency of the phase coexistence becomes small and the volume of the FM

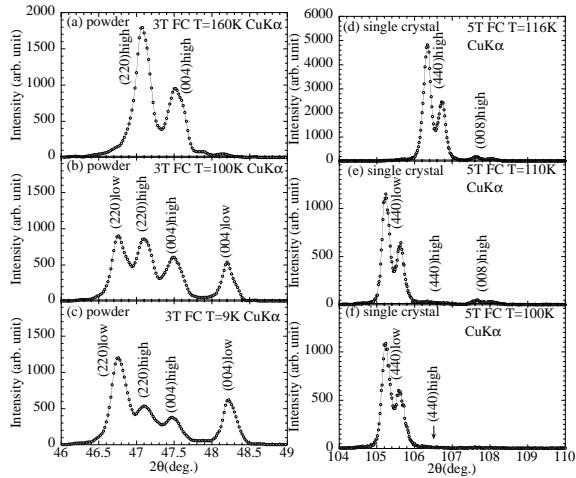


Fig. 1. X-ray diffraction patterns on cooling at 3 T for powder sample (a)-(c) and 5T for single crystal (d)-(f)

phase into the AFM one at low temperature increases with increasing magnetic fields [6]. Moreover, the estimated volume fraction of the FM phases from the X-ray diffraction data is very similar with those defined from the magnetization measurement [6]. Shimomura et al also have reported the X-ray diffraction experimental results in magnetic field for single crystal [7]. According to them, however, no any phase segregations are observed in case of the single crystals. In order to the study this paradox, we carried out the X-ray diffraction experiments for single crystal. Figures 1 (d)-(f) show the X-ray diffraction patterns for the single crystal. In this case, the two reflection peaks of (008) and (440) are observed at the same experimental condition in spite of the single crystal. This means that the single crystal used in this study consists of the two domains with different crystal axes. Since we focus on the difference of the phase segregation phenomena between "bulk" single crystal and powder, this domain structure is not serious problem in this study. We set the crystal so that the (440) reflection of the high temperature FM phase becomes maximum at every measuring temperature. As similar with the case of the powder sample, only high temperature phase are observed and still remains slightly at 100 K below  $T_{co}$ . At 80K, the FM phase remains slightly but is very small compared with the case of the powder sample as shown in Fig. 1 (c). The temperature region of the phase coexistence in this case is from 115K to 100K. Therefore, the phase segregation hardly occurs for the single crystal and this is reasonable to the previous results [7]. If we use various kinds of X-ray sources, the penetration depth can be changed since wavelength of the X-ray is different. Figure 2 summarizes the phase segregation temperature range on cooling at 5 T for various X-ray sources. The calculated penetration depth of CuK $\alpha$ , MoK $\alpha$  and

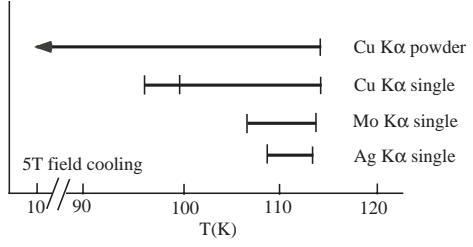


Fig. 2. Phase segregation temperature region on cooling at 5T

AgK $\alpha$  are about 10, 30 and 50  $\mu$ m, respectively. The phase segregation region becomes narrower with larger penetration depth. Hence, the surface with about a few  $\mu$ m in depth plays an important roll for the phase segregation phenomena. Since the powder sample consists of large amount of surface area, it is considered that the phase segregation is enhanced. If powder samples are used for experiments such as Neutron diffraction or the measurements on the surface such as the scanning probe microscopy, much attention is necessary.

In conclusion, we study the phase segregation phenomena on charge ordered phase of  $Nd_{0.5}Sr_{0.5}MnO_3$  by using the high magnetic field and low temperature X-ray diffraction. We found that the phase segregation mainly occurs near the surface area.

## Acknowledgements

X-ray diffraction experiments in magnetic field were carried out at the High Field Laboratory for Superconducting Materials, Institute for Materials Research, Tohoku University.

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