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X-ray diffraction study of commensurate-incommensurate crossover in $La_{2-x}Sr_xNiO_4$

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Introduction

$La_{2-x}Sr_xNiO_4$ (LSNO) is a well-known system to show an incommensurate spin-charge order with incommensurability $\varepsilon \sim x$, so-called a stripe order, in a wide hole-concentration region ($0.1 < x < 0.7$) [1]. Recently, a temperature dependent behavior of the stripe superstructure periodicity was observed by neutron diffraction measurement at around $x = 1/3$ [2], where the stripe order is remarkably stabilized. Similar phenomena have been observed in various kinds of helical magnets, ferroelectric materials, *etc*, which in some cases are called “Devil’s staircase”. In this study, we investigated such commensurate-incommensurate crossover appearing in charge order system as well, utilizing the synchrotron x-ray diffraction with high Q-resolution ability.

Experimental

The samples used in this work are LSNO single crystals grown by floating-zone method, whose average crystal structures are all tetragonal (*I*4/*mmm*). Their compositions were confirmed by ICP measurements. The typical sample size was $2\times2\times1$ mm^3 with (1 1 0) oriented plane polished for its optical flatness. The x-ray diffraction measurement was performed using four-axis diffractometer at BL-4C with a fixed incident photon energy of E_i = 13 keV. The temperature was controlled carefully in the region of 10 - 300 K by using a closed-cycle He refrigerator.

Result and discussion

The temperature dependence of the superlattice peak profiles for LSNO samples (x = 0.31, 0.333, and 0.35) along (h h 1) direction are plotted in Figure 1. The evolution of these superlattice peaks at (2-ε 2-ε 1) corresponds to the appearance of the incommensurate charge stripe modulation. All of the peak intensities are normalized using a standard Bragg reflection. The superlattice peak starts evolving at the highest temperature (~ 230 K) for x = 0.333, with symmetric and sharp profiles compared to other samples. It indicates the stable and long-range characteristics of the charge order at $x = \varepsilon = 1/3$. As for the peak positions, a strongly temperature dependent ε is observed for x = 0.31; ε varies from 0.305 (50 K) to 0.327 (190 K) as increasing the temperature. Such a behavior indicates that the periodicity of the stripe tends to approach $3\times\sqrt{2}\,a$ at high temperature, despite some possible energy cost caused by the deviation of the hole density on the stripe (p_{st}) from p_{st} = 1. On the contrary, such a commensurate-incommensurate crossover is not observed for x = 0.333 and 0.35, with nearly temperature independent incommensurability ($\varepsilon \sim$ 0.332 and 0.346, respectively). It may be related to the asymmetric character of doped particles or holes, since the ε = 1/3 charge ordered state can be viewed as a Mott insulator.

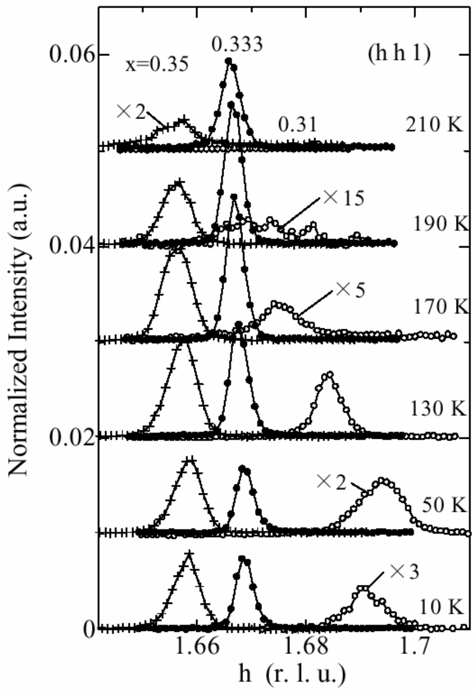

Figure 1: Peak profiles of the (2-ε 2-ε 1) superlattice for x=0.31, 0.333, and 0.35.

References

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