X-ray diffraction study of commensurate-incommensurate crossover in La$_{2-x}$Sr$_x$NiO$_4$

Kyoko ISHIZAKA$^1$, Taka-hisa ARIMA$^2$, Youichi MURAKAMI$^3$, Ryoichi KAJIMOTO$^4$, Hideki YOSHIZAWA$^5$, and Yoshinori TOKURA$^1$

$^1$Univ. of Tokyo, Bunkyo-ku, Tokyo 113-8656, Japan

$^2$Univ. of Tsukuba, Tsukuba, Ibaraki 305-8573, Japan

$^3$Tohoku Univ., Sendai, Miyagi 980-8577, Japan

$^4$Ochanomizu Univ., Bunkyo-ku, Tokyo 112-8610, Japan

$^5$ISSP, Univ. of Tokyo, Tokai, Ibaraki 319-1106, Japan

Introduction

La$_{2-x}$Sr$_x$NiO$_4$ (LSNO) is a well-known system to show an incommensurate spin-charge order with incommensurability $\epsilon$~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 ($I4/mmm$). Their compositions were confirmed by ICP measurements. The typical sample size was $2 \times 2 \times 1$ 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 $(2-\epsilon 2-\epsilon 1)$ 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 $x = \epsilon = 1/3$. As for the peak positions, a strongly temperature dependent $\epsilon$ is observed for $x = 0.31$; $\epsilon$ 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 ($\epsilon \sim 0.332$ and 0.346, respectively). It may be related to the asymmetric character of doped particles or holes, since the $\epsilon = 1/3$ charge ordered state can be viewed as a Mott insulator.

References


* ishizaka@cmr.t.u-tokyo.ac.jp