Inverse Order-Disorder Transition of Charge Stripes - Supplementary Material

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CHARGE AND MAGNETIC CORRELATIONS

Synchrotron X-ray scattering experiments were carried out on the beamlines BL07 and SP12B1 of NSRRC, Taiwan. The sample was mounted on a closed-cycle cryostat mounted on a multi-circle diffractometer, which allows the scans to be performed along any of the reciprocal space crystallographic axes, $H(= 2\pi/a)$, $K(= 2\pi/b)$, and $L(= 2\pi/c)$. Throughout this study for $La_{5/3}Sr_{1/3}NiO_4$, a tetragonal unit cell with lattice parameters of $a = b = 5.4145$ Å $= 2\sqrt{2}d_{\mathrm{Ni-O}}$ and $c = 12.715$ Å was used to index the reflections. There was no realignment of the crystal during the measurements because $La_{5/3}Sr_{1/3}NiO_4$ does not display any structural phase transitions at low temperatures [1]. The incident x-ray energy was set to 10 keV by a pair of high quality single crystals of Si(1 1 1), and a LiF crystal was used in an analyzer to define the scattered x-rays from the sample. The experimental resolution function was determined to be $\epsilon_H^{-1} \sim 0.0019$ Å^{-1}, $\epsilon_K^{-1} \sim 0.001$ Å^{-1}, and $\epsilon_L^{-1} \sim 0.015$ Å^{-1} as measured on the Bragg peak (4 0 0), which is near the charge ordering peaks measured at $T = 140$ K, and the sample mosaic width was found to be $\sim 0.02°$. The peak profiles of the Bragg reflection (4 0 0) were monitored throughout the measurements and showed no changes. The correlation lengths of the charge stripe reflections were extracted from their measured peak profiles convoluted with the resolution functions, and the error bars shown in this study were taken from the square-root of the data points. Measurements were taken as a function of temperature through the Bragg peak and charge stripe satellites along the crystallographic axes of H, K, and L in the reciprocal space.

Figure 1 shows how the peak widths of the charge modulation along the H- and K-directions change as a function of temperature. As can be seen, for temperatures above T_{CO}, the charge modulation is isotropic in the $a \times b$ plane, but as temperature is lowered, there is an anisotropic evolution of the correlation lengths. Figure 2 displays the evolution of the peak profile of charge modulation along c-axis as a function of temperature. As temperature is decreased, the peak narrows at first, indicating an increase in order along the c-axis, but then it widens again, indicating an inverse order-disorder transition.

For the study of spin stripes, in order to enhance the signals from the spin modulations, a resonant soft x-ray diffraction experiment was conducted on the beamline BL05B3 of NSRRC. The measurements were performed to scan the spin stripe reflection (0.66 0 0) through the L edge of Ni. A large resonance from the spin reflection was observed at the L_3 edge of Ni with incident π-polarized x-rays at T = 80 K. Upon warming, the spin ordering

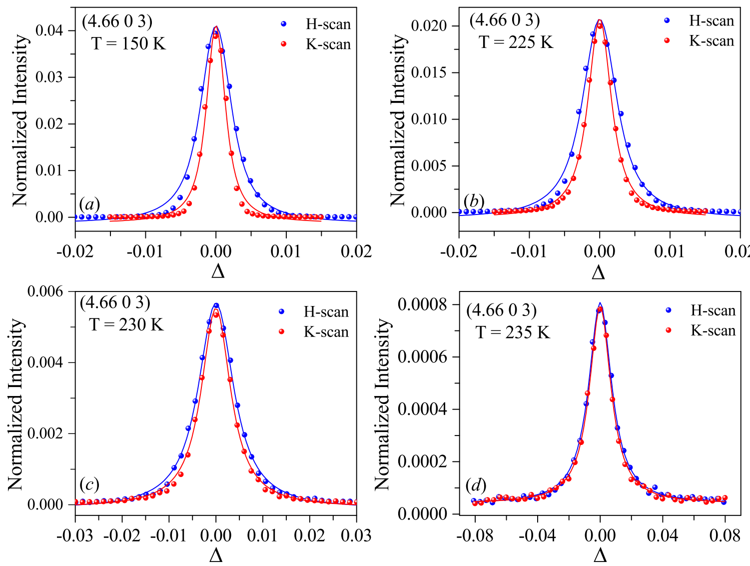

FIG. 1. (Color online) Comparison of the peak profiles along the H- and K-direction at different temperatures. In order to compare the peak profiles along the H- and K-direction, the central positions of the charge stripe reflection (4.66 0 3) are set to zero. (*a*) Below temperature T ∼218 K, the ratio of peak widths along H- and K-directions is almost constant. (*b*) and (*c*) Upon warming, the ratio changes as a function of temperature, (*d*) approaching 1 as the temperature approaches T_{CO} (∼238 K).

reflection was observed to persist at T = 230 K as shown in figure 3.

THERMAL HYSTERESIS

Experiments were also conducted to study thermal effects on the charge modulation. The measurement was done on a second crystal of $La_{5/3}Sr_{1/3}NiO_4$. As shown in figure 4, charge stripes show a hysteresis behavior around the transition boundary under different thermal treatments. This is in accordance with previously described thermal phenomena of an electron liquid crystal [2, 3]. The data shown in Figure 4 were collected during three sequences of warming and cooling. The sample was first cooled down to 130 K from room temperature in approximately 2 hours, and after the alignment at 130 K, the data (as marked by blue triangles in Figure 4) were collected by increasing temperature and scanning the charge stripe reflection (4.66 0 3) along the H, K, and L directions as a function of temperature until T= 250 K, where the reflection becomes very broad and weak. The sample was then warmed up to 260 K and kept at that temperature for approximately

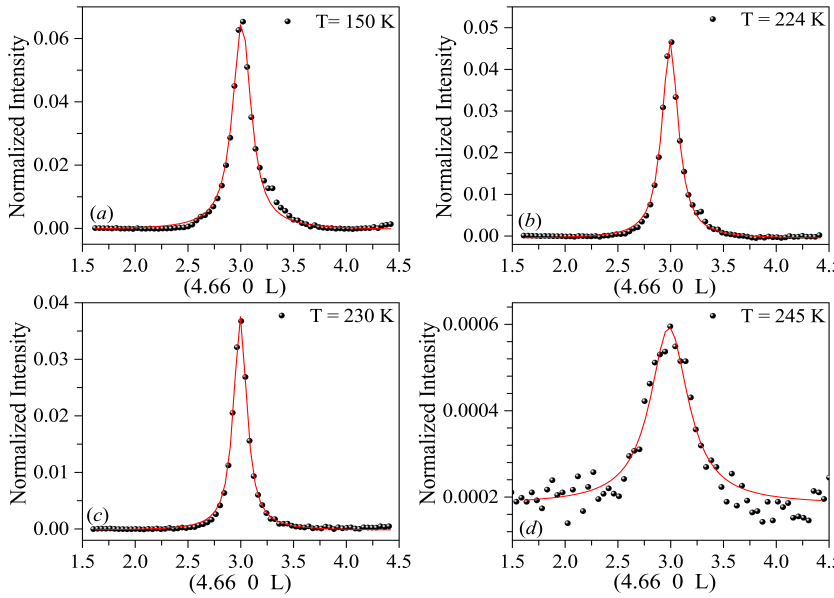

FIG. 2. (Color online) Peak profiles of a charge stripe reflection along the L-direction at different temperatures. Scans through the L-direction (c-axis) of the charge stripe reflection (4.66 0 3) at (a) 150 K, (b) 224 K , (c) 230 K, and (d) 245 K are shown. As temperature is lowered, the peak narrows and becomes sharpest at ∼230 K, but it then widens below 230 K, indicating an inverse order-disorder transition.

half an hour, after which measurements (marked by red dots) were taken as the sample was cooled to $T=$ 140 K. A third round of measurements (marked by open squares) were taken as the sample was warmed up to 250 K once more.

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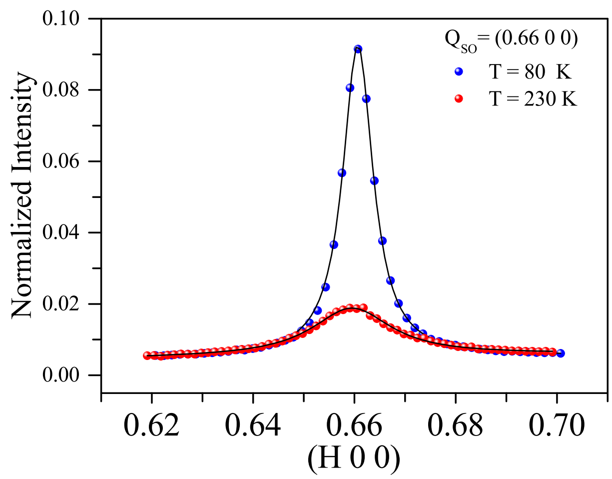

FIG. 3. (Color online) Spin stripe reflection (0.66 0 0) at T = 80 and 230 K. The data were collected by using resonant soft x-ray diffraction at Ni L_3 edge along the a-axis. The black lines are the best fits with a Lorentzian function.

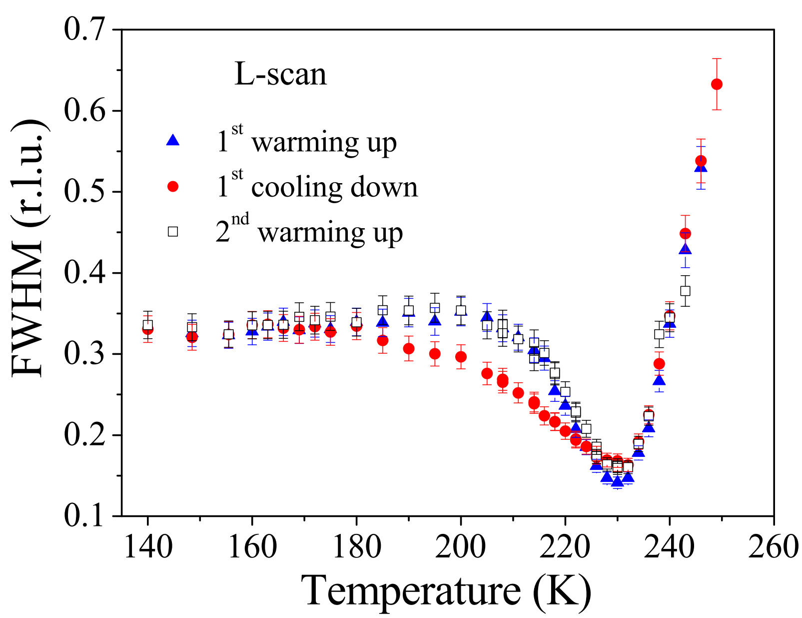

FIG. 4. (Color online) Thermal effects on the charge stripe modulations. Evolution of the peak width (FWHM) of the charge stripe reflection (4.66 0 3) along the L-direction is shown as a function of temperature for the different thermal processes (see the explanation in the supplementary method description). As can be seen, there is some thermal hysteresis.