

## Study of Charge Stripe Order and Oxygen Order in $\text{La}_2\text{NiO}_{4+\delta}$ With $\delta=2/15$ .

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**Introduction:**  $\text{La}_2\text{NiO}_4$  is a Mott-Hubbard insulator consisting of antiferromagnetic  $\text{NiO}_2$  planes alternating with  $\text{La}_2\text{O}_2$  layers. The  $\text{NiO}_2$  planes can be doped with holes; however, contrary to conventional expectations, the material remains nonmetallic up to quite large hole concentrations. There is now considerable evidence that the insulating behavior occurs because the dopant-induced holes tend to order themselves in periodically spaced stripes. These charge stripes run diagonally relative to the square lattice defined by the Ni-O-Ni bonds. In the essentially undoped regions between the stripes the Ni spins can order antiferromagnetically, with the charge stripes acting as antiphase domain walls. This behavior represents an excellent example of “topological” doping in a correlated insulator<sup>1</sup>.

In neutron scattering studies of a  $\delta=0.133$  crystal we observe a discontinuous jump in the intensities of both the magnetic and charge-order superlattice peaks on cooling below  $T_m = 110.5 \text{ K}^2$ . But we find that charge order clearly survives at  $T > T_m$  with a jump in the stripe spacing to a value commensurate with a second harmonic of the interstitial order, whereas the magnetic order disappears. Scattering at this position could come from either magnetic or charge order or both; however, the Q dependence of the structure factor indicated that it corresponds uniquely to charge order. The intensity of these  $\epsilon = 1/3$  peaks ( $Q = G \pm (2\epsilon, 0, 0)$ ; where G is a reciprocal-lattice vector corresponding to the average pseudo-tetragonal unit cell.) disappears at small Q, where the magnetic form factor is largest.

For  $T > T_m$ , the charge-order peak intensity decreases exponentially with temperature, so that no clear transition to the disordered state has been observed. This exponential variation of the intensity is analogous to a Debye-Waller-like behavior. Such a temperature dependence is in sharp contrast to the usual critical behavior associated with an order-disorder transition. The Debye-Waller-like decay of the intensity suggests that the charge correlations are fluctuating about an average ordered configuration determined by the ordered interstitial oxygens. It is possible that the charge order does not entirely disappear until the interstitials disorder. This coupling, together with the simultaneous appearance of interstitial order and charge-stripe order as a function of  $\delta$ , leads one to wonder whether the energy associated with the charge and spin correlations might influence the ordering of the interstitial oxygens.

**Methods and Materials:** A synchrotron scattering experiment can uniquely test the charge order scattering, which is due to lattice distortions and distinguish more clearly between charge and magnetic contributions. Furthermore, resonant scattering techniques at the Ni K-edge can give more information about the Ni valence distribution involved in the charge-stripe formation.

The single-crystal sample used in the present study was grown by radio-frequency skull melting. The oxygen concentration was selected by annealing at  $464^\circ\text{C}$  for 5 days in flowing  $\text{O}_2$  (1 bar) after careful preparation of the crystal surface followed by a quench to room temperature. The cooling and heating history for  $T < T_m$  has an influence on the relative fractions of coexisting magnetic phases, the width of the temperature regions where the magnetic wave vector is locked-in to rational fractions, and the detailed spectral weight of higher order magnetic satellites and their peak widths and possibly on the oxygen order. Therefore, the temperature dependence was studied during slow, careful warming after an initial cool down to 10 K.

**Results:** The temperature dependence of charge and oxygen order superlattice peaks has been studied from 10K up to 325K. At 10K and at 80K, where in the neutron scattering study charge- and spin structure are especially well ordered, energy scans of selected superlattice peaks at the Ni K-edge have been performed. The temperature dependence of the charge order superlattice peaks could be followed up to 308K, which is the highest temperature up to which charge order has been observed so far in nickelates and cuprates. The oxygen order disappeared at approximately 312K. Further experiments with improved signal to noise ratio will have to show, if charge order is connected to oxygen order or if it can also be observed above the oxygen ordering temperature. A detailed analysis of the experimental results is still on-going.

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**References:**<sup>1</sup> S. A. Kivelson and V. J. Emery, *Synth. Met.* **80**, 151 (1996); <sup>2</sup> P. Wochner, J. M. Tranquada, , D. J. Buttrey, and V. Sachan, *Phys. Rev. B* **57**, 1066 (1998).