Structural study of the $La_{2-x}Sr_xNiO_4$ phase diagram in the lower-doped regime

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Charge stripes have been predicted as a combined charge and spin-density wave phenomenon [1-3] before being experimentally observed in $La_{2-x}Sr_xNiO_4$ [4] and in the Nd codoped $La_{2-x}Sr_xCuO_4$ [5]. However, the role of this charge stripe instability for the superconducting pairing mechanism still remains a matter of debate [6,7]. Remarkable attention has been reattracted to the physics of stripe phases due to a recent discovery of a "hour-glass"-shaped magnetic spectrum in $La_{2-x}Sr_xCoO_4$ [8] similar to the observations in the superconducting cuprates [8,9]. Apart from the cobaltates [10] also the nickelates could be a useful reference system since a rather stable diagonal charge stripe order has been observed in these systems at higher hole-doping [4]. However, so far only the higher Sr-/hole-doped $La_{2-x}Sr_xNiO_4$ compounds have been studied in detail and no "hour-glass"dispersion has been found in these systems. The exact onset of stripe ordering is not known properly and has not been studied systematically (to the best of our knowledge). Therefore, we are interested in studying charge stripe phases in the nickelates $La_{2-x}Sr_xNiO_4$ systematically. In particular we were interested in the interplay of stripe ordering and structural distortions. Hence, we measured a whole series of $La_{2-x}Sr_xNiO_4$ samples with low Sr-doping at beamline P02 HRPD in order to study the symmetry and lattice parameters as a function of Sr-/hole-doping.

For this synchrotron radiation powder X-ray diffraction experiment a wavelength of 0.2076 Å could be used in combination with a resolution-optimized sample-to-detector distance that amounts to 1539.63 mm. For cooling a nitrogen flow Cryostream system was available having a temperature range of 81 K to 300 K.

One important value indicative for the structural distortions in $La_{2-x}Sr_xNiO_4$ (LSNO) is the orthorhombic splitting (a-b)/(a+b) that arises from the octahedral tilts after the structural transition I4/mmm → Bmab. We summarized our results in Fig. 1 for a couple of selected samples which may illustrate all important findings best. The undoped parent material exhibits the largest orthorhombic distortion. For this compound the transition temperature is even above room-temperature and could not be reached in this experiment. The size of the orthorhombic splitting as well as the onset temperature decreases with increasing hole-concentration *n*. For a 15%-doped sample the onset temperature is around room-temperature (right arrow in Fig. 1). Finally, for 20% and 21% hole-doped samples it is still possible to see an orthorhombic splitting around very roughly 150 K (left arrow in Fig. 1; the orthorhombic splitting starts increasing below ~150 K compared to the extrapolated dashed/dotted line in this figure). However, no orthorhombic splitting could be observed for a 22% doped sample within the resolution and temperature range of this experiment (see black data points in Fig. 1). This absence of any orthorhombic distortion might be already indicative for the border of the orthorhombic phase in the LSNO phase diagram and a first important result of this experiment.

Furthermore, we also studied the impact of Zn- and Ti- doping on the lattice parameters for samples with 15%-16% of mobile holes **n**. As can be seen in Fig. 1 both samples exhibit orthorhombic distortions. But the Ti-doped sample exhibits a kink around 120 K which is indicative for a stabilization of a LTLO/LTT phase in this system that might be rather interesting and subject to further investigations.

Complementary neutron and synchrotron experiments on single crystals are planned in order to study the stripe ordering in these nickelate compounds. The results found in this experiment will be essential for the interpretation of these experiments and also for the determination of the LSNO phase diagram. Now, further synchrotron radiation powder X-ray diffraction experiments in the still missing low temperature regime (T < 80 K) would be interesting in order to elucidate the structural properties also in this part of the phase diagram.

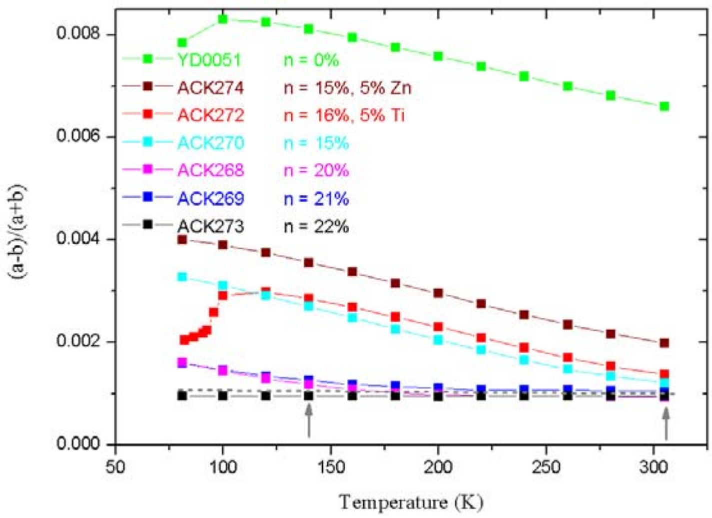

Figure 1: Orthorhombic splitting (a-b)/(a+b) of several selected LSNO samples.
(The dashed grey line is probably the detection limit in this experiment.)

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