



## Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.**

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

*<http://193.49.43.2:8080/smis/servlet/UserUtils?start>*

### ***Reports supporting requests for additional beam time***

Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

### ***Reports on experiments relating to long term projects***

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

### ***Published papers***

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

### **Deadlines for submission of Experimental Reports**

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

### **Instructions for preparing your Report**

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.



**Experiment title: X-Ray Scattering Studies of the overdoped Nickelates**

**Experiment number:**  
HE-1405

**Beamline:**  
BM28

**Date of experiment:**  
from: 02/10/2002 to: 10/10/2002

**Date of report:**  
25/02/2003

**Shifts:**  
24

**Local contact(s):**  
S. D. Brown

*Received at ESRF:*

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### **Report:**

In recent years the nickelate system  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  has been studied greatly due to it being isostructural with the high temperature superconducting cuprates. Our previous results have shown that the charge stripes are most intense, correlated and stable at the commensurate doping level of  $x = 1/3$  [1][2] where the spin stripes occur at the same position as the charge stripes and stabilise the charge ordering pattern. Decreasing the doping level away from  $x = 1/3$  results in the charge stripes weakening in intensity, becoming less correlated and the charge ordering temperature to decrease.

In this experiment we studied the doping levels  $x = 0.4$ ,  $x = 0.48$  and  $x = 0.5$  in order to investigate the behaviour of the charge stripes for doping levels of greater than  $1/3$  and to observe if the doping level of  $x = 0.5$  corresponded to another stable charge ordering regime. Recent neutron scattering measurements have demonstrated a rearrangement of the charge order from an incommensurate stripe pattern at low temperatures to a checkerboard charge order pattern similar to that in the half doped cobaltates [3] and manganites [4] which is stable to high temperatures  $\sim 480$  K. [5]

Measurements were made on beamline BM28 on the using an incident energy of 12.4 keV which is close to the peak flux of the beamline. A Ge (1,1,1) crystal was used as an analyser to provide high resolution triple axis measurements. The  $x = 0.4$  and  $x = 0.48$  samples were mounted in a displax cryostat and the  $x = 0.5$  in a displax cryofurnace that allowed a temperature range of 50 K – 800 K to be accessed. The crystals were mounted with the  $\langle 101 \rangle$  axis surface normal and all were of high quality with rocking curve widths of  $\sim 0.03^\circ$ . The samples were cooled to low temperature and a search was carried out around the (4, 0, 4) Bragg reflection. The charge stripes were located at a position of  $(2\varepsilon, 0, 1)$  around the (4, 0, 4) with  $\varepsilon$  increasing with doping level. Reciprocal space scans were carried out in the  $H$ ,  $K$  and  $L$  directions to determine the temperature dependence of the intensity, correlation length and position..

The charge stripes in all samples were 2-D in nature with them only being poorly correlated in the  $L$  direction. As the doping level was increased from  $x = 0.33$  the charge stripes became weaker, more diffuse with an accompanying decrease in the charge ordering temperature. The charge stripe ordering is weakest in the  $x = 0.5$  doped sample, however on heating a rearrangement of the charge ordering pattern was observed. At low temperatures the charge stripe ordering was observed at a

modulation of (0.88, 0, 1) which corresponds to  $\varepsilon = 0.44$  and the strongest peak was at (4,88, 0, 5) and there was no evidence of the checkerboard pattern at the expected position of (5, 0, 5). On

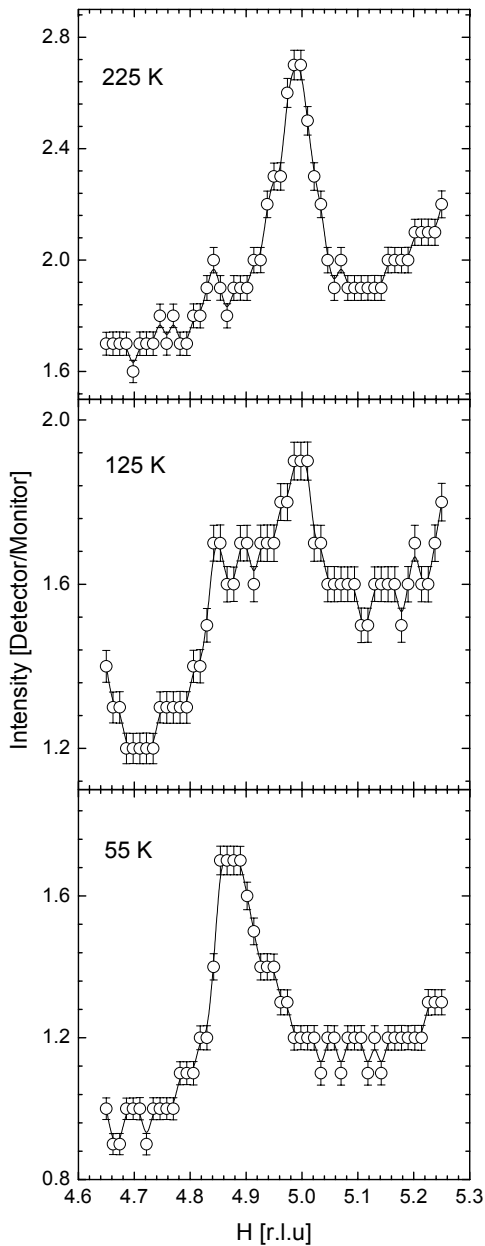


Fig. 1

where the charge stripe order is stabilised by the spin ordering. Deviation away from this doping level results in the charge stripes weakening and becoming less correlated. As the doping level of  $x = \frac{1}{2}$  is reached a new regime is entered where the system orders into a checkerboard pattern at high temperatures when the stripe order is no longer stabilised by the spin ordering. This work is being prepared for a publication [6] and forms the basis of an invited lecture at the CMMP 03 international conference [7]

heating the charge stripe order decreased in intensity and at 125 K a second signal was observed at a modulation of (5, 0, 5) corresponding to the checkerboard charge order. At 150 K only the checkerboard charge order was observed and this increased in intensity until reaching a maxima at 225 K. This is illustrated in Figure 1 which shows scans in the  $H$  direction carried out at 55 K, 125 K and 225 K. The checkerboard pattern was thermally very stable and was observed to 440 K despite being weak and poorly correlated. There was no difference within error in inverse correlation length of the stripe and checkerboard charge ordering indicating that there is no change from a less correlated state to a more correlated state. We postulate that the reason for this arrangement is due to the effects of the spin ordering. If the charge alone is considered the most stable configuration is the checkerboard pattern. However, at low temperatures the stripe order state is stabilised by the spin exchange interactions. The introduction of a discommensuration into the checkerboard pattern increases the types of spin interactions present. The checkerboard pattern only contains interactions between the  $\text{Ni}^{2+}$  and  $\text{Ni}^{3+}$  ions but with the introduction of a discommensuration introduces exchange interactions between  $\text{Ni}^{2+}$ - $\text{Ni}^{2+}$  and  $\text{Ni}^{2+}$ -hole- $\text{Ni}^{2+}$  which are much stronger and stabilise the stripe order at low temperatures. Above 80 K the spin order disappears and the stripe order is no longer stabilised and it begins to collapse. The holes re-order into the checkerboard pattern which is the most stable configuration for the charge ordering. The absence of the checkerboard order in the  $x = 0.48$  sample indicates that the checkerboard order only exists in a narrow window around the  $x = 0.5$  doping level.

This experiment has shown that the most stable doping level for charge stripe ordering is the  $x = 1/3$  doped sample

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- [5] R. Kajimoto, K. Ishizaka, H. Yoshizawa, Y. Tokura, Phys. Rev. B **67**, 014511 (2003)
- [6] P.D. Spencer, S.B. Wilkins, T.A.W. Beale *et al.* Phys. Rev. B in preparation
- [7] P.D. Hatton. 'Charge stripes in Nickelates' symposium of crystallography of charge, orbital and magnetic ordering. CMMP 2003 Belfast