ESRF	**Experiment title:** **Charge Ordering and Structural Distortions at Low Temperature in $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with x = 0.475 and 0.5**	**Experiment number**: 28-01-141
Beamline: BM28	**Date of experiment**: from: 12/6/02 to: 18/6/02	**Date of report**: 30/9/02
Shifts: 17	**Local contact(s)**: Dr. S.D. Brown	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

S.B. Wilkins*, P.D. Spencer*, and P.D. Hatton*

Department of Physics, University of Durham, South Road, Durham, DH1 3LE, UK

P. Prabhakaran and A.T. Boothroyd

Department of Physics, University of Oxford, Oxford, OX1 3PU. UK

Report:

This work has been submitted to the Physical Review B in the format attached.

Charge Ordering and Structural Distortions at Low Temperature in $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with $x = 0.475$ and 0.5

S. B. Wilkins, P. D. Spencer, T. A. W. Beale, and P. D. Hatton*
Department of Physics, University of Durham, Rochester Building, South Road, Durham, DH1 3LE, UK

M. von Zimmermann
HASYLAB at DESY, Notkestr. 85, D-22603 HAMBURG, Germany

S. D. Brown
XMaS CRG, European Synchrotron Radiation Facility, 38043 Grenoble Cedex, France

D. Prabhakaran and A. T. Boothroyd
Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford, OX1 3PU, UK
(Dated: September 30, 2002)

In this paper we present X-ray scattering results of charge and orbital ordering in the bi-layer manganite $La_{2-2x}Sr_{1+2x}Mn_2O_7$ with doping levels $x = 0.5$ and $x = 0.475$. Using high energy X-ray scattering the structural modulation due to the Jahn-Teller ordering and the charge ordering due to the Mn^{3+} / Mn^{4+} pattern have been measured. Both the $x = 0.5$ and $x = 0.475$ samples are found to display charge and Jahn - Teller order. We have confirmed that the wavevectors of the Jahn - Teller order, charge order and orbital order are $\vec{Q} = (0.5, 0.5, 0)$, $\vec{Q} = (0.25, 0.25, 0)$ and $\vec{Q} = (0.25, 0.25, 0)$. The origin of these has been confirmed by resonant X-ray scattering in the vicinity of the Mn K edge using polarization analysis. Contrary to previous studies the Jahn - Teller order is found to be not re-entrant but to reduce in intensity at temperatures below 140 K for both samples. Charge ordering was also detected in the $x = 0.5$ sample below this temperature.

PACS numbers: 61.10.Nz, 71.27.+a, 71.30.+h, 75.30.Vn

I. INTRODUCTION

The hole doped $3d$ transition metal oxides have been studied extensively over the past few years due to their remarkable physical properties; the most interesting being the high temperature superconductivity displayed by the cuprates[1] and the colossal magnetoresistance (CMR) shown by the manganites[2]. The three dimensional half-doped perovskite manganite systems have been studied greatly in order to investigate the relationship between the charge, spin, structural and orbital degrees of freedom and the electronic and magnetic properties shown in an effort to achieve an understanding of the mechanism associated with the CMR behavior. Neutron, electron and X-ray diffraction techniques have allowed the behavior of the charge, spin and structural properties to be investigated thoroughly, and recently it has also been possible to investigate the behavior of the orbital degree of freedom using resonant X-ray scattering technique at third generation synchrotron sources.[3,4] However, more work is needed to understand the relationship of orbital ordering with the other degrees of freedom in the system and the electronic and magnetic properties.

The magnetoresistive properties are very sensitive to the dimensionality of the manganese oxide, lattice and by inserting rock salt layers between manganese oxide sheets it is possible to greatly vary the electronic and magnetic properties of the system. This group of compounds is known as the Ruddlesden-Popper series and they have the general formula $(La, Sr)_{n+1}Mn_nO_{3n+1}$ where n is an integer. The $n = 1$ single layered perovskite $La_{0.5}Sr_{1.05}MnO_4$ shows no metallic or ferromagnetic state[5] while the $n = \infty$, which corresponds to the cubic perovskite structure, shows the CMR effect, although a high magnetic field is required to produce it[6]. By varying the value of n the properties of the system are dramatically changed and they should eventually approach those of the cubic system as n is increased towards infinity.

In this paper we report on the bi-layer system which corresponds to the $n = 2$ member of the Ruddlesden-Popper series and has the general formula $La_{2-2x}Sr_{1+2x}Mn_2O_7$. The basic structure in this series is an alternate stacking of MnO_2 layers alternated with rock salt $(La, Sr)_2O_2$ layers and the stacking occurs along the c-axis and the unit cell is shown in Figure 1.

The bi-layer system has the $I4/mmm$ tetragonal space group with lattice parameters with $a \simeq b = 3.87$ Å and $c \approx 20$ Å. The presence of the rock salt blocker layer has the effect of reducing the dimensionality, and in the $x = 0.4$ doped system there exists a paramagnetic to ferromagnetic transition at 126 K with a huge accompanying change in the CMR[7]. The CMR effect observed was significantly larger than seen previously in the 3-D system and required a smaller magnetic field to produce this effect; at 1 Tesla the CMR effect is ~ 20 times larger in the bi-layer compared to that of the three dimensional system. However, this larger effect is at the cost of a lower transition temperature compared to the 3-D system where the transition occurs at 350 K.