

LATTICE DISTORTIONS RELATED TO CHARGE ORDERING IN $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ M. Braden¹, J. Etrillard², A. Gukasov³, U. Ammerahl⁴, A. Revcolevschi⁴¹II. Physikalisches Institut, Universität zu Köln, Zùlpicher Str. 77, D-50937 Köln, Germany²Groupe Matière Condensée et Matériaux, UMR au CNRS 6626, Université de Rennes I, 35042 Rennes Cedex, France³Laboratoire Léon Brillouin (CEA-CNRS), CEA- Saclay, 91191 Gif sur Yvette Cedex⁴Laboratoire de Physico-Chimie de l'Etat Solide, Université Paris Sud, 91405 Orsay Cedex, France

The spin-ladders compounds with formula $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ or related exhibit a rather complex crystallographic structure due to their composite character. The materials consist of an alternating stacking of two distinct types of layers. One is formed by $\text{Sr}_2\text{Cu}_2\text{O}_3$ with a ladder-like structure and the other contains CuO_2 chains. The incommensurate modulation results from the misfit between the c-lattice-parameters of these planar elements. Whereas the a-parameter, $a \sim 11.47 \text{ \AA}$, is the same in both sub-lattices due to the free arrangement of the two CuO_2 -chains, the c-parameters reflect the different Cu-O-Cu bonding, 180 and 90° in ladders and chains respectively. The b-parameter corresponding to the stacking is the same for both elements. We have studied the crystal structure of this composite material by neutron diffraction focusing on the charge ordering transition occurring near 250K [1,2].

We have first analysed the temperature dependence of the mean crystal structure in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ by collecting large sets of Bragg reflection intensities on a four circle diffractometer; a large number of low order satellite reflections arising from the composite structure were included [1]. The crystal structure remains essentially unchanged between room and low temperature; in particular, the strongest satellite reflections show only little temperature dependence indicating that the main modulation of the composite is not changing with temperature. However the Debye-Waller factors and some weak fundamental reflections depend sensitively on temperature. The former may be attributed to a low lying phonon branch associated with the sliding of the chains against the ladders.

There has been a controversy in the literature about the onset of charge ordering in $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$, Cox et al. [3] reported a quadrupling of the chain lattice at low temperature, whereas Fukuda et al. [4] found superstructure reflection which are indexed by a modulation vector of (0.2,0,0) within the chain sublattice. However, van Smaalen argued that these reflections do not indicate charge ordering but should be interpreted as high order satellite reflections of the composite structure [5].

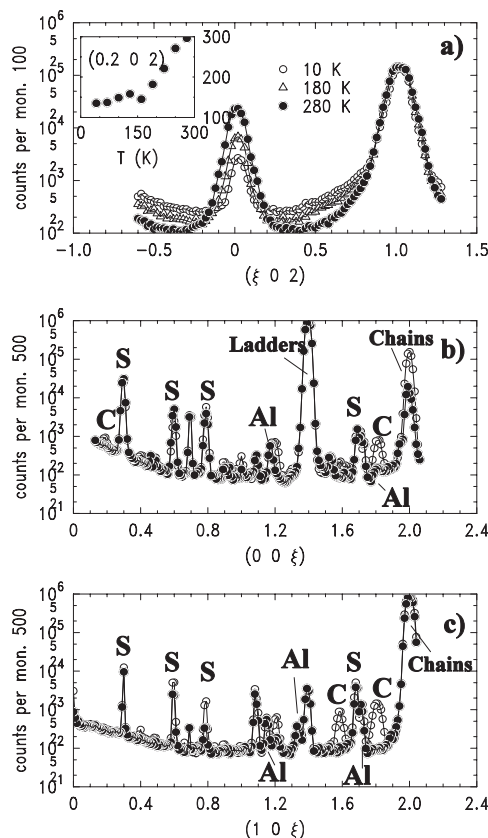


Figure 1. a) Scans along the a^* -direction across the chain lattice Bragg peaks (1 0 2 0) and (0 0 2 0), note that the latter is weaker and strongly temperature dependent. The inset shows the height of the diffuse scattering at (0.2 0 2) as function of temperature. b) Scans along the c^* -direction across the ladder reflection (0 0 0 2)=(0 0 1.395) and the chain reflection (0 0 2 0)=(0 0 2). c) Scans along the c -direction across (1 0 2 0)=(1 0 2). S denotes pure satellite reflections, AI a contamination by the Aluminum sample holder and C a low temperature superstructure peak; $\lambda/2$ denotes a weak contamination through neutrons of half wavelength [2].

We have studied the occurrence of additional reflections on the 4F triple axis spectrometer, which offers a high dynamical range and Q-resolution. The main results are shown in the figures 1 and 2.

Whereas the low order satellites change little in intensity upon cooling, we find additional reflections at low temperature, which are typically an order of magnitude smaller than the strongest satellites. In contrast to the results by Fukuda et al. [4] these reflections completely disappear at high temperature. Also the position does not perfectly match with the previous study. We may index most of the additional reflections by a modulation $(0,0,0.190(4))$ in respect with the chain lattice. However, we find that not only the chain-system but also the ladders are modulated with this modulation. Some weaker additional peaks are observed near but not precisely at the positions where second order reflections are expected. Due to the high resolution of the 4F-spectrometer the observed differences are clearly significant.

The new reflections appear close to the temperature where the transport properties exhibit strong anomalies. One should connect them hence with the onset of charge ordering. However, the totality of the data cannot be explained by a single modulation on the chain subsystem.

We propose an interpretation intermediate between references [4] and [5]. We argue that the reflections, we find to appear at low-temperature, are originating from the charge ordering. But charge ordering does not seem to induce its proper modulation instead the charge order induced modulation seems to fall into the composite modulations existing anyway. All the reflections, we observe, can be indexed as high order satellites of the composite lattice. But in an ordinary modulated system one may not understand why we find so many superstructure reflections corresponding to the special value of $q=(0,0,0.19)$. The modulation of the charge ordering seems to lock into the modulation of the composite

structure, somehow similar to the typical lock-in transitions of structural phase transitions on commensurate values.

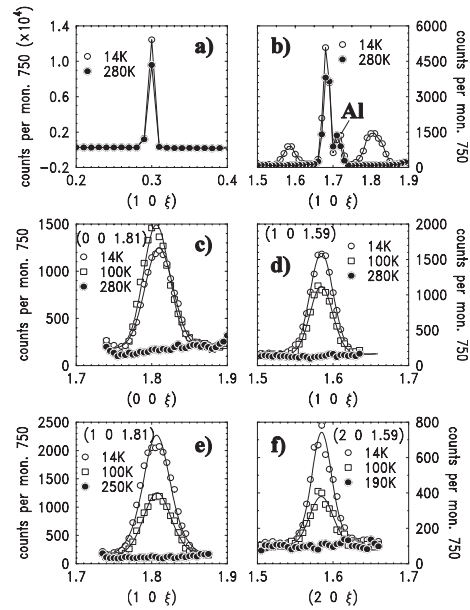


Figure 2. Scans across satellite and the additional peaks demonstrating their different temperature dependencies and peak widths. a) Scan across the satellite $(1\ 0\ 0.3)=(1\ 0\ 1\ -1)$ at 14 and 280K. b) Scan across the satellite $(1\ 0\ 1.69)=(0\ 0\ 1\ 1)$ at 14 and 280 K; near $(1\ 0\ 1.59)$ and $(1\ 0\ 1.81)$ new superstructure peaks appear at low temperature which exhibit significant broadening, **Al** indicates an Al-contamination. c)-d) Scans across the low temperature reflections $(0\ 0\ 1.81)$, $(1\ 0\ 1.59)$, $(1\ 0\ 1.81)$ and $(2\ 0\ 1.59)$ at low and high temperatures. Lines denote fits with a Lorentzian peak profile convoluted with the resolution of the triple axis spectrometer.

References

- [1] J. Etrillard, M. Braden, A. Gukasov, U. Ammerahl, and A. Revcolevschi; *Physica C* **403**, 290-296 (2004).
- [2] M. Braden, J. Etrillard, A. Gukasov, U. Ammerahl, and A. Revcolevschi, *Phys. Rev.* **B69**, 214426 (2004).
- [3] D.E. Cox, T. Iglesias, K. Hirota, G. Shirane, M. Matsuda, N. Motoyama, H. Eisaki, and S. Uchida, *Phys. Rev.* **B57**, 10750 (1998).
- [4] T. Fukuda, J. Mizuki, and M. Matsuda, *Phys. Rev.* **B66**, 012104 (2002).
- [5] S. van Smaalen, *Phys. Rev. B* **67**, 026101 (2003).