EUROPEAN SYNCHROTRON RADIATION FACILITY
INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report

	Experiment title: Intrisic disorder in Sr doped Han Purple	**Experiment number**: 01-02-1081
Beamline: BM01	**Date of experiment**: from: 01/07/2015 to: 07/07/2015	**Date of report**: 28/08/2015
Shifts: 18	**Local contact(s)**: Dmitry Chernyshov	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Björn Wehinger*, University of Geneva and Paul Scherrer Institute, Switzerland

Natalija van Well*, Paul Scherrer Institute, Switzerland

Skirmante Butkute*, Paul Scherrer Institute, Switzerland

Mariusz Kubus*, University of Bern and Paul Scherrer Institute, Switzerland

Christian Rüegg, Paul Scherrer Institute and University of Geneva, Switzerland

Report:

The investigation of Han purple $BaCuSi_2O_6$ and $Ba_{1-x}Sr_xCuSi_2O_6$ single-crystal samples using synchrotron x-ray diffraction and diffuse scattering was performed in order to disentangle the effects of exchange and structural distortions on the microscopic Hamiltonian.

High quality diffuse and single crystal diffraction data were recorded at 4.3K, at 300K, in more detail in the temperature range of 80-120K and at a few representative temperatures between 300 and 800K. The use of the newly build helium cryostream at the lowest temperature, a nitrogen cryostream at intermediate temperatures and a heat blower at high temperatures allowed for a free rotation of the crystals.

Representative reciprocal space reconstructions of scattering intensities in the (*0kl*) plane of $BaCuSi_2O_6$ and $Ba_{0.9}Sr_{0.1}CuSi_2O_6$ at 4.3K are shown in Figure 1 and 2, respectively.

For the first time, we were able to obtain a 3D picture of the incommensurate modulations which appear at low temperature. The symmetry of the incommensurate structure is more complex than expected. A follow-up experiment with higher momentum resolution is required to resolve its structure. The incommensurate reflections are in the order of 10^4 times weaker than the strongest Bragg peaks.

The structural investigation revealed the absence of a previously reported symmetry lowering phase transition at 100K in $BaCuSi_2O_6$ crystals grown with oxygen partial pressure and in $Ba_{0.9}Sr_{0.1}CuSi_2O_6$. The incommensureate modulation, which appear in the pure compound, is not present in the Sr doped sample, see Figure 2.
Both compounds show a high temperature phase above 640K.

Diffuse scattering was recorded at 4.3K, 300K and detailed in the range of 80-120K. The acquisitions covered an angular range of 360 degrees with 0.1 degrees angular step. This data allows for high quality 3D reciprocal space constructions. The analysis is currently ongoing. A detailed shape analysis will allow us to determine the in-plane modulation in the pure compound and the intrinsic disorder induced by doping.

The suppressed symmetry-lowering structural phase transition may allow studies of the generic physics of interacting two-dimensional triplon Bose-Einstein condensates for the first time. The obtained results will enable studying the spin dynamics in this model magnetic compound of high current interest. The incommensurate modulations may also be suppressed by application of external pressure. A single crystal diffraction experiment in a diamond anvil cell at low temperatures will open the possibility to access the quantum critical point without the need of structural disorder induced by Sr doping.

In summary, we could show that our high quality single crystals grown with oxygen partial pressure do not undergo a symmetry-lowering phase transition at 100K but show incommensurate modulations at 4.3 K. The incommensurate structure was resolved in 3D reciprocal space. The Sr doped sample show significant diffuse scattering due to intrinsic disorder. Both, the symmetry-lowering transition at 100K and the incommensurate modulations at 4.3 K are absent.

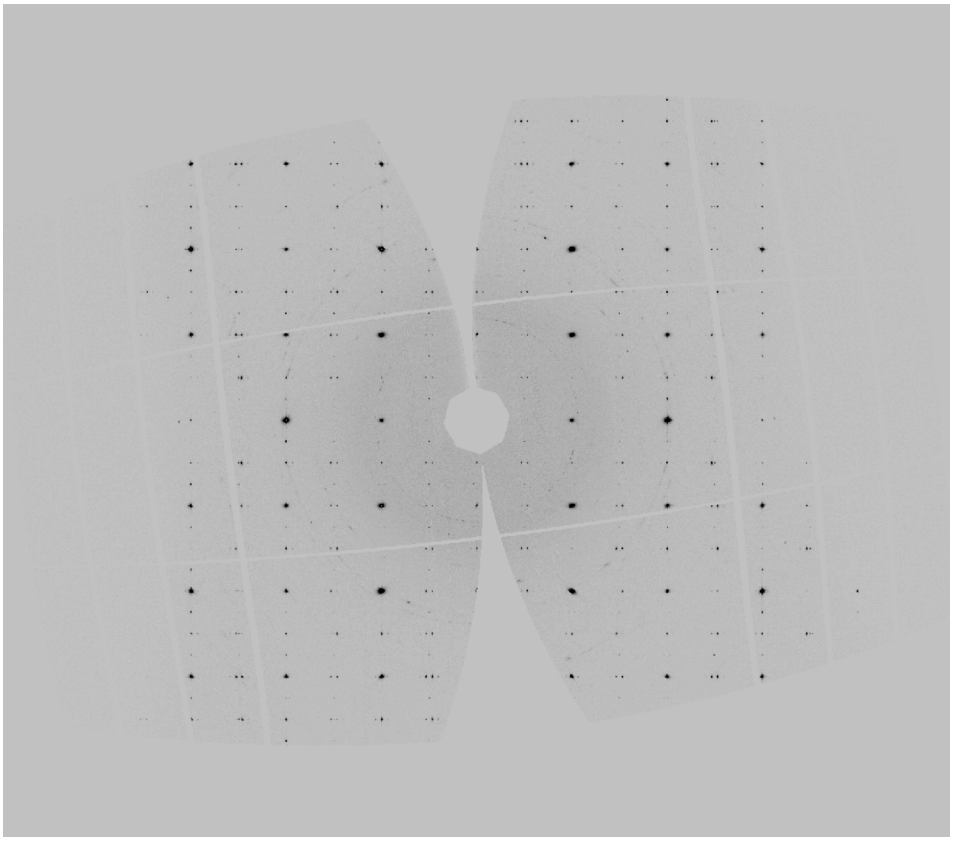

Figure 1. Reconstructed scattering intensities in (*0kl*) reciprocal space plane of $BaCuSi_2O_6$ at 4.3K.

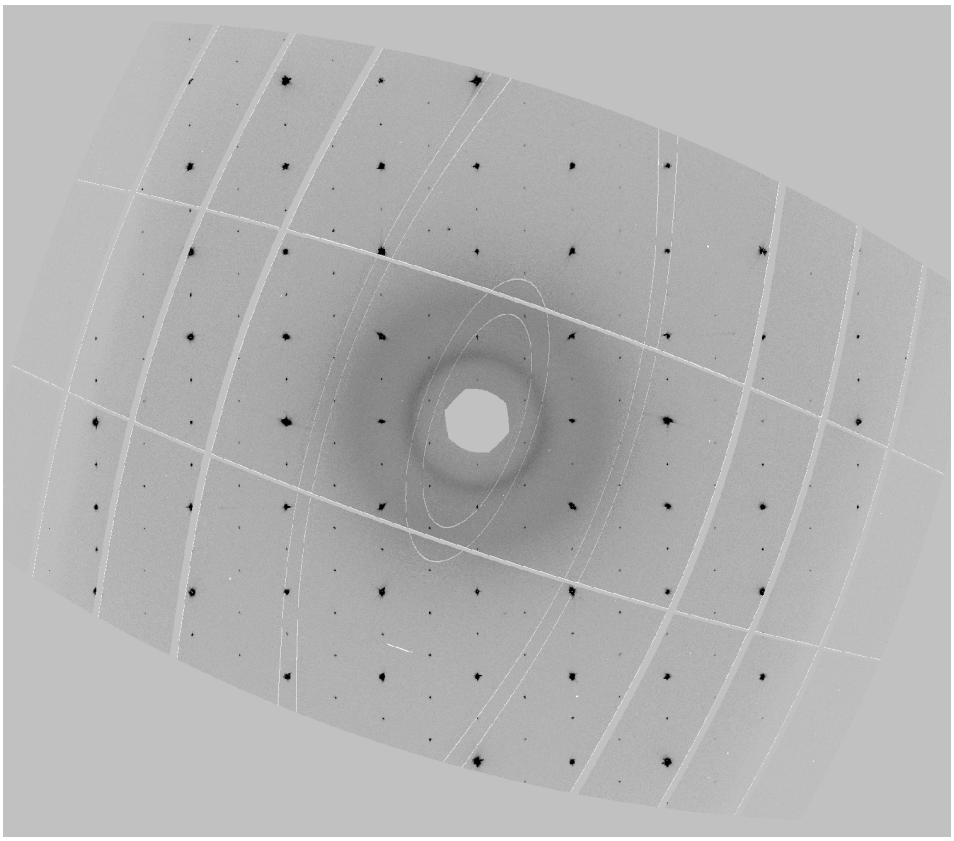

Figure 2. Reconstructed scattering intensities in (*0kl*) reciprocal space plane of $Ba_{0.9}Sr_{0.1}CuSi_2O_6$ at 4.3K.