Experimental Report

12/09/2014

Proposal: **7-02-144** **Council:** 10/2012

Title: Studies of polar nanoregions in (Sr,Ba)Nb2O6 single crystal

This proposal is continuation of: 7-02-128

Researh Area: Physics

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Samples: Sr0.61Ba0.39Nb2O6

Instrument	Req. Days	All. Days	From	To
IN16	8	9	22/05/2013	31/05/2013
IN3	0	2	21/05/2013	23/05/2013

Abstract:

We intend to continue the experiment No. 7-02-128 on IN16B backscattering spectrometer using the new option - fixed window measurements - and characterize polar dynamics of tetragonal Sr0.61Ba0.39Nb2O6 (ferroelectric relaxor) single crystal via studies of diffuse scattering. This proposed study could help better understand polar nanoregions dynamics of (Sr,Ba)Nb2O6 and of relaxor perovskites.

Studies of polar nanoregions in (Sr,Ba)Nb_2O_6 single crystal

The experiment 7-02-144 was performed on the IN3 three-axis spectrometer, IN16, and IN16B backscattering instruments on May 21-31, 2013. The aim of this experiment was to characterize the nanoscale polarization dynamics of tetragonal $Sr_{0.61}Ba_{0.39}Nb_2O_6$ (SBN-61) single crystal via studies of its diffuse scattering in the (001) Brillouin zone. We intended to use the fixed window method (scans at a given energy transfer) since the statistics had been too poor with the usual approach, i.e. continuous measurement of the whole spectrum.

We have investigated a cylindrical SBN-61 single crystal (volume of about 3 cm^3) grown by the Czochralski method. First of all, the crystal was put inside a Nb holder and its crystallographic orientation was rechecked on IN3. Then, the sample was placed in a standard orange cryofurnace and measured in the (*h*0*l*) scattering plane on IN16. Since the setup for fixed window measurements was moved to the newly commissioned IN16B backscattering instrument, the IN16 instrument was operated in its standard configuration: an unpolished Si(111) Doppler monochromator, unpolished Si(111) analysers providing instrumental energy resolution of 0.8 μeV at scattered neutron wavevector of $k_f = 1$ Å^{-1} and operating with a dynamic range of ±4 μeV. Most of the measurements were carried out at momentum transfers close to the 001 Brillouin zone center in three stages:

1. In the static operation mode, small elastic maps around the 001 Bragg reflection were measured from 300 to 480 K across the phase transition (T_C ~ 350 K). The point of this stage was to find the best momentum transfer **Q** at which the signal has less contamination from the Bragg reflection and/or domain wall scattering and shows a peak in diffuse scattering close to the phase transition.
2. In the dynamic operation mode, the whole spectra from –4 to 4 μeV at momentum transfer **Q** ~ (0.15,0,1) were measured at several temperatures in the temperature range 290 – 480 K. In order to get data with sufficient statistics the acquisition times were 20 –35 hours per spectrum.
3. Measurement of the IN16 energy resolution on a hollow cylindrical vanadium sample with a radius of 0.5 cm.

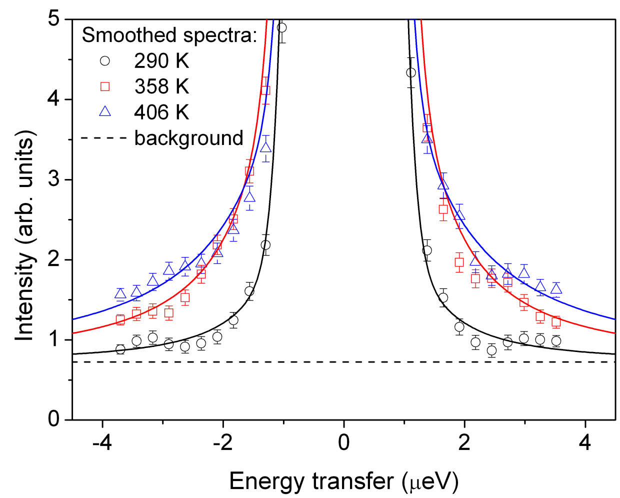

Fig.1. Inelastic neutron spectra at **Q** ~ (0.15,0,1), smoothed with a 0.8 μeV wide sliding window. Solid lines are calculated spectra from convolution of the resolution function of IN16 with the imaginary part of dielectric function multiplied by Bose-Eintstein factor [1,2]. The solid line at 290 K corresponds well to the vanadium resolution spectrum that was not displayed for clarity.

Finally, some data have been taken while having an opportunity of a one-day test on the IN16B backscattering spectrometer, operating in a similar configuration as IN16 with an energy resolution of 1 μeV. Here we were able to draw benefits from the increased flexibility of the new instrument, permitting to carry out directly momentum transfer and temperature scans at a given energy transfer window. Nevertheless, the stage 1 of collecting elastic data as on IN16 was repeated so as to find the best momentum transfer **Q** ~ (0.15,0,1). Then, we measured temperature scans at energy transfers of 0, 3, and 8 μeV.

Despite not having possibility of using fixed window measurement on IN16, we have succeeded to detect inelastic scattering, which is apparent from Fig.1: the spectra at 358 K and 406 K are visibly broader than the spectrum at 290 K which is resolution limited. Note that the spectra in Fig.1 were smoothed with a 0.8 μeV wide sliding window in order to reduce the spread of data. The reason why we were able to detect inelastic scattering in comparison with the former experiment is that the sample was 8 times bigger than the former one, acquisition times were increased 2–3 times, and the range of measured energy transfers was reduced 4 times. So, this resulted in an increase in intensity of about 1–2 orders of magnitude.

To compare data collected from IN16B with those from IN16, we have plotted temperature dependence of intensity obtained from the smoothed spectra in Fig.1. The results are shown in Fig.2a, showing shifting of peak position at energy transfers to higher temperatures. This shift occurs systematically in both IN16 and IN16B data and seems to correspond well to the Vogel-Fulcher law determined from dielectric measurements [1], which will be analyzed in details. Moreover, we have compared the 001 Bragg intensities and found that IN16B gives about one order of magnitude bigger intensity than IN16.

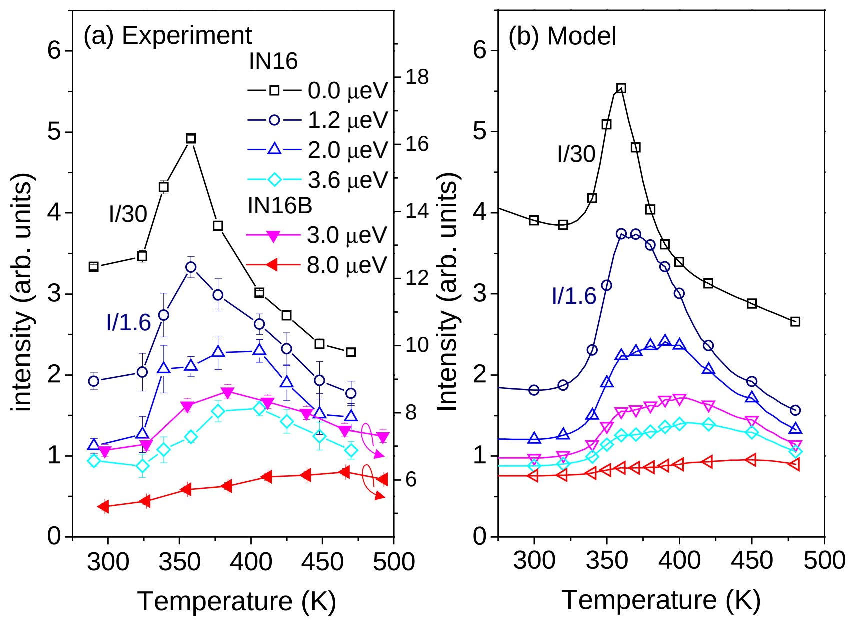

Fig.2. Scattering intensity obtained (a) from experimental spectra measured on IN16 and fixed-window measurements on IN16B at **Q** ~ (0.15,0,1) and (b) from the same model as in Fig.1. Both the experimental and model spectra were smoothed as in Fig.1.

In conclusion, fixed window regime seems to be extremely advantageous for this type of dynamics in relaxor ferroelectric. We have found that diffuse scattering in the (001) Brillouin zone corresponds to polar fluctuations seen by dielectric spectroscopy [1,2]. This is a very important result in relaxor physics since these measurements provide the missing link that allows to relate directly the correlation lengths of diffuse scattering coming from order-parameter fluctuations with the polarization fluctuations seen by dielectric spectroscopy.

References:

[1] E. Buixaderas, et al., *J. Phys.: Condens. Matter* **17**, 653 (2005).
[2] P. Ondrejkovic, et al., in preparation.