Experimental report

17/02/2020

Proposal: 7-02-185 **Council:** 4/2019

Title: Dynamics of nanoscale polar fluctuations in PMN

Research area: Physics

This proposal is a continuation of 7-02-154

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Samples: Pb(Mg1/3,Nb2/3)O3

Instrument	Requested days	Allocated days	From	To
IN3	1	1		
IN16B	7	4	24/01/2020	28/01/2020

Abstract:

We intend to do continuing study of a Vogel-Fulcher relaxation in the relaxor ferroelectric Pb(Mg1/3Nb2/3)O3 single crystal via investigation of its diffuse scattering. This study will give valuable information about time-space correlation lengths of polar fluctuations associated with exceptional dielectric properties of multiaxial relaxor perovskites. In this challenging study, we would like to take advantage of fixed window measurements on the new high-flux IN16B backscattering spectrometer and our experience with this type of experiment.

Dynamics of nanoscale polar fluctuations in PMN

The experiment 7-02-185 was performed on the IN16B backscattering instrument on January 24. – 28. 2020. The aim of this experiment was to study a Vogel-Fulcher relaxation in the relaxor ferroelectric $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) single crystal. This was done via investigation of its diffuse scattering in order to obtain valuable information about time-space correlation lengths of polar fluctuations associated with the Vogel-Fulcher relaxation.

The single crystal (volume of 2.2 cm^3) wrapped in an Al foil and fixed to an Al holder was firstly checked and thoroughly aligned on the IN3 three-axis spectrometer to have its (001) crystallographic plane in the scattering plane. Then, the sample was placed in a standard orange cryofurnace, whose body was masked by Cd bands apart from a 4.5-cm height stripe around equatorial scattering plane. The IN16B instrument was operated in its standard configuration: an unpolished Si(111) Doppler monochromator, unpolished Si(111) analyzers (neutron wavevector of k_f = 1 $Å^{-1}$), and PSD detectors. The instrumental energy resolution was about 0.8 μeV. Most of the measurements were carried out in the 010 Brillouin zone as follows:

1. Small elastic maps around the 010 Bragg reflection were measured at 0 μeV and 3 μeV from 200 to 450 K. The point of this stage was to find the best **Q** at which the diffuse scattering is free of contamination from the 100 Bragg reflection and shows a characteristic temperature change [1].
2. Then, the diffuse scattering at selected momentum transfer **Q** ~ (0.07,1.07,0) was measured at 0, 2, 3, 5, and 8 μeV energy transfers from 450 K to 10 K. The selected data are plotted in Fig. 1. The acquisition times were up to 80 min per point for the highest energy transfer.
3. In the dynamic operation mode, the spectra at the same momentum transfer **Q** ~ (0.07,1.07,0) were measured from -15 to 15 μeV at 10, 250, 290, and 330 K. The acquisition time was up to 4-10 h. Selected spectra are shown in Fig. 2.
4. Since we had obtained four out of seven requested experimental days on IN16B we did not have time to measure dispersion of the diffuse scattering along the [110] directions as intended in the proposal.

The diffuse scattering measured at 0 μeV shows a monotonic increase with decreasing temperature (see Fig. 1) in accord with a previous backscattering study by Stock et al. [1]. In addition, we found an inelastic component showing a peak, whose position corresponds well to dielectric data [2]. Moreover, the peak shifts to higher temperatures with increasing energy

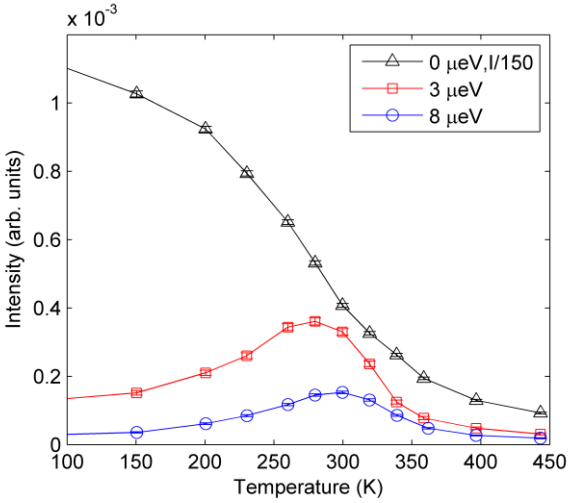

Fig. 1: Temperature dependence of diffuse scattering at **Q** ~ (0.07,1.07,0).

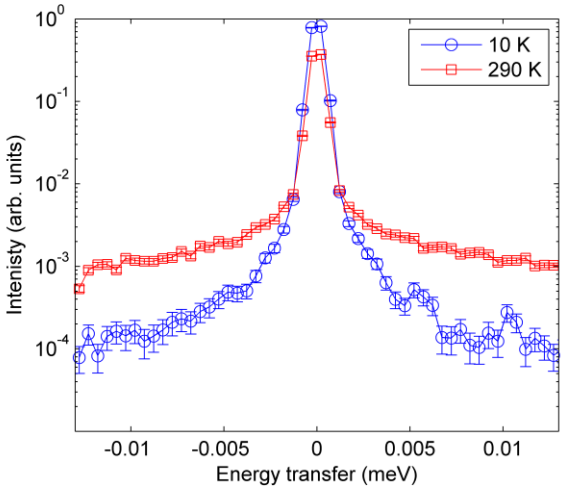

Fig. 2: Inelastic neutron spectra at **Q** ~ (0.07,1.07,0).

transfer which is a clear signature of the dielectric Vogel-Fulcher-like relaxation – polar fluctuations – observed in relaxor ferroelectrics [2,3]. The inelastic component is also seen from the spectrum at 290 K in Fig. 2.

In conclusion, we have found that diffuse scattering in the (010) Brillouin zone corresponds to polar fluctuations seen by dielectric spectroscopy [2] as we proved for the first time in the uniaxial relaxor ferroelectric $Sr_{0.61}Ba_{0.39}Nb_2O_6$ [3]. However, the dispersion of the relaxation remains to be clarified.

References:

[1] C. Stock, *et al.*, *Phys. Rev. B* **81**, 144127 (2010).
[2] V. Bovtun et al., *J. Eur. Ceram. Soc.* **26**, 2867 (2006).
[3] P. Ondrejkovic *et al.*, *Phys. Rev. Lett.* **113**, 167601 (2014).