Experimental report

08/02/2018

Proposal: TEST-2487 **Council:** 10/2014

Title: Low-energy lattice dynamics of therelaxox PMN

Research area:

This proposal is a new proposal

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Samples: Pb(Mg0.33Nb0.66)O3 (PMN)

Instrument	Requested days	Allocated days	From	To
THALES	7	7	07/07/2015	14/07/2015

Abstract:

Giant softening of elastic constants in relaxor ferroelectrics

The experiment no. TEST-2487 was undertaken on the newly commissioned ThALES cold three-axis (TAS) spectrometer and held on June 7-16, 2015. We obtained three extra days of allocated beamtime because of troubles with a velocity selector at the beginning of the experiment. The aim of this study was to investigate transverse low-frequency phonons of a morphotropic phase boundary $Pb(Zn_{1/3}Nb_{2/3})O_3$ with 8 % of $PbTiO_3$ (PZN-8%PT) relaxor ferroelectric single crystal in order to extract information about the elastic softening effect [1].

The spectrometer was equipped with Si(111) monochromator and Si(111) analyser and operated not only in its standard so-called 'w' configuration but also in thermal TAS configuration (so-called 'long chair'), which allowed us to vary the outgoing neutron wave-number on the analyser k_f from 1.4 to 2.8 $Å^{-1}$ and so to reach higher Brillouin zones. A PZN-8%PT single crystal was wrapped into an Al foil on an Al holder, placed in the standard cryofurnace, and measured in the (*hk*0) scattering plane.

'W' configuration

First, we measured in the standard configuration with k_f = 1.4 $Å^{-1}$ and energy resolution of 0.1 meV. Inelastic neutron scattering data were collected mainly in the (110) Brillouin zone which has a strong acoustic structure factor. Spectra were measured along the [1-10] direction with energy transfer up to 2.5 meV and temperatures of 300, 442, and 533 K. Fig.1 shows an example of constant-energy measurements at several energy transfers up to 2.4 meV. This strategy of constant-energy scans allowed us to trace the transverse acoustic (TA) phonon propagating along the [1-10] direction. Unexpectedly, we were not able to separate the TA phonon from diffuse (quasi)elastic scattering in a spectrum measured at **Q** = (1.02,0.98,0) r.l.u. (see Fig.2) as in our earlier results taken on IN12 cold TAS [2]. The transverse Q-resolution was 0.022 r.l.u. (0.035 $Å^{-1}$). We tried to optimize the spectrometer configuration (together with the Restrax program) but without notable success since collimators have not been commissioned on ThALES yet. For that reason we changed our plans and focused on measuring TA phonons at the standard q = 0.1 r.l.u. Since spectra measured at about **Q** = (1.1,0.9,0) in the "W" configuration had low counting rate (see Fig.2), we intended to measure phonons in higher zones (phonon intensity is proportional to $|\mathbf{Q}|^2$) with stronger

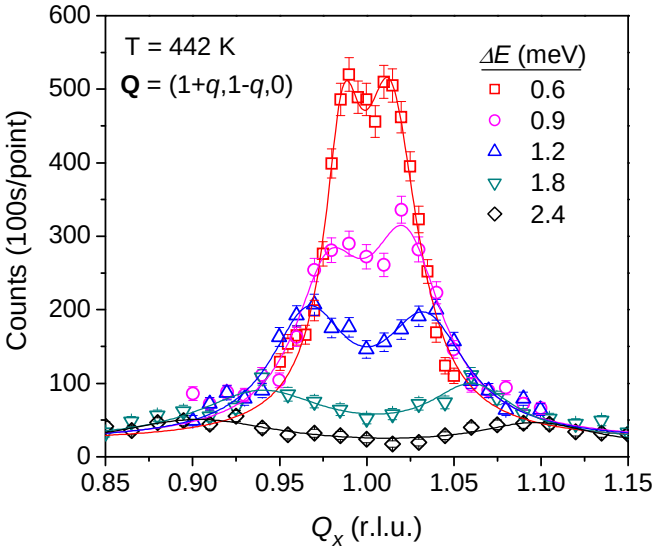

Fig 1. Constant-energy scans at **Q** = (1+*q*,1-*q*,0) and temperature of 442 K. The solid lines are guides for eye (k_f = 1.4 $Å^{-1}$, 'W' configuration).

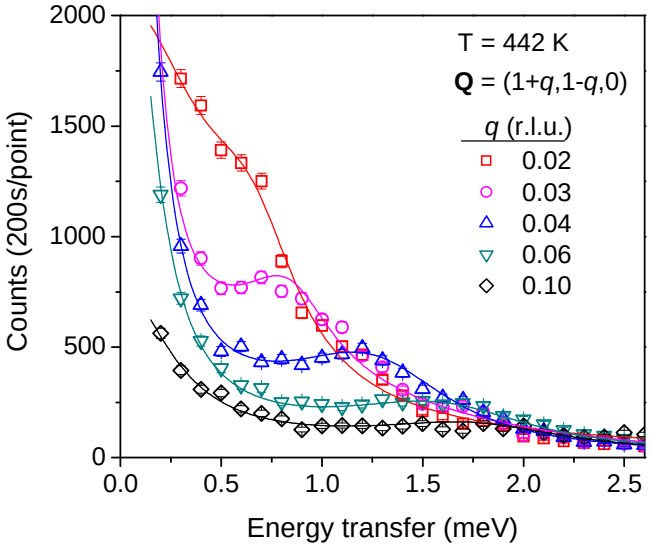

Fig 2. Neutron scattering spectra at **Q** = (1+*q*,1-*q*,0) and temperature of 442 K. The solid lines are guide for eye (k_f = 1.4 $Å^{-1}$, 'W' configuration).

acoustic structure factors. This required bigger k_f and so a change of the instrument configuration.

'Long-chair' configuration

ThALES was used for the first time in the "long-chair" configuration allowing to set k_f to 2 and 2.8 Å^{-1} and measure in the (200) and (220) Brillouin zones, respectively.

In the case of k_f = 2.8 Å^{-1}, the velocity selector was taken out because of its speed limit which would cause a dramatic decrease in neutron flux. We optimized energy resolution to 1.2 meV by setting the vertical analyser curvature to zero in spite of three times longer collecting time. The transverse and longitudinal Q-resolution was rather rough of about 0.045 r.l.u. (0.07 Å^{-1}) and 0.075 r.l.u. (0.12 Å^{-1}), respectively. Thus, this configuration was not appropriate for this study without possibility of using collimators. The only advantage was that the four-dimensional resolution ellipsoid had an elongated shape versus a fairly spherical ellipsoid of the "W" configuration. We measured spectra in the vicinity of **Q** = (2.1,1.9,0) at five temperatures from 300 to 533 K. They showed only minor changes with temperature.

Finally, most of the data were collected with k_f = 2 Å^{-1}, without vertical analyser focalization and the vertical slit between the velocity selector and monochromator reduced to 10. This setting provided us an energy resolution of 0.4 meV and a transverse Q-resolution of 0.035 r.l.u. (0.055 Å^{-1}). This k_f limited us to the (200) zone. We measured qausi-transverse spectra along the [1±10] direction at temperatures from 347 to 530 K across the diffuse phase transition where the most significant changes in the TA phonon frequency were expected according to resonant ultrasound spectroscopy (RUS) measurements [1]. An example of typical spectra at **Q** = (2.1,0.1,0) are shown in Fig.3. There is no significant change in the phonon frequency at first sight and therefore the temperature dependence of the TA branch is much smaller than in RUS measurements [1]. Nevertheless, the temperature-dependent (left-hand) wing and an asymmetrical shape of the mode peak indicate clear mode coupling which could partly explain the temperature dependence of the acoustic phonon.

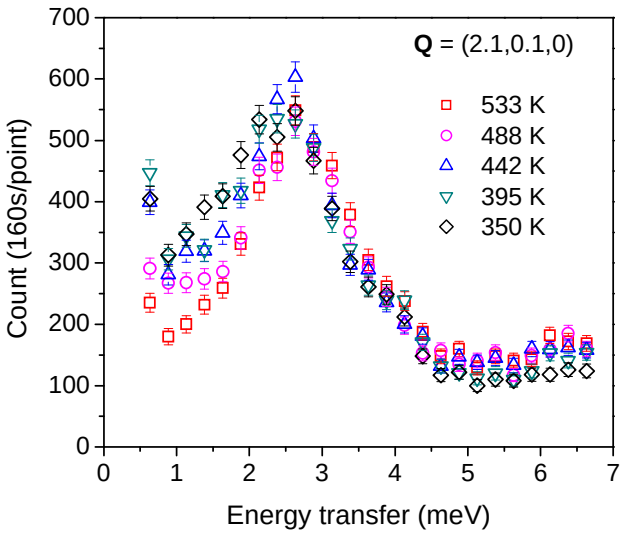

Fig 3. Neutron scattering spectrum at **Q** = (2.1,0.1,0) at several temperatures (k_f = 2 Å^{-1}, 'long-chair' configuration).

In conclusion, we have put in a lot of effort to optimize the spectrometer for the proposed study and testing new configurations. We were missing the possibility of using collimators for improving the Q resolution. The measured data are currently analysed in detail.

References:

[1] S. M. Farnsworth, E. H. Kisi, and M. A. Carpenter, *Phys. Rev. B* **84**, 174124 (2011).

[2] J. Hlinka, S. Kamba, J. Petzelt, J. Kulda, C. A. Randall, and S. J. Zhang, *J. Phys.: Condens. Matter* **15**, 4249 (2003).