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Soft mode dispersion and 'waterfall' phenomenon in relaxors revisited

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Abstract

Results of recent inelastic neutron scattering studies of lead-based relaxor ferroelectrics by Gvasaliya et al. [J. Phys.: Condens. Matter 17, 4343 (2005); J. Phys.: Condens. Matter 19, 016219 (2007)] have put in question the existence of the "waterfall" anomaly–an apparent vertical dispersion segment joining the TA and TO branches–observed earlier in low-energy [ξ00] phonon dispersion curves of these materials. In the present article, we review the results of earlier experiments and model calculations together with the outcome of our recent measurements on PMN using the same instrumental set-up as Gvasaliya et al. to conclude that the "waterfall" feature is not an experimental artefact. We also give some hints on a possible explanation of the results of Gvasaliya et al., by exploring the fact that the reported dispersion of the underdamped transverse optic branch follows the longitudinal acoustic (LA) branch dispersion surprisingly closely.

Keywords:

relaxor ferroelectrics | waterfall phenomenon | phonon dispersions | inelastic neutron scattering

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Notes

Notes

1. In the rest of this article, we use reduced (dimensionless) phonon wave vectors expressed in reciprocal lattice units (r.l.u.) given by $c^* = 2\pi/c$, (e.g. for PMN, $c = 4.04$ Å, i.e. $c^* = 2\pi/c = 1.555$ Å^{-1}).

2. First two hypotheses are close to those invoked in Ref. 17.

3. Apart from the obvious misprint–extra factor of 2 in arguments of all trignometric functions introduced in Ref. 14.

4. This additional mode should not be confused with additional relaxations in the GHz region, often called (narrow) central peaks, which are not in the scope of this article.

5. We have combined the data from the (200) and (300) BZs as D_q should be independent of the BZ choice.

6. In principle, PMN is known to grow also, for example in pyrochlore structure, but in this case both the lattice parameters and the TO mode frequency are completely different.

7. It was argued in 9 that the TO mode cannot couple noticeably to the TA branch because the independent mode intensities do not change with temperature. However, this not a valid argument since the measurements shown in figure of Ref. 9 were done in (20q) zone, where both TA and TO modes have similar structure factors so that eventual eigenvector change has no chance to produce such drastic intensity changes as those observed in the quoted 9 case of $SrTiO_3$.

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