Soft mode splitting in PMN below the Burns temperature observed by hyper-Raman scattering

B. Hehlen[1], M. Al-Sabbagh[1], A. Al-Zein[2], and J. Hlinka[3]

[1]Laboratoire Charles Coulomb (L2C), University Montpellier and CNRS, Place E. Bataillon, Montpellier, France
[2]European Synchrotron Radiation Facility (ESRF), 71 Avenue des Martyrs, Grenoble, France
[3]Institute of Physics, Academy of Sciences of the Czech Republic, Na Slovance 2, 18221 Praha 8, Czech Republic

Hyper-Raman scattering (HRS) is a non-linear inelastic spectroscopy sensitive to polar excitations in solids whatever their crystalline symmetry. This selection rule is very attractive in particular for centro-symmetric structures for which polar modes are inactive in Raman.
HRS has been performed in the cubic relaxors $PbMg_{1/3}Nb_{2/3}O_3$ (PMN) and $PbMg_{1/3}Ta_{2/3}O_3$, with particular attention to the low frequency region, down to 2 cm^{-1}. Spectra have been recorded over a wide temperature range (900K-30K in PMN) providing therefore the framework for a detailed analysis of the polarization dynamics in these systems.
In both materials, the soft mode response exhibits a doublet structure up to the highest temperature investigated [1,2] emphasizing a common property of cubic relaxors. The lowest frequency component is assigned to the primary soft mode of symmetry F_{1u}, while the second likely originates from a local disorder persisting until very high temperatures and lifting the cubic selection rules. On cooling, the soft F_{1u}-mode of PMN becomes overdamped at the onset of the Burns temperature, but very interestingly it splits between 600K and 400K into two components which harden on decreasing further the temperature. This behavior likely highlights the onset of a local anisotropy of the polarization and provides therefore new insight about the relaxor nature.

1. A. Al-Zein, *et al.* Phys. Rev. Lett. **105**, 017601 (2010)
2. S.B. Vakhrushev, *et al.* Phys. Solid State, **52**, 889 (2010)