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Soft mode dynamics of ferroelectric relaxors

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Abstract:

Hyper-Raman scattering is a non-linear inelastic spectroscopy sensitive to polar excitations of solids whatever the crystalline symmetry. This selection rule is very attractive in particular for centrosymmetric 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 $PbMg_{1/3}Ta_{2/3}O_3$ (PMT), with particular attention to the low frequency region, down to about 2 cm^{-1}. Spectra have been recorded over a wide temperature range (900K-30K in PMN) and the results compared to existing infrared and neutron scattering data. This complete set of vibrational information provides the framework for a detailed analysis of the polarization dynamics in these systems.

In particular, in both materials, the soft mode response exhibits a doublet structure up to the highest temperature investigated [1,2] emphasizing therefore 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 which lifts 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).

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