

*Space and time correlations of polar fluctuations in dielectric materials*Marek Paściak¹¹*Institute Of Physics, The Academy Of Sciences Of The Czech Republic, Prague, Czech Republic
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Modern dielectric materials with exceptional piezoelectric properties are widely used in various electromechanical devices (e.g. actuators or transducers). Special features of these materials are usually engineered by managing competing interactions and orders (e.g. by introducing substitutional disorder), often leading to compounds in which structural frustrations create heterogeneity on a nanoscopic level. Understanding structure-property relationship is in this case requires insight into spatio-temporal distribution of polar fluctuations and their interrelation with other structural instabilities as well as short-ranged chemical perturbations.

In this work we concentrate on two particular cases which have recently drawn a significant attention in the ferroelectric community and which pose a challenge for structural analysis.

$\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ (SBN), a uniaxial ferroelectric with excellent electro-optic and piezoelectric properties. The ferroelectric characteristics of SBN changes from normal to relaxor (broad and frequency-dependent maximum of dielectric susceptibility) with the degree of Sr content. The average structure is tetragonal, of a tungsten bronze type (P4bm space group). Local structure is affected by disorder on different levels. The NbO_6 chains create squared and pentagonal channels which are stochastically occupied by Ba and Sr cations. This gives rise to strain fields and in the cases of higher Sr concentrations to incommensurate modulation of octahedra tilts. Despite a considerable effort the source of polarization and huge susceptibility in SBN is not known in detail. The signatures of a short range order, however, are very strong in neutron and x-ray diffuse scattering data and the detailed analysis of them should help revealing missing pieces of understanding on a microscopic level.

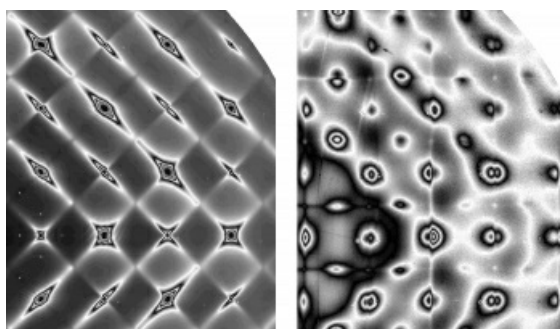
Lead ziconate (PbZrO_3 , PZO) is an antiferroelectric (AFE) material, the end member of a technologically important solid solution $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$. It has a peculiar temperature-driven AFE phase transition (PT) from cubic to orthorhombic state with no direct link between the two phases. There are three lattice modes involved in the PT: antiparallel displacements of Pb atoms and two rotation (tilting) modes of oxygen octahedra. What is the mechanism behind a simultaneous action of these distortions? Recent theories suggest either flexoelectric [1] or trilinear coupling [2] to play a major role, putting accents either on the dominant role of the ferroelectric instability or a soft phonon branch coupled to tilts, respectively. Detailed diffuse scattering ([3], see figure) analysis allow identification of the signals related to the important instabilities and track their dynamics and temperature dependence.

From the methodological point of view we approach both cases in a similar manner: the extensive diffuse scattering datasets are interpreted with the help of a multi-scale modelling, starting at the highest level of theory with density functional-based calculations for smaller supercell representations that allow us to build classical models of atomic interactions. These are consequently used for large-scale molecular dynamics simulations giving access to both time and space resolution of short-range correlations including polar distortions and octahedra tilting.

[1] Tagantsev, A. K. et al. (2013) Nature comms. 4, 2229.

[2] Hlinka, J. et al. (2014) Phys. Rev. Lett. 112, 197601.

[3] Paściak, M. et al. (2015) Phase Transitions 88, 273.



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