	Experiment title:	Experiment number:
	Relaxor ferroelectrics at high-pressure probed by single crystal x-ray diffraction.	HS-1813
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Report:

Relaxor ferroelectrics (called relaxors) form a special class among ferroelectric materials of which the understanding remains a challenging problem. However, relaxors are not only important from a fundamental point of view, but attract currently a large interest due to their potential impact in various applications, especially related to their ultrahigh piezoelectric effects [1, 2].

Although the true origin of the relaxor behavior remains controversial, it is generally admitted that their reported outstanding properties are governed by *a peculiar microstructure that consists of nanoscale polar regions* in a somewhat different matrix .

The aim of our experiment was to understand the effect of pressure on the local structure of the model relaxors $Pb(Mg_{1/3}Nb_{2/3})O_3$ and $(Na_{0.5}Bi_{0.5})TiO_3$, with special regard to short-range ordering, the change of size for polar nano-regions and the detection of instabilities towards pressure. Furthermore, we aimed at a understanding of the different behaviour on the *A*- and *B*-sites, which is a key element to understand intriguing differences between PMN and NBT.

Before this study, only little was known about the effect of high pressure on relaxors, since investigations were limited to high-pressure Raman investigations [3] [4].

Results

X-ray scattering imaging (XRSI) at a synchrotron source, probing a large region of the reciprocal space, has the invaluable advantage of giving within one experiment rapid

simultaneous information about the average structure (via diffraction) *and* local distortions (via diffuse scattering). In this sense XRSI is a powerful technique which allows overcoming the frequent controversy if the difference in phase transition parameters, observed by average and local techniques, is intrinsic or not.

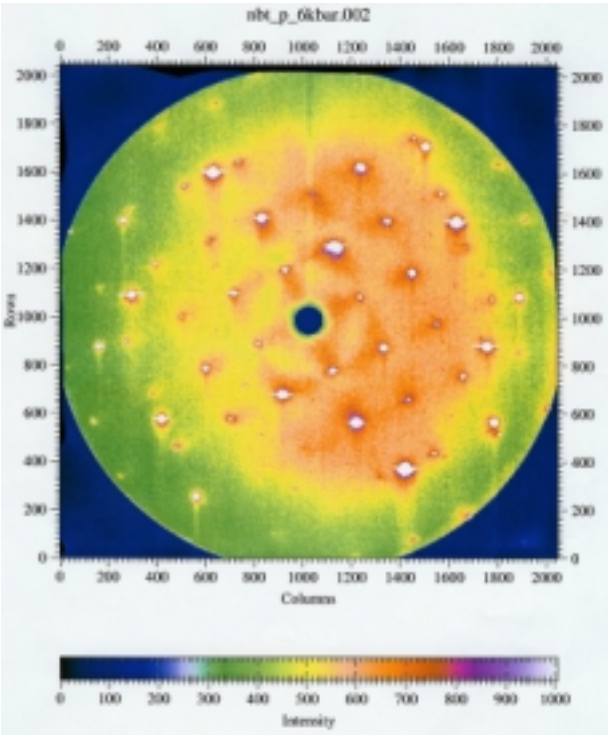
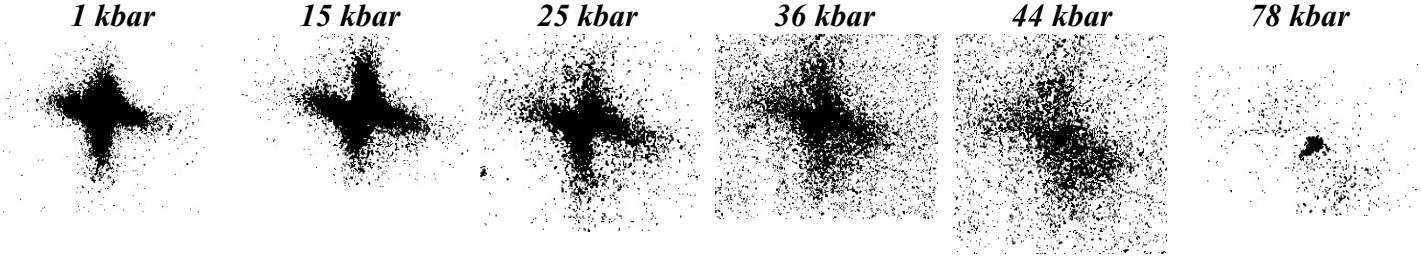


Figure 1 (left) shows an example of a low-pressure x-ray scattering pattern exhibiting average-structure-related Bragg peaks together with a strong diffuse scattering originating from the nano-scaled structure of $(Na_{0.5}Bi_{0.5})TiO_3$. For instance, the observation of diffuse scattering around the direct beam *and* the Bragg peaks suggests a **segregation of Na and Bi atoms on a local scale rather** than a perfect disorder or a superstructure-type arrangement. The anti-symmetric form of the diffuse scattering around the Bragg peaks illustrates a **correlation of the substitution disorder with a displacement-type disorder** leading to a local deformation of the unit cell. The analysis of the Bragg reflection illustrates that the **average structure undergoes two structural transitions** a first between 1.6 and 2.0 GPa and a second between 9.9 and 11.1 GPa.

Figure 2 (below) illustrates the spectacular pressure-dependent evolution of the diffuse scattering around the (300) reflection of $Pb(Mg_{1/3}Nb_{2/3})O_3$. The disappearance of the initially very strong diffuse scattering under the application of high-pressure is related to the ordering of the cations on one crystallographic site.



Our results show that the combination of the external parameter high-pressure with X-ray scattering imaging (XRSI) at a synchrotron source allows investigating the nano-scaled chemical and structural characteristics of relaxors, or more generally nano-scaled oxides, at a heretofore-unidentified degree.

[1] Park SE, Shrout TR. 1997. *J. Appl. Phys.* 82: 1804-11
[2] Fu H, Cohen RE. 2000. *Nature* 403: 281-3
[3] Kreisel J, Glazer AM, Bouvier P, Lucazeau G. 2001. *Phys. Rev. B* 63: 174106
[4] Kreisel J, Dkhil B, Bouvier P, Kiat JM. 2002. *Phys. Rev. B* 65: 172101

- *J. Kreisel will present the main lines of this investigation in the framework of an invited presentation at the 2002 Meeting of the International Union of Crystallography (IUCr).*
- *An article entitled High-pressure X-ray scattering imaging of oxides with a nano-scaled local structure is in preparation.*