

Diffuse scattering in $\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3$ - $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ relaxor solid solutions

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$\text{Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3$ (PIN) and $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ (PMN) are complex perovskites with the chemical formula $\text{A}(\text{B}'_x\text{B}''_{1-x})\text{O}_3$. The electrically balanced 1:1 B-site cation ordered domains, of which directions are along $\langle 111 \rangle$, can form in PIN. In contrast, since the 1:2 stoichiometric cation ratio is the chemical nature of PMN. Antiferroelectricity and sharp phase transition can be observed in the highly ordered PIN; whereas the incompletely ordered and disordered PIN are relaxors, which exhibit diffuse phase transition and slim polarization hysteresis loop. Since the 1:1 order-disorder behaviour in PIN-PMN as well as other complex perovskite materials plays a central role in influencing the physical properties [1-3], TEM studies were carried out to investigate the various structural ordering.

Centred dark-field (CDF) images of the 1:1 B-site ordered domains in the ceramics with $x = 0.0$ (i.e. PMN), 0.3 and 0.6 are shown in Figure 1. The size of the ordered domain increases as increasing PIN contents. We found that the ceramics with $x > 0.3$ have intermediate ordering. The distinct anti-phase boundaries (APBs) are clearly seen in $x = 0.6$, indicating the high degree of order. However, the variation of contrasts in the ordered regions suggests strong compositional fluctuation, i.e. different arrangement and combination of In^{3+} , Mg^{2+} and Nb^{5+} [4].

In addition to the superstructure reflection $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ given by 1:1 B-site ordering, the forbidden reflections $\frac{1}{2} \frac{1}{2} 0$ and diffuse streak and scattering are observed in selected-area electron diffraction (SAED) patterns, shown in Figure 2. The intensity of the features other than Bragg reflections depends on the composition of PIN, similar to the size of B-site ordered domains. The $\frac{1}{2} \frac{1}{2} 0$ reflections indicates that the crystal structure of the solid solutions have a lower symmetry than $Fm-3m$. Diffuse streaks along $\langle 110 \rangle^*$ are observed in all major zone axes, indicating the presence of either planar defects or short-range ordering along the directions. The ordered domains and the electrical properties of the ceramics suggest that the occurrence of diffuse streaks can be contributed by the large amount of short-range anti-parallel polarization [5], polar nanoregions (PNRs) [6], various kind of nanoscale 1:1 ordering [4] and APBs. Relaxor behavior is given by the short-range polarization or polar nanoregions. The directions of the polarization can be in different crystallographic directions but still be stabilized and electrically balanced by the APBs. These are the consequence of three cations in the B-site and the source of random-field. However, the origin of these diffuse streak and scattering in SAED patterns is not fully understood [6,7].

References:

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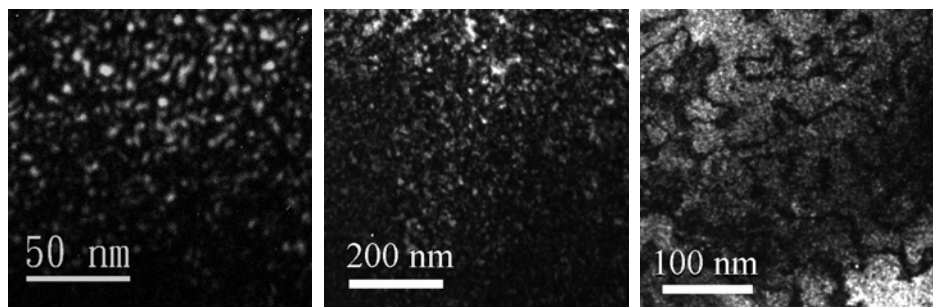


Figure 1. Centered dark-field micrograph taken along a $\langle 110 \rangle$ zone axis using a $\frac{1}{2} \frac{1}{2} \frac{1}{2}$ superstructure reflection (indicated as **g**) from a region of the ceramic with (left) $x=0$, (middle) $x=0.3$ and (right) $x=0.6$.

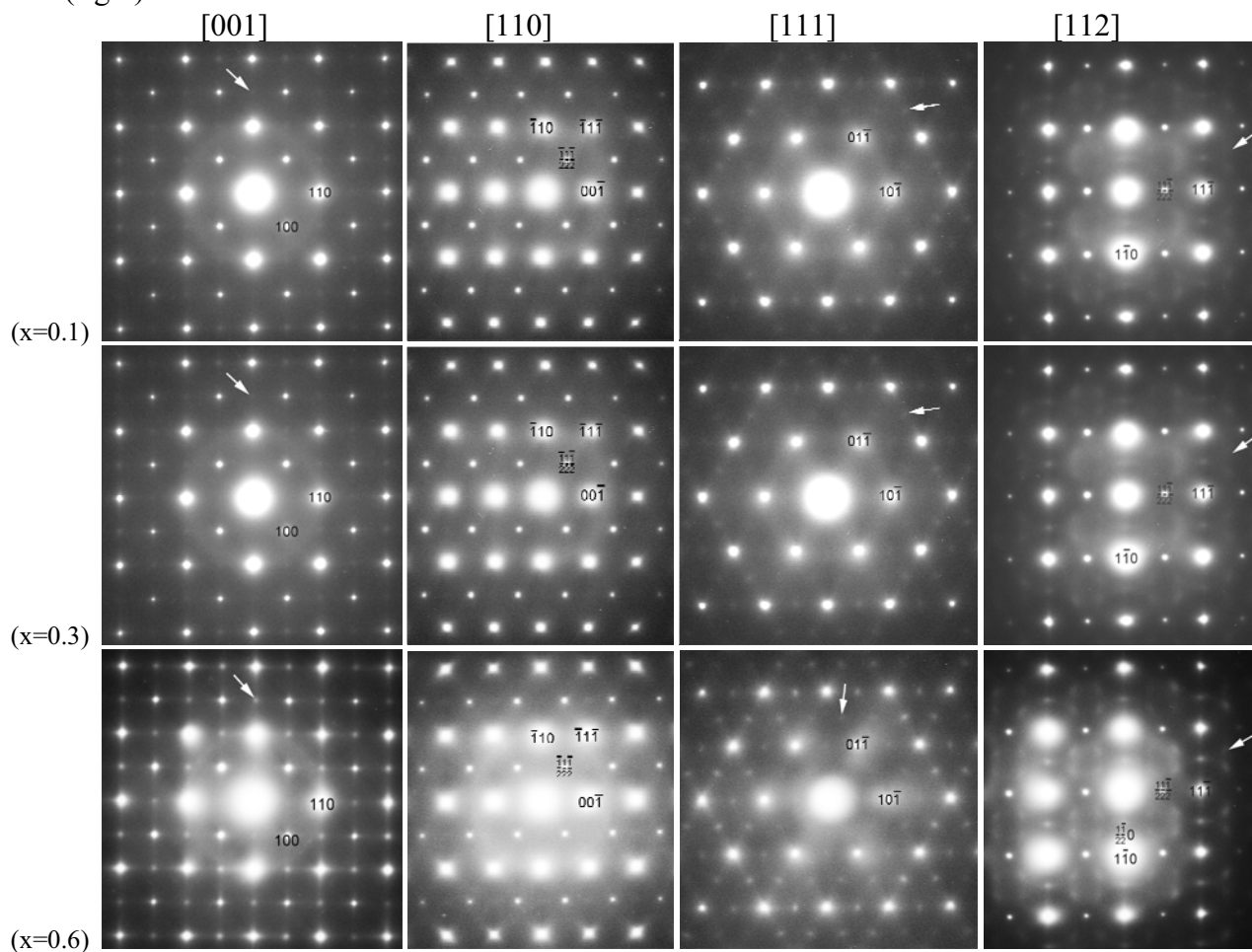


Figure 2. SADPs of the ceramic with $x=0.1$, 0.3 and 0.6 recorded along $[001]$, $[110]$, $[111]$ and $[112]$ zone axes, respectively. The forbidden reflection $\frac{1}{2} \frac{1}{2} 0$ is indicated by an arrow.