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Temperature dependence of gradational system in $Pb_{0.7}Sr_{0.3}TiO_3$ nanoparticles

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Introduction

Technologies for forming uniform and high-quality perovskite ferroelectric materials have been advanced up to controlling the nanosized materials. However, the nanosized materials show unfavorable performances such as decreasing of dielectric permittivity and spontaneous polarization. This deterioration of ferroelectricity has been called 'size effect'. From the structural and theoretical studies in recent years, it has been recognized that nanoparticles have inhomogeneous structure in the vicinity of surface due to the surface effect.

Recently, we have performed diffraction experiment using synchrotron radiation (SR) X-ray for $Pb_{0.7}Sr_{0.3}TiO_3$ (PST7/3) nanoparticles at room-temperature [1]. The FWHM of identical peaks depended on not only the particle size but also the Miller index *l*. Furthermore, the peak shape became asymmetric with increasing the *l*. The *l* dependence and asymmetry have been well explained by an inline dipole series model which shows gradational lattice and dipole in the vicinity of surface. This shows that the gradational system originate from the dipole interaction in ferroelectric phase. We, therefore, performed SR X-ray diffraction measurement to examine temperature dependence of the gradational system in PST7/3 nanoparticles.

Experiment, Results and Discussion

SR X-ray diffraction measurement was performed for PST7/3 nanoparticles up to 680 K using a diffractometer installed at BL-1B in the KEK-PF. The incident beam energy was 8 keV. Rietveld analyses were performed up to 2θ=140˚ for the collected data omitting those from 58˚ to 84˚ which corresponds to background and Bragg peaks from Be window of furnace. All data were normalized by using the integrated intensities of incident beam and corrected by using the scale factor. Measurement temperature at the sample was estimated from lattice constant of NaCl powder.

Figure 1 shows peak profiles of 200 and 002 reflections at various temperatures and particles sizes. As shown in fig. 1, the 002 reflection is distinguishable from 200 at 297 K in each particle sizes. The 002 reflections are broader than 200 and asymmetric tailing toward the high-2θ. The 200 and 002 are confused at about 570 K in the particle size of 50 and 10 nm, however, the peaks narrow with increasing temperature, and all peaks become symmetric at 678 K, while the size-induced broadening is observed at each temperature. This shows that the particle size scarcely affects to T_C. Estimated phase transition temperature is about 630 K and consistent with that of balk. Mean lattice constants were obtained from tetragonal single phase model with the space group *P4mm*. Particle size dependence of mean lattice constants shows similar tendency with those at room-temperature. The lattice constant, *a*, increases with decreasing particle size, while the *c* shows a minimum at the particle size 50 nm (it can be seen as the peak positions of 200 and 002 in fig. 1). On the other hand, the aspect ratio (tolerance factor), *c*/*a*, is independent of the particle size and universally depends on temperature.

Thus, the results suggest that the size effect originates from the strain due to the gradational system in the vicinity of surface. Although the analysis using multilayer model has not been finished, our Rietveld analyses for this data and those obtained by high-resolution diffractometer at BL-4B2 will make them clear.

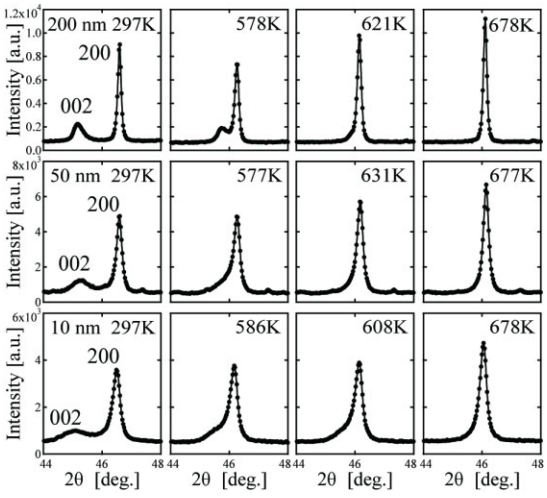

FIG. 1 Peak profiles of 200 and 002 reflections in PST7/3 nanoparticles at various temperatures and particle sizes.

References

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