crystals

MDPI

SUPPLEMENTARY MATERIALS
for:
Raman Response of Quantum Critical Ferroelectric Pb-doped $SrTiO_3$

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Synchrotron powder diffraction data collected at the P24 (EH2) PETRA III beamline, DESY (Hamburg, Germany) using the 2-D marCCD 165 detector. The monochromatic beam at wavelength λ = 0.5607 Å was slitted down to 300 × 300 μm². 200-μm-diameter glass capillaries were used for mounting finely ground powder samples. The sample-to-detector distance (D ≈ 75 mm), the wavelength and the detector parameters were calibrated using a LaB6 NIST reference powder sample; calibration and subsequent integration were performed using Dioptas software [17].

Powder diffraction data refinement was carried out using FullProf suit [18]. The refined parameters were unit cell dimensions, background, and profile parameters (profile match known as Le Bail fitting). The pseudo-Voigt profile is used to model the Bragg reflection profile, which is linear combination of Gaussian and Lorentzian model function with mixing coefficient η:

$$pV(2\theta) = \eta L(2\theta)+(1-\eta)G(2\theta) \qquad 0\leq\eta\leq1 \tag{1}$$

It is well known that intrinsic and extrinsic factors contribute to the broadening of the line profile of Bragg reflections. The extrinsic factor is the contribution from the diffractometer, which, as a result of convolution with the intrinsic profile, gives the observed diffraction pattern. Intrinsic contributions are from the features of the sample, namely, from the small size of the coherent scattering regions and strain in the crystal structure of the sample. The instrumental resolution function (IRF) was calculated from LaB_6 NIST powder diffraction pattern to consider the contribution of the diffractometer to the broadening of the diffraction pattern profile. Taking into account the IRF made it possible to determine the intrinsic parameters of the sample, which affect the broadening of the line profile. In our case, we did not consider the contribution from the size effect, since the grains of the ceramic sample prepared by the standard ceramic technology were more than 200 nanometers.

The intrinsic profile of a particular reflection due to a strain effect has an integral breadth β_{strain}. Fullprof software used the so-called maximum strain, which is derived from the integral breadth β_{strain} as

$$\varepsilon = \frac{\Delta d}{d} = \frac{1}{2}\beta_{strain} d_{hkl} \tag{2}$$

The integral width is calculated individually for each peak, which makes it possible to estimate not only the magnitude of microstrain, but also anisotropy. The results of refining the model of the line profile of the diffraction pattern from the studied samples are shown in Figures S1-S3. At 250 K, all samples are perfectly indexed using a cubic crystal structure model with space group $Pm\bar{3}m$. Lattice parameters, maximal strains with anisotropy and refined parameters of fits presented in Table S1.

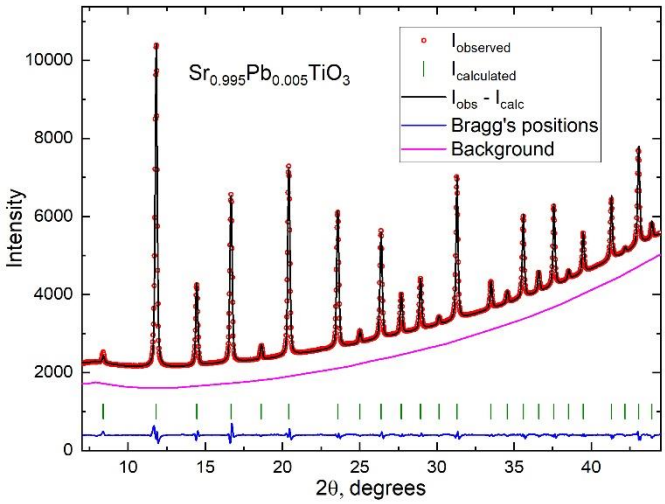

Figure S1: Diffraction pattern and refined model of profile for $Sr_{1-x}Pb_xTiO_3$ with $x = 0.005$

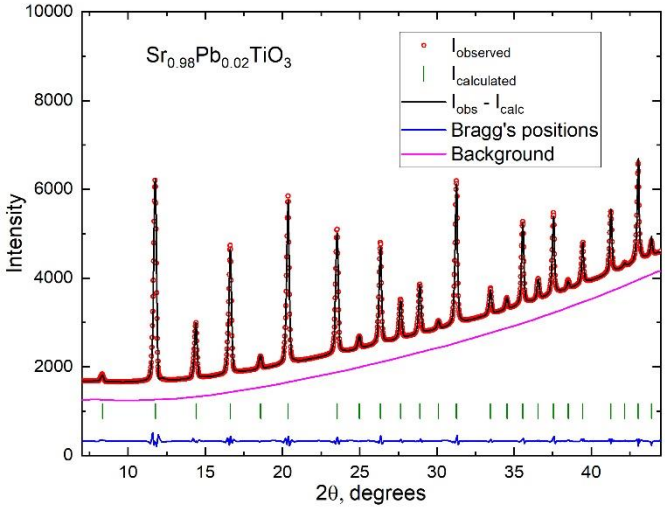

Figure S2: Diffraction pattern and refined model of profile for $Sr_{1-x}Pb_xTiO_3$ with $x = 0.02$

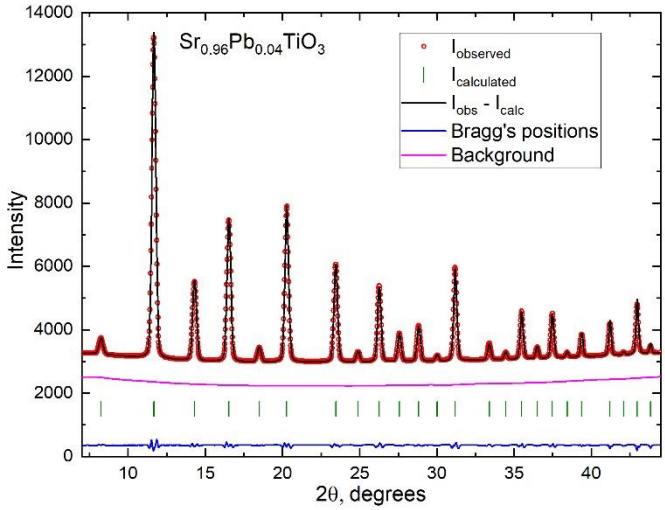

Figure S3: Diffraction pattern and refined model of profile for $Sr_{1-x}Pb_xTiO_3$ with $x = 0.04$

Table S1: Refined lattice parameters from Le Bail fit, maximal strain with anisotropy and reliability factors with all non-excluded points

Parameter	$Sr_{1-x}Pb_xTiO_3$		
	x = 0,005	x = 0,02	x = 0,04
Lattice constant *a*, Å	3,910(1)	3,908(1)	3,903(1)
Maximal strain ε, %% (x10000)	7.2619	18.2894	29.1573
Anisotropy of strain, %%	0.0510	0.3578	0.9077
R_p, %	0.436	0.371	0.395
R_{wp}, %	0.819	0.699	0.614
R_{exp}	1.70	1.85	1.70
χ^2	0.233	0.144	0.131
Bragg R-factor, %	0.084	0.124	0.112

References

17. Prescher, C., Prakapenka, V. B. DIOPTAS : a program for reduction of two-dimensional X-ray diffraction data and data exploration. *High Pressure Research* **2015**, *35*, 223–230.
18. Rodriguez-Carvajal, J. Recent developments of the program FULLPROF. *Comm. Powder Diffr. (IUCr) Newsl*. **2001**, *26*, 12-19.