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Ferroelectric State and Polarization Switching Behaviour of Ultrafine $BaTiO_3$ Nanoparticles with Large-Scale Size Uniformity

Yanan Hao,[1,2] Zunpeng Feng,[1] Soham Banerjee,[3] Xiaohui Wang,[2,*] Simon J. L. Billinge,[3,4,*] Jiesu Wang,[5] Kuijuan Jin,[5] Ke Bi,[1,*] and Longtu Li[2]

1State Key Laboratory of Information Photonics and Optical Communications,School of Science, Beijing University of Posts and Telecommunications, Beijing 100876, China

2State Key Laboratory of New Ceramics and Fine Processing,School of Materials Science and Engineering, Tsinghua University, Beijing 100084, China

3 Applied Physics and Applied Mathematics Departments, Columbia University, New York, NY 10027

4Condensed Matter Physics and Materials Science Department,Brookhaven National Laboratory, Upton, NY 11973

5Beijing National Laboratory for Condensed Matter Physics,Institute of Physics,Chinese Academyof Sciences,Beijing 100190, China

*Correspondence should be addressed to: X. W. (wxh@tsinghua.edu.cn); S. B. (sb2896@columbia.edu); and K. B. (bike@bupt.edu.cn).

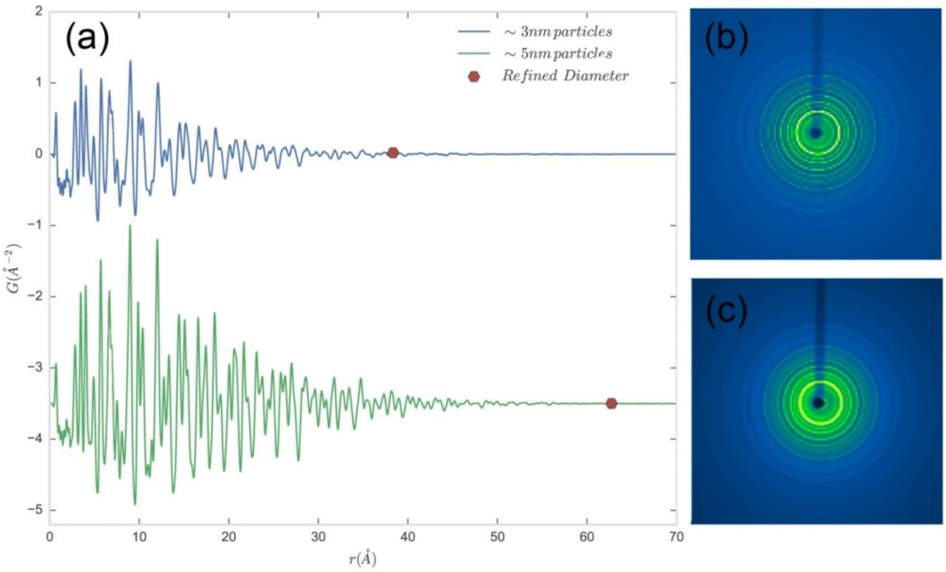

Figure S1. (a) Measured PDFs for the BTO NPs with crystal sizes approximately of 3 nm and 5 nm, respectively. And the refined diameter of the NPs is marked using red hexagon. (b) and (c) are the selected area diffraction patterns for the ～3nm and ～5 nm BTO NPs, respectively.

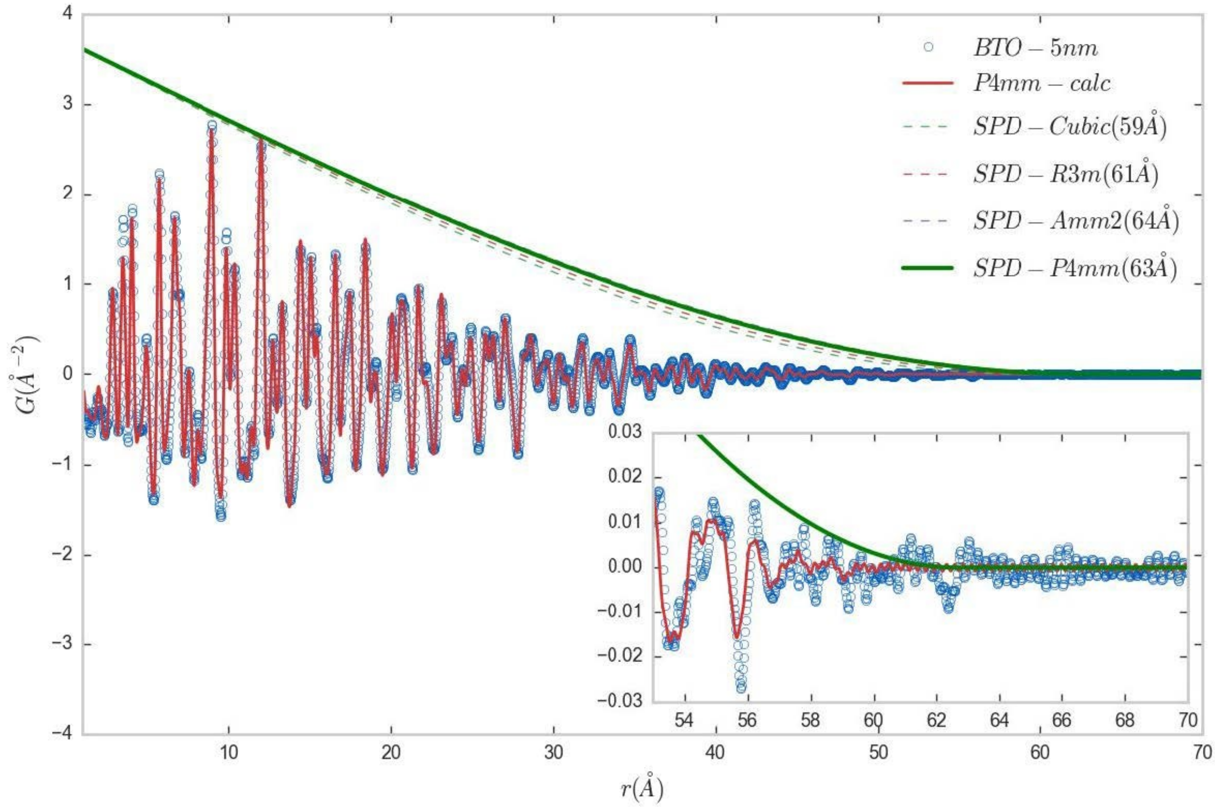

Figure S2. Measured (open circles) and calculated (red solid lines) PDFs, and four candidate $BaTiO_3$ crystallographic phases fit to the experimental PDF for thesub 10 nm nanocrystals. Inset is an amplification of the curves at high *r* range.

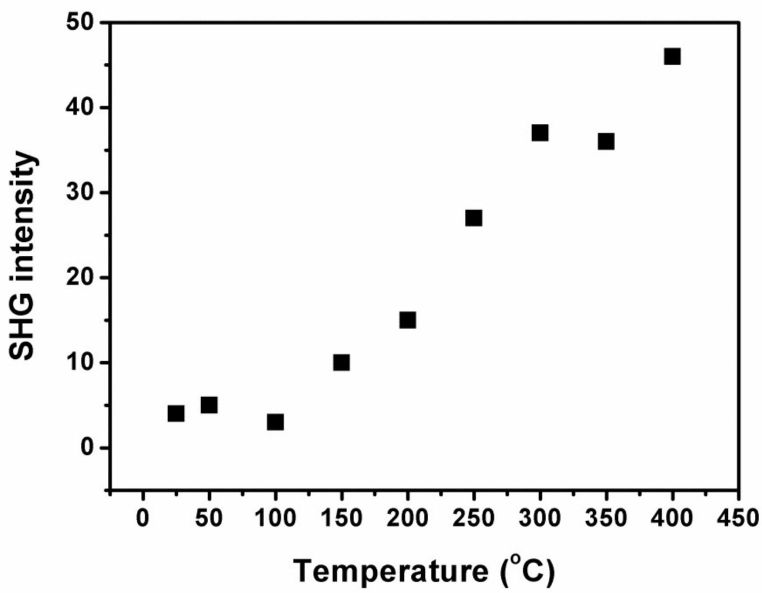

Figure S3. The background SHG signal variation at gradually increased temperatures.

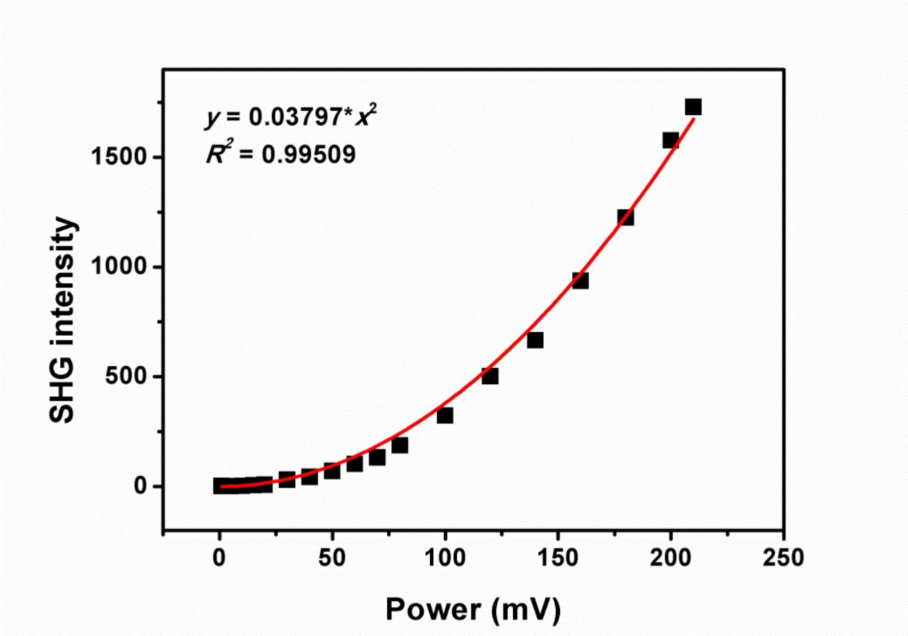

Figure S4. Quadratic dependence between detected SHG signal and the laser power.As the incident light power increases, the SHG signal increases following the fitting curve y=0.3797*x^2 with a high R^2 value, confirming the second nonlinear response of the nanocrystals.

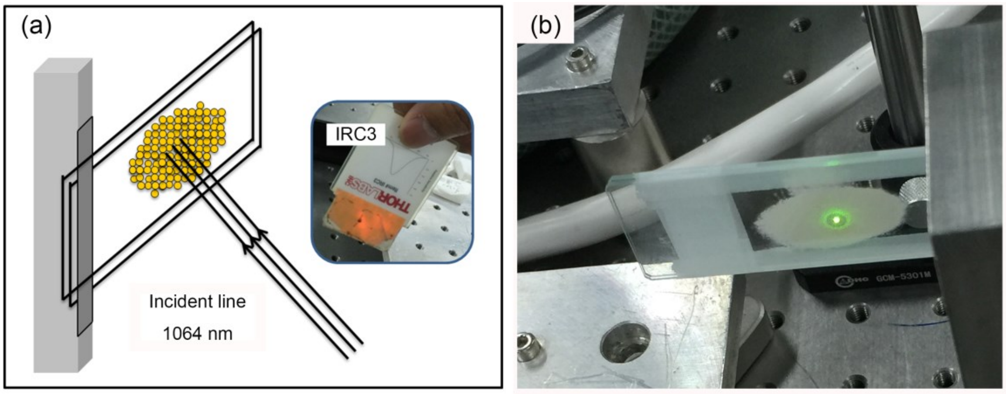

Figure S5. (a) Schematic diagram for the visual SHG measurement, the insertion is the THOR-LABS IRC3 plate which is used to detect the SHG signal. (b) Photograph showing that green second harmonic signal (532 nm) is generated when an incident line of 1064 nm is irradiated on the 4.5 nm NPs.

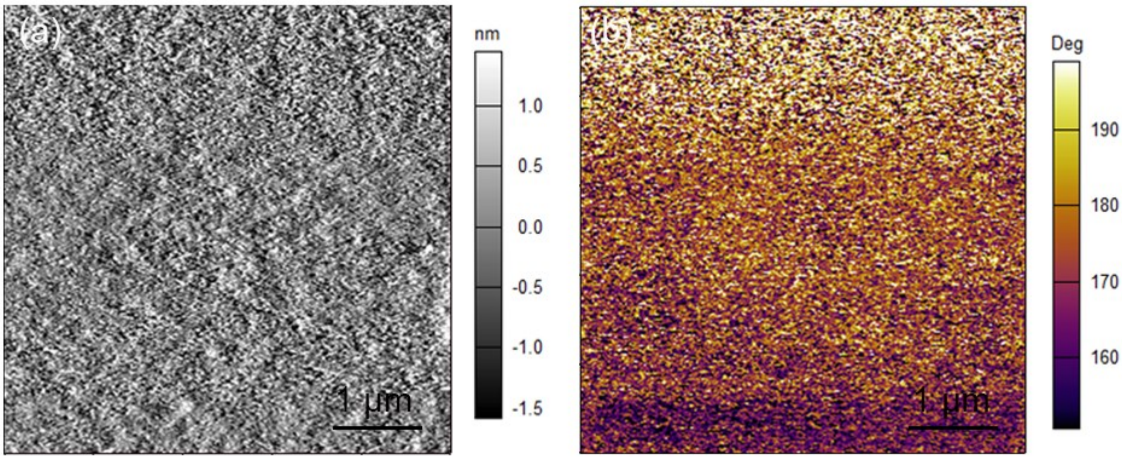

Figure S6. (a) Ferroelectric polarization switching amplitude image and (b) phase image of the amorphous BTO film. No ferroelectric switching behavior observed on this sample.

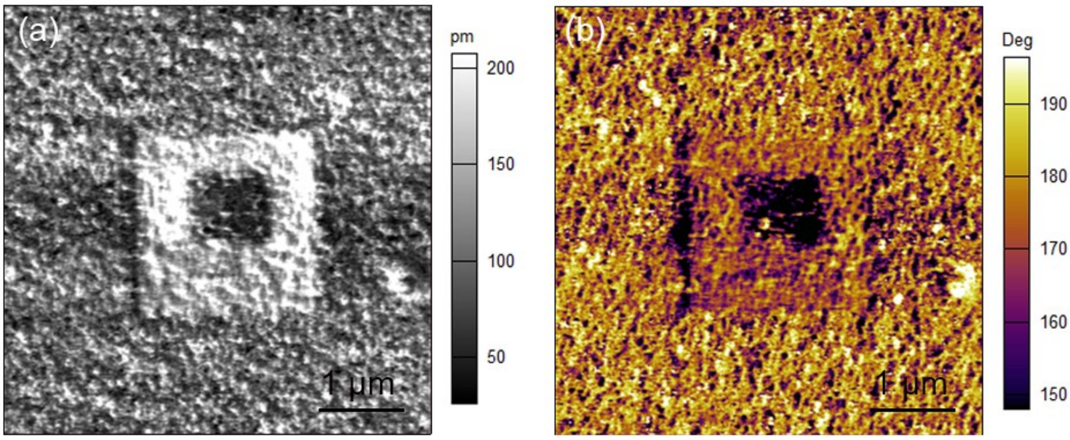

Figure S7. (a) Ferroelectric polarization switching amplitude image and (b) phase image of the BTO nanocrystal film that heated at 400 °C for 10 min.

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