Electronic supplementary information (ESI)

Wide-frequency-range dielectric tuning of BaTiO₃ by embedding metal nanocrystals

Zhengwei Xiong¹, Qian Liu¹, Jinlong Tang¹, Leiming Fang², Xiaoqiang Zhang³, Jun

Li⁴, Yajun Fu¹, Jin Wang¹, Zhipeng Gao^{1,4}, Deli Shi^{1†}

Affiliations:

¹Joint Laboratory for Extreme Conditions Matter Properties, Southwest University of Science and Technology, Mianyang, 621010, China

²Institute of Physics Nuclear and Chemistry, China Academy of Engineering Physics, Mianyang, 621900, China

³ Institute of Electronic Engineering, China Academy of Engineering Physics, Mianyang, Mianyang, 621900, China

⁴Institute of Fluid Physics, China Academy of Engineering Physics, Mianyang, 621900, China

*Corresponding author:

Deli Shi (deli.s@foxmail.com)

[†] Corresponding author. E-mail: 2801465754@qq.com

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We further provided the EELS of Fe- $L_{3,2}$ in the composite films (Fig. S1, Supplementary information). It can be seen that the position of the L_3 and L_2 edge from the area of Fe is 707.2 and 721.3 eV, respectively. It proves that the Fe element presents a metallic state in the composite films instead of iron oxides [1, 2].



Fig. S1 Electron energy loss spectroscopy of Fe- $L_{3,2}$ in the composite films.



Fig. S2(a) Temperature dependence conductivity (σ) of 0.5 wt% Nb:SrTiO₃ substrates and (b) pure epitaxial BTO films at varied frequencies.

Fig. S2 shows the temperature dependence conductivity (σ) of 0.5 wt% Nb:SrTiO₃ substrates and epitaxial BTO films. With the increase of temperatures, the σ of

Nb:SrTiO₃ substrate is almost linearly decreased (Fig. S2a), ascribing that the Nb:SrTiO₃ behaves as a doped semiconductor with electrons being thermally activated. On the contrary, the σ -*T* curves of BTO films present the insulating properties. So we can exclude the influence of 0.5 wt% Nb:SrTiO₃ substrates to the films. By comparison, the conductivities of Fe NCs-BTO composites (Figs. 6c, d) are higher than that of BTO films (Fig. S2b), proving that the embedded Fe NCs could really increase the σ of BTO matrix. The increased leakage conductance further brings the increase of tan δ (Fig. 4b). To sum up, it can be concluded that the embedded Fe NCs could indeed change the dielectric properties of epitaxial BTO films.

[1] J. Yuan, E. Gu, M. Gester, J. A. Bland, L. M. Brown, Electron-energy-loss spectroscopy of Fe thin films on GaAs (001), *J. Appl. Phys.*, 1994, **75**, 6501.

[2] K. F. Chen, S. C. Lo, L. Chang, *et al.*, Valence state map of iron oxide thin film obtained from electron spectroscopy imaging series, Micron, 2007, **38**, 354-361.