Supporting Information: Dislocations Promoted A-Site Non-Stoichiometry and Their Impacts On Proton Transport Properties of Epitaxial Barium Zirconate Thin Films

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S1: BZO target were measured by XRD

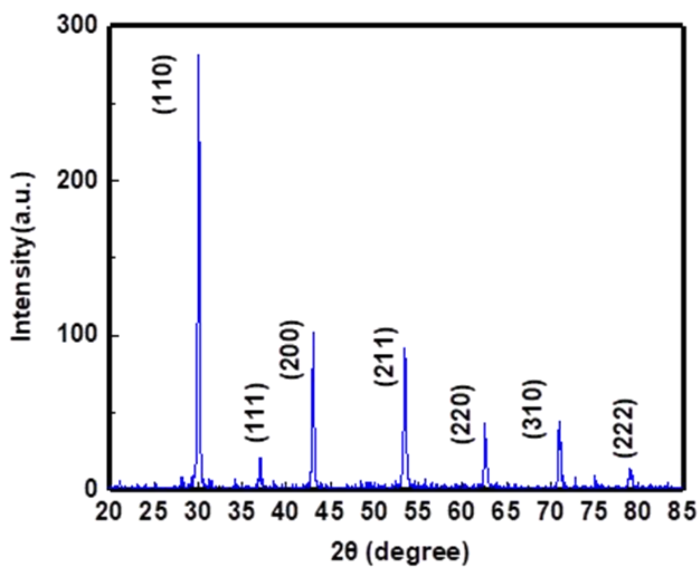

Figure S1. XRD diffractogram of BZO target.

The peaks of the diffractogram can be assigned to the different crystal planes of BZO. All the peaks are assigned to a cubic perovskite structure of BZO (JCPS 06-0399). The estimated lattice parameter from (200) plane is 4.198 Å.

S2: The cation composition of NGO substrate and 500 nm BZO thin film through measurements of Energy Dispersive Spectrometer (EDS)

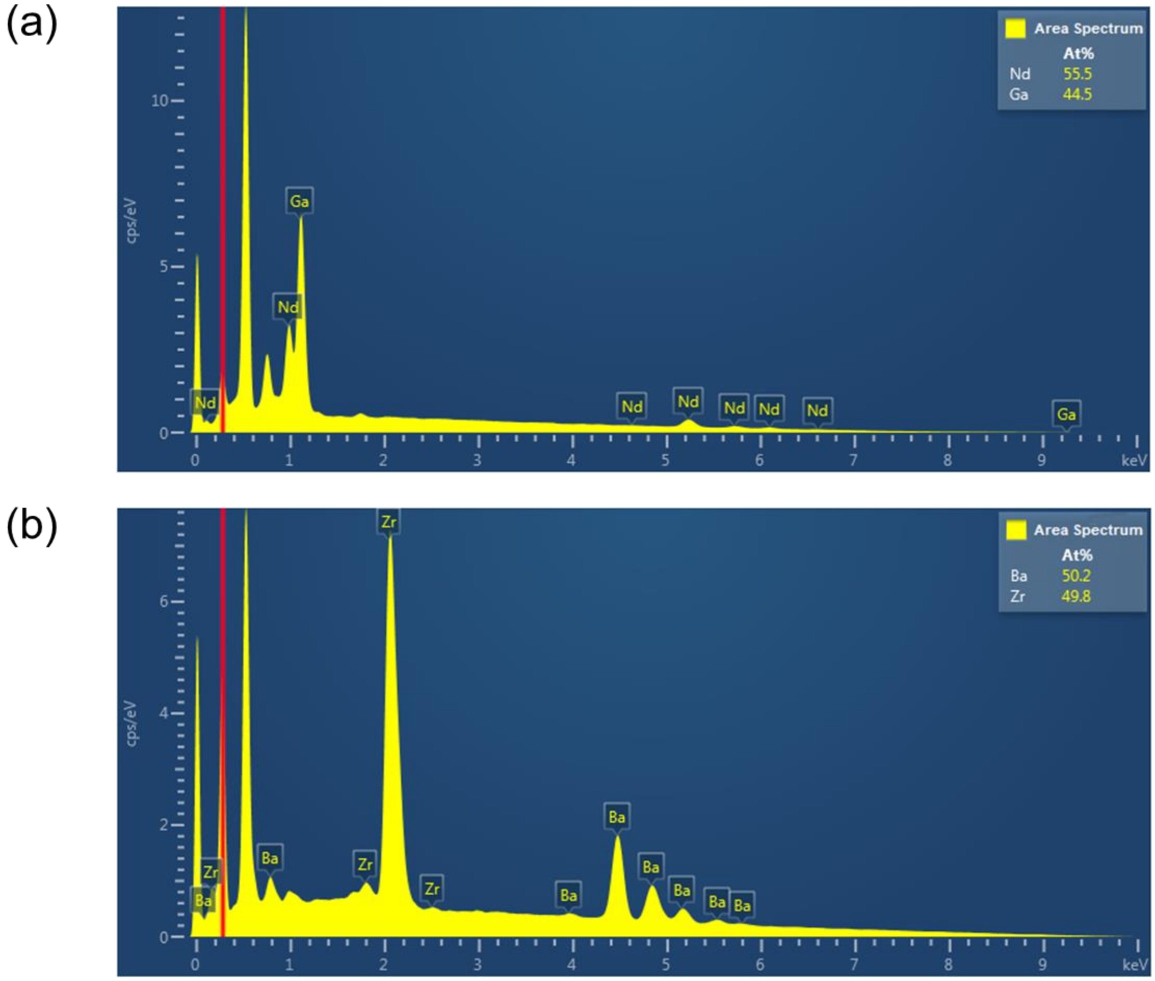

Figure S2. Cation composition obtained from EDS: (a) NGO substrate, (b) 500 nm BZO thin film.

The cation composition of thin films and substrate can be detected through Energy Dispersive Spectrometer (EDS), as shown in Figure S2. After the calculation from EDS measurements, the atomic ratio of Ba and Zr is 1.01 (50.2% / 49.8%) for the 500 nm BZO thin film. The fabricated thin film has Ba : Zr atomic ratio close to 1:1. The atomic ratio between Nd and Ga is 1.25 (55.5% / 44.5%) for the bare NGO substrate. All the results are listed in Table 1. The 500nm BZO sample is chosen for an accurate characterization of atomic composition.

Table 1: Cation composition of NGO substrate and 500 nm BZO thin film

NGO Substrate	Nd	Ga	Nd : Ga
	55.5%	44.5%	1.25
BZO thin film	Ba	Zr	Ba : Zr
	50.2%	49.8%	1.01

S3: Resistance and conductivity of BZO thin films with different thickness as well as bare NGO substrate measured by electrochemical impedance spectroscopy (EIS)

As a supplement to Figure 5, Figure S3 shows the temperature dependent conductivity comparison of the four BZO films with different thickness (20nm, 50nm, 100nm and 500nm) and NGO substrate in the atmosphere of the wet 5% H_2 in Argon. NGO substrate demonstrates the larger resistive components in the high frequency semicircles for the entire temperature range. This observation proves that the contribution in Nyquist plot is dominated by thin film.

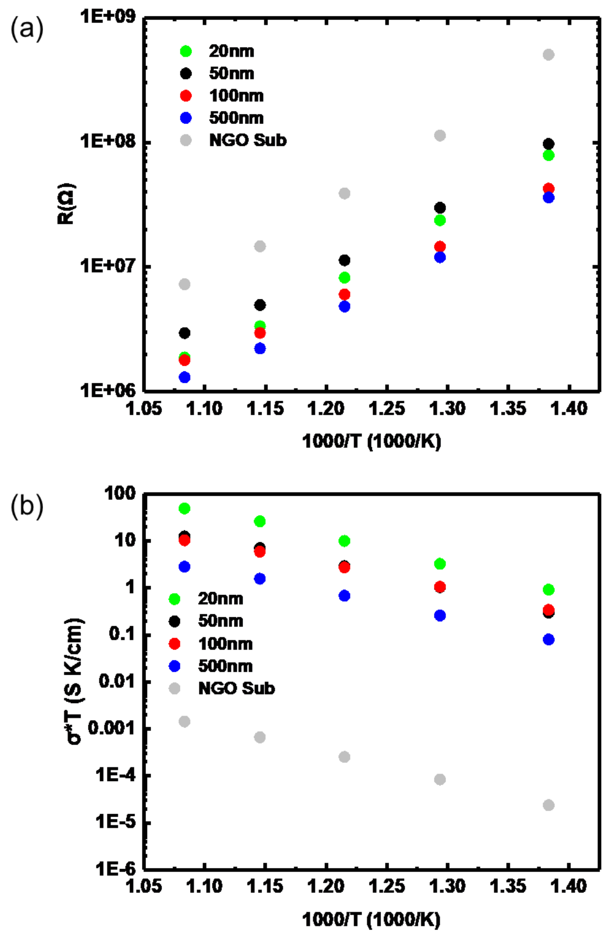

Figure S3. The temperature dependent resistance (a) and conductivity (b) are compared for the different thickness BZO thin films grown on NGO substrates in the wet 5% H_2 in Argon. The conductivity plot of bare NGO substrate are added as a reference.

S4: Conductivity comparison of 100nm BZO, BZY thin film on NGO substrate, and BZO thin film on MgO substrate, in H_2 wet atmosphere

To get more evidence of dislocation effect, more reference samples have been fabricated and characterized in H_2 wet atmosphere. From Figure S4 it can be seen that BZY thin film deposited on NGO substrate has the largest conductivity in whole temperature range, which reveals the influence of the dopants on the proton conductivity, while BZO thin film deposited on MgO substrate has the lowest highest conductivity. This result demonstrates that the dislocations at the mismatched interface could promote significantly the proton conductivity.

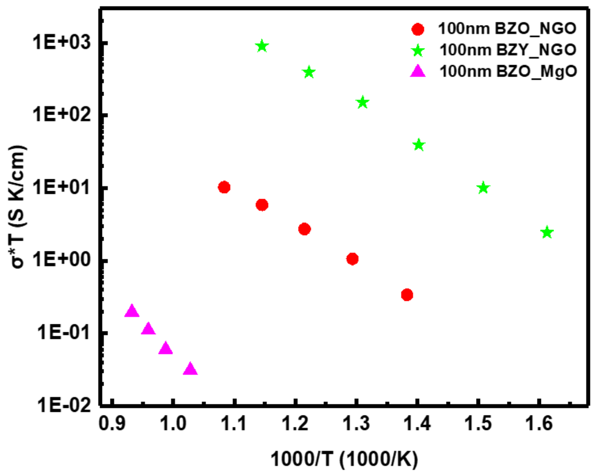

Figure S4. The temperature dependent conductivity is compared for the thickness of 100nm BZO and 100nm BZY thin film on NGO substrate, 100nm BZO thin film on MgO substrate, as well as bare NGO substrate are measured under H_2 wet atmosphere.

S5: Resistance and conductivity of 100 nm BZO thin film as well as bare NGO substrate measured under different atmosphere

To further prove the nature of the proton conduction, the comparisons of the temperature dependent resistance and conductivity plots of the 100 nm BZO films and NGO substrate in the pure Argon, the dry air, and the wet 5% H_2 in Argon were demonstrated in Figure S5.

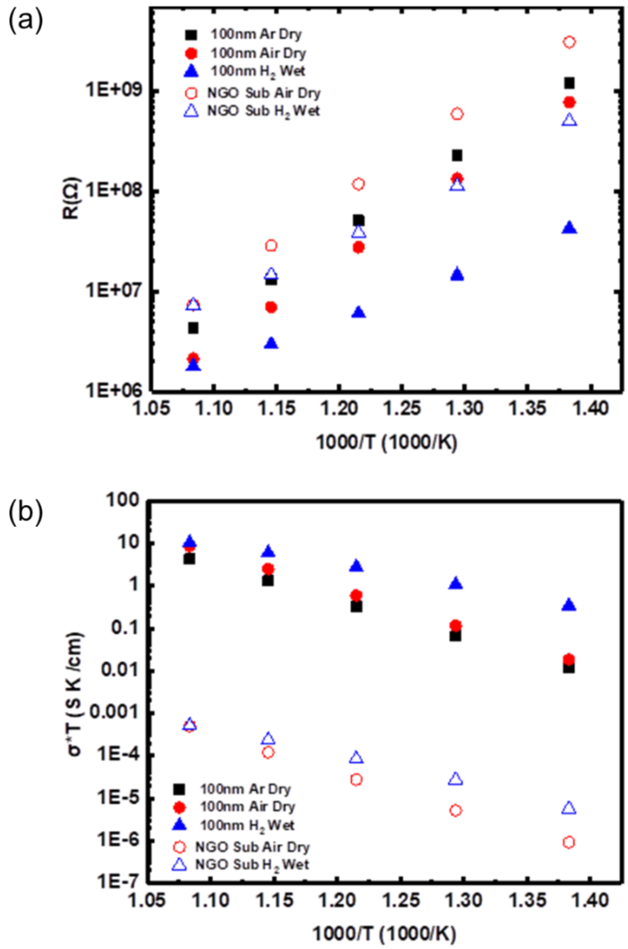

Figure S5. The temperature dependent resistance (a) and conductivity (b) is compared for 100nm BZO films and bare NGO substrate in three different atmospheres.