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Supplementary Information

Adsorption-Controlled Growth and the Influence of Stoichiometry on Electronic Transport in Hybrid Molecular Beam Epitaxy-Grown $BaSnO_3$ Films

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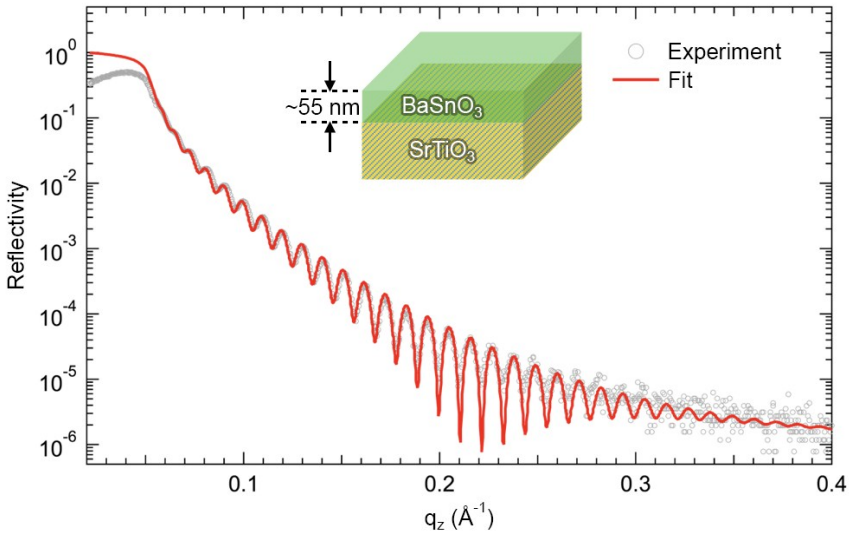
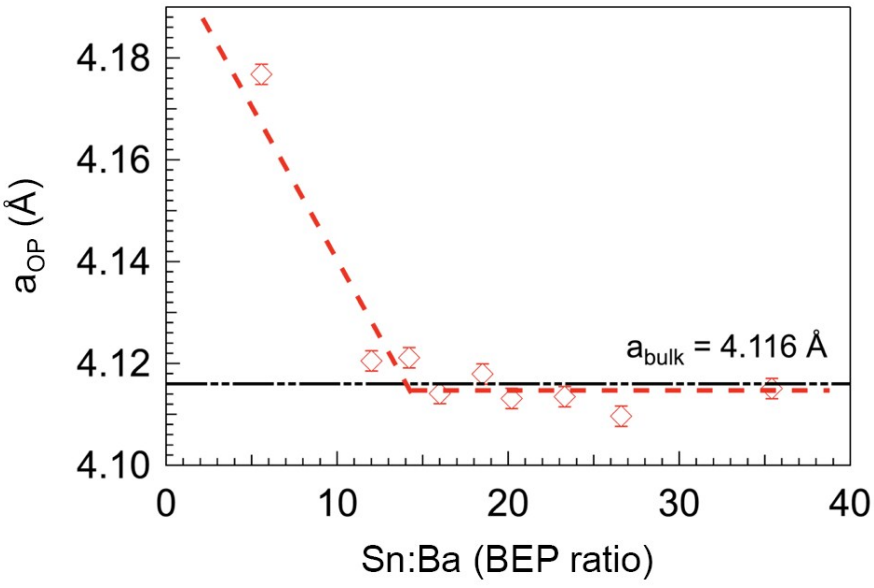
Figure S1



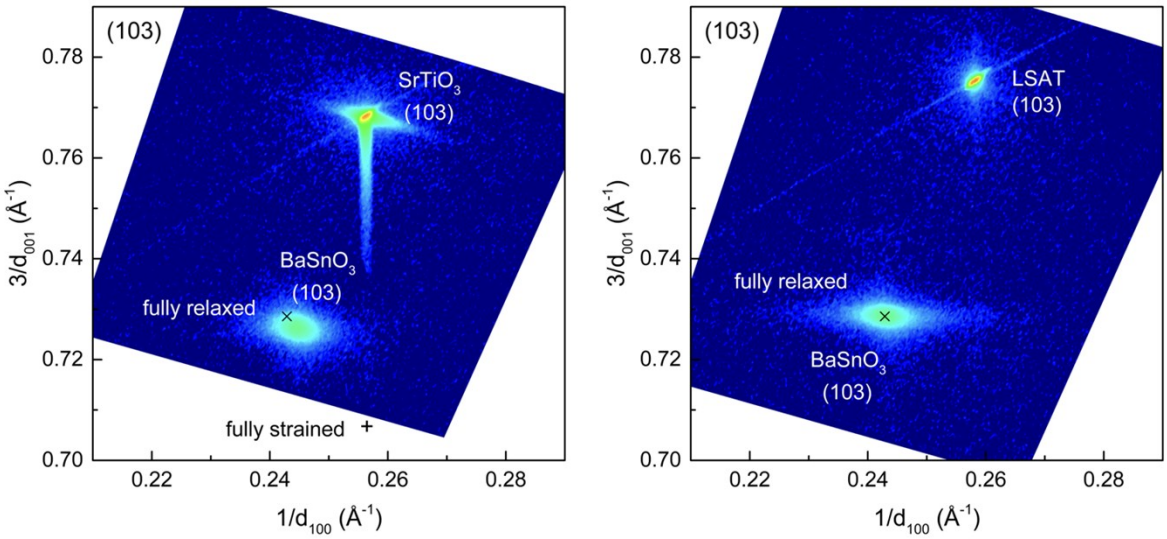
Gracing incidence x-ray reflectivity (GIXR) for a stoichiometric BSO film grown on STO substrate. The thickness determined using GenX fitting was ~ 55 nm. The density of the film was 7.34 g/cm^3 as compared to the bulk value of 7.24 g/cm^3. Roughness of the film from fitting and AFM were 8.2 Å and 7.6 Å, respectively.

Figure S2



Out-of-plane lattice parameter of BSO films grown on LSAT (001) substrates as a function of Sn/Ba BEP ratio. The trend in the a_{OP} is similar to the films grown on STO substrates. Note that RBS cannot be performed on these films since Ba and La are next to each other in the periodic table with atomic number of 56 and 57 respectively.

Figure S3

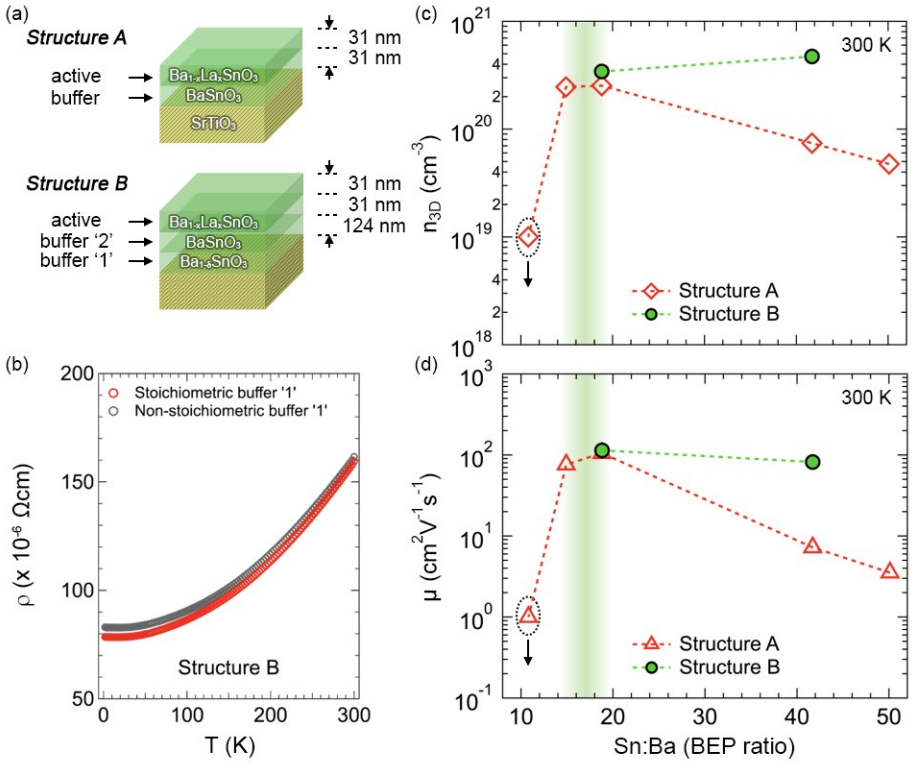


Off-axis reciprocal space maps around (103) plane of substrate of $BaSnO_3$ films on $SrTiO_3$ (left panel) and LSAT (right panel) substrates grown under identical conditions (Sn/Ba BEP = 18.5) for stoichiometric composition. Film thicknesses were between 36-45 nm. Clearly, film on STO (a = 3.905 Å) is not fully relaxed whereas that on LSAT (a = 3.868 Å) is fully relaxed. These results confirm why a_{OP} of films on STO is slightly increased compared to bulk BSO lattice parameter (4.116 Å) whereas a_{OP} is identical to that of bulk for film on LSAT. Note that the expected value of a_{OP} of a fully coherent BSO film on STO and LSAT substrates is 4.255 Å and 4.279 Å respectively.

Figure S4

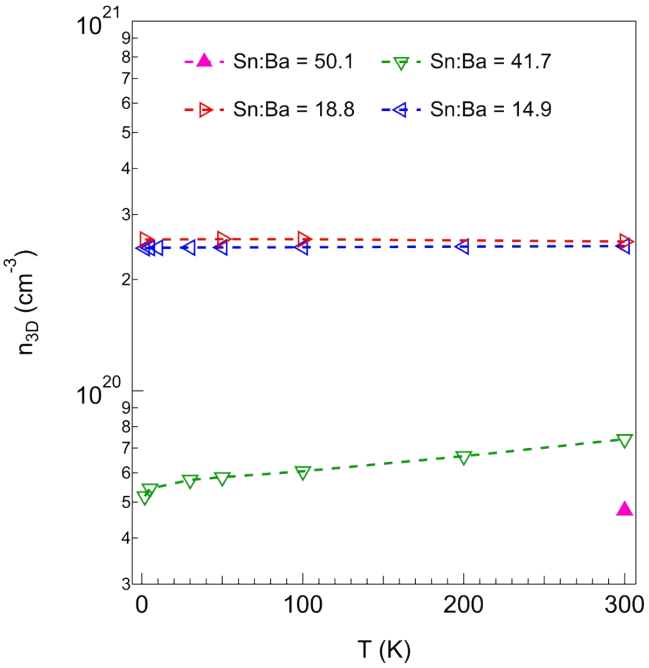
It is conceivable that the incorporation of point defects can influence the threading dislocation density in the doped BSO layer making it non-trivia to disentangle the role of dislocation and point defects on transport. To tackle this question, we grew a series of samples with the undoped buffer layer of varying stoichiometry followed by doped layer where electronic transport is probed. It is expected that if dislocation density in the buffer layer is affected by the introducing point defects, the transport in the active doped layer should likely also be affected. With this assumption, we grew structure A and B as illustrated in Fig. S4a. Structure A is identical to the one discussed in the manuscript where the stoichiometry of both doped layer and buffer layer were varied simultaneously. Structure B, on the other hand, has 124 nm thick Ba-deficient buffer layer (buffer#1) followed by a 31 nm undoped (buffer#2), plus 31 nm doped BSO layer. The difference between structure A and B is thus only in the introduction of an additional buffer#1.

Figure S4b shows T-dependent resistivity of structure B containing buffer#1 of stoichiometric (Sn/Ba BEP = 18.8) and *highly* nonstoichiometric composition (Sn/Ba BEP = 41.7). The corresponding room temperature carrier density and mobility values are shown in Fig. S4 c and d (green circle). For comparison, carrier density and mobility values for structure A as a function of Sn/Ba BEP ratios are also shown in Fig. S4c and d (reproduced from Fig. 5d-e of the main manuscript).



No change in resistivity, n_{3D} and μ for structure B with varying amount of point defects in buffer#1 was observed indicating the threading dislocation density in the active layer is not affected by point defects in the buffer layer. Further investigation using transmission electron microscopy combined with detailed transport and growth variations is needed, which is beyond the scope of this study.

Figure S5



Temperature dependence of n_{3D} for samples grown with different Sn:Ba BEP ratios showing carrier freeze-out for a highly non-stoichiometric (Sn/Ba BEP = 41.7) sample.