

Supporting Information:
Thickness independent vibrational thermal
conductance across confined solid-solution
thin films

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RMS roughness

The RMS roughnesses of the CSTO films prior to CTO deposition was characterized over $3 \mu\text{m} \times 3 \mu\text{m}$ areas via atomic force microscopy. The roughnesses are provided in the following table. As can be seen, all roughness values are approximately 2 \AA , which is one-half of the unit cell step height of STO. As these STO films were prepared with a TiO_2 termination, this is evidence that the 1 unit cell step height was preserved and that the interfacial roughnesses of all samples in this study are similar.

Table 1: RMS roughnesses measured via atomic force microscopy over $9 \mu\text{m}^2$ areas on the CSTO films prior to CTO deposition.

CSTO thickness	RMS roughness (\AA)
0 nm, TiO_2 -terminated SrTiO_3	2.08
1 nm	1.56
3 nm	2.29
5 nm	2.08
10 nm	1.88

Lattice mismatch

The lattice parameter of SrTiO_3 is 3.905 \AA and the pseudocubic lattice parameter of CaTiO_3 is 3.82 \AA . While we do not have a direct measure of the 50:50 CSTO composition's lattice parameter, use of Vegard's law results in a lattice parameter that is approximately 3.86 \AA . The lattice constant mismatches between CSTO and STO and CTO and CSTO are then each approximately 1%. Prior work on SrTiO_3 grown epitaxially on DyScO_3 , for which DyScO_3 has lattice parameters approximately 1 % larger than SrTiO_3 , revealed the onset of strain relaxation at thicknesses of $350\text{-}500 \text{ \AA}$ ($35\text{-}50 \text{ nm}$).¹ This value is well beyond the thickness of the CSTO layers in this work. Further, CaTiO_3 films prepared on LSAT substrates, for which a 1.3 % lattice mismatch exists, have been shown to not strain relax until a thickness of 20 nm .² For this reason, the assumption of pseudomorphic, strained CSTO layers in this work is reasonable. It is likely that the CTO layers are strain relaxed and our XRD data indicates that the degree of strain relaxation is equivalent as the

out-of-plane lattice parameter of all CTO films was 3.85 Å, as measured from the 004_p reflection.

Cation ordering

Cation ordering in the CSTO layer is unlikely. From a crystal chemistry perspective, the following conditions favor chemical ordering: 1) a simple crystal structure, 2) a cation concentration ratio of 1:1, 3) large difference in charge between the ordering cations, and 4) large ion size differences between the ordering cations, and 5) a large A-site ion size (for B-site ordering in the perovskite lattice).³ Of these, CTO and STO may be considered as simple crystal structures and the CSTO mixture satisfies the 1:1 cation ratio. However, there is no formal charge difference between Sr²⁺ and Ca²⁺ and their ionic radii in 12-fold coordination are 1.58 and 1.48 Å, respectively. Because there is no formal charge difference and the ionic radii difference is small (~6 %), it is highly unlikely that cation ordering would occur.

Oxygen vacancies

Oxygen vacancies will exist in all oxides to some degree. These films were prepared from sintered oxide targets with an oxygen plasma containing 20 % oxygen. The addition of oxygen to the plasma was done to compensate for oxygen loss due to gas-phase scattering during the deposition process. All CSTO films were processed in an identical manner and the CTO layers were deposited in the same deposition run. In depositing the CTO layer at the same time, each CSTO layer was heated in the sputter environment at the same time under the CTO sputter atmosphere conditions. This allowed for equilibration of the oxygen point defect concentrations in the CSTO layers, should any differences exist. As such, each film will have the same thermal/oxygen history and the oxygen point defect concentrations should be identical.

References

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