SUPPORTING INFORMATION

Enhanced Thermoelectric Figure of Merit in Thermally Robust, Nanostructured Superlattices based on $SrTiO_3$

Anas I. Abutaha, S.R. Sarath Kumar, Kun Li, Arash. M. Dehkordi, and Terry M. Tritt, and Husam N. Alshareef[*]

[a]Materials Science and Engineering, King Abdullah University of Science & Technology (KAUST), Thuwal 23955-6900, Saudi Arabia

[b]Department of Physics and Astronomy, Clemson University, SC, 29634, USA

1. Thermal conductivity measurement by 3ω method:

We used the differential 3ω method[1, 2] to measure the cross-plane thermal conductivity of the films. In this method, an alternating current passes through metal lines (Au microheaters), patterned on a reference sample and one with additional film (Figure S1a and S1b, respectively). The Au microheater act as both a heater and a thermometer. The alternating current with angular frequency ω creates Joule heating at 2ω ($I^2(\omega)R$, where I is the current and R is the resistance). This heating causes the temperature of the Au microheater to oscillate at 2ω, and hence its resistance oscillates at 2ω. Therefore, a small voltage component ($I(\omega) \times R(2\omega)$) is produced at 3ω.[2] The temperature rise of the Au microheater can be expressed by:[1]

$$\Delta T = 2R(\frac{dR}{dT})^{-1}\frac{V_{3\omega}}{V_{1\omega}}, \quad (1)$$

where R is the resistance of the Au microheater, dR/dT is the temperature dependent coefficient of resistance of the Au microheater, $V_{1\omega}$ and $V_{3\omega}$ are the amplitudes of the first and third harmonic voltage drops along the Au microheater, respectively.

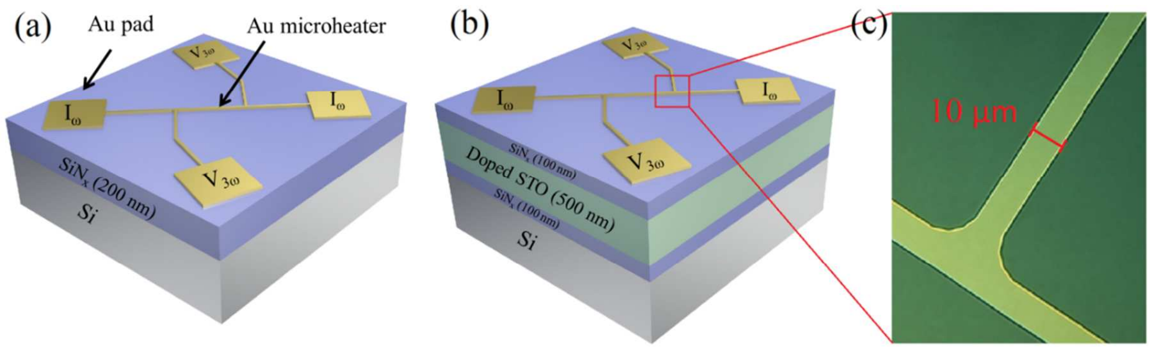

Figure S1. Schematic representations for the (a) reference sample and a (b) sample with additional film (doped STO), used in the differential 3ω method, and (c) optical micrograph of 10 μm wide Au microheater.

In order to pattern Au microheater on the electrically conductive doped STO films, an insulating layer is needed to prevent any leakage current. Therefore, SiN_x film was deposited on doped STO films (Figure S1b) and bare Si substrate (Figure S1a) using sputtering, and we measured the temperature rise from both samples. An algorithm was used to extract the temperature drops across the doped STO films from the rise in temperature across the two samples, using the differential 3ω method. Since the differential 3ω method allows the measurement of the thermal conductivity of the additional film, it removes uncertainties in the thermal properties of other layers.[1]

Since the width of the Au microheater is 10 μm (Figure S1c) which is much larger than the thickness (500 nm) of the film, the heat flow can be described as one dimensional, and the additional film has a frequency-independent temperature rise given by:[3]

$$\Delta T_f = \frac{P_l \, t_f}{w \, \lambda_f}, \quad (2)$$

where P_l, λ_f, t_f, and w are the heating power unit length, the thermal conductivity of the additional film, the thickness of the film, and the width of the microheater, respectively. The plots of measured ΔT as a function of ω at 100K and 300 K are shown in Figure S2a and S2b, respectively. It is evident that ΔT resulted from SPTO film is higher than ΔT resulted from STNO film, which is attributed to lower λ_f of SPTO film compared to STNO film (equation 2).

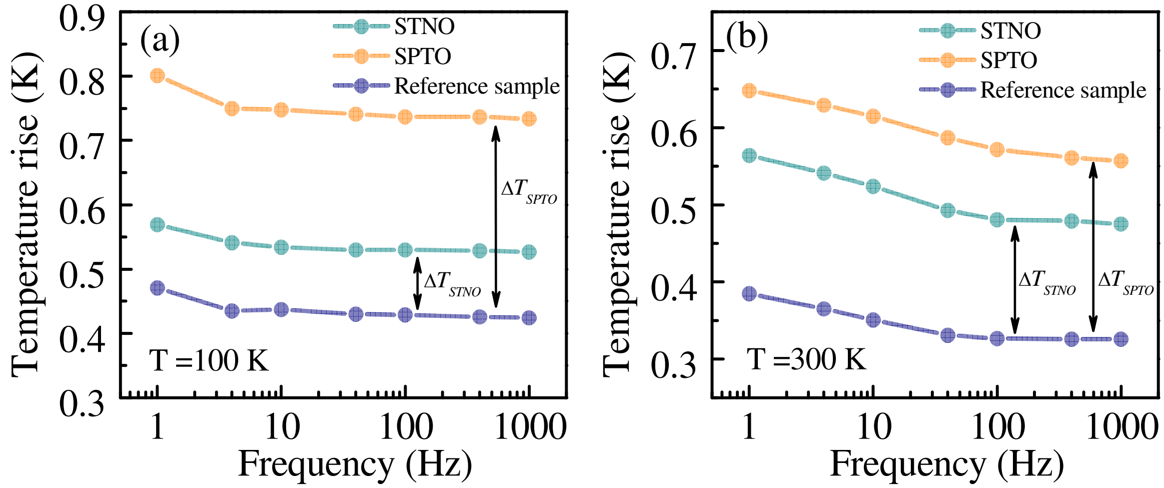

Figure S2. Temperature rise of the Au microheaters as a function of current frequency (ω) at (a) 100 K and (b) 300 K. ΔT_{SPTO} and ΔT_{STNO} are the frequency-independent temperature rises due to the presence of SPTO and STNO films, respectively.

For the anisotropic thermal properties of the films, the combination between w and t_f determines the measurement sensitivity to the in-plane and cross-plane heat flows in the film.[1] If w»t_f, the measured ΔT is mainly assumed to be sensitive to the cross-plane λ_f. On the other hand, if w~t_f, the heat generated by Au microheater spreads in the film, and the measured ΔT is influenced by both cross-plane and in-plane λ_f. Hence, by using a pair of Au microheaters with different widths and applying the two-dimensional heat flow model, the cross-plane and in-plane λ_f can be determined.[1] Due to the limited measurement capability of using the 'two-microheater' method, the cross-plane λ_f can be measured only.

2. Φ-scan XRD:

To verify the in-plane epitaxial relationship between the films/superlattices and LAO substrate, Φ scans were obtained. Figure S3 shows typical Φ scan XRD patterns for the off-axis {011} reflections from the films/superlattices and LAO. Four peaks with almost equal intensity spaced by 90º can be observed indicating that the films/superlattices and the LAO have four-fold symmetry about an axis normal to LAO. The peak positions of the Φ scan patterns indicate that the films/superlattices are in-plane textured and epitaxially aligned on LAO with epitaxial relationships of [100] STO ǁ [100] LAO and [001] STO ǁ [001] LAO.

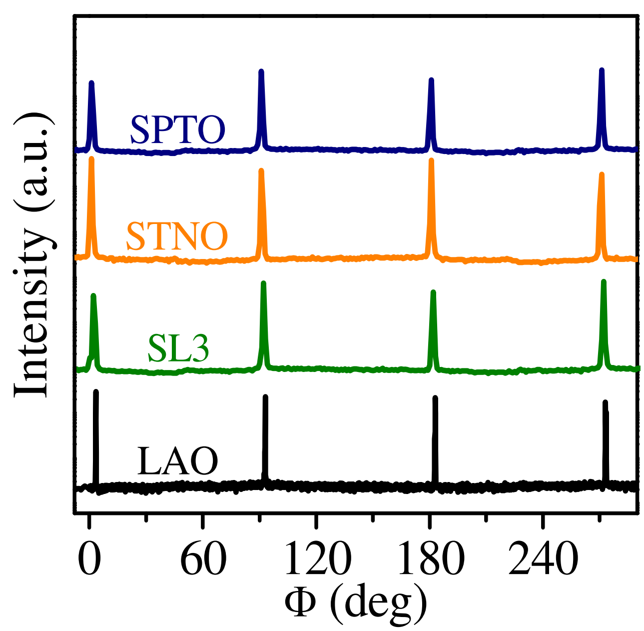

Figure S3. Φ-scan for {011} planes of SL3, SPTO, STNO samples and the substrate, showing four evenly spaced (90^0) peaks that confirm a four-fold symmetry about an axis normal to the substrate.

3. TEM:

TEM images were obtained for SL3 before and after annealing (Figure S4a and S4b, respectively). Both images show no difference caused due to annealing and the layer-by-layer epitaxial structure of the perovskite superlattices is preserved suggesting a high stability of these superlattices under a long-time annealing (~20 hrs) at high temperatures (~1000K).

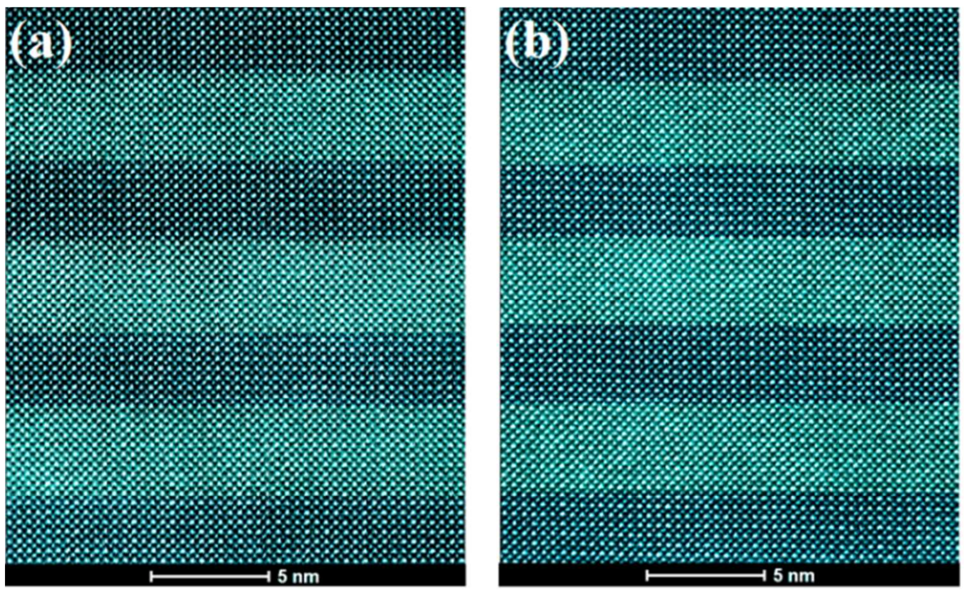

Figure S4. TEM images for (a) as-grown and (b) annealed SL3.

References:

1. Borca-Tasciuc, T.; Kumar, A. R.; Chen, G., *Review of Scientific Instruments* **2001,** *72* (4), 2139-2147.
2. Cahill, D. G., *Review of Scientific Instruments* **1990,** *61* (2), 802-808.
3. Lee, S.-M.; Cahill, D. G., *Journal of Applied Physics* **1997,** *81* (6), 2590-2595.