Supporting information

Stable cubic crystal structure and optimized thermoelectric performance of $SrTiO_3$ based ceramics driven by entropy engineering

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Calculation of Debye Temperature

The Debye temperature θ_D, represents the characteristic energy scale of the crystal lattice, which can be obtained by the following equation:

$$\theta_D = \frac{h}{k_B}\left(\frac{6\pi^2 N}{V}\right)^{1/3} v_D \qquad \text{* MERGEFORMAT (S1)}$$

$$\frac{1}{v_D^3} = \frac{1}{3}\left(\frac{1}{v_L^3} + \frac{2}{v_S^3}\right) \qquad \text{* MERGEFORMAT (S2)}$$

where h, k_B, N, V, v_D, v_L and v_S are Planck's constant, Boltzmann constant, number of atoms per unit volume, volume of the solid, mean velocity of sound, longitudinal and transverse sound velocity respectively. The longitudinal and transverse sound velocity are measured by ultrasonic pulse transmitter receiver(OLYMPUS 5073PR).

Fitting calculation of specific heat equation

A Scaematic drawing of C_p from 0 K to the sintering temperature is given in **Figure S1**.At lower temperatures, the variation of specific heat at constant pressure with temperature satisfies Debye's T^3 law approximately,as shown in the following equation:

$$C_p \approx C_v = \frac{12}{5}\pi^4 N_A k_B \left(\frac{T}{\theta_D}\right)^3 \quad \text{* MERGEFORMAT (S2)}$$

where N_A, k_B and θ_D are Avogadro's constant, Boltzmann constant and Debye temperature respectively. At higher temperatures, the $C_p(T)$ were obtained by least squares fitting of the equation $C_p = a + bT + cT^{-2} + dT^2$.

The calculation of disorder scattering parameter

The disorder scattering parameter (Γ_{total}) is depended on the mass fluctuation (Γ_m) and strain field term (Γ_s), as given by $\Gamma_{total} = \Gamma_m + \Gamma_s$.The mass fluctuation ($\Gamma_m$) and strain field term (Γ_s) parameter are then given by

$$\Gamma_m = \frac{\sum_{i=1}^{n} c_i \left(\frac{\bar{M}_i}{\bar{\bar{M}}}\right)^2 f_i^1 f_i^2 \left(\frac{M_i^1 - M_i^2}{\bar{r}_i}\right)}{\left(\sum_{i=1}^{n} c_i\right)} \text{ and } \Gamma_s = \frac{\sum_{i=1}^{n} c_i \left(\frac{\bar{M}_i}{\bar{\bar{M}}}\right)^2 f_i^1 f_i^2 \varepsilon \left(\frac{r_i^1 - r_i^2}{\bar{r}_i}\right)}{\left(\sum_{i=1}^{n} c_i\right)},$$

where $\bar{M}_i$ is the average atomic mass of the i of sublattice, the $\bar{\bar{M}}$ is the average atomic mass of the compound, f_i is the fractional occupant, r_i is the radius of atom, ε is the phenomenological parameter which is a function of the Grüneisen parameter. The mass fluctuation term and the strain field term would be jointly determined by four or five parameters of $\bar{M}_i$, $\bar{\bar{M}}$, f_i, r_i, ε, respectively.

Table S1. The values of a, b, c, d, ΔH_{298}^{θ}, v_L, v_S and θ_D of $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ samples.

Component x	a	b	c	d	ΔH_{298}^{θ} (kJ / mol)	v_L (m/s)	v_S (m/s)	θ_D (K)
x = 0.1	96.21	37.99	-0.39	15.35	-1555.47	6769	4235	595
x = 0.2	88.65	50.07	-0.36	7.38	-1562.42	6676	4111	576
x = 0.3	89.23	48.67	-0.36	15.72	-1569.36	6667	4455	617
x = 0.4	88.62	69.50	-0.36	-11.32	-1576.28	6417	3972	551
x = 0.5	96.52	47.42	-0.39	12.03	-1583.19	6304	4238	577

Table S2. Lattice constant (a), experimental density (ρ_1) and relative density (ρ_2) of $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ samples.

Component x	a (Å)	ρ_1 (g/cm^3)	ρ_2 (%)
x = 0.1	3.9290	5.171	95.3
x = 0.2	3.9341	5.228	94.4
x = 0.3	3.9491	5.333	95.0
x = 0.4	3.9593	5.355	93.9
x = 0.5	3.9664	5.310	91.5

The relative density is calculated by

$$\text{Relative density} = \frac{\text{Experimental density}}{\text{Theoretical density}} \times 100\% .$$

As listed in **Table S2**, experimental densities of $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ fluctuate around 5.2-5.3g/cm^3. And since the mass of Ba is much higher than that of Sr, the theoretical density of Ba doped samples increases. Finally, the relative density shows a decreasing trend with the increase of Ba amount.

Table S3. Parameters for calculation of lattice thermal conductivity of $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ through Debye-Callaway model. D is the average grain size obtained by Nano Measurer.

Component x	θ_D (K)	v_D (m/s)	D (μm)	A (s^3)	B ($s \cdot K^{-1}$)	C (s)	β
$x = 0.1$	595	4664	0.69	2.19×10^{-42}	1.22×10^{-19}	1.0×10^{-18}	11
$x = 0.2$	576	4535	0.8	2.44×10^{-42}	1.45×10^{-19}	1.0×10^{-18}	10.9
$x = 0.3$	617	4869	0.84	1.85×10^{-42}	6.58×10^{-20}	7.7×10^{-17}	26
$x = 0.4$	551	4380	0.105	2.64×10^{-42}	1.52×10^{-19}	8.0×10^{-17}	10.5
$x = 0.5$	577	4628	0.148	2.04×10^{-42}	7.23×10^{-20}	1.1×10^{-16}	32.1

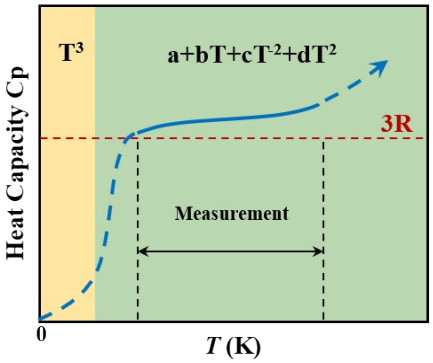

Fig. S1 Schematic drawing of the specific heat capacity of an ideal model from 0 K to the sintering temperature with no phase transition over the whole temperature range.

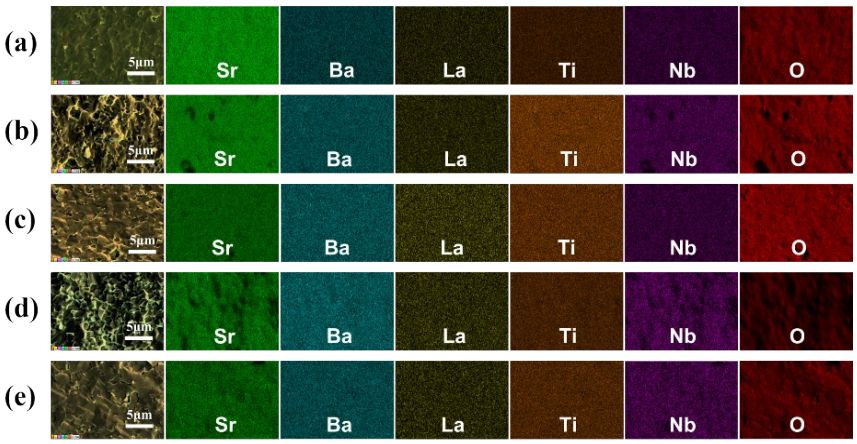

Fig. S2 The EDS mapping of $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ samples: (a) x = 0.1; (b) x = 0.2; (c) x = 0.3; (d) x = 0.4; (e) x = 0.5.

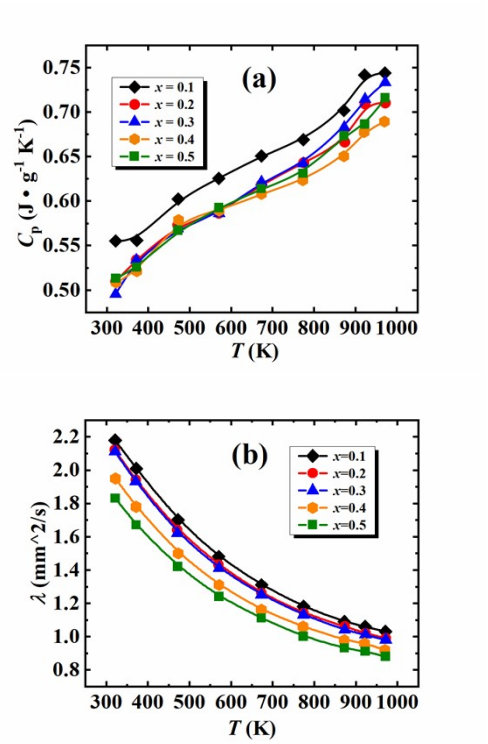

Fig. S3 Temperature dependence of the thermal transport properties for $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ samples: (a) specific heat capacity; (b) thermal diffusion coefficient.

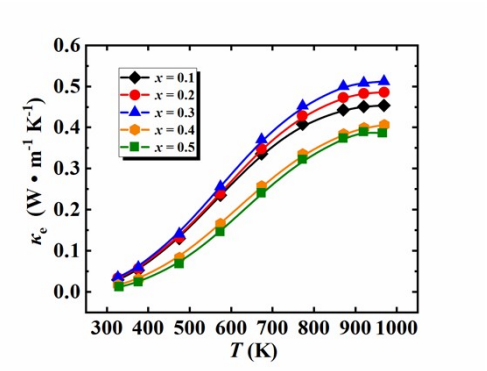

Fig. S4 Temperature dependence of electronic thermal conductivity for $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ samples.

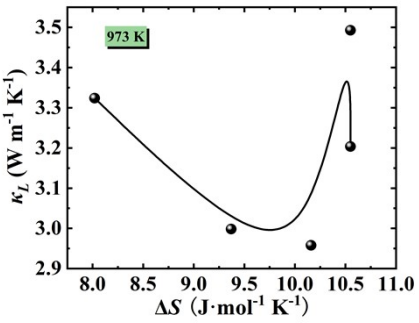

Fig. S5 Lattice thermal conductivity depended on configurational entropy for $Sr_{0.9-x}Ba_xLa_{0.1}Ti_{0.9}Nb_{0.1}O_{3-\delta}$ samples (x = 0.1, 0.2, 0.3, 0.4, 0.5) at 973 K.