# Ultralow Thermal Conductivity and High Thermoelectric Performance Induced by Multiscale lattice Defects in Cu-doped BST Alloy

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## I. Sample preparation.

$BiSbTe_3$ samples were prepared using a KCl solvent method combined with a melt quenching technique. The samples were designated as $BST(KCl)_{3.5}$, $BST(KCl)_5$, $BST(KCl)_{6.5}$, $BST(KCl)_8$, and $BST(KCl)_{9.5}$, corresponding to different stoichiometric ratios of $BiSbTe_3(KCl)_x$ ($x$ = 3.5, 5, 6.5, 8, and 9.5), respectively. Among these samples, $BST(KCl)_{3.5}$ was selected as the base sample for further experiments. Cu doping experiments were performed on the $BST(KCl)_{3.5}$ sample using various doping concentrations denoted as $Cu_xBi_{1-x}SbTe_3{\cdot}KCl_{3.5}$ ($x$ = 0.01, 0.015, 0.02, 0.025, and 0.03), and labeled as S1, S2, S3, S4, and S5, respectively. Comprehensive investigations of the thermoelectric properties were conducted on all the samples. The raw materials were sealed in a vacuum quartz tube, and prepared according to the temperature gradient shown in **Fig.S1**. Ultimately, the lustrous crystals were obtained by dissolving KCl in distilled water.

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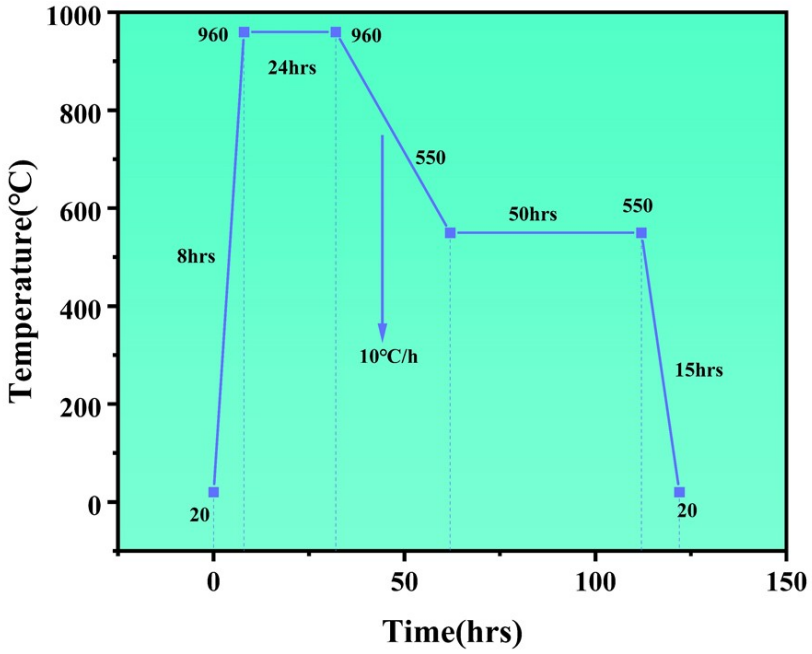


**Fig. S1.** The temperature gradient of sample preparation.

**II. Structure Characterization.**

The phase constitutions of the obtained samples were analyzed via powder X-ray diffraction (XRD, Ultima IV) with Cu Kα radiation (λ = 1.5403 Å). The microstructures and elemental compositions of the samples were obtained by a field emission scanning electron microscope (SEM, FEI Inspect F50) equipped with the energy dispersive X-ray spectroscopy (EDX, Oxford Inca X-act). The accurate composition of elements in the samples was determined by electron probe microanalysis (EPMA, 1720H). The detailed microstructures of the crystal were investigated by transmission electron microscopy (TEM, Tecnai G20) and selected electron diffraction (SAED). The Seebeck coefficient and electrical resistivity were simultaneously measured on bars of ~3 × 3 × 10 $mm^3$ by ZEM-3. The thermal diffusivity is tested by the laser flash technique (LFA 457, Netzsch) at 300-550 K under argon flow. Thermogravimetry–differential scanning calorimetry (TA Q20) was used to measure the $C_p$ of samples. $\rho$ was measured via

Archimedes' method. The Hall coefficient was tested using the Hall tester (HL5500) under a magnetic field of 0.5 T at room temperature.

**III. Table S1: Hall coefficient $R_H$, effective mass $m^*$ of the BST samples at room temperature and the density $\rho$**

| Samples | $R_H$ cm$^3$/C | m*/m$_0$ | $\rho$ (g·cm$^{-3}$) |
|---|---|---|---|
| **Pure-BST** | **0.162** | **1.17** | **6.251** |
| **BST(KCl)$_{3.5}$** | **0.161** | **1.34** | **6.323** |
| **BST(KCl)$_5$** | **0.153** | **1.33** | **6.322** |
| **BST(KCl)$_{6.5}$** | **0.137** | **1.32** | **6.322** |
| **BST(KCl)$_8$** | **0.123** | **1.32** | **6.321** |
| **BST(KCl)$_{9.5}$** | **0.112** | **1.29** | **6.318** |
| **S1** | **0.162** | **1.47** | **6.335** |
| **S2** | **0.147** | **1.46** | **6.334** |
| **S3** | **0.121** | **1.45** | **6.337** |
| **S4** | **0.098** | **1.45** | **6.336** |
| **S5** | **0.078** | **1.44** | **6.338** |

## IV. Thermal diffusivity, specific heat $C_p$ and Electronic thermal conductivity for the samples.

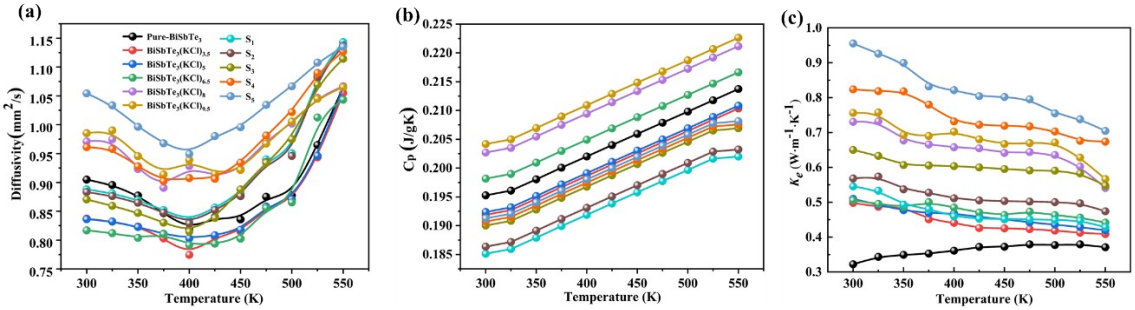


**Fig. S2.** (a) Thermal diffusivity for the BST samples; (b) The specific heat $C_p$ for the BST samples; (c) Electronic thermal conductivity for the BST samples.

## V. Band structure.

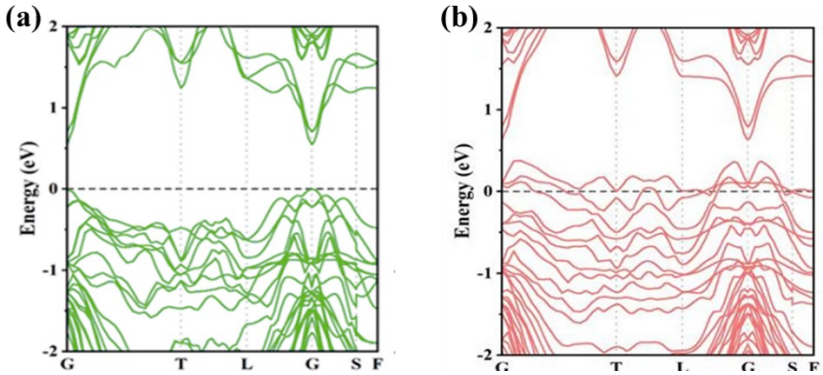


**Fig. S3.** Band structure (a)Pure-$BiSbTe_3$; (b) $BiSbTe_3$ with Cu-doped.