

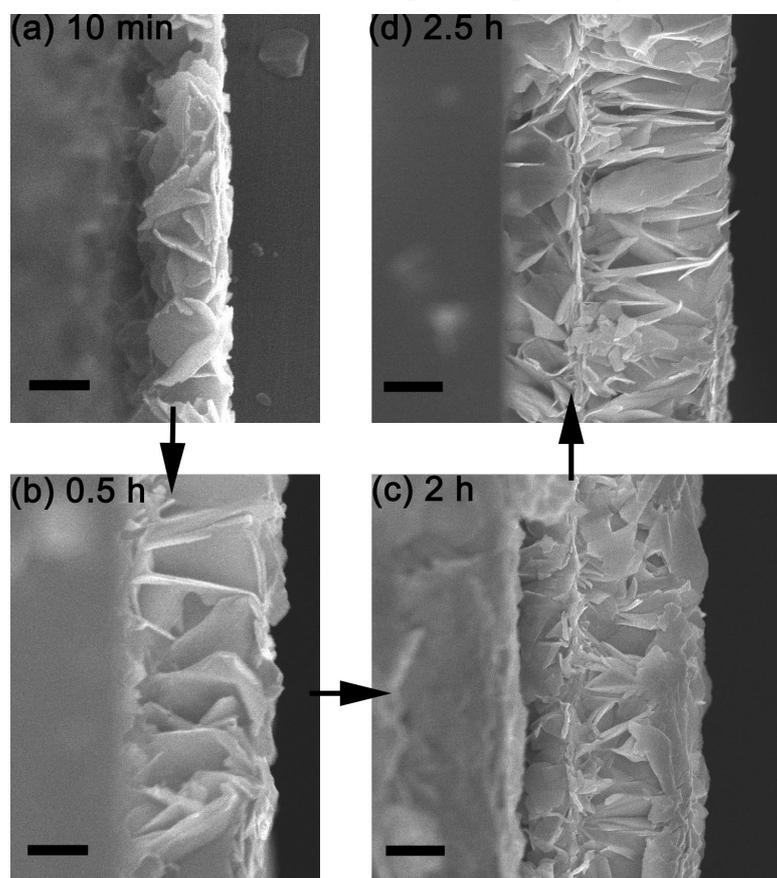
## Enhancing Thermoelectric Performance of Bismuth Selenide Films via Constructing Double-layer Nanostructure

Zhengliang Sun<sup>a,b</sup>, Shengcong Liufu<sup>a,\*</sup>, Xihong Chen<sup>a</sup>, Lidong Chen<sup>a</sup>

CAS Key laboratory of Materials for Energy Conversion, Shanghai Institute of Ceramics, Chinese Academy of Science, 1295 Dingxi Road, Shanghai 200050, PR China.

E-mail address: scliu@hotmail.com

**Figure S1** cross-sectional SEM micrographs of Bi<sub>2</sub>Se<sub>3</sub> films prepared with different reaction times, the scale bars in the figure are 250, 250, 500, 500 nm for (a) 10 min, (b) 0.5 h, (c) 2 h and (d) 2.5 h samples, respectively.



As be indicated in the Figure S1, the 10 min and 0.5 h sample displayed only one layer, combined with the XRD for 0.5 h in Figure 1 in the text, the layer is mainly (001) orientation. On the other hand, the 2 h sample displayed two layer, and in the XRD pattern for 2 h sample, (110) peak turns much stronger than that of 0.5 h sample. This result indicated there is an orientation transformation during the films growth, and the transformation is consistent with the layer structure change. When the reaction time proceeds to 2.5 h, the outer layer became thicker compared with the 2 h

sample, correspondingly the (110) peak became stronger. Based on the XRD results and SEM results, it is reasonable to state that the DLTFs are consisted of (001) orientational inner layer and (110) orientational outer layer.

**The experimental details about how thermoelectric properties were measured:**

The dark electrical resistivity of  $\text{Bi}_2\text{Se}_3$  films was measured by a four-point probe method in van der Pauw configuration with an Accent HL5500 Hall System at room temperature. Silver paste was applied to provide ohmic contact with  $\text{Bi}_2\text{Se}_3$  films. For the Seebeck coefficient measurement, two Pt-Pt/Rh thermocouples were attached to both ends of  $\text{Bi}_2\text{Se}_3$  thin films, and temperature gradients in 3-5 K were generated by a film heater at one end, the Seebeck coefficient was then obtained from the slop of thermoelectromotive force to the temperature difference.