

Fig.S1 EPMA mapping of three elements on polished InSeSn<sub>0.01</sub> surface (a) In, (b)Se, (c) Sn and (d) Energy dispersive x-ray spectrum.



Fig.S2 XPS spectra of (a) In 3d, (b) Se3d, and (c) Sn 3d for InSeSn<sub>x</sub>.



Fig.S3 Total thermal conductivities of  $InSeSn_x$  in  $C \perp (x=0, 0.005, 0.01, 0.02)$  as a function of temperature.



Fig.S4 Total thermal conductivities of  $InSeSn_x$  in  $C_{//}$  (x=0, 0.005, 0.01, 0.02) as a function of temperature.

Table S1 The chemical compositions (relative molars) identified using EPMA for  $InSeSn_x$  (*x*=0, 0.01) (taken from mappings)

Compounds	In	Sn	Se
InSe	1.04		0.96
InSeSn <sub>0.01</sub>	1.0	0.02	0.99

Compounds	In3d (eV)	Se3 <i>d</i> (eV)	Sn3 <i>d</i> (eV)
InSe	444.53	Peak 1: 53.85	
		Peak 2: 54.47	
InSeSn <sub>0.005</sub>	444.44	Peak 1: 53.75	486.29
		Peak 2: 54.43	
InSeSn <sub>0.01</sub>	444.73	Peak 1: 53.99	485.95
		Peak 2: 54.59	
InSeSn <sub>0.02</sub>	444.66	Peak 1:53.88	195 90
		Peak 2: 54.59	483.80

Table S2 Binding energies from the  $In3d_{5/2}$ ,  $Sn3d_{5/2}$  and Se3d core-level photoelectron spectra for the  $InSeSn_x$  (*x*=0,0.005,0.01,0.02) compounds.