High Seebeck Coefficient and Unusually Low Thermal Conductivity Near Ambient Temperatures in Layered Compound $Yb_{2-x}Eu_xCdSb_2$

Supporting Information

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Figures

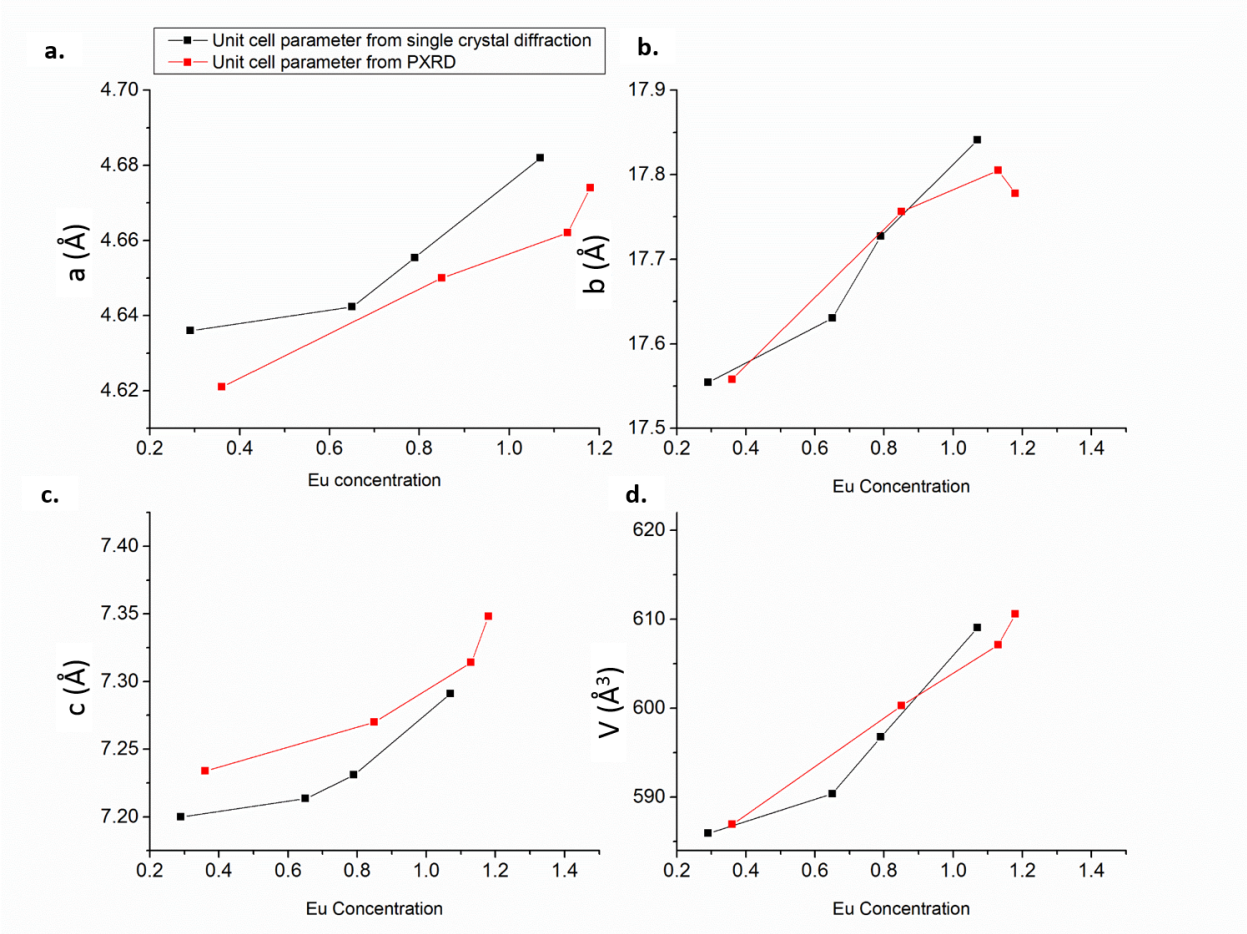

Figure S1: Unit cell parameters (**a.**, **b.** and **c.**) and unit cell volume from X-ray diffraction (**d.**) vs. Eu concentration reported for single crystal $Yb_{2-x}Eu_xCdSb_2$, x = 0.29, 0.65, 0.79, 1.07, (black, ref 21) and powder samples reported herein, x = 0.36, 0.85, 1.13, 1.18, (red, this publication).

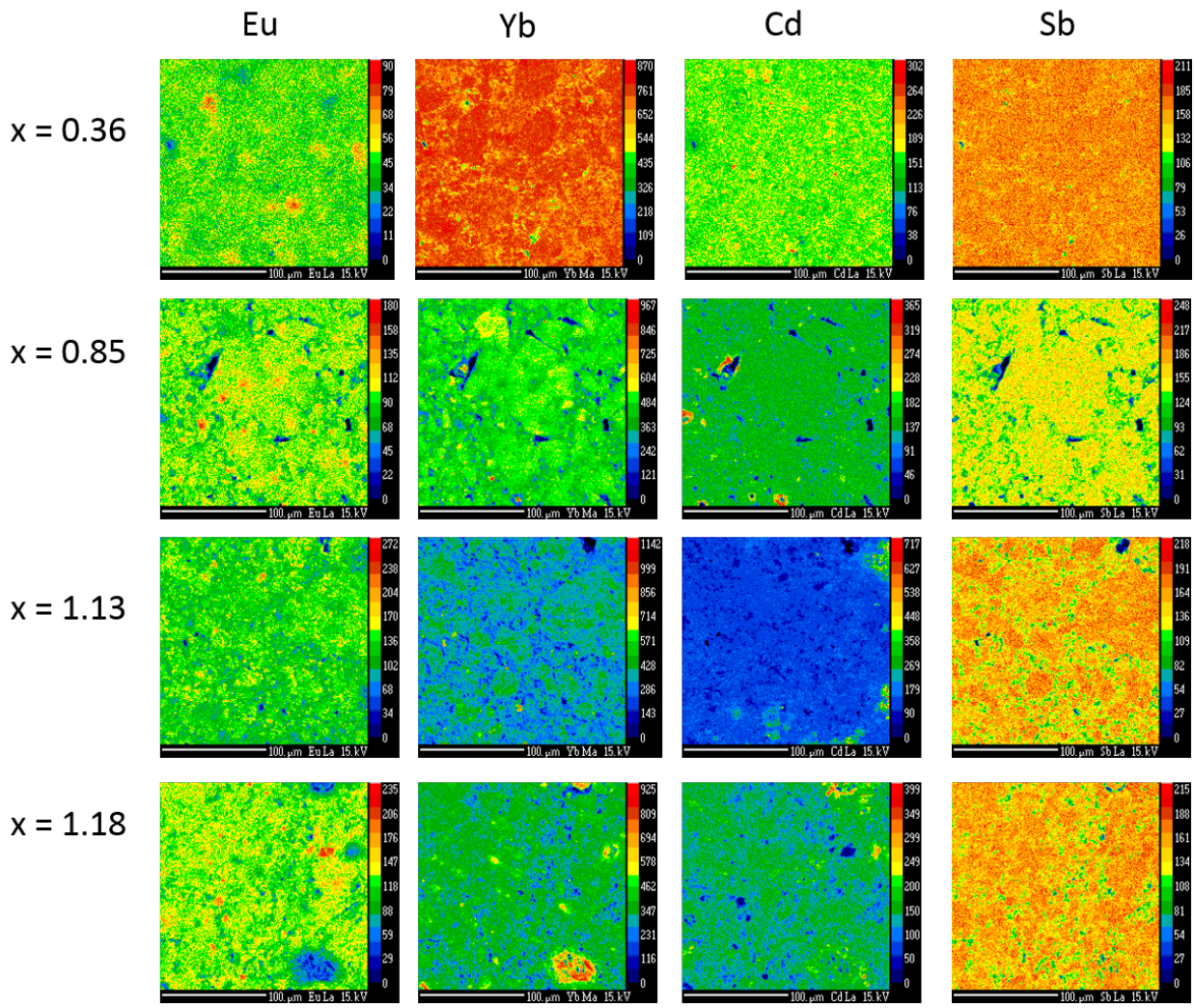

Figure S2: Electron microprobe analysis (EMPA) images for $Yb_{2-x}Eu_xCdSb_2$ samples, x = 0.36, 0.85, 1.13 and 1.18. Colors show element homogeneity over the interrogated region. Scale bar indicates 100 μm.

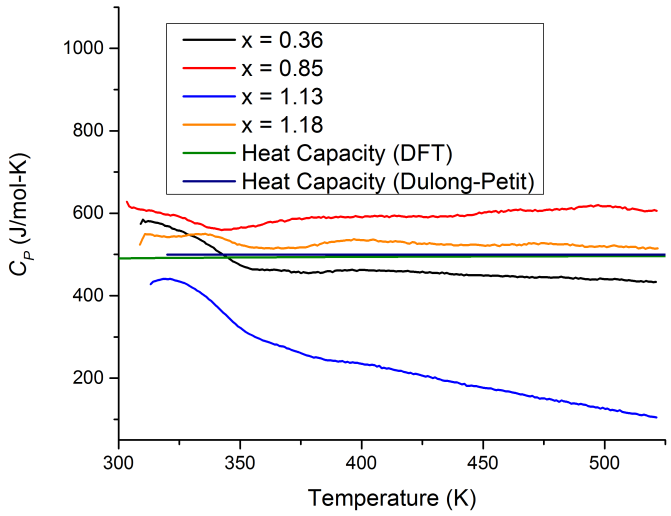

Figure S3: Measured heat capacity values along with DFT calculated heat capacity (green) and Dulong-Petit estimated heat capacity (navy). For conversion of thermal diffusivity to thermal conductivity for presented samples, the Dulong-Petit approximation was used.

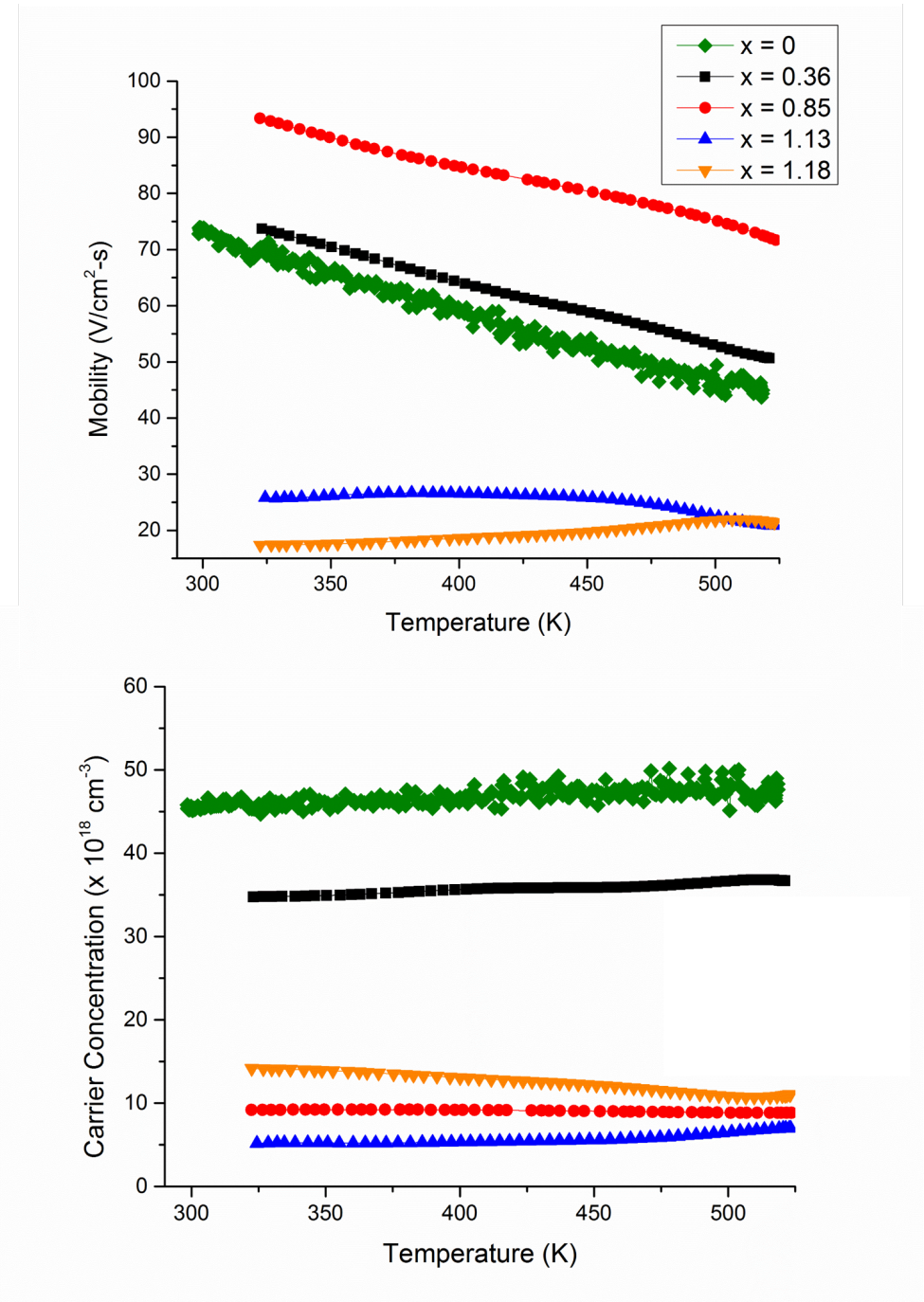

Figure S4: Hall mobility (top) and carrier concentration (bottom) for $Yb_{2-x}Eu_xCdSb_2$.

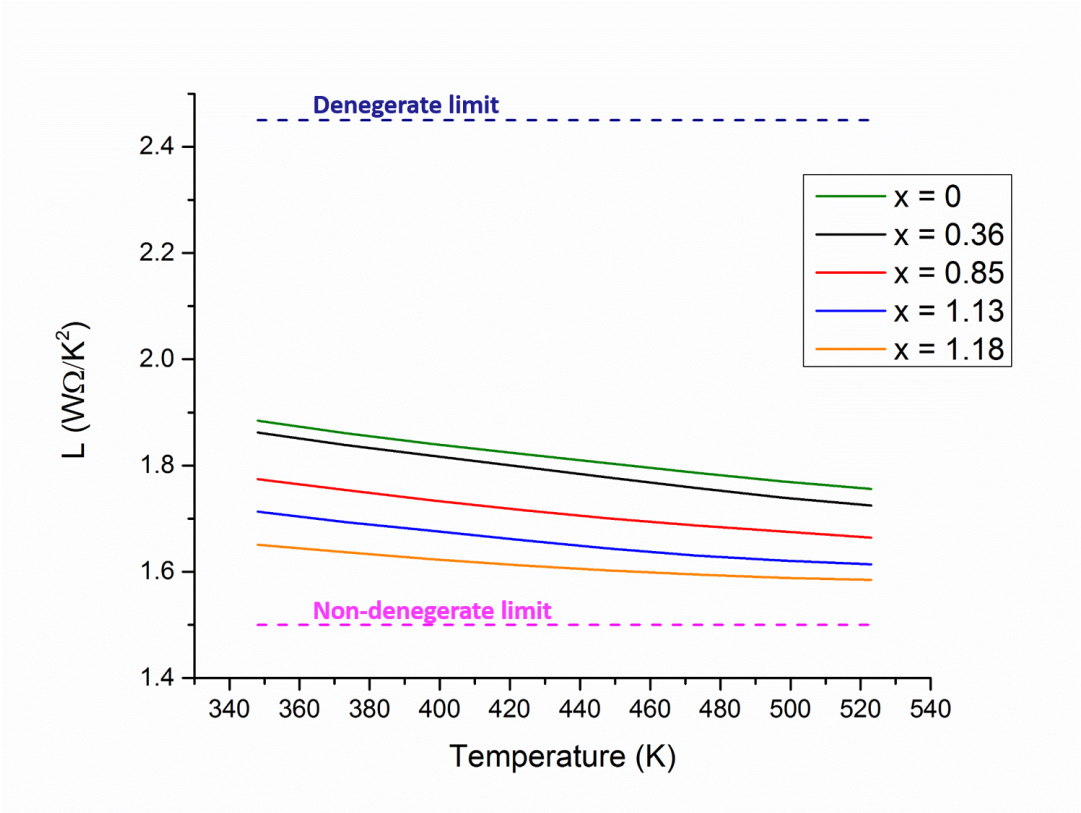

Figure S5: Lorenz number values for each composition, $Yb_{2-x}Eu_xCdSb_2$, as calculated from $L = 1.5 + exp\left(\frac{-S}{116}\right)$.

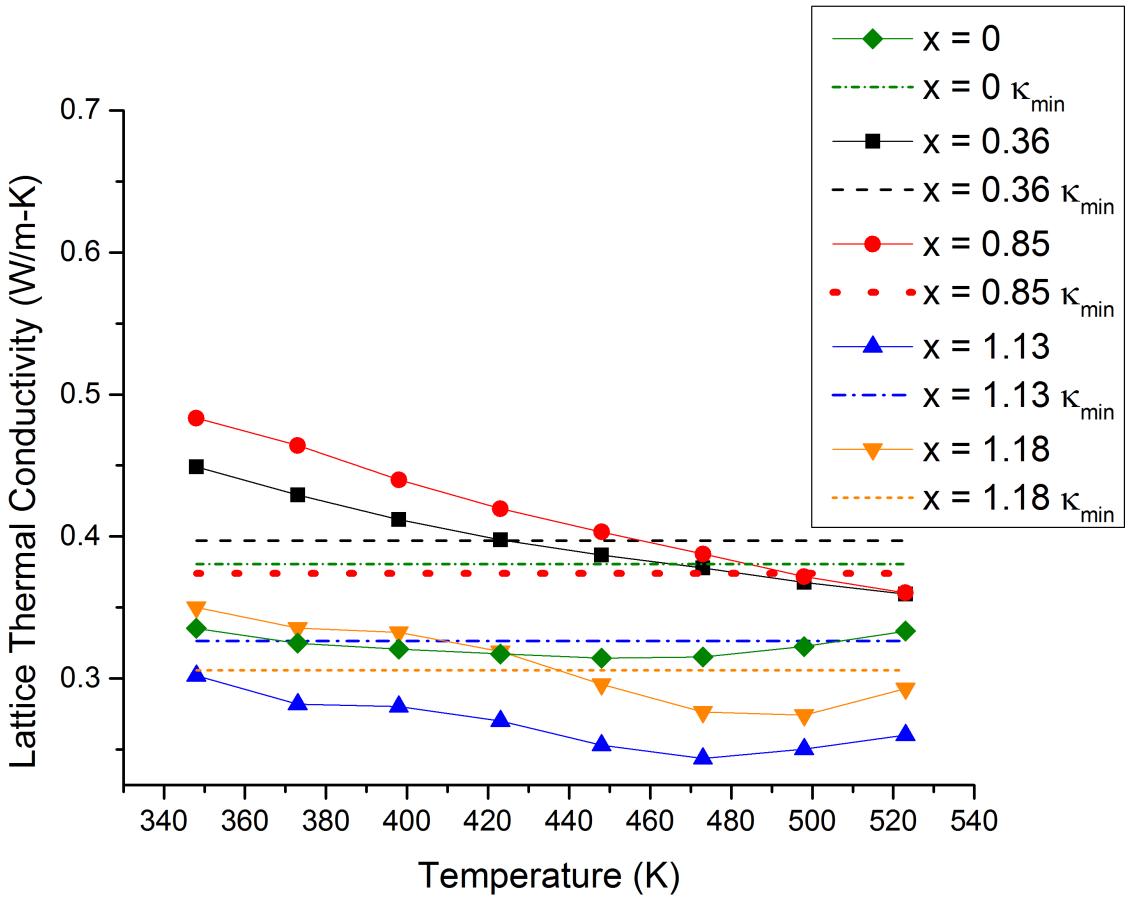

Figure S6: Lattice thermal conductivity for $Yb_{2-x}Eu_xCdSb_2$. Symbols indicate calculated κ_L values and dashed horizontal lines indicate κ_{min} values calculated for each sample based on its unit cell parameters and speed of sound values.

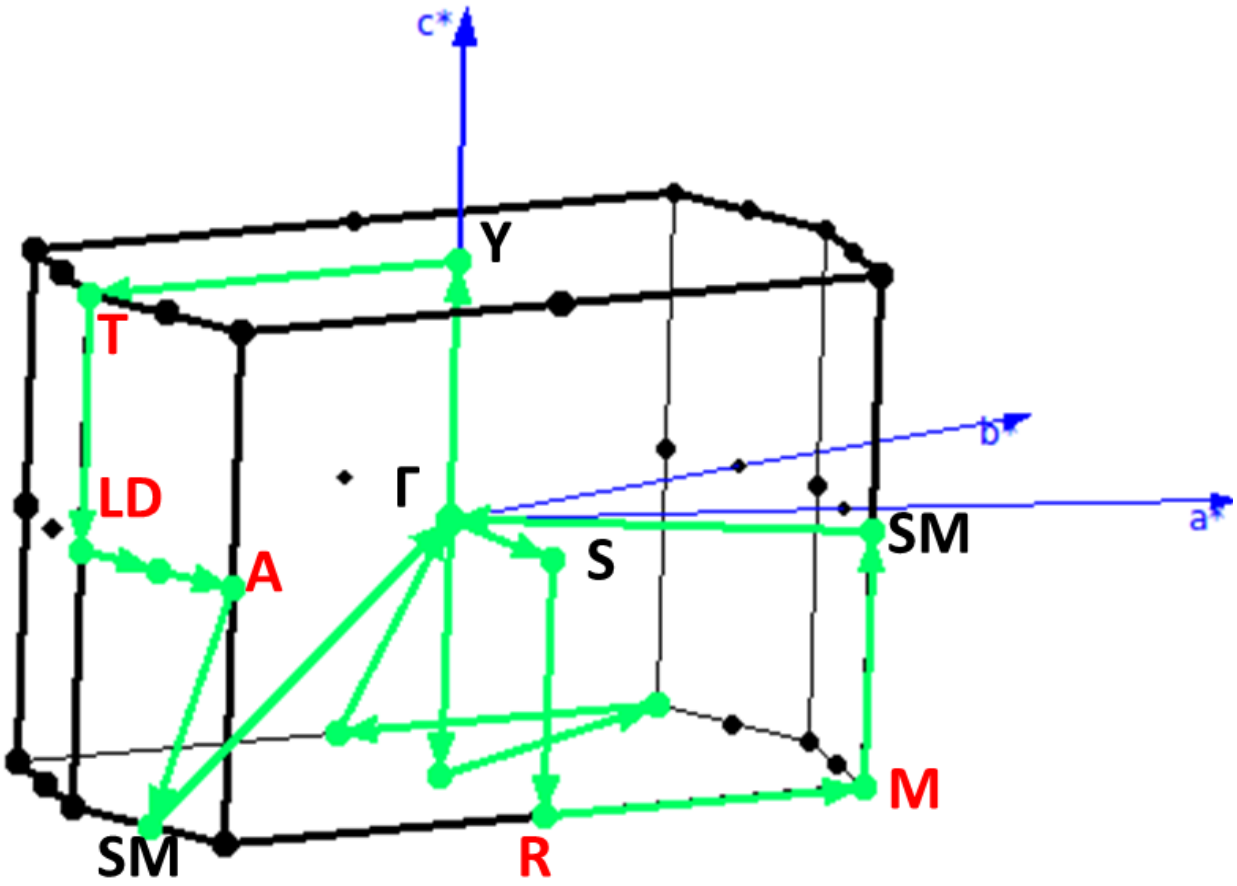

Figure S7: K path used for band plot and phonon dispersion black labels are showing major symmetry points (showing in manuscript) and minor symmetry points (not labeled in plots), marked in red.

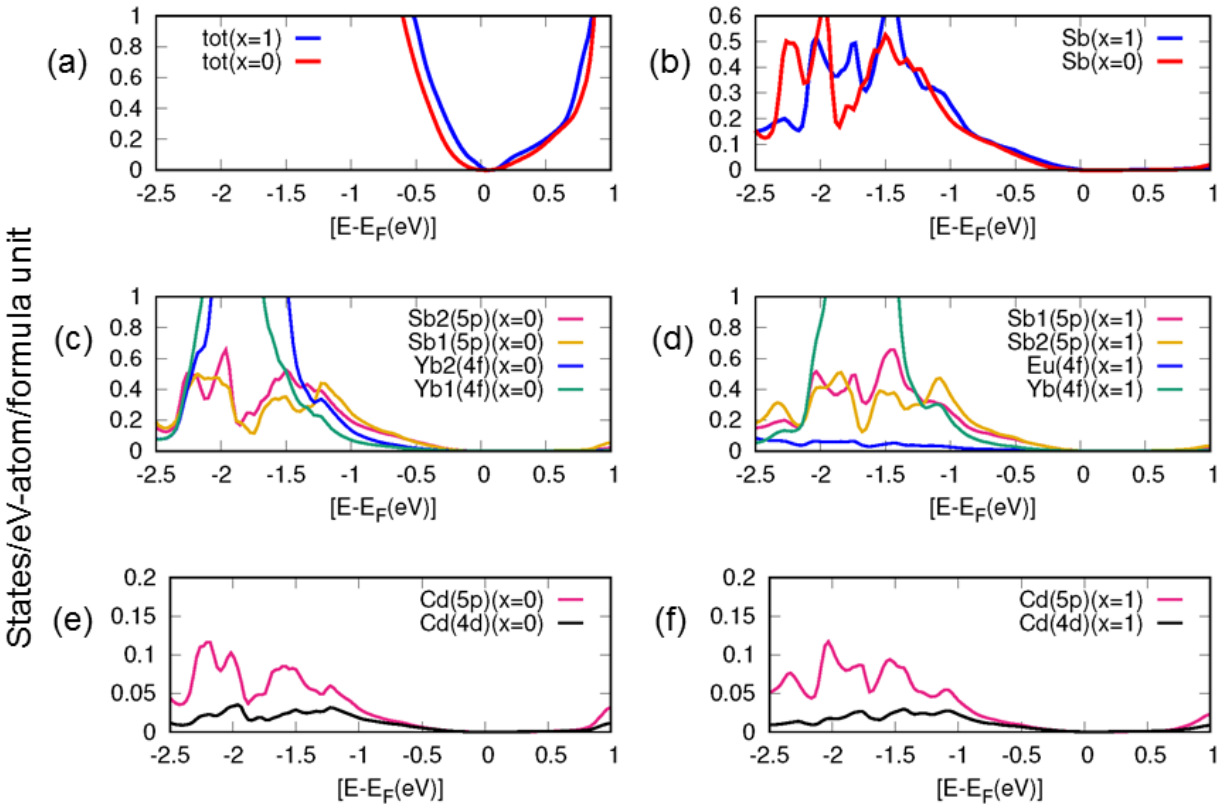

Figure S8: Orbital-projected density of states plots of $Yb_{2-x}EuCdSb_2$ (x = 0, 1) using the DFT+U. Valence band overlaps are shown, (a) total PDOS, (b) Sb (p), (c), (d) with two inequivalent Sb (p) and Yb (f) and Eu(f) orbitals, and (e), (f) Cd (p) (d) orbitals. Majority spin channel have been shown in this plot

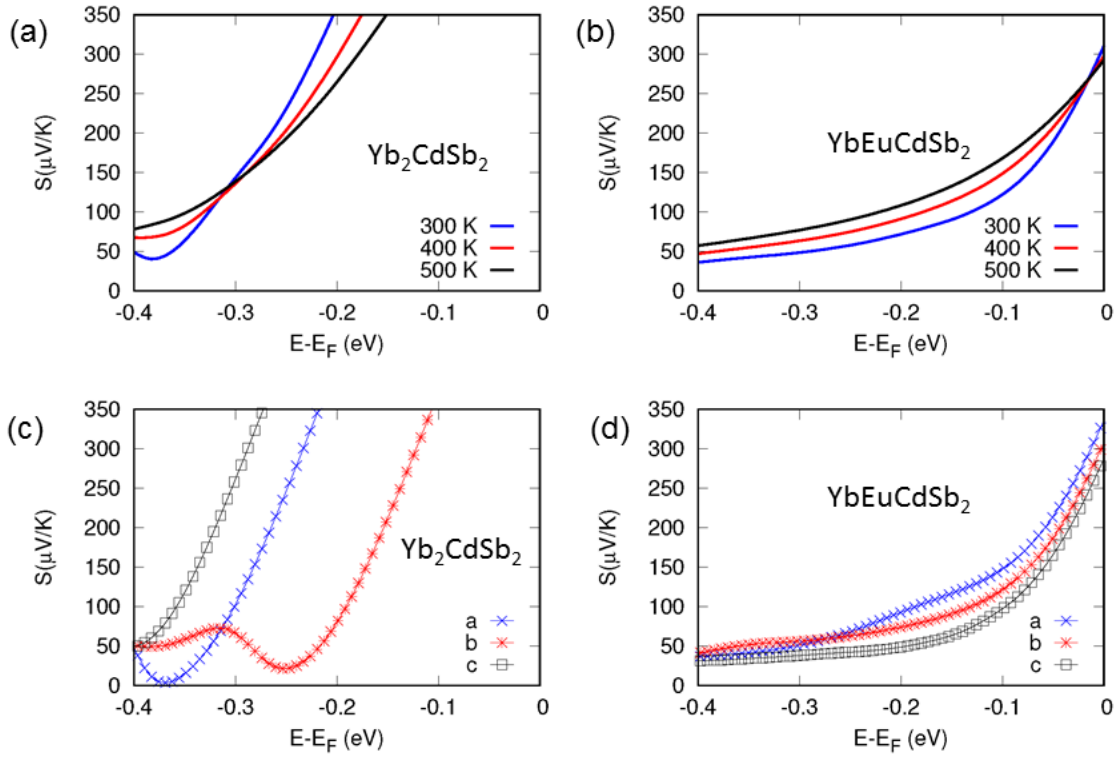

Figure S9: Seebeck coefficient with respect to chemical potential Seebeck coefficient in different temperature (a)Yb_2CdSb_2 and (b) $YbEuCdSb_2$ over a broader chemical potential range (-0.4 eV to 0.4 eV) for (c) Yb_2CdSb_2 , (d) $YbEuCdSb_2$ and Seebeck coefficient along different crystallographic axes

Table S1: Comparison of experimental single crystal (collected at 120 K) and calculated optimized lattice parameters for Yb_2CdSb_2 (x = 0) and $YbEuCdSb_2$ (x = 1). For DFT calculations crystallographic coordinated obtained from Ref. a is used for Yb_2CdSb_2 and coordinates from [ref b] had been used for $Yb_{2-x}Eu_xCdSb_2$

$Yb_{2-x}Eu_xCdSb_2$	Experimental (Å)	Calculated (Å)
x = 0 [a]		
a	4.6184 (17)	4.66
b	17.418(6)	17.49
c	7.178(3)	7.19
x = 1[b]		
a	4.682(2)	4.72
b	17.841(8)	17.89
c	7.291(3)	7.34

[a] S.-Q. Xia and S. Bobev, *Journal of the American Chemical Society*, 2007, **129**, 4049-4057.

[b] B. Saparov, M. Saito and S. Bobev, *Journal of Solid State Chemistry*, 2011, **184**, 432-440.

Table S2: Speed of sound data for all samples.

Eu Concentration (x)	v_s (m/s)	v_L (m/s)
0	1735	3000
0.36	1820	3150
0.85	1750	2990
1.13	1530	2620
1.18	1400	2570

Table S3: Measured thermal diffusivity values for $Yb_{2-x}Eu_xCdSb_2$.

Temperature	Thermal Diffusivity (mm^2/s)	Temperature	Thermal Diffusivity (mm^2/s)
x = 0.36		**x = 0.85**	
74.7	0.525	71.8	0.456
100	0.513	99.1	0.442
124.9	0.504	122	0.427
149.4	0.491	146.6	0.413
174.9	0.48	173.3	0.401
199.8	0.466	198.7	0.389
225.4	0.455	225	0.375
250.4	0.445	250.7	0.365
x = 1.13		**x = 1.18**	
72.9	0.254	73.9	0.294
99.3	0.234	98.7	0.281
122.1	0.237	123.4	0.282
146.4	0.223	146.1	0.27
173	0.219	172.7	0.253
201.7	0.203	199.2	0.237
224.6	0.214	224.5	0.234
250.5	0.221	250	0.25

Table S4: Optimized Cartesian coordinates for Yb_2CdSb_2

	x	y	z
Yb	2.3866	0.3563	5.3033
Yb	2.3866	17.5877	1.6140
Yb	2.3866	14.4471	3.8641
Yb	2.3866	3.4969	0.1749
Yb	0.0000	9.3283	5.3033
Yb	0.0000	8.6157	1.6140
Yb	0.0000	5.4751	3.8641
Yb	0.0000	12.4689	0.1749
Cd	0.0000	1.8446	2.9132
Cd	0.0000	16.0994	6.6025
Cd	2.3866	10.8166	2.9132
Cd	2.3866	7.1274	6.6025
Sb	2.3866	3.2531	3.8796
Sb	2.3866	14.6909	0.1903
Sb	0.0000	16.7340	3.6485
Sb	0.0000	1.2100	7.3377
Sb	0.0000	12.2251	3.8796
Sb	0.0000	5.7189	0.1903
Sb	2.3866	7.7620	3.6485
Sb	2.3866	10.1820	7.3377

Table S5: Optimized Cartesian coordinates for $YbEuCdSb_2$

	x	y	z
Yb	2.6950	0.4151	5.9830
Yb	2.6950	19.8009	1.8249
Yb	2.6950	17.1831	4.9377
Yb	2.6950	3.0328	0.7796
Eu	0.0000	10.5230	5.9830
Eu	0.0000	9.6929	1.8249
Eu	0.0000	7.0752	4.9377
Eu	0.0000	13.1408	0.7796
Cd	0.0000	1.4809	3.1277
Cd	0.0000	18.7351	7.2859
Cd	2.6950	11.5888	3.1277
Cd	2.6950	8.6271	7.2859
Sb	2.6950	2.6970	3.7797
Sb	2.6950	17.5189	7.9379
Sb	0.0000	18.9291	4.2725

Sb	0.0000	1.2868	0.1144
Sb	0.0000	12.8050	3.7797
Sb	0.0000	7.4110	7.9379
Sb	2.6950	8.8211	4.2725
Sb	2.6950	11.3948	0.1144