Supporting Information

Extremely Low Thermal Conductivity and High Thermoelectric Performance in Liquid-like $Cu_2Se_{1-x}S_x$ Polymorph Materials

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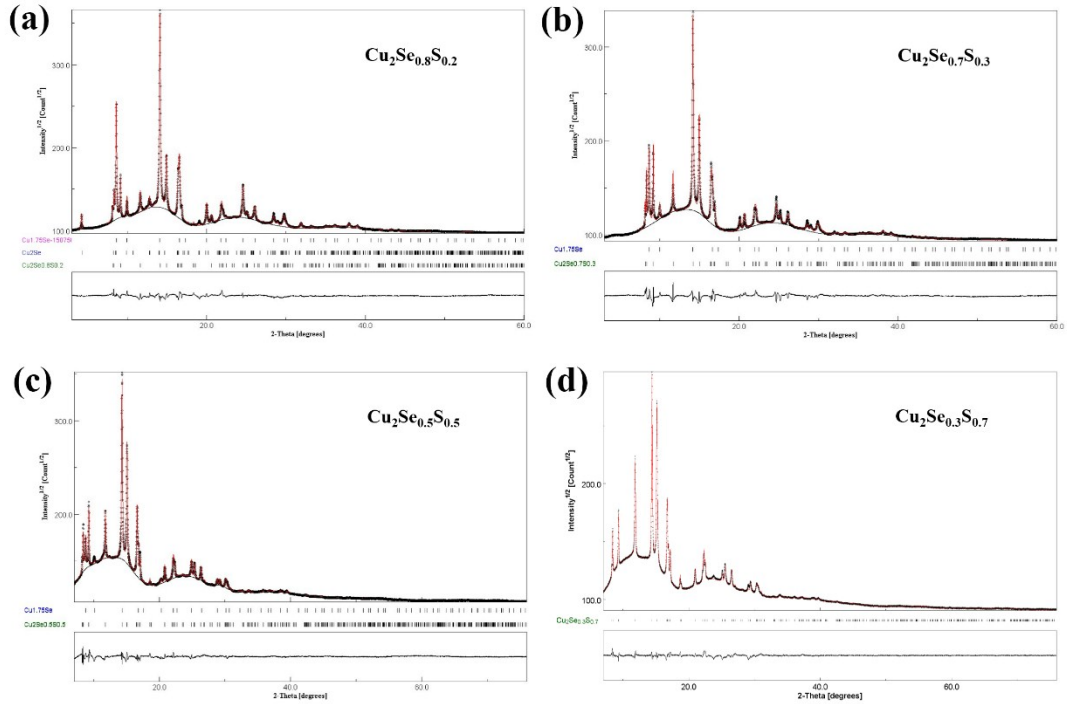

Fig. S1. Synchrotron Rietveld refinements for $Cu_2Se_{1-x}S_x$ (x = 0.2, 0.3, 0.5 and 0.7) samples.

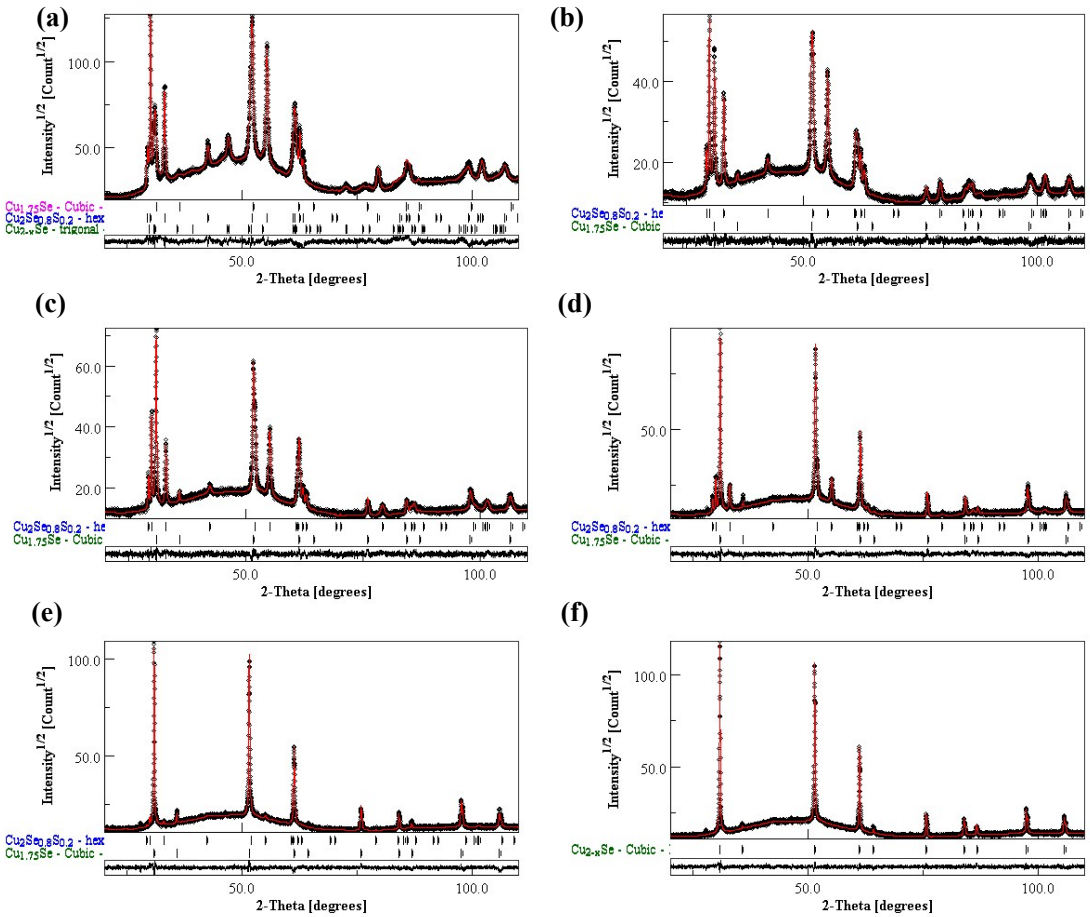

Fig. S2. Rietveld refinements for PXRD with Co-K_α source for $Cu_2Se_{0.8}S_{0.2}$, at **(a)** RT, **(b)** 400 K, **(c)** 450 K, **(d)** 475 K, **(e)** 500 K, and **(f)** 600 K. During refinement, a March-Dollase model for slight preferred orientation for (00l) peaks was used for the hexagonal phase.

Table S1. Detailed Rietveld analysis for the PXRD results shown in Fig. S2.

	Temp.			RT	400 K	450 K	475 K	500 K	600 K
		R_B	(%)	4.35	5.19	5.17	5.60	5.26	4.74
		R_{wp}	(%)	5.76	6.66	6.61	7.06	6.61	6.14
		R_{exp}	(%)	2.65	6.42	6.16	6.20	6.14	5.77
		χ^2		2.18	1.04	1.07	1.14	1.08	1.06
Trigonal $Cu_2Se_{0.8}S_{0.2}$	$R\bar{3}m$	a	(Å)	4.0863(3)	-	-	-	-	-
		c	(Å)	20.268(2)	-	-	-	-	-
	Se/S	Ratio		80/20	-	-	-	-	-
		x		2/3	-	-	-	-	-
		y		1/3	-	-	-	-	-
		z		0.5797(2)	-	-	-	-	-
		U_{iso}	($Å^2$)	0.025(1)	-	-	-	-	-
	Cu2	x		0.6917(3)	-	-	-	-	-
		z		0.002(3)	-	-	-	-	-
		U_{iso}	($Å^2$)	0.6008(8)	-	-	-	-	-
	Cu1a	x		1/3	-	-	-	-	-
		y		2/3	-	-	-	-	-
		z		0.7768(4)	-	-	-	-	-
		U_{iso}	($Å^2$)	0.008(2)	-	-	-	-	-
		Occ.		0.620(7)	-	-	-	-	-
	Cu1b	z		0.7169(7)	-	-	-	-	-
Hexagonal **$Cu_2Se_{0.8}S_{0.2}$**	$P6_3/mmc$	a	(Å)	4.0702(1)	4.0794(1)	4.0863(1)	4.0889(2)	4.090(1)	-
		c	(Å)	6.8974(2)	6.9220(3)	6.9284(2)	6.9305(3)	6.936(1)	-
	Se/S	Ratio		80/20	80/20	80/20	80/20	80/20	-
		x		2/3	2/3	2/3	2/3	2/3	-
		y		1/3	1/3	1/3	1/3	1/3	-
		z		3/4	3/4	3/4	3/4	3/4	-
		U_{iso}	($Å^2$)	0.0227(7)	0.0334(7)	0.0373(9)	0.031(2)	0.0376	-
	Cu1	x = y		0	0	0	0	0	-
		z		¼	¼	¼	¼	¼	-
		U_{iso}	($Å^2$)	0.043(2)	0.042(2)	0.018(2)	0.024(6)	0.059	-
		Occ.		0.735(6)	0.675(6)	0.632(6)	0.59(2)	0.695	-

	Cu2	x		0.3945(9)	0.3985(7)	0.4039(6)	0.408(2)	-	-
		z		0.5671(3)	0.5713(3)	0.5712(4)	0.5803(8)	-	-
		U_{iso}	(Å^2)	0.041(3)	0.042(2)	0.018(2)	0.024(6)	-	-
		Occ.		0.197(2)	0.182(2)	0.182(2)	0.170(5)	-	-
Cubic $Cu_2Se_{0.8}S_{0.2}$	$Fm\bar{3}m$	a	(Å)	5.730(3)	5.7959(2)	5.8121(2)	5.81899(8)	5.82296(6)	5.83337(6)
	Se/S	Ratio		80/20	80/20	80/20	80/20	80/20	80/20
		x = y = z		0	0	0	0	0	0
		U_{iso}	(Å^2)	0.028	0.045(2)	0.061(1)	0.0468(6)	0.0461(5)	0.0498(4)
	Cu1	x = y = z	¼	¼	¼	¼	¼	¼	¼
		U_{iso}	(Å^2)	0.0412	0.12(1)	0.068(2)	0.073(4)	0.063(3)	0.061(3)
		Occ.		0.594	0.73(9)	0.424(5)	0.41(3)	0.36(2)	0.29(2)
	Cu2	x = y = z		0.326	0.33(2)	0.375(2)	0.320(3)	0.318(1)	0.317(1)
		Occ.		0.0714	0.03(2)	0.025(1)	0.093(5)	0.108(3)	0.111(3)

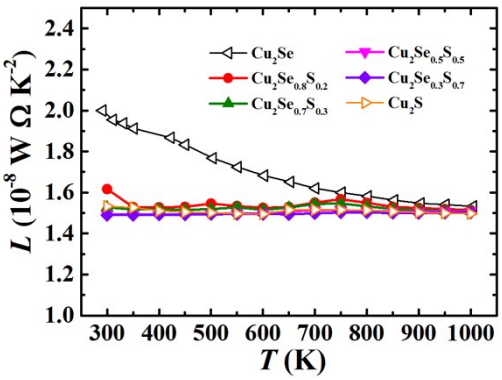

Fig. S3. Temperature dependence of Lorenz number for $Cu_2Se_{1-x}S_x$ (x= 0, 0.2, 0.3, 0.5, 0.7, and 1.0) samples.

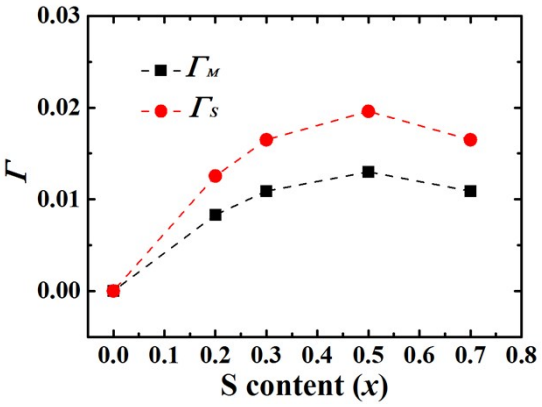

Fig. S4. Mass fluctuation scattering parameter Γ_M and strain field fluctuation scattering parameter

Γ_S as a function of the S-alloying content.

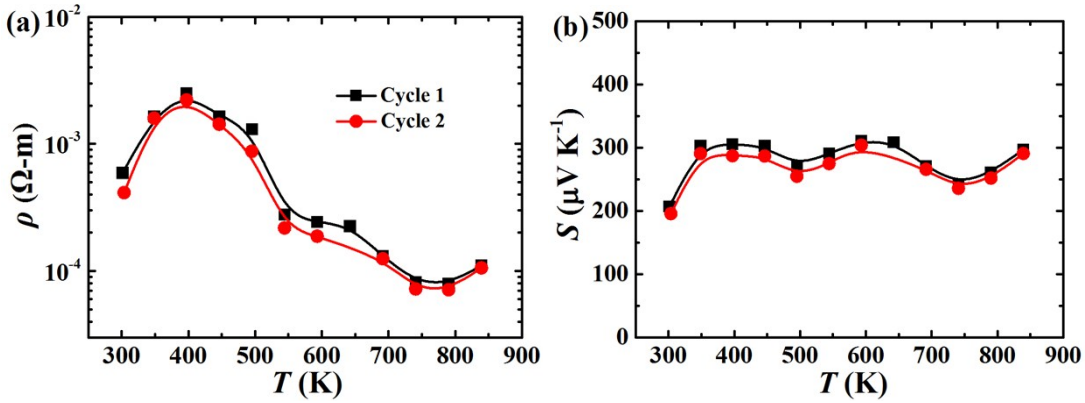

Fig. S5. Repeatability test on electronic transport properties in $Cu_2Se_{0.8}S_{0.2}$ sample.