Journal Name

SUPPORTING



Synthesis, structure and superconductivity of $FeS_{1-x}Se_x$ ($0 \le x \le 1$) solid solution crystals \dagger

Accepted 00th January 20xx DOI: 10.1039/x0xx00000x

Received 00th January 20xx,

Zhongnan Guo,^a Fan Sun,^a Yuyuan Chen,^a Yingluo Mao,^a Lin Wan,^a Xiaoxiao Yan, ^a Yang Yang,^a and Wenxia Yuan^{*a}

www.rsc.org/

Preparation of K_{0.8}Fe_{1.6}S₂ single crystals

The $K_{0.8}Fe_{1.6}(S_{1-x}Se_x)_2$ single crystals (so-called 245 phase) with x=0, 0.05, 0.1, 0.2 0.3, 0.35, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, and 1 were grown via self-flux method with the nominal composition. The pressed $Fe_{1.6}(S_{1-x}Se_x)_2$ disc (Fe: Alfa Aesar, 99.9%; S: Alfa Aesar, 99.9% and Se: Alfa Aesar, 99.95%) and K pieces (China National Accord Medicines Corporation Ltd, 97%;) were weighed as the target composition and added into the alumina crucible, inside a quartz tube. The quartz tube was sealed under vacuum, heated at 1040 °C for 15 hours and then slowly cooled down to 800 °C with 5 °C per hour, followed by furnace cooling to the room temperature.

Nomina x	245 phase (g)	Thiourea (g)	Selenourea (g)
0	0.24	0.23	0
0.05	0.243	0.243 0.22	
0.1	0.246 0.207		0.04
0.2	0.252 0.184		0.08
0.3	0.258 0.161		0.12
0.35	0.261	0.15	0.14
0.4	0.264 0.138		0.16
0.5	0.27	0.27 0.115	
0.6	0.276	0.09	0.24
0.7	0.282	0.07	0.283
0.8	0.289	0.046	0.323
0.9	0.295	0.023	0.36
1.0	1.0 0.3		0.4

Table S1. The detailed mass of raw materials of all the $FeS_{1-x}Se_x$ samples. The mass of Fe powder and NaOH are 0.16

and 0.12 g respectively for all the samples.

^{a.} Department of Chemistry, School of Chemistry and Biological Engineering, University of Science and Technology Beijing, Beijing 100083, China. E-mail: wxyuanwz@163.com

† Electronic Supplementary Information (ESI) available: [details of any supplementary information available should be included here]. See DOI: 10.1039/x0xx00000x

Table S2. The structural parameters, atomic coordinates and isotropic displacement parameters of $FeSe_{1-x}S_x$ solid solution with x = 0, 0.05, 0.1, 0.2, 0.3, 0.35, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, and 1.0.

Chemical	l formula		FeS (x = 0)			Chemical	formula		FeS _{0.7} Se _{0.3} (x	= 0.3)
Space gro	oup		P4/nmm (12	9)		Space gro	oup		P4/nmm (12	9)
a (Å)			3.6798(3)			a (Å)			3.7021(4)	
c (Å)			5.0287(6)			c (Å)			5.2069(8)	
V (Å ³)			67.928(9)			V (Å ³)			71.363(10)	
7 (11) Z			2			Z			2	
Ee-S(Se)	hond leng	th (Å)	$\frac{-}{22317(4)}$			E Fe-S(Se)	hond leng	th (Å)	2 2893(14)	
S(Se)-Fe-	-S(Se) and	e (°)	109.93(6) 1	09.62(6)		S(Se)-Fe-	S(Se) and	دا (۲) ۵ (°)	10791(1) 1	10 26(9)
Atom	v	v ()	7	$U_{\rm L}$ (Å ²)		Atom	v v	v ()	7	$IL(\lambda^2)$
Fo	۸ 0.75	y 0.25	2	$O_{iso}(A)$		Fo	0.75	y 0.25	2	$O_{iso}(A)$
re C/Co	0.75	0.25	0 2510(2)	0.055(2)		ге	0.75	0.25	0 2507(()	0.072(2)
5/Se	0.25	0.25	0.2518(2)	0.042(1)	-	5/Se	0.25	0.25	0.2587(6)	0.050(1)
					_					
Chemical	l formula		$FeS_{0.95}Se_{0.05}$ (2)	x = 0.05)		Chemical	formula		FeS _{0.65} Se _{0.35} ((x = 0.35)
Space gro	oup		P4/nmm (129	9)		Space gro	oup		P4/nmm (12	9)
a (Å)			3.6824(4)			a (Å)			3.7168(5)	
c (Å)			5.0703(8)			c (Å)			5.2588(10)	
V (Å ³)			68.753(13)			V (Å ³)			72.647(20)	
Ζ			2			Ζ			2	
Fe-S(Se)	bond leng	th (Å)	2.2428(13)			Fe-S(Se)	bond leng	th (Å)	2.3072(19)	
S(Se)-Fe-	-S(Se) angl	e (°)	110.35(7) 1	09.03(8)		S(Se)-Fe-	S(Se) angl	e (°)	107.31(1) 1	10.56(9)
Atom	X	y	Z	$U_{\rm iso}$ (Å ²)		Atom	x	v	Z	$U_{\rm iso}$ (Å ²)
Fe	0.75	0.25	0	0.071(2)		Fe	0.75	0.25	0	0.059(2)
S/Se	0.25	0.25	0.2526(2)	0.038(1)		S/Se	0.25	0.25	0.2600(6)	0.047(1)
						- /				
Chemical	l formula		FeSasSeative	= 0.1		Chemical	formula		FeSa Sea (v	= 0.4
Chemical Space gro	l formula		$FeS_{0.9}Se_{0.1}$ (x = P4 / nmm (12)	= 0.1) 9)		Chemical Space gro	formula		$FeS_{0.6}Se_{0.4}(x)$	= 0.4)
Chemical Space gro	l formula oup		$FeS_{0.9}Se_{0.1}$ (x = P4/nmm (129	= 0.1) 9)		Chemical Space gro	formula oup		FeS _{0.6} Se _{0.4} (x P4/nmm (12	= 0.4) 9)
Chemical Space gro <i>a</i> (Å)	l formula oup		FeS _{0.9} Se _{0.1} (x = <i>P</i> 4/ <i>nmm</i> (129 3.6926(3)	= 0.1) 9)		Chemical Space gro a (Å)	formula oup		FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6)	= 0.4) 9)
Chemical Space gro <i>a</i> (Å) <i>c</i> (Å)	l formula oup		FeS _{0.9} Se _{0.1} (x = <i>P</i> 4/ <i>nmm</i> (129 3.6926(3) 5.111(4)	= 0.1) 9)		Chemical Space gro a (Å) c (Å)	formula oup		FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12)	= 0.4) 9)
Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³)	l formula oup		FeS _{0.9} Se _{0.1} (x + P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8)	= 0.1) 9)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³)	formula oup		FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14)	= 0.4) 9)
Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i>	l formula oup		FeS _{0.9} Se _{0.1} (x + P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2	= 0.1) 9)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i>	formula oup		FeS _{0.6} Se _{0.4} (x <i>P</i> 4/ <i>nmm</i> (12 3.7503(6) 5.3618(12) 75.412(14) 2	= 0.4) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se)	l formula oup bond leng	th (Å)	FeS _{0.9} Se _{0.1} (x + P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4)	= 0.1) 9)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se)	formula oup bond leng	th (Å)	FeS _{0.6} Se _{0.4} (x <i>P</i> 4/ <i>nmm</i> (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14)	= 0.4) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe-	l formula oup bond leng -S(Se) angl	th (Å) e (°)	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16)	= 0.1) 9) 108.94(13)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se) S(Se)-Fe-	formula pup bond leng S(Se) angl	th (Å) e (°)	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1	= 0.4) 9) 10.02(8)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom	l formula oup bond leng -S(Se) angl x	th (Å) e (°) <i>y</i>	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) z	= 0.1) 9) 108.94(13) U_{iso} (Å ²)		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom	formula pup bond leng S(Se) angl	th (Å) e (°) <i>y</i>	FeS _{0.6} Se _{0.4} (x <i>P</i> 4/ <i>nmm</i> (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 <i>z</i>	= 0.4) 9) 10.02(8) U _{iso} (Å ²)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe	l formula oup bond leng -S(Se) angl x 0.75	th (Å) e (°) <i>y</i> 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) z 0	= 0.1) 9) 108.94(13) <i>U</i> _{iso} (Å ²) 0.059(2)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se) S(Se)-Fe- Atom Fe	bond leng S(Se) angl x 0.75	th (Å) e (°) <i>y</i> 0.25	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 z 0	= 0.4) 9) 10.02(8) $U_{iso}(Å^2)$ 0.053(2)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se	l formula oup bond leng -S(Se) angl x 0.75 0.25	th (Å) e (°) <u>y</u> 0.25 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) <i>z</i> 0 0.2513(6)	= 0.1) 9) 108.94(13) U_{iso} (Å ²) 0.059(2) 0.041(1)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se) S(Se)-Fe- Atom Fe S/Se	bond leng s(Se) angl x 0.75 0.25	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.6} Se _{0.4} (x <i>P</i> 4/ <i>nmm</i> (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 <i>z</i> 0 0.2523(4)	= 0.4) 9) 10.02(8) <i>U</i> _{iso} (Å ²) 0.053(2) 0.048(1)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se	l formula oup -S(Se) angl <i>x</i> 0.75 0.25	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) z 0 0.2513(6)	= 0.1) 9) 108.94(13) <i>U</i> _{iso} (Å ²) 0.059(2) 0.041(1)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se) S(Se)-Fe- Atom Fe S/Se	bond leng S(Se) angl x 0.75 0.25	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 z 0 0.2523(4)	= 0.4) 9) 10.02(8) <i>U</i> _{iso} (Å ²) 0.053(2) 0.048(1)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se	l formula oup -S(Se) angl x 0.75 0.25 I formula	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) Z 0 0.2513(6) FeS _{0.8} Se _{0.2} (x :	= 0.1) 9) 108.94(13) U_{iso} (Å ²) 0.059(2) 0.041(1) = 0.2)		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical	bond leng S(Se) angl x 0.75 0.25	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.6} Se _{0.4} (x <i>P</i> 4/ <i>nmm</i> (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 <i>z</i> 0 0.2523(4) FeS _{0.5} Se _{0.5} (x	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro	l formula oup -S(Se) angl x 0.75 0.25 I formula oup	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) 2 0 0.2513(6) FeS _{0.8} Se _{0.2} (x : P4/nmm (12 ⁴)	= 0.1) 9) 108.94(13) U_{iso} (Å ²) 0.059(2) 0.041(1) = 0.2) 9)		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro	bond leng S(Se) angl x 0.75 0.25 formula	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.6} Se _{0.4} (x <i>P</i> 4/ <i>nmm</i> (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 <i>z</i> 0 0.2523(4) FeS _{0.5} Se _{0.5} (x <i>P</i> 4/ <i>nmm</i> (12)	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å)	l formula oup -S(Se) angl x 0.75 0.25 I formula oup	th (Å) e (°) <u>y</u> 0.25 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) <i>z</i> 0 0.2513(6) FeS _{0.8} Se _{0.2} (x : P4/nmm (12 ⁴ 3.6861(2)	= 0.1) 9) 108.94(13) U_{iso} (Å ²) 0.059(2) 0.041(1) = 0.2) 9)		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å)	bond leng S(Se) angl x 0.75 0.25 formula	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.6} Se _{0.4} (x <i>P</i> 4/ <i>nmm</i> (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 <i>z</i> 0 0.2523(4) FeS _{0.5} Se _{0.5} (x <i>P</i> 4/ <i>nmm</i> (12 3.7651(8)	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å)	l formula oup -S(Se) angl x 0.75 0.25 I formula oup	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) <i>z</i> 0 0.2513(6) FeS _{0.8} Se _{0.2} (x : P4/nmm (12 ⁴ 3.6861(2) 5.1487(4)	= 0.1) 9) 108.94(13) U_{iso} (Å ²) 0.059(2) 0.041(1) = 0.2) 9)		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å)	bond leng S(Se) angl x 0.75 0.25	th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 z 0 0.2523(4) FeS _{0.5} Se _{0.5} (x P4/nmm (12 3.7651(8) 5.380(2)	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³)	l formula oup -S(Se) angl x 0.75 0.25 I formula oup	th (Å) e (°) 9 0.25 0.25	$\frac{\text{FeS}_{0.9}\text{Se}_{0.1}(\text{x}:}{P4/nmm (12^4)}\\3.6926(3)\\5.111(4)\\69.690(8)\\2\\2.263(4)\\110.53(16)\\\hline z\\0\\0.2513(6)\\\hline \\\hline \\ FeS_{0.8}\text{Se}_{0.2}(\text{x}:\\P4/nmm (12^4)\\3.6861(2)\\5.1487(4)\\69.957(5)\\\hline \\ \hline \\ \end{array}$	= 0.1) 9) 108.94(13) <i>U</i> _{iso} (Å ²) 0.059(2) 0.041(1) = 0.2) 9)		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³)	bond leng S(Se) angl x 0.75 0.25	th (Å) e (°) 9 0.25 0.25	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 z 0 0.2523(4) FeS _{0.5} Se _{0.5} (x P4/nmm (12 3.7651(8) 5.380(2) 76.267(16)	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z	l formula oup -S(Se) angl x 0.75 0.25 I formula oup	th (Å) e (°) <i>y</i> 0.25 0.25	$\frac{FeS_{0.9}Se_{0.1}(x = P4/nmm (124))}{3.6926(3)}$ 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) 2 0 0.2513(6) $\frac{z}{0}$ FeS_{0.8}Se_{0.2}(x = P4/nmm (124)) 3.6861(2) 5.1487(4) 69.957(5) 2	= 0.1) 9) 108.94(13) U_{iso} (Å ²) 0.059(2) 0.041(1) = 0.2) 9)		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z	bond leng S(Se) angl x 0.75 0.25	th (Å) e (°) 9 0.25 0.25	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 z 0 0.2523(4) FeS _{0.5} Se _{0.5} (x P4/nmm (12 3.7651(8) 5.380(2) 76.267(16) 2	= 0.4) 9) 10.02(8) <i>U</i> _{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se)	bond leng -S(Se) angl x 0.75 0.25 I formula oup	th (Å) e (°) 0.25 0.25	FeS _{0.9} Se _{0.1} (x : P4/nmm (12 ⁴ 3.6926(3) 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) Z 0 0.2513(6) FeS _{0.8} Se _{0.2} (x : P4/nmm (12 ⁴ 3.6861(2) 5.1487(4) 69.957(5) 2 2.249 (2)	= 0.1) 9) 108.94(13) U_{iso} (Å ²) 0.059(2) 0.041(1) = 0.2) 9)		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se)	bond leng S(Se) angl X 0.75 0.25 formula	th (Å) e (°) 0.25 0.25	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 z 0 0.2523(4) FeS _{0.5} Se _{0.5} (x P4/nmm (12 3.7651(8) 5.380(2) 76.267(16) 2 2.313(6)	= 0.4) 9) 10.02(8) <i>U</i> _{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe-	bond leng -S(Se) angl x 0.75 0.25 I formula oup bond leng	th (Å) e (°) 0.25 0.25 th (Å)	$\frac{FeS_{0.9}Se_{0.1}(x = P4/nmm (124))}{3.6926(3)}$ 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) 2 0 0.2513(6) $\frac{z}{0}$ $\frac{1}{2}$	= 0.1) $= 0.1)$ $= 0.1)$ $= 0.2)$ $= 0.2(9)$		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe-	bond leng S(Se) angl x 0.75 0.25 formula pup	th (Å) e (°) 0.25 0.25 th (Å)	$\begin{array}{c} {\rm FeS}_{0.6}{\rm Se}_{0.4}({\rm x}\\ P4/nmm(12)\\ 3.7503(6)\\ 5.3618(12)\\ 75.412(14)\\ 2\\ 2.3122(14)\\ 108.38(8)\ 1\\ \hline \\ z\\ 0\\ 0.2523(4)\\ \hline \\ \hline \\ FeS_{0.5}{\rm Se}_{0.5}({\rm x}\\ P4/nmm(12)\\ 3.7651(8)\\ 5.380(2)\\ 76.267(16)\\ 2\\ 2.313(6)\\ 10893(18)\\ \hline \end{array}$	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9) 109.74(17)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe-	l formula oup -S(Se) angl x 0.75 0.25 I formula oup bond leng -S(Se) angl	th (Å) e (°) 0.25 0.25 th (Å) e (°)	$\frac{\text{FeS}_{0.9}\text{Se}_{0.1}(\text{x}:}{P4/nmm (12^4)}$ $\frac{3.6926(3)}{5.111(4)}$ $\frac{5.111(4)}{69.690(8)}$ $\frac{2}{2.263(4)}$ $110.53(16)$ $\frac{z}{0}$ $0.2513(6)$ $\frac{7}{100}$ $\frac{7}{100}$ $\frac{7}{100}$ $\frac{7}{100}$	= 0.1) $= 0.1)$ $= 0.1)$ $= 0.059(1)$ $= 0.00000000000000000000000000000000000$		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- S(Se)-Fe-	bond leng S(Se) angl x 0.75 0.25 formula oup bond leng S(Se) angl	th (Å) e (°) <i>y</i> 0.25 0.25 th (Å) e (°)	$\begin{array}{c} {\rm FeS}_{0.6}{\rm Se}_{0.4}({\rm x}\\ P4/nmm(12)\\ 3.7503(6)\\ 5.3618(12)\\ 75.412(14)\\ 2\\ 2.3122(14)\\ 108.38(8) 1\\ \hline \\ z\\ 0\\ 0.2523(4)\\ \hline \\ \hline \\ {\rm FeS}_{0.5}{\rm Se}_{0.5}({\rm x}\\ P4/nmm(12)\\ 3.7651(8)\\ 5.380(2)\\ 76.267(16)\\ 2\\ 2\\ 2.313(6)\\ 108.93(18)\\ \hline \\ \end{array}$	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9) 109.74(17) U_{i} (Å ²)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom	l formula oup -S(Se) angl x 0.25 -S(Se) angl oup -S(Se) angl x 0.75	th (Å) e (°) <i>y</i> 0.25 0.25 th (Å) e (°) <i>y</i> 0.25	$\frac{FeS_{0.9}Se_{0.1}(x = P4/nmm (124))}{3.6926(3)}$ 5.111(4) 69.690(8) 2 2.263(4) 110.53(16) 2 0 0.2513(6) 7 FeS_{0.8}Se_{0.2}(x = P4/nmm (124)) 3.6861(2) 5.1487(4) 69.957(5) 2 2.249 (2) 110.35(1) 140 2 0	= 0.1) $= 0.1)$ $= 0.1)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$ $= 0.2)$		Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro <i>a</i> (Å) <i>c</i> (Å) <i>V</i> (Å ³) <i>Z</i> Fe-S(Se) S(Se)-Fe- Atom	bond leng S(Se) angl x 0.25 formula oup bond leng S(Se) angl x 0.75	th (Å) e (°) <i>y</i> 0.25 0.25 th (Å) e (°) <i>y</i> 0.25	$\begin{array}{c} {\rm FeS}_{0.6}{\rm Se}_{0.4}({\rm x}\\ P4/nmm(12)\\ 3.7503(6)\\ 5.3618(12)\\ 75.412(14)\\ 2\\ 2.3122(14)\\ 108.38(8) 1\\ \hline \\ z\\ 0\\ 0.2523(4)\\ \hline \\ \hline \\ FeS}_{0.5}{\rm Se}_{0.5}({\rm x}\\ P4/nmm(12)\\ 3.7651(8)\\ 5.380(2)\\ 76.267(16)\\ 2\\ 2.313(6)\\ 108.93(18)\\ \hline \\ z\\ 0\\ \hline \end{array}$	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9) 109.74(17) U_{iso} (Å ²) 0.066(2)
Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se	bond leng -S(Se) angl x 0.75 0.25 l formula oup bond leng -S(Se) angl x 0.75 0.25	th (Å) e (°) <i>y</i> 0.25 0.25 th (Å) e (°) <i>y</i> 0.25 0.25	$\begin{array}{c} {\rm FeS}_{0.9}{\rm Se}_{0.1}({\rm x}:\\ P4/nmm(12^4)\\ 3.6926(3)\\ 5.111(4)\\ 69.690(8)\\ 2\\ 2.263(4)\\ 110.53(16)\\ \hline \\ z\\ 0\\ 0.2513(6)\\ \hline \\ \hline \\ FeS_{0.8}{\rm Se}_{0.2}({\rm x}:\\ P4/nmm(12^4)\\ 3.6861(2)\\ 5.1487(4)\\ 69.957(5)\\ 2\\ 2.249(2)\\ 110.35(1) 1\\ \hline \\ z\\ 0\\ 0,2540(7)\\ \hline \end{array}$	= 0.1) $= 0.1)$ $= 0.1)$ $= 0.2)$		Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se Chemical Space gro a (Å) c (Å) V (Å ³) Z Fe-S(Se) S(Se)-Fe- Atom Fe S/Se	bond leng S(Se) angl x 0.75 0.25 formula oup bond leng S(Se) angl x 0.75 0.25	th (Å) e (°) <i>y</i> 0.25 0.25 th (Å) e (°) <i>y</i> 0.25 0.25	FeS _{0.6} Se _{0.4} (x P4/nmm (12 3.7503(6) 5.3618(12) 75.412(14) 2 2.3122(14) 108.38(8) 1 z 0 0.2523(4) FeS _{0.5} Se _{0.5} (x P4/nmm (12 3.7651(8) 5.380(2) 76.267(16) 2 2.313(6) 108.93(18) z 0 0 2499(20)	= 0.4) 9) 10.02(8) U_{iso} (Å ²) 0.053(2) 0.048(1) = 0.5) 9) 109.74(17) U_{iso} (Å ²) 0.066(2) 0.044(1)

Journal Name

Chemical f	ormula		$FeS_{0.4}Se_{0.6} (x = 0.6)$			
Space grou	ıp		P4/nmm (129)			
a (Å)			3.7758(7)			
c (Å)			5.4007(9)			
V (Å ³)			76.996(13)			
Ζ			2			
Fe-S(Se) b	ond lengtł	n (Å)	2.348(2)			
S(Se)-Fe-S(Se) angle (°)			107.03(10) 110.70(9)			
Atom	Х	у	Ζ	$U_{\rm iso}$ (Å ²)		
Fe	0.75	0.25	0	0.057(2)		
S/Se	0.25	0.25	0.2585(6)	0.042(1)		
Chemical f	ormula		$FeS_{0,3}Se_{0,7}(x =$: 0.7)		
Space group			P4/nmm (129	·)		
a (Å)			3.776(1)	-		
c (Å)			5.4492(5)			
V (Å ³)			77.696(6)			
Z			2			
Fe-S(Se) b	ond lengtł	n (Å)	2.3526(16)			
S(Se)-Fe-S	(Se) angle	ເບ	106.74(8) 110.81(8)			
Atom	X	y	Z	$U_{\rm iso}$ (Å ²)		
Fe	0.75	0.25	0	0.052(2)		
S/Se	0.25	0.25	0.2576(4)	0.044(1)		
Chemical f	ormula		$FeS_{0.2}Se_{0.8}$ (x =	: 0.8)		
Space grou	ıp		P4/nmm (129)			
a (Å)			3.7838(3)			
c (Å)			5.4781(6)			
V (Å ³)			78.431(11)			
Z			2			
Fe-S(Se) bond length (Å)			2.3794(14)			
S(Se)-Fe-S(Se) angle (°)			105.34(8) 111.58(8)			
Atom	X	у	Ζ	$U_{\rm iso}$ (Å ²)		
Fe	0.75	0.25	0	0.067(2)		
S/Se	0.25	0.25	0.2634(4)	0.053(1)		
Chemical f	ormula		$FeS_{0,1}Se_{0,9}(x =$: 0.9)		
Space grou	ıp		P4/nmm (129)			
a (Å)			3.7963(2)			
c (Å)			5.4848(16)			
V (Å ³)			79.046(21)			
Ζ			2			
Fe-S(Se) b	ond lengtł	n (Å)	2.437(5)			
S(Se)-Fe-S	(Se) angle	(°)	102.29(13) 113.18(9)			

Chemical	formula		FeSe (x = 1.0)		
Space gro	oup		P4/nmm (129)		
a (Å)			3.7904(4)		
<i>c</i> (Å)			5.5203(11)		
V (Å ³)			79.31(2)		
Ζ			2		
Fe-S(Se) bond length (Å)			2.433(3)		
S(Se)-Fe-S(Se) angle (°)			102.35(7) 113.15(9)		
Atom	Х	у	Z	$U_{\rm iso}$ (Å ²)	
Fe	0.75	0.25	0	0.071(3)	
S/Se	0.25	0.25	0.2763(6)	0.040(2)	



Fig. 1 PXRD patterns of $K_{0.8}Fe_{1.6}(S_{1-x}Se_x)_2$, which can be indexed by *I*4/*m* space group. The left inset shows the optical photograph of the crystal sample with x=0.5, and the right inset shows the enlarged [002] diffraction peaks.



Fig. S2 Morphology of $FeS_{1-x}Se_x$ crystal sheets with x= (a) 0, (b) 0.1, (c) 0.5 and (d) 0.8. The red squares represent selected area for EDS measurements.

Atom

Fe

S/Se

х

0.75

0.25

y

0.25

0.25

Ζ

0

0.2788(12)

 $U_{\rm iso}$ (Å²)

0.047(2)

0.043(1)



Fig. S3 The Rietveld refinements of $FeS_{1-x}Se_x$ with x = (a) 0, (b) 0.1, (c) 0.5, (d) 0.7, (e) 0.8 and (f) 1.



Fig. S4 Zero-field-cooling (ZFC, in black) and field-cooling (FC, in red) magnetization versus temperature curves of nominal $FeS_{1-x}Se_x$ with x = 0.4 and 0.5 under the field of 20 Oe.



Fig. S5 Zero-field-cooling (ZFC) and field-cooling (FC) magnetization versus temperature curves of FeSe from solid state reaction, showing the diamagnetic transition.



Fig. S6 The Rietveld refinement of FeSe synthesized fromsolidstatereaction.