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

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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]

Preparation of $K_{0.8}Fe_{1.6}S_2$ 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.

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.

Nomina x	245 phase (g)	Thiourea (g)	Selenourea (g)
0	0.24	0.23	0
0.05	0.243	0.22	0.02
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	0.115	0.2
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	0.3	0	0.4

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

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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 formula			FeS (x = 0)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.6798(3)	
c (Å)			5.0287(6)	
V (Å^3)			67.928(9)	
Z			2	
Fe-S(Se) bond length (Å)			2.2317(4)	
S(Se)-Fe-S(Se) angle (˚)			109.93(6)	109.62(6)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.055(2)
S/Se	0.25	0.25	0.2518(2)	0.042(1)

Chemical formula			$FeS_{0.95}Se_{0.05}$ (x = 0.05)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.6824(4)	
c (Å)			5.0703(8)	
V (Å^3)			68.753(13)	
Z			2	
Fe-S(Se) bond length (Å)			2.2428(13)	
S(Se)-Fe-S(Se) angle (˚)			110.35(7)	109.03(8)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.071(2)
S/Se	0.25	0.25	0.2526(2)	0.038(1)

Chemical formula			$FeS_{0.9}Se_{0.1}$ (x = 0.1)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.6926(3)	
c (Å)			5.111(4)	
V (Å^3)			69.690(8)	
Z			2	
Fe-S(Se) bond length (Å)			2.263(4)	
S(Se)-Fe-S(Se) angle (˚)			110.53(16)	108.94(13)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.059(2)
S/Se	0.25	0.25	0.2513(6)	0.041(1)

Chemical formula			$FeS_{0.8}Se_{0.2}$ (x = 0.2)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.6861(2)	
c (Å)			5.1487(4)	
V (Å^3)			69.957(5)	
Z			2	
Fe-S(Se) bond length (Å)			2.249 (2)	
S(Se)-Fe-S(Se) angle (˚)			110.35(1)	109.03(9)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.069(2)
S/Se	0.25	0.25	0.2540(7)	0.049(1)

Chemical formula			$FeS_{0.7}Se_{0.3}$ (x = 0.3)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.7021(4)	
c (Å)			5.2069(8)	
V (Å^3)			71.363(10)	
Z			2	
Fe-S(Se) bond length (Å)			2.2893(14)	
S(Se)-Fe-S(Se) angle (˚)			107.91(1)	110.26(9)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.072(2)
S/Se	0.25	0.25	0.2587(6)	0.050(1)

Chemical formula			$FeS_{0.65}Se_{0.35}$ (x = 0.35)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.7168(5)	
c (Å)			5.2588(10)	
V (Å^3)			72.647(20)	
Z			2	
Fe-S(Se) bond length (Å)			2.3072(19)	
S(Se)-Fe-S(Se) angle (˚)			107.31(1)	110.56(9)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.059(2)
S/Se	0.25	0.25	0.2600(6)	0.047(1)

Chemical formula			$FeS_{0.6}Se_{0.4}$ (x = 0.4)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.7503(6)	
c (Å)			5.3618(12)	
V (Å^3)			75.412(14)	
Z			2	
Fe-S(Se) bond length (Å)			2.3122(14)	
S(Se)-Fe-S(Se) angle (˚)			108.38(8)	110.02(8)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.053(2)
S/Se	0.25	0.25	0.2523(4)	0.048(1)

Chemical formula			$FeS_{0.5}Se_{0.5}$ (x = 0.5)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.7651(8)	
c (Å)			5.380(2)	
V (Å^3)			76.267(16)	
Z			2	
Fe-S(Se) bond length (Å)			2.313(6)	
S(Se)-Fe-S(Se) angle (˚)			108.93(18)	109.74(17)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.066(2)
S/Se	0.25	0.25	0.2499(20)	0.044(1)

Chemical formula			$FeS_{0.4}Se_{0.6}$ (x = 0.6)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.7758(7)	
c (Å)			5.4007(9)	
V (Å^3)			76.996(13)	
Z			2	
Fe-S(Se) bond length (Å)			2.348(2)	
S(Se)-Fe-S(Se) angle (˚)			107.03(10)	110.70(9)
Atom	*x*	*y*	*z*	U_{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 formula			$FeS_{0.3}Se_{0.7}$ (x = 0.7)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.776(1)	
c (Å)			5.4492(5)	
V (Å^3)			77.696(6)	
Z			2	
Fe-S(Se) bond length (Å)			2.3526(16)	
S(Se)-Fe-S(Se) angle (˚)			106.74(8)	110.81(8)
Atom	*x*	*y*	*z*	U_{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 formula			$FeS_{0.2}Se_{0.8}$ (x = 0.8)	
Space group			*P*4/*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*	*y*	*z*	U_{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 formula			$FeS_{0.1}Se_{0.9}$ (x = 0.9)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.7963(2)	
c (Å)			5.4848(16)	
V (Å^3)			79.046(21)	
Z			2	
Fe-S(Se) bond length (Å)			2.437(5)	
S(Se)-Fe-S(Se) angle (˚)			102.29(13)	113.18(9)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.047(2)
S/Se	0.25	0.25	0.2788(12)	0.043(1)

Chemical formula			FeSe (x = 1.0)	
Space group			*P*4/*nmm* (129)	
a (Å)			3.7904(4)	
c (Å)			5.5203(11)	
V (Å^3)			79.31(2)	
Z			2	
Fe-S(Se) bond length (Å)			2.433(3)	
S(Se)-Fe-S(Se) angle (˚)			102.35(7)	113.15(9)
Atom	*x*	*y*	*z*	U_{iso} (Å^2)
Fe	0.75	0.25	0	0.071(3)
S/Se	0.25	0.25	0.2763(6)	0.040(2)

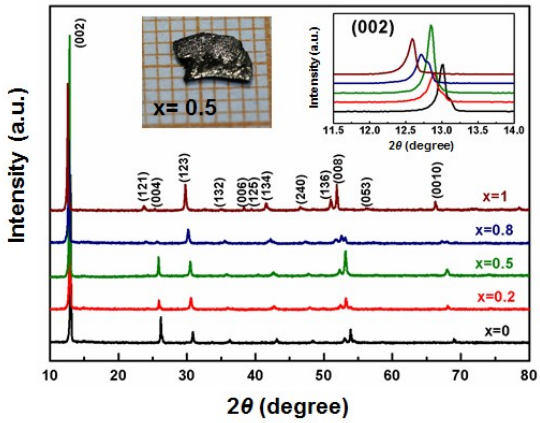

Fig. 1 PXRD patterns of $K_{0.8}Fe_{1.6}(S_{1-x}Se_x)_2$, which can be indexed by *I*4/*m* space group. The left inset shows the optical photograph of the crystal sample with x=0.5, and the right inset shows the enlarged [002] diffraction peaks.

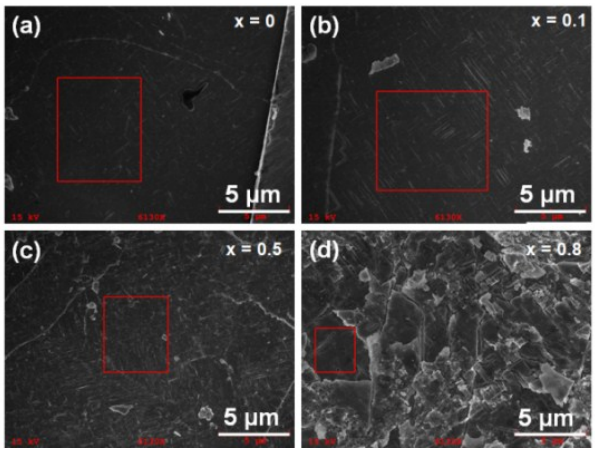

Fig. S2 Morphology of $FeS_{1-x}Se_x$ crystal sheets with x= (a) 0, (b) 0.1, (c) 0.5 and (d) 0.8. The red squares represent selected area for EDS measurements.

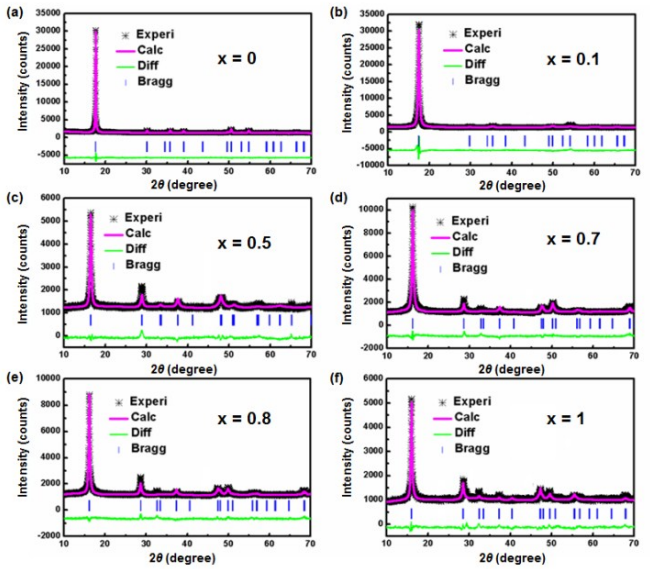

Fig. S3 The Rietveld refinements of $FeS_{1-x}Se_x$ with x = (a) 0, (b) 0.1, (c) 0.5, (d) 0.7, (e) 0.8 and (f) 1.

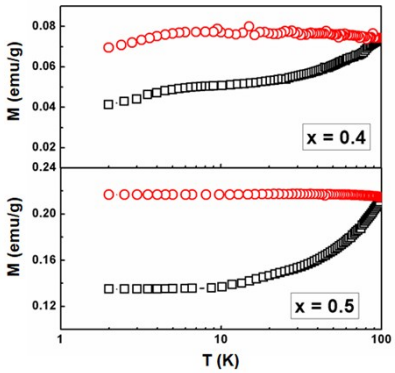

Fig. S4 Zero-field-cooling (ZFC, in black) and field-cooling (FC, in red) magnetization versus temperature curves of nominal $FeS_{1-x}Se_x$ with x = 0.4 and 0.5 under the field of 20 Oe.

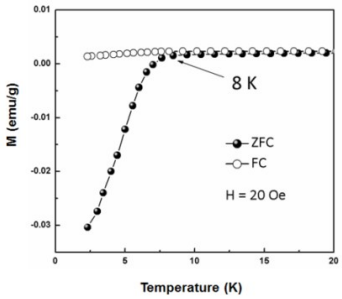

Fig. S5 Zero-field-cooling (ZFC) and field-cooling (FC) magnetization versus temperature curves of FeSe from solid state reaction, showing the diamagnetic transition.

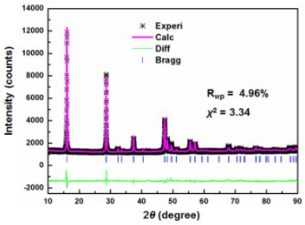

Fig. S6 The Rietveld refinement of FeSe synthesized from solid state reaction.