Supplementary Information

Ferromagnetic interlayer interaction in $KCo_2Se_{2-x}S_x$ ($0 \leq x \leq 2$) and its chemical origin

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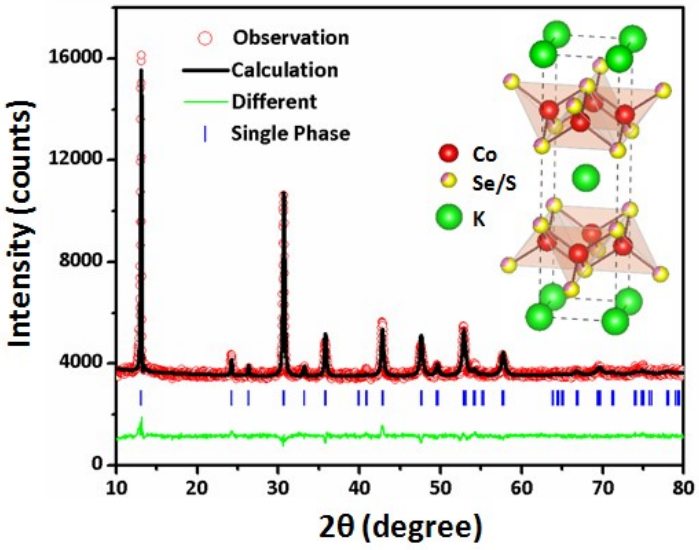

Fig. S1 The Rietveld refinement fits, difference profiles and positions of Bragg peaks of $KCo_2Se_{1.4}S_{0.6}$. The inset shows the crystal structure of $KCo_2Se_{1.4}S_{0.6}$ with the same atom coordinate of Se and S.

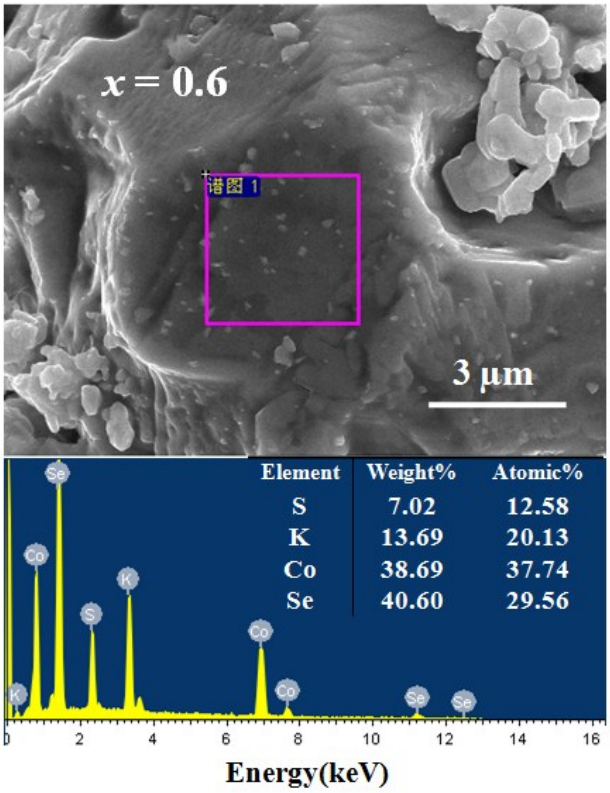

Fig. S2 The sample morphology and EDX result of $KCo_2Se_{1.4}S_{0.6}$.

Talbe S1. Summary of the EDX results and the Rietveld refinement parameters of $KCo_2Se_{2-x}S_x$.

Nominal *x*	0	0.2	0.6	1	1.4	2
x from EDX	0	0.196	0.597	1.011	1.560	2
Space Group	*I*4/*mmm*					
a(Å)	3.8672(7)	3.8510(4)	3.8292(6)	3.7931(10)	3.7583(8)	3.7295(4)
c(Å)	13.695(14)	13.6270(7)	13.5119(11)	13.407(2)	13.2523 (16)	13.063(2)
V(Å^3)	204.812	202.091	198.122	192.895	187.186	181.696
K	0,0,0					
Co	0,0.5,0.25					
Se/S	0,0,0.3396(2)	0,0,0.3493(1)	0,0,0.3461(1)	0,0,0.3370(3)	0,0,0.3348(3)	0,0,0.3399(3)
Co-Se/S-Co(º)	73.53(8)	70.76(6)	71.52(7)	73.95(10)	74.73(9)	73.77(8)
R_{wp}	6.45%	5.18%	5.27%	7.32%	5.58%	4.12%
χ^2	4.85	1.88	2.29	2.32	3.16	1.84

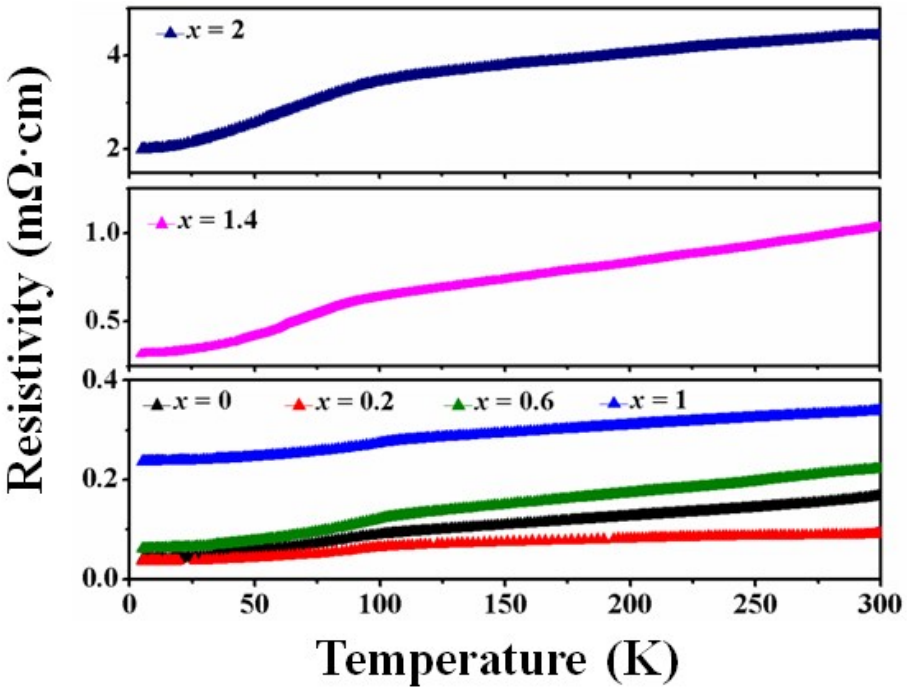

Fig. S3 Temperature dependent resistivity of $KCo_2Se_{2-x}S_x$.

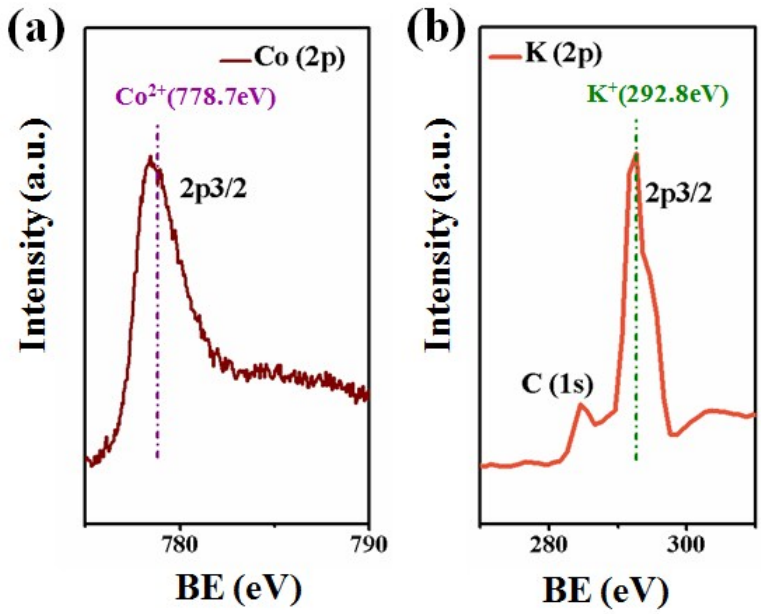

Fig. S4 (a) XPS Co 2p and (b) K 2p of sample $KCo_2Se_{1.4}S_{0.6}$. The two dashed lines correspond to the binding energies of Co +2 valence[1] and K +1 valence. [2] BE denotes the binding energy.

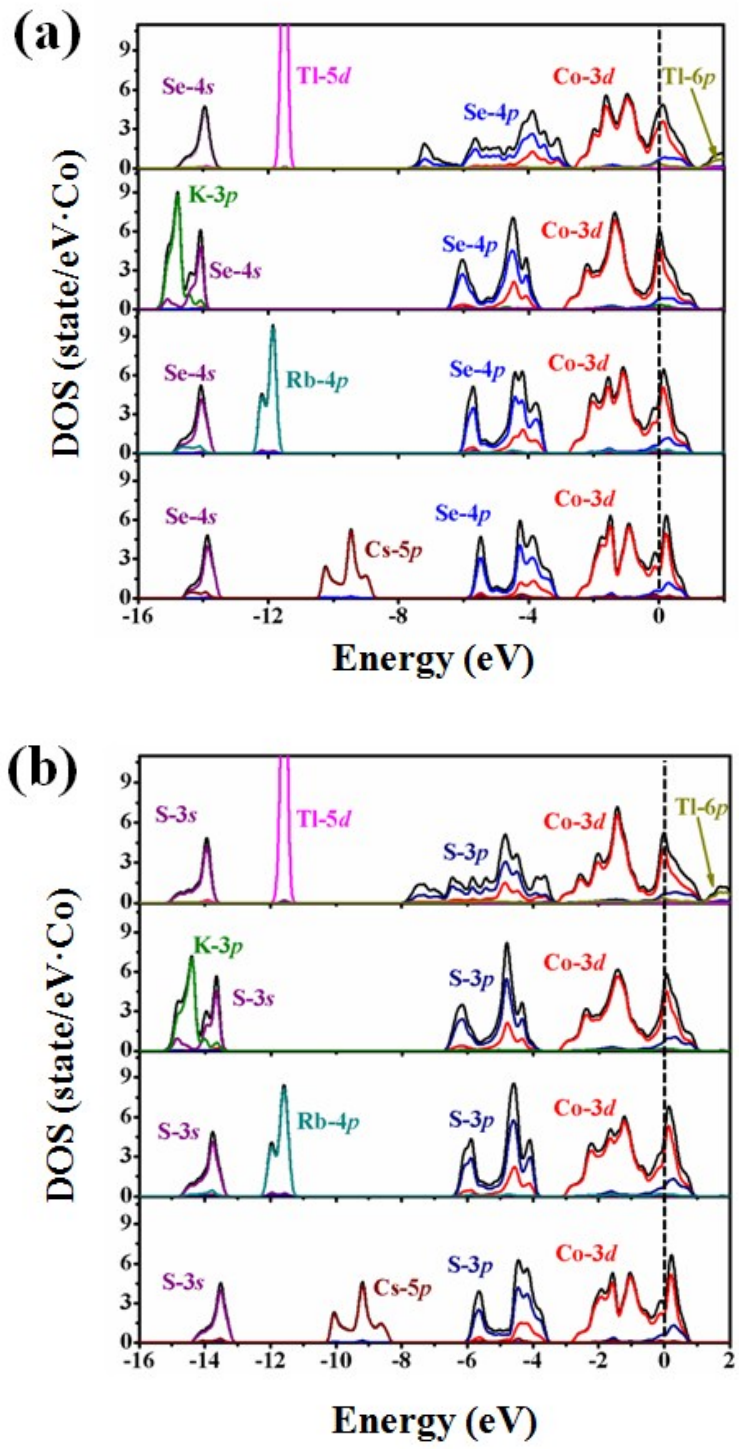

Fig. S5 Total and partial DOS of eight layered cobalt chalcogenides:

(a) four selenides and (b) four sulfides.

Table S2. Structure parameters used for the RKKY model and the obtained results:

Composition	Type	a (Å)	c (Å)	$N(E_F)$ (eV^{-1})	k_F	x	Γ $\times 10^5$
$KCo_2Se_{1.4}S_{0.6}$	F	3.8292	13.5119	4.98	2.2836	30.8552	2.9416
$TlCo_2Se_{1.75}S_{0.25}$	AF	3.8402[3]	13.494[3]	4.51	2.2061	29.7689	-0.1615
$TlCo_2Se_2$	AF	3.8471[3]	13.542[3]	4.41	2.1852	29.5938	-0.8335
KCo_2Se_2	F	3.8672	13.695	5.92	2.3911	32.5805	1.0611
$RbCo_2Se_2$	F	3.825[4]	14.49[4]	3.98	2.072	30.0120	0.7641
$CsCo_2Se_2$	AF	3.824[4]	15.314[4]	2.99	1.8506	28.3408	-4.3731
$TlCo_2S_2$	F	3.741[3]	12.956[3]	5.23	2.3907	30.9744	3.0888
KCo_2S_2	F	3.7295	13.063	5.13	2.3754	30.7728	2.8129
$RbCo_2S_2$	F	3.725[4]	13.74[4]	4.36	2.2132	30.4088	1.9989
$CsCo_2S_2$	F	3.736[4]	14.5[4]	2.81	1.8736	28.0116	-4.4358

where $\Gamma = \frac{x\cos x - \sin x}{x^4}$ and $x = 2k_F d$, and for a tetragonal unit cell the k_F could be obtained from [5]:

$$k_F = 2(6Z\pi^2 / V)^{\frac{1}{3}}$$

Z is the number of the conduction electrons per Co, and for our calculation we use the DOS per Co at the Fermi level $N(E_F)$ as Z.[6]

References

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3. M. Greaney, G. Huan, K. V. Ramanujachary, Z. Teweldmedhin and M. Greenblatt, *Solid State Commun.,* 1991, **79**, 803-810.
4. G. Huan, M. Greenblatt and M. Croft, *Eur. J. Solid State Inorg. Chem.*, 1989, **26**. 193-220.
5. J. Leciejewicz and A. Szytula, *J. Magn. Magn. Mater.*, 1987, **63-64**, 190-192.
6. An integration of the partial DOS of Co 3*d* states near the Fermi level was firstly used as the Z in RKKY model, but the results could not reflect the different

ordering types since the integration of different compounds are basically the same. So we turn to use the $N(E_F)$ directly as Z.