Supplementary Information for "CrRhAs: a member of a large family of metallic kagome antiferromagnets"

Y. N. Huang
Department of Physics, Zhejiang University of Science and Technology, Hangzhou 310023, People's Republic of China

Harald O. Jeschke
Research Institute for Interdisciplinary Science, Okayama University, Okayama 700-8530, Japan

Igor I. Mazin*
Department of Physics & Astronomy, George Mason University, Fairfax, VA 22030, USA and
Quantum Science and Engineering Center, George Mason University, Fairfax, VA 22030, USA.
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Supplementary Note 1. XYZ COMPOUNDS WITH SIGNIFICANT MAGNETISM

Initially, we classified XYZ compounds with space group $P\bar{6}2m$ (no. 189) by groups of elements. By analyzing all P$\bar{6}$2m XYZ compounds in the Materials Project database, we found that X, Y, Z atoms belong to finite groups of elements as shown in Supplementary Table 1.

With poor metal, we refer to Al, Ga, In, Sn, Tl, Pb, Bi. As metalloid, we classify B, Si, Ge, As and Sb. Relevant nonmetals are H, C, N and P. Specifically, the XY combination of element series is shown in Supplementary Table 2.

Supplementary Table 1: XYZ compounds classified by element series

Atom	Element Series
X	actinide, alkaline earth metal, lanthanide, poor metal, transition metal
Y	alkali metal, alkaline earth metal, lanthanide, metalloid, poor metal, transition metal
Z	alkaline earth metal, lanthanide, metalloid, nonmetal, poor metal, transition metal

Supplementary Table 2: XY combination of groups of elements.

X group	Y group
actinide	metalloid
actinide	poor metal
alkaline earth metal	alkali metal
alkaline earth metal	poor metal
alkaline earth metal	transition metal
lanthanide	alkali metal
lanthanide	alkaline earth metal
lanthanide	metalloid
lanthanide	poor metal
lanthanide	transition metal
poor metal	lanthanide
poor metal	transition metal
transition metal	alkali metal
transition metal	alkaline earth metal
transition metal	metalloid
transition metal	poor metal
transition metal	transition metal

We choose P$\bar{6}$2m XYZ compounds with significant magnetism from the Materials Project (MP)[S1], and organize them into Supplementary Table 3. mp-id is the material ID of MP. X is the kagome atom, Y is trimer atom. $|v|(X)$

* imazin2@gmu.edu

and $|v|(Y)|$ are the v values of X and Y respectively where $v = x - 1/2$ is is determined from the fractional coordinate x of the X or Y atom (see also main text). The ICSD column shows whether the compound exists and has been reported to the Inorganic Crystal Structure Database (ICSD).

Supplementary Table 3: XYZ (space group $P\bar{6}2m$) compounds with significant magnetism.

mp-id	compound	X	$\|v\|(X)$	Y	$\|v\|(Y)\|$	ICSD
mp-20665	CeInCu	Ce	0.081851	In	0.252775	yes
mp-15683	CrRhAs	Cr	0.092491	Rh	0.245181	yes
mp-1205827	CrPdAs	Cr	0.103397	Pd	0.232872	no
mp-1087537	CrPdP	Cr	0.105917	Pd	0.231023	yes
mp-4989	CrNiAs	Cr	0.095091	Ni	0.244877	yes
mp-1206397	CrCoAs	Cr	0.086577	Co	0.252504	no
mp-1206083	CrFeAs	Cr	0.087428	Fe	0.253454	no
mp-1079966	CrNiP	Cr	0.091987	Ni	0.244212	yes
mp-1091389	EuInPd	Eu	0.089105	In	0.252248	yes
mp-1225012	FeRhAs	Fe	0.103656	Rh	0.236774	no
mp-1224852	FeRhP	Fe	0.110326	Rh	0.231339	no
mp-1080653	FeNiP	Fe	0.098632	Ni	0.242936	yes
mp-1080612	GdAlCu	Gd	0.085396	Al	0.267555	yes
mp-20332	GdMgPd	Gd	0.086926	Mg	0.257178	yes
mp-22028	GdMgPt	Gd	0.090582	Mg	0.25546	yes
mp-623988	GdCdPd	Gd	0.093488	Cd	0.245842	yes
mp-20224	GdInPd	Gd	0.090917	In	0.242642	yes
mp-1101090	GdAlNi	Gd	0.080304	Al	0.267974	yes
mp-1080178	GdZnPd	Gd	0.096443	Zn	0.249717	yes
mp-30692	GdTlPd	Gd	0.09759	Tl	0.234001	yes
mp-574004	GdMgGa	Gd	0.075155	Mg	0.256293	yes
mp-20058	GdAlPd	Gd	0.082467	Al	0.265203	yes
mp-1101056	GdMgIn	Gd	0.065156	Mg	0.259364	yes
mp-1079690	GdAgSi	Gd	0.083068	Ag	0.249535	yes
mp-1080496	GdInAu	Gd	0.091863	In	0.242823	yes
mp-1079739	GdSnPt	Gd	0.089816	Sn	0.244624	yes
mp-9341	GdAgGe	Gd	0.084516	Ag	0.249724	yes
mp-19745	GdInIr	Gd	0.092568	In	0.245165	yes
mp-623043	GdInRh	Gd	0.091525	In	0.245585	yes
mp-1206951	GdMgTl	Gd	0.070661	Mg	0.256942	no
mp-14210	GdLiGe	Gd	0.077382	Li	0.267255	no
mp-582029	GdInPt	Gd	0.092445	In	0.249518	yes
mp-580154	GdMgAu	Gd	0.08771	Mg	0.254229	yes
mp-21458	GdCdCu	Gd	0.088142	Cd	0.250108	yes
mp-1079035	MnPdAs	Mn	0.092059	Pd	0.238231	yes
mp-1079184	MnRhGe	Mn	0.0912	Rh	0.241729	yes
mp-10049	MnRuAs	Mn	0.095843	Ru	0.242341	yes
mp-1206331	MnPdP	Mn	0.095237	Pd	0.235672	no
mp-610972	MnPdGe	Mn	0.096695	Pd	0.235066	yes
mp-567856	MnRhAs	Mn	0.095212	Rh	0.236511	yes
mp-1079845	MnNiAs	Mn	0.085264	Ni	0.248649	yes
mp-1079806	MnTiAs	Mn	0.109278	Ti	0.238304	yes
mp-1079405	MnRhP	Mn	0.10521	Rh	0.235009	yes
mp-4238	MnFeAs	Mn	0.094506	Fe	0.245038	yes
mp-975423	MnNiP	Mn	0.100394	Ni	0.24237	yes
mp-20314	NpSnIr	Np	0.085981	Sn	0.245747	yes
mp-1091393	PuGaNi	Pu	0.082098	Ga	0.260442	yes
mp-1078774	PuGaRh	Pu	0.086028	Ga	0.252119	yes
mp-1079793	PuAlCo	Pu	0.071542	Al	0.262264	yes
mp-1080562	USnPt	U	0.087948	Sn	0.245606	yes
mp-1078683	UInPd	U	0.080172	In	0.251386	yes
mp-1078582	UInPt	U	0.08236	In	0.249319	yes
mp-1080442	UGaPd	U	0.073034	Ga	0.261463	yes
mp-1079696	USnRh	U	0.081128	Sn	0.248363	yes
mp-1080803	USnIr	U	0.083744	Sn	0.247099	yes
mp-1095127	USbRu	U	0.086204	Sb	0.242608	yes

Supplementary Table 3: XYZ (space group $P\bar{6}2m$) compounds with significant magnetism.

mp-1091404	UInRh	U	0.081975	In	0.253257	yes
mp-21494	USnCo	U	0.081587	Sn	0.249041	yes
mp-1080077	UGaPt	U	0.078517	Ga	0.256914	yes
mp-1079949	UAlPt	U	0.074677	Al	0.262571	yes
mp-5015	UAlRh	U	0.074114	Al	0.265481	yes
mp-1078870	UGaRh	U	0.078603	Ga	0.260529	yes
mp-1079086	UAlNi	U	0.071447	Al	0.264623	yes
mp-21320	UGaNi	U	0.071759	Ga	0.261885	yes
mp-1079219	UAlIr	U	0.07841	Al	0.263225	yes
mp-1080013	UAlRu	U	0.074845	Al	0.266171	yes
mp-1078880	UGaRu	U	0.076241	Ga	0.262093	yes
mp-19811	USnRu	U	0.083475	Sn	0.248264	yes
mp-1078586	UGaCo	U	0.0795095	Ga	0.2635175	yes

Supplementary Note 2. NEAREST NEIGHBOR INFORMATION OF CRRHAS

We summarize the nearest neighbor information to a reference Cr1 atom of CrRhAs in Supplementary Table 4. The in-plane distance is the projected bond length in the xy plane. n_{bonds} is the number of bonds that satisfy the in-plane distance constraint. $d_z = 0\,\text{Å}$, $d_z = 3.718\,\text{Å}$, $d_z = 7.436\,\text{Å}$, $d_z = 11.154\,\text{Å}$ correspond to successive Cr plane distances along the z direction, and Ji in each column is the hopping from the reference Cr1 atom to the corresponding Cr plane.

Supplementary Table 4. Nearest neighbor infomation of CrRhAs. d_z are distances between Cr planes along z direction. Distance $d_{\text{Cr}-\text{Cr}}$ is given in brackets for easy identification of the bond. In the naming scheme of the main paper, the first column of couplings is called $J_0, J_1, J_2, \ldots$, the second column is called $J_0', J_1', J_2', \ldots$, the third $J_0'', J_1'', J_2'', \ldots$, and so on.

d_{inplane} (Å)	Cr1 bond to	n_{bonds}	$d_z = 0\,\text{Å}$	$d_z = 3.718\,\text{Å}$	$d_z = 7.436\,\text{Å}$	$d_z = 11.154\,\text{Å}$
0.	{Cr1}	1	J0 (0 Å)	J2 (3.718 Å)	J9 (7.436 Å)	J25 (11.154 Å)
3.384	{Cr3,Cr2}	4	J1 (3.384 Å)	J4 (5.028 Å)	J12 (8.170 Å)	J26 (11.656 Å)
4.404	{Cr3,Cr2}	2	J3 (4.404 Å)	J5 (5.764 Å)	J14 (8.642 Å)	J29 (11.992 Å)
6.384	{Cr1}	6	J6 (6.384 Å)	J8 (7.388 Å)	J17 (9.800 Å)	J34 (12.852 Å)
6.653	{Cr2,Cr3}	2	J7 (6.653 Å)	J10 (7.622 Å)	J19 (9.978 Å)	J35 (12.988 Å)
7.756	{Cr3,Cr2}	4	J11 (7.756 Å)	J13 (8.601 Å)	J22 (10.745 Å)	J40 (13.585 Å)
9.221	{Cr3,Cr2}	4	J15 (9.221 Å)	J18 (9.942 Å)	J28 (11.846 Å)	J45 (14.472 Å)
9.642	{Cr3,Cr2}	4	J16 (9.642 Å)	J20 (10.334 Å)	J30 (12.176 Å)	J47 (14.744 Å)
10.433	{Cr3,Cr2}	4	J21 (10.433 Å)	J24 (11.076 Å)	J33 (12.812 Å)	J52 (15.273 Å)
11.057	{Cr1}	6	J23 (11.057 Å)	J27 (11.666 Å)	J38 (13.325 Å)	J58 (15.706 Å)
12.593	{Cr2,Cr3}	4	J31 (12.593 Å)	J36 (13.131 Å)	J46 (14.625 Å)	J68 (16.823 Å)
12.768	{Cr1}	6	J32 (12.768 Å)	J37 (13.298 Å)	J48 (14.776 Å)	J70 (16.954 Å)

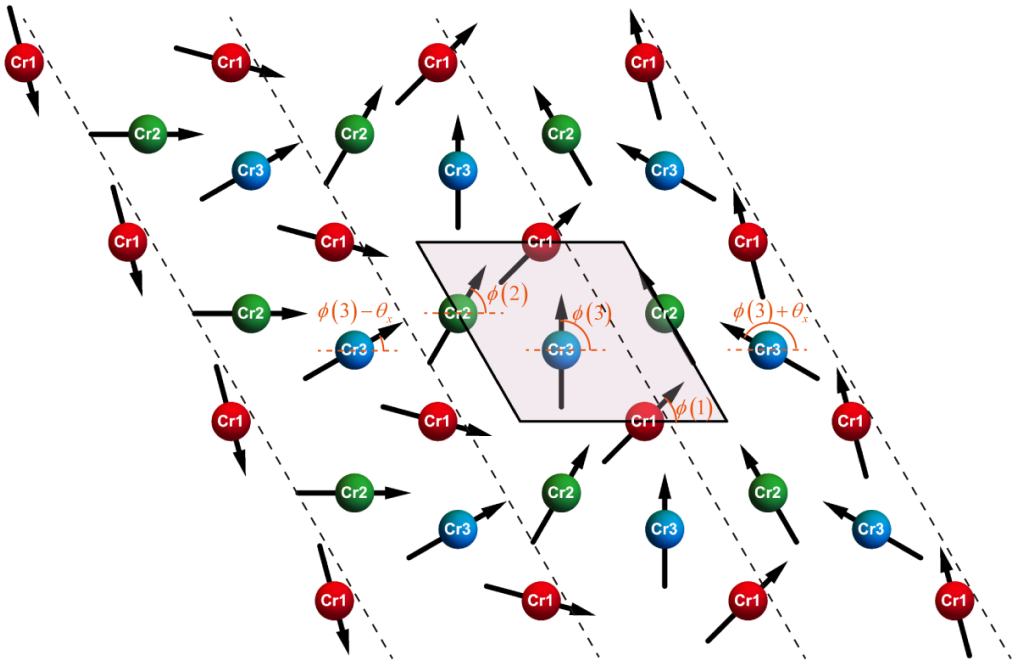
Supplementary Note 3. HAMILTONIAN COEFFICIENT EXPRESSIONS FOR $(q_x, 0, q_z)$ SPIRALS

We derived coefficient expressions $C_i, C_i', C_i'', ...$ and D, D', D'' for Eq. (5) in the main text. They are given in Supplementary Table 5.

Supplementary Table 5. Coefficient expression of C_i and D for $(q_x, 0, q_z)$ spin spirals.

E_0	1
C_1	$\cos(\theta_x - \phi(1) + \phi(2)) + \cos(\theta_x + \phi(2) - \phi(3)) + \cos(\phi(1) - \phi(2)) + 2\cos(\phi(1) - \phi(3)) + \cos(\phi(2) - \phi(3))$
C_2	$\cos(\theta_x - \phi(1) + \phi(2)) + \cos(\theta_x - \phi(1) + \phi(3)) + \cos(\phi(2) - \phi(3))$
C_3	$3(2\cos(\theta_x) + 1)$
C_4	$\cos(\theta_x + \phi(1) - \phi(3)) + \cos(\theta_x + \phi(2) - \phi(3)) + \cos(\phi(1) - \phi(2))$
C_0'	$3\cos(\theta_z)$
C_1'	$2\cos(\theta_z)(\cos(\theta_x - \phi(1) + \phi(2)) + \cos(\theta_x + \phi(2) - \phi(3)) + \cos(\phi(1) - \phi(2)) + 2\cos(\phi(1) - \phi(3)) + \cos(\phi(2) - \phi(3)))$
C_2'	$2\cos(\theta_z)(\cos(\theta_x - \phi(1) + \phi(2)) + \cos(\theta_x - \phi(1) + \phi(3)) + \cos(\phi(2) - \phi(3)))$
C_3'	$6(2\cos(\theta_x) + 1)\cos(\theta_z)$
C_4'	$2\cos(\theta_z)(\cos(\theta_x + \phi(1) - \phi(3)) + \cos(\theta_x + \phi(2) - \phi(3)) + \cos(\phi(1) - \phi(2)))$
C_0''	$3\cos(2\theta_z)$
C_1''	$2\cos(2\theta_z)(\cos(\theta_x - \phi(1) + \phi(2)) + \cos(\theta_x + \phi(2) - \phi(3)) + \cos(\phi(1) - \phi(2)) + 2\cos(\phi(1) - \phi(3)) + \cos(\phi(2) - \phi(3)))$
C_2''	$2\cos(2\theta_z)(\cos(\theta_x - \phi(1) + \phi(2)) + \cos(\theta_x - \phi(1) + \phi(3)) + \cos(\phi(2) - \phi(3)))$
C_3''	$6(2\cos(\theta_x) + 1)\cos(2\theta_z)$
C_4''	$2\cos(2\theta_z)(\cos(\theta_x + \phi(1) - \phi(3)) + \cos(\theta_x + \phi(2) - \phi(3)) + \cos(\phi(1) - \phi(2)))$
D_1	$\cos(\phi(1) - \phi(3))\cos(\theta_x - \phi(1) + \phi(2)) + \cos(\phi(1) - \phi(3))(\cos(\theta_x + \phi(2) - \phi(3)) + \cos(\phi(2) - \phi(3)) + 1) + \cos(\theta_x)\cos(\theta_x - \phi(1) + 2\phi(2) - \phi(3)) + \cos(\phi(1) - \phi(2))\cos(\phi(1) - \phi(3))$
D_2	$\frac{1}{2}(\cos(\phi(2) - \phi(3))(2\cos(\theta_x - \phi(1) + \phi(2)) + 2\cos(\theta_x - \phi(1) + \phi(3)) + 1) + \cos(2\theta_x - 2\phi(1) + \phi(2) + \phi(3)))$

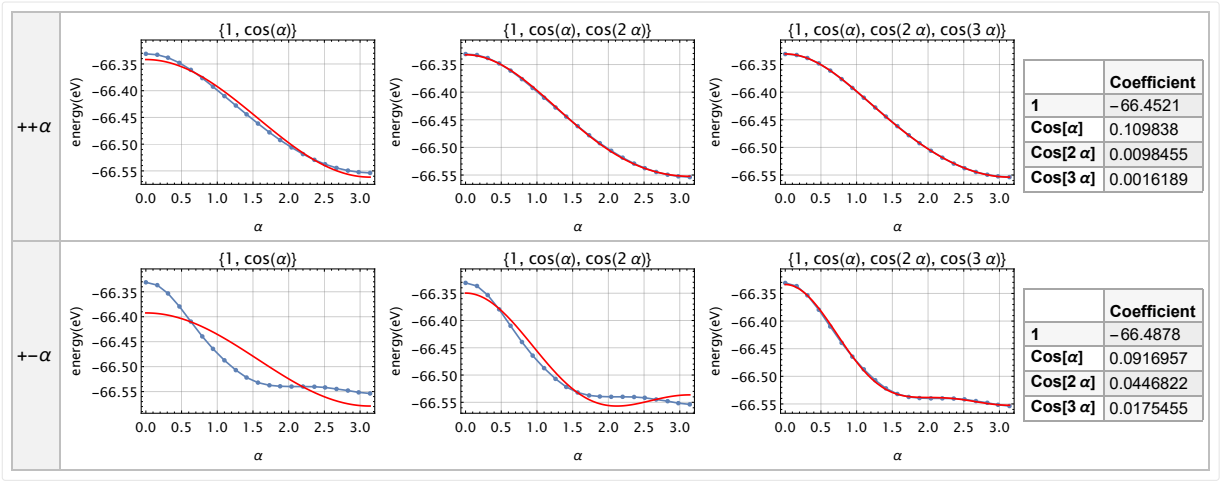
The definition of $\phi(1), \phi(2), \phi(3)$ and θ_x is illustrated in Supplementary Fig. 1. For θ_z, it is a common rotation angle between the current plane and the next plane of all moments.



Supplementary Figure 1. An illustration of a $(q_x, 0, 0)$ spiral. $\phi(1), \phi(2), \phi(3)$ are the polar angles of Cr1, Cr2 and Cr3 moments. θ_x is the spiral angle going from one strip marked by dashed lines to the next one.

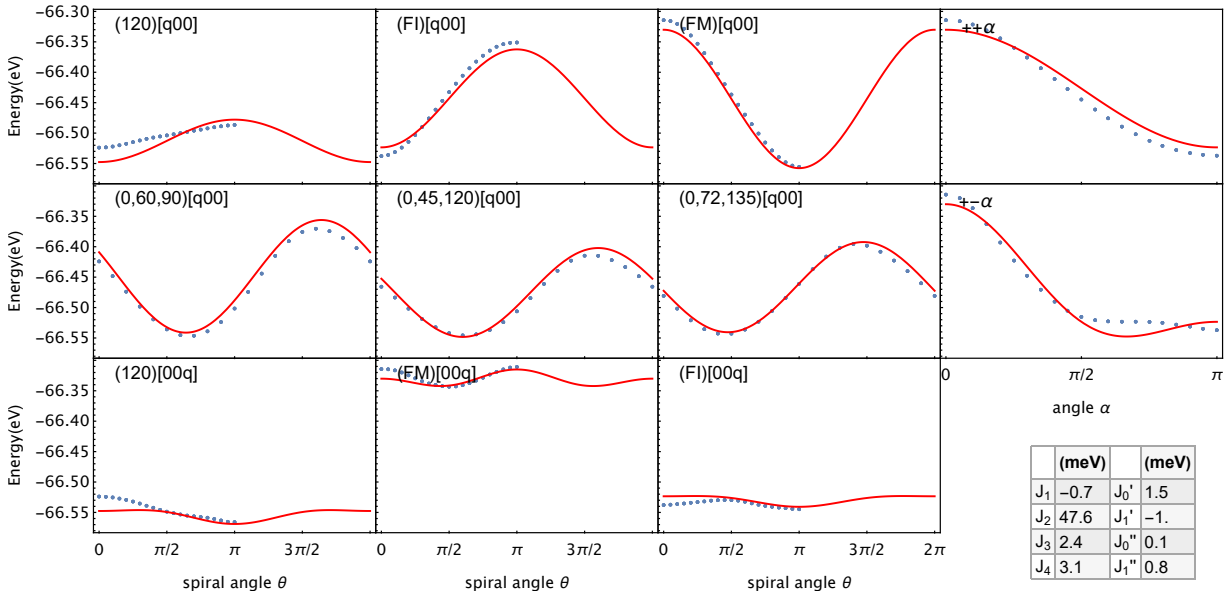
Supplementary Note 4. ADDITIONAL ATTEMPTS OF FITTING DFT TOTAL ENERGIES

Supplementary Figure 2 shows a simple $\cos(\alpha)$ fit of the noncollinear periodic calculations (the $++\alpha$ and $+-\alpha$ series). The deviations of the fit indicate that higher harmonics are necessary for a good fit, *i.e.* a pure Heisenberg Hamiltonian cannot completely represent the noncollinear DFT energies.



Supplementary Figure 2. Pure cosine fitting for $++\alpha$ and $+-\alpha$ series

In Supplementary Figure 3, we show a fit of all spiral energies with Heisenberg exchange but without ring exchange terms. The quality of fit is clearly lower than the full fit shown in Fig. 5 of the main text, especially at $\theta = 0$ for (120)[q00] and (120)[00q].



Supplementary Figure 3. Fitting all spiral energy curves without ring exchange

[S1] A. Jain, S. P. Ong, G. Hautier, W. Chen, W. D. Richards, S. Dacek, S. Cholia, D. Gunter, D. Skinner, G. Ceder, and K. A. Persson, Commentary: The Materials Project: A materials genome approach to accelerating materials innovation, APL Mater. **1**, 011002 (2013).