Supplemental Materials for "Determining the range of magnetic interactions from the relations between magnon eigenvalues at high-symmetry k points"

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A. The symmetry restrictions on the magnetic interactions

As mentioned in the maintext, the microscopic magnetic model with proper parameters is extremely important. Note that the crystal symmetry impose restrictions on the magnetic model and its parameters. Here we consider a general pairwise spin model as shown in the maintext

$$H = \sum_{l,n,l',n'} \mathbf{S}_{ln} \cdot \mathbf{J}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} \cdot \mathbf{S}_{l'n'} \tag{1}$$

where $\mathbf{J}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}}$, a 3×3 tensor, represents the spin exchange parameters. R_l and τ_n represent the lattice translation vector and the position of magnetic ions in the lattice basis, and $\mathbf{S}_{l'n'}$ means the spin at the site of $\mathbf{R}_l + \boldsymbol{\tau}_n$ Translation symmetry will restrict $\mathbf{J}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}}$ to be only related to $\mathbf{J}_{\boldsymbol{\tau}_n,\boldsymbol{\tau}_{\boldsymbol{n'}}+\mathbf{R}_{l''}}$ where $R_{l''} = R_{l'} - R_l$, irrespective of the starting unit cell. Other spatial symmetries will also give restrictions on the magnetic exchange interactions (MEIs). We consider a general space group element $\{\alpha|\mathbf{t}\}$, where the left part represents the rotation and the right part means the lattice translation. Supposing under this symmetry operator, $\mathbf{R}_m + \boldsymbol{\tau}_p$ and $\mathbf{R}_{m'} + \boldsymbol{\tau}_{\boldsymbol{p'}}$ transfer to $\mathbf{R}_l + \boldsymbol{\tau}_n$ and $\mathbf{R}_{l'} + \boldsymbol{\tau}_{\boldsymbol{n'}}$, respectively, meanwhile the transformation of spin becomes $\mathbf{S}_{mp} = M(\alpha)\mathbf{S}_{ln}$, where $M(\alpha)$ is the representation matrix of the proper rotation part of the operation α in the coordinate system, we get the following expression:

$$\begin{aligned} H &= \sum_{l,n,l',n'} \mathbf{S}_{ln} \cdot \mathbf{J}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} \cdot \mathbf{S}_{l'n'} \\ &= \sum_{l,n,l',n'} \mathbf{S}_{ln} M^\dagger(\alpha) M(\alpha) \mathbf{J}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} M^\dagger(\alpha) M(\alpha) \mathbf{S}_{l'n'} \\ &= \sum_{m,p,m',p'} \mathbf{S}_{mp} \cdot M(\alpha) \mathbf{J}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} M^\dagger(\alpha) \cdot \mathbf{S}_{m'p'} \end{aligned} \tag{2}$$

Then the exchange interactions should satisfy the following condition:

$$\mathbf{J}_{\mathbf{R}_m+\boldsymbol{\tau}_p,\mathbf{R}_{m'}+\boldsymbol{\tau}_{\boldsymbol{p'}}} = M(\alpha) \mathbf{J}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} M^\dagger(\alpha) \tag{3}$$

After decomposing the 3×3 tensor $\mathbf{J}$ into scalar Heisenberg term J and vector DM term D as in the maintext, we obtain the following results:

$$\begin{aligned} J_{\mathbf{R}_m+\boldsymbol{\tau}_p,\mathbf{R}_{m'}+\boldsymbol{\tau}_{\boldsymbol{p'}}} &= J_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} \\ \mathbf{D}_{\mathbf{R}_m+\boldsymbol{\tau}_p,\mathbf{R}_{m'}+\boldsymbol{\tau}_{\boldsymbol{p'}}} &= M(\alpha) \mathbf{D}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} \end{aligned} \tag{4}$$

Meanwhile, it is should be noted that the Heisenberg and DM interactions obey the following commutation relations

$$\begin{aligned} J_{\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}},\mathbf{R}_l+\boldsymbol{\tau}_n} &= J_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} \\ \mathbf{D}_{\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}},\mathbf{R}_l+\boldsymbol{\tau}_n} &= -\mathbf{D}_{\mathbf{R}_l+\boldsymbol{\tau}_n,\mathbf{R}_{l'}+\boldsymbol{\tau}_{\boldsymbol{n'}}} \end{aligned} \tag{5}$$

According to the above equations (i.e. Eq. (4) and (5)), one can obtain the symmetry restricted MEIs for any space group. Similarly, for the magnetic space group, the symmetry restriction on MEIs can also be easily obtained. The collinear ferromagnetic system shown in maintext (i.e. BNS 85.59 case) have two generators: the four-fold rotation $\{4^+_{001}|1/2,0,0\}$ and inversion operation $\{\bar{1}|0,0,0\}$. The magnetic ion located at τ_1 position has only two nearest neighbors, i.e. $(\boldsymbol{\tau}_1, \boldsymbol{\tau}_5 + \mathbf{R}_{-100})$ pair and $(\boldsymbol{\tau}_1, \boldsymbol{\tau}_6 + \mathbf{R}_{0-10})$ pair as shown in Table I. These two bonds are equivalent by the inversion symmetry $\{\bar{1}|0,0,0\}$. Meanwhile, performing the four-fold rotation symmetry for the above two pairs, we can get other six pairs in a unit cell, and there are in total eight NN in a unit cell. Based on Eq. (4), it is also easy to prove all of these eight NN exchange paths has the same Heisenberg term, which we denote as J_1 as shown in Table I. The non-collinearity shown in maintext reduces the four-fold rotation $\{4^+_{001}|1/2,0,0\}$ to the two-fold rotation operation $\{2_{001}|1/2,1/2,0\}$, as a result the eight NN exchange path is no longer equivalent as indicated in Table I. Similarly, one can obtain the symmetry restriction on the DM interactions, as also shown in Table I.

For simplicity, we only list the corresponding MEIs for longer range with the collinear FM example (i.e. the case with symmetry of BNS 85.59) and non-collinear example

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TABLE I. The distances and the bond information of corresponding MEIs (with $R_{cut} = 0.5\ a$) for the non-collinear magnetic example in the maintext (i.e. the case with symmetry of BNS 13.65). We also list the results by using the symmetry of BNS 85.59, which is applicable for the collinear FM case as well as the non-collinear case with localized magnetism in the right part.

distance(a)	n	n'	R_l	BNS 13.65		BNS 85.59	
0.35	1	5	$(-1,0,0)$	J_1	(D_1^x, D_1^y, D_1^z)	J_1	(D_1^x, D_1^y, D_1^z)
	1	6	$(0,-1,0)$	J_1	(D_1^x, D_1^y, D_1^z)	J_1	(D_1^x, D_1^y, D_1^z)
	2	5	$(0,0,0)$	J_2	(D_2^x, D_2^y, D_2^z)	J_1	$(-D_1^y, D_1^x, D_1^z)$
	2	6	$(0,-1,0)$	J_2	(D_2^x, D_2^y, D_2^z)	J_1	$(-D_1^y, D_1^x, D_1^z)$
	3	5	$(-1,0,0)$	J_2	$(-D_2^x, -D_2^y, D_2^z)$	J_1	$(D_1^y, -D_1^x, D_1^z)$
	3	6	$(0,0,0)$	J_2	$(-D_2^x, -D_2^y, D_2^z)$	J_1	$(D_1^y, -D_1^x, D_1^z)$
	4	5	$(0,0,0)$	J_1	$(-D_1^x, -D_1^y, D_1^z)$	J_1	$(-D_1^x, -D_1^y, D_1^z)$
	4	6	$(0,0,0)$	J_1	$(-D_1^x, -D_1^y, D_1^z)$	J_1	$(-D_1^x, -D_1^y, D_1^z)$
0.36	1	7	$(0,0,0)$	J_3	(D_3^x, D_3^y, D_3^z)	J_2	(D_2^x, D_2^y, D_2^z)
	1	8	$(-1,-1,0)$	J_3	(D_3^x, D_3^y, D_3^z)	J_2	(D_2^x, D_2^y, D_2^z)
	2	7	$(0,0,0)$	J_4	(D_4^x, D_4^y, D_4^z)	J_2	$(-D_2^y, D_2^x, D_2^z)$
	2	8	$(0,-1,0)$	J_4	(D_4^x, D_4^y, D_4^z)	J_2	$(-D_2^y, D_2^x, D_2^z)$
	3	7	$(0,0,0)$	J_4	$(-D_4^x, -D_4^y, D_4^z)$	J_2	$(D_2^y, -D_2^x, D_2^z)$
	3	8	$(-1,0,0)$	J_4	$(-D_4^x, -D_4^y, D_4^z)$	J_2	$(D_2^y, -D_2^x, D_2^z)$
	4	7	$(0,0,0)$	J_3	$(-D_3^x, -D_3^y, D_3^z)$	J_2	$(-D_2^x, -D_2^y, D_2^z)$
	4	8	$(0,0,0)$	J_3	$(-D_3^x, -D_3^y, D_3^z)$	J_2	$(-D_2^x, -D_2^y, D_2^z)$
0.5	1	2	$(0,0,0)$	J_5	(D_5^x, D_5^y, D_5^z)	J_3	(D_3^x, D_3^y, D_3^z)
	1	2	$(-1,0,0)$	J_5	(D_5^x, D_5^y, D_5^z)	J_3	(D_3^x, D_3^y, D_3^z)
	1	3	$(0,0,0)$	J_6	(D_6^x, D_6^y, D_6^z)	J_3	$(-D_3^y, D_3^x, -D_3^z)$
	1	3	$(0,-1,0)$	J_6	(D_6^x, D_6^y, D_6^z)	J_3	$(-D_3^y, D_3^x, -D_3^z)$
	2	4	$(0,0,0)$	J_6	$(D_6^x, D_6^y, -D_6^z)$	J_3	$(-D_3^y, D_3^x, D_3^z)$
	2	4	$(0,-1,0)$	J_6	$(D_6^x, D_6^y, -D_6^z)$	J_3	$(-D_3^y, D_3^x, D_3^z)$
	3	4	$(0,0,0)$	J_5	$(D_5^x, D_5^y, -D_5^z)$	J_3	$(D_3^x, D_3^y, -D_3^z)$
	3	4	$(-1,0,0)$	J_5	$(D_5^x, D_5^y, -D_5^z)$	J_3	$(D_3^x, D_3^y, -D_3^z)$

TABLE II. The corresponding MEIs for the collinear FM example (i.e. the case with symmetry of BNS 85.59) and non-collinear example (i.e. the case with symmetry of BNS 13.65) shown in maintext.

BNS 85.59	BNS 13.65
J_1	J_1, J_2
J_2	J_3, J_4
J_3	J_5, J_6
J_4	J_7, J_8
$J_5 \tilde{} J_7$	$J_9 \tilde{} J_{14}$
J_8	J_{15}, J_{16}
J_9, J_{10}	$J_{17} \tilde{} J_{20}$
J_{11}, J_{12}	$J_{21} \tilde{} J_{24}$
$J_{13} \tilde{} J_{15}$	$J_{25} \tilde{} J_{28}$
J_{16}	J_{29}, J_{30}
J_{17}, J_{18}	$J_{32} \tilde{} J_{34}$
J_{19}	J_{35}, J_{36}
J_{20}, J_{21}	$J_{37} \tilde{} J_{40}$
J_{22}	J_{41}, J_{42}
J_{23}	J_{43}, J_{44}
$J_{24} \tilde{} J_{27}$	$J_{45} \tilde{} J_{52}$

in the maintext (i.e. the case with symmetry of BNS 13.65), as shown in Table II.

It is worth mentioning that Eq. (3) can also give symmetry restrictions on SIA. The general quadratic expression of SIA could be written as $\sum_{l,n,\alpha,\beta} K^{\alpha,\beta}_{\mathbf{R}_l+\boldsymbol{\tau}_n} S^{\alpha}_{ln} S^{\beta}_{ln}$. Note that the SIA term should be naturally symmetric, i.e. $K^{\alpha,\beta}_{\mathbf{R}_l+\boldsymbol{\tau}_n} = K^{\beta,\alpha}_{\mathbf{R}_l+\boldsymbol{\tau}_n}$. According to Eq. (3), when the action of symmetry operation $\{\alpha|t\}$ keeps the position $R_l + \tau_n$ unchanged, we have $K_{\mathbf{R}_l+\boldsymbol{\tau}_n} = M(\alpha) K_{\mathbf{R}_l+\boldsymbol{\tau}_n} M^{\dagger}(\alpha)$. For the typical FM collinear magnetic material (BNS 85.59) in the maintext, the SIA term for magnetic ions located at $2a$ and $2c$ Wyckoff positions should satisfy that

$$K_{\mathbf{R}_l+\boldsymbol{\tau}_n} = \begin{bmatrix} K^{xx}_{\mathbf{R}_l+\boldsymbol{\tau}_n} & & \\ & K^{xx}_{\mathbf{R}_l+\boldsymbol{\tau}_n} & \\ & & K^{zz}_{\mathbf{R}_l+\boldsymbol{\tau}_n} \end{bmatrix}$$

which is actually the usual form $H_{SIA} = \sum_{l,n} K_{\mathbf{R}_l+\boldsymbol{\tau}_n} (S^z_{ln})^2 + C$ where $K_{\mathbf{R}_l+\boldsymbol{\tau}_n} = K^{zz}_{\mathbf{R}_l+\boldsymbol{\tau}_n} - K^{xx}_{\mathbf{R}_l+\boldsymbol{\tau}_n}$ and C represents the constant term. However, the symmetry does not give restrictions on the SIA term of magnetic ions at $4d$ position. In the maintext, we adopt the usual form $H_{SIA} = \sum_{l,n} K (S^z_{ln})^2$ for simplicity.

B. The parameters $A_{n,n'}$, $B_{n,n'}$, $C_{n,n'}$, $\mathbf{O}_{n,n'}$, $\mathbf{P}_{n,n'}$ and $\mathbf{Q}_{n,n'}$

In the maintext, considering the spin model including the Heisenberg and DM interactions, we perform the standard LSWT and obtain spin Hamiltonian as Eq. (4) in the maintext, where the parameters $A_{n,n'}$, $B_{n,n'}$, $C_{n,n'}$, $\mathbf{O}_{n,n'}$, $\mathbf{P}_{n,n'}$ and $\mathbf{Q}_{n,n'}$ are related to the spin directions at n and n' sites. Here $A_{n,n'}$, $B_{n,n'}$ and $C_{n,n'}$

could be written as:

$$\begin{aligned}A_{n,n'} = &\frac{1}{4}\cos(\theta_n-\theta_{n'})-\frac{1}{4}\cos(\theta_n+\theta_{n'})+\frac{1}{2}\cos(\phi_n-\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n-\theta_{n'}+\phi_n-\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n+\theta_{n'}+\phi_n-\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n-\theta_{n'}-\phi_n+\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n+\theta_{n'}-\phi_n+\phi_{n'})\\&+\frac{i}{4}\sin(\theta_n+\phi_n-\phi_{n'})+\frac{i}{4}\sin(\theta_{n'}+\phi_n-\phi_{n'})\\&-\frac{i}{4}\sin(\theta_n-\phi_n+\phi_{n'})-\frac{i}{4}\sin(\theta_{n'}-\phi_n+\phi_{n'})\end{aligned}\quad(6)$$

$$B_{n,n'} = \sin\theta_n\sin\theta_{n'}\cos(\phi_n-\phi_{n'})+\cos\theta_n\cos\theta_{n'}\quad(7)$$

$$\begin{aligned}C_{n,n'} = &\frac{1}{4}\cos(\theta_n-\theta_{n'})-\frac{1}{4}\cos(\theta_n+\theta_{n'})-\frac{1}{2}\cos(\phi_n-\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n-\theta_{n'}+\phi_n-\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n+\theta_{n'}+\phi_n-\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n-\theta_{n'}-\phi_n+\phi_{n'})\\&+\frac{1}{8}\cos(\theta_n+\theta_{n'}-\phi_n+\phi_{n'})\\&-\frac{i}{4}\sin(\theta_n+\phi_n-\phi_{n'})+\frac{i}{4}\sin(\theta_{n'}+\phi_n-\phi_{n'})\\&+\frac{i}{4}\sin(\theta_n-\phi_n+\phi_{n'})-\frac{i}{4}\sin(\theta_{n'}-\phi_n+\phi_{n'})\end{aligned}\quad(8)$$

Meanwhile, the parameters $\mathbf{O}_{n,n'} = (O^x_{n,n'}, O^y_{n,n'}, O^z_{n,n'})$, $\mathbf{P}_{n,n'} = (P^x_{n,n'}, P^y_{n,n'}, P^z_{n,n'})$ and $\mathbf{Q}_{n,n'} = (Q^x_{n,n'}, Q^y_{n,n'}, Q^z_{n,n'})$ related to the spin directions at n and n' sites could be written as:

$$\begin{aligned}O^x_{n,n'} = &\frac{1}{2}[-\cos\theta_n\sin\theta_{n'}\sin\phi_n+\sin\theta_n\cos\theta_{n'}\sin\phi_{n'}\\&+i(\sin\theta_n\cos\phi_{n'}+\sin\theta_{n'}\cos\phi_n)]\end{aligned}\quad(9)$$

$$P^x_{n,n'} = \sin\theta_n\cos\theta_{n'}\sin\phi_n-\cos\theta_n\sin\theta_{n'}\sin\phi_{n'}\quad(10)$$

$$\begin{aligned}Q^x_{n,n'} = &\frac{1}{2}[-\cos\theta_n\sin\theta_{n'}\sin\phi_n+\sin\theta_n\cos\theta_{n'}\sin\phi_{n'}\\&+i(\sin\theta_n\cos\phi_{n'}-\sin\theta_{n'}\cos\phi_n)]\end{aligned}\quad(11)$$

$$\begin{aligned}O^y_{n,n'} = &\frac{1}{2}[-\cos\theta_n\sin\theta_{n'}\cos\phi_n+\sin\theta_n\cos\theta_{n'}\cos\phi_{n'}\\&+i(-\sin\theta_n\sin\phi_{n'}+\sin\theta_{n'}\sin\phi_n)]\end{aligned}\quad(12)$$

$$P^y_{n,n'} = \sin\theta_n\cos\theta_{n'}\cos\phi_n-\cos\theta_n\sin\theta_{n'}\cos\phi_{n'}\quad(13)$$

$$\begin{aligned}Q^y_{n,n'} = &\frac{1}{2}[-\cos\theta_n\sin\theta_{n'}\cos\phi_n+\sin\theta_n\cos\theta_{n'}\cos\phi_{n'}\\&+i(-\sin\theta_n\sin\phi_{n'}-\sin\theta_{n'}\sin\phi_n)]\end{aligned}\quad(14)$$

$$\begin{aligned}O^z_{n,n'} = &\frac{1}{2}[(1+\cos\theta_n\cos\theta_{n'})\sin(\phi_n-\phi_{n'})\\&-i(\cos\theta_n+\cos\theta_{n'})\cos(\phi_n-\phi_{n'})]\end{aligned}\quad(15)$$

$$P^z_{n,n'} = \sin\theta_n\sin\theta_{n'}\sin(\phi_n-\phi_{n'})\quad(16)$$

$$\begin{aligned}Q^z_{n,n'} = &\frac{1}{2}[(-1+\cos\theta_n\cos\theta_{n'})\sin(\phi_n-\phi_{n'})\\&-i(\cos\theta_n-\cos\theta_{n'})\cos(\phi_n-\phi_{n'})]\end{aligned}\quad(17)$$

If the ground state of this magnetic system is collinear, which means that $\theta_n = \theta_{n'}$ (when magnetic ions at n and n' sites are ferromagnetic) or $\theta_n = \theta_{n'} + \pi$ (when magnetic ions at n and n' sites are antiferromagnetic) and $\phi_n = \phi_{n'}$, the parameters $A_{n,n'}$, $B_{n,n'}$, $C_{n,n'}$ can be simplified as

$$A_{n,n'} = \frac{\zeta_{n,n'}+1}{2}\quad(18)$$

$$B_{n,n'} = \zeta_{n,n'}\quad(19)$$

$$C_{n,n'} = \frac{\zeta_{n,n'}-1}{2}\quad(20)$$

where $\zeta_{n,n'}$ equals to 1 when the spins for the n and n' sites are parallel, otherwise $\zeta_{n,n'}$ equals to -1. More specially, when the ground state of this magnetic system is assumed to be collinear ferromagnetic [PS: do not need to be along z-direction], which means that $\theta_n = \theta_{n'}$ and $\phi_n = \phi_{n'}$, the parameters $A_{n,n'}$, $B_{n,n'}$, $C_{n,n'}$ can be simplified as

$$A_{n,n'} = 1\quad(21)$$

$$B_{n,n'} = 1\quad(22)$$

$$C_{n,n'} = 0\quad(23)$$

C. The eigenvalue problem of magnon eigenvalues

As shown in the maintext, following the LSWT, a general pairwise spin Hamiltonian could be written as Eq. (3) in the maintext. However, it should be noted that the operators in $\psi^\dagger(\mathbf{k}) = [a_1^\dagger(\mathbf{k}), ..., a_N^\dagger(\mathbf{k}), a_1(-\mathbf{k}), ..., a_N(-\mathbf{k})]$ in the Eq. (3) of the maintext satisfy the commutation relation

$$[\psi(k), \psi^\dagger(k)] = \begin{bmatrix} I & \\ & -I \end{bmatrix} = I_-\quad(24)$$

where I represents N×N identity matrix, and N represents the number of magnetic ions in an unit cell. To diagonalize the boson pairing Hamiltonian, we can solve the eigenvalue problem of the general Hamiltonian $H_J(\mathbf{k}) = I_- H(\mathbf{k})$ (i.e., Eq. (6) of the maintext). The first N diagonal elements are the energies of the normal spin wave modes $\omega_{k,n}$ and the last N eigenvalues are equal to the first N eigenvalues multiplied by minus one.

D. Instructions on the program of general relations between magnon eigenvalues

As shown in the maintext, a general pairwise spin model could be expanded as the isotropic Heisenberg Hamiltonian, the DM interactions, and the anisotropic symmetric terms, as shown in Eq. (2) in the maintext. We ignore the third term and perform LSWT to obtain the quadratic spin Hamiltonian as shown in Eq. (3) in the maintext. Note that there is a simple relation between the Fourier transform of MEIs and the SSME, consequently one can easily calculate SSME at arbitrary k point in BZ. Thus, different with the conventional group symmetry analysis, which give the relationship between the magnon energies at the symmetry-related k points, our method produces the relationships between the SSME at high symmetry k points subjected to R_{cut}. We propose a method to obtain the relations between SSME of different high-symmetry k-points. For the given magnetic system in the maintext, the algorithm of the proposed method is implemented in the Mathematica notebook "SR.nb". Using this code, one should first enter the information of magnetic materials, including: (1) the primitive basis and conventional basis; (2) the positions of magnetic atoms; (3) the (magnetic) space group; (4) the spin directions of magnetic atoms; (5) the range of MEIs to be considered (including Heisenberg and DM interactions); (6) whether to consider SIA; (7) the interested k-points. Then the relations between SSME can be automatically obtained. To catch your eye, we have used red color to indicate that the following variable should be specified in the notebook "SR.nb".

The typical magnetic system has the magnetic structure with the magnetic space group (BNS 85.59). As shown in Table I in the maintext, the lattice constant c/a is 0.8. The magnetic ions are located at three nonequivalent crystallographic sites: $4d$ (0, 0, 0), $2a$ (0.25, 0.75, 0) and $2c$ (0.25, 0.25, z) WP and the positions for these eight magnetic ions are summarized in Table I in the maintext. While the $4d$ and $2a$ WP had been completely determined by the symmetry, the coordinates of $2c$ WP have a variable z and here we adopt it as $z = 0.1$. The magnetic state is a collinear ferromagnetic order with spin along the z direction. This case belongs to the type-I magnetic space group (BNS 85.59), and all of the the polar angle and azimuthal angles are equal to 0.

In this notebook "SR.nb", one should specify the parameters as input information, such as:

(*input parameters:*)
(*primitive lattice basis*)
A = {{1, 0, 0}, {0, 1, 0}, {0, 0, 0.8}};
(*conventional lattice basis*)
AA = {{1, 0, 0}, {0, 1, 0}, {0, 0, 0.8}};
(*input positions of magnetic atoms based on conventional lattice basis vectors*)
atoms=8;
$\boldsymbol{\tau}$ = {{0, 0, 0},
{0.5, 0, 0},
{0, 0.5, 0},
{0.5, 0.5, 0},
{0.75, 0.25, 0.},
{0.25, 0.75, 0.},
{0.25, 0.25, 0.1},
{0.75, 0.75, -0.1}};
(*input the serial number of space group or the magnetic space group (in BNS notation).*)
msg=85.59;
(*spin directions of magnetic atoms in spherical coordinates (θ_n, ϕ_n) with the polar angles θ_n and azimuthal angles ϕ_n.*)
ang = {{0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0,0}};
(*the range of Heisenberg and DM interactions to be considered, respectively*)
Jmax = 3;
Dmax = 2;
(*set DIA = "True" or "False" to indicate whether to consider single ion anisotropy.*)
DIA = False;
(*high-symmetry points*)
kk[1] = {0, 0, 0};
kk[2] = {0.5, 0, 0};
kk[3] = {0.5, 0.5, 0};
kk[4] = {0, 0, 0.5};
kk[5] = {0.5, 0, 0.5};
kk[6] = {0.5, 0.5, 0.5};
kname = {"Γ", "X", "M", "Z", "R", "A"};

In the following, we would like to give a description of these parameters one by one:

(1) the primitive basis and conventional basis of Bravais lattice

One should input them in Cartesian coordinates. As the example in the maintext, the space group P4/n (No. 85) crystallizes in a tetragonal lattice, and its primitive basis A and conventional basis AA of Bravais lattice are both $\{\{1,0,0\},\{0,1,0\},\{0,0,0.8\}\}$.

(*primitive lattice basis*)
A = {{1, 0, 0}, {0, 1, 0}, {0, 0, 0.8}};
(*conventional lattice basis*)
AA = {{1, 0, 0}, {0, 1, 0}, {0, 0, 0.8}};

(2) the number and the Wyckoff positions of magnetic atoms (based on conventional lattice basis vectors)

(*input positions of magnetic atoms based on conventional lattice basis vectors*)
atoms=8;
$\boldsymbol{\tau}$ = {{0, 0, 0},
{0.5, 0, 0},
{0, 0.5, 0},
{0.5, 0.5, 0},
{0.75, 0.25, 0.},
{0.25, 0.75, 0.},
{0.25, 0.25, 0.1},

{0.75, 0.75, -0.1}};

(3) the (magnetic) space group of the magnetic system. When the magnetic moments are quite localized, magnetic interactions may still satisfy the symmetries of its space group. In this case, one can enter the serial number of its space group. Otherwise, one should enter the serial number of its magnetic space group (in BNS notation). As the example for collinear ferromagnetic state in the maintext, the magnetic space group (BNS 85.59) has the same symmetries as the space group (SG. 85). In this case, the results of the input "msg=85" and "msg=85.59" are equivalent. Note that if the input parameter is an integer "X", we would use the symmetry of this space group (No. X), otherwise, we will use the symmetry of the magnetic space group as "BNS X.Y".

(*input the serial number of space group or the magnetic space group (in BNS notation).*)

msg=85.59;

(4) the spin directions of magnetic atoms. One should input them in spherical coordinates (θ_n, ϕ_n) with the polar angles θ_n and azimuthal angles ϕ_n. As the example in the maintext, the FM (001) state should be

(*spin directions of magnetic atoms in spherical coordinates (θ_n, ϕ_n) with the polar angles θ_n and azimuthal angles ϕ_n.*)

ang = {{0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0,0}};

(5) One should set up the range of magnetic interactions to be considered. You can set the range of Heisenberg and DM interactions respectively.

(*the range of Heisenberg and DM interactions to be considered, respectively*)

Jmax = 3;

Dmax = 2;

For example, we set Jmax = 3 and Dmax = 2, which means that we consider the range of Heisenberg interactions up to J_3, while the DM interactions are considered up to $\mathbf{D}_2$.

(6) Then one should set up whether to consider SIA by setting DIA = "True" or "False".

(*set DIA = "True" or "False" to indicate whether to consider single ion anisotropy.*)

DIA = False;

(7) Last, one should input the information of high-symmetry points to be considered, including the positions of these high-symmetry points and the labeled names of these high-symmetry points.

(*high-symmetry points*)

kk[1] = {0, 0, 0};
kk[2] = {0.5, 0, 0};
kk[3] = {0.5, 0.5, 0};
kk[4] = {0, 0, 0.5};
kk[5] = {0.5, 0, 0.5};
kk[6] = {0.5, 0.5, 0.5};
kname = {"Γ", "X", "M", "Z", "R", "A"};

After entering the above parameters, our program would output the following information, including:

TABLE III. The Heisenberg and DM interactions for the NNs of the typical material in the maintext for collinear FM state (BNS 85.59) restricted by the crystal symmetry.

distance(a)	n	n'	R_l	J	D
0.35	1	5	$(-1,0,0)$	J_1	(D_1^x, D_1^y, D_1^z)
	1	6	$(0,-1,0)$		(D_1^x, D_1^y, D_1^z)
	2	5	$(0,0,0)$		$(-D_1^y, D_1^x, D_1^z)$
	2	6	$(0,-1,0)$		$(-D_1^y, D_1^x, D_1^z)$
	3	5	$(-1,0,0)$		$(D_1^y, -D_1^x, D_1^z)$
	3	6	$(0,0,0)$		$(D_1^y, -D_1^x, D_1^z)$
	4	5	$(0,0,0)$		$(-D_1^x, -D_1^y, D_1^z)$
	4	6	$(0,0,0)$		$(-D_1^x, -D_1^y, D_1^z)$
0.36	1	7	$(0,0,0)$	J_2	(D_2^x, D_2^y, D_2^z)
	1	8	$(-1,-1,0)$		(D_2^x, D_2^y, D_2^z)
	2	7	$(0,0,0)$		$(-D_2^y, D_2^x, D_2^z)$
	2	8	$(0,-1,0)$		$(-D_2^y, D_2^x, D_2^z)$
	3	7	$(0,0,0)$		$(D_2^y, -D_2^x, D_2^z)$
	3	8	$(-1,0,0)$		$(D_2^y, -D_2^x, D_2^z)$
	4	7	$(0,0,0)$		$(-D_2^x, -D_2^y, D_2^z)$
	4	8	$(0,0,0)$		$(-D_2^x, -D_2^y, D_2^z)$
0.5	1	2	$(0,0,0)$	J_3	(D_3^x, D_3^y, D_3^z)
	1	2	$(-1,0,0)$		(D_3^x, D_3^y, D_3^z)
	1	3	$(0,0,0)$		$(-D_3^y, D_3^x, -D_3^z)$
	1	3	$(0,-1,0)$		$(-D_3^y, D_3^x, -D_3^z)$
	2	4	$(0,0,0)$		$(-D_3^y, D_3^x, D_3^z)$
	2	4	$(0,-1,0)$		$(-D_3^y, D_3^x, D_3^z)$
	3	4	$(0,0,0)$		$(D_3^x, D_3^y, -D_3^z)$
	3	4	$(-1,0,0)$		$(D_3^x, D_3^y, -D_3^z)$

(1) the symmetry of the (magnetic) space group.

output[symmetry]=
(x,y,z | mx,my,mz)
(-y+1/2,x,z | -my,mx,mz)
(y,-x+1/2,z | my,-mx,mz)
(-x+1/2,-y+1/2,z | -mx,-my,mz)
(-x,-y,-z | mx,my,mz)
(y+1/2,-x,-z | -my,mx,mz)
(-y,x+1/2,-z | my,-mx,mz)
(x+1/2,y+1/2,-z | -mx,-my,mz)

where the left part represents the symmetry operation for positions of magnetic atoms, while the right part means the symmetry operation for the orientation of magnetic moment.

(2) The program would also give the distance and the corresponding symmetry restricted MEIs, including the Heisenberg and DM interactions. For example, there are in total 24 MEIs in this magnetic system up to J_3, as summarized in the Table III. Meanwhile, the corresponding symmetry restricted DM interactions $\mathbf{D}(\boldsymbol{\tau}_n, \boldsymbol{\tau}_{n'}, \mathbf{R}_l)$ are also listed here. Note that for 5th NN MEIs, the symmetry makes $D_5^z = 0$, while for 7th NN MEIs, we have

$\mathbf{D}_7 = (0, 0, 0)$. These symmetry restrictions would also be automatically considered in our program.

(3) the main output: the relations between SSME at different high-symmetry k points.

output[relations]=

$M - A = 0$

$X - R = 0$

$\Gamma - Z = 0$

$\Gamma - 2X + M = 0$

where the label "M" means the quadratic sum of the magnon energies at M point "$\sum_i \omega_i^2(M)$", as well as the labels of other high-symmetry k points. We can see that, up to J_3, the quadratic sum of the magnon energies satisfy that $\sum_i \omega_i^2(\Gamma) = \sum_i \omega_i^2(Z)$, $\sum_i \omega_i^2(X) = \sum_i \omega_i^2(R)$, $\sum_i \omega_i^2(M) = \sum_i \omega_i^2(A)$ and $2\sum_i \omega_i^2(X) = \sum_i \omega_i^2(\Gamma) + \sum_i \omega_i^2(M)$, as shown in Table II in the maintext.

As shown above, by entering the information of the typical magnetic model, one can use this code to obtain the relations between magnon eigenvalues in this paper easily. Meanwhile, one can easily extend our method to other magnetic systems by simply modifying this example code.