Determining the range of magnetic interactions from the relations between magnon eigenvalues at high-symmetry $k$ points

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(Dated: December 15, 2021)

Magnetic exchange interactions (MEIs) define networks of coupled magnetic moments and lead to a surprisingly rich variety of their magnetic properties. Typically MEIs can be estimated by fitting experimental results. But how many MEIs need to be included in the fitting process for a material is not clear a priori, which limits the quality of results obtained by these conventional methods. In this paper, based on linear spin-wave theory but without performing matrix diagonalization, we show that for a general quadratic spin Hamiltonian, there is a simple relation between the Fourier transform of MEIs and the sum of square of magnon energies (SSME). We further show that according to the real-space distance range within which MEIs are considered relevant, one can obtain the corresponding relationships between SSME in momentum space. We also develop a theoretical tool for tabulating the rule about SSME. By directly utilizing these characteristics and the experimental magnon energies at only a few high-symmetry $k$ points in the Brillouin zone, one can obtain strong constraints about the range of exchange path beyond which MEIs can be safely neglected. Our methodology is also general applicable for other Hamiltonian with quadratic Fermi or Boson operators.

I. INTRODUCTION

As one of the oldest scientific topics, magnetism is still of great interest [1–4]. Magnetic materials had already been widely used in electromechanical and electronic devices, and its applications in information technology are also continuously growing [1–3]. Especially magnons, as the quanta of spin waves, have received more and more research attention over the past few decades [5, 6]. As an elementary excitations of magnetic systems, magnons became an interesting platform for the study of general wave dynamics [7, 8], Bose-Einstein condensation of magnon [9–11] and so on. In addition, with the development of topological physics in the electron system, topology in magnon spectrum has also attracted significant interests [12–14], including topological magnon insulators [15, 16], magnonic Dirac semimetals [17–21] and Weyl semimetals [22–24]. Besides fundamental research, magnons have also attracted great attention for applications of information transport and processing [25–29]. Analogous to spintronics, the application of magnon are connected with the ability to carry, transport and process information. Potentially, the spins can be manipulated without current, thereby overcoming an important fundamental limitation of conventional electronic devices, the dissipation of energy due to Ohmic losses. Magnon spintronics is therefore an emerging field of modern magnetism, which has spurred significant advances towards computing application recently and is believed to deliver a number of breakthrough developments in the future [25–29]. In order to quantitatively understand the rich phenomenon and wide applications in this highly interdisciplinary field, a microscopic magnetic model with proper parameters becomes extremely important.

Magnetic properties can be typically described by a quadratic spin Hamiltonian \( H = \sum_{i,j} J_{ij} S_i \cdot S_j = \sum_{i,j} J_{ij} S_i^a S_j^a \), where \( J_{ij} \) represents the magnetic exchange interaction (MEI) between the spin at \( i \) site \( S_i \) and spin at \( j \) site \( S_j \) as shown in the following Eq. (1). The sum should take over all possible exchange paths with sizable MEIs. However, it turns out that extracting quantitative \( J_{ij} \) is a highly non-trivial task. By choosing the set of parameters that best fit the experimental results, such as temperature-dependent magnetization, magnetic susceptibility \( \chi(T) \), magnetic excitation spectra \( \omega(q) \) etc, one basically can obtain MEIs \( J \)'s [1–4]. It is well known that the \( J_{ij} \) usually decreases rapidly with increasing of \( R_{ij} \), the distance between magnetic moment at \( i \) and \( j \) sites, and the \( J \)'s with sufficient distance are believed to be negligible. Thus only a number of \( J \)'s within a cut-off range \( R_{cut} \) are needed to be considered. However, a priori knowledge about \( R_{cut} \) is unknown, while the number of MEI used to fit the experimental data obviously affect the obtained \( J \)'s. This leads to the arbitrariness of fitting approach, consequently affected the accuracy of the estimated MEIs, and currently unambiguous fitting is basically impossible. For example, very similar inelastic neutron scattering (INS) experimental results can be fitted by considerably different MEI parameters [20, 21].

In addition to the above approach, theoretical calculations had also been used to evaluate the exchange interaction parameters [30–38]. A popular numerical method is to calculate the total energies of more than \( N \) magnetic

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configurations and map them using a spin Hamiltonian to extract \( N \) MEIs \([30]\). Unfortunately, this method also need to assume a cut-off range \( R_{\text{cut}} \), which again leads to the arbitrariness about the calculated MEIs. An alternative method is based on combining magnetic force theorem and linear-response approach \([31-35]\). Working in the momentum space, this method indeed does not suffer the problem about \( R_{\text{cut}} \). However, the Coulomb interaction which had been incorporated by the parameter \( U \) in first-principles calculations, usually play important role in magnetic systems \([39, 40]\). Thus these theoretical MEIs strongly depend on the choice of the parameter \( U \) \([39, 40]\).

Symmetry imposes constraints about the magnetic model, and one can also use symmetry to check if two exchange paths with the same bond length have the same MEI. Unfortunately, this powerful theoretical method cannot provide any clue about \( R_{\text{cut}} \). The general features, such as sum rule for the spectral weight of the spin correlation function \([41]\) which requires accurate cross-section measurements over the entire Brillouin zone (BZ), also cannot predict the variation of MEIs over distance. Thus to explore possible \( R_{\text{cut}} \)-related generic rules is a very important problem. Certain important subjects on magnetism, such as quantum spin liquids arising from geometrical frustration models \([42]\), novel properties from geometrically frustrated magnet \([43]\), etc., explicitly require small \( R_{\text{cut}} \), hence gaining a wealth of knowledge for \( R_{\text{cut}} \) in a large set of known magnetic materials will also be empirically useful for assessing the relevance of such models.

In this work, based on linear spin-wave theory (LSWT), we find that for a general quadratic spin Hamiltonian, the sum of square of magnon energies (SSME) at \( (\text{LSWT}) \), we find that for a general quadratic spin Hamiltonian with the scalar term \( J_{R_l+\tau_n,R_{l'}+\tau_{n'}} S_{l,n} S_{l',n'}^{\beta} \) \([1-6]\), the experimental SSME start to deviate from the obtained theoretical relationships, one can determine the real-space range of sizable MEIs. Our method can be easily extended to other Hamiltonian with quadratic Fermi or boson operators, thus is useful for the characteristics of the electronic band structure, phonon spectrum, etc.

\[ H = \sum_{l,n,\alpha,l',n',\beta} J_{R_l+\tau_n,R_{l'}+\tau_{n'}}^{\alpha,\beta} S_{l,n} S_{l',n'}^{\beta} \]  

where \( J_{R_l+\tau_n,R_{l'}+\tau_{n'}}^{\alpha,\beta} \) a \( 3 \times 3 \) tensor, represents the spin exchange parameter. Here \( R_l \) and \( \tau_n \) represent the lattice translation vector and the position of magnetic ions in the lattice basis, while \( \alpha \) and \( \beta \) denote \( x, y \) or \( z \) the cartesian components. As a \( 3 \times 3 \) real tensor, \( J_{R_l+\tau_n,R_{l'}+\tau_{n'}} \) could be expanded as three terms, and Eq. (1) could be written as

\[ H = \sum_{l,n,l',n'} J_{R_l+\tau_n,R_{l'}+\tau_{n'}} S_{l,n} S_{l',n'} + \sum_{l,n,l',n'} D_{R_l+\tau_n,R_{l'}+\tau_{n'}} [S_{l,n} \times S_{l',n'}] + \sum_{l,n,l',n'} S_{l,n} \cdot \Gamma_{R_l+\tau_n,R_{l'}+\tau_{n'}} S_{l',n'} \]  

Here the first term describes the isotropic Heisenberg Hamiltonian with the scalar term \( J_{R_l+\tau_n,R_{l'}+\tau_{n'}} \), the second one represents the antisymmetric Dzyaloshinskii-Moriya (DM) interactions with the vector term \( D_{R_l+\tau_n,R_{l'}+\tau_{n'}} \) \([44, 45]\), and the third one is the rest of anisotropic terms with the symmetric tensor term \( \Gamma_{R_l+\tau_n,R_{l'}+\tau_{n'}} \) \([45]\). It is commonly believed that the magnitude of the DM interaction and \( \Gamma_{R_l+\tau_n,R_{l'}+\tau_{n'}} \) are proportional to spin-orbit coupling (SOC) strength \( \lambda \) and \( \lambda^2 \), respectively \([45]\). For the materials with large \( \lambda \), such as ferromagnetic systems, multipolar interactions may become important \([46]\), thus we restrict us on the cases with small \( \lambda \) and ignore the third term in Eq. (2) \(^3\). To take account for non-collinear cases, we use the polar angle \( \theta_n \) and azimuthal angle \( \phi_n \) for the spin orientation of magnetic ion at \( n \) site.

\(^3\) Eq. (1) may also not be suitable for the case with orbitally degenerate \([47]\).
Following the LSWT [5], we perform the Holstein-Primakoff transformation and the Fourier transformation, and the Eq. (1) could be written as

\[ \sum_{k} \psi^\dagger(k) H(k) \psi(k) \]  

where \( \psi^\dagger(k) = [a^\dagger_1(k), ..., a^\dagger_N(k), a_1(-k), ..., a_N(-k)] \), in which \( a^\dagger_i(k) \) and \( a_i(k) \) represent the canonical boson creation and annihilation operators with wave vector \( k \). Here \( i \) runs from 1 to \( N \), and \( N \) is the number magnetic ions per unit cells. The Hermitian matrix \( H(k) \) in Eq. (3) is expressed as

\[ H(k) = \begin{bmatrix} h(k) & h'(k) \\ h'(k) & h(-k) \end{bmatrix} \]  

Here \( h(k) \) and \( h'(k) \) are expressed by

\[
h(k)_{n,n'} = \sum_l S(A_{n,n'}J_{\tau_n,\tau_{n'}+R_l} + B_{n,n'}D_{\tau_n,\tau_{n'}+R_l})
\]

\[
e^{i\mathbf{k} \cdot \mathbf{R}_l} - \delta_{n,n'}\sum_{l,n''}(S(B_{n,n''}J_{\tau_n,\tau_{n''}+R_l} + P_{n,n''}D_{\tau_n,\tau_{n''}+R_l})
\]

\[
h'(k)_{n,n'} = \sum_l S(C_{n,n'}J_{\tau_n,\tau_{n'}+R_l} + Q_{n,n'}D_{\tau_n,\tau_{n'}+R_l})
\]

\[
e^{i\mathbf{k} \cdot \mathbf{R}_l} \]  

where \( \delta_{n,n'} \) is the Kronecker delta function, while \( A_{n,n'}, B_{n,n'}, C_{n,n'}, O_{n,n'}, P_{n,n'} \) and \( Q_{n,n'} \) are parameters related to the spin directions at \( n \) and \( n' \) sites (see SM for details).

Considering the commutation relation of \( \psi(k) \) and \( \psi^\dagger(k) \), we need perform the following transformation (see SM for details):

\[ H_J(k) = I \cdot H(k) \]  

Through numerically diagonalizing the \( H_J(k) \) in Eq. (6), we can obtain the magnon energies \( \omega_i(k)(i = 1, ..., N) \) at wave vector \( k \). In contrary without diagonalization, SSME can be analytically expressed as:

\[ \sum_i \omega_i^2(k) = \frac{1}{2} Tr([H_J(k)]^2) \]

\[ = \frac{1}{2} Tr[|h(k) + h^\dagger(-k)|^2 - h'(k)h'(k)^\dagger - h'(k)^\dagger h'(k)] \]  

As shown in Eq. (5), \( h(k) \) and \( h'(k) \) basically depend on the orientation of magnetic moments and the Fourier transformation of MEIs \( J_{\mathbf{R}_l+\tau_n, \mathbf{R}_l'+\tau_{n'}} \) and \( D_{\mathbf{R}_l, \tau_n, \mathbf{R}_l'+\tau_{n'}} \). Thus, for arbitrary \( k \), \( \sum \omega_i^2(k) \) can be expressed by a quadratic polynomial of MEIs. With the assumption of \( R_{cut} \), which related with how many MEIs had been considered, one can obtain simple relationships between SSME at different wave vectors \( k \).

### III. RESULTS AND DISCUSSION

| WP  | \( \tau_n \) | \((\theta_n, \phi_n)\) |
|-----|-----------|----------|
| 4d  | 1 (0.0, 0) | (0.0) (\(\theta, \pi/2\)) |
| 2   | 2 (0.5, 0, 0) | (0.0) (\(\theta, -\pi/2\)) |
| 3   | 3 (0, 0.5, 0) | (0.0) (\(\theta, \pi/2\)) |
| 4   | 4 (0.5, 0.5) | (0.0) (\(\theta, -\pi/2\)) |
| 2a  | 5 (0.75, 0.25, 0) | (0.0) (0.0) |
| 6   | 6 (0.25, 0.75, 0) | (0.0) (0.0) |
| 2c  | 7 (0.25, 0.25, 0.1) | (0.0) (0.0) |
| 8   | 8 (0.75, 0.75, -0.1) | (0.0) (0.0) |

We illustrate the usage of our results by following typical example. Without loss of generality, we choose space group P4/\(n\) (SG 85) to present our discussion and set the ratio between lattice constant \( c/a \) as 0.8. We put the magnetic ions at three nonequivalent crystallographic sites: 4d (0, 0, 0), 2a (0.25, 0.75, 0) and 2c (0.25, 0.25, z) Wyckoff positions (WPs), as summarized in Table I. While the 4d and 2a WPs had been completely determined by the spatial symmetry, the coordinates of 2c WP have a variable \( z \) and here we adopt it as \( z = 0.1 \). There are two generators for this space group: the four-fold rotation \( \{4z_{\text{inv}}\}1/2, 0, 0 \) and inversion operation \( \{7(0, 0, 0) \} \), where the left part represents the rotation, the right part means the lattice translation, and \( \bar{T} \) denotes the inversion symmetry. We firstly consider the most simple case: isotropic Heisenberg model with all the spins along \( z \). Considering the orientations of the magnetic moments, the space group could be divided into four types of magnetic space groups. The case with this collinear ferromagnetic (FM) ordering belongs to the type-I magnetic
TABLE II. The bonds for the 1st, 2nd and 3rd NNs of the example shown in here and the corresponding Heisenberg exchange interactions of the collinear FM configuration (i.e. the case with BNS 85.59). Each bond is characterized by the positions of the two endings: $\tau_n, \tau_{n' + R_1}$. The unit of distance is taken as the lattice constant $a$.

| distance(a) | n | n' | $R_1$ |
|------------|---|----|-------|
| $J_1$      | 0.35 | 1 | 5 | ($-1, 0, 0$) |
| 1          | 6 | 0, $-1, 0$ |
| 2          | 5 | 0, 0, 0 |
| 3          | 6 | 0, $-1, 0$ |
| 4          | 6 | 0, 0, 0 |
| $J_2$      | 0.36 | 1 | 7 | (0, 0, 0) |
| 1          | 8 | ($-1, 0, 0$) |
| 1          | 7 | (0, 0, 0) |
| $J_3$      | 0.5 | 1 | 2 | (0, 0, 0) |
| 1          | 2 | ($-1, 0, 0$) |
| 1          | 3 | (0, 0, 0) |
| 1          | 4 | (0, $-1, 0$) |
| 1          | 4 | (0, 0, 0) |

As shown in the above formula, the key for SSME is the exchange path between magnetic ions $\tau_n$ and $\tau_{n'} + R_1$, and the related MEI $J_{\tau_n, \tau_{n'} + R_1}$. As shown in Table II, the first and second nearest neighbor exchange paths have similar distances (0.35 $a$ vs 0.36 $a$, $a$ is lattice parameter). Crystal symmetry imposes strong restrictions on the MEIs as shown in SM, and according to the spatial symmetry in this space group, all the first nearest neighbor exchange paths have the same MEI value, and we denote it as $J_1$, same as that, we can label all the second nearest neighbor MEI as $J_2$. Considering only the first two NN interactions $J_1$ and $J_2$, the term of $J_{\tau_n, \tau_{n'} + R_1}$ does not exist as shown in Table II, and the only $k$ dependence of SSME comes from the first term in Eq. (8). Namely, we need to check the non-zero MEIs $J_{\tau_n, \tau_{n'} + R_1}$ and $J_{\tau_n, \tau_{n'} + R_1}$, with the requirement of $R_1 \neq R_1$. As clearly shown in Table II, such kind of exchange path is also not exist.

Thus, although spin wave has dispersion at the entire BZ, we get a surprisingly simple result of $\sum_i \omega_i^2(k) = C$ with considering only $J_1$ and $J_2$.

We further take into account the impact of longer-range exchange paths. With the third NN MEI $J_3$ been considered, there exist more than one exchange paths connect a pair of $\tau_n$ and $\tau_{n'} + R_1$. For example, both $\tau_1, \tau_2$ pair and $\tau_1, \tau_2 + R_{100}$ pair belong to $J_3$ exchange path as shown in Table II. As a result, $\sum_i \omega_i^2(k)$ is no longer equal to constant. Thus, if the observed SSME shows very weak $k$ dependence, one can asserts that the MEIs beyond $J_2$ are ignorable. Since for the high symmetry $k$ points at BZ, $e^{i k \cdot (R_1 - R_1')}$ usually has simple values (equals to $\pm 1$ in this magnetic system), one can expect simple relation between SSME at these $k$ points. We indeed get several simple relationships with the MEIs up to $J_3$: $\sum_i \omega_i^2(T) = \sum_i \omega_i^2(Z)$,
TABLE III. The obtained SSME relationship of the collinear FM example shown here (i.e. the case with BNS 85.59).
The first column $J_n$ represents up to the $x$-th NN MEI. The coordinate of six high-symmetry $k$ points: $\Gamma(0, 0, 0)$, $X(\frac{1}{2}, 0, 0)$, $Z(0, 0, \frac{1}{2})$, $R(\frac{1}{2}, \frac{1}{2}, 0)$, and $A(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$.

| $J_{\text{max}}$ relation | $J_2$ | $\sum_i \omega_i^2(k) = C$ |
|---------------------------|-------|----------------------|
|                           | $J_4$ | $\sum_i \omega_i^2(\Gamma) = \sum_i \omega_i^2(Z)$ |
|                           |       | $\sum_i \omega_i^2(X) = \sum_i \omega_i^2(R)$ |
|                           |       | $\sum_i \omega_i^2(M) = \sum_i \omega_i^2(A)$ |
|                           |       | $2 \sum_i \omega_i^2(X) = \sum_i \omega_i^2(\Gamma) + \sum_i \omega_i^2(M)$ |
|                           | $J_{12}$ | $\sum_i \omega_i^2(\Gamma) = \sum_i \omega_i^2(Z)$ |
|                           |       | $\sum_i \omega_i^2(X) = \sum_i \omega_i^2(R)$ |
|                           |       | $\sum_i \omega_i^2(M) = \sum_i \omega_i^2(A)$ |
|                           | $J_{22}$ | $\sum_i \omega_i^2(I) - \sum_i \omega_i^2(Z) = \sum_i \omega_i^2(X) - \sum_i \omega_i^2(R) = \sum_i \omega_i^2(M) - \sum_i \omega_i^2(A)$ |

The sum of exchange paths in collinear spin ordering case become inequivalent. For example, as shown in Table V of SM, the eight first NN exchange paths in collinear spin ordering case are no longer equivalent and divided into two groups, which are labeled as $J_1$ and $J_2$ for this non-collinear magnetic case. The parameters in Eq. (5) are also depended on the magnetic moment directions, thus non-collinearity results in different relationship between SSME, which are listed in Table IV. We also want to mention that for the localized magnetic systems, the MEIs should not be sensitive to the magnetic configurations as also required by energy-mapping method for calculating MEIs. For such cases, one can still use the symmetry operations in collinear instead of in non-collinear case to determine equivalent exchange path. Namely if this non-collinear magnetism is very localized, the MEIs will still approximately satisfy the Table II. Based on Table II (i.e. the right part of the Table V in SM) instead of the left part of the Table V in SM, we applying our method to this non-collinear case with localized magnetism and list the results in the right part of Table IV. As shown in Table IV, the free parameter $\theta$ about the magnetic moments directions explicitly appear in the relationship about SSME. Thus, for localized non-collinear magnetic materials, one may determine the magnetic moments directions based on the magnon energies at three wave vectors (i.e. $\Gamma$, $X$ and $M$) in the case that MEIs further than $J_3$ are ignorable.

It is worth to mention that our method is also valid for the materials with considerable DM interactions [44, 45]. One can still calculate SSME by Eq. (7) and directly use the program provided in SM to explore the relationship between them. The magnetic anisotropy may also comes from the SIA [1–3]. With the SIA considered, the Hamiltonian becomes $H_{\text{total}} = H + H_{\text{SIA}}$, here $H$ is the term shown in Eq. (2) while $H_{\text{SIA}}$ represents the term of SIA. Here we adopt a popular form $H_{\text{SIA}} = \sum_{\tau} K(S_\tau^2)^2$ [3] where $K$ is the strength of SIA. Based on the standard LSWT, one can easily obtain the spin Hamiltonian at arbitrary wave vector $k$ to be $H_{\text{total}}(k) = H_{\text{J}}(k) + 2SKL_n$. Adding SIA term into the case of Heisenberg model with collinear FM magnetic ordering given in this work, the SSME could be written as

$$\sum_i \omega_i^2(k) = \frac{1}{2} Tr[(H_{\text{total}}(k))^2]$$

$$= \frac{1}{2} Tr[(H_{\text{J}}(k))^2] + 4NS^2K^2 + 4S^2K \cdot$$

$$\sum_n \left[ \sum_{\tau_\alpha, \tau_\beta} J_{\tau_\alpha, \tau_\beta} + R_i + \sum_{\tau_\alpha} J_{\tau_\alpha, \tau_\beta} e^{ikR_i} \right]$$

(9)

Based on Eq. (9), one can easily prove that including SIA will not affect the results given in Table III. For the other cases, one can simply use our code which has implemented effect of SIA to obtain the results.
TABLE IV. The obtained results about SSME for the non-collinear case shown in this work (i.e. the case with symmetry of BNS 13.65). We also give the result for the same non-collinear configuration with the magnetism is very localized in the right part. For the localized magnetism case, the MEIs are not sensitive to the spin ordering, thus one can use the symmetry of SG 85 to determine if the exchange paths have the same MEIs, namely use the results given in the right part of Table V in SM. The x in \( J_x \) still represents up to the x-th NN MEI, and the coordination of six high-symmetry points had been shown in Table III.

| \( J_{\text{max}} \) relation | \( J_{\text{max}} \) relation |
|-----------------------------|-----------------------------|
| \( J_4 \sum_i \omega^2_i(k) = C \) | \( J_2 \sum_i \omega^2_i(k) = C \) |
| \( J_5 \sum_i \omega^2_i(\Gamma) = \sum_i \omega^2_i(Z) \) | \( J_3 \sum_i \omega^2_i(\Gamma) = \sum_i \omega^2_i(Z) \) |
| \( \sum_i \omega^2_i(X) = \sum_i \omega^2_i(R) \) | \( \sum_i \omega^2_i(X) = \sum_i \omega^2_i(R) \) |
| \( \sum_i \omega^2_i(M) = \sum_i \omega^2_i(A) \) | \( \sum_i \omega^2_i(M) = \sum_i \omega^2_i(A) \) |
| \( 1 + \cos 2\theta \sum_i \omega^2_i(X) = \omega^2_i(\Gamma) + \cos 2\theta \sum_i \omega^2_i(M) \) | \( (1 + \cos 2\theta) \sum_i \omega^2_i(X) = \omega^2_i(\Gamma) + \cos 2\theta \sum_i \omega^2_i(M) \) |

IV. CONCLUSION

In summary, appropriate magnetic model play crucial role in investigating various magnetic properties. Unfortunately the current methods for extracting MEIs face a severe limitation about how many MEIs need to be included in the spin Hamiltonian. In this work, we circumvent this methodological bottleneck by noticing that for quadratic spin Hamiltonian, there is a simple connection between SSME and the considered MEIs. Namely, there is \( R_{\text{cut}} \)-related rules between SSME at high-symmetry points. By efficient measurements of magnon energies only at several high-symmetry \( k \) points, one can check up to which \( R_{\text{cut}} \), the experimental SSME start to deviate from the obtained \( R_{\text{cut}} \)-related rules, and subsequently determine the real-space range beyond which MEIs can be safely neglected. For the localized non-collinear magnetic systems, our results may also be used to determine the directions of magnetization. We also provide a program, directly utilizing it, one can get the relationship of SSME for any crystal magnetic materials described by Hamiltonian with pairwise spin. Besides the well used symmetry analysis for the symmetry-related \( k \) points, we expect that similar generic \( R_{\text{cut}} \)-sensitive rules also exist in other Hamiltonian with only quadratic Fermi or boson operators.

VI. SUPPLEMENTAL MATERIALS

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

\[
H = \sum_{l,n,l',n'} S_{ln} \cdot J_{R_l + \tau_n, R_{l'} + \tau_{n'}} \cdot S_{l'n'}
\]

where \( J_{R_l + \tau_n, R_{l'} + \tau_{n'}} \), a \( 3 \times 3 \) tensor, represents the spin exchange parameters. \( R_l \) and \( \tau_n \) represent the lattice translation vector and the position of magnetic ions in the lattice basis, and \( S_{l'n'} \) means the spin at the site of \( R_{l'} + \tau_{n'} \). Translation symmetry will restrict \( J_{R_l + \tau_n, R_{l'} + \tau_{n'}} \) to be only related to \( J_{R_l + \tau_n, R_{l'} + R_{l'}} \) 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 \( \{ \alpha | \tau \} \), where the left part represents the rotation and the right part means the lattice translation. Supposing under this symmetry operator, \( R_m + \tau_p \) and \( R_{m'} + \tau_{p'} \) transfer to \( R_l + \tau_n \) and \( R_{l'} + \tau_{n'} \), respectively, meanwhile the transformation of spin becomes \( S_{n'p'} = M(\alpha)S_{ln} \), where \( M(\alpha) \) is the representation matrix of the proper rotation part of the operation \( \alpha \) in the coordinate system, we get the following expression:

\[
H = \sum_{l,n,l',n'} S_{ln} \cdot J_{R_l + \tau_n, R_{l'} + R_{l'}} \cdot S_{l'n'}
\]

V. ACKNOWLEDGEMENTS

This work was supported by the NSFC (No. 11834006, 12004170, 51721001, and 11790311), National Key R&D Program of China (No. 2018YFA0305704 and 2017YFA0303203) and the excellent programme in Nanjing University. Xiangang Wan also acknowledges the support from the Tencent Foundation through the XPLORER PRIZE.
\[ H = \sum_{l,n,l',n'} S_{ln} \cdot J_{R_l+\tau_n, R_{l'}+\tau_{n'}} \cdot S_{l'n'} \]
\[ = \sum_{l,n,l',n'} S_{ln} M^\dagger(\alpha) M(\alpha) J_{R_l+\tau_n, R_{l'}+\tau_{n'}} M^\dagger(\alpha) M(\alpha) S_{l'n'} \]
\[ = \sum_{m,p,m',p'} S_{mp} \cdot M(\alpha) J_{R_l+\tau_n, R_{l'}+\tau_{n'}} M^\dagger(\alpha) \cdot S_{m'p'} \] (11)

Then the exchange interactions should satisfy the following condition:
\[ J_{R_m+\tau_p, R_{m'}+\tau_{p'}} = M(\alpha) J_{R_l+\tau_n, R_{l'}+\tau_{n'}} M^\dagger(\alpha) \] (12)

After decomposing the 3x3 tensor J into scalar Heisenberg term J and vector DM term D as in the maintext, we obtain the following results:

\[ J_{R_m+\tau_p, R_{m'}+\tau_{p'}} = J_{R_l+\tau_n, R_{l'}+\tau_{n'}} \]
\[ D_{R_m+\tau_p, R_{m'}+\tau_{p'}} = M(\alpha) D_{R_l+\tau_n, R_{l'}+\tau_{n'}} \] (13)

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

\[ J_{R_l+\tau_n, R_{l'}+\tau_{n'}} = J_{R_l+\tau_n, R_{l'}+\tau_{n'}} \]
\[ D_{R_l+\tau_n, R_{l'}+\tau_{n'}} = -D_{R_l+\tau_n, R_{l'}+\tau_{n'}} \] (14)

According to the above equations (i.e. Eq. (13) and (14)), 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_{444}1/2, 0, 0\} and inversion operation \(\{0, 0, 0\}\). The magnetic ion located at \(\tau_1\) position has only two nearest neighbors, i.e. \(\{\tau_1, \tau_2 + R_{-100}\}\) pair and \(\{\tau_1, \tau_6 + R_{0,0,10}\}\) pair as shown in Table V (also see the Table II in the maintext). These two bonds are equivalent by the inversion symmetry \(\{0, 0, 0\}\). 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. (13), 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 V. The non-collinearity shown in maintext reduces the four-fold rotation \(\{4_{444}1/2, 0, 0\}\) to the two-fold rotation operation \(\{2_{000}1/2, 1/2, 0\}\), as a result the eight NN exchange path is no longer equivalent as indicated in Table V. Similarly, one can obtain the symmetry restriction on the DM interactions, as also shown in Table V.

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

It is worth mentioning that Eq. (12) can also give symmetry restrictions on SIA. The general quadratic expression of SIA could be written as \(\sum_{l,n,a,b} K^a_{R_l+\tau_n} S^a_{l} S^b_{l}\). Note that the SIA term should be naturally symmetric, i.e. \(K^a_{R_l+\tau_n} = K^a_{R_{l}+\tau_{n}}\). Accordingly to Eq. (12), when the action of symmetry operation \(\{\alpha||\\}\) keeps the position \(R_l + \tau_n\) unchanged, we have \(K_{R_l+\tau_n} = M(\alpha) M_{R_l+\tau_n} M^\dagger(\alpha)\). 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

\[ K_{R_l+\tau_n} = \begin{bmatrix} K^{zz}_{R_l+\tau_n} & K^{\vec{x}}_{R_l+\tau_n} \\ K^{\vec{x}}_{R_l+\tau_n} & K^{zz}_{R_l+\tau_n} \end{bmatrix} \]

which is actually the usual form \(H_{SIA} = \sum_{l,n} K_{R_l+\tau_n} (S^z_{l})^2 + C\) where \(K_{R_l+\tau_n} = K^{zz}_{R_l+\tau_n} = K^{zz}_{R_l+\tau_n}\) 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 \(H_{SIA} = \sum_{l,n} K (S^z_{l})^2\) [3] for simplicity.

B. The parameters \(A_{n,n'}, B_{n,n'}, C_{n,n'}, O_{n,n'}, P_{n,n'}\) and \(Q_{n,n'}\)

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, where the parameters \(A_{n,n'}, B_{n,n'}, C_{n,n'}, O_{n,n'}, P_{n,n'}\) and \(Q_{n,n'}\) 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:
TABLE V. The distances and the bond information of corresponding MEIs (with \(R_{\text{cut}} = 0.5 \ a\)) for the non-collinear magnetic example in the main text (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) | \(n\ n'\) | \(R_1\) | BNS 13.65 | BNS 85.59 |
|-------------|-----------|--------|-----------|-----------|
| 0.35        | 1 5       | \((-1,0,0)\) | \(J_1\) | \((D^x_1, D^y_1, D^z_1)\) |
|             | 1 6       | \((0,-1,0)\) | \(J_1\) | \((D^x_1, D^y_1, D^z_1)\) |
|             | 2 5       | \((0,0,0)\)  | \(J_2\) | \((-D^y_2, D^x_2, D^z_2)\) |
|             | 2 6       | \((0,-1,0)\) | \(J_2\) | \((-D^y_2, D^x_2, D^z_2)\) |
|             | 3 5       | \((-1,0,0)\) | \(J_2\) | \((-D^y_2, D^x_2, D^z_2)\) |
|             | 3 6       | \((0,0,0)\)  | \(J_2\) | \((-D^y_2, D^x_2, D^z_2)\) |
|             | 4 5       | \((0,0,0)\)  | \(J_1\) | \((-D^1_1, -D^1_1, D^1_1)\) |
|             | 4 6       | \((0,0,0)\)  | \(J_1\) | \((-D^1_1, -D^1_1, D^1_1)\) |

| 0.36        | 1 7       | \((0,0,0)\)  | \(J_3\) | \((D^x_3, D^y_3, D^z_3)\) |
|             | 1 8       | \((-1,-1,0)\)| \(J_3\) | \((D^x_3, D^y_3, D^z_3)\) |
|             | 2 7       | \((0,0,0)\)  | \(J_4\) | \((-D^y_4, D^x_4, D^z_4)\) |
|             | 2 8       | \((0,-1,0)\) | \(J_4\) | \((-D^y_4, D^x_4, D^z_4)\) |
|             | 3 7       | \((-1,0,0)\) | \(J_4\) | \((-D^y_4, D^x_4, D^z_4)\) |
|             | 3 8       | \((-1,-1,0)\)| \(J_4\) | \((-D^y_4, D^x_4, D^z_4)\) |
|             | 4 7       | \((0,0,0)\)  | \(J_5\) | \((-D^y_5, D^x_5, D^z_5)\) |
|             | 4 8       | \((0,0,0)\)  | \(J_5\) | \((-D^y_5, D^x_5, D^z_5)\) |

| 0.5         | 1 2       | \((0,0,0)\)  | \(J_6\) | \((D^x_6, D^y_6, D^z_6)\) |
|             | 1 3       | \((-1,0,0)\) | \(J_6\) | \((D^x_6, D^y_6, D^z_6)\) |
|             | 1 1       | \((0,0,0)\)  | \(J_6\) | \((-D^y_6, D^x_6, D^z_6)\) |
|             | 1 1       | \((0,-1,0)\) | \(J_6\) | \((-D^y_6, D^x_6, D^z_6)\) |
|             | 2 4       | \((0,0,0)\)  | \(J_6\) | \((-D^y_6, D^x_6, D^z_6)\) |
|             | 2 4       | \((-1,0,-1)\)| \(J_6\) | \((-D^y_6, D^x_6, D^z_6)\) |
|             | 3 4       | \((0,0,0)\)  | \(J_6\) | \((-D^y_6, D^x_6, D^z_6)\) |
|             | 3 4       | \((-1,-1,0)\)| \(J_6\) | \((-D^y_6, D^x_6, D^z_6)\) |

TABLE VI. 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 main text.

| BNS 85.59 | BNS 13.65 |
|-----------|-----------|
| \(J_1\)   | \(J_1, J_2\) |
| \(J_2\)   | \(J_3, J_4\) |
| \(J_3\)   | \(J_5, J_6\) |
| \(J_4\)   | \(J_7, J_8\) |
| \(J_5\)   | \(J_9, J_{10}\) |
| \(J_7\)   | \(J_{12}, J_{14}\) |
| \(J_9\)   | \(J_{15}, J_{16}\) |
| \(J_{11}, J_{12}\) | \(J_{17}, J_{18}\) |
| \(J_{13}, J_{15}\) | \(J_{22}, J_{30}\) |
| \(J_{17}, J_{18}\) | \(J_{22}, J_{30}\) |
| \(J_{19}\) | \(J_{35}, J_{36}\) |
| \(J_{20}, J_{21}\) | \(J_{37}, J_{40}\) |
| \(J_{22}\) | \(J_{31}, J_{42}\) |
| \(J_{23}\) | \(J_{33}, J_{44}\) |
| \(J_{25}, J_{27}\) | \(J_{35}, J_{37}\) |

\[A_{n,n'} = \frac{1}{4} \cos(\theta_n - \theta_{n'}) - \frac{1}{4} \cos(\theta_n + \theta_{n'}) + \frac{1}{2} \cos(\phi_n - \phi_{n'}) + \frac{1}{8} \cos(\theta_n - \theta_{n'} + \phi_n - \phi_{n'})\]

\[B_{n,n'} = \sin(\theta_n) \sin(\phi_n - \phi_{n'}) + \cos(\theta_n) \cos(\phi_n - \phi_{n'})\]

\[C_{n,n'} = \frac{1}{4} \cos(\theta_n - \theta_{n'}) - \frac{1}{4} \cos(\theta_n + \theta_{n'}) - \frac{1}{2} \cos(\phi_n - \phi_{n'}) + \frac{1}{8} \cos(\theta_n - \theta_{n'} + \phi_n - \phi_{n'})\]
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:

\[
O_{x,n,n'} = \frac{1}{2} [ \cos \theta_n \sin \theta_{n'} \sin \phi_n + \sin \theta_n \cos \theta_{n'} \sin \phi_{n'} + i(\sin \theta_n \cos \phi_n + \sin \theta_{n'} \cos \phi_{n'}) ]
\]

(18) 

\[
P_{x,n,n'} = \sin \theta_n \cos \theta_{n'} \sin \phi_n - \cos \theta_n \sin \theta_{n'} \sin \phi_{n'}
\]

(19) 

\[
Q_{x,n,n'} = \frac{1}{2} [ \cos \theta_n \sin \theta_{n'} \cos \phi_n + \sin \theta_n \cos \theta_{n'} \cos \phi_{n'} + i(\sin \theta_n \cos \phi_n - \sin \theta_{n'} \cos \phi_{n'}) ]
\]

(20) 

\[
O^y_{n,n'} = \frac{1}{2} [ \cos \theta_n \sin \theta_{n'} \cos \phi_n + \sin \theta_n \cos \theta_{n'} \cos \phi_{n'} + i(\sin \theta_n \sin \phi_n + \sin \theta_{n'} \sin \phi_{n'}) ]
\]

(21) 

\[
P^y_{n,n'} = \sin \theta_n \cos \theta_{n'} \cos \phi_n - \cos \theta_n \sin \theta_{n'} \sin \phi_{n'}
\]

(22) 

\[
Q^y_{n,n'} = \frac{1}{2} [ \cos \theta_n \sin \theta_{n'} \cos \phi_n + \sin \theta_n \cos \theta_{n'} \cos \phi_{n'} + i(\sin \theta_n \sin \phi_n - \sin \theta_{n'} \sin \phi_{n'}) ]
\]

(23) 

\[
O^z_{n,n'} = \frac{1}{2} [(1 + \cos \theta_n \cos \theta_{n'}) \sin(\phi_n - \phi_{n'}) - i(\cos \theta_n + \cos \theta_{n'}) \cos(\phi_n - \phi_{n'})]
\]

(24) 

\[
P^z_{n,n'} = \sin \theta_n \sin \theta_{n'} \sin(\phi_n - \phi_{n'})
\]

(25) 

\[
Q^z_{n,n'} = \frac{1}{2} [(-1 + \cos \theta_n \cos \theta_{n'}) \sin(\phi_n - \phi_{n'}) - i(\cos \theta_n - \cos \theta_{n'}) \cos(\phi_n - \phi_{n'})]
\]

(26) 

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:

\[
A_{n,n'} = \frac{\zeta_{n,n'} + 1}{2}
\]

(27) 

\[
B_{n,n'} = \zeta_{n,n'}
\]

(28) 

\[
C_{n,n'} = \frac{\zeta_{n,n'} - 1}{2}
\]

(29) 

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 [i.e., 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

\[
A_{n,n'} = 1
\]

(30) 

\[
B_{n,n'} = 1
\]

(31) 

\[
C_{n,n'} = 0
\]

(32) 

C. The eigenvalue problem of magnon eigenvalues

As shown in the main text, following the LSWT, a general pairwise spin Hamiltonian could be written as Eq. (3). However, it should be noted that the operators in \( \psi(k) = [a_1(k), a_2(k), a_3(k), a_4(k)] \) in the Eq. (3) of the main text satisfy the commutation relation

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

(33)

where \( I \) represents \( N \times N \) 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 \( H_{ij}(k) = I_+ H(k) \) (i.e., Eq. (6) of the main text). 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 main text, 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 main text. We ignore the third term and perform LSWT to obtain the quadratic spin Hamiltonian as shown in Eq. (3) in the main text. 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 relationships 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 \( R_{cut} \). For any given magnetic system, we propose a method to obtain the relations between SSME of different high-symmetry \( k \)-points. 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". Below, we present an example for the typical magnetic system (BNS 85,59).

As shown in Table 1 in the main text, the lattice constant \( c/a \) is 0.8. The magnetic ions are located at three nonequivalent crystallographic sites: \( 4d \) (0, 0, 0), \( 2n \) (0.25, 0.75, 0) and \( 2e \) (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 2e 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;
\[ \tau = \{0, 0, 0\}, \{0.5, 0, 0\}, \{0, 0.5, 0\}, \{0, 0, 0.8\}, \{0.75, 0.25, 0\}, \{0.25, 0.75, 0\}, \{0.25, 0.25, 0.1\}, \{0.75, 0.75, -0.1\}\];

(*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 (\(\theta_n, \phi_n\)) with the polar angles \(\theta_n\) and azimuthal angles \(\phi_n\).*)

ang = {{0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 0}, {0, 1, 0}, {0, 0, 0.8}};

(*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;

\[ \tau = \{0, 0, 0\}, \{0.5, 0, 0\}, \{0, 0.5, 0\}, \{0.5, 0.5, 0\}, \{0.75, 0.25, 0\}, \{0.25, 0.75, 0\}, \{0.25, 0.25, 0.1\}, \{0.75, 0.75, -0.1\}\];

(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 \((\theta_n, \phi_n)\) with the polar angles \(\theta_n\) and azimuthal angles \(\phi_n\). As the example in the maintext, the FM (001) state should be

(*spin directions of magnetic atoms in spherical coordinates \((\theta_n, \phi_n)\) with the polar angles \(\theta_n\) and azimuthal angles \(\phi_n\).*)

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 \(D_2\).

(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*)

\[
\begin{align*}
kk[1] & = \{0, 0, 0\}; \\
kk[2] & = \{0, 0, 0\}; \\
kk[3] & = \{0.5, 0.5, 0\}; \\
kk[4] & = \{0, 0, 0.5\}; \\
kk[5] & = \{0.5, 0.5, 0\}; \\
kname = \{"\Gamma", "X", "M", "Z", "R", "A"\};
\end{align*}
\]

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

TABLE VII. 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’ | \(R_i\) | \(J\) | \(D\) |
|-------------|------|--------|------|------|
| 0.35        | 1    | (-1, 0, 0) | \(J_1\) | \(D_{11}, D_{21}, D_{31}\) |
|             | 1    | (0, -1, 0) | \(D_{12}, D_{22}, D_{32}\) |
|             | 2    | (0, 0, 0)  | \(D_{13}, D_{23}, D_{33}\) |
|             | 3    | (-1, 0, 0) | \(-D_{11}, -D_{21}, -D_{31}\) |
|             | 3    | (0, 0, 0)  | \(-D_{12}, -D_{22}, -D_{32}\) |
|             | 4    | (0, 0, 0)  | \(-D_{13}, -D_{23}, -D_{33}\) |
|             | 4    | (0, 0, 0)  | \(-D_{14}, -D_{24}, -D_{34}\) |
| 0.36        | 1    | (0, 0, 0)  | \(J_2\) | \(D_{12}, D_{22}, D_{32}\) |
|             | 1    | (-1, -1, 0)| \(D_{13}, D_{23}, D_{33}\) |
|             | 2    | (0, 0, 0)  | \(D_{12}, D_{22}, D_{32}\) |
|             | 2    | (0, 0, 0)  | \(-D_{13}, -D_{23}, -D_{33}\) |
|             | 3    | (0, 0, 0)  | \(D_{14}, D_{24}, D_{34}\) |
|             | 3    | (-1, 0, 0) | \(-D_{11}, -D_{21}, -D_{31}\) |
|             | 4    | (0, 0, 0)  | \(D_{14}, D_{24}, D_{34}\) |
|             | 4    | (0, 0, 0)  | \(-D_{14}, -D_{24}, -D_{34}\) |
| 0.5         | 1    | (0, 0, 0)  | \(J_3\) | \(D_{13}, D_{23}, D_{33}\) |
|             | 1    | (-1, 0, 0) | \(D_{12}, D_{22}, D_{32}\) |
|             | 1    | (0, 0, 0)  | \(-D_{13}, -D_{23}, -D_{33}\) |
|             | 3    | (0, 0, 0)  | \(D_{14}, D_{24}, D_{34}\) |
|             | 3    | (-1, 0, 0) | \(-D_{11}, -D_{21}, -D_{31}\) |
|             | 4    | (0, 0, 0)  | \(D_{14}, D_{24}, D_{34}\) |
|             | 4    | (0, 0, 0)  | \(-D_{14}, -D_{24}, -D_{34}\) |

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

output[symmetry]=

\[
\begin{align*}
(x,y,z & | mx,my,mz) \\
(-y+1/2,x,z & | -my,mx,mz) \\
(y,-x+1/2,z & | my,-mx,mz) \\
(-x+1/2,-y+1/2,z & | -mx,-my,mz) \\
(-x,y,-z & | mx,my,mz) \\
(y+1/2,-x,-z & | -my,mx,mz) \\
(-y+1/2,-x+1/2,-z & | -mx,-my,mz) \\
(x+1/2,y+1/2,-z & | -mx,-my,mz)
\end{align*}
\]

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 VII. Meanwhile, the corresponding symmetry restricted DM interactions \(D(\tau_n, \tau_{n’}, R_l)\) are also listed here. Note that for 5th NN MEIs, the symmetry makes \(D_{5}^\pm = 0\), while for 7th NN MEIs, we have \(D_5 = (0, 0, 0, 0)\). 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]=

\[
\begin{align*}
M & = A = 0 \\
X & = R = 0 \\
\Gamma & = Z = 0 \\
\Gamma & - 2X + M = 0
\end{align*}
\]

where the label "M" means the quadratic sum of the magnon energies at M point "\(\sum_i \omega_i^M(M)\)”, as well as the labels of other high-symmetry \(k\) points. We can see that, up to \(J_5\), the quadratic sum of the magnon energies satisfy that \(\sum_i \omega_i^2(\Gamma) = \sum_i \omega_i^2(Z)\), \(\sum_i \omega_i^2(X) = \sum_i \omega_i^2(R)\), \(\sum_i \omega_i^2(M) = \sum_i \omega_i^2(\Gamma) + \sum_i \omega_i^2(M)\), as shown in Table III in the maintext.

As shown above, by entering the information of magnetic materials, one can use this code to obtain the relations between magnon eigenvalues easily.

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