

Theoretical investigation of magnetic dynamics in α -RuCl₃

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(Received 6 February 2017; revised manuscript received 11 May 2017; published 5 September 2017)

We study spin-wave excitations in α -RuCl₃ by the spin-wave theory. Starting from the five-orbital Hubbard model and the perturbation theory, we derive an effective isospin-1/2 model in the large Hubbard (U) limit. Based on the energy-band structure calculated from the first-principles method, we find that the effective model can be further reduced to the K - Γ model containing a ferromagnetic nearest-neighbor (NN) Kitaev interaction (K) and a NN off-diagonal exchange interaction (Γ). With the spin-wave theory, we find that the K - Γ model can give magnetic excitations, which is consistent with recent neutron-scattering experiments.

DOI: [10.1103/PhysRevB.96.115103](https://doi.org/10.1103/PhysRevB.96.115103)

I. INTRODUCTION

Currently, considerable attention has been attracted to exotic physics driven by the interplay of spin-orbital coupling (SOC), crystal fields, and electronic correlation [1–12]. Especially, in $4d$ or $5d$ transition-metal materials, neither the Hubbard interaction U nor the SOC λ can solely lead to the insulating behavior. However, the interplay between U , λ , and crystal field Δ could induce the so-called spin-orbital assisted Mott insulator [1,3,5]. In d orbitals, an electron has total angular momentum $\mathbf{J} = \mathbf{s} + \mathbf{L}$ with orbital angular momentum \mathbf{L} of five d orbitals and spin angular momentum \mathbf{s} . When d orbitals are subject to an octahedral crystal-field circumstance, these states are split into a t_{2g} triplet and an e_g doublet. For the partially filled d^5 configuration under large crystal field, the low-energy physics is dominated by the t_{2g} orbitals and it is depicted by a single hole which has an effective orbital angular momentum $l = 1$ and an effective total angular momentum $\mathbf{J}_{\text{eff}} = \mathbf{s} - \mathbf{L}'$, where $\mathbf{L}'(\mathbf{s})$ is the effective orbital (spin) angular momentum of the t_{2g} orbitals. Thus, for a large SOC, the t_{2g} multiplet is divided into a $J_{\text{eff}} = 3/2$ quartet and a $J_{\text{eff}} = 1/2$ Kramers doublet with a reduced bandwidth. Therefore, a moderate interaction U can open a Mott gap in the Kramers doublet. The significant consequence of this $J_{\text{eff}} = 1/2$ Mott insulator state is that its low-energy spin model has been shown to be the Heisenberg-Kitaev (HK) model [13], in which the celebrated Kitaev interaction is an unusual bond dependent exchange [14]. The pioneer examples are the $5d^5$ -iridate compounds $A_2\text{IrO}_3$ ($A=\text{Na}, \text{Li}$) [2,4,15–22] which contain honeycomb lattices with low-spin magnetic ions Ir^{4+} and the edge-sharing octahedral crystal field. Unfortunately, the fact that Ir ions have large neutron absorption cross sections hinders neutron studies [4,15]. In addition, the trigonal distortions arouse controversy about the application of the $J_{\text{eff}} = 1/2$ picture to iridates [23].

Recently, α -RuCl₃, which is a $4d^5$ analog of iridates, was suggested as another candidate for the realization of the Kitaev interaction term [24–28]. In contrast to iridates, the RuCl₆ octahedron is much closer to cubic and layers are weakly coupled by van der Waals interactions. Even though the value of SOC is expected to be smaller than that of the $5d$ element,

the intermediate SOC of Ru^{3+} combined with correlation effects in a narrow Ru^{3+} d band could also lead to the $J_{\text{eff}} = 1/2$ picture [24,25,29–32]. Experimentally, due to stacking faults, two different crystalline symmetries have been reported in this compound, including both $P3_112$ [25,29,33,34] ($P3$) and $C2/m$ [26,35–37] ($C2$) space groups. Neutron-scattering [25,26,32], x-ray-diffraction [37], and heat capacity [24–26,34] measurements have pointed towards a zigzag-type magnetic order at $T_{N1} \approx 14$ K and $T_{N2} \approx 8$ K, which are associated with stacking faults. Moreover, above magnetic ordering temperature the broad continuum scattering is observed not only in inelastic neutron scattering (INS) [25,38] but also in Raman scattering [39], which suggests that α -RuCl₃ may realize Kitaev physics. The INS experiments [38] suggest that the Kitaev interaction is antiferromagnetic, but below T_{N1} a spin gap near the M point is observed [25,38,40], which is not consistent with the theoretical results based on the HK model with an antiferromagnetic Kitaev interaction. Therefore, the HK model is not enough to describe the physics in α -RuCl₃. Moreover, many theoretical works have suggested that the Kitaev interaction is ferromagnetic [41–43]. Additionally, in previous work, the crystal field is expected to be large enough so that one can only take the t_{2g} manifold into account at low energies. However, the crystal-field splitting Δ between e_g and t_{2g} orbitals is estimated to be 2.2 eV from the x-ray-absorption spectroscopy data [31,44], which is comparable to Hubbard interaction U . Therefore, it is necessary to study the effect of crystal field Δ on the low-energy behavior by including all of the five d orbitals.

In this paper, based on the tight-binding energy bands from first-principles calculations on α -RuCl₃, we derive a minimal isospin model which contains only the nearest-neighbor (NN) ferromagnetic Kitaev term and isotropic antiferromagnetic off-diagonal exchange interaction, by projecting the five-orbital Hubbard model onto the lowest Kramers doublet. By analyzing the magnetic interactions, we find that the exchange between e_g and t_{2g} orbitals can enhance the NN ferromagnetic Kitaev interaction K and off-diagonal exchange Γ , and reduce the NN ferromagnetic Heisenberg interaction J . Compared with the previous works in which the signs of K are different in the crystal structures $C2$ and $P3$ [41,42], our results reveal that the signs of K are consistent in the two cases. We also find the third-NN Heisenberg interaction J_3 is largely suppressed. We investigate the magnetic dynamics of this

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model, which is consistent with the results of INS experiments [25,38,40] through the SU(2) spin-wave theory [4,45]. We further verify the validity of the minimal isospin model through the comparison to the spin-wave excitations calculated from the exchange model containing all of the $J_{\text{eff}} = 1/2$ and $3/2$ states by use of the SU(6) spin-wave theory.

The paper is organized as follows. In Sec. II, we first introduce the second-order perturbation theory and derive an effective exchange model at the strong-coupling limit. By analyzing the magnetic interactions based on the energy-band structure from the first-principles calculation, we then arrive at a minimal effective exchange model. In Sec. III, we introduce the SU(N) spin-wave theory [46,47] and verify the validity of the minimal exchange model by calculating the spin-excitation spectrum and the spin-spin correlation functions. Finally, the discussion and summary are given in Sec. IV.

II. MINIMAL EFFECTIVE MODEL

We start from the multiorbital Hubbard model, which includes all of the five $4d$ orbitals of Ru^{3+} in $\alpha\text{-RuCl}_3$. It is given as

$$H = H_t + H_\Delta + H_{\text{soc}} + H_{\text{int}}. \quad (1)$$

The kinetic-energy term H_t and crystal field H_Δ are expressed as

$$H_t = \sum_{ij,\sigma} \psi_{i\sigma}^\dagger \mathcal{T}_{ij} \psi_{j\sigma} \quad (2)$$

and

$$H_\Delta = \sum_{i,\sigma} \psi_{i\sigma}^\dagger h_i^\Delta \psi_{i\sigma}, \quad (3)$$

where $\psi_{i,\sigma}^\dagger = (d_{i,z^2,\sigma}^\dagger, d_{i,x^2-y^2,\sigma}^\dagger, d_{i,yz,\sigma}^\dagger, d_{i,xz,\sigma}^\dagger, d_{i,xy,\sigma}^\dagger)$ with $d_{i,m,\sigma}^\dagger$ creating an electron of spin σ at site i in the orbital m . The parameters of \mathcal{T}_{ij} and h_i^Δ for a tight-binding fit of the band structure based on the density-functional theory (DFT) are listed in Appendix A. $H_{\text{soc}} = \sum_i \lambda \mathbf{L}_i \cdot \mathbf{s}_i$ is the electron spin-orbital interaction. The on-site Coulomb interaction H_{int} is given by

$$\begin{aligned} H_{\text{int}} = & \frac{1}{2} \sum_{imm'nn'} \sum_{\alpha\beta\mu\nu} \delta_{\alpha\nu} \delta_{\beta\mu} \{ U \delta_{m=m'=n=n'} (1 - \delta_{\alpha\beta}) \\ & + U' \delta_{mn'} \delta_{m'n} (1 - \delta_{mm'}) + J_H \delta_{mn} \delta_{m'n'} (1 - \delta_{mm'}) \\ & + J' \delta_{mm'} \delta_{mn'} (1 - \delta_{mn}) (1 - \delta_{\alpha\beta}) \} d_{im\alpha}^\dagger d_{im'\beta}^\dagger d_{in\mu} d_{in'\nu}, \end{aligned} \quad (4)$$

where U (U') is the intraorbital (interorbital) Coulomb interaction, J_H and J' are Hund's coupling and pairing hopping, respectively. In this paper, we employ $U = U' + 2J_H$ and $J_H = J'$.

Next, we consider the large U limit and derive an effective exchange model through the second-order perturbation approximation. In the perturbation theory, the total Hamiltonian of Eq. (1) is divided into two parts $H_0 = H_{\text{int}} + H_{\text{soc}} + H_\Delta$ and $H_1 = H_t$. Here, H_0 can be written as $H_0 = \sum_i H_{0i}$ where H_{0i} denotes the Hamiltonian on the site i . Then, by projecting out the states in the high-energy subspace with the second-order perturbation approximation, we can

obtain the effective Hamiltonian in the low-energy subspace as

$$\mathcal{H}_{\text{eff}} = \sum_{ip} E_{ip} |ip\rangle_l \langle ip| + \sum_{i<j} \mathcal{H}_{ij}, \quad (5)$$

where E_{ip} is the eigenenergy of the p th low-energy eigenstate $|ip\rangle_l$ of H_{0i} . Here, the subscript l indicates that the state $|ip\rangle_l$ is in the low-energy subspace of H_{0i} . \mathcal{H}_{ij} is the effective interaction between the sites i and j by projecting the original Hamiltonian in Eq. (1) into this low-energy subspace, and it can be formally expressed as

$$\begin{aligned} \mathcal{H}_{ij} = & \sum_{pp'p_1p_1'nn'} \frac{|ip, jp'\rangle_l \langle ip_1, jp_1'|}{2\Delta E_{pp'p_1p_1'nn'}} \\ & \times (\mathcal{H}_{pp',nn'}^{j \rightarrow i} \mathcal{H}_{nn',p_1p_1'}^{i \rightarrow j} + \mathcal{H}_{pp',nn'}^{i \rightarrow j} \mathcal{H}_{nn',p_1p_1'}^{j \rightarrow i}), \end{aligned} \quad (6)$$

$$\mathcal{H}_{pp',nn'}^{j \rightarrow i} = {}_l \langle ip, jp' | H_1^{j \rightarrow i} | in, jn' \rangle_h, \quad (7)$$

$$\mathcal{H}_{nn',p_1p_1'}^{i \rightarrow j} = {}_h \langle in, jn' | H_1^{i \rightarrow j} | ip_1, jp_1' \rangle_l, \quad (8)$$

$$\begin{aligned} \frac{1}{\Delta E_{pp'p_1p_1'nn'}} = & \frac{1}{E_{ip} + E_{jp'} - E_{in} - E_{jn'}} \\ & + \frac{1}{E_{ip_1} + E_{jp_1'} - E_{in} - E_{jn'}}, \end{aligned} \quad (9)$$

where $H_1^{j \rightarrow i}$ is the hopping term from the j site to i in H_t , $|ip, jp'\rangle_l = |ip\rangle_l \otimes |jp'\rangle_l$, and $|in, jn'\rangle_h = |in\rangle_h \otimes |jn'\rangle_h$. Here, $|in\rangle_h$ is the n th eigenstate with eigenenergy E_{in} of H_{0i} in the high-energy subspace, and the subscript h indicates that $|in\rangle_h$ is in the high-energy subspace of H_{0i} .

In the limit $U \sim \Delta \gg t$ and $\lambda \gg \frac{t^2}{U}$, the local degrees of freedom are governed by the lowest two many-body states of H_{0i} , labeled by $|1\rangle$ and $|2\rangle$, which become the $J_{\text{eff}} = 1/2$ Kramers doublet exactly when Δ tends to infinity and the crystal-field splits in the t_{2g} orbitals (see Appendix A) are zero. Thus, we project H into the subspace of the Kramers doublet and expand the Hamiltonian \mathcal{H}_{eff} in the form of $S_i^\mu S_j^\nu (\mu, \nu = 0, x, y, z)$, i.e., $\mathcal{H}_{ij} = \sum_{\mu\nu pp' mm'} J_{ij}^{\mu\nu} S_{i,pp'}^\mu S_{j,mm'}^\nu |ip, jm\rangle_l \langle ip', jm'|$, where $J_{ij}^{\mu\nu}$ is the coefficient of the exchange interaction, S_i^0 is the identity matrix, and $S_{i,pp'}^{\alpha=x,y,z} = {}_l \langle ip | J_{i,\text{eff}}^\alpha | ip' \rangle_l$ is the element of the isospin matrix. The isospin operators satisfy the commutation relation $[S_i^\alpha, S_i^\beta] = i\epsilon^{\alpha\beta\gamma} S_i^\gamma$ ($\epsilon^{\alpha\beta\gamma}$ is Levi-Civita antisymmetry symbol) exactly if $\Delta = \infty$ and the crystal-field splits in the t_{2g} orbitals are zero. Due to the degeneracy of the Kramers doublet, $J_{ij}^{0\alpha}$ and $J_{ij}^{\alpha 0}$ are zeros, and the first term of Eq. (5) which is just a constant can be dropped. Therefore, we obtain an effective model involving exchange interactions up to the third NN [48]:

$$\begin{aligned} H_{\text{eff}} = & \sum_{(ij) \in \gamma(\alpha\beta)} [J^\gamma S_i \cdot S_j + K^\gamma S_i^\gamma S_j^\gamma + \Gamma^\gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \\ & + \Gamma'^\gamma (S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\beta + S_i^\gamma S_j^\alpha)] \\ & + \sum_{\langle\langle ij \rangle\rangle \in \gamma} (J_2^\gamma S_i \cdot S_j + K_2^\gamma S_i^\gamma S_j^\gamma) \\ & + \sum_{\langle\langle\langle ij \rangle\rangle\rangle \in \gamma} (J_3^\gamma S_i \cdot S_j + K_3^\gamma S_i^\gamma S_j^\gamma). \end{aligned} \quad (10)$$

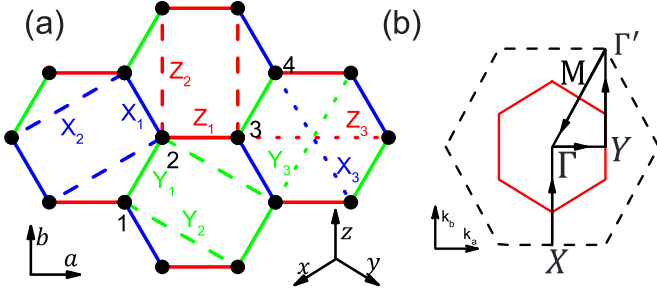


FIG. 1. (a) Lattice structure of Ru^{3+} in $\alpha\text{-RuCl}_3$. Solid, dashed, and dotted lines label first-, second-, and third-NN bonds on the honeycomb lattice, respectively. Red, green, and blue colors denote the Z, Y, and X bonds, respectively. a, b refer to the axes in the honeycomb layer, while x, y, z are the cubic axes of the local octahedron. Sites within a magnetic unit cell for the zigzag order are labeled by 1–4. (b) Structure of the reciprocal space. The red solid lines represent the first Brillouin zone. Γ, Γ', X, Y , and M denote the symmetrical points.

Here, $\langle ij \rangle$, $\langle\langle ij \rangle\rangle$, and $\langle\langle\langle ij \rangle\rangle\rangle$ denote the first-NN, second-NN, and third-NN bonds, respectively. γ represents the direction of each bond as shown in Fig. 1. For Z-type, X-type, and Y-type bonds in Fig. 1, $(\alpha\beta)$'s are (xy) , (yz) , and (zx) respectively. J and K are the magnitude of the Heisenberg and Kitaev interactions; Γ and Γ' are the off-diagonal exchanges. The second- and third-NN exchange interactions are generally smaller than the first-NN interactions since their hopping integrals are much smaller than the first-NN ones (see Appendix B), so only the main terms of the second- and third-NN exchange interactions are retained in Eq. (10). In the case of the $P3$ space group, the interactions are invariant for different directions due to the C_3 symmetry. However, for the low symmetric $C2$ space group, the interactions on the X and Y bonds are equal but different from those on the Z bonds. Moreover, in the case of the $C2$ space group, to make the J and Γ' terms on the X-type and Y-type bonds equal for the spin directions, we will take their average values [41].

The exchange interaction parameters in Eq. (10) depend on the hopping integrals between various orbitals, crystal field Δ , SOC λ , Hubbard interaction U , and Hund's coupling J_H . The hopping integrals are determined from the first-principles calculations as listed in Appendix A. The dependences of the exchange interactions on Δ , λ , U , and J_H are shown in Fig. 2. To simplify the comparison, the values of interactions are bond averaged in the $C2$ case, so the superscript γ is omitted. As the second- and third-NN terms are small in contrast to the first-NN terms, we only present the values of the first-NN terms in Fig. 2. In Figs. 2(a)–2(c), we fix $\Delta = 2.1$ eV, which is suitable for $\alpha\text{-RuCl}_3$. Their Δ dependences are then presented in Fig. 2(d). To investigate the effect of the e_g orbital, we deliberate to choose an unrealistic large $\Delta = 210$ eV and the results are shown in Figs. 2(g)–2(i) for a comparison.

The noticeable overall feature in Figs. 2(a)–2(d) is that the Heisenberg exchange term is much smaller than other terms in an extended range of parameters for $\lambda < 0.15$ eV which is the estimated maximum value for λ [25,31,41,44,49]. This arises from the near offset between the contributions to the J term from the interband e_g - t_{2g} superexchange channels

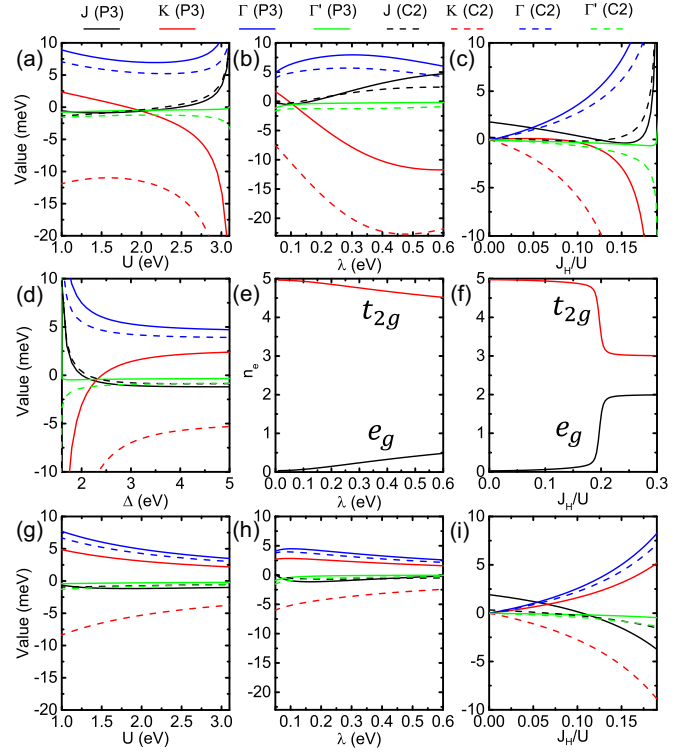


FIG. 2. Dependence of the first-NN interactions on the parameters for the $C2$ case (dashed) and $P3$ case (solid). (a) U dependence with $\Delta = 2.10$ eV, $\lambda = 0.14$ eV, and $J_H/U = 0.14$. (b) λ dependence with $\Delta = 2.10$ eV, $U = 2.31$ eV, and $J_H/U = 0.14$. (c) J_H/U dependence with $\Delta = 2.10$ eV, $U = 2.31$ eV and $\lambda = 0.14$ eV. (d) Δ dependence with $U = 2.31$ eV, $\lambda = 0.14$ eV, and $J_H/U = 0.14$. (g)–(i) With $\Delta = 210$ eV corresponding to (a)–(c). (e) and (f) show the number of electrons in the Kramers doublet per site corresponding to (b) and (c), respectively. The black (red) line is e_g (t_{2g}) orbitals.

and intraband t_{2g} channels [18]. Figure 2(a) shows that the magnitude of the exchange interactions has a trend to decrease and then increase with the increase of U . As U increases the gap between the Kramers doublet and other excited states, the effective exchange interactions will decrease with U according to Eq. (6). In Fig. 2(a), we fix the value of J_H/U , so Hund's coupling J_H increases with U . For the $4d^5$ electron configuration, Hund's coupling will decrease the energies of the excited states which contain a large weight of e_g orbitals, so the exchange interactions increase with J_H . Therefore, there is a competing relation between U and J_H in determining the exchange interactions. We can see this point more clearly in Fig. 2(g), where the crystal field Δ is set at a deliberate large value, so that the effect of the e_g orbitals is excluded and the effect of J_H is suppressed. In this case, all the exchange interactions decrease with U . In Fig. 2(i), the large J_H/U induces the ferromagnetic J interaction and enhances the values of the antiferromagnetic K interaction in the $P3$ case, the ferromagnetic Γ interactions in both cases. The different signs of the K interactions in two cases depend on the hoppings in the t_{2g} orbitals. The antiferromagnetic K term in the $P3$ cases comes mainly from the direct hopping t_3 between the d_{xy} orbitals for the Z bond. The K term in the

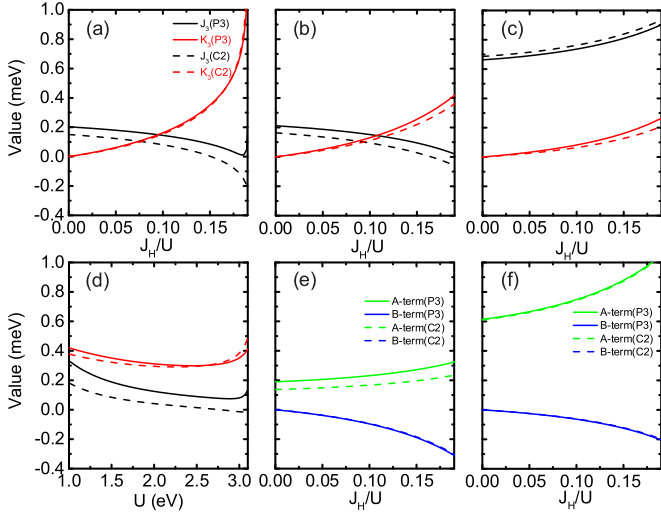


FIG. 3. Dependence of the third-NN Heisenberg and Kitaev interactions J_3 and K_3 on parameters J_H/U and U . The Hubbard interaction $U = 2.31$ eV and SOC $\lambda = 0.14$ eV are used except in panel (d). (a) $\Delta = 2.10$ eV. (b) $\Delta = 210$ eV. (c) For the three-orbital model containing only t_{2g} orbitals. (d) U dependence with $J_H/U = 0.14$, $\lambda = 0.14$ eV, and $\Delta = 2.10$ eV. (e) and (f) The A and B components of the J_3 term (see text), and the parameters correspond to (b) and (c), respectively. Solid (dash) lines denote the P3 (C2) case.

C2 case is attributed to the indirect hopping t_2 between t_{2g} orbitals via chlorine ions. However, when Δ is reduced to be comparable to the Hubbard U , as shown in panel (c), a large J_H leads to the ferromagnetic K in the P3 case and the antiferromagnetic J in both cases. It is also supported by their Δ dependence. This is because a large J_H/Δ increases the mixing of the e_g and t_{2g} orbitals in the Kramers doublet, as shown in panel (f) where the number of electrons n_e in the e_g orbitals increases rapidly for $J_H/U > 0.19$. Moreover, by comparing Figs. 2(c) and 2(i), we find that the exchange channels between the e_g and t_{2g} orbitals can enhance the magnitude of the Γ and K interactions. When the weight of e_g orbitals in the Kramers doublet increases rapidly, the values of interactions are divergent and the $J_{\text{eff}} = 1/2$ picture is no longer applicable. From the λ dependence as shown in Figs. 2(b), 2(e) and 2(h), we can see that in the large Δ limit the values of interactions are suppressed with λ owing to the enhancement of the gap between the $J_{\text{eff}} = 1/2$ and $3/2$ states, which is consistent with previous work [41]. If Δ is reduced, the increase of λ results in the same effect as the increase of J_H/U , as seen in Figs. 2(b) and 2(e). However, when λ is increased to 0.3 eV, the values of interactions increase slowly and even decrease. This is because the effect of the gap between the $J_{\text{eff}} = 1/2$ and $3/2$ states on interactions is greater than that of the e_g - t_{2g} channels.

In α - RuCl_3 , $\Delta \approx 2.2$ eV, $U = 2 \sim 3$ eV, and $\lambda = 0.13 \sim 0.15$ eV [25,31,41,44,49], so we find that the leading exchange interactions are K and Γ terms according to the above discussion. Thus, we arrive at a minimal exchange model:

$$H_{\text{min}} = \sum_{(ij) \in \gamma(\alpha\beta)} [K^\gamma S_i^\alpha S_j^\beta + \Gamma^\gamma (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha)]. \quad (11)$$

The symbols are the same as those in Eq. (10). In the P3 case, the symmetry allows $K^z = K^x = K^y$ and $\Gamma^z = \Gamma^x = \Gamma^y$, while in the C2 case $K^x = K^y = K^z + \delta_1$ and $\Gamma^x = \Gamma^y = \Gamma^z + \delta_2$ with a small amount δ_1 and δ_2 .

In previous works [41,42] where only the t_{2g} orbitals are considered to fit the band structures from the first-principles calculation, the third-NN Heisenberg interaction J_3 is found to have a relatively large value. However, we find that J_3 is suppressed when all five d orbitals are used (see Fig. 3). In addition, as shown in Fig. 3(a), J_3 is smaller than the third-NN Kitaev interaction K_3 when the ratio J_H/U is greater than 0.1. Similarly, for a fixed $J_H/U = 0.14$, J_3 is always smaller than K_3 in the range of U we considered [Fig. 3(d)]. To test whether this suppression of J_3 is induced by the exchange channels between the e_g and t_{2g} orbitals, we artificially increase the crystal field to be $\Delta = 210$ eV to exclude the effects of e_g orbitals, and the results are shown in Fig. 3(b). In this case, J_3 is still suppressed, so the effect of e_g - t_{2g} mixing on the third-NN diagonal magnetic interactions J_3 and K_3 is weak. However, if we use the t_{2g} three-orbital model (see Appendix A) to derive the magnetic interactions, the third-NN Heisenberg interaction J_3 is relatively large, as shown in Fig. 3(c), which is consistent with the previous studies [41,42]. This difference in results between the five-orbital and three-orbital models mainly comes from the change of signs of the intraorbital hopping integrals along the third-NN bond in t_{2g} orbitals. In the three-orbital model, the signs of these hopping integrals are the same (see Appendix A), while for the five-orbital model one of them has a different sign (see Appendix A). According to Eq. (25) in Ref. [41] (also see Ref. [48]), we have

$$J_3 = \frac{4A}{9}(t_{xx} + t_{yy} + t_{zz})^2 + \frac{8B}{9}[-9t_{xz}^2 + (t_{xx} + t_{yy} - 2t_{zz})(t_{yy} + t_{zz} - 2t_{xx})] \quad (12)$$

on the Z bond. Here, the subscripts x , y , and z in the hopping parameters represent the d_{yz} , d_{xz} , and d_{xy} orbitals, respectively. To see the separate effects of the two terms in Eq. (12), we label its first and second terms as the A term and B term, respectively. When the signs of the intraorbital hopping integrals are the same, the intensity of the A term is much greater than that of the B term, as shown in Fig. 3(f). However, when the signs of these hopping integrals are different, the magnitude of the A term is largely reduced, as shown in Fig. 3(e), which leads to a reduction in the third-NN Heisenberg term J_3 .

III. SPIN-WAVE EXCITATION

We now turn to the calculation of the spin-wave excitations. Using the approach of the spin-particle mapping with Schwinger-Wigner bosons [50], we map the low-energy state $|ip\rangle_l$ to $b_{ip}^\dagger|0\rangle$ with condition $\sum_p b_{ip}^\dagger b_{ip} = 1$, where b_{ip}^\dagger creates a boson on site i with quantum number p and $|0\rangle$ is the vacuum state without any bosons. Here, we employ the fundamental irreducible representation of the $\text{SU}(N)$ group with N the number of p . If the ground state of the system is an ordered state, one of the bosonic modes will condense. Therefore, in the local mean-field approximation [47], there exists a stable solution to minimize the ground-state energy $\langle G|\mathcal{H}_{\text{eff}}|G\rangle$, where $|G\rangle = \prod_i \tilde{b}_{i,0}^\dagger|0\rangle$ is the mean-field ground

state represented by the condensed boson $\tilde{b}_{i,0}^\dagger$, which can be expressed as

$$\tilde{b}_{i,0}^\dagger = \sum_p U_{0p}(\mathbf{x}_i) b_{i,p}^\dagger. \quad (13)$$

For the case of $J_{\text{eff}} = 1/2$ discussed above, the local rotation matrix $U_{pp'}(\mathbf{x}_i)$ depends on two parameters [4,45], i.e., $\mathbf{x}_i = (\theta_i, \phi_i)$, which are the parameters of the polar coordinates in the local frame. For the $SU(N)$ spin-wave theory, the local rotation matrix has $2(N-1)$ parameters [48,51,52], i.e., $\mathbf{x}_i = (\theta_{i,1}, \dots, \theta_{i,N-1}, \phi_{i,1}, \dots, \phi_{i,N-1})$. When one of the bosons condenses, the corresponding creation and annihilation operators can be replaced by the number c [47], and they satisfy

$$\begin{aligned} \tilde{b}_{i,0}^\dagger &\simeq \tilde{b}_{i,0} = c = \sqrt{1 - \sum_{p \neq 0} \tilde{b}_{i,p}^\dagger \tilde{b}_{i,p}} \\ &= 1 - \frac{1}{2} \sum_{p \neq 0} \tilde{b}_{i,p}^\dagger \tilde{b}_{i,p} + \dots, \end{aligned} \quad (14)$$

where the $N-1$ bosons $\tilde{b}_{i,p \neq 0}$ become the Holstein-Primakoff bosons now. By substituting Eqs. (14) and (13) into the Hamiltonian \mathcal{H}_{eff} we obtain the Hamiltonian in terms of rotated bosons as follows:

$$\mathcal{H}_{\text{eff}} = \mathcal{H}_0(\{\mathbf{x}_i\}) + \mathcal{H}_1(\{\mathbf{x}_i\}) + \mathcal{H}_2(\{\mathbf{x}_i\}) + \dots, \quad (15)$$

where the subscripts of \mathcal{H} denote the number of rotated bosons. In the linear spin-wave approximation, we only retain the first three terms of Eq. (15). To find the ground state, we minimize the zeroth-order term $\mathcal{H}_0(\{\mathbf{x}_i\})$. When a set of proper parameters $\{\mathbf{x}_i^0\}$ is found, the first-order term $\mathcal{H}_1(\{\mathbf{x}_i\})$ vanishes [45]. Then, the dispersion is obtained by solving the quadratic term $\mathcal{H}_2(\{\mathbf{x}_i\})$ [48].

To search for various possible magnetic ground states including the zigzag order, we choose a magnetic unit cell involving four sites (see Fig. 1) to minimize the ground-state energy. To compare to the INS experiments [25,38,40], we use the $SU(N)$ [47] spin-wave theory to calculate the correlation function $\tilde{S}(\mathbf{q}, \omega)$ (zero temperature), which is defined as

$$\tilde{S}(\mathbf{q}, \omega) = \frac{1}{N} \sum_{ij} e^{iq(r_i - r_j)} \int_{-\infty}^{\infty} \langle \mathbf{Q}_i \mathbf{Q}_j(t) \rangle e^{-i\omega t} dt, \quad (16)$$

with $\mathbf{Q}_j(t) = e^{iHt} \mathbf{Q}_j e^{-iHt}$. For the effective and minimum isospin models, the correlation function of the isospin operator $\mathbf{Q}_i = \sum_{pp'=1,2} \langle p | J_{i,\text{eff}} | p' \rangle b_p^\dagger b_{p'}$ is calculated by the $SU(2)$ spin-wave theory.

First, to obtain suitable values of K and Γ in the minimal isospin model in Eq. (11) for $\alpha\text{-RuCl}_3$, we optimize K and Γ to make the low-energy isospin excitations of Eq. (11) be in accordance with those of Eq. (10) obtained through projecting the five-orbital Hubbard model to the Kramers doublet. The hopping parameters in the five-orbital Hubbard model are from the first-principles calculations as listed in Appendix A, and the interaction parameters are chosen as $U = 2.31$ eV, $J_H = 0.32$ eV, and $\lambda = 0.14$ eV, which are appropriate for $\alpha\text{-RuCl}_3$ [25,31,41,44,49]. Then, we obtain the exchange interaction parameters in Eq. (10), which are listed in Appendix B. By a comparison of the spin-excitation spectrum for the effective exchange model in Eq. (10) and the minimal

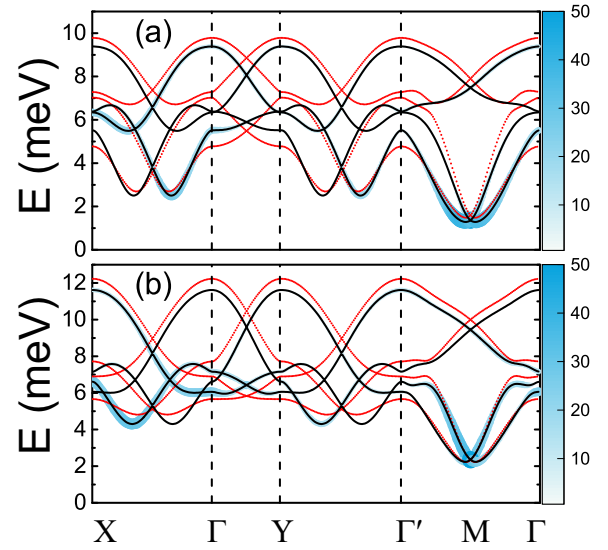


FIG. 4. Spin-wave dispersions along the high-symmetry direction $X - \Gamma - Y - \Gamma' - M - \Gamma$ [see Fig. 1(b)] in the $P3$ case (a) and $C2$ case (b). The black solid and red dashed lines correspond to the results calculated based on the minimal isospin model in Eq. (11) and the effective isospin model in Eq. (10), respectively. The sizes of the colors indicate the magnitude of the isospin correlation function \tilde{S} calculated based on the minimal isospin model in Eq. (11).

isospin model in Eq. (11), we find that $K^y = -5.50$ meV and $\Gamma^y = 7.60$ meV ($K^z = -10.92$ meV, $K^x = -10.86$ meV, $\Gamma^z = 6.20$ meV, and $\Gamma^x = 6.00$ meV) in Eq. (11) can give a consistent fit to those obtained by Eq. (10) in the $P3$ ($C2$) case, as shown in Fig. 4. Moreover, the classical ground states of both the minimal isospin model in Eq. (11) and the effective exchange model in Eq. (10) show the same zigzag magnetic order for these parameters.

We then perform the calculations of the $SU(2)$ spin-wave theory based on the minimal isospin model to compute the correlation function in Eq. (16) for the $J_{\text{eff}} = 1/2$ isospin. The spin-wave Hamiltonian of the minimal isospin model in Eq. (11) is listed in Appendix C. The results are presented in Fig. 4. We find that the isospin excitations show a gap at the M point and the maximal intensity is also near the M point, which agrees well with the INS experiments [25,38,40]. Moreover, the direction of the magnetic moment $\mathbf{m}_i = \langle G | \sum_{pp'} \langle p | \mathbf{s}_i + \mathbf{L}_i | p' \rangle b_p^\dagger b_{p'} | G \rangle$, in which \mathbf{L}_i is the orbital angular momentum of the five d orbitals, tilts 36° (48°) out of the ab plane in the $P3$ ($C2$) case, which roughly coincides with the experimental result of Ref. [37].

As shown in Figs. 4(a) and 4(b), we can find the dispersions show no qualitative difference in the $P3$ and $C2$ cases, though the maximum intensities of the correlation function \tilde{S} near the M point in the two cases are in different branches. In addition, the gaps of the isospin excitations in the $P3$ and $C2$ cases are also consistent with each other, though the values of K and Γ are obviously different.

To further check the validity of the K - Γ model shown in Eq. (11), we construct a more complex effective exchange model by projecting the five-orbital Hubbard model in Eq. (1) to the subspace of the lowest six many-body

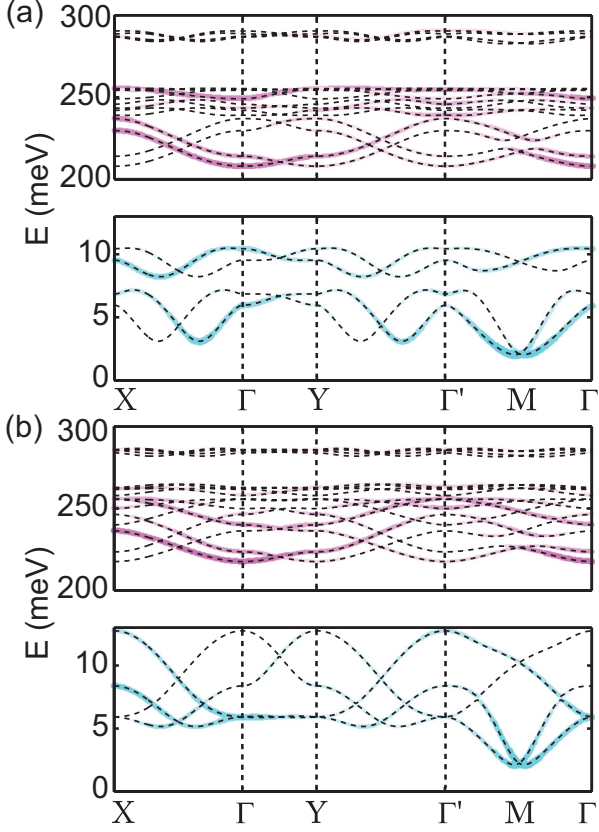


FIG. 5. Spin-wave dispersion (dashed) for the complex effective exchange model in (a) and (b) corresponding to the $P3$ case and $C2$ case, respectively. The sizes of the colors indicate the magnitude of correlation function \tilde{S} , and the cyan and magenta colors represent the isospin and spin-orbital excitations, respectively. The large gap between 200 and 13 meV results from SOC.

states of the $4d^5$ electron configuration. In this enlarged subspace, besides the $J_{\text{eff}} = 1/2$ doublet, the $J_{\text{eff}} = 3/2$ quartet is also included. Thus, in addition to the $J_{\text{eff}} = 1/2$ isospin excitations, there are also the spin-orbital excitations between the $J_{\text{eff}} = 1/2$ and $3/2$ states. In this case, we use the $SU(6)$ spin-wave theory, in which the local rotation parameters become $\mathbf{x}_i = (\theta_{i,1}, \dots, \theta_{i,5}, \phi_{i,1}, \dots, \phi_{i,5})$. We calculate the correlation functions in Eq. (16) of the quantities $\mathcal{Q}_i = \sum_{pp'=1,2} \langle p | 2s_i + \mathbf{L}_i | p' \rangle_l b_{ip}^\dagger b_{ip'}$ and $\sum_{p=1,2} \sum_{p'=3}^6 \langle p | 2s_i + \mathbf{L}_i | p' \rangle_l b_{ip}^\dagger b_{ip'} + \text{H.c.}$, which correspond to the magnetic excitations in the $J_{\text{eff}} = 1/2$ isospin subspace and those between the $J_{\text{eff}} = 1/2$ and $3/2$ states, respectively. Here, s_i and \mathbf{L}_i are the spin and orbital angular momenta, respectively. The factor 2 of s_i is the Landé g factor of spin. By performing the calculations, we find the ground state of this effective model is of a zigzag spin order, in which the Kramers doublet, $\langle G | \sum_{p=1,2} b_{ip}^\dagger b_{ip} | G \rangle$, has the dominant weight. This result provides further support to the $J_{\text{eff}} = 1/2$ isospin picture on which the minimal isospin model is based. More importantly, the low-energy spin-wave excitations (see Fig. 5) calculated from the $SU(6)$ spin-wave theory based on this effective exchange model are also dominated by the $J_{\text{eff}} = 1/2$ isospin, which agrees well with

TABLE I. Bond-averaged values of magnetic interactions (in meV). J_3 represents the third-NN Heisenberg interaction. The results from Refs. [41,42,49] are also presented for a comparison.

Structure	J	K	Γ	J_3
$C2$	-0.3	-10.9	6.1	0.03
$P3$	0.1	-5.5	7.6	0.1
$C2$ [41]	-1.7	-6.7	6.6	2.7
$P3$ [41]	-5.5	7.6	8.4	2.3
$C2$ [42]	-1.0	-8.2	4.2	
$P3$ [42,49]	-3.5	4.6	6.4	0.8

those of the minimal isospin model (see Fig. 4). Thus, the minimal isospin model in Eq. (11) is suitable for describing the low-energy physics in $\alpha\text{-RuCl}_3$, and it can be used to investigate other magnetic properties such as the physics of Kitaev spin liquid. In addition, besides the $J_{\text{eff}} = 1/2$ isospin excitations, we expect that the spin-orbital excitations between the $J_{\text{eff}} = 1/2$ and $3/2$ states revealed by the $SU(6)$ spin-wave calculations in the high-energy parts of Fig. 5 can be observed by the future resonant inelastic x-ray-scattering experiments.

IV. DISCUSSION AND SUMMARY

We derive a minimal effective isospin model from the five-orbital Hubbard model using the energy bands obtained from the first-principles calculations for $\alpha\text{-RuCl}_3$. The minimal model contains the ferromagnetic Kitaev term and the antiferromagnetic off-diagonal exchange term. We find that the e_g - t_{2g} interband superexchange channels play an important role in determining the effective exchange interactions on the NN bonds in $\alpha\text{-RuCl}_3$. In the previous works [41,42,49], the effects of the e_g - t_{2g} mixing on the magnetic interactions have not been investigated in detail. In Refs. [41,42], they only consider the t_{2g} orbitals to study the magnetic interactions and suggest the Kitaev interaction for the $P3$ crystal structure is antiferromagnetic, as shown in Table I. Although the authors of Ref. [49] suggest that the e_g - t_{2g} mixing enhances the antiferromagnetic Kitaev interaction $K > 0$ and the ferromagnetic Heisenberg interaction $J < 0$, they neglect the intra-atomic exchange interaction between the e_g and t_{2g} orbitals. Here, by considering the Coulomb interactions between all five d orbitals, we find that the e_g - t_{2g} mixing induces the ferromagnetic Kitaev coupling $K < 0$ in both the $C2$ and $P3$ crystal structures and reduces the Heisenberg interaction J in both structures. Compared with the previous studies, the third-NN Heisenberg interaction J_3 is also largely suppressed. This is caused by the different signs of the third-NN diagonal hopping integrals in the t_{2g} orbitals. If the signs are all minus, the third-NN Heisenberg interaction J_3 is greater than the third-NN Kitaev interaction K_3 , which is consistent with the result from Ref. [41].

Based on this effective isospin model we investigate the spin-wave excitation using the linear spin-wave theory and find

it is consistent with the recent neutron scattering on α -RuCl₃ [40], especially the gap opening in the magnon dispersion. In our minimal K - Γ model, the basic reason for the gap opening is that it lacks continuous rotation symmetry, which prevents the Goldstone modes emerging in the magnetic ordering phase.

In this paper, we only consider a four-site unit cell in searching for magnetic order within the spin-wave theory, so we cannot exclude the possibility of the incommensurate state as proposed in Ref. [22]. Therefore, the zigzag state may be a local minimum for the minimal model, and it may need small additional magnetic interactions to stabilize the zigzag state as a global minimum. However, since the experimental results have shown a zigzag magnetic ground state, our approach is still appropriate. Indeed, the K - Γ model is just a minimal model to describe the magnetic properties of α -RuCl₃, and it does not completely exclude the possible existence of other small exchange terms. In fact, from Fig. 2 we can find that those exchange terms, which are not included in the minimum K - Γ model, indeed have small nonzero values. These small terms, such as the third-NN Heisenberg interaction J_3 , may stabilize the zigzag state, but are expected to have little effect on the magnetic dynamics described by the minimum K - Γ model.

ACKNOWLEDGMENTS

This paper was supported by the National Natural Science Foundation of China (Grants No. 11374138, No. 11674158, and No. 11190023) and National Key Projects for Research and Development of China (Grant No. 2016YFA0300401).

APPENDIX A: PARAMETERS FOR TIGHT-BINDING MODELS AND REPRESENTATIONS OF ANGULAR MOMENTA

The electronic structure calculations are performed with the generalized gradient approximation for the exchange-correlation functional as implemented in the Quantum ESPRESSO package [53] based on the density-functional theory. To avoid double counting of the SOC [41], the SOC was not included in these calculations. The five-orbital parameters (TB5) in the hopping matrix \mathcal{T}_{ij} from the maximally localized Wannier orbital [54] calculation are shown in Tables II–IV for the first, second, and third NN, respectively. Here, only the parameters along the Z - and X -type bonds are shown; for other bonds in the $P3$ ($C2$) case, the holistic hopping matrix can be recovered by applying inversion operations and C_3 rotations along the c axis perpendicular to the ab plane (C_2 rotations along the Z_1 bond). For comparison, we also list the values of hopping integrals from several previous works [41,42,49], which only have three-orbital parameters. In our $C2$ case, the crystal structure is from Ref. [37]. In our $P3$ case, an ideal chlorine octahedron is considered and the lattice constants are fixed at $a_0 = 5.97$ Å, $b_0 = 5.97$ Å, and $c_0 = 17.2$ Å [25]. The electron operators are expressed as $\psi_{i,\sigma}^\dagger = (d_{i,z^2,\sigma}^\dagger, d_{i,x^2-y^2,\sigma}^\dagger, d_{i,yz,\sigma}^\dagger, d_{i,xz,\sigma}^\dagger, d_{i,xy,\sigma}^\dagger)$ and $\psi_{i,\sigma} = (d_{i,yz,\sigma}, d_{i,xz,\sigma}, d_{i,xy,\sigma}, d_{i,x^2-y^2,\sigma}, d_{i,z^2,\sigma})$ for five-orbital and three-orbital models,

TABLE II. Hopping parameters (in meV) for the first NN. A and B are the sublattice indices; Z_1 and X_1 bonds are shown in Fig. 1 (a). The results from Refs. [41,42,49] are also presented for a comparison.

Bond	\mathcal{T}_{ij}					
	$C2$	$P3$	$C2$ [41]	$C2$ [42]	$P3$ [49]	
$Z_1:$						
$d_z \rightarrow d_z$	22.9	23.0				
$A \rightarrow B$ $d_{x^2-y^2} \rightarrow d_{x^2-y^2}$	-30.4	-91.0				
$d_{yz} \rightarrow d_{yz}$	42.8	61.4	50.9	36.0	65.0	
$d_{xz} \rightarrow d_{xz}$	42.8	61.4	50.9	36.0	66.0	
$d_{xy} \rightarrow d_{xy}$	-117.1	-206.9	-154.0	-62.0	-229.0	
$d_z \leftrightarrow d_{x^2-y^2}$	0.0	0.0				
$d_z \leftrightarrow d_{yz}$	13.8	-0.7				
$d_z \leftrightarrow d_{xz}$	13.8	-0.7				
$d_z \leftrightarrow d_{xy}$	268.0	212.4				
$d_{x^2-y^2} \leftrightarrow d_{yz}$	-7.5	-0.3				
$d_{x^2-y^2} \leftrightarrow d_{xz}$	7.5	0.3				
$d_{x^2-y^2} \leftrightarrow d_{xy}$	0.0	0.0				
$d_{yz} \leftrightarrow d_{xz}$	156.6	108.7	158.2	191.0	114.0	
$d_{yz} \leftrightarrow d_{xy}$	-21.4	-4.5	-20.2	-24.0	-10.0	
$d_{xz} \leftrightarrow d_{xy}$	-21.4	-4.5	-20.2	-24.0	-10.0	
$X_1:$						
$d_z \rightarrow d_z$	-14.9	-62.5				
$A \rightarrow B$ $d_{x^2-y^2} \rightarrow d_{x^2-y^2}$	11.4	-11.3				
$d_{yz} \rightarrow d_{yz}$	-104.5	-206.9	-103.1	-75.0	-229.0	
$d_{xz} \rightarrow d_{xz}$	42.1	61.4	44.9	37.0	65.0	
$d_{xy} \rightarrow d_{xy}$	41.6	61.4	45.8	37.0	66.0	
$d_z \leftrightarrow d_{x^2-y^2}$	-16.8	-49.4				
$d_z \leftrightarrow d_{yz}$	-137.3	-106.2				
$d_z \leftrightarrow d_{xz}$	2.3	-0.6				
$d_z \leftrightarrow d_{xy}$	-9.8	0.1				
$d_{x^2-y^2} \leftrightarrow d_{yz}$	232.7	183.9				
$d_{x^2-y^2} \leftrightarrow d_{xz}$	11.7	-0.5				
$d_{x^2-y^2} \leftrightarrow d_{xy}$	2.8	-0.8				
$d_{yz} \leftrightarrow d_{xz}$	-12.7	-4.5	-15.1	-26.0	-10.0	
$d_{yz} \leftrightarrow d_{xy}$	-13.2	-4.5	-10.9	-26.0	-10.0	
$d_{xz} \leftrightarrow d_{xy}$	159.9	108.7	162.2	182.0	114.0	

respectively. The crystal field in the $P3$ case is given by

$$h_i^\Delta = \begin{pmatrix} \Delta & 0 & \Delta'_2 & \Delta'_2 & -2\Delta'_2 \\ 0 & \Delta & -\sqrt{3}\Delta'_2 & \sqrt{3}\Delta'_2 & 0 \\ \Delta'_2 & -\sqrt{3}\Delta'_2 & 0 & \Delta'_3 & \Delta'_3 \\ \Delta'_2 & \sqrt{3}\Delta'_2 & \Delta'_3 & 0 & \Delta'_3 \\ -2\Delta'_2 & 0 & \Delta'_3 & \Delta'_3 & 0 \end{pmatrix} \quad (A1)$$

with $\Delta = 1980$ meV, $\Delta'_2 = 15$ meV, and $\Delta'_3 = -8.6$ meV. The matrix representation is the same as that defined in Eq. (3). Due to the high symmetry in the $P3$ case, there is only one kind of crystal-field split in the t_{2g} orbitals. However, the low symmetry in the $C2$ case allows three kinds of crystal-field splits in the t_{2g} orbitals, as shown in the following text. For the $C2$ case, the crystal field is written as

$$h_i^\Delta = \begin{pmatrix} \Delta + 4.4 & 0 & 8.1 & 8.1 & -64.2 \\ 0 & \Delta & -60.9 & 60.9 & 0 \\ 8.1 & -60.9 & 0 & \Delta_1 & \Delta_2 \\ 8.1 & 60.9 & \Delta_1 & 0 & \Delta_2 \\ -64.2 & 0 & \Delta_2 & \Delta_2 & \Delta_3 \end{pmatrix} \quad (A2)$$

TABLE III. Hopping parameters (in meV) for the second NN. A is the sublattice index; Z_2 and X_2 bonds are shown in Fig. 1(a). The results from Refs. [41,42,49] are also presented for a comparison.

Bond	T_{ij}				
	C2	P3	C2 [41]	C2 [42]	P3 [49]
Z_2 :					
$d_z \rightarrow d_z$	12.2	4.6			
$A \rightarrow A$ $d_{x^2-y^2} \rightarrow d_{x^2-y^2}$	-1.1	-2.2			
$d_{yz} \rightarrow d_{yz}$	-5.9	-0.3	-4.7		0.0
$d_{xz} \rightarrow d_{xz}$	-5.9	-0.3	-4.7		0.0
$d_{xy} \rightarrow d_{xy}$	-4.6	-0.5	-0.4		0.0
$d_z \rightarrow d_{x^2-y^2}$	-5.7	-1.5			
$d_z \rightarrow d_{yz}$	-15.5	-15.9			
$d_z \rightarrow d_{xz}$	-11.8	-8.4			
$d_z \rightarrow d_{xy}$	73.0	65.9			
$d_{x^2-y^2} \rightarrow d_z$	5.7	1.8			
$d_{yz} \rightarrow d_z$	-11.8	-8.3			
$d_{xz} \rightarrow d_z$	-15.5	-15.8			
$d_{xy} \rightarrow d_z$	73.0	65.8			
$d_{x^2-y^2} \rightarrow d_{yz}$	-2.7	-1.3			
$d_{x^2-y^2} \rightarrow d_{xz}$	4.6	3.5			
$d_{x^2-y^2} \rightarrow d_{xy}$	4.2	2.1			
$d_{yz} \rightarrow d_{x^2-y^2}$	-4.6	-3.6			
$d_{xz} \rightarrow d_{x^2-y^2}$	2.7	1.0			
$d_{xy} \rightarrow d_{x^2-y^2}$	-4.2	-2.1			
$d_{yz} \rightarrow d_{xz}$	-43.5	-37.0	-23.9		-20.0
$d_{yz} \rightarrow d_{xy}$	-0.9	4.6	-1.7		3.0
$d_{xz} \rightarrow d_{xy}$	8.5	6.3	11.6		6.0
$d_{xz} \rightarrow d_{yz}$	-63.1	-58.0	-60.7		-58.0
$d_{xy} \rightarrow d_{yz}$	8.5	6.3	11.6		6.0
$d_{xy} \rightarrow d_{xz}$	-0.9	4.6	-1.7		4.0
X_2 :					
$d_z \rightarrow d_z$	1.9	-0.4			
$A \rightarrow A$ $d_{x^2-y^2} \rightarrow d_{x^2-y^2}$	9.9	2.8			
$d_{yz} \rightarrow d_{yz}$	-4.3	-0.5	-0.4		0.0
$d_{xz} \rightarrow d_{xz}$	-6.7	-0.3	-4.5		0.0
$d_{xy} \rightarrow d_{xy}$	-5.5	-0.3	-3.2		0.0
$d_z \rightarrow d_{x^2-y^2}$	-11.5	-4.6			
$d_z \rightarrow d_{yz}$	-40.2	-34.8			
$d_z \rightarrow d_{xz}$	9.9	9.1			
$d_z \rightarrow d_{xy}$	1.4	1.2			
$d_{x^2-y^2} \rightarrow d_z$	0.2	-1.3			
$d_{yz} \rightarrow d_z$	-33.1	-31.1			
$d_{xz} \rightarrow d_z$	9.6	7.3			
$d_{xy} \rightarrow d_z$	3.9	7.0			
$d_{x^2-y^2} \rightarrow d_{yz}$	61.3	56.0			
$d_{x^2-y^2} \rightarrow d_{xz}$	-11.3	-13.1			
$d_{x^2-y^2} \rightarrow d_{xy}$	-13.0	-9.0			
$d_{yz} \rightarrow d_{x^2-y^2}$	65.3	58.1			
$d_{xz} \rightarrow d_{x^2-y^2}$	-7.4	-5.4			
$d_{xy} \rightarrow d_{x^2-y^2}$	-13.7	-14.2			
$d_{yz} \rightarrow d_{xz}$	10.5	6.3	11.8		6.0
$d_{yz} \rightarrow d_{xy}$	0.4	4.6	1.3		4.0
$d_{xz} \rightarrow d_{xy}$	-44.0	-37.0	-24.3		-20.0
$d_{xz} \rightarrow d_{yz}$	-0.5	4.6	-1.2		3.0
$d_{xy} \rightarrow d_{yz}$	8.8	6.3	8.3		6.0
$d_{xy} \rightarrow d_{xz}$	-63.2	-58.0	-59.1		-58.0

with $\Delta = 2272.5$ meV, $\Delta_1 = -8.1$ meV, $\Delta_2 = -7.0$ meV, and $\Delta_3 = -3.4$ meV. The orbital angular momenta in the five-orbital model are expressed as

TABLE IV. Hopping parameters (in meV) for the third NN. A and B are the sublattice indices; Z_3 and X_3 bonds are shown in Fig. 1(a). The results from Refs. [41,42,49] are also presented for a comparison.

Bond	T_{ij}				
	C2	P3	C2 [41]	C2 [42]	P3 [49]
Z_3 :					
$d_z \rightarrow d_z$	-26.1	-30.6			
$A \rightarrow B$ $d_{x^2-y^2} \rightarrow d_{x^2-y^2}$	56.9	72.8			
$d_{yz} \rightarrow d_{yz}$	6.6	6.4	-8.2		-8.0
$d_{xz} \rightarrow d_{xz}$	6.6	6.4	-8.2		-8.0
$d_{xy} \rightarrow d_{xy}$	-39.9	-44.2	-39.5		-49.0
$d_z \leftrightarrow d_{x^2-y^2}$	0.0	0.0			
$d_z \leftrightarrow d_{yz}$	-6.8	-6.1			
$d_z \leftrightarrow d_{xz}$	-6.8	-6.1			
$d_z \leftrightarrow d_{xy}$	22.5	26.0			
$d_{x^2-y^2} \leftrightarrow d_{yz}$	4.7	5.7			
$d_{x^2-y^2} \leftrightarrow d_{xz}$	-4.7	-5.7			
$d_{x^2-y^2} \leftrightarrow d_{xy}$	0.0	0.0			
$d_{yz} \leftrightarrow d_{xz}$	-10.6	-7.5	-7.4		-5.0
$d_{yz} \leftrightarrow d_{xy}$	12.4	9.0	11.7		9.0
$d_{xz} \leftrightarrow d_{xy}$	12.4	9.0	11.7		9.0
X_3 :					
$d_z \rightarrow d_z$	35.3	47.0			
$A \rightarrow B$ $d_{x^2-y^2} \rightarrow d_{x^2-y^2}$	-5.1	-4.8			
$d_{yz} \rightarrow d_{yz}$	-39.9	-44.2	-41.4		-49.0
$d_{xz} \rightarrow d_{xz}$	6.3	6.4	-7.9		-8.0
$d_{xy} \rightarrow d_{xy}$	6.4	6.4	-7.5		-8.0
$d_z \leftrightarrow d_{x^2-y^2}$	35.1	44.8			
$d_z \leftrightarrow d_{yz}$	-12.4	-13.0			
$d_z \leftrightarrow d_{xz}$	-0.5	-1.9			
$d_z \leftrightarrow d_{xy}$	7.7	8.0			
$d_{x^2-y^2} \leftrightarrow d_{yz}$	18.3	22.5			
$d_{x^2-y^2} \leftrightarrow d_{xz}$	-8.6	-8.1			
$d_{x^2-y^2} \leftrightarrow d_{xy}$	-3.4	-2.4			
$d_{yz} \leftrightarrow d_{xz}$	13.1	9.0	12.7		9.0
$d_{yz} \leftrightarrow d_{xy}$	12.3	9.0	10.7		9.0
$d_{xz} \leftrightarrow d_{xy}$	-10.6	-7.5	-7.8		-5.0

$$L^x = \begin{pmatrix} 0 & 0 & i\sqrt{3} & 0 & 0 \\ 0 & 0 & i & 0 & 0 \\ -i\sqrt{3} & -i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i \\ 0 & 0 & 0 & -i & 0 \end{pmatrix}, \quad (A3)$$

TABLE V. Hopping parameters (in meV) for the three-orbital model in the $P3$ case. A and B are the sublattice indices; Z_1, Z_2 , and Z_3 bonds are shown in Fig. 1(a).

Bond	T_{ij}		
	d_{yz}	d_{xz}	d_{xy}
Z_1 :			
d_{yz}	58.7	113.9	-7.0
$A \rightarrow B$			
d_{xz}	113.9	58.7	-7.0
d_{xy}	-7.0	-7.0	-194.1
Z_2 :			
d_{yz}	-0.7	-27.6	3.6
$A \rightarrow A$			
d_{xz}	-51.9	-0.7	6.2
d_{xy}	6.2	3.6	1.6
Z_3 :			
d_{yz}	-6.3	-4.8	10.7
$A \rightarrow B$			
d_{xz}	-4.8	-6.3	10.7
d_{xy}	10.7	10.7	-43.9

TABLE VI. Hopping parameters (in meV) for the three-orbital model in the $C2$ case. A and B are sublattice indices; $Z_{1,2,3}$ and $X_{1,2,3}$ bonds are expressed in Fig. 1(a).

Bond		d_{yz}	d_{xz}	\mathcal{T}_{ij}	d_{xy}
Z_1 :	d_{yz}	40.7	161.9		-22.9
$A \rightarrow B$	d_{xz}	161.9	40.7		-22.9
	d_{xy}	-22.9	-22.9		-101.5
X_1 :	d_{yz}	-90.7	-19.0		-17.9
$A \rightarrow B$	d_{xz}	-19.0	39.7		164.9
	d_{xy}	-17.9	164.9		39.5
Z_2 :	d_{yz}	-5.2	-27.1		-1.3
$A \rightarrow A$	d_{xz}	-63.1	-5.2		8.8
	d_{xy}	8.8	-1.3		-1.6
X_2 :	d_{yz}	-1.8	9.4		-0.7
$A \rightarrow A$	d_{xz}	-1.8	-5.2		-26.9
	d_{xy}	8.4	-62.8		-4.5
Z_3 :	d_{yz}	-8.4	-8.5		14.2
$A \rightarrow B$	d_{xz}	-8.5	-8.4		14.2
	d_{xy}	14.2	14.2		-39.5
X_3 :	d_{yz}	-39.6	14.5		13.8
$A \rightarrow B$	d_{xz}	14.5	-8.6		-8.3
	d_{xy}	13.8	-8.3		-8.3

$$L^y = \begin{pmatrix} 0 & 0 & 0 & -i\sqrt{3} & 0 \\ 0 & 0 & 0 & i & 0 \\ 0 & 0 & 0 & 0 & -i \\ i\sqrt{3} & -i & 0 & 0 & 0 \\ 0 & 0 & i & 0 & 0 \end{pmatrix}, \quad (A4)$$

$$L^z = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -2i \\ 0 & 0 & 0 & i & 0 \\ 0 & 0 & -i & 0 & 0 \\ 0 & 2i & 0 & 0 & 0 \end{pmatrix}. \quad (A5)$$

The three-orbital parameters (TB3) for the $P3$ ($C2$) space group are also shown in Table V (Table VI), and are qualitatively consistent with Ref. [49] (Ref. [41]). For the three-orbital model in the $P3$ and $C2$ cases, the crystal fields h_i^Δ are expressed as

$$h_i^\Delta = \begin{pmatrix} 0 & \Delta'_3 & \Delta'_3 \\ \Delta'_3 & 0 & \Delta'_3 \\ \Delta'_3 & \Delta'_3 & 0 \end{pmatrix} \quad (A6)$$

with $\Delta'_3 = -6.6$ meV and

$$h_i^\Delta = \begin{pmatrix} 0 & \Delta_1 & \Delta_2 \\ \Delta_1 & 0 & \Delta_2 \\ \Delta_2 & \Delta_2 & \Delta_3 \end{pmatrix} \quad (A7)$$

with $\Delta_1 = -7.9$ meV, $\Delta_2 = -8.4$ meV, and $\Delta_3 = -3.2$ meV, respectively. The orbital angular momenta in the three-orbital model are expressed as

$$L^x = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & i \\ 0 & -i & 0 \end{pmatrix}, \quad (A8)$$

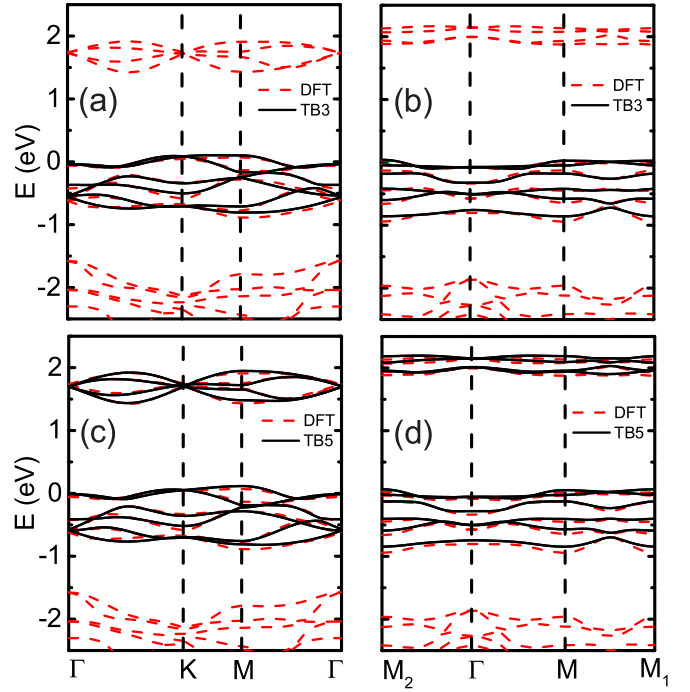


FIG. 6. Band structure of α -RuCl₃ monolayer. The dashed red lines show the result from DFT without SOC. The tight-binding bands from (a) the three-orbital model in the $P3$ case, (b) the three-orbital model in the $C2$ case, (c) the five-orbital model in the $P3$ case, and (d) the five-orbital model in the $C2$ case are denoted by the black solid lines. For the $C2$ case, the high-symmetry points M_1 and M_2 are midpoints of reciprocal-lattice vectors.

$$L'^y = \begin{pmatrix} 0 & 0 & -i \\ 0 & 0 & 0 \\ i & 0 & 0 \end{pmatrix}, \quad (A9)$$

$$L'^z = \begin{pmatrix} 0 & i & 0 \\ -i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (A10)$$

Based on the tight-binding fits, the band structures (black solid) without SOC are shown in Fig. 6. The red dashed lines in Fig. 6 are the band structures from the DFT calculation.

APPENDIX B: PARAMETERS OF THE $J_{\text{eff}} = 1/2$ EFFECTIVE ISOSPIN MODEL IN Eq. (10)

The exchange interaction parameters in Eq. (10) derived from the five-orbital Hubbard model are calculated based on the tight-binding fit to the DFT calculations and with interactions $U = 2.31$ eV, $J_H = 0.32$ eV, and $\lambda = 0.14$ eV. The results are (in meV) $J^\gamma = 0.10$, $K^\gamma = -3.35$, $\Gamma^\gamma = 7.62$, $\Gamma'^\gamma = -0.45$, $J_2^\gamma = -0.37$, $K_2^\gamma = 0.73$, and $K_3^\gamma = 0.42$ ($J^z = -0.40$, $J^{x,y} = -0.23$, $K^z = -10.52$, $K^{x,y} = -10.63$, $\Gamma^z = 5.07$, $\Gamma^{x,y} = 4.75$, $\Gamma'^z = -1.12$, $\Gamma'^{x,y} = -1.14$, $J_2^z = -0.31$, $J_2^{x,y} = -0.31$, $K_2^z = 0.31$, $K_2^{x,y} = 0.31$, $K_3^z = 0.31$, and $K_3^{x,y} = 0.31$). This set of parameters is used to plot the red dashed lines in Fig. 4 in the main text.

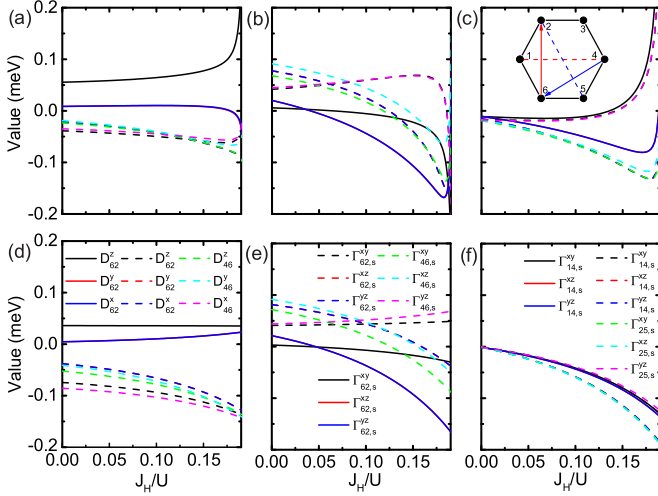


FIG. 7. Dependence of the second- and third-NN magnetic interactions on the Hund's coupling J_H/U . (a) and (d) The second-NN DM interactions. (b) and (e) The second-NN symmetrical off-diagonal interactions. (c) and (f) The third-NN symmetrical off-diagonal interactions. $\Delta = 2.10$ eV, $U = 2.31$ eV, and $\lambda = 0.14$ eV are used in (a), (b), and (c). $\Delta = 210$ eV, $U = 2.31$ eV, and $\lambda = 0.14$ eV are used in (d), (e), and (f). Solid (dash) lines denote the P3 (C2) case. The subscripts of the magnetic interactions denote the sites labeled by the numbers 1 to 6, as shown in the inset of (c). The red and blue solid (dashed) lines in the inset represent the second- (third-) NN Z and X bonds, respectively.

In Eq. (10), we have neglected some terms in the second- and third-NN exchange interactions, which are found to be much smaller than the NN exchange interactions. Here, we show the J_H dependence of the second- and third-NN exchange interactions neglected in Eq. (10) in Fig. 7. Because Hund's coupling J_H has the most obvious effect on the exchange interactions as already seen from Fig. 2, only the J_H dependence is discussed here.

In Fig. 7, the Dzyaloshinskii-Moriya (DM) interaction $D_{ij} = (D_{ij}^x, D_{ij}^y, D_{ij}^z)$ and off-diagonal Γ terms $\Gamma_{ij,s}^{\alpha\beta}$ are shown, which are defined as $D_{ij}^\alpha = (J_{ij}^{\beta\gamma} - J_{ij}^{\gamma\beta})/2$ and $\Gamma_{ij,s}^{\alpha\beta} = (J_{ij}^{\alpha\beta} + J_{ij}^{\beta\alpha})/2$, respectively. The indices i and j are illustrated in the inset of Fig. 7(c). Comparing Figs. 7 and 2, we find that the magnitudes of the DM interactions and off-diagonal Γ term for the second and third NN are much smaller than those of the first-NN exchange interactions. The main reason is that the second- and third-NN hopping integrals are much smaller than those for the first NN. Another reason for the smallness of the DM interactions is that the e_g - t_{2g} mixing

also decreases these interactions on second-NN bonds. If we deliberately increase the crystal field to be the unrealistic value $\Delta = 210$ eV, which reduces the mixing of the e_g and t_{2g} orbitals, the magnitudes of the DM exchange interactions are enhanced as shown in Figs. 7(d) and 7(e). Figures 7(c) and 7(f) show that the e_g - t_{2g} mixing also reduces some third-NN off-diagonal Γ interactions.

APPENDIX C: SPIN-WAVE HAMILTONIAN OF THE MINIMAL MODEL IN EQ. (11)

Here, we show the spin-wave Hamiltonian of the minimal model in Eq. (11) by employing the linear spin-wave theory [4,45] for the zigzag phase. In the zigzag phase, we choose the magnetic unit cell $a \times b$ with $a = 3\hat{a}_0$ and $b = \sqrt{3}\hat{a}_0$, where \hat{a}_0 is the length of the NN bond. For the zigzag order, there are only two degrees of freedom in the magnetic unit cell, i.e., two local rotation parameters (θ, ϕ) . Then the zeroth-order Hamiltonian in the magnetic unit cell is obtained as

$$\begin{aligned} \mathcal{H}_0(\theta, \phi) = & \frac{1}{2} \{ -K^z \cos^2(\theta) + \Gamma^{x,y} \sin(2\theta) [\cos(\phi) + \sin(\phi)] \\ & + \sin^2(\theta) [K^{x,y} \sin^2(\phi) \\ & - \Gamma^z \sin(2\phi) + K^{x,y} \cos^2(\phi)] \}. \end{aligned} \quad (C1)$$

By minimizing the zeroth-order Hamiltonian, we find the rotation parameter ϕ is equal to $\pi/4$ and θ satisfies $\theta = 1/2 \tan^{-1} [-2\sqrt{2}\Gamma^{x,y}/(K^{x,y} + K^z - \Gamma^z)] + \pi/2$. Thus the quadratic Hamiltonian becomes

$$\mathcal{H}_2 = X^\dagger H(\mathbf{q}) X, \quad (C2)$$

where $X^\dagger = (\tilde{b}_{1,q}^\dagger, \tilde{b}_{2,q}^\dagger, \tilde{b}_{3,q}^\dagger, \tilde{b}_{4,q}^\dagger, \tilde{b}_{1,-q}^\dagger, \tilde{b}_{2,-q}^\dagger, \tilde{b}_{3,-q}^\dagger, \tilde{b}_{4,-q}^\dagger)$ and the number i in the subscript of $\tilde{b}_{i,q}^\dagger$ represents the lattice site in the magnetic unit cell (see Fig. 1). The matrix $H(\mathbf{q})$ is given by

$$H(\mathbf{q}) = \begin{pmatrix} A & C_1^* & 0 & B & 0 & C^* & 0 & D \\ C_1^* & A & B^* & 0 & C & 0 & D^* & 0 \\ 0 & B & A & C_1^* & 0 & D_1 & 0 & C^* \\ B^* & 0 & C_1 & A & D_1^* & 0 & C & 0 \\ 0 & C^* & 0 & D_1 & A & C_1^* & 0 & B \\ C & 0 & D_1^* & 0 & C_1 & A & B^* & 0 \\ 0 & D & 0 & C^* & 0 & B & A & C_1^* \\ D^* & 0 & C & 0 & B^* & 0 & C_1 & A \end{pmatrix} \quad (C3)$$

where

$$\begin{aligned} A &= \frac{1}{2} [K^z \cos^2(\theta) - \sqrt{2}\Gamma^{x,y} \sin(2\theta) + (\Gamma^z - K^{x,y}) \sin^2(\theta)], \\ B &= \frac{1}{8} \eta \cos\left(\frac{\sqrt{3}q_b}{2}\right) [3K^{x,y} + K^{x,y} \cos(2\theta) - 2\sqrt{2}\Gamma^{x,y} \sin(2\theta)], \\ C &= \frac{1}{4} \eta^2 (-\Gamma^z + K^z) \sin^2(\theta), \\ C_1 &= \frac{1}{4} \eta^2 [\Gamma^z + \Gamma^z \cos^2(\theta) + K^z \sin^2(\theta)], \end{aligned}$$

$$D = -\frac{1}{4}\eta \left\{ 2 \sin \left(\frac{\sqrt{3}q_b}{2} \right) [K^{x,y} \cos(\theta) + \sqrt{2}\Gamma^{x,y} \sin(\theta)] + \cos \left(\frac{\sqrt{3}q_b}{2} \right) \sin(\theta) [2\sqrt{2}\Gamma^{x,y} \cos(\theta) + K^{x,y} \sin(\theta)] \right\},$$

$$D_1 = \frac{1}{4}\eta \left\{ 2 \sin \left(\frac{\sqrt{3}q_b}{2} \right) [K^{x,y} \cos(\theta) + \sqrt{2}\Gamma^{x,y} \sin(\theta)] - \cos \left(\frac{\sqrt{3}q_b}{2} \right) \sin(\theta) [2\sqrt{2}\Gamma^{x,y} \cos(\theta) + K^{x,y} \sin(\theta)] \right\}, \eta = e^{-i\frac{q_a}{2}}.$$
(C4)

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