Excitonic Magnetism in Van Vleck-type $d^4$ Mott Insulators

Giniyat Khaliullin

$^1$Max Planck Institute for Solid State Research, Heisenbergstrasse 1, D-70569 Stuttgart, Germany

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In Mott insulators with the $t_{2g}^2$ electronic configuration such as of Re$^{3+}$, Ru$^{4+}$, Os$^{4+}$, Ir$^{5+}$ ions, spin-orbit coupling dictates a Van Vleck-type nonmagnetic ground state with angular momentum $J = 0$, and the magnetic response is governed by gapped singlet-triplet excitations. We derive the exchange interactions between these excitons and study their collective behavior on different lattices. In perovskites, a conventional Bose condensation of excitons into a magnetic state is found, while an unexpected one-dimensional behavior supporting spin-liquid states emerges in honeycomb lattices, due to the bond-directional nature of exciton interactions in the case of 90$^\circ$ $d$-$p$-$d$ bonding geometry.

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Many transition metal (TM) compounds fall into a category of Mott insulators where strong correlations suppress low-energy charge dynamics, but there remains rich physics due to unquenched spin and orbital magnetic moments that operate at energies below the charge (Mott) gap. Depending on spin-orbital structure of constituent ions and the nature of the chemical bonding of neighboring $d$ orbitals, the TM oxides host a great variety of magnetic phenomena ranging from classical orderings to quantum spin and orbital liquids.

In broad terms, the magnetism of localized electrons in Mott insulators is governed by several factors: intraionic Hund’s rules that form local spin $S$ and orbital $L$ moments; spin-orbit coupling (SOC) that tends to bind them into a total angular momentum $J = S + L$; crystal fields which split $d$ levels and suppress the $L$ moment, acting thereby against SOC; and, finally, inter-site superexchange (SE) interactions which establish a long-range coherence between spins and orbitals.

In Mott insulators with $d$ orbitals of $e_g$ symmetry like manganites and cuprates, $L$ moment is fully quenched in the ground state (GS), and one is left with spin-only magnetism. In contrast, TM ions with threefold $t_{2g}$ orbital degeneracy possess an effective orbital angular momentum $L = 1$, and a complex interplay between unquenched SOC and SE interactions emerges.

In TM oxides with odd number of electrons on the $d$ shell, $S$ and $J$ are half-integer; hence, the ionic GS is Kramers degenerate and magnetically active. The main effect of SOC in this case is to convert the original exchange interactions among $S$ and $L$ moments into an effective $J$-Hamiltonian operating within the lowest Kramers $J$-manifold. The $t_{2g}$ orbital $L$-interactions are bond dependent and highly frustrated; consequently, the $J$-Hamiltonians inherit this property, too. In short, SOC replaces $S$ and $L$ moments by $J$ that obeys the same spin-commutation rules, but resulting magnetic states may obtain a nontrivial structure, as found for $d^2(J = 1/2)$ [13, 14] and $d^3(J = 3/2)$ [11, 12] compounds. Similar SOC effects can be realized also in non-Kramers $d^2$ oxides [13, 14] with $J = 2$.

A conceptually different situation can be encountered in Mott insulators with TM ions of Van Vleck-type, i.e. when SOC imposes nonmagnetic GS with $J = 0$ and magnetism is entirely due to virtual transitions to higher levels with finite $J$. Such “nonmagnetic” Mott insulators are natural for $4d$ and $5d$ TM ions with $t_{2g}$ configuration, e.g., Re$^{3+}$, Ru$^{4+}$, Os$^{4+}$, and Ir$^{5+}$. These ions realize low-spin $S = 1$ state because of moderate Hund’s coupling $J_H$ (compared to 10$Dq$ octahedral splitting), and, at the same time, SOC $\lambda(S \cdot L)$ is strong enough to stabilize $J = 0$ state gaining energy $\lambda$ relative to the excited $J = 1$ triplet. Since the singlet-triplet splitting for these ions $\lambda \sim 50–200$ meV [13, 16] is comparable to SE energy scales $4t^2/U \sim 50–100$ meV, we may expect magnetic condensation of Van Vleck excitons. This brings us to the “singlet-triplet” physics widely discussed in the literature in various contexts: magnon condensation in quantum dimer models [17, 21], bilayer magnets [22], excitons in rare-earth filled-skutterudites [23, 25], a curious case of $e_g$ orbital FeSc$_2$S$_4$ [26], spin-state transition in Fe-pnictides [27], etc. The underlying physics and, hence, the energy scales involved in the present case are of course different from the above examples.

In this Letter, we develop a microscopic theory of magnetism for Van Vleck-type $d^4$ Mott insulators. First, we derive $S$ and $L$ based SE Hamiltonian and map it onto a singlet-triplet low-energy Hilbert space. We then show, taking perovskite lattices as an example, how an excitonic magnetic order, magnons and the amplitude (“Higgs”) modes do emerge in the model. Considering the model on a honeycomb lattice, we reveal the emergent one-dimensional dynamics of Van Vleck excitons, and discuss possible implications of this observation.

The spin-orbital superexchange.--- Kugel-Khomskii type interactions between $t_{2g}^4$ ions are derived in a standard way, by integrating out oxygen-mediated $d$-$p$-$d$ electron hoppings. We label $d_{yz}, d_{zx}, d_{xy}$ orbitals by $a, b, c$, respectively. In 180$^\circ$ (90$^\circ$) $d$-$p$-$d$ bonding geometry corresponding to corner-shared (edge-shared) octahedra, two
orbitals are active on a given bond (see Fig. 2 of Ref. [3]),
while the third one, say, $\gamma$, is not; accordingly, this
bond is denoted by $\gamma$. Then, nearest-neighbor hops-
pings on the $c$ bond read as $t(a_i^\dagger a_j + b_i^\dagger b_j + h.c.)$ for
180° and $t(a_j b_i + b_i a_j + h.c.)$ for 90° geometries.
In calculations, it is helpful to introduce $A, B, C$ operators
that represent three different orbital configurations
$A = \{a^2 b c\}$, $B = \{a b^2 c\}$, $C = \{a b c^2\}$ of the $t_{2g}$ shell and
its effective $L = 1$ momentum $L_x = -i(B^L - C^L)$,
etc., similar to the $d^1$ case [2]. There is one-to-one cor-
respondence $(A, B, C) \leftrightarrow (a, b, c)$ and $L^{\alpha} \leftrightarrow I^\alpha$ between
d$^1$ and $d^1$ orbital configurations.

The resulting spin-orbital Hamiltonian reads as $H = \sum_{i < ij}[(S_i \cdot S_j + 1) O^{(j)}_i + (L_i^x)^2 + (L_i^y)^2]$, where $U \gg$
t, $J$, $J_H$ is Hubbard repulsion. The bond-dependent or-
bital operator $O^{(\gamma)}$ depends on the above $A, B, C$; we
show it directly in terms of $L$:

$$O^{(c)}_{ij} = (L_i^x T_j^x)^2 + (L_i^y T_j^y)^2 + (L_i^z T_j^z)^2 + (L_i^x T_j^x L_i^y T_j^y + L_i^y T_j^y L_i^z T_j^z + L_i^z T_j^z L_i^x T_j^x),$$ (1)

This result holds for 180° bondage. For 90° geometry,
one has simply to interchange $L_i^z \leftrightarrow L_j^z$; this can be
traced back to the $a_j \leftrightarrow b_j$ relation between 180° and 90°
hoppings given above. Operators $O^{(a)}$ and $O^{(b)}$ for $a, b$
bonds follow from cyclic permutations among $L_x, L_y, L_z$.

The above model $H$ operates within $|M_S, M_L>$ basis.
We project it onto the low-energy subspace spanned by the
GS $J = 0$ singlet $|0\rangle = \frac{1}{\sqrt{3}}((1, -1) - (0, 0) + | - 1, 1)$, and $J = 1$ triplet $|T_0\rangle = \frac{1}{\sqrt{2}}((1, -1) - | - 1, 1)$,$
|T_{\pm 1}\rangle = \frac{1}{\sqrt{2}}((1, 1) - | - 1, -1)$ at energy $E_T = \lambda$, as
ddictated by local SOC. (The high-energy $J = 2$ level
at 3$\lambda$ is neglected.) Calculating matrix elements of $S^\alpha, L^\alpha$, and their combinations within this Hilbert space,
we represent them in terms of hard-core “triplon” $T^\alpha$ with
the Cartesian components $T_{x, y, z} = \frac{1}{\sqrt{2}}(T_i + T_{-i}), T_0 = \frac{i}{\sqrt{2}}(T_i + T_{-i}), T_z = iT_0$, and “spin” $J = -i(T^1 \times T^\alpha)$.
For instance, $S = -i\sqrt{2}(T - T^1) + \frac{2}{3}J$, $L = i\sqrt{2}(T - T^1) + \frac{2}{3}J$. A projection $H(S, L) \rightarrow H(T, J)$ results in
the effective singlet-triplet models $H_{eff}(180^\circ)$ and $H_{eff}(90^\circ)$
discussed below.

In terms of $T$ and $J$, magnetic moment of a $t_{2g}^1$ shell
$M = 2S - L$ reads as $M = -i\sqrt{6}(T - T^1) + g_s J$ with
g$J = 1/2$, or $M = 2\sqrt{6} u + g_s J$, introducing real fields
$u$ and $v$ as $T = u + iv$ with $u^2 + v^2 \leq 1$ [28]. The
two-component structure of $M$ highlights physical distinc-
tion between conventional Mott insulators where $M$ is
simply $g_s J$ with finite $J$ in the GS, and the present
case where the magnetic moment resides predominantly
on singlet-triplet Van Vleck transitions represented by
$T$ exciton (hence the term “excitonic magnetism”). On
formal side, these two components obey different commuta-
tion rules, hard-core boson $T$ vs spin $J$; consequently,
magnetic order is realized here as Bose condensation of $T$
particles, instead of the usual freezing of the prexisting
$J$ moments. The above equations for $S$ and $L$ make it
also clear that $T$ condensation implies a condensation of $S$
and $L$ moments resulting in finite $M$, while the sum
$S + L = J$ may still fluctuate [24]. As in singlet-triplet
models in general, the magnetic exciton condensation in
$\xi_{2g}$ Van Vleck models requires a critical exchange cou-
pling $t^2/|U|$, so there will be magnetic order-disorder criti-
cal point that can be tuned by pressure, doping, etc.

Singlet-triplet model $H_{eff}(180^\circ)$. – This case applies to
perovskites like $ABO_3$ or $A_2BO_4$ with corner-shared $BO_6$
ocathedra (e.g., $Ca_2RuO_4$). We shape the model in the
form of $H_{eff} = \lambda \sum_i n_i + \frac{\xi}{|U|} \sum_{i < ij} (h_2 + h_3 + h_4)_{ij}$,
where $h_2$ term is quadratic in $T$ bosons, while $h_3$ and $h_4$
represent three- and four-boson interactions [30]. For the
$\gamma = c$ bond,

$$h_2^{(c)} = \frac{11}{3} v_i \cdot v_j - v_i v_j \frac{2}{3} (u_i \cdot u_j - u_i u_j),$$ (2)

$$h_3^{(c)} = \frac{1}{24} (v_i \cdot J_j + v_i - v_j) + \frac{1}{4} (J_i J_j + J_j J_i) - \frac{5}{36} n_i n_j,$$

$$h_4^{(c)} = \frac{3}{4} d_{ij} d_{ij} + \frac{1}{2} J_i J_j + \frac{1}{4} (J_i J_j + J_j J_i) - \frac{5}{36} n_i n_j.$$

$h^{(c)}$ for $\gamma = a, b$ follow from permutations among $x, y, z$.
$n = \sum_i T_i^3 T_i$, while $Q_{z < T_i^3 T_i + T_i^3 T_i}$, etc.,
are quadrupole operators of $T_{2g}$ symmetry [31]. As expected,
h$_4$ contains a biquadratic Heisenberg coupling; we show
it here via bond-singlet operator $d_{ij} = \frac{1}{\sqrt{2}}(T_i^3 T_j^3)$ using
the identity $(J_i, J_j)^2 = 3d_{ij} d_{ij} + n_i n_j$.

We quantify exchange interaction by $\kappa = 4t^2/|U|$. On
a cubic (square) lattice, the model undergoes a magnetic
phase transition at $\kappa_c \approx \frac{2}{5} \lambda$ ({$\kappa_c \approx \frac{4}{5} \lambda$}), due to conden-
sation of a dipolar $\upsilon$ part of the $T$ bosons. The den-
sity of out-of-condensate $T$ particles and, hence, $J$ and
$Q_{z < T}$ are very small near critical $\kappa$, e.g., $\langle J_i J_j \rangle \sim 1/8z$
with $z = 6(4)$ for a cubic (square) lattice; thus, the in-
hertions $h_{3,4}$ are not of a qualitative importance for the
180° case, and we focus on a quadratic part $H_2$ of
$H_{eff}$. Also, bond-dependent terms in $h_2$ are weak and
unessential in 180° geometry, so we may average them out:
$v_i v_j v_j \rightarrow v_i v_j / 3$ for simplicity [32]. The resulting
hard-core boson Hamiltonian

$$H_2 = \lambda \sum_i n_i + \kappa \frac{5}{9} \sum_{i < j} (T_i, T_j) - \frac{7}{16} (T_i, T_j + H.c.)]$$ (3)

is treated in a standard way familiar from “singlet-triplet
model” literature (see, e.g., Refs. [17, 22]).

In a paramagnetic phase, $\kappa < \kappa_c$, magnetic exicita-
tions are degenerate, and their dispersion $\omega_{z < y / z}(k) =
\lambda \sqrt{1 + (\kappa / \kappa_c) \phi(k)}$ with $\phi(k) = \frac{1}{2} \sum \cos(\kappa c)$ has a finite gap
$\lambda \sqrt{1 - (\kappa / \kappa_c)}$. At $\kappa = \kappa_c$, the gap closes and, say, $T_z$
boson condenses to give a finite staggered magnetization
$M_z = 2\sqrt{6} \rho(1 - \rho)$ at $\kappa > \kappa_c$, where $\rho = \frac{1}{2} (1 - \tau^{-1})$ is
the condensate density expressed via dimensionless pa-
ter parameter $\tau = \kappa / \kappa_c > 1$. The $M$-length fluctuations, i.e.,
the amplitude "Higgs" mode, has a dispersion \( \omega_z(k) \approx \lambda \sqrt{\Delta^2 + \phi_k^2} \) with the gap \( \Delta = \lambda \sqrt{\tau^2 - 1} \), while \( T_{x/y} \) excitons become gapless Goldstone magnons with the energy \( \omega_{x/y}(k) \approx \lambda \frac{\phi}{\sqrt{1 + \phi_k^2}} \).

We are ready to show our theory in action, by applying it to \( d^4 \) Mott insulator \( \text{Ca}_2\text{RuO}_4 \) \([34]\) where a sizable value of the \( LS \) product has indeed been observed \([35]\). This fact implies the presence of unquenched spin-orbit coupling which is the basic input of our model.

First, we compare the observed staggered moment \( M \approx 1.3 \mu_B \) \([36]\) with our result \( M = \sqrt{6(1 - \tau^2) - \mu_B} \), and find \( \tau \approx 1.18 \), i.e., this compound is rather close to the magnetic critical point. For spin-orbit coupling \( \lambda(\xi/2) \approx 75 \text{ meV} \) \([33]\), this translates into \( \frac{4\lambda^2}{\mu_B^2} \approx 53 \text{ meV} \), a reasonable value for \( t_{2g} \) systems with \( t \sim 0.2 \text{ eV} \) and \( U \sim 3 - 4 \text{ eV} \).

Second, using spin and orbital moments in the condensate \( S = -L = \frac{1}{\sqrt{3\mu_B}}M \), we estimate their product \( LS \approx -0.2 \) which is not too far from \(-0.28 \pm 0.07 \) observed \([33]\).

Third, we obtain from our theory the uniform magnetic susceptibility \( \chi = \frac{\mu_B^2 \lambda^2}{2(1 + \lambda^2)} \approx 2.3 \times 10^{-5} \text{ emu/mol} \), which is consistent with that of \( \text{Ca}_2\text{RuO}_4 \) (\( \sim 2.5 \times 10^{-3} \text{ emu/mol} \) \([31, 36]\)) above Néel temperature, where it is only weakly temperature dependent as expected for Van Vleck-type systems.

With the above numbers at hand (in fact, all extracted from the data), we predict the amplitude-mode gap \( \Delta \sim 45 \text{ meV} \), and the topmost energies \( \sim 115 \text{ meV} \) for all three magnetic modes. We are not aware of inelastic magnetic data for \( \text{Ca}_2\text{RuO}_4 \) to date; resonant x-ray or neutron scattering experiments would provide an crucial test for the theory.

We now turn to compounds with \( 90^\circ \) \( d-p-d \) bonding geometry, where effective interactions lead to remarkable features not present in perovskites.

**Singlet-triplet model \( H_{\text{eff}}(90^\circ) \).** This case is relevant to delafossite \( \text{ABO}_2 \) or \( \text{A}_2\text{BO}_4 \) structures where \( \text{BO}_6 \) octahedra share the edges and TM-ions form triangular or honeycomb lattices (e.g., \( \text{Li}_2\text{RuO}_3 \)). Using the same notations as above, we find

\[
h_2^{(c)} = 3(v_i \cdot v_j - u_i u_j) - \frac{1}{3}(u_i \cdot u_j - u_i u_{jz}),
\]

\[
= \frac{3}{2}(T_{x}T_{x} + T_{y}T_{y}) - \frac{5}{6}(T_{x}T_{z} + T_{y}T_{y}) + H.c.,
\]

\[
h_3^{(c)} = \frac{1}{\sqrt{21}}(3v_{i} \cdot J_{j} + 3v_{iz}J_{jz} - u_{iz}Q_{jz} + u_{iy}Q_{yj} + (i \leftrightarrow j),
\]

\[
h_4^{(c)} = -\frac{3}{4}d_{ij}d_{ij} + \frac{1}{4}(J_{iz}J_{iz} + Q_{iz}Q_{iz}) + \frac{1}{6}(n_{i}n_{j} + n_{iz}n_{jz})
\]

\[
- \frac{1}{12}n_{i}n_{jz}.
\]

Again, \( h^{(\gamma)} \) for \( \gamma = a, b \) follow from \( x, y, z \)-cyclic permutations. While bond-dependent nature of \( h^{(\gamma)} \) is expected for SOC models on general grounds \([3]\), it is surpris-
Three types of bonds on triangular or honeycomb lattices; (a) Three types of T excitations. (b) Three types of bonds on triangular or honeycomb lattices; $xy + yy$ indicates that only $T_x$ and $T_y$ bosons can move along $c$ bonds. (c) On a honeycomb lattice, each type of exciton forms its own zigzag chain, e.g., $z$ path for $T_z$. (d) In momentum space, each $T_i$ boson softens and forms a quasicondensate at the respective edges of the Brillouin zone where $\omega_\gamma = 0$.\cite{17}.

FIG. 1: (color online). Schematic of $T$-exciton dynamics in 90°-bonding geometry. (a) Three types of $T$ excitations. (b) Three types of bonds on triangular or honeycomb lattices; $xx + yy$ indicates that only $T_x$ and $T_y$ bosons can move along $c$ bonds. (c) On a honeycomb lattice, each type of exciton forms its own zigzag chain, e.g., $z$ path for $T_z$. (d) In momentum space, each $T_i$ boson softens and forms a quasicondensate at the respective edges of the Brillouin zone where $\omega_\gamma = 0$.\cite{17}.

as well as numerical methods; this goes beyond the scope of the present work. We may, however, indicate potential instabilities and possible scenarios.

The Hamiltonian (4) possesses a threefold symmetry (originating from $t_{2g}$ orbital degeneracy): $C_3$ rotation of the lattice and permutation among $T_x$, $T_y$, $T_z$ flavors. This discrete symmetry can be broken at finite temperature. One may expect at least three distinct ground states as a function of $\kappa$: a trivial paramagnet below $\kappa_{c1} \lesssim \frac{1}{4} \lambda$; a long-range magnetic order when boson density becomes large at $\kappa_{c2} > \frac{1}{3} \lambda$; and an intermediate phase at $\kappa_{c1} < \kappa < \kappa_{c2}$ hosting the spin singlet GS. The most intriguing option for the latter is a spin-superfluid state, often discussed in the context of spin-one bosons\cite{31} and bilinear-biquadratic spin models\cite{32}.

Here, this state is supported by a flavor-symmetric attraction $-d_{ij}d_{ij}$ between $T_x$, selecting a global spin singlet of $A_{1g}$ symmetry where all three flavors form pairs and condense, but there is a single-particle gap. Another possibility, favored by the bond-directional nature of hoppings and interactions in Eq. (4), is a nematic order, i.e. spontaneous selection of a particular zigzag out of three $x$, $y$, $z$ directions (assisted in real systems by electron-lattice coupling). Once zigzag chain is formed, it can dimerize due to biquadratic interactions\cite{33, 41, 44}. In other words, boson pairs condense into a valence-bond-solid pattern, followed by a suppression of Van Vleck susceptibility. Future studies are necessary to clarify the phase behavior of the model near the magnetic critical point.

In the recent past, some unusual properties of ruthenate compounds have been reported, including the formation of one-dimensional, spin-gapped chains in $\text{Ti}_2\text{Ru}_2\text{O}_7$\cite{45}, and singlet dimers in $\text{La}_2\text{Ru}_2\text{O}_{10}$\cite{46, 47}. Of particular interest is a honeycomb lattice $\text{La}_2\text{Ru}_3\text{O}_5$\cite{48} which forms dimerized zigzag chains. This observation has been discussed in terms of orbital ordering\cite{49}; the present model based on spin-orbit coupling may provide an alternative way. Indeed, $z$-type zigzag chain dimerized by biquadratic exchange (and supported by electron-lattice coupling) would give the same pattern as observed\cite{48}. Future experiments, in particular a direct measurement of the $LS$ product, should tell whether an unquenched $L$ moment (the key ingredient of our model) is present in these compounds, in order to put the above ideas on a more solid ground.

On theory side, apart from low-energy properties of the model itself, important questions are related to doping of Van Vleck-type $d^x$ insulators. A doped electron, i.e. $J = 1/2$ fermion moving on a background of singlet-triplet $d^x$ lattice, should have a large impact on magnetism and vice versa. Unconventional pairing via the exchange of $T$ excitons also deserves attention, in particular, on triangular and honeycomb lattices (where unusual pairing symmetries have been suggested for $d^5$ systems with strong SOC\cite{3, 50–52}). We recall that energy scales involved in $d^x$ systems are large even for 90° $d-p-d$ bonding, so all the ordering phenomena are expected at higher temperatures than in $d^5$ compounds like triangular lattice $\text{Na}_x\text{CoO}_2$ or honeycomb $\text{Na}_2\text{IrO}_3$.

To conclude, unconventional magnetism emerging from exciton condensation, rather than from orientation of the preexisting local moments, can be realized in Mott insulators of Van Vleck-type ions with a nonmagnetic ground state. We derived effective models describing the magnetic condensate and its elementary excitations on various lattices. Of particular interest is the emergence of quasi-one-dimensional condensate of magnetic excitons on a honeycomb lattice. We discussed implications of the theory for candidate Van Vleck-type Mott insulators.

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The \((u,v)\) representation is often more convenient, in particular for semiclassical analysis. Physically, \(v\) takes care of a magnetic dipole carried by the \(T\) exciton (and hence enters in \(S L\), \(L\), and \(M\)), while \(u\) stands for the quadrupolar component of \(T\).

In the \((u,v)\) representation, \(J = 2(u \times v)\). We will see that \(v\) condenses at the critical point but \(u\) remains fluctuating.

We neglect here a small correction to singlet-triplet splitting \(\lambda\) arising from single-ion energy shifts; for a cubic lattice, the renormalization is \(\lambda \rightarrow \lambda - \frac{\pi}{6}\), and even smaller for the low-coordination square and honeycomb lattices.

Quadrupoles \(Q\) arise from the mapping of composite spin-orbital terms \(S^\alpha L^\beta\) in \(H\), e.g., \(S^y L^z = \sqrt{\frac{2}{3}} u_x + \frac{1}{2} Q_x\). Note that \(u\) enters here, revealing its quadrupole nature.

The only effect of small "compass-like" \(v_\alpha v_\gamma\) terms here is to select an easy magnetic axis and open (order-by-disorder) magnon gap that can be routinely worked out \([3]\).

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We may reverse the sign of \(\kappa\) in Eq. (5) by virtue of the four-sublattice transformation \([3]\). Then, zero-energy lines would go through the \(\Gamma\) point, e.g., \(\omega_\nu(k) = 0\) at \(k_y = 0\).

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In the \((u,v)\) representation, \(J = 2(u \times v)\). We will see that \(v\) condenses at the critical point but \(u\) remains fluctuating.

We neglect here a small correction to singlet-triplet splitting \(\lambda\) arising from single-ion energy shifts; for a cubic lattice, the renormalization is \(\lambda \rightarrow \lambda - \frac{\pi}{6}\), and even smaller for the low-coordination square and honeycomb lattices.

Quadrupoles \(Q\) arise from the mapping of composite spin-orbital terms \(S^\alpha L^\beta\) in \(H\), e.g., \(S^y L^z = \sqrt{\frac{2}{3}} u_x + \frac{1}{2} Q_x\). Note that \(u\) enters here, revealing its quadrupole nature.

The only effect of small "compass-like" \(v_\alpha v_\gamma\) terms here is to select an easy magnetic axis and open (order-by-disorder) magnon gap that can be routinely worked out \([3]\).

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We may reverse the sign of \(\kappa\) in Eq. (5) by virtue of the four-sublattice transformation \([3]\). Then, zero-energy lines would go through the \(\Gamma\) point, e.g., \(\omega_\nu(k) = 0\) at \(k_y = 0\).

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