Damped orbital excitations in the titanates

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A possible mechanism for the removal of the orbital degeneracy in RTiO\textsubscript{3} (where R=La, Y, ...) is considered. The calculation is based on the Kugel-Khomskii Hamiltonian for electrons residing in the $t_{2\text{g}}$ orbitals of the Ti ions, and uses a self-consistent perturbation expansion in the interaction between the orbital and the spin degrees of freedom. The latter are assumed to be ordered in a Neél state, brought about by delicate interactions that are not included in the Kugel-Khomskii Hamiltonian. Within our model calculations, each of the $t_{2\text{g}}$ bands is found to acquire a finite, temperature-dependent dispersion, that lifts the orbital degeneracy. The orbital excitations are found to be heavily damped over a rather wide band. Consequently, they do not participate as a separate branch of excitations in the low-temperature thermodynamics.

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I. INTRODUCTION

The Mott insulator lanthanum titanate (LaTiO\textsubscript{3}) is one of the most puzzling materials in the family of transition metals (TM) of the perovskite structure (see, e.g., Ref. 1). Each of the Ti ions in this material has a single valent electron in one of the three-fold degenerate $t_{2\text{g}}$ orbitals, causing the ground state to be enormously degenerate. Whereas in the majority of the TM perovskites the conventional Jahn-Teller distortion lifts the orbital degeneracy, such distortions are believed to be small in the lanthanum titanate, and will be assumed to be zero for the purposes of the present paper. Here we pose the following question: is there an alternative mechanism to the Jahn-Teller one, which is capable of removing the orbital degeneracy?

This question has already come up in materials with doubly-degenerate $e_{\text{g}}$ valent electrons. In these materials, there exists an \textit{orbitally ordered phase}, with coherent excitations (‘orbitons’). These excitations were recently observed in LaMnO\textsubscript{3}. The notion of an orbital disordered, \textit{liquid}, state was developed in Ref. 5 for the hole doped LaMnO\textsubscript{3} with a high hole concentration, where the antiferromagnetic (AF) insulating state gives way to a ferromagnetic metallic phase. Theoretically, the orbitons in doped materials are constructed similarly to spinons, which appear within the framework of the $t-J$ model for the hole-doped cuprates. Another scenario of quantum liquid formation was proposed for the insulating KCuF\textsubscript{3}. According to Ref. 7, the orbital fluctuations (random changes of the bond directions) may be the source for the “quantum melting” of the magnetic order, because of the strong interaction between orbital and spin degrees of freedom. Melting, in this scenario, means the disappearance of a long-range AF order in favor of a \textit{spin liquid} state.

The lifting of the degeneracy of the $t_{2\text{g}}$ orbital triplet in LaTiO\textsubscript{3} is a more difficult problem. In this case, according to the analysis of Kugel and Khomskii (KK), the spin and the orbital degrees of freedom are not separated. Hence, the spin excitations are necessarily intimately involved in the formation of the orbital excitations and \textit{vice versa}. A possible scenario of orbital liquid formation in LaTiO\textsubscript{3} has been proposed in Ref. 9. These authors have conjectured the appearance of a resonance between the spin and the orbital excitations. To describe this resonance, they have used mean-field decoupling, generalizing the approach proposed in Ref. 6 for the spin liquid state in the two-dimensional $t-J$ model. This decoupling leads inevitably to the conclusion that the resonance states should give a contribution linear in the temperature to the low-energy specific heat. Such a Sommerfeld-like contribution signals the lifting of the orbital degeneracy by forming a quantum liquid. Unfortunately, a direct experimental check of this prediction in LaTiO\textsubscript{3} has yielded negative results.

In the present paper, the main idea of Khaliullin and Maekawa is rehabilitated by means of a more refined approach, which goes beyond the mean-field decoupling. We use the methods recently tested in the theory of Kondo lattices, where the spin liquid state is described as short-range correlations of the RVB type, which arise due to a non-local exchange through the Kondo screening clouds between the magnetic ions. We show that the \textit{two-magnon} excitations play a similar role in the formation of the orbital liquid correlations.

Our analysis is based on the KK Hamiltonian (see below). In its simplest symmetric form, this Hamiltonian \textit{does not} support long-range magnetic order at any non-zero temperature, due to some hidden symmetries. However, the Hamiltonian of the real LaTiO\textsubscript{3} contains many additional (possibly small) terms, which restore long range AF spin order below some Néel temperature $T_N$, as indeed observed experimentally. In the follow-
ing, we therefore assume the existence of this ordered magnetic state, and concentrate on its effects on the low temperature dynamics of the orbital degrees of freedom. Once the magnetic order is stabilized, we expect these dynamics at temperatures $T \ll T_N$ to be described by the KK Hamiltonian, as discussed below. Indeed, we find heavily damped orbital excitations, which form a continuum whose energy scale is the same as that of the spin excitations, such that both orbital and spin excitations merge into a common continuum. As a result, a direct experimental observation of the orbital dynamics alone, at low temperatures, is hardly possible.

The paper is organized as follows. In the next section we review the derivation of the KK Hamiltonian, and set the basis for our perturbation expansion. The latter is carried out in Sec. III, where the orbital spectrum is obtained and discussed. Section IV includes our conclusions.

II. THE KUGEL-KHOMSKII HAMILTONIAN

Following the KK approach, we consider a simplified version of the titanate, concentrating only on the sublattice of Ti ions. Each Ti ion possesses a single electron in one of the three-fold degenerate $t_{2g}$ orbitals of the d-shell. Including only a single Coulomb-Hubbard on-site repulsion on the Ti ion, with energy $U$, the simple initial Hamiltonian reads

$$\mathcal{H} = \frac{U}{2} \sum_i \sum_{mm'} \sum_{\sigma\sigma'} n_{im\sigma} n_{im'\sigma'} (1 - \delta_{mm'} \delta_{\sigma\sigma'}) + \sum_{\langle ij \rangle} \sum_{mm'} \sum_{\sigma} t_{ij}^{mm'} d_{i\sigma}^{\dagger} d_{j\sigma'}, \quad n_{im\sigma} \equiv d_{i\sigma}^{\dagger} d_{i\sigma}, \quad (1)$$

where $\langle ij \rangle$ denotes nearest-neighbor pairs. Here, $d_{i\sigma}^{\dagger}$ creates an electron of spin $\sigma$ at site $i$, in one of the $t_{2g}$ levels, denoted $m$, and $n_{im\sigma}$ is the number operator in the $m$ orbital on site $i$ with spin index $\sigma$. Kugel and Khomskii treated this Hamiltonian in second-order perturbation theory, assuming that the overlap integrals $t_{ij}^{mm'}$ are much smaller than $U$, in order to obtain an effective Hamiltonian describing the spin interactions among the Ti ions. Operating within strictly cubic symmetry, KK assumed that

$$t_{ij}^{mm'} = \delta_{mm'} t_{ij}^{m}, \quad (2)$$

where $t_{ij}^{m} \equiv t$ and differs from zero only when

\begin{align*}
\langle ij \rangle & \text{ is along the } z \text{ axis and } m = d_{yz} \text{ or } d_{zz}, \\
\langle ij \rangle & \text{ is along the } y \text{ axis and } m = d_{xy} \text{ or } d_{yz}, \\
\langle ij \rangle & \text{ is along the } x \text{ axis and } m = d_{xy} \text{ or } d_{yz}. \quad (3)
\end{align*}

The resulting exchange Hamiltonian, for a bond along the $z$-direction, reads

$$\mathcal{H}_{\text{KK}} = -\frac{J}{2} \sum_i (a_i^\dagger a_i + b_i^\dagger b_i) + J \sum_{\langle ij \rangle} \left(\frac{1}{4} + S_i \cdot S_j\right) Y_{\langle ij \rangle}^z, \quad (4)$$

where $S_i$ is the spin operator on site $i$, and

$$J = 2t^2/U \quad (5)$$

sets the energy scale of the spin excitations. The orbital degrees of freedom are now described by the operators $a_i^\dagger (b_i^\dagger, c_i^\dagger)$, which create a spinless fermion (termed ‘orbiton’) in the $yz, xy$ orbital on site $i$. The intermingling of the orbiton degrees of freedom with those of the spin is contained in the second term of Eq. (4), in which KK introduced the orbiton operator product $Y^z$, given by

$$Y_{\langle ij \rangle}^z = a_i^\dagger a_j^\dagger a_j + b_i^\dagger b_j^\dagger b_j + a_i^\dagger b_j^\dagger a_j + b_i^\dagger a_j^\dagger b_j. \quad (6)$$

The expressions for $\mathcal{H}_{\text{KK}}^p$ (for bonds along the $x$-direction) and for $\mathcal{H}_{\text{KK}}^y$ (for bonds along the $y$-direction) are derived from Eq. (4) by cyclic permutations of the operators $a, b,$ and $c$. The full exchange Hamiltonian is the sum of the three,

$$\mathcal{H}_{\text{KK}} = \mathcal{H}_{\text{KK}}^x + \mathcal{H}_{\text{KK}}^y + \mathcal{H}_{\text{KK}}^z. \quad (7)$$

Then, the first term in Eq. (4), when added together with the corresponding terms in $\mathcal{H}_{\text{KK}}^x$ and $\mathcal{H}_{\text{KK}}^y$ yields twice the total number of electrons in the system,

$$N = \sum_i \left(a_i^\dagger a_i + b_i^\dagger b_i + c_i^\dagger c_i\right), \quad (8)$$

which is just a constant.

Contrary to the situation in the manganites, the spin and charge degrees of freedom in the titanate are not separated. Moreover, the orbiton-spin interaction has a peculiar structure of two fermions interacting with one magnon (see below). To explore the outcome of this complicated situation, we proceed as follows. Firstly, we work well below $T_N$. Then the spin operators may be treated by the linearized Holstein-Primakoff transformation: assuming a two sublattice bipartite antiferromagnetic order, with moments along the magnetic $z$-direction, we write

$$X_{\langle ij \rangle} = S_i \cdot S_j + \frac{1}{4}$$

$$= \frac{1}{2} \left(B_i^\dagger B_i + B_j^\dagger B_j + B_i B_j + B_i^\dagger B_j^\dagger\right). \quad (9)$$

Here, $B_i^\dagger$ is a boson operator which creates a magnon excitation (related to $S_{ix} + iS_{iy}$). Secondly, the KK exchange Hamiltonian, together with Eq. (9), will be treated as a perturbation, acting on the zeroth order Hamiltonian. The latter consists of the free orbiton Hamiltonian, and the free spin Hamiltonian. For the
first, we impose a constraint ensuring that each site is occupied by a single charge carrier. This constraint is applied globally. For the latter, we introduce an infinitesimally small staggered magnetic field in the magnetic z-direction, $h_i = (−1)^i h$. Hence,

$$\mathcal{H}_0 = \mathcal{H}_O + \mathcal{H}_S,$$

(10)

with

$$\mathcal{H}_O = −\lambda \sum_i \left( a_i^\dagger a_i + b_i^\dagger b_i + c_i^\dagger c_i \right),$$

(11)

$$\mathcal{H}_S = \sum_i h_i \left( \frac{1}{2} - B_i^\dagger B_i \right).$$

(12)

Note that the chemical potential $\lambda$ includes the constant term of the original KK Hamiltonian (7) [cf. Eq. (8)]. In the next section we use these definitions in order to construct a thermodynamic perturbation theory in terms of the Matsubara Green functions.

### III. Perturbation Expansion for the Orboton Spectrum

The orboton spectrum is determined by the self-energy of the orboton Green function. Here we calculate the orboton Green function, which is diagonal in the orboton operators, that is,

$$G_{ij}^{\mu\nu}(\tau) = \langle T_\tau a_{i\mu}(\tau) a_{j\mu}^\dagger(0) \rangle,$$

(13)

where $i$ and $j$ denote lattice sites, $T_\tau$ is the time-ordering operator, and $a_{i\mu} = a, b,$ or $c$. This Green function satisfies the Dyson equation

$$G_{ij}^{\mu\nu} = g_0 \left( \delta_{ij} + \sum_\nu \sum_l M_{il}^{\mu\nu} G_{ll}^{\nu\nu} \right),$$

(14)

where $g_0$ is the ‘bare’ orboton Green function,

$$g_0(\epsilon_m) = (i\epsilon_m + \lambda)^{-1},$$

(15)

with $\epsilon_m$ being the Matsubara frequency. As we show below, the self-consistent calculation of the orboton self-energy, $M_{il}^{\mu\nu}$, requires knowledge of the magnon Green function, which may be represented by the matrix

$$\hat{D}_{ij}(\tau) = \begin{bmatrix} \langle T_\tau B_i(\tau) B_j^\dagger(0) \rangle & \langle T_\tau B_i(\tau) B_j(0) \rangle \\ \langle T_\tau B_i(\tau)^\dagger B_j^\dagger(0) \rangle & \langle T_\tau B_i^\dagger(\tau) B_j(0) \rangle \end{bmatrix}. $$

(16)

Unlike the standard case of a linear fermion-boson interaction, the KK Hamiltonian (7) contains six-tail interaction vertices, of four types (see Fig. 1). These are denoted $V_1$, $V_2$, $V_3$, and $V_4$ and are given by

$$V_1 = -J B_i^\dagger B_j a_{i\mu} a_{j\nu} a_{i\ell}^\dagger a_{j\ell},$$

$$V_2 = -J B_i^\dagger B_j a_{i\mu} a_{j\nu} a_{i\ell} a_{j\ell}^\dagger,$$

$$V_3 = -J B_i^\dagger B_j a_{i\mu} a_{j\nu} a_{i\ell}^\dagger a_{j\ell},$$

(17)

$V_2$ has the same form as $V_1$ with $i$ replaced by $\ell$, and $V_4 = V_j^\dagger$. Hence the KK Hamiltonian involves a fermion-magnon interaction, which is quadratic in the magnon operators and quartic in the fermion operators. This feature is the basis for our results in the following.

**FIG. 1.** The four magnon-orboton vertices. The wavy lines denote the interaction, full (dotted) lines denote the orboton (magnon) operators.

### A. Lowest-order mean-field approximation

To first order, the perturbation expansion corresponds to the mean-field approximation for the self-energy of both the orbotons and the magnons. Then, these self-energies can be obtained by decoupling the product of magnon operators and orboton operators appearing, e.g., in Eq. (4). This procedure is carried out as follows. Ignoring a constant, one first writes

$$\mathcal{H}^z \equiv \sum_{(ij)_z} X_{(ij)} Y_{(ij)}^z \sim \sum_{(ij)_z} \left[ \langle Y \rangle X_{(ij)} + \langle X \rangle Y_{(ij)}^z \right],$$

(18)

where the orboton operators $Y$ and the magnon operators $X$ are given in Eqs. (6) and (9), respectively. The notation $\langle \rangle$ stands for the thermodynamic average of the relevant operators. A similar decoupling is written down for $\mathcal{H}^\nu$ and $\mathcal{H}^z$. When these three approximate Hamiltonians are summed together, the resulting Hamiltonian separates into two terms, $\mathcal{H}_1$ and $\mathcal{H}_2$, the first pertaining to the magnons, and the second to the orbotons.

Let us first consider $\mathcal{H}_1$ and the magnon dynamics it implies. The orboton operator product contained in $\langle Y \rangle$ is further decoupled, by introducing the thermal averages, which will be determined self-consistently (see below),

$$n_\mu = \langle a_{i\mu}^\dagger a_{i\mu} \rangle, \text{ with } n = n_a = n_b = n_c = 1/3,$$

$$\Delta_\mu = \langle a_{i\mu}^\dagger a_{j\mu} \rangle, \text{ with } \Delta \equiv \Delta_a = \Delta_b = \Delta_c, \ i \neq j.$$  

(19)

In other words, we assume an equal average occupation of all three orbitals (i.e., no orbital long range order), and consider only the possible “ordering” of the off-diagonal...
orbital parameter, \( \Delta \), which represents the delocalization of the orbital degrees of freedom. Then \( H_1 \) takes the form
\[
H_1 = \frac{\tilde{J}}{2} \sum_{\langle ij \rangle} (B_i^\dagger B_i + B_i B_j + B_i B_j^\dagger + B_j B_i^\dagger),
\]
with the coupling constant
\[
\tilde{J} = 2J(n^2 - 2\Delta^2).
\]
Thus, \( H_1 \) reduces to the usual spin-wave antiferromagnetic Hamiltonian, with a renormalized exchange coefficient. As usual, this Hamiltonian is diagonalized in Fourier space, utilizing the Holstein-Primakoff rotation:
\[
B_q = C_q \beta_q + S_q \beta_q^\dagger,
\]
where
\[
C_q \equiv \cosh W_q, \quad S_q \equiv \sinh W_q,
\]
such that
\[
-\tanh 2W_q = \frac{1}{3} (\cos q_x + \cos q_y + \cos q_z) \equiv \Phi_q.
\]
Hence, the magnon Green function takes the form
\[
D_q(\omega_n) = (i\omega_n - \omega_q)^{-1},
\]
with the magnon frequency
\[
\omega_q = 3\tilde{J} \sqrt{1 - \Phi_q^2} \tag{26}
\]
Within the mean field approximation of Eq. (18), the energy scale of the orbiton Hamiltonian \( H_2 \) is set by \( \langle X \rangle \), which can be now obtained using the magnon dispersion (26),
\[
\langle X \rangle = \frac{3}{2N} \sum_q \left[ -1 + (2N_q + 1) \sqrt{1 - \Phi_q^2} \right],
\]
with the magnon occupation numbers \( N_q = (\exp(\beta \omega_q) - 1)^{-1} \). Since the operator \( Y^{(i)} \) still involves products of four operators, one needs to apply further contractions. It is then convenient to calculate the orbiton self-energy using the diagrams shown in Fig. 2. From these diagrams one finds the first order self-energy
\[
M_{1,\mu}(k) = 4J \langle X \rangle \left[ n - \Phi_k^\mu \Delta \right],
\]
with the form factors
\[
\Phi_k^\mu = \cos k_y + \cos k_z, \tag{29}
\]
and analogous expressions for \( \Phi_k^b (y, z \rightarrow z, x) \) and \( \Phi_k^c (y, z \rightarrow x, y) \). It therefore follows that the orbiton Green function, in this approximation, becomes
\[
G_k^{\mu}(\epsilon) = \frac{1}{\epsilon - E_k^\mu}, \tag{30}
\]
where
\[
E_k^\mu = 4J \langle X \rangle \left[ n - \Phi_k^\mu \Delta \right] - \lambda, \quad \mu = a, b, c. \tag{31}
\]

![FIG. 2. The first-order diagrams contributing to the self-energy of the orbiton Green function.](image)

It is thus seen that a finite value of \( \Delta \) implies a finite dispersion of the orbiton modes, with an orbiton band width \( 16J\langle X \rangle \Delta \). Moreover, this band-width is temperature-dependent, due to the temperature dependence of the magnon occupations, contained in \( \langle X \rangle \), Eq. (27), and due to the temperature dependence of \( \Delta \) itself. For a given wave-vector \( k \), the three energies \( E_k^\mu \) are generally different, so that the degeneracy of the \( t_{2g} \) band is removed. Indeed, were we to terminate the calculation at this level, keeping only first-order contributions, then the orbiton averages, Eqs. (19), would have to be found self-consistently using the Green function (30). It is very illuminating to examine those self-consistency requirements. Equation (30) yields
\[
f_k^\mu \equiv \langle a_{\mu k}^\dagger a_{\mu k} \rangle = f(E_k^\mu) \equiv \frac{1}{e^{\beta E_k^\mu} + 1}, \quad \mu = a, b, c. \tag{32}
\]
Then the self-consistency equations read
\[
1 = \frac{1}{N} \sum_{\mu} \sum_k f_k^\mu, \quad \Delta(T) = \frac{1}{6N} \sum_{\mu} \sum_k \Phi_k^\mu f_k^\mu, \tag{33}
\]
where the first equation fixes the chemical potential \( \lambda(T) \), so that the three-fold-degenerate orbiton band is \( 1/3 \) filled, and the second determines \( \Delta \).

Evidently, Eqs. (33) are satisfied by setting \( \Delta = 0 \), (since then the Fermi function is independent of \( k \), and \( \sum \Phi_k^\mu \) vanishes). Let us now examine at which temperature \( \Delta \) begins to deviate from zero. To this end we expand Eqs. (33) in powers of \( \Delta \). The first self-consistency equation yields (at \( \Delta = 0 \))
\[
4J \langle X \rangle(T) n - \lambda(T) = \frac{4}{3} J \langle X \rangle(T) - \lambda(T) = \frac{1}{\beta} \ln 2, \tag{34}
\]
while the second allows a non-zero \( \Delta \) only for temperatures lower than the solution of
\[
\frac{1}{\beta} = \frac{4}{9} J \langle X \rangle(T). \tag{35}
\]
Using here the explicit expression for \( (X) \), Eq. (27), and the results Eqs. (21) and (26) for the magnon frequency, we find
\[
\frac{3}{2\beta J} = -1 + \frac{3}{2\beta J N} \sum_q \beta \omega_q \coth \frac{\beta \omega_q}{2}.
\]
(36)

This is an implicit equation for the temperature at which \( \Delta \) begins to deviate from zero. Examination of this result reveals that this occurs at \( k_B T \) of order \( J \). A similar order of magnitude is found from the spin-wave approximation for the Néel temperature (above which the staggered magnetization \((\frac{1}{2} - B_1)\) vanishes). Both of these estimated temperatures are probably above the real Néel temperature, which is determined by small symmetry breaking terms which are not contained in the simple KK model. Hence, in the whole range below the real \( T_N \) we expect to have a finite \( \Delta(T) \), and we do not expect any phase transition at which \( \Delta \) vanishes while the spins are still ordered. Even if this statement is not valid for all \( T < T_N \), we still expect it to hold for sufficiently low temperatures. In the next section we examine the dynamical corrections to the orbiton spectrum, brought about by the second-order corrections, and examine whether and how the above conclusion is modified.

**B. Beyond the mean-field approximation**

The next order contribution to the orbiton self-energy, \( M_{2,\mu}(k,\epsilon) \), requires expansion to second order in the KK Hamiltonian, i.e. to second order in the diagrams listed in Fig. 1. Instead of going through the details of all the diagrams which contribute to \( M_{2,\mu} \), we give here the details of only two diagrams, which describe orbiton propagation assisted by two-magnon processes (Fig. 3). The first of these two diagrams is generated by the vertices \( V_1 \) and \( V_2 \) in Fig. 1, whereas the second is generated by the vertices \( V_3 \) and \( V_4 \). Both diagrams contain a single propagating fermion line. Each of the other two fermion tails (of each vertex) is rolled into a bubble, to give the factor \( n^2 \). Other diagrams involve three internal fermion lines, but do not change the qualitative features discussed below.

FIG. 3. Second-order diagrams contributing to the self-energy of the orbiton Green function.

A lengthy calculation of the contribution of the two diagrams displayed in Fig. 3 yields (after performing the analytical continuation for the Matsubara frequencies)
\[
M_{2,\mu}^{(1)}(k,\epsilon) = \left( \frac{nJ}{N} \right)^2 \sum_{pq} \left( \frac{\Phi_{\mu q}^+ + \Phi_{p-q}^\mu}{2} \right)^2 \times \left[ C_{q,p+q}^+ \frac{N_q(1 + N_{p+q}) + f_{k-p}^\mu(N_{p+q} - N_q)}{\epsilon - \beta \omega_k + \beta \omega_q - \beta \omega_{p+q}} \right. \\
+ \frac{1}{2} C_{q,p+q}^- \frac{N_q N_{p+q} + f_{k-p}^\mu(1 + N_{p+q} + N_q)}{\epsilon - \beta \omega_k - \beta \omega_q + \beta \omega_{p+q}} \\
n + \left( 1 + N_q)(1 + N_{p+q}) - f_{k-p}^\mu(1 + N_{p+q} + N_q) \right) \right],
\]
(37)

for the first diagram, and
\[
M_{2,\mu}^{(2)}(k,\epsilon) = \left( \frac{nJ}{N} \right)^2 \sum_{pq} \left( \frac{\Phi_{\mu q}^+ - \Phi_{p-q}^\mu}{2} \right)^2 \times \left[ C_{q,p+q}^+ \frac{N_q(1 + N_{p+q}) + f_{k-p}^\mu(N_{p+q} - N_q)}{\epsilon - \beta \omega_k + \beta \omega_q - \beta \omega_{p+q}} \right. \\
+ \frac{1}{2} C_{q,p+q}^- \frac{N_q N_{p+q} + f_{k-p}^\mu(1 + N_{p+q} + N_q)}{\epsilon - \beta \omega_k - \beta \omega_q + \beta \omega_{p+q}} \\
n + \left( 1 + N_q)(1 + N_{p+q}) - f_{k-p}^\mu(1 + N_{p+q} + N_q) \right) \right],
\]
(38)

for the second diagram. Here we have introduced the magnon coherence factors,
\[
C_{q,p+q}^+ = (C_q C_{p+q} + S_q S_{p+q})^2; \\
C_{q,p+q}^- = (S_q C_{p+q} + C_q S_{p+q})^2; \tag{39}
\]
which, upon using Eq. (23), take the simpler form
\[
C_{q,p+q}^\pm = \begin{cases} 
\cosh^2(W_q + W_{p+q}) \\
\sinh^2(W_q + W_{p+q})
\end{cases}.
\]
(40)

With the second-order corrections to the orbiton self-energy, the orbiton Green function becomes
\[
G_{k}^{\mu\mu}(\epsilon) = \frac{1}{\epsilon - \beta \omega_k - M_{2,\mu}(k,\epsilon)}, \tag{41}
\]
where \( M_{2,\mu}(k,\epsilon) = M_{2,\mu}^{(1)}(k,\epsilon) + M_{2,\mu}^{(2)}(k,\epsilon) + ... \). We now concentrate on the contributions of Eqs. (37) and (38) to this Green function. Clearly, these contributions yield
a finite, temperature-dependent imaginary part to the orbiton dispersion, namely, the orbiton modes acquire a temperature-dependent damping, as expected. The new dispersion law is obtained from the poles of the Green function (41),

\[ z - E^\mu_k = M_{2,\mu}(k, z), \quad (42) \]

where \( z \) is a complex variable. Examining Eqs. (37) and (38) one may draw the following conclusions. (i) The real part of the second-order self-energy, \( R(k, \epsilon) = \text{Re} M_{2,\mu}(k, \epsilon) \), as function of \( \epsilon \), contains three components which come from the two-magnon processes. The lowest one arises from the two-magnon emission processes and is dominant in the interval \(-2\omega_D \leq \epsilon \leq 0\), where \( \omega_D \) is the maximal magnon frequency [see Eq. (26)]. The second results from the processes in which one magnon is emitted and one is absorbed, which occur in the interval \( -\omega_D \leq \epsilon \leq \omega_D \). The third component comes from the two-magnon absorption processes and prevails at positive frequencies, \( 0 \leq \epsilon \leq 2\omega_D \). It follows that \( R(k, \epsilon) \) has a significant magnitude in the entire range \(-2\omega_D < \epsilon < 2\omega_D \), and falls down rapidly outside this interval. The magnitude of different contributions to \( R(k, \epsilon) \) is regulated by the magnon occupation numbers. In particular, the mixed emission-absorption processes are frozen out at \( T \to 0 \). One may conclude that the superposition of the three types of the two-magnon processes results in a complicated behavior of \( R(k, \epsilon) \). This quantity may have several zeros and peaks within the above energy interval; the heights of these peaks can be estimated as \( \sim \kappa J \), with a numerical coefficient \( \kappa < 1 \). (ii) The imaginary part of the orbiton self-energy, which comes from the second-order terms, \( \Gamma(k, \epsilon) = \text{Im} M_{2,\mu}(k, \epsilon) \), behaves similarly to the real part: expressions (37) and (38) do not show that \( \Gamma \) is essentially smaller than \( R \) in the whole energy interval. (iii) As a result, the orbitons become ill-defined, heavily damped, quasi-particles. Their off-diagonal orbiton parameter \( \Delta \), and the related orbiton band-width, should be again determined self-consistently, using the refined orbiton Green function Eq. (41),

\[ \Delta(T) = -\frac{1}{6N} k \sum \sum \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \Phi^\mu_k \text{Im} G^\mu_k(\epsilon) f(\epsilon), \quad (43) \]

in place of the second of Eqs. (33).

Without going into the details of the solutions of the complicated self-consistent equation for \( \Delta(T) \), Eq. (43), it is clear that going beyond the simple mean-field calculation changes drastically the nature of the orbiton excitations. Whereas in the first order we have obtained a coherent orbiton spectrum with a well-defined wave vector (appearing due to the magnetic order), this picture changes drastically with the second-order corrections. The interaction with the spin wave excitations extends the effective width of the orbiton band and makes it asymmetric relative to its center of gravity. Moreover, the orbiton excitations lose their coherence by acquiring a considerable life-time (given by the imaginary part of the self-energy). Those excitations are thus transformed into a structureless “liquid-like” continuum. Clearly, more complicated processes involving higher-orders of the orbiton-magnon interaction, are not expected to modify this picture.

IV. CONCLUDING REMARKS

We have proposed in this paper a mechanism capable of lifting the degeneracy of the \( t_{2g} \) orbitals in the titanate. In our scenario, the orbital degeneracy is removed due to the interplay between the (ordered) spin and the (disordered) orbital degrees of freedom. We have obtained three dispersive orbiton bands, with a temperature-dependent band-width and a structureless spectrum, that exist together with the coherent spin fluctuations at low temperatures. We have found that the second-order perturbation corrections transform the coherent orbiton spectrum, \( E^\mu_k \) [see Eq. (31)] obtained within the simplest mean-field theory, into an incoherent continuum of heavily damped excitations, which may be termed an ‘orbital liquid’. This, in turn, may explain the experimental observation,\(^{10}\) that the low-temperature heat capacity arises from the spin wave excitations alone.

The incoherent continuum state of the orbital excitations is dominated at low temperatures by the two-magnon emission peak in the orbiton self-energy. The effect of this peak is to shift the weight of the orbiton spectrum into negative energies, thus making the orbiton liquid state thermodynamically preferable as compared to the fully-degenerate orbiton state that prevails when \( \Delta = 0 \), or in the absence of the coupling with the spin degrees of freedom. In particular, our process of degeneracy lifting may possibly overcome the Jahn-Teller mechanism.\(^{13}\) In the latter, the energy gain is restricted by the energy of the Jahn-Teller coupling strength, which is definitely smaller than the exchange coupling, which determines the energy gain in our case.

Finally we mention that, within the framework of the KK Hamiltonian in cubic symmetry, the orbiton-magnon interaction cannot lead to a magnetic anisotropy, that is, it cannot generate a gap in the spin-wave spectrum. To obtain such anisotropies, one should extend the KK Hamiltonian to include the hybridization of the oxygen and titanium orbitals, the possible orthorhombic tilting of the oxygen octahedra, as well as spin-orbit interactions. Apparently, one will also have to treat the Coulomb interaction in a more refined way, taking into account Coulomb-exchange effects.\(^5\)
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