Orbital Wave and its Observation in Orbital Ordered Titanates and Vanadates

Sumio Ishihara

Department of Physics, Tohoku University, Sendai 980-8578, Japan

(November 4, 2018)

We present a theory of the collective orbital excitation termed orbital wave in perovskite titanates and vanadates with the triply degenerate $t_{2g}$ orbitals. The dispersion relations of the orbital waves for the orbital ordered LaVO$_3$, YVO$_3$ and YTiO$_3$ are examined in the effective spin-orbital coupled Hamiltonians associated with the Jahn-Teller type couplings. We propose possible scattering processes for the Raman and inelastic neutron scatterings from the orbital wave and calculate the scattering spectra for titanates and vanadates. It is found that both the excitation spectra and the observation methods of the orbital wave are distinct qualitatively from those for the $e_g$ orbital ordered systems.

PACS numbers: 75.10.-b, 75.30.Et, 78.70.-g, 78.70.Nx

I. INTRODUCTION

When degenerate electron orbitals are partially filled in correlated electron systems, this is recognized to be an internal degree of freedom belonging to an electron as well as spin and charge. This orbital degree of freedom has recently attracted much attention especially in transition-metal (TM) oxides.\textsuperscript{1,2} In particular, the orbital ordering (OO) and fluctuation play a key role in anisotropic electric, magnetic and optical properties in correlated oxides.

The collective orbital excitation in the orbital ordered state is known to be orbital wave (OW), in analogous to the spin wave in magnetically ordered state, and its quantized object is termed orbiton. The theoretical study of OW has started in the idealized spin-orbital coupled model where the continuous symmetry exists in the orbital space,\textsuperscript{3} and progressed in the anisotropic orbital model.\textsuperscript{4} A realistic calculation has been done in LaMnO$_3$ with the doubly degenerate $e_g$ orbitals.\textsuperscript{5} Recently new peak structures observed in the Raman spectra in LaMnO$_3$ are interpreted as scatterings from OW.\textsuperscript{6} Although there are some debates about origin of the newly found peaks, OW or multi-phonons,\textsuperscript{7,8} the energy and polarization dependences of the observed Raman spectra show a good agreement with the calculation based on the OW interpretation.

The perovskite titanates $R$TiO$_3$ and vanadates $R$VO$_3$ ($R$: rare-earth ion) with the triply degenerate $t_{2g}$ orbitals are another class of materials where OW is expected. One of the well known orbital ordered materials is YTiO$_3$ where the nominal valence of the titanium ion is $3^+$ and one electron occupies the triply degenerate $t_{2g}$ orbitals. The orbital ordered state associated with the Jahn-Teller (JT) type lattice distortion has recently new peak structures observed in the Raman spectra in YTiO$_3$ and YVO$_3$. The calculations are based on the effective spin-orbital coupled Hamiltonians associated with the JT type electron-lattice coupling. We propose possible scattering processes for the Raman and inelastic neutron scatterings from OW and calculate the spectra. It is found that the excitation spectra and the observation methods of OW are distinct qualitatively from those for the $e_g$ orbital ordered systems.

termed the $(d_{y(x+z)}/d_{y(x-z)}/d_{x(y+z)}/d_{x(y-z)})$-type from now on, is also supported by previous calculations.\textsuperscript{14–16} Although an exotic orbital state is proposed recently by taking into account the quantum fluctuation,\textsuperscript{17,18} the predicted orbital state being incompatible with the crystal lattice symmetry is different from the above type of OO.

A series of $R$VO$_3$ is systematically examined in the recent studies.\textsuperscript{14,15,19–20} Two electron occupy the triply degenerate orbital in a $V^{3+}$ ion. A sequential phase transition is found in YVO$_3$,\textsuperscript{19–21} the G-type OO (O-G) occurs at $T_{OG1}$=200K and the C-type antiferromagnetic (AFM) ordering (S-C) appears at $T_{N1}$=115K. With further decreasing temperature, another orbital and magnetic transitions appear at $T_{N2}=T_{OG2}=77K$ where the C-type OO associated with the G-type AFM order (the C-G/O-C order) is realized. On the other hand, in LaVO$_3$, the C-type AFM ordering occurs at $T_N$(=143K) and, at slightly below this temperature, the G-type OO (the C-G/O-G order) appears.\textsuperscript{20–22} Types of OO in vanadates are determined that the $d_{xy}$ orbital is occupied at all the vanadate sites and the $d_{xz}$ and $d_{zx}$ orbitals are alternately ordered in the $xy$ plane (the C-type OO), and in all direction (the G-type OO).\textsuperscript{24,25} These kinds of OO are termed the pure OO states, and the OO such as realized in YTiO$_3$ is termed the mixed OO from now on. It is suggested that the experimentally observed type of OO in vanadates associated with the JT type distortion explains the several optical and magnetic properties.\textsuperscript{26,29}
ordered systems such as manganites.

In Sec. II, the model Hamiltonian for titanates and vanadates with the triply degenerate $t_{2g}$ orbitals are introduced. In Sec. III, the dispersion relations of OW for LaVO$_3$, YVO$_3$ and YTiO$_3$ are examined. The scattering spectra for the Raman and inelastic neutron scatterings from OW are shown in Sec. IV. Section V is devoted to summary and discussion.

II. MODEL HAMILTONIAN

The collective orbital excitations in the orbital ordered state are studied in the spin-orbital model derived from the generalized Hubbard Hamiltonian with the triply degenerate $t_{2g}$ orbitals:

$$
\mathcal{H} = \varepsilon_d \sum_{i,\sigma,\gamma} d_{i\gamma\sigma}^\dagger d_{i\gamma\sigma} + \mathcal{H}_{el-el} \\
+ \sum_{(ij),\gamma,\gamma',\sigma} \left( \tilde{t}_{ij}^{\gamma\gamma'} d_{i\gamma\sigma}^\dagger d_{j\gamma'\sigma} + H.c. \right),
$$

with the electron-electron interaction

$$
\mathcal{H}_{el-el} = U \sum_{i,\gamma} n_{i\gamma\uparrow} n_{i\gamma\downarrow} + U' \sum_{i,\gamma,\gamma'} n_{i\gamma\uparrow} n_{i\gamma'\downarrow} \\
+ J \sum_{i,\gamma,\gamma' \neq \gamma'} d_{i\gamma\sigma}^\dagger d_{i\gamma'\sigma}^\dagger d_{i\gamma\sigma} d_{i\gamma'\sigma},
$$

$d_{i\gamma\sigma}$ is the annihilation operator for the $t_{2g}$ electron at site $i$ with spin $\sigma = (\uparrow, \downarrow)$ and orbital $\gamma = (yz, xz, xy)$. The number operators are defined by $n_{i\gamma\sigma} = d_{i\gamma\sigma}^\dagger d_{i\gamma\sigma}$ and $n_{i\gamma} = \sum_{\sigma} n_{i\gamma\sigma}$. $U$ and $U'$ are the intra- and inter-orbital Coulomb interactions, respectively. $I$ is the exchange interaction, and $J$ is the pair-hopping interaction.

In Eq. (1) is the electron transfer integral between site $i$ with orbital $\gamma$ and nearest neighboring (NN) site $j$ with $\gamma'$. In the ideal perovskite structure, the hopping integral is simplified as $\tilde{t}_{ij}^{\gamma\gamma'} = t_0 \delta_{\gamma\gamma'}(\delta_{\gamma=\downarrow(\uparrow)} + \delta_{\gamma=\uparrow(\downarrow)})$ where $l$ indicates a direction of a bond connecting sites $i$ and $j$, and $(h, k, l) = (x, y, z), (y, z, x), (z, x, y)$. The electron hopping occurs through the 0 2$\mathcal{P}$ orbitals in between the NN TM sites.

We derive the effective Hamiltonian for titanates where the nominal electron configuration of the TM ion is $d^3$. This Hamiltonian is defined in the Hilbert space where the electron number per site is restricted to be one or less due to the strong on-site Coulomb interactions. The Hamiltonian is classified by the intermediate states of the perturbational processes as derived in Ref. 13:

$$
\mathcal{H}_{Ti} = \mathcal{H}_{T_1} + \mathcal{H}_{T_2} + \mathcal{H}_E + \mathcal{H}_{A_1},
$$

with

$$
\mathcal{H}_{T_1} = -J_{T_1} \sum_{(ij)} \left( \frac{3}{4} + \tilde{S}_i \cdot \tilde{S}_j \right) \left( A'_l + B'_l - C'_l \right),
$$

$$
\mathcal{H}_{T_2} = -J_{T_2} \sum_{(ij)} \left( \frac{1}{4} - \tilde{S}_i \cdot \tilde{S}_j \right) \left( A'_l + B'_l + C'_l \right),
$$

$$
\mathcal{H}_E = -J_E \sum_{(ij)} \left( \frac{1}{4} - \tilde{S}_i \cdot \tilde{S}_j \right) \frac{2}{3} \left( 2A'_l - C'_l \right),
$$

$$
\mathcal{H}_{A_1} = -J_{A_1} \sum_{(ij)} \left( \frac{1}{4} - \tilde{S}_i \cdot \tilde{S}_j \right) \frac{2}{3} \left( A'_l + C'_l \right).
$$

The superexchange interactions are given by $J_{T_1} = \frac{t_0^2}{E_{11}^{(2)}} (\Gamma = T_1, T_2, E, A_1)$ where $E_{11}^{(2)}$ are the energies of the intermediate states: $E_{11}^{(2)} = U'I - I, E_{11}^{(2)} = U - I, E_{11}^{(2)} = U' + I$ and $E_{11}^{(2)} = U + 2I$ where the relations $U = U' + 2I$ and $I = J$ are used. $J_{T_1}$ is the largest among them. The spin degree of freedom is described by the operator $\tilde{S}_i = \frac{1}{2} \sum_{\gamma,s} d_{i\gamma \sigma}^\dagger d_{i\gamma \sigma}$ with the Pauli matrices $\sigma$. The orbital degree of freedom is represented by the eight orbital operators $O_{l\gamma}$ classified by the irreducible representations of the $O_h$ group as $(\Gamma \gamma m) = (Eu, Ev, T(2)\gamma, (T_2x, T_2y, T_2z), (T_1x, T_1y, T_1z))$. These operators are defined by the generators of the SU(3) Lie algebra, i.e. the 3×3 Gell-Mann matrices $\lambda_m (m = l \sim 8)$ as

$$
O_{l\gamma m} = -\frac{1}{\sqrt{2}} \sum_{\sigma,\alpha,\beta} d_{l\alpha\beta}^\dagger \lambda_{m\beta} d_{l\beta\sigma},
$$

where $(\Gamma \gamma m) = (Eu, Ev, (T_2x, 6), (T_2y, 4), (T_2z, 1), (T_1x, 7), (T_1y, 5), (T_1z, 2))$. The operators $O_{l\Gamma E}$ and $O_{lT_2 \gamma}$ represent the diagonal and off-diagonal components of the electric quadrupole moments, respectively, and $O_{lT_1 \gamma}$ does the magnetic dipole ones, i.e. the orbital angular momentum. By utilizing the above orbital operators, the orbital parts of the Hamiltonian in Eqs. (4)-(7) are given as

$$
A'_l = W_l^i W_l^j \pm O_{lEu}^i O_{lEu}^j,
$$

$$
B'_l = V_l^i W_l^j + W_l^i V_l^j,
$$

$$
C'_l = 2(O_{lT_2} O_{lT_2} \pm O_{lT_1} O_{lT_1}),
$$

with

$$
W_l^i = \frac{2}{3} - \sqrt{\frac{2}{3}} O_{lEu}^i,
$$

$$
V_l^i = \frac{1}{3} + \sqrt{\frac{2}{3}} O_{lEu}^i.
$$
The nominal electron configuration is \( d \) with \( S \). In analogy with the spin Hamiltonian, \( A \) similar way from Eq. (1). The electron number at each orbital, e.g. \( O \), we introduce the spin operator \( \vec{S} \) that a similarity between \( l \) systems, there are two kinds of the JT type interactions; \( \Delta \) in the triply degenerate \( d \) electron picture. Similar types of the spin-orbital coupling, the GdFeO\(_3\)-type lattice distortion, lifts the degeneracy. We suppose that the Jahn-Teller type distortion with the \( T_{2g} \) symmetry, \( g_{Tz}Q_{Tz} \) in Eq. (20), plays a key role to stabilize the observed mixed orbital order. The (S-C/O-G) order for LaVO\(_3\) is reproduced by \( H_V \). The FM (AFM) order along the \( c \) axis (in the \( ab \) plane) is attributed to the alternate (uniform) alignment of the \( d_{yz} \) and \( d_{zx} \) orbitals (the \( d_{xy} \) orbital) along the \( c \) axis (in the \( ab \) plane). The (S-G/O-C) order for the YVO\(_3\) is obtained by \( H_V \) and the Jahn-Teller type interaction with the \( E_g \) symmetry. The AFM spin order and the uniform alignment of the \( d_{yz} \) orbitals along the \( c \) axis is stabilized cooperatively, as discussed in the next section.

III. ORBITAL WAVE

The dispersion relations of OW in the orbital ordered states are obtained by utilizing the Holstein-Primakoff transformation for the generators in the SU(3) algebra. For example, at a site where the \( d_{xy} \) orbital is occupied, there are two excitation modes; an excitation between the \( d_{xy} \) and \( d_{yz} \) orbitals denoted by a boson operator \( y (y^\dagger) \), and that between \( d_{xy} \) and \( d_{zx} \) denoted by \( x (x^\dagger) \). The orbital operators \( O_{iTγ} \) are transformed into

\[
O_{iT_Eu} \text{ and } O_{iT_Ev} \text{ are defined by}
\]

\[
\left( \frac{O_{iT_Eu}}{O_{iT_Ev}} \right) = \left( \begin{array}{cc} \cos \frac{2\pi}{3} m_1 & \sin \frac{2\pi}{3} m_1 \\ -\sin \frac{2\pi}{3} m_1 & \cos \frac{2\pi}{3} m_1 \end{array} \right) \left( \frac{O_{iEu}}{O_{iEv}} \right),
\]

with \( m_i = (1, 2, 3) \) for a direction of the bond \( l = (x, y, z) \).

\[
\text{In analogy with the spin Hamiltonian, } A \text{ similar way from Eq. (1). The electron number at each orbital, e.g. } O \text{ we introduce the spin operator } \vec{S} \text{ that a similarity between } l \text{ systems, there are two kinds of the JT type interactions; } \Delta \text{ in the triply degenerate } d \text{ electron picture. Similar types of the spin-orbital coupling, the GdFeO}_3\text{-type lattice distortion, lifts the degeneracy. We suppose that the Jahn-Teller type distortion with the } T_{2g} \text{ symmetry, } g_{Tz}Q_{Tz} \text{ in Eq. (20), plays a key role to stabilize the observed mixed orbital order. The (S-C/O-G) order for LaVO}_3\text{ is reproduced by } H_V \text{. The FM (AFM) order along the } c \text{ axis (in the } ab \text{ plane) is attributed to the alternate (uniform) alignment of the } d_{yz} \text{ and } d_{zx} \text{ orbitals (the } d_{xy} \text{ orbital) along the } c \text{ axis (in the } ab \text{ plane). The (S-G/O-C) order for the YVO}_3\text{ is obtained by } H_V \text{ and the Jahn-Teller type interaction with the } E_g \text{ symmetry. The AFM spin order and the uniform alignment of the } d_{yz} \text{ orbitals along the } c \text{ axis is stabilized cooperatively, as discussed in the next section.}

where \( g_E \) and \( g_{Tz} \) are the coupling constants and \( Q_{iEγ} \) and \( Q_{iTzγ} \) are the normal modes in an O\(_6\) octahedron with symmetries \( E_g \) and \( T_{2g} \), respectively. \( Q_{Eγ} \) directly modifies the TM-O bond lengths and \( Q_{Tzγ} \) modifies the O-TM-O bond angles.

Energy parameter values have been numerically evaluated by several authors. The effective exchange parameters \( J_S \) in the Heisenberg model are obtain from the spin-wave dispersion relations as \( J_S^2 = 5.5 \text{meV ( } z \text{ axis) and } J_S^3 = 5.8 \text{meV ( } xy \text{ plane) in the (S-G/O-C) phase in YVO}_3\text{, } J_S^2 = 2.2 - 4 \text{meV, } J_S^3 = 2.6 \text{meV in the (S-C/O-G) phase in YVO}_3\text{ (Ref. 26), and } J_S^3 = J_S^2 = 3 \text{meV in YTiO}_3\text{ (Ref. 17). The effective exchange parameters for the orbital operators in LaVO}_3\text{ are also estimated as } J_O^2 = 4t_O^2/(U'-1)=33 \text{meV and } J_O^3 = 2 \text{meV (Ref. 28). The JT stabilization energy is obtained from the LDA+U method}^{28} \text{ as } E_{JT} = 27 \text{meV which is comparable or larger than the exchange interactions. The relativistic spin-orbit interaction which is not taken into account in the present model is about 0.4meV being much smaller than both the exchange and JT energies.}^{13} \text{ This is consistent with the experimental results in the magnetic x-ray scattering in YTiO}_3\text{; the angular momentum separately estimated from the spin momentum is found to be negligible small.}^{34} \text{ Thus, the Hamiltonian } H_{Tz(γ)} + H_{JT} \text{ introduced above is the minimal model for examination of OW. Here we mention the implications of the theoretical model for the observed spin/orbital orders. As we have shown in Ref. 13, in the mean field theory, the large orbital degeneracy remains in the ferromagnetic (FM) ground state in } H_T. \text{ A small perturbation, such as the Jahn-Teller type distortion, the relativistic spin-orbit coupling, the GdFeO}_3\text{-type lattice distortion, lifts the degeneracy. We suppose that the Jahn-Teller type distortion with the } T_{2g} \text{ symmetry, } g_{Tz}Q_{Tz} \text{ in Eq. (20), plays a key role to stabilize the observed mixed orbital order. The (S-C/O-G) order for LaVO}_3\text{ is reproduced by } H_V \text{. The FM (AFM) order along the } c \text{ axis (in the } ab \text{ plane) is attributed to the alternate (uniform) alignment of the } d_{yz} \text{ and } d_{zx} \text{ orbitals (the } d_{xy} \text{ orbital) along the } c \text{ axis (in the } ab \text{ plane). The (S-G/O-C) order for the YVO}_3\text{ is obtained by } H_V \text{ and the Jahn-Teller type interaction with the } E_g \text{ symmetry. The AFM spin order and the uniform alignment of the } d_{yz} \text{ (for the } d_{zx} \text{ orbital) along the } c \text{ axis axis is stabilized cooperatively, as discussed in the next section.}

\[
H_V = H_{A_1} + H_E + H_{T_1} + H_{T_2},
\]

with

\[
H_{A_1} = -J_{A_1} \sum_{\langle ij \rangle} \frac{1}{6} \left( 2 + \bar{S}_i \cdot \bar{S}_j \right) \times (-A_1^l + A_1^l + B^l - 2C_1^l),
\]

\[
H_E = -J_E \sum_{\langle ij \rangle} \frac{1}{6} \left( 1 - \bar{S}_i \cdot \bar{S}_j \right) \times (-A_1^l + A_1^l + B^l + C_1^l),
\]

\[
H_{T_1} = -J_{T_1} \sum_{\langle ij \rangle} \frac{1}{4} \left( 1 - \bar{S}_i \cdot \bar{S}_j \right) (A_1^l + C_1^l),
\]

\[
H_{T_2} = -J_{T_2} \sum_{\langle ij \rangle} \frac{1}{4} \left( 1 - \bar{S}_i \cdot \bar{S}_j \right) (A_1^l + C_1^l).
\]
these boson operators as

\[
\begin{align*}
O_{iE_u} &= \sqrt{2} - \sqrt{3} \left( n_{ix} + n_{iy} \right), \\
O_{iE_v} &= \sqrt{2} \left( n_{iy} - n_{ix} \right), \\
O_{iT_{ax}} &= \left( i \right)_{\alpha} \left( y_i x_i \pm x_i^\dagger y_i \right), \\
O_{iT_{ay}} &= \left( i \right)_{\alpha} \left( \sqrt{1 - N_i y_i} \pm y_i^\dagger \sqrt{1 - N_i} \right), \\
O_{iT_{ax}} &= \left( i \right)_{\alpha} \left( x_i^\dagger \sqrt{1 - N_i} \pm \sqrt{1 - N_i} x_i \right),
\end{align*}
\]

with \( \alpha = (1, 2) \). The plus and minus signs in \( O_{iT_{ax}, ay} \) and \( O_{iT_{az}} \) are for the \( \alpha = 1 \) and \( \alpha = 2 \) cases, respectively. We define \( N_i = n_{ix} + n_{iy} \) with \( n_{ix} = x_i^\dagger x_i \) and \( n_{iy} = y_i^\dagger y_i \). In the linear spin wave approximation, \( \sqrt{1 - N_i} \) is replaced by 1. We have checked that the Green's function method for the operators \( O_{iT_{\gamma \gamma}'} \), i.e.

\[
G_{T_{\gamma} T_{\gamma}'}(t-t') = -i\theta(t-t') \langle [O_{iT_{\gamma}'}(t), O_{iT_{\gamma}'}(t')] \rangle,
\]

with the decoupling approximation, reproduces the calculated results for OW with the boson method introduced above.

In Fig. 1, we present the dispersion relations of OW in the (S-C/O-G) and (S-G/O-C) phases for LaVO\(_3\) and YVO\(_3\) (the low temperature phase), respectively. The parameter values are chosen to be \( I/U' = 0.125 \), \( gE_{Q_E}/J_0 = 0.8 \) and \( gT_2/Q_2 = 0 \). The energy parameters are normalized by \( J_0 = 4t_0^2/J_0 (U' - I) \) which is estimated to be about 33meV for LaVO\(_3\) (Ref. 28) and is supposed to be smaller in YTiO\(_3\) due to the larger GdFeO\(_3\)-type lattice distortion. The ratios of the exchange parameters are represented by the parameter \( R \) as \( J_{T_2}/J_{T_1} = J_E/J_{T_1} = (1 - R)/(1 + 2R) \) and \( J_{A_1}/J_{A_2} = (1 - R)/(1 + 4R) \). Both the (S-C/O-G)- and (S-G/O-C)-phases, there are four modes of OW attributed to the two different orbital occupied sites in a unit cell; there are the excitations \( y_A \) \((d_{xy} \rightarrow d_{yz})\) and \( z_A \) \((d_{xx} \rightarrow d_{xz})\) at site A where the \( d_{xy} \) and \( d_{xz} \) orbitals are occupied, and the excitations \( x_B \) \((d_{xy} \rightarrow d_{xx})\) and \( y_B \) \((d_{xz} \rightarrow d_{xz})\) at site B where the \( d_{xz} \) and \( d_{yz} \) orbitals are occupied. Two of the four, i.e. \( y_A \) and \( x_B \), are the local modes within the linear spin wave theory and do not show dispersions. This character does not depend on the spin arrangements. These local modes originate from the facts that (1) the excited \( d_{xy} \) hole does not hop along the z axis due to the orbital symmetry, and (2) a coherent motion of the excited \( d_{xy} \) hole in the xz plane is impossible, since this motion is associated with increasing the number of the wrong orbital arrangements. The latter implies that the orbital exchange processes do not recover the wrong orbital arrangements, and the triply degenerate orbital model is qualitatively different from the Heisenberg model with \( S = 1 \). The remaining modes, \( z_A \) and \( z_B \), are dispersive along the z direction. The dispersion relation of OW in the (S-C/O-G) phase is explicitly obtained as

\[
E(\vec{k}) = \frac{8}{6} J_{A_1} \sqrt{(K_2^R + K_2^Z)^2 - \left( \frac{2}{3} K_2^R \cos k_z \right)^2}
\]

where we assume \( I = 0 \) and \( gE_{Q_E} = gT_2 Q_2 = 0 \). We introduce

\[
K_2^R(z) = 2 + \langle \vec{S}_i \cdot \vec{S}_{i+\delta(\vec{d}_3)} \rangle \text{ and } K_2^Z(z) = 1 - \langle \vec{S}_i \cdot \vec{S}_{i+\delta(\vec{d}_3)} \rangle.
\]

This energy has its minimum at \( k_z = 0 \) and the energy gap is attributed to the anisotropy in the orbital space, i.e. a lack of the SU(3) symmetry, in the orbital part of the Hamiltonian. In comparison with the OW in the (S-C/O-G) phase, the OW in the (S-G/O-C) phase is barely stable; with decreasing the spin correlation which corresponds to increasing temperature toward \( T_N \), a remarkable softening around \( \vec{k} = (0, 0, \pi) \) occurs. In the case where \( I = 0 \) and \( gT_2 Q_2 = 0 \), the dispersion relation is given as \( E(\vec{k}) = \frac{4}{3} J_{A_1} (2 K_2^Z - K_2^R) \cos k_z + \sqrt{\frac{2 gE_{Q_E}}{3}} \) which has its minimum value at \( k_z = \pi \), and the energy gap is attributed to the JT type interaction. This result suggests an instability of the (S-G/O-C) phase to the (S-C/O-G) one with increasing temperature. This is consistent with the experimental fact that in RVVO\(_3\) the O-C phase appears associated with the S-G order, and is changed into the (S-C/O-G) phase at 77K in YVO\(_3\). In Fig. 2, we present the dispersion relations of OW in the \((d_{y(x+z)}/d_{y(x+z)}/d_{x(y+z)}/d_{x(y+z)})\)-type OO for YTiO\(_3\). The FM and paramagnetic spin orders are assumed, and the exchange parameter is taken to be \( R = I/U' = 0.125 \). The JT type interaction parameters are chosen to be \( gE_{Q_E}/J_0 = gT_2 Q_2/J_0 = 1.2 \) where \( gE_{Q_E} \) is larger than that in vanadates and \( gT_2 Q_2 \) is introduced. This is based on a consideration that, in comparison with vanadates, \( J_0 \) is supposed to be smaller due to the large GeFeO\(_3\)-type lattice distortion, and \( Q_2 \) is found to be
larger in the crystal structure of YTiO$_3$.\textsuperscript{12,22,23} In contrast to the case of vanadates, all the eight modes, attributed to the four different orbitals in a unit cell, are dispersive along all the directions in the Brillouin zone. This originates from the OO states with the mixed orbitals where the excitations propagate along the three directions. The OW dispersions are classified into the two groups: for example, in the directions. The OW dispersions are classified into the orbitals where the excitations propagate along the three levels. This originates from the OO states with the mixed orbitals. The parameter values are chosen to be $I/U’=0.125$, $g_E Q_E/J_0=g_I Q_I/J_0=1.2$. The Brillouin zone for the tetragonally symmetric is adopted.

IV. OBSERVATION OF ORBITAL WAVE

A. raman scattering

As introduced in Sec. I, in orbital ordered LaMnO$_3$, the new peak structures in the Raman spectra were explained successfully as the scattering from OW.\textsuperscript{6} Here we consider the Raman scattering as a possible probe to detect the OW in titanates and vanadates with the triply degenerate $t_{2g}$ orbitals. It is considered the inter-site scattering processes where OW’s are excited through the electronic exchange processes between the NN TM sites.\textsuperscript{37,38} This is attributed to the fact that the lowest electronic excitations in titanates and vanadates occur across the Mott-Hubbard gap, unlike the manganites where the electronic excitations across the charge-transfer gap dominate the excitation of OW. Depending on the types of OO, there are the following two scattering processes: (i) The two-orbiton scattering: Consider a pair of the NN TM sites where the occupied orbitals are different and the pure orbitals (Fig. 3), such as the OO in vanadates. Through the second order processes of the interaction between photons and electrons, electrons at the two sites are exchanged and, at the final state, the occupied orbitals are changed at both the sites. This is the analogous to the two magnon Raman scattering in the antiferromagnets. Another two-orbiton process occurs in a pair of the NN TM site where the same kind of orbitals are occupied. In the intermediate state of the scattering process, where two electrons occupy the same orbital at a site, the occupied orbital is changed due to the pair-hopping interaction. At the final state, the two orbitons are created. (ii) The one-orbiton scattering: When the electrons occupy the so-called mixed orbital such as that in YTiO$_3$, the electron hops from one orbital ($\gamma$) to the different orbital ($\gamma’$) in its NN TM site. When this electron comes back to the orbital $\gamma’$ in the initial site, one orbiton is excited.

There are alternate two scattering processes from OW where one orbiton is created at a TM site. (i) An electron is excited from an orbital $\gamma$ to $\gamma’$ at the same site associated with a creation of odd-parity phonons. Then, these phonons are annihilated by emitting a photon. Such kinds of the Frank-Condon processes have been considered for the orbiton+phonon excitation in the optical conductivity spectra, and for the multiphonon excitation in the Raman spectra.\textsuperscript{39} The total scattering-cross section ratio of this process to the inter-site process is of the order of $10^{-3}$ to $10^{-2}$. (ii) The incident photon excites an electron from the 3$d$ $\gamma$ orbital to one of the 4$p$ orbitals at the same site. Then, this electron is relaxed to the 3$d$ $\gamma’$ orbital by emitting a photon. This scattering-cross section ratio to the inter-site process is estimated for titanates and vanadates to be of the order of $10^{-1}$.

Here, we calculate the Raman spectra for the inter-site scattering process. It is supposed that this process provides the main contribution for the OW scattering, in particular, in the two-orbiton energy regions. The energy, momentum and polarization of initial (scattered) photon are $\omega_i(\omega_f)$, $\vec{k}_i(\vec{k}_f)$ and $\lambda_i(\lambda_f)$, respectively. The differential scattering cross section from OW is given as

![FIG. 2. Dispersion relations of OW in the $(d_y(x+z)/d_y(x-z)/d_x(y+z)/d_x(y-z))$-type OO for YTiO$_3$. The bold and broken lines are for the FM and paramagnetic states, respectively. The parameter values are chosen to be $I/U’=0.125$, $g_E Q_E/J_0=g_I Q_I/J_0=1.2$. The Brillouin zone for the tetragonally symmetric is adopted.](#)

![FIG. 3. Scattering processes in the Raman scattering. (a) and (b) The two-orbiton scattering processes in the pure OO states. (c) The one-orbiton scattering processes in the mixed OO state.](#)
Eqs. (3) and (15) concerning with a bond connecting site \( V \) by the matrix \( \Gamma \) in the Holstein-Primakoff boson operators represented by the intermediate states. The exchange interaction \( \delta H_{\text{ond}} \) extends processes of the interactions between photons and site \( S \left( r, \lambda, y \right) \). Both the incident and scattered photon polarizations are parallel to the \( z \) axis. Other parameter values are the same as those in Fig. 1.

\[
\frac{d^2 \sigma}{d\Omega dE_J} = \sigma_T \frac{m a^2}{\hbar^2} \left( \frac{m a^2}{\hbar^2} \right)^2 \frac{1}{2 \pi \hbar} \int dt e^{i\hbar(\omega_J - \omega)t} \times \sum \psi_{i} \langle \omega \rangle_x \langle \psi_{i} \rangle_y
\]

(22)

with \( \sigma_T = (e^2/mc^2)^2 \) and a bond length \( a \). \( \psi_{i} \) is the polarization factor given by

\[
\psi_{i} = (\hat{e}_{k_i,\lambda_i})_I (\hat{e}_{k_i,\lambda_i})_F (\hat{e}_{k_j,\lambda_j})_I (\hat{e}_{k_j,\lambda_j})_F \quad \text{(23)}
\]

and \( S(t) \) is the dynamical correlation function defined by

\[
S(t) = \langle K(t) K(0) \rangle
\]

(24)

with

\[
K(t) = \sum \Gamma \sum \alpha \beta |H_T(i, i + \delta t)|
\]

(25)

\( H_T(i, i + \delta t) \) is a term of the effective Hamiltonian given in Eqs. (3) and (15) concerning with a bond connecting site \( i \) and site \( i + \delta t \). Here, \( t = T_1, T_2, A_1, E \) classifies the intermediate states. The exchange interaction \( J_T = \frac{T^2}{\Delta E_T} \) in \( H_T \) is replaced by \( \frac{T^2}{\Delta E_T} - (\omega - \omega_f) \) in \( H_T(i, i + \delta t) \). This expression is obtained from the second order processes of the interactions between photons and electronic currents between the NN TM sites.

We introduce the orbiton operators \( \psi(\tilde{k}) \) with the energies \( E(\tilde{k}) \) which are obtained by diagonalizing the Hamiltonian represented by the Holstein-Primakoff boson operators \( \psi(\tilde{k}) \). This Bogoliubov transformation is given by the matrix \( V(\tilde{k}) \) as

\[
\tilde{\psi}_\alpha(\tilde{k}) = \sum \psi_\beta(\tilde{k}) V_{\alpha\beta}(\tilde{k}).
\]

(26)

\( \tilde{\psi}(\tilde{k}) \) has \( 2N \) components in the system where the number of the OW mode is \( N \), and \( \tilde{\psi}_{\alpha}(\tilde{k}) \) is the creation operator with the condition \( \tilde{\psi}_{\alpha}(-\tilde{k}) = \tilde{\psi}_{\alpha}(-\tilde{k})^\dagger \). For example,

\[
\tilde{\psi}(\tilde{k}) = \{ a_1(\tilde{k}), \ldots a_N(\tilde{k}), a_1^\dagger(-\tilde{k}), \ldots a_N^\dagger(-\tilde{k}) \}
\]

(27)

By using the operators, we obtain

\[
\mathcal{K} = \sum \tilde{\psi}_\alpha(\tilde{k})^\dagger h_{\alpha\beta}(\tilde{k}) \tilde{\psi}_\beta(\tilde{k})
\]

\[
+ \frac{1}{2} \sum \delta \left( g_{\alpha}(\tilde{k}) \psi_\alpha(0) + g_{\alpha}^\dagger(\tilde{k}) \psi_\alpha(0) \right)
\]

(28)

where \( h_{\alpha\beta}(\tilde{k}) \) and \( g_{\alpha}(\tilde{k}) \) are the coefficients. In the pure OO states, the second term vanishes, i.e. the one-orbiton scattering is prohibited. Then the Fourier transform of the dynamical correlation function in the two-orbiton Raman scattering is given by

\[
S^{ll}(\omega) = N \sum \tilde{\psi}_\alpha(\tilde{k})^\dagger h_{\alpha\beta}(\tilde{k}) \tilde{\psi}_\beta(\tilde{k})
\]

\[
+ \frac{1}{2} \sum \delta \left( g_{\alpha}(\tilde{k}) \psi_\alpha(0) + g_{\alpha}(\tilde{k})^\dagger \psi_\alpha(0) \right)
\]

(29)

We neglect the orbiton-orbiton interaction for simplicity, and \( S^{ll}(\omega) \) is represented by the convolution of the two OW modes. On the other hand, in the one-orbiton scattering, the dynamical correlation function reflects the OW at the momentum \( \tilde{k} = 0 \) as

\[
S^{ll}(\omega) = 4N \sum g_{\alpha}(\tilde{k}) \delta (\omega - E_\alpha(\tilde{k}))
\]

(30)

In Fig. 4, the Raman spectra by OW in the (S-C/O-G) phase for LaVO\(_3\), and in the (S-G/O-C) phase for YVO\(_3\) (the low temperature phase) are presented. The two-orbiton scattering processes are considered. In spite of this processes, a sharp peak structure appears at the lower edge of the continuum. This is attributed to the one-dimensional character of the OW and the factor \( h_{\alpha\beta}(\tilde{k}) \) in Eq. (28) which enhances the lower edge. As for the selection rule, the Raman scattering is only active for the (zz) polarization where both the incident and scattered light polarizations, \( \hat{e}_{k_i,\lambda_i} \) and \( \hat{e}_{k_j,\lambda_j} \), are parallel to the \( z \) axis. Through the interaction with the \( z \)-polarized lights, two electrons are exchanged between the \( dxz \) and \( d_{xz} \) orbitals in the NN sites along the \( z \) axis. We mention that the local modes discussed in the previous section, i.e. the \( d_{xy} \to d_{yz} \) and \( d_{xy} \to d_{xz} \) excitations, are not detected by the Raman scattering, since the exchange processes do not occur between the \( d_{xy} \) and \( d_{xz} \) orbitals. In Fig. 5, we show the Raman spectra.
and polarization section is given by scattered neutron with momentum scattering cross section in the scattering of initial (scattered) neutron. Formulate the differential equations for our present interest, it is possible to detect by the function is chosen to be 0.25 \( J_0 \). The polarizations of the incident and scattered neutrons are chosen to be parallel to the \( z \) axis. This originates from the one-orbiton scattering and the joint density of states of one-orbiton scattering, and the reciprocal vector is \( \vec{G} = (000) \). Other parameter values are the same as those in Fig. 2.

from OW in the \( (d_x(x+z)/d_y(x+z)/d_z(x+z)/d_x(y-z)/d_z(y-z)) \)-type OO. The one-orbiton scattering is considered in the calculation. The spectra being active for the \( (xx) \) and \( (x'y') \) polarizations are the \( A_{1g} \) modes, and those for the \( (xx') \) and \( (x'y') \) ones are the \( B_{1g} \) ones. Here, the \( x \), \( y \), and \( z \) directions are chosen to be parallel to the TM-O bonds, and \( x' = \frac{1}{\sqrt{2}}(x+y) \) and \( y' = \frac{1}{\sqrt{2}}(-x+y) \). It is worth noting that all modes are inactive for the \( (zz) \) polarization in contrast to the case of vanadates. This originates from a cancellation from the one-orbiton scattering contributions from site \( i \) and its NN site \( i + \delta_z \) along the \( z \) direction where the occupied orbitals have the mirror symmetry in terms of the \( xy \) plane between the two.

B. inelastic neutron scattering

Although the Raman scattering is a possible probe to detect OW as shown in the previous section, its observation is limited to be the OW at zero momentum in the one-orbiton scattering, and the joint density of states of OW in the two-orbiton scattering. In the \( t_{2g} \) OO systems of our present interest, it is possible to detect by the inelastic neutron scattering. Formulate the differential scattering cross section in the scattering of initial (scattered) neutron with momentum \( \vec{k}_i(k_f) \), energy \( \omega_i(\omega_f) \), and polarization \( l_i(l_f) (= x, y, z) \). The scattering cross section is given by

\[
\frac{d\sigma}{d\Omega d\omega_f} = \left( \frac{\gamma c^2}{m_N c^2} \right)^2 \frac{1}{2} g F(\vec{K})^2 \frac{k_f}{k_i} \times \sum_{l_i,l_f} \left( \delta_{l_i l_f} - \kappa_i \kappa_f \right) S^{l_i l_f}(\vec{K}, \omega),
\]

with \( \vec{K} = \vec{k}_i - \vec{k}_f \), \( \omega = \omega_i - \omega_f \), and \( \kappa = \vec{K}/|\vec{K}| \). \( S^{l_i l_f}(\vec{K}, \omega) \) is the Fourier transform of the dynamical correlation function for the angular momentum operators \( L^l_{id} \) defined by

\[
S^{l_i l_f}(\vec{K}, \omega) = 2N \sum_{dd'} \sum_{\alpha = 1}^N \delta \left\{ \omega - E_\alpha(\vec{K}) \right\} \times D^{l_i \ast}_{d\alpha} D^{l_f}_{d'\alpha} e^{iG \cdot \delta_{dd'}},
\]

where \( D^{l_i \ast}_{d\alpha} \) is defined by the Fourier transform of the angular momentum operator

\[
O_{d'Tl}(\vec{K}) = \sum_{\alpha} D^l_{da}(\vec{K}) \psi_{\alpha}(\vec{K}).
\]

\( \delta_{dd'} \) is a vector connecting the \( d \)- and \( d' \)-th TM ions in the same cell, and \( G \) is the reciprocal lattice vector. In contrast to the Raman scattering, the momentum dependence of the dispersion relation is detectable. The magnitude of the scattering intensity is expected to be the same order with that for the magnetic neutron scattering in magnets with \( L = 1 \).
YTiO
also seen in the contour map of the scattering intensity in fied by utilizing the polarized neutron scattering. This is shown in Fig. 6). Therefore, the modes of OW are identi-
z\( (S\text{-}C/O\text{-}G)\) and \((S\text{-}G/O\text{-}C)\) phases for LaVO
for YVO
3
in these systems can be detected by utilizing the Raman
spectra. This is attributed to the or-
ther experiments in
particular, this is remark-
ably seen in vanadates where the so-called pure orbitals are ordered. Thus, the selection rules for the Raman and neutron scatterings are strict. For example, the two-orbiton scatterings with the \( z \) polarized photons dominate the Raman spectra. This is attributed to the orthogonality of the electron transfer integral between the NN vanadium sites. In the actual vanadates, there is the GdFeO
3
-type lattice distortion which may make the one-orbiton scatterings possible. In the recent Raman scat-
tering experiments in RVO
3
, a new peak appears around 60meV in the \((S\text{-}C/O\text{-}G)\) phase.\(^{40}\) It is confirmed that this peak is active in the \((zz)\) polarization configuration. We expect that this peak originates from OW excited by the two-orbiton scattering processes as discussed in Sec. III (Fig. 4).

In the case of YTiO
3
, the dispersion relation of OW and the Raman and neutron scattering spectra are more complicated than those in vanadates. This is because of the mixed OO state with the four different orbitals in a single unit cell. The present results are also distinct from those proposed in Ref. 18; the orbital excitations are exam-
ined in the OO states with high symmetry being different from the \( (d_y(x+z))/d_y(x-z)/d_z(x+z)/d_z(x-z)\)-type OO and incompatible with the crystal symmetry of YTiO
3
. In the present results, as shown in Fig. 2, it is found, in contrast to the previous results,\(^{18}\) that there are the anisotropy of the dispersion relations in the \( xy\) plane and along the \( z\) axis, the two kind groups of the OW with higher and lower energies appear, and the flat bands are not seen along the \((\pi\pi\pi)-(\pi\pi0)\) direction. Actually, the inelastic neutron scattering experiments have started in YTiO
3
.\(^{41,17}\) The detailed comparison between the theo-
retical calculations and the experimental data can reveal nature of OW as well as that of OO in the \( t_{2g} \) orbital systems.

V. SUMMARY AND DISCUSSION

In this paper, a theory of OW in perovskite titanates and vanadates with the triply degenerate \( t_{2g} \) orbitals is present. We examine the dispersion relations of OW in the \((S\text{-}C/O\text{-}G)\)- and \((S\text{-}G/O\text{-}C)\) phases for LaVO
3
 and YVO
3
 (the low temperature phase), respectively and that in the \( (d_y(x+z))/d_y(x-z)/d_z(x+z)/d_z(x-z)\)-type OO for YTiO
3
. We demonstrated that characteristics of OW in these systems can be detected by utilizing the Raman and inelastic neutron scatterings.

In comparison with the OW in the manganites where the \( e_g \) orbital is ordered, both the excitation spectra and the observation methods are distinct qualitatively in the present \( t_{2g} \) orbital system. In particular, this is remark-
ably seen in vanadates where the so-called pure orbitals are ordered. Thus, the selection rules for the Raman and neutron scatterings are strict. For example, the two-orbiton scatterings with the \( z \) polarized photons dominate the Raman spectra. This is attributed to the orthogonality of the electron transfer integral between the NN vanadium sites. In the actual vanadates, there is the GdFeO
3
-type lattice distortion which may make the one-orbiton scatterings possible. In the recent Raman scat-
tering experiments in RVO
3
, a new peak appears around 60meV in the \((S\text{-}C/O\text{-}G)\) phase.\(^{40}\) It is confirmed that this peak is active in the \((zz)\) polarization configuration. We expect that this peak originates from OW excited by the two-orbiton scattering processes as discussed in Sec. III (Fig. 4).

In the case of YTiO
3
, the dispersion relation of OW and the Raman and neutron scattering spectra are more complicated than those in vanadates. This is because of the mixed OO state with the four different orbitals in a unit cell. The present results are also distinct from those proposed in Ref. 18; the orbital excitations are exam-
ined in the OO states with high symmetry being different from the \( (d_y(x+z))/d_y(x-z)/d_z(x+z)/d_z(x-z)\)-type OO and incompatible with the crystal symmetry of YTiO
3
. In the present results, as shown in Fig. 2, it is found, in contrast to the previous results,\(^{18}\) that there are the anisotropy of the dispersion relations in the \( xy\) plane and along the \( z\) axis, the two kind groups of the OW with higher and lower energies appear, and the flat bands are not seen along the \((\pi\pi\pi)-(\pi\pi0)\) direction. Actually, the inelastic neutron scattering experiments have started in YTiO
3
.\(^{41,17}\) The detailed comparison between the theo-
retical calculations and the experimental data can reveal nature of OW as well as that of OO in the \( t_{2g} \) orbital systems.

ACKNOWLEDGMENTS

Author would like to thank S. Maekawa, N. Nagaosa, G. Khalilullin, T. Hatakeyama, and S. Okamoto for their valuable discussions, and also thank Y. Tokura, S. Miyasaka, S. Shamoto, and S. Sugai for providing unpublished experimental data. This work was supported by KAKENHI from MEXT, and KURATA foundation. Part of the numerical calculation has been performed by the supercomputing facilities in IMR, Tohoku University.

1 See, for a review, M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. 70, 1039 (1998).
2 Y. Tokura and N. Nagaosa, Science 288, 462 (2000).
3 M. Cyrot, and C. Lyon-Caen, Jour. Physique 36, 253 (1975).
4 A. G. Komarov, and L. I. Korovin, and E. K. Kudinov, Sov. Phys. Solid State 17, 1531 (1975).
5 S. Ishihara, J. Inoue, and S. Maekawa, Phys. Rev. B 55, 8280 (1997).
6 E. Saitoh, S. Okamoto, K. Tobe, K. Yamamoto, T. Kimura, S. Ishihara, S. Maekawa, and Y. Tokura, Nature 410, 180 (2001).
7 M. Gruninger, R. Ruckamp, M. Windt, P. Reutler, C. Zoel, T. Lorenz, A. Freimuth, and A. Revcolevshi, Nature 418, 39 (2002).
8 E. Saitoh, S. Okamoto, K. Tobe, K. Yamamoto, T. Kimura, S. Ishihara, S. Maekawa, and Y. Tokura, Nature 418, 40 (2002).
9 J. Itoh, M. Tsuchiya, H. Tanaka, and K. Motoya, Jour. Phys. Soc. Jpn. 71, 2086 (2002).
10 M. v. Zimmermann, J. P. Hill, D. Gibbs, S. Ishihara, Y. Taguchi, and Y. Tokura, Phys. Rev. B 66, 184419 (2002).
11 M. Itoh, S. Tsuchiya, H. Tanaka, and K. Motoya, Jour. Phys. Soc. Jpn. 68, 2783 (1999).
12 H. Ichikawa, J. Akimitsu, M. Nishi, and K. Kakurai, Physica B 281&282, 482 (2000).
13 D. A. MacLean, H.-N. Ng, and J. E. Greedan, Jour. Sol. Stat. Chem. 30, 35 (1979).
14 S. Ishihara, T. Hayakeyama, and S. Maekawa, Phys. Rev. B 65, 064442 (2002).
15 T. Mizokawa, and A. Fujimori, Phys. Rev. B 54, 5368 (1996), and T. Mizokawa, D. I. Khomskii, and G. A. Sawatzky, Phys. Rev. B 60, 7309 (1999).
16 H. Sawada, N. Hamada, K. Terakura, and T. Asada, Phys. Rev. B 53, 12742 (1996), and H. Sawada, and K. Terakura, Phys. Rev. B 58, 6831 (1998).
17 M. Machizuki, and M. Imada, Jour. Phys. Soc. Jpn. 69, 1982 (2000), ibid 70, 1777 (2001).
18 G. Khaliullin, and S. Okamoto, M. Reehuis, A. Ivanov, H. He, Y. Taguchi, Y. Tokura, and B. Keimer, Phys. Rev. Lett. 89, 167202 (2002).
19 G. Khaliullin, and S. Okamoto, Phys. Rev. Lett. 89, 167201 (2002).
20 S. Miyasaka, Y. Okimoto, and Y. Tokura, Jour. Phys. Soc. Jpn. 71, 2086 (2002).
21 S. Miyasaka, Y. Okimoto, M. Iwama, and Y. Tokura, Phys. Rev. B 68, 100406 (2003).
22 P. Bordet, C. Chaillout, M. Marezio, Q. Huang, A. Santro, S. -W. Cheong, H. Takagi, C. S. Oglesby, and B. Batlogg, Jour. Sol. Stat. Chem. 106, 253 (1993).
23 H. Kawano, H. Yoshizawa, and Y. Ueda, Jour. Phys. Soc. Jpn. 63, 2857 (1994).
24 M. Noguchi, A. Nakazawa, S. Oka, T. Arima, Y. Wakabayashi, H. Nakao, and Y. Murakami, Phys. Rev. B 62, R9271 (2000).
25 G. R. Blake, T. T. M. Palstra, Y. Ren, A. A. Nugroho, and A. A. Menovsky, Phys. Rev. Lett. 87, 245501 (2001).
26 C. Ulrich, G. Khaliullin, J. Sirker, M. Reehuis, M. Ohl, S. Miyasaka, Y. Tokura, and B. Keimer, (unpublished) cond-mat/0211589.
27 G. Khaliullin, P. Horsch, and A. M. Olés, Phys. Rev. Lett. 86, 3879 (2001), and J. Sirker, and G. Khaliullin, Phys. Rev. B 67, 100408 (2003).
28 Y. Motome, H. Seo, Z. Fang, and N. Nagaosa, Phys. Rev. Lett. 90, 146602 (2003).
29 Z. Fang, N. Nagaosa, and K. Terakura, Phys. Rev. B 67, 035101 (2003).
30 T. H. De Silva, A. Joshi, M. Ma, and F. C. Zhang, (unpublished) cond-mat/03024989.
31 M. Gell-Mann, and Y. Ne’emam. The eightfold way, (Benjamin, New York, 1964).
32 K. I. Kugel, and D. I. Khomskii, Sov. Phys. Solid, State, 17, 285 (1975).
33 K. Kikoin, O. Entin-Wohlman, V. Fleurov, and A. Aharony, Phys. Rev. B 67, 214418 (2003).
34 M. Itoh, H. Adachi, H. Nakao, Y. Murakami, Y. Taguchi, Y. Tokura, K. Kato, E. Nishihori, M. Takata, M. Sakata, H. Miyagawa, S. Nanao, H. Maruyama, E. Arawaka, and K. Namikawa, Meeting Abstract of the Physical Society of Japan, 2001 Autumn Meeting, 56, Part 3, 341 (2001).
35 D. Janssen, R. V. Jolos, and F. Donau, Nucl. Phys. A 224, 93 (1974).
36 A. Klein, and E. R. Marshalek, Rev. Mod. Phys. 63, 375 (1991).
37 J. Inoue, S. Okamoto, S. Ishihara, W. Koshibae, Y. Kawamura, and S. Maekawa, Physica B 237-238, 51 (1997).
38 S. Okamoto, S. Ishihara, and S. Maekawa, Phys. Rev. B 66, 104435 (2001).
39 P. B. Allen, and V. Pereveinos, Phys. Rev. Lett. 83, 4828 (1999), and V. Pereveinos, and P. B. Allen, Phys. Rev. B 64, 085118 (2001).
40 S. Miyasaka, Y. Okimoto, and Y. Tokura (unpublished).
41 S. Shamato, F. Iga, M. Tsubota, T. Kajitani, ISIS Experimental Report, RB Num.13662, Date of Report 03/03/03, MARI.