Orbital Liquid in Three-Dimensional Mott Insulator: $LaTiO_3$

G. Khaliullin[1,2] and S. Maekawa[2]

[1]*Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany*
[2]*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

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We present a theory of spin and orbital states in Mott insulator $LaTiO_3$. The spin-orbital superexchange interaction between $d^1(t_{2g})$ ions in cubic crystal suffers from a pathological degeneracy of orbital states at the classical level. Quantum effects remove this degeneracy and result in the formation of the coherent ground state, in which the orbital moment of t_{2g} level is fully quenched. We find a finite gap for orbital excitations. Such a disordered state of local degrees of freedom on unfrustrated, simple cubic lattice is highly unusual. Orbital liquid state naturally explains observed anomalies of $LaTiO_3$.

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Experiments on transition metal oxides continue to challenge the theory — Keimer *et al.* [1] have recently reported on dynamical quenching of t_{2g} orbital angular moments in Mott insulator $LaTiO_3$ which is not associated with any detectable orbital/Jahn-Teller ordering. More specifically, magnon spectra are found to fit a spin one-half, nearly isotropic Heisenberg model on cubic lattice. At the same time, an anomalous x-ray scattering study has not revealed any evidence of static orbital order. These observations indicate that t_{2g} orbital degrees of freedom of Ti^{3+} ions are mysteriously missing in low energy states of $LaTiO_3$. Keimer *et al.* suggested a picture of strongly fluctuating orbitals to reconcile their results.

Dynamical quenching of local orbital moments in a periodic, simple cubic lattice formed by Ti^{3+} ions in $LaTiO_3$ is fascinating. This poses a serious problem for the canonical Goodenough-Kanamori picture of successive orbital and magnetic orderings — the guiding idea in the modern theory of orbitally degenerate transition metal oxides (see, for review, [2,3]). Loosely speaking, a lifting of the orbital degeneracy without symmetry breaking is formally similar to the spin moment quenching in periodic Kondo lattices. The novelty of $LaTiO_3$ is however that it is an insulator. At the same time, the ideas of quantum disorder due to the weak connectivity or geometrical frustration (as in pyrochlore systems, see [4], for instance) do not apply here either.

As discussed below, there are several physical reasons for the orbital disorder in $LaTiO_3$, and the key factor is a special symmetry of t_{2g}-superexchange interaction on cubic lattice. This results in a quantum resonance between orbital levels, which removes degeneracy present at the classical level by the formation of resonating bonds in the orbital sector — a realization of Anderson's resonating valence bonds idea [5] in a three-dimensional (3D) insulator with help of orbital degrees of freedom. As for spin moments, they show a long-range order as expected on general grounds. However, a staggered moment is considerably reduced, since short-wavelength magnons are actively involved in the local resonance of exchange bonds — in fact, it is the composite spin-orbital excitation which plays a crucial role in the theory presented.

We begin with discussion of the t_{2g}-superexchange interaction. In terms of fermions $a_{i,\sigma}$, $b_{i,\sigma}$, $c_{i,\sigma}$ corresponding to t_{2g} levels of yz, zx, xy symmetry, respectively, the hopping term in the Hubbard model reads as $-t(a^\dagger_{i,\sigma}a_{j,\sigma} + b^\dagger_{i,\sigma}b_{j,\sigma} + \text{H.c.})$ for the bonds along the c direction in a cubic crystal. Correlation energies in doubly occupied virtual states of Ti^{3+} ion are specified as follows: U for electrons on the same orbital; $U' + J_H$, if they occupy different orbitals and form a spin singlet; and $U' - J_H$ for a spin-triplet charge excitation. The relation $U' = U - 2J_H$ holds in the atomic limit. As $J_H \ll U$ usually, the dominant part of the superexchange interaction in a cubic lattice is

$$H_{\rm SE} = -\frac{4t^2}{U} + \sum_{\langle ij\rangle}\left(\mathbf{S}_i\mathbf{S}_j + \frac{1}{4}\right)\hat{J}^{(\gamma)}_{ij}. \qquad (1)$$

On c-axis bonds the orbital structure of the exchange "constant" is given by (hereafter, the energy scale $4t^2/U$ is used)

$$\hat{J}^{(c)}_{ij} = n_{i,a}n_{j,a} + n_{i,b}n_{j,b} + a^\dagger_i b_i b^\dagger_j a_j + b^\dagger_i a_i a^\dagger_j b_j\,. \qquad (2)$$

Similar expressions can be obtained for $\hat{J}^{(a)}_{ij}$ and $\hat{J}^{(b)}_{ij}$ by replacing orbital fermions a, b in this equation by b, c and c, a pairs, respectively. We notice that in Eq. (2) and below fermions have only orbital quantum number (a, b, c orbitons), since a spin component of the original fermions is represented in Eq. (1) by spin one-half operators, as usual. We shall focus now on the Hamiltonian (1), and discuss later the effects of finite Hund coupling corrections (of the order of J_H/U).

As evident from Eq. (1) (consider $\langle \mathbf{S}_i\mathbf{S}_j\rangle = -1/4$), the classical Néel state is infinitely degenerate in the orbital sector, reflecting the fact that cubic symmetry is respected by the Néel state. This emphasizes a crucial importance of quantum effects, as has been pointed out first in the context of e_g systems [6]. The bond directional geometry of e_g orbitals offers a solution [7] in that case: e_g orbital frustration is resolved by order from disorder mechanism selecting a particular (directional) orbital configuration, which maximizes the energy gain from quantum spin fluctuations. t_{2g}

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orbitals are however not bond oriented (they are all planar), and not much spin fluctuation energy can be gained by any pattern of static orbital orderings. The solution of the t_{2g} problem proposed here is different: that is an idea of SU(4) spin-orbital excitations recently discussed in a context of one-dimensional models [8–10].

Reflecting the geometry of t_{2g} orbitals, every bond in Eq. (1) is represented by two equivalent orbitals (such a symmetry is absent for Ising-like e_g orbitals). This is also evident from the representation of Eq. (2) in terms of orbital pseudospin $\boldsymbol{\tau}^i_{ab}$ and density $n^i_{ab} = n_{ia} + n_{ib}$ operators acting on the (a,b)-doublet subspace: $\hat{J}^{(c)}_{ij} = 2(\boldsymbol{\tau}^i_{ab}\boldsymbol{\tau}^j_{ab} + \frac{1}{4}n^i_{ab}n^j_{ab})$. Suppose that $n^i_{ab} = 1$; one then clearly observes an orbital SU(2) symmetry of exchange "integrals." As we know from spin SU(2) $\times$ orbital SU(2) models [8–10], the exchange energy is gained in that case due to the resonance between degenerate local configurations (spin singlet $\times$ orbital triplet, and spin triplet $\times$ orbital singlet), and elementary excitations are mixed spin-orbital SU(4) modes. Figure 1(a) illustrates the idea of correlated spin-orbital fluctuation. In the present problem, spins and orbitals are different: the spin sector is effectively half filled ($n_{i,\uparrow} + n_{i,\downarrow} = 1$) and must show a long-range order in 3D; on the other hand, the orbital occupation number n^i_{ab} is not conserved but rather fluctuates around the average value $2/3$. Because of the presence of a third orbital, one has a less severe constraint $n_{i,a} + n_{i,b} + n_{i,c} = 1$ in the orbital sector. Even though a true SU(4) excitation cannot develop here because of spin-orbital asymmetry, the analogy with 1D models suggests the way how to optimize the exchange energy: That is by the formation of a virtual SU(4) resonance which can be viewed in the present context as a local excitation composed of magnon and orbital fluctuation. This resonance removes the orbital frustration dynamically, and the disordered state of orbitals is precisely what is required to amplify this mechanism in all three directions of cubic lattice.

Technically, the main ingredient of the theory is the three-particle bond variable, $\hat{\chi}^{(ij)}_\alpha = \alpha^\dagger_i \alpha_j (s^\dagger_i + s_j)$. Here, α denotes an appropriate fermionic orbiton, say a or b orbitons for the c-bond pair (ij), with $s^\dagger$ being a magnon excitation about the Néel state. This is an analogy to the SU(4) flavor bond variable [8] in a spin ordered state. The bare magnon dispersion is simply given by the Heisenberg interaction with an average exchange constant $J = \langle \hat{J}^{(\gamma)}_{ij} \rangle$, which will be calculated below. On the contrary, the orbital dynamics is exclusively due to the three-particle resonance, since the spin-only prefactor $\langle \mathbf{S}_i \mathbf{S}_j + 1/4 \rangle$ is almost zero (~ -0.05) in 3D. We may present Eq. (1) as $H_{\rm SE} = H_{\rm sp} + H_{\rm int} + \text{const}$, where $H_{\rm sp}$ describes bare magnons, and $H_{\rm int} = -\frac{1}{2}\sum_{\langle ij \rangle}\sum_{\alpha\beta} \hat{\chi}^{(ij)}_\alpha \hat{\chi}^{(ji)}_\beta$. The strategy is then to decouple $H_{\rm int}$ in terms of bond variables. The solution with uniform bond amplitude χ gives $H_{\rm int} \Rightarrow H_a + H_b + H_c + \text{const}$, where

$$H_a = -\chi \cdot \sum_{\langle ij \rangle_{bc}} \{ a^\dagger_i a_j (s^\dagger_i + s_j) + \text{H.c.} \}. \qquad (3)$$

Here the summation is restricted to (b,c) planes. Similar expressions hold for b and c orbitons. Since the filling factor is only $1/3$ per orbital flavor, the orbital sector is far from nesting conditions, thus a mean-field uniform solution in the spirit of large N theories [11] is believed to be a reasonable starting point to describe a disordered state of orbitals. Equation (3) describes an effective hopping of orbitons accompanied by simultaneous magnon excitations. Thus, we have mapped the superexchange in an undoped system to an effective χ-J model, which in a momentum space reads as

$$H_{\chi\text{-}J} = \sum_{\mathbf{kp}} \sum_\alpha M^{(\alpha)}_{\mathbf{kp}} (\alpha^\dagger_{\mathbf{k}+\mathbf{p}} \alpha_{\mathbf{k}} s_{\mathbf{p}} + \text{H.c.}) + \sum_{\mathbf{p}} \omega_{\mathbf{p}} s^\dagger_{\mathbf{p}} s_{\mathbf{p}} . \qquad (4)$$

Here $M^\alpha_{\mathbf{kp}} = -4\chi(\gamma^{(\alpha)}_{\mathbf{k}+\mathbf{p}} u_{\mathbf{p}} + \gamma^{(\alpha)}_{\mathbf{k}} v_{\mathbf{p}})$, while form factors are $\gamma^{(a)}_{\mathbf{k}} = (c_y + c_z)/2$, $\gamma^{(b)}_{\mathbf{k}} = (c_z + c_x)/2$, $\gamma^{(c)}_{\mathbf{k}} = (c_x + c_y)/2$, where $c_\lambda = \cos k_\lambda$. Bare magnon dispersion $\omega_{\mathbf{p}} = 3J(1 - \gamma^2_{\mathbf{p}})^{1/2}$ with $\gamma_{\mathbf{p}} = (c_x + c_y + c_z)/3$, and $u_{\mathbf{p}}, v_{\mathbf{p}}$ are conventional Bogoliubov transformation coefficients for magnons in the cubic lattice. It is noticed that the coupling constant in Eq. (4) vanishes at the $p = 0$ limit (spin conservation is respected), and it is the short-wavelength magnons which are important. The model is similar to the t-J model for the doped Néel state of cuprates (see [12,13] for comparison), yet the parameters χ and J have to be self-consistently determined. It is noticed that there are three branches of 2D fermions, and the dimensionality of the magnon sector is 3 in the present case.

We calculate first the fermionic spectrum within a self-consistent Born approximation. Different from holes in the t-J model, an orbiton creates a magnon on the site it arrives at, and eliminates a magnon while leaving the site [see Eq. (3)]. Therefore orbiton motion contains a coherent component even in the Ising limit for magnons. The latter approximation is used to simplify a momentum integration in the self-energy, Fig. 1(b). One then obtains

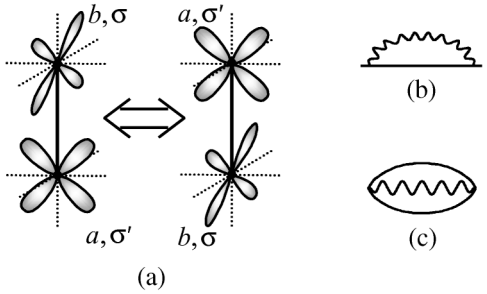

FIG. 1. (a) Spin-orbital resonance between two (left and right) states of the c-bond pair. (b) Self-consistent Born approximation for the fermionic orbital self-energy. (c) Diagram contributing to the bond order parameter χ. Solid (wavy) lines represent orbitons (magnons).

$\mathrm{Re}\Sigma^{(\alpha)}_{\omega}(\mathbf{k}) = f_\omega - \kappa_\omega(\gamma^{(\alpha)}_{\mathbf{k}})^2$. Here $\kappa_\omega = (4\chi)^2 \times \int_0^\infty d\xi\, \rho(\xi)/(\xi + 3J - \omega)$, where $\rho(\xi) = \langle \rho_{\mathbf{k}}(\xi)\rangle_{\mathbf{k}}$ is the fermionic density of states (DOS). f_ω has a similar structure: $f_\omega = (4\chi)^2 \int_{-\infty}^0 d\xi\, \tilde{\rho}(\xi)/(\omega + 3J - \xi)$, where $\tilde{\rho}(\xi) = \langle \rho_{\mathbf{k}}(\xi)\,(\gamma^{(\alpha)}_{\mathbf{k}})^2\rangle_{\mathbf{k}}$. One observes a two-sublattice structure of the orbiton dispersion (imposed by spin order) as expected from the analogy with the t-J model [13]. We estimate the fermionic (unrenormalized) mass as $m \simeq 1/\kappa_0$, and density of states as $\rho(0) \simeq 1/\pi\kappa_0$. Each orbiton forms its own 2D Fermi surface (FS). Say, for a orbitons the FS consists of two almost circles around $(0, 0)$ and (π, π) points in a (k_y, k_z) plane. Assuming constant DOS within the interval of width $W_{\mathrm{orb}} = 1/\rho(0)$, we find $\kappa_0 \simeq 4\chi/\sqrt{\pi}$, and $W_{\mathrm{orb}} \simeq 4\sqrt{\pi}\,\chi$. W_{orb} is the energy scale of orbital fluctuations [14]. Further, we estimate the bond amplitude $\chi = \langle a_i^\dagger a_j (s_i^\dagger + s_j)\rangle$. In the Ising limit for intermediate magnons, Fig. 1(c) gives $\chi \simeq (4/3\sqrt{\pi})\,\langle(\gamma^{(a)}_{\mathbf{k}})^2\rangle_{\mathrm{FS}}$. For $\langle n^{(a)}\rangle = 1/3$ we estimate $\langle(\gamma^{(a)}_{\mathbf{k}})^2\rangle_{\mathrm{FS}} \simeq 0.3$. Spin stiffness in the present model is controlled by J. The mean-field value $2/9$ for J follows from Eq. (2). This number is reduced to $J \simeq 0.16$ due to the corrections shown in Fig. 2(a). The physics behind this reduction is that SU(4) resonance induces some ferromagnetic component in spin interactions.

Thus, we have fixed basic energy scales (in units of $4t^2/U$): $W_{\mathrm{orb}} \simeq 1.6$ for the orbital sector, and $3J \simeq 0.48$ for magnon bandwidth. The spin stiffness is relatively small because the Pauli principle is relevant only if electrons occupy the same orbital, and that probability is reduced at large orbital degeneracy. The main energy gain stems from magnon-orbital resonance: $E_{\mathrm{int}} = -6\chi^2 \simeq -0.31$. (Spin-only contribution is negligible: $E_{\mathrm{SW}} \simeq -0.02$.) This number should be compared with the energy gain that one has with statically ordered planar orbitals: $\simeq -0.17$.

We discuss now physical consequences of the theory in context of experimental data on $LaTiO_3$ [1]. Observed magnon dispersions of cubic symmetry are very natural in the above picture of orbitals fluctuating faster than magnons. The energy scale $4t^2/U \simeq 100$ meV, required to fit $J_{\mathrm{exp}} = 15.5 \pm 1$ meV, is also reasonable in view of values $U \sim 4$ eV [15], and $t \sim 0.3$ eV [16]. This scale means that orbital fluctuation energy in $LaTiO_3$ is $W_{\mathrm{orb}} \sim 160$ meV.

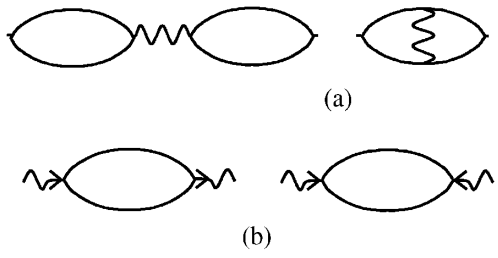

FIG. 2. (a) Interaction corrections to the spin exchange constant J. (b) Magnon self-energies.

Keimer *et al.* observed a very small anisotropy gap, indicating that effects of conventional relativistic spin-orbit coupling are strongly suppressed. This is in fact an unavoidable consequence of our theory, since the ground state has no orbital degeneracy. We may calculate spectral density of orbital angular momentum fluctuations. It is given by the fermionic orbiton excitations ("orbiton Stoner continuum"). Spectral density of the local susceptibility of angular moments $l_i^z = i(a^\dagger b - b^\dagger a)_i$ vanishes in the static, $\omega = 0$ limit: $S(\omega) = 2\rho^2(0)\omega$. This implies complete quenching of orbital moments at low energies. Making parallel with Jahn-Teller (JT) impurity physics [17], we may say that the "effective Ham reduction factor," $\zeta(\bar{\omega}) \sim \{\int_0^{\bar{\omega}} S(\omega)\, d\omega\}^{1/2} \sim \bar{\omega}/W_{\mathrm{orb}}$ is frequency dependent, and the angular moments disappear from low-energy physics linearly in energy. Also, the ratio $\Lambda_{\mathrm{so}}/W_{\mathrm{orb}} \sim 10^{-1}$ ($\Lambda_{\mathrm{so}} \sim 20$ meV [1]) means that relativistic spin-orbit coupling induced corrections are small; we estimate the g-value shift $\Delta g/g \sim 0.06$, consistent with [1].

Despite three dimensionality of magnon spectra, the staggered moment in $LaTiO_3$ is small, i.e., $0.45\mu_B$ [1]—an obvious problem for a spin-wave picture. The present theory resolves this difficulty: The intensity of a spin Bragg peak is partially taken away by a quantum magnon-orbiton resonance and redistributed over the finite frequency region. In other words, fluctuating orbitals generate additional quantum spin fluctuations in the ground state. We have calculated a spin moment reduction in a similar way as it was previously done in the t-J model [18]. Accounting for lowest order interaction corrections [Fig. 2(b)] to the magnons, one finds:

$$\delta S^z_{\mathrm{int}} = \frac{3}{2}(4\chi)^2 \sum_{\mathbf{k}\mathbf{k}'} n_{\mathbf{k}}(1 - n_{\mathbf{k}'}) \times \left\{ \frac{A_{\mathbf{k}\mathbf{k}'}}{(\omega_{\mathbf{p}} + \xi_{\mathbf{k}'} - \xi_{\mathbf{k}})^2} - \frac{B_{\mathbf{k}\mathbf{k}'}}{\omega_{\mathbf{p}}(\omega_{\mathbf{p}} + \xi_{\mathbf{k}'} - \xi_{\mathbf{k}})} \right\}, \tag{5}$$

where $\xi_{\mathbf{k}}$ is the a orbiton dispersion, $\mathbf{k}' = \mathbf{k} + \mathbf{p}$, $A_{\mathbf{k}\mathbf{k}'} = \lambda_+^2/(1 + \gamma_{\mathbf{p}}) + \lambda_+\lambda_-/(1 - \gamma_{\mathbf{p}}^2)^{1/2}$, $B_{\mathbf{k}\mathbf{k}'} = \lambda_+^2\gamma_{\mathbf{p}}/(1 + \gamma_{\mathbf{p}})$, and $\lambda_\pm = \gamma^{(a)}_{\mathbf{k}} \pm \gamma^{(a)}_{\mathbf{k}'}$. By averaging first the matrix elements A, B in Eq. (5) over the Brillouin zone (they are rather regular functions), we evaluate $\delta S^z_{\mathrm{int}} \simeq 0.15$. (Another estimation by using the Ising limit for magnons gives 0.13.) Adding also a conventional 3D spin-wave correction $\delta S^z_{\mathrm{SW}} \simeq 0.075$, one then obtains a staggered moment $0.55\mu_B$ in fair agreement with experiment.

The orbital liquid picture offers a simple explanation of puzzling Fano-type phonon anomalies observed by Raman scattering data in insulating titanates (Reedyk *et al.* [19]). These anomalies are most pronounced in $LaTiO_3$, and we

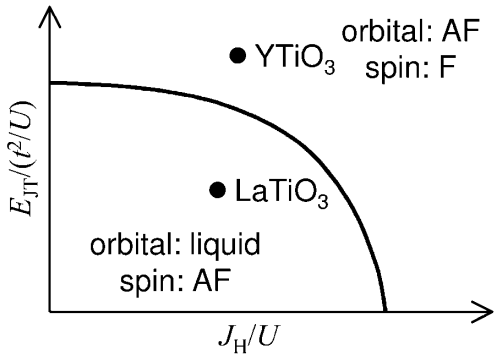

FIG. 3. Proposed phase diagram for the $d^1(t_{2g})$ Mott insulator on cubic lattice. At small Hund's and Jahn-Teller couplings the quantum Néel state with dynamically quenched orbital moments is stabilized. A quantum phase transition line separates this state from the ferromagnetic phase with static orbital/JT order.

identify their origin as due to the coupling of phonons to orbital excitations, noticing that the phonon position ($\sim$300 cm^{-1}) is right in the orbital Stoner continuum.

According to Goodenough-Kanamori rules, local ferrotype orbital correlations are expected in a spin Néel state. Residual interactions (via magnons) between orbitons do indeed produce a triplet pairing of orbitons within each fermionic branch. Treating the problem on the BCS level, we find the finite mean-field order parameter $\langle \alpha_i^\dagger \alpha_j^\dagger \rangle$ of p-wave symmetry (specifically, of $p_x + ip_y$ symmetry for c orbitons, for instance), which opens an orbital gap. It is difficult to calculate reliably the value of this gap (of the order of a few meV); we predict however that a linear γ term in specific heat should be released when a correlation gap in the orbital spectrum is thermally washed out. We estimate $\gamma \sim 40$ mJ/mole K^2. Specific heat measurements in $LaTiO_3$ up to $T \sim 100$ K would be a crucial test for the theory.

From a physical point of view, the proximity of $LaTiO_3$ to Mott transition (charge gap is only 0.2 eV [16]) is actually a key factor, which drives this compound to a superexchange dominated orbital liquid state. This state is stable, however, only if the effective orbiton bandwidth, $W_{\rm orb} \sim 4t^2/U$, is large enough to suppress JT order. When one goes to another end compound, $YTiO_3$ (charge gap $\sim$1 eV), the bandwidth narrows [16], thus reducing the superexchange energy scale, and we expect static JT ordering of spatially more localized orbitals once the ratio $W_{\rm orb}/E_{\rm JT}$ becomes less than critical. With antiferrotype orbital ordering as observed in $YTiO_3$ [20] a classical expectation value of Eq. (2) vanishes, and the Hund's coupling J_H term induces a ferromagnetic state. The competition between the frustrated superexchange on one hand, and JT plus Hund's couplings on the other hand is expected to result in the phase diagram shown in Fig. 3.

To conclude, we have argued that due to large degeneracy and special geometry of orbitals the t_{2g}-superexchange system most likely has an orbitally disordered ground state. The observed anomalies of $LaTiO_3$ find their natural explanations in a proposed orbital liquid picture. In metallic manganites, e_g orbital disorder is enforced by the presence of mobile carriers [21]. Apparently, the case of insulating $LaTiO_3$ is a nice example of the fact that frustrated t_{2g} orbitals can do the same job alone, without doping and well before the Mott transition to a metallic state is reached.

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