Strongly Correlated Electrons on Frustrated Lattices

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Abstract. We review our two recent theoretical works on strongly correlated electrons on typical frustrated lattices. The first topic is about a Mott transition in the single-band Hubbard model on anisotropic triangular lattice, and we discuss a reentrant behaviour of metal-insulator transition, consistent with that in a $\kappa$-type BEDT-TTF salt. The second topic is about heavy fermion behaviour in the vanadium spinel $\text{LiV}_2\text{O}_4$. We study the 3-orbital $t_{2g}$ Hubbard model on the pyrochlore lattice at quarter filling and derive its low-energy effective model. The correlations of spin and orbital degrees of freedom are discussed.

1. Introduction
The effects of frustration have been discussed intensively for many spin systems and some charge orderings, but not for so many itinerant electron systems. Localised nature of degrees of freedom is necessary for frustration, and this is easily understood when the noninteracting electron gas is considered. Whether the lattice is frustrated or not, the ground state is always nondegenerate in the thermodynamic limit and the zero-temperature entropy is zero. The exception is the case that a flat band exists, and as a matter of fact this happens in many frustrated lattices, but this is not the subject of this paper. We discuss strongly correlated electrons on frustrated lattices. Due to strong correlation effects, the double occupancy is suppressed, enhancing low-energy spin fluctuations. Each atom experiences a longer time in single-electron configuration, becoming closer to insulating situation but keeping metallic nature. Spin and charge degrees of freedom are subject to large short-range correlation in real space, as well as orbital degrees of freedom if present. Thus, frustration becomes important as in the spin system, and this is the situation of frustrated metal. We report our recent related works in this paper.

2. Anisotropic triangular lattice Hubbard model
The triangular lattice system is a classic example of frustrated systems, and resonating valence bond state was also discussed in details first for this system [1]. This is a quantum spin liquid without violation of spin rotation symmetry nor of real-space translation symmetry. After a long quest of its realisation in nature, now we have a few real systems which exhibit spin liquid behaviours, and one of them is the organic compound with triangular lattice structure, $\kappa$-(BEDT-TTF)$_2X$ with $X=\text{Cu}_2\text{CN}_3$ [2]. In this material, a pair of BEDT-TTF molecules form a dimer with spin $S=1/2$, and these dimers constitute a triangular lattice. This system is described by the Hubbard model on a triangular lattice. There exist two types of hopping integral depending on bond direction as shown in figure 1(a), but in the case of $X=\text{Cu}_2\text{CN}_3$ the hopping is almost isotropic, $t'/t=1.06$. This ratio is a measure of frustration: $t'/t=1$ and 0 correspond to the highly frustrated regular triangular lattice and the unfrustrated square lattice, respectively.
The \(\kappa\)-type system has a large variety with different X-ions, and one very interesting member is \(\kappa\)-(BEDT-TTF)\(_2\)Cu[N(CN)\(_2\)]Cl [3]. This material shows a reentrant metal-insulator transition (see figure 1(b)), and the ground state at ambient pressure is an antiferromagnetic insulator. Quantum chemistry calculation estimates \(t'/t=0.75\), and the spin frustration is partially relaxed compared with the spin liquid compound \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)CN\(_3\). We investigate this reentrant metal-insulator transition from the viewpoint of interplay of frustration and electron correlation.

The model to study is the half-filled Hubbard model on the triangular lattice with two nearest-neighbour hoppings

\[
H = -t \sum_{\langle i,j \rangle, \sigma} (c_i^\dagger c_j + \text{h.c.}) - t' \sum_{\langle i,j \rangle, \sigma} (c_i^\dagger c_j + \text{h.c.}) + U \sum_i n_i \bar{n}_i, \tag{1}
\]

where the chemical potential term is dropped. We want to investigate a Mott metal-insulator transition in this frustrated system, and therefore need to calculate the electron Green function at finite temperatures. The question is what is an appropriate approach for taking account of frustration and strong electron correlation effects. The Mott transition has been successfully described by the dynamical mean field theory (DMFT) [5]. The system is mapped to a single-site impurity interacting with the reservoir via temporally fluctuating mean field. This mean field is determined self consistently from the electron propagator at the impurity site. Generalising this with using a cluster of multiple sites, we can completely include temporal and spatial correlations inside the cluster. This is the cellular dynamical mean field theory (CDMFT) [6], and we use this approach with a cluster of four sites. The details are explained in Ref. [8]. Due to frustration, the \(k\)-dependence of spin and other correlations is usually suppressed.

**Figure 1.** Phase diagrams of \(\kappa\)-(BEDT-TTF)\(_2\)X salts: (a) X=Cu\(_2\)CN\(_3\) (based on [4]) and (b) X=Cu[N(CN)\(_2\)]Cl (based on [3]). Phase diagrams of the Hubbard model determined by DMFT: (c) single-site DMFT (base on [5]) and (d) considering antiferromagnetic solution. In unfrustrated case, metal-insulator transition takes place at the same time of magnetic instability, and the transition in the insulator phase is not a real one [7].
strongly. Thus, the short-range correlation effects are predominantly important and we can take account of this part with the CDMFT.

The issue to discuss is the boundary of metal and insulator phases. First notice that it has a different slope near zero temperature between the two $\kappa$-type compounds as shown in figure 1 (a) and (b). Upon increasing temperature, the boundary moves to the lower pressure side in the case of $X=\text{Cu}[\text{N(CN)}_2]\text{Cl}$, while to the higher pressure side in the more frustrated case, $X=\text{Cu}_2\text{CN}_3$. The latter case means that the insulating phase has a larger entropy, and it is related to static local spins in the insulating phase. Noticing that applying pressure enhances band width $W$, i.e. reduction of $U/W$, one can explain qualitatively this behaviour by the single-site DMFT shown in figure 1 (c). In the less frustrated case $X=\text{Cu}[\text{N(CN)}_2]\text{Cl}$, the insulating phase is located in the lowest-temperature region, meaning smaller entropy, and this is opposite to the highly frustrated case. To discuss this, we determine the phase diagram of the anisotropic triangular Hubbard model by means of the CDMFT.

The phase diagram is shown in figure 2(a) for the intermediate frustration case $t'/t=0.75$. Here $W$ is the total band width of the noninteracting system and $W=8.33t$. It is determined from double occupancy $D=\langle n_{i\uparrow}n_{i\downarrow}\rangle$. This quantity is a measure of metallicity, and its larger value corresponds to a better metal. When temperature $T$ is fixed and the electron repulsion $U$ is decreased, $D$ shows a jump to a larger value at $U_c(T)$. This is an insulator-to-metal transition, and the transition is first order. When the temperature is set at a higher value, the transition point $U_c(T)$ increases. However, the jump shrinks in size at the same time and disappears around $T/W \sim 0.03$, indicating a critical end point of the first-order transition line. In addition to a jump at low temperature, there exists a crossover at a higher temperature shown by the dotted line in figure 2(a). When the temperature dependence is concerned, $D(T)$ is not monotonic at $U/W < \sim 1.07$. With lowering temperature, $D(T)$ shows a decreasing behaviour above the crossover temperature, while increasing below it.

The first order line at low temperature and the crossover line at higher temperature form a boundary of metal and paramagnetic insulator phases, and it agrees qualitatively with the experimental result of
κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl. Precisely speaking, the position of the critical end point is different, but this may be due to pressure effects on other microscopic parameters.

It is important to clarify the difference between the higher- and lower-$T$ parts of the insulating phase. This can be visualised most clearly with the electron spectral function, i.e., the imaginary part of the Green function. The results are shown in figure 2 (b)-(d) for three typical temperatures. In the higher-$T$ part of the insulating phase, the spectral function has one broad peak at a negative energy and another part of the insulating phase is stable due to large spin entropy of excited states. These excited states are generated by frustration and similar to low-energy states in the highly frustrated systems. At lower temperatures, the system acquires a lower energy through coherent propagation of quasiparticles, and becomes metallic. Due to strong correlations, these quasiparticles are subject to incommensurate spin fluctuations and this drives the low-temperature metal-insulator transition.

In the intermediate metallic phase, interestingly, a new narrow band gradually appears inside the Mott peak at a positive energy. There is a large energy gap between the two, characteristic of the Mott insulator. This can be visualised most clearly with the electron spectral function, i.e., the imaginary part of the Green function. The results are shown in figure 2 (b)-(d) for three typical temperatures. In the higher-$T$ part of the insulating phase, the spectral function has one broad peak at a negative energy and another peak at a positive energy. There is a large energy gap between the two, characteristic of the Mott insulator. In the intermediate metallic phase, interestingly, a new narrow band gradually appears inside the Mott gap. As seen in the figure for $T/W=0.02959$, this new band crosses $\omega=0$, and this confirms the metallic behaviour.

In the standard picture of the Mott transition, the quasiparticle peak around $\omega=0$ disappears and its spectral weight is transferred to upper and lower Hubbard bands. The low temperature transition in the present system takes place in a different way. The low-energy quasiparticle band splits to two bands and a small gap is generated at $\omega=0$. The upper and lower Hubbard bands do not change during this transition.

The result of magnetic susceptibility indicates that the low-temperature metal-insulator transition is driven by spin fluctuations. The susceptibility at an incommensurate wave vector diverges very close to the metal-insulator transition line, and the boundary of the magnetically ordered phase is shown in figure 2(a). The boundary is related to symmetry breaking in spin space and therefore cannot terminate at a point of finite $T$ and $U$, but the calculations do not converge for larger $U$’s.

In summary, the reentrant behaviour of metal-insulator transition in the present anisotropic triangular system is a consequence of frustration and strong electron correlation effects. The higher-temperature part of the insulating phase is stable due to large spin entropy of excited states. These excited states are generated by frustration and similar to low-energy states in the highly frustrated systems. At lower temperatures, the system acquires a lower energy through coherent propagation of quasiparticles, and becomes metallic. Due to strong correlations, these quasiparticles are subject to incommensurate spin fluctuations and this drives the low-temperature metal-insulator transition.

3. An effective Hamiltonian for three-orbital Hubbard model on a pyrochlore lattice

The vanadium spinel LiV$_2$O$_4$ is a heavy fermion system of $d$-electrons instead of $f$-electrons [9]. The observed low-temperature properties indicate a large effective mass of quasiparticles and the characteristic energy scale $T^* \sim 30$ K [10, 11]. Conduction band is primarily composed of $d$-electrons of vanadium ions, which form a frustrated pyrochlore lattice, and each vanadium has $d^{1.5}$ electrons. Various interesting characters have been uncovered for this system by theoretical and experimental studies, [12, 13, 14, 15, 16, 17, 18] but the origin of heavy fermion behaviours is not pinned down yet. In addition to the geometrical frustration of pyrochlore lattice, another important point is the three-fold degeneracy of $t_{2g}$ orbital of conduction electron. We derive a low-energy effective Hamiltonian by a real-space renormalisation group approach, in order to discuss the interplay of orbital and spin degrees of freedom in a pyrochlore lattice.

3.1. Model

The microscopic Hamiltonian of LiV$_2$O$_4$ is the $t_{2g}$ three-orbital Hubbard model on the pyrochlore lattice,

$$
H = \sum_{r\sigma} \left[ \sum_{\alpha<\beta} t^{\alpha\beta}_{rr'} c^\dagger_{r\alpha \sigma} c_{r' \beta \sigma} + \frac{1}{2} U n_{r\alpha \sigma} n_{r\beta \bar{\sigma}} + \sum_{\beta<\alpha'} U' n_{r\alpha \sigma} n_{r\beta \alpha'} + J d^{\dagger}_{r\alpha \sigma} d_{r\beta \sigma} \right],
$$

(2)

where the standard notation is used and $\alpha=(xy, yz, and zx)$ is the orbital index. We consider only the nearest neighbour electron hoppings $t_{\sigma} = -0.527$ eV, $t_{\pi} = -0.1$ eV and $t_{\delta} = 0.25$ eV. The trigonal distortion of oxygen is included in $t^{\alpha\beta}_{rr'}$ and $t_{2g}$ orbitals are split into $a_{1g}$ and $e_g$ orbitals. This crystalline field splitting is defined as $\Delta \equiv \epsilon_{e_g} - \epsilon_{a_{1g}}$. For the interaction term, the relation $U = U' + J$ should be
retained and we set $U = 1.5$ eV. It is useful to consider molecular orbitals in a tetrahedron unit cell. Twelve molecular orbitals form 5 multiplets $A_1, E, T_1, T_2^{(-)}$ and $T_2^{(+)}$ in $T_d$ point group. The energy level scheme is shown in figure 3 and this is consistent with the results of band structure calculations [12, 19].

### 3.2. One-tetrahedron eigenstates

We first analyse multi-electron eigenstates in a one-tetrahedron unit, a building block of the pyrochlore lattice. This is the first step of the real-space renormalisation group and they are to be used as bases for the effective model [20]. We numerically diagonalise the Hamiltonian of one-tetrahedron unit with open boundary conditions. The ground-state phase diagram is shown in figure 4 for the total d-electron number $n_d = 6$. This corresponds to the average density $d^{1.5}$. A wide range of the phase diagram is covered by $^7A_1$ states. Here we use the standard notation $2S + 1\Gamma$ with the spin $S$ and the irreducible representation $\Gamma$ of the $T_d$ point group. The $^7A_1$ state has the maximum spin, and this is stabilised by ferromagnetic double-exchange interactions, whereas the $^3T_1$ state has a partially screened spin. It is important that these ground states are very well described by electron configurations shown in figure 3 despite strong electron correlations. In the following, we concentrate on the parameter region where $^3T_1$ states is the ground state for one-tetrahedron unit, since it is within the realistic parameter region and both orbital and spin degrees of freedom are expected to contribute to low-energy properties.

We can describe most of low-energy states by the Hubbard model of three-fold $T_2^{(-)}$-orbitals. Its vacuum is the ground state of $n_d=4$ electrons. It is spin singlet and orbital singlet, and is described as the configuration with fully occupied $E$-orbitals. As discussed before, the ground state ($^3T_1$) of the $n_d=6$ case is described as in figure 3, and the ground states for $n_d=5$ and $7$ are also explained by this one-tetrahedron molecular orbital picture, as well as most of low-energy excited states in these charge sectors.

### 3.3. Low-energy effective model

We now consider interactions between tetrahedron units. These are generated by the bare electron hopping processes between neighbouring tetrahedra. Apart from real hopping processes, the most important interactions are exchange interactions of the $n_d = 6$ configurations: $S = 1$ and three-fold orbital $T_1$. These include spin-spin and orbital-orbital interactions and also spin-orbital coupled ones. The effective exchange Hamiltonian between tetrahedra $n$ and $m$ is written in terms of spin-1 operators...
that the xy-component of bond direction. To be specific, when the (110) bond is considered, the lowest-energy configuration is such as

\[ S = \left\{ \begin{array}{cl} 0 & \text{for } \mathbf{A} \\
1 & \text{for } \mathbf{B} 
\end{array} \right. \]

original Hamiltonian, the orbital interactions are not isotropic either, and their anisotropy depends on the spatial symmetry.

Figure 5. Two typical orbital states of four-tetrahedron system favoured by \( J^{88}_1 \). Two-electron configurations in the \( T_2^{(-)} \)-orbitals are shown. Wavefunctions drawn represent the spatial symmetry.

There are 30 lowest-energy configurations, each of which has four favoured bonds, and two examples are shown in figure 5 where we draw arrows for the favoured bonds. Due to this degeneracy, this system can be well understood by considering this spatial symmetry. Let us consider the system of a cubic unit cell including four coupled tetrahedra. It is impossible to make all the six bonds take the lowest-energy configuration. There are 30 lowest-energy configurations, each of which has four favoured bonds, and two examples are shown in figure 5 where we draw arrows for the favoured bonds. Due to this degeneracy, this system is expected to have a large entropy in the low-energy sector.

Among the spin-orbital coupled terms, the term \( J^{88}_1 T_8 T_8' S \cdot S' \) is the largest with \( J^{88}_1 > 0 \), compatible in size to the pure spin exchange \( J^{90}_2 \). This term favours antiferromagnetic spin correlation when \( T_8 T_8' > 0 \) and ferromagnetic one when \( T_8 T_8' < 0 \). Indeed, spin-orbital correlations in the four-tetrahedron system can be well understood by considering this \( J^{88}_1 \) term [22]. Let us look at the two bonds A and B in figure 5(a), and consider the presented orbital configuration at both ends of each bond. Figure 6 shows the conditional spin correlations between the two tetrahedra at the ends of each bond, calculated for the three lowest-energy states with different symmetries. This conditional correlation is defined as \( \langle \mathbf{S}_n \cdot \mathbf{S}_{n'} P_n(\gamma) P_{n'}(\gamma') \rangle \), where \( P_n(\gamma) \) is the projector to the specified orbital configuration \( \gamma \) at the tetrahedron \( n \) as shown in figure 5(a). This represents the cross correlation of orbital and spin configurations. As one can see, on the bonds with lowest-energy orbital configuration (like A) favoured by \( J^{88}_1 \) term, the spin correlation is primarily antiferromagnetic. On the unfavoured bonds (like B), the spin correlation is modified towards ferromagnetic side. As well as the other quantities presented in Ref. [22], these results show that the spin and orbital degrees of freedom in this system are strongly entangled.

When we consider the lowest-energy orbital configurations of the \( J^{88}_1 \) term, the \( J^{88}_2 \) terms are ferromagnetic in spin space. These ferromagnetic tendency correlated with the orbital correlations.
interacting tetrahedron units, in which one-particle states are molecular orbital
three-orbital Hubbard model on the pyrochlore lattice. We have derived an effective Hamiltonian of

\[ \text{References} \]

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It is also important to notice another interaction, i.e., ferromagnetic double exchange processes. Combining all these terms, the low-energy effective Hamiltonian becomes \( t-J \) model like:

\[ H_{\text{eff}} = \sum_n \left[ -\mu_{\text{eff}} N(n) + U_{\text{eff}} N^2(n) \right] + \sum_{(n,m)} \sum_{\alpha\beta\sigma} \left( \tau^\alpha_{nm} P a_{n+1\alpha\sigma} a_{m+\beta\sigma}^\dagger + \text{h.c.} \right) \]

\[ + \sum_{(n,m)} \sum_{\mu\nu} \left[ \frac{1}{2} J_1^{\mu\nu}(nm) + J_2^{\mu\nu}(nm) S(n) \cdot S(m) \right] T_{\mu}(n) T_{\nu}(m) + \cdots. \]  

(4)

where \( N(n) = \sum_{\alpha\sigma} a_{n+1\alpha\sigma}^\dagger a_{n\alpha\sigma} \). This is derived particularly for the average density \( d^{1.5} \), corresponding to LiV\(_2\)O\(_4\). Here, \( P \) is the projection operator to the restricted Hilbert space, i.e., the ground states of \( n_d = 5, 6, \) and \( 7 \) spaces of a tetrahedron unit. \( a_{n+1\alpha\sigma} \) represents the quasiparticle in the \( T_2^{(-)} \) orbital. \( S \) and \( T_{\mu} \) are the spin one and triplet orbital operators defined before. \( \mu_{\text{eff}} \) and \( U_{\text{eff}} \) are the effective chemical potential and Coulomb interaction, determined from the one-tetrahedron spectra. \( U_{\text{eff}} \) is renormalized to about 1/5 of the bare \( U \) in our calculations.

In conclusion, we have carried out the first step of the real-space renormalisation group for the three-orbital Hubbard model on the pyrochlore lattice. We have derived an effective Hamiltonian of interacting tetrahedron units, in which one-particle states are molecular orbital \( T_2^{(-)} \) of the tetrahedron unit. In the superexchange interaction terms, various channels compete to each other both in spin and orbital space. In addition, double exchange processes also compete with some of them. These aspects and the geometrical frustration of the pyrochlore lattice are expected to reduce the energy scale of this system. This would lead that the entropy which is originated from spin one orbital triplet degrees of freedom in in the tetrahedron units remains large down to very low temperatures. Due to these competing interactions and strongly entangled spin and orbital degrees of freedom, the effective mass and other properties in the metallic phase will be strongly renormalized. We hope that further study of the effective model (4) will provide useful results for clarifying the origin of heavy fermion behaviour in LiV\(_2\)O\(_4\). This model may also be valuable for investigating the interplay of spin and orbital correlations in metallic systems in general.

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