A Possible Bipolaronic Transition in β-pyrochlore Compounds

T. Fuse[a], Y. Ōno[a,b]

[a] *Department of Physics, Niigata University,Ikarashi, Niigata 950-2181, Japan*
[b] *Center for Transdisciplinary, Reserch, Niigata University, Ikarashi, Niigata 950-2181, Japan*

Abstract

We investigate the Hubbard-Holstein model by using the dynamical mean-field theory combined with the exact diagonalization method. Below a critical temperature $T_{\rm cr}$, a coexistence of two solutions, small lattice fluctuating solution (polaronic state) and large one (bipolaronic state), is found for the same value of the electron-phonon coupling g in the range $g_{c1}(T) < g < g_{c2}(T)$ for a fixed value of the Coulomb interaction U. As the result, the system shows a first-order phase transition at $T_p(< T_{\rm cr})$ from the bipolaronic to the polaronic states as T decreases for $g_{c1}(T_{\rm cr}) < g < g_{c2}(0)$. The effective potential for oscillating atoms, which is renormalized due to the effect of g, is double-well (strongly anharmonic) type for $T > T_p$, while it is single-well (harmonic) type for $T < T_p$. When U decreases, changes in physical quantities such as the lattice and the charge fluctuations at T_p decrease together with decreasing the values of g_{c1}, g_{c2} and T_p. The obtained transition for small U case with small changes in the physical quantities seems to be account for the rattling transition observed in the β-pyrochlore superconductor KOs_2O_6.

Key words: β-pyrochlore compounds, KO_2O_6, bipolaronic transition, Hubbard-Holstein model

β-pyroclore KOs_2O_6 is a transition metal oxide superconductors with it's transition temperature $T_c \sim 9.6$K. Recently, this compound is reported to have another anomalous that the specific heat has a magnetic-independent second peak appearing at $T_p \sim 7.5$K below T_c, implying a structure-dependent first order transition [1]. In addition, the concave-downward resistivity behaivior, which is suggested to have roots in a strong electron-phonon interaction [2], and the disappearance of the T_p transition by applying pressure is also reported. This compound has Os_2O_6 corner-sharing tetrahedral cage structures, and in it, K ion is considered to take an off-center oscilation for neighboring four K ion site. For this reason, it is discussed that this rattling motion relates to the unknown T_p transition.

In this paper, we investigate the Hubbard-Holstein model by using the dynamical mean-field theory (DMFT) with the exact diagonalization method. The Holstein model is given by the following Hamiltonian

$$\begin{aligned} H &= \sum_{\boldsymbol{k}\sigma} \epsilon_{\boldsymbol{k}} c^\dagger_{\boldsymbol{k}\sigma} c_{\boldsymbol{k}\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} + \omega_0 \sum_i {b_i}^\dagger b_i \\ &\quad + g \sum_i \left({b_i}^\dagger + b_i \right) \left(\sum_\sigma n_{i\sigma} - 1 \right), \end{aligned} \tag{1}$$

where $c^\dagger_{i\sigma}$ are creation operators for a electron at site i with spin σ, and the number operators are defined by $n_{i\sigma} = c^\dagger_{i\sigma} c_{i\sigma}$; the lattice displasement is given by $Q_i = \frac{1}{\sqrt{2\omega_0}}({b_i}^\dagger + b_i)$. The quantities, $\epsilon_{\boldsymbol{k}}$, U, g and ω_0 are the energy for a electron with wave vector $\boldsymbol{k}$, the Coulomb interaction, the electron-phonon coupling strength, and the frequency of the Einstein phonon. In the DMFT[3], the model eq.(1) is mapped onto an effective single impurity Holstein model. In the limit of infinite dimensions, the self-energy becomes purely site-diagonal. To solve the impurity Holstein model[4, 5], we use the ED method for a finite-size cluster[5, 6].

In following numerical results, we show 4-site impurity model, finite temperature DMFT investigation. We choose the cutoff of phonon number 20, which is reasonable amount in the range of g of publishing in this paper. We assume a semielliptic density of states for the bare conduction band with bandwidth $W = 1$, and we set $U = 1$, $\omega_0 = 0.2$. We concentrate our attention on the particle-hole symmetric case with total electron number $\langle n \rangle = 1$.

Fig.1 (a) shows g dependence of the lattice fluctuation, i.e., the root mean square value of the displacement $\sqrt{\langle Q^2 \rangle / \langle Q^2 \rangle_0}$ for $T = 0.0025$, where $\langle Q^2 \rangle_0 = \frac{1}{2\omega_0}$ is lattice fluctuation for $g = 0$, i.e., the zero-point oscillation. With increasing g, the lattice fluctuation increases gradually, while it does steeply at $g \sim 0.16$, and then finally it shows linear increament after that. In the inset (b), we expand the region of the steep increase in detail for several termperature $T = 0.001, 0.00175, 0.003$. With increasing and decreasing g, abrupt changes in the lattice fluctuation is found at $g = g_{c1}$ and g_{c2}. For $g_{c1} \le g \le g_{c2}$, a large and a small lattice fluctuating solutions exist for the same g, resulting in the coexistence region, in which it is understood the existence of a first order transition.

Fig.1 (c) shows g dependence of the double-occupancy $d = \langle n_\uparrow n_\downarrow \rangle$, and the inset (d) indicates it's magnification of the vicinity of the first order transition. With increasing g,

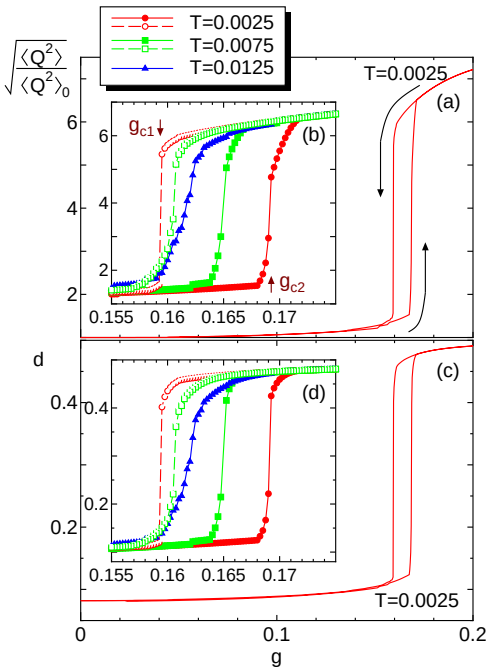

Figure 1: g dependence of the lattice fluctuation $\sqrt{\langle Q^2\rangle/\langle Q^2\rangle_0}$ (a) and the double-occupancy d (c) with increasing and decreasing g for $T = 0.0025$. The inset (b) and (d) shows the magnification of the vicinity of the first order transition of (a) and (c), respectively, for several termperature $T = 0.0025, 0.0075, 0.00125$.

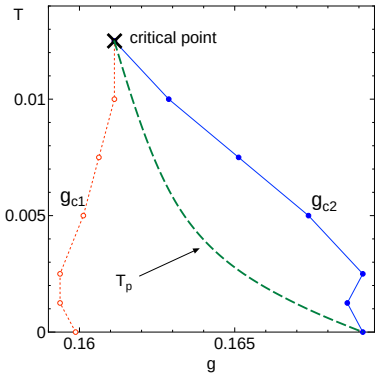

Figure 2: The g-T phase-diagram of g_{c1}, g_{c2}. The dashed curve between g_{c1} and g_{c2} is the expected first order transition point. The cross mark indicate a critical point of the first order phase transition.

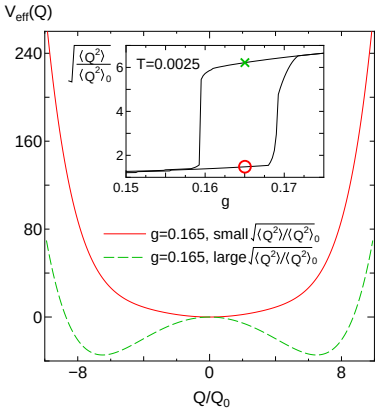

Figure 3: The effective potentials $V_{\rm eff}(Q)$ of an ion in a cage which is renormalized due to the effect of the electron-phonon coupling of the two solutions for the same value of $g = 0.165$ at $T = 0.0025$. The solid line correspond to the small lattice fluctuating solution (circle in the inset) and the dashed to the large one (cross).

d increases gradually for small $g \le g_{c2}$, then the small and the large d solution exist together for $g_{c1} \le g \le g_{c2}$. In the large d solution side, d is almost temperature independent, and shows fairy close to 0.5, at which in our definition, the electron state is represented as in bipolaronic.

In Fig.2, we plot g_{c1} and g_{c2} on the g vs. T diagram. g_{c1} corresponds with g_{c2} at the critical temperature $T = T_{\rm cr} \sim 0.0125$, then they becomes smooth crossover for $T > T_{\rm cr}$. Between g_{c1} and g_{c2}, there is a first order phase transition, which is essentially fixed from a minimam free energy condition. With DMFT, it is inadequate to decide a first order transition point for lack of the accuracy of energy, so we draw the expected one, referring the Mott transition case [5].

Fig.3 shows the effective potentials $V_{\rm eff}(Q)$ for oscillating atoms, which is renormalized due to the effect of g [7], of the two solutions for the same value of $g = 0.165$ at $T = 0.0025$. The effective potential for the small $\sqrt{\langle Q^2\rangle/\langle Q^2\rangle_0}$ solution, for which the electron state is similar to the one for low T below the first order transition side, is single-well (harmonic) type, while it is double-well (strongly anharmonic) type for the large $\sqrt{\langle Q^2\rangle/\langle Q^2\rangle_0}$ solution, for high T side. This result means that the first order transition also brings the change of the effective potentials for oscillating atoms, therefore the atoms take rattling motion or not.

In summary, we investigate the Hubbard-Holstein model by using the dynamical mean-field theory in finite temperature. It is found that a large and a small lattice fluctuating solutions coexist for the same $g_{c1} \le g \le g_{c2}$ in low temperature and strong correlating region, indicating a polaronic-bipolaronic first order transition. By the first order transition, the effective potential changes from harmonic type to anharmonic type, implying the change of the motion of atoms. This may be a explanation of a spin-independent, structure-dependent T_p transition observed in β-pyrochlore coumpoinds KOs_2O_6. In this paper, we discuss $U = 1$ case only. In $U = 0$ case, the authors will discuss quantitatively in detail in another paper later.

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