Kondo effect emerging from a spin-vibronic state

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Abstract. We discuss the Kondo effect in an electron system dynamically coupled with local Jahn-Teller phonons. For the purpose, we consider the two-orbital Anderson model including both Coulomb interactions and the coupling between electrons and Jahn-Teller phonons on an impurity site. Note that in this paper, we consider a quarter-filling case. First we analyze a local problem and confirm a spin-vibronic quartet ground state, characterized by the direct product of spin and vibronic degrees of freedom. Next we include the hybridization term and analyze the model by using a numerical renormalization group method. From the evaluation of entropy, specific heat, and several kinds of susceptibilities, it is found that spin and total angular momenta are simultaneously screened by conduction electrons, leading to the Kondo phenomenon due to the entropy release of $\ln 4$ of the local spin-vibronic state. A possibility of heavy-electron formation in the spin-vibronic Kondo effect is briefly discussed.

1. Introduction
Recently, the Kondo effect originating from phonon degrees of freedom have attracted renewed attention both in the experimental and theoretical research fields of condensed matter physics. In particular, in cage-structure materials such as filled skutterudites and clathrate compounds, local anharmonic oscillations of a guest atom in the cage, called rattling, have been considered to play crucial roles for the emergence of exotic magnetic phenomena [1]. A typical example of such interesting phenomena is heavy-electron state induced by rattling. In fact, in the filled-skutterudite compound SmOs\(_4\)Sb\(_{12}\), it has been observed that the large electronic specific heat coefficient is almost independent of an applied magnetic field up to about 30 Tesla [2], in sharp contrast to the decrease of electronic specific heat coefficient due to the magnetic field in the conventional heavy-electron materials originating from the traditional spin Kondo effect. The mechanism of rattling-induced heavy-electron state has been theoretically investigated from various viewpoints [3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15].

Among several kinds of oscillation modes of the guest atom in the cage, here we focus on local Jahn-Teller mode. When such Jahn-Teller phonons are dynamically coupled with orbital degree of freedom of electrons, the local ground state is known to be a vibronic state characterized by clockwise and anti-clockwise rotational mode of Jahn-Teller phonons. Such geometric degree of freedom is screened by orbital moment of electrons and there occurs the release of an entropy $\ln 2$ of the vibronic state, leading to the Kondo-like phenomenon [16, 17, 18, 19]. Due to this type of Kondo effect, we expect the emergence of heavy-electron state, but this possibility was discussed with the use of the dynamical mean-field theory for the orbital-degenerate periodic Anderson model coupled with Jahn-Teller phonons [20]. When we explicitly consider the spin degree of freedom in the vibronic state, we expect a spin-vibronic quartet ground state, which
should be given by the direct product of spin and vibronic degrees of freedom. We believe that it is an intriguing problem to pursue the Kondo effect emerging from the spin-vibronic state.

In this paper, we numerically analyze the two-orbital Anderson model including both Coulomb interactions and the coupling between electrons and Jahn-Teller phonons on an impurity site. For the convenience of the calculation, we introduce unitary transformations of electron and phonon operators, which naturally express the rotational mode of the Jahn-Teller phonons and remove the pair-hopping interaction term from the original Hamiltonian. By using a numerical renormalization method [21, 22], we evaluate entropy, specific heat, and several kinds of susceptibilities. It is found that the spin and total angular momenta are simultaneously screened by conduction electrons, leading to the Kondo phenomenon due to the entropy release of ln 4 of the local spin-vibronic state. Then, we point out a possibility of heavy-electron formation by the spin-vibronic Kondo effect. Throughout this paper, we use such units as $\hbar = k_B = 1$.

2. Model Hamiltonian and Local Electron States

2.1. Model Hamiltonian

In this paper, we consider the two-orbital Anderson model including both Coulomb interactions and the coupling between electrons and Jahn-Teller phonons, expressed by

$$H = \sum_{k,\ell} \varepsilon_k c_{k\ell}^\dagger c_{k\ell} + V \sum_{k,\ell} (c_{k\ell}^\dagger d_{\ell} + \text{h.c.}) + H_{\text{loc}},$$

(1)

where $\varepsilon_k$ is the dispersion of conduction electron, $c_{k\ell}$ is the annihilation operator of conduction electron with momentum $k$, orbital $\ell$ ($=a$ and $b$), and spin $\sigma$ ($=\uparrow$ and $\downarrow$), $d_{\ell}$ is the annihilation operator of impurity electron, and $V$ denotes the hybridization between conduction and impurity electrons of the same orbitals. The final term $H_{\text{loc}}$ indicates the local Hamiltonian, given by

$$H_{\text{loc}} = U \sum_{\ell} n_{\ell\uparrow} n_{\ell\downarrow} + U' n_{a} n_{b} + J \sum_{\sigma,\sigma'} b_{\sigma}^\dagger b_{\sigma'}^\dagger d_{\sigma} d_{\sigma'} + J' (d_{a\uparrow}^\dagger d_{a\downarrow}^\dagger b_{\uparrow} b_{\downarrow} + \text{h.c.}) + E_d n + \sqrt{\alpha} \omega ((a_2 + a_4 \uparrow) \tau_x + (a_3 + a_4 \downarrow) \tau_z) + \omega (a_2 a_2 + a_3 a_3 + 1),$$

(2)

where $U$ is intra-orbital Coulomb interaction, $n_{\ell\sigma} = d_{\ell\sigma}^\dagger d_{\ell\sigma}$, $U'$ is inter-orbital Coulomb interaction, $n_{a} = n_{a\uparrow} + n_{a\downarrow}$, $J$ and $J'$ denote exchange and pair-hopping interaction, respectively, $E_d$ denotes the level of localized electron, $n$ is local electron number, given by $n = n_{a} + n_{b}$, $\alpha$ is non-dimensional electron-phonon coupling constant, $a_2$ and $a_3$ are annihilation operators for $(x^2 - y^2)$- and $(3z^2 - r^2)$-type Jahn-Teller phonons, respectively, $\tau_x = \sum_{\sigma} (d_{\sigma\uparrow}^\dagger d_{\sigma\downarrow} + d_{\sigma\downarrow}^\dagger d_{\sigma\uparrow})$, $\tau_z = \sum_{\sigma} (d_{\sigma\uparrow}^\dagger d_{\sigma\uparrow} - d_{\sigma\downarrow}^\dagger d_{\sigma\downarrow})$, and $\omega$ is the frequency of local Jahn-Teller phonons. Note that $U = U' + J + J'$ due to the rotational invariance in the orbital space, while $J = J'$ due to the fact that the electron wavefunction is real for the evaluation of Coulomb integrals.

Although it is possible to perform the numerical calculations even in the present form of the Hamiltonian, it is convenient to introduce the following unitary transformations for both electron and phonon operators [23]:

$$c_{k\pm\sigma} = (c_{k\sigma} \mp i c_{k\bar{\sigma}}) / \sqrt{2}, \quad d_{\pm\sigma} = (d_{\sigma} \mp i d_{\bar{\sigma}}) / \sqrt{2}, \quad a_{\pm} = (a_3 \mp ia_2) / \sqrt{2}.$$  

(3)

In the following, we use the index $\gamma$ to distinguish the pseudo-orbital, $+$ and $-$. From the above transformations, eqs. (1) and (2) are, respectively, rewritten as

$$H = \sum_{k,\gamma} \varepsilon_k c_{k\gamma\sigma}^\dagger c_{k\gamma\sigma} + V \sum_{k,\gamma} (c_{k\gamma\sigma}^\dagger d_{\gamma\sigma} + \text{h.c.}) + H_{\text{loc}},$$

(4)
In order to obtain this form, it is important to include Coulomb interaction terms correctly in the original Hamiltonian from the viewpoint of the symmetry. In order to treat the phonon part, we truncate the phonon basis for each Jahn-Teller mode at a finite number as the energy unit for the local problem. For small $U'$, the ground states with double degeneracy are composed of the states of $n = 0$ and $n = 2$ with the same weights, leading to

\[ H_{\text{loc}} = (U' + J) \sum_{\gamma} n_{\gamma \uparrow} n_{\gamma \downarrow} + (U' + J) n_{\uparrow} n_{\downarrow} + 2J \sum_{\sigma, \sigma'} d_{\sigma \sigma'}^+ d_{\sigma'}^+ d_{\sigma}^+ d_{\sigma}^+ \]

where $n_{\gamma \sigma} = d_{\gamma \sigma}^+ d_{\gamma \sigma}$, $n_{\sigma} = \sum_{\gamma} n_{\gamma \sigma}$, and $\tau_{\pm} = \sum_{\sigma} d_{\pm \sigma}^+ d_{\mp \sigma}$. Note that $n$ is invariant under this transformation and we use the relations of $U' = U' + J + J'$ and $J = J'$. Form the comparison of eqs. (2) and (5), we remark that the pair-hopping term disappears after the transformation.

In order to obtain this form, it is important to include Coulomb interaction terms correctly in the original Hamiltonian from the viewpoint of the symmetry. When we use the transformed Hamiltonian, there exists an advantageous point that we can adopt the total angular momentum $J_z$, defined by $J_z = T_z + L_z$, as the conserved quantity, where $T_z = \sum_{\sigma} (n_{\uparrow \sigma} - n_{\downarrow \sigma})/2$ and $L_z = a_\uparrow^d a_\downarrow - a_\downarrow^d a_\uparrow$. Namely, $J_z$ is composed of the electron orbital moment and the rotational moment of Jahn-Teller phonons. Note that this conserved quantity is not easily found in the form of the original Hamiltonian, eqs. (1) and (2). From the viewpoint of the numerical calculations, it is useful to introduce the conserved quantity $J_z$ in addition to total electron number and total spin moment, since we can save the computer resources.

### 2.2. Local electron states

Before proceeding to the numerical results of the Anderson model, let us discuss the local electron state. Since we are interested in the dynamical Jahn-Teller effect, we consider the quarter-filling case with one electron on the impurity site, by setting $E_d$ so as to satisfy $\langle n \rangle = 1$, where $\langle \cdots \rangle$ denotes the operation to take thermal average. In order to treat the phonon part, we truncate the phonon basis for each Jahn-Teller mode at a finite number $N_{\text{ph}}$, which is set as $N_{\text{ph}} = 50$.

In Fig. 1(a), we show the eigenenergies of $H_{\text{loc}}$ of $U'$ for $J = U'/2$ and $\alpha = 1$. Note that we set $\omega$ as the energy unit for the local problem. For small $U'$, the ground states with double degeneracy are composed of the states of $n = 0$ and $n = 2$ with the same weights, leading to
\( \langle n \rangle = 1 \). Note that the state of \( n = 2 \) is characterized by the linear combination of two double-occupied orbital states, \( d_{a_1}^\dagger d_{a_1}^\dagger |0\rangle \) and \( d_{b_1}^\dagger d_{b_1}^\dagger |0\rangle \), where \( |0\rangle \) denotes the vacuum state. On the basis of eq. (5), it is possible to derive the effective interactions mediated by dynamical Jahn-Teller phonons and the pair-hopping attraction is enhanced among several kinds of interactions [24]. Thus, in order to gain the attractive interaction, double-occupied orbital states are stabilized for small \( U' \).

However, when we increase the Coulomb repulsion, such double-occupied orbital states become unstable. Instead of them, there appears a single-occupied orbital state with active spin degree of freedom. Thus, we expect the quartet ground state, when we increase the magnitude of \( U' \), as observed in Fig. 1(a). It is called the spin-vibronic state from the following reason: Since this state includes one electron in the impurity site, the dynamical Jahn-Teller effect becomes active. The \( k \)-th eigenstate of \( H_{\text{loc}} \) labeled by \( \sigma \) and \( J_z \) is given by

\[
|\sigma, J_z; k \rangle = \sum_{j=0}^{\infty} \left[ p_{J_z}^{(k,j)} a_+^\dagger |J_z - 1/2; j\rangle + q_{J_z}^{(k,j)} d_-^\dagger |J_z + 1/2; j\rangle \right].
\]

Here \( |L; j \rangle = |L + j, j \rangle \) for \( L \geq 0 \) and \( |j, j + |L\rangle \) for \( L < 0 \), where \( |n_+, n_- \rangle = (1/\sqrt{n_+!n_-!})(a_+^{\dagger})^{n_+}(a_-^{\dagger})^{n_-}|0\rangle \). Note that \( J_z \) takes half-odd-integer values as \( J_z = \pm 1/2, \pm 3/2, \pm 5/2, \cdots \) and the coefficients satisfy the relations of \( p_{J_z}^{(k,j)} = q_{J_z}^{(k,j)} \) and \( q_{J_z}^{(k,j)} = p_{J_z}^{(k,j)} \). For a fixed spin \( \sigma \), we find the double degeneracy in each eigenstate originates from the rotational mode of Jahn-Teller phonons. The ground state is specified by \( k = 1 \) and \( J_z = \pm 1/2 \) for each \( \sigma = \uparrow \) and \( \downarrow \), suggesting that the ground state is quartet. We understand that the phonon state with \( L_z = J_z + 1/2 \) is coupled with the electron state with pseudo-orbital \( T_z = -1/2 \) \((-1/2)\), leading to the vibronic state specified by total angular momentum \( J_z \).

In Fig. 1(b), we show the local ground-state phase diagram on the plane of \( U' \) and \( J \). Note that we draw the figure in the region for physically realistic parameters of \( U' \geq J \). As easily understood from Fig. 1(a), we find the degenerate states of \( n = 0 \) and \( n = 2 \) in the region of small \( U' \). On the other hand, we observe the spin-vibronic state, when we increase the value of \( U' \). These two states are converted in a manner of the level crossing.

3. Calculated Results

Now we include the effect of hybridization in the model. In order to reveal electronic and phononic properties of \( H \) at low temperatures, it is necessary to evaluate corresponding susceptibilities. The susceptibility of an arbitrary operator \( A \) is given by

\[
\chi_A = \frac{1}{Z} \sum_{k,j} e^{-E_k/T} - e^{-E_j/T} E_j - E_k |\langle j|(A - \langle A \rangle) |k\rangle|^2,
\]

where \( E_k \) is the eigenenergy of the \( k \)-th eigenstate \( |k\rangle \) of \( H \), \( Z \) is the partition function given by \( Z = \sum_k e^{-E_k/T} \), and \( \langle A \rangle = \sum_k e^{-E_k/T} \langle k|A|k\rangle / Z \). In this paper, we calculate the susceptibilities of charge, spin, and total angular momentum, which are defined as \( \chi_c \), \( \chi_s \), and \( \chi_J \), respectively.

For the evaluation of susceptibilities, in this paper, we employ a numerical renormalization group (NRG) method [21, 22]. In this technique, it is possible to include efficiently the conduction electron states in the vicinity of the Fermi energy by discretizing momentum space logarithmically. In actual calculations, we introduce a cut-off \( \Lambda \) for the logarithmic discretization of the conduction band. Due to the limitation of computer resources, we remain \( m \) low-energy states in each renormalization step. In this paper, we set \( \Lambda = 5 \) and \( m = 5,000 \).

In the NRG calculations, the energy unit is \( D \), which is a half of the conduction electron bandwidth \( W \). Hereafter \( D \) is set as unity. In this energy unit, a temperature \( T \) is defined...
as $T_i = \Lambda^{-i(1-i)/2}$ in the NRG calculation, where $i$ is the number of the renormalization step. By using the NRG method, we also evaluate entropy $S_{\text{imp}}$ and specific heat $C_{\text{imp}}$ of localized electron. For the purpose, we calculate it by the numerical derivative of the entropy $S_{\text{imp}}$ through the relation of $C_{\text{imp}} = \partial S_{\text{imp}} / \partial \log T = (S_{\text{imp},i+1} - S_{\text{imp},i}) / \log (\Lambda^{1/2})$, where $S_{\text{imp},i}$ is the entropy at the step $i$.

First we consider the case of the local degenerate ground states of $n = 0$ and $n = 2$. In Figs. 2(a) and 2(b), we depict the results of entropy $S_{\text{imp}}$ and specific heat $C_{\text{imp}}$ for various values of $V$ for $U' = 0.3$ and $J = 0.15$. At $V = 0$, we find the double degeneracy originating from the local ground states of $n = 0$ and $n = 2$. For high temperatures as $T \lesssim 10^{-2}$, irrespective of the value of $V$, $S_{\text{imp}}$ shows large value due to the phonon excited states. With decreasing temperature, $S_{\text{imp}}$ rapidly decreases and indicates a plateau of $\ln 2$ for $V < 0.05$, while $S_{\text{imp}}$ directly goes to zero for $V > 0.05$ at relatively high temperatures.

For $V < 0.05$, in the plateau region, $C_{\text{imp}}$ becomes almost zero. When we further decrease temperature, we observe the release of entropy $\ln 2$ and $C_{\text{imp}}$ forms a clear peak. Here we define the Kondo temperature $T_K$ as the temperature at which $C_{\text{imp}}$ shows the peak. Finally, for $T < T_K$, both $S_{\text{imp}}$ and $C_{\text{imp}}$ eventually vanish. For $V > 0.05$, without showing the peak in $C_{\text{imp}}$ at $T = T_K$, $C_{\text{imp}}$ becomes zero. At temperatures lower than $T_K$, we expect that the system is in the local Fermi-liquid state.

In Fig. 2(c), we depict the susceptibilities for charge, spin, and total angular momentum for $V = 0.001$, in order to clarify what is screened in the present Kondo-like phenomenon. We observe that $T_{\chi s}$ and $T_{\chi J}$ are rapidly suppressed at the temperature higher than $T_K$, while $T_{\chi c}$ becomes zero around at $T_K$. The behavior of $\chi_c$ is quite similar to that of the Anderson-Holstein model with attractive interaction larger than Coulomb repulsion. Namely, we confirm that the charge degree of freedom is relevant to the Kondo-like phenomenon due to the entropy release of $\ln 2$, as naively expected from the local degenerate ground states of $n = 0$ and $n = 2$.

Next we show the results for the local spin-vibronic ground state. In Figs. 3(a) and 3(b), we show the temperature dependence of $S_{\text{imp}}$ and $C_{\text{imp}}$ for several values of $V$. At $V = 0$, we find the four-fold degeneracy due to the local spin-vibronic ground state, leading to $\ln 4$ as residual entropy. In the high-temperature region such as $T \lesssim 10^{-2}$, we also observe large $S_{\text{imp}}$ due to phonon degrees of freedom. With decreasing temperature, $S_{\text{imp}}$ rapidly decreases. For $V > 0.35$, we do not find a plateau and $S_{\text{imp}}$ smoothly goes to zero. On the other hand, for $V < 0.35$, a plateau of $\ln 4$ is clearly observed and in this region, $C_{\text{imp}}$ becomes zero. When the entropy $\ln 4$ is released, we find a peak in $C_{\text{imp}}$, which also defines the Kondo temperature $T_K$. For $T < T_K$,
Figure 3. The temperature $T$ dependences of (a) the entropy $S_{\text{imp}}$ and (b) specific heat $C_{\text{imp}}$ for several $V$ for $U' = 0.6$ and $J = 0.3$. (c) The susceptibilities for charge $T_{\chi_c}$, spin $T_{\chi_s}$, and total angular momentum $T_{\chi_J}$ vs. $T$ for $U' = 0.6$, $J = 0.3$, and $V = 0.15$.

4. Discussion and Summary
We have discussed the Kondo effect of the two-orbital Anderson model which is dynamically coupled with local Jahn-Teller phonons. It has been found that spin and total angular momenta are simultaneously screened by conduction electrons, leading to the release of $\ln 4$, for the case with the local spin-vibronic ground state.

Here we provide a brief comment on the heavy-electron state, where the electronic specific heat coefficient $\gamma$ becomes $10 \sim 10^3$ times larger than that of the normal metal. From the relation of $C_{\text{imp}} = \gamma T$, we consider that the large peak of $C_{\text{imp}}$ implies the enhancement of $\gamma$. On the basis of the local Fermi-liquid theory, we obtain the scaling relation of $\gamma \sim 1/T_K$. For the parameter region of the local spin-vibronic ground state, we find that $T_K$ is lower than that for the case of the local degenerate ground states of $n = 0$ and $n = 2$, when we use the same value of $V$. Namely, we expect that $\gamma$ becomes large in the local spin-vibronic ground state. In our preliminary numerical results on $\gamma$ (not shown here), we observe that $\gamma$ is rapidly increased, when we enter the region of the local spin-vibronic state with the increase of $U'$ for a fixed value of $J$. The results seem to indicate a possibility of heavy-electron state due to the spin-vibronic Kondo effect. It is one of our future problems to confirm this issue.

Note that in the spin-vibronic Kondo effect, we have observed the release of the entropy $\ln 4$, but it is not obvious that the release of entropy $\ln 4$ always occurs irrespective of the parameters. In particular, the adiabaticity seems to be a key issue. In this paper, we set $\omega = 0.4$ in the unit of $D = 1$, but it indicates $\omega/W = 0.2$, where $W$ denotes the electron bandwidth. This value is not large enough to violate immediately the adiabatic condition, but in actual materials, $\omega$ seems to be smaller than the present values. In the adiabatic region, one may consider a possibility of the separation of the screening of spin and total angular momenta. On the other hand, in a purely electronic two-orbital Anderson model, we expect the occurrence of local spin-orbital quartet ground state, leading to the simultaneous release of the entropy $\ln 4$ of spin and orbital degrees of freedom. If the spin-vibronic ground state is continuously connected to the spin-orbital state of the purely electronic model, there may always occur the simultaneous release of the entropy.
ln 4. In any case, it is another future problem to clarify precisely how the entropy release of ln 4 occurs in the present model.

In summary, we have analyzed the two-orbital Anderson model coupled with local Jahn-Teller phonons. Depending on the parameters, we have found a couple of local ground states. Among them, we have emphasized the entropy release of ln 4, originating from the local spin-vibronic quartet ground state. After the release of ln 4, we expect the appearance of heavy-electron state due to the spin-vibronic Kondo effect. The condition of the occurrence of entropy release of ln 4 will be discussed elsewhere in future.

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