Electronic State and Magnetic Susceptibility in Orbitally Degenerate ($J = 5/2$) Periodic Anderson Model

Hiroshi KONTANI and Kosaku YAMADA^1

*Institute for Solid State Physics, University of Tokyo, 7-22-1 Roppongi, Minato-ku, Tokyo 106
^1 Department of Physics, Faculty of Science, Kyoto University, Kyoto 606-01

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Magnetic susceptibility in a heavy fermion system is composed of the Pauli term ($\chi_P$) and the Van-Vleck term ($\chi_V$). The latter comes from the interband excitation, where $f$-orbital degeneracy is essential. In this work, we study $\chi_P$ and $\chi_V$ in the orbitally degenerate ($J = 5/2$) periodic Anderson model for both the metallic and insulating cases. The effect of the correlation between $f$-electrons is investigated using the self-consistent second-order perturbation theory. The main results are as follows. (i) Sixfold degenerate model: both $\chi_P$ and $\chi_V$ are enhanced by a factor of $1/z$ ($z$ is renormalization constant). (ii) Nondegenerate model: only $\chi_P$ is enhanced by $1/z$. Thus, orbital degeneracy is indispensable for the enhancement of $\chi_V$. Moreover, orbital degeneracy reduces the Wilson ratio and stabilizes the nonmagnetic Fermi liquid state.

KEYWORDS : heavy fermion system, Kondo insulator, magnetic susceptibility, Fermi liquid theory, orbital degeneracy

In general, the Ce-compound heavy fermion (HF) system possesses one electron per Ce-atom. Due to strong $t$-$s$ coupling, the lowest $f^1$-eigenstate has the angular momentum $J = 5/2$. In real materials, the sixfold states in $J = 5/2$ are split into three Kramers doublets (KDs) by a weak crystalline electric field (CEF). The higher KDs strongly affect the electronic structure because $f$-level splitting is, at most, $\sim O(10^2)\text{K}$. In HF systems, orbital degeneracy (or orbital freedom) has an essential effect on many physical properties. For example, it causes the anomalous Hall effect, or a quadrupole long range order. Another interesting feature of the orbitally degenerate systems is the existence of the Van-Vleck susceptibility $\chi_V$, which is brought about by magnetic interband excitation. $\chi_V$ should bring about large residual magnetic susceptibility in many Kondo insulators (CeNiSn, Ce$_2$Bi$_4$Pt$_3$, etc), or in superconducting compounds below $T_C$ (UPt$_3$, UBe$_{13}$, etc). Thus, orbital freedom causes various nontrivial phenomena in HF compounds, and should be studied in earnest. In addition, orbital freedom also exists in many d-electron systems (V- or Mn-oxide compounds, etc).

In this work, we study the orbitally degenerate ($J = 5/2$) periodic Anderson model ($J = 5/2$ PAM), which is the appropriate model for Ce-compounds. In this orbitally degenerate model, the magnetic susceptibility $\chi$ is composed of the Pauli term ($\chi_P$) and the Van-Vleck term ($\chi_V$). It is proved that $\chi_V$ appears in the case of $[H, M] \neq 0$, where $H$ and $M$ are the Hamiltonian and the magnetization operators, respectively. Generally, the analysis of $\chi_V$ in the interacting system is not easy. In the mean-field approximation (Gutzwiller approximation), where we replace $V_0$ with $\sqrt{z} \cdot V_0$ ($z \ll 1$ is the renormalization constant), both $\chi_P$ and $\chi_V$ are enhanced by $1/z$. However, this result depends strongly on the assumption that the $f$-electron level $E_f$ is renormalized by $1/z$ to $E_f^*$, which is never trivial and should be confirmed in detail. For $U > 0$, it is also nontrivial whether the quasi-particle like spectrum at $E_f^*$ remains coherent or vanishes to become incoherent.

The aim of this work is to study the electronic structure and the magnetic susceptibility of $J = 5/2$ PAM beyond the mean-field theory. Here, we perform the numerical calculation by using the self-consistent second-order perturbation theory (SC-SOPT) with respect to (w.r.t.) $U$. In principle, (SC-)SOPT is valid only for the weak-coupling regime. Nonetheless, it is known to be very useful for discussing strong coupling properties of, e.g., the impurity Anderson model and the Hubbard model. In addition, we use the local approximation ($d = \infty$ approximation), which is known to work well for nondegenerate PAM. We check the higher correction terms w.r.t. $1/d = 5/2$ PAM, and find that the local approximation works better due to the orbital degeneracy.

We find that, for the six-fold degenerate case (no CEF case), both $\chi_P$ and $\chi_V$ are enhanced by the enhancement factor at the Fermi energy within SC-SOPT. This result coincides with that of our analytical study on the basis of the Fermi liquid theory for the strong correlation region, given in ref. This numerical work is complementary to the analytical work in ref. We also confirm the renormalization of $E_f$ towards the Fermi energy, $\mu$. This result is consistent with the enhancement of $\chi_V$.

The Hamiltonian for $J = 5/2$ PAM is given as

$$H = \sum_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{kM} E_f f_{kM}^\dagger f_{kM} + \sum_{Mk\sigma} (V_{kM\sigma}^e f_{kM}^\dagger c_{k\sigma}$$

$$+ h.c.) + \frac{U}{2} \sum_{kk'qM\neq M'} f_{k-qM}^\dagger f_{k'qM}^\dagger f_{k'qM} f_{kM},$$

where $c_{k\sigma}^\dagger (f_{kM}^\dagger)$ is the creation operator of a conduction electron ($f$-electron), and $c_{k\sigma}$ is the $k$-dependent spectrum for conduction electrons. $M$ is the eigenvalue of $J_z$ ($M = 5/2, 3/2, \cdots, -5/2$). The $c$-$f$ mix-
ing potential $V_{kMσ}$ in (1) is given by $V_{kMσ} = V_0 \cdot \sqrt{4π/3} \sum_m -σ(7/2 - Mσ)/7δ_{m,M−σ,σ}Y_{m,σ}^{\sigma}(θ_k, φ_k)$, where $Y_{m,σ}^{\sigma}(θ_k, φ_k)$ is the spherical harmonic function. We impose the Zeeman energies for only $f$-electrons, $E_M^f = E_f + gµ_B M \cdot H$, where $g$ is Lande’s $g$-factor ($g = 6/7$ for $J = 5/2$), and we set $µ_B = 1$ hereafter. The electronic structure in the case of $U = 0$ is shown schematically in Fig. 1. ∆− = $E_k = 2π$, and $E_k$ is the quasi-particle spectrum, which is the bonding orbit brought about by $ε_k$ and $E_f$. There remain fourfold nonbonding orbits at $E_f$.

The local Green’s function $g(ω)$, with which the self-energy $Σ(ω)$ is constructed in the local approximation, is given at $H = 0$ by

$$g(ω) = \frac{1}{3}G(ω) + \frac{2}{3}G^f(ω), \quad \text{(2)}$$

$$G(ω) = \frac{1}{N} \sum_k (1/Γ_k(ω) - 3V^2/(ω + µ - ε_k))^{-1}, \quad \text{(3)}$$

$$G^f(ω) = (ω + µ - E_f - Σ(ω))^{-1}. \quad \text{(4)}$$

and the $ω$-dependent renormalization factor in the Fermi liquid theory is

$$z(ω) = (1 - \partial Σ_M(ω)/∂ω)^{-1}, \quad \text{(5)}$$

This $ω$-dependence is neglected in the mean-field treatment. Note that both $g(ω)$ and $Σ(ω)$ are diagonal and independent of $M$ in the case of no CEF. Considering the Pauli principle, the self-energy by SC-SOPT in the local approximation is given by

$$Σ(ω) = \frac{5U}{6} n_f - 5U^2 \int \frac{dεdε'}{4π^2} g(ε)g(ε')g(ω + ε - ε') \quad \text{(6)}$$

at $T = 0$. Figure (2) shows the calculated self-energy $Σ(ω)$. The $ω$-dependence of $Σ(ω)$ is complicated, and the energy region, where renormalization takes place by a factor of $1/z(ω) > 1$, is limited to within $2T_0$ from the Fermi energy. (We call $T_0$ the coherent energy in the system.) These features are not derivable from the mean-field treatment.

Figure (3) shows the $f$-electron density of states (DOS), $ρ(ω) = -\frac{1}{π} \text{Im} g(ω + iδ)$, obtained by using (2), (3) and (4). We also introduce the DOS for the nonbonding orbits, $ρ^f(ω) = -\frac{1}{π} \text{Im} G^f(ω + iδ)$. In the numerical calculation, we imposed weak $k$-dependence on $E_f$ for convenience. The noteworthy points are as follows. (i) $E_f$ is renormalized toward $µ$ to be $E_f^*$, and its weight is reduced. We find that the relation $|E_f^* - µ| ≈ z(0) |E_f - µ|$ holds well for smaller $U$ where SC-SOPT is adequate. (ii) Broad satellites (or shoulders) are formed on both sides of $µ$, which are attributed to Mott-Hubbard excitation. Large part of $ρ^f(ω)$ is transferred to the satellites, which is localized at $E_f$ in the case of $U = 0$. This feature is not derivable from the mean-field theory. The renormalization of $E_f$ observed in our calculation means that $E_f^*$ lies inside the coherent region, and $χ_f$ is expected to be enhanced.

Next, we calculate the uniform magnetic susceptibility for the following two typical cases.

(i) Case 1 (there is no CEF and three KDs are degenerate): In the previous work based on the Fermi liquid theory, we found the following result in the strong coupling region:

$$χ/χ^0 = 1/z_H(0) \quad \text{for the metallic case}, \quad \text{(7)}$$

$$χ/χ^0 = 1/z_H(Δ^∗) \quad \text{for the insulating case}, \quad \text{(8)}$$

$$1/z_H(ω) = 1 + \frac{1}{gM H} \frac{∂}{∂H} Σ_M(ω), \quad \text{(9)}$$

whose deviations are of order $O(ζ^0)$, and $1/z_H(ω)$ is the magnetic enhancement factor in the Fermi liquid theory. In general, $1/z(0) ≲ 1/z_H(0)$ through the Landau parameter $F^0_M < 0$. Thus, $χ_{\text{insulator}} = χ_V$ is enhanced. In the metallic state, total susceptibility $χ$ is given by the $k$-limit of the dynamical susceptibility $χ_k(ω)$ (i.e., $χ = \lim_{ε→0} χ_k(0)$) and $χ_V$ should be defined by the $ω$-limit of $χ_k(ω)$. In the insulating state, the two limits coincide.

Using the above relations, we calculate $χ$ in a manner consistent with the self-energy obtained by SC-SOPT, so as to satisfy the conservation law (i.e., the Ward identity) proposed in ref. [8]. Furthermore, we compare it with $1/z_H(0)$ (or $1/z_H(Δ^∗)$) derived using SC-SOPT. Here, the result in Fig. 5. We can see that relations (7) and (8) hold irrespective of the strength of the correlation. Moreover, Fig. (a) clearly shows the enhancement of $χ_V$ in the metallic state.

(ii) Case 2 (where only the lowest KD is active due to strong CEF): The system reduces to the familiar non-degenerate PAM. In this model, $χ_V$ is finite unless the $g$-values for $f$-electrons ($g_f$) and for $c$-electrons ($g_c$) are the same. Here, we set $g_c = 0$. The result of the mean-field approximation is

$$χ^p = g^2_f \cdot ρ(0)/z, \quad \text{(10)}$$

$$χ_V = g^2_f \cdot ρ_c \cdot \text{cot}^{-1}(√z/V/(µ - ε_k)), \quad \text{(11)}$$

where $ρ(ω)$ ($ρ_c$) is the $f$($c$)-electron DOS. $χ^p = \chi^U_{\text{insulator}} = χ^U_{\text{insulator}}$ because $ρ(0) = V^2/(E_f - µ)^2 \cdot ρ_c$. Thus, the enhancement of $χ_V$ is of the order $O(1)$ even in the mean-field approximation. This behavior is recognized more accurately in quantum Monte Carlo simulation within the $d = ∞$ approximation. Here, we calculate $χ$ and $1/z_H$ for the insulating state using SC-SOPT, and show the result in Fig. (4). We can see that $χ$ deviates from...
$1/z_H$ in the $U^2$-term. Especially, the sign of curvature of $\chi$ w.r.t. $U$ and that of $1/z_H$ are opposite.

Here, we compare several properties between case 1 and case 2. The sign of the curvature of $\chi_V$ w.r.t. $U$ are opposite in the two case. Thus, interestingly, orbital degeneracy is indispensable for the large enhancement of $\chi_V$. Next, we discuss the coherent energy $T_0$ for both cases (see Fig. 3). We can see that $T_0$ in case 1 (sixfold degenerate) is much larger than that in case 2 (twofold degenerate) if $1/z$ is the same in both cases. Roughly speaking, within SC-SOPT, we can see that $T_0/z \sim |E_f - \mu|$ for case 1 and $T_0/z \sim |\Delta_+ - \mu|$ for case 2 in the metallic system, where $E_f$ and $\Delta_-$ are not renormalized values.

This result indicates that the paramagnetic state is more stable in case 1. Finally, we discuss the Wilson ratio $R$ for both cases, where $R = (\chi/\chi^{U=0}) \cdot z$ in the local approximation. In Fig. 2, we plot the $U$-dependence of $R$ derived using SC-SOPT for both case 1 and case 2. In case 1, $R$ increases much more slowly w.r.t. $U$. As a result, the orbital degeneracy stabilizes the paramagnetic state against (ferro-)magnetic instability. This tendency is consistent with the discussion on $T_0$ above, and it is also found within the Gutzwiller approximation.

In this work, we have studied the $f$-electron DOS and $\chi$ in $J = 5/2$ PAM by applying the SC-SOPT, in order to study many-body effects beyond the mean-field approximation. We showed that the Coulomb interaction drastically changes the whole electronic structure. This change causes the enhancement of $\chi_V$, which is the interband contribution away from the Fermi level. In Fig. 3, we found that the relations (6) and (8) hold with high accuracy, which means both $\chi_F$ and $\chi_V$ are enhanced by the magnetic enhancement factor, $1/z_H$. This result coincides with our 1996 analytical study based on the Fermi liquid theory. Interestingly, the enhancement of $\chi_V$ is closely related to the orbital degeneracy because it is not observed in nondegenerate PAM (see Fig. 5). Moreover, comparing the Wilson ratio between $J = 5/2$ PAM and nondegenerate PAM, we can see within the scope of SC-SOPT that orbital degeneracy stabilizes the nonmagnetic state.

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FIG. 2. The self-energy of the metallic $J = 5/2$ PAM by SC-SOPT for $U = 0.3$, where $1/z(0) = 1.53$. For $|\omega| < 0.02$, the relation $\text{Im} \Sigma A(\omega) = U^2 \cdot (5\pi/2) \rho(0)^3 \cdot \omega^2 \approx 2.4 \cdot \omega^2$ is satisfied. Other parameters are $\epsilon_k = -1 + 2 \cdot (k/\pi)^3$, $V_0 = 0.4$, $E_f = -0.25$ and $\mu = -0.38$. In each stage of self-consistent calculation, we choose $E_f$ so that $\mu$ is unchanged. Note that $E_f^* \approx 0.1 < T_0$ (see Fig. 3).

FIG. 3. The $f$-electron DOS of the metallic $J = 5/2$ PAM obtained by SC-SOPT for (a) $U = 0$ and (b) $U = 0.3$, where $1/z(0) = 1.53$. Other parameters are the same as in Fig. 2. We can see that $\rho'(\omega)$ for $U = 0.3$ is spread into a wider energy range. $\rho(0)$ is unchanged by $U$ in the local approximation.

FIG. 4. In case 1 (sixfold degenerate case): $\chi$, $1/z_H$ and $1/z$ obtained using SC-SOPT for (a) the metallic case and for (b) the insulating case. For (a), we used the same parameters as those in Fig. 2. For (b), parameters are $\epsilon_k = -1 + 2 \cdot (k/\pi)^3$, $V_0 = 0.4$ and $E_f = 0$. We also imposed $\mu = (\Delta^*_c + E_f^*)/2$ which should be satisfied in the case of a pure insulator at $T = 0$.

FIG. 5. In case 2 (twofold degenerate case): $\chi$, $1/z_H(0)$ and $1/z(0)$ obtained by SC-SOPT for the insulating case. Here we used a symmetric PAM ($\epsilon_k = -1 + 2 \cdot (k/\pi)^3$, $V_0 = 0.4$, $E_f = \mu = 0$).
FIG. 6. $U$-dependence of the Wilson ratio obtained using SC-SOPT for case 1 ($R_{6\text{-fold}}$) and for case 2 ($R_{2\text{-fold}}$). In both cases, we used the same parameters (and $\rho(0)$) as those in Fig 2.