Variational theory for a finite-U periodic Anderson model: Application to heavy-electron materials

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We exhibit a variational wave function for the orbitally nondegenerate Anderson lattice model, which incorporates the effects of onsite Coulomb interaction to deal with essentially metallic systems. The average occupation in the correlated orbitals, the renormalized hybridization matrix element, and the mass enhancement are calculated as a function of Coulomb interaction U and the hybridization matrix element V. Our results for the $U\rightarrow\infty$ limit are in agreement with the existing results for the infinite-U problem. We show that the infinite-U approximation is a good approximation for a class of materials with $U>U_c$ and $V<V_c$. The calculation of the effective mass m^* in the heavy-fermion regime shows that it becomes large mainly because of the small hybridization of f electrons with the conduction band rather than because of large Coulomb correlations. The advantages of our approach are briefly discussed.

There has been, of late, a renewed interest in the periodic Anderson model (PAM), triggered mainly by the remarkable properties of the so-called heavy-electron materials (HEM's).[1-3] This model consists of a conduction band, correlated f-electron states localized at lattice sites, and a hybridization (V) between f and the conduction-band electron states. In the simplest form of the model, only the spin degeneracy is taken into account. Various approaches have been used to study the problem. Owing to the large Coulomb interaction (U), application of a perturbation theory does not look like a promising path (see, however, Ref. 4). Including U in the unperturbed part of the Hamiltonian and treating V as a perturbation, is also beset with technical difficulties.[5] Variational methods[6] offer an attractive possibility for treating the large-U periodic Anderson model, since they are nonperturbative in both U and V. In most of the existing approximations the role of U is not so transparent. In this paper we do the following. (i) We present a simple variational method to deal with metallic systems, which brings out the role of U in a very transparent manner. A close correspondence is maintained between the $U\neq 0$ and the $U=0$ cases. Owing to the ease with which calculations are handled by using the variational wave function, the method is amenable to further modifications and/or extensions. (ii) We estimate values of various physical quantities for a range of values of U and V. (iii) We show that the infinite-U approximation is a good approximation to describe a class of systems having $U>U_c$ and $V<V_c$. (iv) The enhancement of effective mass in the heavy-fermion regime is mainly due to small values of V rather than large values of U.

The Hamiltonian for the orbitally nondegenerate periodic Anderson lattice model is given by

$$H=\sum_{k,\sigma}\epsilon_k c_{k\sigma}^{\dagger}c_{k\sigma}+\sum_{i,\sigma}E_f\hat{n}_{fi\sigma}+V\sum_{i,\sigma}(c_{i\sigma}^{\dagger}f_{i\sigma}+\text{H.c.})+U\sum_i\hat{n}_{fi\uparrow}\hat{n}_{fi\downarrow}\ . \tag{1}$$

The first two terms describe the broad conduction band and the nondispersive f level, respectively. The third term describes the hybridization between f and conduction electrons. The last term refers to the Coulomb interaction between f electrons on the same site.

For $U=0$, Hamiltonian (1) can be diagonalized by the canonical transformation

$$\begin{bmatrix}c_{k\sigma}\\ f_{k\sigma}\end{bmatrix}=\begin{bmatrix}\beta'_{k\sigma} & \alpha'_{k\sigma}\\ \alpha'_{k\sigma} & -\beta'_{k\sigma}\end{bmatrix}\begin{bmatrix}u'_{k\sigma}\\ l'_{k\sigma}\end{bmatrix} \tag{2}$$

to get the two hybridized bands

$$\xi_k^{u,l}=\tfrac{1}{2}(\epsilon_k+E_f)\pm\tfrac{1}{2}[(\epsilon_k-E_f)^2+4V^2]^{1/2}\ , \tag{3}$$

where ϵ_k and E_f are the nonhybridized and uncorrelated conduction band and f-level energy, respectively. When the total number of electrons per site is taken as $1<n<2$, the normalized ground-state wave function is given by

$$|\psi_{U=0}\rangle=\prod_{k<k_f,\sigma}l_{k\sigma}'^{\dagger}|0\rangle\ . \tag{4}$$

For the finite-U case, we construct the trial ground-state wave function around the uncorrelated wave function by introducing the effect of U through a projection operator P_i,[7,8]

$$|\psi_c\rangle=\prod_i P_i|\psi_{uc}\rangle\ , \tag{5}$$

with

$$|\psi_{uc}\rangle=\prod_{k<k_f,\sigma}(\alpha_{k\sigma}c_{k\sigma}^{\dagger}-\beta_{k\sigma}f_{k\sigma}^{\dagger})|0\rangle\ . \tag{6}$$

Here we have taken for $|\psi_{uc}\rangle$ the form (4) with $\alpha'_{k\sigma}$ and $\beta'_{k\sigma}$ replaced by variational functions $\alpha_{k\sigma}$ and $\beta_{k\sigma}$. The simplest possible choice for the projection operator, wide-

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ly used in the literature for $U=\infty$, is $P_i=(1-\hat{n}_{fi\uparrow}\hat{n}_{fi\downarrow})$, which forbids double occupancy. However, the problem with this form of P_i is that it violates the requirement of conservation of total electron density per site,

$$\langle n_{fi\sigma}+c_{i\sigma}^{\dagger}c_{i\sigma}\rangle_c=\langle n_{fi\sigma}+c_{i\sigma}^{\dagger}c_{i\sigma}\rangle_{uc}\ , \tag{7}$$

where $\langle O\rangle_c$ means $\langle\psi_c|O|\psi_c\rangle/\langle\psi_c|\psi_c\rangle$ and $\langle O\rangle_{uc}$ means $\langle\psi_{uc}|O|\psi_{uc}\rangle$. In view of the requirement (7), we choose

$$P_i=1+\sum_{\sigma}s_{\sigma}\hat{n}_{fi\sigma}-\left[(1-d)+\sum_{\sigma}s_{\sigma}\right]\hat{n}_{fi\uparrow}\hat{n}_{fi\downarrow}\ , \tag{8}$$

with d as a variational parameter. P_i projects out the doubly occupied sites to an extent that we determine variationally. From Eq. (7) we obtain

$$s_{\sigma}=\left[\frac{1-n_{f\sigma}-d^2n_{f-\sigma}}{1-n_f}\right]^{1/2}-1\ , \tag{9}$$

where

$$n_{f\sigma}=\frac{1}{N}\sum_{k<k_f}\beta_{k\sigma}^2 \tag{10}$$

and $n_f=n_{f\uparrow}+n_{f\downarrow}$. Here we have used the translational invariance.

The ground-state energy of (1) is

$$E_g=\frac{\langle\psi_c|H|\psi_c\rangle}{\langle\psi_c|\psi_c\rangle}\ . \tag{11}$$

Again assuming the translational invariance, the density matrices are evaluated using (5),

$$\langle c_{i\sigma}^{\dagger}f_{i\sigma}\rangle_c=R_{\sigma}\langle c_{i\sigma}^{\dagger}f_{i\sigma}\rangle_{uc}\ , \tag{12a}$$

$$\langle f_{i\sigma}^{\dagger}c_{i\sigma}\rangle_c=R_{\sigma}\langle f_{i\sigma}^{\dagger}c_{i\sigma}\rangle_{uc}\ , \tag{12b}$$

$$\langle f_{i\uparrow}^{\dagger}f_{i\downarrow}f_{i\uparrow}^{\dagger}f_{i\downarrow}\rangle_c=\frac{d^2(1-n_f)}{(1-n_f)+(1-d^2)n_{f\uparrow}n_{f\downarrow}}\times\langle f_{i\uparrow}^{\dagger}f_{i\downarrow}f_{i\uparrow}^{\dagger}f_{i\downarrow}\rangle_{uc}\ . \tag{12c}$$

Here R_{σ} is the renormalization factor for V, and is given by

$$R_{\sigma}=\frac{[(1-n_{f-\sigma})(1+s_{\sigma})+dn_{f-\sigma}(1+s_{-\sigma})](1-n_f)}{(1-n_f)+(1-d^2)n_{f\sigma}n_{f-\sigma}}\ . \tag{13}$$

In Ref. 9 the expressions for $\langle c_{i\sigma}^{\dagger}f_{i\sigma}\rangle_c$ and $\langle f_{i\sigma}^{\dagger}c_{i\sigma}\rangle_c$ do not satisfy the identity $\langle c_{i\sigma}^{\dagger}f_{i\sigma}\rangle=\langle f_{i\sigma}^{\dagger}f_{i\sigma}\rangle^*$. Such a difficulty does not appear in the present formulation as can be seen from (12a) and (12b).

Minimization of the ground-state energy functional [obtained by using (6), (11), and (12)] by imposing the constraint $\alpha_{k\sigma}^2+\beta_{k\sigma}^2=1$ through a Lagrange multiplier $\lambda_{k\sigma}$, gives us α_k, β_k, and λ_k. Using α_k, β_k, λ_k, and (11), the ground-state energy is given by

$$E_g=\frac{1}{N}\sum_{k<k_f}\{(\epsilon_k+E_f-\mu)-[(\epsilon_k-E_f+\mu)^2+4\tilde{V}^2]^{1/2}\}+\mu n_f+Ud^2\frac{n_f^2(1-n_f)}{(2-n_f)^2-d^2n_f^2}\ , \tag{14}$$

where $n_{f\uparrow}=n_{f\downarrow}=n_f/2$ is assumed, $\tilde{V}=VR$, and μ is the shift in the f level,

$$\mu=4V\frac{\partial R(n_f,d)}{\partial n_f}\frac{1}{N}\sum_{k<k_f}\frac{\tilde{V}}{[(\epsilon_k-E_f+\mu)^2+4\tilde{V}^2]^{1/2}}-Ud^2\frac{\partial}{\partial n_f}\left[\frac{n_f^2(1-n_f)}{(2-n_f)^2-d^2n_f^2}\right]\ . \tag{15}$$

Our method of construction of the ground-state energy functional is similar to that used in Refs. 9 and 11.

Substituting β_k obtained through energy minimization in Eq. (10), the average occupation in correlated orbitals n_f is given by

$$n_f=\frac{1}{N}\sum_{k<k_f}\left[1+\frac{\epsilon_k-E_f+\mu}{[(\epsilon_k-E_f+\mu)^2+4\tilde{V}^2]^{1/2}}\right]\ . \tag{16}$$

We solve the self-consistent equations (15) and (16) and $\partial E_g/\partial d=0$ for n_f, μ, and d. Although our calculations are valid for arbitrary dimensions, for simplicity we assume the conduction band to be linearly ranging from $-b$ to $+b$. In the calculations we have taken $2b=8$ eV and the nonhybridized and uncorrelated Fermi energy of conduction electrons, $\epsilon_F=3$ eV. We have investigated various cases $0<V/|E_f|\leq 1$, $1<n<2$, with $0\leq U<\infty$.

The results for the parameter d, which is the reduction factor for the doubly occupied sites and n_f, are plotted as a function of $U/|E_f|$ in Fig. 1 for $V/|E_f|=1.0$, 0.6, and 0.1. We have taken $n=1.75$ and $E_f=-1$ eV. We find, at $U=0$, $d=1$. Using this in Eqs. (8), (9), (13), and (15), we get $P_i=1$, $R=1$, $\mu=0$, and, therefore, $\tilde{V}=V$. Consequently, $|\psi_c\rangle$ reduces to $|\psi_{U=0}\rangle$ and we exactly reproduce the hybridized bands (3) for the uncorrelated problem. When $U\to\infty$, $d\to 0$. This simply reflects the fact that at infinite U doubly occupied sites are forbidden. Substituting $d=0$ in Eqs. (9) and (13), the renormaliza-

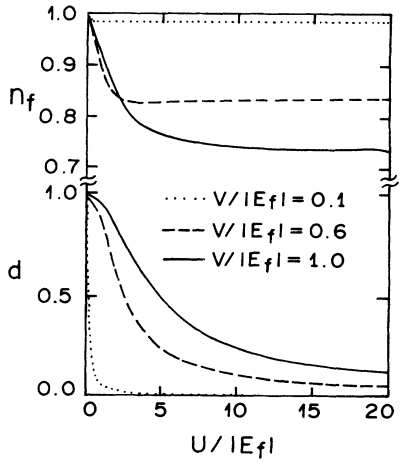

FIG. 1. The average occupation in the correlated orbitals n_f and the reduction factor for the doubly occupied sites d as a function of $U/|E_f|$ for $n=1.75$, $E_f=-1$ eV, and $V/|E_f|=1.0$, 0.6, and 0.1.

tion factor R is given by

$$R=\left[\frac{1-n_f}{1-n_f/2}\right]^{1/2}, \tag{17}$$

which is in agreement with the results obtained by various authors[7,9-12] for the $U=\infty$ case. In the region $0<U<\infty$, d decreases with increasing $U/|E_f|$ for all values of $V/|E_f|$. Similar behavior is observed for n_f, but instead of approaching zero as $U\to\infty$, as is the case for d, it saturates to a nonzero value.

Underlying physics is simple to understand: For the uncorrelated case, there is no restriction on the presence of doubly occupied f sites and the f level, which lies below the Fermi surface, plays no role. In the correlated case, the presence of a double occupied f site raises the energy by an amount U. However, the energy is lowered because of the hybridization process, through which the f electrons change over to the d electrons. The competing roles of U and V lead to a variation of n_f and d, with U and V, shown in Fig. 1.

As in other models, we have an enhancement of effective mass of the carrier fermions. The density of quasiparticle states is given by

$$\rho(E)=\sum_k \delta(E_f-f(\epsilon_k))=1+\frac{\tilde{V}^2}{(E-E_f+\mu)^2}, \tag{18}$$

where

$$f(\epsilon_k)=\tfrac{1}{2}(\epsilon_k+E_f-\mu)-\tfrac{1}{2}[(\epsilon_k-E_f+m)^2+4\tilde{V}^2]^{1/2}. \tag{19}$$

The effective-mass enhancement obtained from Eq. (18) is given by

$$m^*=\frac{m_{(V\neq 0,U\neq 0)}}{m_{(V=0,U=0)}}=\rho[f(\epsilon_F)]\sim\frac{1}{R^2V^2}. \tag{20}$$

This can also be seen from Eq. (14), which for $U\to\infty$ (i.e., $d\to 0$) describes the energy of two noninteracting bands hybridized by a matrix element RV. The enhancement of the effective mass can be understood as follows: When $U=0$ and $V=0$, the Hamiltonian (1) describes a noninteracting conduction band and an f level. We denote the effective mass of the carrier fermions for the noninteracting case as $m_{(V=0,U=0)}$. For $V\neq 0$, the eigenvalues of $H(U=0)$ describe two quasiparticle bands [see Eq. (3)]. The lower quasiparticle band has predominantly f character in the neighborhood of the Fermi energy. Using (3) and (18), the effective mass is given by

$$\frac{m^*_{(V\neq 0,U=0)}}{m_{(V=0,U=0)}}=\frac{1}{V^2}. \tag{21}$$

For the correlated problem, the effective mass of the carrier fermions m^* is given by Eq. (20). The enhancement of effective mass of the carrier fermions produced by Coulomb interactions is

$$m^*_{\text{Coul}}=\frac{m_{(V\neq 0,U\neq 0)}}{m_{(V\neq 0,U=0)}}=\frac{1}{R^2}. \tag{22}$$

Figure 2 presents $m^*(V/|E_f|)^2$ as a function of $U/|E_f|$ for $V/|E_f|=1.0$, 0.6, and 0.1. It shows that (i) m^* increases up to a critical value of U, say, U_c, beyond which it almost saturates. The critical value U_c decreases with decreasing V. This implies that the infinite-U approximation is a good approximation to study a class of materials with $U>U_c$ and $V<V_c$. (ii) m^* increases by a factor of 3000 by decreasing $V/|E_f|$ from $V/|E_f|=1.0$ to 0.1 for a fixed value of $U/|E_f|$ ($U/|E_f|=1$ in this calculation). Therefore, m^* becomes large mainly because of the small values of V rather than the large values of U.

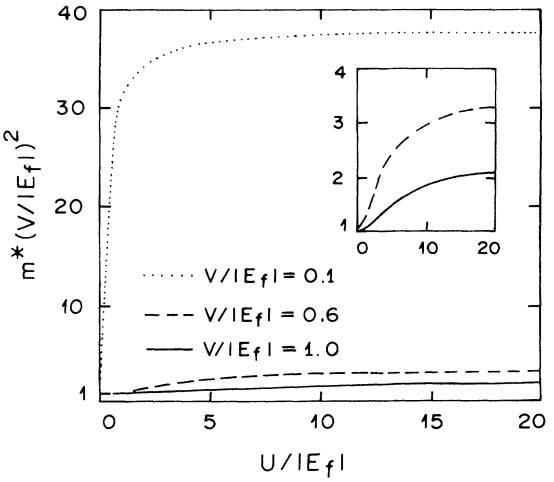

FIG. 2. The effective mass of the carrier fermions m^* as a function of $U/|E_f|$ for $n=1.75$, $E_f=-1$ eV, and $V/|E_f|=1.0$, 0.6, and 0.1.

From Fig. 2, we find that for small $V/|E_f|$ (say 0.1), the effective mass rapidly saturates and the infinite-U approximation would be valid for all $U/|E_f|>5$ eV. This, however, cannot be said for a case where $V/|E_f|=0.6$. For this value of $V/|E_f|$, we see from Fig. 2 (inset) that even for $U/|E_f|=20$ the m^* curve has not saturated.

For heavy-electron materials, it is known[13] that $U=5$–6 eV for Ce-based HEM's and $U\sim 2$ eV for uranium-based materials. In these materials there is no reliable way of estimating V; however, crude estimates[2] put its value in the range $0.2<V<0.6$ eV. For these values of U and V, the results in Fig. 2 show that the infinite-U treatment of the problem does not provide an adequate description of the physics of heavy-electron materials.

In this paper we have presented a simple variational method for the orbitally nondegenerate Anderson lattice model. This approach is much simpler than the Gutzwiller-type variational method used in the literature. The application of the Gutzwiller method to deal with metallic systems ($1<n<2$) presents difficulties and one has to use some uncontrolled approximations. We have shown through this paper how such calculations can be handled more easily by using the variational wave function proposed by us. We have shown the variation of n_f, the effective hybridization matrix element, and the enhancement of effective mass of the carrier fermions with U and V. We have discussed the relevance of the infinite-U approximation to the study of real systems. In

the absence of any numerically exact results for the asymmetric finite-U PAM, it will be interesting to compare our results with quantum Monte Carlo calculations, which hopefully will become possible in the near future. It would be interesting to study the magnetic properties of the ground state by including the possibility of an antiferromagnetic phase. The method can be easily extended to describe the transition metals and their compounds, where the correlations in the d orbitals are much stronger than in the sp orbitals by introducing dispersion in the correlated orbitals.

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[1]See Proceedings of the International Conference on Strongly Correlated Electron Systems, Sendai, Japan [Physica B **186-188**, 1083 (1993)].

[2]N. Grewe and F. Steglich, in *Handbook on the Physics and Chemistry of Rare Earths,* edited by K. A. Gschneider, Jr. and L. Eyring (North-Holland, Amsterdam, 1991), Vol. 14, p. 343.

[3]A. C. Hewson, *The Kondo Problem to Heavy Fermions* (Cambridge University Press, New York, 1993).

[4]H. Schweitzer and G. Czycholl, Z. Phys. B **79**, 377 (1991), and references therein.

[5]N. Grewe, Solid State Commun. **50**, 19 (1984).

[6]D. Volhardt, in *Interacting Electrons in Reduced Dimensions,* edited by D. Baeriswyl and D. K. Campbell (Plenum, New York, 1989), p. 107.

[7]A. Oguchi, Prog. Theor. Phys. **77**, 278 (1987).

[8]J. Bernasconi, Phys. Kondens. Mater. **14**, 225 (1972).

[9]T. M. Rice and K. Ueda, Phys. Rev. Lett. **55**, 995 (1985); **55**, 2093(E) (1985); Phys. Rev. B **34**, 6420 (1986); T. M. Rice, in *Frontiers and Borderlines in Many-Particle Physics,* edited by R. A. Broglia and J. R. Schrieffer (North-Holland, Amsterdam, 1988), p. 171.

[10]C. M. Varma, W. Weber, and L. J. Randall, Phys. Rev. B **33**, 1015 (1986).

[11]V. Z. Vulovic and E. Abrahams, Phys. Rev. B **36**, 2614 (1987).

[12]F. Gebhard and D. Volhardt, in *Interacting Electrons in Reduced Dimensions* (Ref. 6), p. 123.

[13]P. A. Lee *et al.*, Comments Solid State Phys. **12**, 99 (1986).