Antiferromagnetic heavy-fermion and Kondo-insulating states with compensated magnetic moments

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We discuss the magnetic phases in the periodic Anderson model and determine the conditions under which there appears a stable Fermi-liquid state with (i) small antiferromagnetic moment of f electrons, largely compensated by the conduction electrons, (ii) high density of states at the Fermi level, and (iii) almost integer occupation of the f level. We use a modified version of the spin rotationally invariant slave-boson approach, in which a strong nonlinear molecular field acting on f electrons appears already in the saddle-point approximation. For an even number of electrons an antiferromagnetic Kondo insulating state transforms with increasing hybridization into a paramagnetic Kondo insulator. [S0163-1829(97)51346-4]

The heavy-electron Fermi-liquid state in the intermetallic f-electron compounds is modelled with the help of the periodic Anderson model or its derivatives for paramagnetic[1–3] and magnetic[4] cases. Recently, this approach has been applied to the Kondo insulators.[5,6] We analyze the connection between the two states by determining the phase diagram as a function of filling n_e. This also required the determination of the phase boundary between the antiferromagnetic Kondo-insulating state (AKI) with almost compensated magnetic moments (including the crucial negative contribution of conduction electrons) and the paramagnetic Kondo insulator (PKI). In the same manner, we have *complemented* the previous discussion[1–4] of the metallic Kondo-lattice state with the detailed study of the stability of the small-magnetic-moment state resulting from magnetic screening of f electrons both by themselves and by the conduction electrons. In brief, we introduce the Kondo compensating cloud into the heavy-quasiparticle picture.

The method we use is the rotationally invariant slave-boson approach[7] modified slightly[8] to correctly obtain the fermion quasiparticle energy in an applied magnetic field. This correction leads in a natural manner to a *nonlinear molecular field*, which appears already on the level of saddle-point solution. The field is strong and is absent in any of the one-boson approaches[1–4] unless the effective quasiparticle interactions are taken into account.[9] Only in the limit of weak hybridization V can it be viewed as a molecular field coming from the Schrieffer-Wolff type of interaction (the correction to the atomic limit value is $\sim V^2$). Thus, in the limit of almost compensated moments it cannot be easily resolved into the bare Kondo and the RKKY components,[4] the latter being the higher-order contribution. The picture obtained here is that with growing hybridization the antiferromagnetic kinetic exchange becomes important in the almost half-filled situation. A similar effective field arises in the Hubbard model and its role was discussed separately.[8]

We start from the following representation of the atomic (f) n-electron states on lattice site i: $|0,i\rangle = e_i^\dagger|v\rangle$, $|\mathbf{1},\boldsymbol{i}\rangle = \mathbf{f}_i^\dagger \mathbf{P}_i^\dagger |v\rangle$, and $|2,i\rangle = f_{i\uparrow}^\dagger f_{i\downarrow}^\dagger d_i^\dagger |v\rangle$, where $|v\rangle$ is the abstract vacuum state, $\boldsymbol{\tau}$ stands for the Pauli matrices, $\mathbf{f}_i^\dagger \equiv (f_{i\uparrow}^\dagger, f_{i\downarrow}^\dagger)$ represents the pseudofermion field, and $e_i^\dagger$, $\mathbf{P}_i^\dagger \equiv \frac{1}{2}[p_{i0}^\dagger \mathbb{1} + \mathbf{p}_i^\dagger \cdot \boldsymbol{\tau}]$, and $d_i^\dagger$ represent auxiliary Bose fields obeying the constraints

$$e_i^\dagger e_i + \frac{1}{2}[p_{i0}^\dagger p_{i0} + \mathbf{p}_i^\dagger \cdot \mathbf{p}_i] + d_i^\dagger d_i = \mathbb{1}, \tag{1a}$$

$$\sum_\sigma f_{i\sigma}^\dagger f_{i\sigma} = \frac{1}{2}[p_{i0}^\dagger p_{i0} + \mathbf{p}_i^\dagger \cdot \mathbf{p}_i] + 2 d_i^\dagger d_i, \tag{1b}$$

$$\mathbf{p}_i^\dagger \times \mathbf{p}_i = 0. \tag{1c}$$

As noted before,[8] the last constraint implies the equivalence between the representation of the spin operator via pseudofermions $\mathbf{S}_i = \frac{1}{2}\mathbf{f}_i^\dagger \boldsymbol{\tau} \mathbf{f}_i$, and the representation via slave bosons.[7] Therefore, Eq. (1c) can be rewritten as

$$\sum_{\sigma,\sigma'} f_{i\sigma}^\dagger \boldsymbol{\tau}_{\sigma\sigma'} f_{i\sigma'} - p_{i0}^\dagger \widetilde{\mathbf{p}}_i - \widetilde{\mathbf{p}}_i^\dagger p_{i0} = 0, \tag{2}$$

where $\widetilde{\mathbf{p}}_i = (p_{i1}, -p_{i2}, p_{i3})$. We take (1c) in form (2), as it provides correctly the Zeeman term for the emerging quasiparticles.

The constraints (1a) and (1b) and (2) enter the Lagrangian with the (bosonic) Lagrange multipliers. The radial gauge transformation eliminates the path integration over the phases of the fields e_i, p_{i0} and $\mathbf{p}_i$, which are then transformed into real quantities e_i, q_{i0} and $\mathbf{q}_i$. Explicitly, the partition function for the Anderson lattice model reads

$$Z \equiv \exp(-\beta F) = \int [\mathcal{D}c][\mathcal{D}f][\mathcal{D}e][\mathcal{D}q][\mathcal{D}d][\mathcal{D}\alpha][\mathcal{D}\beta] \times \exp\left(\int_0^\beta (L_F + L_B) d\tau \right), \tag{3a}$$

with

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$$L_F=\sum_{i,\sigma} c_{i\sigma}^{\dagger}\left(\frac{\partial}{\partial\tau}-\mu\right)c_{i\sigma}-\sum_{i,j,\sigma} t_{ij}c_{i\sigma}^{\dagger}c_{j\sigma}$$
$$+\sum_{i,\sigma,\sigma'} V\Bigg[c_{i\sigma}^{\dagger}\Bigg(e_i\frac{1}{2}(q_{i0}\mathbb{1}+\mathbf{q}_i\cdot\boldsymbol{\tau})_{\sigma\sigma'}$$
$$+\frac{1}{2}(q_{i0}\mathbb{1}-\mathbf{q}_i\cdot\boldsymbol{\tau})_{\sigma\sigma'}d_i^{*}\Bigg)f_{i\sigma'}+\text{H.c.}\Bigg]$$
$$+\sum_{i,\sigma,\sigma'} f_{i\sigma}^{\dagger}\left[\left(\frac{\partial}{\partial\tau}-\mu+\varepsilon_f+\beta_{i0}\right)\mathbb{1}+(\boldsymbol{\beta}_i\cdot\boldsymbol{\tau})\right]_{\sigma\sigma'}f_{i\sigma'}, \tag{3b}$$

and

$$L_B=\sum_i\left[\alpha_i\left(e_i^2+|d_i|^2+\frac{1}{2}(q_{i0}^2+\mathbf{q}_i^2)-1\right)-\beta_{i0}\left(\frac{1}{2}(q_{i0}^2+\mathbf{q}_i^2)+2|d_i|^2\right)-2q_{i0}\tilde{\mathbf{q}}_{\mathbf{i}}\cdot\boldsymbol{\beta}_i+U|d_i|^2\right]. \tag{3c}$$

Here $\tilde{\mathbf{q}}_i=(q_{i1},-q_{i2},q_{i3})$, α_i, β_{i0}, and $\boldsymbol{\beta}_i$ are complex, time-dependent fields. The field $\mathbf{q}_i$ gives rise to a spin-dependent renormalization of the hybridization term, whereas nonzero field $\boldsymbol{\beta}_i$ spin-splits the bare f level. Note that the latter enters L_F as an applied magnetic field would do. Therefore it can be regarded as a molecular field, which acts only on f electrons.

In what follows we assume $\boldsymbol{\beta}_i\equiv(0,0,\beta_{i3})$, and $\mathbf{q}_i\equiv(0,0,q_{i3})$. Introducing $q_{i\sigma}=1/\sqrt{2}\ (q_{i0}+\sigma q_{i3})$ we obtain the renormalized hybridization matrix element in the standard form[7]

$$V\rightarrow\tilde{V}_{i\sigma}=V\frac{e_iq_{i\sigma}+d_iq_{i-\sigma}}{\sqrt{1-e_i^2-q_{i-\sigma}^2}\sqrt{1-d_i^2-q_{i\sigma}^2}}. \tag{4}$$

In the saddle-point approximation we first represent the Fermi-liquid state as a homogenous broken-symmetry state with nonzero real values for the fields $e_i\equiv e$, $d_i\equiv d$, $q_{i0}\equiv q_0$, $q_{i3}\equiv q_3$, $\alpha_i\equiv\alpha$, and $\beta_{i3}\equiv\beta_3$. Additionally, taking the Fourier transform to $\mathbf{k}$ space, the Lagrangians (3b) and (3c) reduce to

$$L_F=\sum_{\mathbf{k},\sigma}\left[c_{\mathbf{k}\sigma}^{\dagger}\left(\frac{\partial}{\partial\tau}-\mu+\epsilon_{\mathbf{k}}\right)c_{\mathbf{k}\sigma}+\tilde{V}_\sigma(c_{\mathbf{k}\sigma}^{\dagger}f_{\mathbf{k}\sigma}+\text{H.c.})+f_{\mathbf{k}\sigma}^{\dagger}\left(\frac{\partial}{\partial\tau}-\mu+\varepsilon_f+\beta_0+\sigma\beta_3\right)f_{\mathbf{k}\sigma}\right] \tag{5a}$$

and

$$L_B=N\{\alpha(e^2+d^2+q_\uparrow^2+q_\downarrow^2-1)-\beta_0(q_\uparrow^2+q_\downarrow^2+2d^2)-\beta_3(q_\uparrow^2-q_\downarrow^2)+Ud^2\}. \tag{5b}$$

Assuming the featureless form of the density of states (DOS) in the bare band, we obtain the free-energy functional in the form

$$f\equiv\frac{F}{N}=-\frac{1}{\beta W}\sum_{\sigma,s}\int_{-W/2}^{W/2}\ln[1+e^{-\beta(E_\sigma^{(s)}-\mu)}]d\epsilon+\frac{1}{N}L_F+\mu n_e, \tag{6}$$

where the quasiparticle energies in the hybridized subbands ($s=\pm1$) are

$$E_\sigma^{(s)}=\frac{1}{2}[\epsilon+\varepsilon_f+\beta_0+\sigma\beta_3+s\sqrt{(\epsilon-\varepsilon_f-\beta_0-\sigma\beta_3)^2+4\tilde{V}_\sigma^2}], \tag{7}$$

and $n_e=N_e/N$ is the number of electrons per atom. From the conditions $\partial f/\partial x_j=0$, with $x_j=q_\uparrow,q_\downarrow,d,e,\alpha,\beta_0,\beta_3$, and μ, we determine the *paramagnetic* and *ferromagnetic* states. Of special interest are the conduction-band and the f-level fillings (n_c and n_f, respectively) and their magnetic moments ($m_f=q_\uparrow^2-q_\downarrow^2$, and $m_c=1/N\Sigma_{\mathbf{k},\sigma}\sigma\langle c_{\mathbf{k}\sigma}^{\dagger}c_{\mathbf{k}\sigma}\rangle$).

We consider also an *antiferromagnetic* (AF) saddle-point solution with two interpenetrating sublattices, which is modelled by $q_{i\sigma}=q^u+\sigma q^s\cos(\mathbf{Q}\cdot\mathbf{R}_i)$, with the uniform and staggered parts q^u and q^s, respectively; the latter appears together with the molecular field of the form $\beta_{3i}^s=\beta_3^s\cos(\mathbf{Q}\cdot\mathbf{R}_i)$, where $\mathbf{Q}=(\pi/a,\pi/a,\pi/a)$. Leaving all other fields spatially uniform, we disregard charge-density-wave effects. In this picture the renormalized hybridization (4) can be cast in the form $\tilde{V}_{i\sigma}=\tilde{V}^u+\sigma\tilde{V}^s\cos(\mathbf{Q}\cdot\mathbf{R}_i)$, the f-electron magnetic moment is $m_{fi}=4q^uq^s\cos(\mathbf{Q}\cdot\mathbf{R}_i)$, and the f-level filling is $n_{fi}=2[(q^u)^2+(q^s)^2+d^2]$. In effect, the bosonic part of Lagrangian takes the form

$$L_B=N\{-2\beta_0[(q^u)^2+(q^s)^2+d^2]-4\beta_3^sq^uq^s+Ud^2\}, \tag{8a}$$

and the fermionic part can be written in a compact form as

$$L_F=\sum_{\mathbf{k},\sigma}{}' \mathbf{X}_{\mathbf{k}\sigma}^{\dagger}\left(\frac{\partial}{\partial\tau}-\mu+\mathcal{E}_{\mathbf{k}\sigma}\right)\mathbf{X}_{\mathbf{k}\sigma}, \tag{8b}$$

where the primed summation runs over the reduced (halved) Brillouin zone, $\mathbf{X}_{\mathbf{k}\sigma}^{\dagger}\equiv[f_{\mathbf{k}\sigma}^{\dagger},c_{\mathbf{k}\sigma}^{\dagger},f_{\mathbf{k}+\mathbf{Q}\sigma}^{\dagger},c_{\mathbf{k}+\mathbf{Q}\sigma}^{\dagger}]$ is a four-component vector, and $\mathcal{E}_{\mathbf{k}\sigma}$ is a 4×4 matrix, the explicit form of which is not provided here. We have also assumed a perfect nesting, i.e., $\epsilon_{\mathbf{k}+\mathbf{Q}}=-\epsilon_{\mathbf{k}}$ and a rectangular DOS in the halved conduction band. Finally, the free energy functional in this case is essentially of the form (6). The only differences are that the integration runs from $-W/2$ to 0, and that $E^{(s)}$ denotes four ($s=1,2,3,4$) spin-independent eigenvalues of matrix $\mathcal{E}_{\mathbf{k}\sigma}$ for a given value of $\epsilon_{\mathbf{k}}=\epsilon$. The functional f is optimized with respect to q^u, q^s, d, e, α, β_0, β_3^s, and μ. As in the previous case, the quantities of interest are the f-electron (staggered) moment $m_f^s=4q^uq^s$, the (uniform) f-level occupancy n_f, the conduction-band occupancy n_c, and its staggered moment $m_c^s\equiv1/N\Sigma'_{\mathbf{k},\sigma}\sigma[\langle c_{\mathbf{k}+\mathbf{Q}\sigma}^{\dagger}c_{\mathbf{k}\sigma}\rangle+\langle c_{\mathbf{k}\sigma}^{\dagger}c_{\mathbf{k}+\mathbf{Q}\sigma}\rangle]$. The global quantities (per atom) are $n_e=n_f+n_c$ and the staggered magnetization $m^s=\frac{1}{2}g_fm_f^s+\frac{1}{2}g_cm_c^s$, where g_f and g_c are the corresponding Landé factors.

The detailed numerical analysis of stable solutions encompasses: (a) paramagnetic (P) phase $q_\uparrow=q_\downarrow$, $\beta_3=0$ and the total moment $m=m_f+m_c=0$; (b) weak ferromagnetic

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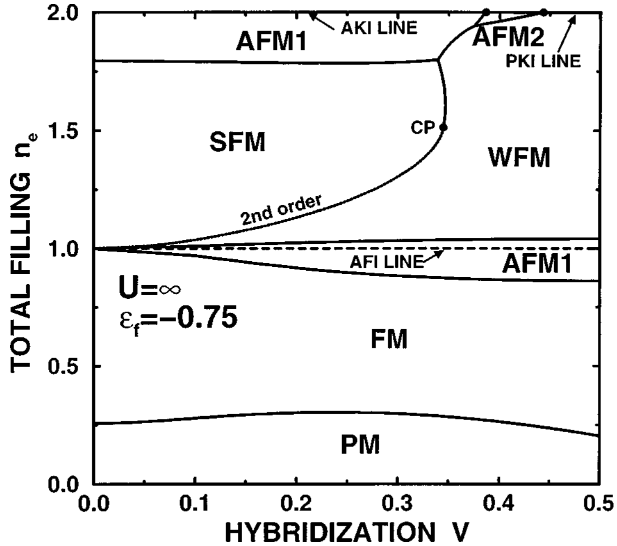

FIG. 1. Phase diagram for the periodic Anderson model. For explanation of the various phases, see main text.

(WF) state $q_\uparrow \neq q_\downarrow$ and $|m| = 2 - n_e$; (c) strong ferromagnetic (SF) state $q_\uparrow \neq q_\downarrow$ and $|m| \rangle 2 - n_e$; and (d) antiferromagnetic states AF1 and AF2, specified below. Also, we attach an additional label, I or M, to the phase label, depending on whether the state is insulating or metallic, respectively. In Fig. 1 we display a representative phase diagram of the system in the filling range $0 \leqslant n_e \leqslant 2$. This diagram comprises the antiferromagnetic-insulator (AFI) line for the total filling per site $n_e = 1$ and the line for $n_e = 2$, along which the antiferromagnetic Kondo insulator (AKI) transforms with increasing hybridization into a paramagnetic Kondo insulator (PKI).[10] There are two antiferromagnetic metallic (AFM) phases, with the band structure inverted with respect to each other (in AFM2 the peak in the density of states dispalyed in Fig. 2 is at the bottom of the second-lowest band). The

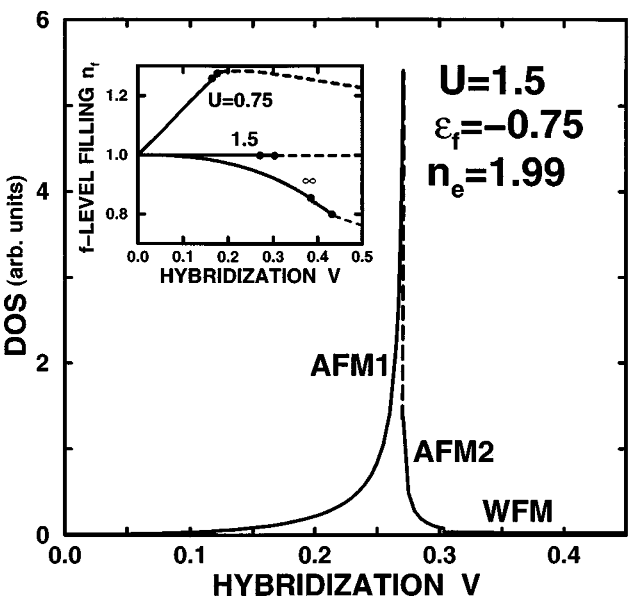

FIG. 2. The density of quasiparticle states at Fermi energy (relative to the value in the bare band, and reduced by a factor 10^{-2}). The inset displays the f-level occupancy for various U. All quantities are a function of bare hybridization V.

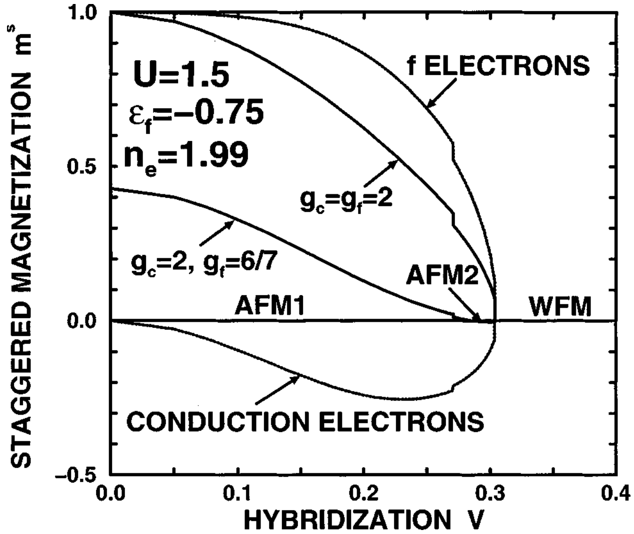

FIG. 3. Sublattice magnetic moments for f and conduction electrons and the total magnetization for the Landé factors $g_f = 2$ and $g_f = 6/7$, respectively.

boundary lines involving AFM phases are of the first order, except on the line for $n_e = 2$, where transitions are continuous. The critical point separating SFM and WFM phases has also been marked. All energies are given in the units of W.

To characterize the nature of the phases we have plotted in Fig. 2 the DOS enhancement of the quasiparticle states relative to the bare-band density of states $(2/W)$, for $n_e = 1.99$, as a function of bare hybridization V. Only in the limit $n_e = 2 - \delta$, with $\delta \ll 1$, do we obtain a stable AFM phase with high density of states. The f-level occupancy is equal to unity for the symmetric Anderson model $(2\varepsilon_f + U = 0)$ and for $\delta = 0$.[5] For $0 < \delta \ll 1$, however, n_f is still very close to unity, as displayed in the inset of Fig. 2. At the same time, the magnetic moment is strongly reduced, as illustrated in Fig. 3, where both the components m_f^s and m_c^s are drawn, together with the total magnetizations (solid lines). Note that m_f^s is almost totally compensated close to the AFM1-AFM2

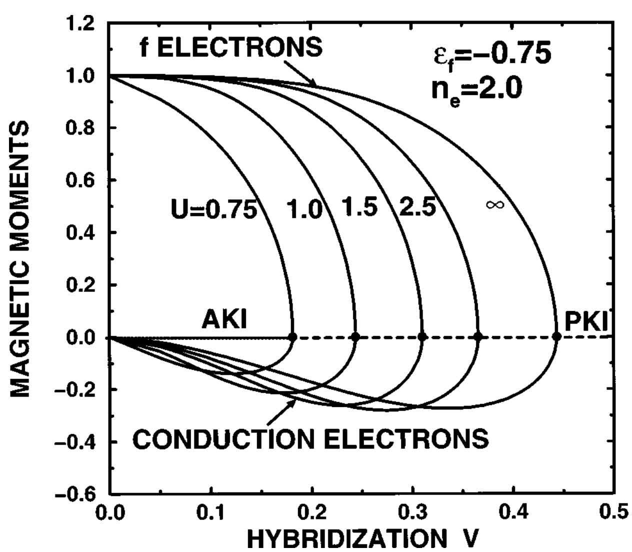

FIG. 4. Magnetic moments (per site) of f and conduction electrons for the Kondo insulator.

boundary, particularly if we take $g_f=6/7$, as is the case for the Ce^{3+} ion. Moreover, the negative polarization m_c^s grows strongly with increasing hybridization, while m_f^s decreases until the almost compensated state is achieved. Thus, our version of the mean-field theory (with the nonlinear molecular field) contains the competition between the Kondo compensation and intersite magnetic interactions introduced by Doniach.[4] However, in the present analysis the f electrons screen themselves, as the sublattice moment decreases with decreasing n_f (cf. inset in Fig. 2). In this respect, the situation is very similar to the disappearance of AF in the Hubbard model in the partially filled-band case. The staggered molecular field limits are $\beta_3^s \sim (\mu-\varepsilon_f)$ for $V\rightarrow 0$, and $\sim |V-V_c|^{1/2}$ for $m_f \rightarrow 0$, where V_c is the critical value of V, below which AF solution exists. Obviously, the Gaussian fluctuations in Bose fields will also introduce the magnetic (RKKY-type) coupling,[4] which, being of the order of T_K,[6] will be substantially weaker than β_3^s. Also, our analysis complements the criteria for the onset of the magnetic Kondo-lattice state derived by Doniach[4] and induces both conduction-electron compensation and the autocompensation of f moments. Note that within the one-boson approach[4] one has to go *beyond* the saddle-point approximation to discuss magnetic instabilities.

For $n_e=2$ the system is always insulating, even though n_f is not always an integer. The situation with almost compensated magnetic moments is displayed in Fig. 4. With increasing hybridization the almost-compensated-moment AKI state transforms into zero-moment PKI state, in the wide range of ε_f and U. It would be interesting to verify this prediction (e.g., by applying the pressure or alloying the KI systems). The PKI state cannot be achieved without the inclusion of the compensating cloud.

In summary, we have shown that an itinerant f-electron antiferromagnetism combined with a comparable negative conduction-electron-polarization effect are important in achieving both the *Kondo-lattice state* of heavy-quasiparticles with almost integer occupancy of the f level and a small and almost compensated magnetic moment at the same time. This state arises from a competition between a strong staggered molecular field coming from f-f correlations and f-c antiferromagnetic coupling, the latter reducing to the Kondo-type coupling only in the $V\rightarrow 0$ limit. For $n_e=2$ the antiferromagnetic Kondo insulating state transforms continuously into a paramagnetic Kondo insulator, whereas for $n_e \lesssim 2$ the antiferromagnetic metal changes into a weakly-ferromagnetic metal. The detailed discussion of the almost compensated state will appear separately.[11]

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