Kondo versus antiferromagnetic ground state of two Anderson impurities

A. Schiller and V. Zevin
The Racah Institute of Physics, The Hebrew University of Jerusalem, 91904 Jerusalem, Israel
(Received 16 December 1992)

A slave-boson formulation of the Anderson model for two degenerate impurities with infinite on-site Coulomb interaction U is presented. The Hamiltonian has many features in common with the slave-boson Hamiltonian for the single Anderson impurity but with finite U. It is shown within the noncrossing approximation that in the absence of any anisotropy an antiferromagneticlike ground state is impossible for $N_f > 2$. In order to make it possible one must either include anisotropy or add an explicit antiferromagnetic exchange term to the original Anderson model.

The old problem of two Anderson impurities embedded in a metal[1] has recently attracted vivid interest. It is believed that the understanding of this problem may cast light on the magnetic properties of some heavy-fermion compounds.[2] In spite of the good understanding of the single Anderson impurity the two-impurity system still poses many questions as to the nature of its ground state,[3–10] impurity-impurity interactions,[11–15] magnetic correlations,[16] and thermodynamic properties.[3,5,17] According to the standard scenario, which is based on the $S = \frac{1}{2}$ Kondo-impurity-spin Hamiltonian or on the Anderson Hamiltonian for the nondegenerate impurities,[3] the system locks either in the Kondo-like ground state (for a ferromagnetic or small enough antiferromagnetic spin interaction between the two impurities) or in the antiferromagnetic (AF) -like singlet (when the AF interaction is large enough compared with the Kondo energy scale T_K).

For many heavy-fermion compounds the system of two degenerate Anderson impurities is a much more realistic model. However, the large degeneracy together with the fact that the spin-spin [Ruderman-Kittel-Kasuya-Yosida (RKKY)] interaction appears in the Anderson model only in $1/N_f^2$ terms,[18] N_f being the impurity degeneracy, make it difficult to handle this model both analytically and numerically. The slave-boson formulation of the problem is one of the possibilities to use conventional theoretical methods within the $1/N_f$ expansion.[19] Recently by introducing two empty-state bosons, one for each impurity site, and two corresponding local constraints, the above-mentioned scenario was confirmed but only in the $N_f \longrightarrow \infty$ (mean field) approximation.[6] The RKKY interaction between the two impurities (which is absent in this limit) was imitated by introducing explicitly a kind of exchange interaction to the model.[20] It is worthy to note that neither crystal-electric-field (CEF) splittings nor anisotropy of the mixing interaction was introduced. Because of the two local constraints it would be difficult to treat the Hamiltonian of Ref. 6 beyond the mean-field approximation. Here we suggest another mapping of the original two-impurity Anderson Hamiltonian which involves only a single constraint. This permits us to formulate the noncrossing approximation[19] (NCA) for the problem. Following Müller-Hartmann's analytical treatment of the single impurity[21] we show in the absence of anisotropy (and of an explicit exchange interaction term) that the AF ground state is possible only for $N_f = 2$.

We use the common Anderson Hamiltonian for two degenerate impurities

$$\mathcal{H} = \sum_{\mathbf{k},m} \epsilon_k c^\dagger_{\mathbf{k},m} c_{\mathbf{k},m} + \sum_m \epsilon_{fm} \left(f^\dagger_{1m} f_{1m} + f^\dagger_{2m} f_{2m} \right) + \frac{U}{2} \sum_{m \neq m'} \left(n_{1m} n_{1m'} + n_{2m} n_{2m'} \right) + \sum_{\mathbf{k},m} V_m \left\{ e^{\frac{i}{2}\mathbf{k}\cdot\mathbf{R}} f^\dagger_{1m} c_{\mathbf{k},m} + e^{-\frac{i}{2}\mathbf{k}\cdot\mathbf{R}} f^\dagger_{2m} c_{\mathbf{k},m} + \text{H.c.} \right\} . \tag{1}$$

Here $c^\dagger_{\mathbf{k},m}$ creates a conduction electron while $f^\dagger_{1m}$ and $f^\dagger_{2m}$ create localized f electrons at sites $\mathbf{R}/2$ and $-\mathbf{R}/2$, respectively. U is the on-site Coulomb interaction which we will take hereafter to be infinite (i.e., no double occupation is allowed on either impurities) and $n_\alpha = f^\dagger_\alpha f_\alpha$, where $\alpha = 1m, 2m$. We introduce a mapping that consists of a single empty-state boson $b^\dagger$, a set of $2N_f$ single occupied-state pseudofermions $f^\dagger_{m,p}$ and a set of N_f^2 double occupied-state bosons $d^\dagger_{m,m',p}$. Here p denotes the parity (under interchange of the two impurities) of the pseudoparticle which could be either odd ($p = -$) or even ($p = +$). If we represent the state where the two impurities are unoccupied by $|0\rangle$ then the proposed mapping is described by $|0\rangle \longleftrightarrow b^\dagger$, $\frac{1}{\sqrt{2}} \left(f^\dagger_{1m} + p f^\dagger_{2m} \right) |0\rangle \longleftrightarrow f^\dagger_{m,p}$ and $\frac{1}{\sqrt{2}} \left(p f^\dagger_{1m} f^\dagger_{2m'} - f^\dagger_{1m'} f^\dagger_{2m} \right) |0\rangle \longleftrightarrow d^\dagger_{m,m',p}$. Note that $d^\dagger_{m,m',p} = -p d^\dagger_{m',m,p}$ describe the same state and that for $m = m'$ only $d^\dagger_{m,m,-}$ exists. The physical space corresponds to the constraint $Q = b^\dagger b + \sum_{m,p} f^\dagger_{m,p} f_{m,p} + \sum_{m,m',p\ (m \geq m')} d^\dagger_{m,m',p} d_{m,m',p} = 1$ while the appropriate Hamiltonian reads

$$\mathcal{H} = \int d\epsilon \sum_{m,p} \epsilon c^\dagger_{\epsilon,m,p} c_{\epsilon,m,p} + \sum_{m,p} \epsilon_{fm} f^\dagger_{m,p} f_{m,p} + \sum_{m,m',p\ (m\geq m')} (\epsilon_{fm} + \epsilon_{fm'})\, d^\dagger_{m,m',p} d_{m,m',p}$$
$$+ \int d\epsilon \sum_{m,p} V_{m,p}(\epsilon) \left\{ \left(f^\dagger_{m,p} b + p d^\dagger_{m,m,-} f_{m,-p} + \frac{1}{\sqrt{2}} \sum_{m',p'(m\neq m')} p d^\dagger_{m,m',p'} f_{m',pp'} \right) c_{\epsilon,m,p} \ + \text{ H.c.} \right\}. \tag{2}$$

Here the parity-channeled conduction-electron creation operators are defined as in Ref. 9 and $V^2_{m,p}(\epsilon) = \rho(\epsilon) N^2_p(\epsilon) V^2_m$ where $\rho(\epsilon)$ is the conduction electrons density of states and $N_p(\epsilon) = \left(1 + p\frac{\sin(kR)}{kR}\right)^{1/2}$ $[k = k(\epsilon)]$. Throughout this paper pp' indicates the product of the parities p and p'. In general the index m denotes the atomic states in the presence of the CEF. If an explicit exchange interaction term is added to the Hamiltonian of Eq. (1) the above mapping may be reformulated in terms of the total local spin states. In the particular case of $N_f = 2$ the only difference in Eq. (2) would reflect in the bare energies of the d bosons. The latter then will exhibit an explicit parity dependence. Similarly a direct hopping term between the two impurities would result in a parity dependence of the bare f energies. The specific case where $\epsilon_{fm} = \epsilon_f$ and $V_m = V$ would be referred to hereafter as the isotropic case. Note that the Hamiltonian of Eq. (2) is analogical to the slave-boson Hamiltonian for the finite-U Anderson impurity (see references in Ref. 22) which provides us with some insight as to its treatment.

We calculate the Green's functions $G_\alpha(z,T) = [z - \epsilon_\alpha - \Sigma_\alpha(z,T)]^{-1}$ of the pseudoparticles after a proper projection into the physical space. Here $\alpha = 0, 1mp, 2mm'p$ and $\epsilon_\alpha = 0, \epsilon_{fm}, \epsilon_{fm} + \epsilon_{fm'}$, respectively. A correct evaluation of the self-energies must account for the competing Kondo and magnetic behaviors that could arise. Each of the two limits is brought about differently. The Kondo ground state survives the $N_f \longrightarrow \infty$ limit and its first correction is of $O(1/N_f)$ whereas the RKKY interaction is only $O\left(1/N_f^2\right)$ and requires at least fourth order in the mixing interaction V. Thus while being committed to produce correctly the $N_f \longrightarrow \infty$ limit we must consider at the same time the $O\left(1/N_f^2\right)$ process that yields the RKKY interaction. Both goals are fulfilled by applying the same NCA scheme that was constructed for the finite-U single impurity. By keeping all diagrams relevant from $1/N_f$ considerations apart from some crossing diagrams that are $O\left(V^6\right)$, we manage to capture the RKKY interaction as well. Thus we arrive to the following set of self-consistent NCA integral equations:[22]

$$\Sigma_0(\omega + i\delta, T) = \sum_{m,p} \frac{\Gamma_m}{\pi} \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \nu(\epsilon) N^2_p(\epsilon) G_{1m,p}(\epsilon + \omega + i\delta, T) I_{m,p}(\epsilon, \omega + i\delta), \tag{3}$$

$$\Sigma_{1m,p}(\omega + i\delta, T) = \frac{\Gamma_m}{\pi} \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \nu(-\epsilon) N^2_p(-\epsilon) G_0(\epsilon + \omega + i\delta, T) I^2_{m,p}(-\epsilon, \omega + \epsilon + i\delta)$$
$$+ \sum_{m',p'} \frac{\Gamma_{m'}}{2\pi} (1 + \delta_{m,m'}) \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \nu(\epsilon) N^2_{pp'}(\epsilon) G_{2mm',p'}(\epsilon + \omega + i\delta, T), \tag{4}$$

$$\Sigma_{2mm',p}(\omega + i\delta, T) = \sum_{p'} \frac{\Gamma_m}{2\pi} \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \nu(-\epsilon) N^2_{pp'}(-\epsilon) G_{1m',p'}(\epsilon + \omega + i\delta, T) \ + \ (m \longleftrightarrow m')$$
$$+ \sum_{p'} \frac{\Gamma_m}{\pi} \frac{\Gamma_{m'}}{\pi} \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \nu(-\epsilon) N^2_{pp'}(-\epsilon) \int_{-\infty}^{\infty} d\epsilon' f(\epsilon') \nu(-\epsilon') N^2_{p'}(-\epsilon') G_0(\omega + \epsilon + \epsilon' + i\delta, T)$$
$$\times G_{1m,p'}(\epsilon + \omega + i\delta, T) I_{m,p'}(-\epsilon', \omega + \epsilon + \epsilon' + i\delta)$$
$$\times G_{1m',pp'}(\epsilon' + \omega + i\delta, T) I_{m',pp'}(-\epsilon, \omega + \epsilon + \epsilon' + i\delta). \tag{5}$$

Here Γ_m is the Anderson width $\Gamma_m = \pi\rho(0) V^2_m$, $f(\epsilon)$ is the Fermi-Dirac distribution function, and $\nu(\epsilon)$ is the reduced conduction electrons density of states $\nu(\epsilon) = \rho(\epsilon)/\rho(0)$. Another key component entering the NCA equations is the vertex correction $I_{m,p}(\epsilon, \omega + i\delta)$ which obeys the integral equation[22]

$$I_{m,p}(\zeta, z) = 1 + \sum_{m',p'} \frac{\Gamma_{m'}}{2\pi} (1 + \delta_{m,m'}) \int_{-\infty}^{\infty} d\epsilon f(\epsilon) \nu(\epsilon) N^2_{p'}(\epsilon) G_{1m',p'}(\epsilon + z, T)\ G_{2mm',pp'}(\epsilon + \zeta + z, T) I_{m',p'}(\epsilon, z). \tag{6}$$

A full self-consistent solution of Eqs. (3)–(6) may provide us with the answer to the question of the competition between the RKKY and Kondo behaviors and may reveal how each of the two is influenced by the other. It is easy to see for $N_f \longrightarrow \infty$ that in the isotropic case $G_{2mm',p}(\omega + i\delta, T = 0)$, $G_{1m,p}(\omega + i\delta, T = 0)$, and $G_0(\omega + i\delta, T = 0)$ from the above equations exhibit delta peak poles at $2\epsilon_f$, $2\epsilon_f - T_0$, and $2\epsilon_f - 2T_0$, respectively (T_0 being the $N_f \longrightarrow \infty$ single impurity Kondo energy), corresponding to a formation of zero, one, and two Kondo singlets. On the other hand, a straightforward fourth-order perturbation expansion of the first two terms in Eq. (5) reveals the RKKY splitting between the odd and even parities. Although the latter is just $O\left(1/N_f^2\right)$ it comes about due to the fact that for $m = m'$ only odd-parity double occupied states exist. A complete numerical study of Eqs. (3)–(6) is currently under investigation; nevertheless, it is possible to derive analytically some conclusions as for the competition between the magnetic and Kondo-like ground states.

The nature of the ground state of the two impurity problem is revealed from the zero-temperature limit of the G_α Green's functions. Since $\alpha = 0, 1mp, 2mm'p$ relate to a Kondo, partly Kondo (i.e., only one magnetic moment screened), and magnetic natured states, respectively, we need to determine which of the corresponding Green's functions (and thus also spectral functions) exhibit a more enhanced behavior near the threshold energy,[21] namely the ground-state energy measured relative to the unperturbed Fermi sea. This indicates the impurity configuration whose direct product with the Fermi sea possesses a greater overlap with the ground state and hence will reveal its basic character. Of course, one should expect the competition to be between G_0 and $G_{2mm',p}$. In the following we shall determine some necessary conditions for a magnetic behavior to dominate. We shall do so by neglecting altogether those terms involving the empty-state boson Green's function in Eqs. (4) and (5) and examining the competition between $G_{1m,p}$ and $G_{2mm',p}$ rather than G_0 and $G_{2mm',p}$. Whenever $G_{1m,p}$ overcomes $G_{2mm',p}$ it indicates that the system already prefers one of the magnetic moments to be quenched over any magnetic behavior so that the true nature of the ground state will surely be of the Kondo type. By omitting G_0 from the NCA equations we twist the results in favor of a magnetic behavior, so that such an analysis may very well fail when suggesting a magneticlike ground state. However, we can deduce from it regimes where the Kondo-like ground state is genuinely preferred. The basis of our treatment, therefore, is the following equations for T = 0:

$$\Sigma_{1m,p}(\omega + i\delta, T = 0) = \sum_{m',p'} \frac{\Gamma_{m'}}{2\pi} (1 + \delta_{m,m'}) \int_{-D}^{0} d\epsilon N_{pp'}^2(\epsilon) G_{2mm',p'}(\epsilon + \omega + i\delta, T = 0), \tag{7}$$

$$\Sigma_{2mm',p}(\omega + i\delta, T = 0) = \sum_{p'} \frac{\Gamma_m}{2\pi} \int_{-D}^{0} d\epsilon N_{pp'}^2(-\epsilon) G_{1m',p'}(\epsilon + \omega + i\delta, T = 0) \; + \; (m \longleftrightarrow m'), \tag{8}$$

where we have adopted a flat conduction electron density of states. Equations (7) and (8) are reminiscent of the single impurity NCA equations that were analyzed by Müller-Hartmann and Kuramoto and Kajima,[21] thus we follow their line of reasoning. The only complications as compared with Ref. 21 are the energy-dependent oscillatory functions $N_p^2(\epsilon)$ appearing in the integrands and the fact that both the single and double occupied-state Green's functions display a parity splitting even in the isotropic case. Yet, as we show, one could overcome these complications. The explicit form of Eqs. (7) and (8) demands the existence of a threshold energy ω_0 at which at least one $G_{1m,p}$ function and one $G_{2mm',p}$ function will simultaneously diverge. The threshold energy is just the ground-state energy of the system (measured relative to the unperturbed Fermi sea) above which all Green's functions start to acquire an imaginary part.[21] Due to the simplifications used the ground-state energy found here could quantitatively be incorrect, but it should not effect the threshold behaviors in case of a magneticlike ground state. Introducing the inverted Green's functions $g_\alpha(\omega) = -G_\alpha^{-1}(\omega + i\delta, T = 0)$, $\alpha = 1mp, 2mm'p$, we may differentiate them with respect to ω by using Eqs. (7) and (8). This yields

$$g'_{1m,p}(\omega) = -1 - \sum_{m',p'} \frac{\Gamma_{m'}}{2\pi} (1 + \delta_{m,m'}) \, N_{pp'}^2(\epsilon) g_{2mm',p'}^{-1}(\epsilon + \omega) \Big|_{\epsilon=-D}^{\epsilon=0}$$

$$+ \sum_{m',p'} \frac{\Gamma_{m'}}{2\pi} (1 + \delta_{m,m'}) \, pp' \int_{-D}^{0} d\epsilon \, \frac{d}{dx} \left(\frac{\sin(x)}{x} \right) \Big|_{x=kR} \frac{R}{\hbar v(\epsilon)} g_{2mm',p'}^{-1}(\epsilon + \omega), \tag{9}$$

together with similar equations for $g'_{2mm',p}(\omega)$. Here $\hbar v(\epsilon) = \frac{\partial \epsilon}{\partial k}$ and $k(\epsilon = 0) = k_F$ are the conduction electron velocity and the Fermi wave number, respectively. Amongst the various terms describing $g'_{1m,p}(\omega)$ and $g'_{2mm',p}(\omega)$, only those involving $g_\alpha^{-1}(\omega)$ (i.e., the ones coming from the $\epsilon = 0$ boundary) are important for the threshold behaviors since they possess singularities when $\omega \longrightarrow \omega_0$. All other terms are typically small (roughly $\sim N_f\Gamma/D$) and do not play an important role near ω_0. Nevertheless, they could be important for determining which of the Green's functions indeed diverges at the threshold energy. Thus for the threshold behavior we may replace the above equations with a simpler set where only the $g_\alpha^{-1}(\omega)$ terms are kept. The latter could be easily integrated to give

$$\sum_{m,p} \ln g_{1m,p} = \sum_{m,m',p(m\geq m')} \ln g_{2mm',p} + C. \qquad (10)$$

Here C is an integration constant and as already pointed out when $m = m'$ only $p = -$ exists. If we first restrict ourselves to the isotropic case where all m dependences fall then Eq. (10) becomes

$$2\ln g_{1,-} + 2\ln g_{1,+} = (N_f - 1)\ln g_{2,+} + (N_f + 1)\ln g_{2,-} + C, \qquad (11)$$

where we have specified only the parities and omitted the unrelevant indices. Though we did not determine explicitly which of the Green's functions indeed diverges at ω_0 the important features could be deduced directly from Eq. (11). If $G_{2,-}(\omega_0, T = 0)$ diverges while $G_{2,+}(\omega_0, T = 0)$ remains finite and negative (which corresponds for $N_f = 2$ to a ferromagnetic coupling) then close enough to the threshold either $G_{1,+} \sim G_{2,-}^{(N_f+1)/2}$ or $G_{1,-} \sim G_{2,-}^{(N_f+1)/2}$ depending on which of the two single occupied-state Green's functions is also divergent at ω_0. Thus we find that for any $N_f \geq 2$ the lower single occupied state exhibits a much more pronounced divergency than $G_{2,-}$, indicating that the system will always prefer a Kondo quenching. If on the other hand $G_{2,+}$ rather than $G_{2,-}$ diverges at ω_0 (corresponds for $N_f = 2$ to an antiferromagnetic coupling) then either $G_{1,+} \sim G_{2,+}^{(N_f-1)/2}$ or $G_{1,-} \sim G_{2,+}^{(N_f-1)/2}$ near the threshold. This case already allows two possibilities. If $N_f \geq 4$ once again the system favors the Kondo quenching; however, if $N_f = 2, 3$ (physically only $N_f = 2$ is relevant) then $G_{2,+}$ diverges stronger and the full set of NCA equations must be considered in order to determine the true nature of the ground state. We clearly see that only for $N_f = 2$ and only in case of an antiferromagnetic coupling can we expect a magnetic behavior at all. Note that we did not make any use of a particular reason why should the odd or even channels be lower in energy. For $N_f = 2$ this may just as well be due to an explicit exchange term added to the original Hamiltonian. Consequently, we in particular recovered the findings of previous scaling technique treatments of the nondegenerate two impurity problem.[3]

Going back to the general case, Eq. (10), we see that anisotropic effects could further split levels within each of the parity channels and therefore reduce the effective degeneracy. As long as no magnetic field is applied each Kramers doublet must share the same bare energy level and the same hybridization strength which implies that the effective degeneracy will always remain an even number. Following the isotropic case we may conclude that for N_f larger than 2 only anisotropic effects that will reduce the effective degeneracy to $N_f^{\text{eff}} = 2$ could yield a magnetic behavior which may result only from the appropriate even-parity channel. The latter serves as the equivalent of the antiferromagnetic channel for spin $\frac{1}{2}$. An alternative way to permit a magnetic ground state for $N_f > 2$ is to explicitly add an appropriate scalar exchange term to the Anderson model.

[1] S. Alexander and P. W. Anderson, Phys. Rev. **133**, A1594 (1964).

[2] See, e.g., N. Grewe and F. Steiglich, 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] C. Jayaprakash *et al.*, Phys. Rev. Lett. **47**, 737 (1981); J. Appl. Phys. **53**, 2143 (1982).

[4] B. A. Jones *et al.*, Phys. Rev. Lett. **61**, 125 (1988).

[5] R. M. Fye *et al.*, Phys. Rev. B **35**, 4901 (1987); R. M. Fye and J. E. Hirsh, *ibid.* **40**, 4780 (1989).

[6] B. A. Jones *et al.*, Phys. Rev. B **39**, 3415 (1989).

[7] O. Sakai *et al.*, Solid State Commun. **75**, 81 (1990).

[8] L. C. Andreani and H. Beck, Solid State Commun. **79**, 17 (1991).

[9] T. Saso, Phys. Rev. B **44**, 450 (1991).

[10] T. Yanagisawa, J. Phys. Soc. Jpn. **60**, 29; **60**, 3449 (1991).

[11] P. Fulde *et al.*, Solid State Phys. **41**, 1 (1988).

[12] N. Grewe and H. Keiter, Phys. Rev. B **24**, 4420 (1981).

[13] J. W. Rasul and A. C. Hewson, Solid State Commun. **52**, 217 (1984).

[14] D. M. Newns and N. Read, Adv. Phys. **36**, 799 (1987).

[15] D. L. Cox, Phys. Rev. B **35**, 4561 (1987).

[16] The zero temperature staggered susceptibility and the linear specific-heat coefficient are of a controversy that stimulated considerable work in the field, see Refs. 4–10 and also J. W. Rasul and P. Schlottmann, Phys. Rev. Lett. **62**, 1701 (1989).

[17] C. Zhou and T. K. Lee, J. Magn. Magn. Mater. **63 & 64**, 248 (1987).

[18] N. Read *et al.*, Phys. Rev. B **30**, 3841 (1984).

[19] N. E. Bickers, Rev. Mod. Phys. **59**, 845 (1987).

[20] The RKKY interaction is often introduced phenomenologically also into the nondegenerate Anderson model as a Heisenberg scalar interaction. It is not clear how innocent this treatment is in the frame of the variational approach or other numerical methods.

[21] E. Müller-Hartmann, Z. Phys. B **57**, 281 (1984); Y. Kuramoto and H. Kajima, *ibid.* **57**, 95 (1984).

[22] The integral equations, Eqs. (3)–(6), are based on the diagrammatic treatment of the degenerate Anderson impurity with a finite U and details may be seen in J. Holm and K. Schönhammer, Solid State Commun. **69**, 969 (1989) and in A. Schiller and V. Zevin, Phys. Rev. B **47**, 9297 (1993). For a more general set of NCA equations see Q. Qin and H. Keiter, Z. Phys. B **84**, 89 (1991).