Impurity correlations in dilute Kondo alloys

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The single-impurity Kondo model is often used to describe metals with dilute concentrations (n_i) of magnetic impurities. Here we examine how dilute the impurities must be for this to be valid by developing a virial expansion in impurity density. The $O(n_i^2)$ term is determined from results on the two-impurity Kondo problem by averaging over the Ruderman-Kittel-Kasuya-Yosida coupling. The nontrivial fixed point of the two-impurity problem could produce interesting singularities in the heat capacity of dilute alloys at $O(n_i^2)$.

I. INTRODUCTION

The physics of the single-impurity Kondo model, the problem describing a magnetic moment interacting with a sea of conduction electrons, is well understood.[1] At high temperatures the moment is free. The spin susceptibility of the impurity obeys a simple Curie-Weiss law, and there is a large additional term in the entropy, $S=k_B \ln 2$. As the temperature decreases, the impurity spin disappears as it forms a singlet with conduction electrons. The characteristic energy scale at which this happens is the Kondo temperature, which is exponentially small at small couplings, $T_K \sim E_F \exp[-1/(J\rho)]$, where J is the s-d exchange constant, ρ is the density of states. The $1/T$ divergence of the spin susceptibility is then effectively cut off by this scale at low temperatures. The singlet state formed by conduction electrons and impurity can be described at low temperatures $T \ll T_K$ as a Fermi liquid with enhanced density of states, $\delta\nu \sim n_i/T_K$, where n_i is the concentration of impurities.

The single-impurity Kondo model is often used as a way to obtain insights into the nature of the ground state for the Kondo lattice problem, a toy model for a number of rare-earth compounds, such as heavy fermions. Enhancement of the density of states due to the Kondo physics explains qualitatively the heavy masses in the heavy fermion materials. The local moments in a Kondo lattice interact through the electron-mediated Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which competes with the Kondo physics. The low-temperature behavior of the Kondo lattice is often thought to be determined by this competition. Conventional mean-field models of the heavy fermions,[2] however, often neglect the RKKY interaction, since it only appears as a correction to the mean-field behavior, and therefore is difficult to calculate.

The simplest model that captures the physics of this competition is the two-impurity Kondo model.[3–5] Depending on the relation between the RKKY interaction and the Kondo temperature and the presence of particle-hole symmetry breaking, the impurity spins can each get compensated by conduction electrons, form a singlet with each other, or a combination of both. A surface of Fermi-liquid fixed points occurs in the problem as well as an unstable nontrivial critical point associated with critical behavior of the thermodynamic functions.[6–11]

Despite extensive theoretical work on the two-impurity problem, we are not aware of any effort to apply the results in a systematic way to experiments. The appropriate way to do this, for dilute alloys, was pointed out in 1970 by Larkin and collaborators.[12] One should develop an expansion of thermodynamic quantities in powers of the impurity concentration n_i, a virial expansion. The pioneering work on this subject[12] only considered the high-T limit where the Kondo interaction could be ignored and only the RKKY interaction considered. At T of $O(T_K)$ or lower the Kondo interaction must be included. The $O(n_i^2)$ term in the virial expansion is completely determined by properties of the two-impurity problem appropriately averaged over the strength of the RKKY interaction. This calculation gives an estimate of the characteristic impurity concentration at which the single-impurity Kondo model breaks down. Analysis of the N-impurity models with $N>2$ could provide a further insight into the structure of this expansion.

We note that deviations from linear dependence of thermodynamic quantities on impurity concentration do not arise solely from the mechanism considered here. The Kondo temperature depends exponentially on the exchange integral, and changes in the composition of an alloy can alter it significantly. Such corrections, however, can be incorporated in the single-impurity picture, even though they alter the linear dependence of the thermodynamic and magnetic properties of a dilute alloy on concentration.

In the next section we discuss the subtle issue of length scales in the Kondo problem. Section III sets up the virial expansion, following Ref. 12. Section IV reviews some relevant results on the two-impurity Kondo problem. The $O(n_i^2)$ term is then considered for $T \gg T_K$ in Sec. V and at $T \leqslant T_K$ in Sec. VI. Quantitative results would certainly require accurate numerical solution of the two-impurity problem for all values or the RKKY coupling. Here we just apply the large-N approximation and make some qualitative remarks on the effect of the nontrivial critical point. Section VII contains conclusions.

II. LENGTH SCALES IN THE KONDO PROBLEM

There are (at least) three different ways of estimating a lower bound on the average separation of impurities neccessary for the single-impurity model to be valid. One could require that each impurity be far from the others compared to the size ξ_K of its screening cloud. Alternatively, one could require that the density of electron states within an energy T_K of the Fermi surface be at least as large as n_i (the Nozières exhaustion principle). This appears to be a neccessary (although not obviously sufficient) condition for the screening cloud wave functions from each impurity to form an approximately orthogonal set. Or, finally, one could require that the average RKKY interaction be small compared to T_K.

As a mechanism of Kondo screening, the picture of spin exhaustion cloud of exponentially large size $\xi_K \sim v_F/T_K$ (v_F is the Fermi velocity) has often been adapted.[1] From the renormalization group point of view, clearly, this length scale is present in the problem, since the low-energy Fermi-liquid theory[13] is only valid for $k-k_F \ll T_K/v_F$, or at distances $r \gg \xi_K$ away from the impurity. Direct calculations of the screening cloud profile[14] for the spin-spin correlators of the single-impurity Kondo model demonstrate that the screening cloud of conduction electrons indeed forms at the distance scale $\sim \xi_K$. Note that this is a more dynamical type of a screening than that which occurs for charge impurities in a Fermi liquid since it involves a linear combination of states where the impurity spin and the screening electron spin are in either an up-down or down-up configuration. In particular, the finiteness of the susceptibility at $T \to 0$ should not be attributed to a static conduction electron polarization cancelling the impurity spin polarization. Rather it results from the tendency of the impurity to form a singlet with the screening electron. This tendency is very well illustrated by a calculation of an equal-time correlator, which provides an instant snapshot of the system, and the zero-frequency spin correlator, or the spin susceptibility.[14] Indeed, it has been shown that the magnetic moment of conduction electrons exactly compensates that of the impurity at the same moment in time. Yet, there is no net polarization of the conduction electrons (as illustrated by a calculation of zero-frequency spin susceptibility), the fact also known as the Anderson-Clogston theorem.

Existence of an exponentially large length scale could have potentially strong effect on the theory of alloys with magnetic impurities. Indeed, typical $T_K \sim 10$ K and $E_F \sim 10$ eV makes $\xi_K \sim 10\,000a$, where a is the lattice spacing, much larger than typical distance between two impurities. This issue was addressed in one dimension for Luttinger liquids with magnetic impurities,[15] where it was found that the crossover happens at $n_i \sim 1/\xi_K$. At higher dimension, however, no such crossover was found experimentally. The physical reason for this is that the screening cloud wave function has an oscillatory character. Let us make a simple estimate of the overlap of the screening clouds from two separate impurities, with one impurity located at a distance R apart from the other. We shall make no particular assumptions about the screening cloud wave function except that it is made up of Fourier modes with $|k-k_F|$ less than or of order of $1/\xi_K$. Thus the overlap is

$$O(R) \equiv \int d^3r\, \psi^*(|\vec{r}+\vec{R}/2|)\,\psi(|\vec{r}-\vec{R}/2|) = \int \frac{d^3k}{(2\pi)^3} e^{i\vec{k}\cdot\vec{R}} |\psi(k)|^2 = \int_0^\infty dk k^2 |\psi(k)|^2 \frac{\sin(kR)}{2\pi^2 kR}. \tag{1}$$

So far we have only used the spherical symmetry of the screening cloud wave function and hence its Fourier transform. Now we assume

$$|\psi(k)|^2 \approx (\xi_K/k_F^2) f[(k-k_F)\xi_K], \tag{2}$$

where the scaling function $f(y)$ obeys the normalization condition

$$\int dy f(y) = 2\pi^2, \tag{3}$$

in order that $O(0)=1$. Thus we get

$$O(R) = (1/2\pi^2) \int dy f(y) \sin[k_F R + (R/\xi_K)y]/k_F R. \tag{4}$$

For $R \ll \xi_K$ this reduces to

$$O(R) = \sin(k_F R)/k_F R. \tag{5}$$

Thus the overlap is small for $k_F R \gg 1$. This calculation can be easily extended to the one-dimensional (1D) or 2D case. For 1D the overlap is essentially $\cos(2k_F R)$ for $R \ll \xi_K$. It does not get small until $R \gg \xi_K$. In 2D it is a Bessel function, $J_0(k_F R) \approx \sqrt{2/\pi k_F R}\cos(k_F R - \pi/4)$ (for $k_F R \gg 1$), again becoming small for $k_F R \gg 1$. We see that in dimensions higher than one the overlap of the two screening cloud wave functions is suppressed by $1/\sqrt{k_F R}$ (2D) or $1/k_F R$ (3D), small factors at large enough interimpurity separation. In 1D this overlap is $O(1)$. Thus the dimensionality of the problem could be the main reason why the large Kondo scale is absent in alloys.

The well-known Nozières' exhaustion principle states that for each impurity spin there should be a conduction electron in the vicinity $\sim T_K$ of the Fermi energy that screens it. This produces the following estimate for the interimpurity separation: $R \sim (\xi_K k_F^{-2})^{1/3}$ in 3D, $R \sim \sqrt{\xi_K k_F^{-1}}$ in 2D, and $R \sim \xi_K$ in 1D.

On the other hand, one can argue that the single-impurity model stops working when the RKKY interaction at the average interimpurity distance becomes comparable to the Kondo temperature, $J_{RKKY} \sim T_K$. This gives a somewhat higher estimate for the concentration at which one needs to account multi-impurity effects (or lower estimate for the average inter-impurity distance), $R \sim (\lambda^2 \xi_K k_F^{-2})^{1/3}$ in 3D, $R \sim \sqrt{\lambda^2 \xi_K k_F^{-1}}$ in 2D, and $R \sim \lambda^2 \xi_K$ in 1D, because of its dependence on the small coupling constant, $\lambda \equiv (J\rho)$.

III. THE VIRIAL EXPANSION

We start with the ordinary s-d Hamiltonian for a collection of magnetic impurities in an electron gas located at arbitrary positions in space:

$$H_{sd}=-\frac{J}{2N}\sum_{j,\mathbf{k},\mathbf{k}'} a^{\dagger}_{\mathbf{k}'\alpha}(\boldsymbol{\sigma}^{\alpha}_{\beta}\cdot\mathbf{S}^{j}_{imp})a^{\beta}_{\mathbf{k}}e^{i(\mathbf{k}-\mathbf{k}')\mathbf{r}_j}. \tag{6}$$

The free energy for N impurities is determined by the formula

$$F_{1,2,\ldots,N}=-T\ln\left\langle \exp\left\{-\int_0^{1/T}H_{sd}(\tau)d\tau\right\}\right\rangle. \tag{7}$$

To obtain a virial expansion, one can introduce the quantities f defined by recurrence relation

$$f_i=F_i,$$

$$F_{ij\cdots n}=\sum_k f_k+\sum_{kr} f_{kr}+\cdots\sum_{kr\cdots m} f_{kr\cdots m}+f_{ij\cdots n}, \tag{8}$$

where the summation is carried out over different sets of indices $ij\cdots n$. The function f vanishes if the distance between any two impurities tends to infinity. For example,

$$f_{ij}=F_{ij}-(F_i+F_j). \tag{9}$$

Averaging Eq. (8) over the distribution of the impurities and going to the thermodynamic limit $N\to\infty$, we obtain an expansion of the free energy in powers of the density. For magnetic impurities interacting with an RKKY interaction such expansion was carried out in Ref. 12. The n_i^2 term in the Kondo regime without the RKKY term was calculated in Ref. 16, although the influence of the RKKY interaction on this result was discussed in a later paper. The ordinary Kondo term is proportional to the density of impurities, $n_i\equiv N_i/V$. The n_i^2 contribution takes the following form:

$$F^{(2)}=N_i\frac{n_i}{2}\int d\mathbf{R}[F_{2i}(\mathbf{R})-2F_{Kondo}]. \tag{10}$$

Here $F_{2i}(\mathbf{R})$ is the free energy of the two impurities separated by a distance $\mathbf{R}$, F_{Kondo} is the usual single-impurity Kondo term. Thus the n_i^2 correction is competely determined by the two-impurity physics. Equation (10) and similar expressions for thermodynamic functions (derivatives of F) could be useful for numerical determination of the temperature dependence of the n_i^2 term in thermodynamic functions from the results on the two-impurity model.

IV. TWO-IMPURITY KONDO MODEL

Before calculating the n_i^2 term let us discuss the relevant physics of the two-impurity model. The study of the critical point for two impurities interacting with the Hamiltonian Eq. (6) starts with a reduction of a three-dimensional problem to a one-dimensional one.[6,9,10] This is done by introducing orthogonal 1D fermionic operators,

$$\psi_{1,2}(k)=\int d\Omega\,\frac{k}{\sqrt{2}}\left[\frac{1}{N_e(k)}\cos(\mathbf{k}\cdot\mathbf{R}/2)\right.$$
$$\left.\pm\frac{i}{N_o(k)}\sin(\mathbf{k}\cdot\mathbf{R}/2)\right]a(\mathbf{k}), \tag{11}$$

where we have integrated over spherical angle, and

$$N_{e,o}(k)=\sqrt{1\pm\frac{\sin(kR)}{kR}}. \tag{12}$$

Then the two-impurity model can be rewritten in 1D form. While the transformation itself is exact, the k-dependent couplings arise. Then these couplings are assumed to be k independent for excitations in the vicinity of the Fermi surface, an assumption questioned in Ref. 17. The main drawback of such assumption is that the RKKY exchange interaction then does not have a correct oscillating character. The reason is that the main contribution to RKKY interaction comes from energies $\sim v_F/R$, where v_F is the Fermi velocity, R is the distance between impurities. Apart from this, however, this procedure is fully justified. One can view the process of cutoff renormalization in two stages. During the first stage, the cutoff Λ is reduced below v_F/R, $\Lambda\ll v_F/R$, and oscillating RKKY interaction appears as a result:

$$H_{int}=\frac{1}{2}\lambda\bar{\psi}^{\dagger}_1(0)\boldsymbol{\sigma}\cdot\mathbf{S}_1\bar{\psi}_1(0)+\frac{1}{2}\lambda\bar{\psi}^{\dagger}_2(0)\boldsymbol{\sigma}\cdot\mathbf{S}_2\bar{\psi}_2(0)$$
$$+I\mathbf{S}_1\cdot\mathbf{S}_2. \tag{13}$$

Here $\lambda=J\rho$ is the Kondo coupling constant and $I=V_0cos(2k_FR)/R^3$ is the usual RKKY term, $V_0=\rho J^2/(16\pi)$. Note that both impurities are now at the same place in the 1D model. During the second stage the 1D Hamiltonian Eq. (13) is renormalized. The wave functions $\bar{\psi}_1$ and $\bar{\psi}_2$ are not orthogonal; they can be written in terms of orthogonal ψ_1 and ψ_2 as follows:

$$\bar{\psi}_1=\alpha\psi_1+\beta\psi_2, \tag{14}$$

$$\bar{\psi}_2=\beta\psi_1+\alpha\psi_2, \tag{15}$$

where

$$\alpha=\frac{N_e(k_F)+N_o(k_F)}{2}\simeq 1, \tag{16}$$

$$\beta=\frac{N_e(k_F)-N_o(k_F)}{2}\simeq\frac{\sin(k_FR)}{2k_FR}. \tag{17}$$

This orthogonal basis differs from the usual ''even'' and ''odd'' parity wave functions. It is seen explicitly in this basis that for $k_FR\gg 1$ channel mixing terms play minor role and can be neglected. Other marginal operators appear in the interaction Hamiltonian Eq. (13). Those, however, are suppressed by factors of k_FR, and therefore can be neglected.

V. HIGH-TEMPERATURE RESULTS

We can now analyze the n_i^2 corrections in a Kondo alloy of spin-1/2 impurities using the 1D Hamiltonian Eq. (13). At high temperatures ($T\gg T_K$) this can be done by including the RKKY term in Eq. (13) in the noninteracting Hamiltonian, and perturbing in the Kondo interaction. Instead of averaging over distances as in Eq. (10) one can do averaging over the strength of the RKKY coupling.[12] Indeed, the interaction $I(R)$ is a product of the smooth function R^{-3} times the rapidly oscillating function $\cos(2k_FR)$. It is convenient to carry out the integration in formula Eq. (10) by first averaging the

integrand over the period of oscillations. Changing the order of averaging and integration, we obtain

$$F^{(2)}=\frac{4Nn_iV_0}{3T}v.p.\int\frac{dy}{y^2}[F_{2i}(y,\lambda)-F_{2i}(0,\lambda)], \quad (18)$$

where $y=I(R)/T$, $v.p.$ stands for ''principle value.'' The behavior of the bulk spin susceptibility and the heat capacity of a Kondo alloy at temperatures $T\gg T_K$ is well known:

$$\chi(T)=\frac{(g\mu_B)^2n_i}{4T}[1-\lambda_{eff}(T)],$$

$$C(T)=\frac{3}{4}\pi^2n_i\lambda_{eff}(T)^4, \quad (19)$$

where $\lambda_{eff}(T)\simeq\lambda+\lambda^2\ln[\Lambda/T]$ is the effective coupling constant at energy scale T. To the lowest orders in the Kondo coupling we get the following result for the n_i^2 correction to the heat capacity:

$$\delta C=\frac{4}{3}\frac{n_i^2V_0}{T}\left(1-\lambda^2\ln\left[\frac{\Lambda}{T}\right]\right), \quad (20)$$

while for the magnetic susceptibility we find

$$\delta\chi=\kappa(g\mu_B)^2\frac{n_i^2V_0}{3T^2}\left(1-\lambda-2\lambda^2\ln\left[\frac{\Lambda}{T}\right]\right), \quad (21)$$

where

$$\kappa=v.p.\int_{-\infty}^{\infty}\frac{dy}{y^2}\frac{1-e^y}{3+e^y}\simeq-0.501\,447, \quad (22)$$

and $\Lambda\ll v_F/\bar{R}\simeq v_F(T_K/V_0)^{1/3}$ is the cutoff of the 1D model. Note that the small bandwidth effective theory of the two-impurity Kondo model is, of course, only valid for calculating low-energy properties. Thus the above results are valid in the range $T_K\ll T\ll\Lambda$. Since the RKKY interaction is relevant at weak coupling, it gives rise to power law singularities in thermodynamic functions at high temperatures.[12] The marginal Kondo interaction gives rise to weaker logarithmic terms in Eqs. (20) and (21). As we will see below in the next section, these singularities are cut off at temperatures of the order of T_K. The parameter of the virial expansion is $n_iV_0/T\ll1$. The major contribution to the n_i^2 term comes from distances $\bar{R}\sim(V_0/T)^{1/3}$, where the amplitude of the RKKY term is of the same order as temperature. The virial expansion fails at high enough densities when the average distance between impurities becomes comparable to $\bar{R}$. Note that the second virial coefficients for the spin susceptibility and the heat capacity Eqs. (20) and (21) cannot be written simply in terms of an effective coupling. Averaging over distances in the virial expansion correspond to different values of RKKY exchange I, which can be quite large. It is clear that I ruins the simple dependence of physical quantities on Λ and bare coupling. Consider what happens when we take I either very large or very small, continuing to treat I exactly and doing perturbation theory in the Kondo couplings. If I is very small (compared to T) we should be able to ignore it and then we just get two decoupled single impurity Kondo problems. In this limit we certainly recover the standard reduction of Λ dependence to renormalized coupling. On the other hand, if $T\ll|I|$ ($I<0$) then, at low energies, we have essentially a two-channel $S=1$ Kondo problem. Again we expect a standard reduction of the dependence on Λ corresponding to the beta function for that problem. However, the evolution of effective Kondo coupling with energy scale is different in these two limits (two decoupled one-channel $S=1/2$ Kondo problems versus one two-channel $S=1$ Kondo problem). Thus as we lower our cutoff past I the nature of the dependence on the cutoff changes. This is to be expected on general grounds. When we have a physical energy scale, like I in the problem we do not in general get simple dependence on the cutoff scale. In a massive quantum field theory, effective coupling constants are only useful at energy scales large compared to the mass.

VI. CORRECTIONS AT LOW TEMPERATURES

Let us now consider this problem at low temperatures. Averaging over the distances between two impurities involves integration over all RKKY couplings [see Eq. (18)], which makes analytical analysis extremely difficult. Instead let us consider first the $SU(N)$ generalization of the two-impurity model, for which explicit results for the linear term in the specific heat (γ) and the bulk magnetic susceptibility were obtained[7] as a function of RKKY coupling strength. For a $U=\infty$ $SU(N)$ generalized two-impurity Anderson model with RKKY interaction they find, in the large-N limit,

$$\gamma=\pi^2\chi=\frac{N\pi}{3}\frac{2e^{-B\delta}\cos\delta}{(1-A^2)T_K^*}, \quad (23)$$

where $A\simeq\sin(k_FR)/k_FR$, $B\simeq\cos(k_FR)/k_FR$, δ is related to the scattering phase shift in even and odd channels, $\delta_{e,o}=\pi/2\pm\delta$, N is the $SU(N)$ expansion parameter, T_K^* is the Kondo temperature. δ is found from the solution of appropriate mean-field equations. To find the n_i^2 correction to γ and χ, we have to average Eq. (23) over distances, or over RKKY interaction. Since for the average distance $k_F\bar{R}\gg1$, $B\to0$, $A\to0$. For these values of parameters the model has a first-order phase transition at $(I/T_K)_c=8/\pi$, with $\delta\simeq0$ when $(I/T_K)<8/\pi$ and $\delta\simeq\pi/2$ if $(I/T_K)>8/\pi$. We can apply the same averaging technique as in Eq. (18) [with $y=I(R)/T_K$]:

$$\chi^{(2)}=\frac{4Nn_iV_0}{3T_K}v.p.\int\frac{dy}{y^2}[\chi_{2i}(y,\lambda)-\chi_{2i}(0,\lambda)]. \quad (24)$$

Performing the integration, we easily find

$$\gamma=\pi^2\chi=N_i\frac{\pi N}{3T_K}\left(1-\frac{\pi n_iV_0}{3T_K}\right), \quad (25)$$

where the first term in Eq. (25) corresponds to the ordinary Kondo effect in the $SU(N)$ model. One important result of the virial expansion is to find the density at which the single-impurity approximation no longer works. As one can see from Eq. (25), this happens at $n_i\sim T_K/V_0$, or when the Kondo temperature becomes comparable to the amplitude of the RKKY interaction at the average interimpurity distance.

The mean-field $N=\infty$ solution suffers from a number of defficiencies. In particular, it was shown[6,11,10] that for two

spin-1/2 impurities the quantum phase transition is second, not first order. The critical point for two spin-1/2 impurities is only present if the model is particle-hole symmetric. Under these conditions it was found[11,10] that the specific-heat coefficient γ diverges at the critical point as $|I/T_K - (I/T_K)_c|^{-2}$, $(I/T_K)_c \simeq 2.2$, while the uniform spin susceptibility χ does not. Let us find out whether this divergence produces any singularity in the thermodynamic quantities. Unlike in the $N=\infty$ model, where there is always a first-order phase transition, one can identify possible singularities at $y \simeq 0$ and $y \simeq y_c$ from Eq. (24). At $y \equiv I/T_K \to 0$ the two Kondo impurities are independently screened. Since impurity spin at the Kondo fixed point should be replaced by the local spin density,

$$\mathbf{S}_1 \propto \frac{v_F \psi_L^\dagger(0)\boldsymbol{\sigma}\psi_L(0)}{T_K}, \tag{26}$$

so that the RKKY interaction can be written as follows:

$$H_{RKKY} \propto \frac{I v_F^2}{T_K^2}[\psi_1^\dagger(0)\boldsymbol{\sigma}\psi_1(0)\cdot\psi_2^\dagger(0)\boldsymbol{\sigma}\psi_2(0)]. \tag{27}$$

Perturbing in H_{RKKY}, one easily finds that the singularity in γ and χ at $y \to 0$ is logarithmic, i.e., principle-value integrable. For $y \simeq y_c$ the bulk susceptibility χ does not have a singularity, whereas γ has a power-law divergence $\sim (y - y_c)^{-2}$. The form of this singularity in the vicinity of y_c was investigated ealier using bosonisation by Gan,[10] who showed that it is effectively cut off at finite temperatures at $\delta y \propto \sqrt{T}$. This produces a square-root singularity in γ,

$$\delta\gamma \propto \frac{N_i}{T_K}\frac{n_i V_0}{\sqrt{T T_K}}. \tag{28}$$

Since χ does not diverge at the critical point, such singularity is absent for the bulk susceptibility. It is known[9] that in the absence of particle-hole symmetry there are relevant perturbations near the zero-temperature critical point of the two-impurity Kondo model. Such perturbations tend to wipe out the singularity at low temperatures, so it is not clear whether this effect could be observed in real materials.

VII. CONCLUSIONS

To summarize, we have considered cluster expansion in ''dense'' Kondo alloys. This expansion ties the two-impurity model, which has been investigated in detail, to experiment. At high temperatures, n_i^2 corrections produce additional $1/T$ and logarithmic singularities in thermodynamic quantities. At low temperatures, the n_i^2 term becomes large when the RKKY interaction at the average distance between impurities becomes of the same order as the Kondo temperature. Since the parameter of expansion in density is n_i/n_c, where $n_c = T_K/V_0 \propto \lambda^{-2}\exp(-1/\lambda)$ ($\lambda = \rho J$ is the dimensionless Kondo coupling constant) is exponentially small, deviation from the single-impurity behavior could be observed in dilute alloys with magnetic impurities. We have also seen that the presence of the intermediate fixed point in the two-impurity problem could lead to a mild low-temperature singularity in the specific heat, but not the bulk magnetic susceptibility. Further numerical investigation of this problem is desirable. In some rare-earth alloys, such as $Yb_xY_{1-x}CuAl$,[1] the behavior of thermodynamic functions is linear in x up to very large $x \simeq 0.9$. This observation could be related to the fact that these ions are often described by the Coqblin-Schrieffer model ($N=8$ in case of Yb^{3+}), where the RKKY interaction is suppressed [$O(1/N^2)$ (Ref. 18)].

Our calculation of the second virial coefficient shows that the density of impurities at which multi-impurity effects become important is *higher* than that given by Nozières' exhaustion principle. According to this principle, at these densities ($n \lesssim n_c$) there are not enough states to screen all impurities. Since Nozières' principle is multi-impurity in nature, it may be necessary to study the convergence properties of the expansion itself rather than the behavior of a given term to claim its failure.

Finally, we note that the reason why the large Kondo length scale is absent in the problem is purely geometric. The channel mixing terms between two impurities are large in the one-dimensional problem. However, in 3D they become suppressed by a small factor of $1/k_F R$, and the physics of the two-impurity model (and related n_i^2 term in the cluster expansion) is fully determined by the competition of the RKKY and Kondo effects.

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