Journal of Magnetism and Magnetic Materials 108 (1992) 147–149
North-Holland

Local approach to the magnetic susceptibility and specific heat of the Anderson lattice

L.G. Brunet, M.A. Gusmão and J.R. Iglesias

Instituto de Física, Universidade Federal do Rio Grande do Sul, Caixa Postal 15051, 91500 Porto Alegre, RS, Brazil

Green's functions relevant to the periodic Anderson Hamiltonian are evaluated via perturbation theory around the atomic limit, within an approximation that reproduces exact results in three different limits: zero band width, zero hybridization and zero Coulomb correlation. The magnetic susceptibility and the electronic specific heat are calculated for both Kondo and intermediate valence regimes. The results are in qualitative agreement with experiments and can be related to other theoretical calculations.

A large class of rare-earth and actinide compounds exhibit properties which vary from Intermediate Valence (IV) behavior (as SmS) to Kondo lattices and heavy-fermion systems (as $CeCu_2Si_2$) [1]. Although specific differences may be stated between IV and Kondo systems, common characteristics can be found, such as Curie–Weiss behaviour for the static susceptibility at high temperatures, Pauli-like susceptibility at low temperatures – indicating absence of magnetic order (not for all heavy-fermion systems), one or more peaks in the specific heat at low temperatures – indicating high electronic effective masses.

One of the most employed models to describe this kind of systems is the Periodic Anderson Hamiltonian (PAH), which extends the original one-impurity Anderson model [2] to the "one-impurity-per-site" description. Different theoretical treatments of the PAH have been explored: Green's function techniques, variational calculations, slave boson approach, etc. Comprehensive reviews of methods and results can be found in the literature (see, for example, ref. [3]).

A particularly simple and clear way to approach the PAH is to solve the isolated ion problem and treat the hopping between lattice sites as a perturbation [4,5]. Within this scheme, we present here a calculation of the static magnetic susceptibility and the specific heat of the Anderson lattice for both Kondo and IV systems. A comparison with experimental results and other theoretical calculations is also performed.

The model Hamiltonian can be written as,

$$H = H_0 + H', \tag{1}$$

with

$$H_0 = \sum_{i\sigma} E n^{\mathrm{f}}_{i\sigma} + \frac{U}{2}\sum_{i\sigma} n^{\mathrm{f}}_{i\sigma} n^{\mathrm{f}}_{i\bar{\sigma}} + \frac{1}{N}\sum_{i\sigma} V\left(c^{\dagger}_{i\sigma} f_{i\sigma} + \mathrm{h.c.}\right) \tag{2}$$

and

$$H' = \sum_{ij\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma}. \tag{3}$$

H_0 can be exactly diagonalized [5], and from the knowledge of its eigenvalues E_m, and eigenvectors, $|m\rangle$, the local Green's functions can be calculated using a spectral representation at finite temperatures (in the grand-canonical ensemble) [6],

$$g^{\alpha\delta}_{ij}(\omega_k) = \frac{1}{Z}\sum_{mn} \langle m | a^{\alpha}_i | n\rangle\langle n | a^{\delta\dagger}_j | m\rangle \times \frac{e^{-\beta E_m} + e^{-\beta E_n}}{E_m - E_n - \mathrm{i}\omega_k}\delta_{ij}, \tag{4}$$

where the superscripts α and β represent either f or c (conduction) electrons, and $\omega_k = (2k+1)\pi T$.

In the calculation of the complete finite temperature Green's function, the usual perturbation expansion [7] generates averages of products of fermion operators. The local character of the unperturbed Hamiltonian allows an exact decoupling of these averages for different sites. The approximation we adopt consists in decoupling the remaining local averages of four or more fermion operators in products of pair averages. This is equivalent to imposing Wick's theorem, which is not valid due to the presence of the Coulomb interaction term in H_0. Within this approximation we can write a Dyson-like equation for the complete Green's function in $\boldsymbol{k}$-space,

$$\mathcal{G}^{\alpha\delta}_{\sigma}(\boldsymbol{k}, \omega_n) = g^{\alpha\delta}_{\sigma}(\omega_n) + g^{\alpha c}_{\sigma}(\omega_n)\epsilon(\boldsymbol{k})\mathcal{G}^{c\delta}_{\sigma}(\boldsymbol{k}, \omega_n), \tag{5}$$

where

$$\epsilon(\boldsymbol{k}) = \sum_j t_{ij}\, \mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R}_{ij}}. \tag{6}$$

The imaginary part of the retarded Green's function obtained by the usual analytic continuation [7] of the above one yields a density of states in which Kondo peaks appear near the Fermi level [4,5,8].

We are interested in the zero-field static susceptibility,

$$\chi = -\frac{g\mu_B}{2}\frac{d}{dh}\langle n_\uparrow - n_\downarrow\rangle|_{h=0}. \tag{7}$$

Taking into account the changes in the energy eigenvalues of H_0 in the presence of a small external magnetic field, we perform numerically the above derivative, with the number of up- and down-spin electrons obtained through the Green's functions (5). Fig. 1 shows results for the Kondo case, in which a Curie–Weiss susceptibility is obtained for high temperatures and a Pauli-like one for low temperatures. The itinerant nature of the low temperature susceptibility can be confirmed by the vanishing of the local contribution (dashed line in fig. 1) due to a complete suppression of the local magnetic moment. This is in qualitative agreement with experimental observations for certain Ce compounds like $CeAl_3$, $CeCu_6$ and $CeCu_2Si_2$ [9,10]. The susceptibility in the IV limit decreases slowly with T for a large temperature range, and the low temperature behavior can be qualitatively compared with experimental results for Sm compounds such as SmS, SmSe and SmTe [11].

The specific heat of the system was calculated by deriving the average energy with respect to temperature, i.e., $C = d\langle H\rangle/dT$. The temperature derivative has been evaluated numerically, and the averages (see eqs. (1)–(3)) have been obtained from the related Green's functions. In order to evaluate the average of the Coulomb term we have introduced the function

$$\Lambda^{\alpha\beta\gamma}_{\sigma\sigma'}(\omega_n) \equiv \langle n^\alpha_\sigma a^\beta_{\sigma'}; a^{\gamma\dagger}\rangle, \tag{8}$$

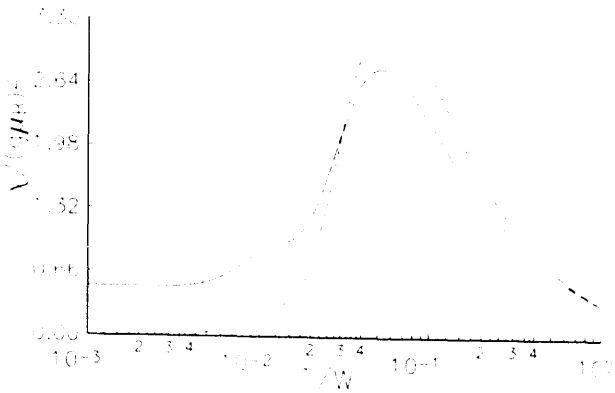

Fig. 1. Static susceptibility vs. temperature, with (full line) and without (dashed line) the hopping term, in the Kondo limit. The parameters are $E/W = -2$, $U/W = 6$ and $V/W = 0.3$, where W is half the bandwidth.

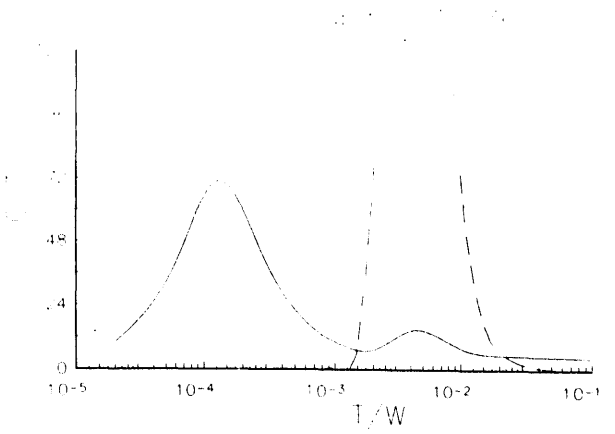

Fig. 2. Ratio of specific heat and temperature vs. temperature with (full line) and without (dashed line) the hopping term, in the Kondo limit. The parameters are $E/W = -2$, $U/W = 4$ and $V/W = 0.1$.

which is calculated exactly in zeroth order, with higher order corrections being decoupled in terms of Λ^0's and g's, in analogy to what has been done for the one-particle Green's function.

Fig. 2 shows the results for the specific heat divided by the temperature for the symmetric Kondo case ($2E + U = 0$) with hybridization $V = 0.1$. In the continuous plot (including hopping) two peaks are observed: the higher temperature one is also obtained in the local (no hopping) limit, and can be associated with the formation of the Kondo singlet; the lower temperature one is due to the high density of states in the vicinity of the Fermi level. The ratio between the amplitudes of these peaks depends on the hybridization and varies from ~ 2 to ~ 10 for V ranging from 0.2 to 0.05 (with $E = -2$). Such structure has recently been observed in $CeCu_4Al$ and $CeCu_4Ga$ [12], although the high temperature peak has been interpreted as a crystal field effect. A similar profile, with less pronounced peaks, is also visible in a $C/T \times T$ plot [10] for UBe_{13}. We should mention the variational treatment made by Brandow [13], where these two peaks are also found and attributed the same origin. In order to compare our results with experimental ones for $CeCu_4Al$ [12] we chose a value for the hybridization which gives the same ratio of temperature of the peaks in both theory and experiment. For an estimated ratio of ~ 42 the corresponding hybridization would be $V \approx 0.08$. With this value and choosing $E = -2$ we found a ratio of ~ 7 for the amplitudes of the corresponding peaks while the experimental ratio is ~ 10. Taking into account that this is a crude comparison the agreement is reasonable.

In conclusion we want to stress that, despite the simple approximation scheme utilized, we have found results in relatively good agreement with experiment. Furthermore, it is worth noticing that many of the

characteristic features of these results are essentially owing to the prominent role played by local interactions in our approach.

We acknowledge fruitful discussions with B. Coqblin and C. Lacroix. This work was partially supported by Brazilian agencies Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Financiadora de Estudos e Projetos (FINEP) and Fundação de Amparo à Pesquisa do Estado do Rio Grande do Sul (FAPERGS).

References

[1] See, for example, B. Coqblin, Magnetism of Metals and Alloys, ed. M. Cyrot (North-Holland, Amsterdam, 1982) p. 295. M.B. Brandt and V.V. Moshchalkov, Adv. Phys. 33 (1984) 373.
[2] P.W. Anderson, Phys. Rev. 124 (1961) 41.
[3] P. Fulde, J. Phys. F 18 (1988) 601.
[4] A.S.R. Simões, J.R. Iglesias, A. Rojo and B.R. Alascio, J. Phys. C 21 (1988) 1941.
[5] L.G. Brunet, R.M. Ribeiro-Teixeira and J.R. Iglesias, Solid State Commun. 68 (1988) 477; J. de Phys. 49 (1988) C8-697.
[6] D.N. Zubarev, Sov. Phys. Usp. 3 (1960) 320.
[7] A.A. Abrikosov, L.P. Gorkov and I.E. Dzyaloshinski, Methods of Quantum Field Theory in Statistical Physics (Prentice-Hall, Englewood Cliffs, 1963).
[8] J.R. Iglesias, A.S.R. Simões, R.M. Ribeiro-Teixeira and L.G. Brunet, Physica B 163 (1990) 692.
[9] K. Andres, J.E. Graebner and H.R. Ott, Phys. Rev. Lett. 35 (1975) 1979.
[10] G.R. Stewart, Rev. Mod. Phys. 56 (1984) 755.
[11] M.B. Maple and D. Wohlleben, Phys. Rev. Lett. 27 (1971) 511.
[12] E. Bauer, N. Pillmayr, H. Müller, J. Kohlmann and K. Winzer, J. Magn. Magn. Mater. 90&91 (1990) 411.
[13] B.H. Brandow, Phys. Rev. B 37 (1988) 250.