Single Impurity in a Paramagnetic Induced-Moment System*

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Using a cluster method it is shown that the local instability observed in the molecular-field calculation and in the Green's-function calculation in the random-phase approximation for a paramagnetic induced-moment system containing a single impurity is spurious.

Recently, the problem of a single impurity in a paramagnetic induced-moment system has been discussed by Fulde and Peschel[1] using a diagram technique in the molecular-field approximation and by Shiles and Wang[2] using a Green's-function method in the random-phase approximation (RPA). One of the results found was that the energy of a localized mode becomes zero and thus the susceptibility diverges as the exchange interaction between the impurity ion and the host ions exceeds a certain value. This normally indicates an instability of the assumed ground state and a state with local polarization was proposed. To investigate the nature of this instability we have recalculated the susceptibility using a cluster method. We solve the impurity cluster which consists of the impurity and its nearest-neighbor host ions exactly and couple the cluster to the rest of the system in an effective-field approximation. Since the interactions between the impurity ion and its surrounding host ions are treated exactly in the calculation, the method is superior to the previous ones in dealing with systems with strong impurity-host coupling. We show that the instability observed in the previous works was a spurious result due to the use of molecular-field approximation or RPA in the Green's-function calculation. This same question was also raised by Fulde and Peschel[3] and discussed in a very complicated manner by Villain and Pataud.[4] Here we sketch the cluster theory as follows.

We assume the same simple induced-moment system as described by Refs. 1 and 2. Each ion is represented by a singlet ground state and a singlet excited state. To describe the system we introduce a pseudospin $S=\frac{1}{2}$ for each ion.[2] The Hamiltonian in the pseudospin variables is then

$$\mathcal{H}=\Delta\sum_n S_n^z+(\Delta-\Delta_0)S_0^z-4\mathcal{J}\alpha^2\sum_{n,\delta}S_n^x S_{n+\delta}^x - 8(\mathcal{J}_0\alpha_0-\mathcal{J}\alpha)\sum_\delta S_0^x S_\delta^x, \tag{1}$$

where Δ and Δ_0 are energy gaps of the two singlets and α and α_0 are the matrix elements of the angular momentum operators between the two singlets for the host ions and the impurity ion. We have also assumed that the exchange interactions are between nearest-neighbor ions only and $\mathcal{J}$ and $\mathcal{J}_0$ are, respectively, the host-host and host-impurity exchange-interaction parameters.

The eigenstates of an isolated impurity cluster can be found easily and the local susceptibilities χ_0 and χ_δ are then calculated by the standard perturbation technique using

$$\mathcal{H}'=-2rhS_0^x-2h\sum_\delta\left(1+4\mathcal{J}\alpha^2\sum_{(\delta'+\delta\neq 0)}{}'\chi_{\delta+\delta'}\right)S_\delta^x \qquad (h=g\mu_B\alpha H,\quad r=g_0\alpha_0/g\alpha,\quad \chi_n=\langle S_n^x\rangle/h) \tag{2}$$

as the perturbation potential, where the exchange-enhancement effect due to the neighboring ions of the cluster has been incorporated using an effective-field approximation. It is then clear that both χ_0 and χ_δ depend on the quantity $\chi_2\equiv\sum'_{\delta'}\chi_{\delta+\delta'}$ which should be determined self-consistently.

To find the self-consistent condition, we note that in the same approximation we can obtain the following equation which relates the local susceptibilities:

$$\sum_n \mathcal{L}_{mn}\chi_n=\sum_\kappa D_{m\kappa}\chi_\kappa+\gamma_m, \tag{3}$$

where

$$\mathcal{L}_{mn}=\delta_{mn}-\frac{a}{z}\sum_\delta\delta_{m,n+\delta}, \tag{4}$$

$$D_{m\kappa}=-\frac{a}{z}\sum_{\delta_1,\delta_2}\delta_{m,\delta_1}\delta_{\kappa,\delta_1+\delta_2}-\frac{a}{z}\sum_{\delta_1}\delta_{m,0}\delta_{\kappa,\delta_1}, \tag{5}$$

$$\gamma_m=\chi_c+\sum_\delta\delta_{m,\delta}(\chi_\delta-\chi_c)+\delta_{m,0}(\chi_0-\chi_c). \tag{6}$$

We have also defined $a=(4\mathcal{J}z\alpha^2/\Delta)\tanh(\Delta/2kT)$ and $\chi_c=(1/\Delta)\tanh(\Delta/2kT)$ and z is the number of the nearest neighbors of an ion. Equation (3) enables χ_n to be expressed in terms of χ_0 and χ_δ. Setting $n=0$ we find that

$$a\chi_2=\chi_c\left(\frac{a}{1-a}\,\frac{1}{G-1}-z-\frac{aG}{G-1}\right)+\chi_\delta\left(z-\frac{a^2G}{G-1}\right), \tag{7}$$

where

$$G=\frac{1}{N}\sum_\kappa\frac{1}{1-\gamma_\kappa}\quad\text{and}\quad z\gamma_\kappa=\sum_\delta e^{i\vec{\kappa}\cdot\vec{\delta}}. \tag{8}$$

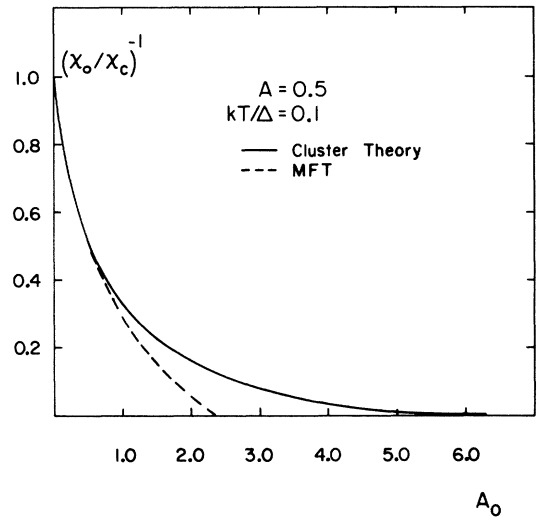

FIG. 1. Inverse local susceptibility at the impurity site χ_0^{-1} in units of χ_c^{-1} (the inverse susceptibility of the pure crystal in the absence of exchange interactions) as a function of $A_0 \equiv 4\mathcal{J}_0\alpha_0\alpha/\Delta$ for $A \equiv 4\mathcal{J}\alpha^2/\Delta = 0.5$, $\Delta_0 = \Delta$, and $g_0\alpha_0 = g\alpha$ at $k_BT = 0.1\Delta$.

This equation which relates χ_2 back to χ_0 is the self-consistent condition for our calculation. In Fig. 1, we compare the result of the present cluster theory (CT) and that of the molecular-field theory (MFT) (Green's-function theory in RPA gives a similar result), for $A \equiv 4\mathcal{J}\alpha^2/\Delta = 0.5$ and $\Delta_0 = \Delta$ at $k_BT = 0.1\Delta$ for a simple-cubic-lattice system. We plot $(\chi_0/\chi_c)^{-1}$ as function of $A_0 \equiv 4\mathcal{J}_0\alpha_0\alpha/\Delta$. We see that they agree very well until the coupling of the impurity and the host ions is so strong that $A_0 \sim 1$. χ_0 of MFT diverges at about $A_0 = 2.3$, while χ_0 of CT remains finite until $A_0 = 6.3$. In fact if A is reduced to 0.3, χ_0 of MFT diverges at $A_0 = 2.4$, but χ_0 of CT will never diverge although it does at a lower temperature. The cause of divergence of χ_0 of CT at the much higher value of A_0 can also be traced to the use of the effective-field approximation in the coupling of the impurity cluster to the rest of the system. We therefore conclude that the instability is spurious and that the polarization would only be possible if the concentration of the impurity is finite and then the critical condition is expected to be concentration dependent.

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