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Spin Fluctuation Effects with Strongly Magnetic Impurities

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Abstract. A phenomenological theory of the effect of spin fluctuations on the magnetic susceptibility of strongly magnetic impurities in metals is developed. The finite zero temperature susceptibility is calculated and qualitative aspects of the temperature dependence are pointed out. The results are expressed in terms of the density of states due to the localized state virtual levels.

I. Introduction

In this paper we investigate some aspects of the influence of the metallic environment on magnetic atoms from the transition or rare earth series when placed in solid solution in a dilute alloy. In simplest terms the contact of the impurity atom with the conduction electron sea has two effects:

- (I) The mixing of the atomic wave functions with the degenerate conduction electron states broadens the atomic impurity state into a virtual state and allows the possibility that the 'pseudo-atom' will change its configuration by adding or subtracting a fraction of an electron. This configuration change is inhibited by the electron-nuclear Coulomb attraction and by the electron-electron Coulomb repulsion, U , both of which favor the atomic configuration. If the level width, Δ , is sufficiently large compared with U , $U/\Delta \lesssim 1$, the atomic magnetism will disappear [1].
- (II) Since the conduction electrons are constantly exchanging with those of the localized magnetic state, spin correlations are induced in the conduction sea in the vicinity of the impurity. This is the Kondo effect [2] and has associated with it the well-known anomalies in the electrical resistivity, thermoelectric power, magnetoresistance, etc.

In this paper we concern ourselves with (I) above in the limit $U/\Delta \gg 1$, and we investigate by means of a simple model the effect on the magnetic susceptibility of the contact of the virtual level density of states at the Fermi surface³⁾. The results imply a

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³⁾ We shall not consider the Kondo effect in this paper as our interest is explicitly in the spin fluctuation effects. The treatment is valid if $\hbar T_k = D \exp(-1/J \rho)$ is very small compared to the temperatures of interest.

finite zero temperature susceptibility in all cases with the expected Curie law becoming dominant only at higher temperatures.

II. Definition of the Problem

We assume the impurity – conduction electron system \mathcal{H} can be described by the one-orbital Anderson Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{mix}$$

where

$$\mathcal{H}_0 = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}} n_{\mathbf{k}\sigma} + E (n_{d\uparrow} + n_{d\downarrow}) + U n_{d\uparrow} n_{d\downarrow} \quad (1a)$$

and

$$\mathcal{H}_{mix} = \bar{V} \sum_{\mathbf{k}, \sigma} (c_{\mathbf{k}\sigma}^+ c_{d\sigma} + c_{d\sigma}^+ c_{\mathbf{k}\sigma}) \quad (1b)$$

and all quantities are as defined by Anderson [1]. In the limit of interest here, \bar{V} small, Schrieffer and Wolff [3] have shown that the above Hamiltonian can be transformed into the well-known s - d Hamiltonian of the form

$$\mathcal{H} = \bar{\mathcal{H}}_0 + \bar{\mathcal{H}}_{s,d} \quad (2)$$

where

$$\bar{\mathcal{H}}_0 = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}} \bar{n}_{\mathbf{k}\sigma} + E (\bar{n}_{d\uparrow} + \bar{n}_{d\downarrow}) + U \bar{n}_{d\uparrow} \bar{n}_{d\downarrow}, \quad (3)$$

$$\begin{aligned} \bar{\mathcal{H}}_{s,d} = & \frac{1}{2N} \sum_{\mathbf{k}, \mathbf{q}} J_{\mathbf{k}\mathbf{q}} \{ (\bar{c}_{d\uparrow}^+ \bar{c}_{d\uparrow} - \bar{c}_{d\downarrow}^+ \bar{c}_{d\downarrow}) (\bar{c}_{\mathbf{k}\uparrow}^+ \bar{c}_{\mathbf{q}\uparrow} - \bar{c}_{\mathbf{k}\downarrow}^+ \bar{c}_{\mathbf{q}\downarrow}) \\ & + 2 \bar{c}_{d\uparrow}^+ \bar{c}_{d\downarrow} \bar{c}_{\mathbf{k}\downarrow}^+ \bar{c}_{\mathbf{q}\uparrow} + 2 \bar{c}_{d\downarrow}^+ \bar{c}_{d\uparrow} \bar{c}_{\mathbf{k}\uparrow}^+ \bar{c}_{\mathbf{q}\downarrow} \}. \end{aligned} \quad (4)$$

In equations (2), (3) and (4)

$$\bar{c}_{d\sigma}^+ = e^{-s} c_{d\sigma}^+ e^s \quad (5)$$

$$\bar{c}_{\mathbf{k}\sigma}^+ = e^{-s} c_{\mathbf{k}\sigma}^+ e^s \quad (6)$$

and the generating function S is given [3] by Schrieffer and Wolff

$$S = \sum_{\mathbf{k}\sigma\alpha} \frac{\bar{V}}{\varepsilon_{\mathbf{k}} - \varepsilon_{\alpha}} \bar{n}_{d-\sigma}^{\alpha} \bar{c}_{\mathbf{k}\sigma}^+ \bar{c}_{d\sigma} - \text{H.c.} \quad (7)$$

with

$$\varepsilon_{\alpha} = \begin{cases} E & \alpha = + \\ E + U & \alpha = - \end{cases} \quad \bar{n}_{d-\sigma}^{\alpha} = \begin{cases} \bar{n}_{d-\sigma} & \alpha = + \\ 1 - \bar{n}_{d-\sigma} & \alpha = - \end{cases}. \quad (8)$$

The conditions for the validity of the Schrieffer-Wolff transformation are (we use the convention $E_F = 0$)

$$\frac{|E|}{\Delta} \gg 1 \quad \text{and} \quad \frac{E + U}{\Delta} \gg 1. \quad (9)$$

In the limit where equations (2), (3) and (4) provide an adequate description of the problem, the impurity susceptibility is immediately seen to be a Curie law, with higher order effects added on due to the Kondo effect as indicated in II above [4]. If one neglects $H_{s,d}$, the examination of equation (3) shows the $\bar{c}_{\mathbf{k}\sigma}$ states to represent a complete set of non-interacting Fermions (the anticommutation relations are preserved in the transformation) with a dispersion relation identical to that of the plane wave states of the pure host. These states thus form a free-particle Fermi sea and contribute to the magnetic susceptibility (and specific heat) by an amount numerically equal to the pure host material. Since the magnetization operator has the same form in the transformed as well as the untransformed representation, the change in the susceptibility upon addition of a single impurity is given by

$$\Delta\chi = \chi_{\text{alloy}} - \chi_{\text{host}} = \frac{g\mu_B}{2} [\langle\langle \bar{n}_{d\uparrow} \rangle\rangle - \langle\langle \bar{n}_{d\downarrow} \rangle\rangle] \quad (10)$$

where the double brackets denote a thermal average.

The transformed states thus represent the best starting point for a discussion of the impurity susceptibility in the limit where the mixing interaction is small, for they are well-defined states with an essentially infinite lifetime so long as the condition (9) is well satisfied. Moreover, there is experimental evidence that the transformed operators are the physically relevant description. Spin resonance experiments have demonstrated clearly that because of the long tails of the admixed wave-function the impurity can probe the spin-orbit interaction at distant impurities [5]. Thus in the ESR experiments, the νf field couples with the transformed state. We emphasize this point at length only because in the literature, the difference between the transformed and untransformed operators is often not explicitly taken into account in the use of the s - d model. Furthermore, the question of the physically relevant operators is not *a priori* clear.

Because of the Kondo effect, we still do not have a complete solution of the problem defined by the s - d Hamiltonian given in equation (2). However, in a qualitative sense the problem is understood. For example, as argued above, when the conditions expressed by equation (9) hold, the Schrieffer-Wolff transformation is valid and any deviation from Curie law behavior is to be attributed to the Kondo effect. This observation has in the past led experimentalists to claim a given system is an example of the Kondo effect whenever departures from a Curie law (e.g. Curie-Weiss behavior) are observed. However, the condition for the validity of the s - d model, i.e. equation (9), raises an additional and separate question. In any real physical system the ratios $|E|/\Delta$ and $(E + U)/\Delta$ are finite. The effect of finite values (perhaps large) for these ratios on measurable parameters, e.g. the susceptibility, is not clearly understood.

The question of the effect of a breakdown in the conditions given in equation (9) is related to the spin fluctuation approach to the impurity problem. When $|E|/\Delta$ and/or $(E + U)/\Delta$ are finite, the \bar{d} states must have a finite lifetime; i.e. the Schrieffer-Wolff transformation is no longer rigorously valid. Note that the lifetime in question is not the one-electron lifetime, Δ , but the lifetime of the \bar{d} -state arising from the finite values of the quantities in equation (9). At present, no satisfactory microscopic theory exists which expresses this lifetime in terms of the fundamental parameters of

the model \bar{V} , E and U . Therefore we treat the \bar{d} -state lifetime as a phenomenological parameter. This finite lifetime can have important consequences for it allows the impurity spin to 'leak' off. The physical point is that for a finite mixing interaction there is a finite probability at any given time of simultaneous occupancy of both the up-spin and down-spin orbitals (or alternatively a finite probability of neither being occupied) with a zero net magnetic moment. This kind of *spin fluctuation* effect leads to a finite zero temperature susceptibility. In the magnetic limit, $E \ll 0$ and $E + U > 0$ (thus making the zero occupancy probability negligible) the probability of such a spin fluctuation is

$$P_0(E + U) \simeq \int_{-\infty}^0 \varrho_{\bar{d}, E+U}(\omega) d\omega \quad (11)$$

where $\varrho_{\bar{d}, E+U}$ is the density of states arising from the broadened virtual \bar{d} -state centered at $(E + U)$. The competitive thermal process, i.e. the probability of a thermal fluctuation, has a magnitude of approximately the Fermi function evaluated at $E + U$ the energy of the peak in $\varrho_{\bar{d}, E+U}(\omega)$

$$P_{\text{Th}} \simeq e^{-\beta(E+U)}. \quad (12)$$

It is only when

$$P_{\text{Th}} > P_0(E + U) \quad (13)$$

that the temperature plays an explicit role so that a Curie-like temperature dependence can be operative. The qualitative argument leading to the relation (13) will be verified by explicit calculation below.

III. Phenomenological Theory of the Susceptibility

Since we are interested in the limit of small level width we begin by considering the case $\bar{V} = 0$. The equation of motion for the localized state Green's function is written

$$\frac{d}{dt} G_{\sigma}(t) = -i \delta(t) + \vartheta(t) \ll\{[\bar{\mathcal{H}}_0, \bar{c}_{d\sigma}(t)], \bar{c}_{d\sigma}^+(0)\} \gg \quad (14)$$

where

$$G_{\sigma}(t) = -i \ll\{\bar{c}_{d\sigma}(t), \bar{c}_{d\sigma}^+(0)\} \gg \vartheta(t) \quad (15)$$

and the double brackets denote a thermal average. Equation (14) can be solved exactly [6-8] leading to the result

$$\int dt e^{i\omega t} G_{\sigma}(t) = G_{\sigma}(\omega) = \frac{1 - \ll\bar{n}_{d-\sigma}\gg}{\omega - E + i\delta} + \frac{\ll\bar{n}_{d-\sigma}\gg}{\omega - E - U + i\delta}. \quad (16)$$

The spectral weight function $A_{\sigma}(\omega)$ is given by

$$A_{\sigma}(\omega) = -\frac{1}{\pi} \text{Im} G_{\sigma}(\omega) = (1 - \ll\bar{n}_{d-\sigma}\gg) \delta(\omega - E) + \ll\bar{n}_{d-\sigma}\gg \delta(\omega - E - U) \quad (17)$$

and consists of a double-peaked structure for each sign of spin; one peak at $\omega = E$ with amplitude $1 - \langle\langle \bar{n}_{d-\sigma} \rangle\rangle$ and the other at $(E + U)$ with amplitude $\langle\langle \bar{n}_{d-\sigma} \rangle\rangle$, as shown schematically in Figure 1. The result is physically reasonable; the two peaks correspond to the two possible configurations of one or two electrons in the localized state. This structure is well-known and was pointed out by Kj ollerstr om, Scalapino and Schrieffer [6] as well as Hewson [7] and Dworin [8].

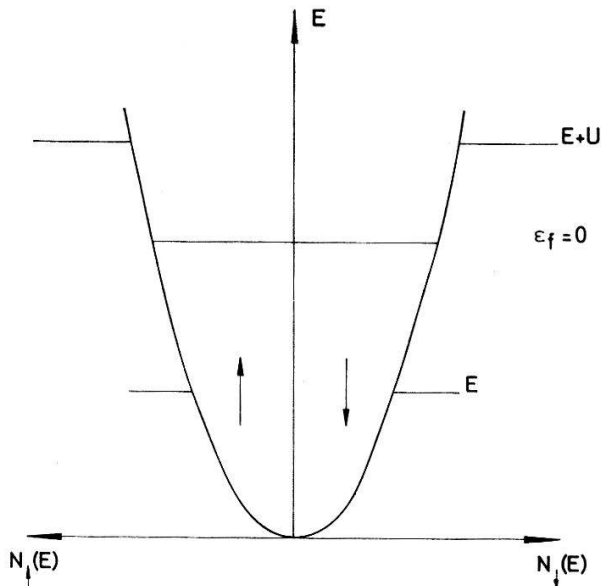


Figure 1
Schematic diagram of the localized state spectral weight function. For the case drawn here $E < 0$, $E + U > 0$ the peaks each have an amplitude of $1/2$.

One cannot use (16) in a diagrammatic expansion in terms of \bar{V} because the poles of G do not correspond to one-particle energies. Moreover, since the \bar{d} operators are complicated functions of the mixing interaction involving a complex superposition of states, a calculation of the \bar{d} spectral weight function appropriate to an impurity in a real metal is a difficult problem indeed. As a result, we adopt a phenomenological point of view and generalize equation (17) as

$$A_{\sigma}(\omega) = (1 - \langle\langle \bar{n}_{d-\sigma} \rangle\rangle) \varrho_{\bar{d},E}(\omega) + \langle\langle \bar{n}_{d-\sigma} \rangle\rangle \varrho_{\bar{d},E+U}(\omega) \quad (18)$$

where $\varrho_{\bar{d},E}(\omega)$ and $\varrho_{\bar{d},E+U}(\omega)$ are the normalized density of states functions centered at E and $(E + U)$ respectively. Equation (18) can perhaps be viewed somewhat more generally. In the limit of interest here, $U/\Delta \gg 1$, one does not expect the basic atomic level structure of the impurity to be altered. Thus the form of equation (18) should be generally valid since as mentioned above, the double peaked structure has its origin in the different possible atomic configurations. One can then view $\varrho_{\bar{d},E}(\omega)$ and $\varrho_{\bar{d},E+U}(\omega)$ either as the experimentally determined \bar{d} virtual state densities in the alloy or as the fundamental quantities to be calculated in a microscopic theory. Since information on virtual states is becoming available through optical absorption and photoemission studies [9], the experimental point of view may be of real practical value (of course the relation of such measurements to the \bar{d} state density must be carefully considered). We emphasize, however, that when $U/\Delta < 1$, the atomic physics of the impurity is strongly perturbed by the metallic environment so that a spectral weight function of the form of equation (18) loses its validity.

We explicitly neglect in equation (18) the effects arising from $H_{s,d}$ which are the source of the Kondo divergence. This is done not because they are unimportant, but because we are interested in the explicit spin fluctuation effects which arise indirectly from the finite level width. In fact, since the characteristic Kondo temperature varies exponentially with J and hence with \bar{V}^2 , the Kondo phenomena are restricted to very low temperature in the limit of small \bar{V} as is appropriate for example for most rare earth impurities in simple metals.

The average number of localized electrons is obtained with the help of equation (18)

$$\langle\langle \bar{n}_{d\sigma} \rangle\rangle = (1 - \langle\langle \bar{n}_{d-\sigma} \rangle\rangle) F(E) + \langle\langle \bar{n}_{d-\sigma} \rangle\rangle F(E + U) \quad (19)$$

where

$$F(E) = \int_{-\infty}^{+\infty} f(\omega) \varrho_{\bar{d},E}(\omega) d\omega \quad F(E + U) = \int_{-\infty}^{+\infty} f(\omega) \varrho_{\bar{d},E+U}(\omega) d\omega \quad (20)$$

and $f(\omega)$ is the fermi function

$$f(\omega) = \frac{1}{e^{\beta\omega} + 1}. \quad (21)$$

The effect of a small external field is to change the energies by an amount $\sigma \omega_e/2 = \sigma g \mu_B H/2$ where $\sigma = +1$ for spin-up and -1 for spin down and μ_B is the Bohr magneton. The resulting changes in population are of the form

$$\begin{aligned} \delta \langle\langle \bar{n}_{d\sigma} \rangle\rangle &= (F(E + U) - F(E)) \delta \langle\langle \bar{n}_{d-\sigma} \rangle\rangle \\ &+ \sigma \frac{\omega_e}{2} (F'(E) + \langle\langle \bar{n}_{d-\sigma} \rangle\rangle (F'(E + U) - F'(E))). \end{aligned} \quad (22)$$

From equation (17), the magnetic susceptibility is seen to be

$$\Delta\chi = - \frac{g^2 \mu^2}{2} \frac{(1 - \bar{n})(F'(E) + \bar{n} F'(E + U))}{1 - F(E) + F(E + U)} \quad (23)$$

where $\bar{n} = 1/2 (\langle\langle \bar{n}_{d\uparrow} \rangle\rangle + \langle\langle \bar{n}_{d\downarrow} \rangle\rangle)$. In the case of the δ -function peaks in $A_\sigma(\omega)$ (equation 12), the evaluation of the susceptibility is straightforward.

For $E < 0$, $E + U > 0$ and $|E|, E + U \gg kT$, one obtains

$$\Delta\chi \cong \frac{g^2 \mu^2}{2 k T} \frac{(1 - \bar{n}) e^{-\beta|E|} + \bar{n} e^{-\beta(E+U)}}{e^{-\beta|E|} + e^{-\beta(E+U)}} \quad (24)$$

$$= \frac{g^2 \mu^2}{4 k T}. \quad (25)$$

The result is always a Curie law as expected on physical grounds. Note however that the Curie law originates from a cancellation (in numerator and denominator of equation (19)) of the small probability factors, $e^{-\beta|E|}$, $e^{-\beta(E+U)}$, which in turn arise from the value of the Fermi function at the peaks in $A_\sigma(\omega)$. Since usually $kT \ll |E|$ and $kT \ll E + U$, these probability factors are very small, and the cancellation is crucial.

In the case of a more realistic $\varrho_{\bar{a},E}(\omega)$, i.e. perhaps experimentally determined, the integral (20) is more difficult to evaluate. However the zero temperature result may be derived quite generally. One finds immediately

$$F(E) = \int_{-\infty}^0 d\omega \varrho_{\bar{a},E}(\omega) = P_0(E) \quad (26)$$

as defined in equation (11) and

$$F'(E) = -\varrho_{\bar{a},E}(0) \quad (27)$$

so that

$$\Delta\chi(T=0) = \frac{g^2 \mu^2}{2} \frac{(1 - \bar{n}) \varrho_{\bar{a},E}(0) + \bar{n} \varrho_{\bar{a},E+U}(0)}{1 - P_0(E) + P_0(E+U)}. \quad (28)$$

From equation (28), one sees that whenever the virtual state densities at the Fermi energy are non-zero, the $T=0$ susceptibility is finite. Only when $E < 0$, $E+U > 0$ and the respective state densities do not cross the Fermi surface is the zero temperature result divergent as for a Curie law. The limit is consistent with that required for the validity of the Schrieffer-Wolff transformation (see equation 9); however the restriction is quite severe. Generally, one does expect overlap (perhaps small) of the localized state density, and as a result, a finite zero temperature susceptibility.

One can obtain a qualitative determination of $\Delta\chi(T)$ by writing, for $E \ll 0$

$$F(E) = \int_{-\infty}^{+\infty} f(\omega) \varrho_{\bar{a},E}(\omega) d\omega \quad (29)$$

$$= P_0(E) + \int_{-\infty}^{+\infty} \{f(\omega) - f(\omega)|_{T=0}\} \varrho_{\bar{a},E}(\omega) d\omega. \quad (30)$$

Since $\varrho_{\bar{a},E}(\omega)$ is assumed to be a sharply peaked function, the major contributions to $F(E)$ can be seen to be

$$F(E) \simeq P_0(E) - e^{-\beta|E|}. \quad (31)$$

A similar expression is valid for $E \gg 0$.

The susceptibility is then given approximately by

$$\Delta\chi(\tau) = \frac{g^2 \mu^2}{2} \times \frac{(1 - \bar{n}) \varrho_{\bar{a},E}(0) + \bar{n} \varrho_{\bar{a},E+U}(0) + (1/kT) \{(1 - \bar{n}) e^{-\beta|E|} + \bar{n} e^{-\beta(E+U)}\}}{1 - P_0(E) + P_0(E+U) + e^{-\beta|E|} + e^{-\beta(E+U)}}. \quad (32)$$

A Curie-like dependence is expected only when the condition expressed in (13) above is fulfilled.

Of course this crude evaluation of $F(E)$ is only approximately correct. Inclusion of the finite width and proper functional dependence of $\varrho_{\bar{a},E}(\omega)$ and $\varrho_{\bar{a},E+U}(\omega)$ intro-

duces temperature dependent multiplicative factors. We have investigated in some detail the cases of $\varrho_{\bar{a},E}(\omega)$ and $\varrho_{\bar{a},E+U}(\omega)$ being Gaussian, square, and exponential functions. Although there are differences in detailed low temperature behavior, the general features are the following. For the case $E + U/\Delta \gg 1$, $|E|/\Delta \gg 1$, (see Fig. 2) the susceptibility obeys a Curie law at high temperature, and saturates to a finite value at low temperature. The cross-over from Curie law behavior is found at $kT \sim \Gamma^2/E$ for a Gaussian and $kT \sim \Gamma$ for an exponential density of states where Γ is the level

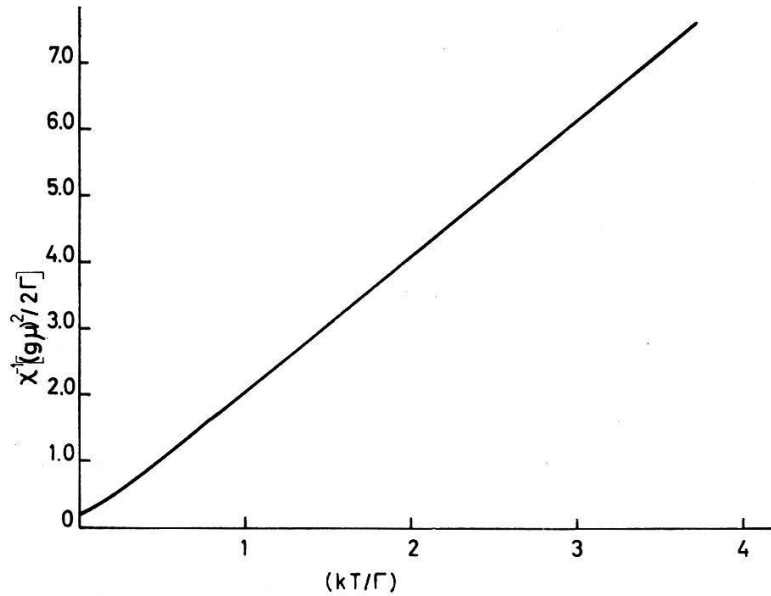


Figure 2

Temperature dependence of the inverse susceptibility for the symmetric case assuming a Gaussian density of states $\varrho_{\bar{a},E} = \gamma \sqrt{\pi} \exp[-\gamma^2 (\omega - E)^2]$ with $|E| = E + U$ and $E = -5\Gamma$. The curve is a spin $-1/2$ Curie law for temperatures larger than Γ^2/E .

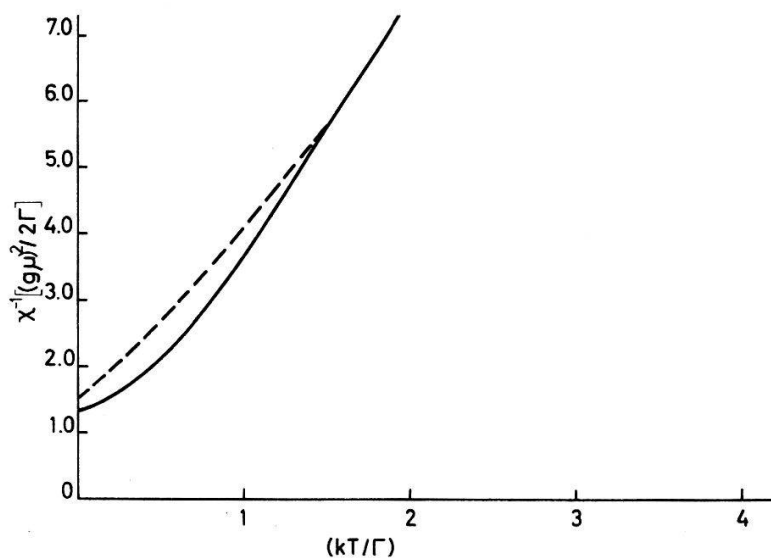


Figure 3

Temperature dependence of the inverse susceptibility for the case when $E + U = 0$. The solid curve assumes a Gaussian density of states, the dashed curve an exponential function $\varrho_{\bar{a},E} = \gamma/2 \exp(-\gamma |\omega - E|)$. Note that the details of the low temperature behavior are sensitive to the form of $\varrho_{\bar{a}}$.

width of the \bar{d} -state. For a Lorentzian function, because of the very long tails, the Curie law is not obtained until very high temperatures ($kT \sim E + U$). For the case $E + U/\Delta \sim 0$, the zero temperature susceptibility as well as the effective moment (as measured by the slope in χ^{-1}) decrease relative to the symmetric case. The detailed behavior again is sensitive to the structure in $\rho_{\bar{d}, E+U}(\omega)$, but can resemble a Curie-Weiss law over a wide range in temperatures (see Fig. 3).

IV. Conclusion

We conclude that these spin fluctuation phenomena can play an important role even in the strongly magnetic limit since a Curie law is obtained at low temperature only when the localized state density does not cross the Fermi surface. These results are in qualitative agreement with the susceptibility measurements of Allali and Donzé [10] on the system of dilute Yb in the alloy $\text{Au}_x\text{Ag}_{1-x}$. The experimental data demonstrate that as the $4f$ virtual level approaches the Fermi surface [11] (i.e. as $x \rightarrow 0$) the zero temperature susceptibility increases as expected from equation (28) [12]. Detailed comparison with theory of the magnitude and temperature dependence of $\Delta\chi$ must await knowledge of the energy dependence of the virtual state density. It is interesting to note further that the optical and photoemission data of Myers and co-workers [9, 13] suggest that for CuMn and AgMn (where the Curie law persists to extremely low temperatures) the localized state density at $\omega = 0$ is very small.

Other effects are immediately suggested. For example the detailed balance relationship inherent in the Hasegawa equations [14, 15] lead to the implication that one should expect deviations from the Korringa relaxation of the impurity spin resonance at low temperatures when χ becomes independent of T .

The effect of the spin fluctuations on the Kondo divergence is being studied. We expect that if the spin fluctuation probability is sufficiently large, the spin 'memory' in the intermediate state will be lost, and the indirect electron-electron interaction [16] responsible for the Kondo effect will disappear.

Finally, comparison with other approaches to the spin fluctuation problem is in order. Zuckermann, Rivier and Sunjic [17], Lederer and Mills [18], Levine and Suhl [19] and Hamann [20] have all approached the spin fluctuation problem starting from the opposite, i.e. non-magnetic limit; and have attempted to selfconsistently form the impurity moment. The latter authors do find a Curie-like temperature dependence [19, 20]. However, the Curie constant is much too small, and the impurity spectral weight function does not show the double-peaked structure characteristic of the magnetic limit (and experimentally observed [9]). In the present case, the particle-particle splitting is built in from the beginning and as a result a Curie constant of the correct magnitude is obtained.

Acknowledgment

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