Alloy-analogy approximation of the degenerate Hubbard model

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In a recent paper [J. van der Rest and F. Brouers, Phys. Rev. B 24, 450 (1983)] it has been claimed that the ground state of the doubly degenerate Hubbard model is always paramagnetic in the alloy-analogy approximation. Here we show that this conclusion was based on an incorrect expression of the spin susceptibility. We give the correct method for calculating the spin susceptibility in the alloy-analogy approximation. In some limiting cases we calculate this susceptibility and show that a ferromagnetic instability can occur. Finally we make some general comments on the applicability and the limitations of the alloy-analogy approximation.

I. INTRODUCTION

Recently van der Rest and Brouers' calculated the spin susceptibility of the doubly degenerate Hubbard model in the alloy-analogy approximation. In order to obtain the expression for the susceptibility they used a method introduced by Brouers and Ducastelle² for the simple Hubbard model. This calculation does not give any ferromagnetic instability: This result contradicts the results obtained in a preceding paper by Lacroix Lyon-Caen and Cyrot, 3 who found that the ground state is ferromagnetic for all values of the band filling between 1 and 2 electrons per atom, in the limit of infinite Coulomb interactions. This result was in agreement with the exact results obtained by the same authors. ⁴

But as was pointed out by van der Rest and Brouers,¹ there is not a unique definition of the energy in the alloyanalogy approximation of the Hubbard model. To avoid this problem we present in this paper a calculation of the spin susceptibility which is exact within the alloy analogy. Thus the results presented in this paper are exact results for the degenerate Hubbard model in the alloy analogy.

In Sec. II we show that the spin susceptibility calculated in Ref. I (see also Ref. 5) was not correct and we give the correct method for calculating the susceptibility. In Sec. III we show that in the limit of infinite Coulomb interaction a ferromagnetic instability can be obtained. In Sec. IV we give some further results in other limiting cases where it is possible to directly calculate the susceptibility. Finally in Sec. V we give some general conclusions on the alloyanalogy approximation.

First we give some definitions and notations which will be used in the following: The doubly degenerate Hubbard model can be written as³

$$
H = H_0 + H_1 = U_1 \sum_{i,m} n_{im} \nvert n_{im} \nvert + U_2 \sum_{i} n_{i1} \nvert n_{i2} \nvert + n_{i1} \nvert n_{i2} \nvert + (U_2 - J) \sum_{i,\sigma} n_{i1\sigma} n_{i2\sigma} + \sum_{\substack{i,j \ m,\sigma}} t_{ij} C_{im\sigma}^{\dagger} C_{jm\sigma} ,
$$
\n(1)

where $m = 1,2$ for the two orbital states. H_0 is the atomic part of H and H_1 describes the conduction band.

In the alloy analogy the motion of the (m, σ) electrons is

described by the Hamiltonian
\n
$$
H_{m\sigma} = \sum_{i,j} t_{ij} C_{jm\sigma} + \sum_{i} E_{i}^{m\sigma} n_{im\sigma} \qquad (2)
$$

where the energy $E_l^{m\sigma}$ depends on the configuration of site i. There are eight possible values of E_i , and the corresponding probabilities $P_i^{m\sigma}$ depend on the electron numbers $\langle n_{m'\sigma'} \rangle$ and on the different correlation function $\langle n_{m\sigma} n_{m'\sigma'} \rangle$, etc. The expressions of E_i and P_i can be found in Ref. 3 where self-consistent calculation of the correlation functions is also explained. The main approximation is that the motion of the (m, σ) electrons is studied by supposing that the other (m', σ') electrons are fixed on some given sites which are randomly distributed; thus dynamical correlations are not well described in this approximation.

II. SPIN SUSCEPTIBILITY IN THE ALLOY-ANALOGY APPROXIMATION

In this section we show how to obtain the exact expression for the spin susceptibility. Our result differs from the expression given by van der Rest and Brouers in Ref. ¹ because some terms were neglected in Ref. 1.

In Refs. 1 and 2 the following formula has been used for the calculation of the susceptibility:

$$
\frac{d\langle n_{1\uparrow}\rangle}{dh} = \frac{\partial\langle n_{1\uparrow}\rangle}{\partial h} + a_1 \frac{d\langle n_{1\downarrow}\rangle}{dh} + a_2 \frac{d\langle n_{2\downarrow}\rangle}{dh} + b \frac{d\langle n_{2\uparrow}\rangle}{dh}
$$
(3)

with

$$
a_1 = \frac{\partial n_{11}}{\partial n_{11}}, \quad a_2 = \frac{\partial n_{21}}{\partial n_{21}}, \quad b = \frac{\partial n_{11}}{\partial n_{21}} \quad . \tag{4}
$$

However, $\langle n_1 \rangle$ depends not only on the other $\langle n_{m',n'} \rangle$ but also on the other correlation functions such as $\langle n_{11}n_{21} \rangle$, etc. Thus, in (4) other terms must be added of the type

$$
\frac{\partial \langle n_{1\uparrow} \rangle}{\partial \langle n_{1\downarrow} n_{2\downarrow} \rangle} \times \frac{d \langle n_{1\downarrow} n_{2\downarrow} \rangle}{dh}
$$

etc. Then similar expressions can be obtained for $d(n_{11}n_{21})/dh$ and there are three independent "susceptibil-

COMMENTS

ities":

$$
X = \frac{d}{dh}(\langle n_{11} \rangle + \langle n_{21} \rangle - \langle n_{11} \rangle - \langle n_{21} \rangle) ,
$$

\n
$$
Y = \frac{d}{dh}(\langle n_{11}n_{21} \rangle - \langle n_{11}n_{21} \rangle) ,
$$

\n
$$
Z = \frac{d}{dh}(\langle n_{11}n_{11}n_{21} \rangle + \langle n_{11}n_{21}n_{21} \rangle - \langle n_{11}n_{11}n_{21} \rangle - \langle n_{11}n_{11}n_{21} \rangle - \langle n_{11}n_{21}n_{21} \rangle) .
$$

\n(5)

(The other derivatives vanish in zero field as, for example, $\langle n_{11}n_{11} \rangle$ must vary as h^2 for small fields.)

We have not calculated X , Y , and Z in the general case because the general expression is rather complicated, but simply in some limiting cases where the calculation can be done very easily, starting directly from the self-consistency equations for $\langle n_{m\sigma} \rangle$ $\langle n_{m\sigma} n_{m'\sigma'} \rangle$, etc. Thus in this way we can be sure that all terms are taken into account.

In Secs. III and IV a semielliptic density of states is considered, as in Refs. 1 and 3, in order to obtain analytical expressions.

III. SUSCEPTIBILITY IN THE CASE U_1 , U_2 , AND $U_2 - J \rightarrow \infty$

In this case, the eight subbands corresponding to the eight values of the energy level E_i are well separated and the density of states in each subband is semielliptic and contains p_{λ} states $\lambda = 1, \ldots, 8$) (Ref. 3).

A. $n < 1$

If $n < 1$ only the lower subband $(\lambda = 1)$ is filled and the susceptibility can be obtained easily:

$$
X = \frac{\rho(E_F)}{1 - b} \quad , \tag{6}
$$

where

$$
b = \frac{1}{\pi} \left(\arcsin \frac{E_F}{W (1 - 3n/4)^{1/2}} + \frac{\pi}{2} \right)
$$

The susceptibility remains finite for all values of n as in the nondegenerate case.⁶

B. $1 < n < 2$

In this case, only the correlation functions $\langle n_{1\sigma}n_{2\sigma}\rangle$ must be considered because the doubly occupied sites must be in the lower-energy state. Thus there are two susceptibilities to calculate: X and Y [Eq. (5)]. The self-consistency equations for this case can be written as

$$
\langle n_{m\sigma} \rangle = \sum_{\lambda=1,2} P_{m\sigma}^{\lambda} \int^{E_{F}} \rho_{m\sigma}^{\lambda}(\omega) d\omega , \qquad (7)
$$

$$
\langle n_{1\sigma} n_{2\sigma} \rangle = \frac{1}{2} \left[P_{1\sigma}^{2} \int^{E_{F}} \rho_{1\sigma}^{2}(\omega) d\omega + P_{2\sigma}^{2} \int^{E_{F}} \rho_{2\sigma}^{2}(\omega) d\omega \right] , \qquad (8)
$$

where

$$
P_{11}^{1} = 1 - \langle n_{11} \rangle - \langle n_{21} \rangle - \langle n_{21} + \langle n_{11} n_{21} \rangle ,
$$

$$
P_{11}^{2} = \langle n_{21} \rangle .
$$

 ρ_{mg}^1 and ρ_{mg}^2 are the density of states of the lowest two subbands in the presence of an applied field h .

$$
\rho_{m\sigma}^{\lambda}(\omega) = \frac{2}{\pi W^2} \left[P_{m\sigma}^{\lambda} W^2 - (z - E_{\lambda} + \sigma h)^2 \right]^{1/2} \quad . \tag{9}
$$

Differentiating directly Eqs. (7) and (8) with respect to h gives, in principle, the susceptibilities X and Y . However, the two Eqs. (7) and (8) lead exactly to the same equation relating X and Y :

$$
Y = \frac{X}{2\pi} \left(\arcsin \frac{E_F - U_2 + J}{W\sqrt{n/4}} + \frac{\pi}{2} \right) + \frac{4}{\pi} \left(\frac{n}{4} W^2 - (E_F - U_2 + J)^2 \right)^{1/2} .
$$
 (10)

Thus we cannot calculate the susceptibility using this method. This result can be related to the fact that Eqs. (7) and (8) are equivalent, i.e., there is a solution for each value of the magnetization M .

In order to obtain another relation between X and Y we have considered the case of finite $J(U_1, U_2 \rightarrow \infty)$ and we have performed a development in powers of $1/J$. Up to second order in $1/J$ the Green's function in the $\lambda = 2$ subband is given by

$$
G^{\sigma}(\omega + U_2 - J) = \frac{2}{W^2} \left\{ \omega + \sigma h - \frac{\alpha}{4} - \left[\left(\omega + \sigma h + \frac{\alpha}{4} \right)^2 - P_2^{\sigma} W^2 \right]^{1/2} - \frac{\alpha'}{4} \left[2(\omega + \sigma h) + [(\omega + \sigma h)^2 - P_2^{\sigma} W^2]^{1/2} + \frac{(z + \sigma h)^2}{[(z + \sigma h)^2 - P_2^{\sigma} W^2]^{1/2}} \right] \right\} ,
$$
 (11)

where

$$
\alpha = \left(\frac{P_3^g}{J} + \frac{P_4^g}{U_1 - U_2 + J}\right) W^2 ,
$$

$$
\alpha' = \left(\frac{P_3^g}{J^2} + \frac{P_4^g}{(U_1 - U_2 + J)^2}\right) W^2 .
$$

The differentiation of Eqs. (7) and (8) now gives two dis-

 $X = \frac{n}{E_F - U_2 + J}$. (12)

Thus, the susceptibility is negative for $E_F < U_2 - J$, i.e., $1 < n < \frac{4}{3}$ and positive for $\frac{4}{3} < n < 2$ [Eq. (7) shows that $E_F = U_2 - J$ if $n = \frac{4}{3}$.

ty. For infinite J we obtain the simple result

The same expression can be obtained if we make the following assumption, which seems physically reasonable, on the number of sites with up and down spin, respectively. We call x_{σ} the fraction of sites with σ spin electrons and we suppose that x_{σ} is proportional to the total number of σ spin electrons: $x_{\sigma} = (n_{1\sigma} + n_{2\sigma})/n$. The fraction of sites occupied by two σ spin electrons is $x_{2\sigma} = \langle n_{1\sigma} n_{2\sigma} \rangle$ and the fraction of sites occupied by one σ spin electron is then $x_{1\sigma} = x_{\sigma} - x_{2\sigma}$. The total number of σ spin electrons is $n_{1\sigma} + n_{2\sigma} = x_{1\sigma} + 2x_{2\sigma}$. Differentiating this equation gives another relation between X and Y :

$$
Y = X(n-1)/n \tag{13}
$$

Inserting Eq. (13) in Eq. (10) gives the same expression for X [Eq. (12)]. In fact, this result was not obvious since the above expression for x_{σ} is not trivial, but the physical meaning is clear: The fraction of doubly occupied sites is the same in the up-spin and down-spin regions (at least to first order in h), $x_2t/x_1 = x_2t/x_1$.

Mizia7 calculated the susceptibility in the same limiting case, and found that $X^{-1} = 0$ for $1 < n < 2$. But his starting formula is not correct as he also neglected some terms of the type $\partial \langle n_{m\sigma} \rangle / \partial \langle n_{1\sigma} n_{2\sigma'} \rangle$.

Thus we have shown that the susceptibility may be negative. Our calculation has been done directly from the selfconsistency equations instead of starting from a more general formula as in Refs. ¹ and 7. The direct calculation is difficult to do in the general case but our result can be used to test the various expressions which have been proposed.

IV. GROUND STATE IN OTHER LIMITING CASES

A.
$$
J=0, U_1=U_2 \rightarrow \infty
$$

If $n < 1$ the ground state is the same as in the preceding case as there is still no doubly occupied sites.

On the other hand, for $1 < n < 2$ the result must be different: The total number of doubly occupied sites is still equal to $n-1$ but the correlation functions $\langle n_{m} \nvert n_{m} \rangle$ no longer vanish. We put $L = \langle n_{m} | n_{m'} \rangle$ (independent of m and m') and $K_{\sigma} = \langle n_{1\sigma} n_{2\sigma} \rangle$.

The three subbands $\lambda = 2$, 3, and 4 are centered around the energy U and we have for these subbands

$$
G_{2}^{g} = G_{3}^{g} = G_{4}^{g} = \frac{G^{\sigma}}{1 + (\Sigma^{\sigma} - U)G^{\sigma}}
$$

$$
= \frac{G^{\sigma}}{P_{2}^{\sigma} + P_{3}^{\sigma} + P_{4}^{\sigma}}.
$$
(14)

We can deduce the expressions of $n_{m\sigma}$, K_{σ} , and L:

$$
\langle n_{m\sigma} \rangle = P_1^{\sigma} + \int^{E_F} \rho^{\sigma}(\omega) \, d\omega \quad , \tag{15}
$$

$$
L = \frac{1}{2} \sum_{\sigma} \frac{P_{\sigma}^{\sigma}}{P_{\sigma}^{\sigma} + P_{\sigma}^{\sigma} + P_{\sigma}^{\sigma}} \int_{-\infty}^{E_F} \rho^{\sigma}(\omega) d\omega , \qquad (16)
$$

$$
K_{\sigma} = \frac{P_{\sigma}^{\sigma}}{P_{\sigma}^{\sigma} + P_{\sigma}^{\sigma} + P_{\tau}^{\sigma}} \int^{E_{F}} \rho^{\sigma}(\omega) d\omega , \qquad (17)
$$

where $\rho^{\sigma}(\omega)$ is the density of states in the subband located at energy U.

Differentiating Eqs. (15), (16), and (17) we can obtain

the susceptibility:

$$
X = \rho(E_F) \frac{2+n}{4(n-1) - b(7n-10)} \tag{18}
$$

where

$$
b = \frac{1}{\pi} \left(\arcsin \frac{2(E_F - U)}{\sqrt{4 - n}W} + \frac{\pi}{2} \right) .
$$

 X^{-1} do not vanish for $1 < n < 2$: If $J=0$ there is no ferromagnetic instability.

B.
$$
U_2-J=0
$$
, U_1 and $U_2 \rightarrow \infty$

In this case, the two subbands $\lambda = 1$ and 2 are at the same energy. As U_1 and U_2 are infinite, $\langle n_{m} \nvert n_{m'} \rangle = 0$. The Green's function in the lower subband is given by

$$
\frac{P_1^{\sigma} + P_2^{\sigma}}{1 + \Sigma^{\sigma} G^{\sigma}} = 1\tag{19}
$$

and the density of states in this subband is

$$
\rho^{\sigma}(\omega) = -\frac{1}{\pi} \operatorname{Im} G^{\sigma}(\omega) ;
$$

we can write easily the self-consistency equations:
\n
$$
\langle n_{m\sigma} \rangle = \int^{E_F} \rho^{\sigma}(\omega) d\omega , \qquad (20)
$$

$$
K_{\sigma} = \frac{P_{2}^{\sigma}}{P_{1}^{\sigma} + P_{2}^{\sigma}} \int_{-\infty}^{E_{F}} \rho^{\sigma}(\omega) d\omega
$$
 (21)

Equation (21) gives

$$
K_{\sigma} = \frac{(n_{m\sigma})^2}{1 - 2n_{m-\sigma} + K_{-\sigma}} \quad . \tag{22}
$$

Equation (22) is valid for all values of *n* between 1 and 2. In the paramagnetic state we have

$$
K_{\sigma} = \frac{1}{2} \left[\frac{n}{2} - 1 + \left[1 - n + \frac{n^2}{2} \right]^{1/2} \right]
$$

and in the ferromagnetic state $n_{m} = n/2$, $K_t = n^2/4$. We. can obtain the susceptibility in the paramagnetic state:

$$
X = \rho(E_F) \frac{(1 - n + n^2/2)^{1/2} - 1 + n}{(1 - n + n^2/2)^{1/2} - 1 + n - nb} \tag{23}
$$

where

(14)
$$
b = \frac{1}{\pi} \left[\arcsin \frac{E_F}{W\sqrt{P}} + \frac{\pi}{2} \right],
$$

$$
P = \frac{1}{2} [1 - n/2 + (1 - n + n^2/2)^{1/2}]
$$

The susceptibility does not diverge for any value of the electron number between 0 and 2.

The same case was studied by Dowson⁸ who found that the ground state is ferromagnetic for $n > 0.45$. However, in this work the chemical potential was not the same in the upand down-spin bands. In fact, when looking at the selfconsistency equations (20) and (21) we can see that the only solution is the paramagnetic solution. We have P_1^{σ} only solution is the paramagnetic solution. We have P_1^{ϵ}
+ P_2^{ϵ} = 1 - n + K - σ ; the width of the σ spin band is given by $(P_1^{\sigma} + P_2^{\sigma}) W$. Thus if $n_1 > n_1$, we must also have

 $K_1 > K_1$ and this implies that $P_1^{\dagger} + P_2^{\dagger} < P_1^{\dagger} + P_2^{\dagger}$ and n_1 cannot be larger than n_1 . Thus, in order to obtain a ferromagnetic solution, Dowson had to shift the \uparrow and \downarrow spin bands by taking different chemical potentials.

We have also calculated the energy of the paramagnetic solution and compared with the energy of the ferromagnetic state with maximum magnetization: As we have shown, this state cannot be a solution of the self-consistency equation but it is a solution of the Hubbard model. The comparison shows that thc ferromagnetic state has a lower energy above the concentration $n_0 \approx 0.5$ close to the value obtained by Dowson.

Now if $U_2 - J$ is small but nonzero, to the first order in $U_2 - J$ the increase in energy is given by ΔE $=(U_2-J)\sum_{\sigma}(n_{1\sigma}n_{2\sigma}).$ In the ferromagnetic state the number of doubly occupied sites is always larger than in the paramagnetic one $[Eq. (22)]$ because in the paramagnetic state $\langle n_{1\sigma}n_{2\sigma}\rangle$ is reduced by the correlations; thus the critical value n_0 should increase with $U_2 - J$. In fact, n_0 is equal to 1 if $U_2 - J \rightarrow \infty$.

V, CONCLUSIONS

In this paper we have obtained the exact spin susceptibility in the alloy-analogy approximation for the degenerate Hubbard model. We have calculated the susceptibility in some limiting cases and we have shown that a ferromagnetic instability can be obtained.

However, for infinite Coulomb interaction we have ob-

tained a ferromagnetic instability only for $1 < n < \frac{4}{3}$, whereas the exact results⁴ show that the ground state is ferromagnetic for $1 < n < 2$. This discrepancy can be due to the approximation introduced by the alloy analogy or it could reflect the fact that the magnetic-nonmagnetic transition is a first-order transition for $\frac{4}{3} < n < 2$.

In fact, the analogy with a disordered alloy is correct in the degenerate case for the description of the spin disorder (for $1 < n < 2$), whereas this analogy is difficult to justify in the nondegenerate Hubbard model. Indeed in the alloy analogy the (m, σ) electrons are moving in a potential due to the other $(m'\sigma')$ electrons which are supposed to be fixed on some given sites. In the degenerate case, in the limit of U_1 , $U_2 \rightarrow \infty$ ($U_2 - J$ being finite or not) it is possible to separate up- and down-spin regions⁴ for $1 < n < 2$, because it is possible to fix the spin of any given site.⁹ This site will have either one or two electrons but always with the same spin. Thus for the up-spin electrons, the down-spin sites are forbidden, and if the down-spin sites are randomly distributed in the crystal, we have really an alloy with infinite energy separation. The alloy analogy is correct for the spin disorder, but not for the orbital disorder because in each of the regions the two orbitals are mixed.

However, as in the nondegenerate case there are some limitations to this approximation: The strongest onc, concerning the magnetic ordering, is that there is no antiferromagnetic instability in this alloy analogy, 10 whereas the exact ground state is known to be antiferromagnetic for half-filled bands.

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