Temperature Dependence of Local Magnetic Moments in Paramagnetic Metals: Hubbard's Alloy Analogy*

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The temperature T dependence of the local magnetic moments and the static uniform spin susceptibility $\chi(T)$ in paramagnetic metals is studied using the Hubbard Hamiltonian for nondegenerate bands. Hubbard's alloy analogy, which correctly describes both strongly interacting and weakly interacting systems of electrons, is applied to determine the average local-moment magnitude $m_l(T) \equiv N^{-1}\sum_i |n_{i\uparrow} - n_{i\downarrow}|$ and $\chi(T)$. It is found that the magnetic-moment magnitude decreases gradually with temperature and does *not* undergo a phase transition to a zero value. This is in contrast to the results of previous theories which predict a sharp phase transition associated with a disappearance at high temperatures of the moments. The moment magnitude which is a function of the ratio of the Coulomb repulsion energy U to the half-bandwidth W and of the number of electrons per site $2n$ approaches the *finite* value $m_l(T \to \infty) = 2n(1-n)$ at temperatures high compared to U and W, which is just the probability that a noninteracting electron and hole of opposite spin are at the same site. At these temperatures $\chi(T \to \infty)$ obeys a Curie law with Curie constant given by $m_l(T \to \infty)$. For $U \ll W$, $m_l(T)$ is nearly temperature independent and approximately equal to $2n(1-n)$. For $U \gg W$ and $kT \ll U$ the local moment has its maximum possible magnitude, which is either $2n$ or $2(1-n)$, corresponding to electrons ($n < \frac{1}{2}$) or holes ($n > \frac{1}{2}$), respectively. In addition, the behavior of the paramagnetic susceptibility $\chi(T)$ which is found to be smoothly varying with temperature does *not* suggest the existence of a phase transition associated with a disappearance of local magnetic moments.

I. INTRODUCTION

The possible existence of two phase transitions occurring in the nondegenerate Hubbard Hamiltonian,[1] which includes only intra-atomic Coulomb interactions, has been recently studied.[2,3] The first of these phase transitions was associated with the disappearance of a macroscopic state with long-range order at a temperature T_0. The second was associated with the complete "melting" of local magnetic moments at a temperature T_l. Depending on the ratio of the Coulomb interaction energy U to the half-bandwidth W, the value of T_0 was found to be larger or smaller than T_l. In these previous theoretical calculations of T_l, T_0, and the local moment at a particular site, assumptions about the moment magnitude and direction at neighboring sites were made.[2,3] In addition, the ordering temperature and the melting temperature were not computed self-consistently, i.e., within the same calculational framework.[2,3] It is felt that such assumptions and approaches greatly restrict the validity of former theories and lead to erroneous conclusions.

It is the purpose of the present paper, using the Hubbard-model Hamiltonian, to present a different method for determining the temperature dependence of the average local-moment magnitude $m_l(T) \equiv N^{-1}\sum_i |n_{i\uparrow} - n_{i\downarrow}|$ in metals, which avoids some of these previous approximations and pitfalls. The local-moment magnitude at a site i is computed independently of any assumptions about the direction or magnitude of the moments on lattice sites $j \neq i$. Furthermore, in contrast to previous calculations, in the present approach $m_l(T)$ and the ordering temperature T_0 are calculated within the same framework. It will be shown here that Hubbard's alloy analogy,[1] within which the one-electron properties of electrons in narrow nondegenerate energy bands are calculated from the electronic properties of an appropriate binary alloy, can be used to obtain in a simple way the average moment magnitude $m_l(T)$ as well as the static uniform spin susceptibility $\chi(T)$. The temperature dependence of $m_l(T)$ and $\chi(T)$ is calculated for a wide range of values of the parameter U/W and for different values of n, where n is the number of electrons per site of a single spin state. While the expressions for $\chi(T)$ calculated in the present paper are identical to those obtained by Hubbard and Jain[4] using more complicated equation-of-motion techniques, they are presented here for several reasons. First, they are used to calculate the ordering temperature T_0 which, because only the uniform susceptibility is considered, is equivalent to the Curie temperature T_C. Second, these expressions for $\chi(T)$ enable one to determine whether the previously predicted[2,3] phase transition of the local moments is reflected in $\chi(T)$. Third, they illustrate how one can obtain in a simple physical way the usual Hartree-Fock ($U/W \ll 1$), the t-matrix approximation ($n \ll 1$), and strong scattering ($U/W \gg 1$) limits of

the susceptibility for the Hubbard Hamiltonian. The present theory is exact in the low-density limit $n \ll 1$ (which is the analog of the dilute alloy[5,6]), the weak-interaction limit $U/W \ll 1$ (which is the analog of the rigid-band alloy[5,6]), and the atomic limit $W = 0$ (which is the analog of the "atomic alloy"[5,6]).

Since the alloy analogy is only appropriate to paramagnetic systems or those with ferromagnetic order,[7] attention here will primarily be confined to paramagnetic metals. In this way it is possible to avoid the difficult problem of ascertaining the nature of the magnetically ordered state below T_0 in the Hubbard Hamiltonian.

The conclusions reached in this paper are as follows. In paramagnetic metals there is *no* phase transition associated with a disappearance of local moments. Instead the moment magnitude, which is always nonzero for $0 < n < 1$, decreases gradually with temperature. At temperatures high compared to U and W, the value of $m_l(T)$ approaches the *finite* value $2n(1-n)$, which is just the probability that a noninteracting electron and hole of opposite spin are at the same site. There is furthermore no evidence in the calculated paramagnetic susceptibility $\chi(T)$ which could suggest a possible phase transition associated with the disappearance of local moments.

An outline of the paper will now be given. In Sec. II Hubbard's Hamiltonian and alloy analogy are reviewed. The equivalence of the coherent-potential approximation (CPA),[5,6] first noted by Velický *et al.*,[5] and the Green's-function truncation procedures introduced by Hubbard[1] are discussed and the local-moment magnitude is expressed in terms of the spin-configuration-averaged resolvent for the Hamiltonian. Using the CPA, general expressions for $m_l(T)$ and $\chi(T)$ are obtained for arbitrary U/W and n. In the high-temperature limit these expressions are shown to reduce to simple and physically understandable results.

In Sec. III an approximate spin-configuration-averaged resolvent for the Hubbard Hamiltonian is calculated for several values of the parameters U/W and n using the alloy analogy and the CPA. The temperature dependence of $m_l(T)$ and $\chi(T)$ is discussed. Numerical examples are used to compare the magnitude of $m_l(T)$ for different parameters U/W and n. Finally in Sec. IV the conclusions of the present calculation, which were stated above, are compared with those of previous theories[2,3] and with experiment.

II. CALCULATION OF LOCAL MAGNETIC MOMENTS AND SPIN SUSCEPTIBILITY WITHIN HUBBARD'S ALLOY ANALOGY

The Hubbard Hamiltonian is given by

$$\hat{\mathcal{H}} = \sum_{i \neq j, \sigma} T_{ij} \hat{C}^{\dagger}_{i\sigma} \hat{C}_{j\sigma} + \tfrac{1}{2} \sum_{i,\sigma} U \hat{n}_{i\sigma} \hat{n}_{i-\sigma} , \tag{2.1}$$

where T_{ij} is the hopping matrix element and U the intra-atomic Coulomb repulsion energy. The operators $\hat{C}^{\dagger}_{i\sigma}$ and $\hat{C}_{i\sigma}$ are the usual creation and annihilation operators for an electron with spin σ at the site i and $\hat{n}_{i\sigma} = \hat{C}^{\dagger}_{i\sigma} \hat{C}_{i\sigma}$.[8] As first pointed out by Hubbard,[1] the motion of electrons with spin $s = \pm\sigma$ can be approximately described by the following alloy Hamiltonian:

$$\hat{\mathcal{H}}_s = \sum_{i \neq j} T_{ij} \hat{C}^{\dagger}_{is} \hat{C}_{js} + \sum_i \epsilon^i_s \hat{C}^{\dagger}_{is} \hat{C}_{is}, \quad s = \pm\sigma \tag{2.2}$$

where ϵ^i_σ is equal to $\epsilon^A \equiv U$ or $\epsilon^B \equiv 0$, depending on whether an electron with spin $-\sigma$ is at site i or not. Equation (2.2) can be viewed as describing two interdependent binary alloys: $A_{n_{-\sigma}} B_{1-n_{-\sigma}}$ of electrons with spin σ and $A_{n_\sigma} B_{1-n_\sigma}$ of electrons with spin $-\sigma$. Here n_σ is the average number of electrons per site with spin σ. The relative concentration of atoms with energy level ϵ^A is $n_{-\sigma}$ or n_σ, and with energy level ϵ^B is $1-n_{-\sigma}$ or $1-n_\sigma$, corresponding to whether the electrons have spin σ or $-\sigma$, respectively. Furthermore, in approximating Eq. (2.1) by Eq. (2.2) it is assumed that for the purposes of calculating the motion of σ spin electrons, electrons of spin $-\sigma$ are fixed at the lattice sites and vice versa. Thus the resonance-broadening correction of Hubbard[1] is not included here.

To calculate the one-electron properties of the Hubbard Hamiltonian, the spin-configuration-averaged resolvent $\hat{G}_\sigma(E)$ for electrons with energy E and spin σ is needed. $\hat{G}_\sigma(E)$ is the propagator for electrons of spin σ in the translationally invariant system in which the spin configuration is identical at each site and is given by the average spin configuration in the solid. $\hat{G}_\sigma$ can be written

$$\hat{G}_\sigma = (E - \hat{\mathcal{H}}^{\mathrm{eff}}_\sigma)^{-1} , \tag{2.3}$$

where

$$\hat{\mathcal{H}}^{\mathrm{eff}}_\sigma = \sum_{i \neq j} T_{ij} \hat{C}^{\dagger}_{i\sigma} \hat{C}_{j\sigma} + \hat{\Sigma}_\sigma(E) . \tag{2.4}$$

Here $\hat{\Sigma}_\sigma(E)$ is the self-energy operator which describes the average effect on an electron with energy E and spin σ of the Coulomb interactions with all the other electrons in the metal. As in most alloy problems $\hat{G}_\sigma$ can only be calculated approximately. However, the form of the alloy analog Hamiltonian equation (2.2) makes application of the CPA[5,6] easy. It follows from Refs. 5 and 6 that the self-energy operator $\hat{\Sigma}_\sigma(E)$ for the alloy Hamiltonian is given within the CPA by

$$\hat{\Sigma}_\sigma(E) = \Sigma_\sigma(E) \hat{1}_\sigma , \tag{2.5}$$

where

$$\Sigma_\sigma(E) = U n_{-\sigma} + \frac{(1-n_{-\sigma}) n_{-\sigma} U^2 F(E - \Sigma_\sigma(E))}{1 - [(1-n_{-\sigma})U - \Sigma_\sigma(E)] F(E - \Sigma_\sigma(E))} \tag{2.6}$$

and

$$F(E-\Sigma_\sigma(E))=\langle i\,|\,\hat{G}_\sigma\,|\,i\rangle\ . \tag{2.7}$$

Here $\hat{1}_\sigma$ is the unit operator and Eq. (2.6) is equivalent to Eq. (4.23) of Ref. 5 except for a difference in the origin of energy. Equation (2.6) was previously derived by Hubbard[1] from equation-of-motion techniques. The observation that Hubbard's truncation scheme is equivalent to the CPA applied to Eq. (2.2) is due to Velický *et al.*[5] It can be noted using Eqs. (2.4) and (2.6) that in the limit $U/W \ll 1$, the self-energy $\Sigma_\sigma(E)=Un_{-\sigma}$ and thus $\hat{\mathcal{H}}_\sigma^{\text{eff}}$, reduce to the usual Hartree-Fock approximation to the Hubbard Hamiltonian. In the atomic limit $U/W=\infty$, in which the resolvent of Eq. (2.1) can be obtained exactly, the calculated expression for $\hat{G}_\sigma$ obtained from Eqs. (2.4) and (2.6) reduces to the exact expression[1] for $\hat{G}_\sigma$. The alloy approximation is thus valid for both large and small values of the parameter U/W. It is therefore expected to be a reasonable interpolation scheme for calculating the spin-configuration-averaged resolvent for the Hubbard Hamiltonian for arbitrary U/W.

The density of states per atom of electrons with spin σ is given by

$$\rho_\sigma(E)=-(\pi N)^{-1}\,\mathrm{Im}\,\mathrm{Tr}\hat{G}_\sigma(E+i0)\ , \tag{2.8}$$

which can be simply related to the density of states for noninteracting electrons $\rho_\sigma^0(E)$ using the equation

$$-(\pi)^{-1}\,\mathrm{Im}\int_{-\infty}^{\infty}dE'\,\rho_\sigma^0(E')(E-\Sigma_\sigma(E)-E')^{-1}=\rho_\sigma(E)\ .$$

The average number of electrons per atom with spin σ satisfies the equation

$$n_\sigma=\int_{-\infty}^{\infty}dE\,(1+e^{(E-\mu)/kT})^{-1}\rho_\sigma(E)\ , \tag{2.9}$$

where the Fermi energy μ is related to the number of electrons per site $2n$ through the identity $2n \equiv n_\sigma+n_{-\sigma}$. In order to compute the self-energy $\Sigma_\sigma(E)$ as a function of $\rho_\sigma^0(E)$, $2n$, and U, Eqs. (2.6) and (2.9) must be solved simultaneously at each temperature for the five unknown quantities $\Sigma_\sigma(E)$, $\Sigma_{-\sigma}(E)$, n_σ, $n_{-\sigma}$, and μ.

Until now this section has been essentially a review of previous work.[1,5] It is the purpose of the remainder of the section to go beyond these earlier theoretical results and to calculate both the local magnetic moment and the static uniform spin susceptibility within the framework which has now been set up.

Because of the difficulty in dealing with nontranslationally invariant systems of particles, what is usually calculated in the Hubbard Hamiltonian is the average spin configuration at a site, i.e., n_σ and $n_{-\sigma}$. It is possible, however, to decompose the average configuration at a site into the component configurations which when properly weighted make up the average. The alloy analogy may now be seen to contain useful information about the probability of the occurrence of the possible atomic configurations. Just as in the binary-alloy problem,[5,6] the average electron density of states $\rho_\sigma(E)$ may be decomposed into the component densities $\rho_\sigma^A(E)$ and $\rho_\sigma^B(E)$ which correspond to the density of states of electrons whose spins are, respectively, compensated ($\epsilon_\sigma^i=U$) or uncompensated ($\epsilon_\sigma^i=0$) by electrons with opposite spin at the same site. It follows from the CPA that $\rho_\sigma^A(E)$ and $\rho_\sigma^B(E)$ satisfy the electron-conservation equation[5,6]

$$\rho_\sigma(E)=n_{-\sigma}\rho_\sigma^A(E)+(1-n_{-\sigma})\rho_\sigma^B(E)\ , \tag{2.10}$$

where

$$\rho_\sigma^i(E)=-(\pi)^{-1}\,\mathrm{Im}\langle i\,|\,\hat{G}_\sigma^i(E)\,|\,i\rangle \quad (i=A,B) \tag{2.11}$$

and

$$\langle i\,|\,\hat{G}_\sigma^i(E)\,|\,i\rangle=\frac{F(E-\Sigma_\sigma(E))}{1-[\epsilon_\sigma^i-\Sigma_\sigma(E)]F(E-\Sigma_\sigma(E))}\ . \tag{2.12}$$

In the following, to avoid the difficulties mentioned in Sec. I, attention will be focused on paramagnetic metals for which $n_\sigma=n_{-\sigma}=n$.

There has been a notable lack of precision in the literature[2,3] in defining the local magnetic moment $m_l(T)$ at a site in a crystal. The following definition for $m_l(T)$ is used here:

$$m_l(T)\equiv N^{-1}\sum_i\,|n_{i\uparrow}-n_{i\downarrow}| \tag{2.13a}$$

$$=\langle\,|n_{i\uparrow}-n_{i\downarrow}|\,\rangle_{\text{config av}}\ , \tag{2.13b}$$

where the symbol $\langle\cdots\rangle_{\text{config av}}$ denotes a weighted average over all the possible spin configurations at the site i, $n_{i\sigma}$ is the expectation value of the number operator[9] at the ith site for a given configuration of spins at the N sites in the lattice, and the vertical bars $|\cdots|$ mean that the absolute value of the quantity between them is to be taken. The reasons for defining m_l as in Eq. (2.13) are threefold. First, the local moment as defined above is a measure of the average (spin) angular momentum at each site; second, m_l is easily related to a physically measurable quantity, namely, the spin susceptibility at high temperatures; and third, m_l can be simply calculated within the alloy-analogy framework.[10]

It follows from Eqs. (2.13) that $m_l(T)$ is the average number of electrons per site with uncompensated spins which is given within the alloy analogy by

$$m_l(T)=2(1-n)\int_{-\infty}^{\infty}dE\,(1+e^{(E-\mu)/kT})^{-1}\rho_\sigma^B(E) \tag{2.14a}$$

$$=2n(1-n)+\frac{2(1-n)}{\pi}\,\mathrm{Im}\int_{-\infty}^{\infty}dE\Big[(1+e^{(E-\mu)/kT})^{-1}$$

$$\times \frac{\langle i|\hat{G}_\sigma|i\rangle \Sigma_\sigma(E)\langle i|\hat{G}_\sigma|i\rangle}{1+\Sigma_\sigma(E)\langle i|\hat{G}_\sigma|i\rangle}\Big]. \qquad (2.14b)$$

From Eq. (2.14b) it may be noted that $m_l(T)$ can be decomposed into a constant term $2n(1-n)$ independent of U and a term which is related to the density-density correlation function and which vanishes as $U \to 0$. In the small-U/W limit, for which $\Sigma_\sigma(E) = Un$, the second term in Eq. (2.14b) reduces to the usual expression for the unenhanced local susceptibility times $2n(1-n)U$. In the large-U/W limit, for $kT \ll U$, the second term is given by $2n^2$ if $n < \frac{1}{2}$, and $2(1-n)^2$ if $n > \frac{1}{2}$. This will be discussed in more detail in Sec. III. The first term in Eq. (2.14b) expresses the fact that for any value of U there will always be on the average electrons with uncompensated spins at a site in a metal unless the electron bands are completely full ($n = 1$) or completely empty ($n = 0$).

From Eq. (2.14b) it follows that at temperatures high compared to $|E^{\max} - E^{\min}|$, where $E^{\min}$ and $E^{\max}$ are, respectively, the lower and upper energy bounds of the bands for interacting electrons, we have

$$m_l(T \to \infty) = 2n(1-n), \quad kT \gg |E^{\max} - E^{\min}| \,. \qquad (2.15)$$

In Eq. (2.15) the limit $\langle i|\hat{G}_\sigma|i\rangle \sim E^{-1}$ as $E \to \infty$ has been used. Equation (2.15) can be derived without using any approximation scheme. This exact result expresses the fact that *at temperatures high compared to U and W the average local moment is given by the probability that a noninteracting electron and hole of opposite spin are at the same site*. These results may be illustrated for the case of a half-filled band, $n = \frac{1}{2}$. When $kT \gg U, W$ there are four equally likely spin configurations which may exist at a given site. These four states are shown schematically in Fig. 1. The first two correspond to a local moment of 1. The last two correspond to zero local moment. The average moment magnitude at a site is thus $\frac{1}{2} = 2n(1-n)$. By contrast the average vector moment $\langle n_{i\uparrow} - n_{i\downarrow}\rangle_{\text{config av}}$ is clearly zero. The metal at high temperatures, therefore, can be thought of as a system of local moments of average magnitude $\frac{1}{2}$.

Finally the uniform static spin susceptibility

$$\chi = \mu_B \lim_{H \to 0} [n_\uparrow(H) - n_\downarrow(H)]/H \qquad (2.16)$$

may be calculated from Eq. (2.9) by replacing the energy levels ϵ_σ^i by the spin- and field-dependent quantities $\epsilon_\sigma^i - \mu_B \sigma H \equiv \epsilon_\sigma^i(H)$. Here μ_B is the Bohr magneton and H the external magnetic field. It should be noted that in the presence of a magnetic field the relative concentration of the energy levels $\epsilon_\sigma^A(H)$ and $\epsilon_\sigma^B(H)$ is field dependent and is given by $n_{\pm\sigma}(H)$ and $1 - n_{\pm\sigma}(H)$, respectively. As a result the self-energy and the configuration-averaged resolvent also depend on H.

The high-temperature limit of the paramagnetic susceptibility $\chi(T \to \infty)$ may be easily calculated and related to $m_l(T \to \infty)$. Since the interactions between electrons are unimportant at temperatures large compared to U and W, $\chi(T \to \infty)$ is given by an expression appropriate to noninteracting electrons:

$$\chi(T \to \infty) = 2\mu_B^2 \frac{\partial n}{\partial \mu}, \quad kT \gg |E^{\max} - E^{\min}| \,. \qquad (2.17)$$

As $T \to \infty$,

$$n \cong (1 + e^{-\mu/kT})^{-1} \int_{-\infty}^{\infty} dE\, \rho_\sigma^0(E) \,.$$

Thus $2\partial n/\partial\mu$ is equal to $2n(1-n)/kT = m_l(T \to \infty)/kT$. It then follows from Eq. (2.17) that

$$\chi(T \to \infty) = \mu_B^2 m_l(T \to \infty)/kT \,. \qquad (2.18)$$

The susceptibility at high temperatures is thus proportional to the local-magnetic-moment magnitude $m_l(T \to \infty)$. It should be noted that Eq. (2.18) exhibits the appropriate symmetry between electrons and holes. However, only in the nearly free-electron limits $n \ll 1$ or $1 - n \ll 1$ does $\chi(T \to \infty)$ reduce to the classical result. This apparent violation of the correspondence principle is due to the fact that Eq. (2.18) represents the intraband contribution to the susceptibility. The contribution to χ of additional bands is thus neglected in this equation; only when these contributions are included by modifying the original Hamiltonian [Eq. (2.1)] will the classical result be obtained for $T \to \infty$.

In Sec. III the temperature dependence of $m_l(T)$ and $\chi(T)$ is discussed for special values of the parameters U/W and n. In addition, the Curie tem-

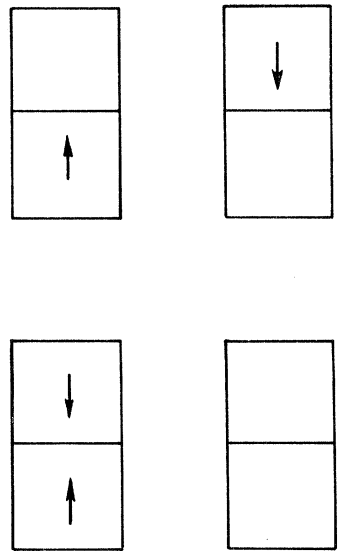

FIG. 1. Local spin configurations for $kT \gg U$, W and for $n = \frac{1}{2}$.

perature T_C, which is the temperature below which the paramagnetic state is unstable with respect to ferromagnetism, is obtained. T_C can be calculated either from the pole in the susceptibility or from the equation $n_\sigma(T) = n_{-\sigma}(T)$. Within the present framework it is not possible to calculate easily the wave-number dependence of the susceptibility. Consequently the Néel temperature T_N, which is the temperature below which the paramagnetic state is unstable with respect to antiferromagnetism, will not be considered.

III. BEHAVIOR OF LOCAL MAGNETIC MOMENT $m_l(T)$ AND SPIN SUSCEPTIBILITY $\chi(T)$ IN SEVERAL LIMITS

In this section the temperature dependence of the local-magnetic-moment magnitude $m_l(T)$ and of the spin susceptibility $\chi(T)$ is studied in three limiting cases in which the electron self-energy $\Sigma_\sigma(E)$ [Eq. (2.6)] may be easily determined. These three limits, for which the analogs in alloy theory have also been extensively discussed,[5,6] are (i) the Hartree-Fock approximation $U \ll W$, corresponding to the rigid-band approximation in alloy theory; (ii) the t-matrix approximation $n \ll 1$, corresponding to the dilute-alloy limit; and (iii) the strong-interaction limit $U \gg W$, corresponding to the split-band limit in alloy theory.

A. Temperature Dependence of $m_l(T)$ and $\chi(T)$ in the Hartree-Fock Approximation

In the Hartree-Fock approximation ($U/W \ll 1$) the electron self-energy in Eq. (2.6) reduces to

$$\Sigma_\sigma(E) = Un \,. \tag{3.1}$$

From Eq. (2.14a) it follows that

$$m_l(T) \cong 2n(1-n) + 2n(1-n)\pi^{-1} U \,\mathrm{Im} \int_{-\infty}^{\infty} dE\, F^2(E) / (1 + e^{(E-\mu)/kT}) + O(U/W)^2 \,. \tag{3.2}$$

It is evident that, except for small corrections of order, (U/W), $m_l \cong 2n(1-n)$ is essentially temperature independent. The second term in Eq. (3.2) which may be related to the non-exchange-enhanced local susceptibility[11] approaches zero for temperatures large compared to W. The temperature dependence of $m_l(T)$ is shown in Fig. 2. Here as in all subsequent figures it is assumed for simplicity that the density of states of noninteracting electrons is given by $\rho_\sigma^0(E) = 2(\pi W)^{-1} \times [1 - (E/W)^2]^{1/2}$. This band shape is sometimes called the Hubbard band model. The characteristic temperature T_l indicated in Fig. 2 is the temperature at which the local magnetic moment (assuming for simplicity $n < \frac{1}{2}$) is reduced to $(2n - 2n^2/3)$. This temperature is in reasonable agreement with the melting temperature of Refs. 2 and 3 in the limit $U/W \ll 1$. Since it has been shown[12] that weakly interacting ($U/W \ll 1$) systems with half-filled bands undergo an antiferromagnetic phase transition, for the case $n = \frac{1}{2}$ the figure is not strictly correct at very low temperatures.

The static uniform spin susceptibility $\chi(T)$ can be obtained from the field-dependent counterpart of Eq. (2.9) by replacing ϵ_σ^i by $\epsilon_\sigma^i(H)$. Expanding Eq. (2.9) about the small parameters $[n_\sigma(H) - n]$ and H it follows that

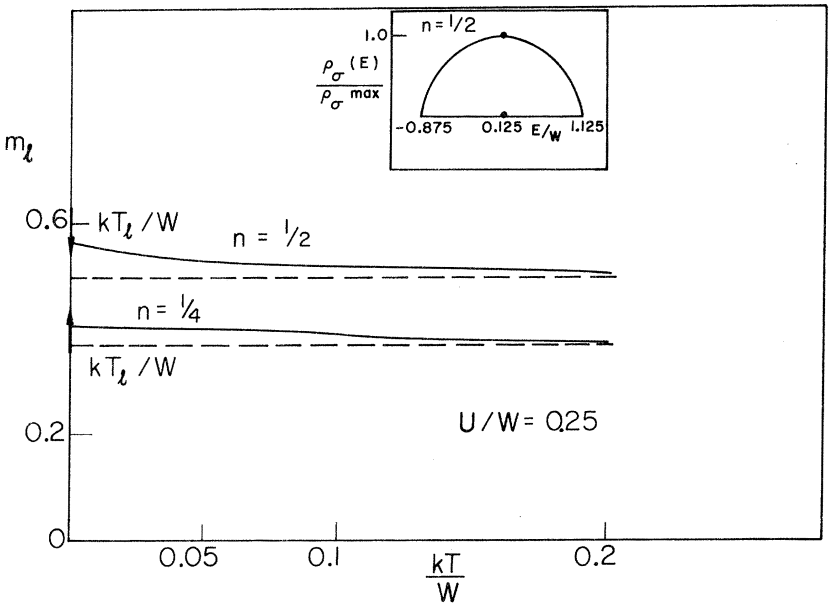

FIG. 2. Temperature dependence of m_l for $U/W = 0.25$ and for $n = \frac{1}{2}$ and $n = \frac{1}{4}$. In the insert is plotted the density of states for interacting electrons.

$$\chi(T)=2\mu_B^2\left(\frac{\partial n}{\partial\mu}\right)_0\Big/\left[1-U\left(\frac{\partial n}{\partial\mu}\right)_0\right] , \qquad (3.3)$$

where $(\partial n/\partial\mu)_0$ is the temperature-dependent density of states per spin at the Fermi energy for noninteracting electrons. Equation (3.3) is the usual expression for χ in the Hartree-Fock limit. Because $U/W \ll 1$ and $(\partial n/\partial\mu)_0 \sim O(W^{-1})$, there is no pole in χ. Thus the system does not undergo a ferromagnetic phase transition for any n.

It may easily be verified from Eq. (3.3) that, for $kT \gg W$, $\chi(T)$ is Curie-Weiss-like. For lower temperatures $\chi(T)$ is predominantly temperature independent.

B. Temperature Dependence of $m_l(T)$ and $\chi(T)$ in the t-Matrix Approximation

In the t-matrix approximation ($n \ll 1$) which is the analog of the dilute-alloy limit in alloy theory, the electron self-energy [Eq. (2.6)] is given by

$$\Sigma_\sigma(E)=nU/[1-UF(E)] . \qquad (3.4)$$

It follows from Eq. (2.14) that

$$m_l(T)=-2(1-n)\pi^{-1}\,\mathrm{Im}\int_{-\infty}^{\infty} dE\,(1+e^{(E-\mu)/kT})^{-1} \times\frac{F\{E-nU/[1-UF(E)]\}}{1+UnF(E)/[1-UF(E)]} . \qquad (3.5)$$

Expanding in powers of n, Eq. (3.5) can be written

$$m_l(T)=2n(1-n)(1+U\pi^{-1}\,\mathrm{Im}\int_{-\infty}^{\infty} dE\,(1+e^{(E-\mu)/kT})^{-1} \times\left(\frac{F^2(E)+\partial F(E)/\partial E}{1-UF(E)}\right)+O(n^2) . \qquad (3.6)$$

Since the integral in Eq. (3.6) is of order n, it follows that

$$m_l(T)\cong 2n+O(n^2) \qquad (3.7)$$

is temperature independent to first order in n. The static uniform spin susceptibility obtained from Eqs. (2.9) and (2.16) is

$$\chi(T)=2\mu_B^2\left(\frac{\partial n}{\partial\mu}\right)\Big/\left[1-U^{\mathrm{eff}}\left(\frac{\partial n}{\partial\mu}\right)\right] , \qquad (3.8)$$

for $kT \ll U, W$. Here U^{eff} is given by

$$U^{\mathrm{eff}}=U/[1+U|F(E^{\mathrm{min}})|] , \qquad (3.9)$$

where $F(E^{\mathrm{min}})$, evaluated at the lower band-edge energy, is real and negative. For the Hubbard band model $F(E^{\mathrm{min}})=-2W^{-1}$. In Eq. (3.8) $\partial n/\partial\mu$ is the density of states at the Fermi energy for interacting electrons. Equation (3.8) may be seen to be equivalent to the usual expression[4,7] for $\chi(T)$ in the limit $n \ll 1$. Because of the smallness of $\partial n/\partial\mu$ near the band edge compared to $(U^{\mathrm{eff}})^{-1} > W^{-1}$, there is no pole in χ for $n \ll 1$. Thus the system does not undergo a ferromagnetic transition.

C. Temperature Dependence of $m_l(T)$ and $\chi(T)$ in the Strong-Interaction Limit

In the strong-interaction limit ($U/W \gg 1$), which is the analog of the split-band alloy, the electron self-energy [Eq. (2.6)] is given by

$$\Sigma_\sigma(E)=nU+n(1-n)U^2/[E-U(1-n)] . \qquad (3.10)$$

It can be seen from Eqs. (3.10) and (2.18) that the density of states contains two well-separated bands, one centered at 0 containing $1-n$ states and one centered at U containing n states. This density of states is illustrated for $n=\frac{1}{2}$ in the insert of Fig. 3. The temperature-dependent local magnetic moment is given by

$$m_l(T)=-2(1-n)\pi^{-1}\,\mathrm{Im}\int_{-\infty}^{\infty} dE\left[(1+e^{(E-\mu)/kT})^{-1} \times\frac{F(E-\Sigma_\sigma(E))}{1+\Sigma_\sigma(E)F(E-\Sigma_\sigma(E))}\right] . \qquad (3.11)$$

In the limit $kT \ll U$, it can be shown using either Eq. (3.11) or the energy-integrated form of Eq. (2.10) and observing that if $n<\frac{1}{2}$, μ lies in the lower band and if $n>\frac{1}{2}$, μ lies in the upper band, that

$$m_l(T)=2n, \qquad n<\tfrac{1}{2} \qquad (3.12)$$

and

$$m_l(T)=2(1-n), \quad n\geq\tfrac{1}{2} . \qquad (3.13)$$

In the atomic limit, for which the single-electron properties of Eq. (2.1) can be exactly solved, the alloy analogy has been shown to be exact.[1] Consequently, if $W=0$ and $kT \ll U$, Eqs. (3.12) and (3.13) are valid independent of any approximation. These two equations are physically reasonable and indicate that, if $U/W \gg 1$ and $kT \ll U$, the local magnetic moment has its maximum value, corresponding either to electrons if $n<\frac{1}{2}$ or holes if $n>\frac{1}{2}$. At temperatures high compared to U/k, where k is the Boltzmann constant the moment falls to the value appropraite to noninteracting electrons: $m_l=2n(1-n)$. These results are shown in Fig. 3. The temperature T_l indicated in Fig. 3, defined as in Sec. III A, may be seen to be in reasonable agreement with the "melting" temperature obtained by other authors[2,3] for $U/W \gg 1$. As in Fig. 2, because of the antiferromagnetic ordering at low temperatures for the case $n=\frac{1}{2}$, the figure is not strictly correct below the (small) Néel temperature $T_N \sim W^2/U$.[13]

It follows from Eqs. (3.10), (2.9), and (2.16) that, for $U/W \gg 1$, $kT \ll U$, and $n<\frac{1}{2}$, the static uniform susceptibility is given by

$$\chi(T)=\frac{2\mu_B^2\rho_\sigma^0[\mu/(1-n)]}{(1-2n)/(1-n)+\rho_\sigma^0[\mu/(1-n)][\mu/(1-n)]} . \qquad (3.14)$$

This result is identical to the expression for $\chi(T)$

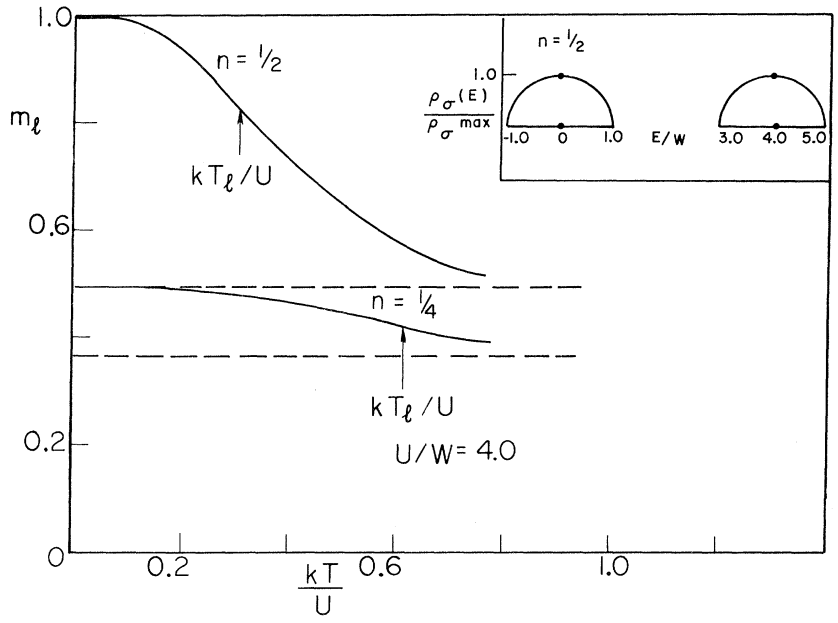

FIG. 3. Temperature dependence of m_l for $U/W=4.0$ and for $n=\frac{1}{2}$ and $n=\frac{1}{4}$. In the insert is plotted the density of states for interacting electrons.

in the strong-interaction limit obtained using equation-of-motion techniques.[4,14] It may be seen from Eq. (3.14) that for $kT \ll W$ the susceptibility does not obey a Curie-Weiss law. This result is felt to be incorrect and may reflect a weakness in the alloy-analogy approximation. However, when $U \gg kT \gg W$, Eq. (3.14) reduces to the expected Curie law, and we get

$$\chi(T) = 2\mu_B^2 n/kT \ . \tag{3.15}$$

Finally, when $kT \sim U$ the Curie-law behavior again breaks down, until $kT \gg U$ when χ is given by Eq. (2.18). These results can easily be extended to the case $n > \frac{1}{2}$. It follows from Eq. (3.14) that the Curie temperature T_C is obtained by solving the equation

$$1 - 2n + \rho_0^0[\mu(T_C)/(1-n)]\,\mu(T_C) = 0 \ . \tag{3.16}$$

The condition for paramagnetic instability has been previously discussed in detail.[14] It is clear that there will be a ferromagnetic phase transition if (a) μ is negative and (b) the density of states at the Fermi energy $\rho_\sigma^0[\mu/(1-n)]$ is sufficiently large. For symmetrical band shapes μ will be negative only if the lower band (centered at $E=0$) is less than half-full. At zero bandwidth it may be seen from Eq. (3.15) that the system is always paramagnetic.

IV. DISCUSSION AND CONCLUSIONS

The fact that the alloy-analogy scheme for the Hubbard Hamiltonian reproduces the usual expressions for the static uniform susceptibility in the three limiting cases considered above suggests that it is reasonable to apply this approach to the more general case of arbitrary U/W and n. In addition, the high- and low-temperature values of the local magnetic moment obtained in Sec. III appear to be physically reasonable.

It can be concluded from the discussion in the previous section that m_l does *not* undergo a phase transition to a zero value. There is instead a partial "melting" of the local-magnetic-moment magnitude, which at temperatures high compared to U and W approaches the *finite* value $2n(1-n)$. Furthermore, there is no structure in the spin susceptibility which can be related to a sudden disappearance of local magnetic moments. This is in contrast to previous results[2,3] which predict a sharp phase transition associated with a disappearance of the local magnetic moment. Because the alloy-analogy scheme is exact in the case $W=0$, in which limit the same conclusions hold, it is evident that these results are not a feature of the approximation used here.

The discrepancy between the results of the present and previous calculations[2,3] can be traced to a difference in the definition of the local moment, in the case of Ref. 2, and to the additional assumptions made in Ref. 3 about the local environment of each local moment. In Ref. 2, in which an antiferromagnetic system was considered, the time-independent-*vector* local moment on each of the two antiferromagnetic sublattices was calculated. This vector local moment was defined to be $(n_\uparrow - n_\downarrow)$ on one sublattice and $(n_\downarrow - n_\uparrow)$ on the other, where $n_\uparrow$ and $n_\downarrow$ represent, respectively, the *average* occupation probabilities of spin-$\uparrow$ and spin-$\downarrow$ electrons calculated within a particular antiferromagnetic sublattice. It is evident that the quantity $(n_\uparrow - n_\downarrow)$

will undergo a phase transition to zero at the Néel temperature.[12] However, this phase transition is clearly characteristic of the bulk rather than the local properties of the system and cannot be considered as distinct from the usual antiferromagnetic-paramagnetic phase transition associated with the disappearance of long-range order.

In Ref. 3 the partition function of the Hubbard Hamiltonian, Eq. (2.1), was calculated using the "static approximation" to the functional integral representation. By minimizing the approximate free energy, a time-independent Ising-vector local field ξ_0 was computed. However, no exact physical interpretation was ascribed to ξ_0. In a single-impurity *alloy* such as described by the Anderson Hamiltonian, this static local field is proportional to plus or minus the absolute value of the impurity spin $\pm|n_\uparrow^{\rm imp}-n_\downarrow^{\rm imp}|$, calculated from an unrestricted Hartree-Fock approximation to the impurity Green's function.[15] Consequently, it seems reasonable to associate ξ_0 in a *metal* with $\pm m_l\times$const, where m_l is the average local magnetic moment defined in Eq. (2.13). In addition to the static approximation a further approximation was made in Ref. 3. In determining ξ_0 at the site i the local fields at sites $i\neq j$ were set equal to 0. This one-center approximation reduces a calculation of the local moments in metals to that of the local moment of an impurity in a metal host. As a consequence of the one-center approximation, at finite temperatures ξ_0 was found to undergo a phase transition to zero similar to that found in very dilute alloys. However, because it has been shown here that the average moment magnitude m_l is nonzero at all temperatures for $0<n<1$, it can be concluded that the value obtained for ξ_0 from the one-center approximation is apparently incorrect.

It is evident that sufficient care must be taken in defining and computing the local magnetic moment. At finite temperatures the local-moment magnitude and direction fluctuates from site to site. The average of the local-moment magnitude $m_l(T)=\langle|n_{i\uparrow}-n_{i\downarrow}|\rangle_{\rm config\ av}$ at temperatures high compared to U and W approaches a finite value $2n(1-n)$. On the other hand, the average of the vector local magnetic moment $\langle n_{i\uparrow}-n_{i\downarrow}\rangle_{\rm config\ av}$ at a site is zero above the ordering temperature T_0. The apparent size of a local magnetic moment in metals is strongly dependent on the nature of the experiment which is performed to measure this quantity. A Mössbauer experiment, for example, is sensitive to the average of the *vector* local magnetic moment if the rate of change of its direction is fast compared to the nuclear precession frequency.[16] Otherwise it measures a distribution of the magnitudes of the local magnetic fields. On the other hand, the static uniform spin susceptibility $\chi(T)$ at high temperatures measures the average local-moment *magnitude*. At these temperatures $\chi(T)$ is described by a Curie law with Curie-Weiss constant $2n(1-n)=m_l(T\to\infty)$.

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[9]For notational convenience, when the site index i is omitted from the quantity n_σ, it is to be understood that n_σ is the average number of electrons per site of spin σ defined by Eq. (2.9). The quantity $n_{i\sigma}$ is defined to be $\langle\hat{C}_{i\sigma}^\dagger\hat{C}_{i\sigma}\rangle$, which denotes an expectation value of the number operator at the site i evaluated for a fixed configuration of spins at the N sites in the metal.

[10]It is clear that Eq. (2.13) is equivalent to $m_l(T)=N^{-1}\sum_i\langle(\hat{n}_{i\uparrow}-\hat{n}_{i\downarrow})^2\rangle$, where $\langle\cdots\rangle$ denotes an expectation value. It should be noted, however, that there is no unique definition of the local-magnetic-moment magnitude.

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