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(Received 12 July 1994)

We derive low-temperature properties of the large-U Hubbard model in two and three dimensions starting from exact series-expansion results for high temperatures. Convergence problems and limited available information prevent a direct or Padé-type extrapolation. We propose a method of extrapolation, which is restricted to large U and low hole densities, for which the problem can be mapped on that of a system of weakly interacting holes. In this formulation an extrapolation down to $T = 0$ can be obtained, but it can be trusted for the presently available series data for $\beta t \lesssim 20$ and for hole densities $n_h \lesssim 0.2$ only. Implications for the magnetic phase diagram are discussed.

I. INTRODUCTION

The single-band Hubbard model is presumably the simplest model for describing the behavior of correlated electrons in a solid. Examples of its applications are its initial use to describe magnetism in transition metals and, most recently, theories of high-temperature superconductivity. Unfortunately, while it seems likely that for the latter phenomenon more complex models are needed, even this simple model is not nearly well understood. For one dimension some rigorous results are known, but in higher dimensions the main results have been obtained from Monte Carlo and finite-lattice calculations only.

We are interested in deriving magnetic properties for the case of large U on a square or simple cubic lattice. A well-known theorem by Nagaoka[1] states that a Hubbard model on a bipartite lattice with one hole and at infinite U has a ferromagnetic ground state. Many authors have investigated whether this one point in the phase diagram is part of a whole region of ferromagnetic behavior. Various methods are being used for this purpose,[2] including exact diagonalization of small systems, Monte Carlo simulations, and mean-field and cluster expansion methods. Two of us[3] as well as various other authors[4–6] have used the last method to calculate high-temperature series expansions for the square and simple cubic lattices. Expressions have been obtained for various thermodynamical quantities, such as the free energy, the magnetization, the magnetic susceptibility, and also for the pair-correlation functions between the z components of the spin at specific sites. These expressions show very well-converged behavior for high temperatures ($kT/t \gtrsim 2$). The aim was to find indications for the onset to ferromagnetic behavior at low temperatures by extrapolating the results of the series expansions. Indeed, these indications can be found, as is shown in Refs. 3 and 6. However, predictions for the ground state, based on these results, are highly unreliable. Due to the fact that we only have five terms in the series expansions (zeroth-, second-, fourth-, sixth-, and eighth-order terms), we found it impossible to rely on standard extrapolation methods like Padé approximants. The obtained results for different extrapolations vary too much to be able to derive any reliable extrapolated value. Henderson *et al.*[6] tried to find an indication for the expected divergence in the uniform susceptibility

$$\chi_{\mathrm{FM}} = \beta \frac{\partial^2}{\partial(\beta h)^2} \ln \mathrm{tr}\, e^{-\beta \mathcal{H}} \Big|_{h=0} \tag{1}$$

by looking for zeros of χ_{FM}^{-1}. The character of the series expansion, shown for infinite U as a function of βt in subsequent approximations in Fig. 1, is such that $\chi_{\mathrm{FM}}^{-1}(\beta t)$ is likely to diverge very quickly for $\beta t > 1$, to plus and minus infinity alternatingly. This means that zeros are to be found for $\beta t \gtrsim 1$ in the fourth- and eighth-order approximations, but no zeros exist at second and sixth

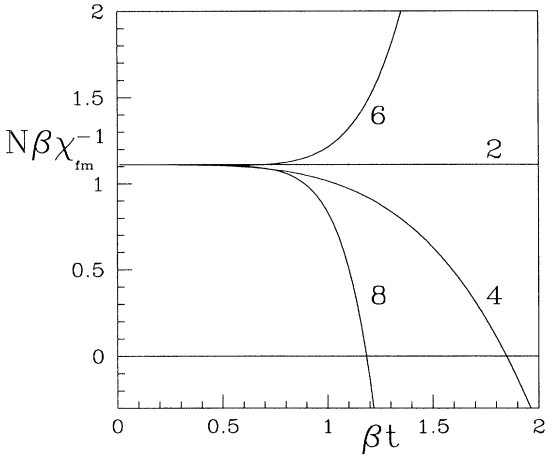

FIG. 1. The inverse ferromagnetic susceptibility as a function of the parameter βt, for the Hubbard model on a simple cubic lattice, with infinite U and particle density $n = 0.9$. Approximations up to order 2, 4, 6, 8 in βt obtained by means of the cluster expansion method.

orders. We feel that there is no reason to believe that the fourth- and eighth-order results should be more reliable than the others.

Furthermore, we have also constructed the antiferromagnetic susceptibility $\chi_{\rm AF}$ by including a staggered-field term in the Hamiltonian, and we have calculated its divergence in the same way as described above. In Fig. 2 we compare the Curie temperature T_C as a function of the particle density, for various values of t/U, to the Néel temperature T_N, for calculations up to eighth order [T_C and T_N are defined by $\chi_{\rm FM}^{-1}(n,U,T_C)=0$ and $\chi_{\rm AF}^{-1}(n,U,T_N)=0$, respectively]. This is an extension of the results presented by Pan and Wang,[5] who make the same comparison but only at fourth order. Qualitatively, our results are very similar to theirs: As the Néel temperature is higher than the Curie temperature for the parameters shown, one should conclude that the system goes into an antiferromagnetic state before the ferromagnetic transition is reached. However, regarding the character of the series expansion, as illustrated in Fig. 1, it is clear that the plots cannot be trusted qualitatively, let alone quantitatively.

In this paper we will consider a method that does not encounter these problems of extrapolation to low temperatures. In this method the density of holes is used as a small parameter. The high-temperature results are expressed in terms of an effective density of states for holes (as was done before by Brinkman and Rice[7]), and extended to interactions between hole levels. With this density of states, expressions for the free energy of the thermodynamic system can be obtained in the whole range of temperatures. In Sec. II we will define a partition function for the holes and express it in terms of an effective chemical potential for the holes. In Sec. III we derive the density of states for noninteracting holes, and we determine its moments, for infinite U. We present an improvement on the noninteracting hole picture in Sec. IV, where we consider interacting holes by introducing a Fermi-liquid-like interaction in energy space. In Sec. V we show how to use the density of states to calculate zeros of the inverse susceptibility. Section VI deals with the noninteracting hole approximation applied for finite U. In Sec. VII we show our conclusions for the magnetic phase diagram, and we discuss the method in Sec. VIII.

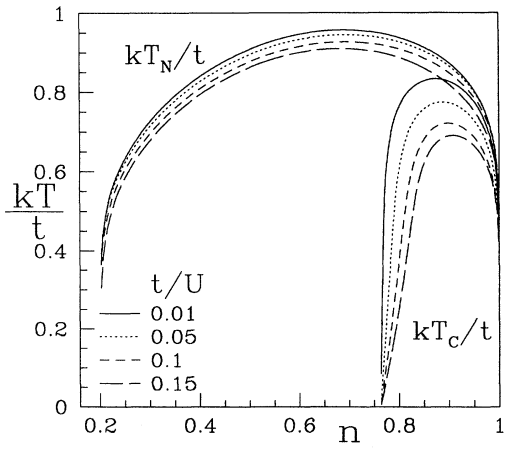

FIG. 2. Néel and Curie temperatures, as a function of the particle density, for the Hubbard model on a simple cubic lattice, at constant t/U.

II. HOLE FORMULATION

We consider the Hubbard Hamiltonian

$$\mathcal{H}=\mathcal{H}_{\rm kin}+\mathcal{H}_{\rm local}-\mu\sum_{i\sigma}n_{i\sigma}-h\sum_{i\sigma}\sigma n_{i\sigma}\ , \tag{2}$$

with

$$\mathcal{H}_{\rm kin}=-t\sum_{\langle i,j\rangle,\sigma}c_{i\sigma}^{\dagger}c_{j\sigma}\ , \tag{3}$$

$$\mathcal{H}_{\rm local}=U\sum_{i}n_{i\uparrow}n_{i\downarrow}\ , \tag{4}$$

where t is the hopping integral between nearest neighbors, U denotes the on-site interaction strength, μ is the chemical potential, and h is the strength of an external magnetic field. The operator $c_{i\sigma}^{\dagger}$ ($c_{i\sigma}$) creates (annihilates) a particle with spin σ at site i, and $n_{i\sigma}=c_{i\sigma}^{\dagger}c_{i\sigma}$ counts the number of particles with spin σ at site i.

To investigate the thermodynamic properties we want to calculate the grand canonical partition function

$$Z_{\rm gr}={\rm tr}\ e^{-\beta\mathcal{H}}\ . \tag{5}$$

For a system consisting of N sites we can rewrite this as

$$Z_{\rm gr}=\sum_{N_s=0}^{2N}e^{\beta\mu N_s}Z_{N_s}\ , \tag{6}$$

with Z_{N_s} the canonical partition function for $N_s=N_\uparrow+N_\downarrow$ particles:

$$Z_{N_s}=\sum_{N_\uparrow=0}^{N_s}e^{\beta h(N_\uparrow-N_\downarrow)}\sum_{j}e^{-\beta\varepsilon_j^{(N,N_s,N_\uparrow)}}\ . \tag{7}$$

Here $\{\varepsilon_j^{(N,N_s,N_\uparrow)}\}$ is the set of eigenvalues of $\mathcal{H}_{\rm kin}+\mathcal{H}_{\rm local}$ for $N_\uparrow$ up spins and $N_\downarrow$ down spins on N sites (note that the ε_j are functions of t and U only). Via the grand potential

$$\Omega=-\frac{1}{\beta}\ln Z_{\rm gr}\ , \tag{8}$$

we can now derive the other thermodynamic quantities by means of the usual manipulations, e.g., the particle density

$$n_s=\frac{\langle N_s\rangle}{N}=-\frac{1}{N}\frac{\partial\beta\Omega}{\partial\beta\mu} \tag{9}$$

or the susceptibility as given by (1).

In order to approach to lower temperatures in the limit of strong interactions and near half filling, we are going to express the partition function in terms of an effective chemical potential for holes. We associate the kinetic part of the Hamiltonian with the motion of the (dilute) holes, and its magnetic part with the background of spins. Thus, we have to divide out the spin degrees of freedom to obtain the canonical partition function for the holes:

$$Z^h_{N_h} \equiv Z_{N-N_h} e^{\beta \epsilon_{\rm HF}(N-N_h)} . \tag{10}$$

Here we define the number of holes,

$$N_h = N - N_s , \tag{11}$$

and we introduce a parameter $\epsilon_{\rm HF}$ which can be viewed as the free energy per spin in the absence of holes (i.e., at half filling; naturally, $Z^h_0 \equiv 1$):

$$\epsilon_{\rm HF} \equiv -\frac{1}{N\beta} \ln Z_N \tag{12}$$

$$= -\frac{1}{\beta} \ln(2 \cosh \beta h) \tag{13}$$

for infinite U. The grand canonical partition function for the holes then is

$$Z^h_{\rm gr} = Z_{\rm gr} e^{\beta(\epsilon_{\rm HF}-\mu)N} \tag{14}$$

$$= \sum_{N_h} Z^h_{N_h} e^{\beta(\epsilon_{\rm HF}-\mu)N_h} , \tag{15}$$

suggesting the definition of an effective chemical potential for the holes [cf. Eq. (6)]:

$$\mu_h \equiv \epsilon_{\rm HF} - \mu . \tag{16}$$

With this definition we can rewrite (14) as

$$\ln Z_{\rm gr} = -\beta \mu_h N + \ln Z^h_{\rm gr} . \tag{17}$$

Note that expression (13) for $\epsilon_{\rm HF}$ is exactly true only in the case of infinite U, as the interaction then prevents particles from occupying the same site. Note also that we do not define the number of holes as the number of sites where no particles are present (a definition which seems obvious), because the interpretation of Eqs. (10) and (15) would then become problematic for finite U. However, if U is very large, as we assume, a pair of electrons located on the same site causes a very high energy, and the contribution of the corresponding hole to the kinetic part of the Hamiltonian is some orders of magnitude smaller than the contribution of a "real" (nonremovable) hole. Therefore we will use (11) also in the case of large, finite U, and we will show that this leads to terms to be added to the expressions for infinite U of order $\frac{1}{U}$ or higher.

III. CONSTRUCTION OF THE DENSITY OF STATES FOR INFINITE U

Let us consider a system near half filling, with, for simplicity, infinitely strong coupling U (the case of finite U will be treated in Sec. VI). We assume that the system can be described in terms of the kinetic energy of noninteracting dilute holes and the magnetic energy of the background particles. We define the spectral distribution of the energy levels of one hole in an otherwise half-filled system, $\rho(\varepsilon, \beta h)$, in terms of the one-hole partition function Z^h_1, through

$$\frac{Z^h_1}{N} \equiv \int d\varepsilon \rho(\varepsilon, \beta h) e^{-\beta t \varepsilon} , \tag{18}$$

where we write $\beta t \varepsilon$ to make the integration parameter ε dimensionless. One can see this as a Laplace tranform, since Z^h_1 is a function of βt. We take ρ to be normalized to 1.

Although we said before that we divide out the magnetic degrees of freedom in the spin background, there is still a dependence of ρ on the magnetic field h. It is not easy to see how the hole motion depends on the field exactly, but one can easily understand why this dependence exists: A magnetic field influences the distribution of the spin background, which in turn determines the behavior of the hole. The hole motion depends on the field only indirectly, and the mechanism that governs the hole dynamics can in fact be much better described in terms of the average magnetization of the spin background than in terms of the field. It is important to understand that, in this picture, one has to treat the spin background as if it were at half filling, with the dilute holes subjected to its magnetization. Therefore we change variables at this level from βh to the magnetization per spin m. This change is easily performed by a Legendre transformation

$$\bar{\epsilon}_{\rm HF}(m) = \epsilon_{\rm HF}(\beta h) + mh , \tag{19}$$

yielding

$$\frac{Z^h_1}{N} \equiv \int d\varepsilon \bar{\rho}(\varepsilon, m) e^{-\beta t \varepsilon} , \tag{20}$$

where $\bar{\rho}(\varepsilon, m)$ is obtained from $\rho(\varepsilon, \beta h)$ via

$$m = -\frac{\partial \epsilon_{\rm HF}}{\partial h} , \tag{21}$$

which becomes $m = \tanh(\beta h)$ for infinite U.

With this definition we can write down a first approximation for the grand canonical partition function. A one-hole level can be occupied, with a Boltzmann weight $e^{-\beta t \varepsilon}$, or it can be unoccupied, in which case there is an electron in the system with Boltzmann weight $e^{-\beta \mu_h}$ (with the magnetic energy included in μ_h). Thus, in the case of noninteracting holes we have (dropping the m dependence of $\bar{\rho}$)

$$\ln Z_{\rm gr} = N \int d\varepsilon \bar{\rho}(\varepsilon) \ln(e^{-\beta t \varepsilon} + e^{-\beta \mu_h}) \tag{22}$$

or equivalently, using (17),

$$\ln Z^h_{\rm gr} = N \int d\varepsilon \bar{\rho}(\varepsilon) \ln\left(1 + e^{-\beta(t\varepsilon - \mu_h)}\right) . \tag{23}$$

This equation becomes exact in a one-dimensional sys-

tem, as in that case the holes cannot disturb the magnetic background of the particles, thus being really noninteracting, and also in a ferromagnetic system (at $m = \pm 1$), for similar reasons. In other, higher-dimensional systems (23) is only correct to first order in $e^{\beta\mu_h}$. We make an expansion of the right-hand side with respect to the small parameter $e^{\beta\mu_h}$ to obtain

$$\ln Z_{\rm gr}^h = N \int d\varepsilon \bar{\rho}(\varepsilon) \left(e^{\beta\mu_h - \beta t\varepsilon} - \frac{1}{2} e^{2\beta\mu_h - 2\beta t\varepsilon} + \cdots \right) . \tag{24}$$

Comparing this to a similar expansion of the logarithm of Eq. (15) we see that this is consistent with the definition of the density of states in the first-order term.

Now, as an illustration of the calculation, let us have a look at the form that Eq. (8) actually takes when evaluating it for this system by means of a cluster expansion method. In Ref. 3 a general formula is presented for finite U, which for infinite U reduces to

$$\frac{\ln Z_{\rm gr}}{N} = \ln Z_{{\rm gr},0} + \sum_{n=1}^{\infty} (\beta t)^{2n} \sum_{m=0}^{2n-1} \sum_{l=-m}^{m} \frac{\Omega_{m,l}^{(n)} e^{\beta(m\mu + lh)}}{(Z_{{\rm gr},0})^{2n}} . \tag{25}$$

Here $Z_{{\rm gr},0}$ denotes the partition function for a system consisting of only one site:

$$Z_{{\rm gr},0} = 1 + 2e^{\beta\mu} \cosh(\beta h) . \tag{26}$$

The $\Omega_{m,l}^{(n)}$ are coefficients, which can, e.g., be calculated by means of a cluster expansion method. By substituting μ_h for μ, using (16) and (13), and expanding in the small parameter $e^{\beta\mu_h}$, we can obtain an expression for the grand potential for the holes again, now in the form of a series expansion:

$$\frac{\ln Z_{\rm gr}^h}{N} = \sum_{N_h=1}^{\infty} e^{N_h \beta\mu_h} \sum_{n=0}^{\infty} (\beta t)^{2n} \Omega(N_h, n, h) , \tag{27}$$

where

$$\Omega(p, 0, h) = \frac{(-1)^{(p-1)}}{p} \tag{28}$$

and

$$\Omega(p, n, h) = \sum_{m=0}^{2n-1} \sum_{l=-m}^{m} \binom{p+m-1}{p+m-2n} \frac{(-1)^p \Omega_{m,l}^{(n)} e^{\beta l h}}{[-2\cosh(\beta h)]^m} \tag{29}$$

for $n \neq 0$. Finally, we obtain a relation between the coefficients $\Omega(1, n, h)$ and the moments of $\rho(\varepsilon)$ $[= \rho(\varepsilon, \beta h)]$ by expanding (18) in powers of βt:

$$\frac{Z_1^h}{N} = \sum_{n=0}^{\infty} (\beta t)^n \frac{(-1)^n}{n!} \int d\varepsilon \rho(\varepsilon) \varepsilon^n . \tag{30}$$

Thus, we see from Eq. (30) and the first-order term in (27) that we have

$$\int d\varepsilon \rho(\varepsilon) \varepsilon^{2n} = (2n)! \Omega(1, n, h) \tag{31}$$

for the even moments of ρ, all odd moments being zero. Although we have restricted ourselves to the case of infinite U here, this expression can easily be extended for the case of finite U, as we will see in Sec. VI. U then enters the equation as a parameter at the right-hand side.

For infinite U there is another, faster way to calculate these moments. They can then be expressed directly in the number of possible paths in state space for a system with one hole. This has been done first by Brinkman and Rice,[7] who calculated the first ten moments of the density of states for ferromagnetic, antiferromagnetic, and paramagnetic spin backgrounds on a simple cubic lattice. Yang *et al.*[8] have presented a large number of moments for the same spin backgrounds on two- to five-

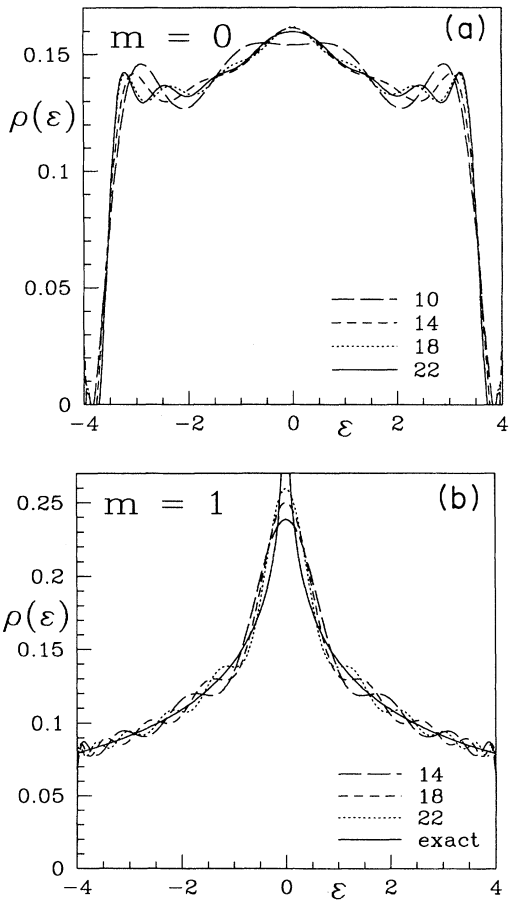

FIG. 3. (a) The density of states for a paramagnetic system on a square lattice, using up to the number of moments indicated. (b) The density of states in the ferromagnetic regime on a square lattice. Exact result, and approximations using up to the number of moments indicated.

TABLE I. Moments of $\bar{\rho}(\varepsilon)$ for $m = 0$ and $m = 1$, for the square and the simple cubic lattices (odd moments vanish).

Square lattice			Simple cubic lattice	
$m = 0$	$m = 1$	n	$m = 0$	$m = 1$
1	1	0	1	1
4	4	2	6	6
30	36	4	72	90
$269\frac{1}{2}$	400	6	$1072\frac{1}{2}$	1860
$2641\frac{3}{4}$	4900	8	$17781\frac{3}{4}$	44730
$27279\frac{15}{16}$	63504	10	$314403\frac{3}{16}$	1172556
$291718\frac{31}{32}$	853776	12	$5804323\frac{47}{64}$	32496156
$3199250\frac{73}{256}$	11778624	14	$110549185\frac{29}{64}$	936369720
$35766660\frac{57}{256}$	165636900	16	$2156004418\frac{11}{256}$	27770358330
$405989247\frac{577}{4096}$	2363904400	18		
$4665921461\frac{101}{16384}$	34134779536	20		
$54182396281\frac{55139}{65536}$	497634306624	22		

dimensional hypercubic lattices, including 18 moments for the square lattice and 14 for the simple cubic lattice. In Appendix A, we outline a method which enables us to enumerate the paths in an efficient way, and by which we have extended their results to 22 and 16 moments, respectively. These moments are presented in Table I for $m = 0$ and $m = 1$, corresponding to a paramagnetic and ferromagnetic system, respectively.

We now approximate $\bar{\rho}(\varepsilon)$ by a polynomial which we fit with the moments. In this way we calculate an approximation for the density of states, to different orders, in order to get an impression of the convergence of subsequent approximations. In Figs. 3(a) and 3(b) we show the result for a paramagnetic and a ferromagnetic system.

For $m = 1$ the exact density of states is known:

$$\bar{\rho}(\varepsilon) = \frac{1}{2\pi^2} K\left[1 - \left(\frac{\varepsilon}{4}\right)^2\right] , \tag{32}$$

with K the complete elliptic integral of the first kind. It has an integrable singularity at $\varepsilon = 0$. This is difficult to approximate and causes some oscillations away from $\varepsilon = 0$. Convergence towards the exact result is rather good. For $m = 0$ convergence is very good, as from 14th order on the difference between subsequent approximations becomes very small.

Meshkov and Berkov[9] fit the density of states by postulating that the integral of $\bar{\rho}^2$ be minimal ("smoothness" criterion), using a discretized $\bar{\rho}$. They claim that this method gives faster convergence than a polynomial fit. Comparing their results for the ferromagnetic density of states with the exact result and the results presented here, however, one may question that claim. We feel that the polynomial fits, when using an equal number of moments, give similar or even better results, which are also easier to handle in further calculations.

Before calculating various quantities which can tell us something about the low-temperature properties of the system, we will in the next section consider a method to improve the approximation of the density of states by including interactions between the holes.

IV. INTERACTING HOLES

The crucial question is to see for which domain of hole densities the assumption of independent holes is justified. This range can be determined from an estimate of the interactions between the holes. Very similar to the theory of the classical dilute gas,[10] the interaction can be deduced from the two-hole partition function as defined by (10) for $N_h = 2$. It is a matter of choice how to represent the hole interaction. One could think of a spatial representation, but one must realize that in this strongly quantal system the interaction is nonlocal, which complicates the transparency of the representation substantially. Having the one-hole system represented by a density of states it is natural here to choose an interaction between energy levels. First we formulate the interaction in terms of discrete levels and then we take the continuum limit as in Eq. (22). The discrete version of this expression can be written in terms of levels ε_i, distributed according to the density $\bar{\rho}(\varepsilon_i)$:

$$Z_{\mathrm{gr}}^{(1)} = e^{-\beta\mu_h N} \sum_{\{n_i\}} e^{-\beta\sum_i (t\varepsilon_i - \mu_h) n_i} , \tag{33}$$

where $\{n_i\}$ with $n_i = 0, 1$ is the occupation of the levels ε_i. We have given the expression a superindex 1 to indicate that $Z_{\mathrm{gr}}^{(1)}$ matches Z_{gr} up to the one-hole terms. The next approximation can be of the form

$$Z_{\rm gr}^{(2)} = e^{-\beta\mu_h N} \sum_{\{n_i\}} e^{-\beta[\sum_i (t\varepsilon_i - \mu_h)n_i + \sum_{(i,j)} f_{ij} n_i n_j]} , \quad (34)$$

where f_{ij} accounts for the interaction between the levels ε_i and ε_j. The second term in the exponent is a sum over all pairs of levels (i,j). In the energy space a distance between levels does not seem to be a measure for the strength of the interaction as in real space, where interactions usually decay sufficiently fast with the distance, such that the sum over pairs does not increase with the square of the number of elements, but only linearly as is necessary for a thermodynamic system. In order to make the exponent in (34) of the correct thermodynamic behavior the interaction should therefore decrease with the size of the system as

$$f_{ij} = \frac{t}{N}\phi_{ij} , \quad (35)$$

with ϕ_{ij} of order unity. An additional advantage of (35) is the fact that interactions of this type can be handled rigorously in the thermodynamic limit by the mean-field theory.[11] Thus we can write

$$\ln Z_{\rm gr}^{(2)} = -\beta\mu_h N + \sum_i \ln(1 + e^{-\beta\tilde{\varepsilon}_i}) + \frac{\beta t}{N} \sum_{(i,j)} \phi_{ij} n(\tilde{\varepsilon}_i) n(\tilde{\varepsilon}_j) , \quad (36)$$

where the $\tilde{\varepsilon}_i$ are the shifted energy levels,

$$\tilde{\varepsilon}_i = t\varepsilon_i - \mu_h + \frac{t}{N} \sum_{j \neq i} \phi_{ij} n(\tilde{\varepsilon}_j) , \quad (37)$$

and $n(\tilde{\varepsilon})$ is the Fermi occupation number,

$$n(\tilde{\varepsilon}) = \frac{1}{1 + e^{\beta\tilde{\varepsilon}}} . \quad (38)$$

Now the interaction ϕ_{ij} must be chosen such that $Z_{\rm gr}^{(2)}$ produces the correct two-hole partition function. Expanding Eq. (34) with respect to the number of holes,

$$N_h = \sum_i n_i , \quad (39)$$

and using Eq. (14) we find

$$Z_2^h = \sum_{(i,j)} e^{-\beta t(\varepsilon_i + t\varepsilon_j + \frac{1}{N}\phi_{ij})} . \quad (40)$$

In our high-temperature expansion we have no direct information on Z_2^h, but we have the coefficient of the second-order term in the hole expansion of $\ln Z_{\rm gr}^h$ [cf. (15)], which is

$$U_2 = Z_2^h - \frac{1}{2}(Z_1^h)^2 . \quad (41)$$

Note that this expression is of order N, and not of order N^2 as are both terms on its right-hand side. Using (40) and the corresponding expression for Z_1^h we may equate

$$U_2 = \sum_{(i,j)} e^{-\beta t(\varepsilon_i + \varepsilon_j + \frac{1}{N}\phi_{ij})} - \frac{1}{2} \sum_{i,j} e^{-\beta t(\varepsilon_i + \varepsilon_j)} . \quad (42)$$

For $N \to \infty$ we may write

$$U_2 = -\frac{\beta t}{N} \sum_{(i,j)} e^{-\beta t(\varepsilon_i + \varepsilon_j)} \phi_{ij} - \frac{1}{2} \sum_i e^{-2\beta t\varepsilon_i} , \quad (43)$$

and we see that U_2 is indeed of order N by virtue of (35). Note that the terms $i = j$ in the second term of (42) are not compensated by the first term. The second term in (43) gives the ideal-gas term of the hole system on the two-hole level.

Since we have moments of U_2 by our high-temperature expansions, and also the last term in (43) is known from our one-hole density of states, it is convenient to split U_2 into an interacting and an ideal part:

$$U_2 = U_2^{\rm int} + U_2^{\rm id} , \quad (44)$$

with

$$U_2^{\rm id} = -\frac{1}{2} \sum_i e^{-2\beta t\varepsilon_i} = -\frac{N}{2} \int d\varepsilon \bar{\rho}(\varepsilon) e^{-2\beta t\varepsilon} \quad (45)$$

[cf. (24)]. ϕ_{ij} must then be determined from $U_2^{\rm int}$. In a continuous version the equation for $\phi(\varepsilon, \varepsilon')$ becomes

$$U_2^{\rm int} = -\frac{\beta t}{2} N \int d\varepsilon \bar{\rho}(\varepsilon) \int d\varepsilon' \bar{\rho}(\varepsilon') e^{-\beta t(\varepsilon + \varepsilon')} \phi(\varepsilon, \varepsilon') . \quad (46)$$

This relation is not strong enough to yield a unique $\phi(\varepsilon, \varepsilon')$, in the same way as the second virial coefficient of a classical gas is not sufficient to determine the interaction potential. The freedom in choice will be reflected upon the efficiency of the program to determine the higher-order interactions. We have chosen to have the dependence of $\phi(\varepsilon, \varepsilon')$ only on the sum variable $\varepsilon + \varepsilon'$, and we approximate it by a polynomial:

$$\phi(\varepsilon, \varepsilon') = \sum_l \phi_l (\varepsilon + \varepsilon')^l . \quad (47)$$

Equating moments in (46) and in

$$U_2^{\rm int} = \sum_k (U_2^{\rm int})_k (\beta t)^k , \quad (48)$$

we have

$$(U_2^{\rm int})_k = \sum_l \frac{(-1)^k N}{2(k-1)!} \int d\varepsilon \bar{\rho}(\varepsilon) \int d\varepsilon' \bar{\rho}(\varepsilon') (\varepsilon + \varepsilon')^{k-1+l} \phi_l . \quad (49)$$

Because we are working on a bipartite lattice, all odd moments of $U_2^{\rm int}$ are identically zero. Hence k is even, and as also $\bar{\rho}(\varepsilon)$ has only even moments, the combination $k-1+l$ must be even and therefore the sums in Eqs. (47) and (49) contain only odd l. The set of equations (49) for a finite number of the moments $(U_2^{\rm int})_k$ determines

TABLE II. Values of $\frac{1}{N}\left(U_2^{\rm int}\right)_k$ for $m=0$ and $m=1$, for the square lattice with $U=\infty$.

k	$m=0$	$m=1$
0	0	0
2	0	0
4	$-\frac{1}{3}$	0
6	$-\frac{47}{80}$	0
8	$-\frac{1713}{4032}$	0
10	$-\frac{989681}{5806080}$	0
12	$-\frac{160327813}{3832012800}$	0

an equal number of coefficients ϕ_l. We have computed $\left(U_2^{\rm int}\right)_k$ for the square lattice at $U=\infty$, up to $k=12$. This involves six terms ($k=2,4,\ldots,12$) and so we can determine six values $\phi_1,\phi_3,\ldots,\phi_{11}$. In the equations we thus need $k-1+l=12-1+11=22$ as the highest moment of $\bar{\rho}(\varepsilon)$, which is just the number of moments we have determined. The values of $\frac{1}{N}\left(U_2^{\rm int}\right)_k$ are given in Table II. For the ferromagnetic system ($m=1$) these coefficients are zero, as the holes do not interact in that case.

Finally we give the continuum form of the expressions (36) and (37) for the grand potential:

$$\ln Z_{\rm gr}^{(2)} = -\beta\mu_h N + N\int d\varepsilon\bar{\rho}(\varepsilon)\ln(1+e^{-\beta\tilde{\varepsilon}}) + \frac{\beta t}{2}N\int d\varepsilon\int d\varepsilon'\bar{\rho}(\varepsilon)n(\tilde{\varepsilon})\bar{\rho}(\varepsilon')n(\tilde{\varepsilon}')\phi(\varepsilon,\varepsilon') , \tag{50}$$

with

$$\tilde{\varepsilon} = t\varepsilon - \mu_h + t\int d\varepsilon'\bar{\rho}(\varepsilon')\phi(\varepsilon,\varepsilon')n(\tilde{\varepsilon}') . \tag{51}$$

V. INVERSE SUSCEPTIBILITY

We return to the uniform susceptibility

$$\chi_{\rm FM} = \left.\frac{\partial M}{\partial h}\right|_{h=0} , \tag{52}$$

with M the total magnetization of the system. As before, we try to find indications of divergences of $\chi_{\rm FM}$, which should be related to second-order phase transitions between a paramagnetic and a ferromagnetic state. It is usually more convenient to express this by stating that the inverse susceptibility must be zero,

$$\chi_{\rm FM}^{-1} = 0 , \tag{53}$$

and to study

$$\chi_{\rm FM}^{-1} = \left.\frac{\partial h}{\partial M}\right|_{M=0} \tag{54}$$

or

$$\beta N\chi_{\rm FM}^{-1} = \left.\frac{\partial\beta h}{n_s\partial m}\right|_{m=0} , \tag{55}$$

where m is the magnetization per spin as defined in Sec. III. In order to find an expression for h, to be able to calculate (55), we construct a generalized (Landau like) free energy

$$\varphi(n_s,\mu,m,h) = -\frac{1}{\beta N}\ln Z_{\rm gr} + \mu n_s - hmn_s , \tag{56}$$

where $\ln Z_{\rm gr}$ is given by (22). φ has to be minimized with respect to μ and m at fixed particle density n_s and field h, to obtain the free energy. Note that this h is *not* the same field as we used before in Sec. III. There we interpreted h as a field that is felt only by the spins in the background, whereas now we obtain the physical external field that would be necessary to yield the given magnetization. Of course, in the case of a finite number of holes (the limit of half filling), these fields are the same, as we will see in the resulting expressions. Note also that, due to the definition of m as the magnetization per spin, its conjugated variable is hn_s, not h.

We can rewrite (56) using (16) and (19):

$$\beta\varphi = \beta\bar{\epsilon}_{\rm HF}n_s + \beta\mu_h n_h - \frac{1}{N}\ln Z_{\rm gr}^h , \tag{57}$$

where we can interpret the first term as the contribution of the background of spins, and the other terms as the contribution of the holes. Minimization leads to the following equations:

$$n_h = \int d\varepsilon\bar{\rho}(\varepsilon)\frac{1}{1+e^{\beta(t\varepsilon-\mu_h)}} , \tag{58}$$

$$\beta h = \beta h_{\rm HF} - \int d\varepsilon\frac{\partial\bar{\rho}(\varepsilon)}{n_s\partial m}\ln\left(1+e^{-\beta(t\varepsilon-\mu_h)}\right) , \tag{59}$$

with

$$h_{\rm HF} = \frac{\partial\bar{\epsilon}_{\rm HF}}{\partial m} . \tag{60}$$

The expression for the inverse uniform susceptibility (55) then becomes

$$\beta N\chi_{\rm FM}^{-1} = \left.\frac{\partial\beta h_{\rm HF}}{n_s\partial m}\right|_{m=0} - \int d\varepsilon\left.\frac{\partial^2\bar{\rho}(\varepsilon)}{n_s^2\partial m^2}\right|_{m=0}\ln\left(1+e^{-\beta(t\varepsilon-\mu_h)}\right) . \tag{61}$$

This can be rewritten in terms of $\rho(\varepsilon)$, using the Legendre transform (19) [thus $\bar{\rho}(\varepsilon,m)=\rho(\varepsilon,\beta h_{\rm HF})$]:

$$\beta N\chi_{\rm FM}^{-1} = \left.\frac{\partial\beta h_{\rm HF}}{n_s\partial m}\right|_{m=0}\left(1-\left.\frac{\partial\beta h_{\rm HF}}{n_s\partial m}\right|_{m=0}\int d\varepsilon\left.\frac{\partial^2\rho(\varepsilon,\beta h_{\rm HF})}{\partial(\beta h_{\rm HF})^2}\right|_{h_{\rm HF}=0}\ln\left(1+e^{-\beta(t\varepsilon-\mu_h)}\right)\right) . \tag{62}$$

Note that $m = 0$ is equivalent to $h_{\rm HF} = 0$, and that, for reasons of symmetry, the first derivative of ρ with respect to $h_{\rm HF}$ vanishes at $h_{\rm HF} = 0$.

According to (53) we want to find values of n_h and βt for which the right-hand side of (62) is zero, with n_h fixed by Eq. (58). One can easily verify that, for infinite U, we have $\beta h_{\rm HF}(m) = \arctan(m)$, and so putting (62) to zero gives

$$\int d\varepsilon \left.\frac{\partial^2 \rho(\varepsilon, \beta h_{\rm HF})}{\partial (\beta h_{\rm HF})^2}\right|_{h_{\rm HF}=0} \ln\left(1 + e^{-\beta(t\varepsilon - \mu_h)}\right) = 1 - n_h \ . \tag{63}$$

This equation can be solved by an iterative procedure to calculate the value of μ_h for a given value of βt. The density of states $\rho(\varepsilon)$, necessary to calculate n_h according to (58), is determined from its moments as described in Sec. III, and its second derivative is calculated in a similar way.

To include the interaction described in Sec. IV, one should use the grand potential as given in (50) rather than the noninteracting-hole approximation of (22). The final equation, equivalent to (62), then involves one extra term which contains the second derivative with respect to $\beta h_{\rm HF}$ of the interaction ϕ. We give a derivation of this equation in Appendix B.

In Fig. 4 we show Curie temperatures for the square-lattice Hubbard model at infinite U, in three different approximations: (a) the noninteracting-hole approximation, with ρ determined by interpolation from 8 of its moments (of which 4 moments are nonzero); (b) the same but with ρ determined from 22 (11 nonzero) moments; and (c) the interacting-hole approximation, with ρ determined from 22 moments and ϕ from 12 (5 nonzero) interaction coefficients.

One can see that the difference between the 8th- and the 22nd-order noninteracting approximations is small. In both approximations, ferromagnetism is stable against paramagnetism for $n_h \lesssim 0.27$, at low T. The interaction does not change this picture very much. It slightly enhances the stability of the ferromagnetic state, up to $n_h \lesssim 0.29$. The difference between the noninteracting and the interacting approximations becomes larger with increasing hole density, as expected. Numerically, the results agree very well for $n_h \lesssim 0.06$.

In the next section we will treat the case of finite U. We have been able to calculate eight moments of the density of states in that case; thus we can do an eighth-order approximation at the most. One can then calculate merely two coefficients ϕ_l of the interaction, resulting in an approximation of the interaction which is rather crude. We have seen that the picture in the noninteracting-hole approximation is qualitatively the same as the one in the interacting-hole approximation, in eighth order already. For small n_h it agrees rather well also numerically. Therefore, we will not include the interaction in the following calculations.

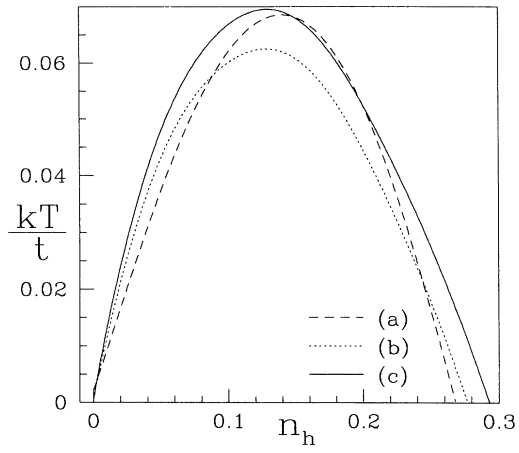

FIG. 4. Curie temperatures (contours of zero inverse ferromagnetic susceptibility) for the square lattice at infinite U. (a) Noninteracting-hole approximation, 8th order; (b) noninteracting-hole approximation, 22nd order; (c) interacting-hole approximation, 22nd order.

VI. NONINTERACTING-HOLE APPROXIMATION FOR FINITE U

As we pointed out before, at finite U, excitations in the spin background become possible due to the creation of pairs of electrons with opposite spin at the same site. This means that extra empty sites are created, and thus the number of empty sites in the system is no longer fixed. Taking U large, however, we can consider the contributions to the partition function due to these excitations to be small corrections of the infinite-U system, and we can neglect the terms that would arise from permanently present electron pairs. To do this, we consider the grand potential of the Hubbard model on a square lattice up to the second-order term (taken from Ref. 3; note that h is the parameter in the Hamiltonian here, not the physical magnetic field we discussed in the previous section):

$$-\frac{\beta\Omega}{N} = \ln\left[1 + 2e^{\beta\mu}\cosh(\beta h) + e^{2\beta\mu-\beta U}\right] + (\beta t)^2 \frac{4e^{\beta\mu}(1 + e^{2\beta\mu-\beta U})\cosh(\beta h) + \frac{8}{\beta U}e^{2\beta\mu}(1 - e^{-\beta U})}{\left[1 + 2e^{\beta\mu}\cosh(\beta h) + e^{2\beta\mu-\beta U}\right]^2} + \cdots \ . \tag{64}$$

In this expression, we will neglect the terms that contain the exponential of $-\beta U$, but we keep terms that are proportional to a power of $1/U$. This precisely distinguishes the terms that are due to permanent electron pairs, which cause an energy βU, from those due to temporary excitations in a system where otherwise no double occupancies are present. It is necessary to make this approximation, as the exponential terms cannot be treated in this method.

However, it can be seen easily that these terms are always exponentially smaller than other terms in the expansion, and thus that this approximation is justified.

First we consider the case of half filling, where we have $\mu = U/2$:

$$-\frac{\beta\Omega_{\mathrm{HF}}}{N} = \ln[2 + 2e^{\beta U/2}\cosh(\beta h)] + (\beta t)^2 \frac{8e^{\beta U/2}\cosh(\beta h) + \frac{8}{\beta U}(e^{\beta U} - 1)}{\left[2 + 2e^{\beta U/2}\cosh(\beta h)\right]^2} + \cdots \quad . \tag{65}$$

Here we can neglect all but the most important terms at large U; i.e., we only take the terms containing the highest power of $e^{\beta U}$, to get

$$-\frac{\beta\Omega_{\mathrm{HF}}}{N} = \frac{\beta U}{2} + \ln\left[2\cosh(\beta h)\right] + (\beta t)^2 \frac{2}{(\beta U)\left[\cosh(\beta h)\right]^2} + \cdots \quad . \tag{66}$$

By definition, this expression must be equal to $\frac{\beta U}{2} + \frac{1}{N}\ln Z_N$, and so using the definition (12) for ϵ_{HF} we get

$$-\beta\epsilon_{\mathrm{HF}} = \ln\left[2\cosh(\beta h)\right] + (\beta t)^2 \frac{2}{(\beta U)\left[\cosh(\beta h)\right]^2} + \cdots \quad , \tag{67}$$

and we see that this is indeed a correction of order $\frac{1}{U}$ in Eq. (13). Note that we obtain the same result if we first omit the $e^{-\beta U}$ terms in (64), and only then substitute $U/2$ for μ. This once more supports our statement that these terms may be neglected.

Off half filling, we have to rewrite (64) (without the $e^{-\beta U}$ terms) in terms of the effective chemical potential μ_h for the holes, as defined by Eq. (16), but now containing the corrected ϵ_{HF} as given by (67). For simplicity, we do this in a few steps. First, we substitute the chemical potential for the holes without the correction terms, as in Sec. III. Then we expand the logarithm and the numerators with respect to the exponential of this chemical potential. Finally, we include the corrected μ_h by expanding the exponentials with respect to the correction terms. Thus, we obtain for the grand potential

$$-\frac{\beta\Omega}{N} = -\beta\mu_h + e^{\beta\mu_h}\left(1 + (\beta t)^2\left[2 - \frac{2}{(\beta U)\left[\cosh(\beta h)\right]^2}\right] + \cdots\right) + \cdots \quad . \tag{68}$$

The coefficient of $e^{\beta\mu_h}$ in this expression again determines the moments of the distribution $\rho(\varepsilon, \beta h)$, as described in Sec. III. Of course these are now functions of βU. In Table III we give the moments that we have been able to derive

TABLE III. Moments of the density of states for the square and simple cubic lattices (odd moments vanish), for large but finite U, at $h = 0$.

n	Square lattice
0	1
2	$2\left(2 - \frac{2}{\beta U}\right)$
4	$24\left(\frac{5}{4} - \frac{2}{\beta U} - \frac{12}{(\beta U)^2} + \frac{3}{(\beta U)^3}\right)$
6	$720\left(\frac{539}{1440} - \frac{59}{48\beta U} - \frac{93}{8(\beta U)^2} - \frac{89}{6(\beta U)^3} + \frac{1}{2(\beta U)^4} + \frac{127}{2(\beta U)^5}\right)$
8	$40320\left(\frac{10567}{161280} - \frac{271}{576\beta U} - \frac{1459}{320(\beta U)^2} - \frac{377}{32(\beta U)^3} + \frac{4531}{96(\beta U)^4} + \frac{4043}{8(\beta U)^5} - \frac{28837}{8(\beta U)^6} + \frac{78593}{8(\beta U)^7}\right)$

n	Simple cubic lattice
0	1
2	$2\left(3 - \frac{3}{\beta U}\right)$
4	$24\left(3 - \frac{9}{2\beta U} - \frac{27}{(\beta U)^2} + \frac{3}{(\beta U)^3}\right)$
6	$720\left(\frac{143}{96} - \frac{67}{16\beta U} - \frac{315}{8(\beta U)^2} - \frac{71}{2(\beta U)^3} - \frac{633}{2(\beta U)^4} + \frac{825}{2(\beta U)^5}\right)$
8	$40320\left(\frac{1129}{2560} - \frac{869}{320\beta U} - \frac{7407}{320(\beta U)^2} - \frac{249}{32(\beta U)^3} - \frac{10551}{32(\beta U)^4} + \frac{13725}{8(\beta U)^5} - \frac{215739}{8(\beta U)^6} + \frac{279837}{4(\beta U)^7}\right)$

from the series-expansion data, for the square lattice, at $h = 0$. Note that the moments for $h = \infty$ are the same as in the case of infinite U (Table I), because U has no significance in a system where all spins point in the same direction.

We can now apply the method described in Sec. V to calculate Curie temperatures for finite U. One has to realize, though, that at half filling the inverse susceptibility depends on the temperature, which was not the case for infinite U. Due to the excitations we get corrections of the type $\beta t^2/U$; thus we still have a series expansion in the parameter βt. The coefficients in this expansion are suppressed by large factors βU, however, and the range of convergence of the expansion is $\beta t \lesssim 30$ or further, depending on the value of βU. Thus, we may hope that convergence is good enough in the region where we expect to find solutions of (53). We give the full expression for the inverse susceptibility at half filling, for the square lattice and up to the $(\beta t)^8$ terms:

$$\beta N \chi_{\mathrm{HF}}^{-1} = \left.\frac{\partial \beta h_{\mathrm{HF}}}{\partial m}\right|_{m=0} = 1 + \frac{4(\beta t)^2}{(\beta U)} + \frac{8\,(-2+\beta U)\,(\beta t)^4}{(\beta U)^3} + \frac{\left[1131 - 648\beta U + 32(\beta U)^2\right](\beta t)^6}{3(\beta U)^5} + \frac{\left[-9129 + 6296\beta U - 1132(\beta U)^2 + 4(\beta U)^3\right](\beta t)^8}{(\beta U)^7} \,. \tag{69}$$

We have checked that (69) does not become zero for any value of βt and βU. Therefore we expect no transition from a paramagnetic to a ferromagnetic state in the half-filled system. Thus, we only have to consider the second factor on the right-hand side of (62), which vanishes at

$$\int d\varepsilon \left.\frac{\partial^2 \rho(\varepsilon, \beta h)}{\partial(\beta h)^2}\right|_{h=0} \ln\left(1 + e^{-\beta(t\varepsilon - \mu_h)}\right) = (1 - n_h)\frac{\chi_{\mathrm{HF}}}{\beta N} \,. \tag{70}$$

We show the results for the square and the simple cubic lattices in the next section.

VII. MAGNETIC PHASE DIAGRAM

We have used the theory described above to calculate Curie temperatures for the square and simple cubic lattices. For both lattices, we find a surface of Curie temperatures in the n_h–$\frac{t}{U}$–T diagram. In Figs. 5 and 6 we display these results in various ways.

In Figs. 5(a) and 6(a), contours of fixed Curie temperature are plotted in the n_h–$\frac{t}{U}$ plane. In the range of temperatures up to about $\frac{kT}{t} = 0.20$ we find a curve enclosing a region of ferromagnetism. For $\frac{kT}{t} \gtrsim 0.07$ these curves are closed and lie away from the $\frac{t}{U} = 0$ axis. Thus, at given density n_h and temperature T_C, one has to go to finite U to find a transition. In other words, allowing for excitations in the spin background enhances the ferromagnetic behavior. Furthermore, curves are generally not enclosed by all contours at lower temperatures. This would imply that, at given n_h and $\frac{t}{U}$, one would find a paramagnetic-ferromagnetic transition when lowering the temperature from a region of high temperature, but also when letting it increase from zero. This reentering of a paramagnetic phase at low temperatures does not seem to be physical. It is in fact an artifact of this method, due to convergence problems at very low temperatures. One can understand this by looking at the expression (69). If the highest-order term becomes of order 1, the series is clearly too short and does not converge properly. This means that the results become unreliable for $\frac{t}{U} \gtrsim \frac{kT}{2t}$ in the case of the square lattice, and $\frac{t}{U} \gtrsim \frac{kT}{4t}$ on the simple cubic lattice. For a few curves we have indicated this by a dashed line. As the approximations are better for

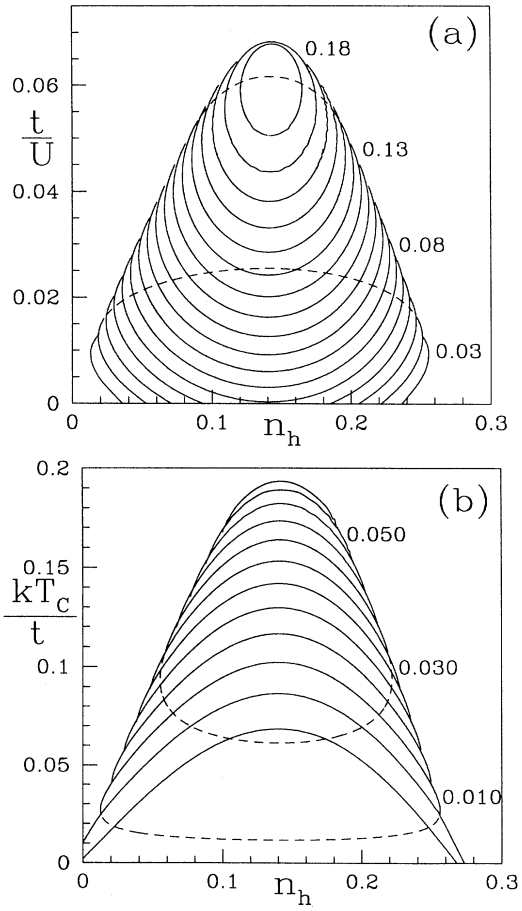

FIG. 5. Magnetic phase diagram for the square lattice. (a) Contours of fixed Curie temperature, with $kT_C/t = 0.03, 0.04, \ldots, 0.19$ (increment 0.01). (b) Curie temperature at fixed $t/U = 0, 0.005, \ldots, 0.055$ (increment 0.005).

higher temperatures, we assume that the actual curve at $T_C = 0$ (for which we can only perform a calculation at infinite U) should enclose all curves shown.

In Figs. 5(b) and 6(b), we show Curie temperatures in contours of fixed $\frac{t}{U}$. Again we see the nonphysical behavior of curves being closed at the low-temperature side, for almost all values of $\frac{t}{U}$. Figure 7 shows Curie temperatures at fixed $n_h = 0.09$, for the simple cubic lattice, indicated by the dot-dashed lines in Figs. 6(a) and 6(b). The dotted line in Fig. 7 indicates the region where the series expansion becomes unreliable, according to the arguments presented above.

There is one other point we want to mention here. As we have stated in the Introduction, we have also constructed the staggered susceptibility by replacing the magnetic field h by a staggered field h_s. Although it is much more complicated to calculate the high-temperature expansions for that case, as the number

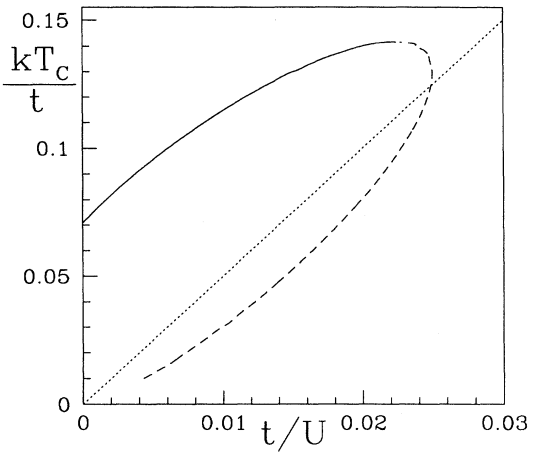

FIG. 7. Curie temperature for the simple cubic lattice, at $n_h = 0.09$. The dashed part of the curve is unreliable, due to lack of convergence (indicated by the dotted line).

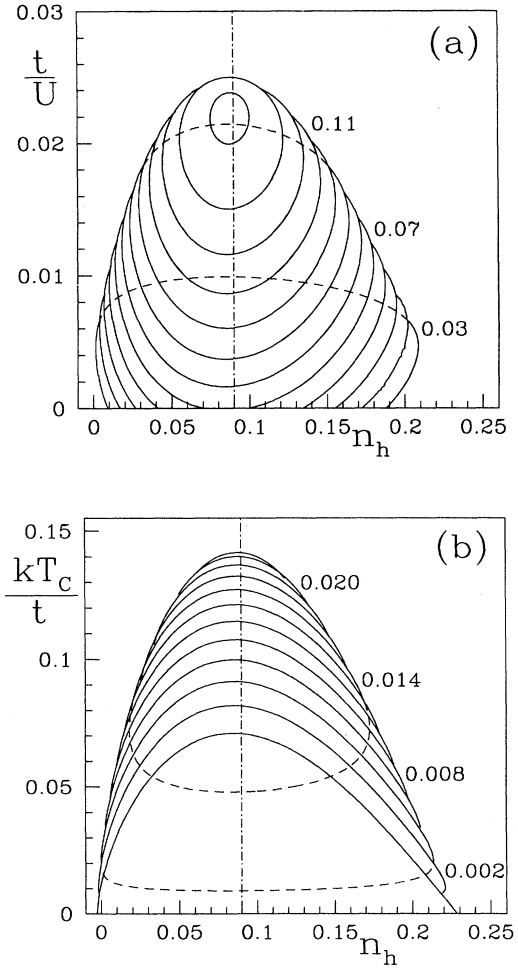

FIG. 6. Phase diagram for the simple cubic lattice. (a) Contours of fixed Curie temperature, with $kT_C/t = 0.03, 0.04, \ldots, 0.14$ (increment 0.01). (b) Curie temperature at fixed $t/U = 0, 0.002, \ldots, 0.022$ (increment 0.002).

of terms involved increases significantly, it is not difficult to obtain expressions for the staggered susceptibility, both at half filling and in the one-hole approximation, for $h_s = 0$. Thus, one may think that it is possible to obtain similar results for the transition between a paramagnetic and an antiferromagnetic state, and conclude which transition occurs first. When putting the inverse staggered susceptibility at half filling [the equivalent of (69) for the antiferromagnetic system] to zero, one finds solutions for all values of the parameter βU. This means that the staggered susceptibility of the half-filled system diverges at a finite temperature. Apparently, the paramagnetic-antiferromagnetic transition is driven by the background itself, and may be disturbed by a finite hole density. In our formulation, however, it is the holes that drive the

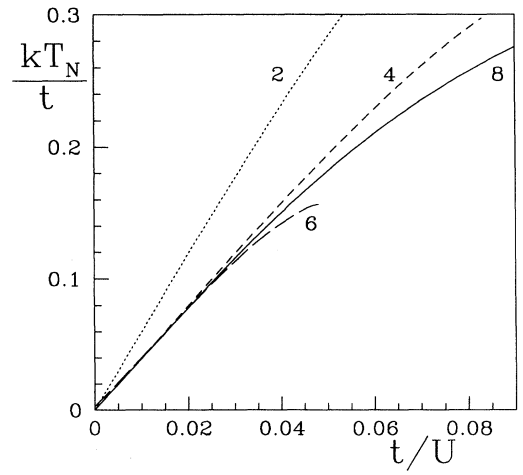

FIG. 8. Néel temperature for the simple cubic lattice at half filling. Approximations to different orders in βt, as indicated.

system into an ordered state, and the background only indirectly contributes to the transition via its interaction with the holes. This formulation is clearly not suitable to describe the transition to an antiferromagnetic state. Therefore we only briefly indicate what we expect for the paramagnetic-antiferromagnetic transition.

In Fig. 8 we plot Néel temperatures for the simple cubic lattice at half filling, in approximations to different orders in the parameter βt. We see that the convergence of the series expansion is very good for large U. A transition from a paramagnetic to an antiferromagnetic phase is expected for all values of U. It is at $T_N = 0$ for infinite U, and at increasing temperatures with decreasing U. For finite hole densities we expect the transition to occur at lower temperatures, and at some point cross the paramagnetic-ferromagnetic transition.

VIII. DISCUSSION AND CONCLUSIONS

We have calculated Curie temperatures for the large-U Hubbard model on the square and simple cubic lattices, by means of an extrapolation method to extract information on low-temperature behavior from high-temperature series expansions. We find a region of ferromagnetic behavior in the magnetic phase diagram, near half filling.

Comparing previous results for the simple cubic lattice, as depicted in Fig. 2, to our current results, shown in Fig. 6, we see that we now find a Curie temperature that is an order of magnitude smaller than before. Furthermore, as we have checked in the case of infinite U, subsequent approximations in the current method do give consistent results, instead of alternatingly producing Curie temperatures or not. These convergence problems in the primitive series expansions are likely due to the Fermi degeneracy of the electron gas. At $\beta t \approx 1$, the wavelength of the electrons becomes equal to the lattice distance, causing this degeneracy and divergences to be present. When applying a straightforward extrapolation technique, one cannot account for this degeneracy, leading to results that are erroneous for $\beta t \gtrsim 1$. In our approximation, using a density of states for holes, we take the Fermi degeneracy into account, and therefore we are able to proceed to lower temperatures. We are confident that our present results do not suffer from the above-mentioned convergence problems.

As we show in Fig. 4, the difference between approximations to different orders in the parameter βt is rather small, and adding the interaction also does not change the result considerably. Thus we believe the eighth-order noninteracting-hole approximation to be sufficient to describe the qualitative behavior, and to obtain a good indication for numerical values. We may add that, as a check, we have compared the free energy from calculations by this method to results following directly from the series expansions, at $\beta t \lesssim 0.5$, where the expansions are almost exact, and that these results agree very well.

Our method works only for large U, low hole density ($n_h \lesssim 0.2$), and, depending on the value of U, sufficiently high temperature. This is clear from Figs. 5–7, where we see that the results are unreliable for $\frac{t}{U} \gtrsim \frac{kT}{4t}$. We believe, however, that our method gives a correct description for the tendencies in the half-filled system at infinite U, and for the qualitative behavior up to $n_h \approx 0.2$.

There are, however, some important limitations to this method, due to which we are not able to predict a ferromagnetic state with certainty.

As we know from a theorem by Ghosh,[12] similar to the Mermin-Wagner theorem for the Heisenberg model, the Hubbard model does not have long-range ordering in two dimensions for finite temperatures. Thus, we must expect a ferromagnetic phase in the two-dimensional case to be of the Kosterlitz-Thouless type. Our method is essentially based on short-range information from the high-temperature expansion (which is obtained via calculations on small systems). It gives similar results for the square and the simple cubic lattices, as can be seen in Figs. 5 and 6, and we cannot distinguish between different kinds of phases occurring.

Also, the method currently fails to describe the case of a paramagnetic-antiferromagnetic transition, due to the fact that a divergent background is not treated correctly. We can therefore calculate only possible second-order phase transitions between a paramagnetic and a ferromagnetic phase, for the case of finite hole density. At half filling, we do find a finite Néel temperature for any finite value of the parameter βU (see Fig. 8). This implies that, near half filling, there is a transition from a paramagnetic to an antiferromagnetic state at a higher temperature than the calculated paramagnetic-ferromagnetic transition. Thus, the paramagnetic-ferromagnetic transition cannot occur, and one must study the antiferromagnetic-ferromagnetic transition to determine the ground-state behavior.

Finally, due to the thermodynamic approach in which all possible states are taken into account, our method cannot distinguish special states that may start to dominate the system at low temperatures. Such states, if any, are not recognized by the high-temperature expansion. An example of this is the fact that it fails to reflect the influence of $m = 1$ states in an $m = 0$ system.

We can compare our results to the work of Putikka *et al.*,[13] who calculate series expansions similar to those used by us, for the related t-J model, and extrapolate to low temperatures by means of Padé approximants. For $J > 0$, in the limit of small J, the t-J model is equivalent to the large-U Hubbard model. They find a region of weak ferromagnetism (i.e., the spins are not fully aligned) for small positive J, at hole density $n_h < 0.28 \pm 0.05$, which is in good agreement with our results.

It is also encouraging to note that some of our results are in reasonable quantitative agreement with results using an approximation of an entirely different nature. By means of the slave-boson mean-field approach (at $T = 0$), Denteneer and Blaauboer[14] find a critical hole density $n_h^c = 1/3$ for ferromagnetism to occur at $U = \infty$, in agreement with the values 0.27–0.29 found here (see Fig. 4). They also find that the value of U/t above which ferromagnetism can occur is $U/t = 20$ (at $n_h = 0.17$), whereas one may extrapolate the results of our Fig. 5(a) to $T = 0$ to find $U/t = 15$ (at $n_h = 0.15$).

von der Linden and Edwards[15] use a variational ap-

proach to find a ferromagnetic region in the $T = 0$ phase diagram of the square-lattice Hubbard model. They rigorously conclude that the state of complete spin alignment is unstable when $n_h > 0.29$, for all U, and when $U/t < 42$, for all n_h. The latter value is significantly higher than the value above which we find ferromagnetism, but we assume that that is due to the fact that they consider only strong ferromagnetism (full alignment of the spins), whereas our method may also include weak ferromagnetism.

Also the results of Barbieri *et al.*,[16] who consider systems with a large (but finite) number of holes, support the existence of ferromagnetic behavior.

A final comparison that can be made is for the relation between the Néel temperature and U/t in the half-filled system. From Fig. 8 one can calculate that the paramagnetic-antiferromagnetic transition occurs for $kT_N \approx 3.85t/U$. The large-U Hubbard model at half filling is known to be equivalent to an antiferromagnetic Heisenberg spin model, for which estimates of the values of the critical temperature are given in Ref. 17. According to the results mentioned there, the relation would be $kT_N \approx 3.80t/U$, which is in very good agreement.

ACKNOWLEDGMENT

This research was supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO).

APPENDIX A: ENUMERATION OF PATHS

In this appendix we describe an efficient way to calculate the moments of the density of states, for the case of infinite U, by which we have calculated 22 of these moments for the square lattice, and 16 for the simple cubic lattice. We start from Eq. (20), which we expand in terms of the parameter βt:

$$\frac{Z_1^h}{N} = \sum_{n=0}^{\infty} (\beta t)^n \frac{(-1)^n}{n!} M_n(m) \ , \tag{A1}$$

with the moments of the density of states defined as

$$M_n(m) \equiv \int d\varepsilon \bar{\rho}(\varepsilon, m) \varepsilon^n \ . \tag{A2}$$

We can write the partition function for one hole according to its definition [cf. (10)] also as

$$Z_1^h \equiv e^{(N-1)\beta\epsilon_{\rm HF}} Z_{N-1} \tag{A3}$$

$$= \binom{N-1}{N_\uparrow(m)}^{-1} \sum_{|i,\alpha_i(m)\rangle} \langle i, \alpha_i(m)| e^{-\beta \mathcal{H}_{\rm kin}} |i, \alpha_i(m)\rangle \ , \tag{A4}$$

where the summation is over all states $|i, \alpha_i(m)\rangle$ with a hole at site i and with a spin background $\alpha_i(m)$ such that the magnetization per spin is indeed m. $N_\uparrow$ denotes the number of electrons with spin up, which depends on m, and the factor $\binom{N-1}{N_\uparrow(m)}$ is the total number of possible background configurations given the location of the hole, which accounts for the spin degrees of freedom. In the thermodynamic limit, this factor is exactly equal to the exponential factor in (A3), as one easily checks by applying Stirling's formula for the binomial, and with (13) for $\epsilon_{\rm HF}$. The summation over i gives a trivial (translational) factor N, and we can expand the exponential in powers of βt to obtain

$$\frac{Z_1^h}{N} \binom{N-1}{N_\uparrow(m)} = \sum_{|\alpha(m)\rangle} \sum_n \frac{(-1)^n}{n!} A_n(\alpha(m)) (\beta t)^n \ , \tag{A5}$$

where

$$A_n(\alpha(m)) = \langle \alpha(m)| \left(\frac{\mathcal{H}_{\rm kin}}{t} \right)^n |\alpha(m)\rangle \tag{A6}$$

is the number of walks of length n in the configuration space that restore the spin background $\alpha(m)$ to its original state. Comparing (A1) and (A5) we see that

$$M_n(m) = \binom{N-1}{N_\uparrow(m)}^{-1} \sum_{|\alpha(m)\rangle} A_n(\alpha(m)) \ . \tag{A7}$$

Thus $M_n(m)$ is precisely the sum over all possible closed walks w_n of length n, summing the fraction of spin backgrounds that is restored by w_n. Such a walk induces a permutation $\mathcal{P}(w_n)$ of the background spins, which can be written as a product of disjunct cyclic permutations $\mathcal{P}_i(w_n)$ with length $|\mathcal{P}_i(w_n)| > 1$. In order to restore the spin background $\alpha(m)$, the direction of the spin on each site must remain unchanged, when applying $\mathcal{P}_i(w_n)$. Thus, all spins that are interchanged by this permutation must point in the same direction. As the number of spins involved is negligible compared to the total number of spins, we may approximate that the probability to find an individual spin pointing up or down is given by $\frac{1+m}{2}$ and $\frac{1-m}{2}$, respectively. Hence the fraction of backgrounds in which the alignment of the spins remains unchanged under the permutation $\mathcal{P}_i(w_n)$ is $\left(\frac{1+m}{2}\right)^l + \left(\frac{1-m}{2}\right)^l$, where $l = |\mathcal{P}_i(w_n)|$ is the number of spins involved in the permutation. Thus, we can calculate M_n as

$$M_n(m) = \sum_{w_n} \prod_i \left[\left(\frac{1+m}{2} \right)^{|\mathcal{P}_i(w_n)|} + \left(\frac{1-m}{2} \right)^{|\mathcal{P}_i(w_n)|} \right] . \tag{A8}$$

For the actual evaluation of this expression we proved an elegant theorem that enables us to significantly extend earlier calculations of the moments to $n = 22$. Defining a *retracing sequence* as two subsequent steps of the hole in opposite directions (thus after two steps the hole is back in its previous position; note that the last and first steps of a closed walk are considered to be subsequent

as well), one can make a distinction between *reducible* and *irreducible* closed walks: An irreducible walk does not contain any retracing sequence, whereas a reducible walk does. A reducible walk can be made irreducible by repeatedly removing its retracing sequences; the result is called the irreducible part of the walk. Note that a retracing sequence does not permute spins, and so the irreducible part of a walk induces the same permutation of the spins as the walk itself. Thus, it is sufficient to study only irreducible walks if one knows of how many reducible walks of a given length it is the irreducible part. We proved the following formula: The number of closed walks of length $l+2n$ on a hypercubical lattice with coordination number z that have a given irreducible part of length $l>0$ is

$$N_{\rm ir}(l,n)=(z-1)^n\begin{pmatrix} l+2n \\ n \end{pmatrix} . \tag{A9}$$

This greatly facilitates the calculation of (A8).

APPENDIX B: INVERSE SUSCEPTIBILITY IN THE INTERACTING-HOLE APPROXIMATION

In this appendix we give the formula for the inverse susceptibility in the interacting-hole approximation, using the theory given in Sec. IV. We start from Eq. (56), which has to be differentiated with respect to m in order to get the equivalent of (59), with (50) for $\ln Z_{\rm gr}$:

$$\begin{aligned}\beta h=\beta h_{\rm HF}+n_h\frac{\partial\beta\mu_h}{n_s\partial m}-\int d\varepsilon\frac{\partial\bar\rho(\varepsilon)}{n_s\partial m}\ln(1+e^{-\beta\tilde\varepsilon})+\int d\varepsilon\bar\rho(\varepsilon)n(\tilde\varepsilon)\frac{\partial\beta\tilde\varepsilon}{n_s\partial m}\\ -\beta t\int d\varepsilon\int d\varepsilon'\frac{\partial\bar\rho(\varepsilon)}{n_s\partial m}n(\tilde\varepsilon)\bar\rho(\varepsilon')n(\tilde\varepsilon')\phi(\varepsilon,\varepsilon')-\beta t\int d\varepsilon\int d\varepsilon'\bar\rho(\varepsilon)\frac{\partial n(\tilde\varepsilon)}{n_s\partial m}\bar\rho(\varepsilon')n(\tilde\varepsilon')\phi(\varepsilon,\varepsilon')\\ -\frac{\beta t}{2}\int d\varepsilon\int d\varepsilon'\bar\rho(\varepsilon)n(\tilde\varepsilon)\bar\rho(\varepsilon')n(\tilde\varepsilon')\frac{\partial\phi(\varepsilon,\varepsilon')}{n_s\partial m} ,\end{aligned} \tag{B1}$$

where n_h is given by

$$n_h=-\int d\varepsilon\bar\rho(\varepsilon)n(\tilde\varepsilon)\frac{\partial\beta\tilde\varepsilon}{\partial\beta\mu_h}+\beta t\int d\varepsilon\int d\varepsilon'\bar\rho(\varepsilon)n(\tilde\varepsilon)\bar\rho(\varepsilon')\frac{\partial n(\tilde\varepsilon')}{\partial\beta\mu_h}\phi(\varepsilon,\varepsilon') . \tag{B2}$$

This may look awkward, but if we look at the derivatives of $\tilde\varepsilon$ [see Eq. (51)] we see that many of these terms cancel. Let us first look at the expression (B2) for the hole density. As we are working at fixed hole density, derivatives of the Fermi factor do not play a role in these equations, and they vanish. We need the derivative of $\tilde\varepsilon$ with respect to $\beta\mu_h$,

$$\frac{\partial\beta\tilde\varepsilon}{\partial\beta\mu_h}=-1+\beta t\int d\varepsilon'\bar\rho(\varepsilon')\phi(\varepsilon,\varepsilon')\frac{\partial n(\tilde\varepsilon')}{\partial\beta\mu_h} , \tag{B3}$$

and so we see that indeed there is a cancellation of terms, leaving us with the relation

$$n_h=\int d\varepsilon\bar\rho(\varepsilon)n(\tilde\varepsilon) . \tag{B4}$$

Then, we rewrite the expression for the magnetic field with

$$\frac{\partial\beta\tilde\varepsilon}{n_s\partial m}=-\frac{\partial\beta\mu_h}{n_s\partial m}+\beta t\int d\varepsilon'\frac{\partial\bar\rho(\varepsilon')}{n_s\partial m}n(\tilde\varepsilon')\phi(\varepsilon,\varepsilon')+\beta t\int d\varepsilon'\bar\rho(\varepsilon')\frac{\partial n(\tilde\varepsilon')}{n_s\partial m}\phi(\varepsilon,\varepsilon')+\beta t\int d\varepsilon'\bar\rho(\varepsilon')n(\tilde\varepsilon')\frac{\partial\phi(\varepsilon,\varepsilon')}{n_s\partial m} . \tag{B5}$$

Using this expression it is straightforward to check that (B1) reduces to

$$\beta h=\beta h_{\rm HF}-\int d\varepsilon\frac{\partial\bar\rho(\varepsilon)}{n_s\partial m}\ln(1+e^{-\beta\tilde\varepsilon})+\frac{\beta t}{2}\int d\varepsilon\int d\varepsilon'\bar\rho(\varepsilon)n(\tilde\varepsilon)\bar\rho(\varepsilon')n(\tilde\varepsilon')\frac{\partial\phi(\varepsilon,\varepsilon')}{n_s\partial m} . \tag{B6}$$

In order to derive the inverse susceptibility from this expression, we have to take the derivative with respect to $n_s m$ again, and put $m=0$. For reasons of symmetry it is easy to show that the first derivatives with respect to m of all functions appearing in the integrals vanish at $m=0$. Thus, in the terms in (B6) we only have to consider the derivatives of the functions that have been differentiated once already:

$$\beta N\chi_{\rm FM}^{-1}=\left.\frac{\partial\beta h_{\rm HF}}{n_s\partial m}\right|_{m=0}-\int d\varepsilon\left.\frac{\partial^2\bar\rho(\varepsilon)}{n_s^2\partial m^2}\right|_{m=0}\ln(1+e^{-\beta\tilde\varepsilon})+\frac{\beta t}{2}\int d\varepsilon\int d\varepsilon'\bar\rho(\varepsilon)n(\tilde\varepsilon)\bar\rho(\varepsilon')n(\tilde\varepsilon')\left.\frac{\partial^2\phi(\varepsilon,\varepsilon')}{n_s^2\partial m^2}\right|_{m=0} . \tag{B7}$$

This can again be expressed in terms of $\rho(\varepsilon)$ (note that also ϕ is being Legendre transformed):

$$\begin{aligned}\beta N\chi_{\rm FM}^{-1}=\left.\frac{\partial\beta h_{\rm HF}}{n_s\partial m}\right|_{m=0}\Bigg(1-\left.\frac{\partial\beta h_{\rm HF}}{n_s\partial m}\right|_{m=0}\Bigg[\int d\varepsilon\frac{\partial^2\rho(\varepsilon)}{\partial(\beta h)^2}\ln(1+e^{-\beta\tilde\varepsilon})\\ +\frac{\beta t}{2}\int d\varepsilon\int d\varepsilon'\rho(\varepsilon)n(\tilde\varepsilon)\rho(\varepsilon')n(\tilde\varepsilon')\frac{\partial^2\phi(\varepsilon,\varepsilon')}{\partial(\beta h)^2}\Bigg]_{h=0}\Bigg) ,\end{aligned} \tag{B8}$$

which is the modification of (62) for interacting holes.

[1] Y. Nagaoka, Phys. Rev. **147**, 392 (1966).
[2] For a summary, see, e.g., E. Müller-Hartmann, Th. Hanish, and R. Hirsch, Physica B **186–188**, 834 (1993), and references therein.
[3] D.F.B. ten Haaf and J.M.J. van Leeuwen, Phys. Rev. B **46**, 6313 (1992).
[4] K. Kubo, Prog. Theor. Phys. **64**, 758 (1980); K. Kubo and M. Tada, *ibid.* **69**, 1345 (1983) and **71**, 479 (1984).
[5] K.K. Pan and Y.L. Wang, Phys. Rev. B **43**, 3706 (1991); J. Appl. Phys. **69**, 4656 (1991).
[6] J.A. Henderson, J. Oitmaa, and M.C.B. Ashley, Phys. Rev. B **46**, 6328 (1992).
[7] W.F. Brinkman and T.M. Rice, Phys. Rev. B **2**, 1324 (1970).
[8] Y.S. Yang, C.J. Thompson, and A.J. Guttmann, Phys. Rev. B **42**, 8431 (1990).
[9] S.V. Meshkov and D.V. Berkov, Phys. Lett. A **170**, 448 (1992).
[10] See, e.g., L.E. Reichl, *A Modern Course in Statistical Physics* (University of Texas Press, Austin, 1980).
[11] See, e.g., N.N. Bogolubov, Jr., *A Method for Studying Model Hamiltonians* (Pergamon Press, New York, 1972).
[12] D.K. Ghosh, Phys. Rev. Lett. **27**, 1584 (1971).
[13] W.O. Putikka, M.U. Luchini, and M. Ogata, Phys. Rev. Lett. **69**, 2288 (1992).
[14] P.J.H. Denteneer and M. Blaauboer, J. Phys. Condens. Matter (to be published).
[15] W. von der Linden and D.M. Edwards, J. Phys. Condens. Matter **3**, 4917 (1991).
[16] A. Barbieri, J.A. Riera, and A.P. Young, Phys. Rev. B **41**, 11697 (1990).
[17] G.S. Rushbrooke, G.A. Baker, Jr., and P.J. Wood, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M.S. Green (Academic, London, 1974), Vol. 3, pp. 305–307.