From the Hubbard model to classical spin-fluctuation theory

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In this paper we develop a microscopic foundation for the Murata-Doniach model of spin fluctuations which has been widely used in connection with band-structure calculations. The main result of the paper is the formulation of the partition function of an itinerant system as a functional integral over magnetization modes, and an explicit formula for the energy functional appearing in the exponent of the Boltzmann factor. Such a derivation is made since former theoretical investigations focus on the interaction part of the partition function Z/Z_0, whereas the Murata-Doniach model is formulated in terms of a functional integral for the complete partition function Z. We start with an approximate description of magnetic excitations of noninteracting fermions within collective modes, and derive a bosonlike partition function for these magnetization modes. This is combined with the well-known result for the interaction part of the partition function in the Hubbard model obtained by functional-integral theory. The leading term of the energy functional appearing in the exponent of the partition function agrees with that of the Ginzburg-Landau expansion for the energy of a classical magnetization field. In the course of the transformation to a bosonlike system we predict that the cutoff wave vector q_c which must be introduced in the classical model is temperature dependent with $q_c \sim T^{1/3}$. It is shown that the frequencies of the collective modes are reduced by the Stoner enhancement factor compared with the one-particle excitation energies of Stoner theory.

I. INTRODUCTION

The importance of spin fluctuations for the magnetic properties of itinerant metallic systems at finite temperature has been shown by extensive investigations since the late 1960s. Several approaches to the theoretical treatment of spin fluctuations have developed so far.

The most recent approach was made by Pindor *et al.*,[1] Oguchi *et al.*,[2] and Györffy *et al.*,[3,4] essentially inspired by the availability of sophisticated computational techniques. Following an idea of Hubbard[5,6] and Hasegawa[7] the authors allow for magnetic fluctuations within the frame of the Korringa-Kohn-Rostoker (KKR) band-structure formalism by admitting local magnetic moments with randomly distributed signs on the lattice sites. The coherent potential approximation (CPA) provides a method to calculate the absolute value of the randomly distributed, disordered local moments self-consistently.

Moriya and Kawabata[8] expressed the correction to the Hartree-Fock free energy in terms of the dynamical and wave-vector-dependent susceptibility. Neglecting the frequency and wave-vector dependence of the interaction contribution to the irreducible susceptibility, and assuming a free-electron-like band, they obtain an implicit equation for the static, uniform susceptibility for a given temperature. As the main result of the self-consistent treatment the theory predicts a Curie-Weiss law for weakly correlated itinerant magnets.

The theory which has been most extensively investigated and widely used in calculations of real systems starts with a representation of the partition function of the system in terms of a functional integral. These investigations focus either on the complete partition function Z or on the interaction part Z/Z_0, Z_0 being the partition function of the noninteracting system.

Discussions belonging to the latter category[5–7,9–21] are microscopically well founded, making use of the Hubbard-Stratonovich technique.[22,23] Most of these papers deal with fluctuations localized in real space and treat the random potentials on the lattice sites with the CPA technique. In the opposite limit Hertz and Klenin[15,16] developed a theory for long wavelength fluctuations, yet difficult to apply to systems of practical interest.

On the other hand Murata and Doniach[24] used a classical functional integral representation for the partition function

$$Z = \int \mathcal{D}[\mathbf{m}] \exp\{-\beta \mathcal{H}[\mathbf{m}]\}, \tag{1}$$

$\mathcal{H}[\mathbf{m}]$ being the energy functional of the system in terms of the magnetization field $\mathbf{m}(r)$. Originally $\mathcal{H}$ was given as a Ginzburg-Landau polynomial, the expansion coefficients being calculated from the Stoner-Wohlfahrt theory.[25] In this form the Murata-Doniach (MD) model was applied to study the magnetic equation of state of weak ferromagnets,[26,27] to calculate the Curie temperature and its pressure dependence of itinerant metals,[28–31] to investigate magnetoelastic anomalies and the Invar problem,[32–35] and to explain anomalies in the temperature dependence of the susceptibility and metamagnetism.[36–39] With the availability of spin-polarized band-structure methods $\mathcal{H}$ could be de-

termined from a polynomial fit of the calculated energy functional.[40–44]

The theory has recently been generalized to arbitrary energy functionals,[27,45,46] which offers the possibility to cover arbitrary fluctuation amplitudes of realistic systems. Although phenomenologically well justified, a microscopic foundation of this approach would be desirable.

In this paper we use the Hubbard model to derive a functional-integral expression for the complete partition function Z. Our approach is intended to be applied to weakly and nearly ferromagnetic metals with mainly delocalized fluctuations. To this end we put all electronic degrees of freedom into a set of collective modes. This is done within the static approximation for spin fluctuations. Subsequent to our formalism this deficiency is compensated in some sense by a proper temperature-dependent cutoff wave vector. A transformation from a fermionic system to bosons was already proposed by Tomonaga[47] in one dimension and the essential approximation only holds in this case. In our approach a transformation of noninteracting systems is made by means of a variational principle. The boson states which we are constructing presumably represent the fermionic system only in a rough approximation, yet the quantities we are really interested in, the frequencies of the bosons and the free energy, match the true values much better. Including the electron-electron interaction through the known expression for Z/Z_0, we arrive at a generalized formula of the Murata-Doniach type.

To derive the MD model some serious approximations have to be made. From a theoretical point of view more rigorous approaches are available, e.g., Korenman's rotational invariant Green's function theory in the local band limit which in principle includes dynamical effects[48–50] or the SCR theory of Moriya and Kawabata for weakly correlated systems.[8,51,52]

However, the generalized MD model benefits from its good applicability to realistic systems, because the energy functional can be calculated with electronic band-structure methods. Thus a particular system is not characterized by a few parameters like in other theories but by the complete energy functional of the system. This allows the model to be critically tested against experiment. Such comparisons of experimental results with predictions from the theory presented here have already been performed by us for the case of PdAg and PdH alloys.[46] The sensitivity of the temperature-dependent susceptibility on the composition of the alloys could be reproduced in good agreement with experiment.

In this paper we derive the MD model from first principles for the first time, being aware of the approximations which are necessary in some steps. All these points are discussed in the text. Although the MD model is only valid in some qualitative sense, it is applied even in its simpler form using a Landau expansion and a temperature-independent cutoff wave vector,[37–39,42–44] and its extended version[45,46] has turned out to be successful for those systems which we have already investigated.

Our paper is organized as follows: In Sec. II a set of creation and annihilation operators for spin fluctuations in a noninteracting fermion gas is introduced, which approximately obey the commutation relations of bosons. By means of second quantization a Fock space for spin fluctuations is constructed. An upper boundary for the free energy of the noninteracting fermions in terms of these spin fluctuation states is given in Sec. III. After a minimization procedure with respect to the boson frequencies this boundary is shown to have the form of the free energy of a noninteracting boson system. It is used as an approximation to the free energy of noninteracting fermions. The corresponding partition function Z_0 is written as a functional integral. In Sec. IV we combine the functional integral of the interaction term Z/Z_0 with that of Z_0 and obtain an expression for Z as indicated in Eq. (1). In Sec. V the formalism is applied to systems with a static magnetic field. A discussion concerning the proper choice for the temperature-dependent cutoff wave vector and a comparison between Stoner and spin-fluctuation excitation energies follows in Sec. VI.

II. CONVERTING A NONINTERACTING FERMION SYSTEM TO NONINTERACTING BOSONS

We define a set of creation operators for magnons of a noninteracting fermion system by

$$\mathcal{O}_{q+}^{\dagger} = \frac{1}{\mathcal{N}_{q+}} \sum_k [c_{(k+)q+} a_{k-q\uparrow}^{\dagger} a_{k\downarrow} + c_{(k-)q+} a_{k+q\downarrow}^{\dagger} a_{k\uparrow}],$$

$$\mathcal{O}_{q-}^{\dagger} = \frac{1}{\mathcal{N}_{q-}} \sum_k [c_{(k+)q-} a_{k-q\downarrow}^{\dagger} a_{k\uparrow} + c_{(k-)q-} a_{k+q\uparrow}^{\dagger} a_{k\downarrow}],$$

$$\mathcal{O}_{qz}^{\dagger} = \frac{1}{\mathcal{N}_{qz}} \sum_k \left[c_{(k+)qz} \frac{1}{\sqrt{2}} (a_{k-q\uparrow}^{\dagger} a_{k\uparrow} - a_{k-q\downarrow}^{\dagger} a_{k\downarrow}) + c_{(k-)qz} \frac{1}{\sqrt{2}} (a_{k+q\uparrow}^{\dagger} a_{k\uparrow} - a_{k+q\downarrow}^{\dagger} a_{k\downarrow}) \right]. \tag{2}$$

Here $+$, $-$, and z indicate the three components of magnetization and q is taken from the positive half-space of wave vectors. $a^{\dagger}$ and a denote creation and annihilation operators of fermions. As will be shown in Sec. III B the absolute values of the constants $c_{(k\sigma)q\nu}$ are given by

$$|c_{(k+)q+}|^2 = \frac{f_{k\downarrow}(1 - f_{k-q\uparrow})}{\epsilon_{k-q\uparrow} - \epsilon_{k\downarrow}},$$

$$|c_{(k-)q+}|^2 = \frac{f_{k\uparrow}(1 - f_{k+q\downarrow})}{\epsilon_{k+q\downarrow} - \epsilon_{k\uparrow}},$$

$$|c_{(k+)q-}|^2 = \frac{f_{k\uparrow}(1 - f_{k-q\downarrow})}{\epsilon_{k-q\downarrow} - \epsilon_{k\uparrow}},$$

$$|c_{(k-)q-}|^2 = \frac{f_{k\downarrow}(1 - f_{k+q\uparrow})}{\epsilon_{k+q\uparrow} - \epsilon_{k\downarrow}},$$

$$|c_{(k+)qz}|^2 = \sum_{\sigma} \frac{f_{k\sigma}(1 - f_{k-q\sigma})}{\epsilon_{k-q\sigma} - \epsilon_{k\sigma}},$$

$$|c_{(k-)qz}|^2 = \sum_{\sigma} \frac{f_{k\sigma}(1 - f_{k+q\sigma})}{\epsilon_{k+q\sigma} - \epsilon_{k\sigma}}, \tag{3}$$

with $f_{k\sigma}$ being the Fermi distribution function for $T = 0$, i.e.,

$$f_{k\sigma} = \Theta(\epsilon_F - \epsilon_{k\sigma}) . \tag{4}$$

Furthermore,

$$\mathcal{N}_{q\nu}{}^2 = \sum_{k\sigma} |c_{(k\sigma)q\nu}|^2 \tag{5}$$

in order to normalize the singly excited state $\mathcal{O}^\dagger_{q\nu} \mid 0\rangle$, with $\mid 0\rangle$ representing the ground state of the fermion system.

Now we set up a set of wave functions each of which represents a spin-fluctuation state with $N_{q\nu}$-fold occupation of the mode $(q\nu)$:

$$\left\{ \prod_{\substack{q_z>0 \\ \nu}} \frac{(\mathcal{O}^\dagger_{q\nu})^{N_{q\nu}}}{\sqrt{N_{q\nu}!}} \mid 0\rangle \right\}_{\{N_{q\nu}=0,1,2,\cdots\}}$$

$$= \{\mid \cdots N_{q_i+} \cdots N_{q_j-} \cdots N_{q_k z} \cdots\rangle\}_{\{N_{q\nu}=0,1,2,\cdots\}} . \tag{6}$$

It is easily seen from the definition of the coefficients $c_{(k\sigma)q\nu}$ that the wave functions do not depend on the order of the creation operators. For the moment we confine ourselves to states whose total occupation number $\sum_{q_z>0,\,\nu} N_{q\nu}$ is small compared with the average number of one-particle excitations N of the fermion system at temperature T (see Fig. 2, Appendix A). In good approximation these states form an orthonormal set, and the Bose commutation relations

$$[\mathcal{O}_{q\nu}, \mathcal{O}_{q'\nu'}] = [\mathcal{O}^\dagger_{q\nu}, \mathcal{O}^\dagger_{q'\nu'}] = 0 , \tag{7}$$

$$[\mathcal{O}_{q\nu}, \mathcal{O}^\dagger_{q'\nu'}] = \delta_{qq'}\delta_{\nu\nu'} \tag{8}$$

hold in the subspace spanned by them (see Appendix A).

Clearly, the total number of fluctuation modes which are necessary to give a reasonable description of the fermion system amounts to N, as mentioned in Appendix A. As an approximation we also apply our results for these physically relevant occupation numbers and take the set of wave functions (6) as a basis for the Fock space of spin fluctuation states. Nevertheless, we perform the calculations in Sec. III as if the total number of fluctuation modes, $\sum_{q_z>0,\,\nu} N_{q\nu}$, were much smaller than the number of one-particle excitations N. This approximation is a part of our model to describe the fermion system within collective modes, and we will treat the spin-fluctuation states as bosons throughout the text.

III. FREE ENERGY OF NONINTERACTING FERMIONS IN TERMS OF A FUNCTIONAL INTEGRAL

A. Variational approximation for the free energy

Based upon the results of the last section, we will now give an approximation of the free energy of noninteracting fermions in terms of the well-known expression of noninteracting bosons. The corresponding frequencies of the boson modes are determined by a minimization procedure. An upper boundary for the free energy F_0 of the actual system is given by the Peierls inequality

$$F_0 \leq F_b + \langle \mathcal{H}_0 - \mathcal{H}_b \rangle_b . \tag{9}$$

Here $\mathcal{H}_b$ is the Hamiltonian of the boson system,

$$\mathcal{H}_b = \sum_{\substack{q_z>0 \\ \nu}} \omega_{q\nu}(\mathcal{O}^\dagger_{q\nu}\mathcal{O}_{q\nu} + \tfrac{1}{2}) , \tag{10}$$

leading to the partition function

$$Z_b = \prod_{\substack{q_z>0 \\ \nu}} \left\{ \sum_n \exp[-\beta\omega_{q\nu}(n+\tfrac{1}{2})] \right\} \tag{11}$$

and the free energy

$$F_b = -\frac{1}{\beta} \sum_{\substack{q_z>0 \\ \nu}} \ln\left\{ \sum_n \exp[-\beta\omega_{q\nu}(n+\tfrac{1}{2})] \right\} . \tag{12}$$

The Hamiltonian of the fermion system,

$$\mathcal{H}_0 = \sum_\sigma \left[\sum_{\substack{k \\ \epsilon_{k\sigma}>\epsilon_F}} \epsilon_{k\sigma} a^\dagger_{k\sigma} a_{k\sigma} - \sum_{\substack{k \\ \epsilon_{k\sigma}\leq\epsilon_F}} \epsilon_{k\sigma}(1 - a^\dagger_{k\sigma} a_{k\sigma}) \right] , \tag{13}$$

measures the energy of excited states relative to the ground state, that is,

$$\mathcal{H}_0 \mid 0\rangle = 0 . \tag{14}$$

$\langle\,\rangle_b$ denotes the statistical mean with respect to the boson states (6).

In Appendix B we use the approximation of small occupation numbers discussed in Sec. II to derive the following explicit form for the Peierls inequality (9):

$$F_0 \leq \frac{1}{\beta} \sum_{\substack{q_z>0 \\ \nu}} \ln\left(2\sinh\frac{\beta\omega_{q\nu}}{2}\right) + \sum_{\substack{q_z>0 \\ \nu}} \left[\frac{\exp(-\frac{\beta\omega_{q\nu}}{2})}{2\sinh\frac{\beta\omega_{q\nu}}{2}} \left(\frac{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2 \Delta\epsilon_{(k\sigma)q\nu}}{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2} - \omega_{q\nu} \right) - \frac{\omega_{q\nu}}{2} \right] . \tag{15}$$

Setting the partial derivatives of F_0 with respect to the $\omega_{q\nu}$ equal to zero we get for the frequencies

$$\omega_{q\nu} = \frac{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2 \Delta\epsilon_{(k\sigma)q\nu}}{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2} . \tag{16}$$

In what follows, we identify the optimized right-hand side of (15) with the free energy F_0. This leads to

$$F_0 = -\frac{1}{\beta} \sum_{\substack{q_z>0 \\ \nu}} \ln\left[\sum_n \exp(-\beta\omega_{q\nu} n)\right], \tag{17}$$

with the corresponding partition function

$$Z_0 = \prod_{\substack{q_z>0 \\ \nu}} \sum_n \exp(-\beta\omega_{q\nu} n) . \tag{18}$$

It is remarkable that the correction term $\langle \mathcal{H}_0 - \mathcal{H}_b \rangle_b$ vanishes and that F_0 takes the form of the free energy of a noninteracting boson system without zero point fluctuations. In this sense F_0 has been approximated up to first order in $\mathcal{H}_0 - \mathcal{H}_b$, by one order better than the approximation of the states themselves (2).

B. Specification of $|c_{(k\sigma)q\nu}|^2$

It is clear from the definition of the operators $\mathcal{O}_{q\nu}$ that only the relative values of $|c_{(k\sigma)q\nu}|^2$ are important. We will discuss the case $\nu = +$.

The square of the norm of the wave function of a singly excited mode is given by

$$\langle 0 \mid \mathcal{O}_{q+}\mathcal{O}_{q+}^\dagger \mid 0\rangle = \frac{1}{\mathcal{N}_{q+}^{\,2}} \sum_{k\sigma} |c_{(k\sigma)q+}|^2 . \tag{19}$$

According to the principle of spectral decomposition of a wave function, $|c_{(k\sigma)q+}|^2$ gives the relative probability that the one-particle excitations $(k\downarrow) \to (k-q\uparrow)$ for $\sigma = +$ and $(k\uparrow) \to (k+q\downarrow)$ for $\sigma = -$ can be measured for the singly excited mode $(q+)$. We construct the mode $(q+)$ as a composition of a right-handed and a left-handed helical magnetization wave with opposite wave vector. Thus

$$\begin{aligned} \langle 0 \mid \mathcal{O}_{q+}\mathcal{O}_{q+}^\dagger \mid 0\rangle &\sim m^+(q) + m^-(-q) \\ &= \chi^{+-}(q)[B^+(q) + B^-(-q)] . \end{aligned} \tag{20}$$

The second identity indicates that the magnetic excitations arise from fluctuating magnetic fields $B^+(q)$ and $B^-(-q)$ with which they are connected by a response function, the transversal susceptibility given by[53]

$$\chi^{+-}(q) = \frac{4}{N_a} \sum_k \left[\frac{f_{k\downarrow}(1 - f_{k-q\uparrow})}{\epsilon_{k-q\uparrow} - \epsilon_{k\downarrow}} + \frac{f_{k\uparrow}(1 - f_{k+q\downarrow})}{\epsilon_{k+q\downarrow} - \epsilon_{k\uparrow}} \right] . \tag{21}$$

In (21) N_a is the number of atoms in the macroblock. Inserting Eq. (21) in (20) and comparing the form of $\langle 0 \mid \mathcal{O}_{q+}\mathcal{O}_{q+}^\dagger \mid 0\rangle$ of (19) with (20) we immediately find that the particle-hole excitations contribute to the mode $(q+)$ with the relative probability given in Eq. (3).

With these coefficients the frequency ω_{q+} is written as

$$\omega_{q+} = \frac{\frac{4}{N_a} \sum_k [f_{k\downarrow}(1 - f_{k-q\uparrow}) + f_{k\uparrow}(1 - f_{k+q\downarrow})]}{\chi^{+-}(q)} . \tag{22}$$

This determines the spin-fluctuation frequencies which may be compared with the Stoner excitation energies. Apart from the factor 4 the numerator denotes the total number of particle-hole excitations contributing to the mode $(q+)$ relative to the number of atoms in the macroblock. The graphical representation of the contributing states in k space is shown in Fig. 1 of Appendix A.

For convenience we introduce the abbreviation η_{q+} for the numerator of Eq. (22):

$$\omega_{q+} = \frac{\eta_{q+}}{\chi^{+-}(q)} . \tag{23}$$

C. Z_0 in terms of a functional integral

In the next step we reformulate the partition function (18) as a functional integral. To this end we use a continuous function set[54] $\{\mid \alpha_{q\nu}\rangle\}_{\alpha_{q\nu}\in\mathbb{C}}$,

$$\mid \alpha_{q\nu}\rangle = e^{-\frac{|\alpha_{q\nu}|^2}{2}} \sum_{N_{q\nu}} \frac{\alpha_{q\nu}^{N_{q\nu}}}{\sqrt{N_{q\nu}!}} \mid N_{q\nu}\rangle, \tag{24}$$

which satisfies the closure relation

$$\frac{1}{\pi} \iint d(\mathrm{Re}\alpha_{q\nu}) d(\mathrm{Im}\alpha_{q\nu}) \mid \alpha_{q\nu}\rangle\langle\alpha_{q\nu} \mid = 1 . \tag{25}$$

Inserting the closure relation twice into the equation

$$Z_0 = \prod_{\substack{q_z>0 \\ \nu}} \sum_{N_{q\nu}=0}^{\infty} \langle N_{q\nu} \mid \exp(-\beta\omega_{q\nu}\mathcal{O}_{q\nu}^\dagger\mathcal{O}_{q\nu}) \mid N_{q\nu}\rangle , \tag{26}$$

we get

$$\begin{aligned} Z_0 = \prod_{\substack{q_z>0 \\ \nu}} \frac{1}{\pi} \iint d(\mathrm{Re}\alpha_{q\nu}) d(\mathrm{Im}\alpha_{q\nu}) \\ \times \exp\{-[1 - \exp(-\beta\omega_{q\nu})][(\mathrm{Re}\alpha_{q\nu})^2 + (\mathrm{Im}\alpha_{q\nu})^2]\} \end{aligned} \tag{27}$$

without further approximations. We then replace $1 - \exp(-\beta\omega_{q\nu})$ by its high-temperature limit $\beta\omega_{q\nu}$ in the exponent of (27). This procedure requires the introduction of a cutoff wave vector q_c to ensure that Z_0 remains finite. A more detailed discussion of this problem is given in Sec. VI.

In our model each q mode is represented by exactly one basis state. However, the number of collective states generating a classical q mode should increase with the number of particle-hole excitations contributing to this mode, i.e., with the size of the hatched region of Fig. 1 in

Appendix A. We therefore define the amplitudes of the classical magnetization modes by

$$|m_{q\nu}|^2 = \eta_{q\nu}|\alpha_{q\nu}|^2 . \tag{28}$$

The factor $\eta_{q\nu}$ allows for a q-dependent scaling of the magnetization amplitude of a classical mode. We believe that the deficiencies arising from the incompleteness of the basis (6) can be compensated with this step.

From Eqs. (27) and (28) we then get the desired functional-integral representation of the partition function of a noninteracting fermion system,

$$Z_0 = \prod_\nu \frac{1}{\sqrt{2\pi}} \int dm_{0\nu} \exp\left[-\frac{\beta|m_{0\nu}|^2}{2\chi_\nu(0)}\right] \times \prod_{\substack{q_z>0 \\ \nu}} \frac{1}{\pi} \iint dm_{q\nu} dm_{-q\nu} \exp\left[-\frac{\beta|m_{q\nu}|^2}{\chi_\nu(q)}\right] . \tag{29}$$

In (29) we used a convenient notation for the integration variables and made a continuous extension to $q = 0$. From now on the components $+,-$ or x, y may be used alternatively in all equations.

IV. PARTITION FUNCTION OF AN INTERACTING ELECTRON SYSTEM

Let us now turn to the question of how the electron-electron interaction modifies formula (29). We consider this interaction in the frame of the Hubbard Hamiltonian

$$\begin{aligned} \mathcal{H}_{\text{int}} &= U \sum_i n_{i\uparrow} n_{i\downarrow} \\ &= U \sum_i (\tfrac{1}{2} n_i{}^2 - \tfrac{2}{3}|\mathbf{S}_i|^2) \\ &= U \sum_i [\tfrac{1}{4} n_i{}^2 - (\mathbf{e}_i \cdot \mathbf{S}_i)^2] \end{aligned} \tag{30}$$

and apply the functional integral technique[17,51] in the rotational-invariant treatment of Hubbard[5] and Prange and Korenman[20] to get the following expression for Z/Z_0:

$$\frac{Z}{Z_0} = \int \left(\prod_\nu \frac{d\xi_{0\nu}}{\sqrt{2}}\right) \iint \left(\prod_{\substack{q_z>0 \\ \nu}} d\xi_{q\nu} d\xi_{-q\nu}\right) \times \exp\left(-\frac{\pi}{2} \sum_{q,\nu} |\xi_{q\nu}|^2 - \beta\Psi\right) . \tag{31}$$

Equation (31) has been written in the static approximation and in wave-vector-dependent notation. The functional Ψ can be expressed with the noninteracting Matsubara function G^0 and the fluctuating potential V:

$$\Psi[\boldsymbol{\xi}] = -\frac{1}{\beta} \mathrm{Tr}[\ln(1 - G^0 V)] . \tag{32}$$

The matrix representations of these one-particle quantities are given by

$$\langle mk'\sigma' \mid G^0 \mid mk\sigma\rangle = \delta_{kk'}\delta_{\sigma\sigma'} G^{0\sigma}_{mk} , \tag{33}$$

$$\langle mk'\sigma' \mid V \mid mk\sigma\rangle = \left(\frac{4\pi U}{N_a\beta}\right)^{\frac{1}{2}} \sum_\nu \xi_{k-k'\nu} \langle \sigma' \mid S_\nu \mid \sigma\rangle , \tag{34}$$

where

$$G^{0\sigma}_{mk} = \frac{1}{i\omega_m - \epsilon_{k\sigma} + \epsilon_F} , \tag{35}$$

and the ω_m are the fermion Matsubara frequencies $(2m+1)\pi/\beta$. We then expand the logarithm in Eq. (32) in powers of G^0V, confining ourselves to the component $\nu = z$ for simplicity. Inclusion of other components would mix the terms of all components with rather involved notation following. With this restriction we obtain

$$\Psi = \frac{1}{\beta} \sum_{n=1}^{\infty} \left[\frac{1}{n}\left(\frac{\pi U}{N_a\beta}\right)^n \sum_{\substack{mk \\ q_1,\cdots,q_{2n-1}}} G^0_{mk} \xi_{q_1} \cdots G^0_{m\,k-q_1-\cdots-q_{2n-1}} \xi_{-q_1-\cdots-q_{2n-1}}\right] . \tag{36}$$

The equation holds for paramagnetic and for ferromagnetic systems. The $\epsilon_{k\sigma}$ are the one-particle energies of the noninteracting electron system; that is, they are independent of the spin index, while all interaction effects are included in the Hubbard parameter U. The notation for Ψ can be simplified with the abbreviation

$$\Phi^{(2n)}(q_1, \cdots, q_{2n-1}) = \frac{1}{N_a\beta n} \sum_{mk} G^0_{mk} G^0_{mk-q_1} \cdots G^0_{mk-q_1-\cdots-q_{2n-1}} . \tag{37}$$

In the special case $n = 1$ this is the noninteracting susceptibility

$$\Phi^{(2)}(q) = -\frac{1}{2}\chi(q) . \tag{38}$$

In order to get the complete partition function Z, Eqs. (29) and (31) have to be multiplied. In evaluating (31), terms of Ψ containing the same ξ_q more than once will be neglected for the moment since their number is small compared with the number of completely mixed terms. For a given q the whole exponent in (31) may then be divided into terms containing $|\xi_q|^2$, ξ_q, ξ_{-q}, or neither ξ_q nor ξ_{-q}. Thus a quadratic form of ξ_q in the exponent results leading to a Gaussian integral with respect to the integration variables ξ_q and ξ_{-q}. The integrations over ξ_q

and ξ_{-q} in Z/Z_0 as well as the integrations over m_q and m_{-q} in Z_0 can be performed analytically. The resulting product is rewritten as one Gaussian integral over the variables m_q and m_{-q}. We thus get one integral over the magnetization mode q with an integrand similar to that of Z/Z_0, however each term in the exponent containing m_q or m_{-q} multiplied by $[\beta/\pi\chi(q)]^{1/2}$.

With this transformation for each q all integrations in Z_0 can be eliminated. Our final result is

$$Z = \int \frac{dm_0}{\sqrt{2\pi}} \iint \Big(\prod_{q_z>0} \frac{1}{\pi} dm_q dm_{-q} \Big) \exp\Big\{ -\beta \mathcal{H}[m] \Big\},$$

$$\mathcal{H}[m] = \frac{1}{2} \sum_q [\chi^{-1}(q) - U] |m_q|^2 + N_a \sum_{n=2}^{\infty} \Big\{ \Big(\frac{U}{N_a} \Big)^n \sum_{q_1 \cdots q_{2n-1}} \frac{\Phi^{(2n)}(q_1, \cdots, q_{2n-1})}{[\chi(q_1) \cdots \chi(-q_1 - \cdots - q_{2n-1})]^{\frac{1}{2}}} m_{q_1} \cdots m_{-q_1 - \cdots - q_{2n-1}} \Big\}. \tag{39}$$

This is the desired form for the complete partition function. Note the remarkable fact that the quadratic term in the spin fluctuations of the energy functional $\mathcal{H}$ equals the first term of the corresponding Ginzburg-Landau expansion, since $\chi^{-1}(q) - U$ is the inverse enhanced susceptibility.

V. APPLICATION TO A UNIFORM EXTERNAL FIELD

In this section we apply our formalism to the case of an external magnetic field and derive a result for the field-dependent partition function consistent with linear response theory. The treatment parallels that of the zero-field case in most aspects.

In order to obtain the partition function of an interacting system in the presence of an external field we have to calculate

$$Z(H) = \exp[-\beta F_0(H)] \frac{Z}{Z_0}(H), \tag{40}$$

with $F_0(H)$ being the field-dependent, noninteracting free energy. We show in Appendix C that the variational scheme of Sec. III yields

$$F_0(H) = F_0(0) - \frac{N_a}{2} \chi(0) H^2 \tag{41}$$

up to order H^2.

The field-dependent expression for the functional Ψ [Eq. (32)] is

$$\Psi = \frac{1}{\beta} \sum_{\sigma} \Big[\sigma \Big(\frac{\pi U}{N_a \beta} \Big)^{\frac{1}{2}} \sum_{mk} G^{0\sigma}_{mk} \xi_0 + \sum_{n=2}^{\infty} \sigma^n \Big(\frac{\pi U}{N_a \beta} \Big)^{\frac{n}{2}} \sum_{\substack{m \\ (k\, q_1 \cdots q_{n-1})_R}} G^{0\sigma}_{mk} \xi_{q_1} \cdots G^{0\sigma}_{m\, k-q_1-\cdots-q_{n-1}} \xi_{-q_1-\cdots-q_{n-1}} \Big], \tag{42}$$

with $G^{0\sigma}_{mk}$ from (C3). In each term of the expansion of (32) n different terms with the same numeric value may be obtained by cyclic permutation of the factors $G^{0\sigma}\xi$. We write only one representative term of the set of permuted elements denoted by $(k\, q_1 \cdots q_{n-1})_R$. The power of this set cancels the factor $1/n$ from the expansion of the logarithm. In fact, different terms are generated by permutation only unless the term itself is constructed by repetition of the same sequence. We will not treat this special case here. In terms of the form

$$\cdots (G^{0\sigma}_{mk} \xi_0)^j G^{0\sigma}_{mk} \xi_{k-k'} \cdots, \quad j = 0, \cdots, n, \quad k - k' \neq 0,$$

we will expand $(G^{0\sigma}_{mk})^{j+1}$ in powers of H. Collecting proper terms for different j we can use the binomial formula to get all terms

$$\cdots \Big\{ G^0_{mk} \Big[\xi_0 - \Big(\frac{N_a \beta}{\pi U} \Big)^{1/2} H \Big] \Big\}^n G^0_{mk} \xi_{k-k'} \cdots, \quad n = 0, 1, \cdots, \quad k - k' \neq 0.$$

In this way the expansion (42) can be transformed into a representation which contains only the zero-field Green functions:

$$\Psi = \frac{2}{\beta}\Big[\sum_{n=1}^{\infty}\Big(\frac{\pi U}{N_a\beta}\Big)^n \sum_{\substack{m \\ (k\,q_1\cdots q_{2n-1})_R}} G^0_{mk}\tilde{\xi}_{q_1}\cdots G^0_{m\,k-q_1-\cdots-q_{2n-1}}\tilde{\xi}_{-q_1-\cdots-q_{2n-1}} - \sum_{n=1}^{\infty}\frac{H^{2n}}{2n}\sum_{mk}\big(G^0_{mk}\big)^{2n}\Big], \tag{43}$$

with

$$\tilde{\xi}_q = \begin{cases} \xi_q & \text{for } q \neq 0, \\ \xi_0 - \big(\frac{N_a\beta}{\pi U}\big)^{1/2} H & \text{for } q = 0\,. \end{cases}$$

Dropping the notation with representatives and changing the integration variable ξ_0 yields

$$\begin{aligned}\frac{Z}{Z_0} = \int \frac{d\xi_0}{\sqrt{2}} \iint \Big(\prod_{q_z>0} d\xi_q d\xi_{-q}\Big) \\ \times \exp\Big\{-\beta\Big[\Big(\frac{N_a\pi}{\beta U}\Big)^{\frac{1}{2}} H\xi_0 + \frac{N_a}{2}\frac{H^2}{U} + \frac{N_a}{2}\chi(0)H^2 + \frac{\pi}{2\beta}\sum_q [1 - U\chi(q)]|\xi_q|^2 \\ + N_a\sum_{n=2}^{\infty}\Big(\frac{\pi U}{N_a\beta}\Big)^n \sum_{(q_1\cdots q_{2n-1})} \Phi^{(2n)}(q_1\cdots q_{2n-1})\xi_{q_1}\cdots\xi_{-q_1-\cdots-q_{2n-1}}\Big]\Big\},\end{aligned} \tag{44}$$

where we have written only the quadratic term in H from the series of Eq. (43). Following the derivation of Sec. IV we get for the field-dependent partition function (40)

$$\begin{aligned} Z(H) &= \int \frac{dm_0}{\sqrt{2\pi}} \iint \Big(\prod_{q_z>0}\frac{1}{\pi} dm_q dm_{-q}\Big) \exp\Big\{-\beta\mathcal{H}(H,\{m_q\})\Big\} \\ \mathcal{H}(H,\{m_q\}) &= \Big[\frac{N_a}{U\chi(0)}\Big]^{1/2} Hm_0 + \frac{N_a}{2}\frac{H^2}{U} + \frac{1}{2}\sum_q [\chi^{-1}(q) - U]|m_q|^2 \\ &\quad + \sum_{n=2}^{\infty}\Big\{\frac{U^n}{N_a^{n-1}}\sum_{q_1\cdots q_{2n-1}} \frac{\Phi^{(2n)}(q_1,\cdots,q_{2n-1})}{[\chi(q_1)\cdots\chi(-q_1-\cdots-q_{2n-1})]^{\frac{1}{2}}} m_{q_1}\cdots m_{-q_1-\cdots-q_{2n-1}}\Big\}. \end{aligned} \tag{45}$$

It can be shown by a shift of the integration variable m_0 that the energy functional is given by

$$\mathcal{H}(H,\{m_q\}) = \frac{1}{2}\sum_q (\chi^U)^{-1}(q)|m_q|^2 - \frac{N_a}{2}\chi^U H^2 \tag{46}$$

up to order U with χ^U as the enhanced susceptibility. Thus the external field contribution to the energy functional agrees with the result of linear response theory.

VI. DISCUSSION AND SUMMARY

The intention of this paper is to derive the partition function of an itinerant magnetic system in the form of a functional integral over magnetization modes. This provides a microscopic foundation of the Murata-Doniach model for spin fluctuations which has been employed by several authors to investigate the magnetic properties of itinerant electron systems based upon spin-polarized band-structure calculations. Now it is possible to write the phenomenological energy functional in terms of the parameters of the Hubbard model. Assuming the number of excited modes to be small compared with the number of thermally excited electrons we find the best approximation for the partition function of noninteracting fermions by a quadratic energy functional. In defining the amplitudes of the classical magnetization modes we have to introduce a q-dependent weight factor to compensate for the incompleteness of the basis states of spin fluctuations. In the course of the transformation to noninteracting bosons all effects of the band-structure are included in an energy functional of quadratic order. On the other hand with the use of functional-integral theory we are able to allow for interaction contributions to the energy functional in all orders. In this way we obtain the fundamental form of the classical partition function as well as the leading term of the Ginzburg-Landau expansion for the energy of magnetization modes.

However, keeping an infinite number of integrals in Eq. (39) causes divergence of the expression for the partition function. This is a consequence of the high-temperature approximation of the static theory, which has been applied in Sec. III C. Similar to the phenomenological theory a cutoff wave vector q_c must be introduced to limit the number of q modes that may be excited thermally. This may be done by regarding F_0 as the free energy of

a boson system with a heat capacity according to the Dulong-Petit rule, or as the free energy of a fermion system with a heat capacity increasing linearly with T. Thus we have

$$C = k_B \sum_{\substack{|q|\le q_c \\ q_z>0 \\ \nu}} 1 = \frac{N_a V_c k_B q_c^3}{4\pi^2} = \gamma T , \tag{47}$$

with V_c being the volume of the primitive cell. This leads to a cutoff wave vector $q_c = q_0 T^{1/3}$, in which q_0 can readily be estimated from the coefficient γ.

The excitation energies for spin fluctuations found in Eq. (22) are in first order modified by the inverse enhancement factor for the interacting case as can be seen from Eq. (39). Therefore

$$\omega_{q\nu}^U = \frac{\eta_{q\nu}}{\chi_\nu^U(q)} . \tag{48}$$

On the other hand applicability of the high-temperature approximation $\omega_{q\nu}^U < 1/\beta$ sets an upper boundary for the q vectors for which our model remains valid:

$$\eta_{q\nu} < \frac{\chi_\nu^U(q)}{\beta} . \tag{49}$$

A straightforward analysis of the geometry of Fig. 1 in Appendix A shows that for $q < k_F$ we have in good approximation

$$\eta_{q+} \approx \frac{8 V_c \pi k_F^2 q}{(2\pi)^3} \tag{50}$$

and in the same q region the wave-vector-dependent inverse susceptibility is given by

$$(\chi_{+-}^U)^{-1}(q) = \chi_{+-}^{-1}(0) - U + \chi_{+-}^{-1}(0)\sigma^2 q^2 . \tag{51}$$

Thus for nearly and weakly ferromagnetic systems $[\chi_{+-}^{-1}(0) \approx U]$ and not too small wave vectors we get from (49)–(51) the following upper boundary for the absolute value of q:

$$q \le \left[\frac{(2\pi)^3 k_B \chi_{+-}(0)}{8 V_c \pi k_F^2 \sigma^2} \right]^{\frac{1}{3}} T^{\frac{1}{3}} . \tag{52}$$

This constitutes the cutoff wave vector q_c. In the case of the strong exchange enhanced paramagnet Pd estimates from Eqs. (47) and (52) are comparable. In Ref. 46 the temperature behavior of the susceptibility of dilute Pd alloys was investigated based on our treatment. It was shown that an agreement of theoretical and experimental curves for a wide temperature range can only be obtained with a temperature-dependent q_c. A $T^{1/3}$ cutoff was also proposed by Murata[17] and Lonzarich and Taillefer.[26]

At very low temperatures corresponding to the excitation of long wavelength modes the asymptotic behavior

$$\chi^U(q) \approx \chi^U(0) \tag{53}$$

suggests a cutoff wave vector $q_c \sim T$. We would like to mention that a violation of the third law of thermodynamics as in the classical theory[24] is avoided with the use of a temperature-dependent q_c.

The different role of the interaction parameter U in spin-fluctuation and in Stoner theory can be seen as follows. In spin-fluctuation theory the mode frequencies of a noninteracting system (22) are a certain mean value of the one-particle excitation energies $\Delta\epsilon_{kq}$ and therefore not essentially different from the Stoner excitations. These mode frequencies are scaled down by the inverse enhancement factor $1-U\chi(q)$ in the interacting case (48). In Stoner theory however the average of the excitation energies

$$\Delta\epsilon_{kq}^U = \epsilon_{k+q} - \epsilon_k + UM \tag{54}$$

does not differ significantly from its noninteracting value. This shows the general trend of the one-particle excitation energies to be higher by the enhancement factor compared with the spin-fluctuation frequencies.

APPENDIX A

Starting from the magnon operators of Eq. (2) and taking the scalar product of the nth excited state $(\mathcal{O}_{q\nu}^\dagger)^n \mid 0\rangle$ with itself we first obtain

$$\langle 0 \mid \mathcal{O}_{q\nu}{}^n \mathcal{O}_{q\nu}^\dagger{}^n \mid 0\rangle = \frac{n! \sum_{(k\sigma)_1 \ne \cdots \ne (k\sigma)_n} \prod_{i=1}^n |c_{(k\sigma)_i q\nu}|^2}{\left(\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2\right)^n} . \tag{A1}$$

We can assume the number of excitations n to be small compared with the number of one-particle excitations contributing to the mode $(q\nu)$. To see this, consider Fig. 1 which shows all one-particle states of the Fermi sphere that might be excited to cause onefold excitation of the mode $(q+)$. Figure 2 shows the occupation of

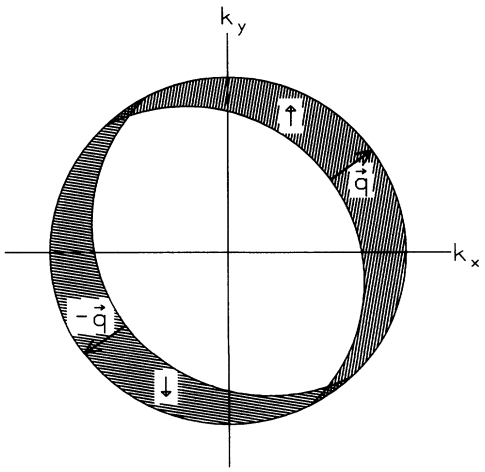

FIG. 1. One-particle states inside the Fermi sphere. The hatched region marks particle states suitable to excite the mode $(q+)$.

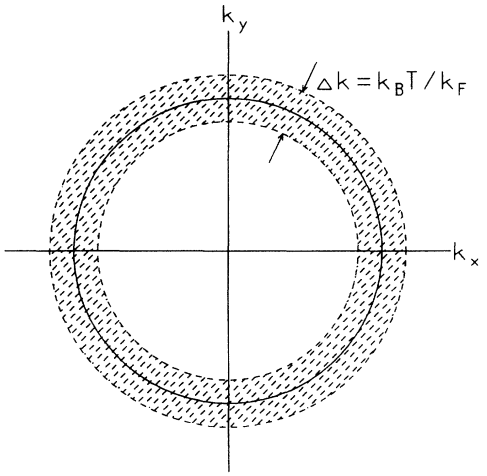

FIG. 2. Fermion system with temperature T. The hatched region marks the range in k space where thermal excitations take place.

states of a fermion system at moderate temperature, especially the smeared region of width $\Delta k = k_B T/k_F$ at the Fermi wave vector k_F. Since the occupation of one $(q\nu)$ mode corresponds to a statistical occupation of one particle-hole pair, the N one-particle excitations in Fig. 2 have to be distributed among N fluctuation excitations. Since many $(q\nu)$ modes are available, the occupation n of a particular mode is small compared with N. The wave vector q of a typical mode and Δk are of the same order of magnitude; that is, the numbers of states in the hatched regions of Figs. 1 and 2 are comparable. This means that the occupation number n of the mode is much smaller than the number of nonvanishing terms in the creation operator $O^\dagger_{q\nu}$. Thus all terms of the denominator of (A1) which are not fully mixed products are neglected. We thus get

$$\langle 0 \mid \mathcal{O}_{q\nu}{}^n \mathcal{O}^\dagger_{q\nu}{}^n \mid 0\rangle = n! \,. \tag{A2}$$

We can therefore define a normalized state $\mid N_{q\nu}\rangle$ with $N_{q\nu}$-fold occupation of the magnetization mode $(q\nu)$ by

$$\mid N_{q\nu}\rangle = \frac{1}{\sqrt{N_{q\nu}!}} (\mathcal{O}^\dagger_{q\nu})^{N_{q\nu}} \mid 0\rangle \,. \tag{A3}$$

As a consequence of Eqs. (A2) and (A3) the action of a creation or annihilation operator on a ket $\mid N_{q\nu}\rangle$ is given by

$$\begin{aligned} \mathcal{O}^\dagger_{q\nu} \mid N_{q\nu}\rangle &= \sqrt{N_{q\nu}+1} \mid N_{q\nu}+1\rangle, \\ \mathcal{O}_{q\nu} \mid N_{q\nu}\rangle &= \sqrt{N_{q\nu}} \mid N_{q\nu}-1\rangle \,. \end{aligned} \tag{A4}$$

The operators $\mathcal{O}^\dagger_{q\nu}$ and $\mathcal{O}_{q\nu}$ obey the commutation relation

$$\langle N_{q\nu} \mid [\mathcal{O}_{q\nu}, \mathcal{O}^\dagger_{q\nu}] \mid N_{q\nu}\rangle = 1 \,. \tag{A5}$$

Since the states (6) do not depend on the order of the creation operators the commutation relations

$$[\mathcal{O}_{q\nu}, \mathcal{O}_{q'\nu'}] = [\mathcal{O}^+_{q\nu}, \mathcal{O}^+_{q'\nu'}] = 0 \tag{A6}$$

hold in the subspace spanned by (6).

A particular basis state is the sum of about $N^{(\sum_{q_z>0,\nu} N_{q\nu})}$ linearly independent terms, since N is an estimate for the number of one-particle states in the hatched region of Fig. 1. Moreover, the terms of other basis states either equal such a term (apart from a constant factor) or are orthogonal to it. Looking for basis states which contain as many terms of the given state as possible it is best to take the given state and to replace two creation operators $\mathcal{O}^\dagger_{q_1\nu}$ and $\mathcal{O}^\dagger_{q_2\nu}$ by operators $\mathcal{O}^\dagger_{q_3\nu}$ and $\mathcal{O}^\dagger_{q_4\nu}$ with $q_1+q_2 = q_3+q_4$. Then one-particle states k, k' can be found with corresponding terms $a^\dagger_{k-q_1} a_k a^\dagger_{k'-q_2} a_{k'}$ and $a^\dagger_{k'-q_3} a_{k'} a^\dagger_{k-q_4} a_k$ being identical. However, the number of these terms is smaller by at least a factor N compared to the total number of terms, $N^{(\sum_{q_z>0,\nu} N_{q\nu})}$. We can therefore assume that the basis states are orthogonal in good approximation.

Now, assuming the total occupation number $\sum_{q_z>0,\nu} N_{q\nu}$ to be small compared to N, a discussion similar to that leading from Eq. (A1) to Eq. (A2) shows the normalization of the states (6). Furthermore, the orthonormal property of these states and Eqs. (A4) can be used to show that the commutation relations

$$[\mathcal{O}_{q\nu}, \mathcal{O}^\dagger_{q'\nu'}] = 0 \quad \text{for} \quad (q\nu) \neq (q'\nu'), \tag{A7}$$

$$[\mathcal{O}_{q\nu}, \mathcal{O}^\dagger_{q\nu}] = 1 \tag{A8}$$

hold in the subspace spanned by the wave functions (6) with small occupation number.

APPENDIX B

The thermal expectation value of an observable A with respect to Bose statistics is defined by

$$\langle A\rangle_b = \frac{\sum_{\{\cdots N_{q\nu}\cdots\}} \langle \cdots N_{q\nu} \cdots \mid A \exp(-\beta \mathcal{H}_b) \mid \cdots N_{q\nu} \cdots\rangle}{\sum_{\{\cdots N_{q\nu}\cdots\}} \exp[-\beta \sum_{q_z>0,\nu} \omega_{q\nu}(N_{q\nu}+\frac{1}{2})]} \,, \tag{B1}$$

where the occupation number $N_{q\nu}$ of each mode runs from zero to infinity. Clearly, $\mathcal{H}_b$ is diagonal with respect to the basis states of (6), and we thus have

$$\langle \mathcal{H}_b \rangle_b = \frac{\sum_{\{\cdots N_{q\nu}\cdots\}} \left\{ \sum_{q_z>0,\nu} \omega_{q\nu}(N_{q\nu}+\frac{1}{2}) \exp[-\beta \sum_{q_z>0,\nu} \omega_{q\nu}(N_{q\nu}+\frac{1}{2})] \right\}}{\sum_{\{\cdots N_{q\nu}\cdots\}} \exp[-\beta \sum_{q_z>0,\nu} \omega_{q\nu}(N_{q\nu}+\frac{1}{2})]} . \tag{B2}$$

$\mathcal{H}_0$ is diagonal in the subspace spanned by the basis states, yet the action of $\mathcal{H}_0$ on a basis state yields a wave function that is not an element of this subspace. This is an effect of the incompleteness of the Fock space, which was constructed to describe spin-fluctuation states but not any state of the fermion system. We get for $\langle \mathcal{H}_0 \rangle_b$

$$\langle \mathcal{H}_0 \rangle_b = \frac{\sum_{\{\cdots N_{q\nu}\cdots\}} \left\{ \sum_{q_z>0,\nu} \langle N_{q\nu} \mid \mathcal{H}_0 \mid N_{q\nu} \rangle \exp[-\beta \sum_{q_z>0,\nu} \omega_{q\nu}(N_{q\nu}+\frac{1}{2})] \right\}}{\sum_{\{\cdots N_{q\nu}\cdots\}} \exp[-\beta \sum_{q_z>0,\nu} \omega_{q\nu}(N_{q\nu}+\frac{1}{2})]} . \tag{B3}$$

Within the approximation discussed in Sec. II the matrix elements of $\mathcal{H}_0$ are given by

$$\begin{aligned} \langle N_{q\nu} \mid \mathcal{H}_0 \mid N_{q\nu} \rangle &= \frac{\sum_{(k\sigma)_1 \neq \cdots \neq (k\sigma)_{N_{q\nu}}} (\prod_{i=1}^{N_{q\nu}} |c_{(k\sigma)_i q\nu}|^2) \sum_{i=1}^{N_{q\nu}} \Delta\epsilon_{(k\sigma)_i q\nu}}{(\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2)^{N_{q\nu}}} \\ &\approx N_{q\nu} \frac{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2 \Delta\epsilon_{(k\sigma)q\nu}}{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2} . \end{aligned} \tag{B4}$$

To obtain the last expression the restricting conditions on the summation indices of the multiple sum have been neglected. The $\Delta\epsilon_{(k\sigma)q\nu}$ denote the energies of the particle-hole excitations. From Eqs. (B2)–(B4) we get

$$\begin{aligned} \langle \mathcal{H}_0 - \mathcal{H}_b \rangle_b = \Bigg[\sum_{\substack{q_z>0 \\ \nu}} \left\{ \sum_{N_{q\nu}} N_{q\nu} \exp[-\beta\omega_{q\nu}(N_{q\nu}+\tfrac{1}{2})] \right\} \left(\frac{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2 \Delta\epsilon_{(k\sigma)q\nu}}{\sum_{k\sigma} |c_{(k\sigma)q\nu}|^2} - \omega_{q\nu} \right) \\ \times \prod_{\substack{(q'\nu')\neq(q\nu) \\ q'_z>0}} \sum_{N_{q'\nu'}} \exp[-\beta\omega_{q'\nu'}(N_{q'\nu'}+\tfrac{1}{2})] \Bigg] \left(\prod_{\substack{q_z>0 \\ \nu}} \sum_{N_{q\nu}} \exp[-\beta\omega_{q\nu}(N_{q\nu}+\tfrac{1}{2})] \right)^{-1} - \sum_{\substack{q_z>0 \\ \nu}} \frac{\omega_{q\nu}}{2} . \end{aligned} \tag{B5}$$

The summations over the occupation numbers $N_{q\nu}$ can be performed exactly, and Eq. (9) takes the form (15).

APPENDIX C

The Hamiltonian of Eq. (13) was chosen for zero ground state energy. In fact, the total energy of a noninteracting system increases as $\frac{N_a}{2}\chi^{-1}M^2$ when a magnetic field H is applied. Therefore, in analogy to Eq. (29), we write for finite magnetic moment M per atom

$$\begin{aligned} F_0(M) = -\frac{1}{\beta} \sum_{\nu} \ln \left\{ \frac{1}{\sqrt{2\pi}} \int dm_{0\nu} \exp\left[-\frac{\beta |m_{0\nu}|^2}{2\chi_{\nu M}(0)} \right] \right\} - \frac{1}{\beta} \sum_{\substack{q_z>0 \\ \nu}} \ln \left\{ \frac{1}{\pi} \iint dm_{q\nu} dm_{-q\nu} \exp\left[-\frac{\beta |m_{q\nu}|^2}{\chi_{\nu M}(q)} \right] \right\} \\ + \frac{N_a}{2} \chi^{-1}(0) M^2 . \end{aligned} \tag{C1}$$

We only consider the component $\nu = z$. In Eq. (C1) the magnetization-dependent susceptibilities which result from the minimization procedure of Sec. III are given by

$$\chi_M(q) = -\frac{1}{N_a \beta} \sum_{\sigma} \sum_{mk} G^{0\sigma}_{mk} G^{0\sigma}_{m\,k-q} , \tag{C2}$$

with

$$G^{0\sigma}_{mk} = \frac{1}{i\omega_m - \epsilon_k + \sigma H + \epsilon_F} . \tag{C3}$$

Expansion of the free energy up to order H^2 gives

$$\begin{aligned} F_0(M) = F_0(0) + \frac{N_a}{2} \chi^{-1}(0) M^2 \\ + \frac{1}{\beta^2} \sum_{q_z>0} \left[\frac{\sum_{mk} (G^0_{mk})^2 (G^0_{m\,k-q})^2}{N_a \chi_0(q)} \right] H^2 . \end{aligned} \tag{C4}$$

The summation index in the last term of (C4) runs up to the cutoff wave vector q_c, and we substitute the summation by an integral in reciprocal space. For an estimate we keep the integrand constant at the value for $q=0$. For

a free-electron-like band we have

$$\chi_0 = \frac{3}{\epsilon_F} \tag{C5}$$

and

$$\frac{1}{2\beta}\sum_{mk}(G^0_{mk})^4 = \frac{5}{64}\frac{N_a}{{\epsilon_F}^3}\,. \tag{C6}$$

Since the integration volume in reciprocal space is smaller than the volume of the Brillouin zone, we get as estimates

$$\frac{1}{\beta^2}\sum_{q_z>0}^{|q_c|}\left[\frac{\sum_{mk}(G^0_{mk})^2(G^0_{m\,k-q})^2}{N_a\chi_0(q)}\right]H^2 \le \frac{5}{576}\frac{N_a}{\beta\epsilon_F}\chi^{-1}(0)M^2 \ll \frac{N_a}{2}\chi^{-1}(0)M^2\,. \tag{C7}$$

Thus the third term of Eq. (C4) will be neglected. The field-dependent free energy is therefore given by

$$F_0(H) = F_0(M) - N_aMH = F_0(0) - \frac{N_a}{2}\chi(0)H^2\,. \tag{C8}$$

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