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Magnetic fluctuations in singlet-ground-state systems*

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This paper discusses the possible effects of coupling between different excitation modes in singlet-ground-state ferromagnets above their Curie points. The picture we use is a Hartree-like one, with every magnetic exciton dressed (in a mean-square sense) by a cloud of other such excitons. This description finds a natural expression in a functional-integral language, where the result can be expressed as a renormalization of the single-ion susceptibility by averaging over a distribution of external fields. We find that the resulting excitation spectrum has a continuum part with a threshold at the crystal-field splitting, in addition to the usual spike at the exciton frequency. The transition temperature is also significantly depressed from its random-phase-approximation value. Some numerical results are presented which suggest that these effects may be important in real systems.

I. INTRODUCTION

In this paper we discuss intrinsic fluctuation effects in the excitation spectrum of induced-moment paramagnets (ferromagnets above T_c). It is well established[1-4] that such systems display a dynamic behavior characteristic of the existence of well-defined magnetic excitations above the transition temperature. Here we investigate the nature and consequences of interactions between these excitations.

In order to treat these effects, which do not appear within the mean-field–RPA (random-phase approximation) theories, we examine a highly simplified model, and develop a microscopic formalism which provides us with a number of possible approximation procedures. We are able to express a generalized Landau free-energy functional as an expansion in powers of a field related to the order parameter. We are able to calculate (at least in principle) the coefficient of each power of the order parameter in the generalized Landau functional, that is, all the bare exciton-exciton coupling vertices.

We work with the Ising model in a transverse field. It is well known that the two-singlet system may be reduced to this model, and it is believed that the dynamic properties of interest in some rare-earth magnets having low transition temperatures can be derived from it. Mean-field–RPA calculations of the spin dynamics predict sharp excitation (magnetic excitons) with a dispersion curve $\omega(\vec{q})$, with $\omega(0)$ going to zero as the transition is approached.

It had been hoped that these simple calculations for this simple model could be taken over more or less intact to explain the data. However, this hope is not borne out by experimental results.[5,6] The theoretical explanation may be simply that the two-singlet model is not sufficiently realistic, and that more crystal-field levels[7-12] and/or coupling to the lattice must be included to give a qualitative account of the experiments.[13] Alternatively or in addition, the problem may lie not with the model itself but with the approximate solutions given hitherto for the model. In this paper we focus exclusively on the latter possibility, and investigate higher-order fluctuation effects within the singlet-singlet model.

There is an additional point of some theoretical interest. The behavior of induced-moment systems is governed by competition between the crystal field, which tends to force the ionic sites into a nonmagnetic ground state at low temperatures, and the magnetic interaction, which tends to hybridize the crystal-field states to produce a magnetic ordering of the crystal. There is, in fact, a critical ratio of magnetic coupling constant to crystal-field energy, such that the system will undergo a magnetic transition only if the ratio exceeds its critical value. Because the systems of experimental interest have nearly critical magnetic coupling constants, one is inevitably in an intermediate-coupling regime where large corrections to mean-field behavior can be expected, if the range of the interaction is not too long.

As we have mentioned, our formal approach involves approximations carried out on the free-energy functional. We find that the physics of the problem, particularly beyond RPA, can be considerably clarified by working in terms of such a Landau functional. Other work which has been done on the transverse Ising model has used decoupling procedures or expansions for the mag-

netic Green's functions themselves. To make contact with other theories, it seems useful to point out that in the present theory, the lowest-order expansion of the free-energy functional in the order parameter corresponds to what is usually called non-self-consistent RPA theory. We display this explicitly. We reserve for a later paper a detailed comparison with theories involving expansion in $1/z$ (z, the number of nearest neighbors).[14,15] These theories correspond in some sense to approximations made after truncating the free-energy functional at the next nontrivial order after RPA in the order-parameter field. The present paper will deal with an approximation which retains fluctuation terms of all orders, and treats them in the simplest self-consistent way. The treatment is similar to that developed by the authors for Hubbard-like paramagnets,[16] and similar validity criteria apply: Because we are doing perturbation theory around RPA theory we do not expect our expression to be valid in the critical region. What we can see is the effect of a fluctuating hybridization of the pure crystal field. The numerical calculations presented are intended to give a semiquantitative estimate of the size of the effect—the limitations on their reliability are discussed in the text.

We mention in passing that our approximation would be the first-order $[O(1)]$ approximation in $1/N$ if our order parameter had N components.[17] This is not very helpful in the present problem, which has a scalar order-parameter field, but may be useful in the singlet-triplet model, where rotational invariance leads to an $N=3$.[18]

We stress that the formal development occurs in two distinct steps. The first—and for the present purposes the more important—is to write down a formal expression for the free-energy functional and to study its general form. The second is to use a variational procedure to approximate the true free-energy functional by a quadratic form. We have emphasized a field-theoretic picture of the problem because of the insight into the exciton-exciton coupling it allows, and because it enables us to take over almost directly the physical picture which we made in the Hubbard-model case. (The present calculation may in fact be done without resorting to these techniques—we have shown elsewhere how to do so.[19]) Incidentally, the effect of competition between crystal-field energies and exchange interaction, which we have mentioned above, has a formal parallel in itinerant magnetism. In both cases we have a characteristic energy of the noninteracting system which must be overcome if the system is to order magnetically. For the Hubbard model, this is the reciprocal of the density of states at the Fermi energy; here we have the ionic-crystal-field splitting. In the Hubbard-model case, our generalized Hartree approximation leads to an effective smearing out of the electronic density of states over an energy range $\approx(Uk_BT)^{1/2}$, where U is the intra-atomic Coulomb interaction, and T is the temperature. Hence for $U \gg k_BT_c$, this effect may be considerably larger than simple thermal broadening. In what follows here, we shall find a similar alteration of the population of the two crystal-field levels, which must be taken into account in computing magnetic properties—crudely stated, these levels develop a width of the order $(Jk_BT)^{1/2}$, where J is now the effective magnetic exchange between sites. We shall make a more precise statement in the text.

In Sec. II we introduce the first part of our formal discussion. We define our model Hamiltonian and obtain a formal expression for the free-energy functional which is the basis of the calculation. In Sec. III we study its properties and their physical meaning. Sec. IV describes our approximation schemes. Section V introduces a static approximation into our description of the mode coupling. This is a separate approximation from the Hartree-like approximation of Sec. IV; it is introduced in order to make illustrative calculations simpler. Section VI discusses these results.

II. MODEL AND FORMALISM

We start from a general Hamiltonian of the form

$$H = H^{\mathrm{CF}} - \frac{1}{2}\sum_{ij} J_{ij}\vec{\mathrm{j}}_i \cdot \vec{\mathrm{j}}_i . \tag{1}$$

Here H^{CF} is the crystal-field Hamiltonian, and $\vec{\mathrm{j}}_i$ is the total-angular-momentum operator for the ith atom. J_{ij} is the effective exchange; translational invariance dictates $J_{ij}=J(R_i-R_j)$. We will assume that the coupling is ferromagnetic, $J_{ij}>0$.

We will consider a very simple version of (1), with only two-crystal-field states on each site; call them $|0_i\rangle$ and $|1_i\rangle$. It is then convenient to express H in terms of Pauli spin operators defined on the space spanned by these two states. Taking the zero of energy at the center of the crystal-field gap, we have

$$H^{\mathrm{CF}} = \tfrac{1}{2}\Delta \sum_i \sigma_i \tag{2}$$

for splitting Δ. We assume that both levels are singlets, $\langle 0_i|\vec{\mathrm{j}}_i|0_i\rangle = \langle 1_i|\vec{\mathrm{j}}_i|1_i\rangle = 0$, but that the two levels are coupled by a matrix element of j_i^x,

$$\langle 1_i|j_i^x|0_i\rangle = M \neq 0 . \tag{3}$$

We assume this is the only nonvanishing matrix element. In terms of the crystal-field-state space

pseudospin operators,

$$j_i^z = M\sigma_i^x , \tag{4}$$

and we can write the Hamiltonian as an Ising model (of the pseudospins, not the real spins) in a transverse field:

$$H = \tfrac{1}{2}\Delta \sum_i \sigma_i^z - \tfrac{1}{2}M^2 \sum_{ij} J_{ij}\sigma_i^x\sigma_j^x \tag{5}$$

$$= \tfrac{1}{2}\Delta\sigma^z(q=0) - \tfrac{1}{2}M^2 \sum_q J(q)\,|\sigma^x(q)|^2 , \tag{6}$$

where the second expression is written in terms of Fourier-transformed pseudospin operators.

In order to write a free-energy functional in a form convenient for our purposes, we transform the partition function derived from the Hamiltonian of Eq. (5) following the Stratonovich-Hubbard procedure, writing it as a Gaussian functional average[20,21]:

$$Z = \int D\xi \exp\left(-\sum_i \frac{1}{\beta}\int_0^\beta dt\, \xi_i^2(t)\right) Z[\xi] , \tag{7}$$

where

$$Z[\xi] = \mathrm{tr}\left[e^{-\beta H_{CF}} T \exp\left(-\int_0^\beta dt \sum_{ij} \xi_i(t) A_{ij}\sigma_j^x(t)\right)\right]$$

$$= Z_{CF}\left\langle T\exp\left(-\int_0^\beta dt \sum_{ij}\xi_i(t)A_{ij}\sigma_i^x(t)\right)\right\rangle_{CF} . \tag{8}$$

Here Z_{CF} is the crystal-field partition function, and the notation $\langle\,\rangle_{CF}$ indicates an average in the presence of the crystal-field Hamiltonian alone:

$$\langle B\rangle_{CF} = \mathrm{tr}(e^{-\beta H_{CF}}B)/Z_{CF} . \tag{9}$$

$\sigma_j^x(t)$ is the interaction-picture operator:

$$\sigma_j^x(t) = e^{tH_{CF}}\sigma_i^x e^{-tH_{CF}} . \tag{10}$$

The kernel A_{ij} is a matrix defined by the interaction constants

$$(A^2)_{ij} = 2M^2 J_{ij}/\beta . \tag{11}$$

The $\xi_i(t)$ play the role of an arbitrary time- and spin-dependent field in the x direction. They correspond to the order parameter of the system in the usual way: The various moments of their distribution correspond to the various thermally averaged spin correlation functions of the full interacting systems. It will sometimes simplify notation to express quantities in terms of

$$h_i(t) \equiv \sum_i A_{ij}\xi_i(t) , \tag{12}$$

which has the dimension of energy (or field in our units) and whose Fourier transform

$$h_q(t) = [2M^2J(q)/\beta]^{1/2}\xi_q(t) . \tag{12'}$$

If we write the functional integral in the form

$$Z = Z_{CF}\int D\xi e^{-\Psi[\xi]} , \tag{13}$$

it is possible to write down directly from (8) a formal expression for the free-energy functional $\Psi[\xi]$ in terms of the fields $\xi_i(t)$ and the dynamic correlation function of the $\sigma_i^x(t)$ thermally averaged with respect to the crystalline-field Hamiltonian. We have

$$\Psi[\xi] \equiv \sum_i \int_0^\beta dt\,|\xi_i(t)|^2 - \ln Z[\xi] = \sum_i \int_0^\beta dt\,|\xi_i(t)|^2 - \ln\left\langle T\exp\left(\sum_{ij}\int_0^\beta dt\,\xi_i(t)A_{ij}\sigma_i^x(t)\right)\right\rangle_{CF}$$

$$= \sum_i \int_0^\beta dt\,|\xi_i(t)|^2$$

$$-\sum_s \frac{1}{s!}\sum_{\substack{i_1\cdots i_s\\ j_1\cdots j_s}} \int_0^\beta dt_1\cdots\int_0^\beta dt_s\;\xi_{j_1}(t_1)\cdots\xi_{j_s}(t_s)A_{i_1j_1}A_{i_2j_2}\cdots A_{i_sj_s}\langle\sigma_{i_1}^x(t_1)\sigma_{i_2}^x(t_2)\cdots\sigma_{i_s}^x(t_s)\rangle_{CF,c} . \tag{14}$$

The extra subscript c on the average denotes the cumulant. To low order, or in certain limiting regimes, one can proceed to evaluate the correlation functions; these, in fact, just describe the response of the crystal-field system to external perturbations. Note that we can describe the problem as a scalar field theory for the $\xi_i(t)$ with the vertices describing their interactions given by the spin correlation functions of the noninteracting system.

Given these vertices, one may write down the free-energy functional to any given order in the ξ fields. It is possible to emphasize various physical aspects of the fluctuations by choosing which terms of the expression are to be included and which are to be discarded. We shall in what follows retain all orders in the expansion, in an average way. This leads us to our generalized Hartree approximation, which we derive by a variational procedure.[22]

Equation (14) is an exact expression for the free-energy functional. It can be written more compactly if we define

$$\Gamma_k(i_1,\ldots,i_k,t_1,\ldots,t_k) = \sum_{\{j\}} A_{i_1 j_1}\cdots A_{i_k j_k}\langle T\sigma_{j_1}(t_1)\cdots\sigma_{j_k}(t_k)\rangle_{\mathrm{CF},c}\,. \tag{15}$$

Then, Fourier transforming with respect to spatial coordinates, as well as with respect to the t_k $[t_k \in (0,\beta)]$, (14) becomes

$$\Psi[\xi] \equiv \sum_q |\xi_q|^2 - \sum_n \sum_{\{q\}} \delta(q_1+q_2+\cdots q_n) \times \Gamma_n(q_1,\ldots,q_n)\xi_{q_1}\xi_{q_2}\cdots\xi_{q_n}\,. \tag{16}$$

[We use the four-vector notation $q \equiv (\vec{q}, i\omega_m)$; $\omega_m \equiv 2\pi m/\beta$.]

III. PROPERTIES OF THE FREE-ENERGY FUNCTIONAL

Let us first consider the free-energy functional we would have if we took the true $\Psi[\xi]$ and kept only terms of order ξ^2.

$$\Psi_2[\xi] \equiv \sum_q |\xi_q|^2 - \sum_q \Gamma_2(q,-q)|\xi_q|^2\,, \tag{17}$$

where $\Gamma_2(q_1 - q)$ is the Fourier transform of

$$\Gamma_2(i_1,i_2;t_1t_2) = \frac{1}{2}\sum_{j_1 j_2} A_{i_1 j_1}A_{i_2 j_2}[\langle T\sigma^x_{j_1}(t_1)\sigma^x_{j_2}(t_2)\rangle_{\mathrm{CF}} - \langle\sigma^x_{j_1}(t_1)\rangle_{\mathrm{CF}}\langle\sigma^x_{j_2}(t_2)\rangle_{\mathrm{CF}}]\,. \tag{18}$$

(The form of H^{CF} ensures that the correlation functions are proportional to $\delta_{j_1 j_2}$ and functions of $t_1 - t_2$ only). One finds

$$\Gamma_2(q,-q) = J(\vec{q})\chi_0(i\omega_m) = J(\vec{q}) \times 2M^2\Delta\frac{\tanh(\beta\Delta/2)}{\Delta^2 - (i\omega_m)^2}\,. \tag{19}$$

That is, χ_0 is just the single-ion susceptibility in the absence of exchange.

We can evaluate the partition function explicitly in this approximation because the functional integral is Gaussian:

$$Z = \int \prod_q \frac{d\xi_q}{\sqrt{\pi}} \exp\left(-\sum_q [1 - J(\vec{q})\chi_0(q)]|\xi_q|^2\right), \tag{20}$$

so that

$$\ln Z = -\frac{1}{2}\sum_q \ln[1 - J(\vec{q})\chi_0(q)]\,. \tag{21}$$

Furthermore the susceptibility is given in terms of the mean-square fluctuations of the field ξ,

$$\chi(\vec{q}, i\omega_m) = [2/J(\vec{q})](\langle|\xi_q|^2\rangle - \tfrac{1}{2})\,, \tag{22}$$

so that we have

$$\chi_{\mathrm{RPA}}(\vec{q}, i\omega_m) = \chi_0(q)/[1 - J(\vec{q})\chi_0(q)]\,. \tag{23}$$

This will be recognized as just the result of standard RPA.[23] We find, in particular, the excitation spectrum from the poles of the denominator

$$\omega(\vec{q}) = \Delta\left[1 - \frac{2M^2J(\vec{q})}{\Delta}\tanh\left(\beta\frac{\Delta}{2}\right)\right]^{1/2}, \tag{24}$$

giving the well-known soft-mode behavior at the transition and the RPA critical value of coupling constant $2M^2J(0)/\Delta > 1$, for which the susceptibility can diverge at finite temperatures.

Equations (17) and (18) relate the RPA propagator to the second functional derivative of $\Psi[\xi]$ evaluated when all ξ_q vanish, and express this quantity in terms of a single-ion susceptibility. It will be of use in subsequent discussions to recognize that the second functional derivative of Ψ, evaluated for an arbitrary field configuration $\xi_i(t)$, is related in the corresponding way to a single-ion susceptibility in the presence of that field:

$$\frac{\delta^2\Psi[\xi]}{\delta\xi_i(t)\,\delta\xi_j(t')} = 2\delta_{ij}\delta(t-t') - 2\sum_{lm} A_{il}A_{jm}[\langle T\sigma^x_l(t)\sigma^x_m(t')\rangle_\xi - \langle\sigma^x_l(t)\rangle_\xi\langle\sigma^x_m(t')\rangle_\xi]\,. \tag{25}$$

The subscript ξ on the averages indicates the expectation value in the presence of $\xi_i(t)$ or, more precisely, the magnetic fields $h_j(t) = \sum \xi_i(t)A_{ij}$ (in the x direction).

The RPA describes what we can think of as free magnetic excitons. We now proceed to examine the higher-order terms in $\Psi[\xi]$ and see what they tell us about the anharmonic coupling between the exciton modes. In a field-theoretical language, we are looking at the bare connected n-point functions, $\Gamma_n(q_1,\ldots,q_n)$ (for $n \geq 4$). We can represent them diagrammatically as circles with n external exciton lines entering or leaving, as shown in Fig. 1. In order to get an idea of the effects of these vertices, we can evaluate them in the limit where all the q_i vanish. It is simple to get an expression for the Γ_n in this limit, and if we substitute these $\Gamma_n(0,\ldots,0)$ in place of $\Gamma_n(q_1,\ldots,q_n)$ in (16), we obtain an approximate *local* field theory with interactions of all orders in ξ. In our calculations in this paper we shall actually do some-

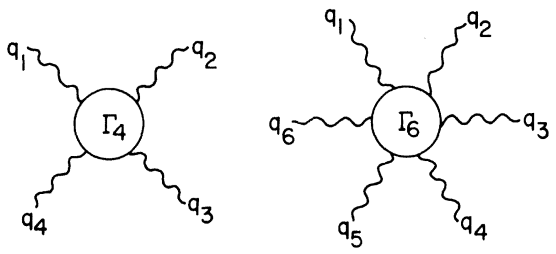

FIG. 1. Diagrammatic representation of exciton-exciton interaction vertices Γ_n.

what better than this approximation and retain some of the q dependence of the Γ_n. But as in the itinerant magnetic case, the local-interaction approximation does serve to illustrate much of the essential physics of the fluctuations. The reason is that since the Γ_n are correlation functions in the presence of the crystal field only, they should vary with frequency on a scale of Δ, and since their spatial variation comes in through the A_{ij}'s of (15), they should vary with wave vector on the scale of r_0^{-1}, the inverse range of the exchange. So if we will primarily be interested in fluctuations whose characteristic frequencies are small relative to Δ and wavelengths long compared with r_0, we may as well replace any $\Gamma_n(q, \ldots, q_n)$ by $\Gamma_n(0, \ldots, 0)$. High-q and -ω divergences are avoided by a cutoff at $\omega \approx \Delta$ and $q \approx r_0^{-1}$. (Actually, this argument is somewhat weaker in the present problem than in the Hubbard-model case, since it turns out that there are anomalous parts of the Γ_n proportional to δ functions of the sums of several of the q_i. These are exponentially weak at low temperature, however, and we shall ignore them in this paper. A discussion of their effects will be given in a subsequent paper.)

Let us now turn to the problem of evaluating the $\Gamma_n(0, \ldots, 0)$. One formal way involves using any of several bilinear fermion representations for the spin operators and evaluating the corresponding contracted $2n$-point fermion Green's functions.[24,25] Because the fermions, unlike the spin operators, have canonical commutation relations, this procedure is straightforward, if tedious. We used such a technique in the Hubbard-model problem (although there the fermions were the real fermions of the metal, not just something introduced for mathematical convenience). However, within the approximations we use in this paper, we do not need to introduce such complications in the present problem.

If the Γ_n ($n \geq 4$) are approximated by their static, uniform limits, expression (16) for Ψ can be written as

$$\Psi[\xi] = \sum_q [\Gamma_2(0,0) - \Gamma_2(q,-q)] |\xi_q|^2 + \frac{1}{\beta} \sum_i \int_0^\beta dt\, \Phi(\xi_i(t)), \tag{26}$$

where the "potential" function Φ is

$$\Phi(\xi) = \xi^2 - \beta \sum_{n=2}^{\infty} \Gamma_n(0, \ldots, 0) \xi^n. \tag{27}$$

Knowing this function, of course, is equivalent to knowing all of the coefficients $\Gamma_n(0, \ldots, 0)$. The form (26) separates $\Psi(\xi)$ into a local piece (the second term) which tells us the cost in free energy of a uniform, static fluctuation of magnitude ξ, and a nonlocal part (the first term) which expresses the free-energy cost of varying ξ in space and time. This "kinetic" term is quadratic in the deviation of ξ from uniformity; a small-q and -ω expansion of (19) produces leading terms of order $\vec{q}^2$ and $q_0^2 = \omega_m^2$. If we were to terminate the expansion at this point, (26) would look formally like the generalized Landau-Ginzburg functional used by Wilson,[26] except that frequency appears on an equal footing with wave vector.[27] Here, however, we keep terms of all powers of $|\vec{q}|^2$ and ω_m^2.

As in many other problems[16,21,26] we can learn a lot about the character of the fluctuations by examining the shape of the potential Φ. To find this function, we ask first about its second derivative Φ'', which turns out to be very easy to calculate. We can then unambiguously construct Φ, since we (conventionally) take $\Phi(0) = 0$ and symmetry rules out any term linear in ξ. Φ'' is easy to calculate because of what we noticed above about the second functional derivative of Ψ: It was simply expressed in terms of the susceptibility of the noninteracting system in the presence of the external field $\xi_i(t)$, Eq. (25). Therefore, since Φ is the value of the general functional Ψ when ξ is static and uniform, Φ'' is simply expressible in terms of the $q = 0$ susceptibility in the presence of a uniform, static external field. Unlike the general expression (25), this susceptibility is easy to calculate, and when we have it, we simply have to integrate twice to get $\Phi(\xi)$. It is this fact that makes the local-interaction approximation so simple to discuss, in contrast to the intractable full problem, which requires the impossible general evaluation of expression (14) or (25).

We therefore digress to make a calculation of the susceptibility of a single ion in the presence of a fixed external field $h = A(0)\xi$. For discussing the shape of the potential, we will actually need to know this susceptibility $\chi_0(q, h)$ only in the limit $q = 0$, but we make the calculation for general q for later reference. The Hamiltonian in the presence of the external field is

$$H = +\tfrac{1}{2}\Delta\sigma^z - h\sigma^x \tag{28}$$

and it is χ_0^{xx} that we want to calculate. The problem is solved by rotating the coordinate axis through an angle $\theta = \tan^{-1}(2h/\Delta)$ around the y axis, diagonalizing H:

$$H = \tfrac{1}{2}\tilde{\Delta}(h)\tilde{\sigma}^z, \tag{29}$$

with

$$\tilde{\Delta}^2(h) = \Delta^2 + (2h)^2. \tag{30}$$

Now in terms of the susceptibilities $\tilde{\chi}_0^{xx}$ and $\tilde{\chi}_0^{zz}$ in the rotated system, the x susceptibility in the unrotated system is

$$\chi_0^{xx} = \cos^2\theta\,\tilde{\chi}_0^{zz} + \sin^2\theta\,\tilde{\chi}_0^{xx} \tag{31}$$

and we can evaluate $\tilde{\chi}_0^{zz}$ and $\tilde{\chi}_0^{xx}$ straightforwardly. For the x component, we have the same problem as in (19) except that Δ has been increased to $\tilde{\Delta}$:

$$\tilde{\chi}_0^{xx}(i\omega_m) = \frac{2\tilde{\Delta}(h)}{\tilde{\Delta}^2(h) - (i\omega_m)^2}\tanh[\tfrac{1}{2}\beta\tilde{\Delta}(h)]. \tag{32}$$

For the z component, note that we are applying a perturbation that commutes with H, so there is no response except at zero frequency. There we can use the classical expression

$$\begin{aligned}\tilde{\chi}_0^{zz}(i\omega_m) &= \beta[\langle(\tilde{\sigma}^z)\rangle - \langle\tilde{\sigma}^z\rangle^2]\delta_{m0} \\ &= \beta\{1 - \tanh^2[\tfrac{1}{2}\beta\tilde{\Delta}(h)]\}\delta_{m0}.\end{aligned} \tag{33}$$

This is just the Curie law for a finite field.

Combining (31)–(33) we obtain

$$\begin{aligned}\chi_0^{xx}(i\omega_m, h) = &\frac{2\Delta^2}{\tilde{\Delta}(h)}\frac{\tanh[\tfrac{1}{2}\beta\tilde{\Delta}(h)]}{\tilde{\Delta}^2(h) - (i\omega_m)^2} \\ &+ \delta_{m0}\frac{4\beta h^2}{\tilde{\Delta}^2(h)}\operatorname{sech}^2[\tfrac{1}{2}\beta\tilde{\Delta}(h)].\end{aligned} \tag{34}$$

The second term is related to the sort of anomalous, singularly frequency-dependent term which, as we mentioned earlier, could occur in the exact expressions for the Γ_n. One can see explicitly here that for $T \ll \Delta$, this term is of relative order $e^{-\Delta/T}$; we will ignore it in this paper. Then we obtain an expression for Φ'' from the static limit of (34):

$$\begin{aligned}\Phi''(\xi) &= 2 - \beta A^2(0)\chi_0^{xx}(0, h) \\ &= 2\left\{1 - \frac{2J(0)M^2\Delta^2}{[\Delta^2 + 8J(0)M^2\xi^2/\beta]^{3/2}}\right. \\ &\quad \left.\times\tanh\left[\tfrac{1}{2}\beta\left(\Delta^2 + \frac{8J(0)M^2\xi^2}{\beta}\right)^{1/2}\right]\right\}.\end{aligned} \tag{35}$$

The result of integrating (35) twice is shown in Fig. 2. Above the mean-field transition temperature T_c^{MF}, where the RPA exciton frequency $\omega(0)$, Eq. (24), goes to zero, Φ has a single minimum

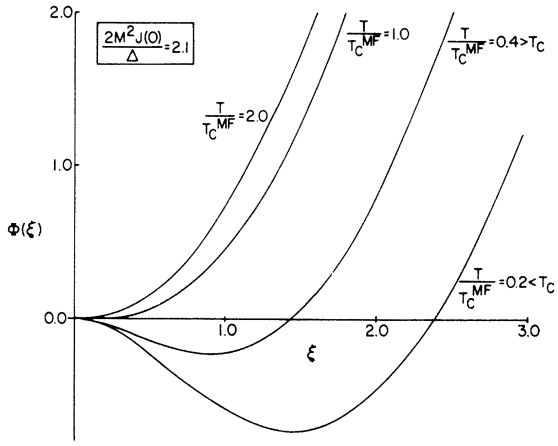

FIG. 2. Shape of the potential Φ.

which flattens out as one approaches T_c^{MF} and the quadratic term goes to zero. Below T_c^{MF}, Φ has a double-well structure. This is the usual sort of behavior for systems undergoing a second-order phase transition.[26]

In the sense of knowing this function, then, we know all the exciton-exciton coupling vertices Γ_n; they are just proportional to the derivatives of Φ at $\xi = 0$. We would then think about doing a diagrammatic perturbation theory for the free energy or for the susceptibility. [It can be derived by expanding the integrand of the functional integral around the RPA (Gaussian) expression from (20).] In calculating χ, we would then generate a set of self-energy diagrams, a few of which are shown in Fig. 3. The single-loop diagram of Fig. 3(a) is the Hartree contribution to the exciton self-energy from the Γ_4 interaction. There are higher-order diagrams in Γ_4, the first of which is that of Fig. 3(b). In this problem, we also have higher-order bare vertices Γ_6, Γ_8, and so on. Some Hartree diagrams for these parts of the interaction are shown in Fig. 3(c). There are, of course, higher-order diagrams in all these Γ_n as well, and diagrams with Γ_n's of differing n's in them, some of which are shown in Fig. 3(a).

In this paper, we will treat all the parts of the interaction in Hartree approximation. That is, we will include all diagrams like those of Figs. 3(a) and 3(c), but none of the higher-order (collisional) diagrams of Figs. 3(b) and 3(d). By omitting any collisional diagrams, we preclude any accurate treatment of the $T \to \infty$ limit.[28] However, this is the simplest approximation one can make for a nonpolynomial interaction such as we have in (27), and that is why we devote this paper to examining its content and consequences.

Stinchcombe[14] has made a Hartree approximation in which only Γ_4 is taken into account, but

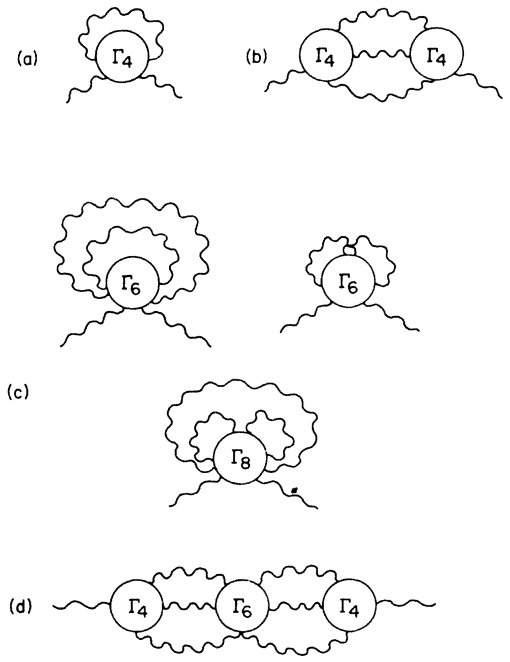

FIG. 3. Exciton self-energy diagrams. (a) Hartree diagram for Γ_4 interaction. (b) Higher-order diagram involving Γ_4. (c) Some Hartree diagrams for Γ_6 and Γ_8 couplings. (d) A higher-order diagram involving Γ_4 and Γ_6.

its frequency and wave-vector dependences are retained. (We simulate this behavior in our calculations below with a large-q cutoff.) Bak's approximation for a related model[15] is analogous to Stinchcombe's.

We have given this discussion of the local interaction theory (26) mainly to illustrate qualitatively the physical content of the full theory. As we noted previously we will be able to do this problem in an approximation slightly better than a completely local one. We shall still be working with a Hartree theory, however, in the sense of retaining only the particular class of diagrams described above. The improvement will be to retain some of the q dependence of the Γ_n; in particular, the dependence of Γ_n on the *external* exciton lines in each self-energy diagram. That is, in the Hartree diagram of Fig. 3(a) or 3(c), the shaded circle stands for $\Gamma_n(0,\ldots,0,q,0,\ldots,0,-q,0,\ldots,0)$, where q is the four-momentum of the external line, rather than $\Gamma_n(0,\ldots,0)$, as in the completely local approximation. This approximation is clearly sensible in that the effects of the dependence Γ_n on internal exciton four-momenta are washed out by the integration over these momenta, while the dependence on the external variables, which are not integrated over, affects the self-energy directly.

Keeping track of the combinatorics in this direct diagrammatic approach is somewhat messy, although the problem can be done this way. However, there is an easier way to do the calculation for a Hartree approximation which avoids these complications, and we shall follow it in Sec. IV. The preceding description of the diagrammatic approach is therefore, as far as this paper is concerned, merely an aid to understanding physically just what processes are included in the approximation.

IV. HARTREE APPROXIMATION

When one uses the term Hartree (or Hartree-Fock) approximation in elementary quantum mechanics, one refers to a variationally optimal approximation of a Hamiltonian which includes interparticle interactions by a self-consistent one-body Hamiltonian. In the field theory we have here, the counterpart of the Hamiltonian is the free-energy functional $\Psi[\xi]$ and the fields $\xi_i(t)$ play the role analogous to the single-particle fields. Hence the quadratic term in Ψ is like a "one-body" Hamiltonian and the quartic and higher-order terms are the analogs of interaction terms in H. A Hartree (-Fock) approximation for this sort of problem then involves the following question: What is the best (in the variational sense) *quadratic* functional $\Psi_0[\xi]$ with which I can approximate $\Psi[\xi]$? We therefore make a parametrization

$$\Psi_0[\xi] = \sum_q a(q)\,|\xi(q)|^2 , \tag{36}$$

where the a's are to be determined variationally, as follows.[22]

If we define the average in the approximate distribution $e^{-\Psi_0[\xi]}$ of a quantity $f[\xi]$ by

$$\langle f[\xi]\rangle_0 = \frac{1}{Z_0}\int D\xi\, e^{-\Psi_0[\xi]} f[\xi] , \tag{37}$$

where

$$Z_0 = \int D\xi\, e^{-\Psi_0[\xi]} , \tag{38}$$

we can obtain an inequality relating the true and approximate free energies:

$$\beta F = -\ln Z \leq -\ln Z_0 + \langle \Psi[\xi] - \Psi_0[\xi]\rangle_0 . \tag{39}$$

Minimizing the right-hand side with respect to $a(q)$ then leads to the condition

$$a(q) = \frac{1}{2}\left(\frac{\partial^2 \Psi[\xi]}{\partial \xi(q)\,\partial \xi(-q)}\right) . \tag{40}$$

We are thus faced with the problem of estimating the averaged second derivative of the *true* free-energy functional using the probability distribution defined by the *trial* functional $\Psi_0[\xi]$.

As we noted earlier, Eq. (25) in the present

problem, the second derivative which appears inside the average in (40) is simply related to the susceptibility of a single ion in a field h, which is proportional to ξ, and this cannot be calculated in general. However, as we also noted, this susceptibility *can* be easily calculated (for general q) if ξ is uniform and static. We therefore make the approximation of replacing the χ_0 in the presence of the arbitrary h by the one in the presence of the uniform, static h. In terms of the diagrammatic description of Sec. III, the replacement of the arbitrary h by the uniform one is equivalent to the neglect of the dependence of the vertices on the internal four-momenta.

To see this point pictorially we give a diagrammatic version of the arguments leading to (40).[16] We can represent the functional $\Psi[\xi]$ by the series of diagrams of Fig. 4(a). Differentiating a diagram once with respect to ξ_q then removes a dotted line (and the factor $1/n$, because any of the n lines could be removed), as in Fig. 4(b). A second differentiation then removes any of the remaining $n-1$ dotted lines, so the diagrams for the second derivative $\partial^2\Psi/\partial\xi_q\partial\xi_{-q}$ look like Fig. 4(c). They are the diagrams one would draw for χ_0 in an external field (specified by the unremoved dotted lines). The averaging over a Gaussian distribution of these external fields then leads to terms which can be represented by tying together in pairs the remaining dotted lines, as in Fig. 4(d). The pairs of dotted lines so tied then stand for averages $\langle\xi_{q_i}\xi_{-q_i}\rangle$. But these diagrams look just like and are evaluated in the same way as those of Figs. 3(a) and 3(c). Therefore when we evaluate the average χ_0 as if ξ were uniform in space and time, we are doing a calculation of a diagram [of Fig. 4(c)] in which all the ξ_{q_i} have $q_i=0$; the vertex is evaluated as if all internal momentum transfers vanished, although the external ξ (with respect to which we differentiated) is allowed to have a finite q. This establishes the equivalence of the diagrammatic prescription of approximating the vertex by its value when internal momentum transfers vanish to the analytic prescription of calculating χ_0 in a uniform field, and averaging over the Gaussian distribution of fields $e^{-\Psi_0[\xi]}$.

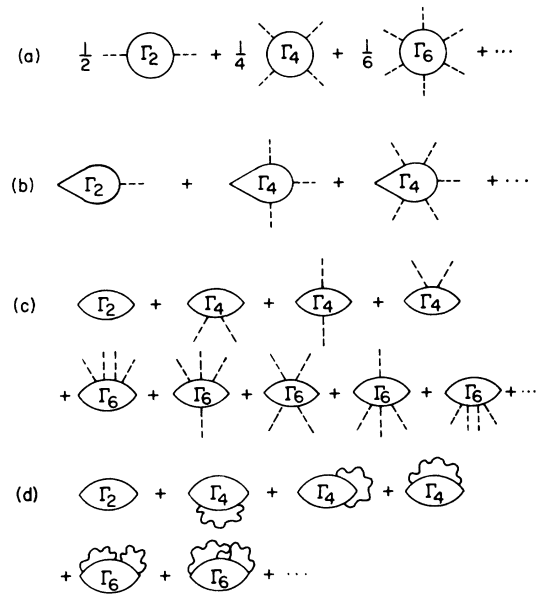

FIG. 4. Diagrammatic representation of double differentiation and averaging of the functional Ψ. (a) Ψ itself (omitting the ξ^2 term from the Gaussian integral). The dotted lines stand for ξ fields. (b) Result of one differentiation of the diagrams of (a). (c) Result of second differentiation of Ψ. (d) Averaging of (c), given that ξ is normally distributed. The wavy line stands for $\langle\xi_{q\omega}\xi_{-q-\omega}\rangle$, where q,ω is the four-momentum it carries.

The single-ion susceptibility in a uniform, static external field was calculated above (34), so the variational coefficients $a(q)$ can be written as

$$a(q)=1-J(q)\varphi(i\omega_m)\,, \tag{41}$$

where

$$\varphi(i\omega_m)=2M^2\Delta^2\left\langle\frac{1}{\Delta^2+4h^2-(i\omega_m)^2}\times\frac{\tanh[\frac{1}{2}\beta(\Delta^2+4h^2)^{1/2}]}{(\Delta^2+4h^2)^{1/2}}\right\rangle_0 . \tag{42}$$

The averaging is now simply over the distribution of *local* fields

$$h_i(t)=\frac{1}{N^{1/2}}\sum_{qm}e^{i\vec{q}\cdot\vec{r}_i-i\omega_m t}h_{qm}$$

whose variance is

$$\sigma^2=\frac{1}{N}\sum_{qm}\langle|h_{qm}|^2\rangle=\frac{2M^2}{\beta}\sum_{qm}J(q)\langle|\xi_{qm}|^2\rangle\,. \tag{43}$$

{This distribution is Gaussian because $h_i(t)$ is a sum of fields h_{qm} which, by our hypothesis about the form of $\Psi_0[\xi]$ [Eq. (36)], are independently (and normally) distributed.} In the limit of small fluctuations ($\sigma^2\to0$), $\varphi(i\omega_m)$ just reduces to $\chi_0(i\omega_m)$ and the RPA, Eq. (20), is recovered. Our result is also quite analogous to what we found for the Hubbard model, where the Lindhard function was averaged over a distribution of magnetic fields. In that problem, placing a cutoff at some q_c on the sum of q in (43) was crudely equivalent to assigning a finite range of $1/q_c$ to the forces coupling different fluctuation modes. Here, the q dependence

of $J(q)$ provides such a cutoff explicitly, and we see that the fluctuation effects are suppressed if the exchange has long range.

Writing the average in (43) explicitly,

$$\varphi(i\omega_m) = \int \frac{dh}{(2\pi\sigma^2)^{1/2}} \exp\left(-\frac{1}{2}\frac{h^2}{\sigma^2}\right) \frac{\Delta^2}{(\Delta^2+4h^2)^{1/2}} \times \frac{2M^2}{\Delta^2+4h^2-(i\omega_m)^2} \tanh[\tfrac{1}{2}\beta(\Delta^2+4h^2)^{1/2}] . \tag{44}$$

We see that the δ functions in the imaginary part of χ_0, Eq. (19), are broadened out into continua with singular thresholds at $\omega = \pm\Delta$:

$$\mathrm{Im}\varphi(\omega+i\delta) = \theta(|\omega|-\Delta)(\pi/2\sigma^2)^{1/2} \times \frac{M^2\Delta^2 \tanh\frac{1}{2}\beta\omega}{|\omega|(\omega^2-\Delta^2)^{1/2}} \exp\left[-\frac{1}{2}\left(\frac{\omega^2-\Delta^2}{4\sigma^2}\right)\right] . \tag{45}$$

The corresponding correction to the real part of φ can be written simply as

$$\mathrm{Re}\varphi(\omega) = 2M^2(\pi/2\sigma^2)^{1/2} \times \mathrm{P}\int_1^\infty \frac{dx}{\pi} \frac{\exp[-\frac{1}{2}(\Delta^2/4\sigma^2)(x^2-1)]\tanh(\frac{1}{2}\beta\Delta x)}{[x^2-(\omega/\Delta)^2](x^2-1)^{1/2}} . \tag{46}$$

The sharpness of the singularity at Δ in $\chi_0(\omega)$ is reduced, but $\mathrm{Re}\varphi$ still diverges like $|\omega-\Delta|^{-1/2}$ near Δ. Figure 5 shows plots of the imaginary and real parts of $\varphi(\omega)$ for a case where σ is comparable to Δ.

This $\varphi(\omega)$ is then substituted back into (41), from which we can calculate the mean-square fluctuations of the ξ field,

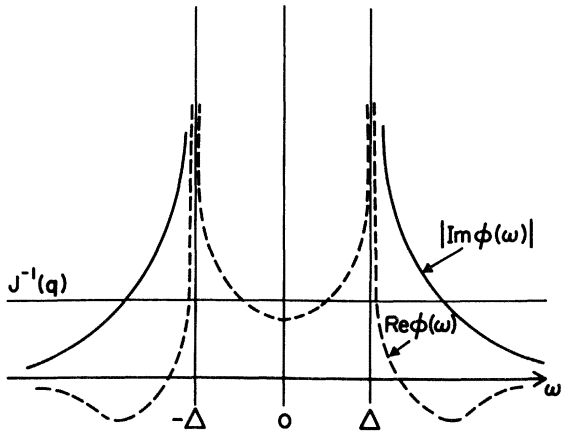

FIG. 5. Real (dashed line) and imaginary (solid line) parts of $\varphi(\omega)$. Quasiparticle excitations occur when $\mathrm{Re}\varphi$ crosses the horizontal line at a height $J^{-1}(q)$.

$$\langle|\xi(q)|^2\rangle = \int \frac{D\xi\,|\xi(q)|^2 e^{-\Psi_0[\xi]}}{\int D\xi\, e^{-\Psi_0[\xi]}} = \tfrac{1}{2}[1-J(q)\varphi(i\omega_m)]^{-1} , \tag{47}$$

and hence from (43)

$$\sigma^2 = \frac{M^2}{\beta N}\sum_{qm} J(q)[1-J(q)\varphi(i\omega_m)]^{-1} . \tag{48}$$

Self-consistency then demands that this σ^2 be the same one which went into calculating $\varphi(i\omega_m)$ [Eq. (44)].

The full dynamical susceptibility is obtained from (22),

$$\chi(q, i\omega_m) = \varphi(i\omega_m)/[1-J(q)\varphi(i\omega_m)] . \tag{49}$$

Its imaginary part (which is what is measured in a neutron scattering experiment) is nonvanishing in the region above Δ (or below $-\Delta$) where $\mathrm{Im}\varphi(\omega)$, Eq. (45), is nonvanishing. It will also have δ-function parts at frequencies $\omega(q)$ where the denominator vanishes, i.e., $\mathrm{Re}\varphi(\omega(q)) = 1/J(q)$ and $\mathrm{Im}(\omega(q)) = 0$. The spectral weight in the spikes is small if $\omega(q)$ is near Δ, and increases with decreasing $\omega(q)$. The strength of the spike is given by

$$z_q = \frac{\pi\varphi^2(\omega(q))}{|\partial\varphi(\omega)/\partial\omega|_{\omega=\omega(q)}} . \tag{50}$$

(Since $\mathrm{Re}\varphi$ is an even function, the spike in $\mathrm{Im}\chi$ would have infinite strength in the limit $\omega(q)\to 0$ if it occurred.) Figure 5 shows that these sharply defined excitations always exist, and that their energies decrease as J or φ is increased. Increasing φ corresponds, of course, to lowering the temperature, as is evident from the tanh factor in (42).

In a previous report of this work,[19] we obtained a small elastic ($\omega = 0$ only) central peak in φ and χ. This was because in calculating the average of χ_0 in an external field (34), we retained the second term proportional to $\delta_{m0}\,\mathrm{sech}^2(\frac{1}{2}\beta\tilde{\Delta})$. Therefore, we found a static χ which exceeded the limit of the finite-ω susceptibility as $\omega\to 0$; consequently, the static χ would diverge (as T was lowered) and the phase transition take place before the $q = 0$ exciton had gone soft. Appealing as this result might be, we no longer believe that it is sensible, for the following reason: It makes sense, in evaluating the average of χ_0 over the distribution of external fields, to pretend that the external field is static only if the exciton-exciton coupling vertices are slowly varying functions of frequency. But the form of the second term in (34) indicates that there are contributions to these vertices with a very singular dependence on the external exciton frequency, for fixed (zero, in this case) internal frequency. We therefore expect these vertices to depend singularly on internal frequencies as well,

and this dependence invalidates the approximation of slow frequency dependence we made in each diagram. In this paper we restrict our attention to temperatures well below the crystal-field splitting, in which limit the singular parts of the vertices are very small. In a subsequent paper we shall direct our attention to the interesting anomalous damping effects which these terms can produce.[14,15]

V. NUMERICAL CALCULATIONS

In making some quantitative estimates of how our calculated excitation spectrum might be observably different from RPA, we have adopted the static approximation, as we did in treating the Hubbard model. This means that all fluctuation modes with nonzero ω_m are ignored in calculating $\varphi(i\omega_m)$, so that Eq. (48) becomes

$$\sigma^2 = \frac{2M^2}{\beta N}\sum_q J(q)\langle|\xi(q)|^2\rangle = \frac{M^2}{\beta N}\sum_q \frac{J(q)}{1-J(q)\varphi(0)}\,. \tag{51}$$

This approximation can be expected to be reasonable whenever T is larger than the exciton energies $\omega(q)$ (provided most of the spectral weight in χ lies in the quasiparticle spike, and not in the continuum).

Structural effects in $J(q)$ are ignored here; we take

$$J(q) = \begin{cases} J(0)(1-q^2/q_0^2) & (q\in R_0) \\ -J(0)[1-(q-q_m)^2/q_0^2] & (q\in R_1)\,, \end{cases} \tag{52}$$

where R_0 and R_1 are spheres of radius q_0 centered about $q=0$ and $q_m \equiv (\pi/a, \pi/a, \pi/a)$, respectively. This agrees to lowest order in q with nearest-neighbor exchange in a cubic crystal, if $q_0 = \sqrt{6}/a$.

Figure 6 shows the critical temperature as a function of the dimensionless coupling parameter $2M^2J(0)/\Delta$, calculated by solving the equation $1 = J(0)\varphi(0)$ for $\beta = T_c^{-1}$, with σ^2 obtained from (51) using the above approximations:

$$\sigma^2 = \frac{4\pi M^2 T_c}{(2\pi)^3}\int_0^{q_0} q^2\,dq\; J(q)\left(\frac{1}{1-J(q)/J(0)} - \frac{1}{1+J(q)/J(0)}\right) = 0.433 M^2 T_c J(0)\,. \tag{53}$$

The RPA result

$$1 = [2M^2J(0)/\Delta]\tanh(\Delta/2T_c) \tag{54}$$

is also plotted, for comparison. As one might expect, the effect of fluctuations in the order parameter is to suppress T_c. Over the range of T_c most likely to be of experimental interest we see that our estimate of the coupling constant necessary for a given T_c is two to three times what one would obtain from RPA. The size of this reduction is, of course, sensitive to our cutoff procedure or, in a more realistic treatment, to the range of the exchange. Nevertheless, the result does show that fluctuation effects can be significant.

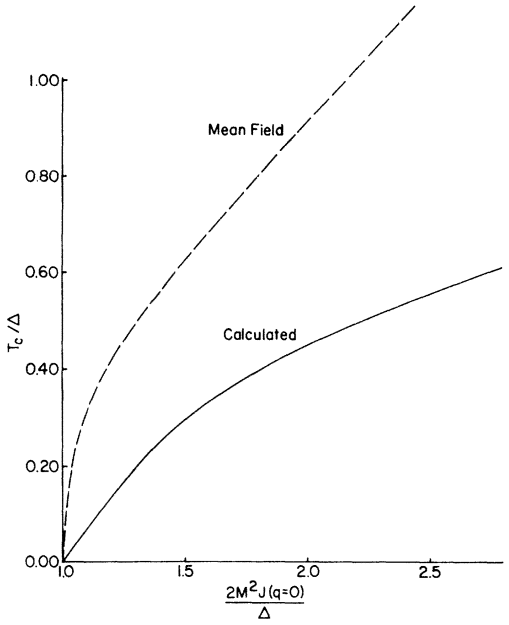

FIG. 6. Critical temperature T_c as a function of coupling, for static approximation (solid line) and RPA (dashed line).

If we tried to apply this approximation at lower temperatures, the fluctuation effects would become smaller, since Eq. (46) gives $\sigma^2 \propto T$. At $T=0$, we would be back to the RPA result, as is evident in the figure, where T_c goes to zero for the same value of the coupling in both cases. Of course, the static approximation cannot be trusted in this limit. A better calculation of T_c would be expected to lead to a higher threshold for magnetism, since the $m\neq 0$ terms in (48) would produce a nonzero σ^2 even at $T=0$.

We turn now to the excitation spectrum, which we have calculated for a case of moderate coupling: $2M^2J(0)/\Delta = 2.1$. T_c is calculated to be $\simeq 0.45\Delta$ (cf. the RPA T_c, which is 0.97Δ). The results are given in Fig. 7 for a range of temperatures between about $1.07T_c$ and $2.0T_c$. Note that the continuum part above the temperature-independent threshold Δ retains significant oscillator strength even close to T_c.

Such effects may be observable in experimental

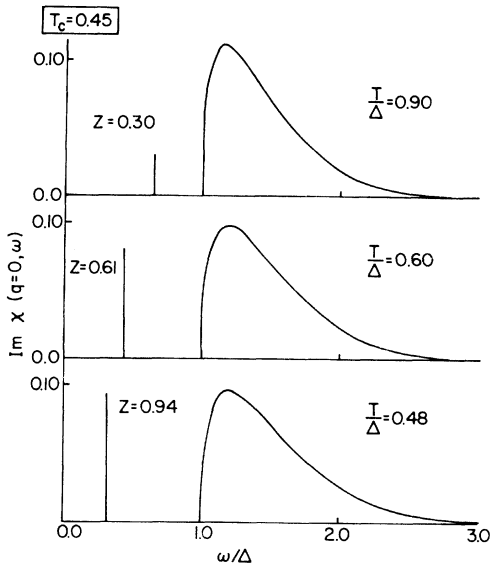

FIG. 7. Magnetic excitation spectrum $\mathrm{Im}\chi(0,\omega)$ for several values of $T > T_c = 0.45\Delta$, $2M^2J(0)/\Delta = 2.10$.

spectra and may even lead to reinterpretation of existing data. For example, it had been thought that observation of satellite peaks in neutron scattering experiments could be taken as evidence of interaction with higher-lying crystal-field levels.[11,12] Here we see that it might be a consequence of the continuum piece of $\mathrm{Im}\chi$ discussed here. It is also possible that at high temperatures, the continuum may obscure the spike part of the spectral weight function, making it difficult to follow as the temperature is lowered.

The somewhat unusual form of $\mathrm{Im}\chi$ can be traced through $\mathrm{Im}\varphi$ [Eq. (45)], to the broadening of the spike in the single-ion susceptibility into a continuum by the distribution of local fields. The broadening is asymmetric and the threshold is sharp because the gap in the presence of any given effective field h is larger than that for $h=0$. Any mechanism broadening the bare-crystal-field energies will also smear the magnetic excitations out into the gap, but as long as the intrinsic magnetic fluctuations of the system are the most important broadening mechanism, the structure of the excitation spectrum should remain largely as shown.

VI. CONCLUSIONS

We have presented here an attempt at estimating the effects of mode coupling in Van Vleck magnets, and seen that a Hartree-like decoupling scheme leads in a static approximation to qualitative deviations from the usual RPA excitation spectrum. In particular, we find, in addition to the RPA-like quasisoft mode, a broad contribution with a threshold fixed at the crystal-field energy. This effect arises from fluctuating hybridization of the pure-crystal-field levels. Numerical estimates indicate that it may be significant experimentally.

We have neglected a number of aspects of the problem which are important in some regimes. The fundamental decoupling scheme breaks down when the magnetic coupling constant approaches its critical value, and there one would have to resort to a version of Wilson's renormalization-group theory to describe the fluctuations.

It should also be mentioned that in a realistic analysis, the role of fluctuation effects in changing the relative contributions of higher-lying crystal-field levels to the instability is very difficult to estimate. We have neglected these considerations entirely here.

Another point concerns the role of crystal structure in determining the character of the transition. This enters both in the form of the crystal-field level scheme and in the range and anisotropy of the exchange coupling. The latter affects the fluctuations [Eq. (43)]. Although such structural effects have been neglected here, we might expect them to be of some interest, not only for crystal-field magnetic systems, but more particularly for certain classes of ferroelectrics, for which the same model Hamiltonian is routinely used.

A serious omission in our calculations is the neglect of finite frequency terms in the sum (48) determining σ^2. We can, however, give a qualitative discussion of what happens to σ^2 at low temperatures. We have

$$\sigma^2 = \frac{M^2}{\beta N}\sum_{qm}\frac{J(q)}{1-J(q)\varphi(i\omega_m)} = \frac{M^2}{\beta N}\sum_{qm}J(q)[1+J(q)\chi(q,i\omega_m)], \tag{55}$$

and since $\sum_q J(q) = J_{ii}$, the intra-atomic exchange, which we take to vanish, we can use the spectral representation of χ to obtain

$$\sigma^2 = \frac{M^2}{N}\sum_q J^2(q)\int\frac{d\omega}{2\pi}\chi''(q,\omega)\coth(\tfrac{1}{2}\beta\omega). \tag{56}$$

At high temperatures the integral just reduced to the static susceptibility $\chi(q)$ times the temperature, which is essentially the form we used in (51). When T is low compared to the frequencies where the spectral weight in χ'' is concentrated, however, the coth may be approximated by a signum function instead of $2/\beta\omega$. Further approximating χ'' by a spike at $\omega(q)$, we find

$$\sigma^2 = \frac{M^2}{2N}\sum_q J^2(q)\chi(q)\omega(q). \tag{57}$$

That is, $\frac{1}{2}\omega(q)$ replaced the temperature in (51).

Since most of the contribution to the sum in (56) comes from large q, a typical $\omega(q) \lesssim \Delta$. Another way to estimate σ^2 is to use the fluctuation-dissipation theorem to write (56) in terms of equal-time correlation functions,

$$\sigma^2 = M^2 \sum_{ijk} J_{ij} \langle j_j^z(0) j_k^z(0) \rangle J_{ki} . \tag{58}$$

Since $\chi(q) \simeq M^2\Delta/\omega^2(q) \simeq M^2/\Delta$, and the correlation function in (53) is of the order of M^2, (52) and (53) are consistent. Either way, we estimate the low-temperature σ^2 as around $M^4 J^2(0)$. The relevant dimensionless parameter for measuring the importance of the fluctuations is σ^2/Δ^2. We therefore expect a σ^2 of order unity when J is large enough to produce magnetic order, using the RPA criterion obtained from (23).

Study of all these problems necessitates some refinement of our rather crude approximations, and further work in this direction is in progress.

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[23]The equivalence of the result of expanding the free-energy functional only to second order in ξ to approximations which have historically been called RPA extends to many different problems, in particular the electron gas and the Hubbard model. The present problem seems somewhat exceptional in that the term RPA has been applied to a somewhat more elaborate approximation scheme (see, e.g., Cooper, Ref. 4) with a higher level of self-consistency. We adhere here to the pattern set in the other problems and use the term RPA to describe the non-self-consistent approximation of Eqs. (20)–(24).

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