# Critical and paramagnetic spin fluctuations in Heisenberg magnets

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(Received 4 April 1988)

The frequency- and wave-vector-dependent spin response function is calculated for a Heisenberg ferromagnet using the mode-coupling theory. Results are presented for the critical and paramagnetic regions. The work is motivated largely by recent, high-resolution neutron spectroscopy studies of EuO and EuS. In view of this, the Heisenberg model used incorporates nearest- and next-nearest-exchange interactions between spins on a face-centered-cubic lattice. The coupled-mode theory is shown to be successful in describing the measurements. Additional features are predicted as possible subjects of future experimental work. We present a review of various derivations of mode-coupling equations and demonstrate the equivalence of some prescriptions. The question of incorporating an external magnetic field in the theory is addressed, and we conclude that readily attainable fields have a minimal effect on the spin dynamics.

## I. INTRODUCTION

Recent high-resolution neutron-scattering experiments on simple insulating ferromagnets[1-6] and computer-simulation studies of Heisenberg magnets[7] have fuelled a resurgence of work on the theory of spin dynamics in Heisenberg magnets in the critical and paramagnetic regions.[8-17] The most promising theories are based on either a renormalization-group rationale[15-17] or the mode-coupling approximation.[10-14] Although the renormalization-group method is established for the critical region, it is plausible to extend it to high temperatures and wave vectors through educated guesses about the form of the scaling function.[17]

The mode-coupling theory, examined in this paper, is more obviously an approximate technique since one derivation, for example, uses a decoupling of the infinite-order equation of motion. However, in the critical region the theory is consistent with dynamic scaling which gives confidence in the quality of the approximation. There is, *a priori,* no reason to question the potential value of the theory outside the critical domain, i.e., at higher temperatures and short wavelengths. The motivation to explore the value of the theory in this region is twofold: first, to explore the range of validity of a long standing and generally successful theoretical approach that is used in a wide variety of physical problems, including localization,[18] the dynamical properties of liquids,[19] and the glass transition,[20] and second, to access the critical domain experimentally is very demanding, and the constraints are weaker in the paramagnetic phase. This comment is valid for both scattering experiments and computer simulations. The aim of the work reported here is to present a comprehensive account of predictions from the mode-coupling theory in the critical and paramagnetic region, i.e., for a wide range of the wave vectors and temperature. We incorporate an external magnetic field and predict that readily attainable fields have a minimal effect on spin dynamics.

The recent availability of samples of EuO and EuS with tolerable neutron characteristics is largely responsible for the spate of experimental work that has been a major motivation factor in undertaking the present study. There is a controversy on the form of the measured response in the vicinity of the critical region,[21] and the wave vector dependence of the damping of the collective excitations in particular. It is well established that a collective mode persists at the zone boundary in a large range above the critical temperature,[1,3,4] albeit strongly damped. Long-wavelength fluctuations in the paramagnetic phase have a diffusive character.[2,4-6] Hence, there is a temperature-dependent wave vector that can be assigned to the crossover from collective to diffusive behavior, and the experimental determination of said wave vector is a vexed question. Our results should help to focus future investigation.

The Heisenberg model used is believed to be a realistic description of EuO and EuS. Spins arranged on a face-centered-cubic lattice are coupled by nearest- and next-nearest-exchange interaction.[22] For EuS the exchange interactions are of opposite sign, and the competition produces pronounced spatial anisotropy in the predicted neutron cross section. We provide a comprehensive report of our findings to encourage future experimental studies of EuO and EuS, which are classic examples of insulating, ferromagnetic salts.

A second objective in our work is to explore the range of validity of the mode-coupling theory and various approximate analytic studies of the integro-differential equations. Our results are derived from a numerical solution of the mode-coupling equations, and we estimate that the results are accurate within $\simeq 2\%$. Availability of computer time and resources has placed limit on the range of wave vectors examined. To make significative

 

progress beyond the results presented here requires, we estimate, many hours of CRAY X-MP/12 CPU time.

## II. FORMULATION OF THEORY

Mode coupling integro-differential equations for the spin correlation functions of a Heisenberg ferromagnet including a small applied magnetic field can be derived by decoupling correlations involving more than two spin operators, which is a method that has been discussed in Refs. 23 and 24 for zero applied field. Similar results are obtained using the scheme employed by Refs. 25 and 26 and these topics are addressed in the Appendix.

The starting point is the Heisenberg ferromagnetic Hamiltonian in which $N$ spins on sites $i,j$ are coupled by exchange interactions $J_{ij}$,

$$\mathcal{H}=-\tfrac{1}{2}\sum_{i,j}J_{ij}\mathbf{s}_i\cdot\mathbf{s}_j-h\sum_i s_i^z \ , \tag{2.1}$$

and this is usefully rewritten in terms of Fourier components:

$$\mathcal{H}=-\frac{N}{2}\sum_{\mathbf{q}}J_{\mathbf{q}}\mathbf{s}_{\mathbf{q}}\cdot\mathbf{s}_{-\mathbf{q}}-Nhs_0^z \ , \tag{2.2}$$

where $h=Hg\mu_B$, for an applied magnetic field $H$ which define the $z$ axis, and the Fourier components

$$s_{\mathbf{q}}^{\alpha}=\frac{1}{N}\sum_l e^{i\mathbf{q}\cdot\mathbf{R}_l}s_l^{\alpha} \ , \tag{2.3a}$$

$$J_{\mathbf{q}}=\sum_l J_{lj}e^{i\mathbf{q}\cdot(\mathbf{R}_l-\mathbf{R}_j)} \ . \tag{2.3b}$$

(in the following, to simplify notation, we suppress the vector symbol for the wave vector index of Fourier transformed quantities). Finally, we define the spin fluctuation operators

$$S_k^{\alpha}=s_k^{\alpha}-\langle s_k^{\alpha}\rangle \ , \tag{2.4}$$

and the new constants $J_{qk}=J_k-J_{q-k}$; moreover, throughout the paper we set the Planck's and Boltzmann's constants $\hbar$ and $k_B$ equal to one.

Taking into account that

$$\langle s_k^{\alpha}\rangle=\langle s^z\rangle\delta_{\alpha z}\delta_{k0} \ ,$$

we obtain the following exact coupled equations of motion:

$$\frac{dS_q^+}{dt}=-\omega_q S_q^+ + i\sum_k J_{qk}S_k^+S_{q-k}^z \ , \tag{2.5a}$$

$$\frac{dS_q^z}{dt}=-\tfrac{1}{2}i\sum_k J_{qk}S_k^+S_{q-k}^- \ , \tag{2.5b}$$

$$\frac{dS_q^-}{dt}=\left[\frac{dS_{-q}^+}{dt}\right]^* \ , \tag{2.5c}$$

where the random-phase approximation (RPA) frequency

$$\omega_q=h+\langle s^z\rangle(J_0-J_q) \ .$$

In analyzing magnetic neutron-scattering experiments, we are interested in the fluctuation spectra $S(q,\omega)$, namely in the Fourier transform of the quantity, $\langle S_q^{\alpha}(t)[S_q^{\alpha}(0)]^{\dagger}\rangle$, which is related to the Fourier transform of the Kubo relaxation function (see for example, Ref. 24),

$$R_q(t)=(S_q^{\alpha}(t),[S_q^{\alpha}(0)]^{\dagger}) = \int_0^{\beta}d\lambda\{\langle S_q^{\alpha}[S_q^{\alpha}(t+i\lambda)]^{\dagger}\rangle\}-\beta\langle S_q^{\alpha}\rangle\langle S_q^{\alpha}\rangle \ , \tag{2.6}$$

by the fluctuation-dissipation theorem ($\beta=1/T$),

$$S(q,\omega)=\frac{\omega}{1-\exp(-\beta\omega)}R_q(\omega) \ . \tag{2.7}$$

We now consider an exact equation for $R_q(\omega)$ derived from a generalized Langevin equation. Let the column vector $\underline{S}$ have components $S_q^+,S_q^z,S_q^-$, with $q$ varying in all the Brillouin zone, and the susceptibility matrix $\chi=(\underline{S},\underline{S}^{\dagger})$. The exact equation of motion for the matrix of normalized relaxation functions,

$$F(t)=(\underline{S}(t),\underline{S}^{\dagger})\chi^{-1} \ , \tag{2.8}$$

can be cast in an appealing form by using the Mori-Zwanzig projection method and it reads

$$\dot{F}(t)=i\Omega F(t)-\int_0^t dt'M(t-t')F(t') \ , \tag{2.9}$$

where a dot is used to denote the differentiation with respect to time, and the frequency matrix is

$$i\Omega=(\dot{\underline{S}},\underline{S}^{\dagger})\chi^{-1} \ . \tag{2.10}$$

Here, the memory function

$$M(t)=(f(t),f^{\dagger}(0))\chi^{-1}$$

is defined in term of the fluctuating force

$$f(t)=e^{i(1-\underline{P})tL}(1-\underline{P})\dot{\underline{S}} \ , \tag{2.11}$$

whose evolution is not controlled by the simple Liouville operator $L$, but entails the projection operator $\underline{P}$ defined by the operation

$$\underline{P}A=(A,\underline{S}^{\dagger})(\underline{S},\underline{S}^{\dagger})^{-1}\underline{S} \ , \tag{2.12}$$

where $A$ is an arbitrary column matrix operator.

Owing to the cubic point group and translational symmetries of the system, the matrices so far defined are diagonal both in wave vectors and Cartesian indices, and we can limit our study to the $3\times3$ subspace with fixed $q$. In the subspace we have

$$\chi_q=\begin{bmatrix}2\chi_q^{\perp} & 0 & 0\\ 0 & \chi_q^{\parallel} & 0\\ 0 & 0 & 2\chi_q^{\perp}\end{bmatrix} \ , \tag{2.13}$$

where the transverse and longitudinal susceptibilities are

$$\chi_q^{\perp}=(S_q^x,S_{-q}^x)=(S_q^y,S_{-q}^y)$$

and $\chi_q^{\parallel}=(S_q^z,S_{-q}^z)$, and the frequency matrix

$$\Omega_q=\begin{bmatrix}-\Omega_q & 0 & 0\\ 0 & 0 & 0\\ 0 & 0 & \Omega_q\end{bmatrix}, \tag{2.14}$$

where

$$\Omega_q=\omega_q-(\tfrac{1}{2}\chi_q^{\perp})\sum_k J_{qk}(S_k^+S_{q-k}^z,S_{-q}^-)\ ,$$

so that

$$f_q(t=0)=\begin{bmatrix} i\sum_k J_{qk}\left[S_k^+S_{q-k}^z-\dfrac{(S_k^+S_{q-k}^z,S_{-q}^-)}{2\chi_q^{\perp}}S_q^+\right] \\ -\dfrac{i}{2}\sum_k J_{qk}S_k^+S_{q-k}^- \\ -i\sum_k J_{qk}\left[S_k^-S_{q-k}^z-\dfrac{(S_k^-S_{q-k}^z,S_{-q}^+)}{2\chi_q^{\perp}}S_q^-\right]\end{bmatrix}. \tag{2.15}$$

Equations (2.9)–(2.15) are exact. To proceed further we are forced to make approximations because we do not know the full effect of the projection operator $\underline{P}$ in the modified evolution operator encountered in (2.11) or how to evaluate the four operator correlation function appearing in the quantity $(f(t),f^{\dagger})$.

The simplest nontrivial solution consists in neglecting $\underline{P}$ in the evolution operator of the fluctuating force and at the same time decoupling scalar products where more than two spin operators appear.

It is worthwhile to notice that both the approximations are essential to give physical results. Although this approach, as shown in Refs. 11, 27, and 28, can be insufficient to reproduce some behavior of the elementary excitations, it is found to give good results in many magnetic problems and in critical dynamics.

After decoupling, all terms containing a scalar product involving three operator disappear, and considering all possible decoupling of the type

$$(S_k^+(t)S_{q-k}^z(t),S_{k'-q}^zS_{k'}^-)\simeq T(S_k^+(t),S_{-k}^-)(S_{q-k}^z(t),S_{k-q}^z)\delta_{k-k'}\ , \tag{2.16}$$

we obtain the following coupled integro-differential equations for the normalized relaxation functions:

$$\begin{aligned}\dot F_q^+(t)&=-i\Omega_qF_q^+(t)-T\sum_k(J_{qk})^2\frac{\chi_k^{\perp}\chi_{q-k}^{\parallel}}{\chi_q^{\perp}}\int_0^t dt'\,F_k^+(t-t')F_{q-k}^z(t-t')F_q^+(t')\ ,\\ \dot F_q^z(t)&=-T\sum_k(J_{qk})^2\frac{\chi_k^{\perp}\chi_{q-k}^{\perp}}{\chi_q^{\parallel}}\int_0^t dt'\,F_k^+(t-t')F_{q-k}^-(t-t')F_q^z(t')\ ,\\ \dot F_q^-(t)&=[\dot F_q^+(t)]^*\ .\end{aligned} \tag{2.17}$$

In the Appendix we show that our result is the same up to term of the first order in three operators scalar products as the one obtained by Kawasaki[26] in the critical region using a quite different approach. We also provide a comparison with the equations obtained by McLean and Blume.[25]

To solve Eq. (2.17), we have to specify the static quantities $\chi_q^{\perp}$, $\chi_q^{\parallel}$, and $\langle s^z\rangle$. In the isotropic case the functional form of $\chi_q$ can be obtained by requiring that the second derivative of $F_q(t)$ has the correct value when $t=0$ and this prescription leads to the susceptibility of the spherical model.[29,30] Applied to our case, the same prescription gives the following equations:

$$\sum_k(J_{qk})^2\chi_k^{\perp}\chi_{q-k}^{\perp}=\frac{2}{N}\sum_k J_{qk}\chi_k^{\perp}\ , \tag{2.18a}$$

$$\sum_k(J_{qk})^2\chi_k^{\perp}\chi_{q-k}^{\parallel}=\frac{1}{N}\sum_k J_{qk}(\chi_k^{\perp}+\chi_k^{\parallel})\ . \tag{2.18b}$$

Equation (2.18a) contains only the perpendicular susceptibility $\chi^{\perp}$. Since it has exactly the same form as the corresponding equation obtained in zero field, we obtain a spherical model solution

$$\chi_q^{\perp}=\frac{1}{N}\frac{1}{\lambda-J_k}\ , \tag{2.19}$$

where $\lambda$ is a temperature-dependent parameter which has to be determined. Subtracting Eq. (2.18a) from (2.18b) we obtain

$$\sum_k J_{qk}(\chi^{\parallel}_{q-k}-\chi^{\perp}_{q-k})\left[J_{qk}\chi^{\perp}_k+\frac{1}{N}\right]=0\ , \tag{2.20}$$

which admits the solution $\chi^{\parallel}_k=\chi^{\perp}_k$. This result, which can appear surprising and inconsistent with the field induced anisotropy of the system, is simply a consequence of the decoupling. To better understand this point, we refer to the Appendix where we show that

$$\chi^{\parallel}_k-\chi^{\perp}_k=\langle s^z\rangle(2\chi^{\perp}_0 T)^{-1}(S^+_0,S^-_{-k},S^z_k),$$

and thereby conclude that the perpendicular and longitudinal susceptibilities are the same in an approximation scheme which incorporates only pair correlations.

## III. NUMERICAL METHOD

In this section we give a brief description of the method followed to obtain a numerical solution of Eq. (2.17) in the absence of an applied magnetic field. In this case, the system of coupled equations (2.17) reduces to only one equation, which, if the static susceptibility is that one of the spherical model, Eq. (2.19), can be rewritten as

$$\dot{F}_q(\tau)=-\frac{2}{N}\sum_k(\gamma_k-\gamma_q)(\gamma_k-\gamma_{q-k})G_k\int_0^\tau d\tau'\,F_k(\tau-\tau')F_{q-k}(\tau-\tau')F_q(\tau')\ , \tag{3.1}$$

where $\tau=J_0SW^{-1/2}t$ is a dimensionless reduced time, the factor $\gamma_q=J_q/J_0$ and

$$G_q=\frac{NTW}{S^2}\chi_q=\frac{(S+1)}{3S}\frac{\Theta}{\mu+1-\gamma_q}\ , \tag{3.2}$$

in which the reduced temperature $\Theta=T/T_c$. The temperature-dependent parameter $\mu$ appearing in (3.2) is obtained by using the sum rule

$$\frac{1}{N}\sum_k\frac{1}{\mu+1-\gamma_k}=\frac{W}{\Theta}\ , \tag{3.3}$$

and the Watson integral $W$ is defined by the equality (3.3) when $\Theta=1$ and $\mu=0$. Previous solutions of Eq. (3.1) were obtained either in the critical region, performing an approximation valid for long time and large wave vectors,[10] or for a simple cubic paramagnet with only nearest neighbors interactions.[31,32] The numerical solution which we present here is valid for a face-centered-cubic lattice with both nearest neighbors and next-nearest-neighbors interactions, which is a realistic model of europium salts.

First of all we must consider the problem of discretization in variables space, which is intrinsic to every numerical method. Wave vectors are defined on a cubic mesh with $2n$ parts on each edge, of length $4\pi/a$, of the cube which represents a double Brillouin zone in the reciprocal lattice. Owing to the symmetry properties of a bcc lattice, many of the $8n^3$ points are equivalent, and therefore a solution of equation (2.17) has to be obtained only for a limited number of them. If we use for the wave vectors the unit $2\pi/na$, a point in our mesh is described by $(k_1,k_2,k_3)$, where $k_1,k_2,k_3$ are integer numbers in the range $-n+1\le k_i\le n$, and the unequivalent points are defined by the relations

$$0\le k_1\le k_2\le k_3\le n\ , \tag{3.4a}$$

$$k_1+k_2+k_3\le\tfrac{3}{2}n\ . \tag{3.4b}$$

The integration of the equations in the time domain has been performed using the simplest method, which is the step-by-step integration retaining only the lowest-order terms. We have exploited the relatively fast decay of $F_q(t)$ at large wave vectors to cut down the running time of the computer program. This is accomplished by using an expanded time increment at large times when $F_q(t)$ has most decayed to zero for high $q$, and it is slowly varying for small $q$.

Another problem which derives from the application of discretization is the fact that the contribution to the sum over the wave vectors of the term with $k=0$ diverges at $T=T_c$. To cope with this singular behavior we have replaced the term with $k=0$ by an integral over a sphere of volume $1/8n^3$ centered in $k=0$. The integral is obtained replacing all elements with their value for $k=0$, except $G_k$ which is replaced by an Ornstein-Zernicke form. Such a procedure is equivalent to the substitution

$$G_0=\frac{(S+1)}{3S}\frac{\Theta}{\mu}\to\bar{G}_0=\frac{(S+1)}{3S}\Theta\int\frac{d^3k}{\mu+s^2k^2}\ , \tag{3.5}$$

in which

$$\bar{G}_0=\frac{(S+1)}{3S}\Theta\frac{4\pi}{s^2}\times\left[\sqrt[3]{3/4\pi}-\frac{\sqrt{\mu}}{s}\arctan\left[\frac{s}{\sqrt{\mu}}\sqrt[3]{3/4\pi}\right]\right]\ , \tag{3.6}$$

where

$$s^2=[(J_1+J_2)/6(2J_1+J_2)]4\pi^2/n^2\ .$$

Obviously such a substitution is strictly necessary only for $T=T_c$, i.e., $\mu=0$; however, we can note that the difference between $\bar{G}_0$ and $G_0$ becomes smaller and smaller in the high-temperature limit.

In the present calculation we have used $n=8$; such a value of $n$ gives us adequate accuracy in the results, and the improvement we can achieve taking, for example, $n=16$ is negligible compared to the large computer time

demanded (about 6 h on a CRAY X-MP/12).

The choice of the time step has been made empirically and it is based on the stability of the solution in respect of its variation. The numerical results we present have been obtained with $\Delta\tau=0.08$ for $0\leq\tau\leq 40$, $\Delta\tau=0.4$ for $40\leq\tau\leq 200$, and so on, multiplying $\Delta\tau$ and $\tau$ using a factor of 5 at each new iteration. We have found that decreasing to an half $\Delta\tau$ the variation of $F_q(\tau)$ for $\tau=2$ is less than 1%.

Once we obtained the relaxation function $F_q(t)$ in the time domain, the respective spectrum shape $F_q(\omega)$ has been computed by using a standard fast Fourier transform routine.

We have verified that our program applied to the simple cubic (sc) and nearest-neighbor Heisenberg model reproduces the results obtained in Refs. 31 and 32. Furthermore the long time behavior is found to be in agreement with low-$q$ expansion used in Ref. 10.

## IV. NUMERICAL RESULTS

The results obtained by using the numerical method outlined in the previous paragraph are here described and compared with experimental data and computer simulation where available. The numerical values of the exchange constants reported in the literature by fitting to the low-temperature spin-wave spectra are in good agreement with each other,[3,22] and the following values are used in the present calculation: $J_1=1.22$ K, $J_2=0.25$ K, and $a=5.12$ Å for EuO; $J_1=0.48$ K, $J_2=-0.24$ K and $a=5.95$ Å for EuS.

### A. Critical behavior

The first topic we consider here is the behavior of the system in the critical region, i.e, at $T=T_c$ and small wave vectors. In discussing the critical dynamics, we refer almost entirely to EuO, because more complete and detailed sets of experimental data are available for this compound; however, we can observe that, as far as the dipolar interaction is neglected, the critical properties of EuS are the same of EuO, apart from a different time scale of dynamical phenomena. This has been verified by our numerical results (see Fig. 6) and it is a consequence of the fact that, as it is well known, the microscopic details of the interactions give only a negligible contribution to large scale properties of the system, which dominate at the phase transition.

The most important result of the large amount of experimental work devoted to explore the critical dynamics of EuO is the confirmation of the dynamical scaling law with a dynamical critical exponent $z=2.5$, which can be derived theoretically using many different approaches, included the mode-coupling theory. The scaling law is confirmed both by conventional neutron scattering and neutron spin-echo experiments, but these same results, which corroborates the scaling, are in some sense surprising. In fact the scaling law is verified for a very large range of wave vectors: up to $q\simeq 0.4$ Å$^{-1}$ (Ref. 5), which corresponds to about $\frac{2}{5}$ of the Brillouin zone, and it is therefore not a very small wave vector. Moreover, the time domain explored in spin-echo experiments by Mezei[2] contains relatively short times in which the universal behavior should not yet be accessed. Finally, and perhaps the more striking fact, Mezei obtained, after a very short time, a simple exponential decay for the relaxation function $F_q(t)$, which is not consistent with the various theories applied to this problem and it is not confirmed by the most recent conventional neutron-scattering experiments,[5] which give a non-Lorentzian shape for the spectrum.

Although the smallest value of $q$ for which we have obtained the numerical solution is

$$q_0=\pi/4a\simeq 0.1534\text{Å}^{-1}\ ,$$

which is 6.4 times larger than the value $q=0.024$ Å$^{-1}$ for which detailed experimental data for $F_q(t)$ are reported by Mezei,[2] we generate the appropriate results for such wave vector from our detailed knowledge for the correlation function at the larger wave vectors.

In fact, the comparison of the shape of $F_q(\tau)$ for $q=q_0$, $\sqrt{2}q_0$, $\sqrt{3}q_0$, and $2q_0$, which lie all in the range of wave vectors for which the scaling has been experimentally verified, shows that they are very similar in their functional forms. Values do not scale with $q^{5/2}$, since, for very short times,

$$F_q(\tau)\simeq 1-\tfrac{1}{2}A_q\tau^2\ ,$$

where $A_q\propto q^4$ in a system where the total spin is conserved. This regime is consistent with an effective scaling exponent $z=2$, and it is valid up to $\tau\simeq 8$ for $q=q_0$, which should correspond to a value $t\simeq 100$ ps for the wave vector of Mezei's experiment; the order of magnitude of this microscope regime time is the same as obtained in Ref. 12. As the time increases, the effective exponent $z$ increases too, and attains a value of about 2.4 only for $\tau>150$ for $q=q_0$ (i.e., $t>2$ns for $q=0.024$ Å$^{-1}$); the values of $z$ obtained are essentially independent of $q$ in the previously mentioned range of wave vectors.

Correspondingly, the Fourier transform of the relaxation function $F_q(\omega)$ shows a scaling with $q^{5/2}$ only for small $\omega(\omega<0.2$ meV for $q=q_0)$ and in this range of $\omega$ it reproduces the universal function calculated in Ref. 31.

In view of these observations we can confidently transfer the results obtained for $Fq_0(\tau)$ to the time scale of $q=0.024$ Å$^{-1}$ using for the scaling the effective exponent $z(\tau)$ derived from our numerical calculations. We emphasize that the procedure adopted does not rely on theoretical speculation but is based firmly on our findings from the detailed numerical solution of the mode-coupling equations. The values displayed in Fig. 1 are obtained from our derived $Fq_0(\tau)$ by evaluating $\tau_{q=0.024}=\tau(q_0/0.024)^{z(\tau)}$. As we can see our results are similar to those obtained by Folk and Iro in the framework of renormalization group (RG) theory, and are thus in contrast with the findings by Mezei. As guessed recently,[33] and confirmed by our numerical calculation, an exponential decay would be explicable if the experiment should not be performed exactly at $T=T_c$, so that a crossover to hydrodynamic regime could occur. However, in this case a different scaling would be present; more-

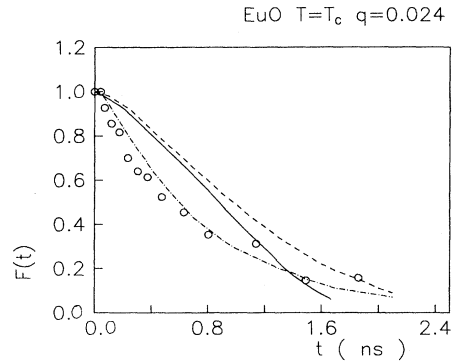


FIG. 1. Comparison of various theoretical results and spin-echo data by Mezei: circle, Mezei data (Ref. 2); dot-dashed line, hybrid theory by Balucani *et al.* (Ref. 12); dashed line, renormalization group calculation by Folk and Iro (Ref. 15); solid line, our numerical results.

over, the onset of this behavior, according to calculation in the framework of the spherical model, could be seen starting from $T \simeq 1.01 T_c$ for $q=0.024$ Å$^{-1}$, which is far from the critical temperature.

As we said the solution of the equation of motion in the critical region for EuS does not give any new features. However new experimental data at $T=T_c$ for small $q$ would be interesting to see how important is the dipolar interaction, so far neglected in the theory, which in this compound is stronger relative to the exchange interaction, than in EuO, and should therefore modify the critical behavior. The dynamical scaling functions for Heisenberg ferromagnets have been calculated recently in the framework of mode-coupling theory, taking into account also the dipolar interaction,[13,14] and the effect of the latter seems to be revealed by very recent experimental studies performed on EuS (Ref. 34).

### B. EuO

The numerical calculation for EuO has been carried out for $T=T_c$, $T=1.68T_c$, and $T=2T_c$. Experimental results are available at these temperatures; moreover computer simulations have been performed at $T=1.68T_c$ and $T=2T_c$.

A large amount of experimental data for EuO reported in the literature are obtained with a powder sample, but the effects of average among the various direction are negligible, because our calculations show that the line shape is almost independent on the direction of wave vectors even at the zone boundary, where the difference is less than 10%.

Finally, in order to normalize the experimental results at $T=T_c$ and $T=1.68T_c$ we have drawn the line shapes derived from the three poles approximation,[35] using for the parameter, $\delta_1$ and $\delta_2$, the values obtained by the fitting to the experimental data and reported by the authors in Ref. 4. Such a procedure also permits some account of the effect of the spectrometer resolution which we do not know in detail, but that turns out to be important for small wave vectors.

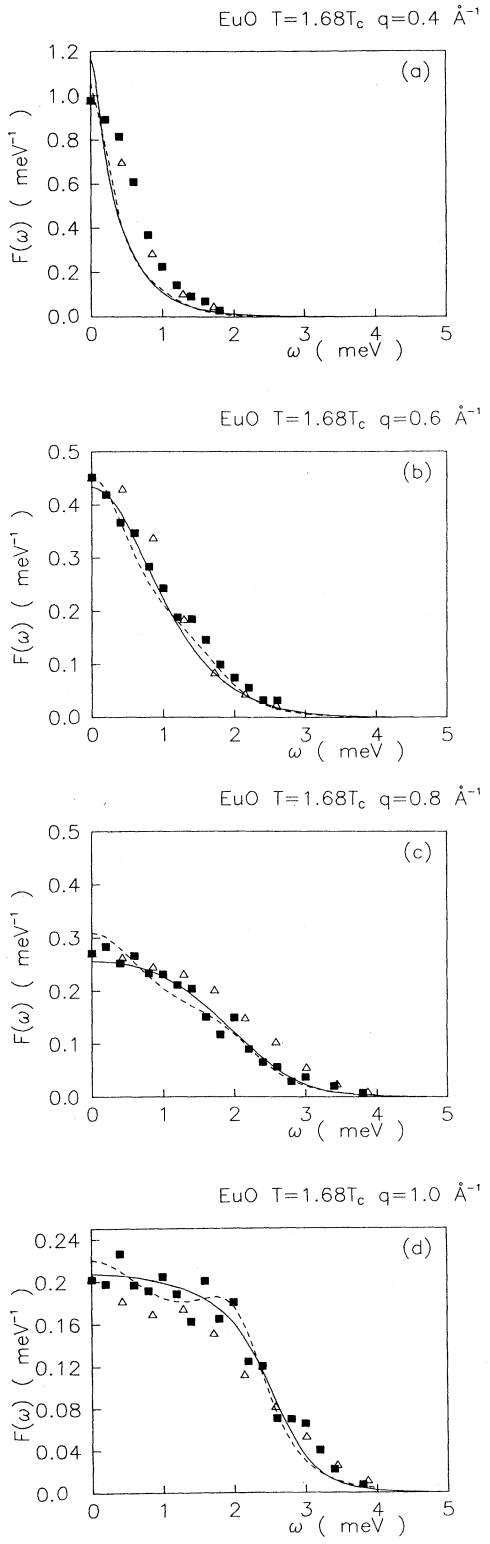


FIG. 2. Comparison of our numerical results with spin dynamics calculation (Ref. 7) and neutron scattering data (Ref. 4) at $T=1.68T_c$ for EuO. Solid line, our numerical results; dashed line, three pole approximation as explained in the text; solid quad, neutron data; open triangle, spin dynamics simulation.

The comparison with spin dynamics results[7] for the spectrum shape $S(q,\omega)$ has been made reporting in our plots the data normalized to the experimental ones as done in Ref. 7. As shown in Fig. 2, our numerical results are in excellent agreement with experimental data for all wave vectors at $T=1.68T_c$. The relevance of resolution corrections is apparent in Fig. 2(a); as a matter of fact, the three poles curve, which practically coincides with our results, also gives very good fit of experimental data.[4] The comparison with spin dynamics calculation is also favorable, but care is required with the normalization because if we try to evaluate

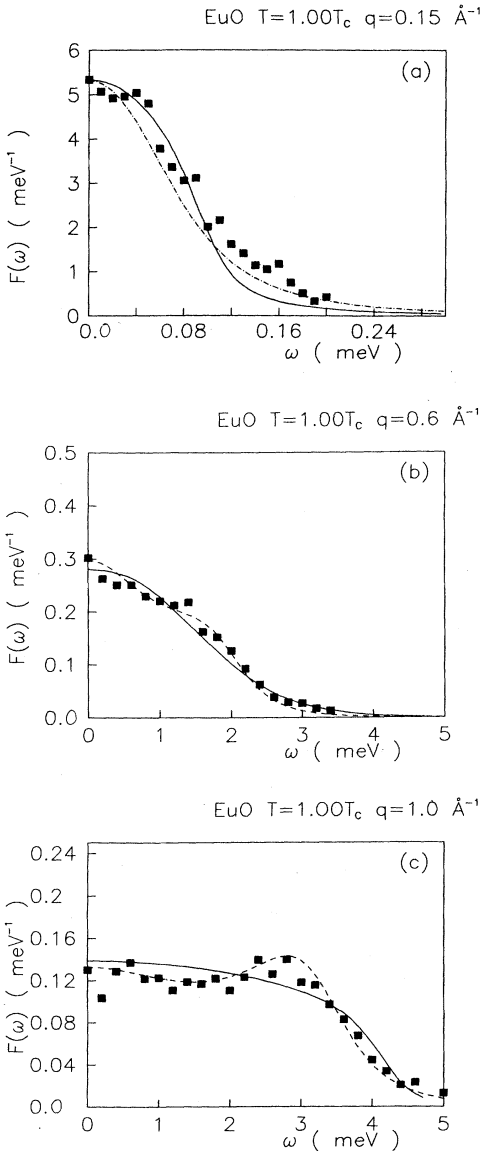


FIG. 3. Comparison of our numerical results with neutron data (Refs. 4 and 5) at $T=T_c$ for EuO. The meaning of various symbols is the same as in Fig. 1, but the dot-dashed line in Fig. 2(a) represents the function proposed by Folk and Iro (Ref. 15) (see the text).

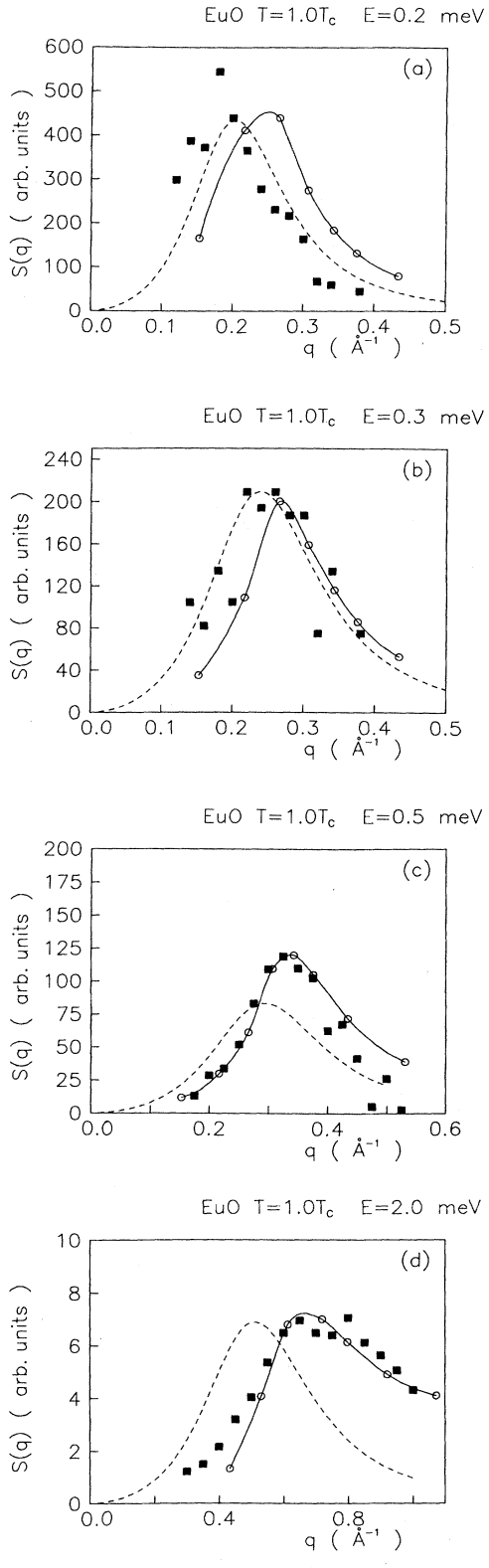


FIG. 4. Experimental constant energy spectra compared with renormalization group theory and our mode-coupling results. Solid quad, experimental data (Ref. 5); dashed line, renormalization group (Ref. 15); open circle and solid line through them, our numerical results.

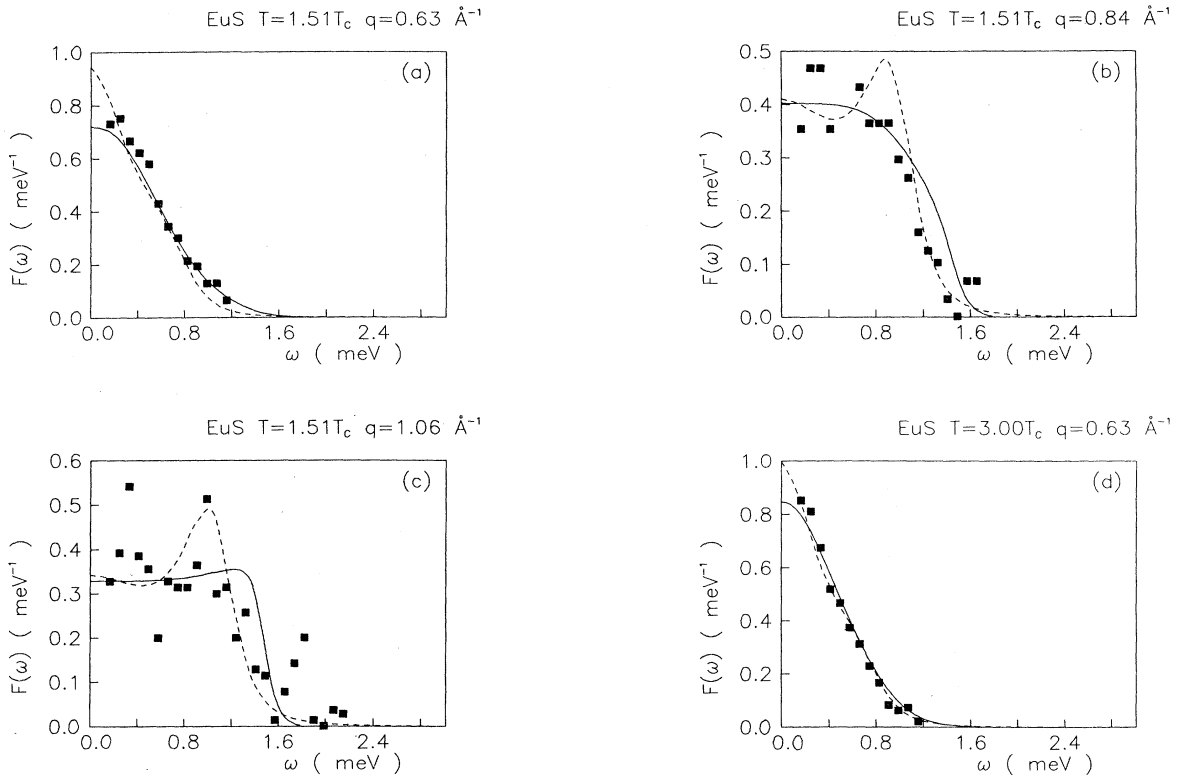


FIG. 5. Comparison of our numerical results and neutron scattering data (Ref. 3) along (1,0,0) direction at $T=1.51T_c$ and $T=3.0T_c$ for EuS. Solid line, our numerical results; dashed line, three pole approximation as explained in the text; solid quad, neutron data.

$$F_q(\omega)=(1/3T\chi_q)S(q,\omega)\ ,$$

using for $\chi_q$ the spherical model susceptibility and for $S(q,\omega)$ the spin dynamics results, we obtain values smaller than ours or those given by the three poles approximation.

In Fig. 3, the comparison with experimental data is given at $T=T_c$ for three wave vectors, showing all the significative information. The dot-dashed line in Fig. 3(a) represents a different analytic form for the relaxation function derived by Folk and Iro[15,16] by means of the renormalization-group approach and used by Böni *et al.* to fit their experimental data[5]; it has been plotted for the same purpose of the three poles in the other figures. Also, for $T=T_c$ the agreement between our theory and experimental results is good for intermediate and small wave vectors, but our numerical calculations fail to reproduce the inelastic shoulder in the line shape at the zone boundary [Fig. 3(c)].

For $T=2T_c$, the comparison with spin dynamics calculation gives almost the same results than that for $T=1.68T_c$. The existing experimental data for this temperature are obtained from a single crystal, but are given only for the two wave vectors $q=0.5q_{ZB}$ and $q=q_{ZB}$ along the [111] direction.[1] In this case we do not have any good criterion to normalize the measured intensity; the agreement is not very good anyway because the experimental data show a well-defined two peaked structure which is not present in our calculation. However, we can observed that these results of Ref. 1 are partially contradicted by those by Böni and Shirane,[4] because the latter show no well-defined inelastic peak at $T=1.68T_c$.

Finally, in Fig. 4, we compare our results with experimental data obtained in constant energy scans[5] and renormalization group predictions. The normalization constant has been fixed in each plot to fit the intensity of the maximum. The experimental data are very well reproduced by the renormalization-group calculation at low energies, but this approach fails at higher energies, where our results become very good. This can be explained taking into account that at higher energies the large wave-vector contributions become more important. These microscopic characteristics of the system cannot be taken into account by a renormalization-group approach, based on the continuum limit of the Heisenberg Hamiltonian.

### C. EuS

For europium sulfide the solution of Eq. (3.1) has been obtained for $T=T_c$, $T=1.08T_c$, $T=1.51T_c$, and $T=3T_c$. Detailed experimental data are available for $T=1.08T_c$, $T=1.51T_c$, and $T=3T_c$, and the solution for $T=T_c$ has been produced mainly for the purpose of comparison with EuO.

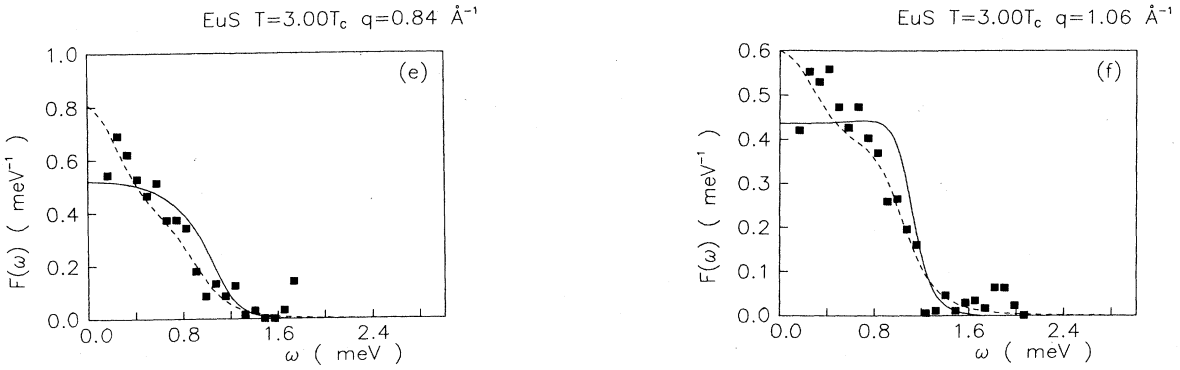


FIG. 5. (*Continued*).

The available experiments were performed on a single crystal of EuS along the [100] direction,[3] and the comparison with our results is shown in Fig. 5; the normalization of the intensity having been made using the same procedure as for EuO. It is apparent from the figures that the agreement is good for $q=0.63$ Å$^{-1}$ at both temperatures. For higher wave vectors the agreement is also fairly good, except for some point around $E \simeq 0.3$ meV. However, we have to observe that the experimental data for EuS do not appear to be as good as those for EuO, not only because of the relatively large error bars,[3] but also because they show an enhanced intensity around $E \simeq 2$

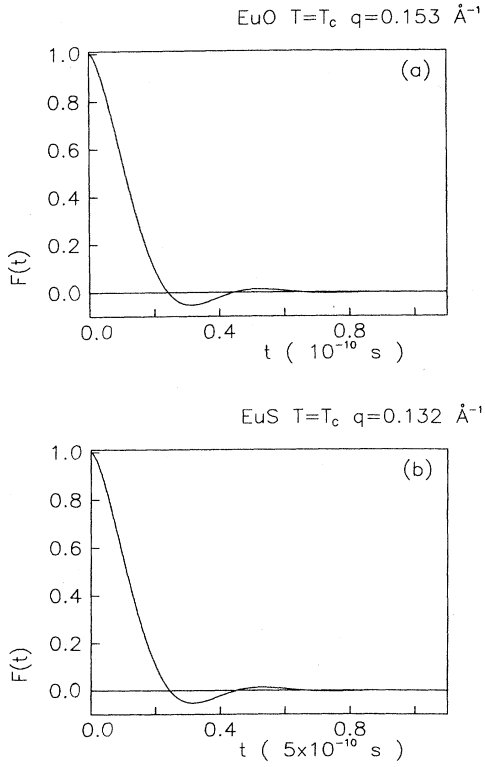


FIG. 6. Comparison of the relaxation functions in EuO and EuS for $q=(2\pi/8a)$, the smallest wave vector given by our numerical solution; as expected, they are equal, except for the different time scale.

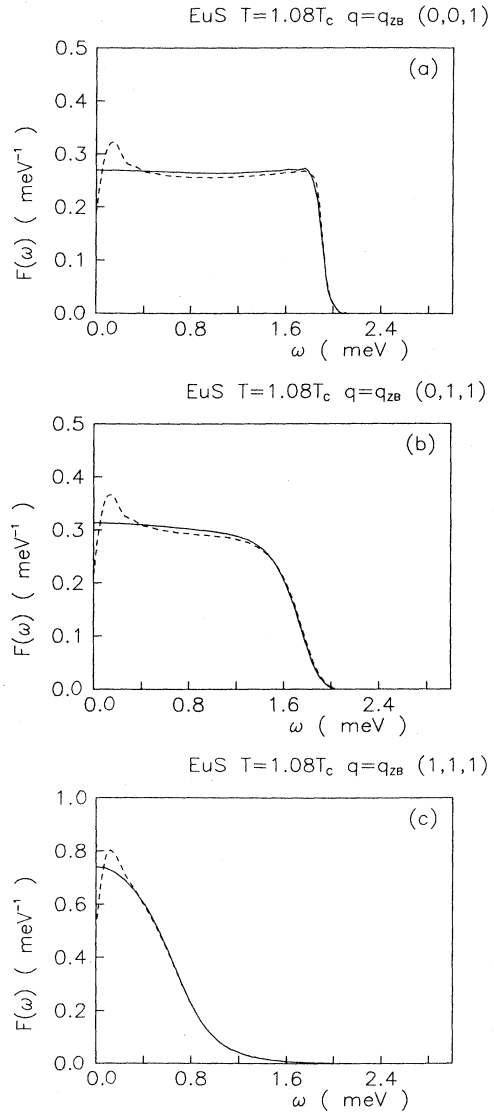


FIG. 7. Mode-coupling theory results for the longitudinal line shape at $T=1.08T_c$ for EuS at the zone boundary along (0,0,1) (0,1,1), and (1,1,1). Solid line, isotropic case; dashed line, with an external field $H=100$ G. They show a marked spatial anisotropy and the very small effects of the applied field.

meV which can be an index of an incomplete subtraction of nonmagnetic effects. As it is clearly shown in Figs. 5(c) and 5(f), our numerical solution of the mode-coupling equations is able to give an inelastic shoulder at the zone boundary, but much less intense than that one produced by three pole approximation. However the question as to whether this peak is really present in the experimental data has, in our opinion, no unambiguous answer because there is no reason to accept the peak in the experimental data at $E \simeq 1$ meV as a true peak and reject a similar peak at $E \simeq 0.3$ meV.

As we already noted in Sec. IV A, the behavior of relaxation function for EuS at $T = T_c$ and small $q$ is the same as obtained for EuO, and this is shown in Fig. 6. For larger wave vectors new important features appear, because the relaxation functions show a strong dependence on the direction of $q$. This effect is a direct consequence of the competing exchange interactions of EuS and can be observed in Fig. 7 for wave vectors at the zone boundary along the three principal directions for $T = 1.08 T_c$. New experiments are required to verify our finding at to test the quantitative reliability of our solution, but some preliminary results[3,6,36] seem to be in agreement with our prediction.

Finally, in Fig. 7 we present some results for the spectrum of longitudinal fluctuations in the presence of an applied magnetic field $H = 100$ G. The magnetization induced by such field at $T = 1.08 T_c$ is sufficiently small that the effect of the applied field can be treated perturbatively; in this framework, retaining only first order terms in $(\langle S^z \rangle / S)$, the static susceptibilities are assumed to be unchanged in respect to the isotropic case. As it is apparent from the figure, the effect of such magnetic field on the spectrum shape in the paramagnetic region is negligible, the only contribution being the appearance of a small inelastic peak at a frequency which practically coincides with the RPA frequency of spin wave $\omega_q$.

## V. CONCLUSION

We have used the mode-coupling theory to study the spin correlations of real Heisenberg ferromagnets EuO and EuS in all paramagnetic phase and across the entire Brillouin zone. For the critical temperatures and wave vectors, the spectral shapes are not, of course, dependent on the details of the exchange interaction, so that the same results are found for EuO and EuS. While they reproduce the results of the renormalization group approach and the neutron-scattering data available for EuO, the relaxation function measured by neutron spin echo[2] is not recovered; an exponential decay is obtained in the hydrodynamic regime, but the latter is not in agreement with a dynamic scaling with $z = 2.5$, and it would imply a mismatch of about 1% between the critical temperature and the experimental temperature for the wave vector considered in Ref. 2. However, very recent calculations[37] have pointed out that the inclusion of the dipolar interaction in the mode-coupling equations is able to reproduce the exponential decay observed in spin-echo experiment with the correct scaling. These results have been confirmed in Ref. 38.

The theory seems to work rather well to explain a lot of different features between EuO and EuS for intermediate and zone boundary wave vectors in the paramagnetic region. The comparison with spin dynamics simulation appears satisfactory, taking into account the accuracy of the latter. Finally, the effects of a small applied magnetic field are shown to be negligible, considering also that they are smaller in EuO than in EuS and that they decrease as the temperature increases.

## ACKNOWLEDGMENTS

We are grateful to North Atlantic Treaty Organization (NATO) Scientific Affairs Division for the Research Grant No. 609/84. Furthermore, one of us (A.C.) thanks the Neutron Division of the Rutherford Appleton Laboratory (RAL) for hospitality and the Neutron Commission of the Italian Research National Council (CNR) for financial support during his stay at RAL, where a major part of this work was performed.

## APPENDIX

This Appendix is devoted to a comparison of the integro-differential equations obtained applying the method of decoupling, with those derived by Kawasaki using a conventional mode-coupling formalism[26] and by McLean and Blume.[25] Moreover, we show the essential role played by static correlations among three operators to take into account the anisotropy between the parallel and perpendicular static susceptibilities induced by the applied field.

From the definition of the Kubo scalar product given in (2.6), it follows that, for any two operators $A$ and $B$,

$$(\dot{A},B) = -i\langle [A,B] \rangle \ .$$

Therefore, using the classical (or high-temperature) limit of fluctuation dissipation theorem, which is consistent with decoupling and used throughout the Kawasaki derivation, we have

$$(\dot{S}_q^+, S_{-k}^- S_{k-q}^z) = -i\langle [S_q^+, S_{-k}^- S_{k-q}^z] \rangle = \frac{-2iT}{N}(\chi_{q-k}^{\parallel} - \chi_k^{\perp}) \ . \tag{A1}$$

The quantity on the left-hand side can also be computed by using the equation of motion for spin operators (2.5) and decoupling four operator scalar products in the same way that we proceed in deriving the equation of motion (2.17); this gives

$$(\dot{S}_q^+, S_{-k}^- S_{k-q}^z) = -i\omega_q (S_q^+, S_{-k}^- S_{q-k}^z) + 2iJ_{qk} T \chi_k^{\perp} \chi_{q-k}^{\parallel} \ . \tag{A2}$$

Comparing (A1) with (A2) for $q = 0$ we have ($J_{0k} = 0$):

$$\chi_k^{\parallel} - \chi_k^{\perp} = \frac{\langle S^z \rangle}{2T\chi_0^{\perp}} (S_0^+, S_{-k}^- S_k^z) \ , \tag{A3}$$

where use has been made of the equality

$$\Omega_q = \frac{\langle S^z \rangle}{N\chi_q^{\perp}} = \omega_q - \frac{1}{2\chi_q^{\perp}} \sum_k J_{qk} (S_k^+ S_{q-k}^z, S_{-q}^-) \ , \tag{A4}$$

which is obtained in a similar way to (A3), evaluating $(\dot{S}_q^+, S_{-q}^-)$ in the two different ways and comparing the results. Equation (A3) shows clearly that correlations among three operators are essential to consider the anisotropy of the system.

Furthermore from (A1), (A2), (A4), and (2.14b) we obtain

$$J_{qk}=\frac{1}{N\chi^{\parallel}_{q-k}}-\frac{1}{N\chi^{\perp}_{k}}+\frac{\langle S^z\rangle}{2NT\chi^{\perp}_{q}\chi^{\perp}_{k}\chi^{\parallel}_{q-k}}(S_q^+,S_{-k}^-S_{k-q}^z)$$
$$-\frac{1}{4T\chi^{\perp}_{q}\chi^{\perp}_{k}\chi^{\parallel}_{q-k}}(S_q^+,S_{-k}^-S_{k-q}^z)\sum_{k'}J_{qk}'(S_{k'}^+S_{q-k'}^z,S_{-q}^-) \tag{A5}$$

and likewise, starting from $(\dot{S}_q^z, S_{-k}^-S_{k-q}^+)$,

$$\frac{1}{N\chi^{\perp}_{q-k}}-\frac{1}{N\chi^{\perp}_{k}}=J_{qk}\ . \tag{A6}$$

To compare out integro-differential equations (2.17) with Eqs. (8.12) and (8.13) given by Kawasaki in Ref. 26, we must consider that Kawasaki retains scalar products containing three operators, but not products among them. Therefore, the approximation which we made, neglecting the term containing $(S_k^+S_{q-k}^z, S_{-q}^-)$ in the definition of the fluctuating force (2.15), to obtain (2.17), is consistent; moreover, the last term in (A5) has to be neglected. Taking this into account, from (A4), (A5), and (A6), it follows that the method of decoupling gives the result derived by Kawasaki, the various different factors $N$ and $T$ in some terms deriving from the different normalization constants in the definition of $S_q$ and $\chi_q$.

Retaining three operators scalar products, Eq. (2.18a) is unchanged, but Eq. (2.18b) becomes

$$\sum_k (J_{qk})^2\chi^{\perp}_{k}\chi^{\parallel}_{q-k}=\frac{1}{N}\sum_k J_{qk}(\chi^{\perp}_{k}+\chi^{\parallel}_{k})+\frac{\langle S^z\rangle}{2NT\chi^{\perp}_{q}}\sum_k J_{qk}(S_k^+S_{q-k}^z,S_{-q}^-)\ , \tag{A7}$$

and so Eq. (2.20) acquires at its right-hand side a new term, and reads

$$\sum_k J_{qk}(\chi^{\parallel}_{q-k}-\chi^{\perp}_{q-k})(J_{qk}\chi^{\perp}_{k}+\frac{1}{N})=\frac{\langle S^z\rangle}{2NT\chi^{\perp}_{q}}\sum_k J_{qk}(S_k^+S_{q-k}^z,S_{-q}^-)\ . \tag{A8}$$

We can observe that Eqs. (A3) and (A4) follow from constraints imposed on the first time derivative of dynamical quantities for $t=0$, and they are able to give the static susceptibilities as function of $\langle S^z\rangle$ and three operators scalar products

$$\chi^{\perp}_{q}=\left[\frac{1}{N}+\tfrac{1}{2}\sum_k J_{qk}\frac{(S_k^+S_{q-k}^z,S_{-q}^-)}{\langle s^z\rangle}\right]\frac{1}{\dfrac{h}{\langle S^z\rangle}+J_0-J_q}\ , \tag{A9}$$

$$\chi^{\parallel}_{q}=\chi^{\perp}_{q}+\frac{\langle S^z\rangle}{2T\chi^{\perp}_{0}}(S_0^+,S_{-q}^-S_q^z)\ . \tag{A10}$$

Since,

$$\lim_{h\to 0}(h/\langle S^z\rangle)=N\chi_0^{-1}$$

and

$$(S_k^+S_{q-k}^z,S_{-q}^-)=0$$

for $h=0$, in the isotropic case, Eqs. (A9) and (A10) become the spherical model susceptibility, which also gives a correct value for the second time derivative for $t=0$ [see (2.18)]. When $h\neq 0$, Eq. (A8) continues to be automatically satisfied, since it is easy to show that it is equivalent to (A5), but inserting (A9) in (2.18a), and retaining only terms of the first order in three operators scalar products, we have

$$\frac{1}{N}\sum_k\frac{J_{qk}}{\dfrac{h}{\langle S^z\rangle}+J_0-J_q}\sum_{k'}J_{kk}'(S_{k'}^+S_{k-k'}^z,S_{-k}^-)=0\ , \tag{A11}$$

which can be seen as a condition on three operators scalar products to obtain the correct value of the second moment.

Regarding the equations for the relaxation functions obtained by McLean and Blume,[25] we must first of all observe that they are derived applying systematically a decoupling procedure which replaces higher-order correlations with products of all possible pairing of operators. This approximation is more natural than the decoupling (2.16), but it forces the use explicitly of the classical fluctuation-dissipation theorem in order to close the system of equations, which otherwise contain not only relaxation functions, but also time-dependent correlation functions. However, one can notice that the classical limit of the fluctuation-dissipation theorem is in some sense implicitly contained in (2.16) (Ref. 23), and so the two treatments are more similar than it might seem at a first sight, and indeed they give exactly the same results in the isotropic case.

On the contrary, in presence of an applied field $h \neq 0$, some differences appear between the two systems of integro-differential equations. In the equation by McLean and Blume for $F_q^+(t)$, the coefficient of the linear term is simply the RPA frequency $\omega_q$, and both the longitudinal and transverse memory functions are different from those appearing in (2.17). By requiring that the second time derivative in $t=0$, given by the equations by McLean and Blume, has the correct value, the following equations can be obtained:

$$\chi_q^{\parallel} \sum_k J_{qk} J_{q-kk} \chi_k^{\perp} = \frac{1}{N} \sum_k J_{qk} \chi_k^{\perp} , \tag{A12}$$

$$\chi_q^{\perp} \sum_k J_{qk} J_{q-kk} (\chi_k^{\perp} + \chi_k^{\parallel}) = \frac{1}{N} \sum_k J_{qk} (\chi_k^{\perp} + \chi_k^{\parallel}) + \frac{\omega_q}{T} \left[ \frac{\langle S^z \rangle}{N} - \omega_q \chi_q^{\perp} \right] . \tag{A13}$$

A possible solution of these equations is

$$\chi_q^{\parallel} = \chi_q^{\perp} = \frac{1}{N} \frac{1}{\dfrac{h}{\langle S^z \rangle} + J_0 - J_q} , \tag{A14}$$

which coincides with that of (A9) and (A10) when scalar products involving three spin operators are neglected; in this approximation also, the memory functions given by the two approaches become equivalent.

Therefore, we conclude that all treatments discussed here give the same results if we neglect the anisotropy between the static susceptibilities; this seems to be a good approximation when the applied field $h$ is small, owing to the fact that, since $(S_0^+, S_{-k}^- S_k^z) \to 0$, when $h \to 0$, Eq. (A3) shows that the difference between longitudinal and transverse static susceptibilities in the paramagnetic phase is of higher order in the applied field than the term already contained in the RPA frequency $\omega_q$.


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