

## Spin dynamics of EuS in the paramagnetic phase

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The spin dynamics of the semiclassical Heisenberg model on the fcc lattice, with ferromagnetic interaction in the first-neighbor shell, antiferromagnetic interaction in the second-neighbor shell, and which undergoes a ferromagnetic transition, is studied in the paramagnetic phase at the temperature  $1.1T_c$  using the Monte Carlo molecular-dynamics technique. The important quantities calculated are the dynamic structure function  $S(\mathbf{q},\omega)$  and the spin autocorrelation function  $\langle \mathbf{S}_i(0) \cdot \mathbf{S}_i(t) \rangle$ . Our results for  $S(\mathbf{q},\omega)$  show the existence of purely diffusive modes in the low- $q$  regime. For  $\mathbf{q}$  close to the zone boundary, our calculated  $S(\mathbf{q},\omega)$  shows a two-peaked or a multip peaked structure depending upon the magnitude and direction of  $\mathbf{q}$  and signifies damped propagating modes. This result disagrees with the theoretical predictions of Young and Shastry for all the principal directions and of Lindgard clearly for the  $\langle 100 \rangle$  direction. Our results for  $S(\mathbf{q},\omega)$  for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction is in fairly good agreement with the recent neutron scattering experiment of Böni *et al.*; however, our results for the  $\langle 100 \rangle$  direction somewhat disagrees with the experiment of Bohn *et al.* Our calculated auto-correlation function shows a diffusive behavior temporally.

### I. INTRODUCTION

Recent neutron scattering experiments on europium chalcogenides (EuO and EuS) by Böni and Shirane<sup>1</sup> and by Böni *et al.*<sup>2</sup> have provided valuable detailed information on the spin dynamics in these materials which are close to ideal realizations of the three-dimensional (3D) isotropic Heisenberg model. The only reliable way at present to calculate the spin dynamics of the Heisenberg model is the technique of the Monte Carlo simulation combined with the molecular dynamics (MCMD). In a previous paper (to be referred to as Ref. 3) we presented the results of the spin dynamics of the paramagnetic EuO, studied by using the MCMD technique. We present, in this paper, the resulting spin correlations for the paramagnetic EuS, which agree fairly well with the recent experimental results from neutron scattering experiments. We also present the spin autocorrelation function  $\langle \mathbf{S}_i(0) \cdot \mathbf{S}_i(t) \rangle$ , in the hope of stimulating further experiments involving local probes like PAC,  $\mu^+$ SR, and ESR.

As in the case of the paramagnetic EuO, the study of spin dynamics for the paramagnetic EuS also brings out the fact that the structure function  $S(\mathbf{q},\omega)$  has interesting and nontrivial structure in the paramagnetic phase and departs very much from Lorentzian or semi-Lorentzian (spin-diffusion) shape forced at small  $q$  by the global spin-conservation laws. Distinct peaks at finite values of  $\omega$  appear for large enough  $q$  and can be thought of as (damped) propagating modes. The shape of the  $S(\mathbf{q},\omega)$  curve for large  $q$  and the frequencies of the propagating modes obtained in our calculation are quite similar to those found in the neutron scattering experiment by Böni *et al.*<sup>2</sup> and Bohn *et al.* (those performed later),<sup>4</sup>

but are quite different from those obtained in the approximate analytical calculations of Young and Shastry (YS)<sup>5</sup> and also of Lindgard.<sup>6</sup> The observations of Böni *et al.*<sup>2</sup> on the single crystal form of EuS also differ from the predictions of both the YS theory and Lindgard's theory as well as from the earlier observations of Bohn *et al.*<sup>7</sup> To elaborate slightly on this point, in the case of the paramagnetic EuS for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction, the YS theory predicts nonexistence of distinct propagating modes even for temperature very close to the transition temperature  $T_c$ . Lindgard's theory predicts the existence of propagating modes only in the central region of the upper half of the magnetic zone in this case. The experimental results of Böni *et al.* show the existence of propagating modes quite clearly with a lot of structure in the constant  $\mathbf{q}$  scan of  $S(\mathbf{q},\omega)$  in the entire upper half of the magnetic zone for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction. In general, the observations of Böni *et al.* for  $S(\mathbf{q},\omega)$  seem to show much more structure than that seen in Bohn *et al.*'s observations.<sup>7</sup> However Bohn *et al.* reported their results for  $\mathbf{q}$  only along the  $\langle 100 \rangle$  direction. The results of the later experiments of Bohn *et al.*<sup>4</sup> performed for  $\mathbf{q}$  along the  $\langle 100 \rangle$  direction also display the existence of propagating modes for  $\mathbf{q}$  close to the zone boundary (ZB), with lots of structure in  $S(\mathbf{q},\omega)$ . Though the results of YS and of Lindgard for  $\mathbf{q}$  along the  $\langle 100 \rangle$  direction show the existence of propagating modes in the upper half of the magnetic zone, the resulting  $S(\mathbf{q},\omega)$  does not have as much structure as found in the experiments of Bohn *et al.*

These stimulating results of Böni *et al.*<sup>2</sup> and Bohn *et al.*<sup>4</sup> inspired us to calculate  $S(\mathbf{q},\omega)$  in the paramagnetic phase of the Heisenberg model, with parameters appropriate to EuS using the MCMD technique. As men-

tioned in Ref. 3, in this rare-earth chalcogenide compound which is an insulator, the magnetic ion  $\text{Eu}^{2+}$  forms a fcc lattice and the exchange interaction is confined to the first- and the second-neighbor shells only. The  $\text{Eu}^{2+}$  ion is in spin- $\frac{7}{2}$  state and the orbital effects are quenched completely since the magnetism is due only to the electrons of exactly half-filled  $f$  shell. In this system, the first-neighbor interaction is ferromagnetic, the second-neighbor interaction is antiferromagnetic, but the system orders ferromagnetically. We present in this paper the  $S(\mathbf{q}, \omega)$  results on an absolute scale for EuS at the reduced temperature  $T/T_c = 1.1$ . We compare these with the available experimental data as well as with the results of some approximate theories.

## II. BRIEF REVIEW OF APPROXIMATE THEORIES AND EXPERIMENTS RELATING TO EuS

Several analytic approximate techniques have been tried to probe the spin dynamics in the paramagnetic phase of 3D isotropic Heisenberg model. The details can be found in Ref. 3. Here for convenience we only briefly present the results of YS and of Lindgard, corresponding to EuS.

The calculation of YS for the paramagnetic EuS, shows no clear existence of propagating modes for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction. For the  $\langle 100 \rangle$  and  $\langle 110 \rangle$  directions, the YS theory predicts the existence of propagating modes close to the zone boundary and diffusive modes in the rest of the  $\mathbf{q}$  space. Unlike the case of the paramagnetic EuO, the existence of the central peak is not very clear in the propagating regime; rather, sometimes there seems to be the existence of a central dip.

For the paramagnetic EuS for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction, Lindgard's theory predicts the existence of propagating modes only in a narrow region of the upper half of the magnetic zone, with no propagating modes at and very close to the zone boundary. However, in the  $\langle 100 \rangle$  direction, the predicted behavior is very similar to that for the paramagnetic EuO. In the propagating regime of the  $\mathbf{q}$  space, the calculated  $S(\mathbf{q}, \omega)$  in the constant  $\mathbf{q}$  scans, show a two-peaked structure with no central peaks (rather the existence of a central dip) and peaks only at finite positive and negative values of  $\omega$ . In the diffusive regime of the  $\mathbf{q}$  space, the calculated  $S(\mathbf{q}, \omega)$  shows only a central peak.

Among the experimental techniques, inelastic neutron scattering has mostly been used to study spin dynamics of the paramagnetic EuS. Bohn *et al.*<sup>7</sup> were one of the first groups to carry out inelastic neutron scattering experiments on the paramagnetic EuS at various temperatures. The sample used was of single-crystal form. They measured  $S(\mathbf{q}, \omega)$  for  $\mathbf{q}$  along all the three principal directions. However, they report their results only for  $\mathbf{q}$  in the  $\langle 100 \rangle$  direction.<sup>7</sup> Their results for  $S(\mathbf{q}, \omega)$  in the constant  $\mathbf{q}$  scans along this direction, showing a three-peaked structure containing a central peak and two peaks at finite values of  $\omega$ , indicating the existence of damped propagating modes for  $\mathbf{q}$  close to the zone boundary. Their results for spin-wave dispersion relations in the ordered phase with  $\mathbf{q}$  along different directions display an appreciable amount of anisotropy in magnetic properties.

Later, Bohn *et al.*<sup>4</sup> again performed more careful high-resolution experiments on the paramagnetic EuS in the single-crystal form using a sample containing less absorbant  $^{153}\text{Eu}$  isotope and determining the contribution from nonmagnetic scattering much more accurately. Their results for  $S(\mathbf{q}, \omega)$  with  $\mathbf{q}$  along the  $\langle 100 \rangle$  direction seem to show a multi-peaked structure in the propagating regime of  $\mathbf{q}$ . However, in their experiment there was some amount of undesirable scattering from the ceramic glue, used to fix the mosaic pieces of the sample, the amount of which was impossible to estimate. Thus, the magnetic scattering cross section could not be measured for the values of  $\omega$  at and close to zero. Recently, Böni *et al.*<sup>2</sup> also performed neutron scattering experiments with high resolution on the paramagnetic EuS in the single-crystal form. They studied  $S(\mathbf{q}, \omega)$  for  $\mathbf{q}$  only along the  $\langle 111 \rangle$  direction. Their results for  $S(\mathbf{q}, \omega)$  in the constant  $\mathbf{q}$  scans show a multi-peaked structure like in Bohn *et al.*'s experiments, for  $\mathbf{q}$  close to the zone boundary, signifying damped propagating modes. In this large  $q$  regime, because of the presence of a huge amount of quasielastic nonmagnetic scattering from the glue, which could not be subtracted with sufficient accuracy, the shape of their observed  $S(\mathbf{q}, \omega)$  curves at  $\omega=0$ , and in general for  $|\hbar\omega| < 0.2$  meV, could not be ascertained properly. For low values of  $q$ , the subtraction of the nonmagnetic scattering could be done more reliably and the resulting  $S(\mathbf{q}, \omega)$  curves in the constant  $\mathbf{q}$  scans show only a central peak, indicating the existence of purely diffusive modes. Their experiments at  $T_c$ , measuring the dynamical critical exponent, confirmed the existence of anisotropy in this system.<sup>2,4</sup>

Until now, PAC or ESR or  $\mu^+$  SR experiments have not been performed on EuS; therefore, there are no experimental results for the spin autocorrelation function of EuS.

## III. THE MCMD APPROACH AND CALCULATIONS

In Ref. 3 we have described in detail the MCMD technique for studying the spin dynamics of the Heisenberg model. We followed exactly the same procedure for the paramagnetic EuS, as was followed in the case of the paramagnetic EuO. However, in the case of the paramagnetic EuS, since the temperature of interest was closer to  $T_c$ , viz., the temperature being  $1.1T_c$ , we stored the spin configurations from 5000 MC steps/spin MC age onwards, for carrying out the dynamical runs. This was done to take into account the slow relaxation of the system to thermal equilibrium at temperature close to  $T_c$ . As in the case of the paramagnetic EuO, here also we present our results for the dynamic structure function  $S(\mathbf{q}, \omega)$  and the spin autocorrelation function  $\langle \mathbf{S}_i(0) \cdot \mathbf{S}_i(t) \rangle$  obtained by using only the direct definition and  $1/t_{\max}$  definition.<sup>3</sup> We used the lattice of size  $12 \times 12 \times 12$  containing 6912 spins. We calculated  $S(\mathbf{q}, \omega)$  for  $\mathbf{q}$  along all the three principal directions. We used Windsor's<sup>38</sup> prescription to extract the quantum-mechanical estimate from our classical one for  $S(\mathbf{q}, \omega)$ . We compared our results with the experimental results of Böni *et al.* and Bohn *et al.* as well as with the results of

the YS theory and Lindgard's theory.

The values of  $J_1$  and  $J_2$  used in our calculation are those extracted from the experimentally obtained spin-wave dispersion curves at low temperature in the ordered phase.<sup>9</sup> They agree very well with those obtained from the *ab initio* calculations.<sup>10</sup> The value of  $T_c$  used in this calculation is the experimental  $T_c$ .<sup>9</sup> This value of  $T_c$  agrees very well with that obtained by the high-temperature series expansion results for the classical Heisenberg model.<sup>9,11</sup> It must however be noted that the value of  $T_c$  obtained from the YS calculation is slightly different from that used in our calculation. For comparing with our results we have calculated the corresponding quantities using the YS theory at the same value of  $T/T_c$  ratio, viz., 1.1. Lindgard's theory also gives a slightly different value of  $T_c$ . We have again compared our results with those of Lindgard for the same value of  $T/T_c$ , viz., 1.1.

All the simulation work reported in this paper was done in a Cyber 170/730 computer. Each dynamical run takes about 7 h of CPU time. The resolution width of our calculation is about the same as that in the experiments of Böni *et al.*, i.e., the half-width at half-maximum (HWHM) equal to 0.07 meV. For comparison with our results we have convoluted the corresponding YS results with the same resolution functions of the same widths as used in our calculations. The instrumental resolution width in the reported results of Bohn *et al.* in the experiments performed later<sup>4</sup> is exactly the same as that in the experiment of Böni *et al.*<sup>2</sup>

Since the reported results of Böni *et al.*<sup>2</sup> and of Bohn *et al.*<sup>4</sup> for  $S(\mathbf{q}, \omega)$  are not in the absolute scales, we have normalized them suitably for comparing with our results. The scheme used for normalization was as follows: at the temperature  $1.1T_c$  for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction for  $\mathbf{q} \equiv 0.66q_{ZB}$ ,  $\mathbf{q} \equiv 0.82q_{ZB}$ , and  $\mathbf{q} \equiv q_{ZB}$ , the normalization constants were determined by making our results and the results of Böni *et al.* coincide at the energy value of 0.50 meV. At the same temperature for  $\mathbf{q}$  along the  $\langle 100 \rangle$  direction, with  $\mathbf{q} \equiv 0.8q_{ZB}$ , the normalization constant was determined by making our results and the results of Bohn *et al.* coincide at the energy value of 1.20 meV. It may be pointed out that we could have chosen a different and probably a better normalization scheme by which we could have brought the curves of Böni *et al.* and of Bohn *et al.* much closer to our curves from the MCMD. All the curves from the convoluted YS are to be multiplied by a factor of 3, to bring them on absolute scale.

#### IV. RESULTS AND DISCUSSIONS

We first state the constants used:  $J_1 = 0.482$  K,  $J_2 = -0.204$  K,  $T_c = 16.57$  K, and a (lattice parameter)  $= 5.95$  Å;  $t_0$  (natural time unit)  $= 4.0 \times 10^{-12}$  sec, and  $t_{step}$  (iteration step size)  $= 0.010t_0$ .

In Figs. 1(a)–1(d), we display the theoretical as well as the experimental results for  $S(\mathbf{q}, \omega)$  with  $\mathbf{q} \equiv 0.3q_{ZB}$ ,  $\mathbf{q} \equiv 0.7q_{ZB}$ ,  $\mathbf{q} \equiv 0.8q_{ZB}$ , and  $\mathbf{q} \equiv q_{ZB}$ , respectively, along the  $\langle 111 \rangle$  direction, at the temperature  $T = 1.1T_c$ . At the same temperature we show the results for  $S(\mathbf{q}, \omega)$  with  $\mathbf{q}$  along the  $\langle 110 \rangle$  direction, in Fig. 2, with  $|\mathbf{q}| = q_{ZB}$

corresponding to the  $\langle 111 \rangle$  direction. The results for  $S(\mathbf{q}, \omega)$  with  $\mathbf{q}$  along the  $\langle 100 \rangle$  direction are shown in Fig. 3, with  $|\mathbf{q}| = q_{ZB}$  corresponding to the  $\langle 111 \rangle$  direction at the temperature  $T = 1.1T_c$ . The results for the spin autocorrelation function at the same temperature are shown in Fig. 4. From the examination of Figs. 1(a)–1(d) we see that our results differ drastically from those of the convoluted YS theory, in the large- $q$  regime along the  $\langle 111 \rangle$  direction. In this regime, our  $S(\mathbf{q}, \omega)$  shows a number of peaks at finite values of  $\omega$ , whereas the YS result shows none at finite  $\omega$ . Only at the zone boundary, the YS curve seems to show very faint shoulders at finite values of  $\omega$ . The qualitative features in our curves find a fairly good agreement with those in the experimental results of Böni *et al.*<sup>2</sup> in this regime of  $\mathbf{q}$ , suggesting the existence of damped propagating modes.

As mentioned earlier, the features of the results of Lindgard's calculations<sup>6</sup> (not shown in the figures) are different. Lindgard's calculations predict the existence of damped propagating modes only in the regime  $0.5q_{ZB} < |\mathbf{q}| < 0.7q_{ZB}$  for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction. Lindgard's calculations also show that the peak position of  $S(\mathbf{q}, \omega)$  for the propagating mode is at the maximum value of  $\omega$  for  $\mathbf{q} \approx 0.6q_{ZB}$ . In our calculation, as well as in Böni *et al.*'s observations, the maximum frequency of the most clearly resolved peak occurs not at the zone boundary. In our theoretical result, the peak is most prominent for  $\mathbf{q} = 0.8q_{ZB}$ . In the propagating regime of  $\mathbf{q}$ , our calculations show sometimes a two-peak (and sometimes a multi-peaked) structure for  $S(\mathbf{q}, \omega)$  with the existence of a central dip in contrast to Lindgard's result of occurrence of always a two-peak structure with a central dip for  $S(\mathbf{q}, \omega)$ . As mentioned earlier, the YS results show a central peak for  $S(\mathbf{q}, \omega)$  for all values of  $|\mathbf{q}|$  in this  $\mathbf{q}$  direction. In the propagating regime of  $\mathbf{q}$  space the results of Böni *et al.* indicate multi-peaked structure for  $S(\mathbf{q}, \omega)$ . In the low- $q$  regime, their experiment performed at  $T_c$  (not shown in the figures) shows the existence of only a central peak, as in our calculations as well as in calculations of YS and Lindgard (not shown in the figures), indicating the existence of purely diffusive modes. It may be mentioned that the positions of the peaks in  $S(\mathbf{q}, \omega)$  from our calculations and from the experimental results of Böni *et al.* are in good agreement for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction.

For  $\mathbf{q}$  along the  $\langle 110 \rangle$  direction, we have only our MCMD results and the YS results to compare, since there are no experimental data available and Lindgard also does not present these results. For  $\mathbf{q} \equiv 0.6q_{ZB}$  along  $\langle 110 \rangle$  we have  $|\mathbf{q}| = q_{ZB}$  appropriate to  $\langle 111 \rangle$ . For this particular value of  $|\mathbf{q}|$ , we find the features in the  $S(\mathbf{q}, \omega)$  curves quite different from those in the corresponding curves from our calculations and the convoluted YS calculations with the same value of  $|\mathbf{q}|$ , but for  $\mathbf{q}$  along the  $\langle 111 \rangle$  direction, as can be seen from Fig. 1(d) and Fig. 2. This shows the violation of isotropy. In the large  $q$  regime in general, both our calculations and the YS calculations show the existence of damped propagating modes, and the existence of a central peak. The YS calculation does not show the existence of a central peak very clearly (taking into account the resolution width of the calcula-

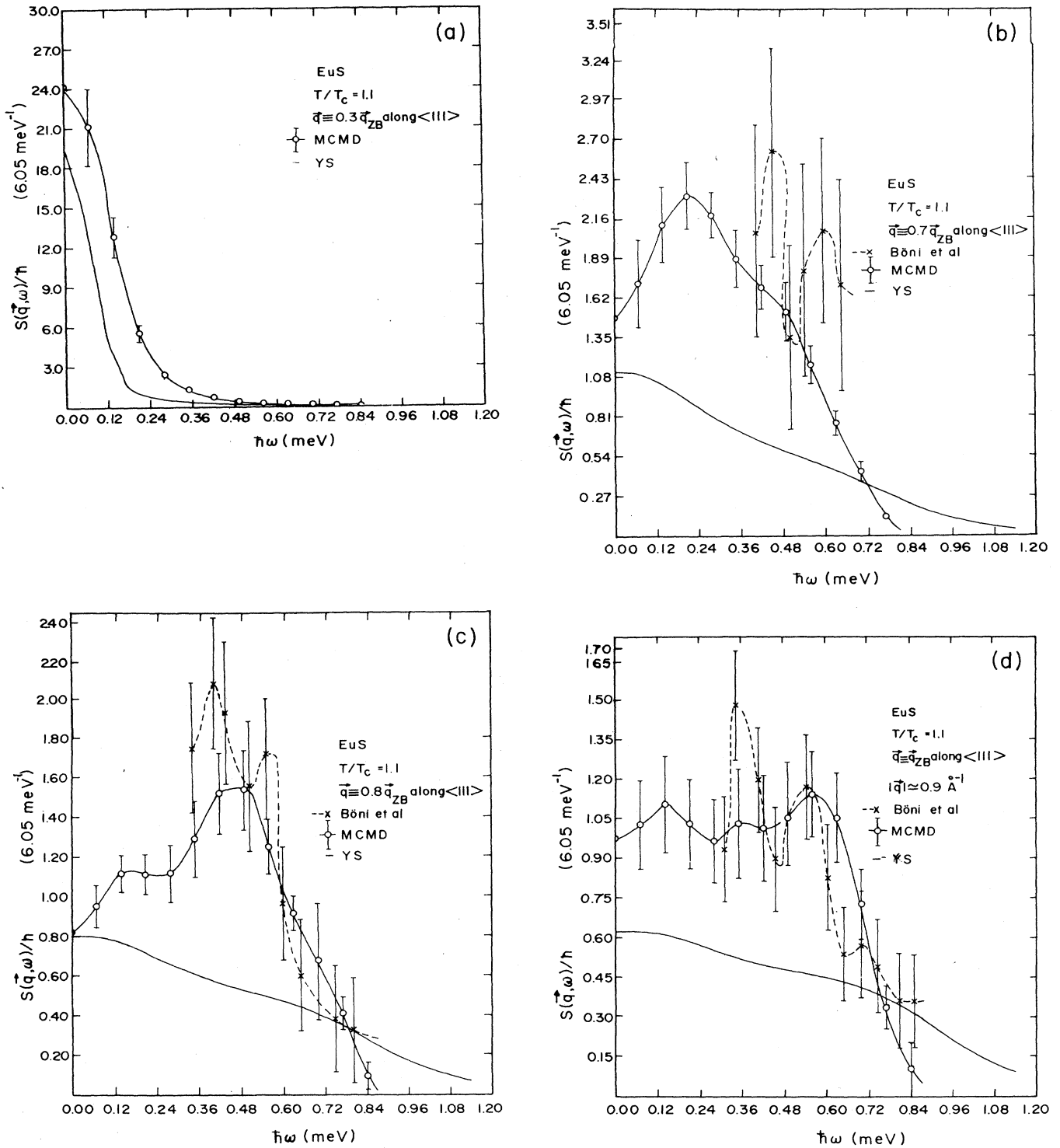


FIG. 1. (a) The plot of dynamic structure function  $S(\mathbf{q}, \omega)$  against energy  $\hbar\omega$  in constant  $\mathbf{q}$  scan, from MCMD and YS calculations; the YS curve is to be multiplied by a factor of 3. (b) The plot of dynamic structure function  $S(\mathbf{q}, \omega)$  against energy  $\hbar\omega$  in constant  $\mathbf{q}$  scan, from MCMD, YS calculations, and experimental results of Böni *et al.*; the YS curve is to be multiplied by a factor of 3. (c) The plot of dynamic structure function  $S(\mathbf{q}, \omega)$  against energy  $\hbar\omega$  in constant  $\mathbf{q}$  scan, from MCMD, YS calculations, and experimental results of Böni *et al.*; the YS curve is to be multiplied by a factor of 3. (d) The plot of dynamic structure function  $S(\mathbf{q}, \omega)$  against energy  $\hbar\omega$  in constant  $\mathbf{q}$  scan, from MCMD, YS calculations, and experimental results of Böni *et al.*; the YS curve is to be multiplied by a factor of 3.

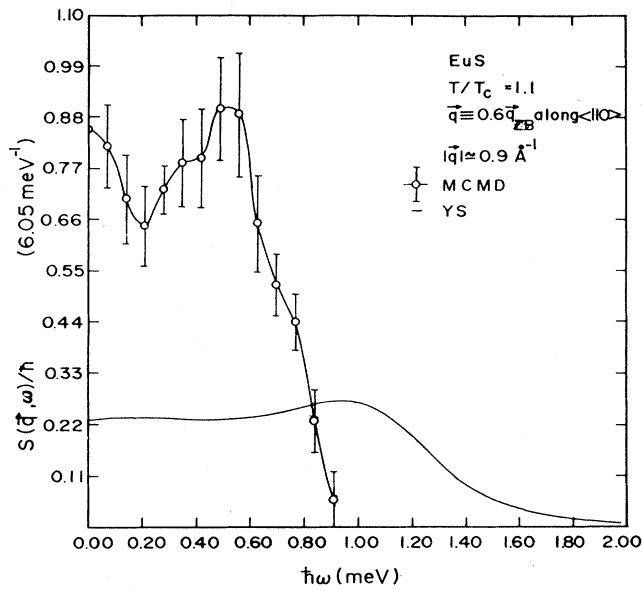


FIG. 2. The plot of dynamic structure function  $S(\mathbf{q}, \omega)$  against energy  $\hbar\omega$  in constant  $\mathbf{q}$  scan, from MCMD and YS calculation; the YS curve is to be multiplied by a factor of 3.

tion) but shows peaks at positive and negative values of  $\omega$  very close to  $\omega=0$  and in addition two more intense peaks at larger values of  $|\omega|$ . In the low- $q$  regime both our calculations and the YS calculations show only a central peak for  $S(\mathbf{q}, \omega)$ , implying the existence of purely diffusive modes (not shown in the figures).

Turning our attention to the  $\langle 100 \rangle$  direction, we again find that for large  $q$  our calculations, the convoluted YS calculations, and the experimental observation of Bohn

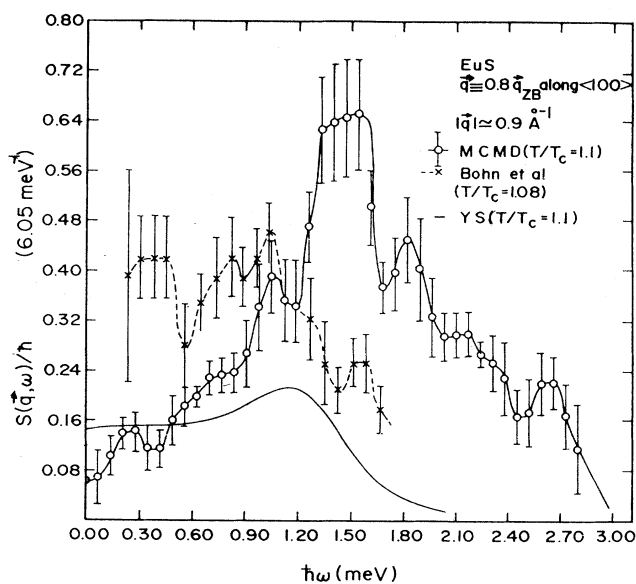


FIG. 3. The plot of dynamic structure function  $S(\mathbf{q}, \omega)$  against energy  $\hbar\omega$  in constant  $\mathbf{q}$  scan, from MCMD, YS calculations, and experimental results of Bohn *et al.*

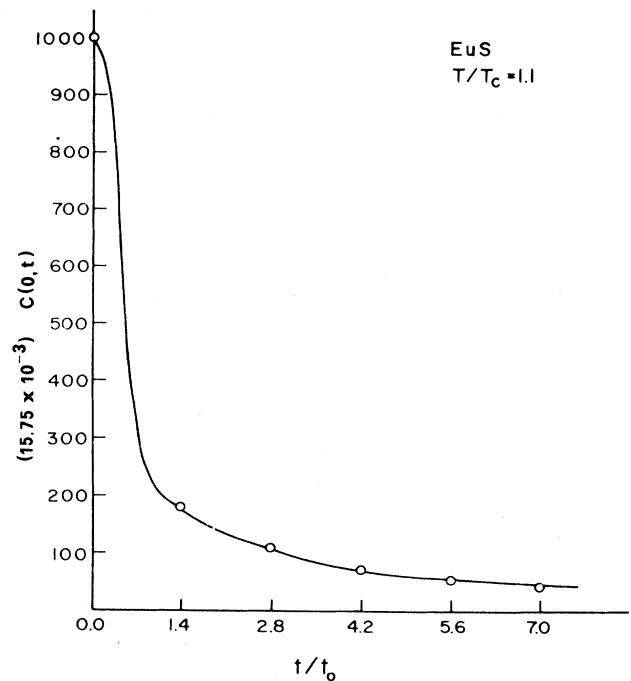


FIG. 4. The plot of spin autocorrelation function  $C(0, t)$  against time  $t$  (in reduced units) from MCMD calculation.

*et al.* (performed at the temperature  $1.08T_c$ ) all (see Fig. 3) show the existence of damped propagating modes. Comparing Fig. 3, Fig. 1(d), and Fig. 2 we see an appreciable difference in the shape and the features of the  $S(\mathbf{q}, \omega)$  curves from our calculations for  $\mathbf{q}$  along the  $\langle 100 \rangle$  direction, the  $\langle 110 \rangle$  direction, and the  $\langle 111 \rangle$  direction with same value of  $|\mathbf{q}|$  viz.  $q_{ZB}$  for the  $\langle 111 \rangle$  direction. This reconfirms the existence of anisotropy in the magnetic correlations in EuS. The YS calculations and the experimental results also support this. The propagating mode is most clearly resolved in the  $\langle 100 \rangle$  direction, but is at a significantly larger frequency than observed experimentally. Our calculated  $S(\mathbf{q}, \omega)$  in this re-

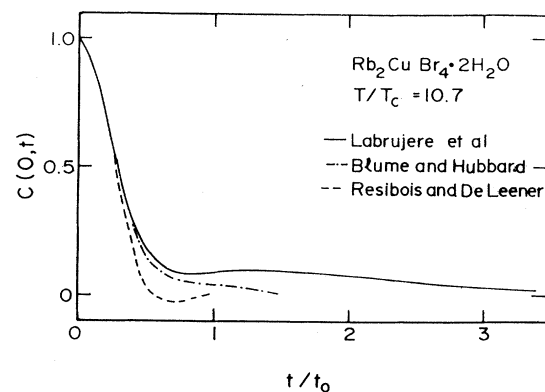


FIG. 5. The plot of spin autocorrelation function  $C(0, t)$  against time  $t$  (in reduced units).

gime shows a multi-peaked structure, as also found in the experimental observations of Bohn *et al.* The YS calculation does not show so much of structure in  $S(\mathbf{q}, \omega)$ . For low values of  $q$ , again we find the existence of only a central peak in the  $S(\mathbf{q}, \omega)$  from our calculations, the YS calculation, and the observations of Bohn *et al.* (not shown in the figures) implying the existence of diffusive modes. It may be noted that in general the magnitude of the error bars in our calculations are also very close to the corresponding experimental error bars.

Our results for the spin autocorrelation function of EuS at the temperature  $1.1T_c$  are displayed in Fig. 4. The qualitative features of the result are quite similar to those for EuO.<sup>3</sup> We again find a purely diffusive temporal behavior of the autocorrelation function. However, the damping is slower here compared to the case with EuO, where we had done the calculations at higher values of  $T/T_c$ . Our results for the spin autocorrelation function for both paramagnetic EuO (Ref. 3) and EuS bear a great similarity to the experimentally obtained spin autocorrelation function in the paramagnetic phase of  $\text{Rb}_2\text{CuBr}_4 \cdot 2\text{H}_2\text{O}$ , a 3D bcc ( $s = \frac{1}{2}$ ) Heisenberg ferromagnet with ferromagnetic interactions up to the second neighbor shell<sup>12</sup> (see Fig. 5). Our results also show similarity to the results from other approximate analytic theories with parameters appropriate to this system. However, for this spin- $\frac{1}{2}$  system, the spin autocorrelation function seems to show a slight propagating character as well, in contrast with our case.

For the paramagnetic EuS, the agreement between our results and the experimental results is not so good, as compared to the case with the paramagnetic EuO.<sup>3</sup> Our calculations show the existence of appreciable anisotropy in the magnetic correlations, assuming purely Heisenberg exchange interactions. The anisotropy can very well arise from the presence of competing exchange interactions. It may be worthwhile to mention that experimentally, the anisotropy observed is linked mainly to the presence of the dipolar interactions.<sup>2,4,7</sup> Our investigation, combined with the recent experimental results, seems to indicate the limitation of both the YS theory

and Lindgard's theory in describing the spin dynamics of the paramagnetic EuS accurately. This can be due to the breakdown of both the two-pole and the three-pole approximation in the case of competing exchange interactions. The failure of the YS theory may also imply the failure of the spherical model approximation in this case. In general, our calculations support the recent experiments, showing that the presence of competing exchange interactions in EuS makes the spin dynamics much more complicated in its paramagnetic phase than the one found in the case of the paramagnetic EuO. This is manifested in the frequent occurrences of the multi-peaked structures in  $S(\mathbf{q}, \omega)$  in the propagating regime of the  $\mathbf{q}$  space for EuS, in contrast to the occurrence of only the three-peaked structures in the corresponding case of EuO.

As for future plans, neutron scattering experiments should be performed for higher values of  $|\mathbf{q}|$  in the paramagnetic EuO and EuS to test our predictions more thoroughly. Also the experimental determination of the spin autocorrelation functions should provide a further check on our calculations. We would also like to study the dynamical scaling properties for both EuO and EuS, by simulation, to answer some of the interesting questions regarding the scaling regime in the  $\mathbf{q}$  space and the dependence of the scaling exponent on the directions in the  $\mathbf{q}$  space.<sup>1,2,4,13-15</sup> This would also help in understanding the role of dipolar interactions in these europium chalcogenides more clearly. Lastly, both theoretical and experimental work in paramagnetic EuSe for studying the spin dynamics would be quite instructive.

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