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Spin-wave modes and low-temperature specific heat in the spin-glass $Eu_xSr_{1-x}S$: $x = 0.54$ and 0.40

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We study the linear spin-wave modes and their contribution to the specific heat in the insulating spin-glass $Eu_xSr_{1-x}S$ for $x = 0.54$ and 0.40. The spin waves are identified with the collective excitations obtained by linearizing the equations of motion of the spins about their equilibrium orientations. The latter were determined by minimizing the energy by successive rotations of the spins into the directions of their local fields. The energies and localization indices of the modes were calculated by diagonalizing the dynamical matrices of ensembles of 270 ($x = 0.45$) and 345 ($x = 0.40$) spins with periodic boundary conditions. The densities of states have appreciable weight down to energies on the order of 0.1 K. The localization indices indicate that except for the high end of the spectrum nearly all the modes are delocalized. Comparison with experimental data indicates that the spin waves are responsible for seventy to eighty percent of the specific heat at $T \approx 0.3$ K with an even greater percentage expected at lower temperatures. The discrepancy between the experimental and theoretical values of the specific heat increases with increasing temperature, an effect which we attribute to the breakdown of the linear theory.

I. INTRODUCTION

One of the most challenging problems in the area of spin-glasses is to understand the character and distribution of the low-lying excitations. Broadly speaking, there are two classes of excitations.[1] One class is associated with multispin transitions between different equilibrium configurations. The existence of a large number of quasidegenerate configurations is believed to be the source of the anomalous ($\ln t$) relaxation of the remanent magnetization. In addition to the multispin transitions, in Ising spin-glasses there is also a spectrum of single-spin excitations which have energies equal to the Zeeman energies of the local fields.

In planar (easy-plane) or Heisenberg spin-glasses the situation is somewhat different in that the commutation relations of the spin components appearing in the Hamiltonian generate a dynamical coupling between the spins. The single-spin excitations are replaced by collective excitations analogous to spin waves in ferro- and antiferromagnets. It must be stressed that our classification of the modes should not be interpreted too strictly. At finite temperatures the distinction between the spin waves (we use the term in the general sense to describe the collective excitations which at low temperatures are obtained from the linearized equations of motion) and the multispin transition modes may break down for modes with energies $\ll kT$.[2]

The detailed study of the spin-wave modes in Heisenberg spin-glasses began with the work of Walker and Walstedt,[3] who calculated the spectrum of excitations in a classical model of CuMn. In their approach a local equilibrium configuration was obtained by successively rotating each spin into the direction of its local field. The modes were calculated by linearizing the equations of motion for the spins about the equilibrium orientations. Walker and Walstedt also calculated the magnetic contribution to the specific heat. Using a value for the exchange integral which was close to that inferred independently from experiment, they obtained reasonable agreement with the experimental data up to temperatures on the order of one half of the freezing temperature.

Calculations of the Walker-Walstedt type have also been reported for the Edwards-Anderson model[4] and PdMn.[5] In PdMn, as in CuMn, agreement was obtained between the calculated and the measured values of the specific heat. This result is somewhat surprising in view of the remarks made earlier about the existence of the other class of excitations.

Both CuMn and PdMn are metallic systems with long-range interactions between the spins. Recently it has been found that the insulator $Eu_xSR_{1-x}S$ shows canonical spin-glass behavior for $0.13 \lesssim x \lesssim 0.6$.[6,7] As in the metallic systems the spin-glass phase arises from a competition between ferromagnetic and antiferromagnetic interactions. However, in this case the interactions are primarily limited to nearest (ferromagnetic) and next-nearest (antiferromagnetic) neighbors.

Meschede *et al.*[8] have reported results for the specific heat of Eu_xSr_{1-x} for $x=0.40$ and 0.54. Because of the differences in the interactions in metallic and insulating systems, it is worthwhile to carry out an analysis of the specific heat of $Eu_xSr_{1-x}S$ similar to that undertaken for CuMn and PdMn. In this paper we report the results of our study of the spin-wave modes and their contribution to the specific heat in $Eu_{0.54}Sr_{0.46}S$ and $Eu_{0.40}Sr_{0.60}S$. Our approach is similar to that of Refs. 3–5 in that we first obtain a local equilibrium configuration by rotating the spins. We then calculate the spin-wave excitations by direct diagonalization of the dynamical matrix associated with the linearized equations of motion. The eigenvalues of the dynamical matrix are the spin-wave energies. From the eigenvectors we calculate the localization indices which provide information about the spatial extent of the modes.

The determination of the equilibrium configurations and the calculation of the energies and localization indices are discussed in Sec. II. In Sec. III we compare our findings for the specific heat with the experimental data reported in Ref. 8. We discuss our results and their implications in Sec. IV. In that section we also comment on the recent work of Krey,[9] who uses a continued fraction expansion to calculate the density of states and dynamic structure factor.

II. NUMERICAL STUDIES

A. Equilibrium configurations

Our numerical calculations were carried out on an fcc lattice of N sites with periodic boundary conditions. A fraction $xN \equiv N_S$ of sites, chosen at random, are occupied by Eu ions. A Heisenberg exchange interaction ($S=\frac{7}{2}$) was postulated:

$$\mathcal{H} = -2\sum_{i,j} J_{ij}\vec{S}_i \cdot \vec{S}_j , \qquad (1)$$

with nearest- and next-nearest-neighbor interactions. Guided by inelastic neutron scattering studies[10] we took

$$J_{nn} = 0.236\text{ K}, \qquad (2)$$

$$J_{nnn} = -0.118\text{ K}. \qquad (3)$$

As mentioned, equilibrium configurations were obtained by beginning with a random distribution of orientations and then rotating each spin in succession into the direction of its local field. The procedure was continued until the energy stabilized, remaining constant to one part in 10^8. This method leads only to a local minimum in the energy. However, our results for the density of states and specific heat showed little variation from configuration to configuration, which gives us some confidence that our findings would be unchanged were we to average over the full manifold of quasidegenerate ground states.

In Fig. 1 we show our results for the equilibrium magnetization, $\langle M\rangle$ defined by

$$\langle M\rangle = (N_S S)^{-1}\left[\left(\sum_{i=1}^{N_S} S_x^i\right)^2 + \left(\sum_{i=1}^{N_S} S_y^i\right)^2 + \left(\sum_{i=1}^{N_S} S_z^i\right)^2\right]^{1/2}, \qquad (4)$$

where N_S is the number of spins. The points are the averages taken over four configurations of 300–500 spins; the error bars denote the root mean square deviations. Although the magnetization fluctuates somewhat from configuration to configuration, it is evident there is a transition from a ferromagnetic to nonferromagnetic ground state in the interval between $x=0.3$ and 0.7.[11] As x approaches zero $\langle M\rangle$ decreases to 0.05, which is close to $N_S^{-1/2}$, the value appropriate to an ensemble of uncorrelated spins.

B. Density of states

The calculation of the spin-wave energies was carried out using the method outlined in Ref. 3. In Fig. 2 we show a histogram of the density of

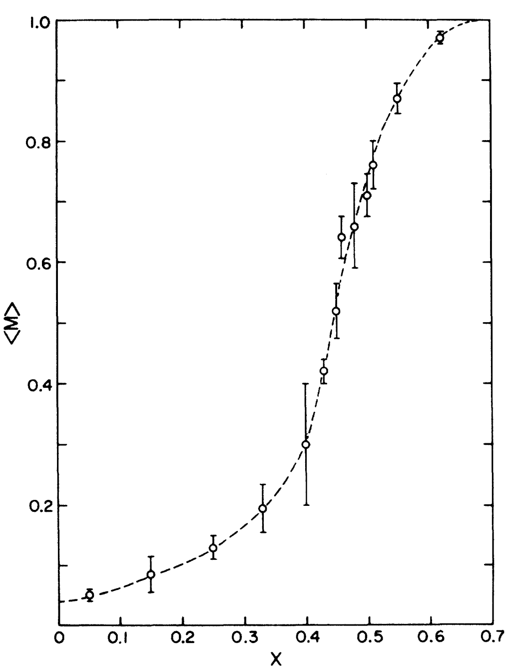

FIG. 1. $\langle M\rangle$ [defined in Eq. (4)] vs x, the Eu concentration in $Eu_xSr_{1-x}S$. The units are the mean values obtained by averaging over four configurations of 300–500 spins. The error bars are the rms deviations. The broken curve is a guide to the eye.

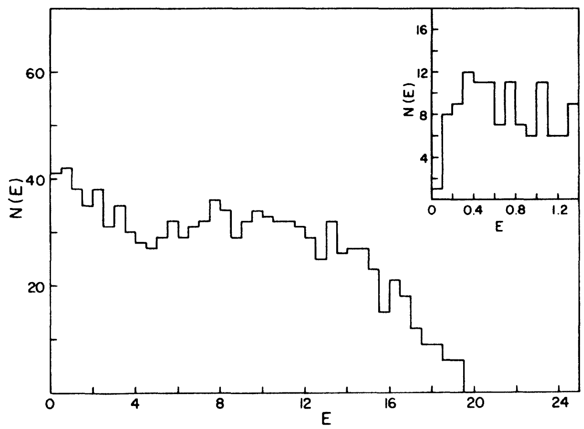

FIG. 2. Histogram of the density of states for $x=0.54$; 270 spins, four configurations. The inset shows the behavior at low energies. The energy is in kelvin.

states, $N(E)$, for $x=0.54$. The data were obtained from four configurations each with 270 spins. Relative to EuS the upper cutoff is shifted downward from 27 K to 19.5 K. The density of states builds up smoothly to a maximum in the vicinity of $E=0$. In the inset we show the behavior near $E=0$ on a finer scale. It is evident that the distribution drops rapidly below $E=0.2$ K and is not inconsistent with $\lim_{E\to 0} N(E)=0$. However, a nonzero value for $N(0)$ cannot be ruled out.

In Fig. 3 we show the corresponding results for $x=0.40$ obtained from three configurations each with 345 spins. As is shown in the inset, there is an increase in the number of modes near $E=0$ relative to $x=0.54$. The evidence is consistent with $N(0)$ being finite. Moreover, if $N(0)$ is zero the density of states must decrease rapidly at energies below 0.1 K. It is plausible that the difference in the low-energy behavior of $N(E)$ revealed in the insets in Figs. 2 and 3 is a consequence of the larger residual magnetization obtained for $x=0.54$. For this value of x $N(E)$ behaves qualitatively similar to $E^{1/2}$, the energy variation obtained for an ordered ferromagnet.

C. Localization indices

In parallel with Ref. 3 we have used the eigenvectors of the dynamical matrix to calculate the localization indices, L_μ, associated with the various modes. These are defined as

$$L_\mu = \sum_{j=1}^{N_S} W_{j\mu}^2 \Bigg/ \left(\sum_{j=1}^{N_S} W_{j\mu} \right)^2 , \tag{5}$$

where $W_{j\mu}$ is given by

$$W_{j\mu} = |\alpha_{j\mu}|^2 + |\beta_{j\mu}|^2 . \tag{6}$$

The $\alpha_{j\mu}$ and $\beta_{j\mu}$ are coefficients in the projection of the jth spin deviation, $\vec{n}_j$, on to $\vec{a}_j$ and $\vec{b}_j$, the local coordinate vectors perpendicular to the equilibrium orientation of the jth spin,[3] viz.,

$$\delta \vec{n}_j(t) = (\alpha_{j\mu}\vec{a}_j + \beta_{j\mu}\vec{b}_j) e^{i\omega_\mu t} + \text{c.c.} \tag{7}$$

Our results for the L_μ for single configurations with $x=0.54$ and 0.40 are shown in Figs. 4 and 5, respectively. It is evident that in both cases nearly all of the modes have localization indices near the minimum value N_S^{-1} for a completely delocalized mode having equal amplitude on all spins. For $x=0.54$ there are a few ($\approx 2\%$) modes with energies below 1 K which have localization indices greater than 0.02. Other modes for which $L_\mu > 0.02$ have energies greater than 17 K. In the case $x=0.40$ there is a large number of modes with indices greater than 0.02 but in no case is L_μ greater than 0.1.

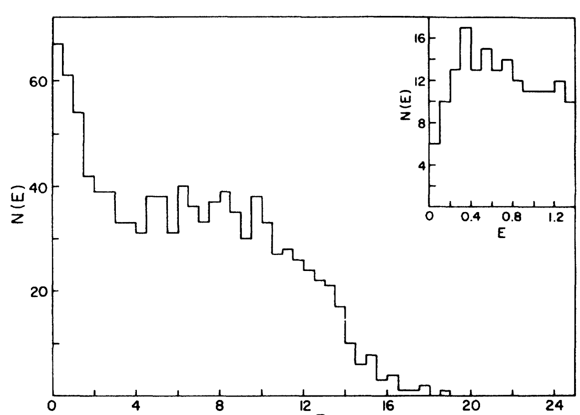

FIG. 3. Histogram of the density of states for $x=0.40$; 345 spins, three configurations. The inset shows the behavior at low energies. The energy is kelvin.

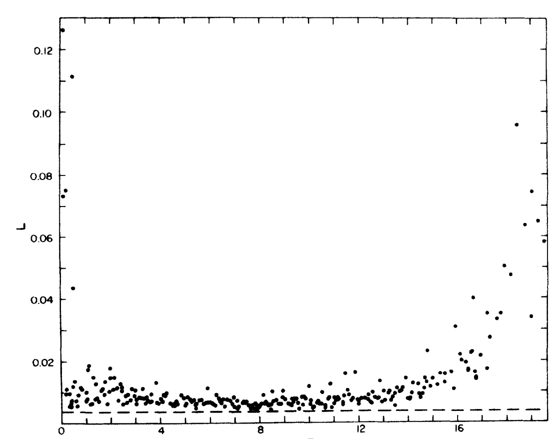

FIG. 4. Localization indices for $x=0.54$; 270 spins, one configuration. The broken line is N_S^{-1}.

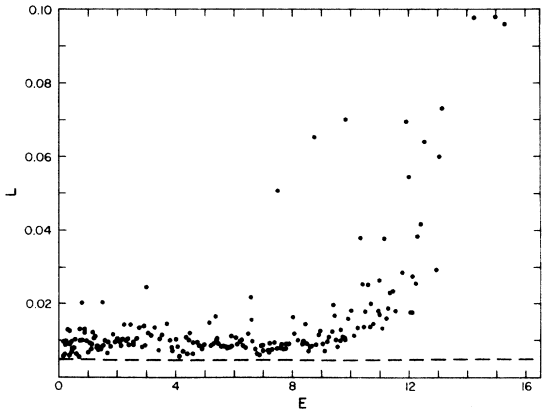

FIG. 5. Localization indices for $x = 0.40$; 200 spins, on one configuration. The broken line is N_S^{-1}.

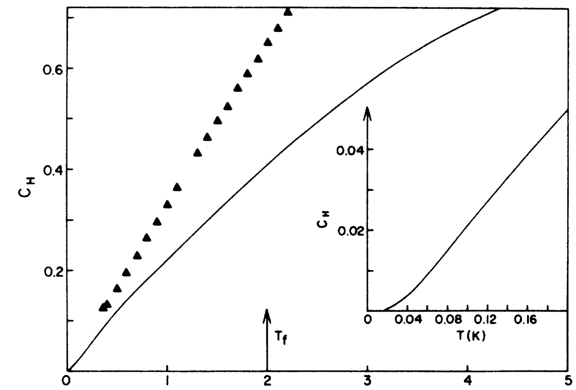

FIG. 6. Specific heat per spin vs T, $x = 0.54$; 270 spins, four configurations. The triangles are experimental data from Ref. 8. The inset shows the behavior at low temperatures. The arrow denotes the spin-glass freezing temperature.

The distribution of localization indices in $Eu_xSr_{1-x}S$ is quite similar to what was obtained for the Edwards-Anderson model with a Gaussian distribution of nearest-neighbor exchange integrals.[4] However, it differs considerably from the distribution calculated for CuMn.[3] In the latter case a large fraction of the modes are substantially localized ($L > 0.1$). Only the low-frequency modes have indices near N_S^{-1}. Presumably this difference is related to the range of the exchange interaction.

III. SPECIFIC HEAT

Numerical values of the specific heat per spin, C_H, were obtained from the equation

$$C_H = \frac{1}{N_S} \sum_{\mu} (E_\mu/T)^2 \frac{e^{E_\mu/T}}{(e^{E_\mu/T} - 1)^2}, \qquad (8)$$

where the E_μ are the energies (in K) of the N_S spin wave modes. In evaluating C_H for $T > 0.05$ K it was found that there was little variation in the results obtained from different configurations. Below 0.05 K the values were sensitive to the distribution of the lowest eigenvalues of the dynamical matrix, which varied from run to run.

In Fig. 6 we show our results for C_H for $x = 0.54$ along with the data from Ref. 8. From the figure it is evident that the spin waves make an important contribution to the specific heat at low temperatures. At $T = 0.4$ K the theoretical value of C_H calculated with no adjustable parameters is only 20% less than the experimental value. The inset shows the behavior of C_H for $T \leq 0.2$ K. Between 0.06 K and 0.3 K the specific heat can be approximated by $0.3(T - 0.03)$ (T in K). From Fig. 7 it is apparent that similar results were obtained for $x = 0.40$. At $T = 0.3$ K the theoretical value of the specific heat is 30% less than the experimental value while between 0.04 K and 0.3 K C_H is approximated by $0.45(T - 0.01)$ (T in K).

We conclude this section with two comments. First, the fact that the deviation between the measured and calculated values of C_H increases with increasing temperature could be taken as evidence of a contribution to the specific heat coming from other types of excitations. However, we think this is unlikely. In our opinion a more plausible explanation is that linear spin-wave theory is breaking down. It is expected that this breakdown first appears in the form of a temperature-dependent reduction of the spin-wave energies as is the case in ferro- and antiferromagnets. In support of this interpretation we note that a *reasonable* extrapolation of the data indicates that at $T = 0.1$ K, where the renormalization effects should be much smaller, the

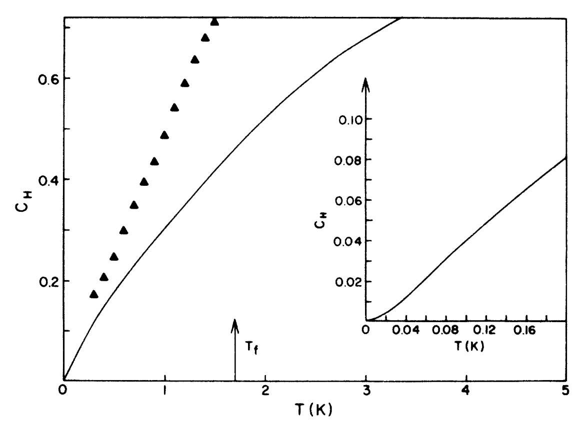

FIG. 7. Specific heat per spin vs T, $x = 0.40$; 345 spins, three configurations. The triangles are experimental data from Ref. 8. The inset shows the behavior at low temperatures. The arrow denotes the spin-glass freezing temperature.

linear theory accounts for essentially all of the specific heat.

The correspondence between the measured and calculated values of the specific heat in $Eu_xSr_{1-x}S$ is not quite as impressive as in CuMn and PdMn. In CuMn, as mentioned, there is good agreement up to one half of the freezing temperature.[3,12] In PdMn the agreement extends (probably fortuitously) all the way to the freezing point.[5] However, these results are colored by the fact that in both cases the experimentally determined exchange integrals were scaled by overall multiplicative factors on the order of 0.7–0.8 in order to make experiment and theory coincide at low temperatures.

IV. DISCUSSION

Our results along with those of Refs. 3 and 5 provide evidence that the spin-wave excitations make the dominant contribution to the magnetic specific heat at low temperatures in both metallic and insulating spin-glasses. In the case of $Eu_xSr_{1-x}S$ these modes are delocalized over nearly the entire band in contrast to CuMn where only the low-energy modes are delocalized. Even in finite clusters of $Eu_xSr_{1-x}S$ there are appreciable numbers of modes with energies as low as 0.2 K. The low-lying excitations give rise to a specific heat which varies approximately linearly with temperature below 0.3 K.

For $x=0.4$ our results for the density of states are in good agreement with the curve obtained by Krey[9] using a continued fraction expansion. This is evidence that the continued fraction method can provide reliable evidence about spin waves in Heisenberg spin-glasses. Krey has also calculated the dynamic structure factor appropriate to inelastic neutron scattering. In the case of $x=0.4$ he finds that the structure factor has a broad peak centered at a nonzero value of the energy. However, there is no evidence for well-defined propagating modes with a linear dispersion relation and quadratic damping which had been predicted for Heisenberg spin-glasses[13] and have been shown to be present in a model of a planar spin-glass.[1,14] It is possible that such modes exist in Heisenberg spin-glasses but nevertheless do not dominate the structure factor.[15]

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