Numerical studies of the field dependence of the magnetic specific heat of $Eu_{0.25}Sr_{0.75}S$ and $Eu_{0.54}Sr_{0.46}S$

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We use numerical simulation methods to determine the magnetic specific heat of $Eu_{0.25}Sr_{0.75}$ and $Eu_{0.54}Sr_{0.46}S$ as a function of field and temperature over the range $0.1 \le T \le 3$ K and $0 \le B \le 6$ T. For $x=0.25$ agreement with experiment is obtained at 1 K for $B > 1$ T; for $x=0.54$ agreement is obtained at 1 K in zero field and for fields above 1 T. Possible explanations for the discrepancy at low fields are advanced. The calculations are carried out in the independent-boson approximation so that as a by-product we obtain data on the field dependence of the distribution of magnon modes. The effect of fields above 3–4 T is to introduce a uniform shift in the magnon energies. At low fields the effect is more complicated as it involves a change in the relative distribution of the modes as well as a shift in their energies. Since the calculations involve no adjustable parameters, they provide additional confirmation of earlier results that magnons make the dominant contribution to the specific heat of Heisenberg spin glasses at low temperatures.

I. INTRODUCTION

The influence of applied magnetic fields on the various properties of spin glasses is proving to be a rich area for experimentation and theory. Recently, data have been presented on the variation of the specific heat of $Eu_xSr_{1-x}S$ with field and temperature.[1-3] Experimental studies of this compound are especially valuable since the microscopic exchange interactions are known.[4] As a consequence, quantitative comparisons can be made between theory and experiment, which are often impossible with metallic spin glasses. In the case of the specific heat, direct contact can be made at low temperatures between experimental results and the predictions of a noninteracting-boson model in which the excitations are magnons associated with small-amplitude spin oscillations.[5]

Previous calculations[6,7] showed that the boson model gave a good account of the zero-field data up to temperatures on the order of $0.5T_f$, where T_f is the freezing temperature as determined by the peak in the zero-field susceptibility. Recently, it was demonstrated that the model also worked well at high fields $B \ge 3$ T, where the ground state is such that essentailly all spins are aligned with the field.[3,8]

In this paper we also carry out calculations of the magnetic specific heat in the independent-boson approximation. Our work differs from that of Refs. 3 and 8 in that we treat low fields, $B < 3$ T, where the spins are not fully aligned. We also differ in that we calculate the density of states by the direct diagonalization of a dynamical matrix, whereas in Refs. 3 and 8 the density of states is determined by a continued-fraction technique. Generally speaking, however, both approaches yield comparable results for similar parameters.

In Sec. II we present our results for the density of states at various values of the field for $x=0.25$ and 0.54. The data on the field dependence of the specific heat are given in Sec. III. Section IV is devoted to a brief discussion of our findings.

II. DENSITY OF STATES

Our results for the density of states for $x=0.25$ and 0.54 are displayed in histogram form in Figs. 1 and 2. In each case the field values are $B=0$ T, 0.23 T, 1.15 T, 2.30 T, 4.61 T, and 10.5 T. The data shown in Fig. 1 ($x=0.25$) came from the analysis of three configurations each with 343 spins. The data in Fig. 2 ($x=0.54$) were obtained from four configurations with 270 spins. In all cases the equilibrium configurations and the magnon energies were obtained by general methods described elsewhere.[5,9] The only modifications brought about by the field are (1) the minimization algorithm involves rotation into the direction of the sum of the external and the exchange fields and (2) the dynamical matrix $P_{j,k}$ [Eq. (3.13) of Ref. 5] is augmented by the addition of the term

$$2\mu_B H \hat{\gamma}_j \delta_{jk}.$$

Here μ_B is the Bohr magneton and $\hat{\gamma}_j$ is the cosine of the angle the applied field H makes with the jth spin in its equilibrium orientation. All calculations were carried out using the values $J_1=0.22$ K and $J_2=-0.10$ K for the nearest- and next-nearest-neighbor exchange integrals, respectively.[4]

The data in Figs. 1 and 2 show that a gap in the density of states is induced at fields above 1–2 T. The gap appears at a point where nearly all the spins are aligned with the field. At higher values, 3–4 T, the effect of the field is to induce a uniform shift in the magnon energies. This behavior can be understood as follows. An examination of the dynamical matrix for a fully aligned array shows that

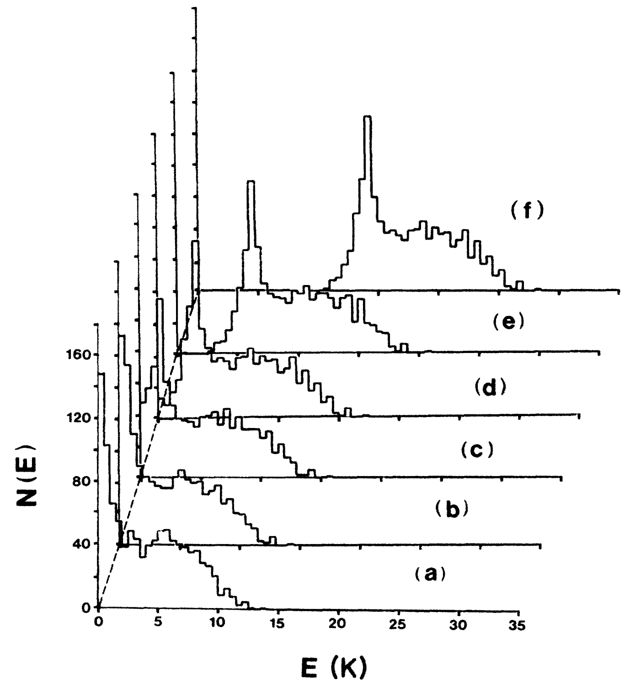

FIG. 1. Density of states for $x=0.25$. (a) $B=0$ T, (b) $B=0.23$ T, (c) $B=1.15$ T, (d) $B=2.30$ T, (e) $B=4.61$ T, (f) $B=10.50$ T. Energy is in Kelvin. Combined data from three configurations, each with 343 spins.

if $H(1)$ and $H(2)$ denote values of the field which are sufficient to produce complete alignment, then the shift in the eigenvalues is $2\mu_B[H(1)-H(2)]$. At low fields, where there is incomplete alignment, the change in the density of states cannot be described solely in terms of a shift but also involves a change in the relative distribution of the modes.

III. SPECIFIC HEAT

In the independent-boson approximation the magnetic specific heat is given by the expression

$$C_H=(1/kT^2)\int_0^\infty dE\, E^2\rho(E)[e^{E/kT}(e^{E/kT}-1)^{-2}],$$

where $\rho(E)$ denotes the density of states. Our results for the field dependence of the specific heat at 1 K are shown in Figs. 3 and 4, where we plot both theoretical and experimental data for $x=0.25$ and $x=0.54$. Here the experimental data are obtained from Ref. 3.

In the case of Fig. 3 ($x=0.25$) the theoretical data, shown as open circles, were obtained from three configurations each with 343 spins. The broken curve, drawn as a guide to the eye, indicates good agreement for fields above 1 T. At lower fields the agreement breaks down. Such a result is to be expected since T_f at this concentration is equal to 0.8 K. As mentioned, in zero field the theory is inapplicable for $T\gtrsim 0.5T_f$.

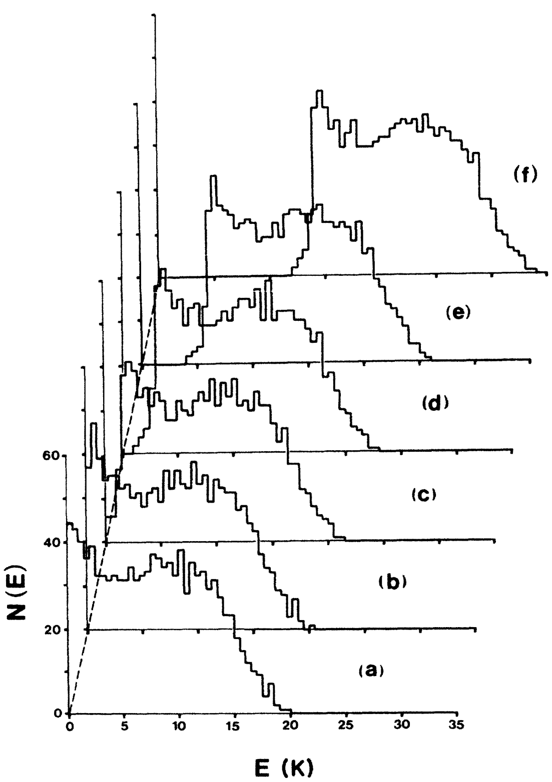

FIG. 2. Density of states for $x=0.54$. Labeling of the histograms the same as in Fig. 1. Combined data from four configurations, each with 270 spins.

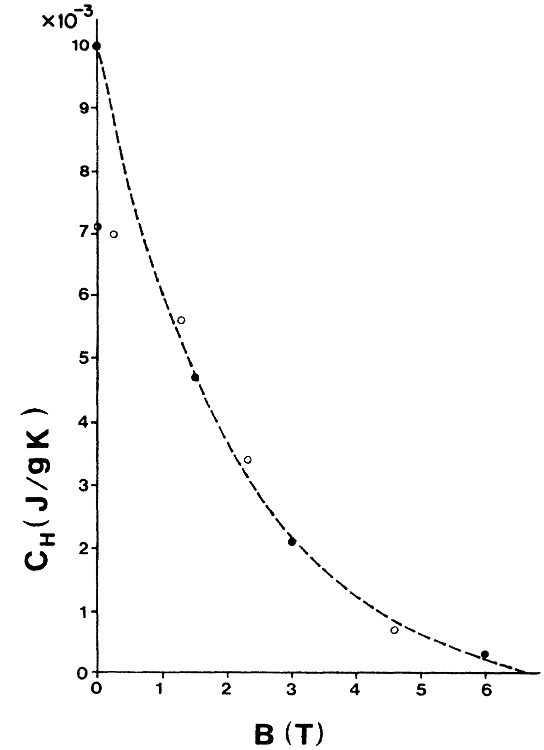

FIG. 3. Specific heat at 1 K vs applied field; $x=0.25$. The solid circles are experimental data from Ref. 3. The open circles are calculated values. Average over three configurations, each with 343 spins.

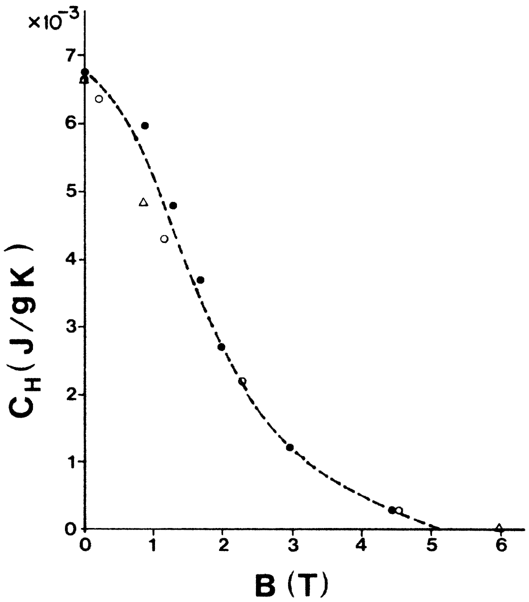

FIG. 4. Specific heat at 1 K vs applied field; $x = 0.54$. The solid circles are experimental data from Ref. 3. The open circles and open triangles are calculated values. Open circle: average over four configurations, each with 270 spins. Open triangle: average over three configurations, each with 467 spins.

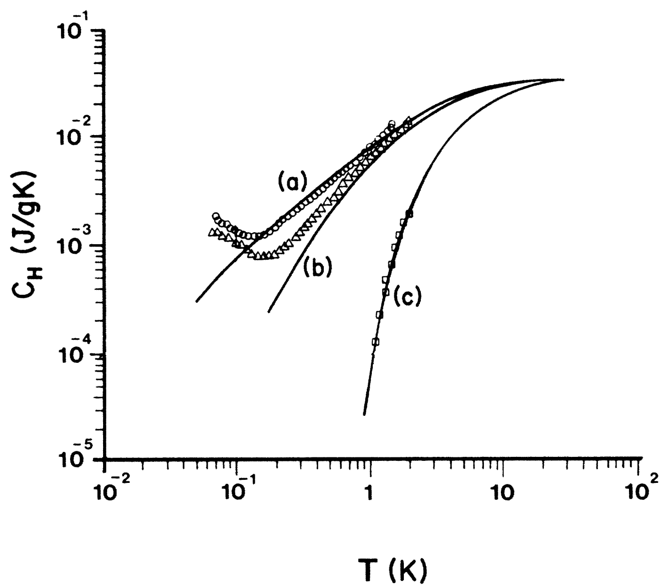

FIG. 5. Specific heat vs temperature; $x = 0.54$. (a) $B = 0$ T, (b) $B = 0.9$ T, (c) $B = 6$ T. The experimental data points are from Refs. 3 and 8. The theoretical curves were obtained by averaging over three configurations, each with 467 spins.

Our results for $x = 0.54$ are displayed in Fig. 4 along with the experimental data from Ref. 3. As in Fig. 3 the experimental data are represented by solid circles. The open circles are obtained from an analysis of four configurations, each with 270 spins; the open triangles are data from three configurations, each with 467 spins. The broken curve is a guide to the eye. There is good agreement between experiment and theory in zero field ($T_f = 2$ K) and above 1 T. Below 1 T the theoretical data fall below the experimental results. This discrepancy appears real since we obtain virtually identical results with both size arrays.

The temperature dependence of the specific heat for $x = 0.54$ is shown in Fig. 5 for $B = 0$ T, 0.9 T, and 6 T. The theoretical results were obtained from an analysis of three configurations, each with 467 spins. Apart from the upturn in the experimental data near 0.1 K, which is due to the contribution from the nuclei, there is good agreement between experiment and theory for both $B = 0$ T and $B = 6$ T. The data for $B = 0.9$ T show a deviation between experiment and theory below 0.5 K, consistent with the low-field results shown in Fig. 4.

IV. DISCUSSION

In assessing the findings reported in Secs. II and III it is important to keep in mind that the theoretical results were obtained with no adjustable parameters. As such they provide direct evidence of the appropriateness of the independent-boson model for the magnetic specific heat of spin glasses with Heisenberg interactions—a result which is consistent with earlier findings for *Cu*Mn[5] and $Cd_{1-x}Mn_xTe$.[10] In the case of $x = 0.25$ we have noted that the breakdown in the theory reflects the failure of the independent-boson approximation at high temperatures and low fields. For $x = 0.54$ the most noticeable discrepancies between experiment and theory occur at low but finite fields, $0 < B \lesssim 1$ T. There appear to be at least two possible explanations for this. It can happen that our minimization procedure does not generate appropriate equilibrium configurations at low fields. Also, since the calculated values of the specific heat lie below the measured values, it may be the case that excitations other than magnons, e.g., tunneling modes, are making a non-negligible contribution in the low-field regime.

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[1]H. v. Lohneysen, R. van der Berg, G. V. Lecomte, and W. Zinn, Phys. Rev. B **31**, 2920 (1985).
[2]H. v. Lohneysen, R. van der Berg, J. Wosnitza, G. V. Lecomte, and W. Zinn, J. Magn. Magn. Mater. **54–57**, 189 (1986).
[3]J. Wosnitza, H. v. Lohneysen, W. Zinn, and U. Krey, Phys. Rev. B **33**, 3436 (1986).
[4]H. G. Bohn, W. Zinn, B. Dorner, and A. Kollmar, Phys. Rev. B **22**, 5447 (1980).
[5]L. R. Walker and R. E. Walstedt, Phys. Rev. B **22**, 3816 (1980).
[6]W. Y. Ching, D. L. Huber, and K. M. Leung, Phys. Rev. B **21**, 3708 (1980).
[7]U. Krey, Z. Phys. B **38**, 243 (1980); **42**, 231 (1981); J. Magn. Magn. Mater. **28**, 231 (1982).
[8]U. Krey, J. Phys. (Paris) Lett. **46**, L845 (1985).
[9]W. Y. Ching, D. L. Huber, and K. M. Leung, Phys. Rev. B **23**, 6126 (1981).
[10]W. Y. Ching and D. L. Huber, Phys. Rev. B **30**, 179 (1984).