Magnetic excitations in $Eu_xSr_{1-x}S$ in high magnetic fields

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We report on measurements of the specific heat of $Eu_xSr_{1-x}S$ ($0.25 \leq x \leq 0.8$) in magnetic fields up to 6.6 T and for temperatures between 1.5 and 30 K. The magnetic specific heat in zero field and in high fields ($B \geq 3$ T) can be quantitatively accounted for in terms of magnonlike excitations without any fit parameter. The density of states $g(E)$ of these excitations is calculated numerically by treating them as noninteracting bosons. The very good agreement between experiment and calculation includes the temperature range of our earlier measurements down to 0.1 K, and extends in high fields up to temperatures close to the freezing temperature or Curie temperature for spin-glass and ferromagnetic samples, respectively.

I. INTRODUCTION

It is well known that spin-glasses exhibit a quasilinear magnetic specific heat over a wide temperature range from the lowest temperatures up to temperatures slightly above the spin-glass freezing temperature T_f. Typically, about one third of the total magnetic entropy is attained by the spin-glass when $T \approx T_f$. The nature of the excitations leading to this large specific heat is, however, not clear, despite considerable experimental and theoretical work. Spin-wave-like excitations have been suggested repeatedly for metallic[1] and nonmetallic spin-glasses.[2,3] Here we focus on the nonmetallic $Eu_xSr_{1-x}S$ spin-glass which has received much attention recently.[4,5] In this system, the magnetic interactions occur via superexchange between nearest (NN) and next-nearest Eu neighbors (NNN) with $J_1/k_B = 0.22$ K and $J_2/k_B = -0.11$ K, respectively. Longer-range interactions can be neglected. A variety of magnetic phases are found at low temperatures when changing the Eu concentration x. For $x < x_p$, superparamagnetism of isolated Eu clusters is observed. $x_p = 0.136$ is the site percolation threshold for an fcc lattice with respect to NN and NNN. For $x_p < x < x_c = 0.51$, spin-glass behavior is found. For $x_c < x < 0.7$ the system is in a reentrant or "frustrated" ferromagnetic state. Finally, for $x > 0.7$, $Eu_xSr_{1-x}S$ behaves like a simple diluted Heisenberg ferromagnet.

Recently, we reported[6] on the specific heat in high magnetic fields of $Eu_xSr_{1-x}S$ with $x = 0.25$ and $x = 0.54$, i.e., for samples in the spin-glass and reentrant-ferromagnetic regimes. With increasing (moderate) field B, the magnetic specific heat C_M was progressively reduced, mainly at low temperatures, resulting in a gradual increase of the slope of the C_M versus T curve from the quasilinearity in zero field. For larger fields, a very strong decrease of C_M towards lower temperatures was found, with a roughly exponential dependence of C_M on T. These results were interpreted in terms of a gradual opening of a gap in the magnetic excitation spectrum. A strong decrease of the apparent nuclear specific heat C_N with increasing B at very low temperatures occurs concomitantly with the opening of the gap. This has been attributed to a strong increase of the nuclear spin-lattice relaxation time.[6]

The numerical calculations[3] of the magnetic excitation spectrum for $Eu_xSr_{1-x}S$ obtained for $B = 0$, have been extended recently to high magnetic fields.[7] Good agreement was found between theory and our low-temperature experimental data, and also with data of other authors for a sample with $x = 0.44$.[8] In high fields, the approximation of noninteracting bosons works well up to rather high temperatures. It seemed, therefore, worthwhile to extend our previous high-field specific-heat measurements[6] to higher temperatures, and also to investigate more samples, in particular towards the ferromagnetic side of the magnetic phase diagram.

In this paper, we present the results of a comprehensive specific-heat study for samples with x between 0.025 and 0.8. The paper is organized as follows. Sec. II contains the experimental details and Sec. III the results. In Sec. IV, a discussion of the theoretical calculations is presented which are compared with the experimental results. The main conclusions are summarized in Sec. V.

II. EXPERIMENTAL DETAILS

The samples used in this investigation were single crystals. First, powder samples of the desired composition

were prepared chemically via mixed oxalates of Eu^{3+} and Sr^{2+}. The obtained powder was melted in a sealed tungsten crucible at about 2500 °C in order to grow single crystals. The specific heat was measured between 1.5 and 30 K with the standard heat pulse method. Because of the rather small sample size (a few mm^3), the samples were attached to a sapphire plate which carried a thin-wire manganin heater and an Allan-Bradley carbon resistance thermometer ground down to a mass of a few mg. The thermometer was calibrated in various magnetic fields against a calibrated[9] carbon-glass thermometer located in the low-field region of the magnet. The sapphire plate was positioned with nylon threads in a Cu frame which was attached to the cold sink. For some measurements, a 20 μm Cu wire between the sample and the cold sink served as an additional thermal weak link (besides the nylon threads and electrical leads).

III. RESULTS

Figures 1–4 show the specific heat C of four $Eu_xSr_{1-x}S$ samples between 1.5 and 30 K in various applied magnetic fields. For the spin-glass sample with $x=0.25$ ($T_f=0.8$ K) and the reentrant-ferromagnetic sample with $x=0.54$ ($T_c=5$ K and $T_f=2$ K), the earlier data taken at lower temperatures[6] are also included to facilitate a comparison (Figs. 1 and 2). In general, the overlap between the low-temperature and the present high-temperature data is quite good; the data join rather smoothly. The small bump observed for $x=0.54$ in zero field at 1.5 K is probably an artifact of the thermometer calibration. The quasilinear specific heat for the two samples with $x=0.25$ and 0.54 in zero field extends to temperatures above the freezing temperature, as found previously.[10] C passes a broad maximum for $x=0.25$ and 0.54 at $T\approx 2T_f$. The rise of C at the highest temperatures is, of course, due to the Debye T^3 contribution becoming dominant well above T_f. The upturn of C below 0.2 K towards low temperatures is due the nuclear specific-heat contribution C_N arising from the hyperfine splitting of ^{151}Eu and ^{153}Eu nuclei.[6]

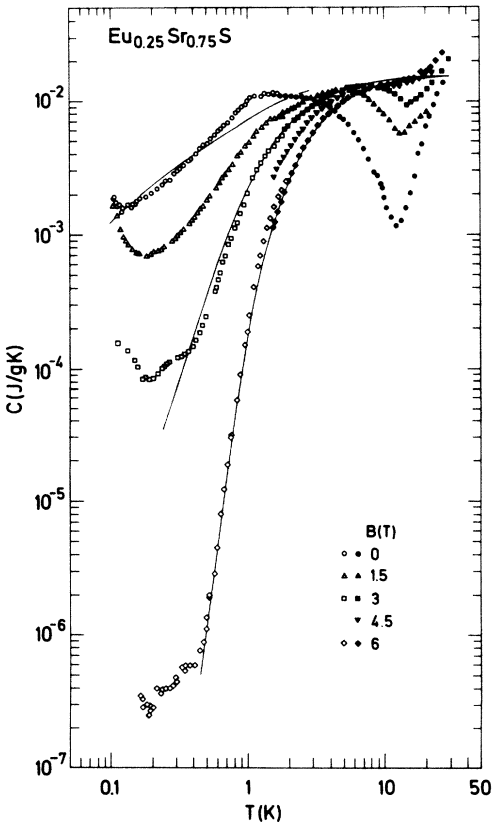

FIG. 1. Specific heat C of $Eu_{0.25}Sr_{0.75}S$ as a function of temperature T for different applied magnetic fields B. Solid lines indicate the calculated specific heat (see Sec. IV) for $B=0$, 3, and 6 T.

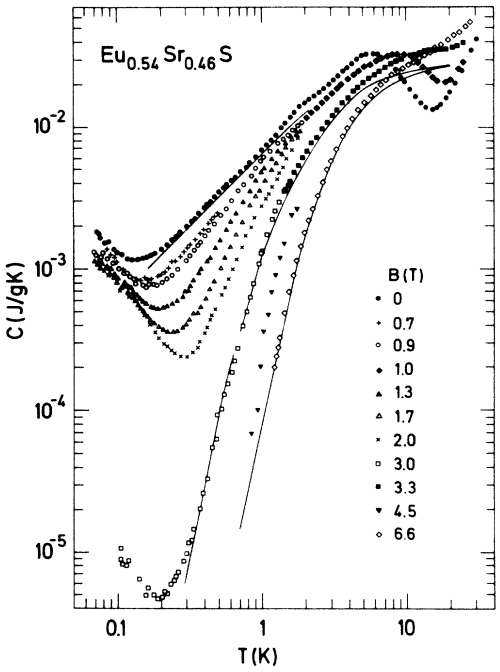

FIG. 2. Specific heat C of $Eu_{0.54}Sr_{0.46}S$ as a function of temperature T for different applied magnetic fields B. Solid lines indicate the calculated specific heat for $B=0$, $B=3$ (3.3) T for $T<0.7$ K (>0.7 K), and 6.6 T.

The zero-field data for the ferromagnetic samples $x=0.7$ and $x=0.8$ (Figs. 3 and 4) indicate a $T^{1.1}$ and $T^{1.3}$ dependence, respectively, well below their Curie temperatures T_c. The data of these samples are in good agreement with earlier zero-field data,[11] including the presence of sharp maxima at T_c. However, the earlier data had been analyzed for temperatures just below T_c in terms of a $T^{3/2}$ law expected for the spin-wave specific heat of ferromagnets. In our view, such a $T^{3/2}$ behavior just below T_c is fortuitious and can be traced back to the presence of the small bump found in pure EuS (Ref. 12) and also in moderately diluted samples (e.g., around 5 K for $x=0.8$; see Fig. 4). It is expected that the limiting $T^{3/2}$ law for long-wavelength ferromagnetic spin-waves will be visible only below 1 K for our ferromagnetic samples. The rise of C well above T_c is again due to the Debye contribution.

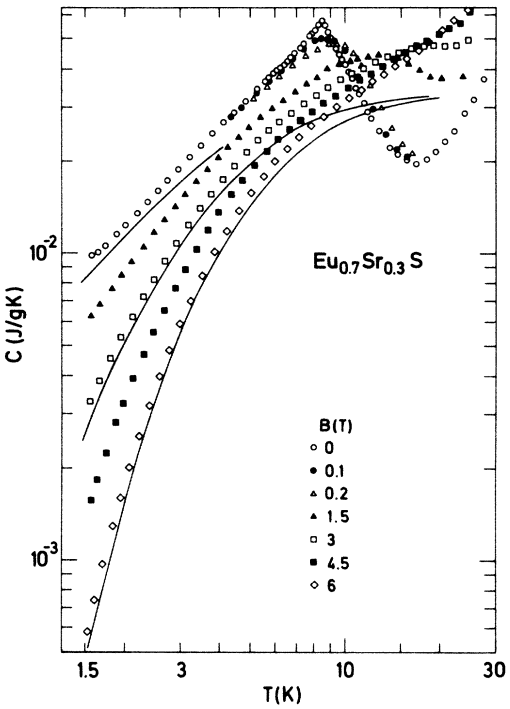

FIG. 3. Specific heat C of $Eu_{0.7}Sr_{0.3}S$ as a function of temperature T for different applied magnetic fields B. Solid lines indicate the calculated specific heat for $B=0$, 3, and 6 T.

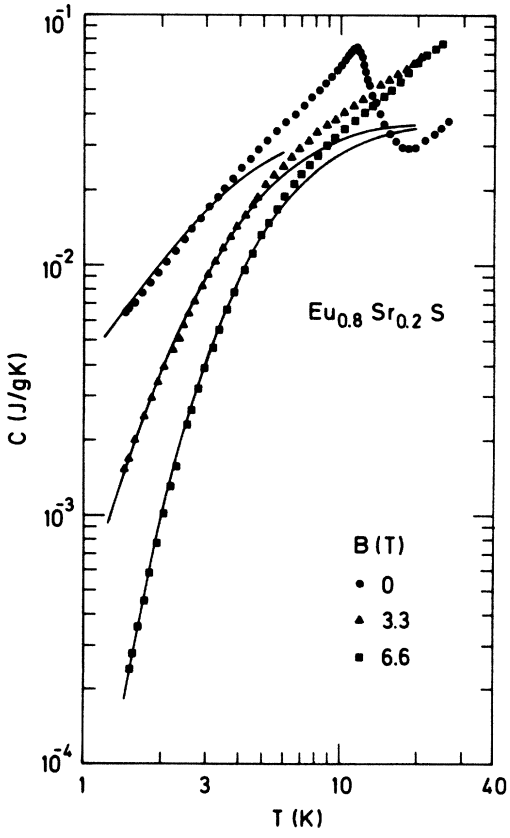

FIG. 4. Specific heat C of $Eu_{0.8}Sr_{0.2}S$ as a function of temperature T for different applied magnetic fields B. Solid lines indicate the calculated specific heat for $B=0$, 3.3, and 6.6 T.

Turning to the magnetic field dependence, an overall similar strong depression of C is found at lower temperatures, with a crossing over towards higher temperatures where the specific heat is increased in a magnetic field. The opening of a gap in the magnetic excitation spectrum for high magnetic fields, previously observed for samples in the spin-glass range,[6] is seen to occur also for higher-concentration samples $x=0.7$ and 0.8. From plots of $\ln C$ versus $1/T$ (not shown) we can obtain the energy gap ΔE, as demonstrated earlier.[6] The present new data, although taken only above 1 K for the ferromagnetic samples, suggest that ΔE is close to $g\mu_B B$ for these samples, in contrast to the spin-glass sample where $\Delta E < g\mu_B B$ was found.[6] The decrease of the apparent nuclear specific heat with increasing magnetic field has been discussed before.[6]

IV. DISCUSSION

A. Density of states of magnon excitations: Theory

For $B=0$ and $T \ll T_f$, already the early specific-heat results of Meschede *et al.*[10] could be explained in terms of magnonlike excitations which were treated in the random phase approximation, i.e., as noninteracting bosons. This corresponds to the following formula for the magnetic specific heat C^* per Eu atom:

$$C^*/k_B = \int_0^\infty dE\, g(E) \left[\frac{E}{k_B T}\right]^2 \frac{e^{E/k_B T}}{(e^{E/k_B T}-1)^2} , \quad (1)$$

where $g(E)$ is the magnon density of states normalized to yield $\int_0^\infty g(E)dE=1$. Equation (1) should apply as long as the local thermal spin fluctuations remain small compared to the total spin $\hbar S$ with $S=\frac{7}{2}$ for Eu^{2+}. For strong magnetic fields, in contrast to $B=0$, this condition will be fulfilled up to temperatures larger than T_f. For instance, in $B=6$ T the validity range extends up to ≈ 7 K for $x=0.44$ [$T_f=1.8$ K (Ref. 8)] as has been demonstrated by one of the present authors.[7] Furthermore, for strong fields the calculation of $g(E)$ is facilitated by the fact that in the ground state all spins will be aligned by the field. In this case the spin-wave Hamiltonian from which $g(E)$ has to be calculated, is simply

$$\mathcal{H}_{SW} = \sum_{l,m} H_{l,m} \, |l\rangle\langle m| \, , \quad (2)$$

with

$$H_{l,m} = 2S\left[\sum_p J_{l,p}\delta_{l,m} - J_{l,m}\right] + g\mu_B B\delta_{l,m} \, . \quad (3)$$

Here l and m enumerate the Eu sites; $|l\rangle$ is a state where at the site l the z component of the spin is reduced from S to $S-1$, and $J_{l,m}$ are the exchange integrals, namely $J_{l,m}/k_B \equiv J_1/k_B = 0.22$ K for nearest neighbors and $\equiv J_2/k_B = -0.11$ K for next-nearest neighbors. Furthermore, for Eu^{2+} the Landé factor is $g=2$, and k_B and μ_B have their usual meanings. Thus, for the present system all relevant parameters are known, and $g(E)$ can be calculated numerically.

The calculation of $g(E)$ has been performed by means of a continued fraction algorithm[3] for an fcc model sys-

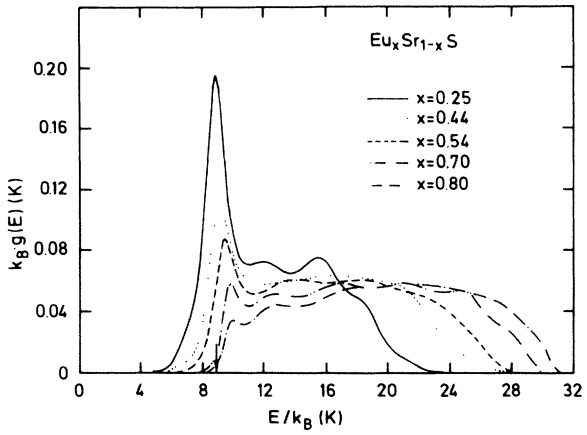

FIG. 5. Spectral density of states $g(E)$ per Eu atom for magnetic excitations in $Eu_xSr_{1-x}S$ for different concentrations x in a field of 6.6 T. The arrow indicates the Zeeman energy $E_B=g\mu_B B=k_B(8.87\ \mathrm{K})$.

tem with 20^3 sites which are randomly occupied by a fraction x of Eu atoms. As usual, periodic boundary conditions have been used. As an example, the resulting $g(E)$ is presented in Fig. 5 for an applied magnetic field of $B=6.6$ T for various concentrations x. The arrow denotes the Zeeman energy $E_B=g\mu_B B$. Three obvious trends with regard to the concentration dependence of $g(E)$ can be inferred from Fig. 5: (i) With decreasing x, the energy range of $g(E)$ shrinks, i.e., the upper band edge decreases. (ii) Simultaneously, a pronounced peak appears around E_B. (iii) Finally, the lower band edge is separated from $E=0$ by an energy gap.

The magnitude ΔE of the energy gap depends on B and x as

$$\Delta E=g\mu_B[B-B_0(x)]\ , \tag{4}$$

where the effective field $B_0(x)$ vanishes for $x=1$ and remains small for $x\geq 0.7$. In the reentrant-ferromagnetic and spin-glass regimes, $x<0.7$, B_0 increases with decreasing x. For instance, for $x=0.25$, B_0 becomes ≈ 2.6 T. This general trend in the calculation agrees well with our low-temperature experimental results already reported earlier.[6] Of course, as long as B remains larger than B_0, a change in the magnetic field would simply correspond to a rigid shift of the $g(E)$ curves in Fig. 5.

For $B\ll B_0$, the calculation of $g(E)$ is much more complicated, since the magnetic moments are no longer aligned. In this case, after the preparation of the ground-state by a Monte Carlo procedure, $g(E)$ is obtained by the method described previously.[3]

B. Specific heat: Comparison between theory and experiment

With the knowledge of $g(E)$, the magnetic specific heat is calculated from Eq. (1). The results are indicated by the solid lines in Figs. 1–4. As noted above, no adjustable parameters enter, since J_1 and J_2 are well-known from a number of other experiments.[4] The agreement with the experimental data is indeed very good except at higher temperatures where the noninteracting-boson approximation breaks down.[7] As already stressed above, the validity of the calculation in high magnetic fields extends to temperatures well above the freezing temperature for the spin-glass samples, and to roughly the Curie temperature for ferromagnetic samples, while the validity range of the zero-field calculation is much more limited.

Also, the opening of a gap ΔE can be quantitatively accounted for. Experimentally, ΔE was found[6] to be somewhat smaller than $g\mu_B B$, in particular in the spin-glass range. For instance, ΔE for $x=0.25$ was found to obey Eq. (4) with $B_0=1.4$ T. The present data for samples with $x\geq 0.7$ indicate that $\Delta E\approx g\mu_B B$, i.e., $B_0=0$. Hence, the calculated concentration dependence of $B_0(x)$ is quite closely observed.

V. CONCLUSIONS

It has been demonstrated that for strong magnetic fields the magnetic specific heat of $Eu_xSr_{1-x}S$ crystals is completely determined by magnonlike excitations up to rather high temperatures (e.g., up to ≈ 7 K for $B=6$ T). In zero field, C_M can be attributed to the same type of excitations for temperatures sufficiently below the freezing temperature in spin-glass samples and the Curie temperature in ferromagnetic samples. Necessarily, these excitations should also play a dominant role in the mechanisms leading to the experimentally observed field dependence of the thermal conductivity.[13]

ACKNOWLEDGMENT

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