Specific heat of $Eu_xSr_{1-x}S$ in high magnetic fields

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The specific heat C of the spin glass $Eu_{0.25}Sr_{0.75}S$ and of the reentrant ferromagnet $Eu_{0.54}Sr_{0.46}S$ has been measured between 70 mK and 2 K in magnetic fields B up to 6 T. C for $B=0$ varies roughly linearly with temperature as found previously, while the steep temperature dependence observed in high fields indicates the gradual opening of a gap in the energy spectrum with increasing B. The presence of a large threshold field B_0 for the existence of a gap in the case of the spin glass is attributed to a peak in the density of states in the vicinity of $E=0$, in agreement with recent numerical calculations.

I. INTRODUCTION

Recent interest in spin-glass research has focused on the $Eu_xSr_{1-x}S$ insulating spin glass.[1] With increasing Eu concentration x, magnetism of isolated Eu^{2+} clusters, spin-glass behavior, reentrant (or "frustrated") ferromagnetism, and simple (diluted) ferromagnetism are observed. The nature of the low-energy excitations in this disordered magnetic system is not entirely resolved. Previous specific-heat measurements revealed the excitation of clusters for $x \leq 0.07$.[2] Spin-wave-like excitations in the spin-glass and reentrant ferromagnetic phases were suggested in some theoretical papers[3,4] prompted by specific-heat studies in this concentration range.[5] At still higher concentrations ($x \geq 0.70$) the specific heat C was interpreted in terms of standard spin-wave theory.[6] Inelastic neutron scattering experiments gave some indications for spin-wave excitations not only for $x=0.70$, but also in the reentrant regime ($x=0.54$).[7] With the exception of the investigation by Meschede *et al.*,[5] which was carried out in magnetic fields up to 1 T, all the above-mentioned work was done in zero field.

In order to obtain more information about the magnetic excitations, we have begun measurements of C in high magnetic fields. In particular, we wanted to look for evidence for spin-wave-like excitations induced by the presence of a magnetic field. Heat transport associated with the propagation of such excitations has been proposed to explain the enhancement of the thermal conductivity with magnetic field observed in recent measurements performed above 1.5 K on $Eu_xSr_{1-x}S$.[8] We present here our first results for $x=0.25$ and 0.54, i.e., for samples in the spin-glass and reentrant ferromagnetic regions, respectively.

II. EXPERIMENTAL DETAILS

Two of the single-crystal samples were the same as used for the thermal-conductivity measurement.[8] In addition, the $x=0.54$ investigated by Meschede *et al.*[5] was also measured. The specific heat was measured with the heat-pulse method in a dilution refrigerator between 0.07 and 2 K. Magnetic fields up to 6 T were provided by a superconducting solenoid with a low-field region (<0.1 T for a central field of 6 T), where a calibrated [against cerium magnesium nitrate (CMN)] Ge resistor was located. Matsushita carbon thermometers located in the high-field region of the magnet were in turn calibrated against the Ge resistor. The magnetoresistance $|\Delta R/R|$ of the carbon resistors was $\sim 5\%$ at 0.1 K and 6 T and much smaller above 0.3 K. For the two $x=0.54$ samples, two different mountings were employed in order to check for problems arising from slow thermal relaxation. (1) In the "standard mounting" in our laboratory the sample was attached to a Si plate carrying an electrical thin-film heater and the carbon thermometer.[9] A weak thermal link between sample and mixing chamber (in the present case 80-μm Cu wire was used) provided for cooling of the sample with sample-bath relaxation times of the order of several minutes for $B=0$. In higher fields ($B \geq 3$T) the relaxation times became so short (due to the decrease in heat capacity) that the Cu link had to be removed and only the nylon threads holding the Si plate provided for the thermal link. Even so, the sample-bath relaxation times were only ~ 10 s in $B=3$ T. In these arrangements, considerable "overshooting" of the temperature upon application of a heat pulse was observed because the Si plate was overheated before thermalizing with the sample. The heat capacity was determined from $C_{tot}=Q/\Delta T$ and ΔT was obtained by extrapolating the long-time temperature decay due to the weak link *(after* the overheating had decayed) to the time when the heat pulse of energy Q was applied. (2) The sample was tied between nylon threads. Thermometer, heater (in this case a constantan strain gauge), and weak link were all directly attached to the sample. Here only a small overheating was observed at low temperatures, presumable because of the slow thermal relaxation between lattice and nuclear spins which contribute to the heat capacity at low temperatures (see below). Both types of measurements yielded heat capacities which agreed within 10%. Therefore, the $x=0.25$ sample was only measured with the "standard mounting."

III. RESULTS

The specific-heat results for $Eu_xSr_{1-x}S$ are displayed in Fig. 1 ($x=0.25$) and Fig. 2 ($x=0.54$) for various applied magnetic fields B between 0 and 6 T. On the whole, the temperature and magnetic field dependence are quite similar for the two concentrations. C varies roughly linearly with T in the intermediate-temperature range for $B=0$ and exhibits a strong decrease at all temperatures with increasing B. The lattice contribution to C amounts to $\sim 1\times 10^{-6}$ J/g K at 1 K and hence plays a negligible role at all temperatures and fields.

We first discuss the results for $x=0.25$ in detail. Between 0.1 and 1 K for $B=0$, C can be adequately represented by

$$C=aT+bT^{-2}, \tag{1}$$

with $a=1.0\times 10^{-2}$ J/g K^2 and $b=6\times 10^{-6}$ J K/g. The first contribution is the well-known spin-glass linear specific heat and the second is due to hyperfine splitting of the ^{151}Eu and ^{153}Eu nuclei. Above 1 K, C levels off towards a maximum presumably occurring near the upper end of our temperature range, ~ 1.5 K. This temperature is roughly twice the spin-glass freezing temperature $T_f \approx 0.8$ K. A similar behavior has been found previously for $x=0.40$.[5] With increasing field, the T dependence of C becomes steeper and also the low-temperature nuclear specific-heat contribution decreases.

For $x=0.54$ (Fig. 2), positive deviations of the spin-glass specific heat from a strictly linear behavior are observed. A better fit for $T<T_f$ can be obtained by introducing a small additional T^2 term:

$$C=aT+bT^{-2}+cT^2 . \tag{2}$$

Such a T^2 term has been used frequently to describe the small deviations from a linear dependence generally observed in spin glasses,[10,11] and was also found previously for $Eu_{0.54}Sr_{0.46}S$.[5] From a fit of Eq. (2) to our data, we obtain the coefficients $a=5.9\times 10^{-3}$ J/g K^2, $b=7.5\times 10^{-6}$ J K/g, and $c=1.3\times 10^{-3}$ J/g K^3 (cf. solid line in Fig. 2).

Alternatively, C can be represented as a power law $C\sim T^{1.05}$. Such a power-law behavior has been suggested for several spin glasses with exponents ranging from 1.2 to 1.7.[12,13] An almost equally good fit to the data is obtained using

$$C=a'T^{1.05}+bT^{-2}, \tag{2'}$$

with $a'=6.8\times 10^{-3}$ J/g K$^{2.05}$ and again $b=7.5\times 10^{-6}$ J K/g. For lack of a detailed theory treating the "linear" specific heat of spin glasses, no further discussion of the small deviations from linearity as modeled by either Eq. (2) or (2′) is possible. At this point we should mention that our data for $x=0.54$ are smaller than those reported previously by Meschede *et al.*[5] by $\approx 30\%$ (see Fig. 2). For our two samples with $x=0.54$—one of which was the very sample measured by Meschede *et al.*[5]—measured in two different mountings as described above, the data agreed to within 10%. We are at present unable to locate the origin of the discrepancy with the earlier results.

Concerning the magnetic field dependence of C which has been investigated in more detail for $x=0.54$, a very gradual decrease of C is observed in small fields, followed by a much faster decrease above 3 T.

IV. DISCUSSION

We first discuss the spin-glass (magnetic) specific heat C_M and then the nuclear specific heat. As mentioned above, a roughly linear temperature dependence of C_M is only observed for $B=0$. For moderate fields B the magnetic specific heat can be represented by $C_M\sim T^{\alpha}$ over an appreciable T range. α is plotted as a function of B in Fig. 3. The smooth dependence of $\alpha(B)$ shows that the previously observed[5] $T^{3/2}$ law for $x=0.54$ and $B=1$ T is

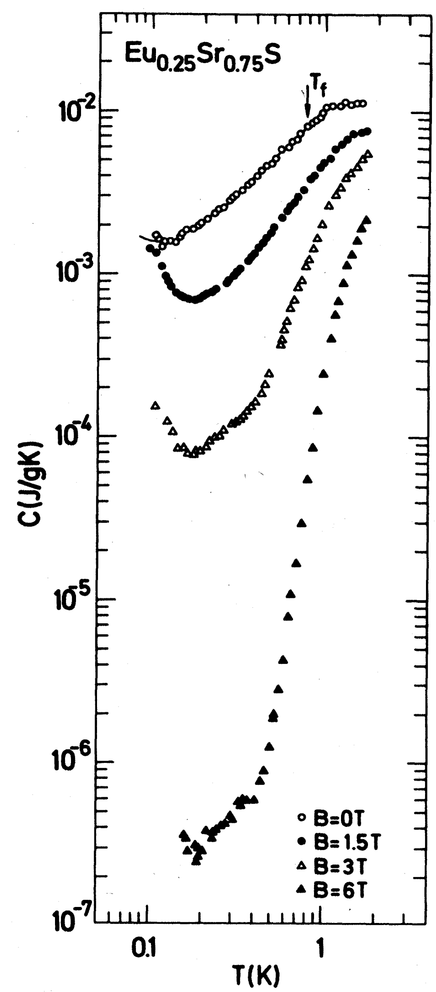

FIG. 1. Specific heat C of $Eu_{0.25}Sr_{0.75}S$ vs temperature T in various applied magnetic fields B. Solid line indicates fit of Eq. (1) to the data for $B=0$. The arrow denotes the spin-glass freezing temperature T_f.

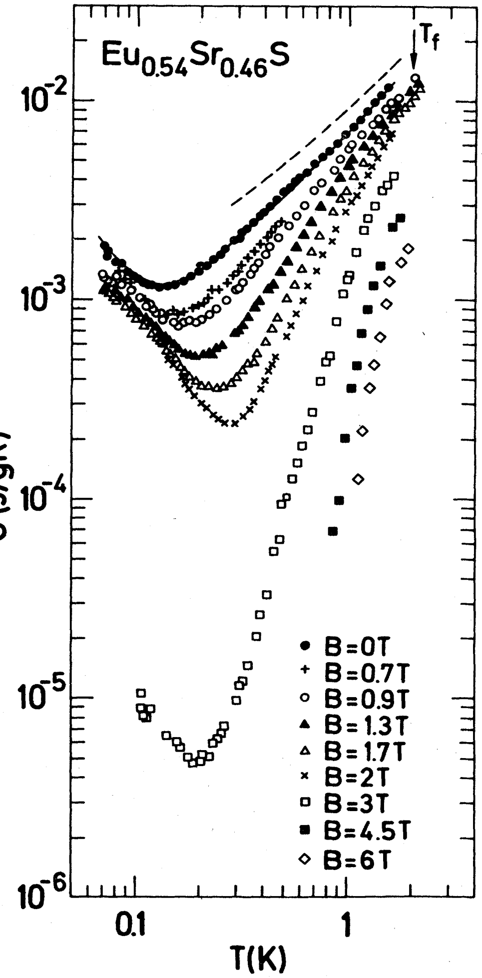

FIG. 2. Specific heat C of $Eu_{0.54}Sr_{0.46}S$ vs. temperature T in various applied magnetic fields B. Data for $B=0$, 0.7, and 2 T were taken with sample mounting type 2, all others with type 1 (see text). Solid line indicates fit of Eq. (2) to the data for $B=0$. Dashed line represents results of Ref. 5. The arrow marked T_f denotes the ferromagnetic$\leftrightarrow$spin-glass transition.

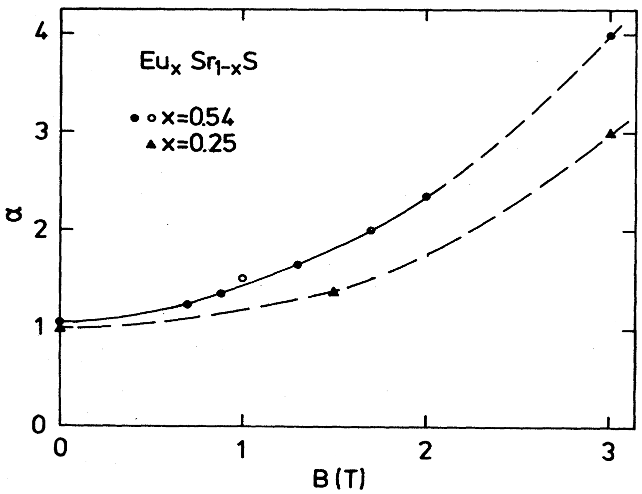

FIG. 3. Exponent α of the magnetic specific heat $C_M \sim T^\alpha$ vs magnetic field B. Open circle denotes datum given by Meschede *et al.* (Ref. 5).

accidental. There is no extended magnetic field regime where such a law, reminiscent of the spin-wave specific heat of a ferromagnet in *zero* field, is found. A simple calculation of the specific heat in the "saturated paramagnetic regime" is not available. We should mention that there appears to be no critical behavior at the spin-glass $\leftrightarrow$ paramagnetic transition for $x=0.25$ in the (B,T) phase diagram.

The faster than linear decay of C_M for $T\to 0$ hints at the gradual opening of a gap at $E=0$ in the spectrum of magnetic excitations as the magnetic field is increased. To our knowledge such a distinct feature has not been observed up to now in either metallic or insulating spin glasses.

For high fields ($B \geq 3$ T) C_M decreases almost exponentially. This can be seen from Fig. 4 where $\ln C$ is plotted versus $1/T$ for $B=3$ T as an example. The slope of the resulting straight lines yields an energy gap ΔE, hence

$$C_M = A \exp(-\Delta E/k_B T) . \tag{3}$$

Figure 5 shows the resulting gaps ΔE versus B. Both sets of points, for $x=0.54$ and for $x=0.25$, can be fitted with straight lines having the same slope: $\Delta E = g\mu_B(B-B_0)$. The magnitude of the slope is that expected for the upward energy shift of spin waves in a magnetic field, in analogy to the case of a simple isotropic ferromagnet.

The presence of B_0 reflects the existence of a threshold field for the opening of the gap, which can be understood qualitatively in the following way: The magnon dispersion curve of a simple ferromagnet is replaced by a broader spectrum of excitations in a spin glass.[4] For $B=0$, the density of states at low energies is approximately constant, and leads to the roughly linear temperature dependence of C observed below T_f. In a moderate magnetic field, the whole excitation band is shifted by an amount $g\mu_B B$, but due to the finite density of states in the vicinity of $E=0$, a gap does not open immediately, and the specific-heat behavior remains accordingly relatively flat. It is only in larger applied fields that the density of states at $E=0$ vanishes, and the system can be described in terms of an effective gap $g\mu_B(B-B_0)$ so that B_0 appears as a measure of the spectrum width at low energies.

For $x=0.54$, B_0 is small (a rms best fit yields $B_0=0.2$ T, but given the measurement's accuracy B_0 could possibly be zero), as could be expected from the composition falling in the frustrated ferromagnetic regime. On the other hand, for $x=0.25$, well in the spin-glass regime, $B_0=1.4$ T corresponds to an energy of $E_0=1.9$ K k_B i.e., a considerable width for the spin-glass excitation spectrum. This difference can also be inferred from a direct comparison of the C-versus-T slopes for moderate fields ($B \approx 1.5$ T) which are larger for $x=0.54$ (cf. Fig. 3). These findings are in qualitative agreement with the predictions of numerical calculations for $Eu_xSr_{1-x}S$.[3,4]

We now turn to the discussion of the nuclear specific

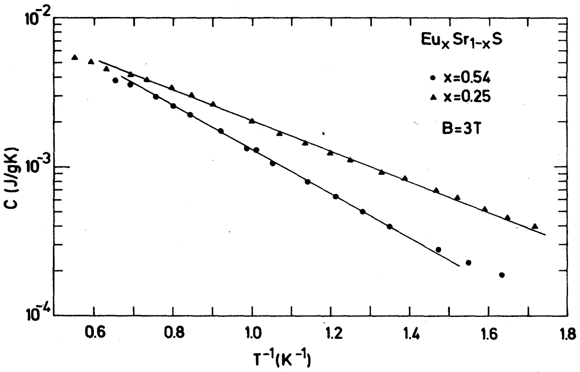

FIG. 4. Specific heat C of $Eu_xSr_{1-x}S$ for $B=3$ T plotted as $\ln C$ vs $1/T$.

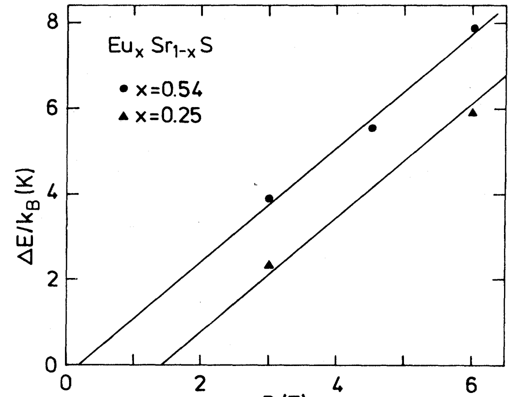

FIG. 5. Energy gap ΔE vs magnetic field B for $B \geq 3$ T. Solid lines represent $\Delta E = g\mu_B(B - B_0)$.

heat C_N arising from the hyperfine splitting of the ^{151}Eu and ^{153}Eu nuclei. The T^{-2} term is the high-temperature tail of a Schottky specific-heat anomaly. We can calculate the expected coefficient b_N of the T^{-2} term:

$$b_N = \frac{xR}{M} \sum_i a_i \frac{I_i + 1}{3I_i} \left[\frac{\mu_i B_{\rm eff}}{k_B} \right]^2 . \quad (4)$$

Here R is the gas constant, M is the molar mass of $Eu_xSr_{1-x}S$, I_i is the nuclear spin, μ_i the nuclear moment, and a_i the relative abundance of the isotope i. For $x = 0.25$, $b_N = 5.5 \times 10^{-6}$ J K/g and for $x = 0.54$, $b_N = 11.3 \times 10^{-6}$ J K/g are calculated from Eq. (4) using the values of $B_{\rm eff}$ obtained from Mössbauer measurements.[14] While for $x = 0.25$ the agreement with the experimentally determined value b (see above) is quite good, only poor agreement is found for $x = 0.54$, b being only $\approx 70\%$ of b_N. The previously reported values of b *exceeding* considerably b_N for $x = 0.40$ and 0.54 were inferred from measurements above 0.3 K only.[5] Hence we feel that the present values are more reliable and there is no need to invoke an excess T^{-2} specific heat. In this respect it is interesting to note that an excess value of b was found[2] in very dilute $Eu_xSr_{1-x}S$ ($x \leq 0.07$) which was interpreted as arising from isolated spins (with respect to nearest and next-nearest Eu^{2+} neighbors). However, as the number of isolated spins quickly drops to zero for x exceeding the percolation threshold $x_p \approx 0.13$, such a contribution should indeed be negligible in our samples.

The apparent nuclear heat capacity $C_N = bT^{-2}$ strongly decreases in large magnetic fields (cf. Figs. 1 and 2). This can be explained with an increase of the nuclear spin-lattice relaxation time T_1 which of course is accompanied by an increase of the thermal relaxation time τ_1 between nuclear spins and lattice. With our quasiadiabatic method, C_N can only be detected if $\tau_1 < \tau$, where τ is the sample-bath thermal relaxation time. Indeed, T_1 has been reported to increase strongly with B at low temperatures in EuS, i.e., approximately $T_1 \sim B^3/T^2$, reaching several hours at 0.05 K in a field of 1.7 T.[15] A magnetic-field-dependent coupling of the nuclear spins to the lattice in $Eu_xSr_{1-x}S$ could occur via magnonlike excitations which are frozen out in high magnetic fields at low temperatures as discussed above.

V. CONCLUSIONS

The present specific-heat measurements show a drastic quenching of the low-temperature magnetic excitations of $Eu_xSr_{1-x}S$ in high fields. While the low-temperature specific heat of $Eu_{0.54}Sr_{0.46}S$ behaves similarly to that of a simple ferromagnet in a field, the reduction of C observed in the spin-glass $Eu_{0.25}Sr_{0.75}S$ is markedly smaller. This suggests the presence of a threshold field for the opening of a gap in the excitation spectrum. Although specific-heat data cannot provide information on the propagating nature of the observed excitations, these results support the picture that in magnetically disordered $Eu_xSr_{1-x}S$ the magnetic field establishes spin-wave-like excitations that can contribute to heat transport, as previously suggested to explain the thermal conductivity (κ) behavior:[8] It shows that down to 1 K and even in 6 T there is sufficient heat capacity available in the magnonlike system to sustain the observed heat transport, with values for the energy gap consistent with the approach toward saturation measured for κ, and also consistent with a larger magnon heat transport in the spin glass than in the frustrated ferromagnet.

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