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Linear and non-linear AC susceptibilities of the spin glass $Eu_{0.4}Sr_{0.6}S$

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Abstract. The linear and non-linear susceptibilities of a spin-glass $Eu_{0.4}Sr_{0.6}S$ system have been measured in the temperature range from 1.2 to 4.2 K, for frequencies between 1 and 234 Hz. The freezing temperature $T_f(\omega \to 0)$ of the system is found to be 1.60 ± 0.02 K, as determined from the Cole–Cole analysis using the linear-susceptibility data. In the vicinity of T_f the non-linear susceptibility varies much more strongly with temperature than the linear susceptibility does. The third-harmonic results show the beginning of a power-law divergence $\chi_3 \propto \epsilon^{-\gamma}$, where $\epsilon = (T - T_f)/T_f$, and the curve quickly becomes rounded. This behaviour suggests a cooperative phenomenon that is greatly modified by strong dynamical and non-equilibrium effects as T_f is approached. The critical exponent γ is estimated to be 2.35 ± 0.20 in the limited temperature region where a fit was obtained. In addition it is experimentally found that even a small DC field has a large effect on the third harmonic.

1. Introduction

In recent years, solid solutions of EuS and SrS have been extensively studied. It is now well known that EuS has an FCC structure similar to NaCl and is a Heisenberg ferromagnet with a Curie temperature $T_C = 16.6$ K [1]. Each magnetic Eu^{2+} ion has 12 nearest and six next-nearest neighbour Eu^{2+} ions. The exchange interactions between the nearest and next-nearest neighbours have been determined by means of neutron-scattering experiments and found to be $J_1/k_B = 0.221 \pm 0.003$ K and $J_2/k_B = -0.100 \pm 0.004$ K, respectively [2]. Hence the nearest-neighbour interaction is ferromagnetic and the next-nearest-neighbour interaction is antiferromagnetic. When EuS is diluted with the isostructural diamagnet SrS to form $Eu_xSr_{1-x}S$, the Curie temperature is rapidly lowered with decreasing x and, near $x = 0.55$, the system becomes a spin glass, remaining so for concentrations down to 0.13 [1–3]. With increasing concentration of Sr, randomness is introduced into the coupling alignments of the Eu moments. Because of the competing ferromagnetic and antiferromagnetic interactions, frustration results and this combination of randomness and frustration creates a good 3D Heisenberg spin glass. If x is reduced below 0.13, then the Eu ions will be too far apart to form a percolating network of first- and second-near-neighbour coupled moments. Accordingly the system splits into domains and gradually freezes into a superparamagnetic state [4].

A distinctive feature of a spin glass is the cusp in the low-field AC susceptibility at the 'freezing temperature' T_f which suggests a thermodynamic phase transition. Whereas the round maximum above T_f in the specific heat, the reversible (field-cooled) and irreversible (zero-field-cooled) magnetization with characteristic long-time relaxation effects below T_f all suggest a kind of blocking phenomenon, leading to non-equilibrium metastable states. At present the nature of the ordering process in spin glasses is not well understood experimentally nor theoretically.

Recently major efforts in the field of spin glasses have been directed towards clarifying the freezing phenomenon, both theoretically and experimentally. On the assumption that the spin-glass freezing is a cooperative phenomenon and within the framework of the mean-field theory, the non-linear susceptibility has been predicted to diverge at T_f according to a power law of the form $\chi_{2n+1} \propto \epsilon^{-n(\gamma+\beta)+\beta}$, for $n \geqslant 1$, where $\epsilon = (T - T_f)/T_f$, and γ and β are the susceptibility and order-parameter critical exponents [5, 6]. This divergent behaviour indicates a phase transition. An experimental investigation of the temperature dependence of χ_{2n+1} thus allows one to seek such divergences and, if so, to determine T_f as well as γ and β. By measuring the third harmonic, T_f and γ can be determined but, to find β, one has to measure fifth or higher harmonics.

In this work we wish to study the nature of the transition at T_f by measuring the linear and non-linear susceptibilities of the spin glass $Eu_{0.4}Sr_{0.6}S$ as a function of temperature for frequencies between 1 and 234 Hz. We present the experimental procedure in section 2, and the results and discussion in section 3; finally section 4 gives our conclusions.

2. Experimental procedure

It is well known that, owing to the thermal fluctuations and the increasing short-range order around T_f, pronounced non-linearities appear in the magnetization of a spin glass. In terms of the applied field H, the magnetization can be expressed as

$$M(H,T) = \chi_1 H + \chi_3 H^3 + \chi_5 H^5 + \cdots \tag{1}$$

where even powers of H are neglected because of the symmetry properties of M. In general, let us take H to be

$$H = H_0 + h(t) \tag{2}$$

where H_0 is a static applied field and $h(t) = h_0 \sin(\omega t)$ is the time-dependent AC driving field. If equation (2) is inserted in equation (1), one obtains

$$\begin{aligned} M(H,T) = {} & [(\chi_1 + \chi_3 H_0^2 + \cdots) + (\tfrac{3}{2}\chi_3 + 5\chi_5 H_0^2 + \cdots)h_0^2 + \cdots] \\ & \times H_0 + [(\chi_1 + 3\chi_3 H_0^2 + 5\chi_5 H_0^4 + \cdots) + (\tfrac{3}{4}\chi_3 + \tfrac{15}{2}\chi_5 H_0^2 + \cdots)h_0^2 + \cdots] \\ & \times h_0 \sin(\omega t) - [(\tfrac{3}{2}\chi_3 + 5\chi_5 H_0^2 + \cdots) + (\tfrac{5}{2}\chi_5 + \cdots)h_0^2 + \cdots] \\ & \times H_0 h_0^2 \cos(2\omega t) - [(\tfrac{1}{4}\chi_3 + \tfrac{5}{2}\chi_5 H_0^2 + \cdots) + (\tfrac{5}{16}\chi_5 + \cdots)h_0^2 + \cdots] \\ & \times h_0^3 \sin(3\omega t) + \cdots . \end{aligned} \tag{3}$$

As can be seen from equation (3), in the absence of the DC field H_0, the first term and the terms involving even harmonics disappear. In this case equation (3) reduces to

$$M(h,T) = (\chi_1 + \tfrac{3}{4}\chi_3 h_0^2 + \tfrac{5}{8}\chi_5 h_0^4 + \cdots)h_0 \sin(\omega t) - (\tfrac{1}{4}\chi_3 + \tfrac{5}{16}\chi_5 h_0^2 + \cdots) \times h_0^3 \sin(3\omega t) + (\tfrac{1}{16}\chi_5 + \cdots)h_0^5 \sin(5\omega t) + \cdots \tag{4}$$

or, in compact form,

$$M(h,T) = \sum_{n=0}^{\infty} \tilde{\chi}_{2n+1} h_0^{2n+1} \sin[(2n+1)\omega t] \tag{5}$$

where

$$\begin{aligned} \tilde{\chi}_1 &= \chi_1 + \tfrac{3}{4}\chi_3 h_0^2 + \tfrac{5}{8}\chi_5 h_0^4 + \cdots \\ -\tilde{\chi}_3 &= \tfrac{1}{4}\chi_3 + \tfrac{5}{16}\chi_5 h_0^2 + \cdots \\ \tilde{\chi}_5 &= \tfrac{1}{16}\chi_5 + \cdots \\ &\;\vdots \end{aligned} \tag{6}$$

By detecting the response of $M(t)$ to the AC magnetic field at the frequencies ω, 3ω, $5\omega, \cdots$, one can obtain $\tilde{\chi}_1$, $\tilde{\chi}_3$, $\tilde{\chi}_5, \cdots$, respectively. These are the measured susceptibilities given in section 3. However, in general, one has to correct these results for demagnetization effects.

In terms of the driving field h, and the demagnetization factor D (known from sample geometry), the internal field h_i can be written as

$$h_i = h - Dm \tag{7}$$

where $m \equiv M/M_0$, M_0 being the saturation value of M. This means that the measured value of the susceptibility is

$$\mathrm{d}m/\mathrm{d}h = \tilde{\chi}_1 = (\mathrm{d}m/\mathrm{d}h_i)(\mathrm{d}h_i/\mathrm{d}h) = (\mathrm{d}m/\mathrm{d}h_i)(1 - D\,\mathrm{d}m/\mathrm{d}h) = \tilde{\chi}_{1c}(1 - D\tilde{\chi}_1)$$

or the corrected linear susceptibility becomes

$$\tilde{\chi}_{1c} = \tilde{\chi}_1/(1 - D\tilde{\chi}_1). \tag{8}$$

For the measured second harmonic we have

$$\begin{aligned} \mathrm{d}^2m/\mathrm{d}h^2 &= (\mathrm{d}^2m/\mathrm{d}h_i^2)(\mathrm{d}h_i/\mathrm{d}h)^2 + (\mathrm{d}m/\mathrm{d}h_i)(\mathrm{d}^2h_i/\mathrm{d}h^2) \\ &= (\mathrm{d}^2m/\mathrm{d}h_i^2)(1 - D\,\mathrm{d}m/\mathrm{d}h)^3. \end{aligned}$$

Thus the corrected second harmonic becomes

$$\tilde{\chi}_{2c} = \tilde{\chi}_2/(1 - D\tilde{\chi}_1)^3. \tag{9}$$

By following the same procedure, one finds the corrected value of the third-harmonic component of the AC susceptibility to be

$$\tilde{\chi}_{3c} = \tilde{\chi}_3/(1 - D\tilde{\chi}_1)^4. \tag{10}$$

As can be seen by comparing equations (8)–(10) the demagnetization correction becomes more important the higher the harmonic of the non-linear susceptibility.

The sample used in this work was previously investigated and described by Baalbergen *et al* [3]. The sample is spherical for which $D = 18.81$ g emu^{-1}. For AC susceptibility measurements the well known mutual inductance technique has been used. The output from the mutual inductance bridge is fed into two lock-in amplifiers. One is operated at ω, the frequency of the applied field, and the other at $n\omega$. With this method the in-phase and out-of-phase components are simultaneously measured [7]. It is natural that the magnitudes of the higher harmonics are much smaller than the linear response. Thus, in order to detect them, one usually needs higher values of the driving AC field. For the sample used in this work an AC field amplitude of 0.7 Oe was sufficient for the linear response. However, it was difficult to obtain clear signals for the higher harmonics with the same field. Thus we had to increase the amplitude which may cause a field-dependent susceptibility. For this reason, one has to compromise between having a clear signal and a field-dependent susceptibility. To clarify this point we have investigated the driving field dependence of the third harmonic $\tilde{\chi}_3$. For third-harmonic frequencies higher than $3f = 9$ Hz, $\tilde{\chi}_3$ has been found to be amplitude independent up to a value of 7 Oe. Therefore, in our non-linear susceptibility measurements, an AC driving field of 7 Oe was used.

3. Results and discussion

In this section, we present our result for the temperature dependence of the linear and non-linear AC susceptibilities of the spin glass $Eu_{0.4}Sr_{0.6}S$ measured at different frequencies. In addition the DC field dependence of the third-harmonic response around T_f is also given. The temperature range used is from 1.2 to 4.2 K, and the AC field amplitudes are 0.7 Oe and 7 Oe for the linear and the non-linear responses, respectively. Note that all the results reported below are normalized to 1 Oe.

The temperature dependence of the measured in-phase component $\tilde{\chi}'_1$ of the linear AC susceptibility is shown in figure 1, for different frequencies. There is little or no frequency dependence at temperatures above the so-called spin-glass freezing temperature T_f characterized by the maximum in $\tilde{\chi}'_1$. However, around and below T_f the magnitude of $\tilde{\chi}'_1$ increases and its maximum shifts to lower temperatures with decreasing frequency in agreement with the results on many other spin-glass systems [3, 7–9]. We should point out here that the data shown in figure 1 for the zero-frequency value have been obtained from figure 2, where the Cole–Cole plots of $\mathrm{Im}(1/\tilde{\chi}_1)$ versus $\mathrm{Re}(1/\tilde{\chi}_1)$ are shown for temperatures between 1.2 and 1.65 K, with frequency as the parameter [3]. In figure 2 the extrapolated intersections of the lines with the $\mathrm{Re}(1/\tilde{\chi}_1)$ axis give the zero-frequency values of $\tilde{\chi}_1$, i.e. $\tilde{\chi}_1(0)$. Here we should also point out that the magnitude of the out-of-phase component $\tilde{\chi}''_1 = \mathrm{Im}(\tilde{\chi}_1)$ is two orders of magnitude smaller than $\tilde{\chi}'_1$ and, in contrast with the behaviour of $\tilde{\chi}'_1$, $\tilde{\chi}''_1(T)$ decreases with decreasing frequency. Furthermore, above 1.6 K, $\tilde{\chi}_1''$ was not detectable except for the two higher frequencies [10]. For temperatures above 2 K it was zero for all frequencies used in this work. Therefore in figure 1 the point for $\tilde{\chi}_1(0)$ is not given for $T > 2$ K. The temperature corresponding to the maximum of $\tilde{\chi}_1(0)$ in figure 1 gives the freezing temperature $T_f(\omega = 0)$ as 1.60 ± 0.02 K which is in good agreement with the $T_f(\omega)$ trends.

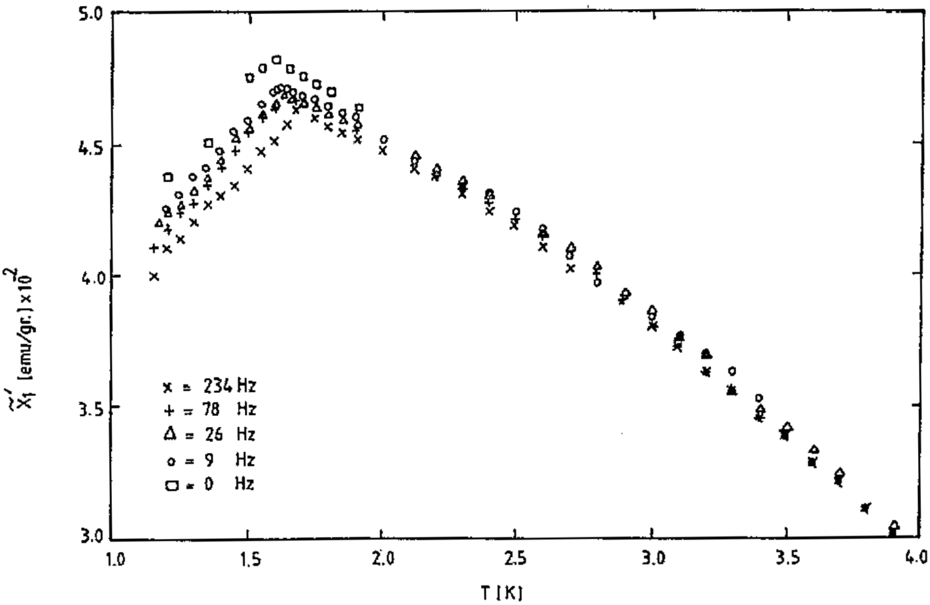

Figure 1. Temperature dependence of the measured in-phase component of $\tilde{\chi}_1$, for different frequencies.

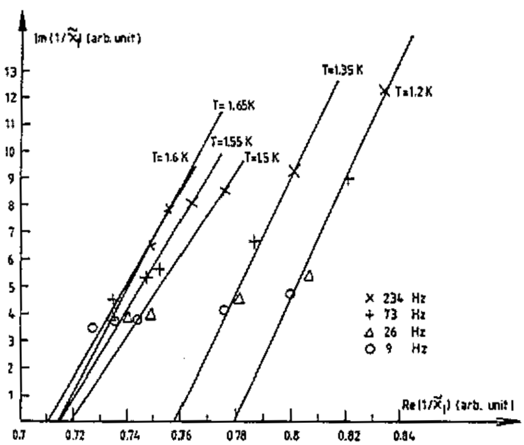

Figure 2. $\mathrm{Im}(1/\tilde{\chi}_1)$ against $\mathrm{Re}(1/\tilde{\chi}_1)$, at several temperatures.

By using the measured values of the linear susceptibility given in figure 1 and the demagnetization factor of the sample ($D = 18.81$ g emu^{-1} for our spherical sample) in equation (8), the corrected susceptibilities $\tilde{\chi}_{1c}$ are obtained. These results are shown in figure 3 for different frequencies. This figure illustrates the frequency dependence of the linear susceptibility much better than figure 1 (compare the vertical scales) and shows the importance of the demagnetization correction.

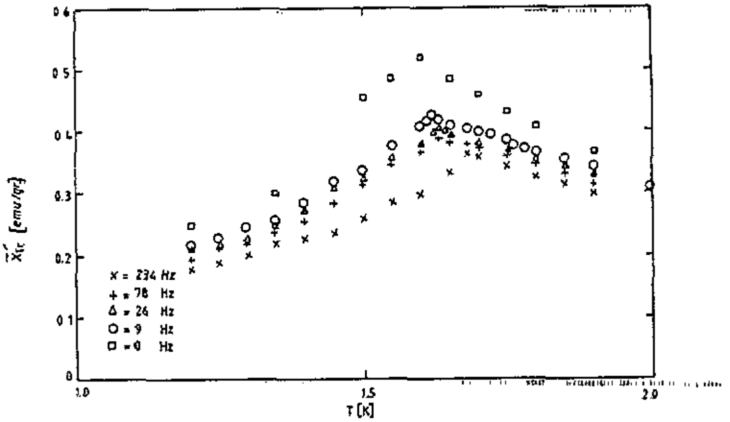

Figure 3. Temperature dependence of the corrected in-phase component of $\tilde{\chi}_1$, for different frequencies.

As was mentioned in section 2, the coefficient of the third harmonic $\tilde{\chi}_3$ is field independent above $3f = 9$ Hz but is dependent below it. Therefore, we separate the results for $\tilde{\chi}_3$ into two regimes: regime I is from $3f = 9$ Hz up to 234 Hz; regime II is from $3f = 9$ Hz down to 3 Hz. Figures 4 and 5 give the temperature dependences of the in-phase component $\tilde{\chi}'_3$ and the out-of-phase component $\tilde{\chi}''_3$ of the third harmonic of the non-linear susceptibility for frequencies from $3f = 9$ Hz to 234 Hz†. It is obvious that the magnitudes of both components increase at around T_f, and the transitions to the spin-glass state shift to lower temperatures with decreasing frequency. From the real and imaginary components, the absolute value of $\tilde{\chi}_3$ given by $\tilde{\chi}_3 = \sqrt{(\tilde{\chi}'_3)^2 + (\tilde{\chi}''_3)^2}$ is calculated and shown in figure 6. The general behaviour of $\tilde{\chi}_3$ is almost the same as that of $\tilde{\chi}'_3$. Since the equilibrium value of T_f is obtained as the frequency goes to zero, we can see from figure 6 that the maximum in $\tilde{\chi}_3$ approaches 1.6 K, which is in very good agreement with the value estimated from $\tilde{\chi}_1(0)$ in figure 1.

The results for regime II are presented in figure 7, where we give only the absolute value of $\tilde{\chi}_3$ for frequencies in the range from 3 to 9 Hz. By comparing figures 6 and 7, one immediately sees the difference between the behaviours of the two regimes, namely, while the $\tilde{\chi}_3$ results above 9 Hz increase with decreasing frequency, those below decrease with decreasing frequency. The behaviour above 9 Hz is as expected; however, that below is, at first glance, surprising. Further consideration shows that it is not. Such behaviour has to do with the strong AC field amplitude dependence of the third harmonic at the lower frequencies. This point has been previously examined by Chikazawa *et al* [11] and by us [7]. As pointed out above for our $\tilde{\chi}_3$ measurements, the amplitude of the driving AC field has been kept constant at

† From now on, for the frequency values given in the text and figures, $3f$ indicates the third-harmonic and $2f$ the second-harmonic values.

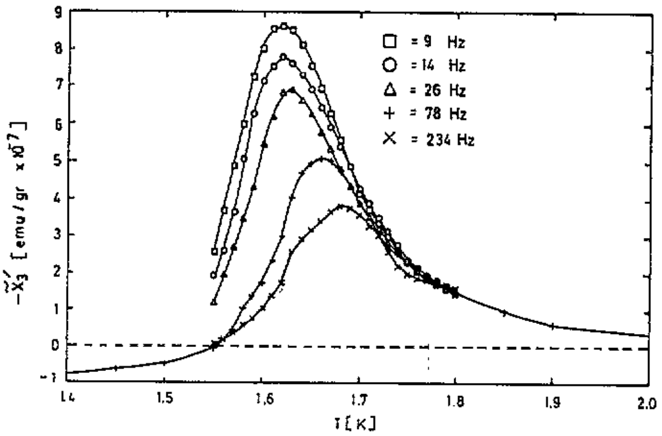

Figure 4. Temperature dependence of the measured in-phase component of $\tilde{\chi}_3$, for different frequencies.

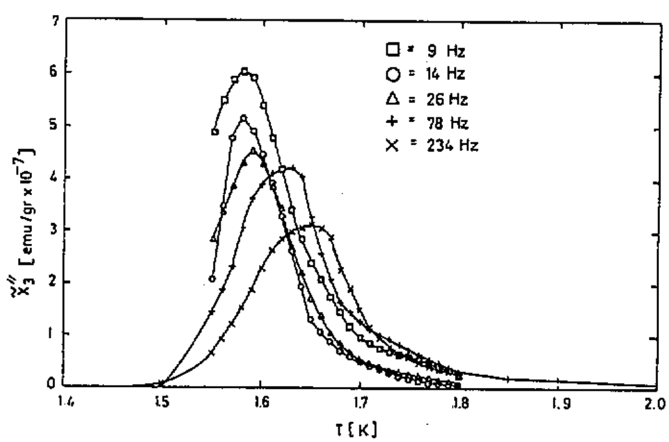

Figure 5. Temperature dependence of the measured out-of-phase component of $\tilde{\chi}_3$, for different frequencies.

7 Oe for all frequencies. If we could have used lower amplitude values for lower frequencies, we would have observed the same behaviour as that above 9 Hz, but signal-to-noise difficulties for the non-linear measurements do not allow us to use lower-amplitude values at lower frequencies.

To describe the frequency dependence of the freezing temperature obtained from the third-harmonic measurements, we plot T_f^{-1} versus $\log 3f$ in figure 8. The results indicate that the system does not obey the Arrhenius law [12] given by the expression $T_f^{-1} \propto \ln(f_0/f)$. If it did, a straight-line plot would be found. Thus the spin-glass transition cannot be explained in terms of the blocking of independent magnetic units such as superparamagnetic clusters. The bending of $T_f(\omega)$ to a probably constant T_f-value at low frequencies may be an indication of an incipient phase transition.

Using equation (10) the demagnetization corrections have been performed on the measured third-harmonic results shown in figures 6 and 7. However, because of

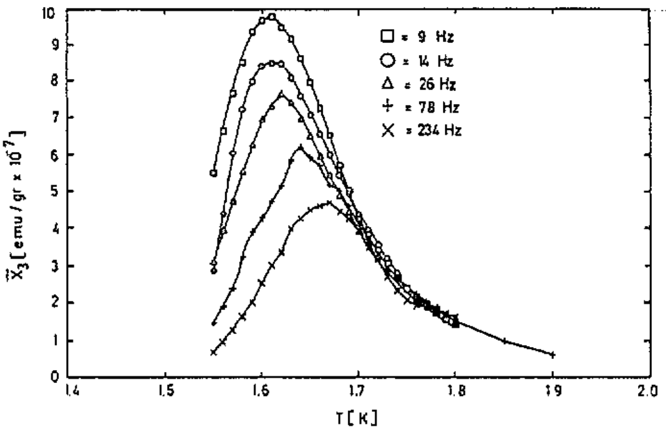

Figure 6. Temperature dependence of the absolute value of $\bar{\chi}_3$, for frequencies between $3f = 9$ Hz and $3f = 234$ Hz.

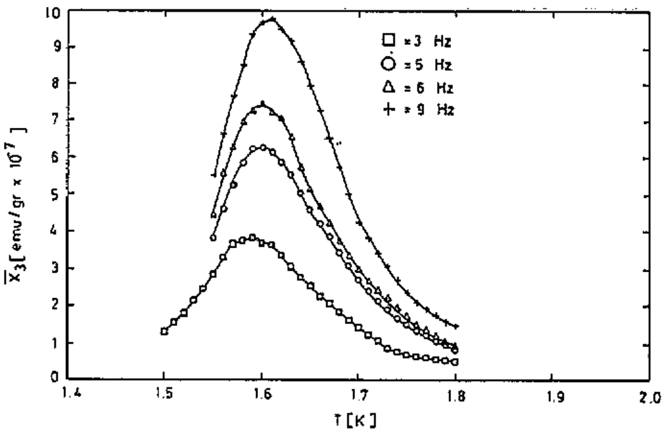

Figure 7. Temperature dependence of the absolute value of $\bar{\chi}_3$, for frequencies between $3f = 3$ Hz and $3f = 9$ Hz.

the unusual low-frequency behaviour, we give in figure 9 only the corrected values for regime I, i.e. the values for frequencies above 9 Hz. The corrected values are almost four orders of magnitude larger than the uncorrected values. Furthermore, the transitions to the spin-glass phase become sharper after the demagnetization correction.

As discussed in section 1, based on the model of Edwards and Anderson [13], Suzuki [5] has proposed a phenomenological theory for spin glasses. He showed that, if the spin-glass freezing is a cooperative phenomenon, the non-linear susceptibilities χ_{2n+1} diverge at T_f according to $\epsilon^{-n(\gamma+\beta)+\beta}$, for $n \geqslant 1$, where ϵ is the reduced temperature equal to $[(T - T_f)/T_f]$. Therefore, for the third harmonic, this power law becomes $\epsilon^{-\gamma}$ for $T > T_f$ and its sign is negative. In figure 10, log $\bar{\chi}_3$ is plotted against log ϵ for frequencies higher than 9 Hz. The value of the critical exponent γ, obtained from the initial slope of the straight line shown in the figure, is found to be

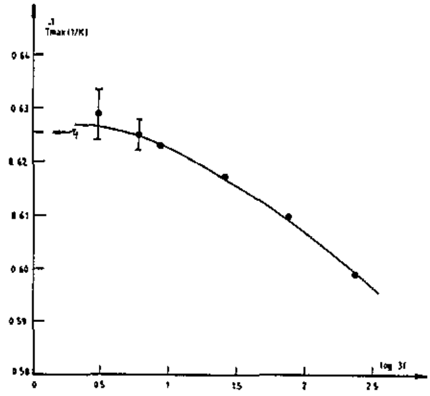

Figure 8. Inverse of the freezing temperature T_f as a function of frequency.

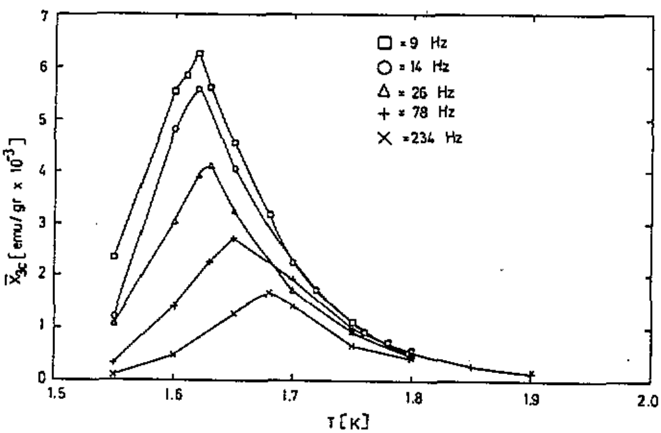

Figure 9. Temperature dependence of the corrected absolute value of $\tilde{\chi}_3$, for frequencies between $3f = 9$ Hz and $3f = 234$ Hz.

2.35 ± 0.20. Note that the power-law behaviour is only in an asymptotic region at rather large values of reduced temperature. This value is in good agreement with the value of 2.3 ± 0.2 reported for AgMn [9]. The rounding of the curves on approaching T_f is due to the magnetic inhomogeneities in the sample and most importantly to the dynamical effects as the system becomes out of equilibrium.

If one applies a DC field in addition to the driving AC field, one can also measure the second-harmonic response of the system (see equation (3)). By applying a DC field of 30 Oe, such second-harmonic measurements have also been carried out on $Eu_{0.4}Sr_{0.6}S$. The results, obtained after the demagnetization correction using equation (9), are shown in figure 11. The general behaviour is the same as that of the third harmonic (see figures 9 and 11). Since no theoretical guidance is available concerning the second-harmonic susceptibility, the results are not analysed further.

Finally, at a frequency of 234 Hz the DC field dependence of $\tilde{\chi}_3$ has also been investigated at $T = 1.68$ K, for two different values of AC field amplitude, namely 7 and 2 Oe. As can be seen from figure 12 the magnitude of $\tilde{\chi}_3$ decreases with

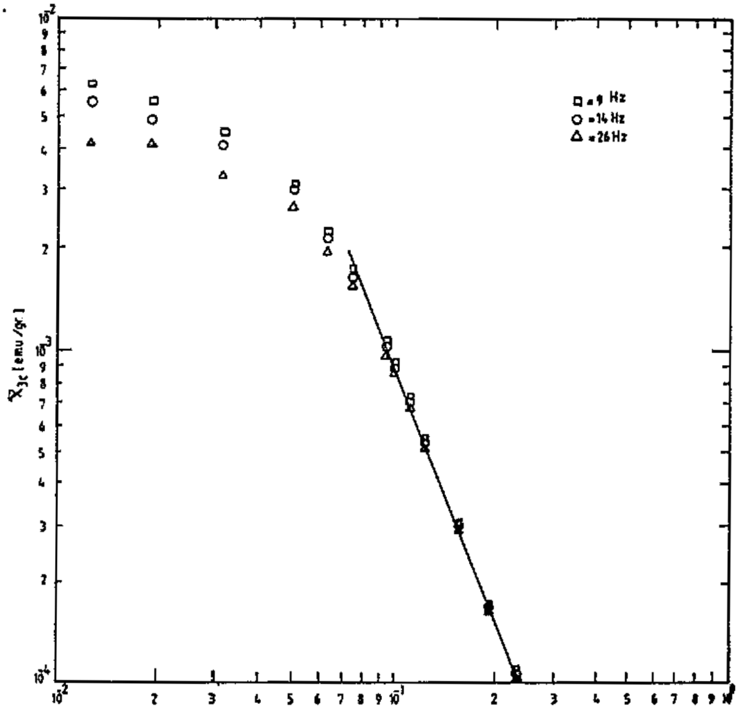

Figure 10. Logarithm of the corrected absolute value of $\tilde{\chi}_3$ against log ϵ.

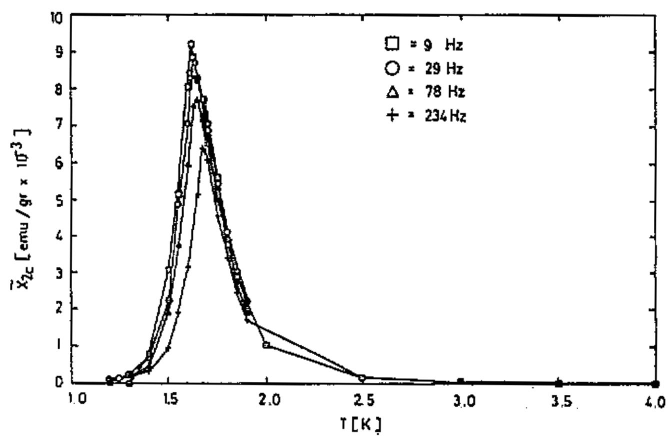

Figure 11. Temperature dependence of the corrected absolute value of $\tilde{\chi}_2$, for different frequencies in an applied field of 30 Oe.

increasing DC field, reaches zero at around 15 Oe and then changes sign. The crossover depends on the value of the AC field amplitude. From this observation, one can conclude that the non-linear susceptibility is extremely DC field dependent and small DC fields may suppress the third harmonic completely.

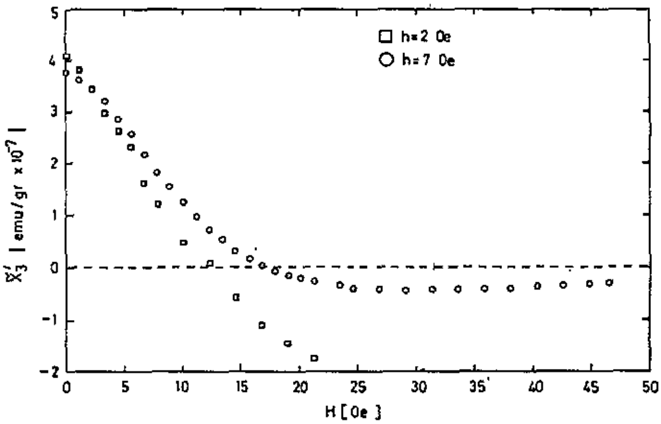

Figure 12. DC field dependence of the measured in-phase component of $\tilde{\chi}_3$ at $T = 1.68$ K for $f = 234$ Hz, and two different AC field amplitudes.

4. Conclusions

Non-linear AC susceptibility measurements on the spin glass $Eu_{0.4}Sr_{0.6}S$, especially the third-harmonic behaviour, indicate that the system has an incipient phase transition at $T_f(\omega = 0)$ rather than an independent cluster blocking phenomenon. We have reached this conclusion by examining

(i) the extrapolated value of $\tilde{\chi}_1(0)$ from the Cole–Cole analysis giving $T_f(\omega = 0) = 1.60$ K,

(ii) the frequency dependence of the freezing temperature, obtained from the third harmonic, which turned out not to obey the Arrhenius law, and

(iii) the initial power-law dependence of the third harmonic.

The third harmonic seems to obey the power law $\epsilon^{-\gamma}$ for $T > T_f$ and diverges on approaching T_f from above, as proposed by Suzuki [5]. However, here one must take into consideration the ferromagnetic clusters in the sample as well as the dynamical effects around T_f. Hence we believe that the non-linear susceptibility measurements are an important tool with which to investigate the nature of the transition for spin glasses, and probably for other magnetic systems. Nevertheless non-linear susceptibilities are extremely DC field dependent and even very small DC fields can suppress the third harmonic completely. Thus, in order to measure $\tilde{\chi}_3$ properly, one has to make sure that the external DC fields, such as the Earth's field, are compensated.

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