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Semi-spin-glass and spin-glass behaviour in $Eu_xSr_{1-x}Se$ with $x = 0.5$ and 0.7

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Abstract. We have investigated the linear and, in detail, the non-linear AC susceptibilities of two insulating spin glasses obtained from the dilution of the antiferromagnetic EuSe with diamagnetic SrSe. The systems studied are the mixed compound $Eu_xSr_{1-x}Se$ with $x = 0.5$ and 0.7. The linear susceptibilities of the samples show a single peak at their spin-glass freezing temperatures of 2.00 ± 0.03 K and 2.88 ± 0.03 K, respectively. However, the non-linear susceptibilities (third harmonics) of the systems exhibit double peaks: one at the spin-glass transition temperature, and the other at a higher temperature somewhat below the Néel temperature of pure EuSe which is 4.6 K. We believe that the origin of the second peak is the remaining antiferromagnetic phase which seems to persist in the systems down to a critical concentration below $x = 0.5$.

1. Introduction

During the past two decades, major efforts in the field of spin glasses [1, 2] have been exerted to elucidate the peculiar characteristics of these random magnetic systems. In spite of the great interest of both experimentalists and theoreticians the spin-glass phenomenon remains somewhat of an enigma. Originally it was generally believed that the 'archetypal' spin-glass phenomenon arises from the existence of Ruderman–Kittel–Kasuya–Yosida (RKKY) interactions causing competition between the magnetic impurities in a non-magnetic metallic matrix [3]. However, later, the spin-glass behaviour was also observed in a certain concentration range of the insulating $Eu_xSr_{1-x}S$ system [4]. In this system the interactions between the ferromagnetic clusters are believed to be short ranged (not like the RKKY type) but the competing ferromagnetic and antiferromagnetic interactions are still present and the randomness of the system causes spin-glass behaviour. It is now well known that all systems exhibiting good spin-glass behaviour share two basic ingredients: randomness and mixed interaction which lead to frustration. Nevertheless, a question may be posed concerning the existence of a spin glass with only antiferromagnetic interactions. Here the $Eu_xSr_{1-x}Se$ system is an interesting example of a material in which the antiferromagnetic coupling dominates the ferromagnetic coupling.

The prominent signature of a spin glass is the sharp cusp found in the low-field AC susceptibility at the so-called 'freezing temperature' T_f. The sharpness [5] suggests a thermodynamic phase transition at T_f. Recently it has been established that non-linear susceptibility χ_{nl} measurements play a decisive role in providing evidence for the occurrence

§ The research work was done at this laboratory.

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of a phase transition at a finite critical temperature [6–8]. Because χ_{nl} directly couples to the spin-glass order parameter, it is more sensitive to the phase transition than is the linear susceptibility. On the basis of the mean-field theory, for instance, the non-linear susceptibilities of spin glasses should diverge according to a power law of the form $\epsilon^{-n(\gamma+\beta)+\beta}$, where γ and β are the critical exponents [9], $n = 1, 2, 3, \ldots$, and $\epsilon \equiv (T - T_c)/T_c$.

EuSe is a metastable antiferromagnet with a very low Néel temperature of 4.6 K and quite a complex magnetic behaviour below T_N [10, 11]. By diluting with non-magnetic SrSe a solid solution is obtained of the form $Eu_xSr_{1-x}Se$ with random magnetic sites. The phase diagram of this system has recently been determined by means of specific-heat and linear AC susceptibility measurements [12]. A spin-glass-like phase has been found for Eu concentrations $x \leqslant 0.7$, similar to that of $Eu_xSr_{1-x}S$ for $x \leqslant 0.55$ [4, 8]. Above $x = 0.8$ the system shows smeared-out antiferromagnetic behaviour, and, in the small intermediate-concentration range, antiferromagnetic and spin-glass ordering coexist.

The present paper is concerned with the AC susceptibility measurements performed on $Eu_xSr_{1-x}Se$ mixed compounds for the nominal x-values of 0.5 and 0.7. We have systematically determined the temperature, frequency and AC-field amplitude dependences of both the linear and the non-linear susceptibilities of the system using a conventional mutual-inductance technique [7, 8, 13]. Thus the nature of the interactions which control the magnetic behaviour of the system in the concentration range of $0.5 \leqslant x \leqslant 0.7$ has been explored.

2. Experimental method

The detailed experimental techniques and procedures have already been provided in our previous publications [7, 8]. The samples used in this work were supplied by Westerholt and Bach [12] and had irregular shapes. Therefore, no attempt was made to correct for demagnetization effects. The temperature range of our measurements is from 1.2 to 4.2 K and the frequency region from 15 to 234 Hz. The AC-field amplitude is 0.7 Oe for the linear susceptibilities and 7 Oe for the non-linear susceptibilities. Furthermore, in order to investigate the AC-field dependence of the third harmonic, amplitudes of 2 and 3 Oe were also used. The reasons for using different AC-field amplitudes for the linear and the non-linear measurements have already been explained in our previous publications [7, 8]. The results are all normalized to 1 Oe, i.e. the third-harmonic magnitudes are divided by the square of the AC-field amplitude.

3. Results and discussion

We first present the linear susceptibility results. Figure 1 shows the temperature dependence of the measured in-phase component $\tilde{\chi}_1'$ of the $Eu_{0.7}Sr_{0.3}Se$ compound for four different frequencies. As can be seen from the figure, the linear susceptibility shows a frequency dependence above as well as below the so-called cusp temperature, similar to other spin glasses. However, this frequency dependence seems to disappear above 4.2 K and is consistent with the high-frequency results obtained by Baalbergen [14] for temperatures of 1–12 K, and frequencies of 58.6 Hz–56.6 MHz. The so-called freezing or cusp temperature is estimated to be $T_f = 2.88 \pm 0.03$ K from the lowest-frequency result in figure 1 and does not depend upon frequency within our experimental error. It was impossible to observe the out-of-phase component of the linear susceptibility in the above frequency range. For the second sample, $Eu_{0.5}Sr_{0.5}Se$, the linear susceptibility measurement was carried out for a frequency value of only 234 Hz, since it has been previously determined by Baalbergen [14]. The freezing temperature of this sample, given by its cusp, is estimated to be $T_f = 2.00 \pm 0.03$ K, which is in good agreement with those obtained by others [12, 14].

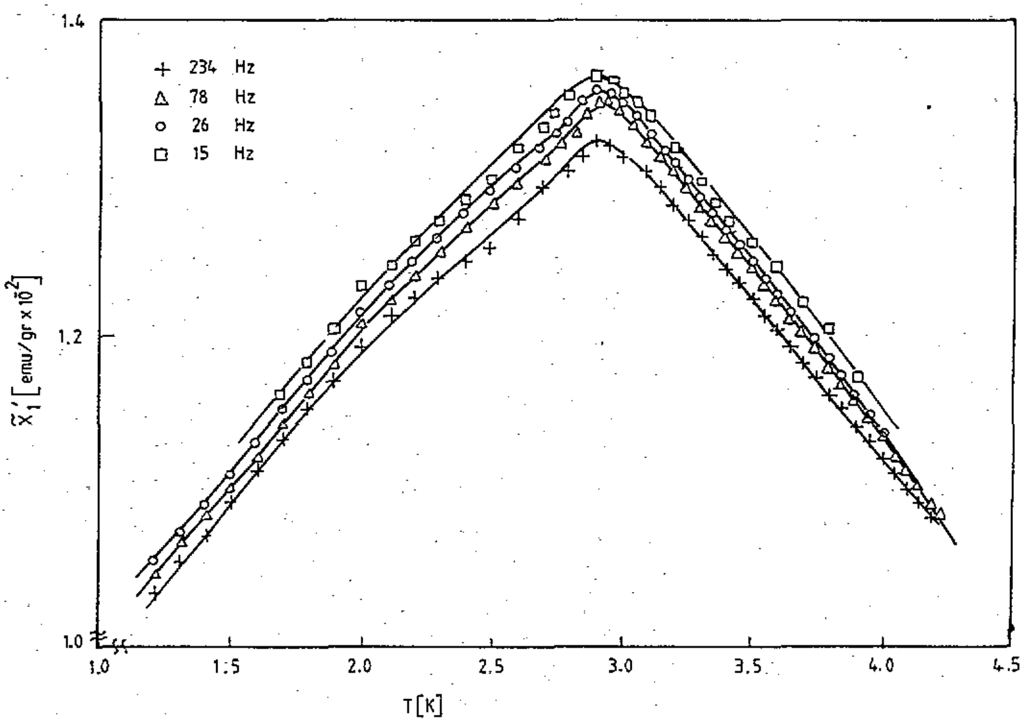

Figure 1. In-phase component of the linear susceptibility of the $Eu_{0.7}Sr_{0.3}Se$ system for several frequencies as a function of the temperature.

Figures 2(a) and 2(b) show the in-phase components $\tilde{\chi}_3'$ of the third harmonics of the non-linear susceptibilities for $Eu_{0.7}Sr_{0.3}Se$ and $Eu_{0.5}Sr_{0.5}Se$, respectively, at different frequencies. The corresponding out-of-phase components $\tilde{\chi}_3''$ of the third harmonics are presented in figures 3(a) and 3(b). As can be seen from the figures, both in the in-phase and the out-of-phase components of the third harmonics contain two well defined peaks. The sharp peaks at the lower temperatures define the spin-glass transition since these maxima occur at the same temperatures as the transition temperatures obtained from the linear susceptibility (see previous paragraph). The magnitudes of the peaks increase with decreasing frequency, similar to the linear susceptibility peaks and as expected. Unexpectedly peaks are found in $\tilde{\chi}_3'$ and $\tilde{\chi}_3''$ at higher temperatures, which are not observed in the linear susceptibility data. By comparing the peaks in figures 2(a) and 2(b) or in figures 3(a) and 3(b) it can be seen that the positions of the higher-temperature peaks shift towards higher temperatures, and they become broader and more dominant with increasing Eu concentration. Furthermore, the magnitudes of these peaks become smaller compared with the spin-glass peaks at the lower Eu concentration. This behaviour indicates that at a critical concentration below $x = 0.5$ the higher-temperature peaks will disappear and the $Eu_xSr_{1-x}Se$ system will become good, i.e. a one-peak spin-glass system. This expectation is further supported by the increase in the magnitudes of the spin-glass transition peaks as the Eu concentration decreases.

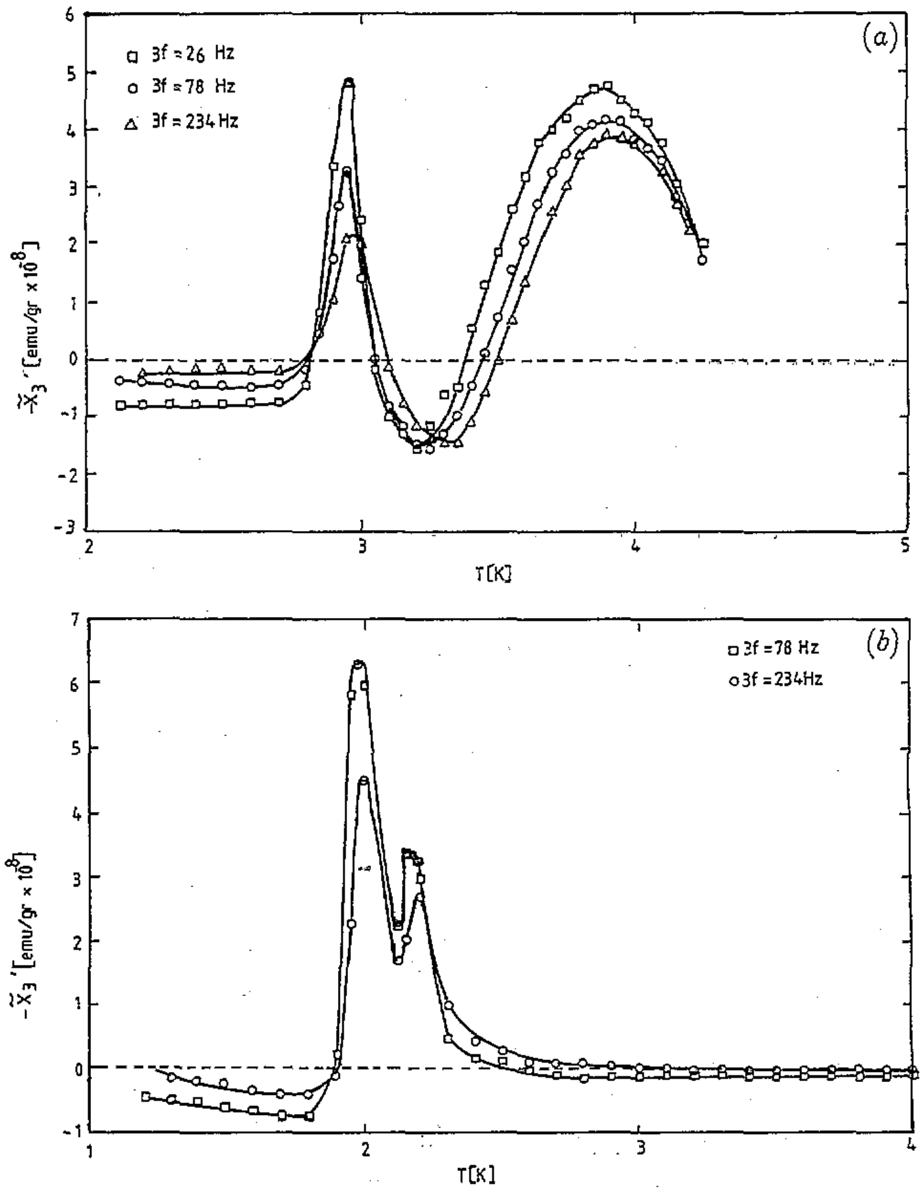

Figure 2. In-phase component of the third harmonic of the non-linear susceptibility of (*a*) the $Eu_{0.7}Sr_{0.3}Se$ and (*b*) the $Eu_{0.5}Sr_{0.5}Se$ systems for several frequencies as a function of the temperature. The measured value of the driving field is $h_m = 7$ Oe.

The shift and increase in the magnitudes of the higher-temperature peaks towards the Néel temperature of EuSe ($T_N = 4.6$ K) with increasing Eu concentration in the $Eu_xSr_{1-x}Se$ system suggest that these peaks are due to the remaining antiferromagnetic structure which possesses sufficient randomness and frustration to exhibit a non-linear response. As the Eu

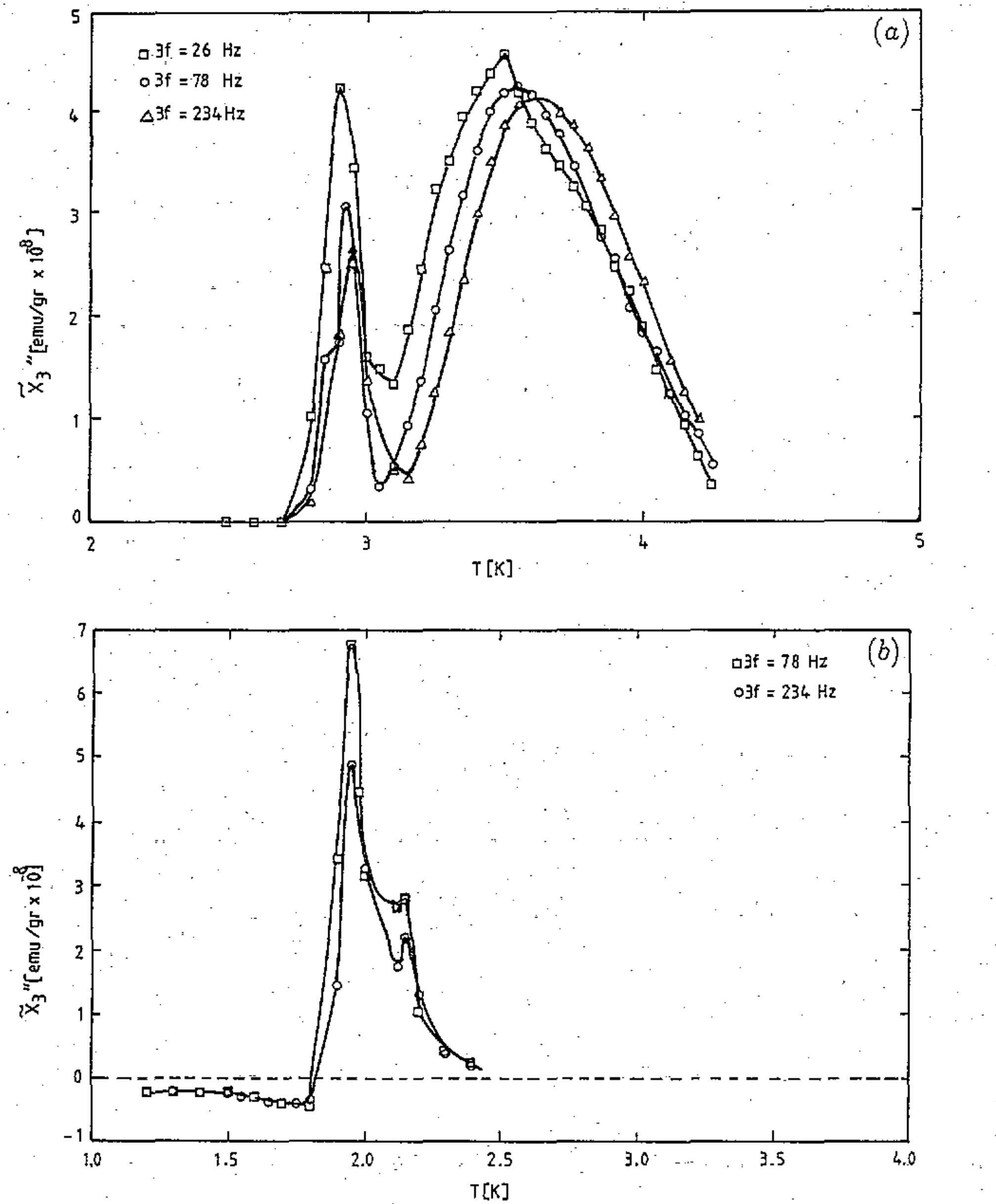

Figure 3. Out-of-phase component of the third harmonic of the non-linear susceptibility of (*a*) the $Eu_{0.7}Sr_{0.3}Se$ and (*b*) the $Eu_{0.5}Sr_{0.5}Se$ systems for several frequencies as a function of the temperature. The measured value of the driving field is $h_m = 7$ Oe.

concentration is reduced, these domains or the 'ghost' of the antiferromagnetism become smaller. Such an explanation can naturally account for the disappearance of the second peak with decreasing Eu concentration of the $Eu_xSr_{1-x}Se$ system. From previous work we know that, in an antiferromagnet, no peak is expected in the third harmonic of the non-linear susceptibility at the Néel temperature for a perfect antiferromagnet [6]. Our measurements on some antiferromagnetic systems confirm this expectation [15]. Thus the higher-temperature peaks observed in our third-harmonic measurements are not due to genuine antiferromagnetic transitions and must be related to the randomness of the mixed compounds. We believe that these peaks are the results of the so-called semi-spin-glass transitions as suggested by De Seze [16] and by Aharony [17]. Here a spin-glass state emerges from a weakened antiferromagnetic state. This latter phase is distinguished by the high-temperature maximum in χ_{nl}.

In order to explore further the origin of the higher-temperature peaks, we have also investigated the AC-field amplitude dependence of the third harmonic for the $Eu_{0.7}Sr_{0.3}Se$ system. As an example in figures 4(*a*) and 4(*b*) we present the in-phase and out-of-phase components of the third harmonic obtained at three different amplitudes for $3f = 78$ Hz.

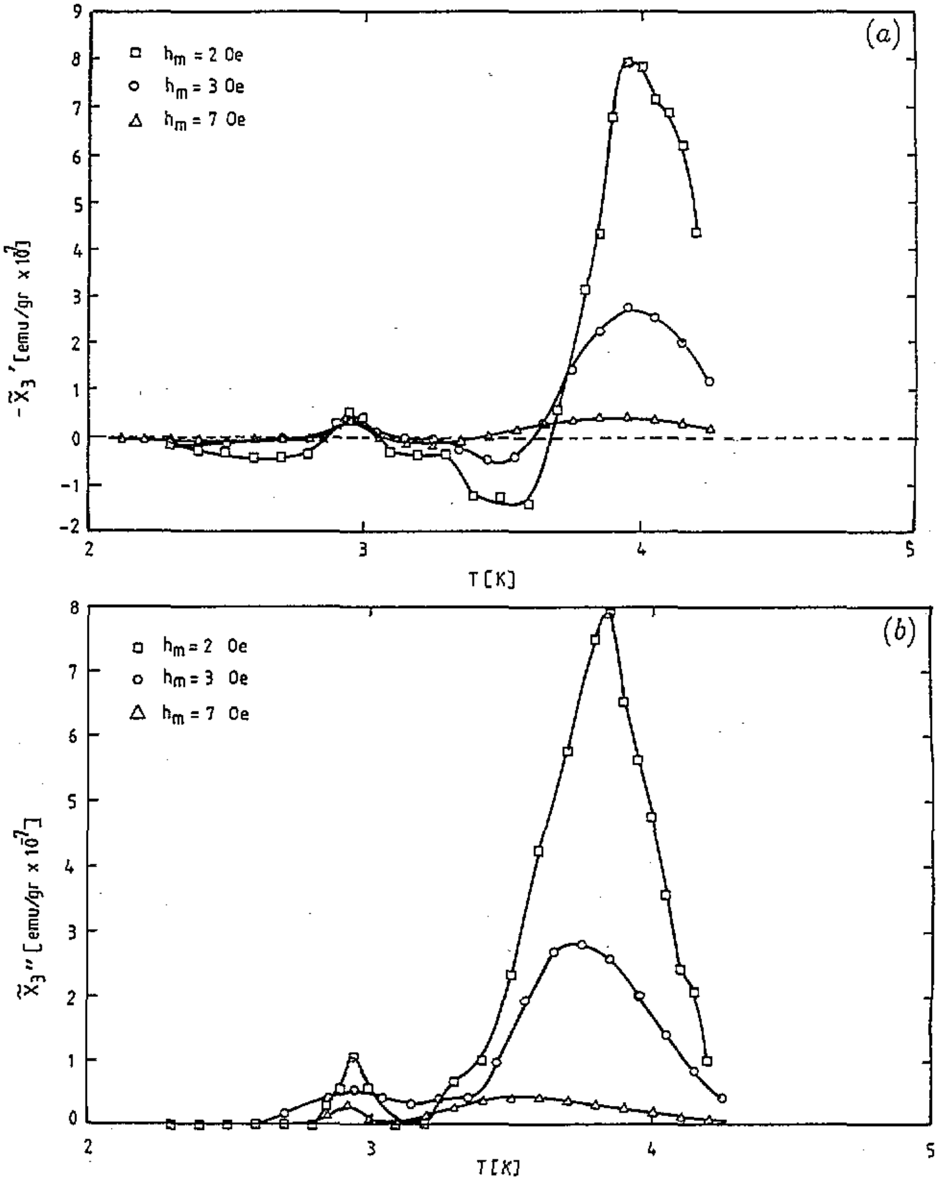

Figure 4. AC-field dependences of (*a*) the in-phase component and (*b*) the out-of-phase component of the third harmonic of the non-linear susceptibility of the $Eu_{0.7}Sr_{0.3}Se$ system for $3f = 78$ Hz as functions of the temperature.

As can be seen from these figures the high-temperature peak is extremely dependent upon the AC-field amplitude while the low-temperature peak (or real spin-glass peak) is hardly affected by the change in the amplitude. A field dependence has also been observed for the linear susceptibility of a diluted antiferromagnet around its transition temperature [18]. Therefore the field dependences of both the linear and the non-linear peaks around the transition temperature seem to be a common feature of a mixed or diluted antiferromagnet.

4. Conclusions and summary

We have investigated the linear and in particular the third harmonic of the non-linear AC susceptibilities of the insulating compound $Eu_xSr_{1-x}Se$, in the spin-glass region of its phase diagram for $x = 0.7$ and 0.5. By examining the linear susceptibilities there seems to be standard non-metallic spin-glass behaviour. However, the third-harmonic measurements indicate that there are two peaks in the temperature range studied. The first peak is at the same temperature as that of the linear susceptibility. Hence it is due to the real spin-glass transition. This peak is almost independent of the AC-field amplitude. On the other hand, the second peak is at a higher temperature and extremely dependent on the AC-field amplitude. Furthermore, this latter peak shifts to higher temperatures towards the Néel temperature of EuSe, and its magnitude increases with increasing Eu concentration. There is evidence that this peak completely vanishes at a particular concentration below $x = 0.5$ and the system becomes a good spin glass. The origin of the second higher-temperature peaks is probably the remains or domains of the pure antiferromagnetic phase with $T_N = 4.6$ K. This phase in the diluted system is represented by a collection of local regions of antiferromagnetic order which form at the temperature of the maximum χ_{nl}. As a conclusion we believe that such antiferromagnetic short-range order prevails in the $Eu_xSr_{1-x}Se$ system down to a critical concentration somewhat below $x = 0.5$.

Acknowledgments

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