

Possibility of dipolar spin-glass in very dilute $(\text{Eu}_x\text{Sr}_{1-x})\text{S}$

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The dynamic susceptibility between 10^{-4} and 10^6 Hz and the magnetic remanence have been investigated on powders and single crystals in the concentration range $0.0005 \leq x \leq 0.15$. Earlier conjectures upon a transition to a dipolar spin-glass associated with either the low- or high-temperature maximum of the susceptibility could not be confirmed. Several arguments are presented that these maxima originate from magnetic blockades of statistically distributed ferromagnetic pairs formed by Heisenberg exchange between first- and third-nearest neighbors. At low temperatures, $T < 0.1$ K, the pair susceptibilities and energy barriers appear to be modified by the weak intercluster interactions.

I. INTRODUCTION

Recently, dilute magnetic insulators have attracted some interest since spin-glass-like phenomena were discovered in such materials. Extensive experimental efforts have been devoted to the system $(\text{Eu}_x\text{Sr}_{1-x})\text{S}$,¹⁻⁸ for which the Heisenberg exchange interactions between the Eu^{2+} are known in much more detail^{9,10} than for metallic spin-glasses. Therefore, a better quantitative interpretation of the spin-glass properties could be expected. In fact, numerical work on the phase diagram, magnetization,¹¹ and excitations¹² of $(\text{Eu}_x\text{Sr}_{1-x})\text{S}$ explains convincingly essential features observed in the region around $x = 0.5$. This spin-glass behavior turned out to originate from frustration of spins or small Eu^{2+} clusters due to the competition between the ferromagnetic near-neighbor ($J_{\text{NN}}/k_B = 0.44$ K) and the antiferromagnetic next-nearest neighbor ($J_{2\text{NN}}/k_B = -0.20$ K) exchange interactions. This "frustrated" spin-glass was found to disappear below the percolation concentration associated with J_{NN} and $J_{2\text{NN}}$, $x_c = 0.13$,¹³ where only finite Eu^{2+} clusters are present.

The nature of the magnetic state below x_c is not yet understood. Measurements of the magnetic dispersion revealed in addition to a maximum at $T > 0.1$ K a second one around 15 mK.² While the former could be ascribed to a blockade of the most frequent cluster, i.e., the ferromagnetic NN pair,^{1,2} the origin of the low-temperature maximum (LTM) remained obscure. Three alternatives were mentioned²: (i) Weak interactions between third-nearest neighbors could create new clusters at low temperature, a conjecture which was renewed by v. Löhneysen studying the specific heat of very dilute $(\text{Eu}_x\text{Sr}_{1-x})\text{S}$.⁶ (ii) Chemical clustering of the magnetic impurities, often occurring in annealed metallic spin-glasses like CuMn ,¹⁴ could generate new spin

configurations not considered by the model of statistically distributed impurities.² (iii) It was argued that the LTM signals the transition to a *dipolar spin-glass*, where the remaining single spins freeze in the random dipolar fields created by themselves and the blocked clusters. The possibility of such a spin-glass originating from the r^{-3} dependence and the randomly alternating sign of the dipolar forces, which are the characteristic features of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction in metallic spin-glasses,¹⁵ too, was also proposed recently by Villain.¹⁶

In contrast, Tholence,⁴ investigating single crystals of $(\text{Eu}_x\text{Sr}_{1-x})\text{S}$, observed both $\chi'(T)$ maxima only in very dilute samples, $x < 0.05$, whereas above this threshold only one cusplike maximum appeared at $T > 0.1$ K. He attributed this cusp temperature, $T_f \sim x$, as a transition to a *dipolar spin-glass* and referred the presence of the additional LTM at $x > 0.05$ in Ref. 2 to an artifact of the powder samples studied there: The long-range dipolar interaction should be reduced in powders compared to single crystals,⁴ so that loose single spins appear which then freeze at the LTM. Obviously, this interpretation introduces $x \leq 0.05$ as a threshold concentration between cooperative freezing and blocking of isolated clusters. This is the essential difference to the former model^{2,7,11} where the percolation limit determines the boundary between freezing of frustrated and isolated Eu^{2+} clusters.

This paper is intended to shed some more light on the central question whether, if at all, a *dipolar spin-glass* exists in $(\text{Eu}_x\text{Sr}_{1-x})\text{S}$. For this purpose we have extended our previous investigations^{2,3} into two directions: (i) Along with powders we also investigated single crystals ($x = 0.0005, 0.005, 0.01, \text{ and } 0.05$), so that a possible difference should directly emerge. (ii) In addition to the dispersion we present results on the absorption and a temporal increment of the magnetization at long times. Both methods probe

only that part of the magnetization which relaxes within the given observation time, $t = \omega^{-1}$, and provide, therefore, a more detailed information on the origin of the magnetic blocking phenomena than the usual measurements of χ' .

II. RESULTS AND DISCUSSION

A. Complex susceptibility

The experimental arrangements for measuring $\chi(\omega) = \chi' - i\chi''$ between 10 Hz and 1 MHz by bridge methods have been described elsewhere.^{2,5} Some representative examples of χ' and χ'' curves at fixed frequency are depicted in Fig. 1. We note that the absorption peaks appearing on the low-temperature flank of the dispersion maxima are much more pronounced and, therefore, better suited for an analysis of the underlying blocking processes than χ' .

If we assume that the blockades simply arise from isolated clusters (configuration index k , see Ref. 2 for the smallest Eu^{2+} clusters in fcc SrS) with

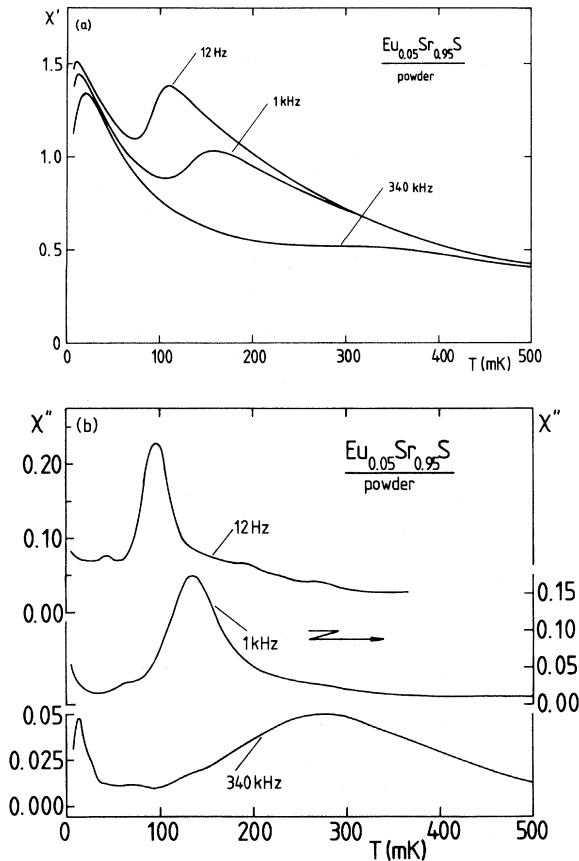


FIG. 1. Temperature dependence of the (a) real and (b) imaginary parts of the dynamic susceptibility at various frequencies in zero magnetic field.

Arrhenius-like flipping rates,

$$R_k = \rho_k \exp\left[-\frac{\Delta_k}{T}\right], \quad (1)$$

then the contribution of the type- k clusters to the time-dependent susceptibility is given by

$$\chi_k(t) = \frac{1}{3} \chi_k^{\parallel} [1 - \exp(-R_k t)] + \frac{2}{3} \chi_k^{\perp}. \quad (2)$$

$\chi_k^{\parallel} = C_k/T$ denotes the Curie susceptibility parallel to the easy direction of the cluster. The transverse susceptibility, χ_k^{\perp} , remains small at $T \ll \Delta_k$, where Eq. (1) is valid. The complex susceptibility is obtained by Fourier transforming $\chi_k(t)$:

$$\chi_k(\omega) = \frac{1}{3} \frac{\chi_k^{\parallel}}{1 + i\omega/R_k} + \frac{2}{3} \chi_k^{\perp}. \quad (3)$$

Provided that the temperature dependence of ρ_k is weak compared to $\exp(-\Delta_k/T)$, the real and imaginary parts of $\chi_k(\omega)$ exhibit maxima at temperatures $T_{m,k}$:

$$\frac{\alpha_k}{T_{m,k}} = \frac{1}{\Delta_k} (\ln \rho_k - \ln \omega). \quad (4)$$

The correction factor α_k is responsible for the observed difference between the positions of the χ' maximum [$\alpha_k' = 1 - (T_{m,k}'/2\Delta_k) \ln(T_{m,k}'/2\Delta_k)$] and the χ'' maximum [$\alpha_k'' = 1 - (T_{m,k}''/\Delta_k)^2$].

Figure 2 reproduces the Arrhenius analyses of the

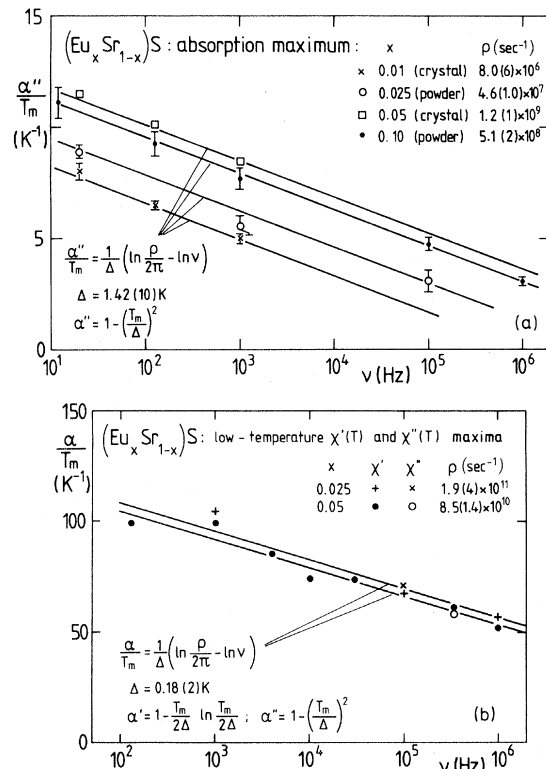


FIG. 2. Arrhenius plots [Eq. (4)] of the (a) high- and (b) low-temperature maxima of the complex susceptibility.

high- and low-temperature maxima. Within the error margins, each set of data can be described by one common value for the energy barrier Δ . The attempt frequencies ρ_k , depending slightly on concentration, assume values in the order of the thermal frequency, $\rho_T = k_B T/\hbar$, in contrast to unphysically high values, $\rho \approx 10^{40}$ Hz, obtained for metallic¹⁷ and insulating¹⁸ spin-glasses. If we compare the barrier, $\Delta = 1.42$ K, associated with the high-temperature maximum (HTM) to the intradipolar barrier of ferromagnetic pairs with diameter r_p ,

$$\Delta^p = 3 \frac{(g\mu_B S)^2}{4\pi\mu_0 r_p^3}, \quad (5)$$

we find good agreement with $\Delta_p(\text{NN}) = 1.20$ K resulting for the most frequent NN pair.

This interpretation is strongly supported by the amplitudes of the χ'' maxima. According to Eq. (3) they are determined by the isothermal susceptibilities

$$\chi_k''(T_{m,k}) = \frac{1}{6} \frac{C_k}{T_{m,k}}, \quad (6)$$

where the Curie constant is given by $C_k = n_k \mu_k^2 / \mu_0 k_B \nu_{\text{spin}}$. For the ferromagnetic NN pair ($\mu_2 = 7\mu_B$) one finds $\chi_2''(T_{m,2}) T_{m,2} = 4.6 n_2$ (K) in good agreement with the data, being plotted for $x \geq 0.01$ in Fig. 3 (after subtraction of a small T -independent background from χ'') versus the nominal concentration of NN pairs, $n_2 = 6x^2(1-x)^{26}$.² The slight deviations at the highest x can be accounted for, if one corrects the Curie susceptibility by a factor

$$\chi_{2,MF}^2 = \frac{\chi_2^2}{1 + 0.2(x/T)^2}, \quad (7)$$

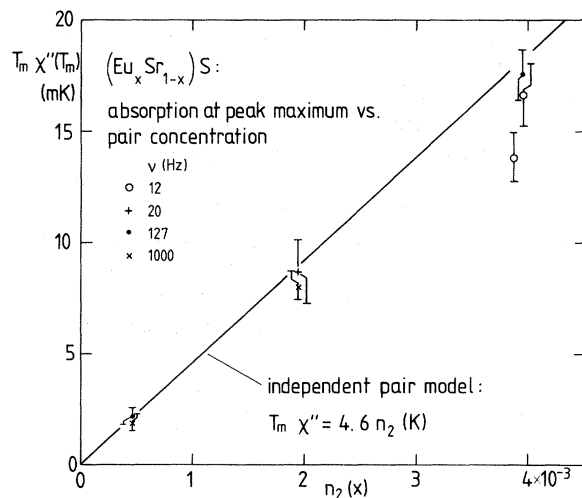


FIG. 3. Amplitudes of the absorption times temperature at maximum vs nominal concentration of ferromagnetic nearest-neighbor pairs, $n_2(x)$, for Eu^{2+} concentrations between 1 and 10 at.%. The full line represents the prediction of the model assuming statistically distributed and noninteracting ferromagnetic Eu^{2+} pairs.

which considers the dipolar interaction of the cluster with its random magnetic environment in a mean-field approximation.² For $T_m = 0.1$ K and $x = 0.1$, e.g., the isothermal susceptibility is reduced by 20%, in reasonable accord with the results (Fig. 3).

Additional support for the present model of independent clusters is provided by the dispersion exemplified by Fig. 4. Above 100 mK, all results can be quantitatively explained by a summing over all clusters including the free single spins, $\chi_1 = C_1(S+1)/3ST$:

$$\chi'(\omega, T) = \chi_1 + \sum_{k \geq 2} \text{Re} \chi_k(\omega), \quad (8)$$

where the flipping rates of the most significant clusters were taken from the experiment.³

The barrier of the LTM, $\Delta = 0.18$ K, is much smaller than expected for all clusters formed by the NN and 2NN exchange (see Table III of Ref. 2). Therefore, it is suggestive to relate the LTM to clusters created by weak interactions between more distant neighbors. In order to illustrate how these interactions come into play, we have calculated the isothermal susceptibilities of Eu^{2+} pairs, $\chi_r^\alpha \propto (S_\alpha^\alpha + S_r^\alpha |S_\alpha^\alpha + S_r^\alpha)_T$, $\alpha = z, \perp$, up to the sixth-nearest neighbors (see Fig. 5) from the pair Hamiltonian

$$H_r = -J_r \vec{S}_0 \cdot \vec{S}_r + H_{\text{dip},r} \quad (9)$$

with J_r from Ref. 10. Obviously, the weak exchange between more distant Eu^{2+} becomes effective below 0.1 K creating, e.g., new pairs as can be seen in Fig. 5, where the susceptibilities change from their bare to the superparamagnetic values. From this picture it is suggestive to ascribe the LTM to the blockade of the

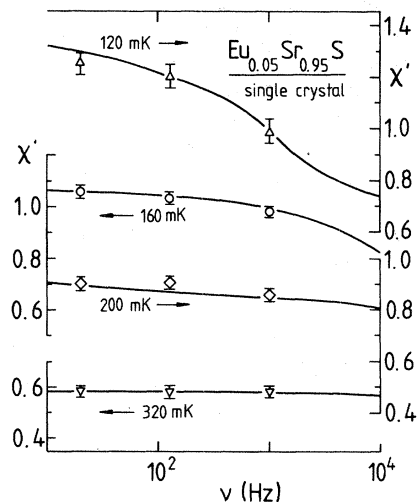


FIG. 4. Dispersion of $(\text{Eu}_{0.05}\text{Sr}_{0.95})_2\text{S}$ at different temperatures. Full lines reproduce calculations based on the model of independent Eu^{2+} clusters, Eq. (8).

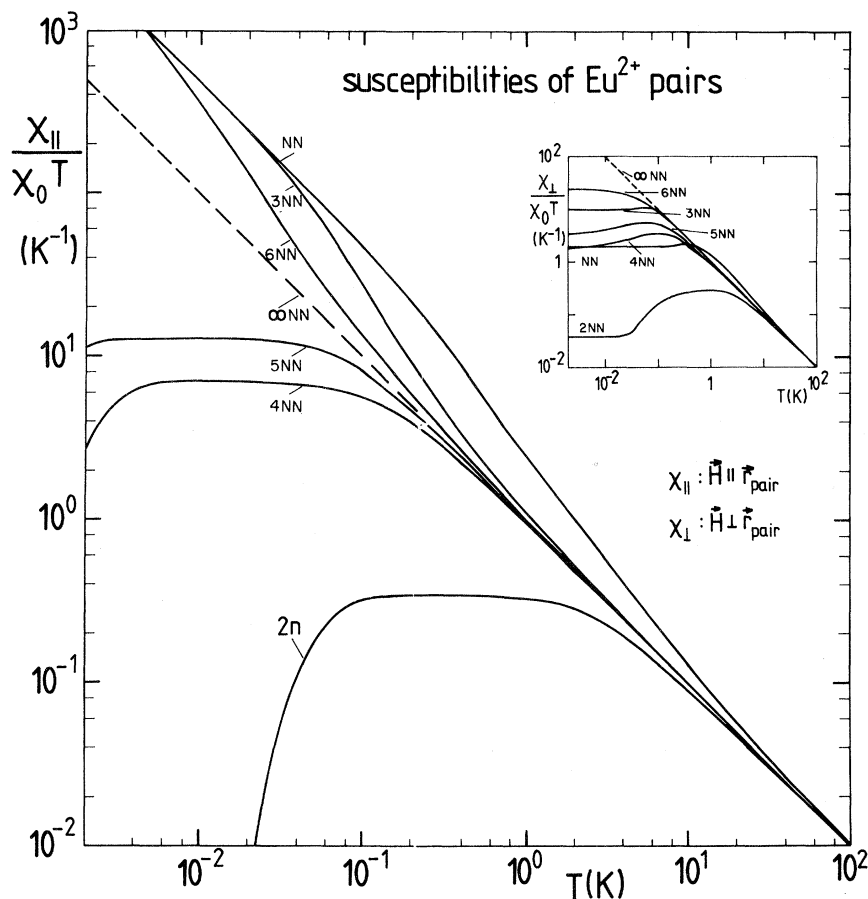


FIG. 5. Temperature dependence of the static longitudinal and transverse susceptibilities of Eu^{2+} pairs formed by exchange and dipolar interaction between the x nearest neighbors ($x=1, \dots, 6, \infty$). χ_0 is the bare susceptibility given by the dotted lines. The calculations are based on the Hamiltonian of Eq. (9).

ferromagnetic 3NN pair, also because it is statistically dominant among the new clusters formed by the long-range forces. In fact, we obtain from Eq. (5) for the barrier, $\Delta_p(3n) = 0.23$ K, which is close to the measured value. We should, however, draw attention to the fact that due to the smallness of $J_{3\text{NN}} = 6\text{mK}k_B$ coherent quantum spin fluctuations may become important.¹⁹ According to a recent estimate,²⁰ the boundary temperature $T_q \approx [J_{3\text{NN}}\Delta_p(3\text{NN})]^{1/2}S/4k_B = 28$ mK is just in the region of the LTM.

In Table I, the absorption amplitudes of the LTM

[see, e.g., Fig. 1(b)] measured on two samples are compared to values calculated from Eq. (6). The concentrations of the 3NN pairs were determined in a "5NN model" taking couplings up to the fifth-nearest neighbors into account as suggested by Fig. 5. The differences between the experimental and calculated data are perhaps related to additional contributions from larger clusters formed by a 3NN pair plus antiferromagnetically coupled 4NN or 5NN spins, since numerical estimates on the dipolar energy barriers of these configurations yielded values close to

TABLE I. Experimental data for the absorption maximum at low temperature compared to calculated values for 3NN pairs, $\chi''_{3\text{NN}}$.

x	ν (kHz)	T_m (mK)	$\chi''(T_m)$	$\chi''_{3\text{NN}}(T_m)$
0.025	100	14	0.22(5)	0.13
0.05	340	16	0.05(1)	0.02

that of the 3NN pair.²¹ However, one should also note that the concentrations are near the percolation limit of the 5NN model, $x_c = 0.035$,¹³ so that the observed enhancement of the LTM may also arise from an unknown cooperative effect.

B. Magnetic remanence and relaxation at long times

According to the magnetization curves in Fig. 6, measured by a SQUID,² after field cooling and reversal of $H = 0.5$ G an irreversible magnetization appears for samples with $x = 0.025$ and 0.05 below temperatures of about 0.1 K. As an interesting feature we found a cusp of the reversible susceptibility, the position of which depended on the warming-up rate dT/dt . This nonequilibrium phenomenon can qualitatively be related to the fact that around the cusp, the growth of χ_{rev} due to the increase of the flipping rates is balanced by the decrease of the isothermal susceptibility of the clusters. Within the model of independent Eu^{2+} clusters, the saturation of χ_{irr} is determined by the susceptibility of the clusters at their blocking temperatures T_k . For a rough estimate, we replace all T_k by a mean value of $T_b = 50$ mK, where half of χ_{irr} has been flipped into the field direction within the observation time, so that we obtain

$$\chi_{irr} = \frac{1}{3} \sum_{k \geq 2} \chi_k^{\delta}(T_b) . \quad (10)$$

According to Table II, the calculated values agree well with experimental data, taken from the maximum difference for χ before and after field reversal. This supports the model of independent clusters and shows that the susceptibility cusp is not associated with a phase transition.

More detailed information on the dynamics of the remanent magnetization can be expected from the

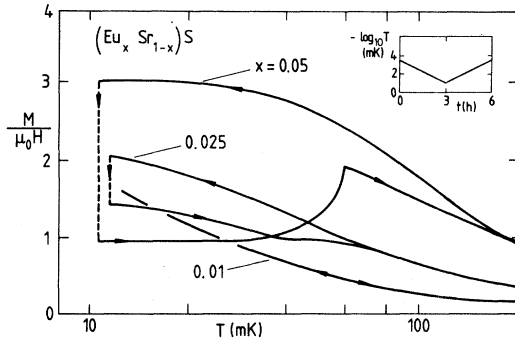


FIG. 6. Magnetic remanence of $(\text{Eu}_x \text{Sr}_{1-x})\text{S}$ single crystals after cooling in a small field, $H = 0.5$ G [$\vec{H} \parallel (100)$], and reversal to $H = 0.5$ G at 10.5 mK. The time scale of the experiment is illustrated by the inset.

TABLE II. Comparison between experimental and calculated irreversible susceptibilities of $(\text{Eu}_x \text{Sr}_{1-x})\text{S}$ at 10 mK.

χ_{irr}	$x = 0.01$	0.025	0.05
Calc.	0.10	0.58	1.88
Expt.	0.0(1)	0.6(1)	2.0(1)

isothermal relaxation below 0.1 K. Comparing the two measuring curves in Fig. 7(a), we note that at 63 mK $\delta\chi(t)$ is near to the isothermal value within the observation window of 100 min, whereas at 36 mK the relaxation process is just in the initial phase. If we evaluate the increment $\delta\chi$ at fixed observation time t as a function of temperature, we find a well-pronounced maximum around 50 mK, which shifts at increasing t to lower temperatures, see Figs. 7(b)–7(d).

Assuming independent clusters, the increment can be obtained from Eq. (1):

$$\begin{aligned} \delta\chi(t) &= \sum_{k \geq 2} \delta\chi_k(t) \\ &= \sum_{k \geq 2} \frac{1}{3} \chi_k^{\delta} [\exp(-R_k t_0) - \exp(-R_k t)] . \end{aligned} \quad (11)$$

Inspection of Eq. (11) shows that at given t each cluster of type k generates a resonancelike maximum as observed in Fig. 7, the position of which, $T_{m,k}^{\delta}$, follows the Arrhenius law, Eq. (4), if we substitute ω by $(t - t_0)^{-1}$ and α_k by $1 + p_k \ln \ln [(R_k t + p_k) / (R_k t_0 + p_k)]$, where $p_k = T_{m,k}^{\delta} / \Delta_k$. Provided that the reference time t_0 is chosen to be small, i.e., $t_0 \ll t$, $R_k^{-1}(T_{m,k}^{\delta})$, the increment maximum of the cluster k should be equal to its isothermal susceptibility,

$$\delta\chi_k(T_{m,k}^{\delta}) = \frac{1}{3} \frac{C_k}{T_{m,k}^{\delta}} . \quad (12)$$

In Fig. 8 we have indicated the product of the measured peak amplitude and temperature as a function of the concentration of the NN pair, $k = 2$, i.e., of the statistically most significant cluster, for $0.005 < x < 0.15$. Obviously at low concentrations, the data can be well described by the Curie constant in Eq. (12) which also includes $x = 0.005$ not shown there.²² The differences at larger x can be explained if we consider the dipolar intercluster interactions by the mean-field approximation, i.e., Eq. (7): see full circles in Fig. 8. This correction is for the $\delta\chi$ maximum more significant than for χ'' (Fig. 3), because the $\delta\chi$ maximum occurs at lower temperature due to the much longer observation time.

Another difference appears when we compare the measured positions of the $\delta\chi$ maximum with the values between 55 and 60 mK following from an ex-

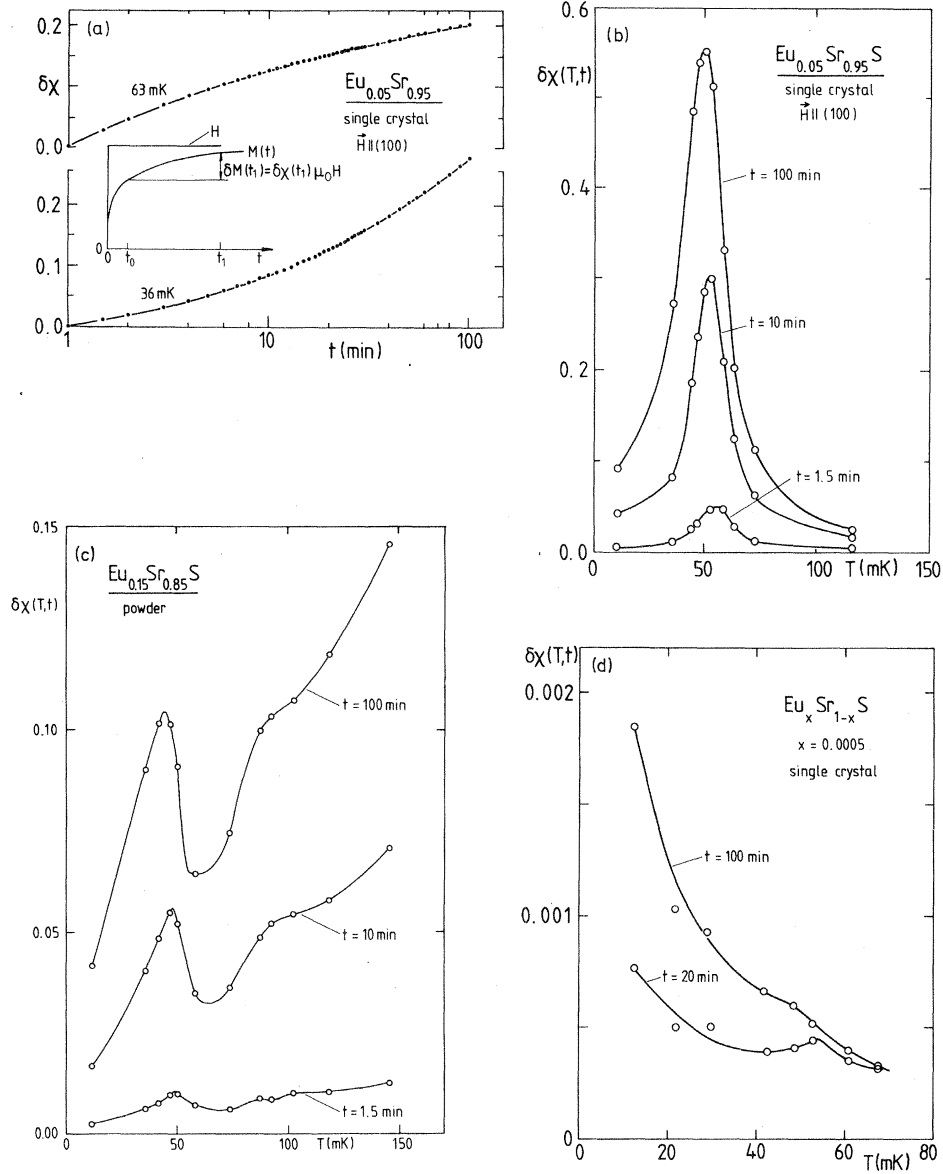


FIG. 7. (a) Increment of the time-dependent susceptibility, $\delta\chi(t)$, measured from $t_0 = 1$ min after turning on the magnetic field (see inset, $H = 0.5$ G). Temperature dependence of the increment measured between t_0 and different final times t for (b) $x = 0.05$, (c) for $x = 0.15$, and (d) for $x = 0.0005$ (Ref. 22).

trapolation of the Arrhenius law, Eq. (4), to the observation time of $\omega^{-1} = 6 \times 10^3$ sec: The experimental values are overestimated by about $\delta T_m = 7(3)$ mK. Since an increase of the attempt frequency is unlikely, a more realistic reason might be a reduction of the energy barrier of the NN pair. This can be done by a local magnetic field H_k acting on the cluster²³:

$$T_{m,k}(H_k) = T_{m,k}(0) \left(1 - \frac{H_k}{H_k^*} \right)^2, \quad (13)$$

where the anisotropy field is given by $H_k^* = 2k_B \Delta_k / \mu_k$. Such a local field may arise from neighbored single spins freezing at the low temperatures via the weak long-range couplings. According to a model proposed for spin-glasses by Ma,²⁴ the ferromagnetic NN pair acts as a nucleus attracting a cloud of loosely coupled spins. In principle, the field H_k exerted on the nucleus by the cloud can be calculated by a self-consistent minimization of the free energy of the whole cluster,²⁴ but due to frustration effects within the cloud a real calculation of H_k seems to be very

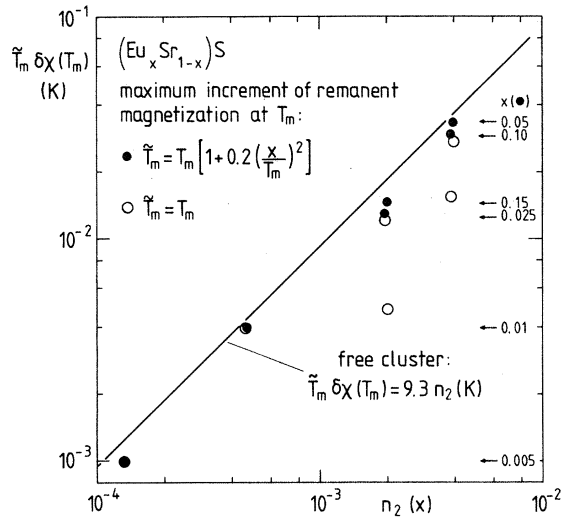


FIG. 8. Amplitude of the magnetization increment times temperature at the maximum (open circles) vs pair concentration. Full circles contain a correction term from intercluster interactions [Eq. (7)].

difficult.¹⁹ We note that the mean dipolar field at the NN pair,² $H_{2,dip} = 35x$ mT, is much smaller than the local field, $H_2 = 36$ mT, required to account for the measured shift $\delta T_{m,2}$ by Eq. (13).

III. CONCLUSIONS

Our experimental material on the magnetization dynamics of $(Eu_xSr_{1-x})S$ reveals no evidence for a transition to a dipolar spin-glass, neither at the HTM nor at the LTM of the susceptibility as was conjectured recently.^{1,2} The dynamical shift and the dispersion, absorption, and increment amplitudes of the HTM show that in the wide range of concentration, $0.0005 \leq x \leq 0.15$, the HTM arises from a blocking

of statistically distributed and independent ferromagnetic NN pairs by their internal dipolar interaction. This also excludes the presence of chemical clusters in our powders and single crystals.²⁵ A cluster effect may perhaps be responsible for the different observation by Tholence *et al.*^{1,4} for $x \geq 0.05$, which would also explain the absence of the LTM in these samples, because the single spins required for it are absorbed within the chemical clusters. Finally, with regard to the r^{-3} decay of the dipolar forces, the postulated breakdown of the dipolar spin-glass transition, $T_{dip} \propto x$, at $x = 0.05$,⁴ appears not to be justified.

From similar analyses of the LTM we conclude that this is most likely associated with a blockade of the ferromagnetic 3NN pair, in accord with a recent suggestion based on the specific heat.⁶ The main argument against a transition to a dipolar spin-glass is that the LTM does not depend on the concentration [see Fig. 2(b)], whereas, e.g., the mean-field approach yields a transition temperature, $T_{dip}^{MF} = 0.22x$ K. Moreover, for strongly random systems this approximation is valid only for $T \gg T_{dip}^{MF}$. There, in fact, the mean-field correction factor, Eq. (7), describes quantitatively the observed reduction of the isothermal susceptibility of the NN pairs. We believe that the answer to the question, whether a cooperative dipolar spin freezing happens in $(Eu_xSr_{1-x})S$, would require measurements on low-concentration samples (no exchange-coupled clusters) at very low temperatures, $T \ll T_{dip}^{MF}$.

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