Spin-Freezing Process in Spin-Glasses

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The authors report on susceptibility (χ) measurements of insulating spin-glasses $Eu_xSr_{1-x}S$ in the high-frequency region 0.1 to 12 GHz. For $x=0.43$ an anomalous dependence of χ on temperature is found with a dip instead of a χ maximum at 8 and 12 GHz. A study at 12 GHz over the $Eu_xSr_{1-x}S$ series shows that the dip is more pronounced near the ferromagnetic phase boundary. The behavior can be understood as arising from a competition between $\chi(T,\nu)$ of spin-glass and ferromagnetic short-range order.

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The nature of the transition into the spin-glass state is still an unsolved problem, in spite of considerable experimental and theoretical efforts.[1] Although the temperature dependence of the ac susceptibility in spin-glasses exhibits sharp maxima, their position T_f is often frequency dependent. It is most remarkable that T_f seems to converge to the static value for low frequencies of the ac measuring field. Recent theoretical work on the short-range Edwards-Anderson model, however, provides evidence against a static phase transition at T_f (see also Binder[2]). Thus it turns out that further studies of the dynamic properties are necessary for a better understanding of the freezing process and the nature of the spin-glass state. Measurements of the magnetic susceptibility χ over a range of frequencies give valuable information about the dynamic behavior. But since the usually studied metallic spin-glasses have large losses at high frequencies, measurements have been restricted to rather low frequencies. The recently investigated[3] insulating spin-glass $Eu_xSr_{1-x}S$ does not have this drawback and is therefore a suitable system for studying the freezing process in spin-glasses.

In this Letter we report on susceptibility measurements on the spin-glass $Eu_{0.43}Sr_{0.57}S$ in the frequency range from 100 MHz to 12 GHz. At these high frequencies we observe new features which were not expected from earlier lower-frequency measurements.[3] The temperature of the χ maximum increases with frequency only up to ~ 500 MHz, and subsequently decreases. At 8 and 12 GHz χ even exhibits a minimum (instead of a maximum) as a function of temperature. Experiments on several Eu concentrations in the $Eu_xSr_{1-x}S$ series reveal that this dip is more pronounced near the reentrant ferromagnetic phase boundary. We argue that the observed anomalous frequency dependence does not arise from two-level systems,[4] but rather from an interplay between $\chi(T,\nu)$ of spin-glass type and ferromagnetic short-range order.

Measurements of the susceptibility were carried out with the cavity perturbation technique.[5] For 136, 516, and 676 MHz, capacitively foreshortened reentrant cavities were used[6] and for 967 MHz a reentrant cavity was used. For 8 and 12 GHz we employed cylindrical resonators operating in transmission in TM_{010} mode. Care was taken to couple the cavities only weakly. To measure the resonance frequency of the cavity we used an automatic frequency control in which the oscillator frequency was locked to the maximum of the resonance curve. The frequency was measured with a Hewlett Packard frequency counter, model HP 5345A, with a temperature-stabilized quartz reference. With this counter and on-line computer averaging we achieved a resolution better than 1 part in 10^7.

The samples of $Eu_xSr_{1-x}S$ ($x=0.10$, 0.15, 0.30, 0.43, 0.54, and 0.70) used in this investigation were from the same lot as in earlier experiments.[3] To ensure good thermal contact, powdered polycrystalline $Eu_xSr_{1-x}S$ samples (10–60 mg) were mixed with vacuum grease and spread in a thin layer on cylindrical walls of the cavities in the high-magnetic-field region.[5,6] The shift Δf in resonance frequency f of the cavity on insertion of the sample was always less than 0.1%. The variation of f of the loaded cavities with temperature in the range 0.4 to 20 K was between 2 parts in 10^5 to 3 parts in 10^4. The quality factor Q_L of the loaded cavities was $\geqslant 100$ for all frequencies, except for the lowest one of 135 MHz where a value of 45 was found. The conditions of the cavity perturbation technique, i.e., $\Delta f \ll f$ and $Q_L \gg 1$, were always satisfied[7] and therefore the change in resonance frequency with temperature is proportional to $\Delta\chi$, the corresponding change in the magnetic susceptibility.[5] For 8 and 12 GHz the proportionality factor was calculated from the known field configuration.[5] For frequencies between 135 and 976 MHz the $\Delta\chi$ values have been scaled by use of arbitrary factors (Fig. 1). We did not apply any correction for the demagnetization factor. We carefully measured the cavities with vacuum grease only to establish the background signal and found the resonance frequency to be constant to within 2 parts in 10^6 between 0.4 and 12 K. At still higher temperatures a small systematic decrease of f with temperature was observed, due to thermal expansion of the cavities. This was corrected for in our data.

In all our figures we have displaced the curves vertically with respect to each other for better clarity. Figure 1 shows the temperature dependence of $\Delta\chi$ for $Eu_{0.43}Sr_{0.57}S$ in the frequency range from 135 to 967 MHz. The susceptibility exhibits a maximum at T_f. The T_f values are frequency dependent as plotted in the inset in Fig. 1 (open circles) where T_f values (crosses) extrapolated to $x=0.43$ from earlier lower-frequency measurements of Maletta and Felsch[3] are also included. With increasing frequency up to about 500 MHz T_f increases but subsequently *decreases*. The large uncertainty in T_f for high-frequency data is due to the broadness of the χ maxima. At very high frequencies of 8.3 and 12.2 GHz we find a *dip in the susceptibility* as a function of temperature (Fig. 2). At 8.3 GHz the dip lies at a higher temperature than at 12.2 GHz. Our data at 967 MHz are quite similar to the data of Hess and DeConde at 840 MHz.[8] But our data at 8.3 and 12.2 GHz differ considerably from the data of Holtzberg *et al.*[9] at 9 GHz on $Eu_xSr_{1-x}S$ with $x = 0.44$ and 0.48. The reason for this discrepancy is not clear. Figure 3 shows $\chi(T)$ of $Eu_xSr_{1-x}S$ with various concentrations x at 12 GHz. We notice that the dip is most pronounced for the $x=0.43$ sample.

We start the discussion by considering first

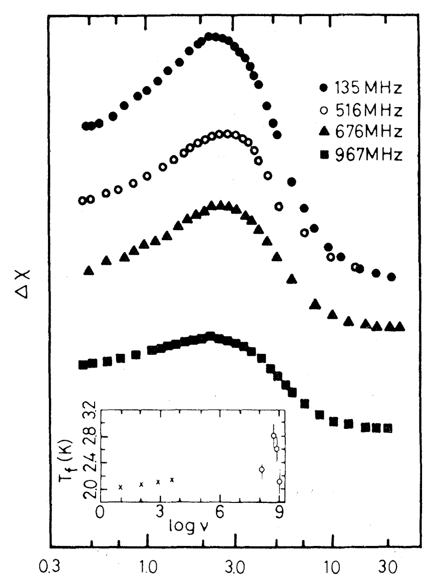

FIG. 1. $\Delta\chi$ (linear scale, scaling factors are arbitrarily chosen for each of the frequencies) vs temperature for $Eu_{0.43}Sr_{0.57}S$ in the frequency range 0.1 to 1 GHz. The temperature T_f of susceptibility maximum is frequency dependent as displayed in the inset.

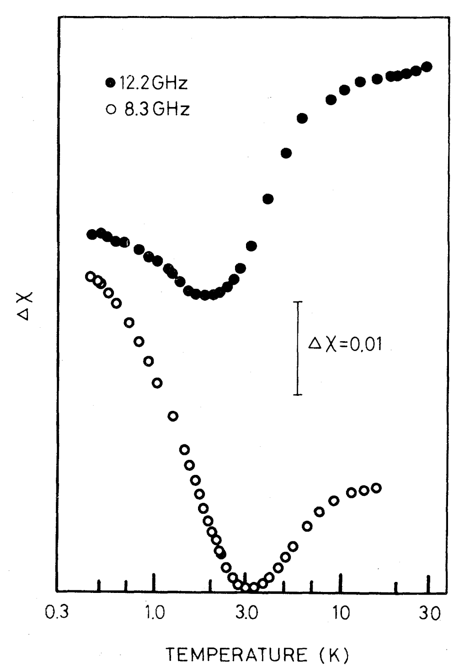

FIG. 2. $\Delta\chi$ vs temperature in $Eu_{0.43}Sr_{0.57}S$ at 8.3 and 12.2 GHz.

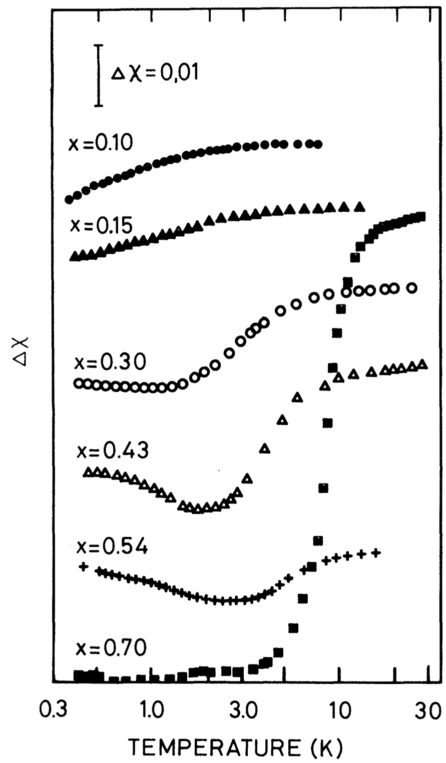

FIG. 3. $\Delta\chi$ vs temperature at 12 GHz in $Eu_xSr_{1-x}S$ with Eu concentrations $x = 0.10$, 0.15, 0.30, 0.43, 0.54, and 0.70.

the highest-frequency data. At 8 and 12 GHz at our lowest temperature we have $h\nu \approx k_BT$. Therefore, it appears quite likely that one may get resonant interaction with the low-energy excitations in spin-glasses, e.g., with *two-level systems*[4] in analogy to the ones found in dielectric glasses. In glasses the dielectric susceptibility indeed shows a dip as a function of temperature, which arises from a competition between the resonant and relaxation absorption of electromagnetic waves by the two-level systems.[10] But we believe that the similarity is superficial for the following reasons: (i) The temperature of the χ minimum shifts with frequency in a way opposite to that found in glasses.[10] (ii) In glasses with constant density of states of two-level systems, the susceptibility decreases as $-\log T$ for $h\nu < 2k_BT$, and the slope of this curve is frequency independent.[10] As is seen from Fig. 2, this is not the case in $Eu_{0.43}Sr_{0.57}S$. We also did not find any evidence for saturation of the microwave absorption even at high input microwave power. We therefore conclude that the observed dip of χ does not arise from two-level systems. Another possibility leading to a minimum in $\chi(T)$ at high frequencies could be the critical slowing down[11] of quasiferromagnetic clusters possibly present in this rather concentrated spin-glass. If this is true, then the dip should be much more pronounced in ferromagnetic samples like $Eu_{0.7}Sr_{0.3}S$, but as shown in Fig. 3 we can rule out this possibility as well.

Finally, we consider the observed behavior in terms of a simple model based upon previous studies of $Eu_xSr_{1-x}S$.[12] The Eu^{2+} spins are coupled via short-range interactions, where the nearest-neighbor exhange is ferromagnetic and the next-nearest neighbor exchange is antiferromagnetic. For x below the percolation threshold $x_p = 0.136$, χ is determined by small independent clusters, in quantitative agreement with ordinary superparamagnetism.[13] For $x \geq x_p$ an infinite connected network of spins exists. Because of competition between ferromagnetic and antiferromagnetic interactions, however, several spins or spin clusters are connected by frustrated and hence weak bonds, and thus samples with $0.13 \leq x \leq 0.51$ show a paramagnetic-to-spin-glass transition.[12] The susceptibility of $Eu_xSr_{1-x}S$ spin-glasses exhibits a maximum for dc and ac measurements[3] at a temperature T_f which is dependent on the measurement frequency ν. The data in Ref. 12 can be interpreted as evidence for a collective freezing process around T_f, as distinguished from thermal blocking of isolated clusters for $x < x_p$. One expects a broad distribution of relaxation times associated with cooperative freezing of the infinite network and blocking of small clusters in spin-glasses. Then, as a result of the dynamic nature of the processes involved, the χ maximum will be broadened and shifted to higher temperature with increasing frequency. We find a drastic slowing down of spin relaxation at T_f,[12] and hence the χ peak will decrease in magnitude with increasing ν. Consequently, the susceptibility at very high frequencies is expected to be dominated by the response of fast spins which are strongly coupled with each other in small-size ferromagnetic clusters.

The tendency to ferromagnetic short-range order gets more pronounced in $Eu_xSr_{1-x}S$ spin-glasses near $x_c = 0.51$. As shown by Maletta *et al.*,[14,15] the interplay of spin-glass type and ferromagnetic order leads to a reentrant ferromagnetic phase boundary at $x = x_c$, and in spin-glasses just below x_c to a ferromagnetic correlation length $\xi(T)$ which always stays finite but shows a maximum versus temperature around T_f. Therefore, one also may expect the relaxation time τ of the strongly coupled spins (probed at high ν) to pass through a maximum according to $\tau(T) \sim d^{-\nu}$, since the normal distance $d(T)$ from the phase line passes through a minimum.[2] These arguments directly explain the observed *minimum* in $\chi(T)$ at very high frequencies in $Eu_{0.43}Sr_{0.57}S$ (Fig. 2) and its concentration dependence (Fig. 3). The compound with $x = 0.54$ also exhibits a spin-glass state at the lowest temperature, but with ξ larger than for $x = 0.43$.[14,15] Hence, the change in ξ with T in $x = 0.54$ is less dramatic, fewer spins are rigidly coupled together in small-size clusters, and thus the dip in $\chi(T)$ at 12 GHz is less pronounced than in the $x = 0.43$ compound (Fig. 3). The anomalous shift of the χ maximum around 0.5 GHz (Fig. 1) may also be attributed to the change of correlated regions with temperature.

In conclusion, the measurement of high-frequency susceptibility of $Eu_{0.43}Sr_{0.57}S$ has led to several interesting new results on the freezing process of concentrated spin-glasses. The maximum in $\chi(T)$ is observed only up to a frequency of ~ 1 GHz. Its position shifts with frequency up to 500 MHz to higher temperature and then to lower T. At still higher frequencies of 8 and 12 GHz one observes a dip instead of a maximum in χ. This anomalous frequency dependence can be qualitatively understood by considering the coexistence of both fast and slow spin relaxations in the freezing process associated with small-size clusters and spin-glass type of order, respectively. From our data it appears important to consider the change of size and relaxation time of ferromagnetically correlated regions with temperature (which must not be simply increasing by lowering temperature) in order to understand the phenomenon of spin-glass freezing.

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