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Phase-transition problem and magnetic short-range order in Heisenberg spin glasses

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We report on measurements of the nonlinear susceptibility for the spin glasses $Eu_xSr_{1-x}S_ySe_{1-y}$ which, depending on the concentration y, possess either ferromagnetic or antiferromagnetic short-range order. Only for the case of ferromagnetic short-range order does the nonlinear susceptibility diverge for $T \rightarrow T_{f0}$; for antiferromagnetic short-range order it is nearly temperature independent for $T \rightarrow T_{f0}$. The results are in favor of a nonequilibrium transition at T_{f0} and demonstrate the misleading role of magnetic short-range order developing above the freezing temperature for the phase-transition problem in real spin glasses.

The possible existence of an Edwards-Anderson type of equilibrium phase transition remains one of the central issues in the physics of spin glasses. For spin glasses with magnetic short-range interactions, computer-simulation studies on small model systems provide the main guidelines for this phase-transition problem. From extensive computer simulations there seems to be general agreement now that in the spatial dimension $d=3$ an equilibrium phase transition can exist in Ising systems, whereas in Heisenberg systems an equilibrium phase transition at finite temperatures cannot exist, the transition temperature being at $T=0$.[1,2]

These conclusions are in puzzling disagreement with experimental results on Heisenberg spin glasses as, e.g., *Cu*Mn,[3] *Ag*Mn,[4] and $Al_{0.63}Gd_{0.37}$,[5] which exhibit a scaling behavior of the nonlinear magnetization consistent with a phase transition at a finite T_{f0}. The values of the critical exponents derived from the experiments are similar to those obtained from computer-simulation studies of three-dimensional (3D) Ising spin glasses.

For spin glasses from the system $Eu_xSr_{1-x}S$, which are good representatives of Heisenberg spin glasses with magnetic short-range interactions, one similarly may conclude from experiments on dynamical scaling[6] and on the critical exponents derived from the susceptibility[7] that a phase transition at T_{f0} exists.

Theoretical arguments were given recently,[8] indicating that a very small anisotropy in Heisenberg spin glasses might be sufficient to establish pure Ising behavior.

The main purpose of this Rapid Communication is to raise some doubt about the conclusions of the existence of an equilibrium phase transition from analysis of the nonlinear susceptibility for spin glasses with higher concentrations of the magnetic atoms, where the development of magnetic short-range order (SRO) above T_{f0} cannot be neglected.

It should be remembered that the interpretation of the nonlinear susceptibility, which plays an essential role in the discussion of the phase-transition problem, is very problematic in spin glasses with magnetic SRO.[9] Only asymptotically with the limits unknown *a priori* does the proportionality of the nonlinear susceptibility χ_{NL} and the Edwards-Anderson order parameter susceptibility χ_{EA} hold in that case.

The spin glasses we analyze in the following are from the pseudoquaternary system $Eu_xSr_{1-x}S_ySe_{1-y}$ which we have introduced in Ref. 10. This system is an extension of the well-known spin-glass system $Eu_xSr_{1-x}S$ and offers some interesting features for the present problem. The ratio of the antiferromagnetic and the ferromagnetic exchange interactions can be varied continuously with the S concentration y: For $y \leq 0.1$ the antiferromagnetic exchange dominates and the magnetic short-range order in the spin glasses is antiferromagnetic;[11] for $y > 0.1$ the ferromagnetic exchange dominates and, correspondingly, ferromagnetic SRO prevails in the spin-glass phase.[12]

It turns out that the typical dynamic behavior in the spin glasses $Eu_xSr_{1-x}S_ySe_{1-y}$ is essentially independent of the type of magnetic SRO present,[13] whereas the nonlinear susceptibility is strongly influenced.

In Fig. 1 we show the magnetic field dependence of the susceptibility χ for a spin glass with ferromagnetic SRO [Fig. 1(a)] and antiferromagnetic SRO [Fig. 1(b)] for temperatures above the freezing temperature T_{f0}. T_{f0} has been determined from the maximum in the field-cooled dc susceptibility measured at $h=1$ Oe. One observes completely different behavior for both samples: In Fig. 1(a) the slope of the susceptibility isotherms is strongly increasing for $T \rightarrow T_{f0}$; in Fig. 1(b) the slope has a reversed sign and is nearly temperature independent. The initial slope of the isotherms in Fig. 1 gives the nonlinear susceptibility χ_{NL}, which is plotted versus the reduced temperature in the lower part of Fig. 2. For the sample with ferromagnetic SRO one finds a very high critical exponent $\gamma_{NL}=3.4$ from the slope of the straight line for reduced temperatures $\tau > 0.1$; for lower reduced temperatures the slope flattens off strongly towards a very low value $\gamma_{NL}=0.2$. For the sample with antiferromagnetic SRO from Fig. 1(b), χ_{NL} is temperature independent in the experimental range. A second sample from the system $Eu_xSr_{1-x}Se$ with lower Eu concentration is shown in addition in Fig. 2; this sample exhibits even a shallow maximum of χ_{NL} at a reduced temperature $\tau \simeq 0.2$ with χ_{NL} decreasing slowly on both sides.

For the two samples with antiferromagnetic SRO in Fig. 2, χ_{NL} does not show any indication of a phase transi-

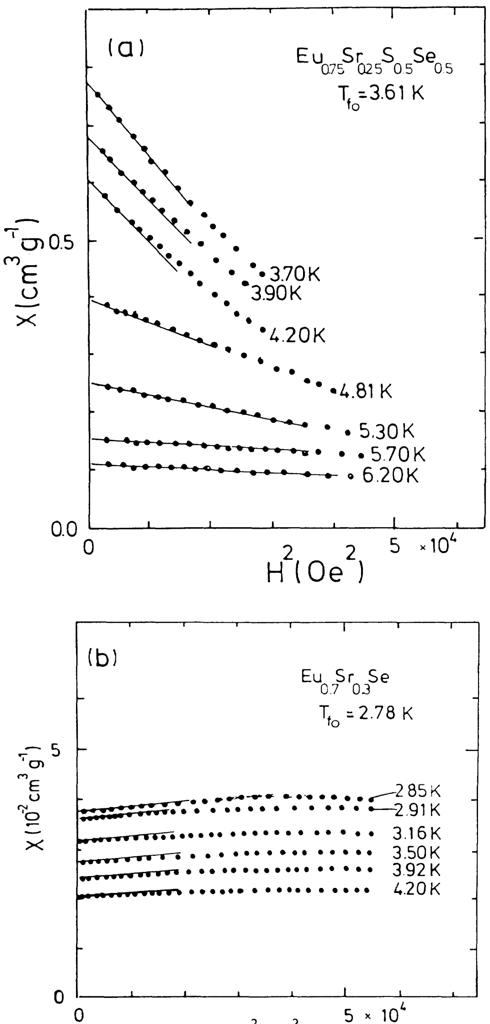

FIG. 1. Magnetic susceptibility vs the squared applied magnetic field at temperatures above the freezing temperature for two spin glasses from the system $Eu_xSr_{1-x}S_ySe_{1-y}$.

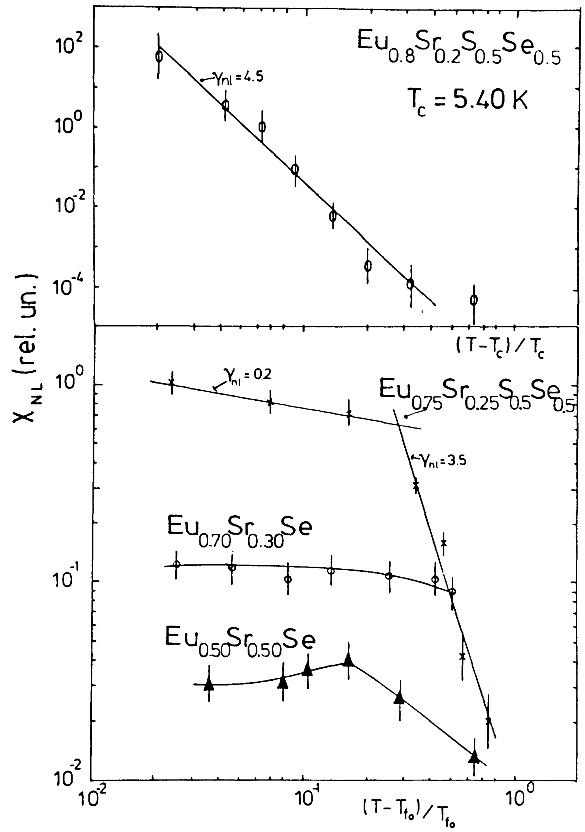

FIG. 2. Nonlinear susceptibility as a function of the reduced temperature for spin glasses (lower figure) and a ferromagnet (upper figure) in direct comparison.

tion at T_{f0}. Interestingly, for the sample with ferromagnetic SRO we can derive an exponent $\gamma_{NL}=3.4$, which is comparable to the values derived for the spin glasses in Refs. 3–5 and is regarded as a characteristic value for a spin-glass phase transition, but it is only valid at rather high reduced temperatures. In addition, we will next present the experimental indications that the strongly temperature-dependent χ_{NL} for $\tau > 0.1$ originates from the development of ferromagnetic SRO and not from the divergence of the Edwards-Anderson susceptibility.

First, we have plotted in the upper part of Fig. 2 the nonlinear susceptibility of a ferromagnetic sample from the system $Eu_xSr_{1-x}S_{0.50}Se_{0.50}$ with slightly higher Eu concentration. Actually, χ_{NL} is not an important quantity in a ferromagnetic phase transition, but it always exists and diverges strongly for $T \rightarrow T_C$ (ferromagnetic order temperature T_C).

One finds that χ_{NL} derived in the same manner as for the spin glasses diverges with an effective exponent $\gamma_{NL}=4.5$; this exponent holds approximately up to rather high reduced temperatures thus showing that the development of ferromagnetic SRO can give rise to a strongly diverging χ_{NL}.

Second, in Fig. 3 we compare the critical behavior of the zero-field susceptibility χ_0 of the same two samples by Fisher-Kouvel plots, which is an important method for the analysis of the critical behavior of the ferromagnet. One finds that the curves for the spin glass with $x=0.75$ and the ferromagnetic sample with $x=0.80$ deviate from the Landau straight line at a temperature of about 20 K and are parallel down to about 5 K. This clearly indicates that the crossover from the Landau range is very similar for both samples, i.e., both samples develop ferromagnetic SRO in a similar manner. Only below about 5 K do the two curves deviate from being parallel: χ_0 diverges asymptotically with an exponent $\gamma=2.1$ for the ferromagnet and crosses over towards a finite value for the spin glass. This crossover behavior of χ_0 for the spin glass correlates with the crossover observed in Fig. 2 from the strongly diverging χ_{NL} towards the weakly diverging χ_{NL}, thus indicating that the ferromagnetic SRO causes the diverging χ_{NL} for $\tau > 0.1$.

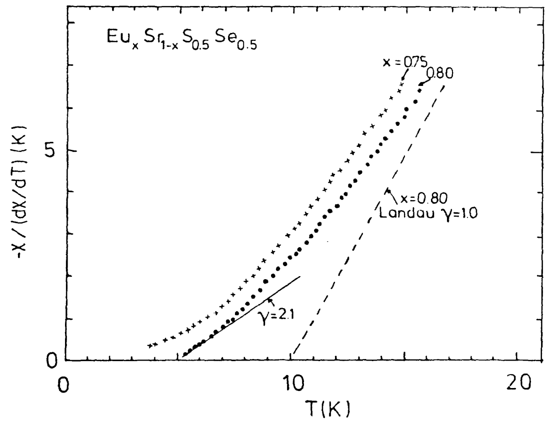

FIG. 3. Fisher-Kouvel plots derived from the zero-field susceptibility for a ferromagnet ($x=0.80$) and a spin glass ($x=0.75$). The dashed line gives the Landau behavior which is reached at temperatures above 20 K.

The results presented here show that in spin glasses the nonlinear susceptibility may be strongly influenced by the development of magnetic short-range order above T_{f0}. The absence of any divergent behavior of χ_{NL} in the case of antiferromagnetic SRO, however, is clearly in favor of a nonequilibrium phase transition at T_{f0} in agreement with the computer-simulation results for Heisenberg spin glasses.

The spin glasses we have chosen above to demonstrate the influence of magnetic SRO have a very high concentration of magnetic ions since in this case the influence of the magnetic SRO is expected to be strongest. Similar characteristic behavior indicating a definite influence of the magnetic SRO seems to exist in other spin-glass systems with higher concentrations of magnetic atoms too.

A crossover from high values of γ_{NL} towards low values at a reduced temperature $\tau \simeq 0.1$ has been observed, e.g., in *Au*Fe,[14] *Pt*Mn,[15] and $(Ti_{1-x}V_x)_2O_3$.[16] This behavior is commonly interpreted as being caused by sample inhomogeneities or the onset of slow magnetic relaxation. But our results indicate that it might as well be due to a crossover from the development of magnetic SRO towards a spin-glass freezing at $\tau \simeq 0.1$.

The correlation of high values of γ_{NL} and ferromagnetic SRO on the one side, and low values for γ_{NL} and antiferromagnetic SRO on the other side, exists in other spin-glass systems too. For $Al_{0.63}Gd_{0.37}$,[5] $Fe_{10}Ni_{70}P_{20}$,[17] and *Cu*Mn (Ref. 3), all with a tendency towards ferromagnetic SRO, high values of $\gamma_{NL} > 2$ have been determined, whereas similar to our $Eu_xSr_{1-x}Se$ spin glasses, *Pt*Mn (Refs. 15 and 18) and $(Ti_{1-x}V_x)_2O_3$, with a tendency towards antiferromagnetic SRO, exhibit low values $\gamma_{NL} \simeq 0$.

Thus, in conclusion, the nonlinear susceptibility in real spin glasses with higher concentrations of magnetic atoms, where the development of a special type of magnetic SRO almost inevitably exists, is difficult to interpret. Only for very dilute Ruderman-Kittel-Kasuya-Yosida-type spin glasses can one rely on the proportionality of the nonlinear susceptibility and the Edwards-Anderson order-parameter susceptibility. For spin glasses with higher magnetic-atom concentrations one must be aware of the fact that the development of ferromagnetic SRO might give rise to a strongly diverging χ_{NL} which can be taken by mistake as an indication of a spin-glass equilibrium phase transition.

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