Evidence for a Phase Transition in the Spin-Glass $Eu_{0.4}Sr_{0.6}S$ from Dynamic Susceptibility Measurements

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The ac magnetic susceptibility of the spin-glass $Eu_{0.4}Sr_{0.6}S$ is found to have a power-law dependence on frequency near the freezing temperature. Extrapolating to zero frequency causes the peak in the real component to sharpen and shift to a lower temperature T_c. That T_c marks a phase transition is supported by exponents describing the divergence of the relaxation time, development below T_c of the order parameter $q_{EA}(T)$, suppression of T_c by a magnetic field, and scaling of the nonlinear susceptibility.

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Most of the interesting and unusual properties that characterize the spin-glass state are of a dynamic nature. In fact "true equilibrium" measurements near T_c may not be possible because of the appearance of very long-relaxation-time phenomena which tend to obscure what may otherwise be a sharp phase transition. It may be possible, however, to infer the nature of this transition by making dynamic measurements and then extrapolating the time dependence to infinitely long measuring times. We report that such measurements of the ac susceptibility $\chi'+\chi''$ of the spin-glass $Eu_{0.4}Sr_{0.6}S$ reveal properties in the zero-frequency limit that indicate the system undergoes a phase transition into the spin-glass state.

Early theories based on mean-field Ising models such as those of Edwards and Anderson (EA)[1] and Sherrington and Kirkpatrick[2] predict the appearance of a novel order parameter $q_{EA}(T)$ below a well-defined temperature T_c. This Edwards-Anderson order parameter is defined by the time-averaged autocorrelation function $q_{EA}=\{\langle S_i(0)S_i(t)\rangle_T\}_J$ in the limit $\tau\rightarrow\infty$. Sompolinsky and Zippelius[3] investigated dynamic generalizations of the EA and the Sherrington-Kirkpatrick models, introducing time-dependent phenomena by means of the Langevin equation. Fischer and Kinzel[4] extended this analysis by including a larger range of frequencies, fields, and temperatures. They predicted that $\chi''\simeq\omega^{\nu(T)}$ near T_f and attributed the abrupt increase of χ'' on cooling through T_f in an applied magnetic field to a crossover from analytic behavior $\chi''\simeq\omega$ at high temperatures to $\chi''\simeq\omega^{\nu(T)}$ on the de Almeida–Thouless line.[5] However, the predicted frequency dependence with $\nu(T)=\frac{1}{2}$ at $T=T_c$ is in disagreement with their analysis of available data, from which values of $\simeq 0.05$ and 0.08 were deduced.

Motivated by this, we have adopted the empirical expression[6]

$$\chi''=A(T)\omega^{\nu(T)} \qquad (1)$$

to describe the temperature and the frequency dependence of our susceptibility measurements on the well-characterized[7] insulating spin-glass $Eu_{0.4}Sr_{0.6}S$ in the temperature regime where peaks in $\chi'(\omega,T)$ are observed. We shall then extrapolate Eq. (1) to the low-frequency limit where such a power law is expected from the Fischer-Kinzel theoretical results. Measurements of χ' and χ'' for a single crystal were carried out with a high-sensitivity ac SQUID magnetometer[8] and lock-in amplifiers over the frequency range 7–5000 Hz. The sample had an approximately ellipsoidal shape, and to minimize demagnetization corrections the ac and dc fields were applied parallel to its long axis. For clarity we distinguish between the susceptibility $\chi_a=M/H_a$ defined by the ac applied field H_a and the internal susceptibility $\chi_i=M/H_i$ defined by the ac internal field $H_i=H_a-4\pi NM$, where $4\pi N=0.76\pm0.05$ is the demagnetization factor. To minimize the perturbation of the spin system, the ac field was kept below 10 mOe, and the cryostat was well shielded from the Earth's magnetic field.

To determine the exponent $\nu(T)$ and coefficient $A(T)$ in Eq. (1), we carried out a least-squares fit to the data for χ_a'' at seven frequencies over the temperature range $1.40<T<1.85$ K, as illustrated in Fig. 1(a) for a few representative temperatures. Since Eq. (1) is expected to hold only in the low-frequency limit, and a systematic discrepancy is shown in Fig. 1(a) for data at our highest frequency (5 kHz), we have not used these data in the subsequent analysis. The temperature dependence of the two parameters is shown in Fig. 1(b) (lines). The coefficient $A(T)$ retains the general features of the $\chi''(T)$ curves. The exponent $\nu(T)$ decreases smoothly from $\nu(T)>0.4$ at $T=1.8$ K to $\nu(T)\simeq0.09$ for $T<1.54$ K, where it levels off.

Figure 2(b) compares the observed $\chi_a''(\omega,T)$ response with that calculated from $A(T)$ and $\nu(T)$. When extrapolating to zero frequency we find that an ultralow-

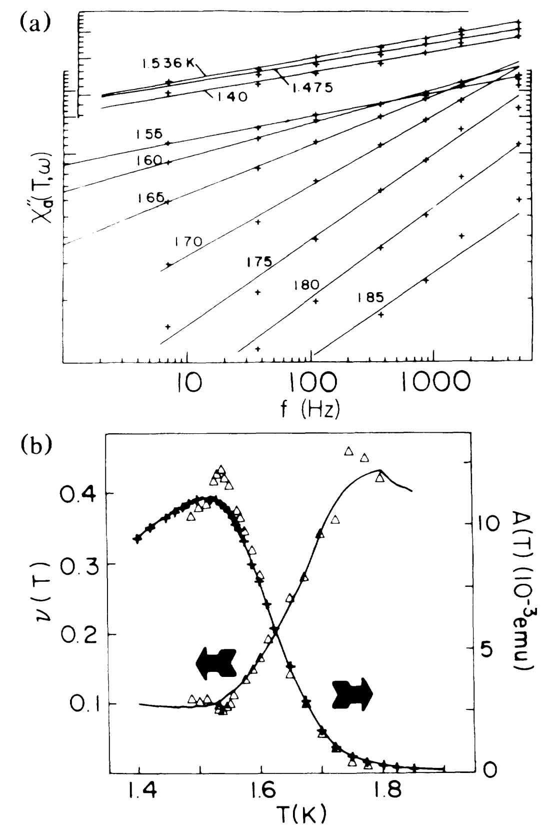

FIG. 1. (a) Frequency dependence of χ''_a for various temperatures near T_f with straight lines showing fits by Eq. (1). (b) Temperature dependence of the amplitude $A(T)$ and the exponent $\nu(T)$ obtained from various fits described in the text.

frequency response to an applied field at 10^{-9} Hz (corresponding to a period of 32 yr) is predicted to display a small, but in principle measurable, signal. For such a measurement, there must also be a corresponding ultraslow cooling rate. This indicates that true equilibrium for this spin-glass is experimentally unattainable.

We have previously shown[9] that the expression $\chi''_a = -(\pi/2)\partial\chi'_a/\partial\ln(\omega)$, which was derived from the Kramers-Kronig relation with the assumption of a broad distribution of relaxation times,[10] is well obeyed for this spin-glass in both zero and finite magnetic fields. This equation may be integrated to predict the behavior of χ'_a in terms of the coefficient $A(T)$ and exponent $\nu(T)$ for χ''_a:

$$\chi'_a = \chi'_{0a}(T) - (2/\pi)[A(T)/\nu(T)]\omega^{\nu(T)}. \quad (2)$$

To demonstrate this we first assume only the values $\nu(T)$ previously obtained from our χ''_a data. This assumption allows the use of a *linear* least-squares-fitting routine for Eq. (2), and thus the two parameters $A(T)$ and $\chi_{0a}(T)$ could be determined from a fit to the χ'_a data (for seven frequencies). The coefficient $A(T)$ obtained in this manner is displayed by the crosses in Fig. 1(b). The agreement with $A(T)$ previously obtained from χ''_a data is excellent.

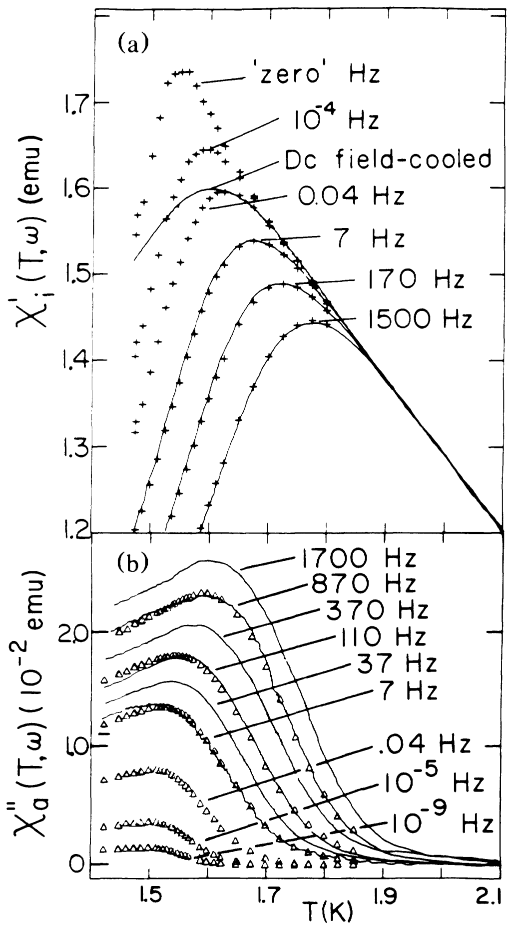

FIG. 2. (a) Measured χ'_i (lines) and predicted behavior (crosses), as described in the text. (b) Measured χ''_a (lines) and predictions (triangles) for a two-parameter least-squares fit by Eq. (1).

Alternatively, all three parameters $\chi_{0a}(T)$, $A(T)$, and $\nu(T)$ may be obtained from χ'_a data alone by means of a *nonlinear* least-squares-fitting procedure [triangles in Fig. 1(b)]. Here the agreement is quite good except within a narrow temperature range near $T \simeq 1.54$ K. The disagreement is most likely due to the peculiarities of the nonlinear three-parameter-fitting procedure, along with the somewhat larger errors associated with the determination of χ'_a. We therefore focus our attention on the first method which has a smaller cumulative error.

For quantitative comparison to theory, the internal susceptibility χ'_i is of interest. This may be calculated from the results of the above analysis by our defining $\chi_i = \chi_a/(1-4\pi N\chi'_a)$, where χ'_a is given by Eq. (2).[11] The results displayed as crosses in Fig. 2(a) are in excellent agreement with the χ_i' data. The extrapolation of the predicted behavior of χ_i to the zero-frequency limit is also shown in the figure. This limit is approached very slowly, with the peak in χ_i' sharpening as it shifts toward lower temperatures. Also shown is the conventional dc susceptibility obtained by cooling of the sample in a constant field of 0.5 Oe at a rate of approximately 200 mK/(10 min). The extrapolated zero-frequency behavior suggests that a phase transition occurs at $T_c = 1.54$ K. We emphasize that the presence of a peak in the zero-frequency limit has been obtained from the measured frequency-dependent susceptibility in the temperature domain where *no peak is directly observed*.

Now we shall demonstrate how the zero-frequency extrapolation provides a self-consistent picture of spin-glass behavior. Figure 2(a) indicates that in this limit the paramagnetic response at high temperatures is continued to much lower temperatures than measurements at finite frequencies suggest. Figure 3 shows that the susceptibility can be accurately described by a Curie-Weiss law down to T_c with $\theta_c \simeq 0.25$ K. The Curie constant is larger than would be deduced from the free-Eu^{++}-ion moment, indicating short-range correlations of approximately 5.3 Eu^{++} ions in the case of ferromagnetic alignment.

The order parameter $q_{EA}(T)$ can be determined from the real component of the magnetic susceptibility by the Fischer relation,[12] which is generalized[13] to

$$\chi \simeq [1 - q_{EA}(T)]/\{T - \theta_c[1 - q_{EA}(T)]\}.$$

We interpret the extrapolation of the susceptibility to the $\omega \to 0$ limit as precisely the desired condition for its determination. The results are depicted in the inset in Fig. 3. From the scaling form $q_{EA}(T) \simeq (1 - T/T_c)^\beta$, the critical exponent is found to be $\beta = 1.02 \pm 0.06$, in excellent agreement with the mean-field prediction of $\beta = 1$.

We turn now to the possibility of critical slowing down just above T_c, which would produce a divergence in the relaxation time[14] as $\tau = \omega^{-1} = \tau_0[T_f(\omega)/T_c - 1]^{-z\nu}$ [the temperature-independent critical exponent ν should not be confused with the temperature-dependent $\nu(T)$ previously discussed]. Previous fits of this divergence expression with experimental data have involved the adjustment of three parameters, τ_0, $z\nu$, and T_c. We carry out a more restrictive fit by assigning T_c the value of 1.54 K predicted by the zero-frequency peak. Figure 4 shows the measured values for T_f, as well as values obtained from the peaks of the low-frequency extrapolated curves and the corresponding best fits. The data and extrapolations are in accord with the divergence expression for values $z\nu = 10.6 \pm 0.6$ and $\tau_0 \simeq 2 \times 10^{-8}$ s, which are in reasonable agreement with predictions.[15] Our exponents also satisfy the relationship $\nu(T_c) = \beta/z\nu$ derived by Malozemoff and co-workers[16] for a fractal cluster model. The power-law exponent is $\nu(T_c) \simeq 0.09$, and the divergence exponents give $\beta/z\nu = 0.094$.

In addition, we briefly mention studies of the susceptibility in finite dc fields H which will be reported in detail in a future publication.[17] We find that the $\nu(T)$ curve for $H = 0$ illustrated in Fig. 1(b) is merely shifted to lower temperatures with increasing field, while maintaining essentially the same shape. We therefore associate the temperature below which $\nu(T,H)$ is independent of T with the transition temperature $T_c(H)$. This avoids questions as to which feature of individual $\chi_a''(\omega,T,H)$

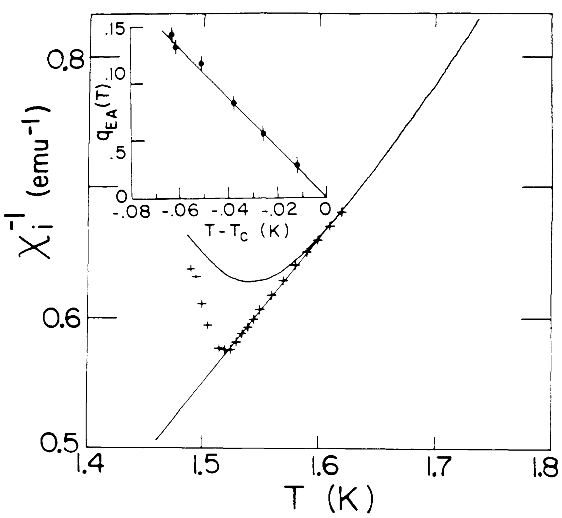

FIG. 3. Inverse of χ_i' measured in 0.5-Oe dc field (heavy line) and the behavior predicted in the zero-frequency limit (crosses). The fit of a Curie-Weiss law is also shown (straight line). Inset: The decrease of the order parameter $q_{EA}(T)$ on approach to T_c.

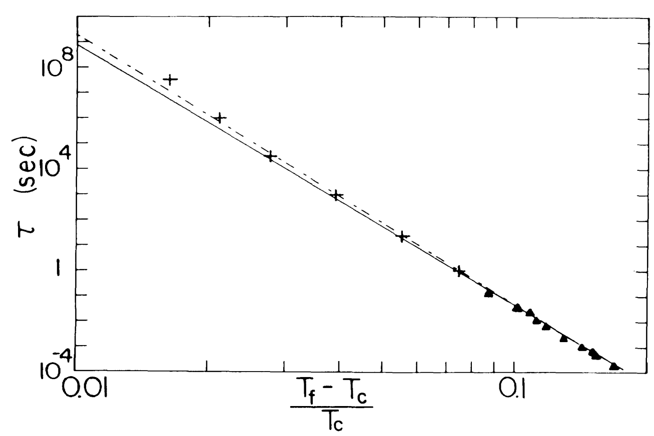

FIG. 4. Temperature T_f of the peak in χ_i for various relaxation times, as measured (triangles) and as obtained from the low-frequency extrapolation (crosses). The solid line is the best two-parameter fit to the data, and the dot-dashed line is the best fit when the data are augmented by the extrapolated values.

curves mark the de Almeida–Thouless line,[18] and in addition produces a frequency-independent result. The suppression of the transition temperature has the form

$$1 - T_c(H)/T_c(0) = K(\mu_B H_i/kT_c)^\theta,$$

where $\theta = 0.70 \pm 0.09$. This is in agreement with the predicted[5,19] mean-field exponent of $\frac{2}{3}$ for Ising spins and is consistent with previous measurements of this spin-glass.[18,20]

Finally, we note results for the scaling of the nonlinear susceptibility above T_c,

$$\chi'_{nl}(T,H) = \chi'(T,H=0) - \chi'(T,H),$$

using the zero-frequency extrapolations of data measured in finite dc fields. With the generalized scaling form $\chi_{nl} \simeq H^{2/\delta} f(t/H^{2/\phi})$, where $t = (T - T_c)/T_c$, a value of $\phi = 3.0 \pm 0.3$ was obtained, in agreement with the scaling relation[21,22] $\phi = 2/\theta$.

In conclusion, the power-law description of the magnetic susceptibility and the extrapolation to zero frequency provide an excellent fit to the experimental results. A consistent picture emerges of a phase transition that could be observed directly if measurements were made with an infinitely long time scale. Nevertheless, the essential features of this transition may be inferred from data obtained at finite frequencies.

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[11]Note added after submission: Alternatively, a linear least-squares fit can be made directly for χ'_i with use of the exponent $\nu(T)$ from χ''_i by analogy with the method previously discussed, the results being identical to those in Fig. 2(a). However, the demagnetization correction gives a larger value for $A(T)$, and more importantly a 10% decrease in $\nu(T)$ at low T because of the frequency dependence of the internal field. Measurements of magnetic noise by W. Reim, R. H. Koch, A. P. Malozemoff, M. B. Ketchen, and H. Maletta [Phys. Rev. Lett. **57**, 905 (1986)] suggest that $\nu(T)$ may depend on frequency at very low frequency; however, the much larger N for their sample has not been taken into account in their analysis.

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