

## Dynamic scaling in the $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ spin-glass

N. Bontemps and J. Rajchenbach

*Laboratoire d'Optique Physique, École Supérieure de Physique et de Chimie Industrielles (ER 05 du CNRS),  
10 rue Vauquelin, 75231 Paris Cedex 05, France*

R. V. Chamberlin and R. Orbach

*Department of Physics, University of California at Los Angeles, Los Angeles, California 90024  
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We have established the zero-field freezing temperature and  $T_f(H, \omega)$  lines associated with a constant relaxation time over a large frequency range ( $10^{-2}$ – $10^5$  Hz) in  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ . The choice of the experimental criterion and the demagnetizing field effects are analyzed in some detail in order to establish the validity and the limits of our analysis. Two different scalings are tested as suggested by Binder and Young:  $T_c=0$ ,  $\ln(\tau/\tau_0)=T^{-zv}f(H/T^\Delta)$ ; and  $T_c\neq 0$ ,  $\tau/\tau_0=(T-T_c)^{-zv}g((T-T_c)^{-\Delta}(H/T))$ . Both are found to work satisfactorily with the following sets of parameters: for  $T_c=0$  K,  $zv=8$ ,  $\Delta=10.5$ , and  $\tau_0\sim 10^{-5}$  s; for  $T_c=1.50$  K,  $zv=7$ ,  $\Delta=2$ , and  $\tau_0\sim 10^{-7}$  s. The latter value of  $\tau_0$  appears to be more consistent with previous relaxation data than the former. Hence, our fitting procedure suggests that  $T_c\neq 0$  K (equal to  $\sim 1.50$  K) for  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ .

### I. INTRODUCTION

Static critical behavior has been extensively investigated in various spin-glasses.<sup>1–3</sup> Susceptibility data appear to exhibit universal scaling, as can be expected in the vicinity of the phase-transition temperature. However, no alternative model has been considered and shown to be inconsistent with the experimental results. It has been recently suggested by Binder and Young<sup>4</sup> that “freezing in spin-glasses can be understood as a consequence of anomalous critical slowing down associated with a zero-temperature phase transition.” They write a dynamic scaling within the assumptions of their model. We shall compare in this paper an analysis of our experimental results on the insulating spin-glass  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$  using their  $T=0$  scaling with dynamic scaling that would be expected in the case of “normal” critical slowing down in the vicinity of a transition at finite temperature.<sup>5</sup>

We rely on dynamic susceptibility data obtained by joining measurements from two experimental procedures and apparatuses, covering a large frequency range on the same sample, in order to test the ability of the two contradictory scaling models to discriminate between the limits of  $T_c=0$  or  $T_c$  finite. We shall show that both scaling models can provide satisfactory fits to the data. However, the characteristic time scale and the critical transition temperature derived on the assumption of a finite  $T_c$  yield values more consistent with previous data on  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ .

We describe briefly in Sec. II the scaling theories for the relaxation time, as suggested by Binder and Young<sup>4</sup> and Binder<sup>5</sup> for the two cases  $T_c=0$  and  $T_c\neq 0$ . We then discuss the actual measurements of the complex susceptibility in Sec. III. We show in Sec. III A which criterion seems the most appropriate in order to establish a constant relaxation-time contour in the  $H$ - $T$  plane. We suggest a new definition of the zero-field freezing temperature associated with a given experimental time scale.

The experimental conditions are specified in Sec. III B.

In Sec. III C we raise and discuss a particular difficulty connected with the high susceptibility and peculiar shape of our sample: the problem of determining the demagnetizing field and its related consequences. We also discuss how we connected the high-frequency measurements performed at the École Supérieure de Physique et de Chimie Industrielles (ESPCI) using the Faraday rotation method with the low-frequency measurements performed at the University of California, Los Angeles (UCLA) using a superconducting quantum interference device (SQUID) magnetometer.

Finally, we present in Sec. IV the two scalings of the experimental data, first in zero field, then in the presence of a magnetic field. We derive the characteristic times and the critical exponents. Our concluding remarks are presented in Sec. V.

### II. SCALING THEORIES FOR THE RELAXATION TIMES

#### A. $T_c=0$

We briefly review in this section the assumptions which appear to be relevant to the model of logarithmic scaling for the case of  $T_c=0$ .<sup>4,6</sup> In this analysis,  $T_c$  is chosen to be zero because the lower critical dimension for spin-glasses is taken to be  $d=4$  for short-range interactions. Near the transition, it is assumed moreover that “fluctuations relating the various locally ordered states in anisotropic systems are ‘walls’; nucleating these walls requires thermal activation . . . [and] one assumes that  $\xi_{EA}$  (the Edwards-Anderson correlation length) also controls the heights of typical free-energy barriers.”<sup>4</sup> It follows that

$$\ln(\tau/\tau_0) \propto \Delta F(\xi_{EA})/T \propto T^{-1}\xi^{z-1/\nu} \propto T^{-zv}. \quad (1)$$

This defines a zero-field freezing temperature  $T_f$  associated with a response time  $\tau$ :

$$T_f \propto [\ln(\tau/\tau_0)]^{-1/zv}.$$

For a given experimental frequency  $\omega$ , taking  $\omega\tau = \text{const}$  (see our analysis in Sec. III A),

$$\ln(\omega/\omega_0) \propto [T_f(\omega)]^{-z\nu}. \quad (2)$$

Equation (2) is the first relation which must be checked experimentally.

Static scaling generates a field dependence for  $\xi_{\text{EA}}$ :

$$\xi_{\text{EA}}(T, H) \propto T^{-\nu} \tilde{\xi}(H/T^\Delta). \quad (3)$$

This yields immediately the field dependence of the relaxation time using Eq. (1):

$$\ln(\tau/\tau_0) \propto T^{-z\nu} f(H/T^\Delta), \quad T_c = 0. \quad (4)$$

Keeping  $\tau$  constant defines a relationship between  $T$  and  $H$  which we call  $T_f(H, \tau)$  or  $T_f(H, \omega)$ ; i.e., a constant-relaxation-time contour. Equations (1) and (4) imply that  $T_f(H, \omega)/T_f(0, \omega)$  is only a function of  $H \cdot \{[T_f(H, \omega)]^{-\Delta}\}$ , so that all  $T_f(H, \omega)$  lines are expected to collapse onto a single line when plotted according to this scaled form.

The numerical values tentatively suggested by Binder and Young are  $z\nu = 4$  and  $\Delta = 7$ . These values are claimed to be consistent with experimental data available for  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ .<sup>7,8</sup> We shall argue that this data, besides exhibiting more scatter than the data reported here, should not be used as a check of the model. We claim that the manner by which  $T_f(\omega)$  and  $T_f(H, \omega)$  have been established in Refs. 7 and 8 is inconsistent within the model itself.

### B. $T_c \neq 0$ .

We now describe the scaling relations that may be expected for the case of "standard" critical slowing down.<sup>5</sup> As one approaches the transition temperature  $T_c$  from above, the response time is assumed to diverge as

$$\tau/\tau_0 \propto \xi_{\text{EA}}^z \propto [(T - T_c)/T_c]^{-z\nu}. \quad (5)$$

Equation (5) defines a zero-field freezing temperature associated with the frequency  $\omega$  taking  $\tau \sim 1/\omega$ :

$$\omega/\omega_0 \propto [(T - T_c)/T_c]^{z\nu}. \quad (6)$$

Upon application of an external static magnetic field, the dependence of the correlation length on field should therefore be

$$\xi_{\text{EA}}(T, H) \propto [(T - T_c)/T_c]^{-\nu} \times \tilde{\xi}((H/T)[(T - T_c)/T_c]^{-\Delta}), \quad (7)$$

which yields the field dependence of the relaxation time,

$$\tau/\tau_0 \propto [(T - T_c)/T_c]^{-z\nu} g((H/T)[(T - T_c)/T_c]^{-\Delta}). \quad (8)$$

Setting  $\tau$  equal to a constant,  $T_f = T_f(0, \omega)$ , and  $T = T_f(H, \omega)$  to simplify notation,

$$[(T_f - T_c)/T_c]^{-z\nu} = [(T - T_c)/T_c]^{-z\nu} \times g((H/T)[(T - T_c)/T_c]^{-\Delta}), \quad (9)$$

so that the ratio  $(T - T_c)/(T_f - T_c)$  is only a function of

$(T - T_c)(H/T)^{-1/\Delta}$ . Thus all  $T_f(H, \omega)$  lines should collapse if plotted in this scaled form. The ratio  $H/T$  has been considered rather than  $H/T_c$  in order to enlarge the critical region.

The above analysis holds for the case of a given relaxation time  $\tau$ . However, spin-glasses do not exhibit a single relaxation time but rather a broad spectrum, even above a "static" transition temperature.<sup>9,10</sup> Mackenzie and Young have shown<sup>11</sup> that this spectrum of relaxation times, within the Sherrington-Kirkpatrick model, diverges below  $T_c$ . Similarly, Binder and Young have argued<sup>4</sup> that the spectrum of relaxation times diverges as  $T \rightarrow 0$  for a zero-temperature phase transition. In both treatments, all times appear to scale in the same way. We shall assume that the same situation is relevant for the distribution of response times in the  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$  spin-glass; that is, that any relaxation time  $\tau$  picked out of the spectrum should follow Eq. (4) or (8), with its own prefactor depending on the model. We shall discuss next which time is actually extracted using our approach, and how this depends upon the choice of the criterion which defines  $T_f(H, \omega)$ .

## III. EXPERIMENTAL DERIVATION OF CONSTANT-RELAXATION-TIME LINES

### A. Discussion of a proper criterion

In this section, we show how we derive a constant-relaxation-time contour from measurements of the real and imaginary part  $\chi'$  and  $\chi''$  of the susceptibility. We also try to make clear which time will be extracted. It is well known that in the simple case of an exponential decay

$$\begin{aligned} \chi' &= \chi_0 / (1 + \omega^2 \tau^2), \\ \chi'' &= \chi_0 \omega \tau / (1 + \omega^2 \tau^2). \end{aligned} \quad (10)$$

Though these relations are certainly incorrect for the magnetization (or susceptibility) of a spin-glass, they exhibit the features which lead to our choice of an appropriate criterion for the determination of the  $T_f(H, \omega)$  lines. The susceptibilities  $\chi'$  and  $\chi''$  depend upon  $\chi_0$  and  $\tau$ , and both of the latter depend on  $H$  and  $T$ . Therefore, such a widely used criterion as the maximum of  $\chi'$  to define  $T_f(\omega)$  does not actually determine the correct relationship between  $H$  and  $T$  because of the dual dependence of  $(\chi_0, \tau)$  on  $T$  and  $H$ . We must use an analysis which generates an  $H$ - $T$  line for a specific response time  $\tau$ . The maximum of  $\chi'$  criterion certainly generates an  $H$ - $T$  line, but  $\tau$  is not necessarily constant along that line. Similarly, choosing the inflection point exhibited by  $\chi''(\omega, T, H)$  as a criterion to establish  $T_f(H, \omega)$  lines is also not correct if these lines are to be associated with a constant relaxation time:  $d^2\chi''/dT^2 = 0$  does not imply that  $\tau(H, T)$  is constant. Yet another criterion, namely,  $\chi''(\omega, T, H) = \text{const}$ , has been used by two of the present authors to derive  $(H, T)$  lines in a limited range of frequency.<sup>12</sup> This is again incorrect for the reasons quoted above. Therefore we do not believe that one may rely on previous data on  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$  (Refs. 7, 8, and 12) to check the scaling theories. One must use an analysis which generates an

$H$ - $T$  line at a specific response time  $\tau$ .

We now discuss what we propose to be an appropriate criterion. It is obvious that in the simple case described by the Eq. (10) the quantity of interest is

$$\tan \phi = \chi''/\chi' = \omega\tau. \quad (11)$$

Thus, setting  $\tan \phi \sim \phi$  to a constant value does define a set of  $T_f(0, \omega)$  values and  $T_f(H, \omega)$  lines for which  $\tau$  is indeed a constant. Of course, Eq. (11) is very restrictive, but in fact any expressions for  $\chi'$  and  $\chi''$  for which the ratio is only a function of  $\omega\tau$  will yield the same conclusion.

We must examine now how this statement may be generalized to the case of a distribution of relaxation times. For a spin-glass, in the paramagnetic regime, following Ref. 9 we write

$$\chi'' = (1/h_0) \int m(\tau)g(\tau)\omega\tau(1+\omega^2\tau^2)^{-1}d\tau, \quad (12)$$

$$\chi' = (1/h_0) \int m(\tau)g(\tau)(1+\omega^2\tau^2)^{-1}d\tau, \quad (13)$$

where  $g(\tau)$  is the distribution of relaxation times  $\tau$  (which may depend upon the magnetic moment  $m$  so that, in turn,  $m$  may depend on  $\tau$  as written).

We now make the following (trivial) remark. The ratio  $\tan \phi = \chi''/\chi'$  is, to within a multiplying factor  $\omega$ , merely the center of gravity  $\bar{\tau}$  of the distribution  $m(\tau)g(\tau)$  of the magnetic moments weighted by the "cut-off" function  $(1+\omega^2\tau^2)^{-1}$ :

$$\tan \phi = \omega\bar{\tau}. \quad (14)$$

The quantity  $\bar{\tau}$  therefore has the dimension of a time, but it is not entirely characteristic of the system since it depends upon  $\omega$ . However, for  $\omega\tau \ll 1$  for the explicit form of Eqs. (12) and (13),  $\bar{\tau}$  will not depend on  $\omega$ . This condition ( $\omega\tau \ll 1$ ) will be realized at high enough temperature.

We can specify the regime  $\omega\tau \ll 1$  operationally because it implies that

$$\chi' \simeq (1/h_0) \int m(\tau)g(\tau)d\tau.$$

Therefore, when  $\chi'(\omega)$  departs from the static value  $\chi_0$  (e.g., typically, when it goes through its maximum), the condition  $\omega\tau \ll 1$  is no longer fulfilled. It has been widely observed<sup>8,10</sup> that the maximum of  $\chi'$  corresponds to the inflection point of  $\chi''$  in zero field. We therefore adopt the following empirical definition for the range of validity of the assumption  $\omega\tau \ll 1$ : The phase angle  $\phi$  must remain small compared to its value at the inflection point which we denote as  $\phi_x$ . When this is achieved, then  $\phi = \omega\bar{\tau}$  extracts a characteristic time  $\bar{\tau}$  of the system. We obtain, therefore, the zero-field freezing temperature  $T_f(0, \omega)$ , as well as the constant-relaxation-time contours  $T_f(H, \omega)$ , by setting  $\phi$  to a constant value. This value can be arbitrary, but must remain small compared to the value at the inflection point,  $\phi_x$ .

We now show that this criterion works; i.e., it is consistent with the experimental results. We first choose, from two set of measurements performed at two different frequencies ( $10^4$  and  $10^3$  Hz), two  $T_f(H, \omega)$  lines. We adjust the chosen values of  $\phi(\omega)$  for the two frequencies in such a way that  $T_f(0, 10^4 \text{ Hz}) \equiv T_f(0, 10^3 \text{ Hz})$ . The inset of Fig. 1, which illustrates this particular choice, also shows that in both cases the chosen value of  $\phi$  is well

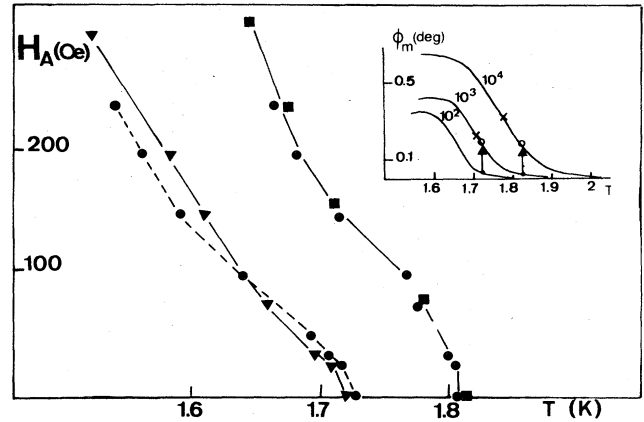


FIG. 1.  $T_f(H, \omega)$  lines reported for two different sets of frequencies as a function of the applied field  $H_A$  (■,  $10^4$  Hz; ●,  $10^3$  Hz; ▲,  $10^2$  Hz) using for each set two different criteria in order to define the same line. The inset shows first the choice of the low-level criterion (see text) for  $10^3$  and  $10^2$  Hz to determine  $T_f(H, \omega)$  from the measurement of the phase  $\phi_m = \chi''/\chi'$ ; then second, how to connect to this choice the high-level criterion for  $10^4$  and  $10^3$  Hz, respectively. The crosses locate the inflection point.

below  $\phi_x$ . The two values are then expected to define the same  $T_f(H, \omega)$  line. Figure 1 shows that this is indeed the case. We now turn to another set of two frequencies ( $10^3$  and  $10^2$  Hz) and again adjust the chosen values of  $\phi(\omega)$  in order that  $T_f(0, 10^3 \text{ Hz}) \equiv T_f(0, 10^2 \text{ Hz})$ . The inset of Fig. 1 shows that this condition is realized for a value of  $\phi(10^3 \text{ Hz})$  which is now quite close to  $\phi_x$  (because of the decrease of  $\phi$  with  $\omega$ ), whereas the corresponding value  $\phi(10^2 \text{ Hz})$  is still much smaller than  $\phi_x$ . The choice of  $\phi$  at  $10^2$  Hz is therefore expected to lead to an incorrect conclusion, whereas it should lead to a correct conclusion for  $10^2$  Hz. If this is true, then the two  $T_f(H, \omega)$  lines should not coincide. Figure 1 shows that indeed they do not, but rather that they depart from each other quite systematically. The experimental results are thus consistent with our use of a constant value for  $\phi$  ( $\phi \ll \phi_x$ ). The data reported in the subsequent sections proceed from this analysis.

## B. EXPERIMENTAL PROCEDURES

In order to cover the largest range of frequencies, we have used two different techniques for measuring  $\chi''$  and  $\chi'$  on a single sample. The high-frequency regime ( $10$ – $10^5$  Hz) has been investigated at ESPCI by measuring the magnetization (or susceptibility) using a Faraday rotation method.<sup>7,12</sup> The low-frequency regime ( $10^{-2}$ – $10$  Hz) has been investigated at UCLA using a SQUID magnetometer.<sup>13</sup> For purely technical reasons, the zero-field experiments have been performed over a larger frequency range ( $10^{-2}$ – $10^5$  Hz) than experiments in the presence of a field ( $1$ – $10^4$  Hz). Faraday rotation requires a platelet-shaped sample. Therefore, both sets of experiments were performed with the same magnetic configuration: ac and dc fields applied perpendicular to the platelet. A set of low-frequency experiments ( $1$ – $10$  Hz)

has also been performed using the SQUID with the ac and dc fields applied parallel to the plane of the platelet in order to probe the effects of platelet geometry.

Experiments were performed at 10 Hz with both techniques to give an overlap measurement region.

### C. Contribution of the demagnetizing field

If the demagnetizing field  $H_D$  is non-negligible compared to the applied field  $H_A$ , our experimental results will be affected by the sample geometry. This appears to be a major problem in our sample in three respects.

- (1) Value of the internal field  $H$ .
- (2) Homogeneity of the internal field because of finite sample size.
- (3) Value of the phase angle  $\phi$ .

We take each in order.

#### 1. Value of internal field

We first approximate our platelet-shaped sample by an ellipsoid of revolution. We can then write

$$H = H_A - N_{\perp} M, \quad (15)$$

where  $N_{\perp}$  is the demagnetizing factor when  $H_A$  is applied perpendicular to the platelet, and  $M$  is the platelet magnetization. Because  $M$  is temperature dependent in the temperature range that we investigate, the internal field  $H$  will also depend on temperature at constant applied field. This effect is quite noticeable in our case.

By measuring the magnetization of our sample in the plane of the platelet and perpendicular to the platelet, we have obtained a fairly accurate (though empirical) estimate of  $N_{\perp}$  (assuming  $2N_{\parallel} + N_{\perp} = 4\pi$ ). We find  $N_{\perp} = (2.4 \pm 0.04)\pi$ .

In order to obtain a quantitative value of the internal field, we also need the absolute value per unit volume of the magnetization (or susceptibility) in low field. We measured  $\chi_{\perp} = (0.13 \pm 0.03)$  cgs units per  $\text{cm}^3$  at 1.8 K. The importance of the demagnetizing field may be exemplified by the following estimates, which rely on the values for  $N_{\perp}$  and  $\chi_{\perp}$  quoted above.

First, we calculate the internal field  $H$  at 1.8 K using Eq. (15). We find

$$H = (3_{-1}^{+2}) \times 10^{-2} H_A,$$

which means that the absolute value of the internal field  $H$  is not accurately known. However, the absolute value will not turn out to be relevant for the determination of the critical exponents (see below).

Second, we calculate the relative *change* of the internal field when the temperature is lowered from 2 to 1.55 K. We find

$$H(1.55 \text{ K})/H(2 \text{ K}) = 0.32_{-0.17}^{+0.22}.$$

These error limits are very serious because the relative location of the low temperature [i.e., low-frequency  $T_f(H, \omega)$  lines] may not be correctly determined. This introduces a systematic error for the determination of the

critical exponents. All of the scalings to be described in Sec. IV have been obtained at the mean and extreme values of the internal field. As indicated above, the effect of the statistical error on the scaling exponents is at least four times smaller than the systematic deviation introduced by the uncertainty in the internal field. As we shall show, while the scaling in zero field and the scaling for finite  $T_c$  are affected little, the exponent  $\Delta$  for field scaling at  $T_c = 0$  is significantly shifted when taking into account the variations in the internal field.

In summary, all static measurement values will henceforward refer to the internal field, having taken into account the mean correction quoted above. The error bars that will be specified for the scaling exponents will not correspond to a standard deviation derived from a best-fit procedure, but rather to the systematic error introduced by the computation of the internal field associated with a given applied field.

#### 2. Homogeneity of internal field

Such a large sensitivity of the internal field to the value of  $N_{\perp}$  raises another question. Is the internal field homogeneous over a sample whose shape is not quite an ellipsoid of revolution? We explain below how we deal with this question.

Referring to the literature on nonellipsoidal bodies,<sup>14</sup> we can obtain the variation of the internal field introduced by a spatial variation of the demagnetizing factor. The variation of  $N_{\perp}$  along the axis of the platelet may be neglected. Defining the mean radius of the platelet as  $r_0$ , we can estimate from Ref. 14 the spatial variation of  $N_{\perp}$  along the radius  $r$ . As long as  $r/r_0 < 0.7$ ,  $N_{\perp}(r)$  is reasonably constant. However, for  $r/r_0 \geq 0.7$ ,  $N_{\perp}(r)$  drops rapidly and reaches, for example,  $N_{\perp}(0.9r_0) = 0.88N_{\perp}(0)$ . This induces a large *increase* of the internal field from the center to the edge. A rough estimate yields  $H(0.9r_0)/H(0) = 7.4$ .

Therefore, a measurement which averages the magnetization response to an applied field over the whole sample is associated with a large inhomogeneity in the internal field. This is the case for the measurements performed with the SQUID. However, when measuring the Faraday rotation, we do not probe the sample's magnetization in the edges. The light beam is focused so as to illuminate only the central part of the sample ( $r < 0.8r_0$ ) where the internal field is roughly constant. The comparison of the experimental curves  $\phi(T, \omega, H)$  is consistent with this interpretation. In zero field, the curves obtained by the optical measurement map onto SQUID data quite satisfactorily. In the presence of the same external magnetic field, the curves are quite different. The SQUID data should be associated with a larger internal field because the edges of the sample represent a non-negligible volume and experience a smaller demagnetizing field, i.e., a larger internal field.

More quantitatively, we have attempted to reconstruct the SQUID results by choosing an internal field distribution appropriate to the geometry of our sample, using the Faraday rotation  $\phi(T, \omega, H)$  curves for specific internal fields. We selected five delta functions corresponding to

five selected values of  $N_{\perp}$  decreasing gradually from 0.6 ( $r \leq 0.825r_0$ ) to 0.48 ( $r = r_0$ ). These delta functions were weighted in a manner so as to represent reasonably well the spatial variation of  $N_{\perp}$  according to Ref. 14. We then used the curves for  $\phi(T, \omega, H)$  as measured by the Faraday rotation at these five values of field, and generated the  $\phi(T, \omega, H)$  curve one would have "expected" to have seen from the SQUID measurements. The resulting curve was quite close to that actually measured with the SQUID at an external field of 13 Oe. The external Faraday rotation fields, when weighted according to the above prescription, averaged to 36 Oe. Therefore, in order to compare the SQUID data with the Faraday rotation data, one should multiply the field applied to the SQUID by a factor of 36 Oe/13 Oe = 2.8. We have found that such a field "recalibration" actually works quite well. For example, the  $T_f(H, \omega)$  lines derived from both sets of experiments fall on top of one another with this field scaling. We shall henceforward rely on the optical (Faraday rotation) measurement to define the proper internal fields. The 1- and 10-Hz SQUID experiments in the presence of an external field will therefore be calibrated with respect to the 10-Hz optical measurements.

### 3. Value of the phase angle

We show now that the geometry also modifies the response of our system. One can visualize the effect of the demagnetizing field on the measurement of  $\phi$  by writing the equation of motion for  $M$  including the demagnetizing field:

$$\tau \frac{dM}{dt} + M = \chi(H_A - N_{\perp}M). \quad (16)$$

Solving Eq. (16) yields immediately

$$\phi = \omega\tau / (1 + N_{\perp}\chi) = \omega\tau(1 - N_{\perp}\chi_{\perp}) \equiv \omega\tau^*. \quad (17)$$

Our analysis therefore actually generates an apparent relaxation time  $\tau^*$ , which is the product of an intrinsic time  $\tau$ , multiplied by  $1 - N_{\perp}\chi_{\perp}$ , a geometry- and temperature-dependent factor. We expect, therefore, when measuring  $\phi$  in two different geometries ( $\phi_{\parallel}$  with  $H_A$  and  $\Delta H_A$  parallel to the plane of the platelet, and  $\phi_{\perp}$  with  $H_A$  and  $\Delta H_A$  perpendicular to the plane), the ratio

$$\frac{\phi_{\parallel}}{\phi_{\perp}} = \frac{1 - N_{\parallel}\chi_{\parallel}}{1 - N_{\perp}\chi_{\perp}} = \frac{\chi_{\parallel}}{\chi_{\perp}}. \quad (18)$$

In fact, Eq. (18) is remarkably well satisfied experimentally both in zero-field and in applied magnetic field measurements.

Following the procedures outlined in Sec. III A [setting  $\phi_{\perp}$  to a constant value in order to define  $T_f(H, \omega)$ ], we define a line associated with a constant  $\tau^* = \tau(1 - N_{\perp}\chi_{\perp})$ . The value of  $\phi_{\perp}$  must therefore be corrected by taking into account the multiplicative factor  $(1 - N_{\perp}\chi_{\perp})$  in order to restore a constant intrinsic relaxation-time contour. This introduces some additional uncertainty on the  $T_f(H, \omega)$  lines, which may be viewed as another systematic error on the value of  $H$ . This error has been estimated and is smaller than the major error discussed above associated with the computation of the internal field.

The following are the conclusions of this section. A defensible criterion has been defined and discussed which provides  $T_f(H, \omega)$  lines associated with specific response times. Demagnetizing field effects have been fully taken into account. They introduce a systematic error into the data analysis when a field is present which will be evaluated in the next section. They make necessary a calibration of the SQUID measurements as compared to the Faraday rotation measurements, and they require a further correction for the phase angle  $\phi$ . This will also be evaluated for the data presented in the next section. In particular, once the apparent values estimated for the characteristic times have been derived from the best fits, intrinsic values are deduced by multiplying (at 1.6 K) the apparent values by  $1 - N_{\perp}\chi_{\perp} \approx 2 \times 10^{-2}$ .

## IV. EXPERIMENTAL SCALINGS

### A. Check in zero field

Following the analysis that we have described in detail in the previous section, we report in Table I freezing temperatures as determined using two different values for  $\phi$  criteria. We plot  $T_f^{-z\nu}(\omega)$  as a function of  $\log_{10}\omega$  in Fig. 2 and look for the value of  $z\nu$  which gives rise to two parallel straight lines. We find  $z\nu = 8 \pm 0.5$ . This is twice the value suggested by Binder and Young.<sup>4</sup> Using Eq. (1), we have also determined the microscopic time  $\tau_0$ :

$$\tau_0 \sim 10^{-5} \text{ s}, \quad T_c = 0.$$

Referring to the model of Binder and Young,<sup>4</sup> one can argue that in the expression  $\ln(\tau/\tau_0) \sim \Delta F(\xi_{EA})/T$  the potential barriers  $\Delta F$  may be distributed, but that  $\tau_0$  has a unique value which is the shortest response time available to the system. We then note that in Ref. 15 a decay time as short as  $10^{-6}$  s (an apparent time of 25 ns) has been observed. A measured response time shorter than  $\tau_0$ , obtained for the  $T_c = 0$  model, is a serious inconsistency.

For the alternate scaling assumption,  $T_c \neq 0$ ,  $T_f - T_c$  is plotted versus  $\omega$  in Fig. 3 on a log-log scale. Two parallel straight lines are obtained by choosing  $T_c = 1.50 \pm 0.02$  K, very close to the observed temperature for the maximum of the d.c. magnetization. Using Eq. (6), the exponent  $z\nu$  is

$$z\nu = 7.2 \pm 0.5.$$

We can also determine a characteristic time  $\tau_0$  in this case. We find

TABLE I. Zero-field freezing temperatures defined by two different criteria  $\phi_1 = 0.08$  deg,  $\phi_2 = 0.04$  deg ( $\phi_x \geq 0.15$  deg). The error bar for  $T_c$  is estimated to be  $\sim 0.01$  K.

| $\nu$ (Hz) | $\phi_1$ | $\phi_2$ |
|------------|----------|----------|
| $10^5$     | 2.0      | 2.065    |
| $10^4$     | 1.86     | 1.890    |
| $10^3$     | 1.76     | 1.780    |
| $10^2$     | 1.695    | 1.705    |
| 10         | 1.645    | 1.655    |
| 1          | 1.605    | 1.612    |
| $10^{-2}$  | 1.550    | 1.560    |

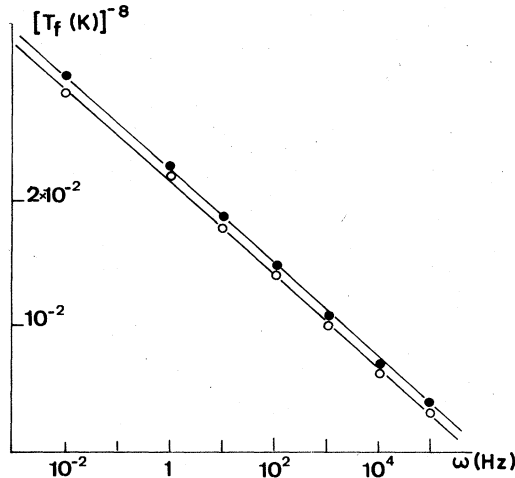


FIG. 2. Check of the frequency dependence of the zero-field freezing temperature  $T_f$  in the model  $T_c=0$ . The best fit is achieved for  $T_f^{-8}$  vs  $\log_{10}\omega$ . The two straight lines correspond to the two criteria:  $\bullet$ ,  $\phi=0.08$  deg;  $\circ$ ,  $\phi=0.04$  deg.

$$\tau_0 \sim 2 \times 10^{-7} \text{ s}, \quad T_c \neq 0.$$

This is still a large value compared to  $k_B T_c / \hbar$ , but is two orders of magnitude smaller than for  $T_c=0$ , and it is now shorter than the relaxation time found in Ref. 15.

The zero-field scalings thus provide critical exponents for both values of  $T_c$ . The value of the exponents, as can be seen through the error bars, are not very sensitive to the geometric corrections on  $\phi$ .

### B. Scalings of $T_f(H, \omega)$ lines

We now report the scalings analysis appropriate to both values of  $T_c$  in the presence of a field. In each case, the only adjustable parameter is  $\Delta$ . Figures 4 and 5 display the best fits, with parameters

$$\Delta = 10.5 \pm 2.5, \quad T_c = 0 \text{ K};$$

$$\Delta = 2 \pm 0.2, \quad T_c = 1.5 \text{ K}.$$

As already stressed, the exponent  $\Delta$  associated with  $T_c=0$  is more sensitive to the magnitude of the internal

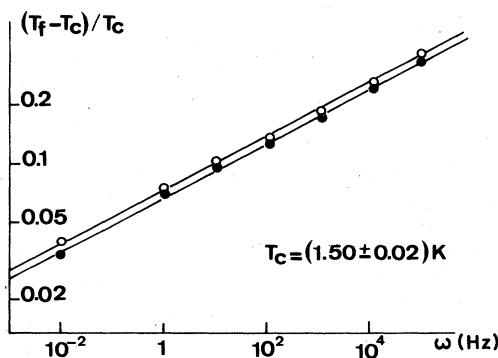


FIG. 3. Check of the frequency dependence of the zero-field freezing temperature  $T_f$  for a  $T_c$ . A best fit to two parallel straight lines corresponding to the two criteria:  $\bullet$ ,  $\phi=0.08$  deg, and  $\circ$ ,  $\phi=0.04$  deg, is achieved for  $T_c=1.5$  K.

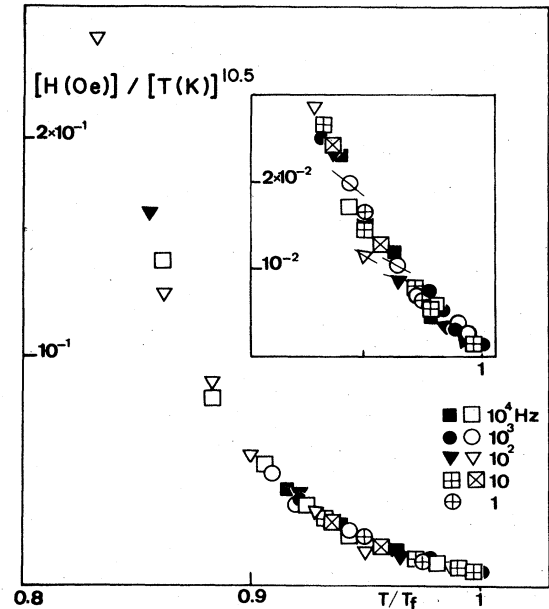


FIG. 4. Scaling of field-temperature  $T_f(H, \omega)$  lines for the case  $T_c=0$ . The inset shows the low-field points with an extended scale. A typical experimental error bar is indicated on a few points.

field. However, the value of  $\Delta$  extracted by Binder and Young ( $\sim 7$ ) may be ruled out. Also, the residual scattering of the points in both cases is entirely consistent with the experimental errors. The value for  $\Delta$  obtained under the assumption of a finite  $T_c$ ,  $2 \pm 0.2$ , is outside the error bars from the value of  $\frac{3}{2}$  expected for the de Almeida- Thouless (AT) line.<sup>16</sup> It is conceivable that this difference derives from the fact that  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$  is a short-range spin-glass, and that the value  $\Delta = \frac{3}{2}$  has been derived for an infinite-range model.<sup>16</sup>

This conclusion ( $\Delta = 2 \pm 0.2$ ) differs from that presented in Ref. 12 using the same raw data (though for a more restricted frequency range:  $10^2$ – $10^5$  Hz). As we have already remarked in Sec. III A, Ref. 12 utilized a different criterion ( $\chi'' = \text{const}$ ) than that adopted in this paper.

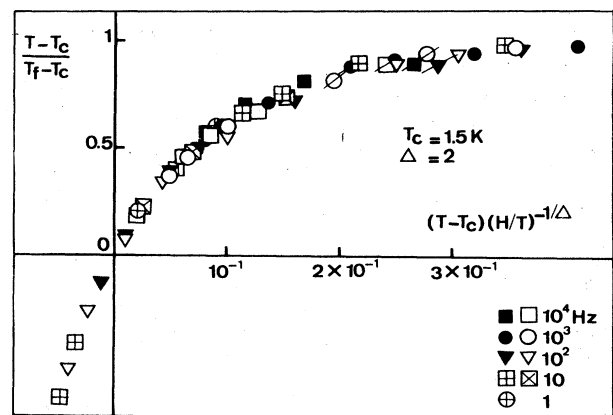


FIG. 5. Scaling of the field-temperature  $T_f(H, \omega)$  lines for the case  $T_c=1.5$  K. Typical experimental error bars are indicated on a few points.

Simply speaking, the method of Ref. 12 did not ensure that  $\tau$  was constant along the  $(H, T)$  line.

Another question arises when one considers the finite- $T_c$  case. The order-parameter susceptibility diverges in the infinite-range Sherrington-Kirkpatrick spin-glass model along the de Almeida—Thouless line.<sup>17</sup> This would imply the divergence of the response time as one approached the transition line. This means that, as  $\omega \rightarrow 0$  (or  $\tau \rightarrow \infty$ ) and  $T_f \rightarrow T_c$ , the field should remain finite. The universal function, defined by the experimental points in Fig. 5, should therefore exhibit asymptotic behavior for a given value of  $(T - T_c)/(H/T_c)^{-1/\Delta}$ . Unfortunately, too few experimental points are available to draw any definitive conclusion. This will be the subject of a future investigation.

The relevance of the AT solution to  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ , in the face of an isotropic exchange interaction where one would normally expect a Gabay-Toulouse transition,<sup>18</sup> derives from the large (0.4 K) local anisotropy energy in this spin-glass. As stated by Campbell *et al.*,<sup>19</sup> "...Kotliar and Sompolinsky [to be published]... predict Ising-like behavior (with an AT-like irreversibility temperature dropping as  $H^{2/3}$ ) below a threshold field related to the anisotropy strength, and a Heisenberg-like behavior (with a GT-like irreversibility temperature) above the threshold

field." All of our measurements have been in very small field ( $\approx 10$  Oe) relative to the local anisotropy field ( $\approx 2500$  Oe). Hence, our reference to the AT transition line should be relevant to  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$ .

## V. CONCLUSION

We have shown that on the basis of scaling, our experimental data for (1) zero-field freezing temperature and (2)  $T_f(H, \omega)$  lines are consistent with both the logarithmic scaling suggested for  $T_c = 0$ , and for the more standard scaling expected for finite  $T_c$ . We note that both the minimum relaxation time  $\tau_0$ , and the comparability of our  $T_c$  extracted from the  $T_f(H, \omega)$  lines ( $T_c = 1.5$  K) with what is already known for the  $\text{Eu}_{0.4}\text{Sr}_{0.6}\text{S}$  system, suggests that  $T_c \neq 0$  may be more applicable.

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