Nonlinear susceptibility and relaxation in the *XY* spin glass *Y*Tb

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Measurements of the nonlinear susceptibility, irreversibility onset, and relaxation of the remanent magnetization are reported for a single-crystal sample of $Y\mathrm{Tb}_{3\ \mathrm{at.}\,\%}$. The data give strong support for the existence of a spin-glass phase transition for spin components in the basal plane, but not for those along the c axis. A scaling analysis of the nonlinear susceptibility leads to the values $\delta=3.2\pm0.2$ and $\phi=3.0\pm0.2$ for the field and cross-over exponents, respectively. These values are consistent with the position of the de Almeida–Thouless line observed below the freezing temperature.

I. INTRODUCTION

Recent numerical simulations of long-range spin glasses in three dimensions suggest that Ising systems undergo a phase transition,[1] while Heisenberg systems do not.[2] It is of considerable importance to examine, therefore, the intermediate case of *XY* spins. In this paper we report a detailed study of the *XY* spin-glass $Y\mathrm{Tb}_{3\ \mathrm{at.}\,\%}$ using dc measurements of the nonlinear susceptibility, the onset of irreversibility, and the relaxation of the thermal remanent magnetization. As the property most closely related to the spin-glass order parameter,[3] the critical behavior of the nonlinear susceptibility must be regarded as crucial evidence for a spin-glass phase transition. The results presented here confirm that such a transition occurs for spin components in the basal plane, but not for components along the hexagonal axis.

Pioneering work by Sarkissian and Coles[4] accurately mapped the phase diagram of the Y-Tb system along with those of other rare-earth—transition-metal alloys. More recently, Fert and co-workers[5] extended this work to single-crystal samples and examined the spin-glass phase diagram as a function of the single-ion anisotropy constant D. For *Sc*Tb, which has a negative value of D, they found that irreversibility sets in at the same temperature when the applied field is along the hexagonal c axis as when the field is in the basal plane. From this evidence, they concluded that spin-glass freezing of both c-axis and basal-plane spin components occurs simultaneously, in disagreement with the phase diagram proposed by Roberts and Bray,[6] and by Cragg and Sherrington.[7] We have shown recently,[8] however, that $Y\mathrm{Tb}_{5\ \mathrm{at.}\,\%}$, which exhibits irreversible properties similar to $Y\mathrm{Tb}_{3\ \mathrm{at.}\,\%}$ and *Sc*Tb, orders in a spiral phase at 27 K. The presence of long-range order for the compound with 5 at. % Tb, but not for that with 3 at. %, is consistent with the phase diagram of Sarkissian and Coles, and suggests that irreversibility alone is not a good measure of a spin-glass transition. The present study addresses this problem in detail for $Y\mathrm{Tb}_{3\ \mathrm{at.}\,\%}$.

II. EXPERIMENTAL PROCEDURE

Samples of *Y*Tb were prepared by repeatedly arc-melting 10-g buttons of the constituents. The ingots were annealed for 24 h at 1350 °C in a vacuum furnace. Large grains, produced by the annealing process, were cut from the ingot using a low-speed diamond saw. The crystals were then electropolished for x-ray diffraction. For magnetization studies, oriented samples were cut in the form of bars approximately 6 mm in length and weighing ~ 60 mg. As noted above, neutron scattering measurements[8,9] on the same crystals revealed the presence of long-range spiral order for $Y\mathrm{Tb}_{5\ \mathrm{at.}\,\%}$ but only diffuse magnetic scattering down to 9 K for $Y\mathrm{Tb}_{3\ \mathrm{at.}\,\%}$.

All magnetic measurements were performed on commercial superconducting quantum interference device magnetometers.[10] Nonlinear susceptibility data were taken by cooling the sample from 36 K ($\sim 2T_g$) to 9 K in a field H and then measuring the magnetization $M(H)$ while heating. When the field was oriented along the hexagonal c-axis, the sample tended to rotate since, near T_g, the basal-plane susceptibility is approximately twenty times larger than the c-axis susceptibility. This rotation was countered by the use of a massive plastic sample holder much longer than the spacing between the magnetometer coils.[11] To determine the onset of irreversibility, samples were cooled to 5 K in zero field [zero-field cooling (ZFC)], the field was applied, and $M_{\mathrm{ZFC}}(H)$ measured to $\sim 2T_g$. The temperature was then stepped downward and $M_{\mathrm{FC}}(H)$ measured in the same applied field [field cooling (FC)]. Relaxation studies were performed with H in both the basal plane and along the c axis. The sample was cooled from 25 K in a magnetic field. The field was then decreased to zero and the decay of the remanent magnetization followed for ~ 1500 s. For the c-axis relaxation measurements, no attempt was made to prevent the establishment of a basal-plane component of the remanent magnetization due to sample rotation. Since the actual measurement was performed in zero applied field, only the c component of the decaying remanent was recorded.

III. NONLINEAR SUSCEPTIBILITY

Magnetization curves for $YTb_{3\,\mathrm{at.\,\%}}$ for magnetic fields up to 34 kOe in the basal plane and along the c axis are shown in Fig. 1 (upper and lower parts, respectively). Considerable nonlinearity is evident at all temperatures for fields in the basal plane; none could be detected in the c direction. Because nonlinearity of the magnetization is the property most closely associated with the spin-glass order parameter,[3,12,13] this result suggests that a spin-glass transition occurs only for spin components in the basal plane.

The nonlinearity in the basal plane is more readily observed in plots of M/H versus temperature, as shown in Fig. 2. Extraction of the nonlinear part of the susceptibility, defined as

$$\chi_{\mathrm{NL}}=\chi_0-M(H,T)/H \;, \tag{1}$$

requires precise knowledge of $\chi_0 \equiv \lim_{H\to 0}(M/H)$, the determination of which is the main experimental challenge. To achieve the required precision (tenths of a percent of χ_0), we use the sample itself to calibrate the magnetometer at low fields, as has been done previously.[14] The magnetization is measured in a nominal field of 40 Oe. The actual field value is chosen so that χ_{NL} vanishes at 40 K ($\sim 2.5T_g$), as Fig. 1 suggests to be true. The adjusted field, approximately 2 Oe lower than that determined by using a Pt standard, incorporates differences in the high- and low-field ranges of the magnetometer and corrections[15] due ot the size and shape of the YTb sample. The inverse of χ_0 thus determined is shown in Fig. 3.

The nonlinear susceptibility determined from (1) using the data of Fig. 3 is shown in Fig. 4. At all fields, it exhibits a maximum near 16 K, which we take to be the freezing temperature T_g. This point lies at a slightly higher temperature than the peak in the low-field susceptibility and below that at which irreversibility can be noted. The rounding of χ_{NL} in the vicinity of T_g reflects the rounding of χ_0. In the scaling analysis below, data at 17 and 16.4 K deviate from the remainder of the data at low fields. The range over which the data can be scaled is improved by using the extrapolation of χ_0 shown by the dashed line in Fig. 3. In previous scaling studies the susceptibility $\chi_0(T)$ was treated as a function to be determined via the scaling process. Here we extrapolate only in the critical region.

The scaling behavior of the nonlinear susceptibility has been examined in some detail for CuMn and amorphous Gd-Al.[13] While supporting the scaling hypothesis, the analysis gives widely varying values of the exponents. The scaling approach predicts that

$$\chi_{\mathrm{NL}}(H,T)/H^{2/\delta}=f(t^{\phi/2}/H) \;, \tag{2}$$

where $t\equiv(T/T_g-1)$ and δ and ϕ are exponents. The scaling function $f(x)$ is constant for small values of x and decreases as $x^{-2+2/\delta}$ for large x. The latter guarantees that χ_{NL} varies as $H^2t^{-\gamma}$ for small fields. Although mean-field theory predicts $\delta=\phi=2$, it has recently been suggested that $\delta\sim 3$ may be more appropriate for three-dimensional systems.[16]

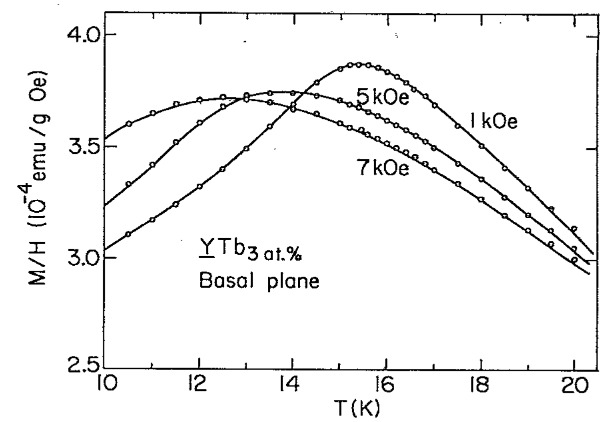

FIG. 2. M/H in the basal plane at several applied fields.

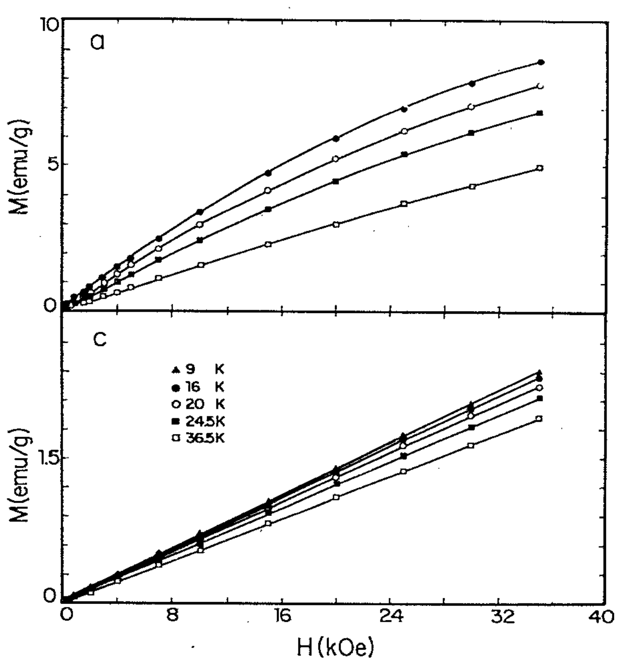

FIG. 1. Magnetization M vs field H. Upper plot: H along the a axis; lower plot: H along the c axis. Nonlinearity is evident for H in the basal plane.

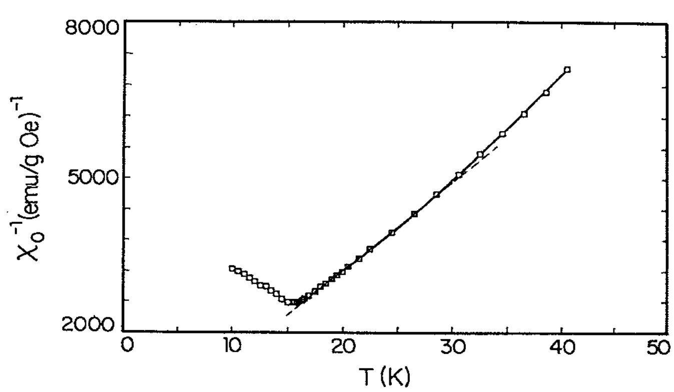

FIG. 3. Low-field values of M/H used for χ_0 in Eq. (1).

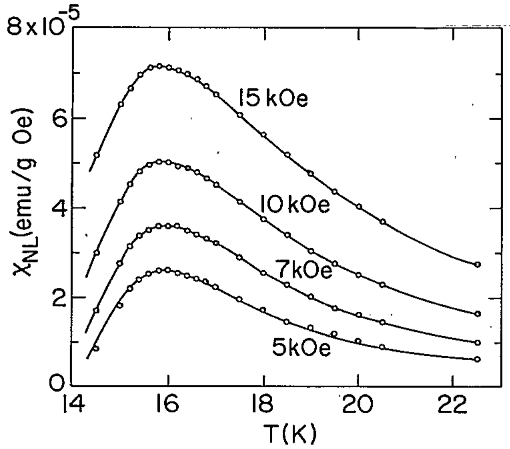

FIG. 4. Nonlinear susceptibility determined from the data of Figs. 2 and 3 according to Eq. (1).

In Fig. 5 we show the χ_{NL} data as in Fig. 4 scaled according to Eq. (2). The parameters T_g, δ, and ϕ were varied to give the best qualitative collapsing of the data to a single curve. Fields between 1 and 40 kOe are included in each isotherm. Satisfactory results could be obtained within the range $T_g=16.0\pm0.2$ K, $\delta=3.2\pm0.2$, and $\phi=3.0\pm0.2$. For the isotherms closest to T_g (16.4 and 17 K) it was necessary to use the extrapolated χ_0 in Fig. 3; there is no correction for other isotherms. Error bars on selected points are estimates of the uncertainty in ascribing an absolute value to χ_0.

The present value of ϕ is identical to that obtained for amorphous Gd-Al, but δ is much smaller. Using the scaling law $\beta=\phi/\delta$ we find $\beta=0.95$, close to values for *Cu*Mn and amorphous Gd-Al. The form of the crossover line $H\sim t^{1.5}$ is, as we show below, satisfyingly close to that of the irreversibility (de Almeida—Thouless) line below T_g. The lowest-field data points for 16.4 and 17 K do not scale properly. This suggests a crossover to a different scaling regime in the region close to T_g. The line in Fig. 5 shows the limiting slope of the scaling function if χ_{NL} is to vary as H^2 for large fields.

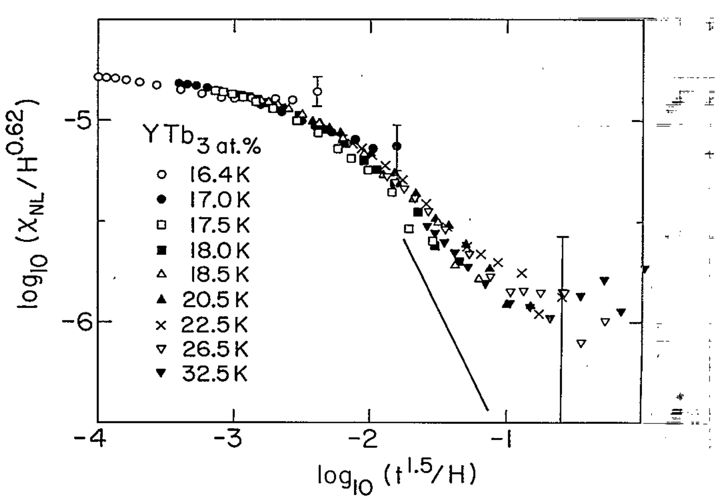

FIG. 5. Scaling plot of the nonlinear susceptibility. Each isotherm includes data from 14 values of the applied field from 1 to 40 kOe. The error bars reflect uncertainty in the determination of χ_0, and are less than 0.5% of the measured M/H.

IV. RELAXATION AND IRREVERSIBILITY

The onset of irreversibility is most conveniently located by the branch-point method.[17] Figure 6 shows that $M_{ZFC}(T)$ and $M_{FC}(T)$ differ below the branch-point temperature $t_c(H)\equiv1-T_c(H)/T_g$. The inset to Fig. 6 shows that the branch points are consistent with de Almeida—Thouless behavior [$H\approx(53\text{ kOe})t_c^{2/3}$] and with the crossover line above T_g, with $T_g=16$ K. At still lower fields, however, the branch point moves *above* the peak in M/H, as seen in Fig. 7. We believe this manifests a change from strong to weak anisotropy behavior within the basal plane.[18] In large fields, the system behaves as a Heisenberg spin glass along a Gabay-Toulouse critical line. However, in low fields, Ising-like behavior sets in and persists to a slightly higher freezing temperature. Note the appearance of irreversibility for fields along the c axis, Fig. 7 (lower part), similar to that reported for *Sc*Tb. In view of the absence of nonlinear effects for fields along the c direction, we will argue below that this reflects basal-plane freezing, rather than indicating the existence of a spin-glass order parameter in this direction.

Similar effects are observed in the decay of the remanent magnetization. For $YTb_{3\text{ at.\%}}$ the remanent magnetization follows the law

$$M_r(\tau)=M_r(0)-S\ln(\tau/\tau_0) \tag{3}$$

for up to 4000 s after the removal of the field; τ_0 is the earliest time (usually $\sim$500 s) at which data can be taken.

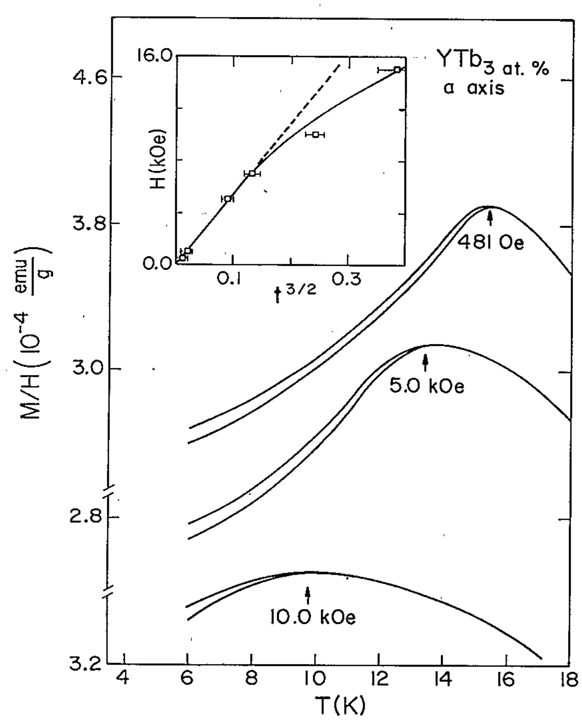

FIG. 6. Determination of the point at which field-cooled (upper) branches and zero-field-cooled (lower) branches join. The inset shows the locus of these points on a plot of H vs $t^{3/2}\equiv(1-T/T_g)^{3/2}$. The dashed line is a de Almeida—Thouless line with a characteristic field of 53 kOe.

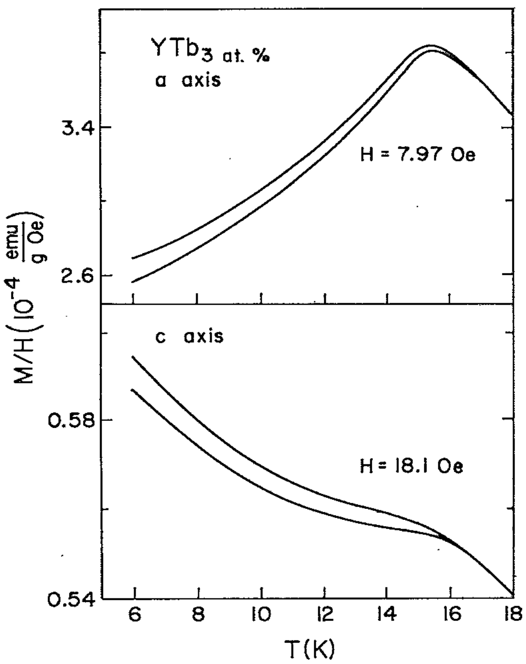

FIG. 7. Low-field susceptibility (upper part) along the a axis and (lower part) along the c axis as in Fig. 6. Although the irreversibility is much smaller (as a fraction of the susceptibility) along the c axis, it cannot be ignored. Note that irreversibility appears near 17 K for both directions.

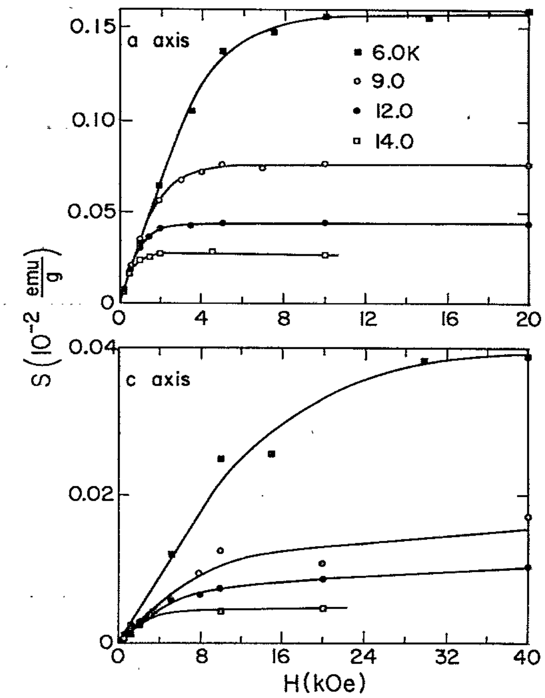

FIG. 8. Amplitude of the decaying portion of the thermal remanent magnetization as a function of the cooling field and temperature. The plateau in the a-axis data indicates that the sample was cooled in fields above the crossover field; its absence in the c-axis direction suggests that a de Almeida—Thouless line is absent.

Figure 8 (upper part) shows the results for cooling fields in the basal plane. The plateau is characteristic[19] of fields well above the irreversibility line and the decrease, of those well below. Estimating the fields at which S levels off, we find them to follow the usual $t^{3/2}$ law, but with a smaller characteristic field than that deduced from the branch point, Fig. 6.

There is also a decay of remanence when the sample is cooled in a field along the c axis, as seen in Fig. 8 (lower part). No attempt was made to restrict sample rotation, and it is possible that the remanence is established with a basal-plane component. However, only the decay of the c axis component is measured. The observed decay is two orders of magnitude too large to be explained by sample misalignment. Clearly the c axis relaxation is qualitatively different from that in the basal plane. There is no clear evidence for a plateau, even to fields double those used in the basal-plane study, nor is there a common low-field behavior at all temperatures. We take this to be further evidence against independent ordering of the spin components along the c axis.

V. DISCUSSION

The nonlinear susceptibility results presented above give clear evidence for a spin-glass transition in $YTb_{3\ \text{at.}\ \%}$ for spin components in the basal plane. The freezing temperature $T_g = 16 \pm 0.2$ K is slightly (≤ 0.5 K) above the peak in the low-field susceptibility, but coincides with the maximum in χ_{NL}. As in amorphous Gd-Al, the crossover exponent $\phi = 3.0 \pm 0.2$ found here is in good agreement with $\frac{2}{3}$-power law obtained from the onset of irreversibility. Unlike the amorphous Gd-Al case, however, the value $\delta = 3.2 \pm 0.2$ found here is consistent with β close to unity, as is the case in other spin glasses.

The branch point measurement gives a characteristic field of 53 kOe, much larger than comparable values in *Ag*Mn.[20] Similarly, nonlinear effects are observable only in rather large fields compared with *Cu*Mn.[12–14] For Tb atoms, the estimated characteristic field,[18] $H_c \equiv \sqrt{2} k_B T_g / \mu \approx 35$ kOe, is quite close to that observed. We conclude that the *Y*Tb system better approximates a single-spin, mean-field-like spin glass than does *Cu*Mn.

Two unusual points remain: the c-axis data show irreversibility without direct evidence for spin-glass ordering from the nonlinear susceptibility, and low-field irreversibility sets in above the apparent T_g, both in the basal plane and along the c axis. Omitted from the discussion of Ref. 5 is the random anisotropy arising from spin-spin interactions. It is this source of anisotropy that gives rise to many of the effects most commonly associated with the spin-glass state. Upon the establishment of spin-glass order, a memory of spin directions is established that is independent of the eventual direction of the magnetization. In the presence of a field, the orientation of this "anisotropy triad"[21] may be moved via the torque exerted on it by the magnetization. For an easy-plane magnet such as *Y*Tb, cooling the sample with a magnetic field applied along the c axis results in the establishment of anisotropy triads whose axes do not lie, necessarily, in the basal plane. Consequently, when the field is removed, the magnetization cannot immediately relax to the basal plane under the influence of single-ion anisotropy, but must remain partially aligned with the anisotropy triad. The c-axis remanence and its decay must, in our view, be associated with the relaxation of the anisotropy triad, oriented during field cooling, into the basal plane.

The same anisotropic spin-spin interactions cause a cross over form XY (high-field) to Ising (low-field) behavior. In the Heisenberg-to-Ising case, the critical behavior occurs along a Gabay-Toulouse-like line at high fields and along a de Almeida—Thouless line at low fields.[18] The temperatures to which the lines extrapolate for vanishing fields differ slightly. We expect the same situation to hold in the XY case, and suspect that the branch-point line above 16 K represents the phase-transition line, rather than the irreversibility crossover line as below 16 K. To resolve these questions requires detailed study of irreversibility and transverse freezing within the basal plane. Such measurements are currently underway.

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