Spin-glass transition in amorphous Tb-Si films

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A recent study established that disordered Tb exhibits a spin-glass transition with a spin-glass freezing temperature ranging from 183 to 53 K, the lowest temperature corresponding to the greatest degree of atomic disorder. This result is confirmed by the present study of amorphous Tb-Si alloys which demonstrates that at low Si content the spin-glass properties of $Tb_{90}Si_{10}$ are essentially identical to those of the most distorted (quasiamorphous) pure Tb film. Furthermore, the spin-glass properties of Tb-Si alloys with Si content up to 50 at. % were studied by ac and low-field dc susceptibility measurements with special emphasis on the composition Tb_5Si_3 which is ferromagnetic in the crystalline state.

I. INTRODUCTION

While crystalline Tb is a helix antiferromagnet[1] with a Néel temperature of 229 K which becomes ferromagnetic at 222 K, disordered Tb obtained by sputtering at 77 K exhibits a spin-glass transition[2] as evidenced by an ac susceptibility peak and an irreversibility in the dc susceptibility. Although irreversibilities and susceptibility peaks have been reported in crystalline solid solutions of Tb-Y (Ref. 3) and amorphous $Tb_{52}Ag_{48}$ (Ref. 4) and $Tb_{52}Au_{48}$ (Ref. 5) films, their origin is different from those reported here for reasons previously discussed.[2] The spin-glass freezing temperature (T_{SG}) ranges from 183 to 53 K, the lowest temperature corresponding to the greatest degree of atomic disorder. Since, in general it is not possible to obtain a pure single element in a totally amorphous state, even the most distorted Tb displays, besides the main susceptibility peak at 53 K, deviations from the Curie-Weiss law and irreversibility at higher temperatures which have been associated[2] with the spin-glass transition of microcrystalline inclusions with higher T_{SG}. To further test this interpretation, it was decided to investigate the magnetic properties of Tb-Si alloys because such alloys should be fully amorphous as a result of the glass-forming ability of Si and should be similar to pure distorted Tb in the limit of low Si content. Furthermore, crystalline Tb_5Si_3 is a ferromagnet with a 50-K Curie temperature,[6] and it should be interesting to investigate the magnetic properties of this compound in the amorphous state.

II. EXPERIMENTAL PROCEDURE

Most of the Tb-Si films were prepared similarly to the Tb films[1] by getter-sputtering an arc-melted Tb-Si button at 2.25 W (1500 V, 1.5 mA) onto sapphire substrates held at 77 K. In the case of Tb_5Si_3, films were sputtered at powers as high as 15 W (1500 V, 10 mA) and at temperatures ranging from 77 to 870 K. The structure of the films was determined by x-ray diffractometry. The ac susceptibility of the films was measured in an applied field of 4 Oe at 10 kHz, while the dc susceptibility was obtained in a susceptometer using a superconducting quantum-interference device (SQUID) in fields as high as 16 Oe. It is important to notice that although the samples for the ac and dc measurements were made simultaneously, two different samples were used in the two measurements. Contrary to the Tb films, the various Tb-Si films were stable in air.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Starting with the low Si alloys, Fig. 1 displays an x-ray diffractometer trace characteristic of an amorphous film and very similar to that shown in Fig. 2 of Ref. 2 for the most distorted pure Tb film: indeed, both patterns are characterized by a broad peak which rises above the diffuse background over a 2θ range of about 6° and is centered at an angle corresponding to the (002) crystalline Tb diffraction peak. As shown in Fig. 2, this similarity in structure is reflected by a similarity in magnetic properties. More specifically, the ac susceptibility has essentially the same value for the most distorted pure Tb film (Tb No. 6) and for amorphous (a) $Tb_{90}Si_{10}$; furthermore, the maxima in susceptibility which correspond to the spin freezing temperatures T_{SG} are also very close (50 K for Tb No. 6 and 58 K for $Tb_{90}Si_{10}$). The slightly higher T_{SG} for a-$Tb_{90}Si_{10}$ is consistent with the fact that T_{SG} increases with increasing Si content ($T_{SG}\simeq70$ K for a-$Tb_{62.5}Si_{37.5}$). There is, however, an important difference around 140 K between the susceptibility of a-Tb No. 6 and a-$Tb_{90}Si_{10}$. In the case of Tb No. 6 there is a relative maximum in the susceptibility at $\simeq$140 K which is better evidenced in the top of Fig. 2 as the maximum deviation from the Curie-Weiss plot. This deviation (referred to as T_{SG} crystalline) has been previously attributed[2] to the presence of microcrystals embedded in the amorphous matrix. This assumption is clearly confirmed by the susceptibility data of a-$Tb_{90}Si_{10}$ which display a perfect fit to the Curie-Weiss law in this temperature range as one would expect from a fully amorphous material. These results can be summarized by stating that except for the presence of some residual crystalline material, the magnetic properties [T_{SG}, p_{eff}, and $\chi(T)$] of amorphous Tb are essentially the same as those of a dilute amorphous Tb-Si alloy.

The study of a-Tb-Si spin glasses will now be continued by examining the effects of both higher Si content and higher deposition temperatures (T_D). The dc and ac susceptibility of an a-$Tb_{62.5}Si_{37.5}$ film deposited at 77 K shown in Fig. 3 clearly establish that such a film undergoes a spin-glass transition at $T_{SG} \simeq 70$ K, while the crystalline counterpart Tb_5Si_3 becomes ferromagnetic[6] at 50 K (this ferromagnetic transition was actually observed in the arc-melted Tb_5Si_3 button used to sputter the film). The spin-glass transition at 70 K is demonstrated by the progressive rounding off of the ac susceptibility cusp with increasing dc magnetic field and by the irreversibility displayed by the dc susceptibility. The zero-field-cooled (ZFC) dc susceptibility curve is in good agreement with the reversible ac susceptibility curve (same T_{SG} of 68 K), considering the previously mentioned fact that the susceptibilities were measured on two different samples. Although the samples were deposited simultaneously at 77 K, the dc susceptibility sample is somewhat thinner than the ac sample, and as will be shown later, the susceptibility is an increasing function of the film thickness. At any rate, these small susceptibility differences are negligible compared to the central result of Fig. 3, which is the difference between the ZFC and the field-cooled (FC) curve which reveals the typical irreversibility of spin glasses. The irreversible part of χ can be separated by removing the applied field after field cooling, which leads to the thermoremanent magnetization per Oe (TRM in Fig. 3) that can be seen to vanish at $T_{SG} \simeq 68$ K.

The effect of film thickness on the magnetic susceptibility can be appreciated from the data shown in Fig. 4 for a-$Tb_{62.5}Si_{37.5}$ No. 2, which is approximately twice as thick as a-$Tb_{62.5}Si_{37.5}$ No. 1 described in Fig. 3, which while leaving T_{SG} essentially unchanged ($\simeq 73$ K) leads to an increase in χ of about 50%. Such an effect can easily account for the small difference between the ac and dc samples analyzed in Fig. 3. The rounding off of the ac susceptibility cusp with applied dc magnetic field is shown again for film No. 2 (Fig. 4). The effect of the temperature of deposition is examined in Figs. 4–6. The susceptibility of a-$Tb_{62.5}Si_{37.5}$ No. 3 (3.8 μm) deposited at

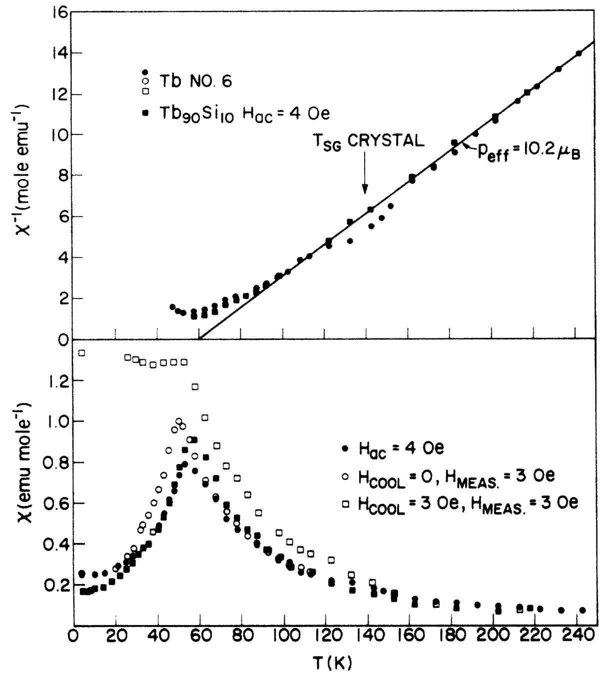

FIG. 2. The data for Tb No. 6 are identical to those shown in Fig. 3 of Ref. 2. Bottom: ac susceptibility (solid circles for Tb No. 6 and solid squares for $Tb_{90}Si_{10}$) and dc susceptibility (open symbols for Tb No. 6 only). Top: inverse ac susceptibility as a function of temperature.

300 K (Fig. 5) is essentially the same as that of No. 2 (4.9 μm) deposited at 77 K (Fig. 4), which suggests that the decrease in χ of No. 3 expected from its smaller thickness has been compensated by an increase in χ resulting from the higher T_D. Furthermore, a plot of the inverse susceptibilities of both films versus temperature is well fitted by a Curie-Weiss law with similar parameters: ferromagnetic Curie-Weiss temperatures of 90 and 95 K and effective local moments of $10.8\mu_B$ and $10.95\mu_B$ which are close to those of a-Tb and a-$Tb_{90}Si_{10}$ ($10.2\mu_B$) and to the free-ion value ($9.72\mu_B$). There is, however, an interesting differ-

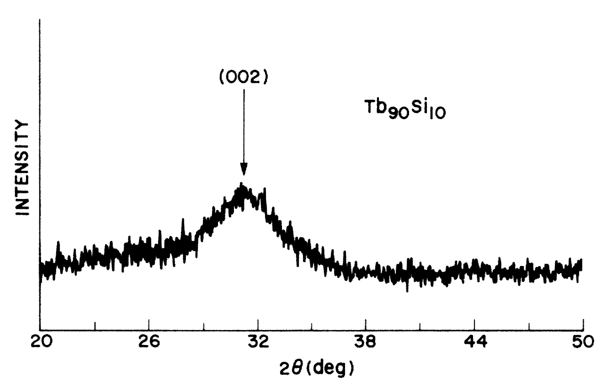

FIG. 1. X-ray diffractometer trace taken at room temperature on a 1.6-μm $Tb_{90}Si_{10}$ film deposited at 77 K. The arrow indicates the position of the (002) diffraction peak for crystalline Tb.

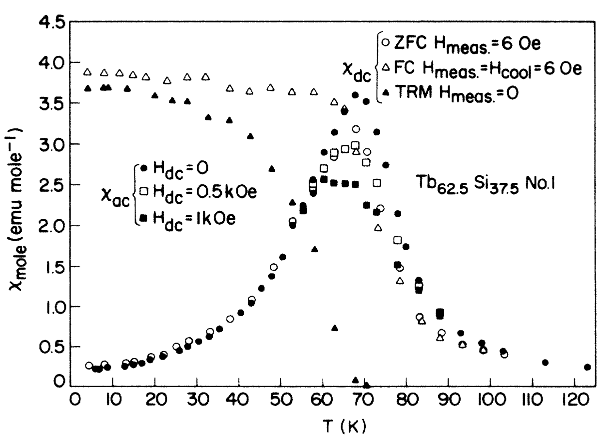

FIG. 3. Temperature dependence of the ac and dc susceptibilities of respectively, a 2.2-μm and a 1.6-μm a-$Tb_{62.5}Si_{37.5}$ film deposited at 77 K.

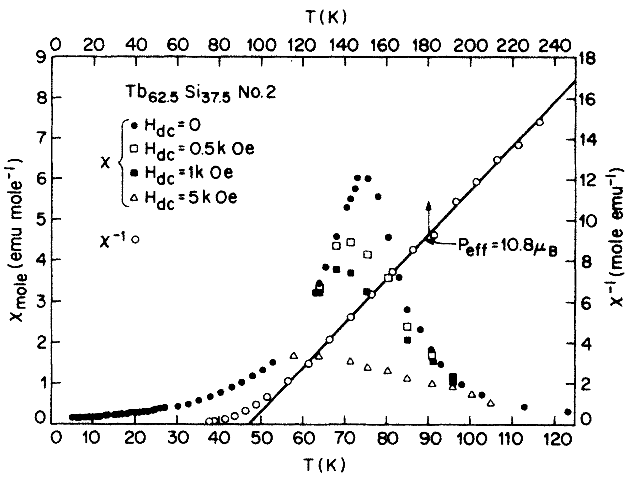

FIG. 4. Temperature and field dependence (bottom abscissas) of the ac susceptibility for a 4.9-μm a-$Tb_{62.5}Si_{37.5}$ film deposited at 77 K. The temperature dependence of the inverse susceptibility is plotted against the upper abscissa scale.

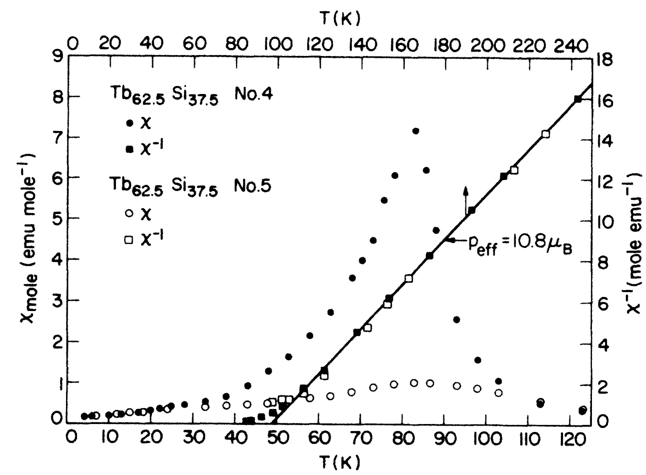

FIG. 6. Temperature dependence of the ac susceptibility (bottom abscissa) for an 11-μm a-$Tb_{62.5}Si_{37.5}$ film deposited at 370 K (No. 4) and for a 9.3-μm crystalline $Tb_{62.5}Si_{37.5}$ film deposited at 870 K (No. 5). The inverse susceptibility is plotted for both films against the upper temperature scale.

ence in the susceptibility of the two films in the vicinity of 50 K where the χ of a-$Tb_{62.5}Si_{37.5}$ No. 3 deposited at 300 K exhibits a pronounced upward deviation. Since the Curie temperature[6] of the crystalline compound Tb_5Si_3 is 50 K, it is tempting to associate this relative maximum with microcrystalline Tb_5Si_3 clusters too small to be detected by x rays which arose as a result of the higher T_D. The effect of T_D is further studied by the data shown in Fig. 6, which compares an amorphous film (No. 4) deposited at 370 K with a crystalline film (No. 5) deposited at 870 K. The susceptibility of the amorphous film (No. 4) displays again an upward deviation around 50 K and remains essentially unchanged in amorphous films deposited at temperatures as high as 700 K. At $T_D \simeq 870$ K the deposited films become crystalline, and as shown by the data for $Tb_{62.5}Si_{37.5}$ No. 5 in Fig. 6, the susceptibility decreases markedly to a value comparable to that measured on the arc-melted button and to the value previously reported[6] for Tb_5Si_3. Furthermore, at the onset of crystallization the spin-glass interaction is still observable as a broad peak centered at 83 K; this peak disappears upon full crystallization and is replaced by a ferromagnetic peak at 50 K. As shown in Fig. 6 the effective moment p_{eff} remains fixed at $10.8\mu_B$ for all deposition temperatures.

This study will now be completed by examining the magnetic properties of $Tb_{50}Si_{50}$ (Fig. 7). In comparison with the other alloys, the presence of 50 at. % Si has led to a smaller susceptibility and to a lower spin freezing temperature ($T_{SG} \simeq 35$ K). On the other hand, the fit to the Curie-Weiss law yields a similar value for p_{eff} ($10.5\mu_B$). Although not shown here, the low-field dc sus-

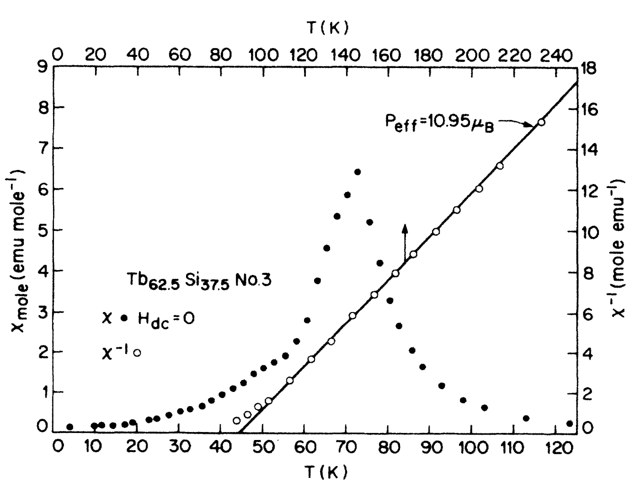

FIG. 5. Temperature dependence of the ac susceptibility (bottom abscissa) and inverse susceptibility (upper abscissa) for a 3.8-μm a-$Tb_{62.5}Si_{37.5}$ film deposited at 300 K.

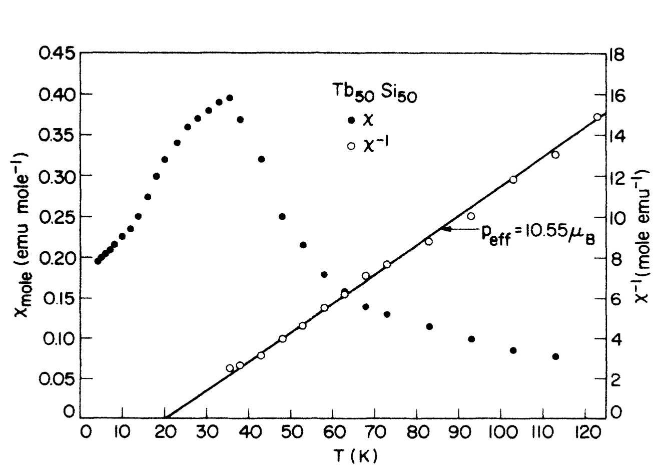

FIG. 7. Temperature dependence of the susceptibility and inverse susceptibility for a 0.8-μm a-$Tb_{50}Si_{50}$ film deposited at 77 K.

ceptibility measurement of a-$Tb_{50}Si_{50}$ is quite similar to that shown in Fig. 3 for a-$Tb_{62.5}Si_{37.5}$.

IV. SUMMARY

The spin-glass properties of a-$Tb_{90}Si_{10}$ are essentially identical to those of quasiamorphous pure Tb and confirm the previous analysis[2] of the spin-glass transition in disordered Tb. Amorphous Tb-Si alloys with Si content as high as 50 at. % display a spin-glass transition. This transition is most pronounced (highest T_{SG} and largest χ at T_{SG}) for the composition which corresponds to the crystalline ferromagnetic compound Tb_5Si_3.

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