Growth-Induced Magnetic Anisotropy in Amorphous Tb-Fe

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Coherent magnetic anisotropy is shown to be induced in vapor-deposited amorphous Tb-Fe by a thermally activated growth process. This process is hypothesized to involve rearrangement of local adatom configurations into energetically favorable orientations which minimize surface energy during the growth, a process analogous to the frequently observed crystallographic texturing of polycrystalline thin films. The anisotropy is found to be independent of the state of stress in the film during the growth, and does not depend on film thickness; these results are inconsistent with recently proposed models.

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When amorphous $\mathrm{Tb}_x\mathrm{Fe}_{100-x}$ (*a*-Tb-Fe) (and other rare-earth–transition-metal alloys) are prepared by vapor-deposition processes, they have long been known to possess a large uniaxial magnetic anisotropy normal to the film plane [1-11]. This anisotropy is critical to the materials' use as magneto-optic recording media. The role of the vapor-deposition process and the structural origin of such an anisotropy in an amorphous material is still, however, not understood. Extensive x-ray and neutron-scattering analyses have been done; all results are consistent with the usual model of a dense random packing of two different-size atoms [11-15], with some recent evidence for a small anisotropy in the radial distribution function (to be discussed below) [11]. *Local* magnetic anisotropy results from the interaction of the nonspherical, *f*-electron state of the Tb ion with its neighboring ions. In the random structure of an amorphous metal, the local environment varies from Tb site to Tb site, causing the local anisotropy direction to vary, which in turn causes the magnetic structure to become noncollinear and destroys the long-range magnetic order [12,16]. For the material to possess a *macroscopic* anisotropy, there must be an orientational coherence to these local anisotropy axis directions. In particular, since the observed macroscopic magnetic anisotropy is perpendicular, the distribution of these axes must be preferentially along the sample normal. This macroscopic anisotropy restores the long-range magnetic order and causes this material to appear in most respects like a conventional ferrimagnet.

A number of different sources of the structural anisotropy underlying the magnetic anisotropy have been postulated, ranging from stress, to a columnar microstructure, to a chemical or topological short-range order which becomes oriented normal to the film due to some generally unspecified vapor-deposition process [1-10]. Most recently, a new stress-induced mechanism has been proposed [11]. This model is based on x-ray scattering evidence of a compressive strain in these films, called a "bond orientational anisotropy" (BOA), which is apparently larger than can be explained by the observed elastic stress. The model proposes that the surface during growth is at a higher temperature than the bulk of the film; the stresses present then cause an *anelastic* strain as the surface cools. This proposed anisotropy mechanism is distinct from and *in addition to* the usual magnetoelastic (elastic stress plus magnetostriction) contribution to the anisotropy. We note here that the anelastic strain and hence the anisotropy due to this mechanism would necessarily change sign if the stress (at the surface of the film during growth) changed from compressive to tensile. There have also been recent suggestions that the anisotropy is due to a *surface* anisotropy of magnetic dipolar origin [17]; this would cause the anisotropy to vary as the inverse film thickness. Finally, it has been proposed that the source is a perpendicular anisotropy in the first monolayer deposited, due, for example, to this same magnetic dipolar mechanism. This leads to a growth-induced anisotropy throughout the film due to magnetic interactions of all subsequently deposited layers with the underlying layer [18]. Such an effect must vanish if the film is grown significantly above its Curie temperature.

We have found that the magnitude of the macroscopic perpendicular anisotropy may be varied by over an order of magnitude by varying the temperature at which the samples are prepared [19]. This increased coherence is induced by *raising* the deposition temperature, including temperatures well above the Curie temperature. The nonrandomness in the amorphous phase is therefore not due to magnetic interactions nor kinetic effects of vapor-deposition growth, such as shadowing and incident atomic-beam directions, which are reduced by raising the deposition temperature [20]. Instead, the nonrandomness is believed to be due to a reorienting of local adatom configurations so as to minimize chemical surface energy during growth, a thermally activated process. We call this a "texturing" of the amorphous phase, analogous to texturing of polycrystalline materials in which low-surface-energy grain orientations grow increasingly preferentially with increasing deposition temperature.

We find that changing the sign of the stress during growth (by manipulating growth conditions) has only a secondary effect on the perpendicular anisotropy, an effect entirely attributable to a conventional magnetoelastic effect. Films grown in both tension and compression exhibit large perpendicular anisotropy, a result inconsistent with the anelastic-strain model. Finally, we find that the perpendicular anisotropy is independent of film thickness (from 700 Å to 1 μm), ruling out surface magnetic anisotropy as a significant source.

In this work, amorphous Tb-Fe has been prepared by magnetron cosputtering onto a variety of substrates held at temperatures ranging from 77 to 700 K. The films are protected from oxidation by a 200-Å-thick Nb coating. X-ray diffraction and transmission electron microscopy have shown materials containing 14 to over 40 at.% Tb to be dense, amorphous materials with no evidence of columns or voids [6,8]. Argon, oxygen, and other impurities were not found at the limit of Auger analysis detection with depth profiling (< 1 at.%). The uniaxial intrinsic anisotropy constant K_{ui} [21] was determined by measuring the torque on the sample as a function of magnetic-field magnitude with the field held at ±45° relative to the magnetic easy axis. Further details concerning preparation and characterization of these materials may be found in Ref. [8].

Figure 1 shows K_{ui} (measured at room temperature) as a function of *deposition* temperature T_s for several compositions. K_{ui} increases by more than an order of magnitude as T_s is raised. These data include deposition temperatures well above the Curie temperature T_C (nearly constant at 400 K for $15 < x < 30$). T_C drops for $x > 30$, causing a decrease in K_{ui}. Samples with $x < 25$ are not plotted; for $18 < x < 25$, K_{ui} is artificially suppressed due to an artifact of analyzing a ferrimagnet near the magnetic compensation point ($x = 22$) [22], and for $x < 18$, a subtle phase separation causes a variety of anomalies in K_{ui} [8]. Films prepared at all deposition temperatures have been examined by x-ray diffraction and show no sign of crystallinity. A more sensitive test was made by annealing them at 350 °C for 2 h. Any small, oriented microcrystals should have grown larger in such an anneal, which would have caused an increase in both M and K_{ui} (T_C and M of the relevant crystalline phases are significantly higher than the amorphous phase). No change in M was found and, as observed by other researchers [7,11], K_{ui} decreased significantly.

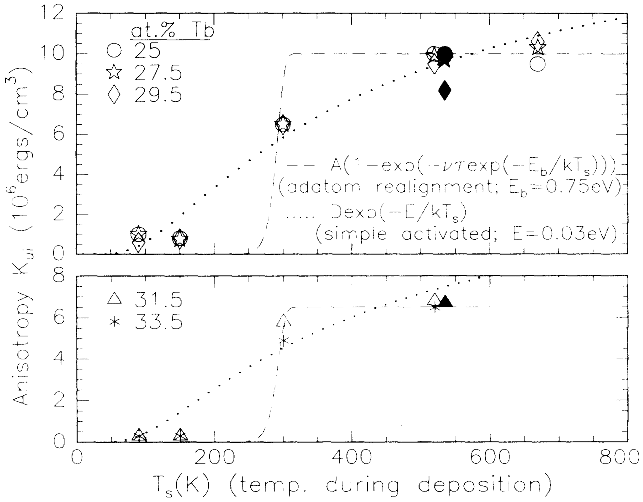

FIG. 1. Macroscopic anisotropy measured at room temperature vs deposition temperature T_s of 5000-Å-thick *a*-Tb_xFe_{100-x}. Open symbols: Ar pressure during growth 5 μm; films in compression at T_s. Solid symbols: Ar pressure 10 μm; films in tension at $T_s = 520$ K (data shown at 535 K for clarity).

We turn now to the effect of stress. It is generally observed that thin films prepared by magnetron sputtering at low Ar pressure are in compression and become tensile with increasing pressure due to thermalization of energetic ions [23]. Preparation of samples on thin (0.001 in.) Kapton polyimide substrates allows an approximate determination of the stress acting on the film, by measuring the curvature of the substrate/film combination [23–25]. Figure 1 shows data for tensile films (solid symbols) prepared at an Ar pressure of 10 μm at a deposition temperature of 520 K. The data are virtually identical to the values for compressive films (open symbols) prepared at an Ar pressure of 5 μm, with all other deposition conditions identical. The latter is in a state of compression at the deposition temperature (the stress $\sigma \approx -2\times10^9$ dyn/cm^2), while the former are slightly tensile ($\sigma \approx 0.75\times10^9$ dyn/cm^2). Thus there is *no* significant effect on the anisotropy of stress *during* the deposition, and hence *anelastic* strain cannot be its origin. The *elastic* stress-plus-magnetostriction contribution is also negligible compared to the intrinsic anisotropy for these samples, as demonstrated experimentally both by numerical estimates (see below) and by the independence of K_{ui} from substrate type (differential thermal contraction is of opposite signs for Kapton and fused quartz substrates). The observation that perpendicular anisotropy is obtained for films prepared in either tension or compression is consistent with the fact that it has been observed in *a*-Tb-Fe prepared by a wide variety of means, some of which produce films in compression and some in strong tension [1–11].

Data have also been taken for compressive and tensile samples grown at room temperature: For 5 μm Ar pressure, the elastic stress is compressive ($\sigma \approx -3\times10^9$ dyn/cm^2) while at 10 μm, it is strongly tensile ($\sigma \approx 4.5\times10^9$ dyn/cm^2) but appears inhomogeneous through the film thickness. Both types of samples have perpendicular magnetic anisotropy, including a remnant perpendicular moment. The value of K_{ui} for the room-temperature tensile film could only be estimated, due to the inhomogeneity, but is smaller than that of the compressive film. The difference can be completely explained by a magnetoelastic component $K_{u\sigma} \approx -3\lambda\sigma/2$, where λ is the magnetostrictive constant $\approx 4\times10^{-4}$ [26]; values of $K_{u\sigma} \approx 1.8\times10^6$ and $\approx -2.7\times10^6$ ergs/cm^3 are found for the compressive and tensile films, respectively, comparable to values in the literature [4,10]. When these contributions to K_{ui} are removed, an intrinsic component $\approx 4\times10^6$ ergs/cm^3 is found for both sets of samples prepared at room temperature.

Figure 2 shows K_{ui} as a function of thickness. No dependence is seen from 700 up to 10000 Å, demonstrating a negligible surface anisotropy contribution.

All data on the perpendicular magnetic anisotropy of *a*-Tb-Fe thus are explained by two components: an intrinsic, growth-induced component which is independent of the stress and a smaller magnetoelastic component.

Despite large changes in anisotropy, no significant dependence on deposition temperature has been found for M, T_C, or Mossbauer hyperfine distribution. Figure 3 shows M measured at room temperature as a function of composition. Compositions and thicknesses were determined by Rutherford backscattering. The lack of change in M or T_C argues against significant changes in exchange coupling or in the local nearest-neighbor structure.

The fact that the anisotropy *increases* with deposition temperature, independent of the stress, suggests that the effect of the deposition temperature is to allow rearrangements of local structural units into energetically favorable orientations, lowering the *surface* free energy *during* the growth, as previously suggested for *a*-Ge [27]. By contrast, nonequilibrium effects, such as a columnar microstructure in the amorphous phase, rely on a limited surface mobility and vanish with increasing deposition temperature [20]. Once the surface is buried by the next layer of incoming atoms, all directions become equivalent. The anisotropic local structure is, however, frozen in by low bulk diffusion rates. These local structures possess a local magnetic anisotropy, and hence their alignment in some preferred orientation will cause a macroscopic magnetic anisotropy.

We suggest that this effect be called a surface-mediated "texturing" of the amorphous phase, analogous to texturing of the crystallographic orientation of vapor-deposited crystalline films. In this better-known process, grains with low-energy surfaces (at which coordination numbers are a maximum) preferentially grow, causing highly oriented polycrystalline films on even amorphous substrates. In the present, amorphous structure, certain orientations of the local atomic arrangement may cause atoms at the surface to have a higher fraction of their bulk coordination. The x-ray scattering data showing bond orientational anisotropy, upon which the anelastic strain model was based [11], may reflect this texturing; measurements on *tensile* films with perpendicular anisotropy would provide a critical test.

The data in Fig. 1 show a dependence on deposition temperature T_s similar to that seen in the structure and properties (anisotropic properties were not discussed) of amorphous Ge [27]. There it was suggested that the dependence on T_s is related to the probability that an initial adatom configuration will transform to a lower-energy configuration before being buried by the next monolayer of adatoms. The fraction of misoriented configurations is

$$F(T_s)=\exp\{-\tau\nu\exp(-E_b/kT_s)\}\,, \qquad (1)$$

where τ is the time to deposit a monolayer (~ 1 sec), ν is the attempt frequency (on the order of phonon frequencies, $\sim 8\times10^{12}$ sec^{-1}), and E_b is the energy barrier separating the configurations. Assuming that the anisotropy in *a*-Tb-Fe is proportional to the fraction "correctly" oriented, the dashed lines in Fig. 1 are drawn for $E_b=0.75$ eV, a reasonable number for the relatively low barriers one might expect for metal adatom configurations ($E_b=1.5$ eV for *a*-Ge). The data shown are consistent with this expression; more deposition temperatures would of course be required to verify it. Equation (1) implies that for $T_s > T_s^c = E_b/k\ln(\tau\nu)$, virtually all possible reorientations have occurred. The saturation seen in the data in Fig. 1 at high T_s may be a consequence of $T_s > T_s^c$, as in this model, or it may result from the competing effect of the inherent annealing as the film grows. We also show in Fig. 1 (dotted lines) fits by a simple activated process, $De^{-E/kT}$. Such a curve would be related to a surface-diffusion model; we do not have a specific model for the process. The value thus obtained for E

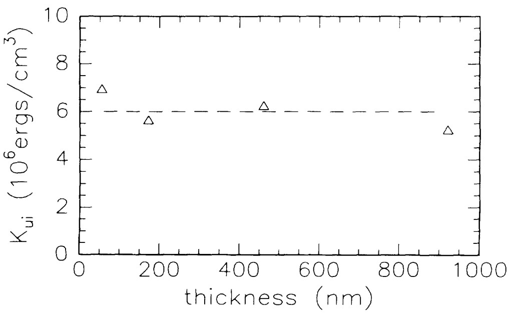

FIG. 2. Macroscopic anisotropy vs film thickness for *a*-$Tb_{28}Fe_{72}$ deposited and measured at room temperature.

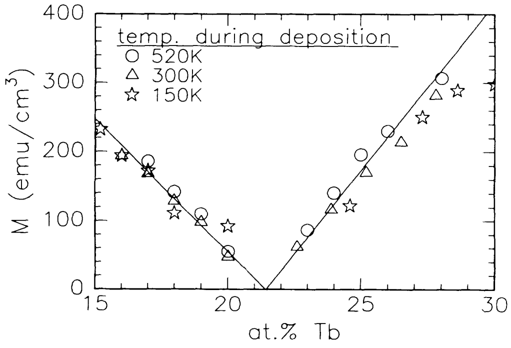

FIG. 3. Magnetic moment for *a*-Tb_xFe_{100-x} measured at 300 K vs x for various T_s.

(≈ 0.03 eV for all x) is small.

In conclusion, we have shown that the perpendicular magnetic anisotropy of amorphous Tb-Fe increases strongly with increasing deposition temperature, implying that surface energy rather than strictly kinetic growth processes are driving it. We have shown in particular that stress at the surface is not the driving force, as recently postulated. We suggest instead that a texturing of the amorphous structure, which lowers the surface energy during the growth, is the cause.

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