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Magnetism and microstructure of compositionally modulated disordered Fe/Ta films

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Results are presented on compositionally modulated disordered magnetic films of the form (X ÅFe/Y ÅTa), as a function of the modulation wavelength $\lambda \equiv X + Y$. Magnetization, ac susceptibility, torque magnetometry, x-ray diffraction, and Mössbauer measurements are used to probe magnetic transitions and microstructure. For X and Y values less than about 12 Å the structure is amorphous within the layers and compositionally modulated in the direction perpendicular to the film. Films with ultrathin individual layers (1–3 atomic diameters) exhibit perpendicular anisotropy while the thicker ones have in-plane magnetization. Spin-glass and ferromagnetic transitions are observed and are associated with specific regions of the multilayered films.

INTRODUCTION

Compositionally modulated disordered magnetic films (CMDMF) repressnt a new class of materials which are of great interest for reasons both pure and applied. In such amorphous and disordered magnetic multilayers, one can hope to study magnetic interfaces, quasi-two-dimensional magnetism,[1] and the effects of artificially induced microstructure on magnetic transitions and properties. From the viewpoint of applied physics these materials also are exciting because of their potential as magnetic information storage media. In particular, magnetic films with perpendicular anisotropy will have applications in erasable magneto-optic storage devices, perpendicular recording media, and bubble-domain materials. The ability to control the fundamental interactions and structure in CMDMF suggests that research on these materials should be fruitful.

We have investigated a variety of magnetic and structural properties in CMDMF of the form (X ÅFe/Y ÅTa), where X ranges from about 5 to 300 Å, and Y ranges from 3 to 180 Å. Of special interest are the magnetic properties, transition temperatures, anisotropy, and interface effects as a function of modulation wavelength, $\lambda \equiv X + Y$.

EXPERIMENT

Fe/Ta multilayers were prepared in a homemade, multiple-gun sputtering system with a microprocessor-controlled, water-cooled rotating substrate. The base pressure of the system is 2×10^{-7} Torr and special field-shaping techniques were employed to sputter Fe in a dc magnetron gun. The pressure during sputtering was held at 5 mTorr in a continuous flow of high-purity argon gas. The samples were deposited on mylar or glass substrates to a total thickness of about 3000 Å. The measurements performed included large and small-angle x-ray diffraction, vibrating-sample magnetometry, ac susceptibility down to 4.2 K, torque magnetometry, and Mössbauer effect. Details of some of these techniques can be found in the literature.[2]

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RESULTS AND DISCUSSION

In this paper we shall focus on two series of samples. In the first the Fe thickness is varied from 5 to 20 Å while holding the Ta thickness at 3 Å. Given the approximate atomic diameters of Fe and Ta of 2.5 and 3.0 Å, respectively, this series may be denoted as (X ÅFe/3ÅTa) or Fe_n/Ta_1, where $2\leqslant n\leqslant 8$, and where the subscript denotes the number of atomic layers. The second series has the form (X ÅFe/0.6X ÅTa) or Fe_{2n}/Ta_n. For this latter series and for $x\geqslant 18$ Å, the diffraction data show the Fe and Ta layers to be crystalline. This is shown for (20ÅFe/12ÅTa) in Fig. 1(a) where the broadened (110) peaks due to bcc Ta and Fe appear at about 40° and 44°, respectively. On the other hand, for (10ÅFe/6ÅTa), Fig. 1(b) shows only a single broad peak characteristic of a Fe-Ta glass (at 42°).[3] Small-angle diffraction data for the same two samples are exhibited in Fig. 2. These data can be explained on the basis of Bragg's law with $\lambda = 37$ Å for (20ÅFe/12 ÅTa) and $\lambda = 18.7$ Å for (10ÅFe/6ÅTa). The order of the diffraction n is shown on the appro-

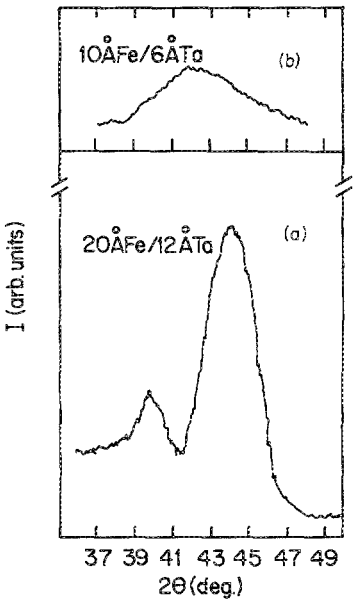

FIG. 1. Cu$K\alpha$ diffraction intensity as a function of 2θ for two of the films.

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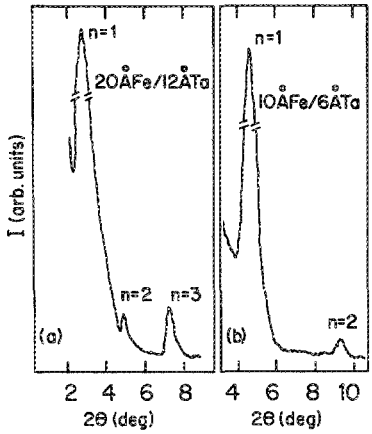

FIG. 2. Cu$K\alpha$ small-angle diffraction intensity for the two films referred to in Fig. 1.

priate peaks. Thus the structure of the (10ÅFe/6ÅTa) film can be described as amorphous within the layers but compositionally modulated with a bilayer wavelength of 18.7 Å. The (20ÅFe/12ÅTa) film is polycrystalline in both the Fe and Ta individual layers.

The "spontaneous" magnetization (extrapolated to $H=0$) measured at $T=300$ K, with the field parallel to the film, is shown for the (X ÅFe/0.6X ÅTa) series in Fig. 3. For Fe thicknesses less than about 15 Å, there is a sharp falloff of $\sigma_{\parallel}$ indicating a highly disordered, weakly magnetic, multilayered film.

The presence of uniaxial anisotropy was investigated with magnetization, Mössbauer, and torque magnetometry measurements. For the thicker Fe layers (e.g., $X=50$ Å, $Y=19$ Å), Mössbauer intensity measurements showed ratios characteristic of the magnetization **M** lying in the film (3:4:1) rather than random orientation (3:2:1). Also the magnetization measurements for **H** parallel and perpendicular to the film are characteristic of **M** in the film plane. However, the thinner samples clearly indicate a perpendicular anisotropy. An example, for (10ÅFe/3ÅTa), is shown in the hysteresis loops of Fig. 4(a). This is confirmed by torque measurements for the same sample shown in Fig. 4(b). From this data the perpendicular anisotropy constant K_u can be calculated from $K_u=\tau_{\max}/V$, where V is the volume of the film. For (10ÅFe/3ÅTa), the result is $K_u=1.0\times10^5$ erg/cm^3 and for (10ÅFe/6ÅTa), $K_u=9.3\times10^3$ erg/cm^3. For these two samples the shape anisotropy energy $2\pi M_s^2$

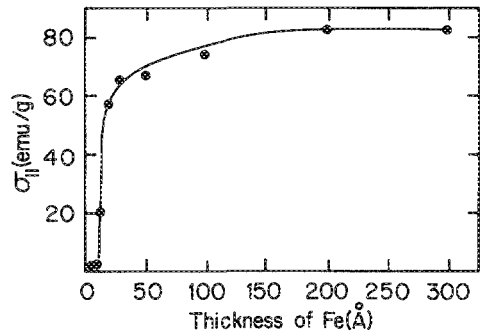

FIG. 3. $\sigma_{\parallel}$ (extrapolated magnetization) as a function of Fe layer thickness in (X ÅFe/0.6X ÅTa) series.

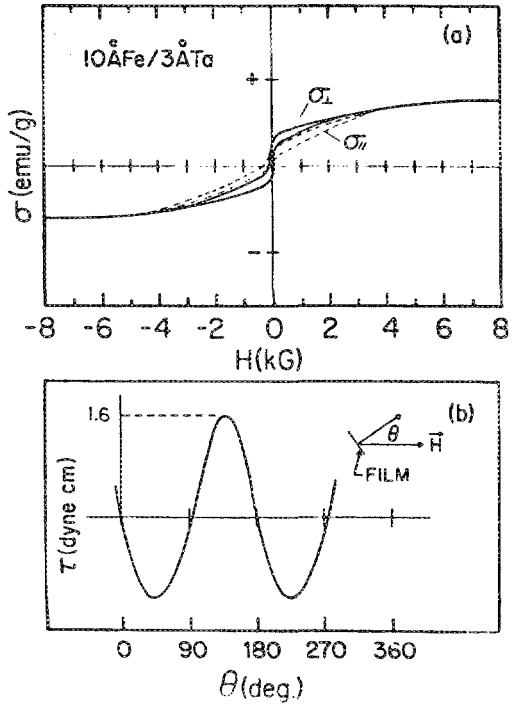

FIG. 4. (a) Hysteresis loop for (10ÅFe/3ÅTa) showing perpendicular anisotropy. (b) Torque as a function of θ for the same sample, also showing perpendicular anisotropy.

equals 1.4 and 2.0×10^3 erg/cm^3, respectively. Thus the perpendicular state of magnetization is consistent with the condition $K_u>2\pi M_s^2$.

It is of interest to consider the magnetic structure of these multilayered films. Clearly, for large X and Y values, the magnetic structure corresponds to widely separated, noninteracting sheets of ferromagnetic Fe. As X and Y approach several monolayers there is the possibility for (a) a complete loss of magnetism, (b) amorphous, quasi-two-dimensional spin-glass-like "interfaces," and (c) complex magnetic ordering of Fe layers due to interlayer coupling through nonmagnetic Ta layers. Figure 5 shows ac susceptibility data for Fe_4/Ta_1, Fe_3/Ta_1, and Fe_2/Ta_1 films. A low-temperature maximum, suggestive of a spin-glass transition, develops at a temperature T_g which decreases as the Fe

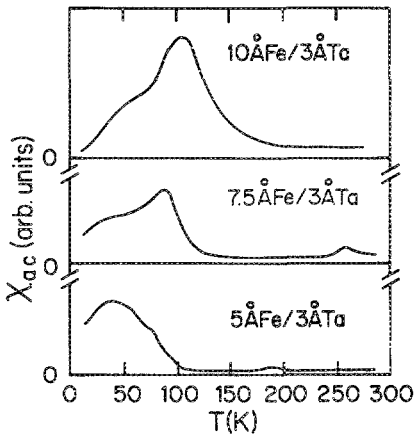

FIG. 5. ac susceptibility (280 Hz, $H_{ac}\approx0.1$ Oe) for three of the Fe_n/Ta_1 (X ÅFe/3ÅTa), samples.

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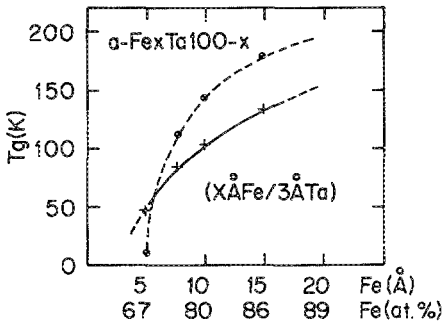

FIG. 6. Spin-glass ordering temperature T_g for the series of Fig. 5. Also shown are T_g values for bulk amorphous $Fe_x Ta_{1-x}$ alloys from Ref. 3.

thickness X decreases. In Fig. 6 a plot of T_g (X) is shown, along with T_g values of Chien *et al.*[3] on bulk, rapidly quenched $Fe_x Ta_{1-x}$ glasses. The similarity of these curves suggests that the susceptibility peaks arise from spin-glass transitions associated with glassy FeTa layers centered on the nominal Ta layers. Also of interest is a clear peak in x_{ac} at 260 K in the (7.5ÅFe/3ÅTa) sample. A smaller peak is seen at about 190 K in the (5ÅFe/3ÅTa) sample. Note that the (10ÅFe/3ÅTa) film does not show a corresponding peak, but if it exists it is likely above room temperature. Since this latter film is clearly ferromagnetically ordered at room temperature [see Fig. 4(a)], the likely explanation of these higher temperature x_{ac} peaks is that they correspond to ferromagnetic transitions of the Fe-rich layers. In these samples with very thin Fe and Ta layers, the Fe-rich layers are quasi-two-dimensional amorphous layers.

In summary, interesting magnetic transitions and properties have been discovered in Fe/Ta amorphous multilayers. In particular, perpendicular anisotropy has been observed in amorphous transition-metal–nonmagnetic-metal multilayered films. Several authors have discussed the importance of various types of anisotropic pair orderings as the source of perpendicular anisotropy in amorphous rare-earth–transition-metal alloys.[4] In the CMDMF under study here, Fe-Ta pairs oriented perpendicular to the films should gain in relative importance as the individual layer thicknesses become very small. This suggests that such pairs may be the origin of the perpendicular anisotropy observed in Fe/Ta. However, further systematic studies, which are now under way, will be required to more fully understand the microscopic origins and other possible sources[5] of the anisotropy.

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