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Optical and structural properties of cobalt-permalloy slanted columnar heterostructure thin films

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ABSTRACT

Optical and structural properties of sequential Co-column–NiFe-column slanted columnar heterostructure thin films with an Al_2O_3 passivation coating are reported. Electron-beam evaporated glancing angle deposition is utilized to deposit the sequential multiple-material slanted columnar heterostructure thin films. Mueller matrix generalized spectroscopic ellipsometry data is analyzed with a best-match model approach employing the anisotropic Bruggeman effective medium approximation formalism to determine bulk-like and anisotropic optical and structural properties of the individual Co and NiFe slanted columnar material sub-layers. Scanning electron microscopy is applied to image the Co-NiFe sequential growth properties and to verify the results of the ellipsometric analysis. Comparisons to single-material slanted columnar thin films and optically bulk solid thin films are presented and discussed. We find that the optical and structural properties of each material sub-layer of the sequential slanted columnar heterostructure film are distinct from each other and resemble those of their respective single-material counterparts.

1. Introduction

Highly ordered three-dimensional nanostructures grown by glancing angle deposition (GLAD), e.g., using electron beam evaporation constitute a material class with highly tunable physical and chemical properties and which render those materials useful for potential device applications. For example, thin films deposited by GLAD form slanted columnar structures, which are highly anisotropic [1–3]. So far, the majority of reports have focused on GLAD thin films consisting of a single material while further interacting physical and chemical properties may increase the range of useful functionality when the nanostructured materials are formed sequentially using multiple source materials [4–11]. For example, inhomogeneous nanowires have long been valued as theoretical model systems for several applications such as for tuning magnetic [12,13] and magnetoresistive properties [14,15], optimizing photocatalytic properties [6,16–18], and as photoelectrochemical electrodes [19].

Characterization of anisotropic, nanostructured thin films is possible with Mueller matrix generalized spectroscopic ellipsometry (MMGE). MMGE has been shown as a robust, non-destructive means of model-based characterization of the optical and structural properties of anisotropic GLAD grown nanostructured thin films [20]. The layered-model approach allows for the separation of sequential material layer properties, which are part of ultra-thin film architectures.

The focus of this work is to gain understanding about the optical and physical properties of slanted columnar heterostructure thin films (SCHTFs) deposited by GLAD. For the SCHTF, two different materials, Co and NiFe, are deposited sequentially by GLAD. We use MMGE and a best-match model approach for the analysis. The MMGE data is analyzed by employing the anisotropic Bruggeman effective media approximation (AB-EMA) formalism to determine the full dielectric function tensor and structural characteristics for each material sub-layer comprising the SCHTF [21–24]. For comparison, single-material slanted columnar thin films (SCTFs) as well as optically bulk solid thin films comprised of Co and NiFe were grown and subsequently analyzed. In addition to ellipsometric model analysis, scanning electron microscopy (SEM) is utilized by analyzing top view and cross-section micrographs to determine structure uniformity between the Co and NiFe sub-layers and to

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compare with our ellipsometric model results. The MMGE analysis indicates notable distinctions between the Co and NiFe material sub-layers with respect to their structural and optical properties. Each SCHTF material sub-layer expresses good agreement with their single-material analogues.

2. Experimental

The Co–NiFe SCHTF was deposited in a custom built ultra-high vacuum chamber by glancing angle electron-beam evaporation onto a silicon wafer coated with a natively grown oxide. A rotatable four-pocket crucible indexer houses the deposition materials inside the ultra-high vacuum chamber with background pressures at or below 10^{-9} mbar. The substrate surface normal was directed 85° from the evaporation material source. The Co SCHTF sub-layer was deposited onto the substrate at a constant rate of 3.0 Å/s monitored by a quartz crystal microbalance deposition controller. A shutter was moved between the source and the substrate to protect the Co slanted columns during source cooling. Without breaking vacuum, the NiFe source was rotated into the electron beam flux for heating and subsequent evaporation. The shutter was opened once the NiFe deposition rate achieved 3.0 Å/s. After NiFe SCHTF sub-layer deposition completion, the substrate was immediately transferred to the atomic layer deposition reactor (Fiji 200, Cambridge Nanotech Inc.) for Al_2O_3 passivation [25]. A total of 45 cycles of trimethylaluminum and nanopure water (18.3 MΩ) were introduced to the Co–NiFe SCHTF with a 20 s vacuum purge after each precursor pulse equating to ~3.5 nm conformal growth. The samples were held at 150 °C with a background pressure of 2.45×10^{-1} mbar throughout the deposition. Few nanometer thickness Al_2O_3 coatings have been shown to protect GLAD grown nanostructured films from structural degradation, corrosion, and thermal degradation [2,26,27]. MMGE measurements were performed on the Co–NiFe SCHTF with a multi-wavelength spectroscopic ellipsometer with a spectral range from 194 to 1690 nm (RC2, J. A. Woollam Co. Inc.). The ellipsometric measurements were gathered from four unique angles of incidence Φ_a= 45°, 55°, 65°, and 75° through a full azimuthal sample rotation $0° \leq \phi \leq 360°$ by 6° intervals, which allows for observation of the full optical anisotropy of the SCHTF samples.

3. Results and discussion

The SEM image in Fig. 1(a) depicts the top view of the Co–NiFe SCHTF. Overall, the slanted columns are narrower at the base of the SCHTF than at the top, indicative of a change in porosity between the bottom and top sub-layers of the SCHTF. A small change in slanting angle between the bottom and top sub-layers can also be seen. Further, the top view SEM image presents a highly ordered, spatially coherent SCHTF growth. The cross-section SEM image in Fig. 1(b) depicts little disjunction at the Co–NiFe interface as well as thickness uniformity of the SCHTF. The total film thickness and average slanting angle, θ, can be estimated to be ~95 nm and ~60°, respectively, from the SEM micrographs.

The MMGE data was subsequently analyzed through a best-match multiple layered-model approach by applying the AB-EMA [20] to describe each of the sub-layers in the SCHTF separately. The approximation is an extension of the isotropic Bruggeman formalism [28]. The AB-EMA allows for description of mixtures of anisotropic polarizabilities into the material model-layer. The depolarization factors L_a^D, L_b^D, and L_c^D represent the relative magnitudes of a polarization ellipsoid of one given constituent within an intrinsic orthogonal coordinate axis system, which can be ascribed to the columnar thin films as shown in Fig. 1(b). Euler angle rotations connect the description of the anisotropic polarizability axes $\boldsymbol{N}_a$, $\boldsymbol{N}_b$, and $\boldsymbol{N}_c$ (Fig. 1(b)) with laboratory coordinate axes x, y, and z

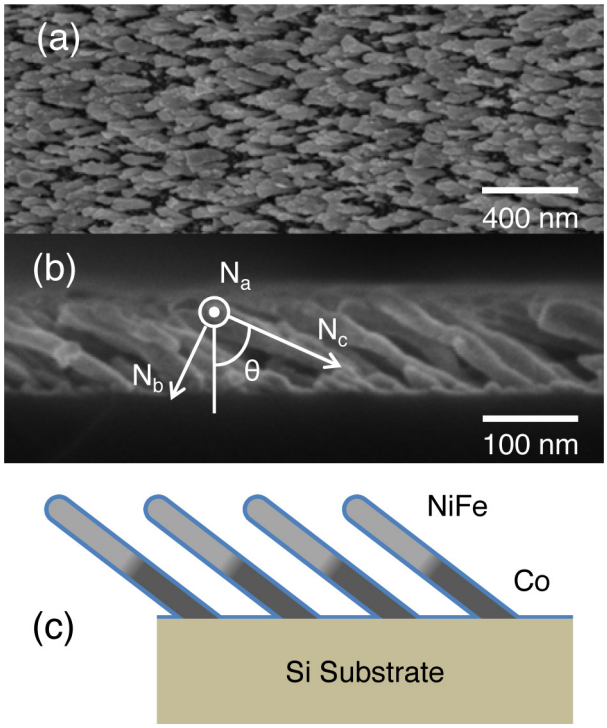

Fig. 1. High resolution SEM micrographs show top view (a) and cross-section (b) of the Co–NiFe SCHTF under investigation with scale references inset as white bars. A schematic representation of the inhomogeneous film (c) with the material layers labeled. The coordinate system depicted on panel (b) displays the intrinsic polarizability axes of the anisotropic optical system.

as described previously in the work of Schmidt et al. [20–22,29]. The AB-EMA formalism allows for optical analysis of SCTFs and provides model parameters such as constituent material fractions, layer thickness, SCTF slanting angle, and polarizability along the major intrinsic axes.

By applying the AB-EMA formalism, the dielectric function tensor components $\varepsilon_{eff,j}$, where $j = a, b, c$, the polarizability directions along the intrinsic SCTF coordinate frame can be determined by

$$\sum_{n=1}^{m} f_n \frac{\varepsilon_n - \varepsilon_{\mathrm{eff},j}}{\varepsilon_{\mathrm{eff},j} + L_j^D(\varepsilon_n - \varepsilon_{\mathrm{eff},j})} = 0, \quad (1)$$

where m is the number of material constituents of the model-layer [30,31]. ε_n is the bulk-like reference dielectric function. The constituent volume fraction contributions, f_n, as well as the depolarization factors, L_j^D, must independently add to unity and unity, respectively.

Fig. 2 depicts spectrally dependent MMGE data from 194 nm to 1685 nm for the Co–NiFe SCHTF at ϕ = 228°. There is near perfect agreement between experimental and best-match model data for the four measured angles of incidence. ϕ = 228° is specifically selected for presentation due to its strong anisotropic response, however, all ϕ angles exhibited exemplary agreement between the experimental and model data. The best-match model parameters of the SCHTF are presented in Table 1, showing the parameters that are allowed to simultaneously vary until the simulated model best-matches the experimental data. The determined values of constituent material fractions, depolarization factors, and slanting angles are consistent with previous reports for an AB-EMA model analysis of GLAD thin films of similar material types [21,32,33]. The best-match model overall thickness of 84.2 nm and θ values of 57.93° and 63.21° for Co and NiFe, respectively, compare well with the values determined from the SEM images in Fig. 1(b). In further agreement to the SEM micrograph analysis, the SCHTF

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