B8 - Thin Films and Multilayers

B8 - O1

X-RAY ANALYSIS OF THIN FILM STRUCTURES USED IN MAGNETIC SENSORS AND MAGNETIC MEDIA

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The performance of multilayer stacks used in magnetic devices such as giant-magnetoresistive (GMR) sensors and magnetic media is intimately linked to the physical properties of the contributing functional films as well as to their crystalline and interfacial structure. A number of surface-sensitive X-ray methods to investigate these parameters is presented. Specular X-ray reflectometry (XRR) has proven to be an absolute method to measure film thicknesses in complicated layer sequences - as e.g. the Ru-nanolayer thickness in GMR stacks - and deposition rates with sub-Angstrom accuracy [1]; it can be used for the routinely offline monitoring of large-area coupons, but also for the calibration of quicker, indirect, inline monitoring methods, e.g. X-ray fluorescence (XRF). Off-specular XRR was successfully applied to monitor the interfacial properties of ion-bombarded Co/Pt-superlattices, a promising candidate for perpendicular recording based on the so-called patterned media approach [2,3]. In-plane X-ray diffraction (XRD) has gained increasing attention with regard to the study of the real structure (crystallite size, preferred orientation, stress) of functional thin films. The method is well-suited to investigate phase transitions in thin films, such as the fcc to fct transition of a GMR sensor's PtMn pinning layer upon annealing [4].

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B8 - O2

SMALL AND LARGE ANGLE X-RAY DIFFRACTION IN METALLIC MULTILAYERS

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Many physical properties of artificially layered structures are strongly dependent on the atomic arrangement at the interface. Indeed, the interfacial roughness affects the electrical and optical properties of semiconductor superlattices, plays an important role in the long-range magnetic coupling in magnetic multilayers, or strongly influences the specular reflectivity from multilayer x-ray mirrors. A detailed characterization of the interface is therefore essential. The most widely used ex-situ structure and interface characterization technique for multilayers is x-ray diffraction (XRD). The nondestructive XRD technique is commonly used in -2 geometry, in which the scattering vector is perpendicular to the sample surface and only –so called – "one dimensional" information in the growth direction is obtained. Nevertheless, a specific multilayer x-ray diffraction profile allows one to deduce some structural parameters by fitting the measured intensity profiles with model calculations using the computer programs SUPREX [1] and SLERF [2]. In this way the quantitative values for the rms roughness of the interfaces may be determined, but the method cannot provide information about the lateral character of the roughness. Indeed, both models are only valid for specular x-ray scattering. An interfacial roughness may be laterally as well as vertically correlated. Both kinds of correlation influence strongly their small- as well as large- angle nonspecular x-ray diffraction profiles. It becomes clear that the most valuable information about a real multilayer structure can be obtained from nonspecular x-ray scattering measurements [3]. Unfortunately, the most of reported in the literature data is based on simple specular -2 scans. This fact is apparently caused by the fact that interpretation of nonspecular scans cannot be performed in the straightforward way, as it is nowadays possible for specular scans. An example of the usefulness of nonspecular x-ray scattering measurements is provided by the stress determination that often is carried out on the basis of so called $\sin^2$ method. This method has been well developed for polycrystalline thin films. However, for multilayered epitaxial systems the method may lead to a false result. Therefore, 3D models must be used to interpret the performed scans in a correct way. In this work, a review of 1D, 2D and 3D models necessary for x-ray diffraction profiles interpretation will be done. Both specular and nonspecular measurements will be described. Application of analytical as well as simulation models will be discussed.

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B2 - O3

MICROSTRUCTURE DETERMINATION AS A FUNCTION OF DEPTH IN THIN MULTILAYER STRUCTURES

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This presentation describes a new approach for extracting microstructural information in polycrystalline multilayers as a function of depth. Most X-ray scattering experiments average information over large depths in multilayers, however in some cases knowledge of surface strains, relaxation and distortions is of interest. Most work on measuring structural information in thin polycrystalline layers as a function of depth has been confined to synchrotron sources, however by modification of standard laboratory equipment and the application of the unique scattering properties of periodic structures this can be performed with a laboratory sealed source.

The method has been applied to Nb-Al multilayers, and it is clear that the single element layers do not necessarily have the same state of strain nor do they necessarily have the same crystallite size. In combination with other X-ray techniques, large area reciprocal space mapping, reflectometry applied to these samples it is possible to build a fairly comprehensive view of these materials.

This method is based on in-plane scattering, whose strength is modulated in depth by a standing wave associated with the density and roughness disruption at each interface. The standing wave is modified with increasing incidence angle. This description of the process will be discussed in terms of the experimental methodology and the detailed theoretical modelling required to interpret the results.

B8 - O4

IN SITU STUDIES OF DIFFUSION AND CRYSTALLIZATION PROCESSES IN THIN ITO FILMS BY TEMPERATURE AND TIME RESOLVED GRAZING INCIDENCE X-RAY DIFFRACTOMETRY

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Studies of diffusion and crystallite growth in thin films have attracted wide attention due to the increased use of multilayered thin film structures in modern electronic devices. Particularly with regard to the time stability of such devices, the investigation of phase transitions or diffusion processes are of special interest. In situ high temperature grazing incidence X-ray diffractometry (HT-GIXD) is well suited, but so far rarely used for characterizing the kinetic parameters of such processes. We present both a quantitative description of oxygen diffusion into plasma enhanced deposited metallic In/Sn films and of the crystallite growth of indium tin oxide thin films.

Tin-doped indium oxide (ITO) films were deposited on Si(100) substrates without external heating by means of dc planar magnetron sputtering. A metallic In/Sn (90/10) target and an argon/oxygen gas mixture were used. The flow of the reactive gas oxygen was varied between 0 and 2 sccm. Bias voltages between 0 and –100 V were used. With increasing oxygen flow, the film structure and composition changes from crystalline metallic In/Sn to amorphous ITO. Oxygen diffusion into metallic In/Sn films and crystallite

growth of ITO films were investigated by in situ HT-GIXD at temperatures ranging from 100 to 300 °C and a vacuum of $5*10^{-3}$ mbar.

The ITO formation is determined by two processes: the diffusion of oxygen into the metallic grains and a fast crystallization process. Kinetic parameters for both processes were derived. A model was developed which allows the determination of the diffusion coefficient D from the time dependence of the integral intensity of the ITO-X-ray reflection. This mathematical model incorporates the following physical basics:

A thin film of the material *n* (In/Sn) converts into a crystalline phase *m* (ITO) in a diffusion limited process. The intensity of an X-ray reflection of phase *m* in the film at time *t* can be calculated using Eq. (1).

$$dI_m \quad B_m K_0 I_0 A A_m(x,t)\frac{C(x,t)}{C_0}dx \qquad (1)$$

with B_m the theoretical intensity factor, K_0 the apparatus constant, I_0 the intensity of the incident X-ray beam and A the irradiated sample area. The ratio $C(x,t)/C_0$ is the fraction of phase *m* in the depth *x.* The absorption correction factor A_m depends on the phase composition: $A_m(x,t) \quad \exp(\ \bar{}\ (x,t)zx)$

$$\bar{}\ (x,t) \quad 1/x_m \int_0^{x_m} ((\ _m \quad _n)\frac{C(x,t)}{C_0} \quad _n)dx$$

and a geometry factor *z*. The time dependence of $C(x,t)/C_0$ is given by Fick's second law

$$\frac{C(x,t)}{t} \quad D\frac{^2C(x,t)}{x^2}$$

The amount of amorphous ITO in the as-deposited films is considered by a factor *f.*

By integration Eq.(1) over the film thickness *d* with the above mentioned assumption, the intensity of a reflection of phase *m* follows:

$$I_m(t) \quad \frac{I \quad _m z}{1 \quad \exp(\quad _m zd)} \int_0^d (f \quad (1 \quad f)\frac{C(x,t)}{C_0} \exp\left\langle \frac{1}{x_m}\int_0^{x_m} \bar{}\ dx \quad zx\right\rangle dx \qquad (2)$$

A detailed description of the model is given in [1, 2]. The diffusion coefficient depends on the applied bias voltage but it is not influenced by the oxygen flow during film deposition. From the temperature dependence of D the activation energies for the diffusion process can be calculated [3]. For $T < 150$°C the activation energy depends on the applied bias voltage. For T > 150°C the *D* values still differ with the bias voltage but the activation energy seems to be independent of the deposition conditions.

The crystallization of amorphous ITO can be understood by application of the Johnson-Mehl-Avrami theory [3]: $y_m(t) \quad 1 \quad \exp\left\langle \quad (t/t_0)^n \right\rangle$ with y_m normalized intensity, *t* the annealing time, *n* the reaction order and $1/t_0$ the rate constant. For our films we get *n*-values between 2.4 and 3, i.e. a two dimensional crystallization process [4].

Our experimental results show that the application of in situ GIXD measurements is well suited to investigate temperature induced processes. This method establishes a wide range of analytical possibilities.

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B8 - O5

DIFFRACTION STRESS ANALYSIS OF STRONGLY FIBRE-TEXTURED Au LAYERS

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Diffraction stress analysis on the basis of the so-called $\sin^2$ -method is a well-established method. However, a straightforward application of the $\sin^2$ -method, using the so-called X-ray elastic constants (XECs) and , is possible only if the specimen is macroscopically elastically isotropic. This implies that the $\sin^2$ -method (based on XECs) can not be applied to crystallographically textured specimens, because for such specimens, macroscopic, mechanical anisotropy generally occurs.

For specimens presenting crystallographic texture that is both strong (i.e. the random texture fraction is small) and sharp (i.e. the orientations of crystallites exhibit only small spreads from the ideal orientations), the so-called crystallite group method has been proposed [1,2]. As proposed the method was intended for single-crystal like textures only. The crystallite group method can also be adapted to deal with specimens presenting a fibre-texture [3].

In this work, the crystallite group method has been employed for the diffraction stress analysis of fibre-textured gold films. The consequences of the macroscopically elastically anisotropic nature have been demonstrated. Further, possible alternative measurement strategies, i.e. the use of one reflection versus the use of multiple reflections, and the corresponding procedures required for calculating lattice strains from measured lattice spacings, have been discussed in particular in view of the susceptibility of the obtained stress results to instrumental aberrations. The ranges of applicability of the crystallite group method in view of texture strength, sharpness and complexity and with respect to possible mechanical loading states (biaxial rotationally symmetric versus biaxial) have also been analysed.

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