ACCURATE TEXTURE MEASUREMENTS ON THIN FILMS USING A POWDER X-RAY DIFFRACTOMETER

MARK D. VAUDIN
NIST, Gaithersburg, MD, USA.

Abstract

A fast and accurate method that uses a conventional powder x-ray diffractometer has been developed for measuring crystalline texture. A θ-2θ scan of a Bragg peak from the textured planes is collected and also a θ scan, or rocking curve, using the same Bragg peak. The method has important advantages over other techniques: a large x-ray footprint can be used, thus obtaining significantly higher intensity which is particularly significant for thin films; no randomly textured specimen is required. The large footprint leads to considerable tilt-induced defocussing during the θ scan – the scattering angle varies along the iradiated length of the specimen as it is tilted out of the symmetric position. To obtain an accurate texture profile from the θ scan, corrections for defocussing and absorption must be applied, and the θ-2θ scan of the Bragg peak, which gives the variation of scattered intensity with angle, is used for the defocussing correction. First principles calculations are used to correct for absorption, with the film thickness and x-ray absorption coefficient as parameters. The technique has been applied to several classes of technologically important materials used in thin film form, such as films of electroplated copper for advanced metallization, and also BST films down to 26 nm thick, which are candidates for DRAM applications.

Keywords: accurate texture; thin films; powder x-ray diffractometer; defocussing

1 Introduction

Crystallographic texture in thin films, i.e. the preferred orientation of particular crystal planes relative to the film substrate, is a common and frequently useful phenomenon. Important materials properties, such as remanent polarization, dielectric constant and elastic moduli, are typically anisotropic, and the most effective use of anisotropic materials in thin film applications often involves controlling the texture of

the film. For example, the use of $PbZr_{1-x}Ti_xO_3$ (PZT) in non-volatile ferroelectric random access memories (NVFRAM) requires optimization of the film texture to best utilize the remanent polarization of PZT while satisfying other processing requirements (Hadnagy 1997).

In a typical laboratory or industrial setting, texture is measured using x-ray diffraction. There are certain problems peculiar to the measurement of texture in thin films, the most significant of which is that the intensity of diffracted x-rays that can be obtained from a thin film is frequently so small that intensity measurements are of low accuracy. Thin films deposited on planar substrates typically display fiber (axisymmetric) texture with the substrate normal as the fiber axis. Substrates patterned with surface features such as trenches for conduction vias may display more complex, three dimensional texture (Lingk et al. 1998). In previous work (Vaudin et al. 1998), a technique was described that measures fiber texture using a conventional x-ray powder diffractometer. The technique required the recording of two scans from the sample: a high resolution θ-2θ scan of a Bragg peak whose diffracting planes are normal to the preferred orientation direction; and a θ scan obtained using this peak. The θ scan gives the variation of scattered intensity with specimen orientation and contains the required texture information, but it must be corrected for defocussing and absorption to obtain the texture profile. In a pole figure measurement, this is typically achieved by measuring a specimen of the same phase composition but with no preferred orientation (i.e. a random specimen) and dividing the intensity from the textured specimen by that from the random specimen, thus obtaining multiples of a random distribution (MRD) as a function of specimen orientation. In the technique of Vaudin et al., the θ scan that would be obtained from a random specimen is calculated from the θ-2θ scan of the Bragg peak, taking into account defocussing and absorption, and the texture profile is determined by dividing the experimental θ scan by the calculated random θ scan. In this way, the shape of the texture profile is determined, but the scaling is arbitrary. The technique has been extensively applied to textured bulk alumina (Seabaugh et al., in preparation).

2 Theory

Axisymmetric thin film texture implies the preferential alignment of a particular set of crystal planes parallel to the substrate. The data collected from the specimen are the two x-ray scans described above, $I_{pk}(2\theta)$, a θ-2θ scan of a Bragg peak (Bragg angle θ_B) from the aligned planes, and a θ scan, $I_{rc}(\omega)$, where the specimen orientation is tilted away from the symmetric orientation through ω about an axis in the specimen surface normal to the diffraction plane, while the scattering angle remains fixed at $2\theta_B$. (In Vaudin et al., α was used for the tilt angle instead of ω.) The θ scan for a randomly oriented specimen of the same material, $I_{rand}(\omega)$, is calculated from $I_{pk}(2\theta)$ by correcting for defocussing and absorption, and the texture profile, $T(\omega) = I_{rc}(\omega)/I_{rand}(\omega)$, is calculated. As the specimen is tilted away from the symmetric orientation in either direction, the x-ray scattering angle, 2θ, varies along the irradiated length of the specimen. The central assumption of the theory is that the intensity of a ray scattered by the specimen through 2θ is proportional to the θ-2θ scan intensity at angle 2θ. Thus, the θ scan intensity at angle ω is proportional to the integral of the Bragg peak intensity, as illustrated in Fig. 1; the scattering angle variation is indicated: $2\theta_- < 2\theta < 2\theta_+$.

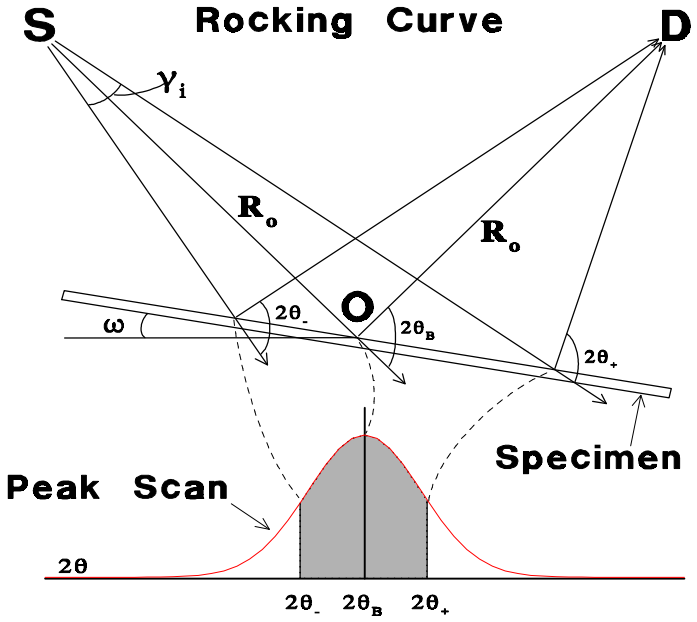

Figure 1: Schematic of x-ray source, specimen and detector showing relation between defocus during θ scan and peak scan intensity

The integral is carried out as a function of γ between $-\gamma_i/2$ and $\gamma_i/2$:

$$I_{rand}(\omega) \propto \frac{1}{\gamma_i}\int_{-\gamma_i/2}^{\gamma_i/2} I_{pk}(2\theta(\omega,\gamma))\, d\gamma \qquad \textbf{(1)}$$

where γ_i is the divergence of the incident beam and γ is the angle between the ray and line SO in Fig. 1, i.e. the divergence from the central ray. The factor that determines the constant of proportionality in eqn.(1) is absorption. From simple geometry, an x-ray striking the specimen surface at angle ϕ, penetrating to depth z in the specimen, scattering through 2θ and returning to the surface is attenuated by $K(z, \phi, 2\theta)$:

$$K(z,\phi,2\theta) = \exp\left\{-\mu z\left(\frac{1}{\sin\phi} + \frac{1}{\sin(2\theta-\phi)}\right)\right\} = \exp\{-\mu D(2\theta,\phi)z\} \qquad \textbf{(2)}$$

where μ is the linear coefficient of x-ray absorption, $D(2\theta, \phi)$ is defined implicitly, and ϕ is the angle of incidence of the ray on the specimen surface: $\phi = \theta_B + \omega - \gamma$. For a ray at γ to SO incident on a film of thickness t, the total effect of x-ray absorption on the recorded intensity, $A(\omega, \gamma)$, is found by integrating through the thickness of the film:

$$A(\omega,\gamma) = \int_0^t K(z,\phi(\omega,\gamma),2\theta(\omega,\gamma))\, dz$$

$$= \frac{1-\exp\{-\mu D(2\theta,\varphi)t\}}{\mu D(2\theta,\varphi)} \qquad \textbf{(3)}$$

To relate I_{pk}, the intensity measured at zero tilt, to the intensity that would be measured during a θ scan when the specimen tilt varies, the I_{pk} values are first divided by $A(0, \gamma)$ and then multiplied by $A(\omega, \gamma)$, giving the correction factor, $F(\omega, \gamma) = A(\omega, \gamma) / A(0, \gamma)$,

and thus we obtain:

$$I_{rand}(\omega) = \frac{1}{\gamma_i}\int_{-\gamma_i/2}^{\gamma_i/2} F(\omega,\gamma) I_{pk}(2\theta(\omega,\gamma))\, d\gamma \qquad \textbf{(4)}$$

Thus, measuring I_{pk} and I_{rc} allows the texture profile $T(\omega)$ to be calculated. When $I_{pk}(2\theta_B)$ and $I_{rc}(0)$ are measured, the diffractometer is in the same configuration, and therefore the measured intensities should be equal within experimental error. Agreement between them is a good check on the reliability of the data.

The units of $T(\omega)$ can be found by considering that when $\omega = 0$, 2θ varies very little with γ (only by the flat specimen correction), and therefore from eqn. (4), $I_{rand}(0) = I_{pk}(2\theta_B)$ to a very good approximation. Since, within experimental error, $I_{pk}(2\theta_B) = I_{rc}(0)$, it can be seen that within experimental error $T(0)$ should equal 1. As ω increases, $T(\omega)$ decreases from 1 for positively textured samples. If $T(\omega)$ were known over all orientation space, the profile could be scaled to be in MRD by multiplying by the MRD value at $\omega = 0$, MRD_0, which can be found from the normalization condition:

$$\int_0^{\pi/2} MRD_0 T(\omega) \sin\omega d\omega = 1 \qquad \textbf{(5)}$$

With this rocking curve technique, ω is limited to a range: $0 < \omega < \theta_B$. However, eqn.(5) can still be used in those cases where the texture profile of a material is narrow enough that it decays to zero within the observed orientation range and can be assumed to be zero in the unobserved orientation range.

3 Experimental

The technique has been designed to be used with a conventional divergent beam powder x-ray diffractometer. The two x-ray scans collected, the θ-2θ and θ scans, were collected under identical conditions; in particular, the incident and receiving slits were the same. In addition, no anti-scatter slits were used; Soller slits could have been used but were not in this case. For both scans, the dwell time was set by experience to be long enough to produce data of an adequately high signal to noise ratio. The angular steps of the θ-2θ and θ scans were at most one tenth of the full width at half maximum of their respective peaks. The θ-2θ and θ scan intensity data were processed using software called Texture[ref], which requires the thickness and x-ray absorption coefficient of the film, the beam divergence, the diffractometer radius and $2\theta_B$ for the θ scan as additional input data.

4 Results and Discussion

Fig. 2(a) shows a log scale plot of a θ-2θ scan from a 1.6 μm copper film electrodeposited on a TaN / SiO_2 / Si (001) substrate. In addition to peaks attributed to the substrate from TaN, Si and a weak, unidentified peak at 81°, all the fcc Cu peaks are present, but relative to their random integrated intensities (Powder Diffraction File 1996), the 111 and 222 peaks are over an order of magnitude larger than the 200, 220 and 311 peaks, indicating significant (111) texture, and also the likelihood of a random fraction. This is confirmed by Fig. 2(b) where the (111) texture profile measured using both 111 and 222 peaks is plotted, arbitrarily scaled to a maximum of 1000. The raw data have been corrected using thicknesses of 0.8, 1.6 and 3.2 μm; the texture plots show that

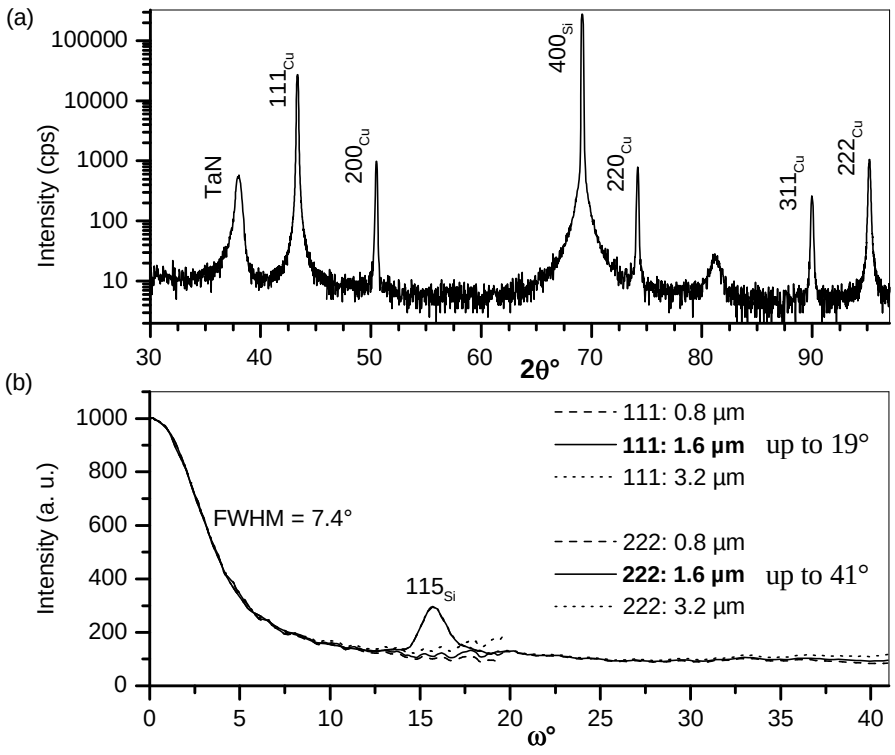

Fig. 2: (a) θ-2θ scan and (b) texture profile from 1.6 µm electroplated Cu film

the best match between the 111 and 222 plots occurs for the 1.6 µm correction. Comparing the FWHM value of 7.4° for the texture profiles with the 6.1° value for the raw θ scans shows that the defocusing and absorption corrections had a significant effect on the profiles. The presence of a significant random fraction is clear from the constant texture level from 15°<ω<40°. When testing the technique on bulk specimens, Vaudin et al showed that a texture profile of constant value was obtained from material that had been shown by other methods to be untextured, indicating that the corrections were correctly formulated. The agreement between 111 and 222 texture in Fig. 2(b) is clearly better when corrected for the true film thickness than double or half the value, which shows the method works well in this case. However, this is not a full validation of the thin film correction, which would require a set of thin film specimens with known textures, not currently available.

Fig. 3(a) shows a θ-2θ scan from a $Ba_{0.7}Sr_{0.3}TiO_3$ (BST) thin film deposited on a thin film of Pt on Si. The position of the 111_{BST} peak is indicated, and, despite the strong overlap with the 111_{Pt} peak, it is clear that the 111_{BST} peak is very weak or absent. This suggests that the film contains very little or no randomly oriented material but consists of two populations of grains with (001) and (110) texture, respectively. Texture profiles collected using the 001, 110 and 002 peaks are shown in Fig. 3(b) and indicate that the two populations are both highly textured with full widths at half maximum (FWHM) values from 2.8° to 3.4°; the agreement between the 001 and 002 FWHM values is good (the FWHM values are accurate to ±0.1°). If the texture profiles (corrected data) are compared with the θ scans (uncorrected data), the raw and corrected data are

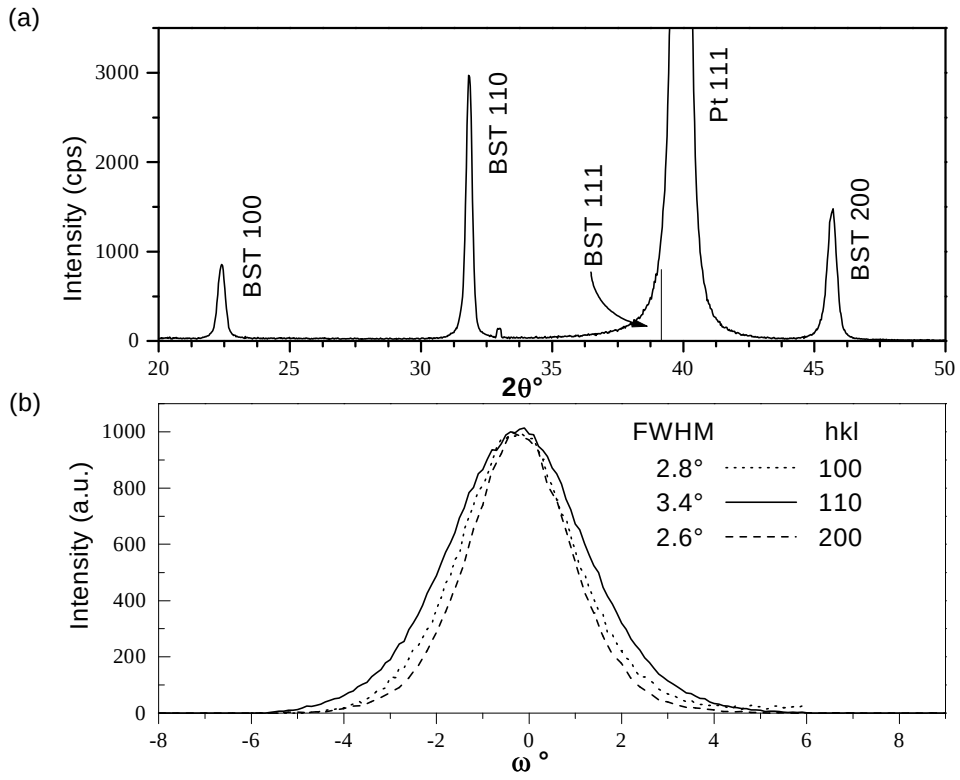

Fig. 3: (a) XRD scan and (b) texture profiles from 36 nm BST film

the same within experimental error, indicating that for highly textured materials, the uncorrected 'rocking curve' data are an accurate measure of texture. However, this would not be known without the analysis methods described in this paper.

5 References

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