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STRESS GRADIENTS AND ANISOTROPY IN THIN FILMS

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ABSTRACT

Indications that steep through-thickness strain gradients occur in vapour-deposited chromium films stemming from previous observations of film curling during spontaneous delamination from substrates, have been substantiated by analysis and simulation of Bragg X-Ray diffraction peaks. The presence of large through-thickness compressive strains, that increase quadratically with distance from the substrate from about zero at the interface to around 0.8% at the film surface, was deduced by empirical computer matching of diffraction peak shapes.

INTRODUCTION

Quantitative interpretation of spontaneous, i.e. residual stress-driven, delamination of thin films from substrates requires precise knowledge of the internal stress state at the interface and throughout the film [1]. The direction and magnitude of stresses in films on substrates can be determined by measurements of substrate distortion. However, any preferred crystallographic texture, steep through-thickness stress gradients or large in-plane stress anisotropy, as well as the occurrence of any brittle, ductile or diffusive stress relaxation processes during or subsequent to film deposition can lead to an erroneous interpretation of substrate bending [2,3]. On the other hand, measurements of residual stresses in crystalline materials by x-ray diffraction are less influenced by localized film relaxation and particularly by cracking.

STRAIN MEASUREMENTS BY X-RAY DIFFRACTION

Representative tracings of 110 Bragg peaks from several thin Cr films vapor-deposited on glass substrates are presented in Figure 1 along with the peak from a reference sample of annealed Cr powder. The large shift in peak position reveals that lattice planes parallel to the surface are in compression with a mean strain of about 0.5%, i.e. the films are in tension. The considerable broadening of the lines is indicative of a substantial range of elastic strains in the films (the pronounced asymmetry of the peaks derives from non-uniformities within the films as discussed below). The observed broadening is a composite of effects due to the fine grain size and non-uniform strain in the films, as well as instrument broadening, is given by [4],

$$B_{obs}^2 = B_I^2 + B_{GS}^2 + B_{\Delta\varepsilon}^2 \qquad (1)$$

The grain size broadening B_{GS} and non-uniform strain broadening $B(\Delta\varepsilon)$ are given by,

$$B_{GS} = \frac{k \cdot \lambda}{t \cdot \cos\theta_B} \qquad (2) \qquad \text{and} \quad B(\Delta\varepsilon) = -2\left(\frac{\Delta d}{d}\right)\tan\theta_B \qquad (3)$$

Subtracting instrument broadening, established with a well-annealed, coarse grain standard, and rearranging terms gives,
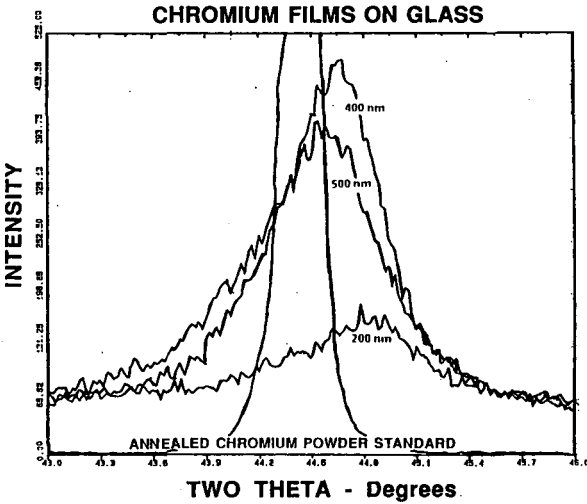
CHROMIUM FILMS ON GLASS

Fig. 1. X-Ray diffraction peaks (110 bcc - Cu K alpha) for thin Cr films vapour-deposited on glass - superimposed peak from annealed Cr powder.

TWO THETA - Degrees

$$\frac{B^2 \cos^2 \theta_B}{\lambda^2} = \frac{k^2}{t^2} + 4 \left(\frac{\Delta d}{d}\right)^2 \frac{\sin^2 \theta}{\lambda^2} \tag{4}$$

Here $B^2 = B_{obs}^2 - B_I^2$,t is the diameter of the grain size, and k^2 is the constant depending on the shape of the grain. It may be seen that the slope and the intercept of a plot of equation (4) using experimental values for B^2, $\cos^2\theta$, $\sin^2\theta$ are directly related to the non-uniform strain and the grain size respectively. The representative results for grain size and strain range as tabulated below in Table I are in the right range but are about a factor of 2 too large.   This will be discussed below in relation to the unusual asymmetry of the x-ray peaks.

TABLE I. Results of grain sizes and non-uniform strains from equation (4)

film thickness	intercept	slope	GS Å	Δe %
300 nm	0.35x10^{-4}	5.856x10^{-4}	169	1.21
600 nm	0.20x10^{-4}	10.858x10^{-4}	223	1.65

A more fundamental approach is to fit the full intensity distribution of both first and second order reflections with a Fourier series (Warren-Averbach) method [5]. Data analysis using proprietary software on a Siemens diffractometer indicated grain sizes in the films of about 150 - 200 Å, in fair agreement with the Cullity method, and gave non-uniform strain values around 0.5%, but with persistent inconsistencies in results. It became clear that for both the Cullity and Warren-Averbach methods, very low diffracted intensities from the films made it impossible to obtain good data from the high order reflections that are needed for reliable analyses. Furthermore the marked asymmetry of line shape, indicative of the presence of contributions to line broadening that are not randomly distributed, is not contemplated in either formalism. In fact, diffraction peaks obtained using more highly absorbed Fe K-alpha varied significantly in shape and peak position from the Cu K-alpha results, indicating a change in strain with depth in the film. Thus it is evident that conventional x-ray methods are not adequate when steep, through-thickness strain gradients are present.

COMPUTER SIMULATION OF X-RAY PEAKS

A more straightforward, albeit empirical, approach to interpreting the x-ray observations is to compute matching intensity distributions based on information about grain size and strain distribution obtained from electron microscopy studies that are summarized below.

When the adhesion of a Cr film to the substrate is poor, it may peel away from the substrate as illustrated in Figure 2. As mentioned previously, the occurrence of a strain gradient is suggested from this scanning electron micrograph which shows that the film has curled into a very tight roll after delamination from the substrate with the outer surface of the film in tension. Peak surface strains greater than 2 % are suggested by the small radius of curvature of the curls. Oblique deposition results in the formation of narrow hoops (by film cracking and followed by delamination) indicating a large stress anisotropy between directions parallel and perpendicular to the line of sight to the source [6]. Transmission electron microscopy of cross-sections of a number of typical films revealed that the grain size was not uniform through the film but increased from the substrate to the top by a factor of about 5 for the thickest films.
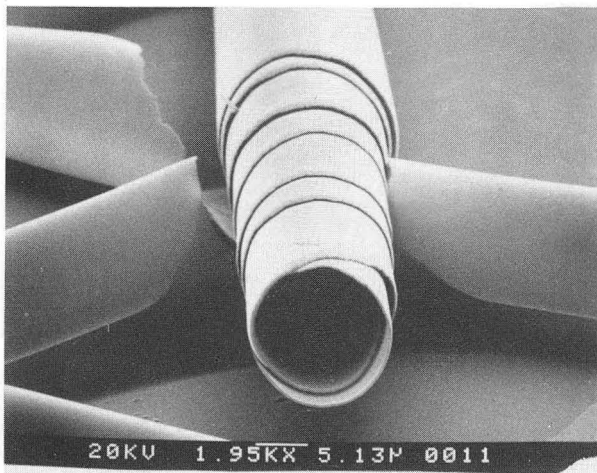

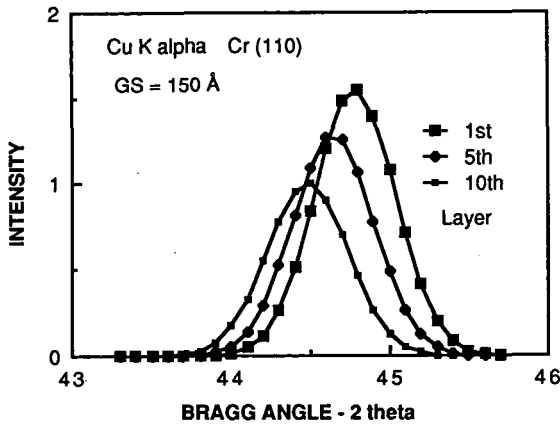Fig. 2. Scanning electron micrograph showing curling of delaminated Cr film.

20KV   1.95KX   5.13P   0011

Fig. 3, Illustration of method of calculating intensities from individual layers in thin films. Effect of absorption in middle and bottom layers may be seen.

In computing x-ray diffraction peaks, each film was considered to be a stack of 10 layers with different (110) d spacings as shown in Figure 3. The intensity diffracted from each layer was corrected for absorption and for grain size and instrument broadening and summed to generate simulated peaks.

Simulated (110) peaks for 3 different variations of d spacing with distances from the substrate are shown in Figure 4. Although the fit is not particularly good for
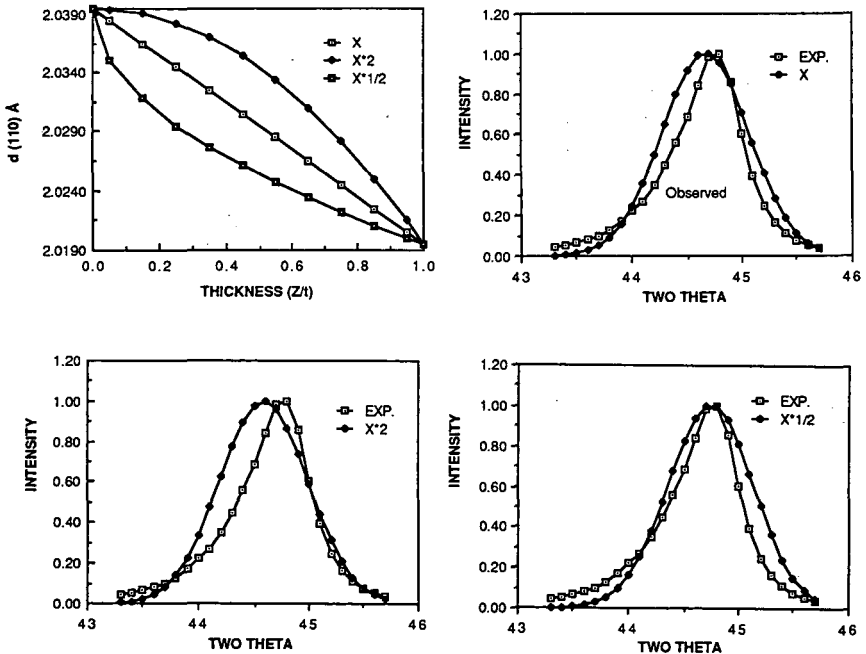

Fig. 4. Comparison of observed and simulated X-ray diffraction peaks for linear and non-linear lattice parameter gradients in a 3000 Å Cr film. Depth correction for absorption included – grain size assumed to be 100 Å.

any of the assumed strain distributions, the general peak shape is about right. Much better fits were obtained by using the actual grain size values determined by electron microscopy as illustrated in Figure 5. Very good fits were also achieved for Fe k-alpha diffraction peaks from the same films. As anticipated, the need to fit experimental peaks obtained with 2 different radiations narrowed the possible range of strain distributions considerably.

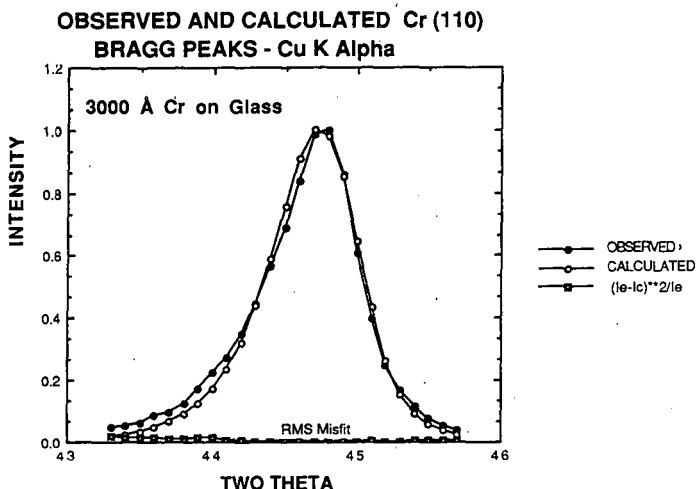**OBSERVED AND CALCULATED Cr (110)
BRAGG PEAKS - Cu K Alpha**

Fig. 5. Observed and simulated X-ray diffraction peaks for 3000 Å of Cr on glass. A linear grain size gradient and a non-linear strain gradient assumed - corrected for absorption.

RESULTS AND DISCUSSION

The elastic strain distributions in two Cr films derived from computer matching of (110) Bragg peaks are shown in Figure 6. The plots illustrate a general feature about strain vs thickness that was noted throughout this study. In particular the strain at the substrate and the rate of increase with distance from the substrate tend to be smaller in the greater final film thickness. Increasing the substrate temperature is known to result in lower internal stresses in Cr films and so it is reasonable that the slight temperature rise that occurs during deposition will also result in lower strain values for thicker films.

Interpretations of film cracking and delamination based on the newly established knowledge of strain gradients in Cr films will be reported separately.

ACKNOWLEDGEMENTS

STRAIN DISTRIBUTION

IN VAPOUR-DEPOSITED CR
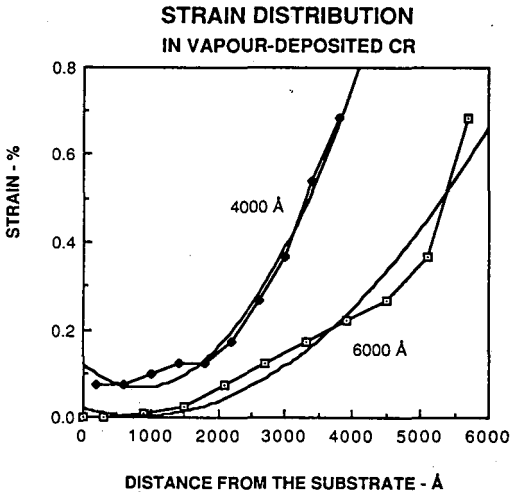

DISTANCE FROM THE SUBSTRATE - Å

Fig. 6. Strain distribution in Cr films determined by simulation of X-ray peaks.

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