SPATIALLY-TRANSIENT STRESS EFFECTS IN THIN FILMS
BY X-RAY DIFFRACTION

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ABSTRACT

We present a review of the application of diffraction stress/strain analysis to small volumes. For cases in which the material properties and/or the stress state are not homogeneous, traditional approaches may yield erroneous stress results. On the other hand, with proper care, relevant mechanical information about the system can be obtained. Through the use of conventional and synchrotron-based X-ray methods, we can determine the amount of strain transfer between thin film features that possess heterogeneous stress distributions and the underlying substrate. Two examples of such studies are presented. The resulting data is used to assess the validity of several models often used to predict the mechanical behavior in thin film/substrate composites.

INTRODUCTION

The traditional diffraction techniques for stress/strain determination in crystalline materials require a priori both the material properties and stress/strain states within the irradiated volume of the sample to be homogeneous. These requirements are rigorously justifiable in few specimen classes. For example, a single crystal specimen with a homogeneously distributed far-field load at its boundary would satisfy both requirements if the irradiated volume were much smaller than the sample volume and located far from the sample edges. For polycrystalline specimens that are quasi-isotropic (randomly textured and with a grain size approximately two orders of magnitude less than the beam size), the analysis is applicable if the average stress/strain state within the irradiated volume is homogeneous.

Even though the use of traditional techniques in measuring small volumes is problematic, recent advances in sources and optics of diffraction systems, and the trends in technology that control properties on ever decreasing scales have resulted in the extension of diffraction methods to smaller volumes. For example, in the microelectronics industry, strain distributions within integrated circuitry impact reliability aspects of device behavior and the mobility of silicon-based transistor junctions. Consequently, particular stress distributions can be advantageous in certain applications and not in others. Modeling and measurement of the stress fields in such structures is rapidly growing. In the following, we present a review of traditional X-ray diffraction stress analysis and two cases of its application to spatially-varying stress distributions in blanket films and finite-sized features.
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THEORY

The basic theory of X-ray diffraction stress analysis is based on measurement of the lattice spacing of crystallites which satisfy the Bragg condition for a particular reflection. Figure 1 depicts the diffraction geometry commonly used in X-ray stress/strain measurements, where the coordinate system of the laboratory, Li, is transformed into that of the specimen, Si, through the use of the angles, \( \varphi \) and \( \psi \). The diffraction plane is defined by L3, the diffraction vector and L1.

![Diffraction geometry of specimen](from reference [1].)

The plane spacing measured at a given set of angles, \( d_{\varphi \psi} \), can be translated into a strain if the unstressed plane spacing, \( d_0 \), is known.

\[
\varepsilon_{\varphi \psi} = \frac{d_{\varphi \psi} - d_0}{d_0}
\]

(1)

By measuring \( d_{\varphi \psi} \) over several orientations, through a rotation about the sample normal (\( \varphi \)), or a tilt of the sample normal with respect to the diffraction vector (\( \psi \)), one can deduce the strain tensor of the sample by solving a second rank tensor transformation equation:

\[
\varepsilon'_{33} = \frac{d_{\varphi \psi} - d_0}{d_0} = a_{3k} a_{3l} \varepsilon_{kl}
\]

(2)

where \( a_{3k}, a_{3l} \) are the direction cosines between the sample and laboratory coordinate systems, Si and Li. We note that, in the case of a general stress state, at least six independent measurements are required to determine the strains in the sample system \( \varepsilon_{kl} \).
The link between strain and stress distributions involves a tensor multiplication with either a fourth-order compliance, $S_{ijkl}$, or stiffness, $C_{ijkl}$, tensor (Hooke’s law). These elastic tensors are single-valued only for single crystal samples. For all other samples, the calculation of the elastic tensor involves an averaging over the mechanical response of the diffracting crystallites for a particular reflection. For a thorough description of the common models, see either reference [1] or [2]. The most basic assumptions employed are either of a uniform strain tensor, the Voigt average [3], or a uniform stress tensor, the Reuss average [4], among all of the grains of a polycrystalline aggregate. These models represent the bounds of the aggregate’s mechanical behavior, although ensembles composed of diffracting crystallites, may lie outside of the Voigt values calculated in the traditional method, which are constant for all X-ray reflections [5]. X-ray elastic constants (XEC) calculated using the Neerfeld-Hill model [6,7] take an arithmetic average of the Voigt and Reuss values. More refined models, such as that of Kroner [8], often produce XEC values close enough to that of the Neerfeld-Hill (N-H) average so that N-H values can be used within the experimental error associated with the X-ray diffraction measurements.

The application of any of these models, which implicitly assume a randomly textured polycrystalline sample, must also include an assumption of the state of stress within the sample. For example, the measured lattice spacing, $d_{\psi\psi}$, for a linear, elastic sample under an in-plane biaxial stress (all out-of-plane stress components $\sigma_{ij3}$ are zero) can be represented using the X-ray equation:

$$\varepsilon_{\psi\psi} = \frac{d_{\psi\psi} - d_0}{d_0} = \frac{1}{2} S_2 \sigma_\psi \sin^2(\psi) + S_1 (\sigma_{11} + \sigma_{22})$$

(3)

where

$$\sigma_\psi = \sigma_{11}\cos^2(\varphi) + \sigma_{12}\sin(2\varphi) + \sigma_{22}\sin^2(\varphi)$$

and $S_1$ and $\frac{1}{2} S_2$ represent the X-ray elastic constants (XEC). By taking the derivative of Equation (3) with respect to $\sin^2(\psi)$, one obtains the commonly used $d$ vs. $\sin^2(\psi)$ form of the

![Graph showing X-ray elastic constants (XEC) for a polycrystalline Cu sample (a) S1 and (b) $\frac{1}{2} S_2$ as a function of the orientation parameter $\Gamma$. Selected X-ray reflections are indicated on the top of the graphs.](image)
X-ray equation:

\[
\frac{\partial d_{\psi\phi}}{\partial [\sin^2(\psi)]} = \frac{1}{2} S_2 \sigma_{\phi} d_0
\]  

(4)

In this form, the error associated with the uncertainty in \(d_0\), which is approximately 1 to 2%, has less impact than that found in subtracting \(d_0\) from \(d\) directly, as indicated in Equation (3). However, the presence of out-of-plane shear stresses requires a more complicated representation than either Equation (3) or (4). For a comprehensive treatment of this case, see reference [1].

Because the elastic response of the sample can be represented by two constants in this form, the mechanical behavior of the polycrystalline aggregate is termed “quasi-isotropic.” Figures 2(a) and 2(b) depict the XEC values calculated for a quasi-isotropic Cu sample. The orientation parameter, \(\Gamma\), is a function of the Miller indices (hkl) for a particular reflection:

\[
\Gamma = \left( \frac{h^2k^2 + h^2l^2 + k^2l^2}{h^2 + k^2 + l^2} \right)^2
\]  

(5)

which spans from 0 to 1/3, corresponding to the (h00) and (hhh) reflections, respectively. As indicated in Figure 2, the anisotropic nature of Cu leads to a large variation in both \(S_1\) and \(S_2\), particularly for small values of \(\Gamma\).

The range in XEC values impacts the resultant strain tensor as calculated through the measurements conducted on different X-ray reflections. To illustrate this point, an example is presented of a Cu polycrystalline sample under an isotropic, in-plane biaxial stress state of 200 MPa. The calculated dependence of strain, \(\varepsilon_{\phi\psi}\), using Equation (3), is depicted in Figure 3, where the Neerfeld-Hill (N-H) approximation was used to generate the XEC values.

The results depicted in Figure 3 indicate not only that the slopes of the \(d\) vs. \(\sin^2(\psi)\) curves associated with different X-ray reflections can vary by over a factor of 2 but that the \(\psi\)-angle, corresponding to zero strain, will be different, changing from 42 to 48 degrees. The calculation of a “stress-free” direction from this value of \(\psi\), will clearly vary depending on the reflection used in the X-ray measurement even for a simple case of a Neerfeld-Hill approximation under a isotropic biaxial stress field. For general cases, in which the true stress state and the exact XEC for a particular material system may be unknown, the determination of \(d_0\) using this approach may result in significant errors in the subsequent strain measurements.

Although the previous example addressed the effects of crystal anisotropy on X-ray stress measurements for a quasi-isotropic sample, an additional complication associated with anisotropy in real, polycrystalline ensembles should also be mentioned. When preferred orientation, or texture, exists in a specimen, the population of crystallites that will diffract for different values of \(\phi\) and \(\psi\) varies. In extreme cases, the selectivity of the Bragg condition produces an insufficient number of diffracting grains at certain orientations, either due to strong texture or incident X-ray radiation with a low divergence angle. The diffraction peaks measured
in such cases will contain an unrepresentative sampling of the d-spacing which may lead to erroneous stress measurements.

![Graph of calculated strain vs. \(\sin^2(\psi)\)](image)

Figure 3. Calculated lattice strain as a function of \(\sin^2(\psi)\) using the Neerfeld-Hill model for isotropic biaxial stress of 200 MPa.

An example of this issue is presented for a strongly fiber-textured (111) Cu film, where the measured (111) fiber plot is depicted in Figure 4(a). By comparing the population of (111) or (hhh) grains for different tilt angles to a value of 1, corresponding to a randomly textured sample, one can locate orientations that would be susceptible to an inadequate number of diffracting

![Graph of Cu (111) intensity vs. tilt angle and \(\sin^2(\psi)\)](image)

Figure 4. Measured Cu (111) fiber texture for blanket film sample (a) fiber plot and (b) \(\sin^2(\psi)\) plot to indicate difference between randomly textured sample and population of diffracting grains for (hhh) stress measurement.
grains. Figure 4(b) contains the same information presented in Figure 4(a) but plotted as a function of $\sin^2(\psi)$ to illustrate sections of a $d$ vs. $\sin^2(\psi)$ plot, such as that at 0.25, where the X-ray stress measurement conducted for a (hhh) reflection will possess 40 times fewer grains than a sample with no texture.

X-ray stress measurements of this sample, performed using the Cu (311), (331), and (420) reflections, are depicted in Figure 5 for two orthogonal sample directions: $\phi = 0^0$ and $\phi = 90^0$. Because the sample possesses in-plane transverse isotropy, measurements conducted in both of these directions should be equivalent. However, values from the (331) reflection, as measured using synchrotron-based techniques, exhibit a distinct asymmetry. The high degree of selectivity imposed by the highly parallel nature of the synchrotron beam exacerbates the poor sampling of diffracting grains for this reflection. In fact, the divergence associated with conventional, laboratory-based X-ray measurements produce consistent stress values of approximately 275 MPa, as indicated in Figure 5.

**FINITE-SIZE FEATURES**

It must be noted that, even when the material is quasi-isotropic, the stress state within the material may have spatially transient terms. One reason for such variation is the proximity of geometric discontinuities, such as boundaries. For example, since a free edge can not support a normal stress, the stress field approaching a free edge must decay in accordance with the equations of equilibrium. To assess the effects of free edges on the stress relaxation in thin film features, a 1 $\mu$m-thick Cu film was evaporated onto a 5 inch Si wafer, possessing a 10 nm Cr layer for adhesion purposes. The wafer was etched to produce arrays of square features either 3 $\mu$m or 14 $\mu$m in size in its central portion while an annular region along the periphery of the
wafer remained unetched. By comparing the mechanical response of the Cu blanket film in the
annular region to that from the etched Cu structure, we can determine the feature size at which
the stress decay due to the presence of free edges dominates the volume-averaged stress values.

Conventional X-ray diffraction measurements were conducted on the samples using a Rigaku
RINT 2100 Ultima theta-theta diffractometer with Fe Kα radiation and a rotating anode source.
A d vs. sin²(ψ) analysis was performed on the Cu features and the blanket film region using the
Cu (311) and (222) reflections. Samples were thermally cycled from room temperature to 450°C,
and held for 30 minutes during each X-ray measurement. Because the beam size was
approximately 10 mm × 15 mm at normal incidence, the diffraction data represented a volume
average of the irradiated Cu features.

The results of the stress measurements are plotted in Figure 6 for both the blanket film and 3 µm
wide Cu features. The Cu blanket film exhibits the traditional behavior associated with metallic
films on substrates with a lower coefficient of thermal expansion (CTE). The decrease in Cu
stress as temperature is increased follows the trend predicted by linear elasticity up to
approximately 100°C. The slope of the decrease agrees well with the calculated values using the
Timoshenko model of a strained, bimaterial composite [9].

![Figure 6. Measured stress for 3 µm wide Cu features and the corresponding blanket film during thermal cycling. Deviation from linear elasticity (dotted line) is observed in the blanket film behavior for temperatures greater than 100°C.](image)

Plasticity effects lead to a deviation in the mechanical response of the Cu film above 100°C,
ultimately leading to a saturated stress value above 300°C where diffusional and creep
deformation dominates. Although the 3 µm Cu features exhibit similar qualitative trends, the
magnitude of stress is much lower not only at room temperature but during the entire thermal
cycle. Subsequent thermal cycles conducted on the 3 µm features were performed to analyze the
elastic region of the heating curve (< 100°C) and revealed that the slope of the Cu stress decrease was approximately 45% of that produced in the blanket film. In contrast, the 14 µm wide Cu features behaved similarly to the blanket films during thermal cycling, within the experimental error of the X-ray stress measurement.

MECHANICAL MODELING OF EDGE EFFECTS

The previous experimental evidence can be used to assess the validity of the response of a strained thin film feature, as predicted by a number of potential mechanical models. A comparison of the attributes of the linear elastic models commonly used to calculate the stress distributions in finite-size features is contained in Table I. In particular, the way in which these models treat the mechanical properties of the common interface between thin film and substrate serves as an important delineation. The transfer of strain between the constituent materials of a multilayer composite can be greatly affected by the mechanical integrity of the common interface. The stress distributions predicted by the edge-force model, proposed by Blech and Meieran [10], assume a constant stress value in the film and no interface between the feature and the underlying substrate. The distributed force approach of Hu [11] applies a strain compatibility condition between the thin film feature and substrate, but is invalid for features of finite width. Although the lap shear model, developed by Suhir [13], assumes a fixed mechanical response of strain transfer between the thin film and substrate, an interfacial region does not exist. Stress distributions predicted from the Eshelby inclusion method [15], which assume a perfectly bonded interface, result in a constant stress distribution within the finite feature and cannot be used to analyze stress relaxation due to features with free edges.

Table I. Compilation of mechanical models of strained features with finite dimensions.

<table>
<thead>
<tr>
<th>Model</th>
<th>Interface treatment</th>
<th>Variable interface</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Edge Force [10]</td>
<td>No</td>
<td>n/a</td>
<td>Point forces on substrate</td>
</tr>
<tr>
<td>Shear Lag [12]</td>
<td>Yes</td>
<td>yes</td>
<td>Equal film and substrate width</td>
</tr>
<tr>
<td>Lap Shear [13]</td>
<td>Yes</td>
<td>no</td>
<td>Equal film and substrate width</td>
</tr>
<tr>
<td>Eigenvalue [14]</td>
<td>Yes</td>
<td>yes</td>
<td>Fourier series</td>
</tr>
<tr>
<td>Eshelby inclusion [15]</td>
<td>Yes</td>
<td>no</td>
<td>Uniform strain in feature</td>
</tr>
</tbody>
</table>

Of the models listed in Table I, only two possess the capability of varying the interfacial mechanical properties or adhesion in the composite. Both the shear lag approach, applied to the analysis of semiconductor features by Chen and Nelson [12], and the eigenvalue method, also known as the Fadle-Papkovich model [14], ascribe a proportionality constant between the
interfacial shear stress, $\tau$, and the magnitude of horizontal slip, $\Delta u$, between the film and substrate. The terms $R$ and $G_0/t_0$ refer to the interfacial compliance ratios for the eigenvalue and shear lag methods, respectively:

$$\tau_{\text{eig}} = \frac{\Delta u}{R} \quad \tau_{\text{S-L}} = \left(\frac{G_0}{t_0}\right) \Delta u$$

(6)

Although these treatments of the interface are identical, the approach commonly used in the Fadle-Papkovich method determines the eigenvalue that possesses a negative real component with the smallest magnitude. A recent comparison between these models indicated that the shear lag model produced a more accurate representation of the decay in the normal stress at the free edges [16].

Because the X-ray stress measurements indicated that, during the elastic portion of the heating curve, the volume-averaged stress distributions of the 3 $\mu$m Cu features are approximately 45% of those in the blanket film, we can solve for the interfacial compliance ratio necessary to produce this response using the shear lag model. Figure 7 depicts the calculated normal stress distributions for 3 $\mu$m wide and 14 $\mu$m wide Cu features on Si, where the value of $G_0/t_0$ has been calibrated so that the average normal stress in the 3 $\mu$m wide Cu features is 0.45 $\sigma_0$, or 45% of the Timoshenko stress value for a blanket film.

![Figure 7: Calculated stress distribution for 14 $\mu$m and 3 $\mu$m Cu features on Si using the shear lag (S-L) model (from reference [17]).](image)

As indicated in Figure 7, the predicted decay in the normal stress extends up to 3.5 $\mu$m from the free edge of the features so that for features less than 7 $\mu$m wide, the interaction regions in the feature created by the edges overlaps. As the feature size diminishes, these regions dominate the overall stress distribution as evidenced in the case of 3 $\mu$m wide Cu features, where no portion of
the sample reaches the value of stress experienced in the blanket film case. Although the model contains assumptions that limit its application to all cases (e.g: linear elastic behavior), the approach is being used to develop and experimentally verify more comprehensive models of the mechanical response of within thin film features.

SUMMARY

The use of X-ray diffraction-based stress measurements to experimentally verify mechanical models in features with free edges has been presented. Microstructural aspects, such as texture, can exacerbate the selectivity associated with X-ray diffraction and must be considered for any $d$ vs. $\sin^2(\psi)$ analysis. The nature of the interface plays an important role in dictating the transfer of strain between thin film and substrate, which is contained in several mechanical models. By comparing the mechanical behavior of Cu features on Si substrates during thermal cycling, one can experimentally verify the elastic response predicted by these models, for which the shear lag approach is the most accurate. However, any method, numerically or analytically based, that are applied at temperatures that exceed the elastic limit of the composite, can lead to erroneous calculations and must be used with caution.

REFERENCES