RESIDUAL STRESS ANALYSIS WITH MULTIPLE HKL RINGS COLLECTED BY AREA DETECTORS

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ABSTRACT
Measurement of residual stresses in thin films or textured materials by X-ray diffraction is always a challenge task. The limited diffraction volume in thin films or the directions with low pole density in sharply textured materials results in a weak diffraction signals. The sharp stress gradient, anisotropic grain shape and inhomogeneous phase and microstructure distribution also add difficulties in stress analysis with the conventional X-ray diffraction method. The nonlinear $\varepsilon_\psi - \sin^2\psi$ behavior, commonly associated with thin films and textured materials, produces poor results. This paper introduces a method using diffraction rings from multiple (hkl) crystalline planes collected with area detectors to analyze residual stresses.

FUNDAMENTALS
The stress measurement with an area detector is based on a direct relationship between the stress tensor and the diffraction cone distortion. The fundamental equation for stress measurement is developed with the matrix transformation defined for the two-dimensional diffraction. Stresses in a sample distort the diffraction cone shape so that $2\theta$ becomes a function of $\gamma$, $2\theta = 2\theta(\gamma)$, this function is uniquely determined by the stress tensor and the sample orientation. The fundamental equation for stress measurement using 2D detector is given [1] as

$$p_{11}\sigma_{11} + p_{12}\sigma_{12} + p_{13}\sigma_{13} + p_{22}\sigma_{22} + p_{23}\sigma_{23} + p_{33}\sigma_{33} = \ln\left(\frac{\sin\theta_0}{\sin\theta}\right)$$  \hspace{1cm} (1)

where

$$p_{ij} = \begin{cases} \left(\frac{1}{E}\right)((1 + \nu) f_{ij} - \nu) = \frac{1}{2} S_2 f_{ij} + S_1 & \text{if } i = j \\ \left(\frac{1}{E}\right)(1 + \nu) f_{ij} = \frac{1}{2} S_2 f_{ij} & \text{if } i \neq j \end{cases}$$

with

$$f_{11} = h_1^2, \quad f_{12} = 2h_1h_2, \quad f_{22} = h_2^2, \quad f_{13} = 2h_1h_3, \quad f_{23} = 2h_2h_3, \quad f_{33} = h_3^2$$

and

$$h_1 = \sin\theta(\sin\phi\sin\psi\sin\omega + \cos\phi\cos\omega) + \cos\theta\cos\gamma\sin\phi\cos\psi$$
$$- \cos\theta\sin\gamma(\sin\phi\sin\psi\cos\omega - \cos\phi\sin\omega)$$
$$h_2 = -\sin\theta(\cos\phi\sin\psi\sin\omega - \sin\phi\cos\omega) - \cos\theta\cos\gamma\cos\phi\cos\psi$$
$$+ \cos\theta\sin\gamma(\cos\phi\sin\psi\cos\omega + \sin\phi\sin\omega)$$
$$h_3 = \sin\theta\cos\psi\sin\omega - \cos\theta\sin\gamma\cos\psi\cos\omega - \cos\theta\cos\gamma\sin\psi$$  \hspace{1cm} (2)
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where \( f_{ij} \)'s are the strain coefficients determined by \( \{h_1, h_2, h_3\} \) which are components of the unit vector of the diffraction vector \( H_{hkl} \) expressed in the sample coordinates. And \( \ln(\sin \theta_0 / \sin \theta) \) represents the diffraction cone distortion at a particular \((\gamma, 2\theta)\) position. For isotropic materials, there are only two independent elastic constants, Young’s modulus \( E \) and Poisson’s ratio \( \nu \) or the macroscopic elastic constants \( S_1 = (1 + \nu) / E \) and \( S_2 = -\nu / E \). The stress tensor can be determined by fitting the data points to equation (1) with the least-squares method.

For biaxial stress state with only in-plane stress components, corresponding to the straight line of \( d \) vs. \( \sin^2 \psi \) plot in conventional method, we have

\[
p_{11}\sigma_{11} + p_{12}\sigma_{12} + p_{22}\sigma_{22} + \frac{1 - 2\nu}{E}\sigma_{ph} = \ln\left(\frac{\sin \theta_0}{\sin \theta}\right)
\]

where \( \sigma_{ph} \) is the pseudo hydrostatic stress component caused by the error initial values of stress free \( 2\theta \). For biaxial stress state with out-of-plane shear stress components, the case corresponding to the \( \psi \)-split in the conventional method, the equation becomes

\[
p_{11}\sigma_{11} + p_{12}\sigma_{12} + p_{22}\sigma_{22} + p_{13}\sigma_{13} + p_{23}\sigma_{23} + \frac{1 - 2\nu}{E}\sigma_{ph} = \ln\left(\frac{\sin \theta_0}{\sin \theta}\right)
\]

For multiple (hkl) rings, it is possible to solve the stress by putting the linear equations generated from multiple rings into one group, and solved by a single least square calculation. Due the increased sampling, the statistical error will be reduced. There are also many cases, such as thin films, or samples with strong texture, where multiple lines are necessary to get more accurate stress measurement. When multiple (hkl) are used for the stress measurement...
all the linear equations for all the stress state are the same as with single (hkl) except different 2θ values for each (hkl) rings. Figure 1 shows a sketch of frame with multiple (hkl) lines, 2θ1, 2θ2 and 2θn corresponds to the diffraction rings from the crystalline planes (h1, k1, l1), (h2, k2, l2) and (hn, kn, ln) respectively. When equation (1) or the derived equations (3) and (4) are used for stress calculation with multiple (hkl) lines, we use

\[ \ln \left( \frac{\sin \theta_{01}}{\sin \theta_1} \right) \text{ to replace } \ln \left( \frac{\sin \theta}{\sin \theta_0} \right), \]

where 01 is from the stress free value for the (h1, k1, l1) ring. The linear equations are then generated for each (hkl) rings at each data points and all linear equations are solved together by least square method. When multiple (hkl) lines are used for stress calculation, it is the most critical that the lattice symmetry and elastic anisotropy be consistent for all (hkl) lines. For example, in biaxial stress condition, the error of initial d0 (or 2θ0) values for each lines should only create pseudo-hydrostatic (dilation or contraction) deformation, not a distortion. In order to maintain the same pseudo hydrostatic stress component in the calculation, the initial stress free values for all (hkl)’s must be consistent to the crystal lattice structure. For instance, for cubic crystal structure,

\[ \frac{\sin \theta_{01}}{\sqrt{h_1^2 + k_1^2 + l_1^2}} = \frac{\sin \theta_{02}}{\sqrt{h_2^2 + k_2^2 + l_2^2}} = \frac{\sin \theta_{0n}}{\sqrt{h_n^2 + k_n^2 + l_n^2}} \]  \tag{5}

The anisotropic effect on the elastic constants should also be considered for each (hkl) rings. The anisotropy effect can be included in the x-ray elastic constants \( \frac{1}{2} S_2(hkl) \) and \( S_1(hkl) \) to replace the macroscopic elastic constants \( \frac{1}{2} S_2 \) and \( S_1 \). The equations for calculating x-ray elastic constants are:

\[ \frac{1}{2} S_2(hkl) = \frac{1}{2} S_2[1 + 3(0.2 - \Gamma(hkl)\Delta)] \]
\[ S_1(hkl) = S_1 - \frac{1}{2} S_2[0.2 - \Gamma(hkl)\Delta] \]

with \( \Gamma(hkl) = \frac{h^2k^2 + k^2l^2 + l^2h^2}{(h^2 + k^2 + l^2)^2} \) and \( \Delta = \frac{5(A_{RX} - 1)}{3 + 2A_{RX}} \)  \tag{6}

Experimental Results

An example is given by stress measurement on a 1 µm textured Cu film on a proprietary substrate using 1.4Å synchrotron beam and CCD detector. The stress calculations with single (331) or (420) peak and combined analysis with both peaks are compared. Figure 2 is a diffraction frame collected in 30 seconds with the sample mounted on a loading
stage. The data frames are collected at various levels of loading. Two frames were collected for each stress measurement at $\omega = 106.1^\circ$ and $79.5^\circ$. The frame contains two diffraction rings (331) and (420). The macroscopic elastic constants for Cu are $E = 129,800$ MPa and $\nu = 0.343$, and the anisotropic factor is $A_{RX} = 1.09$ [3]. The anisotropic elastic constants, calculated with equation (6), are $S_1 = -2.576 \times 10^{-6}$/MPa and $\frac{1}{2}S_2 = 1.015 \times 10^{-5}$/MPa for (331) and $S_1 = -2.678 \times 10^{-6}$/MPa and $\frac{1}{2}S_2 = 1.045 \times 10^{-5}$/MPa for (420). Figure 3 shows the data analysis setting for both (331) and (420) with LEPTOS software. The integration range of (331) is displayed. The range for (420) is the same except a different $2\theta$ range of $118^\circ$-$122^\circ$.

![GADDS: General Area Detector Diffraction System](image)

Figure 2. Diffraction frame of Cu film containing (331) and (420) rings.

Figure 4 shows the stress values measured at various loading strains. The thin solid line is measured from (331) rings only, the dashed line from (420) rings only, and the thick solid line is from both (331) and (420) rings. The standard deviations are displayed in the corresponding colors with the scale on the right. The three lines follow the same trend with increasing loading. The discrepancy between (331) and (420) are most likely from anisotropic elasticity, even though the anisotropic factor of 1.09 has been introduced. The standard error for the (331)+(420) case is relatively smaller than for (331) or (420) only. Considering the least squares regression for the combined (331)+(420) data points has to cover the systematic discrepancy between the two rings,
the statistical error for (331)+(420) is actually much smaller than either (331) or (420). The combined (331)+(420) data points show that the linear relationship between the stress and loading strain is maintained up to approximately 500 MPa which is slightly higher than the yield strength (471 MPa) given by the empirical equation obtained from a series of stress-strain curves of Cu thin films of various thickness on polyimide substrate measured by micro-tensile tester [4]. The discrepancy is likely due to that the Young modulus of the thin film is typically about 20% lower than the value of single crystal Cu. The residual stress without loading was not measured, but can be estimated by the extrapolation as 320 MPa.

**Figure 3. Data integration region and stress calculation settings with LEPTOS software (Bruker AXS).**

**Conclusions**

Since the diffraction frames collected with area detectors typically contains more than one diffraction ring, the stress analysis can be calculated from diffraction rings from multiple (hkl) crystalline planes. First, this will increase the available data points for stress calculation and so to improve the sampling statistics. Secondly, due to the different orientation distributions from different (hkl) planes, the weak diffraction signal
from one (hkl) plane is most likely to be compensated by a strong diffraction signal from another (hkl) plane. The stress results contain less impact from preferred orientation and anisotropic elasticity. By using multiple diffraction rings, it is also possible to reduce the number of sample tilt angles without reduce the angular coverage.

Figure 4. Stresses of Cu film are measured at various loading strains.

References