RESEARCH PAPERS


A Correction of the Seemann-Bohlin Method for Stress Measurements in Thin Films

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Abstract

The deviation of X-ray data measured by the Seemann-Bohlin method from the predicted linear relationship is studied. A linear $d$ versus $\sin^2 \psi$ relation can be achieved by a modification of the theory based on an average of the Voigt and Reuss models. However, because the Seemann-Bohlin method has to use low $2\theta$ values, it is intrinsically less accurate than the Bragg-Brentano method, particularly for low stress values.

1. Introduction

The commonly used Bragg-Brentano (BB) method for X-ray stress measurement detects the lattice displacements of the same crystallographic planes at different specimen orientations. For it to be accurate, the incident angle of the X-ray beam should be as high as possible so that the penetration is deep and varies with the change in the azimuthal angle. When this method is employed on a material with a sharp stress gradient, the deviation of the experimental results from the linear $d$ versus $\sin^2 \psi$ relationship will be large (Valvoda et al., 1990). This problem can be solved by using the Seemann-Bohlin (SB) method, which measures the lattice displacements of different crystallographic planes at different specimen orientations while keeping incident angle constant. However, the SB method introduces errors as a result of elastic anisotropy, i.e., different crystallographic planes have different elastic constants. In this paper, an improvement on the SB method is derived based on an average of the Voigt and Reuss models, and the residual stresses of PVD TiN thin films are obtained.

2. Theory for the SB method and its problem

Fig. 1 shows the geometry of SB diffraction. $f$ is the anode and $M$ the monochromator. The X-ray beam is focused at $f'$ and hits the sample at a fixed glancing angle $\gamma$. $f''$, $f'''$, ... are different peak positions corresponding to the diffraction angles $2\theta''$ and $2\theta'''$. The $\psi$ angle is

$$\psi = \theta - \gamma.$$  (1)

From (1) and the penetration-depth equation for the $\Omega$ goniometer (Noyan & Cohen, 1987), the penetration depth for the SB method is

$$\tau = \sin (2\theta - \gamma) \sin \gamma / \mu [\sin (2\theta - \gamma) + \sin \gamma].$$  (2)

All of the measured $d(hkl)$ lattice spacings are converted to the lattice parameter $d(100) = d$ and the stress is determined from the slope of the $d$ versus $\sin^2 \psi$ plot.

The advantages of the SB method are as follows:

1. For the BB method, the X-ray beam usually penetrates through the film and the diffraction peaks of the substrate appear on the record. However, in the SB method, the substrate peaks for a 5 μm-thick TiN film will not show up at $\gamma = 4^\circ$ and can only be found at $\gamma = 10^\circ$ (Valvoda et al., 1990).

2. The change of the penetration depth with the angle $\psi$ is large for the BB method but small for the SB method, as shown in Fig. 2. So the weighted average stress value in the surface layer can be obtained. If the film is thick, the stress distribution can be determined by the layer-removal method.

Fig. 1. Diffraction geometry of Seemann-Bohlin method.
When the sample is illuminated by the X-ray beam at a glancing angle \( \gamma \), the effective diffraction length is increased by a factor of \( 1/\sin \gamma \) and results in a correspondingly higher diffraction intensity.

The problem for the SB method is the errors induced by the flat surface of the sample and its displacement. A correction for the flat surface has been given by Haase (1985) and Rafaja & Valvoda (1991). The error caused by the sample displacement can be corrected following the method of Valvoda et al. (1990), who suggested the use of a calibration powder slightly dispersed on the sample surface as an internal standard. However, even after the errors mentioned above are corrected, a large error in the lattice parameters still exists, especially for low \( hkl \) Miller indices, as shown in Fig. 3 (Valvoda et al., 1990). This phenomenon has been recognized and addressed by Perry, Sartwell et al. (1992) and Perry, Valvoda & Rafaja (1992). They use the \( a_{\psi} \) versus \( \sin^2 \psi \) plot instead of the \( a_{\psi} \) versus \( \sin^2 \psi \) plot and find that it gives a straight line. When the compliance values \( S_1 \) and \( S_2 \) for each \( hkl \) reflection are known, this method can give good accuracy. But values of the compliances are not always available, especially for new materials. In this article, we deal with the deviation of the \( a_{\psi} \) versus \( \sin^2 \psi \) plot using an average of the Reuss and Voigt models and derive all the requisite information from the \( a_{\psi} \) versus \( \sin^2 \psi \) plot itself.

3. Correction of the SB method for elastic anisotropy

The strain–stress relation under a biaxial stress state is expressed as

\[
\varepsilon_{\phi\phi} = [(1 + \nu)/E] \sigma_{\phi} \sin^2 \psi - (\nu/E)(\sigma_1 + \sigma_2). \tag{3}
\]

For the BB method, the diffraction planes are fixed, \( (1 + \nu)/E \) is constant and \( \varepsilon_{\phi\phi} \) versus \( \sin^2 \psi \) is linear. When different diffraction planes are used for the SB method, the X-ray elastic constants (XECs) are related to the planes, i.e. \( \varepsilon_{\phi\phi} \) and \( (1 + \nu)/E \) are functions of \( hkl \). Now, the question is how to calculate a stress value for the strains of different planes for which the elastic constants are varied.

The XECs of a polycrystalline sample can be calculated from those of a single crystal. There are three models for calculation.

The Voigt constant-strain model assumes that the strains in all grains are equal and the XECs for a cubic material are expressed by

\[
[(1 + \nu)/E]_V = \frac{S_0^0 - S_0^1}{5/(2C_{11} - 2C_{12} + 6C_{44})}
\]

\[
(v/E)_V = \frac{(C_{11} + 4C_{12} - 2C_{44})}{[(C_{11} + 2C_{12})(2C_{11} - 2C_{12} + 6C_{44})].
\tag{4}
\]

The Reuss model assumes that the stresses in all the grains are equal to the average stress applied to the material. The XECs developed from this model are

\[
[(1 + \nu)/E]_R = \frac{S_{11} - S_{12}}{S_{12} - S_{12} - 3S_{ij}},
\]

\[
(v/E)_R = -S_{12} - \frac{S_{ij}}{S_{12}} \Gamma;
\tag{5}
\]

here,

\[
S_0 = S_{11} - S_{12} - \frac{1}{2}S_{44},
\]

\[
\Gamma = (h^2k^2 + k^2l^2 + l^2h^2)/(h^2 + k^2 + l^2)^2,
\]

\( hkl \) are the Miller indices of the planes and \( S_{ij} \) and \( C_{ij} \)

![Fig. 2. Penetration depth of Cu K\alpha in TiN film with an \( \Omega \) goniometer and the SB method.](image)

![Fig. 3. Data for the SB method of Kuzel. \( \psi^* \) is the strain-free angle (Valvoda et al., 1990).](image)
are the compliance and stiffness moduli, respectively, of the tested material in the single-crystal state.

The Kröner model results in a weighted average of the Reuss and Voigt models. For iron, the Voigt:Reuss ratio = 0.583:0.417 (Japanese Material Association, 1981). For convenience, we take 1:1 as the average of the Voigt and Reuss models. The experiments indicate that this average is closer to reality than either of the two models individually.

Now we derive the SB stress-measurement method from the average of the Voigt and Reuss models.

The XECs under the average of the Voigt and Reuss models can be expressed as

\[
\left(1 + \frac{v}{E}\right)_{R+V}^{hkl} = \frac{1}{2} \left(\left(1 + \frac{v}{E}\right)_{R}^{hkl} + \left(1 + \frac{v}{E}\right)_{V}^{hkl}\right)
\]

The values of \(hkl\) and 20 for TiN with Cu K\(\alpha\) radiation

<table>
<thead>
<tr>
<th>(hkl)</th>
<th>111</th>
<th>200</th>
<th>220</th>
<th>311</th>
<th>322</th>
<th>400</th>
<th>331</th>
<th>420</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 (°)</td>
<td>36.8</td>
<td>42.6</td>
<td>62.0</td>
<td>74.2</td>
<td>78.1</td>
<td>94.6</td>
<td>104.8</td>
<td>108.7</td>
</tr>
</tbody>
</table>

With each \(\varepsilon_{R+V}^{hkl}/\varepsilon_{R+V}^{hkl}\), the strains \(\varepsilon_{q\phi}\) of every \(hkl\) after normalization are obtained. Thus, the stress value is calculated with the slope of the \(\varepsilon_{q\phi}\) versus \(\sin^2\psi\) curve.

So far, the Poisson ration \(v\) is taken as a single value. It can be proved that, regardless of the value of \(v\) in an isotropic material, the same stress value can be obtained after elastic normalization (Yu, 1992).

### 4. Experiments and discussion

All samples used in these experiments are PVD TiN films on high-speed steel. Because of the limitations of our equipment, for TiN film with Cu K\(\alpha\) radiation, only eight peaks are recorded, as shown in Table 1.

The instrumental errors were corrected using fine silicon powder dispersed on the sample surface and the diffraction-peak positions of silicon and TiN were measured. The peaks of silicon are sharp and their 20 values are taken as the peak point. The peaks of TiN, on the other hand, are broad and the 20 values were determined by the full width at half-maximum (FWHM) method. The stress-free lattice spacing for TiN is assumed to be 0.4240 nm. (ICDD Powder Diffraction File no. 6-0642.) The normalization of the \(d\) versus \(\sin^2\psi\) plot is carried out as shown above. The stress values of all samples are calculated with the XEC of the (100) plane obtained with (6) using the experimental XEC value of the (422) plane (Yu, 1992). The final results are listed in Table 2. All samples were also measured using the Bragg–Brentano method for comparison. But, because the plots measured by the BB method for samples 3 to 6 are either curved or oscillated at large amplitude, as mentioned by Perry (1990), we do not trust the stress values derived from data of this kind.

Figs. 4 and 5 show the original and corrected results of two samples measured by the SB method. At some values of \(\sin^2\psi\), the correction had no effect as the two data points overlapped. It shows that the corrected data are generally linear. The stress values of samples no. 1 and no. 3 are shown in Table 2.

Fig. 6 shows the correction of the Fig. 3 data that were presented by Valvoda et al. (1990). The data at \(\sin^2\psi = 0.761\) are a combination of the (333) and (511) sets of crystallographic planes and so cannot be corrected. The stress value for Fig. 6 is also listed in Table 2 as that for sample no. 7. The correction shown in Fig. 6 indicates that the deviation at low diffraction angles has been greatly reduced, yet the stress value only changes a little in comparison with the original one.
Table 2. The stress values and their deviations

All specimens are PVD TiN film coated.

<table>
<thead>
<tr>
<th>Specimen no.</th>
<th>Stress (GPa)</th>
<th>Mean square deviation (GPa)</th>
<th>Correlation coefficient</th>
<th>Corrected Stress (GPa)</th>
<th>Mean square deviation (GPa)</th>
<th>Correlation coefficient</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>-8.8</td>
<td>1.2</td>
<td>0.8559</td>
<td>-7.8</td>
<td>0.4</td>
<td>0.9793</td>
</tr>
<tr>
<td>2</td>
<td>-10.0</td>
<td>1.3</td>
<td>0.8686</td>
<td>-8.1</td>
<td>0.3</td>
<td>0.9856</td>
</tr>
<tr>
<td>3</td>
<td>-8.1</td>
<td>1.0</td>
<td>0.8471</td>
<td>-6.4</td>
<td>0.2</td>
<td>0.9908</td>
</tr>
<tr>
<td>4</td>
<td>-8.1</td>
<td>1.0</td>
<td>0.8471</td>
<td>-7.1</td>
<td>0.1</td>
<td>0.9989</td>
</tr>
<tr>
<td>5</td>
<td>-5.1</td>
<td>0.66</td>
<td>0.8508</td>
<td>-4.3</td>
<td>0.1</td>
<td>0.9917</td>
</tr>
<tr>
<td>6</td>
<td>-8.5</td>
<td>1.4</td>
<td>0.7979</td>
<td>-7.5</td>
<td>0.3</td>
<td>0.9850</td>
</tr>
<tr>
<td>7</td>
<td>-7.1</td>
<td>1.2</td>
<td>0.7990</td>
<td>-6.5</td>
<td>0.2</td>
<td>0.9923</td>
</tr>
<tr>
<td>3'</td>
<td>-8.1</td>
<td>1.0</td>
<td>0.8471</td>
<td>-7.1</td>
<td>0.1</td>
<td>0.9989</td>
</tr>
</tbody>
</table>

From the results above, it is shown that elastic anisotropy of TiN contributes most to the non-linearity of the data. The data can be linearized by the corrected equations based on the average of the Voigt and Reuss models.

In the modification above, the $\nu$ value is assumed to be $1/3$, i.e. the $d_{\phi\phi}$ value equals $d_0$ at $\psi^*$ and $\sin^2 \psi^* = 0.5$. In fact, the Poisson ratio of TiN obtained by Perry is found to be 0.3 by a mechanical method and 0.25 by X-ray diffraction (Perry, 1990).

In order to estimate the errors induced by the uncertainty in the Poisson ratio $\nu$, a sample shown in Fig. 5 is analysed as below:

Suppose the X-ray Poisson ratio is 0.2 and $\sin^2 \psi^* = 2\nu/(1 + \nu) = 0.33$. $\sin^2 \psi$ for the (400) plane is 0.356, which is close to 0.33, so the (400) plane can be taken as the strain-free plane, i.e. $d_{\phi\phi}^{400} = d_0$ and the strains of other planes are determined by this plane, as shown in Fig. 7. Comparison of Fig. 5 with
Fig. 7 shows that, even though the $d_0$ value and $\sin^2 \psi^*$ value change with the change of $\nu$, the stress value only deviates by about 10%. The stress values of $\nu$ values 0.33 and 0.2 are shown in Table 2 as those of samples no. 3 and no. 3'.

Because all diffraction peaks are employed for the SB method, the measurement error of the stress should be estimated by the plane with the lowest Miller indices. For TiN, if the error of peak location is $\Delta(2\theta) = \pm 0.02^\circ$, for $2\theta_{111} = 36.8^\circ$, with $|\Delta d/d| = \cotan \theta d\theta$, then

$$|\Delta d/d| = 5.25 \times 10^{-4}, \quad |\Delta \sigma| = 0.4 \text{ GPa}.$$  

But, for the BB method, we take the $2\theta$ value as high as possible. For TiN, if the same error of peak location is taken, $\Delta(2\theta) = \pm 0.02^\circ$ and, for the 422 reflection, $2\theta = 125.8^\circ$. With $|\Delta d/d| = \cotan \theta d\theta$,

$$|\Delta d/d| = 1.786 \times 10^{-4}, \quad |\Delta \sigma| = 0.1 \text{ GPa},$$  

i.e. the minimum error of the SB stress measurement for TiN film is 0.4 GPa and that of the BB method for TiN film is 0.1 GPa.

**Concluding remarks**

The deviation from the linear $d$ versus $\sin^2 \psi$ relation appearing in the Seemann–Bohlin method is predominantly caused by elastic anisotropy and can be corrected by an average of the Voigt and Reuss model. But because of the use of low $hkl$ indices in the measurement, the SB method cannot reach high accuracy in comparison with the Bragg–Brentano method, particularly for low stress values.

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**References**


