X-ray optical thin film deposition and analysis capability at NASA MSFC

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• Optical thin film coating capability at MSFC
  • Deposition system
  • X-ray reflectometer (XRR)
  • A method for the deposition of broadband coatings
• Thin film stress measurement:
  • Ex-situ example
  • In-situ
    • Example: Stress behavior in polycrystalline materials during film growth
    • Current optical methods of in-situ measurement
      • Limitations, sensitivity
    • New method of in-situ stress measurement using fiber optic displacement sensor
      • Two embodiments: circular, cantilever-substrate
      • Sensitivity
      • Repeatability performance
      • Device validation
    • Effect of material interfaces on film stress: Ir/B4C, Ir/Si, Mo/Si, Mo/B4C,…
      • Multilayers to compensate stress in x-ray optical coatings?
        • W/Si example
X-ray optical thin-film coatings

- Single or multilayers thin-films
- Layer thicknesses in the range of tens to hundreds of angstroms (Å)
- Multilayers contain hundreds of layer pairs
- Layer thickness can be numerically designed to elicit a specific spectral response
- Optical performance dominated by interface roughness/diffusion
- Lateral and in-depth thickness gradients enables:
  - collimating and focusing optics
  - bandpass filters, notch filters, etc.
X-ray reflectivity from the multilayer

Modified Fresnel coefficient:

\[ r_{N-j}^s = \frac{\sqrt{n_{N-j}^2(\lambda) - \cos(\theta_0)^2}}{\sqrt{n_{N-j+1}^2(\lambda) - \cos(\theta_0)^2}} \]

\[ r_{N-j}^p = \frac{\left(\frac{n_{N-j}(\lambda)}{n_{N-j+1}(\lambda)}\right)^2}{\sqrt{n_{N-j+1}^2(\lambda) - \cos(\theta_0)^2}} \]

\[ \beta_{N-j} = \frac{2\pi n_{N-j}}{\lambda} \sqrt{1 - \cos(\theta_0)^2} \]

\[ S^V_{j+1} = \frac{r^V_{N-j}}{1 + S^V_j} e^{2i\beta_{N-j}} \]

\[ R = \frac{1}{2} R^s + \frac{1}{2} R^p \]

\[ R^V = S^V S^{V*} \]

\[ d \equiv h_A + h_B \]
Recently developed XRR system at MSFC

The X-ray Reflectometer System

- High flux rotating anode source
  - Cu target 8 keV X-rays
  - 5-35 kV, 10-150 mA
- Series of Tungsten slits to reduce beam size on sample
- High speed silicon drift detector
  - Amptek
  - (Energy res. of 0.14 keV at 5.9 keV)
- Precision sample alignment and positioning
- Custom control + real-time data collection software (LabVIEW)
- Vacuum chuck sample mount
- No monochromator due high detector resolution
- Interlaboratory study conducted to ensure quality of XRR measurements

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Thin Film Stress

- The stress can be compressive or tensile.

- Various components of stress:
  - Intrinsic, $\sigma_i$, which is related to the film’s microstructure.
  - Thermal stress, $\sigma_{\Delta CTE}$, which arises due to the difference in the linear expansion coefficient between the film and substrate and the difference between substrate temperature, $T_s$, during deposition and subsequent cooling to room temperature:
    \[ \sigma_{\Delta CTE} = M_f (\alpha_s - \alpha_f) \Delta T \]
  - Extrinsic, $\sigma_{ext}$, that results due to external forces applied to the film substrate system such as bending of the substrate to produce a figured optic.

- The film stress can be enormous for some materials (i.e. GPa’s)

- The curvature, $\kappa$, of the deformed substrate is proportional to the product of film stress and film thickness, $\sigma h_f$, through a constant that describes the geometric and mechanical properties of the substrate (Stoney’s Equation)--namely, the substrate’s thickness, $h_s$, and biaxial modulus, $\frac{E_s}{(1-\vartheta_s)}$:
  \[ \sigma h_f = \frac{E_s h_s^2}{6(1-\vartheta_s)} \kappa \]
In controlling film stress, the aim is to manipulate the energy of the sputtered atoms and influence the adatom mobility at the film surface.

For a given material, stress is highly process dependent for magnetron sputtering and influenced by deposition conditions:
- Gas Pressure (stress reversal)
- Deposition Rate (cathode power)
- Substrate Temperature
- Substrate bias

There is a trade-off between film stress and film quality (i.e. roughness, density).
- Generally, the deposition conditions needed to achieve good X-ray reflectivity result in high film stress.
The film will delaminate if the stress is greater than the adhesion between the film and substrate.
Detrimental effects of high film stress:

- Cracking, buckling and delamination
  - If the force per unit length due to the stress in the film exceeds the adhesive force, delamination of the film will occur.

  "telephone chord" propagation
  
  High compressive stress (buckling)
  
  High tensile stress (cracking)

- Substrate deformation
  - Of particular concern for grazing incidence X-ray optics since the stress can alter the precise geometrical figure and degrade its focusing or collimating properties.
  - Significant technological challenge for the next generation of lightweight X-ray space telescopes like Lynx:
    - The desire to achieve sub-second resolution has motivated deposition techniques to correct substrate figure errors which rely on a very low stress film (ie. A few MPa)
    - Substrates are only 10’s of microns thick.
    - The X-ray reflective Ir layer is highly stressed (~4 GPa)
Ex-situ measurement of thin film stress

Stoney's Eqn: \( \sigma_f = \frac{E_s h_s^2 \kappa}{6(1 - \nu_s)} \)

Spherical Deformation Mode:
\[
A = \sigma_f \frac{D_s^2}{h_s^3}
\]

\( \kappa_x = 0.0212 \) and \( \kappa_y = 0.0214 \) m\(^{-1}\)

Tallysurf stylus profilometer

\[ \kappa \approx \frac{d^2 w}{d^2 x} = \text{const} \]

Si <111>
Current methods of optical in-situ thin film stress measurement:

Multi-beam stress sensor (MOSS):

These methods determine the substrate curvature by various optical means from which the integrated stress is calculated from the Stoney Eqn.:

$$\sigma h_f = \frac{E_s h_s^2}{6(1 - \nu_s)} \kappa$$

Minimum detectable stress $\Delta\sigma_h$:
- Ranges from 0.5-50 MPa*nm depending on method and substrate (i.e. geometry and mechanical properties)
- MOSS is 50 MPa*nm for 100 µm thick silicon substrate

Draw backs with current optical methods:
- Requires external optical access to the substrate through angled viewports
- Limited to specific deposition geometries
- Complex
- Requires the use of opaque substrates such as crystalline silicon.
- Film side is measured which can result in destructive interference effects when measuring transparent films.
New approach to in-situ stress measurement:

- Utilizes a high resolution (i.e. 5nm) vacuum compatible fiber optic displacement sensor.
- Curvature determined from out-of-plane displacement measurement of the substrate.
- Uses double-side polished substrate.
- Same arrangement can be used for thermal annealing.
- Glass substrates can be utilized.
- Easily implemented into existing deposition systems.
- Very sensitive method.

Stoney Eqn. for circular substrate:

\[ \sigma_f = \frac{4}{3} \frac{E_s}{(1 - v_s)} \left( \frac{h_s}{D_s} \right)^2 \delta \]

Stoney Eqn. for cantilever:

\[ \sigma_f = \frac{E_s h_s^2 \delta}{3(1 - v_s) L^2} \]

Ongoing work:
- Adapting to rotating substrates
- Adapting to curved (i.e. segmented) substrates

Pending publication in Review of Scientific Instruments
Minimum detectable integrated stress, $\Delta(\sigma h_f)$

- The minimum detectable stress is limited by the combined ambient vibrational background of the substrate and electronic noise of the displacement sensor.
- The sensitivity further depends on the mechanical and geometric properties of the substrate.
- The cantilever approach is more sensitive to a given integrated stress but is also more sensitive to vibrational noise—compensating effect.
- The cantilever approach is advantageous because it is flexible in its orientation and easily adapted to various deposition geometries.

Sensitive enough to measure stress in x-ray multilayers

\[
\Delta(\sigma h_f) = \frac{4}{3} \frac{E_s}{(1 - \nu_s)} \left(\frac{h_s}{D_s}\right)^2 \Delta \delta
\]

$15 \text{ MPa*nm}$

\[
\Delta(\sigma h_f) = \frac{E_s h_s^2 \Delta \delta}{3(1 - \nu_s)L^2}
\]

$9 \text{ MPa*nm}$
Stress evolution in polycrystalline films

Depends on:
- Substrate temperature
- Argon pressure
- Mass of sputtered atoms
- Substrate bias
- Surface energy

Nucleation & island growth

Type I
- high $T_m$
- low atomic mobility

Type II
- low $T_m$
- high atomic mobility

Volmer-Weber Growth Mode

Island coalescence

Force per unit width

Depends on:
- Nucleation and growth
- Growth mode

Surface roughness increases with film thickness

Low surface roughness

Anneal

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Stress reversal in polycrystalline films (steady state)

Results are consistent with D.W. Hoffman, Internal stress of sputtered Chromium, Thin Solid Films, 40 (1977) 355-363

Instantaneous stress:

\[ \frac{d(\sigma h_f)}{dh_f} = -\frac{E_s}{3\xi(1-\vartheta_s)} \left( \frac{h_s}{r} \right)^2 \frac{d\delta}{dt} \]

- Efficient for parametrizing the stress
- Independent of film thickness in the steady-state regime of film growth
- Substrate in thermal equilibrium

 Scaling of the critical pressure with film density

- State of stress (i.e. tensile or compressive) at low pressure is strongly influenced by substrate temperature for low density metals like chromium.
- Therefore, the state of stress will depend on the heat transfer mechanisms of the substrate (i.e. how it is mounted) for a given deposition system.

\[ T_s \sim 70^\circ C \]
Stress reversal in Cr with argon pressure has been measured with the instrument. Consistent with the previous work of Hoffman (i.e. stress reversal).

Measurement sensitivity is better than resolution in the control of Argon pressure.
Reduction of Ir film stress with argon pressure optimization

- Surface roughness is too high at the argon critical pressure of ~22 mTorr.
- We can achieve low stress and lower argon pressure by exploiting the zero stress that occurs shortly after island coalescence.
- The use of in-situ stress measurement allows use to tune the stress to within a few MPa.
Near-zero stress in Iridium (15.0 mTorr)

Reduction in the total stress by 3 orders of magnitude (i.e. to -2.89 MPa)

Good adhesion

Promising result: 5Å RMS roughness

Further reduction in the roughness is possible through optimization of Ar pressure
Refined in-situ stress sensor
In-situ stress of single layer thin films

$T_s \sim 27-30^\circ C$, 2.5 mTorr Ar
100µm thick Schott D263

Calibration masses placed on cantilever tip used to validate substrate modulus and linear range of the sensor:

\[
\delta_m = \frac{2mgx^2}{E_s b h^3} (3L - x)
\]

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Device performance

- $\delta, \mu m$
  - $t, \text{sec.}$
  - $-40$ to $-20$
  - $-20$ to $-10$
  - $-10$ to $0$
  - $0$ to $20$
  - $20$ to $40$

- $R, \%$
  - $10^{-7}$ to $10^{-5}$
  - $10^{-5}$ to $10^{-3}$
  - $10^{-3}$ to $10^{-1}$
  - $10^{-1}$ to $10^{0}$

- Model
- Measured

- $\Gamma = 0.38$
- $d = 3.38 \text{nm}$
- $\sigma = 0.40 \text{nm}$

- $\pm 2.5 \%$ run-to-run repeatability
- $\pm 0.5 \%$ within run repeatability
Effect of material interfaces on the film stress (Mo-based)

- Mo on D263
- Mo on B4C
- Mo on Si
- Si on Mo
Effect of material interfaces on the film stress (Ir-based)

![Graphs showing the effect of material interfaces on film stress](image)

- Ir on D263
- Ir on Si
- Ir on B4C
- Si on Ir
- B4C on Ir

**Graphs:**
- Four graphs showing the change in film stress ($\delta$, $\mu m$) over time ($t$, sec.) for different material interfaces.

**Key Observations:**
- The graphs illustrate the stress changes for different interfaces over time.
- The y-axis represents the change in film stress, while the x-axis represents time.

**Notes:**
- The graphs are color-coded to distinguish between different material interfaces.
- The data points and trends are clearly visible, indicating the effect of material interfaces on film stress.
Effect of material interfaces on the film stress (W, Cr-based)

Effect of material interfaces on the film stress (W, Cr-based)
Currently single layer films (i.e. Cr) with tensile stress are used as one technique to compensate the integrated stress in x-ray optical coatings to near-zero:

\[(\sigma h_f)_{Net} = \sigma_A h_A + \sigma_B h_B + (\sigma h)_{CTE} \approx 0\]

The columnar microstructure of metal films in tension results in increasing surface roughness as the film thickness increases—thereby limiting the method’s applicability.

The increased surface roughness can severely degrade the optical coating’s performance; particularly for high energy broadband multilayers.

Multilayers interrupt the columnar growth so roughness doesn’t increase with film thickness (for Glass & Si)

Example:

Stress compensating ML: Cr/B4C, Mo/B4C, …

Future work:
- Influence of substrate temperature and deposition rate.
- Impact to total deposition time.
- Optimization of layer thicknesses.
- Surface roughness characterization.
- Addition of N to B4C based ML’s to increase dep. rate and smooth interfaces.
In-situ stress in W/Si multilayers

$T_s \sim 30^\circ C$
Schott D263
$\xi_W \sim 0.13 \text{ nm/sec}$
$\xi_{Si} \sim 0.18 \text{ nm/sec}$

$d \sim 7.3 \text{ nm}$
$\Gamma \sim 0.80$
$\sigma \sim 0.45 \text{ nm}$
Conclusions

- We have introduced a novel method for the in-situ measurement of film stress using a fiber optic displacement sensor.
- The device is less complex than other current optical methods and easily implemented into an existing deposition system.
- The device’s sensitivity is 0.009 N/m (9 MPa*nm) for a 100 µm thick glass substrate.
- This sensitivity is capable of detecting changes in stress due to small changes in deposition parameters such as argon process pressure (i.e. ±0.02 mTorr).
- The sensitivity can easily detect changes in the integrated stress in the individual layers of multilayer films of sub-nanometer thickness.
- The in-situ stress measured with the device is in good qualitative agreement with the known behavior of metals films (i.e. stress reversal, Volmer-Weber growth).
- We presented the influence of the material interfaces on the evolution of the film stress for several material pairs including: Mo/Si, Mo/B4C, W/Si, Ir/B4C,…
- We have proposed a new stress compensating method that utilizes multilayers
  - This method might be applicable to balance the stress in broadband multilayer that are more than a micron in total thickness.
  - More investigation is needed to study the impact to the total deposition time through optimization of the layer thicknesses