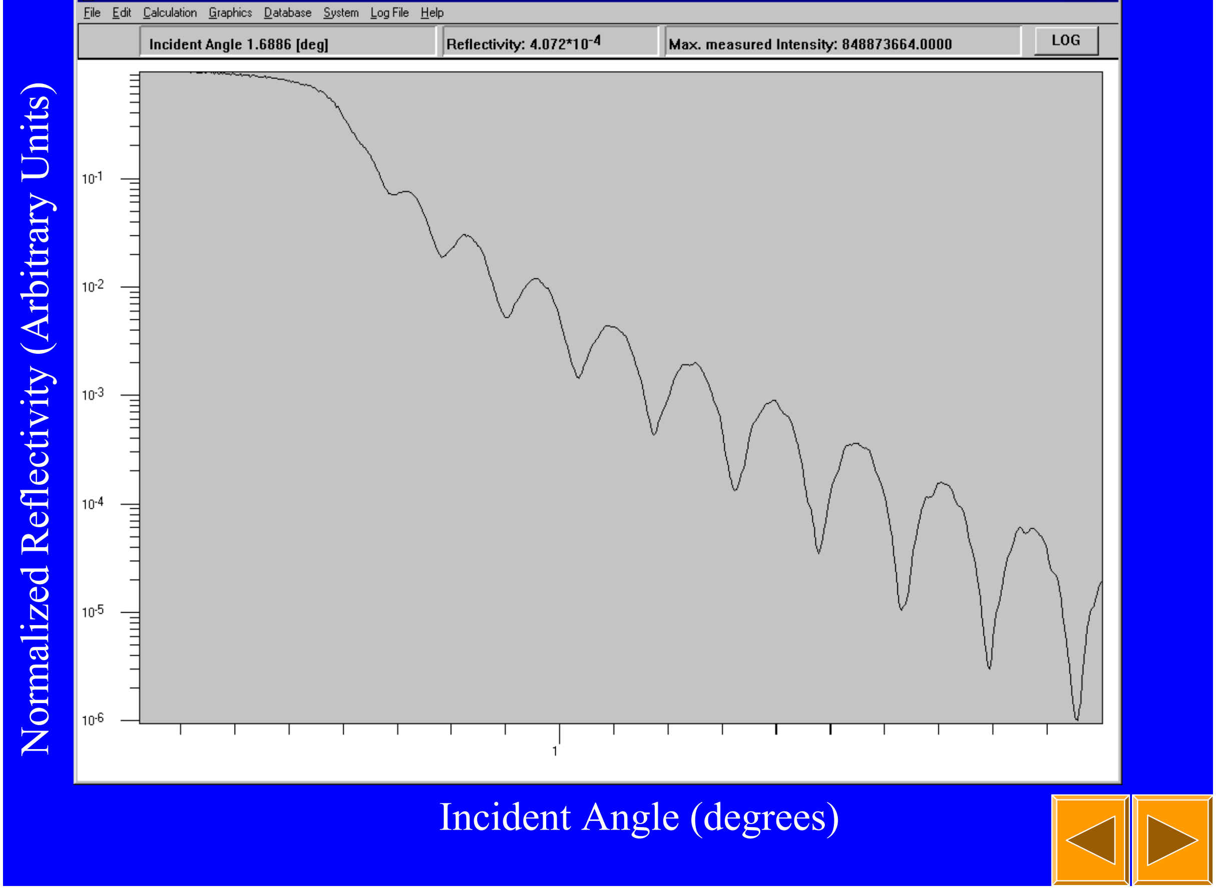
Reflection

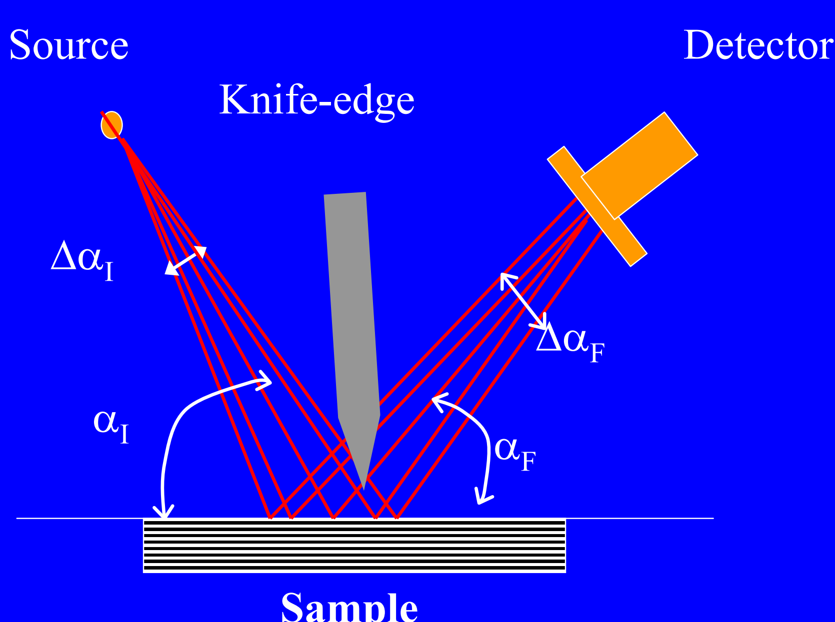
- Specular or mirror like reflection from smooth surfaces
- Diffuse reflection from rough surfaces

Specular X-ray Reflectivity

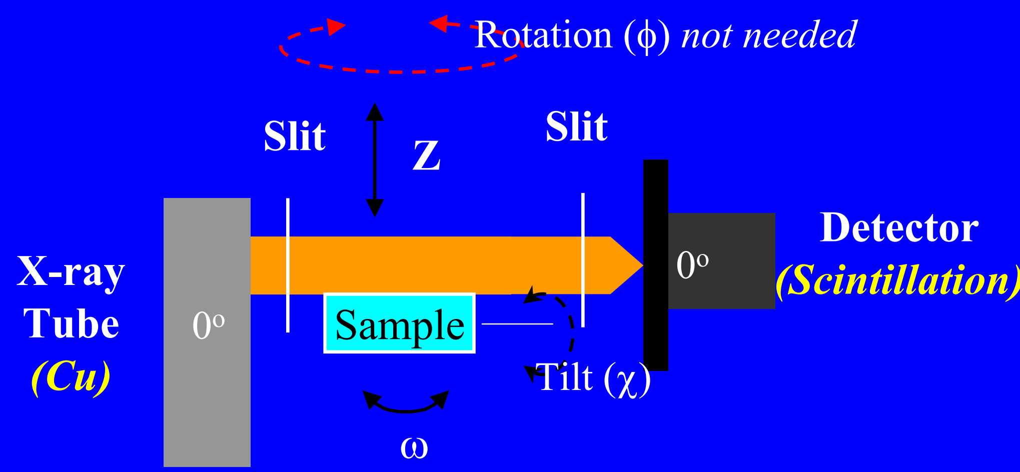
- A non-destructive, routine technique, used for estimation of density, thickness and roughness of thin film structures (single layer and multi-layered)
- Based on total external reflection of X-rays from surfaces and interfaces
- Can be used with amorphous, crystalline and liquid samples
- Used for typical layer thickness between 5 Å and 400 nm and surface roughness from 0 to 20 Å
- *This technique does not work effectively if there is no difference between the electron density of different layers or layer and substrate*



Typical Measurement Setup



Alignment

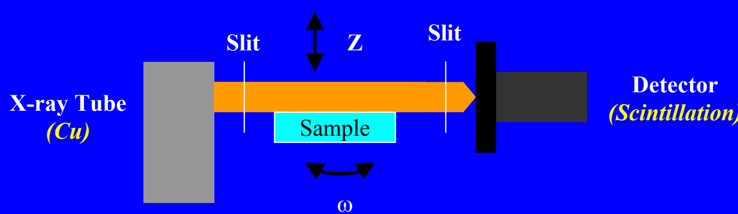


Experimental Setup

- According to α_I^{-4} law of Fresnel reflectivity, the intensity leaving a smooth surface decreases very rapidly on increasing the angle of incidence
- Since XRR requires recording reflected intensity over 5-6 orders of magnitude, highly intense X-ray source and detector with low noise are needed
- In order to measure the angles accurately, thus to minimize error in the results, the rotational axis of the sample circle (ω-circle) has to be aligned exactly with the sample surface. This is accomplished in following steps:

Sample Alignment

- Lateral movement and rocking sample across the primary beam (ω-scan) are iterated until the maximum intensity of ω-scans equals half the intensity of the primary beam, compared with the intensity measured without the sample. With this ω- axis lies on the sample surface and this surface is parallel to the primary beam direction
- The angular position of the sample after the adjustment, however, may not coincide with the zero point of the ω-circle. This is caused by various surface treatments or by the miscut of the sample surface with respect to any crystallographic main axis. The following step is required to redefine the ω-scale.



Sample Alignment: Redefining ω-scale

- To redefine the ω-scale we choose an angle of incidence (α_I) in the range $0< \alpha_I< \alpha_{C;}$ where α_C is the critical angle of incidence for total external reflection of X-rays. Typically this angle is chosen to be 0.2°.
- Angular position of the specularly reflected beam is measured on the detector circle 2θ. The sample surface is also corrected for any tilt (χ-scan). If $2\theta \neq 2\alpha_I$; ω scale is readjusted by ($2\theta/2- \alpha_I$). This procedure may be repeated for different values of α_I to improve the precision of the sample alignment

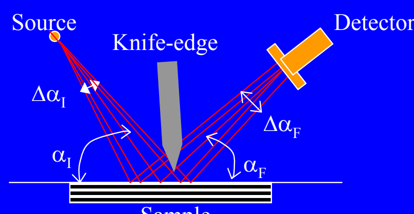
This completes the sample alignment. This procedure is repeated for every sample.

Reflectivity Measurement: Setup

- Reflectivity experiments are optimized in such a way that the specular reflectivity and large features characterizing the sample (Bragg's peak, Kiessig oscillations) appear up to a large value of α_I (~2°).
- Higher angular resolution is needed to separate specular specular from diffuse scattering events. This can be achieved by decreasing angular divergence of incident beam ($\Delta\alpha_I$) and angular acceptance of detector ($\Delta\alpha_F$). In practice low $\Delta\alpha_I$ and ($\Delta\alpha_F$) can be obtained by using narrow slits at incident beam and detector side respectively. However, narrow slits decrease the intensity thus increasing the experimental time.
- To obtain high resolution and good scattered intensity, the following steps are taken

Reflectivity Measurement: Setup

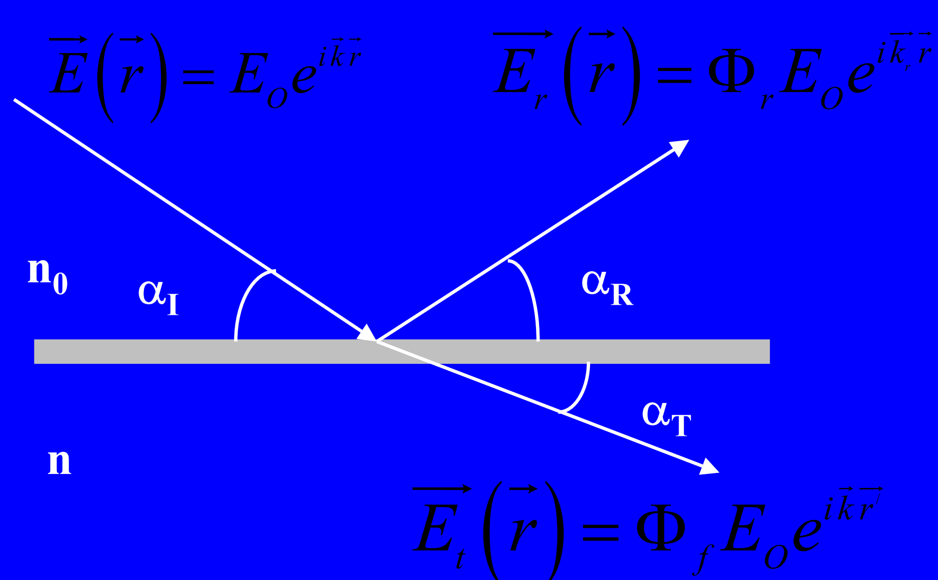
- As a trade-off, higher values $\Delta\alpha_I$ and $\Delta\alpha_F$ (hence wider slits) are used but the irradiated sample area is reduced to achieve sufficient angular resolution.
- In practice, this is achieved by using a knife-edge very close to the axis of sample rotation i.e. to the sample surface.
- Under these conditions only those beams leaving the sample surface directly below the knife-edge arrive at detector.



Reflectivity Measurement

- Different ranges of the reflected curve are selected and recorded under various conditions of angular resolution and counting time. While the measurements near α_C are carried out with highest resolution, angular resolution can be relaxed at higher angles. Typically measurements are setup to have at least 1500 counts for the maxima of each oscillation.
- The specular reflectivity is recorded while running ω-2θ scan, where ω is the angular position of sample circle and 2θ is the angular position of detector. In this scan, α_I and angle of exit (α_F) are changed simultaneously and $\alpha_I = \alpha_F$.
- High resolution measurements with Four-bounce monochromator are typically for films thicker than ~0.5μm

Transmission and Reflection of X-rays



Basic Equations: Density of Single layer

- At X-ray frequencies, the refractive index can be expressed as

$$n = 1 - \delta - i\beta \qquad (1)$$

Where

$$\delta = \left[\frac{\rho_e r_e \lambda^2}{2\Pi}\right] \qquad \beta = \left(\frac{\mu\lambda}{4\Pi}\right)$$

$\delta(\lambda)$ and $\beta(\lambda) \sim 10^{-6}$ and describes the dispersion and absorption terms

- ρ_e = electron density (Z electrons/ atom)
- λ= wavelength of X-ray
- μ=linear absorption coefficient for energies far from X-ray threshold
- r_e= classical electron radius $=e^2/mc^2$

Basic Equations: Density of Single layer

- For specular X-ray reflectivity $\alpha_I = \alpha_R$; angle of incidence is equal to angle of exit.
- Since the real part of index of refraction of materials if less than unity (index of refraction for vacuum) the material is for X-rays less refractive than it is for vacuum.

- According for Snell-Descartes law

$$\frac{\cos\alpha_I}{\cos\alpha_T} = \frac{n}{n_0} \qquad (2)$$

Basic Equations: Density of Single layer

- There is a critical angle of incidence α_C for which the X-rays are totally reflected at the interface, hence α_T =0. Neglecting the absorption in this case, we find

$$n \cong 1-\delta = \frac{\cos\alpha_I}{\cos\alpha_T}$$

- Expanding the cosine for small angles

$$\alpha_c \approx \sqrt{2\delta} = 1.64 * 10^{-3}\sqrt{\rho_m} * \lambda \qquad (3)$$

$$\rho_m = \frac{\rho_e A}{N_A Z}$$

where Z is the atomic number, A is the mass number and N_A is Avogadro's constant

α_C is determined at $R(\alpha_I)$ =R_{max}/2

Basic Equation:Thickness of Single Layer

- Reflectivity of a single layer deposited on a semi-infinite substrate can be expressed as

$$R = \left| \frac{r_1 + r_2 e^{-2ik_{0z}t}}{1 + r_1 r_2 e^{-2ik_{0z}t}} \right|^2 \qquad (4)$$

Where $\mathbf{r_{1,2}}$ are the Fresnel reflectivity coefficients of the free surface and the substrate interface respectively, $\mathbf{k_{0z}}$, is the vertical component of the wave vector of the beam transmitted through the layer and **t** is the layer thickness.

•Intensity maxima occurs whenever **exp(-2ik$_{0z}$t) =1**
i.e. at angle positions α_{im}

Basic Equation:Thickness of Single layer

- Alternatively, for intensity maxima, the path difference between the reflected waves should be an integral multiple of the incident wavelength

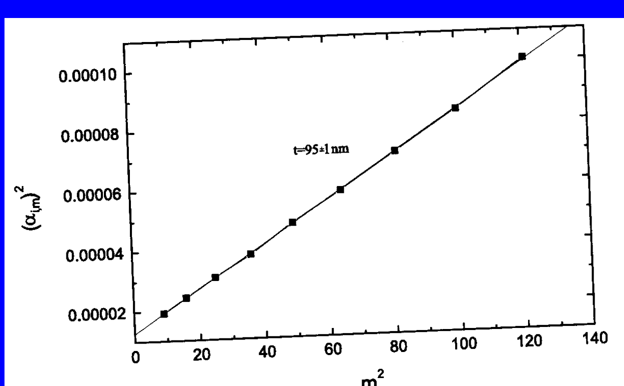
$$2t\sqrt{\sin^2\alpha_{\mathrm{Im}} - \sin^2\alpha_C} = m\lambda \qquad (5)$$

Where “m” is an integer

In most cases, the angle of incidence are small, hence (5) can be expressed as

$$\alpha_{Im}^2 - \alpha_C^2 = m^2\left(\frac{\lambda}{2t}\right)^2 \qquad (6)$$

Thickness Calculation



- Squares of the positions of the intensity maxima versus squares of the Kiessig fringe order “m” is plotted
- The slope of the linear dependence is used to estimate layer thickness “t” while intercept of the line at m=0 is used to determine α_C.

Accuracy in Estimation

- Accuracy in density is defined as

$$\frac{\Delta\rho}{\rho} = 2\left(\frac{\delta\alpha_I}{\alpha_C}\right)$$

Where $\delta\alpha_I$ is the step width of the goniometer

- Accuracy in thickness is defined as

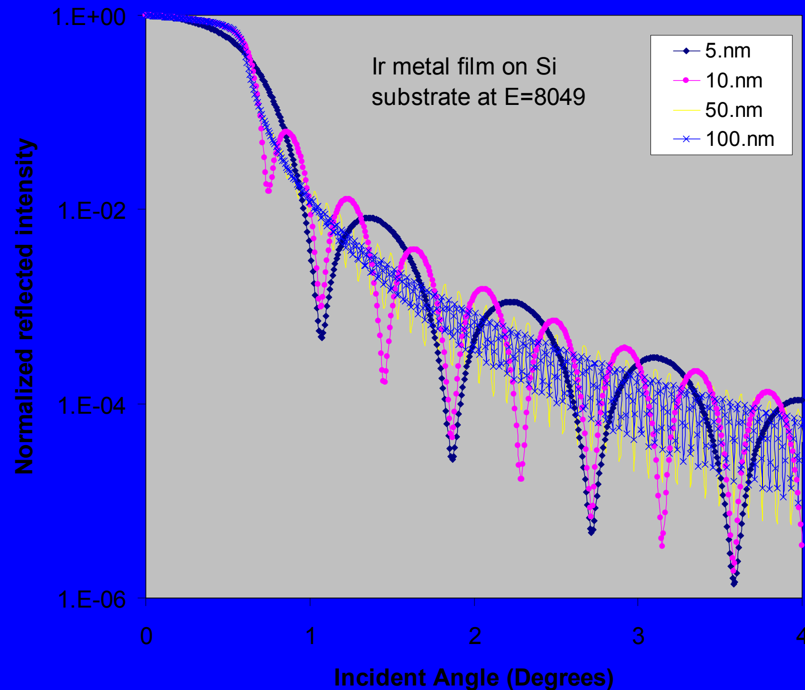
$$\frac{\Delta t}{t} = \left(\frac{\delta\alpha_I}{\alpha_C}\right) \approx \frac{1}{m_{\max}}$$

Where $m_{\max}$ is the largest fringe order that is detected in the reflectivity curve with an accuracy of one-half of a fringe period

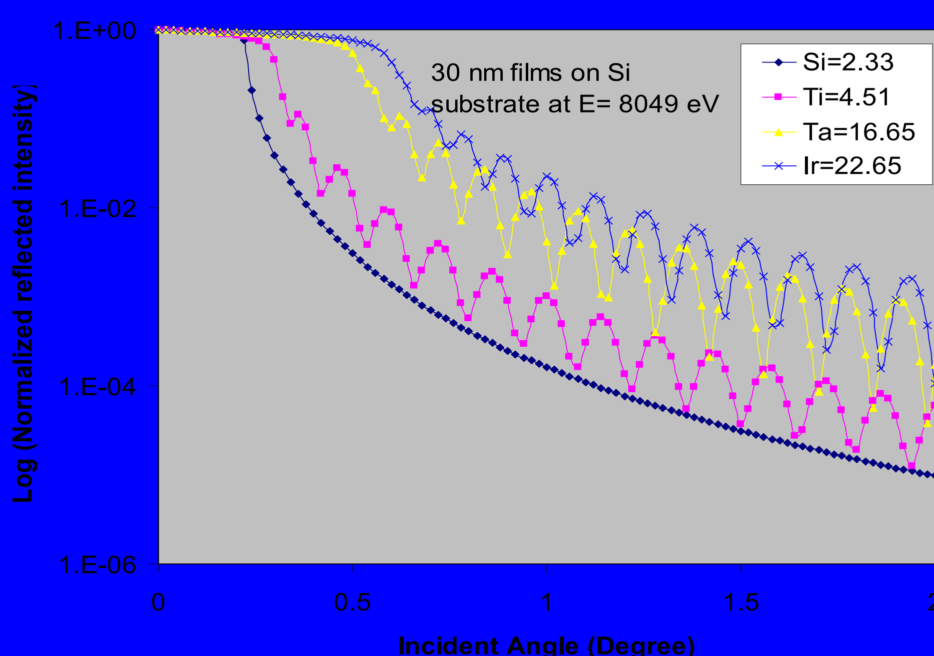
Effect of Roughness

- In real world surfaces/ thin film structures are not perfectly smooth and posses surface and/or interface roughness
- While the presence of surface roughness decreases the specular intensity of the whole curve progressively, interface roughness gives rise to progressive damping of the Kiessig fringes.

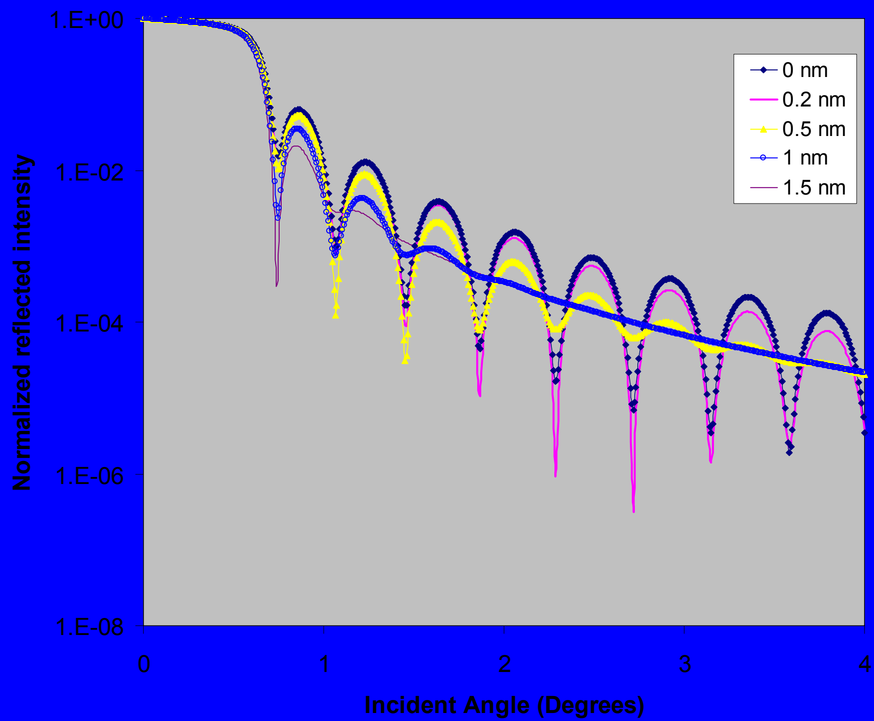
Reflectivity curves for films with different thickness



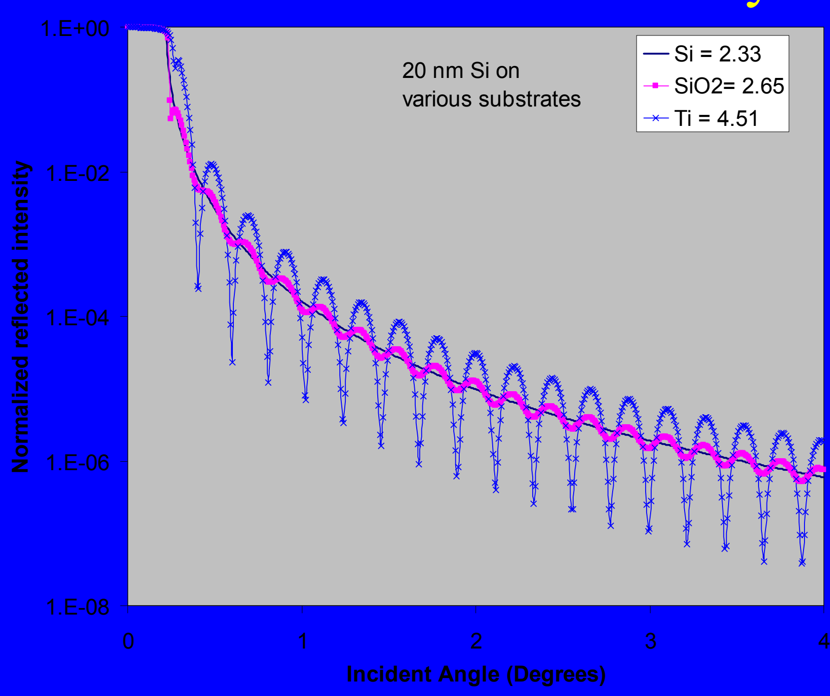
Reflectivity curves for materials with different densities



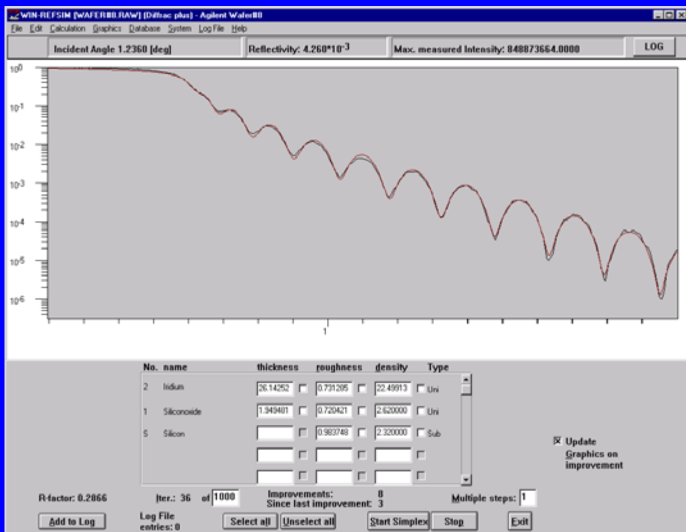
Reflectivity curves for films with different film surface roughness



Reflectivity curves for films with different substrate density



Simulated Results: Ir metal on Si substrate



Red curve= simulated results;
Grey curve = experimental results

- Results are simulated starting with simplest structure; adding complexity as needed
- An attempt is made to have minimum difference between simulated and experimental results.

Suggested Readings

- REFSIM Version 1.2, User's Manual (Bruker AXS)
- High Resolution X-ray Scattering from Thin Films and Multilayers (V. Holy, U. Pietsch, T. Baumbach) Springer Tracts in Modern Physics

Website of Interest

http://cindy.lbl.gov/optical_constants

- X-ray specular reflectivity curves for single and multiple layers
- Estimation of depth penetration as a function of incident angle or energy