Molecular Optics and Thin Films

Danelle E. Brown (deb36@email.byu.edu),
R. Steven Turley (turley@byu.edu),
Brigham Young University

Our studies address microscopic calculations of the optical properties of thin films in the soft x-ray and extreme ultraviolet (EUV) regime when the wavelength is on order of the inter-atomic distances. Investigations include the effects of surface roughness, as well as one-dimensional "amorphous" atomic spacing, on specular and non-specular reflection.

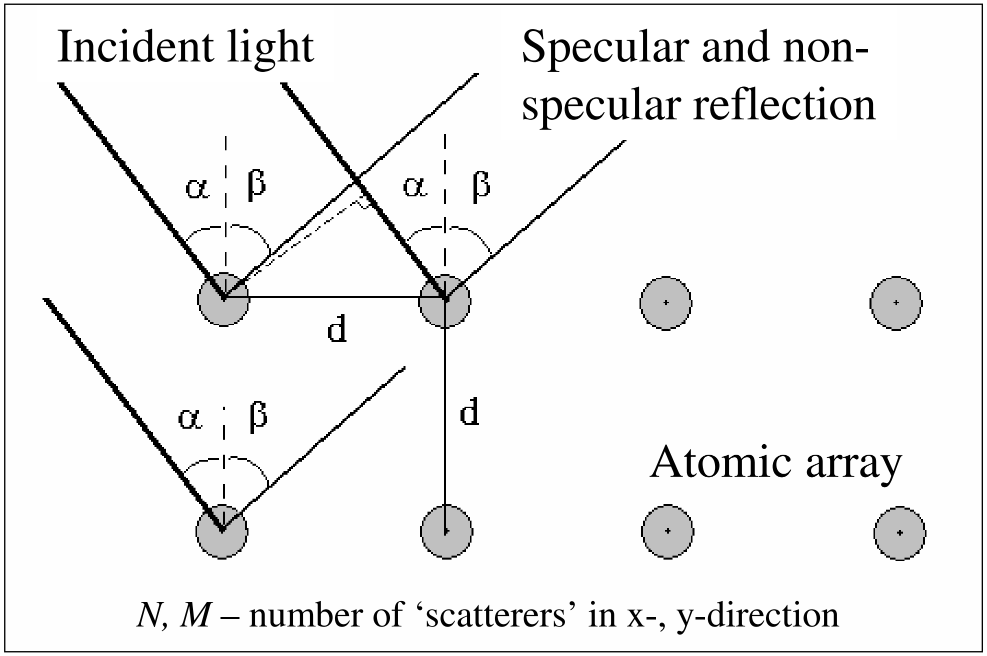
When wavelength gets small…

- When approaching reflection and transmission macroscopically – i.e. when the wavelength of light λ . d, the inter-atomic distance – determining optical properties becomes an optics problem. The interface is, for all practical purposes, "flat."

- When λ is on the same order as d (the EUV and x-ray regime), the interface no longer "looks" flat, but is more correctly treated as an array of scattering atoms, microscopically speaking.

- <u>Our task</u>: Investigate how microscopic atomic properties such as position correlations and scattering amplitudes affect macroscopic reflection
  - The field of Molecular Optics!
  - Side note: BYU's Thin Films research group is experimentally determining optical properties in the EUV regime for UO_x. Could the theoretical microscopic scattering amplitudes be correlated to these experimental results?

Diagrams and Definitions



N, M – number of 'scatterers' in x-, y-direction

The symmetry between the x and y directions has been verified via theory and program, as was expected

Path length difference between two 'atoms'

$$\delta = kd(\sin\alpha - \sin\beta)$$

Expression in 1D, assuming isotropic [s-wave] scattering (including constant scattering probability factor *f*):

$$Field = \sum_{n=0}^{N} f e^{in\delta}$$

$$I(\delta, N, f) = \frac{|f|^2 \cos(\delta N + \delta) - 1}{N \cos(\delta) - 1}$$

Extended to 2D:

$$Field = \sum_{m=0}^{M-1} \left(\frac{e^{im\Delta} f e^{iN\delta}}{e^{i\delta} - 1} - \frac{f e^{im\Delta}}{e^{i\delta} - 1} \right) = f^2 \left(\frac{e^{iN\delta} - 1}{e^{i\delta} - 1} \right) \left(\frac{e^{iM\Delta} - 1}{e^{i\Delta} - 1} \right)$$

looking strangely enough like a separable equation, where

$$\Delta = kd(\cos\alpha + \cos\beta)$$

Dependent Variables

- The path length differences δ and Δ are dependent on atomic spacing d, wave number k, incident angle α, and exit angle β
- That is to say,

$$\delta(d,k,\alpha,\beta) \text{ and } \Delta(d,k,\alpha,\beta),$$

$$\boldsymbol{E}(N,M,d,k,\alpha,\beta) \text{ and } \boldsymbol{I}(N,M,d,k,\alpha,\beta),$$

where $\boldsymbol{I} = |\,\boldsymbol{E}\,|^2$

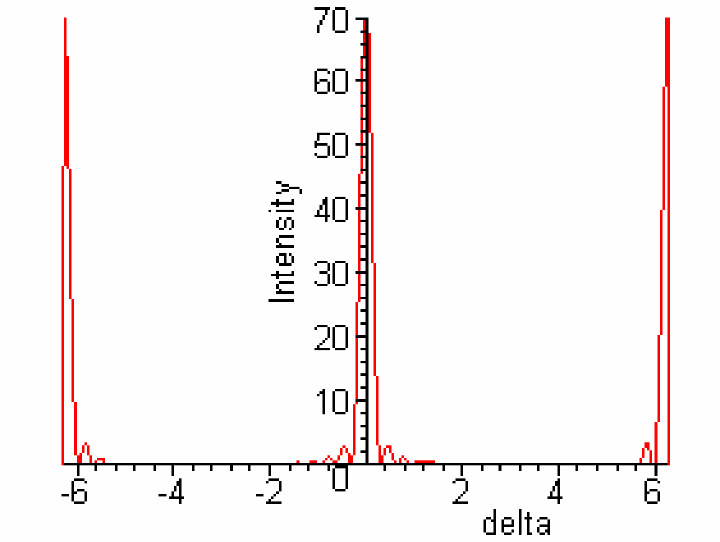
- To more correctly model Δ, an attenuation factor accounting for scattering from each succeeding surface, ξ, was introduced
- For simplicity, lumped into Δ, where $\Delta \rightarrow \Delta + i\xi$
  - As expected, the intensity was affected

Thickness Effects

For two-dimensions:

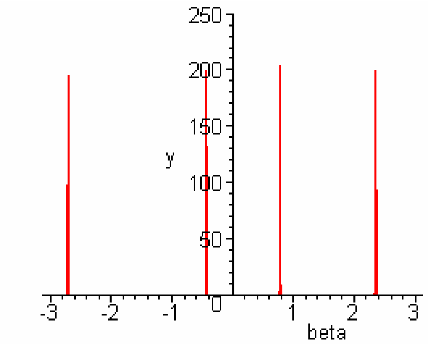
What happens to $I(\delta)$ as M increases?

- Intensity increases to a maximum, then decreases to zero, then increases to a lesser maximum, exhibiting features of a dying sinusoidal exponential
- The first intensity max and min occurs at M=6 and 16, respectively.

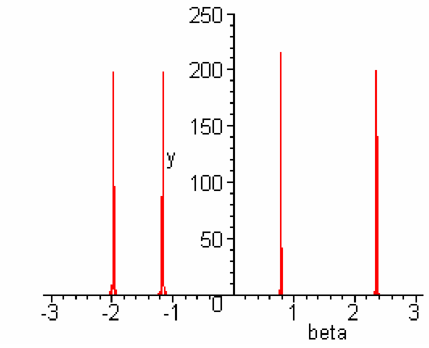


Parameter	Value	Rows (M)	Intensity
N	100	1	20
d in x-dir	135	6	75 (max)
d in y-dir	d_x	16	0
Δ	$\pi/8$	23	22
-	-	32	0
-	-	40	13
-	-	48	0

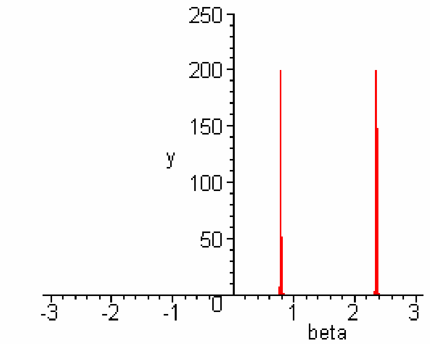
Wavelength Scan



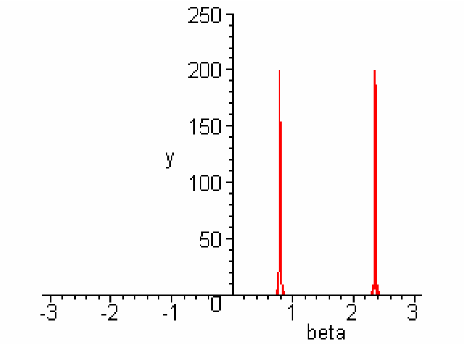
λ=11*d*/8



λ=13*d*/8



λ=14*d*/8



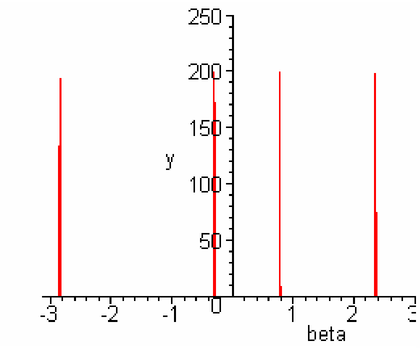
λ=30*d*/8

In each case,
N=200
M=1
d=135
$\alpha=\pi/4$

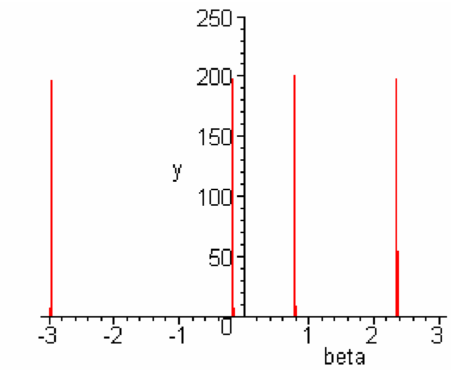
- As λ increases with respect to d, $I(\beta)$ from $-\pi < \beta < \pi$ shifts toward ~1.7 and then diminishes

- There comes a point where, for $\alpha = \pi/4$, β is only $\pi/4$ and $3\pi/4$, where either the incident light follows Snell's Law or passes right through the lattice.

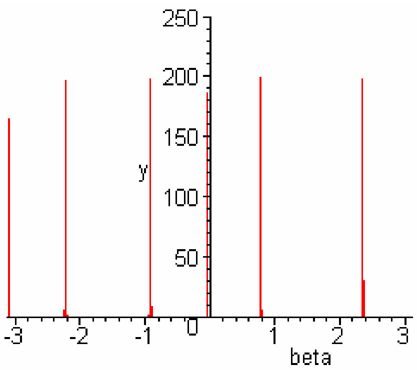
Wavelength Scan



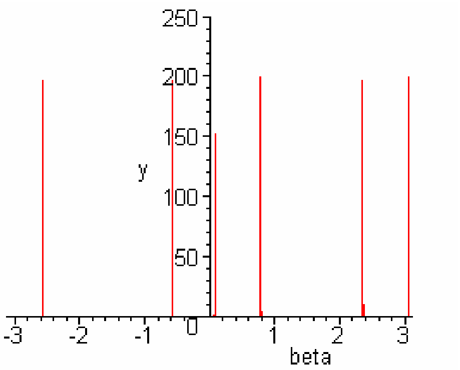
$\lambda=8d/8$



$\lambda=7d/8$



$\lambda=6d/8$



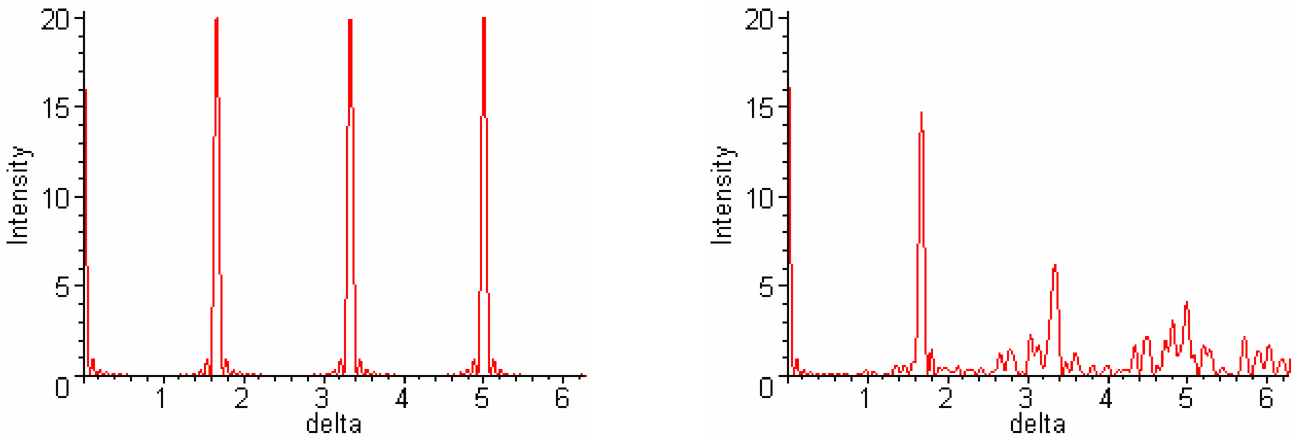
$\lambda=5d/8$

In each case,
$N=200$
$M=1$
$d=135$
$\alpha=\pi/4$

- Intensity as a function of β
- Out of one intensity peak, many, and vice versa
  - As λ increases, the specular spikes move, multiply, and/or decrease

Adding Randomness

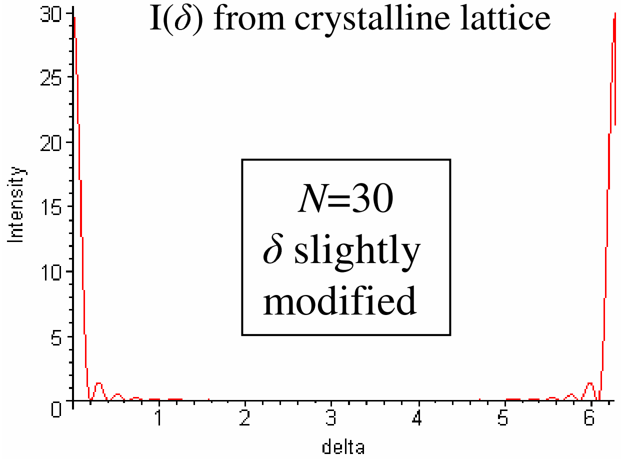
- To better understand the thin film aspect, a Gaussian random number generator was added to take the inter-atomic spacing from the crystalline case to the amorphous case
  - Sputtered thin films are generally amorphous in nature
- Atoms were shifted from their *usual places in the lattice*
- Note: The results are in terms of path length difference δ



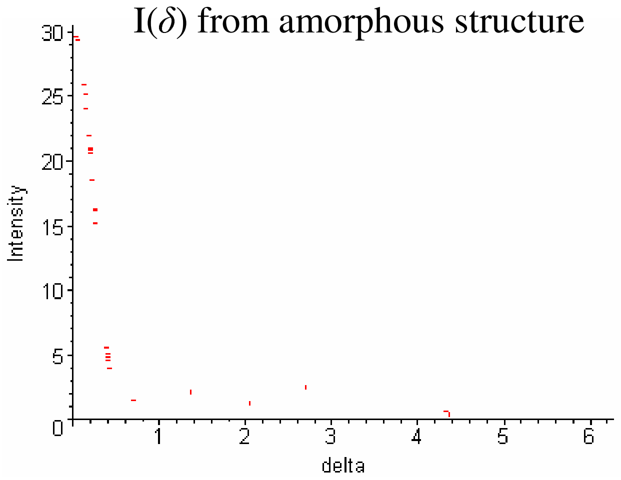
N=10
M=1
d=120
d+(-1,0,1)
λ=100

Amorphous Atomic Arrangement (AAA)

- Variations in atomic spacing *d* simulated by a random number generator
    - Atomic shift for each atom measured *relative to the previous neighboring atom's shift*
- To what extent is diffraction preserved?
- The amorphous plot required more generating of points
    - Why this is yet unknown



I(δ) from crystalline lattice



I(δ) from amorphous structure

Conclusions

- Most of our results are still preliminary, but with continued research we hope to answer the question of theoretical optical constant determination. We have:
  - reproduced specular and Bragg reflections from isotropic scattering processes
  - showed the effects of disorder on the width and height of the Bragg peaks
- We may be able to:
  - predict the order in the atomic positions through height comparison of specular and Bragg peaks
  - perform qualitative studies of the effects of different kinds of roughness on the reflection coefficient of the surface
  - Perform a qualitative study of different scattering cross-sections on reflection

- It may be possible to theoretically calculate *accurate* optical constants for elements in the EUV and soft x-ray regime
  - This refining of optical constants could have major impacts in industries such thin films, affecting everything from EUV lithography to satellite-based observatories

References

M. A. Karam, "Molecular optics approach to electromagnetic wave interactions with stratified media.” J. Opt. Soc. Am. **13** (11), 2208-2218 (1996).

G. C. Reali, “Exact Solution of the Equations of Molecular Optics for Refraction and Reflection of an Electromagnetic Wave on a Semi-Infinite Dielectric.*” J. Opt. Soc. Am. **62** (10), 1165-1174 (1982).

Christopher Palmer, Erwin Loewen, editor. “Diffraction Grating Handbook.” Richardson Grating Laboratory, NY, 2002, p.18.

Notes from interviews with Dr. Turley. Late March to early September, 2002.