Characterization of Radiation Damage Using X-Ray Diffraction

I. C. Noyan

Columbia University, Department of Applied Physics and Applied Mathematics

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- **Diffraction techniques are very sensitive to the material symmetry within a probe volume.**
- **If the material is truly *perfect*, and *large*, we can use *dynamical diffraction* theory to model scattered beams.**
- ***Imperfections*, or *small material volumes*, change the mode of scattering to *kinematical*.**

- Since radiation can cause local damage within the material, the scattering signature should change with exposure.
- Thus, x-ray or neutron diffraction techniques should be able to yield “in-situ” data on the evolution of lattice damage.
- This talk will illustrate an actual real time measurement using “x-rays” to cause the damage and, also, to detect it.

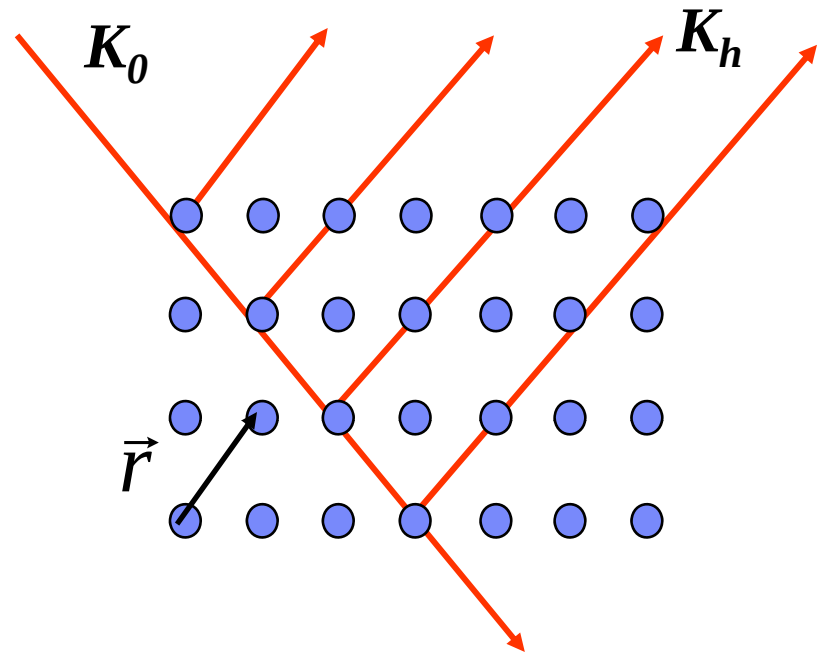
Synopsis

- Structural damage is observed in silicon-on-insulator (SOI) and SiGe on SOI samples illuminated with monochromatic (11.2 keV) x-ray microbeams approximately 250 nm in diameter.
- The x-ray diffraction peaks from the irradiated layers degrade irreversibly with time, indicating permanent structural damage to the crystal lattice.
- We do not see any such damage on bulk samples.
- The damage mechanism is not fully understood at this time.

Outline

- Basic Diffraction Theory: What do we expect to measure?
- Experiments/Results: What we did measure?
- Conclusions.

Basic Kinematical Formulation



- Construct the diffracted wave from the coherently scattered waves from all crystal atoms; assume:
  - Photoelectric absorption only,
  - No multiple scattering,
  - Negligible energy taken by the reflected beam.

    - X-rays are really scattered by electrons: write the electron density distribution in the crystal as:

$$\rho(\vec{r}) = \rho_\infty(\vec{r})\, y(\vec{r})$$

$\rho_\infty(\vec{r})$: Electron density distribution of a triply-periodic *infinite* medium.

$$\rho_\infty(\vec{r}) = \mathrm{v}^{-1} \sum_{hkl} F_{hkl} \exp(-i\, 2\pi\, \vec{h}.\vec{r})$$

F_{hkl}: the structure factor (diffraction from a unit cell).

v: unit cell volume.

$y(\vec{r})$: envelope function of the sample. $y(\vec{r}) = \begin{cases} 1 & \text{inside} \\ 0 & \text{outside} \end{cases}$.

The diffracted wave amplitude, $\mathcal{A}$, can be written as:

$$\mathcal{A}(\vec{\boldsymbol{k}}) = \iiint \rho(\vec{\boldsymbol{r}}) \exp[\boldsymbol{i} 2\pi(\vec{\boldsymbol{k}} \cdot \vec{\boldsymbol{r}})] \boldsymbol{d}^3 \vec{\boldsymbol{r}},$$

$$\mathcal{A}(\vec{k}) = \mathrm{v}^{-1} \sum_{hkl} F_{hkl} Y(\vec{k} - \vec{h}),$$

$$\boldsymbol{Y}(\vec{\boldsymbol{k}} - \vec{\boldsymbol{h}}) = \iiint \boldsymbol{y}(\vec{\boldsymbol{r}}) \exp[\boldsymbol{i} 2\pi(\vec{\boldsymbol{k}} - \vec{\boldsymbol{h}}) \cdot \vec{\boldsymbol{r}}] \boldsymbol{d}^3 \vec{\boldsymbol{r}},$$

$\vec{\boldsymbol{K}}_{\boldsymbol{0}}$, $\vec{\boldsymbol{K}}_{\boldsymbol{h}}$: the incident and diffracted wave vectors, respectively.

$\vec{k} = \vec{K}_h - \vec{K}_0$, is the wave vector.

$Y(\vec{k} - \vec{h})$, Fourier transform of sample shape (***the shape function***.)

$$\boldsymbol{Y}(\vec{\boldsymbol{k}}-\vec{\boldsymbol{h}})=\iiint \boldsymbol{y}(\vec{\boldsymbol{r}})\exp[\boldsymbol{i}2\pi(\vec{\boldsymbol{k}}-\vec{\boldsymbol{h}})\cdot\vec{\boldsymbol{r}}]\boldsymbol{d}^3\vec{\boldsymbol{r}},$$

Crystal size is <u>infinite</u> in 3-d:

$$Y(\vec{k}-\vec{h})=\delta(\vec{k}-\vec{h}),\quad then:$$

$$\vec{K}_d-\vec{K}_0=\vec{h}$$ for no-zero amplitude. -> Bragg's law.

For a crystalline film of finite thickness "t":

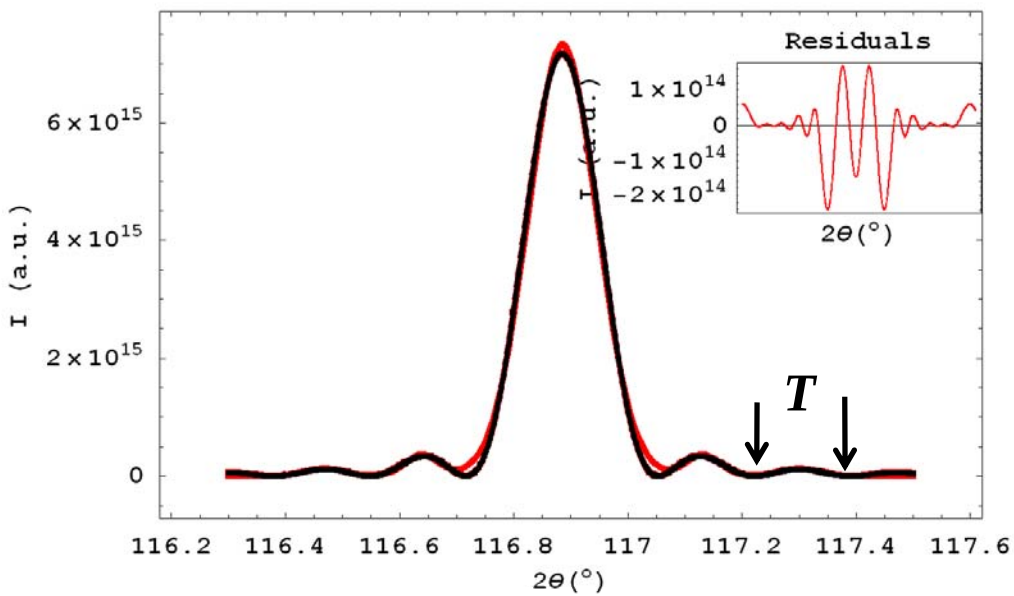
$$\boldsymbol{Y}(\vec{\boldsymbol{k}}-\vec{\boldsymbol{h}})=\int_{-\boldsymbol{t}/2}^{\boldsymbol{t}/2}\boldsymbol{dx}\int_{-\infty}^{\infty}\boldsymbol{dy}\int_{\infty}^{\infty}\exp[\boldsymbol{i}2\pi(\vec{\boldsymbol{k}}-\vec{\boldsymbol{h}})\cdot\vec{\boldsymbol{r}}]\boldsymbol{dz}$$

$$\boldsymbol{Y}(\vec{\boldsymbol{k}}-\vec{\boldsymbol{h}})\approx\boldsymbol{V}\frac{\sin(\pi(\vec{\boldsymbol{k}}-\vec{\boldsymbol{h}})\boldsymbol{t})}{\pi(\vec{\boldsymbol{k}}-\vec{\boldsymbol{h}})\boldsymbol{t}},$$

Kinematic Diffraction Peak Profile from a thin film:

$$I(\Delta 2\theta) \approx N^2 \left|F_{hkl}\right|^2 \frac{\sin^2(t\,\Theta)}{(t\,\Theta)^2},$$

$$\Theta = \pi\, k \cos\theta_B \left(\Delta 2\theta\right)$$



The expected peak shape of the **SOI** layer is given by the **square of the normalized sinc function**.

$$t \to \infty, A = 0 \;\; for \;\; \Delta 2\theta \neq 0$$

$$T = \frac{\lambda}{t\cos(\theta_B)}$$

$$I = \frac{I_{Max}}{2} \;\; for \;\; t\,\pi\,\mathbf{k}\cos\theta\;\Delta 2\theta = \mp 1.39$$

$$FWHM \cong \frac{0.885\lambda}{t\;\cos\theta_B}$$

Samples

All SOI substrates were wafer-bonded; the SOI layer and underlying Si substrate possess an offset of ~0.40°.

We looked at:

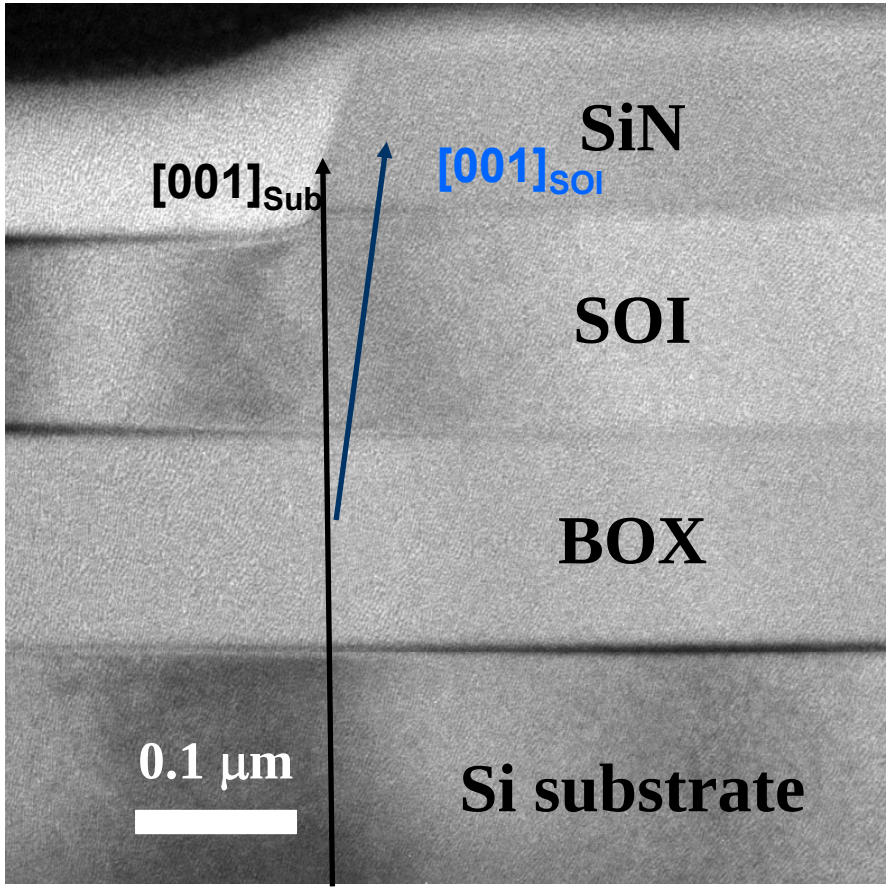
Bare SOI

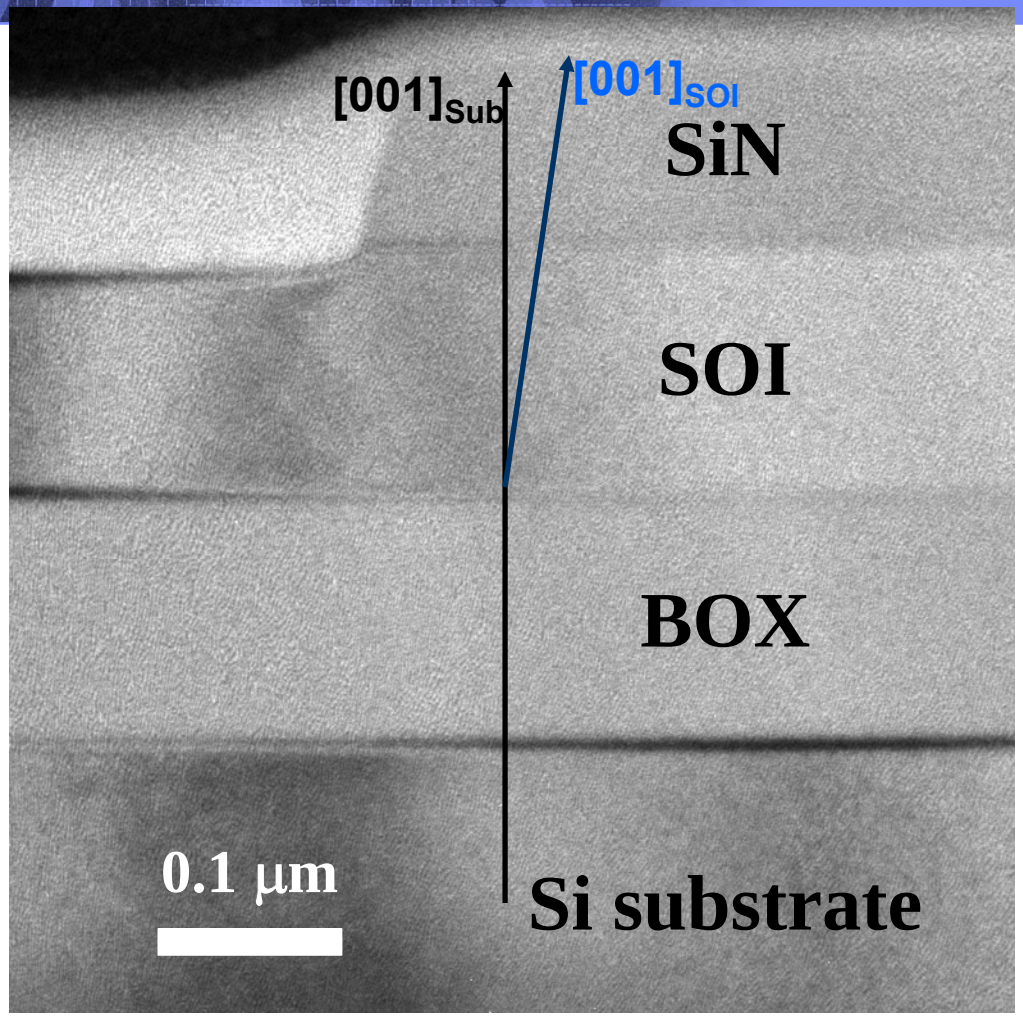
SOI/ SiN

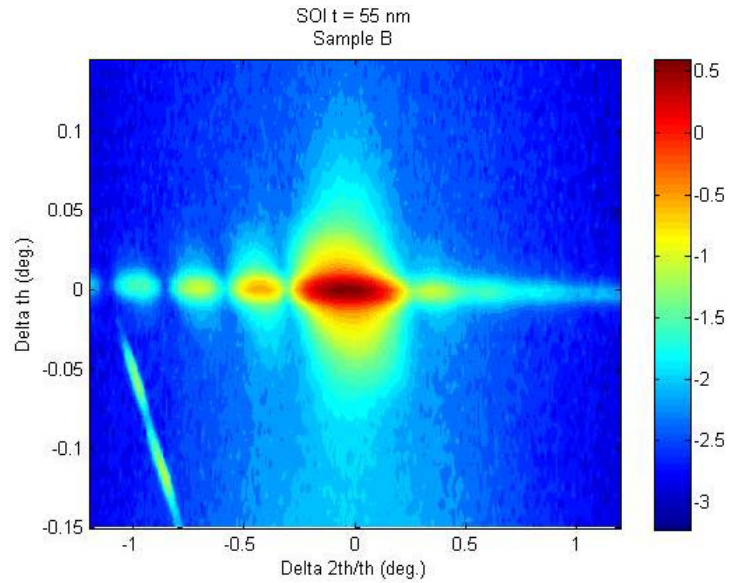
SOI/SiGe

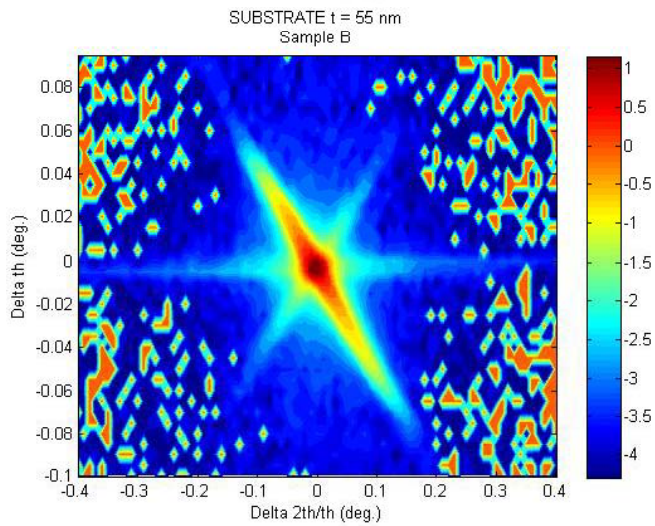
BulkSi/SiGe

Bulk Si

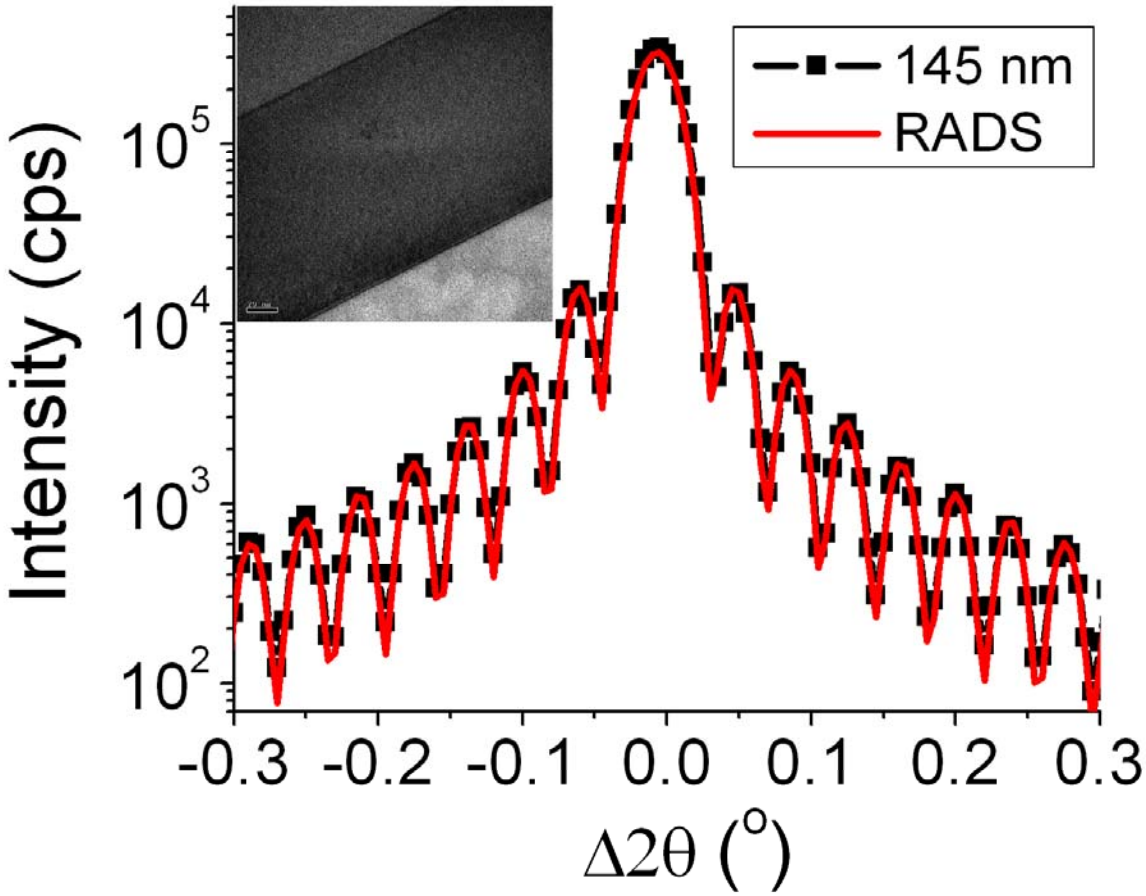



[001]Sub
[001]SOI
SiN
SOI
BOX
0.1 μm
Si substrate


SOI t = 55 nm
Sample B
Delta th (deg.)
Delta 2th/th (deg.)


SUBSTRATE t = 55 nm
Sample B
Delta th (deg.)
Delta 2th/th (deg.)

NSLS X20 Macrobeam Measurements



Si 004

8.05 keV (λ = 1.54 Å)

~10^9 photons/sec

1 x 2 mm^2 beam size

"t" from three techniques (nm)

TEM: 142 (1)

FWHM->143 (1)

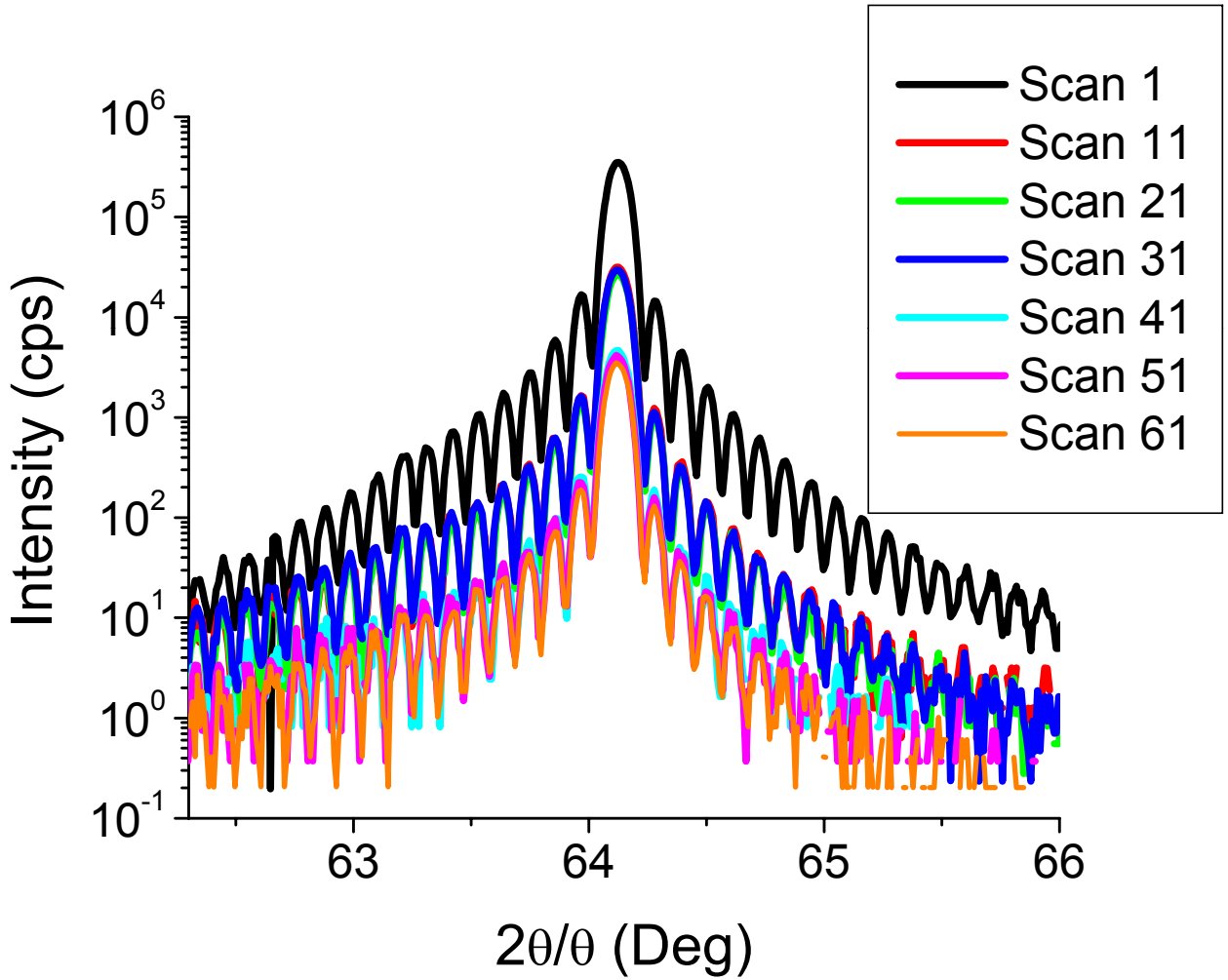
RADS->142.23 (0.02)

- The theory works fine for SOI samples.

J. Appl. Cryst. (2009). 42, 401–410 , Ying, et. al.

The diffraction profile is invariant over 70 measurements, two and a half days.

No radiation damage at X20.

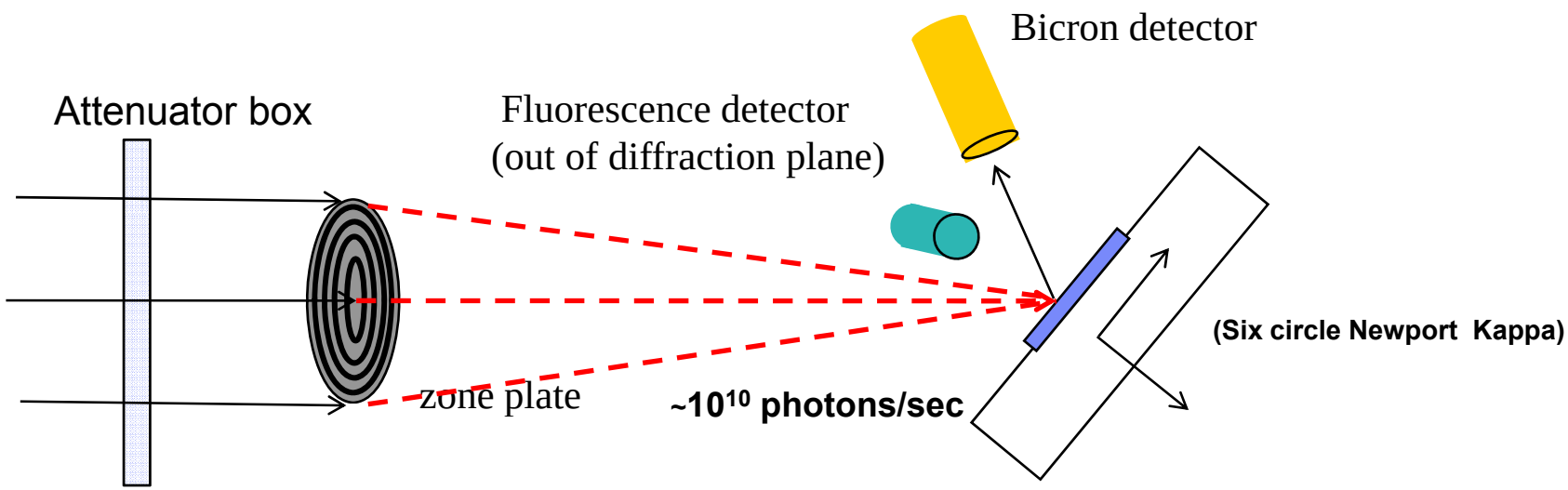


Microbeam measurements

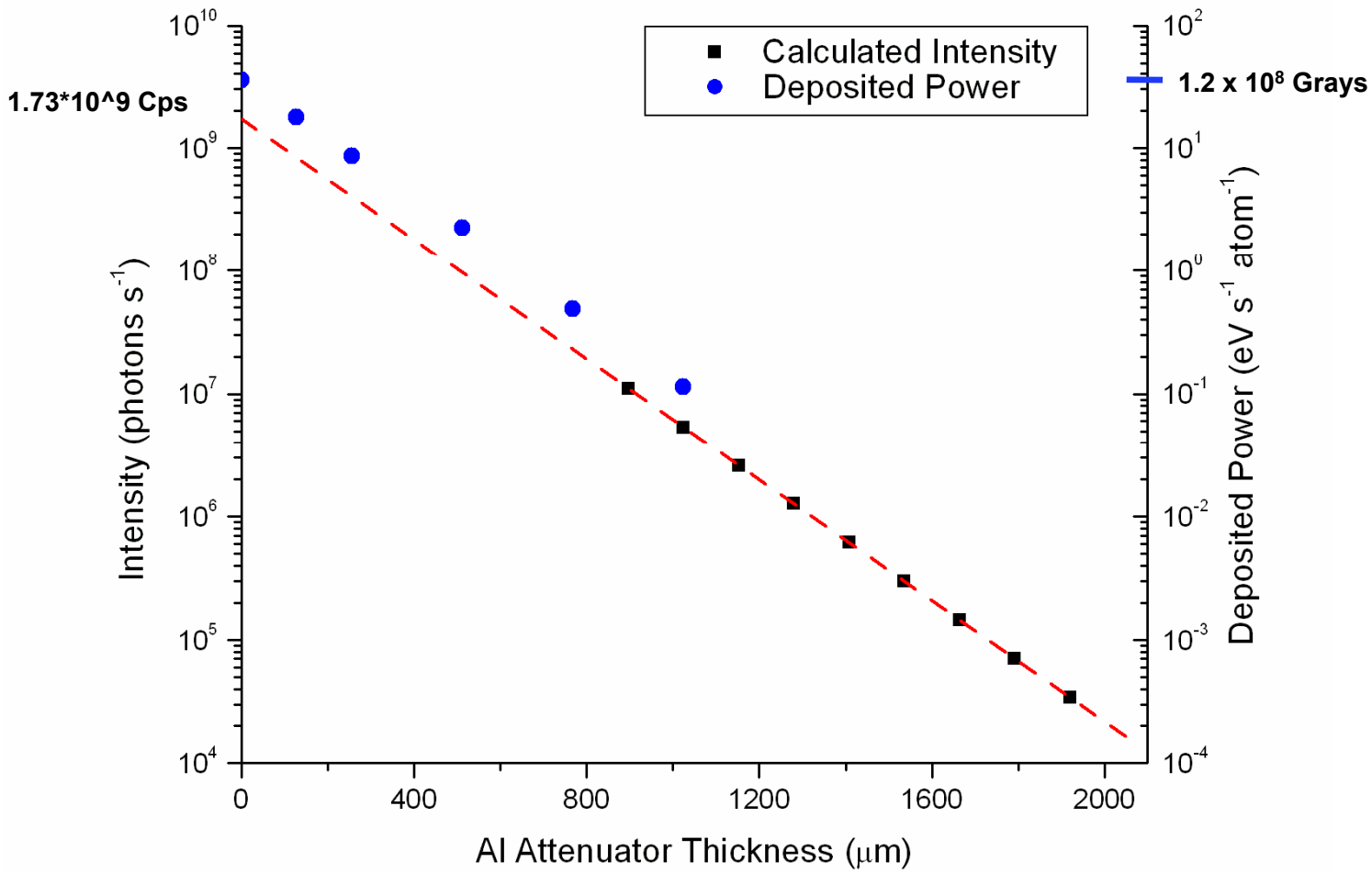
APS 2ID-D: Zone plate optics (~ 0.3 μm FWHM)

E = 11.2 keV (1.107 Å)

- **Slit optics on the diffracted beam path (widths of 100 μm and 10 mm),**
- **Measure SiGe (008) and Si (008) intensity (107° and 109° $2\theta_B$)**
- **Simultaneous measurement of diffraction and Ge Kα fluorescence,**
- **The beam intensity can be attenuated by inserting Al foils in the beam path.**



Incident focused beam as a function of attenuator thickness



Polvino, et. al. *APL*, **92**, 224105, 2008

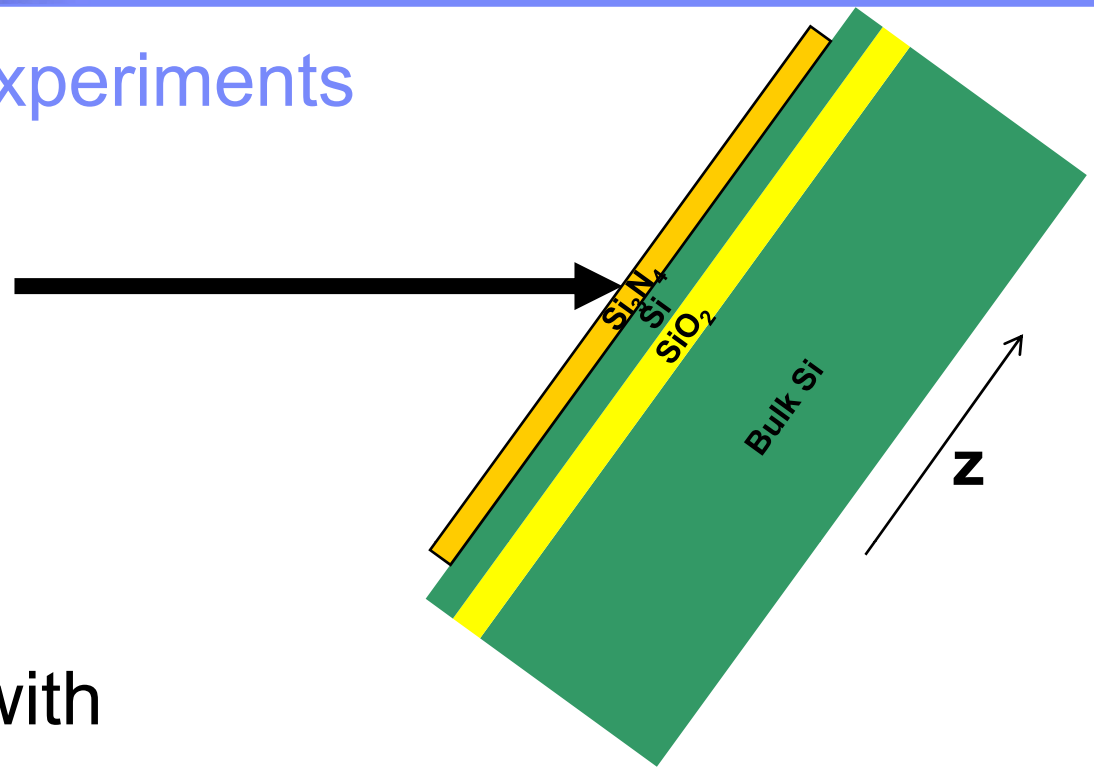
Calculated from XOP XPOWER pkg.

Repeated Diffraction Experiments

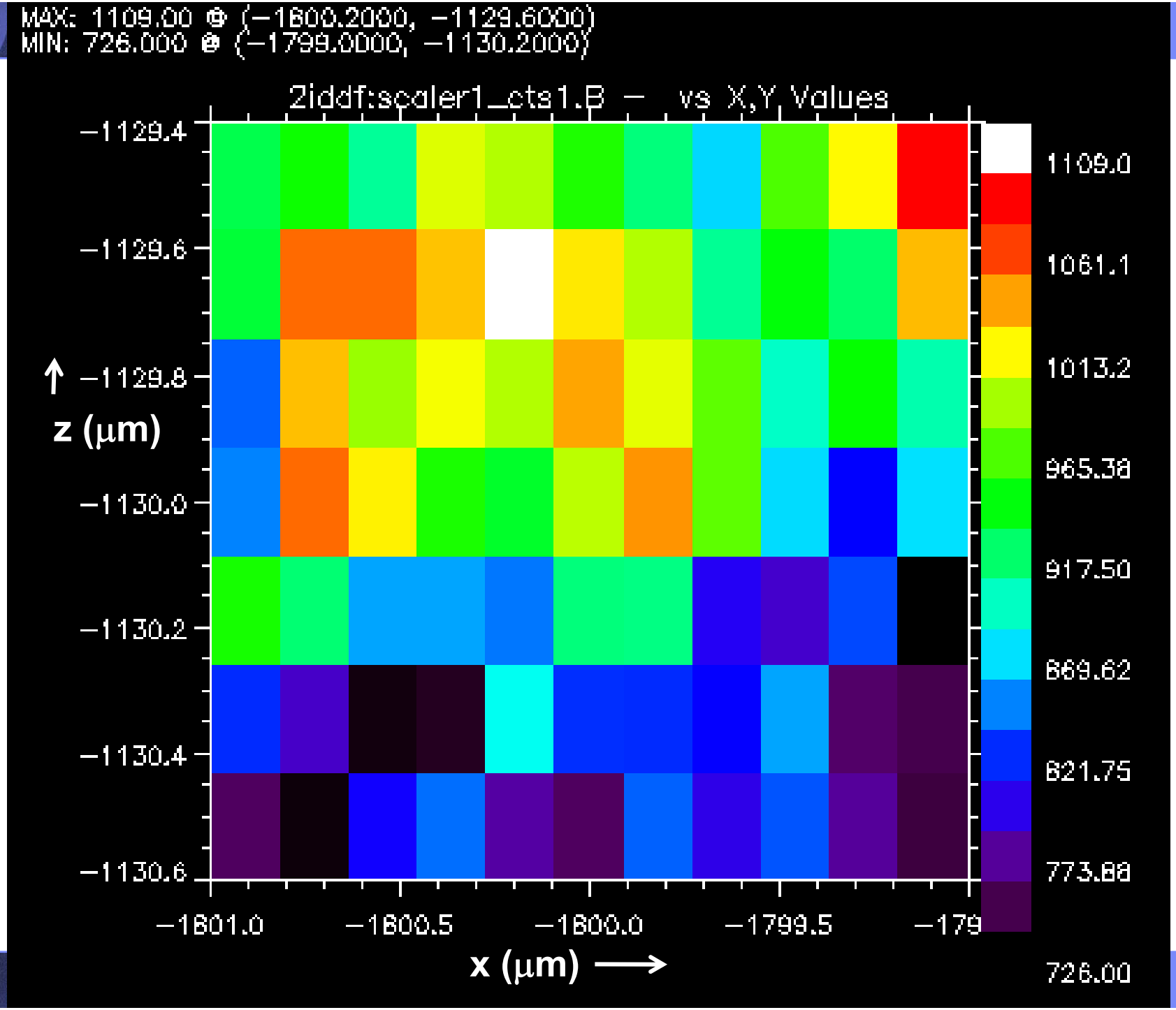
t_{Si3N4} ~ 105 nm

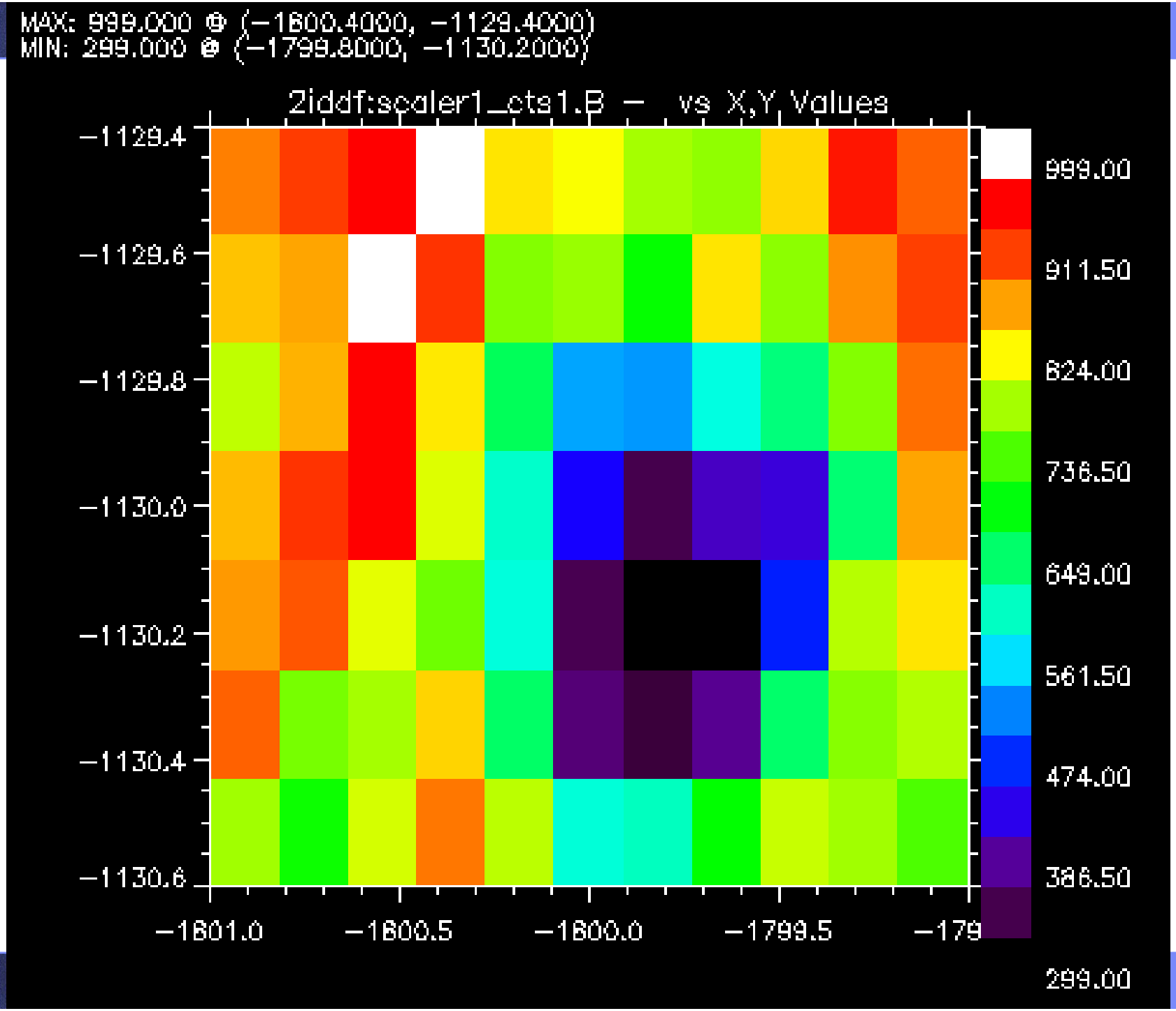
t_{SOI} ~140nm

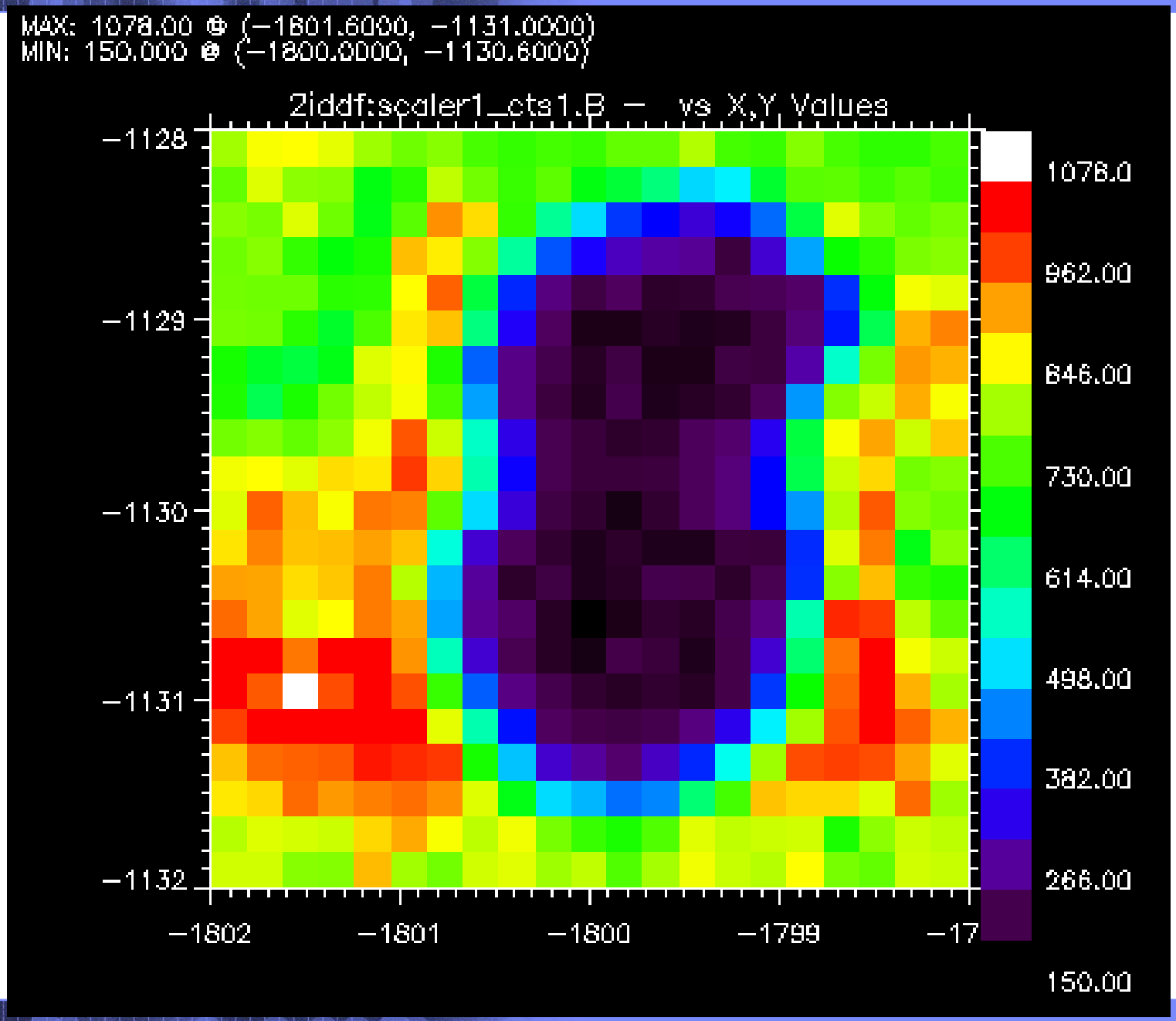
t_{BOX} 140 nm



- Sample is irradiated with unattenuated beam for 10 minute intervals
  - **After each irradiation attenuators are inserted (8 layers Al foil), and the sample is measured in an x-z topographic mesh around the area of irradiation and a θ/2θ scan at the center of irradiation.**


MAX: 1109.00 @ (−1800.2000, −1129.6000)
MIN: 726.000 @ (−1799.0000, −1130.2000)
2iddf:scaler1_cts1.B − vs X,Y Values
−1129.4
−1129.6
−1129.8
−1130.0
−1130.2
−1130.4
−1130.6
z (μm)
−1801.0
−1800.5
−1800.0
−1799.5
−179
x (μm)
1109.0
1061.1
1013.2
965.38
917.50
869.62
821.75
773.88
726.00


MAX: 999.000 @ (-1800.4000, -1129.4000)
MIN: 299.000 @ (-1799.8000, -1130.2000)
2iddf:scaler1_cts1.B - vs X,Y Values
-1129.4
-1129.6
-1129.8
-1130.0
-1130.2
-1130.4
-1130.6
-1801.0
-1800.5
-1800.0
-1799.5
-179
999.00
911.50
824.00
736.50
649.00
561.50
474.00
386.50
299.00

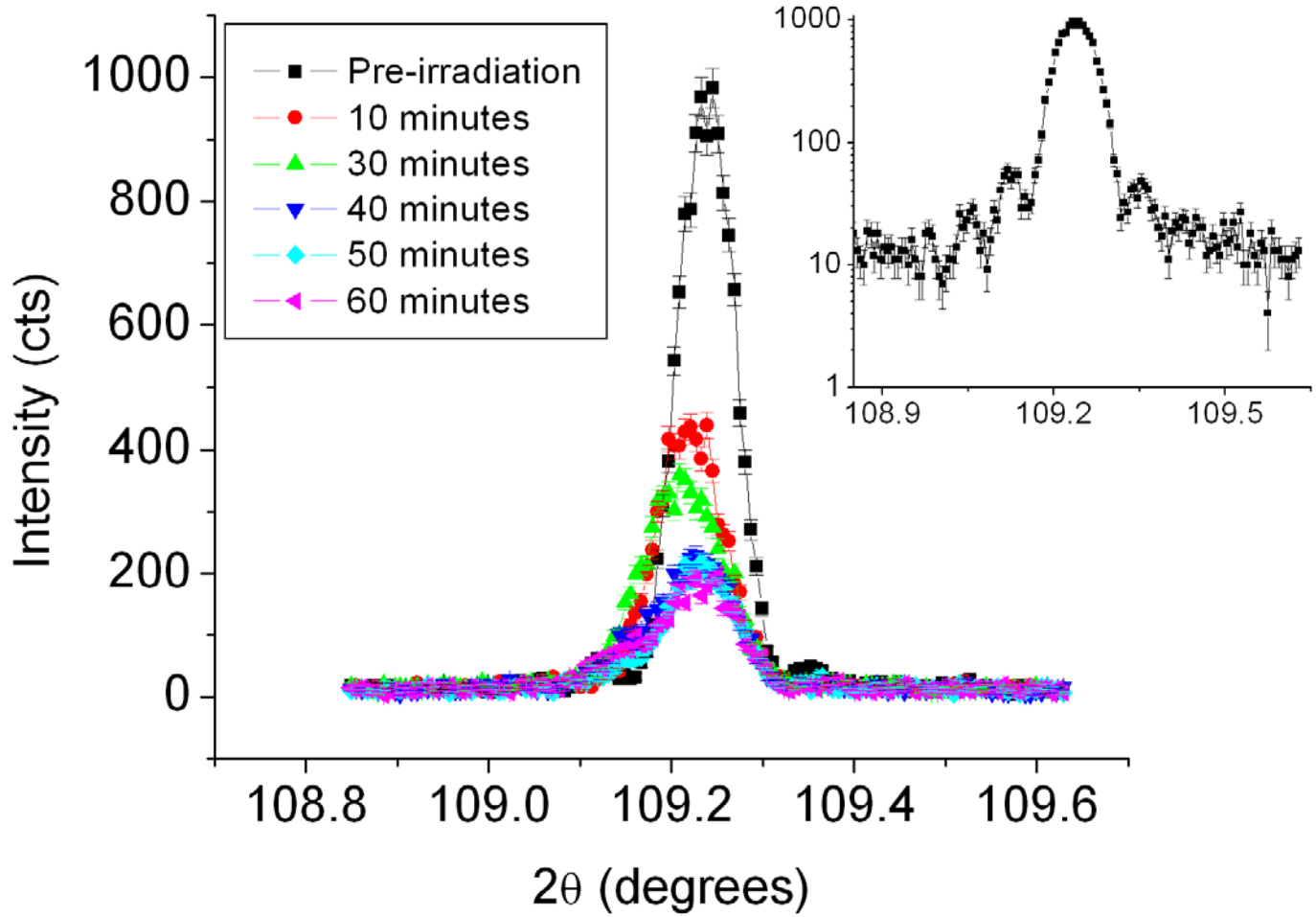

MAX: 1078.00 @ (−1801.6000, −1131.0000)
MIN: 150.000 @ (−1800.0000, −1130.6000)
2iddf:scaler1_cts1.B − vs X,Y Values
−1128
−1129
−1130
−1131
−1132
−1802
−1801
−1800
−1799
−17
1078.0
962.00
846.00
730.00
614.00
498.00
382.00
266.00
150.00

Variation of the 008 diffraction peak profile from the SOI layer with exposure to direct beam .

The inset shows the profile at zero exposure with the intensity in log scale.

The thickness fringes indicate a perfect Si layer.

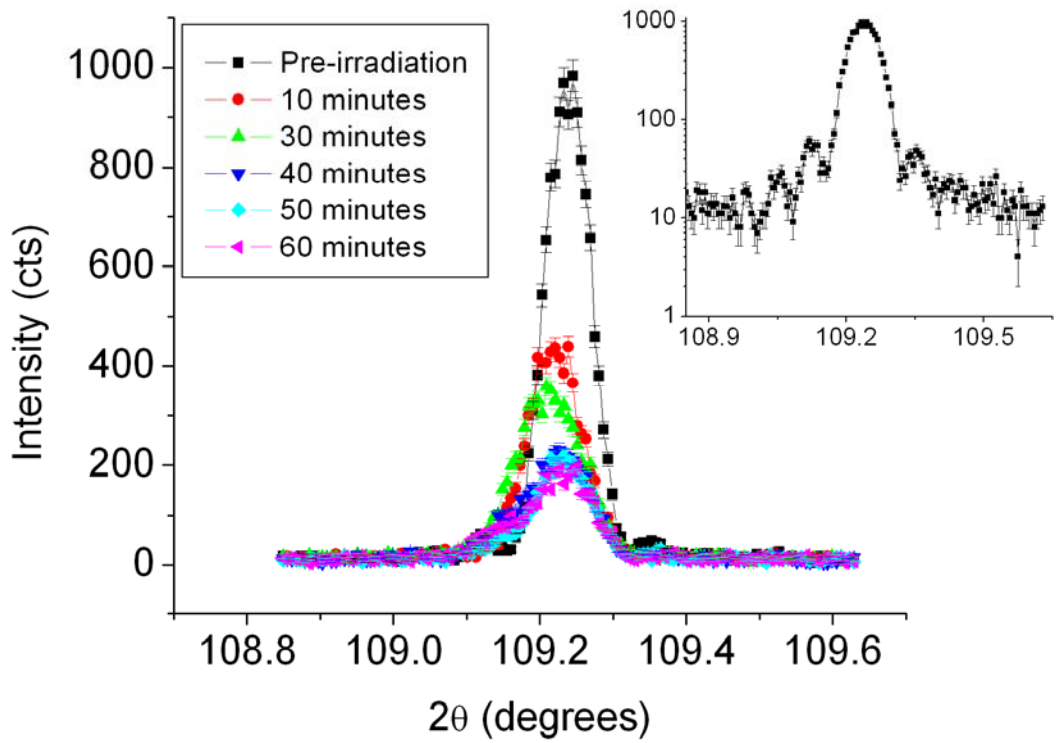
These fringes disappear with irradiation.



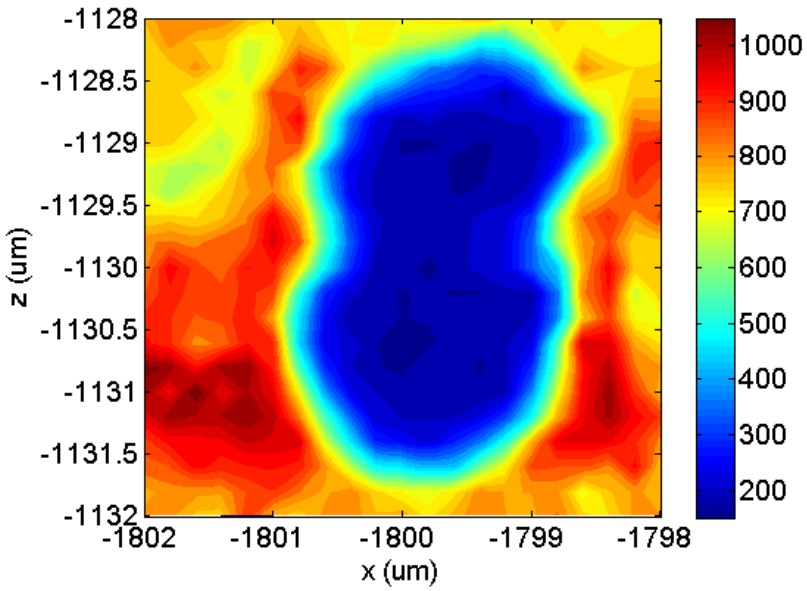
The initial diffracting layer thickness, calculated from the peak-breadth using the Scherrer formula, yields a value very close to the physical thickness of the SOI layer.

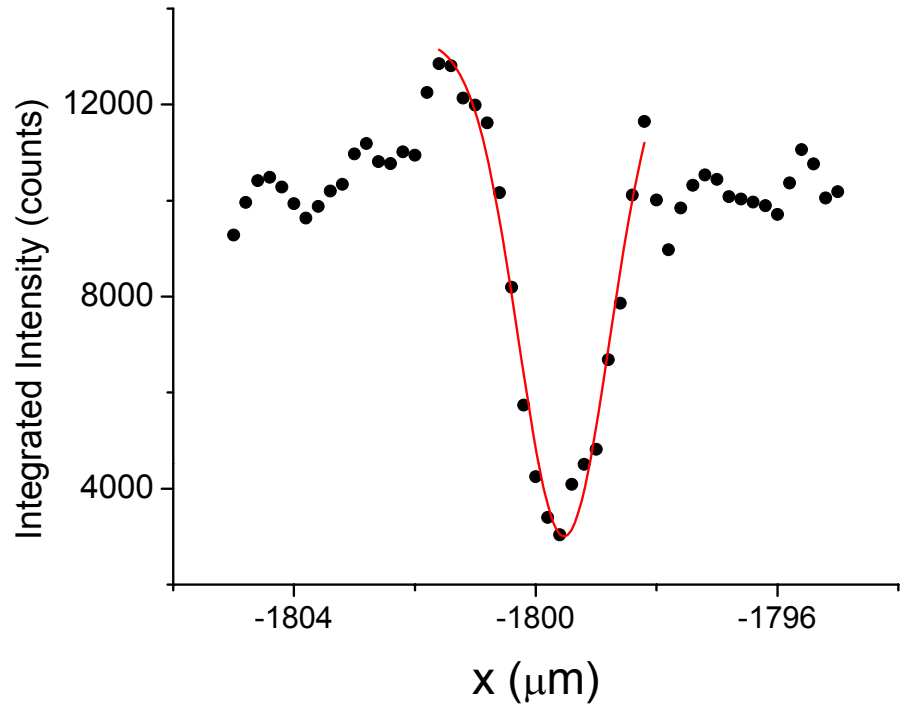
The thickness after 60 minutes of irradiation, calculated from the broadened peak profile, drops to approximately half of its initial value.

The integrated intensity of the broadened peak at this point is approximately 1/3rd of its initial value.



After irradiation, there is less crystalline Si in the irradiated volume.





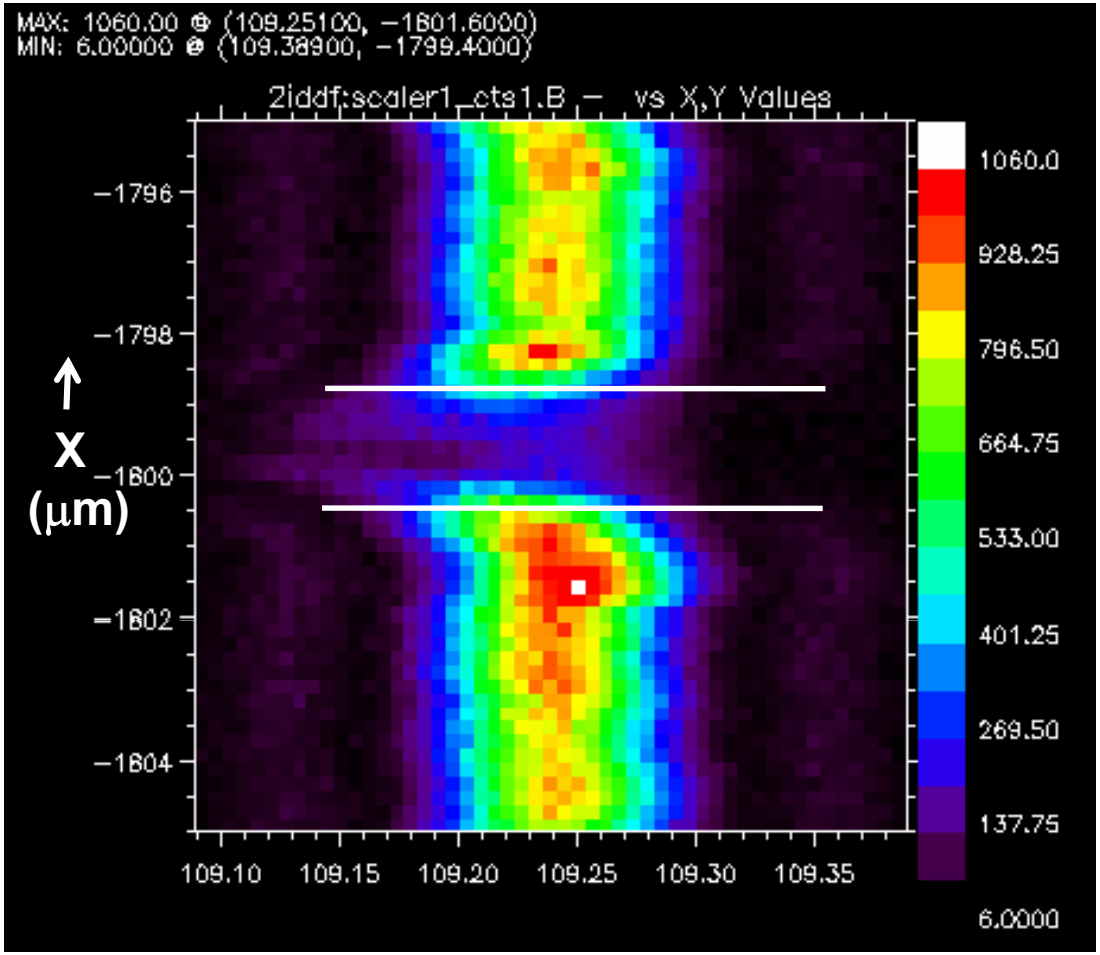
Si 008 intensity map of the radiation-damaged region . In this figure *x* and *z* coordinates define the sample surface.

Damaged region is ~6x larger than the beam.

Variation of integrated intensity across the minor diameter. The red line is a Gaussian fit to the damaged region, centered at x=-1799.5 μm, with FWHM of 1.78 μm.

The damage modifies the elastic strain field set-up due to the nitride overlayer.

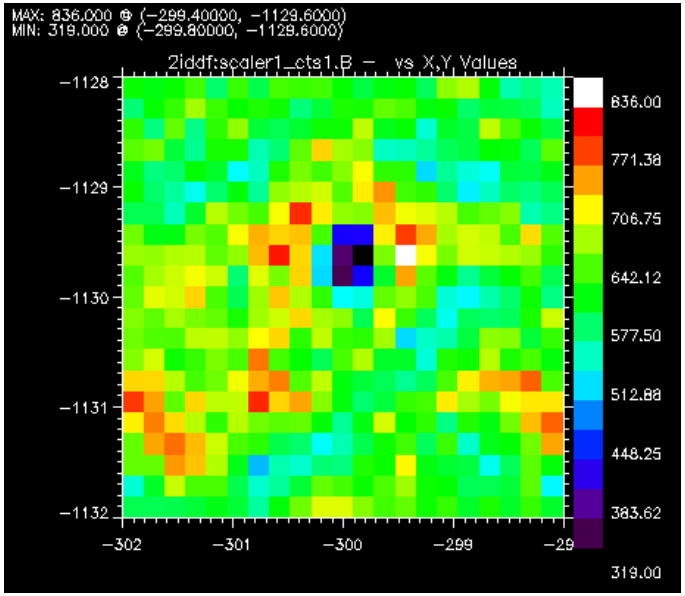
- The $2\theta_B$ peak values change to lower values on either side of the damage region.
- There are two intensity maxima bracketing both sides of the peak.
- **Two new boundaries have been created.**



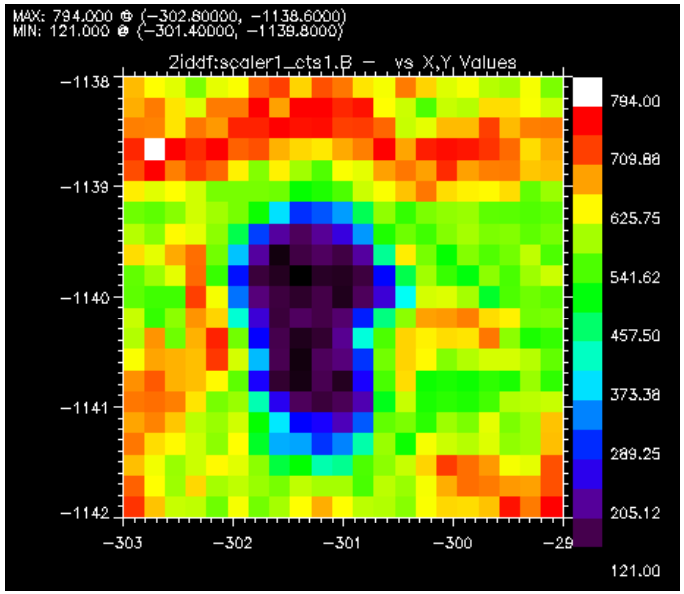
Radial scans as a function of x position (minor diameter)

Other Samples

- We observed similar damage when we irradiated an SOI region with no SiN cover, and one with epitaxial SiGe.
- We able to locate the damaged regions a day after the initial scans were run.
- There was no change in intensities or dimensions.



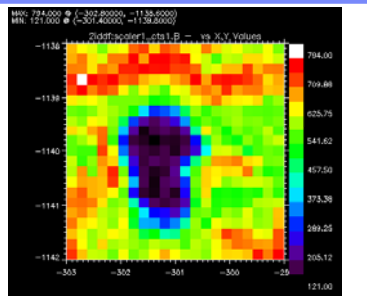
10 min

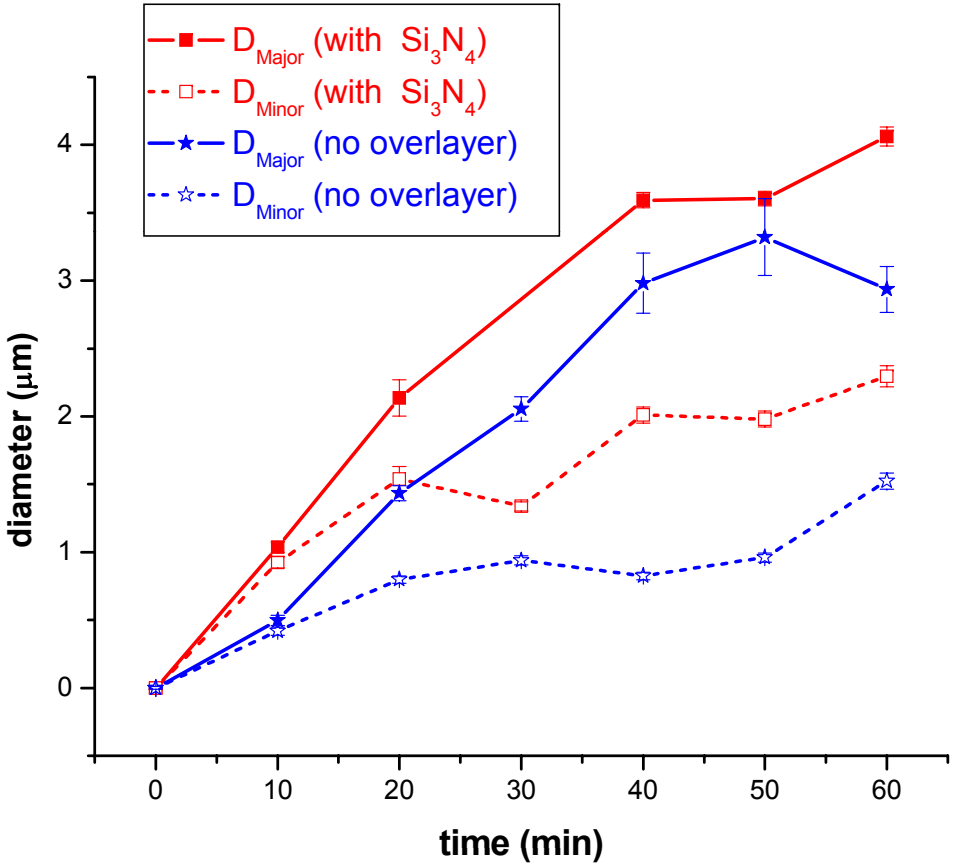


60 min

The growth rates of the damaged regions were ~ parabolic.

Growth saturated after ~ 60 minutes.

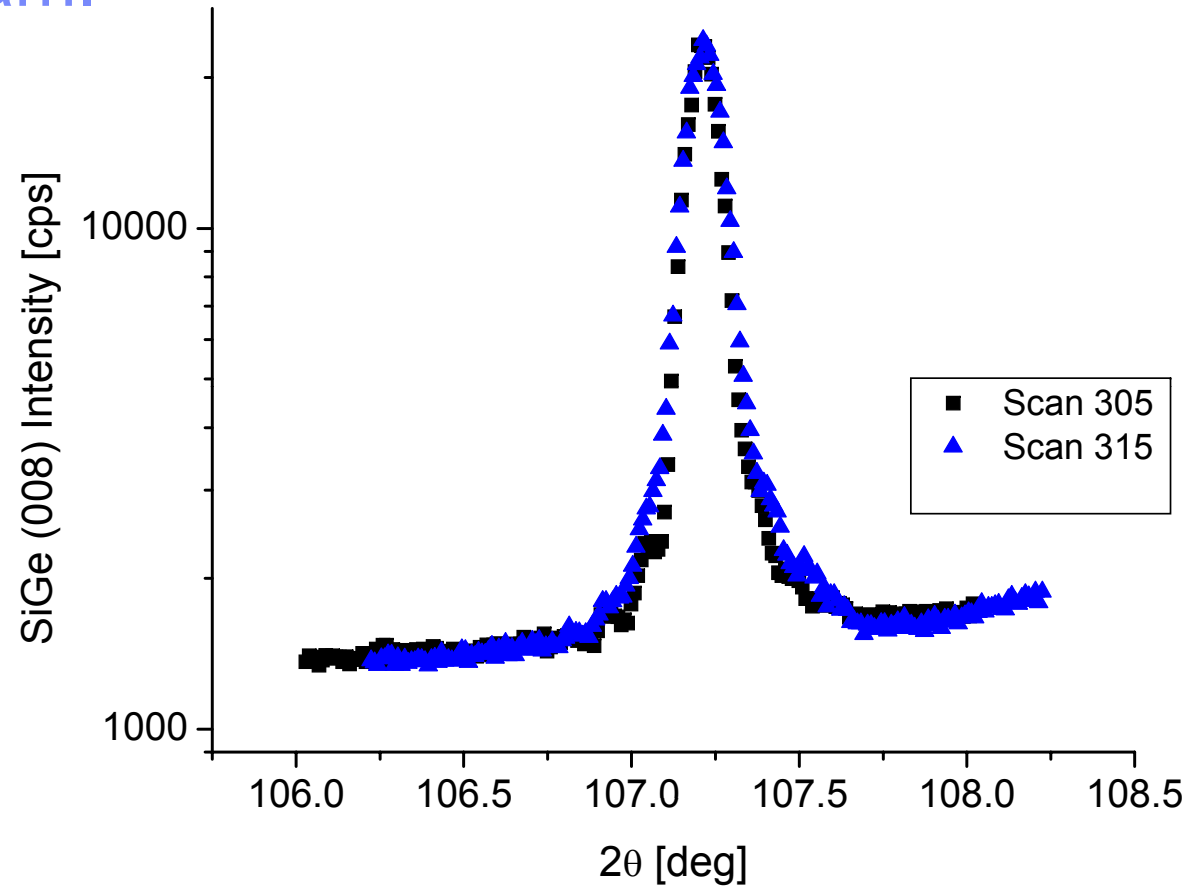




Samples Investigated

	$t_{Substrate}$ (µm)	t_{BOX} (nm)	t_{SOI}	Capping Layer
Sample A	700	140	25 nm	55nm Heteroepitaxial SiGe
Sample B	700	140	140 nm	100 nm Tensile Si_3N_4 Film
Sample C	700	140	140 nm	100 nm Compressive Si_3N_4 Film
Sample D	700	N/A	N/A	80 nm Heteroepitaxial $Si_{0.85}Ge_{0.15}$

The control sample shows no degradation after 9 hr of direct beam.



- **Sample D: 80 nm Heteroepitaxial $Si_{0.85}Ge_{0.15}$ on 700 μm Si.**
- **We observed no damage on uncoated Si wafers either.**

The crystal lattice of the SOI layer suffers damage as a function of irradiation:

- Thickness fringes disappear;
- Peak broadens
- Integrated intensity drops.

These changes occur irrespective of the presence of a capping layer.

The damaged region is stable.

We could not resolve the damage with AFM. SEM or IR microscopy.

Conclusions

- **X-ray diffraction techniques are excellent tools for <u>detecting</u> damage.**
  - These tools are non-contact and "~*non-destructive*" and can be very useful for the study of radiation damage evolution.
- **Diffraction data analysis involves solving an inverse problem: *Data → Model parameters***
  - *existence, uniqueness, stability of the solution*
- **Defining a suitable "test" sample where the problem is constrained might be useful.**

- **Integrated modeling tools are needed to understand the damage mechanism.**
- **A *rigorous* combination of scattering theory and multi-scale damage modeling might yield exciting results.**

ACKNOWLEDGEMENTS

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