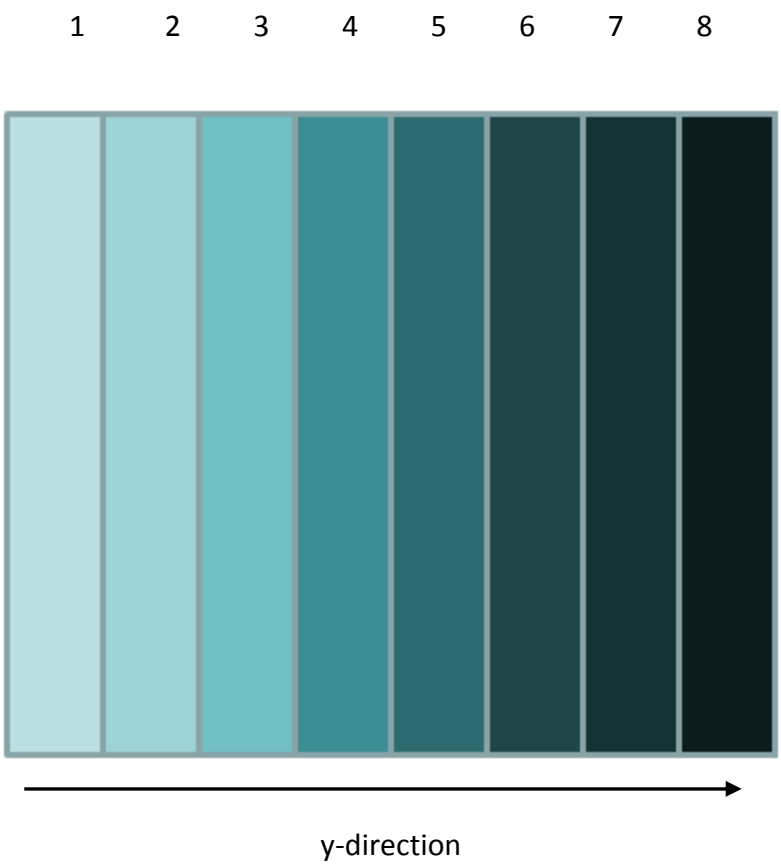
Supplemental information:

Schematic diagram of wedge creation:

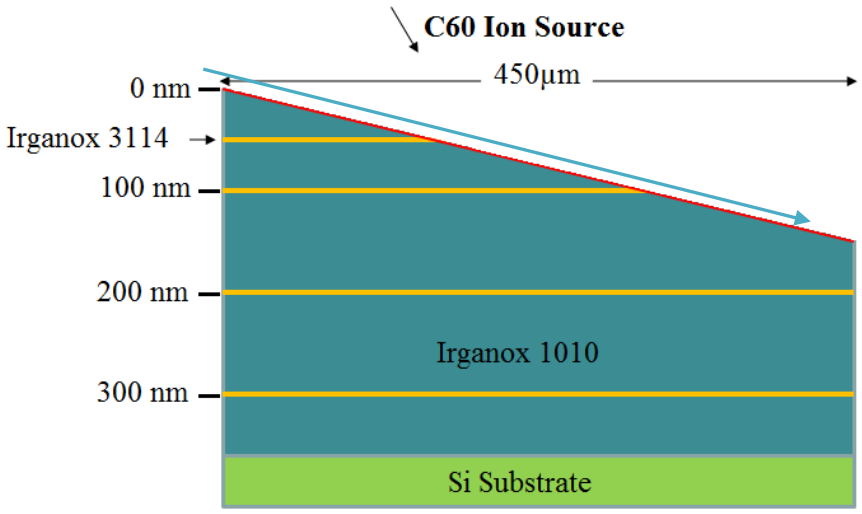


In order to erode a wedge-shaped crater, the raster area was varied from frame to frame by sequentially skipping more and more lines in the *y*-direction.

As shown in the top figure, the ion beam first rastered the whole crater area from 1 to 8. Then the ion beam skipped area 1 and rastered a smaller area in the crater from 2 to 8. The ion beam continued to skip lines in the y-direction until the end of crater is reached. This way, the crater area along the y-direction received an increasing ion dose. In this figure, the color darkness is proportional to the total applied fluence.

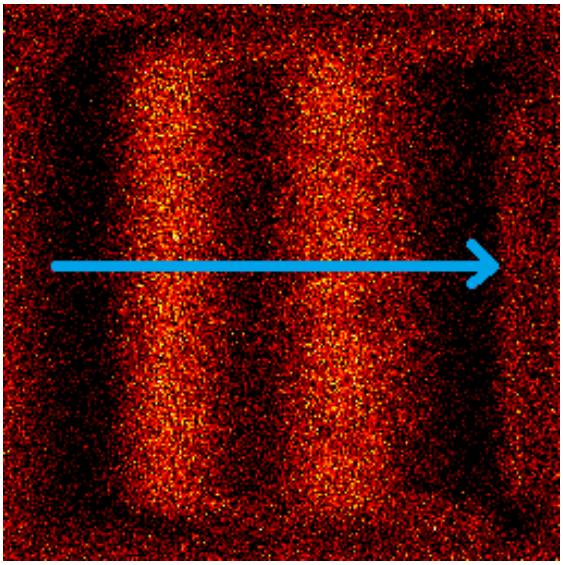
In real experiments, by increasing the sputtering time per cycle, pixels along the y-direction can receive linearly increasing ion fluence.

SIMS of wedge

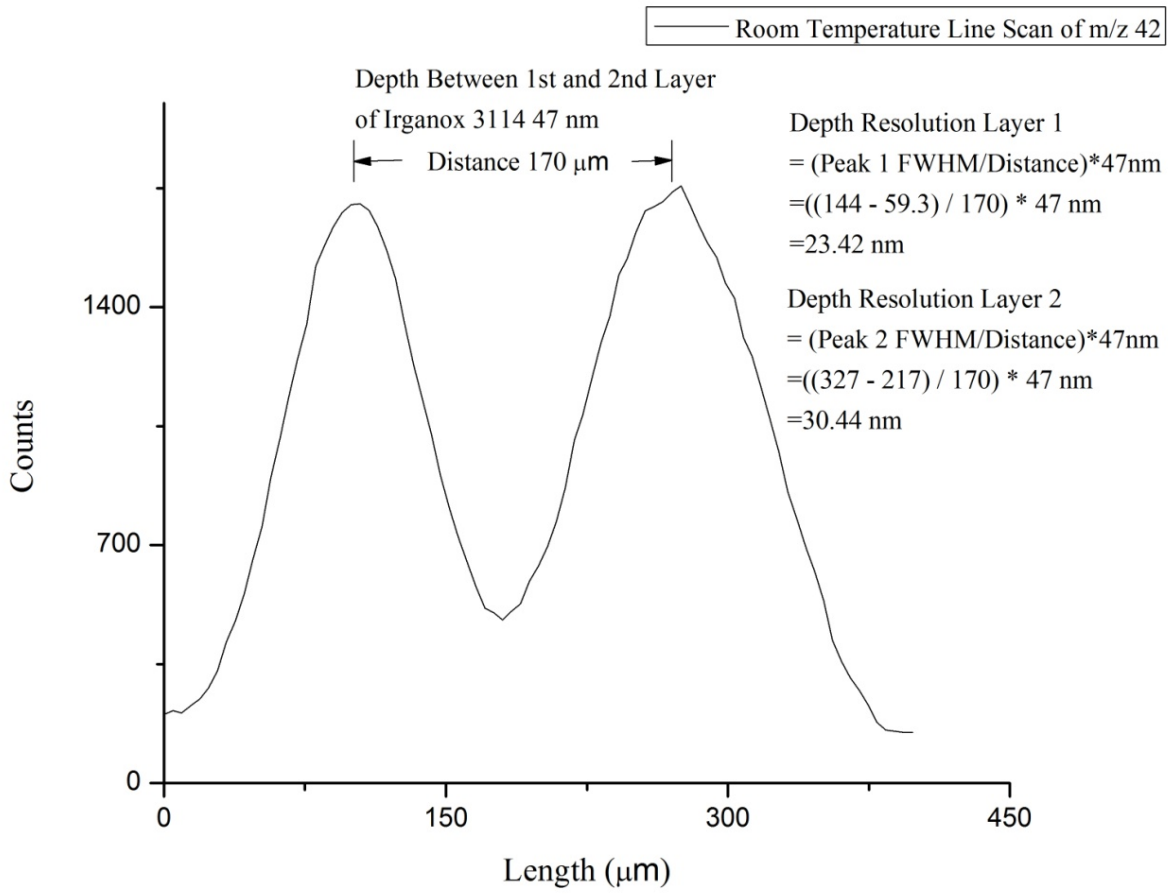


Dark Green: Irganox 1010; Orange: Irganox 3114; Light Green: Si

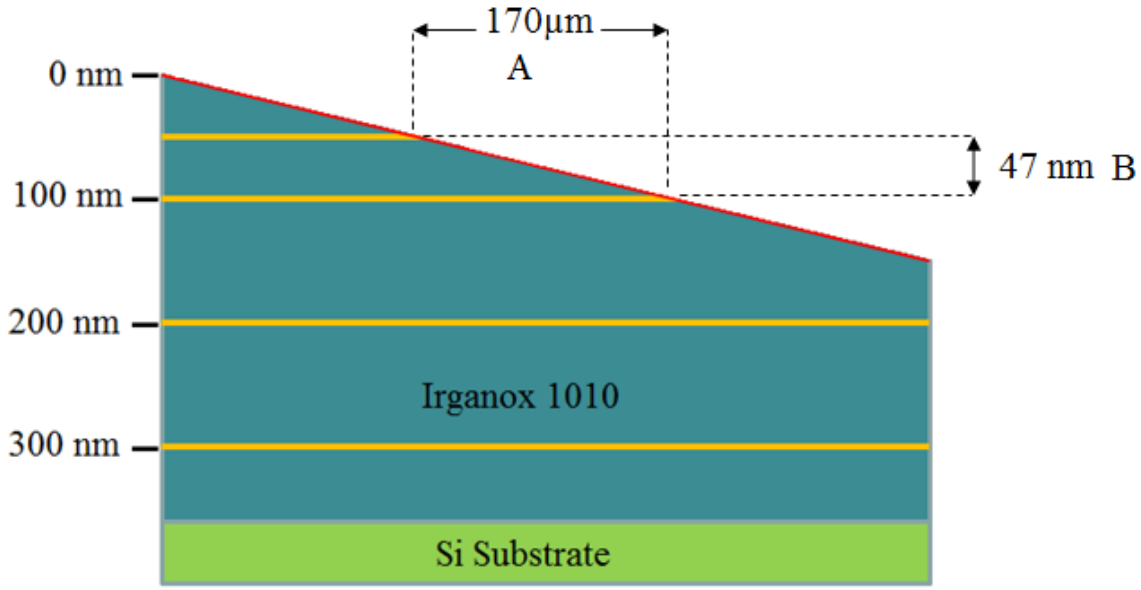
During the wedge depth profile, a SIMS image is recorded at the end of each cycle. In the example above, after 100 seconds of erosion, the deepest side of the crater reached 150 nm. The red line across the wedge crater represents the actual SIMS imaging surface during this sputter cycle. Then image of m/z 42 from this surface is shown as below representing the signal from Irganox 3114.



Then a line scan of m/z 42 across the crater (light blue arrow) is shown below. From the line scan data, we know the distance between the 1st and 2nd Irganox 3114 layer signal on this m/z 42 SIMS imaging is 170 µm (whole crater 450 µm). And from the structure of the film, we also know the depth distance between the 1st and 2nd layer is 47 nm. We can get the FWHM value of each layer from the figure below by locating them in the x axis, 84.7 µm for the 1st layer and 110 µm for the 2nd layer. They can be converted into a depth scale to get the depth resolution of each layer. The detail calculations are shown in the figure.



A further illustration of SIMS image scale and depth scale converting is shown as below.



Also, all of these figures and results represent one SIMS imaging cycle during the whole wedge depth profile. The wedge crater width in the y-direction stays the same at 450 µm. At this point, the deepest side is 150 nm and the angle of the wedge can be calculated as 0.019°. With continued wedge depth profile, the wedge crater will become deeper and angle will become larger till the substrate is reached corresponding to an angle of 0.050°.