

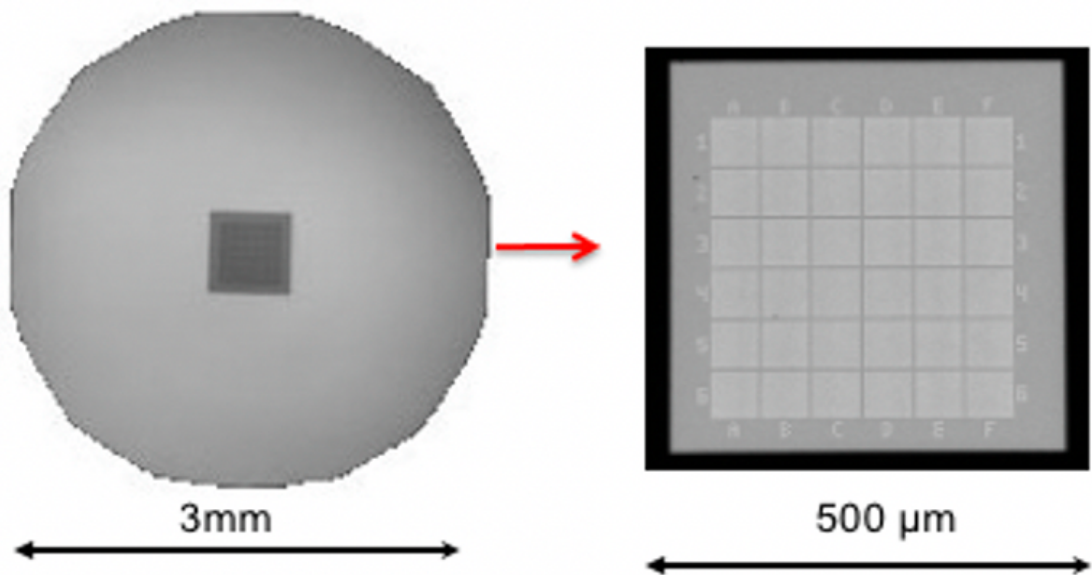
## ***Quantitative Assessment and Measurement of X-ray Detector Performance and Solid Angle in the Analytical Electron Microscope***

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### ***Supplemental Information: Penumbra Measurement Protocol using Ge/SiNx***

The author confirms that no financial benefits are involved with the recommendation of the test specimens or the procedure documented below.

The following steps document a systematic procedure for using a Ge/SiNx test specimen for the measurement of the penumbra of a combined XEDS/microscope system (Zaluzec et al 2016). This procedure utilizes a commercial specimen, GE20-SN20MP2Q05, which is a microporous 20 nm thick Ge/SiNx window, and can be obtained commercially from TEMWindows.com (Figure S1a). Alternatively, a uniformly thick, flat specimen of alternate compositions can also be employed, ideally with holes present to allow beam current verification without withdrawing the specimen.



*Figure S1a) Ge/SiNx test specimen (TEM Windows GE20-SN20MP2Q05) consisting of 20 nm of amorphous Ge on 20 nm of amorphous SiNx on a 100 μm thick 3mm diameter Si disk (left). Central window ~ 500 x 500 μm of the TEM window is magnified on the right.*

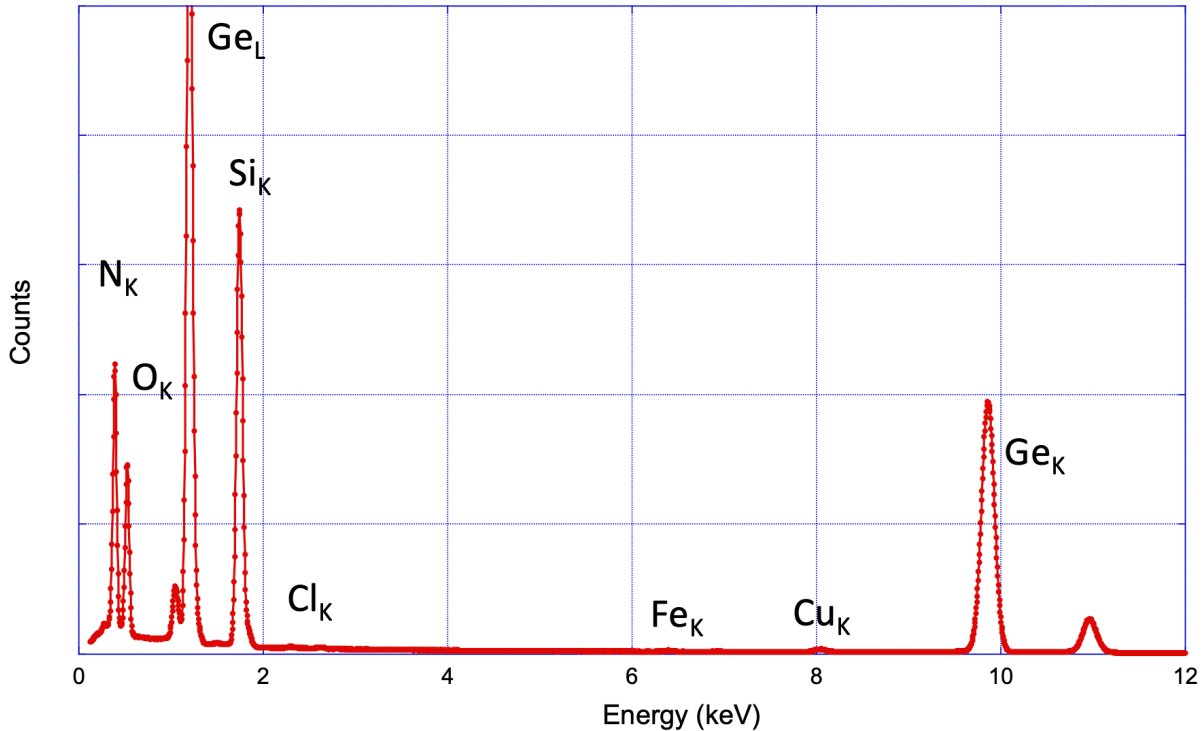
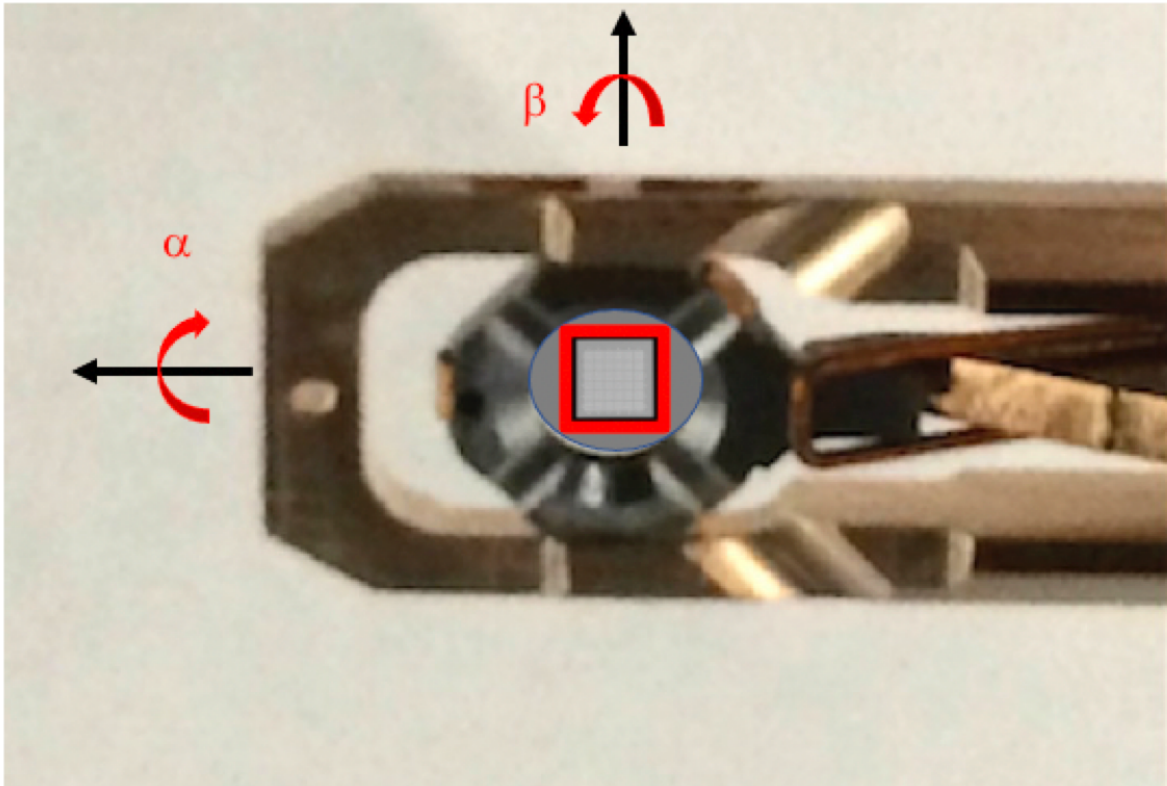


Figure S1b) Typical XEDS data set showing the spectral lines ( $Ge_K$ ,  $Ge_L$ ,  $Si_K$ ,  $N_K$ ,  $O_K$ ,  $Cl_K$ , and system peaks of  $Fe_K$  and  $Cu_K$  from GE20-SN20MP2Q05 at 200 kV, 600 Lsec, 200 pA.

The magnitude of any system peaks (C, Al, Fe, Co, Cu, Pt, Au, Mo) from the microscope or the detector (Al, Zr, Y, La, Pb, Sn) can be assessed by comparing their respective integrals to the Ge Ka peak (Figure S1b). Note that these system peaks will vary with holder and tilt. The small native system Si peak that will be present from the Si-based SDD cannot be assessed with this specimen, in addition, the O, and Cl specimen peaks resulting from the fabrication of the Ge/SiNx will nearly always be present (Zaluzec, 2015). This spectral profile can also be used for energy axis calibration using the Ge K, and the lower energy peaks (Si K, Ge L, or N K).

**Step 1.)** Insert Ge/SiN specimen in the holder and physically align the specimen as well as possible so that the 0.5x0.5 mm window (Red Square Figure S2) is both centered in the specimen stage and that the sides of the square window are aligned with the Alpha/Beta tilt axis, as shown in Figure S2.

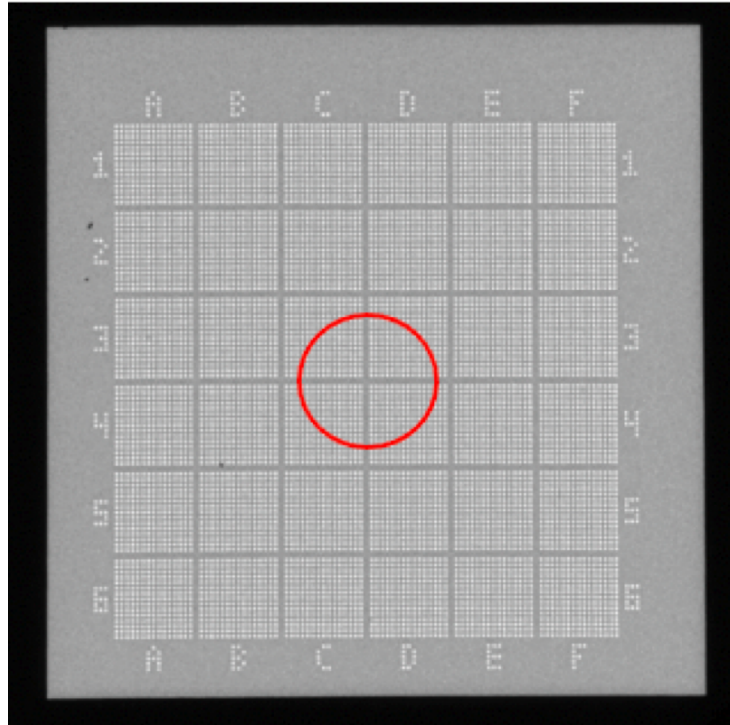


*Figure S2) Ge/SiNx test specimen mounted in a double tilt Beryllium Gimbal holder showing the optimum orientation of the specimen window with the major tilt  $\alpha$  and  $\beta$  tilt axes.*

**Step 2.)** It is recommended, if possible, to do a light quick plasma clean of the holder and specimen to remove any residual mobile hydrocarbon contamination. 15 seconds/10 Watts/150mTorr Argon is generally sufficient.

**Step 3.)** Set instrument accelerating voltage to desired value, and perform nominal TEM column alignment. Insert test specimen into column and adjust the holder to eucentric height and fine tune height to reach TEM focus condition. The penumbra procedure can be completely done in TEM mode, although equivalent results will be obtained in STEM mode.

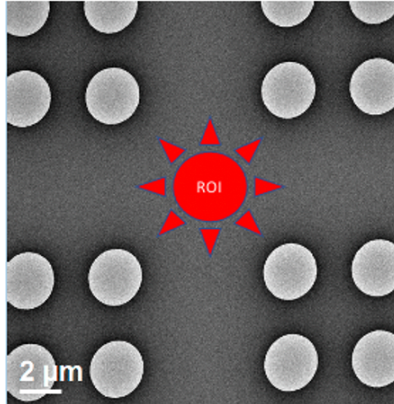
**Step 4.)** Using the coordinates labeled on the perimeter of the 0.5x0.5 mm grid (i.e.ABCDEF, 123456) find and translate to the center of the specimen (between C-D/3-4), illustrated as the red circle in Figure S3. If a good physical alignment was done upon mounting the test specimen in the holder this should be close to center when the goniometer is zeroed.



*Figure S3) Magnified TEM image of the center of the Ge/SiNx specimen indicating the location of the center of the test specimen*

**Step 5.)** Confirm that  $[0,0]$  coordinates of the alpha and beta tilts are correct. The tilt zero position can be wrong if the goniometer is not fully calibrated and centered, or if the specimen is not inserted properly and held down flat. A simple way to confirm this is to image one of the existing circular holes at 0 tilt, then tilt  $+45$  and  $-45$ . The circle should become an ellipse and the aspect ratio of the ellipse should be the same at  $\pm 45$ . If not then zero may not be true. Fix this, or note the position of true zero as necessary to compensate.

**Step 6.)** Return to true zero tilt position. Identify the center of the  $0.5 \times 0.5$  mm grid. This the region of interest (ROI) for measurements. This ROI will have a “wider”  $\sim 5$   $\mu\text{m}$  flat zone surrounded symmetrically by holes. (see figure S4). Store and save this location as well as the location of one of the 4 “holes” for beam current measurements. These is your target area for all measurements.



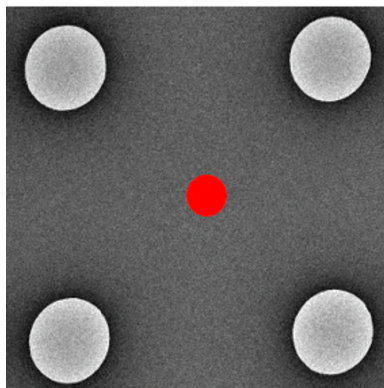
*Figure S4) Region of Interest and large “flat zone” of the test specimen located in between the periodic array of holes which will be used for beam current measurements without removing the specimen from the microscope.*

**Step 7.)** Confirm that upon tilting through the full range of the holder that this nominal area stays in the field of view for the tilt series. This will be reasonably achievable for the alpha tilt series, but beta tilts may be problematic as many goniometers are not double eucentric. If eucentric alignment cannot be achieved, measurements will require manually adjusting the holder position and height during data collection if the ROI moves off center.

**Step 8.)** Set up electron beam conditions.

If absolute solid angle measurements are to also be performed confirm that beam current monitor is calibrated. If absolute solid angle measurements are not the goal then this is absolute beam current calibration not essential, however, relative beam current measurements are still needed to insure that the system is stable during all data acquisition.

Go to a magnification of  $\sim 10\text{-}20\text{ kx}$ . This should be a reasonable magnification to allow both the ROI and some of the holes on viewing screen (Figure S5).



*Figure S5) Location of focused beam in center of ROI for measurements between 4 beam current measurement holes  $\sim 10\text{ kx}$*



Setup to measure the XEDS data

C1 aperture as beam defining  $\sim 70$   $\mu\text{m}$

C2 aperture as hole count aperture  $\sim 150$   $\mu\text{m}$

Spot size and Condensor lenses adjusted to obtain the following conditions:

Adjust the nominal electron beam diameter so that it is  $\leq 1/3$  the diameter of one of the holes (Figure S5). Adjust beam current to  $\sim 0.1$ -  $0.5$  nA, measurement of beam current should be made with the electron beam translated into one of the nearby holes. Adjustment of beam current can be done using best method for the instrument; i.e. monochromator, gun settings, or spot size. Confirm that beam current is stable, by measuring the "in-hole" electron beam signal throughout the course of the measurements. Once set do not change the beam in any way which changes the incident beam current.

Move the beam back to the ROI and check count rate on the x-ray detectors. A reasonable target should be  $\sim 0.5$ - $2$  kcps with dead time ( $< 10\%$ ). If multiple detectors are present the net count rate value should all be about the same (within about  $10\%$ ) on all detectors. If not equal then investigate why not.

**Step 9.)** Start XEDS measurements.

Go to  $\sim 10$ - $20$  kx i.e. a magnification so that allows one to see the ROI and nearest 4 surrounding holes so that you know you are centered. Diameter of the beam should be set to  $\sim 1/3$  a hole diameter. Confirm beam current value and stability by translating beam into hole and then recenter beam on ROI for XEDS measurement.

Confirm that hydrocarbon contamination is nearly absent during the measurement conditions (i.e. look for a  $C_K$  peak in your XEDS data). If not repeat plasma cleaning.

Set Live Time to 100 seconds, record beam current, microscope conditions, and tilt parameters. Record a spectrum from each individual detector, store spectral data with unique file name and record all metadata.

Start all measurements at Alpha=0 Beta =0, then increment Alpha by 5 degrees and repeat spectral measurement, store data and increment tilt until you reach the maximum Alpha Tilt. At each tilt, check visually that the beam is still centered in the ROI. There will always be some movement of the specimen. Manually recenter the stage and/or beam as needed so that ROI approximately remains at the same position at each tilt.

Check beam current in a nearby hole approximately every 5<sup>th</sup> measurement. After reaching the maximum tilt return and repeat [00] measurements of beam current and XEDS for stability check. Now repeat procedure but proceed to Maximum Negative tilt. When finished return to [00] repeat [00] measurements then start a new series using Beta Tilt.

## Step 10.) Data Analysis

Using your preferred spectral processing program process the individual spectra and create a table of results. The Ge K $\alpha$  intensity should be corrected for background and integrated over the full peak width, not the FWHM nor the Peak amplitude (Figure S6). Create a table (Table S1) of results, correct for specimen tilt, beam current and live time and plot (Figure S7). If the current is calibrated in nanoamps, then this data can also be used to calculate the absolute solid angle. For the example data shown below, measurements indicate that the penumbra is minimal for alpha tilt angles > 5 degrees and at a maximum at ~ - 20 degrees.

$$I = \int_{-\infty}^{+\infty} A \exp\left(\frac{-x^2}{2\sigma^2}\right) dx = \sqrt{2\pi} \sigma A$$

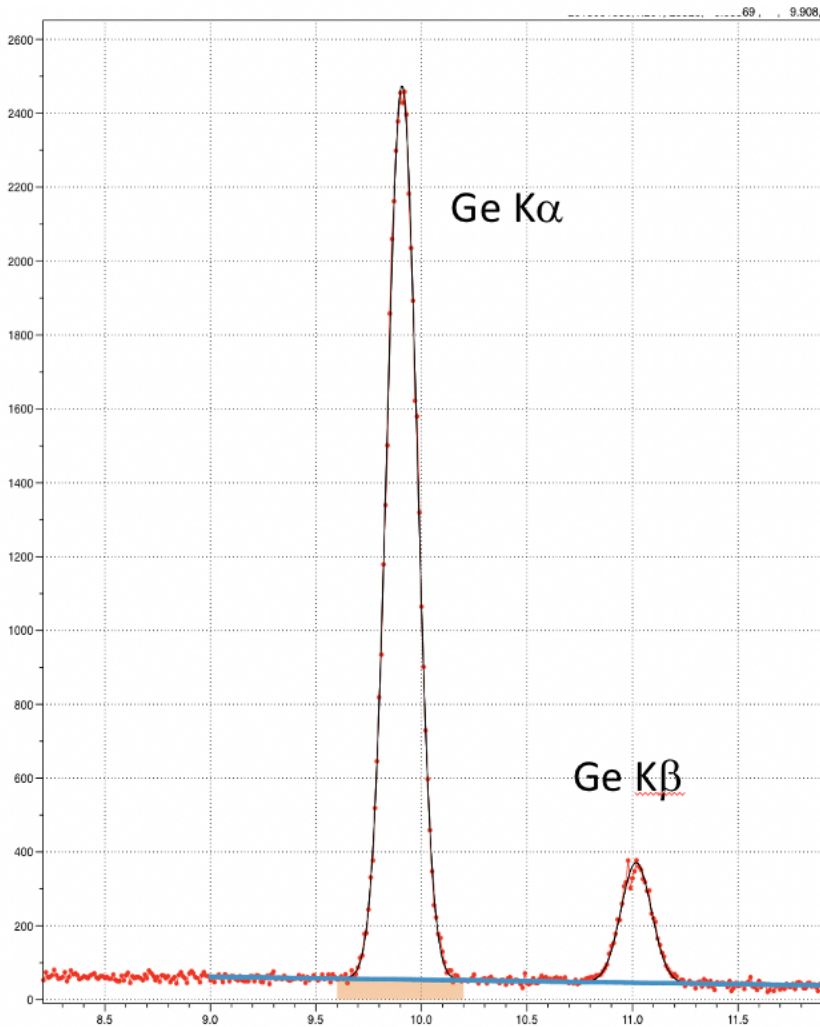


Figure S6) Integration of the GeK $\alpha$  above background. Note the protocol requires an Integration over the full peak and NOT a measurement of the peak Amplitude or FWHM.

Spectra File	Screen Current	kV	LTime (sec)	Holder $\alpha$ Tilt	Net Integral GeK $\alpha$	Net Integral GeK $\beta$	GeK $\alpha$ /Bgd	GeK $\alpha$ *Cos( $\alpha$ )/SC/LTime
2020022101	1.6	200	100	35	12327.67	1814.19	56.81	63.11
2020022102	1.6	200	100	30	12082.99	1796.89	59.47	65.40
2020022103	1.6	200	100	25	11513.52	1682.15	60.07	65.21
2020022104	1.6	200	100	20	11107.03	1722.78	58.89	65.23
2020022105	1.6	200	100	15	10641.2	1633.8	53.22	64.24
2020022106	1.6	200	100	10	10669.93	1598.24	63.74	65.67
2020022107	1.6	200	100	5	10370.18	1525.4	68.5	64.56
2020022108	1.6	200	100	0	9292.29	1344.56	66.12	58.07
2020022109	1.6	200	100	-5	7492.4	1100.18	57.64	46.64
2020022110	1.6	200	100	-10	5251.66	760.11	58.03	32.32
2020022111	1.6	200	100	-15	3172.23	470.47	20.9	19.15
2020022112	1.6	200	100	-20	2194.45	400.9	11.62	12.88
2020022113	1.6	200	100	-25	3046.05	450.29	12.99	17.25
2020022114	1.6	200	100	-30	5255.05	887.71	18.19	28.44
2020022115	1.6	200	100	-35	8561.18	1326.4	25.68	43.83

Table S1) Results from Experimental Penumbra measurements.

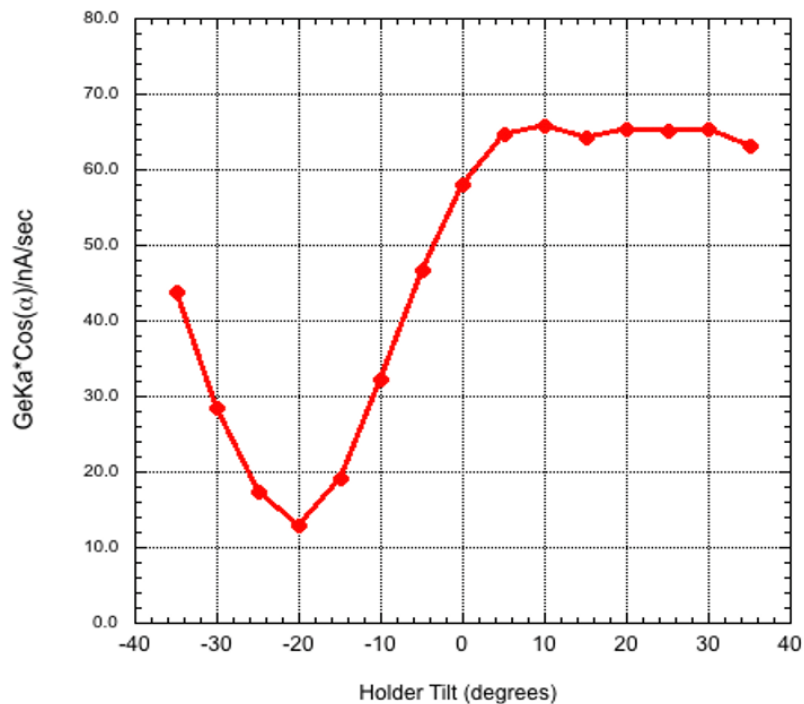


Figure S7) Graphical pot of the analyzed data from Table S1. In this analysis the penumbra of the system is minimized for Holder tilt angles > 5 degrees, and maximized at - 20 degrees.

## References

Zaluzec, N.J. (2015), "When is Si<sub>3</sub>N<sub>4</sub> not Si<sub>3</sub>N<sub>4</sub>? When it is a Low Stress SiN<sub>x</sub> Membrane Window"; Microsc. Microanal. 21, S3, 2015, 959-960 doi:10.1017/S1431927615005590

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