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New Simulation Capability for gamma rays mirror experiments

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1 Executive Summary

This report provides a description of the simulation toolkit developed at Lawrence Livermore National Laboratory to support the design of nuclear safeguards experiments using grazing incidence multilayer mirrors in the energy band of uranium (U) and plutonium (Pu) emission lines. This effort was motivated by the data analysis of a scoping experiment at the Irradiated Fuels Examination Facility (IFEL) at Oakridge National Laboratory in FY13 and of a benchmark experiment at the Idaho National Laboratory (INL) in FY14 that highlighted the need for predictive tools built around a ray-tracing capability.

This report presents the simulation toolkit and relevant results such as the simulated spectra for TMI, MOX, and ATM106 fuel rods based on spent fuel models provided by Los Alamos National Laboratory and for a virgin high ^{240}Pu-content fuel plate, as well as models of the IFEL and INL experiments implemented in the ray tracing tool. The beam position and height were validated against the INL ~60 keV americium data. Examples of alternate configurations of the optics or experimental set-up illustrate the future use of the simulation suite to guide the next IFEL experimental campaign.

2 Motivation

New safeguards technologies using nuclear spent fuel nondestructive assay based on direct measurements of fissile material emission lines aim to verify the plutonium (Pu) content of spent fuel assemblies. Over the past few years, grazing incidence multilayer mirrors have been developed to selectively reflect hard X-ray and soft gamma rays in the 90 to 420 keV energy band of U and Pu emission lines into a 2D high-purity germanium (HPGe) detector shielded from the line-of-sight radiation from spent fuel to overcome the combination of high rates and high background.[1][2][3]

A scoping experiment conducted at the Irradiated Fuels Examination Facility (IFEL) at Oakridge National Laboratory (ORNL) in June 2013 successfully demonstrated the band-pass properties of a stack of five multilayer mirrors.[4][5] The ^{137}Cs background and the difficulty to detect Pu characteristic X-rays led to the design of a benchmark experiment using virgin plutonium fuel plate of known material composition. The experiment conducted at the Idaho National Laboratory (INL) in September 2014 was successful in reflecting the 59.64 keV line of ^{241}Am off a single mirror, but Pu emissions in the 100 keV range were indistinguishable from background. [6]

The above experiments highlighted the need for predictive tools to estimate the optics response under various experimental conditions and inform the design of the final experimental set-up for direct measurements of U and Pu lines. Consequently, a simulation suite was developed that could give some insight in the design of the X-ray optics system as a function of source emission, multilayer coating characteristics and general experimental set-up such as source-mirror distance and collimation systems.

The three main goals were:

- Obtain models of the energy spectrum emitted by the three sources including characteristic X-rays and gamma rays from actinides and fission products,
- Develop a ray-tracing capability that will allow to simulate the beam reflected by flat mirror(s) within the experimental set-up
- Validate these capabilities against data from the INL experiments

Section 3 details all the steps of the simulation suite from source definition to expected reflected signal incident on the detector. Results for the INL and IFEL experiments are summarized in section 4 and an example presented in section 5 illustrates how the ray-tracing capability can be used to explore a different optics concept within the IFEL experimental set-up.

3 Simulation Capability

3.1 Source Energy Spectrum

The energy spectrum of photons escaping through the cladding of a fuel rod or a fuel plate or through the walls of a sample container is a required input for the ray tracing capability to properly weigh the reflected photon spectrum at the detector plane. Spectra of spent fuel rods are simulated with MCNP, and tallied in 0.5 keV wide energy bins.[7] Spent fuel, material composition and emission spectrum are obtained from burn-up calculations and

are provided by Dr. H. Trellue and her team at Los Alamos National Laboratory. For virgin fuel plates, line emission spectra are simulated with radsrcv1.6.[8]

3.2 Mirror Response

The mirror response, or reflectivity as a function of energy and angle, is calculated with IMD, a software for modeling the optical property of multilayer film developed in the IDL programming language (Exelis Visual Information Solutions, Boulder, Colorado).[9] IMD requires a detailed description of the substrate and multilayer characteristics including the substrate composition and micro-roughness, the bilayer materials and d-spacing, the number of bilayers, and the Γ or ratio of the high-Z layer thickness to the total thickness of the bilayer.

3.3 Beam Ray-Tracing

A ray tracing capability was developed in IDL that enables modeling of an experimental set-up including linear or planar sources, collimators, multiple flat mirrors or mirror stacks. The energy range and mirror angle range are user-defined, so is the step size for energy and angle. Stepping systematically through the energy and mirror angle ranges, the position and intensity of the reflected line is tallied at the detector plane. Simulations yield the energy spectrum, position and vertical extent of the reflected beam at the detector plane. Since the energy response of the detector and the background are not taken into account, analysis of the energy spectrum gives some insight into the ideal signal achievable under the experimental conditions modeled and could guide possible set-up optimization. The location and extent of the beam can be compared directly to available data.

3.4 Background

Background due to photon scatter in the experimental set-up can be estimated with classic Monte Carlo photon transport simulations granted the model includes enough details as demonstrated with the MCNP simulations of the ESRF experiments at 384, 508 and 644 keV. [2][10]

4 Results

Ray tracing simulations of the INL and IFEL experiments are presented below: the INL benchmark data set at ~60 keV is used to validate the ray tracing capability; optimal angles for several energy ranges are determined for the IFEL experiment. Note that ongoing work to simulate background in both experiments is not included in this report since the MCNP model of the INL experiment is currently being tested, and set-up information required to build the model of the IFEL experiment has just been gathered enabling the start of the modeling phase.

4.1 Mirror response

The recipe of the multilayer coatings deposited on the single mirror (INL) and the 5-mirror stack (IFEL) are essentially the same. They are both made of N=300 bilayers of tungsten carbide (WC)/silicon carbide (SiC) multilayers with a nominal period thickness d=15.2 Å. The Γ or ratio of the WC layer thickness to the total thickness of the bilayer differs slightly

therefore two distinct IMD models were used. It is worth noting the multilayer coating of the single mirror has been characterized over a wide range of energy from 850 eV to 644 keV resulting in an extensive validation of its model. The IMD response for the single multilayer mirror with a 60 keV incident beam is plotted in figure 1.

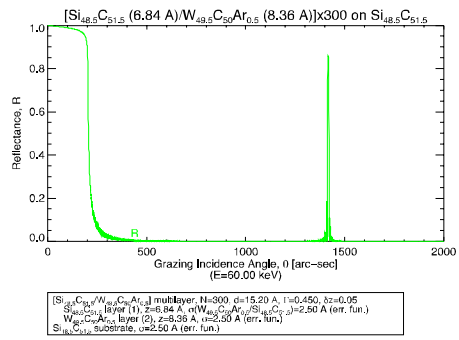

Figure 1. Single mirror response as a function of grazing for 60 keV incident photons.

4.2 INL experiment

The line emission spectrum included in the model of the ZPPR fuel plate was simulated with radsrcv1.6 using input data from report INL/MIS-14-33068.[8] The fuel plate and the experimental set-up are described in reference 12 and 6, respectively. The simulated spectrum in figure 2 shows that the intensity of the 59.54 keV americium line is several orders of magnitude larger than the lines in the 100 keV range and several of the lines with significant intensity in the 100 keV region result from ^{241}Am decay.

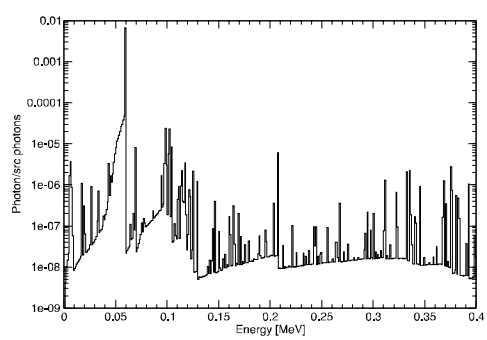

Figure 2. Simulated leakage spectrum for an INL ZPPR fuel plate with high ^{240}Pu content.

The geometric setup of the INL experiment and the simulated spectrum from 0 to 120 keV were used as input for the ray tracing simulation. Mirror angles considered here were:

- 1454 arcsec enhancing the reflection of the 59.54 keV ^{241}Americium (see figure 3)
- 840 arcsec enhances the energies between 100 and 105 keV for Pu K□ line

Ray□tracing simulations were performed for each grazing angle to obtain the reflected beam position, width and spectrum at the detector plane (figure 3 and 4).

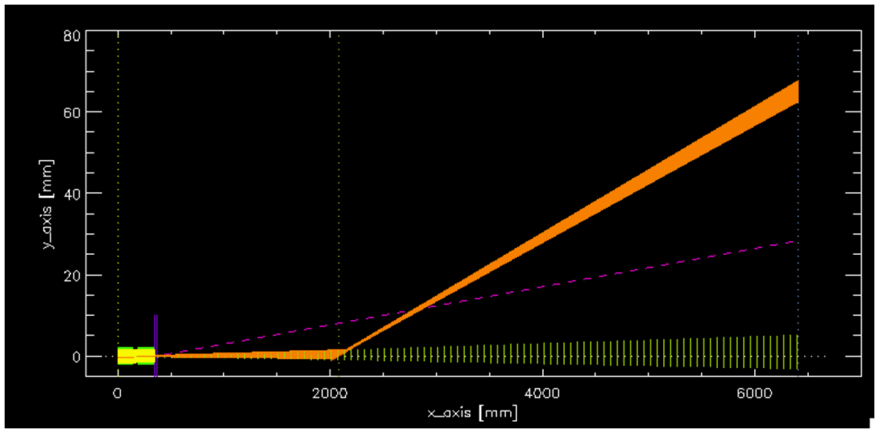

Figure 3. Ray-tracing simulation at 1454 arcsec. Simulated x-rays (orange) are emitted from the left and are reflected in the mirror towards the right where the detector would be located. The mirror is represented in red. In the bottom left corner, surfaces of the copper collimator are shown in green and purple. The region in yellow represents photon traces stopped by the collimator. The purple dotted line shows the field of view of the direct beam exiting the collimation system and the green vertical lines represent the area obstructed by the mirror.

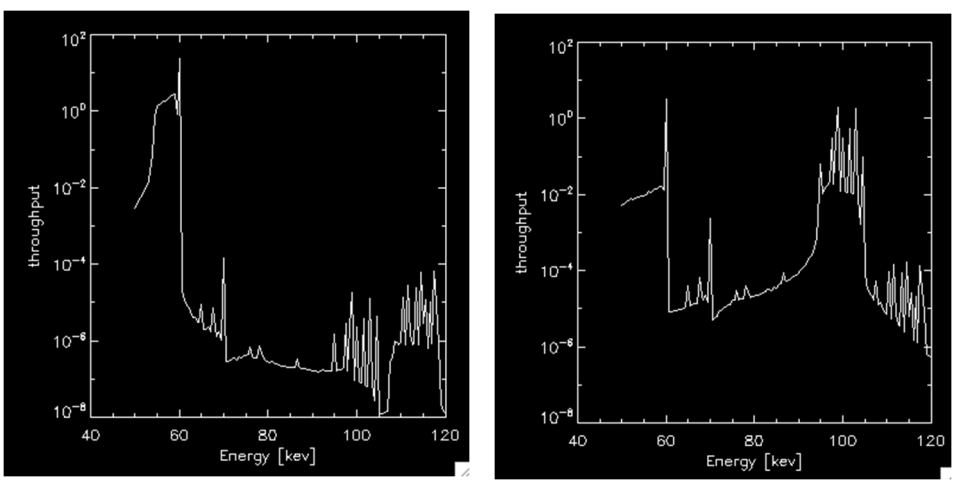

Figure 4. Ray-traced reflected spectra of the ZPPR fuel plate at the detector plane for absolute grazing angles of 1454 (left) and 840 (right) arcsec.

4.2.1 Validation at 59.54 keV

The INL benchmark data included six different absolute grazing angle measurements around the americium fluorescence line. Each run had a total of 450 seconds dwell time and data included the position and energy observed in the detector. 2D intensity maps were generated for the 57-61 keV (figure 5) and 100-105 keV energy range (figure 9).

The rocking curve presented in figure 6 suggests that the mirror reflects the americium line within a small range of angles albeit with decreasing efficiency. Ray tracing was performed for the six angles given in figure 6, and results are shown in figure 7 where color lines represent intensity at different grazing angles around the americium line and the white line is the summed intensity over all angles. Results were then normalized for a direct comparison with the data. The position, width and overall shape of the simulated intensity profiles match the data as shown in figure 8.

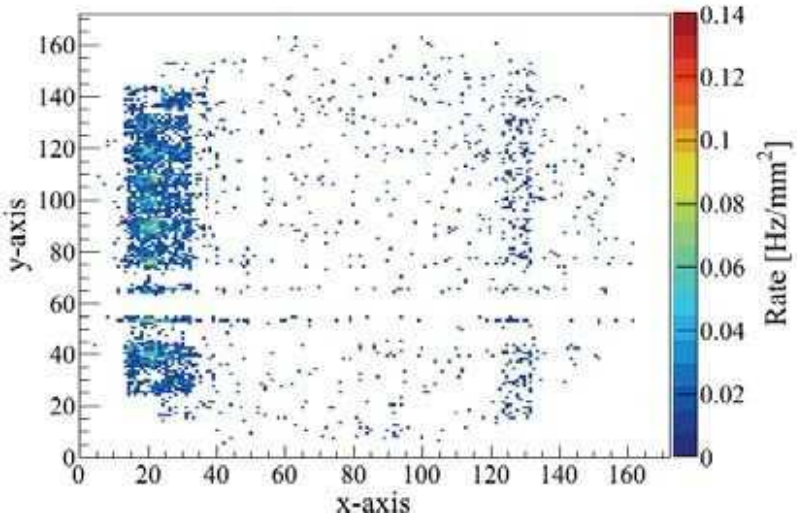

Figure 5. Intensity map in the 57-61 keV energy range at an absolute grazing angle of 1454 arcsec. The large band on the left is an image of the direct beam and the narrow vertical band on the right corresponds to the reflected 59.54 keV Americium line.

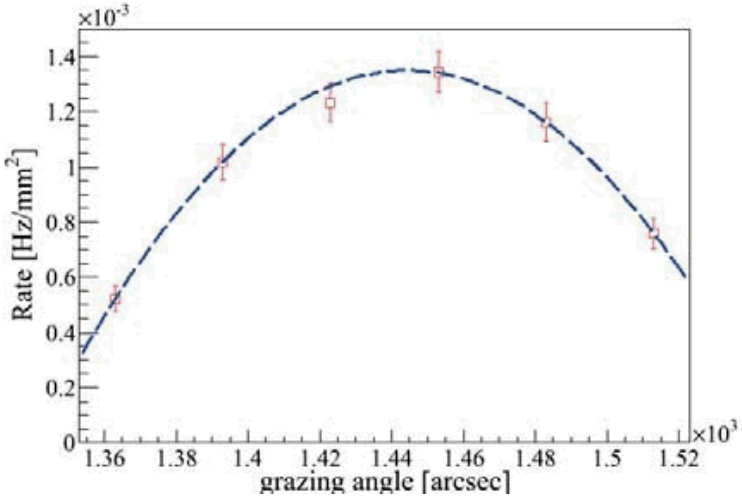

Figure 6. Rocking curve around the 59.54 keV Americium line with data (red square) and fit (dotted blue curve).

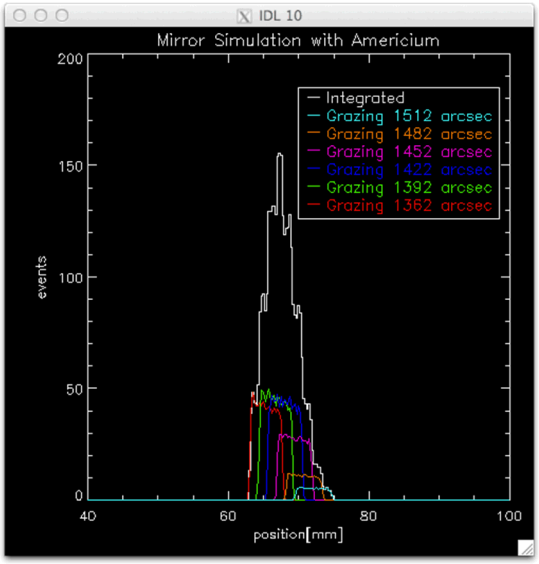

Figure 7. Simulated intensity profile for six absolute grazing angles. The white curve represents the sum of all six contributions.

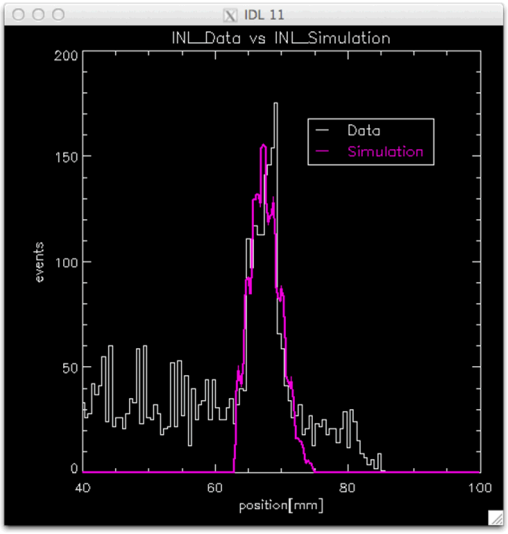

Figure 8. Comparison between the overall intensity profile from the INL americium data (white line) and the simulated overall intensity profile (purple line).

To measure the reflected beam around 100 keV, the mirror was set at an absolute angle of 840 arcsec. The 2D intensity map in figure 9 does not reveal any structure or band above background indicative of the presence of reflected lines unlike the result observed for Americium (see figure 5).

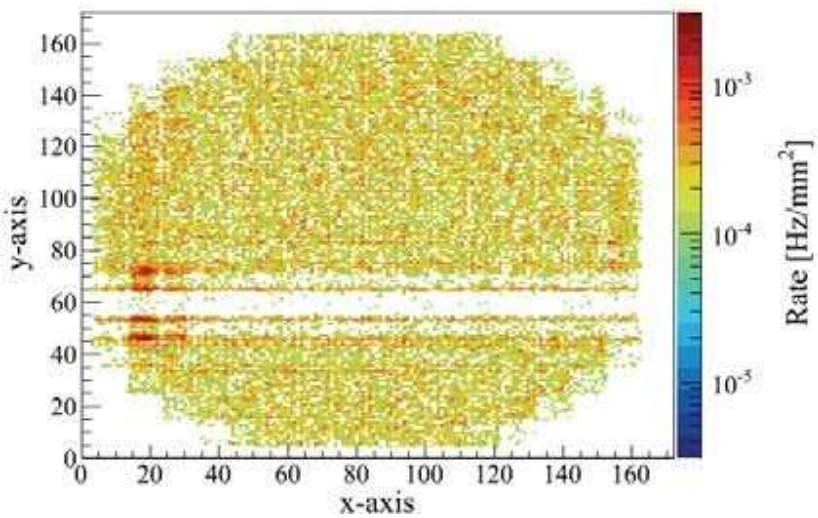

Figure 9. Intensity map in the 98-112 keV energy range for a mirror tilt angle of 840 arcsec.

As shown in figure 4, the difference between the throughput, or reflected beam intensity, at energies around the 60 keV line is almost 9 times larger than for lines around 100 keV. These results seem to indicate that the photon fluence incident on the mirror should be larger by at least an order of magnitude to observe a signal in the 100 keV region.

4.2.2 Maximizing photon fluence at 100 keV

The model was modified to explore different setups with the goal to maximize photon fluence incident on the mirror: the collimated source was moved closer to the center of the mirror from 2084 mm to 750 and 500 mm (see figure 10). Since the collimation was left unchanged, the mirror has a larger angular acceptance and lower energy photons coming in at steeper angles will also be reflected on the detector plane as shown in the spectra in figure 11.

The optimal position of the center of the mirror is at 750 mm from the source and in this configuration the total intensity is 4 times larger than in the setup used for our measurements. The simulation also shows that half of the signal is due to the Americium line, and that moving the mirror any closer will further increase the americium contribution. While an imaging detector provides a direct view of the reflection position, these results clearly show that a detector with high-energy resolution is key.

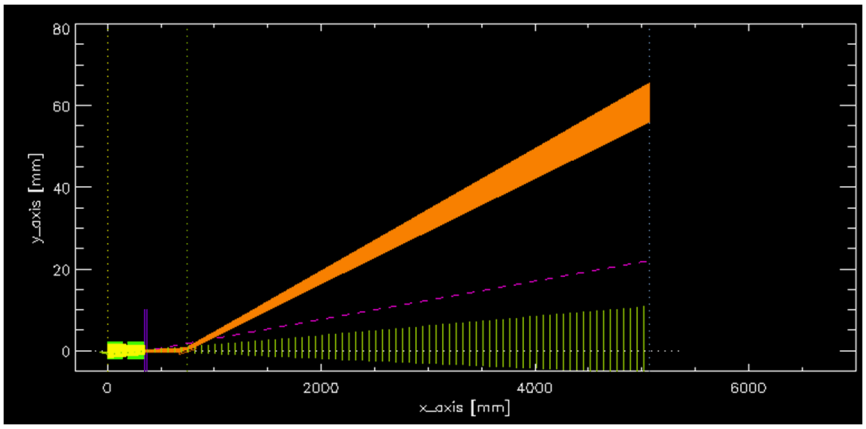

Figure 10. Ray tracing simulation at 840 arcsec and mirror centered at 750mm from the source.

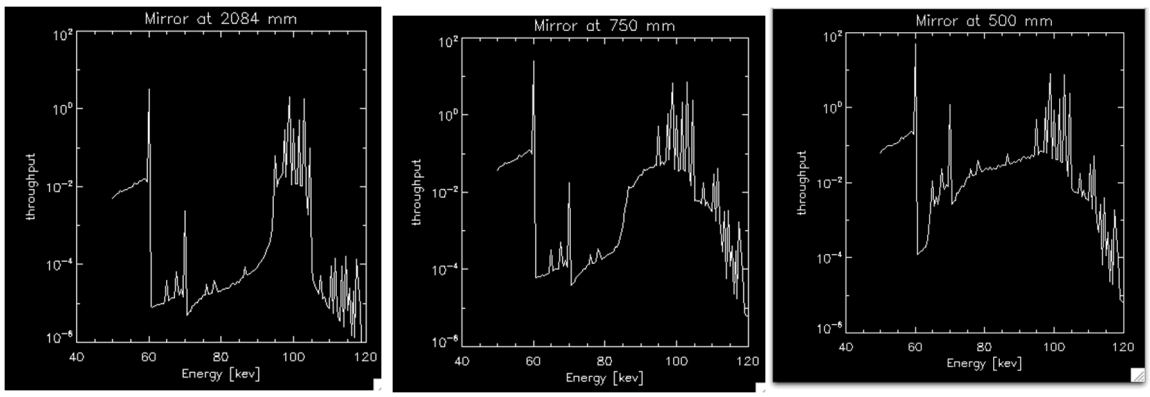

Figure 11. Energy spectrum of the reflected beam at the detector plane for a mirror angle of 840 arcsec and the mirror centered at 2084 mm (left), 750 mm (center) and 500 mm (right) from the fuel plate.

4.3 IFEL Experiment

Details of the set-up can be found in reference 11. A collimated beam emitted by either Three-Mile Island (TMI) or Mixed oxide (MOX) spent fuel rods was reflected off a 5-stack multilayer coated mirror into a 2D imaging detector. Characteristics X-ray of uranium were detected during the IFEL experiment for the TMI and the MOX fuel rods, it was determined that further experiments would be required to isolate the Pu characteristics X-rays.[5]

4.3.1 Sources

Photon leakage spectra for TMI, MOX and ATM106 spent fuel rods were simulated with MCNP. The TMI and MOX rods were used at IFEL (see figure 12), while ATM106 spent fuel samples are available at PNNL. The latter spectrum was simulated to evaluate the possibility of using this fuel for a hard X-ray mirror experiment at PNNL.

4.3.2 Mirror angle optimization

The ray tracing simulations include the geometrical details of the experimental setup at IFEL and reproduce the response of the 5-stack mirror used as band pass filter. The input energy spectrum ranges from 0 to 300 keV as shown in figure 12. The work presented here focuses on the energy region from 100 to 135 keV that contains the $K_{\alpha 1}$ characteristic X-ray

from Pu at 103. 76 keV and two nuclear lines noticeable in the simulated spectra of the MOX and TMI fuels, namely ^{155}Eu at 123.07 keV and ^{144}Ce at 133.51 keV. These lines could be of interest for calibration purpose. Ray-tracing simulations are used to determine the best mirror angle for each line.

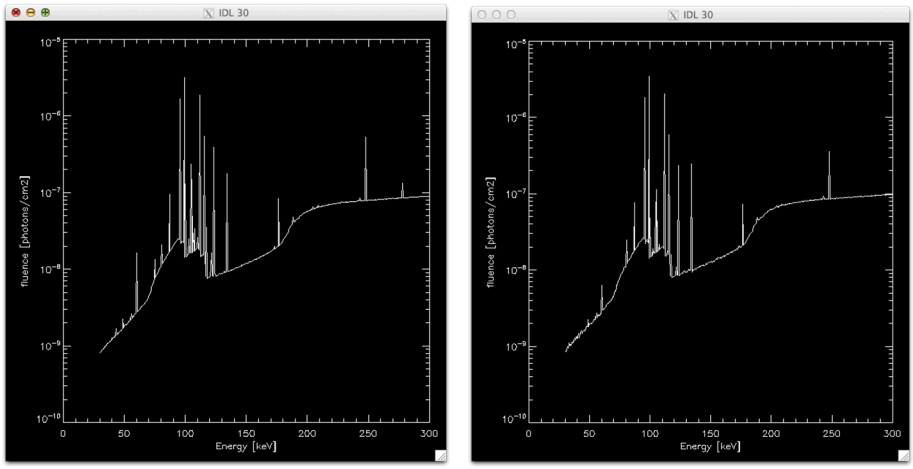

Figure 12. Input spectra for ray-tracing code in the 0 to 300 keV for a) MOX, b) TMI. spent fuel rods.

Preferred grazing angles were determined for energy ranges from 100 to 105 keV, 120 to125 keV, and 130-135 keV by comparing the intensity of the reflected beam in a given range of energy relative to the total intensity integrated from 0 to 300 keV for mirror angles ranging from 500 to 1000 arcsec.

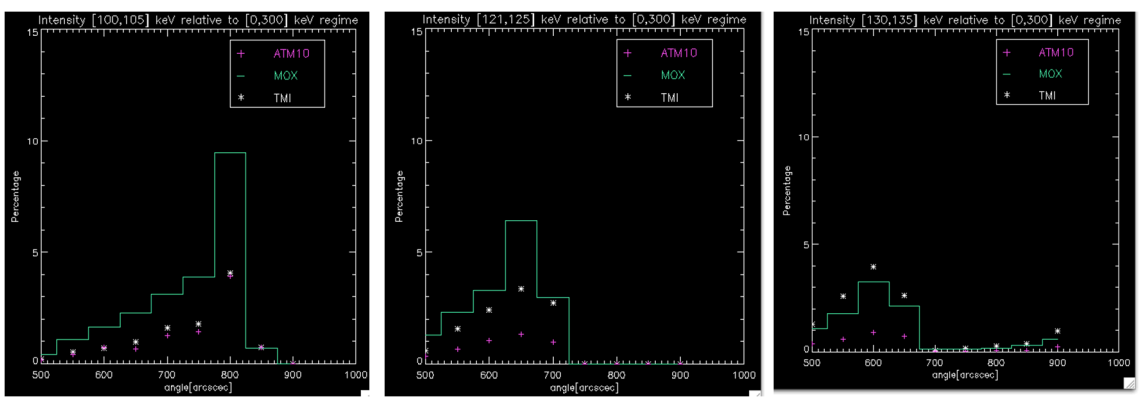

Figure 13. Intensity of the a) 100 to 105 keV, b) 121 to 125 keV, and c) 130 to 135 keV range for the MOX (green line) TMI (white asterisks) and ATM106 (magenta crosses) fuels relative to the total reflected intensity integrated over 0 to 300 keV.

Results shown in figure 13 indicate that optimum mirror angles are:

- 800 arcsec for energies between 100 and 105 keV around U and Pu X-rays
- 650 arcsec for energies between 121 and 125 keV around the ^{155}Eu line
- 600 arcsec for energies between 130 and 135 keV around the ^{144}Ce line

Ray-tracing simulations for the TMI and MOX fuels were then performed at each grazing angle to obtain the position and vertical extent of the reflected beam at the detector plane

and spectral line intensities in the 0 to 300 keV range. Figure 14 shows the reflected beam size increases at higher energies due to lower grazing angles and the beam position shifts closer to the collimated source axis and figure 15 illustrates the mirror passband as a function of mirror angle. At 800 arcsec, lines outside the 85-110 keV range are suppressed, and the Pu $K_{\alpha 1}$ emission line is more prominent in the MOX fuel spectrum. The intensity of the 133.51 keV line increases when the mirror is tilted from 650 and 600 arcsec.

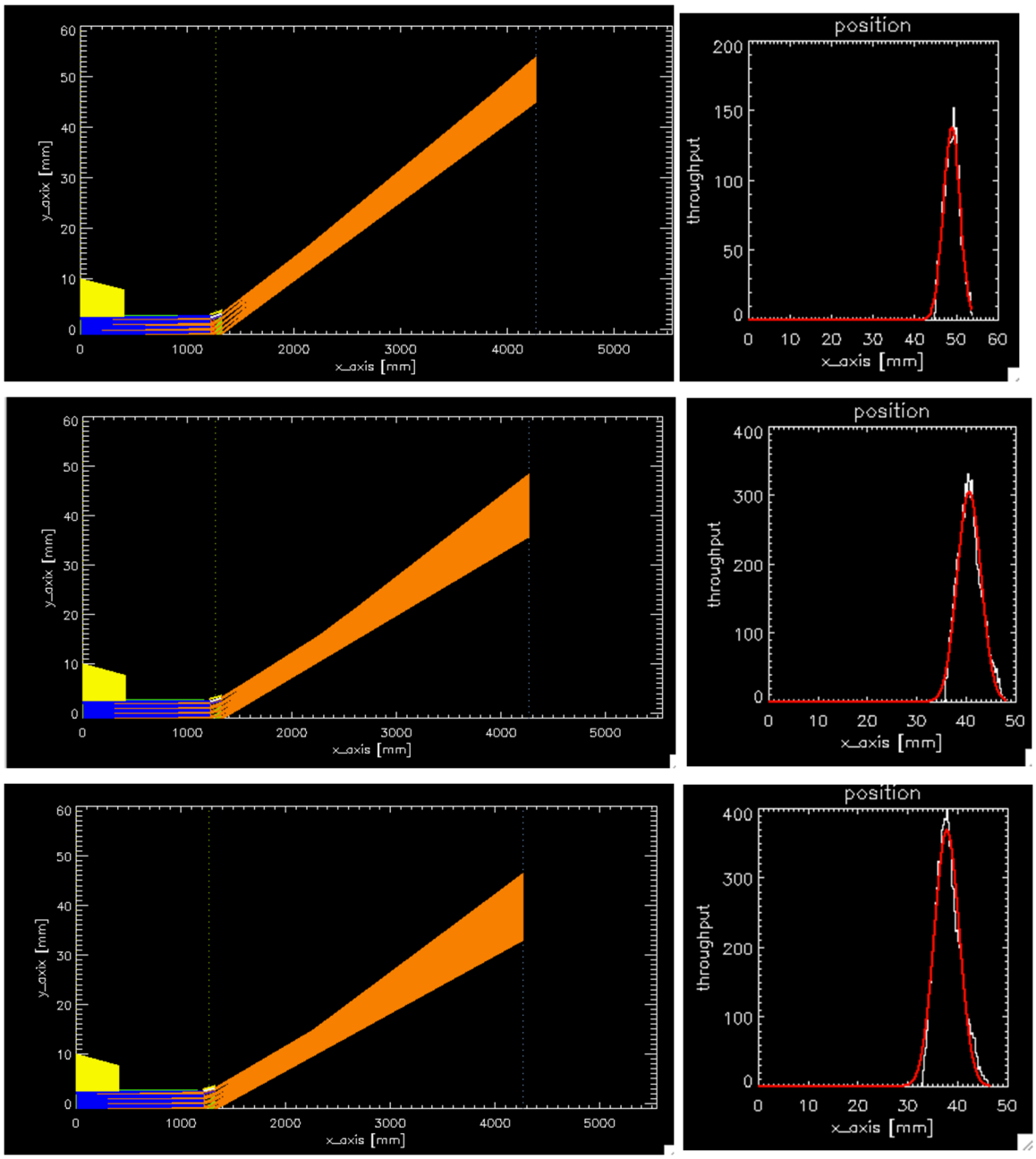

Figure 14. Ray tracing simulation (left) and expected position of the beam in the detector plane (right) at 800 arcsec or 100- 105 keV (top), 650 arcsec or 121-125 keV (middle) and 600 arcsec or 130-135 keV (bottom). The red line is a Gaussian fit to the simulation (white).

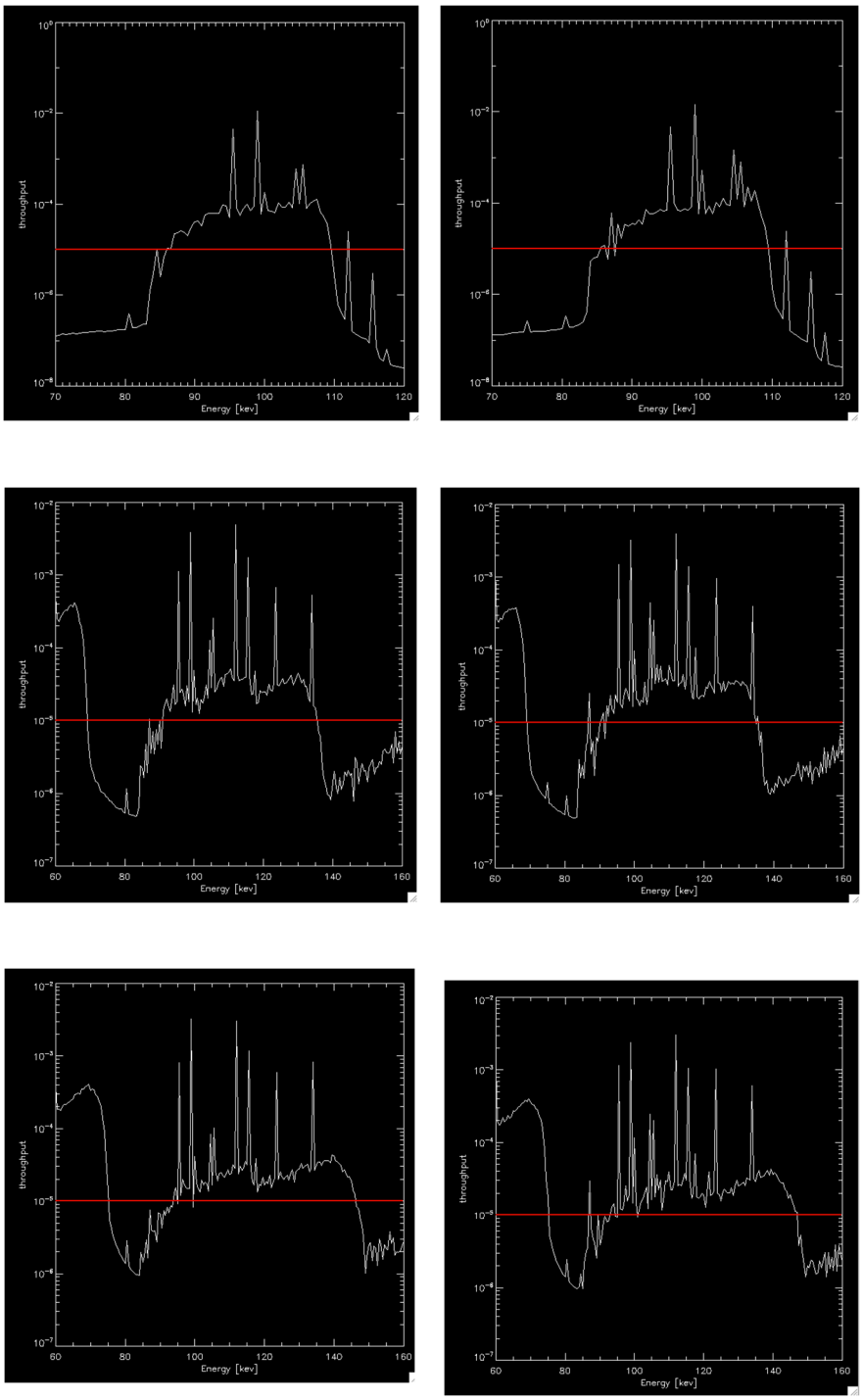

Figure 15. Reflected spectra of the TMI (left) and MOX (right) fuel rods at the detector plane for an absolute grazing angle of 800 arsec (top), 650 arcsec (middle), and 600 arcsec (bottom).

5 Possible optic design for IFEL experiment

The new simulation toolkit opens the possibility to explore the potential cost/benefit of different experimental set-up and optics choices when planning an experiment. Preliminary simulations of experimental set-ups with either two or three successive single mirrors angled at 840 arcsec with respect to the beam or reflected beam are shown in figure 16 and 17 to illustrate possible solutions for the upcoming experimental campaign at IFEL in FY16. All mirrors have characteristics similar to that of the mirror used in the INL experiment. The initial results in figure 17 seems to indicate that a two-bounce system will already suppress most of the signal in the energy band outside of 90-110 keV and could be sufficient compared to a more complex system with three mirrors. However an in-depth study of both systems is required.

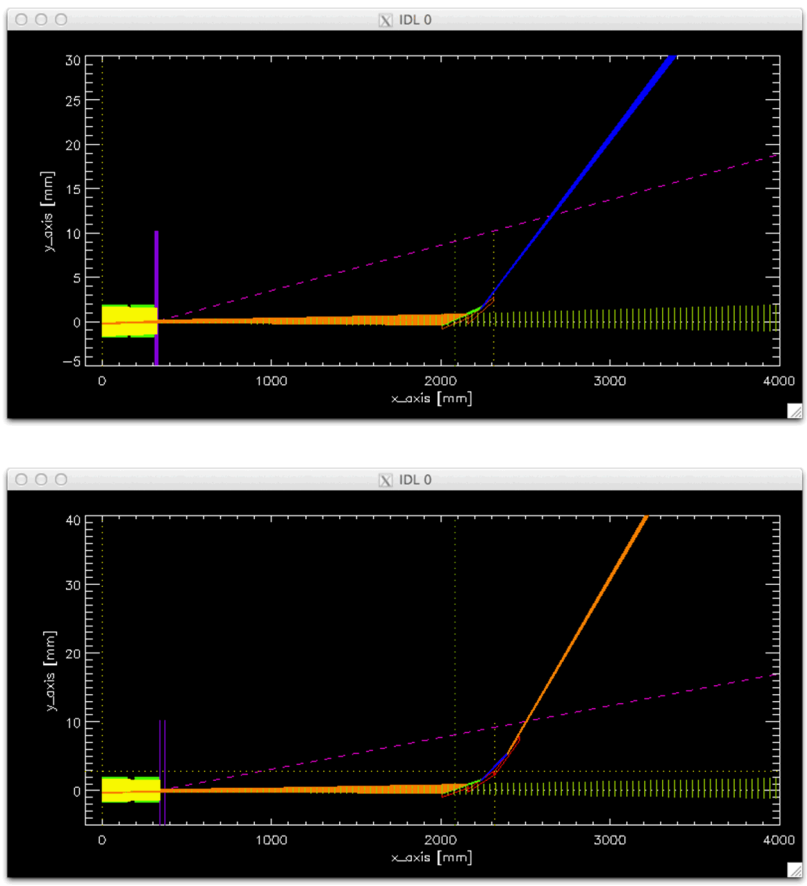

Figure 16. Experimental set-up for a gamma ray mirror measurement of Pu characteristic X-rays with two mirrors (top) and three mirrors (bottom).

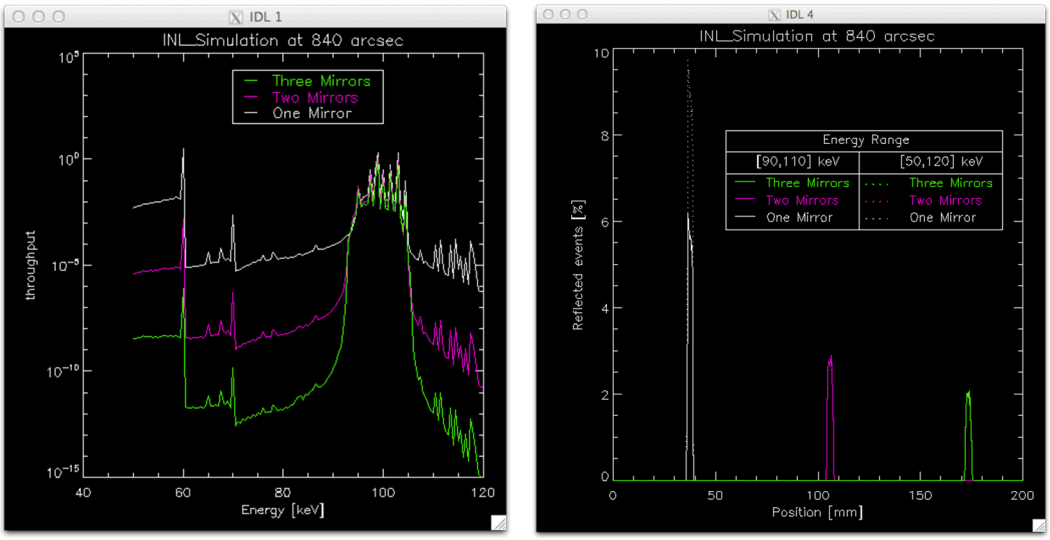

Figure 17. Reflected beam energy spectra (left) and position (right) for experimental set up with one (white), two (magenta), and three multilayer mirrors (green). The right plot also illustrates the loss of signal observed in the range 90-110 keV (full lines) and 50 to 120 keV (dotted lines) as a percentage of the total number of events between 50 and 300 keV when increasing the number of reflections.

6 Conclusion

Over the last year, a suite of models and simulation tools was assembled to enable gamma-ray mirror experiment analysis and design. Two experiments conducted at IFEL and INL respectively, were modeled and initial validation against data from the INL experiment showed that beam position and height were reproduced. Finally, alternative configurations of the experimental set-up or optics used in these two experiments were explored to showcase how this new simulation capability will be used to drive future experimental design.

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