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Supplementary information: Scintillating Thin Film Design for Ultimate High Resolution X-ray Imaging†

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Particle contributions

To further investigate the effects arising when exceeding K-edge energies of elements comprised in the Single Crystalline Film (SCF) and substrate the contributions from the different particles were isolated. With the simulation tool we have isolated the deposited energy distribution of the following particle types: primary X-rays, secondary X-rays, photoelectrons created from interactions with either primary or secondary X-rays and resulting electrons created from electron-electron scattering. It is important to note that in reality, the X-rays do not deposit the energy directly, but generate secondary electrons, that eventually deposit energy. In the Monte Carlo model, an energy threshold is defined for the production of the secondary particles, meaning that secondary particles with an energy lower than the threshold are not generated. The remaining energy is hence counted as deposited by the X-rays in the position of the interaction. The energy threshold is user defined and was set at 250 eV. This approximation is sufficiently accurate since the attenuation length for electrons at 250 eV in the investigated materials is below the spatial resolution range which is studied. It is important to keep in mind that the energy deposited by X-rays depends on the production threshold. In our case, it corresponds to the amount of energy deposited by electrons with a diffusion length shorter than the size of the voxel defined in the simulation. Since we are working at energies lower than 100 keV the contribution from Compton electrons are insignificant, which is why they are not included in the considerations here.

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‡ Additional footnotes to the title and authors can be included *e.g.* 'Present address:' or 'These authors contributed equally to this work' as above using the symbols: ‡, §, and ¶. Please place the appropriate symbol next to the author's name and include a `\footnotetext` entry in the the correct place in the list.

To study the effect on the LSF and MTF when working at X-ray energies around the K-edge of an element in the substrate, we performed simulations of a LAG:Ce SCF supported by a YAG substrate at energies varying from 14 to 24 keV. In Figure S1 the isolated LSFs for the different particle contributions at these energies are presented. All the types of secondary particles (secondary X-rays, secondary photoelectrons and electrons from electron-electron scattering events) deposit significantly more energy far from the initial interaction point when the incoming X-ray energies exceed the substrate K-edge (here Y: 17.04 keV). This is in agreement with fluorescence X-rays generated in the substrate and then interact and deposit energy in the SCF. The primary photoelectrons only deposit energy continuously further away corresponding to the increasing energy they will have from the photoelectric event.

To study the effect on the LSF and MTF when working at X-ray energies around the K-edge of an element in the SCF, we performed simulations of a LAG:Ce SCF supported by a YAG substrate at energies varying from 60 to 70 keV. In Figure S2 the isolated LSFs for the different particle contributions at these energies are presented. When exceeding the SCF K-edge, all types of generated electrons (primary and secondary photoelectrons and resulting electrons from electron-electron scattering) deposit more of their energy at the initial point of interaction giving rise to the sudden decrease in the LSF. This is a result of the increased probability of the photoelectric effect when the incoming X-ray beam have energy above the SCF K-edge. The X-ray fluorescence (secondary X-rays) also slightly reduce the tails of the LSF but due to their large energy and therefore large attenuation length most will escape from the SCF and do not contribute considerably to the image. However, the photoelectrons have sufficiently low energy resulting in short attenuation length, leading them to deposit energy very close to the initial point of interaction. The continuous improvement is caused by photoelectrons created from primary X-rays interactions and subsequent electrons from electron-electron scattering events. We ascribe this effect to the

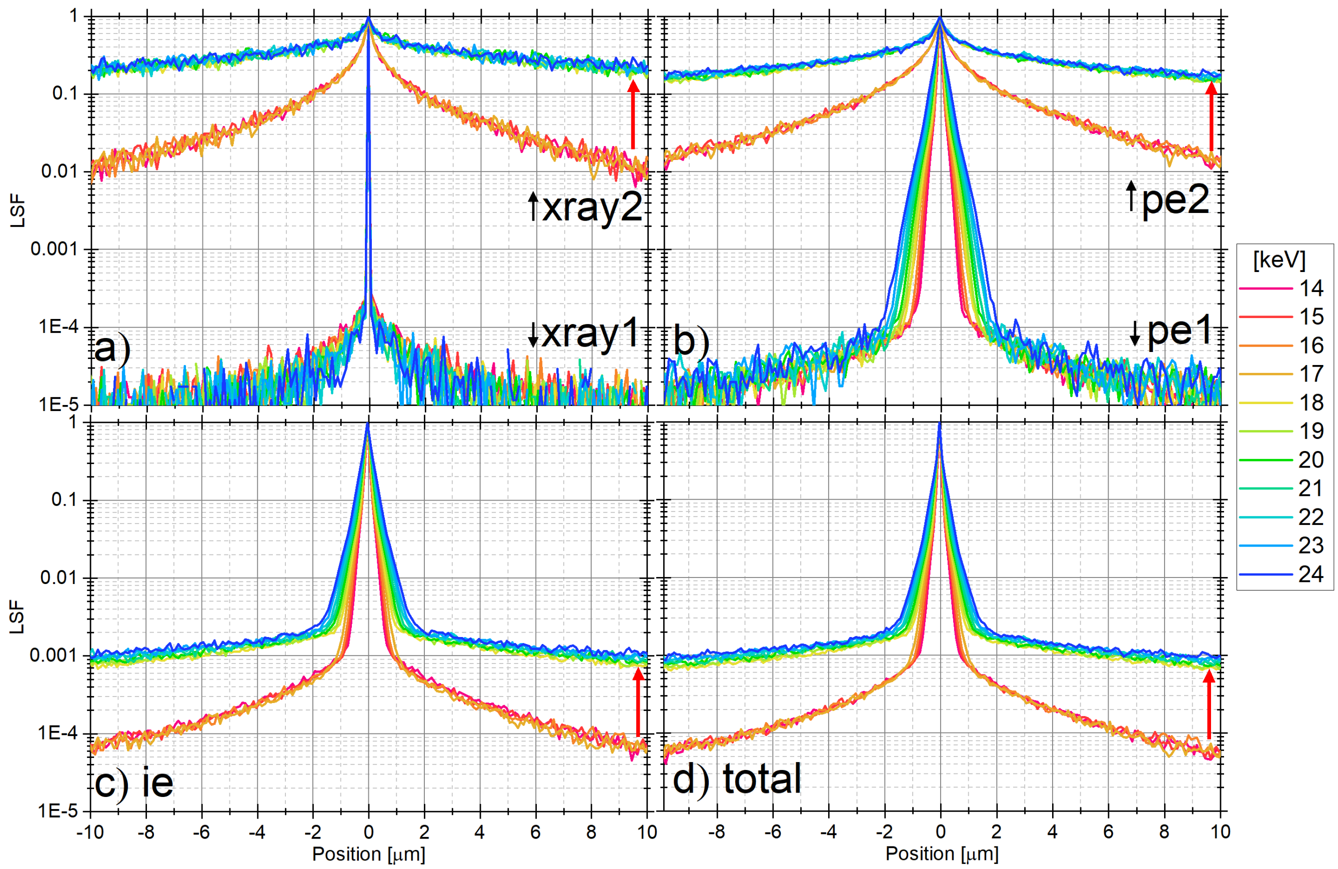

Figure S 1 Isolated LSFs representing the contributions from the different particle types. a) Primary X-rays (xray1) and secondary X-rays (xray2), b) photoelectrons created from primary X-rays (pe1) and secondary X-rays (pe2), c) resulting electrons from electron-electron scattering (ei) and d) the total LSF for better comparison. The simulations are conducted for a 5 μm LAG film supported by 150 μm YAG substrate at X-ray energies from 14 to 24 keV. K-edge energy of Y is 17.04 keV.

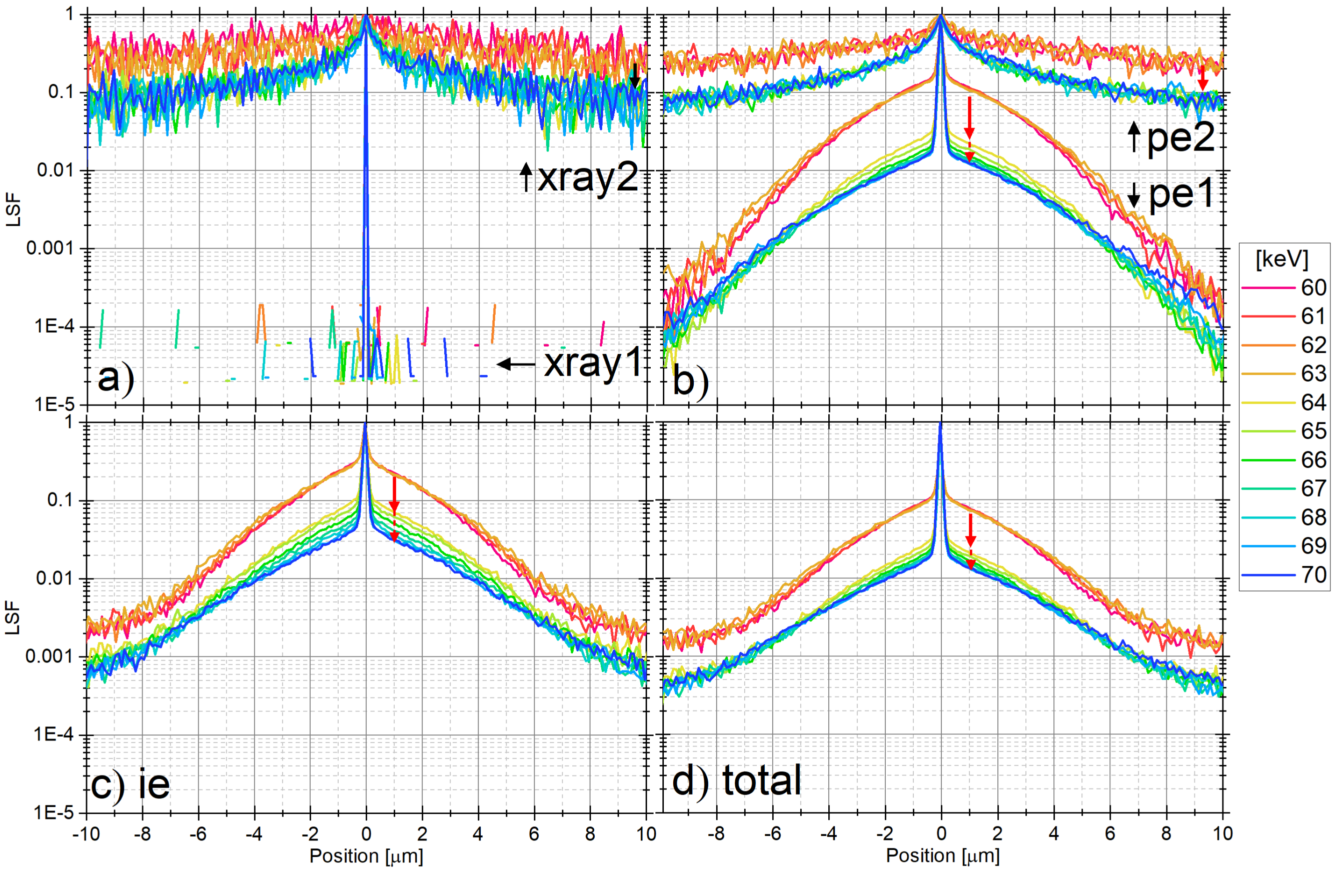

Figure S 2 Isolated LSFs representing the contributions from the different particle types. a) Primary X-rays (xray1) and secondary X-rays (xray2), b) photoelectrons created from primary X-rays (pe1) and secondary X-rays (pe2), c) resulting electrons from electron-electron scattering (ei) and d) the total LSF for better comparison. The simulations are conducted for a 5 μm LAG film supported by 150 μm YAG substrate at X-ray energies from 60 to 70 keV. K-edge of Lu is 63.3 keV.

increase of local energy deposition due the increase of the X-ray beam energy. Indeed, the energy of the primary electron will be $\sim$ 0.7-6.7 keV (assuming they are created from 64-70 keV primary X-ray photons and the K-shell of Lu) the attenuation length will typically be shorter than 0.5 μm (CSDA, NIST [1]), thereby containing the resulting energy deposit very close to the initial point of interaction.

Blurring by optics

The optics applied for the X-ray imaging experiment at different energies has a big influence on the quality of the final image. In order to emphasize this, we performed additional simulations for some of the SCFs with NA: 0.15, 0.45 and 0.80 at different energies as well: 10, 30, 60 and 100 keV. The results from these simulations are summarized in Figure S3.

If your experiment do not require a spatial resolution better than 2.5 μm (Rayleigh criterion), a NA of 0.15 is sufficient and a thicker SCF scintillator can be applied as long as it is not exceeding the depth of field.

At low energy (10 keV) light diffraction and thereby the optics are limiting the spatial resolution. If we included SCFs with a thickness larger than the depth of field as well, we would have observed the degradation caused by defocus.

The role of the specific material becomes crucial when reaching higher energies above K-edge energies in the substrates especially.

Figure of merit

In the main article the figure of merit is constructed from the MTF at 500 lp/mm and the energy deposited in the SCF. The MTF at 500 lp/mm is revealing how good a contrast you have for 1 μm features. This value is chosen because the study is focused on micron to sub-micron resolution X-ray imaging. To expand the view, we here present the figure of merit calculated for the MTF at 100, 500 and 1000 lp/mm (features of size 5, 1 and 0.5 μm) that is presented in Figure S4. The figure of merit can therefore easily be adjusted and applied accordingly to the needs of the X-ray imaging experiment.

Experimental method

As also described in the paper, the MTFs was experimentally obtained by using the slanted edge method [2,3]. A 525 μm thick GaAs edge carefully cleaved and positioned 1-3 mm away from the scintillator was used to absorb part of the X-ray beam. The acquired edge images were corrected by flat-field and dark images and the Edge Spread Function (ESF) was computed as a step function along one direction while being constant in the other. Subsequently, the derivative gives the Line Spread Function (LSF) and a Fast Fourier Transform (FFT) provides the Modulation Transfer Function (MTF). This procedure is presented in Figure S5.

Notes and references

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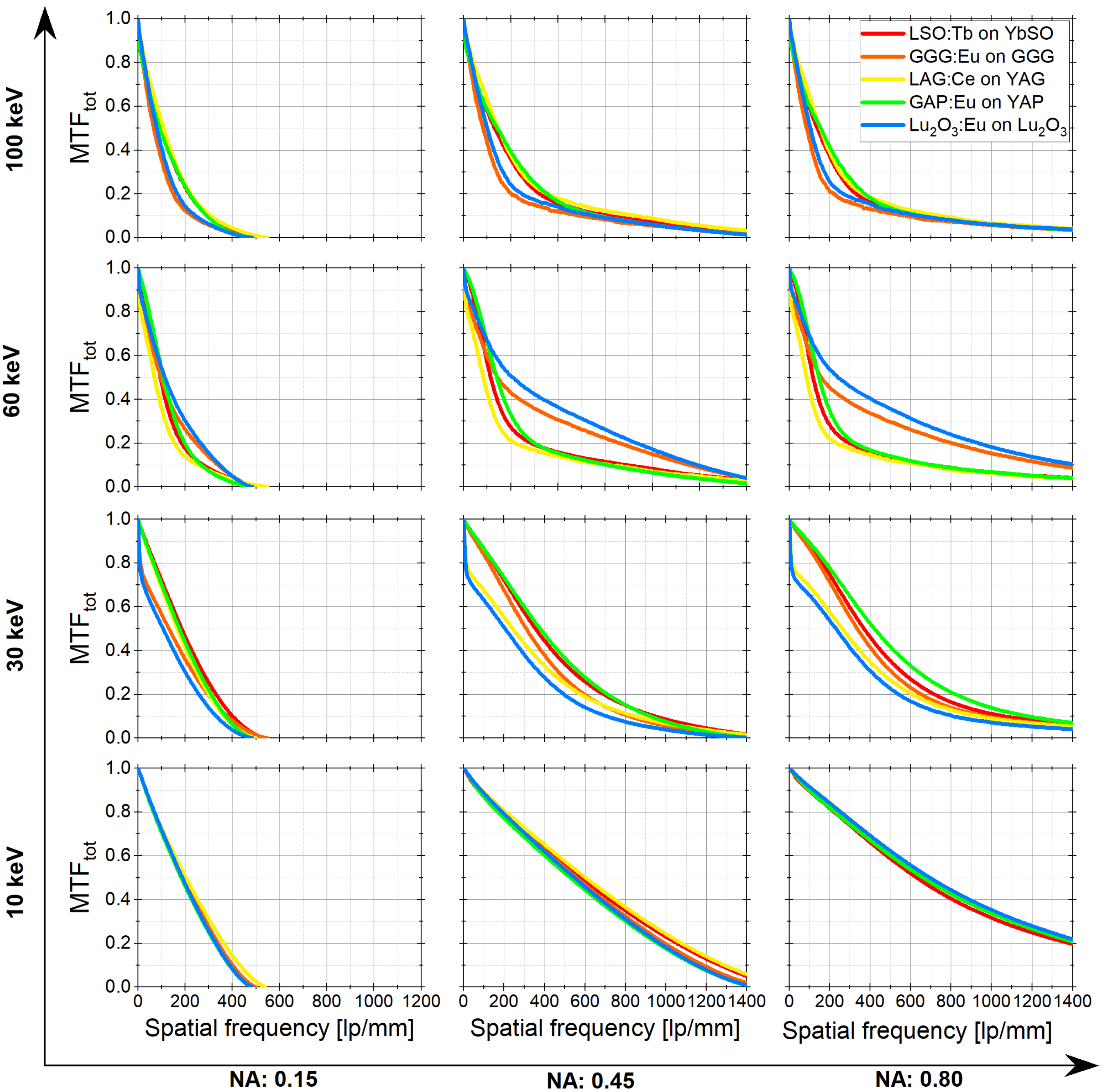

Figure S 3 Simulated MTF$_{tot}$ of various 5 μm SCFs supported by 150 μm substrates evaluated for various NA and X-ray energies. Materials are listed in the legend, NA and X-ray energy is indicated on the two axis.

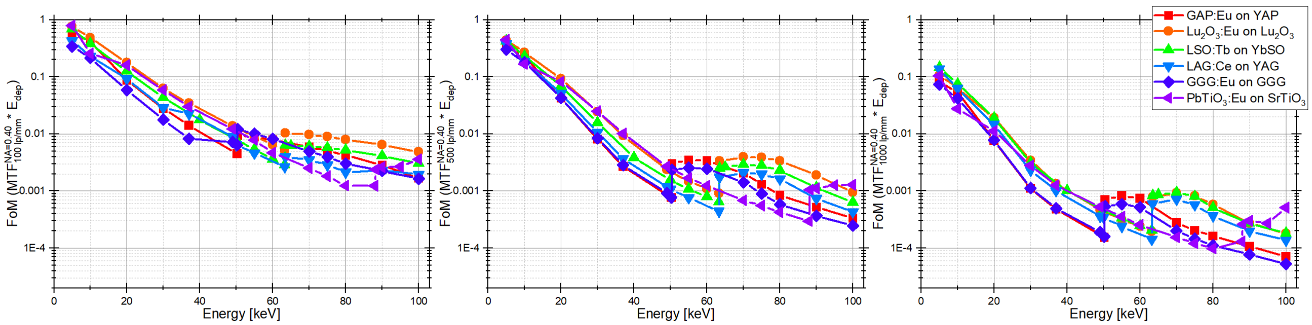

Figure S 4 The proposed Figure of Merit (FoM) calculated from the contrast in the MTF blurred by optics (NA=0.40) at 100, 500 and 1000 lp/mm and the energy deposited in the SCF. Values are extracted from simulations at X-ray energies from 5-100 keV for 5 μm SCFs supported by 150 μm substrates.

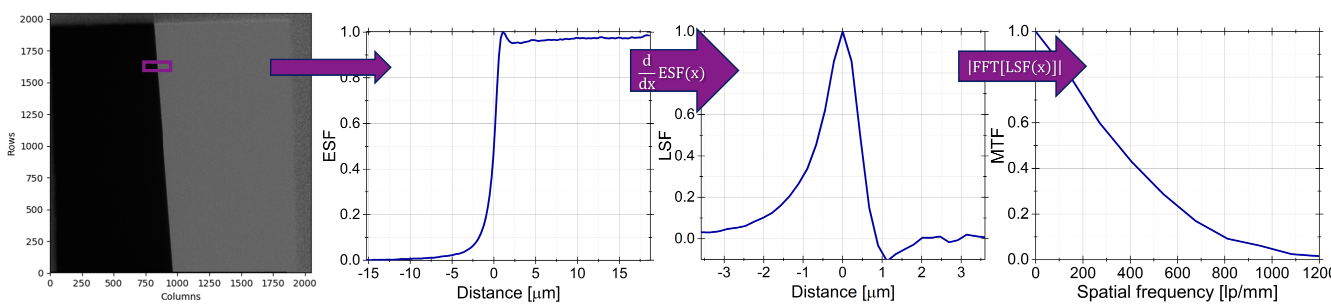

Figure S 5 The procedure to obtain the MTF. From left: An image of the GaAs edge cutting the image in half obtained with the setup described in the paper (camera pixel size: 7.4 μm, effective pixel size: 0.224 μm) where a region of interest is chosen to ease the computing time. The image is rotated to have the edge vertically, to be prepared for the computation of the ESF. The derivative then gives the LSF and the FFT finally gives the MTF.