Response to reviewer #3

Dear Anonymous Reviewer,

Thank you very much for your detailed and thorough comments on our manuscript. Please find our answers listed below:

Responses to major comments

> The paper is written clearly and well-presented but the authors are suggested to check a few grammar mistakes (such as the use of articles) and typos or misplaced text within a sentence including the type of English (American, British or others). I have pointed out some (find them below) but still, authors need to look at such errors throughout the text. My comments and general queries are as follows (P stands for page number):

We are sorry for the encountered typos. Thanks to the other two reviewers, we ironed out already a couple of those mistakes. Still, of course, this manuscript version was is not yet accessible. Besides, we tried to stick to British English.

> Figure 5: do authors see an initial rise in the signal in these samples? The signals used as the examples in Figure 3 have it. Does this mean it is not there in the samples investigated here (Figure 5)? Have they checked this between the grains? How many initial channels (time) were removed in De determination?

This is a good observation! Indeed, in some curves, a slight initial rise is visible, mainly for curves from sample BDX16651. However, we do not consider this a problem. The arbitrary curve in figure 3 shows a stronger initial rise since this was observed in many natural (and partly regenerated) IR-RF curves (cf. Frouin et al., 2017, their supplement). However, again, this is an arbitrary curve, and it is not related to data measured in our manuscript. A general pattern is that the very first channel shows a lower count rate. This is likely due to the mechanical opening of the shutter. For the D_e estimation through sliding, the very first channel was discarded for all analyses.

We did not inspect specifically the variation of the initial rise between single grains since this was not in our analysis's focus, and we did not feel that this is of particular relevance in our manuscript.

> Authors say they see at least four-dose components. Have they compared their dose response curves (DRCs)?

We have compared them during the routine data analysis, nothing particular was attracting our attention.

> Could they share the data?

GChron strongly encourages authors to share their data. We followed this call. The **entire** dataset, i.e. all sequence files, raw image data and partially processed data (around 1.9 GB of data), had been submitted in parallel to a public repository Zenodo (Kreutzer and Mittelstrass, 2020) and is available under the Create Commons Licence.

> The authors have not presented any data comparing DRCs within a sample. This is crucial since one would want to know the extent of variation in the DRCs (i.e. if any saturates later or not) in order to evaluate grain-dependent saturation (or dynamic range).

Yes, you are right. These data are not directly presented in the manuscript. However, as mentioned above, our data are ready for inspection. The files also include processed dose-response data. We even provide the R scripts we used for the data analysis, so any reader can play with the data.

However, we did not see any particular reasons to compare the curves in the manuscript. They add nothing to the story (but again, such processed data is available for inspection to interested readers).

> Do authors think that the grains which were not found bright, in reality, photons were produced but not detected due to the overall noise?

Of course, we cannot entirely exclude that some grains remained undetected because their signal was lower than the background noise. However, we have no evidence for such an effect. Grains showed sufficient IR light, or they did not show IR light at all and where too dim to be analysed.

> In that case, noise can still be suppressed and therefore EMCCD is required since below a threshold detection point EMCCD is still better than sCMOS cameras. Do authors think that it is not important to look for those grains and only the bright grains (and resulting DRCs) will do?

This is an interesting point. At the beginning of our project, we took the EM mode into consideration and experimented with it. However, we rejected the use the of the EM mode for the reasons we stated in section 2.4.3. The by far worst problem is the pixel well overflow caused by bremsstrahlung spots. The image below shows how the resulting effect looks like for one single spot:

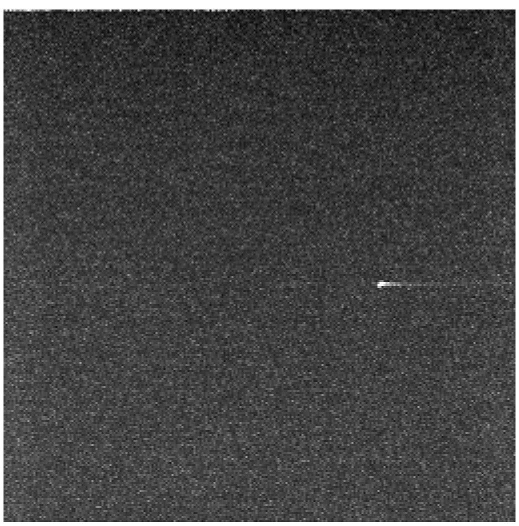

Figure 1: Arbitrary no-signal OSL image taken with 200x EM gain and 0.4 sec exposure time at a Princeton Instruments ProEM512B. The spot is either caused by a cosmic ray or by a bremsstrahlung photon from the ca. 50 cm away located beta source

You can see a signal "tail" or "streak" left-hand of the spot. In case of an IR-RF image with 5 sec exposure time, we see about 150 of bremsstrahlung spots with many of them having more or less intensive streaks. Most IR-RF signals but the brightest of grains become hard to observe in such a case. Also, we found the image processing far more challenging then because our median grouping algorithm ceased to work proberly.

We are aware that we can decrease the spot density by decreasing the exposure time. However, then we would also decrease the SNR because of the less accumulated signal before readout. Please be aware that the readout noise is still significant in EM mode (32 e- at 5 MHz according to the Certificate of Performance). In conclusion, we achieved far better images with the conventional readout mode (EM mode turned off) as we did not observed any pixel well overflows. Also the readout noise (3.2 e- at 100 kHz) is still low enough to ensure the detection of grains with a photon flux of just a few photons per second.

Regarding sCMOS cameras: A modern sCMOS camera can reach better SNR-per-pixel levels than an EMCCD camera at conventional readout. Scientific back-illuminated CMOS cameras have a readout noise of about 1 e- while achieving about the same quantuum efficiency than back-illuminated EMCCD cameras. In addition, sCMOS cameras offer higher image resolution and a larger field of view and thus enable a higher magnification. Also, sCMOS cameras are cheaper. We are quite sure that we would choose a sCMOS camera (but one with large pixels) if we would build a RF reader today. Nonetheless, regarding the maximum achievable sensitivity, an EMCCD camera is probably still better, provided the appearance of bremsstrahlung spots can be reduced to a sufficiently low level.

We clarified our statement regarding bremsstrahlung spot streaks in section 2.4.3.

Responses to specific comments

> P2, line 39-40: I really think this sentence needs a word 'from' before single grains.

Done.

> P2, line 43: It is 'charged coupled device (CCD)'.

Thank you, indeed.

> P3, line 67: Seems 'a' is in the wrong place. Further, I do not think it is important to expand SR again since it is already done earlier. One time explanation is enough.

We rephrased the beginning of this sentence.

> P7, line 148-150: Actually, most of the solar simulator settings differ a bit when compared to Frouin et al. (2015). Therefore, it is good to mention about all of them, not just UV.

Thank you, this was also flagged by the first reviewer, we clarified this now in the text.

> P8, line 178: 'a' setting?

Done.

> P8, line 183: '(2) it reduces the dynamic range and the linearity of the signal acquisition'. . . ! I think this is partially true. There is a limit to which EM gain can still be applied maintaining full well capacity. Under this limit, the EM gain can increase the dynamic range by reducing the read noise.

We do not agree. Like we stated above, EM read out increases read noise and adds excess noise. To compensate these additional sources of noise, a minimum EM gain is necessary, depending on the particular camera settings (to our knowledge and experience an EM gain of about 200x is necessary to gain a significant SNR win). An EM gain high enough to improve the low range would certainly reduce the high range threshold.

> P10, line 210: 'images are combined to one image by taking the median pixel value for each pixel location. . . .! Does this mean the total no. of images are reduced by the group size during median filtering and the measurement time of each image is summed?

That is correct, the total number of images is reduced and the measurement time is summed up. So basically, the filtering reduces the resolution on the x-axis.

> P11, line 222: Authors should provide more information on interpolation methods or provide references, therefore, must expand this paragraph.

The available interpolation methods are integral parts of the *ImageJ* source code and are briefly explained in the *ImageJ user guide* (Ferraira and Rasband 2012). They refer to Burger and Burge (2008) for further details on the `bicubic` interpolation. On the `bilinear` interpolation, no further references are given. We feel that details on the interpolation method are beyond scope of our contribution. Hence, we added just a reference to Ferraira and Rasband (2012) to the paragraph.

> P14, line 290, The sigma here represents error?

We clarified that we quote arithmetic mean ± standard deviation.

> P15, Figure 5: Good to use unit i.e. px for diameter.

Added.

> P16, line 329: define sigma_m.

Well spotted, this might indeed not be intelligible to readers unfamiliar with the cited work. We now clarify that σ_m refers to the intrinsic overdispersion.

> P17, Figure 6: Is it possible to re-number the grains 1-10? As the text says only 10 grains were selected.

Yes, this would be possible, but then it might become more confusing to readers who want to reproduce our results with our original dataset (which is wanted). The numbers are assigned by the algorithm, and relabelling would somewhat disconnect the automated ROI selection from the final results. Therefore we prefer to keep the numbering scheme as shown in the figure. Hopefully, this makes some sense.

> Further, It would be good to keep the horizontal label within the image's boundaries.

The scales are accurate although it looks otherwise. The images had been cropped in order to avoid some issues in the presentation. We clarified the caption text and noted the cropping

> P18, line 356-357: The unit will be in years, not Gy.

Done.

> P19, line 371: It would be good to cite any work relevant to the potential degradation of the filter under ionising radiation.

Done.

> P20, line 408: Not sure what effect?

We rephrased this part earlier, but we hope that it now reads intelligible.

> P21, lin3 433: Close the bracket around SR IR-RF.

Done.

> P23, line 481: the image 'is'.

Nothing changed here, perhaps page and line numbers were not correct?

> P25, line 514: dpixel = 16 μm – Is this value from the manufacturer?

Yes, the pixel size can be deduced from the data sheet of the camera.

> P25, line 520: Luminescence signal (phi_grain) per grain is basically the sum of counts from each pixel in an ROI (that has a fixed number of pixels), isn't it? The difference between eq A1 and A6 lies between how phi_grain is chosen: per pixel or in an ROI. am I correct? I think the sum needs to be mentioned.

Yes, this part is misleading. We expanded and clarified it.

Dirk Mittelstrass, Freiberg and Sebastian Kreutzer, Aberystwyth, March 23, 2021

References

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Burger, W. and Burge, M., 2008. Digital image processing: an algorithmic introduction using Java, 1st ed., Springer, New York. https://imagingbook.com/

Ferreira, T. and Rasband, W., 2012. ImageJ User Guide IJ 1.46r. https://imagej.nih.gov/ij/docs/guide/user-guide.pdf