Dear Editor,

Please find our responses to the referees’ comments (bold text) below.

Changes to the manuscript and SI are highlighted in blue text in hl\_main.pdf and hl\_sup.pdf.

On behalf of all the authors,

Anthony Yoshimura

Lawrence Livermore National Laboratory

Referee 1

**The authors could enlarge the sampling and mention applicability to also other classes of 2D materials of relevance whereby their methodology may be very useful, not least some confined materials containing hexagonal group III nitrides (beyond h-BN) and oxides, namely, e.g., Nanoscale12 (2020) Pages 19470 - 19476; Applied Surface Science 548 (2021) Article number 149275. Such works are supportive to the transferability of the scheme reported in the present manuscript.**

Thank you for this suggestion. We now mention the prospective application to layered and confined 2D systems in the conclusion, citing both articles you mention.

**The authors discuss thermal kinetic energy at room temperature which is part of the scheme assessment. However, isn’t that also important to explicitly compare these energies to the thermal stability of the studied 2D material(s)? Many materials would delaminate/disintegrate at similar energies…**

This is a great point to keep in mind in general. However, to the best of our knowledge, the temperatures considered in our manuscript are those employed in the experiments we reference. Specifically, Cretu et al. [10.1016/j.micron.2015.02.002] irradiated their hBN at temperatures up to 1473 K. Kretchmer et al. [10.1021/acs.nanolett.0c00670] did not mention any specific temperatures in their study, so we assumed that their MoS2 was irradiated at 300 K. Neither articles mention any delamination nor disintegration, so we do not believe such stability issues are a major concern in our study.

**Section S6: Fitting and converging S. How specific is the convergence routine considered each particular 2D material? The conclusion that Sec. S6 leaves the reader with is that it is very specific and needs lots of tests and simulations. Or am I missing something? In any case it would be worth to revised the final sentences of this section.**

Thank you for pointing this out. While it may not have been clear in Section S6, the method for converging S is material independent. The goal of the k-point fitting method is to determine the limit of S for an infinitely dense k-point mesh. This requires calculating S for sufficiently dense k-point meshes, i.e., S(Nk) plotted in figure S4a should appear to approach an asymptote for the densest k-point mesh considered. We found that meshes of 45x45x1 and 36x36x1 were sufficiently dense hBN and MoS2 respectively. These dense k-point meshes have very similar linear k-point spacings of 0.056 and 0.055 inverse angstrom for hBN and MoS2 respectively. The hBN mesh contains more k-points because hBN has a larger Brillouin zone.

Admittedly, this method relies on “eyeballing” the S(Nk) curve, relying on the user’s discretion as to whether the curve is approaching an asymptote. We are currently working to establishing a more formal convergence criteria for future work.

Convergence of S with respect to other parameters is also material independent, and a bit more straightforward. For a given parameter, we chose a value such that any increase in the parameter’s precision changes S by less than 5%.

We have added a paragraph at the beginning of section S6 that hopefully contextualizes the section to better orient the reader. We have also added two sentences to the end of the caption of figure S4 to clarify that the convergence methods for hBN and MoS2 are identical.

**Spell-check and stylistic revision of the paper are still necessary. Some long sentences, misspellings, etc., still are noticeable throughout the text.**

We have proofread the manuscript again and have done our best to fix any remaining errors. Please let us know if there are still any specific errors you would like us to change.

Referee: 2

**Have you calculated the displacement thresholds of B and N in a pristine hBN, instead of on the edge of hBN? How does it compare to the experiment?**

We have not calculated displacement thresholds for B and N sputtering from pristine hBN. However, Kotakoski et al. [10.1103/PhysRevB.82.113404] calculate thresholds of 19.36 eV and 23.06 eV for B and N respectively. As expected, these thresholds are several eV larger than those from the hBN edge. Assuming the excitation lifetime of 240 fs that we used to plot figure 5, these larger thresholds reduce the sputtering cross sections considerably for all beam energies. This is shown in the plot below, where the dashed “pristine” curves sit well below the solid “edge” curves.

**Diagram

Description automatically generated**

There also does not exist an excitation lifetime that aligns the simulated pristine curve to the experimental edge sputtering data of Cretu et al. [10.1016/j.micron.2015.02.002]. Below is our best fit to the data with a lifetime of 20 ps.

**Diagram

Description automatically generated**

Finally, experimental evaluation of the pristine displacement thresholds and corresponding cross sections would require a careful TEM study in which only single B and N vacancies (not multiatom pores) are counted for various TEM doses. To our knowledge, such a study does not exist.

**Is your code open-source and accessible by the public?**

We intend to provide a public GitHub repository soon after publication.