Dear Editor,

Please find our uploaded manuscript "QED theory of electron beam-induced excitation rates and sputtering cross-sections in 2D crystals" that we are hereby submitting for consideration as a Research Article in *Physical Review X* (*PRX*).

Electron beam irradiation by transmission electron microscopy (TEM) is an effective tool for defect engineering in 2-dimensional (2D) materials, potentially with atomic-scale precision. This degree of spatial control can provide powerful solutions to many of today's most pressing challenges in nanotechnology, including quantum device fabrication, hydrogen evolution catalysis, and electronic device miniaturization. Often, beam-induced defects are initiated and sculpted through a process called sputtering, in which a beam electron knocks an atom out of the material. However, current computational models drastically underestimate sputtering rates in insulators, in which beam-induced electronic excitations may weaken the bonding between the irradiated atoms. Most notably, substantial sputtering is experimentally observed at beam energies deemed far too low to drive atomic dislocations by present-day theories. To address this seemingly anomalous behavior, this work develops a quantum electrodynamics (QED)-based method to more accurately describe electron beam-induced sputtering cross-sections in 2D crystals by explicitly calculating the probabilities of beam-induced electronic excitations and their effects on sputtering kinetics. The consideration of excitations yields cross-sections that quantitatively match experiment and correctly predict appreciable sputtering rates at beam energies previously predicted to leave the crystal intact. This new predictive power can pave the way for the use of TEM for top-down atomic-scale defect engineering of 2D insulators.

We feel that this work meets the acceptance criteria of *PRX*, as it significantly advances the state of the art in modelling the response of materials to electron irradiation. The QED-based prediction of excitation rates could also push efforts to simulate both TEM images and electron energy loss spectra towards a more analytical, quantum field theory-based direction. Furthermore, we believe that this article will appeal to the diverse readership of *PRX*. We have attempted to present our findings in a logical and pedagogical manner, including sufficient background information and motivation to make our work accessible to a broad audience. At the same time, the article also provides in-depth derivations and technical details for experts who wish to reproduce our results.

Lastly, we were unable to download REVTeX to our lab's computing server for security reasons. The uploaded manuscript therefore uses LaTeX's default single-column format. Because of this, multi-panel figures are arranged in a "landscape" layout for the sake of readability. However, we have also uploaded figures 2, 3, and 4 with "portrait" layouts that should better suit PRX's two-column format. These PDF files are named with a "_portrait" suffix. Please let me know if you need any further information or materials. We can also resubmit the figure files in .ps format if needed.

This manuscript has not been published and is not under consideration for publication elsewhere. We have no conflicts of interest to disclose.

Thank you for your consideration.

On behalf of all the authors,

Anthony Yoshimura, PhD
Lawrence Livermore National Laboratory
Livermore, CA 94550, USA