I. SUPPLEMENTARY INFORMATION

A. Ion Bombarded Amorphization Threshold

We use Equation 1, from Ghandhi *et al.* [1] to approximate the amorphization threshold for our krypton-bombarded silicon crystal. We assume straggle (ΔR) of 54.3 nm (per SRIM), target atomic density (N) of $5{\times}10^{22}$ atoms cm^{-3}, and a target atom displacement energy (E_d) of 22 eV, to find a critical dose of $6{\times}10^{13}$ ions cm^{-2}. We note that this is in rough agreement with SRIM's predictions of a maximum of 2 displacements ions^{-1} Å^{-1} at the ion end of range, and a 30% displacements-per-atom threshold[2].

$$N_{crit} = \frac{5 * \Delta R * N * E_d}{E_o} \tag{1}$$

B. Predicting structural damage

The popular software The Stopping and Range of Ions in Matter (SRIM) and its associated package Transport of Ions in Matter (TRIM) allow for the prediction of stopping ranges, ion distribution, and damage profiles for arbitrary ions in arbitrary-composition amorphous materials. Simulations are performed using the Kinchen-Pease formalism using built-in silicon and krypton material properties. The damage profile is calculated as an average number of damage events (host atom displacements) per unit depth per ion. This allows for the calculation of damage as a percentage of host atoms affected (displacements per atom) per the following equation:

$$dpa = \frac{d * n}{\rho} \tag{2}$$

where d is the displacements from SRIM (number of displacements per unit depth per ion), n is the dose (ions per surface area), and ρ is the target's atomic density (atoms per volume).

C. Combining 3D contours to narrow uncertainty

Contour plots are typically generated in 2D, where the entire parameter space for 2 fitted parameters is mapped. This is done by generating the TDTR decay curve for each combination of parameters, and comparing the residual fit between data and curve. The quality of fits can then be plotted as a contour, with the two parameters as the x and y axes, and color or contour denoting the quality of fit. for 3 fitted parameters, we simply iterate through all combinations of the 3 parameters, and plot in 3D. The area (for 2D) or volume (for 3D) within a given contour of choice denotes all the combinations of values that yield an acceptable fit, and can then be taken as the uncertainty for the fitted parameters.

Two or more contours (typically 2D) can also be overlapped to reduce uncertainty, e.g. if measurements are taken at multiple frequencies, or if fitting is performed using both signal magnitude and the ratio of in and out of phase signal. Any location within one contour at one frequency that is not also within the contour at the second frequency still represents a poor fit, meaning the overlap (boolean intersection) of the two contour regions is considered the new uncertainty. Extending this to 3D, for each combination of the 3 parameters, for each combination of parameters, we can take the maximimum residual across frequencies. The remaining below-threshold region represents the boolean intersection of the multiple volumes, i.e., all combinations of parameters which find a good fit to data at all modulation frequencies. This is shown in Fig. 1, where we present the contour volumes at each frequency independently, and combined.

The extent to which this strategy is effective can be seen when exploring the results at lower doses. The 10^{12} ions cm^{-2} dose sample sees a much wider swath of functions yielding a good fit as compared to the and 10^{14} ions cm^{-2} dose sample, as seen in Fig. 2.

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[1] Sorab K. Ghandhi. VLSI Fabrication principles: Silicon and gallium arsenide, 1995.

[2] K. R.C. Mok, M. Jaraiz, I. Martin-Bragado, J. E. Rubio, P. Castrillo, R. Pinacho, J. Barbolla, and M. P. Srinivasan. Ion-beam amorphization of semiconductors: A physical model based on the amorphous pocket population. *Journal of Applied Physics*, 98(4), 2005.

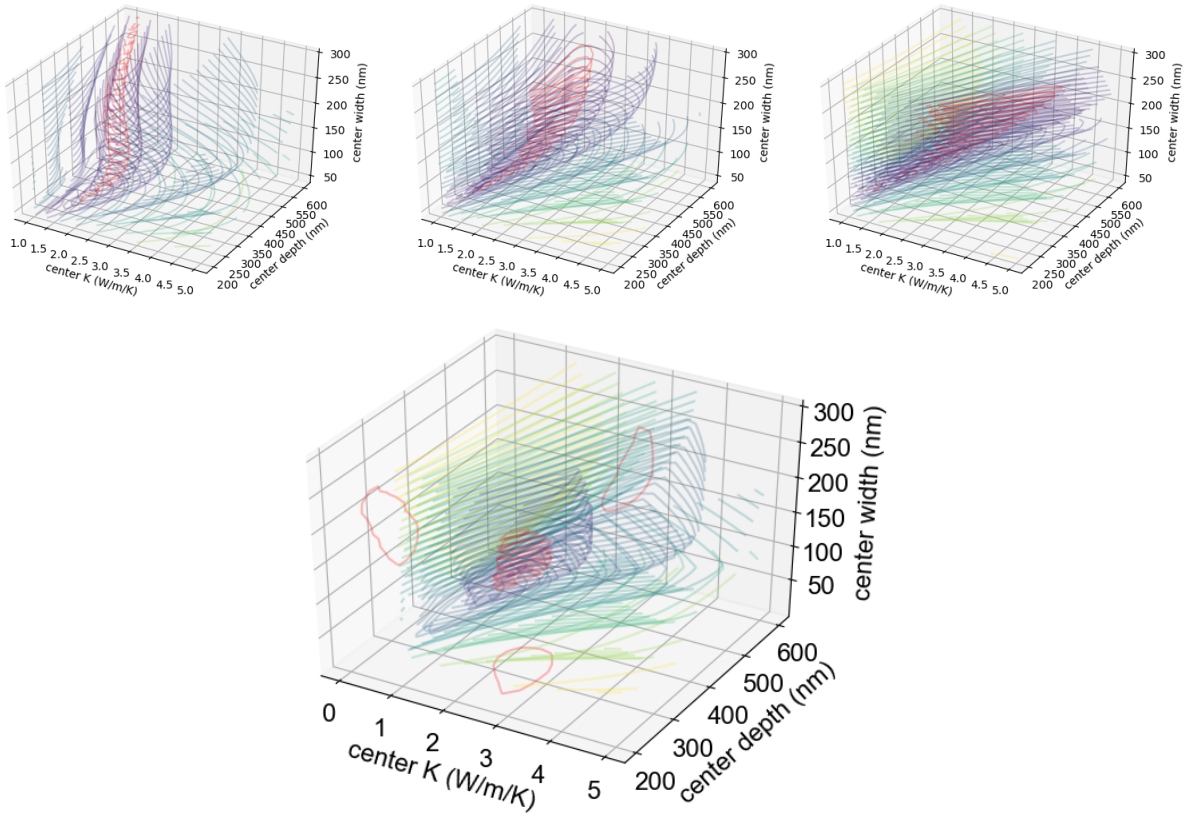

Figure 1. To investigate uncertainty, we create 3D contour plots (top) at each TDTR modulation frequency. Residuals (quality of the model's fit to data) are computed for many combinations of the three gaussian constants, and the volumes containing all residuals below 2.5% are shown in red. The 3 volumes are then the overlapped (bottom).

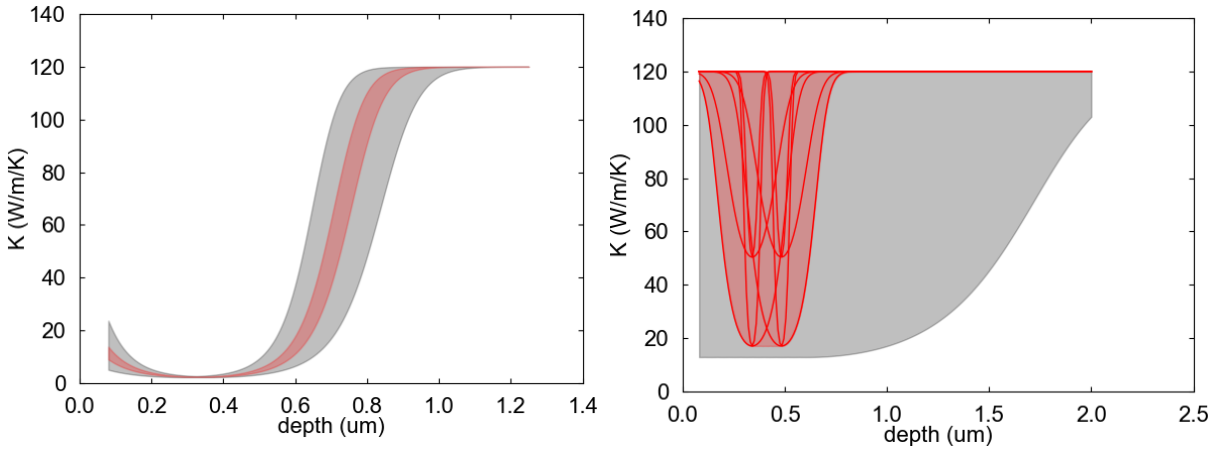

Figure 2. To visualize the growth of uncertainty as dose is reduced, we show the ranges of K(z) functions yielding a 1% residual (red) and 2.5% residual (grey). For the 10^{14} ions cm^{-2} case, there is a very narrow set of functions that yield great fits. For 10^{12} ions cm^{-2} however, we see a much broader range of functions. We also include the curves for the minimum and maximum combinations of parameters for clarity.