Monte Carlo Simulation of Nanoscale Material Focused Ion Beam Gas-Assisted Etching: Ga^+ and Ne^+ Etching of SiO_2 in the Presence of a XeF_2 Precursor Gas Supplemental

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1 Parameter Tuning

To study the effect of E_{act} and α, we simulate a scaled version of the experiment described by Harriott (1993) [1]. Our simulation consists of a 20 keV Ga^+ beam with a 31.25 nm radius cylindrical beam profile and 7.8pA current; the gas flux $\phi = 2 \times 10^{19}$ molecules/cm^2/s. Two setups are considered:

1. Test Setup 1: We populate the surface using Equation (2), from an initial condition of zero gas fraction, for a variable refresh time, and then simulate a single 1 μs beam dwell. This is then repeated 50 times.

2. Test Setup 2: We populate the surface with the equilibrium value gas coverage given by Equation (4). We then run a single beam dwell of varying dwell time. This is then repeated for $5,000$ ions.

For both test setups, we turn voxel updating off (i.e., the substrate remains smooth and has no vacancies throughout the simulation); this is done so that Equation (2) can be used to populate

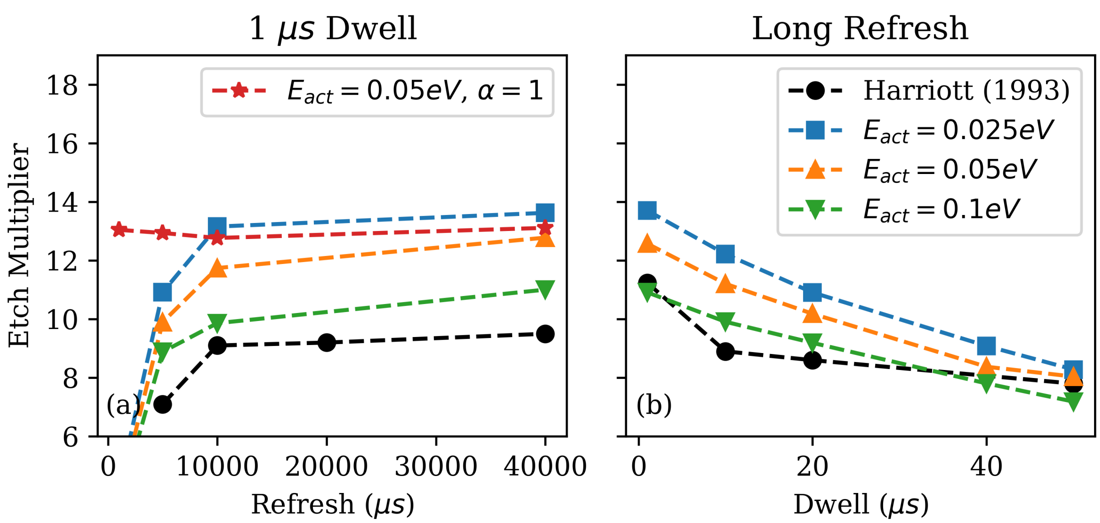

Figure 1: Etch multiplier of simulations, for (a) a fixed dwell time, and varying refresh time, and (b) for a fixed refresh time, with a varying dwell time. All simulations have an flux of $\phi = 2.5 \times 10^{17}$ molecules/cm^2/s, except for the points labeled with $\alpha = 1$, where the flux is $\phi = 10^{19}$ /cm^2/s. All simulations were run with a 20 keV 7.8 pA Ga^+ beam, with a uniform cylindrical profile of radius 31 nm. Simulations were run with voxel updating turned off, i.e., even if an atom is sputtered or etched, the corresponding voxel it originated from remains full. The legends apply to both plots in the figure; dashed lines are shown to guide the eye, however the symbols show the data points.

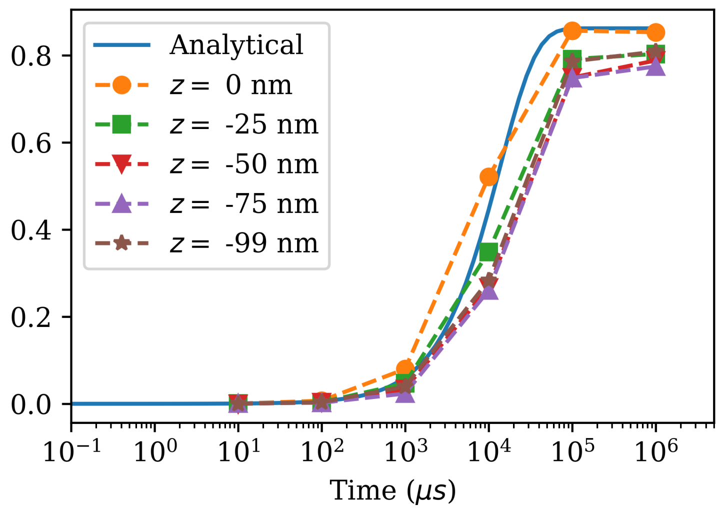

Figure 2: Population of a 100 nm deep, 15 nm FWHM Gaussian via at various depths. The analytical result for a flat surface is shown by the solid line. The gas flux is 4×10^{18} molecules/cm^2/s, with a sticking coefficient of 0.025. The dashed lines are shown to guide the eye; measurements were only made at the times indicated by the symbols.

the surface with XeF_2. For each simulation, we calculate the "etch multiplier" as

$$\text{Etch Multiplier} = \frac{\text{Sputter Yield} + \text{Etch Yield}}{\text{Flat Surface Sputter Yield}}$$

We plot the results of Test Setup 1 in Figure 1(a). A value of $\alpha = 1$ results in no significant sensitivity of the etch multiplier to refresh time, and is unable to reproduce the behavior seen in Harriott (1993) [1]. If we set the sticking coefficient of $\alpha = 0.025$, we obtain agreement with the experimental data in Figure 1(a). While this is equivalent to reducing the gas flux in this case, in all simulations in the main text, we use $\alpha = 1$; we discuss the effect of reducing α, and compare with scaling the flux in Supplemental Section 2. Using this value of α, we plot the etch multiplier for various values of E_{act}; $E_{act} = 0.1$ eV results in good agreement with the experimental etch multipliers in Figure 1(a). Likewise, we plot the results of Test Setup 2 for various values of E_{act} in Figure 1(b); simulations show a more linear decay in the etch multiplier with dwell time, perhaps owing to differences in the surface topography, since in these simulations even as etching occurs the surface remains atomically smooth. For the main text, we use a value of $E_{act} = 0.05$ eV; although Figure 1 suggests that a value of $E_{act} = 0.1$ eV agrees better with Harriott(1993) [1], we find good agreement using a lower value in the experiments reported in the main text.

2 Sticking Coefficient

In the main text, we have set $\alpha = 1$, so that incoming gas molecules always adsorb to the first empty surface site they find. For a flat surface, $\alpha \neq 1$ is equivalent to scaling the incoming gas flux. However, for deep vias, gas ricochet leads to a significant difference between a scaled gas flux, and

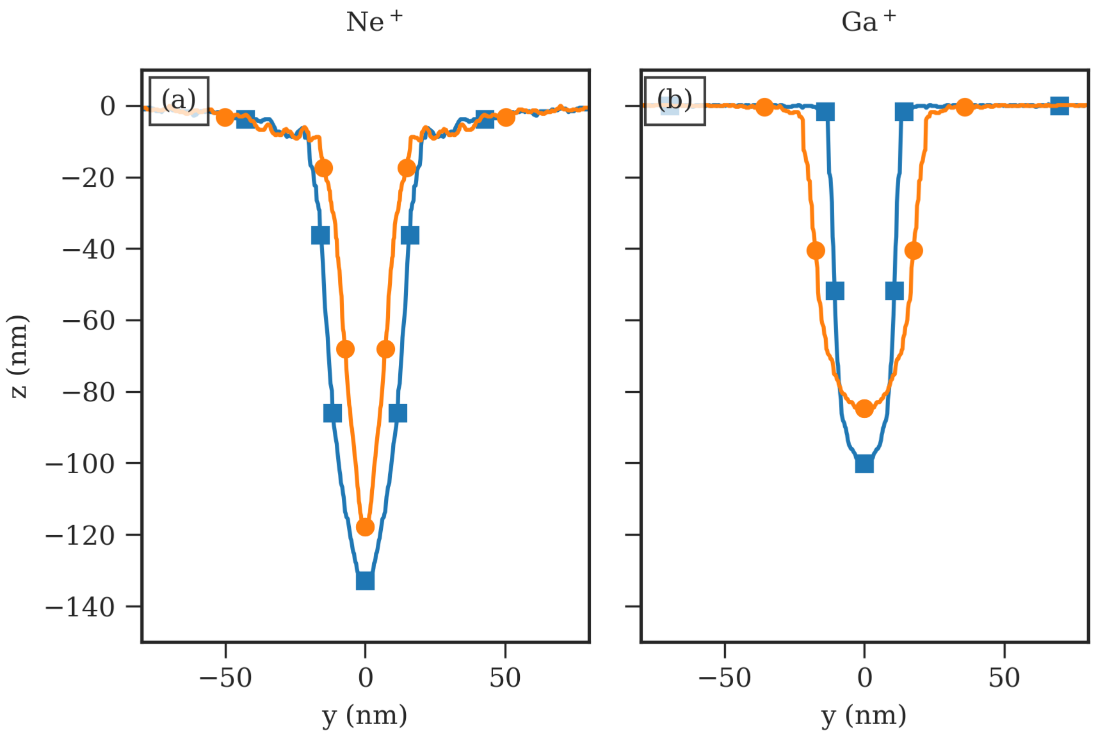

Figure 3: Comparison of etched vias resulting from simulations of the Ne^+ line scan, and the Ga^+ Recipe 5 described in the main text, using the "Measured" and "Effective" beam profiles.

a smaller value of α. In Figure 2, we reproduce the plot of the isotropic population of a Gaussian Via with XeF_2, plotted in the main text in Figure (2); however, in Figure 2, we have reduced the sticking coefficient from 1.0 to 0.025, and increased the gas flux to 4×10^{18} molecules/cm^2/s, from 10^{17} molecules/cm^2/s. On a flat surface, the gas population as a function of time is identical for both figures, as shown by the curve labeled "Analytical" and the "z=0 nm" simulated points. However, deep inside the Gaussian via, a higher flux with a lower sticking coefficient leads to the gas population occurring with a shorter time scale; compare Figure 2 with the main text Figure (2) for all data points evaluated at depths less than or equal to 25 nm. This can be understood by considering the following cases. If a gas molecule is incident upon a flat surface, and fails to adsorb due to the small α, it has no other opportunities to adsorb to the surface. However, if it fails to adsorb after reaching a surface site inside of a via, after desorbing it may find another site inside of the via, and have another opportunity to adsorb to the surface. Therefore, for a flat surface, $\alpha < 1$ is equivalent to scaling the incident flux by α, while inside of a via, the low sticking coefficient actually enhances population deeper in the via.

3 Comparison of Measured vs Effective Beam Profiles

In the main text, we simulate Ne^+ and Ga^+ etching using "Effective" beam profiles which are selected to reproduce the experimental via shape. These beam profiles can be contrasted with "Measured" beam profiles, which are estimated using the method described by Tan et al. [2].

The difference between these two profiles was discussed in detail in our previous work on FIB SiO_2 sputtering [3]. Based on experiments, the "Measured" beam profiles are estimated to be the following [unpublished]:

- Ga^+ beam:
  - "Measured Beam": $(a_1, a_2, a_3) = (4.72, 0, 27.82)$ nm, $(I_1, I_2, I_3) = (0.97, 0, 0.03)$
- Ne^+ beam:
  - "Measured Beam": $(a_1, a_2, a_3) = (2, 27.9, 68.5)$ nm, $(I_1, I_2, I_3) = (0.89, 0.06, 0.05)$

The "Measured" beam is interpreted as more closely representing the inherent beam current distribution, while the "Effective" beam accounts for possible machining artifacts which may decrease etching resolution. We compare the etched vias resulting from using the "Measured" and "Effective" beams in Figure 3. Simulations using the "Measured" beam profile are notably narrower, however the difference is not as large as that observed for pure sputtering [3], suggesting that the lower doses associated with etching may result in less platform level artifacts.

4 Comparison of Ga^+ etching simulations with deeper experiments

In the main text, Figure (6) compares simulated vias sputtered with Ga^+ Recipes 1-5, with profiles extracted from experimental TEM images. Profile extraction was performed by means of a feature detection of the images, using a multilayer perceptron implemented in Scikit-Learn [4], and further image processing with SciPy [5]. The exact shape of the profile is ambiguous, as the boundary in the TEM images is at times poorly defined. Due to this ambiguity, we plot the simulated profiles with the actual TEM cross sections in Figure 4-5.

5 Distribution of Etch Events

The radial distance of etch events was simulated for incident 10 keV Ne^+ ions in SiO_2. For the purposes of the simulation, XeF_2 was set to a full coverage during the simulation (i.e., it was not exhausted in the reactions causing etching). Simulations show that the radial distribution of etch events follows a Cauchy distribution with the form

$$\psi_{Cauchy}(x, y) = \frac{\gamma_C}{2\pi} \left(\frac{\gamma_C}{(x^2 + \gamma_C^2)^{3/2}} \right)$$

with $\gamma_C = 1.8$ nm (see Figure 6).

References

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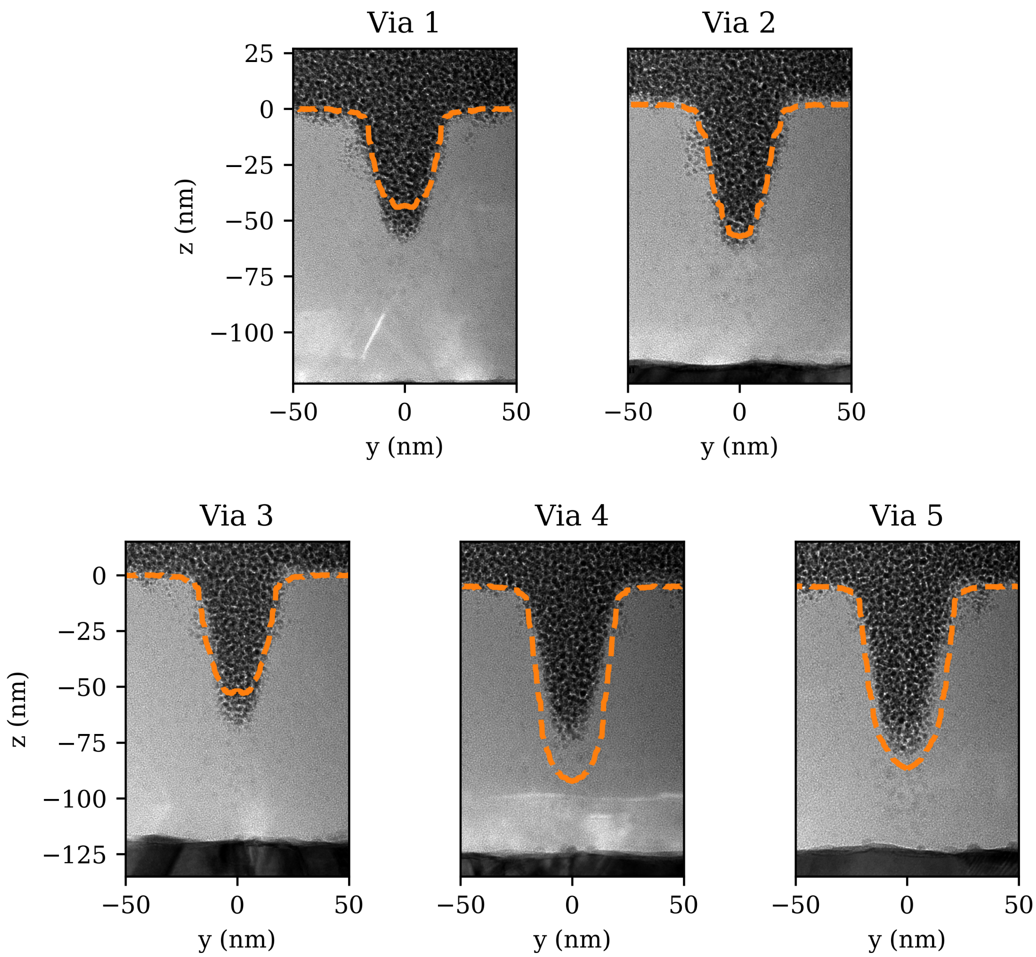

Figure 4: Comparison of simulated etched profile, with the shallower experimental results, for the Ga^+ etched via with Recipes 1 through 5.

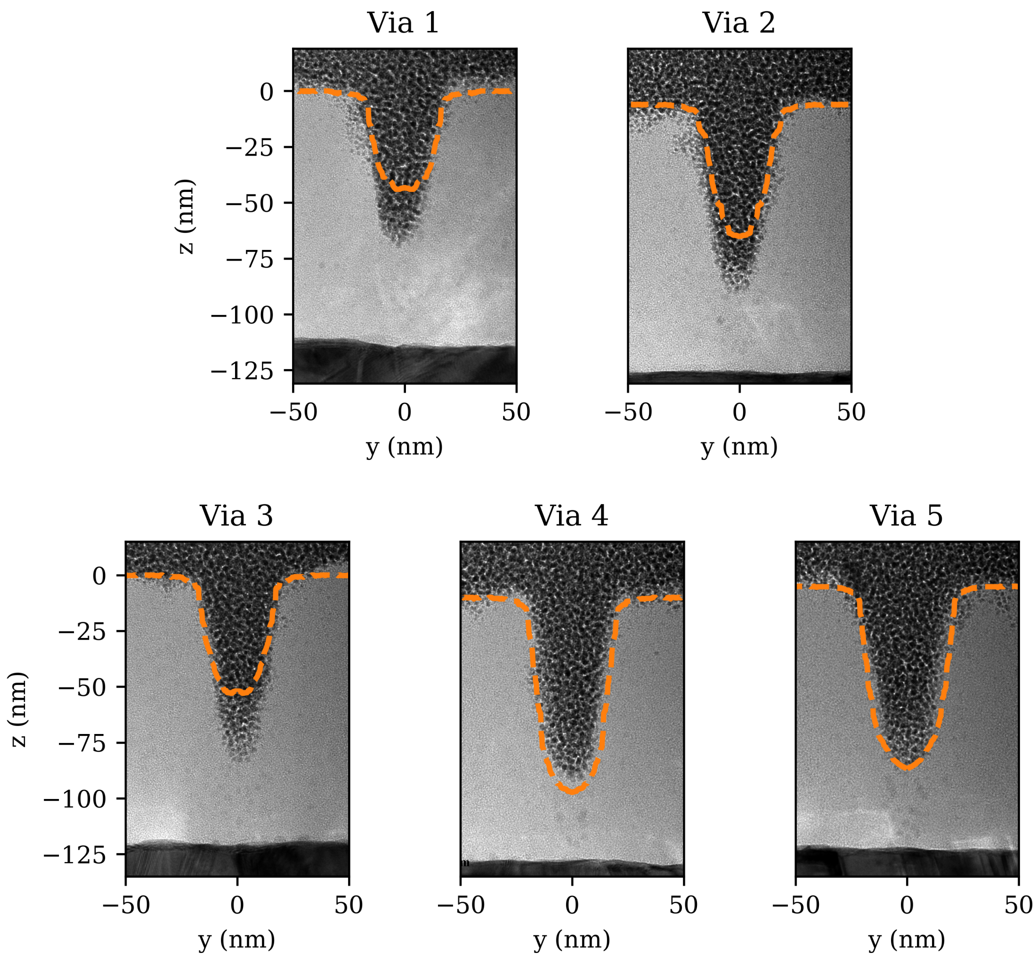

Figure 5: Comparison of simulated etched profile, with the deeper experimental results, for the Ga^{+} etched via with Recipes 1 through 5.

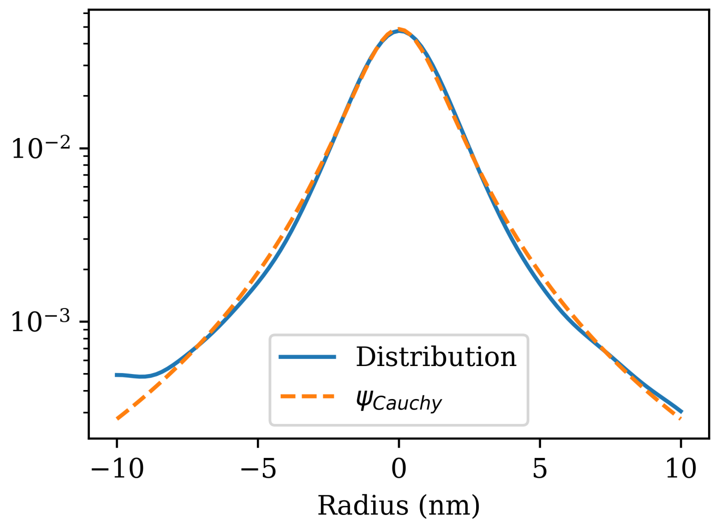

Figure 6: Distribution of the radial coordinate of etch events due to a 10 keV Ne^+ ion incident on the surface at $(x, y) = (0, 0)$ (solid curve), generated from 10,000 incident ions. The distribution was estimated using a Gaussian Kernel Density Estimation implemented in the SciPy package [5]. The dashed curve shows a Cauchy distribution, with $\gamma_C = 1.8$ nm.

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