Calculating dpa levels in single-crystal niobium samples exposed to Si^{3+} ions for

"Inferring radiation-induced microstructural evolution in single-crystal niobium through changes in thermal transport"

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Outline

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1. Sample Information

We have three samples in this study. All are 5mm diameter discs of polished single crystal niobium. One sample is kept as-is (control). The other two samples are cold-worked in a pellet press (1500 lb and 2000 lb force respectively). The samples were all exposed to progressively higher doses of radiation with CLASS using Si^{3+} ions. Samples were characterized with TGS prior to irradiation and after each of five irradiation steps. (See main paper for more information.)

2. Why this document?

In the main paper, we use dpa as a proxy for total dose, as it is the common standard in radiation materials science for describing total dose in terms of radiation damage incurred. However, there are a lot of assumptions that go into calculating dpa in an experiment. It is important that readers are able to understand what assumptions we made in this document, in case they wish to repeat a similar study or confirm our calculations for themselves. This information is also important because a reader may wish to compare our results here to other reporting on radiation damage, and then it is necessary to understand whether or not "1 dpa" in this paper is equivalent to "1 dpa" in another.

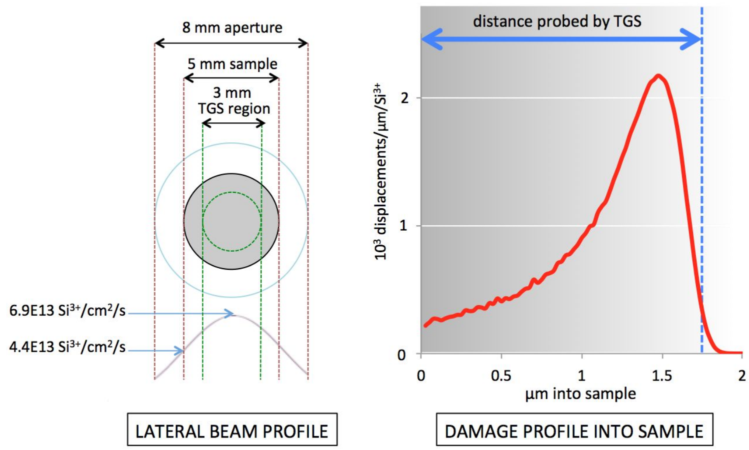
Furthermore, our dpa calculations are derived from a combination of experimentally measured parameters and SRIM results. SRIM itself relies on a set of input parameters, and which choices a researcher makes at this stage will affect their results. There are also multiple paths by which a researcher can extract dpa information from SRIM results. Therefore, it is important that readers can understand which choices we made in SRIM, in addition to the experimental parameters we measured, in order to arrive at the dpa values we report.

At a glance, this document, and the main paper, utilize the SRIM data in the VACANCY.txt output file. Displacements per angstrom per ion data caused by the primary knock-on atoms (PKAs) and the recoil atoms are added together, and these values averaged over the depth region of interest, to determine a basic dpa-per-second rate. However, there is also another method, proposed by Stoller et al., in which the SRIM data from the ENERGY.txt output file is used to calculate dpa. This second method is used to recalculate dpa at the end of this paper to show how things change if one chooses to utilize this approach. The resulting difference in reported dpa levels can be as much as 40%. This large difference further underscores the importance of maintaining clear records as to assumptions and methods used to calculate dpa.

3. Relevant beam parameters

There are two things to consider when calculating dpa from the known irradiation parameters: (1) the lateral beam profile and (2) the damage profile into the sample. These are illustrated schematically in the below figure, which is also in the paper. They also represent the updated assumptions detailed here in Sections 4 (accounting for lateral beam variation) and 5 (recalculating the damage profile with more accurate SRIM input parameters). The table provides important experimental beam parameters, which were used in the initial dpa calculations described in Section 3.

Figure 1.



Beam parameters (CLASS)[1]

Ion species	Si^{3+}
Ion energy	5.3 MeV
Nominal beam current	100 nA
Nominal beam flux density through the 8mm aperture	4.14466E+15 ions/m^2/s

On the left we have the lateral beam profile. This shows the Gaussian flux profile of the Si^{3+} beam: the beam flux is not uniform in lateral space. The beam passes through an 8mm diameter circular aperture, which is larger than the 5mm diameter sample. Note that TGS measurements for each sample were concentrated near the center of the sample: we did not take measurements near the sample edge. This is because of the Gaussian shape of the beam profile: the further from the sample center, the lower the total dose received. The nominal beam flux density is an averaged value based on the nominal beam current and the area of the aperture.

4. Step 1: calculating the beam fluence needed to reach target dpa values

We planned the irradiation experiments by assigning "target" dpa values that we wanted to progressively achieve in each sample. The original target values were **0.01, 0.03, 0.1, 1, and 3 dpa**: these values were used to determine how much integrated charge we needed to measure during each sample irradiation to reach our target dpa. This process is detailed below. Importantly, we did **not** consider the effect of a non-uniform beam profile: the calculations described here in Step 1 assume a uniform beam flux passing through the aperture and reaching the niobium samples. This is why we corrected the reported dpa values, via the methods explained below in Step 2.

[1] http://cstar.mit.edu/class.php

First, we used SRIM to calculate that the Si^{3+} beam is expected to cause **0.218227 displacements/Å/ion** of damage through the first micron in the Nb samples.[2] This result was obtained using data in the VACANCY.txt SRIM output, which is graphed in the figure above ("damage profile into sample"). VACANCY.txt reports vacancies/Å/ion at regular depth intervals in two columns: one is for vacancies/Å/ion caused by the incident radiation, and the other gives vacancies/Å/ion created by recoil atoms. These values are added together at each depth, and these sums are then averaged for sample depths from 0 - 1.75 µm (the depth probed by TGS in this project). Here, "vacancies" are taken to be equivalent to "displacements."

A value of **1.63E-04 dpa/s** in the niobium samples due to the Si^{3+} beam was calculated using the SRIM calculation and the beam current density :

0.218227 dpa/Å/ion × 1E+10 Å/m × 4.14466E+15 ions/m²/s = 1.63E-04 dpa/s

Given a beam current of 100 nA, one can now calculate how much ion charge (as in total ion charge that has impinged on the sample) is required to achieve 1 dpa in damage:

1.63E-04 dpa/s = 6141.67 s/dpa
100 nA × 1A/1E+09 nA = 1E-07 A
10^{-7} A × 1 (C/s)/A × 1E+03 mC/C = 1E-04 mC/s
6141.67 s/dpa × 1E-04 mC/s = 0.614167 mC/dpa

Of course, this was based on assumptions that we later updated. **0.614167 mC/dpa** is the charge required to achieve 1 dpa in the niobium samples using the *initial assumption* of a uniform flux through the beam aperture. Importantly, though, this is the value that was used as a point of reference when irradiating the samples to get to the planned dpa levels.

The charge counter on the experimental setup for this project measures charge in units of 10^{-11} C. As an example, to get from 0 dpa to 0.01 dpa, it was necessary to irradiate until "614617" was read out on the counter:

1E+02 dpa × 0.614167 mC/dpa = 6.14167E-03 mC = 6.1416E-06 C
6.14167E-06 C × 1 count/1E-11 C = 6.14167E+05 counts

The target charge readout for each dpa level is shown in the table below.

Total dpa to reach	Need to "add" this many dpa to the sample	Additional charge this requires (in units of 10^{-11} C), as read on the counter
0.01	0.01	615617
0.03	0.2	1228336
0.1	0.07	4299176
1	0.9	55275120
3	2	122833600

The following table shows the actual charge measured during each irradiation step for each sample.

[2] Using the Stoller method (see Section 6) results in a different value of displacements/Å/ion for the beam into niobium (and thus different values for the "true" dpa incurred in each sample). The assumptions/parameters set at the beginning of the SRIM calculation should be always be noted, as these will influence the reported results of the calculation.

Measured charge during sample irradiation

Sample/dpa	Get to 0.01 dpa (need 615617 readout)	Get to 0.03 dpa (need 1228336 readout)	Get to 0.1 dpa (need 4299176 readout)	Get to 1 dpa (need 55275120 readout)	Get to 3 dpa (need 122833600 readout)
Control	615846	1227203	4299176	55316972	122883237
1500 lb	614529	1228137	4299066	57020123	121658964
2000 lb	615339	1228279	4299915	55295118	122855051

The irradiation fluence for each sample and dpa level is then calculated from these values as follows. It is necessary to account for the +3 charge on the silicon ions. [e] is the elementary charge. The aperture radius was 4 mm.

$$\textbf{Readout charge} \times 10\text{E-}11 \rightarrow \textbf{Charge in [C]}$$
$$\textbf{Charge [C]} \times (1\ Si^{3+}\ \text{ion}/3\ [e]) \times (1[e]/1.6\text{E-}19\ [C]) \times (1/\pi * 0.004^2\ [1/m^2])$$

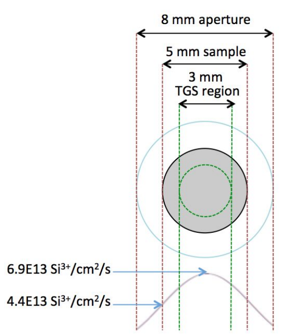
The fluence results are given in the table below.

Fluence in Si^{3+}/m^2:

Sample/dpa	To 0.01 dpa	To 0.03 dpa	To 0.1 dpa	To 1 dpa	To 3 dpa
Control	2.55247E+17	5.08634E+17	1.78186E+18	2.2927E+19	5.09309E+19
1500 lb	2.54701E+17	5.09021E+17	1.78182E+18	2.36329E+19	5.04235E+19
2000 lb	2.55037E+17	5.0908E+17	1.78217E+18	2.29179E+19	5.09192E+19

These are the values that hold if (1) one assumes a uniform beam flux through the beam aperture (2) one uses standard SRIM inputs for binding energy and displacement energy and (3) one relies on the charge readouts supplied above. These fluence values do **not** take into account the Gaussian beam profile or the fact that TGS measurements were concentrated in the middle 3mm diameter of the samples.[3]

5. **<u>Step 2: updating the initially reported dpa values by refining our assumptions</u>**



LATERAL BEAM PROFILE

First, we considered the beam shape. The circular Gaussian beam shape is taken to have a sigma value of 2.67 mm. The beam flux averaged throughout the entire 8mm aperture is 4.14E+11 ions/cm^2/s - we know this from the calculations above. However, if one accounts for the Gaussian beam shape and averages the flux 1.5mm to either side of the beam peak, the flux is much higher: 6.38E+11 ions/cm^2/s. Next, we make a new assumption, which is reasonable based on our experimental observation, that the beam peak was centered at the sample center. Since the TGS measurements were located near the sample center (see image at left), it is reasonable to assume that the true dpa levels we were measuring were higher than those that we would calculate from the aperture-averaged flux.

We divide 6.38E+11/4.14E+11 to get a ratio of 1.54. This can be applied as a scaling factor to the fluences reported above. Updated fluences are listed in the table below.

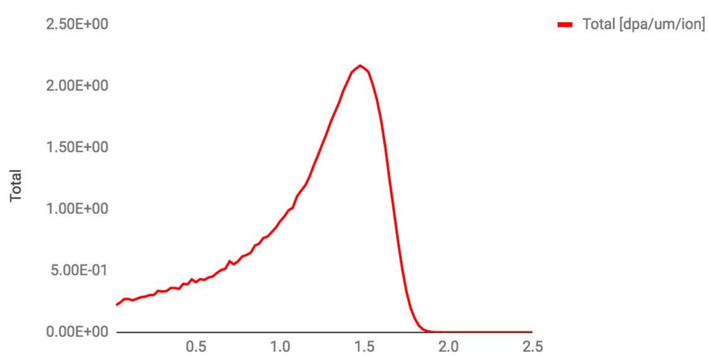
[3] These are the fluence values reported in my thesis. They are updated in the paper - hence why there is not 1:1 correlation between the dpa values reported in the paper and the dpa values reported in my thesis - after we gave the input assumptions some more serious thought. - S.E.F.

Labels are placed in quotes to indicate that these were the nominal, but not the true values, of the dpa actually reached in each sample. (Keeping the old labels here makes for easier cross-referencing with older, un-updated documents.)

Updated fluence in $Si^{3+}/\mu m^2$ (accounting for higher beam flux near sample center):

Sample/dpa	To "0.01 dpa"	To "0.03 dpa"	To "0.1 dpa"	To "1 dpa"	To "3 dpa"
Control	3.93E+05	7.84E+05	2.75E+06	3.53E+07	7.85E+07
1500 lb	3.93E+05	7.84E+05	2.75E+06	3.64E+07	7.77E+07
2000 lb	3.93E+05	7.85E+05	2.75E+06	3.53E+07	7.85E+07

Next, consider the damage profile into the sample. The original SRIM calculations above used an incorrect value of E_d. Values of 60 eV for displacement energy[4] and 0 eV binding energy[5] instead of the default SRIM inputs were used to calculate a new damage profile. (Note that this will, to some extent, counteract the effect of the updated, increased fluences. In the original dpa calculations, we underestimated the fluence and overestimated the extent of the SRIM-calculated into-sample damage) The total dpa per unit depth per silicon ion were calculated using the same procedure outlined in Section 3 (i.e. calculating damage using the VACANCY.txt output). The results are plotted below, with the x-axis showing depth into sample in [μm]. As before, we are interested in damage 1.75 μm into the sample.



Based on the updated SRIM calculations, the average value of vacancies/μm/ion from 0 to 1.75 μm is **906 vacancies created per μm per ion**.

Multiplying fluence (ions/μm^2) by the averaged SRIM data (vacancies/μm/ion) yields a value in units of vacancies/μm^3. This value can then be divided by the number density of Nb (5.56E+28 1/m^3 = 5.56E+10 1/μm^3) to yield total vacancies (displacements) per Nb atom.

Vacancies created/μm^3

Sample/dpa	To "0.01 dpa"	To "0.03 dpa"	To "0.1 dpa"	To "1 dpa"	To "3 dpa"
Control	3.56E+08	7.10E+08	2.49E+09	3.20E+10	7.11E+10
1500 lb	3.56E+08	7.10E+08	2.49E+09	3.30E+10	7.04E+10
2000 lb	3.56E+08	7.11E+08	2.49E+09	3.20E+10	7.11E+10

[4] ASTM, NRT. "E521 (1996) Standard Practice for Neutron Radiation Damage Simulation by Charged-Particle Irradiation." *Annual Book of ASTM Standards* 12.
[5] Stoller, Roger E., et al. "On the use of SRIM for computing radiation damage exposure." *Nuclear instrumments and methods in physics research section B: beam interactions with materials and atoms* 310 (2013): 75-80.

Vacancies created per niobium atom (dpa)

Sample/dpa	To "0.01 dpa" Irradiation 1	To "0.03 dpa" Irradiation 2	To "0.1 dpa" Irradiation 3	To "1 dpa" Irradiation 4	To "3 dpa" Irradiation 5
Control	6.40E-03	1.28E-02	4.48E-02	5.75E-01	1.28E+00
1500 lb	6.40E-03	1.28E-02	4.48E-02	5.93E-01	1.27E+00
2000 lb	6.40E-03	1.28E-02	4.48E-02	5.75E-01	1.28E+00

For a given irradiation step, each of the three samples received roughly the same fluence from the beam. Next, the resultant dpa calculated for each of the three samples is simply averaged at each irradiation step. This is just for the straightforwardness of referring to a single set of dpa values for all three samples. The averaged values are given in the table below.

Irradiation step	Additional dpa incurred in this step
1	0.00640
2	0.01278
3	0.04481
4	0.58119
5	1.27481

These values represent the additional dpa incurred by the samples at each irradiation step. In the paper, we want to report on the total dpa incurred by the sample at each step (previous dpa + additional incurred). These dpa totals are as follows: **0, 0.0064, 0.0192, 0.0576, 0.06260, and 1.8560 dpa.** Since there are a lot of assumptions in these dpa calculations - it's really more of a *best estimate* than a direct calculation, we will round:

0, 0.006, 0.02, 0.06, 0.6, and 1.9 dpa

6. <u>Appendix: recalculating the reported dpa values using the Stoller et al. method</u>