Examination of issues involved when using ion irradiation to simulate void swelling and microstructural stability of ferritic-martensitic alloys in spallation environments

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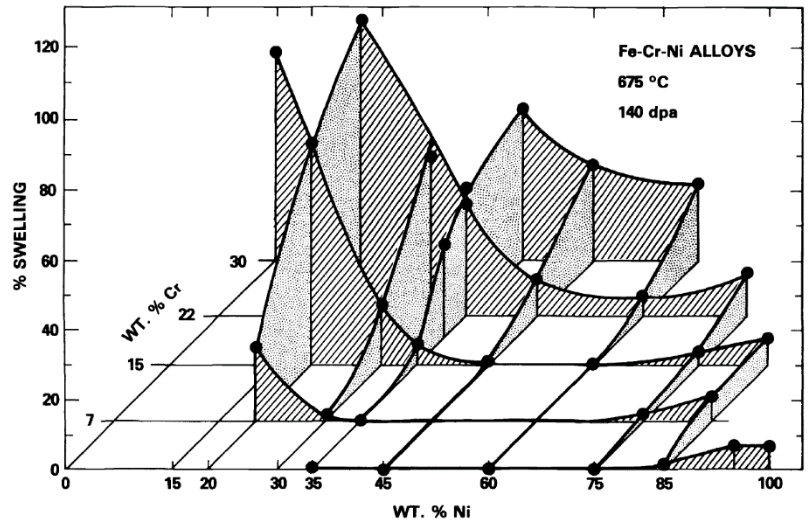
Background

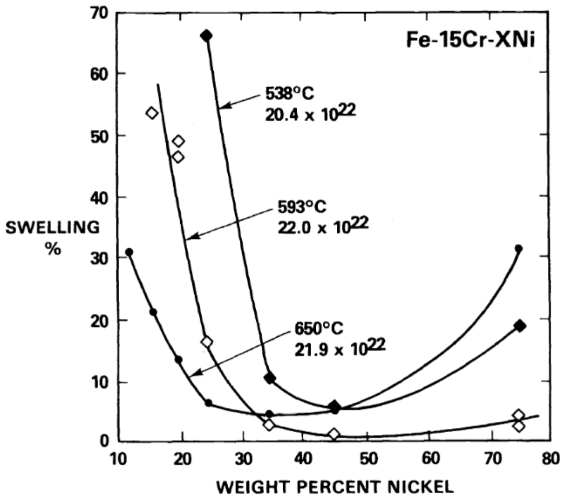
Ion bombardment was used very successfully in the fast reactor materials program to provide <u>guidance</u> on the compositional dependence of void swelling of austenitic alloys before neutron data became available.

5 MeV Ni^+ ions



Johnston et al., 1983

EBR-II neutrons



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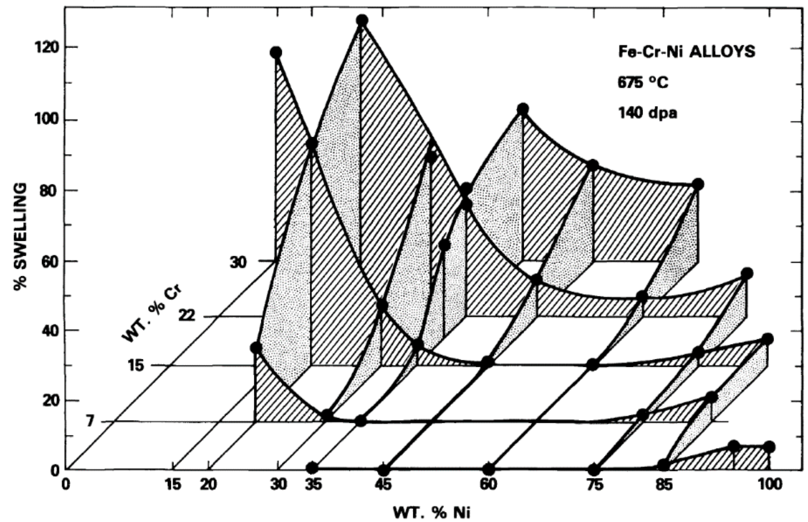

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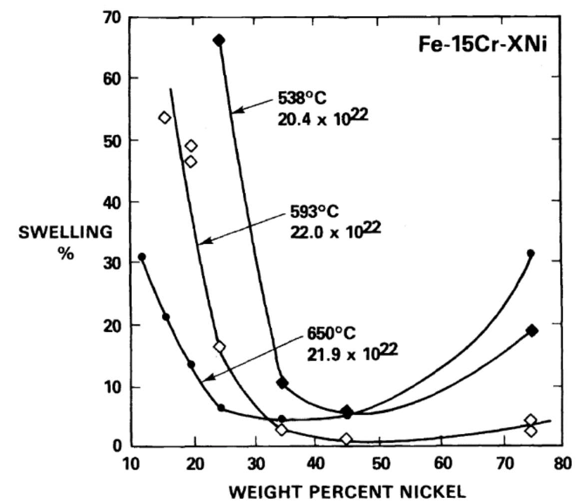


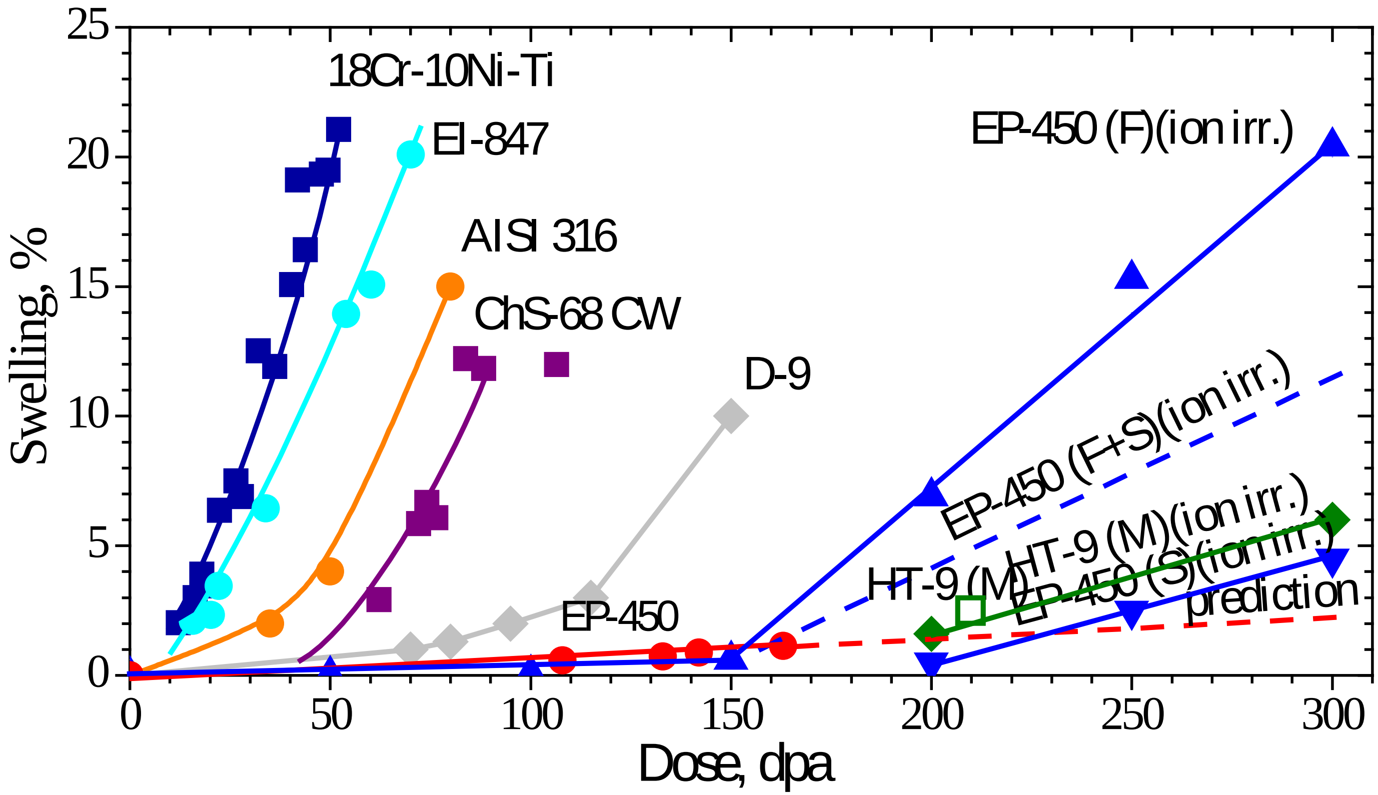
Garner and Brager, 1988

Ion irradiation also showed correctly the role of Cr, Si, P, cold-working, etc.

When conducted correctly, ion results also forecast the steady-state rate of 1%/dpa, independent of composition, cold-working, dpa rate, etc.

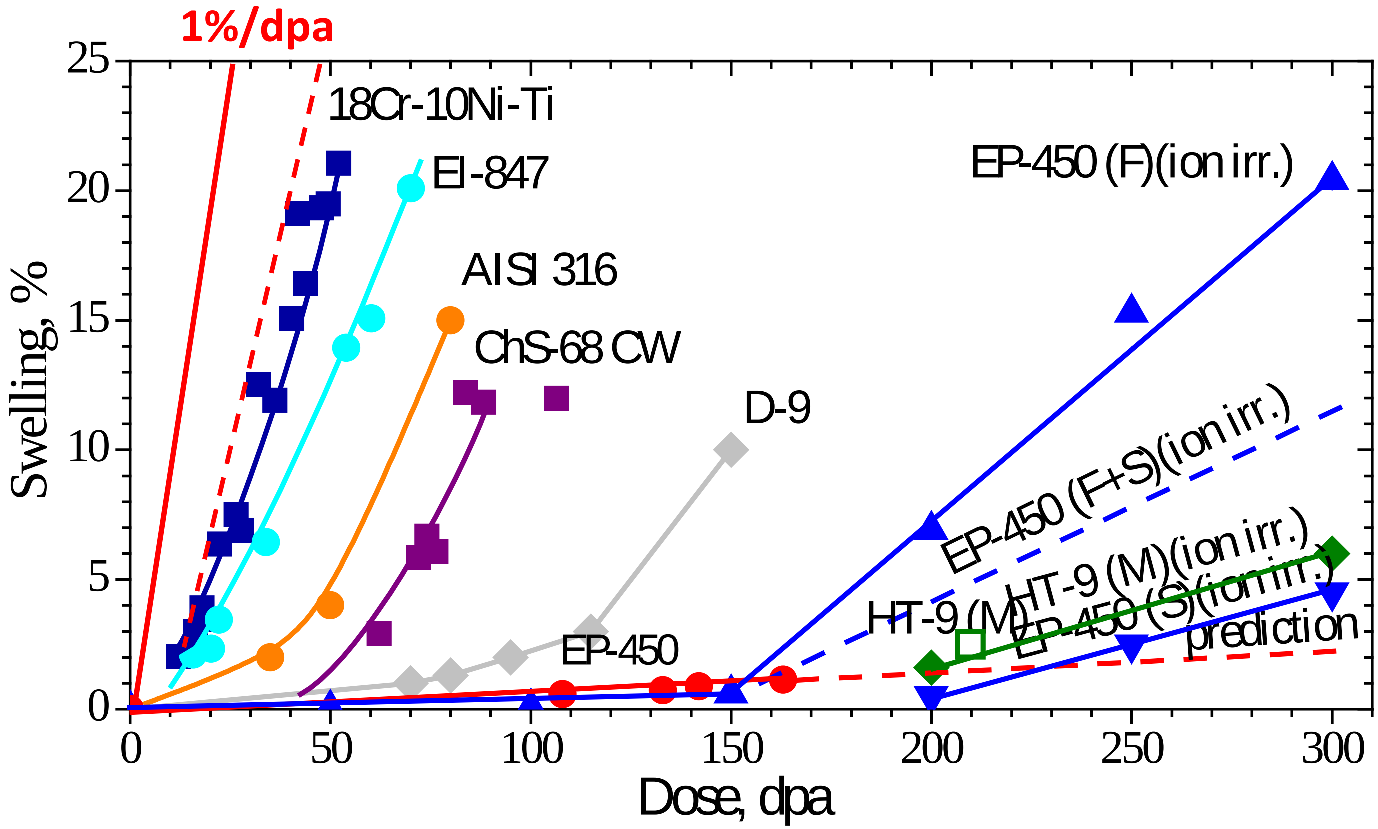
Ion-induced swelling of austenitic and ferritic steels

Conducted at KIPT in Ukraine



Void volume fraction

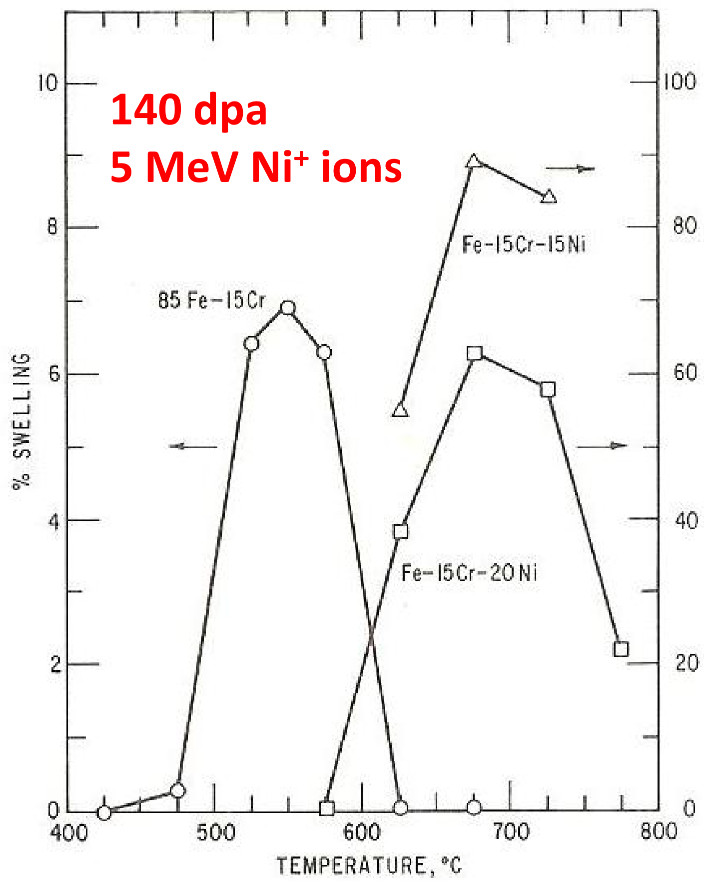
Ion-induced swelling of austenitic and ferritic steels



Void volume fraction → **swelling**

Ion irradiation correctly forecast the different swelling behavior of bcc Fe-Cr and fcc Fe-Ni-Cr alloys

Johnston et al., 1983



Note difference in scales on right and left axis.

1×10^{-2} dpa/sec

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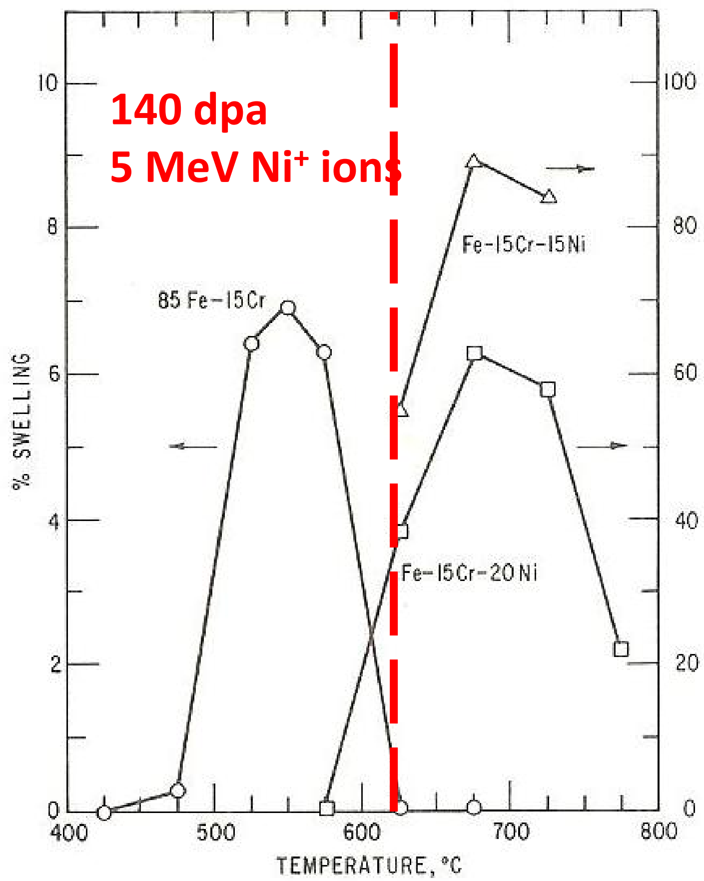
Johnston et al., 1983



Surface of Uranus 50 duplex alloy irradiated at 625°C to 140 dpa

Ferrite grains swell less than austenite grains due to different swelling rate and different temperature regime of swelling

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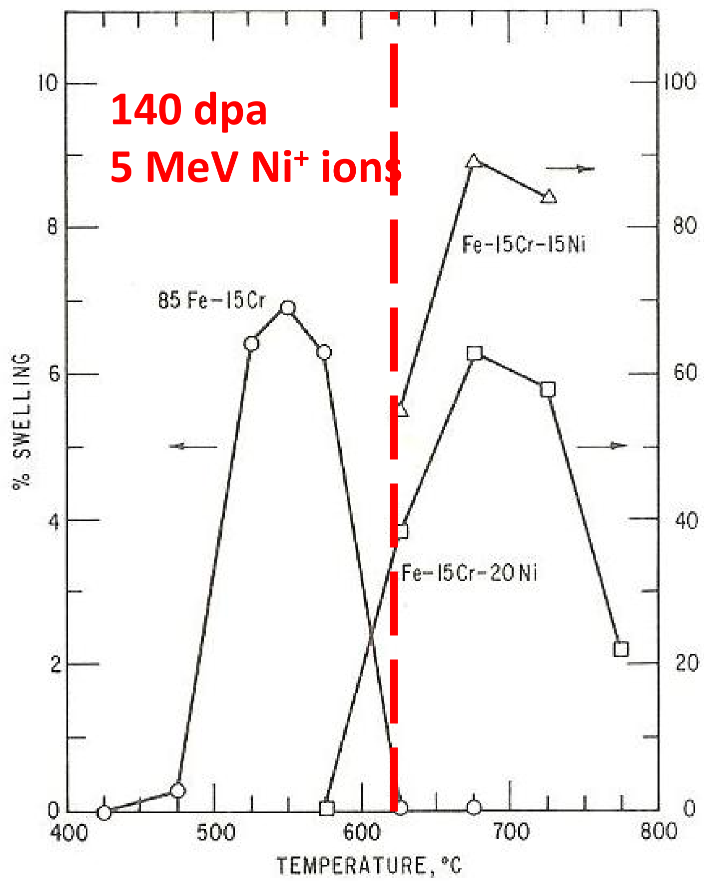

Surface of Uranus 50 duplex alloy irradiated at 625°C to 140 dpa

Ferrite grains swell less than austenite grains due to different swelling rate and different temperature regime of swelling

Fully anisotropic swelling. All mass moves normal to surface.

However, there is not a one-to-one correspondence between ions and neutrons.

- **Primary difference is a very large difference in dpa rates. Radiation-induced phase instabilities that precede onset of steady-state void swelling.**

Point defect balances are very sensitive to dpa rate, leading to classic "temperature shift".

Thermally-driven and radiation-driven phases are known to have different rate constants compared to that of point defects.

Stability of yittria-titania dispersoids probably involves different rate constants.

However, there is not a one-to-one correspondence between ions and neutrons.

- **Primary difference is a very large difference in dpa rates. Radiation-induced phase instabilities.**
- **But there are other very large differences we call "neutron-atypical" variables.**

  Strong effect of nearby surface

  Strong gradients in dpa rate (segregation)

  Compressive stress state in swelling film

  Injected interstitial effect ← very strong!

  Suppression of void nucleation and growth
  Chemical modification by implanted ion

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- **We can use ion bombardment to explore what factors control swelling in ferritic-martensitic alloys.**
- **Other issues: dpa definition, beam sweeping (rastering)**

Calculation of dpa using SRIM code

УДК 620.187:621.039.531

COMMENTS ON DPA CALCULATION METHODS FOR ION BEAM DRIVEN SIMULATION IRRADIATIONS

M.I. Bratchenko, V.V. Bryk, S.V. Dyuldya, A.S. Kalchenko, N.P. Lazarev, V.N. Voyevodin
National Science Center "Kharkov Institute of Physics and Technology", Kharkov, Ukraine

The methodology of application of computer simulation technique to the NSC KIPT advanced simulation studies of radiation materials science at charged particles accelerators is considered with due account of the conformance of simulation methods and algorithms to the working standards of nuclear engineering. The ambiguities of dpa calculations by means of the SRIM code are demonstrated and analyzed using complementary simulations by means of the RaT Monte-Carlo code. The refined guideline of the SRIM dpa calculations is presented.

1. INTRODUCTION

Ion beam simulation irradiation of structural materials is used in NSC KIPT [1–3] and worldwide [4] as a valuable technique of express assessment of their radiation stability under nuclear reactor (n,γ) irradiation. The established standard practice of simulation studies [5] prescribes the calculation of the number of atomic displacements per atom (*dpa* defined [5] as "*a unit of radiation exposure giving the mean number of times an atom is displaced from its lattice site*") as an adopted metric of correlation of the radiation damage relevant effects in metals and alloys subjected to different irradiation environments. This allows comparison of the results of accelerator and reactor based irradiations as well as of those of different experimental groups [4, 5].

The quantification of spatially dependent dpa is a complicated radiation transport problem mostly solved by means of the Monte Carlo (MC) modeling software. The SRIM package [6] is a publicly available [7] practically standard [5] user-friendly tool of such kind of calculations applicable to $\sim 10^{(0\ldots4)}$ keV ion beams irradiation of planar layered targets. The TRIM MC code of the SRIM package simulates depth profiles of irradiation induced vacancy-interstitial Frenkel pairs (FPs) using the binary collisions approximation (BCA) method. Under the assumption that each FP arises in a single atomic displacement, a common practice is to scale dpa at a given ion fluence Φ, cm^{-2}, with the vacancy profile the TRIM code outputs in the VACANCY.TXT file.

The present paper addresses the known issue of this code application to dpa calculations. TRIM offers two options for the FP distributions simulation. The express *"quick damage"* (QD) method simulates only the trajectories of primary ions and the production of primary knock-on atoms (PKAs). The total FP production rate is then calculated analytically within the scope of the modified Kinchin-Pease (K–P) [8, 9] model of the secondary displacement function (SDF) $\nu(T)$, the number of the secondary knock-on atoms (SKAs) produced by a PKA of energy T at a user-supplied value of a stable FP production threshold energy E_d. The alternative *"full cascades"* (FC) damage MC simulation method simulates the overall collision cascade explicitly down to certain cut-off energy $E_{fin} \sim 1$ eV of BCA applicability. The numbers of FPs and atomic replacements are scored collision-by-collision according to certain decision rules based on the values of E_d and the lattice binding energy E_b.

The issue consists in the about twofold discrepancy of the vacancies (and thus dpa) profiles simulated by means of the FC and QD methods (Fig. 1). The ratio is too high to rate it as a reasonable scattering of the estimate of the same physical quantity. It is very probable that the FC/QD methods deviate systematically.

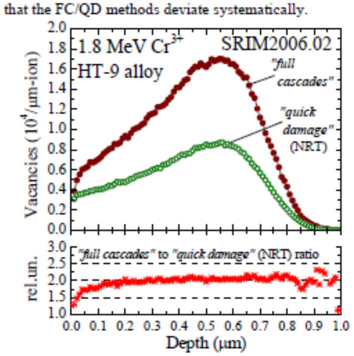

Fig. 1. Depth profiles of the total number of vacancies produced under irradiation of ferritic-martensitic steel HT-9 by 1.8 MeV Chromium ions at the NSC KIPT ESUVI accelerator [2] as calculated using two alternative damage simulation methods of the TRIM BCA code

Neither SRIM manuals [7] nor the ASTM simulation standard [5] comment the origin of this difference. The TRIM simulation method (FC/QD) is seldom specified in publications of experimentalists. This is fraught with misinterpretation of the measured irradiation effect (*e.g.* swelling) in terms of the calculated dpa, *esp.* significant for the topical case of ultra-high (300…600 dpa) damage dose irradiation [2] of prospective reactor materials.

The goals of the present paper are: (i) to uncover physical and algorithmic reasons of the observed discrepancy and (ii) to refine a guideline of the dpa rate calculations by means of the TRIM BCA code.

In sec. 2, we outline the meaning of dpa in radiation material science (RMS) R&D and the methods/models implemented in various codes (*incl.* TRIM) for dpa calculations. Since TRIM is not an open-source software, certain fine details of its algorithms (the calculated dpa seem to be sensitive to) are only poorly documented in [6,7,10] and subjected to changes from one version of the code to another. However, they are extractable from

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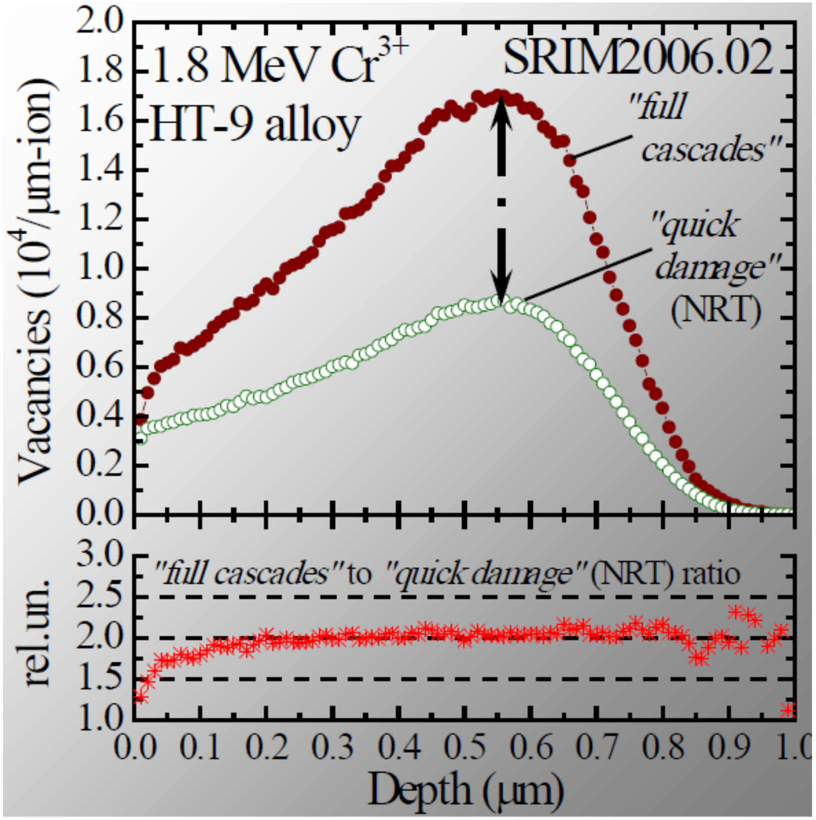

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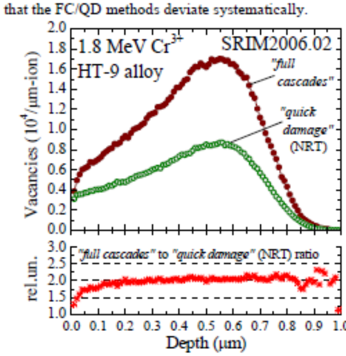

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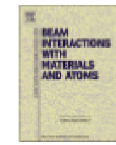

On the use of SRIM for computing radiation damage exposure

CrossMark

R.E. Stoller [a,*], M.B. Toloczko [b], G.S. Was [c], A.G. Certain [b], S. Dwaraknath [c], F.A. Garner [d]

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A R T I C L E   I N F O

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A B S T R A C T

The SRIM (formerly TRIM) Monte Carlo simulation code is widely used to compute a number of parameters relevant to ion beam implantation and ion beam processing of materials. It also has the capability to compute a common radiation damage exposure unit known as atomic displacements per atom (dpa). Since dpa is a standard measure of primary radiation damage production, most researchers who employ ion beams as a tool for inducing radiation damage in materials use SRIM to determine the dpa associated with their irradiations. The use of SRIM for this purpose has been evaluated and comparisons have been made with an internationally-recognized standard definition of dpa, as well as more detailed atomistic simulations of atomic displacement cascades. Differences between the standard and SRIM-based dpa are discussed and recommendations for future usage of SRIM in radiation damage studies are made. In particular, it is recommended that when direct comparisons between ion and neutron data are intended, the Kinchin–Pease option of SRIM should be selected.

Both papers recommend that Kinchin-Pease "quick" option of SRIM must be used to match dpa calculated by SPECTER code for neutron-induced dpa.

Three approaches are being used

- **"Peel the onion" to isolate and identify each neutron-atypical contribution without strong synergisms with other contributions, especially compositional effects**

  Fe, Fe-Cr, Fe-Cr-solutes of increasing complexity

  Effect of production variables: cold work, tempering

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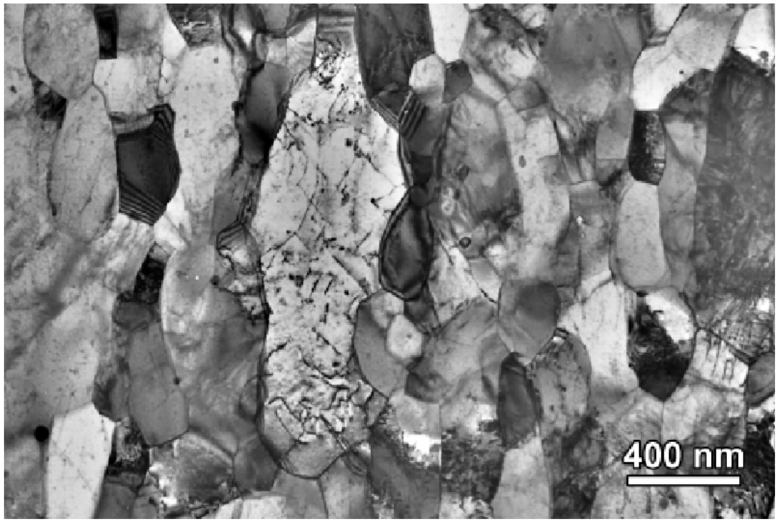
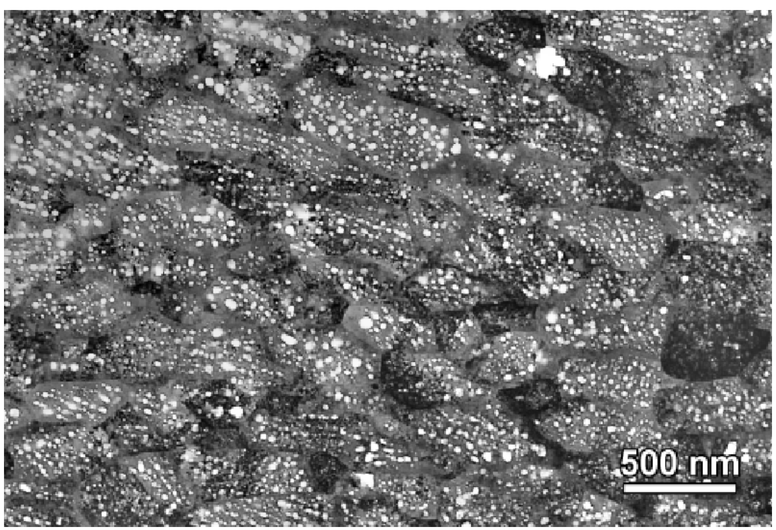
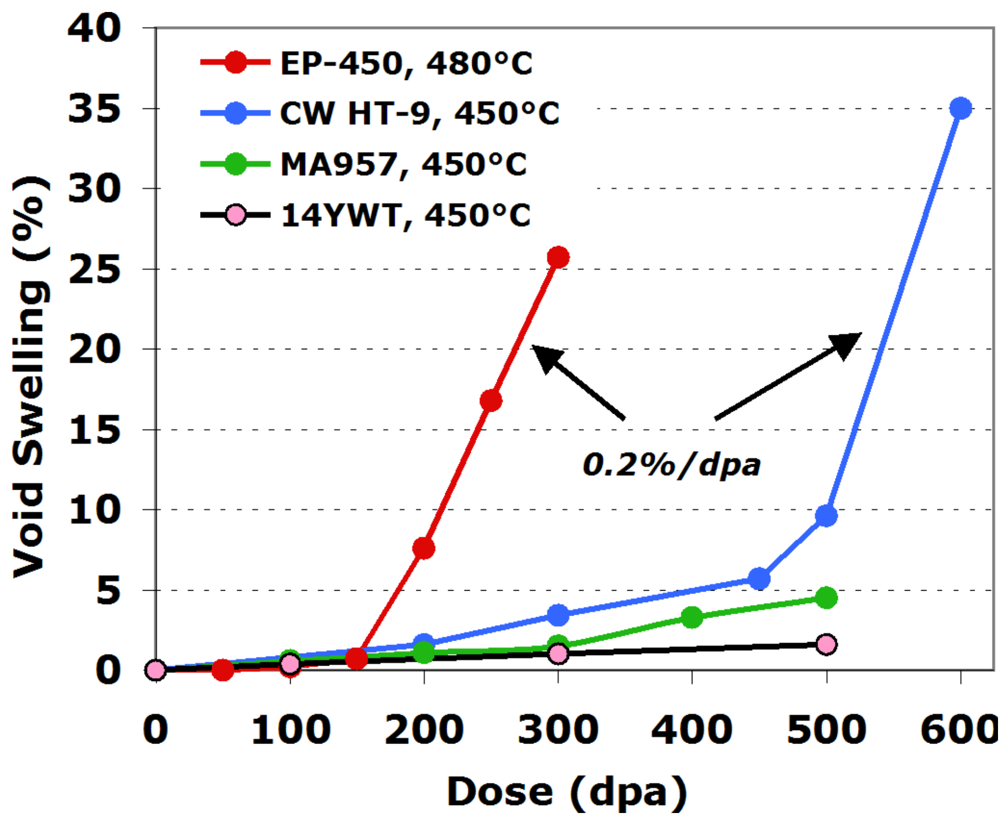
- Examine complex commercial and developmental alloys to identify most promising paths for improvement of alloy radiation resistance.

Comparison of swelling of various ferritic-martensitic and ferritic-ODS alloys (KIPT)





14YWT in unirradiated and irradiated state (500 dpa at 450°C)



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  Effect of production variables: cold work, tempering

- **Examine complex commercial and developmental alloys to identify most promising paths for improvement of alloy radiation resistance.**
- **Determine base behavior of alloy in <u>absence</u> of helium/hydrogen introduction, and then add helium and hydrogen as <u>perturbations</u>.**

“The tail does not wag the dog!”

Three examples from these studies

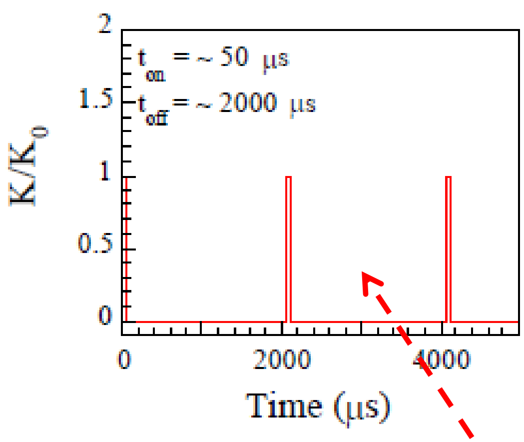
- **Influence of beam-rastering**
- **Injected interstitial suppression of swelling**
- **Stability of various types of dispersoids under irradiation**

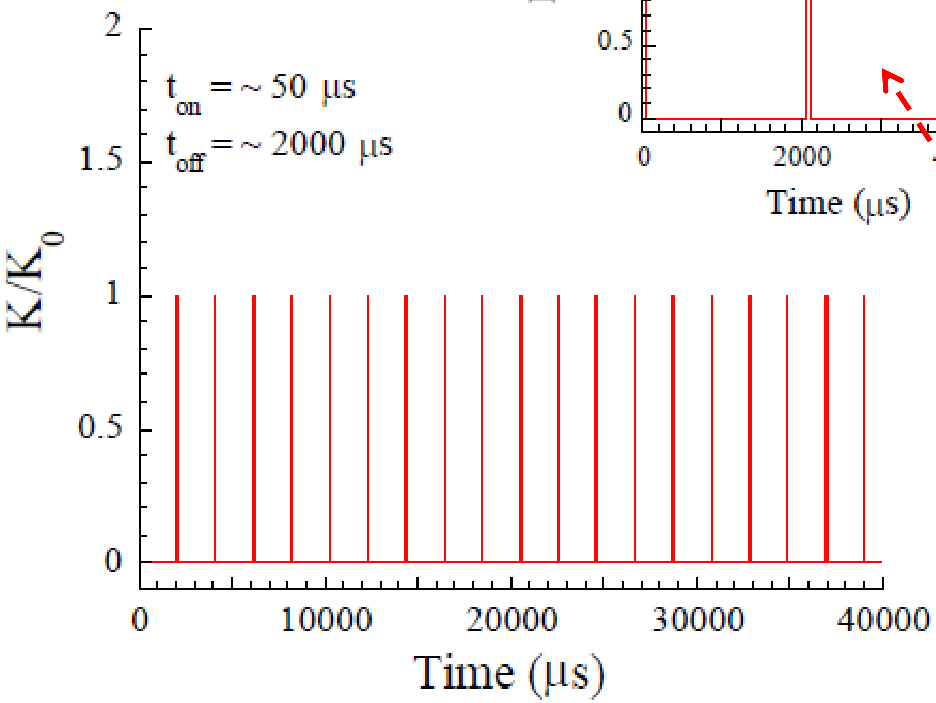
Choice of irradiation conditions

- **Beam conditions: Rastering (beam sweeping) vs. <u>defocused beam</u>**

Duty factor of 2.5%

rastered at
255 Hz horizontally
2061 Hz vertically



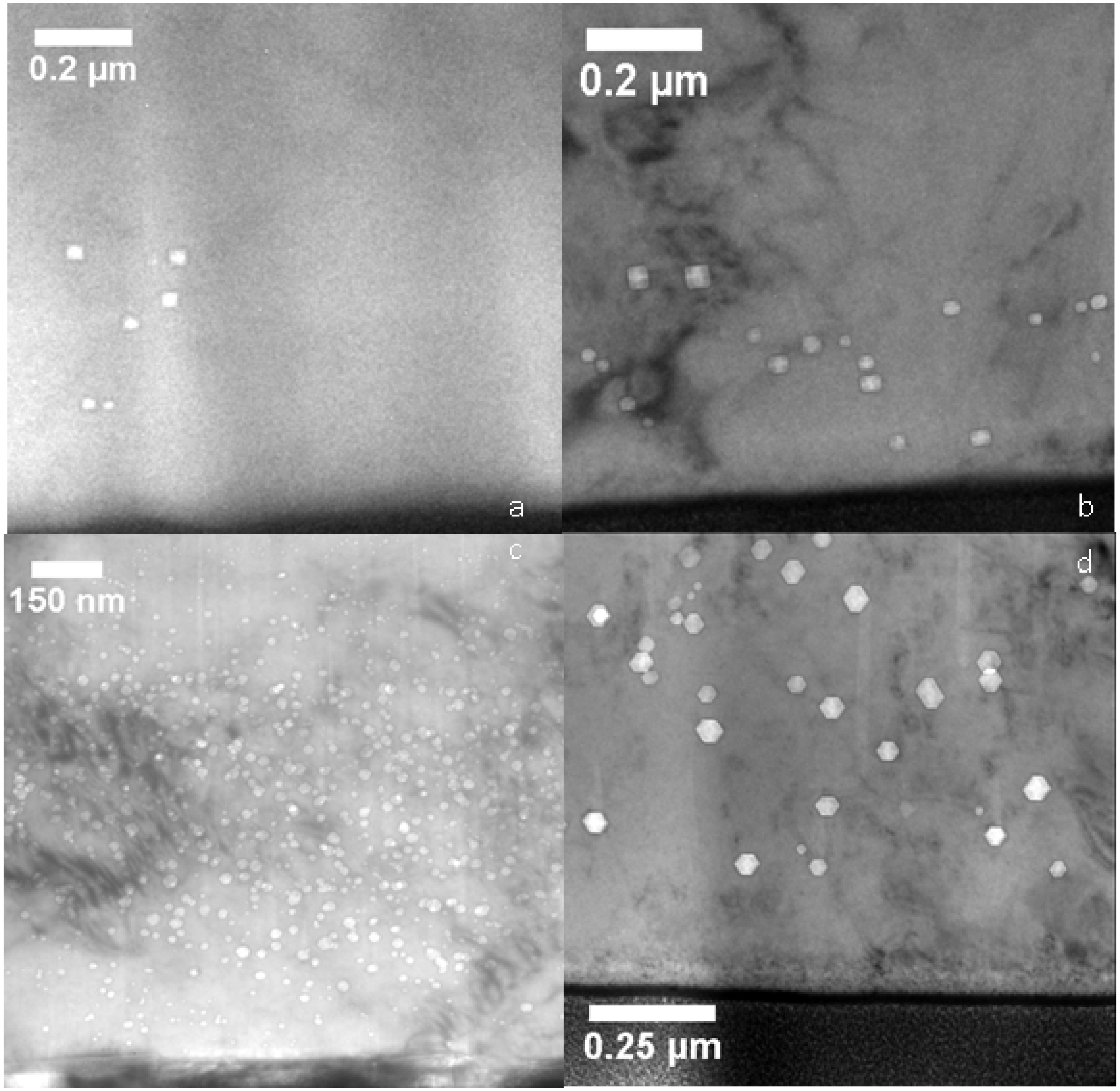


ASTM E521-83. Standard Practice for Neutron Radiation Damage Simulation by Charged-Particle Irradiation

"It is recommended that a rastered beam be avoided for the simulation of a constant neutron flux."

Void nuclei tend to dissolve during between-pulse periods.

Ion-induced swelling of pure iron at 50 dpa



3.5 MeV Fe^{+}
450°C

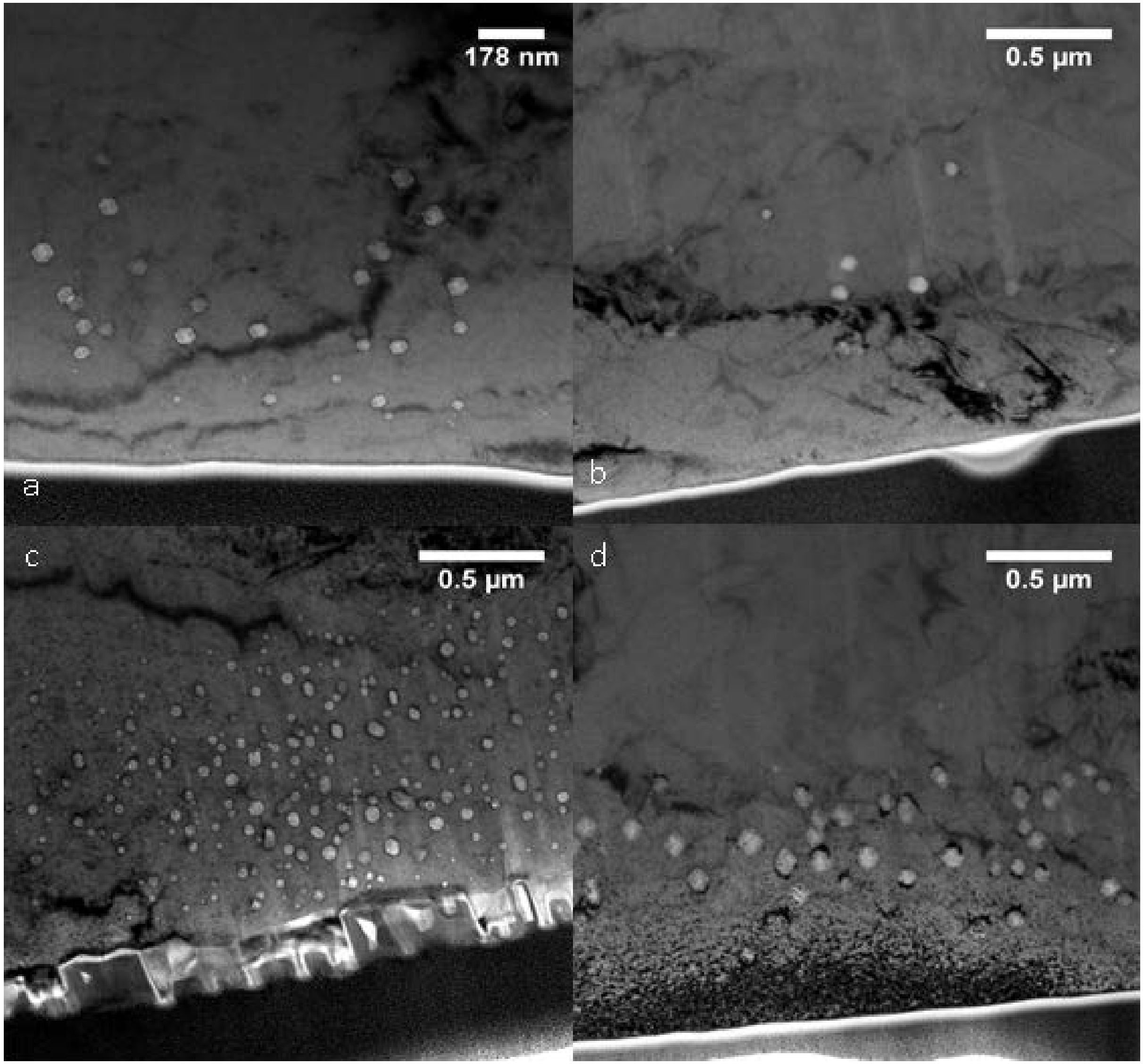
(a) 15.63 Hz

(b) 1.95 Hz

(c) 0.244 Hz

(d) defocused beam

Ion-induced swelling of pure iron at 150 dpa



3.5 MeV Fe^{+}
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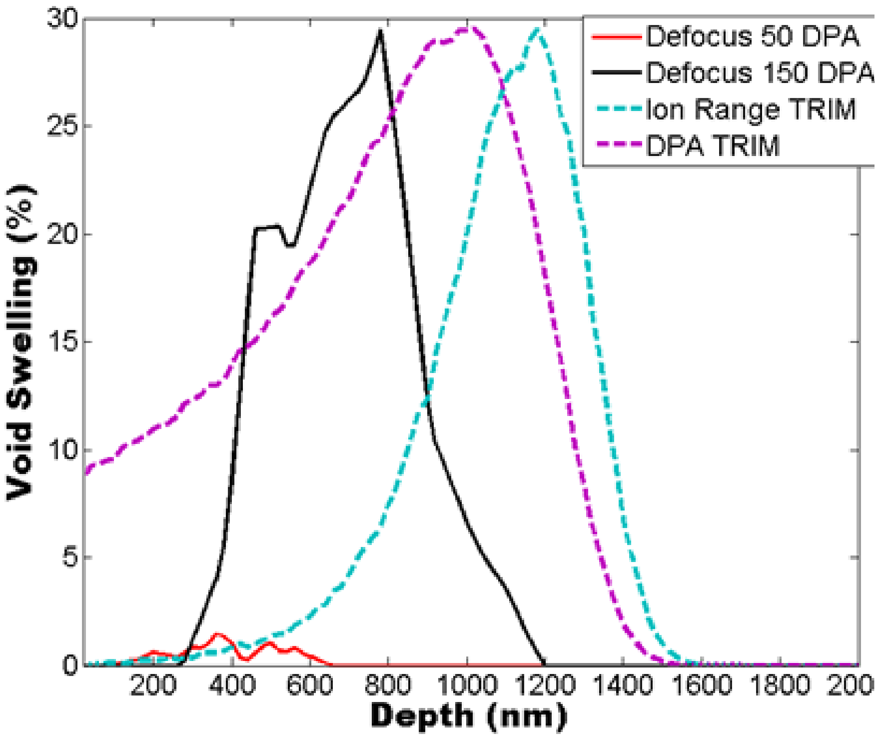
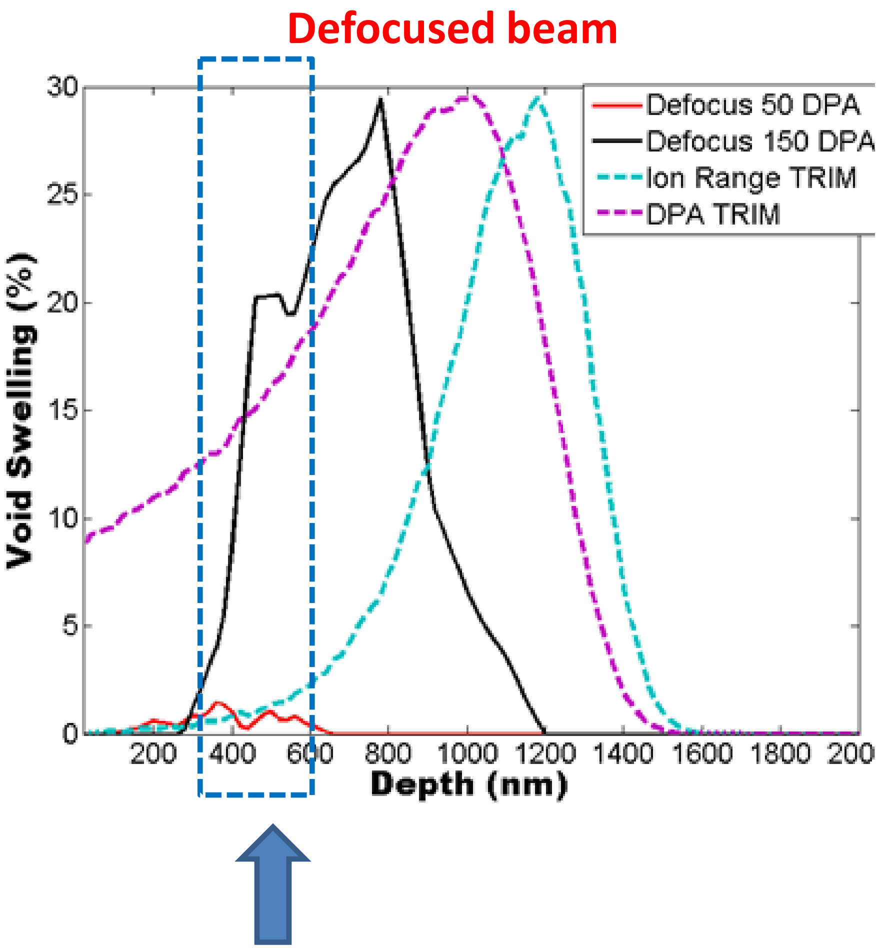
Depth dependence of swelling in raster-no-raster experiments in pure iron at 450°C

Defocused beam



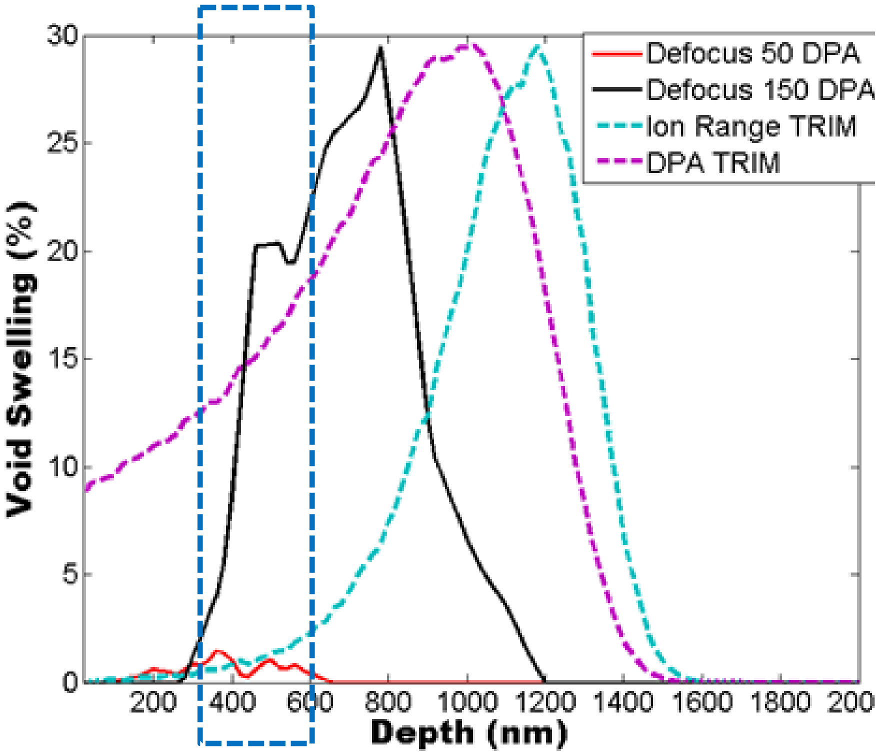
Depth dependence of swelling in raster-no-raster experiments in pure iron at 450°C



Swelling data extracted from here to minimize the effect of injected interstitials.

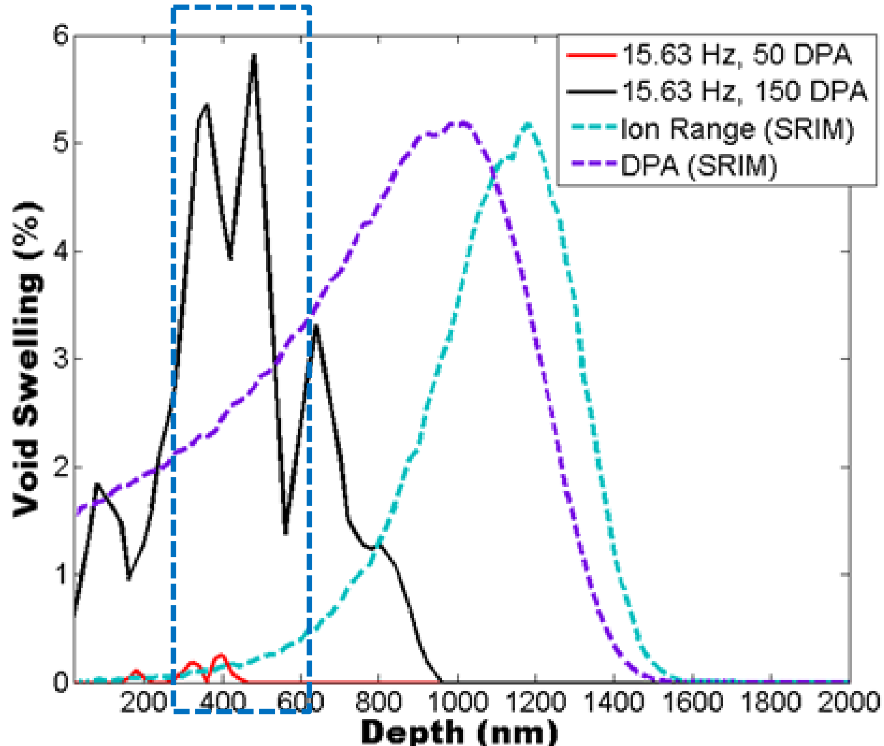
Depth dependence of swelling in raster-no-raster experiments in pure iron at 450°C

Defocused beam



Swelling data extracted from here to minimize the effect of injected interstitials.

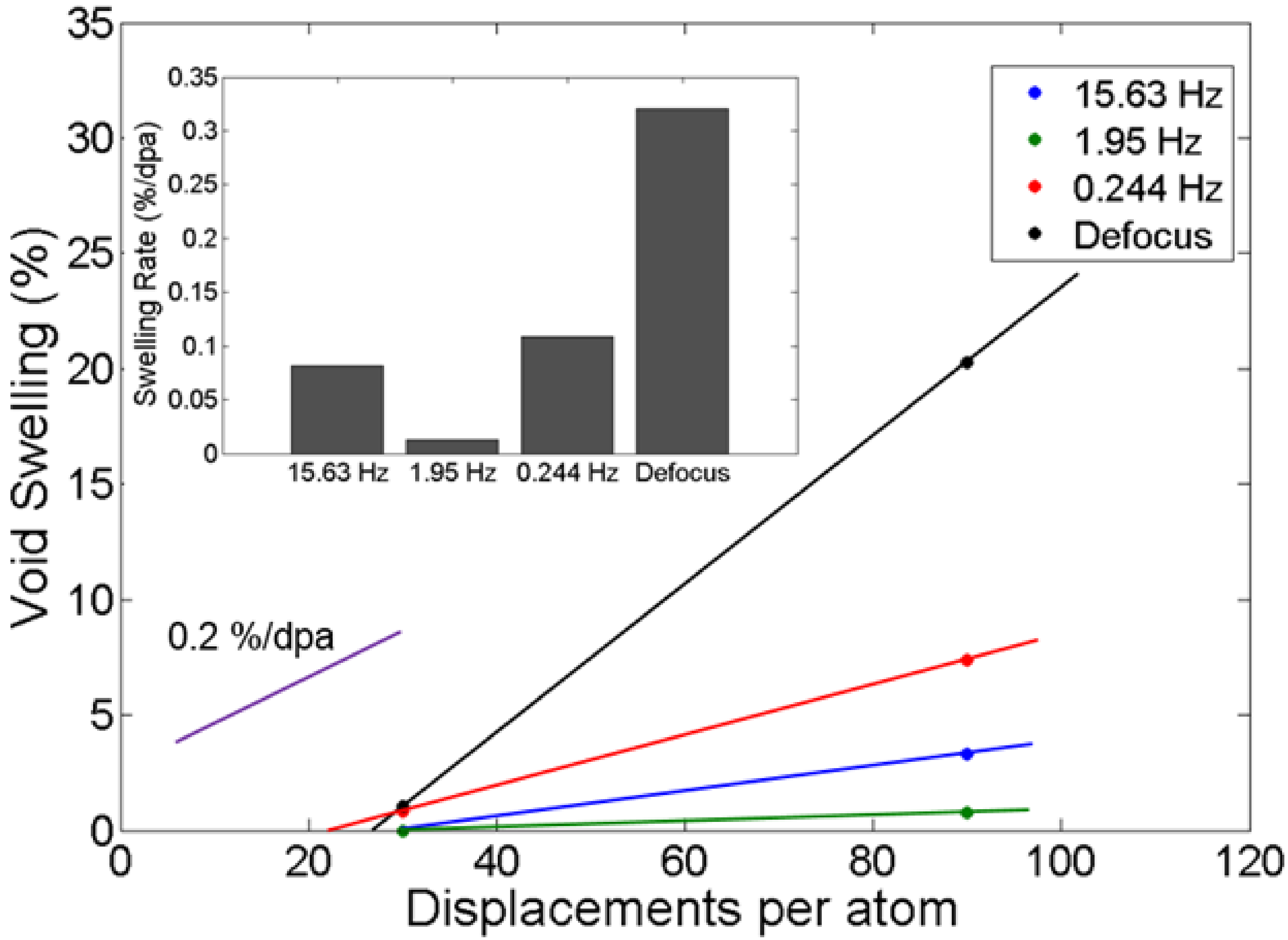
Rastered beam



Vacancy Rich

Interstitial Rich

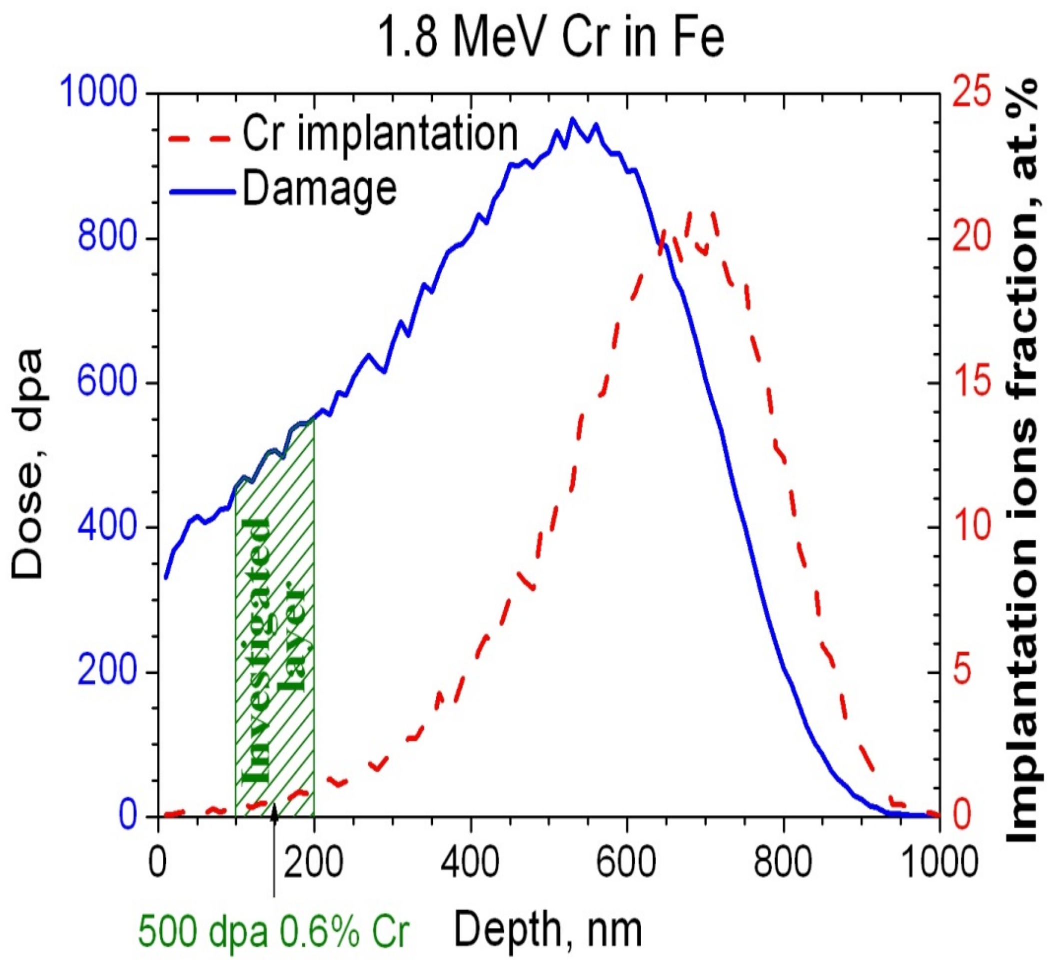
Swelling rate comparison in raster-no-raster experiments



Displacement profile calculated by SRIM code

Incident

ions



~500 dpa in sampled region

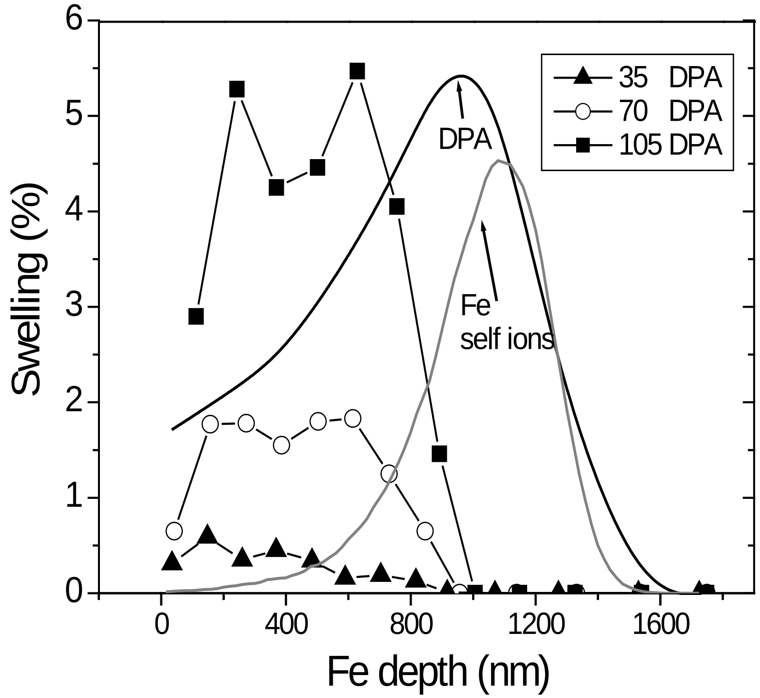
>900 dpa at peak

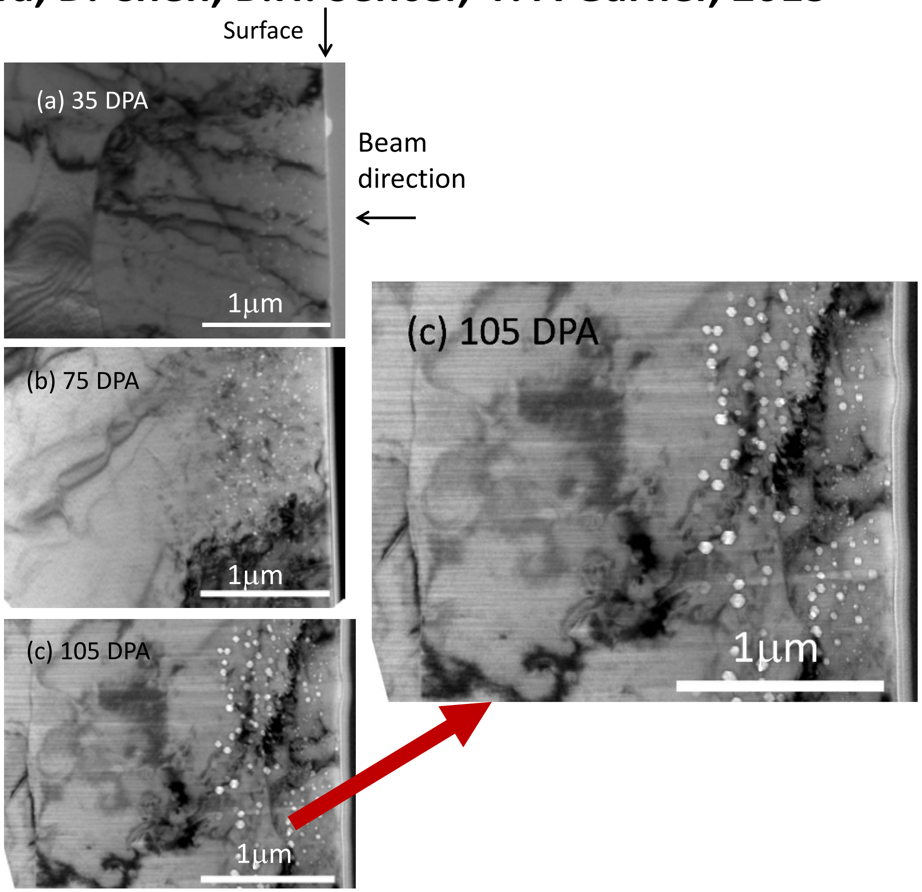
Large addition of chromium by injected ion

Injected interstitial effect to suppress void nucleation

Strong suppression of void nucleation in pure iron by injected interstitial

L. Shao, C.-C. Wei, J. Gigax, A. Aitkaliyeva, D. Chen, B.H. Sencer, F. A Garner, 2013





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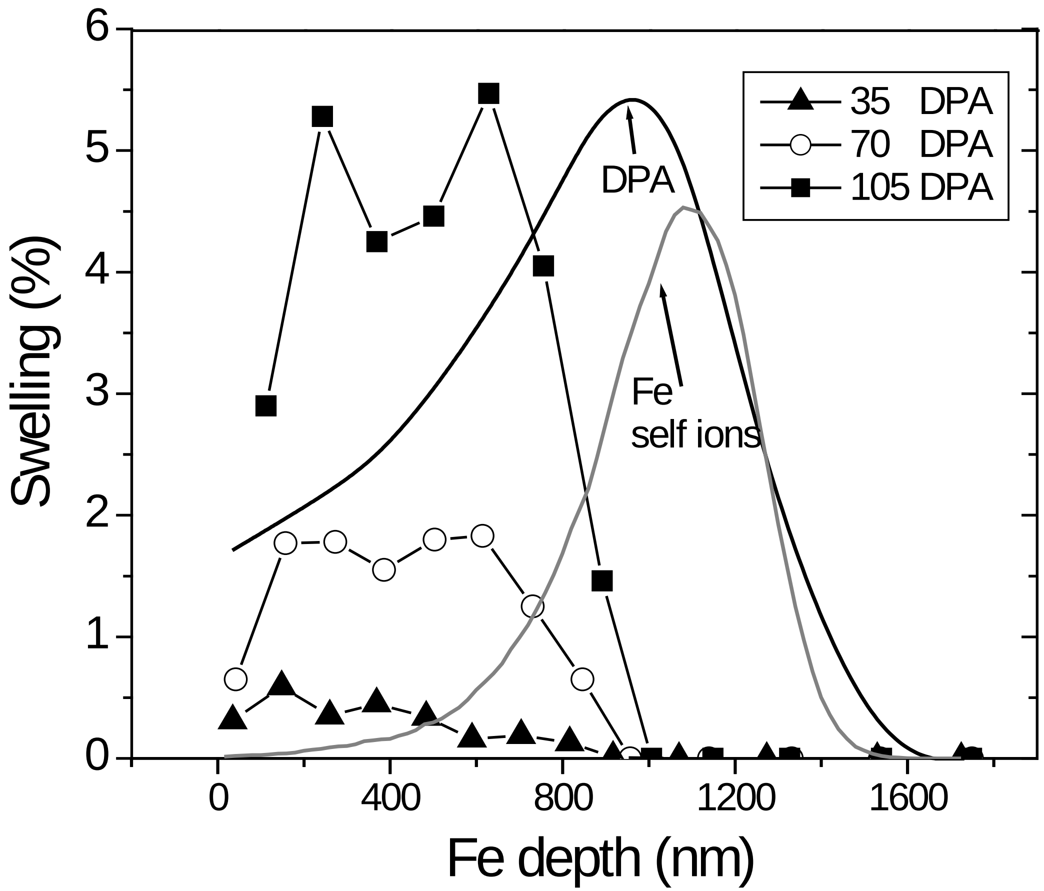


3.5 MeV Fe^{+} at 450°C with rastering

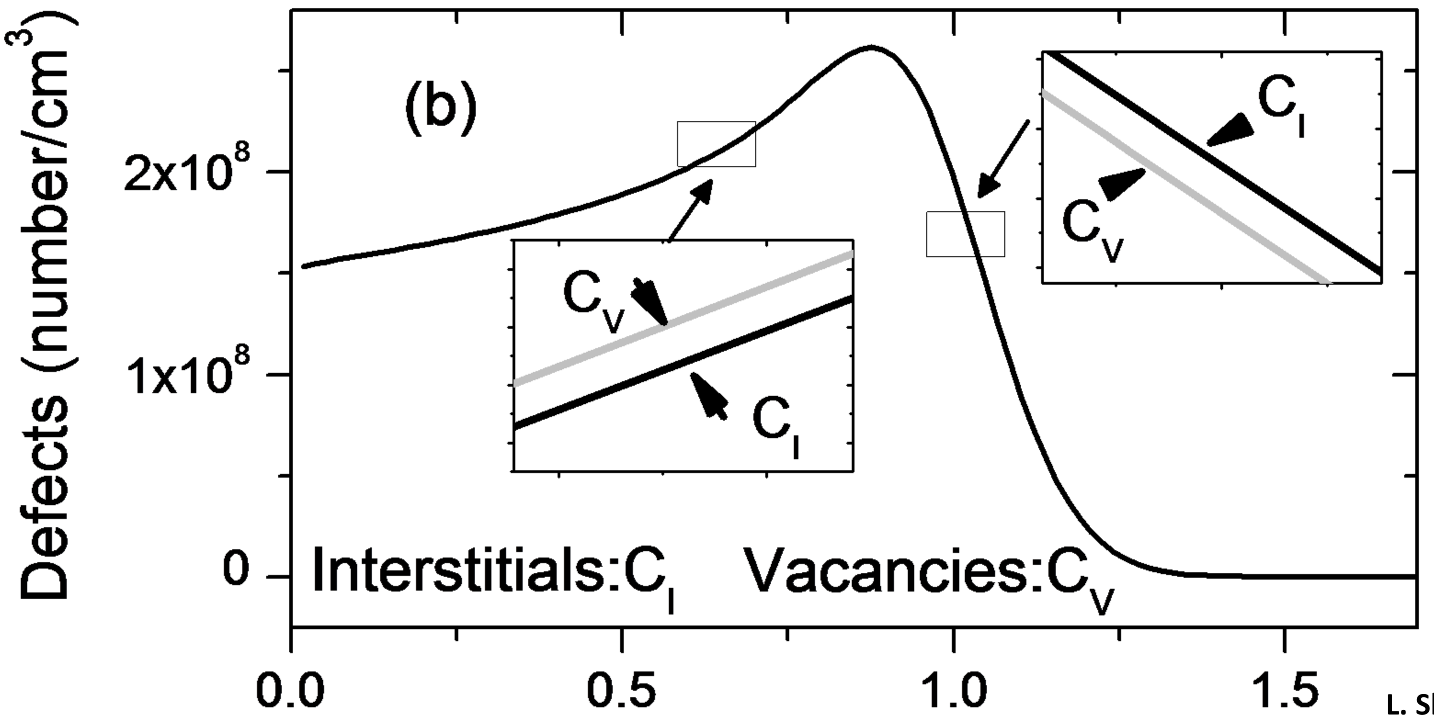
Dose defined at peak damage region.

Double peak swelling is not an artifact but a consequence of "defect imbalance".

Combined influence of forward scattering and injected interstitial

Change in point defect concentrations due to forward scattering

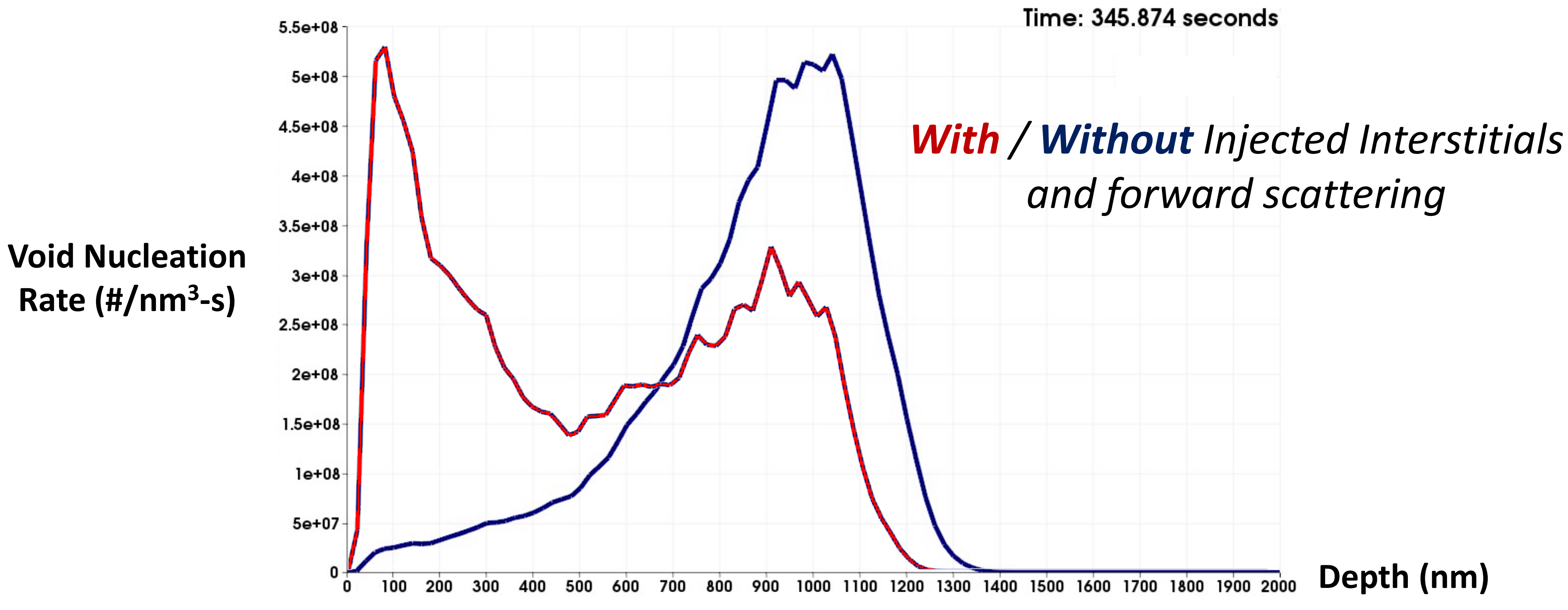
- … and its effect may look very small…



L. Shao et al. (2014)

Change in point defect concentrations due to forward scattering

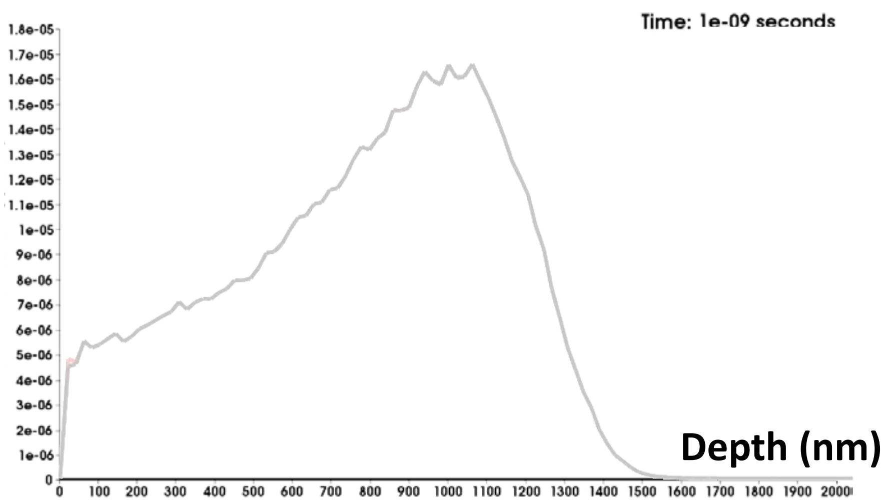
- … but its effects on void swelling are strong!



Results – Point Defects

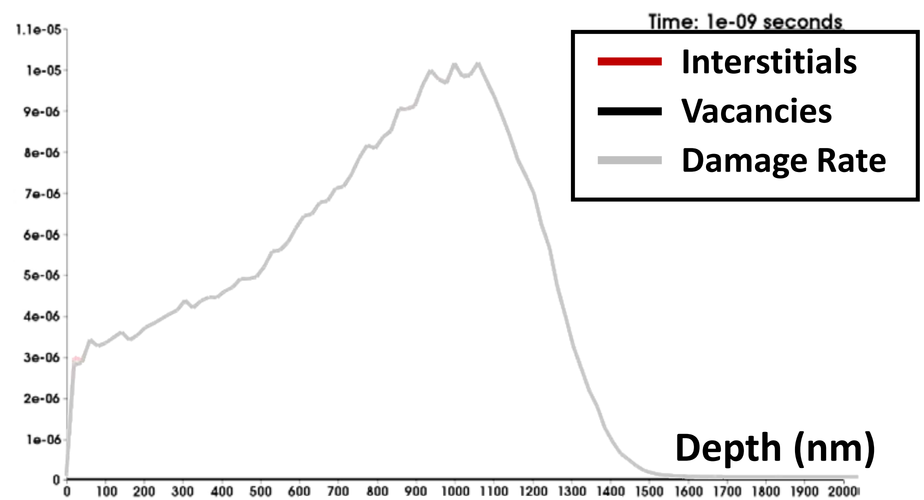
3.5MeV Fe^{+2}, 1mA, $1mm^2$ beam, 450C, $E_M^V = 0.66eV$

Point defects follow SRIM forcing function



Without **injected interstitials**

Point defects do not quite follow SRIM forcing function

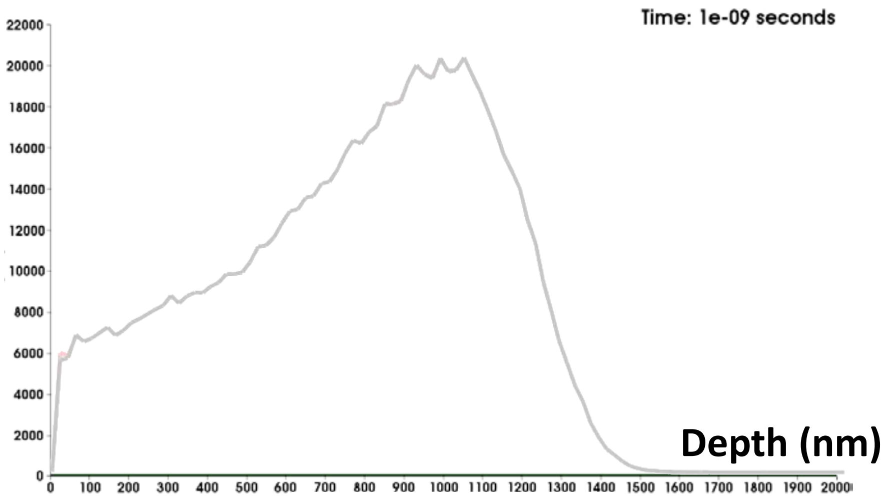


With **injected interstitials**

Results – Vacancy Supersaturation

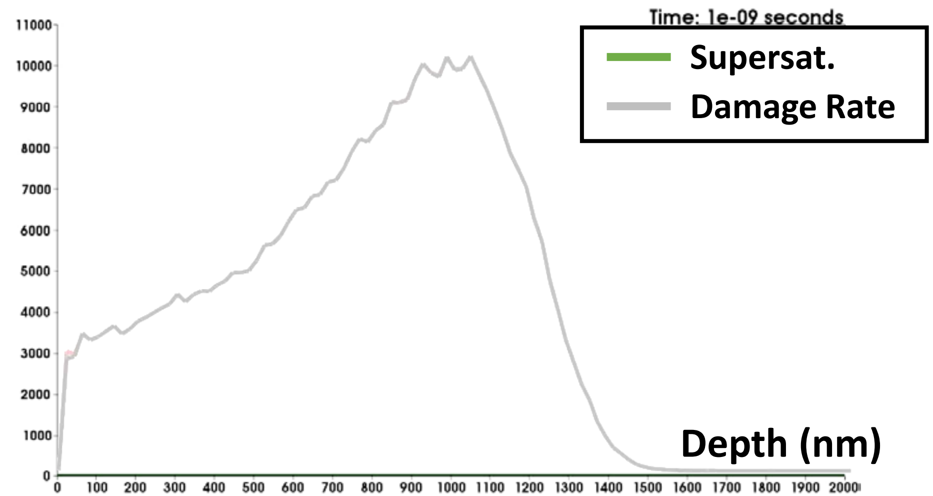
3.5MeV Fe^{+2}, 1mA, 1mm^2 beam, 450C, $E_M^V = 0.66$eV

Peaks near maximum damage region



Without **injected interstitials**

Bimodal distribution, shifted to the left by 100nm

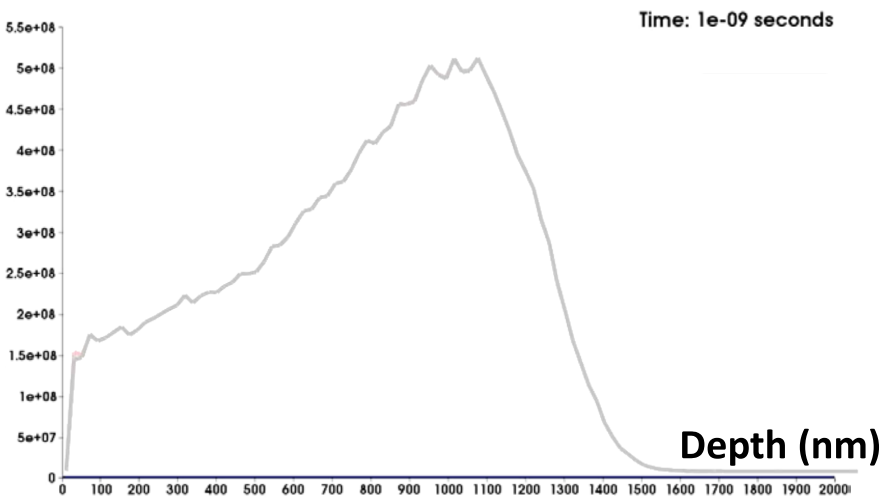


With **injected interstitials**

Results – Void Nucleation Rate

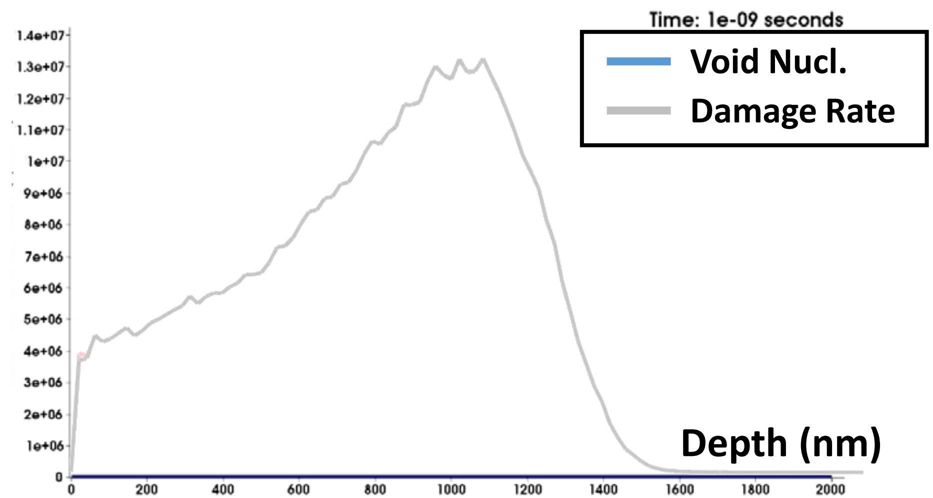
3.5MeV Fe^{+2}, 1mA, 1mm^2 beam, 450C, $E_M^V = 0.66$eV

Clearly peaks at maximum damage region



***Without* injected interstitials**

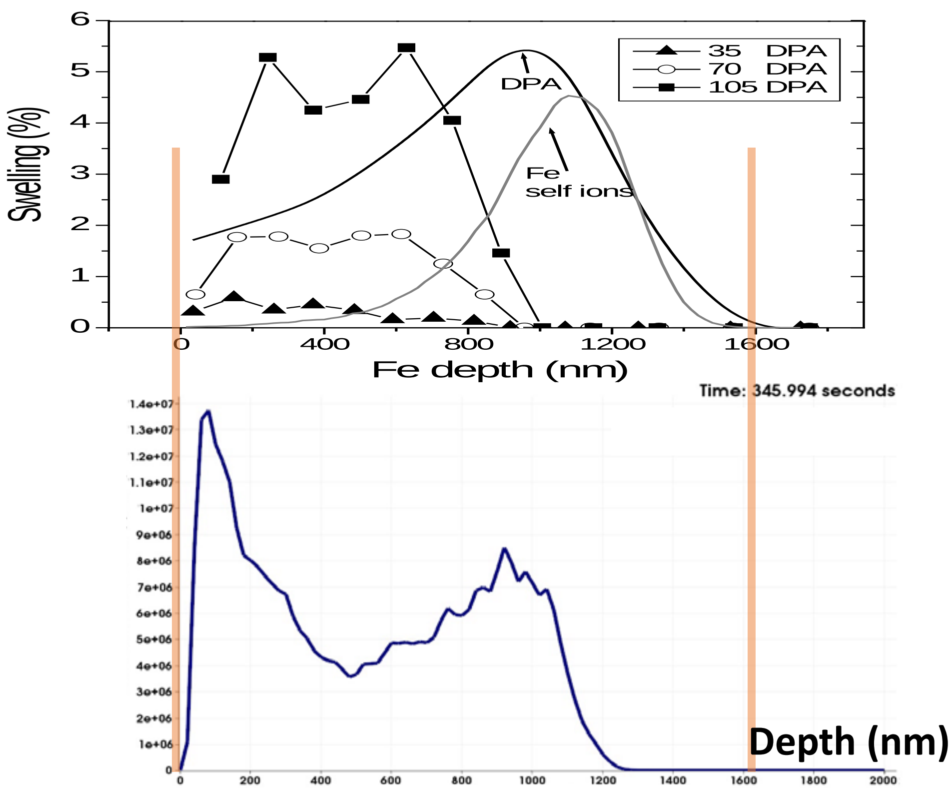
Very bimodal distribution, shifted to the left by 100nm



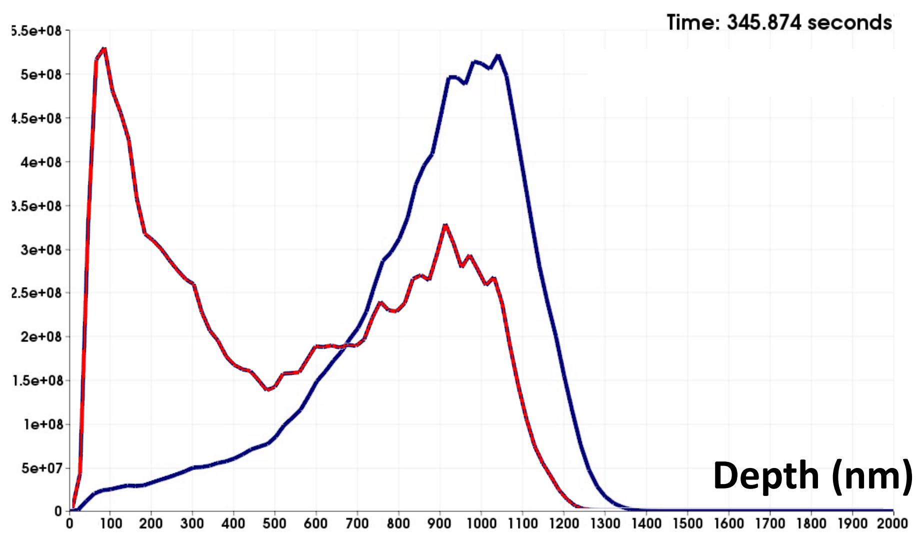
***With* injected interstitials**

Compare with Experiments

3.5MeV Fe^{+2}, 1mA, 1mm^2 beam, 450C, $E_M^V = 0.66$eV

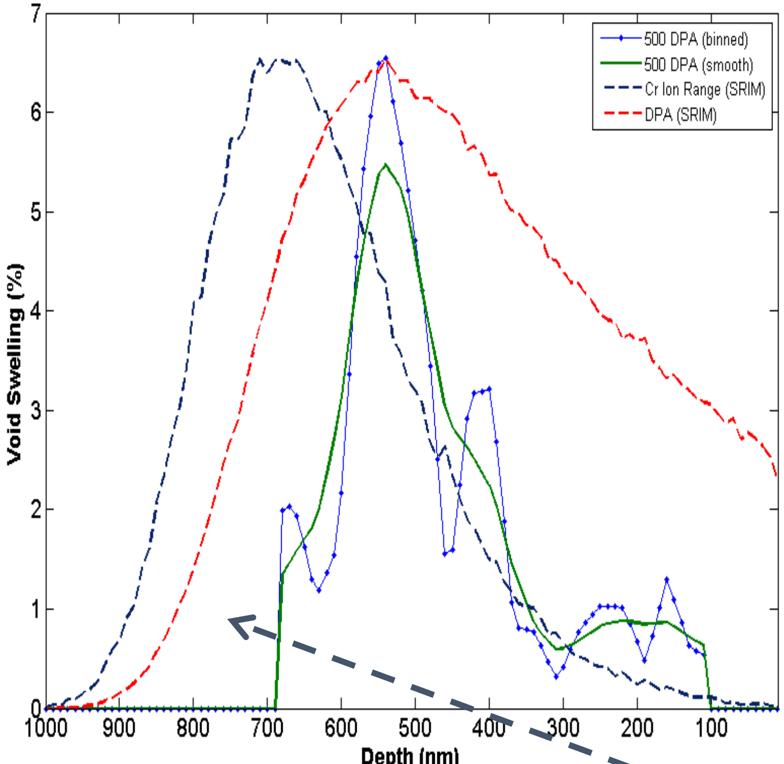


Simulation vs. Experiments

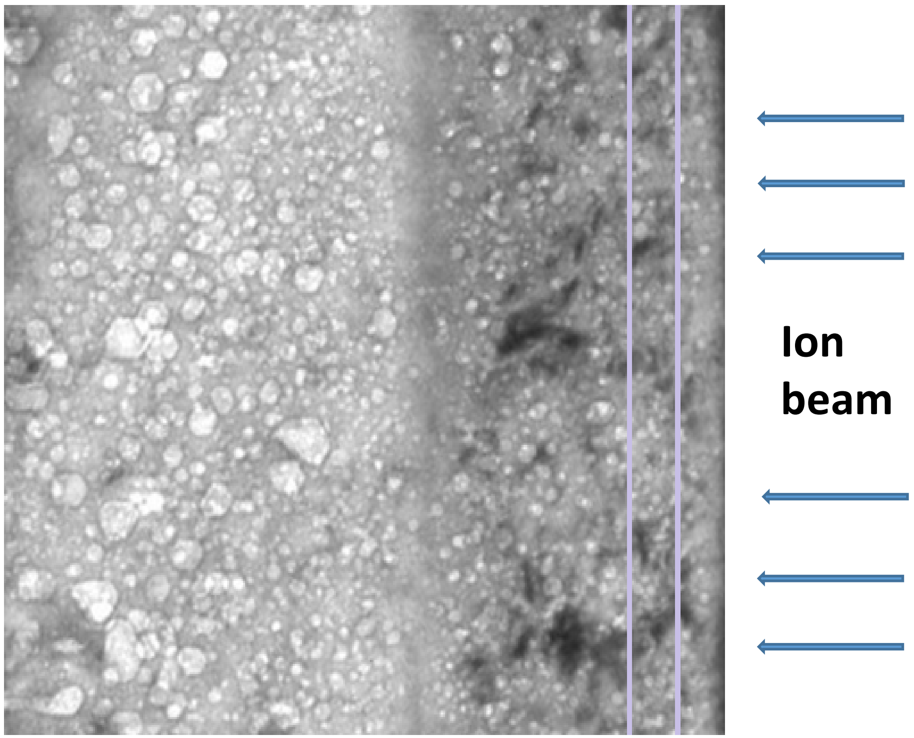


With / Without Injected Interstitials

Depth dependence of swelling in MA957 at 900 dpa peak and 425°C



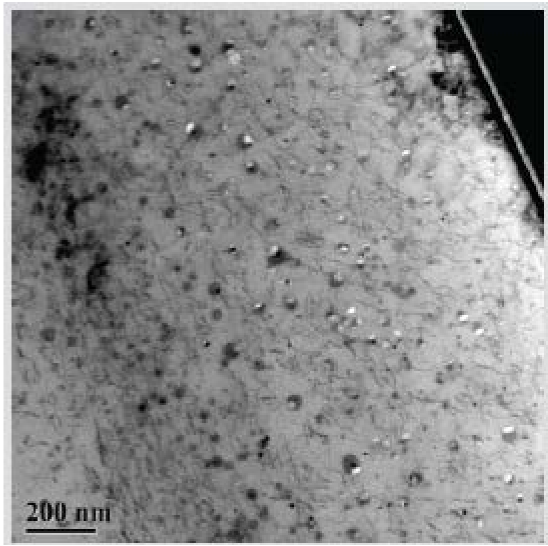
Grain boundaries

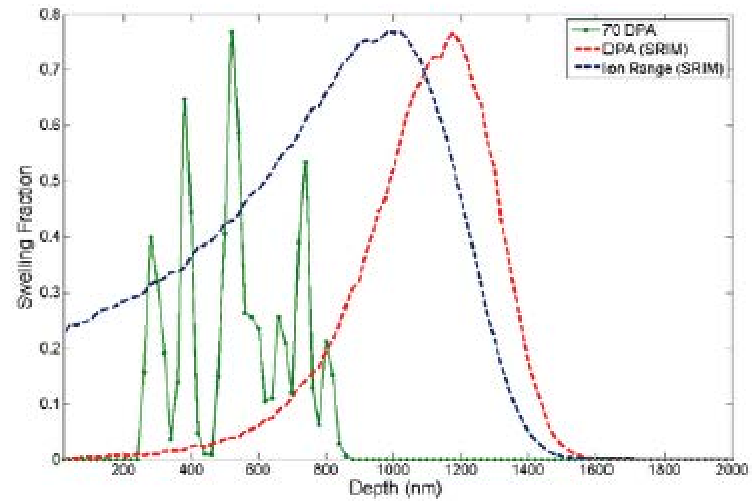


Strong effect of injected interstitial to suppress void nucleation

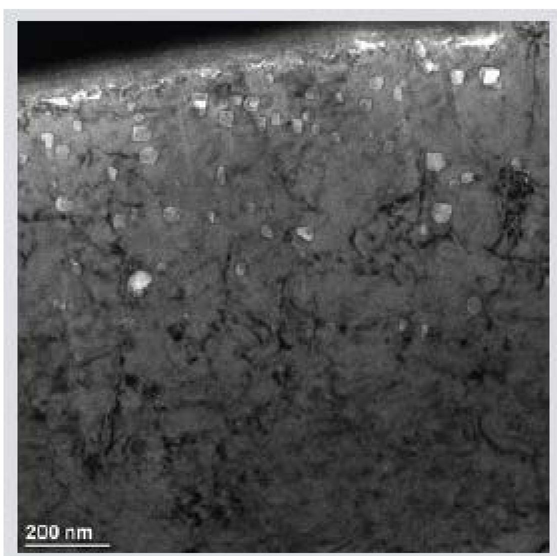
Impact of injected interstitial on swelling of MA956

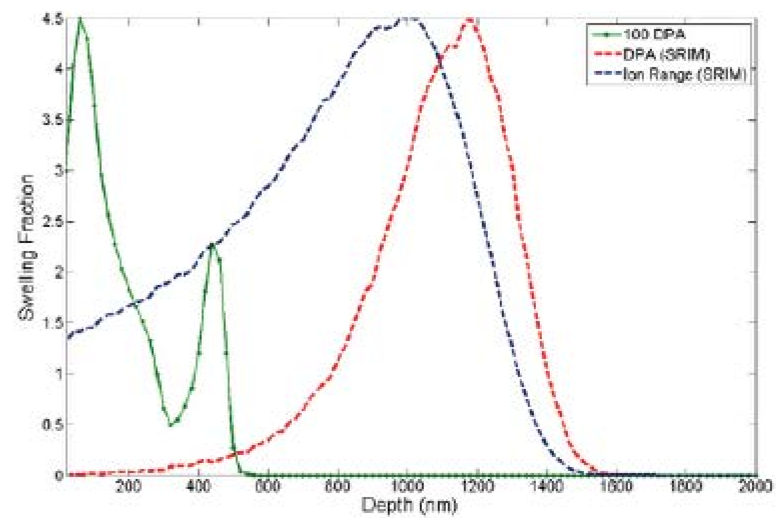
70 dpa peak at 450°C



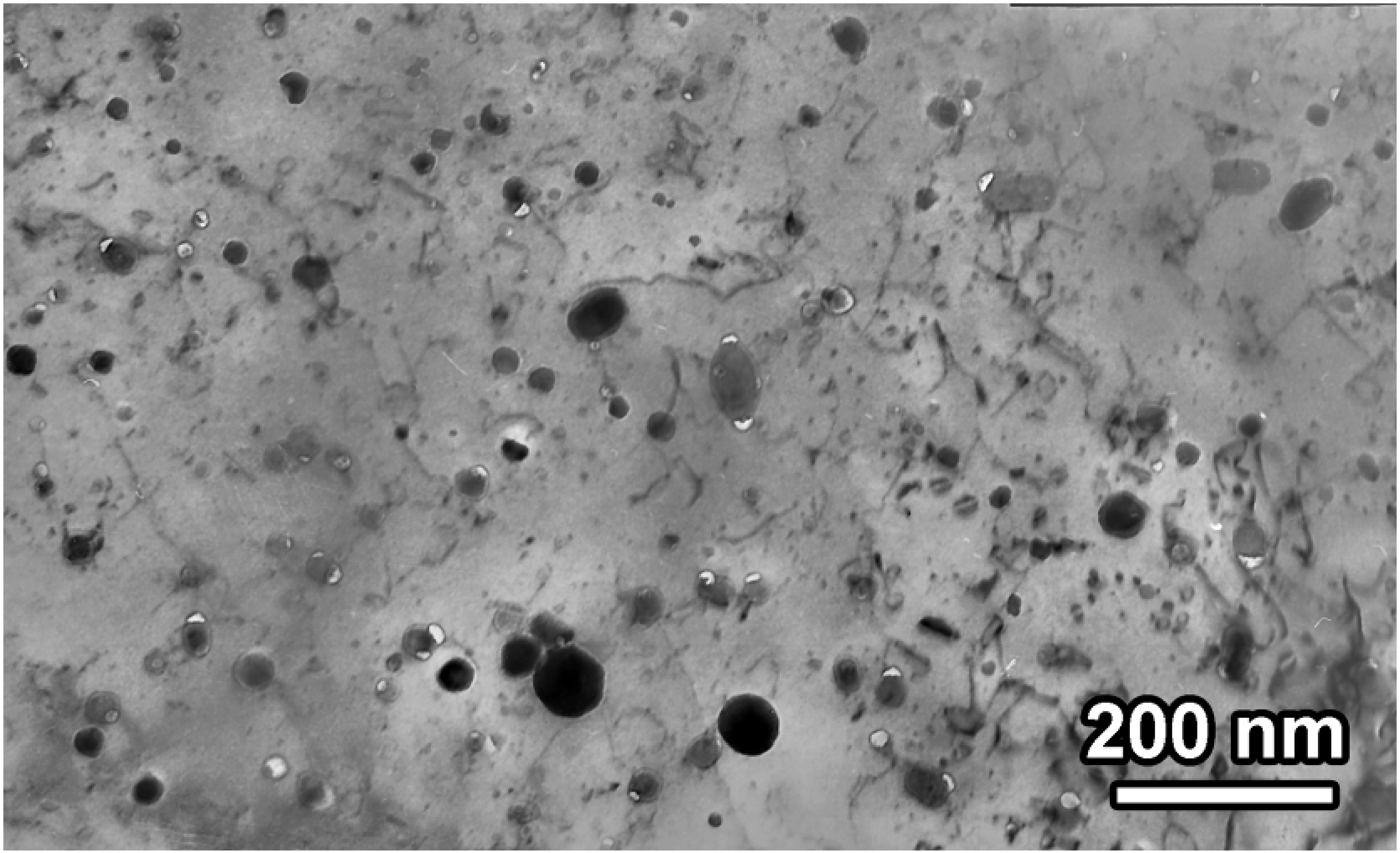


100 dpa peak at 500°C

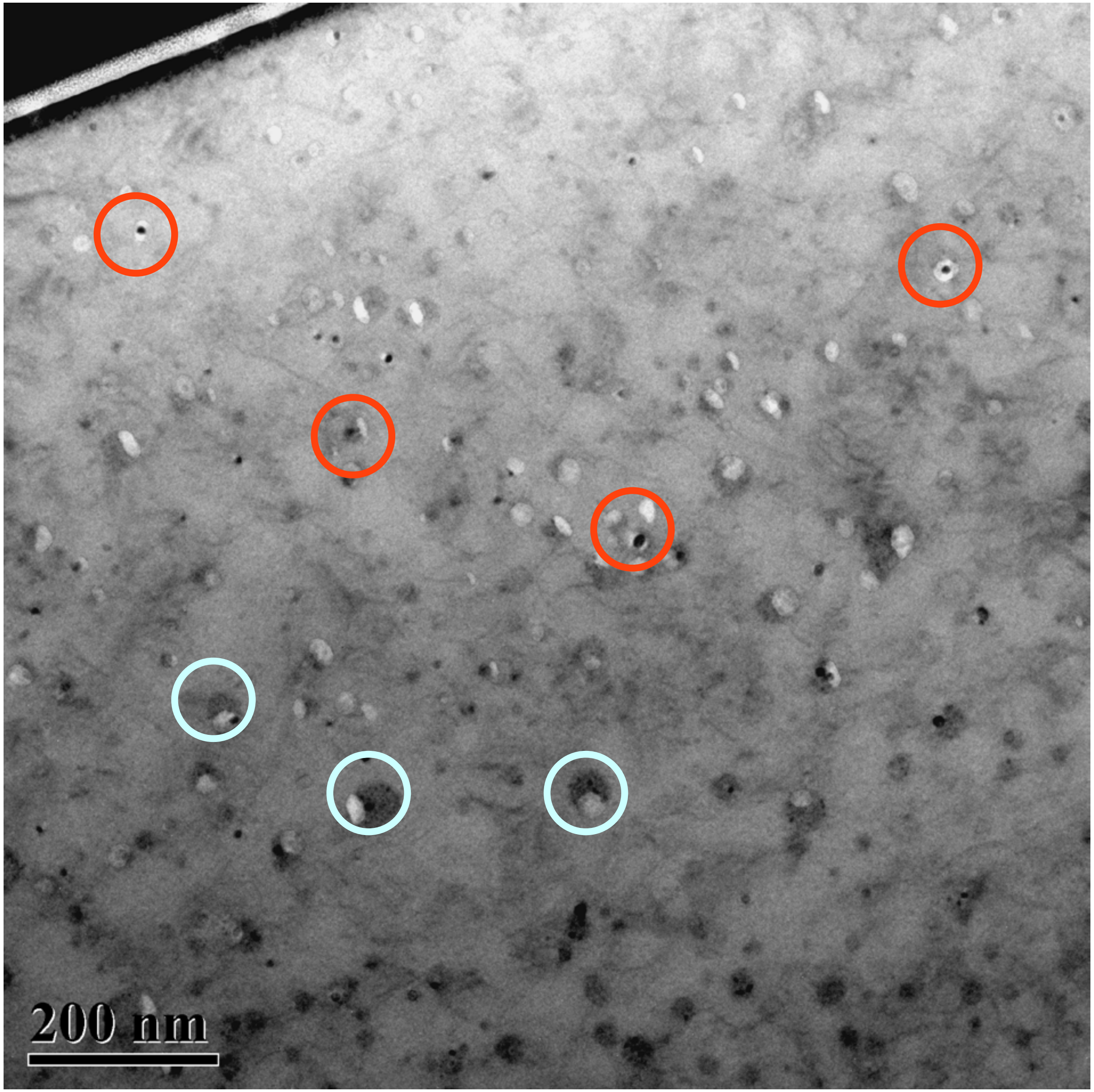




MA956 at 450°C and 100 dpa irradiated with 1.8 MeV Cr^+ ions at KIPT



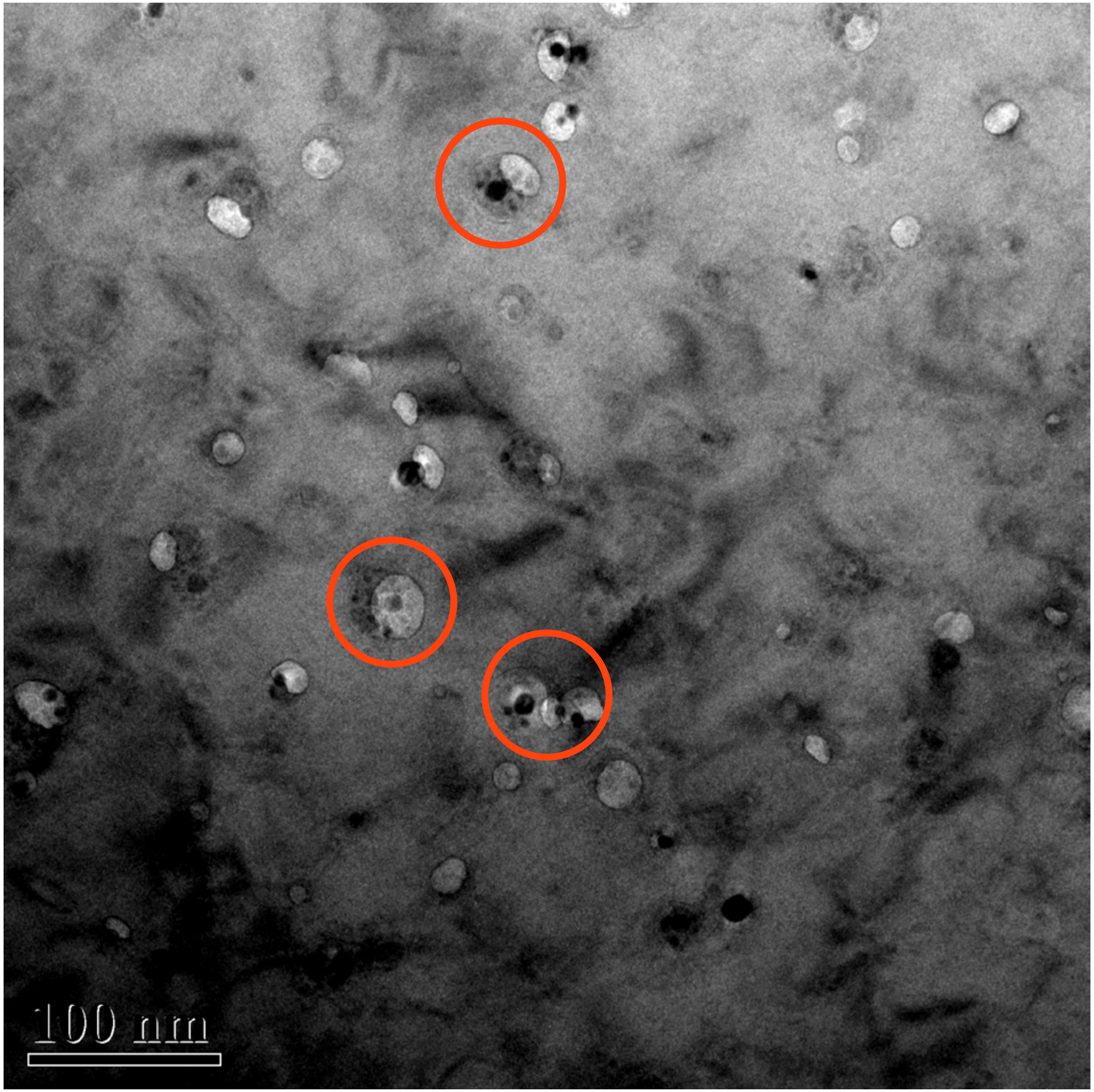
MA956: 3.5 MeV Fe^{+} at 70 dpa peak and 450°C (TAMU)



Voids are generally attached to the interface between oxide particles and matrix.

Oxide particles appear to be decomposing.

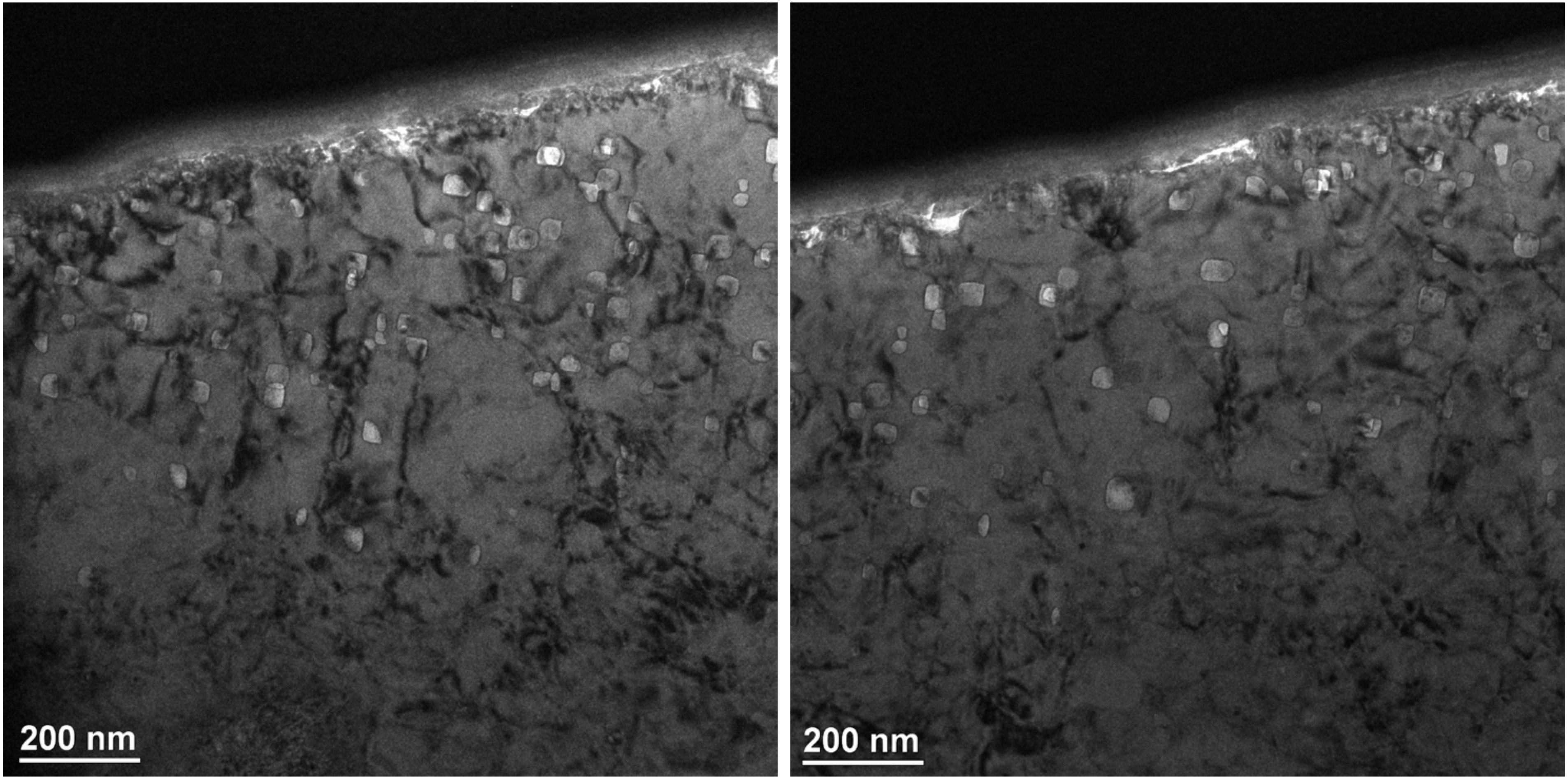
MA956: 3.5 MeV Fe^{+} irradiation at 70 dpa peak and 450°C



Oxide particles appear to becoming amorphous.

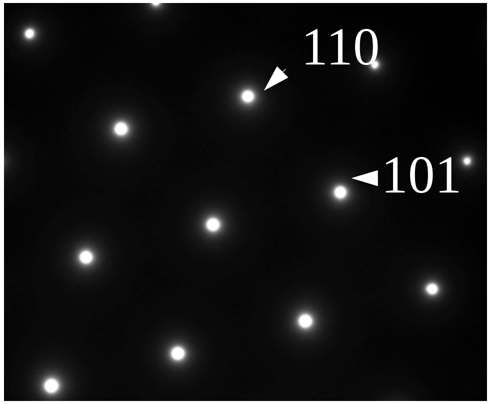
MA956: 3.5 MeV Fe^{+} irradiation at 100 dpa peak and 500°C

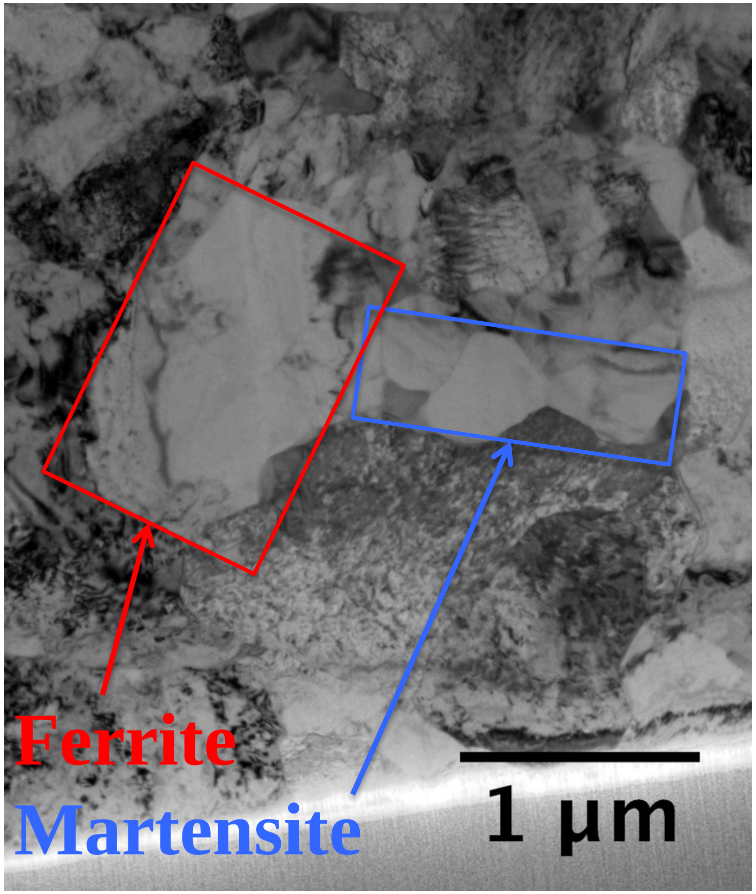
Oxide particles have dissolved in the irradiated region.



Novel alloy with dispersoids in both phases

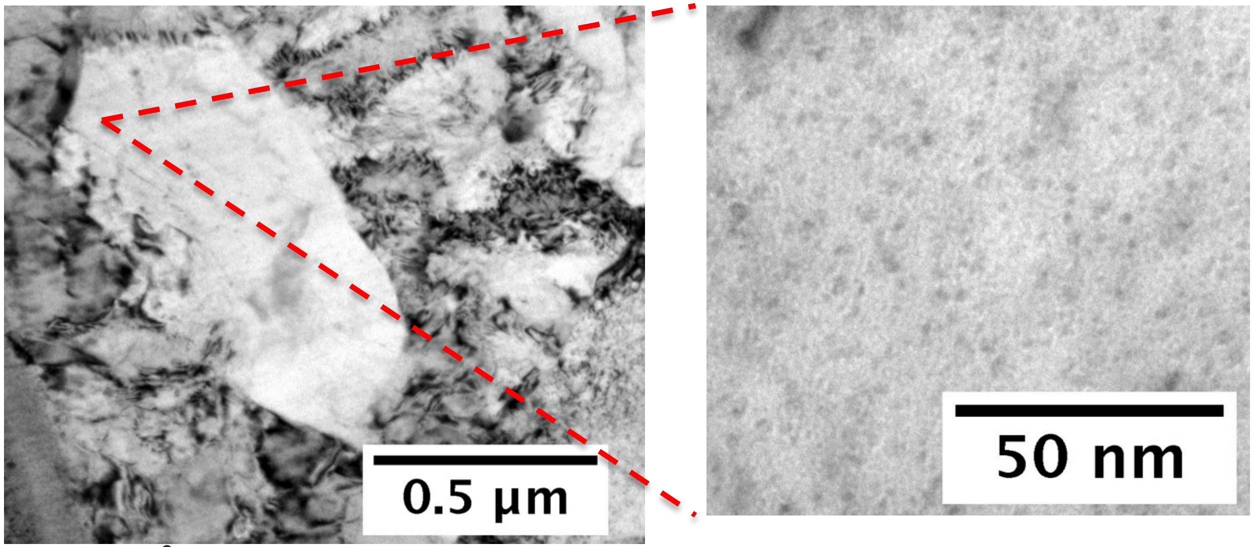
- Residual ferrite:
  - 10%, ~1µm grains
- Martensite:
  - 90%, ~200nm grains



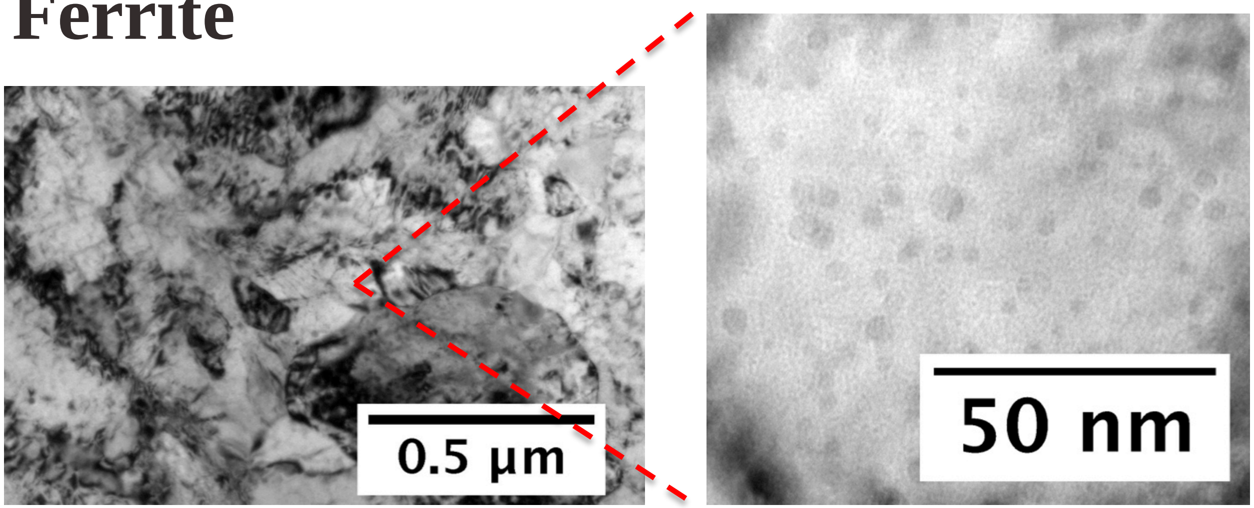


Ele.	C	Cr	Ni	W	Ti	N	Ar	Ex.O	Y_2O_3
Wt%	0.16	11.52	0.3 4	1.44	0.28	0.00 7	0.006	0.144	0.36

As-received sample

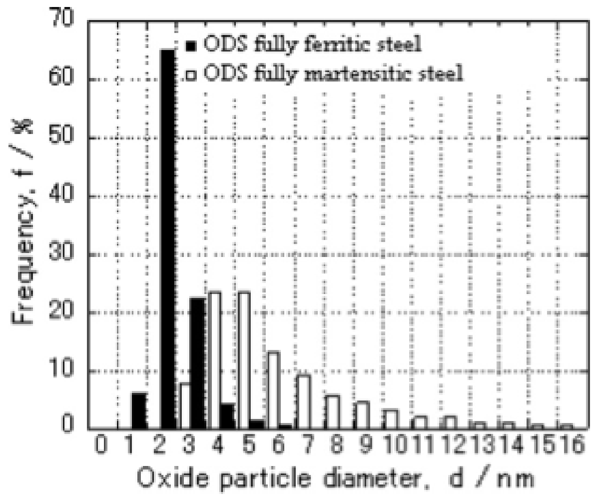


Ferrite

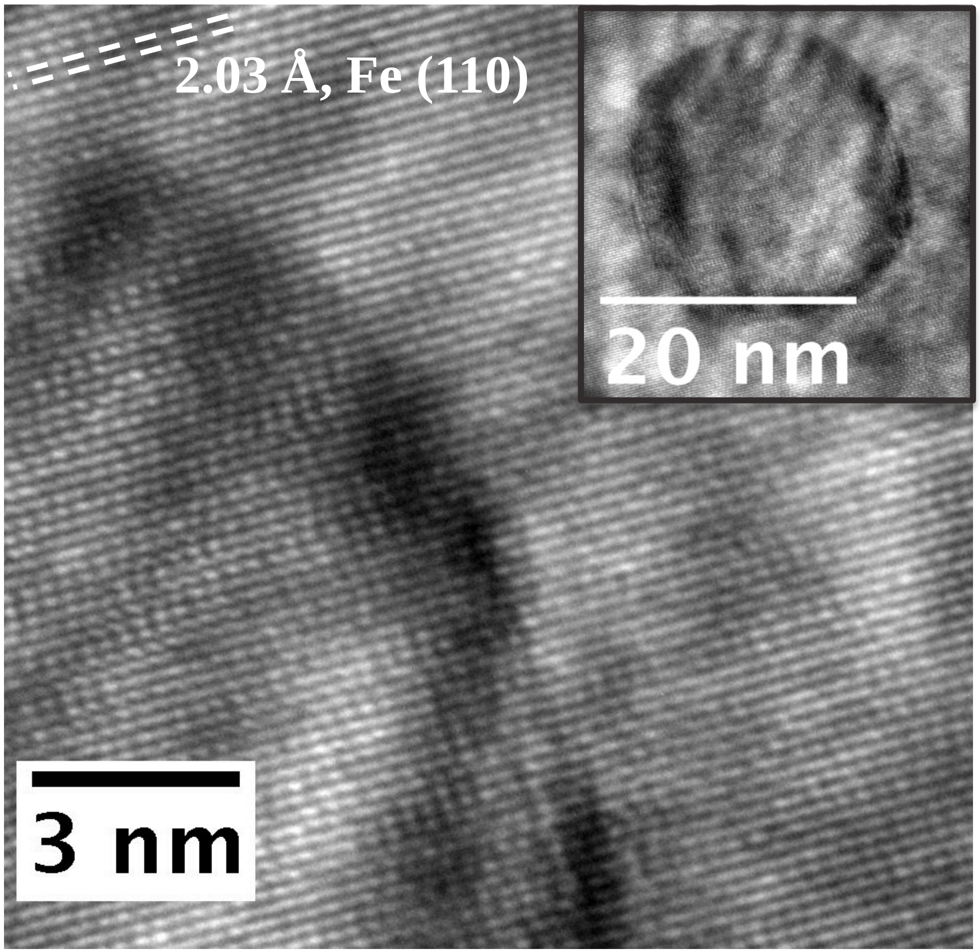
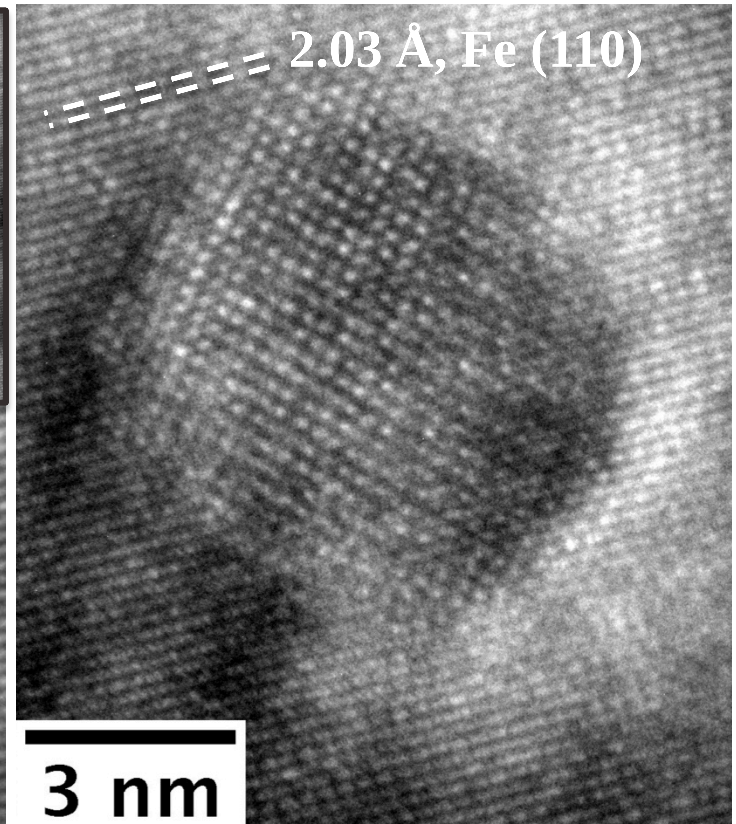


Martensite

Oxides are coarser in martensite than they are in ferrite.



[1] M. Yamamoto et al. JNM, 2011


2.03 Å, Fe (110)
20 nm
3 nm

2.03 Å, Fe (110)
3 nm

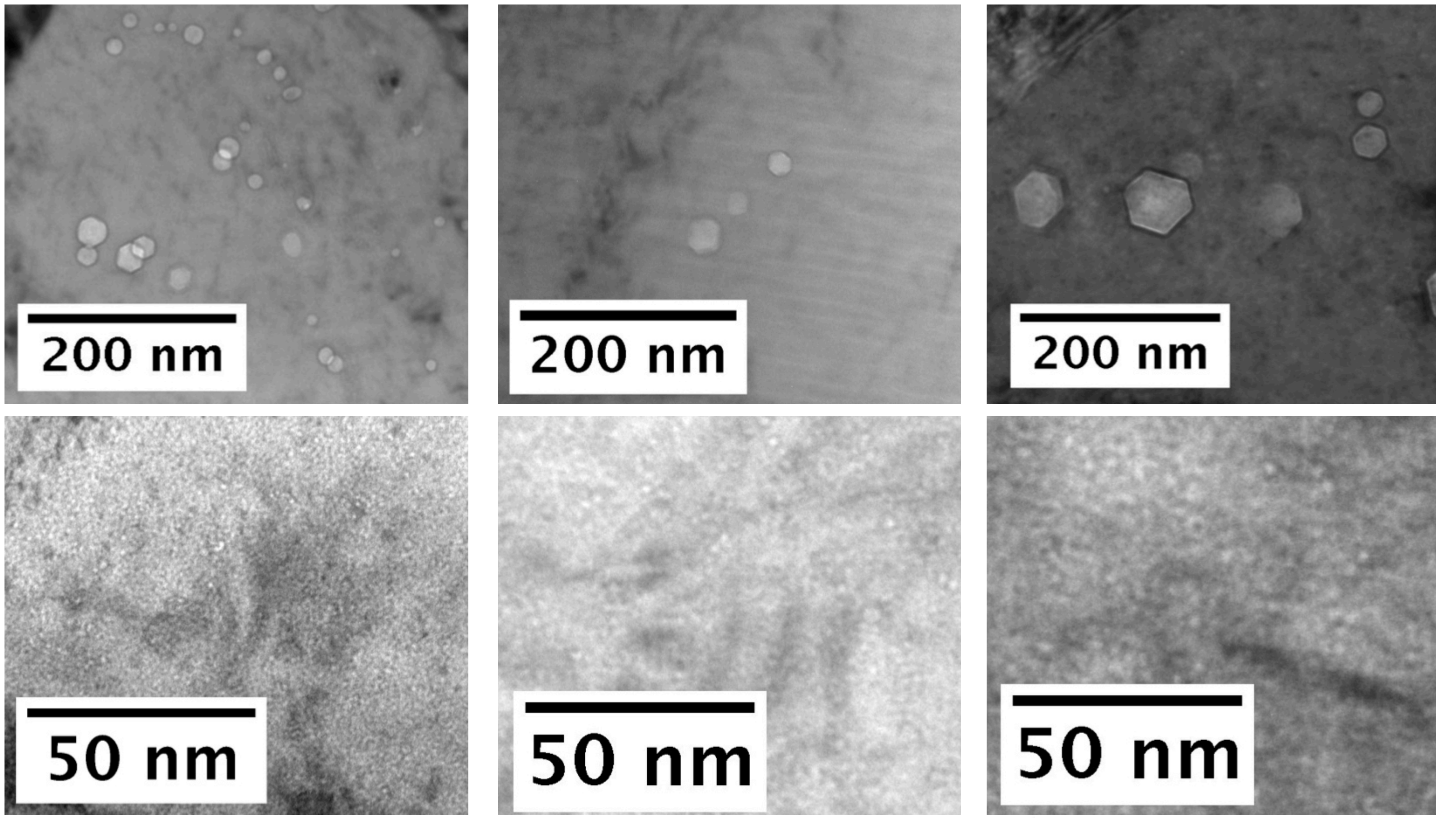
Swelling much smaller in martensite phase

Peak DPA: 100 | 300/400 | 600

Real DPA: ~50 | ~150/~200 | ~300

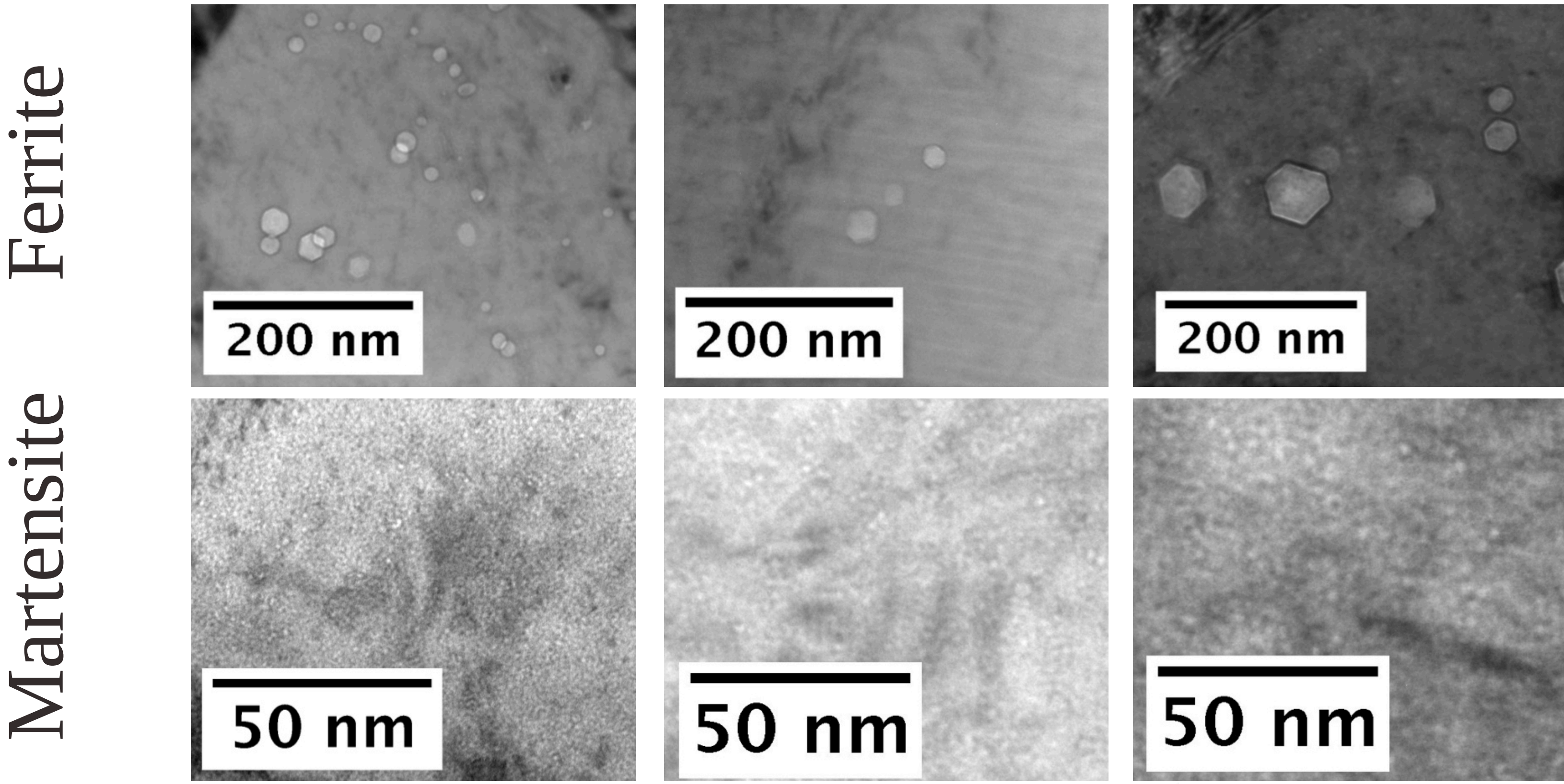
Ferrite

Martensite



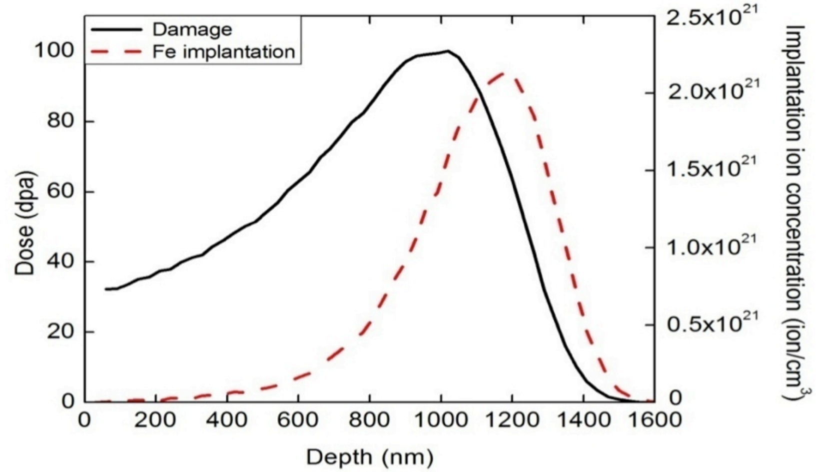
Swelling much smaller in martensite phase

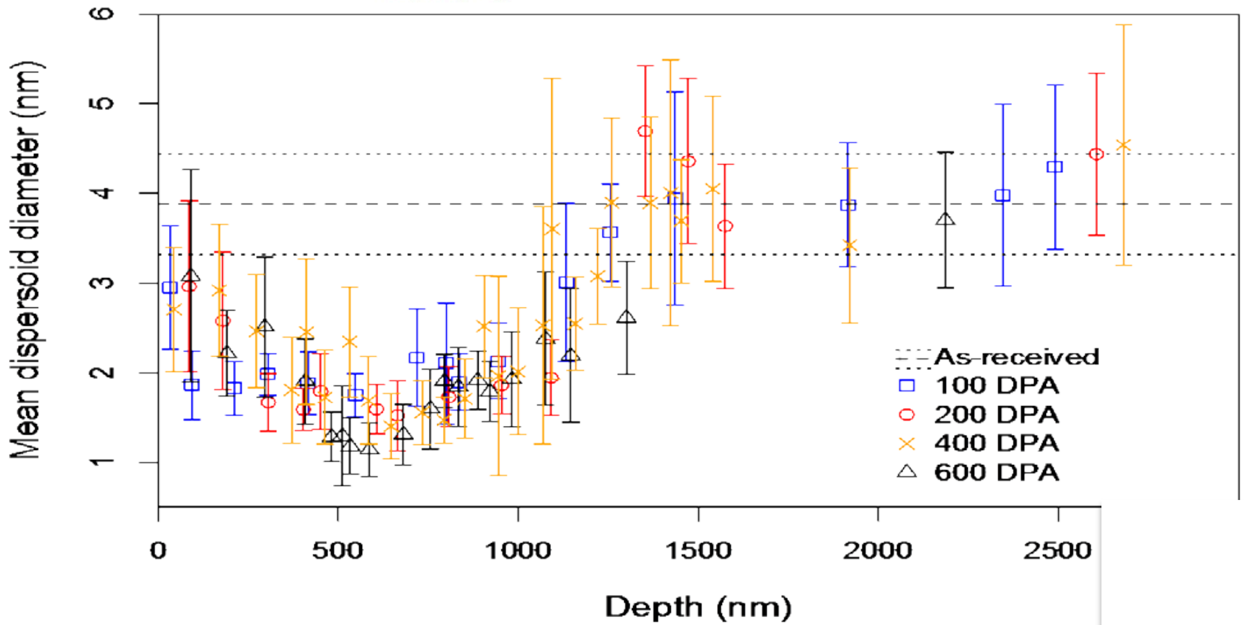
Peak DPA: 100 | 300/400 | 600
Real DPA: ~50 | ~150/~200 | ~300



No voids were found in the injected interstitial region.

Oxide particle size changes strongly with depth but not with dpa level

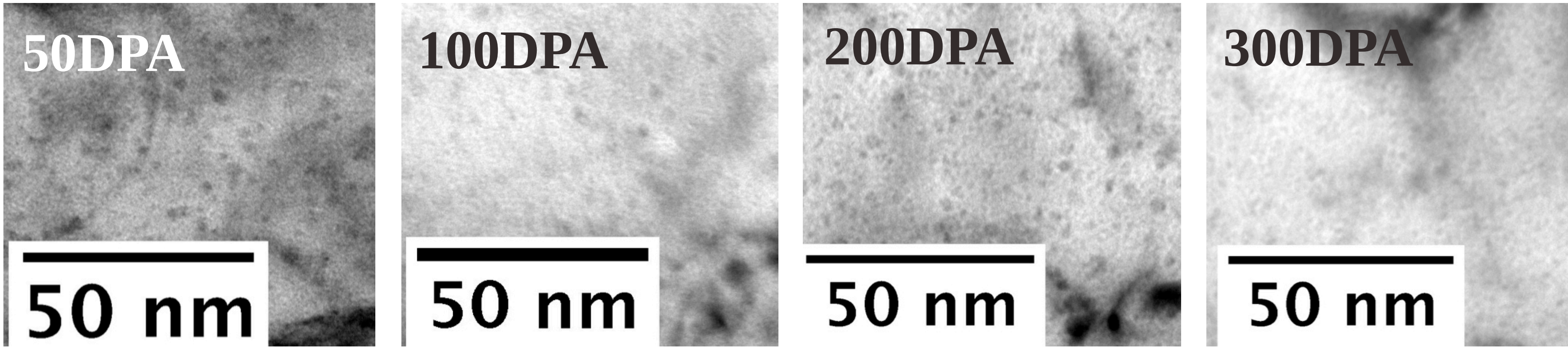


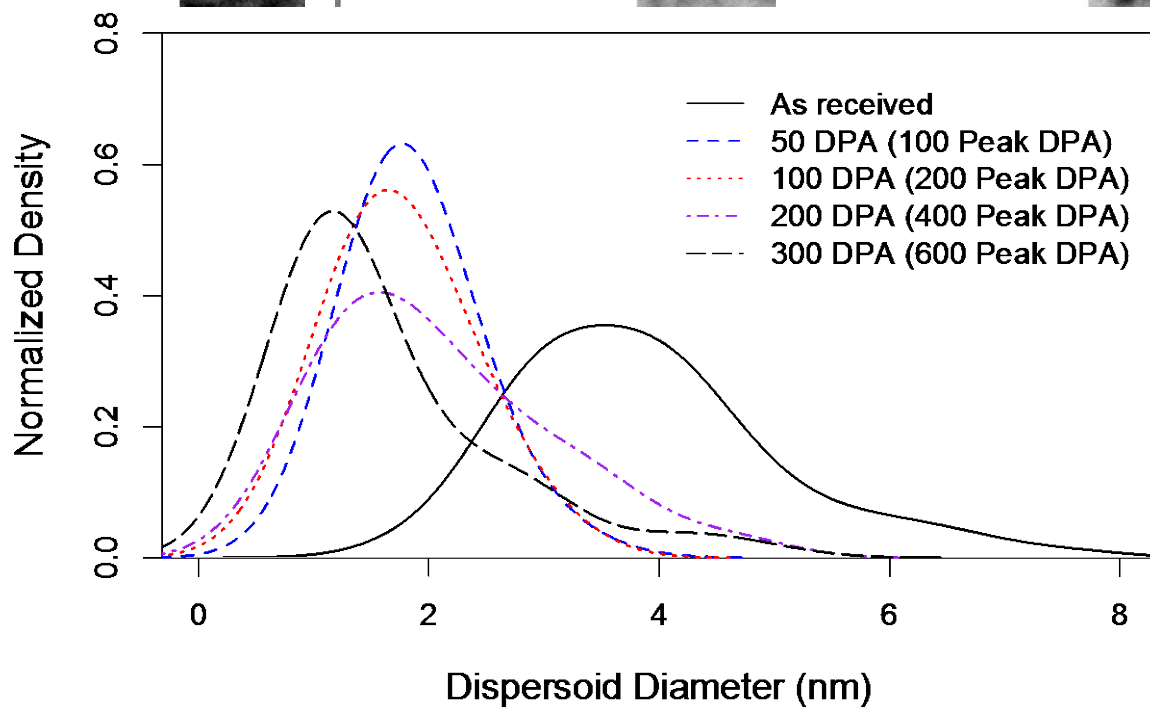


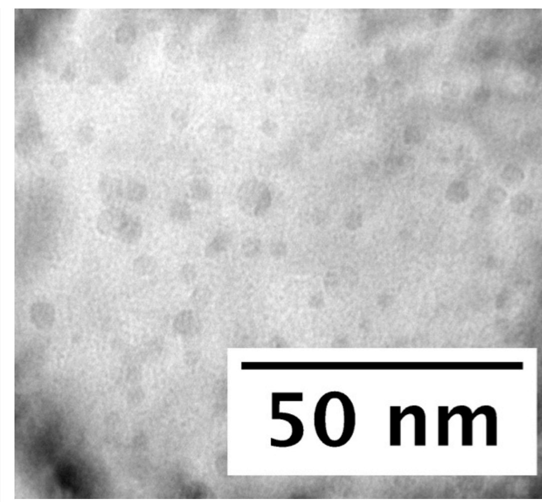
Top: SRIM simulation

Bottom: Dispersoid size distribution

Oxide particle size decreases with dose







Conclusions

- **Ion irradiation can yield excellent insight on void swelling and microstructural evolution.**
- **It cannot provide good predictions of void swelling in neutron environments.**
- **Proper use of ion bombardment requires an understanding of neutron-atypical aspects.**
- **It is important to use a non-rastered beam, to calculate the dpa correctly and to avoid the injected interstitial region.**