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Comments on ‘On the onset of void ordering in metals under neutron or heavy-ion irradiation’

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Abstract. In their recent paper on void ordering, Barashev and Golubov [1] describe cavity lattice formation in austenitic steel under electron irradiation as a bubble lattice rather than a void lattice. While this is convenient for the model they propose, this comment gives several reasons why the bubble lattice assignation cannot be justified. In addition, some remarks made by Barashev and Golubov on the results of 1-d Monte Carlo (MC) simulations [2], and the role of 1-d SIA clusters in creating the planar ordering found in experimental void lattice formation results [3], are critically discussed.

Barashev and Golubov [1] describe a model of void lattice formation based on the one dimension migration of self-interstitial atom (SIA) clusters emanating from collision cascades under neutron or heavy ion bombardment. To avoid the difficulty (to their model) of the observation of void ordering in 1 MeV electron irradiated nitrided austenitic stainless steel [4] they have repeated an interpretation of this observation by Trinkaus and Jäger [5] which suggested that the high concentration of nitrogen reported in the steel had led to the formation of a nitrogen bubble lattice, rather than a void lattice. The bubble lattice was supposedly formed by SIA loop punching from the bubbles. However, in considering the interpretation of Trinkaus and Jäger several points can be made:

- (1). The assignment of the ordered cavities as bubbles is most unlikely. There is no evidence that any excess nitrogen will form bubbles in steel; any excess nitrogen should precipitate as chromium nitride [6].
- (2). It was clear from the investigation that the electron irradiation conditions were carried out exactly in a regime where voids should be expected – as shown by void formation in identical steel but with normal nitrogen content [4].
- (3). The cavity concentration was of the order of $10^{23}/\text{m}^3$, two orders of magnitude lower than the $10^{25}/\text{m}^3$ concentration consistently found in inert gas bubble lattices [7].
- (4). Finally, it is hard to understand how loop punching from small bubbles along close packed directions could lead to the formation of ordering. The punching out of an interstitial loop along a glide cylinder and interacting with a bubble only partially lying on the same glide cylinder must push that bubble *away* from an ordering direction.

These points suggest that the interpretation of the electron irradiation result as nitrogen bubbles cannot be sustained. It is rather easier to believe that the ordering is a void lattice

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3 exactly as first proposed by Fisher and Williams. This observation, and that of void ordering
4 formed under electron irradiation in alpha-iron [8], show clearly that the void lattice formation
5 mechanism cannot wholly be attributed to the migration properties of cascade induced
6 interstitial clusters. A separate mechanism, independent of cascades, is required.
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10 In their remarks to support their model, Barashev and Golubov comment on the MC
11 simulations [2] which show that the influence of 1-d SIA defect movements on void populations
12 do not lead to a perfect void lattice. They suggest that the results of these investigations are
13 suspect since such simulations need to use simulation boxes bigger than the mean free path
14 (MFP) of the 1-d migrating defects. While at first sight this criticism might sound plausible, in
15 both the work of Heinisch and Singh [9], and of Evans [2], 1-d ordering was found when the
16 SIA mean free path was only a few times larger than the inter-void distance. It is then difficult
17 to see why results should be fundamentally different for larger mean free path values. In any
18 case, when impurities are present, the MFP values might be expected to be considerably shorter
19 [10]. The critical point is that the barrier for the precise alignment of voids – toward a perfect
20 lattice – is a short range effect, and can be predicted from a trivially simple diagram [2]. The
21 suggestion that it only occurs in the simulation due to a methodological error associated with the
22 mean free path and box size cannot therefore be justified.
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26 A further comment by Barashev and Golubov concerns the dimensionality of the *initial*
27 ordering, i.e. that one should expect it to follow the dimensionality of the 1-d or 2-d migrating
28 defect [3]. For many years this has been the received wisdom, but in a marked departure from
29 this, they state that according to their equations (16) and (17) based on the 1-d migrating SIA
30 cluster, voids form close packed planes rather than close packed lines during the early stages of
31 void ordering. The inference is that there would be no disagreement with the experimental
32 evidence for early planar ordering shown for molybdenum and the absence of linear alignments
33 in the required close packed directions [3], a result that suggested the migration of an interstitial
34 defect with a 2-d rather than a 1-d character. While it is recognised that at a later stage of
35 ordering, the linear ordering can interact and move toward a 3-d lattice [2], it is extremely
36 difficult in the early stages to see how the claimed planar ordering result can have any
37 mechanistic basis. Recalling the Foreman model, voids become aligned along close packed
38 directions as a result of 1-d SIA fluxes, and therefore only along these directions is it possible
39 for voids to interact and alter their positions. The mechanism is linear, not planar. If any planar
40 effect was somehow taking place, then the linear ordering would bizarrely have to disappear to
41 be consistent with the molybdenum results [3]. In the end it seems rather more logical to stay
42 with the older assumption that the dimensionality of the initial void ordering follows that of the
43 self-interstitial defect.
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Barashev and Golubov in a closing argument emphasise that substantial modelling and experimental evidence to date support 1-d rather than 2-d SIA transport. While there is little argument about the production of 1-d SIA clusters in cascades, there are serious difficulties as discussed above in extending their influence to void lattice formation. One might add further that although these clusters have provided a possible explanation for enhanced swelling occurring near grain boundaries in irradiated metals, there seem to be no reported observations of this result in samples of the bcc metals where void lattice formation is perhaps most clear. Thus it is not apparent that these phenomena always go together.

Returning to the requirement of a void lattice mechanism independent of cascades, such a mechanism would have an advantage if it could also apply to the bubble lattice results formed in helium ion irradiations where, as in the electron irradiation situation, no cascades are formed. In addition, besides allowing the formation of perfect, or near-perfect, void lattices, it should also explain the clear influence of impurities on void lattice formation as seen in the bcc metals Nb and Nb-1%Zr [11], Ta [12], and molybdenum [13] after ion irradiations, a result not addressed in the model put forward by Barashev and Golubov. The work describing the 2-d model [14], where for the bcc metals it was concluded that the di-interstitial could be the diffusing species, showed that all of these essential requirements can be met.

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JHE comments on the second version of the Barashev and Golubov Response

In their revised response, B&S have accepted my point (point 1) that there is no disagreement between us on the void lattice – saturation swelling topic. Since I did not discuss this area in my submitted Comment, I expected that the topic would be dropped in their revised response. However, not only is it still included, but it is introduced on the premise that swelling saturation “it is generally perceived to be a consequence and hence a property of the void lattice....”. Although this was correct in the past, this statement is now simply not true. As an example, I was one of the main proponents of this view, but as already discussed and agreed by B&S, this has been corrected and updated. In the end, I don’t believe this discussion of saturation swelling has any relevance in our exchanges; essentially it is now only a trailer for their forthcoming publication.

As an issue separate from my Comment and the Response, I can take this opportunity to address a query brought up by B&S in their covering letter under item 5. Regarding the nickel result that I quoted, I obtained this from my paper on 1-d SIA simulation where I said correctly that the void lattice had not been seen under neutron irradiation. I of course know of the Kulcinski work. As for vanadium, they are right to ask for a reference, but at the moment, I cannot help them. I originally made this statement in a draft report for Culham but annoyingly did not reference it. I cannot now find the basis for what I wrote but I can’t believe I invented it and will keep looking.