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A THEORY OF VOID LATTICES

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ABSTRACT

A theory of void lattices in metals is presented that is based on the two-interstitial model and makes use of no additional assumptions. Outstanding features of the void lattices, e.g., the fact that they possess the same structure and orientation as the host lattices, are shown to be intimately related to the existence of a one-dimensionally migrating metastable excited self-interstitial state (crowdion). Void ordering is shown to be an example of self-organization by means of a phase transition far from thermodynamic equilibrium in the sense of synergetics.

1. INTRODUCTION

One of the most spectacular phenomena in the radiation damage in crystalline solids is the alignment of voids into regular lattices during irradiation. This effect has been observed in metals (e.g., Ta, W, Nb, Ni, Al, Mg) [1], their alloys, and fluorites (e.g., CaF_2) [2].

Void-lattice formation occurs in four stages: (A) the initial formation and growth of small, *randomly* distributed voids, (B) the growth of the larger voids at the expense of smaller voids, (C) the appearance of small locally *ordered* regions, (D) the spread of these regions to other areas. In most cases, it was observed that a high density of void nuclei facilitates ordering.

The most important characteristics of void lattices are : (a) Their structure and crystallographic orientation are the same as those of the host lattices. (b) Void growth saturates in a void lattice. The ratio of the void spacing to the average saturated void radius is nearly independent of temperature and other irradiation conditions. (c) The size of the voids in a void lattice is uniform. (d) The void lattice structure is in general stable against irradiation and thermal annealing. (e) Alignment within a void lattice is not generally improved by post-irradiation annealing. (f) Though void lattices form over a wide range of temperatures, their perfection and ease of formation (measured by the threshold dose) is best at temperatures just below the temperature of maximum swelling. (g) Gas impurities appear to have a large effect on ordering. In electron irradiations, a minimum gas concentration is a prerequisite. (h) The void lattice is usually imperfect. Displacements by about 10% from the perfect lattice sites, edge dislocations, multiple site occupancies, and vacant sites are often seen. "Interstitial" voids, however, have not been observed. The perfection of the lattice in general increases with dose.

Many theories have been proposed to explain the formation of void lattices [3]. Their relative merits have been discussed by various authors [1-3]. Noticing that under irradiation the crowdion concentration may be much higher than its thermal-equilibrium value, we recently have investigated quantitatively the rôle played by crowdions during void swelling with particular emphasis on the effect of ordering. Based on the so-called two-interstitial model, a successful theory of void lattices in metals and alloys has been formulated that explains all the essential features of void lattices. While the original papers [4-6] develop the mathematical theory, the present paper summarizes the results with emphasis on the physical concepts.

2. THEORY

2.1. *Two-interstitial model.* The two-interstitial model of point defects in metals [7-8] on which the theory is based without any additional assumptions postulates that the self-interstitials may occupy two energetically different states. In fcc (bcc) metals the self-interstitial groundstate is the <100> (<110>) dumbbell configuration [9-11]. In the excited metastable state the self-interstitials possess the <110> (<111>) crowdion configuration. Conversion from the dumbbell to the crowdion configuration, or vice versa, may occur by thermal activation. Hence, the crowdions possess a mean lifetime τ_C which is the average time they spend between their creation and their conversion to the dumbbell configuration. τ_C is related to a corresponding crowdion mean free path (MFP) L_C via $L_C^2 = 2D_C\tau_C$, where D_C is the crowdion diffusivity. Analogous quantities may be defined for the dumbbells.

2.2. *Diffusional anisotropy difference and crowdion-supply-cylinder concept.* In its application to void lattices the most important feature of the two-interstitial model is the one-dimensional migration of the crowdions along close-packed crystallographic directions, which is in marked contrast to the three-dimensional diffusion of the dumbbell interstitials and the vacancies. This diffusional anisotropy difference (DAD) [12] between the crowdions and vacancies provides the source for ordering via a Darwinian selection among randomly distributed voids. The result is the formation of void lattices having the same symmetry and crystallographic orientation as the host lattices. This outstanding feature of the void lattices, which has been observed in both fcc and bcc metals without exception [1], has not been explained by any other void-lattice theory not involving crowdions.

The significance of the DAD between the crowdions and the vacancies in the formation of void lattices is best visualized in terms of the crowdion-supply cylinders (CSCs). Most of the crowdions annihilating at a void must have been produced by displacement events or conversions of dumbbells within cylinders of length L_C extending from the void surface along the close-packed directions. The radius of each CSC is equal to the radius of the void it belongs to. In a bcc (fcc) lattice, there are eight (twelve) of these CSCs for each void. When the separation of two voids along a close-packed direction is sufficiently small,

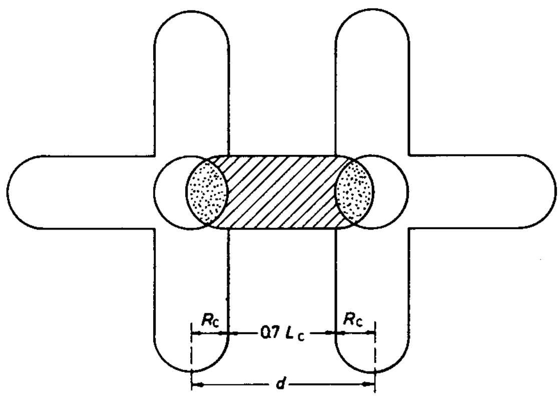

Fig. 1. Neighbouring voids at a distance $d = 2R_C + 0.7\ L_C$. The crowdion-supply cylinders overlap (shaded region) and penetrate into the neighbouring voids (dotted region).

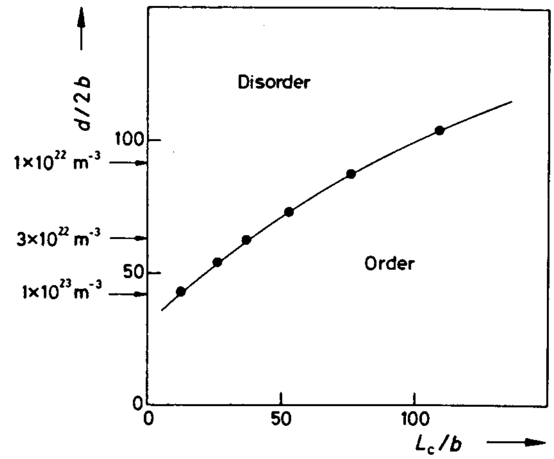

Fig. 2. Computed d—L_C non-equilibrium phase diagram showing the regions of void order and disorder (b = nearest-neighbour distance of the host-lattice atoms). The arrows at the ordinate mark some void-number-density values.

their CSCs overlap or even protrude into the other void (Fig. 1). In this situation of "connected" voids a reduction in the crowdion supply to both voids leads to an increase in the net vacancy fluxes into these voids and thus to their enhanced growth. As "connected" voids increase in size, the vacancy supersaturation decreases, causing a reduction in the net vacancy flux to the smaller, "unconnected" voids. The final result is a preferential growth of voids arranged in a lattice of the same structure and orientation as the atomic host lattice — since for such voids all the CSCs are "connected" — and a simultaneous shrinking away of the other "unconnected" voids.

2.3. *Extended-rate-theory treatment.* The CSC concept shows that void-lattice formation can occur only if the void-number density is high enough, so that the CSCs of neighbouring voids may overlap. Hence, in a mathematical formulation of the void-lattice formation as a problem in self-organization [13], the void-number density or the average void distance d plays the rôle of a control parameter, and disordered void arrangements or void lattices are deducible as solutions for large or small d values, respectively. This has been demonstrated within the framework of an extended rate theory that accounts for spatial fluctuations of the void-number density. The theory predicts that a non-equilibrium phase transition takes place when the radius R of the voids attain a critical value

$$R_C = (d-0.7\ L_C)/2\ . \tag{1}$$

This phase transition is interpreted as the transition from a disordered void arrangement to a void lattice (Fig. 2), since — as illustrated in Fig. 1 — Eq. (1) represents the condition for the overlap of the CSCs of neighbouring voids and the onset of CSC—void protrusions. Eq. (1) can only be fulfilled in regions with a high void-number density. There R_C is small enough to be reached before void-size saturation in the disordered state has occurred.

2.4. *Mechanism of void-lattice formation and void-lattice defects.* When the ordering condition (1) is satisfied, presumably first in isolated regions with a higher-than-average void density, void ordering takes place. Since voids grow if their CSCs are "connected" in the close-packed directions and shrink otherwise, it is clear that the resulting void lattice must have the same structure and orientation as the host lattice. Hence, the present theory explains the most important characteristics of void lattices in a natural way.

Once started, ordering spreads by the growth of ordered islands. When different islands meet, edge dislocations or low-angle grain boundaries are created in the void lattice to accommodate the misfits. The inability of voids in the dislocation cores to join up with all nearest neighbours along close-packed directions necessarily leads to their smaller size [14]. "Interstitial" voids can*not* join up with *any* nearest neighbours along the close-packed directions and therefore cannot exist [15]. Because of the statistical fluctuation of the initial void distribution in the starting islands, void-lattice "vacancies" and fluctuations in the void-lattice parameter are expected, and indeed observed [1,14]. In the present theory, void ordering is a dynamic effect that occurs only under irradiation. Alignment is expected to improve in general by prolongued irradiation, but *not* by post-irradiation annealing.

3. FURTHER PREDICTIONS AND COMPARISON WITH EXPERIMENTS

3.1. *Pre-ordering regime.* The existence of a critical void radius R_C [see (1)] divides the evolution of voids into a pre-ordering regime ($R < R_C$), in which voids grow in random arrangement, and an ordering regime ($r \geqslant R_C$), in which a regular void lattice may develop. The extent of the pre-ordering regime decreases with increasing crowdion MFP, increasing void-number density, and increasing void-growth rate. The crowdion MFP varies from element to element and increases with decreasing impurity content and decreasing temperature. The void-number density depends on the type of irradiation, the alloying elements, and in general increases with increasing gas impurity content and decreasing temperature. The void-growth rate varies with dose rate and temperature and depends on the particular element. From this one may deduce that the extent of the pre-ordering regime may considerably depend on the particular element, its impurity content, the irradiation temperature, and the irradiating particles.

The preceding predictions match the experimental observations well. Void growth in a random distribution is always observed before the onset of ordering [1,3]. The most effective way of producing ordering is to start with a high concentration of nucleii [3]. Gas impurities have a large effect on ordering; in many cases, a minimum gas concentration is a prerequisite [1,16,17]. This is particularly true in the case of electron irradiation because of the lack of displacement cascades that can act as nucleation centers [1]. E.g., in electron-irradiated 20/25 stainless steel, void ordering requires a high concentration of nitrogen [17]. The ordering doses vary from element to element. In Nb [16,18] void lattices have been observed at a dose as low as 5 dpa, while in Ni [19] they are not observable up to 400 dpa. The odering dose also varies with the alloy content. Alloying Ni with Al [20] increases the void-number density almost tenfold and reduces the ordering dose from 400 dpa to 20 dpa. However, as the Al content is increased, the void lattice becomes increasingly more imperfect, presumably due to the reduced effective crowdion MFP. It is interesting to note that the void-number density in ion-irradiated pure Ni [20] increases with dose and at 400 dpa, when the void lattice is observed, has a void-number density roughly equal to that of the Ni—Al alloy. Comparison of the mean spacings between ordered and disordered voids in Nb at different temperatures (Fig. 3 in [1]) clearly shows that the disordered voids are much more separated than the ordered ones, as predicted by the present theory.

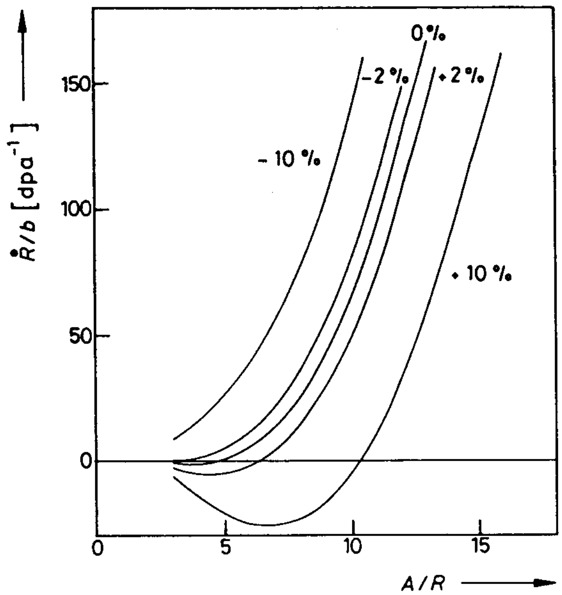

Fig. 3. Computed growth rates $\dot{R}$ of individual voids in a bcc void lattice as a function of A/R. The radii of these voids deviate from the radius R of all other voids by 0%, ± 2%, and ± 10%, respectively.

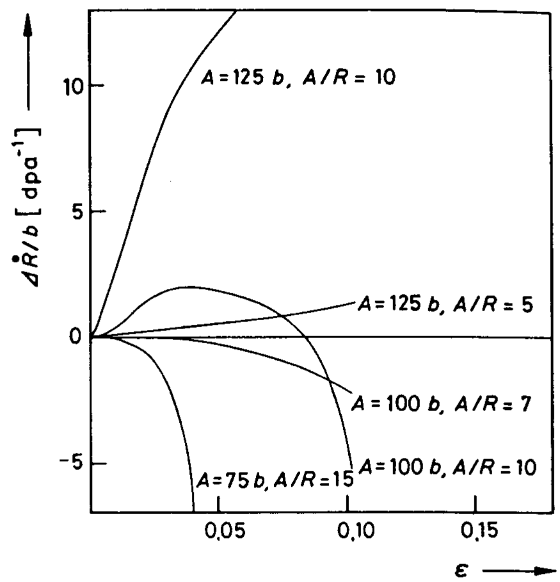

Fig. 4. (In-)stability of bcc void lattices against a relative void displacement ϵ, computed for various pairs of A, R values. For $\Delta\dot{R} > 0$ the lattices are stable and otherwise unstable.

3.2. *Limited growth, size uniformity, and displacive stability of voids in a void lattice.* The present theory [5] predicts that, *as a consequence of the one-dimensionality of the crowdion migration*, (α) smaller voids in a void lattice grow faster than larger ones (Fig. 3), so that eventually all voids have the same size, that (β) the stability or the instability of a void lattice against the displacement of voids from their perfect lattice sites depends on both the void-lattice spacing A and the ratio A/R (Fig. 4), and that (γ) in stable void lattices the growth of voids saturates. The calculated void-spacing/saturated-void-radius ratios [5] in Mo, Nb, and Ni are in good agreement with experimental data [16,18 -22]. Except for Mo, for which the occurrence of void-lattice instability at a very high void-number density is forecast, void lattices are predicted to be stable in all metals in which they have been observed [5]. The patchy void distribution to which the uniform void lattice in neutron-irradiated Mo changes when the irradiation dose reaches 8×10^{26} m^{-2} [23] is an example of the onset of displacive void-lattice instability [5].

3.3. *Temperature dependence of void swelling and ordering.* At first sight it is surprising that void ordering is most pronounced in a temperature regime below the temperature at which swelling passes a maximum [1]. It is a merit of the present theory that it can account for this observation [6]. Here we mention only that this phenomenon is related to the counteraction of the different temperature dependences of the mean free paths of dumbbells and crowdions in the temperature interval of interest. More details and a comparison to observations on Ni and Mo are given elsewhere [6].

4. CONCLUSIONS

In the present paper, we have introduced a quantitative theory of void lattices based on the two-interstitial model of point defects. It has been shown that this theory is capable of explaining, in a simple and straightforward way, all the essential features of void lattices, namely (i) their formation as a disorder–order phase transition in systems maintained far from equilibrium by particle irradiation, (ii) the coincidence of their crystallographic structure and orientation with those of the host lattices, (iii) the saturation of void growth, (iv) the uniformity of void size, (v) their stability or instability, and (vi) the influence of temperature on their occurrence. These phenomena are natural consequences of a rate theory of swelling that differs from the conventional one only in that the existence of a metastable, one-dimensionally migrating configuration of the self-interstitial has been assumed. In addition, many other less spectacular observations than (i) through (vi) also find satisfactory explanations within the present theory. In our opinion, the success of this theory questions the general applicability of the conventional void-swelling theory, which does not consider the one-dimensionally migrating, excited state of the self-interstitials, and strongly supports the two-interstitial model of point defects in metals. According to the present theory, void ordering is an example of self-organization far from thermodynamic equilibrium.

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