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Non-linear dynamics of microstructure evolution and hyper void-lattice formation

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ABSTRACT

Irradiation damage accumulation in metals is studied via a dynamical description of the evolution of a system of interacting crystal defects, focusing on the complex behavior caused by system instability and symmetry-breaking. The case of a supercritical void ensemble in a temperature range where void growth is significantly affected by vacancy emission is specifically considered. Conditions of instability are found in the growth dynamics of the void system, the resulting bifurcation of which causes the shrinkage of some voids and the growth of others, resulting in coarsening of the ensemble. The presence of a small amount of one-dimensionally migrating self-interstitials with mean-free path comparable to the average distance between voids can bias the void coarsening process, such that the non-aligned voids have a much larger probability to shrink than the aligned ones. The post-bifurcation evolution leaves voids aligned along the crystallographic directions to form an imperfect lattice with empty lattice sites eventually filled by preferred nucleation. For this process to occur the irradiation temperatures must be higher than 0.4 of the melting temperature. The typically low number densities of voids at these temperatures necessarily entail a void lattice parameter much larger than when vacancy emission is negligible. The implication of the formation of the hyper void-lattice, an appellation adopted from earlier studies, on properties of one-dimensionally migrating self-interstitials is also discussed.

1. Introduction

Microstructure development during particle irradiation often results in nano-scale ordered structures. Particle irradiation typically produces regions of displacement damage at a rate of 10^{12} collision events per second per cm^3. When the damaged region cools down, crystal defects are 'quenched in'. As irradiation proceeds, crystal defects accumulate and interact, and the microstructure evolves under non-linear driving forces. The evolution of the accumulating defects is conventionally described by coupled rate equations, analogous to chemical processes. The complexity of the dynamics of such coupled nonlinear systems is well known [1]. However, to maintain the manageability of the calculation, a simplifying mean-field approximation is usually adopted, in which the spatial and size distributions of the sinks and the mobile-defect concentrations are averaged out. Despite the apparent simplicity of the equations, the corresponding dynamical behavior is not necessarily simple. Complexity due to dynamical instabilities and bifurcations gives rise to phase-change like behavior of the system [2–4]. This issue is even more pronounced when the spatial and size distributions [5–9] of the reaction partners are explicitly taken into account. Thus, when the long-wave-length solution becomes unstable, dominance of the shorter wave length leads to spatial ordering [5–8]. Instability in the size distribution function leads to void coarsening when the stochastic nature of the irradiation damage and defect accumulation processes are taken into account [9]. The presence at the instability point of a small bias due to one-dimensionally migrating self-interstitials with mean-free path comparable to the average distance between voids influences the void coarsening process such that shrinkage predominantly occurs with the non-aligned voids, and growth with the aligned ones. The post-bifurcation evolution then leaves voids aligned along the crystallographic directions to form an imperfect lattice with empty lattice sites eventually filled by preferred nucleation.

Indeed, the crystallographic structure and the orientation of void lattices in irradiated metals are well known to follow those of the host lattices [5]. For this reason, self-interstitial atoms (SIAs) moving one-dimensionally along the close-packed crystallographic directions have been a prime factor in many studies of void-lattice formation [2–4, 6–11]. Yet, the detail mechanism is still controversial. Recent Monte Carlo simulation [12] suggests that the 1-D self-interstitial transport of either the crowdions or small interstitial clusters may bring about coalescence between neighboring voids along the crystallographic directions. Coalescence must not be overwhelming for void-lattice formation to be feasible. In another aspect, for a void lattice to form from a randomly distributed ensemble of supercritical voids [5,13], non-aligned supercritical voids outside lattice positions have to disappear. As shown in [9], 1-D interstitial diffusion by itself does not cause the non-aligned

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supercritical voids to shrink away. It only leads asymptotically to an ensemble in which smaller non-aligned voids coexist with larger aligned ones.

In some cubic metals such as vanadium and aluminum, molecular dynamics simulations show that intracascade clustering is an event of low probability [14,15]. Available experimental data on void swelling in aluminum is also completely explicable in terms of the concept of standard rate theory based on the dislocation bias [16–19]. Production bias [20] and initial defect clustering in cascades do not seem to play an important role. This indicates that the actual fraction of long-range 1-D moving interstitials needed for void-lattice formation may not have to be significant.

Experiments find that void ordering is usually preceded by the coarsening of randomly distributed supercritical voids in an ensemble, resulting in the growth of the larger voids and the shrinkage of the smaller ones [5,13]. In this regard, one may note that void coarsening may occur due to the dynamic instability of the void-size distribution, caused either by the usual vacancy emission, or as a stochastic effect due to the fluctuation of point defect fluxes received by the voids [9,21–23]. The former occurs because of the instability caused by the positive feedback effect due to the increasing vacancy emission from a shrinking void. In the latter case, when voids shrink under fluctuating point-defect fluxes there is also a positive feed-back action because the probability of dissolution of the supercritical voids increases as the voids shrink [9,21–23]. That void dissolution does happen under this condition has been explicitly demonstrated both analytically via the solution of the Fokker–Planck equation [23], and numerically via the solution of the time-dependent master equation [22].

In a previous paper [9] we showed that when the average net vacancy flux received by the voids became sufficiently low, dynamic instability of the void-size distribution might occur, and a fraction ε_i of self-interstitials as small as ~1% moving one-dimensionally was enough to instigate stochastic dissolution of the non-aligned voids. The critical condition was typically satisfied when the swelling rate $\dot{S}$ dropped below 0.1%/NRT dpa. We also showed that the aligned voids were more resistant to shrinkage than the non-aligned ones because of their larger growth rate. The elimination of non-aligned voids would feed the growth of the aligned ones and at the same time created a partial void lattice with many empty lattice sites where the 'shadowing' [2,3] of neighboring voids produced local depressions of 1-D self-interstitial fluxes. With a void nucleation probability that increases exponentially with the net vacancy flux [23], practically nucleation of all new voids occurs in these SIA-deficient locations [9]. The end result of this development is that only the aligned voids survive and multiply to form a void lattice, as the winning species of the Darwinian competition [9]. We note that in this process, the most important role of the 1-D moving self-interstitials is as a bias favoring the nucleation and survival of the aligned voids during the coarsening process. Due to the highly selective void nucleation sites, void coalescence due to the 1-D self-interstitial transport is unimportant as discussed in [9,24].

Void dissolution due to stochastic fluctuations may occur only when both the void growth rate and the average void radius are sufficiently small [9,21–23]. This condition is satisfied in most cases where void-lattice formation is observed [5,25–27]. Exceptions are found in the case of the so called hyper void lattices, such as in neutron irradiated aluminum, where lattices with very large lattice parameter (200–250 nm) are formed from voids as large as 60–90 nm in diameter, undergoing healthy growth (swelling rate ~0.5%/NRT dpa) [28,29]. Obviously, stochastic fluctuations can hardly have any effect on the evolution of voids of this size.

As mentioned in the foregoing, dynamic instability of the void-size distribution due to the positive feed-back of vacancy emission from a shrinking void may also be reflected in the coarsening of a void ensemble as in Ostwald ripening. Indeed, it has been shown analytically that the dynamics of evolution of a spatially homogeneous distribution of voids is only conditionally stable at a temperature where vacancy emission from the voids is important [30,31]. Since the aligned voids 'shield' each other against 1-D moving SIAs, they receive a reduced flux of self-interstitials [2,3]. As a result, the aligned voids will be larger in general than the non-aligned ones, and hence have lower vacancy emission rates. This suggests that non-aligned voids will dissolve in preference to the aligned ones during coarsening. It is our aim in this paper to examine in greater detail the possible dynamic instability due to vacancy emission as a mechanism for the dissolution of non-aligned voids during void-lattice formation. We shall adopt an analytical approach, and the calculated results will be discussed in comparison with the available experimental data on void hyperlattices.

2. Dynamic stability of the size distribution of a void ensemble

We consider the evolution of a void ensemble, in which the radius of the mth void is denoted by R_m. Taking into account vacancy emission from the voids, the boundary conditions on the void surface is given by,

$$C_v(\mathbf{r})|_{|\mathbf{r}-\mathbf{r}_m|=R_m} = C_s(R_m) = C_\infty \exp(2\gamma_s \Omega / kTR_m). \tag{1}$$

Here $C_s(R_m)$ is the equilibrium concentration of vacancies on its surface at an absolute temperature T, C_∞ is the equilibrium vacancy concentration at T far from any sink, γ_s is the surface tension coefficient, Ω is the atomic volume and k is the Boltzman constant. In Eq. (1) we neglect the gas pressure, and assume that the dominant stress on the void is due to the surface tension. For self-interstitials we assume the zero-boundary conditions on the void surfaces.

Let us first consider the major component of point-defects for which the mobility is three-dimensional (3-D) and assumed isotropic for simplicity. In the space between the voids, their local steady-state concentrations satisfy the conservation equations:

$$G_j + D_j \nabla^2 C_j - D_j Z_j \rho_d (C_j - C_{j\infty}) = 0, \tag{2}$$

where G_j ($j = i, v$) is the effective production rate of point defects, D_j and $C_j(\mathbf{r})$ are the 3-D diffusion coefficient and the concentration of point defects at the location $\mathbf{r}$, respectively, ρ_d is the total dislocation density, Z_j is the reaction constant between dislocations and three-dimensionally moving point defects, and $C_{j\infty}$ is the equilibrium concentration of point defects. Since $C_{i\infty} << C_{v\infty}$, in the following we put $C_{i\infty} = 0$, and $C_{v\infty} = C_\infty$. For simplicity we also assume $Z_v = 1$, and $Z_i = Z$.

Following [31], the solution of Eq. (2) can be written as

$$D_j C_j(\mathbf{r}) = D_j C_j^0 + \sum\nolimits_m \frac{W_{jm}}{|\mathbf{r} - \mathbf{r}_m|} \exp(-\sqrt{Z_j \rho_d}|\mathbf{r} - \mathbf{r}_m|), \tag{3}$$

where C_j^0 is the homogeneous solution of Eq. (2), and W_{jm} are constants determined by the boundary conditions. The summation in Eq. (3) is taken over all voids in the ensemble, which are assumed to be randomly distributed in space with a number density N.

We now consider the dynamical stability of an ensemble of voids characterized by a radius R. Suppose the mth void is subjected to an infinitesimal perturbation and its radius becomes $R_m = R + \delta R_m$. Then, when both $RN^{1/3}$ and $R\rho_d{}^{1/2} << 1$, within the first order approximation we have

$$W_{jm} = W_j + \tilde{W}_j \frac{\delta R_m}{R}, \tag{4}$$

with

$$W_i = \tilde{W}_i = -\frac{RG_i}{(4\pi NR + Z\rho_d)}, \tag{5}$$

$$W_v = -\frac{R}{(4\pi NR + \rho_d)}[G_v - D_v\rho_d(C_s(R) - C_\infty)], \tag{6}$$

$$\tilde{W}_v = W_v - \frac{2D_vC_s(R)\gamma_s\Omega}{RkT}. \tag{7}$$

The corresponding void growth rate is given by

$$\frac{4\pi R_m^2}{\Omega}\frac{dR_m}{dt} = \frac{1}{\Omega}\int_{S_m} dS_m \mathbf{n}_m(D_v\nabla C_v - D_i\nabla C_i) = \frac{4\pi}{\Omega}(W_{im} - W_{vm}), \tag{8}$$

where $\mathbf{n}_m$ is the unit vector of the external normal to the surface S_m of the mth void. Using Eqs. (4)–(7) can be written as

$$R_m^2\frac{dR_m}{dt} = \frac{[G_v - D_v\rho_d(C_s(R) - C_\infty)]R_m}{(4\pi NR + \rho_d)} - \frac{R_mG_i}{(4\pi NR + Z\rho_d)} + \frac{2D_vC_s(R)\gamma_s\Omega}{R^2kT}\delta R_m, \tag{9}$$

or

$$R_m^2\frac{dR_m}{dt} = \frac{\varepsilon_iGR_m}{(4\pi NR + \rho_d)} + \frac{(Z-1)\rho_d}{(4\pi NR + \rho_d)^2}\{[(1-\varepsilon_i)G - G_0]R_m + 2\tilde{G}\delta R_m\}, \tag{10}$$

if we assume $(Z-1) << 1$. Here $G = G_v$, ε_i is the fraction of self-interstitials that do not undergo conventional 3-D migration, i.e., $G_i = (1-\varepsilon_i)G$, and

$$G_0 = \frac{D_v(4\pi NR + \rho_d)(C_s(R) - C_\infty)}{(Z-1)}, \tag{11}$$

$$\tilde{G} = \frac{(4\pi NR + \rho_d)^2}{(Z-1)\rho_d}\frac{D_vC_s(R)\gamma_s\Omega}{kTR^2}. \tag{12}$$

Here G_0 and $\tilde{G}$ are both temperature-dependent quantities related to the vacancy emission rate. Note that Eq. (9) can also be derived from the conventional rate equations. Indeed, the right-hand side of this equation can be written as $R_m[D_vC_v - D_iC_i - D_vC_s(R + \delta R_m)]$, where $\delta R_m << R$, and the point-defect fluxes D_jC_j are determined by the conventional rate equations. The relative importance of the last term of Eq. (9) can be seen by writing the first two terms as $R_m(dS/dt)/k_c^2$, where $k_c^2 = 4\pi NR$ is the void sink strength, and dS/dt is the void swelling rate.

Keeping only terms linear in δR_m, Eq. (10) can be written as

$$R_m^2\frac{d(\delta R_m/R)}{dt} = \frac{R_m^2}{R}\frac{dR_m}{dt} - R_m\frac{dR}{dt} - 2\delta R_m\frac{dR}{dt} = \frac{2(Z-1)\rho_d}{(4\pi NR + \rho_d)^2} \times \left\{\tilde{G} + G_0 - (1-\varepsilon_i)G - \frac{\varepsilon_iG(4\pi NR + \rho_d)}{(Z-1)\rho_d}\right\}\left(\frac{\delta R_m}{R}\right). \tag{13}$$

We assume that all voids are initially supercritical, such that the void number density satisfies $G > G_0$. Let us consider, for illustrative purposes, the simplest case in which free vacancies and free interstitials are produced in equal numbers, i.e., $\varepsilon_i = 0$. Then for $G > \tilde{G} + G_0$, the perturbation δR_m in Eq. (13) decays exponentially with time, and the evolution of the void ensemble is dynamically stable. On the other hand, when $G < \tilde{G} + G_0$, the positive feedback effect of vacancy emission dominates and any perturbation δR_m in the void size distribution grows exponentially, no matter how small it is initially. In other words, the void ensemble becomes dynamically unstable. As a result, some of the initially supercritical voids may turn subcritical and shrink away, while others will continue to grow.

Similar to conventional void coarsening, the effective net vacancy flux decreases with dose as the void sink strength increases, as reflected by the term $(Z-1)\rho_d/(4\pi NR + \rho_d)^2$ in Eqs. (10), (12). As a consequence, the smallest supercritical voids may become subcritical. The difference is that the broadened void-size distribution here is not due to their initial variations, but is characteristic of the dynamics of the evolution of the void-size distribution function. Note that for sufficiently large voids in the initial distribution, i.e., $R > 2\Omega\gamma_s/kT$, $\tilde{G}$ is an increasing function of R. Accordingly, we may define the critical defect generation rate $K_{cr} = (\tilde{G} + G_0)/\varepsilon_c$ below which the homogeneous spatial distribution is dynamically unstable. Here ε_c is the fraction of point defects surviving the intra-cascade recombination.

From Eqs. (11) and (12), it is clear that the self-diffusion energy is the governing parameter responsible for the strong temperature dependence of K_{cr} Therefore, in Fig. 1 we plot K_{cr} in NRT dpa/s [32] as a function of temperature for Al, Nb and Mo only for void densities for which vacancy emission is significant. At the same time, we also note that K_{cr} increases with the void-number density which is a decreasing function of temperature. The temperature dependence of the void-number density should be taken into account if Fig. 1 is to be interpreted quantitatively. It can be seen that under typical reactor dose rates of 10^7–10^6 NRT dpa/s, the spatially homogeneous void distribution of Al and Mo is *dynamically unstable* due to vacancy emission for irradiation temperatures above approximately $0.39T_m$. This is about 90 °C for aluminum and 860 °C for molybdenum (see Table 1). For niobium instability occurs at about similar temperatures $T/T_m \approx 0.41$ (=830 °C) even for dose rates as high as 10^{-3} NRT dpa/s under ion irradiation. Experimental observation of partial ordering of large voids in neutron irradiated copper at 420 °C ($\cong 0.5T_m$) [34] also corroborates the instability condition $K < K_{cr}$.

From the foregoing discussion, it can be seen that vacancy emission may indeed drive the dynamics of the evolution of a void ensemble to instability, resulting in its coarsening. In the presence of a biasing mechanism serving as a selection process, the instability may lead to the development of a spatially heterogeneous distribution with a geometric structure. This forms the subject of discussion in the following section.

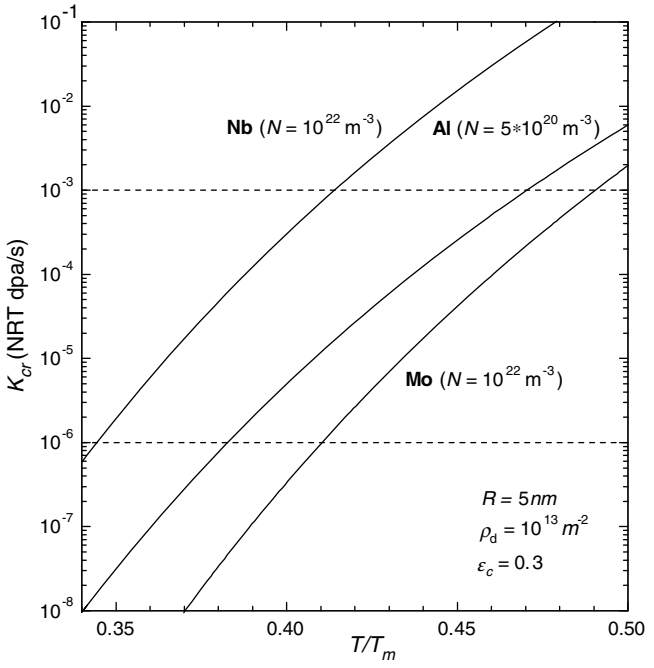

Fig. 1. Temperature dependence of K_{cr}, the critical dose rate below which the homogeneous void distribution is unstable, in various metals for the void densities typical for the corresponding metals ($Z = 1.1$ for Al, and $Z = 1.05$ for Nb and Mo).

Table 1
Material parameters

Parameter[a]	Molybdenum	Niobium	Aluminum
Atomic volume, Ω (m^3)	1.34×10^{-29}	1.617×10^{-29}	1.03×10^{-29}
Vacancy migration energy (eV)	1.5	0.55	0.6
Vacancy formation energy (eV)	3	2.9	0.6
Vacancy diffusivity pre-exponential (m^2/s)	1.0×10^{-5}	1.5×10^{-6}	1.0×10^{-5}
Surface free energy, γ_s (J/m^2)	2.05	2	1.1
Melting temperature, T_m (K)	2898	2690	933

[a] Material parameters, except surface free energy, are taken from Ref. [33].

3. Effects of one-dimension self-interstitial diffusion

The self interstitials generated in irradiated metals often contain a component that adopts the form of a one-dimensional diffuser along the crystallographic close-packed directions. In this section, we investigate the consequence of the presence of this component on the void coarsening process discussed in Section 2.

We call the average distance between consecutive changes of the direction of motion of the 1-D diffuser the mean free path (MFP). Depending on intrinsic parameters such as inter-atomic interaction, crystallographic structure, and extrinsic parameters such as the void-number density, impurity content, etc., the MFP varies. Let us first consider the case where the MFP is much smaller than the average distance between voids.

3.1. Short MFP case

The contribution of 1-D SIAs to the growth rate of the mth void in this case is given by [9]

$$R_m^2 \left.\frac{dR_m}{dt}\right|_{1-D\,\text{interstitials}} = -\frac{\varepsilon_i G R_m^2}{4\pi[N\bar{R}^2 + \rho_d d/4]}, \tag{14}$$

where $\bar{R}^2$ is the average square void radius, and d is the effective diameter of absorption of 1-D SIAs by dislocations. From Eqs. (13) and (14), the dynamic equation for δR_m becomes,

$$R_m^2 \frac{d(\delta R_m/R)}{dt} = \frac{2(Z-1)\rho_d}{(4\pi NR + \rho_d)} \times \left[\tilde{G} + G_0 - (1-\varepsilon_i)G - \frac{\varepsilon_i G(4\pi NR + \rho_d)}{2(Z-1)\rho_d}\frac{4\pi NR^2 + 2\pi\rho_d d - \rho_d R}{4\pi NR^2 + \pi\rho_d d}\right] \times \left(\frac{\delta R_m}{R}\right). \tag{15}$$

This equation shows that the presence of 1-D diffusing component tends to stabilize the dynamics of the void ensemble. Indeed, voids with a smaller than average surface area receive a reduced 1-D interstitial flux and thus grow faster, while the larger voids absorb a higher-than-average flux of such interstitials and grow slower. The presence of 1-D self-interstitials tends to sharpen the void size distribution and thus opposes the broadening effect of vacancy emission.

The presence of the 1-D diffuser changes the stability conditions via the void-growth rates. From Eqs. (10) and (14), the average void growth rate is given by

$$\frac{dR}{dt} = \frac{\rho_d}{R(4\pi NR + \rho_d)^2} \times \left\{(Z-1)[(1-\varepsilon_i)G - G_0] - \varepsilon_i \frac{4\pi NR^2 + \rho_d R}{4\pi NR^2 + \pi\rho_d d}\left(1 - \frac{\pi d}{R}\right)\right\}. \tag{16}$$

Having assumed $\varepsilon_i << (Z-1)$, the conventional dislocation bias can be considered as the major driving force for void growth in the present case. At the point when void lattices are experimentally observable, the voids are usually the major sinks, i.e., $4\pi NR >> \rho_d$. Thus, even when the dislocation bias dominates the void growth, the last term in brackets in Eq. (15) is still non-negligible. This leads to a shift in the instability conditions towards a higher temperature. Nevertheless, due to the exponential dependence of K_{cr} on the irradiation temperature this quantitative shift is not important.

3.2. Long MFP case

When the MFP of the 1-D diffuser is comparable with the average void separation, the qualitative effect of 1-D diffusion becomes important. Indeed, due to the overlap of 1-D diffusing fields, the Woo–Frank theory [2,3] suggests that voids sufficiently close to each other along close-packed crystallographic directions receive a reduced 1-D interstitial flux compared to that given by Eq. (14). This may then cause the dynamic instability of a homogeneously distributed void ensemble, and the resulting bifurcation may lead to the disappearance of the non-aligned voids. The critical dose was calculated neglecting both stochastic effects and vacancy emission from the voids, and was found to be much too high compared with experimental observations [3]. Furthermore, as shown in [9], 1-D self-interstitial diffusion by itself does not cause the disappearance of the non-aligned voids. It only leads to a stationary state in which the aligned and non-aligned voids coexist, saturating at different sizes. Without a mechanism via which the non-aligned supercritical voids selectively disappear during evolution, it remains unclear how a well-defined void lattice may emerge.

In the following we will show that when dynamic stability is lost (i.e., $K < K_{cr}$), the void ensemble would evolve, in the presence of a flux of long-MFP 1-D self-interstitials, in such a way that the non-aligned voids completely dissolve, leaving only the aligned ones to grow further. It is also worth noting that in the present case the characteristic time for instability development in Eq. (15) is comparable with that for the average void growth in Eq. (16).

Thus, when the 1-D diffusion fields among neighboring aligned voids overlap, the 1-D interstitial flux received by each void is given by [9]

$$R_m^2 \left.\frac{dR_m}{dt}\right|_{1-D\,\text{interstitials}} = -\frac{\varepsilon_i G R_m^2 \Phi}{4\pi[N_{AV}\bar{R}_{AV}^2\Phi_{AV} + N_{RV}\bar{R}_{RV}^2 + \rho d/4]}, \tag{17}$$

where N_{RV} and $\bar{R}_{RV}^2$ (N_{AV} and $\bar{R}_{AV}^2$) are the concentration and the average square radius of the non-aligned (aligned) voids, respectively. Φ differentiates between the growth rates of the aligned and non-aligned voids. For the non-aligned voids, $\Phi = 1$. For the aligned voids $\Phi = \Phi_{AV}$, which takes into account the reduction in the 1-D interstitial flux given by [9]

$$\Phi_{AV} = \frac{((2M-q)\tanh(L/\lambda) + q\tanh(L/2\lambda))}{2M}, \tag{18}$$

where L is the nearest-neighbor distance in the void lattice, q is the average number of nearest void-lattice sites occupied by a void, and $\lambda = (D_1\tau_c)^{1/2}$ is the MFP along the close-packed atomic directions. D_1 is the 1-D diffusion coefficient and τ_c the mean lifetime between migration direction changes. In Eq. (18) we have also assumed that if the void is not in the crowdion supply cylinders (CSCs) of its nearest neighbors, then it is in the CSCs of the corresponding next nearest neighbors. Note that Φ_{AV} has a value of less than 1 and decreases with decreasing L/λ in general.

We have shown in [9] that, when the average distance between existing voids is comparable with the MFP of the 1-D diffuser, voids nucleate and grow practically exclusively in the small regions near the empty void lattice sites. This is because the probability of void nucleation is orders of magnitude higher in these localities, where there is a reduced self-interstitial flux [9]. In this regard, the

exponential dependence of the nucleation probability on the net vacancy flux has also been shown in [23]. As a result, two subsystems exist in the void ensemble: aligned voids along the close-packed atomic directions and the non-aligned ones.

To properly describe void nucleation when the vacancy emission is important, the growth of thermally unstable void embryos has to be considered beyond the critical size, which can only be achieved via stochastic fluctuations. This means that the deterministic Eq. (8) for void growth should be generalized to include the effect of stochastic fluctuations in point-defect fluxes received by the voids [23]. However, since the void coarsening considered in this paper is driven by vacancy emission and not stochastic fluctuations, the complete stochastic treatment that has to be performed is much too complex within the present scope. Instead, we adopt a simplified approach in the following and consider only the simultaneous evolution of two subsystems (the aligned and non-aligned voids) under the prevalence of the instability conditions.

When stochastic fluctuations are neglected, we may assume that each subsystem of the void ensemble can be characterized by the corresponding average radius R_{AV} and R_{RV} for the aligned and non-aligned voids, respectively. Then, the mean-field growth rate of voids in each subsystem, due to the 3-D moving point defects, can be found from the conventional rate equations. As a result, with the account of the 1-D self-interstitial flux given by Eq. (17), the total growth rate of aligned voids can be written as

$$R_{AV}\frac{dR_{AV}}{dt} = \frac{[G - D_v\rho_d(C_s(R_{AV}) - C_\infty) - 4\pi N_{RV}R_{RV}(C_s(R_{AV}) - C_s(R_{RV}))]}{(4\pi N_{AV}R_{AV} + 4\pi N_{RV}R_{RV} + \rho_d)} - \frac{(1-\varepsilon_i)G}{(4\pi N_{AV}R_{AV} + 4\pi N_{RV}R_{RV} + Z\rho_d)} - \frac{\varepsilon_i G R_{AV}\Phi_{AV}}{4\pi[N_{AV}R_{AV}^2\Phi_{AV} + N_{RV}R_{RV}^2\Phi_{RV} + \rho d/4]}. \quad (19)$$

Substitution $AV \leftrightarrow RV$ transforms Eq. (19) into the rate equation for the average radius of the non-aligned voids.

4. Results and discussions

Numerical integration of Eq. (19) for the aligned and the non-aligned voids are preformed and the results are presented in

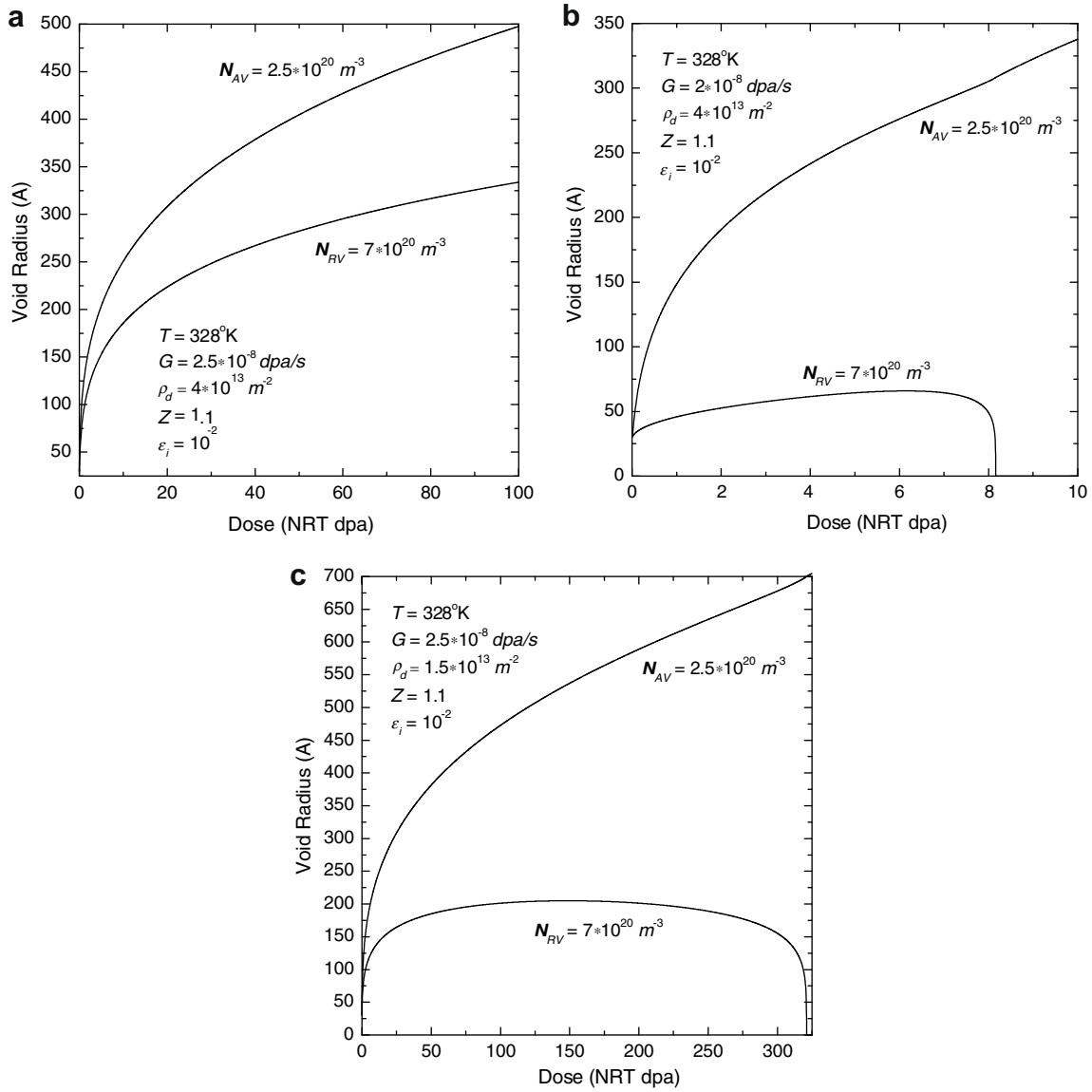

Fig. 2. Time dependence of the average radii of aligned ($\Phi_{AV} = 0.5$) and non-aligned voids ($\Phi_{RV} = 1.0$) at different point defect generations rates and dislocation densities.

Figs. 2(a–c) as a function of dose for aluminum. The number density of aligned voids N_{AV} chosen in the calculations corresponds to a void-lattice constant of ~250 nm. Comparing Figs. 2(a–c) shows the obvious presence of criticality in the behavior of the void ensemble. In Fig. 2(a) both the aligned and the non-aligned voids are monotonically growing, while in Fig. 2(b) the initial growth of non-aligned voids reverses to shrinkage subsequently. The rather drastic difference in the system behavior represented in the two figures is only caused by a small difference in effective generation rates of $G = 2.5 \times 10^{-8}$ in Fig. 2(a) and 2.0×10^{-8} dpa/s in Fig. 2(b). The non-aligned voids in Fig. 2(b), which are initially supercritical, become subcritical during further evolution and, as a result, disappear completely. Similarly, in Fig. 2(c), the total dislocation density is only slightly lower than that in Fig. 2(a). Yet, depending on the irradiation conditions and microstructure characteristics, the irradiation dose required for the shrinkage of the non-aligned voids can vary within a very wide range of values. This also explains why under practically identical experimental conditions (neutron flux $\cong 0.6 \times 10^{18}$ n/m^2/s, $E > 0.1$ MeV, i.e., dose rate $K \cong 5.2 \times 10^{-8}$ NRT dpa/s [18], irradiation dose $Kt \cong 19$ NRT dpa, temperature $T = 323 \pm 5$ K) voids in irradiated aluminum in one set of experiments are found to be randomly distributed [18], while in another the formation of a void hyperlattice was observed [29].

To illustrate the critical nature of the behavior of this two-component void ensemble, we generalize the expressions for G_0 and $\tilde{G}$ in Eqs. (11) and (12), derived for a single-component void system,

$$G_0 \cong \frac{8\pi D_v C_\infty \gamma_s \Omega (1 + \rho_d/k_c^2)(N_{AV} + N_{RV})}{(Z-1)RkT}, \tag{20}$$

$$\tilde{G} \cong \frac{(k_c^2 + \rho_d)^2}{(Z-1)\rho_d} \frac{D_v[C_s(R_{AV})N_{AV}/R_{AV} + C_s(R_{RV})N_{RV}/R_{RV}]\gamma_s \Omega}{(N_{AV} + N_{RV})kT}. \tag{21}$$

Here $k_c^2 = 4\pi(N_{AV}R_{AV} + N_{RV}R_{RV})$ is the total void sink strength. In Fig. 3 we show the time dependence of the ratio of $K_{cr} = (\tilde{G} + G_0)/\varepsilon_c$ to the corresponding nominal generation rate $K = G/\varepsilon_c$ for the three cases presented in Fig. 2. An initial decline in the ratio K_{cr}/K can be seen in Fig. 3, followed by a subsequent increase as the voids grow, leading to the loss of stability when $K_{cr}/K > 1$ (see Figs. 2(b) and (c). At this point, the process of void coarsening starts and the smaller non-aligned voids eventually become subcritical and shrink away. These results corroborate very well experimental observations in void hyperlattice formation in aluminum, in which void sizes just before the void ordering takes place (at 6 NRT dpa) are found to vary in a wide range between 8 and 80 nm in diameter [28].

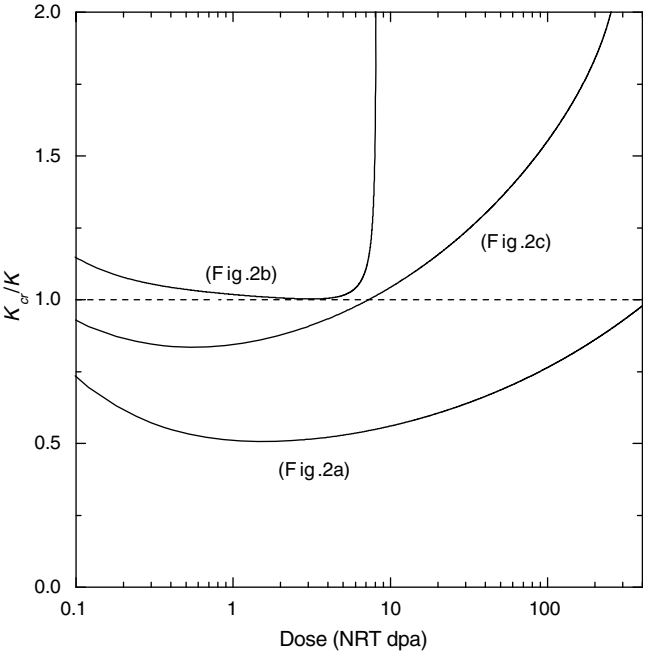

Fig. 3. Time dependence of the critical ratio K_{cr}/K for the cases presented in Fig. 2.

When $K_{cr}/K < 1$, both the aligned and non-aligned voids continue to grow (Fig. 2(a)). In this case, an increase of the 1-D self-interstitial flux received by the non-aligned voids only produce a small reduction in the average void sizes compared to the aligned voids, and does not lead to their dissolution at all. Note also that the difference between the average sizes of the aligned and non-aligned voids decreases with the decreasing ratio K_{cr}/K, due to the reduction of the effect of vacancy emission on the evolution of the smaller voids.

The present results show that for irradiation temperatures higher than $0.4T_m$, vacancy emission from the voids can play an important role in the shrinkage of the non-aligned voids. Unlike stochastic void coarsening, voids in the present case do not have to be small for the shrinkage to occur. On the contrary, it is actually the continuous growth of voids that creates the necessary condition for the dissolution of the non-aligned voids. Indeed, from Eqs. (9) and (12), an increase of the void sink strength increases the controlling parameter $\tilde{G}$ and pushes the system to coarsen. It is the smaller net vacancy flux (see the first two terms in Eq. (9)) that causes the initially supercritical non-aligned voids to become subcritical eventually. Thus, to instigate the shrinkage of non-aligned voids via vacancy emission, the sizes of the voids forming the lattice can be very large (tens or even hundreds of nanometers). However, for either mechanism, the shrinkage of the non-aligned voids is accomplished via sufficiently high void sink strength, as envisioned in the Woo–Frank theory [2]. The difference is that at lower temperatures, when vacancy emission from the voids is insignificant, this is achieved via a high number density of small voids [9].

According to the foregoing theory, the lattice constant of a hyper void lattice may give a good estimation of the MFP of the 1-D diffuser λ. In this regard, the void-lattice constant in niobium at $T = 1010$ °C is found to be 146 nm [35], in Mo at $T = 1120$ °C it is about 100 nm [36]. From the thermodynamic point of view, it is reasonable to assume that the MFP λ at lower temperatures is at least as long as those indicated at a higher temperature. In the temperature range where vacancy emission is unimportant, the void-lattice constants in molybdenum and niobium can be as small as 20–30 nm [5,26,35], corresponding to a ratio of $2\lambda/L \sim 10$, or a value of $\tanh(L/2\lambda) \sim 0.1$. Putting N_{RV} in Eq. (19) to zero and taking into account that at the lower temperatures $\rho/k_c^2 \sim 10^{-2}$, one can deduce that the voids in the lattice should saturate at a size of $R_{AV} = \pi d/\Phi_{AV}/[1 - (Z-1)(1-\varepsilon_i)/\varepsilon_i]$ if $\varepsilon_i > (Z-1)$. For randomly distributed voids, a similar consideration arrives at a saturation size of $R_{RV} = \pi d/[1 - (Z-1)(1-\varepsilon_i)/\varepsilon_i]$ (see also Eq. (16) and Ref. [9]). Note that for randomly distributed voids, the mean distance L_1 between two traps absorbing the one-dimensional random walkers is given by $L_1 = 2/\pi R_{RV}^2 N$. This means that, if the void swelling $S = 4\pi R_{RV}^3 N/3$ is less than 1%, which often is the case [5,26,27,37], the ratio $L_1/\lambda = (2/\pi^{1/3})(4/3S)^{2/3}N^{-1/3}/\lambda) > 3$, even when $\lambda N^{1/3} \sim 10$. Thus, we still have $\Phi_{RV} \approx 1$. Experimental observations at lower temperatures show that the diameter of the voids forming the lattice can be as small as 3–4 nm [5,26]. Then it follows from Eq. (19) that if $\varepsilon_i > (Z-1)$ random voids cannot exist in practice. This is contrary to experimental observations where random voids have average void sizes at least comparable with the aligned voids [26]. Thus, our assumption of $\varepsilon_i \sim 1\%$ is justifiable (see also Introduction and Ref. [9]).

In the present investigation, the role of the 1-D diffusers, which maintain the crystallographic relationship between the void and host lattice, is just to provide a mechanism via which voids destined to grow versus those destined to shrink during the coarsening process caused by the dynamic instability in the evolution of the void ensemble are differentiated. In this regard, it is worth noting that this mechanism does not distinguish between single SIAs in the <1 1 1> dumbbell/crowdion configurations which are often the ground-state configuration in the nonmagnetic bcc metals [38,39], and the crowdion clusters. Due to the relatively easy activation of the dumbbell/crowdion rotation (activation energy ≈ 0.2–0.4 eV), their associated kinetics at elevated temperatures are predominantly three-dimensional in character. The probability of a sufficiently long 1-D motion for hyperlattice formation is expected to be low. Consequently, the effective fraction ε_i of interstitials, which do not participate in the three-dimensional motion, will be low in this case as well. Of course, there is also the possibility that the 1-D self-interstitial diffuser responsible for the two types of void lattices may be different.

5. Summary and conclusion

We have investigated the evolution of a void ensemble in the presence of one-dimensionally migrating self-interstitials in a temperature range where the effect of vacancy emission is important. We find that when the effective point-defect generation rate is sufficiently low, the evolution of the void ensemble may become dynamically unstable, resulting in the coarsening of the void distribution. The presence of one-dimensionally migrating self-interstitials with mean-free path comparable to the average distance between voids can bias the void coarsening process such that the non-aligned voids shrink, leaving the aligned ones to grow. An imperfect lattice with empty lattice sites may then form. Due to the overlapping diffusion fields of neighboring voids, these sites are subjected to a reduced 1-D interstitial flux and are preferred void nucleation sites, which were eventually filled, completing the void lattice formation process.

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