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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

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