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ABSTRACT

Swelling of metals under irradiation is commonly assessed by calculating the volume fraction of voids, which appear at temperatures where vacancies are mobile. However, other clusters are formed, which may also have an impact on swelling. In particular, interstitial loops have recently been considered to give a significant contribution to swelling owing to their large relaxation volume. In this work, we perform calculations in nickel, based on interatomic potentials, to estimate the contributions of the various point defect clusters. We show that voids produce much more swelling than loops and stacking fault tetrahedra, whose contribution is essentially due to the dislocation core field, inducing a dilatation per unit length of around $1b^2$, where b is the Burgers vector. Evaluation of swelling should indeed be done by summing formation volumes, not relaxation volumes, the latter being related to lattice parameter change as measured by x-ray diffraction. We also discuss the case of “lattice swelling” occurring when vacancies are immobile. When self-interstitial atoms cluster as dislocation loops, this swelling mode turns out to be nothing but “void” swelling in a regime where vacancy mobility is so low that vacancies do not cluster appreciably, leaving only interstitial loops visible in transmission electron microscopy.

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I. INTRODUCTION

Swelling of pure metals under irradiation is due to the formation of point defects and point defect clusters. In addition, in metallic alloys, secondary phases can form, modifying the overall density and changing the volume of the material. Above the temperature corresponding to stage III of point defect recovery, at which vacancies become mobile, voids can grow to large sizes by absorption of vacancies. They are considered to have the largest contribution to swelling. That is why swelling is generally estimated from transmission electron microscopy (TEM) observations by counting voids.¹ However, other types of clusters are produced under irradiation and “void swelling” may only be a part of the total swelling. In some recent works, it has been argued that dislocation loops produce significant swelling^{2,3} due to their large relaxation volume,⁴ and that they should be considered to estimate swelling, instead of voids.⁵ Using the example of pure nickel, we determine the contribution of the different point defect clusters (voids, interstitial Frank loops, and stacking fault tetrahedra) to macroscopic swelling. Formulas are given to evaluate the macroscopic swelling of a microstructure containing point defect clusters.

Finally, the evaluation of swelling in metallic alloys is briefly discussed.

II. METHODS

The change in volume upon introduction of a defect in a finite crystal is the relaxation volume V^r . For cubic crystals, it is related to the trace of the dipole tensor \mathbf{P} of this defect through⁶

$$V^r = \frac{\text{Tr}\mathbf{P}}{3K}, \quad (1)$$

where $K = (C_{11} + 2C_{12})/3$ is the bulk modulus and C_{ij} are the elastic constants. The elastic dipole is conveniently calculated from the average residual stress in an unstrained simulation box of volume V , where the defect is introduced^{7,8}

$$\sigma_{ij} = -\frac{1}{V}P_{ij}. \quad (2)$$

Since experimentally the number of atoms remains constant when a defect is formed (except under ion irradiations where atoms are

injected in the material), the formation of clusters made of self-interstitial atoms (SIAs), such as dislocation loops, requires the removal of atoms somewhere else in the material. Likewise, the formation of vacancy clusters is only possible if atoms which are removed are placed at another location in the material. It is convenient to remove or to put these atoms at the surface of the sample where they occupy an atomic volume Ω , so the formation volume of a defect is simply related to the relaxation volume through

$$V^f = V^r - n\Omega \quad \text{for a cluster containing } n \text{ SIAs,} \quad (3)$$

$$V^f = V^r + n\Omega \quad \text{for a cluster containing } n \text{ vacancies.} \quad (4)$$

This relation is quite general and can also be applied to non-cubic materials.

In the following, we calculate the relaxation and formation volumes of defects in nickel using the interatomic potential of Angelo *et al.*⁹ The potential of Foiles *et al.*¹⁰ is also used to simulate SIA Frank loops and investigate the effect of potential. Energy minimization is performed with the conjugate gradient algorithm as implemented in the LAMMPS code.¹¹ Initial configurations of the various defects considered are generated as follows. Voids are created by simply removing atoms within a spherical domain. The radius of a spherical void containing n vacancies is such that $4\pi r^3/3 = n\Omega$, where $\Omega = a^3/4$ and a is the lattice parameter ($a = 3.52 \text{ \AA}$). Interstitial Frank loops are generated by adding a disk of atoms in a $\{111\}$ plane, leading to an extrinsic stacking fault surrounded by a circular Frank partial dislocation. To accelerate convergence, we displace the atoms of the surrounding matrix according to the displacement field generated by a prismatic loop in an elastically isotropic material.¹² The relation between the radius r and the number of SIAs in the loop is $\pi r^2 b = n\Omega$, with b being the magnitude of the Burgers vector ($b = a/\sqrt{3}$). Only perfect stacking fault tetrahedra (SFTs) are considered in this study. The starting configuration consists of a triangular vacancy loop in a $\{111\}$ plane. The Frank partial dislocations surrounding the intrinsic stacking fault dissociate into $1/6[110]$ stair-rod dislocations and $1/6[112]$ Shockley partial dislocations. The latter “propagate” during energy minimization and interact with each other, leaving only stair-rod dislocations surrounding stacking faults on each side of a perfect tetrahedron (Silcox–Hirsch mechanism¹³). The radius of the SFT is defined as the radius of its circumscribed sphere, related to the area of the initial triangular loop S (a face of the SFT) through $S = 2r^2/\sqrt{3}$. Using the same kind of formula as for the interstitial loop $Sb = n\Omega$ leads to the relation between the radius and the number of vacancies in the SFT. For all simulations, the convergence of dipole tensors is checked by varying the size of the box. The retained values of dipole tensors are the ones calculated in a box of size at least equal to ten times the radius of defects.

III. RELAXATION AND FORMATION VOLUMES OF POINT DEFECT CLUSTERS IN NICKEL

Relaxation and formation volumes for the different point defect clusters considered here are shown in Figs. 1 and 2, respectively. The relaxation volume of voids is very small,^{5,14} while the relaxation volume of SIA loops essentially corresponds to $n\Omega$,^{5,15,16}

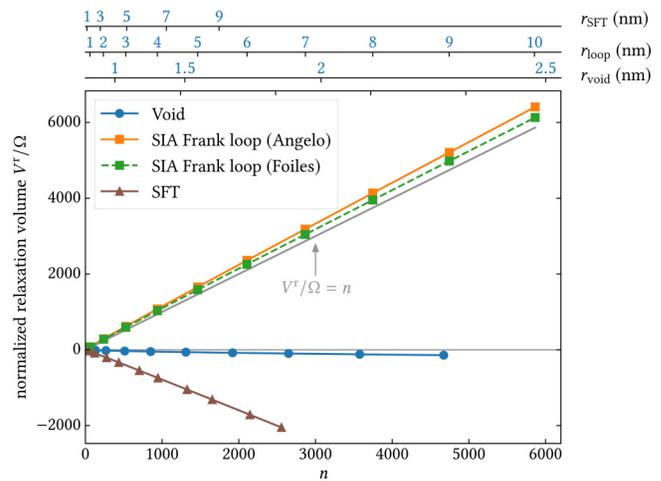


FIG. 1. Normalized relaxation volumes of voids, Frank dislocation loops, and SFTs in nickel, computed with Angelo’s interatomic potential. The relaxation volume of loops is also given for Foiles’s interatomic potential. Index n represents the number of vacancies (for voids and SFTs) or SIAs (for loops) in clusters, while r_x represents the radius of cluster type X .

as predicted by linear elasticity.¹⁷ Small differences appear when the interatomic potential is changed, as can be seen with the results obtained with Foiles’s potential. Relaxation volumes of vacancy loops are expected to be very similar but of opposite sign. Results are not reported here since with the potential used, Frank partial dislocations tend to dissociate in this case, which may not be

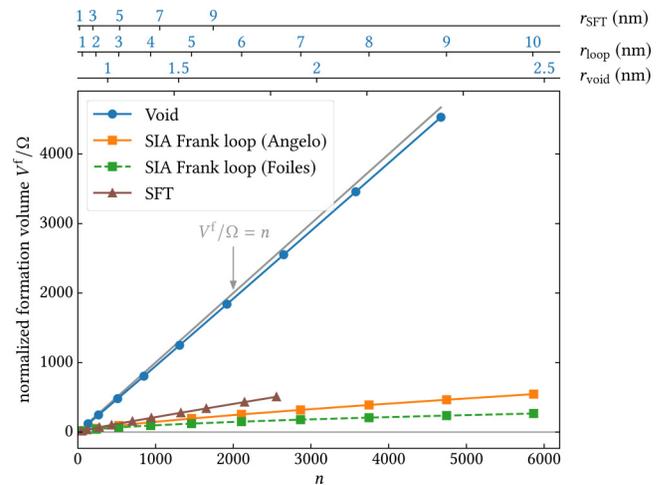


FIG. 2. Normalized formation volumes of voids, Frank dislocation loops, and SFTs in nickel, computed with Angelo’s interatomic potential. The formation volume of loops is also given for Foiles’ interatomic potential. Index n represents the number of vacancies (for voids and SFTs) or SIAs (for loops) in clusters, while r_x represents the radius of cluster type X .

representative of the real behavior of vacancy loops. Finally, as all vacancy clusters, SFTs exhibit negative relaxation volumes, which in magnitude are slightly smaller than SIA loops.

Formation volumes are obtained from relaxation volumes using Eqs. (3) and (4). As shown in Fig. 2, the formation volume of voids is very close to the total volume of atoms removed from the matrix ($V^f \approx n\Omega$), which are now placed at other sinks (surfaces, dislocations, etc.). This is due to the small relaxation of the void surface.

On the contrary, SIA loops generate only a small increase in the volume of the material, since the increase in volume due to the loop itself (relaxation volume) is nearly compensated by a decrease in volume resulting from the removal of atoms somewhere else. Loop swelling is due to two elements: the stacking fault, corresponding to a local hexagonal close packed stacking ($\dots ABCAB \dots \rightarrow \dots ABACAB \dots$), and the dislocation.¹⁸ The stacking fault is expected to induce a variation of volume equal to $\pi r^2 \delta l$, where δl is the expansion per unit area of stacking fault. The contribution of the dislocation is $2\pi r \delta A$, where δA is the dilatation per unit length of dislocation. Since $\pi r^2 b = n\Omega$, a convenient fit of the formation volume in this case is

$$\frac{V^f}{\Omega} = a_0 + a_1 n^{1/2} + a_2 n, \quad (5)$$

where the second term corresponds to the dislocation line and the third term to the stacking fault. A better fit for small sizes, down to the minimum value $r = 1 \text{ nm}$ ($n = 59$) considered here, was obtained by introducing an additional constant term a_0 . We obtain for Angelo's potential $a_0 = -13.22$, $a_1 = 3.54$, and $a_2 = 0.049$ and for Foiles's potential $a_0 = -7.46$, $a_1 = 3.16$, and $a_2 = 0.005$. Since we are mostly interested in clusters which are visible in TEM, we did not explore the validity of these fits for smaller SIA clusters. Equation (5) is not expected to be valid for single SIAs and clusters containing only a few SIAs, which cannot be considered as dislocation loops.

From the values of a_1 , we obtain $\delta A = 1.14b^2$ and $\delta A = 1.01b^2$ for Angelo's and Foiles's potentials, respectively ($b = 2.03 \text{ \AA}$). These values are in line with early estimates given by nonlinear elasticity models¹⁹ ($\delta A \approx 2b^2$ in nickel). More quantitative results are provided by interatomic potential calculations of the core field contribution.^{20–24} The typical value given by such calculations, usually performed in an infinite system using flexible boundary conditions,²⁰ is $\delta A \approx 0.5b^2$. In the present study, the calculation of relaxation and formation volumes from dipole tensors includes the contribution of image fields due to free surface boundary conditions.²⁵ Only a few calculations were performed with flexible boundary conditions and surface relaxation.²¹ They typically lead to $\delta A \approx 1b^2$, in excellent agreement with the present results. It should be kept in mind, however, that interatomic potentials may not correctly capture core effects^{26,27} so that these results should be only viewed as estimates.

The expansion per unit area of stacking fault deduced from the fit is $\delta l = 0.10 \text{ \AA}$ for Angelo's potential and $\delta l = 0.01 \text{ \AA}$ for Foiles's potential. An independent calculation of the expansion due to an infinite extrinsic stacking fault with these two potentials leads to $\delta l = 0.107 \text{ \AA}$ and $\delta l = -0.015 \text{ \AA}$, respectively, in reasonable

agreement with values deduced from the fits. We also note that the trends are consistent with the higher value of c lattice parameter in the hexagonal phase for Angelo's potential ($c = 4.18 \text{ \AA}$) than for Foiles's potential ($c = 4.06 \text{ \AA}$).

The formation volume of SFTs can also be fitted with Eq. (5), leading to $a_0 = -2.40$, $a_1 = 1.46$, and $a_2 = 0.17$. The minimum size considered for SFTs is $r = 1.4 \text{ nm}$ ($n = 45$), so the fit cannot be guaranteed to be valid for smaller clusters. For an SFT of edge length d , the contribution of the dislocation to the formation volume is $6d\delta A$. Since $d = an^{1/2}$, we have $\delta A = 1.09b^2$ ($b = 0.83 \text{ \AA}$), which is consistent with values obtained for Frank loops, although the magnitudes of Burgers vectors are very different. The intrinsic stacking fault ($\dots ABCABC \dots \rightarrow \dots AB \parallel ABC \dots$) produces an expansion of $4S\delta l = \sqrt{3}d^2\delta l$ so that $\delta l = 0.09 \text{ \AA}$. This is in agreement with the expansion per unit area calculated for an infinite intrinsic stacking fault ($\delta l = 0.107 \text{ \AA}$), which is the same as for an extrinsic stacking fault. The rather large contribution of the stacking fault with this potential leads to an overall formation volume larger than the one of Frank loops and to a nearly linear variation with n over the range investigated. This should not be considered as a general result, though. If δl is small, as with Foiles's potential, the contribution of the dislocation will become dominant. In this case, a Frank loop will have a larger formation volume, as can be checked by comparing values of parameter a_1 .

IV. DISCUSSION

As mentioned in Sec. II, one should consider formation volumes to evaluate swelling, since the number of atoms is kept constant during the formation process of a point defect cluster. It is common to say that atoms are put on the surface (for the creation of a vacancy defect) or removed from it (for an SIA defect),²⁸ although it does not necessarily represent the physical process by which the defect is created. In the bulk, a more realistic picture is to move atoms to or from dislocation jogs, which also corresponds to the creation or removal of lattice sites. Under irradiation, if the temperature is low enough to ignore generation of point defects at sinks (dislocations, surfaces, etc.), the main creation mechanism of point defects is by Frenkel pair formation. Although sinks do not play any role in this case, summing formation volumes of defects leads to the correct estimation of swelling (Fig. 3).

The relative contributions of the different point defect clusters to swelling depend on their size and concentration. The very large formation volume of voids make them the overwhelming contributor to swelling at the swelling peak temperature of metals. It is interesting to investigate the less obvious case of lower temperatures. This kind of conditions typically corresponds to irradiations of austenitic steels in pressurized water reactors (PWRs). Results from microstructural observations are reviewed in Ref. 29, and typical values are given in Table I. The loop properties are roughly independent of temperature between 260 and 380 °C, at least in the quasi-steady state arising after a few dpa. This is also the case for the dislocation network,³⁰ but not for voids, which are very sensitive to temperature. That is why two sets of values are reported. Swelling is estimated the following way. For loops, given their small size, the stacking fault is not expected to give a significant contribution, so only volume expansion due to the dislocation is taken into

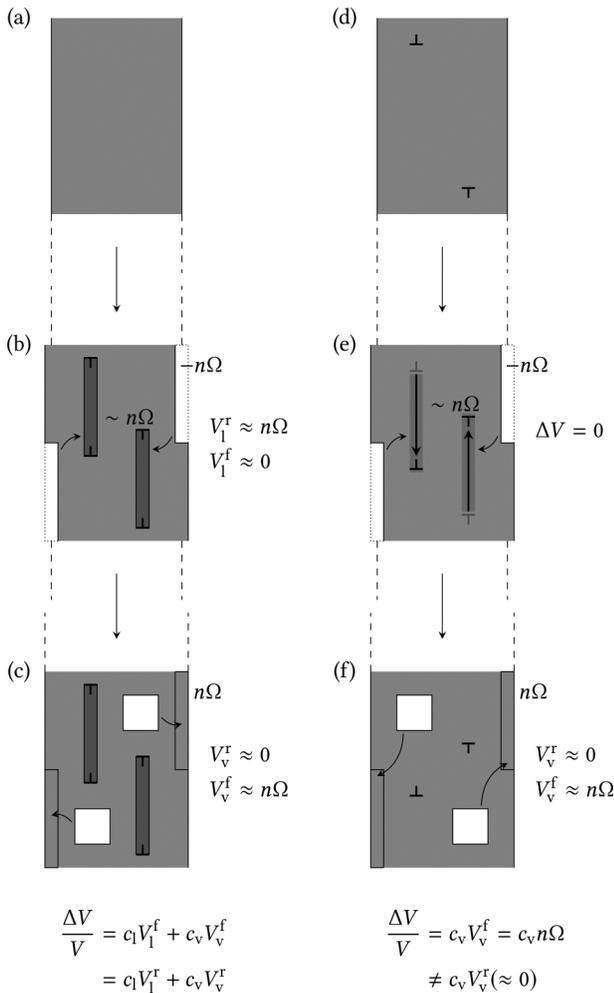


FIG. 3. Estimation of swelling of irradiation microstructures, using artificial steps to highlight the contribution of each defect type. Swelling due to dislocation core effects, stacking faults and relaxation of void surfaces is neglected. The average position of the surface, which is related to macroscopic swelling, is shown with dashed lines. (a) Pristine material, before irradiation. (b) Material with SIA loops; to first order, the relaxation volume of loops is equal to the volume removed from the surface (or dislocations for bulk materials), so loops do not contribute appreciably to swelling. (c) Material with voids: swelling occurs due to the creation of atomic sites. In this very specific case, where equal numbers of vacancies and SIAs are present in clusters, swelling may also be evaluated by summing relaxation volumes. (d) Pristine material, containing some network dislocations. (e) Material after climb of dislocations due to SIAs, which induces no volume change and (f) material with voids. In this case, often seen experimentally, where a defect imbalance exists in clusters, relaxation volumes cannot be used to estimate swelling.

account,

$$\left. \frac{\Delta V}{V} \right|_{\text{loops}} = c_l 2\pi\eta\delta A = c_l 2\pi\eta b^2, \quad (6)$$

where η and c_l are the loop average radius and concentration, respectively. Likewise, the contribution of a density ρ_d of network dislocations, with Burgers vector $1/2[110]$, is

$$\left. \frac{\Delta V}{V} \right|_{\text{dislocations}} = \rho_d b^2. \quad (7)$$

They typically induce slightly more swelling per unit length than loops, owing to their larger Burgers vector. Finally, void swelling is estimated through

$$\left. \frac{\Delta V}{V} \right|_{\text{voids}} = \frac{4}{3}\pi r_v^3 c_v, \quad (8)$$

where r_v is the average radius of voids and c_v is their concentration. From Table I, one sees that at 300 °C, swelling produced by the different defects in the microstructure is of the same order of magnitude, so it is important to take all of them into account. At $T \sim 350$ °C, the contribution of voids is one order of magnitude larger than the other defects, so swelling may be estimated by considering only voids. We note that values of swelling reported here are very small. The contribution of voids to swelling increases with temperature so that at temperatures where swelling is a potential technological problem, it is safe to calculate swelling values with voids.

In some works, relaxation volumes instead of formation volumes were used to estimate swelling.^{2,31–33} They may indeed be used in a very particular case. Let us assume for simplicity that only SIA loops and voids are present in a material of volume V , and let $c_{X,n}$ be the concentration of a loop ($X=l$) or a void ($X=v$) containing n self-defects, of relaxation and formation volumes $V_{X,n}^r$ and $V_{X,n}^f$ respectively. Swelling can be written as³⁴

$$\frac{\Delta V}{V} = \sum_n c_{l,n} V_{l,n}^f + \sum_n c_{v,n} V_{v,n}^f. \quad (9)$$

Using Eqs. (3) and (4), it also reads

$$\frac{\Delta V}{V} = \sum_n c_{l,n} V_{l,n}^r + \sum_n c_{v,n} V_{v,n}^r + \sum_n (c_{v,n} - c_{l,n}) n\Omega. \quad (10)$$

TABLE I. Contribution to swelling of various elements of the microstructure in PWR conditions. Experimental data are extracted from the review in Ref. 29. Between 260 and 380 °C, the loop and dislocation microstructures are roughly independent of temperature, whereas the void microstructure is very sensitive to it. Therefore, two sets of values, at $T = 300$ °C and $T = 350$ °C, are given for voids. The contribution of stacking fault is neglected for dislocation loops. The volume expansion per unit length of dislocation is assumed to be $1 b^2$. The relaxation of void surface is not taken into account. The lattice parameter is $a = 3.6$ Å.

	Density	Radius (nm)	Swelling (%)
Loops	10^{22} m^{-3}	6	2×10^{-3}
Dislocations	10^{15} m^{-2}	...	6×10^{-3}
Voids	$T \sim 300$ °C	10^{23} m^{-3}	5×10^{-3}
	$T \sim 350$ °C	10^{21} m^{-3}	5×10^{-2}

The last term of this equation is the difference between the total atomic fraction of vacancies and the total atomic fraction of SIAs. If they are equal, the last term is zero, so relaxation volumes and formation volumes can be used indifferently to calculate swelling. This is the case shown in Figs. 3(a)–3(c). It arises, for example, in low temperature irradiations, where defect migration is not sufficiently activated to enable defect elimination at sinks. Swelling produced by displacement cascades generated in molecular dynamics simulations, where no sinks are present, can also be estimated by summing relaxation volumes.⁵

In the general case, relaxation volumes are not related to macroscopic swelling but to lattice parameter change Δa as measured by x-ray diffraction.³⁵ For cubic crystals, if defects are uniformly distributed,^{35,36} we have²⁸

$$3 \frac{\Delta a}{a} = \sum_n c_{l,n} V_{l,n}^r + \sum_n c_{v,n} V_{v,n}^r, \quad (11)$$

so the change in macroscopic length ΔL , related to swelling, can be written as

$$\begin{aligned} 3 \frac{\Delta L}{L} &= \sum_n c_{l,n} V_{l,n}^f + \sum_n c_{v,n} V_{v,n}^f \\ &= 3 \frac{\Delta a}{a} + \sum_n (c_{v,n} - c_{l,n}) n \Omega. \end{aligned} \quad (12)$$

If the total atomic fraction of vacancies and SIAs are equal, the last term of Eq. (13) is zero and the change in length is equal to the change in lattice parameter. That is why swelling at low temperature or very low dose [when last term of Eq. (10) or Eq. (13) is negligible] is sometimes referred to as “lattice parameter swelling” or simply “lattice swelling.”^{31,37–39} This naming suggests that this swelling mode is different from void swelling observed at higher temperatures or higher doses, because swelling would be induced by lattice parameter change.⁴⁰ If this were true, a microstructure containing only interstitial loops would give rise to macroscopic swelling, since loops have large positive relaxation volumes and thus induce significant lattice parameter change.⁴¹ However, their relaxation volume is almost compensated by the removal of atoms from some sinks, so swelling due to interstitial loops is nearly zero [Fig. 3(b)]. In other words, in a system without vacancies, the first and third terms of Eq. (10), or equivalently the first and second terms of Eq. (13), approximately balance each other out. So the specific case where the change in length is equal to the change in lattice parameter requires vacancies or vacancy clusters, with the same total amount of vacancies and interstitials in the system. Vacancies and voids have low relaxation volumes but contribute to swelling due to the necessary creation of atomic sites when they are generated, as evidenced in differential dilatometry measurements⁴² [$c_{v,n} n \Omega$ term in Eq. (13)]. The reasoning is still valid if some vacancies cluster as loops or SFTs. These clusters do not contribute appreciably to swelling, so the overall swelling is lower than if all vacancies cluster as voids. They also induce a decrease in lattice parameter due to their negative relaxation volume, so the equality between the change in length and change in lattice parameter still

holds. Therefore, it appears that “lattice swelling” in pure metals is nothing but vacancy and void swelling in a regime where vacancy clusters are so small that they are invisible in TEM, while SIA loops are in general visible.³⁹ If the temperature is so low that SIAs do not cluster appreciably, SIAs and small SIA clusters also contribute to swelling, since $V_n^r/n\Omega$ is larger than one (see Fig. 5 in Ref. 15) and so $V_n^f > 0$. For example, in nickel, the relaxation volumes of SIAs and vacancies are 1.8 and -0.2Ω , respectively,⁶ so in a sample containing an equal concentration of vacancies and SIAs, the contribution of these two species to swelling is roughly the same ($V^f = 0.8\Omega$ for both species). To conclude, to avoid any confusion between the strain created by defects and macroscopic swelling, we think a good practice is to use the term “lattice swelling” only when swelling does not imply the creation of new atomic sites through vacancies, such as swelling due to atomic disordering.⁴³ Otherwise, as there is no general link between strain and macroscopic swelling, “lattice strain” is more appropriate to refer to change in lattice parameter.⁴⁴

Let us consider the case where the amounts of SIAs and vacancies in clusters are different. Under irradiation, such a defect imbalance arises if point defects migrate to sinks, such as grain boundaries, surfaces and dislocations, as shown in Figs. 3(d) and 3(f). Figure 3(f) represents the system after dislocation climb due to a preferential absorption of SIAs and agglomeration of the remaining vacancies as voids. It is clear from the thought experiment (d) to (f) that dislocation climb alone does not produce volume change; it is because some sites are left vacant in the matrix that additional atomic planes are created and that the material swells. Incidentally, we note that dislocations are not necessary to explain swelling; SIAs may be absorbed by other sinks, such as grain boundaries and surfaces. When point defects reach sinks, one typically observes a decoupling between the change in lattice parameter, given by relaxation volumes of defects, and the change in length, given by formation volumes.⁴⁵ In systems without sinks, this decoupling can also occur by coalescence of loops, which form a dislocation network, as seen in simulations of Frenkel pair accumulation.⁴⁶ The transformation of loops into a dislocation network does not modify swelling, but leads to a decrease in lattice parameter change. The evolution of swelling has been explained by the fact that coalescence of loops create new atomic planes, which “preserves the increase in volume due to interstitial defects.”⁴⁶ If we now admit that swelling is due to vacancies and that dislocation loops have nearly zero formation volume, it appears quite natural that swelling is unaffected by transformation of loops into a dislocation network. The lattice parameter decreases because SIA loops, which have large relaxation volume, disappear from the system. The change in lattice parameter can even become negative, which is ascribed to the negative relaxation volume of vacancies and vacancy clusters (especially, vacancy loops and SFTs).^{45,46}

Although in general swelling is only correctly described by formation volumes of defects, there is one case where relaxation volumes may be useful from an experimental point of view. At low dose, vacancy clusters are hardly resolvable with TEM, so an estimation of swelling by formation volumes of voids is difficult. Due to the larger mobility of SIAs and the two-dimensional character of their clustering process, SIA loops are larger and more easily observed. If one assumes that the sinks do not absorb or generate

point defects, swelling can advantageously be evaluated by summing the relaxation volume of loops.⁵ As soon as the numbers of SIAs and vacancies in clusters are not the same, using relaxation volumes would in addition require to track the loss of defects at sinks [last term of Eq. (10)], which is unfeasible experimentally. The more physical and general method based on the formation volumes of defects, especially voids, should be preferred^{1,47-51} (note that in some sources, formation volumes are used but they are named relaxation volumes^{47,48}).

So far, we have only considered point defect clusters. In metallic alloys, precipitates can appear and lead to either swelling or densification,^{52,53} following complex precipitation sequences, where point defects may play a crucial role.⁵⁴ In addition, point defect cluster microstructures are known to be altered if secondary phases precipitate. The approach consisting in summing formation volumes remains valid in alloys, provided formation volumes of precipitates are taken into account and only precipitates which appear under irradiation are considered. For a given precipitate, the formation volume is given by (Fig. 4)

$$V_p^f = V_p^r + \sum_{\alpha} n_{\alpha} V_{\alpha}^f - \sum_{\alpha'} n_{\alpha'} V_{\alpha'}^f, \quad (14)$$

where n_{α} is the number of species α removed to create the void containing the precipitate, $n_{\alpha'}$ is the number of species α' inserted as a secondary phase inside the void, and V_p^r the relaxation volume

of the precipitate. The formation volume is

$$V_{\alpha}^f = V_{\alpha}^r + \Omega \quad \text{for a substitutional atom,} \quad (15)$$

$$V_{\alpha}^f = V_{\alpha}^r \quad \text{for an interstitial atom.} \quad (16)$$

For example, precipitates which lead to densification, such as carbides in austenitic steels, will exhibit a negative formation volume and will have the opposite behaviour from voids. For sufficiently large precipitates, V_p^r is expected to be negligible compared to the second and third terms in Eq. (14). These two terms reflect the difference in density between the matrix and the secondary phase. For a phase denser than the matrix, one typically adds more substitutional species in the precipitate region than one removes from it, so the overall contribution of the two terms is negative and one expects densification of the material.⁵⁴ The reverse trend, i.e., swelling, is expected for the precipitation of a less dense phase. However, if a large atomic fraction of interstitial atoms, such as carbon, must be taken from the matrix to form the secondary phase, the last term of Eq. (14) will give a significant contribution to the formation volume, which can even become negative. This is precisely what occurs with carbides in austenitic steels: although this phase is less dense than the austenite, its formation results in densification due to the contraction of the matrix as carbon concentration decreases.⁵⁵ Therefore, within the present simple model, the negative formation volume of carbides would be mostly due to the positive relaxation volume of carbon atoms in the matrix. This case is particularly important, since the volume change associated with carbides can be comparable to the one associated with voids at low doses. So, austenitic steels containing voids and carbides can either swell or densify, depending on the relative volume fractions of voids and carbides, and to obtain the precise overall volume change, it is necessary to sum the formation volumes of both voids and precipitates.

In this approach, we consider that the composition of the matrix remains roughly constant during the precipitation process. Actually variations in chemical composition of the matrix can affect its density. This has been shown, for example, in Fe-Ni Invar alloys^{56,57} and in Fe-Cr-Mn alloys.⁵⁸ If the density of each phase is known as a function of its composition, a simple rule of mixture⁵⁵ can also be used advantageously instead of the previous model.

V. CONCLUSIONS

Using interatomic potentials in nickel, we have calculated the relaxation and formation volume of voids, dislocation loops, and SFTs and discussed the estimation of swelling due to these defects. The following conclusions are drawn:

- Swelling should be estimated by summing the formation volumes of all defects in the microstructure, not relaxation volumes. Relaxation volumes are related to lattice parameter change and only give a good estimation of swelling if the number of SIAs and vacancies in clusters are the same, which is rarely seen in practice. In particular, as soon as point defects are lost to sinks, using formation volumes is the only relevant method to estimate swelling.

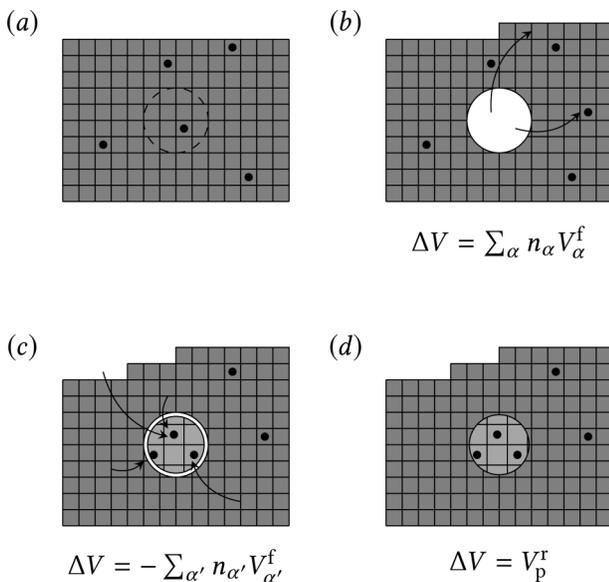


FIG. 4. Calculation of the formation volume of a precipitate. (a) Pristine material, without precipitate. Interstitial atoms are represented by black dots. (b) Removal of the atoms in the precipitate region, thus creating a void, whose surface is not relaxed. (c) Insertion of atoms in the void as a new phase, without relaxation. (d) Relaxation of the system.

- Formation volume of voids is far larger than the formation volume of SIA loops, vacancy loops, and SFTs. It is essentially given by $n\Omega$, where n is the number of vacancies in the void and Ω the atomic volume. The formation volume of SIA or vacancy loops and SFTs is due to the dislocations, which induce a dilatation per unit length of around $1b^2$, whatever the dislocation type (Frank or stair-rod dislocations for loops and SFTs, respectively). The stacking fault can also contribute for large clusters. If a precise evaluation of swelling by dislocation loops and SFTs is required, the volume changes due to dislocations and stacking faults can be evaluated separately through dedicated first principles calculations.²⁶
- Except for low dose irradiation conditions, the contribution of vacancy clusters is, therefore, much more important than the contribution of dislocation loops and SFTs. Even at low temperature, in the so-called “lattice swelling” regime, we have shown that swelling is not due to lattice parameter change mostly induced by dislocation loops but to the creation of new atomic sites, which is only possible because mono-vacancies or small voids are present in the matrix.
- In metallic alloys, although swelling mechanisms also involve precipitation, we have shown that in many cases, the same concept of formation volume can be applied.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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