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Absence of saturation of void growth in rate theory with anisotropic diffusion

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Abstract

We present a first attempt at solution the problem of the growth of a single void in the presence of anisotropically diffusing radiation induced self-interstitial atom (SIA) clusters. In order to treat a distribution of voids we perform ensemble averaging over the positions of centres of voids using a mean-field approximation. In this way we are able to model physical situations in between the Standard Rate Theory (SRT) treatment of swelling (isotropic diffusion), and the purely 1-dimensional diffusion of clusters in the Production Bias Model. The background absorption by dislocations is however treated isotropically, with a bias for interstitial cluster absorption assumed similar to that of individual SIAs. We find that for moderate anisotropy, unsaturated void growth is characteristic of this anisotropic diffusion of clusters. In addition we obtain a higher initial void swelling rate than predicted by SRT whenever the diffusion is anisotropic.

1. Introduction

Until recently, models for the kinetics of evolution of microstructure of materials under cascade damage conditions have been based primarily on either the case of isotropic 3-dimensional (3-D) diffusion of vacancies and self-interstitial atoms (SIAs), or the case where some SIAs are clustered and perform strictly 1-dimensional (1-D) glide. The first case is a principal assumption of Standard Rate Theory (SRT). SRT uses a bias in the absorption of defects by dislocations [1], favouring SIAs over vacancies, and so allows unsaturated void swelling at large neutron doses, as observed experimentally. However, SRT grossly underestimates void growth at low doses and low dislocation densities [2]. The second case, which is the hallmark of the Production Bias Model (PBM) [3], accounts for high swelling rates observed experimentally at small irradiation doses. But with the assumptions and defect interactions considered so far, in every case it predicts complete saturation of void growth at high doses [4–6], which is not always observed experimentally.

This failing of the approximations used so far in the PBM, together with evidence from molecular dynamics (MD) studies that have shown that smaller SIA clusters, at least, do not adhere to strictly 1-D diffusion, has prompted investigation into the effects of deviations from strict 1-D motion. The two physically likely deviations from strict 1-D motion are compared to random isotropic diffusion in Fig. 1. Both deviations can be seen in MD simulations of motion of a 3 SIA cluster in α-Fe at 1000 K [7]. Both the Burgers vector (BV) changes, and the self-climb of SIA clusters are less important for large clusters. The BV changes occur with frequency of order $\exp(-\sqrt{n})$, while the transverse diffusion coefficient due to self-climb is of order $D_{cd}/n^{3/2}$, where n is the number of interstitials in the cluster, and D_{cd} is the dislocation core diffusion coefficient [8]. Thus for large clusters self-climb is the more important of the two effects.

Recent studies have investigated the kinetics of the SIA cluster-void reaction kinetics in the case of random BV changes [8,9]. In this study, we present a theory of

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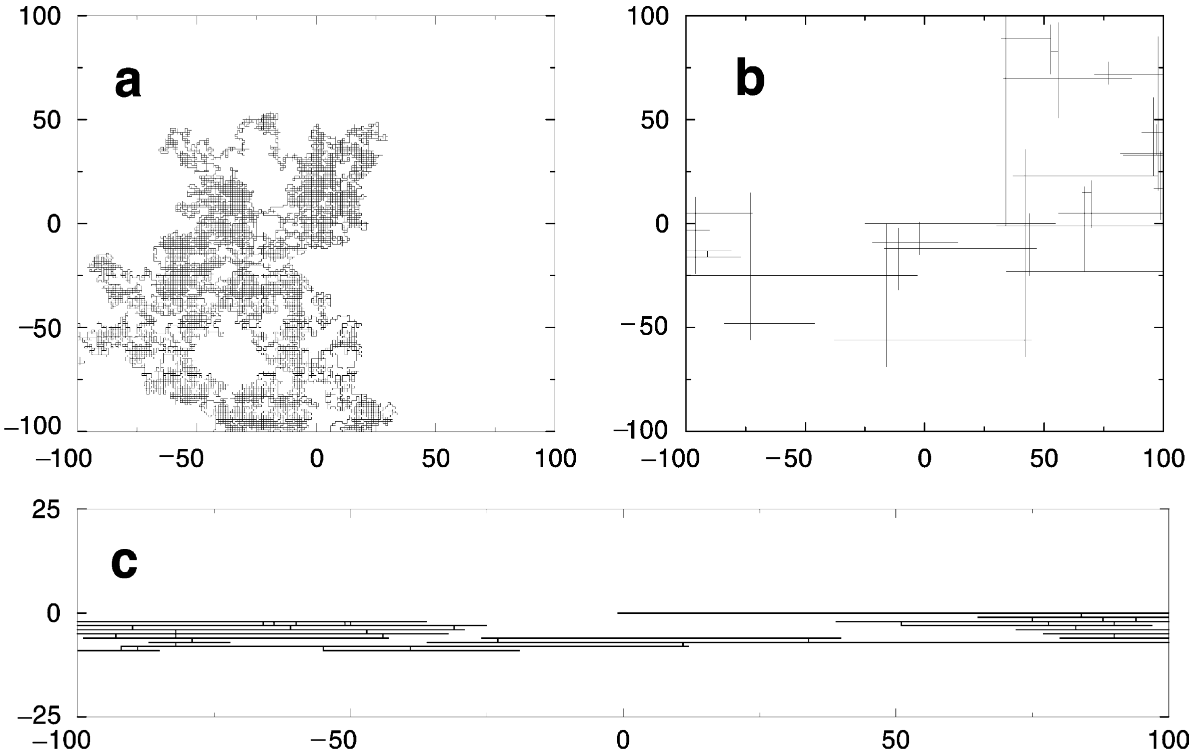

Fig. 1. Diffusion types on a 2-dimensional (2-D) square lattice: (a) random isotropic 2-D, (b) 1-D with rare changes in the direction, analogous to 1-D glide with rare BV changes, (c) strongly anisotropic, analogous to 1-D glide with rare self-climb.

void-size dependent swelling in the presence of anisotropically diffusing (self-climbing) SIA clusters.

2. The model

We model anisotropic diffusion of SIA clusters, with the diffusion coefficient in one axial direction greater than those in the two transverse directions, which are considered to be equal. Physically, if 1-D axial motion postulated by the PBM is considered to be the rapid diffusive motion of a SIA cluster in the form of a bundle of crowdions, then this model would allow an additional smaller component of SIA edge diffusion around the circumference of the SIA cluster, leading to self-climb and to the random variation of the transverse position of the centre of the cluster. Just as in PBM, there will be a population of these defects for each equivalent crystal direction of propagation. Mathematically, we represent the diffusion by

$$\frac{\partial C(\boldsymbol{\rho},z,t)}{\partial t} = D\left(\frac{\partial^2}{\partial z^2} + \epsilon\frac{\partial^2}{\partial \boldsymbol{\rho}^2}\right)C(\boldsymbol{\rho},z,t). \tag{1}$$

Here $C(\boldsymbol{\rho},z)$ represents the concentration of the diffusing interstitial clusters as a function of both the transverse $\boldsymbol{\rho}=(x,y)$, and axial z coordinates, D is the axial diffusion constant, and ϵ is the non-zero ratio between diffusion constants in each of the transverse directions, to that of the axial direction.

3. A single absorbing cavity

We begin by finding the steady state distribution of concentration around a stationary absorbing spherical cavity of radius a. In the steady state the concentration of cavities satisfies equation

$$D\left(\frac{\partial^2}{\partial z^2} + \epsilon\frac{\partial^2}{\partial \boldsymbol{\rho}^2}\right)C(\boldsymbol{\rho},z) = 0 \tag{2}$$

(subsequently represented by $\hat{L}C(\boldsymbol{\rho},z)=0$) outside the sphere, with boundary conditions $C(\boldsymbol{\rho},z)\,|_{|\boldsymbol{\rho}|^2+z^2=a^2}=0$ and $C(\boldsymbol{\rho},z)\,|_{|\boldsymbol{\rho}|^2+z^2=\infty}=C_\infty$, where C_∞ is the homogeneous SIA cluster concentration in the absence of the cavity.

Rescaling the z coordinate allows us to create a direct analogy with the homogeneous Poisson equation describing the distribution of the electrostatic potential around a charged conducting ellipsoid. The corresponding solution [10] is

$$C(\boldsymbol{\rho},z) = C_\infty - \frac{C_\infty \arctan\sqrt{\frac{2(1-\epsilon)a^2}{b^2}}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}},$$
$$b^2 = \epsilon z^2 + |\boldsymbol{\rho}|^2 - (1-\epsilon)a^2 + \sqrt{[(1-\epsilon)a^2 + \epsilon z^2 + |\boldsymbol{\rho}|^2]^2 - 4(1-\epsilon)a^2|\boldsymbol{\rho}|^2}. \quad (3)$$

Our subsequent treatment of an ensemble of randomly distributed cavities requires investigating mathematical properties of this solution. We note that when the operator $\hat{L}$ is applied to the concentration field given above, the resulting expression (representing the effective sink generating the concentration field) is identically equal to zero everywhere except for the surface defined by ($|\boldsymbol{\rho}|^2 \leqslant (1-\epsilon)a^2$) in the plane $z = 0$.

This surface is a circular disc of radius $\rho_0 = a\sqrt{1-\epsilon}$, which is smaller than the radius of the absorbing sphere. The disc is located at the centre of the sphere with its axis pointing in the direction of anisotropy. The disc degenerates to a point in the case $\epsilon = 1$ (corresponding to isotropic diffusion), and fills the cross section of the sphere when $\epsilon = 0$ (corresponding to strictly 1-D diffusion).

The distribution of sink strength is inhomogeneous across the surface of the disc, namely

$$\hat{L}C(x,y,z) = \frac{2D\sqrt{\epsilon}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}} \frac{\delta(z)\Theta(\rho_0^2 - x^2 - y^2)}{\sqrt{\rho_0^2 - x^2 - y^2}} C_\infty, \quad (4)$$

where $\Theta(\rho_0^2 - x^2 - y^2) = 1$ for $x^2 + y^2 \leqslant \rho_0^2$ and $\Theta(\rho_0^2 - x^2 - y^2) = 0$ for $x^2 + y^2 > \rho_0^2$. This gives us a radially symmetric disc, with a very high absorption strength at the edges of the disc compared to the centre of the disc. For comparison, in the case of isotropic diffusion the distribution of sink strength associated with an absorbing cavity is given by a delta-function located at the centre of the cavity.

4. Many absorbing cavities

Using Eq. (4) we consider how a concentration field of SIA clusters is affected by a random distribution of absorbing cavities. We now attempt to evaluate the concentration field of SIAs that does not correspond to a specific configuration of absorbing cavities, but instead represents the result of statistical ensemble averaging over all the possible configurations of positions of cavities. The averaging over positions of the centres of cavities uses the statistical scattering formalism [5,11]:

$$\langle C(\mathbf{r})\rangle = C_\infty(\mathbf{r}) + \int \mathrm{d}\mathbf{R}_a n(\mathbf{R}_a)\,\mathrm{d}\mathbf{R}\,\mathrm{d}\mathbf{r}'\, G_f(\mathbf{r},\mathbf{R}) \times T(\mathbf{R}-\mathbf{R}_a, \mathbf{r}'-\mathbf{R}_a)\langle C(\mathbf{r}')\rangle. \quad (5)$$

Here we introduced a quantity analogous to the T matrix of the theory of scattering:

$$T(\mathbf{r}-\mathbf{R}_a, \mathbf{r}'-\mathbf{R}_a) = \delta(\mathbf{r}'-\mathbf{R}_a) \times \frac{2D\sqrt{\epsilon}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}} \frac{\delta(z-z)'\Theta(\rho_0^2 - (x-x')^2 - (y-y')^2)}{\sqrt{\rho_0^2 - (x-x')^2 - (y-y')^2}}. \quad (6)$$

The Green's function $G_f(\boldsymbol{\rho},z)$ describing anisotropic propagation of SIA clusters through a crystal free of cavities is defined by equation $\hat{L}G_f(\boldsymbol{\rho},z) = \delta(\boldsymbol{\rho})\delta(z)$.

We operate on Eq. (5) with the anisotropic operator $\hat{L}$ and obtain

$$\hat{L}\langle C(\mathbf{r})\rangle = \hat{L}C_\infty(\mathbf{r}) + \int \mathrm{d}\mathbf{R}_a n(\mathbf{R}_a)\,\mathrm{d}\mathbf{R}\,\mathrm{d}\mathbf{r}' \times \delta(\mathbf{r}-\mathbf{R})T(\mathbf{R}-\mathbf{R}_a, \mathbf{r}'-\mathbf{R}_a)\langle C(\mathbf{r}')\rangle. \quad (7)$$

Taking into account the definition of the T matrix, we see that this equation is equivalent to

$$\hat{L}\langle C(\mathbf{r})\rangle = \hat{L}C_\infty(\mathbf{r}) + \frac{2D\sqrt{\epsilon}n(\mathbf{r})}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}} \times \int \mathrm{d}x'\,\mathrm{d}y' \frac{\Theta(\rho_0^2 - (x-x')^2 - (y-y')^2)\langle C(x',y',z)\rangle}{\sqrt{\rho_0^2 - (x-x')^2 - (y-y')^2}}. \quad (8)$$

The integration in the latter equation is performed over the surface of the disc, the size of which is many times smaller than the characteristic scale of variation of the statically averaged concentration field $\langle C(x,y,z)\rangle$. We can therefore treat the average concentration field as approximately constant over the entire range of integration. Using cylindrical polar coordinates centred about (x,y), we obtain

$$\left[\hat{L} - 4\pi D a n(\mathbf{r}) \frac{\sqrt{\epsilon}\sqrt{1-\epsilon}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}}\right]\langle C(\mathbf{r})\rangle = \hat{L}C_\infty(\mathbf{r}). \quad (9)$$

At this point, we include the effect of absorption of interstitial clusters by dislocations. Dislocations are extended linear objects and the influence of anisotropy of diffusion on the rate of absorption by dislocations is likely to be less significant than it is on the absorption by voids. In our treatment we neglect the effect of anisotropy on the absorption by dislocations. We take the mean-field homogeneous sink strength due to dislocations as ρZ_i, where ρ is the dislocation density, and Z_i is the dislocation bias factor which occurs in SRT. More precise evaluation of this term will be necessary for a fully quantitative study based on our model

$$\left[\hat{L} - 4\pi Dan(\mathbf{r})\frac{\sqrt{\epsilon}\sqrt{1-\epsilon}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}} - D\rho Z_i\right]\langle C(\mathbf{r})\rangle = \hat{L}C_\infty(\mathbf{r}). \tag{10}$$

To proceed further, we need to find the average Green's function describing the propagation and absorption of SIAs in a random distribution of cavities. We are looking for the average steady state distribution of the anisotropically diffusing particles generated by a unit point source situated at $\mathbf{r}''$. The Green's function satisfies equation

$$\left[\hat{L} - 4\pi Dan(\mathbf{r})\frac{\sqrt{\epsilon}\sqrt{1-\epsilon}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}} - D\rho Z_i\right]\langle G(\mathbf{r},\mathbf{r}'')\rangle = -\delta(\mathbf{r}-\mathbf{r}''), \tag{11}$$

the solution of which is

$$\langle G(\mathbf{r},\mathbf{r}'')\rangle = \frac{\exp\left(-l(\mathbf{r},\mathbf{r}'')\sqrt{\frac{4\pi an\sqrt{\frac{1-\epsilon}{\epsilon}}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}} + \frac{\rho Z_i}{\epsilon}}\right)}{4\pi\sqrt{\epsilon}Dl(\mathbf{r},\mathbf{r}'')}, \tag{12}$$

$$l(\mathbf{r},\mathbf{r}'') = \sqrt{(x-x'')^2 + (y-y'')^2 + \epsilon(z-z'')^2}.$$

This Green's function has the shape of a prolate spheroid, and in the limit of highly anisotropic diffusion its extends to a length longer than the characteristic distance between absorbing cavities in that particular direction. The limit of highly anisotropic diffusion requires going beyond the mean-field treatment, and this condition defines the range of validity of the treatment based on statistical theory of scattering. Because of this, our subsequent mean-field results cannot be applied quantitatively to systems with extreme anisotropy of cluster diffusion.

5. Void swelling

Assuming that the concentration of cavities in the material is sufficiently low so that anisotropic perturbations of the concentration field associated with different cavities do not overlap, we move closer to our eventual aim of finding the rate of growth of absorbing cavities in the presence of constant homogeneous irradiative generation of vacancy-interstitial pairs. We consider a specific configuration of cavities at $\mathbf{r}_a$, $\mathbf{r}_b$, etc. Denoting by a_0^3 the volume that one extra vacancy contributes to the size of a cavity, we obtain an expression for the flux of particles going into cavity α in the form of an integral over the surface area of the cavity

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\frac{4}{3}\pi a_\alpha^3}{a_0^3}\right) = \oint \mathbf{J}(\mathbf{r})\,\mathrm{d}\mathbf{S}_\alpha. \tag{13}$$

Using Gauss' theorem, we transform this integral into an integral over the volume of the cavity V:

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\frac{4}{3}\pi a_\alpha^3}{a_0^3}\right) &= \int_V \mathrm{div}(\mathbf{J}(\mathbf{r}))\,\mathrm{d}\mathbf{r} = \int_V \hat{L}C(\mathbf{r})\,\mathrm{d}\mathbf{r} \\ &= \int_V \hat{L}C_\infty(\mathbf{r})\,\mathrm{d}\mathbf{r} + \int_V \mathrm{d}\mathbf{r}\sum_a \int \mathrm{d}\mathbf{R}\,\mathrm{d}\mathbf{r}' \\ &\quad \times \hat{L}G_f(\mathbf{r},\mathbf{R})T(\mathbf{R}-\mathbf{R}_a,\mathbf{r}'-\mathbf{R}_a)C_a(\mathbf{r}'),\end{aligned} \tag{14}$$

where the inner integration is performed over entire space and C_a stands for the concentration field that would occur if the cavity a was absent.

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\frac{4}{3}\pi a_\alpha^3}{a_0^3}\right) &= \int \hat{L}C_\infty(\mathbf{r})\,\mathrm{d}\mathbf{r} + \int \mathrm{d}\mathbf{r}\sum_a \int \mathrm{d}\mathbf{R}\,\mathrm{d}\mathbf{r}' \\ &\quad \times \delta(\mathbf{r}-\mathbf{R})T(\mathbf{R}-\mathbf{R}_a,\mathbf{r}'-\mathbf{R}_a)C_a(\mathbf{r}').\end{aligned} \tag{15}$$

Since the volume fraction of absorbers is assumed to be small, defects cannot be produced by cascades inside the cavity and the first term can be neglected. Also, noting the fact that all scattering matrices apart from T_α are identically equal to zero inside the volume of integration V, we find that the summation reduces to only one term, the contribution from the absorber α

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\frac{4}{3}\pi a_\alpha^3}{a_0^3}\right) &= \int_V \mathrm{d}\mathbf{r}\int \mathrm{d}\mathbf{r}'\,T(\mathbf{r}-\mathbf{R}_\alpha,\mathbf{r}'-\mathbf{R}_\alpha)C_\alpha(\mathbf{r}') \\ &= \int \mathrm{d}\mathbf{r}'\,T(\mathbf{R}_\alpha,\mathbf{r}')C_\alpha(\mathbf{r}').\end{aligned} \tag{16}$$

At this point we substitute $C_a(\mathbf{r}) = \int \mathrm{d}\mathbf{r}''\,G(\mathbf{r},\mathbf{r}'')K(\mathbf{r}'')$, where $K(\mathbf{r}'')$ corresponds to the net rate of generation of SIAs per unit volume. This rate describes the generation of defects by irradiation cascades after any initial recombination has occurred.

The difference between absorption rates of vacancies and interstitials governs the net growth of voids. The total number of vacancies and interstitials created in the material per unit time is the same. The defects are generated in close proximity relative to the diffusive scale we are looking at, so $K_i(\mathbf{r}'') = K_v(\mathbf{r}'')$. By making the mean-field approximation, using our average Green's function as an approximation to the exact Green's function for a given configuration, and treating all voids as equally sized, we arrive at a relation for the swelling rate:

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\frac{4}{3}\pi a^3}{a_0^3}\right) &= \int \mathrm{d}\mathbf{r}\,\mathrm{d}\mathbf{r}'\,\mathrm{d}\mathbf{r}''\,(T_v(\mathbf{r},\mathbf{r}')\langle G_v(\mathbf{r}',\mathbf{r}'')\rangle \\ &\quad - T_i(\mathbf{r},\mathbf{r}')\langle G_i(\mathbf{r}',\mathbf{r}'')\rangle)K(\mathbf{r}'').\end{aligned} \tag{17}$$

We treat the vacancies as diffusing isotropically which means that the first term of Eq. (17) is equal to

$$\frac{4\pi aK}{\rho Z_v + 4\pi na}. \tag{18}$$

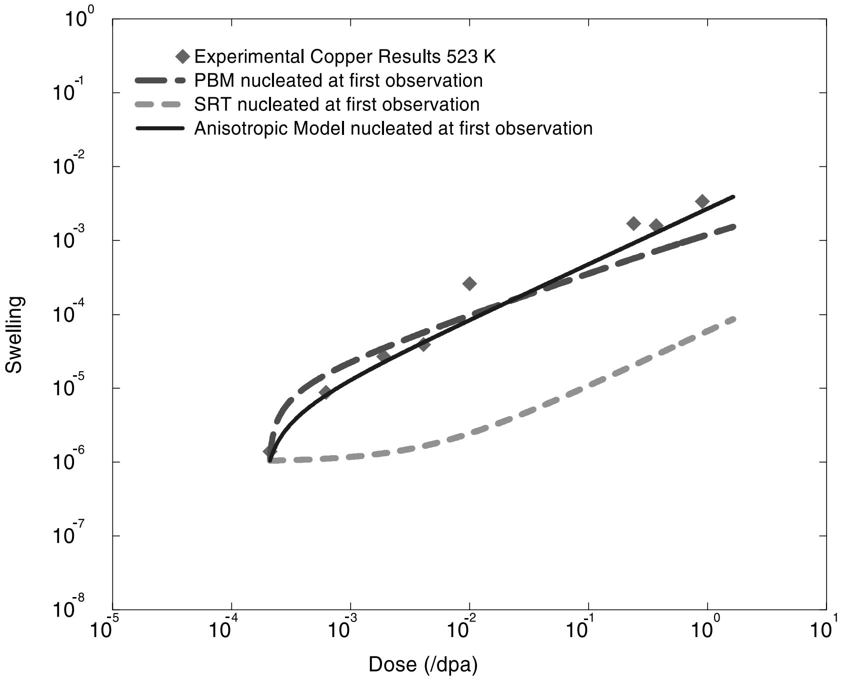

Fig. 2. A comparison of swelling between experiment and the three models discussed in this paper. Theoretical void growth was initiated at the point of the first experimental observation. A typical value of the dislocation bias of 0.02 was used. Parameters varied for a good fit: in this model $\epsilon = 0.01$, and in the PBM the dislocation capture diameter is 4 nm.

For the anisotropically diffusing interstitial clusters, we get the second term of Eq. (17) equal to

$$\frac{K}{2\pi \arctan\sqrt{\frac{1-\epsilon}{\epsilon}}}\int \mathrm{d}\mathbf{r}'\,\mathrm{d}\mathbf{r}''\frac{\delta(z')\exp\left(-Bl(\mathbf{r}'',\mathbf{r}'')\right)}{l(\mathbf{r}',\mathbf{r}'')\sqrt{\rho_0^2-x'^2-y'^2}}, \qquad (19)$$

$$B=\sqrt{\frac{4\pi an\sqrt{\frac{1-\epsilon}{\epsilon}}}{\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}}+\frac{\rho Z_i}{\epsilon}}.$$

Carrying out the integration, we find this equal to

$$\frac{4\pi aK}{4\pi an+\frac{\rho Z_i\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}}{\sqrt{1-\epsilon}\sqrt{\epsilon}}} \qquad (20)$$

and the growth rate is therefore given by

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\frac{\frac{4}{3}\pi a^3}{a_0^3}\right)=4\pi aK\left(\frac{1}{4\pi an+\rho Z_v}-\frac{1}{4\pi an+\frac{\rho Z_i\arctan\sqrt{\frac{1-\epsilon}{\epsilon}}}{\sqrt{1-\epsilon}\sqrt{\epsilon}}}\right). \qquad (21)$$

6. Results and comparison

Eq. (21) predicts that unsaturated void growth occurs at any finite value of dislocation bias.

We examine growth in the limits of small and large voids. For small a, the dose dependence of volume swelling is given by $a^3 \sim t^{3/2}$. Even for large a, we obtain the dose dependence of volume swelling of the form $a^3 \sim t^{3/4}$. This sublinear dependence does not show saturation in the limit of large t and is similar to the dependence observed experimentally.

Fig. 2 shows a comparison between experimental data and the three models discussed in this paper. The experimental results are a compilation [12] of swelling data from copper irradiated by neutrons at 523 K [2,13–16]. As well as predicting the continued lack of saturation in growth of voids even at high doses, this anisotropic model produces a good fit with experimental swelling results.

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