A General Treatment of One- to Three-Dimensional Diffusion Reaction Kinetics of Interstitial Clusters: Implications for the Evolution of Voids

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A General Treatment of One- to Three-Dimensional Diffusion Reaction Kinetics of Interstitial Clusters: Implications for the Evolution of Voids

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Abstract

In recent years, it has been shown that a number of striking features in the microstructural evolution occurring in metals under cascade damage generating irradiation (e.g. enhanced swelling near grain boundaries, decoration of dislocations with SIA loops, saturation of void growth and void lattice formation) can be rationalised in terms of intra-cascade clustering of vacancies and self-interstitial atoms (SIAs), differences in the thermal stability and mobility of the resulting clusters and one-dimensional (1D) glide diffusion of SIA clusters ("production bias model").

The 1D diffusion of SIA clusters is generally disturbed by changes between equivalent 1D diffusion paths and by transversal diffusion by self-climb, resulting in diffusion reaction kinetics between the 1D and 3D limiting cases. In this paper, a general treatment of such kinetics operating in systems containing random distributions of sinks is presented. The complicated partial sink strengths of different components of the system for the annihilation of SIA clusters are expressed by those for the simple 1D and 3D limiting cases. The effects of direction changes and transversal diffusion are first considered separately and are then combined. The significance of the present treatment for damage accumulation under cascade damage conditions is illustrated by applying it to the discussion of void growth characteristics, particularly of the conditions for saturation of void growth.
Quantities and Symbols

\( \mathbf{b} \): Burgers vector of dislocation loop, magnitude \( b \)
\( c \): atomic concentration
\( c_n \): concentration of mobile defects of type \( n \)
\( D \): diffusion tensor of defects (SIA clusters)
\( \bar{D} = D_{lo} / 3 \): effective 3D diffusion coefficient of (SIA clusters)
\( D_{lo} \): longitudinal (glide) diffusion coefficient (for SIA clusters \( \approx 10^{-8} \) m\(^2\)/s)
\( D_{cd} \): self-diffusion coefficient along dislocation core
\( d \): glissile loop absorption diameter of the dislocations
\( E_d \): diffusion energy of small glissile loops
\( I \): total defect flux to sink
\( j \): defect flux density
\( k_{1/3}^2 \): sink strength for 1D/3D diffusing defects
\( k_n^2 \): sink strength for the absorption of defects of type \( n \)
\( k_{i,n}^2 \): partial strengths of sink type \( i \) for the absorption of defects of type \( n \)
\( k_{v,d,i}^2 \): partial sink strengths voids (\( v \)) / dislocations (\( d \)) for the absorption of SIA clusters (cl)
\( k_B \): Boltzmann’s constant
\( l_{ch} = \sqrt{2D_{lo} \tau_{ch}} \): mean 1D diffusion length covered between two direction changes
\( \tau_n = 1/\bar{D}k_n^2 \): mean life-time of defects of type \( n \)
\( N \): number density of defects (voids)
\( p = Z_{i,cl}^n - 1 \): dislocation bias for the absorption of 3D migrating SIAs and SIA clusters
\( P_n \): production rate of mobile defects of type \( n \)
\( R \): radius of spherical sink (void)
\( R_i \): radius of sink of type \( i \)
\( R_s \): saturation size where voids cease to grow
\( T \): temperature
\( T_m \): melting temperature
\( x \): coordinate in 1D diffusion direction
\( x \): independent variable of “master curve”, “disturbance of 1-D diffusion”,
\( y \): dependent variable of “master curve”, sink strength normalised to its value for pure 1D, \( k^2/k_{(1)}^2 \),
\( Z_{i,v}^{v,d} \): efficiency factors for absorption of SIAs and vacancies (i,v) by voids and dislocations (v,d)
\( Z_d = 1 + p_d \): efficiency factor for the absorption of 3D migrating SIA clusters by dislocations
\( \delta = D_u/D_{lo} \): ratio of transversal to longitudinal diffusion coefficients
\( \varepsilon \): fraction of SIAs produced in glissile clusters
\( \kappa = \lambda^{-1} \): reciprocal mean free path of 1D diffusing defects
\( \kappa_1 \): partial contribution of sink to \( \kappa \)
\( \lambda_n = k_n^{-1} \): mean diffusion range of defect of type n
\( \lambda_1 = (\sigma N)^{1/2} \): mean 1D diffusion range
\( \mu \): shear modulus
\( \rho \): dislocation density
\( \sigma \): cross section for absorption of 3D diffusing defects
\( \tau_{ch} \): average time between two direction changes; \( \approx \) for staying in certain defect drainage channel.
\( \tau_n = 1/\overline{D k_n^2} \): mean life-time of defects of type n
\( \tau_{lo/tr} \): average time of a defect between entering and leaving drainage channel by longitudinal/transversal diffusion
\( \Omega \): matrix atom volume
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1. Introduction

The diffusion of mobile lattice defects and their reactions with other mobile or immobile defects are necessary prerequisites for changes in the chemistry and microstructure and the associated macroscopic properties of crystalline solids. The production of defects such as vacancies and self-interstitial atoms (SIAs) in metals under irradiation, particularly with energetic ions or neutrons, enhances and enriches the diffusion reaction kinetics and the resulting evolution of the microstructure substantially. Unfortunately, these irradiation induced changes in the microstructure are associated mainly with undesired changes in macroscopic material properties such as void swelling, loss of ductility and embrittlement which can seriously reduce the service lifetime of structural components in fission and fusion reactors.

Defect accumulations evolving in pure metals under irradiation with energetic particles such as neutrons or ions which produce displacement cascades have been found at low displacement doses (up to about 1dpa) and at temperatures around 0.4T\textsubscript{m} (T\textsubscript{m}: melting temperature) to be highly heterogeneous and segregated: SIAs segregate in the form of dislocation loops in regions close to (grown-in) dislocations which may result in the formation of dislocation walls, while vacancies accumulate at very high rates in the form of voids in between (Singh and Leffers, 1981, Singh et al, 1986) and, even enhanced, in several μm wide regions adjacent to grain boundaries (for review see Foreman et al, 1987). The characteristic length scales of such features (distance between grown-in dislocations and dislocation walls, width of swelling zones near grain boundaries) is of the order of several μm. There is clear experimental evidence that details in the microstructural evolution depends on the crystal structure (Singh and Evans 1995, Golubov et al 2000). In BCC metals, for instance, common features in the evolution of voids at high doses (> 1dpa) is that they cease to grow and form superlattices of strikingly high perfection, whereas in FCC these features, if observed at all, are less well developed. There appears to be a general trend of the microstructure to change with increasing dose, increasing temperature and increasing alloying from extremely heterogeneous towards more homogeneous.

Since the 1960s, defect accumulation and its consequences were commonly treated in terms of a chemical rate theory approach where the following main assumptions were made (Brailsford and Boullough, 1972, 1981): (a) under irradiation, both vacancies and SIAs are solely produced randomly in space and time in the form of mono-defects, (b) they perform three-dimensional (3D) diffusion, which allows them to recombine, to cluster and to react with extended defects such as dislocations and voids; and (c) clusters of both types
of mono-defects (except di- and tri-interstitials) are immobile. It has been shown however by Leffers et al. (1986) that the highly heterogeneous and segregated defect accumulation, i.e. dislocation wall formation and fast void evolution in between, as, for instance, observed in neutron irradiated copper (Singh and Leffers, 1981, Singh et al., 1986), cannot be explained in terms of this approach without making a number of unreasonable assumptions.

Already in the 1950s, it was recognized that high recoil energies transmitted from energetic particles to matrix atoms (> 1 keV) would initiate displacement cascades. According to the picture suggested by Seeger (1958), such a cascade results in a vacancy rich core which may collapse to form a vacancy cluster, and in SIAs which were considered to leave the cascade region by replacement sequences or by thermally activated random migration. Even though results of X-ray scattering experiments on neutron irradiated samples had indicated already since the beginning of the 1970s that not only vacancies but also SIAs are able to cluster in the cascade region (Peisl and Trinkaus 1973, von Guerard et al. 1980), this important feature of cascade damage was recognized seriously by the radiation damage community only at the beginning of the 1990s when it was confirmed by Molecular Dynamics (MD) (English et al. 1990; Diaz de la Rubia and Guinan 1990, 1991). It is interesting to note that in the same time period it was shown independently by analytical calculations that SIAs produced in cascades are likely to form clusters (Woo et al., 1990).

In the temperature range of void swelling, SIA clusters are stable while small vacancy clusters decay into single vacancies. Woo and Singh (1990, 1992) introduced this asymmetry in the concurrent production of stable SIA clusters and unstable vacancy clusters in cascades as a “production bias” representing a potential driving force for void swelling. It was soon recognised by Singh and Foreman (1992) that this driving force can only be maintained if a substantial fraction of SIA cluster is able to escape to remote sinks other than voids, and it was shown by Trinkaus et al. (1992, 1993) that one-dimensional (1D) diffusion-like glide of planar SIA clusters in the form of small perfect dislocation loops represents an efficient and sufficiently long ranging SIA removal mechanism (up to several μm) suited to rationalise the highly heterogeneous and segregated defect accumulation occurring under cascade damage conditions. Since then, the idea of 1D diffusion of SIA clusters has been confirmed by many molecular dynamics (MD) simulations (English et al. 1990, Foreman et al. 1992, Stoller et al. 1997, Soneda and Diaz de la Rubia 1998, Osetsky et al. 2000, Wirth et al. 2000, 2003). It is worth mentioning here that already in 1955 Lomer and Cottrell considered 1D diffusion of single SIAs (in the so-called “crowdion” configuration) to play an important role in defect recovery, an idea which was worked out in detail in the 1960s and 70s.
by Seeger and his co-workers (Frank et al 1965, Gösele and Seeger 1976, for a review, see Gösele, 1984).

During the last decade, the “production bias model”, extended by including the 1D diffusion of SIA clusters, was further worked out and applied to explain several features in the microstructural evolution and their dependence on the main controlling parameters (Trinkaus et al 1993, Singh et al 1997, Golubov et al (2000, 2001) Trinkaus et al 2000). It was, for instance, shown that the several μm large width of swelling zones near grain boundaries correlates well with the large 1D diffusion range, both decreasing in the same way with increasing dose (Trinkaus et al 1993). The decoration of dislocations with SIA type loops was modelled in terms of the trapping of 1D diffusing SIA clusters in the strain field of dislocations (Trinkaus et al 1997a,b), and its role in the post-irradiation deformation behaviour of metals was studied (Singh et al 1997, Trinkaus et al 1997b). Very recently, the plastic deformation of Cu samples under the concurrent production of cascades and dislocations as observed in a new type of in-reactor tensile tests (Singh et al 2004) was modelled in terms of the dynamic decoration of dislocations by 1D migrating SIA loops (Trinkaus and Singh 2008). Furthermore, the recoil energy dependence (Singh et al, 2000, Golubov et al 2000b) and the grain size dependence of void swelling were analysed in detail (Singh et al 2002).

So far, ideal 1D diffusion was assumed in the reaction kinetics (RK) of SIA clusters. It is, however, known, that self-diffusion along the core of dislocation segments becomes important at temperatures above 0.3Tm by which dislocation loops could perform 2D conservative self-climb diffusion transversal to the 1D diffusion direction (Johnson 1960, Eyre and Maher 1971). Moreover, some MD simulations indicate that 1D diffusing SIA clusters are able to change between crystallographically equivalent close-packed directions of the crystal lattice (Foreman et al 1992, Soneda and Diaz de la Rubia 1998, Osetsky et al 2000). Both transversal diffusion and direction changes would disturb the pure 1D RK of SIA clusters. Increasing rates of these processes must be expected to induce a continuous transition from the 1D to the 3D limiting cases.

In fact, various aspects of the microstructural evolution are difficult to understand in terms of pure 1D RK of SIA clusters: for instance its dependence on crystal structure and temperature, its tendency to change with increasing alloying from highly heterogeneous towards more homogeneous, the saturation of void growth in once formed void super-lattices.

Reaction kinetics of defects performing anisotropic diffusion without changing the direction of fastest diffusion was discussed in detail previously by Seeger and his co-workers (for a review, see Gösele, 1984). 1D reaction kinetics disturbed by occasional changes between equivalent 1D diffusion
direction has been analysed only recently by Barashev et al (2001) in terms of the average lifetime of the diffusing defects in a random distribution of sinks. Subsequently, Trinkaus et al (2002) have treated this case in terms of a self-consistent sink embedding procedure, and derived an analytical single-variable function ("master curve") interpolating between the 1D and 3D limiting cases which is in excellent agreement with kinetic Monte Carlo simulations (Heinisch et al 2007).

In the present work, we present a treatment of general 1D to 3D RK of SIA clusters induced by 1D direction changes and (2D) diffusion transversal to the 1D diffusion directions. In the section §2, we describe the main processes, i.e. 1D glide diffusion, direction change and transversal diffusion. In section §3 devoted to 1D to 3D RK, we first treat the effect of changes between equivalent 1D diffusion directions on the RK with fixed sinks. Then we consider the effect of diffusion transversal to the 1D diffusion direction, applying for high sink densities and/or high diffusion anisotropy (a limiting case for which the approximations used in the classical treatment by Seeger and co-workers fail) a treatment analogous to that applied for 1D direction changes. In the following, a generalisation including both 1D direction changes and transversal diffusion is discussed. In the last section §3, the treatment is extended to the more general case where more than one mobile defect performing 3D diffusion are involved. In §4, an application of the theory to some aspects in the evolution of voids typical for 1D to 3D reaction kinetics is presented. Finally, the work is summarised and an outlook on future applications is given in section 5.
2. Mechanisms Controlling Diffusion of SIA Clusters

Before the early nineties, it was only the strikingly heterogeneous microstructure evolving in pure metals under cascade damage conditions which could have provided indirect hints on the 1D diffusion of an important defect involved in the defect kinetics, most likely of SIA clusters (Singh and Leffers, 1981, Leffers et al, 1986, Singh et al, 1986. Trinkaus et al, 1992, 1993). Since then, MD simulations of cascades have provided plenty of evidence for the 1D diffusion of SIA clusters in the form of perfect dislocation loops (or coupled crowdions) and, in addition, some indication of direction (Burgers vector) changes of small members of them (Osetsky et al, 2002; Osetsky et al, 2003; Bacon et al, 2003). MD simulations are, however, still limited to time periods below a few ns, and are thus not able to reveal processes requiring longer times. It was argued on the other hand that direction changes are necessary for the stability of void lattices (Golubov et al 2000).

A negative finding in MD simulations for the occurrence of a certain process, which could be relevant for defect reaction kinetics, may be used, however, to derive a lower bound estimate for the activation energy of that process, \( E_a \). Assuming, for instance, simulation times \( > 1 \) ns, pre-exponential time scales \( < 10^{12} \) s, and high simulation temperatures close to the melting temperature \( T_m \), one finds \( E_a > 7 k_B T_m \) (\( k_B \): Boltzmann’s constant).

Macroscopic (continuum) dislocation theory may be expected to provide useful guidelines for estimating the formation and migration energies of cascade induced SIA clusters in the form of dislocation loops, even though the sizes of such defects are clearly below the validity limit of this approach (Trinkaus et al, 1992, 1993; Wirth et al, 2000 ). In figure 1, the three main processes relevant for the diffusion reaction kinetics of SIA clusters in the form of loops are illustrated from the point of view of macroscopic dislocation theory (Trinkaus et al, 2000).

Figure 2.1a indicates random (diffusion-like) glide of a loop as a whole entity. Estimates of migration energies for this process on the basis of the Peierls energy per dislocation core atom (Trinkaus et al, 1992, 1993) yield values clearly below \( k_B T_m \) for loops containing less than 100 SIAs for both FCC and BCC metals (\( k_B T_m = 0.12 \) and 0.17 eV for Cu and Fe, respectively). Such naive upper bound estimates are consistent with results from MD simulations yielding about 0.2 \( k_B T_m \) for both FCC Cu and BCC Fe. Such simulations show, however, in addition, that this activation energy is virtually independent of cluster size, which has been attributed to the observation of apparently independent spatial fluctuations of individual crowdions within a
cluster of coupled crowdions even over a few atomic distances (Osetsky et al., 2003). The longitudinal glide diffusion coefficient has been found to be of the order of $D_{lo} \approx 10^{-8} \text{m}^2/\text{s}$, almost independent of cluster size and temperature.

Figure 2.1b indicates transversal diffusion of loops by random (conservative) self-climb due to dislocation core diffusion (Johnson, 1960; Eyre and Maher, 1971; Trinkaus et al., 1994). Accordingly, activation energies for this transversal diffusion mode may be considered to be comparable with those for self-diffusion along dislocations and grain boundaries which are in the range of 7 to 10 $k_B T_m$ (Neumann & Neumann, 1972). Thus, this process would be close to the limit of being observable in modern MD simulations. The transversal diffusion coefficient for a loop as a whole entity, $D_{tr}$, decreases with the number $n$ of SIAs per loop as $D_{tr} \sim D_{cd}/n^{3/2}$ where $D_{cd}$ is the dislocation core diffusion coefficient (Trinkaus et al., 1994). Because of the high longitudinal glide diffusion coefficient, the diffusion ratio $\delta = D_{tr}/D_{lo}$ is extremely small (even in the presence of impurities).

In MD simulations, changes in the glide direction of glissile SIA clusters have been observed only for very small clusters containing only a few ($\leq 4$) SIAs (Gao, 2000). In order to get an idea of the frequency of direction changes, we indeed depend in this case on crude estimates based on analytical dislocation theory or on the interpretation of experimentally observed defect features.

Figure 2.1c illustrates a possible mechanism for a Burgers vector (BV) change of a loop from the point of view of dislocation theory. The change in BV from $b_1$ to $b_2$ is considered to occur by the sweeping of a dislocation segment with the difference vector $\Delta b = b_2 - b_1$ across the loop area. Using the standard expression for the energy $E_d$ of a (edge) dislocation segment of length $\Delta l$ (Hirth and Lothe, 1992) and the estimate $\mu \Omega = 35 k_B T_m$, we find $E_d \approx 0.2 \mu \Delta l b^2 \approx 7(\Delta l b^2/\Omega ) k_B T_m$ where $\mu$ is the shear modulus and $\Omega$ is the atomic volume of the metal. Assuming $\Delta b$ to be a perfect BV and the total length change $\Delta l \approx 1 \text{nm}$, we estimate the energy change $\Delta E_d$ to be about 30 $k_B T_m$, which virtually rules out the occurrence of any direction change by the sweeping of a perfect dislocation segment during irradiation at temperatures $< 0.5 T_m$.

Substantially lower values for $\Delta E_d$ are obtained for a two-step-process consisting of a transition from an initially perfect to a faulted one and from there to another perfect configuration by the sweeping of appropriate partials across the loop area (Trinkaus et al., 1997b). Assuming, for instance, for a FCC lattice sweeping of a partial $\langle 1,1,2 \rangle /6$, we find a reduction of $\Delta E_d$ in comparison with the sweeping of a perfect dislocation segment by a factor of 3, i.e. down from 30 $k_B T_m$ to about 10 $k_B T_m$, meaning that this process may be important for defect reaction kinetics even though it can not be observed in MD simulations. An analysis of the decoration of dislocation by SIA loops...
under cascade damage conditions yields somewhat higher energies (Trinkaus et al, 1997a, 1997b). Note that the stacking fault energy required in this two-step-process is smaller than $k_B T_m$ and may therefore be neglected for loop sizes to be considered here even for metals with high stacking fault energy.

A qualitatively new feature in the kinetics of nm-scale loops compared to the conventional macroscopic dislocation dynamics is that, at intermediate temperatures of interest, $0.2 T_m < T < 0.5 T_m$, 1D glide as well as changes in the glide direction must be considered to occur by thermal activation.

So far, we have assumed that the crystal where SIA clusters glide, climb and change their direction are free of impurities. All three processes must be expected, however, to be significantly affected by the presence of impurities interacting with SIAs. The effect of impurities on these basic processes and on the associated RK of SIAs is discussed elsewhere (Trinkaus and Singh).
3. 1D to 3D Diffusion Reaction Kinetics

Damage accumulation in metals under cascade generating irradiation depends on the primary damage, mainly controlled by the recoil energy spectrum, as well as on the intrinsic properties and the associated RK of the defects produced. According to the preceding chapter, §2, SIA clusters produced in cascades in the form of small dislocation loops or coupled crowdions are able to diffuse by 1D random glide through the lattice, to change their 1D diffusion direction and to diffuse transversally to the 1D direction by 2D random self-climb. It is obvious that not only a qualitative understanding but also a quantitative description of the associated RK is required for modelling defect accumulation under cascade damage conditions.

In the following sections, we present a general treatment of the 1D to 3D RK of SIA clusters including 1D diffusion with direction changes as well as (2D) diffusion transversal to the 1D diffusion directions. Before a detailed discussion of the RK, we introduce the general framework for describing the reaction of mobile defects with fixed sinks.

3.1 Reaction of Mobile Defects with Fixed Sinks

3.1.1 General Framework

The aim of this subsection is to provide a basis for general RK between the 1D and 3D limiting cases considered in the following. In defining the relevant physical quantities it is useful to make reference to the customary 3D limiting case. Accordingly, we consider the spatial and temporal evolution of the concentration $c_n$ of mobile defects of type $n$, resulting from their random production at a given production rate $P_n$, their generally anisotropic diffusion in 3D characterized by a diffusion tensor $D$ with effective 3D diffusion coefficient $D$ and their diffusion limited reactions with randomly distributed immobile and quasi-stationary (constant) sinks of modest total strength $k_n^2$ (realized e.g. for small volume fraction of sinks, say < 5%). Reactions of the mobile defects with each other are ignored. In the mean field approximation, suited for treating global reaction kinetics, this problem is described by the linear diffusion reaction equation (see, for instance, Brailsford and Boullough, 1981)

$$\frac{\partial c_n}{\partial t} = P_n + \nabla \cdot \nabla c_n - D c_n k_n^2,$$  (3.1)
where \( D \) is the diffusion tensor of the defects and \( \overline{D} \) is an associated effective 3D diffusion coefficient described by an appropriate scalar representative of \( D \).

For strictly 1D diffusion, \( \overline{D} \) in equation (3.1) could be substituted by the (longitudinal) 1D diffusion coefficient \( D_{lo} \), the Laplacian \( \Delta \) by \( \partial^2 c/\partial x^2 \) for defects diffusing in \( x \)-direction and \( k^2 \) by an appropriate 1D sink strength. Considering, however, that, on a very large spatial scale of a sink-free virtual crystal, even almost perfect 1D diffusion would be effectively 3D with a 3D diffusion coefficient \( \overline{D} = D_{lo} / 3 \), we shall keep the form of equation (3.1) meaning that we choose for general anisotropic diffusion

\[
\overline{D} = \text{trace}(D)/3.
\]  

(3.2)

This definition implies that deviations of diffusion from 3D diffusion must be fully accounted for in the sink strength \( k_n^2 \), which is to be determined by some appropriate procedure. According to equation (3.1), \( k_n^2 \) is the key parameter for the evolution of the defect concentration \( c_n \). Thus, \( k_n^2 \) determines the characteristic spatial and temporal scales of the evolution, i.e. the mean diffusion range \( \lambda_n = k_n^{-1} \) and the mean life-time, \( \tau_n = 1/\overline{D}k_n^2 \), of the defects, respectively (appearing as constants in exponential solutions of equation (3.1)).

Generally, the system contains sinks of different type \( i \) with partial sink strengths, \( k_{i,n}^2 \), for the absorption of defects of type \( n \) such that the total sink strength is given by

\[
k_n^2 = \sum_i k_{i,n}^2.
\]  

(3.3)

We are interested here not primarily in determining the partial sink strengths of specific sinks in quantitative details, but in expressing them formally by their 1D and 3D limiting cases. Consequently, we refer to the shape of specific sinks only where needed.

### 3.1.2 1D Diffusion Reaction Kinetics Disturbed by Direction Changes

1D RK disturbed by changes between equivalent 1D diffusion directions has been treated recently by Barashev et al (2001) in terms of the average life-time of the diffusing defects in a random distribution of sinks and subsequently by Trinkaus et al (2002) in terms of a self-consistent sink embedding procedure. We describe the latter procedure in detail here, in order to prepare the one used later for treating 1D diffusion with slow transversal diffusion (high diffusion anisotropy) which has similar features.
For 1D diffusion of mobile defects with direction changes, the character of the RK depends on the relationship between the following three main types of length scales involved (Trinkaus et al 2000, 2002):

(1) the mean 1D diffusion length, \( l_{ch} = \sqrt{2D_0 \tau_{ch}} \), covered during the time between two direction changes, \( \tau_{ch} \), in a sink free virtual crystal, where \( D_0 \) is the 1D (longitudinal) diffusion coefficient;

(2) the mean 1D diffusion range in a certain microstructure, given, for instance, by \( \lambda_1 = (\sigma N)^{-1} \) for a mono-disperse random distribution of sinks of absorption cross section \( \sigma \) and number density \( N \);

(3) the linear dimension of the sinks, for instance the absorption radius \( R \) of spherical sinks.

The three physically meaningful relationships between these length scales define three characteristic cases in the RK as illustrated in figure 3.1:

(1) The pure (correlated) 1D case defined by for \( l_{ch} >> \lambda_1 \) (figure 3.1a). In this case, any given straight 1D diffusion line is terminated by two unambiguously defined individual sinks, which is associated with a two-sink correlation in the 1D RK (Gösele 1984, Trinkaus et al 1992, 1993 (see below).

(2) An intermediate case defined by \( \lambda_1 >> l_{ch} >> R \) (figure 3.1b). The correlation between sinks characteristic for pure 1D case is obviously broken by direction changes, but an important feature of 1D, the dependence of the kinetics on absorption cross sections rather than linear dimensions of the sinks is maintained. Therefore, we call this case “uncorrelated 1D RK” (see below).

(3) The well known 3D case defined by \( l_{ch} << R \) (figure 3.1c).

We first present a treatment including cases 1 and 2 defined by \( l_{ch} >> R \). To derive an expression for the sink strength \( k^2 \) for the absorption of the mobile defects, we assume steady-state and use the following self-consistent sink embedding procedure. We consider one individual sink of type \( i \) of radius \( R_i \) and cross section \( \sigma_i \) embedded into a homogeneous background of randomly distributed sinks of total strength \( k^2 \) which is to be defined by the rate of absorption of the mobile defects by all individual sinks.

As long as \( l_{ch} = \sqrt{2D_0 \tau_{ch}} >> R_i \), the considered sink defines \( 2n \) linear defect drainage channels, in the case of spherical sinks, for instance, cylinders (2 for each direction), in which defects diffusing in \( n \) crystallographically equivalent 1D directions flow towards the sink (\( n=6 \) for FCC crystals). In figure 3.2, this situation is illustrated in 2D for two perpendicular diffusion directions. Defects flowing towards the sink within a certain drainage channel will leave this after direction change, and defects diffusing transversal to it will join the
flux towards the sink after a proper direction change. Within a certain
drainage channel, only the concentration of the defects diffusing in it is
significantly affected (reduced) due to absorption by the sink whereas the
concentrations of the defects diffusing in other directions are virtually
unaffected for \( l_{\text{ch}} \gg R_i \). Furthermore, the average time for a defect to leave
the drainage channel after a direction change is negligibly small compared to
the stay time \( \tau_{\text{ch}} \).

Using these approximations, we may modify the steady state version of
equation (3.1) for the local concentration \( c_\mu(x) \) of defects of configuration \( \mu \)
produced at a constant partial production rate \( P_\mu \) and diffusing in the direction
\( \mu \) along the corresponding drainage channel of the individual sink considered
(chosen to be oriented in the direction of the \( x \)-co-ordinate)

\[
P_\mu = -3D\partial^2 c_\mu / \partial x^2 + Dk^2(c_\mu + \langle c_\mu \rangle) / 2 + (c_\mu - \langle c_\mu \rangle) / \tau_{\text{ch}}, \tag{3.4}
\]

where \( \langle c_\mu \rangle = c_\mu(x \to \infty) = P_\mu / Dk^2 \) is the average defect concentration far
away from the sink considered. The three terms at the right hand side of
equation (3.4) describe the diffusion of defects \( \mu \) along the drainage channel
(where \( D_{lo} = 3D \) has been used), their absorption by two types of sides of
sinks in the background, and their loss and gain (net gain) by direction
changes, respectively. In the second term, the orientation of the two sides of
the background sinks with respect to the individual sink considered must be
taken into account. Thus, the presence of the latter sink does only affect the
absorption of defects by the sides of the background sinks turned towards it
but not that by the sides turned away from it. According to this “shadowing
effect”, the defect concentration has to be split into local and distant parts,
\( c_\mu / 2 \) and \( \langle c_\mu \rangle / 2 \), respectively. It should be mentioned here that, if this
distinction were not made, the resulting sink strength would be incorrect by a
factor of 2 in the limiting case of pure 1D diffusion. The solution of equation
(3.4), subject to the boundary conditions \( c_\mu(x=0) = 0 \) and \( c_\mu(x \to \infty) \to \langle c_\mu \rangle \),
is

\[
c_\mu = (P_\mu / Dk^2)[1 - \exp(-k'x)], \tag{3.5}
\]

where \( k' \) is defined by \( 3k'^2 = k^2 / 2 + 6 / l_{\text{ch}} \). \tag{3.6}

The flux density defined by equation (3.5), \( j_\mu = -D_{lo} \partial c_\mu / \partial x \), yields for the
two-sided total flux \( I_i \) to a sink of type \( i \)

\[
I_i = 2|j(x = 0)| \sigma_i = 6(P_\mu / k^2)k' \sigma_i. \tag{3.7}
\]

In steady state, the self-consistency condition consists in equating the total
flux to all individual sinks \( I_i \) (equation (3.7), multiplied by the number density
of sinks of type \( i \), \( N_i \), and summed up over \( i \), with the production rate, which
yields:
\[ k^2 = 6k' \sum_i k_i', \quad \text{with } k_i = \lambda_i^{-1} = \sigma_i N_i. \]  

(3.8)

The form of equation (3.8) suggests that \( k_i^2 = 6k'k_i \) can be considered as the partial sink strengths of sinks of type \( i \). Eqs. (3.6) and (3.8) yield quadratic equations for \( k^2 \) and \( k' \), the solutions of which result in an expression for the partial sink strengths

\[ k_i^2 = 3k_i k[1 + \sqrt{(1 + 8/k_i^2 l_{ch}^2)}]. \]  

(3.9)

Expression (3.9) for \( k_i^2 \) is in excellent quantitative agreement with an expression in form of an integral obtained for the same problem by considering average defect lifetimes in a random distribution of sinks (Barashev et al. 2002). We emphasise here that our arguments for the procedure yielding equation (3.9) do not depend on the shape of the sink considered which may even be cylindrical as in the case of dislocations, defining planar plate-like drainage regions.

In equation (3.9) twice of the reciprocal of the function in the square brackets may be considered as a measure of the degree of diffusion correlation between two neighbouring sinks characteristic for the RK of 1D diffusing defects with fixed sinks. This correlation function decreases with decreasing \( l_{ch} \) from 1 for \( l_{ch} \to \infty \) and to 0 for \( l_{ch} \to 0 \). Note that this function depends, via \( k^2 \), only on the total sink structure but not on the partial contributions of its individual components.

In the two limiting cases of correlated (pure) and uncorrelated 1D diffusion, respectively, equation (3.9) simplifies to

\[ k^2 \to k^2_{(1)} = 6k_i k, \quad \text{for } l_{ch} \gg \lambda, \]  

(3.10)

and

\[ k^2 \to 6\sqrt{2}k_i / l_{ch}, \quad \text{for } \lambda \gg l_{ch} \gg R. \]  

(3.11)

Due to the two-sink correlation, the limiting case \( k^2_{(1)} \) for correlated 1D diffusion is quadratic in the sink density, establishing, together with the concentration of the mobile defects, RK of 3\textsuperscript{rd} order. The increase of \( k^2 \) with decreasing \( l_{ch} \) for \( \lambda \gg l_{ch} \gg R \) reflects the increase in the search-and-find efficiency with increasing frequency of direction changes in the 1D diffusion of the defects.

Even though equation (3.9) is restricted to a weak disturbance of the pure 1D diffusion by direction changes, \( l_{ch} \gg R \), it provides a basis for interpolating to the limiting case of 3D diffusion reached when \( l_{ch} << R \) and even for including transversal diffusion. This may be done by generalizing the time of 1D diffusion along a certain drainage channel, \( \tau_{ch} \), contained in the term \( l_{ch} \) in equation (3.4) and, via this, in equation (3.9). We first consider the transition to 3D diffusion by increasingly frequent direction changes.
In this case, we interpret $\tau_{ch}$ as the average time of a defect for staying in a certain drainage channel. When $R_i$ is no longer negligible compared to $l_{ch}$, the average time of a defect for entering and leaving the drainage channel before and after the period of 1D diffusion along it, respectively, $\tau_{lo} \propto R_i^2/D_{lo}$, would have to be included in the average stay time in addition to $\tau_{ch}$. This suggests that $\tau_{ch}$ in equation (3.9) should be substituted by ($\tau_{ch} + \tau_{lo}$). In the corresponding generalisation of equation (3.9), a numerical factors of the order of 1 in $\tau_{lo} \propto R_i^2/D_{lo}$ is to be determined such that the partial sink strength becomes exact, i.e. equal to $k_{(1)y}^4$, in the limiting case of 3D diffusion and low sink volume fraction, defined by $l_{ch} \rightarrow 0$ and $k_{(1)y}^4/k_{(3)y}^4 \rightarrow 0$, respectively. Using in $\tau_{lo} \propto R_i^2/D_{lo}$ that $R_i^2 \propto \kappa_i^2/k_{(3)y}^4$, we find that this requirement is fulfilled by setting $\tau_{lo} \propto 4\kappa_i^2/(D_{lo} k_{(3)y}^4)$. The resulting generalised version of equation (3.9) may then be written as a single variable function in the form of

$$y = 0.5 \left\{ 1 + \sqrt{1 + 4x^2} \right\} \quad \text{with}$$

$$y = k_i^2 / k_{(1)y}^2, \quad x^2 = \left( l_{ch}^2 k_{(1)y}^2 / 12 + k_{(3)y}^4 / k_{(3)y}^4 \right)^{-1}.$$  (3.12)

This function, plotted in figure 3.3, represents a single-variable master curve for the dependence of the sink strength on the three main spatial scales, $l_{ch}$, $\lambda_1$ and $R_i$, involved. Data obtained by Kinetic Monte Carlo simulations have been found to be in perfect agreement with this analytical master curve (Trinkaus et al 2002). We emphasise that we have chosen here as the independent variable x the reciprocal of the one chosen previously (Trinkaus et al 2002), since the present choice is more suited for modifying (and generalising) the master curve for the case of 1D diffusion disturbed by slow transversal diffusion (high diffusion anisotropy) as well as will be shown in the next subsections.

For the case of 1D diffusion disturbed by slow transversal diffusion, we may, however, anticipate the functional trends obtained by the rigorous treatment given below by applying an intuitive procedure analogous to that given above for the transition to 3D diffusion by increasingly frequent direction changes. For 1D diffusion disturbed by a slow transversal diffusion with diffusion component $D_{tr}$, we simply re-interpret the original meaning of the stay time $\tau_{ch}$ of 1D diffusion in the drainage cylinder as the characteristic time for gain and loss of defects by transversal diffusion, $\tau_{tr}$. For spherical sinks of radius $R_i$, for instance, this means substituting in the term $l_{ch}^2 = 2D_{lo} \tau_{ch}$, $\tau_{ch}$ by $\tau_{tr} \propto R_i^2/D_{tr}$, by which equation (3.11) is transformed to $k_i^2 \propto \sqrt{\delta} \kappa_i / R_i$ with $\delta = D_{tr}/D_{lo}$. This functional dependence of $k_i^2$ for uncorrelated 1D diffusion will be confirmed by the rigorous treatment given in the next subsection.
3.1.3 1D Diffusion Reaction Kinetics Disturbed by Transversal Diffusion

As discussed in §2, SIA clusters in the form of dislocation loops are generally able to diffuse by 1D random glide as well as by 2D random conservative climb. An increasing contribution of climb relative to that of glide would result in diffusion changing from the limiting case of 1D for negligible climb over 3D to the limiting case of 2D for sessile loops. Reaction kinetics of defects with such diffusion anisotropy was treated in detail already in the 1960s and 70s by Seeger and his co-workers (for a review, see Gösele, 1984). In the following, we shall apply the procedure of Gösele and Seeger (1976) in a modified form to adapt it to our purposes. We shall extend the approximations used in this treatment for small sink concentrations (and/or “moderate” diffusion anisotropy) to bridge the gap between the 1D and 3D limiting cases.

3.1.3.1 Moderate Diffusion Anisotropy

We consider anisotropic defect diffusion in the presence of sinks. Choosing the Cartesian coordinates parallel to the principal axes of a diffusion tensor with principal values $D_1$, $D_2$, $D_3$, we may write the components of the flux density vector as

$$ j_i = -D_i \frac{\partial c}{\partial x_i}. \quad (3.13) $$

In sink free regions, the requirement of defect conservation results in the anisotropic diffusion equation

$$ \frac{\partial c}{\partial t} = P - \sum_i j_i / \partial x_i = P + \sum_i D_{ij} \frac{\partial^2 c}{\partial x_i^2}. \quad (3.14) $$

A general solution of equation (3.14) would have to satisfy appropriate boundary conditions, for instance $c = 0$ at the boundaries of the sinks. In principle, the absorption efficiency of the sinks or the sink strength follows from the solution of equation (3.14) obeying the boundary condition by integrating the flux density according to equation (3.13) over all sink boundaries and comparing the result with the term $Dck^2$ in equation (3.1).

However, such a procedure is complicated by two different features of the problem:

(i) the diffusion anisotropy associated with the complicated partial differential equation

(3.14) with three second order partial derivatives, and
(ii) the presence of many sinks associated with a multitude of boundary conditions.

Problem (i) may, however be resolved, even though at the cost of the incorrect boundary conditions, by employing a spatial scaling transformation (sketched in figure 3.4 for moderate and high diffusion anisotropy where the meaning of “moderate” and “high” depends on the sink density)

\[ x_i^* = \left( \frac{D_{ii}}{\bar{D}} \right) x_i^* , \quad \bar{D} = (D_{11} D_{22} D_{33})^{1/3}, \quad (3.15) \]

by which equations (3.13) and (3.14) transform to the corresponding ones for isotropic diffusion,

\[ j_i^* = -\bar{D} \frac{\partial c}{\partial x_i^*} , \quad (3.16) \]

\[ \frac{\partial c}{\partial t} = P + \Delta c . \quad (3.17) \]

In equations (3.15 to (3.17), asterisk * denotes coordinates, flux components and Laplacian in the scaled system. The form of \( \bar{D} \) chosen in equation (3.15) guarantees volume and density conservation. For the stationary diffusion problem without defect production, equation (3.17) even reduces to the Laplace equation \( \Delta^* c = 0 \), investigated extensively in electrostatics. The shape of the sink considered changes, however, by the transformation. An originally spherical sink boundary, for instance, would transform to an ellipsoidal boundary as sketched in figure 3.4.

For the treatment of the multi-sink problem suitable approximate procedures are needed, such as the sink-embedding procedure used in section 3.1.2. As long as all sinks are well separated in the transformed coordinate system, i.e for “moderate” anisotropy and/or low sink density (figure 3.4a), a more suited procedures to reduce the multi-sink problem to a single-sink problem is (see Brailsford and Bouluogh 1972 and 1981) to consider the total defect flux to one individual sink, taking the presence of other sinks into account by either properly limiting the sink free region of influence around the individual sink (“cell approximation” with finite \( P \)) or by applying an appropriate remote boundary condition, for instance \( c(r \to \infty) \to c_\infty \) (assuming \( P = 0 \)).

In determining the sink strength by using the latter procedure in conjunction with the scaling transformation, the flux density according to equation (3.16) integrated over the transformed boundary of the sink considered must be compared with the term \( \bar{D} k^2 \) in equation (3.1). For sinks with 3D closed surfaces such as spherical sinks of number density \( N \), this yields

\[ k^2 = -N(\bar{D}/\bar{D}_c) \int \nabla^* c \, dA^* . \quad (3.18) \]
The right hand side of equation (3.18) represents, except for a factor of $N(D/\bar{D})$, the electrical capacitance of a capacitor with the shape of the transformed sink.

For glide-climb diffusion of loops, the shape of a spherical sink such as a void becomes a prolate rotational ellipsoid in the transformed system. Using the formula for the corresponding capacitance (Landau et al 1984, Gösele 1984) we may express the sink strength by that for isotropic diffusion, $k_{(3)}^2$, in the form

$$k^2 = \delta^{1/2} f_{c}(\delta) k_{(3)}^2,$$  \hspace{1cm} (3.19)

where $f_{c}(\delta)$ is a slowly varying function of $\delta$

$$f_{c}(\delta) = \frac{3\sqrt{1-\delta}}{(1+2\delta) \arccos \delta^{1/2}}.$$ \hspace{1cm} (3.20)

The limiting behaviour of $f_{c}(\delta)$ is \(\lim_{\delta \to 1} f_{c}(\delta) \to 1\) for $\delta \to 1$ (isotropy) and \(\lim_{\delta \to 0} f_{c}(\delta) \to 6/\pi\) for $\delta \to 0$ (high anisotropy). Equation (3.19) in conjunction with equation (3.20) confirms the main square root dependence of $k^2$ on $\delta$ derived in section 3.1.2. from the corresponding result for 1D diffusion with direction changes.

The procedure for linear defects such as dislocations is more complicated since in this case not only the scaling transformation of the shape of the reaction volume but also that of the originally 3D network (which becomes a 2D texture in the case of high diffusion anisotropy). The latter transformation is even more important than the former since the sink strength depends linearly on the three different densities in the transformed network but only logarithmically on the shape of the transformed reaction volume. The sink strength of dislocations can be approximated by an expression of similar form as given by equation (3.19) but with another slowly varying function of $\delta$

$$f_{dis}(\delta) = 2(1+\delta^{1/2}/2)/(1+2\delta).$$ \hspace{1cm} (3.21)

The limiting behaviour of $f_{dis}(\delta)$ is \(\lim_{\delta \to 1} f_{dis}(\delta) \to 1\) for $\delta \to 1$ (isotropy) and \(\lim_{\delta \to 0} f_{dis}(\delta) \to 2\) for $\delta \to 0$ (high anisotropy).

### 3.1.3.2 High Diffusion Anisotropy

When the distortion of the shape of the sinks by the scaling transformation becomes so strong that the transformed sinks are no longer well separated (figuratively spoken, when the “normal paste of the sinks” becomes a “puff pastry”) which occurs at high diffusion anisotropy and/or high sink density,
the electrostatic analogue of (single) sinks to (single) capacitors becomes useless. For this regime (which represents the transition to the limiting case of 1D diffusion), we employ a procedure similar to that used in the preceding section 3.1.2 for deriving an expression for the sink strength in the case of 1D diffusion disturbed by direction changes, i.e. we assume steady-state in the framework of a properly adjusted self-consistent sink embedding procedure.

We consider the absorption of mobile defects diffusing with high diffusion anisotropy by one individual sink of type $i$ with cross section $\sigma_i$ embedded into a homogeneous background of randomly distributed sinks of total strength $k^2$ which is to be defined by the rate of absorption of the mobile defects by all individual sinks. As long as $\delta \ll 1$, the considered sink defines two defect drainage regions (in the case of spherical sinks cylinders), one at both sides of the sink, oriented in the main diffusion direction of the defect configuration considered. The situation is similar to that sketched in figure 3.2 except that the coupling with the region outside the channel is now not due to abrupt direction changes but due to continuous transversal diffusion. This is an important qualitative difference since the concentrations of the defects outside the channel is virtually unaffected by the presence of sinks in the former case but is significantly disturbed in the close vicinity of the channel in the latter case. Consequently, in the latter case, the coupling of the diffusion fields inside and outside the drainage channel must be considered explicitly. Because of this coupling, the details in the procedure and the results are no longer independent of the shape of the sink.

In formulating the coupled diffusion problem, we refer to spherical sinks. In this case, defect production, diffusion and absorption within the drainage cylinder is described by an equation similar to equation (3.4), except for the last term accounting for defect gain and loss due to the coupling to the outside region. Choosing the axis of the drainage cylinder to be the z-coordinate of a cylindrical coordinate system $(r,z)$ we write for the region inside the cylinder

$$P = -D_n \frac{\partial^2 c}{\partial z^2} + \bar{D} k^2 (c + \langle c \rangle) / 2 + 2\pi j_r(R, z) \text{ (inside cylinder)}, \quad (3.22)$$

where $j_r(R, z)$ is the $z$-dependent flux density at $r \rightarrow R$ from outside the cylinder due to transversal diffusion. Outside the drainage cylinder, the diffusion flux is approximately radial. Accordingly, we write for this region

$$P = -D_n \Delta_r c + \bar{D} c k^2 \text{ (outside cylinder)}, \quad (3.23)$$

where $\Delta_r$ is the radial part of the Laplacian. Equation (3.23) defines the flux in the third term of equation (3.22) as

$$j_r(R, z) = -D_n c/\partial r (r \rightarrow R, z), \quad (3.24)$$

by which both equations are coupled.
The solution of the coupled equations (3.22) and (3.23) has to satisfy continuity at the boundary \( r = R \) and the remote boundary condition \( c(r \to \infty) \to \{c\} \).

The main steps in the procedure are: Find the formal solution of equation (3.23) containing the concentration inside the cylinder as a parameter, calculate from this the flux density to the cylinder, i.e. the third term in equation (3.22), then solve equation (3.22) and proceed as in the section 3.1.2. By this, an expression similar to equation (3.9) is obtained. The interpolation between this expression for high and that for moderate diffusion anisotropy may be written in the same form as in equation (3.11a) for 1D diffusion disturbed by direction changes, but with a different meaning of the independent variable \( x \)

\[
y = 0.5\left\{1 + \sqrt{1 + 4x^2}\right\}, \quad \text{with} \quad y = \frac{k_i^2}{k_{(i)j}^2}, \quad x = \frac{\delta^{1/2} f(\delta)k_{(i)j}^2}{k_{(i)j}^2}. \quad (3.25)
\]

According to equation (25), the sink strength for the absorption of defects performing 1D diffusion disturbed by transversal diffusion follows a “master curve” with the same form as given by equation (3.12) and shown in figure 3.3 for 1D diffusion disturbed by direction changes except that the independent variable \( x \) has now a different meaning. Figure 3.6 shows that also the present master curve given by equation (3.25) (solid line) is in excellent agreement with the results of kinetic Monte Carlo simulations (symbols) (Heinisch et al 2007).

### 3.1.4 Reaction Kinetics Including 1D Direction Changes and Transversal Diffusion

The 1D RK disturbed by the combined action of both 1D direction changes and transversal diffusion can be treated by including in the term for loss and gain of defects in equations (3.4) or (3.22) the contributions of both disturbances. The treatment which is already rather complicated for transversal diffusion, becomes substantially more complicated if both effects are included.

An expression for the sink strength in the general kinetics interpolating between the 1D and 3D limiting cases can, however, be directly derived from the corresponding expressions (3.12) or (3.25) by simply re-interpreting the original meaning of the defect stay times in the drainage cylinder (or the corresponding frequencies) including in them now defect loss and gain by both direction changes and transversal diffusion and by choosing the numerical factors properly, in a manner similar to what we have done in
section 3.1.2 for the transition from 1D to 3D. According to the physical meaning of defect gain by direction changes and transversal diffusion and their formal description by equations (3.4) and (3.22), the frequencies (reciprocal times) of both effects have to be added. This means that we need to substitute the reciprocal of $\tau_{ch}$ contained in $t_{ch} = \sqrt{2} D_{ch} \tau_{ch}$, and via this in equation (3.9) by

$$1/\tau_{ch} \to 1/\left(\tau_{ch} + \tau_{tr}\right) + 1/\tau_{tr}, \quad (3.26)$$

Numerical factors of the order of 1 in the stay times $\tau_{lo}$ and $\tau_{tr}$ are to be defined such that the sink strengths becomes equal to $k_{3i}^2$ in the limiting case of 3D diffusion. This is achieved by choosing

$$\tau_{lo,tr} = 4k_i^2 / D_{lo,tr}k_{(3i)}^4. \quad (3.27)$$

Equation (3.9) in conjunction with equations (3.26) and (3.27) provides a full description of the diffusion-reaction kinetics of defects, including the limiting case of pure 1D diffusion, the intermediate cases of 1D diffusion with direction changes and transversal diffusion, and the limiting case of isotropic 3D diffusion. Explicitly written, the general single-variable master curve for the dependence of the sink strength on the main parameters $l_{ch}$, $\delta$, $k_{(1)}^2$ and $k_{(3i)}^2$ involved reads

$$y = 0.5 \left\{1 + \sqrt{1 + 4x^2}\right\}, \quad \text{with}$$

$$x^2 = \left(l_{ch}^2 k_{(1)}^4 / 12 + k_{(1)}^2 / k_{(3i)}^4\right)^{-1} + \delta f(\delta)k_{(3i)}^4 / k_{(1)}^4. \quad (3.28)$$

### 3.2 1D to 3D Diffusion Reaction Kinetics between Mobile Defects

In the preceding subsections, we have considered RK where one reactant is mobile but the other (the sink) is immobile. In discussing the extension to the RK of two types of mobile reactants with each other, generally in the presence of other mobile and immobile reactants, we have to consider two aspects in RK: the diffusion of the reactants and their reaction efficiency, described in equation (3.1) by the diffusion constant, $D$, and the sink strength, $k^2$.

It is obvious that for the rate of reaction of two mobile defects of type i and j, only their relative diffusion matters, meaning that we would have to substitute

$$D \to D_{ij} = D_i + D_j, \quad (3.29)$$

in a generalized version of equation (3.1). One obvious consequence of this superposition of the two defect diffusion constants is that the RK of two defects diffusing in different 1D directions is 2D (Gösele and Seeger (1976), Gösele, 1984).
The generalisation of $k^2$ is less evident. First of all, a certain type of defect does not represent an invariable sink for another type of defect but the reaction between both results in a third type of defect. Secondly, the reaction term $D_n c_n k_n^2$ in equation (3.1) is symmetric in the concentrations/densities of the reactants (as it should be for two independent mobile reactants) only for 3D diffusion and moderate anisotropy but not for RK close to 1D where it depends linearly on the concentration of the mobile defect but quadratically on that of the immobile sinks as discussed in subsection 3.1.2. This suggests that equation (3.1) can be generalized by substituting the reaction term $D_n c_n k_n^2$ by a term symmetrical in the two mobile defects. For a homogeneous reaction rate without defect production ($P = \Delta c = 0$) this generalization results in

$$\frac{\partial c_i}{\partial t} = \frac{\partial c_j}{\partial t} = -\overline{D}_y (c_i k_j^2 + c_j k_i^2) / 2, \text{ with } \overline{D}_y = \overline{D}_i + \overline{D}_j,$$  

(3.30)

where, for the sink strength, equation (3.28) is to be used containing the length scales of the system and the anisotropy parameters. The validity of the symmetrisation used in equation (3.28) can be tested by applying this to the evolution of immobile clusters of a certain type of defect by their reaction with mobile mono-defects.

There remains, however, a third problem. The inclusion of the “shadowing effect” in the absorption of 1D diffusing defects by the side of a background sink oriented away from the individual sink considered in equations (3.4) and (3.22) has a well defined meaning only for immobile sinks staying in the drainage channel. Inclusion of this effect by using equation (3.28) is associated with an uncertainty in the sink strength which is, however, restricted to the transition region between “correlated” and “uncorrelated” 1D RK, and there the margin is most likely small but certainly smaller than by a factor of 2. This is illustrated in figure 3.5 where the “master curve” is plotted with and without inclusion of the “shadowing effect”. Consequently, we may recommend the use of equation (3.28) even for the reaction of mobile defects.

4. Application to the Evolution of Voids

The formation and growth of voids (more generally cavities including voids and bubbles) in metals under irradiation and the associated macroscopic swelling is one of the most prominent features of radiation damage. The evolution of voids depends not only on irradiation parameters such as temperature, dose and dose rate but also the crystal structure (Singh and Evans 1995, Golubov et al 2000a). Under neutron irradiation, for instance, voids are formed in a much wider temperature range in BCC metals ($\sim 0.19 T_m$ and above) than in FCC metals ($\sim 0.35 T_m$ and above) (in stage V), and cavity densities formed are substantially higher in BCC than in FCC. At high
doses, voids in a number of BCC metals, cease to grow and form void super-
lattices whereas these features seem to be missing in FCC metals. The aim of
modelling is to describe all of these features adequately and consistently. A
comprehensive treatment of cavity formation and growth in terms of the
general RK presented above is, however, far beyond the scope of the present
paper. In the following we apply 1D RK, disturbed by 1D direction changes
and by diffusion transversal to the 1D direction, to void growth and its
saturation at high doses and discuss the observed differences between BCC
and FCC metals in the light of the revealed features.

4.1 Swelling Rate

Swelling is determined by the biased partitioning of SIA-type defects and
vacancies over voids and other types of defects. This has been discussed in
detail previously in the framework of the standard rate theory where the bias
is assumed to be due to the preferential absorption of single SIAs by
dislocations (“dislocation bias”, see Brailsford and Bullough, 1972, 1981)
and, more recently, in the framework of the extended “production bias
model” where the bias is assumed to be due to the production and 1D
diffusion of SIA clusters (see Trinkaus et al, 1992; Singh et al 1997).
Therefore, we need not repeat here the procedure resulting in appropriate
expressions for the swelling rate.

In the following we assume that mobile SIAs and vacancies surviving intra-
cascade recombination are produced at an effective rate $G$, with partial rates
$(1- \varepsilon)G$ for the production of SIAs in 3D diffusing SIA defects (single SIAs
and di-SIA clusters) and $\varepsilon G$ for the production of SIAs in 1D diffusing
clusters, respectively, and that all these defects are distributed over voids of
(neutral) sink strength $k_v^2$ and dislocations of density $\rho$ (meaning that no
other sinks are present in the system). Generalizing the 1D RK for SIA
clusters used previously by introducing general sink strengths as discussed in
§3 we may write for the swelling rate

$$\frac{dS}{dt} = \rho (1- \varepsilon) \frac{Z_v^i k_v^2 Z_v^d}{(Z_v^i k_v^2 + Z_v^d)(Z_v^i k_v^2 + Z_v^d \rho)} + \varepsilon \left( \frac{Z_v^i k_v^2}{Z_v^i k_v^2 + Z_v^d \rho} - \frac{k_v^2}{k_{vd}^2} \right).$$

(4.1)

Here $k_v^2$ is the (neutral) sink strength of voids for the absorption of the 3D
diffusing defects (single SIAs, SIAs in small clusters and vacancies), $\rho$ is the
dislocation density, $Z_{iv}^d$ are efficiency factors for the absorption of SIAs and
vacancies (i,v) by voids and dislocations (v,d), respectively, for which we
assume, for simplicity, $Z_{iv}^d = Z_v^d = 1$ and $Z_i^d = Z = 1 + p$ where $p$ is the
quantity characterizing the “dislocation bias”, $k_{vd}^2$ and $k_{d,cl}^2$ are the general
sink strengths for the absorption of 1D to 3D diffusing SIA clusters by voids
and dislocations, respectively. The first and the second terms in equation (4.1) are the contributions of the dislocation bias and the production bias, respectively.

For a mono-disperse random distribution of voids of radius $R$ and number density $N$, the expression for pure 1D diffusion of SIA clusters assumed so far in the production bias model (Trinkaus et al, 1992; Singh et al 1997) is obtained by using in the second term of (4.1) the sink strength for 1D diffusing SIA clusters (equations (3.9) for $l_{ch} \to \infty$)

$$k_{1,ccl}^2 / k_{1,cd}^2 = \pi R^2 N / (\pi R^2 N + \pi d \rho / 4). \quad (4.2)$$

The expression for pure 3D diffusing SIA clusters, on the other hand, is obtained by using

$$k_{3,ccl}^2 / k_{3,cd}^2 = 4\pi RN / (4\pi RN + Z_{cl} \rho), \quad (4.3)$$

where $Z_{cl} = 1 + p_{cl}$ is the efficiency for the absorption of SIA clusters on dislocations ($p_{cl}$: cluster dislocation bias).

The contribution of the production bias to the swelling rate at low doses has been shown to be significantly larger than that of the dislocation bias if pure 1D diffusion of the mobile SIA clusters and physically reasonable values for $p$ (<5%) and $\varepsilon$ (> 10%) are assumed (Golubov et al 2001). With increasing dose, however, the contribution of the production bias to the swelling rate decreases since the absorption of 1D diffusing SIA clusters by voids increases faster with the increasing void radius (as $R^2$) than the absorption of the 3D diffusing vacancies by voids (as $R$) which can result in the saturation of void growth and swelling depending on the specific conditions.

### 4.2 Saturation of Void Growth and Swelling

In the following, we discuss the saturation of void growth and, particularly, the void size (radius $R_s$) reached in saturation as a function of the specific conditions. To determine the latter, we have to consider vanishing void growth and swelling rates, i.e. $dR/dt = 0$. This means that we need to find the zero(s) of the right hand side of equation (4.1) with respect to the void radius $R$.

Assuming in equation (4.1) that all SIAs are produced in the form of 1D diffusing glissile clusters, $\varepsilon = 1$, and, for a mono-disperse random distribution of voids of radius $R$ and number density $N$, $k_{ccl}^2 / k_{cl}^2 = \pi R^2 N / (\pi R^2 N + \pi d \rho / 4)$, a surprisingly simple result was obtained by Trinkaus et al (1992) and Golubov et al (2000)

$$R_s^{(D)} = \pi d. \quad (4.4)$$
According to this, the saturation size, $R_s$, is proportional to the absorption diameter of the dislocations, $d$, independent of the sink densities and the other parameters contained in equation (4.1).

Any disturbance of the assumed “ideal conditions” (SIAs only in 1D-diffusing clusters) will, however, increase the value of $R_s$. Possible disturbances of the “ideal conditions” are: (a) the concurrent production of 3D diffusing SIA defects (mono- and di-interstitials) with a non-vanishing dislocation bias, $\varepsilon < 1$, $p > 0$ (mixture of 1D and 3D diffusing SIA defects), (b) 1D direction changes and (c) diffusion transversal to a 1D direction (i.e. disturbances described by equation (4.1) which are of primary interest here) and, (d) the presence of pre-existing sinks in addition to dislocations, and (e) the production of other types of immobile sinks in addition to voids and dislocations such as sessile SIA and vacancy clusters. We consider here only the first three types of disturbances being most interesting in the present context. Because of the many parameters involved, we study their effects on the saturation value of the void size separately.

### 4.2.1 Concurrent Production of 1D and 3D Diffusing SIA Defects

In displacement cascades, a wide spectrum of SIA defects consisting of 3D diffusing mono-interstitials and di-interstitials as well as mobile 1D diffusing and sessile higher order clusters are produced. The ratio of the 1D diffusing SIA clusters to the 3D diffusing mono-interstitials and di-interstitials, $\varepsilon$, increases with increasing recoil energy transmitted from the projectile particle to a matrix atom. We study here the effect of mixing 3D and 1D diffusing SIA defects on void growth saturation by using equation (4.2) in equation (4.1). For the detailed analyses, we introduce the following dimensionless representations for the saturation size $R_s$, void density $N$, dislocation density $\rho$ and ratio of the void to the dislocation density (henceforth called scaled saturation size etc)

$$R*_s = \frac{R_s}{R_s^{(1D)}} = \frac{R_s}{(\pi d)}, \quad (4.5)$$

$$N* = (R_s^{(1D)})^3 N = (\pi d)^3 N, \quad \rho* = \pi d^2 \rho/4, \quad Q* = N*/\rho*. \quad (4.6)$$

With this scaled variables, we may write $dS/dGt = 0$ according to equation (4.1) in the form

$$(\varepsilon Z - p)Q* R^* - \varepsilon (Q* - Z) R^* - (\varepsilon + p) = 0, \quad (4.7)$$

where $Z = 1 + p$. According to equation (4.7), the scaled saturation size $R^*$ depends on totally 3 independent parameters: the production bias and dislocation bias parameters, (i.e. $\varepsilon$ and $p$, respectively) and the scaled density ratio $Q^*$. The physically acceptable positive solution of this quadratic equation for $R^*$ is given by
In figure 4.1(a), this solution for the scaled saturation size $R^*$ according to equation (4.8) is plotted vs scaled density ratio, $Q^*/\rho^*$, for various combinations of the production bias and dislocation bias parameters, $\varepsilon$ and $p$. It can be seen that even for the most extreme case shown (lowest value of $\varepsilon = 10\%$, highest value of $p = 5\%$), the change remains relatively moderate below 80%.

A real solution of equation (4.8) for $R^*$, i.e. the occurrence of void growth saturation, requires the discriminant in equation (4.8) to be positive, i.e.

$$
\varepsilon^2(Q^*-Z)^2 + 4(\varepsilon Z-p)(\varepsilon + p)Q^* > 0.
$$

(4.9)

The physically acceptable positive solution of this quadratic equation for $\varepsilon$ is plotted in figure 4.1(b) vs $Q^*/\rho^*$ for various values of $p$. Saturation occurs for values of $\varepsilon$ above, no saturation for those below the curves shown in fig. 4.1(b). The analysis shows that $\varepsilon > p$ represents a sufficient, though not necessary condition for growth saturation. Since generally $p < 5\%$, we may conclude that even small cascades with a relatively small fraction of SIAs produced in 1D diffusing clusters, $\varepsilon > 5\%$, would result in void growth saturation, provided no other significant disturbance of the 1D RK were operating.

### 4.2.2 Disturbance by 1D Direction Changes

In this section, we discuss the effect of changes in the 1D diffusion direction changes (Burgers vector changes) on void growth saturation by assuming in equation (4.1) $\varepsilon = 1$, and using in the term $k_{ch}^2/l_\text{cl}$ the general 1D to 3D sink strength given by equation (3.12). Introducing the additional scaled dimensionless parameters

$$
\kappa^* = N^* R^* + \rho^* = \kappa d, \quad l^* = \sqrt{2l_\text{ch}}/(3\pi d).
$$

(4.11)

we may write $dS/dGt = 0$ according to equation (4.1) in the form

$$
R^*(1 + \sqrt{1 + [(3\pi\kappa^*/4l^*Z_{cl})^2]}^{-1}) = 1 + \sqrt{1 + [(3\pi\kappa^*/4l^*Z_{cl})^2]}^{-1}.
$$

(4.12)

According to equation (4.12), the scaled saturation size $R^*$ depends on 4 independent parameters involved in the process: the scaled void and dislocation densities $N^*$ and $\rho^*$ contained in $\kappa^*$, the scaled 1D-diffusion length $l^*$, and the efficiency of dislocations for absorbing SIA clusters $Z_{cl}$. 

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We did not succeed in deriving a closed form analytical solution of equation (4.12) with respect to $R^*$ and therefore depended on numerical solutions for various combinations of the parameters. In Figure 4.2, the relative change of the saturated void size, $R^*(l^*)-1$, is plotted vs scaled 1D-diffusion length $l^*$ for various combinations of the cluster dislocation absorption efficiency, $Z$, and scaled cavity and dislocation densities, $N^*$ and $\rho^*$, respectively. Figure 4.2 shows that the effect of direction changes in 1D diffusion is significant only for RK close to the 3D limiting case, where $l^* < 1$, and even then the effect is relatively moderate.

### 4.2.3 Disturbance by Transversal Diffusion

To discuss the effect of diffusion transversal to the 1D direction changes (by self-climb) on void growth saturation we assume in equation (4.1) $\varepsilon = 1$, and use in the term $k_{sc}^2 / k_{cl}^2$ the general 1D to 3D sink strength given by equation (3.25). In this case, we may write $dS/dGt = 0$ according to equation (4.1) in the form

$$R^*(1 + \sqrt{1 + \delta(4/3\pi \kappa^* R^*)^2}) = 1 + \sqrt{1 + \delta(4Z_{cl}/3\pi \kappa^*)^2}$$

(4.13)

Some manipulation of this equation results in a relation between $\kappa^*$ and $R^*$

$$\kappa^* = N^* R^* + \rho^* = \frac{2(Z_{cl}^2 - 1)}{3\pi \sqrt{(R^* - 1)(Z_{cl}^2 R^* - 1)}}$$

(4.14)

by which the scaled void and dislocation densities, $N^*$ and $\rho^*$, can be easily expressed by $R^*$.

According to equation (4.14), the scaled saturation size $R^*$ depends now on totally only 3 independent parameters: new scaled void and dislocation densities, defined by $u = N^*/\sqrt{\delta}$, and $v = \rho^*/\sqrt{\delta}$, respectively, and the efficiency of dislocations for absorbing SIA clusters, $Z_{cl}$.

In figure 4.3 (a) and (b), equation (4.14) has been used to plot the relative change of saturated void size, $R^* - 1$, vs the new scaled void density, $u = N^*/\sqrt{\delta}$, and dislocation density, $v = \rho^*/\sqrt{\delta}$, respectively, for different values of the other density and $Z_{cl} = 1.4$. Figure 4.3 reveals that, differently from the two other cases discussed in the preceding sections, the saturation size increases very significantly with decreasing void and dislocation densities for 1D RK disturbed by transversal diffusion. We emphasize here that an increase in the saturation level is generally associated with an increase in the saturation dose.

This feature could explain why void growth saturation occurs in BCC where void densities are very high, but seem not to occur in FCC where void
densities are more than 1 order of magnitude lower. This interpretation has to be considered, however, with caution since the strong increase of the saturation size occurs in the transition regime between moderate to high diffusion anisotropy where also other features characteristic for 1D RK such as the decoration of dislocations with dislocation loops observed in FCC would tend to disappear.

We emphasize here that the temperature dependence of the void evolution is contained in the intrinsic material and microscopic parameters contained in equation (4.1) and in the related equations for void growth saturation equations (4.8), (4.12) and (4.14): the mean 1D diffusion length $l$, the transversal to longitudinal diffusion ratio, $\delta$, and the void and dislocation densities, $N$ and $\rho$.

We illustrate this here for the effect of diffusion transversal to the 1D direction changes on void growth saturation, assuming for the parameters contained in equation (4.14), $d = 1 \text{nm}$ (absorption diameter of the dislocations), $\delta = 10 \times \exp(-7T_m/T)$ (diffusion ratio) and that voids form the dominant sinks with the densities depending on temperature characteristic for BCC and FCC metals as given by Trinkaus et al (2000). In figure 4.4, the resulting temperature dependencies of the saturated void size are shown. The saturated void size increases strongly with increasing temperature, and this increase occurs at a significantly higher level for FCC than for BCC metals. On the other hand, this means that the temperature, where void growth saturation would not be reached at realistic doses, is lower for FCC than for BCC metals.

4.3 Discussion

Saturation of void growth is the most prominent feature of pure 1D RK of SIAs combined with pure 3D RK of vacancies. For this specific limiting case, a surprisingly simple result for the maximum void size where voids would cease to grow, was obtained as given by equation (4.4), which relates the maximum void radius directly to the absorption length of dislocation. Any disturbance of the pure 1D RK of SIAs shifting the RK towards 3D must be expected to increase the saturation size or even to wipe out this fingerprint of a 1D RK and to make its parameter dependence more complicated.

In the present chapter 4, we have analysed the effect of disturbances of the pure 1D RK on the void growth characteristics as a function of void densities and temperature, and have discussed differences between BBC and FCC metals related to differences in the void densities. Since we wished to illustrate some general trends we could keep the discussion transparent by making a number of simplifying assumptions. First of all, we have made the
general assumptions underlying the preceding discussion of the general 1D to 3D RK: spatially random distributions of sinks limited to a moderate volume fraction (say < 10%). Secondly, we have assumed that voids and dislocations represent the only sinks for SIAs and vacancies. Thirdly, we have assumed that 1D diffusion, direction changes and transversal diffusion are not affected by impurities, i.e. we have considered pure metals. In addition, we have analysed each specific disturbance of 1D RK separately, i.e. we have ignored synergetic effects of all disturbances.

The simplest 1D to 3D RK is represented by a mixture of purely 1D diffusing SIA clusters and 3D diffusing mono-, di-interstitials and perhaps tri-interstitials. Assuming undisturbed 1D diffusion of glissile SIA clusters, we have shown in subsection 4.2.1 that void growth saturates as long as the fraction of 1D diffusing SIAs, the “1D production bias”, is larger than the “dislocation bias” for the 3D diffusing SIAs. Since it is generally agreed that the fraction of SIAs in 1D diffusing clusters is of the order of 10% or even more whereas the “dislocation bias” is restricted to only a few % (2%), at least for mono-interstitials, we may argue that void growth saturation is kept in realistic mixtures of 1D and 3D diffusing SIAs. It is not clear, however, whether the contributions to the effective dislocation bias of 3D migrating SIA clusters (di-interstitials and tri-interstitials) which interact with dislocations stronger than mono-interstitials, could invalidate this argument.

The effects of disturbances of 1D diffusion of SIA clusters on the void growth characteristics are more interesting in the context of the present paper. Direction changes shift the 1D RK of SIAs towards 3D increasing thereby the saturation size. Assuming that all SIAs are produced in the form of glissile loops, we have shown in section 4.2.2, that a significant increase of the saturation size can only be expected for relatively frequent direction changes for which the mean 1D diffusion length \( l_{ch} = \sqrt{2D_{ch} \tau_{ch}} \) becomes comparable to the size of the voids meaning that the RK comes close to the 3D limiting case. MD simulations are not in favour of such frequent direction changes.

The other modification of 1D RK of SIAs is transversal diffusion 1D diffusion of SIA clusters in addition to their 1D diffusion quantified by ratio of the corresponding diffusion components, \( \delta = D_{lo}/D_{tr} \). Assuming again that all SIAs are produced in the form of glissile loops, and neglecting direction changes we have found in section 4.2.3, that, differently from the two other cases discussed, for 1D RK disturbed by transversal diffusion, the saturation size increases very significantly with decreasing void and dislocation densities. These findings may be related to the observation that void growth saturation and void lattice formations seem to be general phenomena in BCC where high void densities are nucleated, but not in FCC where nucleated void densities are more than one order of magnitude lower.
It is useful here to discuss the implications of the assumptions made in the above analysis. Any deviations in the system from the assumption of spatially random distributions of sinks, for instance by spatial correlations in the distribution of voids as in void ordering or between voids and other sinks, must be expected to affect void growth significantly. Such effects cannot be discussed in terms of the RK developed in the present paper. The same holds for high sink densities, particularly for high void densities in BCC where, for significant increase of the saturated void size, the volume fraction can easily exceed the range of validity of the present treatment. Other sinks for mobile SIAs and vacancies than voids and dislocations will most likely not affect our conclusions on void growth saturation. Sessile loops, for instance, produced during cascade irradiation will essentially act as recombination centers and thus reduce the effective defect production rate but most likely may not change the dislocation and production biases significantly. Synergetic effects of the three disturbances of the pure 1D RK will manifest themselves certainly in an enhanced total increase of the void saturation size or even wipe-out the void growth saturation. It is most likely that the disturbance with the strongest separate effect, i.e. transversal diffusion, dominates the general trend.

Finally, our assumption of irradiation of a pure impurity-free material and its implications for our conclusions should be discussed here briefly. First of all it is important to emphasize that even in a pure virgin metal, impurities will be continuously produced as transmutation products during neutron irradiation. Elastic interaction between mobile SIA clusters and impurities, initially present or produced during irradiation, may be generally expected to change the SIA cluster RK from 1D towards 3D. A 1D diffusing SIA cluster may be attracted by an impurity and get (temporarily) trapped by this, or such a cluster may get encaged between two impurities repulsing it. Both types of interaction configurations result primarily in a significant reduction of the longitudinal diffusion component of the SIA clusters, $D_{\text{lo}}$. Even if impurities did not affect the mean time between direction changes, $\tau_{\text{ch}}$, and the transversal diffusion components, $D_{\text{tr}}$, of the clusters, they could significantly reduce the mean diffusion length, $l_{\text{ch}} = \sqrt{2D_{\text{lo}}\tau_{\text{ch}}}$, and the longitudinal to transversal diffusion ratio, $\delta = D_{\text{lo}}/D_{\text{tr}}$, controlling the 1D to 3D RK of the mobile SIA clusters and via this the void growth characteristics. Contrary to possible effects of spatial correlations and high sink (void) volume fractions, such effects of impurities on the RK of the mobile SIA clusters can be treated within the framework of the 1D to 3D RK presented above. A detailed discussion of impurity effects on the 1D to 3D RK of SIA clusters and the associated void growth characteristics is beyond the scope of this paper and will therefore be presented elsewhere (Trinkaus and Singh, 2008).
5. Summary and Conclusions

The evolution of the microstructure in metals under cascade damage conditions is controlled by the diffusion of mobile lattice defects and their reactions with other mobile and immobile defects. In earlier modelling, cascade damage accumulation was assumed to result from the homogeneous production of single vacancies and self-interstitial atoms (SIAs) both of which were assumed to diffuse three-dimensionally. Even though experimental evidence for SIA cluster production in cascades was found already in the 1970s this important feature of cascade damage was recognized seriously only at the beginning of the 1990s when it was confirmed by Molecular Dynamics (MD) (English et al 1990; Diaz de la Rubia and Guinan 1990,1991). Since the beginning of the nineties, it was recognised that in cascades both clusters of vacancies and SIAs are formed (even though experimental evidence for SIA cluster formation in cascades existed already in the 1970s) and that some of the small SIA clusters are able to diffuse one-dimensionally (1D).

The 1D diffusion of SIA clusters must be considered to be generally disturbed by changes in their diffusion direction and by transversal diffusion, resulting in diffusion reaction kinetics between the 1D and 3D limiting cases. Both the frequency of direction changes and the transversal component of the diffusion coefficient are (size and temperature dependent) intrinsic properties of SIA clusters. In the present work, the complicated diffusion reaction kinetics (RK) resulting from such disturbances of the pure 1D diffusion has been treated in great detail.

The first step in our treatment was to establish and test the main features of the mathematical procedure required by considering the effect of direction changes on the RK of SIAs in the presence of a random distribution of fixed sinks. In this case, the partial sink strengths depend on the average diffusion distance between direction changes and the density and size of the sinks considered. An embedded mean field approach was developed for small disturbances of 1D diffusion by direction changes (large 1D diffusion range). The parameter dependence of the expression for the sink strength obtained by this procedure suggests the way how to extrapolate it to large disturbances of 1D diffusion (short 1D diffusion range) up to the limiting case of 3D diffusion. The resulting generalised expression could be written as a single variable function (master curve) for the dependence of the sink strength on the three main parameters mentioned. Data obtained by Kinetic Monte Carlo simulations were found to be in excellent agreement with this analytical master curve.

For the case of 1D diffusion disturbed by transversal diffusion, the treatment by Seeger and co-workers was extended with the aim to bridge the gap between the 1D and 3D limiting cases of diffusion anisotropy and to include
direction changes in addition to transversal diffusion. In the case of transversal diffusion, the main intrinsic property of the defect is the ratio of the transversal to the longitudinal diffusion components. Analogous to the procedure used for 1D diffusion with direction changes, an embedded mean field approach was applied and extrapolated to the limiting case of 3D diffusion. The resulting expression for the sink strength has the same form as the master curve for 1D diffusion with direction changes except that the meaning of the main variable is different. Recent kinetic Monte Carlo simulations were again found to be in excellent agreement with the analytical master curve.

Finally, the master curve was further generalised to include the combined effects of direction changes and transversal diffusion. Moreover, the procedure was extended to the diffusion and reaction kinetics of (two) mobile defects, allowing both to have a diffusion characteristics between 1D and 3D. We think that, with this, the most general form of the reaction kinetics for spatially random distributions of sinks is formulated and thus ready for application in modelling of damage accumulation during irradiation with cascade producing particles.

In order to illustrate the strength of our description of general defect diffusion reaction kinetics of defects, this was applied to specific aspects of the evolution of voids under cascade damage conditions: the saturation of void growth and swelling at high doses and its dependence on the crystal structure. In fact, under neutron irradiation voids are formed in a much wider temperature range in BCC metals than in FCC and cavity densities formed are substantially higher in BCC than in FCC.

An attempt was made to rationalize this difference in terms of differences in the 1D to 2D reaction kinetics of the defects involved. For this, void swelling was described in term of combined contributions of the “dislocation bias” and the “production bias”, allowing some fraction of the SIA clusters to follow the generalized reaction kinetics. The effects of a mixture of 1D and 3D diffusing SIA defects, 1D direction changes and diffusion transversal to a 1D direction were explored separately. The strongest effect on saturation of void growth was found to be generated by an even small transversal diffusion component, depending, however strongly on the ratio of the void to the dislocation densities. The results provide an explanation why void growth saturation occurs in BCC where void densities are very high, but seem not to occur in FCC where void densities are more than one order of magnitude lower.

Impurities and alloying elements must be considered to have substantial effects on the defect reaction kinetics and the resulting damage accumulation. Consideration of such effects is beyond the scope of the present work. We will discuss such effects in a separate paper (Trinkaus and Singh 2008).
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Figure 2.1. Illustration of the main processes relevant for the reaction kinetics of SIA loops from the point of view of macroscopic dislocation theory. (a) diffusional glide as random jumps across the Peierls barrier, (b) random self-climb by diffusion of double jogs (core interstitials and vacancies), (c) Burgers vector change by the sweeping of a dislocation across the loop area.
Figure 3.1. Illustration of the three characteristic cases in the RK of a piecewise 1D diffusing defect with an immobile sink; (a) correlated 1D, (b) uncorrelated 1D, (c) 3D.

Figure 3.2. Illustration of 1D defect diffusion disturbed by direction changes in a drainage channel to a sink. Defects can leave and enter the channel by direction changes.
Figure 3.3. Analytical “master curve” interpolating between the sink strengths for the 1D and 3D limiting cases: Sink strength normalised to its value for pure 1-D, $k^2/k(1)^2$, vs measure for the "disturbance of 1-D diffusion". According to equations (3.12) and (3.25), the meaning of "disturbance of 1-D diffusion" is $(l_{ch}^2 k(1)^2 / 12 + k(1) / k(3)^4)^{1/2}$ and $(\delta^d / f(\delta) k(3)^2 / k(1)^2)$ for disturbance by Burgers vector changes and conservative self-climb, respectively.
Figure 3.4. Illustration of effect of scaling transformation: diffusion becomes isotropic, but shape of sinks changes from spherical to ellipsoidal; (a) moderate anisotropy, (b) high anisotropy. In case (b), the maximum extension of the sinks in the transformed state becomes larger than the shortest distance between the sinks. The normal sink in the form of “paste” in the original state becomes a “puff pastry” in the transformed state.
Figure 3.5. Sink strength normalized to its 1D limit vs. “disturbance of 1D diffusion” described by $\delta^{1/2}/S$ for 0.1% and 10% swelling, defined implicitly by equations (3.25) to (3.27) and (3.32) to (3.34) derived from the approximate sink embedding procedure (symbols) and by the explicit analytical “master curve” (lines). Note the good agreement: deviations less than 20% and 5% for 0.1% and 10% swelling, respectively.
Figure 3.6. Analytical “Master Curve” for 1D diffusion disturbed by transversal equations according to equation (3.25). For comparison, the KMC results for two different conditions involving absorbers of different radius R and number density N are also shown.
Figure 4.1. Effect of mixing 3D diffusing SIAs to 1D diffusing SIA clusters, $\epsilon < 1$, $p > 0$, on the saturation of cavity growth, $R_s$. (a) Scaled saturation size $R_s^* = R_s / R_s^{(1D)}$, vs scaled density ratio of void to dislocation density, $N^*/\rho^*$, for various combinations of the production bias ($\epsilon$) and dislocation bias ($p$) parameters. Even for the extreme case shown, $\epsilon = 10\%$, $p = 5\%$, the change remains relatively moderate below 80\%. (b) Limits of void growth saturation in the ($\epsilon - N^*/\rho^*$) plane for various values of $p$. Saturation/no saturation occurs above/below the curves; $\epsilon > p$ is a sufficient, though not necessary condition for growth saturation.
Figure 4.2. Effect of direction changes in 1D diffusion on the saturation of void growth. Relative change of saturated void size, $R^{-1}$, vs scaled 1D-diffusion length $l^*$ for various combinations of the cluster dislocation absorption efficiency, $Z$, and scaled cavity and dislocation densities, $N^*$ and $\rho^*$, respectively. Note that the change in size is significant only for diffusion-reaction kinetics close to the 3D limiting case, $l^* < 1$. 
Figure 4.3. Effect of diffusion transversal to the 1D diffusion direction, $\delta > 0$, on the saturation of void growth. Relative change of saturated void size, $R^* - 1$, vs (a) scaled void density, $N^*/\sqrt{\delta}$, and (b) scaled dislocation density, $\rho^*/\sqrt{\delta}$, respectively, for different values of the other density and $Z = 1.4$. The saturation size increases significantly with decreasing densities.
Figure 4.4. Temperature dependence of void growth saturation: Relative change of saturated void size, $R^* - 1$, vs reciprocal homologous temperature, $T_m/T$, for void densities typical for FCC and BCC metals. The saturated void size increases strongly with increasing temperature and is significantly larger for FCC than for BCC metals.