A Mesoscopic Approach to Point-Defect Clustering in Solids during Irradiation

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Abstract. Accumulation of point defects in solids during irradiation is often accompanied by self-organization processes which lead to point-defect clustering and thus to the formation of a spatially inhomogeneous defect structure. Within the framework of a mesoscopic phenomenological approach, the conditions for clustering of mobile point defects caused by their elastic interactions are studied. It is shown that differences between the elastic interaction of similar and that of dissimilar defects may lead to such clustering. Further, it is shown that the presence of impurities acting as traps for interstitials may promote the clustering process. The conditions for spatial clustering are studied for characteristic material parameters in order to predict experimental observations of this phenomenon in metals and ionic solids.

PACS: 61.70.Bv, 61.80.−x, 61.90.+d

Segregation of defects of the same kind into clusters during the process of defect accumulation under irradiation is a quite universal phenomenon. It has been studied during the last decades both experimentally and theoretically [1–7]. For the theoretical treatment of segregation processes, two main approaches have been adopted, which will be discussed in what follows.

In the *microscopic approach* [1, 2, 7, 8] the defect distribution is characterized by many-particle densities or by the corresponding defect–defect correlation functions. This allows to take into account the influence of defect correlations on the recombination kinetics. Within this approach, it is possible to show that the very nature of the recombination process provides a tendency towards clustering of defects of the same kind on the microscopic scale (*microscopic self-organization* [1]). In contrast, the phenomenological *mesoscopic approach* towards defect accumulation and clustering characterizes the point-defect distribution by spatially averaged but, in general, space-dependent concentrations C_V and C_I of vacancies and self-interstitials. The recombination kinetics is treated in a *mean-field* procedure [9, 10], neglecting spatial correlations between defects and their impact on the reaction kinetics. Nevertheless, nontrivial behaviour may arise if *additional* interactions, such as elastic attraction between point defects [3, 4] or dynamic radiation-induced interactions [6] are taken into account. In this approach, defect clustering manifests itself as a breaking of the translational symmetry of the defect distribution due to the emergence of spatial modulations of the averaged defect concentrations that occur on a scale which is large compared to the distances between the individual defects, i.e., as a 'classical' self-organization process [11]. Treating point-defect clustering under recombination-controlled conditions, Martin [3, 4] was the first who recognized the possibility of *synergetic behaviour* of defects in irradiated solids.

Defect clustering under recombination-controlled conditions arises from the spatial segregation of the mixture of point defects of two kinds (vacancies and interstitials) into clusters of defects of the same kind, i.e., it requires the occurrence of 'uphill' point-defect fluxes. This is possible if the average drift fluxes arising from an attractive interaction between defects of the same kind are larger than the sum of the fluxes arising from attractive interactions between defects of different kinds and from the usual 'downhill' diffusion. Thus, spatial segregation requires two conditions to be fulfilled: *(i)* There has to be an asymmetry between the mean attraction of defects of different kinds and the mean attraction of defects of the same kind, so that the latter effect may become predominant; *(ii)* Point-defect densities have to be rather high in order to make the drift fluxes arising from mutual interactions sufficiently large.

In Martin's model, condition *(i)* was fulfilled by introducing dislocation sinks at which interstitials, which are more mobile than vacancies, disappear. This causes the attraction between pairs of defects of the same kind (vacancies) to become more important than that between pairs of defects of different kinds, simply because such pairs practically do not exist. This approach, however, fails to describe a simultaneous formation of vacancy *and* interstitial clusters at low temperatures where the interstitial mobilities are too low to make interstitials reach extended sinks within

the time scale of the experiment (although recombination is already possible), or in extremely pure, dislocation-free crystals. We therefore adopt a modified approach by taking into account that there exist *direct* asymmetries in the effective interactions that may give rise to clustering. Further, we take into account the influence of impurities, which may act as traps for interstitials, and study the resulting modifications of the segregation behaviour. We neglect the influence of metastable, low-dimensionally migrating interstitial configurations on the defect dynamics, since we shall deal with situations where point-defect concentrations are high. In this case, due to their interactions with other point defects, metastable interstitials are converted athermally into the stable, three-dimensionally migrating dumbbell-type interstitial configuration [6, 12].

1 Basic Equations, Conditions for Defect Clustering

Within the phenomenological approach used here, the process of diffusion-limited defect recombination is taken into account by a bimolecular reaction term, containing the time-independent recombination constant $K_R = 4\pi D R_a$, where R_a is the recombination radius and $D = D_V + D_I$. Since the diffusivity D_V of vacancies is usually much smaller than the diffusivity D_I of interstitials, we shall neglect vacancy diffusion during our subsequent discussion by assuming that defect transport is only due to the diffusion and drift of interstitials ($D \approx D_I$). This situation is typical for many metals and ionic solids at low temperatures. (The modifications of our results which arise in the case in which vacancy motion is not negligible are briefly discussed in the next section.) Our starting equations thus read

$$\partial_t C_V(\mathbf{r}, t) = P - K_R C_V C_I,\tag{1}$$

$$\partial_t C_I(\mathbf{r}, t) = P - K_R C_V C_I + D\nabla\left[\nabla C_I(\mathbf{r}, t) + \frac{C_I}{k_B T}\nabla E_I(\mathbf{r})\right].\tag{2}$$

Here P is the point-defect production rate, which is assumed to be equal for vacancies and interstitials. The potential energy E_I of an interstitial depends on its interaction with all other defects. E_I may thus be written as

$$E_I(\mathbf{r}) = \int\limits_{V_\infty} C_V(\mathbf{s})E_{VI}(\mathbf{r} - \mathbf{s}) + C_I(\mathbf{s})E_{II}(\mathbf{r} - \mathbf{s})d^3s,\tag{3}$$

where E_{VI} and E_{II} denote the interaction energies between a vacancy and an interstitial or between two interstitials, respectively. All relevant solutions of (1–3) must satisfy the requirement of mass conservation,

$$\int\limits_{V_\infty} C_V(\mathbf{r})d^3r = \int\limits_{V_\infty} C_I(\mathbf{r})d^3r.\tag{4}$$

Here V_∞ denotes the crystal volume. In this work, we shall consider the case of short-range interactions, which are assumed to be given by the simple expressions

$$E_{JK}(r) = -\hat{E}_{JK}f(r), f(r) = \begin{cases} 1, & r \le r_0 \\ 0, & r > r_0 \end{cases};$$

$$J, K \in \{V, I\},\tag{5}$$

where r_0 is the interaction radius. The assumption of an isotropic interaction between interstitials is justified if the energy for rotation of the (dumbbell-type) interstitial is significantly lower than its migrational energy, which holds for ionic solids [13] as well as for some metals [14]. Further, for ionic solids as well as for metals, the value of the interaction constant $\hat{E}_{JK}$ is larger for the interstitial–interstitial than for the vacancy–interstitial interaction [15–17].

In contrast to the model of Martin, our model neglects the influence of dislocations as sinks for point defects. In fact, the asymmetry in the interaction between defects of the same and of different kinds which arises from the stronger mutual attraction of interstitals suffices to give a nontrivial behaviour of the system.

In order to obtain information on the possibility of defect clustering and the development of spatially inhomogeneous defect patterns, we consider the time development of fluctuations around the homogeneous steady-state solution of (1–4), which is given by

$$C_V^0 = C_I^0 = C^0 = \sqrt{P/K_R}\tag{6}$$

and introduce the fluctuation amplitudes δc_V and δc_I via

$$C_V(\mathbf{r}, t) = C^0 + \delta c_V(\mathbf{r}, t), C_I(\mathbf{r}, t) = C^0 + \delta c_I(\mathbf{r}, t).\tag{7}$$

Following the formalism used in [3, 4, 6], we study the stability of the homogeneous steady-state solution (6) by investigating the equations of evolution, which are obtained by inserting (7) into (1) and (2). We linearize these equations, i.e., we consider the case of small fluctuation amplitudes. After introducing dimensionless coordinates via

$$t = C^t t', C^t = (4\pi R_a DP)^{1/2};$$

$$r = C^r r', C^r = \left(\frac{D}{4\pi R_a P}\right)^{1/4},\tag{8}$$

the following linearized equations for the fluctuation amplitudes are obtained:

$$\partial_{t'}\delta c_V(\mathbf{r}', t') = -\delta c_V(\mathbf{r}', t') - \delta c_I(\mathbf{r}', t'),\tag{9}$$

$$\partial_{t'}\delta c_I(\mathbf{r}', t') = -\delta c_V(\mathbf{r}', t') - \delta c_I(\mathbf{r}', t') + \Delta'[\delta c_I(\mathbf{r}', t') + \delta U_I(\mathbf{r}')],\tag{10}$$

$$\delta U_I(\mathbf{r}') = \frac{\sqrt{P/4\pi DR_a}}{k_B T}(C^r)^3\int\limits_{V_\infty}\{\delta c_V(\mathbf{s}')E_{VI}(|\mathbf{r}' - \mathbf{s}'|) + \delta c_I(\mathbf{s}')E_{II}(|\mathbf{r}' - \mathbf{s}'|)\}d^3s'.\tag{11}$$

Fourier transformation of these linear integro-differential equations gives us a set of two coupled ordinary differential equations, which in matrix notation read

$$\partial_{t'}\begin{pmatrix}\delta c_V(\mathbf{k}', t) \\ \delta c_I(\mathbf{k}', t)\end{pmatrix}$$
$$= \begin{pmatrix}-1 & -1 \\ -1 - k'^2 U_{VI}(k') & -1 - k'^2[1 + U_{II}(k')]\end{pmatrix}$$
$$\times \begin{pmatrix}\delta c_V(\mathbf{k}', t) \\ \delta c_I(\mathbf{k}', t)\end{pmatrix}.\tag{12}$$

The $U_{JK}(k')$ are related to the Fourier transforms $\tilde{E}_{JK}(k') = -\hat{E}_{JK}\tilde{f}(k')$ of the interaction potentials $E_{JK}(r')$ according to

$$U_{JK}(k') = -\frac{\sqrt{P/4\pi DR_a}}{k_B T}\hat{E}_{JK}\tilde{f}(k'), \text{ where}$$

$$\tilde{f}(k') = (C^r)^3\frac{4\pi}{k'}\left(\frac{\sin(k'r_0')}{k'^2} - r_0'\frac{\cos(k'r_0')}{k'}\right). \tag{13}$$

The criterion for (local) stability of the spatially homogeneous solution (6) is given by the condition that all spatially inhomogeneous fluctuations are damped, i.e. for all $\mathbf{k}$ both eigenvalues of the linearized evolution matrix are negative. This stability criterion may be written as

$$\varepsilon = \frac{\sqrt{P/4\pi DR_a}}{k_B T}(\hat{E}_{II} - \hat{E}_{VI})\tilde{f}(0) - 1 < 0. \tag{14}$$

The critical eigenvalue branch of the linearized matrix (i.e., the branch that may exhibit positive eigenvalues) is given (in dimensionless units) by the relation

$$\omega'(k')$$
$$= \sqrt{\frac{\{k'^2[1 + U_{II}(k')] + 2\}^2}{4} - k'^2[1 + U_{II}(k') - U_{VI}(k')]}$$
$$- \frac{k'^2[1 + U_{II}(k')] + 2}{2}. \tag{15}$$

In the case of a weak instability, i.e., for small ε, this expression can be rewritten in the following simplified form (which is formally identical with the expression for the amplification factor of concentration modulations in the case of spinodal decomposition [18])

$$\omega'(k')\overset{\varepsilon\to 0}{\cong}\frac{\varepsilon^2}{2\alpha}\left(\frac{k'}{k_c'}\right)^2\left[1 - \left(\frac{k'}{k_c}\right)^2\right]; k_c' = \sqrt{\frac{\varepsilon}{\alpha}}. \tag{16}$$

In (16), α is related to the 'structure factor' $\tilde{f}$ (for $\tilde{f}$ as in (13), $\alpha = (r_0')^2/10$). From (16) it can be seen that, when the stability boundary is crossed, instability occurs first at small k, i.e., at long wavelengths.

Since in the linear approximation the maximum value ω'_{max} of $\omega'(k')$ is just the maximum growth rate for concentration modulations, this quantity, as determined from (16), may be used to estimate the irradiation conditions required to achieve segregation at a given small rate $1/t_c'$,

$$1/t_c' \leq \omega'_{max} = \frac{\varepsilon^2}{8\alpha}. \tag{17}$$

In order to perform explicit calculations, we have to specify the temperature dependence of the diffusion coefficient $D \approx D_I$. This is done by using the usual form

$$D = D_I^0\exp\left(-\frac{E_I^M}{k_B T}\right), \tag{18}$$

where E_I^M is the activation energy for interstitial migration. If (unsaturable) traps are present in a concentration C_{tr}, it is posssible to take into account their influence on the stability behaviour by replacing D by a modified diffusion coefficient

$\tilde{D}$ that is defined by

$$\tilde{D}^{-1} = D^{-1} + 4\pi C_{tr}R_{tr}\nu^{-1}\exp\left(\frac{E^B + E_I^M}{k_B T}\right). \tag{19}$$

Here E^B denotes the binding energy to the trap, R_{tr} the trap capture radius, and ν an attempt frequency of the order of magnitude of the Debye frequency.

2 Results

From the stability condition (14), we obtain the instability diagram of Fig. 1 showing the critical irradiation dose rate as a function of temperature. For a given temperature, defect segregation into clusters occurs at dose rates above the critical one, whereas below the defects remain in a spatially homogeneous distribution. For the small migration energies typical of interstitials in alkali halides [13], it can be seen (curve 1) that at the usual experimental dose rates $P \leq 10^{-3}$ dpa/s instability occurs only at very low temperatures. In metals, however, instability may be quite significant. For $E_I^M = 0.68$ eV (dumbbell interstitials in Cu [19]), at a flux of 10^{-3} dpa/s, interstitial aggregation is expected to occur below 250 K, and for $E_I^M = 1$ eV even up to 370 K. Note that the latter estimate agrees qualitatively with the results obtained from Martin's model [3].

Since real experiments are performed in a limited interval of time, the instability diagram should be corrected at its low-temperature side. Despite the fact that Fig. 1 predicts that even very low fluxes should produce instability at rather low temperature, this instability may not become visible within the duration of the experiment. It may be shown that, at low temperatures, the characteristic time of the segregation process increases exponentially with decreasing temperature. In order to take this into account, the dotted line calculated from (17) has been included in Fig. 1. This shows the redefined critical dose rate P_c required to induce segregation at a characteristic rate larger than 10^{-2} s^{-1}. It can be seen that for Cu P_c is close to 10^{-4} dpa/s between about 100 K and 200 K, but increases at both lower and higher temperatures.
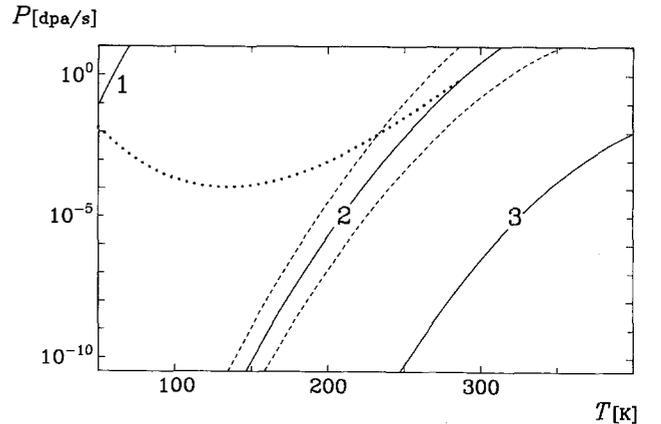
P[dpa/s]

Fig. 1. Instability diagram (irradiation intensity vs temperature) for $E_I^M = 0.1$ eV (curve 1), 0.6 eV (curve 2), and 1.0 eV (curve 3). Vacancies are assumed to be immobile. The elastic interaction energy differences considered are $\hat{E}_{II} - \hat{E}_{IV} = 0.2$ eV (full lines), 0.05 eV (upper broken line), and 0.8 eV (lower broken line). The dotted line corresponds to a finite segregation rate of 10^{-2} s^{-1}

The broken lines above and below the solid curve 2 in Fig. 1 demonstrate how a change in the asymmetry of the elastic interstitial–interstitial and interstitial–vacancy interactions affects the instability diagram. An increase (decrease) of the difference between the interaction energies by a factor of 4 shifts curve 2 only slightly downwards (upwards) parallel to the P axis.

If both interstitials and vacancies are mobile (i.e., if they have similar activation energies of diffusion as in the case of Au [19]), the analysis of the preceding paragraph has to be modified. In this case the segregation condition becomes more rigorous. Instead of the term $(\hat{E}_{II} - \hat{E}_{IV})$ in (14) we have now the term $[\hat{E}_{II} - \hat{E}_{IV} - (D_V/D_I)(\hat{E}_{IV} - \hat{E}_{VV})]$. Since $\hat{E}_{VV}$ is typically less than $\hat{E}_{IV}$, we have thus an additional stabilizing contribution in (14).

The absolute values of the defect interaction energies may vary substantially from material to material. It is known [15] that in alkali halides for nearest neighbours $E_{II} = 0.25$ eV, $E_{VI} = 0.05$ eV, and E_{VV} is even smaller. For metals $E_{II} \simeq 0.5$ eV is quite a common value [17, 20]. What is essential here is that even a *small* asymmetry in the interaction energies, say 0.05 eV, is enough to induce instability and segregation under sufficiently high fluxes. Such asymmetries appear to be a fairly common phenomenon in most materials.

Figure 2 shows dispersion curves, i.e., the characteristic segregation rate versus the reciprocal wavelength of the concentration fluctuations. The values $\omega(k)$ represented by these curves characterize the growth rates ($\omega > 0$) or damping rates ($\omega < 0$) of the density fluctuations. Further, they give qualitative information on the spatial distribution of aggregates of defects of the same kind. The wavelength of maximum amplification λ_{max} gives a rough estimate of the distance between such periodically arranged clusters.

The character of the dispersion curve is defined by the behaviour of the parameters $U_{II}(k)$ and $U_{VI}(k)$ entering (15).

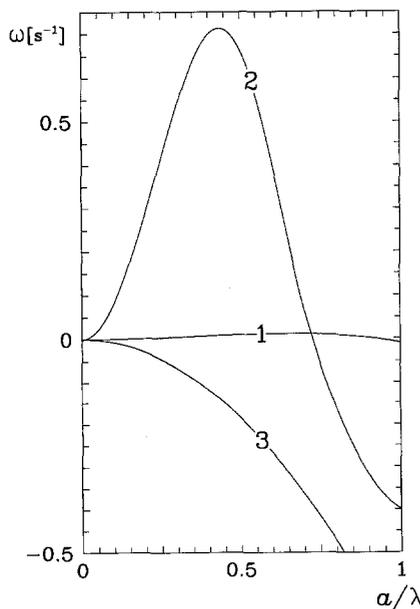Both parameters, which differ only by a factor $\hat{E}_{II}/\hat{E}_{VI}$, are proportional to $P^{-1/2}$ and decrease exponentially with increasing temperature as a result of the T dependence of D. Their dependence on k is defined by the k dependence of the structure factor $\tilde{f}(k)$, which is an oscillating function of the modulus of the wavevector. At low temperatures the U_{JK} are large compared to unity, and $\omega(k)$ is determined mainly by its dependence on k via $\tilde{f}(k)$. On the other hand, at high temperature, the U_{JK} are very small, so that ω is strictly negative and, near $k = 0$, adopts a parabolic behaviour (curve 3 in Fig. 2), i.e., all fluctuations are suppressed. In Fig. 2 we have plotted the dispersion curve for parameters typical of Cu. Note that, although ω is positive for both 170 K and for 250 K, in the former case the absolute values of ω are already very small, corresponding to a characteristic segregation time τ of $1/\omega_{max} \approx 500$ s. (In a plot similar to Fig. 1 this corresponds to a set of parameters in the vicinity of the dotted line.) A problem that is obvious from Fig. 2 as well as previous studies [3, 4] is that the fluctuations of maximum amplification have wavelengths $\lambda \approx 2a$, i.e., of the order of the interatomic spacing, which is in conflict with the assumption that we may use a *mesoscopic* model. This deficiency results directly from the assumed, very short-ranging interaction potential with an interaction radius $r_0 = a$. Thus, the modifications arising from a more realistic *long-range* interaction need be investigated in a more detailed study. (Note, however, that this problem does not affect our discussion of the instability conditions, since according to (16) the fluctuations near the stability limit have always long wavelengths.)

The impact on the stability behaviour of impurities acting as traps for interstitials is shown in Figs. 3 and 4. It is clear that, since traps result in a temporary localization of interstitials, they prevent interstitials from rapid recombination with vacancies. On the other hand, the trapped interstitials remain within the system (in contrast to point defects absorbed by dislocations) and may thus act as nuclei for interstitial clustering. So, at a given irradiation flux, impurities bring about an increase of the interstitial concentration via decreasing the effective diffusion coefficient of the interstitials. In order to
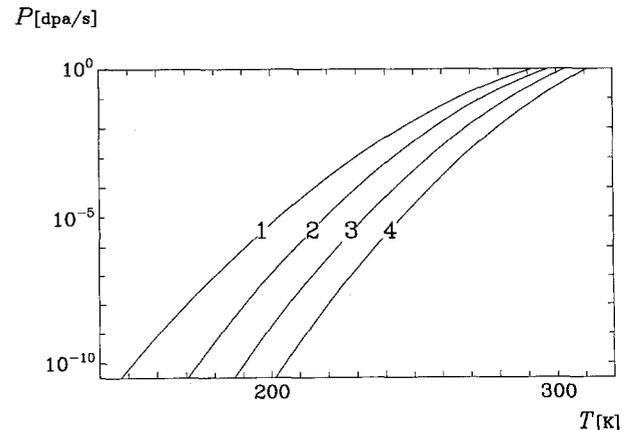

Fig. 2. Dispersion curves (segregation rate vs reciprocal wavelength of concentration fluctuations) for $E_I^M = 0.68$ eV, $\hat{E}_{II} - \hat{E}_{IV} = 0.2$ eV, and $P = 10^{-3}$ dpa/s. The temperatures considered are $T = 170$ K (curve 1), 220 K (curve 2), and 270 K (curve 3)

Fig. 3. Instability diagram in the presence of traps. The binding energies of interstitials to traps have been chosen as $E^B = 0$ (curve 1), 0.3 eV (curve 2), 0.4 eV (curve 3), and 0.5 eV (curve 4), the trap concentration as $C_{tr} = 10^{14}$ cm^{-3}, and the trapping radius as $R_{tr} = 2a$. The further parameters are the same as those used for the calculation of curve 2 in Fig. 1
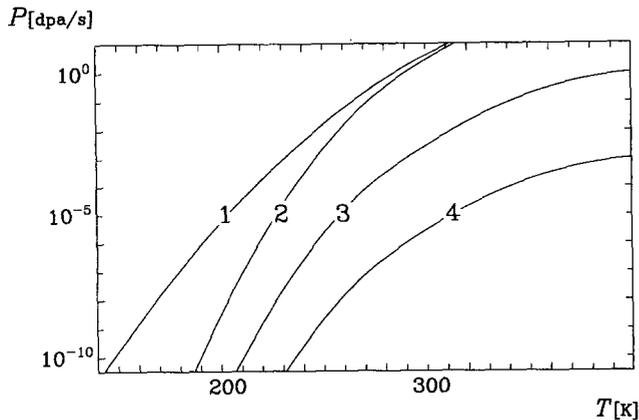
$P_{[\text{dpa/s}]}$

Fig. 4. Effect of the trap concentration on the instability diagram (binding energy $E^B = 0.4\,\text{eV}$; trap concentrations $C_{\text{tr}} = 0$ (curve 1), $10^{14}\,\text{cm}^{-3}$ (curve 2), $10^{17}\,\text{cm}^{-3}$ (curve 3), and $10^{20}\,\text{cm}^{-3}$ (curve 4); other parameters as in Fig. 3

investigate the influence of traps on the instability diagrams, it is possible to redefine the diffusion coefficient according to (19). Since it consists of two exponentials depending on different activation energies, we have at low temperatures

$$\tilde{D} \approx \frac{\nu \exp\{-(E_{\text{I}}^{\text{M}} + E^{\text{B}})/k_{\text{B}}T\}}{4\pi R_{\text{tr}} C_{\text{tr}}}. \tag{20}$$

This means that at low temperatures the diffusion process is controlled by the detrapping rate of interstitials, whereas at high temperatures interstitial diffusion becomes predominant. From Fig. 3 the conclusion can be drawn that in Cu, irrespective of the trap-binding energy, the influence of traps is not very important above about $\approx 350\,\text{K}$. Keeping in mind our corrections for the finite relaxation time (Fig. 1), this suggests that traps in not too high concentrations do not have a considerable effect on instability. However, if the trap concentration increases, a completely different picture emerges (Fig. 4). Since P_c is roughly proportional to D, an increase of the trap concentration by an order of magnitude decreases the critical dose rate in the trap-controlled regime by the same amount. In this case, diffusion that is not controlled by traps becomes predominant at higher temperatures only, such that segregation can occur in a larger temperature range. In other words, the main factor stimulating defect segregation via amplified density fluctuations is the concentration of traps rather than their binding energy. A high trap concentration can shift the stability boundary by such an extent that instability may occur in spite of a low interstitial migration enthalpy. This may occur even in alkali halides under usual experimental dose rates and at ambient temperatures.

3 Conclusions

As shown in this paper, a *sufficient* condition for aggregation of defects of the same kind resulting from the amplification of local density fluctuations at relatively low temperatures and/or high defect-production rates is provided by even small asymmetries in the elastic interaction energies of interstitial–interstitial and vacancy–interstitial pairs (although, at very low temperatures, segregation may not be achieved within the duration of true experiments). We may

thus conclude that the presence of point-defect sinks such as dislocations, as studied in [3, 4] is not essential for the occurrence of segregation. It is further shown that impurities capable of trapping mobile interstitials and thus reducing the effective interstitial mobility reduce considerably the critical irradiation flux P_c required for the segregation. Under these circumstances, the trap concentration rather than the trapping energy plays the decisive rôle.

For interstitials with a migration energy of $0.68\,\text{eV}$ (dumbbells in Cu), P_c is estimated to be about $10^{-5}\,\text{dpa/s}$ at $200\,\text{K}$. However, it rapidly increases with temperature, reaching $10^{-1}\,\text{dpa/s}$ at room temperature. For lower activation energies, say $0.1\,\text{eV}$, as typical of interstitials in alkali halides, it is difficult to achieve aggregation above $100\,\text{K}$, provided neither impurities nor dislocations are present.

Problems which are left for more detailed investigations are the following: *(i)* a more thorough treatment of the interactions between point defects that is based on a $E \propto r^{-3}$-type elastic interaction law [17], *(ii)* an analysis of the size distribution of the aggregates and its temporal evolution, *(iii)* the establishing of a linkage between the mesoscopic approach used here and the microscopic one described elsewhere [1, 8]. Note that a common feature of both approaches is that segregation becomes efficient at low temperatures. Whereas within the mesoscopic approach this process remains diffusion-controlled, in the microscopic approach it occurs even in the limit $D \to 0$ [7, 8].

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