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The influence of non-equilibrium fluctuations on radiation damage and recovery of metals under irradiation

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Abstract

In the conventional theory of radiation damage, it is assumed that the main effect of irradiation is due to formation of Frenkel pairs of vacancies and self-interstitial atoms (SIAs) and their clusters. The difference in absorption of vacancies and SIAs by primary or radiation-induced extended defects (EDs) is thought to be the main reason of microstructural evolution under irradiation. On the other hand, the recovery of radiation damage is thought to be driven exclusively by thermal fluctuations resulting in the vacancy evaporation from voids (void annealing) or dislocations (thermal creep) and in the fluctuation-driven overcoming of obstacles by gliding dislocations (plastic strain). However, these recovery mechanisms can be efficient only at sufficiently high temperatures. At lower irradiation temperatures, the main driving force of the recovery processes may be due to nonequilibrium fluctuations of energy states of the atoms surrounding EDs arising as a result of scattering of radiation-induced excitations of atomic and electronic structure at EDs. In the present paper, the mechanisms of nonequilibrium fluctuations that result in such phenomena as the void shrinkage under irradiation at low temperatures (or high dose rates), irradiation creep and irradiation-induced increase of plasticity under sub-threshold irradiation was considered.

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1. Introduction

A majority of radiation effects studies are connected with creation of radiation-induced defects in the crystal bulk. A subsequent evolution of microstructure is thought to be driven exclusively by thermally activated processes such as the point defect diffusion and evaporation from extended defects obeying essentially the same lows as those without irradiation. However, there is an increasing evidence of that the so called thermally activated reactions may be modified under irradiation. Thus, results of molecular dynamics (MD) simulation [1] have shown that vacancies can be emitted from voids not only by thermal fluctuations but also by the collision events in the vicinity of voids, which result in a biased formation of vacancies due to the lower energy barrier involved. A subsequent diffusion of the ejected vacancies away from the void under certain conditions may result in the radiation-induced void dissolution [2,3]. There is also some evidence

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from MD simulations that a biased formation of vacancies occurs also in the vicinity of the dislocation cores [4]. One of the technologically important consequences of this effect is a mechanism of irradiation creep [5], which is based on the radiation and stress induced difference in emission (RSIDE) of vacancies from dislocations differently oriented with respect to the external stress.

These are examples of the mechanisms based on the *radiation-induced production of Schottky defects* [6], which often act in the opposite direction as compared to the mechanisms based on *Frenkel pair production in the bulk*. Another important class of fluctuation-induced reactions includes the unpinning of dislocations from the obstacles during plastic strain. Defects formed under irradiation in the bulk act as additional pinning centers resulting in the well-known effect of radiation-induced hardening. On the other hand, there is less known but well established effect of the increase in plasticity of metals under sub-threshold irradiation [7,8].

In the present paper, these phenomena will be considered with account of the radiation-induced excitations of atomic structure such as focusing collisions and unstable Frenkel pair production near crystal defects. Effects due to the excitations of electronic structure will be shortly discussed in conclusion.

2. Void evolution with account of radiation-induced emission of vacancies

It is known that not all the energy of the primary knock-on atom (PKA) is spent on the production of stable defects. A considerable part of the PKA energy is spent on the production of *unstable Frenkel pairs* (UFP) [9] and *focusons* that can propagate through the lattice along the close packed directions by the focusing mechanism [10]. Focusons *transfer energy* along close packed directions of the lattice, but there is *no interstitial transport* by a focusing collision, which enlarges their range considerably as compared to that of a *crowdion*. The energy range in which focusons can occur has an upper limit E_F, which has been estimated to be about 60 eV for Cu, 80 eV for Ag and 300 eV for Au [10]. A focuson loses its energy continuously, which determines its propagation range. If the relative energy loss per hit is ε_F then a focuson with initial energy E, will have the energy E_v after propagating at a distance $l_F^v(E, E_v)$ given by

$$l_F^v(E, E_v) = l_F^0 \ln(E/E_v),$$
$$\text{with } l_F^0 = b/\varepsilon_F, \text{ and } \varepsilon_F \approx 10^{-2} \div 10^{-1}, \tag{1}$$

where $b \approx 0.3$ nm is the atom spacing along the close packed directions. In an ideal lattice a focuson *does not produce defects* along its path (Fig. 1(a)). However, if the focuson has to cross an extended defect (ED), a vacancy may be produced in its surroundings (Fig. 1(b)) as has been demonstrated in [1] by means of MD simulations. The focuson propagation range in ideal lattice, l_F^0, decreases with increasing lattice temperature due to random thermal atomic displacements. Thus, for Au it has been estimated to decrease from 66b at 0 K to 39b at 340 K and to 19b at 1190 K [10]. So the vacancy

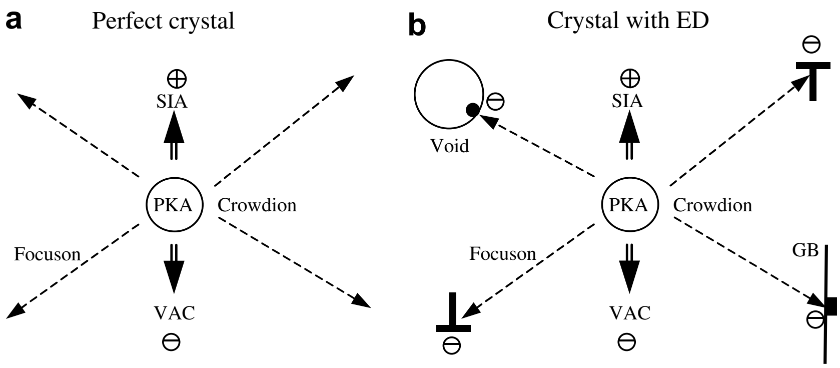

Fig. 1. Illustration of different point defect production schemes in perfect and real crystals: (a) Frenkel pair formation in the bulk by short-ranged crowdions: SIA is the self-interstitial atom, VAC is the vacancy, arrows show the propagation directions of focusons that do not produce defects in ideal lattice; (b) vacancy formation at extended defects due to interaction of long-ranged focusons with voids, dislocations (⊥) and grain boundaries (GB).

production by this mechanism decreases with increasing temperature.

Another mechanism of the radiation-induced vacancy emission from EDs, which is essentially temperature independent, is based on the production of unstable Frenkel pairs (UFPs) near EDs [5]. An ED such as a void or a dislocation is surrounded by a region of a radius R_{cap}, in which a point defect is unstable since it is captured by the ED athermally [9]. The capture radius for self-interstitial atoms is larger than the one for vacancies due to the lower migration energy and stronger interaction with ED. If a regular atom in the region $R^{\mathrm{v}}_{\mathrm{cap}} < r < R^{\mathrm{i}}_{\mathrm{cap}}$ gets an energy $E > E_{\mathrm{v}}$ it may move to an interstitial position, where it can be athermally captured by the ED, leaving behind it a stable vacancy. Since the capture time is about 10^{-11}–10^{-12} s [5], the process can be described as an effective emission of a vacancy by the ED due to its interaction with an UFP. As the energy of the system is increased as a result of vacancy formation, the minimum transferred energy, E_{v}, should exceed the energy of vacancy formation at a particular ED.

These mechanisms have been incorporated in the rate theory [2,5] by modifying the so called *local equilibrium concentrations of vacancies*, $c^{\mathrm{eq}}_{\mathrm{v}}$, which are determined by the rates of vacancy emission from EDs due to thermal or radiation-induced fluctuations of energy states of atoms surrounding the EDs. Generally, $c^{\mathrm{eq}}_{\mathrm{v}}$ is given by the sum of the thermal and the radiation-induced constituents:

$$c^{\mathrm{eq}}_{\mathrm{v}} = c^{\mathrm{th}}_{\mathrm{v}} + c^{\mathrm{irr}}_{\mathrm{v}}, \text{ and } c^{\mathrm{th}}_{\mathrm{v}} = \exp\left(-\frac{E^{\mathrm{f}}_{\mathrm{v}}}{k_{\mathrm{B}}T}\right), \tag{2}$$

$$c^{\mathrm{irr}}_{\mathrm{v}} = \frac{K_{\mathrm{F}} b l^0_{\mathrm{F}}}{D_{\mathrm{v}}} + \frac{K_{\mathrm{UFP}} b (R^{\mathrm{i}}_{\mathrm{cap}} - R^{\mathrm{v}}_{\mathrm{cap}})}{D_{\mathrm{v}}}, \tag{3}$$

where K_{F} is the effective production rate of focusons [2] and K_{UFP} is the UFP production rate [5], $E^{\mathrm{f}}_{\mathrm{v}}$ is the vacancy formation energy at a given ED and D_{v} is the vacancy diffusion coefficient.

Comparison of thermal and radiation-induced equilibrium PD concentrations at a flat surface for a typical neutron flux (Fig. 2(a)) shows that the latter dominates completely at temperatures below 0.5 *TM*, where a majority of radiation effects is observed.

Now the void growth (or shrinkage) rate is given by the usual expression [2]

$$\frac{\mathrm{d}R}{\mathrm{d}t} = \frac{1}{R}\left[Z^{\mathrm{V}}_{\mathrm{v}} D_{\mathrm{v}} \bar{c}_{\mathrm{v}} - Z^{\mathrm{V}}_{\mathrm{v}} D_{\mathrm{v}} c^{\mathrm{eq}}_{\mathrm{v}} - Z^{\mathrm{V}}_{\mathrm{i}} D_{\mathrm{i}} \bar{c}_{\mathrm{i}}\right], \tag{4}$$

where is the void capture efficiency for vacancies (subscript 'v') and SIAs (subscript 'i'), is the mean concentration of point defects determined by the rate equations. The product $D_{\mathrm{v}} c^{\mathrm{eq}}_{\mathrm{v}}$ is the *effective vacancy self-diffusion coefficient*, which determines the rate of the void *thermal* and *radiation-induced* dissolution. The latter does not depend on temperature in contrast to the void growth rate due to the biased absorption of vacancies from the bulk, which decreases with decreasing irradiation temperature due to enhancement of the point defect recombination

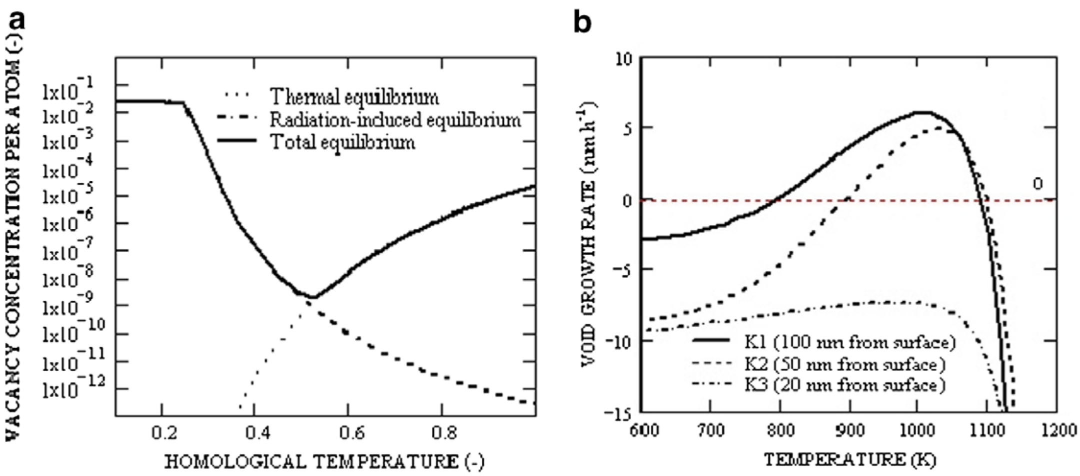

Fig. 2. (a) Comparison of thermal and radiation-induced equilibrium PD concentrations at a flat surface for a typical neutron dpa damage, $K = 10^{-6}\ \mathrm{s}^{-1}$; (b) void growth rates during Ni^{+} ion bombardment of Ni calculated by Eq. (4) for different displacement rates and distances from the surface: $K1 = 10^{-2}\ \mathrm{s}^{-1}$ (100 nm), $K2 = 5 \times 10^{-2}\ \mathrm{s}^{-1}$ (50 nm), $K3 = 20 \times 10^{-2}\ \mathrm{s}^{-1}$ (20 nm). $l^0_{\mathrm{F}} \approx 10b$, $E_{\mathrm{F}} = 60$ eV, $E_{\mathrm{d}} = 30$ eV, $E^{\mathrm{f}}_{\mathrm{v}} = 3$ eV, $k_{\mathrm{FD}} = 0.1$, dislocation bias for SIAs, $\delta_{\mathrm{d}} = 0.1$, other material parameters are given in [3].

in the bulk. As a result, the net growth rate may become negative below some temperature (or above some dose rate).

Some experiments indicate that voids can shrink with decreasing temperature (or increasing dose rate) after they have been formed under more favorable conditions [3,11,12]. According to Steel and Potter [3], voids formed during Ni^{+} ion bombardment of Ni at 923 K shrink rapidly when subjected to further bombardment at temperatures between 298 and 823 K. Between 923 and 973 K, the void growth proceeds to fluences near $3 \times 10^{21}\ m^{-2}$and is followed by the shrinkage as the voids distance to the surface decreases due to sputtering of the surface by the ion beam. The authors have attempted to explain the observations using the rate theory modified to include the interstitials injected by the ion beam. However, this effect is negligible due to a very low 'production bias' introduced by injected SIAs (about 0.1%). Fig. 2(b) shows the results of the present model, which neglects the production bias but takes into account the increasing dose rate with decreasing distance to the surface as it was reported in Ref. [3]. The production rates of focusons, UFPs and stable freely migrating point defects have been expressed via the displacement rate per atom, K, by the following equations

$$K_{\mathrm{F}} \approx K \frac{E_{\mathrm{d}}}{E_{\mathrm{F}}} \int_{1}^{E_{\mathrm{v}}^{\mathrm{f}}/E_{\mathrm{F}}} \ln x \ln\left(x \frac{E_{\mathrm{F}}}{E_{\mathrm{v}}^{\mathrm{f}}}\right) \mathrm{d}x,$$

$$K_{\mathrm{UFP}} \approx K \left(\frac{E_{\mathrm{d}}}{E_{\mathrm{v}}^{\mathrm{f}}}\right)^{\frac{3}{2}}, \quad K_{\mathrm{FD}} \approx K \cdot k_{\mathrm{FD}}, \tag{5}$$

where k_{FD} is the cascade efficiency for producing freely migrating point defects. Assuming $R_{\mathrm{cap}}^{\mathrm{i}} - R_{\mathrm{cap}}^{\mathrm{v}} \approx b$ one finds that the UFP effect dominates over the focuson effect if $l_{\mathrm{F}}^{0} < 50\mathrm{b}$, which is likely to be the case in the temperature range under investigation. It can be seen in Fig. 2(b) that the void growth rate becomes negative with decreasing temperature or the distance from the surface in agreement with experimental data.

A more general trend of the void swelling is its saturation with increasing irradiation time, which is a common feature for a number of materials [12,13]. The present mechanism of radiation-induced void shrinkage may be responsible for the swelling saturation as the void sink strength increases with time thus decreasing a positive constituent to the void growth rate [2].

The saturation phenomenon is intrinsically connected with a void ordering observed in very different radiation environments ranging from metals to ionic crystals [12–19]. It's tempting to suppose that ordering phenomena might be a consequence of the *energy transfer* along the close packed directions provided by focusons. If the focuson length, l_{F}, were larger than the void spacing, l, the voids would shield each other from focuson fluxes along the close packed directions, which would provide a driving force for the void ordering. A serious objection to this hypothesis comes from the temperature dependence of l_{F} estimated in Ref. [10]. Further molecular dynamic simulations of the focuson propagation at various temperatures would be helpful to check the hypothesis.

3. Irradiation-induced dislocation climb and glide

There are two main mechanisms of dislocation movement, namely, dislocation climb and glide, which result in a plastic strain under applied stress. Dislocation climb is a relatively slow diffusion limited process resulting in material creep at high temperatures when the thermal vacancy self-diffusion is effective. Irradiation is known to enhance the creep rate, which becomes temperature independent below $\sim 0.5\ T_{\mathrm{M}}$ under typical neutron fluxes. Conventional *irradiation creep* models are based on the so called stress-induced preferential absorption (SIPA) of radiation-produced point defects by dislocations [20] or other extended defects [21] differently oriented with respect to the external stress. Consequently, these models can yield a significant and temperature independent irradiation creep only when the bulk recombination of point defects is negligible. An alternative mechanism of irradiation creep proposed recently [5] is based on the radiation and stress induced difference in emission (RSIDE) of vacancies from dislocations of different orientations with respect to the external stress. This difference is due to the difference in vacancy formation energies, which is proportional to the external stress. The resulting creep rate may be approximated by a simple expression:

$$\dot{\varepsilon}_{\mathrm{RSIDE}} \approx \rho_{\mathrm{d}} D_{\mathrm{v}} c_{\mathrm{v}}^{\mathrm{irr}} \frac{\sigma\omega}{E_{\mathrm{v}}^{\mathrm{f}}}, \tag{6}$$

which is very similar to the usual expression for the thermal creep rate [5], but the radiation-induced self-diffusion coefficient, $D_{\mathrm{v}}c_{\mathrm{v}}^{\mathrm{irr}}$, stands here for the thermal self-diffusion coefficient, $D_{\mathrm{v}}c_{\mathrm{v}}^{\mathrm{th}}$, and the vacancy formation energy , $E_{\mathrm{v}}^{\mathrm{f}}$, stands for $k_{\mathrm{B}}T$. Consequently, the RSIDE creep is essentially temperature independent in contrast to the SIPA creep rate [5]. Another difference between the RSIDE and SIPA models is that the RSIDE creep should be observed under sub-threshold irradiation, when production rate of stable Frenkel pairs is zero. Sub-threshold irradiation could be used for experimental verification of the RSIDE mechanism.

Such verification has not been performed yet to our knowledge, but there is experimental evidence of plasticity increase under low temperature sub-threshold electron or gamma irradiation, which has been discovered in early sixties [7] and investigated extensively thereafter (see e.g., [8]). Single crystals of Zn, Sn, In and Pb have been irradiated at liquid nitrogen temperature (78 K) with electron flux density ranging from 10^{17} to 10^{18} $\mathrm{m^{-2}\,s^{-1}}$ and energies below and above the threshold displacement energies, the latter being 0.7 MeV (Zn), 0.8 MeV (Sn, In) and 1.2 MeV (Pb). At such low temperatures plastic strain occurs via dislocation glide, the rate of which is limited by thermally activated unpinning of dislocations from local obstacles. The over-threshold irradiation has resulted in the well-known effect of radiation-induced hardening due to formation of additional pinning centers. But with decreasing beam energy below the threshold level the plastic strain rate under irradiation increased as compared to that prior or after irradiation.

The thermally activated plastic strain rate under external stress, σ, can be described approximately by the following equation:

$$\begin{aligned}\dot{\varepsilon}_{\mathrm{T}} &= bL\rho_{\mathrm{d}}^{0}w_{\mathrm{T}},\\ w_{\mathrm{T}} &= \omega_0 \exp\left\{-\frac{U_{\mathrm{a}}-(\sigma-\sigma_{\mathrm{i}})\upsilon_{\mathrm{a}}}{k_{\mathrm{B}}T}\right\},\\ \sigma_{\mathrm{i}} &\approx \frac{\mu b}{2\pi}\sqrt{\rho_{\mathrm{d}}},\end{aligned} \tag{7}$$

where w_{T} is the frequency of the dislocation 'jumps' over the obstacle, ω_0 is the frequency factor, ρ_{d}^{0} is the density of gliding dislocations, ρ_{d} is the total dislocation density, L is the mean distance between the pinning centers, μ is the shear modulus, U_{a} is the binding energy between a dislocation and the pinning center, σ and σ_{i} are the external and internal stresses, respectively, υ_{a} is the activation volume, so that $U_{\mathrm{a}}^{\sigma} = U_{\mathrm{a}} - (\sigma-\sigma_{\mathrm{i}})\upsilon_{\mathrm{a}}$ is the activation energy of the dislocation 'jump' over the obstacle. This energy can be obtained by atoms surrounding the pinning centre either due to the thermal fluctuations (which occurs with a frequency w_{T}) or due to the scattering of radiation-produced excitations such as focusons and random recoil events. In the case of electron irradiation, the frequency of focuson-induced jumps, w_{F}, can be estimated as follows. The production rate of focusons with initial energy E (per atom), $K_{\mathrm{F}}(E)$, is given by [2]

$$K_{\mathrm{F}}(E) = j_{\mathrm{e}}P_{\mathrm{F}}\frac{\mathrm{d}\sigma}{\mathrm{d}E}\,\mathrm{d}E, \tag{8}$$

where j_{e} is the electron flux, $\mathrm{d}\sigma/\mathrm{d}E$ is the differential cross section for producing a PKA of energy and P_{F} is the probability of the focuson production by a PKA [10]. The focusons can transfer energy to the pinning center along $z-2$ close packed directions, where z is the coordination number. Accordingly, the number of focusons of energy higher than U_{a}^{σ} coming to the pinning center per unite time is given by the integration of the product of $K_{\mathrm{F}}(E)$ and the number of atoms in cylindrical region of the length $l_{\mathrm{F}}^{\mathrm{v}}(E, U_{\mathrm{a}}^{\sigma})$ and radius $r_{\mathrm{d}} \approx b$ and the ratio $(z-2)/z$ over the PKA energy:

$$\begin{aligned}w_{\mathrm{F}}(j_{\mathrm{e}}, E_{\mathrm{e}}, U_{\mathrm{a}}^{\sigma}) &= j_{\mathrm{e}} \times l_{\mathrm{F}}^{0} \times \frac{\pi r_{\mathrm{d}}^{2}}{\upsilon}\frac{z-2}{z}\\ &\quad\times \int_{U_{\mathrm{a}}^{\sigma}}^{E_{\mathrm{m}}(E_{\mathrm{e}})} \ln(E/U_{\mathrm{a}}^{\sigma})P_{\mathrm{F}}\frac{\mathrm{d}\sigma}{\mathrm{d}E}\,\mathrm{d}E,\end{aligned} \tag{9}$$

$$E_{\mathrm{m}} = \frac{2E_{\mathrm{e}}(E_{\mathrm{e}}+2m_{\mathrm{e}}c^{2})}{Mc^{2}}, \tag{10}$$

where E_{e} is the electron beam energy, m_{e} and M are the electron and the target masses, respectively, c is the light velocity and E_{m} is the maximum transferred energy, υ is the atomic volume. The frequency of focuson-induced jumps decreases with increasing temperature due to decreasing focuson propagation range.

The temperature independent contribution to the dislocation unpinning comes from random recoil events (RRE). The frequency of RRE-induced jumps, w_{RRE}, can be estimated in a similar way to result in the following expression

$$w_{\mathrm{RRE}}(j_{\mathrm{e}}, E_{\mathrm{e}}, U_{\mathrm{a}}^{\sigma}) = j_{\mathrm{e}} \times \frac{4\pi r_{\mathrm{d}}^{3}}{3\upsilon}\int_{U_{\mathrm{a}}^{\sigma}}^{E_{\mathrm{m}}(E_{\mathrm{e}})}\frac{\mathrm{d}\sigma}{\mathrm{d}E}\,\mathrm{d}E, \tag{11}$$

The total rate of plastic strain under irradiation is given by the sum

$$\begin{aligned}\dot{\varepsilon}(j_{\mathrm{e}}, E_{\mathrm{e}}, U_{\mathrm{a}}^{\omega}, T) &= bL\rho_{\mathrm{d}}^{0}[w_{\mathrm{F}}(j_{\mathrm{e}}, E_{\mathrm{e}}, U_{\mathrm{a}}^{\sigma})\\ &\quad + w_{\mathrm{RRE}}(j_{\mathrm{e}}, E_{\mathrm{e}}, U_{\mathrm{a}}^{\sigma}) + w_{\mathrm{T}}(T, U_{\mathrm{a}}^{\sigma})].\end{aligned} \tag{12}$$

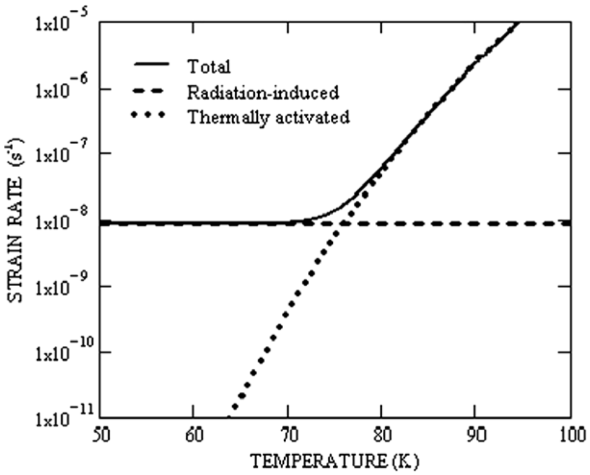

Fig. 3. Temperature dependence of the strain rate Zn under sub-threshold electron irradiation given by Eq. (12) at the following irradiation and material parameters: $J_e = 5 \times 10^{17}\ \mathrm{m}^{-2}\ \mathrm{s}^{-1}$, $E_e = 0.5$ MeV, $\sigma = 1$ MPa, $U_a = 0.3$ eV, $\rho_d = 10^{14}\ \mathrm{m}^{-2}$, $l_F^0 \approx 50b$, $L = 10^{-7}$ m.

Temperature dependence of strain rate calculated by Eq. (12) for the case of sub-threshold electron irradiation of Zn is shown in Fig. 3. It can be seen that irradiation-induced strain rate exceeds the thermally activated one below 80 K in agreement with experimental data. The contributions from the focussing and random recoil events at these low temperatures are of the same order of magnitude.

4. Summary and outstanding problems

Nonequilibrium fluctuations of energy states of the atoms surrounding crystal defects arise as a result of their interaction with radiation-induced excitations in the ionic system. These fluctuations result in radiation-induced recovery processes such as the void shrinkage and plastic strain, which should be taken into account in modeling of the microstructural evolution under irradiation.

Excitations in the ionic system considered in the present paper are not the only reason for the acceleration of fluctuation-driven processes. When swift ions or electrons bombard a solid target, they lose energy mostly by creating electronic excitations. These excitations transfer their energy to the lattice (due to electron-phonon coupling) and to the crystal defects (due to electron-defect coupling) resulting in the ‘thermal’ spikes in the corresponding sub-systems. These spikes may be much more powerful in the defect regions than those in the perfect lattice due to the large difference in the corresponding coupling times (especially at low temperatures when the electron-phonon scattering becomes weak). As a result, the thermally activated processes (such as defect emission, migration, etc.) are enhanced, which also should be taken into account in modeling of the radiation-induced microstructural evolution. This will be done in subsequent work.

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