Internal sinks and the smoothing of the surface structure in solids under irradiation.

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Abstract

We consider in the article the influence of the irradiation and the internal sinks of the point defects on the rate of the flattening of the surface structure in solids. The irradiation produces only the additional external sources of point defects (vacancies and interstitial atoms). The general system of equations is formulated. The solution of the system on the stationary stage of the process is found. It is shown that depending on the values of some parameters of the solid the irradiation can increase or decrease the rate of the surface flattening.
The reason of smoothing of the surface structure in solids is the difference of the equilibrium point defect concentration on the convex and concave parts of the surface. As a result of this difference there are the diffusion flows of point defects which tend to redistribute the point defects to attain the thermodynamically equilibrium state corresponding the flat surface. The expression for the rate of surface flattening without irradiation was received in [1]. Using this expression it is possible to find the surface and volume diffusion coefficients analyzing experimentally the rate of surface smoothing as a function of parameters of the roughness of the surface [2]. In article [3] the irradiation of the solid was introduced into the process of surface flattening. The irradiation was considered only as a external source of point defects. It was shown in [3] that because the depth of allocation of external source of interstitial atoms is bigger then the depth of allocation of external source of vacancies the irradiation increase the rate of the surface flattening. This conclusion can be changed if we take into account the internal sinks of point defects. Our article is devoted to this problem: the evolution of the surface structure of solids with external sources and internal sinks of point defects.

The main system of equations which describes the evolution of surface structure was thoroughly analyzed in [3,4]. For small enough intensity of irradiation we can disregard the annihilation of point defects (vacancies and interstitial atoms). In this case we can find the contribution of vacancies and interstitial atoms into the rate of surface flattening separately. Let us find the contribution of vacancies.

Following the article [3] we assume that the surface of solid is described by the equation:

\[ z = f(x, t) = \sum_n z_n(t) e^{-i\omega x_n} \]  

where prime in (1) means that \( n \neq 0 \) in the sum (i.e. the equilibrium surface is \( z = 0 \)), \( \omega = 2\pi/\lambda \), \( \lambda \) is the distance between two identical roughness on the surface, \( z \)-axis is directed into the bulk of the solid. We will consider \( z_n/\lambda \) as a small parameter and will disregard the second order terms in \( z_n/\lambda \) in all equations below. Under this assumption the curvature of the surface (1) is equal to:

\[ K(x, t) \approx -\frac{\partial^2 f}{\partial x^2} = \sum_n z_n(t) n^2 \omega^2 e^{-i\omega x_n} \]  

where the curvature is proportional to the gradient of the surface.
To find the diffusion (surface and volume) flows of vacancies which modify the surface structure in solids we should find the volume $c_{\text{vac}}$ and the surface $u_{\text{vac}}$ vacancy concentrations as the functions of space and time co-ordinate.

We will find the concentration as the power series in small parameter $z_n/\lambda$ to within the first-order terms in $z_n/\lambda$. With the surface structure in the form (1) such series are

$$u_{\text{vac}}(x, t) = u_{\text{vac}}^{(0)}(t) + \sum_n u_{\text{vac}}^{(n)}(t)e^{-in\omega x} \quad (3)$$

$$c_{\text{vac}}(x, z, t) = c_{\text{vac}}^{(0)}(z, t) + \sum_n c_{\text{vac}}^{(n)}(z, t)e^{-in\omega x} \quad (4)$$

where $c_{\text{vac}}^{(0)}$, $u_{\text{vac}}^{(0)}$ have zero order in $z_n/\lambda$ and $c_{\text{vac}}^{(n)}$, $u_{\text{vac}}^{(n)}$ have the first order in $z_n/\lambda$.

In was shown in [3] that under real conditions the main system of equations describing the spatial and temporal distribution of vacancies $c_{\text{vac}}(x, z, t)$, $u_{\text{vac}}(x, t)$ takes the form:

$$\frac{du_{\text{vac}}^{(0)}}{dt} = -\nu(u_{\text{vac}}^{(0)}(t) - u_{0,\text{vac}}) + \frac{D_{V,\text{vac}}}{a} \frac{\partial c_{\text{vac}}^{(0)}(z, t)}{\partial z} \bigg|_{z=f(x, t)}$$

$$\frac{\partial c_{\text{vac}}^{(0)}}{dt} = D_{V,\text{vac}} \Delta c_{\text{vac}} + I(x, z, t) - \delta_{\text{vac}}(c_{\text{vac}} - c_0) + \frac{D_{V,\text{vac}}}{kT} \nabla c_{\text{vac}} \nabla U(x, z, t)$$

$$\frac{\partial u_{\text{vac}}^{(0)}}{dt} = D_{V,\text{vac}} \frac{\partial c_{\text{vac}}^{(0)}(z, t)}{\partial z} \bigg|_{z=f(x, t)} = (\frac{\partial D_{V,\text{vac}}}{a} c_{\text{vac}}^{(0)}(z = f(x, t), t) - \frac{\alpha}{\tau_S} u_{\text{vac}}^{(0)}(t))$$

$$\frac{df(x, t)}{dt} = D_{S,\text{vac}} a \Delta u_{\text{vac}} + a \frac{D_{S,\text{vac}}}{kT} \nabla S(u_{\text{vac}} \nabla S U(x, z, t)) + D_{V,\text{vac}} \frac{\partial u_{\text{vac}}^{(0)}}{dt} \bigg|_{z=f(x, y, t)}$$

where index ”vac” indicates that the quantities correspond to vacancies; $D_{V,\text{vac}}$ is the volume diffusion coefficient of vacancies; $D_{S,\text{vac}}$ is the surface diffusion coefficient; $\nu$ is the frequency of vacancy absorption by the surface; $I$ is the intensity of a source of vacancies - the number of vacancies generated by irradiation per lattice site and per unit time; $\delta_{\text{vac}}$ is the rate of absorption of vacancies by internal sinks (1/\delta_{\text{vac}} is the vacancy mean free time); $c_0$ is the vacancy concentration far from the surface (the equilibrium vacancy concentration for the flat surface); $c_K = c_0 + c_0 \gamma \Omega K / kT$ is the equilibrium volume vacancy concentration corresponding to the surface with the curvature $K$; $\gamma$ is the surface tension; $\Omega = a^3$, $a$ is the lattice constant; $k$ is
the Boltzmann constant; $T$ is the temperature of the solid; $\Delta_S$ is the surface part of Laplacian $\Delta = \partial_x^2 + \partial_y^2 + \partial_z^2$; $\vec{n}$ is the inward normal to the surface of the solid; $\beta D_{V,vac} = D'$ determines the last jump for a vacancies emerging from the bulk to the free surface, $\beta$ is the dimensionless coefficient that takes into account the presence of a potential barrier for a vacancy emerging at the sample surface $(0 < \beta \leq 1)$; $\tau_S = 1/\nu$; $d\frac{f(x,y,t)}{dt}_{vac}$ is the contribution of vacancies into the rate of the surface smoothing; $U(x,z,t)$ is the elastic potential energy of the vacancies in the elastic field produced by the curved surface. Without irradiation the terms containing $U(x,z,t)$ are very small but under some conditions for solids under irradiation they can be important.

The potential energy $U(x,z,t)$ is given by the equation [5]:

$$U(x,z,t) = |\Omega_{vac}| p(x,z,t)$$

where $\Omega_{vac} < 0$ is the decreasing of the total volume of solid under the appearing of one vacancy, $p(x,z,t)$ is the hydrodynamic pressure in solid. For isotropic case the pressure $p(x,z,t)$ is the solution of Laplace equation [5]:

$$\Delta p(x,z,t) = 0$$

with the boundary condition:

$$p(x,z = f(x,t)) = \gamma K(x,t)$$

Then for the surface of type (1) the potential energy is:

$$U(x,z,t) = |\Omega_{vac}| \gamma \sum_{n \neq 0} z_n(t) \omega^2 n^2 e^{-i\omega x} e^{-\omega x}$$

We have used in the system (5) that under the condition $\nu >> \frac{D_{S,vac}}{x^2}$ it is possible to take $\sum_{n \neq 0} u_{vac}^{(n)}(t) \exp(-i\omega x) = u_K - u_0$ [3], where $u_K = u_0 + u_0 \gamma \frac{\Omega K}{K_K}$ is the equilibrium surface vacancy concentration corresponding to the surface with the curvature $K$.

We shall find the quasi-stationary solution of the system (5) [3]. We have such solution for a long experimental time $t >> \tau_{max} = max \left( \frac{\lambda^2}{D_{S,vac}}, \frac{l^2}{D_{V,vac}} \right)$ (where $l$ is the characteristic depth at which the source of vacancies is located, $\lambda$ is distance between two identical structure on the surface) and for a long characteristic time of variation of roughness of the surface ($f/\dot{f} >> \tau_{max}$) because in this case we can neglect the time derivative $\partial c_{vac}/\partial t$ in the first two
equations of system (5) and obtain the quasi-stationary system to within the terms $\sim \tau_{\text{max}}/t$. The time $t$ is the parameter at this quasi-stationary stage. In other words, we must first determine the quasi-steady-state fluxes for a given shape of the surface relief and then find a closed equation determining the time variation of the surface structure on the basis of these fluxes.

To solve the system of equations (5) we introduce the new variable:

$$\xi = z - f(x, t)$$

and the new function

$$V(x, \xi, t) = V(0)(\xi, t) + \sum_{n \neq 0} V^{(n)}(\xi, t)e^{-\imath n\omega x}c_{\text{vac}}(x, z, t) - c_K(x, t)$$

Then the system of equations (5) on the quasi-stationary stage takes the form:

$$\begin{align*}
-\nu(u^{(0)}_{\text{vac}}(t) - u_{0,\text{vac}}) + \frac{D V^{(0)}_{\text{vac}}}{a} \frac{\partial V^{(0)}(\xi, t)}{\partial \xi} \bigg|_{\xi = 0} \\
\Delta V - f_{xx}V_{\xi} - 2f_xV_{x\xi} = -c_{K,xx} - \frac{I(x, \xi)}{D V_{\text{vac}}} + \frac{\delta_{\text{vac}}}{D V_{\text{vac}}} (V + c_K - c_0) - \frac{1}{kT} \hat{\nabla}V \hat{\nabla}U \\
D_{V,\text{vac}} \frac{\partial V^{(0)}(\xi, t)}{\partial \xi} \bigg|_{\xi = 0} = \left( \frac{\beta D V_{\text{vac}}}{a} V^{(0)}(\xi = 0, t) - \frac{\omega}{\tau_s}(u^{(0)}_{\text{vac}}(t) - u_0) \right) \\
V^{(n)}(x, \xi = 0) = 0 \\
\frac{df(x, t)}{dt} \bigg|_{\text{vac}} = D_{S,\text{vac}} a \Delta_S u_{\text{vac}} + a \frac{D_{S,\text{vac}}}{kT} \hat{\nabla}_S(u_{\text{vac}} \hat{\nabla}_S U(x, \xi, t)) + D_{V,\text{vac}} \frac{\partial V}{\partial \xi} \bigg|_{\xi = 0}
\end{align*}$$

We take the source of vacancies in the form of $\delta$-function:

$$I(x, \xi) = I_0 \Omega \delta(\xi - l)$$

where $I_0$ is the number of vacancies emerging at the surface per unit time and per unit area. When the sample is bombarded by ions, the profile of generated point defects has a Gaussian form, and (7) is a good approximation for a narrow distribution of defects.

We find the solution of the first four equation of the system (12) with the accuracy to within $z_n/\lambda$ [3]. Then we substitute this solution into the fifth equation of the system (12) and find the contribution of vacancies into the rate of the surface smoothing:

$$\frac{dz_n(t)}{dt} \bigg|_{\text{vac}} = -z_n(t)D_{S,\text{vac}} a \frac{\gamma \Omega}{kT} \omega^4 n^4 - z_n(t)D_{V,\text{vac}} a \frac{\gamma \Omega}{kT} \omega^2 n^2 \left( \frac{\omega^2 n^2 + q_{\text{vac}}^2}{\mu_{\text{vac}}} \right) - D_{S,\text{vac}} \frac{l_0 \gamma \Omega \mu_{\text{vac}}}{kT} z_n(t) \omega^4 n^4 e^{-l q_{s,\text{vac}}} + I_0 \Omega \mu_{\text{vac}} z_n(t) \left( e^{-l q_{s,\text{vac}}} - e^{-\mu_{\text{vac}} l} \right)$$

(14)
where \( q_{s,vac} = \sqrt{\frac{\delta_{vac}}{D_{V,vac}}} \), \( \mu_{vac} = \sqrt{\omega^2 n^2 + q_{s,vac}^2} \) and the condition \( a q_s / \beta \) was used.

For interstitial atoms the system of equation (12) has the form [3]:

\[
\begin{cases}
-\nu u_{in}^{(0)}(t) + \frac{D_{V,in}}{a} \frac{\partial V^{(0)}(\xi,t)}{\partial \xi} \bigg|_{\xi=0} = 0 \\
\Delta V - f_{sx} V_\xi - 2 f_{sx} V_x = -\frac{I(x,\xi)}{D_{V,in}} + \frac{\partial_{in}}{D_{V,in}} V - \frac{1}{kT} \nabla^2 V \nabla U \\
D_{V,in} \frac{\partial V^{(0)}(\xi,t)}{\partial \xi} \bigg|_{\xi=0} = (\beta \frac{D_{V,in}}{a} V^{(0)}(\xi = 0, t) - \frac{a}{\gamma} u_{in}^{(0)}(t)) \\
V^{(v)}(x, \xi = 0) = 0 \\
\frac{\partial f(x,t)}{\partial t} \bigg|_{in} = a \frac{D_{s,in}}{kT} \nabla S(u_{in} \nabla S U(x, \xi, t)) + D_{V,in} \frac{\partial V}{\partial \xi} \bigg|_{\xi=0}
\end{cases}
\tag{15}
\]

where we have taken into account that \( c_{0,in} \approx 0 \) (i.e. \( c_{0,in} << c_{0,vac} \)); the index “in” in system (15) indicates that the quantities correspond to interstitial atoms. Solving the system (15) we find the contribution the interstitial atoms into the rate of the surface flattening:

\[
\frac{d z_n(t)}{dt} \bigg|_{in} = -I_0 \Omega \mu_{in} z_n(t) \left( e^{-\ln q_{s,in} - \mu_{in} t} - \mu_{in} \right) - D_{s,in} \frac{I_0 \Omega}{\nu} \left| \frac{\Omega_{in}}{kT} \right| z_n(t) \omega \mu^4 e^{-\ln q_{s,in} - \mu_{in} t} + \mu_{vac} \left( e^{-\ln q_{s,vac} - \mu_{vac} t} \right)
\]

(16)

where \( q_{s,in} = \sqrt{\frac{\delta_{in}}{D_{V,vac}}} \), \( \mu_{in} = \sqrt{\omega^2 n^2 + q_{s,in}^2} \).

Then the total rate of surface smoothing is:

\[
\frac{d z_n(t)}{dt} = -z_n(t) D_{s,vac} a \frac{c_0 \Omega}{kT} \omega \mu^2 n^4 - z_n(t) D_{V,vac} \frac{c_0 \Omega}{kT} \mu_{in} \left( \omega^2 n^2 + q_{s,vac}^2 \right) - D_{s,in} \frac{I_0 \Omega}{\nu} \left| \frac{\Omega_{in}}{kT} \right| z_n(t) \omega \mu^4 e^{-\ln q_{s,vac} - \mu_{vac} t} - D_{s,in} \frac{I_0 \Omega}{\nu} \left| \frac{\Omega_{in}}{kT} \right| z_n(t) \omega \mu^4 e^{-\ln q_{s,in} - \mu_{in} t} - I_0 \Omega \left\{ \mu_{in} \left( e^{-\ln q_{s,in} - \mu_{in} t} - \mu_{in} \right) \right\} - \mu_{vac} \left( e^{-\ln q_{s,vac} - \mu_{vac} t} \right)
\]

and after integration we find:

\[
\ln \frac{z_n(t)}{z_n(0)} = -D_{s,vac} a \frac{c_0 \Omega}{kT} \omega \mu^2 n^4 t - D_{V,vac} \frac{c_0 \Omega}{kT} \mu_{in} \left( \omega^2 n^2 + q_{s,vac}^2 \right) t - D_{s,in} \frac{I_0 \Omega}{\nu} \left| \frac{\Omega_{in}}{kT} \right| \omega \mu^4 e^{-\ln q_{s,vac} - \mu_{vac} t} - D_{s,in} \frac{I_0 \Omega}{\nu} \left| \frac{\Omega_{in}}{kT} \right| \omega \mu^4 e^{-\ln q_{s,in} - \mu_{in} t} - I_0 \Omega \left\{ \mu_{in} \left( e^{-\ln q_{s,in} - \mu_{in} t} - \mu_{in} \right) \right\} - \mu_{vac} \left( e^{-\ln q_{s,vac} - \mu_{vac} t} \right) t
\]

(17)

The first and the second terms in equation (17) describe the evolution of the surface without radiation. For \( q_{s,vac} = 0 = q_{s,in} \) (no internal sinks) these terms were received in [1]. The last three terms in (17) is the contribution of irradiation into \( \ln \frac{z_n(t)}{z_n(0)} \). For \( q_{s,vac} = 0 = q_{s,in} \) we rederive the result of article [3].
For \( q_{s, \text{vac}} \ll \omega n \) \((q_{s, \text{in}} \ll \omega n)\), \( q_{s, \text{vac}} \ll 1/l_{\text{vac}} \) \((q_{s, \text{in}} \ll 1/l_{\text{in}})\) and \( \omega l_{\text{vac}} \ll 1 \) \((\omega l_{\text{in}} \ll 1)\) we find from the equation (17) the corrections to the results of [1] and [3]:

\[
\ln \frac{z_n(t)}{z_n(0)} = -D_{S, \text{vac}} \frac{c_0 \gamma \Omega}{kT} \omega^4 n^4 t - D_{V, \text{vac}} \frac{c_0 \gamma \Omega}{kT} \omega^3 n^3 \left( 1 + \frac{q^2_{s, \text{vac}}}{2\omega^2 n^2} \right) t - \\
- D_{S, \text{vac}} \frac{l_0 \gamma |\Omega_{\text{vac}}|}{kT} \omega^4 n^4 (1 - l_{\text{vac}} q_{s, \text{vac}}) t - D_{S, \text{in}} \frac{l_0 \gamma |\Omega_{\text{in}}|}{kT} \omega^4 n^4 (1 - l_{\text{in}} q_{s, \text{in}}) t - \\
- l_0 \omega^2 n^2 \left\{ l_{\text{in}} \left( 1 - \frac{q_{s, \text{in}}}{\omega n} + \frac{q^2_{s, \text{in}}}{2\omega^2 n^2} \right) - l_{\text{vac}} \left( 1 - \frac{q_{s, \text{vac}}}{\omega n} + \frac{q^2_{s, \text{vac}}}{2\omega^2 n^2} \right) \right\} t
\]

(18)

We see that we have the second order correction \( \frac{q^2_{s, \text{vac}}}{\omega^2} \) to the result without sinks and without irradiation (the second term in equation (18) [1]) and the first order correction \( \frac{q_{s, \text{vac}}}{\omega} \), \( l_{\text{vac}} q_{s, \text{vac}} \) \((q_{s, \text{in}} / \omega, l_{\text{in}} q_{s, \text{in}})\) to the result with irradiation and without sinks [3].

The sign of the last term in eq. (17) depends on the sign of the expression:

\[
\psi = l_{\text{in}} \left( 1 - \frac{q_{s, \text{in}}}{\omega n} + \frac{q^2_{s, \text{in}}}{2\omega^2 n^2} \right) - l_{\text{vac}} \left( 1 - \frac{q_{s, \text{vac}}}{\omega n} + \frac{q^2_{s, \text{vac}}}{2\omega^2 n^2} \right)
\]

(19)

For \( \psi > 0 \) the last term is negative and it increases the rate of the surface smoothing and for \( \psi < 0 \) the last term increases the roughness of the surface. The leading term in the third term have \( \omega^2 \)-dependence on \( \omega \).

For \( q_{s, \text{vac}} \gg \omega \) \((q_{s, \text{in}} \gg \omega)\) the equation (17) takes the form:

\[
\ln \frac{z_n(t)}{z_n(0)} = -D_{S, \text{vac}} \frac{c_0 \gamma \Omega}{kT} \omega^4 n^4 t - D_{V, \text{vac}} \frac{c_0 \gamma \Omega}{kT} q_{s, \text{vac}} \omega^2 n^2 t - \\
- l_0 \omega^2 n^2 \left\{ l_{\text{in}} e^{-l_{\text{in}} q_{s, \text{in}}} - l_{\text{vac}} e^{-l_{\text{vac}} q_{s, \text{vac}}} \right\} t
\]

(20)

where the conditions \( l_{\text{vac}} \omega^2 \ll q_{s, \text{vac}} \) and \( l_{\text{vac}} \omega^2 \ll q_{s, \text{vac}} \) were assumed. In this case the second and the last terms in equation (19) have the same \( \omega^2 \) dependence on the frequency \( \omega \). The sign of the last term is determined by the expression:

\[
\psi_1 = l_{\text{in}} e^{-l_{\text{in}} q_{s, \text{in}}} - l_{\text{vac}} e^{-l_{\text{vac}} q_{s, \text{vac}}}
\]

(21)
Depending on the values of $q_{s,\text{in}}$ ($q_{s,\text{vac}}$) and $l_{\text{in}}$ ($l_{\text{vac}}$) the irradiation tends to decrease (for $\psi_1 > 0$) or to increase (for $\psi_1 < 0$) the roughness of the surface. The expression (15) shows that the interstitial atoms tends to make the surface become flat and the vacancies created by irradiation tends to increase the roughness of the surface. The influence of the point defects on the evolution of the surface is determined by the value of $le^{-lq}$. For example, if the interstitial atoms are absorbed by internal sinks stronger that the vacancies are then the vacancies are the main defects and the expression $\psi_1$ is negative - the irradiation increases the roughness of the surface.

In conclusion, we derived the expression for the rate of the surface smoothing taking into account the external sources of point defects (due to irradiation) and the internal sinks of point defects. We have found that the irradiation has tow contribution into the rate of surface smoothing: the first contribution (the third and the fourth terms in equation (17)) increase the rate of the surface flattening and the influence of the second contribution of irradiation (the last term in equation (17)) on the evolution of the surface structure depends on the sign of the expression $\psi = l_{\text{in}} \left(1 - \frac{q_{s,\text{in}}}{\omega n} + \frac{q_{s,\text{in}}^2}{2\omega^2 n^2}\right) - l_{\text{vac}} \left(1 - \frac{q_{s,\text{vac}}}{\omega n} + \frac{q_{s,\text{vac}}^2}{2\omega^2 n^2}\right)$ (for $q_{s,\text{vac}} << \omega$, $q_{s,\text{in}} << \omega$ and $q_{s,\text{vac}}l_{\text{vac}} << 1$, $q_{s,\text{in}}l_{\text{in}} << 1$) and the expression $\psi_1 = l_{\text{in}}e^{-l_{\text{in}}q_{s,\text{in}}} - l_{\text{vac}}e^{-l_{\text{vac}}q_{s,\text{vac}}}$ (for $q_{s,\text{vac}} >> \omega$, $q_{s,\text{in}} >> \omega$). For $\psi > 0$ ($\psi_1 > 0$) the second contribution of irradiation increases the rate of the surface flattening and for $\psi < 0$ ($\psi_1 < 0$) the it increases the roughness of the surface.

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