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Project No.：2076

Radiation-induced motion of liquid inclusions in crystals

［１］Organization

Project Leader：
Oksana Podshyvalova, Ph.D., Prof. of "KhAI" (National Aerospace University "Kharkiv Aviation Institute", Ukraine)
Representative at RIE：
Toru Aoki, Ph.D., Prof. (Research Institute of Electronics, Shizuoka University, Japan)
Participants：
Ivan Marchenko, D.Sc. (Nat. Sci. Center "Kharkov Institute for Physics & Technology" of the National Academy of Sciences of Ukraine, Ukraine)
Oleksandr Kulyk, Ph.D., Assoc. Prof. (V. N. Karazin Kharkiv National University, Ukraine)
Volodymyr Gnatyuk, Ph.D., Senior Scientist (V. E. Lashkaryov Institute of Semiconductor Physics of the National Academy of Sciences of Ukraine, Ukraine)

［２］Research Progress

The project objective is to investigate the motion of liquid inclusions in alkali halide crystals irradiated by γ-quanta and electrons. Based on the results of experimental studies of radiation-induced motion and transformation of the shape of inclusions as well as the phenomenon of induced transitions of matrix atoms into solution and back [1-5], the physical model of the liquid inclusion motion is developed. Understanding of the mechanism of spontaneous inclusions migration is important for monitoring the integrity of radioactive waste disposal in geological halide sediments.

The participants of the project actively collaborated through electronic media and face-to-face when the Ukrainian partners *O. Podshyvalova* and *O. Kulyk* visited RIE, Hamamatsu in January 2020 (research visits in the frame of the 2019 Cooperative Research at Research Center of Biomedical Engineering). They reported the achieved results, exchanged ideas on join work in frames of the cooperative project, and discussed further cooperation between the partner institutions.

Apart from achieving the research results, described in the next section, the project participants were involved in the holding of the international scientific forums to discuss the research work concerning the project topics and general cooperation issues.

Dr. O. Podshyvalova submitted the abstract with the key research findings to the Book of Abstracts of The Annual Meeting of 2019 Cooperative Research on Biomedical Engineering (Yokohama, Japan, 13 Mar. 2020) and Dr. O. Kulyk presented the project-related results at The 18th International Conference on Global Research and Education, Inter-Academia 2019 (iA-2019) in Budapest, Hungary (4-7 Sep. 2019).

Totally, 5 publications (2 articles & 3 abstracts) related to the project results have been published during the project implementation [2-5, 7].

［３］Results

（３－１）Research results

At gamma and electron irradiation of dielectric single crystals structural defects and, first of all, point defects, such as color centers, are formed in them [1]. The distribution of radiation-induced defects in a crystal depends both on its prehistory and on exposure conditions, and is very inhomogeneous. The "matrix - color centers" system is thermodynamically non-equilibrium and eventually relaxes into the state of minimum free energy. For example, in the presence of liquid inclusions in a water-soluble crystal, their spontaneous motion is one of relaxation mechanisms of the system. Such inclusion motion occurs if the characteristic size of the inhomogeneity of radiation defects significantly exceeds the inclusion size and if the crystal contains a sufficient density of screw dislocations, which are sources of steps on the dissolving inclusion surfaces or evaporating pore surfaces [2-4]. As it was experimentally determined, in a crystal with inhomogeneous distribution of radiation defects, macroscopic inclusions moved as a single whole with almost unchanged shape (Fig. 1).

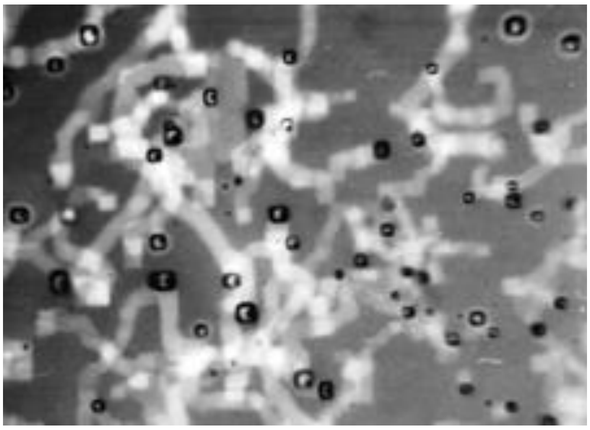

Fig. 1. Traces of inclusions spontaneous migration in an electron-irradiated crystal a month after irradiation.

Well-known phenomenological theories that are used to analyze the motion of inclusions in a crystal describe the matrix-inclusion interaction based on the laws of dissolution, diffusion and crystallization flows conservation. Based on the phenomenon of induced transitions of matrix atoms into the solution and back [5], we have developed a physical model that describes the time dependences of the main parameters of liquid inclusions and allows us to predict the dynamics of their motion.

1. In the reference frame, where a spherical azimuthally symmetric inclusion in a crystalline matrix with inhomogeneous distribution of point defects is at rest when the inclusion size does not exceed the critical size: (or moves in the negative direction of the axis θ $(\theta = \pi)$ at $R > R_0$) (Fig. 2), the matrix velocity is represented as:

$$\vec{U}(R_0) = \left(U_r(R_0); U_\theta(R_0)\right), \qquad (1)$$

where

$$U_r(R_0) = U_{r0} = \vec{U}(R_0)\vec{e}_r, U_\theta(R_0) = U_{\theta 0} = \vec{U}(R_0)\vec{e}_\theta$$

$\vec{e}_r, \vec{e}_\theta$ - are the unit vectors on the radial $\vec{r}$ and polar θ axis, respectively.

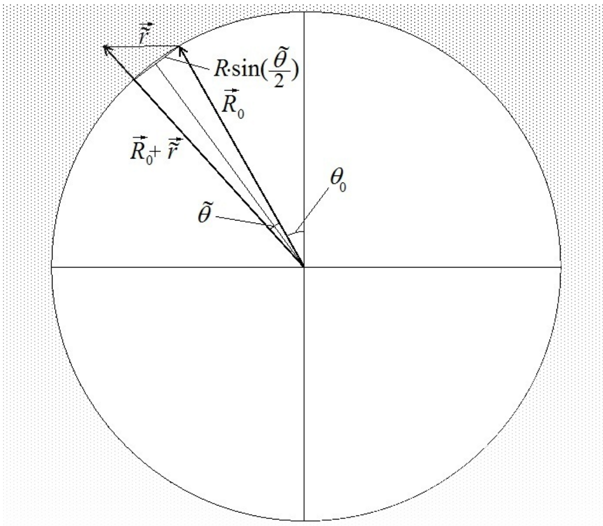

Fig. 2 Layout of a spherical azimuthally symmetric inclusion in the crystal matrix (the inhomogeneous distribution of radiation defects is shown by dots).

2. The elementary processes of transitions of matrix atoms into the solution and back on the frontal and back surfaces of the inclusion are considered on the example of a two-level system [5]. It is obtained the system of equations for the number of matrix atoms in solution n_1, on the front wall n_1, and solvent atoms in the inclusion N:

$$\frac{\partial n_1}{\partial t} + U_{r0}\frac{\partial n_1}{\partial \tilde{r}} + \frac{U_{\theta 0}}{R_0 + \tilde{r}}\frac{\partial n_1}{\partial \tilde{\theta}} = \mu_{12}(n_2 - n_1)N, \qquad (2)$$

$$\frac{\partial n_2}{\partial t} + U_{r0}\frac{\partial n_2}{\partial \tilde{r}} + \frac{U_{\theta 0}}{R_0 + \tilde{r}}\frac{\partial n_2}{\partial \tilde{\theta}} = -\mu_{12}(n_2 - n_1)N,$$

$$\frac{\partial N}{\partial t} + U_{r0}\frac{\partial N}{\partial \tilde{r}} + \frac{U_{\theta 0}}{R_0 + \tilde{r}}\frac{\partial N}{\partial \tilde{\theta}} = -\mu_{12}(n_2 - n_1)N.$$

3. To find solutions of equations (2) the probability of induced transitions μ_{12} is represented in the form of a Taylor series expansion of the small displacements along the radius $\tilde{r}$ and the polar angle $\tilde{\theta}$:

$$\mu_{12}(R_0 + r, \theta_0 + \theta) \approx$$
$$\approx \mu_{12}(R_0, \theta_0)\left(1 + \sum_{n=1}^{2}\lambda_{r,2n-1}r^{2n-1} + \theta\lambda_\theta\right), \qquad (3)$$

where

$$\lambda_{r,2n-1} = \frac{1}{\mu_{12}(R_0, \theta_0)}\frac{1}{(2n-1)!}\frac{\partial^{2n-1}\mu_{12}(R_0, \theta_0)}{\partial r^{2n-1}},$$

$$\lambda_\theta = \frac{1}{\mu_{12}(R_0, \theta_0)}\frac{\partial \mu_{12}(R_0, \theta_0)}{\partial \theta}.$$

4. Based on the type of expansion chosen in [3], by introducing in (3), the introduction of a new variable

$$\xi(t,r,\theta)=t+\frac{r^2\lambda_{r,1}}{2U_{r0}}+\frac{r^4\lambda_{r,3}}{4U_{r0}}+\frac{R_0\theta^2\lambda_\theta}{2U_{\theta 0}},$$

equations (2) can be converted to a simple form:

$$\frac{\partial n_1}{\partial\xi}=\mu(n_2-n_1)N,$$
$$\frac{\partial n_2}{\partial\xi}=-\mu(n_2-n_1)N, \qquad (4)$$
$$\frac{\partial N}{\partial\xi}=-\mu(n_2-n_1)N.$$

5. By setting the constant phase plane:

$$\xi(t,r,\theta)=D, \qquad (5)$$

where D is a constant, and passing into the reference system where the inclusion moves, the dependence of the inclusion velocity on its radius has the form:

$$U_R=\frac{Ax_R}{B+x_R^2+Cx_R^4}, \quad x_R=\frac{R}{R_0}. \qquad (6)$$

6. Based on the energy interpretation of the inclusion velocity [5], the parameters in (6) are interpreted and their numerical values are determined using the experimental data presented in [6]. A comparison of theoretical calculations with experimental data indicates an adequate description of the experimental results by the proposed theoretical model (Fig. 3) [7].

(３－２) Ripple effects and further developments

The proposed model of induced inclusion motion in a crystal with inhomogeneous distribution of radiation defects adequately describes the non-monotonic dependence of inclusion velocity on its size. The physical nature of non-monotonity is due to the confrontation of two forces: the driving force, determined by a gradient of radiation defects and the dissipative force, determined by the effective characteristics of the viscous properties of the matrix material. The validity of the presented physical model of inclusions' induced motion is confirmed for crystals with dislocation and radiation defects and can be applied to crystals with inhomogeneous distribution of defects of another nature.

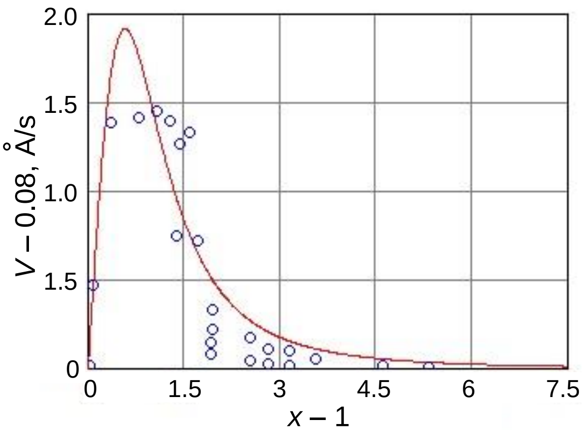

Fig. 3. Dependence of inclusions velocity *V* on their dimensionless longitudinal size *x*. Circles are experimental points, line is the calculation result on the formula $V - 0.08 = 5.9(x-1)/((x-1)^2+1)^2$ [7].

［4］ Achievements (List of Publications)

(1) A.P. Kulik, O.V. Podshyvalova, I.G.Marchenko, Radiation-induced motion of liquid inclusions in alkali halide crystals, *Problems of Atomic Science and Technology* 120(2) 13-19 (2019).

(2) O.P. Kulyk, V.I. Tkachenko, O.Yu. Lisina, V.O. Mikhnych, V.A. Gnatyuk, T. Aoki, Nonlinear effects of diffusion interaction of steps on thermodynamically stable vicinal surfaces, *Physical and Technical Problems of Energy and Their Solutions 2019, Proceedings of the International Scientific and Technical Conference*, P. 13, 2019 (Kharkiv, Ukraine, 19 June 2019). (*Plenary report*)

(3) O.P. Kulyk, L.A. Bulavin, S.F. Skoromnaya, V.I. Tkachenko, Model of induced motion of inclusions in the field of forces of an inhomogeneously stressed crystal, *The 18th International Conference on Global Research and Education in Engineering for Sustainable Future, Inter-Academia 2019, Program and Book of Abstracts*, P. 9-10, 2019. (4-7 September 2019, Budapest and Balatonfüred, Hungary).

(4) O.P. Kulyk, V.I. Tkachenko, O.V. Podshyvalova, V.A. Gnatyuk, T. Aoki, Nonlinear interaction of macrosteps on vicinal surfaces at crystal growth from vapour, *Journal of Crystal Growth*, Vol. 530, P. 125296-1-7, Jan. 2020. *DOI: 10.1016/j.jcrysgro.2019.125296*

(5) O.P. Kulyk, L.A. Bulavin, S.F. Skoromnaya, V.I. Tkachenko, Model of induced motion of inclusions in the field of forces of an

inhomogeneously stressed crystal, *in: A.R. Varkonyi-Koczy (ed.) Engineering for Sustainable Future. Inter-Academia 2019. Lecture Notes in Networks and Systems (LNNS)*, Vol. 101, P. 326-3397, 2020, Cham: Springer. *DOI: 10.1007/978-3-030-36841-8_32*

(6) V.S. Kruzhanov, O.V. Podshyvalova, Motion of liquid inclusions in the crystal caused by radiation defects. Sov. Solid State Physics 32(2), 373-378 (1990).

(7) O. Podshyvalova, V. Tkachenko, O. Kulyk, V. Gnatyuk, T. Aoki, Radiation-induced motion of liquid inclusions in crystals, *The Annual Meeting of 2019 Cooperative Research on Biomedical Engineering, Book of Abstracts,* Abstract No 1-10, 2019, (13 March 2020, Tokyo, Japan), *in press.*

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Travelling Report（Mention each travel by CRP budget.）

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Affiliation : National Aerospace University "Kharkiv Aviation Institute", Chkalov Str. 17, Kharkiv 61070, Ukraine

Period of time：04 January – 20 January 2020

Destination：Research Institute of Electronics, Shizuoka University

Purpose： (1) Study of the processes leading to the motion of liquid inclusions in irradiated crystals with layer-by-layer growth/dissolution. (2) Participation in the scientific meetings at Research Institute of Electronics, Shizuoka University during the period of stay. (3) Discussion of the research results, preparation of the publications and development of the plans of the future research.

Name of receiver：Prof. Toru Aoki

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