

Self-Organization of Radiation Defects: Temporal Dissipative Structures

Pavlo Selyshchev

Abstract—A theoretical approach to radiation damage evolution is developed. Stable temporal behavior taking place in solids under irradiation are examined as phenomena of self-organization in non-equilibrium systems.

Experimental effects of temporal self-organization in solids under irradiation are reviewed. Their essential common properties and features are highlighted and analyzed.

Dynamical model to describe development of self-oscillation of density of point defects under stationary irradiation is proposed. The emphasis is the nonlinear couplings between rate of annealing and density of defects that determine the kind and parameters of an arising self-oscillation.

The field of parameters (defect generation rate and environment temperature) at which self-oscillations develop is found. Bifurcation curve and self-oscillation period near it is obtained.

Keywords—Irradiation, Point Defects, Solids, Temporal Self-organization.

I. INTRODUCTION

THE phenomena which occur in solids under irradiation are characterized by a number of common features. In the first, all of them are non-equilibrium. Material under irradiation is a typical example of an open system of streaming type. Fluxes of energy, matter and entropy maintain states which are far away from thermal equilibrium. Secondly - it is complicated complex phenomenon caused by multiple, interconnected processes. It is often not possible to indicate several the most dominant among them (the phenomenon of synergism). Third, the connections arising under irradiation, as a rule, are inherently nonlinear. They are described by nonlinear equations in framework of nonlinear dynamics. Nonlinear couplings form the mechanisms of feed-backs that drive evolution of radiation damage. Finally appearance of dissipative structure has a threshold character.

A necessary condition for the development of the structure of radiation-induced defects is irradiation. However, the parameters of the structure are determined, first of all, by the internal connections between its elements. The structure is stable with respect to changes in radiation conditions in a wide enough range. Therefore the evolution of the structure of radiation damage can be considered as self-organization. In this case any stable behavior and arising structures are called

dissipative structures in contrast to the equilibrium ones. There are temporal and spatial dissipative structures.

It is known a lot of experimentally observed phenomena which correspond to temporal self-organization of defect density. Well known examples of temporal self-organization are gallops and oscillations of conductivity, non-monotonous dose dependence of microhardness, non-monotonous dose dependence of creep, oscillations of size of vacancy voids, temperature self-oscillation.

II. BACKGROUND

There are the following experimental facts of structured behavior of physical properties and parameters of the microstructure of irradiated materials in time (for stationary condition of irradiation).

Jumps and oscillations of conductivity which is indicator of defect density were observed in metals, semiconductors and ceramic materials [1]-[4].

Regular electrical oscillations with a period is approximately equal to 100 hours and an amplitude is equal to 30% of the mean value was observed during irradiation of aluminum and sapphire samples with 1keV neutron flux ($2 \cdot 10^{16} - 7.2 \cdot 10^{16} \text{ m}^{-2} \text{ s}^{-1}$) at 615°C [1].

The oscillations of resistivity with a frequency of about 10^{-5} s^{-1} were observed during irradiation of a copper sample with 2.2MeV electrons (22.4 mA m^{-2}) at a temperature of 103K [2].

Also periodic variation of the electrical resistivity during irradiation gold specimen by 2.3MeV electrons (11.5 mA m^{-2}) at a temperature 197K is given in [2] too. The oscillations with a frequency of about $2 \cdot 10^{-4} \text{ s}^{-1}$ are modulated by sinusoidal wave with frequency of about 10^{-5} s^{-1} .

Periodic variation of microhardness of Nimonic 90 containing γ -precipitates as a function of dose was observed in [5] during irradiation with 50keV helium ions at temperature near 300K and defect generation rate of the order of 10^{-2} dpa/s .

Not monotone dependence of the relative change of microhardness of 79 Permalloy on dose was observed in [6].

It is well known the cases of non-monotonic dose dependence of the radiation creep [7], [8]. Creep of loaded steel samples during irradiation by neutrons ($2 \cdot 10^{18} \text{ m}^{-2} \text{ s}^{-1}$) at 350°C and defect generation rate $1.7 \cdot 10^{-7} \text{ dpa/c}$ looks like damped oscillations with a period of about 10^7 s .

Variations of temperature of organic crystals CH_4 at the reactor irradiation were observed for the environment temperature below 120K [9].

P. A. Selyshchev is with the Department of Physics University of Pretoria Private bag X20 Hatfield 0028 South Africa (phone: 012-420-6641; fax: 012-362-5288; e-mail: pavel.selyshchev@up.ac.za).

This work was made possible by support from NRF of South Africa.

Finally, oscillations of the void size were observed in nickel samples irradiated with 180 keV nickel ions at 750°C and $5 \cdot 10^{-2}$ dpa/s. Period of the oscillations was of about 10^3 s. [10].

All these phenomena can be sufficiently described in framework of nonlinear dynamics of complex system using power methods of theory of self-organization and complex system analysis [11]. Features of the development of self-oscillations for different dynamic systems depend on the nonlinear feedbacks of the systems for example these feedbacks can be elastic or electrostatic interaction between defects [11], interaction by means of flux of point defect [12] or thermo-concentration interconnection between defect density and temperature of sample that will be consider below. Period of self-oscillation is determined by characteristic times of process that go on inside system.

III. SELF-OSCILLATION OF RADIATION-INDUCED POINT DEFECTS IN SOLIDS UNDER IRRADIATION

As an example of temporal self-organization, let us consider development of temporal self-oscillations of radiation-induced point defects in solids under irradiation. Self-oscillations of defects occur due to the development instability that arises in the system of non-equilibrium excitations with finite life-time for example radiation induced point defects. The mechanism of the instability is following. Consider a crystalline sample under stationary conditions of irradiation. The irradiated sample is heated due to the relaxation of various radiation induced excitations. A big part of the energy of irradiation transforms into heat. Other small part of the irradiation energy (of the order of percent or less depending on the type of irradiation) expends to form of crystal defects: vacancies, interstitials, etc. The defects have finite life-time because they are absorbed by sinks (dislocations, grain boundaries, surface) and recombine. Defects move due to diffusion which grows with temperature. As a rule the dependence is non-linear. Since defects and their sinks are spatially separated, defects can be accumulated in the crystal. The accumulated energy may reach significant amount. During defect annealing (recombination, absorption by sinks) the accumulated energy is converted into heat.

Let us assume that a fluctuation of crystal temperature has increased as result of fluctuation. It leads to increase in diffusion of the defects. The annealing of the defects increases. Release of heat grows too. And temperature increases further. Chain of feedback is closed. Growth of the temperature fluctuation is limited by decrease in the defect density due to annealing. After that temperature of the crystal drops due to thermal contact with the environment. But owing irradiation the defect density increases. When it reaches the certain value, the process is repeated. Self-oscillations of the temperature and defect density are developed. Thus, the steady state that is a priori expected is not unique.

IV. MODEL AND RESULTS

Let the sample has the shape of a plane-parallel plate. The plate thickness is l . Environment temperature is kept constant and equal to T_e . Due to irradiation sample heats with rate Q and defects are created with rate K . The defects are absorbed by dislocations. The dislocation density is ρ_d . When defect is absorbed, some energy releases. It is approximately equal to energy of the defect formation θ .

If the plate is so thin that $h'l/4\chi \ll 1$, temperature of the plate and defect density are approximately constants. Here χ is thermal conductivity of the sample. Absorption of defects by the plate surface compared to their absorption by the internal sinks is neglected. Then the average defect density n and average plate temperature T are described by the system of equations.

$$\frac{dn}{dt} = K - \beta(T)n \quad (1)$$

$$\frac{dT}{dt} = \frac{1}{c}(Q + \theta\beta(T)n - h(T - T_e)) \quad (2)$$

Here $\beta(T) = \rho_d D(T)$ is inverse lifetime of defects, $D(T) = D_0 \exp(-E_m/T)$ is diffusion coefficient defects, E_m is migration energy of the defect. $h=2h'/l$, where h' is heat transfer coefficient between sample and environment.

The heating rate is proportional to the intensity of the irradiation, and therefore it is proportional to rate of defect generation: $Q = \xi \theta K$. Parameter ξ is ratio of energy of irradiation which transforms into heating and energy of irradiation which transforms into defect generation.

The system (1)-(2) is nonlinear as result of the exponential dependence of β on the temperature.

There is the only one possible stationary solution of (1)-(2), that describes the stationary homogeneous density of defects under irradiation,

$$n_s = K / \beta(T_s) \quad (3)$$

where $T_s = T_e + \theta(\xi + 1)K / h$.

The stationary solution (3) takes place physically if it is stable. To exam stability let us consider the evolution of its small perturbations δn and δT . Damping decrement of the small perturbations satisfies equation

$$\lambda^2 + p\lambda + q = 0 \quad (4)$$

where

$$p = h/c - K\theta E_m / cT_s^2 + \beta(T_s) \quad (5)$$

$$q = h\beta(T_s)/c \quad (6)$$

The value of q is positive for all physically admissible

values. The value of p has variable sign. If $K \rightarrow \infty$ and $T_e \rightarrow \infty$, then $p > 0$ and therefore $\text{Re}\lambda < 0$. So the stationary distribution is stable. With decreasing values of K and T_e condition $p > 0$ can be broken.

So far as for any parameters there is a loop without contact which covers the stationary point (3) and all phase trajectories of the system (1)-(2) go in the loop, there is a limit cycle of the system (1)-(2). So self-oscillations of temperature and density of defects are developed.

The self-oscillations are developed if inequality

$$K_s \geq T_s^2 (h + c\beta(T_s)) / \theta E_m \quad (7)$$

is satisfied.

Let all parameters are constants except environmental temperature (T_e) and defect production rate (K). Then space of these parameters can be divided into two fields. For parameters from the first field the stationary homogeneous distribution of defects takes place under irradiation. For parameters from the second field the self-oscillation of density of defects develops. Parametric equations for bifurcation curve are following

$$K = T^2 (h + c\beta(T)) / \theta E_m \quad (8)$$

$$T_e = T - T^2 (\xi + 1) (h + c\beta(T)) / h E_m \quad (9)$$

where sample temperature T is parameter.

The bifurcation value of environment temperature is limited from above, since the second term in (9) for large values of T begins to dominate.

Frequency oscillations near the bifurcation curve is

$$\omega = \sqrt{\frac{h\beta}{c}} \quad (10)$$

Thus, the period of oscillation is the square root of the product of the lifetime of the defects and the characteristic time of sample cooling.

Computer simulation of self-oscillation carries out for models with the parameters of lead and aluminum. It shows the following. Stability diagram for aluminum and lead are similar. For example, the highest environment temperature at which self-oscillation develops in a lead sample is equal to 173K, at defect generation rate $1.3 \cdot 10^{-3}$ dpa/s. Temperature of the sample for these parameters is equal to 273K. The frequency of oscillation is 0.12s^{-1} . When heat-sink decreases, self-oscillation frequency decreases too. The frequency of oscillation depends on the pre-exponential factor of the diffusion coefficient, energy of defect migration and energy of defect formation. If heat capacity of irradiated sample is reduced, frequency of self-oscillation decreases.

In materials with higher density sinks the region of instability is less and the frequencies of the self-oscillation are

higher.

If ratio ξ increases, the region of instability expands and the frequency of the self-oscillation increases.

It is shown that during development of the self-oscillation at low environment temperature the defect density can exceed the steady-state value in ten times. Temperature of the sample can exceed the steady-state values at 100-150K.

At low environment temperature the amplitude of self-oscillations of the defect density near bifurcation curve is about 50% of the average. The amplitude of the self-oscillations of the temperature is about 0.3% of the average.

V. CONCLUSION

Under certain stationary conditions of irradiation self-oscillations of defect density and temperature of irradiated sample are developed. Defect density and temperature during self-oscillations can reach much higher values than the expected steady-state values. Therefore it is necessary to take into account possibility of the self-oscillation for secure operation of nuclear facilities and for environmental protection.

REFERENCES

- [1] Farnum E.H., Clinard F.W., Jr., Sommer W.F., Kennedy III J.C. and Shikama T. "Search for radiation - induced electrical degradation in alumina during spallation - neutron irradiation", *J. Nucl. Mater.*, vol.212-215, pp.1128-1132, 1994.
- [2] Schule W. "Radiation-enhanced diffusion due to interstitials and dynamic crowdions", *J. Nucl. Mater.*, vol.233-237, pp.964-968, 1996.
- [3] Sen P., Aggarwal G., Tiwari U. "Dissipative structure formation in cold-rolled Fe and Ni during heavy ion irradiation", *Phys. Rev. Letters*, vol.80, pp.1128-1132, 1998.
- [4] Kinoshita C., Zinkle S. J. "Potential and limitations of ceramics in terms of structural and electrical integrity in fusion environments", *J. Nucl. Mater.*, vol.233-237, pp. 100-110, 1996.
- [5] Varatharajan K. and Nandedkar R.V. "Microhardness-microstructure study of aged nimonic 90 irradiated with helium" in *Effects of Radiation on Materials*. R.E. Stoller Edr. - Philadelphia, 1989, pp.263-270
- [6] Tetelbaum D.I., Kurilchik E.V., Latisheva N.D. "Long-range effect at low-dose ion and electron irradiation of metals", *Nucl. Inst. and Meth. in Phys. Research B*, vol.127-128, pp. 153-156, 1997.
- [7] Toloczko M.B., Garner F.A. and Eiholzer C.R. "Irradiation creep and swelling of the US fusion heats of HT9 and 9Cr-1Mo to 208 dpa at $\sim 400^\circ\text{C}$ ", *J. Nucl. Mater.*, vol.212-215, pp.604-607, 1994.
- [8] Schule W. and Hausen H. "Neutron irradiation creep in stainless steel alloys", *J. Nucl. Mater.*, vol.212-215, pp. 388 - 392, 1994.
- [9] Carpenter J.M. "Thermally activated release of stored chemical energy in cryogenic media", *Nature*, vol.36, pp. 358-360, 1987.
- [10] Steele J.K. and Potter D.I. "The disappearance of voids during 180 keV Ni⁺ bombardment of nickel", *J. Nucl. Mater.*, vol.218, pp. 95-107, 1995.
- [11] Selyshchev P.A. *Self-organization phenomena in nuclear materials*, Moscow Izhevsk, NITs "Regulyarnaya i haoticheskaya dinamika" 2008, 208 p.
- [12] Selyshchev P.A. Self-organization of microstructure as a new way of using materials under irradiation, Word Academy of Science, Engineering and Technology An International Journal of Science, Engineering and Technology. Cemal ARDIL, EiC, Bali. Issue 58,

Pavlo A. Selyshchev was born in Livny, Orel region, Russia in the 15th of October, 1958. In 1981 he graduated Physics Department of Kyiv National University and has been working in there. He had been a professor of Physics Department of Kyiv National University by 2002, from 2011 he is a professor of University of Pretoria. In 1991 he received the Cand. Sci. (Phys. & Math.) degree in theoretical physics due to thesis "Interaction between defects and formation of dissipative structures in impure crystals under the irradiation"

performed in Kiev Institute for Nuclear Research. In 2001 he received the doctor's degree in theoretical physics due to thesis "The nonlinear couplings and external fluctuations in self-organization of radiation defect structures". He is specialized in theoretical physics for complex non-linear systems and their evolution. His research interests lay in the area of kinetic non-linear processes in materials under irradiation. A theoretical approach to the self-organization phenomena in irradiated materials is developed by him. He has about 200 scientific publications and several monographs.