Radiation Damage as Nonlinear Evolution of Complex System

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***Abstract*—**Irradiated material is a typical example of a complex system with nonlinear coupling between its elements. During irradiation the radiation damage is developed and this development has bifurcations and qualitatively different kinds of behavior.

The accumulation of primary defects in irradiated crystals is considered in frame work of nonlinear evolution of complex system. The thermo-concentration nonlinear feedback is carried out as a mechanism of self-oscillation development.

It is shown that there are two ways of the defect density evolution under stationary irradiation. The first is the accumulation of defects; defect density monotonically grows and tends to its stationary state for some system parameters. Another way that takes place for opportune parameters is the development of self-oscillations of the defect density.

The stationary state, its stability and type are found. The bifurcation values of parameters (environment temperature, defect generation rate, etc.) are obtained. The frequency of the self-oscillation and the conditions of their development is found and rated. It is shown that defect density, heat fluxes and temperature during self-oscillations can reach much higher values than the expected steady-state values. It can lead to a change of typical operation and an accident, e.g. for nuclear equipment.

***Keywords*—**Irradiation, Primary Defects, Solids, Self-oscillation.

I. Introduction

IRRADIATED material is a typical example of a complex system. Firstly, the influence of irradiation has a complex synergistic character. A lot of phenomena take place in material under irradiation. The ion and electron subsystems are excited. The material is heated. Different damages of crystal lattice which drive the microstructure and macroscopic properties changes are created. Secondly, structure, composition, and properties are altered over an extremely wide scale, spanning microscopic processes, meso-scale microstructures and macroscopic properties. All these phenomena are inherently connected with essential nonlinear coupling, it is often impossible to indicate the most dominant of them [1].

One of the most important consequences of radiation influence is the creation of primary radiation defects, namely: interstitial atoms, vacancies and their small clusters, small vacancy and interstitial loops. During irradiation the structure of the radiation damage becomes more and more complex. It is developed and its development is nonlinear. The radiation damage evolution is driven with nonlinear feed-backs and has qualitatively different ways and bifurcations. The system may depend crucially on the numerical values of certain parameters, namely the condition of irradiation and material properties. For instance, the number of stationary states of the system or quality character of its behavior may change abruptly as value of a parameter is changed.

It is usually expected that the stationary state is realized under stationary external conditions. But this statement is not always true for open complex systems with nonlinear feed-backs. Under certain opportune conditions the steady state can become unstable with respect to the development of non-stationary states, e.g. self-oscillations [2].

Let us consider an irradiated sample under stationary conditions of irradiation. The crystal defects (vacancies, interstitial atoms, etc.) are created and accumulated in the sample as a result of irradiation. The significant energy that is equal to the energy of defect formation is accumulated in the sample too. During defect annealing (recombination, absorption by sinks) the accumulated energy is converted into heat. The irradiated sample is also heated due to the relaxation of various radiation induced excitations. As a rule a big part of the energy of irradiation transforms into heat. Another small part of the energy of irradiation (about several percent) expends to form the radiation defects. The environment temperature is fixed. The rate of defect generation and heating are constant.

A mechanism of instability and the development of the self-oscillations is the thermo-concentration nonlinear feedback. Let a small increase of the defect annealing arise as a result of small fluctuation. When defect annealing increases, the energy that is stored by radiation defects is released into heat and the temperature of the sample increases too. As a result, the diffusion of the defects grows, and annealing increases further. The positive feed-back is formed. The temperature grows quickly and the concentration of defects drops, thus defect annealing and the release of energy drops too. The sample cools and the radiation defects are accumulated slowly. After that all processes are repeated. Self-oscillations of the defect density and the sample temperature are developed. Thus there are two ways of the defect density change under stationary irradiation. The first is the accumulation of defects; defect density monotonically grows and aims for the some constant value. The second is the development of self-oscillations of the defect density.

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A theoretical research of the self-oscillations is developed via Poincare formalism within the framework of dynamics of complex system on the plane.

II. Model And Basic Equations

Let us consider a crystal that has a shape of a plane-parallel plate. The plate thickness is l. The environment temperature is kept constant and equal to T_e. Due to irradiation the defects are created in the crystal with a rate of K and the crystal heats with a rate of Q. The defects recombine and are absorbed by dislocations. When a defect is absorbed, some energy releases. It is approximately equal to the energy of the defect formation.

The absorption of interstitial atoms is much more than vacancies since the diffusion of the interstitial atoms is much quicker than the one of the vacancies. Thus the concentration of the interstitial atoms is much more than the vacancy concentration. It allows us to neglect recombination and take into account the vacancies only.

The change of the vacancy density $n(x,t)$ and the temperature of the crystal $T(x,t)$ are described by equations

$$\frac{\partial n}{\partial t} = D\frac{\partial^2 n}{\partial x^2} + K - \beta(T)n \qquad (0 \le x \le l/2) \tag{1}$$

$$c\frac{\partial T}{\partial t} = -\frac{\partial J}{\partial x} + Q + \theta\beta(T)n \qquad (0 \le x \le l/2) \tag{2}$$

The boundary conditions are

$$J\big|_{x=0} = 0\,,\; J\big|_{x=l/2} = -h'\left(T\big|_{x=l/2} - T_e\right).$$

$J = -\kappa\nabla T$ is the heat flux, where κ is thermal conductivity of the crystal. Value h' is the heat transfer coefficient between the crystal and the environment, c is the crystal heat capacities per unit of mass. $\beta(T) = \rho_d D(T)$ is the inverse lifetime of defects with respect to absorption by dislocations, and the dislocation density is ρ_d. The equation $D(T) = D_0\exp(-E_m/T)$ is the diffusion coefficient of defects; E_m is the energy of their migration. The parameter θ is the energy of the defect formation.

We use symmetry and take into account that the flux of the defects on boundary is equal to zero because the absorption of defects by the plate surface compared to their absorption by the internal sinks is neglected. The nonlinear terms (the third in right side of (1) and (2)) connect these equations and describe the nonlinear feedback between the defect density, the rate of annealing and the temperature.

If the internal plate is so thin that $h'l/4\kappa << 1$, the temperature and the defect density are approximately constants. Then the average defect density n and average plate temperature T are described by the system of equations.

$$\frac{dn}{dt} = K - \beta(T)n \tag{3}$$

$$\frac{dT}{dt} = \frac{1}{c}(Q + \theta\beta(T)n - h(T - T_e)) \tag{4}$$

Here $h=2h'/l$. The heating rate is proportional to the intensity of the irradiation, therefore it is proportional to the rate of the defect generation: $Q = \xi\theta K$. The parameter ξ is the ratio of the energy of irradiation which transforms into heating and the energy of irradiation which transforms into defect generation.

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

III. Stationary State and its Stability

There is only one possible stationary solution of (3) - (4) (critical point) that describes the stationary homogeneous temperature and the density of defects under irradiation,

$$n_s = K/\beta(T_s) \tag{5}$$

$$T_s = T_e + \theta(\xi + 1)K/h \tag{6}$$

The stationary solution (3) – (4) is realized if it is stable. To exam the stability let us consider the evolution of its small perturbations δn and δT. The damping decrement of the small perturbations satisfies the equation

$$\lambda = -p \pm \sqrt{p^2 - q} \tag{7}$$

Where

$$p = \left(h/c - K\theta E_m/cT_s^2 + \beta(T_s)\right)/2 \tag{8}$$

$$q = h\beta(T_s)/c \tag{9}$$

The value of q is positive for all physically admissible values. The value of p has a variable sign. If $K \to \infty$ and $T_e \to \infty$, then $p>0$ and therefore $\mathrm{Re}\lambda<0$. So the stationary distribution is stable. With decreasing values of K and T_e the condition of $p>0$ can be broken, and the stationary distribution becomes unstable.

The stationary distribution becomes unstable if inequality

$$\theta E_m K \ge \left(h + c\beta(T_s)\right)\left(T_e + \theta(\xi + 1)K/h\right)^2 \tag{10}$$

is satisfied.

Let all parameters be constants except the environment temperature (T_e) and the defect production rate (K). The space of these parameters can be divided into two fields. For the parameters from the first field the stationary homogeneous distribution of defects is stable and it takes place under irradiation. For the parameters from the second field it unstable and is not realized. Parametric equations for bifurcation curve are the following:

$$K = T^2\left(h + c\beta(T)\right)\big/\theta E_m \tag{11}$$

$$T_e = T - T^2(\xi+1)\left(h + c\beta(T)\right)\big/hE_m \tag{12}$$

where sample temperature T is parameter.

The bifurcation value of the environment temperature is limited from above, since the second term in (12) for large values of T begins to dominate.

The maximum temperature of the irradiated sample $T = T_{max}$ for the area of instability satisfies the equation

$$1 - T_{\max}(\xi+1)\left(h + c\beta(T_{\max})\right)/hE_m = 0 \tag{13}$$

In this case, the bifurcation value of the defect generation rate also reaches maximum value

$$K_{\max} = hT_{\max}/(\xi+1)\theta \tag{14}$$

The line which corresponds to the isotherm $T = T_{\max}$ passes over the bifurcation curve, crossing it only at $T_e = 0$ and $K = K_{max}$.

The maximum temperature of the environment for the area of instability, $T_e^{\max}$, which corresponds to the defect generation rate $K=K^*$ and the temperature of the irradiated sample, $T=T^*$ satisfies the equations

$$2T^*\left(h - c\beta(T^*)\right) + c\beta(T^*)E_m = hE_m/(\xi+1) \tag{15}$$

$$K^* = T^{*2}\left(h + c\beta(T^*)\right)\big/\theta E_m \tag{16}$$

$$T_e^{\max} = T^* - K^*(\xi+1)\theta\big/h \tag{17}$$

The topological type of a stable critical point far away from the bifurcation curve is a stable node. It transforms into a stable spiral point near the bifurcation curve. On the bifurcation curve it transforms into a centre.

Parametric equations for the boundary where the critical point is a stable spiral point are

$$K = T^2\left(h + c\beta(T) - 2\sqrt{hc\beta(T)}\right)\big/\theta E_m \tag{18}$$

$$T_e = T - T^2(\xi+1)\left(h + c\beta(T) - 2\sqrt{hc\beta(T)}\right)\big/hE_m \tag{19}$$

The topological type of an unstable critical point is an unstable spiral or a node.

IV. Self-Oscillations and Their Parameters

The stable spiral becomes unstable when parameters pass through the bifurcation curve. Since for any parameters there is a loop without contact which covers the stationary point (5) – (6) and all phase trajectories of the system (3) - (4) go inside the loop there is a limit cycle of the system (3) - (4). Thus self-oscillations of the temperature and the defect density are developed.

The period of oscillations near the bifurcation curve is

$$\tau = 2\pi\sqrt{c/h\beta} \tag{20}$$

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

There is a phase difference between oscillations of defect density and temperature. The expression for the cosine of the phase difference is given by:

$$\cos\varphi = -1\Big/\sqrt{1 + hc/\beta} \tag{21}$$

If the heat capacity or the thickness of the irradiated plate is reduced, the period of self-oscillations decreases. The period increases together with an increase of the heat transfer coefficient. The period of oscillation depends on the pre-exponential factor of the diffusion coefficient and practically doesn't depend on the energy of the defect migration and the energy of the defect formation. In crystals with higher density sinks the region of instability is less and the frequencies of the self-oscillations are higher.

The parameters of self-oscillation are obtained for several kinds of metals (lead and aluminum) and non-metals (silicon). It shows the following facts. Stability diagram for different materials are similar. The highest environmental temperatures at which self-oscillations develops are about 100 - 200 K, at defect generation rate about 10^{-3}dpa/s. The temperature of the sample for these parameters is about 300 K. The frequency of oscillation is about 10^{-3} – 10^{-2} s^{-1}. 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 10^{-3}dpa/s. The temperature of the sample for these parameters is equal to 273 K. The frequency of oscillation is equal to 0.12s^{-1}. If the ratio ξ increases, the region of instability expands and the frequency of the self-oscillations increases too. During the development of the self-oscillations the temperature of the sample, heat transfer and the defect density can exceed the steady-state value by several times.

The region of the instability increases for more complex systems. They are the systems that take into account the secondary defect formation, e.g. complexes of the defects, voids, etc. The lifetime of secondary defects is much more than lifetimes of primary one. Thus the period of self-oscillation for more complex system grows considerably and can reach several days.

V. Conclusion

The examination of the nonlinear evolution of a complex system on the example of the accumulation of defects in irradiated crystals shows that there are two possible ways of evolution which are realized for different values of the system parameters. The defect density may monotonically grow and tends to its stationary state or may non-monotonically tend into self-oscillation.

The defect density, heat fluxes and the temperature during self-oscillations can reach much higher values than the expected steady-state values. It can lead to change of typical operation and an accident, e.g. for a nuclear reactor.

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