# LONG-RANGE EFFECTS IN SILICON SINGLE CRYSTALS IRRADIATED WITH PROTONS AND ALPHA-PARTICLES

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Radiation effects in silicon single crystals irradiated with protons of the energies $E = 6.8$, 43, and 50 MeV at the fluences $\Phi = (1 \div 3) \times 10^{17}$, $1 \times 10^{17}$, and $5 \times 10^{16}$ cm$^{-2}$, respectively, and with alpha-particles of the energy $E = 27.2$ MeV at the fluence $\Phi = 1 \times 10^{17}$ cm$^{-2}$ have been studied. The extension of a periodic defect structure into the region located behind the ion stopping range has been revealed ("long-range effects"), which cannot be explained in the framework of the available ion implantation theory. The effect of the proton radiation on an increase of the thermal defect generation in crystal growth layers located in this region is found to be more intensive and to occur at a temperature by 50° lower than that in the proton free-path region. In the case of the irradiation with alpha particles, the formation of a defect structure in the form of defect walls oriented perpendicularly to the ion beam and extending over the ion stopping range and behind it was detected. We associate the formation of a periodic defect structure with the self-organization of radiation-induced defects, and the extension of the radiation effect into the region behind the ion stopping range with a probable implementation of the soliton mechanism of propagation.

## 1. Introduction

The sporadic emergence of defects in the region behind the free-path one is one of the effects that accompany the process of heavy ion implantation into a crystal. Such a radiation influence was observed using various experimental techniques in semiconductors and metals at depths that sometimes exceeded the theoretical (projectional) free paths of ions, $R_p$. It is the so-called "long-range effect" (LRE) [1, 2]. Concerning accelerated atomic particles, it means a possibility for a defect structure created by them to extend over extremely far distances. However, the LRE peculiarities have not been studied enough for the action of light ions (protons and alpha particles), which are not used in the ion-doping technology. The LRE mechanism remains obscure till now. The authors of work [2] have analyzed experimental results and theoretical models of LRE at athermal external influences. The body of experimental data testified to an important role of the defect system in initial specimens and the collective processes occurring at its variation under those influences. The following theoretical models of LRE have been examined: diffusion, impact (defect generation under the influence of elastic waves), switching wave (a transition of the irradiated system into another steady state), and autowave (the asymptotic solution of system of nonlinear equations for the diffusion of vacancies and their complexes shows that vacancies propagate in the form of switching autowave) ones. A combination of the mechanisms described in the diffusion and impact models is the mechanism of energy transfer in the form of defect potential energy.

A collective rather than a pair interaction between substance atoms and energetic particles comprises an essentially new mechanism of the LRE model. In this case, the front of radiation-induced damages moves ahead much quicker than it does under conditions of the diffusion motion of individual point defects. In work [3], the analytical description of an irradiated substance with the help of a system of equations similar to hydrodynamic equations was proposed, which is probably admissible for a substance oversaturated with radiation-induced defects. According to the opinion of the authors of works [4–6], for metals and at high levels of radiation damages, the LRE is based on radiation-induced cooperative processes, at which the hydrodynamic motion of the vacancy plasma is realized.

The results of molecular dynamics simulations [7] showed that the generation of soliton-like pulses is possible, provided that either a group of atoms or an individual atom on a free surface is subjected to a high-energy irradiation. The energy, which is injected into the near-surface layers of a substance by means of soliton-like pulses, is transformed into the energy of the crystal defect system, including the corresponding dislocation structure.

In work [8], the model describing the emergence of nano-sized regions of explosive energy release accompanied by the shock-wave radiation emission in a condensed medium was considered. The appearance of such regions ("thermal peaks"), which result from the development of atomic displacement peaks, is a general phenomenon for various kinds of corpuscular irradiation (with particles, the mass of which is larger than that of nucleon). To describe irradiation-induced shock-wave processes analytically, the author used the hydrodynamic equations. The numerical solution of those equations confirmed the validity of the model describing the propagation of a rigid-profile soliton wave in a metastable medium.

Hence, the LRE belongs to challenging issues concerning radiation damages in materials. On the one hand, the distribution of radiation-induced defects and implanted ions behind the penetration depth is of first-order technological interest, because those data are important in many cases for a modification of materials by means of radiation. On the other hand, the research of this effect has an important fundamental value for understanding the nature of the interaction between radiation and the substance, which is described in a simplified manner in the available models. The existing theories, which are based on the concept of pair interaction between fast particles and atoms in the substance, predict similar distribution profiles for radiation-induced defects and implanted atoms [9]: both profiles are Gaussian-like by shape, and the maximum of the defect distribution is located on some surface nearer to the target (TRIM computer codes). Orientational effects can result in a deeper penetration of ions into the target along definite crystallographic directions [10]; however, the fraction of such ions in the general distribution is rather small. Up to now, no unified LRE theory has been developed. When constructing such a theory, our experimental data on the features of radiation action of high-energy light ions in silicon single crystals may appear useful.

## 2. Experimental Part

Silicon single crystals were irradiated with protons of the energies $E = 6.8$, 43, and 50 MeV at the fluences $\Phi = (1 \div 3) \times 10^{17}$, $1 \times 10^{17}$, and $5 \times 10^{16}$ cm$^{-2}$, respectively, and with alpha particles of the energy $E = 27.2$ MeV at the fluence $\Phi = 1 \times 10^{17}$ cm$^{-2}$ on U-120 and U-240 particle accelerators at the Institute for Nuclear Research of the NAS of Ukraine. The particle flux intensity was maintained at a level of $10^{12}$ cm$^{-2}$s$^{-1}$. During irradiation, the specimens were cooled using either liquid nitrogen vapors or flowing water. The specimen temperature did not exceed 100 °C at that.

In order to study the topographical image of a silicon defect structure, the specimen was cut along the irradiation direction into wafers which, after their surface having been subjected to mechanical and chemical treatments, were analyzed on a MIM-8 microscope and a JSEM-90 raster electron microscope. The transverse cross-sections of irradiated specimens were also examined making use of the x-ray topography technique in the Lang transmission-diffraction geometry.

## 3. Experimental Results and Their Analysis

When silicon is irradiated with high-energy charged particles, three defect regions are created, which differ from one another by the type and the character of the defect formation: the free-path region, the stopping region (due to the energy straggling), and the region behind the stopping one.

The results of our researches showed that, just after the irradiation of silicon with 6.8- and 43-MeV protons, the corresponding x-ray topograms (Fig. 1) pronouncedly reveal a straggling region (a black-white contrast characteristic of stresses in the crystalline lattice, which emerge in the proton stopping region) at the depths $R_p = 360$ and 9400 $\mu$m, respectively, from the irradiated surface. In the proton stopping region of specimens preliminary irradiated with 43-MeV protons and isochronously annealed at 800 °C, one may observe the formation of dislocations and their aggregations. If the annealing temperature is increased to about $900 - 1000$ °C, particles of the new phase and accompanying defects are formed in the growth layers of silicon crystals (Figs. 1,*b* and *c*). This effect can be observed in the region behind the stopping one at a temperature that is by 50 °C lower than the corresponding value for the free-path region (Fig. 1,*b*). Hence, the influence of irradiation on structural characteristics of a crystal in the free-path region and behind the stopping one manifested itself as an accelerated generation of thermal defects in the silicon growth layers, which is more intensive in the specimen region behind the free-path one, being observed there at a lower annealing temperature.

For silicon specimens irradiated with 6.8-MeV protons and cut out along the irradiation direction, raster electron micrographs revealed only a single light line at the depth of about $R_p$ (Fig. 2). However, for the same specimen, metallographic analysis found two defect "walls" at distances of 358 and 645 $\mu$m from the irradiated surface.

Metallographic researches of silicon specimens irradiated with 27.2-MeV alpha-particles at the fluence $\Phi = 10^{17}$ cm$^{-2}$ and providing the same projected penetration depth as for 6.8-MeV protons ($R_p = 360$ $\mu$m) revealed a formation of eight periodically arranged defect "walls" (see the first two columns in Table 1).

The raster electron micrographs of specimens irradiated with alpha particles revealed lines (see, e.g., Fig. 3 and the last three columns in Table 1) which can be associated – by their distance from the specimen surface – with the corresponding visually observable defect walls designated in the first column of Table 1 by Roman numerals.

In Fig. 3,*a* and *b*, the micrographs of the transverse cross-section of the specimen-3 surface are depicted; the mentioned lines are the most pronounced in them. The error of determination of the distance separating the irradiated specimen surface and the lines can reach 20 $\mu$m.

In Fig. 4, the computer-assisted photometry of a raster electron micrograph (Fig. 3,*b*) of a transverse cross-section of specimen-3 surface is exhibited. It illustrates the arrangement of defect "walls" with respect to the

**Distances (in micron units) between defect "walls" and the irradiated (alpha particles) surface of silicon specimens measured in metal-microscope and raster electron micrographs of the surfaces of specimen transverse cross-sections**

| No. Wall | Distance from defect "walls" and lines to the specimen surface (in micron units) | | | |
|---|---|---|---|---|
| | Metal-microscopy | Raster electron microscopy | | |
| | | Specimen No. 1 | Specimen No. 2 | Specimen No. 3 |
| I | 132 | 150 | 145 | 140 |
| II | 242 | 282 | | |
| III | 341 | 362 | 337 | 327 |
| IV | 380 | 385 | 380 | 352 |
| V | 423 | | | 441 |
| VI | 627 | 636 | 666 | 637 |
| VII | 720 | 674 | | |
| VIII | 764 | | | |
| | | 855 | 839 | |

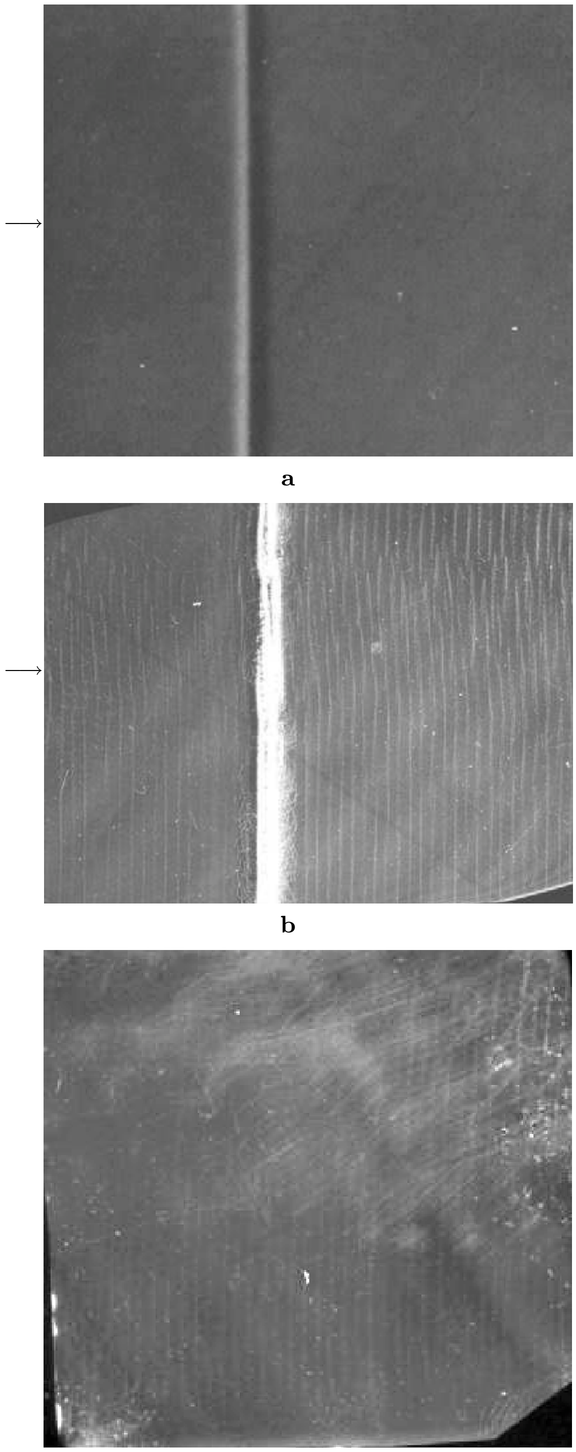
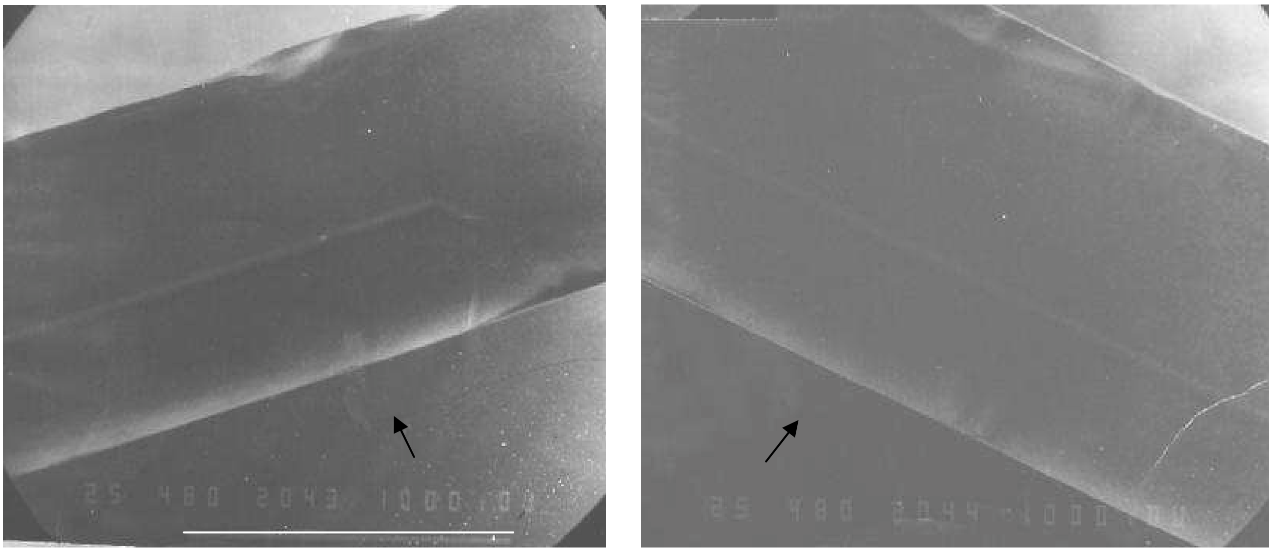
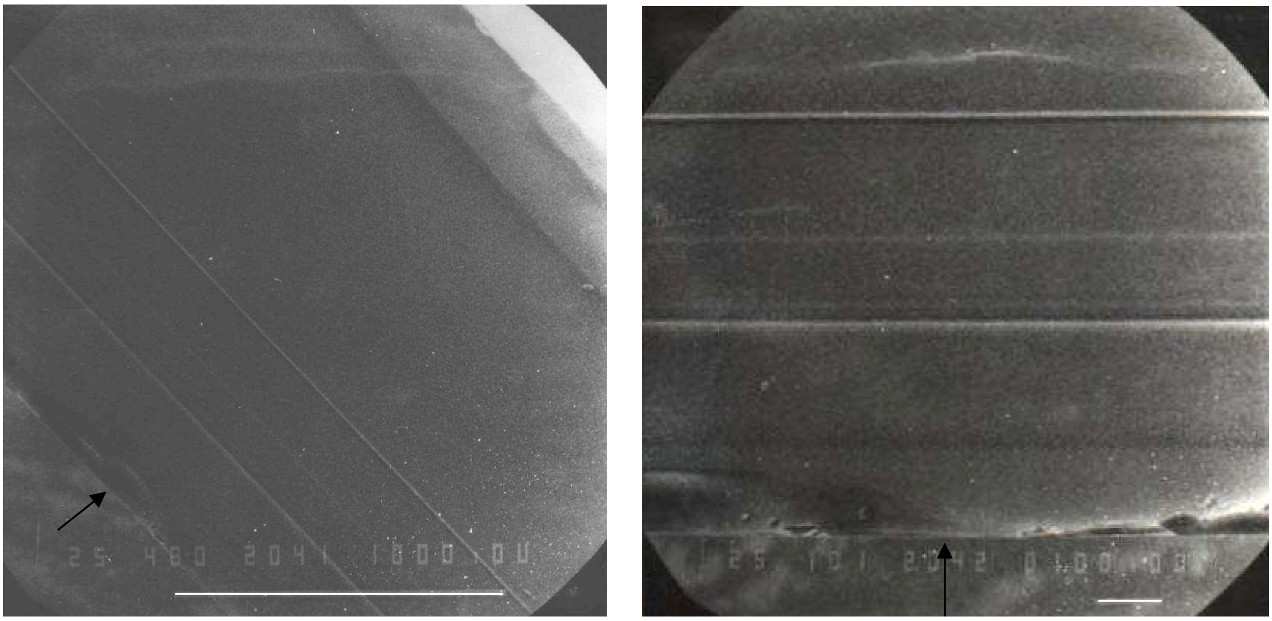



Fig. 1. X-ray topograms (at 9 times magnification) of silicon irradiated with 43- MeV protons at the fluence $\Phi = 10^{17}$ cm$^{-2}$ (*a*) before and (*b*) after the annealing at 1000 °C for 0.5 h; and (*c*) non-irradiated, but annealed at 1000 °C for 0.5 h



a b

Fig. 2. Raster electron micrographs of the silicon surface irradiated with protons ($E$ = 6.8 MeV, $\Phi = 10^{17}$ cm$^{-2}$). The irradiation direction [110] is parallel to the frame plane



a b

Fig. 3. Raster electron micrographs of the surface of a transverse cross-section of silicon specimen 3 irradiated with alpha particles ($E = 27.2$ MeV, $\Phi = 10^{17}$ cm$^{-2}$). The irradiation direction [111] is parallel to the frame plane

specimen surface and the intensities of corresponding lines. The line-to-background intensity ratio $I$ (in per cent units) is reckoned along the ordinate axis, and the distance (in micron units) from the irradiated specimen surface along the abscissa one. The positive values of $I$ correspond to white lines in Fig. 3,*b*, the negative ones to black lines. Attention is drawn by the fact that the intensities of corresponding lines are almost identical in all three defect regions of the crystal. When elucidating the nature of a defect in the "walls", we may only rely on available indirect data of work [11]. In that work, the obtained experimental data were used to calculate the distribution profiles of radiation-induced defects and implanted atoms after silicon having been irradiated with $Si^+$ ions. The calculations were carried out making use of the TRIM computer code. The results of calculations showed that the formation of point defects of the vacancy type prevailed in a region before about $R_p/2$, the formation of large vacancy clusters dominated in the next region located a little bit closer to $R_p$, and the generation of defects of the interstitial type commanded in a region between about $R_p/2$ and $R_p$.

Our researches [12,13] revealed the formation of aggregates (disordered regions) composed of divacancy defects and point-like vacancy-impurity ones in the proton-free-path region of the crystal. The efficiency of generation for defects of the latter type at the proton irradiation is two to three orders of magnitude higher than that at the neutron irradiation. The analysis of the IR absorption spectra of silicon irradiated with 6.8- MeV protons revealed a formation of a number of centers in the proton stopping region. Those centers include hydrogen localized at broken bonds of silicon atoms. The complete annealing of hydrogenous centers occurs at a temperature of 600 °C, being accompanied by the Si–H bond rupture. Provided that the annealing was carried out at a temperature higher that 600 °C, the irradiated (at the fluence $\Phi \geq 10^{17}$ cm$^{-2}$) part of the crystal with the thickness corresponding to the mean-free-path of 6.8- MeV protons in Si (of about 360 $\mu$m) was found to peel off. A by-product of our researches was the discovery of structural defects in the crystal region behind the free-path one, at a depth up to 720 $\mu$m. The concentration of those defects (of about $10^2$ cm$^{-2}$) was 5 to 8 times as high as the defect concentration in the proton free-path region.

According to the data of work [14], radiation-induced defects of the vacancy type are generated not only in the region of the free path of alpha particles, but also behind the region of their stopping. The reported results of researches testify that the light-ion irradiation affects the whole crystal region behind the ion-free-path one (Fig. 1,*b*), so that the LRE is obviously observed. In our case, an annealing temperature of 1000 °C, which invoked the accelerated generation of thermal defects in the growth layers of the irradiated crystal, served as an indicator for the effect detection. We observed a periodic defect system with a period shorter than $R_p$ only in specimens irradiated with alpha particles. Hence, its formation is most likely sensitive to the energy release density. The nature and the structure of defects in the

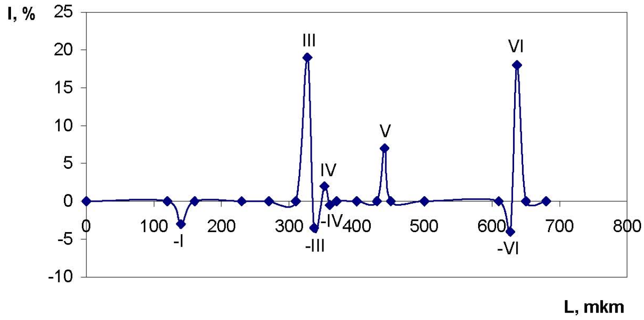



Fig. 4. Photometric analysis of a raster electron micrograph (Fig. 3) of the surface of a transverse cross-section of silicon specimen 3 irradiated with alpha particles. The irradiation direction coincides with that of the abscissa axis

"walls" require further studies. However, comparing the "wall" positions with the data of work [11], one may suppose that the light lines in Figs. 3 and 4 correspond to the aggregates of vacancy-type defects, and the dark ones to interstitial defects. The corresponding structure also extends into the crystal region behind the ion-free-path one.

The periodicity of the defect system and the presence of a fine-scale structure (e.g., in the region near $R_p$) testify to the wave mechanism of its formation. However, the known autowave mechanism [2] cannot explain the irradiation action at long and superlong distances of irradiation penetration into the crystal region behind the free-path one, because it assumes that the amplitude of the propagating concentration front of radiation-induced defects is kept constant owing to a plastic deformation. According to our data [12], a plastic deformation in proton-irradiated specimens takes place at a higher temperature (the rupture of Si–H bonds occurs at a temperature of about 600 °C) than the specimen temperature during irradiation. At the same time, another mechanism of radiation influence propagation was examined in work [7], namely, by engaging soliton-like pulses. In the case of high irradiation fluences and high ion energies, as it was in our work, a huge number of soliton-like pulses can be generated at the particle collision with the surface. It is probable that a new powerful sequence of pulses would be generated in the ion stopping region as well [8]. Owing to a high speed of pulse propagation in the crystal, the energy transfer and the energy redistribution over the crystal volume could be realized quickly enough, which would govern the LRE.

In our opinion, the soliton-like mechanism of radiation effect propagation is the most probable one under our irradiation conditions. It does not contradict any other known wave mechanisms, because solitons are "particle-like" nonlinear waves. In work [15], by analyzing the models of self-organization in various systems, attempts were made to formulate the general conditions needed for periodic coherent processes to be invoked. Under our conditions, the soliton mechanism of periodic process generation may probably be implemented. Hence, the formation of a periodic defect structure can find its explanation in the framework of the synergetic approach, as a result of the defect self-organization in the irradiated crystal [16, 17].

## 4. Conclusions

The radiation effect and the propagation of a periodic defect structure into the specimen region located behind the ion-free-path one ("long-range effects") have been revealed at the irradiation of single-crystalline silicon with high-energy protons and alpha particles. This phenomenon is not predicted by the available theory of ionic implantation. The radiation influence at the proton irradiation manifested itself as the enhanced generation of thermal defects in the silicon growth layers. The acceleration was more intensive in the specimen region located behind the free-path one, and this was observed at an annealing temperature by 50 °C lower than the corresponding temperature for the free-path region.

In the case of the alpha-particle irradiation, the formation of a periodic defect structure in the form of defect walls oriented perpendicularly to the ion beam has been detected. The emerged structure extends before the ion stopping region and behind it. The formation of the periodic defect structure can be a result of the defect self-organization process in the irradiated crystal. The extension of the radiation effect into the region behind the ion-free-path one may probably be implemented by means of the soliton mechanism.

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ОСОБЛИВОСТІ ЕФЕКТУ ДАЛЕКОДІЇ В КРЕМНІЇ ПРИ ІМПЛАНТАЦІЇ ЯДЕР ВОДНЮ ТА ГЕЛІЮ

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Р е з ю м е

У монокристалах кремнію при імплантації ядер водню (протонів) з енергією $E = 6{,}8$ МеВ ($\Phi = 1 - 3 \cdot 10^{17}$ см$^{-2}$), $E = 43$ МеВ ($\Phi = 1 \cdot 10^{17}$ см$^{-2}$), $E = 50$ МеВ ($\Phi = 5 \cdot 10^{16}$ см$^{-2}$) і ядер гелію (альфа-частинок) з енергією $E = 27{,}2$ МеВ, ($\Phi = 1 \cdot 10^{17}$ см$^{-2}$) виявлено радіаційний вплив та поширення періодичної дефектної структури в запробіжну для іонів частину зразків ("ефекти далекодії"), які не передбачені існуючою теорією іонної імплантації. Радіаційний вплив опромінення протонами на

прискорене утворення термодефектів у прошарках росту кристалів кремнію в запробіжній частині зразків є більш інтенсивним і спостерігається при температурі відпалювання на 50 °C нижчій, ніж у пробіжній. При опроміненні альфа-частинками виявлено утворення періодичної дефектної структури у вигляді "стінок" дефектів перпендикулярних до напрямку руху іонного пучка, яка спостерігається як до, так і за областю гальмування іонів. Формування періодичної дефектної структури пов'язуємо з процесом самоорганізації радіаційних дефектів, а поширення радіаційного впливу в запробіжну для іонів частину кристала – з можливою реалізацією солітонного механізму.