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INVESTIGATION OF RADIATION DEFECT FORMATION OF IRRADIATED n-Si<Pt>

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Abstract. In this work, radiation defect formation in silicon irradiated and doped with platinum is studied by the methods of deep level transient spectroscopy (DLTS) and positron annihilation spectroscopy. For the study, single-crystal n-type silicon samples doped with phosphorus during growth were used. These samples were first doped with platinum, then irradiated with 2 MeV protons at a current of 0.5 μA at the EG-5 accelerator. It has been established that the presence of Pt impurity in the bulk of n-Si leads to a slowdown in radiation defect formation: the concentration of vacancy defects in n-Si<Pt> samples is 2-3 times lower than in control samples.

Keywords: *semiconductor, silicon, platinum, irradiation, proton, positron annihilation, capacitance spectroscopy.*

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1. Introduction

It is known that the processes of complex formation in irradiated semiconductors are determined by the concentration ratios of certain defects, their charge state, and the defective structure of the crystal as a whole. The parameters of semiconductor devices, such as speed, forward voltages and reverse currents, the quantum yield of LEDs and lasers, and the gain of transistors are highly dependent on the presence of deep level impurities (DL) (Bogatov *et al.*, 2019; Utamuradova *et al.*, 2023). In addition, the behavior of materials under irradiation is largely determined by the nature and concentration of impurities that interact with primary radiation defects (Utamuradova *et al.*, 2022; 2023b). In some cases, to improve the speed of devices, silicon is specially doped with impurities or exposed to radiation, which leads to the formation of effective recombination centers. The influence of various external factors on the behavior of DLs created by Pt atoms in Si has been studied by different authors for many years, but their

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data are scattered and rather contradictory (Utamuradova *et al*., 2023e; 2023c).

Modification of semiconductor materials by beams of light ions, in particular protons, is one of the most promising and actively developing physical and technological methods in recent years. Interest in the implantation of silicon crystals by protons is due to a wide and controllable range of processed depths (from 0.1 μm to 1 mm) and the absence of complex radiation complexes with a high annealing temperature after irradiation. The main three factors affecting the change in the properties of semiconductors after proton irradiation are: a change in the electrical properties of semiconductors, radiation defect formation, and accumulation of hydrogen atoms (Kozlov & Kozlovskiy, 2001).

The aim of this work is to study radiation defect formation in silicon irradiated and doped with platinum using deep level transient spectroscopy (DLTS) and positron annihilation spectroscopy.

2. Experimental part

The objects under study were n-type silicon wafers 1.5 × 6 × 13 mm in size with a resistivity of 40 Ω cm (KEF-40). The wafers were cut from silicon ingots grown by the Czochralski method. Doping of silicon with platinum was carried out by the diffusion method from a layer of metallic platinum deposited onto the silicon surface in evacuated quartz ampoules at temperatures of 1100 and 1200 °C for 2 hours. Subsequent cooling of the samples was carried out using the thermal regimes given in (Utamuradova *et al*., 2021; 2023a). The phosphorus dopant concentration in the initial n-Si single crystals was 4.2×10^{14} at/cm^3. To carry out capacitive measurements on the samples under study, diode structures were fabricated according to a well-known technique (Komarov *et al*., 2013; Turgunov *et. al*., 2023). The measurements and processing of the spectra are also described in detail in (Komarov *et al*., 2013; Daliev *et al*., 2005).

After cleaning, the doped samples were irradiated with protons with an energy of 2 MeV, at a current of 0.5 μA to obtain a dose of 5.1×10^{14} particles / cm^2 using an electrostatic accelerator "EG-5" at the Laboratory of Neutron Physics of the Joint Institute for Nuclear Research (FLNP JINR).

Structural defects in the irradiated samples were studied by the method of positron annihilation spectroscopy - Doppler broadening of the 511 keV gamma line. The equipment with which the research was carried out is a positron beam equipped with in-depth analysis by changing the positron energy and is located on the territory of JINR, in the Laboratory of Nuclear Problems.

The elemental composition of silicon samples was determined by X-ray spectral microanalysis using a scanning electron microscope (SEM).

3. Results and discussion

Diffusion introduction of platinum atoms into n-Si leads to the formation of two DLs in the upper half of the band gap of n-Si<Pt>: Ec-0.20 eV and Ec-0.25 eV (see Fig.1).

An analysis of the measured DLTS spectra in doped and control samples showed that only one level Ec-0.25 eV is associated with platinum atoms in silicon, and the efficiency of the formation of this high level depends on the technological modes of introducing Pt into n-Si - temperature and diffusion time. The level Ec-0.20 eV is also observed in the control heat-treated (without Pt) samples, but its concentration is half an order of magnitude higher than in the doped samples (Fig. 1, curve 2).

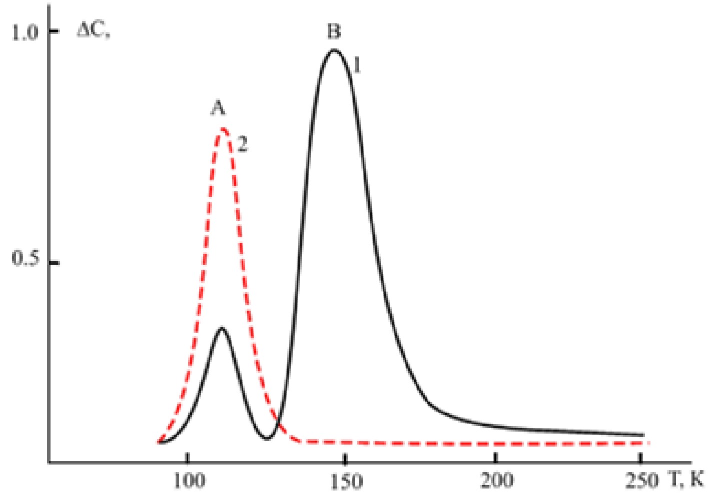

Fig. 1. Typical DLTS spectra of unirradiated n-Si<Pt> samples (curve 1, peaks A and B), control n-Si (curve 2, peak A)

It follows from a comparison of the measured DLTS spectra that as a result of irradiation, both in the control n-Si samples (Fig. 2, curve 3) and in the n-Si <Pt> samples (Fig. 2, curve 4), new levels are introduced with ionization energies Ec-0.17 eV and Ec-0.43 eV. The values of the parameters of this DL refer to the known radiation defects - vacancy-oxygen complexes (A-centers) and vacancy-phosphorus complexes (E-centers) (Utamuradova & Rakhmanov, 2022).

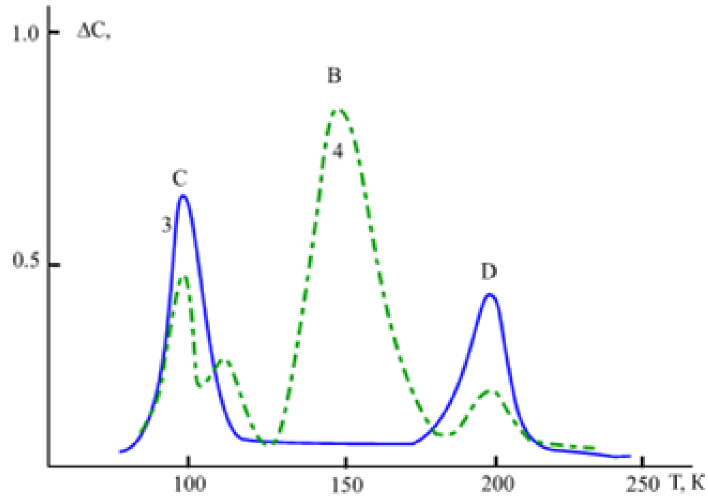

Fig. 2. Typical DLTS spectra of irradiated n-Si samples (curve 3, peaks C and D) and n-Si<Pt> (curve 4, peaks A, B, C, and D)

From the obtained results, it follows that the presence of platinum impurity in the silicon lattice leads to a slowdown in radiation defect formation: the concentrations of A- and E-centers in n-Si<Pt> samples are 2-3 times lower than in control samples. Moreover, the higher the concentration of platinum, the lower the concentration of radiation defects.

Thus, the presence of platinum atoms in the volume of silicon significantly reduces the efficiency of the formation of known radiation defects of A centers (vacancy–oxygen complexes) and E centers (vacancy–phosphorus complexes). This effect, apparently, should be associated with the features of the interaction of specially introduced impurities with defects introduced by irradiation.

The annihilation of positrons with electrons in solids provides information about the momentum distribution of these electrons. The electronic momentum distribution is reflected in the Doppler broadening of the 511 keV annihilation peak. Annihilation of positrons with valence or low momentum conduction electrons results in a small Doppler shift contributing to the center of the peak (Huis van M.A et. al., 2022). Annihilation with high-momentum nuclear electrons results in a large Doppler shift, contributing to the wings of the 511 keV annihilation peak. The shape of the peak at 511 keV is characterized by the parameters S and W, where S is the relative contribution of valence and conduction electrons to annihilation, and the parameter W is the relative contribution of core electrons (see Fig. 3).

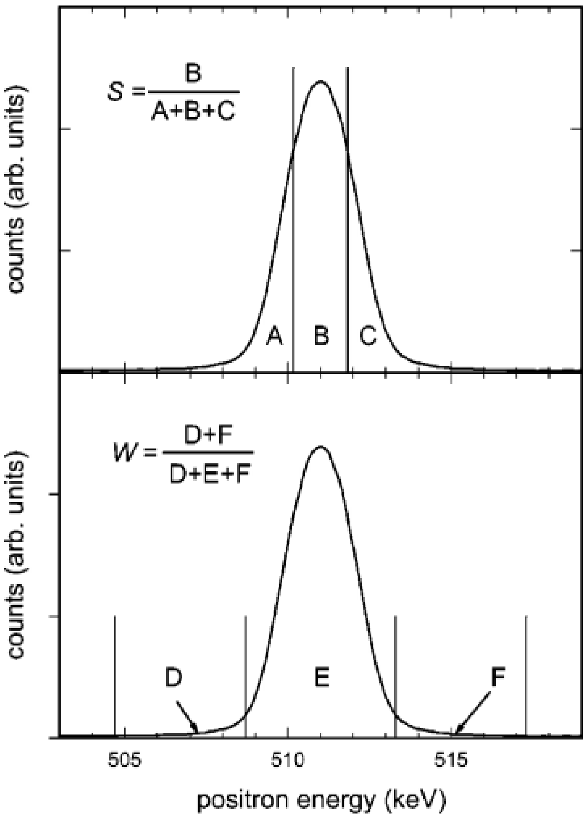

Fig. 3. Definition of the *S* and *W* parameter with the corresponding energy windows used in the analysis of the 511 keV positron annihilation peak (Huis van *et. al*., 2022)

In combination with a slow positron beam, this makes it possible to analyze the positron beam with Doppler expansion. The parameters S and W can be considered as a special volumetric property for each material.

Figure 4 shows the dependence of parameter S on the positron implantation energy in n-Si, n-Si<Pt> samples after irradiation with protons at room temperature, at which a noticeable increase in the peak range is observed (Fig. 4, 1-curve), which corresponds to a positron energy of 1–3 keV. As the radiation corresponding to positron implantation energies of 3–15 keV penetrates deep into the samples, the parameter S drops below the initial value in irradiated n-Si.

S parameter of the irradiated sample n-Si<Pt> is less than that of the initial irradiated sample (Fig. 4, 2-curve). An increase in the S parameter occurs in the positron energy range of 2–7 keV. At great depths, corresponding to positron implantation energies of 7–15 keV, the parameter S falls below the bulk value.

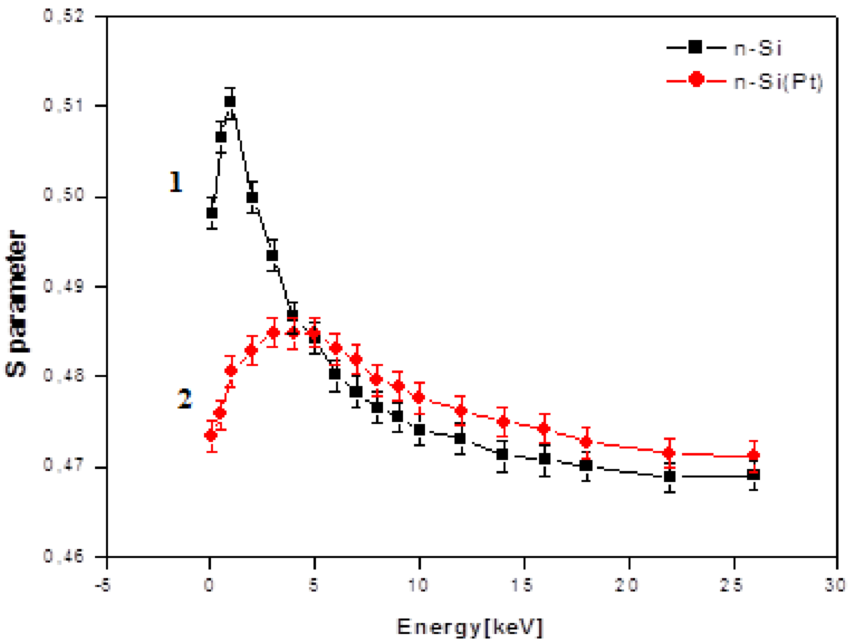

Fig. 4. Dependence of parameter S on the energy (respectively, depth) of annihilation positrons of irradiated n-Si(1) and n-Si<Pt>(2) silicon samples

The parameter S determines how much of the annihilation occurred with valence electrons (Selim, 2021). In these compounds, it is necessary to take into account the contribution of positronium annihilation, which manifests itself in the parameter S, to the total annihilation. Nevertheless, we can neutralize the effect of parapositronium annihilation on the parameter S by comparing the results of the obtained parameter W with the estimate of positron annihilation by nuclear electrons. In the absence of parapositronium destruction, the profiles of the W and S curves should be opposite to each other. This is seen in fig. 5. Curves n-Si and n-Si<Pt> in fig.4 are shifted relative to the maximum of the curve profiles in fig.5. First, if we compare n-Si and n-Si<Pt>, we can say that sample defects decrease with increasing positron energy. The visible result makes sense because the process is like magic. At the same time, in Fig. 4 during implantation, defect repair is observed throughout the entire depth of n-Si. At the same time, along with the radiation defect in the n-Si<Pt> compound, there are also structural defects caused by pressing. This is more clearly observed in the parameter S.

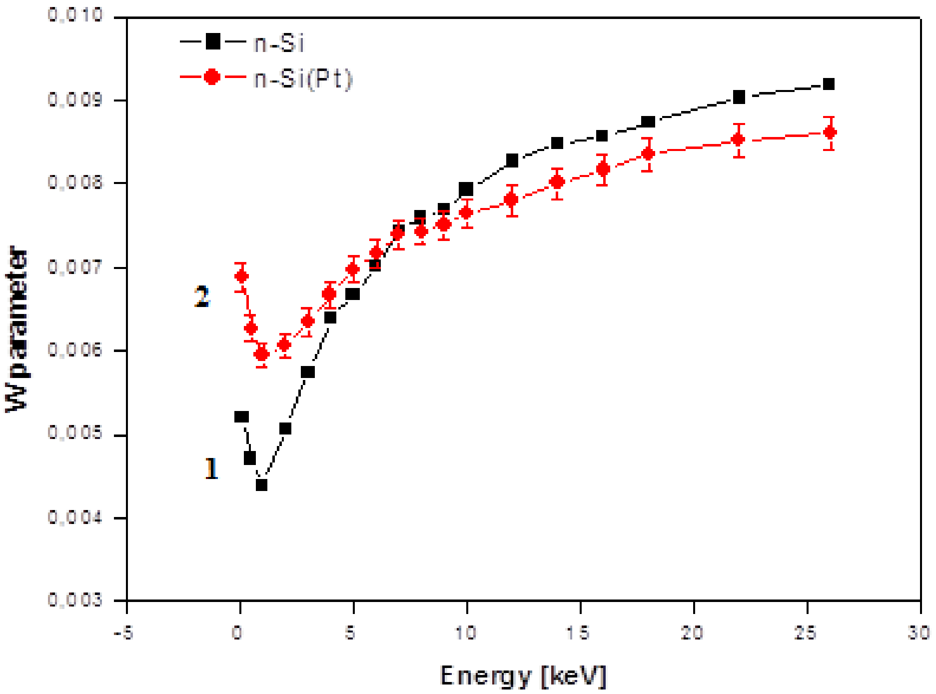

Fig. 5. Dependence of parameter W on energy (respectively, depth) of annihilation positrons of irradiated silicon samples n-Si(1) and n-Si<Pt>(2).

We know (Utamuradova *et al.*, 2023d). that the most significant factor of radiation exposure is radiation defect formation, in particular, the formation of vacancies. These defects include, first of all, oxygen vacancies O_i - vacancy V (A-center), divacancies (V-V) and a phosphorus complex P_S (at the site) - vacancy V (E-center).

It is written in (Brusa *et al.*, 2005) that positrons can get stuck in lattice imperfections. In particular, with defects of the vacancy type, where one or more ions are absent, the Coulomb repulsion felt by the positron decreases, and the positron senses such defects as potential wells. As a result, deep localized positron states are formed on open bulk defects with a binding energy of about 1 eV or more. An important limitation for semiconductors is that a vacancy defect must be either negatively charged or neutral. The Coulomb repulsion caused by the positive charge state prevents positrons from being captured.

In another work (Tuomisto *et al.*, 2010), it was said that the electron density locally decreases at a vacancy defect. This is reflected in the resulting positron lifetime, which is usually 30-80 ps longer than in a defect-free lattice. Therefore, the measurement of the positron lifetime provides information on the presence of vacancy defects in a given sample. Positron annihilation at a vacancy type defect also leads to changes in the momentum distribution studied in the Doppler broadening experiment. The momentum distribution resulting from the annihilation of valence electrons becomes narrower due to the lower electron density. In addition, a localized positron in a vacancy has reduced overlap with ion cores, resulting in a significant reduction in annihilation with high-momentum core electrons.

Evaluation of the so-called S-parameter (shape parameter), defined as the ratio of readings in the central part to the total area of the annihilation line. This parameter, with a value of about 0.5, is sensitive to electrons with low momentum (free or valence). Low-momentum annihilation of electrons, which are also present in open bulk defects, contributes to raising the central part of the peak and, consequently, to an increase in the value of the S-parameter. However, the S-parameter increases when there are more defects in which positrons can be localized (https://ifj.edu.pl/private/jdryzek/page_r18.html).

Based on (Utamuradova *et al.*, 2023d; Brusa *et al.*, 2005; Tuomisto *et al.*, 2010; https://ifj.edu.pl/private/jdryzek/page_r18.html), it can be assumed that after irradiation with protons with an energy of 2 MeV, both samples appeared radiation defects that depend on the vacancy (A-center, E-center, divacancy, etc.). However, the concentration of vacancy defects in irradiated n-Si is almost 2 times higher than in irradiated n-Si<Pt> samples. This shows that doping with platinum atoms leads to a decrease in radiation defects in the lattice of irradiated silicon.

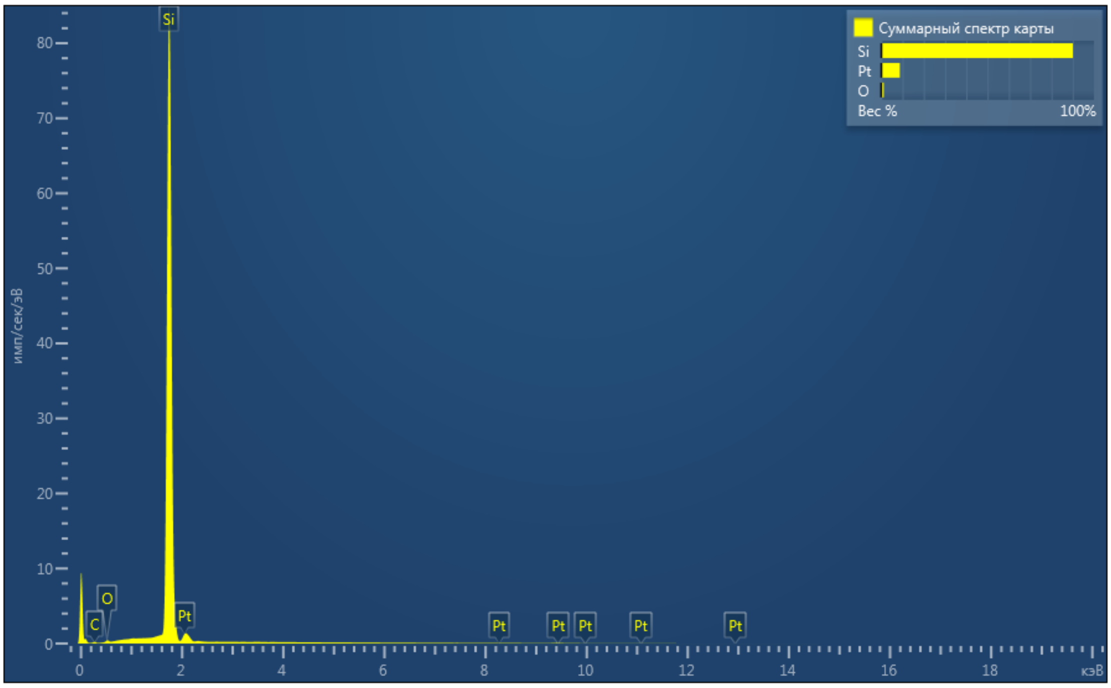

Fig. 6. Energy-dispersive spectra of silicon samples doped with platinum

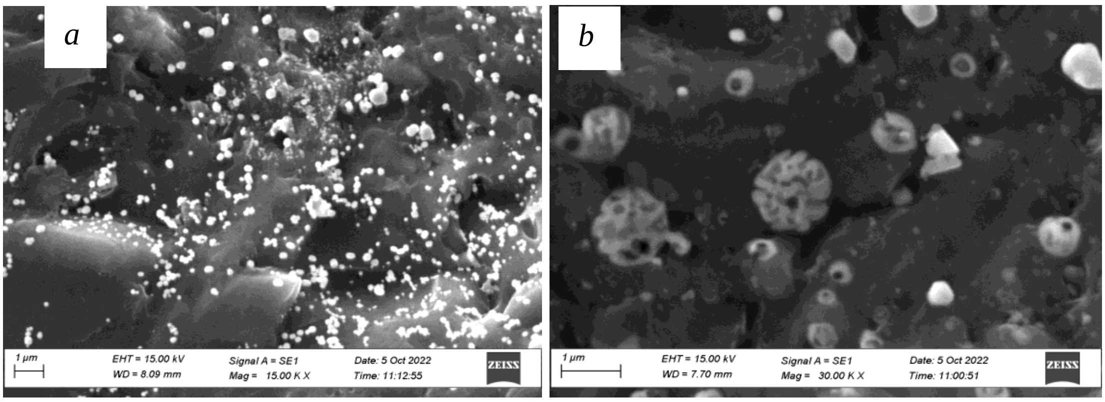

Fig. 7. Micrographs of the surface of silicon single crystal after doping with platinum (a), and after irradiation with protons (b)

The presence of platinum in single crystals is confirmed by X-ray analysis, according to which the content of platinum in the samples is 2 at.% or 12 wt.%. Energy dispersive spectra (Fig. 6) indicate the presence in the composition of the studied samples, in addition to platinum atoms, oxygen and carbon atoms. The content of oxygen atoms in the samples is 0.85 at.% or 1.2 wt.%, carbon atoms 0.7 at.%, respectively.

Microphotographs of silicon doped with platinum (Fig. 7a) indicate the presence of submicron formations in the silicon microstructure, which aggregate into spherical granules after proton irradiation (Fig. 7b).

4. Conclusion

Using DLTS spectroscopy, it has been established that the presence of platinum atoms in the volume of silicon significantly reduces the efficiency of the formation of known radiation defects of A-centers (vacancy-oxygen complexes) and E-centers (vacancy-phosphorus complexes). This effect, apparently, should be associated with the features of the interaction of specially introduced impurities with defects introduced by irradiation.

It was determined by positron annihilation spectroscopy that both samples (n-Si and n-Si <Pt>) after irradiation with protons with an energy of 2 MeV, at a current of 0.5 μA, with a dose of 5.1×10^{14} particles/cm^2, radiation vacancy defects appeared (A-center, E-center, divacancy, etc.). If we compare the original and doped samples, the concentration of vacancy defects in irradiated n-Si is almost 2 times higher than in irradiated n-Si<Pt> samples. This shows that doping with platinum atoms leads to a decrease in radiation defects in the lattice of irradiated silicon.

The energy dispersive spectra indicate the presence in the composition of the studied samples, in addition to platinum atoms, of oxygen and carbon atoms. The content of oxygen atoms in the samples is 0.85 at.% or 1.2 wt.%, carbon atoms 0.7 at.%, respectively.

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