

Грунтуючись на колишні роботи авторів, досліджено роль домішок бору (B) у формуванні вторинних радіаційних дефектів в кристалах кремнію (Si). Залежності цих процесів від температури ізохронного відпалу (в інтервалі 80–600 °C) вивчені з використанням холлівських вимірювань температурних залежностей (в інтервалі 100–300 K) концентрації і рухливості дірок в кремній до і після опромінення електронами з енергією близько 8 MeV при дозі $5 \cdot 10^{15} \text{ см}^{-2}$

Ключові слова: кремній, легуючий бор, радіаційні дефекти і комплекси

Основываясь на прежние работы авторов исследована роль примесей бора (B) в формировании вторичных радиационных дефектов в кристаллах кремния (Si). Зависимости этих процессов от температуры изохронного отжига (в интервале 80–600 °C) изучены с использованием холловских измерений температурных зависимостей (в интервале 100–300 K) концентрации и подвижности дырок в кремний до и после облучения электронами с энергией около 8 МэВ при дозе $5 \cdot 10^{15} \text{ см}^{-2}$

Ключевые слова: кремний, легирующий бор, радиационные дефекты и комплексы

UDC: 538.935

DOI: 10.15587/1729-4061.2015.47224

ROLE OF BORON IN FORMATION OF SECONDARY RADIATION DEFECTS IN SILICON

T. Pagava

Doctor of Physical-Mathematical Sciences, Professor*

E-mail: tpagava@gtu.ge

L. Chkhartishvili

Doctor of Physical-Mathematical Sciences, Professor*

E-mail: chkharti2003@yahoo.com

N. Maisuradze

Associate Professor, Candidate of Physical-Mathematical Sciences*

E-mail: nmaisuradze@gtu.ge

R. Esiava

Candidate of Technical Sciences, Assistant Professor*

E-mail: esrama@mail.ru

Sh. Dekanosidze

Candidate of Technical Sciences, Assistant Professor*

E-mail: Shorenadekanosidze@yahoo.com

M. Beridze

Doctor of Philosophy in Engineering Physics, Senior Lecturer*

E-mail: manchob56@mail.ru

N. Mamisashvili

Candidate of Technical Sciences, Assistant Professor*

E-mail: naniko@gtu.ge

*Department of Engineering Physics

Georgian Technical University

Kostaa, str., 77, Tbilisi, Georgia, 0175

1. Introduction

The p-type silicon (Si), which serves for basic semiconducting material in microelectronics, usually is obtained by the doping with boron (B). So, it is too important to understand the mechanism of interaction between B-dopants with radiation defects in silicon:

- to develop effective radiation treatment technologies for electronic devices and IC (integrated circuits);
- to improve their radiation resistance;
- to design the effective solid-state radiation sensors/detectors, etc.

Here based on authors' previous works, the dependences of the secondary radiation defects formation processes on isochronous annealing temperature are studied utilizing the Hall measurements of holes concentration and mobility in B-doped silicon before and after irradiation with high energy electrons. From these experimentally obtained results two main conclusions are made about boron atoms influence on capacities of active sinks of radiation defects in silicon crystals and their participation in screening the clusters of radiation defects.

2. Analysis of published data

Usually p-Si is obtained by doping with boron because B-atoms substituting Si-atoms in their regular sites form the shallow acceptor-levels. It is why the electron-transport in Si:B is widely investigated. Let us review in brief other properties of Si, which also are affected by doping with B.

In Si:B, the decrease in the lifetime of photo-carriers is believed to be due to their capture directly by dopants with emission of optical phonons [1]. Boron atoms in silicon act not only as isolated dopants, but also interact each with other and background impurities, as well as structural defects of the host Si-lattice. The shift in energy-levels from acceptor to acceptor related to the interactions between nearest neighbor dopants was demonstrated [2] by the single-electron capacitance spectroscopy of individual B-dopants in silicon. The electrostatic model of narrowing of the energy gap between Hubbard bands for boron atoms in silicon with increasing in their concentration and degree of compensation by the donors was advanced [3] taking into account the screening of impurity ions by holes hopping over

the B-acceptors. Studies on the intense highly polarized IR (infrared) luminescence from silicon nanostructures heavily doped with boron ($\sim 5 \cdot 10^{21} \text{ cm}^{-3}$) revealed [4] the ordered system of the dipole centers $\text{B}^+ - \text{B}^-$, which are caused by the elastic reconstruction of the shallow acceptors of boron.

The interaction between boron atoms also affects the process of their migration and consequently, distribution in silicon crystals. A diffusion mechanism of a diboron pair in Si was proposed [5]: a pair of substituting dopants $\text{B}_S - \text{B}_S$ diffuses from one lowest energy configuration to other equivalent structure. The kinetic model with the diffusion activation energy of 1.81 eV suggests that the diboron diffusion plays an important role in processes, which require high boron concentration as well as high annealing temperature.

The dependence of the boron distribution in silicon on the initial concentration of boron within the range $10^{19} - 10^{20} \text{ cm}^{-3}$ was studied [6] after a heat treatment of the samples implanted with B-ions. When the initial boron concentration exceeded the solubility limits in substitutional sites in solid at the annealing temperature, there are two additional peaks, which arose at both borders of the implanted region. Precipitation of boron implanted in Si followed with annealing was also investigated [7] and found that, at so-called critical concentration ($2.5 \cdot 10^{20} \text{ cm}^{-3}$) boron precipitation does not depend on the point defect concentration induced by the ion implantation.

Oxygen is the main background impurity in silicon crystals, especially, grown by Czochralski method. The annealing kinetics of the interstitial pairs B_IO_I formed by electron irradiation in the Cz-Si wafers with different boron concentrations was studied in [8]. The result that, the annealing rate of the B_IO_I complex at a fixed temperature increases monotonously with increasing in doping level, was treated by a model, which takes into account the interactions of interstitial boron with oxygen and substitutional boron atoms. An absorption band situated near 1026.7 cm^{-1} can be detected in Si with high contents both of oxygen and boron subjected to illumination [9]. Both B and O are the components of a defect to which this band corresponds. The co-doping with Ge reduces the efficiency of this formation. The work [10] also argues that the Ge-doping of the crystalline silicon suppresses the boron-oxygen defects responsible for the light induced changes in the carrier lifetime.

Usually, samples of n-type silicon are obtained by the doping with phosphorus (P), which forms the shallow donor-levels within the silicon band gap. Consequently, silicon simultaneously doped with B and P is of practical interests. The features of redistribution of the implanted P-atoms in a B-doped silicon were investigated [11] and it was shown that the high background boron doping ($2.5 \cdot 10^{20} \text{ cm}^{-3}$) drastically reduces the extension of the phosphorus profiles during the annealing. This result can be described in terms of the immobile P-B pairs (with formation energy of 0.6–0.8 eV).

In modern microelectronics, a number of special devices are made from the hydrogenated silicon Si:H. Gap states in B-doped microcrystalline Si:H were examined by analysis of the subgap optical spectra [12]. The absorption of the boron doped material was found to be very high.

It is interesting to consider simultaneous doping of silicon with boron and other group-III elements, e. g. aluminum (Al). According to the investigation [13] of the Frenkel-Pool effect in silicon doped at high and low levels with deep and shallow impurities Ga and B, respectively, a boron-related center is slightly deviated from the ideal single-charged

Coulomb-like center. Parameters of the electrically active centers induced during high temperature diffusion of B and Al into silicon were studied by the Hall-effect-techniques and capacitance spectroscopy measurements [14]. It was found that changes in resistivity of the n-base of p-n structures are controlled by the formation of three additional donor levels.

The defect production processes in p-Si{B,Pt} were studied by means of DLTS (deep-level transient spectroscopy) method [15]. Together B and Pt impurities affect generation of a radiation center. The phenomenon is explained in terms of decays of electrically inactive centers. Experimental data on the long-term relaxation of the photocurrent in compensated samples of Si{B,S} and Si{B,Rh} were analyzed in [16]. The sol-gel derived from nanoscale hybrid organic-inorganic films were used as advanced diffusion sources of B and Gd in silicon [17] and studied the features of their simultaneous diffusion.

Boron atoms in silicon can interact with self-interstitials Si_I . Appearance of the additional peaks on the boron concentration in silicon, which was implanted with boron ions and then annealed, at high initial doping level seems to be due to a knock-on reaction of substitutional boron atoms with self-interstitials Si_I , which have come in from the disordered bulk, and subsequent clustering of the produced excess in interstitial boron atoms [6]. As the concentration of boron atoms in substitutional positions in B-implanted Si crystals decreased to the equilibrium concentration, the boron precipitation takes place only in the regions with low concentration of point defects. However, in the regions with high point defect concentration, boron atoms become inactive because of involvement in clustering of self-interstitials leading to the formation of dislocation loops. A quantitative model was developed in [18]. According to the model of redistribution of B dopants in silicon during the proton-stimulated diffusion, the near-surface concentration peak is caused by migration of neutral B- Si_I pairs to the surface with their subsequent decomposition and accumulation of boron on the silicon surface in a thin layer. Precipitation of boron implanted in Si followed with annealing was investigated depending on the concentration of boron atoms in substitutional positions introduced before implantation [7]. The high value of the effective Hall mobility of holes in p-Si irradiated by high-energy electrons and annealed at 180°C indicates that in this material the p^+ -Si inclusions are formed [19].

Boron atoms in silicon even more intensively react with vacancies. According to the Review [20], in proton-irradiated Si crystals it takes place an irradiation-enhanced diffusion of boron. No doubt that, this effect is related to the radiation vacancies. However, obtained experimental results cannot be explained in the framework of a simple vacancy model. In boron doped p-Si samples irradiated with high-energy electrons, the activation energy of annealing for boron-vacancy complexes BV were identified as 1.6 eV [21, 22]. The substitutional boron-vacancy B_SV complexes in silicon were investigated theoretically in [23]. These results give an explanation of the experimentally reported metastability of the boron-related defect observed in p-type silicon irradiated at low temperature. The complex B_SV is found to have several stable configurations, depending on its charge state. Above mentioned p^+ -Si inclusions frequently are screened by the shells containing vacancy complexes BV characteristic of boron-doped silicon [19]. Some of quasi-chemical reactions that take place between radiation B-defects and impurities in irradiated silicon were described [24].

The substitutional boron–divacancy complex $B_S V_2$ in silicon was investigated [23] theoretically and found to be stable with a binding energy between V_2 and B_S of around 0.2 eV. Complex $B_S V_2$ is likely to be an important defect only in a material heavily doped with B. Its electron energy levels lie close to those of a free divacancy. The quasi-chemical reaction $B+V_2 \rightarrow BV_2$ was experimentally investigated in single-crystalline p-type silicon doped with boron, irradiated with high energy electrons or protons, and subjected to the high-temperature isochronous annealing [24]. Isochronous annealing of radiation defects with almost isoenergetical levels in p-Si, in particular, some (unidentified) vacancy-type and BV_2 complexes with a level of $E_v+0.22$ eV, were determined [25]. In n-type silicon with relatively low content of oxygen, it was found that divacancies are annealed through their decay [26]. Such a mechanism should be compared with direct conversion of divacancies into more complex defects by way of association with dopant B-atoms in p-type silicon. In the course of isochronous annealing of the Czochralski-grown p-Si specimens doped with B up to $6 \cdot 10^{13} \text{ cm}^{-3}$ and irradiated with high-energy electrons the conversion of divacancies, $B_S+V_2 \rightarrow B_S V_2$, takes place [27]. In [28], various radiation-induced defects have been identified in p-Si specimens doped with B and irradiated with electrons.

A study of effect of internal gettering process on the large-scale defect aggregates of the Czochralski-grown boron-doped monocrystalline silicon was made by the low-angle mid-IR light scattering technique [29]. Formation of shallow acceptor centers was found to occur in silicon irradiated by high doses of electrons, neutrons and high energy ions throughout a high-temperature annealing [30]. Acceptor was suggested to be multi-vacancy cluster activated by impurities, most likely by boron atoms.

Boron-related radiation defects in B-doped p-Si were specially studied for Si–SiO₂ structures [31–37]. The system exposed to irradiation with ~10 MeV electrons and also low-dose gamma-rays. These structures were studied by soft-X-ray emission spectroscopy using the variable-exciting-electron-energy optical ellipsometry and nuclear reaction techniques, DLTS measurements, TSC (thermally stimulated current), and quasi-static C–V methods. For the system prepared on a B-doped p-Si substrate, the irradiation-induced oxidation was not observed. It was also found that preliminary boron implantation blocks oxidation of the n-Si substrate. The spectra of non-irradiated samples exhibited only one peak corresponding to a deep level. It was shown that irradiation of structures results in the formation of a trap spectrum: four additional, shallower levels are found. The kinds of radiation-induced interface traps and their concentration depend on the disposition of the maximum of the previously implanted boron ions with respect to the Si–SiO₂ interface. It was shown that the main peak in the irradiated p-type structures correspond to the vacancy–boron complexes.

3. Purpose and objectives of the study

In this study, based on the review given above, including our previous works in the field [19, 21, 22, 24–28], we aim to investigate the role of boron-dopant atoms in formation of secondary radiation defects in silicon.

In particular, we experimentally investigate the dependences of these processes on isochronous annealing temperature based on Hall measurements of temperature-de-

pendencies of charge-carrier concentration and mobility in Si:B before and after irradiation with high energy electrons.

4. Research materials and methods

Boron-doped single-crystalline silicon samples with holes concentration of $\sim 6 \cdot 10^{13} \text{ cm}^{-3}$ were obtained by the zone-melting method.

They were irradiated with 8 MeV electrons at room temperature with exposure dose of $5 \cdot 10^{15} \text{ cm}^{-2}$ at flux density of $5 \cdot 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$.

The irradiated crystals were isochronously annealed in the temperature region 80–600 °C with a step of 10 °C and the 10 min time of sample holding at a fixed temperature.

Every annealing cycle was followed by the measurement of the hole concentration utilizing the Hall method in the temperature region 10–300 K.

Ohm contacts, necessary for such measurements were created by rubbing aluminum onto the surface of silicon samples.

The values of ionization energy for various defects-related centers were determined from the slopes of temperature-dependence of the hole concentration in corresponding temperature intervals.

The measurement errors for electrophysical parameters do not exceed ~10 %.

5. Research results and their discussion

5. 1. Impurity atoms of boron as radiation defects sinks

The dopant B-atoms in irradiated Si-crystals can serve for effective sinks of radiation defects by forming secondary radiation complexes. This conclusion can be derived from the measurements described below.

Namely, the temperature-dependences of hole concentration before and after irradiation and after annealing at different temperatures are shown in Fig. 1. Based on these results, the concentration changes for the majority current carriers – holes and some of radiation-induced defects are shown in Fig. 2 (curves 1 and 2–4, respectively).

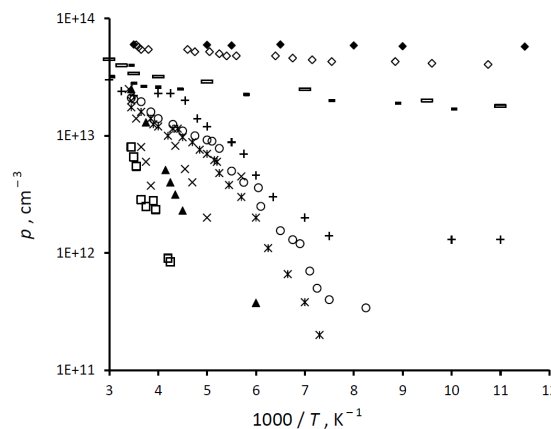


Fig. 1. Temperature-dependence of hole concentration in p-Si crystals 1 – before irradiation (◆), 2 – after irradiation with 8 MeV electrons at the dose of $5.0 \cdot 10^{15} \text{ cm}^{-2}$ before (□) and after annealing at 3 – 80 (▲), 4 – 90 (×), 5 – 120 (*), 6 – 170 (○), 7 – 180 (+), 8 – 380 (■), 9 – 470 (◻), and 10 – 600°C (◇)

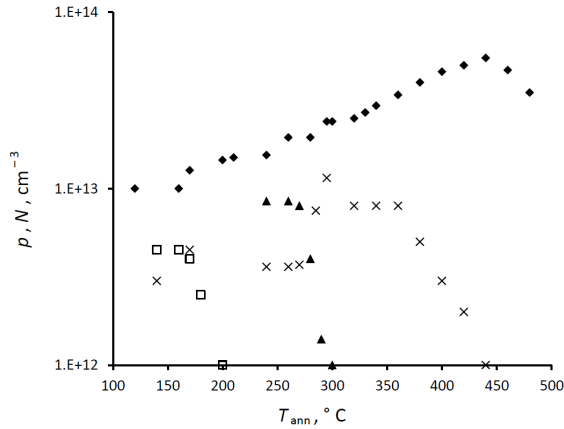


Fig. 2. Concentrations of 1 – holes (◆) and some of radiation defects: 2 – BV complexes (□), 3 – divacancies (▲), and 4 – H-centers (X), in p-Si crystals irradiated with 8 MeV electrons at the dose of $5.0 \cdot 10^{15} \text{ cm}^{-2}$ versus isochronous annealing temperature.

In the annealing temperature interval 170–200 °C, the sharp increase in holes concentration (curve 1) is associated with the annealing of defects with the energy level of $E_v+0.45 \text{ eV}$ and the concentration of $5 \cdot 10^{12} \text{ cm}^{-3}$ (curve 2). This level belongs to the complex of dopant atom, i. e. boron, with vacancy V:BV. These centers are quite deep acceptors and, therefore, up to room temperature their majority is positively charged by the capturing of holes. Thus, it takes place the following quasi-chemical reaction $B^-+V^++e^+ \rightarrow (BV)^+$. In the interval 270–300 °C, a defect with the energy of $E_v+0.28 \text{ eV}$ and the concentration of about $8 \cdot 10^{12} \text{ cm}^{-3}$ is annealed (curve 3). According to these values, it corresponds to divacancies V_2 [38]. In the course of divacancies annealing, a drastic increase of the concentration of defects with the donor level at $E_v+0.22 \text{ eV}$ – so-called H-centers – is observed (curve 4). Moreover, the concentrations of the divacancies decayed in the interval 270–290 °C and the emerged H-centers are equal to each other. It testifies that, H-centers do contain divacancies. In Fig. 1, the third step from below in the step-like curve 6 corresponds to the transition of electrons from the valence band onto a donor level $E_v+0.22 \text{ eV}$, which increases the hole concentration. Fig. 2 demonstrates that, process comes to the end at ~490 °C.

Divacancies-related donors $E_v+0.22 \text{ eV}$ take away their electrons from the valence band in the course of formation. Therefore, the value of holes concentration shows grow at divacancies annealing, although we have practically constant holes concentration in the interval 270–290 °C (curve 1). Probably, the complexes, which are formed at divacancies, contain boron atoms. The concentration of boron atoms, which become locked at the formation of H-centers and of the majority charge carriers, which are formed at divacancies annealing, are equal. Therefore, the variation of the concentration is zero in this annealing interval. These results are in agreement with the opinion [39] about the existence of B_5V_2 complexes of substituting, i. e. dopant, boron atoms in irradiated p-Si crystals, which are finally annealed in the temperature interval of 330–400 °C.

The activation energy of the divacancies migration is 1.30 eV, i. e. less than their binding energy 1.47 eV [38]. Therefore, divacancies can migrate without decaying over the crystal. On the other hand, BV complexes are annealed at a tempera-

ture of about 180 °C (curve 2) and, consequently cannot participate in the formation of B_5V_2 complexes at temperatures of 270–290 °C. So, it is possible to assume that, B_5V_2 complexes are formed by means of direct conversion of divacancies: $B_5+V_2 \rightarrow B_5V_2$, rather than the consequence trapping of a pair radiation-induced vacancies V by a boron atom B_5 .

Fig. 2 demonstrates that, H-centers are annealed in two stages, in the intervals 300–320 and 360–440 °C. Their initial concentration is equal to $3.5 \cdot 10^{12} \text{ cm}^{-3}$, and the same number of centers is annealed at the first stage. At the second stage, the concentration of decayed centers coincides with that of B_5V_2 complexes formed at divacancies annealing ($3.5 \cdot 10^{12} \text{ cm}^{-3}$). Thus, we can assume that B_5V_2 complexes are formed in the interval 270–290 °C in the course of divacancies annealing and dissociate at temperature of 400 °C. Concerning the centers annealed at the first stage, they are identical to B_5V_2 complexes by the ionization energy and are annealed in the temperature interval 300–320 °C.

The variation of hole concentration after the annealing at 300 °C is associated with the decay or formation of O_2V_2 -complexes, so-called K-centers formed as a result of trapping of a divacancy by a CO-center, and other deep centers characterized by a high thermal stability.

5. 2. Impurity atoms of boron as electrostatic screens

The irradiation of silicon crystals by high energy (above 10 MeV) particles leads to a complex structural damage in the form of disordered regions, which according to well-known Gossink model are non-transparent for the majority carriers – so-called “dielectric” inclusions. For such cases within the phonon scattering region the effective Hall mobility of majority carriers decreases due to the reducing the actual volume of the sample. However, in n-Si crystals, irradiated by protons with the energy of ~25 MeV, we observed [40, 41] the opposite effect – a sharp increase in the effective Hall mobility of electrons. It reveals the formation in the sample relatively high-conductive, i. e. “metallic”, inclusions with Ohm junctions at the interfaces with the semiconductor matrix [42]. Apparently, such inclusions are clusters of interstitials which are formed in the silicon irradiated with light ions and heat-treated [43]. Around themselves, they create a field of elastic stresses, which attract non-equilibrium vacancies – products of dissociation of the vacancy-type radiation defects during the isochronal annealing. A part of these vacancies recombine with interstitial atoms in clusters, but the rest of them reacts quasi-chemically with impurity atoms around the clusters creating acceptor centers, which depending on the charge-state and the annealing temperature can form negatively charged shells, non-transparent for the conduction electrons.

Here we explore similar effect in p-type Si crystals doped with boron and irradiated by 8 MeV electrons. Fig. 1 shows the temperature-dependence of hole concentration in samples annealed at different temperatures in the range 80–600 °C. The curve 5 corresponds to depletion of the center with energy level of $E_v+0.45 \text{ eV}$, which is annealed in the range of 170–200 °C – see the curve 2 in Fig. 2 showing the concentration of holes and that of some radiation defects as the function of annealing temperature. Judging from the values of the level energy and the annealing temperature, this center belongs to the complex of the boron atom with a vacancy BV. Of course, there are some other radiation defects with different ionization energies, which appear or disappear during the isochronal annealing. All they are finally annealed at 600 °C.

Fig. 3 shows the curves of the temperature-dependence of the majority charge carrier – hole mobility from the nitrogen up to room temperature. As seen, immediately after the irradiation with dose of $5.0 \cdot 10^{15} \text{ cm}^{-2}$ hole mobility drops sharply and continues to decrease with increasing annealing temperature. After annealing at 90°C , a deep minimum appears in the temperature-dependence. After annealing at 120°C , the curve with a minimum moves upwards, and after annealing at 180°C , it finally disappears, while the temperature-dependence itself is sharply enhanced (curve 7).

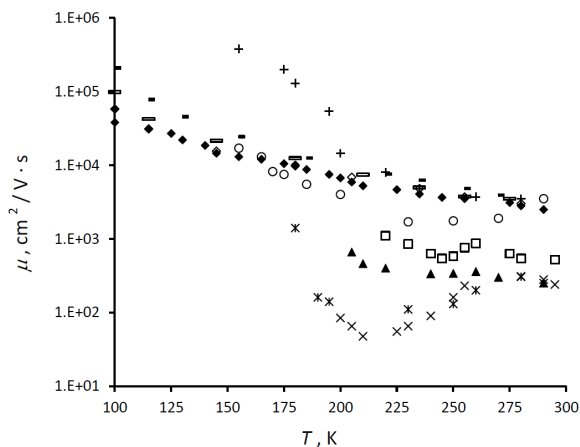


Fig. 3. Temperature-dependence of hole mobility in p-Si crystals 1 – before irradiation (◆), 2 – after irradiation with 8 MeV electrons at the dose of $5.0 \cdot 10^{15} \text{ cm}^{-2}$ before (□) and after annealing at 3 – 80°C (▲), 4 – 90°C (×), 5 – 120°C (×), 6 – 170°C (○), 7 – 180°C (+), 8 – 380°C (■), 9 – 470°C (≡), and 10 – 600°C (◊)

High mobility values obtained in irradiated samples by Hall measurements after their annealing at high temperatures point to the formation of the “metallic” inclusions. If for the simplicity these inclusions, which must be atomic clusters of p-type, are assumed to be spherical the effective Hall mobility of holes is

$$\mu_{\text{eff}} \approx \mu_{\text{H}} \frac{1+3f}{1-6f},$$

where μ_{H} is the Hall mobility in the matrix, and f is the total volume fraction of the “metallic” inclusions [42]. If the parameter μ_{H} takes the value of the Hall mobility at room temperature in the starting material, $2.5 \cdot 10^3 \text{ cm}^2/\text{V}\cdot\text{s}$, and μ_{eff} equals to $3.5 \cdot 10^3 \text{ cm}^2/\text{V}\cdot\text{s}$, the value measured at room temperature immediately after exposure (i. e. before any annealing cycle), we obtain the estimate $f \approx 0.035$. The average atomic radius of the spherical cluster can be determined by the formula

$$R \approx \sqrt[3]{\frac{3f}{4\pi N}},$$

where N is the concentration of inclusions, which in given conditions, according to our previous estimates [41], is equal to $5 \cdot 10^{16} \text{ cm}^{-3}$. From these values we obtain $R \approx 5.5 \text{ nm}$. A significant decrease in the mobility immediately after the irradiation, presumably, is due to the electrostatic screening of highly conductive atomic clusters by the secondary radiation defects of donor-type: the capture of holes makes them positively charged. Such inclusions affect the mobility

in two ways – as “dielectric” inclusions they block the electric current of majority charge carriers and thus reduce the actual volume of the crystal, and as static electric charges at low temperatures they effectively scatter holes. All these lead to a decrease in the mobility of majority carriers both in the phonon- and charged-centers-scattering temperature-regions. Further reduction in the mobility at initial stages of isochronal annealing ($80\text{--}90^\circ \text{C}$) must be caused by the negative annealing of the donor-type secondary radiation defects responsible for holes screening.

Non-monotonic temperature-dependence (presence of a minimum in the curve) of the effective Hall mobility of holes is explained by the non-monotonic variation of the degree of screening with samples irradiation temperature (see the theory in [19] and experimental curves in Fig. 3). As the temperature decreases from 300 to 200 K, the degree of filling of radiation defects, which screen the inclusions, increases. Accordingly, it increases the degree of screening of atomic clusters, what leads to a decrease in the effective Hall mobility. In the vicinity of 200 K, the energy of electrostatic interaction between the positively charged centers in impurities–defects shells around the atomic clusters reaches a critical value ($\sim 0.5 \text{ eV}$ [41]) comparable with the ionization energy of the corresponding deep levels and it starts their deionization. The effective mobility starts to grow, and a minimum appears on the temperature-dependency curve. Judging from the temperature range ($90\text{--}200^\circ \text{C}$) of the annealing of defects forming screening shells around the “metallic” inclusions, they are complexes BV.

The observed oscillations of the carriers Hall mobility in irradiated p-Si samples subjected to isochronous annealing at the constant rate of increasing in temperature, presumably are due to the changes in the degree of screening because the formation or annealing of different radiation defects affecting the screening. Highly conductive p-type clusters themselves are fully annealed at 550°C .

6. Conclusions

In summary, from the above consideration we can make two main conclusions about the role of boron dopants in formation of secondary radiation defects in silicon crystals:

- boron dopant atoms serve as very active sinks of radiation defects in silicon;
- complexes of radiation-induced structural defects in silicon with boron dopant atoms provide space charge-screening of the high-conductive inclusions in form of radiation defects clusters.

It is too important to gain the further insight in the interaction of boron dopants with radiation defects in silicon to develop effective radiation treatment technologies for silicon-based electronic devices and IC, increase their radiation resistance, design solid-state radiation sensors and detectors, etc.

As the rates of quasi-chemical reactions with boron-containing complexes have been found to be dependent on the charge-states of reactants, they are controllable by varying the irradiation conditions – beam-intensity, irradiation temperature and IR light exposure during the irradiation, as well as temperature of annealing of the already irradiated samples.

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