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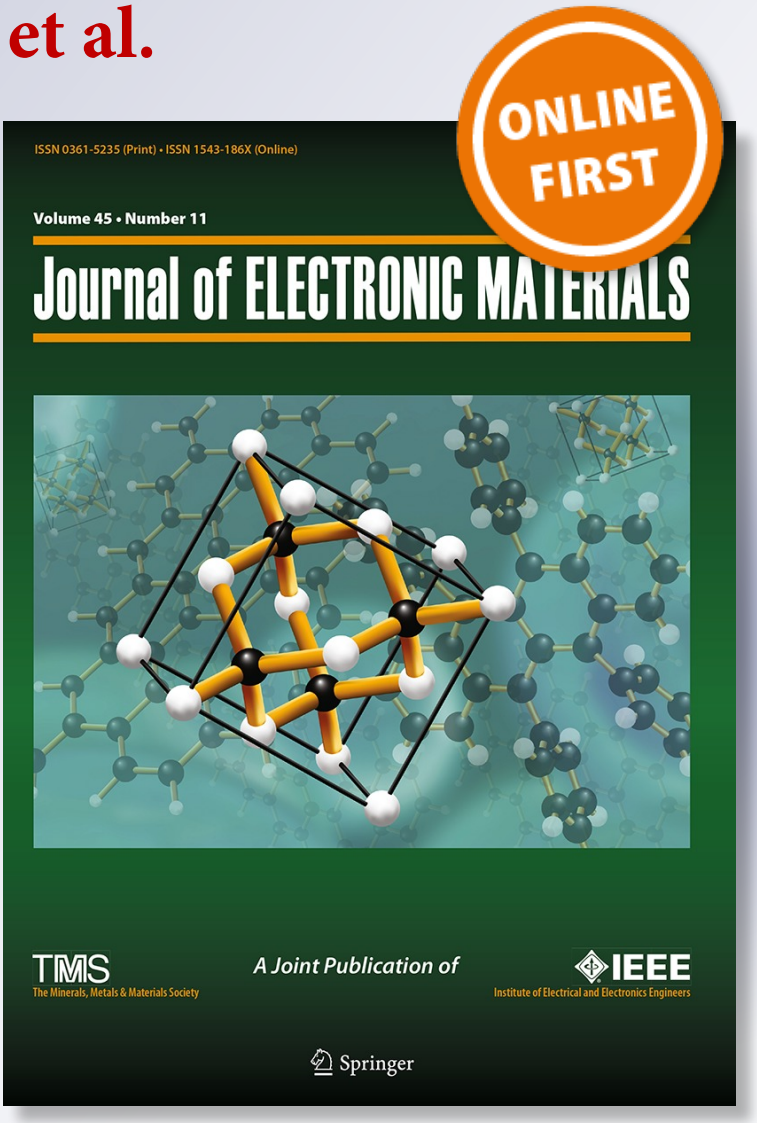

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Simulation of Electronic Center Formation by Irradiation in Silicon Crystals

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We present the results of a study on localized electronic centers formed in crystals by external influences (impurity introduction and irradiation). The main aim is to determine the nature of these centers in the forbidden gap of the energy states of the crystal lattice. For the case of semiconductors, silicon (Si) was applied as model material to determine the energy levels and concentration of radiation defects for application to both doped and other materials. This method relies on solving the appropriate equation describing the variation of the charge carrier concentration as a function of temperature $n(T)$ for silicon crystals with two different energy levels and for a large set of N_1, N_2 (concentrations of electronic centers at each level), and n values. A total of almost 500 such combinations were found. For silicon, energy level values of $\varepsilon_1 = 0.22$ eV and $\varepsilon_2 = 0.34$ eV were used for the forbidden gap (with corresponding slopes determined from experimental temperature-dependent Hall-effect measurements) and compared with photoconductivity spectra. Additionally, it was shown that, for particular correlations among N_1, N_2, and n, curve slopes of $\varepsilon_1/2 = 0.11$ eV, $\varepsilon_2/2 = 0.17$ eV, and $\alpha = 1/2(\varepsilon_1 + \varepsilon_2) = 0.28$ eV also apply. Comparison between experimental results for irradiation of silicon crystals by 3.5-MeV energy electrons and Co^{60} γ-quanta revealed that the $n(T)$ curve slopes do not always coincide with the actual energy levels (electronic centers).

Key words: Silicon, irradiation, Hall effect, carrier concentration, radiation defects, energy levels

INTRODUCTION

Despite the large amount of data in literature concerning the energy levels of various electronic centers in the forbidden gap of semiconductors, especially silicon,[1–9] quantitative and definitive determination of their occurrence and concentrations represents a difficult issue. Determination of doped and other energy-level parameters in the forbidden gap of semiconductors can be achieved by measuring curves from Hall-effect experiments, but it is not always possible to arrive at an unambiguous interpretation of the dependence of the charge carrier concentration on temperature $n(T)$. The absence of clarity on this issue prevents fully confident conclusions from being reached in a number of cases, based on analysis of precise values for the energy-level parameters, and proper interpretation of a number of regularities observed, for example, in irradiated crystals. Therefore, correct determination of energy-level parameters from experimental $n(T)$ curves for samples with known initial properties is interesting from both a scientific and practical point of view. Instead of using such "samples," different sets of values of ε_1, ε_2, N_1, N_2, and n can be considered with the process of $n(T)$

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measurement being replaced by an exact analytical solution for $n(T)$ obtained with the help of relevant computational fitting procedures. In this case, precise evaluation of the $n(T)$ processing results is provided, and satisfactory agreement is found with experimental data.

The simplified model of secondary radiation defect formation from simple Frenkel pairs (a vacancy with an interstitial atom) enables determination of the average number of free vacancies (λ_v) formed by a single absorbed particle (electron or γ-quantum from Co^{60}), following a linear dependence of the secondary radiation defect concentration on the irradiation dose Φ.[1,2,7,8] It is further supposed that λ_v does not depend on the radiation intensity φ and remains constant over time; also, among the various possible types of secondary defect formation, only one is examined, the others being intended subjects for future study. Determination of the radiation defect introduction rate has shown that, in n-Si, the first assumption is approximately satisfied, at least over a definite time interval t. From consideration of the formation of two types of center (A- and E-centers), it was possible to additionally determine some correlations between the parameters defining these centers. The assumed constancy of λ_v is violated in p-Si, which can be attributed to the dependence of λ_v on the concentration of electrons in the conduction band. This dependence may be due to one of the following reasons:

1. Primary defects are considered to be Frenkel bound pairs. The ratio between the decay probability of this pair to a free vacancy with interstitial atom formation and their annihilation probability essentially depends on n (increases with n growth), as capture of an electron by the bound pair greatly increases the relative probability of its decay.[1–3]
2. The diffusion rate of an uncharged vacancy is significantly lower than that of a vacancy capturing an electron.[2] In this case, λ_v represents the average number of negatively charged free vacancies per irradiating particle, which is proportional to n.

In both cases, λ_v will change over time according to the $n(t)$ dependence. If the concentration of nonequilibrium electrons n' exceeds the concentration of equilibrium electrons n_0, λ_v will therefore depend on the radiation intensity (φ). Adopting these considerations, one can obtain the average number of free vacancies in n-Si per irradiating particle, assuming that there are no other sinks for vacancies apart from phosphorus and oxygen atoms: $\lambda_v = (1 \pm 0.3) \times 10^{-2}$ vacancy/quantum for a γ-quantum from Co^{60} and $\lambda_v = (1 \pm 0.5)$ vacancy/el in the case of electron irradiation (energy 1 MeV to 4 MeV),[2,8,10–12] which is also of practical significance.

EXPERIMENTAL PROCEDURES AND DATA PROCESSING

The quantity and electronic structure of radiation defect centers formed in silicon crystals under irradiation by γ-quanta and electrons were both analyzed. Electron irradiation was performed using a linear accelerator with a picosecond pulsed electron beam at energy of 3.5 MeV at the Center for the Advancement of Natural Discoveries Using Light Emission (CANDLE) Synchrotron Research Institute (Armenia); the maximum irradiation dose was 6×10^{13} el/cm^2. Irradiation by γ-quanta was carried out using the Co^{60} source installation at the Physics Institute of Academy of Sciences of Ukraine. Hall-effect measurements were carried out on silicon samples with the following parameters: 1 mm thickness, 3 mm width, 10 mm length, and 6 mm distance between conductivity contacts, having a double-cross shape. Measurements were carried out in the temperature range from 80 K to 300 K, with accuracy of 0.5 K. The electrical parameters of the samples were determined from Hall-effect measurements with magnetic field induction of 0.5 T, direct current through the sample of 100 μA to 2 mA, and voltmeter input resistance of 10^{12} Ω. The concentrations of charge carriers, their mobility, and the concentrations of energy levels produced were estimated by measurements using the four-probe method. Photoconductivity measurements were carried out in the infrared region on the modulated beam at frequency of 180 Hz, normalized to the power of the infrared radiation. These measurements were performed at 300 K and 80 K using illumination from the monochromator of the SF-8 spectrophotometer (Russian production), employing a common measurement scheme: light—sample—loading resistance (with parallel-connected battery)—amplifier—detector (PC).

Experimental results showed that irradiation with Co^{60} γ-quanta produced defects with energy levels of $E_v + 0.22$ eV and $E_v + 0.34$ eV in the forbidden gap of p-Si.[4,5,12–15] These levels were determined from the slopes of curves resulting from Hall-effect measurements. It is further shown from the $p(T)$ measurements in p-Si that, in cases when a slope of 0.34 eV is observed, no deeper levels occur and the concentration of the levels at $E_v + 0.34$ eV may be less than or equal to the initial concentration of free holes in the sample. In the latter case, the presence of weakly compensated deep levels at $E_v + 0.34$ eV ($N_2 - N$) and donor levels at $E_v + 0.22$ eV can result in a slope of $p(T)$ of (0.22 eV + 0.34 eV)/2 = 0.28 eV with wide variation in the concentration of the $E_v + 0.22$ eV level. To explain this phenomenon, detailed analysis of $n(T)$ was performed using the computer model experiment with two energy levels present, and compared with experimental data.

The charge carrier concentration in the presence of two energy levels may be estimated from the

crystal electroneutrality equation describing the conditions for electron transition between the levels and the conduction band (c-band):

$$n + n_1 + n_2 = N, \tag{1}$$

where n is the electron concentration in the c-band, n_1 and n_2 are the electron concentrations in the two energy levels, and N is the total number of electrons in the c-band and the levels, which is proposed to be constant. For the sake of simplicity, we consider an n-type semiconductor and one energy level, for which case Eq. 1 leads to Eq. 2:

$$n + \frac{nN_1}{n + \theta_1} = N, \tag{2}$$

where N_1 is the concentration of the considered electronic center with level ε_1, $\theta_1 = \theta \exp\left(-\frac{\varepsilon_1}{kT}\right)$, θ is the density of states in the c-band, ε_1 is the ionization energy of the level calculated from the bottom of the c-band, k is Boltzmann's constant, and T is the absolute temperature. In this case, the determination of $n(T)$ is simplified and the logarithm of the $n(1/T)$ dependence presents a sequence of straight lines with tangent of the slope angle (multiplied by Boltzmann's coefficient) equal to the energy of the local level: ε_1, $\varepsilon_1/2$, or zero; This has been well described in previous work.[2] Equation 2 was solved for the following particular cases: $N \gg N_1$, $N \approx N_1$, $N_1 \gg N$, and $N_1 \geq N$. However, in these cases, there are wide temperature intervals in intermediate regions where the determination of the slope angle from experimental curves will present great difficulties, and the parameters of levels can be estimated by correct solution of Eq. 2, which, for the sake of simplicity, can be presented as in Eq. 3:

$$n = \frac{N - N_1 - \theta_1 + \sqrt{(N - N_1 - \theta_1)^2 + 4N\theta_1}}{2}. \tag{3}$$

Equation 3 formed the basis for the computer modeling of the time dependence of the energy levels and for estimating their parameters for comparison with experimental data. The resulting sets of curves obtained from irradiation of p-Si by Co^{60} γ-quanta to dose of 6.4×10^{17} cm^{-2} are presented in Fig. 1. Some of these energy levels coincide with experimental results obtained from Hall-effect measurements and photoconductivity measurements. Thus, the presence of real energy levels is indicated, while the other curves correspond to intermediate concentrations of electrons with these corresponding energies. Indeed, for many irradiated silicon samples in the experiments, the presence of curve slopes of 0.28 eV was revealed by Hall-effect measurements.[6,7]

On the basis of the above, it must be realized that the presence of such a slope does not necessarily imply the existence of a local level at E_v + 0.28 eV, but is rather produced by the existence of levels at

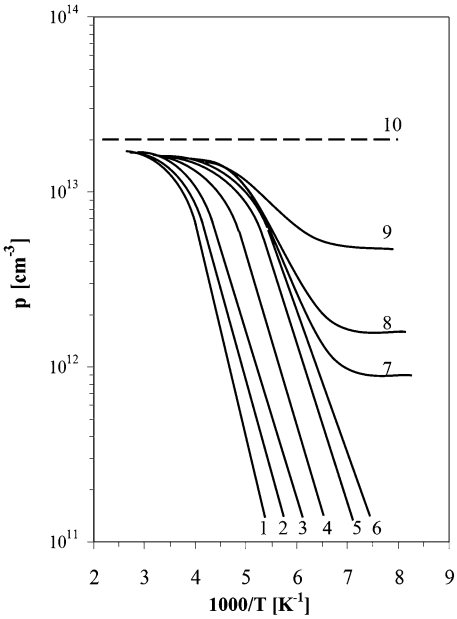

Fig. 1. Computer simulation of temperature dependence of charge carrier concentration for E_v + 0.22 eV level in p-Si after irradiation by Co^{60} γ-quanta (dose 6.4×10^{17} cm^{-2}). The curves were calculated for different concentrations of energetic centers determined as: (1) 2.4×10^{14} cm^{-3}, (2) 2×10^{14} cm^{-3}, (3) 1.5×10^{14} cm^{-3}, (4) 7×10^{13} cm^{-3}, (5) 4.5×10^{13} cm^{-3}, (6) 3×10^{13} cm^{-3}, (7) 1.6×10^{13} cm^{-3}, (8) 1.3×10^{13} cm^{-3}, (9) 1×10^{13} cm^{-3}, and (10) experimental value of charge carrier concentration before irradiation.

E_v + 0.22 eV and E_v + 0.34 eV. However, it is clear that, when a level does exist at E_v + 0.28 eV, the slope of 0.28 eV must also be observed. Therefore, other types of measurements, in addition to those of the Hall effect, must be carried out to estimate which of these cases actually applies. Figure 2 presents calculation results for the hole concentration $p(T)$ at $N_1 = 1.5 \times 10^{14}$ cm^{-3}, $N_2 = 5.15 \times 10^{13}$ cm^{-3}, and $p = 5 \times 10^{13}$ cm^{-3} for boron-doped p-type silicon. As is seen, along with slopes 1 and 3, it is highly likely that slope 2 will also be observed, with a value equal to the half-sum of the other two. Hence, we may stress that use of commonly accepted methods is insufficient for precise determination of the energy levels in such cases when intermediate regions of p values are comparatively high and the temperature interval, in which $p(T)$ measurements are carried out, is relatively narrow. The experimental results for the spectral distribution of photoconductivity served as an additional line of evidence to confirm the existence of the proposed energy levels and an associated radiation defect (Fig. 3).

This condition is essential, in particular for determination of deep energy levels where $n(T)$ decreases very rapidly as a function of temperature and the measurements are complicated (Fig. 4, curves 3, 4). With the above-described "model" presentation, this disadvantage is eliminated.

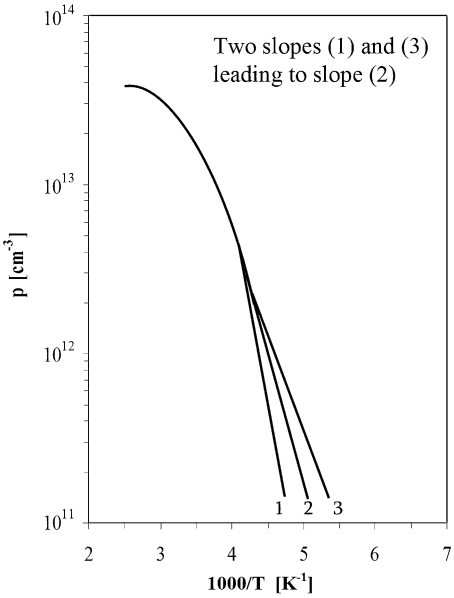

Fig. 2. Computer simulation (special case from Fig. 1) indicating the existence of slope 2, equal to the half-sum of the two other slopes (1 and 3), for the given case at (0.22 eV + 0.34 eV)/2 = 0.28 eV. The curves were calculated for concentrations of charge carrier N and energy levels N_1 and N_2. (1) $N = 5 \times 10^{13}$ cm^{-3}, $N_1 = 1.5 \times 10^{14}$ cm^{-3}, $N_2 = 5.15 \times 10^{13}$ cm^{-3}; (2) $N = 5 \times 10^{13}$ cm^{-3}, $N_1 = 1.5 \times 10^{14}$ cm^{-3}, $N_2 = 5.05 \times 10^{13}$ cm^{-3}; (3) $N = 5 \times 10^{13}$ cm^{-3}, $N_1 = 5 \times 10^{13}$ cm^{-3}, $N_2 = 5.5 \times 10^{13}$ cm^{-3}.

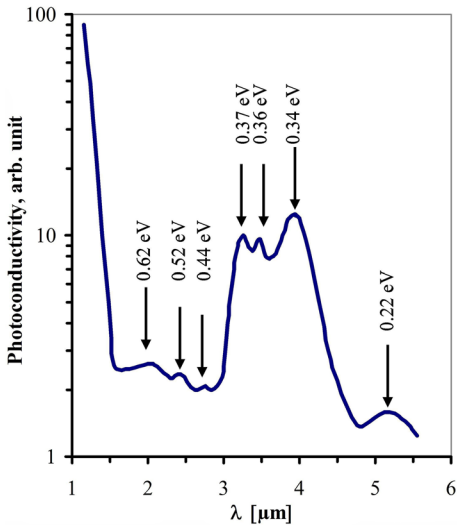

Fig. 3. Photoconductivity of p-Si irradiated by Co^{60} γ-quanta, confirming the existence of energy levels at $E_v + 0.22$ eV and $E_v + 0.34$ eV. Measurements carried out at temperature of 80 K.

However, in the experiments with picosecond (10^{-12} s) pulse electron irradiation, the effect on the materials of electron irradiation with very short pulse length should also be taken into consideration. The latter requires an investigation its own right, but for the present discussion it is not important. We note here that thermal processes do not occur because the irradiation pulse duration is

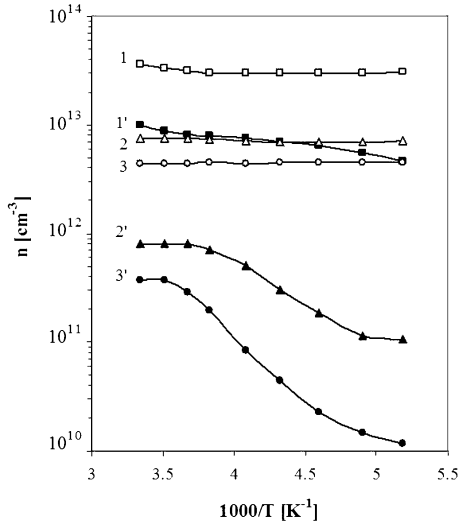

Fig. 4. Temperature dependence of main charge carrier concentrations for n-Si irradiated by picosecond pulse electrons with energy of 3.5 MeV. Specific resistances of samples were 120 Ω cm before (1) and after irradiation with dose of 6×10^{13} el/cm^2 (1′), 720 Ω cm before (2) and after irradiation with dose of 1.2×10^{13} el/cm^2 (2′), and 950 Ω cm before (3) and after irradiation with dose of 1.2×10^{13} el/cm^2 (3′). Curve slope analysis indicated an oxygen-containing cluster center at $E_c - 0.16$ eV.

very short with respect to the thermal effect (order of magnitude in excess of 10^5).

It is known that radiation applied at the same dose, but different intensity, can create different structural defects in semiconductors, which, in turn, can form various energy levels in the forbidden gap of the zone structure of semiconductors.[14–17] Here, we also consider cases where the radiation intensity is significantly (almost an order of magnitude) different, but the dose remains constant. Irradiation at low intensity is called "slow" (S, 10^{11} el/s cm^2), whereas high intensity is called "rapid" (R, 10^{12} el/s cm^2).

Processing of the experimental data showed that, in the p-Si samples after R-irradiation, the conductivity is due to the $E_v + 0.33$ eV level. The concentration of this level (7×10^{12} cm^{-3}) is approximately equal to the concentration of boron atoms, as determined by Hall-effect measurements before the sample was irradiated. S-irradiation of similar samples led to the level responsible for the slope of $E_v + 0.23$ eV with concentration of 1.4×10^{14} cm^{-3}, more than an order of magnitude higher than the concentration of boron atoms. Since photoconductivity measurements revealed a band close to the light quantum energy, it can be assumed that there are energy levels with the $n(t)$ curve slopes given by the Hall-effect measurements. In terms of the introduction rate of radiation defects (radiation defect concentration divided by irradiation dose), it can be argued that, at the same irradiation dose Φ, the introduction rate $\frac{N_2}{\Phi}$ of the $E_v + 0.33$ eV level with concentration N_2 is less than the introduction rate $\frac{N_1}{\Phi}$

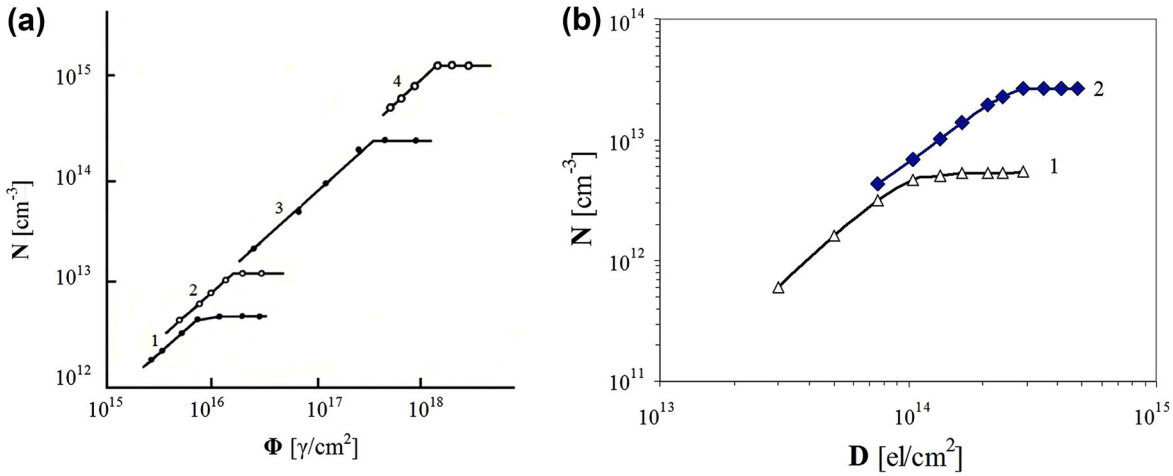

Fig. 5. Introduction of radiation defects in *n*-Si crystal depending on irradiation dose of (a) Co^{60} γ-quanta, and (b) picosecond pulse electrons with energy of 3.5 MeV. The irradiation and measurements were carried out at room temperature. Initial main charge carrier concentrations were (a): (1) $n_0 = 5 \times 10^{12}$ cm^{-3}, (2) $n_0 = 1.2 \times 10^{13}$ cm^{-3}, (3) $n_0 = 2.7 \times 10^{14}$ cm^{-3}, (4) $n_0 = 1.5 \times 10^{15}$ cm^{-3}; and (b): (1) $n_0 = 5 \times 10^{12}$ cm^{-3}, (2) $n_0 = 1.2 \times 10^{13}$ cm^{-3}.

of the $E_v + 0.23$ eV level with concentration N_1, that is

$$\frac{N_1}{\Phi} = \frac{1.4 \times 10^{14}\,\text{cm}^{-3}}{1.6 \times 10^{18}\,\text{cm}^{-2}} = 8.75 \times 10^{-5}\,\text{cm}^{-1}$$
$$\frac{N_2}{\Phi} = \frac{7 \times 10^{12}\,\text{cm}^{-3}}{1.6 \times 10^{18}\,\text{cm}^{-2}} = 4.45 \times 10^{-6}\,\text{cm}^{-1}. \quad (4)$$

Analysis of these introduction rates of radiation defects shows that, in *n*-Si, the prevalence of the introduction of A- and E-centers by irradiation is determined by the initial ratio of oxygen and phosphorus concentrations present in the sample.[7,8,18–20] When the oxygen concentration is less than 10^{16} cm^{-3} and the initial concentration of phosphorus is greater than 10^{16} cm^{-3} in *n*-Si, then E-centers are the main type of defects. If the oxygen concentration is higher than 10^{16} cm^{-3}, the main type of radiation defects are A-centers, which become electrically active as a result of irradiation. The situation with *p*-Si is different. The proportional decrease of λ_v with increase of n' (concentration of nonequilibrium free electrons) under irradiation is explained by the formation of a free vacancy on decay of a Frenkel pair capturing an electron, and by a decrease of the concentration of quasistationary electrons as a function of time as a result of the accumulation of radiation defects. Probably, this is the reason why no radiation transformation of *p*-Si into *n*-Si is observed, and as a result, the radiation hardness of *p*-Si is better than that of *n*-Si.

It can be concluded that this introduction rate of energy levels responsible for irradiation defects clearly determines the rate of introduction of radiation levels only if there is a linear dependence of the concentrations of these levels on the dose. When this dependence is absent or the radiation introduction rate depends on the radiation intensity φ at fixed dose Φ, there is no correspondence between the data for different Φ or φ values. Note that, for the deep levels at $E_v + 0.45$ eV in *p*-Si, the dependence of the concentration on dose is nonlinear. Therefore, the above-quoted values for the radiation introduction rates of the energy levels only apply for the corresponding values of Φ and φ but cannot be extrapolated to other doses or intensities. This fact should be borne in mind when comparing these results with those of other authors,[8–12,21–23] otherwise an apparent discrepancy may arise between the measured values determined for the same apparent radiation introduction rate.

The introduction rates of radiation defect levels for different charge carrier concentrations in *n*-Si as a function of irradiation dose are presented in Fig. 5. The steps shown in Fig. 5 indicate how effectively the radiation defects are introduced, remaining almost invariant with varying irradiation dose up to concentrations comparable to those of the impurities present in the structure of these defects, namely the initial concentrations of oxygen (A-centers) and phosphorus (E-centers) in the samples. Thus, the prevalence of A- and E-centers introduced by irradiation is governed by the initial ratio of the oxygen and phosphorus concentrations present in the sample.

DISCUSSION

As noted above, defects with levels at $E_v + 0.34$ eV, $E_v + 0.22$ eV, and $E_v + 0.28$ eV are considered by many authors to be the principal radiation defects in *p*-Si.[1–3,10] The level at $E_v + 0.34$ eV is observed from the slope of curves obtained from Hall-effect measurements or from the photoconductivity spectrum of oxygen-rich samples.

This has led many authors[4,7] to conclude that the corresponding defect center includes an oxygen atom. It should be noted, however, that the increase of any X center concentration on introduction of an impurity atom does not provide proof that the latter exists within it. Atoms, such as oxygen impurity (O), can act catalytically in the formation of X centers, without being part of their composition. The fact that introduction of oxygen atoms contributes to the appearance of the $E_v + 0.35$ eV level could be attributed, for example, to capture of free vacancies by oxygen atoms with subsequent release of an interstitial atom to form the level at $E_v + 0.35$ eV.

The existence of A-centers in p-Si after electron irradiation was reported previously,[5–7] where irradiation of oxygen-rich samples led to the appearance of an intense photoconduction band near the frequency corresponding to the transition of an electron from the valence band to the level at $E_v + 0.35$ eV.

In fact, carriers from the defect levels at 0.28 eV and 0.34 eV can interact. This is also obvious from the measurements of the spectral dependence of the photoconductivity in Fig. 3. In our opinion, measurements made at liquid-nitrogen temperature show that the bands of photoconductivity at 0.28 eV and 0.34 eV overlap, and a fine structure is noticeable in a wide spectral region from 3 μm to 4.5 μm, which is the aggregate band of the aforementioned dominating and other shallow bands. Many radiation defects are formed in the irradiated sample, creating shallow energy levels in the forbidden gap. These levels are ionized at 300 K and do not make a noticeable contribution to the photoconductivity of the sample; only carriers from deep potential wells of clusters are involved in this process. However, with decreasing temperature (at 80 K), these levels are filled, so when the sample is illuminated, they also become ionized.

It is well known that capture of oxygen atoms by free vacancies is quite effective in irradiated n-Si.[2–4,23,24] The $E_c - 0.17$ eV level, corresponding to the A-center (vacancy + oxygen atom), has been observed in n-Si by various research methods.[2,10,13–15] The existence of A-centers also in p-Si, irradiated by electrons, is verified by photoconductivity spectrum measurements. In these samples containing oxygen, radiation causes a photoconductivity intensity band to appear near the frequency corresponding to the transition of an electron from the top of the valence band to the $E_v + 0.34$ eV level. Apparently, there are a considerable number of other variants that may determine a quasichemical reaction in which oxygen atoms play a catalytic role and contribute to the formation of $E_v + 0.34$ eV. On the other hand, the following constant feature is observed in $E_v + 0.35$ eV level formation: its concentration never exceeds the initial boron atom concentration in the sample but can be equal to it. This has been observed for samples having significantly different numbers of boron atoms in their structure, with concentration from 7×10^{12} cm^{-3} to 1.6×10^{15} cm^{-3}. From these data, it may be deduced that there is one boron atom in the defect structure of the $E_v + 0.35$ eV level, and it is not likely for oxygen to be present in the structure of this center along with boron.

The $E_v + 0.28$ eV level is often interpreted as a divacancy formed in the irradiated silicon crystal. In previous work[11] it was shown that, in Si irradiated by electrons with energy of 1 MeV, there was significant anisotropy in the formation of radiation defects depending on the direction of electron impact with respect to the crystallographic axes of Si. This is explained by divacancy formation being an initial product of electron irradiation. However, we consider that the average electron energy is insufficient to form a divacancy as an initial radiation defect, which can only appear as a result of quasichemical reactions occurring from initial point radiation defects.[17–20]

CONCLUSIONS

The results of this study represent a different point of view regarding the nature of the energy levels responsible for the results of Hall-effect measurements and, hence, the introduction of radiation defects into silicon crystals by irradiation. The slopes of the curves from Hall-effect measurements, for example, a slope of 0.28 eV, were investigated in detail; the latter may be due to the presence of two levels at $E_v + 0.22$ eV and $E_v + 0.34$ eV. In support of this view, we note that there is no corresponding band in the photoconductivity spectrum that could be linked to the $E_v + 0.28$ eV level. The nearest photoconductivity peak on the short-wavelength side ($E_v + 0.32$ eV) is bell-shaped, indicating that the photoconductivity is associated with the transition of a hole, which is related to the excited level of the center, followed by thermal excitation to the zone. The energy of the hole in the ground state is greater (by kT) than the maximum value 0.34 eV of the photon excitation energy. The good radiation hardness of p-Si with respect to n-Si is also explained.

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CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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