EFFECT OF COMPENSATION DEGREE AND CONCENTRATION OF IMPURITY ELECTROACTIVE SELENIUM ATOMS ON CURRENT AUTO-OSCILLATION PARAMETERS IN SILICON

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One of the crucial phenomena is auto-oscillations of current in elementary and binary ($A^{II}B^{V}$, $A^{II}B^{VI}$) semiconductor materials, which allow the creation of solid-state oscillators with a wide frequency range from 10⁻³ to 10⁻⁶ Hz. In this paper, we show the results of a study on the effect of the degree of compensation (K) and the concentration of electroactive impurity selenium atoms on the excitation conditions and parameters (amplitude, frequency) of the auto-oscillation current associated with temperature and electrical instability in silicon. In the research, silicon doped with selenium atoms Si<Se> of identical geometrical dimensions has been used. The compensation degree of the initial boron atoms with impurity selenium atoms in the samples is in the range of K = $2N_B/N_{Se} = 0.94-1.1$. It was found that excitation conditions, the amplitude and frequency of auto-oscillation current significantly vary depending on the degree of compensation of selenium atoms with boron atoms in the initial silicon. Obtained experimental results showed that the autooscillation current in silicon doped with impurity selenium atoms is characterized by ease of control with stable parameters (amplitude and frequency), which makes it possible based on this unique physical phenomenon to develop and create oscillatory circuits in information technology.

Keywords: Selenium; Diffusion; Boron; Amplitude; Frequency; Illumination; Concentration; Auto-oscillation; Silicon; Conduction band; Valence band

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INTRODUCTION

The detected auto-oscillations of current in semiconductors, from the scientific and applied point of view, are one of the most striking and promising physical effects enabling the development of a new scientific direction of nonequilibrium thermodynamic effects in solids and their application to electronics [1-4].

A literature review has presented that there has not been sufficiently studied research on the physical mechanisms of current auto-oscillation excitation and the regularities of parameter changes kinetics (such as amplitude and frequency) and the physics of transients for different types of current auto-oscillations within a same semiconductor material [1-4]. Furthermore, there is a lack of reliable theoretical and experimental data regarding the thermodynamic conditions necessary for the existence of an auto-oscillating environment that could serve as a source of regular, stable, reproducible current auto-oscillations with controllable parameters [5-7].

In the applied aspect, the auto-oscillations of current detected in semiconductors have significant potential, particularly in the development of new functional electronic devices (solid-state generators, memory and storage, recording and transmission information, optoelectronic receivers, etc.) and the possibility of a fundamentally new generation of sensors for physical quantities that utilize an amplitude-frequency output [8-10].

The successful resolution of the scientific and practical challenges associated with auto-oscillation processes requires the implementation of these processes within specially created vibrational media based on semiconductor materials that are in a highly non-equilibrium thermodynamic state. An auto-oscillating environment is one in which each physical small element in it should have potential auto-oscillation properties, and these elements are could be connected through transfer processes, such that the excited oscillation propagates through the volume of the material being investigated [12,13]. Therefore, it requires to investigate the physical properties of the thermodynamic conditions for the existence of autooscillating environments in semiconductors with highly non-equilibrium thermodynamic states, and to determine the technological feasibility of creating such environments. The analysis of the creation of auto-oscillating environments highlights the importance of studying auto-oscillation processes from both scientific and applied perspectives.

It was found that the existence of deep energy levels of impurity atoms, which are the main requirement for current auto-oscillation excitation in semiconductors and semiconductor structures, is necessary for the observation of current auto-oscillation.

When conducting a literature survey [5-12] on the production of silicon doped with impurity atoms, it was discovered that there are several experimental data points that do not fit into the framework of the current theory of deep levels. These anomalies have been identified and include:

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- the presence of sufficiently many energy levels of the same impurities in silicon which cannot be easily explained by the electronic structure or their position in the crystal lattice.

- an unstable state of deep centers where with time or rising temperature new energy levels with different characteristics appear.

- energy level bands with different widths detected for many impurity atoms in silicon cannot be explained using existing theory and no data on energy level distribution inside the bands are available so far.

- for many impurity atoms energy levels with the same values in the band gap of silicon were found.

The above-mentioned data can't be explained by the existing theory of deep-level physics [13,14]. Scientific results which have been obtained by investigation of auto-oscillations of current in silicon diffusively doped with impurity selenium atoms give ground for the creation of a new theory of deep levels.

Researches on the impact of the concentration of electroactive transition group atoms in silicon are important for understanding the nature of deep centers created by these elements in the semiconductor and expanding their use as a new material for semiconductor electronics [11–19]. This study focuses on the effect of the initial concentration of small boron impurity atoms on the concentration of electroactive selenium atoms that create deep energy levels in silicon. The results show that changing the concentration of boron impurity atoms in initial silicon within the range of $N_B = 1.4 \cdot 10^{14} - 3.1 \cdot 10^{16}$ cm⁻³ increases the concentration of electroactive selenium atoms by two orders. However, the concentration of electroactive selenium atoms by two orders. However, the concentration of electroactive selenium atoms by two orders, which create deep energy levels, depends significantly on the type and concentration of the impurity (boron or phosphorus) in the initial silicon. Furthermore, it was found that these impurity atoms interact not only with the basic atoms of silicon and atoms of the initial impurity but also with other uncontrolled impurities and defects in silicon.

METHODS

To develop a reproducible technology for producing compensated silicon doped with selenium atoms, we determined the temperature regimes for obtaining samples with the same resistivity and type of conductivity at different concentrations of electroactive impurity atoms based on those described in [22-24]. These results made it possible to clearly define the technological conditions to produce compensated silicon with the maximum concentration of electroactive impurity atoms of selenium. The developed reproducible technology for the production of compensated silicon with given electrophysical parameters allows to control the electrophysical parameters of the material not only by temperature and time of diffusion annealing but also by the choice of concentration of initial impurity boron atoms in silicon (Table 1).

No	The elasticity of selenium	Type of conductivity of samples	Specific resistance,	Carrier concentration, cm ⁻³	
	atom vapor pressure, atm	obtained after diffusion	Ohm∙cm		
1	0.1	р	18	$1.2 \cdot 10^{15}$	
2	0.3	р	60	$3.5 \cdot 10^{14}$	
3	0.5	р	$3.1 \cdot 10^2$	6.7·10 ¹³	
4	0.75	р	$4.7 \cdot 10^3$	$4.5 \cdot 10^{12}$	
5	1.0	р	6.8·10 ⁴	3.1.1011	
6	1.5	р	$1.25 \cdot 10^{5}$	1.7.1011	
7	2.0	n	$1.1 \cdot 10^{5}$	$6.7 \cdot 10^{10}$	
8	2.5	n	$8.3 \cdot 10^4$	8.2·10 ¹⁰	
9	3.0	n	$2.6 \cdot 10^4$	2.3.1011	
10	4.0	n	5.9·10 ³	8.8.1011	
11	Initial sample KDB-10	р	10	$2 \cdot 10^{15}$	

Table 1. Possibility to control the electrophysical parameters of the material based on the developed technology

The analysis of obtained sample after diffusion showed that impurity selenium atoms strongly destroy the surface of silicon, leading to surface erosion and even complete destruction of the crystal. Therefore, when diffusing selenium atoms into silicon, the main diffusion parameters should be taken into account. One of the main diffusion parameters is the vapor pressure of the diffusant when diffusion occurs at high temperatures. To overcome these difficulties, the task was set to accomplish diffusion at different vapor pressures of diffusant selenium from 0.5 atm to 2 atm.

According to the research that was conducted, it was showed that when the selenium vapor pressure is between 0.5 and 1 atmosphere, there is insignificant erosion observed on the surface of silicon. Further entrainment of diffusant vapor pressure leads to surface erosion and further silicon destruction. Thus, the optimum diffusant vapor pressure for the diffusion of silicon by impurity selenium atoms was established. It was also found that the surface erosion depends on the quality of initial silicon surface treatment - with improvement of silicon surface treatment (in this case, the quality of polishing was changed using White's chemical etchant), the erosion rate was decreased [25]. Thus, it was determined that to reduce surface erosion during diffusion by selenium, it is necessary to treat the surface carefully and to polish the surface of the initial silicon samples.

EXPERIMENTAL PART

For the investigation, initial monocrystalline silicon of p-type with resistivity values of 1, 2, 10, and 100 Ohm·cm was used. The initial silicon samples had boron impurity concentrations (N_B) ranging from $1.4 \cdot 10^{14}$ to $3.1 \cdot 10^{16}$ cm⁻³. These

initial silicon materials were obtained using Czochralski's method, and the oxygen concentration in them was practically the same and in the order of $N_{O2} = (5-7) \cdot 10^{17}$ cm⁻³. To conduct the research, 50 samples of the same geometric dimensions were produced from each of the initial materials. For the study, 50 pieces of samples with the same geometric dimensions were made from each starting material. Diffusion of selenium was carried out in a "Magnetic" diffusion electric furnace from the gas phase in the same conditions (temperature, diffusion time, diffusant vapor pressure, and cooling rate) in evacuated quartz ampoules to P =10⁻⁶ mm Hg. At the same time, two samples from each initial material were placed in each quartz ampoule to ensure the same doping conditions. The experiment was repeated 5 times. Each time, the initial samples were annealed under the same conditions without selenium impurities to assess the influence of selenium impurities in the diffusion process on the electrophysical properties of the initial silicon.

THEORETICAL CALCULATIONS AND RESEARCH RESULTS

Measurements conducted on electrophysical properties of samples derived from KDB silicon after the diffusion of selenium atoms revealed that the concentration of electroactive selenium atoms varied considerably depending on the initial boron concentration in silicon, despite the identical doping conditions of temperature, diffusion time, and cooling rate. Electrophysical parameters of silicon samples after diffusion were measured based on the Hall effect method using Ecopia HMS-3000 Hall Measurement System. The electroneutrality equation was used to determine the overall concentration of electroactive selenium atoms in the samples obtained after diffusion. During computations, we considered sample parameters that accounted for the extent of filling of both energy levels of selenium ($E_1 = E_C-0.24 \text{ eV}$; $E_2 = E_C-0.5 \text{ eV}$) within the silicon band gap [26,27].

Selenium concentration was theoretically determined by the following formula for compensated samples p-Si<B,Se>:

$$N_{Se} = 1/2 \cdot (p_0 - p - N_{TD}), \tag{1}$$

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For overcompensated n-Si samples <B, Se> was determined:

$$N_{Se} = (p_0 + n_0 + f(E_1) \cdot N_{Se} + f(E_2) \cdot N_{Se} - N_{TD}),$$
(2)

$$f(E_2) = N_{Se}/2 \cdot (1 + A \cdot exp(-F - E_2)/k \cdot T),$$
 (3)

$$F=k\cdot T \ln(N_C/n), \tag{4}$$

$$N_{Se} = 1/2 \cdot (p_0 - p - N_{TD}), \tag{5}$$

where p_0 and p are concentrations of holes before and after diffusion of selenium atoms, respectively. N_{TD} is concentration of thermodonors, $f(E_1)$ and $f(E_2)$ are degrees of filling of the 1st and 2nd deep energy levels of selenium, respectively. n_0 and n and electron concentrations before and after diffusion of selenium atoms accordingly, respectively. The values p_0 and p were determined from Hall measurements of samples before and after diffusion. N_{TD} is determined by measuring carriers' concentration before and after diffusion in control (undoped) samples, heat-treated under the same diffusion technological conditions in the temperature range T = 1100-1250°C in time interval t = 5-10 hours. Also, the solubility values of impurity selenium atoms in silicon at a given diffusion temperature were de-fined.

To determine the value of the ionization energy of impurity selenium atoms in silicon, the infrared (IR) and temperature quenching (TQ) photoconductivity of silicon samples doped with impurity selenium atoms were selected with the same resistivity, conductivity type, and geometric dimensions. The concentration of electrically active selenium atoms in the samples studied differed by about 30 times.



Figure 1. Spectral dependence of photocurrent in Si<Se> samples

 $1 - \rho = 8.7 \cdot 10^4 \text{ Ohm cm (initial KDB-1)}; \text{ N}_{\text{Se}} \approx 3.1 \cdot 10^{16} \text{ cm}^{-3}; 2 - \rho = 8 \cdot 10^4 \text{ Ohm cm (initial KDB-10)} \text{ N}_{\text{Se}} \approx 2 \cdot 10^{15} \text{ cm}^{-3}; 3 - \rho = 8 \cdot 10^4 \text{ Ohm cm (source KDB-100)} \text{ N}_{\text{Se}} \approx 1.4 \cdot 10^{14} \text{ cm}^{-3} \text{ at temperature T} = 80 \text{ K}, \text{ E} = 20 \text{ V/cm}.$

Figure 1 illustrates the spectral dependence of photoconductivity under the same background photocurrent value with different concentrations of selenium atoms as electroactive impurities. In Figure 1, the maximum depth of IR extinction of a photocurrent is observed in the samples where the concentration of electroactive selenium atoms is $N_{Se} = 3.1 \cdot 10^{16}$ cm⁻³ and reaches 5 orders of magnitude. It is established that with decreasing concentration of electroactive impurity atoms, the quenching multiplicity decreases, and the energy of the corresponding quenching maximum shifts to high values.

It is shown that with increasing concentration of electroactive selenium atoms, the quenching interval decreases, i.e., photocurrent decreases more sharply, and the energy interval of incident quanta in which IR quenching of photocurrent is observed narrows.

An anomalous course is observed in silicon doped with impurity selenium atoms when studying the temperature dependence of photoconductivity quenching (TDPQ) in silicon doped with selenium atoms. As seen from Figure 2, when a temperature rises from T = 80 K to T = 140 K, the photocurrent does not change essentially. In the temperature range T = 140-180 K, its de-crease is observed, i.e., there is a temperature quenching of photoconductivity well-known in the literature [28,29].

The results of TDPQ studies in compensated silicon samples doped with selenium atoms (Se) have shown that the onset and depth of quenching strongly depend on the background illumination intensity, compensation degree, and concentration of electroactive impurity atoms (Figure 2).



Figure 2. Temperature quenching of photoconductivity at different background illuminations in Si<Se> with $\rho = 8 \cdot 10^4$ Ohm·cm, E = 40 V/cm, 1 - 25 lux, 2 - 10 lux, 3 - 5 lux, 4 - 1 lux, 5 - 0.1 lux.

It is feasible to observe TDPQ over a broad temperature range by adjusting the degree of compensation, background illumination, and photocurrent value in compensated silicon. The obtained data indicate that the depth, rate, and area of both IR and temperature quenching of photoconductivity in silicon samples containing impurity selenium atoms can be controlled by adjusting the concentration of these electroactive impurity atoms. From the results of the analysis, it was found that the energy levels of impurity selenium atoms with the ionization energy Si<Se> – E_C – 0,5 eV, which is associated with the double positive ionization of selenium atoms, are responsible for the IR and TQ photoconductivity in silicon.

The results showed that regardless of the compensation degree of boron atoms in the initial silicon doped with impurity selenium atoms, the current auto-oscillation was excited at a certain integral intensity interval (I_{min} – Imax). With the growth of compensation degree value (resistivity), at the same value of threshold field strength E_{th} at which the auto-oscillation of current is observed, the I_{min} intensity value decreased, and the interval between I_{min} – I_{max} widened (Figure 3). Thus, it was found that conditions of auto-oscillation excitation in silicon samples with different compensation degrees (resistivity) have an identical character. It was also found that the amplitude and frequency of current auto-oscillation, in their turn, strongly depend on excitation conditions.

In this work, we focused on determining the parameters of silicon samples doped with impurity selenium atoms with the most optimal characteristics (forms) of current auto-oscillation. Based on the analysis of the findings, we have determined that the silicon p–Si<Se> - samples with a resistivity of $\rho = 8 \cdot 10^4$ Ohm cm is appropriate for studying current auto-oscillations with reproducible and preset parameters from the compensation degree, which was approximately equal to 0.999. When an electric current intensity of E = 200 V/cm was applied to these samples, the maximum and minimum values of current auto-oscillation amplitude varied within the range of $I_{min} = 10^{-8}$ A to Imax = 2.5 $\cdot 10^{-1}$ A, while the frequency was within the interval of f = $10^{-3} - 10$ Hz.

To elucidate the influence of electroactive concentration of impurity selenium atoms on parameters of current autooscillations, we received samples with the same size and degree of compensation K = 0.999. Thus, the concentration of electroactive selenium atoms was in the interval $N_{Se} = (2-31) \cdot 10^{15}$ cm⁻³ and with increasing of electroactive selenium atoms concentration N_{Se} value of E_{th} and amplitude of auto-oscillations was decreasing and the frequency was increased (Table 2).



Figure 3. The dependence of changing of threshold field E_{th} on integral light intensity for Si<Se> samples with different resistivities (ρ). $1 - 5.3 \cdot 10^3$; $2 - 8.7 \cdot 10^3$; $3 - 3.2 \cdot 10^4$; $4 - 5.8 \cdot 10^4$; $5 - 9.1 \cdot 10^4$; $6 - 2.1 \cdot 10^5$ Ohm cm p-type; at T = 80 K, E = 200 V/cm.

Table 2. Effects of electroactive concentrations	of impurity	v selenium ator	ns in silicon (on auto-oscillation current	parameters
	1 /				1

Paragraph number	The concentration of electroactive	Threshold field E _{th} , V/cm	Amplitude J, mA	Frequency f, Hz	Note
	selenium atoms				
	$N_{Se} = 10^{15} cm^{-3}$				
1	2	250	90	5·10 ⁻²	T = 80 K,
2	8	276	84	9.4·10 ⁻²	$I = 2.4 \cdot 10^{-6} \text{ W/cm}^2 \cdot \text{s}$
3	31	315	78	2.5.10-1	at $hv = 1.1 eV$, with $K = 0.999$

DISCUSSION

It has been confirmed that the auto-oscillation of current in silicon samples doped with selenium atoms is observed when the value of compensation degree (K) is in the interval between 0.94 and 1.1, i.e. in crystals, where the Fermi level is within the compensation degree range from $E_C - 0.45$ to $E_V + 0.35$ eV. It is clear that in this energy interval in the band gap of silicon, there is only one donor level of double ionized impurity selenium atoms with ionization energy $E_C - 0.5$ eV which is connected with the presence of clusters consisting of selenium atoms Se_4 and Se_6 [30]. The presence of clusters of selenium atoms consisting of twice positively ionized atoms leads to the appearance of strongly charged centers which allows the obtained silicon samples Si<Se> to be regarded as inhomogeneous material. Inhomogeneous material for which the barrier model of inhomogeneity proposed by authors of books [31,32] is applicable (Figure 3).

According to barrier model, fluctuations of conduction and valence band topography arise in the semiconductor. The appearance of such inhomogeneities in semiconductors is attributed to the presence of impurity atoms, which can reach concentrations of $N \approx 10^{18} - 10^{19}$ cm⁻³ under strong doping conditions. Due to their inhomogeneous distribution, these impurities cause fluctuations in the conduction and valence bands, resulting in inhomogeneities. In our situation, the concentration of impurity selenium atoms is two orders of magnitude lower ($N_{Se} = 3.1 \cdot 10^{16}$ cm⁻³) than under strong doping conditions. It was shown that this impurity in silicon may be in the binary ionized state and form different complexes as Se₄ and Se₆ molecular compounds with charge always greater than ±2 (n – number of selenium atoms in the cluster). The Dabaev screening radius of such complexes overlaps with each other and results in a fluctuating potential relief in compensated silicon. To explain the physical mechanism of the low-frequency current auto-oscillations observed in compensated silicon, a model based on conduction and valence band relief fluctuations is shown in Figure 4. According to the proposed model, at a constant illumination nonequilibrium electron fill the selenium level, while holes are localized in the potential wells of the valence band. In this case, the conductivity is determined by the concentration of holes at the leakage level, the value of which is determined by the value of the drift barrier.

Based on this model, it is possible to imagine that the excitation of current auto-oscillation occurs through the following mechanism. When Si<Se> samples are illuminated with energy greater than the band gap width of silicon at relatively low temperatures (T = 80-160 K), non-equilibrium carriers - electrons and holes, which are located on energy barriers of conduction and valence bands are generated. Electrons are captured by clusters of doubly positively ionized selenium atoms, while holes are accumulated in energy minima of the valence band. Some of the holes, upon hitting the leakage level, participate in conduction, leading to an increase in photocurrent. Photocurrents are excited when the photocurrent due to holes at the valence leakage level is sufficient for Joule heating of the crystal. When a hole is heated, it acquires sufficient energy to overcome the recombination barrier and recombine with electrons trapped on clusters consisting of impurity selenium atoms. As a result, the current in the circuit and the temperature of the sample decreases, thus repeating the process periodically.



Figure 4. Barrier model of compensated silicon doped with impurity selenium atoms.

 \circ -electrons, \bullet -holes, E_g-silicon band gap width, E_C- conduction band, E_V-valence band, E_{ef} - effective band gap width of inhomogeneous silicon, E_l - is the leakage level for the charge carriers (in this case the hole), E_{rec} - recombination energy.

The excitation conditions and current auto-oscillation parameters change depending on the compensation degree and can be interpreted based on the change of potential relief amplitude. The strongly compensated samples Si<Se> with a compensation degree of K \approx 1 exhibit the highest potential relief values of heterogeneities. Therefore, the most effective separation of photoexcited charge carriers, electrons in conduction band cavities, and accumulation of holes in energy minima of the valence band occurs exactly in these crystals. This explains the maximum amplitude and decrease of the frequency of the current auto-oscillation in Si<Se> samples with resistivity $\rho = 8 \cdot 10^4$ Ohm·cm. In Si<Se> samples, both at n- (K>1) and p- (K<1), the amplitude of formed potential relief due to material heterogeneity decreases due to the charge state decrease of selenium clusters and Debye screening of heterogeneity by uncompensated holes. Thus, the efficiency of separating carriers decreases, which is accompanied by the decrease of hole concentration participating in conductivity. In n-Si<Se> samples, the potential field of clusters could be compensated by increasing of field or illumination intensity, and in p-Si<Se> samples, the barriers are compensated by equilibrium holes.

The observed features of excitation conditions and current auto-oscillation parameters depending on the concentration of electroactive selenium atoms are related to the concentration of multipolarized clusters of impurity selenium atoms.

CONCLUSIONS

We conclude that the temperature quenching effect of photoconductivity in silicon diffusively doped with impurity selenium atoms is responsible for the excitation of low-frequency current oscillations. The technology developed for producing silicon doped with impurity selenium atoms with reproducible and predetermined electro-physical parameters enables the creation of sensors for physical quantities such as temperature, illumination, electric and magnetic field, and pressure with amplitude-frequency output and generators of electromagnetic oscillations based on the obtained experimental results.

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ВПЛИВ СТУПЕНЯ КОМПЕНСАЦІЇ ТА КОНЦЕНТРАЦІЇ ДОМІШКОВИХ ЕЛЕКТРОАКТИВНИХ АТОМІВ СЕЛЕНУ НА ПАРАМЕТРИ АВТОКОЛИВАНЬ СТРУМУ В КРЕМНІЇ

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Одним із вирішальних явищ є автоколивання струму в елементарних і бінарних (A^{IIBV} , A^{IIBVI}) напівпровідникових матеріалах, які дозволяють створювати твердотільні осцилятори з широким діапазоном частот від 10⁻³ до 10⁻⁶ Гц. У даній роботі наведено результати дослідження впливу ступеня компенсації (K) і концентрації електроактивних домішкових атомів селену на умови збудження та параметри (амплітуду, частоту) автоколивального струму, пов'язані з температурою. і електрична нестабільність у кремнії. У дослідженнях використовувався кремній, легований атомами селену Si<Se> однакових геометричних розмірів. Ступінь компенсації вихідних атомів бору домішковими атомами селену в зразках знаходиться в межах K = $2N_B/N_{Se} = 0,94-1,1$. Встановлено, що умови збудження, амплітуда та частота струму автоколивань суттєво змінюються залежно від ступеня компенсації атомів селену атомами бору у вихідному кремнії. Отримані експериментальні результати показали, що автоколивальний струм у кремнії, легованому домішковими атомами селену, характеризується простотою керування зі стабільними параметрами (амплітуда та частота), що дає змогу на основі цього унікального фізичного явища розробляти та створювати коливальні контури в інформаційних технологіях.

Ключові слова: селен; дифузія; бор; амплітуда; частота; освітлення; концентрація; автоколивання; кремній; зона провідності; валентна зона