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Introductory Chapter: Description of the Real Monocrystalline Structure on the Basis of the Vlasov Model for Solids

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1. Introduction

As well known, all characteristics of initial materials for following applications depend on the crystal structural perfection. In the last few decades, our knowledge of the nature of the solid state has sharply increased. At the same time, the areas of application of crystalline solids in various fields of technology are increased. The needs of technology stimulate the rapid development of a relatively young area of modern natural science—solid state chemistry. The main tasks of solid state chemistry are (i) the synthesis of solids, (ii) the research of their physico-chemical properties and reactions with their participation, and ultimately (iii) the creation of materials with predetermined properties.

All real solids contain structural defects. Structural defects are violations of the periodicity of the spatial arrangement of atoms. The impact of defects on the physical properties of crystals is extremely diverse. The introduction or change in the concentration of defects in a solid can change the physical properties of the matter. Such operations lead to the creation of new optical, electronic, mechanical, and other characteristics of the solids. Creating a fairly complete picture of the nature and behavior of various defects is a prerequisite for a scientific approach to control structurally sensitive properties and processes in solids. Therefore, in the research of solids, the most informative is the way of a joint examination of their structure, formation, and properties.

Of all the variety of solids, semiconductor silicon occupies a completely unique position. Semiconductor (or electronic) silicon is the basis of the electronic industry at present and in the foreseeable future. On the basis of silicon, discrete device and integrated circuits are manufactured. For the requirements of microelectronics, it is necessary to grow single crystals (i) in which there are no dislocations completely, (ii) with a uniform distribution of doping and

background impurities, and (iii) with a controlled and limited content of intrinsic point defects. Stringent requirements stimulate intensive research into the nature of defect formation and improvement of methods for obtaining modern dislocation-free single crystals of silicon.

Requirements for the quality of modern microelectronic devices have led to the fact that silicon is the most pure material in the world. To obtain single crystals of silicon, the Czochralski method and the floating zone method are used. Both methods ensure the production of the initial silicon with the total content of residual impurities 10^{11} to 10^{12} cm^{-3} . Semiconductor silicon (a highly pure material with the perfect structure) can be considered as the initial model for the construction of theoretical models of defect formation in other semiconductor materials and metals. The obtained mathematical models and methods proposed for their solution in silicon make it possible to formulate and solve many problems on the kinetics of diffusion processes in solids.

The growth of dislocation-free silicon single crystals by the Czochralski method and floating zone melting is accompanied by the formation of structural imperfections known as grown-in microdefects. The formation of grown-in microdefects is caused by complex processes of interactions between intrinsic point defects (vacancies and interstitial silicon atoms) and impurity atoms of oxygen and carbon in cooling silicon ingots. Depending on the thermal conditions of crystal growth, grown-in microdefects are precipitates of impurities, microvoids, or interstitial dislocation loops [1]. The presence of grown-in microdefects in dislocation-free silicon single crystals significantly affects their mechanical, electrical, optical, and other properties. The problem of controlling the nature, content, size, and nature of the distribution of defects present in the dislocation-free silicon single crystal is primarily related to the development of effective methods for influencing the state of the ensemble of interacting point defects in the grown ingot. Advances in the control of the condition of the ensemble of point defects determine, ultimately, the success of the quality management of electronic devices. The problem of the formation of grown-in microdefects has both the technological aspect (control of the defect structure of the crystal during the growth for the purpose to increase its quality) and the fundamental scientific importance because its solution makes it possible to describe the physics of defect formation in ultra-pure dislocation-free silicon single crystals. The construction of a theoretical model of the formation of grown-in microdefects in dislocation-free silicon single crystals is an important scientific and practical problem from the viewpoint of the development of physical principles of the purposeful creation of new materials with a desired set of physical properties.

In 2010, we created a model of high-temperature precipitation of impurities [2]. In this model, the inclusion of thermal growth conditions of silicon single crystals in the form of the dependence $T(t)$ (where T is temperature, t is time) allows us to theoretically describe the conditions for nucleation, growth, and coalescence of the precipitates in the range from the crystallization temperature to room temperature. The model of high-temperature precipitation is not only adequate to the experimental results of the investigation of grown-in microdefects but also does not contradict the classical theory of nucleation of second-phase particles. This model extends the field of application of the mathematical apparatus of the classical nucleation theory to the formation of nuclei in a solid during its cooling after the growth. The introduction and consideration of thermal conditions of crystal growth in the initial equations of the classical nucleation theory make it possible to demonstrate that the formation of critical nuclei of small

sizes, with a higher probability, occurs in the vicinity of the crystallization front. Cooling of the crystal is accompanied by the growth and coalescence of precipitates. The interaction of impurities with intrinsic point defects has a fundamental character and determines the defect structure of the crystal from the onset of the crystallization to the fabrication of the finished device [3].

In general, in the last century to describe the physical properties of solids with a point of view of their atomic structure, two approaches were suggested. The classical approach posits the notion of the crystal lattice [4]. The classic approach assumes (i) the location of each atom in the vicinity of the fixed lattice site, (ii) the consistency of introducing the concept of probability and of the mechanical description of the behavior of particles, and (iii) the premise that the totality of atoms in a crystal is an integer. Statistical and quantum mechanical methods are used in the classical theory of the crystal. This theory has resulted in important results. In particular, one of such results is the creation of the classical theory of nucleation and growth of second-phase particles in solids [5]. With the help of this theory, we can describe the formation and development (transformation) of the defect structure of a solid, both in the process of its growth and as a result of various technological impacts [3].

In the second approach, a periodic structure of the crystal is not a consequence restriction on the freedom of movement of the atoms in the crystal. Periodic structure is caused by the specificity of statistical laws of motion of particles, where a periodic structure agrees with the freedom of movement of atoms. As a result, the probability of finding an atom in interstitials of the crystal lattice is always different from zero [6, 7]. The second approach is based on the solution of the Vlasov kinetic equation. Vlasov equation represents a system of equations that describe the evolution of the particle continuum with potential of the pair interaction [8]. Vlasov created the physical and mathematical apparatus of system of many particles. He has developed and entered into the reference the concept of collective variables. Vlasov assumed that the method used by him to solve the problem of the plasma has a universal character. This method can be applied to the description of plasma, gas, liquids, and solids. The interaction in those systems can be a short range or long range, weak or strong [9, 10].

For the first time, a solution for solid state was conducted by Vlasov. Vlasov showed that the spatial periodic distribution is one of the particular conditions of particle motion [6, 7]. Vlasov in particular investigated using the linearized Vlasov equation and found the criteria for the origin of the periodic structure in terms of the temperature, density, and microscopic interaction of particles of the medium.

It is noted that in his 1946 paper [11], Lev Landau wrote a very sharp remark: *“these equations were used by A.A. Vlasov for investigation of the vibrations of plasma. However most of his results are incorrect.”* In Ginzburg et al. [12], which is not published in the English version, Lev Landau, Vitalyi Ginzburg, and others were even tougher: *“Recently (in 1944-1945) in print appeared a number of works by AA Vlasov, dedicated to generalization of the concept of the electron plasma and solid state theory. In these works, the author concludes that in the self-consistent field method accounting of the interaction forces at large distances reveals new dynamic properties of polyatomic systems and leads to a change in our perceptions of gas, liquid, solid in the direction of combining them with the concept of plasma etc. Consideration of these works Vlasov led us, however, to the conclusion about their complete failure and about the absence of the results of scientific value.”*

In the field of plasma physics, history has judged critics and Vlasov [13]. However, in his 2000 paper [14] Vitalyi Ginzburg wrote: *"I do not know about any Vlasov achievements in solid state theory and the general theory of many particles (outside of plasma physics)."* Indeed, the Vlasov model for a solid was used only for ideal crystals. Hitherto, this model for solid state for the description of the real structure of the crystal has not been used.

Only in 2016 in the journal "Physics of the Solid State," we have considered the complex formation in the semiconductor silicon in accordance with the Vlasov model for solid state [15]. It was demonstrated that the Vlasov model for solid state can be applied not only for the research of the hypothetical ideal crystals but also for the description of the formation of structure of real crystal. In this article, the formation of silicon-carbon and silicon-oxygen complexes during cooling after the growth of dislocation-free silicon single crystals has been calculated using the Vlasov model of crystal formation. It has been confirmed that the complex formation begins in the vicinity of the crystallization front. The characteristic numbers of complexes (silicon-oxygen and silicon-carbon) were determined.

2. Mathematical model

Vlasov created a physical and mathematical apparatus of many-particle system, developed and introduced the concept of collective variables into circulation. The primary concept of Vlasov physics is the distribution function. The particle is characterized by spectrum of geometric and kinematic properties. Thus, the category of motion is included on the same level with the primacy of categories of space and time. Therefore, in Vlasov model for solid state, the periodic distribution of probability density of the particles is the state (motion) of the particles of the system, not the design.

The Vlasov model for solid state is based on the following fundamental physical positions [6, 10]: (1) rejection of the principle of spatial and speed localization of the particles (in terms of classical mechanics), which takes place regardless of the force interactions; (2) introduction of force interactions in analogy with classical mechanics, but taking into account the new principle of non-localization of particles; (3) the behavior of each particle of the system is described by means of an extended phase space f -function. In this approach, the ideas of continuity and of corpuscular are combined. The method of describing the motion of particle associated with the extended function, and the particle in the form of a point occurs only in the particular case [6].

In general, the equation of Vlasov describes the evolution of the function of distribution $f(x, v, t)$ of the continuum of interacting particles in Euclidean space for speed v and for coordinate x at time point t . The equation has the form

$$\frac{\partial f}{\partial t} + \left(\frac{\partial f}{\partial x}, v \right) + \left(\frac{\partial f}{\partial v}, F \right) = 0 \quad (1)$$

$$F = -\frac{\partial}{\partial x} \int K(x, y) f(y, v, t) dv dy$$

where K is the pair interaction potential, which is in real problem depends on the distance $|x - y|$; F is the total force, with which all the particles act on one of them, which is located at time t at point x [8]. In order for distinguishing between the types of interactions, the systems of equations of Vlasov (equations of the Vlasov-Poisson, Vlasov-Maxwell, Vlasov-Einstein, and Vlasov-Yang-Mills [16]) are usually discussed about.

For a description of stationary properties of the crystal, the concept of distribution density of particles $\rho(r) = \int f(r, v) dv$ is used. The molecular field is determined only by the probable locations of atoms that are described by the potential function. The potential function contains the density of probability of the particles considering the temperature distribution of the particles [6]. The choice of potential of the pair interaction depends on the problem under consideration. Nonlocal model of a crystal is described by nonlinear equations. These equations make it possible to calculate the molecular potential and density of location of particles in the conditions of temperature equilibrium [6]:

$$V(r) = \lambda kT \int_{-\infty}^{\infty} K_{1,2}(r) \exp\left(-\frac{K_{1,2}(r)}{kT}\right) dr \quad (2)$$

$$\rho(r) = \lambda kT \exp\left(-\frac{K_{1,2}(r)}{kT}\right)$$

where k is the Boltzmann constant, $K_{1,2}$ is the potential of the pair interaction, λ is some characteristic number, and T is the temperature. The initial equations are equations for the two particles in a stationary condition $\left(\frac{\partial}{\partial t} = 0\right)$ [6]. Under the characteristic number where the values of a parameter λ are to be understood, Eq. (2) has solutions different from the trivial [6]. If the position of one of the particles is taken as the origin of coordinates, it is possible to determine $\rho(0) = \lambda kT$ [6]. The important task of Vlasov model for the solid state is the determination of characteristic numbers.

The characteristic number λ is determined from the basic criterion of the existence of the crystal state. The conditions of crystallization can be written as follows:

$$\frac{4\pi N}{kT_m} \int_0^{\infty} K_{1,2}^*(\rho) \rho^2 d\rho = 1 \quad (3)$$

where N is the number of particles, T_m is the temperature of melting (crystallization) of crystal, $K_{1,2}^* = -K_{1,2}$ [6].

Eq. (2) is written for the conditions of thermal equilibrium of system. The minima of interatomic potential correspond to the stable equilibrium position of atoms in the complexes

(silicon-oxygen and silicon-carbon). Then, we can determine the density of the distribution of complexes as a function of the cooling temperature of the crystal

$$\rho(T) = \lambda kT \exp\left(-\frac{V_{1min,2min}}{kT}\right) \quad (4)$$

The formation of new phase nuclei takes place near the crystallization front of the crystal [2]. In article [2], the model of high-temperature precipitation of impurities is described. At the second stage of the precipitation process, clusters grow without a change in their number. Assuming that the precipitates have a spherical shape, it is possible to calculate the average radius of the precipitate at the growth stage:

$$R(t) = \sqrt[3]{\frac{3bi(t)}{4\pi}} \quad (5)$$

where b is the quantity of order of the distance between the particles in the cluster; $i(t) = [N(0) - N(t)]/N_c$ is the average number of particles at the nucleation centers; $N(0)$ is the monomer concentration at the initial instant of time; $N(t)$ is the changes in the concentration of monomers over time; N_c is the concentration of nucleation centers; and t is the time [17].

At the third stage, when the new phase particles are large and supersaturation is low, new particles are not formed and the main role is played by coalescence, which is accompanied by the dissolution of small-sized particles and the growth of large-sized particles. Under condition $R(t)/R_{cr}(t) \approx 1$ (where $R_{cr}(t)$ is the critical radius of the precipitate), the precipitate is in equilibrium with the solution ($dR/dt = 0$). The precipitate grows at $R(t) > R_{cr}(t)$ and dissolves at $R(t) < R_{cr}(t)$ [17, 18].

It is possible to determine the critical size of the precipitates in accordance with [19]

$$R_{cr}^O(t) = \frac{2\sigma u V_p}{kT(t) \ln(S_0 S_i^{-\gamma_i} S_v^{\gamma_v}) - 6\mu\delta\epsilon u V_p} \quad (6)$$

$$R_{cr}^C(t) = \frac{2\sigma u V_p}{kT(t) \ln(S_c S_i^{\gamma_i} S_v^{-\gamma_v}) - 6\mu\delta\epsilon u V_p}$$

where $S_o = \frac{C_o}{C_o^s}$, $S_c = \frac{C_c}{C_c^s}$, $S_i = \frac{C_i}{C_i^s}$, $S_v = \frac{C_v}{C_v^s}$ are the supersaturations of the oxygen atoms, carbon atoms, intrinsic interstitial silicon atoms, and vacancies, respectively; σ is the density of the surface energy of the interface between the precipitate and the matrix; μ is the shear modulus of silicon; δ and ϵ are the linear and volume misfit strains of the precipitate and the matrix, respectively; γ_i and γ_v are the fractions of intrinsic interstitial silicon atoms and vacancies per impurity atom attached to the precipitate, respectively; V_p is the molecular volume of the precipitate; and $u = (1 + \gamma_i x + \gamma_v x)^{-1} \cdot \left(\frac{1+\epsilon}{1+\delta}\right)^3$.

The average size of precipitates at the stage of the coalescence is proportional to the cube root of time [17]:

$$R_{av}(t) = \sqrt[3]{R_{cr}^3(t_0) + \frac{4D\beta t}{9}} \quad (7)$$

where D is the diffusion coefficient of impurity atoms; $\beta = \left(\frac{\sigma\Omega}{kT}\right)N(0)$; $R_{cr}(t_0)$ is the initial critical radius; σ is the surface tension at the precipitate-solid solution interface; and Ω is the atomic volume.

As is known, the formation and development of the structure of grown-in microdefects in silicon are determined by thermal conditions of growth and cooling of the crystal [1, 3]. The temperature distribution along the length of the ingot during its cooling varies depending on the thermal parameters of the growth according to the expression $1/T = 1/T_m + Gz/T_m^2$, where z is the distance from the crystallization front; G is the axial temperature gradient at the crystallization front; and T_m is the crystallization temperature [3]. It should be noted that, in the general case, it is necessary to take into account the radial inhomogeneity of the temperature field. We introduce the crystal growth rate (V) into this formula and obtain

$$T(t) = \frac{T_m^2}{T_m + VGt} \quad (8)$$

3. Results and discussion

In the paper [15], the interactions between the atoms of the substances and, consequently, their properties are determined on the basis of information about the potential of interatomic interaction. The exact form of the interaction potential of two atoms is determined from the quantum-mechanical calculations. The obtained potentials are usually described by the functions with a large number of parameters, which complicates their analytical treatment. Therefore, it is usual practice to operate model potentials that contain a small number of parameters.

An unambiguous determination of the interatomic potential for silicon crystals is complicated. Therefore, theoretical predictions within their formalism have used different fitting parameters whose behavior under various temperature conditions is difficult to predict or justify [15]. In the paper [15], characteristic numbers were determined from the basic criterion of the crystalline state (3) with the help of a model potential Mee-Lennard-Jones.

In this paper, the calculations were performed using the following parameters: $U_{1min} = 2.84$ eV (SiO_2), $U_{2min} = 2.71$ eV (SiC), $\lambda_1 = 4.482 \times 10^8 \text{ eV}^{-1}$ (SiO_2), $\lambda_2 = 1.099 \times 10^9 \text{ eV}^{-1}$ (SiC); а также $V_p = 4.302 \times 10^{-2} \text{ nm}^3$ (SiO_2), $V_p = 2.04 \times 10^{-2} \text{ nm}^3$ (SiC), $\sigma = 310 \text{ erg/cm}^2$ (SiO_2), $\sigma = 1000 \text{ erg/cm}^2$ (SiC), $\mu = 6.41 \times 10^{10} \text{ Pa}$, $\delta = 0.3$, $\varepsilon = 0.15$, $D_O = 0.17 \exp(-2.54 \text{ eV/kT})$, $D_c = 1.9 \exp(-3.1 \text{ eV/kT})$, $b = 0.25 \text{ nm}$, $k = 8.6153 \times 10^{-5} \text{ eV/K}$, $N_c = 10^{12} \text{ cm}^{-3}$.

We performed three separate groups of calculations that simulated the processes of precipitation during the growth of crystals of large and small diameters with the use of the Czochralski method (CZ-Si) and crucibleless floating zone melting (FZ-Si). Calculations of the first group (I) were performed using the following parameters: the crystal growth rate was $V = 0.6 \text{ mm/min}$,

the axial temperature gradient was $G = 25$ K/cm, the oxygen concentration was $N(0) = 10^{18} \text{ cm}^{-3}$, and the carbon concentration was $N(0) = 10^{18} \text{ cm}^{-3}$. The corresponding parameters used in calculations of the second group (II) were as follows: the crystal growth rate was $V = 0.3$ mm/min, the axial temperature gradient was $G = 25$ K/cm, the oxygen concentration was $N(0) = 10^{18} \text{ cm}^{-3}$, and the carbon concentration was $N(0) = 10^{18} \text{ cm}^{-3}$. These conditions correspond to the growth of large-sized silicon single crystals with the use of the Czochralski method. For calculations of the third group (III), we used the following parameters: the crystal growth rate was $V = 6$ mm/min, the axial temperature gradient was $G = 130$ K/cm, the oxygen concentration was $N(0) = 10^{18} \text{ cm}^{-3}$, and the carbon concentration was $N(0) = 10^{18} \text{ cm}^{-3}$. Group III corresponds to the conditions for the growth of small-sized silicon single crystals with the use of crucibleless floating zone melting.

In this, the computing experiment is assumed that the concentration of nucleation centers for complexes constitutes $\sim 10^{12} \text{ cm}^{-3}$. This value corresponds to the experimental data obtained by transmission electron microscopy [1]. The densities of the distribution of complexes of silicon-carbon for crystals I and II groups are shown in **Figure 1**. The densities of the distribution of complexes of silicon-carbon for crystals III are shown in **Figure 2**.

In accordance with the Vlasov model for solid state, the process of precipitation begins in the vicinity of the crystallization front due to the disappearance of excess intrinsic point defects on sinks whose role is played by oxygen and carbon impurities. The results of the calculations performed in groups I, II, and III allow us to compare the processes of precipitation in the CZ-Si and FZ-Si crystals and to analyze them for almost maximum contents of the oxygen and carbon impurities. Changing growth conditions of crystal (in particular, an increase in the growth rate and the axial temperature gradient) results in reduced time of the nucleation stage. Besides, the calculations in group III showed that the change in thermal conditions of the growth leads to a decrease in the average radius of precipitates in the FZ-Si crystals as compared to the CZ-Si crystals at the stage of the growth of precipitates, as well as to the corresponding decrease in the precipitate sizes at the stage of the coalescence.

Figure 1. Changing the density of distribution of complexes silicon-carbon for crystals I (1) and II (2) groups.

Figure 2. Changing the density of distribution of complexes silicon-carbon for crystals III group.

The condition providing changeover to the stage of the coalescence is written in the form $R(t) \approx R_{cr}(t)$, which is satisfied at the temperature $T \approx 1423\text{K}$. The stage of the coalescence in large-sized silicon single crystals begins at temperatures close to the temperatures of the formation of clusters of intrinsic point defects [17]. **Figure 3** shows the variation of the average size of oxygen and carbon precipitates at the stage of the coalescence in the temperature range of cooling from 1423 to 300 K.

Changing of growth conditions for small-sized FZ-Si single crystals (high growth rates and axial temperature gradients) gives what the stage of the coalescence begins far in advance (at $T \approx T_m - 20\text{K}$).

In the classical theory of the formation of second-phase particles during heat treatments of single crystals, the formation of critical nuclei of small sizes occurs at low temperatures. In this case, an increase in the temperature leads to the growth and coalescence of precipitates. In real silicon single crystals, there are grown-in microdefects that, during heat treatments, serve as sinks for intrinsic point defects and impurity atoms. Therefore, the growth of precipitates formed during the growth of single crystals can occur simultaneously with the formation of new precipitates.

However, the application of the Vlasov model for solid state to the processes of formation complexes during heat treatment of silicon crystals gives different result (**Figure 4**).

In accordance with the Vlasov model for solid state, the formation of complexes at temperature $T \approx 723\text{K}$ is the unlikely. This temperature corresponds to the average temperature of the formation of thermal donors. The formation of complexes in silicon during heat treatment is possible only at high temperatures. Excess intrinsic point defects and impurity atoms disappear at sinks whose role in this process is played by initial structural defects. This process leads to the growth and transformation of the initial defect structure of crystal.

At the same time, it is known that the thermal treatments of silicon lead to the formation of electrically active centers. If the formation of complexes in the temperature range $300\text{ K} \leq T \leq 600\text{ K}$

Figure 3. Variation of the average size R_{av} of (1) oxygen precipitates at the stage of the coalescence during cooling of the CZ-Si crystals in the temperature range from 1423 to 300 K.

Figure 4. Dependence of the density of distribution for the silicon-oxygen complexes from the temperature during the annealing of the crystal.

is unlikely, then the processes of coalescence occur in crystals of any diameter. Then, in accordance with the Vlasov model for solid state, it can be assumed that the grown-in precipitates, which dissolve, lead to the formation of electrically active centers.

In conclusion, we note that two theories of nucleation second-phase particles, which are based on different gnoseological approaches (classical theory of nucleation and growth of second-phase particles in crystals and the Vlasov model for solid state), lead to identical results. Both approaches describe the processes of high-temperature precipitation of impurities. In turn, the high-temperature precipitation of impurities is the basis of the process of defect formation in crystals.

4. Conclusions

The main task of the solid body is a description of the physical properties of solids in terms of their atomic structure. Despite the successful development of the classical approach in the last 100 years, the theoretical description of the process of defect formation in crystals has not received a satisfactory solution.

Until now, the probabilistic approach (Vlasov model for the solid state) for real crystals has not been applied. At the same time, joint use of both approaches would allow for a fresh look at the already-known facts and discover new phenomena and laws in the study of real solids. This work represents the first attempts in the last 70 years to obtain the solution of Vlasov model for solid state. Questions of the application of the Vlasov model for solid state for the description of the nucleation of defect structure, taking into account the thermal conditions of the crystal growth, were discussed. There are the following main results:

1. The concepts and principles of Vlasov physics are fully applicable to solid state.
2. The Vlasov model for solid state describes the processes of complexation during the growth of real crystals adequately classical theory of nucleation and growth of second-phase particles in solids.
3. The method of calculation of the initial defect structure of crystals was proposed. This method includes the Vlasov model for solid state and the classical theory of nucleation and growth of second-phase particles in solids.
4. With the help of the Vlasov model for solid state, it was shown that during low-temperature treatments of crystals, the complexation is unlikely. In this case, we have the processes of coalescence of grown-in defects.

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