Physics – Uspekhi **40** (4) 387 – 392 (1997)

REVIEWS OF TOPICAL PROBLEMS PACS numbers: 66.30.Fq; 66.30.Lw, **66.90.r**, 78.50.Ge

Atomic migration and related changes in defect concentration and structure due to electronic subsystem excitations in semiconductors

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Contents

Abstract. Exciting the electronic subsystem of a semiconductor via photoionization or ionization by charged particles, or, alternatively, injecting nonequilibrium charge carriers into a semiconductor stimulates atomic migration, generates new structural defects, and modifies the nature of those present. These effects change the major electrical and physical parameters of semiconductors, in particular of those crucial for modern solid-state electronics. Current data on the subject are presented and discussed.

1. Introduction

The solid-state (semiconductor) electronics is an integral component of the modern civilization [1]. Presently it is difficult to realize how people lived without radio and television, simple calculators and state-of-the-art computers, or electronic copiers (xerox). The lifetime of semiconducting devices, however, is usually significantly shorter than the human lifetime (seventy years, according to the Bible).

One of the main causes for degradation and failure in semiconductor devices is gradual accumulation of point defects or larger structural irregularities. Detailed information on the nature and energy spectrum of such defects in some semiconductors, primarily in single-crystal silicon is available in Refs [2, 3]. The progress in growing silicon single crystals and epitaxial films achieved in recent years allows one to fabricate samples with a low concentration of grown-in point defects and dislocations. In particular, the concentration of point defects has been reduced to 10^{10} cm^3, and that of dislocations, down to several units per square centimeter. This is the reason why the basic parameters of silicon are so susceptible to the generation of additional nonequilibrium defects, for example, in physical processes initiated by 'hard' radiation (gammas, fast electrons, fast neutrons, etc.) [4, 5].

Initially the researchers of radiation damage in solids supposed that the dominant mechanisms of the defect generation were impact displacements of atoms or local overheats (thermal spikes), which led to complete disorder in the crystal structure for a short time.

However, excitations in the semiconductor electronic subsystem, i.e., the presence of nonequilibrium charge carriers, may also facilitate migration of defects and impurities and, hence, structural changes, such as the generation of new defects and complexes including point defects and impurities.

The prediction (estimate) of the life time of solid-state devices has invariably been a topical problem. 'Impact' mechanisms of the generation of radiation defects can fairly accurately be described based on the concept of a threshold energy of generation of Frenkel pair (an interstitial atom plus a vacancy). Until the present time, calculations of the number of displaced atoms have usually been based on experimental data about the threshold energies required to displace atoms from their sites. The calculated number of displaced atoms is always slightly higher than the real one for the reasons described in Ref. [6]. As a result, the service life of devices exposed to hard radiation estimated by this method has a considerable safety margin.

Table 1 lists approximate threshold energies E_d required for an impact displacement of an atom from its site, i.e., generation of a Frenkel pair [6, 26].

In the next sections of this review, we will consider the processes induced by excitations in the semiconductor electronic system and leading to migration (displacements) of host atoms and structural rearrangements of defects.

2. Athermal mechanisms of atomic migration

Atomic diffusion in solids due to thermal excitation has been studied in much detail by the present time [7, 8]. In addition to these mechanisms, there are athermal mechanisms of atomic migration assisted by structural points defects due to hard radiation (radiation-stimulated diffusion, RSD) and by the presence of nonequilibrium charge carriers. RSD was first

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Received 15 July 1996
Uspekhi Fizicheskikh Nauk **167** (4) 407 – 412 (1997)
Translated by A V Malyavkin; edited by S N Gorin

Table 1. Threshold displacement energies for various materials.

Material	E_d, eV	T, K
Ge	~ 30	80
	$\leqslant 23$; 22.3; 14.5; 18; 15.5	300
	18.0	21
	14.5	79
	12.7	263
	14	78, 269
Si	20.9; 13	300
	14 (*n*-conductivity); 21 (*p*-conductivity)	80, 300
	22	80
	11	300
	20	80
	~ 13; 45	300
Diamond	80	300
Graphite	24,7	290
	60	15
	33; 31	300
	(23; 40.0); (31, 30)	6, 80
	(28, 42.0)	285
InSb	5.7 (In), 6.6 (Sb)	78
	6.4 (In), 8.5 – 9.9 (Sb)	80
InAs	6.7 (In), 8.3 (As)	77
InP	6.7 (In), 8.7 (P)	77
GaAs	9.0 (Ga); 9.4 (As);	300
	15	77
GaSb	6.2 (Ga); 7.5 (Sb)	77
ZnSe	7.6 (Zn); 8.2 (Se)	85
ZnTe	7.4 (Zn); 9.7 (Zn); 6.7 (Te)	10
	4.2 (Zn)	77
	7.35 (Zn)	10
ZnS	9.9 (Zn) or 20.2 (S); 9.9 (Zn);	10
	15 (S)	
CdS	8.7 (S)	300
	7.3 (Cd)	77
CdSe	8.6 (Se)	77
	8.1 (Cd); 8.6 (Se)	5
CdTe	7.8 (Te)	77
	5.6 (Cd); 7.8 (Te)	5
MgO	60 O)	300
ZnO	30 (Zn); 57 (Zn); 57 (O)	300
BeO	76 (O)	300

reported by P Baruch and J Pfister in 1963 [9]. They demonstrated that proton irradiation of silicon crystals leads to acceleration of the diffusion of electrically active impurities such as P, B, and Ga. The authors ascribed this effect to the large number of vacancies caused by generation of Frenkel pairs in the regions where proton trajectories terminate. It was clearly demonstrated in subsequent publications that vacancies really stimulate impurity diffusion. In recent years, the effect of radiation stimulated diffusion has extensively been used in the technology of planar microelectronic devices [5, 6].

Bourgoin and Corbett [10] suggested an alternative model of RSD to account for the stimulated diffusion in nonmetallic solids. This model was based on Weiser's theory [11], which took into account the difference between the energies of a charged impurity atom in an interstitial tetrahedral position (T) and in a hexagonal position (H). According to Weiser's model, the difference between these energies for Ge and Si is about 0.75 eV. Hence, a change in the charge state may lead to a sequence of 'hops' of an interstitial atom, which is observed as RSD. Figure 1 shows a diagram where the potential energy for the two possible positions of an interstitial atom, i.e., the tetrahedral (T) and hexagonal (H) positions, is plotted.

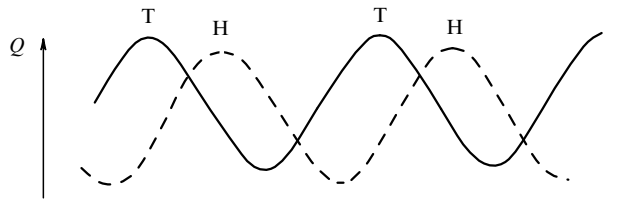

Figure 1. Variation of the potential energy of two possible positions of interstitial atoms, namely, tetrahedral (T) and hexagonal (H) (schematic).

The mechanism of defect delocalization in nonmetallic solids was proposed independently by Oksengendler [12]. The model suggested also implied that the potential acting on a defect in a crystal depended on the state of the electron localized on it. The motion of a defect over a macroscopic distance consisted of hops due to electronic transitions between the ground and excited states.

3. Electrostatic (Coulomb) instability as an alternative driving force for atomic migration in semiconductors

As early as in 1938, the dissociation caused by ionization of organic molecules was ascribed to the 'Coulomb explosion' of a doubly ionized molecule owing to the repulsion between the molecular fragments with like charges [13, 14].

In 1954 Varley [15] proposed a model of a similar process in ionic crystals. The central point of the model was the assumption that a doubly ionized atom at a lattice site was in a state unstable toward a transition to one of neighboring interstitial positions, and the transition could be driven by thermal excitation [15]. After further consideration, the applicability of Varley's model to ionic crystals was questioned because the lifetime of a multiply charged ion in a crystal was known to be short [6, 16]. Semiquantitative estimates concerning the motion of Cl^+, Cl^{2+}, and Cl^{3+} ions in the KCl lattice were given by Chadderton et al. [17], who assumed that the lifetime of these ions (τ_d) should be sufficiently long so that an energy $\Delta E \geqslant E_d$, where E_d is the minimal energy needed to generate a defect, could be transferred to the ion, i.e., an energy of several electronvolts. In other words, a force of about 1 eV Å^{-1} should act during a time of the order of the lattice oscillation period ($> \omega_0^{-1} \simeq 10^{-13}$ s), whereas for impact displacement a larger force ($\gg 1$ eV Å^{-1}) acting during a shorter time ($< \omega_0^{-1}$) is required. This suggests that for 'adiabatic' (nonimpact)

displacement of an ion from its site, its lifetime τ_d should be longer than $10^{-13}-10^{-12}$ s. A detailed analysis of the lifetime τ_d of ionized states was given by Elango and Kiv [16]. According to their preliminary conclusions, doubly charged ions in alkali-halide crystals cannot accumulate sufficient energy for their displacement, and the lifetime of triply charged ions is too short. According to Ref. [16], however, a mechanism of atomic displacement similar to Varley's mechanism can be effective in semiconductors since the force due to the Coulomb repulsion between two neighboring atoms can act for a time corresponding to the Auger process in an inner shell driven by ionizing radiation. A similar mechanism can generate divacancies in silicon [18]. The models proposed by Chadderton et al. [17] and Iskenderova et al. [18] were criticized by Norris [19] and by Mooney and Bourgoin [20], who claimed that the effects observed in experiments with Si could be caused by processes on surfaces or near contacts. Experimental data indicated that the effects of 'Coulomb' instability in semiconductors such as Ge and InSb were stronger in the presence of shallow donors [21]. This observation was confirmed by later experiments [22–24].

Generation of point defects due to repulsion between an ionized host atom and a charged impurity (the so-called impurity-ionization mechanism) was also analyzed in detail by Karpov and Klinger [25] and other authors [21, 22]. The additional potential due to an ionized atom with charge q was treated as an abrupt perturbation of the phonon spectrum under the condition $\tau_a \ll \omega_0^{-1}$. It was demonstrated that in the case of a typical Auger process involving a 'deep' hole ($2 \leqslant q \leqslant 10$ and $\tau_a \simeq 10^{-15}-10^{-14}$ s) the probability of athermal displacement of an ionized atom was larger, whereas in the absence of charged impurity atoms the probability was small.

Generation of defects by this mechanism is possible within a sphere of radius

$$R_i \simeq \frac{q_1 q_2 e^2}{\varepsilon E_d}$$

around an impurity. Since such a sphere contains Z host atoms, the probability of generating a defect is proportional to $Z\sum_j \sigma_j$, where σ_j is the probability (cross section) of ionization of the jth inner shell of an isolated host atom. According to the experimental data obtained by Mashovets et al. [21, 23], the efficiency of defect generation in Ge and InSb by soft x-rays or low-energy electrons can be described using quite realistic values of Z. Moreover, in some semiconductors this mechanism of defect generation can dominate [23] even over the concurrent impact generation of defects by gammas from Co^{60} if the impurity concentration in a semiconductor is higher than $\simeq 3 \times 10^{17}$ cm^{-3}. To the best of our knowledge, no systematic data concerning the efficiency of the impurity-ionization mechanism have been published as yet.

Vavilov et al. [26] reported on the generation of free vacancies in diamond irradiated by electrons with energies several times lower than the threshold for impact displacement of atoms from their lattice sites. The issue of 'non-impact' mechanisms of defect generation was discussed in Ref. [6]. Milevskiĭ and Garnyk [27] observed in silicon single crystals that primary Frenkel pairs generated by local irradiation with fast 2.2 MeV electrons could be separated into the components. After irradiation of n-type samples with an initial electron concentration of 10^{13} cm^{-3}, the photo-emf established between the irradiated and irradiated parts of the sample was measured by the standard technique used for testing the homogeneity of materials.

Figure 2 shows curves of photo-emf plotted against the radiation dose under different experimental conditions. An electric field directed along the sample (in which case its maximum intensity is near the boundary between the exposed and shielded regions) and an increase in the temperature during the exposure shift the photo-emf curve to the left, i.e., to the range of lower doses. The recorded spectra indicate that the photo-emf is due to electron transitions from the valence to conduction band. The photo-emf signal intensity versus the electric field across the sample during its exposure to radiation is shown in Fig. 3. It clearly demonstrates that the external electric field increases the emf intensity by about one order of magnitude. The photo-emf is proportional to the resistivity gradient [29]; hence, the increase in the signal is due to a lower concentration of charge carriers in the exposed section of the sample because of generated radiation defects. If the number of fast electrons required to generate one Frenkel pair is assumed to be independent of the electric field applied to the exposed sample, one can conclude from these experimental data that the components of a vacancy–interstitial pair have opposite elementary electric charges. In this case the annihilation of these particles should strongly depend on the electrostatic interaction between them. The jump in the photo-emf at a definite electric field intensity (Fig. 4) is caused by the inhibition of annihilation of primary radiation-induced vacancies and interstitial atoms; as a consequence, the concentration of secondary radiation defects in the exposed section of the sample is higher.

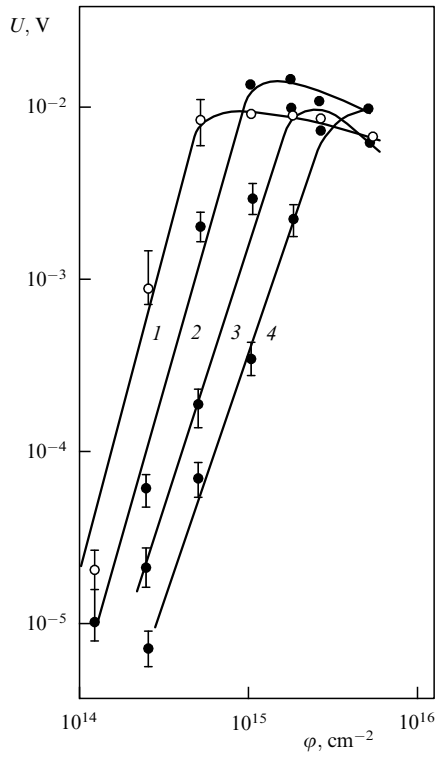

Figure 2. Variation of the photo-emf U as a function of the radiation dose φ. The electron flux density is 2.6×10^{12} cm^{-2} s^{-1}; $T = 300$ K (*1*, *3*) and 100 K (*2*, *4*); $E = 110$ V (*1*, *2*) and 0 (*3*, *4*).

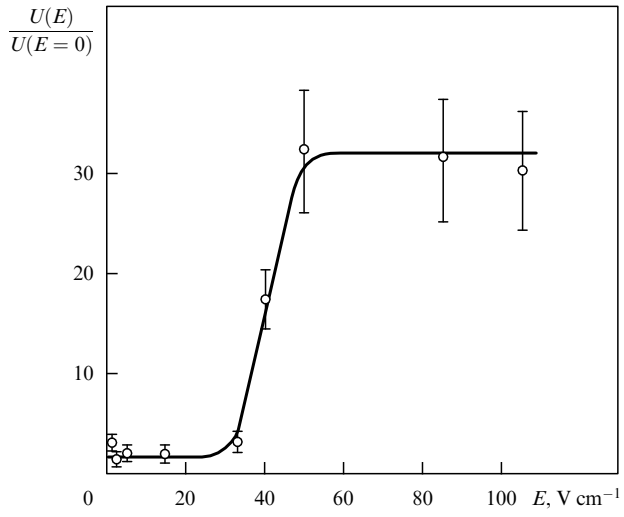

Figure 3. Variation of the relative photo-emf as a function of an applied electric field ($\varphi = 3.9 \times 10^{14}$ cm^{-2}).

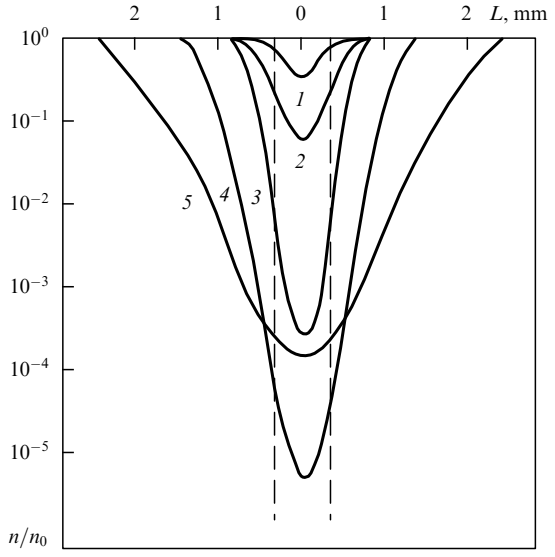

Figure 4. Relative change in the bulk concentration of carriers in the region exposed to radiation and neighboring regions. The exposure times are (*1*) 100, (*2*) 200, (*3*) 400, (*4*) 700, and (*5*) 2000 s. $T \simeq 300$ K. The dashed lines outline the boundaries of the exposed region.

It is reasonable to assume that the components of Frenkel pairs annihilate in an isotropic medium with a dielectric constant equal to that of silicon, and that the electric field generated by a pair and the applied field are directed oppositely. Then, we can estimate the separation between the vacancy and interstitial silicon atom 'genetically related' to one Frenkel pair by substituting the electric field corresponding to the photo-emf jump into the Coulomb formula. This separation is close to 10^{-5} cm, i.e., several hundreds of interatomic distances. This is ten times as small as the mean separation between positively charged phosphorus atoms that control the conductivity type and the equilibrium concentration of majority carriers in the sample studied. Milevskiĭ and Garnyk [27] detected, in addition, a notable broadening of the photosensitive region beyond the region exposed to fast electrons. They ascribed this broadening to the diffusion of primary radiation defects away from the exposed region. This is, apparently, the reason for the saturation of curves in Fig. 2. The drop in the photo-emf observed when irradiation is performed at lower temperatures is also in qualitative agreement with the existing concepts of thermally activated motion of vacancies and interstitial atoms [30].

Vavilov et al. [26] revealed that the concentration of luminescence centers including vacancies in diamond crystals increased rapidly as the electron beam energy grew to 6–7 keV, which corresponds to the ionization threshold of *K* shells of transition-metal atoms (Mn, Fe, Co), which are invariably present in diamond in the form of impurities. They interpreted this effect in terms of the impurity-ionization mechanism. The results discussed in this section lead us to the conclusion on the important role of the 'Coulomb instability' in semiconductors. The issue of the efficiency of this process in silicon deserves further investigation with a view to add new information to the experimental data given in Ref. [6].

4. Ionization effects during ion implantation

In connection with the wide application of ion implantation in semiconductors, primarily in single-crystal silicon [31], ways of reducing concentration of defects inevitably generated by ion implantation have actively been sought in recent years. Two methods have been, in particular, examined, namely, 'hot' ion implantation, when a target is heated during the exposure, and the application of UV radiation [32]. After the first positive results of the UV exposure, the authors ascribed the effect to heating. But later experiments [33] indicated that the effect of the UV radiation on the ion implantation in silicon was athermal.

The effect of ionization on the generation of radiation defects was discussed by Ascheron et al. [34]. In their experiments, they used accelerated ions with various energies, masses, and charges. Since multiply charged ions have very short lifetimes, displacement of atoms of targets near their surfaces in III–V and II–VI compound semiconductors and in germanium single crystals was studied. In these experiments, ions with masses $M = 1-40$ a.u. and energies ranging between several keV and tens of MeV were used. The defect concentration as a function of the ion range was derived from Rutherford back-scattering (RBS) spectra of channeled ions [35]. The quantity of the material ablated from the surface was derived from the amount of substance deposited on a plastic (capton) film.

Calculations were performed using the TRIM code [36]. Programs of this type, which have been widely used recently, as well as the initial Seitz's models and those described in Ref. [4], assume that a Frenkel pair is generated when an energy higher than the threshold energy E_d is transferred to an atom. The TRIM program also takes into account the possibility of spontaneous recombination if a displaced atom stops near a vacancy at a distance smaller than a limiting 'recombination radius,' which determines the instability sphere of a Frenkel pair. Replacement collisions and generation of secondary, tertiary, etc. Frenkel pairs are also included in the model. According to some estimates, the calculation uncertainty is $\pm 30\%$ [30]. Calculations by Ascheron et al. [34] are given in Fig. 5. They are in qualitative agreement with concepts developed before the computer era. These plots clearly illustrate the distribution of primary

Frenkel pairs around the trajectory of an implanted ion shown by black dots. Generation of radiation defects in semiconductors was experimentally studied by many authors, and most of them found direct proportionality between the energy lost in nuclear collisions and the number of generated defects.

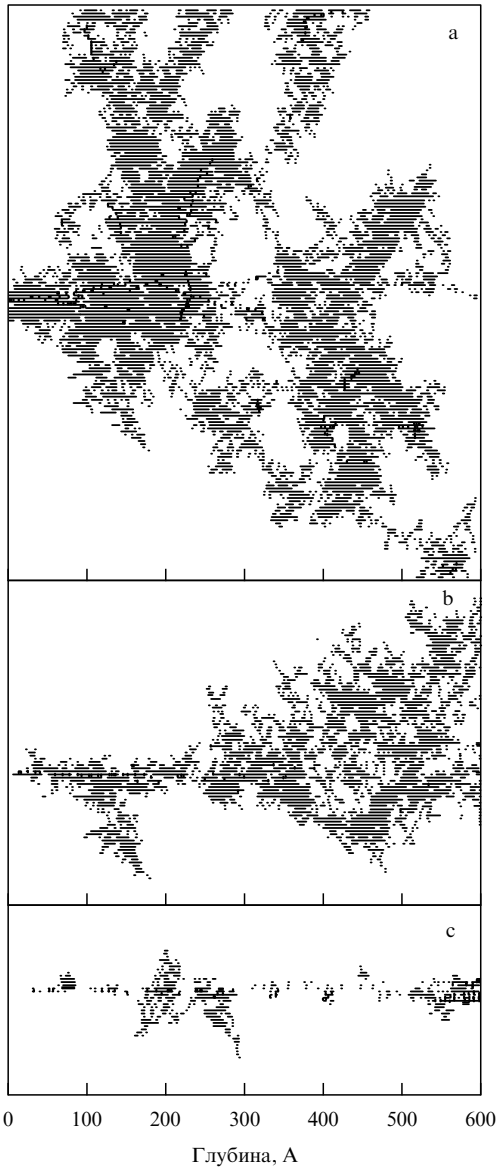

Figure 5. Patterns of collision cascades generating Frenkel pairs. Calculation using the TRIM code for Ge irradiated with Ar^+ ions with energies of 50 keV (a), 500 keV (b), and 5000 keV (c) [34].

Deviations from the direct proportionality are usually attributed to replacement collisions and a larger fraction of mutually annihilating vacancies and interstitials at high energy losses. Both these effects lead to a sublinear dependence of the number of defects on the dissipated energy per unit volume. An additional drop in the number of defects is due to interaction with electrons, which carry away a fraction of kinetic energy that would otherwise be spent for generating a cascade of displaced atoms. According to Ascheron et al. [37], in some cases the number of generated defects, however, is considerably larger than the TRIM calculations. The experimental data plotted in Fig. 6 indicate that ionization processes are of prime importance in the generation of defects. There is a clear correlation between the number of defects and specific ionization losses S_e. The number of generated defects is a function not only of S_e, but also of the sort (mass) of implanted ions. This may be caused by the dependence of the probability of annihilation between Frenkel-pair components on the ion mass, since displacement cascades become more compact at higher ion masses. Unfortunately, the dependence between the number of generated defects and concentrations of impurities in semiconductors [34] has not been studied in detail by this time. In this connection, it is too early to discuss the possibility of controlling generation of defects through impurity-ionization mechanisms [25] and other processes discussed above and in Ref. [16].

Ascheron et al. [34] concluded from their experimental data that it is the stripping off of the valence electrons from the atoms of the implanted material in the process of development of a collision cascade that played an important role. The number of generated defects increased under certain conditions by a factor of nearly 1.6. According to [34], this increase was caused by a lower displacement energy of ionized atoms. Experiments with GaP and other III–V compound semiconductors, and with ZnSe (II–VI compound) provided evidence in favor of this effect, whereas it was not found in elementary semiconductors such as Ge and Si [35].

As was noted above, in order to rule out the effect of intense annihilation of defects in the process of and after the development of displacement cascades, sputtering from bombarded target surfaces was studied [35]. The authors of this publication assumed that sputtering data yielded information directly related to the initial 'collisional' stage of the displacement cascade development before the onset of thermal (or rather quasi-thermal) effects. Detailed information about the sputtering caused by ion beams is contained in [38] and [39]. A diagram illustrating processes due to interaction between accelerated atoms and a solid is given in Fig. 6. In experiments by Ascheron et al. [34] the energy lost to

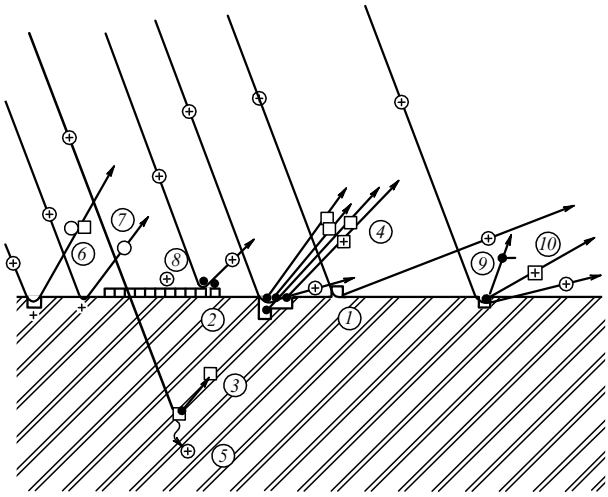

Figure 6. Reactions due to interaction between ions and a semiconductor (schematic) [39]: (*1*) ion scatterng by atoms; (*2*) subsurface dislocations; (*3*) dislocations in the bulk; (*4*) physical sputtering; (*5*) ion implantation; (*6*) chemical sputtering; (*7*) charge transfer; (*8*) ion adsorption; (*9*) electron emission; (*10*) emission of surface ions.

excitations in the electronic subsystem was measured by varying the energy of ions bombarding a target (a germanium single crystal). The total amount of sputtered atoms was determined by integrating their angular distribution. The comparison between the theory and experimental data indicated that nuclear collision processes can account for the experimental data only at ion energies lower than approximately 10 keV. At higher energies, when the energy lost by an ion to ionization produced in a semiconductor becomes essential, the experimentally determined sputtering yields are larger than the theoretical prediction. This increase in the sputtering yield can be taken into account by introducing into the TRIM calculation decreased binding energies of electrons at the surface and in the bulk of a crystal. It seems most probable that the decrease in the energy of interatomic bonds in the bulk is due to internal ionization, i.e., delocalization of valence electrons inside an excited crystal.

5. Conclusions

The material discussed in this review indicates that the problem of defect generation in semiconductors exposed to ionizing radiation or driven to a nonequilibrium state by other agents deserves further investigation and will remain a topical problem in the near future.

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