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Nonlinear Diffusion in Excited HgCdTe and Si Crystals

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We discuss the properties of the nonlinear diffusion equation for the case of diffusion in excited systems. The diffusion coefficient is directly proportional to the concentration of impurities and depends on time in a special way. For the description of the excited systems, we used a special temperature function, which defined the time dependent diffusion coefficient and the Boltzmann distribution of the excited vacancies or impurity atoms in solids. This model was used for the approximation of indium concentration profiles in HgCdTe of a rapid diffusion component and very fast diffusion of metastable vacancies irradiated by soft X-rays in an excited Si crystal.

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1. Introduction

We discuss the properties of the nonlinear diffusion equation in the non-isothermical case. In our previous papers [1, 2] we used the diffusion coefficient that was directly proportional to the concentration of the impurities, which depends on time in a special way. With this aim in mind, we have chosen a temperature function, whose negative value reflects time dependence of the diffusion coefficients in the excited systems. The obtained unique analytical solutions define the rapid thermal diffusion profiles of indium in HgCdTe sample irradiated by 4.14 eV photons from the tungsten halogen lamps and diffusion of vacancies irradiated in Si crystal by soft X-rays. The obtained analytical solutions define the rapid thermal diffusion profiles in the case of the excited systems where vacancies and impurity atoms are not in thermal equilibrium with the lattice. In this situation, the diffusion coefficients for diffusion of atoms or vacancies are increased by thousands of times.

2. Similarity solution of the nonlinear diffusion equation for excited systems

We consider the properties of solutions of the nonlinear diffusion equation for excited systems. The diffusion coefficient in our case is equal to zero in the region where impurities are absent [1]. The nonlinear diffusion equation for the excited systems or for the non-isothermal case can be written in the following way [2]:

$$\frac{\mathrm{d}N}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}x}\left(D\frac{\mathrm{d}N}{\mathrm{d}x}\right), \quad D = D_n N, \quad D_n = \frac{D_0}{N_s}\mathrm{e}^{-E/kT},$$

$$T(t) = \frac{1}{a - b\ln(tc)}. \tag{1}$$

Here, $N(x,t)$ and N_s are the concentrations of impurities in the bulk of the crystal and in the surface layer, respectively. D_0 is the pre-exponential factor and E is the activation energy. The temperature function $T(t)$ depends on parameters a, b, c and it has a singularity at the time moment t_0. Expressing the time t in units of t_0, we obtain

$$T(t) = -\frac{1}{b\ln(z)}, \quad t = zt_0, \quad t_0 = \frac{1}{c}\mathrm{e}^{a/b}. \tag{2}$$

It is very important that the temperature function $T(t)$ corresponds to the Boltzmann statistics for the description of the population ratio $P(t)$ of two quantum levels $E_2 > E_1$:

$$P(t) = \frac{\omega(E_2)}{\omega(E_1)} = \exp\left[-E_{21}/kT(t)\right],$$

$$P(t) = z^{E_{21}b/k}, \quad E_{21} = E_2 - E_1. \tag{3}$$

If $b < 0$, then all excited vibration states E_2 of the atoms taking part in diffusion are populated at the initial time, when $z \to 0$. By introducing the similarity variable

$$\xi = \frac{x}{\sqrt{D_\mathrm{s}t}}, \quad D_\mathrm{s} = D_n N_\mathrm{s},$$

$$N_\mathrm{s} = \mathrm{const}, \quad N(x,t) = N_\mathrm{s} f(\xi), \tag{4}$$

the nonlinear diffusion Eq. (1) can be transformed into the following expression:

$$2\frac{\mathrm{d}}{\mathrm{d}\xi}\left(f\frac{\mathrm{d}}{\mathrm{d}\xi}f\right) + \alpha(\xi - \xi_0)\frac{\mathrm{d}}{\mathrm{d}\xi}f + \alpha\xi_0\frac{\mathrm{d}}{\mathrm{d}\xi}f = 0,$$

$$\alpha = 1 + \frac{Eb}{k}, \quad N(x,t) = N_s f(\xi), \quad \xi \in (0, \xi_0). \tag{5}$$

The solution of the latter equation was obtained by expanding it in the following power series at the maximum penetration point ξ_0 of impurities [3]:

$$f(\xi) = \sum_{n=1}^{\infty} b_n(\xi - \xi_0)^n, \quad 0 \le \xi \le \xi_0. \tag{6}$$

The following boundary conditions must be satisfied:

$$f(0) = 1, \quad f(\xi_0) = 0. \tag{7}$$

Substituting the power series (6) in Eq. (5), we obtain the relations between coefficients b_n:

$$2n \sum_{m=1}^{n} (n+1-m) b_{n+1-m} b_m + \alpha(n-1) b_{n-1} + n\alpha\xi_0 = 0,$$

$$n = 1,\ 2,\ 3 \ldots \tag{8}$$

Solution of the system of Eqs. (8) consisting of five equations with the boundary conditions (7) yields the following exact results:

$$b_1 = -\frac{\alpha}{2}\xi_0,\ b_2 = -\frac{\alpha}{8},\ b_3 = -\frac{\alpha}{144\xi_0},\ b_4 = -\frac{\alpha}{1152\xi_0^2},\ b_5 = -\frac{11\alpha}{172800\xi_0^3}, \tag{9}$$

and the approximate values of maximum penetration points $\xi_{0,n}$:

$$\xi_{0,1} = \sqrt{\frac{2}{\alpha}},\ \xi_{0,2} = \sqrt{\frac{8}{3\alpha}},\ \xi_{0,3} = \frac{12}{\sqrt{55\alpha}},\ \xi_{0,4} = \frac{8\sqrt{2}}{7\sqrt{\alpha}},\ \xi_{0,5} = \frac{120\sqrt{12}}{\sqrt{66161\alpha}}, \tag{10}$$

depending on the number n of included coefficients b_n. When $\alpha \ll 1$ and $b < 0$ in (5), we have very fast diffusion of vacancies or diffusing atoms, which are excited at the initial moment of time. The values of ξ_0 converge very rapidly, and from (10) we obtain $\xi_{0,4} = 1.6162/\sqrt{\alpha}$, $\xi_{0,5} = 1.6161/\sqrt{\alpha}$. Then from (4) we obtain the following expression of the coordinate of the maximum penetration depth of impurities:

$$x_{0,5} = 1.6161\sqrt{\frac{D_\mathrm{s} t}{\alpha}}, \tag{11}$$

which depends on the excitation parameter α. The convergence and uniqueness of the solution of Eq. (5) and nonlinear diffusion Eq. (1) with $\alpha = 1$ were proved in [3]. For small α and when $\xi_0 \gg \xi$, Eq. (6) yields

$$\lim_{x \to 0} f(\xi) = \sum_{n=1}^{\infty} b_n (-\xi_0)^n, \quad f(\xi) = 1. \tag{12}$$

Taking into account that $\xi_{0,2} = 1.63/\sqrt{\alpha}$ and $\alpha \ll 1$, we can conclude that in order to fit the experimental impurity profiles with the theoretical ones, it is sufficient to include only the first two terms in the series (6). Then from (4) , (9), (10) we obtain the approximate expression

$$N(x,t) \approx N_\mathrm{s}(1 - 0.4075\sqrt{\alpha}\xi - 0.125\alpha\xi^2),$$

$$0 \le \xi \le \xi_{0\mathrm{e}}, \quad \xi_{0\mathrm{e}} = 1.63/\sqrt{\alpha} \tag{13}$$

for the theoretical profile of impurities or diffusion of excited vacancies. Introducing the diffusion coefficient for the excited system D_ex and a new similarity variable

$$D_{\text{ex}} = \frac{D_{\text{s}}}{\alpha}, \quad \xi_{\text{e}} = \frac{x}{\sqrt{D_{\text{ex}} t}}, \tag{14}$$

we can obtain the solution similar to the one given by (6), but with the new values (14) of diffusion coefficient D_{ex} and similarity variable ξ_{e}. In this case, instead of (13), we have the theoretical profile

$$N(x,t) \approx N_{\text{s}}(1 - 0.4075\xi_{\text{e}} - 0.125\xi_{\text{e}}^2), \quad 0 \le \xi_{\text{e}} \le \xi_0, \quad \xi_0 = 1.63. \tag{15}$$

By comparing it with the experimental profile, we can find D_{ex}. With this aim in mind, we use the derivative of the impurity concentration profile (15) at the surface of the crystal

$$D_{\text{ex}} = \frac{0.4075^2}{t} \left(\frac{1}{N_{\text{s}}} \frac{\mathrm{d}N(x,t)}{\mathrm{d}x} \right)^{-2} \Bigg|_{x=0}. \tag{16}$$

In the case of diffusion in the excited systems, there is a wide region $\xi < 1$ or $x < \sqrt{D_{\text{ex}} t}$, where the concentration profiles can be represented by straight lines. Such profiles were obtained for rapid thermal diffusion of indium [4] into a HgCdTe crystal. The high-energy photons with energy $E = 4.14$ eV from tungsten halogen lamps reduced the activation energies for 0.001 part of diffusing indium atoms. If the excitation parameter α is very small, then the maximum penetration depths (11) of the impurities are very large. The maximum penetration depths in this case can be used for finding the excitation parameters. The rapid diffusion can be obtained after the irradiation of the diffusion area with a small absorption coefficient in the ground lattice. Also we must get the resonance absorption for transferring the impurity atoms into the excited states and generating the excited vacancies. We introduced the excited and very fast vacancies in an Si crystal [5] by generating the vacancies at the surface of the crystal with soft X-rays. The excited metastable state of Si crystal with a high concentration of rapid vacancies was obtained at room temperature [5]. It is possible to achieve rapid vacancy-assisted diffusion of impurities in the Si crystal even at room temperature.

It is interesting that when α vanishes to zero (5) we have superdiffusivity [2]:

$$\lim_{\alpha \to 0} b = b_0, \quad b_0 = -\frac{k}{E}. \tag{17}$$

When the maximum penetration of impurities (10) into the crystal tends to infinity (11), a more handy expression for the dependence of maximum penetration depth on parameter α can be obtained by substituting (2) into (11):

$$x_{05}(t) = 1.6161\sqrt{D_0 t_0 z^{\alpha}/\alpha}, \quad t = z t_0. \tag{18}$$

When $0 < \alpha \ll 1$, then $z^{\alpha} \approx 1$. In this situation, the superdiffusion can be achieved for the time period t_0. In this case we have $x_0 = 1.6161\sqrt{D_0 t_0/\alpha}$, and the maximum depth of penetration depends on the excitation parameter α and the system relaxation time t_0. Excited atoms of impurities or excited vacancies involved in the fast diffusion process are not in thermal equilibrium (3) with the

crystal lattice. The possibility to create singly charged vacancies of high mobility for a rather long time (about 200 min) in Si crystal (the size of the crystal was $3.33 \times 3.06 \times 3.09$ mm^3) was demonstrated experimentally [5]. For small values of the excitation parameters α, the maximum penetration of impurities (18) is proportional to $z^{\alpha/2} \approx 1$ and it weakly depends on diffusion time. In this case we have diffusion with a specific inertia. The nonlinear heat transport equation can be written in the same way as the diffusion Eq. (1).

The thermal inertia of the response of metals to ultrashort laser pulses and strong localization of hot electrons were obtained during the fast laser heating of solids [6]. When the heat is transported via the electron gas, which is out of equilibrium with lattice, the effect of ultrafast heat transport can be achieved. It is possible to explain the ultrafast diffusion using the above-presented model. Very fast diffusion can be obtained when the excited vacancies or diffusing atoms of a system [4] are in metastable states [7] out of equilibrium with the lattice for a sufficiently long time. We can understand rapid diffusion better if we take into account that the diffusion coefficient is directly proportional to the population [2] of the excited states

$$D_{\mathrm{s}} = D_0 P(t), \quad P(t) = z^{\alpha-1}, \quad \alpha - 1 = \frac{Eb}{k}, \quad t = zt_0. \tag{19}$$

When $\alpha < 1$, at the initial moment of time all ground states E_1 are empty and all excited states E_2 are populated. In this case, diffusion rate depends on the system relaxation time, which can be characterized by parameter t_0 when $\alpha < 1$.

3. Results and conclusions

In the rapid thermal diffusion experiments [4] indium concentration profiles in the HgCdTe sample were obtained. Heating was provided by tungsten halogen lamps. Two essentially different kinds of concentration profiles of diffusing indium were obtained. Indium concentration profiles of the first kind were successfully fitted with the complementary error functions, whereas the profiles of the second kind, which represent the rapid diffusion, were not fitted with any realistic model. Less than one percent of indium atoms were involved in this fast diffusion. These experimentally detected fast components of diffusing indium atoms in the HgCdTe crystal were activated by tungsten–halogen ultraviolet radiation. They can be represented [4] by straight lines up to the depth of 12 μm. These experimental profiles of rapid diffusion of indium can be fitted with the presented theoretical profiles (15). Using the rapid components of the diffusion profiles for three different temperatures $T_1 = 453$ K, $T_2 = 423$ K, $T_3 = 393$ K presented [4] in Fig. 4 in conjunction with formula (16), the following diffusion coefficients and maximum penetrations depths were obtained: $D_{\mathrm{ex},1} = 14.4 \times 10^{-8}$ cm^2/s, $D_{\mathrm{ex},2} = 9.8 \times 10^{-8}$ cm^2/s, $D_{\mathrm{ex},3} = 0.35 \times 10^{-8}$ cm^2/s, $x_{05,1} = 39$ μm, $x_{05,2} = 27$ μm, $x_{05,3} = 23$ μm. The slow diffusion components of indium atoms are significantly

smaller and are equal to 3.5 μm, 3 μm, 1.5 μm, respectively. Using parameters [4] for slowly diffusing indium components $D_0 = 1.45 \times 10^{-2}$ cm^2/s, $\Delta E = 0.772$ eV, we calculated diffusion coefficients (1), (4) $D_1 = 3.83 \times 10^{-11}$ cm$^2/s$, $D_2 = 9.44 \times 10^{-12}$ cm$^2/s$, $D_3 = 1.88 \times 10^{-12}$ cm$^2/s$, Comparing the obtained coefficients (14) for rapid and slow components, we obtained the following excitation parameters: $\alpha_1 = 2.66 \times 10^{-4}$, $\alpha_2 = 1.6 \times 10^{-4}$, $\alpha_3 = 5.37 \times 10^{-4}$.

Now we can evaluate the last term in the theoretical profile (15): $0.125\xi_{\mathrm{e}}^2 = 0.031$ for the maximum experimental depth $x_{\mathrm{e}} = 12$ μm and $D_{\mathrm{ex},1}$. We see that this term is small. In this case the theoretical profiles, as well as the experimental profiles for the three fast components for the different diffusion conditions of indium atoms in the region $0 \le x \le x_{\mathrm{e}}$, $x_{\mathrm{e}} = 12$ μm, can be represented by straight lines

$$N_i(x,t) \approx N_{\mathrm{s}i}(1 - 0.4075\xi_{\mathrm{e}i}),\ i = 1,\ 2,\ 3,$$
$$N_{\mathrm{s1}} = 10^{17}\ \mathrm{cm}^{-3},\ \xi_{\mathrm{e}i} = \frac{x}{\sqrt{D_{\mathrm{ex},i}t_i}},\ t_1 = 40\ \mathrm{s},$$
$$N_{\mathrm{s2}} = 0.9 \times 10^{17}\ \mathrm{cm}^{-3},\ t_2 = 30\ \mathrm{s},\ N_{\mathrm{s3}} = 3 \times 10^{16}\ \mathrm{cm}^{-3},\ t_3 = 600\ \mathrm{s}, \quad (20)$$

which are in good agreement with the experiment [4]. The slow and rapid profiles are obtained when photons excite a part of diffusing indium atoms in HgCdTe crystal for a sufficiently long period of time.

The rapid vacancies in Si crystal were obtained by irradiation of the sample with soft X-rays [5]. The dependence of the measured conductivity for continuous current of the Si sample on the square root of irradiation time is presented in Fig. 1 and Fig. 2 at 22°C and 13°C. Voltage of the chromium anode was 9 kV and current in the tube was 23 mA in both cases.

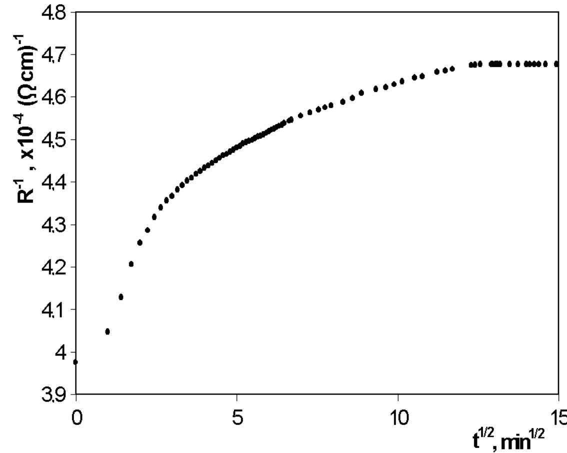

Fig. 1. Dependence of conductivity R^{-1} (Ω cm)$^{-1}$ on square root of the irradiation time t when sample temperature is 22°C.

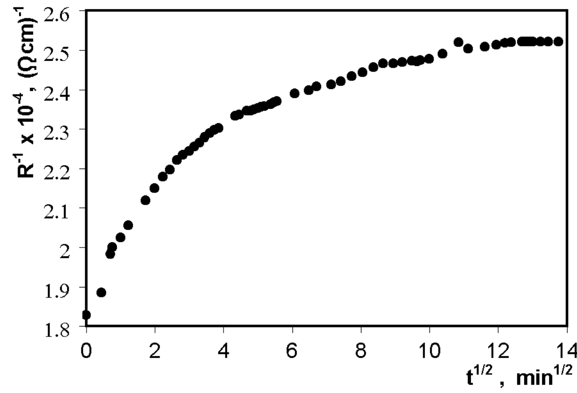

Fig. 2. Dependence of conductivity R^{-1} $(\Omega\ \mathrm{cm})^{-1}$ on square root of the irradiation time t when sample temperature is 13°C.

The number N_v of introduced negatively charged vacancies [8, 9] in the volume $V = 0.3 \times 0.3 \times 0.3\ \mathrm{cm}^3$ of Si crystal can be expressed in the following form:

$$N_\mathrm{v} = 0.274 N_\mathrm{s} x_{05}, \quad x_{05} = 1.6161\sqrt{Dt}. \tag{21}$$

Here, N_s is concentration of vacancies at the irradiated surface, $x_{05} = 0.3$ cm is the maximum vacancy penetration and D is the vacancy diffusion coefficient in Si crystal. We see that in this case the number of negatively charged vacancies [5] or concentration of holes in an Si crystal is proportional to the square root of irradiation time. From the conductivity dependence on square root of the irradiation time presented in Fig. 1 and Fig. 2 it follows that two kinds of vacancies are diffusing in the crystal, when the Cr anode voltage is 9 kV [10] and the Roentgen tube current is 23 mA. After irradiation time $t_1 = 13$ min at a temperature of 22°C, the crystal is saturated by fast singly charged vacancies, and by the time $t_2 = 160$ min the crystal is saturated by more slowly diffusing doubly negatively charged vacancies. At a temperature of 13°C, we obtain that the crystal was saturated by fast vacancies at 9 min, and it was saturated by slow doubly negatively charged vacancies at 160 min. Now, using (11) we obtain

$$\frac{D_-(22°\mathrm{C})}{\alpha_-(22°\mathrm{C})} = 4.418 \times 10^{-5}\ \mathrm{cm}^2/\mathrm{s}, \quad \frac{D_{--}(22°\mathrm{C})}{\alpha_{--}(22°\mathrm{C})} = 3.589 \times 10^{-6}\ \mathrm{cm}^2/\mathrm{s}, \tag{22}$$

$$\frac{D_-(13°\mathrm{C})}{\alpha_-(13°\mathrm{C})} = 6.381 \times 10^{-5}\ \mathrm{cm}^2/\mathrm{s}, \quad \frac{D_{--}(13°\mathrm{C})}{\alpha_{--}(13°\mathrm{C})} = 3.589 \times 10^{-6}\ \mathrm{cm}^2/\mathrm{s}. \tag{23}$$

Diffusion coefficient of vacancies obtained by thermal heating [11] is many times smaller

$$D_\mathrm{v} = (3\ \mathrm{cm}^2/\mathrm{s})\exp[-1.0\ \mathrm{eV}/kT] + (10^{-6}\ \mathrm{cm}^2/\mathrm{s})\exp[-0.2\ \mathrm{eV}/kT]. \tag{24}$$

Using this formula, we can calculate diffusion coefficients of vacancies obtained by thermal heating

$$D_{\mathrm{v}}(13°\mathrm{C}) = 2.981 \times 10^{-10}\ \mathrm{cm}^2/\mathrm{s},\ D_{\mathrm{v}}(22°\mathrm{C}) = 3.8192 \times 10^{-10}\ \mathrm{cm}^2/\mathrm{s}. \quad (25)$$

Taking into account (11) and (25), we obtain the following excitation parameters:

$$\alpha_{-}(22°\mathrm{C}) = 8.645 \times 10^{-6}, \quad \alpha_{--}(13°\mathrm{C}) = 4.672 \times 10^{-6}. \quad (26)$$

From (3) we can conclude that at the initial moment of time all charged vacancies obtained by the Auger effect are in excited states.

These fast excited vacancies can be used to obtain very fast impurity diffusion in silicon at room temperatures, like fast diffusion of indium atoms in the HgCdTe crystal, which was activated by tungsten–halogen ultraviolet radiation [6]. This opens the possibilities for new methods in nanotechnologies, where soft X-rays can be used.

We proposed an original method for the definition of vacancy diffusion coefficients. We obtained fast negatively charged vacancies using soft X-rays instead of 1–3 MeV electron irradiation which was generally used for this purpose [12]. There was confirmed that the movement energy of vacancies contained a part of energy of their producing. We also determined that diffusion coefficient of singly negatively charged vacancies increases with decreasing temperature (see (22), (23)), and the diffusion coefficient of doubly negatively charged vacancies is the same for temperatures 13°C and 22°C.

The negatively charged vacancies cause an increase in hole concentration Δp and hence an increase in conductivity [13]:

$$\Delta\sigma = \Delta p\ e\mu_p. \quad (27)$$

Using hole mobility [13] $\mu_p = 500\ \mathrm{cm}^2/(\mathrm{V\ s})$ and the expression [5, 8]:

$$\Delta p\ V = 0.2745 N_{\mathrm{s}} x_0 \quad (28)$$

for the number of vacancies introduced by diffusion into the volume $V = 0.3 \times 0.3 \times 0.3\ \mathrm{cm}^3$ of the crystal, when density of irradiated vacancies on the surface was N_{s}, in conjunction with the measurement results presented in Fig. 1 and Fig. 2, we obtained the following results for the temperature 13°C:

$$\Delta p_{-} = 3.745 \times 10^{11}\ \mathrm{cm}^{-3}, \quad N_{\mathrm{s}-} = 4.548 \times 10^{12}\ \mathrm{cm}^{-1},$$

$$\Delta p_{--} = 3.745 \times 10^{11}\ \mathrm{cm}^{-3}, \quad N_{\mathrm{s}--} = 2.274 \times 10^{12}\ \mathrm{cm}^{-1}, \quad (29)$$

and for the temperature 22°C:

$$\Delta p_{-} = 3.745 \times 10^{11}\ \mathrm{cm}^{-3}, \quad N_{\mathrm{s}-} = 4.548 \times 10^{12}\ \mathrm{cm}^{-1},$$

$$\Delta p_{--} = 3.745 \times 10^{11}\ \mathrm{cm}^{-3}, \quad N_{\mathrm{s}--} = 2.274 \times 10^{12}\ \mathrm{cm}^{-1}. \quad (30)$$

Those results indicate that the number of produced vacancies does not depend on temperature. Using the resistance of the sample $R_1 = 18240\ \Omega$, $R_2 = 8384\ \Omega$ at temperatures $T_1 = 286$ K, $T_2 = 295$ K, respectively, we obtained the forbidden gap width $\Delta E_g = 1.257$ eV.

The negatively charged vacancies behave like acceptors and increase electrical conductivity of silicon. The diffusion coefficients of singly charged vacancies ($D_- = 3.05 \times 10^{-6}$ cm^2/s) and doubly charged vacancies ($D_{--} = 1.47 \times 10^{-6}$ cm^2/s) at room temperature (about 290 K), which was obtained from Fig. 1 and (21). They are about 10^4 times larger than diffusion coefficients of vacancies obtained by thermal heating [11], and this fact can be explained by excitation of Si lattice. A very long duration (more than 400 min) of existence of the excited vacancies in Si crystal and their ability to move very fast opens up new possibilities for realization of technologies where X-rays can be used in production of new microprocessors and new devices. We can say that the predicted possibility of superdiffusion when the atoms that surround vacancies are in excited states [1] has been practically realized experimentally [5].

The presented model of diffusion in excited systems and experimental results [5, 9] can be used for the investigation of rapid diffusion in excited semiconductors. The proposed model can be also useful for the investigation of vacancy diffusion in excited crystals.

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