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THE NONLINEAR DIFFUSION
IN THE NONISOTHERMICAL CASE

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We will discuss the properties of the nonlinear equation in the nonisothermical case. In our previous papers the diffusion coefficient directly proportional to the concentration of the impurities was proposed and it was exactly defined. Now the nonlinear diffusion equation is solved for the temperature and for the diffusion coefficient depending on time in a special way. The temperature function $T(t)$ has the singularity at the free chosen time moment t_0. The obtained analytical solutions define the diffusion profiles for increasing temperatures and in the case of excited systems when the vacancies and the impurity's atoms are not in the thermal equilibrium with lattice. Considering the connection between temperature function and the population of excited states for atoms surrounding vacancies the possibility of the superdiffusion is shown.

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1. Introduction

The diffusion coefficient must equal zero in the region where the impurities are absent. In papers [1, 2] the nonlinear diffusion equation was proposed

$$\frac{\mathrm{d}}{\mathrm{d}t}N = D_n\frac{\mathrm{d}}{\mathrm{d}x}\left(N\frac{\mathrm{d}}{\mathrm{d}x}N\right), \tag{1}$$

where the diffusion coefficient and current density were proportional to the impurities concentration

$$D(t,N) = D_nN(x,t), \quad j = -D\frac{\mathrm{d}}{\mathrm{d}x}N, \tag{2}$$

where

$$D_n = \frac{D_0}{N_a}\mathrm{e}^{-E/kT}, \quad T(t) = \frac{1}{a - b\ln(tc)}, \tag{3}$$

a, b, c — free chosen constants. If $b = 0$ we have the isothermical diffusion. We got $N_a = N(0,t)$ after we considered the flux density on the frontier [3]

$$j = \frac{D_0\mathrm{e}^{-E/kT}}{\rho\left(x_0 - \frac{1}{2}\Delta\right)}\rho(x_0)\left(\frac{\mathrm{d}}{\mathrm{d}x}\rho\right)_{x_0}. \tag{4}$$

The diffusion coefficient in point x_0 must be proportional not only to the gradient of the impurity's in this point but also to the probability of presence of impurity's atom in this point before the elementary jump Δ.

2. Similarity solution of the nonlinear nonisothermical diffusion equation

Suppose that the impurity concentration satisfies the following boundary:

$$N(0,t) = N_s, \quad N(\infty,t) = 0 \tag{5}$$

and the initial condition

$$N(x,0) = 0. \tag{6}$$

Introducing the similarity variable

$$\xi = \frac{x}{\sqrt{D_s t}}, \quad D_s = D_n N_s \tag{7}$$

into Eq. (1) we can get the nonlinear differential equation [4]

$$2\frac{\mathrm{d}}{\mathrm{d}\xi}\left(f\frac{\mathrm{d}}{\mathrm{d}\xi}f\right) + \alpha\xi\frac{\mathrm{d}}{\mathrm{d}\xi}f = 0, \tag{8}$$

$$\alpha = 1 + \frac{Eb}{k}, \tag{9}$$

where we express the solution of the nonlinear equation (1) in the power series

$$N(x,t) = N_s f(\xi), \tag{10}$$

$$f(\xi) = \sum_{n=0}^{\infty} a_n \xi^n. \tag{11}$$

From the boundary condition (5) we obtain $a_0 = 1$. Substituting (10) and (11) into (8) we obtain the system of the equations

$$\begin{aligned} &2\sum_{m=0}^{n}(n+1-m)(m+1)a_{n+1-m}a_{m+1} + 2\sum_{m=0}^{n} c_{nm} + n\alpha a_n = 0, \\ &c_{nm} = (n+2-m)(n+1-m)a_{n+2-m}a_m, \quad n = 0,1,2,\ldots \end{aligned} \tag{12}$$

Taking in Eq. (11) a finite number of the coefficients and solving the system of Eqs. (12) we obtain the second series of approximate solutions

$$N_3 = N_s\left(1 - \frac{\sqrt{\alpha}}{\sqrt{5}}\xi - \frac{\alpha}{10}\xi^2 - \frac{\alpha\sqrt{\alpha}}{60\sqrt{5}}\xi^3\right), \quad 0 \le \xi \le \xi_{03},$$

$$\xi_{03} = \sqrt{\frac{5}{\alpha}}\left(20^{\frac{1}{3}} - 2\right), \quad x_{03} = 1.597\sqrt{\frac{D_s t}{\alpha}}, \quad 0 \le x \le x_{03}, \tag{13}$$

$$N_4 = N_s\left(1 - \frac{\sqrt{\alpha}\xi}{2.253} - \frac{\alpha\xi^2}{10.15} - \frac{\alpha\sqrt{\alpha}\xi^3}{148.1} + \frac{\alpha^2\xi^4}{2792}\right), \quad 0 \le \xi \le \xi_{04},$$

$$\xi_{04} = \frac{1.615}{\sqrt{\alpha}}, \quad x_{04} = 1.615\sqrt{\frac{D_s t}{\alpha}}, \quad 0 \le x \le x_{04}, \tag{14}$$

$$N_5 = N_s \left(1 - \frac{\sqrt{\alpha}\xi}{2.254} - \frac{\alpha\xi^2}{10.16} - \frac{\alpha\sqrt{\alpha}\xi^3}{149.9} + \frac{\alpha^2\xi^4}{2499} + \frac{\alpha^2\sqrt{\alpha}\xi^5}{46361}\right), \quad 0 \le \xi \le \xi_{05},$$

$$\xi_{05} = \frac{1.617}{\sqrt{\alpha}}, \quad x_{05} = 1.617\sqrt{\frac{D_s t}{\alpha}}, \quad 0 \le x \le x_{05}. \tag{15}$$

From the above expressions we can see that the power series converges very fast when

$$0 < \alpha \le 1, \quad -\frac{k}{E} < b \le 0. \tag{16}$$

We can express diffusion coefficient (3) in the other way

$$D_n(t) = \frac{D_0}{N_s} e^{-(E/k)a} (tc)^{(E/k)b}. \tag{17}$$

When $b = 0$ we have the isothermical diffusion and then a must be > 0. For $b \neq 0$ temperature's expression (3) has singularity for the time moment

$$t_0 = \frac{1}{c} e^{a/b}, \quad b \neq 0. \tag{18}$$

From (17) when $b > 0$ and $0 \le t < \infty$ we obtain $0 \le D_n(t) < \infty$. For $b < 0$ we get $\infty > D_n(t) \ge 0$ and have singularity for $D_n(t)$ in the initial time moment. In case $b > 0$ we have the following intervals for temperature:

$$0 \le T(t) < \infty, \quad 0 \le t < t_0, \tag{19}$$

$$-\infty < T(t) \le -0, \quad t_0 < t < \infty. \tag{20}$$

We can get into the situation $b < 0$ after heating the diffusion area by impulse laser or using other radiation with small absorption coefficients [5] for ground lattice and resonance absorption of excited vibration states for the atoms surrounding vacancies

$$-0 \ge T(t) > -\infty, \quad 0 \le t < t_0, \tag{21}$$

$$\infty > T(t) > 0, \quad t_0 < t < \infty. \tag{22}$$

The generation of vacancies in Si and Ge can be induced by 2 eV photons and the movement of vacancies in Ge can be activated by photons with energies 1.25 eV [6]. The negative absolute temperature expresses the ratio of the populations of two levels of vibration states of the atoms surrounding vacancies. When parameter α is very small the maximum penetration of impurities x_{05} (15) is very large. Condition (2) can be satisfied for increasing temperatures $T(t_1) < T(t_2) < T(t_3)$ in finite time interval $t_3 < t_0$. The three constants in temperature expression (3) can be obtained from any three different temperatures in the following manner:

$$a = \frac{1}{T(t_1)} + \frac{\frac{1}{T(t_1)} - \frac{1}{T(t_2)}}{\ln\left(\frac{t_2}{t_1}\right)} \ln(t_1, c), \quad b = \frac{\frac{1}{T(t_1)} - \frac{1}{T(t_2)}}{\ln\left(\frac{t_2}{t_1}\right)},$$

$$c = \frac{1}{t_3}\left(\frac{t_2}{t_1}\right)^{[1/T(t_1) - 1/T(t_3)]/[1/T(t_1) - 1/T(t_2)]}. \tag{23}$$

3. Superdiffusivity and diffusion in excited systems

We have superdiffusivity when α vanishes to zero

$$\lim_{\alpha\to 0} b = b_0, \quad b_0 = -\frac{k}{E}, \tag{24}$$

and the maximum penetration of impurities (15) in the crystal

$$x_{05} = 1.617\sqrt{D_0 e^{-(E/k)a}\ c^{(E/k)b}\,\frac{t^\alpha}{\alpha}} \tag{25}$$

tends to infinity.

Constants b_0 can be calculated using activation energies [7]. For phosphorus or arsenic diffusion in silicon we obtain

$$b_{0,\mathrm{P}} = -4.198\times 10^{-5}\ \mathrm{K}^{-1}, \quad b_{0,\mathrm{As}} = -2.396\times 10^{-5}\ \mathrm{K}^{-1}. \tag{26}$$

From (3) and (18) we can get a very useful expression

$$bT(t) = -\frac{1}{\ln z}, \quad t = zt_0, \quad 0 \le z \le \infty. \tag{27}$$

From (27), Fig. 1 and Fig. 2 we can see that the temperature changes abruptly when time varies from t_0 very slightly and it changes monotonously when $t > t_0$.

Superdiffusion in excited systems can be obtained when the temperature changes like in Fig. 2

$$T_0(t) = -\frac{1}{b_0 \ln z}, \quad t = zt_0. \tag{28}$$

Considering this problem we can use Boltzmann statistics for the definition of ratio of the populations of two quantum levels with energies $\varepsilon_2 > \varepsilon_1$

$$P = \frac{w(\varepsilon_2)}{w(\varepsilon_1)} = \exp\left(\frac{-E_{21}}{kT}\right), \tag{29}$$

where $E_{21} = \varepsilon_2 - \varepsilon_1$. When $P \ge 1$ we have $T < 0$.

Taking the temperature from (27) and substituting into (29) we obtain

$$P = z^{E_{21}b/k}. \tag{30}$$

For interpretation of expression (1) we can assume that the vacancy mechanism can be realized for the atoms with the energies many times lower than the activation energy. Now we can understand that when $b > 0$ we have the diffusion with the process of saturation. This situation, when $P = z$, is represented in Fig. 1. When all atoms surrounding vacancies are in excited vibration states ε_2 we have $P = z^{-1}$ and $b < 0$. This situation is represented in Fig. 2. From (29) and (30) it follows that superdiffusion is possible when

$$P = z^{-E_{21}/E}, \quad E_{21} \ge E. \tag{31}$$

In this case the vibration levels ε_2 of the diffusing atoms must be fully saturated in the initial time. The transition velocity to the ground states ε_1 is larger when

$$E_{21} \ge E. \tag{32}$$

From (29) and (30) we find that superdiffusion is possible when the surrounding vacancies atoms in initial time moment are in the excited vibration states.

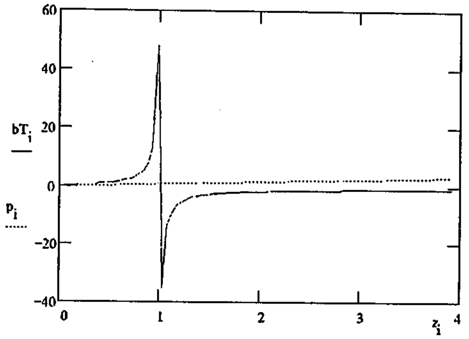

Fig. 1. Traces of dependence bT and P on time zt_0.

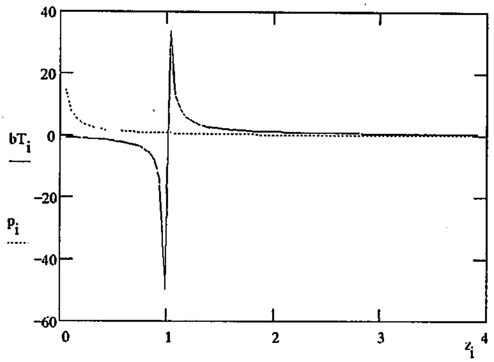

Fig. 2. Traces of dependence bT and P on time zt_0 in the excited systems.

For the practical calculations we can express maximum penetration of the impurities (17) using (27) in a very handy form

$$x_{05} = 1.617\sqrt{\frac{D_0 z^{\alpha}}{\alpha} t_0}. \tag{33}$$

Using (17), (27) we can express the similarity variable in a second way

$$\xi = \frac{x}{\sqrt{D_0 z^{\alpha} t_0}}. \tag{34}$$

The obtained expression will be useful when we calculate the profiles of impurities in the excited systems. From (34) and (15) we can obtain that the relaxation time τ is approximately equal to t_0. Considering the earlier obtained connection $N_a = N_s, D_s = D_n N_s$ and (30), (29), (7), (3) we have

$$D_s = D_0 z^{Eb/k}, \quad P = z^{E_{21} b/k}. \tag{35}$$

The obtained equations can be conformed only when

$$E_{21} = E. \tag{36}$$

Then the diffusion coefficient must satisfy the following connection:

$$D = D_0 P. \tag{37}$$

From (9) and (35) we can obtain

$$D_s(t) = D_0 P(t), \quad P(t) = z^{\alpha-1}, \quad t = z t_0. \tag{38}$$

In the case of superdiffusivity $\alpha \to 0$. When energy of excitation E_{21} equals activation energy E, the diffusion coefficient is directly proportional to the population of the excited state. This population can be increased using radiation of special frequency which can excite local vibration states [8] of the atoms surrounding vacancies with the sufficient energies for diffusion jumps.

When α is large and z is small the magnitude of excited state population is small and we have usual slow diffusion. When $\alpha < 1$ we have a very fast diffusion in the strong excited system in the initial time. The diffusion profile weakly depends on time like in Fig. 4.

4. Results of calculations

The nonisothermical phosphorus profile in silicon presented in Table is calculated for the increasing temperatures $T(t_1) = 300$ K, $t_1 = 3.446$ s, $T(t_2) = 1154$ K, $t_2 = 1373$ s, $T(t_3) = 1373$ K, $t_3 = 1500$ s. From (9), (14), (19), (23) and [7] we have obtained $b = 4.583 \times 10^{-4}$ K^{-1}, $E = -2.17$ eV, $\alpha = 12.53$, $c = 0.1960$ s^{-1}, $a = 3.153 \times 10^{-3}$ K^{-1}, $t_0 = 4966$ s, $N_s = 6 \times 10^{20}$ cm^{-3}, $D_s(t_3) = 10^{-11}$ cm^2/s, $x_{04} = 0.5537$ μm. The dependence of the temperature on time in the presented moments was approximated with the expression $T = nt^{\frac{1}{4}}$. In paper [3] there was calculated a profile for the isothermical phosphorus diffusion in the silicon when $T = 1373$ K, $t = 1500$ s. In this case the maximum penetration of phosphorus in the silicon 1.98 μm must be more than 0.5537 μm for increasing temperatures.

TABLE

Nonisothermical phosphorus profile calculated for the increasing temperatures $T(t_0) = 0$ K, $t_0 = 0$ s, $T(t_1) = 300$ K, $t_1 = 3.44$ s, $T(t_2) = 1154$ K, $t_2 = 1373$ s, $T(t_3) = 1373$ K, $t_3 = 1500$ s.

x [μm]	0	0.06	0.11	0.17	0.22	0.28	0.33	0.39	0.48	0.50	0.56
$N_4 \times 10^{-20}$ cm^{-3}	6	5.55	5.08	4.57	4.02	3.44	2.83	2.18	1.49	0.76	0.00

Using (34) and (15) we calculated the arsenic profiles for the diffusion in excited systems (35), (9) for $\alpha = 0.1$ and diffusion in exciting process when $\alpha = 1.9$. In both cases we have a very fast diffusion but in the first case it is much faster. The obtained results are presented in Fig. 3 and Fig. 4. When $\alpha = 0.01$ and diffusion time $t = 2 \times 10^{-8}$ s the maximum penetration depth is 9.12 μm. The same penetration depth in the nonexcited systems can be obtained for a very long diffusion time 2.659×10^7 s.

5. Conclusions

Diffusion equations (1), (3) describe diffusion and heat conductivity in the excited systems. In this case the properties of the materials heated with ultrashort laser pulse can be investigated.

When the frequency of local vibrations in solids for the atoms of impurities [8] does not lie in the region of the normal vibrations, we suppose that the

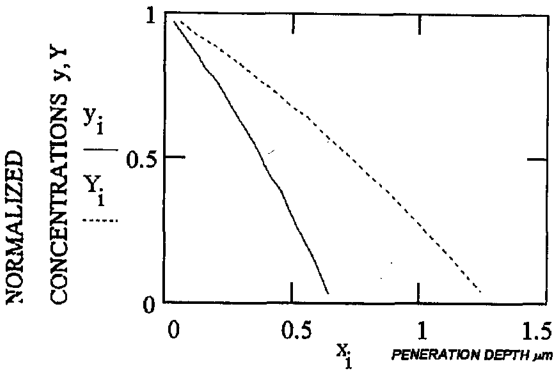

Fig. 3. Profiles of arsenic diffusion in silicon. $y = N(x)/N_s, \alpha = 1.9, z = 1, t_0 = 10^{-8}$ s, $D_0 = 0.32$ cm^2/s, $t = zt_0$; $Y = N(x)/N_s, \alpha = 1.9, z = 2, t_0 = 10^{-8}$ s, $D_0 = 0.32$ cm^2/s.

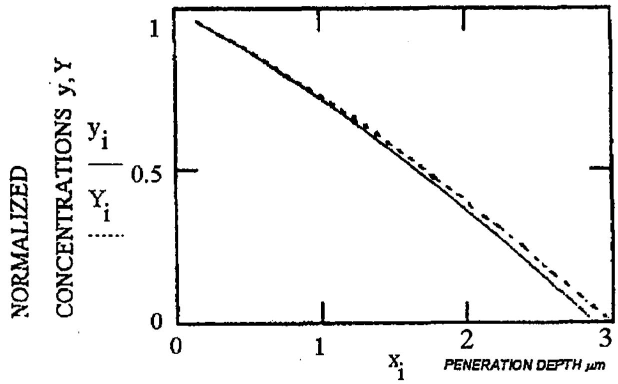

Fig. 4. Profiles of arsenic diffusion in silicon. $y = N(x)/N_s, \alpha = 0.1, z = 1, t_0 = 10^{-8}$ s, $D_0 = 0.32$ cm^2/s, $t = zt_0$; $Y = N(x)/N_s, \alpha = 0.1, z = 2, t_0 = 10^{-8}$ s, $D_0 = 0.32$ cm^2/s.

resonance absorption can be realized and a very fast diffusion (24), (25) can be obtained. From the profiles presented in Fig. 4 we can see that when $b < 0$ and $\alpha = 0.1$ the maximum penetration of impurities weakly depends on the time of diffusion when $t > 0.1t_0$. This situation shows some diffusion inertia and coincides with the thermal inertia and localization of the thermal excitation during fast laser heating of solids [9]. When the heat is transported via the electron gas, which is out of equilibrium with the lattice, the effects of ultrafast heat transport can be observed [10, 11]. We can assume that the ultrafast diffusion can be realized when the vacancies and the impurities atoms will be out of equilibrium with the lattice. They can be excited by beam of 0.1–0.4 MeV electrons or heated by ultrashort laser radiation pulses with the resonance absorption. The conditions for ultrafast diffusion will depend on the manner of diffusion. From (38) we can see that the velocity of diffusion did not depend on the way of population of excited vibration states of atoms surrounding vacancies whose energies must be approximately equal to activation energy. We have the very fast diffusion when in initial time all vacancies are excited and diffusion coefficient tends to infinity. We must initiate the very fast vacancy's movement and its generation.

In paper [12] the picosecond dynamics of nonequilibrium charge carriers the large average ambipolar diffusion coefficient values were obtained. The higher values of the ambipolar diffusion coefficient for hot carriers are larger than for thermalized carriers. This can be explained as the diffusion in excited system.

Rapid thermal diffusion of indium in HgCdTe sample activated by tungsten--halogen radiation was described in paper [13]. The penetration depth of the fast component weakly depends on time. The slow atomic diffusion can occur through nonexcited vacancies.

The real conditions for superdiffusion will be more complicated than the above considered conditions.

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