

ON THE KINETICS OF GROWTH OF CONDUCTIVITY ELECTRON CONCENTRATION IN DIELECTRIC CRYSTAL IRRADIATED BY INTENSIVE LIGHT*

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The kinetics is studied of electron avalanche in insulating crystals under the influence of intense light beams. To obtain solutions of kinetic equations, the method of successive approximations is developed. The multistage character of optical damage in insulators is discussed.

1. Introduction

It is well known that the electronic structure of dielectric crystals is such that there are no electrons in the conduction band, which is detached from a completely filled valence band by an energy gap ΔE . Let us suppose that such a crystal is illuminated by a beam of light with frequency $\omega < \Delta E/\hbar$, where \hbar is Planck's constant. If the problem of interaction between the beam of light and the crystal is treated in linear approximation in respect of the intensity of light, then the pure dielectric crystal must not absorb light at all. At the same time there is experimental evidence that laser radiation can give rise to a damage of a transparent insulator if the intensity of light is high enough [1].

One of the possible mechanisms of the optical damage of dielectric crystals is a development of conductivity electron avalanche [1—6]. This process can be imagined as follows. As a consequence of a multiphoton absorption or thermal fluctuations a few electrons appear in the conduction band. Absorbing photons from the incident radiation these primary electrons can gain an amount of energy greater than the minimum energy I which is necessary for impact ionization. The concentration of conductivity electrons increases as a consequence of their multiplication and after all that results in crystal damage.

In theoretical papers dedicated to the avalanche development the concentration of the conductivity electrons $n(t)$ is usually represented as $n_0 e^{\gamma t}$ and the avalanche development rate γ is actually calculated with small values of $n(t)$. As a consequence, nonlinear in $n(t)$ effects remain in the background.

In the present paper the kinetics of the electron avalanche is studied in the case of intensity of incident light close to the damage threshold with due regard to the above

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mentioned nonlinear effects. The frequency of light is supposed to belong to the visual range. The following physical effects which are nonlinear in concentration of conductivity electrons are taken into account. Firstly, the excitation of vibratory modes of the crystal by conductivity electrons and holes should be taken into consideration if only this process can cause melting of some portion of the crystal. In its turn, this melting is an important intermediate stage of the damage process as a whole.

Secondly, at definite values of $n(t)$ ($\sim 10^{15} \text{ cm}^{-3}$) electron—electron collisions become essential [7]. Such collisions result in a redistribution of energy between electrons and, in consequence, an electron distribution function approximates the Boltzmann one.

Finally, the recombination processes should be taken into account. They reduce the concentrations of electrons and holes. In absence of recombination processes the optical damage of transparent pure crystals has actually no intensity threshold provided that the pulse duration of the incident light is unlimited.

The direct radiative electron—hole recombination cannot apparently be an effective mechanism of reduction of the charge carrier concentration because the cross-section of this transition is too small ($\sim 10^{-19} \text{ cm}^2$). The latter can be found from the experimental data on light absorption in crystals making use of the relation of van Roosbroek and Shockley [8]. But recombination transitions can also occur through intermediate states (excitons in the present case). The appropriate elementary process is binding of an electron and a hole in an exciton [9—11]. It has a rather large cross-section $\sigma_{\text{ex}} \sim 10^{-13} \text{ cm}^2$. The exciton loses energy when scattering on phonons and at length transforms into a photon [11, 12] that leaves the crystal. This process proves to be an effective mechanism of decrease of charge carrier concentration in spite of the competition of a reverse process. This is exciton dissociation to an electron and a hole due to its interaction with a phonon [9].

In Section 2 we present the approximate method of solution of kinetic equations for electron and phonon subsystems of the crystal. That is the method of successive approximations. In Section 3 we shall briefly discuss the multistage character of optical damage of dielectric crystals.

2. Approximate solution of kinetic equations

Let us consider a dielectric crystal in which conduction and valence bands are the standard parabolic bands. We assume also that $m_e \ll m_h$, where m_e and m_h are effective masses of a conductivity electron and a hole, respectively. Suppose that the crystal is irradiated by the light beam, starting from the time moment $t = 0$. We shall describe the plane light wave propagating in the crystal by the vector potential

$$\mathbf{A} = A_0 \mathbf{a} \cos(\mathbf{qr} - \omega t),$$

where A_0 is the amplitude of the potential, \mathbf{a} is the unit polarization vector,

$\mathbf{E} = -\frac{\partial \mathbf{A}}{\partial t}$. Coupling of this wave with conduction electrons can be characterized by the dimensionless parameter $y = (\hbar\omega m_e)^{-1} e^2 A_0^2$, e is the electron charge. We shall consider such intensities of light when $y \ll 1$.

As far as absorption and emission of visible-light photon by a conductivity electron have quantum nature we shall use quantum theory, proceeding from the Hamiltonian $H = H_0 + H_{\text{int}}$. Here H_0 is the Hamiltonian of noninteracting quasi-particles (electrons, holes, phonons and photons), and H_{int} describes the Coulomb interaction between charge carriers, coupling of electrons with photons and phonons, and anharmonic phonon—phonon interaction.

Kinetic equations for occupation numbers of electrons, holes, and phonons, averaged with crystal density matrix $\hat{\rho}$ such as $n_{\mathbf{k}}(t) = \text{Tr}(\hat{\rho}(t)\hat{n}_{\mathbf{k}})$, (master equations) can be obtained within the framework of nonequilibrium quantum statistical mechanics with the aid of Kadanoff—Baym equations and summing up the secular terms in perturbation theory series [13, 14]. Here Tr denotes the trace operation. The problem under consideration is rather specific: there are some small parameters and, besides, absorption and emission of a photon by a conductivity electron must be accompanied by absorption or emission of a phonon to obey the conservation laws of energy and quasi-momentum.

In the course of avalanche development the Fermi gas of conductivity electrons remains nondegenerate. In this case the master equation for electron occupation probabilities has the form

$$\frac{\partial f_e(\mathbf{p}, t)}{\partial t} = (\hat{S}_{ph} + \hat{S}_r + \hat{S}_i + \hat{S}_{ex} + \hat{S}_c) f_e(\mathbf{p}, t). \quad (1)$$

where $f_e(\mathbf{p}, t)$ is the conduction electron distribution function and \hat{S} are transition probabilities. \mathbf{p} is electron quasi-momentum. Distribution function of holes in the valence band $f_h(\mathbf{p}, t)$ satisfies the analogous equation. The terms $\hat{S}f$ have the following common structure. Each of them is the sum of products of the normalizing factor, modulus of the matrix element squared, deltafunction standing for energy conservation law in the elementary process, electron and hole distribution functions in the initial states, and factor $N_{\mathbf{k}}(t)$ or $N_{\mathbf{k}}(t) + 1$ in the case of absorption or emission of phonon. It is too cumbersome to write down the explicit expressions here and we shall refer the readers to the relevant literature. It is essential that in the case of nonpolar crystals the interaction of electrons with acoustic and optical phonons (scattering on the acoustic and optical deformation potentials) is taken into consideration in H_{int} . If the crystal has polar optical vibratory modes, we suppose that their interaction with conductivity electrons is weak. It is also taken into account in H_{int} .

The term $\hat{S}_{ph} f_e$ describes phonon absorption and emission by conductivity electrons [7, 15]. Absorption of photons from the light beam by electron and photon

emission in the same mode with simultaneous scattering on phonons is presented by $\hat{S}_r f_e$ [15]. Due to $y \ll 1$ we restrict ourselves to one photon processes. The term $\hat{S}_i f_e$ describes the impact ionization of electron and hole both with [3] and without photon absorption [16, 17]. The term $\hat{S}_{ex} f_e$ corresponds to the binding of electrons and holes in excitons [9] and $\hat{S}_c f_e$ takes into consideration electron—electron and electron—hole collisions in Born approximation [14].

Strictly speaking, Eq. (1) should be supplemented by the kinetic equation for excitons in which the following processes are taken into account: exciton dissociation, exciton—phonon scattering and luminescence of excitons (polaritons) [11, 12]. We shall consider these processes in a phenomenological way multiplying σ_{ex} by a coefficient $A < 1$. Accordingly, Eq. (1) contains an effective probability of the disappearance of electron and hole as a final result of the binding process.

Kinetic equations for the average numbers of phonons $N_{\mathbf{k}j}(t)$ of the mode j with quasi-momentum \mathbf{k} can be written as

$$\frac{\partial N_{\mathbf{k}j}(t)}{\partial t} = (\hat{L}_a + \hat{L}_e)N_{\mathbf{k}j}(t). \quad (2)$$

Here $\hat{L}_e N_{\mathbf{k}j}(t)$ represents the variation of $N_{\mathbf{k}j}(t)$ with time as a result of phonon—phonon interaction, and $\hat{L}_a N_{\mathbf{k}j}(t)$ describes the phonon absorption and emission in the course of the above electronic transitions [7, 15].

A kinetic equation such as (1) was considered in [3] with some difference: the last two terms on the r.h.s. of Eq. (1) were omitted and, accordingly, the above mentioned nonlinear effects were not considered. For the approximate solution of the kinetic equation the method of slowly varying amplitudes was useful in [3]. It had been worked out earlier in the paper [18] where the problem of gas breakdown by laser radiation was studied. In [3] the function $f_e(\mathbf{p}, t)$ was presented as $\exp(\gamma t)f(\mathbf{p})$. In doing so the impact ionization was considered as a slow process in contrast to the quick establishment of energy distribution in the electron system.

We intend to solve the system of equations (1), (2) at light intensity values close to the threshold of optical damage. These equations are nonlinear: operators \hat{S} depend on $N_{\mathbf{k}j}(t)$, \hat{L}_e contains $f_{e,h}(\mathbf{p}, t)$ and, besides, the last two terms on the r.h.s. of (1) are the bilinear expressions in respect of $f_{e,h}$. Keeping in mind the arguments given in the Introduction we shall generalize the method of slowly varying amplitudes and look for an approximate solution of (1) in the form $n(t)\tilde{f}_e(\mathbf{p}, n(t))$. Here $n(t)$ is the concentration of conductivity electrons at the time t and the function $\tilde{f}_e(\mathbf{p}, n(t))$ satisfies

$$(\hat{S}_{ph} + \hat{S}_r + \hat{S}_c)\tilde{f}_e(\mathbf{p}, n(t)) = 0. \quad (3)$$

Then Eq. (1) reduces to

$$\frac{dn(t)}{dt} = a(t)n(t) - b(t)n^2(t), \quad (4)$$

where $a(t)$ and $b(t)$ $n(t)$ are the probabilities of impact ionization and electron—hole binding in an exciton averaged with $\tilde{f}_e(\mathbf{p}, n(t))$ and the respective function for holes $\tilde{f}_h(\mathbf{p}, n(t))$.

The system of equations (2)—(4) is still too complicated to be solved exactly. As it was shown in [14], the conservation laws should be taken into consideration when looking for approximate solutions of kinetic equations, i.e. the approximate solutions must satisfy them. When solving (2)—(4) we propose to use the method of successive approximations. We obtain the first approximation $f_{e,h}^{(1)}(\mathbf{p}, t)$, if we put in (3)—(4) $N_{\mathbf{k}j}(t)$ to be equal to their values at $t=0$. The latter are determined by Planck's formula for the initial temperature T_0 of the crystal. Then we put the solutions of the resulting equations into Eq. (2) and find the time dependent average occupation numbers of phonons $N_{\mathbf{k}j}^{(1)}(t)$. Now we can write down equations for $f_{e,h}^{(2)}(\mathbf{p}, t)$ by substituting $N_{\mathbf{k}j}^{(1)}(t)$ in (3), (4) for $N_{\mathbf{k}j}(t)$ and so on. It is obvious how to continue the iteration procedure.

If this procedure is quickly convergent, we shall have a rather precise solution after a few steps already. In any case, even $N_{\mathbf{k}j}^{(1)}(t)$ reveals crystal lattice heat-up produced by conduction electrons and holes. And in $f_{e,h}^{(2)}(\mathbf{p}, t)$ the reverse influence of this heat-up on the distribution functions of charge carriers is duly taken into consideration.

The main obstacle to the solution of Eq. (3) consists in the quantum nature of absorption and emission of light by electrons: as a result of these transitions the electron energy $\varepsilon(\mathbf{p})$ changes by a quantity $\sim \hbar\omega$ which is commensurable with the impact ionization threshold I . Making use of the transition probabilities averaged on polarization directions of the incident light we shall look for the solution of Eq. (3) holding that $\tilde{f}_e(\mathbf{p}, n(t))$ depends on $|\mathbf{p}|$ only. Then this solution can be expanded in terms of the complete orthonormal set of functions $\varphi_n(x) = L_n(x) \exp\left(-\frac{x^2}{2}\right)$ as follows

$$\tilde{f}_e(\mathbf{p}, n(t)) = \sum_{n=0}^{\infty} c_n(t) \varphi_n(x), \quad x = \sqrt{\frac{|\mathbf{p}|^2}{m_e k T_e}}. \quad (5)$$

Here $L_n(x)$ are Laguerre polynomials, and k is the Boltzmann constant. The advantage of such an expansion consists in the fact that $\varphi_0(x) = \exp[-|\mathbf{p}|^2/2m_e k T_e]$. At $n(t) > n_c \sim 10^{15} \text{ cm}^{-3}$ collisions between electrons will result in the redistribution of energy and all $c_n(t)$, besides $c_0(t)$, will become zero. Also the expansion (5) contains a free parameter, that is the electronic temperature T_e . Its value can be determined in such a way that the energy conservation law is fulfilled for the approximate solutions.

Let us find out an approximate distribution function of conductivity electrons by putting in (5) all $c_n(t)$, besides $c_0(t)$, to be equal to zero for all time values. Note that we can look for corrections to such an approximation also with the aid of the expansion (5).

In the course of the iteration process we shall determine a value of $T_e(t)$ from the energy balance condition: the energy $n(t) \left\langle -\frac{d\varepsilon}{dt} \right\rangle$ lost in unit of volume by conductivity

electrons per unit time must be equal to the energy SK obtained by them in light absorption and emission processes. Here S is the intensity of incident light and K is the light absorption coefficient. In the case of nonpolar crystals this condition is

$$-\left\langle \frac{d\varepsilon}{dt} \right\rangle_a - \left\langle \frac{d\varepsilon}{dt} \right\rangle_{od} = \frac{1}{n(t)} S(K_a + K_{od}). \quad (6)$$

Here the quantities averaged with the function φ_0 appear and

$$-\left\langle \frac{d\varepsilon}{dt} \right\rangle_a = \frac{8\sqrt{2} E_1^2 m_e^{5/2}}{(\pi)^{3/2} \hbar^4 \rho} (kT_e)^{3/2} \left(1 - \frac{T}{T_e}\right).$$

Here $T(t)$ and ρ are the temperature and the density of the crystal, respectively, E_1 is the deformation potential constant [7]. Also

$$-\left\langle \frac{d\varepsilon}{dt} \right\rangle_{od} = \frac{m_e^{3/2} D^2 \sqrt{\hbar\omega_0} \sqrt{\lambda z} K_1(\lambda z)}{\pi^{3/2} \hbar^2 \rho \operatorname{sh}(z)} \operatorname{sh}[(1-\lambda)z],$$

where ω_0 is the maximum frequency of optical phonon, $\lambda = T/T_e$, $z = \frac{\hbar\omega_0}{2kT}$, and D is the optical deformation potential constant [15]. From now on $K_n(x)$ is the modified Bessel function, $n=0, 1, 2$. The contribution to the right hand side of Eq. (6) from the above mentioned radiative transitions accompanied by absorption or emission of an acoustical phonon is given by

$$\frac{1}{n(t)} SK_a = A_0^2 \omega C (2m_e \lambda kT)^{1/2} \operatorname{sh}\left(\frac{\hbar\omega}{2kT_e}\right) K_2\left(\frac{\hbar\omega}{2kT_e}\right),$$

where $C = e^2 E_1^2 (3\pi^{3/2} \hbar \rho u_e^2)^{-1}$ and u_e is the sound velocity in the crystal [15]. This is absorbed radiation energy attributed to one electron. The same quantity in the case when an optical phonon is emitted or absorbed is [15]

$$\frac{SK_{od}}{n(t)} = A_0^2 \omega^{-1} B (2kT_e)^{3/2} \frac{\operatorname{sh}(z_+ - z) z_+^2 K_2(z_+) + \operatorname{sh}(z_- + z) z_-^2 K_2(|z_-|)}{\operatorname{sh}(z)},$$

where

$$B = \frac{e^2 D^2 m_e^{1/2}}{6\pi^{3/2} \hbar^4 \rho \omega_0}, \quad z_{\pm} = \frac{\hbar\omega \pm \hbar\omega_0}{2kT_e}.$$

Eq. (6) defines T_e as a function of A_0^2 and T .

Let us turn our attention to polar insulating crystals. If the dimensionless polar constant $d_p \ll 1$, then T_e can be found from the energy balance condition

$$-\left\langle \frac{d\varepsilon}{dt} \right\rangle_a - \left\langle \frac{d\varepsilon}{dt} \right\rangle_{od} - \left\langle \frac{d\varepsilon}{dt} \right\rangle_{op} = \frac{1}{n(t)} S(K_a + K_{od} + K_{op}). \quad (7)$$

Here [15]

$$-\left\langle \frac{d\varepsilon}{dt} \right\rangle_{op} = \frac{2^{3/2} dp (\hbar\omega_0)^2 \sqrt{\lambda z} \operatorname{sh}[(1-\lambda)z] K_0(\lambda z)}{\pi^{1/2} \hbar} \frac{1}{\operatorname{sh}(z)},$$

$$\frac{1}{n(t)} SK_{op} = A_0^2 \frac{dpe^2 (\hbar\omega_0)^{3/2}}{3\pi^{1/2} \hbar^2 \omega m_e} (kT_e)^{1/2} [\operatorname{sh}(z)]^{-1} \times$$

$$\times [\operatorname{sh}(z_+ - z)z_+ K_1(z_+) + \operatorname{sh}(z_- + z)|z_-| K_1(|z_-|)].$$

The analogous equations determine a temperature of holes (we remind that $m_e \ll m_h$).

The first approximation of the iteration method can be obtained if we put $T = T_0$ in Eqs. (6), (7). It is obvious that quantities $T_e^{(1)}$ and $T_h^{(1)}$ determined in such a way are time-independent. Now we can evaluate the coefficients a and b in Eq. (4). In doing so we shall average the probabilities of impact ionization $w_0(\varepsilon)$ and $w_1(\varepsilon)$ (without the participation of photons [16, 17] and with the absorption of one photon [3], respectively) and the quantity $|\mathbf{v}_e - \mathbf{v}_h| \sigma_{ex}$ with functions $\tilde{f}_{e,h}^{(1)}(\varepsilon)$ (\mathbf{v}_e and \mathbf{v}_h are velocities of electron and hole, respectively)

$$a = \langle w_0(\varepsilon) + w_1(\varepsilon) \rangle, \quad b = \langle |\mathbf{v}_e - \mathbf{v}_h| \sigma_{ex} \rangle A.$$

The solution of Eq. (4) is ($n_0 = n(0)$)

$$n_1(t) = \frac{n_0 a \exp(at)}{a - n_0 b + n_0 b \exp(at)}. \quad (8)$$

The asymptotic behaviour of (8) is $n_1(t) \rightarrow a/b$, when $t \rightarrow \infty$. Turning to Eq. (2) we shall bear in mind that as a result of the anharmonic interaction the energy received by a phonon subsystem from conductivity electrons and holes tends to be quickly redistributed between the vibratory modes. Accordingly, $N_{\mathbf{k}j}(t)$ will be given by Planck's formula with time-dependent temperature $T(t)$, which in turn satisfies

$$\frac{dT(t)}{dt} = \frac{n(t)}{c_v} \left[-\left\langle \frac{d\varepsilon_e}{dt} \right\rangle - \left\langle \frac{d\varepsilon_h}{dt} \right\rangle \right], \quad (9)$$

where c_v is the specific heat of solid, $T(0) = T_0$ and the magnitude $-\left\langle \frac{d\varepsilon_e}{dt} \right\rangle$ is given by

the left hand side of Eq. (6) or Eq. (7). Let us substitute $n_1(t)$ given by Eq. (8) for $n(t)$ in Eq. (9) and integrate. The result is

$$T(t) = T_0 + (c_v b)^{-1} \ln \left[\frac{b}{a} n_0 (e^{at} - 1) + 1 \right] \left\{ - \left\langle \frac{d\varepsilon_e}{dt} \right\rangle - \left\langle \frac{d\varepsilon_h}{dt} \right\rangle \right\}. \quad (10)$$

The second approximation for $f_e(\mathbf{p}, t)$ can be obtained by substitution of Eq. (10) for T in (6), (7). By solving the equations we find the time-dependent quantity $T_e(t)$. Also the coefficients a and b in (4) will be time-dependent. Due to inequality $\frac{d}{dt} \left[\ln \left(\frac{n_0 b(t)}{a(t)} \right) \right] \ll a(t)$ the approximate formula can be obtained which describes the time-dependence of conductivity electron concentration with due regard to crystal lattice heat-up

$$n_2(t) = n_0 \frac{a(t) \exp \left(\int_0^t a(\tau) d\tau \right)}{a(t) - n_0 b(t) + n_0 b(t) \exp \left(\int_0^t a(\tau) d\tau \right)}. \quad (11)$$

Note that $n(t)$ can also be given by (11) in such a case when light intensity depends on time.

Now we shall give some numerical estimates making use of the parameter values which are characteristic of dielectrics with wide forbidden band: $m_e \sim 10^{-27}$ g, $u_e \sim 7 \cdot 10^5 \frac{\text{cm}}{\text{s}}$, $\hbar\omega_0 \sim 0,1$ eV, $I \approx 6$ eV, $E_1 \approx 11$ eV (these values refer to sapphire), $D = 5 \cdot 10^8 \frac{\text{eV}}{\text{cm}}$. At $T = 300$ K we find from Eq. (6) that $kT_e \sim 0,5$ eV corresponds to vacuum electrical field strength in the incident beam of order $10^5 \frac{\text{V}}{\text{cm}}$. We shall assume that at $\varepsilon > I$ $w_0(\varepsilon)$ linearly depends on their difference [16] as $r(\varepsilon - I)$. In this case $\langle w_0(\varepsilon) \rangle \approx \frac{2r}{\sqrt{\pi}} (kT_e I)^{1/2} \exp(-I/kT_e)$. If $r \sim 0,5 \cdot 10^{14} \text{ eV}^{-1} \text{ s}^{-1}$ then $a \sim 10^{10} \text{ s}^{-1}$. And at $A\sigma_{\text{ex}} \sim 10^{-14} \text{ cm}^2$ we obtain $n(\infty) \sim 10^{16} \text{ cm}^{-3}$.

At the initial stage of avalanche development in alkali halide crystals our treatment cannot be used directly because the polar constant $\alpha_p > 1$. It can be considered only as a qualitative one. But at high values of $n(t)$ the electron—phonon interaction is screened, i.e. $\alpha_p = \alpha_p(n)$ [19]. Owing to this, we can use our treatment starting from such values of $n(t)$ when $\alpha_p(n(t)) < 1$. Also we must assign initial conditions $n(t_0) = n_0$ at some moment $t_0 > 0$. However, some modifications are necessary. The first approximation for $n(t)$ is given by a formula like (11) where now $a(t)$ and $b(t)$ must be determined in the self-consistent way. And $T(t)$ takes approximately the form of Eq. (10) with the substitution of e^{at} by $\exp \left(\int_{t_0}^t a(\tau) d\tau \right)$. Note that the same formula with $t_0 = 0$ describes $T(t)$ in the case of time-varying light intensity.

3. Multistage character of optical damage process

In experiments on optical damage only some part of the crystal is usually under the influence of intensive light. If use is made of the focussed light beams, then it is the caustic region. In such a case the results of Section 2 should be referred to such a small region only.

Here we want to lay emphasis on the fact that laser damage in insulating transparent crystals has multistage character, although it is a fleeting phenomenon. The stages differ from each other both by the phase state of matter in the irradiated region and the light absorption mechanism and they are dominant physical processes. The electron avalanche development discussed in Section 2 can be considered as the first stage of the entire optical damage process. In due time the irreversible variations take place in the crystal at high values of $n(t)$. Let t_1 be the solution of equation $T(t) = T_m$, where T_m is the melting temperature of the crystal and $T(t)$ is given by (10). Then at $t > t_1$ the crystal melts in the above small region. There is, at the same time, a considerable concentration of free charge carriers (electrons and holes).

The arrangement of ions in the liquid state has been described by Bernal as "random close packing". Therefore the localization of some amount of valency electrons on the outer shells of ions, as it was in the crystalline state, becomes energetically unfavourable. It seems reasonable to suppose that in a small part of the solid a new, e.g. a liquid metal phase appears, with the free charge carriers concentration of the order of $10^{20} - 10^{21} \text{ cm}^{-3}$. This results in an ever increasing absorption of incident light (photon absorption by the electron is accompanied by electron—ion scattering). The temperature in the considered region also increases quickly.

The next stage of optical damage consists in the formation of electron—ion plasma whose temperature is fairly high and the density of free electrons is $\gtrsim 10^{23} \text{ cm}^{-3}$. At this stage the continuous radiation is emitted from the crystal [20, 21]. Its origin is probably connected with electron bremsstrahlung and recombination radiation. The experimental study of spectrum of radiation emitted from alkali halide crystals in the course of their laser damage gives the value of plasma temperature up to 27 000 K [20, 21]. Again the lines of doubly ionized aluminium were identified when the damage region in ruby was investigated spectroscopically [21].

The volume filled with plasma increases as a consequence of the energy transfer from plasma particles to ions of crystal phase on the interface. The low heat conductivity of the insulating crystal results in the onset of considerable temperature gradients in the frontier of the crystal, inducing thermal stress [22].

When the thermal stress reaches its critical value, the stress crack arises in the cleavage plane of the crystal. The crack is developing, while the electron—ion plasma is penetrating into its cavity. The total absorption of light falls. A movement of the crack front is accompanied by the same processes as in the case of the brittle failure under

mechanical influence: the electron emission from the newly formed solid surfaces [23], the triboluminescence [24], and the radio-frequency radiation [25].

The damage process ends with the thermochemical processes in the damage region: plasma cools down and its matter precipitates on the new solid surfaces. The precipitation is accompanied by considerable crystal disordering in close proximity to the above surfaces (formation of vacancies, interstitial atoms and so on). This supposition can be indirectly corroborated by the coloration that takes place in the alkali halide crystals near the zone of fracture [26].

4. Discussion

When the kinetic equation is solved in the diffusive approximation upon the energy variable [2, 4, 5], then function $f(\varepsilon + \hbar\omega)$ is substituted by $f(\varepsilon) + \hbar\omega f'(\varepsilon) + \frac{(\hbar\omega)^2}{2} f''(\varepsilon)$ for all values of ε . This is equivalent to the neglect of all the other terms in the Taylor expansion. In order to clarify the question for what types of functions $f(\varepsilon)$ this procedure is legitimate, we shall write down the remainder of Taylor series in the Lagrange form as $R_2 = \frac{(\hbar\omega)^3}{3!} f'''(\varepsilon + \theta\hbar\omega)$, where $0 < \theta < 1$. Thus the terms neglected in the diffusive approximation are negligible indeed if the inequality

$$\max |f'''(\varepsilon)| \ll \frac{3!}{(\hbar\omega)^2} \left| f'(\varepsilon) + \frac{\hbar\omega}{2} f''(\varepsilon) \right|$$

is valid for all energy values. On the left side here there is a maximum value of $|f'''(\varepsilon)|$ within the energy interval under consideration. If the photon energy $\hbar\omega > 1.5$ eV, it is comparable to the impact ionization threshold I . Then in interval $0 < \varepsilon < I$ the inequality is satisfied only by functions $f(\varepsilon)$ obeying $f'''(\varepsilon) \approx 0$, i.e. $f(\varepsilon) \approx c_1 + c_2\varepsilon + c_3\varepsilon^2$. Again the energy balance condition must be kept in mind. For its fulfilment the average number of electron transitions per unit time, with an incident photon absorbed, must exceed the average number of transitions with induced photon emission. This rules out functions $f(\varepsilon)$ monotonically increasing with the growth of energy.

As soon as $n(t)$ reaches the values when electron—electron collisions become essential, function $f(\varepsilon)$ approximates to the Boltzmann distribution function. Then the above inequality takes the form $\frac{2}{3} \left(\frac{\hbar\omega}{2kT_e} \right)^2 \ll \left| \frac{\hbar\omega}{2kT_e} - 1 \right|$. It is not fulfilled if the intensity of light is close to its damage threshold value and $\hbar\omega > 1.5$ eV.

In this paper heat diffusion from the part of the crystal which is exposed to the incident radiation was left out of account. It is of importance at $t > \tau \sim 10^{-3}$ s. If the asymptotic value of $n(t)$ does not cause irreversible variations in the crystal at $t < \tau$, then at $t > \tau$ some quasi-stationary magnitude of T is set in. In fact, it does not differ much

from T_0 . Photoconductivity and exciton luminescence can be recorded in these subthreshold conditions.

The plasma frequency $\omega_p = \left(\frac{4\pi n(t)e^2}{m_e} \right)^{1/2}$ can also be evaluated by means of the expressions obtained here. A discovery of light scattering on plasma oscillations at subthreshold intensities could bear out the electron avalanche mechanism of optical damage in insulating crystals.

The authors consider it an honour to be able to publish this paper in the special issue of the Journal dedicated to the 70th birthday of Professor I. Tarján, who has made important contributions to the physics of dielectric and, particularly, ionic crystals. We are happy to mention that our research cooperation with Professor I. Tarján has been in progress for more than two decades.

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