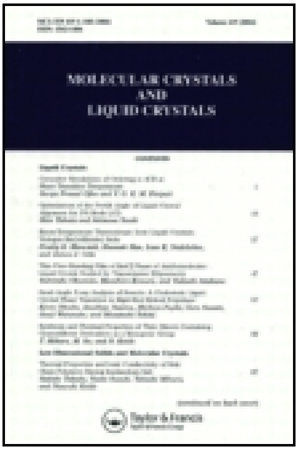

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Radioluminescence Mechanism and Charge Transport Phenomena in Organic Solids

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Radioluminescence Mechanism and Charge Transport Phenomena in Organic Solids

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The overview of our previous works devoted to development of the modern theory of the radioluminescence of organic condensed media is given in this paper to show that Prof. E.A. Silinsh ideas have been conformed by this theory as well as by well-known experimental facts of scintillation technique. Only some of his ideas are discussed, namely, the Gaussian distribution model for local energetic states of a polarization origin in an organic crystal energy diagram, and the molecular – lattice polaron model. It is shown that the appearance of very shallow trapping centres of polarization origin cause the additional delay of a moment of a radioluminescence photon emission in organic solid scintillators.

Keywords: radioluminescence; polarons; molecular solids; and liquids

1. INTRODUCTION

Progress of science may be compared with stairway. Like a stairway, which consists of of separate steps, science is built on significant results achieved by scientific schools and scientific workers. Such an analogy is in good agreement with the scientific heritage of Prof. E.A.Silinsh. Indeed, the Gaussian distribution model for local energy states of polarization origin of the energy diagram in an organic crystal, as proposed by him (1), has become today one of the key aspects in understanding the physical processes of charge transport in organic crystalline systems. According to this model, electronic polarization effects in the crystal by charge carriers determine the energy eigenvalues of conductivity levels and local trapping states. The energy spectrum of local states of structural origin is dis-

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cussed within the framework of the Gaussian distribution model. The results of energy structure studies for number of aromatic and heterocyclic molecular crystals confirm this phenomenological model(1). Evolution of the molecular polaron theory in combination with computer simulation allowed E.A.Silinsh and his colleagues to take the next step and to ground the molecular – lattice polaron model (polarons of "nearly small" radius (2)) for organic molecular crystalline systems (1,2). The possibility of interpretation of an overwhelming majority of transport features as those, in which the charge carrier mobility μ is not of activated nature, i.e. $\mu \sim T^{-n}$, where T is the temperature and n is a constant, was demonstrated by computer simulation for aromatic organic crystals within the scope of the molecular polaron theory. According to the lattice polaron model, i.e. in the case of thermally activated mobility, it is described as $\mu \sim \exp(-E_a/kT)$, where E_a is the depth of a trap and k is the Boltzmann constant. In other words, for the former case we have a decrease in μ -value and for the latter one an increase with rise in the temperature. E.A.Silinsh has shown that in a case when the charge state mobility is independent of temperature, combined of molecular and lattice polaron models are more expedient (2).

This paper shows that the aforementioned ideas of Prof. E.A. Silinsh have been confirmed by modern theory of radioluminescence processes in condensed organic media as well as by well-known experimental facts of scintillation technique.

2. THEORETICAL PREDICTIONS

Luminescence, which excited by ionising radiation, is accepted to be called radioluminescence (RL) (3–10). To emphasise its practical application, another name for this kind of luminescence is used, i.e., scintillation. The materials in which RL can appear are known as scintillators (3–9). In contrast to photoluminescence, in the case of RL the molecules of organic system, practically, are not directly excited by ionizing radiation (7,10). They are excited in the recombination process of charge carriers; i.e. the process of RL excitation has its specific and very important "preliminary" stage. The luminescence in organic molecular crystals, plastics as well as in liquid solutions of organic luminophors is molecular in behaviour (8–10). This means that such a process can be presented as luminescence of individual molecules in spite of a kind of excitation. The molecular nature of luminescence of organic solids and liquids has been shown for photoexcitation (8,9), as well as in comparative studies of RL and photoluminescence spectra (10). It is caused by the low energy of intermolecular interactions in the systems under discussion (1,2,7–10).

The properties of molecular solids as well as of liquid solutions of organic luminophors, brought out in the late forties, allowed developing the theory of their photoluminescence (8,11). In particular, the expressions describing a photoluminescence pulse shape for such an object were obtained on condition that the time distribution of excitation can be presented by a δ – function. This postulation is reasonable because it is equivalent to assumption that molecules are directly excited by light, and the duration of the excitation of an organic molecule into the lowest excited singlet state (S_1) is negligible. In such a situation, the process of excitation energy transfer from the points of excitation to luminescent centres of the object determines the shape of luminescence pulse rise and the process of photon emission by such a centre determines the shape of luminescence pulse decay. There are two limiting cases (8). For solid solutions the probability of energy transfer between molecules, which are the donors of the electronic excitation energy, and molecules, which are its acceptors, decrease during the time of light-pulse formation. It is caused by an increase in the average donor-acceptor distance during this process (8). For liquid solutions with low viscosity, such an effect is impossible owing to intense molecular diffusion and, therefore, the probability of electronic excitation energy transfer does not change in time. Thereby and in respect to the molecular nature of the luminescence, it is easy to obtain that the photoluminescence pulse shape is described by

$$i(t) = f(t) * \exp(-t/\tau), \qquad (1)$$

where t is the time after excitation, τ is the decay time constant, f (t) is a probability density function, which describes the time distribution of the moments of excitation of the luminescent centres. In equation (1) * is the symbol of a convolution, i.e.

$$h(t) * g(t) = \int_0^t h(t - \vartheta) \cdot g(\vartheta) d(\vartheta). \qquad (2)$$

The RL theory of organic molecular solid and liquids has been developed in the sixties (3–6). This theory proceeds from some assumptions. In particular, it is considered that the influence on kinetics of prompt RL of all the processes, which take place in plasmon, superexcited and charge states, is negligible. In such an approach, the process of excitation by ionizing radiation "loses" its specific preliminary stage and so it gives the possibility to use the expressions, which have been deduced in the photoluminescence theory. Thus, according to the theory (3), the rise of a RL pulse for pure solid scintillator must be instantaneous and the pulse shape is described by a single exponential function, because in equation (1) f (t) is a δ – function. For a binary solid scintillator it must have a fast initial rise, followed by a decrease in pulse slope owing to decrease in excitation energy transfer probability between excited molecules of the base (donors of excitation) and molecules of the luminescent solute (acceptors of excitation) with increase in duration of the process (8). The rise time has to decrease with solute concentration increase. For a δ – excitation of low viscosity liquid solutions f (t) is exponential and

$$i(t) \sim \frac{\tau \cdot \tau_1}{\tau - \tau_1}[\exp(-t/\tau) - \exp(-t/\tau_1)], \tag{3}$$

where τ_1 is the rise time constant of a prompt RL pulse, which must decrease with solute concentration increase (3,8).

3. EXPERIMENTS

The lack of precise instruments gave no possibility to examine aforementioned RL theory predictions up to the seventies. The first attempts made by Bengston and Moszynski (see review of their results (12)) to study the precise scintillation pulse shape for vinyltoluene plastic scintillators led them to equation (1) with f (t) as

$$f_G(t) = [(2\pi)^{1/2}\sigma]^{-1} \cdot \exp[-(t - m\sigma)^2/2\sigma^2], \tag{4}$$

where m = 3 and σ is the time constant, which defines the rise time of a prompt RL pulse. It was found that the σ- value was the same for pure, binary and ternary (i.e., containing molecules of a quenching agent) systems. Galligaris et al have obtained the same results (13). So, the σ- value in contrast with τ (See equation (1)) did not depend on concentration and type of the molecules of a luminescent solute or a quench agent. In contrast with theory prediction, a slow initial pulse rise with its following speeding-up was observed. It can be interpret in the framework of existing RL theory (3–6) only as the experimental detection of the presence of unknown additional delay of a photon emission (10,12). This delay was described by function (4) and it appearing cannot be explained without any change in RL theory. Bengston and Moszynski set up the states hypothesis that the σ- value in equation (4) described the decay of highest excited states of molecules. In such a case, the process of excited S_1 states quenching or variation in density of excitation cannot change the σ- value. The experiments with the same objects, but for essentially higher density of excitation (12,14,15) (see Table I) have shown that rise time of RL pulse and it duration can decrease with increase of a density of excitation.

In the sixties, it was considered that RL light output must be essentially dependent on temperature (3). Now (see the review of such materials (10)), it is well known that in the range of-60... +60 °C the RL characteristics of organic solids can vary with temperature in limits of ±5%. It is comparable with error in the determination of temperature dependence of the photomultiplier tubes used with such a scintillator. All these result in the crisis of RL theory in the eightieth.

TABLE I The σ-values (see Equation (4)) for some organic solid scintillators obtained from pulse shape measurements.[a]

Scintillators	*σ, ns*	*Ref*
Plastics		
Both single and binary component systems		
Base		
Styrene	0.26	16,19
Vinyltoluene	0.20	12,13,21
Vinyltoluene[b]	0.17...0.09[c]	14,15
Vinylxylene	0.20	17,19
Vinilxylene[b]	0.13	18,21
Organic Single Crystals		
Stilbene		
The root-mean-square of random orientation of single crystal mosaic structure (minutes of arc)		
160...25	0.18...0.21	16,21
p-Terphenyl		
Diphenylbutadiene concentration (mol.%)		
No doping	0.38	
0.042	0.30	
0.083	0.24	16,21
0.415	0.18	

a. In order to reduce the influence of the light-collection processes on light pulse shape all experiments of such a type have been made at room temperature (10,12–21). The use of a light guide and light collection systems needed for low-temperature RL experiments (in contrast to the case of photoexcitation) potentate the effect of the light-collection process on the time spread of photon registration in pico- and subnanosecond ranges. May be, thereby there are no experimental data for temperature dependence of σ-value.

b. The density of excitation was high and tracks were overlapping.

c. The estimated range of σ-values, which was calculated on the base of the data obtained for different densities of excitation (14, 15).

We have found (16–21) that equation (1) modified by the Gaussian function (4) is correct for all organic solid scintillators except those in which the process of radiative transport is of primary importance. The σ- value did not depend on the type and concentration of added agents in plastics, but it was determined by the type of the polymer macromolecule and the density of excitation (see Table I).

For organic crystals the σ-value weakly depends on crystal structure perfection just as much as it is connected with the concentration of a charge carrier deep trap. For liquid scintillators of low viscosity (based on toluene, xylene) no deviation from equation (3) was observed (17,18,22). Figure 1 demonstrates the effect of additional delay, which is described by function (4), for plastic and crystal scintillators in comparison with liquid one. The comparison between the fluorescence spectra of organic crystals, plastics and liquid scintillators for the cases of radio- and photoexcitation (excitation of base molecules in S_1 – energy state) showed their identity (10).

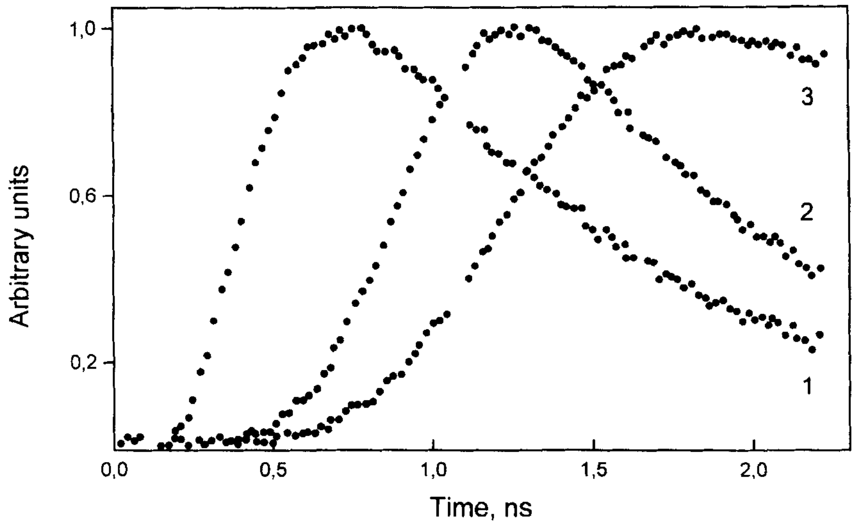

FIGURE 1 Normalized time distribution spectra of the radioluminescence pulse from liquid (curve 1), plastic (curve 2) and single crystal (curve 3) binary scintillators (18). Compositions: liquid toluene scintillator contains 10g/dm^3 PBD (2-phenyl-5- (4-biphenylyl) – 1,3,4- oxadiazole); plastic polyvinyltoluene scintillator contains 3.46 mol.% PBD; crystal scintillator is the single crystal of p-terphenyl, which contains 0.1% diphenylbutadiene (substitutional solid solution)

4. DISCUSSION

The equation (1) holds always, because in multicomponent systems RL is of molecular and sensitized nature (3,7–9). We have verified this by comparative experimental study of RL and photoluminescence spectra (10). Description of a prompt RL pulse shape by equation (1) with $f(t) = f_G(t)$ (4) is only possible when the existence of the "preliminary" stage of the RL excitation is taken into account (18). Thereby the expressions describing a pulse shape of RL for condensed organic media must be obtained on such a condition when the time distribution of excitation can be presented by a function differing from a δ-one.

According to experimental data, an additional delay in the moments of photon emission was observed (see Figure 1). It is described by the probability density function (4) with m = 3 and $\sigma \sim 0.2\ldots0.26$ ns for plastics, and $\sigma \sim 0.18\ldots36$ ns for organic crystals (see Table I). Therefore, the following situation has to be analysed. For a multicomponent system, two cases can exist: 1) luminescence takes place in primary excited molecules and 2) luminescence is sensitized. For the latter situation one has to consider the system, which consists of molecules × and y in S_0 – state (the ground state). The excitation of molecules × (donors of excitation energy) is described by function G (t). The excitation of molecule y (acceptor) appears when the process of energy transfer from molecules × to molecule y takes place. The first situation is the special case for the second one (the case when the probability of energy transfer is equal to 0). Thus, it is necessary to examine the system of equations

$$\begin{cases} \frac{dC^x(t)}{dt} = G(t) - \lambda(t)\cdot[\mathrm{Y} - C^y(t)]\cdot C^x(t) - \nu C^x(t) & (5) \\ \frac{dC^y(t)}{dt} = [\gamma + \lambda(t)]\cdot[Y - C^y(t)]\cdot C^x(t) - \beta C^y(t) & (6) \end{cases}$$

In Equations (5) and (6): Y is the concentration of acceptor molecules; $C^x(t)$ and $C^y(t)$ are the concentrations of excited donors and acceptors for the time t after excitation, correspondingly; λ(t) is the probability density function, which describes nonradiative transfer from excited molecules × to molecules y; γ is the rate constant of radiative electronic excitation energy transfer from × to y; $\nu \equiv 1/\tau_0$ is the rate constant of donor luminescence for the case when energy transfer to acceptors is absent, $\beta \equiv 1/\tau$ is the rate constant of acceptor luminescence. We shall consider this problem for the following initial conditions:

$$\begin{cases} C^x(0) = 0 & (7) \\ C^y(0) = 0 & (8) \end{cases}$$

Let us denote the total probability of deactivation of donors′ excitation as

$$u(t) \equiv \lambda(t)\cdot[Y - C^y(t)] + \nu \boxminus \varphi(t) + \nu \sim \lambda(t)\cdot Y + \nu \qquad (9)$$

It should be mentioned that for the RL the concentration of excited molecules is considerably (usually, 10^5 times (7,10)) lower than the total concentration of molecules, i.e. always $Y \gg C^y(t)$. Therefore in Equation (9) $u(t) \approx \varphi(t) = \lambda(t)Y$.

The analytical solution (23) of this problem for two limiting cases of energy transfer (i.e. for solids and liquid systems with low viscosity) allows reaching the following conclusions. The description (1) with f (t) = f_G (t) = G (t) (See Equations (1) and (4)) is valid if the process of generation of the base molecules excited in S_1- state (donors of excitation energy) is described by the Gaussian function (4), and the σ- value exceeds the characteristic time of the subsequent excitation energy transfer. The solutions are analogous to the ones of the photoluminescence theory (8), when the case of δ- function excitation is being considered (23).

Using the central limit theorem (24) one can obtain the same results for the general case as well. Indeed, let $\{\xi_k, k\geq 1\}$ be a sequence of mutually independent random variables with distribution functions $G_k(X) = p\{\xi_k > X\}$, which possess the finite mathematical expectation $M\xi_k$ and variance $D\xi_k = \sigma_k^2$ $\left(\sum_{k=1}^{n}\sigma_k^2 > 0\right)$. Suppose that the sequence $\{\xi_k, k\geq 1\}$ and a similar one $\{\eta_k, k\geq 1\}$ describes the time distribution of the moments of excitation of the base molecules in S_1- state and the solute ones (in the process of donor-acceptor energy transfer), respectively. If the population of the random variables ξ satisfies conditions of the central limit theorem, and

$$\sum_{k=1}^{n} M|\xi_k - M\xi_k|^3 \Big/ \left|\sum_{k=1}^{n} D\xi_k\right|^{3/2} \tag{10}$$

is small for $n\to\infty$, then the distribution law of a sum of random variables $\xi_1+\xi_2+\ldots+\xi_n$ is close to a normal one, no matter how ξ- variables are distributed. This may cause the shape of the function (4), which has been obtained by experiment for pure solids.

Let define

$$\zeta_k = \xi_k + \eta_k \tag{11}$$

In the case under consideration, a sequence of mutually independent random variables $\{\zeta_k, k \geq 1\}$ describes the probability of a luminescent centre excitation at the moment t. If a condition similar for the (10) one is satisfied for ζ- value, then the distribution law of a sum of summands $\zeta_1+\zeta_2+\ldots+\zeta_n$ is close to a normal one. Condition (10) is violated as soon as the variance of some summands (e.g. η_k) exceeds the variance of the rest. In such a case, their sum is not close to the normal distribution law. Therefore the distribution of the sum $\zeta_1+\zeta_2+\ldots+\zeta_n$ is defined by a distribution law of the sum $\eta_1+\eta_2+\ldots+\eta_k$, when the probability of energy transfer is low (i.e. the σ- value in (4) is smaller than the values of characteristic time of the process and its variance). The sum of independent random variables ζ is distributed according to normal law, if the sum of independent random variables ξ also follows this law, and the probability of energy transfer is high (i.e. the variance of η- value distribution is less than one of ξ- value distribution), no matter how η- variables are distributed.

For effective organic scintillators, which are really used, the probability of energy transfer is high, and the characteristic time of such a process is about $10^{-12}...10^{-11}$s$^{(9,11)}$. It is less than $\sigma \sim 10^{-10}$s (see Table I). Therefore, the answers for two questions must be found in order to understand the origin of the additional delay described by function $f_G(t)$ in equation (1). What is the cause of such a delay origin? Why σ is about 10^{-10}s?

The only possible explanation for the origin of the additional delay, which is described by Equation (4), has to be associated with "preliminary" stage of RL process $^{(10,25)}$. Indeed, the duration of other energy exchange processes such as interaction of ionizing radiation with molecules of matter, generation of plasmons and superexcited states, nonradiative intermolecular and intramolecular energy transfer does not exceed 10^{-11}s $^{(3,\ 7,11)}$. The charge carriers generated by ionizing radiation before their recombination (consequently before a base molecule excitation) can localize on a trap

$$E_t = kT \cdot \ln(\tau_t \nu) \tag{12}$$

in depth. In Equation (12) k is the Boltzmann constant, T is the room temperature (see the note for Table I), τ_t is location time, ν is the attempt-to-escape frequency. If we take $\tau_t = 3\sigma$ (according to equation (4)) and $\nu \sim 10^{12}$s$^{-1(1,18,19)}$, then for the σ- values (see Table I) we obtain $E_t \leq 0.17$ eV.

Let ΔE to be the difference in depth between the trap with location time τ_t and one with location time $\tau_t+\Delta\tau$. For $\Delta E < kT$ it is not difficult to obtain from expression (12) that

$$\Delta E \cong \Delta\tau \cdot (kT/\tau_t). \tag{13}$$

For a given temperature and sample kT/τ_t is a constant. Hence, for a shallow trap system both the location time distribution and the energy one should be described by a function of the same type, i.e., according to expression (4), by Gaussian functions. That is just what has been obtained during the study of the energy distribution of trapping centres, which are formed in regions of local compression and tension of individual edge dislocations of organic crystals by E.A. Silinsh $^{(1,2)}$.

Trapping centres with values of $E_t \leq 0.17$ eV can be associated with low- and medium – angle boundaries of organic crystal lattices or spatial non-uniformities of polymer macromolecules $^{(2,21)}$. Notwithstanding this, such an explanation of the problem discussed is not complete enough. Indeed, the following questions arise. Why are the differences between E_t-values obtained for organic solids with greatly different structure so small (see Table I)? Why is the description (3), but not (4) valid for liquids?

Having fallen into a trapping centre a charge carrier polarizes the molecules of organic matter, those surrounding this centre. The result of the interaction of the charge with dipoles induced on neighbouring molecules can be represented as deepening of a trapping centre by the value of δE_t. This value is about 0.1 eV for the wide class of organic materials (1,2). Hence, the estimated depth of the initial structural trapping centre is $E_t - \delta E_t \leq 0.07$ eV or about 2kT. Therefore the appearance of very shallow trapping centres for charge carriers, most of which can even be produced by temperature-activated vibrations of the organic crystal lattice of fragments of macromolecules of polymers, completely explains the appearance of the additional delay of the emission of RL photons. Such a dynamic trapping centre of polarization origin can be formed during a charge carrier localization on every molecule, and the concentration of such centres is equal to the concentration of all localized charge carriers, i.e. it is very high. It is just described by expression (4).

Thus, the Gaussian form of function (4) only testifies of the statistical nature of the processes described. The same result follows from the above analysis based on central limit theorem. If the charge carrier motion were defined only by transfer integrals, then the corresponding time of charge carrier localization would be about 10^{-14}... 10^{-12}s. It exceeds the time of electronic (10^{-15}...10^{-16}s) and vibrational (10^{-15}...10^{-14}s) polarization, and therefore a molecular polaron can be formed. For organic crystals, a local lattice polarization takes place, and a potential well δE_t in depth appears. Thereby, the formation of dynamic trapping centres of polarization origin raises the possibility of increasing the time of charge carrier localization up to 10^{-10}s. It exceeds the lattice polarization time (10^{-12}...10^{-11}s) (2). A presence of traps of a structural origin causes the same result (i.e. increase of a lattice polaron formation probability). The effective value of μ is formed under the influence of all the processes, which define the transport of a charge carrier in organic solid. For effective organic scintillators the intensity of prompt RL is caused mainly by the efficiency of the process of charge state recombination, which takes place during the time not longer than a RL pulse decay time (25). Therefore, the extremely weak temperature dependence of a RL signal can find a reasonable explanation only on the base of the idea of molecular – lattice polaron model, as analysed by E.A. Silinsh (2).

For liquids, equating the thermal energy to the polarization one at a distance R from the ion, we can estimate the dimension of a zone of it stable polarization surrounding(18,25)

$$R = (e^2\alpha/2kT)^{1/4} \sim 1.0nm, \qquad (14)$$

where e is electronic charge, α is the mean molecular polarizability ($\alpha \sim 2{\cdot}10^{-23}$ cm^3 (10)), k is the Boltzmann constant, and T is the temperature. According to expression (14), R is about a molecule size (1,7,9). Therefore, the generation of excited molecules in liquids is practically due to recombination, which takes place between geminate ions. Hence, the recombination process duration is determined by the characteristic collision frequency of molecules (i.e. ~$10^{12}s^{-1}$), and no delay, which described by expression (4) with characteristic time about 10^{-10}s, can appear.

5. CONCLUSION

The main features of the prompt RL of condensed organic media are determined by its molecular nature. The process of charge carrier energy exchange defines the light pulse formation. For liquids, this process mainly takes place between geminate ions and therefore its duration is limited by their characteristic collision frequency. For organic solids the description of the pulse shape of the RL pulse fast component by expression (1) with $f_G(t)$, which is described by expression (4), is valid when the probability of excitation energy transfer is high, and the spread of the moments of excitation of luminescent centres of the scintillator is defined by the process of preliminary stage of charge carrier generation, localization and recombination. In such a case, the localization of charge carriers on dynamical trapping centres of a polarization origin results in an additional delay of a RL photon emission, which is described by clipped Gaussian function (4). This process of polaron states formation, transport, and recombination will be sensitive to the structure perfection of the object just as much as it will be connected with the concentration of charge carrier traps of a structural origin. The appearance of very shallow trapping centres (most of which can be initiated by the temperature activated vibrations of the organic crystal lattice or fragments of macromolecules of polymers) can result in formation of such dynamic states of polarization origin. The only way to reduce the probability of such a state formation is to generate a high density of charge carriers, which may have not enough time to localize before their recombination. That is why the σ- value decreases for a high density of excitation (see Table I). When the probability of nonradiative energy transfer is low, the RL pulse shape should be approximated by function (3), and the light-collection and reabsorption processes would cause the τ_1-value.

The material discussed has shown that Prof. E.A. Silinsh ideas are not only useful for understanding semiconductive properties of organic crystals, but can be used as the key points in investigating the physical processes taking place in organic condensed media, in particular, for development the RL theory of these objects.

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