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Nature of the Additional Delay of Prompt Radioluminescence Photon Emission in Organic Solids and its Description by Gauss Function

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In contrast to photoluminescence kinetics of organic solid materials for radioluminescence one an additional delay of the moments of photon emission is observed. We have carried out the detailed physical ground of application of Gauss function for description of this delay. The precise analysis of the experimental results let us the possibility to prove the plural nature of the processes causing formation of the radioluminescence pulse.

Keywords: radioluminescence; organic scintillators; Gauss function

INTRODUCTION

The theory of the radioluminescence (RL) of organic molecular systems was grounded on a series of assumptions accounting only in part for the RL specific features[1]. The quickness of processes preceding excitation of S_1-states (the lowest singlet excited states) was believed to allow for the assumption that the kinetics of the luminescence pulse fast component is the same under both photo- and radioexcitation. Therewith, δ-function used to describe the RL kinetics[2]. However for the case of the RL molecules of organic systems are not directly

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excited, but are excited in the recombination process of charge states[3]. The first investigation of the RL kinetics led to unexpected results[4]. A slow initial pulse rise with its following speeding-up was observed for plastic scintillators. In contrast to the expected situation when the scintillation pulse maximum for a "rigid"[2] solution doesn't appear before the maximum for a nonviscous one appears after it. Thus, for organic solids the additional delay of the moments of photon emission was observed[4]. An existence of this delay is caused by the process of charge carrier localization on shallow dynamical trapping centers of a polarization origin. The solution of kinetic equations with a function of "excitation" which defers from δ-function has shown[5] that the additional delay of the moments of RL photon emission for organic solid scintillators is due to only the processes of generation and recombination of charge states.

For organic solids the RL pulse shape is described as

$$i(t) = f(t) * \exp(-t/\tau) \equiv \int_0^t f(t-x) \cdot \exp(-x/\tau) dx. \qquad (1)$$

In Eq.(1) τ is a decay time constant, t is the time after excitation and "*" is a symbol of a convolution. Function $f(t)$ describes the additional delay of the moments of RL photon emission. The mathematical treatment of the experimental curves shows that the form of the RL pulse for organic solid scintillators is described best of all when

$$f(t) \equiv f_G(t) = \left\{const/(\sqrt{2\pi}\sigma)\right\} \cdot \left\{\exp\left[-(t-\Delta)^2/2\sigma^2\right]\right\}, \qquad (2)$$

where Δ and σ are time parameters. But why function $f(t)$ described the additional delay of the moments of photon emission must have a Gaussian form? Is the choice of function $f(t)$ in the form of $f_G(t)$ physically grounded? Try to answer these questions.

EXPERIMENT

Fast RL of stilbene single crystals with controlled structural perfection, *p*-terphenyl single- and polycrystals doped by diphenylbutadiene,

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plastic scintillators on the base of polyvinyltoluene was investigated. To measure time parameters σ and τ of the RL pulse fast component we used a single-photon technique. The set-up with subnanosecond time resolution was used[6]. For all organic solids the additional delay of the moments of photon emission was observed and the RL pulse shape was described by Eq.(1) with $f(t)$ (Eq.(2)) where $\Delta \cong 3\sigma$. Table 1 presents the results of measurements of the values σ and τ, as well as the values of relative light yield J for some organic scintillators. The σ_c-value in Table 1 is the room-mean-square random orientation of the mosaic blocks of stilbene single crystals, C_1 is the concentration of diphenylbutadiene molecules in the scintillators on the base of *p*-terphenyl, C_2 is the concentration of PBD molecules in plastic scintillators on the base of polyvinyltoluene.

TABLE 1. Characteristics of the RL pulse for some organic single crystals (OSC), polycrystals (OPC) and plastic scintillators (PS).

Scintillator	σ, ns	τ, ns	J, rel. Units	N, Photons/ MeV
OSC, stilbene (σ_c=26 min.of arc)	0.21	3.71	1.00	$1.40 \cdot 10^4$
OSC, stilbene (σ_c=160 min.of arc)	0.18	3.32	0.87	$1.22 \cdot 10^4$
OSC, *p*-terphenyl (C_1=0)	0.40	4.05	1.03	$1.45 \cdot 10^4$
OSC, *p*-terphenyl (C_1=0.1%)	0.25	3.65	1.23	$1.72 \cdot 10^4$
OPC, *p*-terphenyl (C_1=0.007%)	0.65	3.17	0.74	$1.04 \cdot 10^4$
OPC, *p*-terphenyl (C_1=0.2%)	0.55	3.57	1.25	$1.75 \cdot 10^4$
PS, polyvinyltoluene (C_2=0)	0.20	14.0	0.06	$8.40 \cdot 10^2$
PS, polyvinyltoluene (C_2=3.46%)	0.20	0.9	0.17	$2.38 \cdot 10^3$

DISCUSSION

So, the mathematical treatment of the experimental results of the measurements of kinetical parameters of the RL pulse shows that the

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additional delay of the moments of photon emission is described best of all by Gauss function $f_G(t)$ (2). Try to argue by another way the regularity of application of function $f_G(t)$ (2) for description of the additional delay. As it mentioned this delay is caused by charge carrier localization on shallow trapping centers of polarization origin. The charge carrier recombination may result in the appearance of RL photons with the delay for the time τ_i. The localization time of a charge carrier on a trap having the depth E_i can be estimated as[7]

$$\tau_i = \nu^{-1} \cdot \exp(E_i / kT), \tag{3}$$

where ν is the attempt-to-escape frequency, k is the Boltzman constant, T is a temperature. Let us consider Eq.(3) for the situation where a certain local state E_t is the most probable one within the energy diagram of a molecular crystal for local states E_j distribution. Designating $\Delta E = E_j - E_t$, we obtain from (3)

$$\Delta\tau = \nu^{-1} \cdot \exp(E_t / kT) \cdot [\exp(\Delta E / kT) - 1]. \tag{4}$$

If $\Delta E/kT \ll 1$ it follows from (3) and (4):

$$\Delta\tau \cong \Delta E(\tau_t / kT). \tag{5}$$

Thus, the scatter of the charge localization time on shallow traps should be defined by the distribution of the same kind as the scatter of energies for those traps. The statistical nature of shallow traps in organic single crystals is known[7] to result in that their distribution in the energy diagram of a crystal is of Gaussian character. Thus, if the duration of the charge carrier localization process on shallow traps of polarization origin exceeds the characteristic one of the electron excitation energy transfer[2], the shape of the RL pulse fast component should be described by Eq.(1) and its additional delay of the moment of photon emission by function $f(t)$ in the form of $f_G(t)$. But such an argument has only a qualitative character.

Let $\{\xi_k,\ k \geq 1\}$ be a sequence of mutually independent random variables with distribution function $G_k(x) = p\{\xi_k \geq 1\}$ which posses the finite mathematical expectation $M\xi_k$ and variance $D\xi_k$. Suppose that the sequence $\{\xi_k,\ k \geq 1\}$ describes the time distribution of the moments of

excitation of the base molecules in S_1-state. If the population of the random variables satisfies the central limit theorem condition[8] and

$$\sum_{k=1}^{n} M \mid \xi_k - M\xi_k \mid^3 / \mid \sum_{k=1}^{n} D\xi_k \mid^{3/2} \qquad (6)$$

is small for $n \to \infty$, then the distribution law of a sum of random variables $\xi_1 + \xi_2 + \ldots + \xi_k$ is close to normal one, no matter how ξ-variables are distributed. So, this analysis shows that the Gaussian description of the processes that cause the features of scintillation pulse formation is physically grounded when these processes are plural ($n \to \infty$) and variance of every singular process is not so many as a variance of the summary process.

Firstly, let us evaluate the number of initial states (plasmons, superexcited states) created by one ionizing particle. The energy of plasmon creation is about 20 eV[1]. Taking the energy of the source of gamma excitation ^{22}Na is equal to 511 keV and assuming that the losses of energy are absent we may obtain that the number of plasmon states is about $2.55 \cdot 10^4$ (or $5.1 \cdot 10^4$ plasmons for 1 MeV of particle energy). The number of plasmon states for 1 MeV of β-excitation has a similar value. It should be noted that we discuss the processes proceeding in the regions of low activation density when a track is not created and therefore the additional losses, practically, is absent. Such a huge number of plasmon states and a little number of different types of systems of trapping centers for charge carriers in these organic systems testify about the plural nature of the scintillation mechanism.

These estimations are in a good agreement with the results of measurements of light yield J (see Table 1). The light yield J of the stilbene single crystals with σ_c=26 min. of arc we have taken for 1. For the case of γ-excitation, according to[9], the number of photons N for 1 MeV of particle energy in the fast component of the scintillation pulse is equal to $1.4 \cdot 10^4$ (see Table 1). Using the known value of relative light yield J we can calculate parameter N for other scintillators. Table 1 demonstrates some examples of the results of such calculation. For all single crystals parameter N is about 10^4 photons/MeV, for activated

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plastic scintillators N is about 10^3 photons/MeV, and for non activated plastic scintillators N is about 10^2 photons/MeV.

Taking the value N for nondoped p-terphenyl single crystals and the values J for p-terphenyl polycrystals we can estimate the values N for polycrystals. The calculated values N for organic polycrystals are $1.0...1.8 \cdot 10^4$ photons/MeV. So, a particle having the energy 1 MeV creates, approximately, $5 \cdot 10^4$ plasmons in organic scintillators. It causes generation, approximately, 10^4 and 10^3 photons in the RL pulse fast component for organic crystals and plastic scintillators, consequently. This fact testifies to a plural character of the process that causes the appearance of the additional delay of the moments of RL pulse emission for organic scintillation materials. So, the description of the additional delay of the moments of RL pulse photon emission by Gauss function is not only convenient approach, but is physically argued one.

Acknowledgments

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References

[1] J.B. Birks. *The Theory and Practice of Scintillation Counting.*Pergamon Press, London, 1964.
[2] V.M. Agranovich, M.D. Galanin. *Excitation Energy Transfer in Condensed Media.* North Holland, Amsterdam, 1982.
[3] S.V. Budakovsky, B.V. Grinev, O.A. Tarasenko, D.V. Vasilyev. *Mol. Cryst. Liq. Cryst.***324**, 145 (1998).
[4] B. Bengston, M. Moszynski. *Nucl. Instrum. and Methods.* **81**, 109 (1970); **155**, 221 (1978).
[5] N.Z. Galunov, O.A. Tarasenko. *Functional Materials.* **4**, 272 (1997).
[6] N.Z. Galunov, O.A. Tarasenko, V.V. Jarychkin, D.V. Vasilyev. *Functional Materials.* **5**, 117 (1998).
[7] E.A. Silinsh, V. Capek. *Organic molecular crystals. Interaction, localization and Transport Phenomena.* American Institute of Physics, New York, 1994.
[8] D.I. Hudson. *Statistics.* Geneva, 1964.
[9] N.Z. Galunov, N.I. Shishov. *Mol. Cryst. Liq. Cryst.* **252** 125 (1994).

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