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Electronic Energy Transfer in Linear Polymer Chains

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Abstract—The effect of the size of a polymer molecule and the character of the motion of its units in solution on the luminescence kinetics of chromophores attached to the polymer chain was studied.

Electronic excitation energy transfer between donor and acceptor chromophores attached to a polymer chain is widely used as a tool for studying the structure of polymers [1, 2]. The time dependence of the donor and acceptor fluorescence after pulsed excitation can be related to the interchromophore distance via the rate of the Förster energy transfer [3]. These experiments are usually carried out in very diluted solutions in which energy transfer between different polymer molecules negligible. It is clear that intermolecular energy transfer cannot be neglected at moderate polymer concentrations. The interpretation of experimental results obtained for such systems appears to be a much more complex problem requiring the many-body problem of excitation transport to be solved [1, 4].

There are several types of chromophore attachment to polymer molecules. First, each polymer molecule can contain exactly one donor and one acceptor chromophore [5–8]. Some biomolecules belong to this class. Second, donors and acceptors can be randomly distributed along the polymer chain [1, 4, 6, 9–15]. Third, each unit of a polymer molecule can be occupied by a donor or acceptor chromophore with a probability equal to unity [1, 2, 16–22]. Aromatic polymers represents an example of such systems.

The aim of this paper is to briefly review our results obtained in the study of incoherent energy transfer between chromophores attached to a flexible polymer chain. The polymer concentration is considered low enough to neglect interaction between the chromophores attached to adjacent polymer chains. It is assumed that the solvent is good, and a polymer is above the θ transition point. It is known that a polymer chain forms a globule below the θ temperature, and the probability density for the polymer chain to have a given end-to-end distance will differ from that used in this paper. We will study the case of isotropic dipole–dipole (dd) interaction between chromophores, when the rate of energy transfer $w(r)$ is defined by $w(r) = \tau^{-1}(R_0/r)^6$. Here, τ is the lifetime of the excited state of

a donor, R_0 is the Förster radius, and r is the distance between chromophores.

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In this case, the luminescence kinetics upon pulse excitation $I(N, t)$ of donor chromophores attached to the polymer chain containing N units is described by the equation [1–15]

$$I(N, t) = 4\pi \int_0^{\infty} r^2 g_N(r) e^{-tw(r)} dr. \quad (1)$$

The exponential term in Eq. (1) describes the kinetics of irreversible excitation energy transfer from a donor to an acceptor. The distance r between the donor and the acceptor depends on the chain conformation. The averaging over the conformations is performed with the use of a $g_N(r)$ function, which is the probability density of finding a polymer containing N units, in which the distance between the chain ends is r .

Equation (1) does not involve the excitation decay caused by a finite lifetime of the fluorescent state of the donor, because this decay channel is independent of energy transfer. Hence, the overall decay kinetics is the product of multiplying by $\exp(-t/\tau)$ and a decay function due to energy transfer to acceptors.

It is known [5, 23] that, in the three-dimensional case for an ideal (Gaussian) polymer chain,

$$g_N(r) = A_1 \exp(-B_1 r^2), \quad (2)$$
$$A_1 = \left(\frac{3}{2\pi \langle R_g^2 \rangle} \right)^{3/2}, \quad B_1 = \frac{3}{2 \langle R_g^2 \rangle}$$

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and for a polymer chain with excluded-volume interaction (nonideal chain) [5, 7, 8, 23],

$$g_N(r) = A_2 r^{5/18} \exp(-B_2 r^{5/2}), \quad (3)$$

$$A_2 = 0.289058 / \langle R_g^2 \rangle^{59/36}, \quad B_2 = 1.22271 / \langle R_g^2 \rangle^{5/4},$$

where R_g is the radius of a polymer coil (radius of gyration), $\langle R_g^2 \rangle = a^2 N$ for the ideal chain and $\langle R_g^2 \rangle = a^2 N^{6/5}$ for nonideal chain, and a is the length of a statistical segment (unit). One can see from Eqs. (2) and (3) that, at $r \rightarrow 0$, the distribution function is equal to zero for the chain with the excluded-volume interaction and is maximum for the ideal chain.

Substituting Eq. (2) into Eq. (1), we obtain an expression for the luminescence kinetics of the ideal polymer chain [7, 8, 15]:

$$I_{id}(N, t) = \frac{2}{\pi^{1/2}} \int_0^\infty y^{1/2} \exp\left(-y - \frac{27T}{8y^3}\right) dy, \quad (4)$$

$$T = \frac{R_0^6}{\langle R_g^2 \rangle^3} \frac{t}{\tau}, \quad \langle R_g^2 \rangle = a^2 N.$$

It should be pointed out that the kinetics as defined by Eq. (4) depends only on the parameter T . The integral in Eq. (4), although written in a less convenient form (without introduction of the universal parameter T) was analyzed numerically in [6] for particular values of R_0 , R_g , and τ , and its asymptotics at long times ($t/\tau \gg 1$) was derived in [5]. By numerical calculations [7, 8], we found that Eq. (4) can be approximated with an accuracy higher than 1% by the function:

$$I_{id}(N, t) = (1 + 2.45T^{1/4}) \exp(-2^{5/4} T^{1/4}). \quad (5)$$

Similarly, we obtained the following expression for a nonideal chain [7, 8, 15]:

$$I_{nid}(N, t) = \frac{1}{\Gamma(1 + 14/45)} \times \int_0^\infty y^{14/45} \exp\left[-y - \left(\frac{\Gamma(19/9)}{\Gamma(59/45)}\right)^3 \frac{T}{y^{12/5}}\right] dy. \quad (6)$$

The parameter T has the same form as in Eq. (4), but $\langle R_g^2 \rangle = a^2 N^{6/5}$. The asymptotics of Eq. (6) at long times was calculated in [5]. From the numerical analysis, we found [7, 8] that kinetics (6) was reproduced with an accuracy higher than 3% by the function

$$I_{nid}(N, t) = (1 + 1.75T^{73/306}) \exp(-2.1122T^{5/17}). \quad (7)$$

Comparing Eqs. (5) and (7) with experimental data, it is possible to determine the type of the chain under study and the radius of the polymer coil. These expressions are also very useful for studying the effect of inhomogeneous broadening of the chromophore spectra and

the segmental motion of a polymer chain on the luminescence kinetics [7, 8, 14, 15].

Polymer Chain with Random Distribution of Chromophores Along the Chain

In this part of the paper, we consider a polymer chain containing chromophores that can be attached to each unit with a certain probability. The concentration of donor chromophores is assumed to be low in order to disregard energy migration over the donors.

When investigating excitation transport in a system of a limited size (a donor and an acceptor are separated by a finite number of monomeric units), it is necessary to make two types of space averaging. The first is averaging over all possible positions of acceptors relative to the position of the primarily excited donor, and the second is averaging over all possible positions of the primarily excited donor (these positions are not equivalent in a size-bounded system). This problem for an ideal polymer chain was solved numerically in [4, 6, 9–12].

The problem of excitation transport in a polymer chain is simplified significantly when the chain under study is long enough (infinite). In this case, the second averaging is unnecessary, because all positions of the primarily excited donor are equivalent in the infinite chain. Since the distance between two not very close sites of the polymer chain divided by N units satisfies the same distribution function as the end-to-end distance of the polymer chain consisting of N units [23], the luminescence kinetics is described by the expression [14, 15]:

$$I(t) = \exp\left[-c \int_{-\infty}^\infty dN \int_0^\infty dr 4\pi r^2 g_N(r) (1 - e^{-tw(r)})\right], \quad (8)$$

where c is the average number of acceptor chromophores per monomer unit ($c \ll 1$). For the dd interaction, both integrals in Eq. (8) are calculated exactly. As a result, we obtain the following expressions for the luminescence kinetics of the ideal polymer chain $I_{id}(t)$:

$$I_{id}(t) = \exp\left[-6\Gamma\left(\frac{2}{3}\right) c \frac{R_0^2}{a^2} \left(\frac{t}{\tau}\right)^{1/3}\right] \quad (9)$$

$$= \exp\left[-8.125 c \frac{R_0^2}{a^2} \left(\frac{t}{\tau}\right)^{1/3}\right],$$

and the nonideal polymer chain $I_{nid}(t)$:

$$I_{nid}(t) = \exp\left[-3.872 c \left(\frac{R_0}{a}\right)^{5/3} \left(\frac{t}{\tau}\right)^{23/54}\right]. \quad (10)$$

Luminescence intensity (10) decays with time at a significantly slower rate than given by Eq. (9) (see the numerical factors), because the distribution function $g_N(r) \rightarrow 0$ at $r \rightarrow 0$ for the nonideal polymer chain

and $g_N(r) \rightarrow \text{const}$ at $r \rightarrow 0$ for the ideal polymer chain.

MOTION OF POLYMER CHAIN UNITS

The effect of the motion of a polymer chain in dilute solutions on the rate of energy transfer was studied earlier in [24–28]. The diffusion equation was customarily used to describe this motion. These equations were solved numerically for short polymer chains ($N = 4\text{--}22$) and the particular form of the distribution function $g_N(r)$ in [24, 27, 28] and for ideal polymer chains in [25, 26]. Comparison with experimental data showed that the diffusion coefficient D could attain a value of 10^{-5} cm²/s [26–28]. Since the equations depend on several parameters (D , R_0 , R_g , and τ), their domain in which the diffusion motion of polymer chain units should be taken into account was not established.

Polymer Chain with Two Chromophores Attached to Its Ends

When the conformation of the chain changes, the luminescence kinetics of a donor is described by the expression similar to Eq. (1),

$$I_{dif}(N, t) = 4\pi \int_0^{\infty} r^2 g_N^*(r, t) dr. \quad (11)$$

However, here $g_N^*(r, t)$ is the distribution function over the distances between the excited donor and unexcited acceptor, which are attached to the ends of the polymer chain containing N units. At the initial point of time, $g_N^*(r, t = 0) = g_N(r)$. In the static case, $g_N^*(r, t) = g_N(r)\exp[-tw(r)]$ (see Eq. (1)). It is known [23–25] that loose units are involved in the Brownian motion in a dense solvent with a diffusion constant D , and the distribution $g_N(r)$ can be interpreted as the energy term if the interaction energy of the units $V(r) = U(r)/kT_0$ (k is the Boltzmann constant and T_0 is the absolute temperature) is introduced so that

$$g_N(r) = \exp(-U(r)/kT_0) = \exp(-V(r)). \quad (12)$$

Then, after introducing new variables (the dimensionless distance $x = r/\sqrt{\langle R_g^2 \rangle}$, time T (see (4)), and diffusion constant $d = D\tau \langle R_g^2 \rangle^2 / R_0^6$) and the new function $f(x, T)$ defined by $g_N^*(x, T) = f(x, T)g_N(x)$, we have for the ideal polymer chain the following expression instead of Eq. (11) [7, 8]:

$$I_{dif}(N, T) = 4\pi \int_0^{\infty} x^2 f(x, T) g_N(x) dx, \quad (13)$$

where $g_N(x) = (3/2\pi)^{3/2} \exp(-3x^2/2)$, with the diffusion equation for $f(x, T)$ being written in the form

$$\frac{\partial}{\partial T} f(x, T) = d \frac{\partial^2}{\partial x^2} f + d \frac{\partial}{\partial x} f \left(\frac{2}{x} - \frac{\partial V}{\partial x} \right) - \frac{1}{x^6} f. \quad (14)$$

Equation (14) can be solved with the following boundary and initial conditions

$$\frac{\partial}{\partial x} f(x, T)|_{x=x_0} = 0, \quad f(x, 0) = 1. \quad (15)$$

The first two terms on the right-hand side of Eq. (14) describe a change in the distribution function $g_N^*(r, t)$ caused by the Brownian motion of (chromophore-containing) units in a spherical field $U(r)$, and the third term describes a decrease in the distribution function caused by dipole–dipole energy transfer from the excited donor to the acceptor. The boundary condition means that chromophores cannot be approach each other to a closer distance than $r_0(x_0)$.

Equation (14) actually depends on one parameter d (at small x_0). It was solved numerically. The results of the calculation of the luminescence kinetics show [7, 8] that diffusion is important if $d = D\tau \langle R_g^2 \rangle^2 / R_0^6 > 10^{-2}$.

We found [7, 8] that the kinetics became exponential with increasing d ($I_{dif}(N, t) \sim \exp(-k_{dif}t)$, where k_{dif} is the rate constant). In the limit of very fast diffusion, we derived for the ideal polymer chain the following expression for the rate constant k_{dif}^{id} :

$$k_{dif}^{id} \cong \frac{1}{\tau} \frac{R_0^6}{\langle R_g^2 \rangle^{3/2} r_0^3}. \quad (16)$$

We derived a similar expression of the rate constant for the nonideal chain k_{dif}^{nid} [7, 8]. The difference between k_{dif}^{id} and k_{dif}^{nid} is very small.

It should be noted that the diffusion equation can be used to describe the luminescence kinetics only when the distance between chromophores changes during the motion of units by small steps compared to the Förster radius R_0 [29].

In general, another type of the motion of a polymer molecule, which is alternative to the diffusion motion, can be suggested. One can assume [17, 20, 30] that there are conformational transitions (the distance between chromophores changes simultaneously), and these transitions between stable conformations occur suddenly and randomly in time and space. This is the so-called hopping model of polymer motion. After each hop, the distance between chromophores changes significantly in comparison with the Förster radius. By introducing the mean time of a conformation change τ_{conf} and assuming the conformational change to be a Poisson process, the following integral equation for the

luminescence kinetics $I_{hop}(N, t)$ was obtained [7, 20, 30]:

$$I_{hop}(N, t) = I(N, t) \exp\left(-\frac{t}{\tau_{conf}}\right) + \frac{1}{\tau_{conf}} \int_0^t I(N, t_1) \exp\left(-\frac{t_1}{\tau_{conf}}\right) I_{hop}(N, t-t_1) dt_1, \quad (17)$$

where $I(N, t)$ is the luminescence intensity in the absence of conformational changes, as described by Eq. (5) or (7) for the ideal or the nonideal chain, respectively.

After introducing the dimensionless time as given by Eq. (4), Eq. (17) was solved numerically for the ideal polymer chain: $I(N, t) = I_{id}(N, t)$ in Eq. (17). The calculations have shown [7] that it is necessary to take into account the polymer motion if the dimensionless time of the change of chain conformation $T_{conf} = R_0^6 \tau_{conf} / \langle R_g^2 \rangle^3 \tau < 20$.

We have found that, as T_{conf} decreases, the luminescence kinetics becomes exponential: $I_{hop}(N, t) \sim \exp(-k_{hop}t)$, where k_{hop} is the rate constant. In the limit of very small values of T_{conf} , we derived the rate constant k_{hop}^{id} for the ideal polymer chain as [7]

$$k_{hop}^{id} = \frac{1}{\tau_{conf}} T_{conf}^{1/2} = \frac{1}{\tau_{conf}} \frac{R_0^3}{\langle R_g^2 \rangle^{3/2}} \left(\frac{\tau_{conf}}{\tau}\right)^{1/2} \quad (18)$$

and the rate constant $k_{hop}^{nid} = T_{conf}^{56/108} / \tau_{conf}$ for the nonideal chain. One can see that the difference between k_{hop}^{id} and k_{hop}^{nid} is insignificant.

Comparison of the expressions for k_{dif} and k_{hop} shows that the two motion types (diffusion and hopping) depend in different manners on R_0 and $\langle R_g^2 \rangle$. Hence, it is possible to establish, in principle, the type of motion of a polymer molecule.

Polymer Chain with Random Distribution of Chromophores Along the Chain

For polymer molecules with the random distribution of chromophores along the chain, the following equation holds for the luminescence kinetics [15]:

$$I_{dif}(t) = \exp\left[-c \int_{-\infty}^{\infty} dN \int_{r_0}^{\infty} dr 4\pi r^2 (g_N(r) - g_N^*(r, t))\right]. \quad (19)$$

From Eq. (19), it follows that it is necessary first to numerically solve Eq. (14) for each value of N and then to average over N . This problem is difficult to fulfill. Instead of this, we obtained an analytical solution to

Eq. (14) in the limit of weak diffusion. As a result, the luminescence intensity $I_{dif}^{id}(t)$ is expressed by [14, 15]:

$$I_{dif}^{id}(t) = \exp\left[-8.125c \frac{R_0^2}{a^2} \left(\frac{t}{\tau}\right)^{1/3} - 12c \frac{Dt}{a^2}\right], \quad (20)$$

for the ideal polymer chain and

$$I_{dif}^{nid}(t) = \exp\left[-3.872c \left(\frac{R_0}{a}\right)^{5/3} \left(\frac{t}{\tau}\right)^{23/54} - 6.535c \frac{D\tau}{a^2} \left(\frac{a}{R_0}\right)^{1/3} \left(\frac{t}{\tau}\right)^{17/18}\right]. \quad (21)$$

for the nonideal polymer chain.

Equations (20) and (21) are valid if the second term in brackets is smaller than the first term (in the time interval of an order of τ). This means that inequality $D\tau < R_0^2$ has to be fulfilled. The second terms in Eqs. (20) and (21) are the corrections to static quenching caused by the diffusion motion of polymer chain units (see Eqs. (9) and (10)). One can see that the polymer motion enhances luminescence quenching.

Equation (14) can also be solved in the limit of very fast diffusion. In this case, the diffusion is so strong that a decrease in the function $g_N^*(r, t)$ at short distances r is immediately compensated by diffusion from farther regions. Therefore, the distribution function $g_N^*(r, t)$ conserves its form coinciding with the form of $g_N(r)$, but its absolute value decreases exponentially with time.

In this limit, we obtained [14, 15]

$$I_{dif}^{id}(t) = \exp\left[-5.358c \frac{R_0^4}{r_0^2 a^2} \left(\frac{t}{\tau}\right)^{2/3}\right], \quad (22)$$

for the ideal chain and

$$I_{dif}^{nid}(t) = \exp\left[-3.605c \left(\frac{R_0^6}{r_0^{3-5/18} a^{3+5/18}} \frac{t}{\tau}\right)^{30/59}\right]. \quad (23)$$

for the nonideal chain. It is seen that the luminescence kinetics remain nonexponential even in the limit of very fast diffusion. This is due to the fact that donor-acceptor pairs move within a restricted space, which may have different sizes determined by N , the number of units between a donor and an acceptor.

In the case of the hopping mechanism of motion, it is more convenient to rewrite the luminescence kinetics equations for $I_{hop}(t)$ in the form [15]

$$I_{hop}(t) = \exp\left[-c \int_{-\infty}^{\infty} dN (1 - I_{hop}(N, t))\right]. \quad (24)$$

The calculation of the kinetics with the use of Eq. (24) is a difficult task, because it is first necessary to find $I_{hop}(N, t)$, solving numerically Eq. (17) for each value of N separately, and then to average over N . Analytical solutions were obtained only in the limit of very small τ_{conf} . In this limit, we got for the luminescence kinetics of the ideal polymer chain $I_{hop}^{id}(t)$ [15]

$$I_{conf}^{id}(t) = \exp\left[-5.358c \frac{R_0^2}{a^2} \left(\frac{\tau_{conf}}{\tau}\right)^{1/3} \left(\frac{t}{\tau_{conf}}\right)^{2/3}\right], \quad (25)$$

and for the nonideal chain ($I_{hop}^{nid}(t)$)

$$I_{hop}^{nid}(t) = \exp\left[-3.605c \frac{R_0^{5/3}}{a^{5/3}} \left(\frac{\tau_{conf}}{\tau}\right)^{5/18} \left(\frac{t}{\tau_{conf}}\right)^{30/59}\right]. \quad (26)$$

Comparing the luminescence kinetics as calculated according to Eqs. (25) and (26) for the hopping model of the motion of polymer chain units with the kinetics (22, 23) calculated with the diffusion model, we see that they depend in the identical manner on time but in different manners on R_0 . This observation can be used for revealing the type of polymer motion in a solvent (by substitution of chromophores attached to a polymer chain).

CONCLUSIONS

The kinetics of irreversible energy transfer between chromophores attached to an isolated flexible polymer chain were studied. In the static case, when the polymer chain does not change its conformation during the lifetime of an excited chromophore state, simple equations were derived for the luminescence kinetics for ideal and nonideal polymer chains.

In the dynamic case, the diffusion and hopping models of the motion of a polymer molecule were studied. It has been shown that it is necessary to take into account these types of motion when the dimensionless diffusion constant d is large enough ($d > 10^{-2}$, in the diffusion limit) or when the dimensionless time of the conformation variation is short ($T_{conf} < 20$, in the hopping limit). Useful analytical expressions were derived.

It should be noted once again that the overall luminescence intensity differs from that given in this article by a factor of $\exp(-t/\tau)$ taking into account the finiteness of the lifetime of the excited state of a donor chromophore.

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