Eur. Phys. J. B **15**, 187–192 (2000)

THE EUROPEAN
PHYSICAL JOURNAL B
EDP Sciences

Reversible charge transfer in solids; binary approximation

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Received 26 May 1999

Abstract. A common theory of reversible charge transfer (RCT) kinetics for low and high excitation power with the use of the conditional concentration of acceptors is presented. A comparison with recently published RCT theory is given. The results are similar or identical only in restricted or fractal spaces of low dimension, and in the case of low concentration of acceptors, *i.e.* in all the cases when the conditions of the binary approximation are fulfilled. The deviation from the binary approximation results in a physically incorrect picture, the cause being due to the different space averaging procedure. The probability of the donor's cation state is an increasing function of the concentration of the acceptor and of the characteristic parameters of the rate constants.

PACS. 78.20.Bh Theory, models, and numerical simulation – 89.90.+n Other topics of general interest to physicists

1 Introduction

Much interest has been devoted recently to the study of the reversible charge transfer (RCT) kinetics in infinite and restricted solutions in terms of the molecular parameters [1–10]. The theoretical results of various works on this problem are essentially different [5–10], as a result of different space averaging procedures. The proper treatment of the back charge transfer in the kinetic theory of this process is of crucial importance. It has been pointed out that the problem of the back charge transfer, which prevents the charge separation, is complex and leads to the complex averaging procedure [5–9]. In reference [10] there are presented and compared several methods for description of the charge transfer kinetics. Such important and interesting problems as dependencies of the cation state probability on the acceptor concentration C_A and on the parameters of the rate constant of charge transfer, have not been discussed so far. However, they give important information about the validity of the available formulations of this problem.

In this work we present a common theory of RCT both for low and high excitation power. We compare some dependencies of the cation state probability $P^+(t)$ of the donor molecule according to the results of two theories [5–7], which were derived for the low concentration of the donors surrounded by randomly distributed acceptors in solids. The method developed in reference [5] uses the so called "novel" averaging procedure (NAP). Our analysis shows that this NAP method is valid only if the criterion of the binary approximation can not be violated until the donor excited state probability can be approximated by the condition $N(t) \sim \exp(-t/\tau)$, where τ is the life-time of the donor's excited state.

2 The common theory of the reversible charge transfer in solids

Here we give a more detailed description of the charge separation kinetics, which has been discussed recently [5–10]. The probability of the cation state of donor S may be obtained [6,7] using a method first proposed by Antonov-Romanovskii and Galanin [11] for the electronic excitation energy transfer in a solid solution.

Let the $4\pi C_\mathrm{A} p^+(r,t) r^2 \mathrm{d}r$ be the average number of acceptor anion molecules A^- which are located at a distance r, $r+\mathrm{d}r$ from the donor-cation molecules S^+, calculated per one excited molecule S^* [6,7,14,16b]. C_A is the concentration of acceptor molecules, which can be also a function of time $C_\mathrm{A}(t)$ in the general case. $p^+(r,t)$ is the probability of cation-anion separation by distance r, $r+\mathrm{d}r$. The variation of conditional acceptor concentration [11] $c^+(r,t) = C_\mathrm{A} p^+(r,t)$ with intermolecular distance r occurs in time due to the forward electron transfer between S^* and A molecules, and due to the back charge transfer from A^- to S^+. The donor molecule can be in three states: excited, ground, and cation state. For the relevant probabilities N, it exists:

$$N^*(t) + N_\mathrm{gr}(t) + N^+(t) = 1. \tag{1}$$

We are interested in the time dependence of the probability of the donor non-cation state $N_\mathrm{nc}(t)$ [6,7]:

$$N_\mathrm{nc}(t) = N^*(t) + N_\mathrm{gr}(t), \tag{2}$$

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for which it is easy to write kinetic equations describing RCT. The probability of the cation state is given by:

$$N^+(t) = 1 - N_{\rm nc}(t) \tag{3}$$

with initial conditions

$$N_{\rm nc}(0) = 1; \quad N^+(0) = 0. \tag{4}$$

The rate equation for $N_{\rm nc}(t)$ can be written as:

$$\mathrm{d}N_{\rm nc}(t)/\mathrm{d}t = -F(t)4\pi \int_{R_{\rm m}}^{\infty} \partial p^+(r,t)/\partial t\, r^2 \mathrm{d}r. \tag{5}$$

From the above definition of $c^+(r,t)$ it follows that $F(t)$ is the concentration of the excited donor-acceptor pairs, as the integral term in equation (5) determines the derivative of the donor cation-state probability calculated per one initially excited donor-acceptor pair. $R_{\rm m}$ is the sum of radii of two molecules. For point particles, $R_{\rm m}$ is equal to zero. If charge back transfer takes place, we can write:

$$F(t) = N_{\rm nc}(t)[C_{\rm nc}(t) + C_{\rm A}(t)] \tag{6}$$

as a function of the acceptor concentration.

For low excitation power, ($C_{\rm nc}(t) \ll C_{\rm A}(t)$), the concentration of donor molecules in the non-cation state is derived from the equation [6,7]:

$$\mathrm{d}N_{\rm nc}(t)/\mathrm{d}t = -[N_{\rm nc}(t)]4\pi C_{\rm A} \int_{R_{\rm m}}^{\infty} \partial p^+(r,t)/\partial t\, r^2 \mathrm{d}r. \tag{7}$$

In the case of high excitation power, ($C_{\rm nc}(t) \gg C_{\rm A}(t)$), we get using the equation (6) [16]:

$$\mathrm{d}N_{\rm nc}(t)/\mathrm{d}t = -N_{\rm nc}^2(t)4\pi C_{\rm so} \int_{R_{\rm m}}^{\infty} \partial p^+(r,t)/\partial t\, r^2 \mathrm{d}r. \tag{8}$$

$C_{\rm so}$ is the initial concentration of S*. The differential operator $\partial/\partial t$ in the integral of equations (7) and (8) means that the change of $p^+(r,t)$ is due to the charge transfer and to the molecular motion and $C_{\rm A}(t)$ is independent of this operator. The equations for $N_{\rm nc}(t)$ are reduced to the modified equations of the theory of the electron energy transfer of Forster and Dexter [11–15], when molecular diffusion is also taken into account [14,15].

Deactivation of donor occurs due to natural decay of its excitation and to intermolecular charge transfer with the rate constants [12,13]:

$$k_{\rm o} = 1/\tau, \tag{9}$$
$$k_{\rm f} = (1/\tau)\exp[(R_{\rm f} - r)/a_{\rm f}], \quad \text{forward transfer}, \tag{10}$$
$$k_{\rm b} = (1/\tau)\exp[(R_{\rm b} - r)/a_{\rm b}], \quad \text{back transfer}. \tag{11}$$

k with the appropriate subscript denotes the rate constant of the relevant process. r is the donor-acceptor separation. $R_{\rm f}$, $R_{\rm b}$, $a_{\rm f}$ and $a_{\rm b}$ are the molecular parameters, which characterize the distance scales of the forward and the back charge transfer rates. τ is the life time of the donor's excited state. $c^+(r,t)$ satisfies the equation for solids:

$$\dot{c}^+(r,t) = k_{\rm f}(r)n(r,t) - k_{\rm b}(r)c^+(r,t), \tag{12}$$
$$c^+(r,0) = 0. \tag{13}$$

The pair distribution function of reactants, $n(r,t)$, satisfies the equation:

$$\dot{n}(r,t) = -k_{\rm f}(r)n(r,t) - 1/\tau n(r,t). \tag{14}$$

By using the correlation between the probabilities of the donor cation and the non- cation states (Eq. (3)), we can use the equality of their derivatives

$$\dot{N}^+(t) = -\dot{N}_{\rm nc}(t) \tag{15}$$

and get the cation-state probability of donor for low excitation power:

$$N^+(t) = 1 - \exp[-4\pi C_{\rm A} \int_{R_{\rm m}}^{\infty} p^+(r,t) r^2 \mathrm{d}r], \tag{16}$$

and for high excitation power:

$$N^+(t) = 1 - [1 - 4\pi C_{\rm so} \int_{R_{\rm m}}^{\infty} p^+(r,t) r^2 \mathrm{d}r]^{-1}. \tag{17}$$

3 The NAP method

In the NAP model the "novel" averaging procedure is used to treat the kinetics of the forward and back charge transfer [5]. Instead of solving the differential equation for the radical-ion population and then averaging over all of the acceptor configurations, the equation is first averaged over $N-1$ acceptors, and then solved. The thermodynamic limit is taken in order to get the total cation state probability. This method is discussed further in references [5,8–10]. At time $t = 0$ the ensemble of donors is optically excited by the low power source. The cation state probability of the donor is given by this model as:

$$P^+(t) = 4\pi C_{\rm A} \int m^+(r,t) r^2 \mathrm{d}r. \tag{18}$$

According to NAP model for solids, $m^+(r,t)$ satisfies the equation [5,9]:

$$\dot{m}^+(r,t) = k_{\rm f}(r)n(r,t)N(t) - k_{\rm b}(r)m^+(r,t). \tag{19}$$

Both forward and back charge transfer are taken here into account.

At time $t = 0$ and for $r > R_{\rm m}$ the acceptor molecules are uniformly distributed, the function and $m^+(r,0) = 0$. $N(t)$ is the excited state probability of donors, which obeys the conventional kinetic equation [11–15]:

$$\mathrm{d}N(t)/\mathrm{d}t = -C_{\rm A}N(t) \int k_{\rm f}(r)n(r,t)\mathrm{d}^3 r - N(t)/\tau. \tag{20}$$

In this model the pair distribution function of reactants is evaluated from:

$$\dot{n}(r,t) = -k_{\mathrm{f}}(r)n(r,t), \tag{21}$$

$$n(r,0) = 1. \tag{22}$$

It should be remarked that the natural lifetime of a donor's excitation τ is introduced in different ways in the present and in the NAP theory (Eqs. (14, 20)) [9]. Calculations show that in the absence of back charge transfer, both theories yield identical results for the donor cation state probability (Eqs. (16, 18)).

The equation (19) for solids results in [5]:

$$P^{+}(t) = 4\pi C_{\mathrm{A}} \int\limits_{R_{\mathrm{m}}}^{\infty} k_{\mathrm{f}}(r) \exp[-k_{\mathrm{b}}(r)t] \times \int\limits_{0}^{t} \exp\{-[k_{\mathrm{f}}(r) - k_{\mathrm{b}}(r)]t'\} N(t')\mathrm{d}t' r^2 \mathrm{d}r. \tag{23}$$

The donor excited state probability $N(t)$ is given by [14,15]:

$$N(t) = \exp(-t/\tau)\exp\left\{-4\pi C_{\mathrm{A}} \int\limits_{R_{\mathrm{m}}}^{\infty} [1-\exp\big(-k_{\mathrm{f}}(r)t\big)] r^2 \mathrm{d}r\right\} = \exp(-t/\tau)S(t). \tag{24}$$

The essential difference between equation (19) in the NAP model and equation (12) in the presented model [6] is determined only by the function $N(t)$ in equation (19). As will be seen below, only the approximation

$$N(t) \sim \exp(-t/\tau) \tag{25}$$

in equations (18–23) results in a physically reasonable behaviour of the donor cation state probability for varying reaction parameters. At a low concentration C_{A} of molecules A, or at small $k_{\mathrm{f}}(r)$, the integrand in equation (16) has a small value α. The decomposition of equation (16) with a low parameter α gives the result [5,9] – equation (18).

In equations (16) or (24), the acceptor concentration may be high. The only condition which must be fulfilled is that acceptors deactivate the donor independently from each other: the presence of any acceptor does not influence the action of the other acceptors [15].

4 Discussion

In a solid solution and in a binary approximation, the donor cation state probability, because of reversible charge transfer independently of the space character (the Euclidean infinite or restricted space or fractal media of various dimensions), must satisfy the following conditions as the effect of the principle of causality:

(i) The probability $P^{+}(t)$ must be an increasing function of the acceptor concentration for any ratio of the rate constants of the forward and back charge transfer. This probability is an increasing function of C_{A} in the case of $k_{\mathrm{b}}(r) = 0$ [17]. Such behaviour is expected also in the case when $k_{\mathrm{b}}(r) > 0$. The following consideration illustrates this statement:
Let an excited donor molecule be found at the center of some sphere with an acceptor molecule on its surface. Then, with the increase of the number of the acceptors on this surface, the maximum value $P^{+}_{\max}(t)$ of probability $P^{+}(t)$ can only increase, independent of back transfer reaction, provided that the acceptors act independently on the donor: every donor with surrounding acceptors represents an independent "vessel" [14b]. In regular (or fractal [19]) space (a set of spherical layers with definite thickness) with many donors ("vessels") and acceptors, this time-dependent character cannot change if the charge transfer rate constants do not change. With the increase of the number of acceptors in any layer, the donor cation state probability can only increase, since every acceptor acts independently [15].
From this discussion it follows also:

(ii) $P^{+}(t)$ must be an increasing function of the characteristic distance $R_{\mathrm{f}} = R_{\mathrm{b}}$ in any case.

(iii) $P^{+}(t)$ must be a decreasing function of a_{b} (determined by the overlap of the wave functions of ions) and an increasing function of a_{f}, which is determined by the overlap of the wave functions of neutral molecules of the donor and the acceptor.

In the calculations, the numerical procedure based on the expanded DCR program [23] was used. The donor excited state natural decay, was not taken into account since the donor's excitation lifetime was already included by multiplying with the factor $\exp(-t/\tau)$ in equation (24).

4.1 RCT in confined space

The fractal structures are self-similar and display dilational symmetry, which determines their site density function $g_{\mathrm{fr}}(r)$ in terms of the Euclidean dimension d and the Hausdorf fractal dimension $\overline{d}$ ($\overline{d} \leq d$) [19–22]. RCT kinetics in a fractal space has been considered in reference [17] in which the above argument and results are depicted. The fractal structure of the reaction space results in a slowing down of the charge separation [17]. The following discussion is based on conclusions reached in that work.

In a restricted space, we must introduce into the space integrals the site density characteristic for a sphere of radius R [19–22]:

$$g_{\mathrm{rst}}(r) = 2\pi r^2(1 - r/2R)\rho. \tag{26}$$

$2\pi r^2\rho$ is the limiting value of $g(r)$ for $R = \infty$. The Forster-type decay follows for $R \to \infty$ by taking $g_{\mathrm{rst}}(r) = 2\pi r^2\rho =$ const [21]. Space restriction slows down the cation state

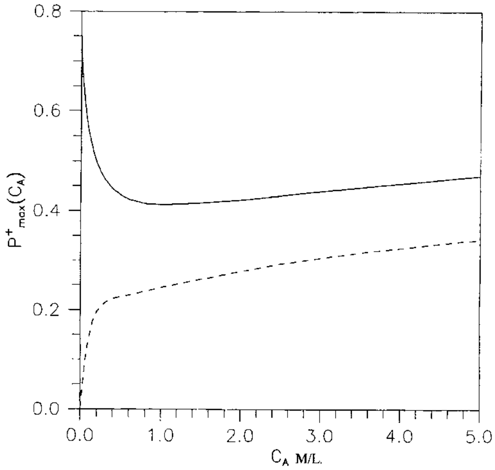


Fig. 1. The dependence of $P^+_{\rm max}(C_{\rm A})$ on the acceptor concentration according to the NAP model, equation (18). $R_{\rm f} = R_{\rm b} = 12$ Å, $a_{\rm f} = 3$ Å, $a_{\rm b} = 1$ Å, $R_{\rm m} = 5$ Å. Bold line, infinite Euclidean space. Dotted line, restricted space: $R_{\rm m} = 4$ Å, $R_{\rm max} = 11$ Å.

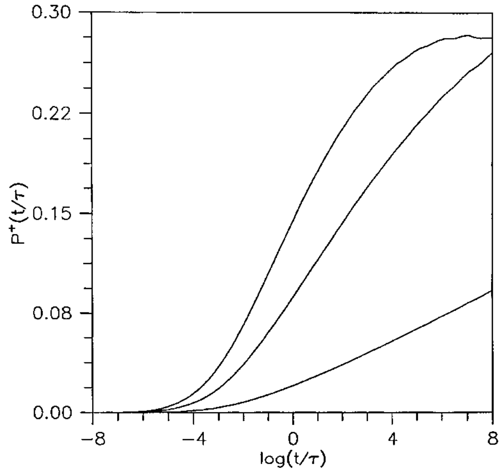


Fig. 2. The time-dependence of $P^+(t)$ according to the equation (18) for different concentrations of the acceptors (from the top to the bottom) $C_{\rm A} = 1$; 0.5 and 0.1 M/l for fractal space with fractal dimension $\overline{d} = 1,5$. $R_{\rm f} = R_{\rm b} = 10$ Å, $a_{\rm f} = 2$ Å, $a_{\rm b} = 1$ Å, $R_{\rm m} = 5$ Å.

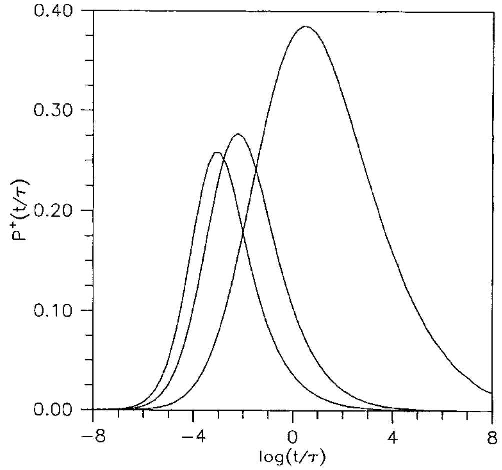


Fig. 3. The same dependence as in Figure 2, (from left to right) $C_{\rm A} = 1$; 0.5 and 0.1 M/L. $R_{\rm f} = R_{\rm b} = 10$ Å, $a_{\rm f} = 2$ Å, $a_{\rm b} = 1$ Å, $R_{\rm m} = 5$ Å. Euclidean space, $d = 3$.

probability. In such space, the value of $N(t)$ can be approximated by $\exp(-t/\tau)$. As a consequence, the probability $P^+(t)$ becomes an increasing function of the acceptor concentration. This is demonstrated in Figure 1. The back charge transfer does not affect this behaviour. In restricted space, according to both theories, the maximum of cation state probability $P^+_{\rm max}(t)$ is a non-decreasing function of the acceptor concentration.

In order to take into account the fractal properties of the solid space, it is necessary to include in space integrals (Eqs. (16–18, 23, 24)) the site's density function [19–21]:

$$g_{\rm fr}(r) = Ar^{\overline{d}-d}, \tag{27}$$

where A is the proportionality constant. In the case of the maximum of the fractal dimension the behaviour of $P^+(t)$ repeats the behaviour, founded in regular three-dimensional Euclidean space. For a low fractal dimension, we have the correct dependencies of $P^+_{\rm max}(t)$ (Fig. 2). Thus, in restricted and fractal spaces, for which the space integrals in equations (17) and (19) are small, the donor cation state probability $P^+(t)$ has the correct behaviour – it satisfies conditions (i)–(iii). As we shall see below, there exists a reverse picture in Euclidean infinite space.

4.2 RCT in Euclidean space

In Euclidean infinite space $N(t) < N_{\rm q}(t)$, where q denotes restricted or fractal space [20,21]. The first term in the right side of equation (19) decreases with the increase of $C_{\rm A}$ and $R_{\rm f}$, and is sensitive to the space character. In particular, it is minimum in infinite Euclidean space, and this, in turn, increases the role of the second term on the right hand side of equation (19). The effect of charge back transfer increases with the increase of the reaction space and its dimension. Such an increase is not physically meaningful.

Considering condition (i), the cation state probability (for any back transfer rate constant and at any time t) must be a non-decreasing function of the acceptor concentration as well as of the rate constant of the forward charge transfer. The function (18) does not satisfy these requirements (Figs. 1–3). Here it is assumed that the concentration of acceptors is much larger than that of the donor, the acceptor-acceptor excluded volume effects being neglected.

Figure 1 shows the dependence of $P^+_{\rm max}$ – as a function of the acceptor concentration $C_{\rm A}$. There is a decreasing branch followed by an increase, in contradiction to condition A. However, in the restricted space the decreasing branch is missing. Calculations for the restricted space were carried out by using equation (19) and including the factor $g_{\rm rst}(r)$ in the integrals. The presence of the decreasing branch is a consequence of the inclusion of $N(t) = \exp(-t/\tau)S(t)$ in equation (19). In restricted space, the integral in equation (24) is small and as

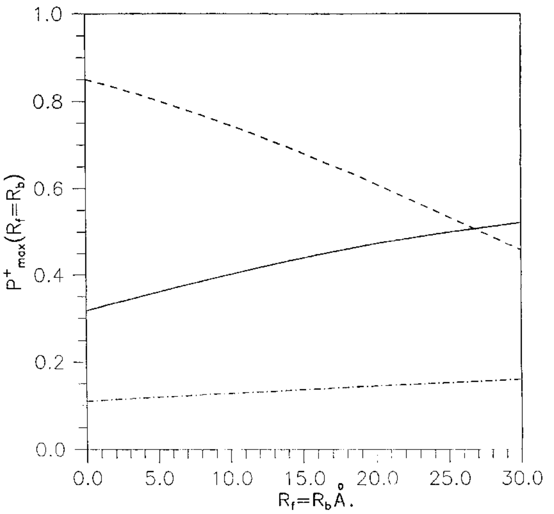


Fig. 4. The dependence of $P^{+}_{\mathrm{max}}(R_{\mathrm{f}} = R_{\mathrm{b}})$; $a_{\mathrm{f}} = 3$ Å, $a_{\mathrm{b}} = 1$ Å. NAP model, equation (18). Dotted line - $C_{\mathrm{A}} = 0.01$ M/L. Bold line - $C_{\mathrm{A}} = 0.001$ M/L. Dash-dot line - $C_{\mathrm{A}} = 0.01$ M/L - fractal space with fractal dimension $\overline{d} = 2$.

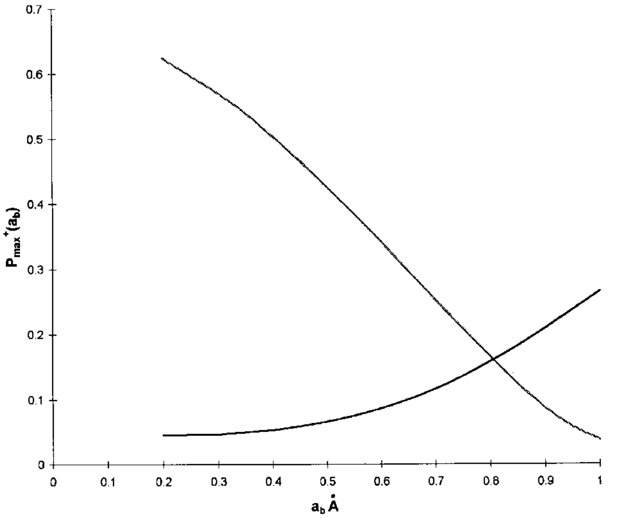
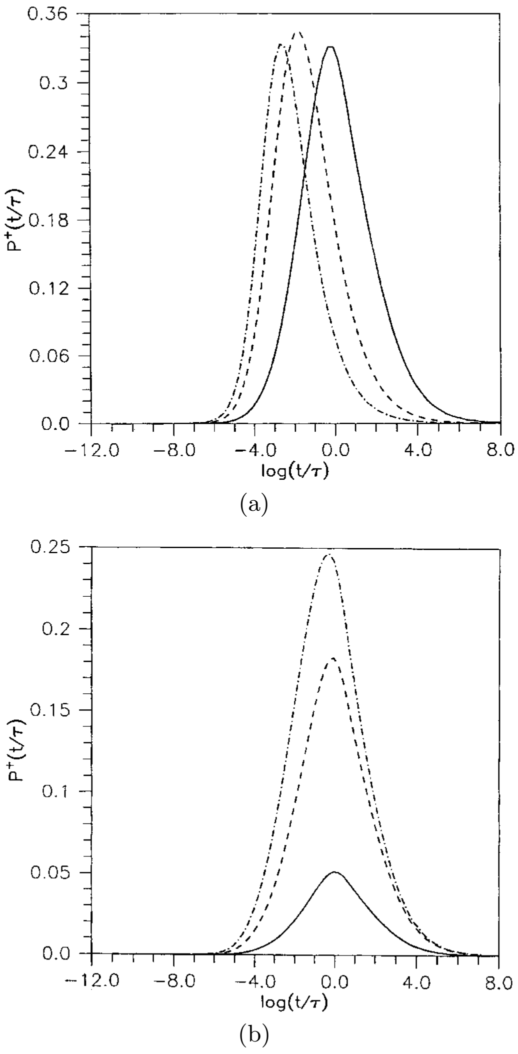


Fig. 5. The $P^{+}_{\mathrm{max}}(a_{\mathrm{b}})$ as function of a_{b} according to the equation (18). $R_{\mathrm{f}} = R_{\mathrm{b}} = 12$ Å, $a_{\mathrm{f}} = 1$ Å. Lower line - $C_{\mathrm{A}} = 1$ M/L. Upper line - $C_{\mathrm{A}} = 0.01$ M/L.



(a)

(b)

Fig. 6. The different t_{max}, for which $P^{+}(t)$ has maximum value. $R_{\mathrm{f}} = R_{\mathrm{b}} = 10$ Å, $a_{\mathrm{f}} = 2$ Å, $a_{\mathrm{b}} = 1$ Å. (a) Euclidean space, (different t_{max}). $C_{\mathrm{A}} = 1$; 0.5; 0.1 M/L (from left to right). (b) Fractal space; $\overline{d} = 2$ (coinciding t_{max}).

a result, $N(t) \sim \exp(-t/\tau)$. When $S(t) \approx 1$ the donor cation state probability becomes a normal increasing function of the acceptor's concentration. Any circumstance, which gives $S(t) \approx 1$, ensures correct behaviour of the probability $P^{+}(t, C_{\mathrm{A}})$; it is required by the binary approximation. If $S(t) \ll 1$ (violating the criterion of binary approximation) the behaviour of the curves is different: the decreasing curve with increase of C_{A} does not correspond to physically meaningful behaviour. In the NAP model, if C_{A} increases the effect of back transfer increases with no limit, while the increase of the effect of forward transfer is restricted by the nonexponential law $N(t)$.

Figure 4 shows the dependence P^{+}_{max} on $R_{\mathrm{f}} = R_{\mathrm{b}}$. In infinite regular space, this function is (in NAP model) a decreasing function of $R_{\mathrm{f}} = R_{\mathrm{b}}$. However, in restricted, fractal space of low dimension or at low concentration of acceptors, $P^{+}_{\mathrm{max}}(R_{\mathrm{f}} = R_{\mathrm{b}})$ is an increasing function. Here we can see again the disturbing influence of $N(t)$ in equation (19).

Figure 5 shows the dependence P^{+}_{max} on a_{b}. Here we see the increase of P^{+}_{max} with increase of a_{b} – the characteristic of the ions' wave function's overlap (donor-cation and acceptor-anion). Here also a violation of condition (iii) takes place.

Hence, the behaviour of functions (18) and (19) does not correspond to the physical sense of the donor-cation state probability when the conditions of the binary approximation are not strictly fulfilled. As we see, the condition $S(t) \approx 1$ provides the correct behaviour of the cation state probability P^{+}; this is the demand of the binary approximation. If $S(t) \ll 1$ (in the case of breaking the criterion of the binary approximation) the behaviour of the curves in two theories, references [5,6], is different: in the second case we cannot explain the curves behaviour. The considered theories take into account the back charge transfer differently. In the case of $k_{\mathrm{b}}(r) = 0$ the NAP results are correct for any value of C_{A}, R_{f}, t and the reaction volume [9], and the results of the theories [5,6] coincide.

From the expressions (18) and (19) it follows that the cation state probability $P^{+}(t)$ has maximum value $P^{+}_{\max}(t)$ at different times at different acceptor concentrations C_{A}, keeping the rate constants of the charge transfer k_{f} and k_{b} at constant values (Fig. 6). However, it is physically clear that these maximum meanings of $P^{+}(t)$ for different C_{A} must happen at the same time, if the rate constants are not varied. The following simple argument illustrates this statement. If an excited donor molecule is found at the center of a sphere (of radius R) with an acceptor molecule on its surface, then with the increase of the number of acceptors, the maximum value of $P^{+}(t)$ appears for the same invariable time. This time does not change with variation of the acceptors quantity if the acceptors influence on the donor is independent: every donor with surrounding acceptors represents an independent system [14d]. Now, in the regular (or fractal [19]) space (set of the spherical layers with definite thickness) with many donors and acceptors this time-dependence character can not change if the charge transfer rate constants are the same. Hence, the behaviour of functions (18) and (19) does not correspond to the physical sense of the donor-cation state probability. Figure 6 shows that the time $t_{\max}$, for which $P^{+}(t)$ has a maximum value, depends on the acceptor concentration in Euclidean space, and $t_{\max}$ does not depend on C_{A} in fractal space.

So, the considered theories take into account the charge back transfer differently. According to the NAP model, the effect of back transfer increases without limit, if C_{A} increases and $S(t) \ll 1$, while the increase of the forward transfer effect is restricted by the non-exponential law $N(t)$. The condition $S(t) \approx 1$ ensures a correct behaviour of the cation state probability.

Our thanks are due to Professors J. Klafter, D. Avnir and M. Kramer for helpful discussions. This work was supported by Academy of Humanities and Sciences of Israel and the Ministry of Absorption of Israel.

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