Redistribution of electronic excitation energy in impurity crystals

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A model which permits obtaining kinetic law for excitation decay in the presence of clusters of impurity ions of arbitrary length in a dielectric crystal matrix is proposed. The phenomenon of electronic excitation trapping in impurity paired centers (PCs) in doped dielectric crystals is investigated by using the theory of the Markovian processes. It is shown that the delay of excitation lifetime in PCs can exert a vital influence on the kinetics of excitation decay in the whole impurity subsystem. The dependences of excitation lifetime on the concentration of impurity ions and pump power are found.

I. INTRODUCTION

Cooperative processes taking place in doped dielectric crystals, in particular, the processes of nonradiative transfer of the electronic excitation energy (NTEEE) between impurity ions, lead to redistribution of excitation energy in the impurity subsystem. This essentially affects the kinetics of excitation decay, dynamics of population and depopulation of electronic levels of impurity ions, etc.[1–5] At least two problems arise when studying the processes of excitation relaxation in an impurity subsystem: (i) determination of probability of elementary NTEEE acts in donor-acceptor pairs of impurity ions induced by multipole-multipole, exchange, and other interactions of these ions[3–8] and (ii) summation of NTEEE probability over a variety of donor and acceptor ions, the result of which certainly depends on distribution of impurity ions in the crystal matrix.[1,3,9–11]

The case of inhomogeneous distribution of impurity ions in a crystalline matrix is of certain interest because it leads to the origin of clusters of two or more impurity ions between which effective NTEEE takes place.[12,13] As a result, the lifetime of excitation in such formations is raised (excitation trapping), which obviously influences kinetics of excitation decay in the impurity subsystem as a whole and, hence, the spectroscopic characteristics of materials.[14–21]

The model proposed in the present paper allows obtaining kinetic law for electronic decay in an impurity system taking into consideration the NTEEE processes between impurity ions in the presence of impurity paired centers (PCs) in a crystal matrix. It must be noted that strongly inhomogeneous distribution of impurity ions in a crystal matrix substantially complicates analytical solution of kinetic equations initially written for the whole impurity system.[15–17] Consequently, in the model proposed, the whole impurity system is split into two subsystems, namely, a PC subsystem and a solitary impurity center subsystem, with *a priori* different excitation decay times. In addition, we imply the following assumptions: (i) the impurity ions are local centers of small radius (i.e., optical electrons of such impurity ion are localized close to nucleus of impurity ion); (ii) impurity ions, which form PC, retain their identity (i.e., the interaction energy of two impurity ions is much less than the energy of intra-atomic interactions); and (iii) the impurity ions are excited by pulses of much shorter duration as compared to that of relaxation processes in a single impurity ion.

In order to calculate excitation lifetime in a PC, the kinetics of excitation decay in a PC is studied for a given value of probability of elementary NTEEE acting between the ions composing a PC. The excitation lifetime in a solitary center determined by intracenter relaxation processes is considered known. Further, the excitation decay law over the whole impurity system is determined taking into account NTEEE processes between solitary and paired centers. Such an approach allows one to significantly simplify calculations of kinetic parameters of the whole system and, on the other hand, to perform calculations also in the case of the presence of more complex cluster formations consisting of three or more impurity ions.

II. EXCITATION DECAY IN PAIRED CENTER

Let us consider the process of excitation decay in a PC consisting of similar impurity ions A, one of which is initially ($t=0$) excited, while the other is in the ground state. Let us denote the number of such a (A-A*) PC by n_{10}. We suppose that the probabilities of the direct and opposite processes of NTEEE in PC are the same, and the excitation can be frequently transferred from one impurity ion to another until it is lost. For the description of the excitation decay process in the PC, we use the theory of the Markovian processes.[22] The process, in which the excitation relaxes after $(n-1)$ jumps, can be described by the directed Markovian chain of n length, where each site corresponds to a single excitation state (Fig. 1).

Excitation evolution described by the chain of n length is determined by the system of the Kolmogorov equations,
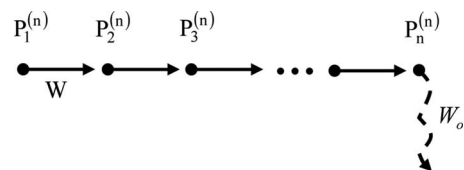
$$\dot{P}_1^{(n)}(t) = -WP_1^{(n)}(t),$$

FIG. 1. Markovian chain of n length; W and W_0 are the probabilities of a jump and intercenter transitions and $P_k^{(n)}(t)$ is the probability that the excitation is on the kth point of the chain of n length.

$$\dot{P}_2^{(n)}(t) = -WP_2^{(n)}(t) + WP_1^{(n)}(t),$$

$$\dot{P}_n^{(n)}(t) = -W_0 P_n^{(n)}(t) + WP_{n-1}^{(n)}(t), \quad (1)$$

where $P_k^{(n)}(t)$ is the probability to find an excitation at time t in the kth site of the chain of n length, W_0 is the probability of intracentral (radiative or nonradiative) relaxation of excitation, and W is the probability of a jump from one site to another. In particular, $P_n^{(n)}(t)$ is the probability of excitation loss in the nth site of the chain of n length after $(n-1)$ jumps. Solving the equation system [Eq. (1)] with the initial condition $P_k^{(n)}(0) = \delta_{k,1}$, we obtain

$$P_n^{(n)}(W,t) = \left(\frac{W}{W-W_0}\right)^{n-1}$$
$$\times e^{-W_0 t}\left[1 - \frac{\Gamma[n-1,(W-W_0)t]}{\Gamma(n-1)}(1-\delta_{n,1})\right], \quad (2)$$

where $\Gamma(n,t)$ is the incomplete gamma function and $\Gamma(n)$ is the gamma function. In particular, $n=1$ corresponds to the case of excitation decay without jumps; for $n=2$, the excitation decays after a single jump, etc.

Obviously, the realization of a certain length chain depends on the magnitude of the ratio $a = W/W_0$: the higher a is the higher the probability of realization of a longer chain is. Let us denote the probability distribution of realizations of a chain with the n length by $Q_n(a)$. It is evident that the function $Q_n(a)$ should have the following properties: (1) $Q_n(0) = \delta_{n,1}$, i.e., at $a=0$ (absence of jumps) the chain with a single site is surely realized; (2) $\lim_{n\to\infty} Q_n(a) = 0$ for all finite positive values of parameter a, i.e., at finite values of the jump probability, the excitation lifetime in the PC is finite; and (3) $\sum_{n=1}^{\infty} Q_n(a) = 1$ is a condition of completeness for a set of probable chains. The simplest model function satisfying the above-stated conditions reads (see Appendix)

$$Q_n(a) = \left[1 - \exp\left(-\frac{\gamma}{a}\right)\right]\exp\left(-\frac{\gamma(n-1)}{a}\right). \quad (3)$$

The numerical value of the positive parameter γ can be determined by comparing the calculated and experimental values of kinetic characteristics of excitation decay. Thus, for a given value of parameter a, the probability of excitation decay at the instant t is equal to

$$P(a,t) = \sum_{n=1}^{\infty} Q_n(a)P_n^n(t). \quad (4)$$

Using Eqs. (2)–(4), we obtain, for the average excitation lifetime in the PC,

$$\frac{\tau_a}{\tau_0} = W_0^2 \int_0^\infty tP(a,t)dt = 1 + \frac{1}{a(e^{\gamma/a} - 1)}. \quad (5)$$

It is easy to see that for the absence of jumps, $\lim_{a\to 0}\tau_a = \tau_0$ (the lifetime of excitation coincides with the lifetime of the excited electronic level of an impurity ion, $\tau_0 = W_0^{-1}$). The dependence of τ_a/τ_0 on the jump probability is given in Fig. 2. It is seen that the excitation jumps lead to prolongation of
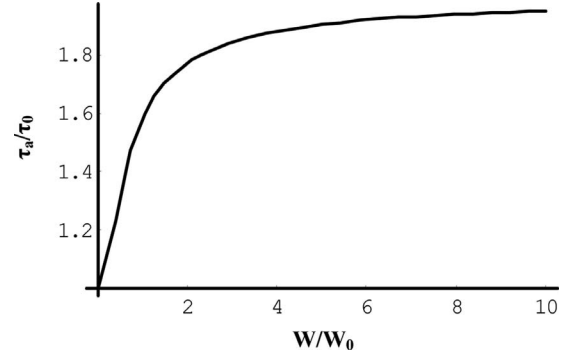

FIG. 2. Dependence of excitation lifetime in a PC on the probability of a jump at $\gamma=1$.

excitation lifetime, and at $W=W_0$ (for $\gamma=1$), when the process rate in PC equals the rate of intracentral relaxation, the excitation lifetime increases by ~ 1.6 times. In the limit of fast NTEEE $(a \gg 1)$, from Eq. (5) we have $\lim_{a\to\infty}\tau_a = (1+1/\gamma)\tau_0$.

The process of excitation decay in the PC consisting of two different ions (A and B) undergoing effective resonant energy exchange (i.e., electronic excitation can be transferred from one ion to another without energy losses) can be described in a similar way. It is presumed that at the initial instant $t=0$, ion A is excited and ion B is in the ground electronic state. Therefore, at the initial moment the PC is of (A*-B) type, the quantity of which we denote by n_0. It should be noted that in the general case, the rates of direct (from ion A* to ion B) and inverse (from ion B* to ion A) transitions are different. However, assuming the presence of resonant NTEEE channels, we can suppose that the rates of direct and inverse NTEEE are equal. The Markovian chains shown in Fig. 1 can also describe the evolution of excitation in a PC of (A-B) type. However, now the odd sites of the chain correspond to the excited state of ion A with an intracenter relaxation time $\tau_{01} = W_{01}^{-1}$, and the even sites correspond to the excited state of ion B with intracenter relaxation time $\tau_{02} = W_{02}^{-1}$ (for certainty, we suppose that $\tau_{01} > \tau_{02}$). Differential equations describing the excitation decay occurring in accordance with the chain of n length coincide with Eq. (1), where W_0 should be replaced by W_{01} for odd values of n and by W_{02} for even values. The solutions of the obtained system of differential equations with initial condition $P_k^{(n)}(0) = \delta_{k,1}$ $(1 < k \le n)$ coincide with Eq. (2) after replacement of parameter W_0, as stated above. For probability distribution of realizations of chains with n length, we take the following function:

$$\Omega_n(a) = \Omega_0^{-1}\exp\left(-\frac{\gamma(n-1)}{a}\right), \quad (6)$$

$$\Omega_0 = \frac{1}{2}\left[\frac{1}{\sinh(\gamma/a_2)} + \frac{\exp(\gamma/a_1)}{\sinh(\gamma/a_1)}\right],$$

$$a = \frac{a_1 + a_2}{2} + (-1)^n\frac{a_2 - a_1}{2}, \quad (7)$$

where $a_1 = W/W_{01}$ and $a_2 = W/W_{02}$. From Eq. (7), it follows

that $a=a_2$ for even n and $a=a_1$ for odd n. It is also seen that expression (6) represents a generalization of formula (3) because for $a_1=a_2=a$ (ions A and B are identical), it transforms to Eq. (3). Thus, the probability of excitation decay in a PC of type (A-B) at the instant t is

$$P(a,t) = \sum_{n=1}^{\infty} \Omega_n(a) P_n^n(t). \qquad (8)$$

The average lifetime of excitation in a PC of (A-B) type is obtained by expression (8),

$$\tau_a(W) = \frac{1}{2W\Omega_0}\left[\frac{1 + a_1 \exp(\gamma/a_1)\sinh(\gamma/a_1)}{\sinh^2(\gamma/a_1)}\right.$$
$$\left. + \frac{a_2 \sinh(\gamma/a_2) + \cosh(\gamma/a_2)}{\sinh^2(\gamma/a_2)}\right]. \qquad (9)$$

It is easy to show that at $a_1=a_2=a$, expression (9) converts into Eq. (5). As a rule, in real crystal systems, the case of rapid energy transfer is realized: the probability of NTEEE between ions in a PC is much higher as compared to intracentral relaxation ($W \gg W_{01}, W_{02}$). In this limiting case, supposing that the relaxation times $\tau_{01}>0$ and $\tau_{02}>0$ have finite values and $a_1 \to \infty$ and $a_2 \to \infty$, we obtain

$$\tau_a(\infty) = \left(1 + \frac{1}{\gamma}\right)\frac{(\tau_{01}^2 + \tau_{02}^2)}{\tau_{01} + \tau_{02}}. \qquad (10)$$

It is seen that the delay of excitation decay in a PC takes place for all finite values of τ_{02}/τ_{01} and γ. The minimum value of excitation lifetime $\tau_a^{(min)}(\infty) \cong 0.828(1+1/\gamma)\tau_{01}$ is reached at $\tau_{02}=(\sqrt{2}-1)\tau_{01}$.

III. EXCITATION DECAY IN IMPURITY SUBSYSTEM OF SIMILAR IONS

For description of excitation decay kinetics in the impurity system, let us divide it into two subsystems: subsystem of a PC and subsystem of a single impurity center. It is assumed that the concentration of impurity ions is small (~ 1 at. %), so that the rate W of NTEEE inside a PC is much higher than the rate of NTEEE W_1 between the ions which did not form a PC. Consequently, the NTEEE processes between PCs, as well as the energy transfer from PC to single centers, can be neglected. Thus, it is believed that the excitation confined in a PC does not leave it before relaxation. This allows considering the PC as an "acceptor" with excitation relaxation lifetime τ_a determined by formulas (5) and (9) for PCs of (A-A) and (A-B) types, respectively, and single impurity ions as "donors" with relaxation lifetime $\tau_d=\tau_0$.

We now consider the case when the impurity system consists of ions A, and the processes of excitation migration between single centers are neglected. The kinetic equation for the number of excited donors n_d and acceptors n_a is

$$\dot{n}_d = -\frac{1}{\tau_d}n_d - W_1 n_d, \quad \dot{n}_a = -\frac{1}{\tau_a}n_a + W_1 n_d, \qquad (11)$$

with initial conditions $n_d(0)=n_{20}$ and $n_a(0)=n_{10}$. Solving system (11) for the simplest case when W_1 does not depend
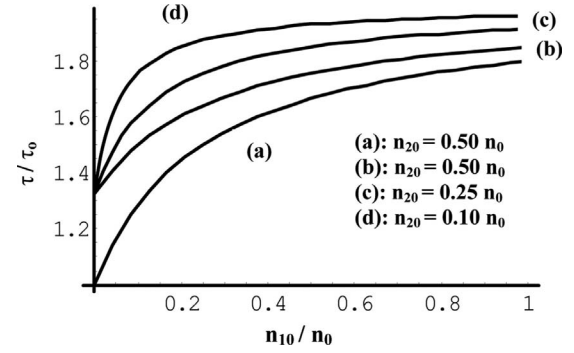

FIG. 3. Dependence of excitation lifetime on concentration of excited PCs at $T=300$ K and $\gamma=1$ in the case when (a) $W_1=0$, and [(b), (c), and (d)] $W_1=200$ s^{-1}.

on time, we obtain the excitation decay law, $n(t)=n_d(t)+n_a(t)$,

$$n(t) = n_0\left(\frac{\tau_a}{\tau_0} + W_1\tau_a - 1\right)^{-1}\left\{y\left[W_1\tau_a \exp\left(-\frac{t}{\tau_a}\right)\right.\right.$$
$$\left.+ \left(\frac{\tau_a}{\tau_0} - 1\right)\exp\left(-\frac{t}{\tau_a} - W_1 t\right)\right]$$
$$\left.+ x\left(\frac{\tau_a}{\tau_0} + W_1\tau_a - 1\right)\exp\left(-\frac{t}{\tau_a}\right)\right\}, \qquad (12)$$

and for excitation lifetime in the impurity subsystem,

$$\frac{\tau}{\tau_0} = \frac{\tau_a}{\tau_0} + \frac{1 - \tau_a/\tau_0}{1 + W_1\tau_0}\frac{y\tau_0}{x\tau_a(1 + W_1\tau_0) + y\tau_0(1 + W_1\tau_a)}, \qquad (13)$$

where $x=n_{10}/n_0$ and $y=n_{20}/n_0$ (here n_0 is the initial concentration of all excited impurity ions). In particular, in the limiting case of $\tau_a=(1+1/\gamma)\tau_0$, we get

$$\frac{\tau}{\tau_0} = 1 + \frac{1}{\gamma} - \frac{y}{(1 + W_1\tau_0)[x + \gamma(x + y) + (1 + \gamma)(x + y)W_1\tau_0]}. \qquad (14)$$

Note that since the parameters τ_a, τ_0, and W_1 depend on the temperature, formulas (13) and (14) determine the temperature dependence of excitation lifetime τ. Figure 3 shows the dependence of excitation lifetime on the concentration of the initially excited paired and single centers at $\gamma=1$ for the case where $W_1=0$ (energy migration from single centers to PCs is absent) and $W_1>0$. It is seen that the energy migration plays an essential role at small concentrations of the initially excited PC, and even at $n_{10}=0$ the excitation lifetime increases by more than 30%. It is seen also that the maximum value of excitation lifetime is reached at $n_0=n_{10}$, where all excited doping ions initially form a "regular" PC.

For obtaining the more detailed pattern for the case of small concentrations of impurity ions by solving Eq. (6), it is necessary to take into account the time dependence of migration rate W_1 (depletion effect), which is determined by the mechanism of NTEEE.[5,10] In the case of a dipole-dipole mechanism of NTEEE, $W_{DD} \sim R^{-6}$ (R is the distance between the interacting ions), we have[1]

$$W_1(t) = (\tau_0 t)^{-1/2} q, \tag{15}$$

where $q = \frac{2}{3}\pi^{3/2}R_c^3 C_A$ [C_A is the concentration of PCs and R_c is the critical distance of NTEEE in the "donor-acceptor" pair determined from the condition $W_{DD}(R_c)\tau_0 = 1$]. Solving Eq. (11) and taking into account Eq. (14), we obtain

$$n(t) = n_{10}\exp(-t/\tau_a) + n_{20}[\exp(-t/\tau_0 - 2q\sqrt{t/\tau_0}) + F(t)], \tag{16}$$

where

$$F(t) = \sqrt{\pi}q_1\exp\left(-q_1^2 - \frac{t}{\tau_a}\right)\{\Phi(q_1 + \sqrt{\alpha t}) - \Phi(q_1)\},$$

$q_1 = q(1 - \tau_0/\tau_a)^{-1/2}$, $\alpha = \sqrt{1/\tau_0 - 1/\tau_a}$, and $\Phi(z) = 2/\sqrt{\pi}\int_0^z \exp(-x^2)dx$ is the error integral.

Using Eq. (15), we obtain, for excitation lifetime in the impurity system,

$$\tau = \frac{n_{10}\tau_a^2 + n_{20}\tau_0^2 A}{n_{10}\tau_a + n_{20}\tau_0 B}, \tag{17}$$

where

$$A = \frac{1}{2}\{2(1 + q^2) - \sqrt{\pi}qe^{q^2}(3 + 2q^2)[1 - \Phi(q)]\} - \frac{\tau_a^2}{\tau_0^2}\Phi(q_1)$$

$$+ \frac{1}{\tau_0^2}\int_0^\infty t\exp(-t/\tau_a)\Phi(q_1 + \alpha\sqrt{t})dt, \tag{18}$$

$$B = 1 - \sqrt{\pi}qe^{q^2}[1 - \Phi(q)] - \frac{\tau_a}{\tau_0}\Phi(q_1)$$

$$+ \frac{1}{\tau_0}\int_0^\infty \exp(-t/\tau_a)\Phi(q_1 + \alpha\sqrt{t})dt. \tag{19}$$

In the case when only PCs are initially excited ($n_d = 0$, $n_a \neq 0$), we have $\tau = \tau_a$. If only single centers are excited ($n_d \neq 0$, $n_a = 0$), $\tau = (A/B)\tau_0$.

Analytical calculation of coefficients A and B yields complicated expressions. However, in each specific case, it is not difficult to perform numerical estimation of the values of A and B. Thus, for the LiNbO$_3$:Yb^{3+} system, the numerical evaluation performed by using the values of parameters at room temperature,[21] $\tau_0 = 728.2$ μs, $\tau_a = 2\tau_0$, $C_A = 0.2 \times 10^{20}$ cm^{-3} ($C_{Yb} = 1$ at. $\% = 1.6 \times 10^{20}$ cm^{-3}), $R_c = 15$ Å, $q = 0.2506$, $q_1 = 0.4092$, and $\alpha = 22.6929$, leads to magnitudes $A = 2.8132$ and $B = 1.6184$. Figure 4 shows the dependence of excitation lifetime in the impurity subsystem on the number of paired and single centers initially excited. Comparison of Figs. 3 and 4 shows that the dependence of excitation lifetime on number n_{10} of regular PCs initially excited has similar characteristics for both constant and time dependent migration rates. However, quantitatively, they differ. In particular, when $n_{10} = 0$, the time dependent migration rate leads to a larger increase in excitation lifetime ($\sim 75\%$).

It is obvious that the number n_{10} of regular PCs and the number of initially excited single centers n_{20} depend on the intensity and conditions of pumping. Assuming that the pumping is performed by short pulses with duration τ_p and
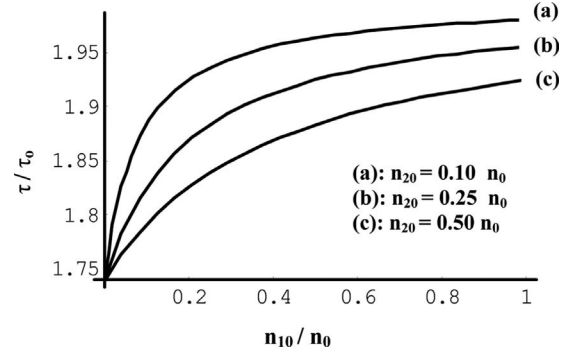

FIG. 4. Dependence of excitation lifetime on concentration of excited PCs at temporal variation of energy migration rate.

that the processes of radiation absorption by impurity ions are independent, the values of n_{10} and n_{20} can be approximated by the following expressions:

$$n_{10} = 2N_{10}\exp(-\sigma_p F\tau_p)[1 - \exp(-\sigma_p F\tau_p)], \tag{20}$$

$$n_{20} = N_0[1 - \exp(-\sigma_p F\tau_p)]\left[1 - \frac{2N_{10}}{N_0}\exp(-\sigma_p F\tau_p)\right], \tag{21}$$

where σ_p is the transverse cross section of absorption by impurity ions, F is the photon flux density in the incident wave pulse, and N_{10} and N_{20} are concentrations of impurity ions. In Eq. (21), we take into account also PCs, in which both ytterbium ions are excited. The relaxation characteristics of such "nonregular" PCs, the number of which evidently depends on the pump intensity, do not differ from relaxation characteristics of single centers. Expressions (20) and (21), together with Eqs. (16) and (17), specify analytical dependences of the decay law and excitation lifetime on the pump power $F\tau_p$ and concentration of PCs.

IV. CONCLUSIONS

The presence of PCs in a crystal matrix, even for small concentrations of impurity ions, essentially influences the kinetics of electronic excitation in the impurity subsystem. The correlation between kinetic characteristics and concentration of PCs can be established by analysis of excitation decay kinetics at different pump intensities. In particular, it is possible to determine the concentration of PCs by using the set of theoretical curves of electronic excitation decay (Figs. 3 and 4) and measured values of excitation lifetimes at given pump intensities.

Obviously, analytical expressions for the excitation decay law in the case when the impurity subsystem consists of two different ions A and B can also be derived in the same way. Here, of course, one needs to solve equations of Eq. (11) type written for a wider class of impurity formations: solitary centers consisting of A and B ions, as well as PCs of (A-A), (B-B), and (A-B) types.

It must be noted that as distinct from the well-known technique of rate equations[15–17] (which are initially written for the whole impurity system), the mathematical apparatus
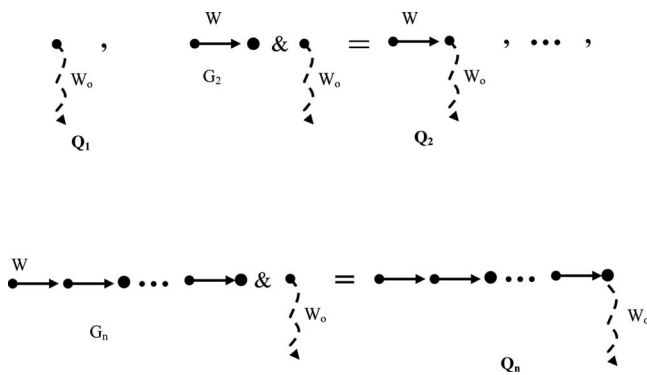
FIG. 5. Splitting of the Markovian chain of finite length.

of the Markovian processes (which is successfully used, along with density matrix method, for solution of problems of statistical physics) allows describing of the excitation decay process in a separate PC. Furthermore, by using the rate equation method in the frame of a chosen distribution model of impurity ions, one can determine kinetic characteristics of the whole impurity system.

We believe that the developed theoretical constructions, which are based on the theory of the Markovian processes, can be naturally generalized for more complicated formations including three and more impurity ions. However, the theoretical investigation of kinetics of electronic excitation decay in such formations requires the use of topological objects with more complicated structures. Thus, for description of excitation states in triple centers, two-dimensional gratings should be used instead of linear chains.

APPENDIX

The process of excitation decay following n-time jumps in the PC described by a chain of length n (Fig. 1) can be represented as an alternating implementation of processes of n-time excitation jumps (with probability of realization G_n) and subsequent excitation decay (with probability of realization Q_1) (Fig. 5).

Considering these two processes as independent from each other, one can write

$$Q_n = G_n Q_1. \tag{A1}$$

Because of the Markovian nature of excitation jump processes and independence of each jump, we have[19] G_{n+m}

$= G_n G_m$. As a result, an exponential dependence law follows for probability of appearing the chain of n length without excitation decay on the number of jumps, which can be presented as

$$G_n = \text{const} \cdot e^{-\beta(n-1)}, \tag{A2}$$

where β is a positive quantity. The minus sign in the exponent means decrease in G_n probability under unlimited increase in n. Substituting Eq. (A2) in Eq. (A1) and introducing an exponent normalization constant in Q_1, we get from the completeness condition $\Sigma_{n=1}^{\infty} Q_n = 1$,

$$Q_1 = (1 - e^{-\beta}). \tag{A3}$$

So, we get a Poisson-type distribution for the probability of realization of a chain of n length,

$$Q_n = (1 - e^{-\beta})e^{-\beta(n-1)}. \tag{A4}$$

The excitation lifetime can be found by formulas (2) and (4),

$$\frac{\tau_a}{\tau_0} = 1 + \frac{1}{a(e^{\beta} - 1)}. \tag{A5}$$

One should assume from physical reasoning that τ_a, as a function of the a parameter, is limited and monotonically increasing. From the finiteness requirement for τ_a, it follows that under unlimited increase in the a parameter, the $\beta(a)$ function decreases not faster than $1/a$ [one can prove it by developing the exponent in Eq. (A5) to power series on β]. Hence, one can set $\beta(a) = \gamma/a^s$, where $0 < s \le 1$ and $\gamma > 0$. However, one can see from Eq. (A5) that at $0 < s < 1$, the $\tau(a)$ function has a local maximum and therefore does not monotonically increase over the whole region of its definition. Consequently, $s = 1$, and

$$Q_n = (1 - e^{-\gamma/a})e^{-\gamma(n-1)/a}. \tag{A6}$$

It is seen that for $n > 1$, the function $Q_n(a)$ reaches its maxima $(Q_n)_{\max} = (n-1)^{n-1}n^{-n}$ at the points determined by $a_n = \gamma[\ln(n/n-1)]^{-1}$. It is important to note that the increase in n shifts the point of maximum to the right. It means that with the increase in probability of the elementary act of jump, the probability of realization of the Markovian chain of longer length increases.

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