

SOLID-STATE
SPECTROSCOPY

Description of Inhomogeneously Broadened Luminescence Spectra of Doped Media within the Framework of Coherent-Potential Approximation

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Abstract—Equations describing inhomogeneously broadened luminescence spectra of doped media are derived within the framework of coherent-potential approximation (CPA). The dependence of the shape of these spectra on the dopant concentration at a low temperature is studied. High precision of the CPA is shown by comparison with Monte Carlo simulations.

INTRODUCTION

The coherent-potential approximation (CPA) is widely used in the calculation of various features of disordered materials [1–3]. It was developed in [3–12] for the problems of the theory of luminescence. Only the positional disorder of impurity centers (donors and acceptors of excitation) was taken into account. The CPA was used for calculations of coefficients of diffusion of excitations within the system of identical centers (donors) for both exchange and dipole–dipole interaction, the quantum yield of luminescence of donors, anisotropy of luminescence, and the rate of concentration self-quenching [3, 12]. Comparison with Monte Carlo simulations of these quantities showed the good accuracy of CPA [3, 12].

The principle of the CPA is the following. A real disordered system of donor centers is replaced by a regular lattice (for example, cubic) with donors located in its sites. A lattice constant is determined by the concentration of donors. Interaction in such a lattice (effective medium) occurs only between the nearest neighbors. This interaction (the rate of excitation jump from one center to another and the coherent potential) is considered to be dependent on time and is chosen in such a way that the properties of an actual medium (described by the Green function G) coincide with those of an effective lattice (described by the Green function G^c):

$$G = G^c. \quad (1)$$

The CPA was not directly used for calculations of spectral properties of media with inhomogeneous broadening of impurity centers. In [13, 14], this approximation was used to study dispersive transport of charge carriers in disordered systems. The probability of a charge carrier jump from one localized state to another decreased exponentially with increasing jump distance. The results were compared with Monte Carlo simulations. It was found that the results obtained by these

methods were in good agreement at temperatures higher than 50 K. At lower temperatures, the theory [13, 14] yielded inadequate results. Therefore, the authors of [15, 16] used another approach for describing the transport of charge carriers at low temperatures. This approach virtually coincides with that developed in [17–20] and is the extension of the method of continuous time random walk (CTRW) to the case of spectral disorder of localized states.

Note that the problem of dispersive transport of charge carriers is equivalent to the problem of migration of triplet excitations over the system of molecules with different triplet energy levels [16]. The only difference is that, in the latter case, it is necessary to take into account the finite lifetime of triplet states.

In this work, the CPA is used in its conventional statement [3, 7]. Equations that describe a change of the luminescence spectrum as a function of the concentration of impurity centers are obtained. By comparing the results with Monte Carlo simulations, the high precision of the CPA is shown at low temperatures.

STARTING EQUATIONS

In the description of incoherent migration of excitations over an ensemble of randomly distributed impurity centers, the starting point is a system of balance equations

$$\frac{d}{dt}P(x_i, x_j, t) = -\frac{1}{\tau_0}P(x_i, x_j, t) + \sum_k^N [w(x_i, x_k)P(x_k, x_j, t) - w(x_k, x_i)P(x_i, x_j, t)], \quad (2)$$

$$P(x_i, x_j, 0) = \delta_{ij}.$$

Here, subscripts i and j numerate impurity centers; N is the number of centers; x_i is the coordinate of the center with number i , which denotes a combination of the Cartesian coordinate \mathbf{x}_i and the transition energy E_i between the ground and excited states here and in the following; $P(x_i, x_j, t)$ is the probability that the excitation is localized on the i th donor at the instant t , provided that initially the j th center was excited; τ_0 is the intrinsic lifetime of the excited state (which is assumed here and in the following to be independent of \mathbf{x}_i); $w(x_i, x_j)$ is the rate of excitation hopping from j th to i th center (in the general case, $w(x_i, x_j) \neq w(x_j, x_i)$); and δ_{ij} is the Kronecker symbol.

For multipole interaction of impurity centers, the jump rate can be written in the form

$$w(x_i, x_j) = \frac{1}{\tau_0} \left(\frac{R_0}{R} \right)^s J(E_i, E_j), \quad (3)$$

where $s = 6, 8,$ and 10 for dipole-dipole, dipole-quadrupole, and quadrupole-quadrupole interaction, respectively; $R = |\mathbf{x}_i - \mathbf{x}_j|$; and R_0 is the characteristic radius of energy transfer. The function $J(E_i, E_j)$ determines the dependence of hopping rate on transition energies in a pair of interacting centers. If these centers are molecules that have broad spectra, then $J(E_i, E_j)$ is expressed in terms of the overlap integral for luminescence and absorption spectra of molecules with transition energies E_i and E_j , respectively. If these centers are rare-earth ions whose spectra at low temperatures consist of a narrow zero-phonon line and a weak phonon wing then [21-28]

$$J(E_i, E_j) = \left| \frac{E_i - E_j}{\sigma} \right|^k, \quad E_i > E_j, \quad (4)$$

$$k = 0, 1, 2, 3, 5.$$

In most cases, dependence (4) follows from the overlap of the zero-phonon line of one center with the one-phonon wing of another center [28]. The exception is the modulation mechanism of energy transfer studied in [21-23].

An averaged solution of a system of equations (2) is of interest. It is expressed in terms of the Green function $G(x, x', t)$, which represents the probability density of excitation localized at the point $x = \{\mathbf{x}, E\}$ at the instant t , provided that the excitation was at the point $x' = \{\mathbf{x}', E'\}$ at $t = 0$. The Green function is defined as follows [3]:

$$G(x, x', t) = \frac{\left\langle \sum_{ij}^N \delta(x, x_i) P(x_i, x_j, t) \delta(x_j, x') \right\rangle}{\left\langle \sum_i^N \delta(x_i, x') \right\rangle}, \quad (5)$$

$$G(x, x', 0) = \delta(x, x').$$

Here, $\delta(x, x') = \delta(\mathbf{x} - \mathbf{x}')\delta(E - E')$ is the delta function. Angular brackets denote an averaging over spatial and energy distributions of impurity centers. This averaging consists in integration over coordinates of all centers with the distribution function $g(x_1, x_2, \dots, x_N)$

$$\langle A \rangle = \int dx_1 dx_2 \dots dx_N \quad (6)$$

$$\times g(x_1, x_2, \dots, x_N) A(x_1, x_2, \dots, x_N).$$

By assuming the spatial distribution of impurity centers to be homogeneous and uncorrelated with the distribution over transition energies, we have

$$g(x_1, x_2, \dots, x_N) = \prod_{i=1}^N \frac{1}{V} g(E_i), \quad (7)$$

where V is the system volume, and $g(E)$ is the normalized distribution function of centers over transition energies.

All quantities measured in experiments (the luminescence spectrum and decay quantum yield, luminescence anisotropy, diffusion coefficient, etc.) can be expressed in terms of the Green function [3]. For example, the distribution $I(E, t)$ of excited centers over transition energies at any given moment t has the form

$$I(E, t) = \int dx \int dx' \int dE' G(x, x', t) \Lambda(x'), \quad (8)$$

where $\Lambda(x')$ is the initial distribution of excited centers in space and over transition energies after pulsed excitation. In the case of spatially homogeneous excitation,

$$\Lambda(x) = \frac{1}{V} \Lambda_0 g^*(E), \quad (9)$$

where $g^*(E)$ satisfies the normalization condition

$$\int dE g^*(E) = 1.$$

Upon nonselective excitation,

$$g^*(E) = g(E), \quad (10)$$

i.e., the function $g^*(E)$ coincides with the distribution of centers over transition energies in the ground state.

The instantaneous luminescence spectrum $I_{\text{lum}}(\epsilon, t)$ is expressed in terms of the distribution function $I(E, t)$

$$I_{\text{lum}}(\epsilon, t) = \int dE I_0(\epsilon, E) I(E, t). \quad (11)$$

Here, $I_0(\epsilon, E)$ is the homogeneously broadened luminescence spectrum of molecules having transition energy E . The spectrum is assumed to be normalized

$$\int I_0(\epsilon, E) d\epsilon = 1. \quad (12)$$

If the homogeneous width of the spectrum is far smaller than the inhomogeneous width (rare-earth ions in crystals and glasses), then one can set in (11)

$$I_0(\epsilon, E) = \delta(\epsilon - E).$$

Then, from (11), follows

$$I_{\text{lum}}(E, t) = I(E, t). \quad (13)$$

In this case, the distribution function $I(E, t)$ corresponds to the instantaneous luminescence spectrum; herefore, we will further call this function the instantaneous luminescence spectrum.

EQUATIONS OF THE CPA

In deriving the Green function G^c for an effective medium, we proceed as follows. As in the case of the CPA, we replace the real disordered system of centers by a regular lattice. Because the impurity centers in the problem are described not only by spatial coordinates, but by transition energies as well, the lattice is four-dimensional. The fourth axis of coordinates displays the transition energy that can take only discrete values with the interval equal to ΔE . Some points on the fourth axis correspond to equal values of energy. The number of such points (a degree of degeneracy) $N(E)$ is determined by the distribution function $g(E)$ of centers over transition energies

$$N(E) = N_0 g(E) \Delta E.$$

Here, N_0 is the total number of points (permissible transition energies) on the energy axis.

In this lattice, only nearest neighbors (sites) interact with each other. It is assumed that this interaction w_c depends on time and transition energies of impurity centers. It is chosen such that condition (1) is valid.

Under these conditions, the Green function G^c of the effective medium must satisfy the equation

$$\begin{aligned} \frac{d}{dt} G_{ij}^c(E_i, E_j, t) &= -\frac{1}{\tau_0} G_{ij}^c(E_i, E_j, t) \\ &- z \int_0^t dt' \sum_{E_m} N(E_m) w_c(E_m, E_i, t-t') G_{ij}^c(E_i, E_j, t') \\ &+ \sum_{m=1}^z \sum_{E_m} N(E_m) \int_0^t dt' w_c(E_i, E_m, t-t') G_{mj}^c(E_m, E_j, t'), \end{aligned} \quad (14)$$

$$G_{ij}^c(E_i, E_j, 0) = \delta_{ij} \delta(E_i - E_j).$$

The summation in (14) over m is performed over sites of the three-dimensional lattice that are nearest to the center i , z being their number ($z = 6$ for the cubic lattice).

The instantaneous luminescence spectrum $I(E, t)$ is related to G^c by the equation

$$I(E, t) = N(E) \sum_{ij} \sum_{E_j} G_{ij}^c(E, E_j, t) g^*(E_j). \quad (15)$$

Going to the Laplace representation in equation (14)

$$\tilde{G}_{ij}^c(E_i, E_j, p) = \int_0^\infty dt e^{-pt} G_{ij}^c(E_i, E_j, t), \quad (16)$$

we obtain

$$\begin{aligned} p \tilde{G}_{ij}^c(E_i, E_j, p) - \delta_{ij} \delta(E_i - E_j) &= -\frac{1}{\tau_0} \tilde{G}_{ij}^c(E_i, E_j, p) \\ &- z \sum_{E_m} N(E_m) \tilde{w}_c(E_m, E_i, p) \tilde{G}_{ij}^c(E_i, E_j, p) \\ &+ \sum_{m=1}^z \sum_{E_m} \tilde{w}_c(E_i, E_m, p) N(E_m) \tilde{G}_{mj}^c(E_m, E_j, p). \end{aligned} \quad (17)$$

To satisfy condition (1), it is necessary to appropriately choose w_c . A simple way of finding w_c is reported in [2, 7] and discussed in detail in [3]. In an effective medium, the rate w_c of excitation hopping between a pair of nearest neighbors (for example, with numbers 1 and 2 and transition energies E_1 and E_2) is replaced by the true hopping rate $w(R, E_1, E_2)$. Then, the Green function G_v of such a perturbed medium is averaged over all possible distributions of the rate w . The function $\langle G_v \rangle$ must coincide with the Green function of G^c of the effective medium, because, in the considered approximation, w_c completely takes into account all fluctuations of the random quantity $w(R, E_1, E_2)$. This results in the equation [3]

$$\langle T \rangle = \left\langle \frac{U}{1 - \tilde{G}^c U} \right\rangle = 0. \quad (18)$$

Here, the angular brackets denote averaging over distributions of w , and U is the matrix that has the following nonzero elements:

$$\begin{aligned} U_{11} &= -U_{21} = \tilde{w}_c(E_2, E_1, p) - w(R, E_2, E_1), \\ U_{22} &= -U_{12} = \tilde{w}_c(E_1, E_2, p) - w(R, E_1, E_2). \end{aligned} \quad (19)$$

Equation (18) is equivalent to the system of equations for the elements of the matrix T . Under the studied conditions (19), this system reduces to a single equation

$$\begin{aligned} \langle U_{11} / \{ 1 - U_{11} [\tilde{G}_{11}^c(E_1, p) - \tilde{G}_{12}^c(E_1, E_2, p)] \\ - U_{22} [\tilde{G}_{22}^c(E_2, p) - \tilde{G}_{21}^c(E_2, E_1, p)] \} \rangle = 0. \end{aligned} \quad (20)$$

Equations (14), (15), (19), and (20) completely describe luminescence spectra of disordered media in the CPA.

Note that, in the absence of inhomogeneous broadening, all sites of the effective lattice become identical, and, therefore,

$$U_{11} = U_{22}, \quad \tilde{G}_{11}^c = \tilde{G}_{22}^c, \quad \tilde{G}_{12}^c = \tilde{G}_{21}^c. \quad (21)$$

Equation (20) therewith reduces to the well-known equation in the CPA [3, 7, 8], which is used for calculation of the effective rate w_c of excitation migration.

EFFECTIVE RATE OF MIGRATION AT LOW TEMPERATURE

As was mentioned in the Introduction, the most interesting for us is the case of low temperatures. In this limit, equation (20) can be substantially simplified and analytically solved.

Indeed, at low temperatures, excitations jump from centers with higher transition energy to those with lower energy. The inverse process is unlikely. The migration of excitations becomes unidirectional. Therefore, if $E_1 > E_2$, then

$$\tilde{G}_{12}^c = 0, \quad U_{22} = 0,$$

and equation (20) takes the form

$$\left\langle \frac{\tilde{w}_c(E_2, E_1, p) - w(R, E_2, E_1)}{1 - \tilde{G}_{11}^c(E_1, p)[\tilde{w}_c(E_2, E_1, p) - w(R, E_2, E_1)]} \right\rangle = 0. \quad (22)$$

Averaging in (22) is accomplished by means of the nearest-neighbor distribution function, this neighbor having one of the allowed transition energies in the effective medium [3, 6, 7]

$$\rho(R)dR = \frac{4\pi}{z_0} R^2 n \exp\left(-\frac{4\pi}{3z_0} R^3 n\right) dR. \quad (23)$$

Here, n is the concentration of impurity centers, and $z_0 = zN_0$ (The total number of sites in the effective four-dimensional medium is equal to N_0N , where N is the number of sites in the spatial lattice.)

Subsequent calculations are possible after specification of the dependence of the hopping rate w on the distance between impurity centers. We chose it in the form of (3) with $s = 6$ (for dipole-dipole interaction). Then, equation (22) is reduced to a dimensionless form

$$1 - \frac{y^2}{\tilde{G}_{11}^c(E_1, p)\tilde{w}_c(E_2, E_1, p)} \int_0^\infty dx \frac{\exp(-x)}{x^2 + y^2} = 0, \quad (24)$$

$$y^2 = b \frac{\tilde{G}_{11}^c \tilde{w}_c}{1 - \tilde{G}_{11}^c \tilde{w}_c}, \quad b = \frac{d^2}{z_0^2 \tau_0 \tilde{w}_c}, \quad (25)$$

$$d = \frac{4\pi}{3} R_0^3 n J^{1/2}(E_2, E_1).$$

Provided that

$$\tilde{w}_c \tilde{G}_{11}^c \ll 1, \quad y \ll 1, \quad (26)$$

equation (24) has the solution

$$\tilde{G}_{11}^c \tilde{w}_c = \frac{\pi}{2} y, \quad (27)$$

from which, according to (25), follows

$$\tilde{w}_c(E_2, E_1, p) = \frac{\pi}{2} z_0^{-1} d(E_2, E_1) [\tau_0 \tilde{G}_{11}^c(E_1, p)]^{-1/2} \quad (28)$$

Note that the function $G_{11}^c(E_1, t)$ (the diagonal part of the Green function of the effective medium) means the probability of localization of excitation on site 1 with the transition energy E_1 at the instant t if the same site was initially excited. This function depends only on the transition energy and time.

In deriving the equation, we assumed that the migration was unidirectional. For this case, the analytic form of the function $G_{11}^c(E, t)$ is known [17–20]

$$G_{11}^c(E, t) = \exp[-t/\tau_0 - \gamma(E)\sqrt{t/\tau_0}],$$

$$\gamma(E) = \gamma_0 \int_{-\infty}^E dE' g(E') J^{1/2}(E', E), \quad (29)$$

$$\gamma_0 = \sqrt{\pi} c, \quad c = 4\pi R_0^3 n/3.$$

In the following, only the values \tilde{w}_c and \tilde{G}_{11}^c at $p = 0$ will be required. Then, for low concentrations ($c \ll 1$), from (29) and (28), we obtain

$$\tilde{G}_{11}^c(E, 0) = \int_0^\infty dt G_{11}^c(E, t) \approx \tau_0, \quad (30)$$

$$\tilde{w}_c(E_2, E_1, 0) = \frac{\pi}{2z_0\tau_0} c J^{1/2}(E_2, E_1).$$

This result is exact [17–19, 29].

For high concentrations ($c \gg 1$),

$$\tilde{G}_{11}^c(E, 0) \approx \frac{2\tau_0}{\gamma^2(E)}, \quad (31)$$

$$\tilde{w}_c(E_2, E_1, 0) \approx \frac{\pi}{2\sqrt{2}z_0\tau_0} c J^{1/2}(E_2, E_1) \gamma(E_1).$$

The analogous dependence of the effective hopping rate $\tilde{w}_c(E_2, E_1, 0)$ on the transition energy (within a constant factor) can be obtained by means of the methods of CTRW [17–19] and GAF [29] developed in [30].

THE KINETICS OF LUMINESCENCE AT LOW TEMPERATURE

The observable quantity is the luminescence spectrum. By means of the relation between the Green function and the kinetics of the luminescence decay (15)

and replacing the summation over transition energies in (14) by integration

$$\sum_{E_i} N(E_i) = \sum_{E_i} N_0 g(E_i) \Delta E = N_0 \int g(E) dE,$$

we obtain

$$\begin{aligned} \frac{d}{dt} I(E, t) = & -\frac{1}{\tau_0} I(E, t) - z_0 \int_0^t dt' \int_{-\infty}^E dE' g(E') \\ & \times w_c(E, E, t-t') I(E, t') \\ & + z_0 g(E) \int_0^t dt' \int_0^E dE' w_c(E, E', t-t') I(E, t'). \end{aligned} \quad (32)$$

The limits of integration over energy take into account that, at low temperatures, the energy transfer occurs only from the high-energy centers to the low-energy centers.

Let us introduce the effective rate $W(E, E', t)$ of energy migration in the form

$$W(E, E', t) = z_0 w_c(E, E', t). \quad (33)$$

Then, equation (32) can be rewritten as

$$\begin{aligned} \frac{d}{dt} I(E, t) = & -\frac{1}{\tau_0} I(E, t) - \int_0^t dt' \int_{-\infty}^E dE' g(E') \\ & \times W(E, E, t-t') I(E, t') \\ & + g(E) \int_0^t dt' \int_0^E dE' W(E, E', t-t') I(E, t') \end{aligned} \quad (34)$$

and takes the form of a simple balance equation.

This equation can also be written in another form in the Laplace presentation, taking into account initial condition (9)

$$\begin{aligned} p \bar{I}(E, p) - \Lambda_0 g^*(E) = & -\frac{1}{\tau_0} \bar{I}(E, p) \\ & - \int_{-\infty}^E dE' g(E') \bar{W}(E, E, p) \bar{I}(E, p) \\ & + g(E) \int_E^{\infty} dE' \bar{W}(E, E', p) \bar{I}(E, p). \end{aligned} \quad (35)$$

However, for the diagonal part of the Green function, from equation (17), we obtain

$$\begin{aligned} p \bar{G}_{11}^c(E, p) - 1 = & -\frac{1}{\tau_0} \bar{G}_{11}^c(E, p) \\ & - \int_{-\infty}^E dE' g(E') \bar{W}(E, E, p) \bar{G}_{11}^c(E, p). \end{aligned} \quad (36)$$

It follows from (36) that

$$\int_{-\infty}^E dE' g(E') \bar{W}(E, E, p) = \frac{1}{\bar{G}_{11}^c(E, p)} - p - \frac{1}{\tau_0}. \quad (37)$$

By substituting (37) into (35), we find

$$\begin{aligned} \bar{I}(E, p) = & \Lambda_0 g^*(E) \bar{G}_{11}^c(E, p) \\ & + \bar{G}_{11}^c(E, p) g(E) \int_E^{\infty} dE' \bar{W}(E, E', p) \bar{I}(E, p). \end{aligned} \quad (38)$$

In the time representation,

$$\begin{aligned} I(E, t) = & \Lambda_0 g^*(E) G_{11}^c(E, t) + g(E) \int_0^t dt' G_{11}^c(E, t-t') \\ & \times \int_0^{t'} dt'' \int_0^E dE' W(E, E', t'-t'') I(E, t''). \end{aligned} \quad (39)$$

Thus, we obtained two equivalent equations, (34) and (39), for calculation of the instantaneous luminescence spectrum (the kinetics of luminescence). The dependence of the effective rate of migration W on E and E' with consideration of (33) is described by equations (30) and (31) in the regions of low and high concentrations of impurity centers, respectively. To find W in the general case, one needs to solve equation (24), taking into account explicit form (29) of the function $G_{11}^c(E, t)$.

THE STEADY-STATE LUMINESCENCE SPECTRUM AT LOW TEMPERATURE

Now we consider the simpler case of steady-state excitation of impurity centers.

The distribution function $I(E, t)$ of excited centers over transition energies at the instant t upon pulsed excitation is related to the distribution function $I^{st}(E)$ of excited centers upon steady-state excitation as follows:

$$I^{st}(E) = \int_0^{\infty} dt I(E, t) = \bar{I}(E, p) \Big|_{p=0}. \quad (40)$$

The steady-state luminescence spectrum $I_{lum}^{st}(E)$ is expressed in terms of the distribution function $I^{st}(E)$

$$I_{lum}^{st}(E) = \int I_0(\epsilon, E) I^{st}(E) dE. \quad (41)$$

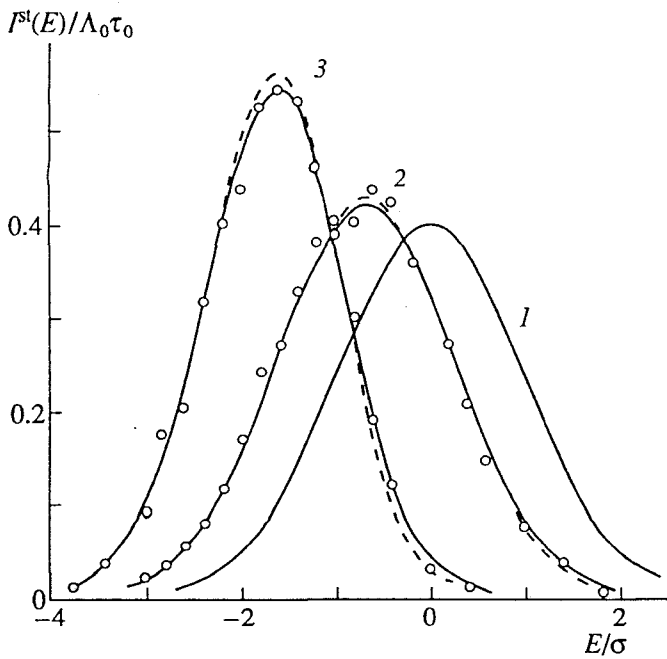


Fig. 1. Steady-state spectra upon nonselective excitation. Solid lines are calculations in the CPA, and circles are Monte Carlo simulations $k = 3$; $s = 6$; and $c = (1) 0$, (2) 0.56, and (3) 5.6.

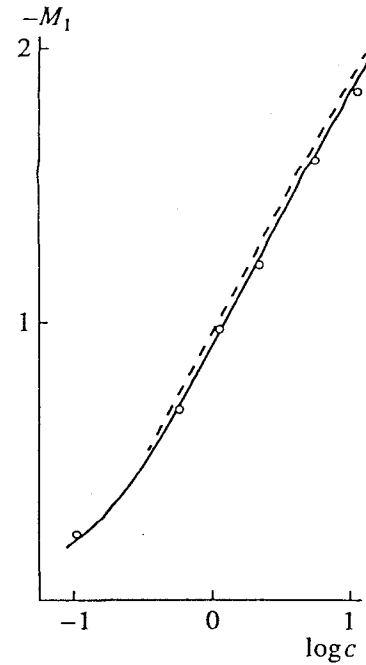


Fig. 2. Concentration dependence of the first moment ($-M_1$) of the distribution function $I^{st}(E)$ upon nonselective excitation. The notations are as in Fig. 1.

It follows from (40) that the equation for $I^{st}(E)$ can be obtained from (35) and (38) if the Laplace parameter ρ in them is set to zero. Then,

$$I^{st}(E) = \tau_0 \Lambda_0 g^*(E) - \int_{-\infty}^E dE' g(E') \tau_0 \tilde{W}(E, E', 0) I^{st}(E') + g(E) \int_E^{\infty} dE' \tau_0 \tilde{W}(E, E', 0) I^{st}(E'), \quad (42)$$

$$I^{st}(E) = \Lambda_0 g^*(E) \tilde{G}_{11}^c(E, 0) + g(E) \tilde{G}_{11}^c(E, 0) \int_E^{\infty} dE' \tilde{W}(E, E', 0) I^{st}(E'). \quad (43)$$

We shall use equation (43) in specific calculations.

The effective rate of migration $\tilde{W}(E, E', 0)$ will be calculated as follows. According to (30), (31), and (33), at low temperatures and for dipole-dipole interaction of impurity centers,

$$\tilde{W}(E, E, 0) = \frac{\pi c}{2\tau_0} J^{1/2}(E, E). \quad (44)$$

At high concentrations,

$$\tilde{W}(E, E, 0) = \frac{\pi c}{2\sqrt{2}\tau_0} J^{1/2}(E, E) \gamma(E). \quad (45)$$

Note that these two expressions differ only by a factor that depends on the transition energy E . Therefore, instead of solving equation (24) for \tilde{w}_c (which appears to be a rather time-consuming procedure), we approximate \tilde{W} in the entire range of concentration [taking into account (44) and (45)] as follows:

$$\tilde{W}(E, E, 0) = \frac{\pi c}{2\tau_0} J^{1/2}(E, E) A(E, c). \quad (46)$$

Here, the function $A(E, c)$ depends on E and c and must be defined. It is known that

$$A(E, c) = 1, \quad c \ll 1, \quad (47)$$

$$A(E, c) = \frac{1}{\sqrt{2}} \gamma(E), \quad c \gg 1.$$

To calculate $A(E, c)$, we shall use equation (37) to obtain

$$A(E, c) = \frac{\tau_0 \tilde{G}_{11}^c(E, 0) - 1}{\frac{\pi c}{2} \int_{-\infty}^E dE' g(E') J^{1/2}(E', E)}. \quad (48)$$

In the case under study (low temperature and dipole-dipole interaction of impurity centers), the function $G_{11}^c(E, t)$ is well-known and described by (29).

Thus, the distribution $I^{st}(E)$ is calculated in the CPA in the following way. At first, the factor $A(E, c)$ is found from (48) by means of explicit form (29) for the func-

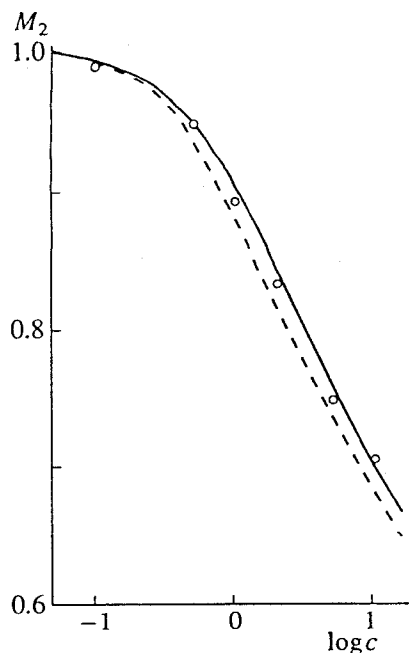


Fig. 3. Concentration dependence of the variance of the distribution function $I^{\text{st}}(E)$ upon nonselective excitation. The notations are as in Fig. 1.

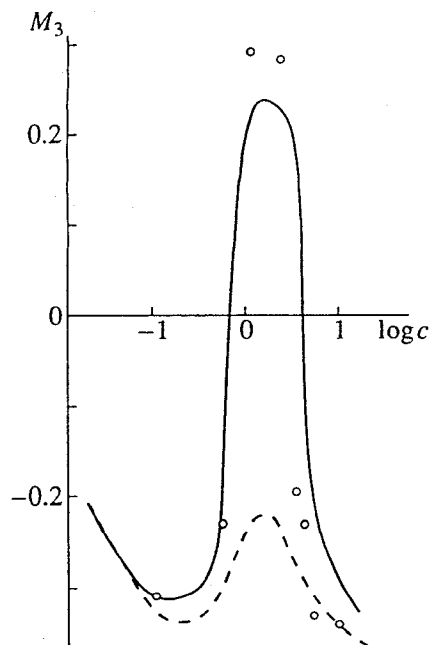


Fig. 4. Concentration dependence of the asymmetry coefficient of the distribution function $I^{\text{st}}(E)$ upon nonselective excitation. The notations are as in Fig. 1.

tion $G_{11}^c(E, t)$. Then, the effective rate of migration $\bar{W}(E, E, 0)$ (46) is substituted into equation (43), and the required distribution $I^{\text{st}}(E)$ is calculated by iteration procedure for a given concentration c of impurity centers and specified pump conditions $g^*(E)$.

NUMERICAL RESULTS AND CONCLUSIONS

The above procedure of calculations was carried out for dipole-dipole interaction of impurity centers (3), where $J(E_i, E_j)$ was taken in the form (4).

The distribution $g(E)$ of centers over transition energies was described by a Gaussian

$$g(E) = \frac{1}{\sqrt{2\pi}\sigma} \exp\left[-\frac{(E - E_g)^2}{2\sigma^2}\right], \quad (49)$$

where σ is the inhomogeneous width, and E_g is the center of the distribution.

We calculated not only the shape of the function $I^{\text{st}}(E)$, but also its first three moments:

the center of gravity

$$M_1 = (\langle E \rangle - E_g)/\sigma, \quad \langle E \rangle = \int E I^{\text{st}}(E) dE; \quad (50)$$

the variance

$$M_2^2 = \langle (E - \langle E \rangle)^2 \rangle / \sigma^2, \quad (51)$$

$$\langle (E - \langle E \rangle)^2 \rangle = \int (E - \langle E \rangle)^2 I^{\text{st}}(E) dE;$$

and the asymmetry

$$M_3^3 = \langle (E - \langle E \rangle)^3 \rangle / \sigma^3, \quad (52)$$

$$\langle (E - \langle E \rangle)^3 \rangle = \int (E - \langle E \rangle)^3 I^{\text{st}}(E) dE.$$

Figures 1–4 present the results of these calculations

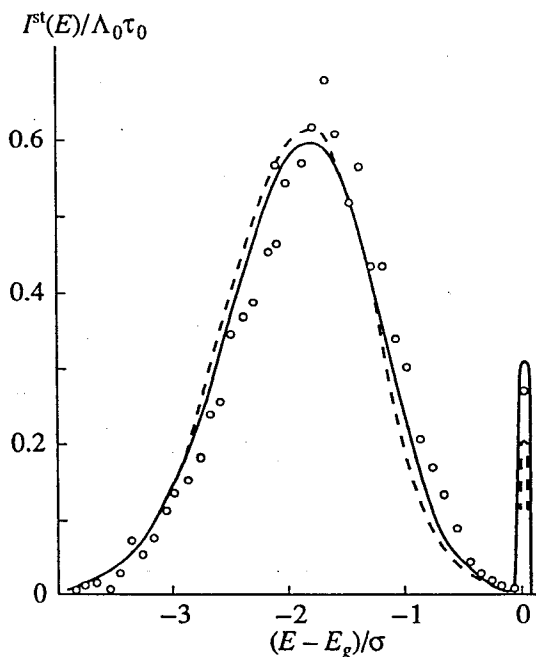


Fig. 5. Steady-state spectra upon selective excitation. Solid lines are the calculations in the CPA, and circles are Monte Carlo simulations $k = 3$, $s = 6$, and $c = 10$.

(solid lines) for $k = 3$ in equation (4) upon nonselective excitation when

$$g^*(E) = g(E). \quad (53)$$

For comparison, these figures also present the results of Monte Carlo simulation (circles) and the self-consistent diagrammatic GAF method (29) (dashed lines).

In Monte Carlo simulation, an algorithm similar to that described in [31, 32] was used. The periodic boundary conditions were used (the medium was treated as a crystal with 200 impurity centers per unit cell). Averaging was carried out over 5000 different distributions of impurity centers.

Note that the third moment M_3 was calculated by Monte Carlo method with the least accuracy, because it is determined by the wings of the distribution function $g(E)$, where the number of centers is small.

Figure 5 presents the results of calculations of the distribution $I^{\text{st}}(E)$ upon selective excitation of centers (at the center of the inhomogeneous band):

$$g^*(E) = \delta(E - E_g). \quad (54)$$

The notations are the same.

Note that the results of calculations performed by CTRW technique [17, 19] coincide within the accuracy of presentation in the figures with those obtained in the CPA.

Comparison of calculations performed by the CPA and Monte Carlo methods shows that equations obtained in this work describe very well (in contrast to equations of [13, 14]) the incoherent migration of excitations in-disordered media at low temperature.

Under the same conditions, the GAF method in the two-particle self-consistent approximation less well describes the luminescence spectra (especially the third moment).

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