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Spontaneous and Stimulated Transitions in Impurity Dielectric Nanoparticles

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1. Introduction

In recent years, great interest has been expressed by researchers in the optical properties exhibited by nanomaterials, including theoretical and experimental studies of the spontaneous lifetime of optical centers in nanosized samples (Christensen et al., 1982; Meltzer et al., 1999; Zakharchenya et al., 2003; Manoj Kumar et al., 2003; Vetrone et al., 2004; Chang-Kui Duan et al., 2005; Dolgaleva et al., 2007; Guokui Liu & Xueyuan Chen, 2007; Song & Tanner, 2008). A change in the spontaneous lifetime of optical centers (OCs) in nanoobjects as compared to bulk materials is of considerable interest for both fundamental physics and practical applications in the field of laser materials and phosphors. For example, the increased lifetime of a metastable level in a lasing medium makes it possible, by increasing the pump-pulse duration several times, to reduce the power and cost of the diode laser-pump source and superluminescence losses while keeping the output-radiation energy and power intact. The adequate theoretical interpretation of the experimental results is of primary importance at the current stage of investigations. It is of great interest to derive a formula describing the spontaneous decay rate of an excitation in a nanosized object and reveal its differences from the corresponding expression for the bulk sample.

The existence of spontaneous emission postulated in 1917 by Einstein in his quantum theory of the interaction between the equilibrium radiation and matter (Einstein, 1917). It is shown in this paper that the statistical equilibrium between matter and radiation can only be achieved if spontaneous emission exists together with the stimulated emission and absorption. The quantum-mechanical expression for the Einstein coefficient of spontaneous emission A equal to the probability of spontaneous emission from a two-level atom in a vacuum has been obtained by (Dirac, 1927; Dirac, 1982). In 1946, (Purcell, 1946) it is shown that the spontaneous emission probability can drastically increase if the radiating dipole is placed in a cavity (see also (Oraevskii, 1994; Milonni, 2007) and references therein). The inverse phenomenon, i.e., the inhibition of the spontaneous emission, can take place in three-dimensional periodic dielectric structures (Yablonovitch, 1987). Variations in the probability of the spontaneous emission from optical centers near the planar interface of the dielectrics have been the subject of active studies since the 1970s (Drexhage, 1970; Kuhn, 1970; Carniglia & Mandel, 1971; Tews, 1973; Morawitz & Philott, 1974; Agarwal, 1975; Lukosz & Kunz, 1977; Chance et al., 1978; Khosravi & Loudon, 1991; Barnes, 1998).

Modifications of the spontaneous emission from OCs located in the vicinity of a metal mirror also has been analyzed (Amos & Barnes, 1997; Brueck et al., 2003). Chew (Chew,

Source: Recent Optical and Photonic Technologies, Book edited by: Ki Young Kim,
 ISBN 978-953-7619-71-8, pp. 450, January 2010, INTECH, Croatia, downloaded from SCIYO.COM

1987; Chew, 1988) considered the modification of the spontaneous emission from an optical center inside and outside the dielectric sphere by modeling the optical center with an oscillating dipole. His analytical results were confirmed later by Fam Le Kien et al. (Fam Le Kien et al., 2000) devoted to the spontaneous emission from a two-level atom inside a dielectric sphere and by Glauber and Lewenstein (Glauber & Lewenstein, 1991). Klimov et al., 2001, considered the problem of the spontaneous emission from an atom near a prolate spheroid. The problem of the spontaneous emission from an atom in the vicinity of a triaxial nanosized ellipsoid is analyzed in a recent paper (Guzatov & Klimov, 2005). Of course, these short overviews far from being comprehensive.

It is well known now that spontaneous emission rate is not necessarily a fixed and an immutable property of optical centers but can be controlled.

The Chapter's central theme is the radiative characteristics of the small-radius optical centers (dopant ions of transition elements) in the subwavelength nanocrystals embedded in a dielectric medium. Our main aim is to provide answer the question "How the expressions derived for the radiative characteristics of optical centers in a bulk material should be modified upon changing over to a nanoobject?".

The rest of the Chapter is organized as follows. In Section 2 expression for the spontaneous radiative decay rate of OCs in a bulk crystal is presented and problem of the local field correction factor is briefly discussed. The expressions for the spontaneous radiative decay rate of OCs in the spherical nanocrystals are presented in Section 3. In Section 4 the expressions for the integrated emission and absorption cross - sections for spherical nanoparticles are given. The expressions for the spontaneous radiative decay rate of OCs in the ellipsoidal nanocrystals are presented in Section 5. Section 6 discusses the applicability of the Judd-Ofelt equation for nanoparticles. In Section 7 the experimental confirmation of the model for spontaneous radiative decay rates of rare-earth ions in the crystalline spherical nanoparticles of cubic structure embedded into different inert dielectric media is presented. The Chapter concludes in Section 8 showing directions for future research and conclusions.

2. Spontaneous radiative rate in a bulk crystal

The coupling between atom and the electric field in the dipole approximation is given by the electric-dipole interaction Hamiltonian

$$H_{\text{int}} = -\hat{\mathbf{d}} \hat{\mathbf{E}} \quad (2.1)$$

where $\hat{\mathbf{d}}$ is operator of the dipole moment and $\hat{\mathbf{E}}$ is the electric field operator, evaluated at the dipole position. In vacuum

$$\hat{\mathbf{E}}^{(vac)} = i \sum_{\mathbf{k}, \sigma} \sqrt{\frac{2\pi\hbar\omega_{\mathbf{k}}}{V}} \mathbf{e}_{\mathbf{k}, \sigma} \left[a_{\mathbf{k}, \sigma} - a_{\mathbf{k}, \sigma}^+ \right] \quad (2.2)$$

where $\mathbf{k}$ is wave vector; σ denotes the state of polarization; $a_{\mathbf{k}, \sigma}$ and $a_{\mathbf{k}, \sigma}^+$ are the photon destruction and creation operators for field eigenmodes, which specified by indices $(\mathbf{k}, \sigma)$; $\mathbf{e}_{\mathbf{k}, \sigma}$ is the polarization vector; V is the quantization volume. Photon frequency $\omega_{\mathbf{k}}$ is connected with wave number $k = |\mathbf{k}|$ by the linear dispersion relation $\omega_{\mathbf{k}} = c_0 k$ where c_0 is

light velocity in vacuum. Fermi's golden rule leads to the following expression for the electric-dipole spontaneous emission rate in free space (Dirac, 1982):

$$A_0 = \left(4\pi^2 \omega |\mathbf{d}|^2 / 3\hbar \right) \rho_{vac}(\omega). \quad (2.3)$$

Here ω is frequency of transition from excited atomic state i to lower-energy state j ;

$$\rho_{vac}(\omega) = \omega^2 / \pi^2 c_0^3 \quad (2.4)$$

is the photon density of states in vacuum; $|\mathbf{d}|^2 = d_x^2 + d_y^2 + d_z^2$, where $d_\alpha = \langle i | \hat{d}_\alpha | j \rangle$ ($\alpha = x, y, z$)

are the electronic matrix elements of the electric-dipole operator $\hat{\mathbf{d}}$ between the states i and j . The quantization of the electromagnetic field in a dielectric medium was first carried out by Ginzburg (Ginzburg, 1940). The macroscopic electric-field operator in a linear, isotropic, and homogeneous medium is given by

$$\hat{\mathbf{E}} = i \sum_{\mathbf{k}, \sigma} \sqrt{\frac{2\pi\hbar\omega_{\mathbf{k}}}{V\varepsilon}} \mathbf{e}_{\mathbf{k}, \sigma} \left[a_{\mathbf{k}, \sigma} - a_{\mathbf{k}, \sigma}^\dagger \right] \quad (2.5)$$

with dielectric function $\varepsilon(\omega_{\mathbf{k}})$ and the dispersion relation $\omega_{\mathbf{k}} = c_0 k / n$, where n is the refractive index of a dielectric. This dispersion relation results in changes in the photon density of states of a dielectric:

$$\rho(\omega) = n^3 \rho_{vac}(\omega) \quad (2.6)$$

Considering Eqs. (2.5) - (2.6) one can neglect the local-field effect for a moment and obtain for the electric-dipole spontaneous emission rate in the continual approximation (Nienhuis & Alkemade, 1976):

$$A = n A_0 \quad (2.7)$$

However, in general case the electric-dipole interaction Hamiltonian has the form

$$H_{\text{int}} = -\hat{\mathbf{d}} \hat{\mathbf{E}}^{(loc)}, \quad (2.8)$$

where $\hat{\mathbf{E}}^{(loc)}$ is the local electric field operator acting at the position of the optical center. The local electric field in a crystal differs from the macroscopic electric field in a crystal. For this reason the expression for the electric-dipole spontaneous radiative rate of the small-radius optical centers in a bulk crystal is given by (Lax, 1952; Fowler & Dexter, 1962; Imbush & Kopelman, 1981)

$$A_{\text{bulk}} = n_{cr} (E^{(loc)} / E^{(cr)})^2 A_0 = n_{cr} f_L A_0. \quad (2.9)$$

Here n_{cr} is the refractive index of a crystal; $E^{(loc)}$ and $E^{(cr)}$ are the strengths of the microscopic and macroscopic electric fields acting at the position of the optical center, respectively. Ratio

$f_L = (E^{(loc)} / E^{(cr)})^2$ is so called the local-field correction factor. In all existing local-field models f_L is a function of the crystal refractive index n_{cr} (see the comprehensive review of all currently available local-field models in paper (S. F. Wuister et al., 2004)); i.e., $(E^{(loc)} / E)^2 = f_L(n_{cr})$ and $f_L(1) = 1$. Most commonly used the local-field models are models of real cavity and virtual cavity (Rikken & Kessener, 1995). In case of an empty, real spherical cavity,

$$f_L(n) = \left[\frac{3n^2}{2n^2 + 1} \right]^2. \quad (2.10)$$

In case of a virtual cavity (Lorentz model)

$$f_L(n) = \left[\frac{n^2 + 2}{3} \right]^2. \quad (2.11)$$

The next sections will answer the question: How expression (2.9) for the electric-dipole spontaneous radiative rate of the small-radius optical centers in a bulk crystal should be modified upon changing over to a nanoobject?

3. Spontaneous radiative rate in the spherical nanocrystal

We shall refer to nanocomposite for dielectric nanocrystals embedded into different homogeneous dielectric media with refraction index n_{med} . The nanocrystals are assumed to be small enough compared with wavelength λ , but large compared with lattice constant a_L , so that the nanocrystals can be characterized by refraction index, which coincide with that of bulk crystal n_{cr} . The light wave propagates through the nanocomposite with amplitude E and a velocity c_0 / n_{eff} . The electric field E is macroscopic field averaged over volumes large enough as compared to the scales of inhomogeneities:

$$E = (1-x)E^{(med)} + x E^{(cr)} \quad (3.1)$$

where x is the volume fraction of nanocrystals in the medium (filling factor), $E^{(med)}$ and $E^{(cr)}$ are macroscopic fields in the dielectric medium and nanocrystals, respectively (Bohren & Huffman, 1998). So, expression (2.9) should be replaced by

$$A_{nano} = n_{eff} (E^{(loc)} / E)^2 A_0. \quad (3.2)$$

After some obvious transformations of this expression we obtain (Pukhov et al., 2008; Basiev et al., 2008)

$$A_{nano} = n_{eff} (E^{(cr)} / E)^2 (E^{(loc)} / E^{(cr)})^2 A_0, \quad (3.3)$$

or,

$$A_{nano} = n_{eff} f_N f_L(n_{cr}) A_0, \quad (3.4)$$

where

$$f_N = (E^{(cr)} / E)^2 \quad (3.5)$$

is the correction factor that accounts for the difference between the macroscopic electric field $E^{(cr)}$ at the position of the optical center and the macroscopic electric field E in the nanocomposite. We assume here that the local-field correction factor is the same as in the bulk crystal because of macroscopic size of nanocrystals. This assumes that the microscopic surrounding of the optical centers is the same in a nanocrystal and in a bulk crystal. Of course, it is not valid for the optical centers located near the nanocrystal surface at distances smaller than perhaps ten lattice constant (Kittel, 2007). The arguments in support of the inference that the correction f_N differs from unity were clearly and thoroughly described by Yablonovitch et al. (Yablonovitch et al., 1988). Here, we will not repeat these arguments and note only that relationship (3.4) differs from the corresponding expression given by Yablonovitch et al. (Yablonovitch et al., 1988). The difference lies in the appearance of the factor f_L in relationship (3.4).

At last, we have (Pukhov et al., 2008; Basiev et al., 2008)

$$A_{nano} = n_{eff} f_N f_L(n_{cr}) A_0 = (n_{eff} / n_{cr}) f_N n_{cr} f_L(n_{cr}) A_0 = (n_{eff} / n_{cr}) f_N A_{bulk} \quad (3.6)$$

and for the A_{nano}/A_{bulk} get the following expression

$$A_{nano}/A_{bulk} = (n_{eff}/n_{cr}) f_N. \quad (3.7)$$

An important consequence of relationship (3.6) is that the ratio A_{nano}/A_{bulk} can be estimated without recourse to a particular local-field model. The problem of the theoretical determination of the ratio A_{nano}/A_{bulk} is reduced to the problem of determining the correction $f_N = (E^{(cr)}/E)^2$ (and, of course, to the problem of determining the effective refractive index n_{eff}).

Let us calculate the correction $f_N = (E^{(cr)}/E)^2$ for subwavelength spherical nanocrystals that have the radius R satisfying the condition $a_L \ll 2R \ll \lambda/2\pi$. The electrostatic approximation is applicable at this condition as it follows from the Lorenz-Mie solution to Maxwell's equations. In framework of the electrostatic approximation the electric field $E^{(cr)}$ within a dielectric sphere placed in the external electric field $E^{(med)}$ is equal to (Landau & Lifshitz, 1984)

$$E^{(cr)} = [3/(\varepsilon + 2)] E^{(med)} \quad (3.8)$$

where $\varepsilon = \varepsilon_{cr}/\varepsilon_{med} = n_{cr}^2/n_{med}^2$ is relative permittivity.

On the lines of the Maxwell Garnett theory (Maxwell Garnett, 1904; Maxwell Garnett, 1906) we obtain

$$f_N^{spher} = \{3 / [2 + \varepsilon - \varepsilon(\varepsilon - 1)]\}^2. \quad (3.9)$$

So, the spontaneous emission rate of a two-level atom in the spherical nanoparticle is given by expression (Pukhov et al., 2008; Basiev et al., 2008)

$$A_{nano}^{spher}/A_{bulk} = \frac{n_{eff}}{n_{cr}} \left[\frac{3}{2 + \varepsilon - x(\varepsilon - 1)} \right]^2. \quad (3.10)$$

(Although for definiteness, we consider nanocrystals, all the inferences refer equally to nanoparticles from a dielectric material with the refractive index n_{cr}).

The Eq. (3.1) together with relation

$$P = (1-x)P^{(med)} + x P^{(cr)}, \quad (3.11)$$

where P , $P^{(med)}$ and $P^{(cr)}$ are average polarizations on nanocomposite, medium and nanocrystal, lead to well known Maxwell Garnett mixing rule for ε_{eff} (Maxwell Garnett, 1904; Maxwell Garnett, 1906):

$$\varepsilon_{eff} = n_{eff}^2 = \varepsilon_{med} \left[1 + \frac{3x\beta}{1 - x\beta} \right], \quad (3.12)$$

where $\beta = (\varepsilon - 1)/(\varepsilon + 2)$. The Maxwell Garnett mixing rule predicts the effective permittivity ε_{eff} of a nanocomposite where homogeneous spheres of isotropic permittivity ε_{cr} dilutely mixed into isotropic medium with permittivity ε_{med} (see book (Bohren & Huffman, 1998) for details). As it can be seen from Eq. (3.10) and Eq. (3.12), the spontaneous emission rate in nanocomposite is enhanced for $\varepsilon < 1$ and inhibited for $\varepsilon > 1$.

From the expressions (3.10) and (3.12), for $x \rightarrow 1$, we obviously have the limiting case of $A_{nano}^{spher} \rightarrow A_{bulk}$. In the limit $x \rightarrow 0$, we obtain

$$A_{nano}^{spher}/A_{bulk} = \frac{n_{med}}{n_{cr}} \left[\frac{3}{2 + \varepsilon} \right]^2. \quad (3.13)$$

The derived expression is consistent with both the result obtained by Yablonovitch et al. (Yablonovitch et al., 1988) and result derived by Chew (Chew, 1988) also without regard for the local-field effect. Thereby, formula of Eq. (3.10) yields the correct result for $x \rightarrow 1$ and fit the results of Refs. (Chew, 1988) and (Yablonovitch et al., 1988) for $x \rightarrow 0$. It is not yet clear whether this formula is applicable for the intermediate values of filling factors x as the experimental data are scarce.

It should be mentioned that in some papers (Meltzer et al., 1999; Zakharchenya et al., 2003; Manoj Kumar et al., 2003; Vetrone et al., 2004; Chang-Kui Duan et al., 2005; Dolgaleva et al., 2007; Liu et al., 2008) expression (2.9) for the spontaneous radiative rate in a bulk crystal is transformed into the formula for the decay rate of an optical center in a crystalline nanoparticles A_{nano} by direct replacing the refractive index of the crystal n_{cr} by the effective refractive index n_{eff} and the local-field correction $f_L(n_{cr})$ by the corresponding correction $f_L(n_{eff})$ with the use of a particular local-field model:

$$A_{nano} = n_{eff} f_L(n_{eff}) A_0. \quad (3.14)$$

This leads to some arbitrariness in the interpretation of experimental data owing to the choice of the particular expression for the local-field correction $f_L(n)$ (this problem is discussed in the paper (Dolgaleva et al., 2007).

For the ratio between the excitation lifetimes of an optical center in a nanoparticle and a bulk crystal, expression (3.13) can be rearranged to give

$$\tau_{nano} / \tau_{bulk} = \frac{n_{cr}}{n_{med}} \left[\frac{2 + \varepsilon}{3} \right]^2 \quad (3.15)$$

Fig. 1. (1–3) Theoretical dependences of the ratio τ_{nano}/τ_{bulk} (the right axis) on the ratio n_{cr}/n_{med} for crystalline matrices with volume fractions (1) $x \rightarrow 0$, (2) $x = 0.2$, and (3) $x = 0.4$. (4–6) Theoretical dependences of the ratio $\sigma_{nano}/\sigma_{bulk}$ (the left axis) on the ratio n_{cr}/n_{med} with volume fractions (4) $x = 0.4$, (5) $x = 0.2$, and (6) $x \rightarrow 0$. Dependences of the measured ratio of the decay time in a nanocrystal to the decay time in a bulk crystal τ_{nano}/τ_{bulk} on the ratio n_{cr}/n_{med} for the $^4F_{3/2}$ metastable level of Nd³⁺ ions in the YAG crystalline matrix ($n_{cr} = 1.82$) (Dolgaleva et al., 2007) (circles) and the 5D_0 metastable level of Eu³⁺ ions in the Y₂O₃ crystalline matrix ($n_{cr} = 1.84$) (Meltzer et al., 1999) (squares).

The calculations have demonstrated that, for $x = 0$ and $n_{med} = 1$ (one nanoparticle is suspended in air), the lifetime τ_{nano} of excitation of an optical center in a nanoparticle can

increase as compared to the corresponding lifetime τ_{bulk} in a bulk crystal, for example, in Y_2O_3 ($n_{cr}/n_{med} = 1.84$) and YAG ($n_{cr}/n_{med} = 1.82$), by a factor of approximately 6 (Fig. 1, curve 1). According to expressions (3.10) and (3.12), an increase in the volume fraction x leads to a decrease in the ratio τ_{nano}/τ_{bulk} (Fig. 1, curves 2, 3).

4. Integrated emission and absorption cross - section

Apart from the lifetime of optical centers, the integrated emission and absorption cross - sections are important characteristics of laser materials. The integrated cross - section of the electric dipole emission in the band $i \rightarrow j$ for a bulk material can be represented in the form (Fowler & Dexter, 1962)

$$\sigma_{bulk}^{em}(i \rightarrow j) = A_{bulk}(i \rightarrow j) / [8\pi c_0 n_{cr}^2 \bar{\nu}^2], \quad (4.1)$$

where $A_{bulk}(i \rightarrow j)$ is the probability of spontaneous decay in the channel $i \rightarrow j$ for a bulk crystal, $\bar{\nu}$ is the average energy of the transition $i \rightarrow j$ (in cm^{-1}).

It is evident that, in order to determine the integrated cross - section of the electric dipole emission in the band $i \rightarrow j$ for a nanocrystal, it is necessary to replace the probability of spontaneous decay $A_{bulk}(i \rightarrow j)$ for the bulk crystal by the probability of spontaneous decay $A_{nano}(i \rightarrow j)$ for the nanocrystal and the refractive index n_{cr} by the effective refractive index n_{eff} in relationship (4.1). As a result, we obtain (Pukhov et al., 2008; Basiev et al., 2008)

$$\sigma_{nano}^{em}(i \rightarrow j) = A_{nano}(i \rightarrow j) / [8\pi c_0 n_{eff}^2 \bar{\nu}^2]. \quad (4.2)$$

After substituting the expression

$$A_{nano}(i \rightarrow j) = \frac{n_{eff}}{n_{cr}} f_N A_{bulk}(i \rightarrow j), \quad (4.3)$$

which was derived in much the same manner as expression (3.6) into relationship (4.2), we find (Pukhov et al., 2008; Basiev et al., 2008)

$$\sigma_{nano}^{em} = \frac{n_{cr}}{n_{eff}} f_N \sigma_{bulk}^{em}. \quad (4.4)$$

The same relationship holds true for the integrated cross - section of the electric dipole absorption; i.e., the integrated cross - sections of electric dipole processes of all types are described by the expression (Pukhov et al., 2008; Basiev et al., 2008)

$$\sigma_{nano} = \frac{n_{cr}}{n_{eff}} f_N \sigma_{bulk}. \quad (4.5)$$

In the special case of spherical nanoparticles, substitution of relationship (3.9) for the correction f_N^{spher} into expression (4.5) gives

$$\sigma_{nano} = \frac{n_{cr}}{n_{eff}} \left[\frac{3}{2 + \varepsilon - x(\varepsilon - 1)} \right]^2 \sigma_{bulk} \quad (4.6)$$

It is worth noting that the factor n_{cr}/n_{eff} in expressions (4.4) – (4.6) is the reciprocal of the factor n_{eff}/n_{cr} , which enters into the right-hand sides of relationships (3.7), (3.10), and (4.3). As a consequence, we have (Pukhov et al., 2008; Basiev et al., 2008)

$$\sigma_{nano}/\sigma_{bulk} = \left[\frac{n_{cr}}{n_{eff}} \right]^2 [A_{nano}/A_{bulk}] = \left[\frac{n_{cr}}{n_{eff}} \right]^2 [\tau_{bulk}/\tau_{nano}], \quad (4.7)$$

where $\tau_{bulk} = 1/A_{bulk}$ and $\tau_{nano} = 1/A_{nano}$.

It should be noted that, for example, at the refractive index $n_{cr} = 1.82$ (YAG), an increase in the time of radiative decay of optical centers in nanoparticles in an aerosol by a factor of 5 (as compared to that of the bulk crystal) results in a decrease in the corresponding emission cross - section by only 42% (Fig. 1, curves 1, 6). Moreover, the effect of an increase in the volume fraction x of nanoparticles on the decrease in the ratio $\sigma_{nano}/\sigma_{bulk}$ becomes considerably weaker as compared to that of the ratio τ_{nano}/τ_{bulk} (compare the changes in the ratios τ_{nano}/τ_{bulk} (Fig. 1, curves 1 - 3) and $\tau_{nano}/\sigma_{bulk}$ (Fig. 1, curves 4 - 6)). An insignificant decrease in the pump absorption and emission cross - sections cannot bring about a negative effect on the laser medium, whereas a fivefold increase in the lifetime at the same pump power makes it possible to increase the product $\sigma_{abs} \tau$ and, therefore, inversion accumulated in a laser generator or an amplifier by a factor of 5. Therefore, an increase in the lifetime of metastable levels in laser media makes it possible to decrease the power and the cost of a diode laser pumping source and to reduce the superluminescence losses without changes in the energy and power of the output emission owing to a several fold increase in the time of pulse pumping.

5. Influence of the shape of samples: Ellipsoidal nanoparticles

5.1 An isolated ellipsoidal nanoparticle ($x \rightarrow 0$)

We now analyze the influence of the shape of nanoparticles on the decay rate of optical centers in subwavelength ellipsoidal dielectric nanoparticles as an example. The mathematical complication arising in the analysis of ellipsoidal nanoparticles lies in the fact that the electric field $E^{(cr)}$ inside an ellipsoidal dielectric nanoparticle placed in the external electric field $E^{(med)}$ is not parallel to the field $E^{(med)}$ (Landau & Lifshitz, 1984). Let us consider this problem in more detail. First, we shall restrict our consideration to a special case $x \rightarrow 0$ (an isolated ellipsoid). In this case $n_{eff} \rightarrow n_{med}$ and $E^{(med)} \rightarrow E$. The electric fields $E^{(cr)}$ and E are related by the linear relation $E^{(cr)} = \hat{g}E$ (Landau & Lifshitz, 1984) with the tensor $\hat{g}$ principal values given by

$$g_{\alpha} = 1/[1 + (\varepsilon - 1)N_{\alpha}] \quad (\alpha = a, b, c) \quad (5.1)$$

where a, b, c are the principal axes of the ellipsoid and N_{α} are the depolarization factors

$(N_a + N_b + N_c = 1)$. Generally $g_a \neq g_b \neq g_c$.

The depolarization factors N_a are expressed through the elliptic integrals (Landau & Lifshitz, 1984):

$$N_a = \frac{abc}{2} \int_0^\infty \frac{ds}{(s + a^2)\sqrt{(s + a^2)(s + b^2)(s + c^2)}} \quad (5.2)$$

As a result, the Hamiltonian of interaction of electric field $\mathbf{E}^{(cr)}$ with dipole moment $\mathbf{d}$ takes the form (Pukhov, 2009)

$$H_{\text{int}} = -\hat{\mathbf{E}}^{(cr)} \hat{\mathbf{d}} = -\hat{\mathbf{d}} \hat{\mathbf{g}} \hat{\mathbf{E}} = -i \sum_{\mathbf{k}, \sigma} \sqrt{\frac{2\pi\hbar\omega_{\mathbf{k}}}{V\varepsilon_{\text{med}}}} \hat{\mathbf{d}} \hat{\mathbf{g}} \mathbf{e}_{\mathbf{k}, \sigma} \left[\mathbf{a}_{\mathbf{k}, \sigma} - \mathbf{a}_{\mathbf{k}, \sigma}^+ \right] \quad (5.3)$$

The averaging of the quantity $\left| \mathbf{d} \mathbf{e}_{\mathbf{k}, \sigma} \right|^2$ over all orientations of the polarization vector $\mathbf{e}_{\mathbf{k}, \sigma}$ in the isotropic field results in the usual expression $|\mathbf{d}|^2 / 3 = \sum_{\alpha} |d_{\alpha}|^2 / 3$ (where d_{α} are the components of the transition dipole moment $\mathbf{d}$), whereas the corresponding averaging in the anisotropic field leads to the expression $\sum_{\alpha} g_{\alpha}^2 |d_{\alpha}|^2 / 3$.

As a result, instead of expression (3.13) for a sphere, we obtain the following relationship for an ellipsoid (Pukhov et al., 2008; Basiev et al., 2008; Pukhov, 2009):

$$A_{\text{nano}}^{\text{ell}} / A_{\text{bulk}} = \frac{n_{\text{med}}}{n_{\text{cr}}} \sum_{\alpha=a,b,c} \left[\frac{\gamma_{\alpha}}{1 + (\varepsilon - 1)N_{\alpha}} \right]^2 \quad (5.4)$$

where $\gamma_{\alpha} = |d_{\alpha}| / \sqrt{\sum_{\alpha} |d_{\alpha}|^2}$ are the direction cosines of the transition dipole moment in the principal axes a , b , and c of the ellipsoid. (In the case of sphere, $N_a = N_b = N_c = 1/3$, so that Eq. (5.4) reduces to Eq. (3.13) for a sphere.) This means that, now, the ratio $A_{\text{nano}} / A_{\text{bulk}}$ depends on the dipole orientation with respect to the principal axes of the ellipsoid.

The anisotropy factor K , which is equal to the ratio of the probability of a transition in an ellipsoid $A_{\text{nano}}^{\text{ell}}$ to the probability of a transition in a sphere $A_{\text{nano}}^{\text{spher}}$ can be written in the form

$$K = \frac{A_{\text{nano}}^{\text{ell}}}{A_{\text{nano}}^{\text{spher}}} = \left[\frac{\varepsilon + 2}{3} \right]^2 \sum_{\alpha=a,b,c} \left[\frac{\gamma_{\alpha}}{1 + (\varepsilon - 1)N_{\alpha}} \right]^2 \quad (5.5)$$

For a sphere, we have $N_a = N_b = N_c = 1/3$ and, after the corresponding substitution and transformation, formula (5.5) leads to $K = 1$, as it must.

Ellipsoids of Revolution

From here on, we will consider only ellipsoids of revolution (where the c axis is the axial symmetry axis and the lengths of the semiaxes a and b are equal; i.e., $N_a = N_b$). In this case, expression (5.5) takes the form

$$K_{axial} = \left[\frac{\varepsilon + 2}{3} \right]^2 \left[\frac{1 - \gamma_c^2}{[1 + (\varepsilon - 1)N_a]^2} + \frac{\gamma_c^2}{[1 + (\varepsilon - 1)N_c]^2} \right], \quad (5.6)$$

where $N_a = N_b = (1 - N_c)/2$.

The elliptic integrals given by formula (5.2) are expressed through elementary functions for all ellipsoids of revolution (Landau & Lifshitz, 1984). For a prolate ellipsoid of revolution ($c > a = b$) with the eccentricity $e = \sqrt{1 - a^2/c^2}$, we have

$$N_c = \frac{1 - e^2}{e^3} (\text{Arth} e - e), \quad (5.7)$$

$$N_a = N_b = (1 - N_c)/2. \quad (5.8)$$

When the ellipsoid is closely similar to a sphere ($e \ll 1$), the depolarization factors are approximately represented by the formulas (Landau & Lifshitz, 1984)

$$N_c = 1/3 - 2e^2/15, \quad (5.9)$$

$$N_a = N_b = 1/3 + e^2/15. \quad (5.10)$$

For an oblate ellipsoid of revolution ($c < a = b$) with the eccentricity $e = \sqrt{a^2/c^2 - 1}$, the depolarization factors are written as (Landau & Lifshitz, 1984)

$$N_c = \frac{1 + e^2}{e^3} (e - \text{arctg} e), \quad (5.11)$$

$$N_a = N_b = (1 - N_c)/2. \quad (5.12)$$

For the eccentricity $e \ll 1$, the depolarization factors are given by the expressions (Landau & Lifshitz, 1984)

$$N_c = 1/3 + 2e^2/15, \quad (5.13)$$

$$N_a = N_b = 1/3 - e^2/15. \quad (5.14)$$

It can be seen from expression (5.6) that the dependence of the anisotropy parameter K_{axial} on the orientation of the transition dipole moment of the optical center is completely determined by the quantity γ_c , i.e., the projection of the transition dipole moment onto the axis of revolution c . For illustrative purposes, we will consider below several cases of the orientation of the transition dipole moment in long cylinders ($c \gg a = b$) and thin disks ($c \ll a = b$).

Case A

The transition dipole moment has nonzero components along the axis of revolution c and in the plane perpendicular to this axis ($\gamma_c^2 = (\gamma_a^2 + \gamma_b^2)/2 = 1/3$). The orientation of the crystallographic axes is chosen to be arbitrary with respect to the axes of the ellipsoid.

Cylindrical nanoparticles ($\gamma_c^2 = 1/3$). A dielectric cylinder is characterized by the depolarization factors $N_a = N_b = 1/2$ and $N_c = 0$. Setting $\gamma_c^2 = 1/3$ in relationship (5.6), we obtain

$$K_{cyl}(\epsilon) = \left[\frac{\epsilon + 2}{3(\epsilon + 1)} \right]^2 \frac{\epsilon^2 + 2\epsilon + 9}{3}. \quad (5.15)$$

Disk-shaped nanoparticles ($\gamma_c^2 = 1/3$). A dielectric disk is characterized by the depolarization factors $N_a = N_b = 0$ and $N_c = 1$. Setting $\gamma_c^2 = 1/3$ in relationship (5.6), we find

$$K_{disk}(\epsilon) = \left[\frac{\epsilon + 2}{3} \right]^2 \frac{2\epsilon^2 + 1}{3\epsilon^2}. \quad (5.16)$$

In both variants, the function $K(\epsilon)$ monotonically increases from the minimum at $\epsilon = 1$. This indicates that the lifetime of optical centers in nonspherical nanocrystals is shortened as compared to their lifetime in spherical nanocrystals. It should be noted that, for Y_2O_3 nanocrystals in air ($\epsilon = 3.4$), the lifetimes of optical centers in cylindrical and disk-shaped nanoparticles are shorter than that in spherical nanoparticles by factors of 1.5 and 2.2, respectively.

Case B

The transition dipole moment is perpendicular ($\gamma_c^2 = 0$) or parallel ($\gamma_c^2 = 1$) to the axis of revolution c .

Cylindrical nanoparticles ($\gamma_c^2 = 0$ or $\gamma_c^2 = 1$). For dipoles oriented perpendicular to the axis of revolution of the cylinder, we calculated $N_a = N_b = 1/2$, $N_c = 0$, and $\gamma_c^2 = 0$ in relationship (5.6) and derive the following expression (Fig. 2, curve 1):

$$K_{cyl}^{\perp}(\epsilon) = \left[\frac{2(\epsilon + 2)}{3(\epsilon + 1)} \right]^2. \quad (5.17)$$

For dipoles aligned parallel to the axis of revolution of the cylinder, we calculated $N_a = N_b = 1/2$, $N_c = 0$, and $\gamma_c^2 = 1$ in relationship (5.6) and obtain (Fig. 2, curve 2)

$$K_{cyl}^{\parallel}(\epsilon) = \left[\frac{\epsilon + 2}{3} \right]^2. \quad (5.18)$$

Disk-shaped nanoparticles ($\gamma_c^2 = 0$ or $\gamma_c^2 = 1$). For dipoles oriented perpendicular to the axis of revolution of the disk, we calculated $N_a = N_b = 0$, $N_c = 1$, and $\gamma_c^2 = 0$ in relationship (5.6) and find (Fig. 2, curve 2)

$$K_{disk}^{\perp}(\epsilon) = \left[\frac{\epsilon + 2}{3} \right]^2. \quad (5.19)$$

It should be noted that, according to relationships (5.18) and (5.19), we have the equality $K_{disk}^{\perp}(\epsilon) = K_{cyl}^{\parallel}(\epsilon)$.

For dipoles aligned parallel to the axis of revolution of the disk, we calculated $N_a = N_b = 0$, $N_c = 1$, and $\gamma_c^2 = 1$ in relationship (5.6) and derive (Fig. 2, curve 3)

$$K_{disk}^{\parallel}(\epsilon) = \left[\frac{\epsilon + 2}{3\epsilon} \right]^2. \quad (5.20)$$

Fig. 2. Dependences of the ratio of the optical excitation lifetime in a nanoparticle in the form of an ellipsoid of revolution to the optical excitation lifetime in a nanosphere on the ratio $(n_{cr}/n_{med})^2$ at different ratios between the lengths of the a , b , and c axes and the directions of the dipole moment $\mathbf{d}$ with respect to the axes of the ellipsoid: (1) nanocylinder for $\mathbf{d} \perp c$, (2) nanocylinder for $\mathbf{d} \parallel c$ and nanodisk at $\mathbf{d} \perp c$, and (3) nanodisk for $\mathbf{d} \parallel c$.

Therefore, the radiative decay time of optical centers in nonspherical nanoparticles can vary over a wide range as compared to that in spherical nanoparticles. For example, the radiative decay time of optical centers in the Y_2O_3 matrix doped with rare-earth ions can increase by a factor of about 3.5 in disk-shaped nanocrystals with a transition dipole moment aligned parallel to the axis of revolution of the disk and, on the contrary, can decrease by a factor of about 3.3 in disk-shaped nanocrystals with a transition dipole moment oriented perpendicular to the axis of revolution of the disk or in cylindrical nanocrystals with a transition dipole moment directed parallel to the axis of revolution of the cylinder (Fig. 2, curves 2 and 3 at $\varepsilon = 3.39$, respectively).

The anisotropy factor K substantially complicates the interpretation of experimental data: in order to calculate the value of K , it is necessary to know the mutual orientation of the principal axes of the ellipsoid (a , b , and c) with respect to the crystallographic axes of the nanocrystal. Let us consider the simplest case in which the nanoparticle is a nanocrystal with cubic symmetry and the crystallographic axes of the nanocrystal are parallel to the principal axes of the ellipsoid. Owing to the cubic symmetry of the crystal, the appearance of optical centers with a transition dipole moment aligned parallel, say, to the $[100]$ axis brings about the formation of an equal number of optical centers with transition dipole moments directed parallel to the $[010]$ and $[001]$ axes. This implies that the cubic crystal should contain optical centers of three types: optical centers of the first type are oriented parallel to the $[100]$ direction ($\gamma_a^2 = 1$, $\gamma_b^2 = \gamma_c^2 = 0$), optical centers of the second type are aligned parallel to the $[010]$ direction ($\gamma_b^2 = 1$, $\gamma_a^2 = \gamma_c^2 = 0$), and optical centers of the third type are directed parallel to the $[001]$ direction ($\gamma_a^2 = \gamma_b^2 = 0$, $\gamma_c^2 = 1$). Optical centers of all three types in a bulk crystal are equivalent and have the same lifetime, whereas a qualitatively different situation occurs with a nanocrystal. In the latter case, only the first two types of optical centers turn out to be equivalent (owing to the axial symmetry).

The luminescence decay kinetics of optical centers should be described by two exponential components (it is a manifestation of the "latent anisotropy" (Feofilov, 1961; Feofilov & Kaplyanskii, 1962) with a weight ratio of 2:1. In this case, the "fast and slow exponential components" should be characterized by lifetimes that are shorter and longer, respectively, than those observed in a nanosphere. For example, the luminescence kinetics of optical centers in the $\text{Y}_2\text{O}_3:\text{Yb}^{3+}$ nanocylinder ($n_{cr} = 1.84$) in air should exhibit a nonexponential behavior (Fig. 3a, curve 1) owing to the presence of two exponential components with equal weights and the same decay time $\tau_1 = 6.4$ ms (Fig. 3a, curve 2) (the transition dipole moment is perpendicular to the axis of revolution of the cylinder) and one exponential component with the decay time $\tau_2 = 1.3$ ms and a weight of 1/3 (Fig. 3a, curve 3) (the transition dipole moment is parallel to the axis of revolution of the cylinder). For comparison, Fig. 3a also shows the luminescence decay curves of optical centers in a nanosphere with the decay time $\tau_{sphere} = 4.3$ ms (Fig. 3a, curve 4) and in a bulk crystal with $\tau_{bulk} = 0.72$ ms (Fig. 3a, curve 5). It can be seen from Fig. 3a that the radiative decay time of optical centers in the $\text{Y}_2\text{O}_3:\text{Yb}^{3+}$ nanocylinder can increase by a factor of approximately 9 upon changing over from the bulk crystal to the cylindrical nanocrystal and by a factor of approximately 6 upon changing over to the spherical nanocrystal. A more pronounced nonexponential behavior is observed for the $\text{Y}_2\text{O}_3:\text{Yb}^{3+}$ nanodisk (Fig. 3b, curve 1). The fast and slow exponential components differ by a factor of approximately 12; in this case, we have $\tau_1 = 1.3$ ms with a weight of 2/3 (Fig. 3b, curve 2) (the transition dipole moment is perpendicular to the axis of revolution of the disk) and $\tau_2 = 15.3$ ms with a weight of 1/3 (Fig. 3b, curve 3) (the transition dipole moment is

Fig. 3. (a) Luminescence kinetic curves for (1) cubic symmetry optical centers in the $\text{Y}_2\text{O}_3:\text{Yb}^{3+}$ nanocylinder ($n_{cr}=1.84$) suspended in air for which the transition dipole moment $\mathbf{d}$ in equal proportion is parallel to one of the axes of the ellipsoid of revolution (a , b , or c); (2, 3) optical centers with the transition dipole moments (2) $\mathbf{d} \perp c$ and (3) $\mathbf{d} \parallel c$; (4) the nanosphere; and (5) the bulk crystal
 (b) Luminescence kinetic curves for (1) cubic symmetry optical centers in the $\text{Y}_2\text{O}_3:\text{Yb}^{3+}$ nanodisk ($n_{cr} = 1.84$) suspended in air for which the transition dipole moment $\mathbf{d}$ in equal proportion is parallel to one of the axes of the ellipsoid of revolution (a , b , or c); (2, 3) optical centers with the transition dipole moments (2) $\mathbf{d} \perp c$ and (3) $\mathbf{d} \parallel c$; (4) the nanosphere; and (5) the bulk crystal.

parallel to the axis of revolution of the disk). It is clear that, in the absence of axial symmetry, the process of luminescence decay should involve three exponential components. The obtained theoretical inference that the kinetics of radiative decay of excited optical centers of the same type in cubic nanocrystals exhibits a strongly nonexponential behavior is confirmed by the experimental data available in the literature (Christensen et al., 1982). It should be mentioned, that in a particular case of the cylinder nanocrystals with elliptical cross section (axes a, b) and an in-plane dipole moment ($d_c = 0$) we obtain from Eq. (5.4)

$$A_{nano}^{ell}/A_{bulk} = \frac{n_{med}}{n_{cr}} \left[\left(\frac{\gamma_a}{1+(\varepsilon-1)N_a} \right)^2 + \left(\frac{\gamma_b}{1+(\varepsilon-1)N_b} \right)^2 \right] \quad (5.21)$$

With the exception of the factor n_{med}/n_{cr} this expression coincides with result of Rogobete and Henkel for the cylinder nanocrystals with elliptical cross - section (see Eq.(A8) in Ref. (Rogobete&Henkel, 2004)). From our point of view the factor n_{med}/n_{cr} is important and cannot be dropped from Eq.(5.21).

It should also be noted that the known Judd–Ofelt parameters Ω_k (Krupke, 1966) for trivalent rare-earth ions in ellipsoidal nanocrystals should depend on the depolarization factors N_α and the orientation of the crystallographic axes with respect to the principal axes of the ellipsoid (see next Section 6).

A situation complicates when the spontaneous emission process involves several decay channels and each channel is determined by its own direction of the transition dipole moment. In this case, the branching ratio can also change for a nanocrystal. However, this circumstance can be used to obtain information on the shape of nanocrystals. The cases under consideration, of course, do not cover a wide variety of real optical centers in nanocrystals and, most likely, have only an illustrative character, because they have demonstrated the influence of the shape of nanoparticles on the spontaneous decay rate of optical centers. Nonetheless, the above analysis has shown that controlling the shape of samples is of primary importance for an adequate theoretical interpretation of experimental results. And visa versa, the deviation of the rate of kinetics of radiative decay from the theoretically predicted value for spherical samples has revealed morphological features of nanoparticles and can serve as an effective method for controlling their sphericity.

5.2 An ensemble of nanoellipsoids distributed in sizes and orientations ($x \neq 0$)

Let us now shortly consider a nanocomposite with the subwavelength ellipsoidal dielectric nanoparticles, which are identical in composition and shape but may be different in volume and orientation. We shall assume that all orientations are equally likely. Follow to Bohren and Huffman (Bohren & Huffman, 1998) we shall use equation

$$\mathbf{E} = (1-x)\mathbf{E}^{(med)} + x\langle \mathbf{E}^{(cr)} \rangle \quad (5.22)$$

where symbol $\langle \dots \rangle$ denotes averaging over orientations of ellipsoidal nanoparticles and the averaged fields $\mathbf{E}^{(med)}$ and $\langle \mathbf{E}^{(cr)} \rangle$ are connected by relation

$$\langle \mathbf{E}^{(cr)} \rangle = g\mathbf{E}^{(med)} \quad (5.23)$$

with

$$g = \sum_{\alpha} g_{\alpha} / 3. \quad (5.24)$$

Eqs.(5.22)-(5.24) together with the corresponding equations for averaged polarizations give a generalization of Maxwell Garnett mixing rule for ε_{eff} (Bohren & Huffman, 1998):

$$\varepsilon_{eff} = \varepsilon_{med} \left[1 + \frac{xg(\varepsilon - 1)}{(1-x) + xg} \right]. \quad (5.25)$$

For spheres ($g = 3/(\varepsilon+2)$) formula (5.25) leads to Maxwell Garnett mixing rule (3.12), as it must.

It follows from Eqs.(5.22)-(5.23) that

$$\langle \mathbf{E}^{(cr)} \rangle = \frac{g}{(1-x) + xg} \mathbf{E} \quad (5.26)$$

However, it is obvious that Eq. (5.26) is inappropriate for calculation of the decay rate of optical centers. With assumption that macroscopic electric field inside the specific ellipsoidal nanoparticle is given by

$$\mathbf{E}^{(cr)} = \frac{(1-x)\hat{\mathbf{g}} + xg}{(1-x) + xg} \mathbf{E}, \quad (5.27)$$

the Hamiltonian of interaction of electric field $\mathbf{E}^{(cr)}$ with dipole moment $\mathbf{d}$ takes the form

$$\begin{aligned} H_{int} &= -\hat{\mathbf{E}}^{(cr)} \hat{\mathbf{d}} = -\hat{\mathbf{d}} [(1-x)\hat{\mathbf{g}} + xg] \hat{\mathbf{E}} / [(1-x) + xg] = \\ &= -i [(1-x) + xg]^{-1} \sum_{\mathbf{k}, \sigma} \sqrt{\frac{2\pi\hbar\omega_{\mathbf{k}}}{V\varepsilon_{eff}}} \hat{\mathbf{d}} [(1-x)\hat{\mathbf{g}} + xg] \mathbf{e}_{\mathbf{k}, \sigma} [a_{\mathbf{k}, \sigma} - a_{\mathbf{k}, \sigma}^{\dagger}]. \end{aligned} \quad (5.28)$$

As a result, we obtain

$$\begin{aligned} A_{nano}^{ell} / A_{bulk} &= \\ &= \frac{n_{eff}}{n_{cr}} [(1-x) + xg]^{-2} \left[(1-x)^2 \sum_{\alpha=a,b,c} \gamma_{\alpha}^2 g_{\alpha}^2 + 2x(1-x)g \sum_{\alpha=a,b,c} \gamma_{\alpha}^2 g_{\alpha} + x^2 g^2 \right]. \end{aligned} \quad (5.29)$$

We lead to expression (5.4) when $x \rightarrow 0$. It follows from Eq. (5.25) that $n_{eff} \rightarrow n_{med}$ either. In the limit $x \rightarrow 1$ ($n_{eff} \rightarrow n_{cr}$), we obviously have $A_{nano}^{ell} \rightarrow A_{bulk}$. Hence, Eq. (5.29) can be considered as a "matching" formula both yielding the correct result for the limiting cases $x \rightarrow 0$ and $x \rightarrow 1$. In the case of spherical nanoparticles, $N_a = N_b = N_c = 1/3$, $g = g_a = g_b = g_c = 3/(2+\varepsilon)$ so that Eq. (5.29) reduces to Eq. (3.10), as it must.

6. Is Judd-Ofelt equation valid for nanocrystal?

All above mentioned in Section 5 results are derived for a two-level atom. Now let us consider the multilevel systems, such as rare-earth (RE) ions. The Eq. (5.4) leads to following expression for the spontaneous emission rate of $J \rightarrow J'$ multilevel transition of RE ion in the ellipsoidal nanoparticles (Pukhov, 2009)

$$A_{nano}^{ell}(J, J')/A_{bulk}(J, J') = \frac{n_{med}}{n_{cr}} \left(\frac{3}{2 + \varepsilon} \right)^2 S_{nano}^{ell}(J, J')/S_{bulk}(J, J'). \quad (6.1)$$

Here $S_{bulk}(J, J')$ is the line-strength of $J \rightarrow J'$ transition in a bulk crystal and $S_{nano}^{ell}(J, J')$ is “anisotropic line-strength” defined as

$$S_{nano}^{ell}(J, J') = \sum_{\alpha=a,b,c} h_{\alpha}^2 r_{\alpha}^2(J, J') \quad (6.2)$$

where

$$h_{\alpha} = \frac{\varepsilon + 2}{3} g_{\alpha} = \frac{\varepsilon + 2}{3[1 + (\varepsilon - 1)N_{\alpha}]}, \quad (6.3)$$

$$r_{\alpha}^2(J, J') = \sum_{B, B'} |(B|D_{\alpha}|B')|^2 \quad (6.4)$$

(in Eqs. (6.2)-(6.4), the coordinate axes coincide with the principal axes of the ellipsoid a, b, c). Hereinafter we use Judd notations (Judd, 1962). B and B' denote Stark states of the J - and J' -manifolds, respectively. From Eqs. (6.2)-(6.4), we have $S_{nano}^{spher}(J, J') = S_{bulk}(J, J')$ for a sphere, as it

must be because of invariance of the sum $\sum_{\alpha=x,y,z} \sum_{B, B'} |(B|D_{\alpha}|B')|^2$ with respect to rotations of coordinate axes.

According to Judd-Ofelt theory (Judd, 1962; Ofelt, 1962), the linestrength for trivalent RE ions is given by equation

$$S_{bulk}(J, J') = \sum_{\lambda=2,4,6} \Omega_{\lambda} \left| (\kappa J \parallel U^{(\lambda)} \parallel \kappa' J') \right|^2 \quad (6.5)$$

where Ω_{λ} are Judd-Ofelt intensity parameters and $(\kappa J \parallel U^{(\lambda)} \parallel \kappa' J')$ is the reduced matrix element of the unit tensor operator $U^{(\lambda)}$ of rank λ that specifies the $J \rightarrow J'$ transition.

Is the Judd-Ofelt equation valid for $S_{nano}^{ell}(J, J')$?

It can be shown using Judd's results (Judd, 1962) that a sum $\sum_{B, B'} |(B|D_{\alpha}|B')|^2$ ($\alpha = x, y, z$) can be expressed in terms of matrix elements $(\kappa J \parallel U^{(\lambda)} \parallel \kappa' J')$ in the arbitrary coordinate system. Namely,

$$r_{\alpha}^2(J, J') = \sum_{BB'} \left| \langle B | D_{\alpha} | B' \rangle \right|^2 = \sum_{\lambda} \Omega_{\alpha}^{(\lambda)} \left| \langle \kappa J \| U^{(\lambda)} \| \kappa' J' \rangle \right|^2 \quad (6.6)$$

where

$$\Omega_x^{(\lambda)} = \frac{1}{2} \left[\Omega_{1,1}^{(\lambda)} + \Omega_{-1,-1}^{(\lambda)} - \Omega_{1,-1}^{(\lambda)} - \Omega_{-1,1}^{(\lambda)} \right] \quad (6.7a)$$

$$\Omega_y^{(\lambda)} = \frac{1}{2} \left[\Omega_{1,1}^{(\lambda)} + \Omega_{-1,-1}^{(\lambda)} + \Omega_{1,-1}^{(\lambda)} + \Omega_{-1,1}^{(\lambda)} \right] \quad (6.7b)$$

$$\Omega_z^{(\lambda)} = \Omega_{0,0}^{(\lambda)}. \quad (6.7c)$$

In Eqs. (6.7)

$$\Omega_{qq'}^{(\lambda)} = (2\lambda + 1) \sum_{p,t,t'} \begin{pmatrix} 1 & \lambda & t \\ q & -p-q & p \end{pmatrix} \begin{pmatrix} 1 & \lambda & t' \\ q' & -p-q & p+q-q' \end{pmatrix} A_{tp} A_{t'p+q-q'}^* \Xi(t, \lambda) \Xi(t', \lambda) \quad (6.8)$$

(expressions for A_{tp} and $\Xi(t, \lambda)$ are given in Judd's paper (Judd, 1962)).

As a result we have that expression (6.2) for $S_{nano}^{ell}(J, J')$ takes the form (Pukhov, 2009)

$$S_{nano}^{ell}(J, J') = \sum_{\lambda} \Omega_{\lambda}^{ell} \left| \langle \kappa J \| U^{(\lambda)} \| \kappa' J' \rangle \right|^2 \quad (6.9)$$

where intensity parameters are equal to

$$\Omega_{\lambda}^{ell} = \sum_{\alpha=a,b,c} h_{\alpha}^2 \Omega_{\alpha}^{(\lambda)}. \quad (6.10)$$

Hence the expression for the spontaneous emission rate of $J \rightarrow J'$ transition of RE ion in the subwavelength ellipsoidal nanocrystals is of the same form as the Judd-Ofelt expression for a bulk crystal. However, the Judd-Ofelt intensity parameters for trivalent rare-earth ions in ellipsoidal nanocrystals should depend on the depolarization factors and the orientation of the crystallographic axes with respect to the principal axes of the ellipsoid. The intensity parameters for a sphere are equal to the bulk intensity parameters.

7. Comparson of the theoretical data with the experimental data results

In this section, the ratios $\tau_{nano} / \tau_{bulk}$ calculated from relationship (3.15) for spherical nanoparticles with a radius $2R \ll \lambda$ and a volume fraction $x \rightarrow 0$ are compared with the corresponding measured quantities taken from paper (Dolgaleva et al., 2007) for the ${}^4F_{3/2}$ level of Nd^{3+} ions in YAG nanocrystals and from paper (Meltzer et al., 1999) for the 5D_0 level of Eu^{3+} ions in Y_2O_3 nanocrystals in suspensions as a function of the ratio n_{cr}/n_{med} (Fig. 1). In the case when nanoparticles have an identical shape (morphology) but are not spherical, instead of expression (3.15), it is necessary to use the general formula (5.4), which leads to the following relationship for the ratio between the decay times:

$$\frac{\tau_{nano}^{ell}}{\tau_{nano}^{spher}} = \left[\frac{3}{n_{cr}^2/n_{med}^2 + 2} \right]^2 \sum_{\alpha=a,b,c} \left[\frac{1 + (n_{cr}^2/n_{med}^2 - 1)N_{\alpha}}{\gamma_{\alpha}} \right]^2. \quad (7.1)$$

As can be seen from relationship (7.1), the smaller the ratio n_{cr}/n_{med} , i.e., the closer the refractive index of the nanoparticle n_{cr} to the refractive index of the surrounding medium n_{med} , the smaller the change in the rate of radiative relaxation of the rare-earth ion upon deviation of the shape of the nanoparticle from spherical, i.e., the weaker the effect of the nanoparticle morphology on the radiative relaxation rate. This tendency is clearly illustrated by the measured rates of radiative relaxation of the $^4F_{3/2}$ level of Nd^{3+} ions in YAG:(0.9 at % Nd^{3+}) nanoparticles with a diameter $2R = 20$ nm and a small volume fraction ($x = 1.1 \times 10^{-3}$) in different immersion liquids (Fig. 1, circles). The size of the circles in Fig. 1 corresponds to the measurement error. It can be seen from Fig. 1 that the experimental points fit the theoretical curve fairly well at a ratio $n_{cr}/n_{med} < 1.18$. The deviation of the experimental data from the theoretical results becomes noticeable at a ratio $n_{cr}/n_{med} > 2$ and increases with an increase in its value. It should be remembered that, even at the late stage of the luminescence decay kinetics, the nonradiative relaxation due to the Nd-Nd concentration quenching can not be ignored at a Nd^{3+} content of 0.9 at % (Voron'ko et al., 1973). This can also result in a deviation of the experimental dependence τ_{nano}/τ_{bulk} versus n_{cr}/n_{med} from the theoretical curve obtained for YAG:Nd nanocrystals. In order to minimize the contribution of the nonradiative quenching to the luminescence kinetics, the lifetime, for example, in the YAG:Nd $^{3+}$ crystal ($\tau_{bulk} = 255 \mu s$ (Basiev et al., 1974), was determined at a considerably lower concentration of Nd^{3+} ions as compared to that in the nanoparticles studied in (Dolgaleva et al., 2007).

Much better agreement with the theoretical curve, already at the ratio $n_{cr}/n_{med} = 1.4$, is observed for the decay time of the fluorescence associated with the 5D_0 level (not affected by quenching) of Eu^{3+} ions in Y_2O_3 :(0.1% Eu^{3+}) nanoparticles with a diameter $2R = 7-10$ nm (Fig. 1, squares) (Meltzer et al., 1999). The deviation observed at a ratio $n_{cr}/n_{med} \rightarrow n_{cr}$ can quite possibly be explained by the considerable volume fraction of the nanoparticles, their agglomeration, or deviation from sphericity.

8. Conclusion

Two principal physical reasons provide a basis for a change of spontaneous emission lifetime of an optical center, which moves from bulk crystal to nanocrystal. First, it is a change in photon density of states; second, it is a change of zero-point amplitude of electric-field modes, which are responsible for spontaneous emission. On this basis, an analytical expression is obtained for the electric-dipole radiative-decay rate of an excited optical center in an ellipsoidal dielectric nanoparticles (with sizes much less than the wavelength of light) embedded in a dielectric medium. It is found that the ratio of the decay rate A_{nano} of an excited optical center in the nanoparticle to the decay rate A_{bulk} of an excited optical center in the bulk sample is independent of the local-field correction and, therefore, of the adopted local-field model. The relation of the ratio A_{nano}/A_{bulk} with refractive indexes of the nanoparticles (n_{cr}) and the medium (n_{med}) and a volume fraction x of nanoparticles in the nanocomposite is established. The ratio of the absorption/emission cross-section $\sigma_{nano}/\sigma_{bulk}$ for spherical nanoparticles in dielectric medium to that in bulk crystal $\sigma_{nano}/\sigma_{bulk}$ is derived

as well. Here, the functional dependence of $A_{\text{nano}}/A_{\text{bulk}}$ and $\sigma_{\text{nano}}/\sigma_{\text{bulk}}$ ratios are found to be different. Strong increase in the radiative decay time in a nanoparticle in comparison with the bulk crystal gives rise to only a slight decrease in the corresponding cross - section for absorption and emission.

Comparison of the experimental and theoretical values of TR^{3+} metastable levels decay time τ in the YAG and Y_2O_3 spherical nanocrystals with those in bulk crystals proves the theory.

Thus, the above analysis of the theoretical expressions and the experimental results has demonstrated that the radiative characteristics of nanoparticles differ significantly from those of a bulk crystal. By varying the volume fraction x of nanoparticles in a suspension or an aerosol, the refractive index of the medium n_{med} surrounding the nanoparticles and their morphology it is possible to control the rates of spontaneous transitions and absorption and emission cross - sections of induced transitions. Thus, it is possible to increase $\sigma \times \tau$ product, an important laser parameter, several times and raises a population inversion in nanocomposite laser medium. By this method, one can control the laser properties of the nanocomposite materials and, thus, to design novel laser and luminescent media with improved characteristics.

The results obtained can be used in developing the fluorescence kinetic method for controlling the shape of nanoparticles and the degree of their agglomeration during subsequent applications to the synthesis of optical laser ceramic materials, control of the luminescence lifetime of nanophosphors, and observation of nanoagglomerates in organic and biological structures.

9. References

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Recent Optical and Photonic Technologies

Edited by Ki Young Kim

ISBN 978-953-7619-71-8

Hard cover, 450 pages

Publisher InTech

Published online 01, January, 2010

Published in print edition January, 2010

Research and development in modern optical and photonic technologies have witnessed quite fast growing advancements in various fundamental and application areas due to availability of novel fabrication and measurement techniques, advanced numerical simulation tools and methods, as well as due to the increasing practical demands. The recent advancements have also been accompanied by the appearance of various interdisciplinary topics. The book attempts to put together state-of-the-art research and development in optical and photonic technologies. It consists of 21 chapters that focus on interesting four topics of photonic crystals (first 5 chapters), THz techniques and applications (next 7 chapters), nanoscale optical techniques and applications (next 5 chapters), and optical trapping and manipulation (last 4 chapters), in which a fundamental theory, numerical simulation techniques, measurement techniques and methods, and various application examples are considered. This book deals with recent and advanced research results and comprehensive reviews on optical and photonic technologies covering the aforementioned topics. I believe that the advanced techniques and research described here may also be applicable to other contemporary research areas in optical and photonic technologies. Thus, I hope the readers will be inspired to start or to improve further their own research and technologies and to expand potential applications. I would like to express my sincere gratitude to all the authors for their outstanding contributions to this book.

How to reference

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K.K. Pukhov, Yu.V. Orlovskii and T.T. Basiev (2010). Spontaneous and Stimulated Transitions in Impurity Dielectric Nanoparticles, Recent Optical and Photonic Technologies, Ki Young Kim (Ed.), ISBN: 978-953-7619-71-8, InTech, Available from: <http://www.intechopen.com/books/recent-optical-and-photonic-technologies/spontaneous-and-stimulated-transitions-in-impurity-dielectric-nanoparticles>

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