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Modeling optical low-threshold exciton nonlinearity in dielectric nanocomposites

D.V. Storozhenko[a]*, V.P. Dzyuba[a], Y.N. Kulchin[a,b], A.V. Amosov[a]

[a] *Institute of Automation and Control Processes, FEB Russian Academy of Sciences, 5 Radio Street, Vladivostok, 690041, Russia*
[b]*Far Easern Federal University, 8 Suhanova Street, Vladivostok, 690950, Russia*

Abstract

We report on calculations of exciton nonlinearity in dielectric nanocomposites. The effect of various parameters on the spectrum of nonlinear increment to the refractive index, such as size and form factor of the nanoparticles shown. Numerical simulations of the optical response of dielectric nanoparticles Al_2O_3 presented.

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Keywords: dielectric nanoparticles; low-threshhold nonlinearity; photoluminescence; exitons

1. Introduction

Unusual optical properties of dielectric nanocomposites actively investigated in the last decade. Special attention focused to features that are weak or absent in bulk dielectrics. For example, detected of additional environmental impact properties (Ganeev and Usmanov(2007)), an external field (Kecherenko andNalbanyan(2012)), the size and nature of the particle shape (Dzuba et. al.(2008)) on the transmission spectra and luminescence. Also found in(Dzuba et. al. (2010), Ganeev et. al.(2008)), the appearance of the nonlinear optical response of some dielectric nanocomposite materials in the range of intensity of the order of less than 1 kW/cm^2, i.e. insufficient to run the multiphoton processes, photoionization or other non-linear processes. In (Milichko et al. (2013),Dneprovskii et

* Corresponding author. Tel.: +79242402258;
*E-mail address:*dbrados@dvo.ru

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al.(2010), Kupchak et al. (2008), He et al.(2004), Dneprovskii et al.(2013)) the authors, discussing the nature of the nonlinearity of this type conclude that the reason for increased impact defective or exciton levels in the dielectric particles on the overall transmission spectrum. In (Milichko et al. (2013)) invited the conditions conducive to emergence of resistant exciton states in a substance consisting of dielectric nanoparticles embedded in a transparent dielectric matrix liquid with linear optical properties in the optical range. Published in (Dzuba V. et al.(2013)) results of experimental studies of such substances have shown that at intensity of radiation about 150-250 W/cm^2 the nonlinear addition to the refractive index near the resonant frequency of the absorption band was $n_n=10^{-4}\div10^{-5}$. This is consistent with the proposed in [14] the theoretical model of such non-linearity. However, for a good prediction of the optical properties of nanocomposite dielectric materials necessary to investigate the influence of the size, shape and concentration of nanoparticles, it is possible with the help of numerical calculations with partial use of empirical data when setting the simulation parameters.

2. Description theory of exciton nonlinearity

The basis of the model used in the study is the excitation of resonant two-tier system (Shen(1984)), in which a nonlinear optical response of the medium is proportional to the complex susceptibility per unit volume of the optical path. Considering that in the range of intensity up to 1000 W/cm^2 total susceptibility dielectric substance $\tilde{\chi}$ is the sum of the linear and nonlinear susceptibilities:

$$\tilde{\chi} = \tilde{\chi}_0 + \tilde{\chi}_R \tag{1}$$

$$\tilde{\chi}_R = \frac{Np^2 \Delta\rho_{ng}(\omega, I)}{\hbar} \cdot \frac{(\omega-\omega_0) + i\Gamma}{(\omega-\omega_0)^2 + \Gamma^2} \tag{2}$$

$$\Delta\rho_{ng}(\omega, I) = \Delta\rho^0 \left[1 - \frac{I/I_s}{(\omega-\omega_0)^2 + \Gamma^2(1 + I/I_s)}\right] \tag{3}$$

Where $\tilde{\chi}_0$ is linear part of the susceptibility of the dielectric nanoparticles, $\tilde{\chi}_R$ – the resonant nonlinear correction to the susceptibility of the dielectric nanoparticles, ω_0 – resonance frequency, Γ – the half-width of the absorption line, $\hbar$ – Planck's constant with a bar, N – number of charge carriers in the bulk of the optical path, $\Delta\rho_{ng}$ – the difference between the populations of the energy levels of states $|n>$ and $|g>$, $p =< n|e \cdot r_z|g >$ – the projection of the total electric dipole moment of the transition of the electron with charge e nanoparticles out of $< n|$ in state $|g >$ on direction of polarization of the external optical radiation, $\Delta\rho^0$ – the equilibrium difference between the thermal of the population in the absence of an external field, I_s – the level of saturation of a two-tier system in which the excited state has moved half of the charge carriers.

Note that a priori unknown dipole moment $p =< n|e \cdot r_z|g >$, which does not know the value of the vector projection r_z. Therefore, the introduction of orientation factor $A(I)$ can decide this problem:

$$p = p_0 A(I) \tag{4}$$

We believe that the maximum value of the dipole moment $p_0 = a \cdot e$ is proportional to the size of the nanoparticles. The dependence of orientation factor from intensity I given in the form of a power model:

$$A(I) = 1 - \mathrm{e}^{-I/\alpha} \tag{5}$$