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The theory of heterogeneous dielectric nanostructures with non-typical low-threshold nonlinearity

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ABSTRACT

The recently discovered, ultralow-threshold, nonlinear refraction of low-intensity laser radiation in dielectric nanostructures has an atypical dependence on radiation intensity in the pulsed and continuous modes. In this study, we present a theoretical explanation. The theory suggests that the nonlinearity is photoinduced in nature, rather than thermal, and depends directly on the nanoparticle electronic structure and the relationship between permittivities of the dielectric matrix and the nanoparticles.
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1. Introduction

Over the last 20 years, significant scientific attention has been paid to nanostructures and nanocomposites based on nanoparticles of semiconductor materials (1 eV < E_{gap} < 3 eV). This response occurs in the visible and infrared region of the electromagnetic spectrum and reaches a maximum then decreases to zero with increasing intensity [1–5]. The dielectric nanostructures show other unexpected nonlinear optical properties. The non-linear interaction of the high-intensity radiation of different frequencies results in the generation of harmonics in conventional dielectric media. In the case of the propagation of the low-intensity radiation of different frequencies in the dielectric nanostructures, the nonlinear interaction is manifested in the dependence of the light beam intensity on the intensity of another collinearly propagating light beam [6]. The two-frequency interaction observed in the nanostructures does not prevent the generation of harmonics, but this process requires radiation intensities four orders of magnitude higher.

The study of nonlinear optical properties of dielectric nanostructures containing nanoscale objects of different chemical natures, shapes and sizes has shown that the existence of a low-threshold optical response is due to a number of conditions. The first is the presence of defect levels in the band gap of nanoparticles' charge carriers, which is manifested in the form of absorption bands in the nanoparticles' transmission spectrum [1,2,7–9]. Second, the radiation forming the nonlinear response of the nanostructure must have a frequency lying within the absorption band [1,2]. Third, the size and shape of the nanoparticles have to lead to the formation of a wide range of exciton states due to the quantum size effect [10–14]. Fourth, the matrix permittivity must be less than that of the nanoparticle material because the chemical nature of the matrix material significantly affects the formation of long-lived exciton states [2,7,15]. Fifth, the value of the electric dipole moments induced by electron phototransition should be substantially larger than the dipole moments in the bulk material, which allows for observation of the optical nonlinearity of nanostructures with a low concentration of nanoparticles under low-intensity optical fields.

The theoretical description of the observed effects [16–18] is based on the fact that the occurrence of non-typical optical nonlinearity requires the existence of defect levels and a broad band of exciton states in the energy band gap of charge carriers. The radiation causes electron transition from the defect to the exciton levels, thereby creating the photo-induced population difference. This process is accompanied by the appearance of the nanoparticle electric dipole moment, whose module depends nonlinearly on the intensity and the light wavelength. The theory conclusions and theoretical modelling of the transmission spectrum and the behaviour of the nonlinear refractive index are similar to the experimental results [2]. It follows from the theory that the nature of the nonlinearity is determined not only by the behaviour of the

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photo-induced dipole moment module in an external field but also by the nanoparticle's orientation along the vector E. However, this orientation has a minor contribution to the nonlinearity, so the observed nonlinear optical response can occur in the case of unpolarized light and solid nanostructures, which is in agreement with the experiment. This chapter is an original quantitative study of the nonlinear refraction and absorption of continuous low-intensity laser radiation in different heterogeneous dielectric nanostructures and compares these data with theoretical results. In addition, the theory of nonlinear light transmission by dielectric nanostructures is discussed.

2. Preface

The theoretical description of the physical and optical properties of heterogeneous nanocomposites containing nanoparticles is a complex problem. It seems impossible to calculate correctly the physical characteristics of an individual nanoparticle as a system consisting of a great number of particles obeying the quantum mechanics laws. Attempts to apply the methods of solid state physics to describe the nanoparticles' properties run into problems because it is not possible to disregard the effects caused by surface defects, as well as crystal lattice defects. The optical properties of a quantum mechanical system are associated with the features of the energy spectrum of the charge carriers (electrons and holes).

The optical and electric properties of nanoparticles have significant differences compared to bulk samples due to the features of the energy spectra. These differences are caused by three effects. First, the band gaps of nanoparticle charge carriers contain the allowed energies zone, and the energy structure is defined by the high density of the surface structural defects and the irregular shape of the nanoparticles. Second, the excitons and discrete energy spectra are formed below and into the conduction band due to the small nanoparticle size and size-quantization effect, respectively. In turn, the size quantization effect is caused by spatial confinement of the charge carriers' wave functions. Third, the electric dipole moments of the electronic transitions in such quasi-zero dimensional systems can be larger than that of the bulk sample. The formation of the above states is of threshold character, and the threshold depends on the nanoparticle dimensions. Specifically, for a spherical nanoparticle (with permittivity ε_2) dispersed in a medium (ε_1), such states can be formed if the nanoparticle radius α is smaller than some critical radius α_c:

$$\alpha \le \alpha_c = 6|\beta|^{-1}\alpha_{e,h} \tag{1}$$

where

$$\frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2} \tag{2}$$

Here, $\alpha_{e,h}$ is the Bohr radius of the charge carriers in the nanoparticle material [19].

Some properties of the quantum states' spectrum can be clarified by studying the nanocomposites' transmittance spectra. As a rule, experimental studies are concerned with the transmittance spectra of nanoparticles' arrays embedded in a solid matrix or deposited on a transparent material surface. In this case, the electronic structure of nanoparticles is substantially influenced by the matrix material and the interaction between nanoparticles. Because of these effects, it is not possible to consider the transmittance spectra as the spectra of no photonics of heterogeneous dielectric nanostructures interacting nanoparticles' arrays. Nanocomposites containing low concentrations of nanoparticles almost satisfy the condition due to the lack of the above interactions; however, to study the optical properties of such composites, one cannot take into account the effects of the optical field on the distribution of the particles through the degrees of freedom. In this case, given the low-intensity radiation, the optical field effect on the coordinates of a gravity centre of a nanoparticle can be disregarded, which cannot be said for the distribution of particles throughout the rotational degrees of freedom.

There is no well-known theoretical approach taking into account the characteristics of the nanoparticle dimensions, the orientation of nanoparticles in the external field of laser radiation, and the dependences of the scattering and absorption cross-sections on the propagating radiation intensity. In this context, it is necessary to develop a theoretical model of the scattering and absorption cross-sections in dielectric nanocomposites with the abovementioned features of such systems.

In this study, we suggest a semiphenomenological model of the optical transmittance of the array of non-interacting, small-sized ($\alpha < \alpha_c$), dielectric nanoparticles embedded in the dielectric matrix. We show that the basic mechanisms of the low-threshold effects of nonlinear scattering and the absorption of laser radiation in the HDN are: the photo induction of electric dipole moments of nanoparticles in the external optical field and the orientation of nanoparticles along the polarization direction of this field. In addition, we discuss the behaviour of the HDN transmittance in the central frequency vicinity of the absorption band and the dependence of the band depth on the radiation intensity.

3. Theoretical approach

We consider the HDN consisting of a low concentration (the number of nanoparticles N per unit volume) of dielectric nanoparticles embedded in an isotropic transparent dielectric matrix with a small coefficient of viscosity and linear optical properties within the visible spectral range. In our case, the multiple scattering of radiation by nanoparticles and the nanoparticles' interaction with each other can be neglected. Let us introduce two coordinate systems with the same origin (Fig. 1).

One of the systems $\{\alpha_1, \alpha_2, \alpha_3\}$ corresponds to the coordinates coinciding with the principle axes of the particle polarization tensor with the unit vectors (n_1, n_2, n_3). The other system is the Cartesian laboratory coordinate system {x, y, z} with the unit vectors (n_x, n_y, n_z). We suggest that the electromagnetic wave polarized along the z-axis E = {0, 0, E} is incident on the composite. We chose the x-axis to be directed collinearly with a wave vector.

The optical transmittance of the HDN depends on the extinction coefficient, the path of the light beam in the material and the

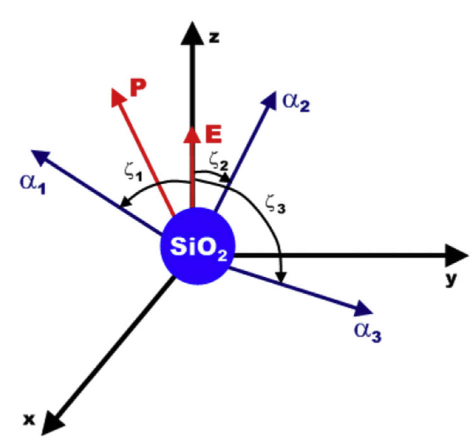

Fig. 1. The coordinate system used in the theoretical study.

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