ISSN 1995-0780, Nanotechnologies in Russia, 2008, Vol. 3, Nos. 11–12, pp. 743–750. © Pleiades Publishing, Ltd., 2008.
Original Russian Text © A.G. Vitukhnovskii, A.A. Isaev, V.S. Lebedev, 2008, published in Rossiiskie nanotekhnologii, 2008, Vol. 3, Nos. 11–12.

EXPERIMENT

Light-Induced Nonlinearity of CdSe/ZnS Quantum Dots with a Millisecond Relaxation Time

A. G. Vitukhnovskii[a]*, A. A. Isaev[a], and V. S. Lebedev[a, b]

[a] *Lebedev Physical Institute, Russian Academy of Sciences, Leninskii pr. 53, Moscow, 119991 Russia*
[b] *Moscow Institute of Physics and Technology (State University), Institutskii per. 9, Dolgoprudnyi, Moscow oblast, 141700 Russia*
* *e-mail: alexei@sci.lebedev.ru*

Received January 23, 2007

Abstract—The laser beam transmission through a colloidal solution of CdSe/ZnS quantum dots in toluene is investigated for the case of a continuous sequence of femtosecond pulses (100 fs) at a frequency of 100 MHz and nanosecond pulses (15 ns) at a frequency of 10 kHz. It is found that a giant nonlinearity is photoinduced in the system. The magnitude of the photoinduced nonlinearity is determined from the number of rings in the interference structure of the intensity distribution in a distant zone of the beam transmitted through the cell. It is established that the induced nonlinearity of the colloidal solution of quantum dots depends linearly on the average power of laser beams. The time required to attain a stationary regime of the induced nonlinearity of the colloidal solution of quantum dots in toluene (i.e., the transient period) is determined and lies in the millisecond range. From analyzing the photoinduced nonlinearities measured under different excitation conditions, it is concluded that the observed nonlinearity exhibits a nonthermal nature. Possible physical mechanisms of the processes that can lead to long-lived induced nonlinearities in the medium are discussed. The general regularities are revealed in the processes responsible for the appearance of a giant nonlinearity of quantum dots in the toluene solution and the processes giving rise to blinking effects of quantum dots in organic matrices. The results of the investigations performed are of considerable interest for the development of nonlinear optical switches and nanophotonic devices based on composite nanomaterials.

DOI: 10.1134/S1995078008110128

INTRODUCTION

One of the directions in nanotechnology is associated with the development of the circuitry type of nanophotonic devices of the new generation, such as nonlinear optical shutters, switches, and nanophotonic integrated circuits [1, 2]. Composite nanomaterials based on cadmium chalcogenide binary semiconductor quantum dots hold much promise for use in the design of the aforementioned devices [3, 4]. Investigation of these materials is of fundamental and practical interest owing to the possibility of controlling their optical characteristics, including absorption and luminescence spectra and nonlinear optical responses. This provides, in particular, the basis for the effective solution of the problem regarding the dynamic control over transmission of ultrashort light pulses both through colloidal solutions of semiconductor quantum dots and through films prepared from polymers with embedded quantum dots. Since the dynamics of charge carriers in quantum-confined objects differs substantially from their dynamics observed in massive materials, the corresponding spectral and nonlinear optical characteristics also differ significantly.

The nonlinear optical response of massive dielectric and semiconductor materials was previously investigated in [5–8]. For quantum-confined systems, apart from the well-known mechanisms responsible for induction of nonlinear susceptibility [5–8], it is necessary to take into account other light-induced processes that are associated with the structure of the energy spectrum, which, in this case, differs from the structure of the energy spectrum inherent in massive materials. At present, the systems containing cadmium chalcogenide quantum dots have been studied with particular emphasis placed on the femtosecond dynamics of charge carriers, which are promising for use in many applications, as well as on the related nonlinear optical response [9, 10]. These investigations have been performed with the use of femtosecond and picosecond laser pulses in single-pulse or very short pulse-train modes with very low values of the average power [11, 12].

It is worth noting that, in real devices (for example, nonlinear optical switches, lasers, light-emitting devices), quantum-dot semiconductor materials should operate under the conditions providing for both high-frequency switching and high values of the average radiation power. These operation conditions of quantum-confined systems have not been adequately studied. In this respect, the purpose of the present work was to investigate the optical properties and effects of interaction of composite semiconductor nanomaterials (containing cadmium chalcogenide quantum dots) with

radiation of repetitively pulsed lasers that ensure high values of the average radiation power and high pulse-repetition rates.

In our investigations, we measured the changes observed in the transverse intensity distribution of a laser beam during its transmission through a colloidal solution of CdSe/ZnS quantum dots in toluene. It is known that these changes are associated primarily with the light-induced nonlinear refractive index. Since the medium under consideration exhibits an inertialess nonlinear optical response [9–13], the investigations performed upon excitation with femtosecond pulses at a high repetition rate are of particular interest. For this purpose, we used a femtosecond (100 fs) titanium sapphire laser with a pulse-repetition rate of 100 MHz. In the investigations with high average powers of laser radiation, we used a repetitively pulsed copper vapor laser with a nanosecond (15 ns) excitation. The investigations performed in this work have revealed that, in these two excitation regimes, the initial Gaussian intensity distribution of the laser beam incident on a cell transforms into a distribution with a characteristic interference pattern containing a large number of rings. The nonlinear refractive index of the medium was calculated from the number of interference rings. The analysis of the results obtained in our experiments and the experimental data available in the literature made it possible to establish general regularities in the processes responsible for the appearance of a giant nonlinearity of quantum dots in the toluene solution and the processes giving rise to blinking effects of single quantum dots in organic matrices. The results obtained are in good agreement with the electronic mechanism of the generation of the light-induced nonlinearity.

OBJECT OF INVESTIGATION AND EXPERIMENTAL TECHNIQUE

The object of our investigation was a solution of CdSe/ZnS quantum dots (each composed of the CdSe core and the ZnS shell) in toluene. The CdSe/ZnS quantum dots were obtained from Evident Technology LLC (United States). We used two types of CdSe/ZnS quantum dots with diameters equal to 2.6 and 1.9 nm. The concentration of CdSe/ZnS quantum dots in the toluene solution was approximately equal to 10^{14} cm^{-3}. The toluene solution with quantum dots was poured into quartz cells 1 and 5 mm thick. The quartz cells were placed in a laser beam, and the spot of the light beam transmitted through the quartz cell was observed on a screen separated from the cell by a distance of 2.5 m. The beam spot was photographed with a digital camera.

Laser beams were obtained from two lasers used as sources: (i) a femtosecond titanium sapphire laser with a pulse duration of 100 fs and a pulse-repetition rate of 100 MHz (fundamental frequency with the wavelength $\lambda = 872$ nm and the spectral half-width $\Delta\lambda = 24$ nm, the second harmonic with $\lambda = 436$ nm and $\Delta\lambda = 6$ nm); and (ii) a nanosecond copper vapor laser with a pulse duration of 15 ns and a pulse-repetition rate of 10 kHz (wavelengths $\lambda_1 = 510.5$ nm and $\lambda_2 = 578.2$ nm, spectral half-width ~0.2 cm^{-1}). The average power of the titanium sapphire laser was 0.5 W at the fundamental frequency and reached 30 mW at the second harmonic frequency. The average power of the copper vapor laser was as high as 1.5 W. The average powers were measured with an IMO-2 calorimetric power meter. The duration of nanosecond pulses was measured using a FEK-16 coaxial photocell (resolution time, 0.5 ns) and an S7-13 stroboscopic oscilloscope with an effective bandwidth up to 3 GHz.

The beam from the femtosecond titanium sapphire laser at both the fundamental and second harmonic frequencies was divergent (with a divergence of $\sim 1 \times 10^{-3}$). In this case, the diameter of the laser beam was 2 mm on the cell and ~8 mm on the observation screen. The diameter of the beam from the copper vapor laser was 2 cm. Consequently, we used a focusing optics (i.e., a lens with a diameter of 2 cm and a focal length of 45 cm). The copper vapor laser had an unstable cavity with the magnification $M = 100$. Therefore, the beam quality approached the diffraction limit and the beam divergence was approximately equal to $\approx 10^{-4}$. The laser beams exhibited a Gaussian cross-sectional intensity distribution. Correspondingly, the diameter of the laser beams on the cell with the use of the focusing lens was estimated from standard relationships for Gaussian beams.

The dependence of the induced nonlinear optical response of the medium with quantum dots on the average power of the laser beams was investigated using their chopping by shutters with attenuation factors of 1 : 3, 1 : 10, and 1 : 100. This chopping made it possible to attenuate only the average power with no change in the peak power.

The linear absorption spectra of CdSe/ZnS quantum dots in the toluene solution were measured on a computerized Specord 40M spectrophotometer with the use of two cells with different thicknesses of 1 and 5 mm. The spectral dependences of the absorption coefficient of the colloidal solution for quantum dots with diameters of 2.6 and 1.9 nm at concentrations of ~10^{14} cm^{-3} are plotted in Fig. 1. Vertical lines in this figure indicate the spectral positions of the laser radiation lines used in the experiments on the transmission of the laser beams through the quartz cells. Figure 1 also shows the spectral dependence of the refractive index of the medium according to the results obtained from the measurements of the transmission of the light beam through the empty cell and the cells with toluene and a colloidal solution of quantum dots. The refractive index of the medium was calculated from the Fresnel formulas for the normal incidence of light with the use of the experimental data obtained for the absorption coefficient of the solution under investigation.

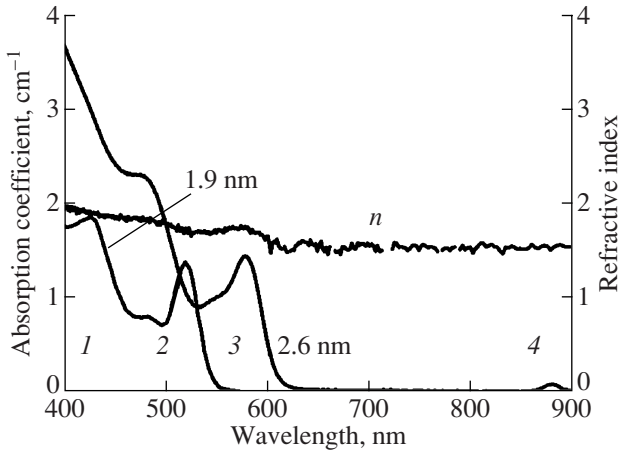

Fig. 1. Linear absorption spectra of the colloidal solutions of CdSe/ZnS quantum dots (2.6 and 1.9 nm in diameter) in toluene. Numerals indicate the laser spectral lines at wavelengths of (*1*) 436, (*2*) 510, (*3*) 578, and (*4*) 872 nm.

RESULTS

Figure 2 shows the photographs and the intensity distributions for the light beams (power, 30 mW; λ = 436 nm) transmitted through the cells with colloidal solutions containing quantum dots with diameters of 2.6 and 1.9 nm. It can be seen from Fig. 2 that, at the same power of the laser beam, the interference pattern of the solution with quantum dots 2.6 nm in diameter has more contrast and contains a larger number of rings than the interference pattern of the solution with quantum dots 1.9 nm in diameter. Similar interference patterns were obtained upon illumination of the cell with radiation from the nanosecond copper vapor laser. It should be noted that the peak powers of the femtosecond and nanosecond pulses were close to each other and equal to approximately 10 kW. The energy of the nanosecond light pulses was five orders of magnitude higher than that of the femtosecond light pulses, and the average power of the former pulses exceeded the average power of the latter pulses by a factor of 20. Under these conditions, the divergence of the light beams transmitted through the cell with quantum dots 2.6 nm in diameter increased several tens of times and the transverse intensity distribution exhibited more than 30 clear interference rings. No interference pattern was observed upon illumination of the cell with toluene (without quantum dots).

It was revealed that, after the onset of illumination of the cell, the stationary interference pattern is attained not instantaneously but for a subsecond time in both cases of femtosecond and nanosecond excitations. In order to estimate the time required to attain the stationary interference pattern, we filmed the process at a frame rate of 15 frames per second, which corresponds to a frame period of 66 ms. The frame-by-frame analysis of the film has demonstrated that, within the first 66 ms after switching on the laser beam (λ = 510 nm; average power, 0.6 W), the interference pattern in the beam is absent and the divergence of the transmitted beam is already close to the divergence of the beam with the formed stationary interference pattern. In the

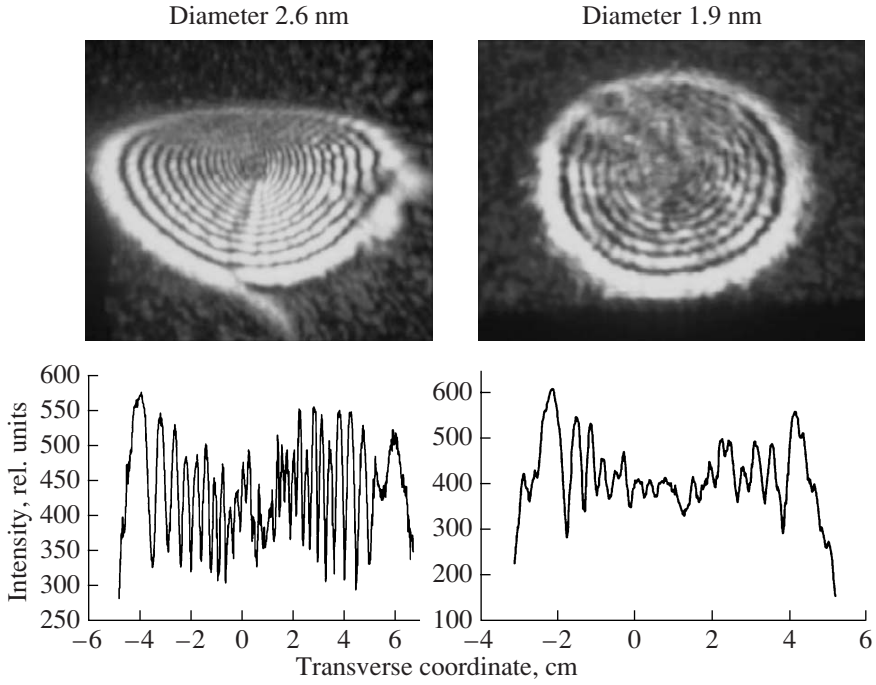

Fig. 2. Images of the light beams transmitted through the cells with colloidal solutions containing quantum dots with diameters of 2.6 and 1.9 nm upon illumination with laser radiation at the wavelength λ = 436 nm.

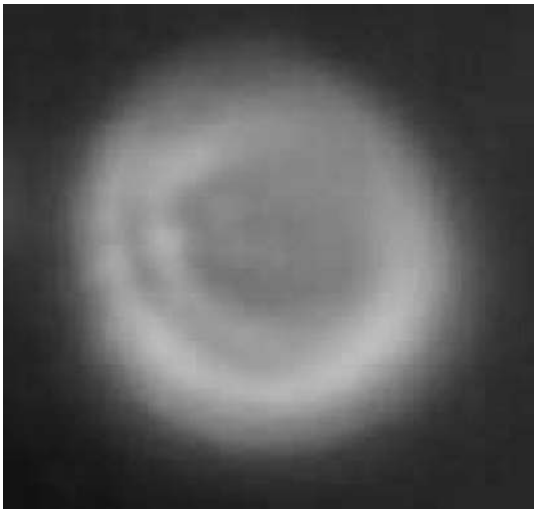

Fig. 3. Photograph of the spot of the light beam with the wavelength λ = 872 nm after its transmission through the cell containing a toluene solution of quantum dots with a diameter of 2.6 nm.

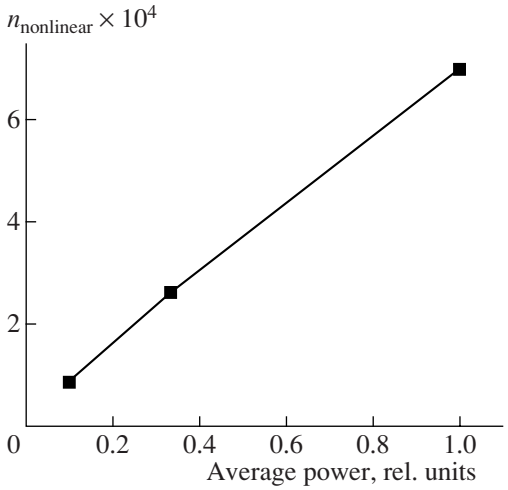

Fig. 4. Dependence of the nonlinear refractive index $n_{\text{nonlinear}}$ on the average power of the laser beam.

subsequent 66 ms (the time elapsed from the onset of illumination is t = 132 ms), characteristic interference rings arise at the periphery of the beam and their shape remains nearly unchanged in the subsequent time intervals. The interference pattern appears to be almost completely formed over the entire screen by the time of the fifth frame (i.e., after 264 ms). Subsequently, the rings change only slightly from the circular to elliptic shape with no change in their total number. The transformation is complete in a time interval of ~1 s.

The changes observed in the intensity distribution of the laser beam with a photon energy less than the band gap after its transmission through the cell with a toluene solution of quantum dots were measured with the use of radiation from the titanium sapphire laser at the fundamental frequency. A photograph illustrating the intensity distribution in the light beam (λ = 872 nm) with an average power of 0.5 W is displayed in Fig. 3. In this case, also, there arises a characteristic interference pattern, which, however, contains a considerably smaller number of rings. The negative sign of the light-induced nonlinearity was specially determined by displacing the cell along the focused laser beam (an analog of the *Z*-scan method). It turned out that, in all cases, the cell located in the convergent beam (before the focal waist) focuses the beam and the beam defocusing occurs in the divergent beam (after the focal waist). It is this behavior that is associated with the negative sign of the induced nonlinearity.

It can be seen from these figures that, after the light beam is transmitted through the quartz cell with a solution of quantum dots, it transforms into a set of concentric interference rings with characteristic features, namely, the first broad bright ring and subsequent bright rings with a decreasing period and a decreasing width. The number of rings depends on the excitation conditions. This value was used to determine the light-induced nonlinear refractive index n_2 of the medium under investigation.

As was noted above, the influence of the average power of the laser beam on the induced nonlinearity was investigated in the pulse-train illumination mode (a sequence of pulse trains with a frequency of 80 Hz and a duration dependent on the attenuation factor of the average power). The experiments were carried out under the following conditions: the pulse-repetition rate was 80 Hz, and the pulse-train duration upon attenuation of the average power was 1.25 ms for an attenuation factor of 1 : 10 and 4.13 ms for an attenuation factor of 1 : 3. The mode without chopping corresponds to a pulse-train duration of 12.5 ms at a pulse-repetition rate of 80 Hz. The results of the measurements performed under these conditions are as follows: eight rings were observed without attenuating the average power, three rings were revealed at an attenuation factor of 1 : 3, and one ring was observed at an attenuation factor of 1 : 10. In the case where the medium with the length l = 0.5 cm is excited by radiation with the wavelength λ_0 = 436 nm, from relationship (1), we obtain $n_{\text{nonlinear}}(E_0^2) = 7.0 \times 10^{-4}$, 2.6×10^{-4}, and 8.7×10^{-5} for the three aforementioned attenuation factors, respectively.

It follows from analyzing the obtained results that the induced nonlinear contribution to the refractive index due to the ring structure of the light beam is proportional to the pulse-train duration and, hence, depends linearly on the average power of the laser beam in accordance with the relationship $n = n_0 + |n_2|I$, where I is the average power of the laser beam. This relationship is illustrated by the dependence of the nonlinear contribution $n_{\text{nonlinear}}$ to the refractive index of the medium on the average beam power (Fig. 4).

DISCUSSION OF THE RESULTS

A similar structure with a characteristic increase in the width of the rings toward the periphery of the laser beam had been repeatedly observed earlier upon illumination of different media with a radiation beam from a continuous-wave argon laser. In particular, similar structures were observed in an alcohol solution of fuchsin [14] and nitrobenzene [15], as well as in experiments on the reorientation of liquid crystals in a light wave field [16, 17]. Interference patterns of this type are usually referred to as aberration rings. The origin of these rings was explained, for example, in [18]. The total number of bright rings depends only on the phase incursion at the center of the wave beam:

$$N = \frac{|\Phi_{\text{nonlinear}}(0)|}{2\pi} = \frac{|n_{\text{nonlinear}}(E_0^2)|l}{\lambda}, \quad (1)$$

where N is the number of rings, $n_{\text{nonlinear}}(E_0^2)$ is the nonlinear contribution to the refractive index, λ is the wavelength, and l is the length of the medium. Therefore, knowing the specified values of the thickness of the nonlinear medium, the radiation wavelength, and the number of rings, we can calculate the nonlinear change in the refractive index according to the procedure described in [19]. It is within this approach that the nonlinear contribution $n_{\text{nonlinear}}(E_0^2)$ to the refractive index was calculated in the present study. The accuracy in the determination of the nonlinear contribution depends on the number of interference rings. Since the number of rings can be easily determined accurate to within one-half the band, the error in the determination of the nonlinear contribution to the refractive index is equal to 4% when the number of rings is 13.

The maximum nonlinearity at the wavelength λ = 436 nm, which corresponds to 13 rings, is observed for an average power of 30 mW. Hence, according to relationship (1), we have $|n_{\text{nonlinear}}(E_0^2)| = 1.13 \times 10^{-3}$. This value corresponds to the nonlinear refractive index coefficient $|n_2| = 1.18 \times 10^{-3}$ cm^2/W $= 1.18 \times 10^{-10}$ CGS. The interference pattern observed at the fundamental frequency (λ = 872 nm) for an average power of 0.5 W contains three rings, which, in turn, corresponds to the nonlinear refractive index coefficient $|n_2| = 8.21 \times 10^{-14}$ CGS. The quantities $|n_2|$ determined under different excitation conditions are presented in the table.

As follows from the results obtained in this study, the time required to attain a stationary interference pattern due to the light-induced nonlinearity of the medium is approximately equal to ~1 s. According to [14–16], these long times are characteristic of thermal processes.

It is known that a stationary temperature profile and, consequently, a stationary thermal lens have a finite transient period associated with the heat diffusion. According to [20], the time required to attain a stationary temperature distribution (i.e., the transient period) in a region of radius r can be determined from the relationship

$$\tau_{\text{tr}} = \frac{\pi r^2 \rho c_p}{\xi}. \quad (2)$$

The time required to attain a stationary temperature profile in the studied medium with quantum dots in the toluene solution was estimated with the use of the following parameters: molecular weight of toluene, 92.14 g/mol; density ρ = 0.8669 g/cm^3; heat capacity c_p = 0.1049 kJ/(mol K); and thermal conductivity $\xi = 132.5 \times 10^{-3}$ W m^{-1} K^{-1} [21]. The calculation from expression (2) for the illuminated region of the medium with the radius r = 0.1 cm leads to the transient periods τ_{tr} = 23 s upon femtosecond excitation at the wavelength λ = 436 nm and τ_{tr} = 14 ms upon nanosecond excitation with subsequent illumination by focused radiation from the copper vapor laser (λ = 510 nm).

According to experimental estimates, the time required to attain the stationary profile of the intensity of the laser beam transmitted through the cell upon illumination by radiation from the copper vapor laser (λ = 510 nm) was determined to be 0.9 s. This value considerably exceeds the corresponding transient period (τ_{tr} = 14 ms) estimated from relationship (2). In turn, upon excitation with the femtosecond laser, the time required to attain the stationary interference pattern in the laser beam transmitted through the cell was found to be 1.1 s, which is substantially shorter than the time τ_{tr} = 23 s estimated from relationship (2).

It is known [18] that the induced thermal nonlinearity is proportional to the absorption coefficient and the power density of the laser beam. A comparison of the first and third rows in the table demonstrates that, upon changing over from the femtosecond excitation at the wavelength λ = 436 nm to the nanosecond excitation at λ = 510 nm, an increase in the power density of the laser beam by four orders of magnitude leads to a decrease in the nonlinear refractive index coefficient $|n_2|$ by four orders of magnitude, whereas the absorption coefficients of the medium for these regimes differ by a factor of only 1.9 (Fig. 1). This indicates that the light-induced nonlinearity under investigation exhibits a nonthermal nature.

It follows from a comparison of the first and third rows in the table that, upon changing over from the femtosecond excitation (λ = 436 nm) to the nanosecond

Table

λ, nm	P_{av}, W	I, W/cm^2	$\lvert n_{\text{nonlinear}}(E_0^2)\rvert$, 10^{-3}	$\lvert n_2\rvert$, CGS
436	0.03	1.0	1.13	1.18×10^{-10}
872	0.5	64	0.05	8.21×10^{-14}
510	1.0	2.5×10^4	3.57	1.4×10^{-14}

excitation ($\lambda = 510$ nm), an increase in the power density of the laser beam by four orders of magnitude brings about a decrease in the nonlinear refractive index coefficient $|n_2|$ by four orders of magnitude with a decrease in the absorption coefficient of the medium by a factor of only 1.9 (Fig. 2). This fact cannot be explained in terms of the mechanism providing the appearance of the light-induced thermal nonlinearity.

Let us now discuss other possible processes that can be responsible for the appearance of long-lived nonlinearities. The well-known mechanism providing the formation of the negative nonlinear contribution to the refractive index is associated with the filling of states in the conduction band. This process leads to saturation of the absorption of the medium and contributes to negative dynamic nonlinearity of a system with quantum dots. However, this mechanism of the generation of the nonlinearity has failed under our conditions, because its characteristic relaxation times are governed by radiative recombination of charge carriers and lie in the nanosecond range [22, 23]. These times are significantly shorter than the characteristic relaxation time (200 ms) of the nonlinearity observed in the present work.

In recent years, the blinking of single quantum dots (including CdSe/ZnS quantum dots) has been investigated under conditions of continuous excitation [24–26]. It has been established that the most probable time interval of blinking ("on–off") lies in the subsecond range (several hundred milliseconds). This corresponds to the characteristic time required to attain the stationary light-induced nonlinearity in our experiments.

One of the possible explanations for the blinking of single quantum dots upon their photoexcitation was offered in [27, 28]. The luminescence is quenched when one of the charge carriers (an electron or a hole) is trapped by the matrix surrounding the quantum dots. This mechanism of luminescence quenching is confirmed by some experimental observations. It was revealed that the luminescence exhibits a qualitatively identical behavior upon addition of quenchers, both electrons and holes [29], which makes these charged quantum dots nonluminescent ("dark"). A direct charge transfer (photoionization) from a CdSe quantum dot to the environment was observed in [30] with the use of an electrostatic-force microscope (EFM). The EFM measurements of the potential relief upon photoexcitation with photon energies greater than the band gap directly proved the photoionization of quantum dots. According to the nonlinear Kramers–Kronig relation [6] connecting the real part (refractive index) and the imaginary part (absorption index) of the complex permittivity

$$\Delta n(\omega, \Delta N) = \frac{c}{\pi} V.P. \int_0^\infty \frac{\Delta\alpha(\omega', \Delta N)}{\omega^2 - \omega'^2} d\omega'$$

(where *V.P.* stands for the principal-value integral and ΔN is the density of charge carriers escaped from quantum dots as a function of the excitation conditions), a decrease in the number of oscillators leads to a decrease in the refractive index of the medium, i.e., to a negative light-induced nonlinear contribution. This induced decrease in the refractive index manifests itself in the form of a giant long-lived nonlinearity revealed in our study.

It should be noted that the excitation of quantum dots in the yellow-green spectral range with a sequence of nanosecond pulses leads to a decrease in the nonlinearity by four orders of magnitude. In our opinion, the observed decrease is associated with the fact that, despite the considerably higher level of photoexcitation of quantum dots, this regime, in contrast to the femtosecond regime, does not bring about a quasi-continuous excitation. It is evident that the effective charge transfer to trap states with relaxation times on the order of several seconds can be provided by long-lived photoexcited carriers, which at a pulse-repetition rate of 10 kHz are almost absent.

The use of femtosecond laser radiation at the fundamental frequency ($\lambda = 872$ nm) can provide only intraband excitation of quantum dots. However, it follows from the experimental data that the nonlinearity photoinduced in this case turns out to be higher than the nonlinearity induced under excitation in the yellow-green spectral range. The reason is that the induced nonlinear optical response is determined by the two-photon excitation at the wavelength $\lambda = 872$ nm, which is equivalent to the photoexcitation in the blue spectral range. The two-photon excitation at wavelengths of 510 and 578 nm falls in the spectral range in which the absorption in toluene is very high.

It should also be noted that the nonlinearity revealed and investigated in our work leads to the formation of a negative nonlinear lens. The application of negative nonlinear lenses was discussed in detail in [31]. It is predominantly based on the use of thermal nonlinear lenses with transient periods on the order of several seconds. In particular, the use of these lenses in devices intended for optical measurements made it possible to decrease the threshold of the measurable absorption coefficients to $\sim 10^{-6}$ cm^{-1}. The "slow" nonlinearity examined in our work is not related to the thermal heating but is determined by the charge transfer from quantum dots to their environment. Therefore, this nonlinearity can be used in systems of optical measurements, which are intended, for example, for determining the transfer rate of charge carriers (photoionization) in the synthesis of semiconductor nanocrystals under different conditions. This provides a means for using nonlinear optical media with cadmium chalcogenide quantum dots in nanotechnological devices based on efficient control over the profile of the refractive wavefront and the parameters of light beams transmitting through this medium.

CONCLUSIONS

(1) The systems of CdSe/ZnS quantum dots (2.6 and 1.9 nm in size) were examined upon excitation with sequences of femtosecond and nanosecond laser pulses.

(2) It was found that, at all the radiation wavelengths under investigation, the systems of CdSe/ZnS quantum dots are characterized by a giant photoinduced nonlinearity, which manifests itself in the form of an interference pattern. The number of rings in the interference pattern was used to measure the coefficient $|n_2|$ determining the nonlinear contribution to the refractive index of the medium.

(3) It was established that the induced nonlinearity of the toluene solution of CdSe/ZnS quantum dots depends linearly on the average power of laser beams. The time required to attain a stationary regime of the induced nonlinearity of the medium (i.e., the transient period) was determined to be in the millisecond range.

(4) From analyzing the induced nonlinearities measured in the medium under different excitation conditions and the estimates obtained for the times required to attain a stationary thermal lens, it was concluded that the observed nonlinearity exhibits a nonthermal nature.

(5) An analysis was made of the possible physical mechanisms of the processes occurring with the participation of the CdSe/ZnS quantum dots, which can lead to long-lived giant nonlinearities of the medium with millisecond relaxation times. It was assumed that the blinking effect of quantum dots in organic matrices and the light-induced nonlinearity observed in our experiments are common in nature.

(6) Possible applications of the obtained results for the use in nanophotonic and nonlinear optical devices based on the efficient control over the profile of the refractive wavefront and the parameters of the light beams were considered.

ACKNOWLEDGMENTS

This study was supported by the Russian Foundation for Basic Research (project nos. 06-02-16399, 06-02-17089, 07-02-00656, 08-02-01447a, 06-02-08120ofi, and 05-03-34821-MF), the Presidium of the Russian Academy of Sciences (program "Femtosecond Optics and Optical Materials"), the Branch of General Physics and Astronomy of the Russian Academy of Sciences (programs "Coherent Optical Radiation of Semiconductor Compounds and Structures" and "Optical Spectroscopy and Frequency Standards"), and the Council on Grants from the President of the Russian Federation for Support of Leading Scientific Schools of the Russian Federation (grant no. NSh-4365-2008.2) and performed within the framework of the State Contract (no. SU/07/564/NTB/k, FIAN).

REFERENCES

1. G. Moeller and S. Coe-Sullivan, "Quantum-Dots Light-Emitting Devices for Displays," Inf. Disp. **2**, 1–6 (2006).
2. M. Ohtsu, K. Kobayashi, T. Kawazoe, et al., "Nanophotonics: Design, Fabrication, and Operation of Nanometric Devices Using Optical Near Fields," IEEE J. Sel. Top. Quantum Electron. **8** (4), 839–862 (2002).
3. C. Burda, X. Chen, R. Narayanan, et al., "Chemistry and Properties of Nanocrystals of Different Shapes," Chem. Rev. **105**, 1025 (2005).
4. D. V. Talapin, A. L. Rogach, A. Kornowski, et al., Highly Luminescent Monodisperse CdSe and CdSe/ZnS Nanocrystals Synthesized in a Hexadecylamine–Trioctylphosphine Oxide–Trioctylphosphine Mixture," Nano Lett. **1** (4), 207–211 (2001).
5. M. Sheik-Bahae, A. A. Said, T. H. Wei, et al., "Sensitive Measurements of Optical Nonlinearities Using a Single Beam," IEEE J. Quantum Electron. **26**, 760 (1990).
6. M. Sheik-Bahae, D. J. Hagan, and E. W. van Stryland, "Dispersion and Band-Gap Scaling of the Electronic Kerr Effect in Solids Associated with Two-Photon Absorption," Phys. Rev. Lett. **65**, 96 (1990).
7. M. Sheik-Bahae, D. C. Hutchings, J. David, et al., "Dispersion of Bound Electronic Nonlinear Refraction in Solids," IEEE J. Quantum Electron. **27**, 1296 (1991).
8. E. J. Canto-Said, D. J. Hagan, J. Young, et al., "Degenerate Four-Wave Mixing Measurements of High Order Nonlinearities in Semiconductors," IEEE J. Quantum Electron. **27**, 2274 (1991).
9. V. I. Klimov, "Optical Nonlinearities and Ultrafast Carrier Dynamics in Semiconductor Nanocrystals," J. Phys. Chem. B **104**, 6112 (2000).
10. V. S. Dneprovskii, E. A. Zhukov, D. A. Kabanin, et al., "Nonlinear Absorption and Refraction of Light in a Colloidal Solution of CdSe/ZnS Quantum Dots upon Two-Photon Resonant Excitation," Fiz. Tverd. Tela (St. Petersburg) **49** (2), 352–356 (2007) [Phys. Solid State **49** (2), 366–370 (2007)].
11. A. J. Nozik, "Spectroscopy and Hot Electron Relaxation Dynamics in Semiconductor Quantum Wells and Quantum Dots," Annu. Rev. Phys. Chem. **52**, 193 (2003).
12. S. A. Crooker, T. Barrick, J. A. Hollingsworth, et. al., "Multiple Temperature Regimes of Radiative Decay in CdSe Nanocrystal Quantum Dots: Intrinsic Limits to the Dark-Exciton Lifetime," Appl. Phys. Lett. **82**, 2793 (2003).
13. S. N. Sharma, Z. S. Pillai, and P. V. Kamat, "Photoinduced Charge Transfer between CdSe Quantum Dots and *p*-Phenylenediamine," J. Phys. Chem. B **107**, 10088 (2003).
14. S. A. Akhmanov, D. P. Krindach, A. P. Sukhorukov, and R. V. Khokhlov, "Nonlinear Defocusing of Laser Beams," Pis'ma Zh. Eksp. Teor. Fiz. **6** (2), 509 (1967) [JETP Lett. **6** (2), 38 (1967)].
15. J. R. Whinneru, D. T. Miller, and F. Dabby, "Thermal Convection and Spherical Aberration Distortion of Laser Beams in Low-Loss Liquids," IEEE J. Quantum Electron. **3**, 382 (1967).
16. A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, et al., "The Effect of an Optical Field on the Nematic Phase of the Liquid

Crystal OCBP," Pis'ma Zh. Eksp. Teor. Fiz. **32** (2), 170 (1980) [JETP Lett. **32** (2), 158 (1980)].

17. A. S. Zolot'ko, V. F. Kitaeva, N. Kroo, et al., "Light-Induced Fréedericksz Transition in an MBBA Crystal," Pis'ma Zh. Eksp. Teor. Fiz. **34** (5), 263 (1981) [JETP Lett. **34** (5), 250 (1981)].
18. A. P. Sukhorukov, "Diffraction of Light Beams in Nonlinear Media," Soros. Obraz. Zh., No. 5, 85–92 (1996).
19. M. I. Barnik, I. A. Budagovskii, V. A. Enikeeva, et al., "Orientation Effect of Femtosecond Light Pulses on Nematic Liquid Crystals," Preprint No. 10 FIAN (P. N. Lebedev Physical Institute, Russian Academy of Sciences, Moscow, 2003).
20. V. P. Isachenko and V. A. Osipova, and A. S. Sukomel, *Heat Transfer* (Energiya, Moscow, 1969; Mir, Moscow, 1969).
21. I. S. Grigoriev and E. Z. Meilikhov, *Handbook of Physical Quantities* (Energoatomizdat, Moscow, 1991; CRC Press, Boca-Raton, FL, United States, 1996).
22. G. Schlegel, J. Bohnenberger, I. Potapova, et al., "Fluorescence Decay Time of Single Semiconductor Nanocrystals," Phys. Rev. Lett. **88**, 137401 (2002).
23. V. Zwiller, H. Blom, P. Jonsson, et al., "Single Quantum Dots Emit Single Photons at a Time: Antibunching Experiments," Appl. Phys. Lett. **78**, 2476–2478 (2001).
24. R. Verberk, A. M. van Oijen, and M. Orrit, "Simple Model for the Power-Law Blinking of Single Semiconductor Nanocrystals," Phys. Rev. B: Condens. Matter **66**, 233202 (2002).
25. K. T. Shimizu, R. G. Neuhauser, C. A. Leatherdale, et al., "Blinking Statistics in Single Semiconductor Nanocrystal Quantum Dots," Phys. Rev. B: Condens. Matter **63**, 205316 (2001).
26. R. J. Cook and H. J. Kimble, "Possibility of Direct Observation of Quantum Jumps," Phys. Rev. Lett. **54**, 1023 (1985).
27. L. Efros and M. Rosen, "Random Telegraph Signal in the Photoluminescence Intensity of a Single Quantum Dot," Phys. Rev. Lett. **78**, 1110 (1997).
28. C. Burda, S. Link, M. Mohamed, et al., "The Relaxation Pathways of CdSe Nanoparticles Monitored with Femtosecond Time-Resolution from the Visible to the IR: Assignment of the Transient Features by Carrier Quenching," J. Phys. Chem. **105**, 12286 (2001).
29. M. Shim, C. Wang, and P. Guyot-Sionnest, "Charge-Tunable Optical Properties in Colloidal Semiconductor Nanocrystals, J. Phys. Chem. B **105**, 2369 (2001).
30. T. D. Krauss, S. O'Brien, and L. E. Brus, "Charge and Photoionization Properties of Single Semiconductor Nanocrystals," J. Phys. Chem. B **105**, 1725 (2001).
31. G. B. Al'tshuler and M. V. Inochkin, "Nonlinear Lenses and Their Applications," Usp. Fiz. Nauk **163** (7), 65 (1993) [Phys.—Usp. **36** (7), 604 (1993)].