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Wavelength-controlled manipulation of colloidal quasi-resonant quantum dots under pulsed laser irradiation

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ABSTRACT

Production of nanostructures consisting of semiconductor nanoparticles (NPs) is of interest for number of applications. Development of new methods of NPs' manipulation and aggregation of NPs into nanostructures with pre-defined geometry is also of considerable interest from the fundamental point of view. Under laser irradiation with properly chosen wavelengths excitonic excitations of semiconductor NPs will be induced. Electrodynamical interaction between excited NPs is rather universal and allows formation of wide variety of nanostructures both of homo- and heterogeneous content. Theoretical approach for study of interaction of NPs' ensembles with laser light includes dipole-dipole approximation for NPs' attraction. Experimental results are obtained for TGA stabilized CdTe QDs with the excitonic resonance at 520 nm. Six different samples of the same colloid solution were irradiated at wavelengths from 540 to 570 nm. Modifications of absorption spectra of solutions after irradiation was detected, being most prominent at 555 and 560 nm irradiation wavelengths. Analysis of spectra shows that up to 47% of QDs were assembled into pairs with 10 nm inter-QD distance. Therefore, possibility of precise QDs manipulation via laser-induced electro-dynamical interaction is demonstrated.

Keywords: Self-assembly, nanostructures, quantum dots, absorption spectra, dipole-dipole interaction, laser control

1. INTRODUCTION

Controlled formation of nanostructures composed of nanoparticles of the same chemical content or different ones may lead to creation of new materials and nanosized objects with unique physical properties and, consequently, new functionality for wide range of applications[1-4]. Methods of control of the shape and size of forming nanostructures investigated up to date include approaches using various lithographic techniques[5,6], and various processes of self-organization in ensembles of nanoparticles[1,2, 7-11]. In the latter case, the shape of final nanostructures is controlled by selective chemical interactions, temperature, using templates on the surface, or static electric and magnetic fields [2]. Employment of illumination by ambient or laser light is found to be a factor of structures' control in some cases, mainly due to thermal or chemical effects produced under this illumination [12]. Special case is the use of molecules experiencing isomeric transition under the illumination as the stabilizers of nanoparticles; that isomeric transition leads to formation of colloid crystals or to considerable displacements of nanoparticles [13]. In all cases mentioned above, the light plays the initiating role in the self-assembly process. Oppositely, at certain level of intensity noticeable mechanical effects of light are known like radiation pressure, or gradient force used in optical tweezers, and in these cases the light is the main source of particles' motion [14, 15]. Another similar example is the electro-dynamical interaction of polarization induced on nanoparticles under external laser illumination; this effect was studied in a number of theoretical papers, and several experiments are done with metal nanoparticles [16-18]. To produce self-assembly of silver nanoparticles, cw laser radiation detuned from surface plasmon frequency by several plasmon resonance widths was used to induce localized plasmons on them, and their self-assembly is explained by strong attractive force acting in the gap between particles [18]. Plasmon resonances of metal nanoparticles have rather large width (FWHM = 90 nm) [16, 18], and this leads to rather low depths of potential wells associated with nanoparticles' attraction [16]. Employing narrower exciton resonances in semiconductor quantum dots (QDs) [19] is attractive since potential wells are predicted to become deeper at the same level of laser light intensity. Possibilities to create complex nanostructures comprising both metal and semiconductor nanoparticles, as well as control of the shape of complex multiparticle's objects are also theoretically shown [20].

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In the present paper we summarize the theoretical results obtained in dipole-dipole approximation, on resonant light-controlled effects on formation of nanostructures that contain semiconductor nanoparticles and results of the experiments on the forming the pairs of colloidal quantum.

2. CALCULATION MODEL

To create stable structure, nanoparticles in a colloid solution must approach each other to the distance where van der Waals energy will exceed the energy of thermal motion. If no means to ensure stability of individual nanoparticles are employed, than fast spontaneous, and therefore, uncontrolled aggregation will occur. Structures formed via spontaneous aggregation are fractal ones with random structure. To prevent spontaneous aggregation, stabilizing potential barrier must be formed between nanoparticles with the height well above thermal motion energy. Light-controlled aggregation must assume addition of electrodynamic potential to van der Waals potential and stabilizing potential so that cumulative potential would enable nanoparticles of desired kind to selectively override stabilizing potential and to form the structure predefined by experimenter [21].

When metal or semiconductor nanoparticles in the state of Brownian motion interact with laser field, then electric dipole polarization is induced on them. Interaction of this polarization with laser field results in optical trapping in the regions with the dimensions of order of wavelength. Increased density of nanoparticles results in increased aggregation rate within an optical trap. Aggregates produced are commonly disordered on the nanoscale, but can bear the signs of ordering on the sub-wavelength scale due to a variety of processes depending on the medium containing nanoparicles [9, 22]. When several particles in the course of Brownian motion occasionally appear closely enough to each other, then their polarizations interact each other. This is one of manifestations of near-field effects. If energy of electromagnetic interaction between nanoparticles exceeds the energy of thermal motion kT and the energy of a barrier preventing the aggregation then irreversible formation of nanostructures will happen.

Let us consider an ensemble of N nanoparticles with coordinates r_i and interacting each other through their polarization induced by laser field. The dimensions of an ensemble are supposed to be much less than the wavelength. Then external field strength can be considered homogeneous within the ensemble. Then electric field strength is considered to depend only on time: $\mathbf{E} = (1/2)\mathbf{E}_0 \exp(i\omega t) + c.c.$ Local field acting on every nanoparticle is a sum of external field and the fields induced by all the rest particles. In the dipole-dipole interaction approximation, the dipole moment vector at the frequency ω induced on i -th particle is:

$$\mathbf{d}_i = \chi_0 \left(\mathbf{E} + \sum_{j \neq i} \mathbf{E}_j \right) \quad (1)$$

Here $\chi_0 = d_{12}^2 / \hbar(\Omega + i\Gamma_i)$ is linear polarizability of isolated nanoparticle, $|d_{12}|^2$ is the squared module of electric dipole moment of the transition between ground (1) and excited (2) states for i -th particle, $\Omega = \omega_{oi} - \omega$ is the resonance detuning, \hbar is the Planck constant, Γ_i is the homogeneous width of the transition, \mathbf{E}_j is the field produced by j -th nanoparticle in the position of i -th particle [23]:

$$\mathbf{E}_j = \frac{3(\mathbf{d}_j \mathbf{r}_{ij}) \mathbf{r}_{ij} - \mathbf{d}_j r_{ij}^2}{r_{ij}^5} \quad (2)$$

where $r_{ij} = r_i - r_j$ is the distance between particles. Interaction energy of i -th and j -th particles with excited dipole polarization will be [24]:

$$W_{dij} = -\mathbf{d}_i \cdot \frac{3(\mathbf{d}_j \mathbf{r}_{ij}) \mathbf{r}_{ij} - \mathbf{d}_j r_{ij}^2}{r_{ij}^5} = \frac{(\mathbf{d}_i \mathbf{d}_j) r_{ij}^2 - 3(\mathbf{d}_i \mathbf{r}_{ij})(\mathbf{d}_j \mathbf{r}_{ij})}{r_{ij}^5} \quad (3)$$

Calculation of interaction energy between particles, therefore, requires solution of the system of N equations with dipole moments of every particle defined by expression (1). Also, renormalised linear polarizability of every particle $\hat{\chi}_i$ can be obtained, that is in general case a tensor connecting dipole moment vector induced on i -th particle with the electric field

strength vector: $\mathbf{d}_i = \hat{\chi}_i \mathbf{E}$. Imaginary part of renormalized polarizability determines the absorption of light by i -th particle. Consider the binary interaction of two particles in a dipole approximation. As an example let us choose a semiconductor nanoparticle of CdSe [19, 25]; optical properties of these nanoparticles are well studied.

Using (1) and introducing the frequency shift of i -th particle resonance due to interaction with neighbor particle $\omega_{si} = |d_{i2}|^2 / \hbar r^3$, we obtain expressions for d_{ix} ($x \parallel r_{12}$) and d_{iy} ($y \perp r_{12}$) components of induced dipole moments of a pair of nanoparticles:

$$\begin{aligned}
 d_{1x} &= r^3 E \omega_{s1} \cos \alpha \left[\frac{(\Omega_2 + 2\omega_{s2})(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2}) + \Gamma_2 (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} + \right. \\
 &\quad \left. + i \frac{\Gamma_2 (\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2}) - (\Omega_2 + 2\omega_{s2})(\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} \right] e^{i\omega t} + c.c. \\
 d_{1y} &= r^3 E \omega_{s1} \sin \alpha \left[\frac{(\Omega_2 - \omega_{s2})(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2}) + \Gamma_2 (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} + \right. \\
 &\quad \left. + i \frac{\Gamma_2 (\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2}) - (\Omega_2 - \omega_{s2})(\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} \right] e^{i\omega t} + c.c. \\
 d_{2x} &= r^3 E \omega_{s2} \cos \alpha \left[\frac{(\Omega_1 + 2\omega_{s1})(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2}) + \Gamma_1 (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} + \right. \\
 &\quad \left. + i \frac{\Gamma_1 (\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2}) - (\Omega_1 + 2\omega_{s1})(\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - 4\omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} \right] e^{i\omega t} + c.c. \\
 d_{2y} &= r^3 E \omega_{s2} \sin \alpha \left[\frac{(\Omega_1 - \omega_{s1})(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2}) + \Gamma_1 (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} + \right. \\
 &\quad \left. + i \frac{\Gamma_1 (\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2}) - (\Omega_1 - \omega_{s1})(\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)}{(\Omega_1 \Omega_2 - \Gamma_1 \Gamma_2 - \omega_{s1} \omega_{s2})^2 + (\Omega_1 \Gamma_2 + \Omega_2 \Gamma_1)^2} \right] e^{i\omega t} + c.c. \tag{4}
 \end{aligned}$$

Here α is the angle between the line connecting the centers of the nanoparticles and the vector \mathbf{E} (Fig.1).

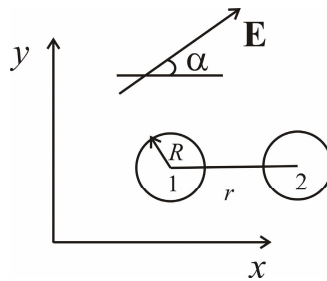


Figure 1. Scheme of mutual orientation of the particles and the polarization of laser field.

It is suitable to present the expression for the energy of dipole-dipole interaction through the amplitudes and phases of induced dipole moments of every particle. Then, after averaging on time, expression (3) will be:

$$W = \frac{|d_{1y}| |d_{2y}| \cos(\phi_{1y} - \phi_{2y}) \sin^2 \alpha - 2 |d_{1x}| |d_{2x}| \cos(\phi_{1x} - \phi_{2x}) \cos^2 \alpha}{2r^3} \tag{5}$$

where $|d_{1y}|, |d_{2y}|, |d_{1x}|, |d_{2x}|$ are modules, and $\phi_{1y}, \phi_{2y}, \phi_{1x}, \phi_{2x}$ are phases of y and x components of time-averaged dipole moments of the first and the second particles.

Results of the calculation of the dipole-dipole interaction energy of a pair of quantum dots are presented in [26]. Narrow resonance enables achieving electrodynamic interaction energy of a pair of QDs up to 10^2 kT (T is the temperature of solution, and k is Boltzmann constant) at interparticle distance 10 nm and laser intensity 10^6 W/cm². This result of the calculations evidences the possibility of the implementation of self-assembly of a pair of colloidal quantum dots at the modest external field intensity.

3. EXPERIMENTAL RESULTS

To demonstrate the effect of electrodynamic interaction on the QDs self-assembly we compared the spectra of QDs solution before and after irradiation by the laser light. Solution of CdTe (average diameter 2.9 nm, purchased from PlasmaChem GmbH, Berlin, Germany) particles in water was used as the object. QDs were stabilized against spontaneous aggregation with addition of thioglicole acid. Excitonic absorption band of CdTe QDs ensembles in solutions is usually 30 nm wide; however, luminescence measurements show that individual QD linewidth is of order 3 nm [19]. Hence, observed absorption linewidth is due to inhomogeneous broadening caused by the scatter of QDs size. For our experiment, absorption and luminescence spectra of initially prepared QD solution are presented in Figure 2; experimental exciton peak is at 523 nm in absorption and at 551 nm in the luminescence. These spectra are similar to those typically observed for CdTe QDs [19].

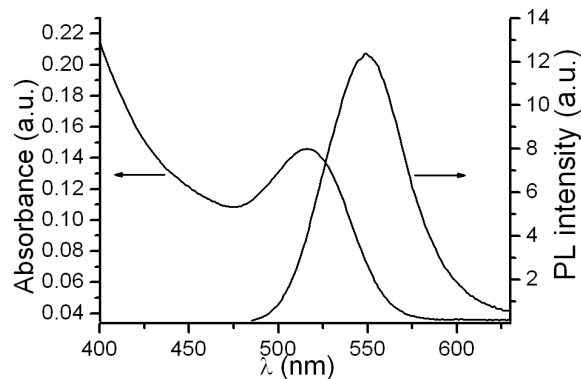


Figure 2. Optical density of 0.9 cm thick CdTe solution (Perkin-Elmer Lambda 35, left scale) and luminescence spectra excited at 480 nm (HoribaJobinIvon Fluorolog3, right scale).

Electrodynamic interaction of QDs was induced using Opotec Vibrant 355 optical parametric oscillator by the laser pulses of 10 ns duration with the peak power 1.5 MW at 10 Hz repetition rate. The peak intensity of laser radiation at the irradiated sample was $2 \cdot 10^7$ W/cm². Assuming refractive index of the solution $n = 1.33$, that corresponds to amplitude of electric field strength 10^7 V/m. Six different samples of the same solution were irradiated at the wavelengths 540, 550, 555, 560, 565 and 570 nm. Each sample was irradiated during 5 minutes by 3000 laser pulses. Most prominent and well-analyzable laser-induced persistent changes were obtained for irradiation wavelengths 550, 555, 560, and 565 nm. Spectra of irradiated solutions are presented in Figure 3.

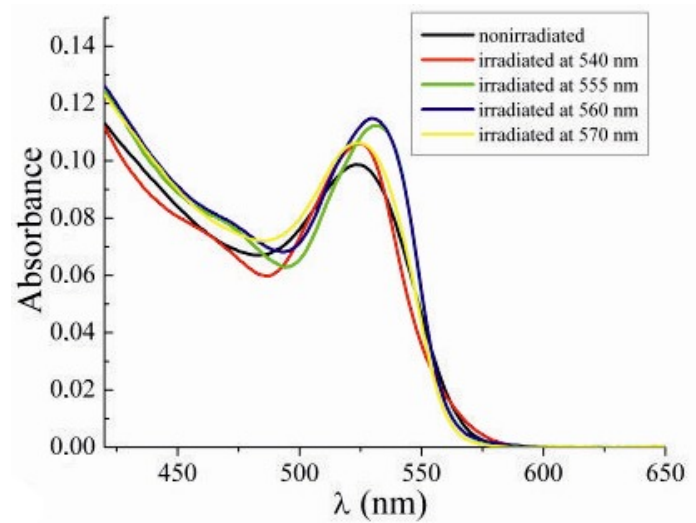


Figure 3. Selected absorption spectra of CdTe (TGA) solutions in water after intense laser irradiation at different laser wavelengths.

To make these variations more pronounced, difference spectra were obtained by the subtraction of the spectrum of unirradiated solution from the spectra of solutions irradiated at different laser wavelengths. These spectra are presented in Figure 4 by thick lines.

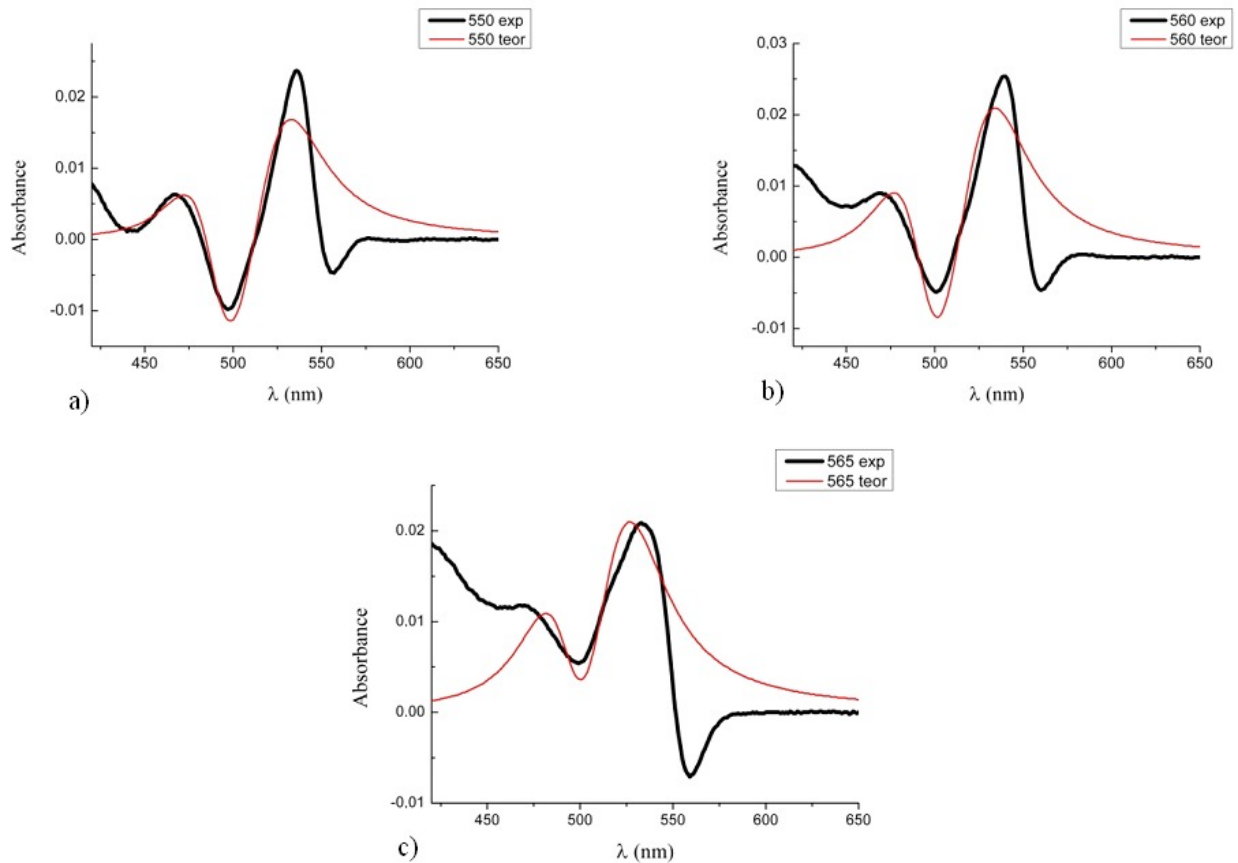


Figure 4. Difference spectra of CdTe QDs solutions after and before irradiation at the wavelengths 550 (a), 560 (b) and 565 (c) nm.

Difference spectra obtained by above-mentioned subtraction are composed by the contributions from the paired QDs spectra which are dependent on the inter-QD distance, and the spectra of the rest of isolated QD that are independent on that distance. The shape of difference spectra $F(p, r)$ can be, therefore presented by the following formula:

$$F(p, r) = p \operatorname{Im}(\chi(r)_{pair}) - 2p \operatorname{Im}(\chi_{iso}) \quad (6)$$

where $\chi(r)_{pair}$ is the linear susceptibility of a pair of QDs averaged over all orientations of the pair with respect to electric field strength vector, χ_{iso} is the linear susceptibility of isolated QD, p is the number of pairs formed after irradiation normalised to the number of isolated QDs in the unirradiated solution. Therefore, p lies in the range from 0 to 0.5. Least square procedure was used to fit theoretical shape $F(p, r)$ to the experimental difference spectra. Two parameters to be determined were used in the least square fit procedure: 1) the part of paired QDs (p) and 2) the mean inter-QD distance in pairs of QD (r). Difference spectra (thick lines) for irradiation at different wavelength and fitted theoretical spectra (thin lines) are presented in Figure 4. Theoretical spectra fairly describe two main maxima formed after the irradiation, and the main discrepancies in the vicinity of laser wavelength, as the consequence of persistent hole-burning due strong absorption and destroying of the resonant QD pairs, and at the short-wavelength edge of the spectra. The latter regions is formed by interband CdTe QDs' transition which was not included in the theoretical model. Corporate graph presenting experimental dependence of paired QDs percentage on the self-assembling laser wavelength is presented in Figure 5.

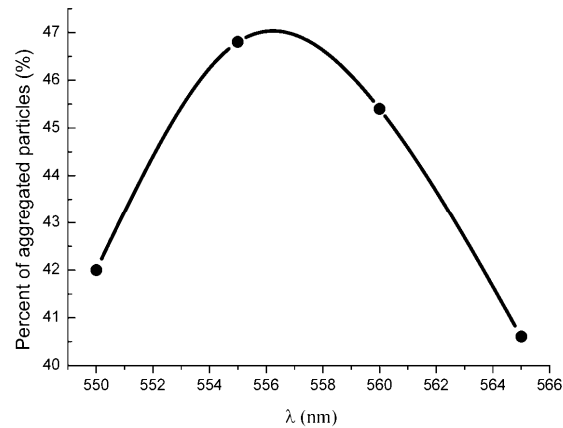


Figure 5. Percentage of CdTe QDs that undergone self-assembly under laser irradiation at the wavelengths in the range 550 - 565 nm. Points are the data extracted from experimental spectra of CdTe solutions after the laser irradiation, and solid line is the guide to the eye.

The spectra obtained after irradiation at 540 and 570 nm also contain noticeable features due to the formation of pairs. However, the amplitude of these features is much smaller than in case of irradiation at 550, 555, 560, and 565 nm. They are small in comparison with persistent hole dip and with the interband transition edge. As the result, least square fitting for the wavelengths 540 and 570 nm is much less convincing, and the percentage of paired QDs can be estimated with less certainty to be surely less than 10%. Therefore, efficient self-assembly is induced in the range of laser wavelengths 550 - 565 nm and rapidly drops for wavelengths outside this region.

4. CONCLUSION

In conclusion, we theoretically and experimentally consider the possibility of laser-controlled formation of nanostructures with required geometry containing semiconductor nanoparticles. The geometry of the structure is governed by frequency and polarization of laser radiation. Presented experimental results show that, under sufficiently intense pulsed laser irradiation with properly chosen wavelength, about a half of quasiresonant isolated QDs are self-assembled into pairs. For CdTe QDs with average diameter 2.9 nm, this effect is efficient in the laser wavelength range 550–565 nm only, and this wavelength range is redshifted from the resonant wavelength of most of isolated QDs as

predicted by the theory of electrodynamic interaction of polarizations induced on QDs. The technology under study allows precise manipulation of QDs not only in the solution, but also on technological substrates.

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