SELF-ORGANIZED AGGREGATION OF A TRIPLE OF COLLOIDAL QUANTUM DOTS INTO STABLE STRUCTURES WITH VARIOUS SHAPES CONTROLLED BY A LASER FIELD

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Abstract

Dynamical model of self-assembly of nanoparticles in the field of laser radiation is developed and applied to the investigation of the possibility of the assembly of variously shaped structures comprised of the particles. Specifically, the computer model of the process of formation of structures with pre-defined geometry from a set of three initially isolated nanoparticles. The possibility of control of the geometry of the formed structure via the choice of the wavelength of external field.

Keywords: Nanoparticles, nanostructure, self-assembly, quantum dots

1. INTRODUCTION

In recent decades, nanostructures with unique properties other than properties of a bulk sample and dependent on both composition and shape have been studying actively. Therefore, the problem of developing a universal method of nanostructures formation is a subject of interest of many scientists. The least expensive method of obtaining colloidal crystals, which doesn't require a local physical impact on the system, is based on the ability of nanoparticles to self-organize in the process of random Brownian collisions in real disperse systems [1-4]. However, in such case it is impossible to control the processes of formation of nanostructures with predetermined shapes. Therefore, one of the possible solutions of this problem is the physical impact on the ensembles of nanoparticles that allows the formation of complex nanostructures without additional surface modifications [5-8]. However, it is not always possible to achieve the selectivity of the self-assembly process.

Earlier in [9-11], it was shown that the interaction of resonant nanoparticles in the field of laser radiation makes possible the formation of predetermined structures, since the energy of the induced dipole-dipole interaction is sufficient to overcome the barrier that prevents spontaneous aggregation (which makes possible the formation a stable structure), and their geometry depends on the wavelength and the polarization of the external field. In this case, the formation of structures, which scale is much smaller than the wavelength of the laser radiation caused this polarization, becomes possible. The presence of optical resonances in the particle leads to an increase of the interparticle interaction and is the basis for the selective formation of different structures with a predetermined position of the particles in the aggregate. It becomes possible because the energy of the interparticle interaction in the laser radiation field depends on its frequency, the resonance frequencies of the particles, and on the orientation of their group relatively to the plane of polarization.

The experiments [10] on the formation of pairs of colloidal quantum dots in the field of laser radiation have shown the possibility of realizing this method. The further formation of more complex structures (three or more particles) can be realized in the way of a step-by-step process, when a third particle is added to an already formed pair of particles, via selecting the wavelength of the external field.

In this study, a dynamic self-assembly model for a triple of particles in a laser field using Brownian dynamics is proposed. The possibility of forming a three-particle structure with a predetermined geometry from triple of isolated particles is studied. The obtained results allows to make significant progress in the study of the method of structures self-assembly in the field of laser radiation, which can be use as universal method to form structures with specified properties that will find application as sensors, photodiode elements and solar cells.

2. RESULTS AND DISCUSSIONS

In the present study, we used the computational model described in [11]. Using this model, the possibility of formation the structures under the action of laser field via a two-stage process was earlier shown [11], when a third particle was adjoined to a previously formed pair of particles. Such approach, however, requires certain complications of the experimental procedures. Present study the possibility of formation of structures with pre-defined geometry in the course of self-assembly of three separated particles under the action of the field of short pulse laser radiation. The parameters of the medium and of the particles are analogous to those considered in [11]. The nanoparticle's parameters correspond to the parameters of CdTe quantum dots with average diameter 3 nm (the wavelength of the first exciton transition λ=525 nm, Δλ=3 nm, electric dipole moment of the transition $|d_{12}|^2 = 1.91 \cdot 10^{-44}$ J·m^3). The choices of the material and of the size of QDs are the factors that determine both the resonant wavelength of exciton transition and the stability of QDs under the action of laser radiation.

As the result of computer modeling, the dependences of the probabilities of structures' formation on the external laser radiation wavelength were obtained. As it was shown earlier in [9,10], the interaction of dipole polarizations induced on QD by laser field in the given structure leads to the shift of resonances of QDs in comparison to those of isolated particles, and this shift is dependent on the position of QDs with respect to the polarization plane of laser field. Therefore, interparticle interaction energy is determined by the laser wavelength, intensity and polarization, which leads to the spectral selectivity of the structure obtained. **Figure 1** depicts such dependence of maximum probability of linear structure formation. In general, it was found that at certain wavelengths choices the formation of three types of stable structures: "line" (**Figure 1**, θ=0º), "angle" (θ=90º) and "pyramid" (θ=120º).

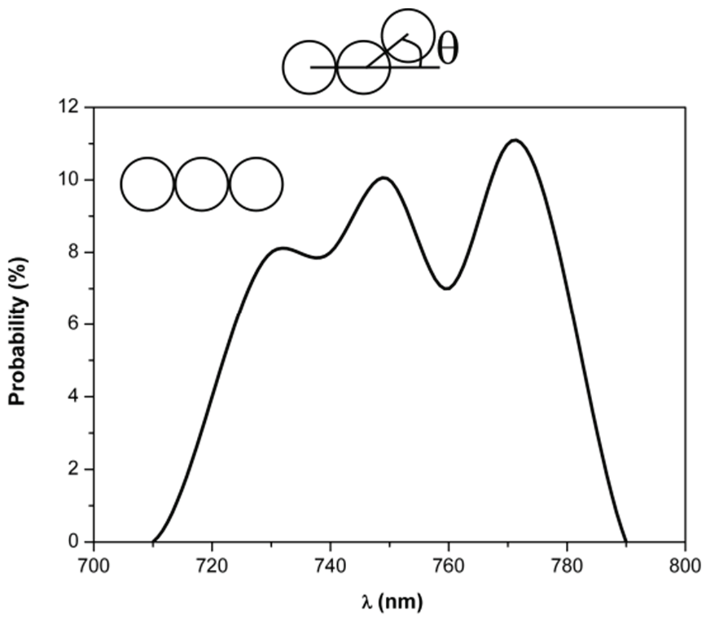

Figure 1 Dependence of the maximum of the probability of formation of the structure of "line" type on the external field wavelength

As one can see from **Figure 1**, the probability of "line" type structure formation reaches 12 % per a single pulse with 10 ns duration; in case when laser irradiation is performed in the long wavelength part of the spectrum, only line type structures are formed, evidencing high selectivity of self-assembly process. Irradiation in the short wavelengths part of the spectrum are shown to be favorable for the formation of other types of stable structures as can be seen in **Figure 2**.

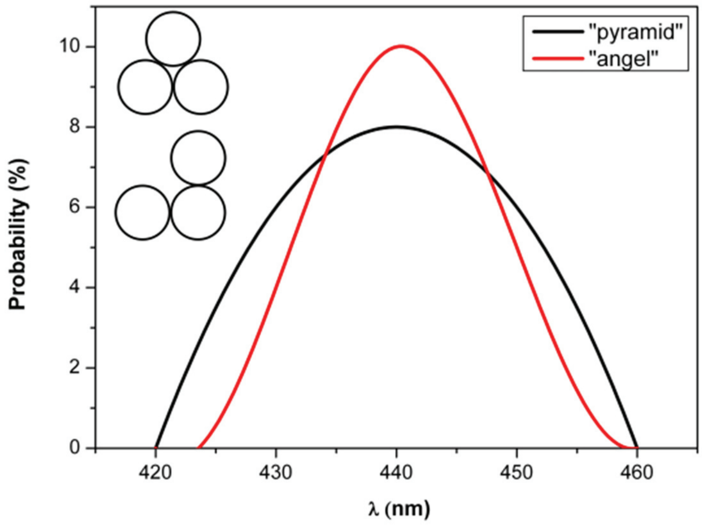

Figure 2 Dependence of maximum probability of formation of structures of a pyramid and angle types on the wavelength of external field

As one can see in **Figure 2**, radiation in the short wavelength range allows formation of two types of stable structures ("pyramid" and "angle") with a good selectivity on the wavelength. However, since for three particles unfixed in space it is impossible to use the selection on the laser field polarization, in these case the formation of both structures is expected to happen with approximately equal probability (10 % и 7.5 %).

Therefore, formation of structures is shown to be possible with the help of a laser with 10 ns pulse duration. Using of longer pulses as well as higher intensity is not unambiguously useful since they will lead not only increase of formation probability but decrease of the selectivity formation of pre-determined structure on the laser frequency. Increase of repetition rate of laser pulses will lead to the increase of overall probability of structures' formation per the unit of time, however, it can produce the radiation heating of a solution.

3. CONCLUSION

The possibility of formation of structures with pre-defined configuration in the field of laser radiation is shown via computer modeling. Using laser pulse with short duration of order of 10 ns, three types of stable structures can be obtained. The geometry of structures depends on the external field wavelength that allows performing their selective formation. Linear structures will be formed for the laser wavelengths in the 720-780 nm range, and the mixture of structures of two types ("pyramid" and "angle" will be formed under irradiation in the 420-460 nm range.

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