Special Section Guest Editorial: Optics, Spectroscopy, and Nanophotonics of Quantum Dots

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This special section of the *Journal of Nanophotonics* is focused on optics, spectroscopy, and nanophotonics of quantum dots.

The study of optical properties of colloidal quantum dots (QDs) and nanostructures, which are formed based on them, is actual for modern nanophotonics. Interest in these structures is caused by the widest range of practical applications in various fields. The most actual applications of photonics of QDs are fluorescent labeling of biological objects, biosensors, and minimally invasive biomedical technology, including thermal and photodynamic therapy of severe human diseases.[1–3] Primarily, the unique optical properties of colloidal QDs are size-dependent optical absorption and photoluminescence spectra, a wide range of luminescence excitation, high photostability of nanocrystals, etc. These properties of colloidal QD spectroscopy provide opportunities for the development of optically distinguishable codes, identifying various diseases, markers of diseased cells, tissues, and organs with its subsequent visualization. The possibility of specific binding of bioconjugated QDs with different targets provide opportunities for labeling of cells and a variety of protein molecules both *in vitro* and *in vivo*.

The basis of marked applications is fundamental processes. They are[1–3]: absorption of light by colloidal QDs; the formation of excitons; radiative and nonradiative annihilation of excitons; radiative recombination of localized excitons; relations of processes of recombination luminescence with QD size and their composition (special in the case of substitutional solid solutions); the exchange of electronic excitations between colloidal QDs and organic structures, which interact with interface of quantum dots.

Thus, the authors' papers[1–3] develop these important areas of optics and spectroscopy of colloidal quantum dots.

A simple model of a quasi-zero-dimensional structure in the form of a spherical QD of radius a and permittivity ε_2, embedded in a medium with permittivity ε_1, was discussed in Ref. 4. An electron (e) and a hole (h) with effective masses m_e and m_h were assumed to travel within the QD. We assume that the permittivities satisfy the relation $\varepsilon_2 \gg \varepsilon_1$ and that the conduction and valence bands are parabolic.

The theory of exciton states in QDs under conditions of dominating polarization interaction of an electron and a hole with a spherical (QD – dielectric matrix) interface are developed in Ref. 4. It is shown that the energy spectrum of a heavy hole in the valence band QD is equivalent to the spectrum of a hole carrying out oscillator vibrations in the adiabatic electron potential.[5] It is shown that the absorption and emission edge of QDs is formed by two transitions of comparable intensity from different hole size–quantization levels and into a lower electron size–quantization level.[6]

The interband absorption of light in QDs was studied theoretically in Ref. 6 using the dipole approximation in the framework of the model [4] considered here, and under the assumption that the absorption length $\lambda \gg a$. An expression for the quantity $K(\bar{s}, \omega)$ defined by the hole optical transition from the energy level $t_h = 2n_h$ (t_h is the hole main quantum number, n_h, is the hole radial quantum number) to the lowest electron level ($n_e = 1$, $l_e = m_e = 0$) (here

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n_e, l_e, m_e are the main, orbital and magnetic quantum numbers of an electron) was derived in the following form[6]:

$$K(\bar{s}, \omega) = A \sum_{n_h} L_{n_h}(\bar{S}), \tag{1}$$

where ω - incident light frequency, $(\bar{S}) = (a/a_h)$, $a_h = \varepsilon_2 \hbar^2/(m_h e^2)$ is the Bohr radius of the hole in QD, and A is proportional to the square of the absolute value of the dipole moment matrix element calculated with Bloch functions. The quantity $K(\bar{s}, \omega)$ (1) connects the energy absorbed by QD in a time unit with the time average of electric field square of incident wave. Moreover, the product of $K(\bar{s}, \omega)$ and QD concentration in the dielectric matrix gives electric conductivity of the considered quasi–zero–dimensional system for the frequency ω, which is connected with light absorption coefficient in the usual way.

We determine the quantity $K(\bar{s}, \omega)$ (1) corresponding to hole optical transition from the energy level $t_h = 2n_h$ to the lowest electron level ($n_e = 1, l_e = m_e = 0$). In this case, the expression for the quantity $L_{n_h}(\bar{s})$, given by the square of the overlap integral of the electron and hole wave functions, takes the form (see article S.I. Pokutnyi, O.V. Ovchinnikov, T.S. Kondratenko[1])

$$L_{n_h}(\bar{s}) = \frac{2\pi^{5/2}}{\left(1 + \frac{2}{3}\pi^2\right)^{3/4}} \frac{n_h + 1}{2^{2n_h} n_h!} (\bar{s})^{-3/4}. \tag{2}$$

In the interband optical absorption spectrum of QD each line corresponding to given values of the radial n_e and orbital l_e quantum numbers turns into a series of close lying equidistant levels, corresponding to various values of the main hole quantum number t_h.[7] This conclusion follows from Eqs. (1) and (2), and is a direct consequence of the Coulomb and polarization interactions of an electron and a hole in QD.

In Ref. 1, estimated value of the overlap integral square $K(\bar{s}, \omega)/A$, using (1), (2) and experimental results of the absorption of colloidal CdS QDs, synthesized by an aqueous synthesis in a gelatin matrix was investigated. For the hole transitions from the equidistant quantum levers: ($n_h = 0; l_h = m_h = 0$), ($n_h = 1; l_h = m_h = 0$), ($n_h = 2; l_h = m_h = 0$), ($n_h = 3; l_h = m_h = 0$) (here l_h, m_h are the orbital and magnetic quantum numbers of an hole), to the lowest electron size – quantized level ($n_e = 1, l_e = m_e = 0$), we have

$$\frac{K(\bar{s}, \omega)}{A} = L_1(\bar{s}) + L_2(\bar{s}) + L_3(\bar{s}), \tag{3}$$

where $L_0 = 1.639 \, \bar{s}^{-3/4}$, $L_1 = 0.5 \, L_0$, $L_2 = 9.38 \cdot 10^{-2} \, L_0$, $L_3 = 10^{-2} L_0$. From the above expression, it follows that the main contribution in the light absorption coefficient is a cadmium sulphide QD from the hole spectral lines corresponding to quantum numbers ($n_h = 0$; $l_h = m_h = 0$) and ($n_h = 1$; $l_h = m_h = 0$) the transition oscillator strengths of which are dominant.[1] The contribution of higher exited hole lines ($n_h \geq 2$; $l_h = m_h = 0$) is negligible.

This way, in the framework of the considered model of the quasi–one–dimensional system [4–6] it was shown that the absorption and emission edge of a cadmium sulphide QD is formed by two transitions of comparable intensities.[7] Estimations of average values of CdS QDs radius were realized using the developed formalism for UV-Vis absorption spectra. These data were compared with experimental values of this parameter, obtained using transmission electron microscope.[1]

The paper (see Ref. 2) presents the results of studies of formation of luminescent properties of hydrophilic colloid solutions, containing hybrid associates, constructed from Ag_2S QDs (2.5 nm) with J-aggregates of 3,3'-di-(γ-sulfopropyl)-4,4',5,5'-dibenzo-9-ethylthiacarbocyanine betaine pyridinium salt (Dye1) and thionine molecules (Dye2) in gelatin. For Dye1 molecules, the tendency to form cis- and trans-isomeric forms is known along with J-aggregation. Cations of Dye2 molecules distinguish their tendency to dimerization and H-aggregation.

The effect of photosensitization of IR luminescence excitation (1205 nm) of colloidal Ag_2S quantum dots (QDs) with average size of 2.5 ± 0.6 nm in gelatin at 600 to 660 nm by molecules of Dye1 and Dye2 was registered. Cis-J-aggregates of Dye1 and cations monomer of Dye2

conjugated with Ag_2S QDs take part in this process. The photosensitization of luminescence excitation of colloidal Ag_2S QDs was interpreted by resonance nonradiation transfer of electronic excitation energy from cis-J-aggregates of Dye1 and cations of Dye2 to centers of recombination luminescence of Ag_2S QDs.[2]

The hybrid association of colloidal Ag_2S QDs with molecules of Dye1 and Dye2 provides the formation of heterostructures of the first type. Under conclusions of this location of energy states of the hybrid associates component, the exchange of electronic excitation between molecules of Dye1 and Dye2 and Ag_2S QDs is possible under excitation of dye molecules due to non-radiative resonance energy transfer of electronic excitation. At the same time, in the case of Dye1, excited cis-J-aggregates sensitize optical transitions with a lower probability, which lead to the excitation of luminescence centers with the participation of deep levels of size quantization of electrons (excited states of holes). In the case of Dye2, monomers take part in this process. The excitation of a luminescence center by light of 440 nm is possible due to its properties. In this case, lights action of excitation radiation is probably. Its mechanism is to overcome the finite potential well by a free hole, which is due to the spatial limitation of Ag_2S crystal. Tunneling of the free hole into the matrix and its localization at macroscopic states caused by a jump of the dielectric constant in the interface between matrix and QD will led to a decrease in the intensity of radiative recombination. For long wavelength photons with lower energy of 2.33 eV (532 nm), 1.95 eV (635 nm), and 1.88 eV (660 nm), the probability of ejection of a hole into the matrix is less. Molecules of Dye1 and Dye2 sensitizing optical transitions for these wavelengths decrease this probability. This is due to decreasing of energy of photosensitizing photons due to Stokes losses and scattering on phonons of energy of excited dye molecules during non-radiative transfer of electronic excitation energy. In the case of Dye1, the decrease in intensity of infrared luminescence of Ag_2S QDs under excitation of 660 and 635 nm is due to the photodegradation of trans- J-aggregates in a gelatin matrix. Thus, decreasing of efficiency of excitation of Ag_2S QDs infrared luminescence under prolonged excitation is due to ionization of Ag_2S QDs with involvement of deep size-quantized states in the valence band. The main mechanism of photosensitization of luminescence is resonance nonradiative transfer of electronic excitation energy from Dye1 and Dye2 forms, which are active in this process to radiative recombination centers in Ag_2S QDs. This channel of exchange of electron excitation has not been considered in the photophysics of hybrid associates. It can be used for fluorescent labeling for the near-IR region, including biological window transparency. Founding the possibility of association of Ag_2S QDs with cations of Dye2 shows the possibility of simultaneous sensitization of singlet oxygen near the hybrid associate.[2]

The paper (see Ref. 3) analyzes the dependence of properties of optical absorption and photoluminescence spectra and morphology of $Cd_xZn_{1-x}S$ QDs, obtained by method of aqueous synthesis in gelatin on the ratio of concentrations of cadmium and zinc atoms in the crystal lattice. Aqueous synthesis of mixed cadmium and zinc sulfides colloidal QDs has been successfully realized. The continuous shift of reflexes on XRD patterns indicates the formation of $Cd_xZn_{1-x}S$ solid solutions. With increasing zinc content in QDs, there is a nonlinear change of lattice parameter from composition, which indicates the deviation from Vegard's law. Similar behavior is also characteristic of the dependence of the effective band gap on composition.

Colloidal $Cd_xZn_{1-x}S$ QDs are formed in a cubic crystal lattice with particle size of ~2 nm. The blue shift of optical absorption spectra from 420 to 295 nm and recombination photoluminescence from 646 to 483 nm with increasing zinc content in QDs was observed. Optimum photoluminescence intensity occurs for QDs with $Cd_{0.3}Zn_{0.7}S$ composition. With increasing zinc content up to $Cd_{0.3}Zn_{0.7}S$, luminescence intensity increases and decreases when zinc content is larger than 0.7.[3]

A model is proposed according to which during the synthesis of QDs in a gelatin matrix, isoelectronic impurity, for example, oxygen atoms in the solvent, replaces one of the sulfur atoms in the elementary tetrahedron. This disturbs the balance of forces acting on the central metal atom.[3]

The increase in photoluminescence intensity is explained by the increase in the number of point defects, such as complexes of interstitial metal atoms–metal vacancies $[Me_i - V_{Me}]$. Such complexes occur due to displacement of metal atom at the center of the elementary

tetrahedron due to substitution of one from four sulfur atoms by an impurity atom, such as an oxygen atom.[3]

This pair of defects is the center of luminescence of the donor–acceptor type. In conclusion, it should be noted that $Cd_xZn_{1-x}S$ QDs with changeable properties can be potentially applied in biology and medicine as fluorescent labels. Moreover, after the removal of the dielectric matrix, $Cd_xZn_{1-x}S$ QDs can be used in light-emitting diodes (LEDs) and QD-LED displays [3].

In A. Sergeev, et al.,[7] CdS-silicate nanocomposite was gradually heat treated up to 120°C. Step-by-step changing of its structural and optical characteristics was studied. Being more energy-effective than $\lambda = 405.9$ nm laser radiation, thermal treatment may lead to a higher degree of nanocomposite optical property modification. That could be coincided as the basis for detailed understanding of processes.

It's known, that the QD absorption band is determined by its sizes and blue-shifted with size decreasing. Under annealing the edge of the fundamental absorption band changes from $E_g = 3.22$ eV for initial nanocomposite to $E_g = 2.55$ eV for nanocomposite annealed at 180°C. The luminescence spectrum of the initial nanocomposite (20°C) has one wide emission band that consists of three Gaussian bands with maxima at 2.65, 2.35, and 2.05 eV. The FWHM of these bands are 0.42, 0.54, and 0.63 eV, respectively. Under thermal treatment, longwave spectral broadening of the luminescence occurs. This changes indicate that thermal annealing leads to equilibrium state between two structural modifications: non-crystalline and randomly ordered with defects.

It can be assumed that thermal treatment primarily affects QDs separated by distance less than the radius of gyration. Increasing the temperature leads to increasing of particle mobility. At the first stage (up to 100°C) the particle mobility initiates the increase of QD size due to aggregation and, to a lesser degree, affects their structure. Further annealing (100–160°C) leads to structural changes in the aggregates of nanoparticles without affecting the single isolated nanoparticles.

The changes of QD optical properties mentioned above allow us to suggest possible changing in QD dimensions and/or structure. By using a small and wide-angle x-ray scattering (SWAXS) technique, it was found that the annealing process leads to significant structural reordering of nanocomposite. The difference between SWAXS patterns of annealed at 120°C and as-synthesized nanocomposite correspond to structural re-ordering of nanoparticles. Under laser exposure to $\lambda = 405.9$ nm similar structural changes in small and medium angles regions occur, which indicate agglomeration of nanoparticles into larger systems. As to as-synthesized nanocomposite, wide diffusion halo with maximum near 27 deg is observed. This pattern consists of broad peaks of hexagonal and cubic structures due to the smaller particle size. It also cannot be determined as a polytype structure, allowing us to suggest non-ordered (amorphous-like) structure of these CdS QDs. After thermal annealing diffusion halo resolves into two narrow peaks at 25.5 deg and 27 deg, accomplished with randomly ordered crystalline structure.

"Pump&probe" studies have shown that the increasing of annealing temperature reduces the exposure dose required for formation of modified area during the primary exposure. At temperatures ≥100°C the primary exposure dose value tends to zero, and there is no need for primary modification. Thus, laser radiation exposure and thermal treatment perform similar effects on the nanocomposite and initiate its structural and interparticle changes.

Thus, one can suppose that presence of non-crystalline CdS QDs is the main reason for appearance of photoinduced changes in the absorption coefficient of nanocomposite. QDs of that kind have emission maximum at 2.7 ± 0.05 eV. Exposure to laser light at $\lambda = 405.9$ eV (3.05 eV) leads to partial ordering of the QD structure. Partial structural changes caused by modifying laser radiation leads to the returning of optical characteristics of nanocomposite to initial level. However, this mechanism is valid only for isolated quantum dots. In the case of several quantum dots at a distance less than their radius of gyration, the modifying radiation causes their aggregation, along with formation of CdS nanoparticles with randomly ordered crystalline structure. The presence of such nanoparticles gave rise to linear absorption coefficient of the nanocomposite, and slightly dissipates modifying radiation during the measurement process. Their presence can be defined by emergence of emission bands in the range of 1.4–1.7 eV.

The study by V. Mykhaylovsky, V. Sugakov, I. Goliney[8] suggests an idea of a generator of traveling pulses of the excitonic condensed phase in laser illuminated double quantum well heterostructures. The pulses would manifest themselves as the bright spots in the emission moving along the plane of the double quantum well despite the time-independent steady pumping. The predicted phenomenon is similar in a way to the Gunn's effect in semiconductors.

The object of the study is a system of indirect excitons of high density created by a laser illumination. Indirect excitons are a type of excitation in a double quantum well in which the electron and the hole are separated to different wells of the double quantum well structure with an external electric field. The small overlap of the wave functions of the electron and the hole that comprise an indirect excitons makes them long living quasi-particles and allows their accumulation to high densities at moderate intensity pumping, thus making them a popular research object in the field of non-linear effects.

For the last decade and a half, a number of interesting effects were discovered in the high-density systems of indirect excitons. The low temperature luminescence from the laser irradiated double quantum wells comes from rings at significant distances from the laser spots and is frequently fragmented into bright spots. This suggests some kind of excitonic condensation. The Bose-Einstein condensation has been suggested to be responsible. Yet, the authors of the paper have argued that the Bose-Einstein condensation requires exciton coherence at the distances far exceeding the exciton diffusion length, and have been advancing a different explanation via the self-organization in the high-density systems of interacting quasi-particles with finite lifetimes. Attraction required for the condensation of the excitons may originate from the exchange and Van der Waals exciton-exciton interactions. This approach allowed authors to recreate quantitatively the experimentally observed patterns of fragmentation, a feat other models could not achieve.

Further analysis has shown that the periodical luminescent structures can be set to motion in an external on-plane bias, for example if the system is set up as a slot in an electrode (see references to papers by Sugakov with co-authors in Ref. 8).

The paper[8] suggests an experimental setup in which a generator of traveling pulses can be realized. Analysis of the kinetic equation describing the evolution of the density of indirect excitons with the Landau-type expression for the free energy and account for the external bias shows that there is a region of parameters (exciton density, exciton lifetime) within which the system may support both the uniform distribution of exciton density and the non-uniform distribution where some of the excitons gather into islands of the condensed phase while others are in the gas phase. There is a dynamic equilibrium between the islands of the condensed phase and the surrounding. Because of the finite exciton lifetime the uniformly created excitons decay within the islands and in the gas phase. The attractive interaction between excitons allows the islands to harvest excitons from the surrounding thus stabilizing their density at the level sufficient to maintain the condensed phase. When the pumping is stronger, the only stable solution of the kinetic equations is the uniform condensed phase, if it is weaker, the only solution is the excitonic gas. There is also a region of parameters in which the solution are periodical but cannot be controlled in a desired way.

The proposed generator of the traveling pulses consists of two regions. The main region where the pulses can travel is broad and pumped with excitons to the level that supports both the uniform exciton distribution and the nonuniform solution in the form of islands of the condensed phase surrounded by the excitonic gas. An inplane bias is applied to this region in order to create a driving force for the motion of condensed phase islands. The second region is narrow and pumped with excitons to the level at which only the condensed phase is possible. As the calculations show, the traveling pulses of the exciton density are born at the boundary of two regions and periodically in time move away drifting in the broad main space. The regular in time generation of pulses occurs at the steady in time illumination.[8]

Numerical simulation of the system shows that the pulse generation starts if the pumping rate and the driving force exceed certain threshold values. As the driving force increases while keeping the fixed pumping rate in the propagation area, the velocity of the pulses' drift and the frequencies of the generation increase.[8] The authors claim that the considered system is potentially applicable in laser controlled opto-electronic devices. It may be useful for the energy and information transfer in microsystems.[8]

J. Zribi et al.[9] report on a chemical beam epitaxy growth study ofInGaAs/GaAsInGaAs/GaAs quantum dots engineered using an *in-situ* indium-flush technique. The emission energy of these structures has been selectively tuned over 225 meV by varying the dot height from (7 to 2) nm. A blue-shift of the photoluminescence (PL) emission peak and a decrease of the intersublevel spacing energy are observed when the dot height is reduced. Numerical investigations of the influence of dot structural parameters on their electronic structure have been carried out by solving the single-particle one-band effective mass Schrödinger equation in cylindrical coordinates, for lens-shaped QDs. The correlation between numerical calculations and PL results is used to better describe the influence of the In-flush technique on both the dot height and the dot composition.

The problem of finding effective characteristics of the nanostructure consisting of spherical shells (nanomatryoshka) is solved using the matrix homogenization method (see article I.A. Starkov, A.S. Starkov[10]). According to the problem formulation, each of the system layers possesses piezoelectric and/or piezomagnetic properties. Thus, the mutual influence of the elastic, electrical, and magnetic characteristics of the shells on their homogenized values is investigated. In particular, the dependence of the dielectric and magnetic permittivity on the geometrical parameters of the layers is studied and analyzed. The performance of the model is illustrated for a two-layer structure.

Electronic states and direct interband light absorption in the ensemble of prolate spheroidal quantum layers (SQL) are considered. The problem of finding the one-electron wave function and energy spectrum have been solved exactly (see article D.A. Baghdasaryan, D.B. Hayrapetyan, E. M. Kazaryan[11]). For light absorption coefficient for strong and intermediate regimes, we have the following expression:

$$K(\Omega) = K_0 \sum_{\nu,\nu'} \left| \int \Psi_\nu^e \Psi_{\nu'}^h dV \right|^2 \delta(\hbar\Omega - E_g - E_\nu^e - E_{\nu'}^h), \tag{4}$$

where E_g is the energy gap of the semiconductor, Ω is frequency of the incident light, $\nu = \{n, l, m\}$ and $\nu' = \{n', l', m'\}$ are the sets of quantum numbers of the electron and the hole respectively. In the regime of strong size quantization the energy of Coulomb interaction between an electron and a hole is much smaller than the energy caused by the walls of the SQL. In this approximation, the Coulomb interaction between particles can be neglected. Note that the selection rule for azimuthal quantum numbers $m_e = -m_h$ was immediately revealed from the expression for absorption coefficient [Eq. (4)]. For the orbital-like and principal quantum numbers there is following selection rules $l_e = l_h$ and $n_e = n_h$.

The dependence of absorption edge in the regime of strong size quantization have been obtained and this dependence monotonically reaches $\hbar\Omega_{100} = E_g$ because with the increase of the small semiaxes impact of the size quantization on the system becomes less, thus the absorption edge of the system tends to the absorption edge of the bulk sample.[11] The dependences of the absorption coefficient [Eq. (4)] on the frequency of incident light for the both cases of Gauss and Lifshits–Slezov distribution functions are calculated. The intensity of the first pick that corresponds to the transition between the ground states of electron and hole has the biggest value. The intensities of subsequent transitions are decreasing.

Absorption edge dependence on the thickness of the layer in the strong size quantization regime has been obtained. The effect of nonparabolicity of the dispersion law of energy levels and optical absorption have been taken into account and calculations are carried out for the cases of both parabolic and Kane's dispersion laws. Selection rules have been revealed. Absorption coefficient [Eq. (4)] dependence on the frequency of incident light has been obtained, taking into account dispersion of nanolayer thicknesses for the cases of both symmetric and asymmetric distribution functions.[11]

We present a combined experimental and simulation study of a single self-assembled InGaAs quantum dot coupled to a nearby (~25 nm) plasmonic antenna (see Ref. 12). Microphotoluminescence spectroscopy shows a ~2.4× ~2.4× increase of intensity, which is attributed to spatial far-field redistribution of the emission from the QD antenna system. Power-dependent studies show similar saturation powers of 2.5 μW for both coupled and uncoupled QD emission in polarization-resolved measurements. Moreover, time-resolved spectroscopy reveals the

absence of Purcell enhancement of the QD coupled to the antenna as compared with an uncoupled dot, yielding comparable exciton lifetimes of $\tau \sim 0.5$ ns. This observation is supported by numerical simulations, suggesting only minor Purcell-effects of $< 2\times < 2\times$ for emitter–antenna separations >25 nm. The observed increased emission from a coupled QD–plasmonic antenna system is found to be in good qualitative agreement with numerical simulations and will lead to a better understanding of light–matter coupling in such semiconductor–plasmonic hybrid systems.

A surface plasmon polariton is an electromagnetic wave that propagates along an interface between two materials with dielectric permittivity of opposite signs. Such waves can be focused by metal waveguides of special geometry (see article P.A. Golovinski, V.A. Astapenko, E.S. Manuylovich[13]). The spatial distribution for a near-field strongly depends on a linear chirp of the laser pulse, which can partially compensate the wave dispersion. Field distribution is calculated for different chirp values, opening angles, and distances. The spatial selectivity of excitation of quantum dots using focused fields is shown using Bloch equations.

We hope that the readers of the *Journal of Nanophotonics* will enjoy this special section. It should bring new ideas to a wide audience of scientists, researchers, and students working in the field of optics, spectroscopy, and nanophotonics nanostructures containing semiconductor and dielectric quantum dots.

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