

Semiconductor nanocrystal based saturable absorbers for optical switching applications

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Abstract—This document presents experimental and theoretical results of an effort to develop semiconductor nanocrystal doped thin films for optical switching applications. Films doped with high quality PbS and PbSe nanocrystals have been fabricated. Measurements of the absorption spectra are compared with theoretical predictions. The general theoretical framework for treating the optical properties of multi-layer dielectric structures containing thin nanocrystal doped layers is presented. A simple model (the particle-in-a-sphere) is used as a starting point, however, more sophisticated models (such as those based on $k \cdot p$ theory, or fits to experiment) can easily be incorporated. A prototypical micron-scale Fabry-Perot optical cavity has been constructed and is discussed. Theoretical predictions for the switching behavior of such a structure, upon introduction of the semiconductor nanocrystal material into the optical cavity, are presented.

Index Terms—optical switches, quantum dots, optical saturation, optical bistability, optical materials, optical propagation in nonlinear media, optical planar waveguides, microcavity, cavity resonators, Fabry-Perot resonators, optical films, semiconductor switches

I. INTRODUCTION

CONSIDERABLE progress in the development of all-optical switching technology has been achieved in recent years fueled by the promise of widespread commercial applications [1], [2]. Many optical switching schemes have been proposed, but one thing they all have in common, however, is the use of some sort of non-linear optical material, that is, a material exhibiting optical properties that depend on the state of the electromagnetic fields present.

Another important application of non-linear optic materials is the use of solid state saturable absorbers in mode-locked laser applications. A saturable absorber is a material that exhibits an optical absorption band, the strength of which decreases with increasing field intensity as, for example:

$$\alpha(E) = \frac{\alpha_0}{1 + |E|^2/|E_{sat}|^2} \quad (1)$$

where α_0 , and E_{sat} are constants with respect to the electric field strength but, in general, depend on frequency. In practical applications, one would like to have a material that exhibits the lowest possible value for the saturation field strength E_{sat} since the optical power required to achieve a given degree of saturation is proportional to $|E_{sat}|^2$.

In recent years, a number of researchers have demonstrated the feasibility of using transparent materials (such as glass)

doped with semiconductor nanocrystals as saturable absorbers for laser applications [3]–[7]. PbS and PbSe nanocrystals were used in these studies since they exhibit a size-tunable absorption resonance in the neighborhood of the $1.55\mu m$ wavelength used in telecommunications applications. These materials are promising because they are inexpensive and have a relatively low value of the saturation intensity ($\sim 0.18 MW/cm^2$) [6].

In addition to the promising results of these studies, the recent commercial availability of high-quality semiconductor PbSe nanocrystals [8], which can easily be incorporated into polymer thin films, suggests the possibility that these materials might be of use for optical switching applications.

The present work documents preliminary experimental and theoretical efforts aimed at using semiconductor nanocrystal doped thin films for optical switching applications. Experimental absorption spectra are presented and discussed in comparison with the results of a simple model. The general framework for treating the optical properties of such materials is presented which can easily incorporate more sophisticated models. A micron-scale prototypical realization of a Fabry-Perot optical cavity is presented along with theoretical predictions of the switching behavior of such a structure upon introducing semiconductor nanocrystals into the optical cavity.

II. THEORETICAL APPROACH

Our objective in this section is to develop simple models of the non-linear optical properties of a semiconductor nanocrystal based saturable absorber material. The lead-salt quantum dots (PbS and PbSe) are well suited to telecommunications applications because their first exciton absorption peak can be size-tuned to coincide with $1.55\mu m$ wavelength of choice for optical fiber applications.

We consider PbS and PbSe quantum dots in the size range from 3 to $10nm$ in order to tune the first exciton resonance to wavelengths neighboring $1.55\mu m$. Dots of these sizes are considered to be in the *strong confinement* limit, since the bulk Bohr radii ($18nm$ for PbS, and $46nm$ for PbSe) are much larger than the size of the nanocrystal [9]. In the strong confinement limit we expect the kinetic energy to dominate over potential energy.

The simplest model, which neglects potential energy, is sometimes called the *particle-in-a-sphere model*. In this model, the electrons and holes are treated as non-interacting particles confined to the quantum dot (which is assumed to be a sphere) by an infinitely high potential barrier. The energies and wave functions are obtained by solving a single-particle Schrödinger equation in which the mass is taken to be the

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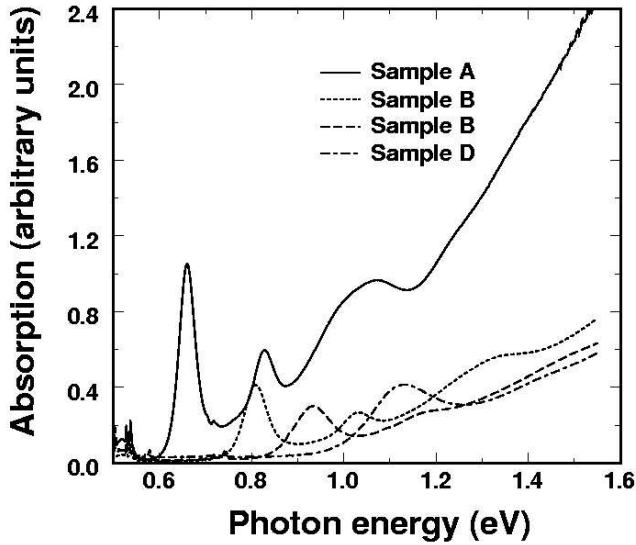


Fig. 1. Measured absorption spectra for PbSe quantum dots of various sizes clearly indicating the quantum confinement effect (size dependent shift of exciton resonances). For the largest dots (Sample A) several exciton peaks are present.

reduced mass of the electron hole pair (band edge effective masses are used), and parabolic band dispersion is assumed.

The particle-in-a-sphere model was asserted to be inadequate to correctly describe the electronic structure of the PbS and PbSe quantum dots because of the assumption of parabolic bands [10]. The lowest energy bulk transitions in PbS and PbSe occur near the L-points in the Brillouin zone. The energy dispersion of these bands is markedly non-parabolic. Therefore, in [10] an improvement over the particle-in-a-sphere model was developed based on the four-band $k \cdot p$ approach. In that work, it was claimed that the particle-in-a-sphere model was incapable of correctly describing the exciton resonances in PbS and PbSe, however no direct comparisons of the excited state spectra were compared.

A. Measured optical properties

In this work we will make detailed comparisons between the simple particle-in-a-sphere model and measurements of the excited state spectra. This work is motivated by the recent availability of high-quality PbS and PbSe quantum dots that exhibit multiple exciton peaks [8]. Figure 1 presents the measured absorption spectra for four samples containing PbSe quantum of different sizes. The extremely high quality of the material allows one to discern several exciton peaks for the sample containing the largest quantum dots (Sample A). By focusing on Sample A, which contains multiple exciton peaks, we have the opportunity to test the predictions of various models for the excited state spectrum. We begin with the simple particle-in-a-sphere model.

B. Particle-in-a-sphere model

The derivation of the simple particle-in-a-sphere model proceeds as follows. The energies and wave functions of the electron-hole states (excitons) are obtained by solving the

Schrödinger equation in the parabolic effective-mass approximation. The confining potential is taken to be zero inside the quantum dot and infinite outside. This potential translates into the boundary condition that the wave function must vanish at the surface of the quantum dot. Because of the spherical symmetry of the potential, the wave functions factorize into radial and angular pieces:

$$\psi_{nlm} = R_{nl}(r)Y_{lm}(\theta, \phi), \quad (2)$$

where Y_{lm} are spherical harmonics. The radial function satisfies the following equation:

$$-\frac{1}{r} \frac{d^2}{dr^2} (rR_{nl}(r)) + \left(\frac{l(l+1)}{r^2} - k^2 \right) R_{nl}(r) = 0. \quad (3)$$

The normalized solutions, which vanish on the boundary of the sphere of radius “a”, are given by:

$$R_{nl}(r) = \sqrt{\frac{2}{a^3}} \frac{j_l(\kappa_{nl}r/a)}{j_{l+1}(\kappa_{nl})}, \quad (4)$$

where, κ_{nl} is the n 'th root of the spherical Bessel function j_l , as discussed in [11]. The corresponding energy spectrum is given by:

$$E_{nl}(a) = E_g + \frac{\hbar^2}{2m_r} \left(\frac{\kappa_{nl}}{a} \right)^2, \quad (5)$$

where, E_g is bulk band gap, m_r is the electron-hole reduced mass:

$$\frac{1}{m_r} = \frac{1}{m_e^*} + \frac{1}{m_h^*}, \quad (6)$$

obtained from the L-point electron and hole effective masses m_e^* , and m_h^* , respectively.

C. Optical properties

Our primary interest in this work is the saturation behavior of the linear dielectric properties. We therefore compute the linear susceptibility as:

$$\chi(\omega) = -\frac{\text{const}}{\omega V} \sum_{nl} \frac{2l+1}{\hbar\omega - E_{nl}(a) + i\gamma} + c.c., \quad (7)$$

where, $c.c$ denotes the complex conjugate, as discussed in [11]. In this equation, we have made the simplification that all of the allowed ($\Delta l = 0$) inter-band matrix elements are equal. The quantity V is the sphere volume, and γ is a phenomenological constant describing homogeneous broadening. The constant in front of the susceptibility is treated as a fitting parameter. The dielectric constant for a single quantum dot of radius a is obtained from the susceptibility as:

$$\epsilon_a(\omega, a) = \epsilon_\infty + 4\pi\chi(\omega, a). \quad (8)$$

where ϵ_∞ , is the high-frequency dielectric constant of the corresponding bulk material. The following parameters:

$$E_g = 0.28\text{eV}; m_r = 0.038; \epsilon_\infty = 23, \quad (9)$$

are assumed for PbSe and:

$$E_g = 0.41\text{eV}; m_r = 0.062; \epsilon_\infty = 17, \quad (10)$$

are assumed for PbS [10].

In order to account for a distribution of sizes, we average the dielectric function over a Gaussian distribution peaked about a particular radius a_0 :

$$\langle \epsilon_d(\omega, a_0) \rangle = \epsilon_\infty + 8\pi \sqrt{\frac{\eta}{\pi}} \int_0^\infty \chi(\omega, a) e^{-\eta(a-a_0)^2} da. \quad (11)$$

From this point on we shall drop the angular brackets indicating the average over size distribution and simply write $\epsilon_d(\omega)$ without the quantum dot radius a_0 . It is convenient to characterize the distribution in terms of the *broadening* y expressed as a percent. This is defined as follows. We define the *width* w of the (normalized) probability distribution:

$$P(a, a_0) \equiv 2\sqrt{\frac{\eta}{\pi}} e^{-\eta(a-a_0)^2}, \quad (12)$$

in terms of the point \bar{a} at which the distribution is reduced to e^{-1} of its value at the maximum. More precisely, \bar{a} , is defined by:

$$\eta(\bar{a} - a_0)^2 = 1. \quad (13)$$

From this we define the width as:

$$w = 2|\bar{a} - a_0| = \frac{2}{\sqrt{\eta}}. \quad (14)$$

Next we define the *broadening* y as:

$$y \equiv \frac{w}{a_0} = \frac{2}{a_0\sqrt{\eta}}, \quad (15)$$

which is a fraction $0 < y \leq 1$. Thus when we speak of, say, 10% broadening we mean the we have chosen $y = 0.1$. Thus to complete the specification of the distribution function, we pick a value for y and obtain the parameter η as:

$$\eta = \frac{4}{(ya_0)^2} \quad (16)$$

D. Another simple model

Our objectives in this work are twofold: (1) to compare the results of the simple particle-in-a-sphere model with against experiment as well as, (2) to consider a possible design for an optical switching device. For the second purpose we choose to use a model that was fit to experiment for CdS and is taken from the literature [12]:

$$\epsilon_d(\omega, S) = \epsilon_\infty + \frac{\beta(\delta + i)}{\delta^2 + 1 + S/S_{sat}}, \quad (17)$$

where $i = \sqrt{-1}$, and the parameter S allows one to express the squared electric field in intensity units:

$$\left| \frac{E_d}{E_{sat}} \right|^2 \equiv \frac{S}{S_{sat}}, \quad (18)$$

where E_d is the electric field strength in the quantum dot, and $|E_{sat}|^2$, the squared saturation field strength expressed in intensity units, is given as $S_{sat} = 58W/cm^2$. The other parameters are defined as follows: $\epsilon_\infty = 6$, $\beta = 40$,

$$\delta = \frac{\omega_x - \omega}{\gamma_x} \approx 4.75, \quad (19)$$

is the detuning from the bound exciton resonance $\omega_x = 2.555eV$, and the homogenous broadening is taken to be

$\gamma_x = 0.015meV$. This model will be used to investigate the switching characteristics of a non-linear material placed in a Fabry-Perot cavity. Effects due to inhomogeneous broadening will not be considered.

E. Optical propagation

In order to study optical propagation in a material consisting of quantum dots in a non-absorbing dielectric matrix material, we assume the volume fraction of dots to be small $N_d \leq 0.001$, allowing use to use an effective medium theory for wave propagation [12], [13]. In this theory we use the Maxwell-Garnett approximation [14] to obtain the effective dielectric function for the medium (quantum dots in a dielectric host matrix):

$$\epsilon(\omega) = \epsilon_h + \frac{3N_d\epsilon_h(\epsilon_d(\omega) - \epsilon_h)}{2\epsilon_h + \epsilon_d(\omega)}, \quad (20)$$

where, ϵ_h is the dielectric constant of the host medium. For the materials considered in this work the host medium dielectric function is on the order of 2.0 (for example for glass, $\epsilon_h = 2.25$, for polyvinyl alcohol, $\epsilon_h = 2.0$, etc.).

F. Non-linear optics

Of primary interest is the fact that the optical properties of the quantum dot based material depend on the strength of the optical electric field in a non-linear way. The expression for the linear optical susceptibility for the quantum dot, Eq. 7, is strictly valid only in the limit of weak fields. In general, $\chi(\omega)$ should be multiplied by the quantity $(1 - 2f)$, where f is the fraction of excited states. In general, there will be a separate parameter f for each transition. For simplicity we consider only a single such transition and develop the rate equation for f . The value of f is determined by the following rate equation:

$$\frac{df}{dt} = -\frac{f}{\tau} + \frac{S\sigma}{\hbar\omega D(\omega)}(1 - 2f), \quad (21)$$

Where τ is the excited state lifetime, S is a quantity proportional the squared electric field *in the dot*, $D(\omega)$ is the quantum dot density of states, ω is the angular frequency of light, \hbar is Planck's constant, and σ is the optical absorption cross section and is given by:

$$\sigma \equiv \frac{Im[\epsilon_d(\omega)]\omega V}{c\sqrt{\epsilon_h}}, \quad (22)$$

where, c is the speed of light in vacuum, and V is the volume of the quantum dot

$$V = \frac{4}{3}\pi a^3. \quad (23)$$

The factor of $\sqrt{\epsilon_h}$ in the definition of σ is for convenience so that squared electric fields can be expressed in intensity units relative to the field intensity in the host matrix material:

$$S_h = \frac{c\sqrt{\epsilon_h}}{8\pi} |E_h|^2. \quad (24)$$

Further elaboration is in order. The absorption per unit volume in the quantum dot is given by [15]:

$$A = \frac{\omega Im[\epsilon_d]}{8\pi} |E_d|^2, \quad (25)$$

where E_d is the electric field strength in the dot. Next we have a non-linear relation between the electric field in the host material and in the dot:

$$E_d = g(E_d)E_h, \quad (26)$$

(which must be solved self-consistently) to be presented shortly. Using this relation we define the quantity S (introduced earlier in the rate equation for f) as:

$$S = g^2 S_h = c\sqrt{\epsilon_h} \frac{|E_d|^2}{8\pi}, \quad (27)$$

which has units of intensity but is *not the same as the actual intensity in the dot*. Using this definition we obtain the following expression for the absorption per unit volume in the dot:

$$A = \frac{\omega \text{Im}[\epsilon_d]}{c\sqrt{\epsilon_h}} S \equiv \frac{\sigma S}{V}, \quad (28)$$

which explains the previously introduced definition of the absorption cross section σ .

We can solve the rate equation in the steady state by setting the time derivative equal to zero and solving for the excited state filling fraction:

$$f = \frac{1}{2 + S_{sat}/S}, \quad (29)$$

where we have introduced the saturation intensity:

$$S_{sat} \equiv \frac{\hbar\omega D(\omega)}{\sigma\tau}. \quad (30)$$

Thus we see that in the steady state the maximum value of the excited-state filling fraction is $f = 1/2$ which is obtained asymptotically as $S \rightarrow \infty$. In transient situations, however, f can take on any value $0 \leq f \leq 1$. For $f < 1/2$ we have absorption, $f > 1/2$ corresponds to gain, and $f = 1/2$ corresponds to transparency.

G. Non-linear field relation

The electric field strength inside the quantum dot is related to the field in the matrix material by the following non-linear relation:

$$E_d = \frac{3\epsilon_h}{2\epsilon_h + \epsilon_d(E_d)} E_h, \quad (31)$$

which must be solved self-consistently [12], [14]. In this equation we have temporarily suppressed the size and frequency dependence of the quantum dot dielectric function. Equation 31 is obtained by solving an electrostatic boundary value problem (Laplace's equation) assuming a given value of the field E_h in the host material. We have assumed that the wavelength of light in the material is considerably longer than the size and spacing of the quantum dots. In this approximation the wave equation reduces to the Laplace Equation:

$$\nabla^2 E + \epsilon \frac{\omega^2}{c^2} E \approx \nabla^2 E. \quad (32)$$

H. Multi-layer dielectric structures

Having developed an approximate theory of optical propagation in an infinite dielectric matrix containing a distribution of quantum dots, we now turn our attention to the question of how light interacts with a realistic structure of finite size. We restrict our attention to planar multi-layer structures and we assume the lateral dimensions of the structure to be much larger than the thickness. By making this approximation we can treat thin films and multi-layer structures as being effectively infinitely extended in the plane of the film.

We assume the composite material (dots in a dielectric matrix) to be weakly absorbing. This allows us to use standard techniques employed in the description of multi-layer dielectric structures with lossy dielectrics. In particular the transmission, reflection, and absorption of the structure will be calculated using the transfer matrix formalism, which is presented in numerous texts in which multi-layer dielectric structures are considered [16].

Consider first the propagation through a single dielectric layer of complex dielectric constant

$$\eta \equiv n + i\kappa = \sqrt{\epsilon}, \quad (33)$$

sandwiched between two half spaces having (real) dielectric constant n_0 . The transfer matrix which relates the electric and magnetic fields entering the structure (incident side) to those exiting the structure (transmitting side) is given by:

$$M = \begin{bmatrix} \cos(\delta) & i \sin(\delta)/\eta \\ i\eta \sin(\delta) & \cos(\delta) \end{bmatrix}, \quad (34)$$

where the complex phase shift δ is given by:

$$\delta \equiv -\frac{2\pi\eta d}{\lambda} \quad (35)$$

where λ is the free-space (vacuum) wave length, and d is the thickness of the layer. The relative electric and magnetic field strengths on the incident side, are related to those on the transmitting side by the relation:

$$\begin{bmatrix} F \\ G \end{bmatrix} = M \begin{bmatrix} 1 \\ n_0 \end{bmatrix} \quad (36)$$

where, F and G denote the electric and magnetic fields on the incident side of the structure, respectively, and we have assumed unit electric field strength on the transmitting side (which implies n_0 for the magnetic field strength on the transmitting side).

From the amplitudes F and G the reflection R , transmission T , and absorption A , of the structure are calculated as follows [16]:

$$R = \left| \frac{G - n_0 F}{G + n_0 F} \right|^2, \quad (37)$$

$$T = \frac{4n_0}{|G + n_0 F|^2}, \quad (38)$$

and

$$A = \text{Re} \left(\frac{4n(FG^* - n_0)}{|G + n_0 F|^2} \right). \quad (39)$$

The above formulae for a single dielectric layer are easily generalized to a multi-layer structure. The transfer matrix for

the entire structure is obtained by simply replacing the transfer matrix for a single layer, by the product of all the matrices corresponding to the layers in the structure (ordered from left to right corresponding to layers going from the incident to the transmitting side):

$$M = \prod_l M_l \quad (40)$$

The above equations were derived assuming fixed optical properties of the material. Since we are dealing with a non-linear problem, however, we must solve a self-consistent problem to determine the electric fields inside the matrix material as well as inside the quantum dots. Therefore we need to know the relation between the incident electric field outside the structure and the electric field in the matrix material. The relation between the fields in the matrix and the dots has already been presented.

In order to determine the fields in the matrix material we need one further transfer matrix – the matrix that determines the field quantities at an arbitrary point in a given layer of the structure. For example, suppose that we wish to determine the field quantities at a point xd_j , where d_j is the thickness of layer “ j ” and x (a dimensionless variable which takes on values $0 \leq x \leq 1$) measures the distance in the slab starting from the side closest to the transmitting edge. The transfer matrix corresponding to the point x in layer j has the same form as previously introduced for a single layer with the simple modification that the phase is given by:

$$\delta_{jx} = -\frac{2\pi xd_j \eta_j}{\lambda} \quad (41)$$

The field quantities at the point x in layer j are then given by:

$$\begin{bmatrix} F_{jx} \\ G_{jx} \end{bmatrix} = \tilde{M} \begin{bmatrix} 1 \\ n_0 \end{bmatrix} \quad (42)$$

where now the matrix \tilde{M} transforms the fields on the transmitting side to those at point x in layer j :

$$\tilde{M} = M_{jx} \prod_{l>j} M_l, \quad (43)$$

where again the product is ordered from left to right corresponding to layers going from left to right if we assume the transmitting side to be on the right. Layers are numbered from 1 to n starting with the layer adjacent to the incident medium. The matrix M_{jx} denotes the position dependant matrix which is obtained from Eq. 34 upon substituting the phase δ_{jx} from Eq. 41. Now that the field quantities have been obtained for layer j at position x we obtain the enhancement factor which relates the intensity in the material to that of the incident field:

$$\frac{S_{jx}}{S_i} = \frac{4n_0^2 |F_{jx}|^2}{|G + n_0 F|^2} \quad (44)$$

We will be primarily concerned with situations in which the non-linear material (dots in matrix) is confined to a single layer. For example, in pump-probe measurements of non-equilibrium optical properties the material will generally be contained in a thin film. For a practical example of a switching device, the non-linear material will be contained

in the central slab of a multi-layer structure such as a Fabry-Perot optical cavity. We assume the non-linear material to be weakly absorbing. For this reason, a reasonable approximation is to average the intensity over the thickness of the layer and define that to be the intensity of the host material S_h that was introduced previously:

$$S_h = \int_0^1 S_{jx} dx, \quad (45)$$

where we have assumed the non-linear material to reside in the j 'th layer of the multi-layer structure.

I. Self-consistent solution

The self-consistent solution for the steady state problem is solved as follows. For a given value of the incident field strength E_i , the dielectric properties of the composite are determined by choosing a value for the field strength in the dot E_d . For the dielectric properties, so determined, the electric field strength in the matrix E_h is determined through the use of the transfer matrix approach as described previously. The non-linear field relation, Eq. 31, is then used to determine the field in the dot \tilde{E}_d from the field in the matrix E_h . In general, the field in the dot \tilde{E}_d determined from the field in the matrix E_h , is different from the one E_d that was chosen in order to determine the dielectric properties of the composite. Therefore a new value of the dot field E_d is chosen (generating new values for the dielectric properties) and the procedure is continued until a self-consistent solution, $\tilde{E}_d \approx E_d$, is found.

III. RESULTS

We now turn to a comparison of the models introduced above to experimental results for PbS and PbSe colloidal nanocrystals (measured results provided by Evident Technologies [8]).

A. PbS absorption spectrum

The absorption spectrum for PbS nanocrystals in a polyvinyl alcohol matrix is shown in Fig.2. The parameters of the model were adjusted to obtain the best fit to the data. This fit is obtained by assuming a 20% size distribution (i.e. we set $y = 0.2$ in Eqs. 15 and 16) centered around a quantum dot radius given by $a_0 = 4.4nm$. The position and shape of the first excitation resonance is described reasonably well by this model as is the position of the second exciton peak. The strength of the second peak, however, is exaggerated. This is consistent with the assertion of [10] that the particle-in-a-sphere model is incapable of correctly describing the higher energy resonances.

B. PbSe absorption spectrum

The absorption spectrum for several samples of PbSe quantum dots in hexane where presented in Fig.1. We now consider the results labeled “sample A”. We focus on this sample as it has the largest number of visible exciton resonances against which we can test the model. These results along with the particle-in-a-sphere prediction are plotted in Fig.3 The

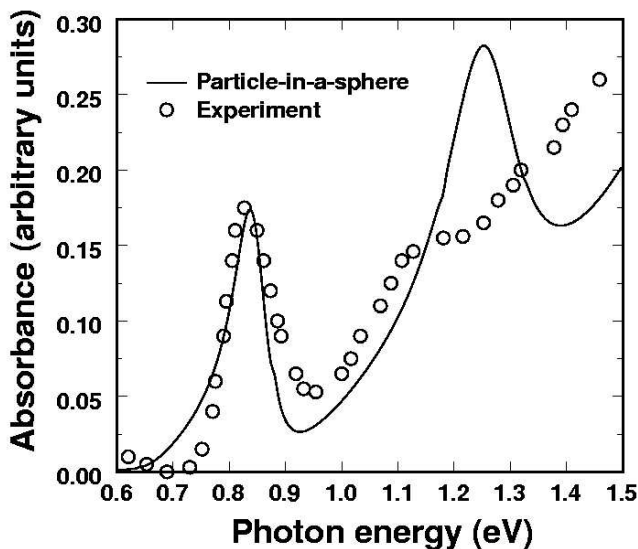


Fig. 2. Measured absorption spectrum for PbS along with the prediction given by the particle-in-a-sphere model. The shape of the first peak is reasonably represented however the strength of the second peak is exaggerated.

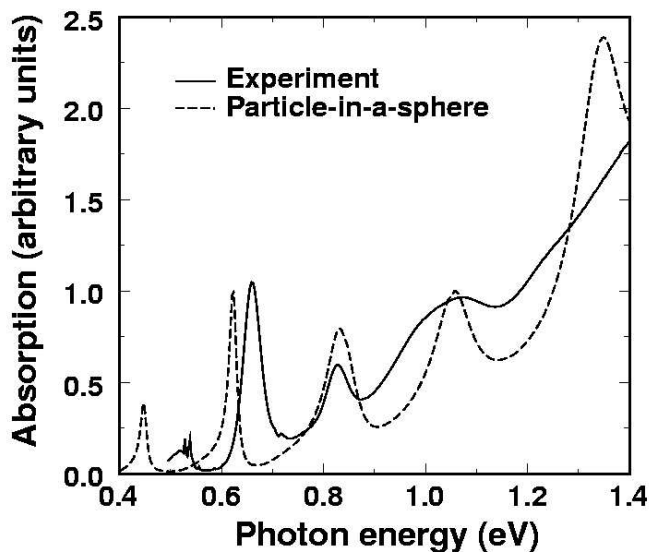


Fig. 3. Measured absorption spectrum for PbSe nanocrystals in comparison with predictions of the particle-in-a-sphere model. The sphere radius ($a_0 = 8.4nm$) and broadening (10%) were chosen to produce a close fit with the second exciton peak.

only reasonable fit to this data was obtained by choosing the parameters to fit the second exciton peak. Given the crudeness of the approximations in this model it is surprising that the model gives reasonable predictions for the higher energy resonances in terms of position and relative strengths. Here we have assumed that the lowest energy feature in the data is the first exciton peak. The predicted relative strengths of the various peaks are consistent with this assignment. At this time, however, we don't have any other supporting evidence (such as an independent measurement of the quantum dot radius) to support this assignment.

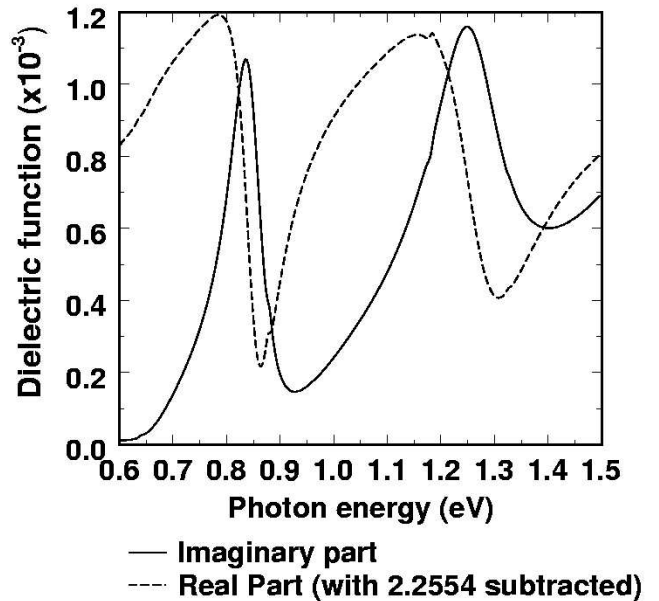


Fig. 4. Low-field frequency dependent dielectric function for PbS dots in a matrix of polyvinyl alcohol. The parameters in the model dielectric function were determined from pump-probe measurements

C. Absorption saturation of PbS nanocrystals

Pump-probe measurements were carried out for PbS nanocrystals in polyvinyl alcohol ($\epsilon_h = 2.0$) and were presented in [17]. Results of these experiments indicated that the excited state lifetime is on the order of picoseconds. Therefore we set $\tau = 1ps$. We choose the homogeneous broadening $\gamma = 0.005s^{-1}$, and the volume fraction of dots in the matrix $= 0.001$. The only remaining parameter is the overall constant in front of the linear susceptibility, which was chosen to produce an estimated absorption constant $\alpha \approx 30cm^{-1}$ at the first exciton peak. This value is consistent with previous measurements in the literature. Using these parameters fixes the value of the saturation intensity $S_{sat} \approx 94kW/cm^2$. Unfortunately a direct comparison with the measured results of [17] is not possible since only changes in transmission were presented and not the actual transmission values.

The parameters listed in the previous paragraph, along with the assumption of $\epsilon_h = 2.0$ (corresponding to polyvinyl alcohol matrix material), determines the field and frequency dependent dielectric function which is displayed in Fig. 4 for low field strength. This dielectric function was then used to determine the self-consistent solution relating the squared electric field strength (expressed in intensity units S) in the dot to the incident intensity. The results of this calculation are presented in Fig. 5 for a $150\mu m$ thick film.

We see that although the saturation intensity of the quantum dot is $S_{sat} \approx 94kW/cm^2$, the incident field intensity corresponding to this value is considerably higher ($S_i \approx 2.5MW/cm^2$). This emphasizes the fact that the effective saturation intensity of a structure (i.e. intensity in the incident medium) can be considerably different from that corresponding to the non-linear components due to dielectric effects. In certain situations, the field strength in the dot can be

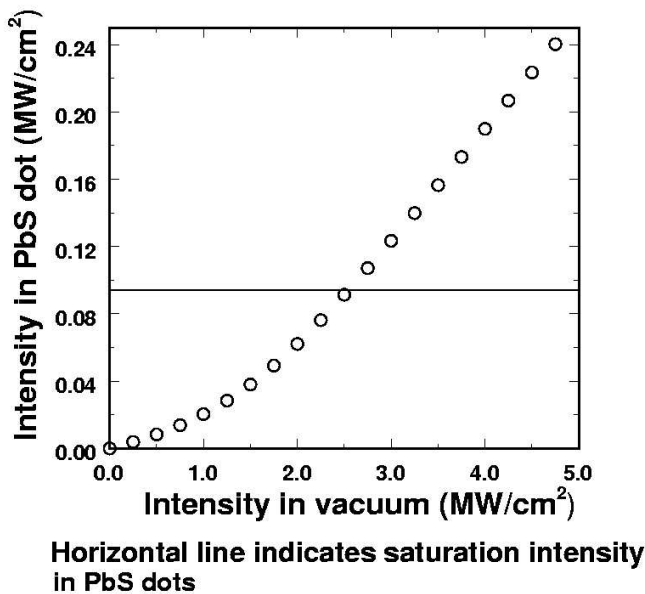


Fig. 5. Squared electric field strength in PbS quantum dots (expressed in intensity units S) vs. the intensity of the incident fields for a $150\mu\text{m}$ film. The data in this figure are the results from the solution of a self-consistent problem.

enhanced relative to the incident intensity (in contrast to being suppressed as in Fig. 5) as was found for metal coated quantum dots in [14]. The fact that the electric field in the quantum dot can be enhanced or suppressed depending on its surroundings implies that a certain flexibility exists which will be beneficial in practical applications.

The predicted behavior of the absorption constant (at the first exciton peak) for PbS vs. the incident field intensity is plotted in Fig. 6. The interesting non-monotonic behavior of the absorption constant is due to dielectric effects. More precisely, as the field in the dot changes, the dielectric properties of the entire structure change. As a result the overall structure can become more or less reflective which, in turn, changes the magnitude of the fields in the matrix.

D. Optical switching applications

One of the motivations of the present study is the desire to use the newly developed, high-quality semiconductor nanocrystals for optical switching applications. By placing quantum dots (or other non-linear optical materials) in a Fabry-Perot cavity, a structure exhibiting optical switching properties can be obtained through careful control of dielectric and structural parameters. Experimental development efforts are underway to construct such a structure. For example, a Fabry-Perot structure was constructed in a ridge wave guide as shown in Fig. 7. This structure was fabricated at the University at Albany, State University of New York by etching away material so as to form two Distributed Bragg Reflectors that define the cavity. The width and height of the structure are approximately $4\mu\text{m}$. This structure was fabricated as a proof of principle demonstration. No measurements of its optical properties are yet available. Further research is needed in order to introduce quantum dots into the cavity.

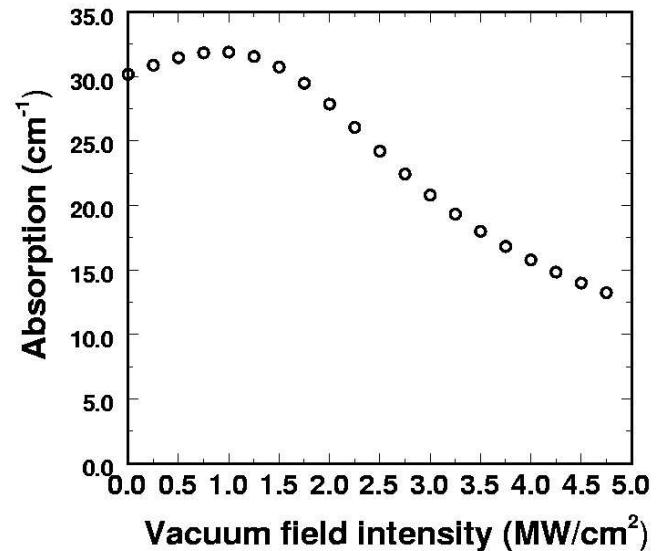


Fig. 6. Absorption coefficient for PbS dots in a thin-film matrix (at the first exciton peak) vs. the intensity of the incident field. The non-monotonic behavior of the absorption coefficient is due to dielectric effects in the film.

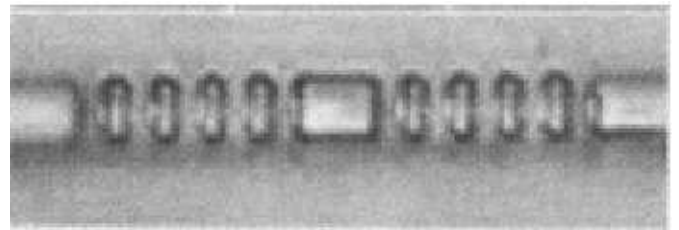


Fig. 7. Fabry-Perot cavity constructed in a ridge wave guide by etching away material so as to form two distributed Bragg reflectors which define the cavity. The width and height of the structure is approximately $4\mu\text{m}$.

Preliminary investigations of switching behavior in a Fabry-Perot cavity containing a non-linear quantum dot based material have been carried out. The analysis was simplified by assuming the multi-layer dielectric structure to be infinitely extended in the lateral dimension. By making this assumption we are able to use the transfer matrix approach for planar films introduced earlier. Obviously this assumption is inappropriate for a structure such as the one depicted in Fig. 7. For a realistic structure a more sophisticated approach such as the Finite-Difference Time Domain (numerical) method will be needed. In any case, the simplified approach will be beneficial in developing insight into the problem.

Simulation results for a quantum dot based Fabry-Perot optical switch are presented in Fig. 8. We assumed the structure to contain CdS quantum dots and use the simple model that was presented earlier. We chose this model as it was fit to experiment so should yield reasonably realistic results. Note, however, that inhomogeneous broadening effects have not been included. Clearly the transmission changes sharply from low ($\approx 10\%$) to high ($\approx 80\%$) as the intensity is increased. The reflectivity also changes abruptly at the same applied field strength. Unfortunately the absorption is quite high. The absorption can be reduced by increasing the detuning away

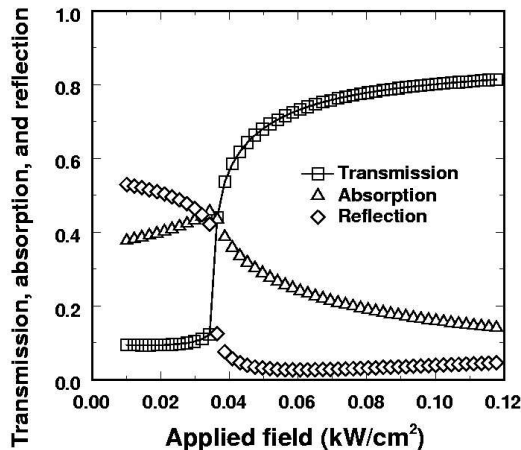


Fig. 8. Simulation results for a structure designed to behave as an optical switch. This simulation assumes the structure to consist of semiconductor quantum dots (CdS) embedded in a Fabry-Perot cavity.

from the resonance at the price of reducing the switching amplitude of the transmission (not shown).

IV. CONCLUSION

This study represents the beginning of a theoretical/experimental effort aimed at utilizing the unique optical properties of high-quality lead-salt based semiconductor nanocrystals for use in non-linear optics applications. We have presented the general framework for treating the optical properties of a multi-layer dielectric structure containing semiconductor quantum dots as the non-linear optical component. The theory assumes a dilute concentration of nanocrystals in a dielectric matrix material and takes the single-dot dielectric function as input.

The simple particle-in-a-sphere model was presented and shown to be inadequate to accurately predict the detailed features of the absorption spectra of PbS and PbSe quantum dots in a matrix as anticipated previously in the literature. An improved model based on the $k \cdot p$ theory or more sophisticated calculation can easily be incorporated into the present theory. The behavior of multilayered optical structures containing a quantum dot based non-linear material was discussed for several cases demonstrating a strong non-linear switching of the transmission.

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