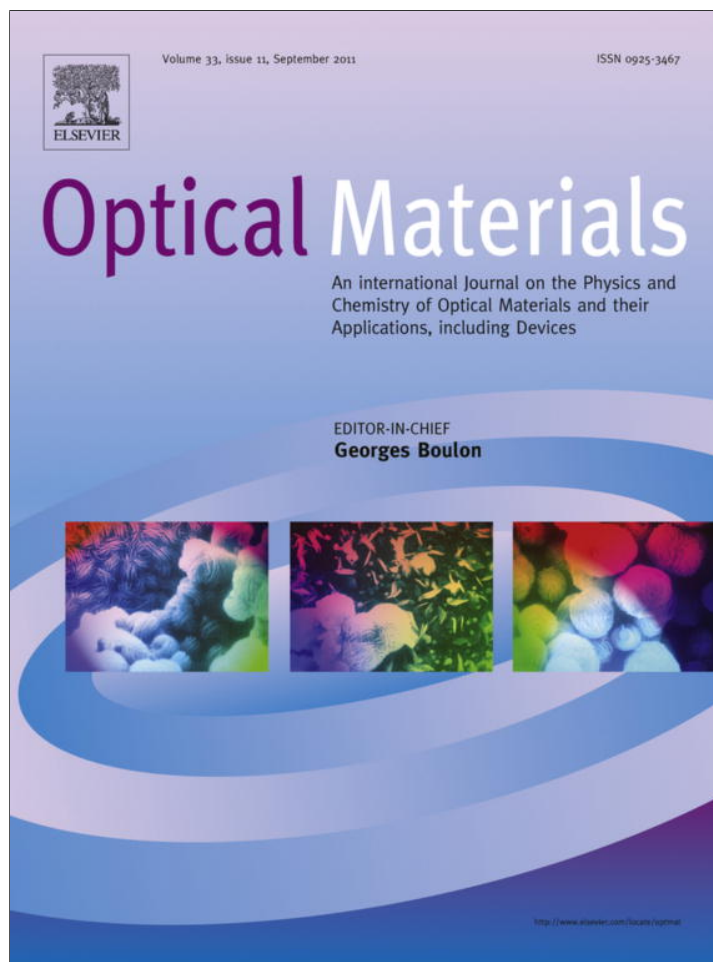


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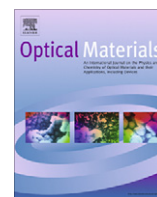
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## Selective IR absorption in molecular nanofilms

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## ARTICLE INFO

## Article history:

Received 15 November 2010

Received in revised form 21 March 2011

Accepted 7 April 2011

Available online 7 May 2011

## Keywords:

Nanofilm

Excitons

Permittivity

Absorption index

Green's functions

## ABSTRACT

We have formulated a microscopic theory of optical properties of ultrathin molecular films (nanofilms), i.e. quasi 2D systems parallel to *XY* planes bounded by two surfaces. Exposure of nanofilms to the external electromagnetic fields has result in creation of excitons – but different than bulk ones. Harmonic exciton states were calculated using the method of two-time, retarded, temperature dependent Green's functions. It has been shown that two types of optical excitations can occur: bulk and surface exciton states. Exciton energy dispersion law shows discrete behavior with non-zero values. Analysis of the dielectric properties of these crystalline systems for low exciton concentration shows that the permittivity strongly depends on boundary parameters and the thickness of the film. In addition, permittivity shows very narrow and discrete dependence of external electromagnetic field frequency, which is a consequence of both resonance and quantum size effects. Influences of boundary conditions on optical characteristics (through analyses of dynamical absorption coefficient) of these nanostructures were specially and in details explored.

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## 1. Introduction

The absorption of photons in dielectric solid can lead to located excitations – Frenkel's excitons, which are responsible for organic crystals optical characteristics [1]. Ultrathin organic (dielectric) films are of potential importance for future electronic compounds and optic devices and they are in the focus of current intensive studies [2]. Theoretical studies of quasi-two-dimensional exciton systems became rather intense, especially in application of device packaging. Electronic components are used today in extreme physical conditions and in that sense, ultrathin dielectric crystalline films could be used as surface layers for electronic component protection [3,4].

In our previous papers [5,6], we investigated properties of ultrathin dielectric films (which were not thicker than a dozen of atomic planes) using a specially adjusted method (Green's functions) for these very discrete crystalline structures. We study the basic physical characteristics of ultrathin molecular crystalline films and one can see that essential properties of these systems arise with per-

turbation conditions which appear *at* and *within* their surface layers [7,8]. We have shown the influence of every boundary parameter (there are five of them) at both microscopic (energy spectra and states) and macroscopic properties (permittivity). We have also found conditions for the occurrence of localized (Tamm's) states at boundary areas and in boundary planes of a nanofilm.

Now we found out that the presence of boundaries and changes in boundary parameters will lead to the presence of discrete absorption and a possibility for a selective absorption of a single line of incoming electromagnetic radiation.<sup>1</sup>

Investigations of exciton response of a symmetrical film to an external electromagnetic excitation are the topic of this paper: theoretical investigations were carried out starting from a standard expression for effective exciton Hamiltonian [1]:

$$H_{\text{ex}} = \sum_{\vec{n}} \Delta_{\vec{n}} P_{\vec{n}}^+ P_{\vec{n}} + \sum_{\vec{n}, \vec{m}} X_{\vec{n}\vec{m}} P_{\vec{n}}^+ P_{\vec{m}} + \sum_{\vec{n}, \vec{m}} Y_{\vec{n}\vec{m}} P_{\vec{n}}^+ P_{\vec{m}}^+ P_{\vec{m}}. \quad (1)$$

where  $P_{\vec{n}}^+$  and  $P_{\vec{n}}$  are creation and annihilation Pauli-operators of exciton at the site  $\vec{n}$  of a crystal lattice,  $\Delta_{\vec{n}}$  is energy of isolated

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<sup>1</sup> The preliminary results of this work are presented at ICOM 2009 (see Ref. [9]).

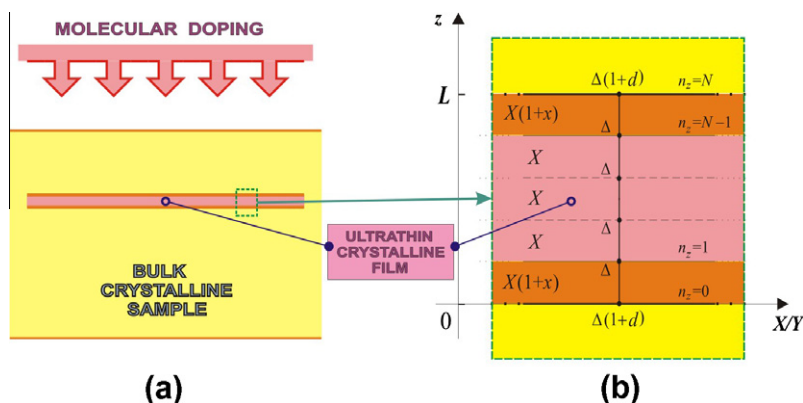


Fig. 1. (a) Schematic presentation of symmetric film-making. (b) Our model of symmetric ultrathin exciton film with boundary perturbation.

exciton in that site, while  $X_{\bar{n}\bar{m}}$  and  $Y_{\bar{n}\bar{m}}$  are matrix elements of dipole–dipole interaction which describe energy of exciton transfer from site  $\bar{n}$  to  $\bar{m}$ . For low exciton concentrations, in harmonic (Bose) approximation [10], and nearest-neighbors approximation (because orbitals overlap only for neighbor molecules), we get:

$$H = \sum_{\bar{n}} \Delta_{\bar{n}} B_{\bar{n}}^{\dagger} B_{\bar{n}} + \sum_{\bar{n}, \bar{z}} X_{\bar{n}\bar{z}} B_{\bar{n}}^{\dagger} B_{\bar{n}+\bar{z}}, \quad (2)$$

where  $B_{\bar{n}}^{\dagger}$  and  $B_{\bar{n}}$  are creation and annihilation Bose-operators of exciton at the site  $\bar{n}$  of a crystal lattice, and  $X_{\bar{n}\bar{z}} \gg Y_{\bar{n}\bar{z}}$  are matrix elements of excitons transfer energy from site  $\bar{n}$  to neighboring site removed for  $\bar{z}$ .

## 2. Model of exciton ultrathin film

Because the most of well-controlled ultrathin crystalline films are made by doping of massive specimens [11,12], nanofilm is formed within the bulk structure (Fig. 1a). In this case, boundary parameters of a film cannot be independent or different from each other. This is why we have a model of a symmetrical nanofilm (in the literature called “cut off” structures, Fig. 1b) whose parameters are changing in following boundaries:

$$\begin{aligned} \Delta_{\bar{n}} &\equiv \Delta[1 + d(\delta_{n_z,0} + \delta_{n_z,N})]; & X_{\bar{n},\bar{n}+\bar{z}} &\equiv X[1 + \chi(\delta_{n_z,0} + \delta_{n_z,N-1})]; \\ &\times X_{\bar{n},\bar{n}-\bar{z}} &\equiv X[1 + \chi(\delta_{n_z,1} + \delta_{n_z,N})], \end{aligned} \quad (3)$$

where parameter  $d \in [-0.2; +0.2]$  defines perturbation at the site of boundary areas, and parameter  $\chi \in [-0.99; +2.0]$  represents perturbation of transfer within boundary planes,<sup>2</sup> along the  $z$ -axis;  $n_z = 0, 1, 2, \dots, N$ .

## 3. Investigation method

Theoretical analysis has been provided by the Green's functions method [1,4–9]. We observe Green's function  $G_{\bar{n}\bar{m}}(t) = \langle\langle B_{\bar{n}}(t) | B_{\bar{m}}^{\dagger}(0) \rangle\rangle \equiv \Theta(t) \langle [B_{\bar{n}}(t), B_{\bar{m}}^{\dagger}(0)] \rangle$ , which satisfies following equation of motion:

$$i\hbar \frac{d}{dt} G_{\bar{n}\bar{m}}(t) = i\hbar \delta(t) \langle [B_{\bar{n}}(t), B_{\bar{m}}^{\dagger}(0)] \rangle + \Theta(t) \langle [[B_{\bar{n}}(t), H], B_{\bar{m}}^{\dagger}(0)] \rangle, \quad (4)$$

After expanding and calculating the commutators [14], in nearest neighbor's approximation next form is obtained:

$$i\hbar \frac{d}{dt} G_{\bar{n}\bar{z}}(t) = i\hbar \delta(t) \delta_{\bar{n},\bar{n}+\bar{z}} + \Delta_{\bar{n}} G_{\bar{n}\bar{z}}(t) + \sum_{\bar{l}} X_{\bar{n}\bar{l}} G_{\bar{l}\bar{z}}(t). \quad (5)$$

Having in mind the Hamiltonian expressions (Eq. (2)), boundary conditions (Eq. (3)), and the Green's equation of motion (Eq. (5)) we can derive the system of  $N + 1$  non-homogenous algebraic-differential equations for the Green's functions (for details, see in [4]):

$$\begin{aligned} G_{n_z, m_z} \left[ \rho - \frac{\Delta}{|X|} d(\delta_{n_z,0} + \delta_{n_z,N}) \right] + G_{n_z+1, m_z} [1 + \chi(\delta_{n_z,0} + \delta_{n_z,N-1})] \\ + G_{n_z-1, m_z} [1 + \chi(\delta_{n_z,1} + \delta_{n_z,N})] \\ = \frac{i\hbar}{2\pi|X|} \delta_{n_z, m_z}, \end{aligned} \quad (6)$$

where

$$\rho = \frac{\hbar\omega - \Delta}{|X|} + F_{xy}; \quad F_{xy} \equiv 2(\cos ak_x + \cos ak_y). \quad (7)$$

The system of Eq. (6) has  $N + 1$  solution for the required Green's functions:

$$G_{n_z} \equiv G_{n_z, m_z} = -\frac{i\hbar}{2\pi|X|} \sum_{v=1}^{N+1} \frac{g_{n_z}(\rho_v)}{\rho - \rho_v}, \quad (8)$$

where  $g_{n_z}(\rho_v)$  are spectral weights (probability) of possible values (states) occurrence:

$$\sum_{n_z=0}^N g_{n_z}^v = 1; \quad \sum_{v=0}^{N+1} g_{n_z}^v = 1; \quad \sum_{n_z=0}^N \sum_{v=1}^{N+1} g_{n_z}^v = N + 1,$$

and

$$\rho_v \equiv \frac{\hbar\omega_v - \Delta}{|X|}; \quad v = 1, 2, 3, \dots, N + 1, \quad (9)$$

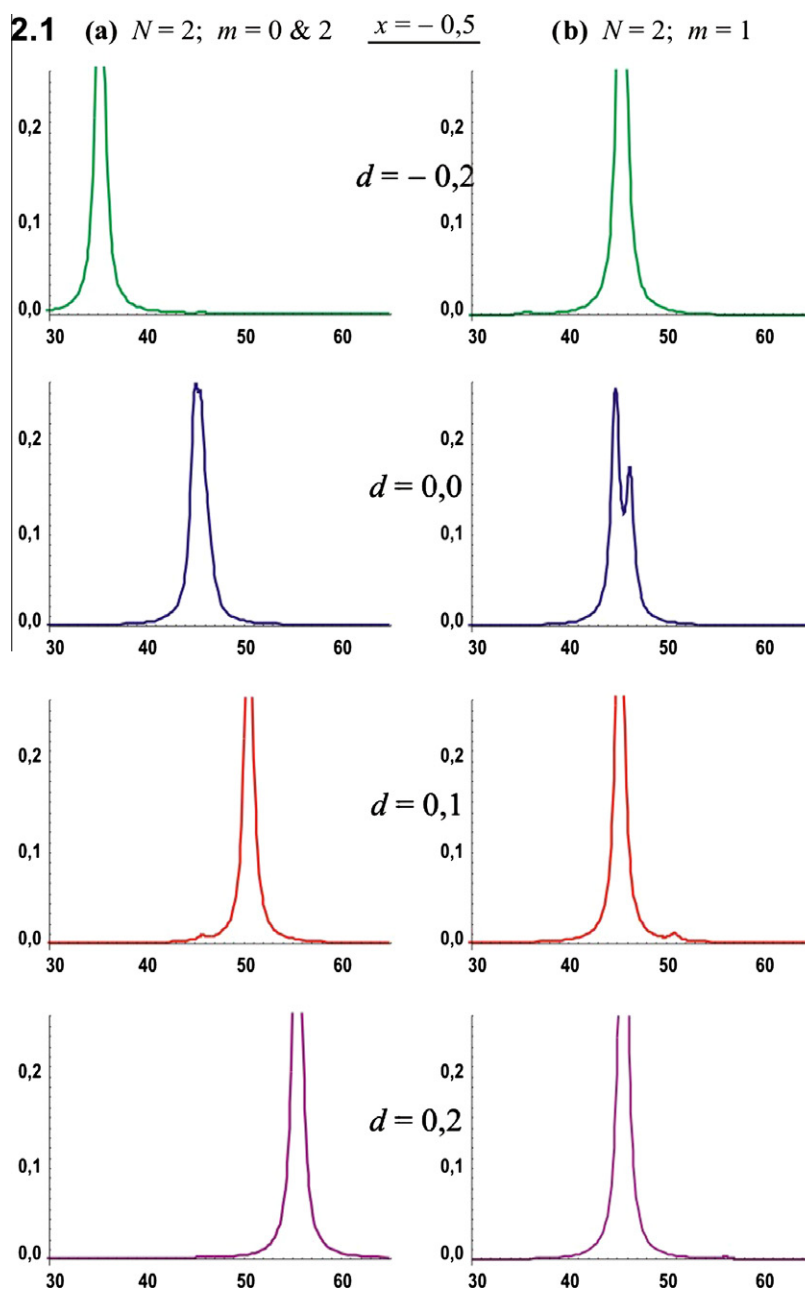
are the Green's function poles, i.e. the roots of determinant of the system of Eq. (6).  $\rho_v$  represents reduced and non-dimensional exciton energy ( $E_v \equiv \hbar\omega_v$ ) also.

Finding these roots is reduced to finding zeros of the determinant of the system of Eq. (6). The determinant, and its zeros, depends on three parameters:  $N$  (i.e., ultrathin film thickness  $L = Na$ ), then  $d$  and  $\chi$  (i.e., the boundary perturbation parameters of exciton system), i.e.

$$\begin{aligned} \text{Det} \equiv \text{Det}_{N,d,\chi}(\rho_v) = 0 \Rightarrow \rho_v = \rho_v(d, \chi); \\ \times v = 1, 2, 3, \dots, N + 1. \end{aligned} \quad (10)$$

In this paper, we decided to analyze the potential of the model's thinnest ultrathin film, and this is a film with two layers ( $N = 2$ ) or with three crystal planes – there are two border ( $n_z = 0$  i 2) and one in the middle of the film ( $n_z = 1$ ). The reason for this choice is that today is possible technologically and technically to produce practically almost monatomic structures on surfaces or substrates [3,4,12]. On the other hand, in a previous paper [15] we found the

<sup>2</sup> The real increase or decrease the parameter  $\Delta_{0/N}$  is up to 20% [3,12,13]. The real increase the parameter  $X_{0,1/N-1,N}$  is up to 200%, while decrease of them is up to 50% [3,12,13].



**Fig. 2.1–2.4.** Dynamical absorption index of perturbed symmetrical two-layered film. Influence of change of parameter  $x$  is shown on all pictures: when the transfer of excitons in border layers is half the value comparing to the bulk, then when there is no change of transfer (ideal film:  $x = 0.0$ ), than for the two times higher border transfer ( $x = +1.0$ ), to the case when that transfer is tripled ( $x = +2.0$ ). On each of these four pictures influences of changes of value of parameter  $d$  are shown (ranging from the case when this parameter is lowered for 20% through the case when value of parameter  $d$  is unchanged comparing with bulk case, to the cases when this parameter is increased for 10% and 20% in the node of border area of film), while values of parameter  $x$  are fixed. Each picture has eight figures: four on left side – which show positions of absorption peaks on border plane of film, while four figures from right-side show positions of absorption peaks in internal plane of film. On each figure, ordinate has relative values of absorption index (comparing with bulk values), while abscise has non-dimensional values of frequencies of external electromagnetic field (defined in paper [15]).

energy spectra and possible exciton states in these nanostructures, as well as its dependence on perpendicular permittivity on the frequency of external electromagnetic field. We found conditions for the occurrence of localized states (critical values of parameters  $d$  and  $x$ )<sup>3</sup> and for occurrence of three resonant permittivity lines in

<sup>3</sup> We have shown the existence outbulk or localized excitation states. Under these conditions the model best suited Tamm surface states, but the physical distribution – these are actually extrinsic surface states [11,16], which occur exactly at the interfaces between two materials (such as a semiconductor or semiconductor–oxide–metal interfaces) as well as on surfaces with defects, where the translational symmetry of the surface is broken.

the IR region. In the present paper we want to find how all of this reflects to the absorption characteristics of ultrathin film, because they are the most important in molecular crystal system that tend to be better luminophores [17].

#### 4. Optical properties of excitonic nanofilm

So, in this way are determined multipole Green's functions, so we can move on to calculate the macroscopic characteristics of the observed molecular film.

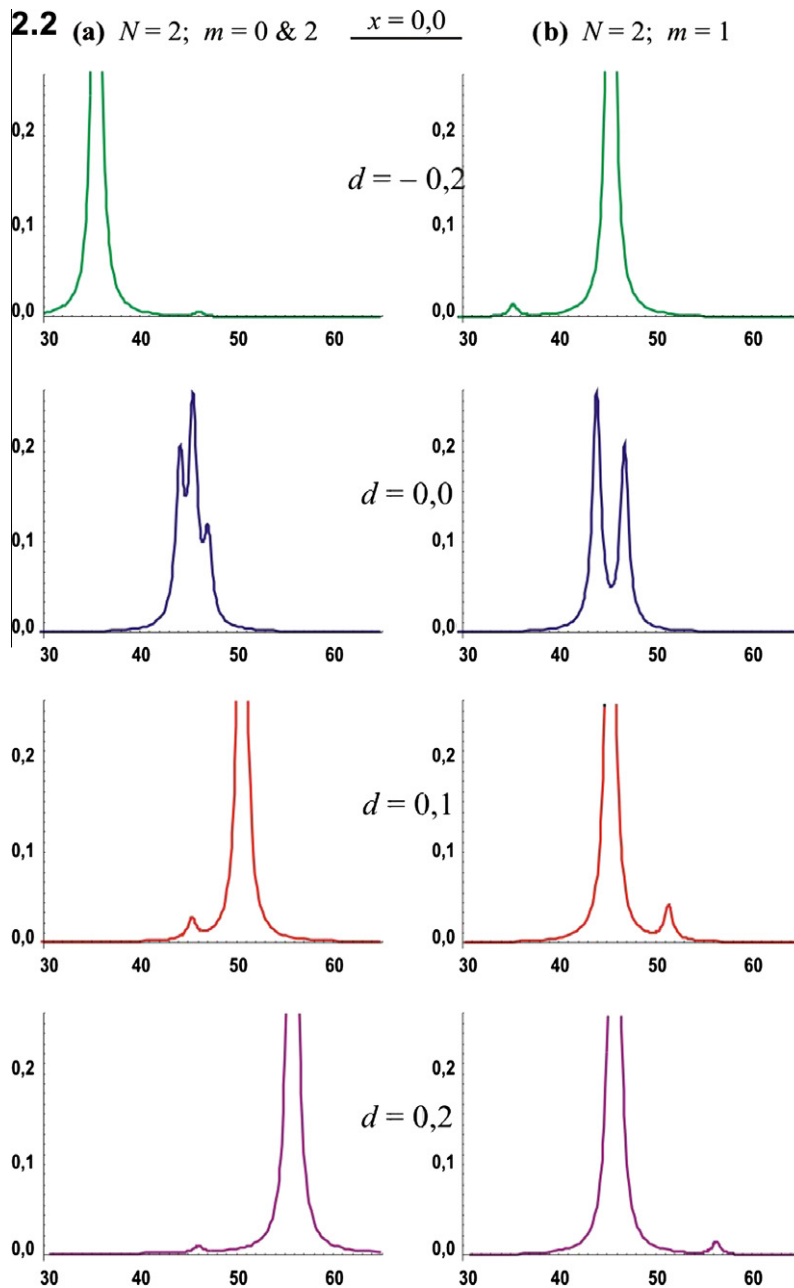


Fig. 2 (continued)

In definition of dynamical permittivity of a film, we will use the general expression [1,4], adjusted to the discrete and strongly space-bounded structures:

$$\varepsilon_{n_z}^{-1}(\omega) = 1 - 2\pi i S [G_{n_z}(\omega) + G_{n_z}(-\omega)], \quad (11)$$

where  $S$  is a parameter corresponding to external perturbation energy and system structure [1,11]. After arranging, we obtain:

$$\varepsilon_{n_z}(\omega) = \left\{ 1 + \frac{2\hbar S}{|X|} \sum_{v=1}^{N+1} g_{n_z}^v \frac{\frac{A}{|X|} + F_{xy} - \rho_v}{\left(\frac{\hbar\omega}{|X|}\right)^2 - \left(\frac{A}{|X|} + F_{xy} - \rho_v\right)^2} \right\}^{-1} \quad (12)$$

This expression represents the dependence of relative dynamical permittivity on the frequency of initial electromagnetic excitation, i.e. the dielectric response of observed symmetrical molecular film on the external electromagnetic field.

In our previous papers [7–9,15] we have proven this dependence and analyzed in detail the effect of boundary parameters  $d$  and  $x$  values to the width and position of the absorption zone – which encompasses resonant permittivity lines (when  $\varepsilon_{n_z}(\omega_r^v) \rightarrow \pm\infty; v = 1, 2, 3, \dots, N$ ). Here we will show the results of our further investigations of optical, i.e. absorption properties of dielectric nanofilms.

The refraction ( $n$ ) and absorption ( $\kappa$ ) indices are usually defined in the literature [18–20] by permittivity term:  $\sqrt{\varepsilon} = n + i\kappa$ . Introducing the complex frequency:  $\omega \rightarrow \omega + i\nu$  in expression for permittivity (12), we get complex permittivity:  $\varepsilon = \varepsilon' + i\varepsilon''$ , where:  $\varepsilon' \equiv \varepsilon'(n_z) = \text{Re}\{\varepsilon\}$  and  $\varepsilon'' \equiv \varepsilon''(n_z) = \text{Im}\{\varepsilon\}$ . Based on this, we can find the expression for absorption index in the following form:

$$\kappa_{n_z}(\omega) = \sqrt{\frac{\varepsilon''}{2} \left[ \sqrt{1 + \left(\frac{\varepsilon''}{\varepsilon'}\right)^2} - 1 \right]}. \quad (13)$$

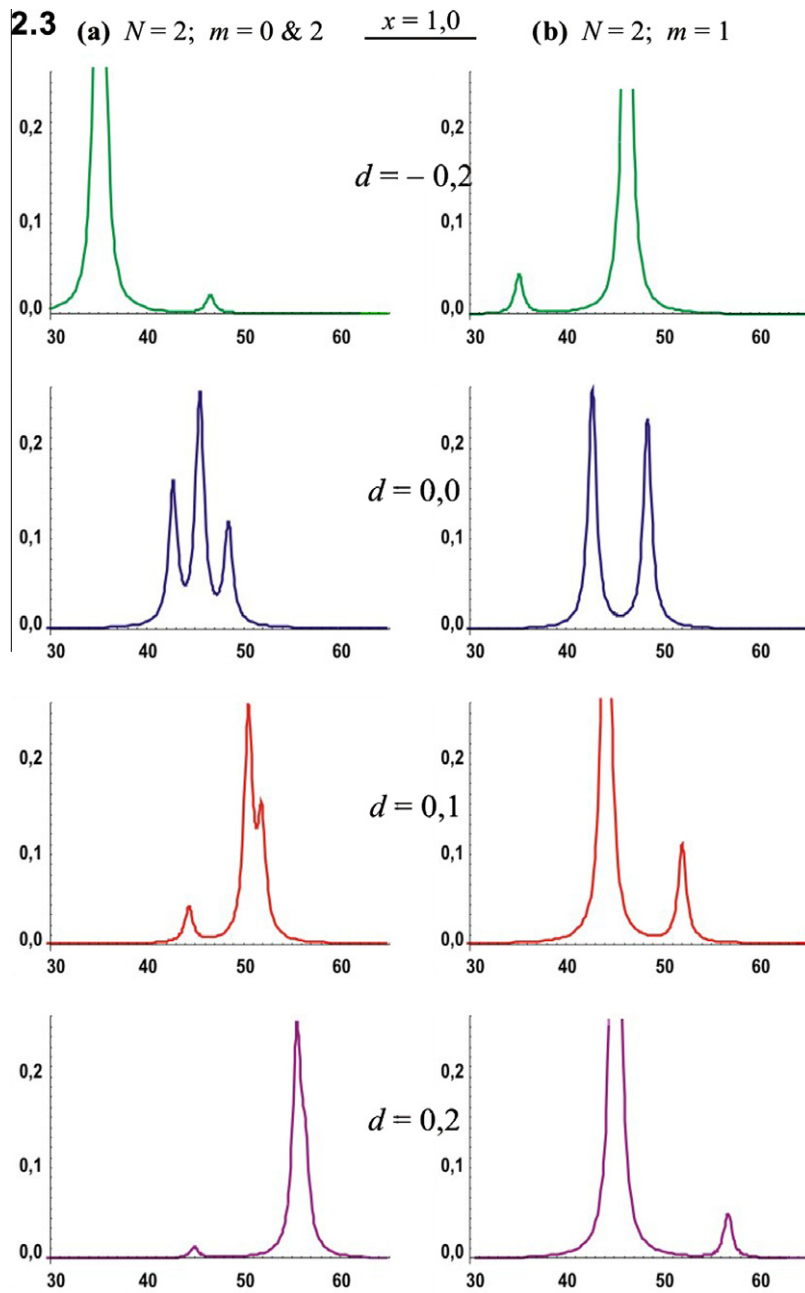


Fig. 2 (continued)

From this and expression (12) one can see that dynamical absorption index as well as permittivity depends on position of film-layer ( $n_z$ ) as well on perturbation at and within ultrathin film.

By numerical calculations, we can analyze absorption index behavior on plane position and on values of boundary parameters. Figs. 2.1–4 are showing the dependence of dynamical absorption index on the reduced energy of external electromagnetic radiation for a symmetrical two-layered (three-lattice-planed) film, as well as their transversal distribution perpendicular to the film (along  $n_z = 0, 1, 2$ , i.e.  $N = 2$ ). These figures/graphics are given by numerical calculations for chosen sets of elementary and boundary parameters of the observed molecular film-structure:  $\Delta/|X| = 50$ ;  $d = -0.2; 0.0; 0.1$  and  $0.2$  as well as  $x = -0.5; 0.0; 0.1; 1.5$ . The minimal number of atomic planes perpendicular to z-axis in our model is 3 ( $N = 2$ ):

$$\begin{aligned}
 & \text{a – (left) boundary planes : } m \equiv n_z \\
 & \quad = 0 \text{ and } 2; \quad \text{b – (right) central inside plane :} \\
 & \quad m \equiv n_z = 1.
 \end{aligned} \tag{14}$$

The number of resonant peaks directly depends on the film width, i.e. on the number of layers  $N$ . General rule is that the number of resonance peaks decreases inside layers of the film, although with influence of perturbation parameters  $d$  and  $x$  that rule can be broken and in that case number of peaks is  $N + 1$ , with possibility of some resonance peaks fading. The wideness of resonant peaks depends directly on spectral weights for corresponding exciton states on each film-layer. There is no resonance when spectral weights are (or tend to) zero.

These figures show occurrence of resonant peaks, whose number and distributions are highly dependent on the boundary per-

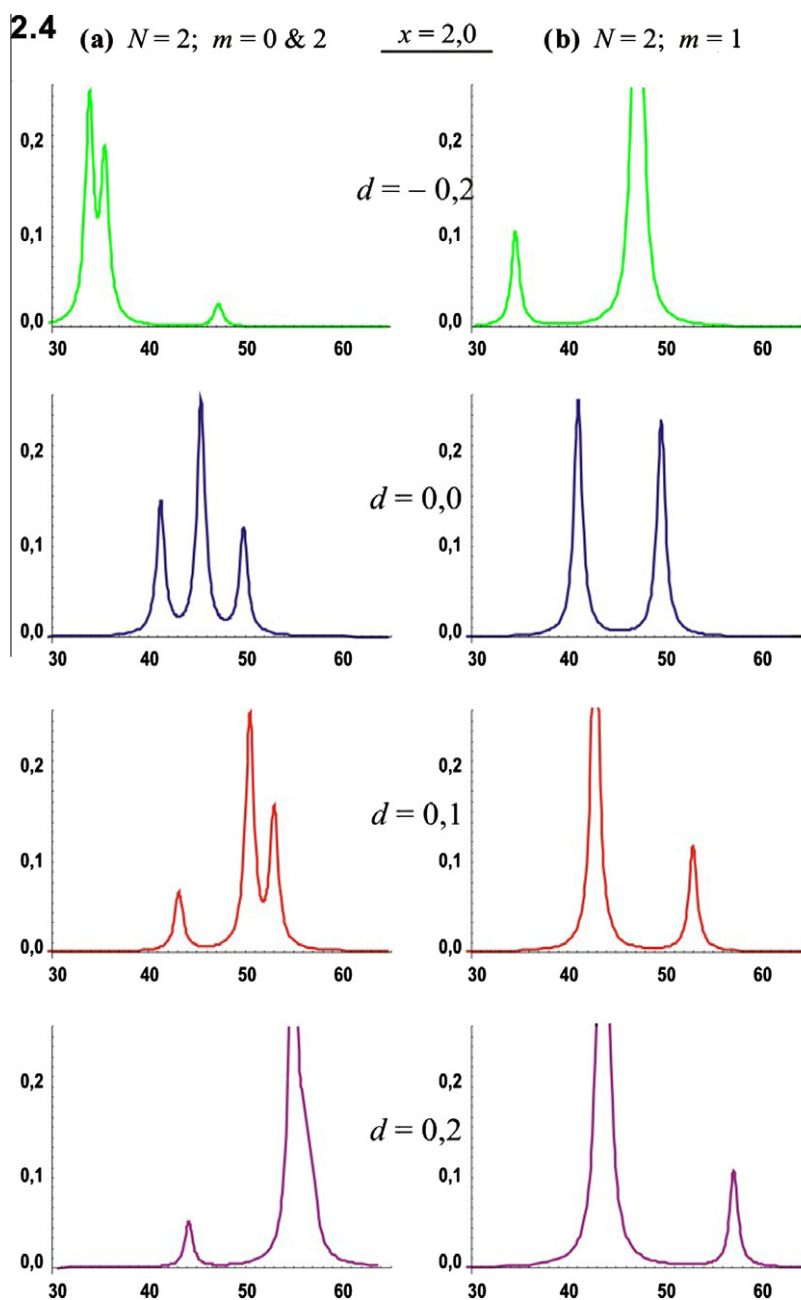


Fig. 2 (continued)

turbation parameters. Increasing the value of parameter  $x$ , the energy zone of resonance broadens, while decreasing it below zero the resonance zone becomes distinctly narrower. Increasing or decreasing the value of parameter  $d$ , the resonance zone shifts towards higher or lower energies, respectively. All these perturbation effects are significant only on the first (i.e. last) film-layer, i.e. on the film boundaries.

The highest change effect is seen at the first and the last layer (left graphs on upper and lower pictures) with highest perturbation, when a zone of absorption peaks is widening towards the higher energies, and where peaks take the largest part of the dielectric response which is moved towards higher energies. The absorption index through the central lattice (all right-side graphs) has associated bulk-properties, i.e. influences of boundary conditions are lower.

One can see from the graphs that 20%  $d$  perturbation gives a significant dielectric response on external electromagnetic field only on border/boundary planes, i.e. upper and lower boundary film surfaces. Inside the film that response is fading, so we show up to  $n_z = 1$ , further dependence will be same. Increasing  $d$ , moves resonance peak to higher energy. From the same graphs one can see too, that positive  $x$  perturbation broadens peak zone and fades internal peaks on first-boundary layer. External peaks vanish on the internal (central) layers. Consequently with space symmetry, we get symmetrical situation for peaks distribution.

## 5. Conclusion

Results of these analyses have shown considerable differences in the dielectric response of excitons between the bulk and

ultrathin film-structures. It is not exclusive consequence of quantum size effects, because influence of exchanged boundary parameters is high as well.

The absorption index of the nanofilm shows unique properties, i.e. the presence of discrete resonant absorption peaks at precisely determined energies (frequencies) of external electromagnetic field, whose number and distribution depends on the number of layers in a film and on perturbation parameters. These properties are giving an advantage to films in comparison to the bulk structures (whose absorption index is continuous within certain range of energy), since in this case films may be used as a kind of filter of external radiation.

Characteristic resonant peaks appear in the dependence of absorption index of ultrathin film on frequency of external electromagnetic field. The peaks width increase with the increasing of exciton spectral weights and there is a possibility to manipulate film dielectric response with suitable selection of boundary perturbation parameters.

All peaks are in infrared region and respond to absorption of corresponding external electromagnetic frequencies. It means that discrete and selective absorption appears.

This result may be explained by experimental facts regarding resonating optical peaks in similar molecular layered nanostructures. In papers [21–23], this was evidenced in perylene chemical compounds and explained by resonating effects at specific unoccupied levels. These effects are manifested by narrow optic absorption in close infrared band. Good agreement in resonating absorption may be attributed and explained by presence of boundary conditions and quantum size effects. Differences in profile of absorption lines appear because we observed only electron line spectra in our work, disregarding oscillatory and rotational contributions.

#### Acknowledgments

This work was partially supported by the Ministries of Sciences and Technological Development of the Republic Serbia and Republic of Srpska. Grant No: OI-171039.

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