JOURNAL OF CRIMINALISTICS AND LAW
ŽURNAL ZA KRIMINALISTIKU I PRAVO
NBP 2019, 24(3): 31–41

Submitted: 2019-09-12
Published: 2019-11-14

UDK: 539.23
620.191.5
doi:10.5937/nabepo24-23159

DISCRETE RESONANT ABSORPTION IN MOLECULAR NANOFILMS

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Abstract: We have analysed the significant different optical absorption of molecular nanofilm crystalline formations. The main reason for this behaviour is in the existence of boundaries on the film structures and we needed to investigate these properties theoretically. In this process, we have done some combinations of (where possible) analytical calculations and (in other cases) numerical calculations. We have calculated the allowed energies of exciton (quasi)particles, and their appearance in nanofilms but in particular direction - perpendicular to the surface of the film. We have analysed why surface exciton localization appears in a film, and after that we calculated dielectric permittivity and optical indices (absorption and refraction). We have established that boundary parameters greatly influence optical phenomena. This influence is resonant, i.e. frequency-related and selectable (depends on what layer is calculated). We even found some particular conditions for the appearance of one (single) absorption line.

Keywords: excitons, ultra-thin films, dielectric permittivity, absorption, refraction.

INTRODUCTION

Nanostructures such as ultra-thin films, quantum wires, or quantum dots have some extraordinary properties when compared to the large (bulk) structures. Obviously this is caused by the confinement of (at least one) dimension of nanostructures. At the same time, nanostructures are usually quite hard to char-

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acterize or to obtain, due to highly expensive equipment or demands for nanostructures without impurities or structural distortions. For those reasons, we need to enforce theoretical research to obtain reliable theoretical models with precise prediction of extremely different properties of these samples with nanometer size thickness. Today usage of nanostructures mainly refers to practical application in electronics where these structures have an important role (Wood et al., 2008).

Today we know that boundary surfaces and processes which take place are very important to change the general properties of these materials. Particularly, in some conditions there arise some very unusual phenomena (which are the consequence of effects of quantization of dimensions (Tringides, Jatochawski and Bauer, 2007)) if we compare them to the macro (dimension) samples (Davison and Steslicka, 1996). We have limited ourselves in studying of exciton sub-systems, mainly knowing that excitons play key role in optical (and optical to electrical) properties in anthracene-type crystals (known as molecular or dielectric crystals) (Agranovich and Ginzburg, 1979). We have analysed films with very small thickness of the film – up to 20 atomic layers, made of dielectric material (e.g. anthracene type), where excitons appear as a result of electrostatic interaction between electrons (inside the dielectric crystal) and electromagnetic field (outside of the crystal). Hereinafter we will show the procedure how to analyse the above mentioned optical values.

MODEL & METHODS

We will use exciton Hamiltonian which is given in harmonic approximation (Agranovich and Ginzburg, 1979; Mirjanić, Kozmidis-Luburić, Marinković and Tošić, 1982):

$$H = \sum_{\vec{l}} D_{\vec{l}} B^{+}_{\vec{l}} B_{\vec{l}} + \sum_{\vec{l},\vec{s}} X_{\vec{l}\vec{s}} B^{+}_{\vec{l}} B_{\vec{s}} \qquad (1)$$

where $B^{+}_{\vec{l}}$ represents exciton (bosonic[2]) operators of creation and $B_{\vec{l}}$ represents Bose operator of annihilation (excitons are Frenkel type – meaning that they are localized on the node of the lattice of the crystal), $D_{\vec{l}}$ is energy of isolated exciton (on node), and $X_{\vec{l}\vec{s}}$ represents exciton transfer between node $\vec{l}$ to node $\vec{k}$. Our model presumes that $D_{\vec{l}}$ is significantly higher than $X_{\vec{l}\vec{s}}$ (in a factor of ~10^2 times).

Among various ways of solving, theoretical analysis could be implemented by the method of Green functions (Rickayzen, 1980; Mahan, 1990). Let us observe

$$G_{\vec{l}\vec{s}}(t) = \left\langle \left\langle B_{\vec{l}}(t) \middle| B^{+}_{\vec{s}}(0) \right\rangle \right\rangle,$$

where the above function is in accordance with the appropriate equation of motion:

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[2] Exact representation of the excitonic Hamiltonian includes Pauli operators, but it can be shown that the Bose operators in the first neighbor conditions can replace this type of operators.

$$i\hbar\frac{d}{dt}G_{\vec{l}\vec{s}}(t)=i\hbar\delta(t)\delta_{\vec{l}\vec{s}}+\mathrm{D}_{\vec{l}}G_{\vec{l}\vec{s}}(t)+\sum_{\vec{j}}X_{\vec{l}\vec{j}}G_{\vec{j}\vec{s}}(t) \tag{2}$$

To obtain Green's function in inverse space (of the wave vectors space) we have to do full Fourier transformations through time and space. After finding the poles of Green's function, we obtain the allowed energies of the excitons:

$$\hbar\omega_{\vec{k}}=\mathrm{D}+2X_x\cos\, a_xk_x+2X_y\cos\, a_yk_y+2X_z\cos\, a_zk_z\,, \tag{3}$$

and, when we replace terms: $-|X|\equiv X_x=X_y=X_z$ and $a\equiv a_x=a_y=a_z$, the equation (3) could be written in non-dimensional form:

$$E_{\vec{k}}\equiv\frac{\hbar\omega-\mathrm{D}}{|X|}=R_{xy}+S_z;$$
$$R_{xy}\equiv 2\cos ak_x+2\cos ak_y;\quad S_z\equiv 2\cos ak_z. \tag{4}$$

One typical result of exciton dispersion law for bulk is presented in the graph in Figure 1.

In literature the universal expression for dielectric permittivity is well known (Dzialoshinski and L. P. Pitaevski, 1959; Pelemiš, Škipina, Vučenović, Mirjanić and Šetrajčić, 2008):

$$\varepsilon(\omega)=\frac{1}{1-2\pi iF\left[G(\omega)+G(-\omega)\right]} \tag{5}$$

where F represents one general term related only to internal structure. If we calculate Green's functions and then substitute in (5), we obtain dynamic permittivity of bulk:

$$\varepsilon^{-1}=1+2F\frac{\omega_{\vec{k}}}{\omega^2-\omega_{\vec{k}}^2} \tag{6}$$

How permittivity depends on frequency of external radiation is shown in Figure 2.

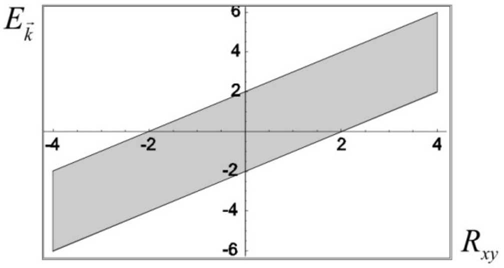

Figure 1. *Allowed energies of excitons in bulk*

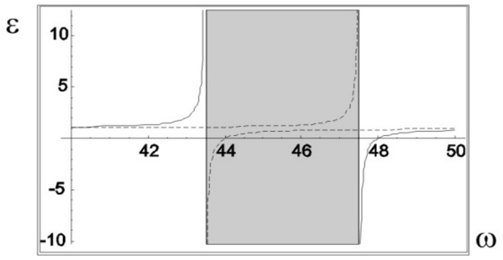

Figure 2. *Dielectric permittivity in bulk*

Dielectric permittivity is a complex value, i.e. that value consists of the real and imaginary parts: $\varepsilon(\omega)=\varepsilon^{/}(\omega)+\varepsilon^{/}(\omega)$. The terms $\varepsilon^{/}$ and $\varepsilon^{//}$ are related via Kramers–Kronig relations, i.e. with refraction and absorption indices.

James Maxwell showed that the dielectric permittivity is square of the refraction index: $\varepsilon(\omega)\equiv\eta^{2}(\omega)$, where the refraction index has a complex value: $\eta=n+i\kappa$. Then we can easily obtain: $\varepsilon^{/}(\omega)=n^{2}-\kappa^{2}$; $\varepsilon^{//}(\omega)=2n\kappa$.

Furthermore, we can calculate the terms for the absorption and refractive index expressed through terms $\varepsilon^{/}$ and $\varepsilon^{//}$ in the following form:

$$\kappa(\omega)=\sqrt{\frac{2}{\varepsilon^{/}(\omega)}\left\{\sqrt{1+\left[\frac{\varepsilon^{/}(\omega)}{\varepsilon^{//}(\omega)}\right]^{2}}-1\right\}};\quad n(\omega)=\sqrt{\frac{2}{\varepsilon^{/}(\omega)}\left\{\sqrt{1+\left[\frac{\varepsilon^{/}(\omega)}{\varepsilon^{//}(\omega)}\right]^{2}}+1\right\}}. \tag{7}$$

The above indices and their dependence from external radiation are shown in Figures 3 and 4.

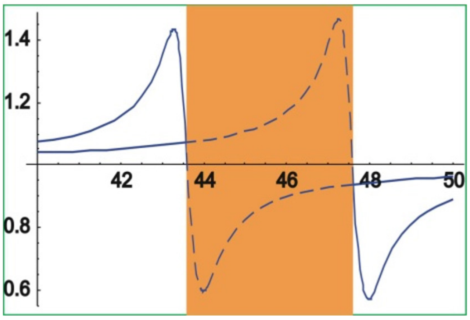

Figure 3. *Absorption index of bulk-crystal*

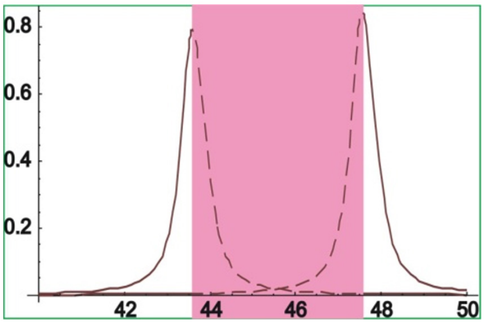

Figure 4. *Refraction index of bulk-crystal*

With dimensionally unlimited materials, the allowed energies could take continuous values in some particular extent of the allowed energies. This defines the dielectric response of bulk. That means that the absorption zone lays within certain interval of energies that bulk "swallows" (i.e. that zone of frequencies of external electromagnetic field is completely absorbed by the bulk).

DIELECTRIC FILM

Film-structure presents alternately arranged layers, limited by two parallel surfaces (Cottam and Tilley, 1989; Šetrajčić et al., 2008). Theoretically we could presume that film is in *XY* (or plane) direction unlimited, while perpendicular to *XY*, (or along z-direction) a film has the final number (*N*) of planes, with film width equal to $L = Na$ ($N < 10$).

We will observe ultrathin (or nano) crystal film (Figure 5), made by controlled deposition of dielectric material (Šetrajčić et al., 2008). Exchanges of the energies between nodes and between borders (i.e. between $l_z = 0$; $l_z = N$ and $l_z = 1$; $l_z = N - 1$) are perturbed, due to the existence of border surfaces (Pelemiš et al., 2008; Šetrajčić et al., 2008; Šetrajčić, Vučenović, Mirjanić, Markoski and Šetrajčić, 2008; Šetrajčić, et al., 2005; Pelemiš et al., 2009):

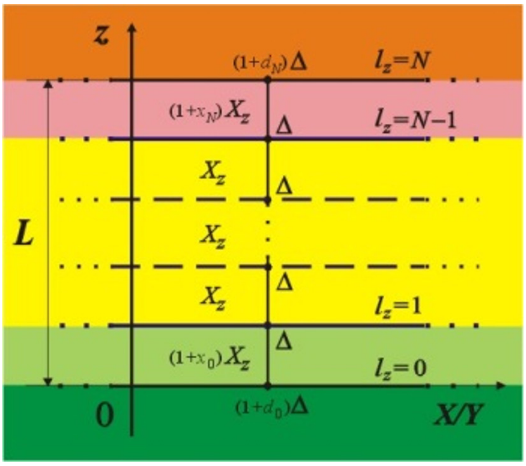

Figure 5. *Dielectric nanofilm model*

$$\mathsf{D}_{\vec{l}} \equiv \mathsf{D}\left[1+\left(d_0\delta_{l_z,0}+d_N\delta_{l_z,N}\right)\right]\ ;$$

$$X_{\vec{l},\vec{l}+\vec{\lambda}} \equiv X\left[1+\left(x_0\,\delta_{l_z,0}+x_N\delta_{l_z,N-1}\right)\right]\ ;$$

$$X_{\vec{l},\vec{l}-\vec{\lambda}} \equiv X\left[1+\left(x_0\delta_{l_z,1}+x_N\delta_{l_z,N}\right)\right],$$

where term *d* shows how energy is perturbed on sites on border planes, and term *x* shows how energy transfer between layers on boundaries is perturbed.

As in the bulk, we analyse exciton behaviour in film, by applying Green's functions with the same manner as in the bulk. However, here instead of full spatial – we must perform partly Fourier transformations[3]. The detailed procedure could be found in (Šetrajčić et al., 2008; Šetrajčić et al., 2008; Šetrajčić, et al., 2005; Pelemiš et al., 2009). One of the interesting steps in calculations is the following relation:

$$\begin{gathered}G_{l_z,s_z}\left[\rho-\frac{\mathsf{D}}{|X|}\left(d_0\delta_{l_z,0}+d_N\delta_{l_z,N}\right)\right]+G_{l_z-1,s_z}\left[1+\left(x_0\delta_{l_z,0}+x_N\delta_{l_z,N-1}\right)\right]+\\+G_{l_z+1,s_z}\left[1+\left(x_0\delta_{l_z,1}+x_N\delta_{l_z,N}\right)\right]=\frac{i\hbar}{2\pi|X|}\delta_{l_z,s_z}\ ,\end{gathered}\tag{8}$$

where we introduce the following denotation:

$$\rho=\frac{\hbar\omega-\mathsf{D}}{|X|}+2\cos ak_x+2\cos ak_y\ .$$

The expression (8) is in fact system[4] of *N*+1 equations for Green's functions. We could write (8) in matrix form, and calculate the determinant of that matrix. In order to find the allowed energies of excitons in film, it is enough to find the poles of the determinant (Šetrajčić et al., 2008; Šetrajčić et al., 2008; Šetrajčić, et

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[3] Film is confined along z-axe. This is the reason for partial Fourier transformation.

[4] This system is algebraic and not homogenuos.

al., 2005; Pelemiš et al., 2009). In such a way we avoid to calculate the exact Green's function, but we get N+1 solutions $\rho \equiv \rho_\nu;\;\; \nu = 1,2,\ldots,N+1$.

If we chose to transfer to non-dimensional form of exciton dispersion law, we get the values of reduced energies:

$$E_\nu = \frac{\hbar\omega - \Delta}{|X|} \equiv R_{xy} - \rho_\nu ,$$

and as one can see the above relation depends on the function $R_{xy} \equiv 2(\cos ak_x + \cos ak_y)$.

The dispersion laws are presented in Figure 6 for symmetrical perturbed film, made of 5 layers. The allowed energy levels of excitons in film are drawn with full lines, and between dotted lines lay energy continuums for excitons in bulk. First, one can see two important facts: zero energy exciton does not exist in film, and those energies are all discrete. One fact that is more interesting is that, when film is unperturbed, the number of possible allowed exciton states (or energies) is equal to the number of film layers.

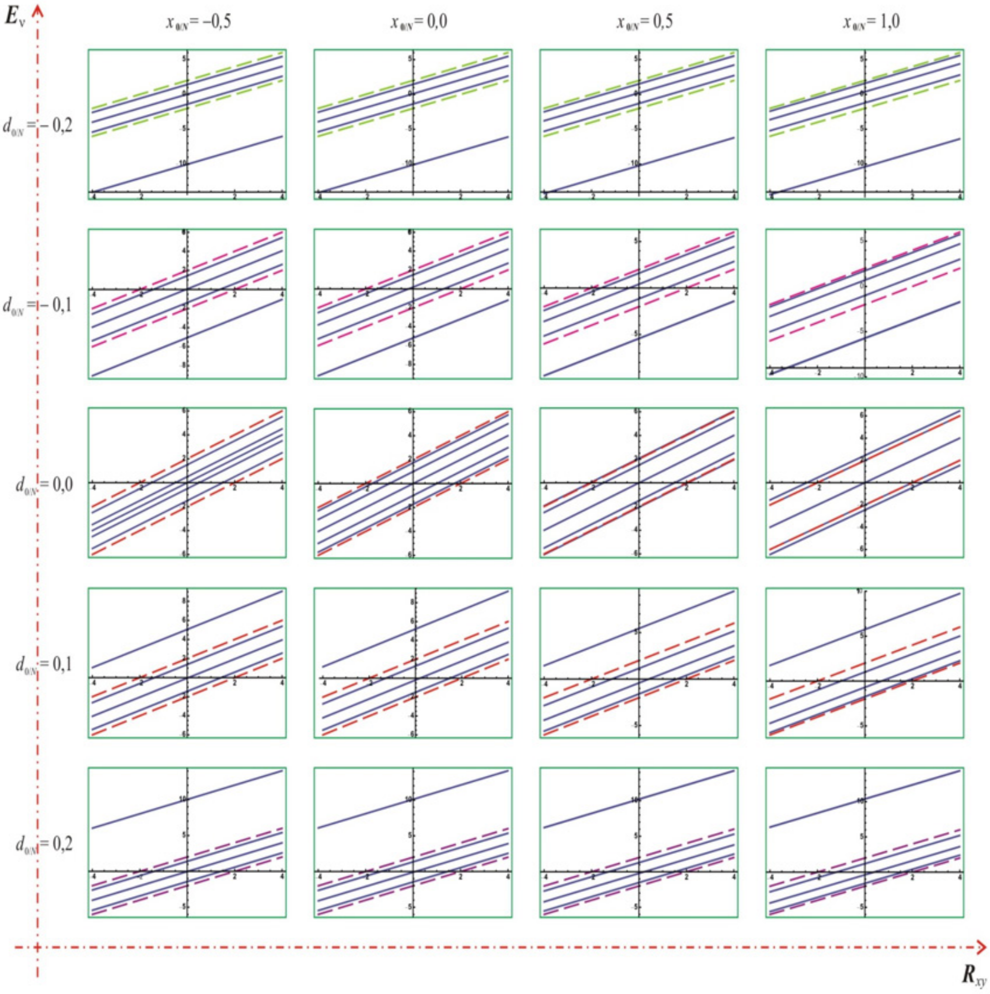

Figure 6. *Dispersion law of d- and x- perturbed five-layer film*

However, with perturbation acting the energy spectrum is somewhat changed. One can see that with increased d – perturbation, the allowed energies spread out of the (dashed) bulk borders. That is happening in such a manner that one energy level (or even two) has higher energy (Figure 6) compared with bulk. These are well known as localized or Tamm's states (Agranovich and Ginzburg, 1979). Furthermore it is evident that with increased x parameter the spectrum expands. That is happening in such a manner that two energy levels (or four) go out of the bulk borders.

We have analysed one specific film structure in this paper, which we call "ideal", where the applied perturbation is symmetrical one, and with further analysis it could be showed that localized levels overlap each other. That condition could happen only for d-perturbation.

Spectral weights of individual Green's functions represent the probability of finding (or appearance of excitons) in the film. To calculate spectral weights, we must rewrite the system (8) in the following form:

$$\hat{D}_{N+1}\tilde{G}_{N+1} = \hat{K}_{N+1} \tag{9}$$

where $\hat{D}_{N+1}$, $\tilde{G}_{N+1}$ and $\hat{K}_{N+1}$ are system matrices, the vectors of Green's function and the vector made of Kronecker's deltas, respectively. When we operate from the left side with inverse matrix $\hat{D}^{-1}_{N+1}$ in (9), we can calculate Green's functions. In the obtained functions the numerators are actually spectral weights $g_{l_z}(\rho_\nu)$, for each exciton state ρ_ν (Pelemiš et al., 2008; Šetrajčić et al., 2008; Šetrajčić et al., 2008; Šetrajčić, et al., 2005; Pelemiš et al., 2009):

$$G_{l_z} = -\frac{i\hbar}{2\pi\left|X\right|}\sum_{\nu=1}^{N+1}\frac{g_{l_z}\left(\rho_\nu\right)}{\rho-\rho_\nu} \tag{10}$$

From the general expression (5) we can calculate dynamic permittivity of the film, knowing that Green's functions, and the permittivity consequently, depend on crystallographic planes l_z:

$$\frac{1}{\varepsilon_{n_z}\left(\omega\right)} = 1-2\pi i\,F\;\left[G_{n_z}\left(\omega\right)+G_{n_z}\left(-\omega\right)\right] \tag{11}$$

Including (10) into (11), we obtain:

$$\varepsilon^{-1}_{n_z}\left(\omega\right) = 1-\frac{\hbar F}{\left|X\right|}\sum_{\nu=1}^{N+1}\sum_{s=+,-}\frac{g^{\nu}_{n_z}}{\rho_s-\rho_\nu} \tag{12}$$

where: $\rho_\pm = \frac{\mp\hbar\omega-\mathrm{D}}{\left|X\right|}+2\cos ak_x+2\cos ak_y$. Finally, (12) is transformed to:

$$\varepsilon_{n_z}\left(\omega\right) = \left\{1-\frac{2\hbar F}{\left|X\right|}\sum_{\nu=1}^{N+1}g^{\nu}_{n_z}\frac{\rho_\nu-\frac{\Delta}{\left|X\right|}-2\left(\cos ak_x+\cos ak_y\right)}{\left(\frac{\hbar\omega}{\left|X\right|}\right)^2-\left[\rho_\nu-\frac{\Delta}{\left|X\right|}-2\left(\cos ak_x+\cos ak_y\right)\right]^2}\right\}^{-1} \tag{13}$$

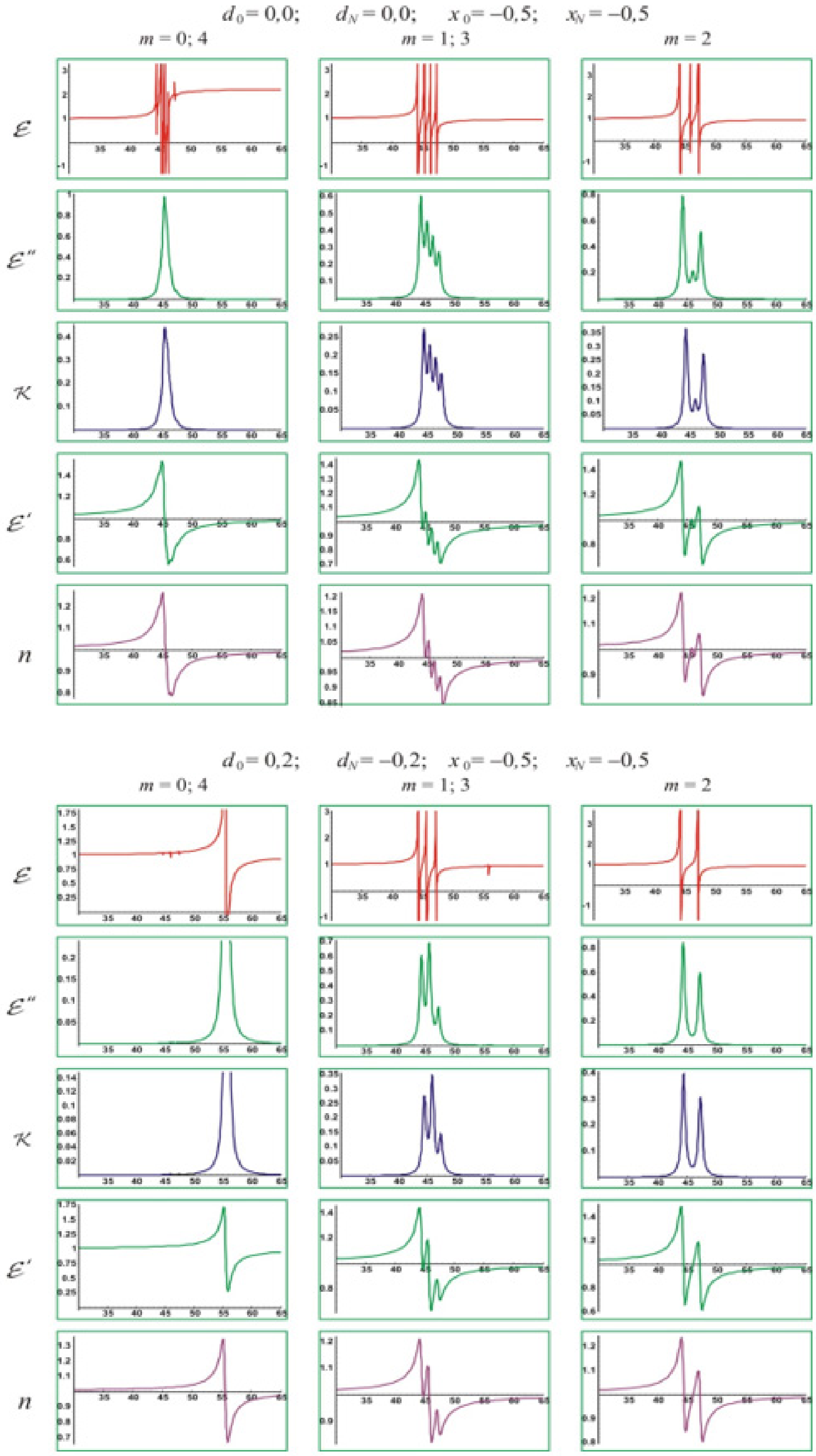

Figure 7. *Optical indices (permittivity, absorption and refraction) for five-layer film*

Expression (13) represents dielectric behaviour, i.e. how symmetrical molecular film reacts to the electromagnetic field. If we again link (13) with complex permittivity $\varepsilon_{n_z}(\omega)=\varepsilon^{/}_{n_z}(\omega)+i\varepsilon^{//}_{n_z}(\omega)$, we can express and calculate optical indices of absorption and refraction for the film:

$$\kappa_{n_z}(\omega)=\sqrt{\frac{\varepsilon^{/}_{n_z}(\omega)}{2}\left\{\sqrt{1+\left[\frac{\varepsilon^{//}_{n_z}(\omega)}{\varepsilon^{/}_{n_z}(\omega)}\right]^2}-1\right\}}\,;\qquad n_{n_z}(\omega)=\sqrt{\frac{\varepsilon^{/}_{n_z}(\omega)}{2}\left\{\sqrt{1+\left[\frac{\varepsilon^{//}_{n_z}(\omega)}{\varepsilon^{/}_{n_z}(\omega)}\right]^2}+1\right\}}\,. \tag{14}$$

Figure 7 shows the prediction how a 5-layered film will optically react when we put it in an electromagnetic field.

When we change parameter Δ by perturbation of d_0 (or d_N) relative permittivity is changing and this is presented in Figure 7. One can see that the number of resonant peaks (those relative frequencies where $\varepsilon\,(\omega_r) \to \pm\infty$) is not equal on every atomic plane. That number is function of the number or position and of the value of perturbation $d_{0/N}$. However, the dominant resonant peak is at the borders of the surface of the film ($n_z = 0$ or 4) under increased perturbation. We could already expect this result, while spectral weights (Šetrajčić et al., 2008) are the highest ones, i.e. they show that excitons would be localized on film surfaces.

One interesting result is mirror symmetry of the first and the last row ($d_{0/N} = -\,0{,}2$; $d_{0/N} = +\,0{,}2$), or the second and the fourth ($d_{0/N} = -\,0{,}1$; $d_{0/N} = +\,0{,}1$) in Figure 7.

The analysis of how perturbation factor X influences permittivity was conducted earlier in (Škipina et al., 2011; Šetrajčić-Tomić, Rodić, Šetrajčić, Sajfert and Šetrajčić, 2018), where we demonstrated perturbation parameter d is evidently much "stronger" where the whole energy spectrum "shifts". Here we focused only on the extreme case (where $x = 1{,}0$).

Every layer in film has different number of resonant lines: this number is at least one (graph below), and it could be up to five (graph on the top). This number strongly depends on the magnitude of the perturbations parameters of the boundary surfaces (to be exact, on boundary layer) of the crystalline film. Localized states are the consequence of certain threshold boundary parameters.

The second row of graphics displays dependence on the imaginary part of relative permittivity, while the third row displays dependence of the index of absorption. It is evident that the behaviour of these two values is almost identical, because of $\kappa \approx \sqrt{\varepsilon}$. Therefore, the position of absorption peaks remains the same, but may vary in size.

When we compare permittivity with absorption, we see that the number of absorption peaks is reduced. Conclusion is that, in that case, some (dominant) frequencies were absorbed.

The real part of the reduced permittivity is shown in the fourth line and in the fifth the refractive index dependence on the reduced frequency of external electro-magnetic field. It can easily be seen that their arrangement is equal, with resonant peaks at the same frequencies, only with different sizes. This could be anticipated regarding relation $n \approx \sqrt{\varepsilon}$. In all of the displayed graphics, it can be seen that in the region where we see increment of absorption - the refractive index is decreasing. Where the absorption index is rapidly changing (toward either lower or higher values), we find the singularities in the refraction of the film. This result is in good agreement with the Kramer's theory, but we must notice that this rapid change occurs only on the "tails" of the absorption zone.

CONCLUSION

In this paper we have analysed the differences between nano-structured film and macroscopic samples (bulk), regarding the allowed excitons energies (dispersion law) and optical properties (dielectric response). These differences are the consequences of film confinement perpendicular to film surface (or along one direction) and different boundary conditions represented through the perturbation parameters on border film surfaces and border layers. We could summarize the analysis in some conclusions remarks:

- Allowed energies of excitons in symmetrical film take discrete values. Without any perturbations (ideal film) the number of allowed energies will be exactly the same as the number of layers in film.
- With some additional interaction on the border of the film (interaction between film and substrate or the surrounding environment) excitons will have higher energies, and this results in shifting the whole spectrum toward higher energies. Other types of perturbation in energy transfer will increase their values and energy spectrum will expand, i.e. excitons will have both lower and higher energies.
- Appearance of Tamm's (or localized) states is highly possible with the increase of perturbations.
- Ultrathin films, under some characteristic perturbations, could be used as very narrow and almost discrete absorber. Energies which define such dielectric response are tuneable, i.e. directly depend on perturbations (interaction of the ultrathin film and environment) and structural characteristic of the film (layers number). This is potentially the most desirable characteristic and advantage of the film comparing with bulk.

ACKNOWLEDGMENTS

These researches were partially supported by the Ministry for Scientific and Technological Development, Higher Education and Information Society of Government of Republic of Srpska (grant No. 19/6-020/961-21/18 and 19/6-020/961-35/18).

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