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Confinement Effects on Absorption of Molecular Crystalline Nanofilms

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INTRODUCTION

Based on the formed model of nanofilm crystal structures, changes of optical properties due to the presence of borders for the case of symmetrical ultrathin films are theoretically investigated in this paper. Influence of five border parameters on the occurrence of localized exciton states is examined, as well as their relation with the effects of discretization and selection of resonant absorption of present electromagnetic radiation. Used combined analytical-numerical calculation to find the allowed energy states of excitons and their spatial distribution (per layers) along the axis perpendicular to surface planes. We determined permittivity for the observed models of these ultrathin dielectric films and explored the influence of boundary parameters on the occurrence of discrete and selective absorption.

EXCITONS IN MOLECULAR STRUCTURES

In this theoretical research of optical properties of nanostructure materials we have to start from the assumption that excitons are generated in materials as response on the external electromagnetic field. Although excitons are not the only (quasi) particles that can be find in nanostructures when external electromagnetic field is turned on, this statement is satisfactorily correct if we use molecular crystals. The Hamiltonian is:

$$H = \sum_{\vec{n}} \Delta_{\vec{n}} B^{+}_{\vec{n}} B_{\vec{n}} + \sum_{\vec{n},\vec{m}} X_{\vec{n}\vec{m}} B^{+}_{\vec{n}} B_{\vec{m}} .$$

We use Green's function method and corresponding equation of motion

$$G_{\vec{n}\vec{m}}(t) \equiv \left\langle\left\langle B_{\vec{n}}(t) \middle| B^{+}_{\vec{m}}(0)\right\rangle\right\rangle;$$

$$i\hbar \frac{d}{dt} G_{\vec{n}\vec{m}}(t) = i\hbar\delta(t)\delta_{\vec{n}\vec{m}} + \Delta_{\vec{n}} G_{\vec{n}\vec{m}}(t) + \sum_{\vec{l}} X_{\vec{n}\vec{l}} G_{\vec{l}\vec{m}}(t) ,$$

in approximation of nearest neighbor, but including dimensional restrictions related to configuration and internal organization of nanostructure, in particular we would observe ultra thin films. The next step is transition from direct space to *k*-space, i.e. performing the time and space Fourier's transformations [3,5]. Advantage of this transition is direct and elegant calculation of energy dispersion law.

DIELECTRIC PERMITTIVITY AND OPTICAL PROPERTIES

Using the expression for Green's functions and Dyaloshinski-Pitaevski relation, we can obtain following expression:

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

which represents dependence of relative dynamic permittivity on energy or frequency of initial electromagnetic excitation, i.e. dielectric response of the observed symmetrical molecular film to external electromagnetic field.

The refraction (n) and absorption (κ) indices are usually defined in the literature by permittivity term: $\sqrt{\varepsilon} = n + i\kappa$. Introducing the complex frequency: $\omega \to \omega + i\nu$ in expression for permittivity, we get complex permittivity: $\varepsilon = \varepsilon' + i\varepsilon''$. Based on this, we can find the expression for absorption and refraction indices in the following form:

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

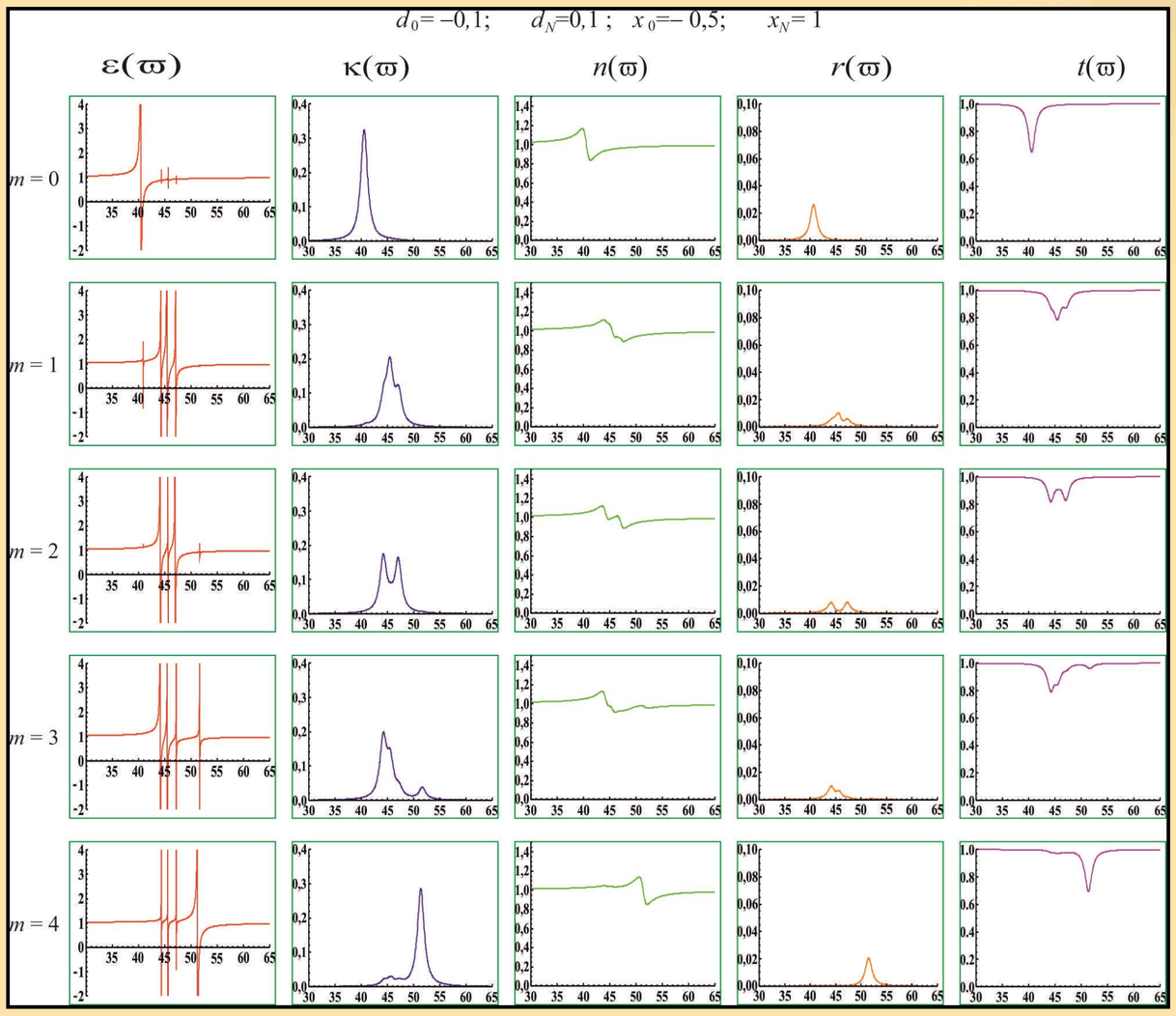
In order to determine optical properties of the medium n and κ, we will observe the diffraction of light from the flat surface of the object. Apart from this, reflection and transparency indices are also very important characteristics of the sample. If light falls vertically to the surface, the index of reflection r and tranparent index τ is defined using the following expression [13–15]:

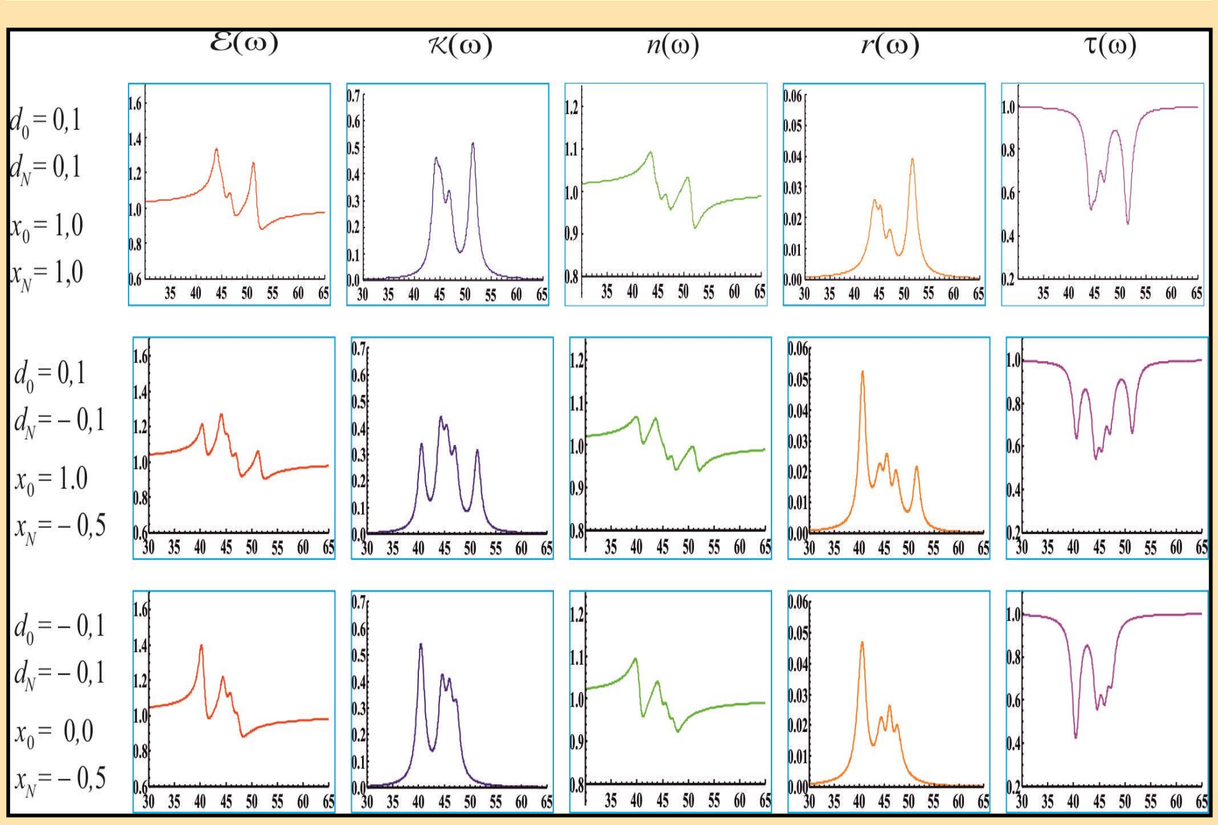
$$\tau_{n_z}(\omega) \equiv 1 - \kappa_{n_z}(\omega) - r_{n_z}(\omega); \qquad r_{n_z}(\omega) = \frac{\left(n_{n_z} - 1\right)^2 + \kappa^2_{n_z}}{\left(n_{n_z} + 1\right)^2 + \kappa^2_{n_z}}.$$

For whole film

$$n_F(\omega) = N\left[\sum_{n_z} n^{-1}_{n_z}(\omega)\right]^{-1}; \qquad \kappa_F(\omega) = \sum_{n_z} \kappa_{n_z}(\omega);$$

$$r_F(\omega) = 0; \qquad \tau_F(\omega) = 1 - \kappa_F(\omega).$$





CONCLUSION

Optical, i.e. absorption, refraction, reflection and transparent properties of these nanostructures demonstrate very narrow or discrete characteristics, where relative dielectric permittivity and optical indices dependence from external electromagnetic field indicate existence of discrete resonant piques, which number is, in general, equal to the number of atomic layers in nanostructure.

Consequently with space symmetry of the observed ultrathin film model, we get symmetrical situation for peaks distribution.

These results may be better explained by experimental facts regarding resonating optical/luminescence peaks in similar molecular layered nanostructures. These effects are manifested by narrow optic absorption an refraction in close infrared band. Very good agreement in resonating absorption may be attributed and explained by presence of boundary conditions and quantum size effects for nano-sized samples.

Acknowledgement

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