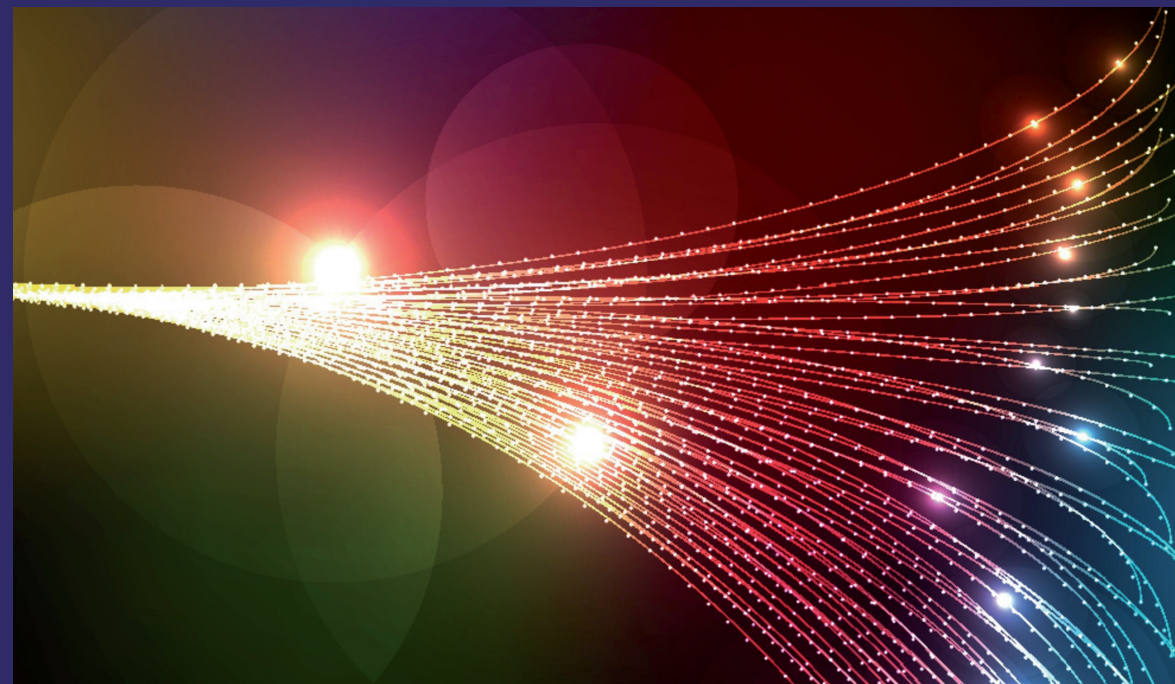


The brochure is devoted to phenomenological theory of optically active nonideal one-dimensional multilayer systems. Numerical simulation is carried out for concentration dependence of the specific rotation angle in the specified systems. Gyrotropy features caused by various disordering types are elucidated. Frequency dependence of the specific rotation angle of polarization plane of linearly polarized light is studied for molecular-crystal multilayers whose layers contain point defects. The developed theory creates additional possibilities for modeling and design of optically active multilayer composite materials.



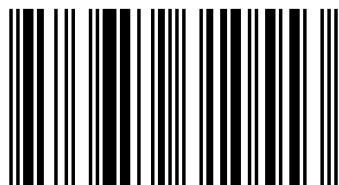
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Theory of Optically Active Imperfect Composite Materials

Selected Topics



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Introduction

As of now there are a lot of synthesized organic complexes and polymers, which happen to be optically active either due to peculiarities of their structure or because of optical activity of their constituent molecules [1-3]. A considerable attention has been devoted to investigation of optical properties of ideal and nonideal crystals [4-9], including polymers and liquid crystals [10]. Special interest in these objects is due to their high sensibility to external fields and strong anisotropy of dielectric permittivity. A method to account for concentration dependence of light refractive index in imperfect lyotropic lamellar systems developed in Ref. [10] proves useful for modeling the composite materials based on liquid crystals with prescribed parameters without consideration of spatial dispersion. At the same time incorporating spatial dispersion into the model permits to widen the range of the studied phenomena and examine such effects as natural optical activity, electrogyration, circular dichroism, etc. Investigation of dispersion of gyrotropic characteristics allows to reveal structural peculiarities of the corresponding media, and to determine important dynamic parameters of their constituent molecules [11]. It also helps to account for the spatial dispersion effects in polarization measurements of spectral characteristics of non-linear optical processes. Gyrotropy is very often the sole tool for determination of certain stereo- and crystal-chemical parameters, as well as fine structural details in spatially dispersive structures.

An important feature of many biological systems is molecular chirality, which is of interest for bio-chemical and pharmaceutical research. Chiral systems are exemplified by amino acids, sugars, proteins and enzymes. While biomolecules have been primarily examined as constituents of liquid solutions, their properties in solid-state phases are of great importance for pharmaceutical industry. Of special interest are the cases when molecules lose their optical activity in solutions, but regain it in the solids. Understanding of gyrotropy mechanisms in organic molecular crystals and crystal complexes is essential for elucidation of the character of their interaction with

electromagnetic irradiation. Along with this, investigation of gyrotropy in crystalline objects proves instrumental for the study of organic objects (including biological) possessing any degree of orderliness.

Studies on various manifestations of optical activity in different types of media pioneered by Arago in 1811 have developed since then into a broad and ramified field of expertise. The authors do not claim to have covered the material in its full generality. They rather concentrated on the issues immediately related to their area of research.

Below are considered some of gyrotropy effects occurring in molecular crystalline systems in the exciton spectral range. The first section is devoted to the exciton theory of natural optical activity in molecular crystals. It deals with the effect of the structure of exciton states on the natural optical activity of crystals and on its frequency dispersion. Mechanisms of gyrotropy are clarified for crystalline structures with various types of intermolecular interaction.

The second section is devoted to molecular crystalline structures, whose symmetry classes allow for the onset of spatial dispersion effects (e.g. optical activity) only under external influences such as mechanical action (the so-called induced gyrotropy). Microscopic analysis of induced gyrotropy is possible due to the use of the theory of excitons in the crystals subject to mechanical stress.

Since real crystalline systems are always imperfect, the correct interpretation of the corresponding experimental data and distinguishing between various gyrotropy mechanisms require development of a microtheory which would take into account the existing structural defects. In the third section a microtheory of gyrotropy is constructed for molecular systems containing point defects (substitutional impurities, vacancies, systems with orientationally disordered molecules etc.).

In the fourth section configurational averaging technique is adopted for phenomenological description of natural optical activity of nonideal multilayer materials. We construct a model of one-dimensional superlattice with randomly included impurity layers, which differ by their width and physico-chemical parameters from the layers of an ideal structure. A numerical modeling is performed

for the specific rotation of polarized light propagating along the layerwise optical axes of the described structure. To be specific we consider a two-sublattice SiO_2 -liquid-crystal structure. Peculiarities of gyrotropy behavior are discussed in their connection to the corresponding types of disordering in the studied systems. The developed phenomenological theory permits to investigate the frequency dependence of the specific rotation angle of polarization plane of plane-polarized light in molecular crystal multilayers with point defects. The obtained results create additional possibilities for modeling optically active multilayered composite materials.

The major part of this review is the result of the authors' original research. It is authors' sincere hope that their work will stimulate a wider interest in the outlined range of problems.

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1. Natural optical activity

1.2 Excitonic theory of optical activity of molecular crystals

Phenomenological theory of natural optical activity (NOA) has been sufficiently developed in book [12]. Phenomenological approach has admittedly a limited applicability and cannot provide answers to a number of questions concerning frequency dispersion of angular rotation of polarization plane of linearly-polarized monochromatic electromagnetic waves, connection between the rotation angle and the structure of exciton states, numerical evaluation of certain quantities of interest etc. Tackling these issues requires microscopic treatment. Construction of microtheory of NOA for molecular crystals became feasible after Agranovich [13] and Davydov [14] developed a theory of Frenkel excitons in molecular crystals. In Ref. [13] exciton states are analyzed in the frame of Heitler-London approximation

(H-L) without considering intermolecular interaction. In Ref. [15] H-L approximation is adopted too, but with mixing interaction taken into account in the first-order perturbation theory. The theory of NOA of crystals not based on H-L approximation was developed in Ref. [16], However it addressed only uniaxial crystals and ignored mixing of molecular states as well as the dependence of exciton wave functions on wave vector \mathbf{k} . Finally, in the above mentioned works [13,15,16] derivatives of exciton characteristics (energies and wave functions) with respect to the wave vector appeared as independent parameters and were not connected to specific crystalline structure (medium microparameters). This resulted in cumbersome formulas for gyrotropic tensor and rotatory power, proving not quite suitable for analysis of various experimental situations. These flaws were eliminated in paper [17].

According to Refs. [13,14] in the model of stationary molecules Hamiltonian \hat{H} of a molecular crystal has the form

$$\hat{H} = \sum_{\bar{n}\alpha} \hat{H}_{\bar{n}\alpha} + \frac{1}{2} \sum_{\bar{n}\alpha, \bar{m}\beta} \hat{V}_{\bar{n}\alpha, \bar{m}\beta} (1 - \delta_{\bar{n}\bar{m}} \delta_{\alpha\beta}), \quad (1.1)$$

where $\hat{H}_{\bar{n}\alpha}$ is Hamiltonian of an isolated molecule α in site \bar{n} ; $\hat{V}_{\bar{n}\alpha, \bar{m}\beta}$ is a Coulomb interaction operator for molecules $\bar{n}\alpha$ and $\bar{m}\beta$. Separation of exciton part of Hamiltonian (1.1) is performed by consecutive application of approximate secondary quantization method to the case of molecular excitons [14]. According to this method transition to the second quantization representation is carried out with the use of a set of wave functions $\varphi_{\bar{n}\alpha}^{(f)}$ characterizing the states of individual molecules in crystal. The method of approximate secondary quantization suggests a way to find such wave functions. As applied to Hamiltonian (1.1) it yields the following self-consistent system of equations for unknown $\varphi_{\bar{n}\alpha}^{(f)}$:

$$\begin{cases} \left(\hat{H}_{\bar{n}\alpha} + \hat{W}_{\bar{n}\alpha} \right) \varphi_{\bar{n}\alpha}^{(f)} = \varepsilon_{\alpha}^{(f)} \varphi_{\bar{n}\alpha}^{(f)} \\ \hat{W}_{\bar{n}\alpha} = \sum_{\bar{m}\beta} \left\langle \varphi_{\bar{n}\alpha}^{(0)} \left| \hat{V}_{\bar{n}\alpha, \bar{m}\beta} \right| \varphi_{\bar{m}\beta}^{(0)} \right\rangle, \end{cases} \quad (1.2)$$

where $\varepsilon_{\alpha}^{(f)}$ is the $\bar{n}\alpha$ -molecule energy corresponding to the state $\varphi_{\bar{n}\alpha}^{(f)}$.

Using these $\varphi_{\bar{n}\alpha}^{(f)}$ to transform exciton part of Hamiltonian to the second-quantized representation we arrive at the following formulas:

$$\hat{H}^{ex} = \sum_{\mathbf{k}} \hat{H}^{ex}(\mathbf{k}) = \sum_{\bar{n}\alpha f} E_{\alpha}^{(f)} B_{\bar{n}\alpha f}^{+} B_{\bar{n}\alpha f} + \frac{1}{2} \sum_{\substack{\bar{n}\alpha f \\ \bar{m}\beta g}} V_{\bar{n}\alpha\bar{m}\beta}^{fg} (B_{\bar{n}\alpha f}^{+} + B_{\bar{n}\alpha f}) (B_{\bar{m}\beta g}^{+} + B_{\bar{m}\beta g}), \quad (1.3)$$

where

$$V_{\bar{n}\alpha\bar{m}\beta}^{fg} = \left\langle \varphi_{\bar{n}\alpha}^{(f)} \varphi_{\bar{m}\beta}^{(0)} \left| \hat{V}_{\bar{n}\alpha\bar{m}\beta} \right| \varphi_{\bar{n}\alpha}^{(0)} \varphi_{\bar{m}\beta}^{(g)} \right\rangle, \quad E_{\alpha}^{(f)} = \varepsilon_{\alpha}^{(f)} - \varepsilon_{\alpha}^{(0)},$$

$B_{\bar{n}\alpha f}^{+}, B_{\bar{n}\alpha f}$ are creation and annihilation operators of molecular excitation, $V_{\bar{n}\alpha\bar{m}\beta}^{fg}$ is the matrix of resonance intermolecular interaction. Exciton states $\Psi_{\mu}(\mathbf{k}), E_{\mu}(\mathbf{k})$ necessary for subsequent calculation of normal electromagnetic waves in spatially dispersive crystals can be found by diagonalization of Hamiltonian (1.3) by Bogolyubov-Tyablikov transformation.

As shown in Ref. [13], the main gyrotropic characteristics of molecular structures are expressible through the gyrotropy tensor $\gamma_{jl}^{\perp}(\mathbf{s}, \omega)$ [17] defined as:

$$\gamma_{jl}^{\perp} = \frac{1}{i} \frac{\partial \chi_{jl}^{\perp}(\mathbf{k}, \omega)}{\partial k} \Big|_{k=0}, \quad (1.4)$$

$\mathbf{s} = \mathbf{k} / k$. Dielectric permittivity tensor $\chi_{jl}^{\perp}(\mathbf{k}, \omega)$ appearing in Eq. (1.4) has the form

$$\chi_{jl}^{\perp}(\mathbf{k}, \omega) = \left(1 - \frac{\omega_p^2}{\omega^2} \right) \delta_{jl} + \frac{8\pi}{V\omega^2} \sum_{\mu} \frac{E_{\mu}(\mathbf{k}) \langle \Psi_0(\mathbf{k}) | J^j(-\mathbf{k}) | \Psi_{\mu}(\mathbf{k}) \rangle \cdot \langle \Psi_{\mu}(\mathbf{k}) | J^l(\mathbf{k}) | \Psi_0(\mathbf{k}) \rangle}{E_{\mu}^2(\mathbf{k}) - \hbar^2 \omega^2}, \quad (1.5)$$

its evaluation requires knowledge of the states of Coulomb excitons $\Psi_{\mu}(\mathbf{k}), E_{\mu}(\mathbf{k})$.

In Eq.(1.5) $\mathbf{J}(\mathbf{k}) = \sum_{\bar{n}\alpha} \exp(i\mathbf{k}\mathbf{r}_{\bar{n}\alpha}) \mathbf{J}_{\bar{n}\alpha}(\mathbf{k})$ is the Fourier transform of the crystal current density operator, $\mathbf{J}_{\bar{n}\alpha}$ is the Fourier transform of the current density operator of molecule $\bar{n}\alpha$, ω_p is the frequency of plasma oscillations; V is the crystal volume. Upon substituting Eq. (1.5) into Eq. (1.4), the gyrotropy tensor appears as a sum of two terms

$$\gamma_{ij}^{\perp}(\mathbf{s}, \omega) = \frac{8\pi S^i}{v_0} \left\{ \frac{\hbar}{i} \sum_{\mu} \frac{P_{0;s\mu}^j Q_{s\mu;0}^{il} + P_{s\mu;0}^l Q_{0;s\mu}^{ij}}{E_{\mu}^2(\mathbf{s}) - \hbar^2 \omega^2} + \sum_{\mu\nu} \frac{E_{\mu}(\mathbf{s}) E_{\nu}(\mathbf{s}) P_{0;s\mu}^j W_{\mu\nu}^t(\mathbf{s}) P_{s\nu;0}^l}{[E_{\mu}^2(\mathbf{s}) - \hbar^2 \omega^2][E_{\nu}^2(\mathbf{s}) - \hbar^2 \omega^2]} \right\}, \quad (1.6)$$

where

$$W_{\mu\nu}^t(\mathbf{s}) = \left\langle \psi_{s\mu} \left| \left(\frac{i\partial H^{(ex)2}(\mathbf{k})}{\partial |\mathbf{k}|} \right) \right|_{\mathbf{k}=0} \psi_{s\nu} \right\rangle;$$

$$Q_{0;s\mu}^{il} = \sum_{f\alpha} \left\langle \varphi_{\bar{n}\alpha}^{(0)} \left| \left(\frac{i\partial J_{n\alpha}^l(\mathbf{k})}{\partial k^i} \right) \right|_{k=0} \varphi_{\bar{n}\alpha}^{(f)} \right\rangle [u_{f\alpha\mu}(\mathbf{s}) - v_{f\alpha\mu}(\mathbf{s})];$$

$u_{f\alpha\mu}(\mathbf{s}), v_{f\alpha\mu}(\mathbf{s})$ are the $u-v$ coefficients of Bogolyubov-Tyablikov transformation.

It is evident that the first term in Eq. (1.6) originates from expansion in \mathbf{k} of characteristics of individual molecules and if the intermolecular interaction is neglected it goes into the gyrotropy tensor of oriented gas. The second term is due to expansion of exciton quantities and under neglected intermolecular interaction it becomes zero. Hence the two terms in Eq. (1.6) may be called the molecular and the crystal components of gyrotropy, respectively.

Using the explicit expression (1.3) for matrix $V_{\bar{n}\alpha \bar{m}\beta}^{fg}$ of resonance intermolecular interaction, it can be shown that in crystals with primitive lattices the crystal component of gyrotropy equals zero. This statement is rather general and does not depend on multipolarity of the considered interaction.

Unlike the previously used [13,15,16] expressions, the obtained relation for gyrotropy tensor (1.6) allows to establish the relationship between gyrotropy characteristics with the medium microparameters and elementary excitations characteristics.

1.2 Optical rotary dispersion

The microscopic consideration of the previous section allows to determine an important quantitative characteristic of gyrotropy, namely the crystal rotatory power $\rho(\mathbf{s}, \omega)$. Using formula (1.6) and expression (6.13) in Ref. [13] we have:

$$\rho(\mathbf{s}, \omega) = \frac{2\pi\omega^2}{v_0 c^2} s^i \left\{ \frac{2\hbar}{i} \sum_{\mu} \frac{Q_{s\mu;0}^\mu [\mathbf{s} \times \vec{P}_{0s\mu}]^i}{E_\mu^2(\mathbf{s}) - \hbar^2 \omega^2} + \sum_{\mu\nu} \frac{E_\mu(\mathbf{s}) E_\nu(\mathbf{s}) [\vec{P}_{0s\mu} \times \vec{P}_{s\nu;0}] \cdot \mathbf{s} W'_{\mu\nu}(\mathbf{s})}{[E_\mu^2(\mathbf{s}) - \hbar^2 \omega^2][E_\nu^2(\mathbf{s}) - \hbar^2 \omega^2]} \right\}. \quad (1.7)$$

Similarly to terminology adopted with regard to the structure of gyrotropy tensor, the first term in Eq. (1.7) corresponds to molecular component (MC), and the second – to the crystal component (CC) of rotatory power.

From the experimental point of view the behavior of $\rho(\mathbf{s}, \omega)$ in the proximity of excitonic resonances is of special interest. It should be noted that in the classic study [13] molecular frequencies $\omega_f = E_f / \hbar$ are regarded to be the resonant ones. However, it is natural to expected that resonances in crystals should be observed at crystals' eigenfrequencies (in this case on the frequencies of Coulomb excitons). Thus, during the further study of dispersion $\rho(\mathbf{s}, \omega)$ the frequencies of Coulomb excitons $\Omega_\mu = E_\mu(\mathbf{s}) / \hbar$ are considered the resonance ones. Let us examine the dependence $\rho(\mathbf{s}, \omega)$ for those directions \mathbf{s} which result in exciton states being degenerated. As is known, if these directions coincide with the crystal optic axes then $\rho(\mathbf{s}, \omega)$ is the specific rotation angle of polarization plane of linearly polarized monochromatic wave.

For the specified \mathbf{s} in the area of isolated resonance $\rho(\mathbf{s}, \omega)$ can be written in the following way:

$$\rho(\mathbf{s}, \omega) \approx \frac{\omega^2 [K_1^{(MC)}(\mathbf{s}) + K_2^{(KC)}(\mathbf{s})]}{\Omega_\mu^2 - \omega^2} + \frac{\omega^2 K_3^{(KC)}(\mathbf{s})}{(\Omega_\mu^2 - \omega^2)^2} \quad (1.8)$$

The first and the second terms in (1.8) are called Drude and Lommel terms, respectively []. Their main difference resides in the fact that under transition of the incident light frequency through resonance the former changes its sign whereas the latter does not. Formula (1.8) shows that Lommel terms correspond to the crystal component of gyrotropy, their contribution into the polarization plane rotatory angle can be experimentally defined by the following procedure. The value $\rho(\mathbf{s}, \omega)/\omega^2$ is measured at two values of ω (quite close to frequency Ω_μ): $\omega_1^2 = \Omega_\mu^2 + \Delta^2$, $\omega_2^2 = \Omega_\mu^2 - \Delta^2$. At $|\rho(\mathbf{s}, \omega_1)|/\omega_1^2 = |\rho(\mathbf{s}, \omega_2)|/\omega_2^2$ Lommel term is absent, at $|\rho(\mathbf{s}, \omega_1)|/\omega_1^2 \neq |\rho(\mathbf{s}, \omega_2)|/\omega_2^2$ Lommel term contributes to the rotatory angle.

From the above discussion it follows that for a primitive crystal lattice the crystal component of gyrotropy is zero and thus the Lommel term in (1.8) is absent.

Let us analyze the case of nondegenerate exciton states. That is when in the neighborhood of excitonic resonance holds the following approximate equality:

$$\rho(\mathbf{s}, \omega) \approx \frac{\omega^2 \left[K_4^{(MC)}(\mathbf{s}) + K_5^{(KC)}(\mathbf{s}) \right]}{\Omega_\mu^2 - \omega^2} . \quad (1.9)$$

In Ref. [13] for nondegenerate exciton states $\rho(\mathbf{s}, \omega)$ is defined by the sum of Drude and Lommel terms, but in this case – by Drude term only (1.9). The reason for this difference lies in the fact that here the crystal frequencies are considered to be resonance ones, while in [] the resonance frequencies are molecule ones.

It should be noted that under negligible role of mixing calculation of rotatory power in the neighborhood of exciton resonances can be significantly simplified by omitting the corresponding terms in the intermolecular interaction matrix $V_{\bar{n}\alpha\bar{m}\beta}^{fg}$. The same simplification applies to the study of optical rotary dispersion in H-L approximation. The corresponding parameters of smallness are given in Ref. [17].

2. Gyrotropy of molecular crystals subject to uniform deformations

2.1. Introductory notes

In the previous section the main attention was devoted to development of microscopic description of gyrotropy, investigation of its various mechanisms, elucidation of its connection to microparameters of the medium and frequency dispersion. Such studies enable to use gyrotropy as a delicate experimental method to study structural features of crystalline media. They allow performing highly accurate polarization measurements of spectral characteristics of nonlinear optical processes in spatially-dispersed molecular crystals. At the same time it is well known that for certain symmetry classes some of spatial dispersion effects (e.g. optical activity) are possible only in the presence of external fields or mechanical stress (the so called induced gyrotropy) [18]. For this reason among other studies [11,13,18] significant interest lies in microscopic analysis of gyrotropic properties of molecular crystals taking into account the mentioned factors. Relevance of such investigations is all the more obvious as gyrotropy is very often the sole sufficiently sensitive indicator of external influences to be used for determination of various stereo- and crystallochemical parameters of crystals [13,14].

Microscopic treatment of various optical effects in systems subject to external actions apparently requires knowledge of the dependence of parameters of normal electromagnetic waves in crystals on the magnitude of external input. Finding of this dependence demands in turn to answer the questions relating to modification of energy spectrum in the considered media and the subsequent calculation of optical matter tensors, which are responsible for the medium response. In the present section these problems are solved for molecular crystals homogeneously deformed by external mechanical stress with the use of the exciton model. In the frame of this model we obtain a microscopic expression for transverse tensor dielectric permittivity, which is then used to find a fundamental gyrotropic quantitative

characteristic, namely rotatory power $\rho(\hat{\epsilon}, \mathbf{s}, \omega)$ of crystal with an arbitrary number of sublattices ($\hat{\epsilon}$ denotes the deformation tensor).

Results of this section permit to analyze in detail the frequency dispersion of rotatory power for all possible types of homogeneous deformations. Below such analysis is carried out for uniaxially deformed crystal systems with primitive lattice and systems subject to shear stress.

2.2 Rotatory power of homogeneously deformed molecular crystals.

For a given character of deformations Hamiltonian of molecular crystals (as follows from [29]) has the form:

$$\hat{H}(\hat{\epsilon}) = \sum_{\bar{n}\alpha} \hat{H}_{\bar{n}\alpha} + \frac{1}{2} \sum_{\bar{n}\alpha, \bar{m}\beta} \hat{V}_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon}). \quad (2.1)$$

In (2.1) \bar{n} , \bar{m} are lattice vector, α, β are the numbers of sublattices, $H_{\bar{n}\alpha}$ is Hamiltonian of an isolated $\bar{n}\alpha$ -th molecule, $V_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon})$ is the Coulomb interaction operator of molecules $\bar{n}\alpha$ and $\bar{m}\beta$ dependent on deformation tensor $\hat{\epsilon}$. A primed summation sign (2.1) means that the terms with $\bar{n}\alpha$ equal to $\bar{m}\beta$ are dropped.

It follows from Refs. [13] that operator of Coulomb interaction between the molecules $\bar{n}\alpha$ and $\bar{m}\beta$ in a homogeneously deformed crystal has the form:

$$V_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon}) = \frac{(\mathbf{P}_{\bar{n}\alpha} \mathbf{P}_{\bar{m}\beta}) |\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon})|^2 - 3(\bar{\mathbf{P}}_{\bar{n}\alpha} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon}))(\mathbf{P}_{\bar{m}\beta} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon}))}{|\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon})|^5}, \quad (2.2)$$

Here $\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}(\hat{\epsilon}) = \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)} + \hat{\epsilon} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)}$.

In (2.2) $\mathbf{P}_{\bar{n}\alpha}$ is molecule $\bar{n}\alpha$ dipole moment operator, $\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)}$ is the radius vector connecting the $\bar{n}\alpha$ -th and $\bar{m}\beta$ -th sites in a free (unstrained) crystal lattice. In the linear in $\hat{\epsilon}$ approximation

$$V_{\bar{n}\alpha\bar{m}\beta}(\hat{\mathcal{E}}) = V_{\bar{n}\alpha\bar{m}\beta}(0) + P_{\bar{n}\alpha}^i P_{\bar{m}\beta}^j \cdot \left[-3 \frac{\delta_{il} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)l} + \delta_{il} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)l} + \delta_{il} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)i}}{|\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)}|^5} + 15 \frac{\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)i} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)l} \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)l}}{|\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)}|^7} \right] \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}^{(0)p} \mathcal{E}_p \equiv V_{\bar{n}\alpha\bar{m}\beta}^{(0)} + V_{\bar{n}\alpha\bar{m}\beta}^{(1)}(\hat{\mathcal{E}}). \quad (2.3)$$

Eq. (2.3) can be used to calculate molecular wave functions and corresponding energies in the linear in $\hat{\mathcal{E}}$ approximation with the help of perturbation theory. To this end wave functions $\varphi_{n\alpha}^f(\hat{\mathcal{E}})$ should be presented in the form of expansion in molecular wave functions $\varphi_{n\alpha}^f$, corresponding to the free crystal:

$$\varphi_{n\alpha}^f(\hat{\mathcal{E}}) = \sum_g C_{fg}(\hat{\mathcal{E}}) \varphi_{n\alpha}^{(g)}. \quad (2.4)$$

Upon substituting formulas (2.3) and (2.4) into Eq. (2.1) and separating out the terms linear in $\hat{\mathcal{E}}$ we obtain the following expression for coefficients $C_{fg}^{(1)}(\hat{\mathcal{E}})$ representing the first-order perturbation theory:

$$C_{f(\alpha)g(\alpha)}^{(1)}(\hat{\mathcal{E}}) = \frac{1}{E_{f(\alpha)}^{(0)} - E_{g(\alpha)}^{(0)}} \cdot \sum_{\bar{m}\beta} \left[\langle \varphi_{\bar{n}\alpha}^{g(\alpha)} \varphi_{\bar{m}\beta}^{(0)} | V_{\bar{n}\alpha\bar{m}\beta}^{(1)} | \varphi_{\bar{n}\alpha}^{f(\alpha)} \varphi_{\bar{m}\beta}^{(0)} \rangle + \sum_{h(\beta)} C_{0h(\beta)}^{(1)} \langle \varphi_{\bar{n}\alpha}^{g(\alpha)} \varphi_{\bar{m}\beta}^{(0)} | V_{\bar{n}\alpha\bar{m}\beta}^{(0)} | \varphi_{\bar{n}\alpha}^{f(\alpha)} \varphi_{\bar{m}\beta}^{h(\beta)} \rangle \right], \quad (2.5)$$

$$C_{0h(\alpha)}^{(1)} = \sum_{g(\alpha)} \left[\hat{A}^{-1} \right]_{h(\alpha)g(\alpha)} B_{g(\alpha)}(\hat{\mathcal{E}}), \quad (2.6)$$

where

$$\left[\hat{A} \right]_{h(\alpha)g(\alpha)} = (E_0^{(0)} - E_{h(\alpha)}^{(0)}) \delta_{h(\alpha)g(\alpha)} - 2 \sum_{\bar{m}\beta} \langle \varphi_{\bar{n}\alpha}^{g(\alpha)} \varphi_{\bar{m}\beta}^{(0)} | V_{\bar{n}\alpha\bar{m}\beta}^{(0)} | \varphi_{\bar{n}\alpha}^{(0)} \varphi_{\bar{m}\beta}^{h(\beta)} \rangle, \\ B_{g(\alpha)}(\hat{\mathcal{E}}) = \sum_{\bar{m}\beta} \langle \varphi_{\bar{n}\alpha}^{g(\alpha)} \varphi_{\bar{m}\beta}^{(0)} | V_{\bar{n}\alpha\bar{m}\beta}^{(1)}(\hat{\mathcal{E}}) | \varphi_{\bar{n}\alpha}^{(0)} \varphi_{\bar{m}\beta}^{(0)} \rangle.$$

Eq. (2.5) permits to easily find molecular energy of the first-order in $\hat{\mathcal{E}}$:

$$E_{f(\alpha)}^{(1)} = \sum_{\bar{m}\beta} \left\{ \langle \varphi_{\bar{n}\alpha}^{f(\alpha)} \varphi_{\bar{m}\beta}^{(0)} | V_{\bar{n}\alpha\bar{m}\beta}^{(1)} | \varphi_{\bar{n}\alpha}^{f(\alpha)} \varphi_{\bar{m}\beta}^{(0)} \rangle + \right. \\ \left. + 2 \sum_{h(\beta)g(\beta)} \left[\hat{A}^{-1} \right]_{g(\beta)h(\beta)} B_{h(\beta)}(\hat{\mathcal{E}}) \langle \varphi_{\bar{n}\alpha}^{f(\alpha)} \varphi_{\bar{m}\beta}^{(0)} | V_{\bar{n}\alpha\bar{m}\beta}^{(0)} | \varphi_{\bar{n}\alpha}^{f(\alpha)} \varphi_{\bar{m}\beta}^{g(\beta)} \rangle \right\}. \quad (2.7)$$

Microscopic calculation of optical characteristics corresponding to exciton spectral range assumes the explicit form of the corresponding Hamiltonian $\hat{H}^{(ex)}(\hat{\mathcal{E}})$ to be

known. For molecular crystals separation of $\hat{H}^{(ex)}(\hat{\varepsilon})$ from (2.1) is easily done by a step-by-step procedure of approximate secondary quantization [14]. The latter requires the wave functions $\varphi_{\bar{n}\alpha}^f(\hat{\varepsilon})$ of molecules in crystal field to satisfy the system of self-consistent integro-differential equations, resulting from solution of the corresponding variation problem. It can be easily shown (using the results of Ref. [288, 300]) that in the considered case the mentioned above equation system and exciton Hamiltonian appear correspondingly as:

$$\left[\hat{H}_{\bar{n}\alpha}(\hat{\varepsilon}) + \hat{W}_{\bar{n}\alpha}(\hat{\varepsilon}) \right] \varphi_{\bar{n}\alpha}^f(\hat{\varepsilon}) = \varepsilon_{f\alpha}(\hat{\varepsilon}) \varphi_{\bar{n}\alpha}^f(\hat{\varepsilon}), \quad (2.8)$$

where

$$\hat{W}_{\bar{n}\alpha}(\hat{\varepsilon}) = \sum_{\bar{m}\beta} \langle \varphi_{\bar{m}\beta}^0 | \hat{V}_{\bar{n}\alpha\bar{m}\beta}(\hat{\varepsilon}) | \varphi_{\bar{m}\beta}^0 \rangle,$$

and

$$\hat{H}^{ex} = \sum_{\vec{k}} \hat{H}^{ex}(\vec{k}) = \sum_{\bar{n}\alpha f} E_{\alpha}^{(f)} \hat{B}_{\bar{n}\alpha f}^{+} \hat{B}_{\bar{n}\alpha f} + \frac{1}{2} \sum_{\substack{\bar{n}\alpha f \\ \bar{m}\beta g}} V_{\bar{n}\alpha\bar{m}\beta}^{fg} (\hat{B}_{\bar{n}\alpha f}^{+} + \hat{B}_{\bar{n}\alpha f}) (\hat{B}_{\bar{m}\beta g}^{+} + \hat{B}_{\bar{m}\beta g}) \quad (2.9)$$

In (2.9) $V_{\bar{n}\alpha\bar{m}\beta}^{fg} = \langle \varphi_{\bar{n}\alpha}^{(f)} \varphi_{\bar{m}\beta}^{(0)} | \hat{V}_{\bar{n}\alpha\bar{m}\beta} | \varphi_{\bar{n}\alpha}^{(0)} \varphi_{\bar{m}\beta}^{(g)} \rangle$, $E_{\alpha}^{(f)} = \varepsilon_{\alpha}^{(f)} - \varepsilon_{\alpha}^{(0)}$, $\hat{B}_{\bar{n}\alpha f}^{+}, \hat{B}_{\bar{n}\alpha f}$ are molecular excitation creation and annihilation operators.

Hamiltonian (2.9) determines the state of Coulomb excitons, necessary for calculation of transverse tensor of dielectric permittivity $\hat{\chi}^{\perp}(\hat{\varepsilon}, \mathbf{k}, \omega)$, which, in its turn, helps to determine all basic optical characteristics, including the target rotatory power of the deformed medium. As $\hat{H}^{(ex)}(\hat{\varepsilon} \neq 0)$ and $\hat{H}^{(ex)}(\hat{\varepsilon} = 0)$ have the same form (due to crystals translational invariance under homogeneous deformations), tensor $\hat{\chi}^{\perp}(\hat{\varepsilon}, \mathbf{k}, \omega)$ can be found employing formulas (3), (5)-(7) of Ref. [19] and using substitutions $\varphi_{\bar{n}\alpha}^{(f)} \rightarrow \varphi_{\bar{n}\alpha}^{(f)}(\hat{\varepsilon})$, $\varepsilon_{\bar{n}\alpha}^{(f)} \rightarrow \varepsilon_{\bar{n}\alpha}^{(f)}(\hat{\varepsilon})$, $v_0 \rightarrow v_0(1 + Sp\hat{\varepsilon})$, where v_0 is the volume of elementary cell in a free (unstressed) crystal. For the part of $\hat{\chi}^{\perp}(\hat{\varepsilon}, \mathbf{k}, \omega)$ dependent on \mathbf{k} and for $\rho(\hat{\varepsilon}, \mathbf{s}, \omega) = -\frac{i\omega^2}{4c^2} \left(\frac{\partial \chi_{il}^{\perp}(\hat{\varepsilon}, \mathbf{k}, \omega)}{\partial |\mathbf{k}|} \right)_{|\mathbf{k}|=0} e_{ill} S^t$ (where Einstein

summation convention is implied and e_{ilt} is the Levy-Civita symbol) this procedure correspondingly yields:

$$\Delta\chi_{il}^\perp(\hat{\varepsilon}, \mathbf{k}, \omega) = \frac{8\pi}{v(1+Sp\hat{\varepsilon})\omega^2} \sum_{\mu} \frac{E_{\mu}(\hat{\varepsilon}, \mathbf{k}) \langle \psi_0(\hat{\varepsilon}) | I^i(-\mathbf{k}) | \psi_{\mathbf{k}\mu}(\hat{\varepsilon}) \rangle \langle \psi_{\mathbf{k}\mu}(\hat{\varepsilon}) | I^l(\mathbf{k}) | \psi_0(\hat{\varepsilon}) \rangle}{E_{\mu}^2(\hat{\varepsilon}, \mathbf{k}) - \hbar^2 \omega^2}, \quad (2.10)$$

$$\begin{aligned} \rho(\hat{\varepsilon}, \mathbf{s}, \omega) = & \frac{-2\pi i \omega^2 e_{ilt} S^t}{c^2 v_0 (1+Sp\hat{\varepsilon})} \left\{ 2\hbar \sum_{\mu} \frac{\langle \psi_0(\mathbf{s}, \hat{\varepsilon}) | P^i | \psi_{s\mu}(\mathbf{s}, \hat{\varepsilon}) \rangle \langle \psi_{s\mu}(\mathbf{s}, \hat{\varepsilon}) | S^j Q^j | \psi_0(\mathbf{s}, \hat{\varepsilon}) \rangle}{E_{\mu}^2(\hat{\varepsilon}, \mathbf{s}) - \hbar^2 \omega^2} + \right. \\ & + \sum_{\mu\nu} \left[\frac{E_{\mu}(\hat{\varepsilon}, \mathbf{s}) E_{\nu}(\hat{\varepsilon}, \mathbf{s}) \langle \psi_0(\mathbf{s}, \hat{\varepsilon}) | P^i | \psi_{s\mu}(\mathbf{s}, \hat{\varepsilon}) \rangle \langle \psi_{s\nu}(\mathbf{s}, \hat{\varepsilon}) | P^l | \psi_0(\mathbf{s}, \hat{\varepsilon}) \rangle}{[E_{\mu}^2(\hat{\varepsilon}, \mathbf{s}) - \hbar^2 \omega^2][E_{\nu}^2(\hat{\varepsilon}, \mathbf{s}) - \hbar^2 \omega^2]} \times \right. \\ & \left. \left. \times \left\langle \psi_{s\mu}(\mathbf{s}, \hat{\varepsilon}) \left| \left(i\partial H^{(ex)^2}(\hat{\varepsilon}, \mathbf{k}) / \partial |\mathbf{k}| \right) \right|_{|\mathbf{k}|=0} \psi_{s\nu}(\mathbf{s}, \hat{\varepsilon}) \right\rangle \right] \right\} \quad (2.11) \end{aligned}$$

Here $\psi_{\mathbf{k}\mu}(\hat{\varepsilon})$ and $E_{\mu}(\hat{\varepsilon}, \mathbf{k})$ are exciton wave functions and energy, $\mathbf{I}(\mathbf{k})$ and \mathbf{P} are correspondingly operators of current density and crystal dipole moment, v is the volume of a strain free crystal, the content of other signs in (2.10), (2.11) being the same as in [17-19]. It follows from Eqs. (2.8)-(2.11) that the dependence of rotatory power on $\hat{\varepsilon}$ cannot be expressed analytically in its general form and finding this dependence for arbitrary deformations should be done individually in each particular case with the account for specifics of the considered systems and with the use of the most suitable approximation with adjustable parameters $V_{\bar{n}\bar{\alpha}\bar{m}\bar{\beta}}^{fg}(\hat{\varepsilon})$. At the same time it is obvious that at moderate crystal deformations (not resulting in irreversible structural changes) the rotatory power to sufficient accuracy can be written in linear in $\hat{\varepsilon}$ approximation with the use of standard formulas of perturbation theory (see Eqs. (2.5)-(2.7)). In this case the corresponding (approximate) microscopic expression for $\rho(\hat{\varepsilon}, \mathbf{s}, \omega) \cong \rho^{(0)}(\mathbf{s}, \omega) + \rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ has a universal form for each class of crystal systems with the same number of sublattices. As can be seen from (2.11) $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ is expressed as a sum of terms reflecting the corresponding mechanisms of induced gyrotropy.

It follows from the said above that the most important in an experimental study analysis of frequency dispersion of rotatory power comes to the analysis of each of the terms and most easily fulfilled for systems with primitive lattices (for which the second term in (2.11) is zero). In the latter case function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ looks as follows:

$$\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = -Sp\hat{\epsilon}\rho^{(0)}(\mathbf{s}, \omega) + \rho_M^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_C^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \equiv \frac{\omega^2}{2c} s_i s_l g_{ilpt}^\perp(\mathbf{s}, \omega) \epsilon_{pt} \quad (2.12)$$

The first term in (2.12) is due to variation of lattice constants under crystal deformation and in optically non-active free media it is zero for \mathbf{s} collinear to the optical axis. Function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is connected with expansion in $\hat{\epsilon}$ molecular characteristics. It is determined mainly by molecules gyrotropy. The third term in (2.12) is conditioned by the dependence of exciton characteristics on $\hat{\epsilon}$ - in the model of oriented gas it also equals zero. The form of pseudotensor of the fourth rank $g_{ilpt}^\perp(\mathbf{s}, \omega)$ is determined by the group $G_s(\hat{\epsilon} = 0)$. A more detailed analysis $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ needs specification of the character of external actions. In such a case the deformation tensor has the form $\epsilon_{il} = \sigma s_{ilpt} q_p q_t$, where σ is strain, s_{ilpt} is the tensor of elastic compliance coefficients. The systems subject to shear stress were studied in Ref. [18]. In the last case the deformation tensor has the form $\epsilon_{il} = \sigma s_{ilrt}(p_r q_t + q_r p_t)$, where \mathbf{p}, \mathbf{q} are unit vectors corresponding to shear and direct stress.

2.3. Dispersion of induced gyrotropy in uniaxially deformed molecular crystals

Of the highest interest in the experimental research of induced gyrotropy is its behaviour in the proximity of exciton resonances. From formulae (2.11) and (2.12) it is evident that in this frequency range for arbitrary \mathbf{s} and \mathbf{q} function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ can be approximated by a linear combination of Drude $\rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ and Lommel

$\rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ terms, proportional correspondingly to $(E_\mu^2(\mathbf{s}) - \hbar^2 \omega^2)^{-1}$ and $(E_\mu^2(\mathbf{s}) - \hbar^2 \omega^2)^{-2}$. It is easy to show that the first type of terms (with specified \mathbf{s} and \mathbf{q}) is conditioned by three functions only, included in (2.12), while the second type appears from $\rho_C^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ only. For $H^{(ex)}(\hat{\epsilon}, \mathbf{s})$ possessing certain symmetry, the form of function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is significantly dependent upon \mathbf{q} and, as will be evident from the examples, can vanish for certain \mathbf{q} 's. In this case revealing of microscopic structure of $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ requires a group-theoretical analysis of the matrix elements in (2.11). It is obvious that this analysis is most topical for those crystalline classes whose symmetry allows existence of induced (by mechanical stress) optical activity. As shown in Ref. [18], such are symmetry classes C_{3v} , C_{4v} , C_{6v} , C_{3h} , D_{3h} , T_d .

For each of the mentioned groups the symmetry conditions for function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ becoming zero are presented below together with a detailed analysis of $\rho^{(0)}(\mathbf{s}, \omega)$, $\rho_M^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ and $\rho_C^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ contribution to Drude and Lommel terms.

1. Group C_{3v}

1.1. Vector \mathbf{s} is directed along triad axis.

In this case $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ becomes zero at: 1) $\mathbf{q} \parallel \mathbf{s}$, 2) \mathbf{q} perpendicular to one of the symmetry planes, 3) \mathbf{q} perpendicular to \vec{s} and lying in one of three symmetry planes. In case with arbitrary \mathbf{q} for degenerate excitonic energies $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$, and for energies of all other symmetry types $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$. At that the contribution to $\rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is made by both $\rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ and $\rho_C^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ ($\rho^{(0)}(\mathbf{s}, \omega) = 0$).

1.2. Vector \mathbf{s} lies in one of three symmetry planes. ($\rho^{(0)}(\mathbf{s}, \omega) = 0$).

For such \mathbf{s} $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is zero under the same \mathbf{q} , as in the previous case. For arbitrary \mathbf{q} this function is described by terms of Drude type only.

1.3. Vector \underline{s} is perpendicular to one of three symmetry planes.

This case is similar to #1.2.

2. Group C_{4v}

2.1. Vector \underline{s} is directed along tetrad axis ($\rho^{(0)}(\underline{s}, \omega) = 0$).

For such \underline{s} $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega) = 0$ at any \underline{q} . In the second order in $\hat{\epsilon}$ $\rho(\hat{\epsilon}, \underline{s}, \omega) \neq 0$ in case of deformation along arbitrary directions.

2.2. Vector \underline{s} is perpendicular to one of the symmetry planes ($\rho^{(0)}(\underline{s}, \omega) = 0$).

Function $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega) = 0$ at $\underline{q} \perp \underline{s}$. For an arbitrary \underline{q} $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$, at that contribution $\rho_D^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$ is conditioned by both $\rho_M^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$ and $\rho_C^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$.

3. Group C_{6v}

3.1. Vector \underline{s} is directed along hexad axis ($\rho^{(0)}(\underline{s}, \omega) = 0$).

Function $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$ equals zero at 1) $\underline{q} \parallel \underline{s}$, 2) \underline{q} perpendicular to one of the symmetry planes, 3) \underline{q} lying in one of six symmetry planes. For arbitrary \underline{q} in cases of degenerate excitonic states $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \underline{s}, \omega) + \rho_L^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$, and $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$ in all other cases.

3.2. Vector \underline{s} is perpendicular to one of the symmetry planes

($\rho^{(0)}(\underline{s}, \omega) = 0$). Function $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$ equals zero at the same \underline{q} , as in 3.1. For an arbitrary direction of contraction axis always holds $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \underline{s}, \omega)$.

4. Group C_{3h}

4.1. Vector \underline{s} is directed along triad axis ($\rho^{(0)}(\underline{s}, \omega) = 0$).

In this case $\rho^{(1)}(\hat{\epsilon}, \underline{s}, \omega) = 0$ at $\underline{q} \perp \underline{s}$. For arbitrary \underline{q} in case of dipole active (complex conjugate) exciton states with corresponding energies

$\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$, and in all other cases
 $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$.

4.2. Vector \mathbf{s} lies in symmetry plane ($\rho^{(0)}(\mathbf{s}, \omega) = 0$).

Function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ equals zero at the same \mathbf{q} , as in 4.1. For an arbitrary \mathbf{q} holds
 $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$.

5. Group D_{3h}

5.1. Vector \mathbf{s} is directed along triad axis ($\rho^{(0)}(\mathbf{s}, \omega) = 0$).

In this case $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ at 1) $\mathbf{q} \perp \mathbf{s}$ and 2) \mathbf{q} , lying in any of three vertical planes of the symmetry. For arbitrary \mathbf{q} in the case of degenerate excitonic states
 $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$, and $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ in any other case.

5.2. Vector \mathbf{s} is directed along one of three twofold symmetry axes.

Function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ in this case equals zero at the same \mathbf{q} as in 5.1, but in the exciton resonances proximity it is always approximated by terms of Drude type.

5.3. Vector \mathbf{s} lies in any of three vertical symmetry planes.

In this case function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ equals zero at $\mathbf{q} \perp \mathbf{s}$. For arbitrary \mathbf{q}
 $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$.

5.4. Vector \mathbf{s} lies in a horizontal symmetry plane.

Function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ in this case equals zero at the same \mathbf{q} as in 5.1. For any other \mathbf{q}
 $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$.

6. Group T_d

For any \mathbf{s} and \mathbf{q} function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$. In the second order in $\hat{\epsilon}$ rotatory power differs from zero for arbitrary \mathbf{q} .

2.4 Gyrotropy of molecular crystals induced by external shear stress

It has been mentioned above that induced gyrotropy in the proximity of exciton resonances arouses the biggest interest for experimental studies. In this connection we study function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ in this area. Form formulae (2.11) and (2.12) it follows that in this frequency range for arbitrary (non-symmetrical) \mathbf{s} $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is always different from zero and, as in the previous section, can always be approximated by the linear combination of Drude $\rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ and Lommel $\rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ terms. The first type of terms (at the specified \mathbf{s}) is conditioned by all three functions present in (2.12), while the second one appears from $\rho_C^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ only. Revealing microscopic structure of $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ requires the group-theoretical analysis of matrix elements in (2.11). At the same time for definite symmetrical \mathbf{p} , \mathbf{q} the specified function can become zero. It is natural that such analysis is the most topical for those crystals whose symmetry allow for the existence of optical activity induced by external shear stress. As it is shown above (in section 6.2.3) such classes are crystal symmetry classes C_{3v} , C_{4v} , C_{6v} , C_{3h} , D_{3h} , T_d . For each of the mentioned groups the symmetry conditions of function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ becoming zero are defined below. Its behaviour in the proximity of exciton resonances is studied in detail. It should be noted that function $\rho^{(1)}(\mathbf{s}, \omega)$ for the specified groups is zero not only at \mathbf{s} , directed along the optical axes, but also as it is revealed by group-theoretical analysis, for all other symmetrical directions. It is taken into account below that the contribution of $\rho^{(0)}(\mathbf{s}, \omega)$ to Drude terms for such \mathbf{s} is zero.

1 Group C_{3v}

1.1. Vector \mathbf{s} is directed along triad axis C_3

In this case $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ becomes zero at: 1) $\mathbf{p} \parallel \mathbf{s}$ and \mathbf{q} , lying in one of the symmetry planes; 2) $\mathbf{q} \parallel \mathbf{s}$ and \mathbf{p} are in one of the symmetry planes. For all other \mathbf{p}

and $\mathbf{q} \quad \rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) + \rho_L^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ for degenerate exciton levels and $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ for energies of other symmetry types. In such case contribution to $\rho_D^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ is made by both $\rho_M^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ and $\rho_C^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$.

1.2. Vector \mathbf{s} lies in one of the symmetry planes.

For such \mathbf{s} $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ is zero at: 1) $\mathbf{q} \parallel C_3$ and \mathbf{p} , lying in the same plane as \mathbf{s} ; 2) $\mathbf{p} \parallel C_3$, and \mathbf{q} is in the same plane as \mathbf{s} . For all other \mathbf{p} , \mathbf{q} $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ is defined by Drude terms only.

1.3. Vector \mathbf{s} is perpendicular to one of three symmetry planes.

At such \mathbf{s} the function under consideration is zero at: 1) $\mathbf{q} \parallel C_3$ and $\mathbf{p} \perp \mathbf{s}$; 2) $\mathbf{p} \parallel C_3$ and $\mathbf{q} \perp \mathbf{s}$. For other \mathbf{p} , \mathbf{q} $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ is always true.

2. Group C_{4v} .

2.1. Vector \mathbf{s} is directed along tetrad axis C_4 .

At such \mathbf{s} $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ for any \mathbf{p} , \mathbf{q} . In the second order in $\hat{\varepsilon}$ $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) \neq 0$ in case of deformations with arbitrary \mathbf{p} , \mathbf{q} .

2.2. Vector \mathbf{s} is perpendicular to tetrad axis and lies in one of the symmetry planes.

In this case the analysed function is zero at: 1) $\mathbf{q} \parallel C_4$ and arbitrary \mathbf{p} ; 2) $\mathbf{p} \parallel C_4$, \mathbf{q} is an arbitrary vector. For other \mathbf{p} and \mathbf{q} $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$ is always true.

2.3. Vector \mathbf{s} lies in one of the symmetry planes and not perpendicular to C_4 .

For such \mathbf{s} function $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) = 0$ at: 1) $\mathbf{q} \parallel C_4$ and \mathbf{p} lies in the same plane as \mathbf{s} ; 2) $\mathbf{p} \parallel C_4$, and \mathbf{q} lies in the studied symmetry plane. In case with arbitrary \mathbf{p} , \mathbf{q} $\rho^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\varepsilon}, \mathbf{s}, \omega)$.

3. Group C_{6v} .

3.1. Vector \mathbf{s} is directed along hexad axis C_6 .

At such \mathbf{s} $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ at: 1) $\mathbf{q} \parallel \mathbf{s}$ and arbitrary \vec{p} ; 2) $\mathbf{p} \parallel \mathbf{s}$ and arbitrary \mathbf{q} . For other \mathbf{p} , \mathbf{q} in case of degenerate exciton states $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$; in all other cases $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$.

3.2. Vector \mathbf{s} lies in one of the symmetry planes and is perpendicular to C_6 .

Function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ at: 1) $\mathbf{q} \parallel C_6$, \mathbf{p} is an arbitrary vector; 2) $\mathbf{p} \parallel C_4$ and arbitrary \mathbf{q} . For all other \mathbf{q} $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is always true.

3.3. Vector \mathbf{s} lies in one of the symmetry planes and is not perpendicular to C_6 .

Function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ for such \mathbf{s} at: 1) $\mathbf{q} \parallel C_6$ and \mathbf{p} lying in the symmetry plane under consideration; 2) $\mathbf{p} \parallel C_6$ and \mathbf{q} are in the same plane as \mathbf{s} . In case when \mathbf{q} is not parallel C_6 $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is always true.

4. Group C_{3h}

4.1. Vector \mathbf{s} is directed along triad axis C_3 .

In this case function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ at \mathbf{p} and \mathbf{q} lying in the symmetry plane. For arbitrary \mathbf{p} , \mathbf{q} : $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_L^{(2)}(\hat{\epsilon}, \mathbf{s}, \omega)$ - only for dipole active (complex conjugate) exciton states, in all other cases $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$.

4.2. Vector \mathbf{s} lies in the symmetry plane.

For such \mathbf{s} the function under consideration is zero at $\mathbf{p} \perp C_3$ and $\mathbf{q} \perp C_3$. In case with arbitrary \mathbf{p} , \mathbf{q} behaviour of $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is similar to that analyzed in 4.1.

5. Group D_{3h} .

5.1. Vector \mathbf{s} is directed along triad axis C_3

In this case $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ at: 1) \mathbf{q} parallel to one of the twofold axis (C_2) and arbitrary \mathbf{p} ; 2) \mathbf{p} along one of axes C_2 and arbitrary \mathbf{q} . For arbitrary \mathbf{p} , \mathbf{q} in the case of degenerate exciton states $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) + \rho_L^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ and $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ in all other cases.

5.2. Vector \mathbf{s} is directed along one of the twofold axes C_2 .

For data \mathbf{s} the function under consideration is zero at: 1) $\mathbf{q} \parallel \mathbf{s}$ and arbitrary \mathbf{p} ; 2) $\mathbf{p} \parallel \mathbf{s}$ and arbitrary \mathbf{q} . At arbitrary \mathbf{p} , \mathbf{q} $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is always true.

5.3. Vector \mathbf{s} lies in one of the vertical symmetry planes and not perpendicular to C_3

For such \mathbf{s} $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ at: 1) $\mathbf{q} \parallel C_3$ and \mathbf{p} , parallel to the corresponding twofold axis; 2) $\mathbf{p} \parallel C_3$ and \mathbf{q} , lying in the same symmetry plane as \mathbf{s} . In all other cases $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is always true.

5.4. Vector \mathbf{s} lies in the horizontal symmetry plane and does not coincide with C_2

For these \mathbf{s} function $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$ at: 1) \mathbf{q} is parallel to one of the twofold axes, and $\mathbf{p} \perp C_3$; 2) \mathbf{p} coinciding with one of the twofold axes and $\mathbf{q} \perp C_3$. In all cases $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) \approx \rho_D^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega)$ is always true.

6. Group T_d .

For any \mathbf{s} and \mathbf{p} , \mathbf{q} $\rho^{(1)}(\hat{\epsilon}, \mathbf{s}, \omega) = 0$. In the second order in $\hat{\epsilon}$ the rotatory power is different from zero for arbitrary \mathbf{p} , \mathbf{q} .

In conclusion, this subsection presents a microscopic analysis of shear-stress-induced gyrotropy in molecular crystals. It is evident that the above-used Frenkel exciton model for microscopic description of induced gyrotropy is applicable not only to molecular crystals proper (in particular to simple cryocrystals such as solid hydrogen, hexagonal close-packed Ar, Kr, Ne etc.), but also to valence bond crystals

(diamond-like structures with T_d symmetry), whose electron excitations can be described by quasi-molecular model [18]. Crystals with the analyzed symmetry are optically inactive at the absence of external action. For other types of homogeneous deformation tensor $\hat{\epsilon}$ has a different form from the discussed form. Thus, the character of frequency dispersion of rotatory power is different from the analysed case. Nevertheless, the developed technique allows to carry out the microscopic analysis of molecular crystals gyrotropy in the exciton spectral region for other types of homogeneous deformation as well. The technique of experimental check of Drude and Lommel terms is described in Ref. [19]. Paper [20] is useful in finding the ways of experimental study of crystals optical activity induced by homogeneous deformation.

It's worthwhile noting that along with the microscopic approach, developed herein, for molecular crystals there exists another one [20] based on the effective Hamiltonian method. Monograph [20] covers issues in the theory of electronic phenomena in crystals distorted by weakly non-uniform deformations. It presents the results of investigation of the general properties of the corresponding effective Hamiltonian and particular problems on the energy spectrum and electron dynamics in deformed crystals. There is also an examination of screening of deformation potential by conduction electrons in metals and semiconductors as well as kinetic phenomena in deformed conductors and superconductors.

3. Gyrotropy in topologically ordered imperfect molecular crystals

3.1. Introductory notes.

Construction of exciton theory of gyrotropy, considered above is feasible due to well-known exciton states in perfect molecular crystals. Yet real crystalline systems are always imperfect. Therefore an adequate interpretation of experimental data, identification of various gyrotropy mechanisms, understanding of their peculiarities in each particular case are possible only in the context of a microtheory, where existing structural imperfections are taken into account. Such theory is the most easy

to develop for topologically ordered defect structures exemplified by molecular crystals containing substitutional impurities and vacancies with no considerable distortion of their surroundings. Vacancies can formally be considered as substitutional impurities with zero values of molecular currents, dipole moments, energies and corresponding elements of intermolecular interaction matrix. The systems mentioned above allow application of the averaged Green functions technique [21]. The latter define renormalization of exciton states and allow evaluation of optical tensors, which in turn are sufficient to find all the corresponding characteristics of normal electromagnetic waves including gyrotropic parameters. In the considered case gyrotropic parameters are functions not only of frequency but of defect concentration as well. In the present section a single-level model is adopted to find microscopic expressions for transverse tensor of dielectric permittivity, gyrotropy tensor and rotary power of topologically disordered non-ideal systems with an arbitrary number of sublattices.

3.2 Transverse tensor of dielectric permittivity of randomly disordered molecular crystals.

Similar to the calculation carried out in Section 1.1 the calculation of rotatory power is fulfilled with the help of the dielectric permittivity transverse tensor. This tensor is calculated by the linear response method and it is expressed by Fourier transform of the two-time retarded Green function $\langle\langle J^j(-\mathbf{k}) | J^l(\mathbf{k}) \rangle\rangle_\omega^r$ from the current density operator [14]. In impurity crystals finding the dielectric permittivity tensor by the specified method requires averaging of the calculated quantities over all possible configurations of point defects. In the result of this calculation method the part of dielectric permittivity transverse tensor $\Delta\chi_{il}^\perp$ depending on \mathbf{k} can be presented in the form

$$\Delta\chi_{il}^\perp(\mathbf{k}, \omega, \{C_\alpha^{\nu(\alpha)}\}) = \frac{4\pi}{V\omega^2\hbar} \sum_{\bar{n}, \bar{m}=1}^{\bar{N}} \sum_{\alpha, \beta=1}^{\kappa} \exp(-i\mathbf{k} \cdot \mathbf{r}_{\bar{n}\alpha\bar{m}\beta}) \hat{P} J_{\bar{n}\alpha; f0}^l(-\mathbf{k}) \times \left\langle \left\langle \hat{B}_{\bar{n}\alpha}^+ - \hat{B}_{\bar{n}\alpha} \left| \hat{B}_{\bar{m}\beta}^+ - \hat{B}_{\bar{m}\beta} \right. \right\rangle \right\rangle_\omega^r J_{\bar{m}\beta; f0}^l(\mathbf{k}). \quad (3.1)$$

In (3.1) N is the number of elementary cells in volume V , \hat{P} is the operator of configurational averaging [22], $C_\alpha^{\nu(\alpha)}$ is the concentration of point defects of $\nu(\alpha)$ -th sort in α -th sublattice. Fourier transform of the two-time retarded Green function

$$\left\langle \left\langle \hat{B}_{\bar{n}\alpha}^+ - \hat{B}_{\bar{n}\alpha} \left| \hat{B}_{\bar{m}\beta}^+ - \hat{B}_{\bar{m}\beta} \right. \right\rangle \right\rangle_\omega^r = \left\langle \Psi_0^{(ex)} \left| \left(\hat{B}_{\bar{n}\alpha}^+ - \hat{B}_{\bar{n}\alpha} \right) \left(\hbar\omega - H^{(ex)} \right)^{-1} \left(\hat{B}_{\bar{m}\beta}^+ - \hat{B}_{\bar{m}\beta} \right) \right| \Psi_0^{(ex)} \right\rangle - \left\langle \Psi_0^{(ex)} \left| \left(\hat{B}_{\bar{m}\beta}^+ - \hat{B}_{\bar{m}\beta} \right) \left(\hbar\omega + H^{(ex)} \right)^{-1} \left(\hat{B}_{\bar{n}\alpha}^+ - \hat{B}_{\bar{n}\alpha} \right) \right| \Psi_0^{(ex)} \right\rangle \quad (3.2)$$

of the difference of creation and annihilation operators $\hat{B}_{\bar{n}\alpha}^+$ and $\hat{B}_{\bar{n}\alpha}$ corresponds to exciton Hamiltonian $\hat{H}^{(ex)}$, which for all considered systems has the standard form :

$$\hat{H}^{ex} = \sum_{\bar{n}\alpha} E_{\bar{n}\alpha} \hat{B}_{\bar{n}\alpha}^+ \hat{B}_{\bar{n}\alpha} + \frac{1}{2} \sum_{\substack{\bar{n}\alpha \\ \bar{m}\beta}} V_{\bar{n}\alpha\bar{m}\beta} \left(\hat{B}_{\bar{n}\alpha}^+ + \hat{B}_{\bar{n}\alpha} \right) \left(\hat{B}_{\bar{m}\beta}^+ + \hat{B}_{\bar{m}\beta} \right) \quad (3.3)$$

From (3.1) it is seen, that microscopic calculation $\Delta\chi_{il}^\perp(\mathbf{k}, \omega, \{C_\alpha^{\nu(\alpha)}\})$ is directly connected with the configuration averaging of the production of molecular currents by Green function (3.2). This procedure is easy to carry out if one expresses molecular characteristics $E_{\bar{n}\alpha}$, $V_{\bar{n}\alpha\bar{m}\beta}$ and $\mathbf{J}_{\bar{n}\alpha}(\mathbf{k})$ through configurationally dependent random quantities $\eta_{\bar{n}\alpha}^{\nu(\alpha)}$. The latter ones have the following meanings:

$\eta_{\bar{n}\alpha}^{\nu(\alpha)} = 1$, if in site $\bar{n}\alpha$ there is point defect of the $\nu(\alpha)$ -th sort and in $\eta_{\bar{n}\alpha}^{\nu(\alpha)} = 0$ in any other case. Molecular characteristics are expressed through random quantities in the following way:

$$\mathbf{J}_{\bar{n}\alpha}(\mathbf{k}) = \sum_{\nu(\alpha)=1}^{r(\alpha)} \mathbf{J}_\alpha^{\nu(\alpha)}(\mathbf{k}) \eta_{\bar{n}\alpha}^{\nu(\alpha)}, \quad (3.4a)$$

$$E_{\bar{n}\alpha} = \sum_{\nu(\alpha)=1}^{r(\alpha)} E_{\alpha}^{\nu(\alpha)} \eta_{\bar{n}\alpha}^{\nu(\alpha)}, \quad (3.4b)$$

$$V_{\bar{n}\alpha\bar{m}\beta} = \sum_{\nu(\alpha), \mu(\beta)=1}^{r(\alpha, \beta)} \eta_{\bar{n}\alpha}^{\nu(\alpha)} W_{\bar{n}\alpha\bar{m}\beta}^{\nu(\alpha)\mu(\beta)} \eta_{\bar{m}\beta}^{\mu(\beta)} \quad (3.4c)$$

At that

$$\sum_{\nu(\alpha)=1}^{r(\alpha)} \eta_{\bar{n}\alpha}^{\nu(\alpha)} = 1 \quad . \quad (3.4d)$$

where $\mathbf{J}_{\bar{n}\alpha}^{\nu(\alpha)}(\mathbf{k})$ and $E_{\alpha}^{\nu(\alpha)}$ are matrix element of Fourier transform of current density operator and exciton energy of the α -th molecule of $\nu(\alpha)$ -th sort; value $W_{\bar{n}\alpha\bar{m}\beta}^{\nu(\alpha)\mu(\beta)}$ corresponds to Coulomb interaction of point defect of $\nu(\alpha)$ -th sort in site $n(\alpha)$ with point defect of $m(\beta)$ -th sort in site $\bar{m}\beta$ (if in sites $n(\alpha)$ or $m(\beta)$ there is a vacancy, the corresponding $W_{\bar{n}\alpha\bar{m}\beta}^{\nu(\alpha)\mu(\beta)}$ equals zero); $r(\alpha)$ is the number of molecular groups in α -th sublattice, each of which corresponds to a particular sort of corresponding defects.

From formula (3.4d) it follows that concentrations $C_{\alpha}^{\nu(\alpha)}$ are subject to constraint

$$\sum_{\nu(\alpha)=1}^{r(\alpha)} C_{\alpha}^{\nu(\alpha)} = 1 \quad (3.5)$$

Function $\left\langle \left\langle \hat{B}_{\bar{n}\alpha}^{+} - \hat{B}_{\bar{n}\alpha} \left| \hat{B}_{\bar{m}\beta}^{+} - \hat{B}_{\bar{m}\beta} \right. \right\rangle \right\rangle_{\omega}^r$, appearing in (3.1), is expressed through the quantities $E_{\bar{n}\alpha}$, $V_{\bar{n}\alpha\bar{m}\beta}$ and for exciton Hamiltonian $\hat{H}^{(ex)}$ quadratic in operators $\hat{B}_{\bar{n}\alpha}^{+}$, $\hat{B}_{\bar{m}\alpha}$ can be determined accurately. Employing the method of motion equations [23] we obtain:

$$\left\langle \left\langle \hat{B}_{\bar{n}\alpha}^+ - \hat{B}_{\bar{n}\alpha} \left| \hat{B}_{\bar{m}\beta}^+ - \hat{B}_{\bar{m}\beta} \right. \right\rangle \right\rangle_{\omega}^r = -2\hbar \left[\left[\hbar^2 \omega^2 - (\hat{E} + 2\hat{V}) \hat{E} \right]^{-1} (\hat{E} + 2\hat{V}) \right]_{\bar{n}\alpha\bar{m}\beta} \quad (3.6)$$

where $\left[\hat{E} \right]_{\bar{n}\alpha\bar{m}\beta} = E_{\bar{n}\alpha} \delta_{\bar{n}\bar{m}} \delta_{\alpha\beta}$, $\left[\hat{V} \right]_{\bar{n}\alpha\bar{m}\beta} = V_{\bar{n}\alpha\bar{m}\beta}$.

Formulas (3.1) and (3.4a) show that finding the frequency and concentration dependences $\Delta\chi_{jl}^{\perp}(\mathbf{k}, \omega, \{C_{\alpha}^{v(\alpha)}\})$ is immediately connected to configuration averaging of the following production of random quantities:

$$\Phi_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} \equiv \eta_{\bar{n}\alpha}^{v(\alpha)} \cdot \left\langle \left\langle \hat{B}_{\bar{n}\alpha}^+ - \hat{B}_{\bar{n}\alpha} \left| \hat{B}_{\bar{m}\beta}^+ - \hat{B}_{\bar{m}\beta} \right. \right\rangle \right\rangle_{\omega}^r \cdot \eta_{\bar{m}\beta}^{\mu(\beta)}.$$

Taking into account (3.46) and (3.4b) $\hat{P}\Phi_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)}$ can be written in the following way:

$$\begin{aligned} \hat{P}\Phi_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} &= 2\hbar \left\{ \left(C_{\alpha}^{v(\alpha)} / E_{\alpha}^{v(\alpha)} \right) \delta_{\bar{n}\bar{m}} \delta_{\alpha\beta} \delta_{v(\alpha)\mu(\beta)} - \hbar^2 \omega^2 \sqrt{g_{\alpha}^{v(\alpha)}(\omega) / E_{\alpha}^{v(\alpha)}} \right\} \cdot \\ &\cdot \left\{ \left[\hat{\sigma}^{-1}(\omega, \{C_{\alpha}^{v(\alpha)}\}) - \hat{u}(\omega) \right]^{-1} \right\}_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} \times \sqrt{g_{\beta}^{\mu(\beta)}(\omega) / E_{\beta}^{\mu(\beta)}} \end{aligned} \quad (3.7)$$

Here

$$\begin{aligned} \hat{\sigma}(\omega, \{C_{\alpha}^{v(\alpha)}\}) &= \hat{P}\hat{\eta} \sum_{q=0}^{\infty} \left[\hat{u}(\omega) \hat{Q}\hat{\eta} \right]^q, \\ \left[\hat{u}(\omega) \right]_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} &= \sqrt{2E_{\alpha}^{v(\alpha)} g_{\alpha}^{v(\alpha)}(\omega)} W_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} \sqrt{2E_{\bar{m}\beta}^{\mu(\beta)} g_{\bar{m}\beta}^{\mu(\beta)}(\omega)}, \\ g_{\alpha}^{v(\alpha)}(\omega) &= \left[\hbar^2 \omega^2 - (E_{\alpha}^{v(\alpha)})^2 \right]^{-1}, \quad \hat{Q} = \hat{I} - \hat{P}, \\ [\hat{\eta}]_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} &= \eta_{\bar{n}\alpha}^{v(\alpha)} \delta_{\bar{n}\bar{m}} \delta_{\alpha\beta} \delta_{v(\alpha)\mu(\beta)}, \end{aligned}$$

\hat{I} is the unity operator. Derivation of formula (3.7) relies on the projection operator formalism [21], which is convenient for the above relation of $E_{n\alpha}$, $V_{n\alpha m\beta}$, $\mathbf{j}_{n\alpha}(\mathbf{k})$ to the random quantities $\eta_{\bar{n}\alpha}^{v(\alpha)}$. Since the four-index matrix (3.7) has the same structure

as the two-index mass operator for the averaged resolvent [22], $\hat{\sigma}(\omega, \{C_\alpha^{v(\alpha)}\})$ can be evaluated by the same methods and approximations as in Refs. [21,22]. In the simplest one-site approximation (approximation of average T-matrix) the specified function is diagonal by all indices and has the following form:

$$\left[\hat{\sigma}(\omega, \{C_\alpha^{v(\alpha)}\}) \right]_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} = C_\alpha^{v(\alpha)} \delta_{\bar{n}\bar{m}} \delta_{\alpha\beta} \delta_{v(\alpha)\mu(\beta)} \quad (3.8)$$

After the substitution of (3.7) into (3.1) and simple transformation we will receive the microscopic expression for $\chi_{jl}^\perp(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\})$ in the excitonic spectral range:

$$\begin{aligned} \chi_{jl}^\perp(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\}) &= (1 - \omega_0^2 / \omega^2) \delta_{jl} + \frac{8\pi}{v_0 \omega^2} \sum_{\alpha=1}^{\kappa} \sum_{v(\alpha)}^{r(\alpha)} \tilde{J}_\alpha^{jv(\alpha)}(-\mathbf{k}) \tilde{J}_\beta^{l\mu(\beta)}(\mathbf{k}) C_\alpha^{v(\alpha)} - \\ &- \frac{8\pi\hbar^2}{v_0} \sum_{\alpha, \beta=1}^{\kappa} \sum_{v(\alpha)}^{r(\alpha)} \sum_{\mu(\beta)}^{r(\beta)} \tilde{J}_\alpha^{jv(\alpha)}(-\mathbf{k}) \tilde{J}_\beta^{l\mu(\beta)}(\mathbf{k}) F_{\alpha\beta}^{v(\alpha)\mu(\beta)}(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\}), \end{aligned} \quad (3.9)$$

where

$$\begin{aligned} F_{\alpha\beta}^{v(\alpha)\mu(\beta)}(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\}) &= \sqrt{g_\alpha^{v(\alpha)}(\omega) g_\beta^{\mu(\beta)}(\omega)} \left\{ \left[\hat{\sigma}^{-1}(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\}) - \hat{u}(\mathbf{k}, \omega) \right]^{-1} \right\}_{\alpha\beta}^{v(\alpha)\mu(\beta)}, \\ \left[\hat{\sigma}^{-1}(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\}) \right]_{\alpha\beta}^{v(\alpha)\mu(\beta)} &= \sum_n \left[\hat{\sigma}(\omega, \{C_\alpha^{v(\alpha)}\}) \right]_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} \exp(-i\mathbf{k}\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}), \\ \left[\hat{u}(\mathbf{k}, \omega) \right]_{\alpha\beta}^{v(\alpha)\mu(\beta)} &= \sum_n \left[\hat{u}(\omega) \right]_{\bar{n}\alpha\bar{m}\beta}^{v(\alpha)\mu(\beta)} \exp(-i\mathbf{k}\mathbf{r}_{\bar{n}\alpha\bar{m}\beta}), \end{aligned}$$

$\tilde{J}_\alpha^{jv(\alpha)}(\mathbf{k}) = J_\alpha^{jv(\alpha)}(\mathbf{k}) \sqrt{E_\alpha^{v(\alpha)}}$, $v_0 = V/N$. Expression for plasma frequency is given in Ref. [13] (where one can also find the reason of the appearance of not depending on \mathbf{k} first term in (3.9)).

The obtained expression for $\chi_{jl}^\perp(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\})$ in the form of Eq. (3.9) has a universal form for all randomly disordered molecular crystals and it is found within the framework of a singular model, without any simplifying assumptions (unlike in

Ref. [24], where function $\Phi_{\vec{n}\alpha\vec{m}\beta}^{v(\alpha)\mu(\beta)}\left(\omega, \{C_\alpha^{v(\alpha)}\}\right)$ was taken in approximate form). The calculation of characteristics of normal electromagnetic waves with the help of (3.9) can be carried out taking into account and ignoring spatial dispersion.

3.3 Calculation of rotatory power and analysis of its dependence on the point defect concentration.

Using (3.9) the microscopic expression for rotatory power $\rho\left(\mathbf{s}, \omega, \{C_\alpha^{v(\alpha)}\}\right)$ of the system under consideration can be simplified to the form:

$$\begin{aligned} \rho\left(\mathbf{s}, \omega, \{C_\alpha^{v(\alpha)}\}\right) = & i \frac{2\pi}{v_0} \left(\frac{\omega}{c}\right)^2 S^t \sum_{\alpha, \beta}^{\kappa} \sum_{v(\alpha)}^{r(\alpha)} \sum_{\mu(\beta)}^{r(\beta)} \left\{ \hbar \sqrt{E_\alpha^{v(\alpha)} / E_\beta^{\mu(\beta)}} \left[\mathbf{s} \times \mathbf{P}_\alpha^{v(\alpha)} \right]^l \mathcal{Q}_\beta^{\mu(\beta)tl} \cdot \right. \\ & \cdot \left[F_{\alpha\beta}^{v(\alpha)\mu(\beta)}\left(\mathbf{s}, \omega, \{C_\alpha^{v(\alpha)}\}\right) + F_{\beta\alpha}^{\mu(\beta)v(\alpha)}\left(\mathbf{s}, \omega, \{C_\alpha^{v(\alpha)}\}\right) \right] \left[\mathbf{s} \times \mathbf{P}_\alpha^{v(\alpha)} \right]^l \mathcal{Q}_\beta^{\mu(\beta)tl} \cdot \\ & \cdot \left[F_{\alpha\beta}^{v(\alpha)\mu(\beta)}\left(\mathbf{s}, \omega, \{C_\alpha^{v(\alpha)}\}\right) + F_{\beta\alpha}^{\mu(\beta)v(\alpha)}\left(\mathbf{s}, \omega, \{C_\alpha^{v(\alpha)}\}\right) \right] + \sqrt{E_\alpha^{v(\alpha)} E_\beta^{\mu(\beta)}} \left[\mathbf{P}_\alpha^{v(\alpha)} \times \mathbf{P}_\beta^{\mu(\beta)} \right] \cdot \\ & \left. \cdot \mathbf{s} \left(\partial F_{\alpha\beta}^{v(\alpha)\mu(\beta)}\left(\mathbf{k}, \omega, \{C_\alpha^{v(\alpha)}\}\right) / \partial k^t \right) \right\}_{|\mathbf{k}|=0}, \end{aligned} \quad (3.10)$$

where repeated indices imply summation, c is the speed of light. The last term in Eq. (3.10) for crystal with primitive lattice is zero (as well as for nonideal systems). Expression (3.10) becomes simpler for systems where the only point defects are vacancies and for orientationally disordered systems (with or without vacancies). The last is due to the fact that in the former case $E_\alpha^{v(\alpha)} \equiv E_\alpha$, $g_\alpha^{v(\alpha)}(\omega) \equiv g_\alpha(\omega)$ for all $v(\alpha)$, while in the latter case $E_\alpha^{(1)} = E_\alpha^{(2)} = \dots = E_\alpha^{r(\alpha)}$, $g_\alpha^{(1)}(\omega) = g_\alpha^{(2)}(\omega) = \dots = g_\alpha^{r(\alpha)}(\omega)$. Concentration dependence of rotatory power and peculiarities of electro-optical activity for various strengths of external electric field is studied for such systems in Ref. [25].

When studying natural optical activity of nonideal systems considerable interest lies in concentration dependence of rotatory power $\rho(\mathbf{s}, \omega, \{C_a^{v(\alpha)}\})$ in various frequency domains. Below such dependence is illustrated on the example of molecular crystals with primitive lattice that have only vacancies as point defects. In this case $\kappa=1$, and formula (3.5) is of the form: $C + C^v = 1$ (C is molecule concentration, C^v is vacancies concentration). For such system the rotatory power in single-site approximation (see formula (3.8)) is described by the formula:

$$\rho(\mathbf{s}, \omega, C^v) = i \frac{4\pi}{v_0 \hbar c^2} s'_l Q'' [\mathbf{s} \times \mathbf{P}]_l \frac{\hbar^2 \omega^2 (1 - C^v)}{\hbar^2 \omega^2 - E_0^2 - 2E_0 W(\mathbf{s}) (1 - C^v)}. \quad (3.11)$$

Here E_0 and $W(\mathbf{s})$ are correspondingly molecule excitation energy and Fourier transform of the matrix of resonance molecular interaction in an ideal crystal. Formula (3.11) shows that for the systems under study there are two frequency areas that have significantly different corresponding values of rotatory power on C . The graphs of the specified dependences for $\hbar\omega_1 = 3,63 \cdot 10^4 \text{ cm}^{-1}$ and $\hbar\omega_2 = 4,00 \cdot 10^4 \text{ cm}^{-1}$ are shown (in relative units) in figure 3.1.

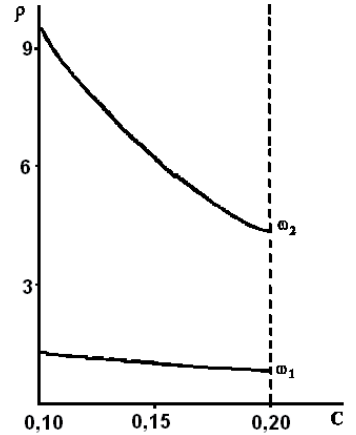


Fig. 3.1. Concentration dependence of rotatory power for two frequency areas.

The calculations use representative values of E_0 and $W(\mathbf{s})$, which are $3 \cdot 10^4 \text{ cm}^{-1}$ and $0,7 \cdot 10^4 \text{ cm}^{-1}$ correspondingly. Figure 3.1 shows that in the first frequency area optical activity is less sensitive to the changes in vacancies concentration than the second one. Such behavior of rotatory power is connected to renormalization of exciton energy and its dependence on C^v . Relatively wide (compared with $\rho(\mathbf{s}, \omega_1, C^v)$, at that $\hbar\omega_1 > \sqrt{E_0^2 - 2E_0 W}$) range of values $\rho(\mathbf{s}, \omega_2, C)$

(one should note that $\hbar\omega_2 > \sqrt{E_0^2 - 2E_0W}$) testifies to the possibility of significant changes in NOA connected with vacancies presence.

It is evident that in the case of systems with complex lattices there are more (than in the given example) frequency areas, whose rotatory power dependences on C are qualitatively different from each other. From Ref. [24] it follows that the above microscopic analysis of NOA is easily generalized for the case of mixed and orientationally disordered systems with vacancies. For those the characteristics and peculiarity of rotatory power are determined by dependences $\mathbf{J}_{\bar{n}\alpha}(\mathbf{k})$, $E_{\bar{n}\alpha}$, $V_{\bar{n}\alpha m\beta}$ on the configurations of both vacancies and impurity molecules. As even for the systems of one type these dependences can be significantly different (like with the case of orientationally disordered crystals with various reorientation angles [18]), the role of the examined defects in NOA of corresponding media can be essentially different.

4. Optical activity of a nonideal dielectric 1D-superlattice

4.1. Introduction

Investigations of the optical properties of dielectric superlattices [3,4,26-28] occupy a prominent place in condensed matter physics. These studies are mainly stimulated by the needs of electrical engineering and electronics in layer structures and, therefore, by the need to simulate composite materials with the given properties. The study of the spatial dispersion effects is of considerable interest, since they are a valuable and often unique tool for revealing subtle structural features of spatially dispersive media [11]. In connection with this, the development of the theory of the aforementioned effects that reveal of their specificity in multilayer systems and finding corresponding frequency characteristics based on model representations are of current interest. The urgency of these investigations is evident because, at present, there are many complicated organic complexes and polymers that are optically active due to the features of their structure or the optical activity of molecules that enter into

their composition [10, 29]. The problem of finding normal electromagnetic waves (which are necessary to calculate gyrotropy characteristics) in spatially dispersive superlattices is unsolved. Nevertheless, it is evident that, in the case when the thickness of the layers of a multilayer system considerably exceeds the characteristic scales of spatial dispersion, the corresponding values can be calculated approximately if the contribution of each layer to the gyrotropy is considered to be independent. With regard to natural optical activity (NOA), this means that the knowledge of only the specific rotation angles of the layer $\rho_{n\alpha}(\omega)$ (ω is the light frequency, n is the number of a unit cell of the one-dimensional lattice, α is the number of a layer in the cell) and the concentration of extraneous layers (if they are present) is sufficient to find the specific rotation angle $\rho(\omega)$ of the polarization plane. In this work, the aforementioned approximation is used to calculate $\rho(\omega)$ of a one-dimensional superlattice that contains randomly distributed extraneous layers, which differ from the corresponding layers of an ideal system in the physico-chemical composition and/or in the thickness. The expression for $\rho(\omega)$ obtained below makes it possible to numerically simulate the concentration dependence of the optical activity, which was done for a SiO_2 -liquid-crystal multilayer system. With the known microscopic expressions for $\rho_{n\alpha}(\omega)$, the approach described in this work makes it possible to reveal and identify the predominating gyrotropy mechanisms, which are important for experiment, to establish the relation of the above-mentioned function with microcharacteristics of the medium (such as the dipole, quadrupole, and magnetic dipole moments of structural units), and to find the corresponding frequency characteristics. Superlattices that consist of macroscopically homogeneous systems with pointlike defects are of special interest. In this case, $\rho(\omega)$ is a function of not only the concentration of defects in extraneous layers, but also of the concentration of pointlike defects. In this work, numerical calculations were performed for the frequency dispersion of the optical activity of a nonideal superlattice made up of layers of impurity molecular crystals. The latter condition widens the possibilities of simulating composite materials because it permits one to continuously vary their

gyrotropy properties and the disordering parameters (with respect to the composition or/and thickness).

4.2 Specific rotation angle of light polarization plane in a nonideal 1D superlattice

In keeping with the above mentioned approximate approach, the rotation angle $\tilde{\rho}(\omega)$ of the polarization plane of light has the following form:

$$\tilde{\rho}(\omega) = \sum_{n=1}^N \sum_{\alpha=1}^{\sigma} \rho_{n\alpha}(\omega) a_{n\alpha} . \quad (4.1)$$

In (4.1), $\rho_{n\alpha}(\omega)$ and $a_{n\alpha}$ are the configuration-dependent thickness of the α -th layer of the n -th unit cell and the specific rotation angle of the plane of polarization of light caused by this layer, respectively, and σ is the number of layers of the unit cell. It is evident that formula (4.1) is valid for the case of light propagation along layer optical axes perpendicular to the plane of the layers of a nonideal, topologically ordered, one-dimensional superlattice consisting of N unit cells. We suppose that the number of cells N is so large that it is possible to form a configuration that averages in correspondence with the general principles of physics of disordered systems [30, 31].

According to [30, 31], the experimentally measured rotation angle is defined as $\hat{P}\tilde{\rho}(\omega) \equiv \langle \tilde{\rho}(\omega) \rangle$, where \hat{P} is the configuration averaging operator [22] that acts on the configuration-dependent function $\tilde{\rho}(\omega)$. There are two types of disorders in the considered nonideal 1D superlattice; therefore, there are two types of configuration dependences. The disordering of the first type is caused by the fact that the superlattice contains extraneous (defective) layers that differ from the corresponding layers of an ideal system in the physico-chemical composition (the corresponding configuration-dependent quantity is $\rho_{n\alpha}(\omega)$). The second disordering is caused by the presence of layer defects that differ from an ideal superlattice in thickness (the corresponding configuration-dependent quantity is $a_{n\alpha}$). It is evident that these factors do not depend on each other.

The connection of $\rho_{n\alpha}(\omega)$ and $a_{n\alpha}$ with the configuration-dependent stochastic variables $\eta_{n\alpha}^{\mu(\alpha)}$ and $\eta_{n\alpha}^{\nu(\alpha)}$ has the following form:

$$\rho_{n\alpha}(\omega) = \sum_{\mu(\alpha)=1}^{r(\alpha)} \rho_{\alpha}^{\mu(\alpha)} \eta_{n\alpha}^{\mu(\alpha)}, \quad a_{n\alpha} = \sum_{\nu(\alpha)=1}^{s(\alpha)} a_{\alpha}^{\nu(\alpha)} \eta_{n\alpha}^{\nu(\alpha)}, \quad (4.2)$$

and

$$\sum_{\mu(\alpha)=1}^{r(\alpha)} \eta_{n\alpha}^{\mu(\alpha)} = 1, \quad \sum_{\nu(\alpha)=1}^{s(\alpha)} \eta_{n\alpha}^{\nu(\alpha)} = 1. \quad (4.3)$$

where $\eta_{n\alpha}^{\mu(\alpha)} = 1$, if the α -th layer of the n -th unit cell is the layer of $\mu(\alpha)$ -type ($\mu(\alpha) = 1.2 \dots r(\alpha)$) and $\eta_{n\alpha}^{\mu} = 0$ - in any other case; $\eta_{n\alpha}^{\nu(\alpha)} = 1$, if thickness of the α -th layer of the n -th unit cell equals $a_{\alpha}^{\nu(\alpha)}$ ($\nu(\alpha) = 1.2 \dots s(\alpha)$) and $\eta_{n\alpha}^{\nu} = 0$ - in any other case. $\rho_{\alpha}^{\mu(\alpha)}(\omega)$ is the specific rotation angle of the α -th layer of $\mu(\alpha)$ type. Now and then index μ enumerates layers of variable composition, ν - of variable thickness.

Using formulas (1)–(3) and the averaging rules [22,30,31] for $\langle \tilde{\rho}(\omega) \rangle$, we obtain the following expression:

$$\begin{aligned} \langle \tilde{\rho}(\omega) \rangle = N \sum_{\alpha=1}^{\sigma} & \left[\rho_{\alpha}^{(1)}(\omega) a_{\alpha}^{(1)} + a_{\alpha}^{(1)} \sum_{\mu(\alpha)=1}^{r(\alpha)} \Delta \rho_{\alpha}^{\mu(\alpha)}(\omega) C_{\alpha}^{\mu(\alpha)} + \rho_{\alpha}^{(1)}(\omega) \sum_{\nu(\alpha)=1}^{s(\alpha)} \Delta a_{\alpha}^{\nu(\alpha)} C_{\alpha}^{\nu(\alpha)} + \right. \\ & \left. + \sum_{\mu(\alpha)=1}^{r(\alpha)} \sum_{\nu(\alpha)=1}^{s(\alpha)} \Delta \rho_{\alpha}^{\mu(\alpha)}(\omega) \Delta a_{\alpha}^{\nu(\alpha)} C_{\alpha}^{\mu(\alpha)} C_{\alpha}^{\nu(\alpha)} \right], \quad (4.4) \end{aligned}$$

$\Delta \rho_{\alpha}^{\mu(\alpha)} = \rho_{\alpha}^{\mu(\alpha)} - \rho_{\alpha}^{(1)}$, $\Delta a_{\alpha}^{\nu(\alpha)} = a_{\alpha}^{\nu(\alpha)} - a_{\alpha}^{(1)}$ as well as $C_{\alpha}^{\mu(\alpha)}$, $C_{\alpha}^{\nu(\alpha)}$ are the concentrations of defects of layers that differ from the layers of the basic compound (for which the layers of the first sort are conventionally taken) in the composition and/or thickness, respectively. The first summand in (4.4) corresponds to the rotation angle of the light polarization plane of an ideal 1D superlattice made up of layers of the first sort. The second summand is caused by the disordering of the superlattice

with respect to the composition; it vanishes in the absence of variations in the composition. The third summand reflects disordering in the thickness (in the absence of this disordering, this summand vanishes). The last summand is caused by the simultaneous disordering of the superlattice with respect to both the composition and the thickness of the layers. The absence of at least one of the disorderings leads to the vanishing of the fourth summand in (4.4). Each of the four summands of (4.4) in the sum over α has the meaning of the rotation angle per one unit cell. Unlike $\rho_{n\alpha}^{\mu(\alpha), \nu(\alpha)}$ (which is measured in degrees/unit of length), these angles are measured in degrees. To illustrate the obtained results, let us consider NOA in a nonperfect 1D superlattice with two elementary layers in a unit cell the first layer of which is SiO_2 (α -quartz) ($\rho_1^{(1)} = 780$ degrees/mm, $\lambda = 152.3$ nm) and the second layer is a model liquid crystal ($\rho_2^{(1)} = 2000$ degrees/mm). Let the first sublattice contain impurity layers that only differ from the basic (ideal) sublattice in thickness and let both the thickness and the composition vary in the second liquid crystal sublattice ($\rho_2^{(2)} = 2500$ degrees/mm). Let the concentration and thickness of the layer of the basic compound in the first and the second sublattices be denoted as $C_1^{(1)}$, $a_1^{(1)}$ and $C_2^{(1)}$, $a_2^{(1)}$, respectively, and those of the impurity as $C_{1C(T)}^{(2)}$, $a_1^{(2)}$ and $C_{2C(T)}^{(2)}$, $a_2^{(2)}$ (index $C(T)$ denotes the variations in the impurity layers in the composition or in the thickness). Based on formula (4.4), simple transformations permit one to obtain the following concentration dependence of the specific rotation angle of the light polarization plane in the considered two_sublattice nonideal 1D superlattice ($C_{1C}^{(2)} = 0$) as follows:

$\rho = \rho \left(C_{1C(T)}^{(2)}, C_{2C(T)}^{(2)} \right) \equiv \langle \tilde{\rho} \rangle / N \langle d \rangle$. Here, $\langle d \rangle$ is the averaged period of the cell of the 1D superlattice, i.e., $\langle d \rangle = a_1^{(1)} + a_2^{(1)} + \left(a_1^{(2)} - a_1^{(1)} \right) C_{1T}^{(2)} + \left(a_2^{(2)} - a_2^{(1)} \right) C_{2T}^{(2)}$. The diagram of the concentration dependence of the specific rotation angle $\rho = \rho \left(C_{1C(T)}^{(2)}, C_{2C(T)}^{(2)} \right)$ of the light polarization plane in the studied nonideal superlattice is presented in Figs. 1

and 2. Figure 1 reflects the variation of the superlattice layers only in thickness, and Fig. 2 presents the function $\rho = \rho(C_{2C}^{(2)}, C_{2T}^{(2)})$.

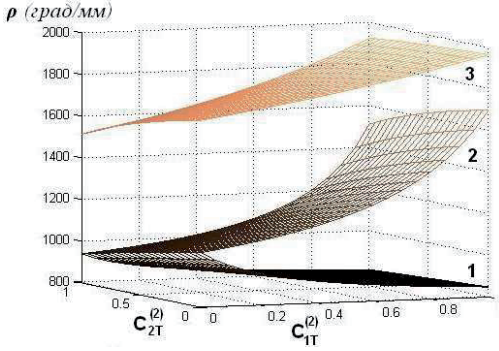
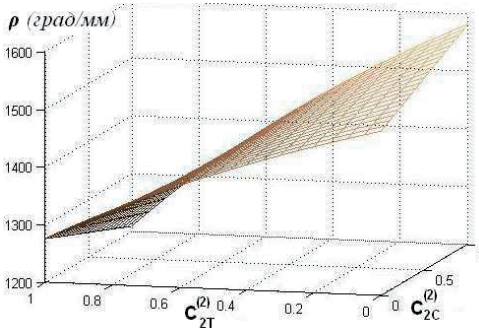


Fig. 4.1. Concentration dependence of the specific rotation angle $\rho = \rho(C_{1T}^{(2)}, C_{2T}^{(2)})$ of the light polarization plane in the studied nonideal superlattice: $a_2^{(2)} / a_2^{(1)} = 0.5$; $a_2^{(1)} / a_1^{(1)}$ and $a_1^{(2)} / a_1^{(1)}$ are, respectively, (1) 0.3 and 0.5, (2) 0.3 and 0.1, and (3) 3 and 0.1.

a)



b)

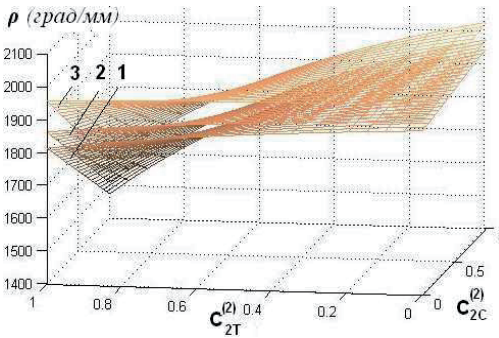


Fig. 4.2. Concentration dependence of the specific rotation angle $\rho = \rho(C_{2C}^{(2)}, C_{2T}^{(2)})$ of the light polarization plane in studied nonideal superlattice: (a) $a_2^{(1)} / a_1^{(1)} = 3$, $a_1^{(2)} / a_1^{(1)} = 5$, $a_2^{(2)} / a_2^{(1)} = 0.5$, and $C_{1T}^{(2)} = 0.3$; (b) $a_2^{(1)} / a_1^{(1)} = 10$, $a_1^{(2)} / a_1^{(1)} = 0.1$, and $a_2^{(2)} / a_2^{(1)} = 0.5$; concentration $C_{1T}^{(2)}$ is (1) 0.1, (2) 0.3, and (3) 0.9.

Moreover, Fig. 4.2a shows a considerable variation of the specific angle ρ with variation of the relative thicknesses $a_2^{(1)} / a_1^{(1)}$, $a_1^{(2)} / a_1^{(1)}$, $a_2^{(2)} / a_2^{(1)}$ of the layers at constant $C_{1T}^{(2)}$, and Fig. 2b permits one to conclude that, for particular values of the relative thicknesses of the layers, the value of depends rather weakly on the variation of the superlattice composition.

The study of these composite materials (polymer and LC superlattices) is important due to the variety of their functional properties in a wide application area [3,4,10,29]. Below, by the example of systems with only one type of disorder (in thickness), we study the NOA of two-layer superlattices made up of layers of binary mixed (the first layer) and orientation-disordered (the second layer) molecular crystals. In the following section of this work, using the microscopic expressions obtained previously [32] for $\rho_{na}(\omega)$, features of the frequency-concentration dependence $\rho(\omega)$ related to the relative positions of excitonic energies of both sublattices and with layer thicknesses are studied for the excitonic region of the spectrum.

4.3. Microscopic theory of optical activity of imperfect superlattices

The expression for the specific rotation angle

$$\rho(\omega, C_{mi}, C_{or}, C^{(1)}, C^{(2)}) \equiv \langle \tilde{\rho}(\omega) \rangle / N$$

of the polarization plane of linearly polarized light passing through a nonideal two-layer molecular superlattice (the first layer is a mixed molecular crystal and the second layer is orientationally disordered), as follows from (4.4), has the form

$$\rho(\omega, C_{mi}, C_{or}, C^{(1)}, C^{(2)}) = \rho_{mi}(\omega, C_{mi}) (a_1 + \Delta a_1 C^{(1)}) + \rho_{or}(\omega, C_{or}) (a_2 + \Delta a_2 C^{(2)}), \quad (4.5)$$

where $\rho_{mi}(\omega, C_{mi})$, $\rho_{or}(\omega, C_{or})$ - specific rotation angles for mixed and orientationally disordered molecular crystals; a_1, a_2 - thicknesses of first and second

layers of corresponding perfect superlattice; C_{mi}, C_{or} - concentrations of isotopic impurities and orientationally disordered molecules, $C^{(1)}, C^{(2)}$ - concentrations of foreign layers in relevant sublattices with thickness differing $\Delta a_1, \Delta a_2$ from those of the perfect system. Here each of the summands has the sense of the rotation angle (by a corresponding sublattice) per one unit cell.

In the most general case, the specific rotation angle $\rho(\omega, \{C\})$ for a multicomponent topologically ordered impurity (including both mixed and orientationally disordered) molecular crystal with a primitive lattice in the exciton region of the spectrum is, within the framework of single-level model, of the form:

$$\rho(\omega, \{C\}) = \frac{2\pi\omega^2\hbar}{vc^2} \sum_{\nu, \mu=1}^r A^{\nu\mu} F^{\nu\mu}(\omega, \{C\}). \quad (4.6)$$

Here v - volume of the unit cell of molecular crystal, r - number of molecular groups, each relating to a definite ν -th or μ -th type of molecules; $\{C\} \equiv C_1, C_2, \dots, C_r$. Quantities $A^{\nu\mu}$ and $F^{\nu\mu}$ are:

$$A^{\nu\mu} = \left[(E_\nu/E_\mu)^{1/2} P_{0f}^{iv} Q_{f0}^{z\mu} + (E_\mu/E_\nu)^{1/2} Q_{of}^{ziv} P_{f0}^{l\mu} \right] e_{ilz}, \quad (4.7)$$

$$F^{\nu\mu}(\omega, \{C\}) = \frac{1}{\sqrt{\hbar^2\omega^2 - (E_\nu)^2}} \left\{ \left[\hat{\sigma}^{-1}(\mathbf{s}, \omega, \{C\}) - \hat{U}(\mathbf{s}, \omega) \right]^{-1} \right\}^{\nu\mu} \frac{1}{\sqrt{\hbar^2\omega^2 - (E_\mu)^2}}. \quad (4.8)$$

In formulas (4.7), (4.8) e_{ilz} is the Levy - Civita tensor, $\mathbf{s} = \mathbf{k} / k$, E_ν, E_μ - molecular excitation energies, P_{of}^ν, P_{f0}^μ - matrix elements of the dipole-moment operators, $Q_{0f}^{ziv}, Q_{f0}^{zi\mu}$ - matrix elements of operators derived from operators of molecular currents $J_{f0}^{v(\mu)}(\mathbf{k})$ of the ν -th and μ -th molecular types, respectively,

$$Q_{0f}^{ziv} = -Q_{f0}^{zi\mu} = i \left[\partial J_{f0}^{ziv}(\mathbf{k}) / \partial k^z \right] \big|_{\mathbf{k}=0}. \quad (4.9)$$

Matrix elements of locator function $\hat{\sigma}$ and matrix \hat{U} (proportional to matrix $W^{\nu\mu}$ of the resonance intermolecular interaction) are of the form:

$$\left[\hat{\sigma}^{\nu\mu}(\mathbf{s}, \omega\{C\}) \right]^{v\mu} = \left\{ \sum_n \left[\hat{\sigma}(\omega, \{C\}) \right]_{nm}^{v\mu} \exp(-i\mathbf{k} \cdot \mathbf{r}_{nm}) \right\}_{|\mathbf{k}|=0}, \quad (4.10)$$

$$\left[\hat{U}^{\nu\mu}(\mathbf{s}, \omega) \right]^{v\mu} = \left\{ \sum_n \left[\hat{U}(\omega) \right]_{nm}^{v\mu} \exp(-i\mathbf{k} \cdot \mathbf{r}_{nm}) \right\}_{|\mathbf{k}|=0}. \quad (4.11)$$

Matrix elements of locator function in the nodal representation $\left[\hat{\sigma}(\omega, \{C\}) \right]_{nm}^{v\mu}$ (in (10)) are expressed through matrix $\hat{U}(\omega)$ as follows:

$$\left[\hat{\sigma}(\omega, \{C\}) \right]_{nm}^{v\mu} = \left\{ \hat{\Pi} \hat{\eta} \sum_{p=0}^{\infty} \left[\hat{U}(\omega) (\hat{I} - \hat{\Pi}) \hat{\eta} \right]_{nm}^p \right\}. \quad (4.12)$$

Expression (4.12) is written in terms of projection operator $\hat{\Pi}$, \hat{I} - unit operator. $[\hat{\eta}]_{nm}^{v\mu} = \eta_n^\mu \delta_{nm} \delta_{v\mu}$, configuration-dependent random unit η_n^μ equals 1, if in node n there is an ν -type molecules, and it is zero in any other case. Matrix elements $[\hat{U}(\omega)]^{v\mu}$ relate to the matrix of resonance intermolecular interaction (figuring in exciton Hamiltonian [33]) as:

$$\left[\hat{U}(\omega) \right]_{nm}^{v\mu} = 2 \left[E_\nu E_\mu \left(\hbar^2 \omega^2 - (E_\nu)^2 \right)^{-1} \left(\hbar^2 \omega^2 - (E_\mu)^2 \right)^{-1} \right]^{1/2} W_{nm}^{v\mu} \quad (4.13)$$

For binary systems, values $F^{11}, F^{22}, F^{12}, F^{21}$ in mononodal approximation are:

$$F^{v\mu} = \frac{1}{\varphi(\omega, C_1, C_2)} \begin{bmatrix} C_1 (\hbar^2 \omega^2 - E_2^2 - 2C_2 E_2 W^{22}) & 2C_2 C_1 (E_1 E_2)^{1/2} W^{12} \\ 2C_2 C_1 (E_1 E_2)^{1/2} W^{21} & C_2 (\hbar^2 \omega^2 - E_1^2 - 2C_1 E_1 W^{11}) \end{bmatrix}, \quad (4.14)$$

$$\begin{aligned} \varphi(\omega, C_1, C_2) \equiv & \left\| \hat{\sigma}^{-1}(\omega, \{C\}) - \hat{U}(\omega) \right\| = \hbar^4 \omega^4 - \hbar^2 \omega^2 (2E_1 C_1 W^{11} + 2E_2 C_2 W^{22} + \\ & + E_1^2 + E_2^2) + 4E_1 E_2 C_1 C_2 (W^{11} W^{22} - W^{12} W^{21}) + 2E_1 E_2 W^{11} C_1 + 2E_2 E_1 W^{22} C_2 + E_1^2 E_2^2 \end{aligned} \quad (4.15)$$

$$C_1 + C_2 = 1.$$

In expression (4.15) C_1 is the molecular concentration for the basic substance of a mixed or orientationally disordered molecular crystal, and $C_2 \equiv C_{mi(or)}$.

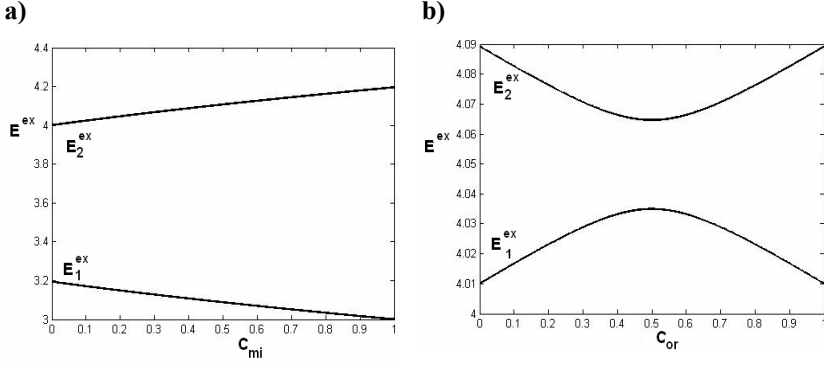


Fig.4.3. Concentration dependence exciton energy levels of a molecular crystal: a) $E_1^{ex}(C_{mi})$, $E_2^{ex}(C_{mi})$; b) $E_1^{ex}(C_{or})$, $E_2^{ex}(C_{or})$. E_1^{ex} , E_2^{ex} is given in units of 10^{-4} cm^{-1} .

Zeros of function $\varphi(\omega, C_1, C_2)$ define renormalized exciton energy levels $E_1^{ex}(C_{mi(or)})$, $E_2^{ex}(C_{mi(or)})$ of a molecular crystal, each lying in a definite interval of values (see Fig.4.3).

4.4. Results and discussion

The character of the frequency-concentration dependence of rotator power is specified by the relative position of exciton energies and by values of parameters $A^{\nu\mu}$, C_{mi} , C_{or} , $C^{(1)}$, $C^{(2)}$, $a_1, \Delta a_1$, $a_2, \Delta a_2$. It follows from formula (4.15) that for frequencies of light running over one of the regions of $E_1^{ex}(C_{mi(or)})$, $E_2^{ex}(C_{mi(or)})$ values (see Fig.4.3) the rotator power $\rho(\omega, C_{mi}, C_{or}, C^{(1)}, C^{(2)})$ is divergent if damping is neglected. For numerical calculations we limit ourselves with the two cases out of variety of types of the optical-activity concentration dependence for certain frequencies of electromagnetic wave. In the first case, the energy of

electromagnetic excitation is in the interval $E_1^{ex}(C_{mi}) < \hbar\omega < E_1^{ex}(C_{or})$, and the corresponding concentration dependence of the specific rotation angle of light passing through the layered structure under consideration is illustrated in Fig.4. In the second case (Fig.4.5), which is described by inequalities $E_2^{ex}(C_{mi}) < \hbar\omega < E_2^{ex}(C_{or})$, for definite values of frequencies of light, the energy of electromagnetic excitation lies in the neighborhood of exciton resonance.

In particular calculations it is considered that for a mixed molecular crystal it can be, within a good accuracy, assumed [14] $W^{11} \approx W^{12} \approx W^{21} \approx W^{22} \equiv W_{mi}$, $\vec{P}_{of}^1 \approx \vec{P}_{f0}^2$, $\hat{Q}_{of}^1 \approx \hat{Q}_{f0}^2$ (as a consequence, all $A^{vu} = const \equiv A^{mi}$). For orientationally disordered molecular crystals $E_1 \approx E_2$ [15].

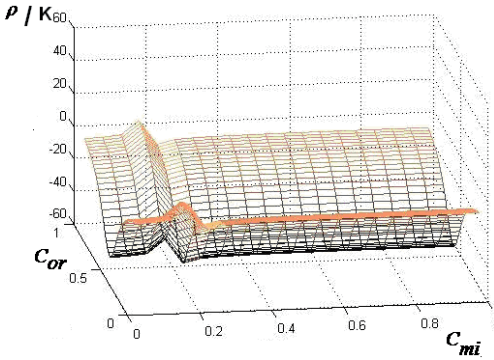


Fig. 4.4. Concentration dependence of the specific rotation angle $\rho = \rho(C_{or}, C_{mi})$ for $\hbar\omega = 3.99 \cdot 10^4 \text{ cm}^{-1}$.

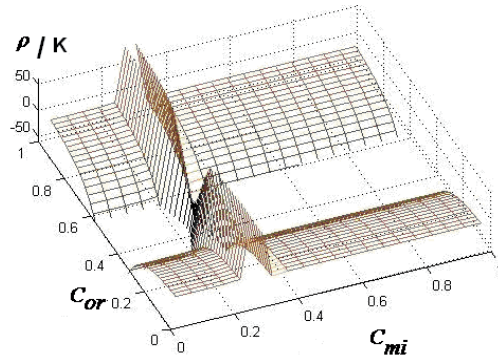


Fig. 4.5. Concentration dependence of the specific rotation angle $\rho = \rho(C_{or}, C_{mi})$ for $\hbar\omega = 4.06 \cdot 10^4 \text{ cm}^{-1}$.

It is assumed that for a mixed molecular crystal $E_1^{mi} = 3 \cdot 10^4 \text{ cm}^{-1}$, $E_2^{mi} = 4 \cdot 10^4 \text{ cm}^{-1}$, $W_{mi} = 2 \cdot 10^3 \text{ cm}^{-1}$, whereas for the orientationally disordered one - $E_1^{or} \approx E_2^{or} = 4.01 \cdot 10^4 \text{ cm}^{-1}$, $W_{or}^{11} \approx W_{or}^{22} = 0.8 \cdot 10^3 \text{ cm}^{-1}$, $W_{or}^{12} \approx W_{or}^{21} = 0.3 \cdot 10^3 \text{ cm}^{-1}$ and $A_{or}^{12} / A^{mi} \approx A_{or}^{21} / A^{mi} = 3$, $A_{or}^{11} / A^{mi} \approx A_{or}^{22} / A^{mi} = 2$. Here, the numerical simulation was done for the following values of concentration of heterogeneous superlattice layers: $C^{(1)} = 0.2$, $C^{(2)} = 0.3$, and for relative thicknesses $\Delta a_1 / a_1 = \Delta a_2 / a_2 = 0,1$ ($a_1 = a_2$). The concentration dependences of the specific rotation angle of light are graphically shown in Fig 4.4-4.6 in relative units ρ / K , where $K = \frac{2\pi a_1 A^{mi}}{v c^2 \hbar}$ (see (4.6)). The frequency dependences of the specific rotation angle $\rho = \rho(\omega, C_{or}, C_{mi})$ for specific values of concentration C_{or} and C_{mi} is illustrated in Fig. 4.6 (a, b).

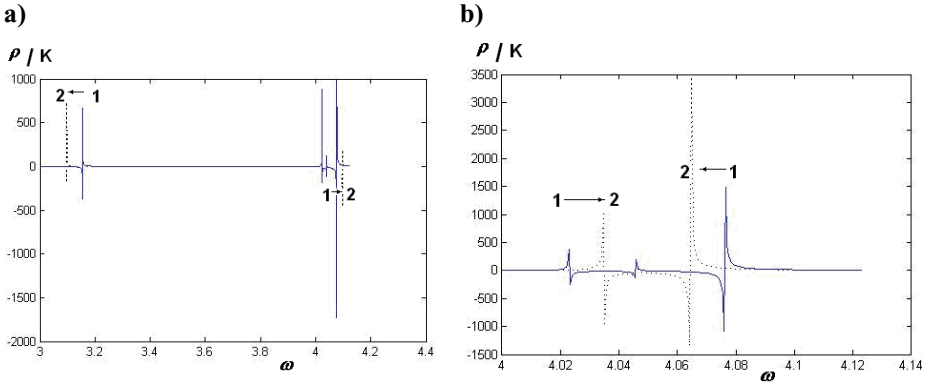


Fig. 4.6. Frequency dependences of the specific rotation angle $\rho = \rho(\omega, C_{or}, C_{mi})$ for values of concentration C_{or} and C_{mi} equal, respectively, to: case a) 1 – 0.2 and 0.17; 2 – 0.2 and 0.45; case b) 1 – 0.2 and 0.2; 2 – 0.5 and 0.2 (ω is given in units of 10^4 cm^{-1}).

It is seen that changes in concentration C_{mi} with C_{or} constant result in mutual removal of resonance frequencies (case a), whereas changes in C_{or} with C_{mi} constant – in their approaching (case b). So the curves $\rho = \rho(\omega, C_{or}, C_{mi})$ are determined by

the specific dependence of exciton energy levels $E_1^{ex}(C_{or(mi)}), E_2^{ex}(C_{or(mi)})$ on the concentration of point defects considered superlattice (see Fig.4.3).

Conclusion

The theory developed in the first three sections of the present review can serve as a solid basis for further investigation of behavior of gyrotropy under external actions such as electric and magnetic fields, mechanical stress etc. Of particular interest are low-dimensional structures and superlattices.

Studies on optical properties of dielectric superlattices comprise a significant part of the modern-day condensed matter physics [3,4,36]. Their importance is enhanced by the constantly growing demands of electronics and electrical engineering for high-performance layered materials with prescribed optical characteristics. The related phenomena of spatial dispersion are of particular interest as they provide a valuable and very frequently the sole tool for revealing fine structural features in spatially dispersive media [37]. This calls for theoretical models, permitting to elucidate the said phenomena in multilayers and would hopefully allow to calculate the corresponding frequency characteristics. The problem appears even more topical in the light of the vast diversity of synthesized complex organic polymers, which happen to be optically active either due to their structural features or due to optical activeness of the comprising molecules [1,2].

In contrast to the previous works, where microscopic analysis was given for optical activity of ideal crystals (see e.g. [38]), here we utilize microscopic approach to study the optical activity of imperfect 1D-multilayer materials. Our method is helpful for numerical simulations of the frequency-concentration dependence of the specific rotation angle of molecular superlattices in the exciton region of spectrum. The system is modeled to be comprised by two sublattices with parameters typical of orientationally disordered and mixed molecular crystals. Types of gyrotropy typical of different disorder types are studied. A 1D-superlattice with point defect containing

layers is investigated. The developed theory opens additional possibilities for simulation of optically active multilayer composite materials.

Finally, in more general terms, the results presented in this work contribute to development of our views of the processes in externally influenced matter. They can also be helpful in technological and engineering developments.

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