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Analytical Models of Systems with Photoinduced Spiral Spatial Microstructure: Interaction with Polarized Optical Radiation

O. D. Asenchik and E. G. Starodubtsev

Gomel State Technical University
October av. 48, 246746 Gomel, Belarus
Fax: + 375 - 232 - 479165; email: starodub@ggtu.belpak.gomel.by

Abstract

Optical model of a photochromic medium with spiral spatial microstructure of centres distribution, which is formed at the record in photoactive materials of polarization state and intensity of two interfering light beams, is developed. The expressions for dielectric permittivity tensor are derived and the corresponding boundary problem for the photochromic layer is investigated. Some possibilities of bistability and multistability regimes for such layers are considered.

1. Creation of Systems with Photoinduced Spiral Spatial Microstructure

Important examples of media with spiral spatial microstructure are cholesteric liquid crystals (CLC). On the other hand, there is a possibility of creating spiral symmetric structures by the optical method: at interference of opposing coherent beams in layers from photochemically active materials. Such materials are polymeric solid solutions of photochromic dyes (indigoide colours, anthracenes, spiropirans, etc.) in polymeric glass (polymethyl methacrylate, polystyrene, etc.) [1,2]. Under polarized optical excitation owing to various photophysical and photochemical processes (saturation of metastable states, photoisomeric change, photoreduction, and others), in these materials dichroism and (or) birefringence is photoinduced. Thus, systems with optically controllable character of the anisotropy arise. The change of the polarized radiation characteristics allows gaining materials with given properties of optical anisotropy, which are not masked by the natural anisotropy (as a rule, initially the samples are isotropic). In this connection, it is interesting to use CLC optics methods and results for the investigation of optical properties of the photochromic layers.

The aims of the paper are: 1) development of optical model of a medium with spiral spatial microstructure of photochromic centres distribution, which is formed at the record in the material of polarization state and intensity of two interfering coherent light beams; 2) investigation of linear and nonlinear interaction of polarized optical radiation with layers from such materials.

Let us accept the following kinetic scheme describing dynamics of photophysical and photochemical processes in a plane-parallel photochromic layer: 1) excitation of molecules of photosensitive component D with rate $R(\Omega_d)$ due to absorption of photons; 2) the later conversion of the molecules D to a photoproduct DP (at monochromatic excitation $R=\sigma_D(\Omega_d)I$, where $\sigma_D(\Omega_d)$ is the interaction cross-section on the excitation frequency of the dipole oscillator with orientation Ω_d, I is photons flux density). Solving kinetic equations describing such scheme and making the corresponding averaging on the ensemble, one can derive dependencies of concentrations $[D](t,\Omega_d),[DP](t,\Omega_d)$ on time and orientation. Function of distribution $f(\Omega_d)$ of non-rotating photochromic centres on orientations Ω_d of the transitions dipole moments can be written in the form

$$f(\Omega_d)=[D](t,\Omega_d)/[D](0,\Omega_d)=f(\sigma_D(\Omega_d)It). \qquad (1)$$

According to quantum mechanics representations: $I\sigma_D(\Omega) \sim \sigma_D |\mathbf{E}\cdot\mathbf{d}|^2$, where **E** is the strength of the recording electric field in the medium, **d** is unit vector of the transition dipole moment of the photochromic center (in the spherical coordinate system $\mathbf{d} = (\sin\vartheta\cos\varphi, \sin\vartheta\cos\varphi, \cos\theta)$).

Let us make the consideration in frames of the following assumptions: 1) optical density of photochromic layer on the record frequency is small, that corresponds to the optically thin layer; 2) absorption of photon by the photoactive center occurs due to dipole-dipole transition between the main and electronically excited states; 3) initial photochromic layer is homogeneous and optically isotropic, with scalar permittivity ε_0.

The recording field in the medium is $\mathbf{E} = \mathbf{E}_+ + \mathbf{E}_-$, where vectors $\mathbf{E}_+$ and $\mathbf{E}_-$ characterize plane elliptically polarized waves with wave vectors $\mathbf{k}_+ = k(-\sin\alpha, 0, \cos\alpha)$ и $\mathbf{k}_- = k(-\sin\alpha, 0, -\cos\alpha)$ (axis Z of the square coordinate system is perpendicular to the layer boundaries), and

$$\mathbf{E}_+ = -E_0(\cos\alpha, -ib, \sin\alpha)\exp(-i\mathbf{k}_+\cdot\mathbf{r}), \quad \mathbf{E}_- = -rE_0(\cos\alpha, ib, -\sin\alpha)\exp(-i\mathbf{k}_-\cdot\mathbf{r}). \tag{2}$$

In Eq. (2) α is the angle of waves incident on the layer, $E_0 = E(1+b^2)^{-1/2}$, b is the ellipticity parameter ($b = 0$ at linear and $b = \pm 1$ at circular polarizations); E is amplitude of wave $\mathbf{E}_+$; r is the relation of amplitudes of waves $\mathbf{E}_-$ and $\mathbf{E}_+$.

2. Optical Properties of the Structure: Boundary Problem

Permittivity tensor of the considered system can be written in the form ($i, j = 1,2,3$)

$$\varepsilon_{ij} = \varepsilon_0 + \frac{\Delta\varepsilon}{4\pi}\int_0^{4\pi} d_i d_j f(\Omega_d)d\Omega_d = \varepsilon_0 + \frac{\Delta\varepsilon}{4\pi}\sum_{m=1}^{\infty}\int_0^{4\pi} d_i d_j a_m(H)(\sigma_D(\Omega_d)/\sigma_D)^m d\Omega_d, \tag{3}$$

where function $f(\Omega_d)$ in Eq. (1) has been expanded in the Taylor series about factor $\sigma_D(\Omega_d)\phi It$ with coefficients $a_m = f^{(m)}(0)H^m/m!$ which do not depend on angle Ω_d, $H = \sigma_D I\phi t$ is the exposure and $\Delta\varepsilon$ is depth of permittivity modulation; ϕ is quantum yield of conversion.

The general form of the expression determining dependence of quantity $\sigma_D(\Omega_d)/\sigma_D$ on the angles of the spherical coordinate system is relatively awkward

$$\begin{aligned}\sigma_D(\Omega_d)/\sigma_D &= \cos^2 K\big((1-r)^2\cos^2\vartheta\sin^2\alpha + (1-r^2)\cos\vartheta\sin\vartheta\cos\varphi\sin 2\alpha\big) + \\ &+ \sin^2\vartheta\big((1+r)^2\cos^2\alpha\cos^2\varphi + b^2(1-r)^2\sin^2\varphi\big) + \\ &+ \sin^2 K\big((1+r)^2\cos^2\vartheta\sin^2\alpha + (1-r^2)\cos\vartheta\sin\vartheta\cos\varphi\sin 2\alpha\big) + \\ &+ \sin^2\vartheta\big((1-r)^2\cos^2\alpha\cos^2\varphi + b^2(1+r)^2\sin^2\varphi\big) + 2br\cos\alpha\sin 2K\sin^2\vartheta\sin 2\phi,\end{aligned} \tag{4}$$

where $K = kz\cos\alpha$. So let us consider important particular cases when Eq. (4) is significantly simplified. At linear on exposure H expansion of function $f(\Omega_d)$ substitution of Eq. (4) to Eq. (3) gives

$$\begin{aligned}&\varepsilon_{11} = \varepsilon_0 + (\Delta\varepsilon/15)\big[(1+r^2)(2+b^2+\cos 2\alpha) + 2r\cos(2K)(1-b^2+2\cos 2\alpha)\big]a_1, \\ &\varepsilon_{12} = \varepsilon_{21} = \varepsilon_0 + (4\Delta\varepsilon/15)(br\sin(2K)\cos\alpha)a_1, \quad \varepsilon_{13} = \varepsilon_{31} = \varepsilon_0 - (\Delta\varepsilon/15)((r^2-1)\sin(2\alpha))a_1, \\ &\varepsilon_{22} = \varepsilon_0 + (\Delta\varepsilon/15)\big[(1+r^2)(1+3b^2) + 2r\cos(2K)(\cos(2\alpha)-3b^2)\big]a_1, \\ &\varepsilon_{33} = \varepsilon_0 + (\Delta\varepsilon/15)\big[(1+r^2)(2+b^2-\cos(2\alpha)) - 2r\cos(2K)(1+b^2-2\cos(2\alpha))\big]a_1, \\ &\varepsilon_{23} = \varepsilon_{32} = 0.\end{aligned} \tag{5}$$

In case of arbitrary parameters describing the recording waves, tensor ε form depends on the photoreaction type and transformation extent. Even in simple model situations the derived expressions are very awkward. Moreover, analysis of Eq. (5) leads to some general conclusions: 1) $\varepsilon_{23}=\varepsilon_{32}=0$; 2) $\varepsilon_{13}=\varepsilon_{31}\neq 0$ if $r\neq 0$, and quantities ε_{13} и ε_{31} do not depend on z; 3) at $\alpha\neq 0$ there is a harmonic dependence $\varepsilon_{33}(z)$, which can be absent at the relations between parameters r, b, α: $b=\sqrt{2\cos(2\alpha)-1}$ or r=0.

In another particular case of α=0, b=1, r=1 (opposing propagation of the circularly polarized waves with the equal amplitudes) we have from Eq. (4): $\sigma_D(\Omega_d)/\sigma_D=(\cos(K+\varphi)\sin(\vartheta))^2$. Substitution of Eq. (4) in Eq. (3) gives ($(2/15)\Delta\varepsilon(H)\rightarrow\Delta\varepsilon$ at $H\rightarrow\infty$)

$$\varepsilon=\begin{pmatrix}\varepsilon_0+(2-\cos(2kz))\Delta\varepsilon(H) & \Delta\varepsilon(H)\sin(2kz) & 0\\ \Delta\varepsilon(H)\sin(2kz) & \varepsilon_0+(2+\cos(2kz))\Delta\varepsilon(H) & 0\\ 0 & 0 & \varepsilon_0\end{pmatrix}. \qquad (6)$$

Due to specific dependence of relation $\sigma_D(\Omega_d)/\sigma_D$ on angles ϑ and φ the form of the permittivity tensor in Eq. (6) does not depend on particular mechanism of the photoreaction which leads to the record of the electromagnetic field state and extent of photochemical transformations in the medium.

The form of Eq. (6) corresponds exactly to the permittivity tensor of uniaxial CLC with the spiral step $H=1/(2k)$ [3]. This analogy allows to use the known analytical expressions for the proper waves and solutions of the boundary problems for CLC [3] at analysis of optical properties of the exposed photochromic layers. In particular, one can show that the circularly polarized proper waves in the layer experience a selective diffractional reflection. In general case, it is represented to be possible investigation of the polarized waves transmission and reflection in a wide frequency range (and not only near the Bragg frequency, as at the traditional approach to investigations of polarization holograms [4]), and with account of the multiple reflections on the layer boundaries.

The calculated at using results [3] dependencies of the transmissivity of the circularly polarized light incident on the layer with thicknesses $50\lambda_0$ (1), $100\lambda_0$ (2), $430\lambda_0$ (3), and $730\lambda_0$ (4) on wavelength λ are shown in Fig. 1 (λ_0=0.44μm). The parameters values $\varepsilon_0=3$, $\Delta\varepsilon=0.01$, b=1 are taken, indexes of refraction beyond the layer are equal to 1. It is seen from Fig. 1, that with increasing the layer thickness and at small tunes from λ_0 the system is similar to the Fabry-Perot interferometer, despite of small Fresnel's reflectivities on the layer boundaries. Mutual influence of the multibeam interference and diffraction on the periodic structure cause such behaviour of the system.

Obviously, the permittivity tensor characterizing the structure, has the form similar to Eq. (6) not only in the case of parallel propagation of the recording beams ($\alpha=0$). For example, as it follows from Eq. (5), at $b^2=2\cos 2\alpha-1$ and $r=1$ this tensor has the form as in Eq. (6) with the structure period $H=1/(2k\cos\alpha)$. That allows to control the structure period at a choice of the record geometry.

The nonlinear properties of the Fabry-Perot interferometers were studied enough explicitly. One of the most interesting features of such systems is bistable and multistable responses at high incident intensities. In connection with the marked similarity of the explored system to the Fabry-Perot interferometer, it is interesting to investigate opportunities of occurrence of multistable regimes in our system. Taking into account, that the photochromic layer with spiral spatial microstructure has specific polarization properties, the analysis of the mentioned problem can have a practical interest.

Let under incident of the probe radiation on the layer of material characterized by the tensor Eq. (6) the averaged permittivity ε varies on quantity $\Delta\varepsilon_{nl}=\varepsilon_2 P$, where P is intensity, and parameter ε_2 is determined by the concrete mechanism of a nonlinearity. Figure 2 illustrates the graphical solution of the transcendental equation determining transmittance of the layer $T(\varepsilon)$:

$$T(\varepsilon)=(\varepsilon-\varepsilon_0)/(\varepsilon_2 P). \qquad (7)$$

The wavelength of the radiation differed from quantity λ_0, determining the spiral microstructure period, on quantity $\lambda_0\delta/3$, where $\delta = 4\Delta\varepsilon/(15\varepsilon_0 + 8\Delta\varepsilon)$ (see Fig. 1). From Fig. 2 it is seen, that at various intensities (straight lines 1, 2, 3, 4 correspond to increasing intensity) of incident radiation, Eq.(9) has a various number of the solutions. That corresponds to the multistable response of the system in the range of intensities relevant to curves 1-3 (Fig. 2). Taking $\varepsilon_2 = 10^{-3}\, cm^2/kW$ (typical value for the thermal nonlinearity [5]), from Fig. 2 one can estimate the minimal intensity for observation of the bistability and multistability effects in the considered layers: $P = 5kW/cm^2$.

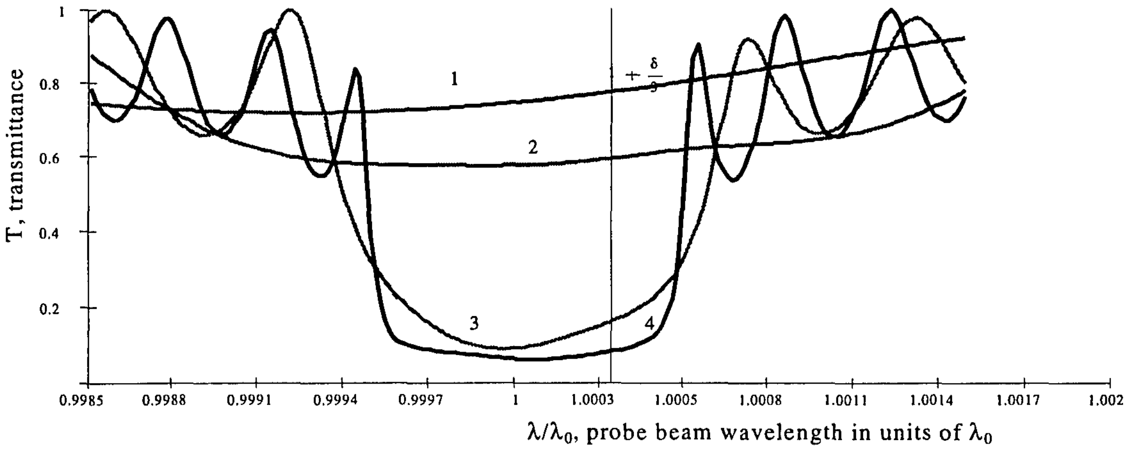

Fig. 1

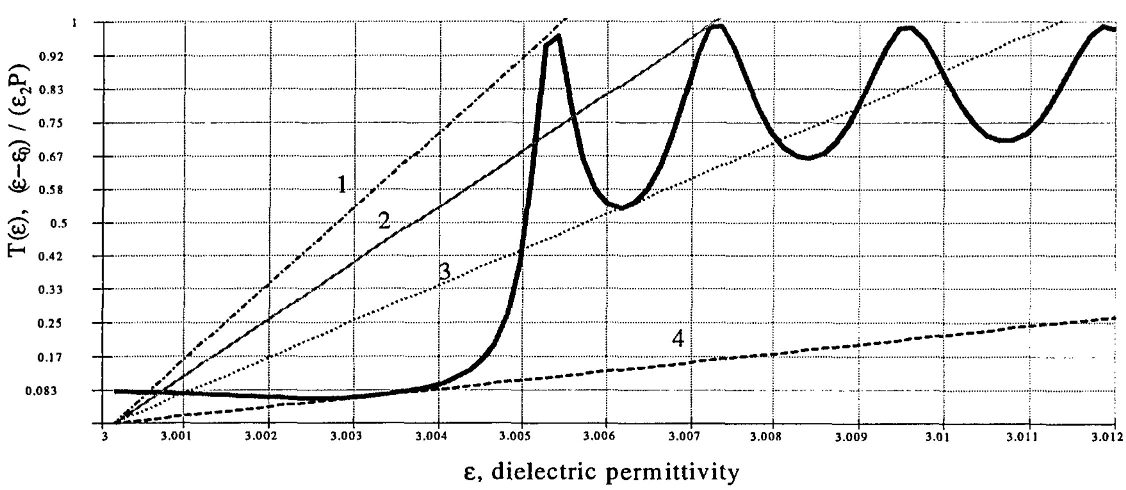

Fig. 2 $\varepsilon_0 = 3$; $\Delta\varepsilon$=0.01; $b = 1$; $\Delta\varepsilon_{nl}$=0.0053 (1), 0.0071 (2), 0.0112 (3), 0.045 (4)

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