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SOLITON MECHANISM OF OPTICAL ANISOTROPY PHOTOINDUCTION IN LAYERED MOLECULAR STRUCTURES

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Abstract. A theory of the influence of linearly polarized light on ordering in layered molecular systems is given.

We consider a two-dimensional system of anisotropic molecules whose centers form a square lattice. Its orientational energy per unit area has the form

$$F = I(\nabla\varphi)^2 - \frac{1}{2}A\cos 4\varphi \tag{1}$$

where $\varphi(\vec{r})$ is the angle between the large molecule axis and the coordinate axis X,I and A are positive energy constants. We will discuss the orientational distributions that characterized by fourth order symmetry axis (optical isotropic distributions). The simplest example of such a distribution is

$$\varphi = -\arctan\frac{\sinh\alpha y}{\sinh\alpha x}, \quad \alpha^2 = 2A/I \tag{2}$$

It is seen that this vortex-like structure may be regarded as an intersection of two mutually perpendicular $\frac{\pi}{2}$-kinks situated on the lines $y = \mp x$. When a molecular layer is irradiated by polarized light $\vec{E}(\vec{r},t) = \vec{E}\cos(\vec{Q}\vec{r} - \omega t)$ (ω is the electronic eigenfrequency of the layer) some molecules occur excited and in the orientational energy there are the terms that depend on the number of excited molecules

$$F_{int}(\vec{r}) = -\frac{1}{2}A_{ex}N(\vec{r})\cos 4\varphi(\vec{r})$$

where $A_{ex} \sim (\mu_{ex} - \mu_{gr})^2$ ($\mu_{ex}(\mu_{gr})$ is the molecular dipole moment in the excited (ground) state), $N(\vec{r})$ is the probability for finding the r-th molecule in the excited state. The order in the irradiated layer can be described by the equations [1]:

$$I\nabla^2\varphi(\vec{r}) - (A + A_{ex}N(\vec{r}))\sin 4\varphi(\vec{r}),$$

$$N(\vec{r}) = N_{ex}\cos^2(\varphi(\vec{r}) - \psi) \tag{3}$$

where $N_{ex} = (Ed)^2/(\Gamma\gamma)$ is the ratio of the number of excited molecules to the total number of molecules (d is the transition dipole moment, $\Gamma(\gamma)$ is the transversal (longitudinal) rate of the excited state), ψ is the angle between $\vec{E}$ and the X axis. The analysis of (3) shows that under the light action the walls $y = \pm x$ are moved to the new positions: in the vicinity of vortex center at $\rho \leq 1$ $(x = \rho\cos\theta, \quad y = \rho\sin\theta)$

$$\theta = \pm\frac{\pi}{4} \mp \frac{N_{ex}}{32}(\alpha\rho)^2 \ln(\alpha\rho)\cos 2\psi;$$

far from the center at $\vec{r}$ satisfying inequalities

$$1 < \sinh^2 \alpha x + \sinh^2 \alpha y < \exp(\frac{2}{N_{ex}})$$

$$\sinh \alpha y = \pm(1 + \frac{N_{ex}}{8}\cos 2\psi \ln(\sinh^2 \alpha x))\sinh \alpha x.$$

At $x, y \rightarrow \infty$ the kinks are moved almost freely with constant velocity [1]

$$v = -\frac{5c^2\alpha}{24\lambda\sqrt{2}} N_{ex}\cos 2\psi$$

where the parameter λ characterizes the dissipation process in the orientation system, c is the velocity of libration waves.The estimations show that e.g. in molecular Langmuir-Blodgett films $v = 10^1 - 10^2 cm/s$. Thus as one areas reduce the others grow and optical anisotropy in the layer arises.

References

[1] Yu.B.Gaididei and A.S.Trofimov Journ.of Mol.Electronics 5(1989),239.