Nature of the aberration pattern formed as a result of self-focusing of a light beam caused by reorientation of the director in liquid crystals

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Experimental and theoretical investigations were made of the characteristics of the aberration pattern (rotation of the plane of polarization, time dependences of the intensity of the central spot in the pattern, and elongation of the aberration rings) formed as a result of self-focusing of a light beam caused by the Fréedericksz transition.

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In our earlier investigations[1-3] relating to the discovery and investigation of the Fréedericksz effect in an optical field we drew attention to the fact that a nonlinear self-focusing lens, which appears in a nematic liquid crystal under the influence of a laser beam, exhibits strong aberrations (a characteristic ring structure appears on a distant screen). However, our earlier papers did not give a detailed description (and even less of an analysis) of the nature of the aberration pattern observed on reorientation of the director and the report given below is intended to at least partly fill this gap.

We shall consider the influence of the Frank elastic constants on the self-focusing of a light beam, the relationship between oscillations of the intensity of the central spot of an aberration pattern and formation of aberration rings, and the polarization effects which appear as a result of orientational self-focusing.

Our investigation was carried out on homotropically oriented octyl cyanobiphenyl (OCBP) and methoxybenzylidene butylaniline (MBBA) crystals, which were placed at a constriction of an argon-ion laser beam ($\lambda = 4880$ Å); the apparatus used is shown in Fig. 1. In front of a crystal we placed a double Fresnel rhomb for rotation of the plane of polarization of the light beam incident on a crystal and behind the crystal there was a film Polaroid and a screen for an analysis and display of the light beam transmitted by the crystal. The screen was usually placed at a distance of $d \approx 200$ cm from the crystal. A photodiode with a stop was sometimes placed in front of the screen and the signal from the photodiode was recorded with a one-coordinate plotter. The photodiode measured the intensity of the central spot in the aberration pattern. The diameter of the stop in front of the photodiode was of the order of or less than the diameter of the central spot.

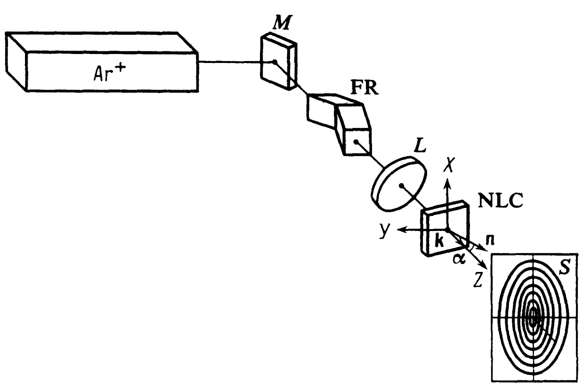

FIG. 1. Schematic diagram of the apparatus. Here, M is a rotatable mirror, FR is a Fresnel rhomb, L is a lens, NLC is a neumatic liquid crystal, S is a screen, and Ar^+ is an argon-ion laser.

The reliability of the results obtained in investigations of nematic liquid crystals are largely determined by the degree of their orientation, particularly in the region crossed by a laser beam. It should be noted that the quality of the orientation of an investigated crystal could be determined from the scattering of laser radiation in a nematic liquid crystal representing, as is well known, an optically turbid medium. A conoscopic pattern, used to judge the quality of a crystal, was displayed on a screen located close to a crystal (≈ 1.5–2 cm) and in this case the laser beam power was relatively low (≈ 30–50 mW). This provided an extremely simple method for the control of the orientation of the investigated nematic liquid crystals.

EXPERIMENTAL RESULTS

1. Rotation of the plane of polarization

The polarization of the radiation transmitted by a liquid crystal differed from the polarization of the incident radiation and it was found that the polarization effects were a function of the angle of incidence α of the laser radiation on a crystal, of the angle θ representing nonlinear deviation of a ray from the beam axis, and of the angle Ψ measured in the plane of the screen from the line of intersection of the screen with the plane of polarization of the incident beam. Only the rays characterized by $\Psi = 0$ retained the polarization of the incident beam. In the case of other rays one could distinguish two cases.

a) $\alpha \gtrsim 20°$. In this case on illumination of a crystal with a linearly polarized (in the YZ plane) laser beam it was found that the polarization of the rays transmitted by the crystal was also linear, but the plane of polarization was rotated. The angle of rotation φ depended on α, θ, and Ψ.

b) $0 \leqslant \alpha \leqslant 20°$. In this case the polarization of the rays transmitted by the crystal was elliptic. The position of the major semiaxis of the polarization ellipse was also governed by the angles α, θ, and Ψ.

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The angle of rotation φ of the plane of polarization (of the major semiaxis of the polarization ellipse) was maximal for $\Psi = \pi/2$. In the case of constant values of α and Ψ, the angle of rotation increased on increase in θ, i.e., on increase in the distance of an aberration ring from the center. Rotation of the plane of polarization at diametrically opposite points of a ring was in opposite directions.

Figure 2 shows the experimental dependences of φ on θ for $\alpha = 30°$ and $\alpha = 0°$ in the case of the OCBP crystal ($\Psi = \pi/2$).

2. Intensity of the central spot in the aberration pattern

It was found visually that the intensity of the central spot of the aberration pattern varied in the course of formation of the pattern and the nature of this variation was a function of the position of the crystal relative to the constriction of the laser beam. We used the photodiode described above to determine the time dependences of the intensity at the center of the aberration pattern for three positions of the crystal relative to the beam constriction (Fig. 3). We found that the changes in the intensity did not start immediately after the beginning of illumination of the crystal (Fig. 3) but only after a certain delay time T_d which was a function of the power of the incident radiation.[1] When the crystal was in a diverging beam (beyond the constriction, Fig. 3a), the intensity at the beam center increased compared with the initial value, reached a maximum of amplitude which depended on the distance δ from the crystal to the constriction, and then fell. The general nature of the pattern was retained on approach of the crystal to the contriction (Fig. 3b), but the increase in the intensity became less; at the constriction itself and in front of it (in a converging beam, Fig. 3c) the increase became practically zero. Oscillations were exhibited at the stage of the fall of the intensity in all three cases. The number of these oscillations was always equal to the number of the aberration rings, which was typical of given experimental conditions. The threshold power needed for the appearance of the aberration pattern naturally increased when the crystal was displaced out of the constriction.

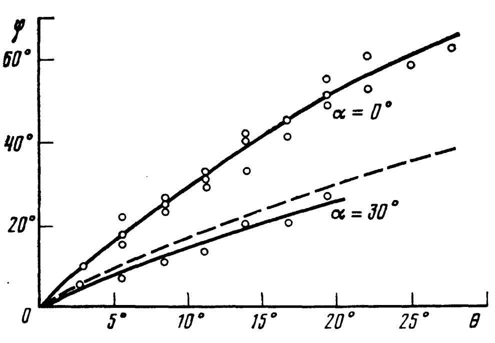

FIG. 2. Dependences of the angle of rotation of the plane of polarization (for $\alpha = 30°$, linear polarization) and of the angle of rotation of the major axis of the polarization ellipse ($\alpha = 0°$, elliptic polarization) on the angle of deviation of a ray θ (for rays deviated in a vertical plane). The results were obtained for an OCBP crystal, $L \approx 150\,\mu$ thick, at $t = 36$ °C. The points and the continuous curve are the experimental results, and the dashed curve is calculated.

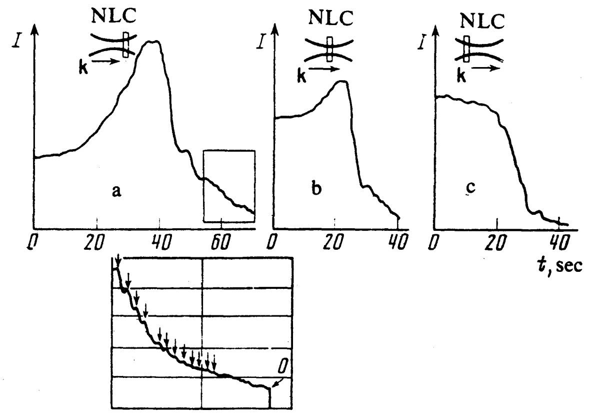

FIG. 3. Time dependences of the intensity at the center of an aberration pattern obtained for different positions of an OCBP crystal ($L = 150\,\mu$, $t = 36$ °C) relative to a constriction in the incident laser beam ($f = 170$ mm): a) nematic liquid crystal (NLC) located in the diverging part of the beam ($\delta = 5$ mm); b) crystal located near the constriction; c) crystal located in the converging part of the beam ($\delta = 5$ mm). In all cases the angle of incidence is $\alpha = 0$.

3. Elongation of the aberration pattern

The aberration rings were of oval shape.[1] They were elongated in a direction perpendicular to the direction of polarization of the incident radiation. Depending on the angle α and the laser beam power P, the elongation amounted to 10–40%.

DISCUSSION OF RESULTS

1. We shall show that the rotation of the plane of polarization is related to bending of the beam rays as a result of its self-focusing. Let us assume that a homotropically oriented crystal is illuminated obliquely ($\alpha \neq 0$) by a linearly polarized (in a horizontal plane YZ) laser beam. We shall assume that the molecules at the walls of a cell are perpendicular to the wall and that their binding to the wall is rigid. An extraordinary wave is excited in such a crystal. The vector of its electric induction $\mathbf{D}$ should be perpendicular to a unit vector $\mathbf{l}_0(\mathbf{r})$ tangential to a ray at a given point and it lies in a plane defined by the vectors $\mathbf{n}(\mathbf{r})$ and $\mathbf{l}_0(\mathbf{r})$ (Figs. 4a and 4b). Since the orientation of the director $\mathbf{n}(\mathbf{r})$ varies along a ray because of reorientation of the investigated crystal in the optical field and the direction $\mathbf{l}_0(\mathbf{r})$ varies because of nonlinear refraction, the direction of $\mathbf{D}$ also varies. It is clear from Fig. 4a that in the case of a ray deviated in a horizontal plane the vector $\mathbf{D}$ changes its direction but it remains in the horizontal plane, i.e., the polarization of the incident light is retained.

A different pattern is obtained for rays lying in other planes. For example, in the case of a ray traveling inside a crystal in a vertical plane E, forming an angle α_0 with $\mathbf{n}$, the vector $\mathbf{D}$ is rotated near the exit wall A' (as demonstrated in Fig. 4b) relative to the initial position and the rotation is by an angle of φ_0 so that the vector is no longer in the horizontal plane (the plane defined by the vectors $\mathbf{n}$ and $\mathbf{l}_0$, in which the vector $\mathbf{D}$ of the extraordinary wave is located, is no longer horizontal).

In the case of an arbitrary ray emerging from a crystal along a direction $\mathbf{l}'$ the position of the unit electric field vec-

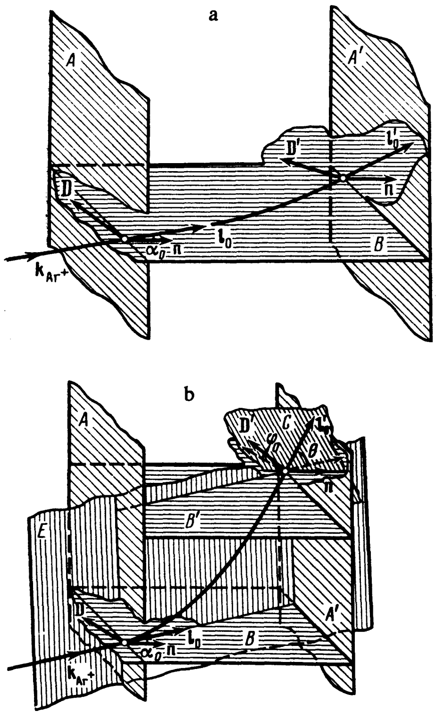

FIG. 4. a) Changes in the polarization of the rays deviated in a horizontal plane B. b) Changes in the polarization of rays deviated in a vertical plane E (rotation of the plane of polarization). Here, AA' are the walls of a cell containing a nematic liquid crystal, BB' are horizontal planes, $\mathbf{n}$ is the director, $\mathbf{l}_0$ is a unit vector tangential to a ray at the entry wall of the cell, $\mathbf{l}_0'$ is the corresponding unit vector tangential to the ray at the exit wall of the cell (inside the crystal), $\mathbf{D}$ and $\mathbf{D}'$ are the electric induction vectors; at the entry wall of the cell the vector $\mathbf{D}$ lies in the plane B for both rays under discussion (which are deviated in the horizontal plane B and in the vertical plane E); at the exit wall the vector $\mathbf{D}'$ also lines in the plane B for the ray deviated in the same plane, but for the ray deviated in the plane E the vector $\mathbf{D}'$ lies in a plane C; θ is the angle of deviation of the ray, α_0 is the angle between $\mathbf{n}$ and $\mathbf{k}$ (inside the crystal), and φ_0 is the angle of rotation of the plane of polarization in the crystal.

tor $\mathbf{e}$ is governed by the vectors $\mathbf{l}$ and $\mathbf{n}$ (naturally, the vectors $\mathbf{D}'$ and $\mathbf{l}_0'$ lie in the same plane). Obviously, the following relationships now apply:

$$(\mathbf{e}\mathbf{l}')=0, \quad \mathbf{e}[\mathbf{l}'\times\mathbf{n}]=0, \quad (\mathbf{e}\mathbf{e})=1,$$

from which it follows that

$$\mathbf{e}=(\mathbf{l}'(\mathbf{l}'\mathbf{n})-\mathbf{n})/|\mathbf{l}'(\mathbf{l}'\mathbf{n})-\mathbf{n}|. \tag{1}$$

Using Eq. (1), we can now estimate the angle of rotation of the plane of polarization for the investigated experimental case, i.e., for rays in the vertical plane XZ (Fig. 1).

In a coordinate system (Fig. 1) with the Z axis parallel to the axis of a beam incident on a crystal, the Y axis is horizontal and the X axis is vertical, and then the vectors $\mathbf{l}'$ and $\mathbf{n}$ have the following coordinates:

$$\mathbf{l}'(\sin\theta,\ 0,\ \cos\theta), \quad \mathbf{n}(0,\ -\sin\alpha,\ \cos\alpha).$$

Equation (1) yields the coordinates of $\mathbf{e}$:

$$\mathbf{e}(R^{-1}\operatorname{ctg}\alpha\sin\theta\cos\theta, \quad 0, \quad -R^{-1}\operatorname{ctg}\alpha\sin^2\theta),$$

$$R=(1+\operatorname{ctg}^2\alpha\sin^2\theta)^{1/2}, \tag{2}$$

and the angle of rotation of the plane of polarization is now different

$$\varphi(\theta)=\operatorname{arctg}[(e_x^2+e_z^2)/e_y^2]^{1/2}=\operatorname{arctg}(\operatorname{ctg}\alpha\sin\theta). \tag{3}$$

The results of a calculation carried out for the above relationship are plotted in Fig. 2 (dashed curve). It is clear from this figure that the agreement between the experiment and calculations is fully satisfactory.

The above analysis of the rotation of the plane of polarization in the nematic phase of a liquid crystal is valid only if normal waves propagate independently.[4] Our estimates obtained for an OCBP liquid crystal $L=150\,\mu$ thick indicate that this is true if $\alpha \gtrsim 20°$. In the range of angles of incidence $\alpha \lesssim 20°$ the propagation of normal waves is no longer independent. In our experimental investigation the angle of incidence was $\alpha=0$ and the polarization of rays emerging from a nematic liquid crystal was elliptic, as predicted above.

It is worth noting that in the $\alpha \gtrsim 20°$ case the position of the plane of polarization of rays emerging from a crystal is governed by the vectors $\mathbf{l}$ and $\mathbf{n}$. A change in the position of either of them should alter the position of the plane of polarization, i.e., a study of rotation of the plane of polarization can be used, in particular, to investigate reorientation of molecules on the walls of a cell if there is a change in the molecular orientation.

2. The diagram in Fig. 3 shows, above all, that reorientation of the director of a nematic liquid crystal (NLC) is an optical field results in self-focusing of a light beam. In fact, in the process of reorientation of the director in an inhomogeneous optical field a crystal can be regarded as a lens with a time-dependent focal length and changes in the intensity at the center of the beam reflect in fact changes in time of the focal length of the nonlinear lenses which are induced optically. An analysis of the results obtained for a nematic liquid crystal in diverging and converging beams (Fig. 3) shows that an induced lens has a positive focal length, i.e., that self-focusing should occur. An analogous result is obtained also for the case of oblique incidence of a light beam on a crystal.

We shall now estimate the maximum increase in the intensity at the center of a beam as a result of self-focusing in a nematic liquid crystal displaced by a distance δ into the diverging part of the beam. We shall assume that the nematic liquid crystal behaves as an aberration-free lens with a varaible focal length. Employing the relationships describing the propagation of a Gaussian beam[5] and bearing in mind that a nonlinear lens alters the radius of curvature of the wavefront of a beam, we obtain a relationship between the maximum intensity $I_{\max}$ at the center of the beam and the initial intensity I_0:

$$I_{max}/I_0=[1+(\lambda\delta/\pi w_0^2)^2]\,[1+(\pi w_0^2/\lambda d)^2]. \tag{4}$$

If $f=280$ mm, the beam radius is $w_0=77\,\mu$ and the calculated maximum increase in the intensity for $\delta=1.5$ cm and $d=200$ cm is 1.17. The experimental value of this increase is 1.11.

We shall now consider oscillations of the intensity at the center of an aberration pattern. Similar oscillations have been observed earlier in the course of thermal self-focusing of a laser beam[6] and they have been attributed to changes in the refractive index of a substance during its heating by a laser beam. In our case there is also a change in the refractive index of an extraordinary wave in the process of reorientation of the director, which gives rise to oscillations of the intensity at the center of an aberration pattern. In fact, at the center of this pattern we can expect interference between axial rays ($\rho = 0$) and rays located far from the center ($\rho = \infty$) for which the nonlinear refraction angle vanishes.[3] In the process of interaction of a light beam with a crystal the molecules become gradually reoriented on the beam axis and there is a continuous change in the phase shift

$$S(\rho=0)-S(\rho=\infty)=k\int_0^L\left(\frac{n_e n_0}{[n_0^2\sin^2\psi+n_e^2\cos^2\psi]^{1/2}}-n_0\right)dz \tag{5}$$

between the interfering rays, which gives rise to observed intensity oscillations. The number N of such oscillations is clearly equal to the factor by which the phase advance on the beam axis exceeds 2π:

$$N=[S(\rho=0)-S(\rho=\infty)]/2\pi. \tag{6}$$

This relationship is identical with the expression for the number of aberration rings.[3] Assuming, in accordance with the boundary conditions, that the angle of rotation of the director on the beam axis varies harmonically

$$\psi(z)=\psi_m\sin(\pi z/L), \tag{7}$$

we find from Eqs. (5) and (6), bearing in mind that $n_e - n_0 < n_0$, the expression

$$N=(L/2\lambda)\psi_m^2(n_e-n_0), \tag{8}$$

where ψ_m is the angle of rotation of the director at the center of a crystal. It follows from Eq. (8) that the number of oscillations is related directly to the angle of rotation of the director and, consequently, the changes in the intensity at the center of an aberration pattern describe the dynamics of reorientation of the director in the field of an optical wave.

3. We shall show that elongation of the aberration rings is due to the fact that the Frank elastic constant K_2 is less than the constants K_1 and K_3 (Refs. 7 and 8). We shall do this by considering the special case when $K_1 = K_3 = K$. Let us assume that a laser beam polarized in a horizontal plane (YZ) propagates along the Z axis and that the distribution of the electric field intensity in the beam has the Gaussian form

$$E=E_0\exp\left\{-\frac{x^2+y^2}{w^2}\right\}. \tag{9}$$

A crystal is oriented homotropically and it is normal to the incident beam ($\mathbf{n}\|\mathbf{k}$). We are assuming that the optical field rotates the director only in the YZ plane and that the angle of rotation is ψ (the angle ψ is measured from the Z axis). Then, the components of the director in this plane are $n_y = \sin\psi$ and $n_z = \cos\psi$.

The elastic energy of a nematic liquid crystal during deformation of the director is a function of the Frank constants. In the YZ plane along the X and Y axes it is proportional to K_2 and K, respectively. Since $K_2 < K$, it follows that the value of ψ and, therefore, the refractive index of an extraordinary wave vary rapidly away from the beam axis along X. This results in the observed elongation of the aberration pattern along the X axis.

If we use the expression for the density of the free energy given in Ref. 9 and average (with respect to time) the square of the electric field intensity, we find that the expression for the density F of the free energy of a nematic liquid crystal subjected to an external electric field can be represented in the form

$$F=\frac{K}{2}(\psi_y^2+\psi_z^2)+\frac{K_2}{2}\psi_x^2-\frac{\Delta\varepsilon E_0^2}{16\pi}\exp\left\{-\frac{2x^2+2y^2}{w_0^2}\right\}\psi^2. \tag{10}$$

In this expression we have restricted ourselves only to the terms quadratic in ψ. We shall seek the function ψ in the form

$$\psi(x,y,z)=A\exp\left\{-\frac{x^2}{a^2}-\frac{y^2}{b^2}\right\}\sin\frac{\pi z}{L}, \tag{11}$$

where A, a, and b are variable positive parameters. We shall find their values by minimizing the free energy $\int F dV$ (the indicated integration is extended over the whole crystal). This gives the relationship

$$2\frac{b}{a}-2\mu\frac{a}{b}+\frac{1}{\xi^2}\frac{abw_0^4(b^2-a^2)}{(a^2+w_0^2)^{3/2}(b^2+w_0^2)^{3/2}}=0, \tag{12}$$

where

$$\mu=K/K_2>1,\quad \xi^{-2}=\Delta\varepsilon E_0^2/8\pi K_2,$$

and ξ is the coherence length. It follows from Eq. (12) that if $\mu > 1$, then $b > a$.

The angle θ_H of deviation of a ray emerging from a crystal and lying in a horizontal plane is given by the following relationship, which is valid in the case of small deviation angles ($\theta_H \ll 1$):

$$\theta_H=k^{-1}\partial S(y)/\partial y, \tag{13}$$

where

$$S=k\int_0^L n_e(y,z)\,dz,\qquad n_e(y,z)=n_0+\Delta n\psi^2(0,y,z),$$

$$\Delta n=n_e-n_0,\quad n_e=\varepsilon_\|^{1/2},\quad n_0=\varepsilon_\perp^{1/2}.$$

Substituting Eq. (11) in Eq. (13), we obtain

$$\theta_H=2A^2\Delta n(Ly/b^2)e^{-2y^2/b^2}. \tag{14}$$

We can easily see that the maximum value of θ_H corresponds to $y = b/2$ and it is

$$\theta_H^{max}=A^2\Delta nL/e^{1/2}b. \tag{15}$$

Similarly, the vertical angular size of an oval is

$$\theta_V^{max}=A^2\Delta nL/e^{1/2}a. \tag{16}$$

It follows from Eqs. (14) and (16) that

$$\theta_V^{max}/\theta_H^{max}=b/a. \tag{17}$$

Since $b > a$, it is clear from Eq. (17) that in the case of the horizontal polarization of a beam incident on a crystal the vertical size of the aberration pattern is greater than the horizontal size. As shown above, this direction of elongation of aberration rings is indeed observed experimentally.

We shall now estimate the degree of elongation of aberration rings given by Eq. (17) when the power is slightly higher than the threshold ($P - P_{th} \ll P_{th}$). We shall do this using Eq. (12) and the relationships for the threshold coherence length and the resultant distribution of the director.[3] Then, on the assumption that the degree of elongation is small (i.e., that $b/a - 1 \ll 1$), we obtain

$$q=\theta_V^{\max}/\theta_H^{\max}-1=(\mu-1)\left(\mu+1+\frac{2}{1+g}\right)^{-1}, \quad g=\frac{\sqrt{2}L}{\pi w_0}. \tag{18}$$

In the case of an MBBA crystal at $t = 25$ °C it follows from Ref. 8 that the constants are $K_1 = 6\times10^{-7}$ dyn, $K_2 = 4\times10^{-7}$ dyn, and $K_3 = 7.5\times10^{-7}$ dyn. Assuming that $K = (K_1 + K_3)/2$ and $g \sim 1$, we can use Eq. (18) to estimate that degree of elongation of the aberration rings: $q \approx 0.21$. The experimental value for a beam of angular divergence 4×10^{-2} amounts to $q \approx 0.12$, i.e., the calculation and experiment are only in qualitative agreement, which is to be expected in the case when a one-constant approximation and the variational method are used.

We shall conclude by drawing once again attention to the special feature of the reorientation of molecules in light beams, namely that it is accompanied by aberration self-focusing, which makes it possible to study not only the process of reorientation of molecules in an optical field, but also the properties of the crystal itself, which are reflected in the aberration pattern.

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