# THEORY OF THE CONVERSION OF OPTICAL RADIATION POLARIZATION IN THE THERMODYNAMICALLY IRREVERSIBLY CRYSTALLIZING MEDIA

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*The physical meaning of a new electro–optical effect of conversion of polarization of optical signal during its propagation through a layer of thermodynamically irreversibly crystallizing liquid is given. Its effect on a linearly polarized signal is analogous to that of the Pockels electro-optical effect being at the same time more pronounced.*

*The transformation of the optical signal in an anisotropically crystallizing substance engendered by the irreversibility of the phase transition provides the basis for this effect, with the longitudinal dispersion of the natural–frequency of vibration of the molecular oscillator in a highly gradient crystallizing electric field playing the most important role.*

*New properties of optical radiation when it interacts with the substance in a thermodynamically nonequilibrium state are considered for water as an example.*

*The effect can be used as a new information characteristic of the strength of hazardous natural phenomena as well as of some transient phase transitions in production processes.*

Previously unknown effect of conversion of laser signal polarization has been discovered in a crystallizing water.[1] An electric field and structural anisotropy of a medium are formed in synchronism in a thermodynamically and electrically perturbed region adjacent to a crystallization front. Double electric layers are formed on both sides of the phase boundary. The perturbed region modulates the polarization of the optical signal propagating through it.

Let us consider the new effect in the approximation which is employed for explaining the Pockels linear electro-optical effect.[2,3] The main difference between these two effects is that the new effect[1] occurs in an originally isotropic medium. Moreover, it takes place in a highly gradient electric field engendered by thermodynamically irreversible crystallization rather than in the external homogeneous electric field.

Let us write, as usual,[2,3] the equation for a molecular oscillator placed in the electric field with intensity $E$ in the form

$$\ddot{r} + 2\gamma_1\dot{r} + \left(\omega_0^2 - \frac{4\pi Ne^2}{3m}\right)r + \gamma_2 r^2 + \frac{e}{m}E = 0\,, \qquad (1)$$

where $r$ is the coordinate of electronic oscillations, $e$ and $m$ are the charge and reduced mass of an electron, $\omega_0$ is its natural–frequency, $N$ is the concentration of oscillating electrons, and $\gamma_1$ and $\gamma_2$ are the constants of the crystallizing medium.

In our case the vector of the intensity $\mathbf{E}(r)$ is perpendicular to the crystallization front,[1,4] therefore the oscillator vibrations parallel to the crystallization front are not affected by the electric field, while the vibrations in the direction of the frontal motion occur under maximum effect of the intensity gradient of the crystallizing electric field. Disregarding the vibration asymmetry in the direction of the frontal motion and in the opposite direction in the crystallizing region it is possible to assume, in accordance with Ref. 4,

$$E = E_0 \exp\left(-\frac{r}{\beta}\right)\,, \quad \varphi_0 = E_0\beta\,, \qquad (2)$$

where $E_0(\tau)$ and $\varphi_0(\tau)$ are the intensity and potential of the electric field at the crystallization front and $\beta(\tau)$ is the parameter of the field gradient in the crystallizing region. These values are determined by the degree of irreversibility of crystallization. In real situations they are the functions of time $\tau$ and have the period of variation being many orders of magnitude larger than the characteristic period of vibrations.

Let us determine the crystallization–induced detuning of the frequency of vibration in the region of maximum $E_0$ and employ it for determining the dispersion of the refraction index of light. The frequency modulated signal is treated as an extraordinary ray which engenders birefringence.

With real values $r < \beta$ the following approximation is valid

$$E = E_0\left(1 - \frac{r}{\beta}\right)\,. \qquad (3)$$

Then we have

$$\ddot{r} + 2\gamma_1\dot{r} + \left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{eE_0}{m\beta}\right)r + \gamma_2 r^2 + \frac{e}{m}E = 0\,, \qquad (4)$$

Due to the crystallization–induced electric field ,the equilibrium position of the oscillator $\dot{r}$, $\ddot{r} = 0$, is displaced at the amount $r = r_0$ approximately equal to

$$r_0 = -\left[1 - \frac{e\,E_0}{m\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}\right)}\right]\times$$

$$\times\left\{\left[1 - \frac{2\gamma_2\, e\,\varphi_0}{\beta m\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}\right)}\right]\right\}\,. \qquad (5)$$

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It should be noted that approximate solution (5) of quadratic equation (4) for $\dot{r}$, $\ddot{r} = 0$ is obtained under physically justified condition that the third term in parentheses of Eqs. (4) and (5) is relatively small but significant. In specific calculations we must estimate the importance of this term and the second term in square brackets on the whole.

Transforming to a new vibrational coordinate $q = r - r_0$ in Eq. (4), we obtain the following relation

$$\ddot{g} + 2\gamma_1\dot{g} + \left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2} + 2\gamma_2 r_0\right) g + \gamma_2 g^2 = 0 \ . \quad (6)$$

The frequency detuning in a crystallizing layer turns out to be equal to

$$\Delta\,\omega_0^2 = -\left(\frac{e\varphi_0}{m\beta^2} - 2\gamma_2 r_0\right) =$$

$$= \left\{\frac{e\varphi_0}{m\beta^2} + \frac{2\gamma_2\, e\ \varphi_0}{\beta m\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}\right)} \times \right.$$

$$\left. \times \left[1 - \frac{2\gamma_2\, e\ \varphi_0}{\beta m\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}\right)}^2\right]\right\} . \quad (7)$$

The basic parameters are the field intensity and its longitudinal gradient in the crystallizing region, both of these parameters are determined by the degree of irreversibility of crystallization.[4]

Accordingly, the refraction index of medium ($n$) varies. Let us use the formula for the dispersion of the index of optical refraction neglecting the absorption in the following form:[2,3]

$$-\frac{\partial n^2}{\partial\omega^2} = \frac{\partial n^2}{\partial\omega_0^2} = -\frac{4\pi Ne^2}{m\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \omega^2\right)^2} , \quad (8)$$

where $\omega$ is the cyclic frequency of a sounding optical signal.

Hence, the difference between the refractive indices of ordinary and extraordinary rays engendered by the irreversibility of crystallization is given by the formula

$$\Delta n = \frac{\partial n^2}{\partial\omega_0^2}\Delta\omega_0^2 = \frac{2\pi Ne^3\,\varphi_0}{nm^2\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \omega^2\right)^2} \times$$

$$\times \left\{\frac{1}{\beta^2} + \frac{2\gamma_2}{\beta\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}\right)} \times \right.$$

$$\left. \times \left[1 - \frac{2\gamma_2\, e\ \varphi_0}{\beta m\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}\right)^2}\right]\right\} . \quad (9)$$

Then the phase shift of the polarization of sounding signal when it propagates through a crystallizing layer with the depth $\beta$ is

$$\delta = 2\pi\Delta n\,\frac{\beta}{\lambda} \simeq \frac{4\pi Ne^3\varphi_0}{\lambda nm^2\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \omega^2\right)^2} \times$$

$$\times \left\{\frac{1}{\beta} + \frac{2\gamma_2}{\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}}\left[1 - \frac{2\gamma_2\, e\ \varphi_0}{\beta m\left(\omega_0^2 - \frac{4nNe^2}{3m} - \frac{e\varphi_0}{m\beta^2}\right)^2}\right]\right\} \simeq$$

$$\simeq \frac{4\pi^2 N\, e^3\ \varphi_0}{\lambda nm^2\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \omega^2\right)^2}\left[\frac{1}{\beta} + \frac{2\gamma_2}{\omega_0^2 - \frac{4\pi Ne^2}{3m} - \frac{e\varphi_0}{m\beta^2}}\right] , \quad (10)$$

where $\lambda$ is the wavelength of the optical signal in the crystallizing medium.

Thus at the start of the process of irreversibility, while

$$\omega_0^2 \gg \gamma_2\,\beta \ , \quad (11)$$

the phase shift is proportional to the intensity of the crystallized electric field. It is equivalent to possible neglect of the term $\gamma_2 r^2$ in Eq. (1), with the vibrations being harmonic.

As the irreversibility of crystallization intensifies the contribution of this term increases and the vibrations become substantially anharmonic. In the limiting case of large values of $\beta$ we have

$$\omega_0^2 \ll 2\gamma_2\,\beta \ , \quad (12)$$

and the phase shift can be set to be proportional to the crystallization potential $\varphi_0$

$$\delta \simeq \frac{8\pi^2 N\, e^3\,\gamma_2\,\varphi_0}{\lambda nm^2\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \omega^2\right)^2\omega_0^2} . \quad (13)$$

Comparing the experiments with formulas (10) and (13) we see that the main problem is insufficient definiteness of the parameters of the crystallizing medium first of all, $\omega_0^2$ and $\gamma_2$. According to Refs. 3 and 6, let the relation between $\omega_0^2$ and electron polarizability ($A$) be

$$\omega_0^2 = \frac{e^2}{mA} . \quad (14)$$

Then with real values of polarizability of crystallizing water the term in parentheses of denominator (8) and, correspondingly, in subsequent equations involves a small difference between the large values whose insufficient definiteness could change the order of magnitude of the calculated result $\delta$.

Based on the results of the experiments of Ref. 1 with equilibrium distilled water, formula (13) can be represented in the form

$$\dot{\delta} \simeq 2\alpha\dot{\varphi}_0 \ , \quad (15)$$

where

$$\alpha = \frac{4\pi^2 N\, e^3\,\gamma_2}{\lambda nm^2\left(\omega_0^2 - \frac{4\pi Ne^2}{3m} - \omega^2\right)^2\omega_0^2} = 0.20\ \text{rad}\cdot\text{V}^{-1} . \quad (16)$$

With such a value of α and the maximum value of the potential $\partial_0 = (50 - 150)$ V the polarization vector is capable of making several revolutions during crystallization while the signal amplitude still remains sufficiently large.

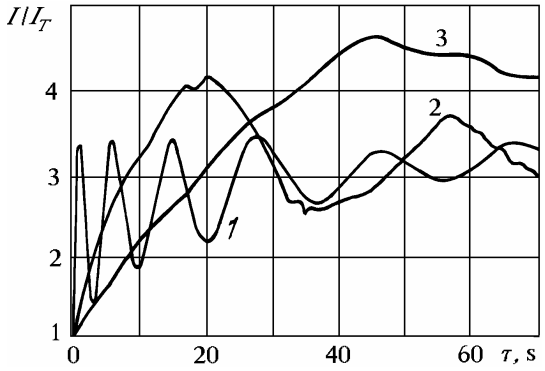


*FIG. 1. Rotation of the polarization vector of laser signal in terms of the ratio of photoelectric current to a dark current $I/I_d$ for different maximum crystallization potentials at the front* ($\varphi_0$): *1)* 140; *2)* 28; *3)* 12 V.

In the standard cases the potential $\varphi_0$ increases monotonically in time when $\dot{\varphi}_0$ decreases. Consequently, the polarization vector revolution occurs with the decreasing velocity of rotation. Using the Nicol prisms we obtain an output photoelectric signal in the form of a periodic curve with an increasing period and decreasing amplitude. The latter monotonically decreases due to the signal depolarization and absorption in the ice layer with increasing thickness. This pattern was found in Ref. 1. The present paper is concentrated on maximum attainable irreversibility of crystallization, the foregoing experimental value of α was obtained when condition (12) was satisfied. Later the experiments on the conversion of polarization were repeated in Ref. 5 with different degree of irreversibility, the combined result is shown in Fig. 1. It confirms the fact that the velocity of rotation of the polarization vector is proportional to the degree of irreversibility which is characterized by the maximum crystallization potential $\varphi_0$.

To involve the theory we must introduce the aforementioned asymmetry of oscillations in the direction of the field gradient and opposite to it. Moreover, in calculating the phase shift of vibrations we must go over from multiplication by β to integration over the entire crystallizing region taking into account different dependence of $\Delta n \left[\mathbf{E}(r), \frac{\partial \mathbf{E}}{\partial r}\right]$ on both sides of the phase boundary.

With increase of the wavelength of a sounding signal while going over from the optical wave range ($\lambda \lesssim \beta$) to the radio wave range ($\lambda \gg \beta$) the effect under study must be less pronounced when radiation propagates through the crystallization front. However for the pulsed sounding of crystallizing clouds and fogs new factors come into force which act in favour of the radio wave region (increase in a strobing volume and others).

## REFERENCES

1. L.G. Kachurin and B.Kh. Petkov, Dokl. Akad. Nauk SSSR **318**, No. 3, 586-589 (1991).
2. D.V. Sivukhin *Optics* ,(Nauka, Moscow, 1980) 751 pp.
3. A. Lösche, *Molecular Physics* (Mir, Moscow, 1987), 228 pp.
4. L.G. Kachurin, in: *Surface Phenomena in Liquids* (Leningrad State University Publishing House, Leningrad, 1975), pp. 137-153.
5. V.A. Malikov, *Conversion of Polarization Characteristics of Laser Radiation Propagating Through a Thermodynamically Irreversibly Crystallizing Water Layer*, (Diploma Project, Leningrad State Hydrometeorological Institute, Leningrad, 1991) 31 pp.
6. M.F. Vuks, *Light Scattering in Gases, Liquids and Solutions* (Leningrad State University Publishing House, Leningrad, 1970) 320 pp.