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Polarization dynamics of an ordinary light wave interacting with a nematic liquid crystal

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Experimental observations and theoretical calculations of the interaction of an ordinary light wave with a homeotropically aligned nematic liquid crystal (NLC) are presented. It was observed that by increasing the light power, aperiodic, damped, periodic and stochastic oscillations of the poiarization of the light wave take place. Theoretically, the self-consistent problem of the interaction of the director of a homeotropic NLC with a plane light wave of ordinary type was treated and the change in the polarization state of the transmitted light wave was calculated. Calculations reproduced the existence of aperiodic, damped and periodic oscillations, in qualitative agreement with the experimental results.

1. Introduction

The light induced Fréedericksz transition (LIFT) in nematic liquid crystals (NLC) has some specific features which are absent in non-optical fields. (For details of these features see for example [1–4].) Among the effects characteristic only for light fields, the self-oscillations of the director of a homeotropically aligned NLC in the field of an ordinary light wave (o-wave) are of special interest.

Self-oscillations of the director of a NLC in the field of an o-wave were first reported (to our knowledge) by the authors [5]. Director self-oscillations appeared as oscillations in the aberrational self-focusing picture, i.e. in the temporal changes of the number of aberrational fringes and correspondingly in the temporal changes of the divergence of the transmitted light beam. It was shown later [6] that the character of the oscillations depends on both the power P and the angle of incidence α of the light beam and, in the frame of a simple mathematical model describing the interaction of the NLC director with the light wave, that a limiting self-oscillation cycle exists. Further detailed investigations have shown [7] that, at a given angle α , with increasing light power, aperiodic, damped, and stochastic oscillations can be excited. A closed area in

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the (P, α) plane was experimentally determined, where pure periodic oscillations of the aberrational picture and consequently that of the director field could be observed.

The Fréedericksz transition due to an o-wave has been studied also by other authors [8,9]. Zel'dovich *et al.* [8], observed director self oscillations and measured how the LIFT threshold depends on the angle of incidence of the light beam. They have shown that the observed angle dependence can be described fairly well by a theory based on the perturbation method. Aleverdian *et al.* [9], found that with increasing light power the period of self-oscillations increases too. This latter result contradicts earlier observations [6, 7], though it has to be mentioned that in [9] a much broader light beam was used than in [6, 7].

It should be noted too that oscillations of the director field can also be excited at normal incidence of the light wave if the light polarization differs from linear. Different types of oscillations and also director precession in elliptically and circularly polarized fields have been observed and investigated by different authors [9–13].

A principal feature of the interaction of the NLC and the light field is the reverse effect of the NLC with modified director field on the modifying light wave itself. Significant changes of the electric field of the light wave take place.

The aim of the present paper is to report experimental and theoretical investigations of the dynamics of the field of an ordinary light wave crossing a NLC and exciting in it director self-oscillations.

2. Experimental study of the polarization dynamics

The measuring method of the polarization state of the light wave transmitted by the NLC has been described elsewhere in detail [14]. It makes use of the fact that the parameters of the polarization ellipse can be correlated—at every moment of time—with the projections of the electric field vector **E** in three (non-parallel) directions of the x-z plane, orthogonal to the light wave vector **k** (see figure 1). (Due to the light field, the homeotropic NLC turns to an inhomogeneous, anisotropic medium resulting, generally, in elliptical polarization of the transmitted wave.) Practically, the light intensities have to be measured for three different positions of the analyser: along the X axis, $I_x = E_x^2$; along the Z axis, $I_z = E_z^2$ and in a third direction CC', forming an angle ϕ with the X axis $(0 < \phi \pi/z)$, $I_{\phi} = E_{\phi}^2$. The polarization ellipse parameters:



Figure 1. Polarization ellipse of the transmitted light wave.

inclination angle of the major axis ζ ; light intensity along the major axis I_A , and the degree of ellipticity $e = I_B/I_A$ (I_B being the light intensity along the minor axis) can be expressed in terms of I_x , I_z and I_{ϕ} using the following simple relations:

$$\operatorname{tg} 2\zeta = \frac{2(I_{\phi} - I_x \cos^2 \phi - I_z \sin^2 \phi)}{(I_z - I_x) \sin 2\phi},\tag{1}$$

$$I_{\rm A} = \frac{1}{2} \left(I_x + I_z + \frac{I_z - I_x}{\cos 2\zeta} \right), \tag{2}$$

$$e = \frac{(I_x + I_z)\cos 2\zeta - I_z + I_x}{(I_x + I_z)\cos 2\zeta + I_z - I_x}.$$
(3)

The experimental set-up is shown schematically in figure 2. The NLC cell consisted of a homeotropically aligned OCB (4-octyl-4'-cyanobiphenyl) film of 150 μ m thickness sandwiched between two glass plates in a thermally controlled holder. During the measurements, the cell temperature was kept at 37°C. The values I_x , I_z and I_{ϕ} were recorded successively, i.e., after turning the analyser into the corresponding positions. The turning time was always kept less than 0.5 s, while the recording time in each position was 1-5 s, still significantly shorter than the period T of the observed oscillations (T = 40-90 s). As an example, the result of a set of such successive measurements is shown in figure 3. From these measurements, the temporal dependence of the three intensity components I_x , I_z and I_{ϕ} could be easily



Figure 2. Experimental set up: L focusing lens, P polarizer-analyser, D diaphragm, PD photodiode, R. recorder. The electric field E of the incident wave is perpendicular to the plane of k and n.



Figure 3. An example for the experimental determination of the temporal changes of the light intensities I_x , I_z and I_{Φ} , transmitted at the corresponding positions of the analyser.



Figure 4. Dynamics of the electric field E, observed on the beam axis, at different power levels.

reconstructed and, using the relations (1)-(3), I_A , ζ and e could be determined. The time dependence of these values, especially the trajectory of the motion of the radius vector I_A gives information on the dynamics of the electric field of the light wave passing through the NLC.

Two sets of experiments were carried out. In the first, the polarization state of the central part of the light beam was studied. In this case, the polarization was found to be nearly linear (e < 0.2). The polarization dynamics are illustrated in figure 4. The magnitude of **E**, which is proportional to $I^{1/2}$, changes in the centre of the beam due to self-focusing [15]. Below a threshold value $P_{\rm th} = 75 \,\mathrm{mW}$, no change in the light polarization takes place (see figure 4(*a*)), indicating the absence of any reorientation of the director. Above the threshold ($P > P_{\rm th}$), the change in the polarization showed that the director was reoriented.

In the interaction of the above-threshold field and the director three interaction modes can be distinguished:

- (i) Aperiodic deformation and damped oscillation mode $(P_{th} < P < P_{osc})$. Here, first the electric field vector of the light wave tilts from the initial vertical direction monotonously and reaches a new stationary position characterized by the angle ζ_{st} (see figure 4(b)). At higher powers, the stationary state is reached through damped oscillations (see figure 4(c)). By further increase of the power, the amplitude of the oscillations increases, while the damping decreases and the angle ζ_{st} increases until it reaches a maximum value $\zeta_{st_{max}}$.
- (ii) Regular oscillation mode ($P_{osc} < P < P_{stoch}$). Above the new threshold power P_{osc} , periodic self-oscillations are excited which can be characterized by periodic changes of the angle ζ and the field amplitude *E*. The end-point of the vector **E** moves along loops lying close to each other (see figures 4 (*d*) and 4 (*e*)).
- (iii) Stochastic oscillation mode ($P_{\text{stoch}} < P$). In this mode, which occurs when P is higher than a third threshold power value, P_{stoch} , the magnitude and the direction of the vector E changes randomly, within some limits, and in the trajectory of E many bends and crossings can be observed.

To sum up, in figure 4 the different types of temporal variations of the polarization of the transmitted light are shown: aperiodic, damped, periodic and stochastic oscillations, respectively.

In the second series of experiments, polarization parameters integrated over the whole light beam were determined. The aperture was replaced here by a lens collecting



Figure 5. Dynamics of the integrated light field parameters \overline{I}_A and ζ of the transmitted wave at different power levels ($\alpha = 6^\circ$).

all the light onto the detector. Evidently, in this case $I_x + I_z = \text{constant}$ and therefore it was sufficient to measure I_z and I_{ϕ} only.

Measurements were carried out partly at a fixed angle of incidence $(\alpha = 6^{\circ})$ by changing the light power, and partly at a fixed light power by changing α ($2^{\circ} \le \alpha \le 8^{\circ}$). Results are given in figure 5 and figure 6.

As shown in figures 5 (a) and (b), at increasing light powers, first periodic changes in the inclination angle ζ of the polarization ellipse take place while the length of I_A remains practically constant. At larger powers the trajectory of I_A has an oval-like shape (see figure 5 (c)) and the degree of ellipticity $e = (I - I_A/I_A)$ increases. By further increase of the power, a competition of the different types of regular self-oscillations can be observed. In figure 5 (d), the trajectories of two competing self-oscillation modes are shown. Transitions from one type to the other occur randomly. At even larger powers, the temporal changes of the polarization ellipse become fully stochastic. The radius vector I_A follows non-reproducible trajectories, with rather complex transient changes. In figure 5 (e), a peculiar doubling of the oscillation period is shown.

As figure 6 shows, polarization parameters of the light wave transmitted by the NLC strongly depend on the angle of incidence α . The smaller the angle α , the larger is the amplitude of the self-oscillation and also the complexity of the trajectory of I_A . With decreasing α , the period of self-oscillation increases from several tens of seconds to



Figure 6. Dynamics of the integrated light field parameters \overline{I}_A and ζ of the transmitted wave at different angles of incidence (P = constant).

several minutes. In the region $2^{\circ} \leq \alpha \leq 6^{\circ}$, relatively small changes in the light power cause significant changes in the character of the self-oscillations. The degree of ellipticity is here nearly unity ($e \approx 1$), i.e. the polarization is nearly circular. At $\alpha > 6^{\circ}$ the polarization of the transmitted light remains nearly linear.

3. The theory of director and ordinary wave interaction

In most of the published theoretical work dealing with the problem of the interaction of an o-wave with a homeotropically aligned NLC, attention has been paid mainly to the calculation of the threshold of the Fréedericksz transition, induced either by an infinite plane wave [8, 16–18] or by a laser beam of finite size [19, 20]. The above-threshold behaviour of the director field of the NLC and that of the inducing light wave proved to be a rather serious and complex problem, and it has been studied only to a smaller extent. The theoretical description of director reorientation at normal incidence of an o-wave on a NLC with oblique director orientation was treated by Zolot'ko *et al.* [21]. They succeeded in describing periodic director oscillations in that geometry.

In the present work, improving the approach outlined in [21], we treat periodic oscillations of the director and the polarization vector of a plane light wave in the case in which the NLC is homeotropically aligned and the light wave of o-type has an oblique incidence.

The electric field E of a harmonic wave can be described by the Helmholz equation

$$\Delta \mathbf{E}(\mathbf{r},t) - \nabla [\nabla \mathbf{E}(\mathbf{r},t)] + \frac{\omega^2}{c^2} \hat{c}(\mathbf{r},t) \mathbf{E}(\mathbf{r},t) = 0, \qquad (4)$$

in which the dielectric permittivity tensor $\hat{\varepsilon}$ is connected with the NLC director **n** by the relation

$$\varepsilon_{ij} = \varepsilon_{\perp} \delta_{ij} + \Delta \varepsilon n_i n_j. \tag{5}$$

Here $\Delta \varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$; ε_{\parallel} and ε_{\perp} are the dielectric permittivities for the electric field of the light wave parallel or normal to the director, respectively; δ_{ij} is the Kronecker symbol; n_i is the projection of **n** on the coordinate axes; $|\mathbf{n}| = 1$, $(n_i n_i = 1)$.

The reorientation of the director **n** due to the light wave can be described, in the oneconstant approximation, by the following equation of motion [22]

$$\frac{\gamma_1}{K}\frac{\partial n_i}{\partial t} + \Delta n_i + \frac{\Delta\varepsilon}{4\pi K}E_i n_j E_j = \lambda(\mathbf{r})n_i.$$
(6)

Here K is the Frank elastic constant, $\gamma_1 = \mu_3 - \mu_2$; μ_2 and μ_3 are Leslie viscosity coefficients, and Δ is the Laplace operator. The $\lambda(\mathbf{r})$ function can be determined taking into account that $|\mathbf{n}| = 1$.

In a homeotropic NLC, the unperturbed director \mathbf{n}_{o} is directed normal to the surface of the crystal (see figure 7). The cartesian coordinate system is chosen in such a way, that the axis Y' is parallel to \mathbf{n}_{o} , while the axes X' and Z' lie in the plane of the surface. The orientation of the perturbed director \mathbf{n} can be described by the polar and azimuthal angles ψ and φ

$$n_{x'} = \sin\psi\cos\varphi, \quad n_{y'} = \cos\psi, \quad n_{z'} = \sin\psi\sin\varphi.$$
(7)



Figure 7. Interaction geometry for a homeotropic NLC with an o-wave of oblique incidence.

The electric field of the plane light wave, incident on the NLC film at an angle α with respect to the Y' axis, can be described by

$$\mathbf{E}(\mathbf{r},t) = (1/2)\mathbf{A}(y',t)\exp(i\omega x \sin \alpha/c + iky' - i\omega t) + c.c$$
(8)

where $k = \omega/C$ $(\varepsilon_{\perp} - \sin^2 \alpha)^{1/2}$, and the amplitude A is a slowly varying quantity.

The description of the light field by (8) means that the spatial limitation of the wave is not taken into account.

At the front boundary of the NLC (at y'=0) the vector A is oriented ordinarily

$$A_{x'} = 0, \quad A_{y'} = 0, \quad A_{z'} = B,$$
 (9)

and the director \mathbf{n} at both the front and the rear boundary is oriented normal to them

$$\psi(0) = \psi(L) = 0. \tag{10}$$

It is more convenient to describe the field polarization in terms of the Stokes parameters [23], which can be given by

$$S_{1} = (A_{e}A_{o}^{*} + A_{e}^{*}A_{o})/|B|^{2},$$

$$S_{2} = i(A_{e}^{*}A_{o} - A_{e}A_{o}^{*})/|B|^{2},$$

$$S_{3} = (|A_{e}|^{2} - |A_{o}|^{2})/|B|^{2}.$$
(11)

Here

$$A_{e} = A_{x}, \cos \varphi + A_{z}, \sin \varphi, A_{o} = -A_{x}, \sin \varphi + A_{z}, \cos \varphi.$$
(12)

In the case of normal incidence ($\alpha = 0$), the quantity A_e gives the projection of A on the plane of the wave vector and the director (the latter determining the direction of the local optical axis). Hence, for normal incidence, A_e can be treated as the amplitude of the extraordinary wave (and in an analogous manner, the quantity A_o as that of the ordinary wave).

Substituting (8) and (12) into (4), and eliminating the field component $A_{y'}$, with the aid of (11), the following set of equations can be achieved for the Stokes parameters:

$$\frac{\partial S_1}{\partial \eta} = -v_3 S_2 + v_2 S_3,$$

$$\frac{\partial S_2}{\partial \eta} = v_3 S_1 - v_1 S_3,$$

$$\frac{\partial S_3}{\partial \eta} = v_1 S_2 - v_2 S_1.$$
(13)

Here $\eta = \pi y'/L$ is a dimensionless coordinate, $v_1 = a \sin 2\varphi - b \cos 2\varphi$, $v_2 = -2(\partial \varphi/\partial \eta)$, $v_3 = -a \cos 2\varphi - b \sin 2\varphi$, $a = 2N(u^2 - n_z^2)$, $b = 4Nun_z$, $N = \omega L\Delta n/2\pi c$, L is the crystal thickness, $\Delta n = \varepsilon_{\perp}^{1/2} \Delta \varepsilon/2\varepsilon_{\parallel}$, and $u = n_x - \sin \alpha$. In the particular case of $\alpha = 0^\circ$, equations (13) are equivalent to those derived by Santamato *et al.* [24].

In the following, some approximations are applied. If only the low order space harmonics of the director field deformation are used [13]:

$$\left. \begin{array}{c} \psi(\eta,t) = \psi_0(\tau) \sin \eta, \\ \phi(\eta,t) = \phi_0(\tau) + \phi_1(\tau) \cos \eta, \end{array} \right\}$$

$$(14)$$

where $\tau = t/\tau_0$, $\tau_0 = (\gamma_1 L^2/\pi^2 K)$, and the coefficients in (13) are substituted by their mean values averaged over the spatial coordinate η , then (13) can be solved easily

$$S_{1} = S_{1}^{0} \cos \Gamma \eta + \bar{V}_{1} d(1 - \cos \Gamma \eta) + \bar{V}_{2} S_{3}^{0} \Gamma^{-1} \sin \Gamma \eta,$$

$$S_{2} = \bar{V}_{2} d(1 - \cos \Gamma \eta) + (\bar{V}_{3} S_{1}^{0} - \bar{V}_{1} S_{3}^{0}) \Gamma^{-1} \sin \Gamma \eta,$$

$$S_{3} = S_{3}^{0} \cos \Gamma \eta + \bar{V}_{3} d(1 - \cos \Gamma \eta) - \bar{V}_{2} S_{1}^{0} \Gamma^{-1} \sin \Gamma \eta.$$
(15)

Here $\Gamma = (\overline{V}_1^2 + \overline{V}_2^2 + \overline{V}_3^2), \quad \overline{V}_1 = N(-8\alpha\psi_0\sin\varphi_0/\pi + 2\alpha^2\sin 2\varphi_0), \quad \overline{V}_2 = 4\varphi_1/\pi, \\ \overline{V}_3 = N(-\psi_0^2 + 8\alpha\psi_0\cos\varphi_0/\pi - 2\alpha^2\cos 2\varphi_0), \quad d = (V_1S_1^0 + V_3S_3^0)/\Gamma^2, \quad S_1^0 = \sin 2(\varphi_0 + \varphi_1), \\ S_3^0 = -\cos 2(\varphi_0 + \varphi_1).$

Substituting (14) and (15) into (6), eliminating λ , and expressing the amplitudes of the spatial harmonics of the director field deformation, the following set of ordinary differential equations can be achieved:

$$\frac{\partial \varphi_{0}}{\partial \tau} = (\delta/2)(W_{120} + \alpha h_{1}/\psi_{0}),
\frac{\partial \varphi_{1}}{\partial \tau} = -3\varphi_{1} + 2\delta(W_{121} + \alpha h_{2}/\psi_{0}),
\frac{\partial \psi_{0}}{\partial \tau} = -\psi_{0} - \frac{3\varphi_{1}^{2}\psi_{0}}{4} + \frac{\delta}{2} \left[\psi_{0}(1 + W_{320}) - \frac{\psi_{0}^{2}}{2N^{1/2}} \left(1 + \frac{4W_{340}}{3} \right) - \alpha h_{3} \right].$$
(16)

Here $h_1 = g_1 \sin \varphi_0 + g_2 \cos \varphi_0$, $h_2 = g_3 \sin \varphi_0 + g_4 \cos \varphi_0$, $h_3 = g_2 \sin \varphi_0 + g_5 \cos \varphi_0$, $g_1 = 4/\pi - W_{310} + \varphi_1 W_{111}$, $g_2 = -W_{110} - \varphi_1 W_{311}$, $g_3 = -W_{311} + \varphi_1 W_{112}$, $g_4 = 4\varphi_1/3\pi - W_{111} - \varphi_1 W_{312}$ and $g_5 = 4/\pi + W_{310} - \varphi_1 W_{111}$,

$$W_{nmk} = \frac{2}{\pi} \int_0^{\pi} S_n(\eta) \sin^m \eta \cos^k \eta \, \mathrm{d}\eta,$$

and δ is a dimensionless quantity, proportional to the light power density (dimensionless power density)

$$\delta = \frac{\Delta \varepsilon}{16\pi K} \frac{L^2}{\pi^2} \frac{\varepsilon_\perp}{\varepsilon_\parallel} I$$

where $I = |A_e|^2 + |A_o|^2$.

The set of equations (16) was solved numerically and the polarization parameters were determined from (15). The magnitude and angle of inclination of the vector I_A can be given by the Stokes parameters as

$$I_{\rm A} = [1 + (1 - S_2^2)^{1/2}]/2, \tag{17}$$

$$\zeta = \varphi + \frac{1}{2} \operatorname{arctg} \left(S_1 / S_3 \right). \tag{18}$$

(Equations (17) and (18) are strictly valid only for normal incidence of the light wave, i.e. if the coordinate systems (X, Y, Z) and (X', Y', Z') coincide. For small angles, however, the use of these equations is still possible.)

Results of calculation for the power dependence of the light polarization are illustrated in figure 8, and that for the stationary angle of inclination ζ_{st} in figure 9 (for $\alpha = 6^{\circ}$). It can be seen from figure 8, that if δ is less than a threshold value, $\delta < \delta_{th} = 1.22$, the director field remains unaffected and the polarization of the transmitted light does not change. In the region $1.22 < \delta < 2.1$, the director field becomes unstable and the polarization of the transmitted wave changes: the major axis of the polarization ellipse is deflected from the vertical direction.

After some time, the angle of deflection reaches a stationary value ζ_{st} (it may occur after a few oscillations). As shown in figure 9, the value of ζ_{st} increases monotonically with increasing power, approaching a maximum value, $\zeta_{st_{max}}^{theor} = 37^{\circ}$, at $\delta = 1.7$; i.e. at $\delta/\delta_{th} = 1.4$. This theoretical value is larger than the experimental $\zeta_{st_{max}}^{exp} = 25^{\circ}$. It should be mentioned, however, that the experimental value was achieved at the same relative power level ($P/P_{th} = 1.4$), as given by the theory.

At $\delta > \delta_{osc} = 2.1$, undamped periodic oscillations of the director and the light polarization take place, the period at $\delta = 2.5$ being $T \approx 20 \tau_0$. Using the data of Kujumchyan *et al.* [25], we obtained $\tau_0 = 17$ s and $T \approx 330$ s. This theoretical value is, however, significantly larger, than the experimental $T_{exp} \approx 60$ s.

For $\delta > 2.3$, the calculation gives periodic solutions for which the trajectories of the vector I_A are symmetric with respect to the vertical direction. This however contradicts



Figure 8. Calculated trajectories of the motion of I_A at different values of the dimensionless light power density δ ($\alpha = 6^\circ$).



Figure 9. Calculated dependence of the stationary inclination angle ζ_{st}^{theor} on the light power density δ ($\alpha = 6^{\circ}$).

the observations. Moreover, in experiments in this range, oscillations become stochastic. Apparently this behaviour is not reflected by the present theory.

Summing up the theoretical considerations, our model for the description of the behaviour of the director of a homeotropic NLC in the field of an ordinary light wave at oblique incidence, though it is strongly approximate, reproduces qualitatively the observed damped and undamped periodic oscillations of the light wave polarization vector. For a more accurate description of the phenomena, however, especially of the observed stochastic processes, a more complete theoretical model is needed.

The o-wave-NLC interaction at oblique incidence, treated as above can be compared with the case of normal incidence of an elliptically polarized light wave, investigated by Santamato *et al.* [12].

It is characteristic for both cases that for director reorientation, a threshold intensity exists which is connected with the orthogonality of the electric field and the unperturbed director, and that different oscillation modes can be excited, due to the strong reverse effect of the NLC on the polarization state of the light wave propagating it.

There are, however, significant differences. At oblique incidence of an o-wave, the e-wave develops only inside the liquid crystal, while at normal incidence of an elliptically polarized wave, the intensity of the e-wave differs from zero even at the front boundary of the liquid crystal. (In fact, at normal incidence, at the boundary of the homeotropic NLC, the polarization planes of the e- and o-waves are undetermined. In the deformed NLC, however, they are determined at any distance from the boundary.) At normal incidence, in the special case of circular polarization, the problem has an axial symmetry, while at oblique incidence of an o-wave, there is always a preferred plane determined by the wave vector and the unperturbed director. At oblique incidence, due to the non-collinearity of the wave vector and the unperturbed director, the phase difference between the ordinary and extraordinary waves propagating inside the liquid crystal can be large, even at small director reorientation angles, unlike the case for normal incidence of an elliptically polarized wave.

These differences are reflected in the character of the possible oscillation modes in the two cases. At normal incidence of an elliptically polarized light wave, precession of the director can be excited, while this cannot occur at oblique incidence of an o-wave. On the other hand, at large angles of incidence of an o-wave, an adiabatic light propagation mode builds up [26] resulting in a reorientation threshold many times higher than that at small angles. This is absent in the case of normal incidence.

All these investigations on the interaction of light waves with a NLC have revealed a large variety of effects which are worthy of further study.

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