Optical bistability, multistability, and instabilities in liquid crystals

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The physical aspects of the manifestation of optical bistability, instabilities, and stochasticity in strongly nonlinear media in the form of liquid crystals subjected to laser radiation fields are discussed. The stress is on experimental research. Optical bistability and multistability are considered for a number of light-induced effects in homogeneous and inhomogeneous liquid crystals. The special features of these effects in liquids crystals, associated with the field-induced structural phase transitions, are stressed and the concept of intrinsic (mirrorless) optical bistability is introduced. General nonlinear systems with optical bistability are considered. An analysis is made of instabilities and stochasticity manifested in experiments on light-induced reorientation of liquid crystals in the presence of fields of different origin, giving rise to oscillatory and chaotic processes in the case of self-modulation of the transmitted light.

CONTENTS

I. INTRODUCTION

1.1 The phenomenon of optical bistability or multistability is a common property of nonlinear optical systems with feedback and it means that there are two or more stable states of a system corresponding to different amplitudes or polarizations of the optical field (see, for example, Refs. 1–3, 5 and 11).

Although the first demonstrations of optical bistability were reported over twenty years ago for active amplifying systems,[13,102] work on this topic became significant only in the eighties.

The main concepts have now been formulated and a classification of systems with optical bistability is available (see, for example, Ref. 1). They are usually divided into hybrid systems and those with intrinsic optical bistability. In the former case, the system is subjected not only to an optical field, but also to a static (electric) field; the dependences of the parameters of a system on the intensity or polarization of light are manifested because of an electrical signal (provided by a detector forming a part of a feedback loop) interacting with a nonlinear medium and controlling the intensity or polarization of an optical wave. In the latter case a system is completely optical and the dependences of the parameters of a nonlinear medium on the intensity or polarization of light are due to the direct interaction of light with matter. Sometimes intrinsic bistability is considered in a narrower sense: only when a feedback in the system is active in the absence of mirrors (mirrorless optical bistability—see Ref. 57).[1)]

Optical bistability may be manifested by active amplifying systems as well as by passive (unexcited) systems. The latter are more interesting from the point of view of applications (they are simple, compact, and require little power).

Bistability may result from nonlinear absorption, known as absorption optical bistability, when the imaginary part of the nonlinear susceptibility is involved, or it may be due to nonlinear dispersion, which is known as dispersion optical bistability, when a laser field alters the refractive index of a medium and the real part of the nonlinear susceptibility is involved. In some cases both optical bistability mechanisms act simultaneously.

Typical characteristics of systems exhibiting optical bistability are illustrated in Fig. 1 (Ref. 2). An increase in the intensity I_{in} of optical radiation incident on a system results in a continuous increase of the optical intensity of light I_{out} emerging from the system until I_{in} reaches a critical value I'_{in} when I_{out} changes abruptly, i.e., a system switches to a different state (Fig. 1a). When I_{in} is reduced, the value of I_{out} follows the upper branch of the curve corresponding to this second state of the system down to $I_{in} = I''_{in}$, where $I''_{in} < I'_{in}$, when there is a reverse abrupt change in I_{out} to the lower branch of the curve, i.e., the system returns to the initial (first) state. Between the two critical points I'_{in} and I''_{in} the system can be in two stable states (there are two values of I_{out}) for a given value of I_{in}, i.e.,

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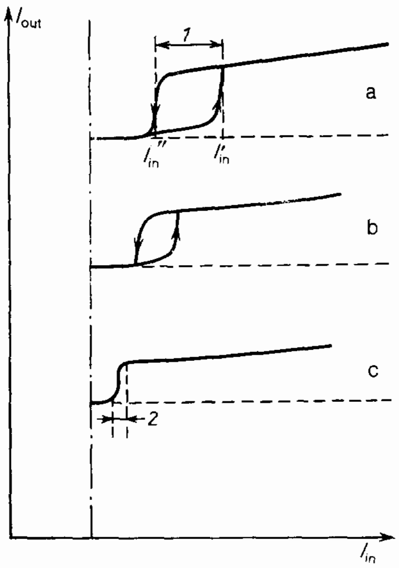

FIG. 1. Typical characteristics of an optically bistable system.[2] Effective reduction of a macroscopic feedback can reduce the width of the bistability region until it disappears (as shown consecutively in Figs. 1a–1c). Optical bistability (1) and differential amplification (2) regions are identified by arrows in Figs. 1a and 1c, respectively.

hysteresis is observed; on both sides of the bistable region a change in the system is described as switching from one state to another. Therefore, in systems exhibiting optical bistability the state of a system depends on the direction of variation of the controlling parameter (I_{in} in the present case), i.e., these systems exhibit a memory effect. This is due to the multivalued nature of the regimes which can be established in a nonlinear optical system. The actual form of a hysteresis loop[2)] is governed by feedback established in the system (Fig. 1b); hysteresis may even disappear (Fig. 1c) and the dependence of I_{out} on I_{in} then becomes single-valued. When the slope of this curve is sufficiently steep, a small change in I_{in} results in major changes in I_{out}, i.e., differential amplification takes place (this happens for $\delta I_{out}/\delta I_{in} > 1$ in the switching region in Fig. 1c). Therefore, in the presence of two incident beams (one weak I_w and the other strong I_s) it is possible to alter considerably the transmitted intensity (I_s) by a small change in I_w, as is usually done in electronic transistors. Such optical devices are called transphasors. On the other hand, if $\delta I_{out}/\delta I_{in} \ll 1$ a system of this kind acts as a power limiter. Such operational capabilities can also be utilized in the construction of logic elements (OR, AND, etc.) so that in principle the complete range of Boolean algebra operations can be performed.[1]

Optical bistability based on interference between two or several light beams requires use of coherent optical fields, so that laser radiation must be used. No-interference systems can operate, in principle, also when wide-band light sources are used, but in the case of purely optical systems it is necessary to employ laser sources in order to provide radiation power sufficient for the experimental observation of optical bistability. Moreover, in the case of stimulated processes it is necessary to overcome the excitation threshold so that once again laser fields are essential.[16]

In some cases (laser systems with amplification; stimulated processes with an excitation threshold) a hysteresis curve differs from that shown in Fig. 1: switching to the upper branch and back again begins from zero value of the controlled parameter I_{out} (Ref. 70).[3)] However, this distinction is not of fundamental importance: for example, in the case of lasers it is determined by the actual nature of the field dependence of the laser gain. In most cases this dependence represents a curve with one maximum. In the case of a more complex dependence (in particular, when the gain curve of a laser resonator with an active medium and a nonlinearly absorbing cell has a minimum and a maximum) a hysteresis loop of a laser system has the standard form expected for a passive system as shown in Fig. 1 (Ref. 1). This has been demonstrated experimentally.[104]

Any of the systems exhibiting optical bistability and belonging to the three classes mentioned above (hybrid, mirror, and mirrorless) can be operated in various regimes: steady-state, quasisteady (transient) and self-oscillatory stochastic. All these regimes have been investigated to a greater or lesser degree, but each time the use of a new nonlinear system reveals new features and different aspects important in the physical picture of optical bistability and in its practical applications.

The use of semiconductor materials, in which strong exciton interactions are utilized,[1,113,114] has made a major difference to the subject. Rapid progress in the technology of these materials (particularly in the construction of superlattices in the form of multiple quantum well structures—see, for example, Refs. 108 and 109) has made optically bistable devices of practical importance. These materials have already been used in optical triggers with a response (switching) time of 10^{-12} sec and a switching signal energy of 10^{-12} J (Ref. 1). These triggers together with the progress made in the development of fiber waveguides and a realization of stable nonlinear propagation regimes of picosecond and femtosecond laser pulses (formation of optical solitons) must be regarded as a major step in the development of ultrafast optical computers (Ref. 106).[4)] We have in mind not only a new basis for device construction, but also a new computer architecture involving parallel data processing.[1,106] The simplest model of such an optical computer will be constructed in the next few years[1] and the important point is not so much the speed of each component, but the number of components used in two-dimensional systems operating in parallel.

It thus follows that research on optical bistability involves tackling not only new physical phenomena, but dealing also with interesting applications.

1.2. These two aspects of physics and applications are manifested fully when the nonlinear medium is a liquid crystal. It is difficult to overestimate the role which liquid crystals have played primarily in the study of a new class of linear optical phenomena combined under the general name of optical bistability. It is in the case of liquid crystals that we can carry out a practically complete range of investigations of optical bistability. The main achievements so far are, firstly, a detailed investigation of nonsteady and transient optical bistability regimes in nonlinear Fabry–Perot resonators followed by a quantitative comparison of the theory and experiment, first carried out on liquid crystals (Ref. 41).[5)] Secondly, true intrinsic optical bistability in the absence of mirrors has been achieved in liquid crystals.[57,62] In addition to the widely used distributed-feedback systems,[10,31] there are also certain threshold effects specific to liquid crystals[12] which in terms of the physics of interaction of strong radiation with matter represent laser-induced phase transitions that do not involve temperature changes.[34,106] Thirdly, liquid crystals allow us to investigate instabilities and laws governing the transition to chaos in nonlinear optical systems, and such studies can be extended also to the polarization characteristics.[38,111] It is particularly important to note that the development of these processes can be studied in detail experimentally. This applies, in particular, to such a fundamental task as the identification of the actual scenario of the transition to chaos in a real physical process.[15] Finally and fourthly, the use of hybrid systems involving the application not only of optical but also of quasistatic fields to liquid crystals,[39] as well as changes in the boundary conditions,[80,84] greatly extend the qualitative picture of optical bistability, reduce the threshold characteristics, and shorten relaxation times. These last two properties are already being utilized in phase transparencies of sufficient resolution and response time for correction of phase fronts of light waves, including those in the infrared range (see, for example, Ref. 39).

All these investigations are of great interest from the point of view of physics and they are possible because of unusual properties of liquid crystals in which strong nonlinear optical effects can appear in optical fields generated by cw lasers of relatively low power: in some cases the changes in the refractive index induced by a laser field can reach values of the order of a few tenths for radiation intensities not exceeding 100 W/cm^2 (Ref. 12). Such a very strong optical orientational nonlinearity of nematic liquid crystals is associated with a strong anisotropy of liquid crystal molecules and the collective nature of their interaction with an external field; this anisotropy has now been thoroughly investigated (for details see, for example, Refs. 12, 90, and 92).

Although a strong optical nonlinearity of liquid crystals is characterized by a fairly slow response and the main advantages of the use of liquid crystals are related to identification of the physical picture of optical bistability, applications of liquid crystals are also of interest.[6)] We have mentioned already hybrid systems which are at present most promising for practical purposes.[39] Moreover, in many cases (in various technological processes, during prolonged monitoring of the operation of various devices, in construction of sensors of prolonged cumulative effects, etc.) one frequently requires systems with long response times whose properties (for example, transmission) vary slowly. Systems of this kind are being developed actively in molecular electronics.[40] Liquid crystals may become very valuable materials for the construction of optical devices characterized by a high immunity to brief external perturbations and by a dynamic memory representing the capability of accumulation of certain effects from one pulse to another.

Our review will be concerned mainly with the physical aspects of the use of liquid crystals in investigations of optical bistability and various instabilities that arise due to light-induced effects in liquid crystals. A clearer idea of the physical problems to be tackled can be gained from our introductory section (Sec. 2), which follows.

2. PHYSICAL DESCRIPTION OF DISPERSION OPTICAL BISTABILITY

Since in the majority of cases liquid crystals exhibit dispersion optical bistability, it will be useful to preface the main text with a short summary of the data on the operation of the simplest bistable device demonstrating dispersion bistability, which is a Fabry–Perot resonator filled with an optically transparent medium characterized by a nonlinear refractive index.[2] Feedback is established by repeated reflection of light from mirrors (d is the distance between the mirrors and R is the reflection coefficient) inside the resonator; the refractive index of the medium filling the resonator is $n = n_0 + n_2 I$, where I is the intensity of light inside the resonator.[6]

2.1. Steady-state case

We shall now write the standard formula for the intensity I_{out} of light transmitted by a system of interest to us and we shall supplement it by a phase shift due to the nonlinearity of the medium.

The simplest calculation method which can be used as the first approximation is based on a procedure adopted in linear optics when the optical field at the exit from a resonator is regarded as a sum of a series for field amplitudes due to successive transits of light across the nonlinear medium because of multiple reflections by the resonator mirrors.[7)]

In this case the relationship between the incident (I_0) and transmitted ($I_{out} = GI$) intensities is described by the usual expression

$$I = I_0 G^{-1}\left(1 + F\sin^2\frac{\Phi}{2}\right)^{-1}, \qquad (1)$$

where $F = 4R/(1-R)^2$; $G = n_0(1-R)/(1+R)$; $\Phi = (4\pi n_0 d/\lambda) + (4\pi n_2 d/\lambda)I \equiv \Phi_0 + \Phi_2 I$; Φ_0 represents the initial phase shift from a resonance of the transmission by a Fabry–Perot resonator; λ is the wavelength of light.

Since Φ depends on I, Eq. (1) establishes an implicit

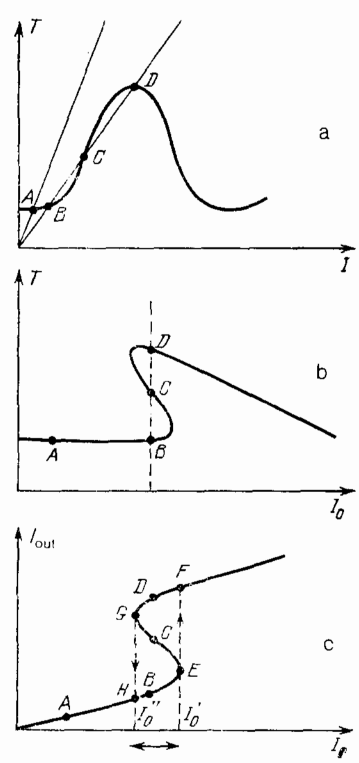

FIG. 2. Graphical solution of Eq. (1) and of the expression $T = I_{out} I_0^{-1}$, giving rise to optical bistability (explanations in text).[17]

relationship between I and I_0. The solution of Eq. (1) should be made graphically and this can be used to reveal various regimes of operation of a nonlinear Fabry–Perot resonator.[16,17]

In fact, the points of intersection of straight lines in Fig. 2a, representing an equation for the transmission $T \equiv I_{out}/I_0 = GII_0^{-1}$, with a curve representing Eq. (1) describe operating regimes of such a resonator. At low values of I_0 there is only one solution (point A); at higher values of I_0 we can have three solutions (points B, C, and D). The dependence of T on I_0 is demonstrated in Fig. 2b as a bistable characteristic of a resonator which is the dependence of I_{out} on I_0 and is shown in Fig. 2c.

We shall now analyze in greater detail the conditions for the appearance of various regimes in the case when $\Phi \ll 1$ (on the assumption that $n_2 > 0$ and $\Phi_0 < 0$).[8)]

Introducing $I_0 \equiv x_0$ and $I \equiv x$, we can readily reduce Eq. (1) to

$$x_0 = x\left\{1 + F\left[\frac{1}{2}(\Phi_0 + \Phi_2 x)\right]^2\right\} G. \qquad (2)$$

The right-hand side of Eq. (2) is a function of x, i.e., it is $f(x)$, and Eq. (2) describes the dependence $y = f(x)$. This dependence is represented in Fig. 3, where $x_{1,2}$ are found from the equation $df/dx = 0$:

$$x_{1,2} = [-2\Phi_0 \mp (\Phi_0^2 - 12F^{-1})^{1/2}] (3\Phi_2)^{-1}.$$

Several cases can now be distinguished.

a) When there are two unequal solutions of Eq. (2), a Fabry–Perot resonator exhibits optical bistability. The critical values of the light intensities I_0' and I_0'' at which the bistability is observed, i.e., at which hysteresis appears (Fig. 2), are found from the conditions $x_0 = f(x_2)$, representing an upward jump, and $x_0 = f(x_1)$, representing a downward jump. These values of I_0 naturally depend on Φ_0 subject to the condition $|\Phi_0| > 12/F$.

We shall now quote some numerical estimates for the case when a liquid crystal is the nonlinear medium in a Fabry–Perot resonator. For example, if $R \sim 0.9$, $\Phi_0 = -0.15$, $\lambda = 0.5\,\mu$, and if the parameters of the medium are typical of a nematic liquid crystal ($n_2 \approx 0.01$ cm^3/erg, $d = 100\,\mu$, $n_0 \approx 1.5$), we obtain $I_0' \approx 0.1$ W/cm^2, which is readily attainable using low-power cw lasers.

b) If Eq. (2) has one solution ($x_1 = x_2$), there is no optical bistability. This corresponds to the condition $\Phi_0 = \Phi_{0,min} \equiv -(12/F)^{1/2}$, which determines the threshold intensity $I_{0,th}$ for the observation of optical bistability:

$$I_{0,th} = c \cdot (8\pi)^{-1} \cdot 16 \cdot (3\sqrt{3})^{-1} G\Phi_2^{-1} F^{-1/2}. \qquad (3)$$

Equation (3) is identical, apart from numerical factors, with the value of $I_{0,th}$ given in Ref. 16 (compare also with Ref. 17). Substitution of the above values of the parameters in Eq. (3) gives $I_{0,th} \approx 0.03$ W/cm^2, i.e., a threshold which is very low compared with traditional values for nonlinear media.

c) If $|\Phi_0| < 12/F$, the curve in Fig. 3 has no inflection, i.e., a single-valued relationship between x and x_0 is established in a Fabry–Perot resonator. Two regimes are now possible: differential amplification, $\partial x/\partial x_0 > 1$, and power limitation, $\partial x/\partial x_0 \ll 1$ (Ref. 2).

We can easily show that in this case the first of these conditions is always satisfied at low values of I: $I < 2/\Phi_2(3FG)^{1/2}$, where $1/G \gg 1$; this gives $\Phi^{nl} \equiv \Phi_2 I < 0.2$ for $R = 0.9$ and $n_0 = 1.5$. If $\Phi_0 \to \Phi_{0,min}$, i.e., if $x_1 \to x_2$, then $\partial x/\partial x_0 \to \infty$. Therefore, each of the branches of the hysteresis loop ensures the most effective differential amplification regime. A similar analysis can be also carried out in the case of power limitation. For example, if $\Phi_0 \sim 0$, then Φ^{nl} obeys the condition $\Phi^{nl} \gg 2(3FG)^{-1/2}$, so that for the same values of R and n_0 as above we have the approximate estimate $\Phi^{nl} \gg 0.2$. Hence, a transmission maximum of a Fabry–Perot resonator ($x \sim x_0$) with typical parameters ($n_2 \sim 10^{-2}$ cm^3/erg, $d = 100\,\mu$m, $\lambda = 0.5\,\mu$m) occurs in the range $I_0 \gg 0.5$ W/cm^2. The same estimate for CS_2 ($n_2 \sim 10^{-11}$ cm^3/erg, $d = 1$ cm, $\lambda = 1\,\mu$m) gives a much higher value of $I_0 \gg 2.5$ MW/cm^2.

We can readily analyze also the conditions for the appearance of multistability in a Fabry–Perot resonator. For example, a second hysteresis (second jump) should be observed for $F \gg 1$ if $x_0 \sim GFx \sim (GF/\Phi_2)\pi$; in the case of a liquid crystal this would require $I_0 \sim 1$ kW/cm^2.

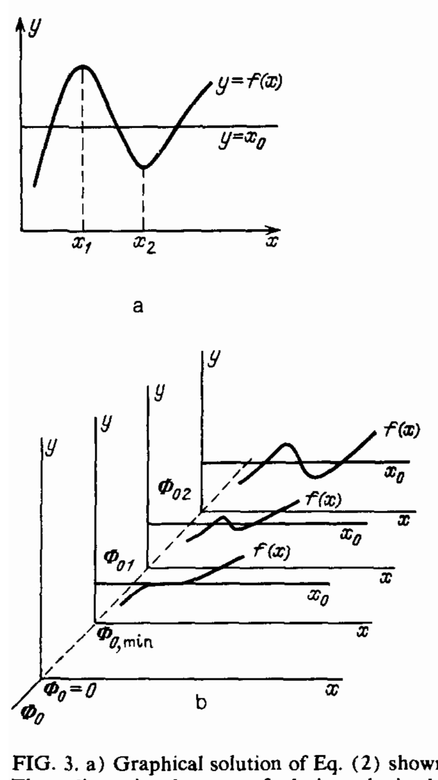

FIG. 3. a) Graphical solution of Eq. (2) shown in the (x, y) plane. b) Three-dimensional pattern of solutions obtained for different values of $\Phi_0 < 0$: $|\Phi_{0,min}| < |\Phi_{01}| < |\Phi_{02}|$; for each value of Φ_0 this solution is represented by the point of intersection of the $y = f(x)$ and $y = x_0$ lines in the (x, y) plane.

2.2. Transient regimes and instabilities

We shall now analyze the operation of a nonlinear Fabry–Perot resonator subjected to pulsed radiation. The physical processes which then occur are naturally governed by time-dependent parameters of the problem and we have to introduce such concepts as the duration of a laser pulse τ_p, the relaxation time of a nonlinear response of the medium filling the resonator τ_M, and the round trip time of a light wave in the resonator $t_R = 2dn/c$. Different regimes are obtained for different relationships between these time constants.

Nonsteady and transient regimes in a nonlinear Fabry–Perot resonator were investigated in detail earlier[41] and a quantitative comparison of the theory and experiment demonstrated excellent agreement. The nonlinear medium in this case exhibited the Kerr nonlinearity and was in the form of the isotropic phase of a nematic liquid crystal. A numerical analysis for the nonsteady case was carried out using the standard calculation method already mentioned: an allowance was made for repeated reflections of light from the resonator mirrors and an infinite series of the field amplitudes of transmitted radiation (E_T) was summed. In the approximation of small changes in the field amplitudes during one round trip in the resonator allowing for the Debye mechanism of the nonlinear response of the medium (time τ_M), it was found that

$$\tau_M \frac{\partial \Phi}{\partial t} + \Phi = \frac{3\omega d n_2}{c} (|E_F|^2 + |E_B|^2), \quad (4)$$

$$|E_T|^2 = n_0 T |E_F|^2 = n_0 T |E_B|^2 R^{-1}, \quad (5)$$

where E_F and E_B are the amplitudes of, respectively, the forward and backward waves inside the resonator; T and R are the intensity reflection and transmission coefficients of the mirrors; $\Phi = (\omega/c)\oint[\delta n(t,z)]dz$ is a nonlinear phase shift in one round trip through the resonator; $\delta n(t,z) = (\delta n_F + \delta n_B)$ is a correction to the refractive index n_0 in the quasisteady case, dependent on the intensity of light:

$$\delta n_{F,B} = n_2 |E_{F,B}|^2 + 2n_2 |E_{B,F}|^2.$$

The integral form of Eq. (4) is

$$\Phi(t) = \frac{3\omega d n_2 (1+R)}{n_0 c T \tau_M} \int_{-\infty}^{t} |E_T(t')|^2 \exp[-(t-t')\tau_M^{-1}]\, dt'. \quad (6)$$

Equations (4)–(6) describe dynamic properties of a nonlinear Fabry–Perot resonator and make it possible to analyze the problem numerically for different relationships between τ_p, τ_M, and t_R.

The main regimes which are possible in a nonlinear Fabry–Perot resonator are as follows.

If $t_R \gg \tau_M$, we can expect such effects such as a phase jump (switching),[18] appearance of oscillations with a period $2t_R$, and transition to a state of chaos (optical turbulence).[19,20] The switching time in this case is $> t_R$.

If $t_R \sim \tau_M$, various transient regimes are observed such as controlled phase switching and dispersion broadening of a pulse.[21] The switching time is now $\gtrsim \tau_M, t_R$.

If $t_R \ll \tau_M$, a self-oscillatory regime[4] is observed and the parameters of the system are slowed down critically, which is typical of a first-order phase transition.[22] The switching time is $\gtrsim \tau_M$ (which can be much shorter in the case of pulsed excitation[23]).

Some results obtained specifically for optical bistability in liquid crystals will be given in Sec. 3. Instabilities can be observed experimentally most conveniently in a ring resonator when an optical wave travels in just one direction. Then, $\Phi(t)$ is still described by Eq. (4), but the right-hand side is now different. In the case of practical situations, this situation can be rewritten in the form[19]

$$\tau_M \frac{\partial \Phi(t)}{\partial t} + \Phi(t) \approx A^2 [1 + 2B \cos(\Phi(t - t_R) - \Phi_0)], \quad (7)$$

where A and B are certain coefficients and Φ_0 still represents the initial detuning from the maximum transmission by the resonator. The optical field is then

$$|E(t)|^2 \approx A^2 + 2A^2 B \cos(\Phi(t) - \Phi_0). \quad (8)$$

A numerical solution of Eq. (7) which describes the time dependence of the output light intensity is presented in Fig. 4 for two values of the parameters A and B. This figure includes also the spectral information. We can see that the period is doubled (Figs. 4a and 4b) and that chaos sets in (Figs. 4c and 4d).

Figure 5 gives the experimental results[19] for a hybrid bistable device exhibiting period doubling and the appearance of chaos due to a change of the relationship between t_R and τ_M.[9)]

It must be stressed that the transition to chaos is not associated in these cases with the noise in the system. Such instabilities are usually called the Ikeda instabilities.[1]

There are also other optical instabilities: regenerative pulsations of the intensity of laser radiation transmitted by a Fabry–Perot resonator filled with a medium characterized

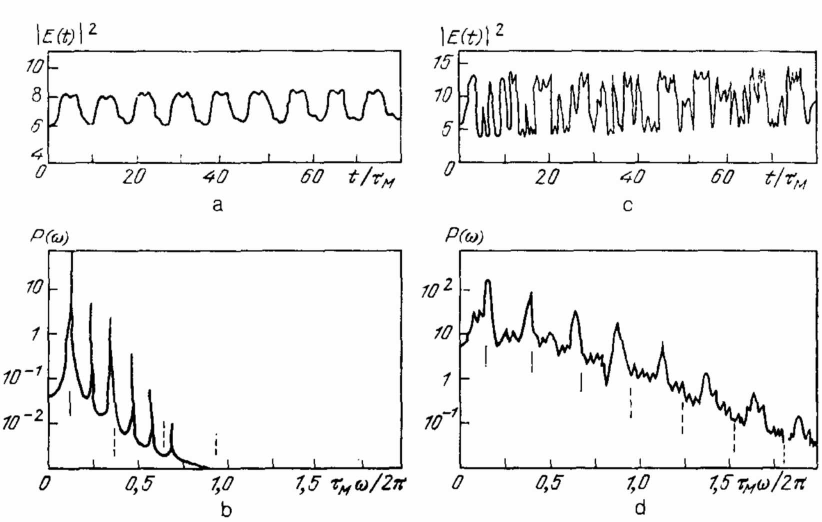

FIG. 4. Time dependence of $|E|^2$ plotted on the basis of Ref. 19. $B = 0.3$, $t_R/\tau_M = 3.5$, and $A = 2.17$ (a) or $A = 2.85$ (c). The lower part of the figure shows $P(\omega)$ spectra plotted for $B = 0.3$, $t_R/\tau_M = 3.5$, and $A = 2.17$ (b) or $A = 2.85$ (d). The continuous curves and the dashed vertical lines in Figs. 4b and 4d identify the values of $\tau_M\omega/2\pi$ for unstable and stable regimes, respectively.

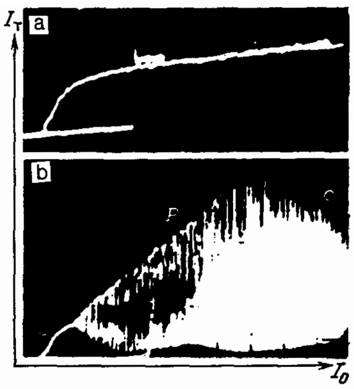

FIG. 5. a) Dependence of I_T on I_0 obtained by gradual increase and reduction in I_0, from zero to some maximal value and back again in $t = 30$ sec (Ref. 19); $t_R = 160\,\mu\text{sec} \ll \tau_M = 1$ msec. b) Dependences as in Fig. 5a, but for $t_R = 40\ \text{msec} \gg \tau_M = 1$ msec. Here, S, P, and C represent stable, periodic, and chaotic states of the system on the upper branch of the hysteretic curve of Fig. 5a.

by two competing optical nonlinearity mechanisms and with different nonlinear response times.[4] The appearance of these pulsations is easily understood on the basis of a graphical analysis presented in greater detail in Fig. 2 (Ref. 43). In fact, under steady-state conditions the total phase shift during one trip in a Fabry–Perot resonator can be written in the first approximation as follows:

$$\Phi = \Phi_0 + \Phi_\theta + \Phi_T, \qquad (9)$$

where

$$\Phi_\theta = K\alpha_\theta T I_0, \quad \Phi_T = K\alpha_T T I_0,$$

Φ_0 is independent of the intensity, K is a constant governed by the resonator parameters, α_θ and α_T are the coefficients of proportionality corresponding to the two mechanisms[10)] inducing Φ, and T is the transmission coefficient of the resonator. The dependence (9) is plotted in Fig. 6 and we can see that it is a straight line. The working point of a Fabry–Perot resonator is then determined by one of the points of intersection of this line with the transmission curve I_T of the resonator. However, the working point, for example O (Fig. 6) actually describes stable operation of the resonator only if this point is stable against rapid thermal fluctuations (because the orientational mechanism does not have sufficient time to develop). Therefore, the thermal mechanism may shift the working point to a position A (or B). Then, because the laser intensity I in the resonator corresponding to the point A is lower (or greater) than at the point B, the orientational mechanism reduces (or increases) Φ_θ. This shifts the working point from A to C (or from B to C') and the dashed line in Fig. 6 shifts to the left (or right). Beyond the point C (or C') the thermal mechanism results in a shift to a position denoted by D (or D'). The resultant increase (or reduction) in I now increases (or reduces) Φ_θ because of the orientational mechanism and shifts the working point from D to C' (or from D' to C), which is followed by switching to D' (or D) and a shift toward A (or B). Then, the cycle $ACDC'D'A$ is repeated and the self-oscillatory operation of a Fabry–Perot resonator, which is not limited in time, is observed. The period of such oscillations (pulsations) is of the order of the orientational relaxation time τ_θ.

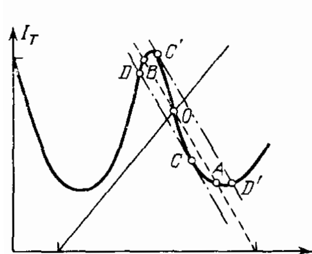

FIG. 6. Explanation of a self-oscillatory regime.[43] Here, O is the working or operating point; the dashed and chain lines represent the phase shift due to heating by radiation, the continuous line is the solution of Eq. (9), and the continuous thick curve represents transmission by a Fabry–Perot resonator.

This description is valid for $\tau_T \ll \tau_\theta$ and we can readily see that the threshold intensity of the fluctuations I_{fluct} should be found from the condition that the dashed line in Fig. 6 is a tangent to the transmission curve of a Fabry–Perot resonator at the point of inflection:

$$|K\alpha_T I_{fluct}|^{-1} = \left|\frac{dT}{d\Phi}\right|_{max} \approx \frac{F^{1/2}}{2}, \qquad (10)$$

where F is the sharpness (finesse) coefficient of the resonator.

Such phenomena are particularly numerous in laser systems of various types so that under certain conditions instabilities of this kind are the rule rather than the exception[1] (see also Ref. 123).

An interesting regime in a passive nonlinear system with optical bistability is predicted in Ref. 105: under certain conditions[11)] it is possible to generate a time sequence of short light pulses using a single "seed" pulse injected into the system; an experimental demonstration of this effect was reported so far only for a model device in which an acoustic wave was excited (see Ref. 123).

2.3. Other optical systems with bistability

A relatively simple system in the form of a nonlinear Fabry–Perot resonator has been discussed by us in the preceding sections. However, there are other optical systems in which optical bistability and unstable processes may appear. Even in the case of a Fabry–Perot resonator it is possible to think of more complex systems. In particular, the complexity may increase because of the presence of various elements operating by transmission or reflection.[110] In the latter case one can use selective reflectors based on dielectrics (interference due to plates; diffraction by apertures and various—including those induced by external fields—structures; reflection near the total internal reflection and Brewster angles, etc.), thin films of metals or semiconductors deposited on dielectrics. The more general problem of propagation of light in a nonlinear layer of finite thickness is related to the case of a nonlinear Fabry–Perot resonator; the boundary conditions give rise to reflected waves and these establish a feedback.

The phenomena due to feedback can cover a wide range: since a feedback is established in respect of the fields, new nonlinear effects can be expected: transverse optical bistability due to spatial modulation of the beam,[35] polarization optical bistability and multistability, and polarization chaos.[106] The problems related to the polarization of radiation have now become an independent topic and are of exceptional importance. In the case of spatially confined light beams one can expect various self-maintained spatially inhomogeneous regimes, in particular traveling switching waves in a Fabry–Perot resonator.[35]

Essentially, optical bistability can appear in any nonlinear system with feedback and is a fairly universal property.[1] In fact, Eq. (2) is cubic. Substitution of the variable in the form $x' = x + 2\Phi_0(3\Phi_2)^{-1}$ makes it possible to rewrite this equation in a more convenient form:

$$\Phi_2^2 x'^3 + \left(4F^{-1} - \frac{1}{3}\Phi_0^2\right) x' + \left[4x_0 G^{-1}F^{-1} - 2\cdot\frac{1}{3}\Phi_0\Phi_2^{-1}\left(4F^{-1} - \frac{1}{3}\Phi_0^2\right)\right] = 0. \quad (11)$$

A dynamic equation describing oscillations of a nonlinear (anharmonic) oscillator can be readily reduced[8,27] to the form of Eq. (11). A detailed analysis of the regimes which can then appear has been made using the theory of nonlinear oscillations; the first treatment of these problems in connection with optical bistability was given in Ref. 27. From the microscopic point of view, we are speaking here of a well-known problem in nonlinear optics, which is allowance for the anharmonicity of atomic systems. In this case a local nonlinear response of a medium can no longer be regarded as small.[106] A feedback loop in a nonlinear oscillator is established by the dependence of its natural frequency on the oscillation amplitude, i.e., by the property of nonisochronism[8] (see also Ref. 9).

A natural result of the use of this oscillator model, frequently called the Duffing oscillator,[12)] in the case of wave processes is the appearance of wave bistability: optical bistability and hysteretic effect are transferred to a propagating wave.[106] At high intensities the process of stochastization of oscillations of a nonlinear oscillator[107] results in stochastization of the wave process.[26,121] At present these effects are attracting most interest.[106] This type of optical bistability is exhibited by a large class of nonlinear optical effects.[13)] Many systems exhibiting optical bistability can effectively be reduced to this case.

Optical bistability can appear in a nonlinear system if there is some point near which its properties change considerably. Then, in the case of an initial "detuning" from this point, the nonlinearity of the system and a strong feedback which appears in the system induce a transition between two stable states[14)] governing optical bistability. The appearance of such a singular point may be related to some resonance in the system (nonlinear Fabry–Perot resonator[17] or anharmonic oscillator[27]) or to total internal reflection,[29] excitation of surface waves,[30] distributed feedback,[31] light-induced bleaching of an initially inhomogeneous medium,[32] etc. Another possibility is that the physics of the phenomenon itself permits interactions with a light-intensity threshold (self-focusing,[33] threshold reorientation of molecules typical of liquid crystals,[12,34] etc.). We shall make no distinction between the two types of hysteresis, the first when only the state of the optical field changes [15)] and the second when the state of the nonlinear medium itself is modified. We shall regard these two cases as representing optical bistability regimes.

Optical bistability based on the interaction between two plane waves propagating opposite to one another in an isotropic medium with the Kerr nonlinearity and inducing anisotropy in the medium has a characteristic feature. The dependences of the output parameters of a light wave on the input parameters are not only multivalued (this gives rise to a hysteresis in the system), but also have specific isolated semiinfinite branches when the number of upward and downward jumps is not the same.[37] Moreover, in a quasi-steady regime there are also branches from which a system can only drop down (downward jumps), but cannot go up. The solutions are related to an internal resonance in a nonlinear system induced by a field in the case of a four-wave interaction. They appear for a specific form of the cubic nonlinearity of a medium and can play an important role in the appearance of instabilities in the system (see Ref. 120). However, these solutions may be associated with the simplification of the model of a nonlinear medium and the approximations used in the calculations. In particular, neglect of absorption can play a decisive role: it is well known, for example, that it is absorption (attenuation) which ensures a transition between different states when optical instability occurs in an anharmonic oscillator.[8] We have to allow also for the nonsteady nature of the problem (see Refs. 75 and 120).

3. OPTICAL BISTABILITY IN LIQUID CRYSTALS

Many of the phenomena which result in optical bistability have already been investigated for liquid crystals. Before discussing some of the topics considered above, it would be desirable to begin with a brief overall review of the results obtained, so as to gain an idea of the range of investigations already carried out on liquid crystals.

Detailed quantitative theoretical and experimental studies of the optical response of a nonlinear Fabry–Perot resonator, including the nonsteady case, were reported in Ref. 41, where the isotropic phase of a nematic liquid crystal was used as a nonlinear medium of the Kerr type.

Measurements were made for different values of the phase shift $\Phi_0 = 0$, -0.1π, and -0.2π, where $\Phi_0 = 2\omega n_0 d/c$, each of which corresponded to a specific mode of a nonlinear Fabry–Perot resonator in a stable state. Figure 7 shows three typical examples. Theoretical curves of the output pulses were obtained from Eqs. (4)–(6). One can also show that if $t_R \ll \tau_p \ll \tau_M$, then the dependence of $|E_T(t_{max})|^2$ on $|E_0(t_{max})|^2$, where $t = t_{max}$ corresponds to the maximum value of Φ, should reach a characteristic curve for a steady state of a nonlinear Fabry–Perot resonator (Fig. 8). We can distinguish three types of operation: power limitation (Fig. 8a), differential amplification (Fig. 8b), and bistable (Fig. 8c).

A reduction in τ_M modifies these dependences. In the regime of Fig. 8c this modification is manifested mainly in the negative curvature region. In the limit of very small values of τ_M, the curve becomes similar to a characteristic switching curve of the phase, which is in good agreement with the dependence mentioned above in the case of quasi-steady operation at the maximum intensity of input pulses $I_{max} = 0.3\ \mathrm{MW/cm^2}$.

The experiments reported in Ref. 41 yielded also the dependences $I_T(t)$ and $\Phi(t)$ as a function of $I_0(t)$ when the molecular relaxation time was reduced from $\tau_M \gg \tau_p \gg t_R$ to $\tau_M \ll t_R \ll \tau_p$. For $\tau_M \gg \tau_p$, the $\Phi(t)$ curves demonstrated a nonsteady response of a medium to the field in a Fabry–Perot resonator; however, if $\tau_M \ll \tau_p$, the medium could follow almost instantaneously the changes in the field in the

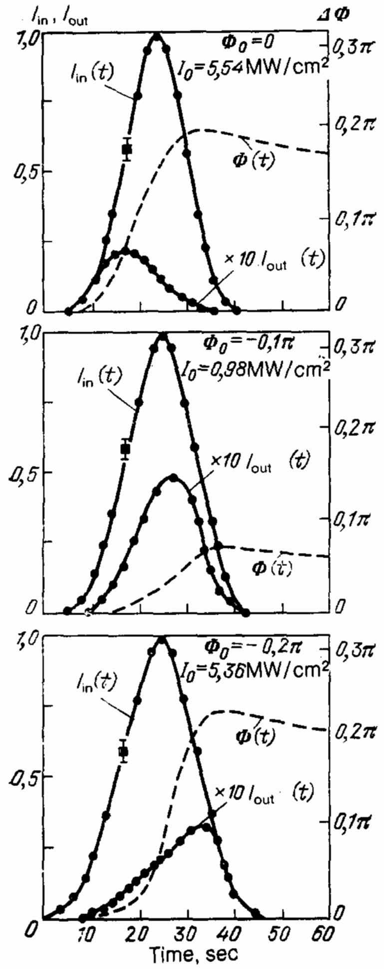

FIG. 7. Input $I_{in}(t)$ and output $I_{out}(t)$ intensities obtained for a nonlinear Fabry–Perot resonator as a function of time for three different values of the initial phase shift Φ_0 (Ref. 41). The continuous curves are calculated using Eqs. (4)–(6) and the dashed curve represents field-induced phase shift $\Phi(t)$; I_0 is the peak intensity of the input signal.

resonator so that $\Phi(t) \propto I_T(t)$. A quantitative study was made of the transition from a nonsteady to a quasisteady case. In the power limitation regime ($\Phi_0 = 0$) the dependence of I_T on I_0 in the nonsteady case was in the form of a loop which readily collapsed to form a line in the cases far from the transition. In the differential amplification case ($\Phi_0 = -0.1\pi$) we observed the same nature of the dependence, but the hysteresis curve for I_T considered as a function of I_0 was quite pronounced in the quasisteady limit. Bistable operation ($\Phi_0 = -0.2\pi$) demonstrated a more abrupt transition between the two limiting cases under discussion. In the nonsteady limit the dependence $I_T(I_0)$ was also in the form of a loop, but the direction along which the loop was followed was opposite to that in the cases discussed above.

The results obtained for $\tau_M \ll t_R \ll \tau_p$ corresponded to the conditions under which the Ikeda instabilities should develop, but in the experiments of Ref. 41 the detector recording the optical radiation was too slow. Moreover, appearance of the Ikeda instabilities would require light intensities 50 times higher[1] than those used in the experiments.

Strong optical bistability effects in a Fabry–Perot resonator with a liquid crystal in the nematic phase were first

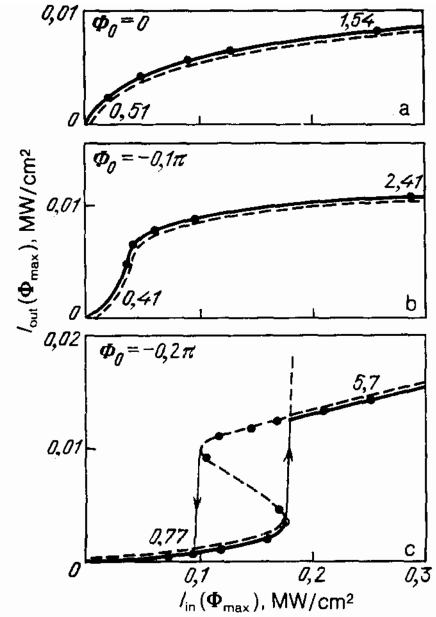

FIG. 8. Dependences of I_{out} on I_{in} for a maximum phase delay Φ_{max} and the three values of Φ_0 (a–c).[41] The dashed curves represent the steady-state behavior of a nonlinear Fabry–Perot resonator. The continuous curves are calculated numerically for the quasisteady case ($\tau_r = 2$ nsec, $I_0 = 0.3$ MW/cm²); the numbers alongside the experimental points give the peak intensity I_0 in our experiments (MW/cm²).

considered in the reports cited as Ref. 42. Optical bistability and multistability had been observed[43] in the field of an Ar^+ laser and a transition to a self-oscillatory regime was also found (the nonlinearity of the medium was governed by

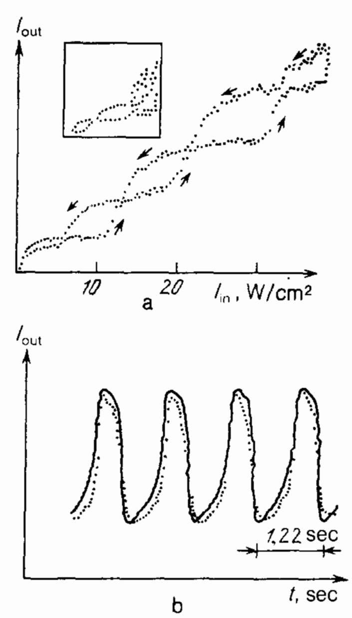

FIG. 9. Multistable characteristics (a) and oscillations (b) exhibited by a Fabry–Perot resonator containing a nematic liquid crystal, observed experimentally in Ref. 43. The top of Fig. 9a shows loops and the appearance of spontaneous oscillations in the case of other (more favorable) initial conditions. The measured values of the parameters were as follows: $\tau_\theta = 1.6$ sec, $\tau_T = 0.015$ sec, $\alpha_\theta = 0.011$ cm²/W, $\alpha_T = 0.0013$ cm²/W. The measured value of the threshold intensity above which spontaneous oscillations appeared in the investigated Fabry–Perot resonator amounted to $I_{osc} \approx 63$ W/cm². An estimate based on Eq. (10) allowing for the resonator parameters found experimentally[43] ($K = 7$, $F = 20$) gave $I_{osc} \approx 50$ W/cm². The measured pulsation period was 1.2 sec.

orientational and thermal mechanisms); see Fig. 9 and compare with Fig. 6. A similar regime was also observed when a cell with a nematic liquid crystal was placed inside the resonator of an Ar^+ laser.[44] Optical bistability appeared in a Fabry–Perot resonator when the field was in the form of a train of nanosecond laser pulses: in view of strong feedback, just a few pulses were sufficient to manifest the accumulation effects.[42]

The main features of the transient processes in liquid crystals in the mesophase were due to the fact that the time τ_M was independent of the cell thickness and of the characteristic scale of the resultant deformations.[12] Therefore, the value of τ_M could be varied continuously within wide limits (in practice from 10^{-6} sec to tens of seconds). Moreover, τ_M essentially represents the switching-off time of a nonlinearity when an external field is removed ($\tau_M \equiv \tau_{off}$); the switching-on time τ_{on} itself depends on the intensity and can be varied continuously In certain geometries (threshold reorientation) there is moreover a specific delay time τ_0 because of the inequivalence of two possible but opposite directions of the initial reorientation of the director.[24] It is necessary to allow also for the effects of accumulation of nonlinear properties of liquid crystals under the action of a train of short (nanosecond and possibly picosecond) laser pulses.[25] Therefore, the dynamics of orientational processes in liquid crystals can be quite complex.

Polarization optical bistability of a cholesteric liquid crystal exhibiting a helical structure in space was discussed in Ref. 45: in this case a Fabry–Perot resonator exhibits not only scalar (frequency), but also polarization modes (optical bistability of the polarization of transmitted waves was demonstrated experimentally for nematic liquid crystals some time ago[46]). Chaotic regimes in the polarization and amplitude characteristics revealed by an analysis of a nonlinear Fabry–Perot resonator containing the isotropic phase of a cholesteric liquid crystal, exhibiting optical activity, were reported in Ref. 112.

Mirrorless optical bistability systems had also been realized experimentally for liquid crystals, but the investigations had been so far in the nature of demonstrations. Nonlinear reflection (in the case of the thermal nonlinearity mechanism) at an interface between a nematic liquid crystal and glass was reported in Ref. 47 for total internal reflection. An optical hysteresis on excitation of surface plasmons at an interface between a nematic liquid crystal and a metal was recorded in Ref. 48 (Fig. 10). Surface phenomena were involved in optical bistability observed in a system comprising a photoconducting semiconductor and a nematic liquid crystal[49]: a static field which appeared in the semiconductor because of the anomalous photo-emf effect resulted in surface reorientation of the liquld crystal characterized by hysteresis as the incident light was increased and then reduced (Fig. 11). A cell with two substrates carrying semiconductor films readily exhibited a tristable characteristic (double hysteresis) illustrated in Fig. 11c: the scattering of light in the bulk of a liquid crystal manifested hysteresis because of the effect of the field on the liquid crystal on the exit substrate, which occurred at higher intensities of the incident light than on the entry substrate. An additional feedback component (reflecting mirror) widened the optical bistability region (Fig. 11b). Nonlinear bleaching of a nematic liquid crystal layer with two free surfaces was described in Ref.

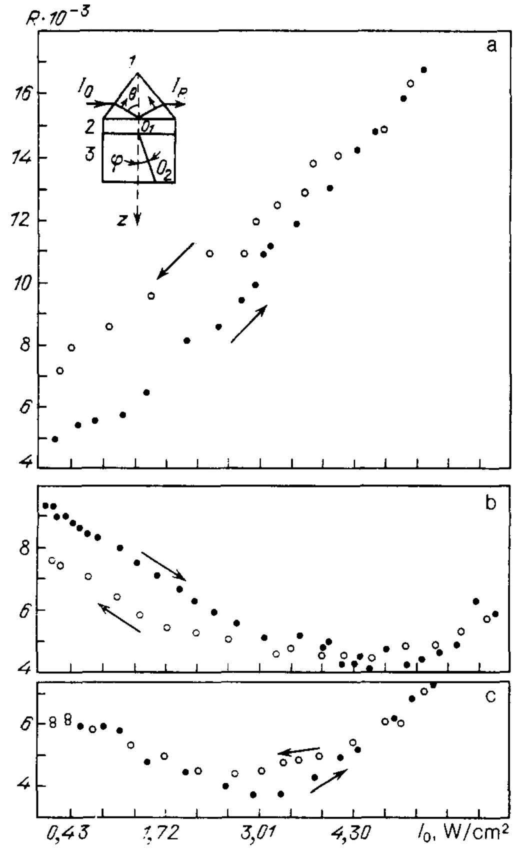

FIG. 10. Hysteresis curves obtained on excitation of nonlinear surface electromagnetic waves in an experiment involving frustrated total internal reflection in a system (inset in Fig. 10a) consisting of a prism (1), a metal film (2), and a nematic liquid crystal (3), based on Ref. 48. Here, O_1O_2 is the direction of the optic axis of the nematic liquid crystal; I_0 and I_R are the intensities of the incident and reflected light, respectively; $R \equiv I_R/I_0$. The initial deviation from the angle θ_r corresponding to a resonance of surface plasmons was as follows: a) $\theta_r - \theta \approx -1°$; b) $\theta_r - \theta \approx 1°$; c) $\theta_r - \theta \approx 30'$. The actual shape of the hysteresis loop is governed by the initial deviation $\theta_r - \theta$ from the resonance and by the state of the system after a change of the direction of going round a hysteresis loop (starting with the direction corresponding to an increase in I_0). The experiments were carried out using a He–Ne laser ($\lambda = 0.633$ μm) and a nematic liquid crystal 5CB; $\varphi = 0^0$.

50.[16]) A relatively simple theory of nonlinear bleaching of nematic liquid crystals, based on a molecular theory of nonlinear scattering of coherent light under conditions of optical orientation of molecules,[71,73] resulting in optical bistability, was developed in Ref. 55 (one could speak thus of one more optical bistability mechanism; compare with Ref. 6).

Optical bistability in a liquid crystal with distributed feedback can be due to self-diffraction effects, when periodic structures are induced by the laser radiation itself[51,53-55] or when a liquid crystal is itself spatially inhomogeneous (cholesteric and smectic liquid crystals).[83] In the former case the appearance of optical bistability should be associated with saturation of the nonlinearity (when the nonlinear coupling coefficient γd, where d is the thickness and γ is governed by the nonlinear correction to the refractive index Δn^{nl}, should obey the inequality $\gamma d \gtrsim 10$, as shown in Ref. 31; current experiments[56] have achieved only the condition $\gamma d \sim 2$). In the latter case the phenomenon of optical bistability is due to Δn^{nl} induced under the Bragg resonance conditions (Ref. 57)[17] (the polarization form of optical bistability was dis-

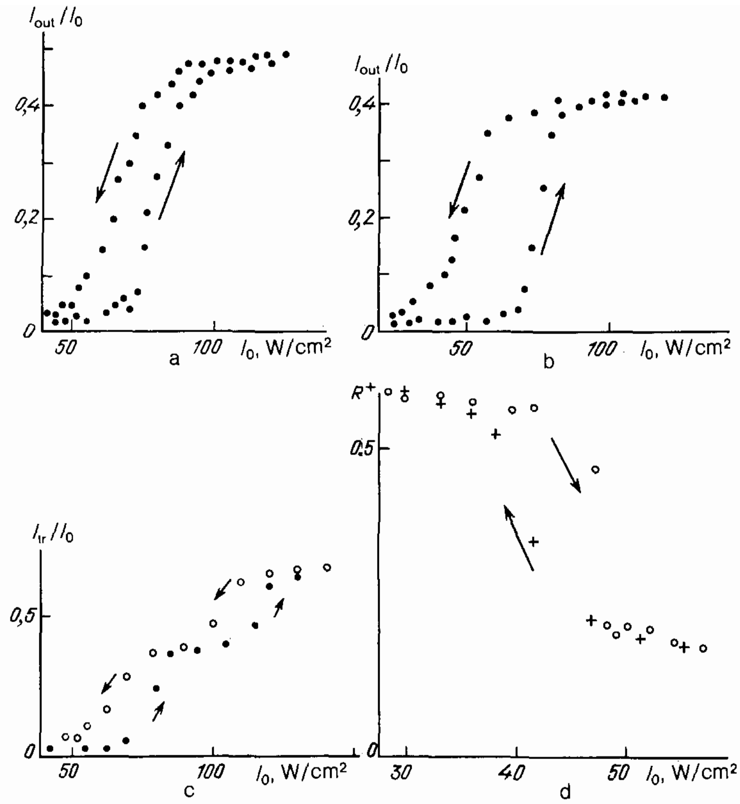

FIG. 11. a) Hysteresis of the dependence of the intensity of the transmitted light I_{out} on the intensity of the incident light I_0 in a system comprising a photoconducting semiconductor and a nematic liquid crystal when the value of I_0 was gradually increased and then reduced. b) Broadening of a hysteresis loop due to a mirror reflecting partly ($r = 60\%$) transmitted radiation. c) Double hysteresis (tristability) in a nematic liquid crystal cell with semiconductor films on both substrates; the light-induced static electric fields due to the anomalous photo-emf were parallel on both substrates. d) Hysteresis of the dependence of the reflection coefficient of light $R^+ \equiv I_{refl} I_0^{-1}$ (under conditions of a Bragg resonance for a mixture of cholesteric liquid crystals) on the intensity of the incident light I_0 when this intensity was gradually increased and reduced (as indicated by arrows). Measurements were carried out at stationary points when an equilibrium thermal state was established in a sample for each value of I_0. In all cases the experiments were carried out using an He-Ne laser ($\lambda = 0.633\,\mu$m) and a nematic liquid crystal 5CB (a–c)[49] or a mixture of cholesteric liquid crystals (d).[59]

cussed for this case in Ref. 58). The latter effect was observed experimentally[59] (Fig. 11d) in the case of the thermal mechanism of the nonlinearity of a mixture of cholesteric and nematic liquid crystals with a dye subjected to the field of a low-power He-Ne laser. (Some systems exhibiting optical bistability in cholesteric liquid crystals, similar to those reported in Refs. 32, 33, and 57, were discussed in Ref. 60.) Hysteresis during pulsed excitation of a cholesteric liquid crystal, due to distortion of the profile of the light pulses resulting from relaxation of the medium (observation of a hysteresis in this case did not necessarily imply optical bistability[7]), was reported in Ref. 61.

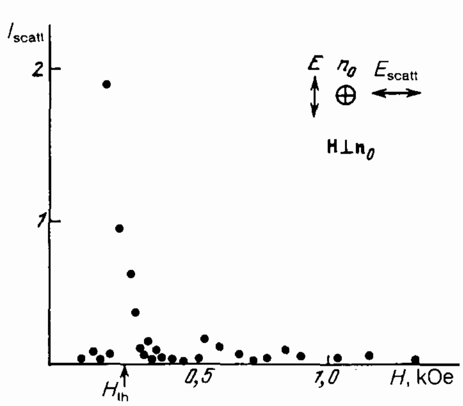

FIG. 12. Dependence on H of the intensity I_{scatt} of light scattered (in the direction of the incident beam) by transverse fluctuations of the director $\mathbf{n}_0$ at the threshold of the Fréedericksz transition in a static magnetic field H[63]. The top part of the figure shows the initial geometry (homeotropic cell containing a nematic MBBA liquid crystal) in a plane perpendicular to the direction of propagation of light; the polarization of light was $\mathbf{E} \| \mathbf{H}$; the arrow identifies the threshold value of the magnetic field H_{th}.

The first experimental observation of the true intrinsic phenomenon of optical bistability in a nematic liquid crystal, associated with a threshold reorientation of molecules in a laser field (field-induced phase transition[34]) was reported in Ref. 62. Light scattering experiments[63] revealed a strong rise of the amplitude of fluctuations at the reorientation threshold of a nematic liquid crystal (Figs. 12), which was an analog of the critical opalescence in the case of thermal phase transitions. Critical behavior had been observed also in the case of the time parameters [54,62]

Optical bistability associated with aperture effects such as the self-focusing of light[33,72] etc., in which a change in the transverse structure of a light beam results in feedback (which could be established also by optical components such as reflecting mirrors, lenses, stops, polarizers, etc.) was reported for a nematic liquid crystal in Ref. 64. These effects were found to be strongest in liquid crystals and they gave rise to a specific ring structure of the transmitted light.[65]

Therefore, a wide range of nonlinear optical phenomena in liquid crystals can give rise to optical bistability. In the next sections we shall discuss in detail some of the effects which at present are attracting most interest.

4. INTRINSIC OPTICAL BISTABILITY ASSOCIATED WITH A LIGHT-INDUCED STRUCTURAL PHASE TRANSITION IN A NEMATIC LIQUID CRYSTAL

A characteristic feature of the light-induced effects in liquid crystals is the existence of an optical intensity threshold I_{th} above which reorientation of the director $\mathbf{n}_0$ takes place (the director represents the average orientation of the molecules, i.e., the optic axis) so that the system undergoes a transition from an initial state with a homogeneous orientation of the medium to an inhomogeneous state. The existence of a singular point which is the threshold of appearance of this effect is not dependent on the presence of a resonator, so that we can speak here of intrinsic optical bistability. Two states of the system in which this bistability can be observed correspond to the incident light intensities $I<I_{th}$ and $I>I_{th}$. A transition between these two stable states may be accompanied by hysteresis which can occur, for example, in the case of a nonlinear phase shift due to a gradual increase and reduction in I if the feedback is selected suitably.[2]

In this case feedback is governed by the self-consistency of the problem: an optical field distorts the structure of a nematic liquid crystal, which influences the parameters of the transmitted radiation: self-modulation of light takes place. Elastic properties of the medium ensure that the nonlinear response is nonlocal. Therefore, hysteresis characterizing a first-order phase transition may appear for suitable values of the material parameters of the medium itself. In fact these parameters determine the macroscopic conditions in an experiment and can be altered readily by, for example, selecting the boundary conditions or using not only an optical field but also additional electric or magnetic fields.[34]

The same conclusion can be reached from a somewhat different standpoint[12]: since we are speaking here of light-induced phase transitions, it follows that the external conditions (the existence of an additional external field or a change in the angle of incidence of light) can alter the nature of the phase transition converting it from one of second order (without hysteresis) to one of first order (with hysteresis). In the case of nematic liquid crystals this means that when the threshold is overcome ($I>I_{th}$) then in the former case the angle of reorientation θ of the molecules (and, consequently, the nonlinear phase shift Φ^{nl} of the transmitted radiation) changes continuously, whereas in the latter case it changes abruptly.[34]

The results of the first observations of hysteresis of a light-induced (in a field **E**) threshold reorientation of a nematic liquid crystal in the presence of a static magnetic field **H** were reported in Ref. 62. The experiments involved determination of the amplitude and time characteristics of Φ^{nl} induced in 4-n-pentyl-4′-cyanobiphenyl (5CB) by Ar^+ laser radiation ($\lambda = 0.51\,\mu$) both at different but fixed values of I as a function of $H(\mathbf{E}\perp\mathbf{H}, \mathbf{H}\|\mathbf{n}_0)$ or vice versa. The results obtained in the latter case are presented in Fig. 13. We can see that hysteresis (of the dependence of Φ^{nl} on I) occurred in fields $H\geqslant H_c$. The values of I_c and H_c could be estimated most accurately from measurements of the switching-on (τ_{on}) and switching-off (τ_{off}) times of the nonlinearity, corresponding to the "instantaneous" opening and stopping the aperture transmitting the Ar^+ laser beam. The values obtained in this way were $I_c\approx 84$ W/cm^2 and $H_c\approx 181$ Oe.

At low values of I the dependences of τ_{on}^{-1} on I obtained at different values of H were linear. At high values of H they were nonmonotonic and exhibited critical behavior.

The role of a magnetic field in this geometry was simply to increase I_{th} and τ_{on} and to reduce τ_{off} provided I was not too high ($I<200$ W/cm^2).

In the case when $\mathbf{E}\|\mathbf{H}$ ($\mathbf{E}, \mathbf{H}\perp\mathbf{n}_0$) it is possible to observe under certain conditions (with ordinary and extraordinary waves inside a nematic liquid crystal) an interesting effect in the form of disappearance of reorientation in fields

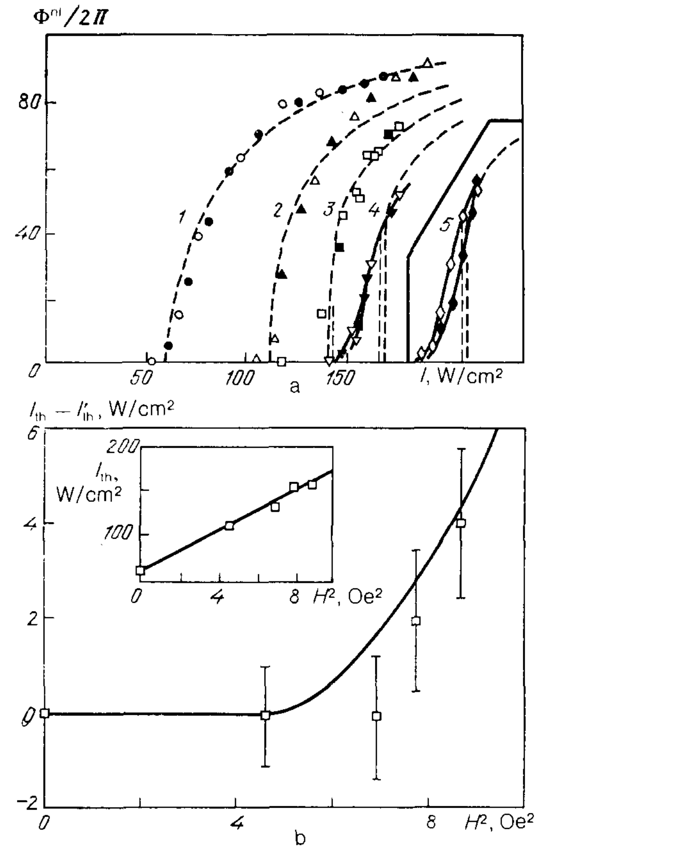

FIG. 13. a) Dependences of the nonlinear phase shift Φ^{nl} induced by an optical field **E** on the intensity of light I obtained in the geometry of the light-induced Fréedericksz transition, obtained in Ref. 62 for different values of the magnetic field H/H_0: 1) 0; 2) 0.92; 3) 1.15; 4) 1.32; 6) 1.35. Here, H_0 is the threshold value for the Fréedericksz transition in a magnetic field when $\mathbf{E}\perp\mathbf{H}$. The symbols are the experimental values obtained by increasing I (black) or by reducing I (open); the curves are calculated. b) Width of the hysteresis loop $I_{th}-I'_{th}$ plotted as a function of H on the basis of the results reported in Ref. 62. The two threshold values of the intensities were recorded for increasing and decreasing I, respectively. The inset shows an increase in I_{th} on increase in H in the case of a nematic liquid crystal 5CB ($d=380\,\mu$m).

$H>H_0$ in the absence of an optical field (H_0 is the threshold of the Fréedericksz transition in a magnetic field), which is discussed in Sec. 6.

A standard expression for the density of the free energy F in a nematic liquid crystal, considered as a function of the amplitude of the small angle of reorientation θ_m which acts as the order parameter, is (see, for example, Ref. 34)

$$F=-C\theta_m^2+\frac{1}{2}B\theta_m^4+\frac{1}{3}G\theta_m^6+\ldots,\qquad \theta=\theta_m\sin\frac{\pi z}{d},\tag{12}$$

where the coefficient $C=(I/I_{th}^0)^{1/2}-1$ determines the threshold of the light-induced Fréedericksz transition; the parameter B in the case when $\mathbf{E}\perp\mathbf{H}$ and $\mathbf{H}\|\mathbf{n}_0$ is governed by the parameters of the medium and by H:

$$B=\frac{1}{4}\left(1-\frac{K_{33}-K_{11}}{K_{33}}\right)-\frac{9}{4}\left(1-\frac{\varepsilon_\perp}{\varepsilon_\|}\right)-\frac{9}{4}\left(1-\frac{\varepsilon_\perp}{\varepsilon_\|}\right)\frac{\chi_a H^2}{K_{33}(\pi/d)^2},$$

where K_{ii} are elastic constants; $\varepsilon_\perp$ and $\varepsilon_\|$ are the components

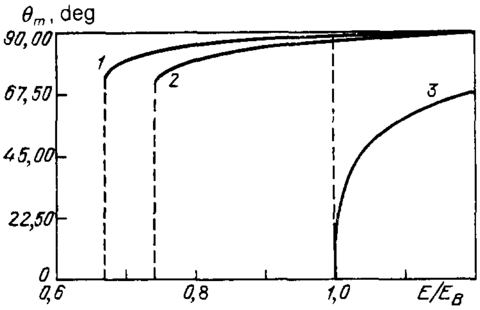

FIG. 14. Calculated dependences of the maximum of the reorientation angle θ_m on the normalized intensity E/E_B of a quasistatic electric field of frequency f applied to a nematic liquid crystal RO-TN-200 in the geometry of the threshold Fréedericksz transition.[62] Parameters: $K_{11} = 9.21\times10^{-7}$ dyn, $K_{33} = 14.83\times10^{-7}$ dyn, $\varepsilon_\perp = 7.58$; E'_B is the threshold value of the field during its reduction. Curve 1: $f = 10$ kHz, $\varepsilon_\parallel = 26.00$, $B = -0.376$, $E'_B = 0.67E_B$ (first-order transition). Curve 2: $f = 140$ kHz, $\varepsilon_\parallel = 21.50$, $B = -0.330$, $E'_B = 0.74E_B$ (first-order transition). Curve 3: $f = 560$ kHz, $\varepsilon_\parallel = 9.36$, $B = 0.013$ (second-order transition); $E_B = \pi d^{-1}(4\pi K_{33}\varepsilon_\parallel/\varepsilon_a\varepsilon_\perp)^{1/2}$.

of the permittivity across and along the director, respectively; χ_a is the anisotropy of the magnetic susceptibility; the last constant in the above equation is $G > 0$ for known nematic liquid crystals. Equation (12) does not have terms which are even in powers of θ_m because of the equivalence of two directions of the director reorientation ($\pm\theta_m$).[24]

If $H = 0$, then in the case of the majority of nematic liquid crystals (particularly for 5CB), we have $B > 0$ and a light-induced second-order phase transition takes place; if H is increased, then the sign of B can change to $B < 0$; then, a first-order phase transition takes place in a system and this transition exhibits a hysteresis. This explains the results shown in Fig. 13, However, the change in Φ^{nl} (and, therefore, in θ_m) at the reorientation threshold occurs quite smoothly in experiments. This departure from the predictions of the simple theory of Ref. 34 is clearly due to the need to allow for thermal fluctuations of the director (see Fig. 12) and also due to the finite dimensions of the laser beam.

The value $B = 0$ in the case when $C = 0$ [when the coefficient in front of θ_m^3 in Eq. (12) automatically vanishes] corresponds to a tricritical point of the system. A theoretical estimate of the tricritical parameters of 5CB ($B = 0$, if $H = H_c = 0.92H_0$ or $I = 1.78I_{th}^0$) gives values close to those found experimentally.

The possibility of hysteresis in the case of reorientation is not a specifically optical effect.[66] Figure 14 shows, by way of example, the results of calculations carried out for a nematic liquid crystal RO-TN-200 subjected to an external quasistatic electric field. An interesting feature of this case is the ability to change the nature of the phase transition by altering the field frequency f, which determines the sign of the anisotropy of the permittivity of the medium.[67]

5. REORIENTATION OF A NEMATIC LIQUID CRYSTAL INDUCED BY EXTERNAL FIELDS AS AN ANALOG OF A NONLINEAR OSCILLATOR

It is of interest to consider light-induced effects in nematic liquid crystals from the point of view of general systems exhibiting optical bistability because this makes it possible to reveal in a systematic manner the various regimes including the transition to dynamic chaos. It is useful to follow here an analogy with the problem of a nonlinear (anharmonic) oscillator.[55]

The description of optical bistability and instabilities within the framework of a model of an anharmonic oscillator (Duffing equation) applies to vibrations of microscopic systems (atoms, molecules) with a strong local nonlinearity, but even in the case of extended systems (when optical fields in a nonlinear medium interact or self-interact) the use of this model can be very fruitful. It is found that in many cases the equations describing such phenomena can be reduced to a form similar to the equation for a nonlinear oscillator.[18)] This makes it possible to carry out a general analysis of the potential regimes in nonlinear dynamics of these systems and identify the conditions for their observation. In particular, this approach has been found to be successful in the case of liquid crystals (particularly in the initial stages of the analysis) which can be subjected to external fields in various geometries. Use of this analogy makes it possible to determine more precisely the conditions for the appearance of various regimes. One can distinguish here two aspects.[55]

One can readily show that the usual equation for reorientation of the director of a nematic liquid crystal can be represented by a spatial analog of the anharmonic oscillator equation. A manifestation of the general property of nonisochronism of such an oscillator[8] in the case of a nematic liquid crystal is the field-dependence of the characteristic scale of a field-induced grating of the refractive index. A resonance of forced oscillations is ensured in the general case by multibeam geometry when the structure of strains created by optical fields acting as an external force becomes similar to that already induced in a nematic liquid crystal by a different field.

In fact, we can show that, for example, the equation for the threshold Fréedericksz transition in a static magnetic field $\mathbf{H}\|\mathbf{x}$, when a nematic liquid crystal with a homeotropic orientation (director $\mathbf{n}_0\|\mathbf{z}$) is subjected also to an optical field $\mathbf{E}$, considered in the geometry of threshold-free reorientation (angle of incidence α; $\widehat{\mathbf{E}\,\mathbf{n}_0} \neq 0°, 90°$) for reorientation angles $\theta \ll 1$, can be written as follows:

$$\frac{\partial^2\theta}{\partial z^2} + R'_0\theta + \beta'\theta^3 = pI_z, \tag{13}$$

where

$$R'_0 = \left(\frac{\pi}{d}\right)^2, \quad \beta' = -\frac{2}{3}\left(\frac{\pi}{d}\right)^2 \frac{H^2}{H_{th}^{02}},$$
$$p = -\varepsilon_a\varepsilon_\perp^{1/2}\,\mathrm{tg}\,\alpha\left[K_{33}\varepsilon_\parallel c\left(1 + \frac{\varepsilon_\perp}{\varepsilon_\parallel}\mathrm{tg}^2\alpha\right)^{1/2}\right]^{-1},$$

and I_z is the z component of the Poynting vector of light. Equation (13) is now a spatial analog of the equation for an anharmonic oscillator subjected to a periodic external force[8] [the right-hand side of Eq. (13) can be represented in the form $I_z = I_{z0}\sin R^{1/2}z$, where $R^{1/2}$ is the spatial frequency of the oscillations)]. This can be generalized allowing for damping or fluctuations. Then, in the former case the left-hand side of Eq. (13) is supplemented by a term containing the viscosity γ: $-\gamma K_{33}^{-1}\partial\theta/\partial t$; if for the sake of simplicity we assume that θ depends exponentially on time ($t \gtrsim 0$) in accordance with $\theta = \theta_0\exp(at)$, where θ_0 is an rms fluctuation and a is a parameter dependent on H and I_z, the quantity R'_0 is redefined. In the latter case the right-hand side of Eq. (30) is supplemented by a random "force" $f(z)$ which

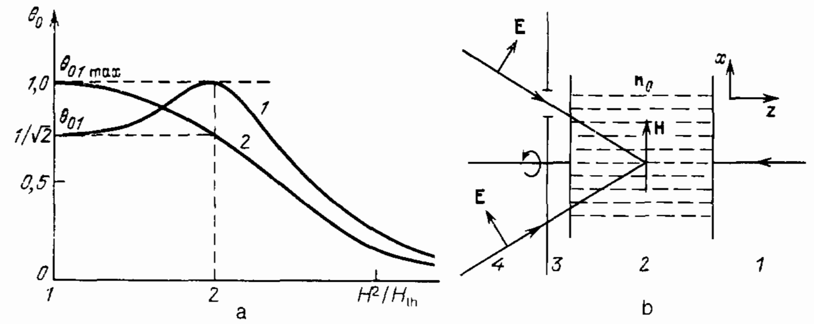

FIG. 15. a) Dependence of the amplitude of the angle of deviation θ_0 of the director on the applied magnetic field H, governing the natural frequency of oscillations of the system ω_0, obtained in Ref. 55 for two cases: 1) $\mathfrak{J}\neq 0$; 2) $\mathfrak{J}=0$; in the $\mathfrak{J}\neq 0$ case the values of θ_{01} and $\theta_{01\,max}$ correspond to $\omega_0=0$ $(H/H_{th}=1)$ and $\omega_0=\Omega[H/H_{th}=(H/H_{th})_{max}]$; the calculations were carried out for the following conditions: $\mathfrak{J}=3\times 10^{-5}$ g/cm, $d=10\,\mu$m, $\Omega=10^3$ Hz, $K=10^{-6}$ dyn, $\gamma=10^{-2}$ g·cm·sec^{-1}, $I/I_{th}=1$, $(H/H_{th})^2_{max}=2$. b) Geometry of the problem: 1) probe beam; 2) liquid crystal; 3) modulator capable of varying the intensity in accordance with the law $I=I_0\sin R^{1/2}t$; 4) pump.

is δ-correlated in space and we have $\langle f(z)\rangle=0$ and $\langle f(z)f(z')\rangle=2D\delta(z-z')$, where D is the diffusion coefficient of the orientation defined by $D\approx R_0^{\prime 2}K_{33}/\gamma$ (see Ref. 68).

However, direct utilization of Eq. (13) in the analysis of optical bistability of liquid crystals meets with considerable difficulties because this equation does not include a term containing the first derivative with respect to z and in the case of an anharmonic oscillator a similar term containing $\delta/\delta t$ (in the time dependence) and describing damping ensures a transition between two states of a bistable system.[8]

Moreover, an analogy with an anharmonic oscillator can be followed by conservation of the moment of inertia $\mathfrak{J}$ in the dynamics of nematic liquid crystals (see Ref. 69).

We shall consider the specific case when a homeotropically oriented nematic liquid crystal is subjected to a field of two crossing light waves [forming a field $\mathbf{E}=(E_{x,0},E_z)$] in the presence of a static magnetic field $\mathbf{H}=(H_x,0,0)$ (Fig. 15). The standard procedure for variation of the free energy of the system, consisting of the fields and the medium, subject to an allowance for the time derivatives of the angle of reorientation $\theta(z,t)$ of the director $\mathbf{n}=(\sin\theta,0,\cos\theta)$ gives the following equation[19] accurate to within terms which are cubic in θ:

$$\frac{d^2\theta_0}{dt^2}+R_0\theta_0+\beta\theta_0^3+\alpha_0\frac{d\theta_0}{dt}=B\cos\Omega t \qquad (14)$$

(we can show that if $\theta_0>0.1$, $H/H_{th}\sim 2$, and $I\gtrsim 1$ kW/cm^2, the terms proportional to $\sim\theta_0^2$ can be ignored). For simplicity, we shall make a number of assumptions (which are usual in the case of nematic liquid crystals), in particular, we shall ignore the anisotropy of the elastic properties of the liquid crystal (elastic constant K) and we shall consider only the first harmonic of the orientational distortion of the initial orientation angle $\theta=\theta_0\sin\pi z/d$, where $\theta_0=\theta_{01}e^{i\Omega t}$, and assume that the parameters are as follows: $\beta=-\chi_a H^2/4\mathfrak{J}$;

$$R_0=\left[\chi_a H^2-K\left(\frac{\pi}{d}\right)^2\right]\frac{1}{\mathfrak{J}}\equiv\chi_a H_{th}^2\frac{1}{\mathfrak{J}}\left(\frac{H^2}{H_{th}^2}-1\right)$$

determines the natural frequency ω_0 of oscillations in a system of this kind $(R_0\equiv\omega_0^2)$; H_{th} is the threshold value of the magnetic field for the Fréedericksz transition; χ_a is the magnetic susceptibility anisotropy; d is the thickness of the investigated nematic liquid crystal; $B=(\varepsilon_a/4\pi^2\mathfrak{J})(\mathscr{E}_x^*\mathscr{E}_z+\mathscr{E}_x\mathscr{E}_z^*)$; ε_a is the optical anisotropy of the permittivity; $|E_x|^2=|\mathscr{E}_x|^2\cos\Omega t$, $|E_z|^2=|\mathscr{E}_z|^2\cos\Omega t$;[20] $\theta_{01}=B\times[(\omega_0^2-\Omega^2)+i\gamma_0\Omega]^{-1}$; $\alpha_0=\gamma/\mathfrak{J}$; γ is the viscosity. For simplicity, we shall ignore the correction for the local field so that $|E|^2=(8\pi/cn)$, where I is the light intensity and n is the refractive index, and assume that there is no phase shift between $\mathscr{E}_x$ and $\mathscr{E}_z$, which then yields $B=(2\varepsilon_a I/\pi cn\mathfrak{J})$.

The nonlinear equation (14) with $\alpha_0\equiv 0$ is the Duffing equation.[8] If $\beta>0$ and $R_0>0$, all the solutions of Eq. (14) are periodic. The principal difference between these solutions and the motion described by the linear equation $(\beta\equiv 0)$ is that the frequency of oscillations described by Eq. (14) is determined by $R_0^{1/2}$ and depends on the amplitude θ_{01}, i.e., the nonisochronism effect is observed. If $\beta<0$ and $R_0>0$, then low energies W of the system correspond to motion which qualitatively resembles harmonic oscillations. An increase in W above a critical value W_c suppresses periodic motion which cannot exist in the range $W>W_c$. This critical value is given by $W_c=R_0^2 4^{-1}|\beta|^{-1}$ and it corresponds to $\theta_{01}=\pm(-R_0\beta^{-1})^{1/2}$. Infinite motion can also occur for $W<W_c$ provided the initial deviation is sufficiently large.

If $R_0<0$ and $\beta>0$, there are two stable equilibrium positions characterized by $\theta_{01}=\pm(-R_0\beta^{-1})^{1/2}$ and corresponding to two possible directions of the tilt of the director, as well as one unstable with $\theta_{01}=0$. In the last case the motion is only periodic.

Therefore, the condition $R_0=0$ determines the threshold value of H at which deformation of a nematic liquid crystal begins $(\theta\neq 0)$. Forced nonlinear oscillations described by Eq. (14) give rise to hysteresis of the dependence of the oscillation amplitude $|\theta_{01}|$ on the frequency $R^{1/2}\equiv\Omega$ of the driving force; the dependence $|\theta_{01}(R)|$ exhibits a resonance $(\alpha_0\ll R^{1/2}, R\sim R_0)$. If R has a constant value, hysteresis appears in the course of successive rise and fall of I and it is again a consequence of continuous deformation of the resonance curve. The condition for the existence of a resonance $\alpha_0\ll R^{1/2}$ in fact determines the range of values of the parameters of a nematic liquid crystal in which the oscillatory regime is observed.

We can now readily estimate the threshold optical intensity I_{OB} which in the geometry under discussion should correspond to optical bistability. We shall therefore seek the solution of Eq. (14) in the form $\theta_0=A(t)\sin[\Omega t+\varphi(t)]$; we can then readily derive reduced equations for $A(t)$ and $\varphi(t)$ by the Van der Pol method and then estimate I_{OB} using the expression (see Refs. 8 and 115)

$$I_{OB}=\frac{4\pi cn\gamma\Omega}{\varepsilon_a}\left(\frac{5\gamma\Omega}{6\chi_a H^2}\right)^{1/2}.$$

If we select the parameters (see below) to be $\mathfrak{J}=10^{-5}$ g/cm, $\gamma=10^{-2}$, $\Omega=10^3$ Hz or $\mathfrak{J}=10^{-8}$ g/cm, $\gamma=10^{-2}$, and $\Omega = 10^5$ Hz, we find that $I_{OB} \sim 50$ kW/cm^2 or $I_{OB} \sim 50$ MW/cm^2, respectively.

The most interesting is the case when hysteresis disappears at the reorientation threshold, because we can then speak of a field-induced first-order phase transition (see Ref. 34). The physical reason for optical bistability is then related to indeterminacy of the initial reorientation: both directions of deviation of the director are equivalent. When the threshold is approached from the high-field side, one of the possible states is already in existence in the system, so that the cases of rising and falling fields are not equivalent.

In other geometries we can show that Eq. (14) has to be replaced by an analog of the nonlinear Mathieu equation and the growth of parametric instabilities in such a case is well known (Ref. 8).[21)]

Equation (14) can also be generalized allowing for fluctuations, i.e., by considering the growth of thermal fluctuations of the director in the presence of an optical field; this can be done by introducing a random force $f(t)$ on the right-hand side of this equation. The solution of Eq. (14) with $f(t)$ in the Gaussian form and in the case of δ correlation in time is analyzed in Ref. 68. Its statistical behavior is governed by the sign of R_0; if $R_0 = 0$, there is a change from a regime with a maximum at $\theta = 0$ for a steady-state distribution of the probability $P(\theta)$ of the quantity θ in the case with maxima of $P(\theta)$ at $\theta = \pm(-R_0\beta^{-1})^{1/2}$ (for simplicity, we shall assume that $I = 0$). Then, fluctuations increase strongly for $R_0 = 0$ and, consequently, the scattering of light should also increase (Fig. 12). This gives rise to an additional energy loss channel which disappears when the reorientation threshold is exceeded. Therefore, hysteresis appears because of the scattering of light associated with reorientation in the threshold region as the field is increased and then reduced. This effect has been observed experimentally.[63] The presence of a scattering maximum in the critical region of the growth of transverse fluctuations of the director, at the threshold of the Fréedericksz transition, can naturally be predicted also by the ordinary methods of thermodynamics of liquid crystals,[24] but this maximum is of very general importance and it is characteristic specifically of first-order phase transitions.[15,70]

An analysis of optical bistability regimes in the case of nematic liquid crystals can also be carried out for other geometries and also allowing for an inhomogeneity of the field in the medium, relaxed boundary conditions (which determine the nature of phase transitions induced by the field), etc.

It is desirable to describe the reorientation effects in the form of Eqs. (13) and (14) of universal validity because it helps to detect the transition to dynamic chaos due to interaction of external fields with a liquid crystal (see Ref. 121). The use of a liquid crystal to identify the actual scenario of the transition to chaos is particularly attractive in experimental studies. The ranges of the parameters occurring in Eqs. (13) and (14) in which the behavior of a system is chaotic are identified in Ref. 26. Therefore, it is realistic to carry out a deliberate experimental search for these extremely important states of the system.

The main question which is encountered in the time dependences is the need to retain the term with $\Im \neq 0$ in Eq. (14). Usually this term is regarded as negligible, of the order of the moment of inertia of a single molecule. However, in the case of a nematic liquid crystal the orientational effects are governed by the collective reorientation phenomena in an ensemble of molecules affecting the director, so that we can assume that the macroscopic value of $\Im$ need not be small[22)] (see Ref. 69).

A simple estimate of the macroscopic moment of inertia can be obtained for the director of a nematic liquid crystal per unit volume ($\Im$) using the dimensions of that local region with a characteristic scale r_d which is associated with the director. In fact, by definition we have $\Im = m_d r_d^2/V_d = \rho r_d^2$, where ρ is the density of the investigated nematic liquid crystal ($\rho \approx 1$ g/cm^3).[23)] We can estimate r_d using dimensional analysis and we then readily obtain[96] the relationship $\gamma = r_d v \rho$, where γ is the viscosity and v is a parameter with the dimensions of velocity (see Ref. 69). In the case of typical nematic liquid crystals we have $\gamma \sim 0.1$–1 P; assuming that $v \sim 10^5$ cm/sec (velocity of sound),[24)] we find that $r_d = 10^{-6}$–10^{-5} cm so that $\Im \sim 10^{-12}$–10^{-10} g/cm. This value of r_d corresponds to the correlation coherence length of nematic liquid crystals.[24] If v is associated with the propagation of orientational waves (solitons), then r_d is equal to the thickness d of a nematic liquid crystal layer; for $r_d \sim d \sim 10\,\mu$m, we have $\Im \sim 10^{-6}$ g/cm. In this case a similar estimate of $\Im$ can be obtained from the relationship $v = (K/\Im)^{1/2}$, where K is the elastic coefficient.[69]

The problem can be solved by experiments. The equations of motion of a nematic liquid crystal in an external (magnetic) field derived allowing for the moment of inertia $\Im$ of the director have soliton solutions.[69] Experimental observation of such behavior makes it possible to estimate $\Im$. However, it is much simpler to find $\Im$ by direct experiments in which a nematic liquid crystal is in an oscillatory regime (see Fig. 15). In fact, if we write down Eq. (14) in the ap-

TABLE I. Reorientation angles θ_{01} and $\theta_{01\,max}$ (Fig. 15) and corresponding phase shifts Φ_1^{nl} and Φ_{max}^{nl} for different parameters of liquid crystals.

$\Im$, g/cm	γ, P	d, μm	Ω, Hz	H/H_{th}	I/I_{th} *)	θ_{01}	Φ_1^{nl}	$\theta_{01\,max}$	Φ_{max}^{nl}
10^{-6}	10^{-1}	100	10^5	10^2	10	10^{-4}	$7\cdot10^{-3}$	10^{-4}	$7\cdot10^{-3}$
10^{-5}	10^{-1}	100	10^4	10	10	$7\cdot10^{-4}$	$5\cdot10^{-2}$	10^{-3}	$7\cdot10^{-2}$
10^{-5}	10^{-2}	10	10^3	2	1	$1/\sqrt{2}$	5	1	7
10^{-5}	1	10	10^4	10^2	1	10^{-3}	$7\cdot10^{-3}$	10^{-3}	$7\cdot10^{-3}$
10^{-5}	1	100	10^4	10^4	10	10^{-1}	$7\cdot10^{-3}$	10^{-4}	$7\cdot10^{-3}$
10^{-3}	10^{-1}	100	10^3	10^4	10	10^{-3}	$7\cdot10^{-2}$	10^{-2}	$7\cdot10^{-1}$

*I_{th} is the threshold light intensity needed to induce the Fréedericksz transition in a homeotropic cell illuminated normally.

proximation which is linear in θ_0, then its solution allowing for the phase shift between the resultant oscillations and the driving force obtained for two cases ($\Im = 0$ and $\Im \neq 0$) yields the dependences shown in Fig. 15. The actual values of the parameters used in such calculations are given in the caption of Fig. 15.

Table I lists the values of the reorientation angles θ_{01} and $\theta_{01\max}$ and the corresponding phase shifts Φ_1^{nl} and $\Phi_{\max}^{nl}$ of a probe light beam used to detect reorientation of a nematic liquid crystal; these angles are given for characteristic points on the curves in Fig. 15 obtained for different values of the material parameters of a nematic liquid crystal and different experimental conditions. For example, we can readily demonstrate that the condition $(\theta_{01\max} - \theta_{01})/\theta_{01\max} \gtrsim 0.1$, which ensures a sufficient difference between the curves in Fig. 15 for experimental detection, leads to the requirement $\Omega \gtrsim 0.45\gamma/\Im$. In this case the difference between the values of H/H_{th} for characteristic points of the curve with $\Im \neq 0$ (curve 1 in Fig. 15) also exceeds 0.1, i.e., it should be easy to detect it experimentally.

The data of Table I show that in real cases it is possible to measure $\Im$ more readily for specially selected liquid crystals with exotic properties such as a low viscosity γ and high values of r_d, which govern $\Im$. However, if r_d is increased, then—in spite of the fact that both $\Im$ and γ both increase and make opposite contributions when differences between the two curves in Fig. 15 are observed—the rise in the former case is faster ($\Im \propto r_d^2$ and $\gamma \propto r_d$).[25] Therefore, an increase in the degree of collective interaction [26] improves the conditions for realization of oscillatory regimes in the dynamics of liquid crystals.

6. INSTABILITIES AND STOCHASTICITY IN EXPERIMENTS ON LIGHT-INDUCED REORIENTATION OF THE DIRECTOR OF LIQUID CRYSTALS UNDER CONDITIONS OF EXCITATION OF VOLUME GRATINGS

6.1. Effects of a laser field on a liquid crystal

A laser field destabilizes the initial equilibrium state of a medium in which reorientation effects begin to appear and these effects govern field-induced structural phase transitions.[24] In the case of a propagating wave this results in creation of volume gratings, i.e., a distributed feedback is established in the system.

The processes of self-modulation of light which then develop are governed by the mutual influence of normal waves in a medium.[74,75] In the case of nematic liquid crystals a characteristic spatial scale in which energy transfer occurs between two waves with orthogonal polarizations is governed by the quantity $1/\nu A_i^2$ (Ref. 54), i.e., it is governed by beats (with the characteristic parameter q_z) between two oscillatory components of the field (here, A_i^2 is the intensity of each of the components of the field identified by $i = 1$ or 2, $\nu = q_z^{-1}\varepsilon_a \cdot 16^{-1}\pi^{-1}K_{22}^{-1}$, q_z is the z component of the vector $\mathbf{q} = \mathbf{k}_e - \mathbf{k}_o$, and $\mathbf{k}_e$ and $\mathbf{k}_o$ are the wave vectors for waves with the extraordinary and ordinary polarizations, respectively). In the case of cholesteric liquid crystals such a periodic redistribution of energy between waves gives rise to so-called pendulum solutions.[76] The presence of two orthogonal polarizations inside a medium and their interaction results in time instabilities of the transmitted light. The appearance of oscillations as a result of optical excitation of nonadiabatic deformations in nematic liquid crystals was first discovered in a series of experiments reported in Ref. 38. Different oscillation regimes were observed: damped, exactly periodic, and stochastic. There are many factors which can give rise to waves of different polarizations in liquid crystals. One of them is the appearance of a second polarization due to light-induced reorientation processes (Ref. 54).[27] Another possibility is that waves with the two polarizations can travel in a liquid crystal even in the linear case. This is either due to the ellipticity of the polarization of light incident on an initially homogeneously oriented liquid crystal or, in the case of linear polarization of light, it is due to an inhomogeneity of the liquid crystal structure. It is determined by the asymmetry of the boundary conditions (samples with hybrid orientation[80]) and by the specific "twistedness" of the structure (cholesteric liquid crystals). Another possibility is the use of multibeam systems when a liquid crystal is subjected to several waves with different polarizations. The nonlinear interaction of light with a medium (characterized by a threshold) ensures formation of complex gratings in all these cases as a result of the combined effects of different components of the field. Finally, the scattering of light in a liquid crystal in the orthogonal polarization can also give rise to gratings if the intensity of the incident light is sufficiently high.

Various combinations are also possible when a liquid crystal is subjected not only to an optical field, but to other quasistatic (in particular, magnetic) or acoustic fields which themselves create inhomogeneities in a medium and can excite various instabilities.[85,99]

We shall now analyze some of the instabilities which appear because of a nonlinear optical interaction of light with a liquid crystal, as manifested in the intensities and polarizations of the transmitted light. It is convenient to divide such instabilities into two groups: regenerative pulsations in time[4] and Ikeda instabilities.[19] The Ikeda instabilities describe the transition to optical chaos and identification of such oscillatory time dependences is important in studies of the stochasticity of wave nonlinear-optical interactions in inhomogeneous anisotropic media.

6.2. Experiments

Oscillations in time had been observed experimentally[111,116] in the course of propagation of optical waves with orthogonal polarizations in nematic liquid crystals. Various forms of dynamic self-diffraction of light[75] in anisotropic media[111] had been observed: (a) due to excitation of nonadiabatic deformations in the geometry of threshold reorientation of nematic liquid crystals in the case of oblique incidence of light on a sample[54]; (b) due to interaction of optical radiation with an inhomogeneously (in a hybrid manner) oriented nematic liquid crystal[80]; due to normal incidence on a nematic liquid crystal of (c) two coherent opposite waves with noncoincident directions of linear polarizations of light and (d) of elliptically polarized light. Oscillations appear in all these cases because of the exchange of energy between different components of the polarization of an optical field as a result of nonlinearity of the medium.

Homeotropically oriented samples of 5CB nematic liquid crystals were used in these experiments. A laser-induced reorientation of a liquid crystal (Ar^+ laser, $\lambda = 0.51\ \mu m$,

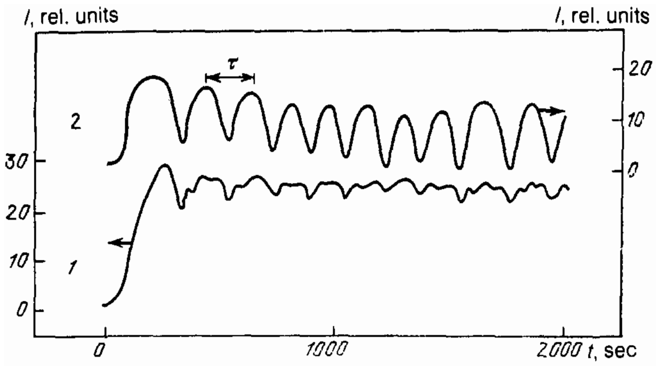

FIG. 16. Typical oscillations in time in the case of threshold reorientation of a homeotropic nematic liquid crystal in the field of an obliquely incident ordinary wave, obtained for the pump beam (1) and for the probe radiation (2).[116] The excess above the threshold was $I/I_{th} \approx 1.5$; the oscillation period was $\tau \approx 170$ sec. Parameters: nematic liquid crystal 5CB; $d = 130\ \mu m$; $T = 24.8\ ^\circ C$. The oscillations appeared more clearly in the case of the probe beam (probed region of size $10\ \mu m$, diameter of the main beam $600\ \mu m$). When I/I_{th} was increased, the oscillation pattern became more complex and irregular processes appeared at $I/I_{th} \approx 3$.

beam diameter $600\ \mu m$, power up to 3 W) was deduced from a characteristic aberration pattern[65] and also using a weak He-Ne laser beam acting as a probe ($\lambda = 0.63\ \mu m$, effective probed region of $\sim 10\ \mu m$ size), which made it possible to investigate a local region of a nematic liquid crystal (at the center of the Ar^+ laser beam) and not the overall pattern, so that the oscillations were observed more clearly (Fig. 16).

Figure 17 shows the results in the form of the dependences of the period τ of the oscillations on the intensity of the incident light I obtained in the geometries (a) and (b). In the former case (Fig. 17a) it was found—in contrast to Ref. 38—that an increase in I increased τ; the oscillations were not observed at high values of I and the reorientation pattern was stable; a further increase in I induced irregular processes. In the latter case (Fig. 17c) the period τ fell on increase in I; saturation occurred in the range $I \gtrsim$ kW/cm^2.

In the geometry (c) these oscillations appeared in every beam and were in antiphase (Fig. 18). For the same beam intensities these oscillations were damped out over approximately ten periods; the maximum duration in time and the amplitude corresponded to the case when the polarizations of the waves $\mathbf{E}_1$ and $\mathbf{E}_2$ were orthogonal; when the angle between $\mathbf{E}_1$ and $\mathbf{E}_2$ was reduced, the oscillations were manifested less strongly and in the $\mathbf{E}_1 \| \mathbf{E}_2$ case the reorientation pattern became steady. Different spontaneous oscillation regimes were observed as a result of a relative change in the intensities of the components of the field and because of a change in the angle $(\widehat{\mathbf{E}_1 \mathbf{E}_2})$.

In the case (d) the threshold intensity I_{th} for the reorientation increased on transition from linear polarization (along x, component E_x) of the incident light to elliptic polarization, when the second component of the field E_y appeared (Fig. 19). On increase in E_y (when the reorientation due to the action of E_x was retained) it was found that beginning from a certain value $E_y > E_{y0}$ once again undamped oscillations appeared in time (see Ref. 77). In the case when $E_y \sim E_x$ the oscillations appeared when the intensity of the component E_y was of the order of I_{th} and they were damped out in several periods. A steady-state threshold reorientation pattern reappeared in the range $E_y \gg E_x$.

Oscillations of the intensity and polarization of the transmitted light in the field of a circularly polarized incident wave were reported in Ref. 119.

In the case of cholesteric liquid crystals we could expect appearance of oscillations in time of a different type, which were due to the Bragg conditions of selective reflection of light dependent on I (Ref. 59). In the experiments this case could be observed conveniently utilizing the mechanism of laser heating of cholesteric liquid crystals. Typical oscillations in time reported in Ref. 55 for this case are plotted in Fig. 20 for a mixture of cholesteric liquid crystals which made it possible to use the field of cw He-Ne laser radiation ($\lambda = 0.63\ \mu m$) interacting with a cell $10\ \mu m$ thick when the intensity was $I \sim 40$ W/cm^2.

Instabilities and oscillations were observed also as a result of a light-induced Fréedericksz transition in cholesteric liquid crystals with the homeotropic orientation at the walls, which was homogeneous and stable for $d < d_c = K_{33}p/2K_{22}$, where p is the pitch of the helix of a free cholesteric liquid crystal.[82] Oscillations in time appeared also in the case of oblique incidence (at an angle α) of Ar^+ laser radiation of either the ordinary or extraordinary polarization (Fig. 21); the nature of the oscillations was governed by the angle α: for $\alpha = 45^\circ$ the oscillations were undamped.[116]

6.3. Discussion

We shall now consider the case when light propagates in a spatially inhomogeneous modulated medium. The parameter which changes under the influence of light is the angle of deviation φ of the director.[28)]

The calculation method was based on the geometric-

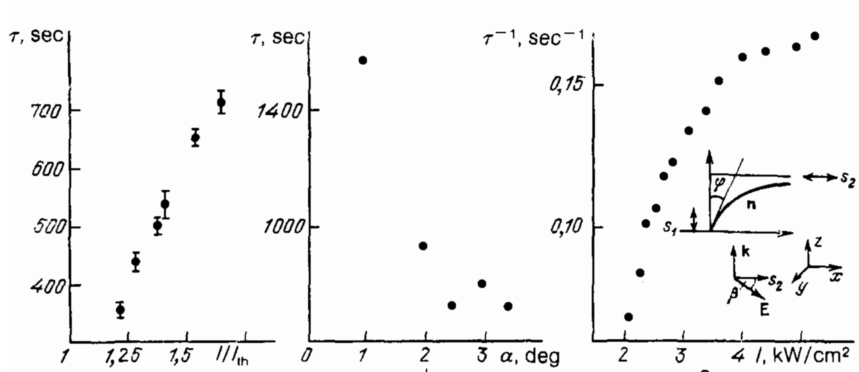

FIG. 17. a), b) Dependences of the oscillation period τ on I for a fixed value of $\alpha = 2.5^\circ$ (a) and on α for a fixed value of $I/I_{th} \approx 1.6$ (b). The experimental parameters were the same as in Fig. 16 (Ref. 116). c) Period of the time oscillations τ plotted as a function of I for $d = 50\ \mu m$ and $\beta = 90^\circ$ (hybrid cell)[116]; the inset shows the experimental geometry.

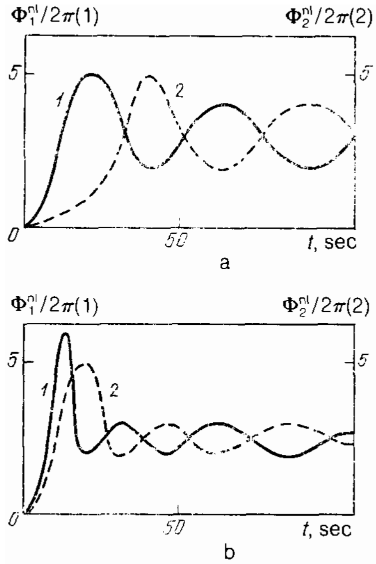

FIG. 18. Oscillations observed when a nematic liquid crystal was subjected to two opposite coherent laser beams with linear polarizations $\mathbf{E}_1$ and $\mathbf{E}_2$ which did not coincide in direction.[116] The intensities of the two beams were the same: $I_1 = I_2 = 230$ W/cm^2. The thickness of the nematic liquid crystal layer was $d = 200\,\mu$m. The angle between $\mathbf{E}_1$ and $\mathbf{E}_2$ was 90° (a) or 80° (b).

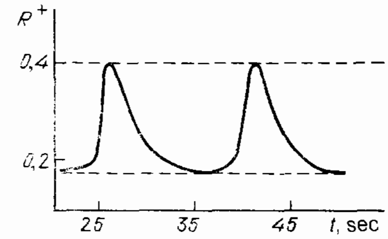

FIG. 20. Pulsations of light reflected by a cholesteric liquid crystal observed initially after an "instantaneous" increase in I (Ref. 59); $R^+ \equiv I_{\text{refl}} I_0^{-1}$.

optics approximation,[74] i.e., a local medium was regarded as uniaxial and the field inside it had components with the ordinary (o) and extraordinary (e) polarizations. The details of the calculations carried out for the case of nematic liquid crystals can be found in Ref. 111.

The physics of the observed phenomena is as follows. When two waves with orthogonal polarizations are acting inside the medium, the reorientation of the director occurs in two planes.[12] In the simple case of the equality of the elastic coefficients K of a nematic liquid crystal the equations of motion of the director $\mathbf{n} = (\sin\psi\cos\varphi, \sin\psi\sin\varphi, \cos\psi)$, where $\varphi = \varphi(z,t)$ and $\psi = \psi(z,t)$ are the reorientation angles, can be written in the following clear form

$$K\sin^2\psi\,\frac{\partial^2\varphi}{\partial z^2} = -Me_z + \gamma\sin^2\psi\,\frac{\partial\varphi}{\partial t},$$

$$K\left[-\sin\psi\cdot\cos\psi\left(\frac{\partial\varphi}{\partial z}\right)^2 + \frac{\partial^2\psi}{\partial z^2}\right]$$

$$= -M(e_z\cos\varphi - e_x\sin\varphi) + \gamma\,\frac{\partial\psi}{\partial t}, \qquad (15)$$

where

$$M = \frac{\varepsilon_a}{16\pi}\{(\mathbf{nE})[\mathbf{nE}^*] + (\mathbf{nE}^*)[\mathbf{nE}]\},$$

γ is the viscosity, and $e_{x,y,z}$ are unit vectors along the axes indicated in the subscript. The system of equations (15) represents equality of the moments of rotation of the director because of the action of elastic forces (on the left hand side) and of an optical field (first terms on the right hand side). In the latter case the first equation of the system (15) maintains the moment of rotation for a fixed value of φ, whereas the second equation contains the corresponding moment for a fixed value of ψ. The second terms govern the dynamics of the process.

In the present case, because of the anisotropy of the medium, there is a change in the polarization of the transmitted light and, in accordance with Eq. (15), the reorientation of a nematic liquid crystal is inhomogeneous along the z axis: a volume grating of the refractive index is induced. Its

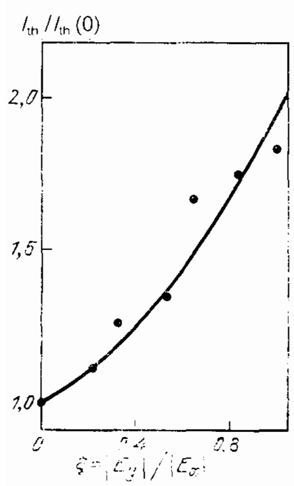

FIG. 19. Dependence of I_{th} on the ellipticity $\xi = |E_y|/|E_x|$ of the polarization of light incident normally on a cell containing 5 CB (Ref. 116). The thickness of the nematic liquid crystal layer was 200 μm. Experimental points were taken from Ref. 116 and the curve was calculated using the formula $I_{\text{th}}(\xi)/I_{\text{th}}(0) = 1/[1 + (1 - \xi^2)^{1/2}]$ based on Ref. 79.

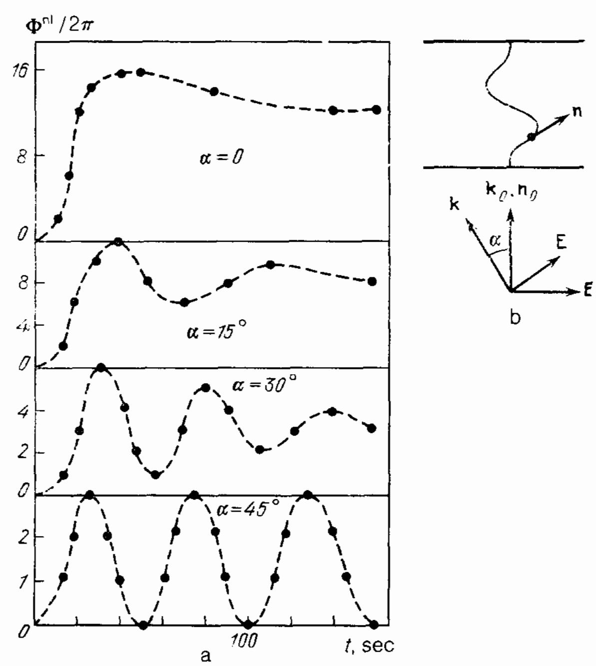

FIG. 21. a) Oscillations in the field of an obliquely incident extraordinary wave of $I = 2$ kW/cm^2 intensity observed for different angles α of incidence of this wave on a homeotropically oriented cholesteric liquid crystal (5 CB, $d \approx 60\,\mu$m) with a chiral admixture of 0.017% by weight, which ensured that in the course of reorientation the director was tilted out of the $(\mathbf{E}, \mathbf{k})$ plane and waves with orthogonal polarizations propagated inside the medium. b) Experimental geometry.

distinguishing feature is a characteristic scale (or "period" representing the separation between zeros of the function), which varies along the z axis and depends not only on the anisotropy of the medium (q_z), but also on the intensity of light I inside a nematic liquid crystal. When the angle α is varied, a complex transformation of the grating takes place. Firstly, there is a change in the scale, whereas the number of "periods" which can be fitted in the thickness of the sample remains constant. Secondly, at some values α_i there is a change in the number of these periods. The light intensity necessary to form a modulated structure in a nematic liquid crystal is governed by the scale of the induced distortions (gratings with distortions varying smoothly along z are formed more easily than those with rapid variation of the distortions) and by the total number of such distortions in the thickness of a sample (i.e., the energy integrated over the thickness of a nematic crystal is important). Therefore, in the former case we have the dependence $I_{th}(\alpha)$, whereas in the latter case there is a discontinuity of I_{th} at α_i, which determines a singular point of the system.

Figure 22a shows field-induced gratings expected for different values of α.

In the case of excitation of nonadiabatic deformations the angular dependence of the threshold intensity $I_{th}(\alpha)$ readily shows that hysteretic characteristics should be observed when α is gradually increased and then reduced. The number of upward and downward jumps is not the same (Fig. 22b).[37] It is important to note that at high values of α, which require large values of I, different branches corresponding to successive jumps of I_{th} and, which is particularly important, the corresponding values of the angle α_i become more similar, so that in practice a system may be in various states and this corresponds to the transition to chaos [on reduction in α the system follows one (the important point is which) of the branches in Fig. 22b]. Such states can be observed in experiments if in addition to an optical field (**E**) there is also a magnetic field **H**‖**E**; even low intensities $I \sim I_{th}$ ($\alpha = 0$) are then sufficient and jumps appear when H is varied.[111]

In the case of an optical field inside such an inhomogeneously anisotropic medium we can write down the equations for the ordinary (A) and extraordinary (B) components of the polarization; moreover, this can be done in a general geometry when both forward (A_+, B_+) and backward (A_-, B_-) waves interact in a medium. These equations describe the time dependences of the amplitudes and polarizations of the transmitted waves.

We can readily show that, for example, in the case of circularly polarized waves the polarization oscillates in time immediately if we make the simplifying assumption that $\partial\varphi/\partial z = \text{const}$ (and it follows hence that in the limit $t \to \infty$, we have $\partial\psi/\partial t = 0$ and $\partial\varphi/\partial t = \text{const}'$)[29] (see Ref. 119). On the other hand, oscillations of the intensity of an optical wave (producing a ring pattern of the transmitted light) require that the problem be solved without this assumption, but this meets with serious difficulties even when the solution is obtained by numerical analysis (the transfer of energy is then related to the ellipticity of the polarization of light). An even more difficult task is to provide a qualitative explanation of the oscillations of the intensity with time.

In fact, since I_{th} exists only for the ordinary wave, the transfer of energy from this wave to the extraordinary component reduces the intensity for the former so that the threshold can no longer be exceeded, i.e., the system returns

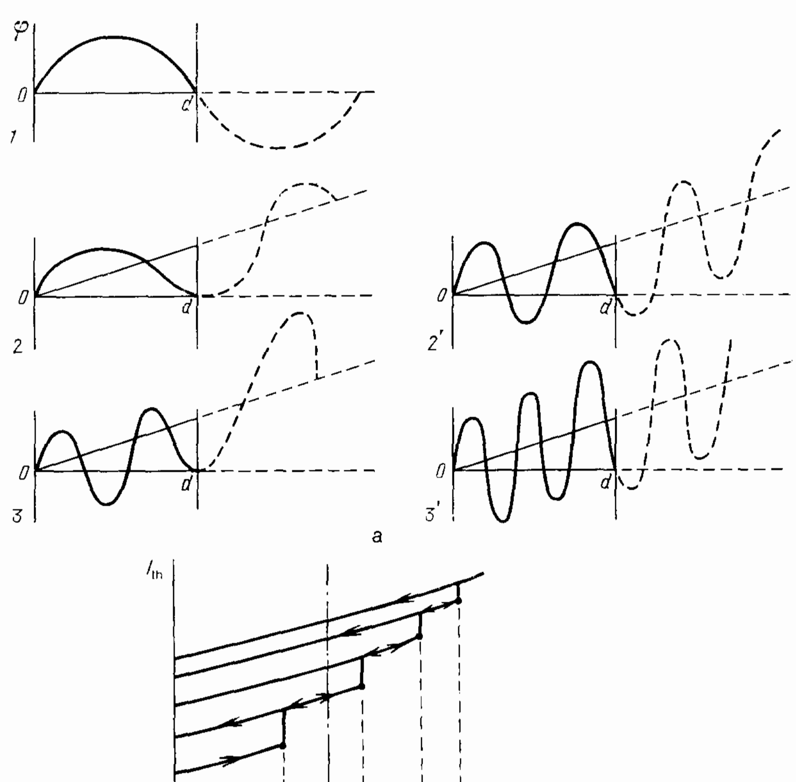

FIG. 22. a) Structure of the distortions which appear in a nematic liquid crystal under the influence of an optical field (threshold reorientation of a homeotropic sample in the case of oblique incidence of an ordinary wave) plotted for different angles of incidence of light α: 1) $\alpha = 0$, $\varphi \propto \sin(\pi z/d)$; 2) $\alpha_1 \approx 5.7°$ (first jump, I_{th}), $\varphi \propto \sin[(3\pi z/2d) + (z/d)]$; 3) $\alpha_2 \approx 7.5°$ (second jump), $\varphi \propto \sin[(5\pi z/2d) + (z/d)]$; here, φ is the reorientation angle. The dependences 2, 3, and 2', 3' correspond to modulated structures for, respectively, the lower and upper branches at jumps of I_{th}. The calculations were carried out for MBBA of thickness $d = 100\,\mu m$. The formula for the reorientation angle was

$$\varphi(z,\ t) \propto ((\chi^2 - q_z^2)\,\chi^{-1} \sin \chi z + q_z^2 z)\, e^{\nu t/\nu},$$

where $\chi = (Q/K)^{1/2}$, $Q = (\varepsilon_a/8\pi)(k_0/k_e)|A_{lim}|^2 + Kq_z^2$, where A_{lim} is the limiting value of the field amplitude and K is the elastic coefficient. b) Hysteresis of $I_{th}(\alpha)$. The chain vertical line identifies possible states of the system which it can assume for a given value of α when the light intensity is varied.

to its initial state. (This energy exchange occurs both during transient regimes and as a result of a grating which is shifted, relative to the optical field, as a consequence of the dependence of the period on I—see Ref. 75.) However, the extraordinary component of an optical wave inside a medium then disappears and the intensity of the ordinary component is again sufficient to excite reorientation. It is important to note that this mechanism operates only near the reorientation threshold when the transfer of energy to the extraordinary component is a fairly weak effect ($\sim\psi$), but it still has a strong influence on the reorientation process. This is confirmed by the experimental results in which oscillations of the values of I are observed near I_{th}.[30)] It should be noted that other possible explanations do not satisfy this experimental observation.[38] An increase in the oscillation period τ on increase in I (Fig. 17a) can also be understood on this basis: the higher the value of I (for a fixed angle α), the less energy is transferred to the extraordinary component and if $I \gg I_{th}$ the reorientation is a steady-state effect so that there are no oscillations. The reduction in τ on increase in α (for a fixed value of I) is determined (Fig. 17b) by an increase in $I_{th}(\alpha)$ in the selected experimental geometry and, consequently, there is a relative increase in the energy transfer to the extraordinary component in the course of reorientation. The very fact of the appearance of oscillations in time, which are in antiphase for two components of the field in an experiment with two opposite waves (see Fig. 18), confirms the correctness of the hypothesis of an oscillation pattern based on energy exchange between waves characterized by different polarizations.

If the appearance of the second polarization component inside the investigated medium is not due to the appearance of deformations, but is governed by the polarization of the incident light (elliptically or circularly polarized radiation) or by an initial inhomogeneity of the medium (hybrid cells, cholesteric liquid crystals),[31)] the competition between these polarization components because of the nonlinear interaction in the medium also gives rise to oscillations of similar type (Figs. 20 and 21).[32)] An increase in the intensity I of the incident light then reduces the oscillation period (see, for example, Fig. 7c) and each of the components has a sufficient intensity to induce distortions in the medium, whereas an increase in I makes the energy exchange more effective. If these components create distortions of the medium characterized by different relaxation times (due to different intensities of the components or due to induced gratings with different periods, which is true of adiabatic and nonadiabatic deformations of a medium in orthogonal planes), regenerative pulsations may appear (Refs. 4 and 43).[33)] Oscillations in the case of cholesteric liquid crystals (Fig. 20) depend on the initial state of the system, i.e., they depend on the point on a Bragg reflection curve at which the process begins when a laser is switched on; in particular, under certain conditions there may be oscillations which do not decay with time (because of the difference between the times taken to establish the temperature of a sample and the pitch of the helix of a liquid crystal).

It should be stressed that in the case of competing interactions of two fields with a liquid crystal, the resultant distorted structure is not simply a sum of two deformations: even in the case of the threshold Fréedericksz transition in a magnetic field $\mathbf{H}$ there may be no reorientation in the range $H > H_{th}$ in the presence of an optical field $\mathbf{E}\|\mathbf{H}$ in the geometry when this field induces (in a threshold manner) nonadiabatic deformations.[111] The opposite effect is also possible. In a sufficiently strong optical field the competing (in respect of the geometry see, for example, Fig. 15) polarization components ($\mathbf{E}_1 \perp \mathbf{E}_2$) can increase the total reorientation: in this case inclusion of terms which are higher in respect of the reorientation angle φ (it is sufficient to retain terms proportional to φ) gives rise to contributions to φ which have the same signs for both components (this plays the main role when $I_1 \approx I_2$ and depends on $\tan^2\alpha$).[116] Another possibility is light-induced reorientation of the director of a nematic liquid crystal occurring in a perpendicular plane ($\mathbf{E}_1$, $\mathbf{E}_2$). We shall conclude by noting the influence of the bounded nature of a laser beam (radius r_0, curvature of the phase front $1/R$). The phase shift $\Phi(r)$, where r is the transverse coordinate, for a transmitted wave is (see Ref. 74)

$$\Phi(r) = k_0 \left[r^2 (2R)^{-1} + \int_0^d \delta n(r, z)\, dz \right],$$

where $k_0 = 2\pi/\lambda$; $\delta n > 0$ is a nonlinear correction to the refractive index; then, depending on whether a converging ($R < 0$) or a diverging ($R > 0$) beam is used in the experiments, the resultant ring structure will be quite different.[34)] If in the course of reorientation there is a reversal of the sign of R (for example, along z at high values of d; for typical nematic liquid crystals when $I \sim 1$ kW/cm^2, $r_0 \sim 50\,\mu$m, and $\lambda \sim 0.5\,\mu$m, the self-focusing length is estimated to be 500 μm), then oscillations are possible. A reduction in r_0 as a result of self-focusing may also create oscillations because of the dependence $I_{th}(r_0)$ (Ref. 38).[35)] Clearly, these are the effects which are manifested experimentally[38] because in their case an increase in I should reduce τ (self-focusing of light is a faster process).

We shall end with the comment that oscillatory light-induced effects in liquid crystals may be due to fairly general factors.[92,93] In fact, in the case of slowly varying media[74] the correction to the quasisteady values of the parameters of the medium (and particularly to the permittivity, which is governed by the nature of light-induced distortions) has an imaginary (anti-Hermitian) part which differs from zero even in the absence of absorption (see, for example, Ref. 74).[36)] This part describes an additional phase shift between the vectors of the electric induction and intensity when this shift is induced by reorientation of the investigated medium.

7. CONCLUSIONS

The results reviewed above demonstrate that liquid crystals are unique media in which it is possible to observe strong nonlinear interactions of optical waves, optical bistability and multistability, instabilities, and chaos.

The principal reason for the occurrence of these processes is that in the case of liquid crystals we are dealing with very strong nonlinearities which are responsible for field-induced structural phase transitions so that the nature of the resultant instability originates from the physical aspects of the phenomena. This is the reason why it has been possible to observe true intrinsic (resonatorless) optical bistability resulting from real phase transitions that occur in a nonlinear medium subjected to an optical field. A feedback necessary for the appearance of optical bistability is in this case determined by the nature of the self-interaction effects of light in a liquid crystal, namely by the nonlocality of the nonlinear response of a medium in which a propagating wave experiences the action of the medium at some point and is itself capable of influencing the state of the medium at this point.

In the case of conventional resonator systems this feedback is ensured by the reflection of a wave from a mirror. In systems with distributed feedback the appearance of a backward wave is a consequence of the Bragg reflection by a grating. In the case of nematic liquid crystals an important feature is that such feedback may appear also in the absence of a backward wave because of, for example, elastic properties of a medium which give rise to a nonlocality of the interaction of light with matter (when the equations contain spatial derivatives of $\mathbf{n}$). In fact, light-induced reorientation of nematic liquid crystals at each subsequent point during propagation of a wave influences, because of the elasticity of the medium, the state of the medium also at the preceding point, which in turn alters the properties of the transmitted wave.[37)] Therefore, in particular, optical bistability becomes possible also in the case of nonlinear scattering of light under conditions ensuring optical orientation of molecules.[52,55] The time parameter, similar to the round-trip time of a resonator, is the relaxation time describing establishment of deformation in a medium; it governs instabilities in the system (for example, the Ikeda instabilities[19]).

In the case of propagating waves the dominant feature is the occurrence of nonlinear interactions of several waves with different polarizations. The random nature of the process (due to fluctuations of the director) plays a decisive role in the appearance of a new state of a liquid crystal under the influence of an external field. A nonlinear interaction of waves in liquid crystals is characterized by a small number of degrees of freedom; these degrees are two components of the polarization of an optical wave and the energy exchange between these is essentially governed by a four-wave parametric process (in the steady-state and the nonsteady cases). A description of these phenomena can be provided using the "language" of stimulated scattering typical of nonlinear optics when effective energy exchange occurs for waves with different frequencies (a moving displacement relative to the grating field forms in a medium); this approach is developed in Ref. 120 for a medium with the Kerr nonlinearity[38)] and an analysis of the appearance of various unstable (chaotic) regimes is made there.

The investigations carried out so far have already yielded some important physical results and will help in further growth of this promising branch of nonlinear dynamics of strongly excited systems.

We should mention first of all the extensive opportunities that liquid crystals provide for detailed investigations of time dependences of strongly nonlinear optical effects, including a quantitative picture of the transition to optical turbulence (chaos). The experimental aspect of such investigations is one of the main topics in the current stage of investigation of this universal state of strongly nonlinear systems (see Refs. 94 and 100, as well as Ref. 122 where recent experiments were reported).

Liquid crystals provide in fact the means for systematic investigations of the manifestations of multistability and chaos in real two- and three-dimensional fields: the first steps have already been made and some promising results, particularly those for hybrid systems have been obtained.[117] It is necessary to consider these effects also in purely optical cases which will undoubtedly be of major practical importance to the development of a new architecture of computers based on nonlinear images and not on standard binary logic.[106]

Scenarios of the transition to chaos are many and in the case of orientational effects in liquid crystals we can use the method often adopted for liquids[15]: a steady (spatially homogeneous) state passes via a nonequilibrium phase transition to another steady (but spatially inhomogeneous) state, which in turn changes to an oscillatory state[39)] so that a limit cycle is established. Then, instead of one there should be two fundamental oscillation frequencies and transitions of this type may continue indefinitely. This behavior is naturally not universal. In particular, as shown in Sec. 5, orientational effects in external fields can be described by analogy with an anharmonic oscillator.

Specific studies of these effects, identification of the conditions under which they appear, and the requirements in respect of the parameters of liquid crystals which ensure that these regimes can be realized experimentally is a necessary stage in the current status of research. Hydrodynamic effects in liquid crystals are very interesting. In particular, Kapustin–Williams domains which appear in nematic liquid crystals can be considered from the point of view of formation of periodic structures on transition to dynamic chaos.[88] The appearance of higher (and fractional, subharmonic) orders of reflection of light by cholesteric liquid crystals (the case of normal incidence of light is considered in Ref. 87) can also be analyzed on the basis of the transition to chaotic behavior by period doubling. The same approach can be used to consider multiple diffraction of light by light-induced gratings in nematic liquid crystals encountered in conventional systems for dynamic self-diffraction[12] (see also Ref. 81).

It should be stressed that such a great variety of instabilities of this type is not accidental and, therefore, it would be desirable to attempt to describe all these effects in liquid crystals from a unified standpoint using fairly general nonlinear equations (see Ref. 100).[40)] A promising approach is an analysis of these phenomena on the basis of multicomponent phase transitions. A consistent description of these and other effects should be statistical and should allow for fluctuations[94] by using, for example, the Fokker–Planck equations (see, for example, in particular Ref. 95).[41)]

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[1)]Distributed-feedback systems are now increasing in importance (in these systems optical waves interact continuously in many sections of a nonlinear medium).[31,106]

[2)]It should be pointed out that a hysteretic nature of the dependence of I_{out} on I_{in} does not necessarily mean that a system exhibits optical bistability; this applies in particular to pulsed systems, and not only to nonlinear ones but even to linear ones with dispersion.[7]

[3)]The possibility of hysteresis during lasing is the feature which allows us to introduce the concept of a first-order phase transition.[102] These topics are considered in Ref. 103 for the case of spatial fluctuations of radiation at the lasing threshold.

4)The speed of optical logic elements is now already superior to any other elements and the shortest response time is 10^{-14} sec (Ref. 1). Pumping can be conveniently provided by frequency-tunable semiconductor laser diodes (considerable progress has been made recently[14] in the manufacture of these diodes).

5)The switching rates which are achieved here are usually low.[12]

6)In particular, the operation of a liquid-crystal optical transistor was demonstrated in Ref. 78.

7)In this calculation the validity of Eq. (1) is limited to low light intensities (see Ref. 98), when the principle of superposition can still be used.

8)Derivation of the necessary conditions for optical bistability in a more general case, when an allowance is made for the nonlinearity of the system manifested not only by its phase but also by its amplitude parameters, can be found in Ref. 105.

9)The operation of such a liquid-crystal device was recently demonstrated in Ref. 118 (see also Ref. 122).

10)We can assume that, for example, the first of these mechanisms (molecular reorientation) is strong but slow ($\tau_\theta \sim$ sec), whereas the second (laser heating) is weak but fast ($\tau_T \sim 0.01$ sec), which is usually true of nematic liquid crystals.[43]

11)The duration of a "seed" pulse should be less than its travel time in the resonator; a pulse has a pedestal (it satisfies the steady-state condition), so that the system is in the region of hysteresis (in the simplest case it is sufficient to consider a nonlinear medium with an instantaneous response).

12)Multistability in this case is ensured by generalization to the case of an oscillator with several natural oscillation frequencies.

13)In particular, these may be four-wave interactions,[28] stimulated scattering,[16,27] etc.

14)There may be some regimes in which the upper state is not steady in the system under consideration and in this case oscillations may appear[19] (see Fig. 5).

15)Sometimes optical bistability is defined only for this case,[1] but this is naturally a question of definition.

16)This was observed experimentally for nematic liquid crystals using light scattering[51] and was studied separately on a different occasion.[52]

17)It is interesting that the transmission of a system can in this case be described by Eq. (1).

18)Aperiodic damping of orientational deformations is usually assumed for liquid crystals, but at least during the initial stage of the reorientation process one can speak of oscillations of the director. This approach to the analysis of the dynamics of reorientation of liquid crystals in the presence of dissipation is developed in Ref. 121 allowing for the reverse fluxes.

19)It is interesting to note that in this geometry weak optical fields exert a competing influence on the reorientation of a nematic liquid crystal; in strong fields such interactions are summed. (Experiments were reported in Ref. 116.)

20)In the experiments it is simplest to generate step-like light pulses of opposite polarities; calculations can easily be extended to this case.

21)An attractive approach involves description of the orientational effects in liquid crystals on the basis of a model of coupled oscillators[101] (with two field components inside the liquid crystal).

22)For example, in the case of an orientational nonlinearity its value for the mesophase is many orders of magnitude greater than the nonlinearity of an isotropic solution of the same molecules and this is precisely due to the collective nature of the interaction.[12]

23)In the case of the molecular moment of inertia ($\Im_M$), we find that $\Im_M = m_M h^2/V_M \sim 10^{-14}$ g/cm (we shall assume that molecules are cylinders of height $h = 2\times10^{-7}$ cm and with the base radius $r_M = 5\times10^{-8}$ cm; $m_M \sim 4\times10^{-22}$ g).

24)The acoustic wavelength λ_{ac} which must be allowed for in the case of waves propagating in a medium with the size of inhomogeneous regions l_i, is determined by the condition $\lambda_{ac} < l_i$.

25)We shall assume that v remains constant.

26)This appears in the region of thermal phase transitions,[24] in the course of stochastization of the process,[96] etc.

27)It is interesting to note that a laser beam with a polarization component of lower intensity becomes self-focused faster because of the transfer of energy from the stronger component, i.e., equalization of the intensities of the components takes place.[12]

28)We are speaking here of deformations of a nematic liquid crystal which are nonadiabatic in space when, by analogy with time dependences, we need to allow for terms $\delta\varphi/\delta z \sim q_z^{-1} \ll d$.

29)The angle ψ determines the deviation of the director from the initial reorientation and creates a nonlinear phase advance in the case of the transmitted light [ψ satisfies the boundary conditions $\psi(z=0) = \psi(z=d) = 0$]; for each value of ψ the rotation of the director is governed by the angle φ (at an angular velocity $\delta\varphi/\delta t$), which is responsible for the rotation of the polarization of light (representing an analog of the Mauguin limit for cholesteric liquid crystals[12]).

30)The possibility of oscillations is clear directly from Eq. (15); when ψ is increased ($\delta\psi/\delta t > 0$) we can easily see that the anisotropy of the medium increases effectively so that $\delta\varphi/\delta z$ becomes larger; this reduces the effective field inside the medium so that the reorientation threshold is not exceeded and the system returns to the initial state ($\delta\psi/\delta t < 0$); the process is then repeated.

31)If $\beta \neq 0$, the effect of light on a hybrid nematic liquid crystal tilts $\mathbf{n}$ out of the $(\mathbf{s}_1, \mathbf{s}_2)$ plane (see Fig. 17c), so that two components of the polarization of the transmitted light are observed.

32)In the case of circularly polarized light such pulsations had been observed, as already mentioned, in nematic liquid crystals[119] and also in cholesteric liquid crystals.[59]

33)In the opposite case, an equilibrium state is established after a time and this state corresponds to reorientation by a certain effective (and smaller than in the case of action of a single field component) angle. This accounts for the results presented in Fig. 18.

34)A report of the observation of this pattern was given in Ref. 78.

35)Some role may be played also by laser heating of the medium because $I_{th} = I_{th}(T)$, where T is the temperature of a sample[12]; in general, the important dependence is that of I_{th} on any parameter of the problem which changes in the process of reorientation (in particular, this is true of the dependence on the polarization of the transmitted light shown in Fig. 19, which changes inside the medium).

36)In this case we need to consider separately the energy invariants of the problem.[74,97]

37)A feedback is established also in the case of reorientation in static fields; however, in this case the field has to be inhomogeneous inside the medium and this is an analog of the state of a system for a propagating wave.

38)The difference between the frequencies ω and ω' is related to the processes of energy dissipation in a system (which may be of thermal, orientational, and other nature); the maximum gain (for the wave with the lower frequency) corresponds to the condition $\omega - \omega' = 1/\tau$, where τ is the relaxation time of the nonlinearity (in the case of liquid crystals the value of τ amounts to several seconds).

39)When the parameter controlling the system increases, the molecular parameters become cooperative and they characterize the system as a whole.[96]

40)This analysis, which makes it possible to reveal instability regions, was made in Ref. 89 for nematic liquid crystals with periodic flexural deformations affected by optical radiation. In the presence of two components of the polarization field in a nonlinear medium it would be useful to provide a description in terms of a model of two coupled (orthogonal) oscillators[8]: we can easily show that the conditions for the excitation of regenerative pulsations are then satisfied, and also that stochastic states are possible.

41)We are speaking here of open systems, so that the main thermodynamic concepts as well as the procedure for the variation of the free energy and finding its minimum for a system comprising a liquid crystal and a field would require refinement.[91,94,124]

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