Periodic phase transition driven in a nematic liquid crystal by the IR beam from a CO_2 laser

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The conditions for observing a periodic phase transition in a nematic liquid crystal during cw illumination by a CO_2 laser are described. A qualitative interpretation of the phenomenon is offered. The times required for a transition from one state of aggregation to another are estimated.

In a study of the effect of laser light with a wavelength $\sim 10.6\ \mu$m on a nematic liquid crystal, carried out in an effort to study the nonlinear response of the crystal, we have observed an interesting periodic phase transition. Phase transitions in nonequilibrium systems have recently attracted increased research interest,[1] so it seems worthwhile to describe the conditions under which this periodic phase transition occurs and to offer a qualitative interpretation of it.

The changes which occur in the nematic liquid crystal during cw illumination by the IR beam from a CO_2 laser are detected from the change in the birefringence by an interference method at the wavelength of the probe light (from an incandescent light bulb). For this purpose, the cell holding the nematic liquid crystal is placed between crossed polarizers in the optical arrangement shown in Fig. 1. The director makes an angle of 45° with the transmission direction of the polarizers. In this case we know that the intensity of the light transmitted through the crystal is the result of an interference of the ordinary and extraordinary rays, and it depends on both the wavelength of the probe light and the temperature of the sample. We chose a wavelength such that the initial intensity of the transmitted probe light, before the IR laser was turned on, was minimal. After the IR light was turned on (at an intensity of the order of 10 W/cm^2), we observed an increase in the transmission of the nematic liquid crystal for the probe light.

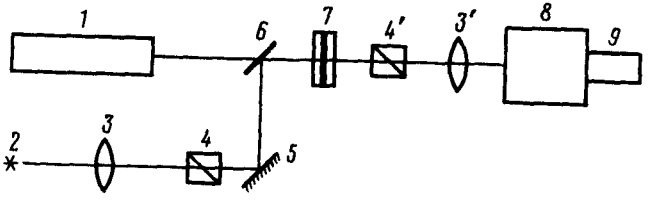

FIG. 1. Optical layout of the apparatus. 1—CO_2 laser; 2—incandescent light bulb; 3, 3'—lenses; 4, 4'—polarizers; 5—mirror; 6—GaAs beam splitter; 7—sample; 8—monochromator; 9—photomultiplier.

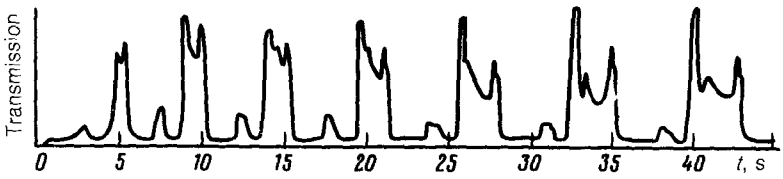

FIG. 2. Time evolution of the transmission for the probe light during cw IR illumination.

The test samples are cells 50–100 μm thick fabricated from KBr single crystals cleaved in a cleavage plane, so that the planar orientation of the director of the nematic liquid crystal is set by the direction of the easy-orientation axis of the substrate.[2] As the nematic liquid crystal we used 4-cyan-4′-*n*-pentyldiphenyl (D-205).

Figure 2 shows some typical results. The increase in the transmission during the cw IR illumination is periodic with a period which increases very slowly. Special measurements revealed that the minimum transmission corresponds to the isotropic state, and the maximum transmission to the liquid-crystal state, of the substance. The structure which can be observed in the liquid-crystal state stems from a temperature-induced change in the interference transparency, due to the strong temperature dependence of the birefringence near the point of the phase transition. This result is evidence that the constant IR illumination causes periodic changes in the temperature in the region of the probe beam.

To explain the periodic phase transition which is observed in this nematic liquid crystal, we need to consider the heating of the system due to the absorption of IR light by the crystal and the cooling due to heat removal into the substrate from the region through which the IR beam passes through the crystal. A study of the IR absorption spectrum in the region of the output from the CO_2 laser showed that the absorption coefficient for light polarized along the optic axis of the liquid crystal was larger everywhere than that for light polarized perpendicular to the optic axis. In the isotropic state, the absorption coefficient takes on an intermediate value. At the transition to the isotropic state due to heating accompanying the absorption of light with polarization along the direction, the absorption coefficient thus decreases. If the absorption is of such a nature that the steady state corresponds to a temperature below the phase transition, the liquid crystal will generally cool off, to the temperature of the phase transition. Since this first-order phase transition is approximately a second-order transition, the cooling occurs to a temperature slightly below the temperature of the phase transition. In the liquid-crystal phase, the absorption coefficient increases, as does the amount of heat evolved, and the temperature of the system begins to rise to that of the isotropic state, etc.

For quantitative estimates we need to consider the heat-conduction equation

$$\rho C \frac{\partial T}{\partial t} = \kappa \frac{\partial^2 T}{\partial R^2} - \frac{h}{d}(T - T_0) + Q(T, S) , \qquad (1)$$

along with the relaxation equation for the order parameter,

$$\gamma \frac{\partial S_{\alpha\beta}}{\partial t} = -\frac{\delta F}{\delta S_{\alpha\beta}} . \tag{2}$$

Here ρ C, κ, and γ are the density, heat capacity, thermal conductivity, and viscosity, respectively, of the nematic liquid crystal, and h is the coefficient of heat transfer to the substrate. The free energy density of the liquid crystal, F, can be written[3] in terms of invariants constructed on the tensor order parameter $S_{\alpha\beta} = S(R)(n_\alpha n_\beta - 1/3\delta_{\alpha\beta})$

$$F = \frac{1}{2} \{ L(\nabla S_{\alpha\beta})^2 + a S_{\alpha\beta}^2 + b S_{\alpha\beta}^3 + C S_{\alpha\beta}^4 \} , \tag{3}$$

where $a = a_1(T - T_c)$, $b < 0$, $C > 0$, and the homogeneous part of the free energy density describes a phase transition of first order, which is close to that of second order, from the isotropic state to the liquid-crystal state at the temperature $a(T_{1N}) = b^2/4C$ with a corresponding jump in the order parameter $S(T_{1N}) = -b/2C$. We assume that the heat evolved in the sample, $Q(T,S)$, can be written as follows as a function of the absorbed energy and the characteristics of the state of the substance:

$$Q(T, S) = Q_1 \theta (T_{1N} - T) + Q_0 , \tag{4}$$

where $\theta(T_{1N} - T) = 1$ at $T_{1N} > T$ and $\theta(T_{1N} - T) = 0$ at $T_{1N} < T$.

If we ignore effects of heat conduction within the sample, so that the temperature is established primarily by heat transfer into the substrate, we can estimate the time of the transition of the nematic crystal into an isotropic liquid in the region $R < R_0$ from

$$\tau_1 = \frac{d\rho C}{h} \ln \frac{Q}{Q - \frac{\dot{n}}{d}(T_c - T_0)} . \tag{5}$$

The relaxation time of the order parameter can be found from the rate at which a localized inhomogeneity R_0 with an order parameter $S = 0$ collapses to the nonzero value of the order parameter of the liquid crystal from the dynamics of a quasisteady switching wave[4]:

$$\tau_2 = R_0 \gamma / 2\sqrt{La} . \tag{6}$$

Adopting the parameter values corresponding to the experimental conditions of Ref. 3, i.e.,

$h \cong 2 \times 10^5$ erg/(cm^2·s·deg), $C = 4.3 \times 10^7$ erg/(g·rad), $\gamma = 0.2$ P,

$T_c = 34$ °C, $T_0 = 24$ °C, $R_0 = 0.3$ cm, $d \sim 10^{-2}$ cm, $(La)^{1/2} \sim 1 - 10^{-2}$ cgs units,

and

$Q \sim 5 \times 10^9$ erg/(cm^3·s),

we find the plausible values $\tau_1 \sim 2$s and $\tau_2 \sim 0.1 - 10$ s.

In conclusion we would like to point out that this periodic phase transition, as an example of an unusual synergetic system, can be used to advantage in a long list of

studies, to learn about nonlinear effects of electromagnetic radiation on such a "soft" system as a liquid crystal and also to determine the nature of nonequilibrium states.

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