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Temporal Dependences of Optical Nonlinearities of Nematic Liquid Crystals and the Unusual Polarization Dependence of Self Diffraction in 4-4′-Bis(Heptyloxy) Azoxybenzene

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TEMPORAL DEPENDENCES OF OPTICAL NONLINEARITIES OF NEMATIC LIQUID CRYSTALS AND THE UNUSUAL POLARIZATION DEPENDENCE OF SELF DIFFRACTION IN 4-4'-BIS (HEPTYLOXY) AZOXYBENZENE

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Abstract We present a quantitative analysis of the temporal dependences of optical nonlinearities of nematic liquid crystals induced by laser of pulse duration from nanosecond to milliseconds (or longer). The nonlinear mechanisms we have considered include thermal, density and reorientational effects. This analysis provides a useful guideline for using such nonlinear optical responses in practical devices. We also report the preliminary observation of an unusual polarization dependence in a particular nematic liquid crystal.

INTRODUCTION

Recently, there have been considerable research and development efforts in the application of nonlinear optical materials to practical devices. Materials of great promises include semiconductors (in bulk or superlattice or quantum confined forms), polymers, liquid crystals, fibers, photorefractive materials and a variety of engineered materials. In this paper, we present a detailed analytical study (based on experimentally observed phenomena) of the optical nonlinearities of nematic liquid crystal under cw or pulsed laser illumination. Our aim is to clearly define the so-called "nonlinearities" of the material, as a function of the time scale involved, in order to aid their incorporation in practical device design. Figure 1 shows an example of such a device. How fast will the device function, and what input laser power (or fluence) is needed to perform switching effect depends on the magnitude and temporal dependence of the liquid crystal nonlinearity.

In such devices, as in many others (e.g., nonlinear Fabry Perot, nonlinear filter, nonlinear transmissive elements, etc.) the fundamental optical parameter involved is the optically induced

refractive index change, which is related to the so-called third order nonlinear polarization induced by the laser beam. In liquid crystals, under laser pulses from cw to nanoseconds regime, the principal nonlinearities are due to laser induced director-axis reorientation, thermal and density changes (c.f, figure 1b).[1,2] Because of the birefringent nature of liquid crystals, the nonlinearities is highly dependent on the interaction geometry. Figure 2 shows a general one where the optical propagation wave vector makes an angle β with the nematic axis.

Following the theoretical framework presented in previous studies for the laser induced reorientational, density and temperature changes, the equations are, respectively,[3,4]

$$\frac{\mu\partial^2\Delta\theta}{\partial t^2} + \gamma\frac{\partial\theta}{\partial t} + \frac{1}{2}(\gamma_1-\gamma_2\sin 2\Delta\theta)\frac{\partial v}{\partial z} = \frac{K\partial^2\Delta\theta}{\partial t^2} + \frac{\Delta\varepsilon E_{op}{}^2\sin 2}{8\pi}(\beta+\Delta\theta) \quad (1)$$

$$\frac{\partial^2}{\partial t^2}(\Delta\rho) + \frac{v^2}{\gamma}\nabla^2(\Delta\rho) + \frac{v^2\beta\rho_o}{\gamma}\nabla^2(\Delta T) + \eta/\rho_o\frac{\partial}{\partial t}\nabla^2(\Delta\rho) = \frac{\gamma e}{8\pi}\nabla^2(E_{op}{}^2) \quad (2)$$

and

$$\rho_o C_p\frac{\partial}{\partial t}(\Delta T) - \lambda_T\nabla^2(\Delta T) = \frac{\alpha nc}{4\pi}\mathrm{Re}(E_{op}E_{op}^*) \quad (3)$$

where μ is the moment of inertia, γ's the viscosity coefficient, K the elastic constant, $\Delta\varepsilon$ the optical anisotropy, v the velocity of sound, β the bulk modulus, ρ_o the density, η the viscosity, γ_e the electrostrictive coefficient, λ_T the thermal conductivity, α the absorption constant, n the effective refractive index of the liquid crystal, and c_p the heat capacity.

The first term on the left hand side is associated with the moment of inertia of the liquid crystal[5] whereas the third term describes the flow effects. In recent reports,[5,6] it was shown that

these terms may be important under intense laser pulse excitations. In our present analysis, these terms are ignored for the sake of simplicity and also for the fact that in the practical switching devices, such processes will be avoided by choice.

The quantitative time dependences and magnitude of optical field induced refractive index changes are obtained by solving equations (1)-(3) for a specific measurement. In the present case, our theories and experimental results are based on the usual grating diffraction experiments as schematically depicted in figure 3. The two equal intensity coherent pump lasers b_1 and b_2 create a spatially periodic intensity grating on the field

$$E_{op}^2 = E_1^2 + E_2^2 + 2|E_1E_2| \cos (\vec{k}_1 - \vec{k}_2) \cdot \vec{x}$$
$$= E_0^2 (1 + \cos \vec{q} \cdot \vec{x}) \tag{4}$$

where we have written $\vec{q} = \vec{k}_1 - \vec{k}_2$, and let $|E_1| = |E_2| = E_0/\sqrt{2}$ be the real amplitude of the optical field. The dc component of E_{op}^2 gives rise to spatially uniform changes in ρ, T and ϑ, which therefore does not contribute to the diffraction of the probe beam. The spatially periodic parts may be expressed as

$$\Delta T = T(t) \cos \vec{q} \cdot \vec{x} \tag{5a}$$

$$\Delta \rho = \rho(t) \cos \vec{q} \cdot \vec{x} \tag{5b}$$

$$\Delta \vartheta = \vartheta(t) \cos \vec{q} \cdot \vec{x} \tag{5c}$$

Substituting equations (5a-5c) into equations 1-3, after some lengthy algebra[7] (similar to reference 4), the solution for the grating amplitudes T(t), $\rho(t)$ and $\vartheta(t)$ are given by, assuming a step-function input laser pulse [i.e., $t \leq 0$, $E_{op}^2 = 0$; $0 \leq t$; $E_{op}^2 = E_{op}^2$], for $t > 0$,

$$\vartheta(t) = \frac{\Delta\varepsilon\ E_{op}^2 \sin 2\beta}{8\pi\Gamma_\vartheta}\ [1 - \exp(-\Gamma_\vartheta t)] \tag{6}$$

$$T(t) = \frac{\alpha cn\ E_{op}^2}{4\pi\rho_0 C_p \Gamma_R}\ [1 - \exp(-\Gamma_R t)] \tag{7}$$

and

$$\rho(t) = \frac{\gamma_e\ E_{op}^2}{4\pi\ v^2}\ [1 - \exp(-\Gamma_B t)\ \cos\ \Omega t]$$

$$\frac{-\beta\alpha cn\ E_{op}^2}{4\pi C_p\ \Gamma_R}\ [1 - \exp(-\Gamma_R t)] \tag{8}$$

where

$$\Gamma_\vartheta = \frac{Kq^2 - (\Delta\varepsilon E_{op}^2 \cos 2\beta)/4\pi}{\gamma} \tag{9}$$

$$\Gamma_B = \eta q^2/2\rho_o \tag{10}$$

and

$$\Gamma_R = \lambda_T q^2/\rho C_p \tag{11}$$

are the reorientational, Brillouin and thermal relaxation time constants, respectively.

DISCUSSION

These changes in orientation, temperature and density give rise to an index grating. Nonlinear optical effects, for example, self-diffraction or probe beam diffractions from these laser induced index gratings have been observed and reported before.[2] In the context of this analysis, i.e., to see how induced grating amplitudes ($dn(\vartheta) = \frac{\partial n}{\partial \vartheta} \cdot \vartheta(t)$; $dn(T) = \frac{\partial n}{\partial \rho} \cdot T(t)$ and $dn(\rho) = \frac{\partial n}{\partial \rho} \cdot \rho(\varepsilon)$) depend on the laser pulse duration, it is important to point out that both the reorientational and the thermal effects are diffusive type (i.e., non-propagating), whereas the density effect contains a component (the one proportional to the electrostrictive coefficient

γ_e) that is a propagating effect associated with sound wave at the sound frequency Ω (and propagating in the direction of the grating wave vector).[8]

For typical nematics, the parameters involved in these relaxation time constants or "rise" times are known: the refractive index n is ~ 1.5 [actually $n_{||} \approx 1.7$ and $n_{\perp} \approx 1.5$], the viscosity constant $\eta \sim 7\times10^{-2}$ kg $m^{-1}s^{-1}$; v ~ 1540 ms^{-1}; $\rho_0 \sim 10^3$ kgm^{-3}; $\lambda_T/\rho_o c_p \sim 0.79 \times 10^{-7}$ m^2/s; $\gamma \sim 0.015$ kg/m.s; $K \sim 7 \times 10^{-12}$N; $\Delta\varepsilon \sim 0.65$; $\alpha \sim 10\text{-}100cm^{-1}$ [depending on wavelength, doping, material, this can vary a lot]; $\rho_o \sim 10^3$ kg/m^3.

It has been observed and measured,[8] that for a grating spacing of 17μm, the thermal relaxation time Γ_R^{-1} is on the order of 100μs (0.1ms), whereas the Brillouin decay time Γ_B^{-1} is on the order of 100ns (0.1μs). These relaxation times are inversely proportional to the grating spacing. Hence, for a 100μm spacing, the corresponding thermal relaxation time is 3.6ms, and the Brillouin decay time is 3.6μs (an important point to note is that this time scale is comparable to the time it takes the sound to propagate away from a 2mm (the typical size of the laser beam incident on the sample) interaction region!).

Considering a 100μm grating spacing (or sample thickness), we note therefore that these various contributions to the index grating will build up, or decay at widely varying time scales, as noted in previous report.[3,5-7] The so-called optical nonlinearities, therefore, are also critically dependent on the laser pulse durations. In this paper, we shall consider two distinct region: (i) steady-state and (ii) transient, for each of these three contributions.

ORIENTATIONAL NONLINEARITY:

(i) Steady state ($t >> \Gamma_\vartheta^{-1}$; $\Gamma_\vartheta \sim 1$ sec)

$$\Delta n = \frac{\Delta\varepsilon\ E_{op}^{2}\ \sin 2\beta}{8\pi\Gamma_\vartheta}\left(\frac{\partial n}{\partial\vartheta}\right)$$

$$= n_{22}^{ss}(\vartheta) I_{op} \tag{12}$$

Typically[9] observed steady state nonlinear coefficient $n_2^{ss}(\vartheta)$ associated with reorientation effect is on the order of 10^{-4} cm^2/Watt, if the optical intensity I_{op} is expressed in the frequently used unit of Watts/cm^2

(ii) transient ($t<<\Gamma_\vartheta^{-1}$; $\Gamma_\vartheta^{-1} \approx 1$ sec $= \tau_\vartheta$),

$$\Delta n(\vartheta) = \frac{\Delta\varepsilon\ E_{op}^{2}}{8\pi\Gamma_\vartheta} \sin 2\beta\left(\frac{\partial n}{\partial\vartheta}\right) t\ \Gamma_\vartheta$$

$$= n_2^{ss}(\vartheta)\ (t\Gamma_\vartheta)\ E_{op}^{2}$$

$$= n_2^{t}\ I_{op} \tag{13}$$

The "transient" nonlinear coefficient $n_2^{t}(\vartheta)$ is related to the steady state value by

$$n_2^{t}(\vartheta) = n_2^{ss}(\vartheta)\left(\frac{t}{\tau_\vartheta}\right) \tag{14}$$

For a microsecond laser pulse, therefore, we have $n_2^{t}(\vartheta) = 10^{-6}\ n_s^{ss}$ $(\vartheta) \sim 10^{-10}$ cm^2/Watt

THERMAL NONLINEARITY

(i) Steady state ($t \ll \Gamma^{-1}$; $\Gamma^{-1} \approx$ ms)

Typically observed thermal effect is on the order of (for $\alpha \approx 1$ cm^{-1}) the orientational effect or larger (if $\alpha \approx 100 cm^{-1}$), and is on the order of 10^{-3} cm^2/Watt or larger, in the steady state. In this case,

$$\Delta n(T) = \frac{\alpha c n E_{op}^2}{4\pi \rho_o c_\rho \Gamma_R} \left(\frac{\partial n}{\partial T}\right) = n_2^{ss}(T)\, I_{op} \qquad (15)$$

ii) Transient ($t \ll \Gamma_R^{-1}$; $\Gamma_R^{-1} \approx$ ms = τ_R)

$$\Delta n(\vartheta) = \frac{\alpha c n E_{op}^2}{4\pi \rho_o c_\rho \Gamma_R} \left(\frac{\partial n}{\partial T}\right) = t\Gamma_R$$

$$= n_2^{t}(T)\, I_{op}$$

where the "transient" nonlinear coefficient $n_2^{t}(T)$ is related to the steady state value by

$$n_2^{t}(T) \simeq n_2^{ss}(T)\left(\frac{t}{\tau_R}\right)$$

For a microsecond laser, therefore, $n_2^{(t)}(T) \approx 10^{-6}$ cm^2/Watt, which ranks amongst the largest of known nonlinear materials currently being investigated for optical switching applications. It is important to note here that the thermal index coefficient $\left(\frac{\partial n}{\partial T}\right)$ is also highly sensitively dependent on the temperature at temperatures near T_c, $\left(\frac{\partial n}{\partial T}\right)$ can be <u>two orders of magnitude</u> larger than its value at temperatures far from T_c.

<u>Density Effect</u>

There are <u>two</u> time-scales involved in the relaxation of the density component, the Brillouin lifetime $\tau_B = \Gamma_B^{-1}$ and the thermal

relaxation time τ_R^{-1}. Studies[6,8] have shown that in the transient case, the density effect is comparable in magnitude to the purely thermal effect (mentioned above) at temperatures far from T_c. However, at temperatures nearer to T_c, the density effect is much smaller in magnitude. Hence they may not be very useful for practical nonlinear switching in the microsecond time scale. However, its relatively shorter response (≈100ns) and the fact that there are special chiral nematic liquid crystals in the isotropic phase (where large interaction length is possible) with much larger density coefficients indicate that this effect is a potentially ideal candidate for nanosecond laser pulse switching application.

Unusually polarization dependence in 4-4'-Bis (heptyloxy) azoxybenzene

The liquid crystal used is procured from Eastman Kodak under the trade name EK10122, whose chemical structure is shown in figure 4. The sample used is homeotropically aligned and maintained in a temperature controlled environment so that the mesophases (smectic, nematic and isotropic) may be studied. The thermotropic ranges are: crystals → smectic at 74°C; smectic to nematic at 93°C; nematic → isotropic at 123°C. Sample thicknesses used range from 75μm to 100μm. The experimental setup for self-diffraction study is similar to that given in figure 3, except that the Nd:Yag laser (b_1 and b_2) are replaced by a cw Ar^+ laser (5145 Å or 4880Å). The laser is divided into two roughly equal intensity beams (1 watt) which are then combined on the sample at an almost normal incidence, with a beam crossing angle of about 0.5°. The beam spot size on the sample is 2mm in diameter. The forward transmitted pump beams are observed on a screen placed about 3 meters downstream, where the self-diffracted beams on both sides of the pump beams are clearly

separated.

The extraordinary phenomenon observed is the dependence of the self-diffraction on the relative orientation of the optical polarizations of the two incident laser beam:

1) When the two incident beams are co-polarized (i.e., both are polarized in the x-direction, which is perpendicular to the director axis of the liquid crystal in the nematic phase), very little side diffractions are observed, even though the sample is highly absorbing. This holds true for all phases (smectic, nematic and isotropic).

2) When the two incident laser beams are orthogonally polarized, very strong side diffractions are observed for the sample maintained in the nematic phase. Up to second order side diffractions are easily observed with a laser power on the order of 1 Watt (beam diameter is 2mm) with an on and off time of tens to 100's of milliseconds. The side diffractions, however, are not observed (or are extremely weak, just as case (1) above) if the liquid crystal is maintained at the smectic or isotropic phase.

The origin(s) of such cross-polarized light induced grating diffraction effect remains a mystery. Purely thermal or thermal induced orientational effects are ruled out since the co-polarized beams (case (1)) yield very small (almost invisible) diffraction. The same reasoning also rules out laser induced reorientation effect. We believe that the nonlinear mechanism is most likely similar to photoinduced anisotropy in some materials used in polarization holographic recording.[11] The fact that the effects (self-diffractions in the cross-polarized two-wave mixing process) is prominent only in the nematic phase indicates that some structural or molecular ordering changes are involved, rather than purely electronic changes, following the laser absorption.

Currently, a detailed experiment involving lasers of various time scales, wavelength in a variety of interaction geometries are

underway to conclusively ascertain the origin of this highly polarization dependent and highly nonlinear effect.

This peculiar wave mixing effect was first observed by me in 1982; it was recently reconfirmed in my present laboratory with the participation of my graduate students Hong Li, Yu Liang, Tony Peroni, P. LoPresti, Bob Lindquist and Bob Michael.

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References

1. I. C. Khoo and Y. R. Shen, Opt. Eng. 24, 579 (1985).
2. I. C. Khoo, "Nonlinear Optics of Liquid Crystals," in Progress in Optics, Vol. XXVI, Ed. E. Wolf (North Holland, 1988).
3. H. Hsiung, L. P. Shi and Y. R. Shen, Phys. Rev. A30, 1453 (1984).
4. I. C. Khoo, J. Y. Hou, G. L. Din, Y. L. He and D. F. Shi, Phys. Rev. A42, 1001, 1990.
5. See, for example, the paper by Eichler and Macdonald in this issue.
6. I. C. Khoo, et al., J. Appl. Phys. (In Press, 1990); see also I. C. Khoo, R. R. Michael, R. G. Lindquist, R. J. Mansfield and P. LoPresti, Technical Digest, Nonlinear Optics: Material, Phenomena and Devices, NLO '90. Hawaii, July 1990, p. 145-146.
7. See also I. C. Khoo, R. R. Michael and P. Y. Yan, IEEE J. Quant. Electron. QE23, 267 (1987).
8. I. C. Khoo and R. Normandin, IEEE J. Quantum Electronics, QE-21, 329 (1985); I. C. Khoo and R. Normandin, Opt. Letts. 9, 238 (1984).
9. I. C. Khoo and T. H. Liu, Phys. Rev. A39, 4036 (1989).

10. I. C. Khoo, J. Mod. Optics (In Press, 1990); Opt. Eng. 28, 1108 (1989).

11. T. Todorov, L. Nikolova and N. Tomova, Applied Optics 23, 4309; ibid 4588 (1984) and references therein.

Figures

Fig. 1 An example of a nonlinear optical switching device that combines (a) self-defocusing and (b) transmission to total-internal reflection effects where the temporal response of the liquid crystal is important. At high input power, the incident beam will self-defocus and/or switch to the total internal reflection state and thus self-limiting effect is provided.

Fig. 1b A schematic depiction of the principal nonlinear effects occurring at different time scales in nematic liquid crystals induced by 66ps Nd:Yag 0.53μm laser pulses (reference 6). The nonlinear self-diffraction effects in the picosecond regime are due to a variety of individual molecular reorientation and or electronic absorption processes not discussed in this paper, but may be found in the references quoted in references 1 and 2.

Fig. 2 General interaction geometry between a linearly polarized light with an aligned nematic liquid crystal.

Fig. 3 Schematic diagram of the experimental setup for measuring laser induced orientational, thermal and density changes.

Fig. 4 Chemical structure of the liquid crystal 4-4'-Bis (heptyloxy) azoxybenzene.

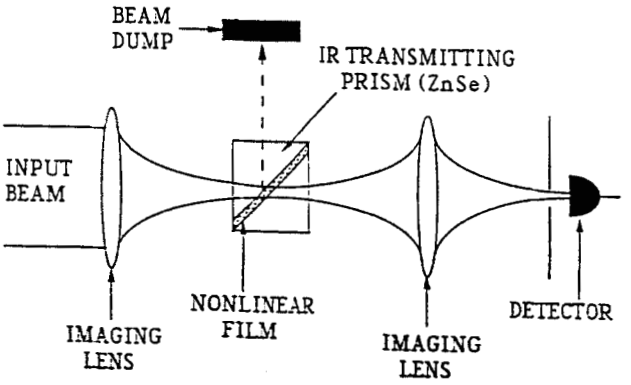

Fig. 1

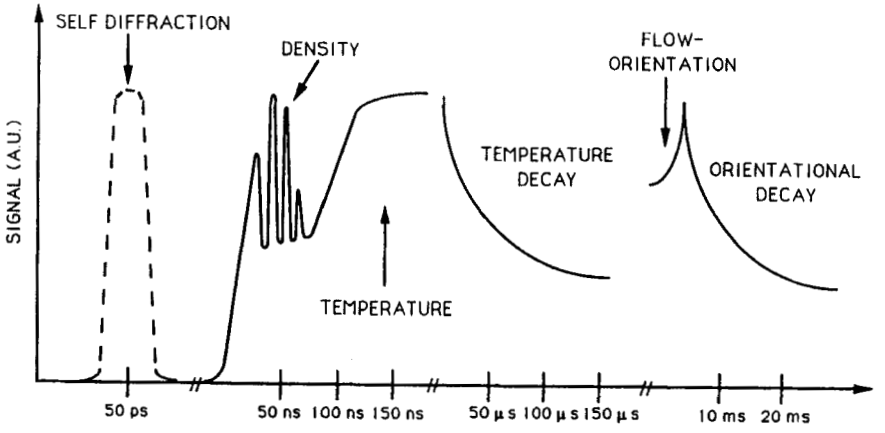

Fig. 1b

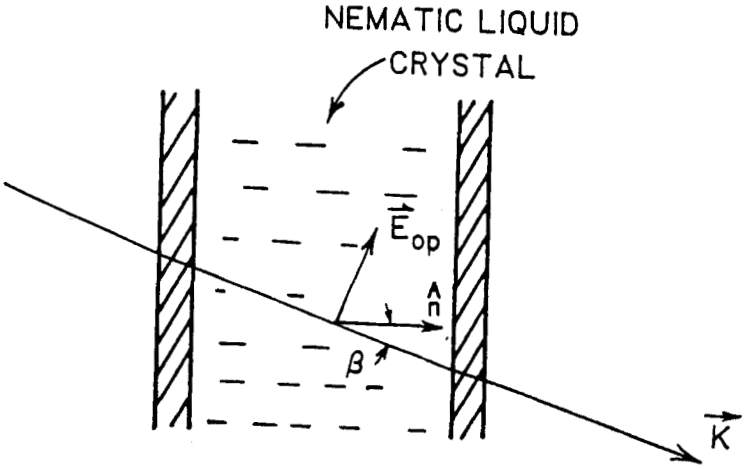

Fig. 2

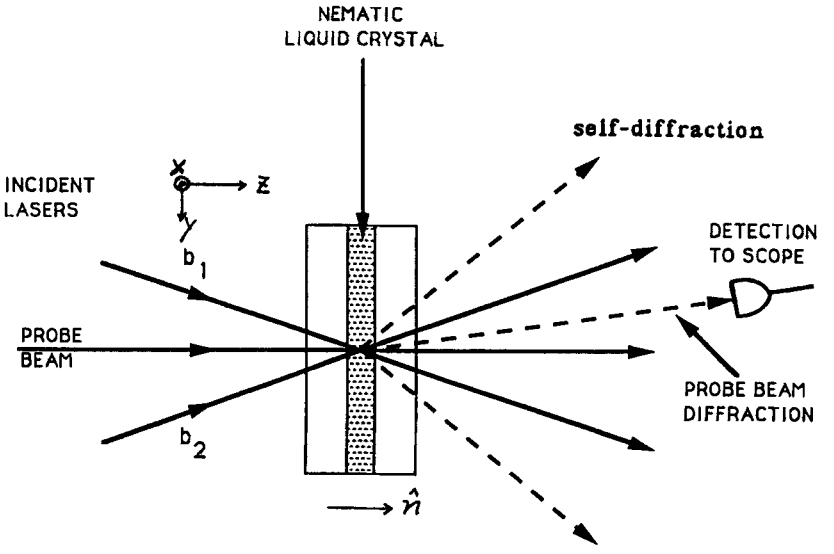

Fig. 3

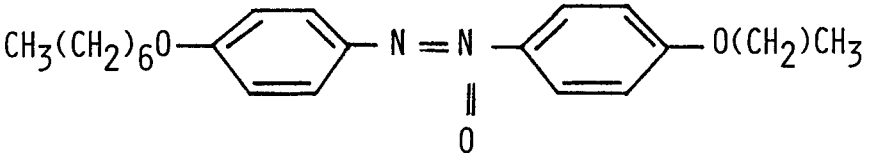

Fig. 4