

Measurement of Weak Kerr Birefringence during the First Microsecond of Field Action

A novel apparatus for the study of slowly varying 10 s transient electric birefringence mechanisms in liquids is constructed, and quantitative results are reported. The apparatus permits to perform pulsed measurements of the relative Kerr constant at a well defined moment of time after switching the electric field on. The Kerr effect dynamics is measured for carbon tetrachloride and cyclohexane in time intervals from 200 ns to 1.6 μ s following the switching on of the field. The increase in induced Kerr anisotropy amounted to 15% and 10%, respectively, and is attributed by the authors to a slowly growing transient of electrostrictive anisotropy of the medium.

1. Introduction

It is now commonly recognized that the physical properties of naturally isotropic media, acted on by a strong external electric field, become spatially anisotropic [1, 2]. Studies applying AC fields of various frequencies have confirmed the presence of numerous molecular mechanisms [3–8] of anisotropy. Kerr's effect has been studied in a wide variety of media [9–13] by means of various measuring systems permitting i.e. absolute visually [14–18] and photoelectrically recorded [19–23] measurements, assessments of the electrostrictive contribution [24–26], sine field [25, 27–29] and rectangular pulse [30–34] studies, as well as Kerr effect measurements in electric fields of optical frequencies applying nanosecond [38–41] and picosecond [42–44] pulses.

We report here results obtained with a new concept of measurement permitting to follow, from the very start, the process whereby electrically induced birefringence arises in a liquid. Our relative measurements of the Kerr constant extended over approximately 30 ns, commencing after a lapse of time ranging from 200 ns to 2 μ s, counted from the moment at which the field was switched on. Results for carbon tetrachloride and cyclohexane point to

a slow growth of birefringence in these liquids. The latter were selected because of their small anisotropy of linear polarizability, facilitating the recording of the delayed process leading to birefringence. In particular, the theoretical variation in their refractive index due to electrostriction is 5–10 times larger than the variation in refractive index due to polarizability anisotropy [45–47].

2. Theoretical foundations of the measuring method

Within the extensive framework of theoretical work on the nature of induced birefringence [3–8], the semi-macroscopic theory based on classical electrodynamics and statistical mechanics has proved to be specially effective [48, 49]. This approach includes the various molecular mechanisms which can cause a medium to exhibit birefringence, thus molecular distortion, reorientation, radial and angular correlations, redistribution, fluctuations of the molecular field and shape, etc. The electric field-induced variation in refractive index of the naturally isotropic medium is given as [48]:

$$n_{or} - n_0^2 \delta_{or} = \left(\frac{n_0^2 + 2}{3} \right)^2 [(A_2 + A_2^{es}) \delta_{or} E_L^2 + (B_2 + B_2^{es})(3E_{Lr} E_{Lr} - \delta_{or} E_L^2)]. \quad (1)$$

The quantities A_2 , A_2^{es} describe the isotropic variation in index due, respectively, to linear

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and nonlinear perturbation of molecular processes, and to electrostriction. B_2 , B_2^{es} describe the anisotropic variation due, respectively, to nonlinear perturbation of the molecular processes and to anisotropic electrostriction.

By (1), the difference in indices for orthogonally polarized beams, propagating perpendicularly to the externally applied field, is:

$$n_{\parallel} - n_{\perp} = \lambda(B_{\lambda} + B^{\text{es}})E_L^2, \quad (2)$$

where:

$$B_{\lambda} = \frac{3}{2n_0 V_m \lambda} \left(\frac{n_0^2 + 2}{3} \right)^2 \left(\frac{\epsilon + 2}{3} \right)^2 (B_m^{\text{NL}} + B_m^{\text{SF}}); \quad (3)$$

$$B_{\lambda}^{\text{es}} = \frac{3}{2n_0 V_m^2 \lambda} \left(\frac{n_0^2 + 2}{3} \right)^2 \left(\frac{\epsilon + 2}{3} \right)^2 B_m^{\text{es}}. \quad (4)$$

B_m^{NL} , B_m^{SF} and B_m^{es} are molecular Kerr constants accounting, respectively, for processes of nonlinear molecular distortion, statistical-fluctuational processes, and the process of electrostriction anisotropy.

The intensity of the light beam traversing the system of crossed polarizers is a function of the birefringence of the medium. Hence, the measurement of B reduces to that of intensity. If the electric vector \mathbf{E}_L of the field within the Kerr cell is at an angle of 45° to the polarization plane of the crossed polarizers (as it was in our experiments), the intensity recorded by the photomultiplier is $I/51$:

$$I = a^2 \sin^2(\delta/2). \quad (5)$$

where a^2 denotes the intensity of the analysing light beam on subtraction of losses in the elements of the optical system, and δ the dephasing angle between the plane waves polarized parallelly and perpendicularly to \mathbf{E}_L .

Since the shift in phase $\delta = \frac{2\pi l}{\lambda} (n_{\parallel} - n_{\perp})$ and with regard to (2) we have by (5):

$$I = a^2 \sin^2[\pi l (B_{\lambda} + B_{\lambda}^{\text{es}}) E_L^2] \\ \cong a^2 \pi^2 l^2 (B_{\lambda} + B_{\lambda}^{\text{es}})^2 E_L^4. \quad (6)$$

For relative measurements, we have in place of (6):

$$\frac{I_x}{I_c} = \frac{(B_{\lambda} + B_{\lambda}^{\text{es}})^2}{(B_{\lambda c} + B_{\lambda c}^{\text{es}})^2}. \quad (7)$$

Putting $B_{\lambda c} \gg B_{\lambda c}^{\text{es}}$, we have by (7):

$$B_{\lambda}^{\text{es}} = B_{\lambda c} \sqrt{\frac{I_x}{I_c}} - B_{\lambda}. \quad (8)$$

In benzene, used by us as reference liquid when measuring the Kerr constant, the electrostriction effect is experimentally negligible: $B_{\lambda c} \gg B_{\lambda c}^{\text{es}}$ (52).

The relaxation constant of the electrostrictive effect is much larger than that of electronic polarization [47] thus permitting the resolution of the two effects, provided a pulsed method is applied. E.g. by performing a series of pulsed measurements of the Kerr constant at various moments of time after switching the electric field on, and approximating the experimental curve to $t = 0$, we find $B_{\lambda}(t = 0)$. Next, with $\frac{I_x}{I_c}$ available from measurement at $t = T$ and $B_{\lambda c}$, $B(t = 0)$ from (8), we find B_{λ}^{es} for given T . This procedure permits the determination of the electrostrictive contribution and, moreover, the contributions from other delayed effects.

3. Experimental conditions and apparatus

Our experiments were aimed at a study of the transient of induced birefringence in media composed of centrosymmetric molecules or ones having a very small anisotropy of linear polarizability. We had recourse to the simplest method i.e. to relative measurements. As standard, we chose benzene; its Kerr constant has been measured repeatedly [52] and, moreover, is known as a function of the light wavelength. Also, the per cent contribution of electrostriction to birefringence is well known for benzene [53].

The measurement of electrically induced birefringence proceeded in three steps, consisting in the obtaining of a birefringence effect, its measurement (by determination of the light intensity traversing the system of crossed polarizers), and determination of the time at which the effect was measured. In producing the effect, we applied a voltage pulse of growth time about 200 ns and controlled amplitude up to 30 kV (corresponding to a field strength of $E = 60$ kV/cm within the cell). The voltage pulse shape is shown in Fig. 1b. The Kerr constant was measured by means of the beam of a ruby laser, which provided for high power of the measuring beam (permitting easy determination of weak birefringences), pulsewise operation (eliminating the problem of photo-

multiplier noise), monochromaticity (making a monochromator unnecessary), and directivity.

The value of the relative Kerr constant was determined in accordance with Eq. (8) expressing the intensities I as the intensity ratio of the beam emerging from the crossed polarizers, and the incident beam. The signal amplitudes of the two beams are shown in Fig. 1a. The pulse amplitude (right hand side of Fig. 1a) is proportional to the intensity of the beam transmitted by the polarizers. The amplitude of this signal multiplied by the transmittivity

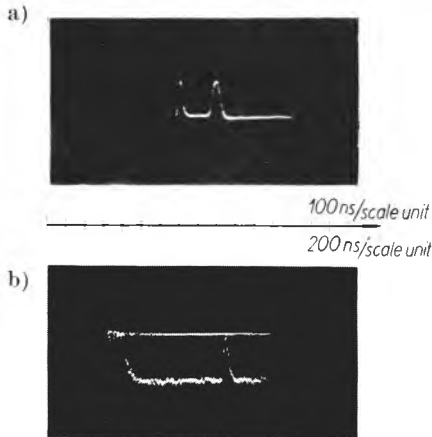


Fig. 1. Diagram of the method of Kerr constant measurement in liquids

a) oscillogram of intensity pulses of the analyzing beam before and after traversing the system of crossed polarizers;

b) oscillogram of voltage pulses on the Kerr cells (descending curve) and of the analyzing beam intensity

of the filters placed just before the window of the Kerr cell and divided by the amplitude of the first signal (left hand side of Fig. 1a) yielded the intensity value I for a given sample. The filters caused no damping of the primary signal. An identical measurement was performed for the benzene sample. Insertion of the I_c and I_x values into Eq. (8) yielded the relative Kerr constant and electrostriction contribution.

Since our setup permitted to perform relative measurements of the light intensity emerging from the system of crossed polarizers, there was no need to calculate changes in intensity due to losses (the Glazebrook prisms, Kerr cell windows, filters) except for those related to the transmission coefficients of the NG filters. The latter were placed immediately before the window of the Kerr cell in order to reduce the light intensity traversing the crossed polarizers when the cell contained a liquid of high

induced birefringence, such as benzene. Their transmission was measured with a Cary 16 spectrophotometer.

In measuring the time of Kerr effect determination, we applied a parallel system of crossed polarizers with an identical Kerr cell (Fig. 3), connected parallelly to the measuring Kerr cell and supplied from the same voltage source. In the time measurement system, we applied a He-Ne laser beam and benzene cell. The He-Ne laser beam intensity, on traversing the time measurement system, provided information on the time-evolution and value of the voltage applied to the measuring cell (Fig. 1b). On the same oscillogram, the pulse amplitude of the ruby laser beam was recorded. The distance of the laser pulse from the origin of the voltage pulse multiplied by the oscilloscope time base constant defined the time of measurement. The complete measurement involved the recording of four pulses i.e. the taking of two oscillograms as shown in Fig. 1. A series of complete measurements for different measuring times led to the determination of the time-dependence of the Kerr constant for the liquid. The optics and electronics of the measuring circuit are shown in Figs 2, 4 and 5.

The two identical Kerr cells are shown in Fig. 2. Their outer wall consisted of a boron-silicate Pyrex type glass tube of dimensions $\varnothing_{\text{ext}} = 57$ mm, $\varnothing_{\text{int}} = 51$ mm, $l = 30.7$ cm. Within the tube, a system of electrodes consist-

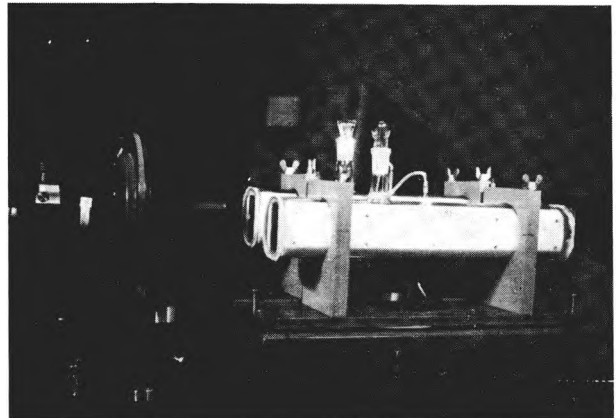


Fig. 2. Kerr cells in the measuring system, on removal of copper shield

ing of two $19 \times 19 \times 295$ mm³ brass rods, maintained at a constant distance of 5 mm by two 10 mm thick teflon plates, was placed. The electrode leads supplying the voltage were screwed into the rods through holes bored in the glass mantle and stopped with French made

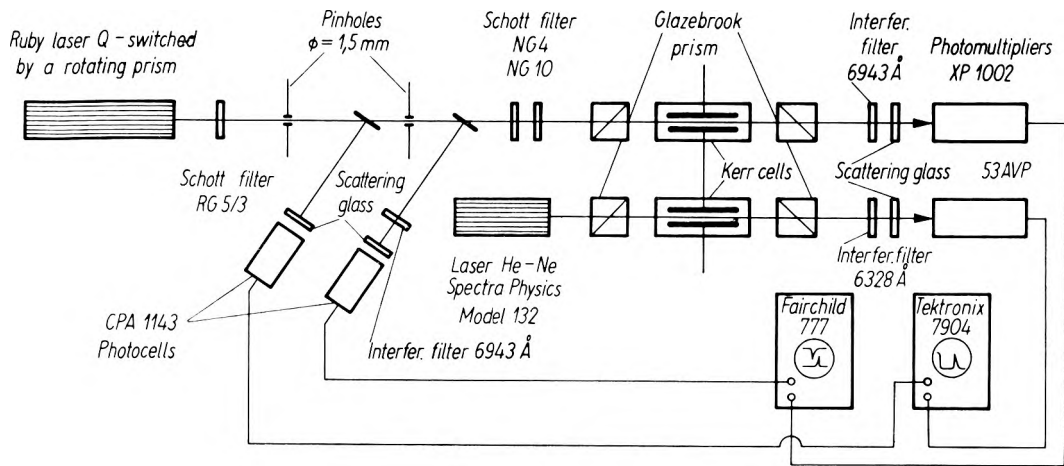


Fig. 3. Laser device for pulsed measurement of the relative Kerr constant value at a particular moment of time

rubber cement of the Conexil type. The ends of the cells were closed off with windows, made of glass taken from Kodak slides of dimensions $60 \times 60 \times 0.7$ mm³. Previous to fixing the windows onto the tube with Conexil type cement, they were selected in polarized light. Parallelity of the windows was checked by means of the He-Ne laser beam.

The two sets of crossed polarizers were a high accuracy element of the setup. The one was placed in the path of Kerr constant measurement (Fig. 3), whereas the other was in the time determination path. The polarizers were Glazebrook prisms of dimensions $10 \times 10 \times 25$ mm³, from Fichou (Paris). The attenuation accuracy of their polarization planes was checked both visually and by means of a photomultiplier, applying a rotating interrupter of the emergent light beam. The Kerr cell, filled with the liquid, was situated between the polarizers. With the latter crossed, the ratio of transmitted unattenuated and incident light intensities was tested for linearity applying light from a ruby laser with Q-switched active resonator. For cyclohexane, the ratio of unattenuated and measured light intensities amounted to about 1:10 at a voltage of 16 kV on the Kerr cell. On filling with another liquid, the cell was put in its former position and a measurement was made to check whether the unattenuated light intensity (at zero field) exhibited its initial value. Fluctuations in unattenuated light intensity did not exceed 0.1 of its amplitude.

The direction and cross-section of the measuring beam was defined by a system of two identical pinholes of diameter $\varnothing = 1.5$ mm distant by 112 cm. The diameter of the fluctuation

area of the beam spot position did not exceed 4 mm when measured at a distance of about 5 cm behind the back window of the Kerr cell.

When measuring the transmitted beam intensities we applied two photomultipliers, the one being of the XP 1002 and the other of the 53 AVP type, made by La Radiotechnique (France), operating at a 50 Ω load and supply voltage 1300 V. The maximal measured signal amplitude did not exceed 0.5 V. Linearity was checked up to 2 V. In dealing with strongly birefringent liquids, a set of calibrated NG filters was used in order to reduce the intensity of the signal measured. The filters were inserted just in front of the window of the cell containing the liquid under investigation (Fig. 3).

Three problems pertaining to the electronics were solved simultaneously in the measuring circuit: that of the production of the voltage pulse setting off the Kerr effect, that of synchronization of the pulse and the shot from the laser emitting the measuring beam, and that of the simultaneous intensity measurement of short pulses from 4 mutually independent radiation detectors.

In order to produce voltage pulses of some tens of kV with a steep growth slope, we had recourse to an earlier method of obtaining voltage pulses by relaxational discharge of a condenser (Fig. 4). A 2nF/30 kV DC charged condenser was connected parallelly to the Kerr cell through a spark gap, operating in an atmosphere of nitrogen at a pressure of e.g. 4 atm at a discharge voltage of 16 kV. The steepness of the condenser discharge pulse front was adjusted by varying the 330 Ω resistance (Fig. 4).

Synchronization of the ruby laser light pulse and Kerr voltage pulse was ensured by a rotating prism, which acted also as Q-switch modulator of the active resonator. The air driven

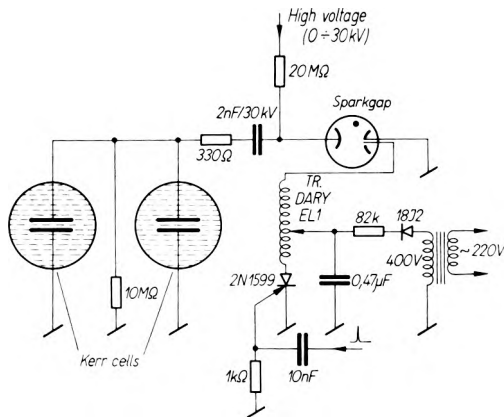


Fig. 4. Block diagram circuit producing voltage pulses on the Kerr cells

turbine operating the prism rotated at a rate of 24,000 rev/min, to within about 2%.

A block diagram of the electronic system for synchronization of the high voltage pulse and laser shot is shown in Fig. 5. Its operation is explained in Fig. 6.

The detector in the laser turbine casing emitted two electronic pulses U_1 per revolution of the prism. The first pulse arose about 0.8 ns before the prism attained a position enabling the release of laser emission. The second pulse arose approximately 2 μs before the prism

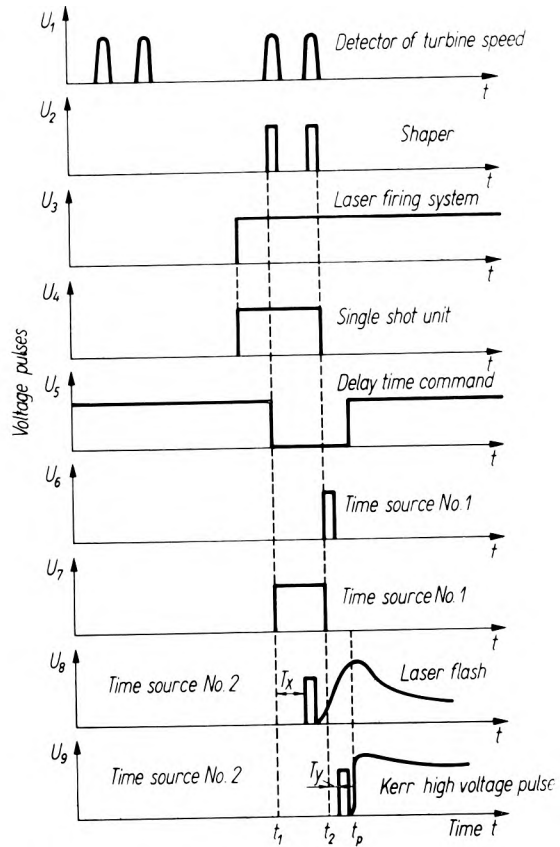


Fig. 6. Voltage forms in the synchronization system

came into emission position. The necessity of producing two pulses was imposed by considerations of turbine rotation stability. It was not possible to assume the first pulse as reference for Kerr pulse command because of the large fluctuation of its position amounting to

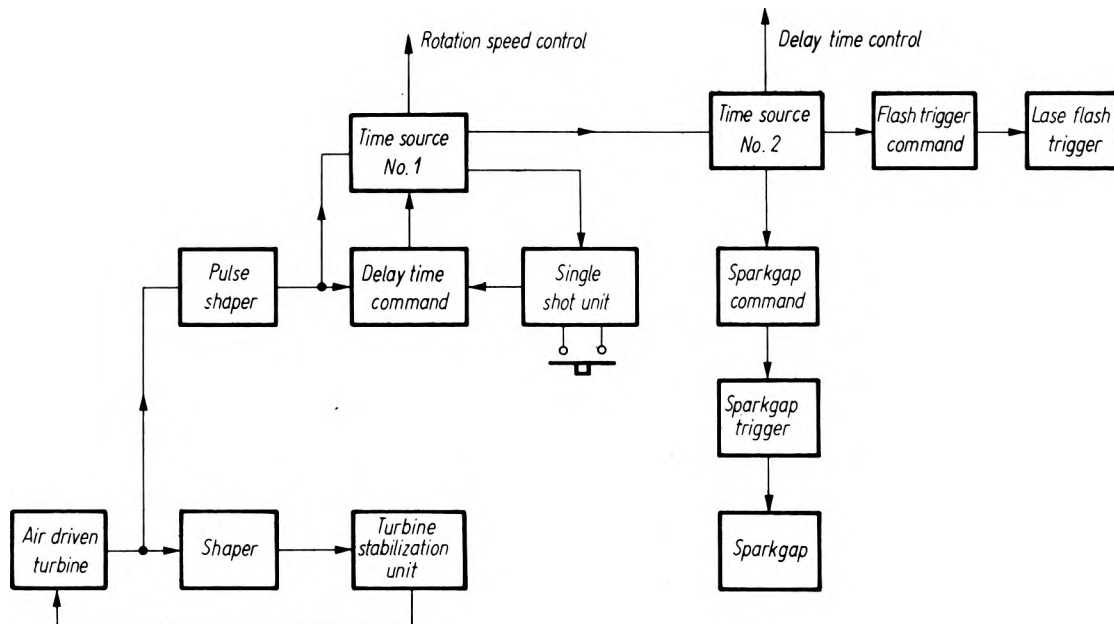


Fig. 5. Circuit for synchronization of the Kerr voltage pulse and ruby laser shot, schematically

2% of the number of prism revolutions i.e. to about 18 μs , as this would involve a time interval of reference signal fluctuation exceeding that of programming the measurement of the effect amounting maximally to 2 μs . The pulse shaper transformed the pulse U_1 into pulses U_2 of regular shape. On pressing the button of the laser firing system a voltage pulse U_3 appeared. The front slope of U_3 opened the single shot unit giving rise to a pulse U_4 , which then controlled the delay time command. A basic element of the electronic synchronization system consisted of the time source No. 1, composed of an RS computer. The first pulse of each pair of pulses, U_2 , was fed to the input S of the time source. This pulse released the time source only if compatible with the delay time command; this occurred by removal of the block voltage U_5 . Feeding the second pulse of each pair to the input R stopped the time source No. 1. The computer switched under the action of the back slope of this pulse. Simultaneously, the back (falling) slope of the second pulse released the single pulse generator, which opened the delay time command by way of the single shot unit. The shaped voltage pulse U_7 controlled the operation of the time source No. 2, on the output of which two pulses with controlled delay time were obtained. The pulse U_8 controlled the laser flash trigger by way of the command system permitting to go over from TTL systems to discrete DTL ones. The second pulse served as spark gap command. The spark discharge caused earthing of the electric charge on one of the plates of the 2nF/30 kV condenser (see Fig. 4) and gave rise to a potential difference on the Kerr cells. Spark gap operation was stable at well defined conditions of voltage and nitrogen pressure.

4. Discussion of the experimental results

By the method described in Section 2, we studied the value of the $B_x/B_{C_6H_6}$ ratio throughout the interval of times from about 200 ns to 1.6 μs , counted since the moment of time at which the electric field was applied to the sample, for two liquids: carbon tetrachloride, and cyclohexane. The results are given by the graph of Fig. 7. The position of the straight lines was determined by the least squares method. Time dispersion of $B_x/B_{C_6H_6}$ is more marked for carbon tetrachloride than for cyclo-

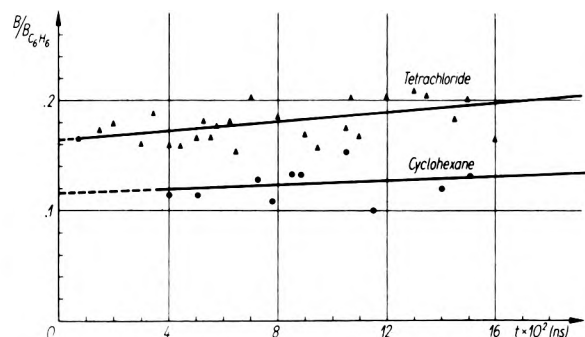


Fig. 7. Kerr constant as a function of the time of its measurement counted from the moment of time at which the electric field is applied to the cell

hexane, amounting to 15% and, respectively, 10% of the increase with time from 200 ns to 1.6 μs , counted from the moment of field application onwards. With regard to the rather slow process of growth, the interpretation of the observed increase in Kerr constant is by no means easy. Considering the known mechanisms of birefringence, the observed increase can be explained as due to the contribution of a slowly growing process of electrostriction. Since the generally accepted value of the electrostriction relaxation constant is approximately 10^{-9} s, our measurements are evidence that the electrostrictive process grows much more slowly or that two growth segments are present: a segment of rapid electrostriction, and one of slow electrostriction. This hypothesis, however, is hardly plausible.

On approximating the straight lines of Fig. 7 to $t = 0$, the relative Kerr constants of carbon tetrachloride and cyclohexane have been determined as 0.17 ± 0.01 and 0.11 ± 0.03 , respectively. These values are compared with those of other authors in Table, where they are seen to lie between those derived from electric birefringence and optical birefringence measurements. Since our results are derived by extrapolation of the final stage of the Kerr effect transient, the relation observed appears to be justified. A further corroboration is to be found in the high sensitivity of the measuring method proposed.

The apparatus described above permits to perform investigations of a wide variety of phenomena in which molecular processes are dependent on an external electric field. In particular, it enables to study Kerr effect transients at several wavelengths, namely 6943; 3471; and 10600 \AA (obtained by an exchange of the laser element in the head of the ruby laser),

Medium studied	Author	Wavelengths of the polarizing and analyzing beams		Kerr constant from electric birefringence measurement		Kerr constant from optical birefringence measurement	
		λ_{pol} [Å]	λ_{an} [Å]	$B_e \times 10^9$ [cm esE ²] ⁻¹	$\left(\frac{B_{medium}}{B_{C_6H_6}}\right)_0$	$B_0 \times 10^9$ [cm esE ²] ⁻¹	$\left(\frac{B_{medium}}{B_{C_6H_6}}\right)_0$
Benzene C ₆ H ₆	M. Paillette 1969	6943	4880	36.7 ± 1 ₂	1	40 ± 8 ₁	1
	A. Proutière 1973	pulsed field	6328		1		1
	R. W. Hellwarth 1968	pulsed field	6328		0.20 ± 0.02 ₃		
Carbon tetrachloride CCl ₄	M. Paillette 1969	6943	4880	8.3 ± 1 ₂	0.22 ± 0.03 ₂	5.1 ± 1 ₁	0.13 ± 0.03 ₁ 0.065 ₄
	J. R. Lalanne 1971	10600	5300				
	A. Proutière 1973	pulsed field	6328				
	present authors	pulsed field	6943				
Cyclohexane C ₆ H ₁₂	M. Paillette 1969	6943	4880	4.4 ± 1 ₂	0.12 ± 0.03 ₂	4.1 ± 0.8 ₁	0.1 ± 0.02 ₁
	A. Proutière 1973	pulsed field	6328				
	present authors	pulsed field	6943				



Fig. 8. Disposition of optical elements and detectors in the apparatus for measurements of weak Kerr birefringences during the first microsecond of external electric field action

variations induced in the absorption of the analyzing beam and its harmonics (linear and nonlinear induced dichroism), as well as electric

field-induced fluorescence, excited by the analyzing beam. With regard to this wide range of applications, we did not hesitate to give an extensive description of the method (general view — Fig. 8).

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Les mesures de faibles de Kerr doublement réfractées au cours de la première microseconde de l'action dans le champ

On a construit un nouveau appareil pour mesurer le mécanisme transiént (10⁻⁷ g) de biréfringence électrique changeant lentement dans les liquides, en donnant des résultats quantitatifs. L'appareil permet d'effectuer des mesures d'impulsions de constante relative de Kerr après le branchement du champ électrique au moment précisément défini. La dynamique du phénomène du Kerr a été mesuré pour le tétrachlorure et cyclohexane dans les intervalles de 200 à 1,6 μs après le branchement du champ électrique. L'accroissement du caractère anisotrope induit des facteurs mentionnés a été de 15% et 10%; il est du à une lente augmentation de l'anisotropie électrostrictive transiént du centre.

Измерение двупреломленных слабых Керра в первую микросекунду действия в поле

Построен новый аппарат для исследования медленно изменяющегося переходного (10⁻⁷) механизма электриче-

ского двупреломления в жидкостях; приведены количественные результаты. Аппараты позволяют производить измерения импульсов относительной постоянной Керра после включения электрического поля в строго определенный момент времени. Динамика эффекта Керра измерена для тетрахлорида и циклогексана в промежутках времени от 200 нс до 1,6 мкс после включения поля. Рост индуцируемой анизотропии названных факторов составил 15 и 10%, что связано с медленно повышающейся электрострикционной переходной анизотропией среды.

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