Physics of Our Days

NEW OPTICAL METHODS OF STUDYING RAPID PROCESSES

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Usp. Fiz. Nauk 104, 77–94 (May, 1971)

1. INTRODUCTION

METHODS of direct measurement and study of the time and kinetics of rapid processes are of great value in different branches of science and technology. Modern optical direct-measurement methods in the nanosecond and picosecond time range have become possible because of the successful development of the technique of generating intense short and ultrashort pulses of light, and have begun to be developed during the last two—three years. In this article we report the main principles of new methods, and will not devote much space to a discussion of the physical results obtained by these methods. The latter question is worthy of an independent detailed study.

Prior to the appearance of pulsed laser sources of light, rapid processes were studied with the aid of different types of mechanical time scanning of the image of the investigated phenomenon. This method has made it possible to study processes much shorter than 10^{-7}—10^{-8} sec[1,2].

Processes occurring within a time $\sim 10^{-9}$ sec already required a more complicated procedure, using modulation of the light either in an ultrasonic cell, or in Kerr or Pockels cells[3-5]. Direct methods of investigating processes developing within less than 10^{-9} sec encountered seemingly unsurmountable difficulties. To increase the ultrasonic modulation frequency above 10^9 Hz calls for the use of cryogenic techniques, which greatly complicates the use of this method and is limited to a frequency $\sim 10^{10}$ Hz.

The use of a Kerr cell to modulate light (at a higher frequency than 10^9 Hz) seemed utterly impossible following the publication of[6], since the anisotropy relaxation time in nitrobenzene and chloroform was found there to equal 2×10^{-9} sec. Such a result contradicted our indirect determination of the anisotropy relaxation time[7,8], and it was soon reported[8] that the conclusions of[6] were incorrect. However, modulation at high frequencies, based on the use of the Kerr effect for the measurement of times shorter than 10^{-9} sec, was not realized.

The light-modulation technique based on the Pockels effect was developed successfully. This method has made it possible to modulate light with a frequency $\sim 10^{10}$ Hz and with an even higher frequency[4,9], but insofar as we know this method was not used for direct measurements of rapid processes, although it has many strong points and possibly will still find application in the future.

In recent years, a method has been effectively developed for measuring rapid processes by using electronic image scanning in instruments of the type of the electron-optical image converter[10-12], and even now makes it possible to study processes with duration 10^{-10}—10^{-11} sec.

We shall not discuss here this and the already mentioned "old methods of direct measurement and study of rapid processes, but will concentrate our attention on new optical methods.

The new methods which will be discussed below can be arbitrarily subdivided into two types:

1) Methods in which two crossed or collimated light pulses are used, one of which is intense and causes the investigated phenomenon (nonlinear optical phenomenon), and the other is a weak pulse of equal or unequal duration and wavelength, and serves to applaud or sound the phenomenon during different instants of its development.

2) A method likewise employing two light pulses, with the intense pulse producing the high-frequency Kerr effect in a cell (for example with carbon disulfide); if this pulse is short (for example, of picosecond duration), it produces an ultrafast optical shutter. The weak pulse, which can also be of equal or unequal wavelength and duration, can carry information concerning the rapidly developing phenomenon and can be registered during different phases of the development of the phenomenon, by means of picosecond exposures.

The only requirements imposed on the low-intensity beam are that the phenomenon be registered during the exposure time and that this light not cause any nonlinear effects. As yet, there are only a few papers devoted to the development of the new methods, but these methods have already been used for the study and measurement of such phenomena and quantities as the time of nonradiative transitions, the time of bleaching and recovery of absorption in saturable solutions, the time of fluorescence quenching, the lifetime of phonons in a crystal at different temperatures, and the development kinetics of a plasma produced in the focused pulse of intense light.

2. DETERMINATION OF THE LIFETIME OF PHONONS PRODUCED IN STIMULATED MANDEL'SHTAM-BRILLOUIN SCATTERING

The study of the absorption of high-frequency sound in crystals at low temperatures has made considerable progress in recent years. The use of the method proposed by Baranskiĭ[13] has made it possible in a number of cases to study the absorption of hypersound up to frequencies 114 GHz. The absorption of high-frequency sound could be studied earlier only at very low helium temperatures or close to them[14-15]. The phonon lifetime region that is most difficult to study lies between 10^{-7} and 10^{-8} sec. This region was recently investigated by using nanosecond pulses of a ruby laser[16,17]. The experimental setup is shown in Fig. 1a. The beam from a Q-switched ruby laser, with peak power 15 MW and half-width 16 nsec, was directed into a sample of Z-cut quartz placed in a cryostat. A small fraction of the light was diverted, delayed in time, and transmitted in the same direction with the initial polarization. The bulk of the beam (with polarization rotated through 90°) produced stimulated Mandel'shtam-Brillouin scattering in the single-crystal quartz sample and this in turn generated an intense hypersound wave moving in the direction of the exciting light[12]. The weak, diverted light beam, which we shall call here and henceforth the probing or sounding beam, was delayed relative to the exciting light by 48 nsec.

If this wave has not had time to attenuate by the instant it arrives in the volume where the hypersonic wave was generated, then the probing light is reflected almost completely from the acoustic "grating." The larger the damping, the less intense the reflection. In Fig. 1b, curve 1 corresponds to the Mandel'shtam-Brillouin light without delay, and curve 2 shows how, at the same delay, the attenuation of the hypersound increases with increasing temperature from 32 to 143°K.

The results of the measurement of the phonon lifetime and the corresponding hypersound absorption coefficients at 29 GHz are shown in Fig. 2. It is seen

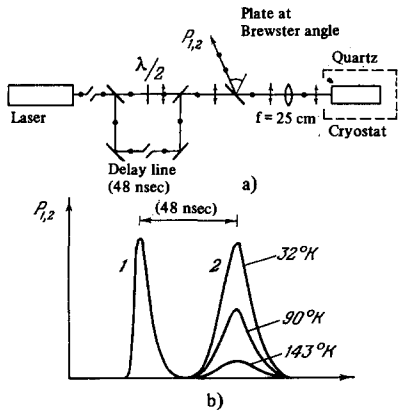

FIG. 1. a) Setup for the determination of the lifetime of phonons generated in stimulated Mandel'shtam-Brillouin scattering [16,17]. b) Oscillograms of current produced by light reflected backwards at different quartz temperatures. 1—Pulse of SMBS light scattered backward without delay; pulses with the same delay (48 nsec) at three quartz-sample temperatures.

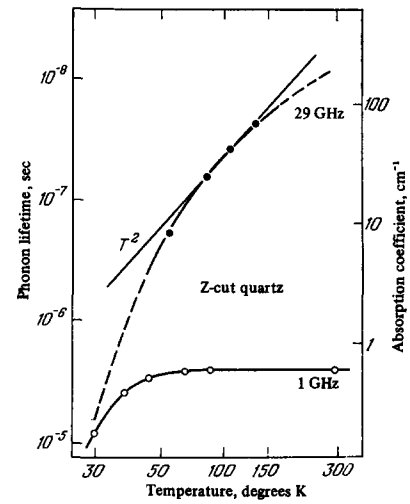

FIG. 2. Lifetime of phonons in quartz at 28 GHz in a quartz crystal at different temperatures. The dashed lines give an extrapolation, quadratic in the frequency, of the phonon lifetime. For comparison, earlier data are presented for 1 GHz frequency [16,17].

from Fig. 2 that in the temperature region ~100°K the absorption coefficient for 29 GHz decreases like the square of the temperature and the plateau obtained for the frequency 1 GHz (which is shown for illustration in the same figure), does not appear.

If it is assumed that the absorption of the hypersound is due to the interaction with thermal phonons (i.e., phonon-phonon interaction[18,19]), then it must be proposed that $\omega\tau_{ther} \geq 1$ and for 29 GHz, up to a temperature of 150°K, the lifetimes of the corresponding thermal phonons (including U- and N-processes) are $\tau_{ther} \geq 0.5 \times 10^{-11}$ sec.

A study od phonon-phonon interaction by investigating the absorption of sound in crystals, particularly at low temperatures, has become an interesting and important branch of solid-state physics[20]. In the present article we shall not touch at all on this important question, but merely call attention to a new and simple method of measuring the lifetime of phonons produced in stimulated Mandel'shtam-Brillouin scattering.

3. METHODS OF DIRECT DETERMINATION OF THE TIME OF RECOVERY OF ABSORPTION OF CERTAIN DYE SOLUTIONS BLEACHED BY INTENSE LIGHT

It is well known that cells filled with solutions of cryptocyanine, phthalocyanine, or other dyes in various solvents, usually designated by some definite code, are used as Q-switches in lasers.

The cell with a solution of such a dye is placed inside the optical resonator and spoils its Q to such an extent, that no light pulse is generated at any level population inversion capable of producing stimulated emission in the absence of the cell with the absorber.

Such a "delay" of the stimulated emission leads to an even larger inversion, as a result of which there is produced a stimulated emission, passing many through the cell, of intensity sufficient to transfer almost all the molecules of the light absorber to an excited state, making the solution transparent, and generation begins

at a large degree of inversion. After the action of the intense light has ceased, the excited dye molecules return to the ground state and the solution becomes opaque. It is important to measure the lifetime of the molecules in the excited state or the time of recovery of the earlier absorption. These times are very short, and therefore the previously available direct methods were not suitable for their measurement.

The appearance of lasers with mode locking, which emit trains of picosecond pulses of several picoseconds duration separated by time intervals of several nanoseconds, has made it possible to develop methods for direct measurement of the lifetime of the excited state.

Recognizing that the lifetime of such dyes in the excited state amounts to several picoseconds, the time elapsed between pulses is sufficient to permit the system to return completely to the initial equilibrium state. In other words, the longest relaxation time of the system is much shorter than the time between two neighboring pulses.

The experimental method for directly measuring the picosecond lifetime of the molecules in the excited state or the time of nonradiative depletion of the excited level was described by Shelton and Armstron[21] and was used by Scarlet et al.[22]. The kinetics of excitation and depletion of this rapidly developing process can be traced. Indeed, if one has a Q-switched laser with mode locking such that in the train of picosecond pulses each individual pulse is separated from its nearest neighbors by a time, say, 4×10^{-9} sec, as in the experiment of Scarlet et al.[22], then it is possible to trace with the aid of a probing picosecond pulse the process of bleaching of the solution and of the recovery of its absorption.

By way of an example, Fig. 3 shows the setup used in the experiment of[22]. A reflecting mirror is placed at a distance 60 cm behind the investigated dye solution (Kodak 9860) and the light filter F. If the time between two neighboring picosecond pulses is 4nsec, then the light of the n-th pulse reflected from the mirror M_1 will encounter in the sample the (n + 1)-st pulse entering the sample. The intensity of the probing pulse I_0 is registered with a photodiode, and the intensity of the probing pulse leaving the sample at a certain instant is registered with another photodiode. By varying the

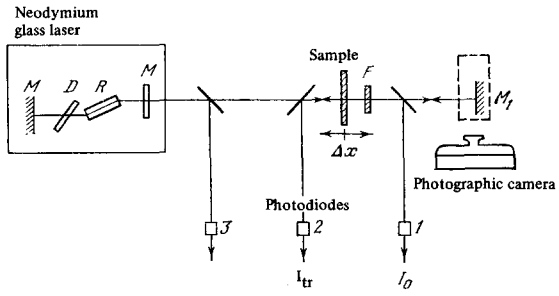

FIG. 3. Setup for the direct measurement of the time of recovery of absorption of saturable dye solutions [22]. M and M_1–mirrors, R–neodymium-glass rod; D–saturable dye solution; 1–3–photodiodes, I_0–intensity of the probing light; I_{tr}–intensity of the probing beam transmitted through the dye solution. The mirror M_1 can be placed in the cell (shown shaded) to determine the duration of the pulse by the two-photon luminescence method.

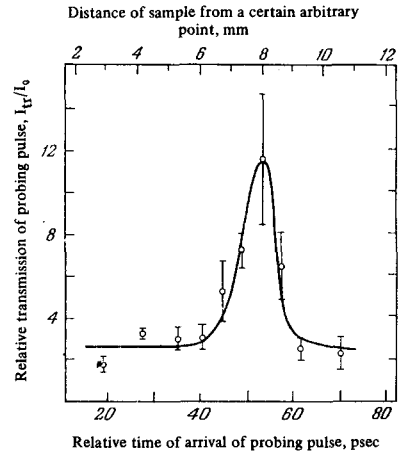

FIG. 4. Plot of the ratio of the intensities I_{tr}/I_0 against the delay time of the probing pulse relative to the bleaching pulse [22].

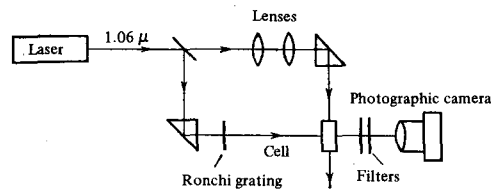

FIG. 5. Diagram of setup for the direct determination of the time of recovery of absorption of a saturable dye solution [24]. The delay of the probing beam relative to the bleaching beam is with the aid of quartz plates placed in the path of the probing beam.

position of the sample along the axis of the entire setup, it is possible to probe the solution during different instants of occurrence of bleaching and recovery of the absorption. Figure 4 shows a plot of I_{tr}/I_0 against the relative shift of the appearance of the bleaching and probing pulses in the sample.

Measurements of the intensity with an oscilloscope having a relatively large persistence, and consequently averaging over different pulses, can be regarded as permissible, since the filter F attenuates the bleaching pulse to such an extent that it cannot produce any nonlinear effects as a probing pulse, and it must be assumed that I_{tr} is proportional to I_0. A change Δx in the position of the cell corresponds to a time delay $\Delta t = 2\Delta x/c$.

The curve shown in Fig. 4 does not reveal the expected rapid exponential growth and decrease. In the opinion of the authors of[22], this is due to the finite dimensions of the sample solution and the finite length of the pulse.

The time of travel of the light in the solution is $T_W \approx 3 \times 10^{-12}$ sec, and the pulse duration is estimated at $T \approx (3-5) \times 10^{-12}$ sec. Taking these data into consideration and taking the half-width of the curve of Fig. 4 approximately equal to $2T_W + \tau/2$, where τ is the lifetime in the ground electronic vibrational state, we find that the measurement of the half-width yields the value $\sim 9 \times 10^{-12}$ sec, and therefore $\tau \approx 6 \times 10^{-12}$ sec.

The authors of[22] propose that the value of τ obtained by them in the preliminary experiments can

actually differ from the true one by a factor of two, although it is good agreement with the results of an indirect determination of this value, $\tau = 8 \times 10^{-12}$ sec.

Recognizing that the duration of the picosecond pulses was determined by the two-photon luminescence method[23], which actually does not give a good idea of the characteristic of the picosecond pulses, the error may be even larger, but in this case the measurement accuracy is not so important as possibility opened for investigating processes that last several microseconds, something utterly impossible by earlier measurement methods.

Malley and Rentzepis[24] proposed a simple method of determining the bleaching time of saturable solutions. Their experimental setup is shown in Fig. 5. A Q-switched laser with mode locking produces radiation at a wavelength $\lambda = 1.06\ \mu$ in the direction indicated by the arrow. A beam-splitting plate diverts a small fraction of the beam (probing beam), which is deflected by a prism and passes through a thick Ronchi grating so as to make the light beam sufficiently broad and uniform. This beam illuminates the entire cross section of the cell. The directly-transmitted intense light is compressed by a telescopic system to a cross section 2 mm^2 and is directed to the cell, the width of which is exactly 2 mm. Consequently, the intense light bleaches the solution throughout the thickness of the cell. By placing quartz plates of different thicknesses in the path of the probing beam, it was possible to vary the delay time, and a camera placed behind the cell could record the degree of transparency of the solution. Figure 6 shows the corresponding photographs and microphotograms.

The observed pictures are, of course, the result of a convolution of the functions describing the bleaching and the probing pulses, with the function characterizing

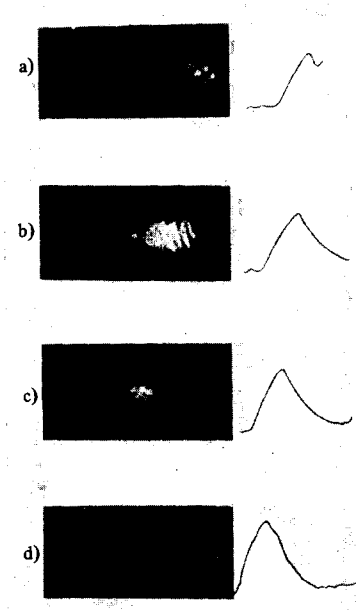

FIG. 6. Photographs of the probing-beam light passing through the cell with different delay times, and microphotograms of the photographs of this light. a) Without delay; delay was produced by plates of thickness: b) 6.3 mm; c) 12.5 mm; d) 18.8 mm.

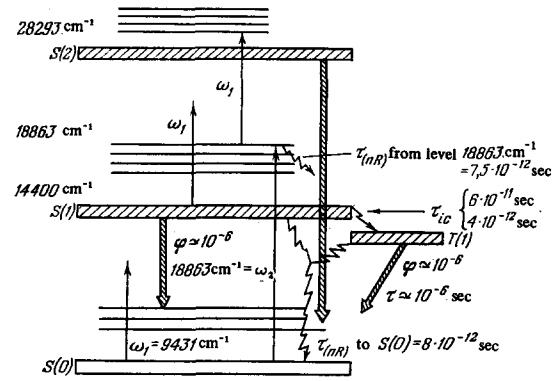

FIG. 7. Energy-level scheme of the azulene molecule. τ_{nR}—time of nonradiative transition; τ_{ic}—time of internal cross relaxation in the transition S(1) → T(1) [27].

the depletion of the excited state of the molecules of the dissolved substance. The authors propose that the duration of the bleaching pulse ($\sim 10^{-12}$ sec) is short compared with the relaxation time of the solution molecules and with the time of recovery of the absorption by the solution, and they therefore propose the following simple model, which describes satisfactorily the optical density of the solution.

It is proposed that the time dependence of the optical density $\alpha(t)$ is given by

$$\alpha(t) = \begin{cases} \alpha_0 & t < 0, \\ \alpha_0 (1 - e^{-t/\tau}), & t \geqslant 0, \end{cases} \qquad (1)$$

where $t = 0$ is the time of arrival of the bleaching pulse and α_0 is the optical density prior to bleaching. The optical density $\alpha(x)$ along the bleaching beam is

$$\alpha(x) = \begin{cases} \alpha_0, & x < 0, \\ \alpha_0 \left[1 + \frac{\delta}{d} (1 - e^{-x/\delta}) \right], & 0 \leqslant x < d, \\ \alpha_0 \left[1 + \frac{\delta}{d} (1 - e^{d/\delta}) e^{-x/\delta} \right], & d \leqslant x; \end{cases}$$

x is the coordinate in the direction of propagation of the bleaching beam, d is the cell thickness (2 mm), and $\delta = v\tau$, where v is the group velocity of the pulse (2×10^{10} cm/sec). These formulas describe well the experimental results and yield for the Kodak-9740 dye solution a relaxation time $\tau \approx 10^{-11}$, which is in good agreement with the value of τ obtained by indirect methods.

The described method can also be used for many other investigations, and its variants have already been used[24] to determine the luminescent emission in a number of solutions and organic liquids.

A method for measuring the lifetime in the electronic vibrational state, also based on the use of picosecond pulses, was developed by Rentzepis[25].

The same method and its modification have enabled Drant et al.[26] and Rentzepis[27] to measure in certain organic liquids the lifetime in the electronic vibrational state, the lifetimes of non-radiative transitions, and the values of the cross relaxation.

By way of example, Fig. 7 shows the energy-level scheme of azulene and indicates the picosecond times measured in the cited investigations.

In the original Rentzepis method[23], a train of picosecond pulses of wavelength $\lambda_1 = 1.06\ \mu$ passed

through a KDP crystal, from which light pulses $\lambda_1 = 1.06\ \mu$ (ω_1) and $\lambda_2 = 0.53\ \mu$ (ω_2) emerged together. These pulses entered a cell with a liquid (bromobenzene), where the pulses were spatially separated as a result of the differences in the group velocities of ω_1 and ω_2, and later entered a cell with azulene, the pulse ω_1 being the first to enter. Azulene has the property that it does not fluoresce when excited to the vibrational level S(1), and fluoresces only when the molecule is excited to the next vibrational state S(2). The pulse ω_1 cannot excite S(2) by either the single-photon or the two-photon mechanism. Two-photon excitation of S(2) by the pulse ω_2 is possible, but it has such a low intensity that the fluorescence light cannot be observed. Excitation to the S(2) level is possible in a stepwise manner. The pulse ω_2 is capable of exciting the level S(1) ($\nu \approx 5$). The pulse ω_1, which has already passed through the cell, is reflected from the mirror, returns to the cell, and encounters the pulse ω_2 and the molecules excited by it at the level S(1). This pulse ω_1 is capable of transferring the molecules from the excited state S(1) to the state S(2), from which fluorescence will be observed. Thus, the pulse ω_2 is the probing one, and the fluorescence is a function of the relative delay of the pulses ω_1 and ω_2. The corresponding width of the emission spectrum is determined by the duration of the laser pulse and by the time of vibrational relaxation[25].

A certain modification of this method has made it possible to measure also the cross relaxation[27]. In all cases, the character of the train of light pulses emitted by the Q-switched laser with mode locking was investigated with a high-speed oscilloscope having a time resolution 0.5 nsec. Such an oscilloscope shows clearly a "comb" of pulses separated by 4–5 nsec from each other. Naturally, the oscilloscope cannot give an idea of the duration of an individual pulse of this train. To measure this important parameter one usually uses the well-known and already mentioned method of two-photon luminescence[23]. However, as shown by Kuznetsova's convincing theoretical investigation[28], the method of two-photon luminescence cannot give a correct idea of the duration and waveform of the individual pulse. Yet knowledge of these quantities is utterly essential for exact measurements in picosecond duration band. It is therefore perfectly possible that the measurement accuracy in the methods considered above is overestimated. This question can be resolved together with the development of a reliable method of measuring the waveform and duration of picosecond pulses.

4. ULTRAFAST OPTICAL SHUTTER AND SOME OF ITS APPLICATIONS

Indirect investigations show that the anisotropy relaxation time obtained by the earlier direct methods is patently overestimated.

A study of the spectrum of depolarized scattering in liquids with anisotropic molecules has indicated that there exist at least two anisotropy relaxation times, which are different for different liquids and lie in the range from 10^{-10} sec to 10^{-13} sec[8]. There was, however, no direct method of measuring such short times.

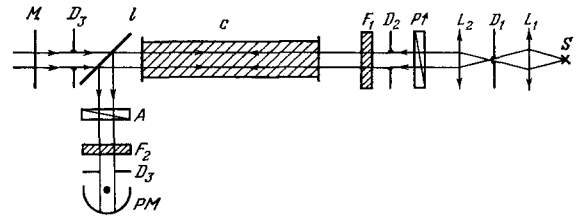

FIG. 8. Diagram of setup for the observation of birefringence produced in a liquid by an intense light pulse [29]. S–source of weak light; L_1, L_2–lenses, P and A–polarizer and analyzer; D_1, D_2, and D_3–diaphragms; C–cell with investigated liquid; *l*–semitransparent plate; PM–photomultiplier; M–output mirror of ruby-laser optical resonator.

The most recent investigations have made it possible to obtain a quantitative estimate of the anisotropy relaxation time by directly measuring this quantity.

Mayer and Gires[29] have proved experimentally that birefringence is produced in a beam of intense light in a cell filled with a liquid with anisotropic molecules.

Their experimental setup is shown in Fig. 8. Light from the source S is transformed into a parallel polarized beam, the electric field vector of which lies in the plane of the figure. The beam passes through a cell and is reflected by plate *l*. If there is no birefringence in the cell, then the light from the source S does not reach the photomultiplier PM, but is stopped by the polarizer A. A Q-switched ruby laser sends a parallel beam from the output mirror M to the right. The electric vector of the light-wave field makes an angle 45° with the plane of the figure. The laser beam subtends the entire beam from the source S and is absorbed in the filter F_1. The birefringence produced by the laser beam opens a path to the photomultiplier for the beam of the source S. The laser pulse duration was 5.5×10^{-8} sec, and the energy was 0.14 J. Under these conditions, birefringence was observed in carbon disulfide, nitrobenzene, chlorobenzene, and benzene. No birefringence was observed in water.

The magnitude of the birefringence is proportional to the laser intensity, and the signal of the photomultiplier PM is proportional to the square of the difference between the refractive indices Δn of the ordinary and extraordinary rays, so long as the corresponding path difference is shorter than the wavelength of the light. For a quantitative determination of the Kerr constant $B(\Delta n = \lambda B E^2)$ the cell C was replaced by a standard cell, and electric pulses of known amplitude were applied to the electrodes of the latter.

In the experiment of[29], the field of the light beam $(\overline{E^2})^{1/2}$ was 39 kV/cm. In carbon disulfide, within the limits of experimental error, such a static field produces an equal birefringence. As expected, for dipoleless molecules the Kerr constant at optical frequencies and in the quasistatic or static regime is the same. This pertains, for example, to carbon disulfide and to benzene, but the Kerr constant differs strongly for such dipolar liquids as nitrobenzene and chlorobenzene. From the point of view of the Langevin theory[8,30], this result can be easily explained. In molecules that have no constant dipole moment but are characterized by a noticeable anisotropy, which is manifest, for example, in the presence of depolarization of the

scattered light, the electric field of the light wave induces a moment that, generally speaking, does not coincide with the direction of the field, as a result of which a force couple is produced and tends to align the molecule's maximum-polarizability axis with the direction of the electric field.

When the sign of the field is reversed, the induced moment is also changed, and the orienting force retains its direction. Such an orientation is produced by the predominent direction of the maximum-polarizability axes along the field, and a gaseous or liquid medium that is isotropic as a whole acquires the properties of a uniaxial crystal. Birefringence is produced in such a medium.

A constant dipole moment, which exerts such a strong influence on the Kerr constant (for example in nitrobenzene), cannot play any role at optical frequencies, owing to the inertia of the molecule, with which the constant dipole moment is rigidly coupled. In such molecules, only anisotropy plays a role at optical frequencies. This is precisely why the Kerr constant in nitrobenzene is approximately the same or even somewhat smaller than in carbon disulfide[30], although in a static or low-frequency field it is larger by 100 times than in carbon disulfide. It is now important to ascertain the minimum operating time of a Kerr cell with a given liquid for use as an optical shutter.

The refractive indices in the directions parallel ($n_{\parallel}$) and perpendicular ($n_{\perp}$) to the direction of the electric field controlling the cell are written as follows[8,31]:

$$n_{\parallel} = n + \frac{2}{3}\lambda B E^2(t) \text{ and } n_{\perp} = n - \frac{1}{2}\lambda B E^2(t),$$

where n is the refractive index in the absence of a field. If $\delta n_{\parallel} = n_{\parallel} - n$ and $\delta n_{\perp} - n$, then

$$\Delta n = \delta n_{\parallel} - \delta n_{\perp} = \lambda B E^2(t). \quad (2)$$

Relation (2) characterizes the stationary case. The time variation obeys the equation[8,31]

$$\frac{d(\Delta n)}{dt} + \frac{1}{\tau}\Delta n = \frac{1}{\tau}\lambda B E^2(t). \quad (3)$$

It follows from (3) that the resultant birefringence, when left alone after turning off the field E(t), decreases exponentially:

$$\Delta n = \text{const} \cdot \exp\{-t/\tau\}. \quad (4)$$

From the last expression it is clear that the attenuation of the birefringence cannot occur in a time shorter than τ. As already stated, τ has the meaning of the anisotropy relaxation time or of the time of orientation of the anisotropic molecules in the electric field. The nature and the minimal values of the birefringence attenuation time in an electric field will be discussed later.

A direct measurement τ in cells filled with carbon disulfide, nitrobenzene, methyl iodide, and dichloroethane was performed by Duguay and Hansen[32] by a new optical method employing picosecond pulses.

Their experimental setup is shown in Fig. 9. A light pulse of power ~100 MW from a Q-switched neodymium laser with mode locking is incident on a KDP-crystal frequency converter. The wavelength is $\lambda = 1.06\ \mu$. The duration of each picosecond pulse is estimated at ~5 psec.

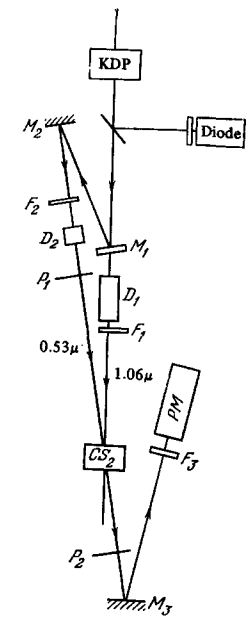

FIG. 9. Setup for the measurement of anisotropy relaxation time in liquids [32]. F_1, F_2, and F_3 – light filters; M_1 – beam-splitting mirror; M_2 and M_3 – mirrors; P_1 and P_2 – crossed polaroids; D_1 and D_2 – delay lines; PM – photomultiplier.

The KDP is mounted in such a way that only ~1% of the light incident on it is transformed into the second harmonic $\lambda = 0.53\ \mu$. The second-harmonic light is purposely made so weak that it cannot produce in the cell any noticeable birefringence or other nonlinear effects, and serves only as a probe.

The pulse of infrared light $\lambda = 1.06\ \mu$ produces birefringence in a cell with a liquid (CS_2), and at the same time, the pulse of green light, the path of which is shown in Fig. 9, can pass through the cell and reach the photomultiplier PM. The delay lines D_1 and D_2 can change the relative time of encounter between the green and infrared pulses in the cell with the liquid. Thus, the character and the growth time of the birefringence in the liquid can be traced by following the PM signal.

The intensity of the light passing through the analyzer is given by the well-known formula

$$I_A = I_0 \sin^2\varphi, \quad (5)$$

where $\varphi = (2\pi/\lambda)\Delta n L$; here L is the path length of the beam in the cell with the liquid, and Δn is determined in the nonstationary case by formula (4). If φ is small, then

$$I_A = \text{const} \cdot (\Delta n)^2. \quad (6)$$

In the scheme of Fig. 9, the photomultiplier signal is proportional to the intensity I_A, and consequently to the square of Δn.

Figure 10 shows the transmission curves of a cell filled either with carbon disulfide or with nitrobenzene[32]. The ordinates represent the logarithm of I_A in arbitrary units, and the abscissas the time of relative delay of the intense and probing pulses. For carbon disulfide, the transmission at the maximum reached 20% in the best experiment[32].

The half-width of the symmetrical curve for carbon disulfide, drawn dashed in Fig. 10, is ~8 psec. It is not advisable to determine the anisotropy relaxation time τ from the character of this curve, since this

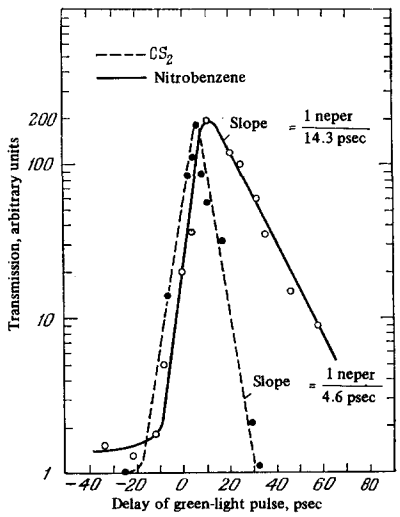

FIG. 10. Dependence of the transmission of an "optical" Kerr cell on the delay time of the green light pulse relative to the control pulse [32].

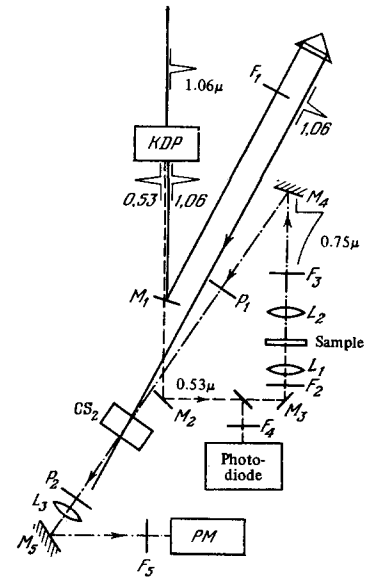

FIG. 11. Setup for the measurement of the luminescence decay time of polymethine dyes with the aid of an ultrafast optical shutter [37]. F_1, F_2, F_3, F_4, and F_5—light filters; P_1 and P_2—crossed polaroids; M_1—beam-splitting mirror; M_2, M_3, M_4, and M_5—mirrors; PM-photomultiplier.

time is known to be small. As already stated, according to indirect determinations of this quantity from the half-width of the intense section of the Rayleigh-line wing[8,33-35] τ lies in the range from 2.4 to 1.8 psec.

On the other hand, in the experiments of[32] the waveform of the light pulse was complicated and unknown, and the duration was known only approximately. The transmission curve as a function of the relative time of arrival of the pulses at the cell with the nitrobenzene is shown in Fig. 10 by the solid curve.

It is easily seen that the result for nitrobenezene differs both qualitatively and quantitatively from that obtained for carbon disulfide. In this case the transmission at the maximum is only 1.4%, and the maximum is shifted towards larger delays. In Fig. 10, the maxima of both curves are made to coincide for convenience in comparison.

The curve for nitrobenzene (Fig. 10) shows an exponential decrease after 20 psec and obeys formula (6) with allowance for formula (4). An important contribution to the course of this curve is made by the anisotropy relaxation time, which is determined from the slope of the exponential section of the curve. An average of several measurements yields $\tau = 32 \pm 6$ psec.

An indirect determination of the anisotropy relaxation time from the narrow section of the Rayleigh-line wing, performed for nitrobenzene by Starunov et al.[36], yielded $\tau = 50$ psec. The results of the direct and indirect determinations of the anisotropy relaxation time can be regarded as being in good agreement.

The obtained anisotropy relaxation times indicate that the birefringence process is due principally to the effect of orientation of the anisotropic molecules in the electric field.

A study of the distribution of the intensity in the Rayleigh line wing has shown[8] that for liquids consisting of anisotropic molecules one can speak of at least two anisotropy relaxation times—a "long" time, which fluctuates for different liquids between 10^{-10} and 10^{-12} sec, and a short relaxation time, $\sim 10^{-13}$ sec or even shorter.

Thus, the orientational anisotropy relaxation times make it possible to study processes of duration to 10^{-13}—10^{-14} sec.

The Kerr effect due to electron polarization makes it possible to reduce the "proper time constant" of the shutter to a value $\sim 10^{-15}$ sec.

Many solids can have considerable birefringence as a result of electronic polarizability in an electric field. The progress in the development of fast optical shutters depends principally on the progress in obtaining the shortest possible intense-light pulses.

A Kerr cell controlled by picosecond light pulses was already used by Duguay and Hansen[37] to study the luminescence kinetics of two polymethine dyes, namely cryptocyanine (1, 1'-diethyl-4, 4'-dicarbocyanine iodide) and DDI (1, 1'-diethyl-2, 2-dicarbocyanine iodide), the fluorescence emission time in which lies in the picosecond range. The experimental setup is shown in Fig. 11. A Q-switched neodymium laser with mode locking emits light pulses $\lambda = 1.06\ \mu$ of duration ~ 8 psec with peak power 800 MW and beam diameter 5 mm. A KDP crystal produces a second harmonic $\lambda = 0.53\ \mu$ with peak power 80 MW. The mirror M_1 causes the 1.06 μ and 0.53 μ beams to take different paths. The infrared light falls on a roof prism, movement of which can change the time of arrival of the 1.06 μ pulse at the cell with CS_2.

The green light, after reflection from mirrors M_1 and M_2, is focused into a vessel 1 mm thick filled with polymethine dye ($\sim 10^{-3}$M). The luminescence emission $\lambda = 0.75\ \mu$ is gathered by lens L_2 and is directed to the cell with CS_2, from which it goes to photomultiplier PM. The light filters F in different parts of the setup cut off the wavelengths that must not pass in certain directions. The photodiode behind the filter F regis-

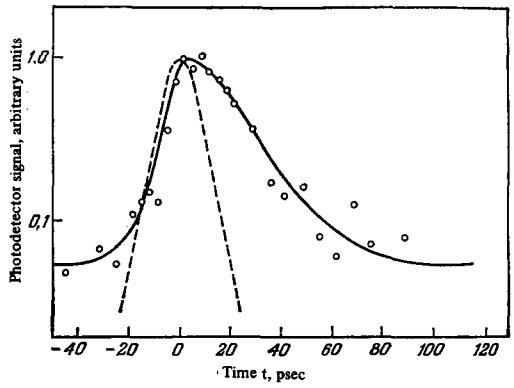

FIG. 12. Plot of photomultiplier signal (Fig. 11) against the relative advance or delay time of the control pulse ($\lambda = 1.06\,\mu$) relative to the pulse of luminescence of DDI in methanol [37] (solid–DDI in methanol, dashed–apparatus function).

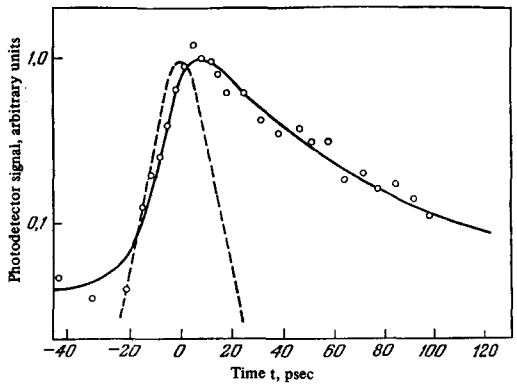

FIG. 13. Photomultiplier signal (Fig. 11) vs. relative advance or delay time of the control pulse ($\lambda = 1.06\,\mu$) relative to the luminescence light of cryptocyanine in methanol [37]. (Solid–cryptocyanine and methanol, dashed–apparatus function).

ters the energy of the green light. The photodiode and photomultiplier signals are registered with a two-beam oscilloscope.

The giant neodymium-laser pulse consisted of approximately 20 picosecond pulses separated by a time interval ~5 nec. In the described experiment, the only pulses chosen were those giving the maximum signal in both oscilloscope channels. There were 4–5 such ultrashort signals at the center of the pulse. To determine the "system response" time or the temporal apparatus function of the setup, the dye solution was replaced by a ground glass, for which the "lifetime in the excited state" is equal to zero. This response determines the time resolution of the apparatus. In Figs. 12 and 13, the apparatus function is shown dashed. In this case, the cell remained open each time for ~10 psec.

The solid curves in Figs. 12 and 13 are the results of computer calculations of the convolution of the apparatus function ("system response") with one exponential or with a sum of two exponential functions. The time necessary for the excitation, the upper limit of which is ~5 psec, is neglected, but account is taken of the dc component, which is clearly seen on the growth side of the curve.

For a solution of DDI in methanol and acetone at 23° (Fig. 12), the experimental data are well described by the convolution of the apparatus function with an exponential function of the type (6) and (4), with a lifetime 14 ± 3 psec.

The data for cryptocyanine cannot be described so unambiguously. In this case one must assume two lifetimes, 10 and 45 psec, with equal weights. This can be regarded as equivalent to assuming that the fluorescence of the cryptocyanine attenuates by a factor e within a time 22 ± 4 psec, if a δ-like form of the excitation function is assumed.

However, the choice of 10 and 45 psec is not unique; the authors of[37] found also a different pair of times, with which the results of the experiment in Fig. 13 can be satisfactorily described. In their opinion, the complex picture of the luminescence attenuation in cryptocyanine can be due to the presence of two or several isomers of cryptocyanine and to different lifetimes at different fluorescence levels.

As already mentioned, the minimum time of opening of an ultrafast optical shutter is approximately 10 psec and consists of two parts: 1) the duration of the picosecond pulse (about 8 psec) and 2) the anisotropy relaxation time in the cell with carbon disulfide (approximately 2 psec).

Thus, almost everything is determined by the duration of the pulse with the intense light, and in view of the possibility of working with short relaxation times in the interval 10^{-13}–10^{-15} sec, as mentioned above, the operating time of the optical shutter is determined for the time being only by the duration of the light pulse controlling the cell.

Progress in the technique of compression of picosecond pulses and other possible methods of shortening the duration of light pulses will apparently make it possible in the near future to increase the shutter operating speed appreciably.

5. USE OF PULSED LIGHT SOURCES IN THE SHADOW METHOD (TOEPLER METHOD)

The use of a series of light flashes in the shadow (Toepler) method has been known for a long time[38], but the shortest exposures, using all possible tricks, were of the order of 10^{-9} sec[39].

Q-switched lasers with mode locking have uncovered entirely new possibilities also in this field of optical research.

An example of the use of such light sources is the research of Alcock et al.[40]. They investigated the kinetics of the development of a spark obtained by focusing a ruby-laser light pulse of duration from 14 to 30 nsec and power from 35 to 3000 MW. The experimental setup is shown in Fig. 14. It consists of two lasers, a ruby laser (RL) in which the Q-switch is a Pockels cell, and whose characteristics were indicated above, and a Q-switched neodymium laser with mode locking (MLL). The pulse duration ranged from 0.4 to to 1 msec, with a total energy 0.4 J. The pulse consists of a train of picosecond pulses, separated by time intervals 4–6 nsec, with an individual-pulse duration ~5 psec (determined by the two-photon luminescence method). The entire pulse is investigated with an oscilloscope having a time resolution 0.5 nsec. The MLL

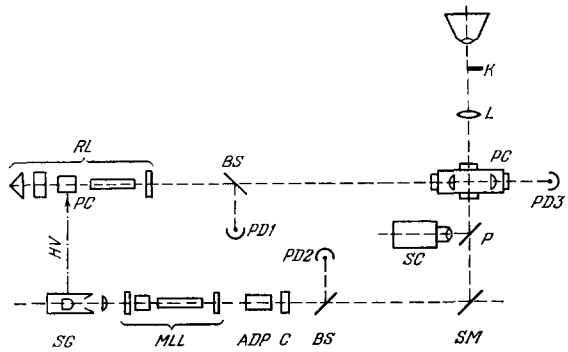

FIG. 14. Setup for the study of the development of a plasma in the focus of a laser pulse by the shadow method (Toepler method). MLL—mode-locking Q-switched laser; RL—ruby laser; PC—Pockels cell; SG—Spark gap, ADP—second-harmonic generator, C—copper-sulfate filter; BS—beam splitter; SM—silvered mirror; PD3—photodetector; L—lens; K—knife; P—plate; SC—camera; PD1 and PD2—photodiodes.

laser radiates forward. An ADP crystal generates the second harmonic, which is then used as a weak probing light passing through the vessel PC, which is filled with different gases. The same laser also radiates in the backward direction, thereby igniting a spark gap that controls the voltage on the Pockels cell and thus determines the instant of generation of the ruby laser RL.

On the path of the beam containing the train of picosecond pulses, after this beam passes through the cell PC with the investigated gas, the lens L focuses the image of the location of the focus of the laser RL on the edge of the knife K. After the plasma is produced in the vessel, it is eliminated by a train of pulses of 5 psec duration, following each other at intervals of 5.5 nsec. A photographic camera registers the picture of the series of "frozen" fronts of the spreading plasma. These pictures make it possible to determine the rates of longitudinal and transverse expansion of the plasma as a function of the time, the dependence of the expansion rate on the pressure in different gases, the plasma temperature, and a number of other important characteristics of the plasma. Of course, such a method is suitable not only for the study of plasma, and is much more universal.

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Translated by J. G. Adashko