PHYSICAL REVIEW A VOLUME 12, NUMBER 3 SEPTEMBER 1975

Optical Kerr effect induced by ultrashort laser pulses

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(Received 3 December 1974)

A generalization of the classical molecular-optics theory of the optical Kerr effect (OKE) is presented which permits a study of the effect when induced by ultrashort laser pulses. Explicit relationships between results from this general theory and often-used phenomenological equations governing the temporal behavior of the OKE are established. Numerical results are presented for the liquids carbon disulfide and nitrobenzene, illustrating in detail the temporal nature and intensity of the birefringence induced in these liquids by ultrashort laser pulses. The importance of these results is discussed in connection with a number of self-induced nonlinear phenomena arising from the OKE as well as in conjunction with the OKE shutter switched by an ultrashort laser pulse. In addition, a discussion is given of the nature of the birefringence induced by picosecond laser pulses as well as aspects of the extension of OKE photography to the subpicosecond domain.

I. INTRODUCTION

The optical Kerr effect (OKE) was first experimentally reported in 1964, independently by Mayer and Gires,[1] and Maker, Terhune, and Savage.[2] However, it was not until the introduction of mode-locked lasers, capable of producing high-intensity coherent-light pulses of picosecond duration, that this effect found wide application to the ultrafast diagnosis of a number of phenomena. These include the measurement of picosecond fluorescent lifetimes and molecular relaxation times,[3-10] the measurement of picosecond pulsewidths,[11-17] optical pulse compression,[18-20, 28] mode locking of lasers,[21-26] nonlinear intensity-dependent discriminators,[17, 27, 28] picosecond time-resolved emission and absorption spectroscopy,[5, 6, 29-31] the temporal measurement of short CO_2 pulses,[32-34] the picosecond framing photography of laser-produced plasmas,[35, 36] and picosecond time-resolved laser emission spectroscopy.[37] In particular, the use of the OKE induced by mode-locked laser pulses in configurations which provide a high-speed optical gate or photographic shutter show considerable promise in extending the range of ultrafast two-dimensional photography.[11, 12, 14, 35-37] For although significant advances have been made in time-resolved one-dimensional photography with the design of image-converter streak cameras[38-41] capable of temporal resolutions ~1 psec,[40, 41] the use of electro-optic techniques appears to be severely limited in permitting two-dimensional photography in this range.

This approach, be it with the aid of Pockels or Kerr cell optical shutters, or with the use of gated-image intensifiers, is primarily limited by the intrinsic response of these devices to fast high-voltage pulses.[42-46] Typically, maximum temporal resolutions of several hundred psec are recorded. Fortunately, the OKE shutter circumvents these limitations and offers a relatively simple approach to picosecond and subpicosecond two-dimensional photography.

However, in using an OKE shutter for high-speed photographic analysis, particular consideration must be given to its transmission characteristics. These are strongly dependent both on the type of optical configuration used in the photographic system and on the characteristics of the ultrashort optical pulses used to gate the shutter. Before this can be carefully attempted it is necessary to have a thorough knowledge of the response of the Kerr-active medium to the ultrashort pulse.

It is the intent of this paper to analyze in detail the temporal and spatial nature of the birefringence induced in such media by ultrashort laser pulses. This is achieved by a generalization of a classical molecular optics theory of the OKE to determine the response of the birefringence to laser pulses of arbitrary temporal and spatial character. From this analysis, the importance of delineating the relative effects of the molecular orientational and electronic contributions to the induced birefringence is clearly illustrated, and their relative effects for particular pulse characteristics defined. This has led to a firmer understanding of the temporal and spatial nature of the birefringence induced in commonly used liquids (such as carbon disulfide and nitrobenzene), and also to the proposal that under suitable conditions, they may be used in OKE photographic schemes providing temporal resolutions in the subpicosecond range (10^{-12}–10^{-14} sec). Thus this analysis should aid in understanding the nature and possible limitations peculiar to most applications of the OKE, particularly to its use in ultrafast two-dimensional photography; in addition, it should be relevant to the study of many self-induced nonlinear phenomena arising in part or in whole from the OKE.

II. THEORY OF THE OPTICAL KERR EFFECT INDUCED BY ULTRASHORT LASER PULSES

The most fundamental approach to a study of optically induced birefringence is through the use of classical molecular-optics theory in conjunction with nonequilibrium statistical fluid mechanics. This was first applied to the Kerr and optical Kerr effects by Buckingham,[47-49] who made the first explicit prediction of the optical Kerr effect in 1956,[49] and subsequently by Kielich,[50-55] who enlarged and developed the theory in a number of papers. Both of these authors derived expressions for the various optical Kerr coefficients in terms of liquid and molecular parameters, and found expressions for the induced birefringence in the case of a plane-wave optical field.[49,50]

In the present paper, this molecular-optics theory of the OKE has been extended to include the more general case of an optical-pulse envelope with an arbitrary spatial and temporal dependence. The mainstaff of the method described by Buckingham and Kielich is the adoption of a relaxation-diffusion equation originally proposed by Debye[56] which governs the spatial and temporal behavior of a nonequilibrium molecular distribution function.[50] The form of the induced birefringence is found directly from this distribution function. By selecting a distribution function of known form but with undetermined coefficients, the Debye relaxation-diffusion equation is solved to determine these unknown coefficients and hence find the induced birefringence.

To generalize this approach to include the case of a short-duration optical pulse, an essentially iterative method was used. Beginning with the known solution for a plane-wave optical field,[50] the constant coefficients of this solution were replaced with expressions incorporating the assumed pulse-envelope model and a new set of variable but undetermined coefficients. Then, using an expression for the molecular-field interaction potential[49,50] corrected to correspond to the plane-wave solution, the relaxation-diffusion equation was applied a second time to solve for this new set of coefficients.[35] This method was straightforward and general, and readily yielded solutions for a variety of assumed pulse models.

Specifically, in the present paper, the model adopted for the optical pulse is a three-dimensional Gaussian pulse described by

$$E_\omega^2(\vec{r},t)=2E_0^2\exp[-A_x^2(x-vt)^2-A_y^2y^2-A_z^2z^2]\times\cos^2[\omega(t-x/v)]\,, \tag{1}$$

where A_x, A_y, and A_z are arbitrary parameters determining the spatial and temporal variation in the pulse envelope. The pulse travels in the positive x-axis direction with a (group) velocity $v=c/n_\omega$. The factor of 2 appearing in Eq. (1) is included for convenience so that the time-averaged optical pulse envelope is

$$\overline{E_\omega^2}(\vec{r},t)=E_0^2\exp[-A_x^2(x-vt)^2-A_y^2y^2-A_z^2z^2]\,. \tag{2}$$

The full-width-at-half-maximum (FWHM) duration of the pulse described by Eqs. (1) and (2) is given by $T_\omega=2(\ln 2)^{1/2}/A_xv$, and it is assumed that the pulse envelope varies slowly relative to the period of the optical electric field, i.e., $T_\omega\gg 2\pi/\omega$.

For the Gaussian optical pulse, the iterative molecular-optics theory yields[35] the refractive indices $n_\parallel$ and $n_\perp$ for, respectively, polarizations parallel and perpendicular to $\vec{E}_\omega$:

$$n_\parallel=n_0+\delta n_\parallel$$

$$=n_0+\tfrac{3}{2}n_2^eE_\omega^2(\vec{r},t)+n_2^o\,\frac{\pi^{1/2}}{3A_xv\tau_o}\,\overline{E_\omega^2}(\vec{r},t)\exp\left(A_x^2(x-vt)^2+\frac{x-vt}{v\tau_o}+\frac{1}{4A_x^2v^2\tau_o^2}\right)\operatorname{erfc}\left(A_x(x-vt)+\frac{1}{2A_xv\tau_o}\right) \tag{3}$$

and

$$n_\perp=n_0+\delta n_\perp$$

$$=n_0+\tfrac{1}{2}n_2^eE_\omega^2(\vec{r},t)-n_2^o\,\frac{\pi^{1/2}}{6A_xv\tau_o}\,\overline{E_\omega^2}(\vec{r},t)\exp\left(A_x^2(x-vt)^2+\frac{x-vt}{v\tau_o}+\frac{1}{4A_x^2v^2\tau_o^2}\right)\operatorname{erfc}\left(A_x(x-vt)+\frac{1}{2A_xv\tau_o}\right)\,, \tag{4}$$

where n_0 is the linear refractive index at the angular optical frequency ω' of the "probe" or "test" beam, and τ_o is the mean orientational relaxation time of the Kerr medium. The induced birefringence $\Delta n=n_\parallel-n_\perp$ is given by

$$\Delta n=\Delta n^o+\Delta n^e\,, \tag{5}$$

where the orientational and electronic contributions to the induced birefringence are, respectively,

$$\Delta n^o = n_2^o \, \frac{\pi^{1/2}}{2A_x v\tau_o} \, \overline{E_\omega^2}(\vec{\mathrm{r}}, t) \exp\left(A_x^2 (x-vt)^2 + \frac{x-vt}{v\tau_o} + \frac{1}{4A_x^2 v^2 \tau_o^2}\right) \operatorname{erfc}\left(A_x (x-vt) + \frac{1}{2A_x v\tau_o}\right) \tag{6}$$

and

$$\Delta n^e = n_2^e E_\omega^2(\vec{\mathrm{r}}, t) = n_2^e \overline{E_\omega^2}(\vec{\mathrm{r}}, t)[1 + \cos 2\omega(t - x/v)] . \tag{7}$$

The contribution to the total nonlinear refractive index $n_2 = n_2^o + n_2^e$ resulting from the electronic hyperpolarizability[48, 49, 55] is readily found from the molecular optics theory to be[48-55]

$$n_2^e = \frac{2\pi\rho}{3n_0} \gamma \left(\frac{n_0^2+2}{3}\right)^2 \left(\frac{n_\omega^2+2}{3}\right)^2 , \tag{8}$$

where ρ is the number density of the Kerr medium, and $\gamma = \frac{1}{5}\gamma_{ii\,jj}^{\omega,\,\omega'}$ $(i, j = 1, 2, 3)$ where $\gamma_{ijkl}^{\omega,\,\omega'}$ is a component of the fourth-rank tensor describing the electronic (second) hyperpolarizability at the angular frequency ω' of the probe light induced[47, 49, 50, 55] by the optical field of angular frequency ω. The orientational contribution n_2^o to the nonlinear refractive index n_2 resulting from an anisotropy in the molecular polarizability is given by[48-55]

$$n_2^o = \frac{2\pi\rho}{15kTn_0} \delta^2 \left(\frac{n_0^2+2}{3}\right)^2 \left(\frac{n_\omega^2+2}{3}\right)^2 , \tag{9}$$

where k is Boltzmann's constant, T is the temperature of the medium in °K, and $\delta^2 = \frac{1}{4}(\alpha_{ii}^{\omega'} - \alpha_{jj}^{\omega'}) \times (\alpha_{ii}^{\omega} - \alpha_{jj}^{\omega})$ $(i, j = 1, 2, 3)$ is a measure of the anisotropy in the molecular polarizabilities $\alpha_{ij}^{\omega'}$ and α_{ij}^{ω} at frequencies ω' and ω, respectively. In the definitions of γ and δ^2, the tensor indices i, j refer to the principal axes of the Kerr-medium molecules so that, for example, $\alpha_{ij} = \alpha_i \delta_{ij}$, with similar simplifying relations among the γ_{ijkl} components.[47, 55]

The above expressions for the electronic Kerr coefficient n_2^e and the orientational Kerr coefficient n_2^o were derived from a model which assumed complete independence for the individual molecules composing the Kerr medium.[50] While this assumption is acceptable for the electronic effect in both gases and liquids, it is invalid for the orientational effect, particularly in the case of liquids. Values of n_2^o calculated from Eq. (9) are generally considerably larger than experimentally known values[57] of n_2. It is thus necessary to reexamine the derivation of n_2^o, taking into account the effect of multiple molecular interactions on the orientating influence of an applied electric field. The most important interaction in a medium composed of anisotropic molecules is that of angular correlation between members of pairs of molecules.[48-55, 58-60] Because of the permanent and/or induced dipole moments in the molecules, a given molecule is no longer completely free to align itself (i.e., its principal dipole axis) parallel to the applied electric field, but is hindered in doing so by its tendency to simultaneously align itself with nearest-neighbor molecules which, in general, have various different (though not random) orientations. Hence the net effect of binary angular correlations among the molecules is to reduce the effective value of the orientational Kerr coefficient n_2^o to a first-order corrected coefficient, ${}_c n_2^o$, given by[50]

$${}_c n_2^o = n_2^o (1 + J_a) , \tag{10}$$

where J_a is the integral parameter

$$J_a = \frac{\rho}{2V\delta^2} \sum_{s,\,t=1}^{3} \alpha_s^\omega \alpha_t^{\omega'} \int\int (3\cos^2\theta_{st}^{pq} - 1) \times g^2(\Omega_p, \Omega_q)\, d\Omega_p \, d\Omega_q , \tag{11}$$

giving a measure of the degree of angular correlation for pairs of molecules. Here, θ_{st}^{pq} denotes the angle between the principal axes s and t of molecules p and q, $g^2(\Omega_p, \Omega_q)$ is the correlation function for pairs of molecules having orientations specified by Ω_q and Ω_p, V is the volume of the medium, and the integration is over the entire range of the set of angular coordinates symbolized by Ω_q and Ω_p. In practice, calculations of J_a using Eq. (11) are very approximate, since the exact form of the correlation function $g^2(\Omega_q, \Omega_p)$ is ambiguous. However, the same parameter J_a also appears in the expression for the depolarization of scattered light D as[61]

$$D = \frac{6\delta^2(1 + J_a)}{45\rho kT\beta_T(\alpha^\omega)^2 + 7\delta^2(1 + J_a)} , \tag{12}$$

where $\alpha^\omega = \frac{1}{3}(\alpha_1^\omega + \alpha_2^\omega + \alpha_3^\omega)$ is the mean polarizability and β_T is the isothermal compressibility of the medium. Solving for J_a and substituting into Eq. (10) gives

$${}_c n_2^o = \frac{6\pi\rho^2(\alpha^\omega)^2\beta_T}{n_0} \, \frac{D}{6 - 7D} \left(\frac{n_0^2+2}{3}\right)^2 \left(\frac{n_\omega^2+2}{3}\right)^2 . \tag{13}$$

In those cases where a value for α^ω is unknown but δ^2 is known, the Lorentz-Lorenz equation may be used to arrive at an alternative but more approximate expression for ${}_c n_2^o$ given by

$$ {}_c n_2^o \simeq \frac{3\beta_T}{8\pi n_0}\,\frac{D}{6-7D}\left(\frac{n_0^2+2}{3}\right)^2 (n_\omega^2-1)^2 . \tag{14}$$

The usefulness of this last expression is that, apart from the refractive indices n_0 and n_ω, only a knowledge of β_T and D are required in order to calculate a reasonable estimate of the orientational optical Kerr coefficient. With $n_2 = {}_c n_2^o + n_2^e$, the ratio n_2^e/n_2 is given from Eqs. (8) and (13) by the expression

$$ \frac{n_2^e}{n_2} = \left(1 + \frac{9\rho(\alpha^\omega)^2\beta_T}{\gamma}\,\frac{D}{6-7D}\right)^{-1} . \tag{15}$$

For those liquids for which a value of γ is known (see the review by Kielich[55]), values of n_2 calculated using Eqs. (8) and (13) consistently give better than 10% agreement with experimentally measured values[62, 63] of n_2. Such agreement lends strong support to at least the first-order correctness of the above expressions for ${}_c n_2^o$ and n_2^e. In addition, the calculations reveal that the contribution made by n_2^e to n_2 is generally considerable, lying in the range 0.1–0.3 for all the liquids studied. Specifically, using Eq. (15) with experimentally determined γ values,[55] the ratio n_2^e/n_2 is found to be 0.193 in carbon disulfide (CS_2), 0.130 in benzene (C_6H_6), 0.143 in cyclohexane (C_6H_{12}), 0.181 in toluene ($C_6H_5CH_3$), and 0.191 in chloroform ($CHCl_3$), while in nitrobenzene ($C_6H_6NO_2$), $n_2^e/n_2 = 0.262$. This finding that the contribution of n_2^e is generally substantial concurs with that expressed by Brewer and Lee,[64] Brewer and McLean,[65] and Alfano and Shapiro,[66-68] from their work on the self-phase-modulation spectra of picosecond pulses. Saikan and Takima[69] have concluded that the contribution of n_2^e to n_2 is considerable from studies of the self-trapping of picosecond pulses, and Topp and Rentzepis[70] have arrived at the same conclusion from studies on the transmission characteristics of an OKE gate.

Substituting ${}_c n_2^o$ in place of n_2^o in Eq. (6), it is found that the orientational part Δn^o of the induced birefringence obeys the partial differential equation

$$ \tau_o \frac{\partial(\Delta n^o)}{\partial t} + \Delta n^o = {}_c n_2^o \overline{E_\omega^2}(\vec{r}, t) . \tag{16}$$

In addition, provided that the condition $\omega\tau_e \gg 1$ is met, then the electronic contribution Δn^e to the induced birefringence [Eq. (7)] satisfies a similar equation,

$$ \tau_e \frac{\partial(\Delta n^e)}{\partial t} + \Delta n^e = n_2^e E_\omega^2(\vec{r}, t) , \tag{17}$$

where τ_e is the relaxation time for the electronic nonlinearity (approximately equal to the inverse of the ultraviolet cutoff frequency of the medium). Moreover, since Eq. (17) yields physically acceptable and intuitively expected results even when the condition $\omega\tau_e \gg 1$ is not met, this equation may be considered valid for arbitrary ω. Estimates of τ_e in most media[11, 12, 17, 64] lie in the range $10^{-15} - 10^{-16}$ sec, an upper limit for τ_e in several media of 1.5×10^{-14} sec having been deduced experimentally.[68] Values of τ_o in most media,[71, 72] on the other hand, are $\gtrsim 10^{-12}$ sec. The distinction between these two radically different response times τ_o and τ_e is reflected in the appearance of the time-averaged field-squared $\overline{E_\omega^2}(\vec{r}, t)$ in Eq. (16) governing the orientational birefringence Δn^o and the need to use the instantaneous field-squared $E_\omega^2(\vec{r}, t)$ in Eq. (17) governing the electronic birefringence Δn^e. Equations (16) and (17) have the integral solutions

$$ \Delta n^o(\vec{r}, t) = \frac{{}_c n_2^o}{\tau_o}\int_{-\infty}^{t} \overline{E_\omega^2}(\vec{r}, t'')\exp\left(\frac{-(t-t'')}{\tau_o}\right) dt'' ,$$

$$ \Delta n^e(\vec{r}, t) = \frac{n_2^e}{\tau_e}\int_{-\infty}^{t} E_\omega^2(\vec{r}, t'')\exp\left(\frac{-(t-t'')}{\tau_e}\right) dt'' . \tag{18}$$

Using the generalized molecular-optics approach described above, the induced birefringence was calculated using a number of pulse models besides the Gaussian model of Eq. (1). These included a triangular pulse, a hyperbolic sech-squared pulse, a Lorentzian pulse, and others. In all of these cases, the final solutions for Δn^o and Δn^e were found to obey Eqs. (16) and (17), respectively. From this it is surmised that these equations as written are valid in general for an arbitrary pulse shape.

The derivation of Eqs. (16) and (17) from the Buckingham-Kielich molecular-optics theory provides, to the authors' knowledge, the first real link between this very fundamental approach to the OKE and the often-used phenomenological equation found in the literature and generally written[3, 4, 11, 12, 14, 17, 19, 25-27]

$$ \tau_o \frac{\partial(\Delta n)}{\partial t} + \Delta n = n_2 \overline{E_\omega^2}(\vec{r}, t) \tag{19}$$

or in an equivalent integral form similar to that of Eq. (18). Equation (19) is quite similar in form to Eqs. (16) and (17), with the important exception that Eq. (19) lumps together the orientational and electronic contributions and regards the total birefringence Δn as governed by a single relaxation equation characterized by the response time τ_o. Thus, this latter equation fails to distinguish between the two separate contributions, Δn^o and Δn^e, with their radically different response times, τ_o and τ_e, and consequently could give rise to two potentially serious errors. The first stems from the fact that Eq. (19), as written, ignores the fast-time variation $[\propto E_\omega^2(\vec{r}, t)]$ inherent in the contribution from Δn^e. In certain phenomena, such as self-phase-modulation, the role played by this fast electronic contribution can be dominant even though the ratio $n_2^e / {}_c n_2^o$ is small.[68] The second error implicit in Eq. (19) concerns the magnitude of the induced birefringence Δn when the pulsewidth T_ω is $\lesssim \tau_o$. Under these conditions, the orientational effect becomes damped out, so that the electronic effect, constant in intensity as long as $T_\omega > \tau_e$ (where τ_e is $\ll \tau_o$), becomes increasingly important as T_ω decreases, and eventually becomes the dominant effect when $T_\omega \ll \tau_o$. This behavior, predicted by Eqs. (6) and (7) [or Eqs. (16) and (17)], but not by Eq. (19), will be explicitly shown by the numerical results given in the following section. In those applications where the fast-time variation of Δn^e may be ignored, a time-averaged Δn^e, given by $\Delta n^e = n_2^e \overline{E_\omega^2}(\vec{r}, t)$ may be used. In the limit $T_\omega \gg \tau_o$, Eq. (6) simplifies to $\Delta n^o = {}_c n_2^o \overline{E_\omega^2}(\vec{r}, t)$ so that the total birefringence is simply

$$\Delta n = ({}_c n_2^o + n_2^e) \overline{E_\omega^2}(\vec{r}, t) = n_2 \overline{E_\omega^2}(\vec{r}, t) . \tag{20}$$

Only under the above two conditions is this equation valid. For example, it could be applied to describe the phenomenon of self-focusing or self-trapping in liquids by nanosecond-duration Q-switched laser pulses, but would be incorrect for most liquids when picosecond or subpicosecond pulses were used.

A final consideration concerns the behavior of $n_\parallel$ or $\delta n_\parallel$ separately from that of Δn. For many self-induced OKE phenomena, such as self-focusing and self-phase-modulation, the behavior of $n_\parallel$ rather than the birefringence Δn is principally of concern. The molecular-optics theory provides a specific solution for $n_\parallel$ [Eq. (3)], while the phenomenological approach represented by Eq. (19) cannot specify $n_\parallel$ separately without making further simplifying assumptions about the relationship between $\delta n_\parallel$ and $\delta n_\perp$. This latter equation is tantamount to assuming an orientational effect only ($n_2^e = 0$) from which the relationships $\delta n_\parallel = -2\delta n_\perp = \frac{2}{3}\Delta n$ follow. However, examination of Eqs. (3), (6), and (7) shows that, in the limit $T_\omega \gg \tau_o$, the total birefringence Δn depends on ${}_c n_2^o$ and n_2^e exactly as given in Eq. (20), while

$$\delta n_\parallel = (\tfrac{2}{3} {}_c n_2^o + \tfrac{3}{2} n_2^e) \overline{E_\omega^2}(\vec{r}, t) \tag{21}$$

in this limit. Thus, the role of the electronic Kerr effect is enhanced in the case of $\delta n_\parallel$ alone, since the ratio of the electronic contribution to the orientational contribution is 2.25 times larger for $\delta n_\parallel$ than it is for Δn. This picture differs distinctly from that offered in connection with Eq. (19) and the assumption $\delta n_\parallel = \frac{2}{3}\Delta n$, because this relationship predicts equal ratios of electronic to orientational contributions for both $\delta n_\parallel$ and Δn. Using CS_2 as an example, Eqs. (20) and (21) predict that the electronic contribution to the birefringence Δn is $\simeq 19\%$ (for $T_\omega \gg \tau_o$) while for $\delta n_\parallel$ the electronic contribution is $\simeq 35\%$. Both of these percentage figures will increase when the pulsewidth T_ω is $\lesssim \tau_o$.

III. NUMERICAL MODEL OF THE OKE INDUCED BY ULTRASHORT LASER PULSES

Comparing Eqs. (6) and (7) to Eq. (1), it is seen that the y and z dependence of $\Delta n(\vec{r}, t)$ is identical to that of the optical pulse $E_\omega^2(\vec{r}, t)$ while both $\Delta n(\vec{r}, t)$ and $E_\omega^2(\vec{r}, t)$ depend upon the variables x and t only in the form $(x - vt)$. Thus, for the purposes of studying the form of the induced birefringence, x, y, and z may be set to 0 throughout the calculations with the implicit knowledge that the spatial dependence may be recovered at any point by simply multiplying by $\exp(-A_y^2 y^2 - A_z^2 z^2)$ and replacing t by $(t - x/v)$. Defining the dimensionless parameters

$$\tau_o' \equiv \tau_o / T_\omega , \quad t' \equiv t / T_\omega , \quad s \equiv t'' / T_\omega , \tag{22}$$

Eqs. (7), (6), and (18) may then be simplified to

$$\frac{\Delta n^e}{n_2 E_0^2} = \frac{n_2^e}{n_2} \exp[-K^2 t'^2] , \tag{23}$$

$$\frac{\Delta n^o}{n_2 E_0^2} = \frac{{}_c n_2^o}{n_2} \frac{\pi^{1/2}}{2K} \frac{1}{\tau_o'} \exp\left(-\frac{t'}{\tau_o'} + \frac{1}{(2K\tau_o')^2}\right) \times \operatorname{erfc}\left(-Kt' + \frac{1}{2K\tau_o'}\right) , \tag{24}$$

and

$$\frac{\Delta n^o}{n_2 E_0} = \frac{{}_c n_2^o}{n_2} \frac{1}{\tau_o'} \int_{-\infty}^{t'} \exp\left[-K^2 s^2 - \left(\frac{t' - s}{\tau_o'}\right)\right] ds , \tag{25}$$

where K is the constant $K = [4 \ln 2]^{1/2}$, and where the time-averaged expression for Δn^e has been used. The quantities Δn, Δn^o, and Δn^e were calculated in units of $n_2 E_0^2$ for a number of different liquids. Figures 1 and 2 show the results of the calculations for several τ_o' values in the liquids carbon disulfide (CS_2) and nitrobenzene ($C_6H_5NO_2$). In these figures, the derived values of ${}_c n_2^o / n_2 = 0.807$ and $n_2^e / n_2 = 0.193$ in CS_2, and ${}_c n_2^o / n_2 = 0.738$ and $n_2^e / n_2 = 0.262$ in $C_6H_5NO_2$ were used. For small values of τ_o', Eq. (25) was used, while for larger τ_o', it proved more practical to use Eq. (24) and adopt a rational approximation for the erfc function.[73]

Figures 1 and 2 illustrate a number of important characteristics of the behavior of the induced birefringence Δn with increasing values of τ_o', i.e., with values of T_ω decreasing below τ_o. The most obvious feature is the decreasing contribution made by the orientational birefringence Δn^o as τ_o' increases. The peak value of Δn decreases from its maximum value of $n_2 E_0^2$ at $\tau_o' = 0$ to the limit $n_2^e E_0^2$ when $T_\omega \ll \tau_o$ (but $T_\omega > \tau_e$). Figure 3 summarizes this particular feature by plotting the values of $\Delta n_{\max}/n_2 E_0^2$ versus τ_o' for both liquids CS_2 and $C_6H_5NO_2$. The convergence of $\Delta n \rightarrow \Delta n^e$ is noticeably quicker in the case of nitrobenzene than for carbon disulfide, since the electronic contribution to the induced birefringence, measured by the ratio n_2^e/n_2, is considerably larger in $C_6H_5NO_2$

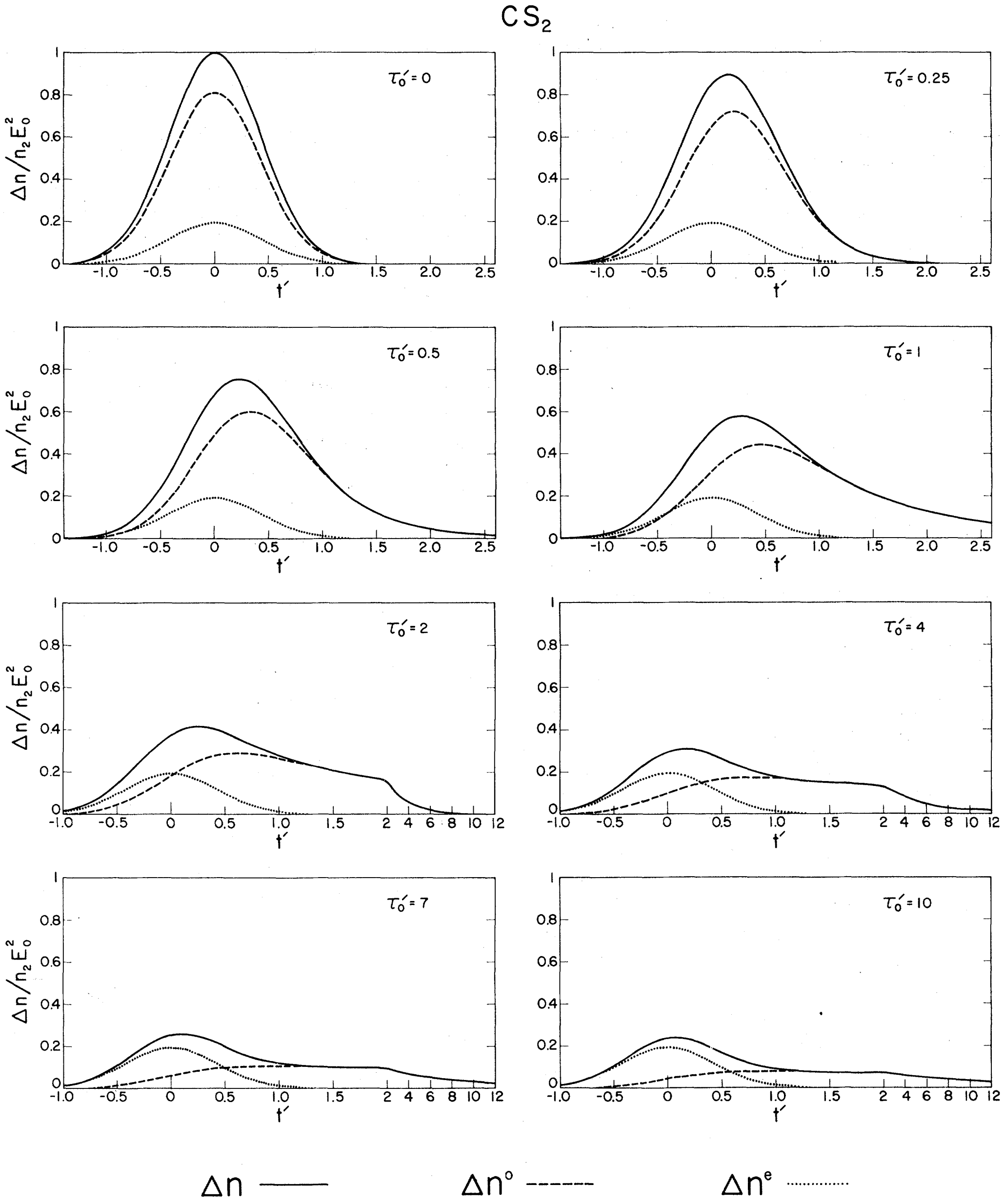

FIG. 1. Induced birefringence in liquid CS_2 for selected pulsewidths ranging from $T_\omega >> \tau_o$ to $T_\omega = 0.1\tau_o$. Each set of curves shows the birefringence Δn (—) and the separate contributions of Δn^o (---) and Δn^e (···). Note the change of scale in the last four figures.

than in CS_2. Since τ_o in CS_2 is $\simeq 2$ psec,[10, 71, 72, 74] while in $C_6H_5NO_2$ $\tau_o \simeq 32$ psec,[3] it should be noted that, in comparing the curves in Fig. 1 to those in Fig. 2 for a given pulsewidth, the corresponding τ_o' values will be very different. For example, for a pulse duration of $\simeq 4$ psec, the induced birefringence in CS_2 would correspond approximately to the curve marked $\tau_o' = 0.50$ in Fig. 1, while the induced birefringence in nitrobenzene would be approximately that shown in the curve marked $\tau_o' = 7$ in Fig. 2. Thus, in this example, the major contribution to Δn in CS_2 comes from the orientational effect, while in $C_6H_5NO_2$ the orientational effect is largely damped out and the electronic effect dominates.

A second important characteristic of the induced birefringence which emerges from Figs. 1 and 2 is the existence of a time lag between the peak of

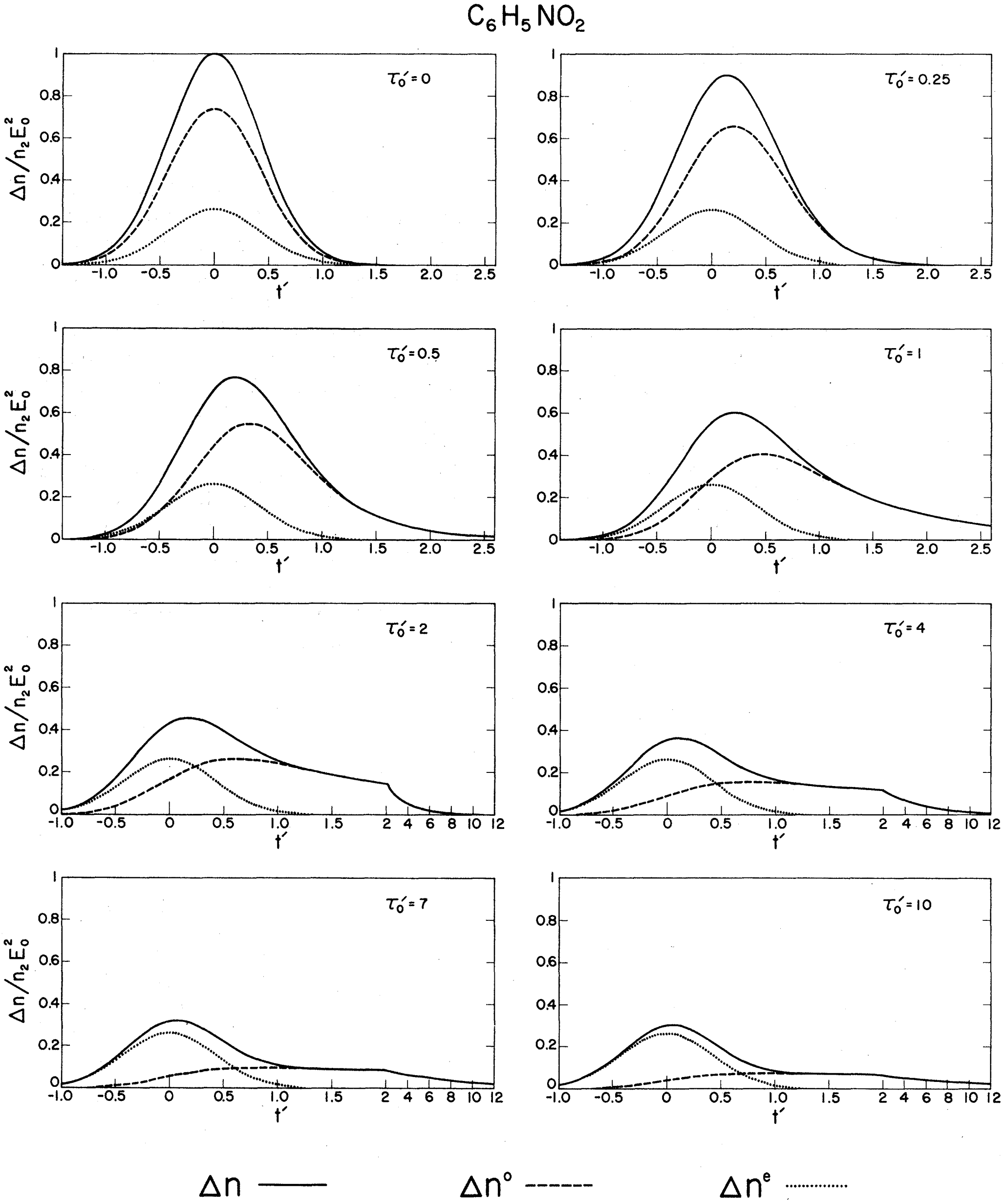

FIG. 2. Induced birefringence in liquid $C_6H_5NO_2$ for selected pulsewidths ranging from $T_\omega >> \tau_o$ to $T_\omega = 0.1\tau_o$. Each set of curves shows the birefringence Δn (—) and the separate contributions Δn^o (---) and Δn^e (···). Note the change of scale in the last four figures.

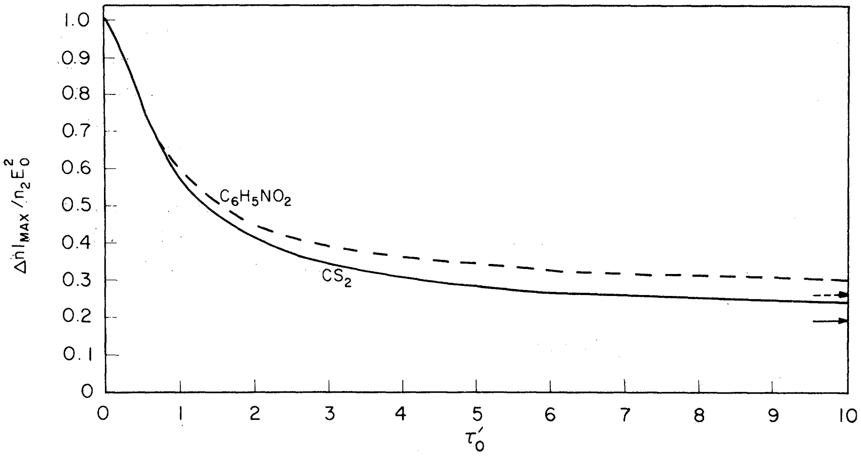

FIG. 3. Peak value of the induced birefringence versus the parameter $\tau_0' = \tau_0/T_\omega$ in the liquids CS_2 and $C_6H_5NO_2$. The small right-hand markers indicate the limit of $\Delta n|_{\max}$ for $\tau_0' \gg 1$ for carbon disulfide (→) and nitrobenzene (--→).

the optical pulse (by definition always at $t' = 0$) and the peak value of the induced birefringence. Defining $t_m' \equiv t_m/T_\omega$ as the time at which Δn is a maximum, $\Delta n(t_m') = \Delta n|_{\max}$, it may be seen from Figs. 1 and 2 that t_m' is initially zero when $\tau_0' = 0$, increases to a maximum as τ_0' increases, and then returns to zero in the limit of $\tau_0' \gg 1$. This behavior of t_m' for carbon disulfide and nitrobenzene is shown explicitly in Fig. 4. The fact that, for a given τ_0' value, the value of t_m' in $C_6H_5NO_2$ is always less than t_m' in CS_2, arises from the relatively larger orientational contribution in CS_2 (${}_cn_2^0/n_2 = 0.807$) than in $C_6H_5NO_2$ (${}_cn_2^0/n_2 = 0.738$). The existence of this time lag t_m' has important consequences in the understanding of the phenomena of self-focusing and self-trapping[75, 76] and the self-steepening[77] of picosecond light pulses.

A third characteristic observed from Figs. 1 and 2 is the behavior of the duration of the induced birefringence versus the parameter τ_0'. Defining the dimensionless parameters $\Delta' \equiv$ (FWHM of Δn)/T_ω, and $\Delta_0' \equiv$ (FWHM of Δn^0)/T_ω, one observes in Figs. 1 and 2 that initially $\Delta' = 1$ when $\tau_0' = 0$, increases to a maximum as τ_0' increases, then returns to the value $\Delta' = 1$ when $\tau_0' \gg 1$, while Δ_0' increases without limit as τ_0' increases. Figure 5 shows the values of Δ' and Δ_0' vs τ_0' for both liquids CS_2 and $C_6H_5NO_2$. The behavior illustrated in this figure is of considerable significance to the understanding of the temporal transmission characteristics of an OKE gate or shutter.

The information contained in Figs. 1 and 2 and summarized in Figs. 3–5 presents a very general picture of the temporal behavior of the OKE in-

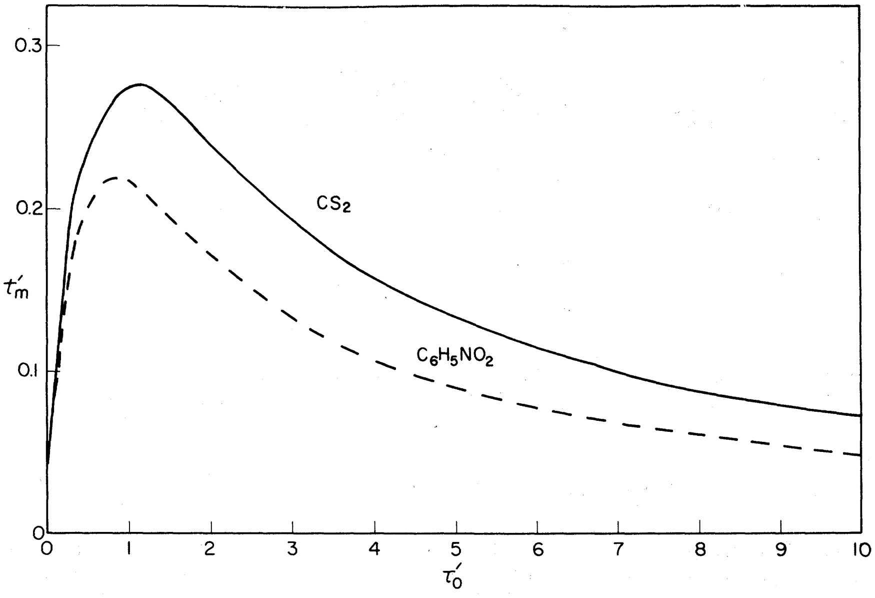

FIG. 4. Time lag t_m' in units of T_ω between $\Delta n|_{\max}$ and the peak of the optical pulse versus $\tau_0' = \tau_0/T_\omega$ in liquids carbon disulfide and nitrobenzene.

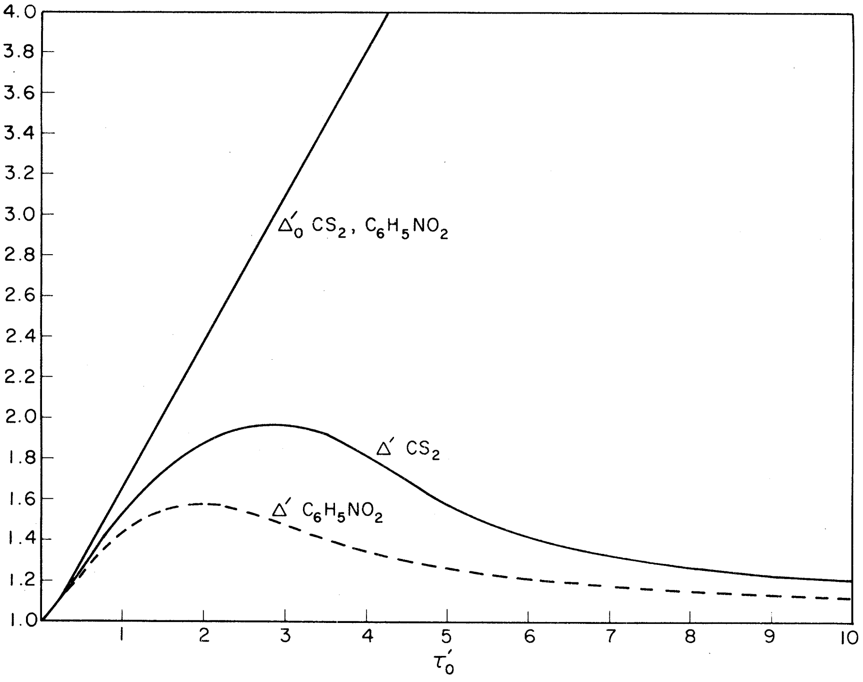

FIG. 5. FWHM durations of the birefringence Δ' and the orientational birefringence Δ_0' in units of T_ω vs the parameter $\tau_0' = \tau_0/T_\omega$ in carbon disulfide and nitrobenzene.

duced by a short-duration laser pulse. Initially, when the pulsewidth T_ω is much greater than the orientational relaxation time τ_o, the intensity of the induced birefringence is a maximum with both the orientational part Δn^o and the electronic part Δn^e in synchronization with the optical pulse. There is no time lag between the induced birefringence and the pulse, and the FWHM of the birefringence is identically that of the pulse. However, when $T_\omega \lesssim \tau_o$, the orientational contribution Δn^o begins to lessen in its intensity and to lag somewhat behind the peak of the optical pulse. Because of this, the total induced birefringence Δn is also reduced in intensity and lags behind the pulse. In addition, the birefringence, because of its relaxing orientational part, is more spread out in time, so that its FWHM is larger than that of the pulse. In the limit $T_\omega \ll \tau_o$ (but $T_\omega > \tau_e$), the orientational contribution Δn^o is completely damped out, with the result that $\Delta n = \Delta n^e$ with no time lag and the FWHM is once again equal to the pulsewidth.

This picture of the temporal behavior of the OKE induced by ultrashort laser pulses offers a practical suggestion concerning the problem of the extension of OKE photography to the subpicosecond regime. Since the time that picosecond-resolution OKE photography was first reported, it has been postulated that, with the availability of intense subpicosecond pulses, this form of photography could be extended to time scales much less than a picosecond by using glass or some similar solid as the optical Kerr medium.[12,14] In such materials, the induced birefringence is due solely to the electronic effect, and is characterized[11,12,17,64] by response times of ~10^{-15} to 10^{-16} sec and optical Kerr coefficients $n_2 \lesssim 10^{-12}$ esu.[78-80] Aside from the practical disadvantage of requiring optically pure samples which would be subject to irreversible laser-produced damage, the use of glass would present problems of very low ($\ll 1\%$) shutter transmissions stemming from the low value of n_2 and the fact that the transmission is a highly nonlinear function[3,11] of n_2. However, the results shown in Figs. 1 and 2 point to the possibility of using a Kerr liquid with relatively large τ_o as the medium for a subpicosecond OKE shutter. For $T_\omega \ll \tau_o$, only the electronic contribution Δn^e with its fast response time $\tau_e \sim 10^{-15}$ sec would remain. As an example, using nitrobenzene ($\tau_o \sim 32$ psec) and a 0.1-psec pulse, the orientational part Δn^o would be $\cong 0$ ($\tau_o' > 300$), and thus $\Delta n \cong \Delta n^e$. Such a Kerr cell under these conditions could give subpicosecond exposure times. Moreover, the cell would be simple in construction, not subject to laser-produced damage, and, most importantly, the comparatively large value of n_2^e in $C_6H_5NO_2$ of $\simeq 3 \times 10^{-12}$ esu [from Eq. (8)] would result in a marked improvement in net transmission over a comparable shutter employing glass. While it is necessary to first isolate the electronic response of a Kerr medium in order to obtain subpicosecond resolution with an OKE shutter, this condition is not in itself sufficient. Consideration must also be given to gating geometry, the effect of finite image and pulse dimensions, and so on. Discussions of some of these more important considerations and their effect on the resolution capabilities of OKE photography are given elsewhere.[12,31,81]

IV. THE OPTICAL KERR EFFECT INDUCED IN SPHERICALLY SYMMETRIC MOLECULES

The theory and calculations given above have been limited to anisotropic molecules ($\delta^2 \neq 0$). However, for spherically symmetric molecules $\delta^2 = 0$, and thus, from Eqs. (12) and (13), ${}_c n_2^o = 0$. Thus, the zero-order approximation for liquids composed of spherically symmetric molecules would be simply $n_2 = n_2^e$. However, as in the case for anisotropic molecules, it is necessary to consider the effect of multiple-molecular interactions explicitly in the calculation of Δn. It is found that the most important correction arises from the existence of radial correlations between members of pairs of molecules.[50-55] This leads to the phenomenon of "molecular clustering"[82-84] with a contribution to the induced birefringence represented by the optical Kerr clustering coefficient[50]

$$n_2^c = \frac{6\pi\rho(\alpha^{\omega})^2(\alpha^{\omega'})^2}{5kTn_0}\left(\frac{n_0^2+2}{3}\right)^2\left(\frac{n_\omega^2+2}{3}\right)^2 J_r\,, \tag{26}$$

where

$$J_r = \frac{2\rho}{V}\int\int r_{pq}^{-6}\, g^2(\Omega_p, \Omega_q)\, d\Omega_p\, d\Omega_q \tag{27}$$

is an integral parameter describing the strength of the radial correlation between members of pairs of molecules. In Eq. (27), r_{pq} is the distance between the centers of symmetry of molecules p and q. As in the case of J_a and for the same reasons, calculations of J_r using Eq. (27) are generally very approximate. However, this same parameter J_r also appears in the formula for the depolarization of scattered light D' for media composed of spherically symmetric molecules as[61]

$$D' = \frac{6(\alpha^{\omega})^2 J_r}{5\rho kT\beta_T + 7(\alpha^{\omega})^2 J_r}\,. \tag{28}$$

Solving for J_r from Eq. (28) and substituting into Eq. (26) gives the result

$$n_2^c = 6\pi\rho^2(\alpha^{\omega'})^2\beta_T\,\frac{D'}{6-7D'}\,\frac{1}{n_0}\left(\frac{n_0^2+2}{3}\right)^2\left(\frac{n_\omega^2+2}{3}\right)^2, \tag{29}$$

with the nonlinear refractive index n_2 given by $n_2 = n_2^c + n_2^e$. Using Eqs. (8) and (29), the values calculated for CCl_4 are $n_2^c = 0.14\times10^{-12}$ esu and $n_2^e = 0.11\times10^{-12}$ esu (for a gating-pulse wavelength 0.69 μm and a probe wavelength of 0.48 μm). The resulting value of $n_2 = 0.25\times10^{-12}$ esu is in good agreement with the experimental value of Paillette.[62,63] In addition, the calculated value of $n_2^e/n_2 = 0.44$ agrees well with the experimentally measured value of 0.54 ± 0.16 reported by Hellwarth *et al.*[84]

Although the results of the numerical calculations presented in Figs. 1–5 pertained specifically to liquids composed of anisotropic molecules for which $n_2 = {}_c n_2^o + n_2^e$, the results of those calculations may be applied, at least qualitatively, to liquids composed of spherically symmetric molecules for which $n_2 = n_2^c + n_2^e$. This follows from the fact that, like the orientational mechanism, the clustering process involves the (translational) motion of the entire molecule, and so is characterized by a relaxation time τ_c which is of the same order of magnitude as the orientational relaxation time[64] τ_o. Thus the clustering birefringence has an effect similar to that of the orientational birefringence in the numerical studies of Sec. III. Most of the conclusions arrived at there remain correct when ${}_c n_2^o$ is replaced by n_2^c, and τ_o by τ_c. Also, since it would appear that, in general, the ratio n_2^e/n_2 is substantially larger (~0.5) in media composed of spherically symmetric molecules than in media composed of anisotropic molecules ($n_2^e/n_2 \sim 0.1$–0.3), the convergence of Δn to Δn^e with increasing τ_c' ($= \tau_c/T_\omega$) will occur considerably faster in liquids composed of spherically symmetric molecules.

V. CONCLUSIONS

Although the above analysis of the optical Kerr effect induced in isotropic and anisotropic liquids by ultrashort laser pulses is based on a generalized theory, some idealized assumptions have been made, particularly with regard to the temporal form of the laser pulseshape. Nonetheless, several important conclusions are apparent which should be of particular relevance to the use of the OKE in the temporal diagnosis of the spatial and/or spectral features of ultrafast phenomena. It is observed that consideration of the relative contributions of the electronic and orientational effects in anisotropic liquids is essential to a correct interpretation of the induced birefringence. The temporal development of the birefringence induced by Gaussian laser pulses in carbon disulfide and nitrobenzene has been explicitly described. The theory and mode of analysis are obviously applicable to other liquid media. In addition, although a Gaussian temporal form for the laser pulseshape has been used in the present derivation of the induced birefringence, it is concluded that the analysis also holds for other forms (triangular, hyperbolic sech-squared, Lorentzian, etc.) of the laser pulseshape. Although it is recognized that these do not adequately describe the random multistructured temporal form of the output of many of today's mode-locked lasers, the general conclusions arrived at above would still remain valid. This follows, since for a structured picosecond optical pulse, the modulation in the induced birefringence will always be much less than that of the optical pulse in liquids exhibiting an orientational Kerr effect with the result that, in a first approximation, the birefringence may be treated as smoothly varying. Moreover, the choice of a Gaussian profile is still the most logical pulseshape to include in the analysis. Bandwidth-limited pulses apparently commonly occur in the initial generation of mode-locked Nd:glass laser pulses,[85,86] and in addition, there have been numerous reports recently of bandwidth-limited pulse outputs from mode-locked ruby, Nd:YAG, Nd:glass, and dye lasers.[87]

The present investigation also points to the possible use of the OKE induced in liquids in the subpicosecond domain. This may partially be more suitable than utilizing the weak electronic effect induced in solid transparent media. With the provisions of laser pulses of duration much less than 10^{-12} sec, the OKE would provide, through its incorporation into an optical or photographic shutter, not only a means of analysis of such pulses themselves, but also a large number of phenomena induced by ultrashort laser pulses.

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