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IN AN ISOTROPIC MEDIUM AND ITS DETERMINATION

C. Y. She, M. J. Moran, and R. L. Carman

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THE DEGENERATE CUBIC NONLINEAR SUSCEPTIBILITY TENSOR
IN AN ISOTROPIC MEDIUM AND ITS DETERMINATION

C. Y. She^{*}, M. J. Moran, and R. L. Carman

Lawrence Livermore Laboratory, University of California
Livermore, California 94550

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I. INTRODUCTION

In this report, we address the questions of how to accurately measure the full cubic nonlinear tensor $\chi^{(3)}$ with estimates of the accuracy to be expected. Preliminary experimental data demonstrating feasibility will be commented upon. Anticipating relevant experimental results in the near future, we present a brief yet comprehensive account of the theory of the degenerate or near-degenerate cubic susceptibility of an isotropic medium, such as glass. While the detailed knowledge of the frequency dependence of the cubic susceptibility tensor may be more of academic interest, it is believed that the values of all the independent tensor elements at the relevant laser frequency are needed in order to explore directly the relative importance of various mechanisms at 100 psec to 1 nsec, as well as the consequence of nonlinear refractive index effects on the high power beam propagation in the laser medium, windows, lenses, and isolators. In addition, the effect of light-by-light scattering on high power beam propagation is also briefly discussed.

^{*}Permanent address: Colorado State University, Fort Collins, Colorado.

II. SELF FOCUSING OF OPTICAL BEAMS

The optical nonlinearity responsible for self focusing may be expressed in terms of an intensity dependent dielectric constant and the associated index of refraction. Following Dawes and Marburger,¹

$$D = \epsilon E = [\epsilon_0 + \epsilon_2 \langle |E|^2 \rangle_{av}] E \quad (1)$$

$$n = \sqrt{\epsilon} = \sqrt{\epsilon_0} + \frac{\epsilon_2}{2\sqrt{\epsilon_0}} \langle |E|^2 \rangle_{av} = n_0 + n_2 \langle |E|^2 \rangle_{av} \quad (2)$$

where $\langle |E|^2 \rangle_{av}$ is the rms value of $|E|^2$. For a Gaussian beam with intensity distribution $I = I_0 \exp(-r^2/a^2)$, propagating with an initially plane phase front, the curvature of the phase front will increase as the beam propagates due to diffraction. The index nonlinearity will just cancel out the diffraction effect on axis, at a total power equal to the Talanov critical power P_1 , defined as

$$P_1 = \frac{c \lambda_0^2}{32 \pi^2 n_2}, \quad \lambda_0 = \frac{2\pi n_0}{k} \quad (3)$$

When the incident power in the beam significantly exceeds P_1 , self focusing will occur. The relation between the total power in a Gaussian beam and the self focusing distance Z_f has been obtained numerically as

$$\left(\frac{2P}{P_1} \right)^{1/2} = 2.322 + \left[\left(\frac{ka^2}{Z_f} \right)^2 - 0.148 \right]^{1/2}$$

For $P \gg P_1$, Eq. (3) reduces to

$$Z_f = \frac{a}{2} \sqrt{\frac{n_0}{\Delta n}} = \left[\frac{c n_0}{16 n_2 P} \times 10^{-7} \right]^{1/2} a^2 \quad (4)$$

where the relations $\Delta n = n_2 \langle |E|^2 \rangle_{av}$ and $P = [(c n_0 / 4\pi) \langle |E|^2 \rangle_{av} (\pi a^2)] \times 10^{-7} \text{ W}$ have been used. Using $n_0 = 1.62$, $n_2 = 10^{-11} \text{ esu}$ for CS_2 , and $n_0 = 1.56$, $n_2 = 2 \times 10^{-13} \text{ esu}$ for ED-2 glass, the critical powers for self focusing are estimated respectively for CS_2 and ED-2 glass as 10^5 W and $3 \times 10^6 \text{ W}$. For a beam with $a = 0.3 \text{ cm}$ and $P \approx 10^8 \text{ W}$ one gets $Z_f \approx 60 \text{ cm}$ and $\Delta n \approx 10^{-5}$ for CS_2 , and $Z_f \approx 350 \text{ cm}$ and $\Delta n \approx 3 \times 10^{-7}$ for ED-2 glass.

When the length of the nonlinear medium ℓ is much shorter than the self focusing length Z_f , the nonlinear refractive medium may be approximated by a thin lens with an equivalent focal length

$$f_{eq} = \frac{a^2}{4\Delta n \ell} = \frac{Z_f^2}{n_0 \ell} \gg Z_f, \quad \Delta n = n_2 \langle |E|^2 \rangle_{av} \quad (5)$$

It is interesting to note that f_{eq}^{-1} in the thin lens limit is proportional to $\Delta \ell = \Delta n \ell$, while Z_f^{-1} is proportional to $\sqrt{\Delta n}$. This difference must be considered from experimental points of view.

In practice, small scale beam trapping at high power usually takes place even when $\ell \approx 0.1 Z_f$. This has been noticed in nonlinear liquids⁴ and was attributed to micro-scale distortion in the beam profile present in the beam perhaps due to random fluctuations in the optical properties of the medium. Speck and Bliss⁵ have also noticed this phenomenon in glass when they reported

the beginning of the small scale trapping at $1-4 \text{ GW/cm}^2$, while the calculated Z_f at this power level is at least ten times longer than the sample length used. The reason for this is thought to be the growth of small scale instabilities.

III. CUBIC NONLINEAR SUSCEPTIBILITY TENSOR

The nonlinear refractive indices responsible for self focusing and related phenomena are the result of the induced polarization of third order in the electric field strength. The nonlinear susceptibility in question is a fourth rank tensor and for isotropic materials such as CS_2 and glass, there are three independent elements in this tensor.⁶ Because this nonlinear property leads to such interesting effects as third harmonic generation, stimulated Raman scattering, three wave mixing, two photon absorption, and above all self focusing and self trapping of laser beams, and because it is nearly impossible to extract relevant information quickly from the literature, it is useful to briefly review the properties of this tensor and to comment on the relations that must be considered for the determination of nonlinear refractive indices.

If the electric field strength of an optical beam is expressed as the Fourier transform pair

$$E(t) = \int_{-\infty}^{\infty} d\omega E(\omega) e^{-i\omega t}, \quad E(\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dt E(t) e^{i\omega t}, \quad (6)$$

the first and third order polarizations $P^{(1)}(t)$ and $P^{(3)}(t)$ may be expressed in terms of a second rank tensor $\chi^{(1)}(\omega)$ and a fourth rank tensor $\chi^{(3)}(\omega_1, \omega_2, \omega_3)$.

respectively as

$$P_{11}^{(1)}(t) = \int_{-\infty}^{\infty} d\omega \chi_{11a}^{(1)}(\omega) E_a(\omega) e^{-i\omega t} \quad (7a)$$

$$P_{11}^{(3)}(t) = \int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 \int_{-\infty}^{\infty} d\omega_3 \chi_{11a_1 a_2 a_3}^{(3)}(\omega_1, \omega_2, \omega_3) \times E_{a_1}(\omega_1) E_{a_2}(\omega_2) E_{a_3}(\omega_3). \quad (7b)$$

The susceptibility tensors defined in this way may be extended to higher orders. The expressions (7a) and (7b) were obtained on the assumptions that the induced polarization may be expanded in a power series of electric field strength and that the dynamical properties of the medium under discussion are independent of time. The susceptibilities as defined above are also subject to many symmetry conditions,⁶ one of which, called the intrinsic permutation symmetry, states that $\chi_{11a_1 a_2 a_3}^{(3)}(\omega_1, \omega_2, \omega_3)$ is invariant under all 3! permutations of the pairs $a_1 \omega_1, a_2 \omega_2$, and $a_3 \omega_3$.

In practice, the applied electric field consists of several monochromatic or quasi-monochromatic components as

$$\vec{E}(t) = \frac{1}{2} \sum_f \vec{E}_f(\omega_f) e^{-i\omega_f t} \quad (8)$$

It is straightforward to substitute Eq. (8) into Eqs. (7) and to show⁶ that

$$\vec{P}^{(3)}(t) = \sum_{f_1 \dots f_3} \chi^{(3)}(\omega_{f_1}, \omega_{f_2}, \omega_{f_3}) : E_{f_1}(\omega_{f_1}) E_{f_2}(\omega_{f_2}) E_{f_3}(\omega_{f_3}) \times \exp \left[-i \sum_{m=1}^3 \omega_{f_m} t \right] \quad (9)$$

Following the convention of References 7 and 8, we note that the tensor $\chi^{(3)}$ in Eq. (9) equals one-fourth times its counter part in Eq. (7b).

Effects such as third harmonics and three wave mixing will not be considered here; the quasi-monochromatic components are limited to at most two. For an electric field having one monochromatic component, the induced third order polarization and the associated first order equivalent susceptibility for an isotropic medium are⁷

$$\begin{aligned} P_i^{(3)}(\omega) = & 6\chi^{1122}(-\omega; \omega, \omega, -\omega) E_i(\omega) E_j(\omega) E_j^*(\omega) \\ & + 3\chi^{1221}(-\omega; \omega, \omega, -\omega) E_j(\omega) E_j(\omega) E_i^*(\omega) \end{aligned} \quad (10a)$$

$$\begin{aligned} \Delta\chi_{ij}^{(1)}(\omega) = & 6\chi^{1122}(-\omega; \omega, \omega, -\omega) E_k(\omega) E_k^*(\omega) \delta_{ij} \\ & + 3\chi^{1221}(-\omega; \omega, \omega, -\omega) E_j(\omega) E_i^*(\omega) \end{aligned} \quad (10b)$$

where the summation convention (repeated indices are summed) has been assumed. For two monochromatic components, one has⁸

$$\begin{aligned} P_i^{(3)}(\omega_1) = & 6\chi^{1122}(-\omega_1; \omega_1, \omega_2, -\omega_2) E_i(\omega_1) E_j(\omega_2) E_j^*(\omega_2) \\ & + 6\chi^{1212}(-\omega_1; \omega_1, \omega_2, -\omega_2) E_i(\omega_2) E_j^*(\omega_2) E_j(\omega_1) \\ & + 6\chi^{1221}(-\omega_1; \omega_1, \omega_2, -\omega_2) E_j(\omega_1) E_j(\omega_2) E_i^*(\omega_2) \end{aligned} \quad (11a)$$

$$\begin{aligned} \Delta\chi_{ij}^{(1)}(\omega_1) = & 6\chi^{1122}(-\omega_1; \omega_1, \omega_2, -\omega_2) E_k(\omega_2) E_k^*(\omega_2) \delta_{ij} \\ & + 6\chi^{1212}(-\omega_1; \omega_1, \omega_2, -\omega_2) E_i(\omega_2) E_j^*(\omega_2) \\ & + 6\chi^{1221}(-\omega_1; \omega_1, \omega_2, -\omega_2) E_j(\omega_2) E_i^*(\omega_2) \end{aligned} \quad (11b)$$

It should be noted that when $\omega_1 = \omega_2$, the intrinsic permutation symmetry implies $\chi^{1122}(-\omega_1; \omega_1, \omega_1, -\omega_1) = \chi^{1212}(-\omega_1; \omega_1, \omega_1, -\omega_1)$. Equations (11) should reduce to Eqs. (10). The fact that they still differ by a factor of two is due to the convention and bookkeeping method used in References 6-8. There the susceptibility with two identical frequency arguments was defined as being twice as large as the same susceptibility with all distinct frequency arguments. ($-\omega$ is distinct from ω .)

Strictly speaking, the permutation symmetry, $\chi^{1122}(-\omega; \omega, \omega, -\omega) = \chi^{1212}(-\omega; \omega, \omega, -\omega)$ is valid only for monochromatic electric fields. If the inducing field is a quasi-monochromatic laser pulse, this condition may not be valid; and Eqs. (11) should be used with the factor 6 replaced by the factor 3. Another condition is often used in the literature^{8,9}: for a lossless system, one may assume $\Delta\chi_{ij}^{(1)}(\omega_1) = \Delta\chi_{ji}^{(1)}(\omega_1)$. This, according to Eq. (11b), implies that $\chi^{1212}(-\omega_1; \omega_1, \omega_2, -\omega_2) = \chi^{1221}(-\omega_1; \omega_1, \omega_2, -\omega_2)$. The validity of these conditions will be discussed and in general, these conditions will not be used here because which microscopic processes are involved is not well understood.

IV. PHYSICAL ORIGINS

In general, both electronic and nuclear motion may be induced by the applied electric field in a nonlinear manner. The basic idea of dividing the origin of the physical mechanisms responsible for the third order nonlinear susceptibilities into electronic and nuclear has been given in References 10 and 11.

According to References 10 and 11, the third order polarization may be written (approximately) as the sum of an electronic contribution and a nuclear contribution

$$\vec{p}^{(3)}(t) = \vec{p}_e^{(3)}(t) + \vec{p}_n^{(3)}(t) \quad (12a)$$

$$\vec{p}_e^{(3)}(t) = \frac{1}{2} \sigma \vec{E}(t) \vec{E}(t) \cdot \vec{E}(t) \quad (12b)$$

$$\begin{aligned} \vec{p}_n^{(3)}(t) = & \vec{E}(t) \int a(t-s) \vec{E}(s) \cdot \vec{E}(s) ds \\ & + \vec{E}(t) \cdot \int b(t-s) \vec{E}(s) \vec{E}(s) ds \end{aligned} \quad (12c)$$

Here, $\vec{p}_e(t)$ results from a distortion of the electronic orbits about the nuclei in a fixed configuration. The intensity dependent electronic polarizability is considered instantaneous because its lifetime, 10^{-14} sec, is much shorter than the duration of the laser pulse used. A single constant is needed to describe this effect for an isotropic medium. The response of the electronic current surrounding a nuclear arrangement also forces the nuclei to change positions in order to minimize the system energy. This in turn represents a change in electronic polarizability (susceptibility) which depends on nuclear coordinates and on applied electric fields in second order. For an isotropic medium, two functions are necessary to account for nuclear movements "parallel" and "perpendicular" to the responding electronic current. Since the nuclear displacements are much slower, one must use two time response functions $a(t-s)$ and $b(t-s)$. The Fourier transform of these functions are defined as

$$\alpha(\omega) = \int a(t) e^{i\omega t} dt, \quad \beta(\omega) = \int b(t) e^{i\omega t} dt \quad (13)$$

Notice, again following the literature, the differences in definition between

Eq. (13) and Eq. (6). If one ignores the sum frequency and the second harmonic contributions in $\alpha(\omega)$ and $\beta(\omega)$, the third order polarization of Eqs. (12) may be expressed in terms of monochromatic electric field components. For one or two monochromatic field components, one respectively has,

$$P_i^{(3)}(\omega) = \frac{1}{4}[\sigma + 2\alpha(0) + \beta(0)]E_i(\omega)E_j(\omega)E_j^*(\omega) + \frac{1}{4}\left[\frac{1}{2}\sigma + \beta(0)\right]E_j(\omega)E_j(\omega)E_i^*(\omega) \quad (14a)$$

and

$$P_i^{(3)}(\omega) = \frac{1}{4}[\sigma + 2\alpha(0) + \beta(\omega_1 - \omega_2)]E_i(\omega_1)E_j(\omega_2)E_j^*(\omega_2) + \frac{1}{4}[\sigma + 2\alpha(\omega_1 - \omega_2) + \beta(0)]E_i(\omega_2)E_j^*(\omega_2)E_j(\omega_1) + \frac{1}{4}[\sigma + \beta(0) + \beta(\omega_1 - \omega_2)]E_j(\omega_1)E_j(\omega_2)E_i^*(\omega_2) \quad (14b)$$

Comparing Eqs. (14) and Eqs. (10a) and (11a), one may relate the susceptibility tensor elements as defined in Eq. (9) to the parameters representing nuclear and electronic contributions. They are:

$$\begin{aligned} 6\chi^{1122}(-\omega_1; \omega_1, \omega_2, -\omega_2) &= \frac{1}{4}[\sigma + 2\alpha(0) + \beta(\omega_1 - \omega_2)] \\ 6\chi^{1212}(-\omega_1; \omega_1, \omega_2, -\omega_2) &= \frac{1}{4}[\sigma + 2\alpha(\omega_1 - \omega_2) + \beta(0)] \\ 6\chi^{1221}(-\omega_1; \omega_1, \omega_2, -\omega_2) &= \frac{1}{4}[\sigma + \beta(0) + \beta(\omega_1 - \omega_2)] \end{aligned} \quad (15)$$

Expressions for $\chi^{1122}(-\omega; \omega, \omega, -\omega)$ and $\chi^{1221}(-\omega; \omega, \omega, -\omega)$ may be obtained from Eqs. (12) by letting $\omega_1 = \omega_2 = \omega$. It is interesting to note from Eq. (15) that $\chi^{1122} = \chi^{1212}$ if $\omega_1 = \omega_2$ in agreement with the intrinsic permutation symmetry and that $\chi^{1212} = \chi^{1221}$ if either $\alpha(\omega_1 - \omega_2) = \beta(\omega_1 - \omega_2) = 0$, or $\omega \gg \alpha, \beta$. The lossless condition here means that the electronic contribution dominates.

The dispersion of the nuclear responses $\alpha(\omega)$ and $\beta(\omega)$ ignoring resonances may be roughly sketched as shown in Fig. 1. τ is essentially the relaxation time, probably 0.1-100 psec, of the nuclear response to the inducing light pulse. With the aid of this figure, several conclusions can be made. If the effect is induced by a quasi-monochromatic pulse with a duration $T \gg \tau$, the dominant contribution of $\alpha(\omega)$ and $\beta(\omega)$ comes from low frequency fluctuations at $\omega \approx 0$. In this case, one has $\chi^{1122}(-\omega_1; \omega_1, \omega_2, -\omega_2) = \chi^{1212}(-\omega_1; \omega_1, \omega_2, -\omega_2)$. If $T \lesssim \tau$, the permutation symmetry relation is not valid. In this case the two constants take their respective average values over a frequency band of $\sim 1/\tau$ in width. The condition $\chi^{1212}(-\omega_1; \omega_1, \omega_2, -\omega_2) = \chi^{1221}(-\omega_1; \omega_1, \omega_2, -\omega_2)$ is valid if either α is dominant or $|\omega_1 - \omega_2| \gg 1/\tau$. This latter criterion may be met in an induced birefringence (Kerr effect) measurement when the probing beam has a different frequency from the inducing beam.

V. MEASURABLE PARAMETERS

Both the self induced nonlinear refractive indices and the nonlinear refractive indices induced by a strong beam at a different frequency will be considered. For the self induced effects by a quasi-monochromatic laser pulse, Eqs. (11) with 6, ω_1 , and ω_2 respectively replaced by 3, ω , and ω will be used, because χ^{1122} may not in general equal χ^{1212} . If $\chi^{1122}(-\omega; \omega, \omega, -\omega) =$

$\chi^{1212}(-\omega; \omega, \omega, -\omega)$, these equations will be reduced to Eqs. (10). If an elliptically polarized laser pulse with circular components $E_+(\omega) = [E_x(\omega) \pm iE_y(\omega)]/\sqrt{2}$, is propagating along the z axis, the induced third order polarization becomes

$$\begin{aligned} P_+^{(3)}(\omega) &= [3(\chi^{1122} + \chi^{1212})|E_+(\omega)|^2 + 3(\chi^{1122} + \chi^{1212} + 2\chi^{1221})|E_-(\omega)|^2]E_+(\omega) \\ P_-^{(3)}(\omega) &= [3(\chi^{1122} + \chi^{1212} + 2\chi^{1221})|E_+(\omega)|^2 + 3(\chi^{1122} + \chi^{1212})|E_-(\omega)|^2]E_-(\omega) \end{aligned} \quad (16)$$

where $|E(\omega)|^2 = |E_+(\omega)|^2 + |E_-(\omega)|^2 = |E_x(\omega)|^2 + |E_y(\omega)|^2$. The quantities in the brackets may be considered as the intensity dependent susceptibilities $\Delta\chi$. Depending on relative values of $E_+(\omega)$ and $E_-(\omega)$, different expressions for $\Delta\chi$ may be obtained. Straightforward but laborious manipulation of Eq. (16) allows us to derive $\Delta\chi_{\parallel}$, $\Delta\chi_{\odot}$, and $\Delta\chi_{\odot\odot}$ which are induced linear equivalent susceptibilities (or susceptibility) changes of a linear, circular, and elliptically polarized beam, respectively. They can be shown to be:

$$\begin{aligned} \Delta\chi_{\parallel}(\omega) &= 3(\chi^{1122} + \chi^{1212} + \chi^{1221})|E(\omega)|^2 \\ \Delta\chi_{\odot}(\omega) &= 3(\chi^{1122} + \chi^{1212})|E(\omega)|^2 \\ \Delta\chi_{\odot\odot}(\omega) &= \Delta\chi_+ - \Delta\chi_- = 6\chi^{1221}[|E_-(\omega)|^2 - |E_+(\omega)|^2] \end{aligned} \quad (17)$$

All these quantities $\Delta\chi_{\parallel}$, $\Delta\chi_{\odot}$, and $\Delta\chi_{\odot\odot}$ may be experimentally determined. Due to the fact that χ^{1122} and χ^{1212} appear as a sum in Eq. (17), only two independent quantities $\chi^{1122} + \chi^{1212}$ and χ^{1221} may be experimentally determined.

Much more can be learned if the nonlinear property of the medium probed

at ω_1 is induced by a strong beam at a different frequency ω_2 . In fact, ω_1 may be varied in order to map out the frequency dependence of these susceptibility tensor elements. Using Eqs. (11), it is easy to derive the following relations if the inducing beam is polarized linearly along the x axis, i.e., $E_y(\omega_2) = 0$:

$$\begin{aligned} p_x^{(3)}(\omega_1) &= 6(\chi^{1122} + \chi^{1212} + \chi^{1221})|E(\omega_2)|^2 E_x(\omega_1) \\ p_y^{(3)}(\omega_1) &= 6\chi^{1122}|E(\omega_2)|^2 E_y(\omega_1) \end{aligned} \quad (18)$$

where the χ 's depends on both ω_1 and ω_2 . The following induced susceptibilities are measurable:

$$\Delta\chi_{||}(\omega_1, \omega_2) = 6(\chi^{1122} + \chi^{1212} + \chi^{1221})|E(\omega_2)|^2 \quad (19a)$$

$$\Delta\chi_{\perp}(\omega_1, \omega_2) = 6\chi^{1122}|E(\omega_2)|^2 \quad (19b)$$

$$\Delta\chi_B = \Delta\chi_{||} - \Delta\chi_{\perp} = 6(\chi^{1212} + \chi^{1221})|E(\omega_2)|^2 \quad (19c)$$

As much as the model described by Eqs. (12) is valid, experimental determination of σ , $\alpha(\omega)$, and $\beta(\omega)$, keeping Eq. (15) in mind, would be possible. This may be done by first determining σ , $\alpha(0)$, and $\beta(0)$ with two measurements using a long pulse laser (thus nearly monochromatic) and a third measurement of induced birefringence having $|\omega_1 - \omega_2| \gg 1/\tau$. The induced susceptibilities may be written as,

$$\Delta\chi_{||}(\omega) \approx \frac{1}{4} \left[\frac{3}{2}\sigma + 2\alpha(o) + 2\beta(o) \right] |E(\omega)|^2 \quad (20a)$$

$$\Delta\chi_{\odot}(\omega) \approx \frac{1}{4} [\sigma + 2\beta(o)] [|E_{-}(\omega)|^2 - |E_{+}(\omega)|^2] \quad (20b)$$

$$\Delta\chi_{||}(\omega_1, \omega_2) - \Delta\chi_{\perp}(\omega_1, \omega_2) \approx \frac{1}{4} [2\sigma + 2\beta(o)] |E(\omega_2)|^2 \quad (20c)$$

Equations (20b) and (20c) may be determined respectively by measuring ellipse rotation and Kerr constant. In fact, References 10 and 11 used these values to determine the relative importance of σ and β , thus accessing the relative importance of electron and nuclear contributions to the third order susceptibilities. After σ , $\alpha(o)$, and $\beta(o)$ are determined, the dispersion of $\alpha(\omega)$ and $\beta(\omega)$ may be obtained by measuring $\Delta\chi_{||}(\omega_1, \omega_2)$ and $\Delta\chi_{\perp}(\omega_1, \omega_2)$ as a function of $(\omega_1 - \omega_2)$.

In terms of σ , α , and β , the two self induced susceptibility constants (by a laser pulse with duration $T = 1/\Delta\omega$) may be expressed as

$$\begin{aligned} 3(\chi^{1122} + \chi^{1212}) &= \frac{1}{2}(A + B) \\ 6\chi^{1221} &= B \end{aligned} \quad (21)$$

where $A(\Delta\omega) = (1/4) [\sigma + 2\alpha(o) + 2\alpha(\Delta\omega)]$ and $B(\Delta\omega) = (1/4) [\sigma + \beta(o) + \beta(\Delta\omega)]$.

The values of $\alpha(\Delta\omega)$ and $\beta(\Delta\omega)$ are the averages of $\alpha(\omega)$ and $\beta(\omega)$ over the laser spectral width $\Delta\omega = 1/T$. They approach their dc values $\alpha(o)$ and $\beta(o)$ if

$T \gg \tau$ (life time of the nonlinear processes) and become zero if $T \ll \tau$.

This means that the self induced susceptibilities leading to self focusing of a laser pulse are independent of pulse duration if the electronic processes

dominate and that they are twice bigger for a long pulse ($T \gg \tau$) if the nuclear processes, which are unable to respond to fast changes, dominate.

In terms of experimental determinations to be discussed below, it is appropriate to express Δn in terms of refractive index changes $\Delta n = 2\pi\Delta\chi/n_0$ as

$$\Delta n_{\parallel}(\omega) = n_2'' |E(\omega)|^2/2; \quad n_2'' = 12\pi(\chi^{1122} + \chi^{1212} + \chi^{1221})/n_0 \quad (22a)$$

$$\Delta n_{\perp}(\omega) = n_2^{\circ} |E(\omega)|^2/2; \quad n_2^{\circ} = 12\pi(\chi^{1122} + \chi^{1212})/n_0 \quad (22b)$$

$$\Delta n_B(\omega_1, \omega_2) = [n_2''(\omega_1, \omega_2) - n_2^{\circ}(\omega_1, \omega_2)] |E(\omega_2)|^2/2;$$

$$n_2'' - n_2^{\circ} = 12\pi(\chi^{1212} + \chi^{1221})/n_0 \quad (22c)$$

VI. INTERFEROMETRIC MEASUREMENTS

It is well known in optics that fringes produced by the interference of two optical beams may be used to determine the refractive index of a medium. In fact, the Mach-Zehnder refractometer has been used to study slight changes in linear refractive index in gases.¹² The usefulness of this method is limited for solid medium because the insertion of a piece of solid in one of the beams in the Mach-Zehnder interferometer produces a discontinuous shift of interference fringes which can not be counted in practice. That the interference technique may in principle be used to measure the intensity dependent refractive index has long been recognized⁸; its success, however, depends critically on the ability of resolving the induced effect in time, so that the shift in fringes corresponding to different intensities (thus

different Δn) may be separately recorded. This technique was used to study thermal blooming.¹³ Only recently, with the availability of a smooth optical pulse and high resolution (~10 psec) streaking cameras, has it been used to measure fast responding, nonlinear refractive index changes.⁵

There are several improvements made on our Mach-Zehnder interferometer currently set up for the measurement of nonlinear refractive indices. Unlike the system used by Speck and Bliss,⁵ two delay compensators, one for each beam have been inserted to produce equal path fringes to within the streak camera resolution. Since its value of n_2'' is known to three significant figures,¹⁴ the experiments are to be carried out as ratio measurements with respect to CS_2 as proposed by Carman.¹⁵ Thus, only the relative intensities of the two beams need to be determined. This can be done quite accurately using the multiple image camera technique with 1-Z plates. Because the n_2 value for CS_2 is about two orders larger than that for glass, 10% wedged plates are used as mirrors in the Mach-Zehnder. Due to the ratio technique used, better accuracy may be achieved with a "null" measurement. With this technique not only the linear, but also the nonlinear index effects in the glass sample are compensated for in the second beam. To our knowledge this is the first time this has been attempted.

We may measure n_2'' readily with a linearly polarized laser pulse. With the insertion of the two quarter wave plates before and after the glass sample, n_2° can be measured by the same set up. For both these cases, the same beam is used as the inducing and the probing beam. When the intensities of the two beams are adjusted to achieve the null condition, i.e., straight streaking fringes, the n_2 of glass may be determined in terms of the n_2 of CS_2 as

$$n_{2\text{glass}} = n_{2\text{CS}_2} \cdot \frac{I_{\text{CS}_2}}{I_{\text{glass}}} \cdot \frac{l_{\text{CS}_2}}{l_{\text{glass}}} \quad (23)$$

Since the three quantities in the right hand side of Eq. (23) can be determined more accurately than one's ability to detect straight streaking fringes, the error of the interferometric measurement is determined by the straightness of the interference fringes. For example, with a beam of $a = 10^{-1}$ cm, $P = 7 \times 10^7$ W, the change of refractive index in glass is roughly $\Delta n \approx 2 \times 10^{-6}$. For a rod 50 cm long, it would produce a shift of $\Delta n l / \lambda \approx 1$ fringe. If the ability of detecting straight fringes is (realistically) 0.1 fringe, the error of the n_2 measurement would be 10%. Increasing the laser power by a factor of four, the maximum shift would be four fringes and the error of the measurement would be 5%. With the help of intensity analysis of the interference fringes, 1% error should be possible. For less accuracy shorter rods may be used.

Due to the distortion inherent in YAG rods used in the amplifier chain, the far field of the optical beam has been used for the preliminary experiments. This is accomplished by mildly focusing the optical pulse with a 2 m lens which produces a beam diameter of approximately 2mm near the focal point extending over 50 cm. In order to achieve a high accuracy of the measurement, one may have to use incident powers so high that some self focusing instabilities have in practice occurred. Because the length of the sample according to calculation is much shorter than Z_f for a power corresponding to less than one fringe distortion, the gross beam is in the regime of thin lens self focusing and the interference fringes are still observable. However, the occurrence of small scale self focusing would reduce the contrast of the interference fringes, as experimentally observed and shown in Fig. 2, further

increases in power are not meaningful. A clean beam is therefore essential for conducting this experiment with better than 20% accuracy. We are now about to start operating with glass amplifiers replacing the Nd:YAG Amplifiers which should accomplish this.

As mentioned before there are only two independent susceptibility constants, $\chi^{1122} = \chi^{1212}$ and χ^{1221} , for the propagation of a long quasi-monochromatic pulse ($T \geq 1$ nsec). The measurement of $\Delta n_{||}$ and $\Delta n_{\perp}$ described above would be sufficient for their determination. However, in terms of Hellwarth's model,^{10,11} one needs a third measurement to determine σ , α , and β . The induced birefringence, $\Delta n_B(n_2'' - n_2')$ when the probe beam and inducing beam have different frequencies, can also be measured with a Mach-Zehnder interferometer. Figure 3 shows a setup that may be used. The inducing beam is at 1.06μ and the probe beam is its second harmonic at 0.53μ . For maximum fringe visibility, the probe beams at 0.53μ should have nearly the same intensity in both arms. The inducing beams at 1.06μ in both arms need not have exactly the same intensity; their relative intensity must be monitored. This relative intensity may be adjusted to achieve the null condition for straight fringes. Alternatively, a short cell of CS_2 may be inserted in one arm of the interferometer to determine $\Delta n_B(\omega_1, \omega_2)$ in terms of n_2 of CS_2 . The errors involved are similar to those considered previously. If a dye laser is used, the frequency dependence of $\Delta n_B(\omega_1, \omega_2)$ would be determined. Because the optical components used in Fig. 3 must respond properly to both the fundamental and second harmonics, the interferometric method may not be as practical as the usual induced birefringence measurement to be discussed later.

VII. NON-INTERFEROMETRIC MEASUREMENTS

This class of experiments is an alternative to the interferometric method described above. The three pieces of information, $\Delta n_{||}$, $\Delta n_{\odot}$, and $\Delta n_B \propto (n_2'' - n_2')$ may be determined respectively by dynamic self focusing, ellipse rotation, and induced birefringence measurements. The determination of $\Delta n_{||}$ and Δn_B of glasses using the non-interferometric techniques has been proposed by Carman¹⁵ and $\Delta n_{\odot}$ measurement has been carried out by Owyong.⁹

By determining the on-axis intensities (or equivalently beam cross sections) due to self focusing, we can propagate a linearly (or circularly) polarized laser pulse through the medium of interest to determine n_2'' (or $n_2^{\odot}$). Within the thin lens limit, the equivalent focal length f_{eq} due to the induced nonlinear refractive index is given by Eq. (5). The time dependent (i.e., intensity dependent) beam cross section in the far field may be conveniently determined with an ultrafast streak camera. We can then determine f_{eq} , from which n_2'' and $n_2^{\odot}$ may be calculated. More precisely, depending on the relative positions of the streak camera, different streaking beam profiles may be obtained for the determination of the f_{eq} . This is shown in Fig. 4. The ratio technique relative to CS_2 can again be used. While the thin lens formulation of the problem within the constant spatial shape approximation, similar to the on-axis intensity measurements, is only accurate to about 50%, the parametric dependence is very good.¹⁶ This means that the ratio measurement would give accurate values of n_2 relative to that of CS_2 . We need only make identical far field patterns for the two media.

The result of the measurement depends on the property of the thin lens. For example, consider a laser beam with $P = 10^9$ W, $a = 0.3$ cm, with 1 mrad

divergence. For this beam one finds $\Delta n = 3 \times 10^{-6}$, $Z_f = 110$ cm, and $f_{eq} = 300$ cm for $\lambda = 20$ cm. For a beam divergence of 1 mrad the gross beam would focus to 1 mm if a $f = 1$ m focal length lens is used. At peak intensity the equivalent focal length of the "lens pair" would be $f' = (1/1 + 1/3)^{-1} = 0.75$ m. The 6 mm beam will have a diameter of about 2 mm at one meter from the lens. If the streaking camera is placed in the position (3), the beam diameter will vary by a factor of two. The trick of this experiment depends on proper selection of incident power to give a maximum clearly detectable variation in the beam diameter. A beam with a smooth spatial distribution and smaller divergence clearly helps. The error of this measurement comes from the determination of far field beam cross sections. It is more difficult to change the beam cross section by self focusing than to increase the number of interference fringe shifts required for the interferometric method. Thus, measurement better than 20% would be difficult with this method.

The induced birefringence experiment^{17,18} can be used to determine the value of $\Delta n_B(\omega_1, \omega_2) \propto (n_2'' - n_2')$. The Nd laser at 1.06 μ may be used to induce the nonlinearity, and its second harmonic radiation may be used as the probe beam for the experiment.^{15,18} In general, a dye laser providing variable ω_1 would be a better choice. The intensity of the probe beam $I_{\perp}$ and the induced light passing through a set of crossed polarizers $I_{\parallel}$ are measured. In terms of these quantities, the induced birefringence is

$$\Delta n = (n_2'' - n_2') \langle |E|^2 \rangle_{av} = \frac{\lambda}{\pi d} \left(\frac{I_{\perp}}{I_{\parallel}} \right)^{1/2} \quad (24)$$

where d is the diameter of the inducing beam. Care must be applied and

calibration must be made in determining the ratio of $I_{\perp}/I_{\parallel}$.^{15,18} The induced refractive index $\Delta n_{\odot}$ may be measured by the method of ellipse rotation.^{9,19} Both the induced birefringence and ellipse rotation measurements have been discussed in many publications in the literature; their details will not be repeated here. Basically, these experiments measure intensity ratios of orthogonally polarized components in the (probe) beam due to the differences in either $\Delta\chi_{\parallel} - \Delta\chi_{\perp}$ or $\Delta\chi_{+} - \Delta\chi_{-}$. Because it is easy to detect a polarization change which gives a rotation of several degrees, these methods are in principle very sensitive and a small change in $\Delta\chi_{\parallel} - \Delta\chi_{\perp}$ or $\Delta\chi_{+} - \Delta\chi_{-}$ can be detected. Consequently, one can use relatively lower powers in the induced birefringence and ellipse rotation measurements. However, due to the difficulties in time resolution and intensity determination, errors less than 15% in a single shot and 5% after averaging over many shots would be difficult.⁹

It is clear that the comparison between interferometric and non-interferometric methods depends very much on experimental conditions. In general, $\Delta n_{\parallel}$ can be determined more accurately with the interferometric method, while the non-interferometric method is comparable to or may perhaps be better than the interferometric determination of $\Delta n_{\odot}$ and Δn_R .

VIII. LIGHT-BY-LIGHT SCATTERING

The light-by-light scattering may be considered as a four photon parametric process with zero or near zero frequency shift.^{4,20} A weak wave crossing a strong inducing beam at an angle θ are coupled through the intensity dependent dielectric constant. As a result, the signal of the weak wave is amplified and at the same time, a second weak wave at $-\theta$ is stimulated both at the expense of the strong beam. The optimum gain of the process, according to the

plane wave theory, occurs at (for the linearly polarized wave),

$$\theta_{\text{opt}} = \sqrt{\frac{\Delta n}{n_0}}, \quad \Delta n = n_2'' |E|_{\text{av}}^2 \quad (25)$$

By measuring θ_{opt} in theory one can determine n_2'' . The value of n_2'' determined by this method may be somewhat different from other methods discussed because the result given in Eq. (25) for light-by-light scattering was based on plane waves.

The detailed theory of light-by-light scattering has not been worked out for finite beams. Nor has the experiment in any solid material been performed. Thus, it would be quite interesting to pursue these experiments in glasses. More importantly, it has been pointed out that self trapping can be viewed as the steady-state limit of light-by-light scattering, where the various (spatial) Fourier components lose as much as they gain by scattering and diffraction into each other.²⁰ There has been experimental evidence of this fact in liquids.⁴ However, self trapping does not occur¹⁸ as per Reference 20, thus raising many questions.

The relevance of a study of light-by-light scattering to LLL programs is that small perturbations in the spatial intensity distribution of laser beams will initially grow exponentially as well as simultaneously generate and amplify complimentary beams. In large aperture disk amplifiers, these effects will undoubtedly dominate the degradation of the beam since self focusing across the whole aperture depends on the intensity over beam diameter squared. The source of consequential local perturbations is likely to be small angle scattering, index inhomogeneities, and other local imperfections which are

almost inevitable in such large volume samples. A more complete understanding of this subject could allow theoretical assessments of the practicality of very large aperture systems, especially due to the already much poorer non-linear optical response due to the low n_2/g where g is the operational exponential gain coefficient, compared with larger numbers of smaller aperture systems.

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Fig. 1. Dispersion of nuclear responses.

Fig. 2. Example of small scale self focusing
reducing contrast of interference
fringes.

Fig. 3. Experimental setup to measure induced birefringence.

Fig. 4. Positioning to obtain different streaking beam profiles.

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