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CHARACTERIZATION OF SELF-FOCUSING MECHANISMS

IN LASER MATERIALS

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ABSTRACT

We have determined the time dependence of the nonlinear optical susceptibility tensor of fused quartz and the laser glasses ED-2 (from Owens Illinois) and LSO, C1020, and C2828 (from American Optical). This will allow computation of any nonlinear optical effect involving frequencies below the electronic bandgap and above frequencies at which nuclear motions respond. We have also determined the relative contributions of the "electronic" and "nuclear" nonlinearities to the self-focusing index for linearly polarized light in these glasses; the electronic contribution dominates (~85%) in each. We discuss the implications of these results for laser-material selection.

1. INTRODUCTION

In this report we have:

- a) Determined the entire frequency dependences of the nonlinear optical susceptibility tensor of six laser glasses, over the range of frequencies below their electronic bandgaps, and
- b) Determined the relative contribution to the self-focusing indices of "electronic" and "nuclear" mechanisms. These results, summarized in Section 2, may be used not only to predict nonlinear effects in particular device configurations, but also show trends in laser-material properties which may aid in choosing, or designing, preferred laser-amplifier host materials as well as laser-window glasses.

We find in the summary of results in Section 2, that our results are consistent with other, more specialized, self-focusing measurements. Also, effects such as stimulated Raman scattering which have not previously been considered in laser component design may affect performance in some cases. The theory by which we construct the complete susceptibility functions is given in Section 3. The details of the experimental measurements are given in Section 4.

1.1 Materials

The materials chosen for this study are:

- 1.1.1 Fused quartz (Homosil).
- 1.1.2 American Optical LSO Laser Glass and its Neodymium-doped form C1020.
- 1.1.3 Owens Illinois ED-4 Laser Glass and its Neodymium-doped form ED-2
- 1.1.4 American Optical Neodymium-Doped Laser Glass C2828.

1.2 Method of Approach

We demonstrate theoretically in Section 3, that the desired nonlinear susceptibility tensor of a glass can be closely determined from

- a) its polarized and depolarized differential Raman-scattering cross-sections (as a function of frequency), and
- b) its coefficient for intensity-induced polarization changes (IIPC).

We have performed the absolute measurements of the necessary Raman cross-sections for the above materials, the results of which are given in Figures 1 - 5.

We have used the published measurements of Owyong, Hellwarth and George for the IIPC coefficients of the above glasses ^{1,2} to complete the determination of their third order nonlinear susceptibilities and physical mechanisms.

2. RESULTS

2.1 The Third Order Nonlinear Susceptibility Function

Equation (1) of Section 3 gives the nonlinear optical polarization P^{NL} (below breakdown intensities) for any isotropic material (such as our glasses) in the Born-Oppenheimer approximation, valid for frequencies below the electronic bandgap. In (1), P^{NL} is completely specified by two real functions $a(t)$ and $b(t)$ and a real number σ . The Fourier transforms A_ν and B_ν of $a(t)$ and $b(t)$ are related to the polarized and depolarized "Raman" (i.e., inelastic) scattering cross-sections $d^2\sigma/d\Omega d\omega$ given in Figs. 1-5 by the relations (5) - (8) of Section 3. These are summarized by

$$\text{Im}B_\nu = \frac{\pi c^4}{\hbar \omega_0^3} \frac{d^2\sigma_\perp}{d\Omega d\omega} (1 - e^{-\hbar\nu/kT}),$$

where ω_0 is the frequency of the incident beam and $\omega = \omega_0 - \nu$ the frequency of the inelastically scattered beam. T is temperature. The same relation holds for $\text{Im}A_\nu$ but with $\frac{1}{2}(\sigma_{||} - \sigma_\perp)$ replacing $\sigma_\perp$. Here the subscripts $||$ and $\perp$ denote the polarized and depolarized cross-sections respectively. The real parts (Re) of A_ν and B_ν are related to the imaginary parts given above by the Kramers-Kronig relation (8) of Section 3.

2.2 The Nonlinear Mechanisms

The instantaneous nonlinear (σ) polarization in (1) is that due to the nonlinear response of the electronic motions as if the nuclei were fixed. We refer to this mechanism as "electronic". Other terms in (1) involve the nuclear-response time-delay kernels $a(t)$ and $b(t)$ that arise from the instantaneous

linear response of the electrons in the presence of field-induced alterations in the motions of the nuclei. We refer to the mechanisms that give rise to these terms as "nuclear" as they depend explicitly on nuclear motions and responses.

The intensity - induced polarization changes (IIPC) measured by Owyong, et al.^{1,2} are governed by $\sigma + 2B_0$ as shown in eqn. (2) of Section 2. The Raman scattering spectra gives B_0 (and A_0) via (9) of Section 2. Using these facts, we have made the determinations of σ , A_0 , and B_0 listed in Table I for various laser glasses.

2.2.1 Self-Focusing Mechanisms

The self-focusing of nanosecond-range pulses is governed by the combination of constants given in equation (4) of Section 2 for the "nonlinear refractive index" for linear polarization, n_2 . Our results for this index are listed in Table I and compared with values obtained by other methods. The fraction f_e of this index arising from the "electronic" mechanism is derived from Eqn. (10) of Section 2 and recorded for each glass in Table I. It is seen that the electronic mechanism dominates this effect, contributing generally 80 to 90 % of it. The fraction of each mechanism underlying any other effect may be found similarly.

2.2.2 Stimulated Raman Scattering

A strong monochromatic beam in a transparent material creates exponential gain by which noise at lower frequencies can build to spurious and troublesome optical oscillations. The maximum values of this gain can be read from our cross-sections

in Figs. 1-5 using the relations (7) of Section 2. These values are listed in Table II for a strong beam at $1.06 \mu\text{m}$. Our results for the gain in fused quartz are in agreement with those reported by Stolen and Ippen.³ One can see that, for example, 1% of accidental feedback for a gain path of 10 cm would give rise to stimulated Raman oscillations above 30 GW/cm^2 for all glasses. This effect therefore must be considered in any particular configuration as a potential difficulty. It is well known that higher gains than given by (7) are possible for phase-matched, coupled, Raman-scattered waves and, if difficulties seem possible, such refinements must be considered.

2.3 Comparisons With Other Measurements

The only laser glass in our list for which the "static" nonlinear index n_2 of Eqn. (3) has been measured directly is the Owens Illinois Nd-doped laser glass ED-2. Bliss and Speck⁴ measured n_2 for (unpumped) ED-2 at $1.06 \mu\text{m}$ by an absolute interferometric measurement of the change in refractive index. Moran et al.,⁵ also measured n_2 for ED-2 at $1.06 \mu\text{m}$ by an interferometric comparison with carbon disulphide. Their results are listed in Table I and are seen to agree with our more indirect determination of n_2 to within our experimental errors of $\pm 10\%$.

2.4 Materials Implications

Our results show that simple self-focusing arises mainly from purely electronic nonlinearities. Glass, et al., have shown that these ought to be correlated within the linear refrac-

Material	Refractive Index (647nm)	Raman Data (this work)		IIPC data (ref. 1)	$10^{14}\sigma$ (Raman + IIPC)	$10^{13}n_2$ (eq. 2)	$10^{13}n_2$ Measured	f_e Electronic fraction of n_2 (eq. 10)
		$10^{15}B_o$	$10^{15}(A_o+B_o)$	$10^{14}(\sigma+2B_o)$				
Fused Quartz	1.46	1.2 $\pm$.2	4.0 $\pm$.4	3.1 $\pm$.2	2.9 $\pm$.2	1.0 $\pm$.1		.84 $\pm$.03
		1.1 $\pm$.2 ^a	4.8 $\pm$.4 ^a		2.9 $\pm$.2			.82 $\pm$.03
American Optical LSO Glass	1.51	2.4 $\pm$.4 ^a	6.1 $\pm$.6 ^a	4.6 $\pm$.3	4.1 $\pm$.3	1.4 $\pm$.1		.83 $\pm$.03
American Optical C1020	1.51	1.7 $\pm$.3	4.2 $\pm$.4	4.6 ^d	4.3 $\pm$.3	1.4 $\pm$.1		.89 $\pm$.03
American Optical C2828	1.54	2.2 $\pm$.3	4.3 $\pm$.4	--				
Owens Illinois ED-4	1.56	--	--	5.7 $\pm$.4	5.2 $\pm$.4	1.7 $\pm$.1	1.9 $\pm$.3 ^b	.88 $\pm$.03
Owens Illinois ED-2	1.56	2.5 $\pm$.4	5.4 $\pm$.5	--			1.5 $\pm$.3 ^c	
^a Raman excitation at 514.5 nm ^b ref. 4 ^c ref. 5 ^d Not measured, but assumed to be same as for LSO.								

TABLE I

Predictions of our Raman data for the coefficients A_o and B_o , derived from the cross-sections in Figures (1-5) with eq. (9). Raman data were taken with 647.1 nm excitation except as noted. Given also is the IIPC coefficients of ref. 1. Together these give values for the coefficients A_o and B_o from which we predict the static self-focusing column is given the fraction f_e of n_2 which our results indicate as arising from electronic nonlinearities. (See eq. (10)).

Material	Raman Gain Coefficient (10^{-6} cm/MW) at $1.06\mu\text{m}$	ω cm^{-1}
Fused Quartz	9 ± 1	420
AO LSO	9 ± 1	535
AO C1020	6 ± 1	535
AO C2828	6 ± 1	580
OI ED-2	7 ± 1	615

TABLE II

Maximum stimulated Raman gain coefficients as calculated by reading the maximum Raman cross-sections in Figures (1 - 5) and inserting them in (7). These coefficients have the pump wave intensity in (MW cm^{-2}) divided out and are renormalized to a pump at $1.06\mu\text{m}$. Also listed are the approximate Stokes shifts ω at which this maximum gain would occur.

tive index and its dispersion.⁶ They have successfully correlated IIPC coefficients with the linear index data. That this correlation is reasonable is seen from the predominance of α over $2B_0$ evidenced in Table I, which shows that IIPC is mainly "electronic" in origin in these glasses.

One conclusion to be drawn is that glasses of lower linear index have a lower index for self-focusing and the nonlinear index is a strong function of the linear.⁶

A second conclusion is that Raman scattering and stimulated Raman thresholds are not clearly correlated with the linear refractive index, and are best estimated directly from Raman scattering data.

3. THEORY

To a good approximation the nonlinear polarization $\vec{P}^{NL}(\vec{r}, t)$ of a glass when the macroscopic electric field strength $\vec{E}(\vec{r}, t)$ is well below damage threshold, and contains only frequencies well below the band gap, is ²

$$\begin{aligned} \vec{P}^{NL}(\vec{r}, t) = & \frac{1}{4} \sigma \vec{E}(\vec{r}, t) E^2(\vec{r}, t) \\ & + \vec{E}(\vec{r}, t) \int ds a(t-s) E^2(\vec{r}, s) \\ & + \int ds b(t-s) \vec{E}(\vec{r}, t) \cdot \vec{E}(\vec{r}, s) \vec{E}(\vec{r}, s). \end{aligned} \quad (1)$$

The first term represents the instantaneous nonlinear "electronic" response, that is, the part which would exist if the nuclei were fixed in their average positions, about which the electronic motions deviate from their linear response.

The second and third terms in (1) represent "nuclear" contributions through the two independent scalar time-delay functions $a(t)$ and $b(t)$ allowed by the isotropy of the glasses.

It is easy to verify that if a monochromatic beam at ω of elliptical polarization propagates a distance z through a glass, its polarization ellipse rotates by an angle

$$\theta = \frac{\pi \omega z \langle E^2(t) \rangle (q + 2B_0) \cos 2\phi}{8nc} \quad (2)$$

when $\theta \ll 1$. Here $B_0 = B_\omega$ at $\omega=0$ and B_ω is the Fourier transform of $b(t)$. The tangent of the angle ϕ is the ratio of right to left circularly polarized field amplitudes. The linear refractive index is n and c is the velocity of light in vacuum.

It is also easy to verify that a linearly polarized monochromatic beam experiences a refractive index change (due

to the nonlinear polarization (1))

$$\Delta n = n_2 \langle E^2(t) \rangle \quad (3)$$

where

$$n_2 = \pi \left(\frac{3}{2} \sigma + 2B_0 + 2A_0 \right) / n \quad (4)$$

and $A_0 = A_\omega$ at $\omega=0$ where, similarly as for B_ω , A_ω is the Fourier transform of $a(t)$.

One can also use (1) in Maxwell's equations to discover that a strong linearly polarized "pump" wave of amplitude F at angular frequency ω_0 creates an exponential gain for a weaker superposed wave at $\omega = \omega_0 - \omega'$, whose gain coefficient is

$$g_{||} = \frac{2\pi\omega F^2}{nc} \operatorname{Im} (A_{\omega'} + B_{\omega'}) \quad (5)$$

if the weak wave is polarized parallel to the pump wave, and

$$g_{\perp} = \frac{\pi\omega F^2}{nc} \operatorname{Im} B_{\omega'} \quad (6)$$

if it is polarized perpendicular to the pump wave. This effect is simple "stimulated Raman scattering" whose gain coefficient at ω is known to be related to the ordinary differential Raman scattering cross-section per unit volume $d^2\sigma/d\Omega d\omega$ (cm^{-1}sec) per unit solid angle per unit (angular) frequency interval by ⁷

$$g_{\perp}(\omega) = \frac{\pi^2 c^3 F^2}{\hbar \omega_0 \omega^2 n} \frac{d^2 \sigma_{\perp}(\omega)}{d\Omega d\omega} (1 - e^{-\hbar\omega'/kT}), \quad (7)$$

where the cross section $\sigma_{\perp}$ is for depolarized scattering. The gain $g_{||}$ for parallel polarizations is also given by (7) but with the cross section $\sigma_{||}$ for polarized scattering.

As it is obvious by causality from (1) that $a(t)=b(t)=0$ for $t \leq 0$, we have the relation

$$\operatorname{Re} B_{\omega} = \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{d\nu}{\nu - \omega} \operatorname{Im} B_{\nu} \quad (8)$$

and similarly for A_{ω} . Furthermore, since $\operatorname{Im} B_{\omega}$ is odd in ω , $B_0 = \operatorname{Re} B_0$.

Therefore the quantity B_0 needed in (2) for IIPC and (4) for self focusing is, from (6) - (8)

$$B_0 = \frac{2c^4}{\hbar\omega_0} \int_0^{\infty} \frac{d\nu}{\nu\omega^3} \frac{d^2\sigma_{\perp}(\omega_0 - \nu)}{d\Omega d\nu} (1 - e^{-\hbar\nu/kT}) \quad (9)$$

Similarly, A_0 is given by (9) except with $\frac{1}{2}\sigma_{||} - \sigma_{\perp}$ substituted for $\sigma_{\perp}$.

It is evident from (5) - (7) that Raman scattering cross-sections give directly the Fourier transforms A_{ω} and B_{ω} of the functions $a(t)$ and $b(t)$ needed to completely specify the nonlinear polarization (1). Furthermore, the integrals over the cross sections as in (9) give the coefficients A_0 and B_0 needed in the description of self-focusing and IIPC. The observed coefficients can be compared to the values A_0 and B_0 from Raman scattering to obtain the purely electronic contribution σ_e . The fraction f_e of the self-focusing index n_2 that is electronic in origin is, from (4),

$$f_e = \frac{1}{1 + \frac{4}{3} \frac{B_0 + A_0}{\sigma}} \quad (10)$$

This fraction as calculated from our preliminary Raman data is given in Table I for the four glasses studied.

4. EXPERIMENT

We have measured the absolute Raman cross-sections at frequency shifts of 20 to 2000 cm^{-1} for the glasses under study by comparing these cross-sections with that of the 992 cm^{-1} Raman band of benzene. Our measurements were carried out using the 647.1 line of a krypton ion laser and the 514.5 nm line of an argon-ion laser as the exciting sources. The scattered light was analyzed with a double monochromator and detected with DC techniques.

The value used for the benzene cross-section is that determined by Kato and Takuma for the total polarized integrated cross-section $\frac{d\sigma}{d\Omega}$ of the 992 cm^{-1} band.^{8,9} These authors have measured $\frac{d\sigma}{d\Omega}$ for 514.5 nm and 632.8 nm excitation. The cross-section for 647.1 nm excitation was obtained from that for 632.8 nm excitation by assuming a λ^{-4} variation of the cross-section in that region.

The experimental comparison yielded the absolute polarized cross-section for each glass at one scattering frequency. The spectra were then normalized to this point. The absolute value of the depolarized cross-sections were obtained from a direct measurement of the depolarization ratio at the one frequency, and a subsequent normalization. The scattering frequency of 95 cm^{-1} chosen for these measurements lies near the peak of all the depolarized spectra.

Corrections of the type described by Kato and Takuma were performed to account for the difference in refractive index among the various samples. The index of refraction determines

the reflection losses for radiation entering and leaving the sample. For a given scattering wavelength λ_R and for a given incident laser irradiance the observed signal for one of the glass samples is proportional to

$$S(\lambda_R) \frac{(1-R)^2}{n^2} \frac{d^2\sigma}{d\Omega d\omega} d\Omega$$

Here R is the reflectivity of the sample, (assumed to be the same for incident and scattered light), $\frac{d^2\sigma}{d\Omega d\omega}$ the differential cross-section per unit frequency, n the index of refraction, $d\Omega$ the geometrical collection solid angle, and $S(\lambda)$ is the spectral sensitivity function of the detection system. This function was measured with a quartz-iodine standard lamp whose standardization is traceable to the NBS.

The choice of exciting laser line is complicated by the twin problems of absorption and luminescence. The strong 514.5 nm line of the argon ion laser falls in the middle of a strong Nd^{3+} absorption in all of the three Nd-doped glasses. Hence, it is unsuitable for quantitative measurements of absolute cross-section. A similar situation holds for other strong argon lines. For the 647.1 nm Kr line, on the other hand, the doped samples are almost completely transparent. However, all of the doped glasses show a strong luminescence starting at about 650 nm and growing towards the red. This luminescence was observed in the LSO glass as well, hence, it is not due to the Nd ion. Indeed, the luminescence was a good deal stronger for the undoped glass. We have corrected for this luminescence according to the following procedure. The spectral shape of the luminescence was measured in LSO

glass using 514.5 nm excitation. (Thus, there was no overlap with Raman scattering). For this excitation though, there was a small amount of luminescence observed at the Kr^+ laser wavelength 647.1 nm. Hence, we subtracted this background level from the luminescence spectrum for wavelengths greater than 647.1 nm. From Raman spectra taken with 514.5 nm excitation we concluded that none of the glasses showed any significant amount of scattering for frequencies greater than 1250 cm^{-1} . So, we assumed that all of the signal observed with 647.1 nm excitation and shifted greater than 1250 cm^{-1} was due to luminescence. We then normalized the luminescence spectrum so that it cancelled out the total spectrum for scattered frequencies greater than 1250 cm^{-1} , and subtracted it from the observed total spectrum. This procedure was followed for all three of the Nd-doped glass samples; that is we assumed the luminescence had the same spectral shape for all three of the glasses. This assumption is reasonable since the luminescence is probably due to an impurity present in all the glasses.

For comparison purposes, we also made quantitative measurements of the absolute Raman intensities of fused quartz and LSO glass with 514.5 nm excitation. In Figures 1 - 5, we have plotted both the observed spectra and the spectra corrected for luminescence. The measurements by Stolen and Ippen³ of the polarized Raman spectrum from fused quartz at 514.5 nm give cross-sections which agree with those in Fig. 1B to within 10%.

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FIGURE CAPTIONS

- 1A Absolute Raman cross-sections of fused quartz excited by 614.7 nm. The solid curves are the Raman cross-sections obtained after correction for luminescence. The dashed curves show the cross-sections before the correction was applied.
- 1B Absolute Raman cross-sections of fused quartz excited by 514.5 nm.
- 2 Absolute Raman cross-sections of American Optical Glass C1020 excited by 614.7 nm. The solid curves are the Raman cross-sections obtained after correction for luminescence. The dashed curves show the cross-sections before the correction was applied.
- 3 Absolute Raman cross-sections of American Optical Glass LSG excited by 514.4 nm.
- 4 Absolute Raman cross-sections of Owens Illinois Glass ED-2 excited by 614.7 nm. The solid curves are the Raman cross-sections obtained after correction for luminescence. The dashed curves show the cross-sections before the correction was applied.
- 5 Absolute Raman cross-sections of American Optical Glass C2828 excited by 614.7 nm. The solid curves are the Raman cross-sections obtained after correction for luminescence. The dashed curves show the cross-sections before the correction was applied.

FUSED QUARTZ

Figure 1A

STOKES FREQUENCY SHIFT (cm^{-1})

FUSED QUARTZ

Figure 1 B

STOKES FREQUENCY SHIFT (cm⁻¹)

AMERICAN OPTICAL GLASS LSO

Figure 3

OWENS ILLINOIS GLASS ED-2

AMERICAN OPTICAL GLASS C 2828

Figure 45

STOKES FREQUENCY SHIFT (cm⁻¹)