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ABSTRACT

Commercial thin borosilicate glass sheets have been evaluated for use as a single-shot optic "debris shield" to separate the radiation and contamination produced by the inertial confinement fusion (ICF) experiment from the expensive precision laser optics which focus and shape the 351-nm laser beam which irradiates the target. The goal of this work is identification of low cost materials that can deliver acceptable beam energy and focal spots to the target. The two parameters that dominate the transmitted beam quality are the transmitted wave front error and bulk absorption. This paper focuses on the latter. To date, the materials with the lowest linear 351-nm absorption have also generally demonstrated the lowest non-linear absorption. Commercial materials have been identified which approach the beam energy and focus requirements for many ICF missions.

Keywords**:** borosilicate glass, non-linear absorption, ultra-violet light

1. INTRODUCTION

The interaction of high intensity laser light with glass can result in both reversible and permanent changes in optical properties. Excellent treatises on (reversible) two-photon absorption in dielectric materials can be found in Chase[1] and Smith[2]. The interaction of solar or laser radiation can also produce free electrons or holes which can be trapped in structural or chemical defects in the glass resulting in 'color centers' or 'solarization'. Bishay[3] and Griscom [4,5] published detailed discussions of color centers induced in multicomponent glasses including borosilicates. The linear and non-linear optical properties of potential fusion-laser materials were investigated extensively in the early 80's[6]. Non-linear absorption, solarization, bulk damage (platinum inclusions) and nonlinear refractive index all limit the utility of borosilicate optical glasses, such as Schott BK-7, as thick transmissive optics in high-peak-power applications at 351-nm. We propose that thin commercial borosilicate glass sheets could function as low cost optics that can deliver acceptable beam energy for a limited shot sequence on an ICF laser. In order to assess the feasibility of specific commercial thin glass sheets, we have measured their transmittance under irradiance by 351-nm, 3-ns laser pulses at various fluence levels.

2. EXPERIMENT

2.1 Materials preparation and measurements

All materials were tested as-received from the vendor. With the exception of the Hoya NA-35 substrates, all test sample surfaces were as-formed – i.e. drawn or float glass surfaces. The Hoya NA-35 substrates were mechanically polished after forming. All optics were cleaned, but not anti-reflection coated before testing. Material composition is shown in Table 1.

Table 1: Test matrix materials

Sample ID	Test Thickness (mm)	Forming process	Type	Major constituents (in addition to SiO2)
Schott Borofloat®[1]	1.1 and 3.3	float	borosilicate	B_2O_3, Na_2O, Al_2O_3
Schott D263[1]	1.1	drawn	zinc borosilicate	B_2O_3,Na_2O, K_2O, ZnO, Al_2O_3,TiO2
Corning 0211[2]	0.4	drawn	zinc borosilicate	B_2O_3,Na_2O, K_2O, ZnO, Al_2O_3,TiO2
Hoya NA-35[3]	0.9	Drawn & polished	aluminosilicate	Al_2O_3, B_2O_3, BaO, CaO, SrO
Schott BK-7	for reference	cast	borosilicate crown	B_2O_3,Na_2O, K_2O, BaO

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[2] Corning Inc., Corning, NY
[3] Hoya Corporation USA, Fremont, CA

2.2 Test facilities

Two test facilities were used in these experiments. Initial experiments were conducted on Corning 0211 zinc borosilicate glass sheet on our Optical Sciences Laser (OSL) using a setup similar to that previously described in Whitman[8]. The Q-switched Nd-glass OSL laser produces 1053-nm pulsed output that can be frequency tripled to 351-nm; the output beam has an approximate "top-hat" temporal and spatial intensity profile. Figure 1 shows the test system and diagnostics layout. Dichroic mirrors were used to separate the 351-nm beam from the residual light at 1053 and 527 nm. A plane near the KDP crystals was imaged by a single lens onto the sample. Input diagnostics allowed measurement of the pulse energy, temporal waveform, and the fluence distribution in an "equivalent sample plane". Fractions of the output beam were observed by a calorimeter and a photodiode that were used to monitor relative changes in the laser output that occurred when the sample was inserted into the beam and when the fluence in the input beam was increased. The output beam pulse length was 3-ns at 351-nm.

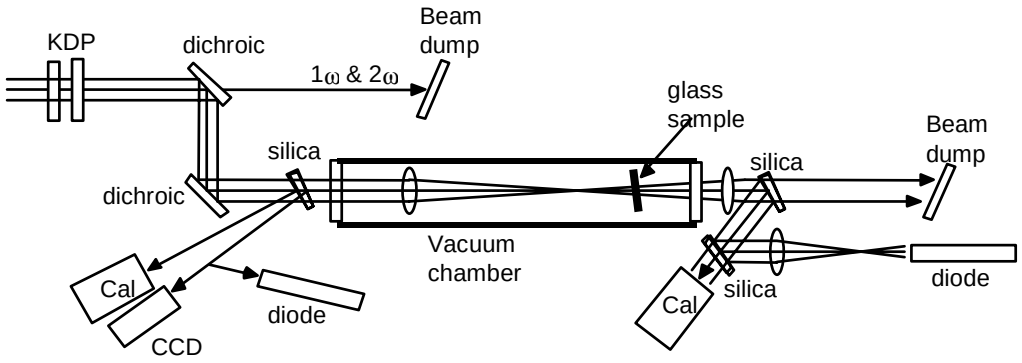


Figure 1. Experimental arrangement for the Optical Sciences Laser (OSL)

The second test facility was assembled specifically for these experiments. A Coherent Infinity tripled Q-switched Nd:YaG laser provided the ~ 3-ns (FWHM) 355-nm Gaussian (temporal and spatial) test beam. Figure 2 shows the layout for this experiment. Vacuum photodiodes and a boxcar integrator recorded the intensity of each 3-ns, 355-nm pulse as a voltage. The transmittance was determined by the ratio of voltages before and after the sample and normalized by the ratio without a sample in place. All samples were illuminated at a two-degree angle of incidence.

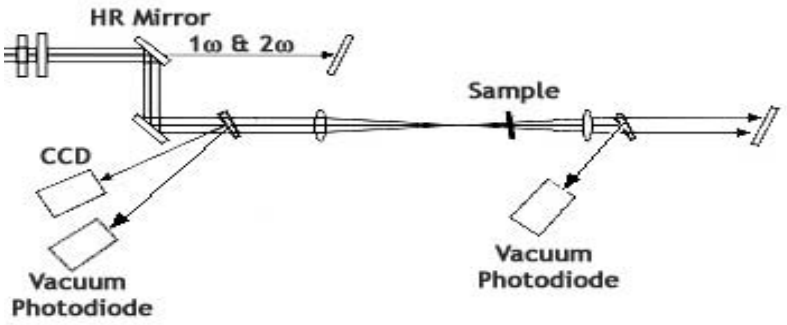


Figure 2: Experimental arrangement for the Coherent Infinity laser experiments

3. RESULTS

3.1 Transmittance of a 351-nm flat pulse

Figure 3 gives the results of our measurements of transmittance as a function of incident 351-nm fluence for the 3-ns flat beam produced in the OSL experimental setup described above. Transmittance was defined to be the ratio of output to input energy that was measured with the sample in the beam, divided by a value of the same ratio that was measured before the sample was installed. From a line that was fitted through the data, we estimate that the transmittance varied from about 88% to 76% as the fluence increased from 0 to 10 J/cm^2. The low power measurement of 88% is close (within error bars) to the

spectraphotometer transmission measurement of 90%. The linear variation of the transmission with 351-nm fluence is indicative of a two-photon absorption process whose source has not yet been identified.

Although there was no visible surface damage to the sample for these single shot irradiation experiments, there was visible sample discoloration ('browning') for incident fluences around 5 J/cm^2 or higher. This is possibly some form of color center formation. The sample discoloration increased with increasing fluence levels. We have not done a systematic study of its causes or tested whether thermal bleaching could recover the original sample transmittance.

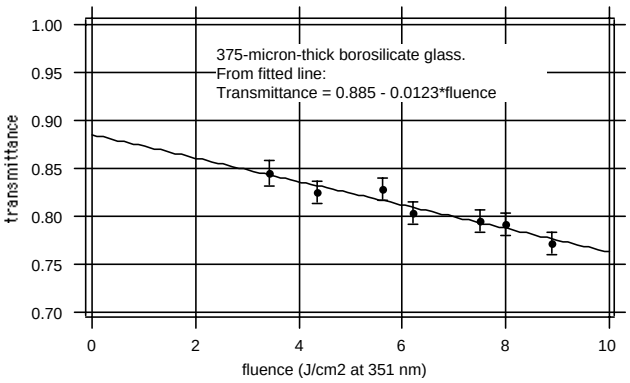


Figure 3. Transmittance of unpolished borosilicate glass sheet (Corning 0211) as a function of incident fluence in a 3-ns, 351-nm pulse.

For each shot, waveforms were captured by SCD50000 digitizers. Figure 4 shows the waveforms for one shot that was recorded before the sample was loaded, and for all seven shots that were used to determine the transmittance. They were normalized such that voltages within the half-power points have average value of unity. The systematic variation of these waveforms with increase in fluence is slight. At low fluence the output power tended to be slightly low at the first of the pulse and slightly high at the end of the pulse. That trend was reversed for shots at highest fluence, see Fig. 4. However, the power reduction at the end of the pulse was not as large as the 8% calorimetric deficit. The absence of pulse shape distortion also indicates that the cause of the transmittance loss is most likely a bulk effect (such as a two-photon-induced absorption process) in the glass itself.

3.2 Transmittance of a 355-nm Gaussian pulse

Figure 5 shows the results of our measurements of transmittance as a function of incident 355-nm fluence for the ~ 3-ns Gaussian pulse. All of the samples measured exhibited behavior that is characteristic of nonlinear absorption. Several displayed a linear variation in transmittance as a function of fluence that is indicative of two-photon absorption. Of the samples examined, Hoya NA-35, 0.9 mm in thickness, had the least loss overall. The transmittance varied with intensity from 89% at 4 J/cm^2 to 87% at 12 J/cm^2.

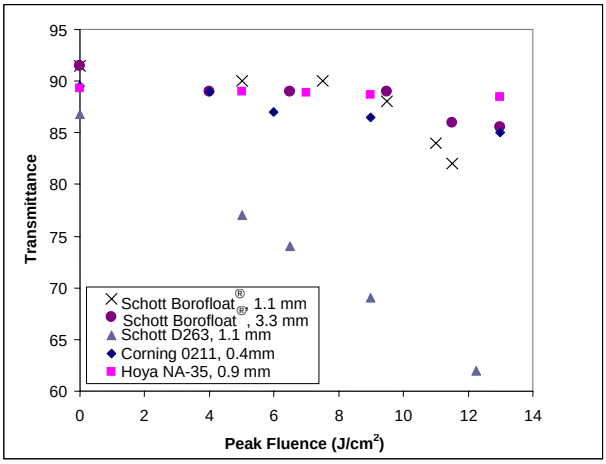


Figure 5: Comparison of transmittance of thin borosilicate glass sheets as a function of incident fluence in a 3-ns FWHM Gaussian 355-nm pulse.

Table 2 summarizes the result of linear least squares regression fits to the data. The extrapolated y-intercept should represent the linear absorption plus the Fresnel reflection losses; for most samples there is good correlation between the transmittance measured on a spectrophotometer and the y-intercept. For borosilicate materials (refractive index around 1.53), Fresnel losses limit the maximum transmittance to no more than ~90%. Extrapolated y-intercepts greater than 90%, in combination with the poor linear fit suggest that non-linear absorption cannot fully account for the observed transmittance of the Schott Borofloat® samples. In this experimental setup, the beam size was approximately 1-mm FWHM – so small surface damages or 'browning' would most likely be undetectable without magnification.

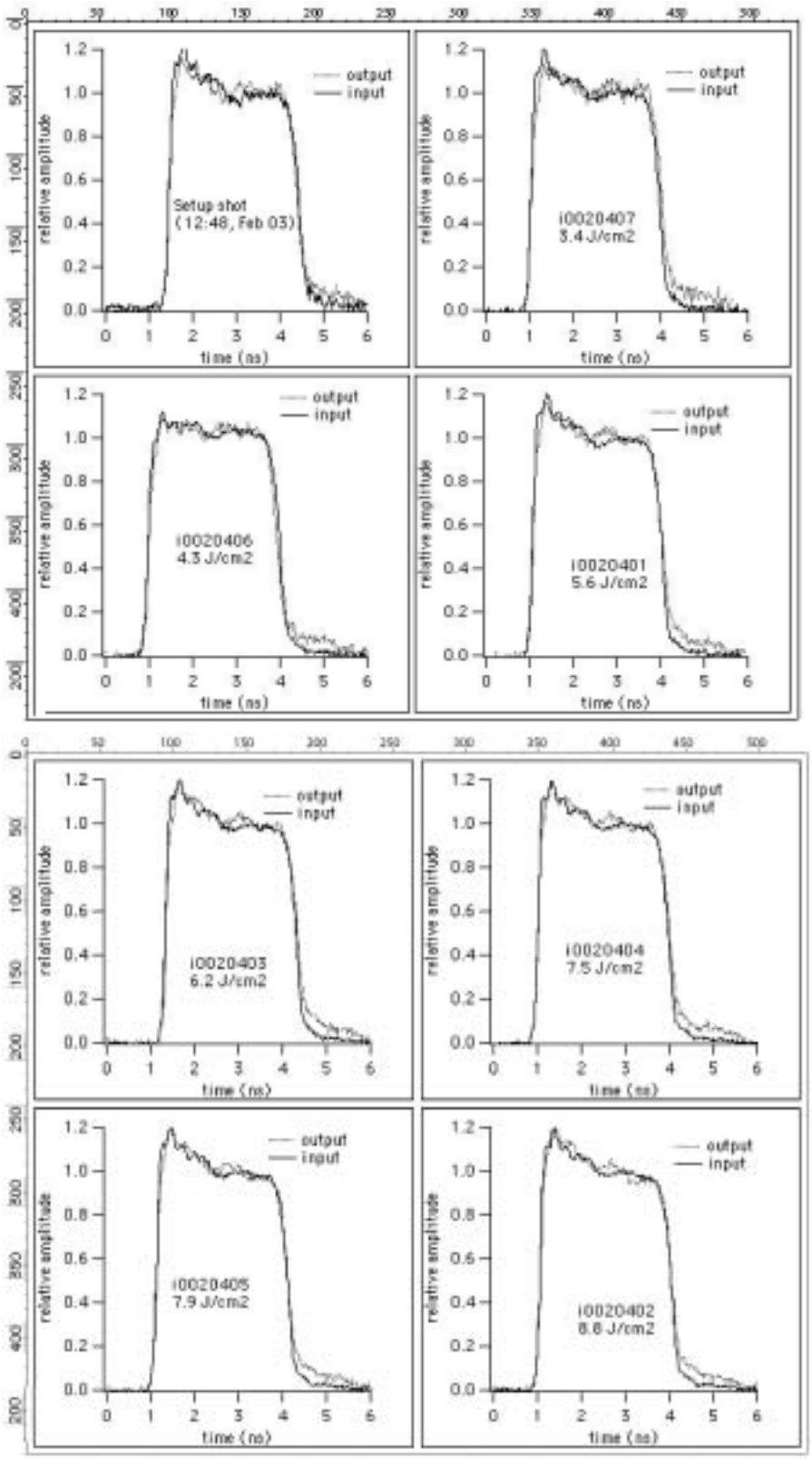


Figure 4. Comparison of the 351-nm input and output pulses through a 375-μm-thick zinc-borosilicate glass sheet (Corning 0211) at various fluence levels. The frame at the upper left is without the sample in the beam path and provides a measure of the errors in the pulsed shape measurement.

Table 2: Comparison of transmittance parameters for four different commercial borosilicate glasses

Test sample	Measured transmittance[1]	Extrapolated "zero-fluence" intercept	Intensity-dependent absorption loss at 355-nm (%/J/cm^2)	Quality of linear fit (R^2)
1.1mm Schott Borofloat®	*91.5%*	*96.2%*	*-1.0*	*0.878*
3.3mm Schott Borofloat®	*91.7%*	*91.4%*	*-0.40*	*0.784*
1.1mm Schott D263	*84.8%*	*86.8%*	*-1.97*	*0.996*
0.4 mm Corning 0211	*89.8%*	*84.7%*	*-0.30*	*0.905*
0.9mm Hoya NA-35	*91.2%*	*89.3%*	*-0.02*	*0.797*

3.3 Extraction of non-linear absorption coefficient

In order to predict material performance for different irradiance pulse shapes, we must extract the non-linear absorption coefficient (β) from the raw data. To do this, both the temporal and spatial profiles of the incident laser pulse must be taken into consideration. Following Smith[2], the change in intensity, I, propagating in the z-direction through an optic with one- and two-photon absorption coefficients α and β, and one-and two-photon-induced solarization s_1 and s_2 respectively is described by equation 1:

$$dI/dz = -\alpha I - \beta I^2 - s_1 I - s_2 I \qquad (1)$$

Ignoring the solarization terms, the total transmittance through thickness L is becomes:

$$T = T_L T_{NL} = I(z)/I_{INC} \qquad (2)$$

where I_{inc} and I(z) are the incident and transmitted intensity, respectively, and the linear transmission factor, T_L and the non-linear transmission factor, T_{NL}, are defined as follows:

$$T_L = (1-R)^2 e^{-\alpha z} \qquad (3)$$

$$T_{NL} = (1+Q)^{-1} \qquad (4)$$

and

$$Q = \beta(1-R) I_{INC} (1-e^{-\alpha z})/\alpha \qquad (5)$$

where R is the Fresnel surface reflectance. Then for a flattop –square pulse incident beam, such as we have on OSL,

$$1/T = 1 + \beta I_{INC} z \qquad (6)$$

and the slope of a plot of 1/T vs. incident intensity yields the non-linear absorption coefficient, β, directly. For a flat-square spatial and temporal profile, the total transmittance, T_F, can be rewritten:

$$T_F = T_L \frac{1}{1+Q} \qquad (7)$$

In contrast, the total transmittance for a Gaussian-Gaussian spatial and temporal profile, T_G, is given by Smith[2] as:

$$T_G = T_L \sum_{n=1}^{\infty} (-Q)^{n-1} n^{-1.5} \qquad Q<1 \qquad (8)$$

To convert the Gaussian beam data to the transmittance expected for a flat beam, we must calculate Q and then multiply the measured transmittance by the ratio of T_F/T_G. Figure 6 shows results from such a calculation for the Corning 0211 zinc borosilicate sample. The excellent agreement between the flat beam and the Gaussian beam experiments gives us confidence that the non-linear absorption coefficients extracted using the small-beam Gaussian experiment setup are reliable.

To determine the non-linear absorption coefficient β for the remaining materials, we use the following algorithm. T_L is known (measured). Guess β, calculate Q and T_G. Minimize the difference between the measured and calculated transmittances, T. The results from these calculations are shown in Figures 7a – c, and summarized in Table 3. For comparison, one- and two-photon absorption coefficients for BK7 and BK10 glasses are also included in Table 3.

Two different sample thicknesses were measured for one material (Schott Borofloat®). Unfortunately, this was the only material which deviated substantially from the bulk absorption model, as is evident from the higher losses observed in the thinner (rather than the thicker) part as well as the deviation from linear fit in Fig. 7c. Although surface damage was not evident with the naked eye, it cannot be ruled out. Alternately, this particular glass is manufactured by a float process and it is possible that a thin surface contamination layer could have strong non-linear absorption. We have reported non-linear absorption coefficients for this material, but one can at best interpret this data as an upper bound.

Test Sample	UV-edge	α/cm	β cm/GW
Schott Borofloat®, 1.1 mm thick[1]	4.3 eV	0.016	0.435
Schott Borofloat®, 3.3 mm thick[1]	4.3 eV	0.032	0.110
Hoya NA-35, 0.9 mm thick[1]	4.4 eV	0.118	0.360
Corning 0211, 0.4 mm thick[2]	3.9 eV	0.937	1.245
Corning 0211, 0.4 mm thick[1]	3.9 eV	0.637	0.950
Schott D263 , 1.1 mm thick[1]	3.9 eV	0.708	2.376
Schott BK7[3]	4.4 eV	0.036	0.006
Schott BK10[3]	4.7 eV	0.007	0.0045
Schott BK10, antisolarant[3]	4.7 eV	0.013	0.01

Table 3: UV-cutoff edge (5 cm^{-1}), linear and non-linear absorption coefficients for test matrix are compared to literature values for BK optical glasses. [1]Calculated from this work, 3-ns Gaussian beam. [2]Calculated from this work, 3-ns Flat Pulse (OSL). [3]from Smith[6].

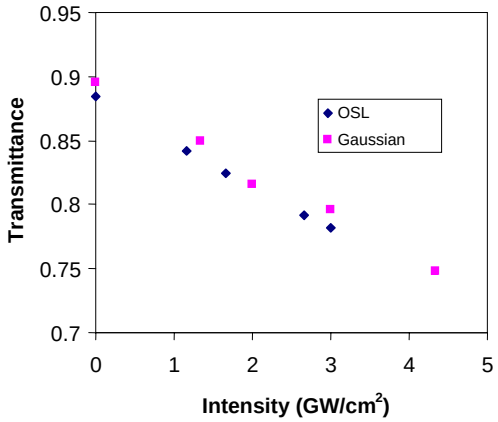
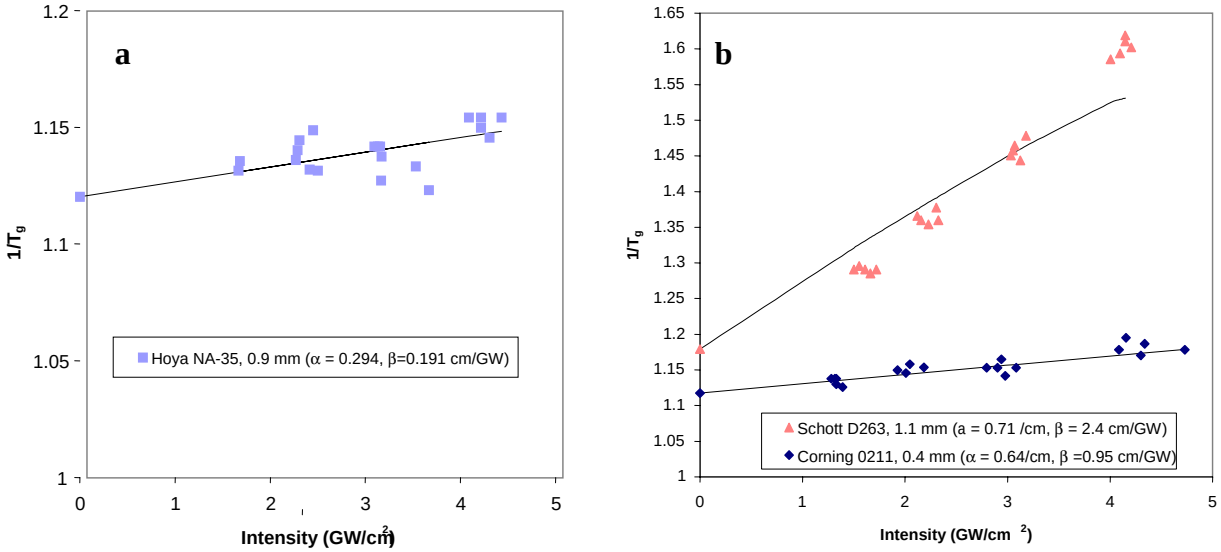


Figure 6. Comparison of Gaussian-Gaussian beam data converted to flat-square beam (α = 0.94, β= 1.24) vs. data measured on OSL using flat-square beam for Corning 0211 borosilicate glass sheet.



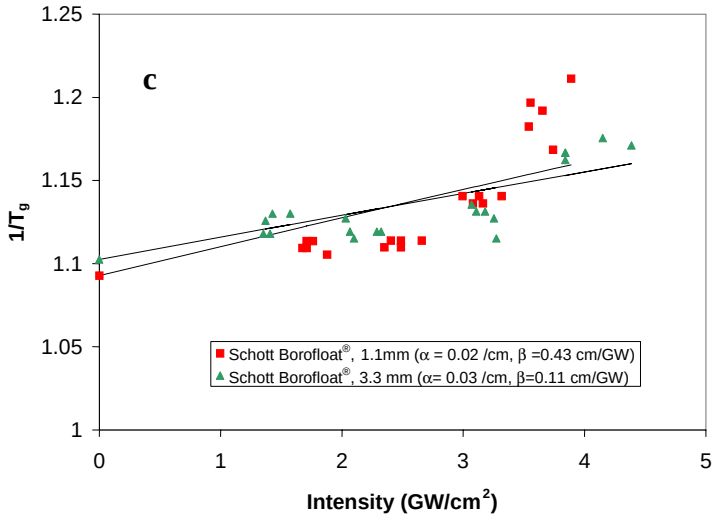


Figure 7: Transmission of 3-ns flat-square pulse through a) 0.9mm Hoya NA-35 aluminoborosilicate; b) through 1.1mm Schott D263 and 0.4mm Corning 0211 zinc borosilicate, and c) 1.1mm and 3.0mm Schott Borofloat® borosilicate. Lines represent regression fit to the transformed data; slope and intercept are 2-photon and 1-photon absorption coefficients β and α, respectively.

4. DISCUSSION

Published values for two-photon absorption in transparent dielectric materials, such as the borosilicates that were measured in this work, are typically 1 – 2 orders of magnitude smaller than our reported non-linear absorption coefficients. In the case of the Schott Borofloat® material, it is clear that linear and non-linear absorption alone cannot explain our observed losses. In this case, it is suspected that surface loss is a dominating influence. But the remaining samples follow a linear fit with 1/T, suggesting linear and non-linear absorption is the dominating loss mechanism. Because we have good correlation between the flat and Gaussian beam experiments conducted in two different laboratories, it is unlikely that the measurements themselves are awry. Rather it is more likely that we are measuring a combination of two-photon and other excited-state absorption phenomena that follow the nonlinear absorption. The observation of sample discoloration ('browning') after a single shot is confirmation that at least some long-lived induced absorption is occurring.

To first order, non-linear absorption in our commercial borosilicates appears to be strongly correlated with linear absorption, see Figure 8. Linear absorption is expected to follow the Urbach rule[9]: the log of the absorption coefficient is proportional to the difference between the energy of the incident photon and the fundamental band gap. For all materials tested, the two-photon energy (6.99 eV) exceeds the UV-edge (5 cm^{-1}), and two-photon absorption is energetically expected.[1]

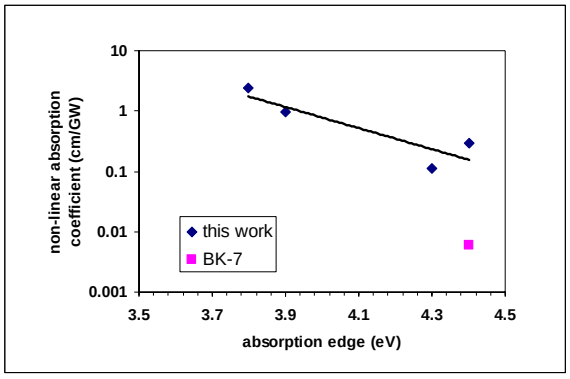


Figure 8. The non-linear absorption coefficient β is strongly correlated with the UV absorption edge (5 cm^{-1}).

The two materials with highest non-linear (and highest linear absorption) are both zinc borosilicates. The complex interaction of melting conditions, photochemical reactions, structure, multivalent additives, and trace impurities which lead to induced absorption make it difficult to predict performance of these multi-component glasses a priori. Efimov[10,11] investigated the role of iron impurities at concentrations up to 0.1% in multi-photon absorption and photo-induced changes in crown alkali silicate glasses such as Schott BK-7. The presence of arsenic or antimony can enhance solarization[12] by providing an effective trap for the ionized electrons produced for example by photo-ionization of "colorless" Fe^{2+} to the strongly-absorbing Fe^{3+}. The materials in this study are all used in optical or flat panel applications where visible absorption must be kept to a minimum. Hence, the concentrations of known colorants such as iron are at the ppm level, and the vendor-to-vendor variation that we see appears to be driven more by base glass composition and forming conditions than impurities per se. Indeed, Figure 9 shows no correlation of absorption with any of the common trace elements. Compared to pure amorphous silica, B and Al would be expected to lower the UV cutoff wavelength; Ti, Li, Na, K, and Ba would be expected to raise the UV cutoff (although much less effectively than Fe^{3+}) and Zn should either raise or have no significant effect on UV-cutoff depending upon forming conditions and other constituents[13].

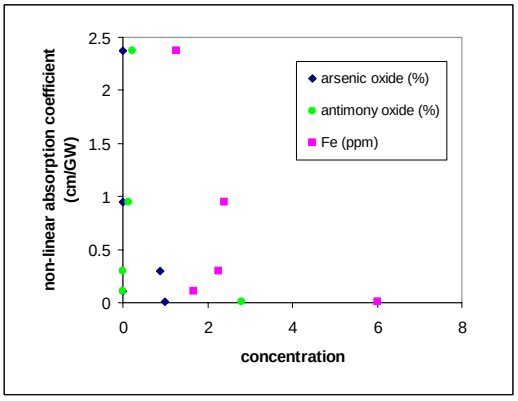


Figure 9. There is no strong correlation between non-linear absorption and Fe concentration with or without As and Sb.

5. CONCLUSIONS

We have measured linear and non-linear absorption losses for commercial borosilicate glasses. Observed losses include linear and non-linear absorption as well as scatter from damage during the pulse. A strong correlation was found between linear absorption and transmittance at 351-nm for laser intensities in the GW/cm^2 range. Several promising debris shield candidates have been identified for single (or a few) shot service at these high intensities.

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