Intensity-dependent loss properties of window materials at 248 nm

P. Simon* and H. Gerhardt

Laser-Laboratorium Göttingen e.V., Robert-Bosch-Breite 10, D-3400 Göttingen, Federal Republic of Germany

S. Szatmári†

Max-Planck-Institut für biophysikalische Chemie, Abteilung Laserphysik, Postfach 28 41, D-3400 Göttingen, Federal Republic of Germany

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Transmission of fused silica, CaF_2, LiF, and MgF_2 is measured using 450-fsec, 248-nm pulses in the range 10–120 GW/cm^2. Different loss mechanisms such as scattering of transmitted radiation, color-center formation, and multiphoton absorption were studied separately. For fused silica a two-photon absorption mechanism is found, while for CaF_2, LiF, and MgF_2 three-photon absorption and absorption due to color-center formation are found as dominant absorption mechanisms.

With existing high-brightness KrF laser systems, optical powers of the order of typically 10–100 GW are generated.[1–7] The power densities of the output beams of these systems typically range from 10 to 100 GW/cm^2. At those high power densities nonlinear-optical properties of window materials become important.

Previous studies of two-photon absorption were performed at 355 and 266 nm.[8] Measurements of nonlinear absorption of window materials at 248 nm were recently reported in Refs. 6, 9, and 10. However, the relative contributions of the different loss mechanisms (multiphoton absorption, color-center formation, and scattering) were not considered. In Ref. 8 the two-photon absorption mechanism found for fused silica was simply adapted for CaF_2. Surprisingly, the two-photon coefficient of CaF_2 was dependent on the sample studied. Even in the case of LiF and MgF_2, where two-photon absorption is improbable, an upper bound for a two-photon absorption coefficient was defined. In addition, the results reported in the publications cited above are not in total agreement.

The above problems and the importance and necessity of having exact data led us to carry out comparative measurements on the intensity-dependent transmission of UV windows of different materials and from different suppliers. In this Letter we report on the nonlinear loss mechanisms observed in fused silica, CaF_2, LiF, and MgF_2 using 450-fsec pulses at 248 nm. While for fused silica our results show good agreement with the two-photon absorption mechanism found in Refs. 8 and 9, we found that the data for CaF_2, LiF, and MgF_2 cannot simply be approximated by two-photon absorption but can be described by the combined effect of three-photon absorption, scattering, and color-center formation.

For the transmission measurements, 8-mJ, 450-fsec pulses at 248 nm from a high-power KrF laser system[6] were used. The pulse energy is measured with a Gentec ED-500 detector. The pulse duration is determined by an autocorrelation measurement using two-photon ionization in NO.[2,6,11,12] From the autocorrelation trace a ±4% fluctuation is obtained for the energy and the pulse width. The amplified spontaneous emission is ~5% of the total energy.

The experimental setup is as follows: The relatively homogeneous middle part of the beam is selected by an approximately 3.5-mm-diameter circular aperture. The plane of this aperture is imaged onto the sample by a 1-m focal-length fused-silica lens. The spot size and the intensity distribution on the sample were checked by a linear diode array placed in the position of the sample. The intensity incident upon the sample is varied by a variable attenuator put just before the aperture. The sample serves as output window of an evacuated tube that is used to prevent air breakdown in the focus. The CaF_2 input window of this tube is just behind the imaging lens.

The transmitted energy through the sample is measured by a Laser Precision RJP 735 pyroelectric detector attached to a sample-and-hold circuit and a chart recorder at a series of well-defined settings of the variable attenuator. The energy incident upon the sample is measured at the same settings of the attenuator. For this measurement the input window of the vacuum tube is disassembled and placed into the beam with the energy meter just behind it. The intensity on the sample ranged from 10 to 120 GW/cm^2. The values of initial transmission corresponding to zero intensity were determined using a Perkin-Elmer Lambda 7 spectrophotometer. The pulse duration seen by the sample was determined by an autocorrelation measurement at the position of the sample. It is worth noting that the optical components traversed by the beam broadened the pulse duration from 400 to 450 fsec through group-velocity dispersion.

The results of the transmission measurements showed that the overall transmission of the samples is given by the combined effect of multiphoton absorption, light scattering, and color-center formation. In

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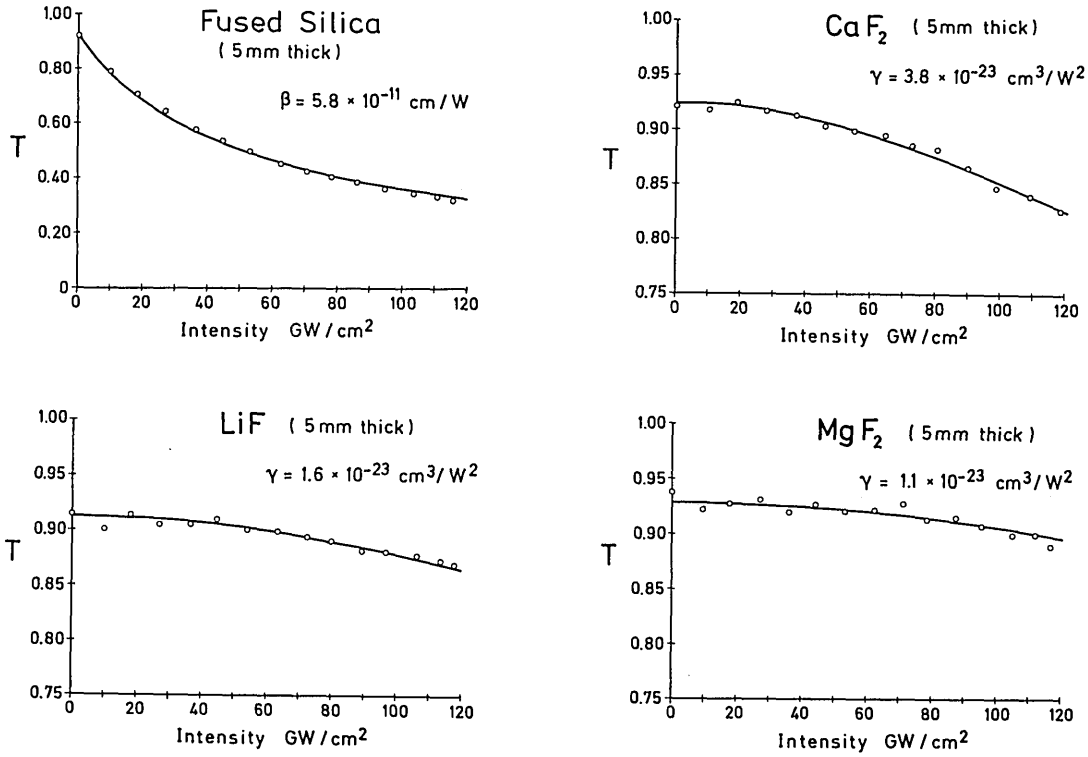

Fig. 1. Measured values of transmission as a function of intensity for fused silica, CaF_2, LiF, and MgF_2. The solid curves are the best fits to the measured data assuming a two-photon absorption mechanism for fused silica and a three-photon absorption mechanism for CaF_2, LiF, and MgF_2.

order to get information on the net effect of multiphoton absorption, the sample was illuminated only for a limited number of shots, until absorption due to color-center formation became important. Care was taken to collect all the transmitted energy, including the scattered part, which was distributed in a solid angle of $\sim 10^{-2}$ sr. The transmission values showing the net effect of multiphoton absorption are indicated in Fig. 1 for different materials as a function of power density incident upon the sample. The fused-silica sample was Suprasil, supplied by Heraeus, the CaF_2 samples were from Oyokoden and Caramant, and the LiF_2 and MgF_2 samples were from Korth.

For fused silica the measured data can be fitted assuming a two-photon absorption process, while for CaF_2, LiF, and MgF_2 the best fits are found assuming a three-photon absorption mechanism. To determine the two-photon and three-photon absorption coefficients, we assumed that only a single multiphoton process is involved and that the input intensity $I_{in}(t)$ is spatially uniform and temporally $\mathrm{sech}^2(t)$ shaped. The general expression describing the change of intensity as the pulse propagates through a sample is $dI/dz = -\alpha I^n$, where α is the absorption coefficient and $n = 2$ and 3 denotes two-photon and three-photon absorption, respectively. The solution of this equation, assuming a $\mathrm{sech}^2(t)$ pulse shape and a reflection R at each surface of the sample of thickness l, is

$$I_{out}(t) = \frac{I_i(1-R)^2}{\{(n-1)\alpha l[(1-R)I_i]^{n-1} + [\mathrm{sech}(t)]^{-2(n-1)}\}^{1/(n-1)}}, \quad (1)$$

where I_i is the peak input intensity. By integrating Eq. (1), the transmission can be calculated as

$$T = \frac{(1-R)^2}{2} \times \int_{-\infty}^{\infty} \frac{dt}{\{(n-1)\alpha l[(1-R)I_i]^{(n-1)} + [\mathrm{sech}(t)]^{-2(n-1)}\}^{1/(n-1)}}. \quad (2)$$

Best fits of Eq. (2) to the measured data are given in Fig. 1 by the solid curves. For the absorption coefficients, the values listed in Table 1 are found. The relative uncertainty of the transmission measurements is ±10%. (Note that each experimental point in Fig. 1 represents an average over 10 shots.) The γ value of CaF_2 is practically independent of the samples from different suppliers.

The interpretation of the results is straightforward for fused silica, LiF, and MgF_2. For fused silica, since its band-gap energy (7.8 eV) lies well below the energy of two 248-nm photons (10 eV), two-photon absorption is permitted. For LiF and MgF_2, having band-

Table 1. Multiphoton Absorption Coefficients at 248 nm

Material	Absorption Mechanism	Absorption Coefficient
Fused silica	$n = 2$	$\beta = 5.8 \times 10^{-11}$ cm/W
CaF_2	$n = 3$	$\gamma = 3.8 \times 10^{-23}$ cm³/W²
LiF	$n = 3$	$\gamma = 1.6 \times 10^{-23}$ cm³/W²
MgF_2	$n = 3$	$\gamma = 1.1 \times 10^{-23}$ cm³/W²

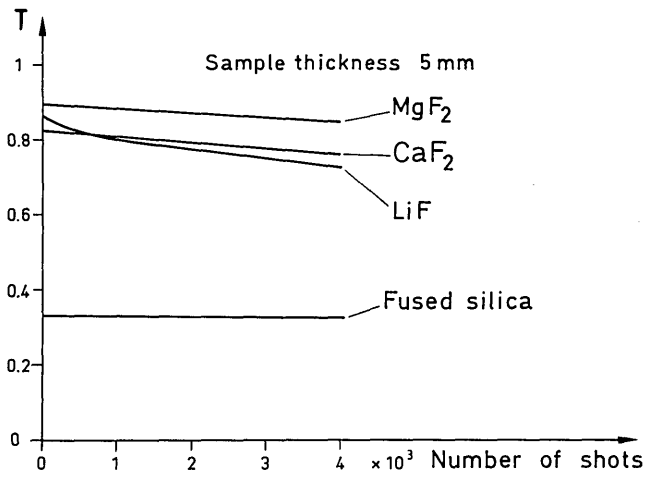

Fig. 2. Transmission of fused silica, CaF_2, LiF, and MgF_2 as a function of the number of shots at 120 GW/cm^2.

Table 2. Ratio of the Scattered to the Incident Energy for Different Materials at 120 GW/cm^2

	Fused Silica	CaF_2	LiF	MgF_2
$\frac{I_{\text{scattered}}}{I_{\text{incident}}}$	0.06	0.33	0.05	0.11

gap energies of 11.6 and 11.8 eV, respectively, two-photon absorption is not allowed but three-photon absorption is certainly possible.

The situation with CaF_2 is somewhat more complicated, since its band-gap energy (10 eV) is just equal to the two-photon energy of 248-nm radiation. However, one might expect that the probability of two-photon absorption is not a steplike function of the photon energy but can be described by a continuous function that reaches its maximum only at higher photon energies. This means that the two-photon absorption coefficient of CaF_2 just at 248 nm is probably small compared with the three-photon coefficient, which can explain the observed behavior.

Besides the multiphoton processes, the other loss mechanisms, excluded in the above measurements, were also studied in a separate measurement. One of these processes is absorption due to color-center formation. We tested this effect at 120 GW/cm^2. In Fig. 2 the change of transmission of the samples is displayed as the number of pulses incident upon the same area of the sample increases. This implies that for a careful measurement of multiphoton absorption, especially at power densities exceeding ~100 GW/cm^2, a new area must always be irradiated and only the first few shots give true information on the material constants.

An additional observation is strong scattering of transmitted radiation at powers exceeding ~100 GW/cm^2. This effect is likely due to nonlinear refraction. Considering that the intensity distribution is not flat-topped but modulated, any kind of intensity-dependent refractive-index change results in significant scattering. At 120 GW/cm^2 we measured that portion of the transmitted energy that travels in the original direction defined as the direction of the beam at low (10 GW/cm^2) intensity. The remaining scattered portion we then related to the incident energy, as listed in Table 2. Although the values in Table 2 are also connected to our specific beam profile, their relative magnitudes are characteristic of the different materials.

In conclusion, intensity-dependent loss mechanisms occurring in window materials at 248 nm were studied. It is found that the loss is partly due to light scattering and absorption. Absorption in fused silica is mainly two-photon absorption, while in CaF_2, LiF, and MgF_2 the combined effect of color-center formation and three-photon absorption must be considered. The evolution of the absorption due to color-center formation and the values of the two- and three-photon absorption coefficients for the different materials are also given.

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* Permanent address, Department of Experimental Physics, Jozsef Attila University, Dóm tér 9, H-6720 Szeged, Hungary.

† Permanent address, Research Group on Laser Physics of the Hungarian Academy of Sciences, Jozsef Attila University, Dóm tér 9, H-6720 Szeged, Hungary.

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