Nonlinear Processes in UV Optical Materials at 248 nm

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Abstract. Nonlinear processes in UV optical materials were investigated by using 280-fs, 248-nm pulses. Nonlinear absorption in CaF$_2$ was confirmed to be a two-photon process by using the luminescence of self-trapped excitons, which was also used for the single shot pulse width measurement. The absorption bands due to F centers were identified in CaF$_2$, MgF$_2$, and LiF after several hundred shots at 100 GW/cm$^2$. Absorption at 248 nm was considerable especially in MgF$_2$ and LiF. Self-focusing and self-phase modulation were observed in CaF$_2$.

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A multiterawatt KrF excimer laser (248 nm) is an attractive light source for studying multi-photon processes and X-ray lasers [1]. Many efforts have been devoted to achieve high peak power in KrF lasers [2–5]. A peak power of up to 4 TW has been obtained with an active cross section of 320 cm$^2$ [6]. More recently a TW-class KrF excimer was operated at 10 Hz with a moderate aperture size of 15–30 cm$^2$ in our institute [7]. For such high-power laser systems, the nonlinear effects in window materials such as absorption, color center formation and self-phase modulation are important because they place a potential limit on the achievable peak intensity.

The nonlinear absorption coefficients of UV window materials have been measured by several authors [8–10]. At the KrF wavelength, there are only a few UV materials available including CaF$_2$, MgF$_2$, and LiF whose band gaps are greater than twice the 5 eV photon energy. Especially in CaF$_2$, the band gap is almost the same as the two-photon energy. In this case, it is quite difficult to determine the order of nonlinear absorption from the simple measurement of the transmittance. The inverse transmittance varies only in a small range as an input intensity while spatial and temporal fluctuations of the input beam can easily result in large errors. Moreover, the effects of other processes, such as self-focusing and color center formation, cannot be discriminated at a high intensity region. In this paper, the luminescence of self-trapped excitons was successfully used to confirm nonlinear absorption in CaF$_2$ as a two-photon process. This luminescence was also used for the single-shot pulse width measurement of subpicosecond KrF laser pulses. In previous papers the two-photon fluorescence at 172 nm of XeF and the three-photon fluorescence at 480 nm of XeF were used for this purpose [11, 12]. In the method reported in this paper, the vacuum system and gas supply are not necessary. The contrast ratio of the autocorrelation trace was nearly 3, which is further evidence of a two-photon process.

Other nonlinear effects such as color center formation, self-focusing and self-phase modulation were investigated for CaF$_2$, MgF$_2$, and LiF. Color center formation is serious especially for high repetition rate operation because it accumulates during operation. Electron-hole pairs created by nonlinear absorption not only relax through the luminescence of excitons and nonradiative recombination, but also form color centers. In CaF$_2$, color centers were visible along the beam path after a few shots at a high intensity. The absorption spectrum of these color centers was due to an F-center band with a peak at 380 nm. In MgF$_2$ and LiF, the F-center absorption bands were observed with peaks around 248 nm.
Self-focusing, which reduces beam uniformity and focusable power, was observed in CaF$_2$. The spectral broadening due to self-phase modulation was observed when self-focusing occurred in the sample. Detailed results will be presented in later sections.

1. Measurement of Nonlinear Absorption Coefficients

Initially, two-photon absorption coefficients were measured for three materials, 1-cm thick fused quartz, 1.5-cm thick CaF$_2$ and 0.6-cm thick MgF$_2$. The experimental setup was essentially the same as in [10]. A 1.75-m focal-length mirror was used for soft focusing. An energy monitor of the transmitted light was placed well before the focal point to eliminate self-focusing in the air. The subpicosecond laser was described in detail in [13].

The spectrum of the subpicosecond pulse used in this experiment is shown in Fig. 1. The pulse width of each shot was measured by the three-photon fluorescence method [12], resulting in $280 \pm 40$ fs throughout the experiment. The beam profile was nearly of flat-topped shape. In Fig. 2, we show a set of data plotted with measured values of inverse transmittance ($1/T$) versus laser intensity. The intersection of the $1/T$ axis was fixed to be $1/(1-R)^2$ because linear absorption was negligible; here $R$ is the surface reflectance. The slope of the straight line gives the two-photon absorption coefficient ($\beta$). The reported band gap of fused quartz is 7.8 eV [9], which is greater than the two-photon energy at 248 nm (10.0 eV). Liu et al. measured the two-photon absorption coefficients of UV window materials at 266 nm (the two-photon energy of 9.36 eV). The value of $\beta$ in fused quartz was $1.7 \times 10^{-11}$ cm/W, while it was quite small in materials with band gaps greater than 9.36 eV [9]. In our measurement nonlinear absorption was clearly observed in fused quartz below 70 GW/cm$^2$. The value of $\beta$ was estimated to be $1.4 \times 10^{-11}$ cm/W. But no significant absorption was observed in CaF$_2$ and MgF$_2$ as shown in Fig. 2. This result contradicts previous reports on CaF$_2$ [10]. The reason for the difference is not clear. Nonlinear absorption depends on the materials. In CaF$_2$, an anomalous decrease of the transmittance was frequently observed above 70 GW/cm$^2$. But it was confirmed to coincide with beam break-up due to self-focusing. Color centers also increased absorption, especially in high intensity regions.

2. Two-Photon Absorption in CaF$_2$

To determine the order of nonlinear absorption in CaF$_2$ is important at 248 nm since the band gap of 10.0 eV is close to the two-photon energy. As mentioned in the preceding section, no significant nonlinear absorption was measured but visible luminescence was observed whenever a laser pulse was transmitted...