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HTML ABSTRACT + LINKS

Ultraviolet poling of pure fused silica by high-intensity femtosecond radiation

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We demonstrate UV poling of a pure fused silica sample by applying to it an electric field of 200 kV/cm and irradiating it with high-intensity (~40 GW/cm^2) femtosecond (220 fs) laser pulses at 264 nm. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1868075]

The possibility of inducing a second-order nonlinearity in glass by poling has some very intriguing possibilities for fiber optics in particular and photonics in general, as it has opened the way for creating different built-in devices in a standard telecom fiber; amongst them modulators, switches, and harmonic generators. The method of UV poling, first realized by Fujiwara *et al.*,[1] consists of the irradiation of highly Ge-doped (15.7 mol %) fused silica glass by nanosecond 193 nm laser pulses in the presence of an applied electric field of 100–300 kV/cm. Fujiwara *et al.* reported the induction of a nonlinear second-order coefficient as high as 3.4 pm/V. Despite his following publications on this subject,[2–4] there is growing skepticism about these results in scientific literature,[5,6] as they are not reproducible (maybe due to a special nature of the sample used by him). In this investigation, we tried to repeat Fujiwara's experiments using a high-intensity UV femtosecond laser source and undoped fused silica samples, excited by a two-photon mechanism.[7,8] After irradiation, we observed a strong second-harmonic (SH) signal in the areas of the sample that had been subjected both to the electric field and to the UV light.

The experimental setup is shown in Fig. 1. The fused silica samples used for poling were made from Herasil 1 glass (Heraeus Quarzglas GmbH) and had dimensions $37\times11\times0.5$ mm^3. All the sides of the samples were polished and aluminum electrodes, 11×5 mm^2, were deposited on both plane sides of the samples in such a way that the edges of each electrode were at a distance of about 3 mm from the end of the plate and side edges (see Fig. 1).

For sample irradiation, we used the UV pulses from a femtosecond Nd:glass laser (Twinkle, Light Conversion Ltd., Vilnius, Lithuania)[9] with the following parameters: the pulse energy, ε_p, is around 200 μJ; the wavelength, λ, is 264 nm; the pulse duration, τ, is 220 fs [full width at half maximum (FWHM)]; the beam diameter, w, is 0.3 cm (FWHM); and the repetition rate, Δf, is 27 Hz. The UV radiation was focused into the narrow side of the sample by a cylindrical lens with focal length 21.8 cm. The sample was placed at a distance of 21 cm from the lens. The laser beam size at the entrance of the sample was about 3×0.15 mm^2 (at FWHM). A high voltage block FC30P4 (Glassman High Voltage, Inc.) was used to create an electric field of about 200 kV/cm through the sample.

The peak beam intensity and the incident fluence were calculated from the following:[8]

$$I=\frac{\varepsilon_p}{\left(\frac{\pi}{\ln 2}\right)^{3/2}\frac{\pi w^2}{4}\left(\frac{F-S}{F}\right)}, \tag{1}$$

$$E=\frac{\varepsilon_p N}{\frac{\pi w^2}{2\ln 2}\left(\frac{F-S}{F}\right)}, \tag{2}$$

where ε_p is the pulse energy incident on the sample; F is the focal distance of the lens at 264 nm; S is the distance between the lens and the sample (more precisely, between the principal plane of the lens and the sample); and N is the total number of pulses incident on the sample. With the high voltage applied across the sample, the sample was irradiated with the high-intensity UV laser light for approximately 3 h (total incident fluence of around 8 kJ/cm^2) with peak incident intensity of around 200 GW/cm^2 (at the narrow edge of the sample). Using our experimental values on the two-photon absorption coefficient for Herasil glass,[7] we can estimate the light intensity value entering the volume inside the sample under high voltage as 40 GW/cm^2 (the pulse energy, entering the poling volume, is about 40 μJ). The total irradiation fluence regarding the poling volume was about 2 kJ/cm^2.

A digital oscilloscope LT372 (LeCroy) was used to measure the photoelectric current induced in the sample. Immediately, at the start of the irradiation, a strong photocurrent pulse with an amplitude of up to a 20 V appeared, which corresponded to a conductivity value of $\sigma=2\times10^{-9}$ cm^{-1} Ω^{-1} (resistivity value $\rho=5\times10^8$ cm Ω, which is about ten orders less than the standard reference value for room temperature $\rho=10^{18}$ cm Ω[10]). The calculated initial quantum yield of charged particles between the electrodes was about 2×10^{-6}, if we presume that the time resolution of our oscilloscope was 50 ns. During the irradiation, the pulse amplitude value degraded rather quickly (during some seconds) to a level of some volts (Fig. 2). A horizontal movement of the sample with respect to the laser beam led to a revival of the photocurrent signal. After switching off the applied voltage, the photocurrent pulse changed its polarity and continued to degrade. No current was observed in the absence of high-intensity UV light. The observed photocurrent behavior is qualitatively consistent with the one induced

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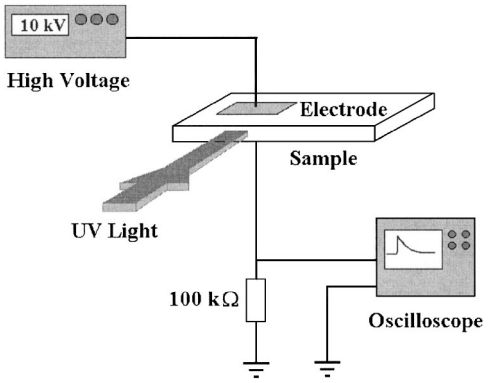


FIG. 1. Schematic of UV poling experiment with induced photocurrent measurement.

in earlier experiments on the two-step ionization $S_0 \rightarrow S_1 \rightarrow S_N$ of oxygen deficient fused silica by KrF excimer laser radiation with $\lambda = 248$ nm and $\tau = 25$ ns.[11]

After poling and removal of the electrodes (by etching), the sample was tested for evidence of second-harmonic generation. As a source of fundamental radiation, a mode-locked and Q-switched Nd:YAG laser was used ($\lambda = 1064$ nm). For the Maker's fringe measurement,[12] the sample was placed on a rotation stage and the laser beam was focused on the poled area, with the polarization parallel to the plane of incidence. The generated SH light was detected with a photomultiplier. The measurement of the periodicity of the fringes in the Maker's fringe pattern allows us to determine the thickness of the poled region in the sample. The fit to the experimental data (Fig. 3) reveals that the region with an induced $\chi^{(2)}$ nonlinearity corresponds to the whole bulk volume between the two electrodes. The comparison of the SH signal value with that obtained from the reference sample (crystalline quartz) with a known $\chi^{(2)}$ value of 0.6 pm/V,[13] allowed us to estimate the maximum value of the induced second-order nonlinearity as about 0.02 pm/V. A rectification model[14] gives for the induced second-order nonlinearity a value of

$$\chi^{(2)} = 3\chi^{(3)}E_{\mathrm{dc}}, \tag{3}$$

where $\chi^{(3)}$ is the third-order nonlinear susceptibility, equal to 4×10^{-22} m^2 V^{-2} for fused silica,[15] and E_{dc} is the applied electric field. If we take E_{dc} equal to 200 kV/cm, we will

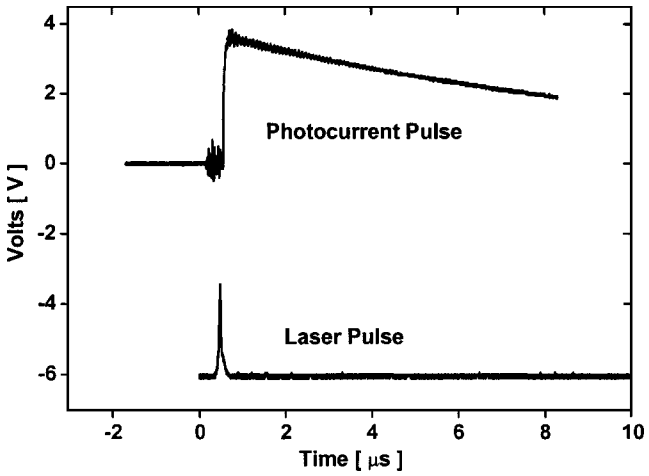


FIG. 2. Oscillogram of an induced photocurrent signal together with femtosecond UV pulse.

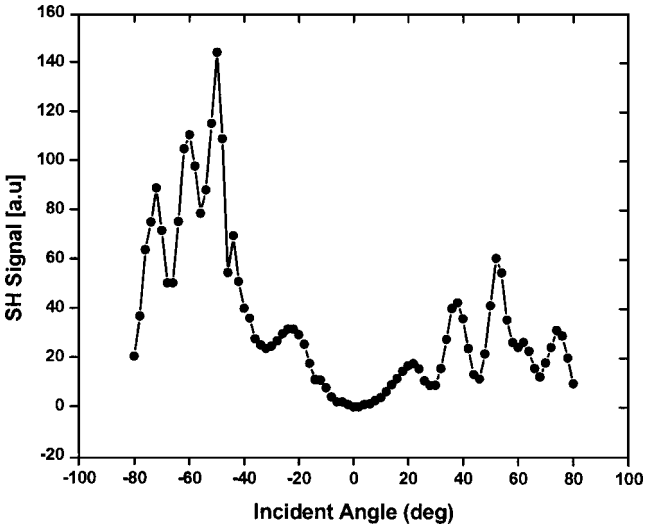


FIG. 3. Maker's fringe pattern registered in the poled sample of Herasil glass.

obtain a predicted $\chi^{(2)}$ value of 0.024 pm/V, which is in remarkable agreement with the measured one. If the same experiment is performed in twin-hole fibers,[16] where the maximum applied field would be only limited by the dielectric strength of silica at room temperature ($>3 \times 10^8$ V/m,[17] $\sim 7 \times 10^8$ V/m,[18] $>10^9$ V/m[16]), we can tentatively predict a value of $\chi^{(2)}$, reaching 1 pm/V.

We also scanned the pump beam across the surface of the sample to test the distribution of the second-order nonlinearity, using the setup described in our earlier work[19] (Fig. 4). The geometry was the same as for the Maker's fringe measurements described earlier. The angle of incidence was chosen to be close to normal incidence (10°) to reduce the effects of nonuniformities. A stronger SH signal is observed near the side where the laser beam entered the sample which could be explained by the decrease of UV intensity caused by two-photon absorption in the glass. During the inscription procedure, we poled several regions under the electrodes by

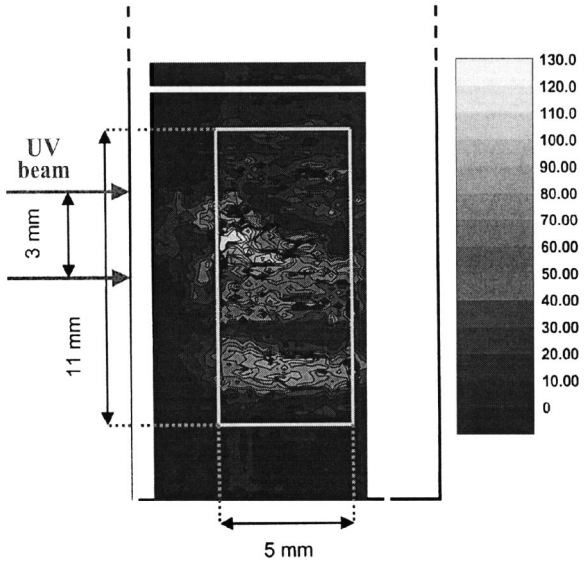


FIG. 4. The two-dimensional distribution of the second-harmonic signal in the poled fused silica sample. The sample geometry is marked by the black lines and the area covered by the electrodes is marked by the white lines. The UV beam entered the sample from the left.

translating the sample. It is clearly seen that the second-harmonic signal is observed *only* in the area under the electrodes illuminated by the UV light beam, indicating that the poling takes place only under the simultaneous action of both UV light and the applied electric field.

In conclusion, using high-intensity ($\sim$40 GW/cm^2) femtosecond UV light combined with a high electric field (200 kV/cm) we were able to induce a second-order nonlinearity of 0.02 pm/V in undoped fused silica.

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