Structural Changes in Vitreous SiO_2 Irradiated Using a Femtosecond Laser

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Introduction

Since 1997, photoinscription of waveguides in a bulk glass has become a novel very promising application of femtosecond lasers [1]. Multiphoton absorption induces a local fusion of the glass in any chosen point of the bulk sample, probably accompanied by a densification of the material after its re-solidification. This process leads to a variation of the refractive index and allows a waveguide to be realised in the bulk glass interior and/or 3D devices. Little is known about the nature of photoinduced phenomena produced by ultrashort laser pulses. We started a systematic study of glass structure variations on mesoscopic, intermediate- and short-range scales using hard x-rays and neutrons.

Methods and Materials

The laser pulses used for photoinscription were produced by a regenerative amplifier operating at a wavelength of 800 nm, with a duration of 160 fs. The peak power density was varied between 10 and 40 TW cm^{-2}. The SiO_2 glass samples were rectangular plates with a thickness of 100 μm. High-energy x-ray diffraction, small-angle neutron and x-ray scattering were used to study structural changes in the irradiated samples. Further experimental details and some preliminary results were reported elsewhere [2,3].

Results

High-energy x-ray data were taken on the Basic Energy Science Synchrotron Radiation Center Collaborative Access Team (BESSRC-CAT) 11-ID-C beamline and analysed using the ISOMER-X software package [4]. The obtained structure factors (Fig. 1) were found to be very similar except the first sharp diffraction peak (FSDP) at 1.6 $Å^{-1}$, which decreases by $\approx$2.4% for a 40 TW cm^{-2} irradiated sample (see the insert in Fig. 1). As expected, no visible changes in the short-range order were detected after Fourier transform of the data.

Changes on the mesoscopic scale were found to be much more spectacular. Both small-angle x-ray scattering (SAXS) (measured using the BESSRC 12-ID beamline) and small-angle neutron scattering (SANS) (Institute Laue-Langevin, France) show a significant increase of the scattering intensity for irradiated samples and a pronounced anisotropy (Fig. 2).

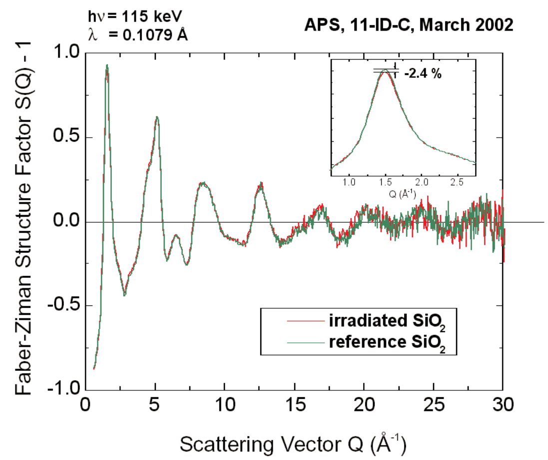

FIG. 1. The Fiber-Ziman structure factor S(Q)-1 for non-irradiated and fs laser-irradiated SiO_2 (40 TW cm^{-2}). The insert shows a decrease of the FSDP on irradiation.

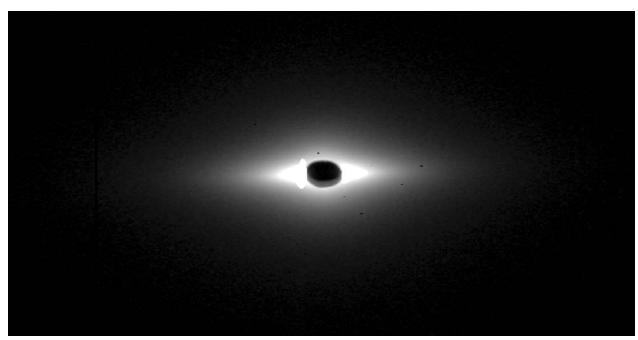

FIG. 2. Typical SAXS raw 2-D image of irradiated SiO_2 glass samples with a pronounced anisotropy. Non-irradiated SiO_2 exhibits the isotropic signal with a much lower scattering intensity.

The difference scattering intensity $\Delta I(Q)$ for fs laser-irradiated SiO_2 samples increases by 5 to 6 orders of magnitude below Q = 0.1 $Å^{-1}$ (Fig. 3) and shows a critical behaviour with a threshold peak power density of $\approx$9 TW cm^{-2} (Fig. 4).

Discussion

The available neutron and x-ray diffraction data for densified SiO_2 clearly indicate a distinct correlation

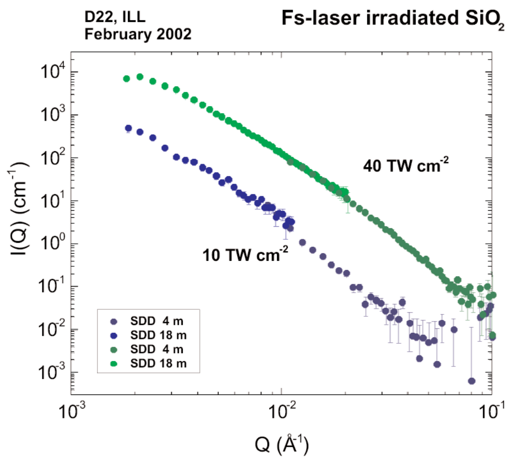

FIG. 3. The difference scattering intensity ΔI(Q) for selected SiO_2 samples. The ΔI(Q)s were obtained by subtracting the signal from non-irradiated SiO_2.

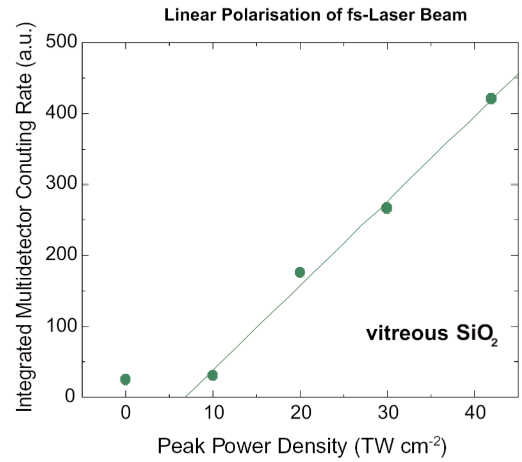

FIG. 4. Integrated multidetector counting rate for investigated SiO_2 glass samples. A nearly linear increase is observed above the threshold power density of ≈9 TW cm^{-2}.

between the FSDP amplitude and glass network compaction [5,6]. In our case, the irradiation results in a ≈2% densification consistent with a density increase estimation of 1.8% from the refractive index change. Moreover, a red shift of the ω_4 (TO) lattice mode, observed for fs laser-irradiated SiO_2 and interpreted as a decrease of the Si-O-Si bond angle by 1.5° [7], also confirms the above estimation.

A structural hypothesis for the densification is a change in the intermediate-range order, i.e., an increase in the number of three- and four-membered silica rings within the irradiated region instead of usual silica rings composed by six $SiO_{4/2}$ tetrahedra.

Small-angle scattering data show that the densification process caused by irradiation has a threshold power density consistent with a non-monotonic change in the optical properties (refractive index, birefringence). The interface between densified and nondensified regions seems to be rather well-defined, since the ΔI(Q)s follow the Porod law with a slope (∂logΔI(Q)/∂logQ) of -4. A characteristic size of the densified regions is a few hundreds Å.

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