

## OPTICAL HYSTERESIS IN FREE-STANDING POROUS SILICON FILMS

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The porous silicon (PS) is a material with a large structural inhomogeneity on nanometer scale. This would produce the unique optical properties of this material [1]. Some of them have been connected with the potential applications of PS in visible silicon optoelectronic technologies, including optical nonlinear devices. The strong optical nonlinearities revealed for PS as early as 1993 [2] indicate the possibility of application of this material not only for effective light-emitting diodes, but also for optical switches in all-optical processing and optical communication systems.

The nonlinear light transmission in PS is studied in a large temporal range, but the most of papers are dealing with millisecond and picosecond/femtosecond time scale. The observed nonlinearity relaxation time constants vary from milliseconds to several picoseconds [1]. The slow optical bistable input-output power characteristics in PS, when the relaxation time of the optically induced absorption change is about 1 ms, were demonstrated in [1]. The fast hysteresis effect was revealed in [3], where the laser pulse propagation through the PS films indicates the fast-induced absorption changes. The revealed laser pulse shape deformation, especially of the tailing edge of the pulse, indicates a pronounced hysteresis cycle. As a rule, the slow nonlinearity of the response in PS is related to the carrier localization on the surface states of the Si microcrystals [1, 3] and the fast recovery is attributed to rapid thermalization of the photoexcited carriers to the surface states [1, 4], or to bimolecular radiative recombination [2]. The thermal effects due to laser heating may also play a significant role, since PS has a very low thermal conductivity [1].

In [5] we have reported the optical power limiting effect in PS during the nonlinear propagation of strong laser pulses in free-standing films. In this letter are prepared the experimental measurements with the time resolution in order to reveal the influence of induced absorption on the laser pulse shape, thus demonstrating microsecond optical transmission hysteresis in free-standing PS films.

The PS films (thickness  $d = 16 - 25 \mu\text{m}$ ) were prepared by anodization from commercial bulk Si substrates (KES 0.01),  $0.01 \Omega\text{-cm}$ , (111) cut orientation. An electrolyte HF (49%): ethanol 1:1 was used. Anodization was carried out at room temperature in two chambers unit cell with  $1 \text{ cm}^2$  surface area at  $j = 7 \text{ mA/cm}^2$  density current about 20 min. For the separation of free-standing film the current density was switched sharply to  $j = 70 \text{ mA/cm}^2$ . Free-standing PS films were then rinsed by ethanol and dried in air stream.

The experiments were done in a single-beam transmission/reflection setup, by measuring synchronously the temporal shape of incident laser pulse ( $I_0$ ), transmitted ( $I$ ) and reflected ( $I_r$ ) ones. The measurements were carried out using a rhodamine 6G flash-lamp pumped dye laser pulses ( $h\nu = 2.09 \text{ eV}$ , pulse duration (FWHM)  $\tau \approx 0.9 \mu\text{s}$ , bandwidth  $\Delta\lambda \approx 0.1 \text{ nm}$ ) with a smooth temporal profiles. The measurement time resolution was 20 ns. The spot diameter of the focused on the sample laser beam was 0.2 - 0.3 mm. The PS samples were measured when irradiated in the spectral region of strong absorption of the PS material (where the absorption coefficient  $\alpha \geq 2 \times 10^3 \text{ cm}^{-1}$ , see inset in Fig. 1). All experiments were done at room temperature.

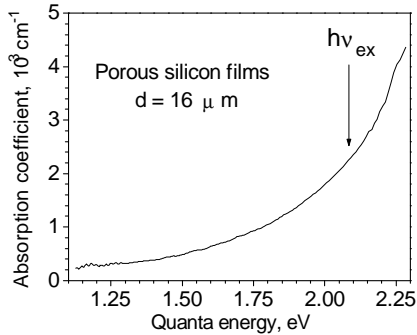


Fig. 1. Optical absorption spectra of free-standing PS films with 0.16  $\mu\text{m}$  thickness. Arrow – laser wavelength position.

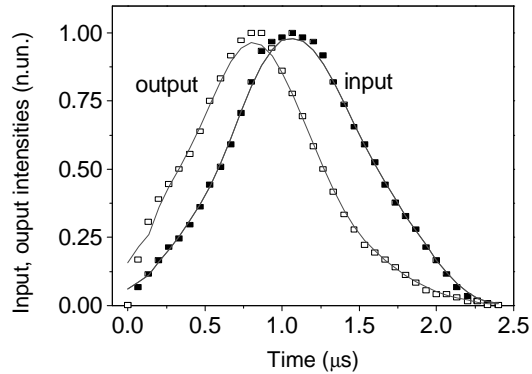


Fig. 2. Laser pulse profile deformation in PS. Input pulse intensity 275  $\text{kW}/\text{cm}^2$ .

We detected the pulse profile deformation when pulse intensity increases over some threshold value ( $I_t = 70 - 100 \text{ kW}/\text{cm}^2$ ). Fig. 2 shows the oscillograms of transmitted and incident ( $I_o$ ) laser pulses, when  $I_o > I_t$ . The change of the pulse profile increases with the increasing of the input light intensity  $I_o$  up to surface damaging of the sample (damaging pulse intensity  $I_d \approx 350 \text{ kW}/\text{cm}^2$ ). Up to  $I_d$  no changes in the reflected laser pulse profiles were detected. The change of the time profile of the laser pulses leads to the hysteresis-like dependencies of the transmitted intensity on the corresponding value of the input one (Fig. 3). This optical hysteresis cycles have a pronounced form in the clockwise direction, typically for nonlinear optical effects with the increase of absorption [6]. The form of optical hysteresis cycles shows a typical character for transient effects and does not imply an optical bistability [6]. This means that in our measurements the laser pulse duration is shorter than relaxation time of the detected nonlinear photoinduced phenomena.

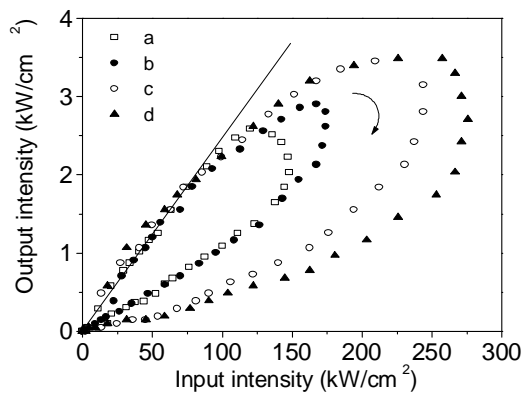


Fig. 3. Optical hysteresis in PS films. Input pulse intensity: 145 (a), 175 (b), 245 (c) and 275  $\text{kW}/\text{cm}^2$  (d).

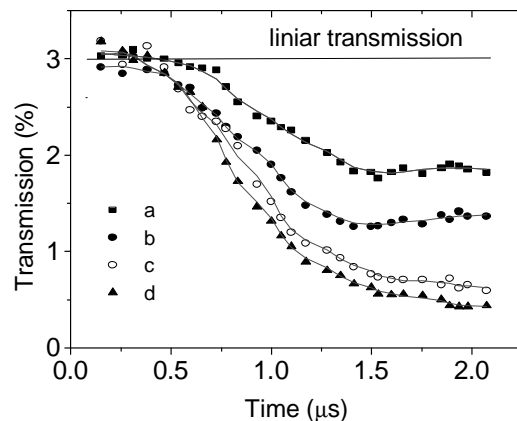


Fig. 4. Induced photodarkening in PS in the field of microsecond laser pulses. Input pulse intensity as for Fig. 3.

The kinetics of the PS films induced darkening has been determined. Fig. 4 presents the transmission changes during the PS film irradiation by the laser pulses of microsecond duration. The characteristic time for the formation of the new PS light absorption state, corresponding to the almost horizontal part in Fig. 4, in the field of laser pulses is less than the laser pulse duration. The photo-induced increase of the PS light absorption coefficient in the field of the laser pulses has a reversible character, i.e. the medium is fully restored to its initial transmission state after the ending of the laser

pulse, and if the same place on the sample is irradiated with another laser pulse the effect can be repeated.

We suggest that, as in the case of optical limiting [5], optical hysteresis arises from nonlinear photoinduced absorption of microsecond laser pulses. Analogous nonlinear photodarkening and optical hysteresis phenomena were measured in non-crystalline semiconductors, including a-Si:H films [7, 8], and were explained by the mechanism of nonlinear light absorption, which take into account the interaction with nonequilibrium phonons and localized vibrational modes. This mechanism can be, also, applied to the case of PS, since this material possesses a large specific surface and, consequently, a large concentration of structural inhomogeneities of crystalline silicon at nanometric scale, that may lead to spatial localization of the phonon vibrational modes and, as a result, to the formation of nonequilibrium state for a high level of laser excitation [7, 8].

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