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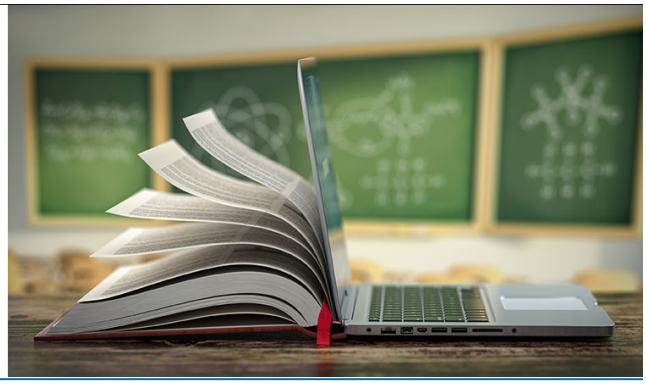

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Time resolved nonlinear response of $Sn_2P_2S_6$:Sb to nanosecond pulse excitation

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Abstract. The temporal variations of SPS:Sb transmission are measured within a 10 orders of magnitude time range (from a few nanoseconds to ten seconds) with a red diode laser beam, after illumination of the sample with a nanosecond pulse of green light. The measurements revealed a sequence of darkening and bleaching processes that are sensitive to the intensities of pre-illumination and testing beams, or the sample temperature, or both. This allowed for identifying particular nonlinear processes that manifest themselves in changes of transmission.

1. Introduction

$Sn_2P_2S_6$:Sb (SPS:Sb) is a fast and efficient photorefractive crystal sensitive in red up to 800 nm [1]. Its response can be enhanced by pre-illumination to white light [2]; its gain depends on the recording intensity. The comparison of photorefractive and EPR data allowed for identifying Sb as a secondary photorefractive center responsible for optical sensitizing [3]. In the present contribution, we study the variation of SPS:Sb transmission at 633 nm within the interval from 10^{-9} s to 10 s, after its illumination with 20 ns pulse of green light. Such measurements are done for different intensities of the exciting and testing radiation, within the temperature range from -109 to 40°C.

2. Experiment

A 2.4 mm-thick sample of SPS that contains 1% of antimony was fixed on a copper cold finger in a vacuum chamber; the temperature control was ensured with a cold nitrogen flow and a heater. The chamber had two optical windows to illuminate the sample with a short powerful pulse from Continuum Panther OPO (532 nm, 1.3 J/cm^2, 20 ns) and to monitor the sample transmission with a diode laser beam (0.633 nm, 1.5 W/cm^2). Two polarizers and two half-wave phase retarders allowed for polarization adjustments of both beams and for the control of their intensities.

Figure 1 shows a representative example of temporal variations of the diode laser red beam transmitted through SPS:Sb sample at ambient temperature during and after exposure to the green pulse, plotted in a log time scale. Both, laser beams are linearly polarized along the sample *x*-axis. An entire temporal range was measured in several attempts with different sampling times and different total times of measurement, to ensure a sufficient temporal resolution in every particular time interval.

During the pulse duration (≈ 20 ns) the sample transmission is gradually decreasing, pointing to photoexcitation of free carriers that might further form polarons [4].

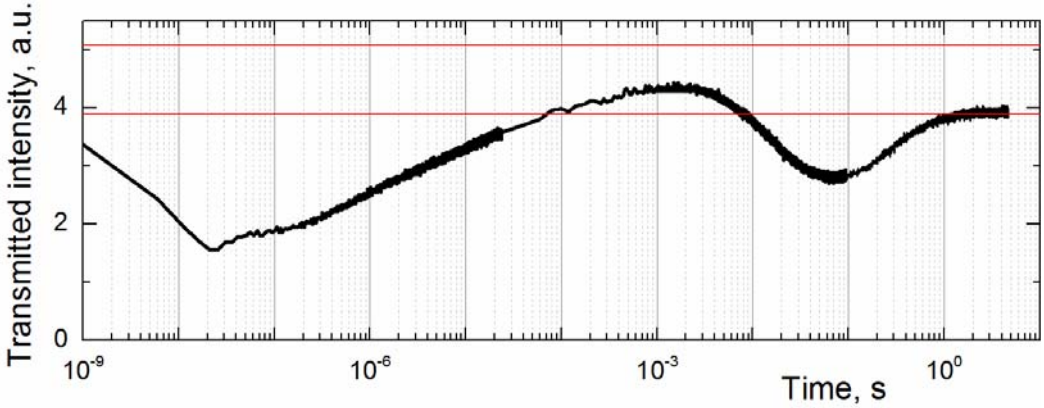

Figure 1. Temporal dynamics of SPS:Sb sample transmission at 633 nm and ambient temperature during and after 20 ns green pulse illumination (black curve). Two red lines mark the initial level of red beam transmission (upper line) and transmission level of partially exhausted for photorefractive scattering red beam in the virgin sample (with no green pulse exposure).

These photoinduced entities decay within the time range from 0.1 ms to 1 ms or even longer. The exact time is shadowed by a new extinction process that starts from a few milliseconds and is related to the development of photorefractive scattering known as beam fanning. Finally, we observe the second bleaching process from ≈ 50 ms to several seconds, with a contribution of the formation of the compensation gratings by moving charges of opposite sign [5].

The next experiments aimed to extract the characteristic build-up and decay times for three processes that occur after termination of the exciting green pulse. The sets of measurements were conducted for every process at different red light intensities and at different sample temperatures. This allowed for identification of their physical origin and for estimating the activation energies from the Arrhenius plots. The comparison of the obtained data with the activation energies extracted from EPR measurements [3,4] allowed for the conclusion that the initial light-induced absorption is caused by hole polarons (Sn^{2+} with a trapped hole [4]), while it is the antimony dopant which is responsible, indirectly and directly, for beam fanning and the development of compensation gratings [5].

The question may arise, why in the present experiment we don't see 14 s decay time as observed in [2] with 633 nm pre-exposure. This might be due to two reasons. In a few seconds the cw red beam may generate a comparable Sb^{2+} density as that done by the green-pulse pre-exposure so that the Sb^{2+} density is close to its steady-state value. The second reason might be related to the hyperbolic decay character of the $Sb^{2+} \rightarrow Sb^{3+}$ relaxation revealed in [6] at low temperatures.

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