

L. VOSTRIKOWA, AND M.K. BALAKIREV

Russian Laser Center, Russian Academy of Sciences, Novosibirsk, Birjuisk 5/65, Russia

Anomalous Growth of the Absorption of Light and Second Harmonic Generation by Optical Poling of Oxide Glass

Introduction

An illumination of glasses by two mutually coherent sources with different frequencies $E_1(\omega)$ and $E_2(2\omega)$ leads to the formation of a long-lived reversible static polarization [1–7]. This process is called as optical poling [1]. By optical poling a glass loses the symmetry and acquires the properties of single-axis crystal. The appearance of the asymmetry in glass leads to the removal of the ban on the three-wave interactions. Therefore, the second harmonic generation (SHG) [1, 3–7] and parametric amplification [8] of light become possible in this medium.

The optical poling is associated [3] with the formation of a spatially periodic electrostatic field $E_0(\mathbf{r})$ in glass. Such field appears as a result of a charge separation by means of direct current due to the coherent photogalvanic effect (CPhGE) [9,10]:

$$\mathbf{J} = \mathbf{j}(E_1^2 E_2^* + \text{c.c.}) \quad (1)$$

A basic peculiarity of optical poling is the spatial periodicity of photoinduced field with the period of q^{-1} ($q = 2\mathbf{k}_1 - \mathbf{k}_2$, \mathbf{k}_1 and \mathbf{k}_2 are the wave vectors of basic and doubled frequencies). Therefore, the periodic modulation

of the refractive index $\Delta n \sim \chi^{(3)} E_0^2$ with the period of $(2q)^{-1}$ (anisotropic Δn -grating [2]) and the second-order effective polarizability $\chi^{(2)} \sim \chi^{(3)} E_0$ with the period of q^{-1} ($\chi^{(2)}$ -grating [3]) appear by optical poling of glass. By certain angles of the convergent beams the formation of the grating leads to the self-diffraction of incident light [2]. The spatial periodicity of $\chi^{(2)}$ promotes the effective SHG [3-7] and parametric amplification of light [8] since the process of the conversion of light in this gratings is phase-matched.

In the present letter the big reversible growth ($\geq 10^3$ times) of the absorption of second harmonic radiation by optical poling of the oxide glass was detected. This absorption is the cause of the restriction of the maximum value of photoinduced SHG efficiency. The influence of the absorption on the process of optical poling and SHG was investigated and the models of the appearance of such anomalous absorption are discussed.

Experimental results and discussion

The experiments were performed in the prefabricated measuring plate PM-40 (oxide glass Soviet trade mark K-8). From several samples we choose the sample with the maximum efficiency of optical poling. The transmission of our sample is displayed in Fig.1, the refractive indices are $n_1 = 1.506$ and $n_2 = 1.518$ for $\lambda_1 = 1.079 \mu\text{m}$ and $\lambda_2 = 0.54 \mu\text{m}$, correspondingly. Photoinduced SHG in the glass of this sort of trade mark was first observed in the work [11].

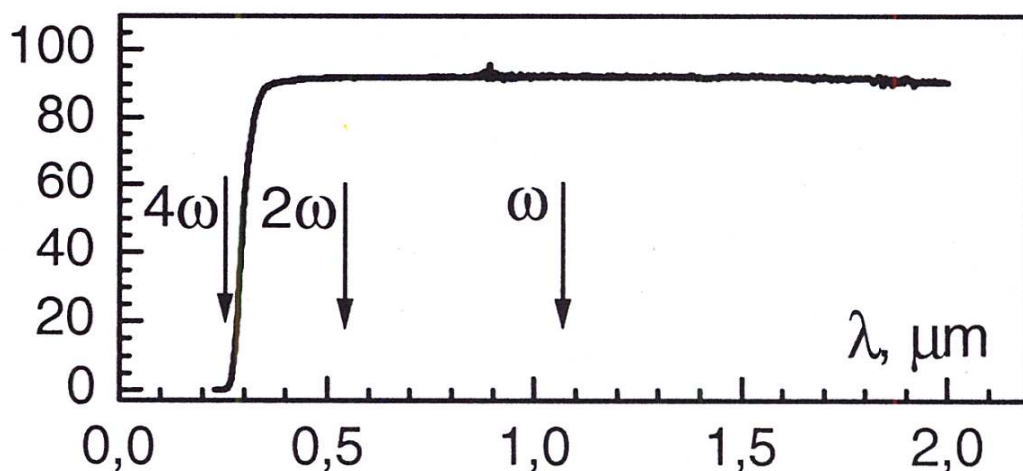


Fig. 1. Transmission of the sample (length is 4 cm)

The scheme of the experimental setup is shown in Fig. 2.

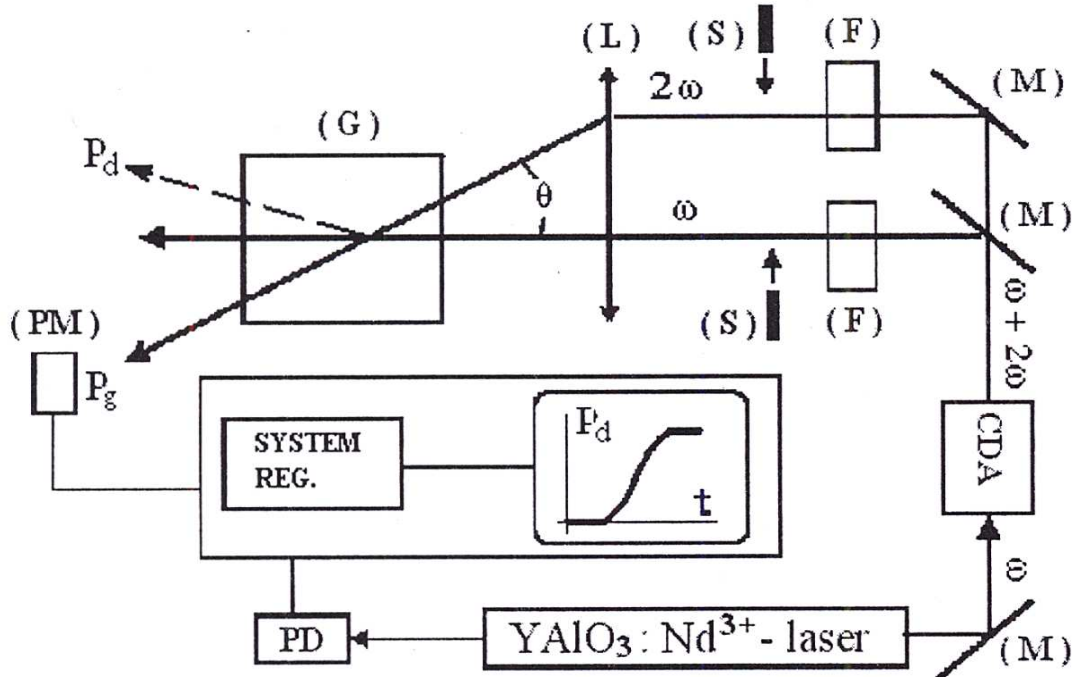


Fig. 2. Experimental setup: (M) — mirrors, (F) — filters, (S) — opaque shades, (PM) — photomultiplier, (L) — spherical lens, (G) — sample (glass), CDA — frequency doubler, PD — photodiode

The experiment was consisted of the following. The focused beams of pulsed $\text{YAlO}_3:\text{Nd}^{3+}$ — laser ($\lambda_1 = 1.079\mu\text{m}$) and its doubled harmonic were intersected in the sample at the small angle $\sim 3.6^\circ$. The beams were polarized in the plane of their convergence, the duration of light laser pulses was 15ns, the repetition frequency was 12,5Hz. The diameter of the beams at the focal point was $\sim 260\mu\text{m}$, the intensity at the waist being $P_\omega \sim 10^9 \text{ W/cm}^2$ and $P_{2\omega} \sim 10^8 \text{ W/cm}^2$ for the basic and doubled harmonic, correspondingly. The angle between the axes of the beams was chosen so that the Bragg diffraction condition on the photoinduced refractive index grating would be satisfied for the beam at the first harmonic

$$(\cos \theta \approx \frac{n_1}{n_2}) \quad (2)$$

During this both frequencies illumination (glass poling) the electrostatic field grating \mathbf{E}_0 and, correspondingly, Δn and $\chi^{(2)}$ -gratings were produced in glass. In experiments we registered the peak power $P_d(\omega)$ of the self-diffracted on the Δn — grating light of the basic frequency, the peak power $P_{2\omega}$ of the

passed through the sample radiation of doubled harmonic and the peak power $P_g(2\omega)$ of the photoinduced on the $\chi^{(2)}$ — grating second harmonic. In the process of the registration of SHG the incident light of the doubled harmonic was overlapped at the entrance to the sample. The signals were detected by a photomultiplier in the remote zone. After the photomultiplier the signal was transformed by the analog-digital voltage converter and was processed on the computer. In doing so, the recorded signal was averaged over 15–20 pulses which decreased the influence of radiation instability. The sensitivity threshold of the measuring system was $1\mu\text{W/pulse}$.

The measurement results are shown in Figs. 3–6. In Fig. 3 (left part) the typical dependences on time for the P_d , $P_{2\omega}$ and the intensity $I_{2\omega}$ on the axis (the split $\sim 40\mu\text{m}$ was in the center of the registered beam) of the passed through the grating light with doubled frequency are shown in the process of the optical poling all the way to saturation ($t \approx 70\text{min}$).

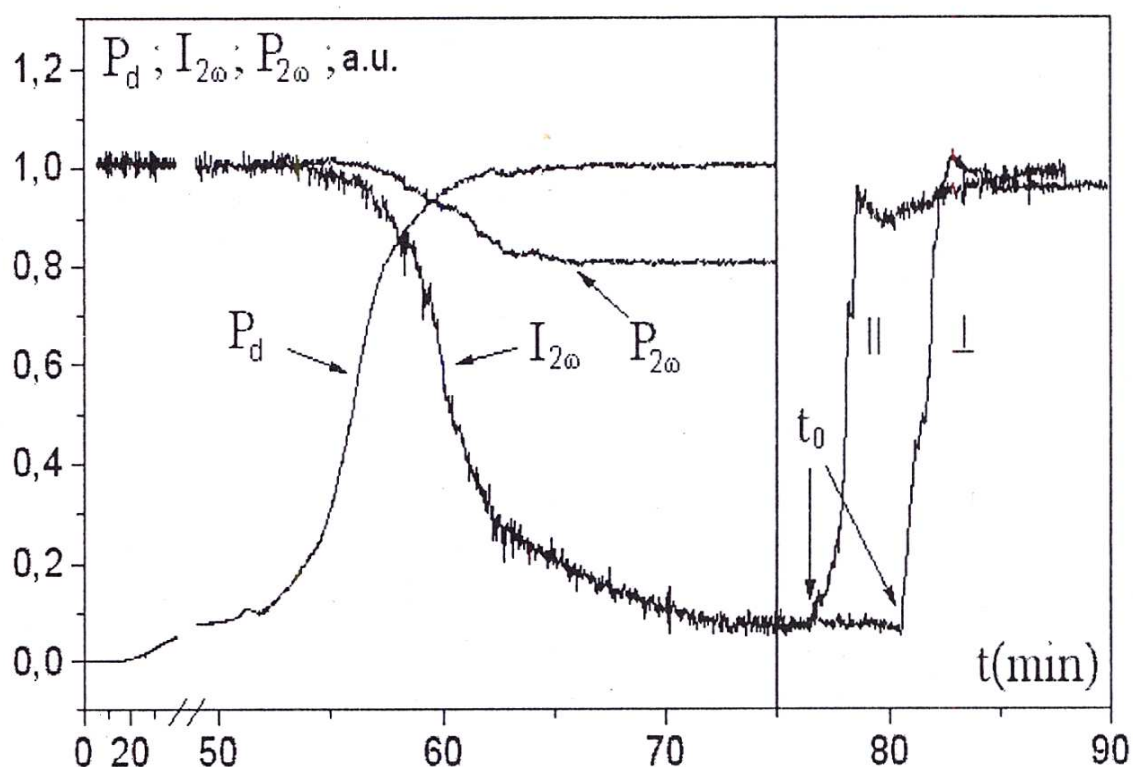


Fig. 3. Dependence of the P_d , $I_{2\omega}$, $P_{2\omega}$ on time by optical poling of glass (left part), the evolution of the intensity of the passed through the grating doubled frequency radiation with parallel and perpendicular polarizations (right part, in the moment t_0 the basic beam was overlapped)

In Fig. 4 the beam distribution $I_{2\omega}(d)$ (d is the distance from the center of the beam) of the passed through the grating light with doubled frequency in the steady-state region is shown. The beam distribution was received by moving the split (with diameter $\sim 40\mu\text{m}$) in the plane of the convergence of incident beams. The split was placed at $\sim 2\text{cm}$ distance from the back side of the glass.

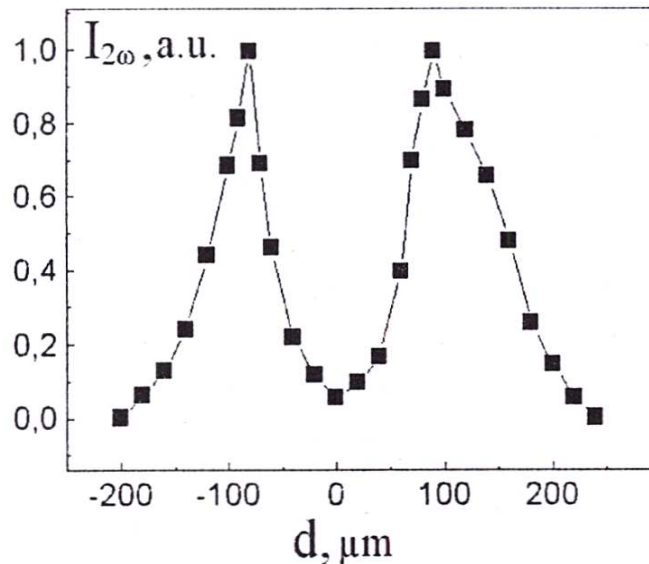


Fig. 4. Distribution of the beam of passed radiation with doubled frequency

The experiments show that at the some value of the amplitude of grating the sharp grows of the absorption of the light with doubled frequency is appeared. We controled the amplitude of the grating by the peak power $P_d(\omega)$ of the self-diffracted light of the basic frequency. The maximum absorption takes place on the axis of doubled frequency beam. The doubled frequency radiation, as it passes through the grating, can be attenuated on the beam axis by a factor of 30. If it is assumed, for a rough estimate, that the decay over the length of the grating is exponential, then the observed attenuation corresponds to absorption $\sim 7\text{cm}^{-1}$. The charge in the absorption compared with an unperturbed medium is three orders of magnitude. Since the transverse size of the field grating is somewhat smaller than the beam diameter, the obtained distribution (Fig. 4) approximately reflects the dependence of the attenuation on the transverse distribution of the amplitude of the grating. The integral absorption of the light was not so big and added up to 20%. We note that some increase in green light absorption has been observed in optical fiber with prolonged passage of mutually coherent bichromatic light with frequencies ω and 2ω through it [12].

We made some special experiments to check the polarization dependence of the absorption. The experiment was consisted of the following. After the optical poling of the glass up to saturation ($t \geq 70\text{min}$) the basic beam was overlapped and we observed the evolution of the intensity of the passed through the grating light with doubled frequency. The results for two different polarisations of the light with doubled frequency are shown in Fig. 3 (right part). In the first case the polarization of light coincided with the polarization of the writing beam. In the second case the polarization was perpendicular. One can see from Fig. 3 (right part) that the absorption is maximum in the initial moment and sharply increases with time and then reaches the value as it was in the undisturbed sample. This behaviour is a result of the erase of grating by the light with doubled frequency. In the process of the erase of grating the absorption of light decreases. We connect the erase with the existence of the photoconductivity in the sample which creates the screen for photoinduced electrostatic field grating. One can see that the photoinduced absorption is polarization-independent, i.e. the dychroism is absent in the grating. The experiment shows also that under our conditions the observed effect maybe considered as a new physical phenomenon.

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