

# Optical frequency doubling in bulk glass patterns

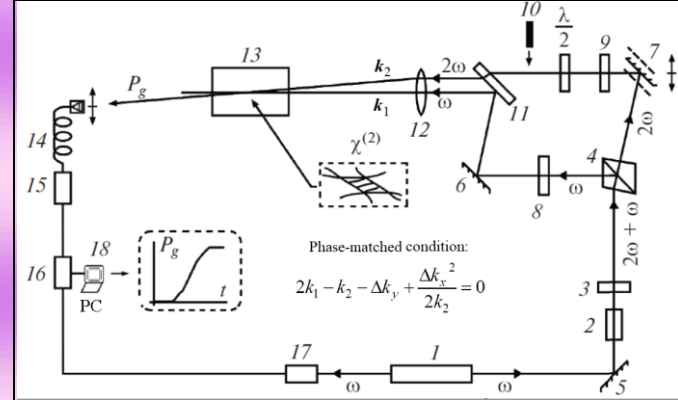
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Compare of optical frequency doubling in different bulk glass patterns are presented. Typical set up is shown in Fig.1. During illumination with use the powerful inter-coherent two-frequency radiation  $\omega+2\omega$  (as a result of coherent photovoltaic current or local reversible static polarization) inside isotropic mediums the spatial-periodic electrostatic field  $E$  arises and the corresponding photo-integrated micro-structure of second-order susceptibility  $\chi^{(2)}$  accumulates in region of  $\omega+2\omega$  interaction:

$$E \sim \frac{1}{\sigma} e_E E_1 E_2 \cos[(2k_1 - k_2)r], \quad \chi_{ijk}^{(2)}(2\Omega; \Omega, \Omega) = \chi_{ijk}^{(3)}(E_i \delta_{jk} + 2E_k \delta_{ij}), \quad \chi^{(3)} = 2\pi\chi_{iii}^{(3)}(\Omega, \Omega, 0).$$

The nonlinear three-wave conversions of light waves can took place in investigated isotropic samples on accumulating second-order susceptibility  $\chi^{(2)}$ . And here the arising optical nonlinear frequency doubling process is studied.



To observe nonlinear process of optical frequency doubling, when writing micro-structures of second-order susceptibility  $\chi^{(2)}$  by laser radiations  $\omega$  and  $2\omega$  up to saturation in different glass patterns, incident radiation  $2\omega$  was periodically shuttered for several seconds at entrance to sample, and frequency doubling radiation  $P_g$  appears on induced  $\chi^{(2)}$  as nonlinear transformation of basic frequency radiation. Peak power  $P_g$  is registered on PC in real time. Efficiency is  $\eta_g = P_g/P_{\omega}$ .

**Fig. 1.** Scheme of the experimental setup: (1) YAG:Nd<sup>3+</sup> laser, (2) converter to the second harmonic based on a KTP crystal, (3) phase-shifting plate, (4) Glan prism with oblique faces, (5–7) mirrors, (8, 9) filters for the radiations of the fundamental and doubled frequencies, (10) shutter, (11) polarization element, (12) lens, (13) sample, (14) light guide, (15) photomultiplier, (16) strobe voltage converter, (17) photodiode, and (18) computer.

Table 1 contains summary results of investigations of nonlinear optical frequency doubling in second-order susceptibilities  $\chi^{(2)}$  photo-integrated in different bulk glass materials. The lengths of interaction regions  $L$  in Table 1 have values less than 1 cm since strong focusing of interacting beams in volume of samples was applied. The performed experiments show that there are sufficiently different efficiencies (up to some orders of values, see Table 1) for generation of nonlinear frequency doubling in photo-induced micro-structures of second-order susceptibility  $\chi^{(2)}$  in glass materials containing different chemical elements. On base of presented experimental results can be performed comparative analysis of influence of chemical elements. So, some samples with sufficiently big values up to  $10^{-4}$  have been obtained in glass media with content the concentrations of lead-oxide and rare-earth elements. Writing times for  $\chi^{(2)}$  micro-structures up to maximum saturation in different studied samples are some minutes. But lifetimes of photo-integrated  $\chi^{(2)}$  micro-structures in all investigated volumetric glasses, see Table 1, are sufficiently small and this is basic task which must be solved in following researches. The photo-integrated  $\chi^{(2)}$  micro-structures of second-order susceptibility can be used in future for creation of broad-band sources of nonlinear transformations for micro- and may be for nano-optoelectronics but additional investigations must be performed for obtaining as more high efficiencies as long lifetimes. Thanks to groups of Nikolaev Institute of Inorganic Chemistry and Vavilov State Optical Institute for some synthesized samples. Work was made as Russian State Task FWGW-2025-0019.

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**Table 1.** Results of optical frequency doubling in various bulk glass patterns.

Elements, mol. %	Laser data	$I_{\omega}, \text{GW/cm}^2$ $I_{2\omega}, \text{GW/cm}^2$	$d, \mu\text{m}$ $L, \text{cm}$	$t_w, \text{min}$	$t_d; t_{\omega};$ $t_{2\omega}, \text{min}$	$\eta_g$
SiO <sub>2</sub> 20,5%PbO16%Na <sub>2</sub> O+6%TiO <sub>2</sub> 1,7%CeO <sub>2</sub>	ML, QS*	7 0,05	50 0,05	10		$10^{-4}$
76%Pb(PO <sub>3</sub> ) <sub>2</sub> 5,5%CeO <sub>2</sub> ; {38%PbO;4%P <sub>2</sub> O <sub>5</sub> }	PS	35 0,05	10 0,01	30		$10^{-4}$
Pb(PO <sub>3</sub> ) <sub>2</sub> Ba(PO <sub>3</sub> ) <sub>2</sub> +1,5%CeO <sub>2</sub> ; {16%PbO}	PS	35 0,05	10 0,01	40	$10^4$ 60 -	$10^{-5}$
BS7 {45%PbO}	ML, QS	17 0,07	10 0,006	4	15 15 5	$10^{-5}$
BS7 {45%PbO}	ML, QS	30 2	10 0,006	5	120 15 -	$10^{-6}$
BS7 {66%SiO <sub>2</sub> 19%PbO}	ML, QS*	7 0,05	50 0,05	10	$2 \cdot 10^4$ - -	$10^{-6}$
SiO <sub>2</sub> 20,5%PbO16%Na <sub>2</sub> O	ML, QS*	7 0,05	50 0,05	10		$10^{-6}$
Pb(PO <sub>3</sub> ) <sub>2</sub> -R(PO <sub>3</sub> ) <sub>2</sub> ; R=Ca, Sr, Ba; {5÷40%PbO}	PS	25 0,04	10 0,01	60	100 60 -	$10^{-6}$
SiO <sub>2</sub> 20,5%PbO16%Na <sub>2</sub> O+(7÷22%)TiO <sub>2</sub>	ML, QS*	7 0,05	50 0,05	10		$5 \cdot 10^{-6}$
JS4 {36%PbO}	ML, QS	50 1,2	5 0,002	10		$2 \cdot 10^{-6}$
JS4 {66%SiO <sub>2</sub> 16%PbO}	ML, QS*	7 0,05	50 0,05	10	$2 \cdot 10^4$ - -	$10^{-6}$
PS5 {55%PbO}	ML, QS	17 0,07	10 0,006	4,5		$10^{-6}$
PS5 {69%SiO <sub>2</sub> 26%PbO}	ML, QS*	7 0,05	50 0,05	10		$3 \cdot 10^{-8}$
SK5 {39%SiO <sub>2</sub> 40%BaO+15%B <sub>2</sub> O <sub>3</sub> 5%Al <sub>2</sub> O <sub>3</sub> }	ML, QS	13 3	100 0,1	10	$3 \cdot 10^3$ - -	$10^{-7}$
F8 {50,2%SiO <sub>2</sub> 39,7%PbO+3,8%Na <sub>2</sub> O5,6%K <sub>2</sub> O}	ML, QS	13 $10^{-3}$	100 0,1	10	60 - -	$5 \cdot 10^{-8}$
BS4	ML, QS	17 0,07	10 0,006	5		$8 \cdot 10^{-8}$
K8 {plate PM15}	ML, QS	17 $10^{-6}$	10 0,006	10	120 120 -	$10^{-8}$
K8 {74%SiO <sub>2</sub> }	ML, QS*	7 0,05	50 0,05	10		$10^{-9}$
K8 {plate PM40}	PS*	4 0,4	260 0,7	40		$10^{-8}$
BS8 {55,7%PbO}	ML, QS*	7 0,05	50 0,05	10		$10^{-8}$
BS8 {63%SiO <sub>2</sub> 26%PbO}	ML, QS*	7 0,05	50 0,05	10		$3 \cdot 10^{-8}$
JZS19 {62%SiO <sub>2</sub> 35%PbO}	ML, QS*	7 0,05	50 0,05	10		$4 \cdot 10^{-8}$
HS24, S24 {crystallite}	ML, QS*	7 0,05	50 0,05	10		$10^{-8}$
BS3	ML, QS	17 0,07	10 0,006	5		$5 \cdot 10^{-9}$
JS18	ML, QS	1 $2 \cdot 10^{-3}$	20 0,01	3	- 0,05 -	$5 \cdot 10^{-9}$
JS18	ML, QS	50 1,2	5 0,001	3		$2 \cdot 10^{-9}$
BS12	ML, QS	17 0,07	10 0,006	5		$10^{-9}$
GeO <sub>2</sub> (10÷40%)PbO	ML, QS*	7 0,05	50 0,05	10		$10^{-8}$
GeO <sub>2</sub> (10÷50%)PbO	ML, QS	30 0,06	4,5 0,001	10		$10^{-11}$
SiO <sub>2</sub> 40%PbO	ML, QS*	7 0,05	50 0,05	10		$10^{-8}$
SiO <sub>2</sub> 3%GeO <sub>2</sub> +0,5%P	ML, QS	50 0,8	90 0,1	30		$2 \cdot 10^{-7}$

$I_{\omega}, I_{2\omega}$  - intensities of incident radiations  $\omega$  and  $2\omega$ ;  $d, L$  - diameter and length of all-optically induced  $\chi^{(2)}$ ;  $t_w$  -  $\chi^{(2)}$  writing time,  $t_d$  - time of  $\chi^{(2)}$  dark relaxation,  $t_{\omega}$  - relaxation time in presence of radiation  $\omega$ ,  $t_{2\omega}$  - relaxation time in presence of radiation  $2\omega$ ;  $\eta_g$  - efficiency of process of fully optical frequency doubling on  $\chi^{(2)}$  (photoinduced second harmonic generation); PS - pulsed YAG: Nd-laser,  $\tau=30\div50\text{ns}$ ,  $f=10\text{Hz}$  ( $\tau$  - pulse length,  $f$  - repetition frequency); ML, QS - pulsed YAG: Nd mode-locked and q-switched laser,  $\tau=100\div600\text{ps}$ ,  $f=76\div125\text{MHz}$ ,  $t=200\div300\text{ns}$ ,  $F=1\div6\text{kHz}$  ( $t$  - duration of bending around set of impulses,  $F$  - repetition frequency of set of impulses); ML, QS\* -  $\tau=30\text{ps}$ ,  $f=76\text{MHz}$ ,  $t=60\text{ns}$ ,  $F=12,5\text{Hz}$ ; PS\* -  $\tau=10\text{ns}$ ,  $f=12,5\text{Hz}$ ; BS3, BS4, BS7, BS8, BS12, JS4, JC18, JZS19, PS5 - plates from set of optical filters (GOST-9411-81); F8 - flint, SK5, K8 - crowns.