Subm. to QEL'S' 683

Influence of PbO concentration on photoinduced second harmonic generation in lead glasses

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Generation of permanent second-order susceptibility in lead-silicate glass fibers was reported in [1]. High-efficiency photoinduced second-harmonic generation (SHG) in lead-silicate bulk glasses was observed in [2,3].

We report here measurements of photoinduced SHG efficiency for lead-silicate glasses. Dependencies of photoinduced second-harmonic generation efficiency on PbO concentration and third-order nonlinearity are observed and interpreted on the base of photovoltaic model.

The experimental setup was described in [3]. Schott and Soviet standard lead glass samples were studied. Samples were prepared with 720 mW of pump (1.064 μm) and 1 mW of seeding (0.532 μm). Preparation time was 1.5 min and saturation was not completely achieved in samples with low PbO concentrations. The reading pump power was 500 mW. PbO concentrations of the samples were measured by scanning microscope JSM-840. It was found that Soviet standard glasses contained several percents of rare-earth and transition dopants.

We observed some deviation of measured values of the photoinduced second-harmonic (SH) signal $P_{2\omega}$ for Soviet standard glasses from those for Schott glasses with the same PbO concentration which was probably caused by dopants. Using data from [4] we calculated $\chi^{(3)}$ values for all the investigated samples. It was found that the dependence of $P_{2\omega}^{1/2}$ on $\chi^{(3)}$ is approximately linear at lower values of $\chi^{(3)}$ in accordance with photovoltaic model [5]:

$$P_{2\omega}^{1/2} \propto \chi^{(2)} = \chi^{(3)} \frac{\beta}{\sigma} I_\omega I_{2\omega}^{1/2}, \tag{1}$$

where $\chi^{(2)}$ is an amplitude of photoinduced second-order susceptibility grating, β is a component of coherent photovoltaic effect tensor, σ is photoconductivity, I_ω and $I_{2\omega}$ are intensities of pump and SH seeding. The decrease of SH signal for glasses with higher values of $\chi^{(3)}$ can probably be caused by phasemismatching because of the difference between writing and reading pump intensities. It can be also explained by an increase of third-harmonic (TH) intensity $I_{3\omega} \propto \chi^{(3)} I_\omega^3$ from third-harmonic generation that evidently leads to an increase of conductivity $\sigma_{3\omega} \propto I_{3\omega}$ due to photoionization by TH. We do observe a strong decrease of relaxation time $\tau = \epsilon\epsilon_0/\sigma$ of photoinduced SH signal in these glasses. Considering the following expression for photoconductivity

$$\sigma = \sigma_\omega + \sigma_{2\omega} + \sigma_{3\omega}, \qquad (2)$$

where $\sigma_\omega \propto I_\omega^2$ and $\sigma_{2\omega} \propto I_{2\omega}$ are conductivities due to photoionization by pump and SH, one can see from (1) that there probably exists an optimal pump intensity, which is lower than the intensity used in our experiments.

References

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[3] E.M.Dianov, P.G.Kazansky, D.S.Starodubov, and D.Yu.Stepanov, *Sov. Lightwave Commun.*, 1992, 2(1), 83–88.

[4] Catalogue of color optical glasses, Ed. by G.T.Petrovskii, Moscow, 1990; Catalogue of Schott glass.

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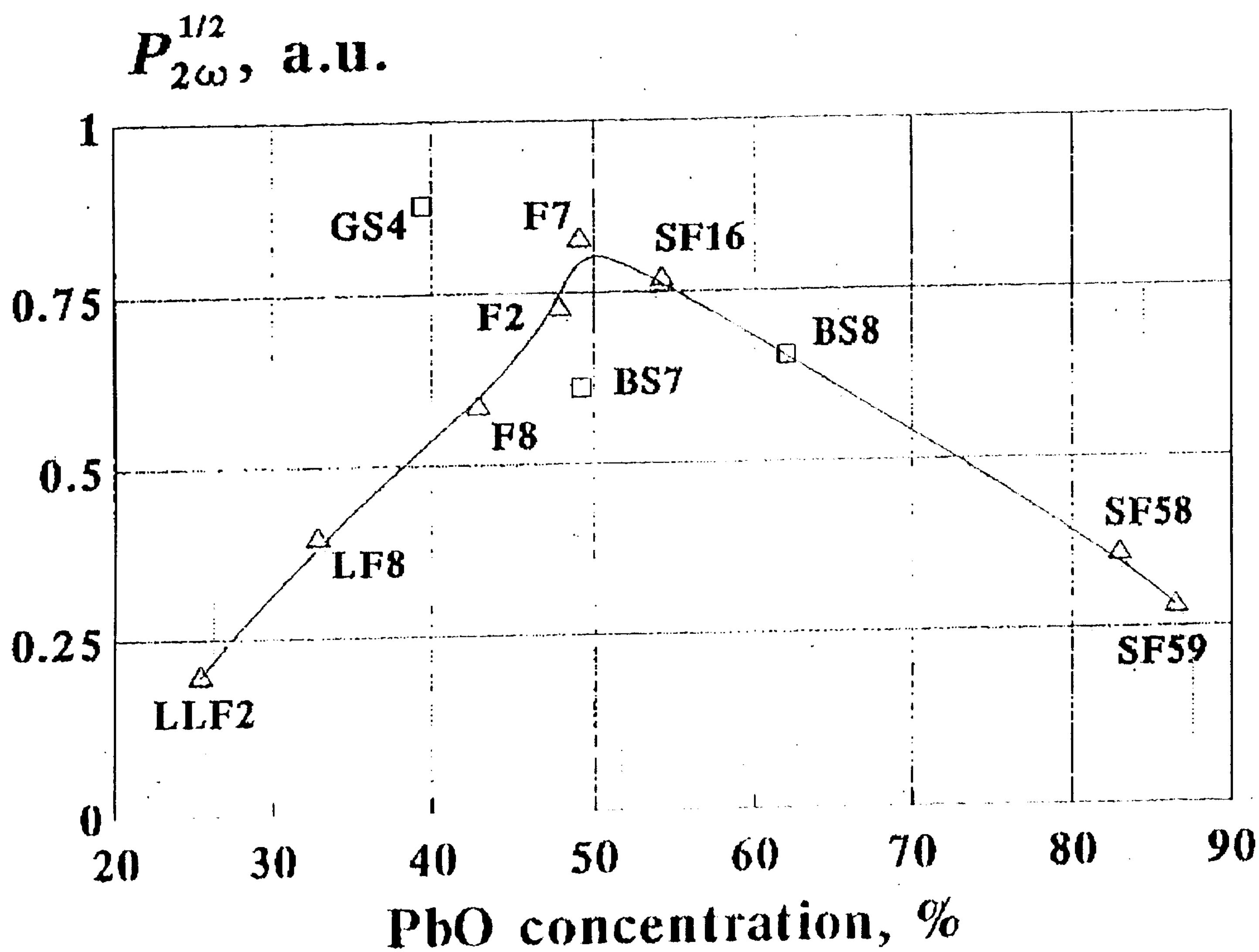
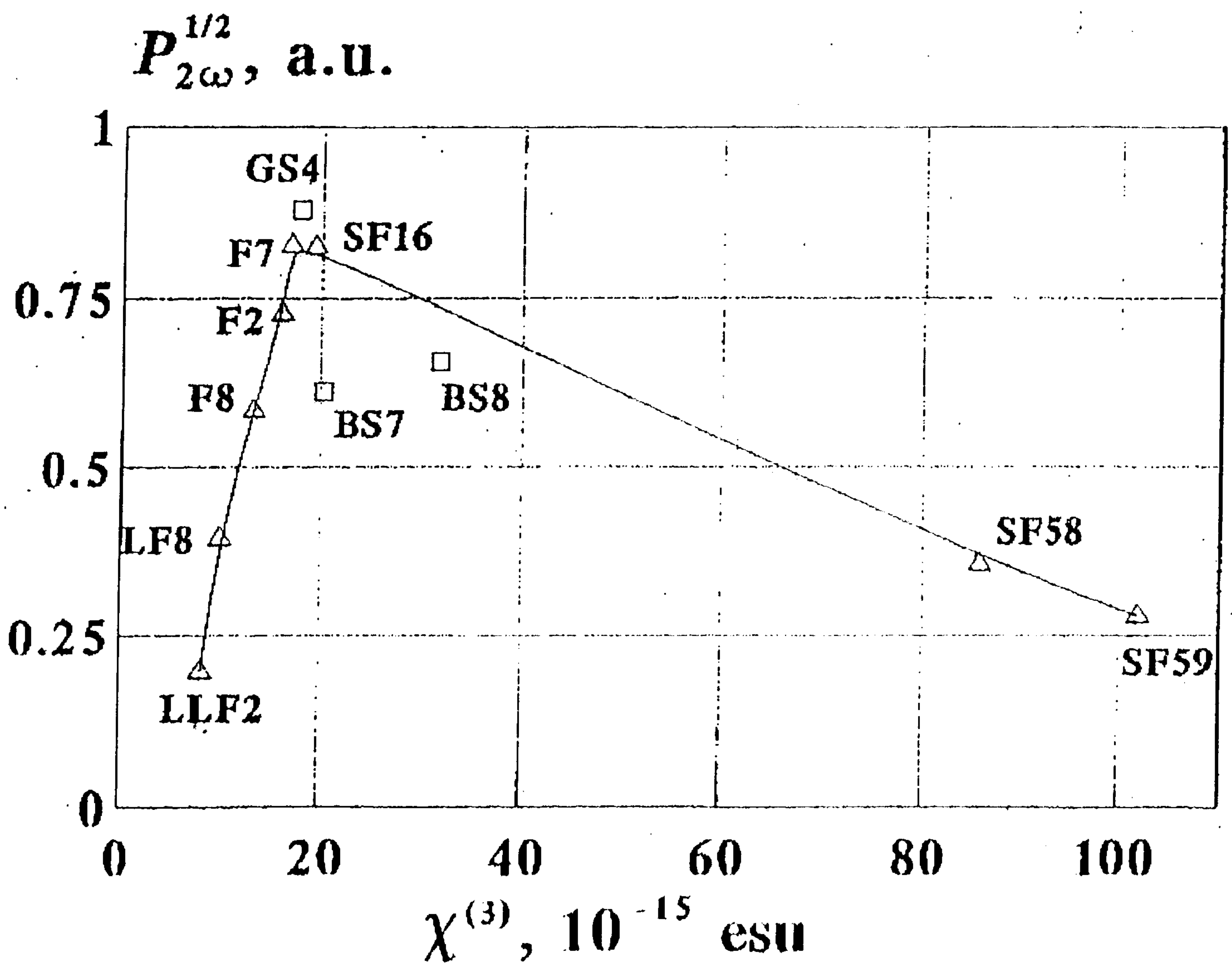
Captions to figures

Figure 1. Square-root of photoinduced SH signal $P_{2\omega}$ versus PbO concentration for Schott (△) and Soviet standard (□) glasses.

Figure 2. $P_{2\omega}^{1/2}$ dependence on $\chi^{(3)}$ for Schott (△) and Soviet standard (□) glasses.



$P_{2\omega}^{1/2}$, a.u.
1
0.75
0.5
0.25
0
20
30
40
50
60
70
80
90
PbO concentration, %
GS4
F7
SF16
F2
BS8
BS7
F8
LF8
LLF2
SF58
SF59



$P^{1/2}_{2\omega}$, a.u.
1
0.75
0.5
0.25
0
GS4
F7
SF16
F2
F8
BS7
BS8
LF8
LLF2
SF58
SF59
0
20
40
60
80
100
$\chi^{(3)}$, 10^{-15} esu