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IR enhancement of photoinduced second harmonic generation

E. M. Dianov, D. S. Starodubov

General Physics Institute, Russian Academy of Sciences,
38 Vavilov Street, Moscow 117942, Russia

P. G. Kazansky

Optoelectronics Research Centre, University of Southampton,
Southampton S017 1BJ, U. K.

Abstract

The increase of saturation efficiency and preparation rate of photoinduced second harmonic generation by counter-propagating two-photon IR pumping of short-lived photovoltaic centers for the third-order photovoltaic effect has been experimentally demonstrated.

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The explanation of photoinduced second harmonic generation (SHG) by charge separation is connected with two types of photovoltaic models : the first one considers third-order coherent photovoltaic effect,[1] the second one is based on higher-order coherent processes.[2]

Quadratic-in-time approximation of early-time growth rate of photoinduced second harmonic signal $I_{\text{ph}}(2\omega)$ makes it possible to determine the order of the whole process:

$$I_{\text{ph}}(2\omega) = \Gamma t^2 + \text{offset}, \tag{1}$$

where Γ depends on mechanism of photoinduced SHG and IR reading power. For third-order coherent photovoltaic effect Γ is given by:

$$\Gamma \propto A^2 I_{\text{r}}^2(\omega) \propto \beta^2 I_{\text{pr}}^2(\omega) I_{\text{pr}}(2\omega)\, I_{\text{r}}^2(\omega) \tag{2}$$

where $I_{\text{pr}}(\omega)$ and $I_{\text{pr}}(2\omega)$ - IR and SH preparation intensities correspondingly, and $I_{\text{r}}(\omega)$ - IR reading intensity, β is a component of coherent photovoltaic effect tensor.

It was shown, that in fiber preform [3] both the fundamental and SH power laws are faster than predicted by third-order process and seems to agree reasonably well with higher-order process.

However for elucidation of the order of coherent photovoltaic effect it's necessary to reveal the possible contribution of intermediate exited states, because the population of these states can be interpreted as intensity dependence of β in (2) and can increase the power law of entire process.

Pre-exitation experiments during the preparation of $\chi^{(2)}$-gratings in glasses have shown, that either the ω or 2ω beams prior to encoding had no effect on $\chi^{(2)}$ values [4].

We report the experimental technique, which uses counter-propagating fundamental beam for both incoherent pumping of intermediate states and reading of the second-order susceptibility grating, produced by preparation beam. Q-switched (repetition rate $\sim 6.3\,\text{kHz}$) and mode-locked (pulse duration $\sim 600\,\text{ps}$) Nd:YAG laser with KTP frequency doubling was used. We studied Ce- and Ti-doped lead glass.

In Fig.1 the dependence of photoinduced SH saturation level on IR reading pulse delay is shown. Coincidence of the pulses led to essential increase of photoinduced SH saturation level. The strong decrease of preparation time was also observed. The symmetry of obtained picture means, that the lifetime of IR-induced photovoltaic centers is small as compared with pulse duration.

For clarifying the difference in preparation process for coincided and non-coincided pulses we have measured SH growth rate dependencies on IR reading and preparation powers for these two cases. The dependence of growth rate on IR reading power is shown in Fig.2. For non-coincided pulses the growth rate is in good agreement with quadratic function of reading intensity: 1.9 ± 0.4 (the reading pulse do not influence on $\chi^{(2)}$-grating formation). For coincided pulses we have obtained 5.8 ± 0.6 power law, which gives two-photon pumping of intermediate state. One can see the increase in SH growth rate by two orders of magnitude for coincided pulses. Moreover, the increase of IR reading power resulted in decrease of threshold IR preparation power (up to peak IR power $> 60\,\text{W}$).

Figure 3 illustrates SH growth rate dependencies on IR preparation power. The power law approximation gives the value 1.8 ± 0.5 for coincided pulses and the value 5.2 ± 0.8 for non-coincided pulses. These curves show, that counter-propagating IR light decreases the power law of IR reading power by two-photon process. The power of coherent process seems to agree well the three-photon model of coherent photovoltaic effect.

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Fig.1 The dependence of second harmonic saturation level on time delay of IR reading pulse. IR reading pulse duration was ~600 ps.

Fig.2 IR reading power dependencies of SH growth rate for coincided (△) and noncoincided (□) pulses. The 6-power law and 1.9-power law approximations are shown for 0 ps delay and 800 ps delay of reading pulse.

Fig.3 IR preparation power dependencies of SH growth rate for coincided (△) and noncoincided (□) pulses. The 5.4-power law and 1.7-power law approximations are shown for 0 ps delay and 800 ps delay of reading pulse.

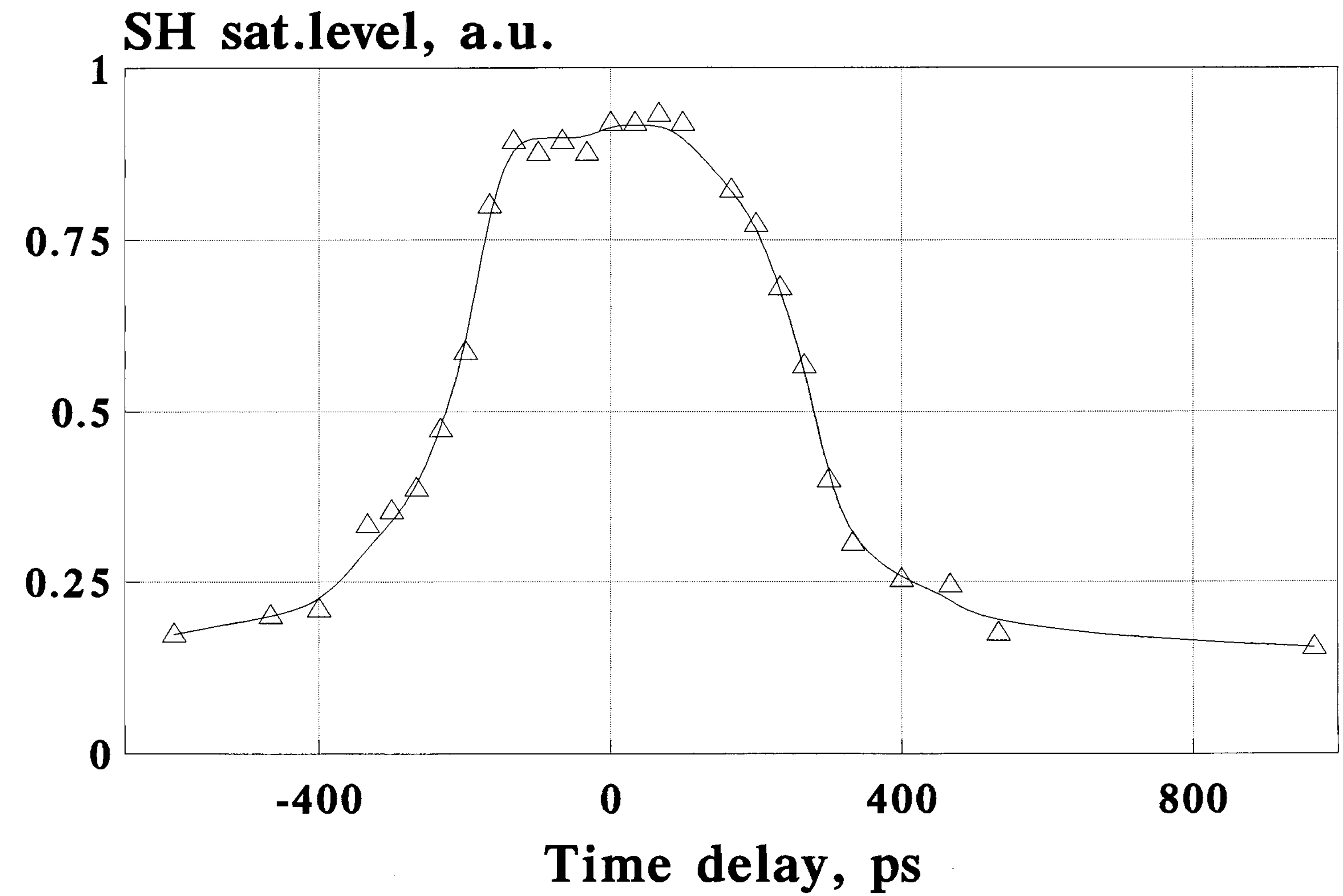
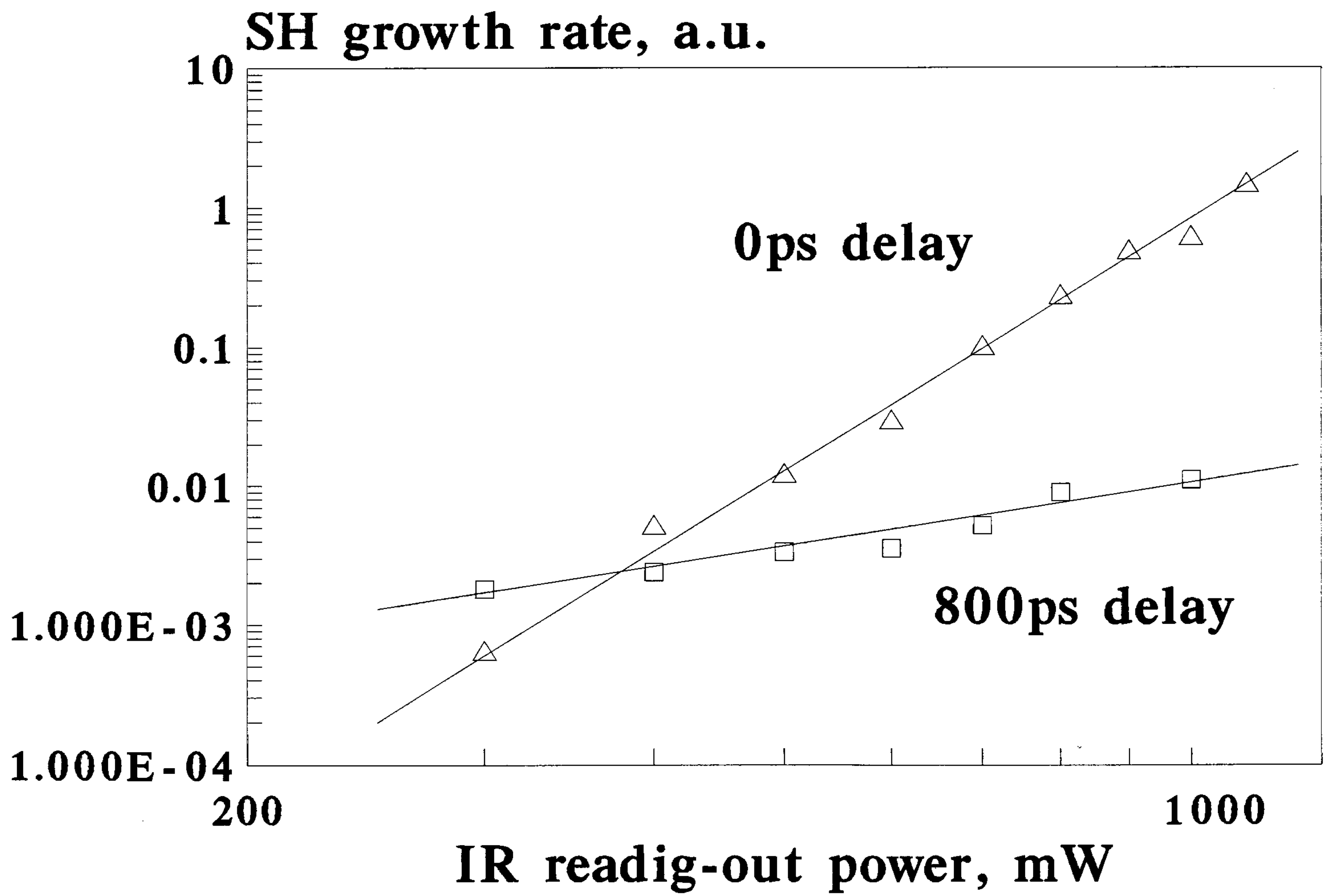
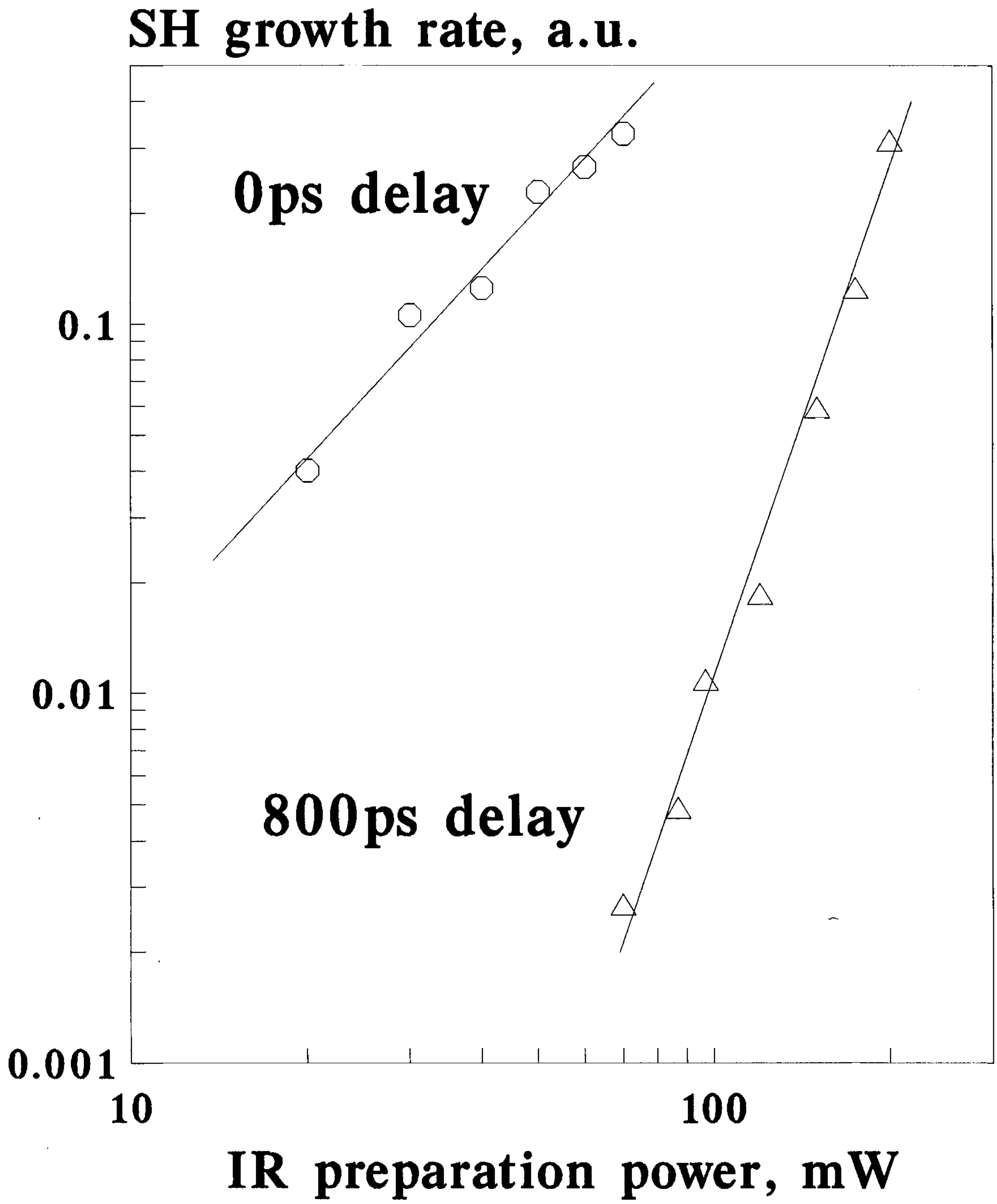


Fig. 1



SH growth rate, a.u.
10
1
0.1
0.01
1.000E-03
1.000E-04
200
1000
0ps delay
800ps delay
IR readig-out power, mW

Fig. 2



SH growth rate, a.u.
0ps delay
800ps delay
0.1
0.01
0.001
10
100
IR preparation power, mW

Fig. 3