

PACS numbers: 42.65.Ky; 42.65.Re; 42.70.Mp
DOI: 10.1070/QE2005v035n06ABEH006618

Highly efficient generation of second and third harmonics of a femtosecond Cr : forsterite laser in nonlinear optical crystals

V.M. Gordienko, S.S. Grechin, A.A. Ivanov, A.A. Podshivalov

***Abstract*. Highly efficient generation of the second ($\eta_{2\omega}$= 69 %) and third ($\eta_{3\omega}$= 26 %) harmonics of a femtosecond Cr : forsterite laser radiation is achieved. It is shown that for the pump intensity exceeding 50 GW cm^{-2} ($\eta_{2\omega}$= 60 %), the third-order nonlinearity begins to play a significant role, resulting in a deterioration of the spectral parameters of the generated harmonics.**

Keywords: *harmonic generation, femtosecond radiation.*

In this paper, we describe the results of investigation of highly efficient generation of second and third harmonics of a repetitively pulsed femtosecond Cr : forsterite laser [Cr^{3+} : Mg_2SiO_4 (Cr : F)] emitting 1-mJ, 110-fs pulses at 1.24 μm. The second and third harmonic generation efficiencies achieved in experiments were 69 % and 26 %, respectively. The study was performed for a wide range of intensities, including the saturation regime caused by the third-order nonlinearity.

A high conversion efficiency of Cr : F laser radiation into the second and third harmonics is important because it allows a significant enhancement of the contrast of femtosecond pulses. The second harmonic generation (SHG) of femtosecond Cr : F laser radiation can be realised under conditions close to those of group velocity matching of interacting waves [1 – 3] which makes it possible to obtain a high efficiency of nonlinear optical conversion. As the pump intensity is increased to achieve the maximum conversion efficiency, the third-order nonlinearity effects $\chi^{(3)}$ become significant [4]. These effects cause, first, a phase mismatch between the generated waves, thereby reducing the conversion efficiency and, second, they distort the spectrum of the generated radiation. SHG of femtosecond Cr : F laser radiation ($\tau \sim 200$ fs) with the maximum conversion efficiency $\eta_{2\omega} \sim 50$ % was achieved in [5] using LBO crystals under conditions when the effect of the third-order nonlinearity was insignificant.

V.M. Gordienko, S.S. Grechin, , A.A. Podshivalov International Laser Center, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119992 Moscow, Russia; e-mail: gord@femtosrv.phys.msu.ru, grechin@psi.phys.msu.su;
A.A. Ivanov Photochemistry Center, Russian Academy of Sciences, ul. Novatorov 7a, 117421 Moscow, Russia

Received 7 April 2005
Kvantovaya Elektronika **35** (6) 525 – 526 (2005)
Translated by Ram Wadhwa

An optimal crystal for generation of harmonics of a femtosecond Cr : F laser radiation can be selected by analysing the dispersion and nonlinear properties of crystals [3], supplemented by the data on cubic nonlinearity $\chi^{(3)} \sim n_2$. Table 1 presents the effective nonlinearity coefficients $d_{\rm eff}$, group lengths $L_{\rm gr}$ (τ = 100 fs), and normalised characteristic coefficients K for the most efficient crystals for each type of interaction [3], which were used to select the optimal crystal. From the data available in the literature on the nonlinearity coefficient n_2, we chose the values at wavelengths closest to the wavelengths of the Cr : F laser radiation and its harmonics.

Table 1. Parameters of the most efficient crystals for generation of second and third harmonics of a Cr : F laser radiation.

Radiation harmonics	Crystal	$d_{\rm eff}^2/n^3$/ pm^2 V^{-2}	$L_{\rm gr}$/ mm	K	$n_2/10^{-16}$ cm^2 W^{-1}
second	LBO	0.23	10.6	1	2 (616 nm) [6]
	CDA	0.04	9.3	0.13	–
	DCDA	0.04	6.7	0.07	–
third	BBO	0.41	2.3	8.3×10^{-2}	5.2 (400 nm) [7]
	LBO	0.24	1.0	9.3×10^{-3}	2 (616 nm)
	KDP	0.05	1.9	6.8×10^{-3}	2.8 (1064 nm) [8]
	KDP	0.05	1.9	6.8×10^{-3}	8.4 (400 nm) [9]

In the SHG experiments we used a 4-mm long LBO crystal ($\varphi = 0$, $\theta = 87°$) with an AR coating on both faces. Figure 1 shows the experimental dependence of the second harmonic conversion efficiency on the pump intensity. The maximum conversion efficiency of 69 % was achieved for a pump intensity of 180 GW cm^{-2}. However, the second harmonic radiation spectrum was broadened displaying the influence of the third-order nonlinearity effect when the conversion efficiency exceeded 60 % (see inset in Fig. 1). Figure 1 also shows the theoretical dependences of the conversion efficiency on the pump intensity, calculated by using the spectral model [10] with and without consideration of the self-phase and cross-modulation. The following values of coefficients were used in the calculations: $d_{\rm eff} = 0.72$ pm V^{-1}, n_2 (1240 nm) = 10^{-16} cm^2 W^{-1}, and n_2 (620 nm) = 2×10^{-16} cm^2 W^{-1} [6].

The obtained data indicate that for pump intensities exceeding 50 GW cm^{-2}, the effects caused by the third-order nonlinearity become significant and the spectral parameters of the generated radiation begin to deteriorate. Thus, $\eta_{2\omega} = 60$ % is the maximum SHG efficiency for the Cr : F laser with preserved parameters of the generated

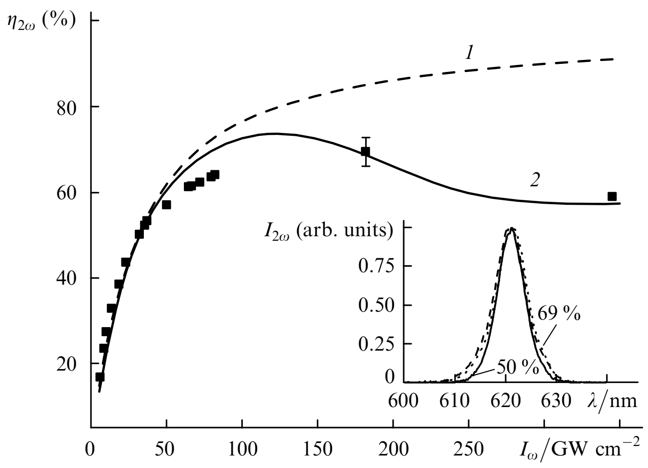

Figure 1. Dependence of SHG efficiency $\eta_{2\omega}$ on the pump intensity I_ω at the fundamental frequency, obtained (*1*) without and (*2*) with consideration of the self-phase and cross-modulation effects. The inset shows the spectra of the second harmonic radiation pulse for various conversion efficiencies.

radiation. The duration of the second-harmonic radiation pulse measured by the cross-correlation technique is equal to 80 ± 5 fs, which corresponds to the calculated pulse duration of 85 fs. This also confirms the fact that second-harmonic conversion takes place in the regime close to group velocity matching.

For the third harmonic generation (THG), we used 1-mm long KDP ($\varphi = 45°$, $\theta = 45°$) and LBO ($\varphi = 0$, $\theta = 8°$) crystals without AR coatings. Figure 2 shows the dependences of the THG efficiency on the fundamental radiation intensity. The ratio of intensities of the fundamental radiation and second-harmonic radiation incident on the crystal varied from 1 : 1 to 1 : 2.

Theoretical calculations were made by using the tabulated data for the KDP crystal ($d_{36} = 0.42$ pm V^{-1} [11] and $n_2 = 8.4 \times 10^{-16}$ cm^2 W^{-1} [9]) and the data for the LBO crystal obtained from the analysis of the SHG experiments.

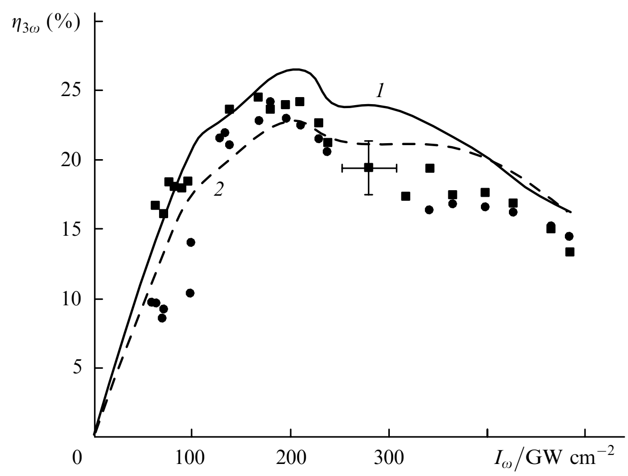

Figure 2. Dependence of the THG efficiency $\eta_{3\omega}$ on the pump intensity I_ω at the fundamental frequency upon radiation conversion in LBO [squares, curve (*1*)] and KDP [circles, curve (*2*)] crystals. Circles and squares are the experimental data; the curves are calculated using the spectral model.

The theoretical results are found to be in good agreement with the experimental data.

The maximum conversion efficiency for LBO and KDP crystals was found to be 26 % and 24 % respectively [taking into account the Fresnel reflection (~ 4 %) of radiation from each crystal face]. In this case, the spectrum of the generated radiation broadened, when the conversion efficiency in the LBO crystal exceeded 20 %. Thus, the THG efficiency $\eta_{3\omega} = 20$ % is the maximum for the LBO crystal under conditions of preserved parameters of the generated radiation.

Acknowledgements. This work was partly supported by the Russian Foundation for Basic Research (Grant Nos 03-02-16973a and 05-02-16476).

References

1. Lukashev A.A., Magnitskii S.A., Pryalkin V.I. *Izv. Ross. Akad. Nauk, Ser. Fiz.*, **59** (12), 123 (1995).
2. Gordienko V.M., Ivanov A.A., Podshivalov A.A., Pryalkin V.I. Kvantovaya Elektron., **31**, 391 (2001) [Quantum Electron., **31**, 391 (2001)].
3. Grechin S.S., Pryalkin V.I. *Kvantovaya Elektron.*, **33**, 737 (2003) [*Quantum Electron.*, **33**, 737 (2003)].
4. Razumikhina T.B, Telegin L.S., Kholodnykh A.I., Chirkin A.S. *Kvantovaya Elektron.*, **11**, 2026 (1984) [*Sov. J. Quantum Electron.*, **14**, 1358 (1984)]; Akhmanov S.A., Dmitriev V.G. *Vestnik Mosk. Univer., Ser. Fiz. Astr.*, (3), 85 (1963).
5. Shcheslavskiy V., Petrov V., Noack F., Zhavoronkov N. *Appl. Phys. B*, **69**, 167 (1999).
6. Bayanov I.M., Gordienko V.M., Djidjoev M.S., Dyakov V.A., Magnitskii S.A., Pryalkin V.I., Tarasevitch A.P. *Proc. SPIE Int. Soc. Opt. Eng.*, **1800**, 2 (1991).
7. Rodriguez G., Taylor A.J. *Opt. Lett.*, **23**, 858 (1998).
8. Kurnit N.A., Shimada T., Sorem M.S., Taylor A.J., Rodriguez G., Clement T.S., Fearn H., James D.F., Milonni P.W. *Proc. SPIE Int. Soc. Opt. Eng.*, **3047**, 387 (1997).
9. Aoyama M., Harimoto T., Ma J., Akahane Y., Yamakawa K. *Opt. Express*, **9** (11), 579 (2001).
10. Grechin S.S. *Kvantovaya Elektron.*, **35**, 257 (2005) [*Quantum Electron.*, **35**, 257 (2005)].
11. Dmitriev V.G., Gurzadyan G.G., Nikogosyan D.N. *Handbook of Nonlinear Optical Crystals* (Berlin: Springer-Verlag, 1999).