Third-harmonic generation of femtosecond laser radiation in β-BaB_2O_4 crystals

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Abstract

We describe the results of experimental investigation of the third-harmonic generation (THG) in β-BaB_2O_4 crystal pumped with a femtosecond laser radiation and cut at the angle θ_S permitting a process of direct phase-matched generation of *oooe* type. A theory of direct THG in uniaxial crystals, which assumes that the effective nonlinear coefficient has an additive nature with respect to the contributing processes, has been used when interpreting the experimental data. The angular dependence of the third harmonic intensity and its dependence on the incident pump power have been analysed in order to ascertain the process, the contribution of which is dominant. The analysis of our experimental results obtained under the condition of pumping β-BaB_2O_4 ($\theta_S = 53°$) sample with femtosecond pulses ($\lambda = 800$ nm, $\tau = 70$ fs, and $P_{max} = 100$ GW/cm^2) has proven that the contribution of second-order cascaded processes to the THG is prevailing.

Keywords: third-harmonic generation, effective coefficient of nonlinear conversion, femtosecond lasers

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1. Introduction

Transformation of laser radiation using nonlinear optical materials allows dramatic widening of the frequency range of generated coherent radiation. In its turn, UV laser radiation is often used for investigating different properties of materials and has a wide application range in spectroscopy, medicine and many other areas.

Femtosecond fields of high densities can give rise to higher-order nonlinear optical effects. β-BaB_2O_4 abbreviated hereafter as BBO is a highly nonlinear crystal [1], which can be efficiently used for observation of parametric generation, stimulated Raman scattering, generation of higher harmonics, sum-frequency generation, etc.

As shown in [2], a considerable contribution to the process of phase-matched third-harmonic generation (THG) using nonlinearity of the third order in uniaxial crystals is originates from the cascaded THG. Radiation of the second harmonic, generated as a result of 'asynchronous' (not phase-matched) process involving the second-order nonlinearity, interacts with the pump radiation, leading to sum-frequency generation of the third

harmonic. The results for the THG effect in the β-BaB_2O_4 crystal cut at the phase matching angle for the case of direct generation with the third-order nonlinearity have been reported by different scientific groups [2, 3]. They differ in the magnitudes of the cascaded and direct contributions to the THG. According to the data presented in the work [2], utilisation of femtosecond laser pulses ($\lambda = 1053$ nm, $\tau_p = 350$ fs, and $P_{\omega\ \max} = 250$ GW/cm^2) for pumping of a 3-mm long BBO crystal sample stipulates a dominant contribution of the second-order processes. The authors [3] have investigated the THG process using samples with the lengths of 1.0, 1.5 and 2.0 mm and pumping with 100-fs laser pulses (the pump pulse density of 250 GW/cm^2).

The results [3] suggest that the contribution of the direct phase-matched THG process is dominating. In other words, the THG effect in the BBO crystals still cannot be regarded as thoroughly studied.

The present work is dedicated to detailed experimental studies of the process of direct THG in the BBO crystal cut at an angle θ appropriate for the THG of the type I, under the condition of pumping with a powerful radiation of a femtosecond Ti:Sa laser. The analysis of possible nonlinear optical processes, which could give rise to the THG, is carried out. In order to reveal the process, of which contribution is dominating, angular dependence of the THG power and dependence of the THG efficiency on the pumping radiation power are investigated.

2. Theoretical description of the THG process in BBO

It is known from the theory that three types of phase matching are possible for the THG in optically uniaxial crystals:

$$\text{type I: } ooo \rightarrow e,\ \vec{k}_o^1 + \vec{k}_o^1 + \vec{k}_o^1 = \vec{k}_e^3,$$

$$\text{type II: } ooe \rightarrow e,\ \vec{k}_o^1 + \vec{k}_o^1 + \vec{k}_e^1 = \vec{k}_e^3,$$

$$\text{type III: } oee \rightarrow e,\ \vec{k}_o^1 + \vec{k}_e^1 + \vec{k}_e^1 = \vec{k}_e^3,$$

In the present work we have investigated the THG as a result of both the direct THG process under the phase matching condition of the type I and the sum-frequency generation process.

As shown in [4], the dependence of the THG intensity generated in nonlinear crystals, upon the incident laser power may be calculated using the formula

$$P_{3\omega} = \left(\frac{12L^2\omega^2}{\varepsilon_0^2 c^4 A^2}\right)\frac{1}{n_\omega^o n_{3\omega}^e} C_{eff}^2 P_\omega^3 \mathrm{sinc}^2\left(\frac{\Delta kL}{2}\right), \qquad (1)$$

where L=3 mm is the sample length, ω =3.91×10^{-1} s^{-1} the pump radiation frequency, A=3.14 mm^2 the bea m cross section area, n_ω^o=1.66 the refractive index of the ordinary wave at the frequency of the first harmonic, $n_{3\omega}^e$=1.61 the refractive index of the extraordinary wave at the frequency of the third harmonic, C_{eff} the effective nonlinear

interaction coefficient, $P_\omega = 45\,\mathrm{GW/cm^2}$ the pump radiation power density, c the free space velocity, ε_0 the dielectric constant, and Δk the phase mismatch of the first and third harmonic waves.

The authors of the work [3] have observed anomalously high efficiency of transformation into the third harmonic radiation in the BBO crystal, which has been claimed to be associated with a considerable contribution to the effect of the second-order nonlinearity. According to the assumption [5], the overall effective coefficient of nonlinear conversion into the third harmonic may be calculated as a sum of effective coefficients describing the processes participating in the THG:

$$C_{eff} = C_{eff}^{(3)} + K\sum_{i=1}^{n} k_i C_{i,eff}^{(2)}\,, \tag{2}$$

where $C_{eff}^{(3)} = \chi^{(3)}/4$ means the effective coefficient of the third-order nonlinear interaction, $C_{i,eff}^{(2)}$ the ith effective coefficient of the nonlinear interaction governed by the second-order nonlinearity, which refers to the both second harmonic generation and the sum-frequency generation process, K the inverted value of sum-weighting coefficient of the third-order processes, and k_i the coefficient which describes contribution of the ith effect to the THG process.

Basing on the results [2], one has to put $k_i = 0.5$ and $K = 1$ for the phase matching of the type I. For this type of phase matching and the crystal point symmetry group 3m, in which the BBO crystallises, Eq. (2) takes the following form [2]:

$$\begin{aligned} C_{eff} = \frac{\omega d_{22}}{c}\Bigg[& d_{22}\frac{\sin 6\varphi}{2}\left(\frac{\cos\theta}{n_{2\omega}^{o}\Delta k_{SFG}^{ooe}} - \frac{\cos^3\theta}{n_{2\omega}^{e}\Delta k_{SFG}^{oee}}\right) \\ & + d_{15}\cos 3\varphi\left(\frac{\cos^2\theta\sin\theta}{n_{2\omega}^{e}\Delta k_{SFG}^{oee}} - \frac{\sin\theta}{n_{2\omega}^{o}\Delta k_{SFG}^{ooe}}\right)\Bigg] + C_{10}\cos 3\varphi\sin\theta, \end{aligned} \tag{3}$$

where d_{22} and d_{15} denote nonzero components of the tensor of nonlinear susceptibility coefficients of the second order, θ and φ respectively the polar and azimuthal angles, $n_{2\omega}^{o}$ and $n_{2\omega}^{e}$ respectively the ordinary and extraordinary refractive indices of the BBO crystal, C_{10} an effective nonzero coefficient of the third-order nonlinearities, and Δk_{SFG}^{ooe} and Δk_{SFG}^{oee} the phase mismatches:

$$\Delta k_{SFG}^{ooe} = k_{3\omega}^{e} - k_{2\omega}^{o} - k_{\omega}^{o}\,,$$

Here $k_{3\omega}^{e}$, $k_{2\omega}^{e}$ are the wave vectors of the third- and second-harmonic extraordinary waves, respectively, and $k_{2\omega}^{o}$, k_{ω}^{o} the wave vectors of the second- and first-harmonic ordinary waves, respectively.

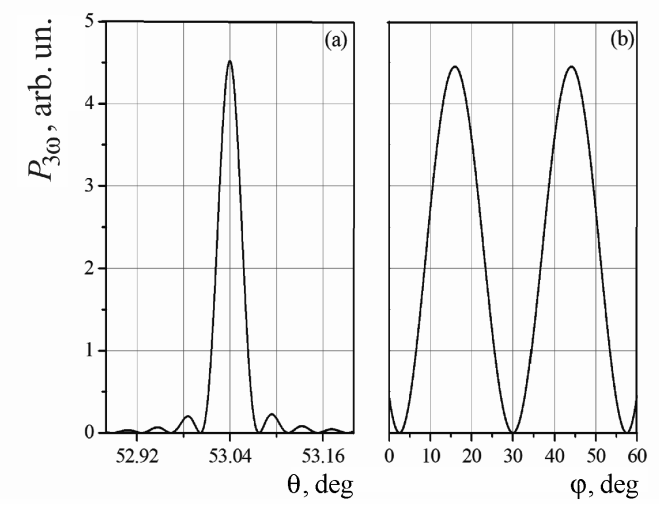

Fig. 1. Theoretical dependences of the third-harmonic power density on the polar (a) and azimuthal (b) angles.

The results calculated for the angular dependences of the third-harmonic power are presented in Fig. 1. For the case of BBO pumped with the Ti:Sa laser, for which $\lambda_p = 780$ nm, the theoretical polar and azimuthal phase matching angles are equal to $\theta_S = 53.04°$ and $\varphi_S = 15°$, respectively. Those θ_S and φ_S values correlate with those presented in [3] ($\theta_S = 55.04°$ and $\varphi_S = 7°$ for 100-fs laser pump pulses of 800-nm wavelength).

The phase matching angles obtained by us have been used when calculating dependence of the third harmonic power density on the pump power density. The dependence $P_{3\omega}(P_\omega)$ calculated using Eq. (1) for the range of $P_\omega < 90$ GW/cm^2 is displayed in Fig. 2.

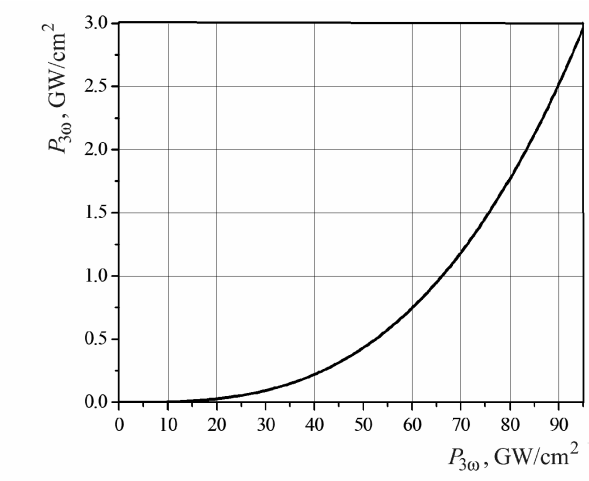

Fig. 2. Dependence of the third-harmonic power density $P_{3\omega}$ on the pump power density P_ω for a 3-mm long BBO sample.

The results show that the theoretical efficiency of transformation into the third-harmonic radiation amounts to 3 per cent in the power range used in our experiments. When analysing contributions of the processes of second and third orders, one should admit that the choice of the method for their separation is essential. Usually such separation is made by comparing the shape of theoretical and experimental polar dependences of the third-harmonic power, as well as the theoretical and experimental phase-matching widths. In the present work we suggest analysing the

relative contributions of the processes of cascaded and direct THG, basing in addition on dependence of the third-harmonic power on the pump radiation power. It is known from the theory [4] that this dependence should be quadratic:

$$P_{2\omega} = \frac{L^2\omega^2}{2\varepsilon_0^2 c^3 A}\frac{1}{n_\omega^{o\,2} n_{3\omega}^e} d_{eff}^2 P_\omega^2 \operatorname{sinc}^2 \frac{\Delta kL}{2} \ .$$

To sum up, we take account of (a) the assumption [3] about additivity of the effects contributing to the THG process (see Eq. (2)), (b) the quadratic character of the dependence $P_{2\omega}(P_\omega)$, and (c) the cubic character of the dependence $P_{3\omega}(P_\omega)$. As a consequence, the dependence $P_{3\omega}(P_\omega)$ obtained experimentally can be approximated by a polynomial containing both quadratic and cubic terms:

$$P_{3\omega}^{app}(P_\omega) = B_1 P_\omega^{b1} + B_2 P_\omega^{b2} + C P_\omega^c , \tag{4}$$

where the terms $B_1 P_\omega^{b1}$ ($b1 = 2$) and $B_2 P_\omega^{b2}$ ($b2 = 2$) are brought into compliance with the processes of direct 'asynchronous' second-harmonic generation and generation of the third harmonic as a result of sum-frequency generation, while the term CP_ω^c ($c = 3$) describes the process of direct THG due to the third-order nonlinearity.

It should be pointed out that any light losses caused by elements present in the optical path can also affect the character of $P_{3\omega}(P_\omega)$ dependence and so they would result in inaccurate $b1$, $b2$ and c values. Finally, the contribution of each optical process to the THG may be described by the weighting coefficient of the corresponding polynomial term.

3. Experimental results

Fig. 3 shows the experimental setup used by us for the THG studies in the BBO crystal. In all the experiments, a 3-mm long crystal BBO plate was used. It was cut at the angle of $\theta_S = 53°$ and one of its facets coincided with the crystallographic plane (110). The maximum nonlinear transformation efficiency was achieved by orienting the sample at the azimuthal angle $\varphi_S \approx 13°$.

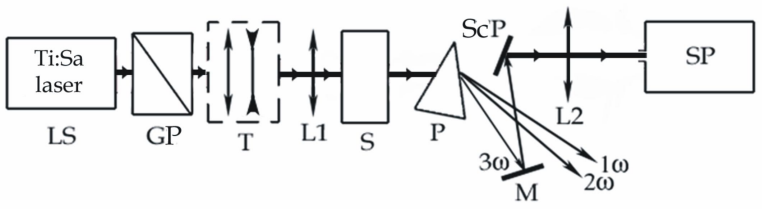

Fig. 3. Experimental setup for measuring the THG in the BBO crystal: LS – femtosecond Ti:Sa laser; GP – Glan prism; T – lens telescope; L1 – focusing lens; S – BBO sample; P – prism; M – mirror; ScP – scattering plate; L2 – exit focusing lens; SP – spectrophotometer.

A Ti:Sa laser (Coherent Mira Optima 900-F As) was employed as a source of femtosecond pulses (the wavelength of 800 nm and the repetition pulse rate of 1 kHz). An external compensator Coherent SPO-I allowed getting pulses of 70 fs duration. The spectra were detected by a spectrophotometer SP-2500i with the aid of CCD camera (CCD-Spec-10:256E/TEPLUS 1024×256 Open-electrode, Marconi CCD 30-11), which was sensitive in the wavelength range of 200–1000 nm.

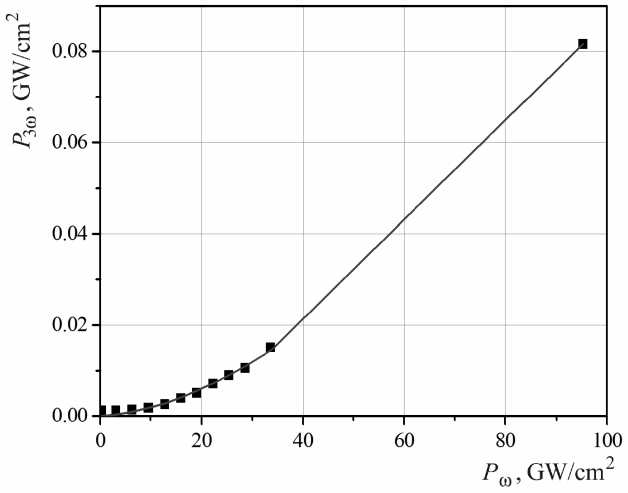

Fig. 4. Third-harmonic power density versus pump power density: dots imply the experiment and a solid line the polynomial given by Eq. (4).

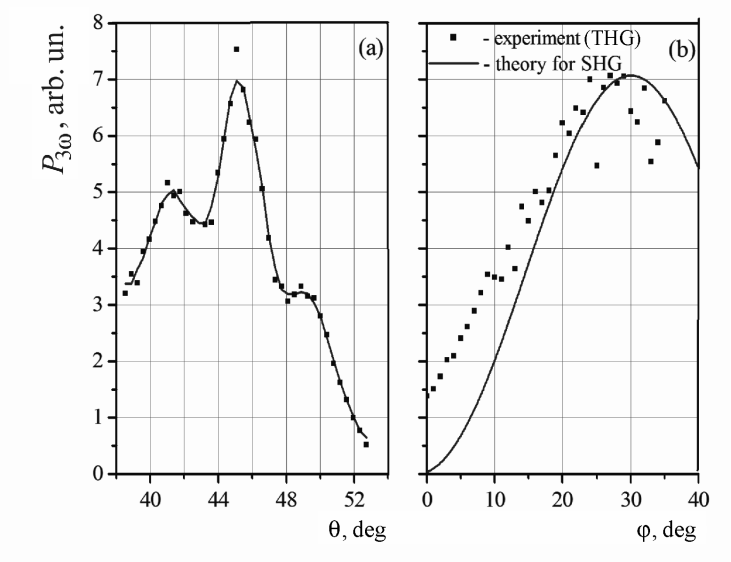

Fig. 5. Dependences of the third-harmonic power density on the angles θ (at φ = 13 deg) (a) and φ (at θ = 45 deg) (b).

In order to decrease laser beam diameter and increase the pump power density, while preserving the beam parallelism, a lens telescope was used. Finally, variations of the pump radiation power were achieved by placing a Glan prism into the optical beam path.

In Fig. 4 we present the dependence of the third-harmonic power density on the pump power density obtained experimentally. Besides, Fig. 5 shows the experimental results concerned with dependences of the third-harmonic intensity on the polar and azimuthal angles.

4. Discussion

As pointed out above, the processes of both direct and cascaded THG could likely be potential sources of the third-harmonic radiation. As the BBO represents a highly nonlinear crystal [1], we assume that the parametric generation process could also be one of the sources of the mentioned radiation. The calculated polar angles, at which the processes of collinear and noncollinear parametric generation with the wavelength of the third harmonic (266.7 nm) could take place, are equal to 20.6° and 30.2°, respectively. Then, taking account of the fact that the polar angle is changed in the region of 38–54° in the course of our experiments, one can claim that the parametric processes do not contribute to the THG.

As we have already mentioned, the conventional method adopted in the analysis of contributions of different processes to the THG is to compare azimuthal and polar curves of the third-harmonic intensity predicted by theory and obtained experimentally. The theoretical angular phase-matching width with respect to the polar angle amounts to $\sigma_\theta = 0.04°$ (see Fig. 1), while the corresponding experimental value is $\sigma_\theta = 7°$ (see Fig. 5). At the same time, the angular phase-matching width calculated for the noncollinear cascaded THG as a result of summation of the first and second harmonics is $\sigma_\theta = 6°$.

Hence, the third-harmonic power oscillations due to variations of the polar angle (see Fig. 5a) are associated for the most part with the corresponding second-harmonic power oscillations. Indeed, the latter lead to the third-harmonic oscillations while interacting with the pump radiation. The calculated polar-angle period of the second-harmonic intensity oscillations correlates well with that of the third-harmonic oscillations obtained experimentally. The analysis of dependence of the third-harmonic intensity upon the azimuthal angle also proves that the role of cascaded THG under the conditions of our experiments should be dominating. The experimental phase-matching width and the shape of the curve (see Fig. 5b) are close to the results of theoretical calculations for the second-harmonic power oscillation depending on the azimuthal angle.

The contributions of the direct and cascaded THG have also been estimated basing on the analysis of our experimental dependence $P_{3\omega}(P_\omega)$. The approximation of the experimental curve $P_{3\omega}(P_\omega)$ with the polynomial given by Eq. (4) is presented in Fig. 4. The parameters of the polynomial presented in Table 1 also indicate a dominant contribution of the cascaded THG process.

It should also be pointed out that the experimental value of the relative third-harmonic power density observed at the maximum pump power density ($P_{\omega\ \max} = 95$ GW/cm^2) is equal to 0.01%. According to Fig. 2, the theoretical third-

harmonic power density $P_{3\omega\ \max}$ should be 3 per cent. This discrepancy may be explained in the following way. It is known from the common theory of nonlinear processes that the pump radiation should meet certain conditions for the most efficient transformation of that radiation into the radiation of higher harmonics: the angular divergence of the output radiation should be smaller than the angular phase-matching width and its spectral width should not exceed the frequency phase-matching width [6]. Otherwise not all of the pump power would take part in the process of nonlinear transformation. Since the duration of the pump radiation has been 70 fs in our experiments, its spectral width 10 nm and the angular divergence has remained significant due to beam focusing, the conditions mentioned above are not fulfilled. This has resulted in substantial decrease of the THG efficiency.

Table 1. Parameters of polynomial given by Eq. (4) used for fitting the experimental dependence presented in Fig. 4.

Parameter	B_1, GW/cm^2	$b1$	B_2, GW/cm^2	$b2$	C, GW/cm^2	c
Value	3×10^{-5}	1.7	2×10^{-5}	1.7	3.1×10^{-10}	3

In order to illustrate these facts, one can mention the results presented in the works [2, 3]: under the conditions of comparable pump power densities ($P_\omega \approx 300$ GW/cm^2) and differing pulse lengths (350 fs in [2] and 100 fs in [3]), the efficiency of the THG differs by 6 times, being 6 per cent in the study [2] and only 1 per cent in [3]. This implies that a decrease in the pulse duration would result in decreasing contribution of the processes induced by the second-order nonlinearity to the THG.

An extremely low third-harmonic power obtained at the polar angle corresponding to the phase-matched THG also gives evidence of the absence of frequency transformation induced by the third-order nonlinearity. One can assume that changing experimental conditions by reducing the angle divergence of the pump beam and increasing the pump power density should yield in different relative second- and third-order contributions to the THG. In its turn, this will result in a shift of the $P_{3\omega}(\theta)$ dependence towards the angular region calculated for the case of direct phase-matched THG, as well as in increasing phase-matching width.

5. Conclusions

Basing on our results and their analysis, one can conclude that only the optical process induced by the second-order nonlinearity is observed experimentally under the conditions valid for the present study (the pump power density as low as 100 GW/cm^2, τ_p = 70 fs, and the BBO crystal cut prepared for the phase-matched THG). The oscillations of the third-harmonic power appearing due to changes in the polar angle should be related to the power oscillations of the second harmonic, which is generated ‘asynchronously’ (i.e., out of the phase matching). This proves that the THG is a result of only one of the possible nonlinear processes.

The low THG efficiency got in the course of our experiments (0.01%) is caused by such factors as a comparably low pump power density, rather high spectral width, and a substantial angular divergence of the pump beam.

Investigations of dependence of the third-harmonic power density on the pump pulse duration and polar and azimuthal dependences of the THG intensity, for the case when the THG effect originates mainly from the cubic nonlinearity $\chi^{(3)}$, are under progress. Their results will be reported in a forthcoming article.

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Анотація. *Описані результати дослідження генерації третьої гармоніки в кристалах | β-BaB_2O_4, вирізаних під кутом θ_S, який забезпечував пряму генерацію в умовах фазового синхронізму типу - ооое. При інтерпретації експериментальних даних використовувалась теорія прямої генерації третьої гармоніки в одновісних кристалах, яка передбачає адитивність нелінійно-оптичного коефіцієнта. Для встановлення домінуючого внеску в генерацію третьої гармоніки проаналізована кутова залежність її інтенсивності і залежність інтенсивності від потужності хвилі нагнітання. На основі аналізу експериментальних результатів, за умови нагнітання кристалів β-BaB_2O_4 ($\theta_S = 53°$) фемтосекундними імпульсами ($\lambda = 800$ нм, $\tau = 70$ фс, і $P_{max} = 100$ ГВт/см^2) показано, що основним внеском в генерацію третьої гармоніки є каскадний процес генерації за рахунок квадратичної нелінійності.*