Nonlinear absorption at 266 nm in BBO crystal
and its influence on frequency conversion

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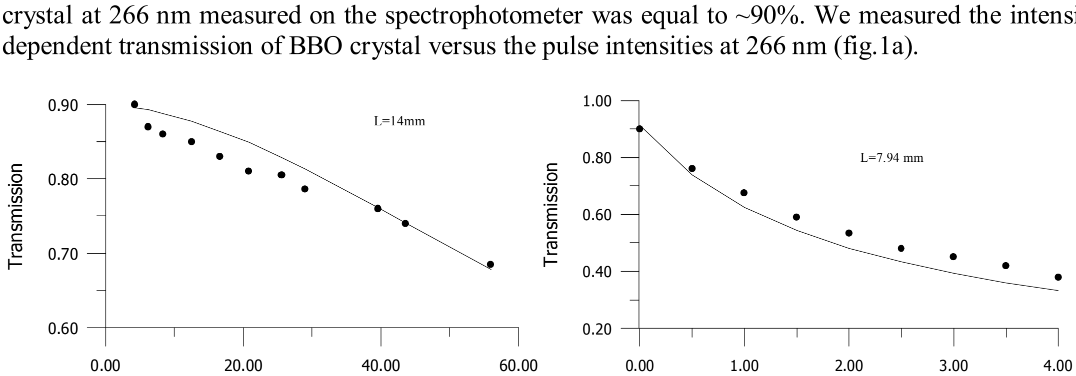
ABSTRACT

In the present paper we represent the results of investigations of an ultraviolet (266-nm) –induced nonlinear absorption in BBO crystals and its influence on frequency conversion. It was made the assumption that in BBO crystals at two-photon absorption of law intensive radiation < 100 MW/cm^2 at 266 nm dynamic color-centers are generated. With the help of computational modeling it was defined that the absorption cross section of dynamic color-centers at 266 nm is equal to ~ $8\cdot10^{-17}$ cm^2.

On the basis of experimental investigations and computational modeling it was shown that the main factors restricting the fourth-harmonic generation efficiency of a Nd:YAG laser in BBO crystals are: law angular bandwidth, nonlinear absorption at 266 nm and effects of thermal self-actions. It was shown that in strong energy exchange conditions between interactive waves in BBO crystal nonlinear losses on pump radiation at 266 nm are becoming smaller and the OPA efficiency in UV range considerably surpasses the OPO efficiency. In the OPA on 14 mm long BBO crystal with pump energy ~ 35 mJ and intensity ~40 MW/cm^2 at 266 nm the output energy in UV range was up to 10 mJ and efficiency >25% at 327 nm.

1. NONLINEAR ABSORPTION IN BBO CRYSTALS

KDP, BBO and LBO crystals are widely used for radiation generation in UV portion of spectrum. It is known that one of the factors restricting the efficiency of non-linear conversions in UV portion of spectrum is two-photon absorption of high intensive UV radiation in crystals[1]. Two-photon absorption coefficients for KDP, BBO and LBO crystals at 264 nm are equal to 0,26 cm/GW, 0,93 cm/GW и 0,15 cm/GW respectively[2]. As a rule, two-photon UV radiation absorption process is accompanied with electron crossing from the valence band into the conduction band with the following color-centers creation. These color-centers auxiliary absorb radiation over the whole three frequencies involved in nonlinear interactions[3]. It was found out that in BBO-crystals non-linear absorption of low-intensive <100 MW/cm^2 radiation of the 4th harmonic of Nd:YAG laser exists. In our experiments we used commercial Q-switched Nd:YAG laser with the 2nd and the 4th harmonic generators (model LQ-727, Solar Laser Systems). This system operated a 10 Hz repetition rate and provided 80 mJ, ~5 ns pulses at 266 nm. With the help of hard aperture diaphragm the central part of the beam of the 4-th harmonic was cut off, where the distribution of intensity was homogeneous and close to the rectangle profile. We used Fresnel attenuator for varying the 4th harmonic intensity from 4 MW/cm^2 to 65 MW/cm^2. The nonlinear losses were measured with 14 mm long AR-coated BBO-crystal, cut at the angle θ=38,3°. The reflection coefficient of AR-coating at 266 nm was equal to ~2,5%. The transmission of BBO-crystal at 266 nm measured on the spectrophotometer was equal to ~90%. We measured the intensity-dependent transmission of BBO crystal versus the pulse intensities at 266 nm (fig.1a).



a) b)

Fig.1. Intensity-dependent transmission curve for a) – nanosecond pulses and b) – subpicosecond pulses[2] at 266 nm. The symbols depict the experimental data, and the solid curves show the best numerical fit.

This nonlinear absorption takes place only at the time of laser pulses action. We assumed that during two-photon absorption process at 266 nm inside BBO crystal the dynamic color-centers are generated. Thus, nonlinear absorption in BBO crystal is caused by both two-photon absorption and dynamic color-centers absorption.

2.NUMERICAL MODELING OF NONLINEAR ABSORPTION IN BBO CRYSTALS

In our calculations we assumed that color-centers are created immediately as the result of two-photon absorption of 266 nm light and their bleaching during the whole pulse is negligibly small. In this case the equations governing the color-centers density N and intensity $I_{4\omega}$ have the form[3]:

$$\frac{dN(z,t)}{dt} = \frac{\beta \cdot I_{4\omega}^2(z,t)}{8\hbar\omega}, \qquad (1)$$

$$\frac{dI_{4\omega}(z,t)}{dt} = -\alpha_{4\omega} \cdot I_{4\omega}(z,t) - N(z,t)\cdot\sigma_{4\omega}\cdot I_{4\omega}(z,t) - \beta \cdot I_{4\omega}^2(z,t), \qquad (2)$$

with an initial conditions of $N(z,t=0)=0$, (3)

where β-is two photon absorption coefficient, $\sigma_{4\omega}$ - color-centers absorption cross section at 266 nm, $\alpha_{4\omega}$ - linear absorption coefficient at 266 nm, $\hbar\omega$ - fundamental photon energy at 1064 nm. In the equation (2) the first term describes linear absorption, the second – color- centers absorption and the third – two-photon nonlinear absorption. The temporal pulse shapes can be described by the function:

$$f(t) = \exp\left\{-\left[2\sqrt{\ln 2}\cdot\left(\frac{t}{t_p} - 1{,}29\right)\right]^2\right\} \qquad (4)$$

where t is the current time in $[0,\infty]$, t_p – pulse width for 266 nm light. For the numerical calculations the pulse envelope of the radiation being converted was approximated by a step-wise function with Δt step-width and constant field intensity inside the each step: $I_n(0,t_n) = I_{4\omega}^{max}\cdot f(t_n)$, where n is the step number, $I_{4\omega}^{max} = 0{,}94\cdot\frac{4\cdot E_{4\omega}(0)}{\pi\varnothing^2 t_p}$ - maximum pulse intensity, $E_{4\omega}(0)$ - input energy, ∅ - beam diameter. For each step eq. (1) has explicit solution and color-centers density is

$N(z,t_n) = N(z,t_{n-1}) + \frac{\beta\cdot I_{4\omega}^2(z,t_n)}{8\hbar\omega}\cdot\Delta t$, where $N(z,t_{n-1})$ – is the color-centers density for previous interval of time $[0,t_{n-1}]$. Equation (2) was solved with a Runge-Kutta numerical routine for each spatial slice in the crystal and output energy at 266 nm was calculated as $E_{4\omega}(\ell) = \frac{\pi\varnothing^2}{4}\cdot\sum_{n=1}^{N} I_n(\ell,t_n)\cdot\Delta t$.

Table 1 presents the parameter values used for modeling the data in fig.1.

Table 1

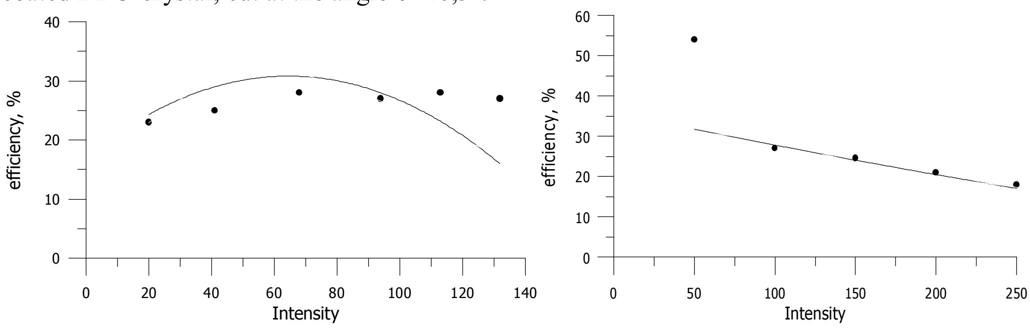
parameters (units)	for nanosecond pulses	for picosecond pulses [2]
Input energy $E_{4\omega}(0)$, mJ	27	<3
Pulse-width τ, ns	5	$\sim 0{,}8\cdot10^{-3}$
Beam diameter ∅, mm	3,5	3,3
Linear absorption coefficient $\alpha_{4\omega}$, cm^{-1}	0,02	0,02
Two-photon absorption coefficient β, cm/GW	0,93	0,93
AR coating reflectivity, %	0,025	0,025
Crystal lenght ℓ, mm	14	7,94

Taking into consideration experimental data we selected color-centers absorption cross section value $\sigma_{4\omega} = 8\cdot10^{-17}$ $см^2$, when calculated dependence of BBO crystal transmission on the nanosecond pulses has a good correlation with experimental results (fig. 1a)

3. NONLINEAR ABSORPTION IN BBO CTYSTAL AND IT'S INFLUENCE ON THE EFFICIENCY OF THE 4TH HARMONIC GENERATION

In the combination of their qualities BBO crystals have good prospects for effective and stable generation of the 4th harmonic of Nd:YAG lasers. However in the major of published papers[1] is reported that the efficiency of the 4th harmonic generation in BBO crystals quickly saturates and is less than 40%. The efficiency $\eta_{4\omega}$ of the fourth harmonic generator of Nd:YAG laser Coherent Infinity 40-100 on BBO crystals was investigated[4]. It was shown that in 5 mm long BBO crystal the efficiency $\eta_{4\omega}$ reaches 68% at intensity $I_{2\omega}\approx 50$ MW/cm^2 and pulse repetition rate f=10 Hz. However, when frequency increases up to 100 Hz the efficiency $\eta_{4\omega}$ decreases down to 20%. In previously mentioned works it's suggested that the efficiency $\eta_{4\omega}$ depends on nonlinear absorption and effects of thermal self-actions.

In the present paper nonlinear absorption and effects of thermal self-actions influences on the efficiency $\eta_{4\omega}$ in BBO crystals is being modeled. As the pump source was used the commercial Q-switched Nd:YAG laser with the 2nd harmonic generator (model LQ-727, Solar Laser System). This laser operated at 10 Hz repetition rate and provided 180 mJ, 5 ns pulses at 532 nm. The divergence of 5.7 mm diameter light beam of the 2nd harmonic was ~0.7 mrad. The spatial radiation beam profile was homogeneous and close to the rectangle one. The measurements were carried out with 7 mm long AR-coated BBO-crystal, cut at the angle θ=48,3°.



a) b)

Fig.2. Fourth harmonic conversion efficiency vs. second harmonic intensity at 10 Hz repetition rate for a) – LQ-727 and b) – Infinity Nd:YAG lasers[4]. The symbols depict the experimental data and the solid curves show the best numerical fit.

The efficiency $\eta_{4\omega}$ of Nd:YAG laser LQ_727 is saturated at the level ~ 28% at intensities $I_{2\omega}>20$ MW/cm^2 (fig.2a). The efficiency $\eta_{4\omega}$ of Infinity in 7mm long BBO crystal quickly decreases at intensities $I_{2\omega}>70$ MW/cm^2 (fig.2b)[4].

To explain these peculiarities we carried out numerical calculations of of the efficiency $\eta_{4\omega}$ in BBO crystal taking into consideration nonlinear absorption at 266 nm and effects of thermal self-actions. The equations governing the fields E_2 and E_4 during nonlinear frequency conversion in BBO from 2ω to 4ω are the following[3]:

$$\begin{cases} \dfrac{dE_2(z,t)}{dz} = -\dfrac{\omega_2 \cdot d_{eff}}{n \cdot c_o} \cdot E_2 \cdot E_4 \cdot \sin\theta - \dfrac{1}{2} \cdot \alpha_{2\omega} \cdot E_2, \\ \dfrac{dE_4(z,t)}{dz} = 2 \cdot \dfrac{\omega_2 \cdot d_{eff}}{n \cdot c_o} \cdot E_2^2 \cdot \sin\theta - \dfrac{1}{2} \cdot (\alpha_{4\omega} + \sigma_{4\omega} \cdot N(z,t)) \cdot E_4 - \dfrac{1}{4} \cdot \beta \cdot n\varepsilon_o c_o E_4^3, \\ \dfrac{d\theta}{dz} = \Delta k - \dfrac{2\omega_2 \cdot d_{eff}}{n \cdot c_o} \cdot (E_4 - \dfrac{E_2^2}{E_4}) \cdot \cos\theta, \\ \dfrac{dN(z,t)}{dz} = \left(\dfrac{1}{2} \cdot n \cdot \varepsilon_o \cdot c_o\right)^2 \cdot \dfrac{\beta}{2\hbar\omega_4} \cdot E_4^4, \end{cases}$$

where d_{eff} is effective nonlinear coefficient,, n – refractive index, $\alpha_{2\omega}$ and $\alpha_{4\omega}$ - linear absorption coefficients for 2w and 4w, c_o –speed of light in vacuum, ε_o – permittivity of vacuum, $\hbar\omega_4$ - photon energy of the 4th harmonic, Δk – total mismatch. The total mismatch $\Delta k=\Delta k_L+\Delta k_{TSA}(z)$ consists from linear mismatch Δk_L, which does not depend on the temperature and the mismatch due to the thermal self-actions $\Delta k_{TSA}(z)=\frac{4\pi}{\lambda_2}\cdot\left[n_2^o(z,T)-n_4^e(z,T)\right]$, where n_2^o and n_4^e - refractive indexes for 2w and 4w at point (z) with temperature T, λ_2 is the wavelength of 2w. For numerical calculations temperature wave mismatch Δk_{TSA} was used in the form $\Delta k_{TC}=278\cdot\frac{\Delta T}{\Delta T_c}\cdot\frac{\ell}{0{,}01}$ [rad/m], where ℓ - is BBO crystal length (in meters), ΔT–crystal temperature changing due to absorption of the 4th harmonic radiation,. ΔT_c –temperature bandwidth In the case of quasi-static approximation, when the 2nd harmonic pulse approximates with the step-wise function, for the step number n at point (z) temperature changes for the time n·Δt can be calculated $\Delta T(z,t_n)=\Delta T(z,t_{n-1})+\delta T(z,t_n)$,where $\Delta T(z,t_{n-1})$ - temperature changes for a previous interval of time $[0,t_{n-1}]$, δT_n - temperature jump after heating, which can be found from the thermal conductivity equation: $c\cdot\rho\cdot\delta T_n(z,t_n)=\left[\alpha_{4\omega}+\sigma_{4\omega}\cdot N(z,t_n)\right]\cdot I_{4\omega}(z,t_n)\cdot\Delta t$, where c – specific heat, ρ - crystal density, $I_{4\omega}(z,t_n)=\frac{1}{2}n\varepsilon_o c_o E_4^2(z,t_n)$ - fourth harmonic intensity.

The calculated dependencies of the efficiency $\eta_{4\omega}$ versus the intensity $I_{2\omega}$ are given in fig.2. In both cases there is a good correlation between experimental data and calculated dependencies for both lasers LQ-727 and Infinity.

4. NONLINEAR ABSORPTION IN BBO CRYSTAL AND IT'S INFLUENCE ON THE PARAMETRIC CONVERSIONS

In the formerly published article[5] the results of experimental investigations of the efficiency of a BBO OPO pumped by the 4th harmonic of a Nd:YAG laser were given. At the pump intensity ~46 MW/cm^2 we obtained the output tunable in the range 300…2340 nm with the efficiency ~15% at double the oscillation threshold at λ_s=340 nm.

On the basis of carried out researches we can make the conclusion that in conditions of weak energy exchange on the linear stage when the intensities of idler and signal waves are small, the pump radiation losses caused by nonlinear absorption are at the most. In the conditions of strong energy exchange between interactions waves in nonlinear crystal the intensity of pump radiation is considerably less and consequently the pump energy losses caused by nonlinear absorption must be less. Proceeding from this assumptions we can make the conclusion that the OPA efficiency could be larger than the OPO one if there are effects of nonlinear absorption of pump radiation.

In our experiments we used commercial Q-switched Nd:YAG laser with the 2nd and the 4th harmonic generators (model LQ-727, Solar Laser Systems). The average pump intensity of BBO OPA was equal to ~40 MW/cm^2 and pump energy was equal to ~35 mJ. The nonlinear losses were measured in 14 mm long AR-coated BBO-crystal, cut at the angle θ=38,3°. Simultaneously with the pump pulses at 266nm the injection pulses at 1427 nm with bandwidth ~2,5 were used. Injected aperture restricted beam ~5 mm in diameter had divergence ~5 mrad. BBO-crystal transmission at 1426 nm was equal to ~84%. Initially BBO-crystal was installed in the position, when phase matching conditions for OPA at λ_i=1426 nm took no place. In this case the output pulse energy at 266nm was smaller and equal to $E_p(l)$=24 mJ (the crystal transmission at 266 nm ≈ 69%). At injection pulse with energy $E_p(o)$=6 mJ in the OPO output the total pulse energy of two waves $E_i(l)+E_p(l)$=29 mJ, where $E_i(l)$=5 мmJ and $E_p(l)$=24 mJ. When BBO-crystal was installed in the position, when phase matching conditions for the OPA at λ_i=1426 nm took place, signal wave at λ_s=324 нм was generated and in the OPA output the total pulse energy of three waves $E_i(l)+E_s(l)+E_p(l)$=32 mJ, where $E_p(l)$=16 mJ, $E_i(l)+E_s(l)$=16 mJ. Thus, one can see, that in the strong energy-exchange between interacting waves energy losses in OPA approximately 3 mJ less as compared with the case of energy exchange absence.

At the wavelength of injected radiation tuning in the range 725…2350 nm the tunable radiation in UV range from 300 nm to 420 nm was obtained. The OPA output energy in UV range was up to 10 mJ and efficiency >25% at 327 nm, fig.3.

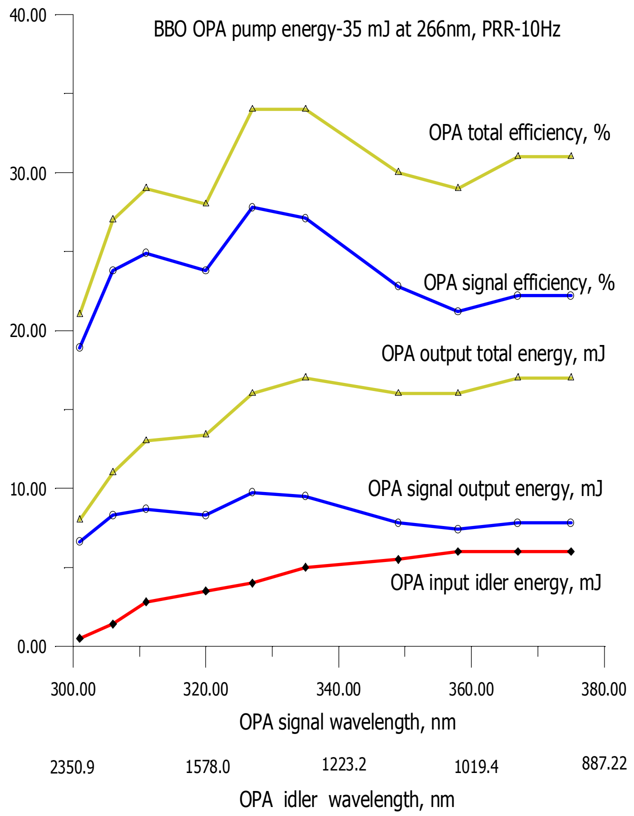


Fig.3. High efficiency BBO OPA system in the range 300…400nm pumped by the 4th harmonic Nd:YAG laser

From the represented dependencies one can see that at the same pump intensities the OPA efficiency in UV range considerably surpasses the OPO efficiency, measured formerly[5].

5. REFERENCES

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