Pis'ma v ZhETF, vol. 74, iss. 12, pp. 655 – 659

Theoretical treatment, simulation of new UV crystal $CsLiB_6O_{10}$ on third-harmonic generation

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Submitted 5 Nowember 2001

We have calculated the theoretical properties of the nonlinear crystal $CsLiB_6O_{10}$ (CLBO) in third-harmonic generation (THG). Such as the phase matching angle, effective nonlinear coefficient, walk-off angle, permitted angle, permitted wavelength, and have found out that the CLBO has smaller walk-off angle, large permitted parameters as compared with BBO. The numerical simulation curves of the conversion efficiency are obtained, when the CLBO is used in THG of the Q-switched Nd:YAG laser with the wavelength of 1064 nm and of the different pump power, and the optimized efficiency is as high as 22%, all of which followed that the CLBO crystal is more suitable to generating a higher order harmonic radiation of intensity power.

PACS: 42.65.-k

1. Introduction. The exploration of the solid ultraviable (UV) laser is valuable for its high power, little volume, and had been the pioneers of the UV lasers on the latest nonlinear crystal $CsLiB_6O_{10}$ (CLBO) [1, 2]. However, the shortest transparent band of them is always very long, for example, the shortest transparent band of BBO is only 190 nm and 175 nm of the CLBO [3, 4], and also has high damage threshold, and many other beneficial optical properties. The present work on CLBO is its SHG and third-harmonic generation (THG) in the Nd:YAG laser of picosecond (ps) or nanosecond (ns) and with the fundamental wavelength of 1064 nm in experiment, and there is no report on its properties in theory, especially on its THG. In this paper, we calculated, and numerical stimulated its theoretical curves on the phase matching (PM) angle, effective nonlinear coefficient, walk-off angle, permitted angle and wavelength, and conversion efficiency of CLBO in THG, and have obtained where there is smaller walk-off angle, broader permitted parameters and high conversion efficiency on UV, all of being proved CLBO is more suitable to produce higher order radiation of high power, and present its prospect in the high power UV solid lasers.

2. Computer simulation of permitted parameters on CLBO. Take the collinear interaction into consideration, we supposed that ω_1 (λ_1) and ω_2 (λ_2) were the frequency (wavelength) of the incident radiation respectively, and ω_3 (λ_3) denoted frequency (wavelength) of the third harmonic wave, and ω_1 was lower than ω_2, both of which were lower than ω_3.

2.1. *PM angle (θ)*:

A) PM angle of type-I ($o + o \to e$). According to the momentum and energy conservation laws, we can obtain:

$$n_1(\omega_1) + 2n_2(\omega_2) = 3n_3(\omega_3, \theta), \quad (1)$$

$$\omega_1 = \omega_2/2 = \omega_3/3. \quad (2)$$

Where n_1, n_2 and n_3 are the reflective indices of the three waves. From (1) and (2), the PM angle (θ) of THG in type-I could be derived:

$$\theta(\mathrm{I}) = \arcsin\left(\frac{\dfrac{9n_0^2(\omega_3)n_e^2(\omega_3)}{[n_1(\omega_1)+2n_2(\omega_2)]^2} - n_e^2(\omega_3)}{n_o^2(\omega_3) - n_e^2(\omega_3)}\right)^{1/2}. \quad (3)$$

B) PM angle of type-II ($e + o \to e; o + e \to e$). We define the type-II(1) PM in THG is as follows: $e+o \to e$, it means that the radiations which the frequency of ω_1 and ω_2 are of e and o polarized respectively; and the other was type-II(2), $o + e \to e$, where the waves of ω_1 and ω_2 were o and e polarized light, respectively. The θ of type-II(1) and type-II(2) in THG were written as

$$3\left[\frac{\cos^2\theta(\mathrm{II}(1))}{n_0^2(\omega_3)} + \frac{\sin^2\theta(\mathrm{II}(1))}{n_e^2(\omega_3)}\right]^{-1/2} = 2n_0(\omega_2) + \left[\frac{\cos^2\theta(\mathrm{II}(1))}{n_o^2(\omega_1)} + \frac{\sin^2\theta(\mathrm{II}(1))}{n_e^2(\omega_1)}\right]^{-1/2} \quad (4)$$

$$3\left[\frac{\cos^2\theta(\mathrm{II}(2))}{n_0^2(\omega_3)} + \frac{\sin^2\theta(\mathrm{II}(2))}{n_e^2(\omega_3)}\right]^{-1/2} = n_0(\omega_1) + 2\left[\frac{\cos^2\theta(\mathrm{II}(2))}{n_o^2(\omega_2)} + \frac{\sin^2\theta(\mathrm{II}(2))}{n_e^2(\omega_2)}\right]^{-1/2}. \quad (5)$$

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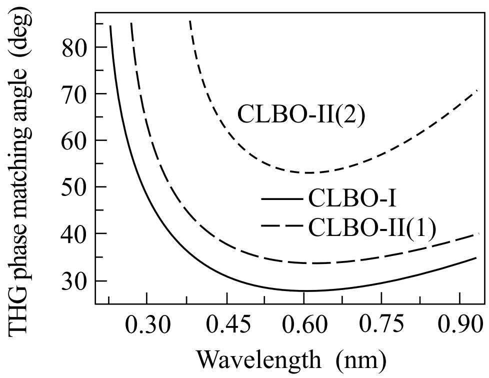

Fig.1. Curves of the phase matching angle versus wavelength of the third harmonic radiation of different type THG for the CLBO crystal

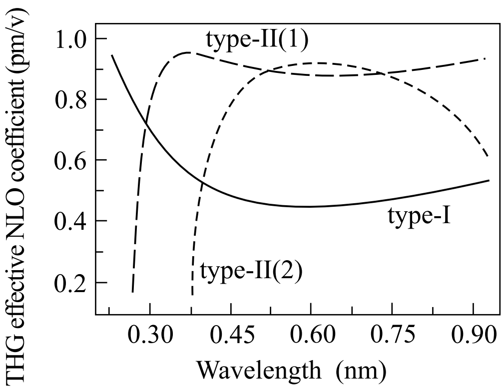

Fig.2. Curves of the effective nonlinear coefficient about the wavelength of third harmonic wave for THG of the CLBO crystal

From (3), (4), (5) and the sellimier equations of CLBO [1]

$$n^2_{o(\lambda)} = 2.2145 + \frac{0.00890}{\lambda^2 - 0.02051} - 0.01413\lambda^2 \,\square, \quad (6)$$

$$n^2_{e(\lambda)} = 2.0588 + \frac{0.00866}{\lambda^2 - 0.01202} - 0.006073\lambda^2. \quad (7)$$

We could get the θ of CLBO in THG. The curves of CLBO's θ versus the wavelength of the THG wave is shown in Fig.1, where we could find out that the shortest wavelength of CLBO's THG is 277 nm, when CLBO was used as type-I PM and the θ is 85 deg. The range of the λ_3 wave is the broadest in the type-I THG, where the θ is about from 25 deg to 85 deg, and the range is the shortest in the type-II(2) PM, where the θ is from 55 deg to 85 deg. It is also presented in Fig.1 that there is no PM angle in type-II THG about CLBO when the wavelength of the λ_1 is between 680 nm and 800 nm, and the type-I PM must be utilized. It is necessary that the crystal has a large birefringence in the type-II(2) THG, and then it is seldom used in practice [5].

2.2. *Effective nonlinear coefficient (d_{eff}).* The CLBO is a negative uniaxial crystal with the space group of $\bar{4}2m$, and we could derive its d_{eff} in considering the kleinman equation [6]

$$d_{\text{eff}}(\text{I}) = d_{36} \sin\theta \sin(2\phi), \quad (8)$$

$$d_{\text{eff}}(\text{II}) = d_{36} \sin 2\theta \cos(2\phi), \quad (9)$$

where θ and φ are the orientation angle of CLBO. It is supposed that φ was 45 deg and 0 deg in type-I and type-II THG, respectively, and θ is inserted in (8) and (9) of the Fig.1, at the same time, d_{36} is caused to be 0.95 pm/V [1], then we could get the d_{eff} curves versus the wavelength of ω_3 of CLBO in THG, as was shown in Fig.2. In Fig.2, the optimized d_{eff} is 0.95 pm/V, when the wavelength of ω_3 is 227 nm and 377 nm in type-I and type-II(1) respectively.

2.3. *Walk-off angle (α).* The formula of the walk-off angle [7]:

$$\alpha = \tan^{-1}\left[\frac{1}{2}\frac{(n_e^2(\lambda) - n_o^2(\lambda))}{n_o^2(\lambda)\sin^2\theta + n_e^2(\lambda)\cos^2\theta}\sin(2\theta)\right], \quad (10)$$

where θ is the angle in Fig.1. Considering (6) and (7), we insert θ of correspondent PM in Fig.1 into (10), and then get the curves of the walk-off angle of CLBO in type-I, type-II(1) and type-II(2) THG, which are shown in Fig.3, Fig.4, and Fig.5, where BBO's walk-off angles are obtained in the same way, and axis-x is the wavelength of the ω_3 between 227 nm and 933 nm. From Fig.3, Fig.4 and Fig.5, we can found out that the walk-

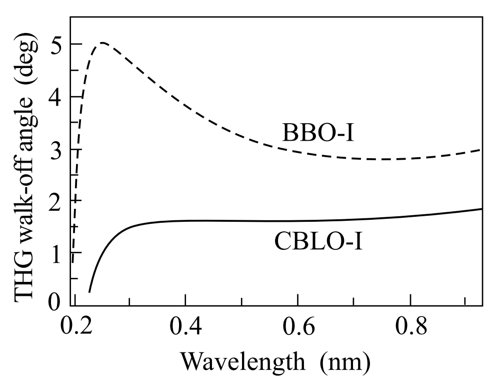

Fig.3. THG walk-off angle curves of type-I phase matching for CLBO and BBO

off angles of CLBO are all far smaller than the ones of BBO under the same circumstance.

2.4. *Permitted parameters*:

A) Permitted angle ($\Delta\theta$). The permitted angle $\Delta\theta$ of CLBO in THG of different PM state is as follows:

$$\Delta\theta = \pm \left| \frac{\pi/l}{\frac{\omega}{c} n_e^3(\omega_3,\theta)[n_e^{-2}(\omega_3) - n_o^{-2}(\omega_3)]\sin(2\theta)} \right| \cdot l, \tag{11}$$

$$\Delta\theta(\mathrm{II}(1)) = \pm \left| \frac{\pi/l}{\frac{\omega_3}{2c} n_e^3(\omega_3,\theta)[n_e^{-2}(\omega_3) - n_o^{-2}(\omega_3)]\sin(2\theta) - \frac{\omega_1}{2c} n_e^3(\omega_1,\theta)[n_e^{-2}(\omega_1) - n_o^{-2}(\omega_1)]\sin(2\theta)} \right| \cdot l, \tag{12}$$

$$\Delta\theta(\mathrm{II}(2)) = \pm \left| \frac{\pi/l}{\frac{\omega_3}{2c} n_e^3(\omega_3,\theta)[n_e^{-2}(\omega_3) - n_o^{-2}(\omega_3)]\sin(2\theta) - \frac{\omega_2}{2c} n_e^3(\omega_2,\theta)[n_e^{-2}(\omega_2) - n_o^{-2}(\omega_2)]\sin(2\theta)} \right| \cdot l. \tag{13}$$

As long as θ is inserted as the related ones of Fig.1 into (11), (12) and (13), the curves of the permitted angles would be obtained versus the wavelength from 227 nm to 933 nm of the ω_3, in type-I, type-II(1) and type-II(2) of CLBO in PM THG, as shown in Fig.6, also the BBO's correspondent permitted angles are presented, where we had supposed that the crystal length l was 10 mm. In the Fig.6, we could find out that the permitted angles of CLBO about any kinds PM is broader than the ones on BBO in the correspondent state in THG, especially when the type-II(2) is taken, the permitted angle $\Delta\theta$ of CLBO is as large as 65.6 mrad·mm, and that of BBO is only 4.42 mrad·mm.

B) Permitted wavelength ($\Delta\lambda$.) The permitted wavelength $\Delta\lambda$ of CLBO in THG of each PM way should be written:

$$\Delta\lambda_1(\mathrm{I} = \pm \left| \frac{\lambda_1^2/l}{6\left[\frac{dn_e(\lambda_3,\theta)}{d\lambda_3}\lambda_3 - n_e(\lambda_3,\theta)\right] - 4\left[\frac{dn_o(\lambda_2)}{d\lambda_2}\lambda_2 - n_0(\lambda_2)\right] - 2\left[\frac{dn_o(\lambda_1)}{d\lambda_1}\lambda_1 - n_o(\lambda_1)\right]} \right| \cdot l, \tag{14}$$

$$\Delta\lambda_1(\mathrm{II}(1)) = \pm \left| \frac{\lambda_1^2/l}{6\left[\frac{dn_e(\lambda_3,\theta)}{d\lambda_3}\lambda_3 - n_e(\lambda_3,\theta)\right] - 2\left[\frac{dn_e(\lambda_1,\theta)}{d\lambda_1}\lambda_1 - n_e(\lambda_1,\theta)\right] - 4\left[\frac{dn_o(\lambda_2)}{d\lambda_2}\lambda_2 - n_o(\lambda_2)\right]} \right| \cdot l, \tag{15}$$

$$\Delta\lambda_1(\mathrm{II}(2)) = \pm \left| \frac{\lambda_1^2/l}{6\left[\frac{dn_e(\lambda_3,\theta)}{d\lambda_3}\lambda_3 - n_e(\lambda_3,\theta)\right] - 4\left[\frac{dn_e(\lambda_2,\theta)}{d\lambda_2}\lambda_2 - n_e(\lambda_2,\theta)\right] - 2\left[\frac{dn_o(\lambda_1)}{d\lambda_1}\lambda_1 - n_o(\lambda_1)\right]} \right| \cdot l. \tag{16}$$

Also the crystal length l is to be 10 mm, and we could obtain the curves of CLBO's and BBO's permitted wavelength according to (14), (15) and (16) in similar way working out $\Delta\theta$, as shown in Fig.7, where x-xis is the wavelength from 227 nm to 933 nm of the ω_3 in the THG. From the Fig.7, we found that when the wavelength of ω_3 was between 330 nm and 933 nm, CLBO's permitted wavelength of type-I and type-II(2) THG, was larger than those of BBO.

It is shown in the curves from Fig.3 to Fig.7 that, in the THG of CLBO, the walk-off angle is smaller, and the permitted parameters, such as permitted angles and permitted wavelength, are larger, which helps the CLBO to generate high order harmonic generation of high-quality.

2.5. *Conversion efficiency*. When the incident wave is of the shape with plane, and the walk off effect is ignored, the THG efficiency in CLBO is:

$$\eta = \frac{\omega_3 t^2}{\omega_1 + \omega_2 t^2}\mathrm{sn}^2\left(\sqrt{\frac{1}{\omega_1 + t^2\omega_2}}\,Dz, t\right) \;\Box t < 1\Box, \tag{17}$$

$$\eta = \frac{\omega_3 t'^2}{\omega_2 + \omega_1 t'^2}\mathrm{sn}^2\left(\sqrt{\frac{1}{\omega_2 + 't^2\omega_1}}\,Dz, t'\right) \;\Box t' < 1\Box, \tag{18}$$

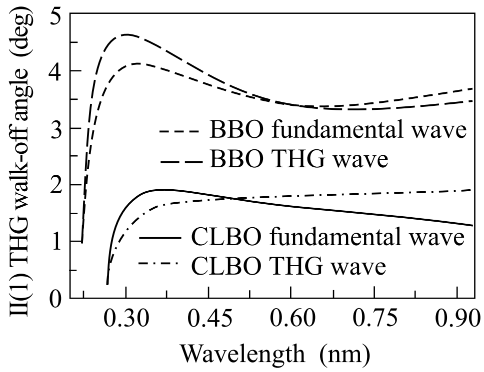

Fig.4. THG walk-off angle curves of tupe-II(1) phase matching for CLBO and BBO

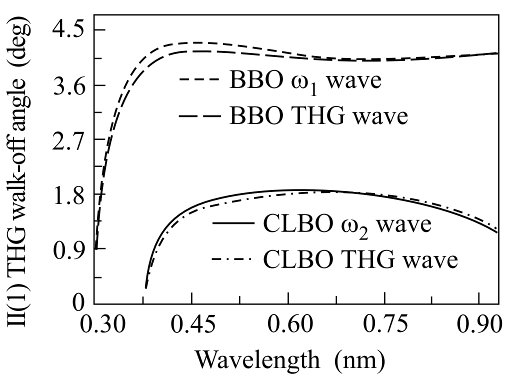

Fig.5. THG walk-off angle curves of tupe-II(2) phase-matching THG for CLBO and BBO

$$D = \left(\frac{16\pi^3 d_{\text{eff}}^2 I_s}{n_1 n_2 n_3 \lambda_1 \lambda_2 \lambda_3 \varepsilon_0}\right)^{1/2}. \tag{19}$$

Where, if it is defined that E_{20} and E_{10} are the original electricity vector, then we can obtain:

$$t = \sqrt{\frac{n_2}{2n_1}}\frac{E_{20}}{E_{10}},$$

$t' = I/t$, at the same time, $\text{sn}\,(x, y)$ is the first kind Jacobi elliptic function, I_s is the incident power intensity, and n_1, n_2, n_3 are the refractive indices of the waves with the frequency of ω_1, ω_2, and ω_3, respectively.

We have stimulated the conversion efficiency of CLBO's of type-II(1) PM in THG, when the pumped light is the fundamental wave of 1064 nm in the Q-switched Nd:YAG laser. The former CLBO crystal is for SHG of type-I, whose length is 0.7 cm, and it is tuned for $\varphi = 45°$, $\theta = 28.7°$, respectively, the latter CLBO crystal is for THG of type-II(1), and its θ and φ are 48.3° and 0°, respectively. Then we'll find out in Ref.[7] and in Fig.2, that the d_{eff} of the two crystals are to be 0.46 and 0.94 pm/v. It is supposed in the latter

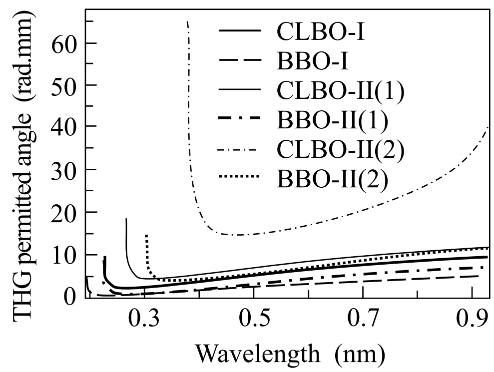

Fig.6. THG permitted angle curves for CLBO and BBO versus the wavelength of third harmonic generation

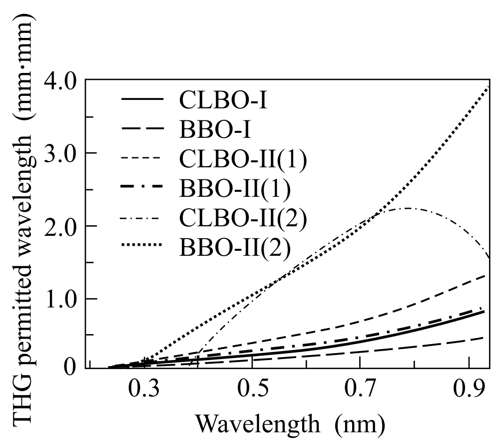

Fig.7. Permitted wavelength curves for CLBO and BBO crystals in THG

one, that one of the pumped wave, $E_1(z)$, is of the wavelength of 1064 nm, its refractive index n_1 is 1.4568, and its walk-off angle is 0.03 rad; the other, $E_2(z)$, is of the wavelength of 532 nm, and it is o light with the refractive index n_2 is 1.4981. So we calculated out that the THG, $E_3(z)$, is with the refractive index n_3 of 1.4844, and its walk-off angle is 0.028 rad. From all of the above, it is obvious that the walk-off effect in the CLBO is very smaller, and we can take it for granted that (17), (18) and (19) are suitable for THG of CLBO.

We got $D = 1.745 \cdot 10^4 \sqrt{I_s}$ according to (19), it was supposed that the beam diameter of the incident wave, and we stimulated the curves of the conversion efficiency versus the latter crystal's length, when the total pumped power P is $1 \cdot 10^8 W$□$0.7 \cdot 10^8$ W and $0.5 \cdot 10^8$ W, respectively, which is shown in Fig.8. From Fig.8, we should find when P is $1 \cdot 10^8$ W, the optimized efficiency is as high as 22% in THG of CLBO, and when the incident power intensity is increased, the efficiency of the THG would be enhanced, if provided the crystal with the appropriate length. The damage threshold of CLBO is as

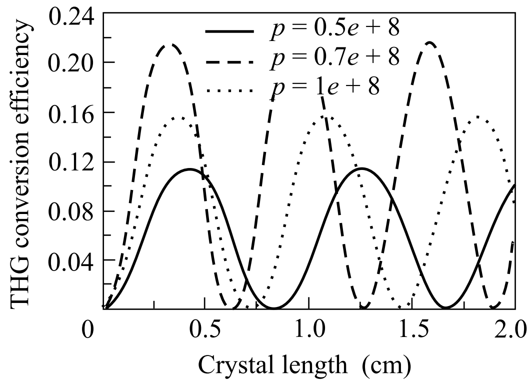

Fig.8. THG conversion efficiency of type-II(1) by different intensity pumped in PM

high as 26 GW/cm^2 in 1064 nm, and its volume can be very large, of which would be an effective way to enhance the THG efficiency in CLBO.

To the CLBO crystal, the walk-off angle is smaller, and the permitted parameter is larger, which make it easy to produce the high-quality radiation of the third or even higher order harmonic radiation in CLBO. The CLBO delivered higher THG conversion efficiency and better output stability than BBO at optimized crystal lengths When CLBO is pumped by the Q-switched Nd: YAG laser, with the beam diameter of 8 mm, and the power of $1 \cdot 10^8$ W, the THG efficiency in CLBO could be 22%. At the same time, its damage threshold is high, from which we could speculate that it can be used in the solid UV laser of high power very intensively in the future.

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