



HAL
open science

Visible and UV effective non-linear optical coefficients of β -BaB₂O₄ as function of the growth technique

Régine Maillard, Alain Maillard, Katalin Polgár

► To cite this version:

Régine Maillard, Alain Maillard, Katalin Polgár. Visible and UV effective non-linear optical coefficients of β -BaB₂O₄ as function of the growth technique. *Optical Materials*, 2009, 31 (6), pp.899-901. 10.1016/j.optmat.2008.10.043 . hal-00333078

HAL Id: hal-00333078

<https://hal.science/hal-00333078>

Submitted on 15 Aug 2022

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution - NonCommercial 4.0 International License

Visible and UV effective non-linear optical coefficients of β -BaB₂O₄ as function of the growth technique

R.S. Maillard^{a,*}, A. Maillard^a, K. Polgár^b

^aLaboratoire Matériaux Optiques Photonique et Systèmes, CNRS-UMR 7132, Université de Metz et Supelec, 2 rue E. Belin, 57070 Metz, France

^bResearch Institute for Solid State Physics and Optics of the Hungarian Academy of Sciences, Budapest 1121, Konkoly-Thege M. út. 29-33, Hungary

We report second and fourth harmonic generation measurements in β -BaB₂O₄ single crystals (β -BBO) cut in the type I phase matching direction for the conversions from 1064 to 532 nm and from 532 to 266 nm. The absolute values of the effective non-linear optical coefficients d_{eff} are determined by measuring the conversion efficiency towards the visible and the UV.

Crystals grown by the Czochralski method (directly pulled from a supercooled pure melt) are compared with crystals produced by the usual high temperature top seeded solution growth method. No difference in the effective non-linear optical coefficients was observed for the conversion from 1064 to 532 nm. In contrast, a systematically slightly higher d_{eff} value of crystal grown by the Czochralski method is obtained for the conversion from 532 to 266 nm.

1. Introduction

Beta barium metaborate β -BaB₂O₄ (β -BBO) is a well known non-linear optical material for applications in second harmonic generation of Nd:YAG laser or optical parametric oscillators. Generation of UV light with wavelength as short as 186 nm is possible [1].

Commercially available β -BBO crystals are generally produced by TSSG (Top Seeded Solution Growth). This method provides large diameter and good quality crystals but presents a very slow pulling rate (0.5 mm/day). In addition, TSSG crystals generally exhibit a Na impurity contamination due to its presence in the flux and may contain macroscopic defects, such as bubbles.

In order to reduce these drawbacks, we have grown β -BBO crystals by the Czochralski (Cz) method which is an interesting alternative to the solution growth. We use the supercooling properties of borate melt in order to reach lower growth temperatures without any solvent as Na₂O [2–4].

The purpose of this work is to compare the BBO crystals grown by the above methods in terms of their non-linear optical performance. The absolute effective non-linear optical coefficients are characterized for the conversions from 1064 to 532 nm and from 532 to 266 nm.

2. Theoretical considerations

In contrast to conventional methods which give relative values of the non-linear coefficients, in this work we have determined the effective coefficient d_{eff} in an absolute way. Our method was described in Ref. [5] and does not require a reference crystal. The specific response of the material on the focusing conditions is taken into account, what permits the direct comparison of different samples.

The technique combines the precise measurement of the conversion efficiency with the theoretical calculations of the conversion efficiency in the case of gaussian beams. Crystals cut along the phase matching direction are used.

As shown by Boyd and Kleinman [6] and by Zondy [7,8], the conversion efficiency can be expressed by

$$\Gamma_{\text{SH}} = \frac{P(2\omega)}{P(\omega)^2} = \frac{2\omega_2 d_{\text{eff}}^2 L_2}{\pi \epsilon_0 c^3 n(\omega)^2 n(2\omega) \omega_0^2} \exp(-\alpha_{2\omega} L) G(L, \omega_0, \rho, \eta, \Delta k, \alpha) \quad (1)$$

where:

- $P(\omega)$ is the fundamental power at the entrance of the crystal and $P(2\omega)$ the harmonic power generated leaving the sample.
- ω_0 is the waist of the fundamental beam.

* Corresponding author. Tel.: +33 87 37 85 44; fax: +33 87 37 85 59.
E-mail address: rmaillard@metz.supelec.fr (R.S. Maillard).

- $\alpha_{2\omega}$ and α_{ω} are the absorption constants of the harmonic and fundamental waves, respectively, and $\alpha = \alpha_{2\omega} - \alpha_{\omega}$.
- L is the length of the crystal.
- c is the velocity of the light.
- G is a factor, taking into account the relative focal position η ($0 < \eta < 1$) inside the sample, the size of the fundamental beam waist w_0 , the length of the sample L , the phase matching type, the phase mismatch Δk , the refractive indexes as well as absorption and double refraction ρ . G is obtained by integration of the intensity distribution on the transverse spatial variables.

The knowledge of the whole set of variables on the right-hand side of (1) allows the calculation of d_{eff} from the measured conversion efficiency.

3. Measurements of d_{eff} for the conversion from 1064 to 532 nm

A Nd-YAG laser ($\lambda = 1.064 \mu\text{m}$) is used as fundamental beam. In a first stage, these measurements have been performed in continuous mode with an average fundamental power of 6 W. TSSG and Cz BBO crystals cut in the type I phase matching direction ($\theta_{\text{PM}} = 22.8^\circ$), in the YZ propagation plan were used.

For a 4.5 mm length TSSG crystal, grown in the Research Institute for Solid State Physics and Optics in Budapest and a beam waist size equal to $92 \mu\text{m}$. The average value of $d_{\text{eff}1}$ was found to be equal to $1.76 \pm 0.1 \text{ pm/V}$. This value is comparable to those obtained in a previous work [5], on TSSG BBO crystals cut in the same directions with

- 4.5 mm length and beam waist size of $56 \mu\text{m}$: $d_{\text{eff}1} = 1.75 \pm 0.05 \text{ pm/V}$.
- 4.5 mm length and beam waist size of $80 \mu\text{m}$: $d_{\text{eff}1} = 1.77 \pm 0.07 \text{ pm/V}$.
- 7 mm length and beam waist size of $56 \mu\text{m}$: $d_{\text{eff}1} = 1.75 \pm 0.07 \text{ pm/V}$.
- 7 mm length and beam waist size of $80 \mu\text{m}$: $d_{\text{eff}1} = 1.78 \pm 0.06 \text{ pm/V}$.

The same measurements were performed on two Cz BBO crystals, grown in our laboratory, cut in the same direction with 4.5 and 5.1 mm length. The $d_{\text{eff}1}$ values obtained with a beam waist size of $92 \mu\text{m}$, were respectively $1.78 \pm 0.1 \text{ pm/V}$ and $1.79 \pm 0.1 \text{ pm/V}$.

These results suggest that for the generation of green light there is no significant difference between TSSG and Cz grown samples.

In a second stage, these measurements have been extended to the pulsed regime in order to check whether a higher fundamental

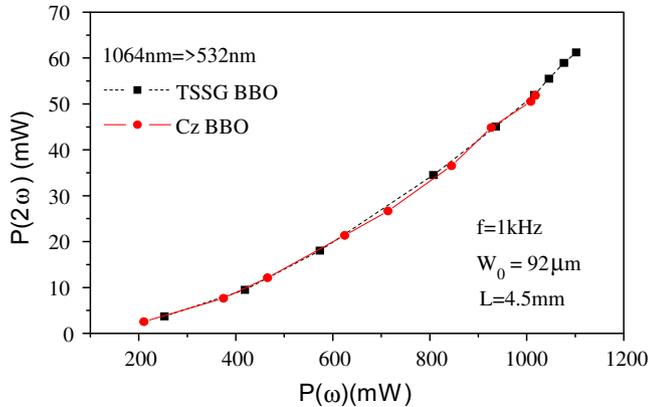


Fig. 1. Average second harmonic power (532 nm) at the exit of the crystal as function of the incident infrared power (1064 nm) for TSSG and Cz samples of 4.5 mm length.

power has an influence on these comparative results. The same procedure as in continuous mode has been used for the calculation of the effective coefficient d_{eff} , but with the $P(2\omega)$ and $P(\omega)$ terms in Eq. (1) corresponding to the peak power. These peak powers were deduced from the average power measured on a thermopile and a photodiode, knowing that the repetition rate was 1 kHz and that the half maximum widths of the pulses measured at 1064 and 532 nm were respectively found to be equal to 100 ns and 60 ns. The two TSSG BBO and Cz BBO of 4.5 mm common length were used for the pulsed measurements.

The average green power at the exit of these samples was measured as function of different fundamental powers at the entrance of the crystals. The results are reported in Fig. 1. The deduced d_{eff} values are $1.74 \pm 0.2 \text{ pm/V}$ for the TSSG sample and $1.78 \pm 0.2 \text{ pm/V}$ for the Cz sample. These values are in good agreement with those found in the CW regime. No significant difference can be observed between TSSG and Cz samples and the superposition of the two curves shows the overlap of the results for different incident powers.

It is not easy to compare these values with literature, in which a significant dispersion of the d_{eff} values can be found, due to the relative character of most measurements: a value of 2.06 pm/V is given by Becker [9] or Sasaki [10] by referring to Chen [11] measurements then Nikogoshian [12] and Eckart [13] indicate respectively d_{eff} values of 1.59 pm/V and 1.94 pm/V .

4. Measurements of d_{eff} for the conversion from 532 to 266 nm

We obtain the fourth harmonic of the Nd-YAG laser ($\lambda = 266 \text{ nm}$) by performing two successive steps of second harmonic generation. BBO crystals cut in the type I phase matching direction were used with $\theta_{\text{PM}} = 22.8$ and $\theta_{\text{PM}} = 47.6$ for the first conversion from 1064 to 532 nm and for the second conversion from 532 to 266 nm, respectively. These measurements were performed only in the pulsed regime because the green power obtained in continuous mode after the first conversion is too low to generate detectable ultraviolet light.

As for the preceding conversion, TSSG and Cz BBO crystals were compared. Two TSSG BBO and Cz BBO have been cut with length of 3 mm in the beam propagating direction. The beam waist size of the 532 nm pump beam was $60 \mu\text{m}$, the half maximum width of the pulse measured at 532 nm and 266 nm was found to be equal to 60 ns and 40 ns, respectively, and the repetition rate was 1 kHz.

As described before, the average UV powers were measured at the exit of the samples as a function of the green power at the entrance. The results are reported in Fig. 2.

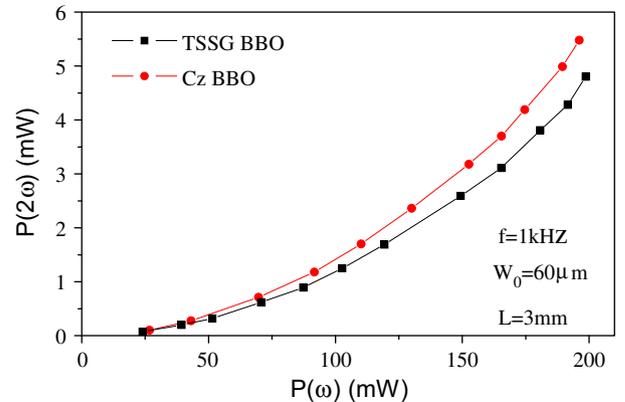


Fig. 2. Average fourth harmonic power (266 nm) at the exit of the crystal as function of the incident green power (532 nm) for the TSSG and the Cz samples of 3 mm length.

In contrast to what is observed for the conversion from 1064 to 532 nm, the two curves obtained in the Fig. 2 are not overlapping and a systematic increase of the generated UV power can be noticed for the Cz sample. The deduced d_{eff} values are 1.28 pm/V for the TSSG sample and 1.38 pm/V for the Cz sample.

Same measurements performed on two Cz BBO and TSSG BBO of 4.5 mm length lead to the same conclusions and give comparable values of d_{eff} with 1.27 pm/V for the TSSG sample and 1.36 pm/V for the Cz sample.

Even if this increase of d_{eff} with the Cz growth method is weak, its systematic character for various incident powers and different crystals lengths shows that it exists indeed.

These values are comparable with the value of 1.32 pm/V given by Sasaki [10] for this conversion from visible to UV.

It is also possible to calculate the expected d_{eff} value in the following way. The d_{eff} development as function of the non-linear coefficients for type I conversion and BBO 3 m symmetry is given by

$$d_{\text{eff}} = d_{31} \sin \theta - d_{22} \cos \theta \sin 3\phi \quad (2)$$

where θ is the angle between the wave vector and the Z optical axis and ϕ is the angle between the projection of the wave vector in the XY plane and the X-axis. In a previous work [5], the d_{22} and d_{31} non-linear coefficients of TSSG BBO crystals was found to be respectively equal to 2.11 ± 0.1 pm/V and 0.26 ± 0.05 pm/V with an opposite sign of these two coefficients. These measurements were performed for the conversion from 1064 to 532 nm. The crystal being cut in the type I phase matching direction ($\theta = \theta_{\text{PM}} = 22.8^\circ$) in the YZ propagation plan ($\phi = 90^\circ$), the calculated value for the conversion from 1064 to 532 nm were $d_{\text{eff}} = 1.84 \pm 0.1$ pm/V. Thus, for the conversion from 532 to 266 nm, we can use these d_{22} and d_{31} values to calculate the expected d_{eff} value, with the assumption that the dispersion of the d_{ij} values is not significant in this wavelengths range. With $\theta = \theta_{\text{PM}} = 47.6^\circ$ and $\phi = 90^\circ$, the calculated value for the conversion from 532 to 266 nm is $d_{\text{eff}} = 1.24 \pm 0.05$ pm/V which is in good agreement with the d_{eff} values measured for the TSSG grown crystals.

5. Conclusion

Comparative non-linear effective coefficients measurements has been performed on BBO crystals grown by the Czochralski method and on BBO crystals obtained by top seeded solution growth. For the conversion from 1064 to 532 nm, no significant difference in the values of d_{eff} is observed for the two types of crystal. For the conversion from 532 to 266 nm, the d_{eff} of the Cz BBO is found to be slightly higher than the one measured for the TSSG BBO.

The observed increase of the UV conversion efficiency is probably connected to a higher purity and/or the smaller number of scattering centres of the samples grown by the Czochralski technique. The fact that such a phenomenon was not observed for the green conversion efficiency confirms the assumption because impurities and light scattering centres have a more important effect on the effective absorption in UV than in the visible range. These results are in good agreement with comparative measurements of scattered light in Cz BBO and flux-grown BBO before and after annealing performed by Kouta and Kuwano [14]: Scattering centers are shown to be due to structural defects in Cz BBO and to impurities in flux BBO.

Non-linear absorption measurements in the visible [15] and UV [16] range have been performed on TSSG BBO but no comparison with Cz BBO are available. Complementary non-linear absorption measurements in UV range should be performed.

In conclusion, the Czochralski growth technique allows to obtain good quality β -BBO crystals with a pulling rate 24 times higher and a conversion efficiency in UV range which is slightly higher than the one obtained for crystal grown by TSSG method.

Acknowledgements

This work was supported by "Groupe de Recherche GDR CNRS 1148 Matériaux Laser LASMAT-CMDO+", the Hungarian Scientific Research Fund (OTKA Nos. T-046481 and K-60086), the Centre of Excellence Program (Contract No. ICA1-1999-75002).

References

- [1] H. Kouta, Y. Kuwano, *Optics Lett.* 24 (1999) 1230.
- [2] K.L. Ovanesyan, A.G. Petrosyan, G.O. Shirinyan, *Cryst. Res. Technol.* 24 (1989) 859.
- [3] K. Itoh, F. Marumo, Y. Kuwano, *J. Cryst. Growth* 106 (1990) 728.
- [4] H. Kouta, Y. Kuwano, K. Ito, F. Marumo, *J. Cryst. Growth* 114 (1991) 676.
- [5] R.S. Klein, G.E. Kugel, A. Maillard, A. Sifi, K. Polgar, *Opt. Mater.* 22 (2003) 163.
- [6] G.D. Boyd, D.A. Kleinman, *J. Appl. Phys.* 39 (1968) 3597.
- [7] J.J. Zondy, *Opt. Commun.* 81 (1991) 427.
- [8] J.J. Zondy, M. Abed, A. Clairon, *J. Opt. Soc. Am. B* 11 (1994) 2004.
- [9] P. Becker, *Adv. Mater.* 10 (1998) 979.
- [10] T. Sasaki, Y. Mori, M. Yoshimura, Y.K. Yap, T. Kamimur, *Mater. Sci. Eng.* 30 (2000) 1.
- [11] C. Chen, B. Wu, A. Jiang, G. You, *Sci. Sinica (Ser. B)* 28 (1985) 235.
- [12] D.N. Nikogosyan, *Appl. Phys. A* 52 (1991) 359.
- [13] R.C. Eckardt, H. Masuda, Y.X. Fan, R.L. Byer, *IEEE J. Quantum Electron.* 26 (1990) 922.
- [14] H. Kouta, Y. Kuwano, *Appl. Opt.* 38 (1999) 1053.
- [15] H. Li, F. Zhou, X. Zhang, W. Ji, *Opt. Commun.* 144 (1997) 75.
- [16] L.I. Isaenko, A. Dragomir, J.G. McInervey, D.N. Nikogosyan, *Opt. Commun.* 198 (2001) 433.