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Stephan P. Velsko
David Eimerl

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NONLINEAR MATERIALS FOR FREQUENCY CONVERSION*

Stephan P. Velsko and David Eimerl
Nonlinear Optical Materials Group
Lawrence Livermore National Laboratory
Livermore, CA 94550

ABSTRACT

Two figures of merit, the threshold power (P_{th}) and the limiting volume (V_{min}) can be used to compare the relative efficiency and economy of new harmonic generating crystals. The properties of barium metaborate and L-Arginine phosphate are used to illustrate the effect of nonlinearity, birefringence, and damage threshold on these figures of merit.

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INTRODUCTION

High efficiency harmonic generators are crucial components of modern laser technology. Advances in both basic science (e.g. spectroscopy), and applications (e.g. medical lasers) can depend on extending the number of useful wavelengths of laboratory lasers through frequency conversion. The success of certain strategic technologies, such as laser driven inertial confinement fusion (ICF), may depend on the availability of new, high performance nonlinear materials. New materials can also significantly alter the available markets for particular commercial lasers.

The way in which a frequency conversion material is selected for a particular application is still primarily determined by the limited number of commercially available harmonic generating crystals and their cost. However, as the number of available new materials for frequency conversion increases, it will become important to have sharper criteria for comparing different materials for a given application. Two such figures of merit which have recently been suggested are the threshold power (P_{th})[1] and the limiting volume (V_{min}).[2]

The threshold power[1] is the minimum power a diffraction limited beam must have for efficient (>50%) conversion to the second harmonic by a given crystal. This quantity is given by the expression

$$P_{th} = \left(\frac{\lambda \beta}{C}\right)^2$$

The parameter β is the angular sensitivity, $\partial \Delta k/\partial \theta$. Alternatively, the walkoff angle of a critically phasematched doubler may be substituted for the $\lambda\beta$ product. The C coefficient is the effective nonlinear coupling, expressed in units of power$^{-1/2}$. For efficient conversion, the laser peak power must exceed the threshold power of the chosen nonlinear material.

An important conclusion of the analysis leading to expression (1) is that the beam aperture and crystal length can always be scaled to achieve the same conversion efficiency at any incident intensity, given a particular power.[1] In this way, deleterious effects such as stimulated Raman scattering or optical damage can always be avoided without sacrificing conversion efficiency, but with a necessary increase in crystal volume.

The concept of aperture/length scaling leads naturally to the second figure of merit, the limiting volume[2]. This is given by the product of the SHG gain length and the minimum device aperture allowed by intensity limiting processes such as optical damage:

$$V_{min} = \frac{P}{C\ I_{max}^{3/2}}$$

This figure of merit is an unambiguous way to distinguish two materials with similar threshold powers. It is clearly related to the economic impact of a given choice of materials, given equal efficiencies.

In this paper we will compare two recently discovered nonlinear crystals, barium metaborate (BBO)[3] and L-Arginine phosphate (LAP),[4] to illustrate the influence of nonlinearity, angular sensitivity, and damage threshold on these figures of merit. We will also compare these crystals to the older materials KTP and KDP.

BARIUM METABORATE

We have recently completed a thorough evaluation of BBO for high average power frequency conversion.[5] The salient properties of this material are its large birefringence and low dispersion, which lead to extremely short noncritical phasematch wavelengths. For example, the noncritical wavelength for type I frequency doubling is 410 nm,[5,6] which makes this crystal extremely useful for generating ultraviolet light. However, this same property leads to very narrow angular bandwidths (or large walkoff angles) for most harmonic generating processes in the near infrared and visible. Table 1 gives the angular sensitivity values for doubling and tripling the Nd:YAG fundamental at 1.064 μm. For comparison, types I and II doubling in KDP have angular sensitivities of 4900 and 2500 cm^{-1}/rad, respectively, 2 to 3 times smaller. Thus, although the nonlinear coefficient of KDP is smaller than that of BBO for these processes,[3] a larger length of crystal can be used. A perhaps surprising result of this is that the threshold power of BBO at 1.064 μm is not significantly different than that of KDP, as illustrated in Fig. 1.

L-ARGININE PHOSPHATE

L-Arginine phosphate (LAP) is a new, efficient organic crystal which can be grown in large sizes. We have recently characterized its properties to evaluate its potential as a replacement for KDP in the next generation of large aperture, high power lasers for ICF.[7]
Unlike BBO, which is inorganic and uniaxial, LAP is a biaxial organic

salt. Extensive hydrogen bonding in this crystal leads to significant optical absorption in the near infrared. However, deuteration can reduce the losses at 1.06 μm to less than 1% cm^{-1}. We have fully characterized the dispersion and thermo-optic coefficients as well as the nonlinear coefficients of this crystal, and have evaluated the phasematching properties. Figure 2 shows the loci for type I and II phasematching for 1.06 μm as well as the variation in effective nonlinear coupling along each locus. Table 2 gives the values of nonlinear coupling, angular sensitivity and threshold power for various processes. For type II doubling the threshold power is nearly three times smaller than that of KDP. This is a result of a significantly larger nonlinear coupling and only modest increase in angular sensitivity.

DAMAGE THRESHOLDS AND LIMITING VOLUMES

For lasers with very high peak powers, the threshold power criterion for efficient conversion will be satisfied by many crystals, and therefore 1 becomes a less discriminating figure of merit. The question then turns to how small a volume of a given material is necessary to assure a robust and efficient device. The most common effect which limits the aperture size is the damage threshold. Table 3 shows the results of damage testing at LLNL for several nonlinear crystals. The damage threshold for LAP is comparable to that of high temperature oxides such as BBO or KTP, and more than twice that of commercially available KDP. The combination of high nonlinearity (short crystal length) and high damage threshold (small aperture size) can lead to a significant reduction in device volume. Table 4 shows the relative volumes of KDP, BBO, KTP and LAP based on the damage thresholds and nonlinear coupling values for type II doubling of 1.06 μm. Note that for this process, KTP has a significant advantage over the others both in threshold power and device volume. A small, optimized device volume may, of course, offset the cost per unit volume of a given material and may also lead to reductions in the total cost of a laser system where size and weight are important factors. However, we currently believe that LAP, which can be grown in large sizes from aqueous solution, and has a significantly lower threshold power and limiting volume than KDP, is the material of choice for large aperture ICF lasers.

CONCLUDING REMARKS

We have evaluated BBO and LAP for frequency conversion using two figures of merit derived from a consideration of scaling in high efficiency harmonic generation. Both BBO and LAP are high damage threshold materials with significantly smaller limiting volumes than KDP. BBO is capable of harmonic generation at very short wavelengths, but the narrow angular bandwidth offsets its large nonlinear coupling for doubling or mixing near infrared wavelengths. The doubling efficiency of LAP in the near infrared exceeds that of KDP and BBO. Like KDP, it can be grown in large sizes at moderate cost.

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FIGURE CAPTIONS

1. Threshold power for Type I and II doubling vs. wavelength for BBO and KDP.
2. Phasematching loci for frequency doubling of 1.06 μm in LAP. Open circles denote positions where $d_{eff} = 0$, solid circles mark local maxima.

Table 1. Phasematching angles and angular sensitivities of BBO for Nd:YAG harmonics.

Process (1.06 μm)	Angle	Angular Sensitivity (cm^{-1}/mrad)
2ω I	22.7°	10.7
2ω II	32.4	7.61
3ω I	31.1	21.7
3ω II	38.4	16.4
3ω III	58.4	7.0
4ω I	47.3	34.0
4ω II	81.0	7.4

Table 2. Nonlinear coupling, angular sensitivity and threshold power for generating Nd:YAG harmonics in d-LAP.

Process	d_{eff}*	ß (cm^{-1}/mrad)	P_{th} (MW)
2ω I	2.57	5.3	100
2ω II	2.35	3.7	26
3ω I	2.51	15.3	190
3ω II	2.24	8.5	87
4ω I	2.10	-	-

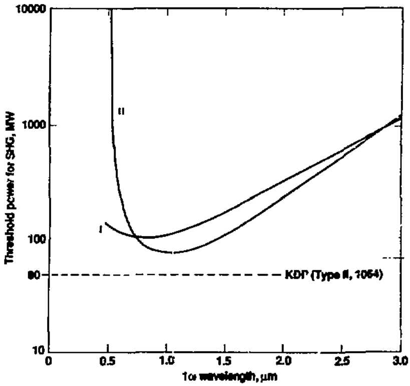
*Relative to d_{36} (KDP)

Table 3. Damage fluences for 1 ns pulses at 1.064 μm.

Material	Available Volume (cm^3)	Source	J_D (Joules/cm^2)
KDP	10x10x10	varies	5 ± 1
LAP	8x7x2	Phillips Cleveland Crystals	13 ± 1
BBO	2x2x0.5	Fujian (Chen)	13 ± 2
KTP	0.5x0.5x0.5	Phillips Beijing	15 ± 2

Table 4. Threshold powers and limiting volumes for type II doubling of 1 ns pulses at 1.064 μm.

Material	P_{th} (MW)	Relative Volume ($I_{max} = J_D/\tau$)
KDP	70	1
KTP	0.5	0.02
BBO	80	0.07
LAP	26	0.13



10000
1000
100
80
10
Threshold power for SHG, MW
II
I
KDP (Type II, 1064)
0
0.5
1.0
1.5
2.0
2.5
3.0
1ω wavelength, μm

Figure 1

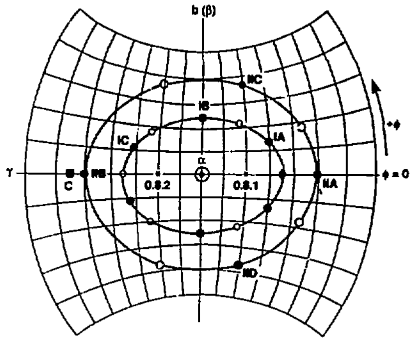


Figure 2