High efficient harmonic generation in two BBO crystals by means of walk-off compensation

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We report on a highly efficient frequency-doubling set-up of two BBO crystals in which the second crystal compensates the walk-off in the first. This yields a maximum conversion efficiency of 50% from 527nm to 263nm.

Introduction

An efficiency-limiting factor in second harmonic generation in critically phase-matched crystals is the occurence of walk-off between the fundamental and second harmonic beam [1]. It is possible to compensate for the walk-off, by using a second crystal in which the walk-off is in the opposite direction. Such a method has been used to enhance the performance of optical parametric oscillators [2]. We will show that it can also be used to increase the efficiency of second harmonic generation processes.

This contribution will start with some physics of birefringent crystals and the origin of walk-off. Then the walk-off compensation scheme will be discussed, and it will be applied to a frequency doubling experiment, in which the fourth harmonic of a Nd:YLF laser system is produced.

Walk-off in birefringent crystals

The walk-off effect was already described by Christiaan Huygens in 1690, who observed a beam of sunlight passing through what he called Islandic crystal ($CaCO_3$) [4]. Even though the incident beam was perpendicular to the crystal surface, Huygens observed that it split in two beams that propagated at an angle of 6.6°. One beam was called the ordinary ray, and the other was called the extraordinary ray since it did not seem to obey Snell's law of refraction.

Many optical crystals exhibit birefringence. The physical origin of this effect is the anisotropy of the crystal, that expresses itself in the direction dependence of the dielectric constant. This results in a different refractive index for the two directions of polarization. Moreover, the refractive index depends on the propagation direction through the crystal.

The ordinary beam is the optical beam with its electric field perpendicular to the plane formed by the propagation direction and the optical axis. The extraordinary beam has its electric field in the same plane as the propagation vector and the optical axis. If an optical beam propagates along the optical axis Z, the extraordinary and ordinary beam are interchangeable (see figure 1).

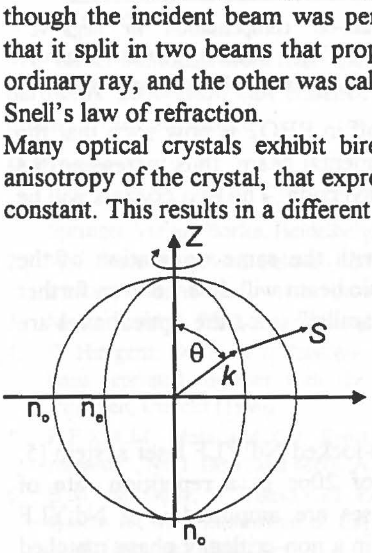

Figure 1: Walk-off in a negative uniaxial crystal.

Due to the direction dependence of the dielectric constant, the electric displacement ***D*** is in general not in the same direction as the electric field *E*. As a result, the wavevector $k = D \times H$ will usually not be in the same direction as the Poynting vector $S = E \times H$. In words, the energy of the beam propagates at an angle to the propagation direction of the wavefront. This effect is called walk-off. It can be shown [3] that the Poynting vector *S* is always perpendicular to the surface of the index ellipsoid, at the point where it is intersected by the wave vector. Figure 1 gives an example of walk-off in a negative uniaxial crystal. The walk-off angle depends on the propagation direction of the wavefront. In negative uniaxial crystals, the walk-off is directed away from the optical axis; in positive uniaxial crystals it is directed towards the optical axis.

Walk-off compensation

The birefringence of non-linear optical crystals is used for phase-matching in second harmonic generation. In long crystals the walk-off between the fundamental and the harmonic beam will limit the conversion efficiency. For instance, in a β-barium borate (BBO) crystal cut for frequency-doubling at 527 nm, the angle between the Poynting vectors of the second harmonic and the fundamental is 4.9°. In a crystal with a length of 7 mm, the fundamental and second harmonic beams will diverge a distance of 0.6 mm over the length of the crystal.

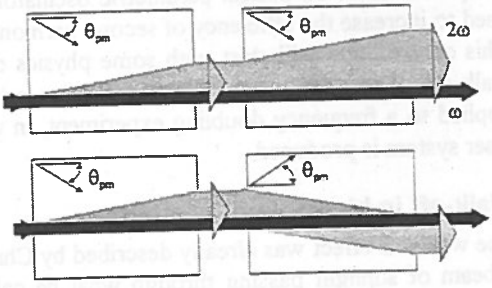

Figure 2: Walk-off compensation in negative uniaxial crystals. Top: parallel crystals; below: crossed crystals. θ_{pm} is the phase matching angle.

Although the Poynting vectors of ordinary and extraordinary rays are not parallel, the wave vectors of the fundamental and second harmonic beams are. Thus the beams, having passed through the non-linear optical crystal, will propagate parallel in free space. This allows for the possibility of walk-off compensation (see figure 2). We can position a second BBO crystal after the first one, which is rotated 180° on the propagation axis with respect to the first crystal. The first crystal will be referred to as BBO1, the second as BBO2. The walk-off in BBO2 is now such that the second harmonic beam moves back towards the fundamental beam, thus increasing the spatial overlap of the beams and the efficiency of the conversion. The two crystals will be called "crossed" since the optical axes are not parallel.

If BBO2 is aligned in the same way as BBO1, i.e. with the same orientation of the principle axes, then the fundamental and second harmonic beam will diverge even further in the second crystal. The two crystals will be called "parallel" since the optical axes are parallel.

Experimental results

In our set-up, we create the fourth harmonic of a mode-locked Nd:YLF laser system [5, 6]. This system emits laser pulses with a duration of 20ps at a repetition rate of 81.25MHz and a wavelength of 1053nm. These pulses are amplified in a Nd:YLF amplifier chain, after which they are frequency-doubled in a non-critically phase matched $5 \times 5 \times 18$ mm lithium triborate (LBO) crystal, with an efficiency of 50%. This second harmonic is frequency-doubled in two BBO crystals, to 263nm.

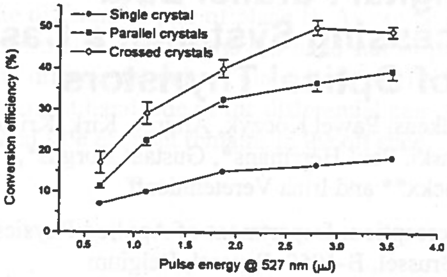

Figure 3: Efficiency of the sum-frequency process for three different BBO configurations.

The efficiency of the fourth harmonic generation process as a function of the pulse energy in the visible is given in figure 3. A conversion efficiency of 20% from the second to the fourth harmonic has been obtained, using a single BBO crystal. Two parallel BBO crystals yield a maximum conversion efficiency of 40%. When the BBO crystals are crossed, the maximum conversion efficiency is 50%, which is more than twice the efficiency obtained with a single crystal.

An additional bonus of the walk-off compensation scheme is an improved beam quality in the ultraviolet. Figure 4 shows the beam profiles of the fourth harmonic beam in the far field. The left figure shows the profile when only one single BBO crystal is used in the conversion. When two crystals in parallel are used, the beam profile consists of two spots that overlap partially, as one would expect from figure 2. When the BBO crystals are crossed, the resulting beam profile is much better than was obtained with two parallel crystals.

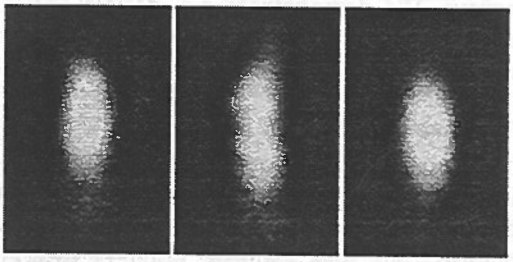

Figure 4: Beam profiles corresponding to three BBO configurations. Left: single BBO, centre: parallel BBOs, right: crossed BBOs.

Conclusions

We have demonstrated that the use of a walk-off compensation scheme in second harmonic generation can enhance the efficiency of the process, and improve the beam quality of the harmonic.

References

1. V.G. Dmitriev, G.G. Gurzadyan, D.N. Nikogosyan, *Handbook of non-linear optical crystals*, Springer Verlag, Berlin, Heidelberg, New York (1991).
2. W.R. Bosenberg, W.S. Pelouch and C.L. Tang, "High-efficiency and narrow-linewidth operation of a two-crystal β-BaB_2O_4 optical parametric oscillator", Appl. Phys. Lett. 55, 19 (1989) p1952 –1954.
3. M. Born and E. Wolf, *Principles of Optics*, Pergamon Press, Oxford (1980).
4. C. Huygens, *Traité de la Lumière* , Van der Aa, Leiden (1690). The original French version has been reprinted together with the Dutch translation, as *Verhandeling over het licht*, Epsilon Uitgaven, Utrecht (1990).
5. R.F.X.A.M. Mols and G.J. Ernst, "Performance of a MOPA laser system for photo cathode research", Nucl. Instr. and Meth. A. 341 (1994) p481 – 483.
6. B.M. van Oerle, D. Bisero, G.J. Ernst, J.W.J. Verschuur and W.J. Witteman, "A Nd: YLF laser system for the illumination of Cs_2Te photo cathodes in the TEU-FEL accelerator", submitted to Nucl. Instr. and Meth. A.