High efficiency Second Harmonic Generation

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Introduction :

Visible ultrashort pulses have numerous uses in spectroscopy [1], non-linear microscopy [2] or micro-machining [3] for instance. However good broadband laser gain media are not available for the visible range. Thus frequency doubling of near infrared pulses is often used to produce femtosecond visible pulses. It is also often used to generate the pump for parametrical amplification schemes [4,5] that have several applications in biophotonics [6,7,8]

Some of the parameters that control the second harmonic generation (SHG) process are well known [9,10], such as the pulse energy, the crystal non-linear properties, the pulse duration or the focusing conditions. The importance of spectral phase was predicted [11] and observed for the non-linear crystals BBO and LBO. In our work we studied more thoroughly the effect of spectral phase on SHG efficiency, but also on beam quality, through modelling.

Results :

I modelled frequency doubling of a 1030 nm ultrashort (300 fs transform limited duration) radiation in the nonlinear crystal LBO. I used the full Sellmeier's equations [12] to model propagation, and the nonlinear susceptibility for the SHG [13]. Modelling is performed using a split-step fourier method [14]. I chose a 4.8mm crystal, which is the group velocity mismatch length between the second harmonic and fundamental. The beam diameter was set at 300um, corresponding to a Rayleigh length of 110 mm. Thus diffraction should be neglectable (though it is fully modelled in the program)

I modelled the dependence of SH conversion efficiency on group delay dispersion (fig.1) at different peak fundamental intensities. At all intensities peak efficiency is achieved at GDD close to 0 fs^2, corresponding to a transform limited pulse. One of the effects of GDD is to stretch the pulse. The dashed curves correspond to the effect of pulse stretching, when depletion of the fundamental and phase effects are neglected. We observe a shift of behavior between the low intensity (and efficiency) cases and the high intensity cases. At low intensity pulse stretching accounts relatively well for the drop in efficiency. The actual curve is slightly flatter because depletion of the fundamental reduces efficiency near the peak. But when intensity is higher, the behavior drastically changes. Efficiency rapidly falls when GDD is introduced. However, much higher intensity can be achieved when GDD = 0 fs^2. This surprisingly high sensitivity to GDD is limiting for practical applications. Before the compensator typical values of GDD in a femtosecond laser are of the order of $30ps^2$. A change of 0,01% would correspond to a 12% efficiency drop at 45 GW/cm^2, which would be prohibitive.

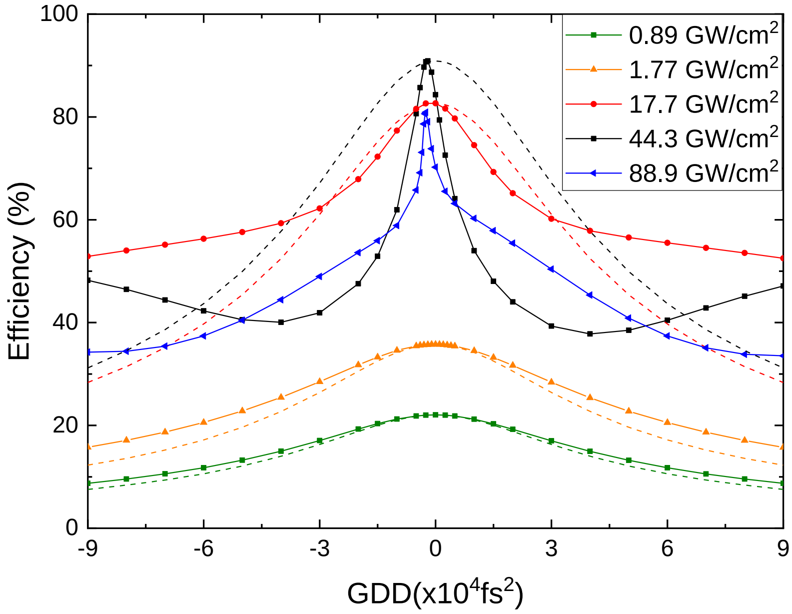

Fig. 1. Numerical simulation results of conversion efficiency from fundamental to SH as a function of GDD applied to fundamental pulses for different values of the fundamental intensity (solid curves) and a relative efficiency using a simple model disregarding depletion of fundamental and phase effects (dashed curves), normalized to the peak efficiency from numerical simulation.

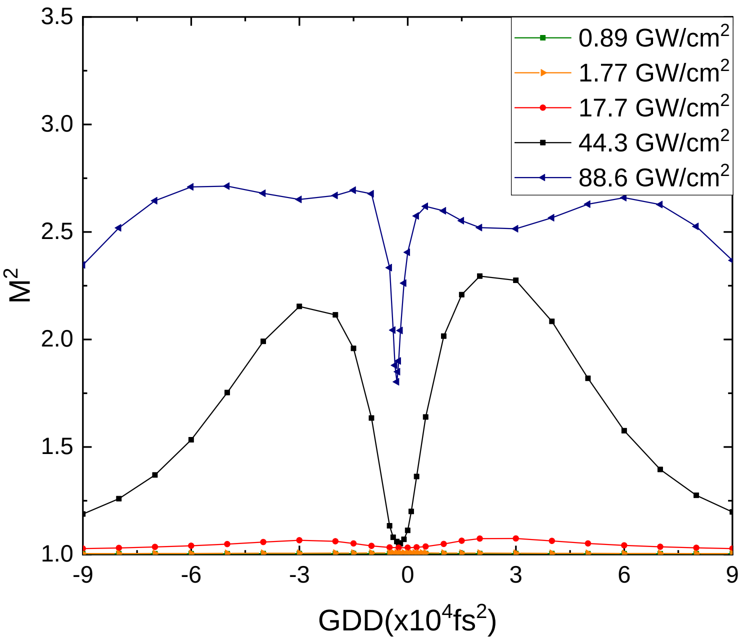

Fig. 2. Beam quality parameter M^2 of the SH beam as a function of GDD applied to the fundamental pulse for different values of the intensity of the fundamental beam.

I also investigated the dependence of the beam quality parameter M^2 for the SH beam on GDD (Fig. 2). Once again the behaviour of M^2 is quite surprising. At low intensity beam quality remains almost ideal (close to 1) for all values of GDD. Even at higher intensity beam quality remains good for transform limited pulses (GDD= 0 fs^2). Only for intensities of ~90GW/cm^2 does it significantly worsen. But at high intensity beam quality is highly sensitive to GDD and quickly worsens when small amounts of GDD are introduced. M^2 reaches peaks for values of GDD corresponding to minima of efficiency (here ±30 000 fs^2) . The higher the intensity, the higher the peaks. The curves mirror each other.

The evolution of SH conversion efficiency and SH beam quality are accounted for by back conversion from the SH to the fundamental. Because back conversion depends on intensity, I has a strong adverse effect on beam quality (and obviously reduces efficiency because the SH gets depleted). When GDD is introduced we observe back conversion starts earlier in the crystal, while when GDD= 0 fs^2 we observe no back conversion up to the intensity of 90 GW/cm^2. At this highest intensity we can also notice a change in the dependence on GDD of both efficiency and beam quality. This is because when relatively large amounts of GDD are introduced (more than 10 000 fs^2) there is a full cycle. Early in the crystal there is conversion to the SH, then back conversion to the fundamental, and further on conversion restarts !

Conclusion :

Good control of the spectral phase (including GDD, but also higher order dispersion) is is a limiting factor for both SH beam quality and SH conversion efficiency. While in practice it is difficult to achieve efficiency of more than 65% and keep good beam quality [9, 10, 12, 13], we showed that in theory efficiency as high as 90% or more could be achieved for transform limited pulses. The effect of spectral phase needs to be carefully considered when choosing the crystal type, its length and the focusing conditions.

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