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Efficient self phase matched third harmonic generation of ultrashort pulses in a material with positive dispersion

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Abstract. An ultrafast transient refractive index grating, produced in barium fluoride, a material with positive dispersion, promotes very efficient third harmonic generation. The significant enhancement of the generation up to a conversion efficiency of about 3% is due to self phase matching, involving the instantaneous grating. At the same time several diffraction orders of the third harmonic signal are observed behind the sample.

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Third harmonic generation (THG), particularly in gases, has been studied intensively during the last few decades and presents a proven way to generate coherent UV radiation [1, 2]. The conversion efficiency, however, is low and limited by the condition that the fundamental and the TH propagate in phase through the medium [3]. This phase matching implies that the difference between the phase velocities of the fundamental and generated waves, due to the dispersion of the material, is compensated. Phase matching for THG has been achieved, for instance, through the birefringence of some crystals [4] or through the mixture of materials with different dispersive behaviour [5].

Shelton and Shen [3] demonstrated phase matching in a cholesteric liquid crystal, exploiting the spatial variation of the dielectric constant in such materials. They described this process, in analogy to electrons propagating in a periodic lattice, as a coherent optical umklapp process, or nonlinear Bragg reflection [6]. In this case the phase mismatch is compensated by the grating or umklapp period, formed as a result of the helical structure of the material. Recently, Kapteyn, Murnane and collaborators [7–9] obtained efficient THG of sub-ps pulses in a gas contained in a hollow fiber. They achieved phase matching by adjusting the gas pressure such that its dispersion compensated the modal dispersion of the fiber.

All these techniques are very sensitive to the variation of the input parameters. Nonlinear phase mismatch or spatial self-interaction of the fundamental fields can destroy the phase matching. On the other hand Glushko et al. [10] demonstrated self phase matching in atomic vapours using focused beams with curved wavefronts and therefore a wide distribution of k directions [11]. They achieved a conversion efficiency of $\approx 1.5\%$ independent of the input parameters. However, they had to use a material with negative dispersion, a situation which is only usually met for frequencies near electronic resonances.

Here, we report for the first time on a new mechanism of self phase matched direct THG with a conversion efficiency of $\approx 3\%$ in barium fluoride, a material with positive dispersion. We assume that this high conversion efficiency is a result of phase compensation by coupling to a transient Kerr-index grating, induced by the interference of two non-collinear femtosecond laser pulses, and propagation through the sample together with these pulses [12]. Also, this grating leads to a diffraction of the generated TH light [13].

1 Experimental principle and set-up

If in a conventional non-collinear pump–probe arrangement [14], pump and probe pulses overlap in time during their propagation in the material, they interfere with each other, resulting in a periodic intensity pattern. For high optical intensities – as in our case – the refractive index of the material becomes intensity dependent due to the optical Kerr effect [15]. Consequently, the intensity pattern is translated into a corresponding refractive index modulation, representing a transmission grating [16]. Since in a widely transparent material such as barium fluoride the photon energies involved (1.53 eV, 4.6 eV) are far from resonance (band gap 10 eV), the origin of the nonlinearity is a mere distortion of the electron orbits, with a response time of the order of one optical cycle (≈ 1 fs). Therefore, the grating formation is an instantaneous response of the material to the interference pattern of

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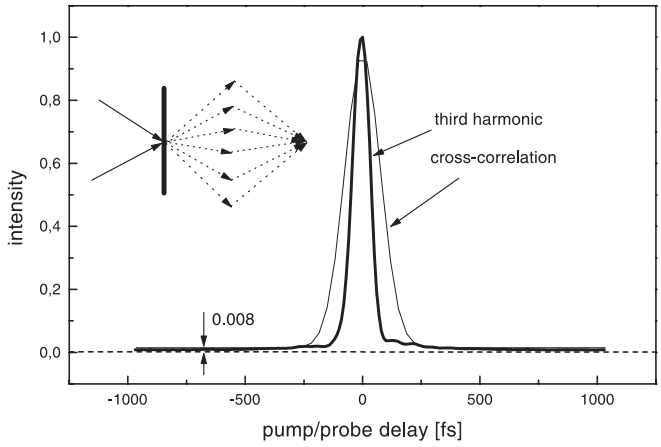


Fig. 1. Dynamical behaviour of the TH for small angles. The diffracted orders were collimated with a quartz lens. Note that for the temporal coincidence of pump and probe beams the THG is enhanced ≈ 125 times as compared to the sum of single beam THG from pump and probe outside the coincidence range

the beams. As a result, the refractive index grating is a thin slide with the thickness of the spatial pulse extension, propagating through the material synchronously with the excitation pulses.

Our detailed experimental set-up has been described in a previous article [12]. In short, we used an amplified titanium-sapphire laser system (wavelength 810 nm, pulse duration < 100 fs, repetition rate 1 kHz) as the fundamental source. The output beam is split into two s-polarized beams (pump and probe) by means of an appropriate 50/50% beamsplitter. The probe beam can be optically delayed to allow exact time overlap, achieved by slightly focussing the two beams onto a cleaved barium fluoride crystal (thickness ≈ 1 mm). The angle between the two beams was set to 1.2°, while the total incident intensity of $\approx 1 \times 10^{11}$ W/cm^2 was always below the threshold for white light continuum generation of $\approx 1 \times 10^{12}$ W/cm^2 [17]. For the measurement of the dynamic behaviour of the THG (as shown in Fig. 1), we collimated the different orders with a quartz lens and separated the TH from the fundamental with a Pellin Broca prism. Then, we varied the path length of the probe pulse with regard to that for the pump pulse and recorded the TH signal as a function of the resulting delay between the two pulses.

2 Results and discussion

As we reported earlier, the travelling grating leads to very efficient self diffraction of the fundamental input beams [12]. However, at the same time, strong TH radiation is generated which is so intense that the resulting fluorescence from a white paper screen can be observed with the naked eye [13]. Spots of blue light are clearly visible between the transmitted pump and probe spots and between the diffracted orders of the fundamental beam spots. Further TH beams are observed collinear to the transmitted fundamental beams and their higher order diffraction.

It is shown below that the observed TH pattern emerges in the following way: due to the transient index grating, two strong self-phase matched TH beams are generated emerging between the two zero order fundamental beams (central blue spots). From the relative intensities of the different TH spots in the observed pattern, we can derive that these are higher diffracted orders of the two central TH beams. This is different to a cascaded process involving higher order diffracted fundamental beams, similar to a recent report by Crespo et al. [18].

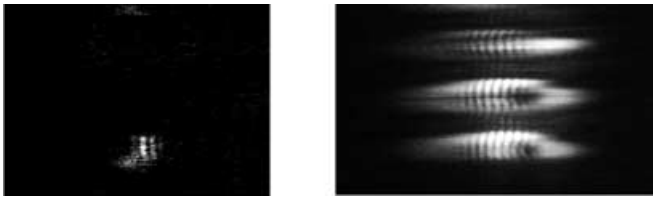
Figure 1 shows the dynamical behaviour of the sum of all diffracted TH beams. For comparison, the cross-correlation between the pump and probe beams, generated at the sample surface via background free sum frequency generation, is also depicted in Fig. 1.

Obviously, in the centre of the temporal coincidence of the two pulses, when the intense parts of the fundamental beams interfere and hence an index grating with a high contrast is present in the material, the TH is enhanced by more than two orders of magnitude compared to THG from a single input beam. This is demonstrated in Fig. 2, which shows the central part of the TH diffraction pattern for two different pump–probe delays. (For the sake of clarity, only the TH beam collinear to the probe and the two central TH beams are present in the figure. The beam collinear to the pump and all higher diffracted orders were blocked using a diaphragm.) In Fig. 2a, the probe beam reaches the sample 1 ps before the pump beam arrives. Consequently, only single beam THG (here from the probe beam) occurs, being barely detectable. When the pump and probe beams arrive simultaneously and the index grating is present (Fig. 2b), a very strong TH is observed, yielding two bright spots between the zero order transmissions of pump and probe and one collinear beam (bottommost spot), which is also much stronger than in the single beam case. The overall conversion efficiency was measured with a photodiode to be $\approx 3\%$, i.e. for a total incident pulse energy of 200 μJ we obtained 6 μJ of UV pulses.

This efficient enhancement cannot be ascribed simply to the higher fundamental intensity during the temporal overlap of the pulses. Instead, we attribute this increase to a compensation of the phase mismatch, due to the transient grating.

If the angle between pump and probe is increased in a way that only the two central TH beams and the two TH beams collinear to the fundamentals are observed, the collinear beams are much weaker than the other two. This makes us believe that, in fact, in the presence of the grating the main THG occurs along the two central maxima and that the amplified collinear beams result from their first order diffraction.

In order to investigate this assumption in more detail, we measured the intensity dependence of the two central (zero order) TH beams on the probe beam intensity, while the pump



a $\Delta\tau$=-1ps **b** $\Delta\tau$=0

Fig. 2a,b. TH spots in the direction of the probe and between pump and probe for two different delays. **a** No grating exists; **b** the two fundamental beams interfere and create an index grating. In order to observe the weak signal in **a**, the detector sensitivity was enhanced

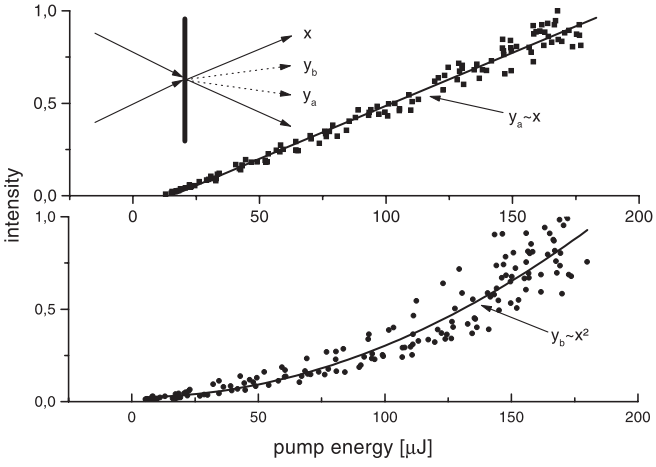


Fig. 3. Dependence of the two zero order TH beams on the probe beam intensity (x) for fixed pump intensity. The angle between the fundamental beams and hence the grating period was adjusted such that no higher orders could be observed

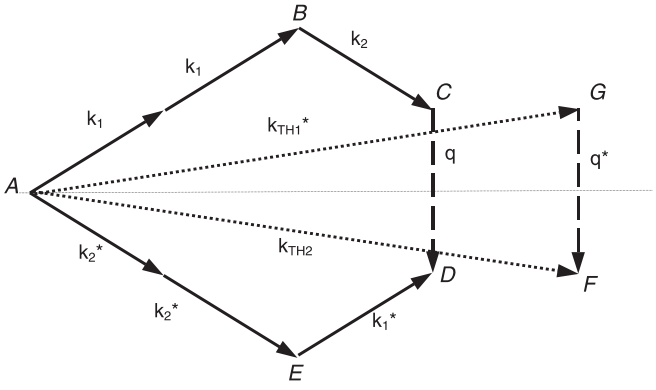


Fig. 4. Diagram of the wavevectors ($\boldsymbol{k}^*$ means the c.c. of $\boldsymbol{k}$) involved in THG and phase matching. *Solid arrows*: fundamental waves; *dotted arrows*: TH waves; *dashed arrows*: $\boldsymbol{q}$; $\boldsymbol{q}^*$: period vector of the transient grating. Note that in the drawing the phase mismatch is strongly exaggerated

intensity was kept fixed. The result is shown in Fig. 3. (Here we chose an angle of 5° between pump and probe such that no higher orders are observed. In fact, the effect appears to be independent of the angle between the beams. High intensities were measured for angles in the range from 5° up to 15°). For the TH spot close to the probe beam, Fig. 3 shows a quadratic intensity dependence on the probe intensity, whereas the TH signal close to the pump beam depends linearly. This suggests that the TH close to the probe beam (TH1) is generated from two probe photons and one pump photon, whereas the TH close to the pump beam (TH2) results from two pump photons and one probe photon. Figure 4 summarises this model by showing the corresponding $\boldsymbol{k}$ vectors for this process.

Since barium fluoride exhibits positive dispersion, the wavevector of the TH is larger than the sum of corresponding fundamental wavevectors. This should be equivalent to significant phase mismatch. Our experiment indicates, however, that the THG must proceed in a phase matched way. A possible model of this phase matching involves the induced index grating and is also sketched in Fig. 4. In a diagram such as that presented in this figure, phase matching, i.e. the conservation of momentum, is equivalent to a closed loop of the $\boldsymbol{k}$ vectors involved. In our model, we have to consider both branches, TH1 and TH2, simultaneously, coupled by momentum exchange with the grating. Thus, the two fundamental pathways ABC and AED are coupled by transferring momentum to the grating via the grating period vector, $\boldsymbol{q}$. In the same way, TH1 and TH2 are coupled, taking up momentum from the grating via $\boldsymbol{q}^*$. As a result, the complete loop ABC–DEA–FGA is closed and the total momentum is conserved. Obviously, the process is thus automatically self phase matched.[1]

In conclusion we have shown experimental results for very efficient THG in a material with positive dispersion. The enhancement of the generation process is due to self phase matching via a travelling grating induced by two non-collinear input beams. This enhancement is independent of the input parameters. At the same time the index grating leads to diffraction of the generated TH.

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[1]Note, that this model does not imply higher than third order interaction, since both the THG channels and the grating are all $\chi^{(3)}$ processes. Thus, all interactions are only contributions to a general $\chi^{(3)}(t)$ susceptibility.