Second-harmonic generation of amplified femtosecond Ti:sapphire laser pulses

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We study theoretically and experimentally second-harmonic generation (SHG) of 150-fs-duration amplified Ti:sapphire laser pulses at a wavelength of 780 nm in the nonlinear crystal KDP of different lengths and at different power densities as high as 150 GW cm^{-2}. The experimentally observed SHG conversion efficiency does not exceed 50%. It is shown theoretically that one possible process limiting the SHG efficiency at low as well as at high intensities is the modulation of the phase of the fundamental wave. In addition, continuum generation is observed at high intensities and can decrease the SHG efficiency.

Over the past few years, reliable laser systems delivering high-energy amplified femtosecond pulses have become available. This development has triggered a renewed interest in the generation of optical harmonics and other nonlinear-optical effects that can be used to convert the frequency of laser light. Nonlinear frequency conversion efficiencies of 30–50% from femtosecond lasers were previously reported,[1,2] but no detailed considerations of factors limiting the efficiency were carried out.

In this Letter we study both experimentally and theoretically the efficiency of conversion of 150-fs-duration amplified pulses at a wavelength of 780 nm to the second harmonic in different nonlinear crystals. In our experiments we use a commercial Ti:sapphire regenerative-amplifier laser system (Clark CPA-1) that yields pulses with an energy of 0.6 mJ at a 1-kHz repetition rate.[3] The spectral width of the pulses is 15 nm, and the diameter and the angular divergence of the laser output beam are 5 mm and 0.08°, respectively. Without any focusing of the beam the intensity achieved by this laser is ~20 GW cm^{-2}.

On one hand, because of their high intensity the amplified femtosecond pulses create a large-amplitude second-order nonlinear-optical polarization of the doubling crystal media. This, of course, facilitates an efficient conversion of the energy of the fundamental beam into the second harmonic. Another factor that influences the efficiency of second-harmonic generation (SHG) positively is the high damage threshold of the frequency-doubling crystals at subpicosecond pulse durations. On the other hand, there are negative factors that may disturb the efficiency of SHG. First, the amplitude and phase profiles of the fundamental pulse and the second-harmonic pulse develop in time and space in a different manner as the light propagates through the crystal. Second, other processes, which are due to higher-order optical nonlinearities of the media, may occur and start to compete with the SHG process.

We have studied SHG under the type I phase-matching condition in three different nonlinear crys-

tals—KDP, β-barium borate (BBO), and LiIO$_3$. All these crystals are transparent at the fundamental wavelength as well as at the second-harmonic wavelength. Some of the important nonlinear-optical parameters of these crystals are listed in Table 1. We studied SHG in all these crystals; however, most of the results that we present concern the KDP crystal.

In the theoretical part of our study we have calculated numerically the conversion efficiency and the time shape of the fundamental and the second-harmonic pulses, using the method described in Refs. 4–6. In our present model we have included the effect of the group-velocity mismatch between the fundamental and the harmonic wave. The set of coupled equations that describes the propagation of complex amplitudes A_1 and A_2 belonging to plain waves of the fundamental and of the second-harmonic frequency, respectively, is given by

$$
\begin{aligned}
\frac{\partial}{\partial z} A_1(z,t) &+ \frac{1}{u_1} \frac{\partial}{\partial t} A_1(z,t) \\
&= -i\sigma_1 A_1^* A_2 \exp(-i\Delta z), \\
\frac{\partial}{\partial z} A_2(z,t) &+ \frac{1}{u_2} \frac{\partial}{\partial t} A_2(z,t) \\
&= -i\sigma_2 A_1^2 \exp(-i\Delta z),
\end{aligned}
\tag{1}
$$

where u_1 and u_2 are the group velocities, σ_1 and σ_2 are the nonlinear coupling coefficients of the two waves propagating in the z direction, and $\Delta = 2k_1 - k_2$ is the phase mismatch. These equations describe a scalar interaction between two waves propagating at an exact phase-matching angle that corresponds to the type I synchronism. We solve these equations by integrating along the z axis from 0 to L, where L is the length of the crystal. In our model calculations, the initial intensity time profile of the fundamental pulse is taken to be Gaussian, with a full width at half-maximum of 150 fs. Most of the SHG efficiency loss is caused by the mismatch between the group velocities of the two waves rather than by the dispersion of the group velocity at one of the wavelengths.

Table 1. Nonlinear-Optical Parameters of SHG Crystals

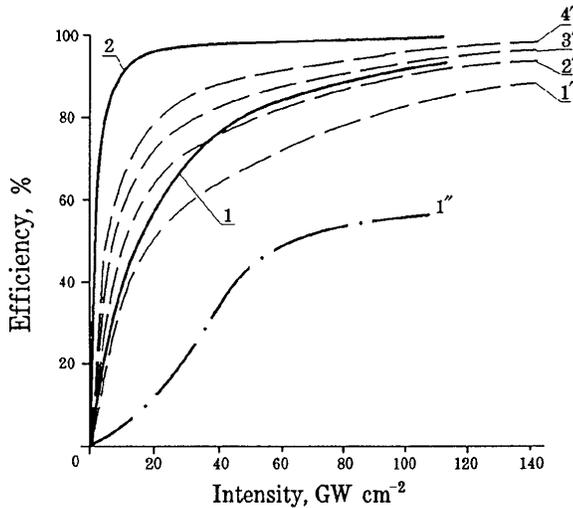| Crystal	Type I Synchronism Angle Θ	Effective Nonlinear Coefficient d_{eff} (pm V^{-1})	Group-Velocity Mismatch (fs mm^{-1})	Synchronism Angular Width $\Delta\Theta$ (min cm^{-1})	Synchronism Spectral Width $\Delta\lambda$ (nm cm^{-1})
KDP	$43°\ 42'$	0.31	83	4	1.6
LiIO$_3$	$44°\ 03'$	4.6	820	1.2	0.29
BBO	$29°\ 56'$	0.88	280	1.7	0.74

Fig. 1. Calculated conversion efficiency of SHG as a function of the fundamental intensity for KDP crystal lengths of 3 and 10 mm without consideration of group-velocity mismatch (curves 1 and 2, respectively), KDP crystal lengths of 3, 5, 7, and 10 mm with the effects of group-velocity mismatch considered (curves $1'$, $2'$, $3'$, and $4'$, respectively), and a 5-mm-long KDP crystal with the effects of group-velocity mismatch and phase modulation considered ($\pi/2$ during 150 fs; curve $1''$).

For this reason we are neglecting the dispersion of the group velocities in our present calculation.

Figure 1 shows the conversion efficiency of SHG as a function of the intensity of the fundamental pulse, calculated for different lengths of KDP crystal. The solid curves result when we do not take into account the group-velocity mismatch, and the dashed curves show the results when the group-velocity mismatch is accounted for. These curves show that, according to the coupled-wave equations, at the fundamental pulse intensities greater than 50 GW cm^{-2} more than 70% of the energy should convert to the second-harmonic wave and that, in the limit of even higher intensities, the conversion efficiency should approach 100%. The dotted–dashed curve is obtained if we take into account the fact that the phase of the fundamental pulse changes in time by $\pi/2$ from the beginning of the pulse to its end in a linear fashion. In contrast to the results discussed above, in this case a substantial decrease of the conversion efficiency occurs at high as well as at low fundamental intensities. From these results we can conclude that, if phase modulation of the fundamental wave occurs (for example, that owing to residual chirp at the output of the grating pulse compressor), then its influence on decreasing the conver-
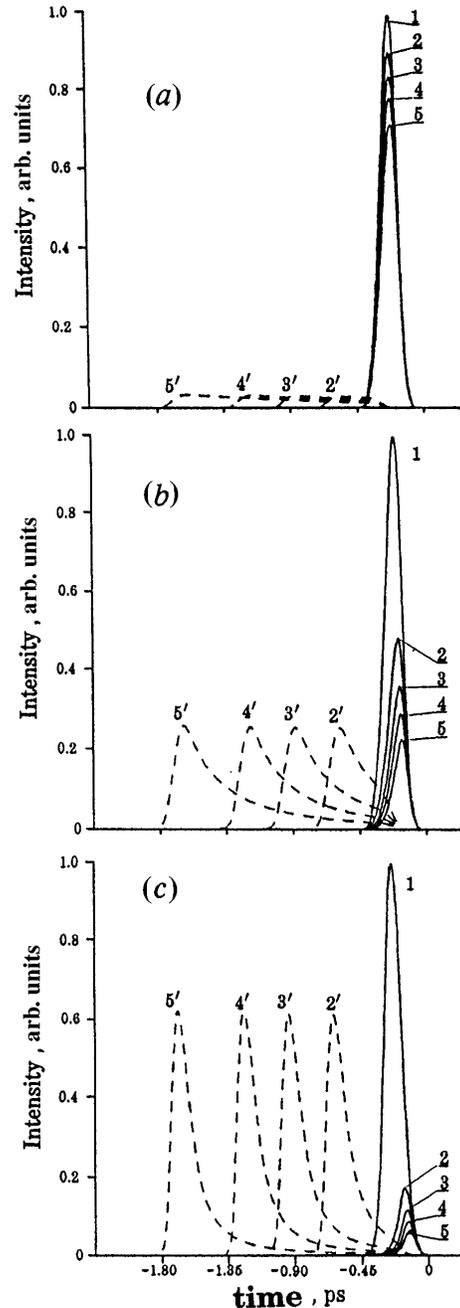

Fig. 2. Calculated pulse profiles at different fundamental beam intensities I of (a) 3 GW cm^{-2}, (b) 30 GW cm^{-2}, and (c) 150 GW cm^{-2}. Curve 1 is for $\lambda = 780$ nm at the input of the KDP crystal. Curves 2, 3, 4, and 5 are for $\lambda = 780$ nm and curves $2'$, $3'$, $4'$, and $5'$ are for $\lambda = 390$ nm at the output of KDP crystal lengths of 3, 5, 7, and 10 mm, respectively.
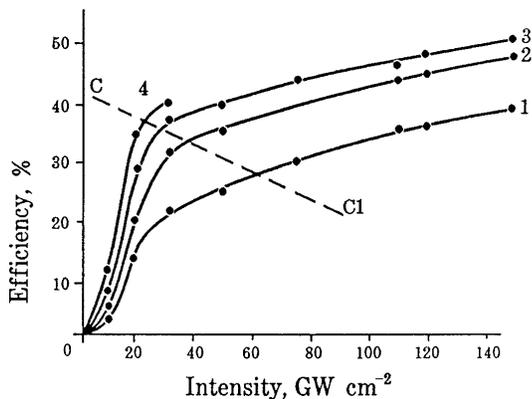
Fig. 3. Experimental conversion efficiency of SHG as a function of the fundamental intensity for KDP crystal lengths of 3, 5, 10, and 40 mm (curves 1, 2, 3, and 4, respectively).

sion efficiency should be even larger than that owing to group-velocity mismatch.

Figure 2 shows the calculated pulse time profiles of the fundamental intensity (solid curves) and the second-harmonic intensity (dashed curves) at different lengths of the KDP crystal and at three different input power levels of the fundamental beam. We carry out this calculation by taking into account the mismatch of the group velocities but without accounting for the phase modulation. At a relatively low pump intensity of 3 GW cm^{-2} the conversion to the second harmonic grows in proportion to the length of the crystal. However, because the group velocities are different the second-harmonic pulse is stretched out in time as the pulses travel through a thicker crystal. Because the conversion efficiency is smaller at lower powers the harmonic wave is produced more or less equally over the whole length of the crystal. A rather different situation occurs, however, when the power of the pump beam is chosen to be 50 times higher (150 GW cm^{-2}). In this case an efficient conversion already takes place within a short distance from the input face of the crystal. The second-harmonic pulse containing the major part of the optical energy is formed before the group-velocity mismatch can have any significant effect on the shape and the duration of the pulses. The remaining relatively small fraction of the fundamental radiation energy is also subsequently converted into the second harmonic, but at a later time, as light travels through the bulk of the crystal. This last circumstance gives rise to the long tail that follows the main pulse. It should be noted, however, that in this case the second-harmonic pulse does not broaden significantly and is almost as short as the fundamental pulse. Our calculation also shows that, at an intermediate intensity of 30 Gw cm^{-2}, the shape and the duration of the harmonic pulse vary with the length of the crystal.

In our experiments we measured the SHG efficiency in KDP crystals of four different lengths: 3, 5, 10, and 40 mm. We also performed similar measurements in LiIO$_3$ crystals of 5-, 10-, and 15-mm lengths and in a BBO crystal of 3-mm length. We used a 300-mm focal-length cylindrical lens to focus the beam, which permitted us to increase the pump beam intensity from 20 to 150 GW cm^{-2}.[5] To exclude side effects that occur when light propagates in the focal plane of a lens, we placed the nonlinear crystals 10 mm closer to the lens than the actual focal length. To attenuate the fundamental beam, we used a set of calibrated glass neutral-density filters.

Figure 3 shows the measured conversion efficiency to the second harmonic as a function of the pump intensity in KDP crystals of different lengths. In the LiIO$_3$ and BBO crystals, similar dependences were obtained, with maximum conversion not exceeding 50% but with the saturation occurring at lower pump intensities. We see that our experimental data are closer to the theoretical prediction in which we accounted for the modulation of the phase of the fundamental wave. At higher intensities we observed generation of a spectrally broad continuum with a maximum at 780 nm. The threshold for this process observed visually in KDP crystals is shown in Fig. 3 by the dashed line $C-C1$. The continuum generation may also contribute to the reduction of the SHG efficiency. It should be noted that, in the BBO and LiIO$_3$ crystals, the power threshold of the continuum generation is lower than that in KDP.

In conclusion, our results indicate that the SHG conversion efficiency of femtosecond high-power Ti:sapphire laser pulses is limited principally to ~50%—at least in the types of nonlinear crystal that we have investigated. Our model calculations show that the reason for this limitation may be the modulation of the phase of the pump pulses. The phase modulation decreases the conversion efficiency at high as well as at lower fundamental intensities, which is also what we observed in the experiment.

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