Efficient conversion of picosecond laser pulses into second-harmonic frequency using group-velocity dispersion

Yanjie Wang and R. Dragila
Laser Physics Centre, Research School of Physical Sciences, The Australian National University, P.O. Box 4, Canberra, Australian Capital Territory 2601, Australia
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A computer model has been used to investigate frequency doubling of 1-psec duration high-power pulses in potassium dihydrogen phosphate (KDP) for type-II phase matching. It has been found that group-velocity dispersion can be used to advantage by predelaying the ordinary and extraordinary polarizations appropriately in a thin KDP crystal with its axes aligned at 90° to the main conversion crystal. In that situation power conversion > 100% from the fundamental to the second harmonic can be obtained with simultaneous "compression" of the output pulse duration by up to a factor of 5.

I. INTRODUCTION

The role of various physical effects in the interaction of a laser radiation with matter usually depends on the wavelength of the radiation. For example, for laser driven fusion it is desirable to use short wavelengths heating radiation to increase the absorption and reduce detrimental effects, such as the generation for fast particles (see, e.g., Ref. 1). As a result, radiation from high-power Nd-glass lasers is usually converted into its higher harmonics to meet these requirements.

Relatively recently considerable interest has been focused on the interaction of very short and intense laser pulses with matter both for understanding of the fundamental physics and for the development of x-ray lasers. Specifically, these experiments are aimed to investigate absorption mechanisms;[2] multiphoton ionization;[3] the generation of short bursts of x-ray radiation for time-resolved studies of solids and high-density plasmas;[4] the pumping of x-ray lasers,[4,5] and kinetic effects within a very high temperature high-density (which is nearly uniform due to the absence of hydrodynamics) plasma.[6] Optical pulse compression is a standard technique[7] used in the development of compact TW/psec solid-state lasers. Unfortunately, an inherent feature of the pulse compression process is the production of a low-intensity long prepulse on which the main high-power pulse is superimposed. Since the energy in the prepulse can be a large fraction of that in the main pulse, its presence severely limits the usefulness of the laser for plasma physics experiments where low prepulse levels are essential. One method of reducing the prepulse intensity is to frequency double the laser output and use the normal nonlinear response of the doubling crystal to reduce the prepulse energy.

In the present paper we model frequency doubling of ≅1-psec duration high-power pulses in a potassium dihydrogen phosphate (KDP) crystal. For such short pulses both phase and group-velocity mismatch must be considered in determining the final conversion efficiency. There are two geometries of the doubling process: (i) type I, when two waves with ordinary (*o* rays) polarization coalesce to produce the second harmonic with extraordinary polarization (*e* ray), and (ii) type II, when *e* and *o* rays coalesce and produce a second-harmonic *e* ray. Since there is only a very small angular range available for the type-I process, type-II conversion is more frequently used, despite the fact that the group-velocity mismatch is smaller in the type-I doubling process. Although nominally the group-velocity dispersion more severely limits the harmonic conversion of very short pulses for type-II phase matching, we have found that it can, in fact, be used to advantage by predelaying the *o* and *e* rays appropriately in thin KDP crystal with its axes aligned at 90° to the main conversion crystal. In this case > 100% power conversion from the fundamental to the second harmonic can be obtained.

II. FORMULATION OF THE PROBLEM

Let us consider a situation when a type-II crystal slab ($0 \le z \le d$) in vacuum is irradiated by two pulses with *e* and *o* polarizations with center frequencies $\omega_e = \omega_0 = \omega$. As these two pulses propagate through the crystal they coalesce into a second-harmonic *e*-polarized pulse with center frequency $\omega_2 = 2\omega$. The electric fields associated with each of the pulses are assumed to have the following form:

$$E_j(t,z) = A_j(t,z) e^{i(\omega_j t - k_j z)} , \tag{1}$$

where $j = e, o, 2$ and $k_j = n_j \omega_j / c$. Here n_j is the refractive index, $n_j^2 = \epsilon_j = (1 + 4\pi\chi_j)$, ϵ_j is the dielectric permittivity, $\chi_j(\omega)$ is the linear dielectric susceptibility, and c is the speed of light. The conversion mechanism is then described by the following set of equations:[8]

$$\frac{\partial A_e}{\partial z} + \frac{1}{v_e} \frac{\partial A_e}{\partial t} = -K_e A_2 A_0^* e^{-i\Delta k z} - \alpha_e A_e , \tag{2}$$

$$\frac{\partial A_0}{\partial z}+\frac{1}{v_0}\frac{\partial A_0}{\partial t}=-K_0 A_2 A_e^* e^{-i\Delta k z}-\alpha_0 A_0 , \quad (3)$$

$$\frac{\partial A_2}{\partial z}+\frac{1}{v_2}\frac{\partial A_2}{\partial t}=-K_2 A_e A_0 e^{i\Delta k z}-\alpha_2 A_2 , \quad (4)$$

where v_j are corresponding group velocities, α_j represent the dissipation rate of each of the frequencies, $\Delta k=k_2-k_e-k_0$ represents the phase velocity mismatch,

$$K_j=\frac{2\pi i\omega_j^2}{k_j c^2}\chi_{\rm NL}$$

are the constants of nonlinear coupling, and $\chi_{\rm NL}$ is the nonlinear dielectric susceptibility. In what follows we will assume that the interacting waves are perfectly phase matched ($\Delta k=0$) and suffer no losses ($\alpha_j=0$). The set of coupled equations (2)–(4) has been solved numerically with the initial condition $A_j(t=0,0\le z\le d)=0; j=e,o,2$ and for various boundary conditions $A_e(t,z=0)=A_{e,0}(t-\tau_e)$, $A_o(t,z=0)=A_{o,0}(t-\tau_0)$, and $A_2(t,z=0)=0$, where $A_{e,0}$ and $A_{o,0}$ are amplitudes of the incident pulses in vacuum and $\tau_{e,o}$ are delays with which the peaks of the pulses reach the crystal/vacuum boundary $z=0$.

III. RESULTS

In all calculations presented below the wavelength of the incident radiation was assumed to be 1.054 μm and, correspondingly, the values of the nonlinear coupling constants were set as follows:[9] $K_2=8.4\times10^{-4}$ CGSE, $K_{e,o}=K_2 k_2\omega_{e,o}^2/k_{e,o}\omega_2^2$. Also, according to Ref. 10, the group velocities of the pulses involved are $v_0=1.966\,36\times10^{10}$ cm/s, $v_e=2.018\,77\times10^{10}$ cm/s, $v_2=1.988\,06\times10^{10}$ cm/s.

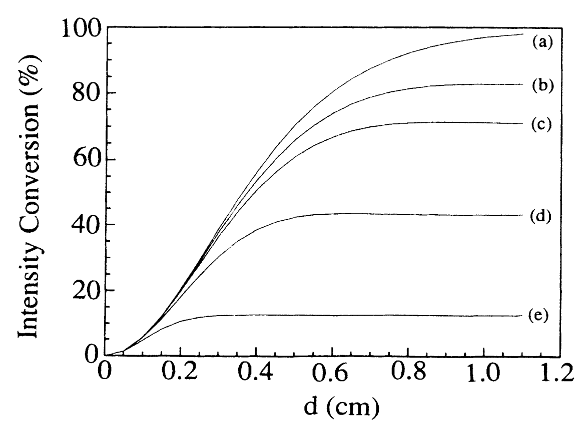

FIG. 1. Net intensity conversion efficiency vs the crystal thickness d for e and o pulses with equal intensity $I_e=I_0=3$ GW/cm^2 and equal pulse durations τ_L (FWHM): curve (*a*), $\tau_L\gg\tau_D=d|1/v_e-1/v_o|$; curve (*b*), $\tau_L=1.5$ psec; curve (*c*), $\tau_L=1.0$ psec; curve (*d*) $\tau_L=0.5$ psec; curve (*e*) $\tau_L=0.2$ psec.

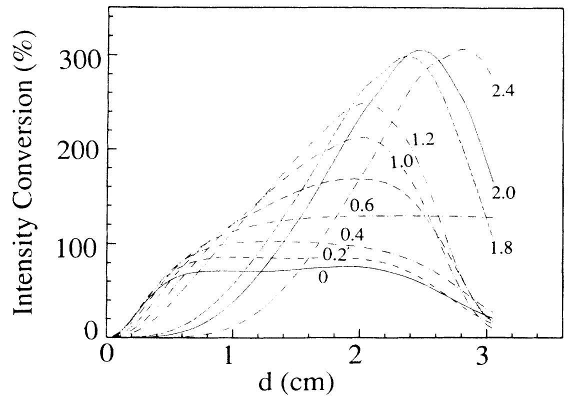

FIG. 2. Net intensity conversion as a function of the crystal thickness d with the predelay $\tau_p=\tau_e-\tau_o$ of the e pulse as a parameter (the corresponding numbers in the figure are in psec). The input pulse parameters are $\tau_L=1$ psec $I_e=I_o=3$ GW/cm^2.

A. The role of group-velocity dispersion

To demonstrate the effect of the group-velocity dispersion on the efficiency of the conversion we have run the first set of calculations assuming that the two pulses are incident simultaneously on the crystal. In this case the net intensity conversion $I_2/(I_e+I_0)$ versus the crystal thickness d is shown in Fig. 1 for e and o pulses with equal intensity $I_e=I_0=3$ GW/cm^2 and equal pulse durations τ_L [full width at half maximum (FWHM)]: curve (*a*) $\tau_L\gg\tau_D=d|1/v_e-1/v_0|$, i.e., the case of a long pulse when the group-velocity dispersion is unimportant; curve (*b*), $\tau_L=1.5$ psec; curve (*c*), $\tau_L=1.0$ psec: curve (*d*), $\tau_L=0.5$ psec; curve (*e*) $\tau_L=0.2$ psec. As can be seen, the group-velocity dispersion causes a substantial decrease in the efficiency of the frequency-doubling process for very short pulses and saturates for the crystal thickness $d_s\cong\tau_L|1/v_e-1/v_0|^{-1}$. The conversion efficiency can, however, be dramatically increased if one predelays the e

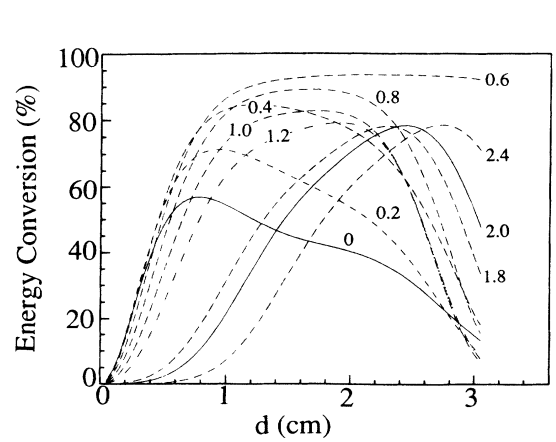

FIG. 3. Net energy conversion as a function of the crystal thickness d with the predelay $\tau_p=\tau_e-\tau_o$ of the e pulse as a parameter (the corresponding numbers in the figure are in psec). The input pulse parameters are $\tau_L=1$ psec, $I_e=I_o=3$ GW/cm^2.

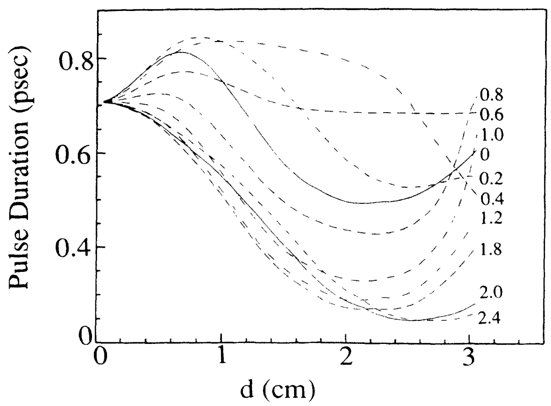

FIG. 4. Second-harmonic pulse duration as a function of the crystal thickness d with the predelay $\tau_p = \tau_e - \tau_o$ of the e pulse as a parameter (the corresponding numbers in the figure are in psec). The input pulse parameters are $\tau_L = 1$ psec, $I_e = I_o = 3$ GW/cm^2.

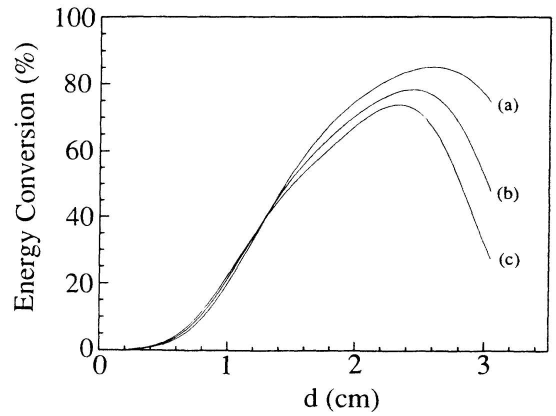

FIG. 6. Net energy conversion vs the crystal thickness for $\tau_p = 2$ psec and $\tau_L = 1$ psec. The three curves correspond to curve (a), $I_e = I_0 = 2.5$ GW/cm^2; curve (b), $I_e = I_0 = 3.0$ GW/^{2}; curve (c), $I_e = I_o = 3.5$ GW/cm^2.

pulse ($v_e > v_0$) to create conditions for longer overlap and, therefore, more efficient interaction of the two pulses.

B. The role of predelay

The role of such a predelay is illustrated in Figs 2–4. In all three cases both incident pulses have the same pulse duration $\tau_L = 1$ psec and same intensity $I_e = I_o = 3$ GW/cm^2. In Fig. 2 the intensity conversion is plotted as a function of the crystal thickness d with the predelay $\tau_p = \tau_e - \tau_0$ of the e pulse as a parameter. As can be seen, the net intensity conversion efficiency increases from $\cong 80\%$ for $\tau_p = 0$ up to $\cong 300\%$ for $\tau_p = 2$psec. The crystal thickness for which the peak intensity conversion efficiency occurs also increases with τ_p. Notice that for $\tau_p \cong 0.6$ psec the intensity conversion, energy conversion (Fig. 3), and pulse duration (Fig. 4) all saturate and remain independent of d within the caculated range. It means that for this particular predelay and the range of d considered the reconversion into the fundamental is negligible. Although the intensity conversion saturates for relatively long predelays, the energy conversion reaches its maximum for $\tau_p \cong 0.6$ psec (see Fig. 3). The existence of this maximum results from the short interaction times for short predelays on the one hand and reconversion back into the fundamental for too long a predelay. The effect of reconversion is also obvious from Fig. 4 where we have plotted the duration of the second-harmonic pulse versus τ_p. As can be seen, very short second-harmonic pulses ($\cong 0.25$ psec, which represents a compression by a factor of 4 relative to the incident pulse) can be obtained for relatively large delays ($\tau_p \cong 2$ psec) and for a crystal thickness $d \cong 2.5$ cm. The net intensity conversion still remains high ($\cong 300\%$) in these conditions although the net energy conversion drops slightly to $\cong 80\%$. An increase in the crystal thickness then leads to increasing reconversion which, obviously, results in a drop in the energy conversion and in an increase of the second-harmonic pulse duration.

The dependence of the intensity conversion on the

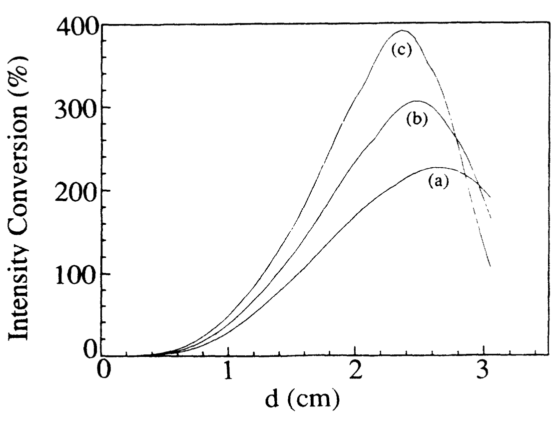

FIG. 5. Net intensity conversion vs the crystal thickness for $\tau_p = 2$ psec and $\tau_L = 1$ psec. The three curves correspond to curve (a), $I_e = I_o = 2.5$ GW/cm^2; curve (b), $I_e = I_o = 3.0$ GW/cm^2; curve (c), $I_e = I_0 = 3.5$ GW/cm^2.

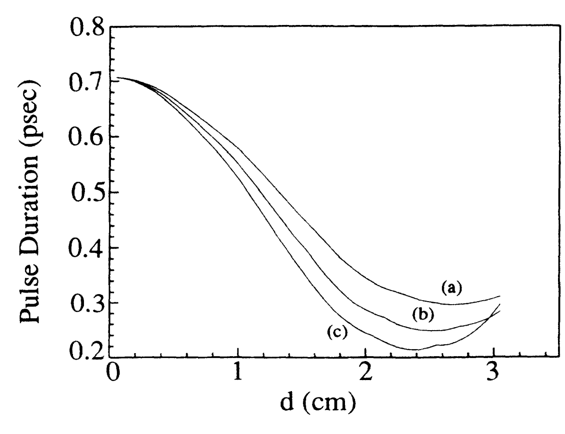

FIG. 7. Second-harmonic pulse duration vs the crystal thickness for $\tau_p = 2$ psec and $\tau_L = 1$ psec. The three curves correspond to curve (a), $I_e = I_o = 2.5$ GW/cm^2; curve (b), $I_e = I_0 = 3.0$ GW/cm^2; curve (c), $I_e = I_0 = 3.5$ GW/cm^2.

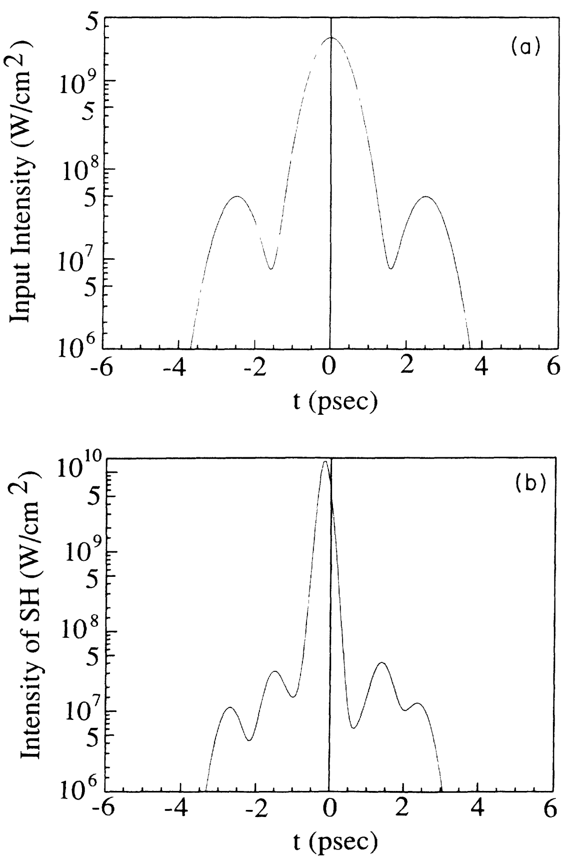

FIG. 8. Illustration of an improvement in the intensity contrast ratio by frequency doubling a pulse with satellites which model complex wings characteristic for compressed pulses: (a) the shape of the model input pulse, (b) the second-harmonic pulse obtained when two such pulses with $I_e = I_0 = 3$ GW/cm^2 and with the relative predelay $\tau_p = 2$ psec are launched into the type-II KDP crystal with the thickness $d = 2.5$ cm.

crystal thickness for various intensities of the incident pulses is shown in Fig. 5. Here the predelay τ_p is set to a fixed value $\tau_p = 2$ psec and both incident pulses have the same duration 1 psec. The three curves correspond to curve (*a*), $I_e = I_o = 2.5$ GW/cm^2; curve (*b*), $I_e = I_0 = 3.0$ GW/cm^2; curve (*c*), $I_e = I_0 = 3.5$ GW/cm^2. As could be expected, the intensity conversion efficiency increases with the increasing pump intensity, in contrast to the energy conversion which drops at the same time (see Fig. 6). The crystal thickness necessary to obtain maximum intensity or energy conversion efficiencies decreases with the increasing intensity of the pump. Also, the second-harmonic pulse duration decreases with the pump intensity and its minimum is relatively flat in the vicinity of $d \cong 2.5$ cm (see Fig. 7).

C. Contrast improvement

Finally, we have investigated the effect of the frequency doubling on the contrast ratio for the pulse involved. Theoretically, when a laser pulse is frequency chirped and subsequently compressed (e.g., by using an optical fiber and a pair of gratings) the output consists of a short main pulse and two wings each containing a series of satellites. Therefore the contrast of the compressed pulse is typically very low. Frequency doubling, as a nonlinear process, thus appear as a possible way of improving the contrast of compressed short laser pulses. To test this we have modeled the compressed pulse in the form shown in Fig. 8(a), i.e., we have replaced the complex wing structure by the first dominant satellite. The power contrast ratio in this case is assumed to be $\simeq 60$. Now, two such pulses with $I_e = I_0 = 3$ GW/cm^3 with the relative predelay $\tau_p = 2$ psec are launched into the type-II KDP crystal with the thickness $d = 2.5$ cm. The output second-harmonic pulses is shown in Fig. 8(b). The net intensity conversion dropped from $\cong 300\%$ (the case without the wings, Fig. 5) down to $\cong 200\%$ due to conversion of some portion of energy of the input pulses into the wings of the second-harmonic pulse. As could be expected the contrast did improve and is now $\cong 300$. At the same time, the energy contrast improved by a factor of $\cong 3$. This, however, is still not enough to make such pulses suitable for short-pulse interaction experiments because the prepulse still contains a considerable amount of energy and hence it can substantially modify the initial conditions before the main pulse arrives. Our results therefore indicate that, in those cases when the pulse contrast is crucial, it is better, though not necessarily simpler, to construct an oscillator that generates the final ≈ 1-psec duration pulse directly and this is then decompressed, amplified, and recompressed to generate high power rather than using a fiber-grating compressor system. In accordance with the scenario proposed above, the pulse can then be further "compressed" by frequency doubling (see Figs. 4 and 5) with the aim of achieving pulse durations < 1 psec.

IV. CONCLUSION

To summarize, we have shown that the apparent disadvantage of type-II second-harmonic conversion of short laser pulses—group-velocity dispersion—can be used, in fact, to advantage by predelaying the ordinary and extraordinary polarizations appropriately. This can be achieved by using a thin KDP crystal with its axes aligned at 90° to the main conversion crystal. There is an optimum predelay of the *e* pulse and thickness of the KDP crystal that, for given intensity and duration of the pulses at the fundamental frequency, results in a minimum pulse duration of the second-harmonic pulse and maximum intensity and energy conversion. For example, for $I_e = I_0 = 3$ GW/cm^2 input 1-psec pulses, $\tau_p = 2$ psec and $d = 2.5$ cm one obtains the net intensity conversion $\cong 300\%$, the net energy converions $\cong 80\%$ and the output pulse duration $\cong 0.25$ psec, i.e., a "compression" by a factor of 4. This compares favorably with the net intensity conversion rate of 80% and energy conversion of $< 60\%$ obtained in the standard frequency-doubling scheme without the predelay. We have also tried to simulate the frequency doubling of laser pulses compressed by the fiber-grating compressor scheme with the aim to improve characteristically poor contrast of such pulses. The particular example analyzed here showed the intensity contrast improvement by a factor of 5 (the energy contrast by a factor of 3) which, however, in absolute terms is not sufficient for the frequency-doubled pulses to be used in short-pulse interaction experiments.

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