Second Harmonic Generation of Femtosecond Laser at One Micron in a Partially Deuterated KDP *

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Partially deuterated KDP is an ideal nonlinear crystal for second-harmonic generation (SHG) of femtosecond lasers at 1 μm, which features with vanished group-velocity mismatch at its retracing point of phase-matching. We numerically investigate the characteristics of SHG with femtosecond lasers in a partially deuterated KDP, which shows that group-velocity dispersion plays an important role. This spectrally non-critical phase-matching configuration can support both high efficiency and large acceptance bandwidth.

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Over the last decade there have been spectacular developments in ultrafast laser technology, due to the introduction of broadband solid-state laser materials and of Kerr lens mode-locking and amplification techniques. With these advances, high-power femtosecond lasers can produce peak powers approaching petawatt at fixed wavelengths of 800 nm and 1 μm. Second-harmonic generation (SHG) is a powerful technique of wavelength extension. To increase the conversion efficiency and to avoid harmonic pulse broadening in the regime of ultrashort lasers, the overall group-velocity mismatch (GVM) between the fundamental (FH) and the second-harmonic (SH) pulses in the crystal needs to be sufficiently small compared with the FH pulse duration, or equivalently in frequency domain the spectral acceptance of the crystal needs to be sufficiently large compared with the FH pulse bandwidth.[1,2] In order to meet this requirement, very thin crystals must be used, which results in low conversion efficiency.[3]

Nonlinear crystals normally exhibit inevitable GVM. Several approaches, such as achromatic phase-matching (APM),[4−6] multi-crystal sequence,[7] tilted quasi-phase-matched gratings,[8] Čerenkov phase-matching,[9] have been tried to broaden the effective spectral acceptance bandwidth of the doubler. Unfortunately, these approaches are not possible to be used in practice due to the complexity and quite strict requirements for alignment of the optics. Thus, it is interesting to search for nonlinear crystals with zero-GVM. In this case, the doubling configuration is just like the conventional one and no modifications of the input light are needed, which makes it more practical in the sense of simplicity, reliability and conversion efficiency. The crucial is the availability of proper crystal for a specific wavelength corresponding to zero-GVM. Several mature crystals were investigated and found to be suitable for broadband second-harmonic generation at certain wavelengths, such as BBO for 1.5 μm,[10] LBO for 1.3 μm,[11] MgO-doped PPLN for 1.55 μm communication band,[12] partially deuterated KDP in the region from 1.034 μm to 1.179 μm and potentially KDP analogs from 1.013 μm to 1.278 μm.[13,14] Among these crystals, partially deuterated KDP is particularly interesting since it can be made large in size and matches with the femtosecond laser from Nd:glass system.

In many common situations of SHG, group-velocity dispersion (GVD) is much less important than GVM and is thus neglected usually. For example, the characteristic dispersion length of a 100-fs pulse at 800 nm in KDP is about 5 cm, while the GVM length (the length over which the FH and SH pulses separate by the pulse duration owing to GVM) is nearly 1 mm. When the effect of GVM is negligible, GVD will be a dominant factor of SHG for femtosecond lasers. SHG in this situation is important for efficient conversion, and has not been addressed in previous studies. In this Letter, we study the SHG of femtosecond lasers in a partially deuterated KDP where GVD plays a major role. Since the first-order wavelength-sensitivity of wave-vector mismatch $\partial\Delta k/\partial\lambda$ (i.e., GVM) vanishes, the spectral acceptance bandwidth is roughly determined by the second-order wavelength-sensitivity parameter (i.e., GVD) and is inversely proportional to the square root of the crystal length. The characteristics of this spectrally non-critical phase-matching configuration of SHG are investigated systematically, which show unique features compared with conventional SHG of femtosecond lasers with significant

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GVM.

To study SHG with femtosecond pulses, the standard nonlinear-coupled wave equations in the time domain are solved numerically, which neglect the transverse effects and all the derivatives of the linear index of refraction beyond the second.[15,16] We first study the phase-matching properties of a partially deuterated KDP (Fig. 1). For SHG of the type-I phase-matching, there exists a retracing point on the phase-matching curve around 1 μm (Fig. 1(a)). The retracing point (i.e., wavelength) occurs at a specific phase-matching angle for a certain nonlinear crystal, and depends on the deuteration level. As the first order of approximation, this retracing behaviour is a mechanism of non-critical phase-matching in the frequency domain. Equivalently, non-critical phase-matching exactly corresponds to zero-GVM at retracing wavelength in the time domain (Fig. 1(b)). Therefore, partially deuterated KDP is an ideal crystal for SHG of femtosecond lasers at 1 μm. The wavelength for retracing or zero-GVM can be tuned with the deuteration level, which is important for practical use. For instance, partially deuterated KDP with a deuteration level of ~12% cut at $\theta = 41^\circ$ corresponds to a retracing wavelength at 1054 nm that matches with the Nd:glass laser system.

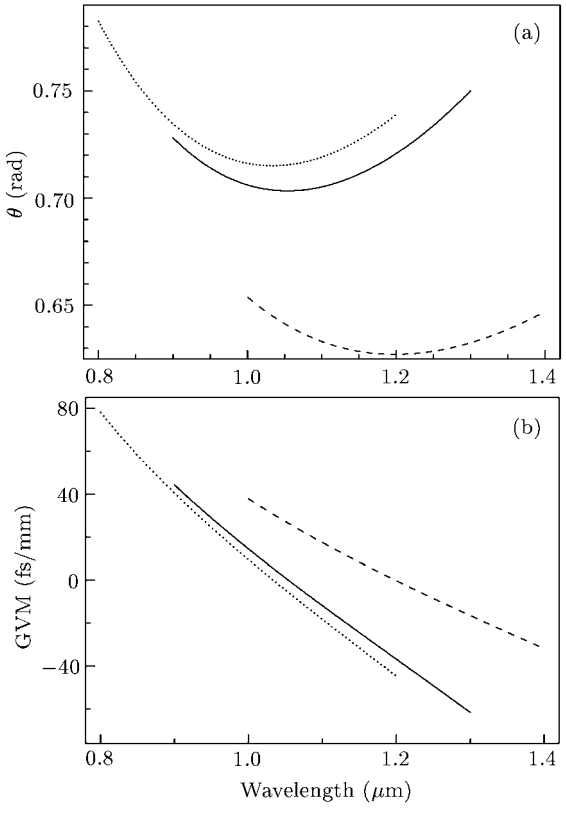

Fig. 1. (a) Phase-matching angle θ and (b) the corresponding GVM versus wavelength $\lambda_{1\omega}$ for type-I doubling in KDP (dotted line), KD*P (dashed line) and 12% partially deuterated KDP (solid line).

When GVM vanishes, the dispersion effect and wave-mixing processes will significantly alter the output SH spectrum. For the sufficiently broad input FH spectrum, SHG of femtosecond pulses at retracing point does not really independent of wavelength. The nonlinear length, $L_{NL} = n\lambda_p/(\pi\chi^{(2)}A_0)$, is used as a measure of FH intensity in the simulations, where n, λ_p and A_0 are the refractive index, wavelength and field amplitude, respectively. Detailed numerical results of the output SH spectra are shown in Fig. 2, where ΔKL, an equivalent measure of the crystal orientation angle θ, is the phase-mismatch at the retracing wavelength (i.e., 1.054 μm). The two main peaks in Figs. 2(a) and 2(b) correspond to the phase-matched components, while other minor peaks can be attributed to those spectral components where phase-mismatch are odd multiple of π. The locations of all these peaks on the spectrum depend on the orientation of the crystal. With the decreasing ΔKL, i.e., θ approaches to the retracing point, the two phase-matched peaks move to the retracing wavelength symmetrically, and link together to form a single peak when $\Delta KL = 0$ is exactly reached, as shown in Fig. 2(c). The spectral width of this peak is much broader than those of the individual peaks, indicating physically that the spectrally non-critical phase-matching is satisfied.

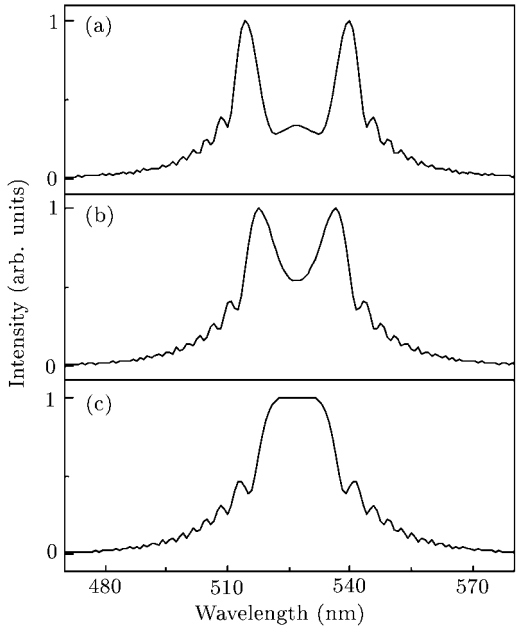

Fig. 2. Numerical results of the output SH spectra with a 1-cm-long partially deuterated KDP at (a) $\Delta KL = 2.5\pi$ (b) $\Delta KL = 0.5\pi$ (c) $\Delta KL = 0$. The simulations are carried out under the regime of low pump intensity ($L_{nl} \gg L$). A Fourier-transform limited Gaussian pulse with bandwidth of 150 nm is adopted, and dispersion values are taken to be $\mathrm{GVD}_{1W} = -13\,\mathrm{fs}^2/\mathrm{mm}$ and $\mathrm{GVD}_{2W} = 70\,\mathrm{fs}^2/\mathrm{mm}$.

The results shown in Fig. 2 also imply that there

exists an acceptance bandwidth for the non-critical phase-matching beyond which phase cannot be effectively matched. To evaluate the acceptance bandwidth governed by GVD, the output SH spectra are calculated for different input bandwidths of FH pulses (solid lines in Fig. 3). For comparison, the SH spectra under conditions of infinite acceptance bandwidth (neglecting GVD) are also illustrated (dashed lines). As is expected, the SH spectrum is identical to that in neglecting GVD when the FH bandwidth is relatively small. For a 10-mm long crystal, GVD will start to limit the output SH spectra significantly if the FH bandwidth is larger than 27 nm. Fig. 4 shows the simulated conversion efficiency. With an FH bandwidth of 27 nm, the efficiency is very close to that of the ideal case with narrow FH bandwidth. Thus we determine that the acceptance bandwidth $\Delta\lambda$ is 27 nm in a 10-mm crystal, and the scaling of $\Delta\lambda$ with crystal length L can be written as

$$\Delta\lambda \cdot \sqrt{L} = 27\text{nm} \cdot \text{cm}^{1/2} \qquad (1)$$

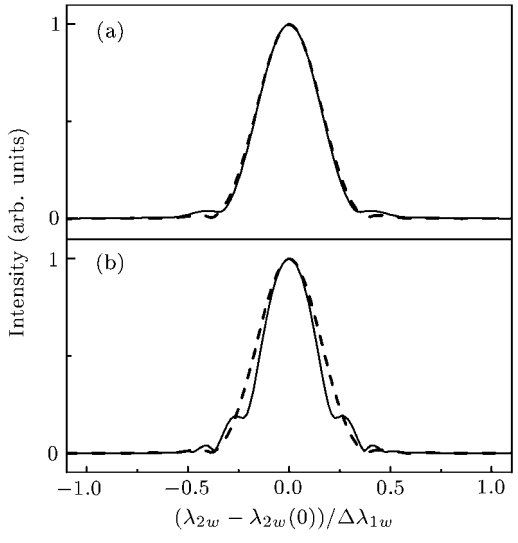

Fig. 3. Numerical results of the output SH spectra at retracing ($\Delta KL = 0$) for different values of FH bandwidth: (a) $\Delta\lambda_{1\omega} = 27$ nm, and (b) $\Delta\lambda_{1\omega} = 54$ nm. The simulations are carried out with a 1-cm-long partially deuterated KDP under the regime of high pump intensity ($L_{nl} = 3.5$ mm). For comparison, the ideal SH spectra under infinite acceptance bandwidth are also illustrated (dashed lines).

It is worth noting that the present definition of acceptance bandwidth is under a condition of high efficiency. Compared to the conventional definition based on the small-signal approximation, this definition is closer to the practical application of design. For efficient SHG of a femtosecond laser with bandwidth $\Delta\lambda$, the crystal length L can be directly determined by Eq. (1). At high intensity of about 200 GW/cm^2, an efficient SHG can be designed with a crystal length of 1 mm, which corresponds to an extremely large value of acceptance bandwidth of about 85 nm. As a comparison, the acceptance bandwidth of high efficiency in the conventional KDP crystals is only about 15 nm under the same condition. Therefore, the retracing of phase-matching will greatly enhance the acceptance bandwidth by nearly an order of magnitude.

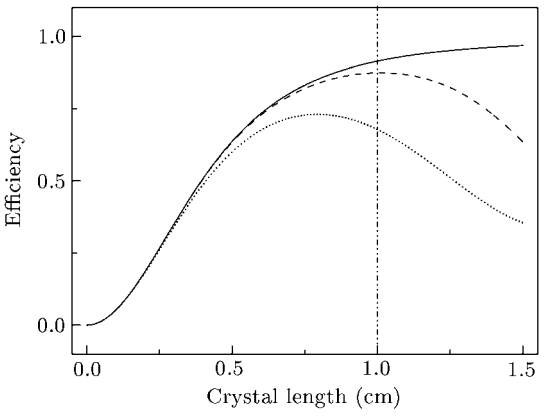

Fig. 4. Calculated conversion efficiency versus crystal length under high pump intensity ($L_{nl} = 3.5$ mm) for different values of FH bandwidth: (1) $\Delta\lambda_{1\omega} = 0$ (solid line), (2) $\Delta\lambda_{1\omega} = 27$ nm (dashed line), and (3) $\Delta\lambda_{1\omega} = 54$ nm (dotted line). A vertical line at 1 cm denotes the design of crystal length for $\Delta\lambda_{1\omega} = 27$ nm.

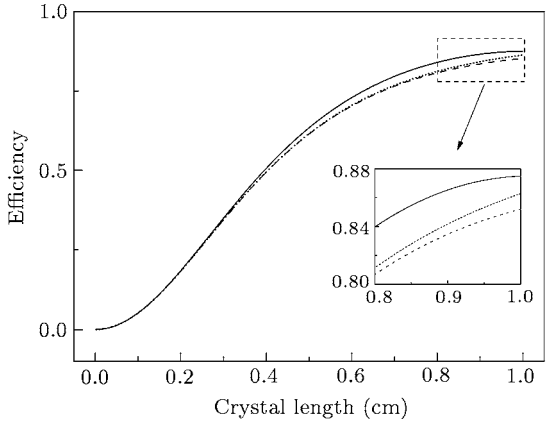

Fig. 5. Calculated conversion efficiency versus crystal length under high pump intensity ($L_{nl} = 3.5$ mm) for different deuterations: (1) optimal deuterated level of 12% (solid line), (2) 16% deuterated level (dashed line), and (3) 16% deuterated level but phase-mismatching $\Delta KL = -0.08\pi$ (dotted line).

It is important to note that phase-matching at retracing is not strictly independent of wavelength. The phase-mismatching due to GVD will be significant when increasing the crystal length beyond the design value, and the SHG of femtosecond laser at retracing point will be limited by GVD eventually. As clearly shown in Fig. 4, the crystal length can be designed to be 10 mm for an FH bandwidth of 27 nm, and the efficiency will be significantly lower for a longer crystal ($L > 10$ mm). On the other hand, spectral bandwidth

is the most important parameter in SHG at the retracing point. We have proven during the simulations that the conversion efficiency for a highly chirped pulse is almost identical to that with femtosecond pulses at fixed intensity. Thus, the obtained results can be valid for broadband lasers with various pulse durations from tens of femtosecond to nanosecond.

Since the retracing behaviour of phase-matching is crucial, it is important to examine the sensitivity of the conversion efficiency of SHG on deuteration level. A deuteration deviated from the optimal value will cause a reduction of efficiency (Fig. 5). From the calculated results, we can conclude that a deuteration level of (12 ± 2) % is practically acceptable for retracing wavelength of 1054 nm. Obviously, a deuteration level beyond (12 ± 2) % will result in a smaller value of acceptance bandwidth and reduced conversion efficiency as well. It is interesting that this effect can be compensated for to some extent by introducing a proper phase-mismatching as also shown in Fig. 5. This may be explained easily by Fig. 1(a), i.e. assuming that the retracing wavelength is longer than that of the laser, a proper phase-mismatching at the central wavelength will favour the overall phase-matching condition for all laser components if there is a perfect phase-matching towards the longer wavelength.

In conclusion, we have numerically studied the SHG of femtosecond pulses in a partially deuterated KDP crystal at its retracing point vicinity. The spectral characteristics of the generated SH pulses and its conversion efficiencies in the strong pump regime are investigated in detail, which shows that GVD plays an important role. This spectrally non-critical phase-matching configuration can support both high efficiency and large acceptance bandwidth, which makes this approach attractive. We conclude that partially deuterated KDP crystal is an ideal nonlinear crystal for femtosecond lasers at 1 μm.

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