Phase-matching temperature shifts in blue generation by frequency doubling of femtosecond pulses in KNbO$_3$

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We have demonstrated temperature rises that result from nonlinear absorption in single-pass frequency-doubling experiments using femtosecond pulses and a 3-mm-thick KNbO$_3$ crystal. These temperature changes shift the phase-matching curve and must be accounted for to optimize the conversion efficiency. We obtained a maximum second-harmonic generation (SHG) efficiency of 66% at an input power of 107 mW and a slope efficiency of ~1.5%/mW at low input powers. We have investigated, for the first time to our knowledge, the focusing dependence of the phase-matching temperature. We have also found that the temperature-dependent SHG efficiency in femtosecond SHG experiments is significantly different from that obtained for frequency doubling of continuous-wave light.   © 1999 Optical Society of America [S0740-3224(99)01008-5]

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1. INTRODUCTION

Frequency conversion through nonlinear processes is widely used to generate coherent radiation in spectral regions that are void of direct sources. A commonly used nonlinear optical frequency conversion is second-harmonic generation (SHG). SHG in the wavelength range from 420 to 530 nm is a topic of considerable research interest because of its numerous applications including optical data storage, optical printing, visual displays, photolithography, and medical uses. Potassium niobate (KNbO$_3$) has been used successfully for efficient frequency doubling and parametric oscillation through noncritical phase matching because of its high nonlinear coefficient.[1,2] Several studies of high blue output power and high conversion efficiency by frequency doubling of cw and pulsed lasers in an external cavity or in a single pass through a nonlinear crystal (NLC) have been reported.[3–8] The efficiency of frequency doubling in a NLC by use of broad-bandwidth femtosecond pulses is usually subject to limitations[9–15] imposed by a group-velocity mismatch (GVM) between the fundamental and the second-harmonic (SH) waves, which is intrinsic to the nonlinear material. It was previously shown that SHG with femtosecond pulses in a thick KNbO$_3$ NLC with large GVM leads to good slope efficiency at low input powers because of the high intensity of femtosecond pulses,[16] although group-velocity walk-off broadens the pulse into the picosecond regime and restricts phase-matching to a narrow SH frequency range. However, the SHG efficiency saturates at roughly 60% for higher input powers. Measurement of transmitted fundamental infrared (IR) and blue powers showed that this saturation was linked to nonlinear absorption. This saturation and nonlinear absorption are most likely related to a blue-induced IR absorption in KNbO$_3$ previously measured in cw laser experiments.[17–19] However, the possibility of an additional nonlinear absorption effect owing to two-photon absorption arising from the high intensity of the femtosecond pulses is not excluded. Femtosecond SHG experiments in a thick lithium niobate (LiB$_3$O$_5$) NLC have also been reported,[6] with efficiency reaching ~41%.

Here we show that the nonlinear or blue-induced IR absorption in KNbO$_3$ leads to a significant temperature rise in the femtosecond experiments, which must be accounted for in setting the phase-matching conditions. We investigate, for the first time to our knowledge, the focusing dependence of the phase-matching temperature. We find that the optimum phase-matching temperature depends significantly not only on the input power but also on the focusing, which is attributed to the heating of the crystal that results from nonlinear absorption. As was pointed out for the cw experiments,[18,19] the heating effect that results from absorption of both fundamental and SH waves in a NLC deteriorates the conversion efficiency for high input powers. Polzik and Kimble[4] theoretically calculated a radially varying temperature distribution within the crystal driven by the absorbed power. We experimentally demonstrate that for tighter focusing at high input power the peak of the conversion efficiency versus the focal position in the crystal shifts as the crystal temperature changes, an effect that is most likely related to a spatially varying index of refraction associated with a lateral and longitudinal temperature variation that is due to nonlinear absorption.

2. THEORY ON THE EFFICIENCY OF SECOND-HARMONIC GENERATION BY USE OF FEMTOSECOND PULSES

We briefly discuss a few theoretical points that are pertinent to SHG by use of femtosecond pulses with large GVM. First we introduce a walk-off length $l_T = t_p/\alpha$

over which the walk-off in time is equal to one pulse width t_p, where α is the GVM, which is defined as the difference in the group velocities v:

$$\alpha = \left(\frac{1}{v_{2\omega}} - \frac{1}{v_\omega} \right). \tag{1}$$

In the short-pulse limit, l_T is much smaller than the crystal length L ($l_T \ll L$) and, assuming chirp-free (transform-limited) pulses and weak focusing, the SHG process is essentially independent in each thickness l_T of the crystal, and therefore we obtain total conversion efficiency by adding the efficiency in each length l_T of the NLC. As a result, for low conversion efficiency (no pump depletion) the conversion efficiency in terms of the SH and fundamental pulse energies $U_{2\omega}$ and U_ω, respectively, is approximately given by[16]

$$\frac{U_{2\omega}}{U_\omega} = \gamma \left(\frac{U_\omega}{t_p} \right) \left(\frac{l_T^2}{b} \right) \left(\frac{L}{l_T} \right) = (\gamma U_\omega) \frac{L}{\alpha b},$$

$$\gamma = \frac{4 \omega_0^2 d_{\text{eff}}^2}{n^2 c^3 \epsilon_0 \lambda}. \tag{2}$$

Here ω_0 is the center frequency of the input pulse, n is the refractive index, d_{eff} is the effective nonlinear coefficient of the crystal, $b = (2\pi w_0^2 n/\lambda)$ is the depth of the focus, and w_0 is the beam radius at the focus. Therefore, for weak focusing, the conversion efficiency is linearly proportional to L/b. We can also handle stronger focusing by taking $b = [2\pi w^2(z) n/\lambda]$ in Eq. (2) to be z dependent according to the Gaussian beam formula and then integrating over the length of the crystal. This approach is valid provided that $l_T < b$, i.e., the temporal walk-off length remains shorter than the depth of focus. The result as derived in Ref. 16 is

$$U_{2\omega}/U_\omega = (\gamma U_\omega/\alpha)\tan^{-1}(L/b), \tag{3}$$

where no pump depletion is assumed, as before. Therefore, for strong focusing, we can expect that the SHG efficiency may be increased as the focusing is tighter and the GVM is smaller. In the limit of tight focusing ($l_T < b \ll L$), the conversion efficiency in Eq. (3) is approximately given by

$$U_{2\omega}/U_\omega \cong (\pi/2)(\gamma U_\omega/\alpha). \tag{4}$$

Thus, for $l_T < b \ll L$, the conversion efficiency is independent of focusing.

Now we discuss the SHG spectral width, which depends on α, L, and b. We can calculate a SH bandwidth from the phase-matching spectral response function $H_{\text{PM}}(\Omega)$, which is given by[13–15]

$$|H_{\text{PM}}(\Omega = 2\omega - 2\omega_0)|^2 = \frac{\sin^2(\Delta k L/2)}{(\Delta k L/2)^2}, \tag{5}$$

where $\Delta k = \alpha(2\omega - 2\omega_0)$. The full width at half-maximum (FWHM) of $|H_{\text{PM}}(\Omega)|^2$ is calculated to be 2.77 rad. Therefore, for weak focusing, the SH bandwidth $\Delta\nu$ is expressed by

$$\Delta\nu = 0.88/\alpha L. \tag{6}$$

Larger GVM results in narrower SHG bandwidths; this is consistent with the increased SH pulse width of order

αL.[13–15] For tight focusing ($l_T < b \ll L$), L should be replaced with the effective interaction length b, i.e., a SH bandwidth $\Delta\nu \approx 0.88/\alpha b$. Therefore for tight focusing the SH pulse width is decreased and the bandwidth is increased.

3. EXPERIMENTAL RESULTS AND DISCUSSIONS

We performed experiments with a mode-locked Ti:sapphire laser with pulse width of ~120 fs at an 80-MHz repetition rate at 860 nm under various focusing conditions and an a-cut, 3-mm-thick $KNbO_3$ crystal that was temperature tuned for noncritical type I phase matching for SHG to 430 nm in a single pass through the crystal. The GVM for a $KNbO_3$ crystal is 1.2 ps/mm[1] and is much larger than those for β-barium borate (β-BaB_2O_4) and LiB_3O_5 crystals.[5] Despite the $KNbO_3$ crystal's large GVM, high-efficiency blue generation by frequency doubling is achieved because it has a large nonlinear optical coefficient and because noncritical 90° phase matching (no spatial walk-off) can be realized by temperature tuning the crystal.[16] Curves for SHG versus input power were acquired over the range 5–285 mW with a variable attenuator. Input IR and output SH powers were measured with a calibrated powermeter before the $KNbO_3$ crystal and after a red blocking filter, respectively, as shown in Fig. 1. We also measured the transmitted IR power through the crystal with a blue-blocking filter after lens L_2. A prism pair was used to ensure that the fundamental pulse from the laser was chirp free. The $KNbO_3$ crystal was temperature controlled by a thermoelectric cooler within 0.05 °C. The input beam before focusing lens L_1 was circular, with measured beam diameter of 2 mm. Data were taken with a series of nine lenses with focal lengths ranging from 25 to 140 mm, yielding calculated depths of focus (b) of 0.34 to 10.7 mm and L/b values from 8.8 to 0.28.

Figure 2 shows SHG efficiencies at different crystal temperatures and optimum phase-matching temperatures versus input power for a focal length of 31 mm (depth of focus $b \approx 0.53$ mm). The highest efficiency for a high input power of 282 mW is achieved at a crystal temperature of 20.4 °C (denoted high power optimization), whereas for an input power of 5 mW it is achieved at 24.3 °C (denoted low power optimization), where the input wavelength is fixed at 860 nm and the spacing d between the crystal and focusing lens L_1 is the same at high power optimization. The conversion efficiency at low power optimization is significantly decreased for input powers above 100 mW because of nonoptimum phase-
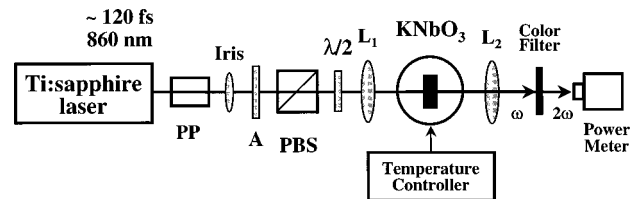

Fig. 1. Experimental setup for SHG by frequency doubling of femtosecond pulses: PP, prism pair; A, variable attenuator; PBS, polarizing beam splitter.

matching that results from an increase in the crystal temperature. When the externally applied crystal temperature is reset at each input power, the SHG efficiency is increased considerably. As a result, a maximum SHG efficiency of 66% was achieved at an input power of 107 mW and a crystal temperature of 23 °C. Moreover, the conversion efficiency rises very fast at low powers, with a slope efficiency of ~1.5%/mW for a focal length of 31 mm.

Polzik and Kimble reported that the crystal temperature for optimum phase matching at high input powers was 1–3 °C lower in cw operation than for swept operation. In our femtosecond experiments the crystal temperature for optimum phase matching decreases with increasing input power at a rate of ~13.3 °C/W for this focusing condition because of the thermal effect that results from nonlinear absorption. The importance of the power dependence of the phase-matching temperature was not carefully investigated in our previous femtosecond experiments with $KNbO_3$ (Ref. 16); properly resetting the temperature with power permits an improved optimization in the current experiments. Note that, although the phase-matching temperature is optimized at each input power, the efficiency still saturates and begins to roll over at high powers because of direct loss from the nonlinear absorption or blue-induced IR absorption and possibly because of incomplete phase matching owing to a spatial temperature variation.

Figure 3 shows the absorbed power and the temperature rise versus input power for a focal length of 31 mm. We measured the temperature rise as the difference between the phase-matching temperature optimized at each input power and at low power optimization. For fixed focusing, the temperature rise increases with greater input power. In addition, the absorbed power, which we measured by subtracting the output blue power and the transmitted red power from the input power, also increases with increasing input power. The temperature rise and the absorbed power exhibit similar behavior, which clearly links these two effects. These temperature rises caused by nonlinear absorption are expected to be spatially inhomogeneous and therefore should impose a second limit on the conversion efficiency in addition to limits that are due to nonlinear absorption, through spatial variation of the phase-matching temperature.

Fig. 3. Absorbed power and temperature rise versus input power for a focal length of 31 mm.
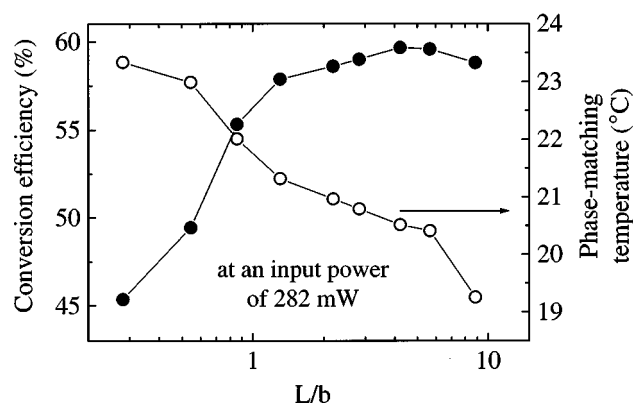
Fig. 4. Optimum phase-matching temperatures and SHG efficiencies versus focusing at an input power of 282 mW and an input wavelength of 860 nm.

We also investigated, for the first time to our knowledge, the focusing dependence of the phase-matching temperature. Figure 4 shows SHG efficiencies and optimum phase-matching temperatures versus focusing at an input power of 282 mW when the fundamental wave is fixed at a wavelength of 860 nm. For weak focusing ($L/b \leq 1$), the conversion efficiency increases with increasing L/b. For tight focusing ($L/b \gg 1$), however, the conversion efficiency is almost independent of focusing. The highest efficiency is achieved for a focal length of 31 mm. For a stronger focusing of 25 mm, however, the SHG efficiency decreases, presumably because of stronger nonlinear absorption (possibly self-phase modulation as a result of the nonlinear refractive index may also play some role[9–12]). The phase-matching temperature decreases as the focusing is tighter. For tighter focusing at high input powers, to maintain phase matching one should decrease the crystal temperature to compensate for the thermal effect that results from nonlinear absorption or blue-induced IR absorption.[17–19] Tighter focusing concentrates the heat generated by the nonlinear absorption or blue-induced IR absorption in a smaller volume of the crystal, leading to a greater optically induced temperature rise in the focal volume and therefore to a lower externally applied phase-matching temperature.
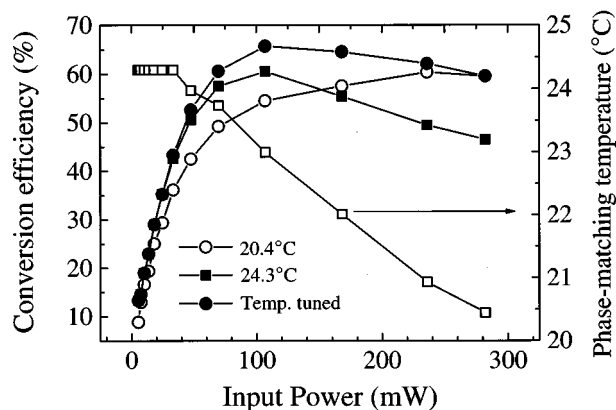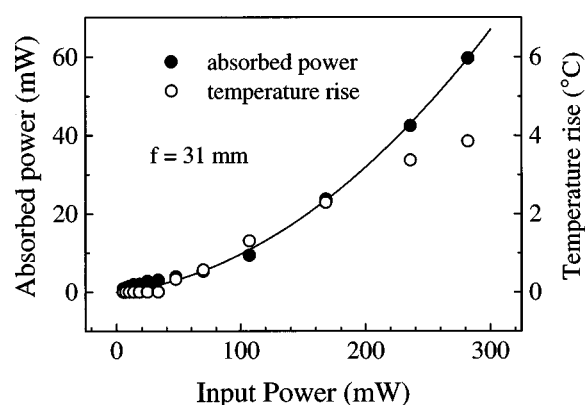

Fig. 2. SHG conversion efficiencies at different crystal temperatures and optimum phase-matching temperatures versus input power for a focal length of 31 mm.

Figure 5 shows the temperature dependence of the SHG efficiency at an input power of 282 mW and a fundamental wavelength of 860 nm for focal lengths of 31 and 80 mm. The phase-matching temperature dependence for both focal lengths is very broad, with a FWHM of ~10 °C. This is in sharp contrast to the cw case, for which a temperature FWHM[3,8,20] of 0.3–0.5 °C and an ~0.3-nm/°C shift[2,8] in the phase-matched SH wavelength were reported. For the cw case we can calculate the temperature FWHM from Eq. (6). For a 3-mm-thick $KNbO_3$ crystal with a GVM α of 1.2 ps/mm, the SH bandwidth $\Delta\lambda$ is calculated to be 0.15 nm, and thus the temperature
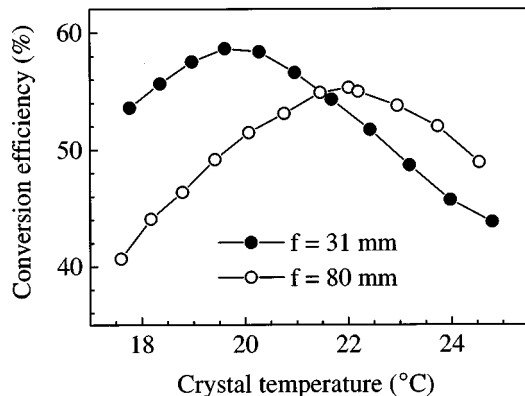

Fig. 5. Temperature dependence of the SHG efficiency at an input power of 282 mW for focal lengths of 31 and 80 mm.
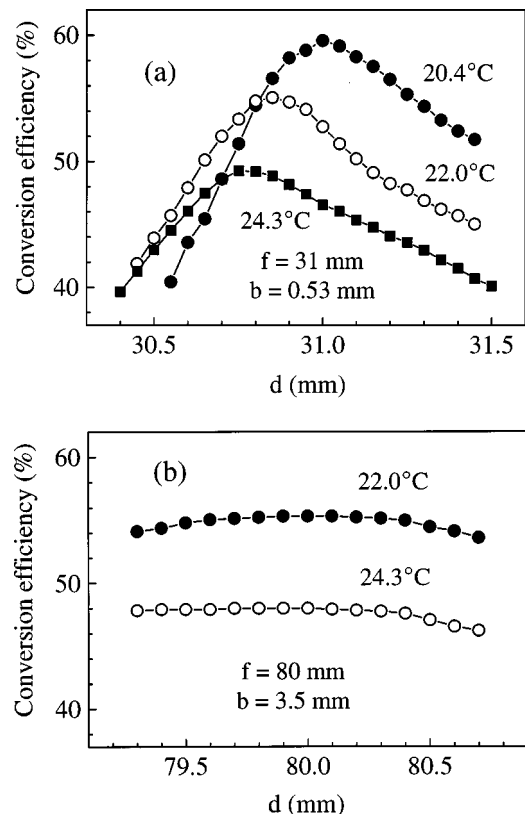

Fig. 6. SHG efficiencies versus spacing d between the focusing lens and the crystal at an input power of 282 mW for focal lengths of (a) 31 and (b) 80 mm. The experiments were performed at an input wavelength of 860 nm.

FWHM is estimated to be 0.5 °C (0.15 nm ÷ 0.3 nm/°C). But our fundamental femtosecond pulse has a spectrum of ~8 nm, which drives nonlinear polarization within a bandwidth $\Delta\lambda_{2\omega}$, which is given by $\Delta\lambda_{2\omega} \approx (\sqrt{2}/4)\Delta\lambda_\omega$. (The factor 1/4 arises because $\Delta\lambda = (\lambda^2/c)\Delta f$; the factor $\sqrt{2}$ arises because the duration of the nonlinear polarization is roughly $\sqrt{2}$ shorter than the input intensity, leading to a corresponding increase in Δf.) Therefore the bandwidth $\Delta\lambda_{2\omega}$ of the SHG nonlinear polarization is roughly 3 nm, which explains the observed ~10 °C temperature FWHM ($\approx$3 nm ÷ 0.3 nm/°C) in Fig. 5. The temperature peaks in Fig. 5 shift by ~2 °C with different focusing conditions. The broad width of the curves in Fig. 5, which is due to the wide femtosecond input spectrum, significantly reduces the sensitivity to temperature rise compared with that in the cw case; however, important effects are still evident.

As was pointed out in the previous cw experiments,[4,17–19] SH intensity may vary with position in the crystal because of the lateral and longitudinal temperature variations that result from nonlinear absorption or blue-induced IR absorption. Here we have experimentally demonstrated that for tighter focusing the conversion efficiency depends on the focal position in the crystal, which is most likely related to a spatially varying index of refraction associated with a temperature variation.

Figure 6 shows the SHG efficiencies versus spacing d between the focusing lens and the crystal for focal lengths of 31 and 80 mm at an input power of 282 mW. For a focal length of 31 mm and a crystal temperature of 20.4 °C, the highest conversion efficiency is achieved when the beam is focused near the center of the NLC. However, the peak position of the conversion efficiency for a focal length of 31 mm shifts toward the back side of the crystal at a rate of ~0.06 mm/°C as the externally applied crystal temperature increases, whereas for a focal length of 80 mm it is almost unchanged. For focal lengths of 31 and 80 mm, the depths of focus b are calculated to be 0.53 and 3.5 mm and L/b values are 5.7 and 0.86, respectively. Therefore, for a focal length of 31 mm, the depth of focus b is much smaller than the crystal length, a case of tight focusing. Moreover, the beam size changes appreciably within the crystal and thus the conversion efficiency is changed with the crystal position. On the other hand, for a focal length of 80 mm, the depth of focus b is comparable with the crystal length, a case of weak focusing. The beam size changes only slowly within the crystal, and thus the conversion efficiency is almost unchanged for small displacements of the crystal.

4. CONCLUSION

We have observed power-dependent temperature shifts in the phase-matching curves for femtosecond SHG in a thick $KNbO_3$ crystal. The temperature shifts are closely related to an induced or nonlinear absorption process and must be taken into account in optimizing the phase-matching conditions. We found that the SHG efficiency is increased when the phase-matching temperature is reset at each input power. We obtained a maximum SHG efficiency of 66% at an input power of 107 mW and a slope efficiency of ~1.5%/mW at low input powers for a focal

length of 31 mm. In addition, the temperature-dependent SHG efficiency and the input-power-dependent phase-matching temperature in our femtosecond experiments are significantly different from those in the cw case. We found that the optimum phase-matching temperature depends significantly not only on the input power but also on the focusing, which is attributed to the heating of the crystal that results from the nonlinear absorption. We also observed that for tighter focusing the peak of the conversion efficiency is shifted toward the back side of the crystal as the crystal temperature increases. Our results provide important information on optimizing the SHG efficiency in frequency-doubling experiments with intense and broad-bandwidth femtosecond pulses.

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