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HIGHLY EFFICIENT THIRD HARMONIC GENERATION BY MEANS OF FOUR-WAVE DIFFERENCE-FREQUENCY MIXING IN FUSED SILICA

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We report on highly efficient third harmonic generation of 1-ps laser pulses in transparent isotropic solid state medium through phase-matched non-collinear four-wave difference-frequency mixing. Third harmonic pulses at 351 nm with 230 μJ energy and 15% energy conversion efficiency were generated in 3-mm-thick UV-grade fused silica sample by use of non-collinear phase matching, cylindrical beam focusing geometry, and pulse-front tilting.

Keywords: third harmonic generation, four-wave mixing

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1. Introduction

Third harmonic (TH) generation in isotropic media is widely used for femtosecond pulse frequency conversion in the ultraviolet spectral range. In a medium with $\chi^{(3)}$ nonlinearity there are two four-wave mixing (FWM) configurations, which may lead to generation of radiation at 3ω. The first approach considers direct frequency tripling, that is $3\omega = \omega + \omega + \omega$. The phase matching condition could be fulfilled only in media with anomalous dispersion, e. g. metal vapours [1], however the overall process is inefficient due to high absorption. On the other hand, in transparent media with normal dispersion, the phase-mismatch could be greatly reduced by means of tight beam focusing and occurrence of non-perturbative effects at high intensity limit [2, 3]. In this case, however, energy conversion process is limited by low intrinsic nonlinearity of gaseous media and short interaction length. The efficiency of TH generation could be improved by use of high pressure gases [4] or by the guided wave propagation in the filamentation regime [3, 5], but still does not exceed 1%. In condensed media, TH generation efficiency in the filamentation regime still remains remarkably low (10^{-6}) due to severe phase-matching limitations [6]. The second approach is based on the four-wave difference-frequency mixing ($3\omega = 2\omega + 2\omega - \omega$) and is more flexible in terms of the interaction geometry. Using this approach, femtosecond TH pulses were generated in a noble-gas-filled capillary waveguide [7] and in an extended light filament generated in noble gasses at atmospheric pressure [8] with improved energy conversion efficiency of 2–4%. The use of cascaded four-wave mixing processes enables further frequency conversion to fourth and fifth laser harmonics [9]. The perfect phase matching may be achieved by setting a non-collinear interaction geometry, for instance, by coupling of Bessel and Gaussian beams at fundamental and second harmonics, respectively [10]. In a condensed medium, enhanced TH generation in water by the use of elliptical light beams was reported [11].

The limiting factor for efficient TH generation in a solid state media is the optical damage. Therefore cylindrical beam focusing enables to scale the energy of the interacting beams keeping the overall fluence below the damage threshold, as demonstrated in recent four-wave parametric amplification experiments [12–14]. In this paper we report on highly efficient TH generation in fused silica with milijoule pumping, using non-collinear phase matching, cylindrical beam focusing, and pulse-front tilting of one of the pump pulses.

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2. Experimental results

The experiment was performed with a Nd:glass laser system (*Twinkle*, Light Conversion Ltd.), which delivered 1-ps, 1054-nm pulses at 10 Hz repetition rate. The laser output was split into two parts. The first portion of the laser radiation was frequency doubled and then recombined with a fresh fundamental harmonic so as to produce TH through the four-wave difference-frequency mixing process, i. e. $3\omega = 2\omega + 2\omega - \omega$. Co-polarized pump beams (fundamental and second harmonics) were focused into the UV-grade fused silica sample (KU-1) using cylindrical lenses of $f_x = \infty$, $f_y = +500$ mm and $f_x = \infty$, $f_y = +750$ mm respectively, so as to achieve mode matching. The FWHM size of both pump beams on the input face of the fused silica sample was measured as 5 mm × 50 μm. The fundamental and second harmonic beams were crossed in the plane containing the major axes of the ellipses in order to ensure good spatial overlap along the entire sample length. The crossing angle was found from the phase-matching condition, which is described by a vector equation $\mathbf{k}_{3\omega} = 2\mathbf{k}_{2\omega} - \mathbf{k}_{\omega}$, where $|\mathbf{k}(w)| = n(w)\, w/c$ is the wave number. After simple algebra, one finds the (internal) phase matching angle θ between the fundamental and second harmonic beams:

$$\cos\theta = \frac{4k_{2\omega}^2 + k_{\omega}^2 - k_{3\omega}^2}{4k_{\omega}k_{2\omega}}, \qquad (1)$$

which yields $\theta = 11^\circ$ under our particular experimental settings.

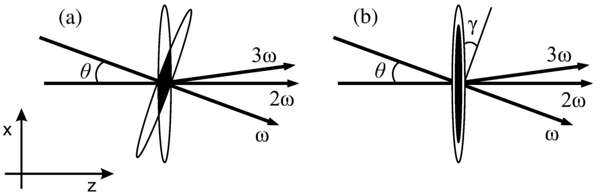

Fig. 1. Schematic representation of pump pulse overlap (indicated by black area) with (a) untilted pulses, (b) tilted-front fundamental harmonic pulse.

Since the elliptical pump beams carry ultrashort light pulses (1-ps pulse occupies only a narrow width of ≈200 μm along the z axis in fused silica), large beam crossing angle results in noticeable reduction of the beam/pulse physical overlap area in the nonlinear medium, as illustrated in Fig. 1(a), which in turn greatly reduces the dimension of the generated TH beam along the x axis as shown in Fig. 2. We have improved the overlap geometry by introducing the pulse-front tilt on the fundamental harmonic pulse, as illustrated in Fig. 1(b). The pulse-front tilt angle $\gamma = 8.6^\circ$ was produced by means of accomplishing four passes through the TF5 glass prism with an apex angle of 60° at the minimum deviation geometry. Further we refer to the above configurations as untilted and tilted pulse cases, respectively. The effect of increased spatial overlap due to pulse-front tilt on the TH beam dimension is illustrated in Fig. 2, where TH beam intensity profiles in the case of untilted and tilted pulses are compared. Improvement of the interaction geometry via pulse-front tilting has led to a substantial improvement of the TH pulse energy as well.

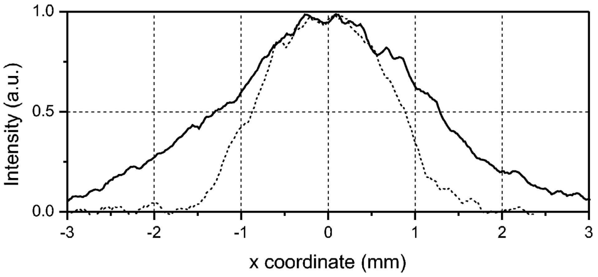

Fig. 2. Measured central cross-sections of TH beams along x axis in the case of untilted (dashed curve) and tilted (solid curve) pulses.

Figure 3 compares the TH pulse energy dependence measured versus pump energy E_{pump} with untilted and tilted pulses in 3-mm-thick fused silica sample. The pump energy here is defined as $E_{\mathrm{pump}} = E_{\omega} + E_{2\omega}$, with fixed energy ratio between the pump pulses that was set at $E_{\omega}/E_{2\omega} = 1/4$ according to Manley–Rowe relation. The highest value of applicable E_{pump} was limited by the threshold for the optical damage of fused silica. With tilted pulses, the highest TH pulse energy of $E_{3\omega} = 230$ μJ is measured at $E_{\mathrm{pump}} = 1.55$ mJ, suggesting as high as 15% energy conversion efficiency from the pump. Besides the improvement in the interaction geometry, tilting of one of the pulses leads to a better group velocity matching along the propagation axis (z axis) for non-collinearly propagating ultrashort pump pulses, which might be important for frequency conversion using femtosecond light pulses.

Figure 4 plots the energy conversion efficiency, $\eta = E_{3\omega}/(E_{\omega} + E_{2\omega})$, versus pump energy, measured in fused silica samples of different thickness (3, 5, and 8 mm). The measured curves indicate that the TH conversion efficiency saturates at nearly the same $\eta \approx 15\%$ in all samples, however, at different pump energy as a result of highly nonlinear propagation dynamics.

And finally, we have measured the focusability of the TH beam generated in 3-mm-thick fused silica sample. For many practical applications the beams with

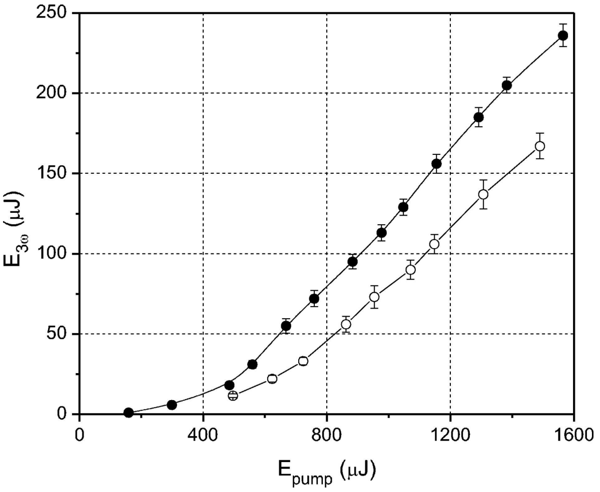

Fig. 3. Third harmonic pulse energy $E_{3\omega}$ versus pump energy E_{pump} measured in 3-mm-thick fused silica sample with untilted (open circles) and tilted (solid circles) pulses.

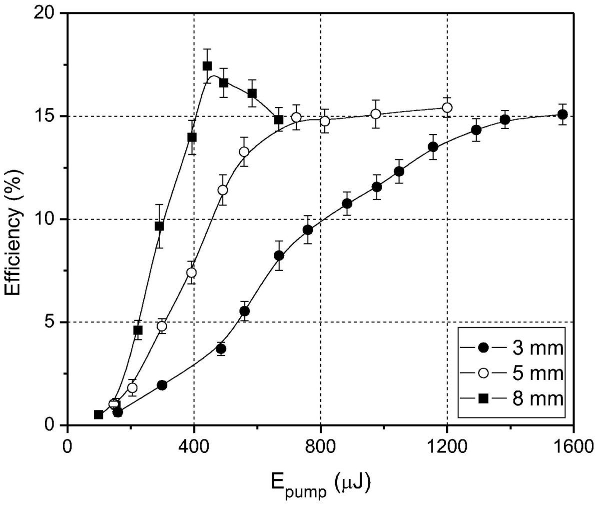

Fig. 4. TH energy conversion efficiency versus pump energy measured in 3, 5, and 8 mm thick fused silica samples.

circular symmetry are required, therefore the circular symmetry of the elliptical TH beam was restored using $f_y = +100$ mm cylindrical lens. Figure 5 shows the measured TH beam intensity profile and its relevant cross-sections at the focal plane of $f = +1$ m spherical lens. The focused TH beam exhibits good focusability and the estimated beam quality is ≈1.5 times of the diffraction limited Gaussian beam.

3. Conclusion

In conclusion, we have experimentally demonstrated efficient third harmonic generation of 1-ps Nd:glass laser pulses through four-wave difference-frequency mixing in fused silica with milijoule pumping. TH pulses with energy as high as 230 μJ with 15% energy conversion efficiency were obtained using non-collinear phase matching combined with cylindrical beam focusing geometry. We also have shown that under these geometrical settings, the efficiency of the TH generation process is notably improved by suitable pulse-front tilting of one of the pump pulses. The proposed set-up could be readily implemented for the frequency conversion of the femtosecond light pulses and for the generation of higher order harmonics in deep-UV and VUV-transparent solids, in particular.

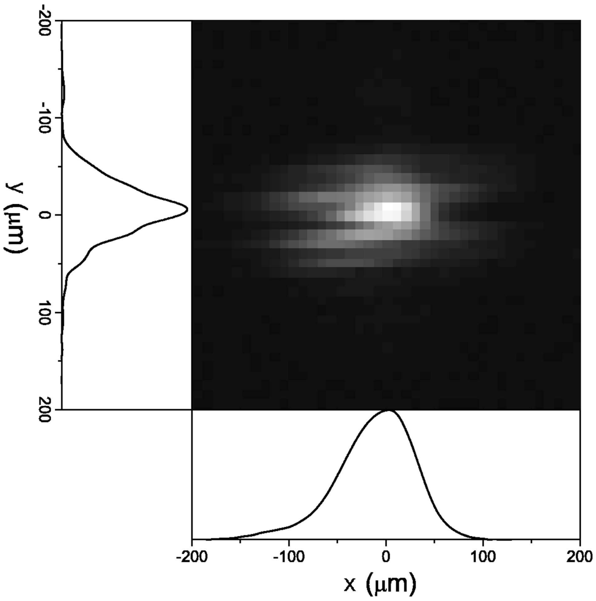

Fig. 5. Intensity distribution of the TH beam and its central cross-sections along the principal axes at the focal plane of an $f = +1$ m spherical lens.

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NAŠUS TREČIOSIOS HARMONIKOS GENERAVIMAS NAUDOJANT KETURBANGĮ SKIRTUMINIO DAŽNIO MAIŠYMĄ LYDYTAME KVARCE

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Santrauka

Pademonstruotas efektyvus 1 ps trukmės šviesos impulsų trečiosios harmonikos generavimas, naudojant faziškai sinchronizuotą keturbangį skirtuminio dažnio maišymą skaidrioje izotropinėje kietakūnėje terpėje. Taikant nekolinearų fazinį sinchronizmą, cilindrinį pluoštų fokusavimą bei impulso fronto pokrypį, 3 mm storio lydyto kvarco plokštelėje ties 351 nm bangos ilgiu generuoti trečiosios harmonikos impulsai, kurių energija siekė 230 μJ, o energijos keitimo efektyvumas – 15%.