Phase-Matched Optical Parametric Conversion of Ultrashort Pulses in a Hollow Waveguide

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Abstract. We demonstrate for the first time nonresonant phase-matched frequency conversion of ultrashort pulses in gases. Broad-bandwidth ultrafast pulses, tunable around 270nm, were generated from a Ti:sapphire amplifier system using 2ω + 2ω - ω parametric wave mixing in a capillary waveguide. Both the fundamental and the second-harmonic light were coupled into the lowest-order (EH_{11}) mode. The output pulses have an energy >4μJ at a 1kHz repetition rate, in the EH_{11} spatial mode. This method can be made to generate 10-20fs pulses, and is the first phase-matching technique which is applicable to frequency conversion into the deep- and vacuum-ultraviolet regions of the spectrum.

INTRODUCTION

In recent years there has been much progress in developing reliable sources of ultrashort-pulse light. Most of these new lasers are based on titanium-doped sapphire, and thus operate in the near infrared.[1] However, many applications of these light sources require photons in other wavelength ranges. For example, many studies of molecular dynamics require light in the deep-ultraviolet or even the vacuum ultraviolet. Ultrashort pulses in the infrared can be converted to the UV using nonlinear-frequency conversion in crystals such as BBO,[2] and parametric generation can be used to generate ultrashort pulses tunable from the near-UV into the mid-infrared. However, these techniques suffer from limitations inherent to the propagation of ultrashort light pulses in solid-density materials: opacity in the VUV, and very high group-velocity walk off which makes it difficult to generate pulses in the UV much shorter than a couple of hundred femtoseconds.

In this work, we present an entirely new method for the efficient, phase-matched conversion of ultrashort pulses into the UV, which is equally applicable in the VUV and possibly even in the XUV region of the spectrum.[3] In this technique, guided-wave optical parametric generation, the nonlinear-medium is a gas (in this case argon). Since many gasses are transparent from the VUV into the far-infrared, the potential tuning range of this technique is much larger than in the case of crystal conversion. Furthermore, dispersion in the gasses is very low, making it a technique suitable for the frequency conversion of extremely short (<10 fs) pulses.

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THEORY

Frequency conversion in crystals typically relies on the use of $\chi^{(2)}$ materials to obtain high nonlinearity, and on birefringence to phase match propagation vectors of the fundamental and the harmonic. Since a gas is homogeneous and isotropic with respect to light propagation, neither of these concepts is applicable in gasses. $\chi^{(3)}$ or higher odd-order nonlinearities must be used, making it essential to increase the interaction length to make-up for the lower magnitude of the nonlinearity. Phase-matching in free-space propagation does not normally occur except in special cases near atomic resonances. [4,5]

In this work, we show theoretically and demonstrate experimentally that phase-matched conversion with increased interaction length can be accomplished using propagation of the light in a waveguide rather than in free space. Recent work has demonstrated the utility of hollow-core fibers (simple gas-filled capillary tubes) for self-phase modulation and compression of ultrashort pulses.[6] Propagation of light in a waveguide introduces a negative phase shift on the wave to counter the tendency of the wavefront to move forward due to diffraction. This phase shift is greater for longer wavelengths, resulting in a net negative dispersion in the hollow waveguide; i.e. a phase velocity that is inversely proportional to wavelength. This negative dispersion can be counterbalanced by the positive dispersion of the gas filling the center of the waveguide, resulting in a value of pressure at which dispersion at a particular wavelength vanishes.

Propagation in an optical waveguide offers several options for phase matching parametric processes, such as mixing of light propagating at different frequencies and/or in different spatial modes.[7,8] The modal properties of a dielectric capillary were given by Marcateli et al.[9] The propagation constant for a capillary filled with a medium of index n_g is given by -

$$k=\frac{2\pi n_g(\lambda)}{\lambda}\left[1-\frac{1}{2}\left(\frac{u_{nm}\lambda}{2\pi a}\right)^2\left(1+\mathrm{Im}\left(\frac{v_{EH}\lambda}{\pi a}\right)\right)\right] \qquad (1)$$

where a is the core radius, u_{nm} is the modal constant, $v_{EH} = 1/2\ (v^2 + 1)/((v^2 - 1)^{0.5})$, and v is the ratio of the refractive index of the capillary material to that of its contents. Although $|v| > 1$, Fresnel reflections at the gas-glass interface allow lossy guiding of optical beams. The field loss rate for hybrid modes (EH_{nm}) is given by -

$$\alpha=\left(\frac{u_{nm}\lambda}{2\pi}\right)^2\frac{\lambda^2}{a^3}\mathrm{Re}(v_{EH})\ . \qquad (2)$$

Consider the case of difference frequency mixing between two beams propagating in an optical fiber at central frequencies ω_1 and ω_2, with modal constants u_1 and u2. The phase mismatch Δk in the case of difference frequency mixing, $\omega_3 = N\omega_2 - M\omega_1$, where $\omega_2 = R\omega_1$, $\Delta k = Nk_2 - Mk_1 - k_3$, may be written as -

$$\begin{aligned}\Delta k &= \Delta k_{\mathrm{mode}} - \Delta k_{material} \\ &= \frac{\lambda_1}{4\pi a^2}\left[\frac{u_3^3}{RN-M} + Mu_1^2 - \frac{Nu_2^2}{R}\right] - \frac{2\pi p}{\lambda_1}((RN-M)\delta_3 + M\delta_1 - N\delta_2)\end{aligned} \tag{3}$$

where the index is written as $n_g = 1 + p\delta_g$, with p the gas pressure. The phase mismatch is a balance of modal dispersion ($\Delta k_{mode} \propto 1/\pi a^2$) material dispersion ($\Delta k_{material} \propto p$). In gases (or plasmas) with normal dispersion, $\Delta k_{mat}>0$. If the modes of the pump and signal are chosen such that $\Delta k_{mode}>0$, there will exist a pressure for which the process is phase-matched. For the difference frequency-mixing case where two 400 nm photons are combined and one 800 nm photon is given-up, N=2, M=1, and $u_1 = u_2$,. Therefore $\Delta k_{mode} > 0$ for any signal mode. In the present experiment, R = 2, and any pair of modes ($u_1 = u_2$) can be phase matched to a particular mode, u_3. It is most desirable, and gives the largest model overlap and nonlinearity, when the TEM_{00} free-space mode is coupled into the EH_{11} mode of the waveguide.

EXPERIMENT

In the experiment, a laser system generating 20 femtosecond pulses with up to 4 mJ energy at 1 kHz repetition-rate was used.[10] A 100 μm LBO crystal was used to frequency double the 800 nm light, with 20% conversion efficiency. The two colors were then separated, and later recombined with a relative time delay, before being focused into the capillary (127 μm core diameter, 60 cm long). The divergence of the input telescope was adjusted to optimize the coupling of the 400 nm light (35% throughput); the fundamental beam was not as well optimized (<10% throughput). Figure 1 shows the 3ω output energy versus argon gas pressure. In the case of krypton gas, the output vs. pressure is has a similar structure, but is shifted to lower gas pressure due to the higher dispersion of krypton. The positions of peaks in conversion efficiency are in excellent agreement with the values calculated from Eqn. 3 for conversion into various modes. The highest conversion efficiency is observed into the EH_{11} mode, at a measured and calculated optimum pressure of 85.1 and 89.7 Torr, respectively. The output mode is very nearly Gaussian. With 30 μJ at 2ω and 64 μJ at ω leaving the fiber, the output energy at 3ω was 4 μJ, giving 13% conversion from 2ω to 3ω.

Figure 2 shows the spectrum of the output under two conditions. Calculated phase-matching bandwidths (>20 nm) and values of group-velocity walk off (<10 fs) lead one to expect excellent characteristics for the frequency conversion of ultrashort pulses, as is observed. Figure 2(a) is the case of moderate intensity of the 800 nm idler field. The spectral bandwidth, which has a bandwidth corresponding to a 12 fs pulse, is consistent with what one would expect given the input bandwidths and a perturbative frequency-conversion process using $\chi^{(2)}$ in LBO and $\chi^{(3)}$ in argon. Figure 2(b) is the case where the intensity of the 800 nm idler light is increased to a level similar to that used in hollow-core fiber pulse compression experiments.[6] In this case, cross-phase modulation broadens the spectrum of the UV light to the point where the bandwidth could support a 4 fs pulse.

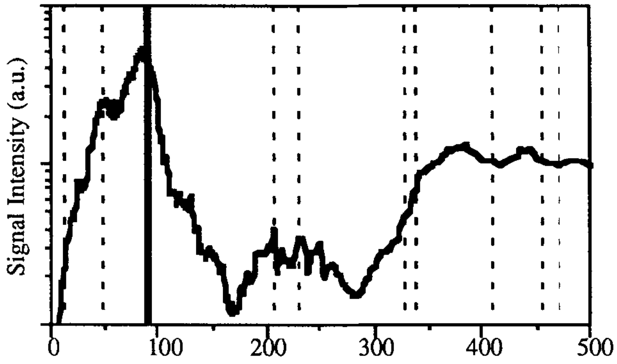

FIGURE 1: Signal intensity of third harmonic of 800 nm, as a function of argon gas pressure within a 127 μm diameter, 60 nm long argon-filled capillary waveguide. The solid line represents the calculated pressure for phase-matched conversion into the EH_{11} mode, while the dashed line represents phase-matching pressures corresponding to high-order modes.

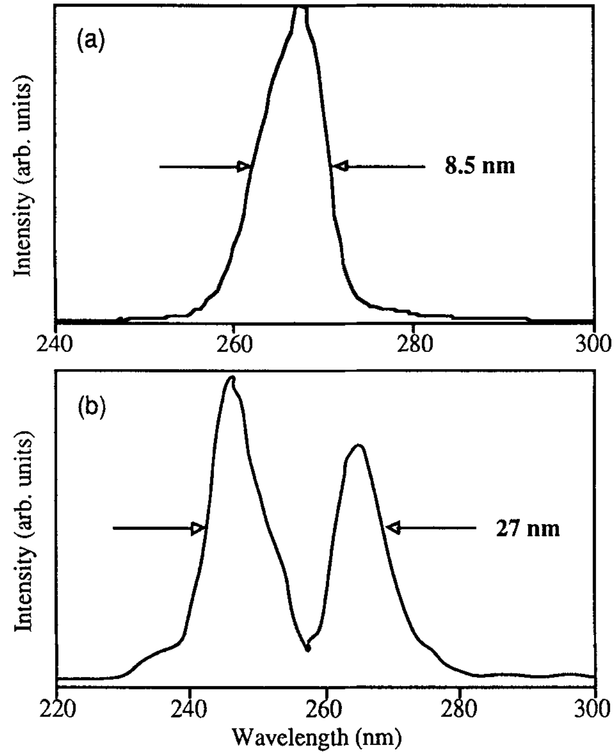

FIGURE 2: Output pulse spectrum (a) in the case of moderate intensity of the 800 nm "idler" pulse, showing a spectrum corresponding to a 12 fs pulse. (b) when the intensity of the 800 nm light is increased, cross-phase modulation of the UV by the nonlinearity caused by the 800 nm pulse broadens the spectrum of the output. The bandwidth could be compressed to ~4 fs duration.

In conclusion, we have demonstrated a new technique for efficient, phase-matched frequency upconversion of light. This is the first phase-matched conversion technique suitable for the generation of ultrafast pulses in the deep-UV, VUV, and possibly even the soft x-ray regions of the spectrum. This technique also lends itself well to the implementation of a tunable ultrafast UV source, with the use of tunable infrared from an optical parametric amplifier as the idler source.

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