Generation and characterization of few-cycle mid-infrared pulses from 2800 to 4600 nm

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Abstract: Sub-five-cycle pulses in the spectral region from 2800 to 4600 nm are generated using a potassium niobate optical parametric amplifier employing a germanium pulse compression scheme. The sub-65-fs mid-infrared pulses are fully characterized by XFROG.

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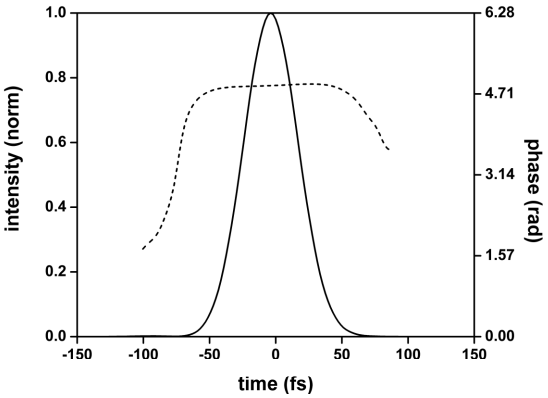
Generation of ultrashort pulses in the mid-infrared (mid-IR) spectral region is a goal that has been pursued intensely, with the major interest being the extension of traditional nonlinear optical measurements to the study of molecular vibrations. While much activity has focused on the region of carbonyl stretching vibrations near 5000 nm [1], the 3000 to 4000 nm wavelength range is of great importance due to the hydrogen-bond-sensitive O-H and N-H stretching modes in this region. While several groups have studied the dynamics of the O-H stretching vibration in isoptopically-diluted water, the pulses used in these experiments have been >100 fs; improved time-resolution is needed to resolve the dephasing of these broad transitions. Furthermore, KTP-based OPA schemes commonly used for mid-IR pulse generation typically have been limited to the region near 3000 nm, requiring the addition of new parametric amplification stages to study the deuterated analogs of these vibrations [2]. The availability of a sub-100-fs source of pulses spanning the full 3000 to 4000 nm region therefore would be a great advance toward the study of hydride stretching dynamics in a wealth of samples, particularly aqueous systems.

Potassium niobate ($KNbO_3$) has shown promise as a nonlinear crystal for ultrashort mid-IR pulse generation due to its large second-order nonlinearity, transparency beyond 4000 nm, and broad phase matching bandwidth for a pump wavelengths near the Ti:sapphire fundamental. However, previous mid-IR generation from this crystal has shown that while pulses generated have broad spectral bandwidths, they are typically not transform limited. Combining a short-pulse seeded $KNbO_3$ OPA design with a mid-IR pulse compression scheme should allow the generation of sub-100 fs mid-IR pulses from 3000 nm to beyond 4000 nm. This paper presents a $KNbO_3$ optical parametric amplifier (OPA) that achieves this goal. The OPA generates mid-IR pulses containing five or fewer optical cycles with center wavelengths from 2800 nm to 4600 nm in a single crystal device. These pulses are suitable for probing the ultrafast vibrational responses of X-H and X-D modes (X = C, N, O). The pulses are fully characterized in amplitude and phase using cross-correlation frequency-resolved optical gating (XFROG) [3].

The OPA was pumped with 800 nm light from a home-built regenerative Ti:sapphire amplifier producing pulses ~ 42 fs in duration. Near-IR signal pulses were obtained from a white light continuum generated in 2 mm of [0001]-cut sapphire. The pump and near-IR signal were combined with a dichroic mirror for collinear type-I optical parametric amplification in the xz-plane of a 1 mm $KNbO_3$ crystal (VLOC; $\theta = 40.5°$off b toward c, $\phi = 0°$). The mid-IR output of the OPA was tuned from 2800 to 4600 nm by adjusting the temporal overlap of the pump and signal pulses; minor tuning of the θ-angle was also required. The design is similar to that of Kafka and Watts [4] and Cussat-Blanc, et al. [5], but uses shorter pump pulses and a thinner OPA crystal.

Mid-IR pulses were characterized by sum-frequency generation (SFG) cross-correlation frequency-resolved optical gating (XFROG). The SFG cross-correlation was generated in a thin (100 or 300 μm) $KNbO_3$ crystal cut at $\theta = 37.6°$. The signal was imaged onto the entrance slit of a 0.32 m monochromator, and XFROG spectrograms were obtained using a CCD array to acquire spectra at each delay. Dispersion compensation of the negative group velocity dispersion (GVD) of the $KNbO_3$ crystal and CaF_2 optics was accomplished using germanium disks of varying thicknesses, which have positive GVD in this spectral region. The gate pulse was characterized via second-harmonic generation autocorrelation and FROG. The intensity and phase of the mid-IR pulses were extracted from spectrograms using FROG 3.06 software (Femtosoft Technologies). All traces were corrected for the spectral response of the detection system prior to analysis. The intensity and phase of a mid-IR pulse at 3600 nm is shown in Figure 1. The FWHM duration of this pulse is 48 fs, corresponding to four optical cycles. The performance of the system over the full tuning range explored is given in Figure 2; pulses are shorter than 65 fs in all but one case, equivalent to five or fewer optical cycles per pulse. To our knowledge, these are the shortest pulses reported to date



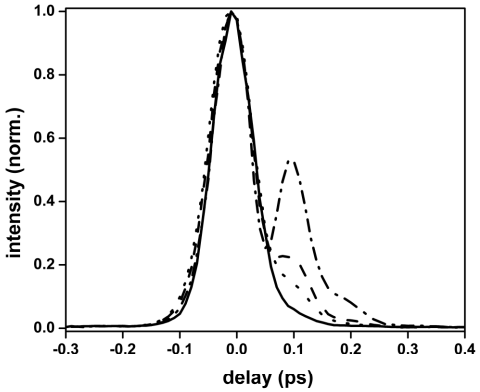


Fig. 1. (Top) Intensity (solid) and phase (dotted) of a four-cycle pulse at 3600 nm (48 fs FWHM).

Fig. 3. (Bottom) Pulse profiles after propagation through HDO:D_2O.

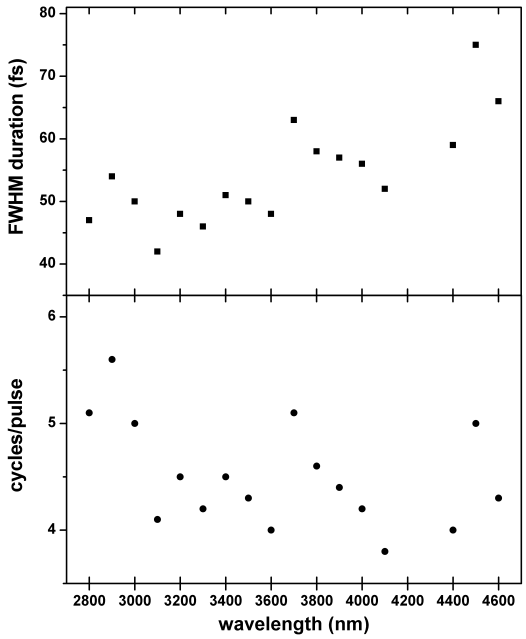


Fig. 2. FWHM duration (top) and cycles/pulse (bottom) of compressed mid-IR pulses.

from a femtosecond mid-IR OPA throughout this spectral range; the duration of these pulses is sufficient to time-resolve the vibrational dephasing of hydrogen-bonded modes in condensed phase systems.

We have used this system and the XFROG technique to examine coherent pulse propagation in optically dense samples of HDO in liquid D_2O [6]. Figure 3 shows the wavelength-integrated profiles of pulses resonant with the OH-stretching vibration of HDO (~ 2950 nm) after traversing samples with optical densities of 0.5 (dotted), 0.9 (dashed), and 1.7 (dot-dashed) at the absorption maximum (solid = instrument response). Severe distortions are observed due to interference between the finite-duration initial pulse and the free-induction decay field that is 180° out of phase with the driving field. These features are chirp-dependent, and the associated phase shifts between them have been determined using XFROG.

[1] M. T. Zanni, S. Gnanakaran, J. Stenger and R. M. Hochstrasser, "Heterodyned two-dimensional infrared spectroscopy of solvent-dependent conformations of acetylproline-NH_2," J. Phys. Chem. B **105,** 6520-6535 (2001).
[2] M. F. Kropman, H.-K. Nienhuys, S. Woutersen and H. J. Bakker, "Vibrational relaxation and hydrogen-bond dynamics of HDO:H_2O," J. Phys. Chem. A **105,** 4622-4626 (2001).
[3] S. Linden, H. Giessen and J. Kuhl, "XFROG - a new method for amplitude and phase characterization of weak ultrashort pulses," Phys. Stat. Sol. B **206,** 119 (1998).
[4] J. D. Kafka and M. L. Watts, "A potassium niobate OPA pumped by an amplified Ti:sapphire laser," in *Ultrafast Phenomena X*, edited by P. F. Barbara, J. G. Fujimoto, W. H. Knox and W. Zinth (Springer-Verlag, 1996), pp. 38-39.
[5] S. Cussat-Blanc, A. Ivanov, D. Lupinski and E. Freysz, "$KTiOPO_4$, $KTiOAsO_4$, and $KNbO_3$ crystals for mid-infrared femtosecond optical parametric amplifiers: analysis and comparison," Appl. Phys. B **70,** S247-S252 (2000).
[6] J. A. Gruetzmacher and N. F. Scherer, "Few-cycle mid-infrared pulse generation, characterization, and propagation," submitted (2002).