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# Generation and multi-octave shaping of mid-infrared intense single-cycle pulses

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## Figures:

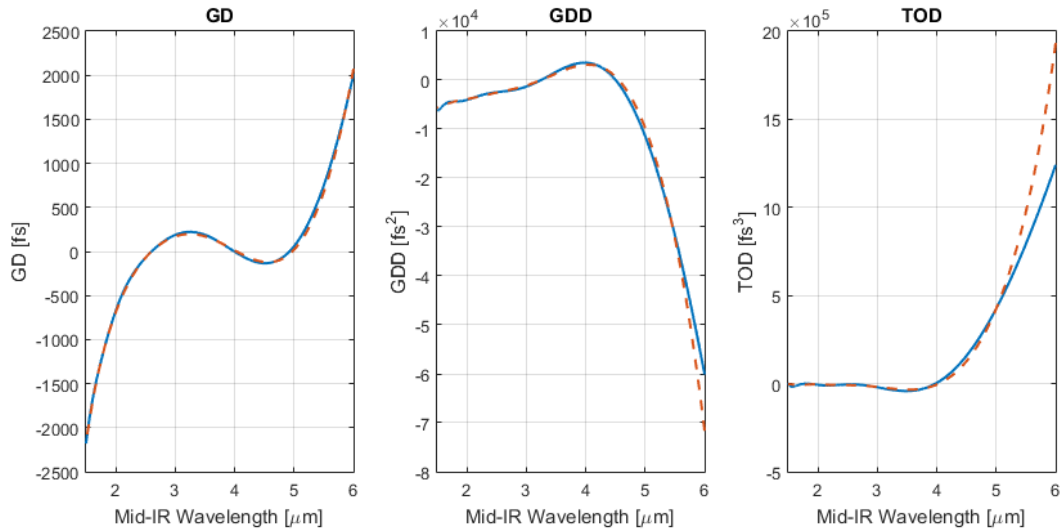


Figure S1: A comparison of the group delay (GD), group delay dispersion (GDD) and third-order dispersion (TOD) produced by the ADFG device as calculated from simulations (solid) and the analytic formula for the delay (dashed).

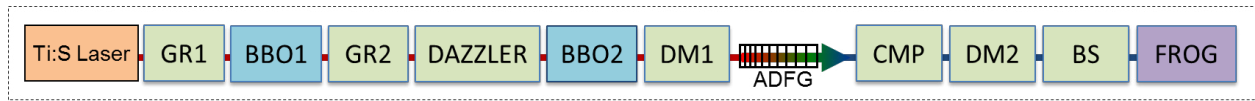


Figure S2: Diagram of Dispersive elements.

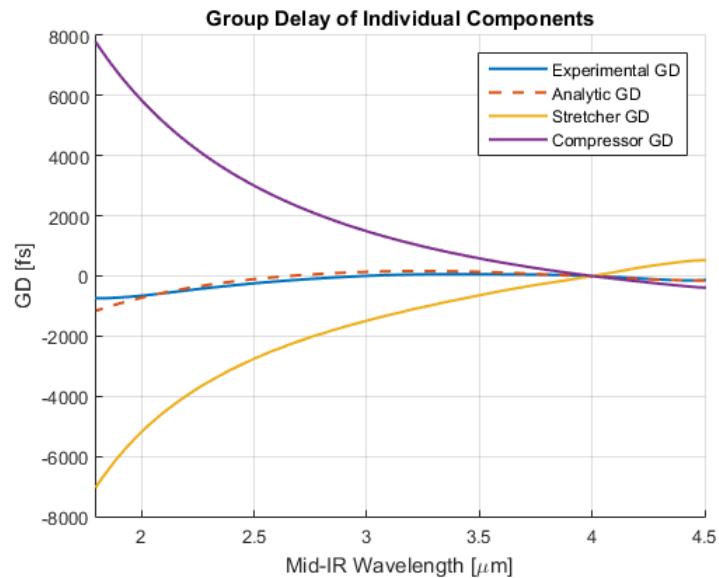


Figure S3: Experimentally and analytically determined group delays, plotted together with the group delays for the stretcher and compressor components.

**Tables:**

Table 1:

Component	Dispersion (GDD)	Dispersion (TOD)	Central Wavelength
Grism 1 (GR1)	-10,200 fs <sup>2</sup>	7,900 fs <sup>3</sup>	780nm
BBO 1 (BBO1)	300 fs <sup>2</sup>	250 fs <sup>3</sup>	780nm
Grism 2 (GR2)	-14,300 fs <sup>2</sup>	10,000 fs <sup>3</sup>	780nm
Dazzler (DAZZLER)	14,500 fs <sup>2</sup>	-24,300 fs <sup>3</sup>	780nm
BBO2 (BBO2)	200 fs <sup>2</sup>	150 fs <sup>3</sup>	780nm
Dichroic Mirror (DM1)	200 fs <sup>2</sup>	200 fs <sup>3</sup>	780nm
ADFG (calculated)	-500 fs <sup>2</sup>	-18,600 fs <sup>3</sup>	3000nm
Compressor (CMP)	10,200 fs <sup>2</sup>	17,400 fs <sup>3</sup>	3000nm
Dichroic Mirror (DM2)	500 fs <sup>2</sup>	900 fs <sup>3</sup>	3000nm
Beam Splitter (BS)	-400 fs <sup>2</sup>	2000 fs <sup>3</sup>	3000nm

**Text:**

Here we present supporting evidence for the analytical dispersion formula for an adiabatic frequency converter presented in the main text. Our analytic expression for the group delay dispersion is empirically verified through comparison to (1) the numerical solutions to our pulse propagation model, and (2) the experimentally measured dispersion of the device. The latter is obtained by evaluation of the dispersion management scheme employed to achieve transform-limited pulse compression of the generated mid-IR pulses; we experimentally determine the group delay dispersion of the ADFG device by calculating the difference in the total group delays accumulated in optical components coming before and after it.

For a sufficiently long crystal (compared to the conversion length of the ADFG process), we find the group delay (GD) imparted on the final mid-IR field is comprised of two terms:

$$\tau(\omega) = k'(\omega + \omega_p)z_c(\omega) + k'(\omega)(L - z_c(\omega))$$

Where  $k(\omega + \omega_p)$  and  $k(\omega)$  are the near-IR and corresponding mid-IR wavenumbers,  $k'(\omega)$  is  $\partial k/\partial\omega$ ,  $z_c(\omega)$  is the frequency dependent conversion position, and  $L$  is the length of the crystal. The first term represents the propagation of the near-IR field until it is converted at  $z_c(\omega)$ , while the second term represents propagation as a mid-IR field through the remaining length of the crystal. We can take successive derivatives to obtain higher order dispersion terms such as the group delay dispersion,

$$\tau'(\omega) = k''(\omega + \omega_p)z_c(\omega) + k''(\omega)(L - z_c(\omega)) + (k'(\omega + \omega_p) - k'(\omega))z'_c(\omega),$$

where  $k''(\omega)$  is  $\partial^2 k/\partial\omega^2$ , and  $z'_c(\omega)$  is  $\partial z_c/\partial\omega$ . We note the unusual final term that is a consequence of the frequency dependent conversion position.

We define the frequency dependent conversion position,  $z_c(\omega)$ , as the position of zero phase mismatch for each frequency  $\omega$ . To calculate  $z_c(\omega)$  we start by determining the QPM grating periods  $\Lambda(\omega)$  for perfect phase matching, *i.e.*, such that  $\Delta k_{eff}(\omega) = k(\omega + \omega_p) - k(\omega) - k(\omega_p) - \frac{2\pi}{\Lambda(\omega)} = 0$ . By comparing  $\Lambda(\omega)$  to the actual grating period of the crystal, we can determine the position of zero wavevector mismatch for each frequency  $\omega$ , and, hence, the conversion position  $z_c(\omega)$ .

Given this definition of  $z_c$ , the physical interpretation of the accumulated GD is that a near-IR device of frequency dependent length  $z_c(\omega)$  is followed by a mid-IR device of frequency dependent length  $[L - z_c(\omega)]$ , with instantaneous frequency conversion at the boundary point. To test the validity of the analytical formula for  $\tau(\omega)$  and the derived group delay dispersion (GDD) and third order dispersion (TOD), we compared these analytical quantities against the same quantities calculated by numerical solution of the coupled wave equations with pulse propagation, using a split-step method to propagate through the ADFG device (see *Methods*). Fig. S1 shows an excellent match between the simulated and analytic

group delays between 1.6–6  $\mu\text{m}$ , and very strong match for the GDD and TOD, with deviations beginning above 5  $\mu\text{m}$ . Note, when plotting group delay we have subtracted off a constant (*i.e.*, wavelength independent) value for ease of comparison across the bandwidth.

The experimental group delay imparted by the ADFG device was calculated by taking the difference of the GD prior to it (including the stretcher, pulse shaper, and OPA dispersive components) and after it (*i.e.*, imparted by the compressor). The dispersive components of the experiment are shown in Fig. S2 and are tabulated in Table 1.

Figure S3 shows a comparison between the experimentally and analytically determined relative group delays of the ADFG device. We plot these quantities for the full wavelength range, 1.8–4.5  $\mu\text{m}$ , over which the FROG trace of Fig. 3 in the main text demonstrates a flat spectral phase. We find a slight mismatch between the two, which is greatest at 1.8  $\mu\text{m}$  (500 fs). Since the experimental GD is determined by calculating the difference between stretcher and compressor delays, we expect its experimental error to be the uncertainties in the stretcher and compressor delays added in quadrature. Because the stretcher and compressor delays are opposite in sign and very close in magnitude, covering a spread of 15,000 fs at 1.8  $\mu\text{m}$ , a 500 fs error in the determined ADFG device group delay would be caused by errors in the stretcher and compressor delays of only 5%. Thus, this comparison constitutes an experimental confirmation of the analytic expression within reasonable measurement error.