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Rovibrational Wave-Packet Dispersion during Femtosecond Laser Filamentation in Air JOHANAN ODHNER, DMITRI A. ROMANOV, ROBERT J. LEVIS, Department of Chemistry, Department of Physics, and Center for Advanced Photonics Research, Temple University, Philadelphia, PA 19122 — An impulsive, femtosecond filament-based Raman technique producing high quality Raman spectra over a broad spectral range (1554.7–4155 cm) is presented. The filamentation dynamics of a loosely focused high-energy femtosecond pulse in air shortens the pulse, leading to impulsive excitation of all vibrational and rotational Raman-active modes present in the filament channel. For particular Raman modes, impulsive excitation requires pulse duration on the order of the characteristic oscillation periods of the mode, thus revealing the degree of pulse-shortening. The temperature of gas-phase molecules can be recovered by measuring the dispersion of the impulsively excited vibrational wave-packets using a delayed narrow-band pulse. This technique reveals that the initial rovibrational temperature in the filament is 300 K for both N_2 and O_2. The temperature-dependent wave-packet dynamics are interpreted using an analytic anharmonic oscillator model. The wave-packets reveal a 1/e dispersion time of 3.9 ps for N_2 and 2.8 ps for O_2. Pulse self-shortening up to 8 fs temporal features within the filament is directly measured by impulsive vibrational excitation of H_2.

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