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**Upconversion-Detected Ultrafast Two-Dimensional Infrared Spectroscopy**[1] KEVIN KUBARYCH, University of Michigan, MANUEL JOFFRE, Laboratoire d'Optique et Biosciences Ecole Polytechnique — Two-dimensional infrared (2DIR) spectroscopy provides direct access to ultrafast molecular dynamics by measuring time- and structure-dependent couplings between vibrational transitions. Biological molecules, such as proteins, have rich vibrational spectra that relate to key structural elements including secondary structure ($\alpha$-helix, $\beta$-sheet), hydrogen bonding and protonation state. The ability to reliably measure 2DIR spectra in biological molecules represents a major step towards an atomic-level picture of biochemical dynamics. A key limitation of ultrafast IR spectroscopy has been the measurement of the spectrum in a grating-based spectrometer due to HgCdTe detectors limited to linear arrays of 128 or fewer pixels. We have circumvented this problem by converting the coherently generated four-wave mixing 2DIR signal into the visible spectrum, and recording it using a 1340x100 pixel silicon CCD camera. The IR signal is mixed in a MgO:$LiNbO_3$ crystal with a chirped, $<$10 $\mu$J, 0.5 ns, 800 nm pulse. Signal detection is sufficient to measure single-shot dispersed vibrational echo spectra, as well as heterodyne detected 2DIR in $Mn_2(CO)_{10}$. The IR emission temporally overlaps only a narrow frequency range of the chirped near-IR pulse resulting in negligible spectral broadening.

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