Infrared kinetic spectroscopy of molecular transients using a widely tunable difference-frequency source

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Recent advances in electro-optic material technology make it possible to generate coherent, widely tunable, continuous-wave (cw) infrared radiation by means of difference-frequency generation (DFG) based on materials such as $AgGaS_2$ and $AgGaSe_2$. Covering the spectral region from 3 to 18 μm, which contains a rich fraction of the primary-fingerprint infrared, this source represents a very valuable tool for high-resolution, high-sensitivity infrared absorption spectroscopy.

The generation of infrared light, continuously tunable from 3 to 9 μm, by mixing two cw single-frequency dye/Ti: sapphire lasers in a 45-mm-long $AgGaS_2$ crystal at room temperature with 90° type I noncritical phase matching has been demonstrated by our group.[1,2] By using optimum focusing conditions, cw infrared DFG powers at the $\frac{1}{4}$-mW level can now be provided reliably for spectroscopic applications. The infrared frequency can be tuned continuously over a width of 30 GHz. Absorption spectra of several stable molecular systems, e.g., H_2O, N_2O, NH_3, and CO, have been observed.

We report on the application of the difference-frequency source to infrared kinetic spectroscopy (IRKS) of transient molecules. These molecules can be generated conveniently by excimer-laser flash photolysis by using an appropriate precursor. For sensitivity enhancement an anastigmatic 1-m White cell with a variable effective optical path length of as much as 60 m has been developed. Special attention has been given to the optimization of the geometrical overlap of the UV excitation beam and the IR probe beam in the White cell. The molecular absorption signal is acquired by a computer-interfaced transient digitizer. First kinetic spectra of free radical species will be presented.

1. P. Canarelli, Z. Benko, R. F. Curl, and F. K. Tittel, J. Opt. Soc. Am. B **9**, 197 (1992).
2. A. H. Hielscher, C. E. Miller, D. C. Bayard, U. Simon, K. P. Smolka, R. F. Curl, and F. K. Tittel, J. Opt. Soc. Am. B **9** (1992).