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Detecting excited-state vibrational dynamics by broadband infrared or Raman probes; A unified picture based on loop diagrams KON-STANTIN E. DORFMAN, BENJAMIN FINGERHUT, SHAUL MUKAMEL, University of California, Irvine — Vibrational motions in electronically excited states can be probed either by time and frequency resolved infrared or by off resonant stimulated Raman techniques. Using loop diagrams, which represent forward and backward propagation of the wavefunction we derive similar multipoint correlation function expressions for both signals which are suitable for quantum microscopic simulations. The effective temporal (Δt) and spectral ($\Delta \omega$) resolution of the techniques is not solely controlled by experimental knobs since it also depends on the system dynamics being probed. The Fourier uncertainty $\Delta \omega \Delta t > 1$ is never violated.

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