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Simultaneously spectral phase and amplitude characterization of coherent molecular vibrations and its application in Coherent Anti-Stokes Raman Spectroscopy XIAOJI XU, STANISLAV KONOROV, JOHN HEPBURN, VALERY MILNER, University of British Columbia — The authors propose a new approach to vibration spectroscopy based on the coherent scattering of broadband ultrashort laser pulses. The proposed method reveals both the amplitude and the phase of molecular vibrations by utilizing cross-correlation Frequency Resolved Optical Gating technique (XFROG) in Coherent anti-Stokes Raman Scattering (CARS). The spectrum is measured as a function of the time delay between the laser-induced molecular vibrations and a well characterized broadband probe pulse. The iterative XFROG algorithm provides simultaneous complete characterization of molecular vibrations both in frequency and time domains with resolution better than pulse bandwidth or duration respectively. They experimentally demonstrate the feasibility of the proposed method and two of its potential applications: disentangling the time behavior of mixture of vibrationally excited molecules and CARS spectroscopy under high non-resonant background. Pulse shaping technique is used for further improvement of accuracy and stability against noise.

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