Advances in Low-Frequency 3-color Broadband Coherent Raman Spectroscopy of Condensed Phase Samples

LASZLO UJJ, University of West Florida — Low-frequency dispersive spontaneous Raman spectroscopy is a very useful method to measure phonon frequencies in crystals or characterize collective vibrational motions of macromolecules. The coherent version of the method has not been fully explored yet. It is shown here that the 3-color Broadband Coherent Raman scattering can be a very powerful extension to not only gas phase but condensed phase low frequency (5-500 cm$^{-1}$) vibrational measurements with large frequency separation between the narrowband and broadband radiation generating the signal. The spectral measurements presented here used volumetric Bragg filters for the first time to record coherent Raman spectra. Specific spectral analysis using model independent methods to derive the vibrational information is also presented. The technique can be extended to measure electronic resonance enhanced spectra by tuning only the frequency of the narrowband laser close to the electronic transition frequencies. This makes the method suitable for coherent Raman microscopy. The polarization properties of the signal is also explained and experimentally verified.

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