Title:	A Revisit on Impulsive Stimulated Raman Spectroscopy: Importance of Spectral Dispersion of Chirped Broadband Probe
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Abstract:	The usefulness of a chirped broadband probe and spectral dispersion to obtain Raman spectra under nonresonant/resonant impulsive excitation is revisited. A general methodology is presented that inherently takes care of phasing the time-domain low-frequency oscillations without probe pulse compression and retrieves the absolute phase of the oscillations. As test beds, neat solvents (CCl4, CHCl3, and CH2Cl2) are used. Observation of periodic intensity modulation along detection wavelengths for particular modes is explained using a simple electric field interaction picture. This method is extended to diatomic molecule (iodine) and polyatomic molecules (Nile blue and methylene blue) to assign vibrational frequencies in ground/excited electronic state that are supported by density functional theory calculations. A comparison between frequency-domain and time-domain counterparts, i.e., stimulated Raman scattering and impulsive stimulated Raman scattering using degenerate pump–probe pairs is presented, and most importantly, it is shown how impulsive stimulated Raman scattering using chirped broadband probe retains unique advantages offered by both.
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