SPECTROSCOPIC LINE SHAPES OF BROAD BAND SUM FREQUENCY GENERATION

HIMALI D JAYATHILAKE , IGOR V STIOPKIN, CHAMPIKA WEERAMAN, ACHANI K YATAWARA and ALEXANDER V BENDERSKII , *Department of Chemistry, Wayne State University, Detroit, MI 48202*.

Using a combination of theoretical modeling and experimental measurements we show how the visible pulse shape and delay can be used to simultaneously maximize the spectral resolution and signal intensity, avoiding the trade-off between the two inherent in the broad band SFG schemes. It is found that at negative time delays (visible pulse come before IR pulse), the instantaneous non resonant background is suppressed by revealing the slow-decaying resonant contribution. Symmetric CC (triple bond) vibrational mode of propiolic acid was used as a test system. Spectra with narrow, symmetric line shapes close to the true transition line width were obtained at negative time delays without sacrificing signal intensity and resolution. Model calculations were performed in order to fit data with different time delays and extract the true line width of the particular vibrational mode. Etalon and stretcher based visible pulses were used to compare the VSFG spectral characteristics. Stretcher based visible pulses were found to produce VSFG spectra with high signal to noise ratio possibly due to the smoothness of the temporal profile of the pulse where as etalon based visible pulse has a singularity at time zero.