Calculation of Raman and infrared spectra by coherent-phonon stimulation

JONATHAN YATES, IVO SOUZA, University of California, Berkeley and Lawrence Berkeley National Laboratory — We propose a novel method for the efficient first principles prediction of Infra-Red and non-resonant Raman spectra. The method is inspired by the experimental technique of impulsive- stimulated Raman scattering. We apply initial impulsive forces to the ions in the system. For IR spectroscopy these forces correspond to the first order forces induced by a static electric field; for Raman spectroscopy they are the second order forces. We show how the corresponding vibrational spectrum can be obtained from the ensuing short-time dynamics of the system. The method has better scaling with system size than existing techniques. We present applications of the method to various clusters and molecules.