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Semiclassical Treatment for Small-Molecule Dynamics in Low-Temperature Crystals Using a Fixed Vibrational Basis and Gaussian Bath Wave Packets XIAOLU CHENG, CRAIG CHAPMAN, JEFFREY CINA — The dynamics of small molecules in low-temperature crystals has been studied by several methods of time-resolved coherent nonlinear optical spectroscopy. Successful interpretation of these ultrafast signals will help illuminate basic aspects of quantum molecular dynamics in condensed media. We outline a mixed quantum/semiclassical theory that enables the systematic simulation of experimental signals from samples featuring a high-frequency vibrational mode immersed in a lower-frequency environment. We treat as the "system" a small number of high- frequency intramolecular degrees of freedom, those driven to large-amplitude motion by direct laser excitation. The low frequency bath of crystal phonons experiences indirectly induced, smaller-amplitude motion. We expand the overall wave function using the eigenstates of the system Hamiltonian as a vibrational basis, and adopt a Gaussian ansatz for the multi- dimensional bath wave packet. Computational details will be described along with representative numerical simulations illustrating the strengths and limitations of the theory.

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