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Beyond vibrational dissipation and decoherence. A mixed quantum/semiclassical theory for small-molecule and host-lattice dynamics in low-temperature media. CRAIG CHAPMAN, XIAOLU CHENG, JEFFREY CINA, University of Oregon, Department of Chemistry, Oregon Center for Optics — Bath-mediated dynamics play an important role in condensed phase processes. Coupling of a small-molecule chromophore vibration to a cryogenic host can lead to coherent motion of the lattice atoms as in addition to giving rise to vibrational decoherence and vibrational energy relaxation, which can in turn affect ultrafast spectroscopic signals. We present a mixed quantum/semiclassical theory to simulate the dynamics of these systems that makes use of several key features of cryogenic chromophore-host complexes. In particular we exploit their well defined structure, and weak coupling aspects by framing a theory that treats the large-amplitude vibrational motion and the small-amplitude bath dynamics with differing degrees of rigor. The system is treated fully quantum mechanically while the bath is treated as a collection of multidimensional Gaussian wave packets. Calculated linear absorption spectra and linear wave packet interferometry signals for a simple test case are presented. A nonlinear version of wave packet interferometry has been shown to be an effective method of reconstructing molecular wave packets using a variety of pulse durations and interpulse delays, and may have applications in low-temperature environments as well.

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