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Non-Markovian harmonic bath model for molecular systems: Influence of the bath spectral density JAN RODEN, Department of Chemistry, University of California, Berkeley, California 94720, USA, ALEXANDER EISFELD, Department of Chemistry and Chemical Biology, Harvard University, 12 Oxford Street, Cambridge, MA 02138, K. BIRGITTA WHALEY, Department of Chemistry, University of California, Berkeley, California 94720, USA — In quantum mechanical simulations of the electronic excitation dynamics in molecular complexes, like natural or artificial light-harvesting complexes, often open quantum system descriptions are applied, to treat a large number of degrees of freedom involved. A popular approach is then, to include only the electronic degrees of freedom into the system part and to couple them to a non-Markovian bath of harmonic vibrational modes. The coupling to the bath, representing intra-molecular as well as external vibrations, is usually described via the bath spectral density, which therefore is an important ingredient in this approach. Here, we discuss different aspects of the influence of the bath spectral density on dynamics and optical spectra. In particular, we consider structured spectral densities, consisting of multiple broadened peaks. It is often assumed that the strong coupling to an intra-molecular vibrational mode, which is damped by coupling to other vibrational modes, is described reasonably by such a broadened peak in the spectral density. Here we demonstrate that this interpretation should be used with caution, because the damping of the mode differs from the model for an intra-molecular mode that one would usually apply when including the mode directly in the system part.

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