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Exciton Scattering approach for conjugated macromolecules: from electronic spectra to electron-phonon coupling

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The exciton scattering (ES) technique is a multiscale approach developed for efficient calculations of excited-state electronic structure and optical spectra in low-dimensional conjugated macromolecules. Within the ES method, the electronic excitations in the molecular structure are attributed to standing waves representing quantum quasi-particles (excitons), which reside on the graph. The exciton propagation on the linear segments is characterized by the exciton dispersion, whereas the exciton scattering on the branching centers is determined by the energy-dependent scattering matrices. Using these ES energetic parameters, the excitation energies are then found by solving a set of generalized "particle in a box" problems on the graph that represents the molecule. All parameters can be extracted from quantum-chemical computations of small molecular fragments and tabulated in the ES library for further applications. Subsequently, spectroscopic modeling for any macrostructure within considered molecular family could be performed with negligible numerical effort. The exciton scattering properties of molecular vertices can be further described by tight-binding or equivalently lattice models. The on-site energies and hopping constants are obtained from the exciton dispersion and scattering matrices. Such tight-binding model approach is particularly useful to describe the exciton-phonon coupling, energetic disorder and incoherent energy transfer in large branched conjugated molecules. Overall the ES applications accurately reproduce the optical spectra compared to the reference quantum chemistry results, and make possible to predict spectra of complex macromolecules, where conventional electronic structure calculations are unfeasible.