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Watching excitons move: the time-dependent transition density matrix[1]

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Time-dependent density-functional theory allows one to calculate excitation energies and the associated transition densities in principle exactly. The transition density matrix (TDM) provides additional information on electron-hole localization and coherence of specific excitations of the many-body system. We have extended the TDM concept into the real-time domain in order to visualize the excited-state dynamics in conjugated molecules. The time-dependent TDM is defined as an implicit density functional, and can be approximately obtained from the time-dependent Kohn-Sham orbitals. The quality of this approximation is assessed in simple model systems. A computational scheme for real molecular systems is presented: the time-dependent Kohn-Sham equations are solved with the OCTOPUS code and the time-dependent Kohn-Sham TDM is calculated using a spatial partitioning scheme. The method is applied to show in real time how locally created electron-hole pairs spread out over neighboring conjugated molecular chains. The coupling mechanism, electron-hole coherence, and the possibility of charge separation are discussed.

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