Reduced-Density-Operator Description for Single-Photon and Multi-Photon Processes in Many-Electron Atomic and Molecular Systems

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Abstract. A reduced-density-operator formulation is developed for single-photon and multi-photon processes in quantized many-electron systems, taking into account environmental collisional and radiative interactions. A fundamental microscopic quantum-mechanical description for the spectral line shapes is provided, which is based on the derivation of precise expressions for the tetradic matrix elements of the frequency-domain Liouville-space self-energy operator occurring in the general expression for the Liouville-space transition operator. Using a perturbation expansion of the frequency-domain Liouville-space self-energy operator, the spectral-line widths and shifts are systematically evaluated in the isolated-line, short-memory-time (Markov), and lowest-order (Born) approximations. The general reduced-density-operator formulation can also be employed in the more difficult spectral description that is applicable to overlapping lines. Applications of interest include spectral simulations for atomic and molecular systems. In addition to the standard data sets consisting of collisional cross sections and radiative transitions rates, the corresponding transitions matrix elements may also be required.

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