# Time-Dependent Two-Particle Reduced Density Matrix Theory: Application to High-Harmonic Generation

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**Synopsis** We follow a new approach for calculating high-harmonic spectra for multi-electron atoms and molecules by propagating the two-particle reduced density matrix. Calculated results are in very good agreement with state-of-the-art many-body wavefunction-based benchmark calculations.

High-harmonic generation (HHG) is one of the fundamental processes in strong field physics whose applications range from attosecond metrology [1], tunable table-top XUV/Soft X-ray sources [2] to high precision spectroscopy [3] and orbital imaging [4]. On the atomic level the theoretical description of HHG is challaging because of the multi-electron nature of the underlying process. While simple models such as the single-active-electron approximation (SAE) or time-dependent Hartree-Fock (TDHF) are well suited to describe qualitative features of HHG, advanced theories capable of correctly treating electron correlation are needed for a quantitative description [5]. However, with increasing system size conventional wavefunction-based methods such as the multi-configurational time-dependent Hartree-Fock (MCTDHF) method soon become unfeasible due to their exponential scaling with particle number. One way to overcome this exponential barrier is to abandon a wavefunction-based description and propagate time-dependent reduced densities instead. Considerable success along this line has been achieved by time-dependent density functional theory (TDDFT) [6]. However, accurate calculations in TDDFT face the difficulty of unknown exchange-correlation functionals that are difficult to improve systematically. We propose to go beyond the limitations of TDDFT by propagating the time-dependent two-particle reduced density matrix (TD-2RDM). As a hybrid between the electron density and the many-body wavefunction the 2RDM fully includes two-particle correlations. We have implemented the TD-2RDM method to describe high-harmonic generation of fully three-dimensional multi-electron atoms [7]. To obtain accurate results for the electronic response we use an advanced closure scheme [8] that is suited to conserve all constants of motion associated with symmetries of the Hamiltonian. We benchmark the performance of the TD-2RDM method by comparing it to a state of the art MCTDHF calculation [9] as well as to TDDFT calculations (see Fig.1). We find very good agreement between the TD-2RDM and the MCTDHF method while TDDFT within the local density approximation shows clear deviations indicating that the correct treatment of two-particle correlations is essential to obtain accurate HHG spectra.

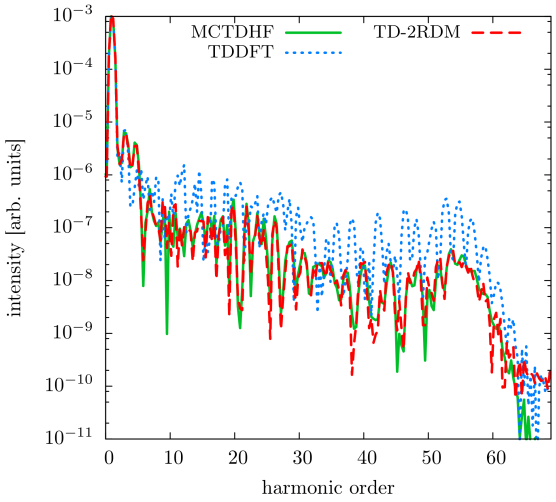


**Figure 1**. HHG spectrum of beryllium subject to a 3-cycle laser pulse with intensity $I = 4.0 \times 10^{14}$W/cm$^2$. MCTDHF(—), TD-2RDM(- -), TDDFT(···).

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