# The role of orbital angular momentum constraints in the variational optimization of the two-electron reduced-density matrix

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In order to use ab initio methods to correctly predict the properties of chemical systems containing heavy elements, one must account for both correlation and relativistic effects, including spin-orbit coupling. The complete active space self-consistent-field method (CASSCF) provides a reliable description of nondynamical correlation effects, but the steep scaling of configuration interaction (CI)-based CASSCF precludes its application to large systems. The variational two-electron reduced-density-matrix (v2RDM) driven methods provide a computationally efficient alternative to CI-based approaches. Both scalar relativistic and spin-orbit coupling effects can be captured within an exact two component (X2C) extension of v2RDM-driven CASSCF. We have developed a complex generalized implementation of the complete active space v2RDM approach (v2RDM-CASCI) that is suitable for X2C-v2RDM-CASCI computations. Our preliminary investigation considered the effect of the orbital and total angular momentum constraints on the reduced-density matrices (RDMs) of atomic systems. Multiple angular momentum states can be modeled by placing constraints on the expectation values of both the square of the orbital angular momentum operator and the z-projection of the orbital angular momentum operator. We have found that these constraints not only allow us to describe electronically excited states that are otherwise inaccessible, but they can improve the description of previously accessible ones.