General implementation of the relativistic coupled-cluster method

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We report the development of a general order relativistic coupled-cluster (CC) code. Our implementation is based on Kramers-paired molecular spinors, utilizes double group symmetry, and is applicable with the full Dirac-Coulomb and several approximate relativistic Hamiltonians. The available methods include iterative and perturbative single-reference CC approaches with arbitrary excitations as well as a state-selective multi-reference CC ansatz. To illustrate the performance of the new code benchmark calculations have been performed for the total energies, bond lengths, and vibrational frequencies of the monoxides of Group IVa elements. The trends due to the simultaneous inclusion of relativity as well as higher-order electron correlation effects are analyzed. The newly developed code significantly widens the scope of the *ab initio* relativistic calculations, for both molecules and atoms alike, surpassing the accuracy and reliability of the currently available implementations in the literature.