Harmonium atoms, i.e. assemblies of electrons trapped in a harmonic potential, are encountered in diverse branches of chemistry and physics. In physics, they emerge in description of quantum dots, particles in dusty plasmas, and ions in cold traps, whereas in quantum chemistry they are often employed in calibration of density and density matrix functionals. Despite the importance of harmonium atoms, their properties have been systematically studied only in the two-electron case.

When used in conjunction with appropriate extrapolation schemes, full configuration interaction (FCI) calculations employing systematic sequences of spherical Gaussian primitives with even-tempered exponents shared by functions of different angular momenta are capable of affording the ground-state energies of the two-electron harmonium atoms with a few-micro-Hartree accuracy that is sufficient for calibration and benchmarking of approximate electron correlation theories of quantum chemistry. The present approach, which is subsequently employed in computations of electronic properties of harmonium atoms with three electrons, calls for a series of between 15 and 17 FCI runs involving basis sets with between four and eight Gaussian primitives of the sp, spd, spdf, and spdfg type.

Details of these calculations, which are limited by linear dependencies among basis functions that become significant for small values of the force constant, are presented.