

broad rubric of time-dependent (TD) DFT. Although considerable theoretical progress has been made for real-space dynamical studies of atoms/molecules under strong fields within TDDFT (see, for example, [39], for a review), several nontrivial problems are encountered for arbitrary atomic/molecular system. Extension in these cases is not straightforward, for they pose considerable challenge. On the other hand, extension of these above-mentioned LCAO-MO-based DFT approaches within ACG is quite difficult in the TD domain. In order to proceed further in that direction, here, for the first time, we report full all-electron calculation of several atoms using the basis set method, completely in CCG. This constitutes an essential first step (the “structure” part) much needed for real-time TDDFT studies. To this end, total energies, component energies, orbital energies as well as HOMO energies from these full calculations are compared systematically with reference literature values. Section II gives a brief overview of the methodology used; discussion on our results are given in Section III, while we end with a few concluding remarks in Section IV.

## II. METHODOLOGY

The method employed in this work has been presented before [24–26] in some detail. Hence will not be repeated here; only essential details are given. Unless otherwise mentioned, atomic units implied henceforth.

In KS DFT, the problem of calculating total ground-state electronic energy of a many-electron system is transformed into solving the following single-particle KS equation,

$$\left[ -\frac{1}{2}\nabla^2 + v_{ext}(\mathbf{r}) + v_h(\mathbf{r}) + v_{xc}(\mathbf{r}) \right] \psi_i(\mathbf{r}) = \epsilon_i \psi_i(\mathbf{r}) \quad (4)$$

Here  $v_{ext}$  signifies the external potential in which electrons move, containing an electrostatic potential due to the presence of nuclei, but may also include other terms (in present occasion, none);  $v_h$  denotes classical Coulomb potential arising because of the electron distribution; and finally  $v_{xc}$  corresponds to a multiplicative XC potential that depends on electron density, but not on wave functions explicitly.

As already mentioned, KS MOs are built from localized Gaussian type basis functions as in Eq. (3). The LCAO-MO approach is quite efficient; for it can give very accurate results and also it provides basis for creation of new methods such as order-N, Green’s function approaches, etc. Note, full self-consistent DFT procedure requires specification of