with respect to α, a value within the range of 6–8 seemed satisfactory and we employed 7 for all the results reported in this work. This has produced highly satisfying results for a modest number of atomic (3) and molecular (12) systems in our previous work [23]. The short-range Fourier integral is calculated analytically, whereas the long-range part is obtained from FFT of the real-space values.

As already mentioned, a major thrust of the current communication is to demonstrate the feasibility and viability of our current scheme in the context of so-called "non-local" (gradient and Laplacian-dependent) XC functionals which would be necessary for future chemical applications. For this, we first test this with two of the widely used functionals, namely the Becke exchange [24] and LYP correlation [25] (for convenience, an alternative equivalent form [33] containing only the first derivative has been mostly used in practice). The homogeneous electron-gas correlation of Vosko-Wilk-Nusair (VWN) [34] is used in all the LDA calculations. Following [35], the gradient-dependent functionals can be treated without evaluating the density Hessians by using a finite-orbital basis expansion. To this end, XC contributions of the KS matrix is written in the following working form,

$$F_{\mu\nu}^{XC\alpha} = \int \left[\frac{\partial f}{\partial \rho_\alpha} \chi_\mu \chi_\nu + \left(2 \frac{\partial f}{\partial \gamma_{\alpha\alpha}} \nabla \rho_\alpha + \frac{\partial f}{\partial \gamma_{\alpha\beta}} \nabla \rho_\beta \right) \cdot \nabla(\chi_\mu \chi_\nu) \right] d\mathbf{r} \qquad (7)$$

where $\gamma_{\alpha\alpha} = |\nabla \rho_\alpha|^2$, $\gamma_{\alpha\beta} = \nabla \rho_\alpha \cdot \nabla \rho_\beta$, $\gamma_{\beta\beta} = |\nabla \rho_\beta|^2$ and f is a function only of the local quantities ρ_α, ρ_β and their gradients. The BLYP functionals are implemented using the Density Functional Repository program [36]. The two-electron matrix elements cannot be evaluated analytically; here we use direct numerical integrations on the CCG grid. Note that some of the existing DFT codes [37] use an alternative route of fitting these by an auxiliary basis set, the so-called discrete variational method [38–40]. The generalized matrix-eigenvalue problem is solved using the standard LAPACK routine [41] accurately efficiently. Self-consistent set of MOs, density and energy are obtained in the usual manner subject to the convergence of (a) potential (b) total energy and (c) eigenvalues. Tolerance of 10^{-6} a.u. was used for (b) and (c), while a value of 10^{-5} for (a). Atomic units employed, unless otherwise mentioned.