The density functional theory (DFT) has become the workhorse in electronic structure calculations. Its success derives from the ability to produce accurate results with reasonable computational effort. The drawback of the DFT is that in principle it is bound to the ground-state properties. However, the time-dependent DFT (TDDFT) extends the applicability of the DFT to excited states. In recent years, TDDFT has been applied successfully to several problems, such as the calculation of optical absorption spectra of broad variety of systems, nonlinear optical response, and coherent control [1].

We present an all-electron implementation of the DFT and linear response TDDFT which employs hierarchial high-order finite-element basis. Our mesh generation scheme in which structured atomic meshes are merged to an unstructured molecular mesh allows highly nonuniform discretization of the space. Thus it is possible to represent the core and valence states using the same discretization scheme, i.e., no pseudopotentials or similar treatments are required. The nonuniform discretization also allows the use of large simulation cells, and therefore avoids any boundary effects.

Figure 1: Cut plane of molecular finite element mesh of $C_6H_6$: a) Complete mesh (diameter of 55 Å), b) atomic mesh near a carbon nucleus, and c) close-up of the molecular region.

Figure 2: Optical absorption spectrum of $C_6H_6$ calculated using finite element based linear-response TDDFT.