Real-space All-electron Band Structure Calculations

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Abstract

An all-electron numerical framework for band structure calculations is presented. A 3D finite element mesh is used to discretized the DFT/Kohn-Sham problem in real space using full ionic local potential. The mathematical model allows flexibility for addressing various combinations of Bloch and Dirichlet boundary conditions for low-dimensional nanostructures, and the resulting large-scale eigenvalue problems are solved using the FEAST solver [1]. The complexity of numerical framework scales linearly with the number of atoms, and it is flexible to address arbitrary impurities, defects and roughness.

Model

Band structure calculations are performed on a finite element mesh with DFT utilizing the full ionic all-electron potential and Bloch periodic boundary conditions.

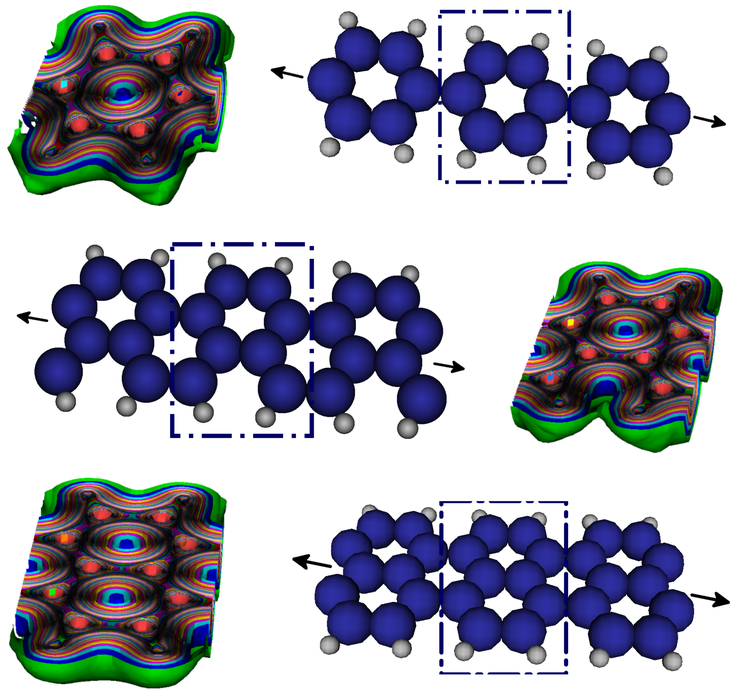
$$(-\frac{\hbar^2}{2m}\triangle^2 + V_{eff}[\rho(r)])\psi_{i,k}(r) = E_i\psi_{i,k}, \quad \rho(r) = \sum_i \int |\psi_{i,k}|^2 dk$$

$$V_{ion}(r) = \frac{1}{4\pi\epsilon}\sum_i \frac{-qZ_i}{r}, \quad -\triangle V_H(r) = \frac{\rho(r)}{\epsilon}, \quad V_H(\Gamma) = \frac{q}{4\pi\epsilon}\int \frac{\rho(\Gamma)}{|\Gamma - r'|}d\Gamma$$

The Hartree potential is calculated from the Poisson equation with both periodic and Dirichlet boundary conditions and is solved using the Conjugate Gradient method with an incomplete Cholesky preconditioner. The local density approximations with Perdew-Zunger correction is used for exchange and correlation terms. The resulting eigenvalue problem is solved using FEAST[1]. Self consistency is achieved using Pulay mixing.

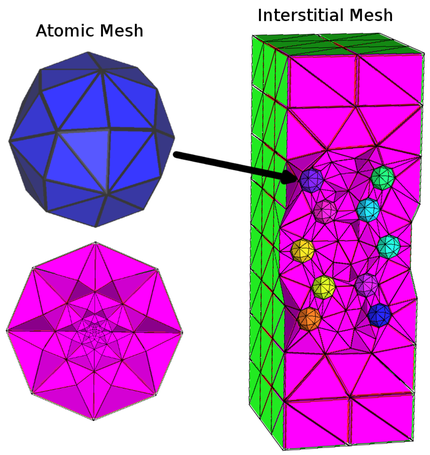
FEAST

The FEAST eigenvalue algorithm [2] offers many important and unique capabilities for achieving accuracy, robustness, high-performance and scalability on parallel computing architectures. At first, the algorithm can operate in parallel to obtain core and valence electrons independently spanning different energy ranges. Secondly, solving the original eigenvalue problem within a given energy range (i.e. search interval) is mainly reformulated into solving a set of well-defined independent linear systems along a complex energy contour. A high performance implementation of the algorithm is freely available available online [1].

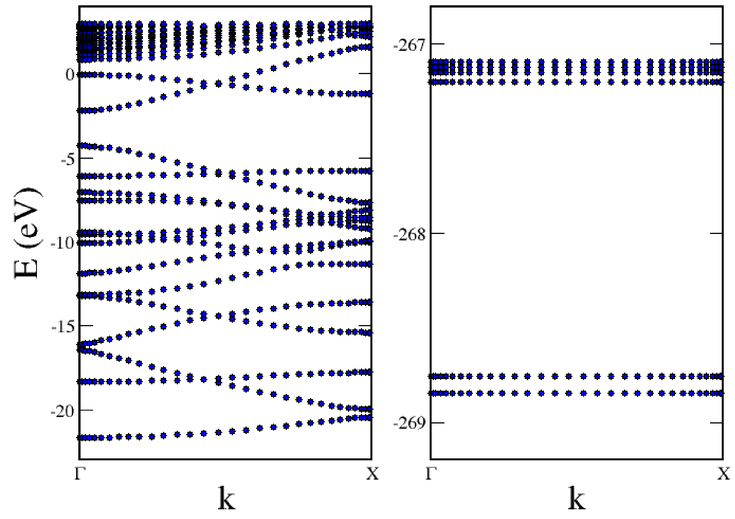


All-Electron Density for 3-AGNR (PPP), 4-AGNR and 5-AGNR

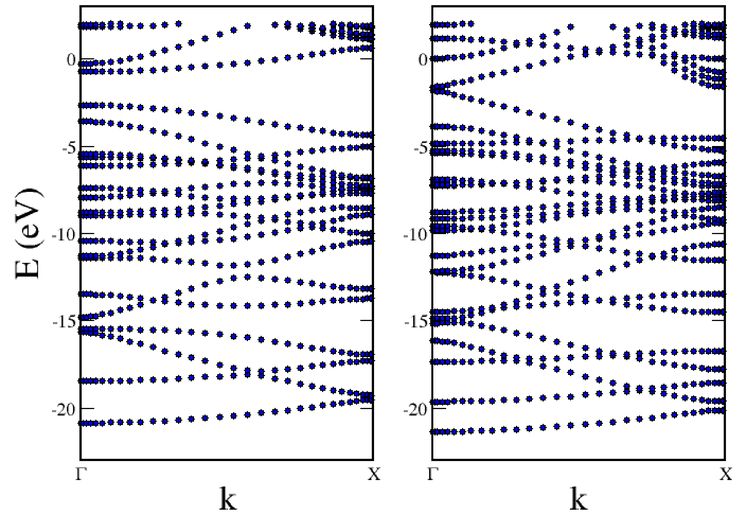
Discretization



A cubic FEM Muffin-tin mesh is constructed from Interstitial and Atomic local meshes (show above for a PPP unit cell). The non-linear eigenvalue problem arising from the muffin-tin decomposition of the real-space domain is reformulated[4] and solved exactly within the FEAST framework[2]. Additionally, this grants a third level of parallelism for the FEAST eigenvalue solver [1].



Band structure of Valence (left) and core (right) states of PPP molecule. Results are in strong agreement with [5].



The band structure of the semiconducting 4-AGNR (left) and metallic 5-AGNR (right).

References

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