



A fast, precise, and flexible DFT code for ab-initio atomistic simulation

Case study

OLEDs

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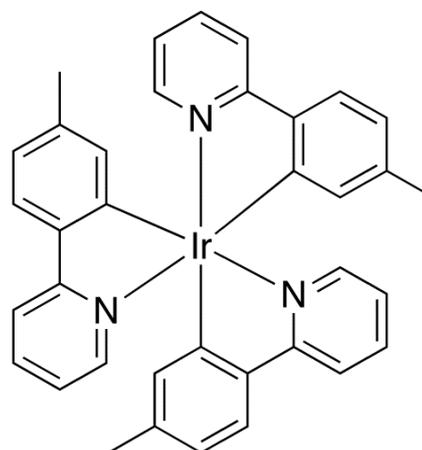
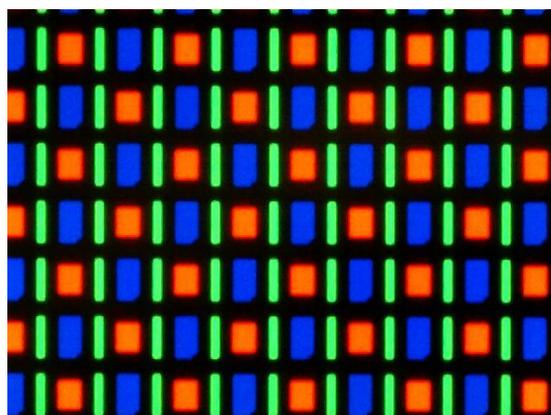
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The Challenge

Organic electronics involve the study of organic molecules offering technologically relevant properties. They present advantages with respect to inorganic electronics, such as light weight, low cost, and flexibility, with applications such as organic **light-emitting diodes (OLEDs)**, **photovoltaic cells** and **transistors**.

A typical material for OLEDs consists of a matrix of host molecules doped with a small proportion of optically active guest molecules (~8%) where holes and electrons recombine and emit radiation. **Iridium (Ir)** and **Platinum (Pt)**-based complexes recombine electron-hole pairs efficiently, and are thus the base for the majority of guest molecules. However, Ir and Pt are expensive and their extended use turns out to be unsustainable. **Computational methods** can help to **explore** the vast chemical search space of guest candidates to find **efficient and purely organic alternatives**.

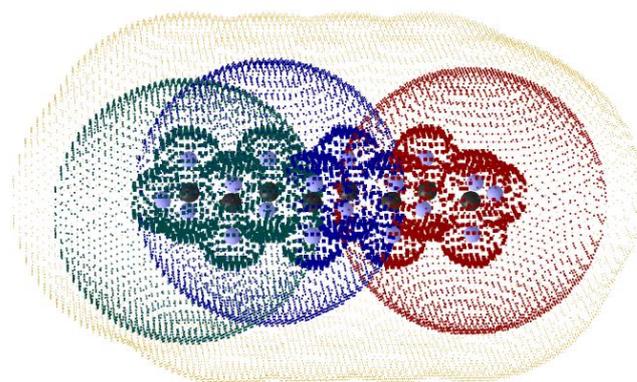
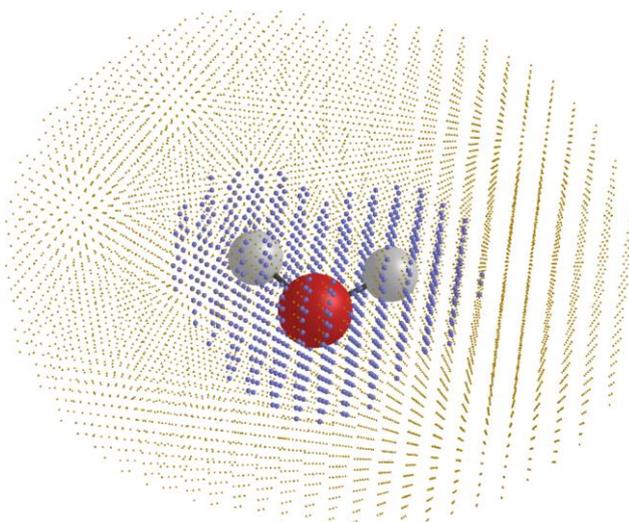


The Approach

The typical approach to study charge transport properties in such systems consists of several steps. First, the structure is created using classical molecular dynamics. Then, transport properties of the structure are calculated with semiempirical or ab initio methods. Finally, the calculated parameters are used to describe macroscopic properties. The bottleneck of this approach is the calculation of **transport properties from first principles**. Therefore, it is crucial to use an ab initio code that is at the same time **fast and accurate**.

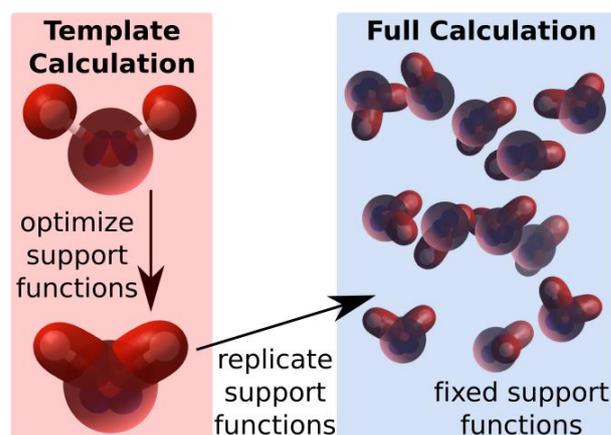
Unlike other ab initio codes, BigDFT is particularly well suited to tackle large calculations of such inhomogeneous systems. This is due to the use of **Daubechies wavelets** as a basis set, which are a set of systematic but still localized functions that create two levels of resolution in an **adaptive way**, namely a finer level close to the atom cores and a coarser one farther away. Thanks to this **multiresolution** and the **efficient parallelization** of the code, large systems of the order of 1,000 atoms can be studied.

Furthermore, BigDFT also exploits the **“nearsightedness” of large quantum systems** to express the solutions in terms of a small set of localized **support functions** centered on the atoms. They are **optimized in situ** (i.e. they are susceptible to the chemical environment of each atom) and therefore yield an **outstanding precision**. The use of the optimized support functions leads to **O(N) ab initio calculations**, i.e. the computational time increases only linearly as a function of the number of atoms. In this way systems containing **up to 10,000+ atoms can be studied**.

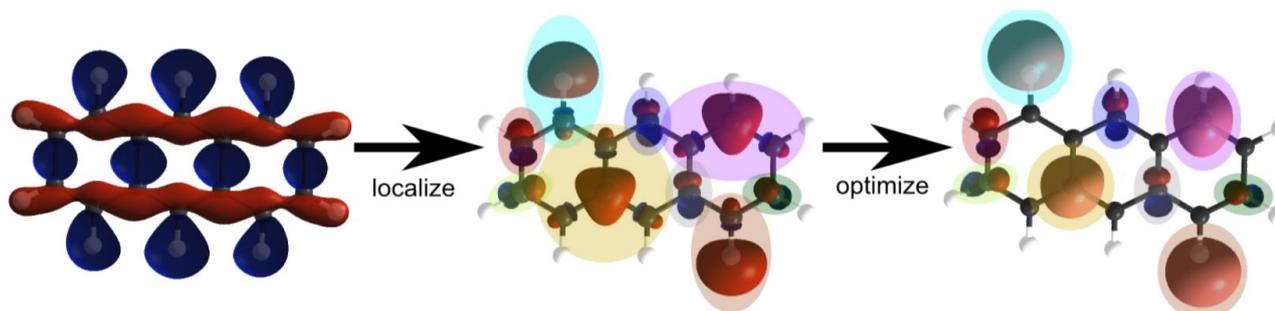
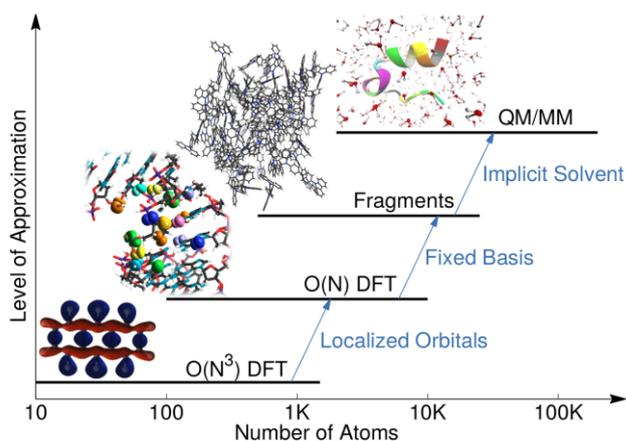


The Approach

In addition to the linear scaling approach, BigDFT also implements an algorithm exploiting the **similarity between parts of the system** (e.g. same molecules in similar chemical environments). These sub-parts of the system are denoted as **fragments**. In this setup, the support functions are optimized for one “template” fragment and then rototranslated to be used as support functions of other equivalent fragments of the system at different positions and orientations. This approach **improves the efficiency** of the calculation even further.

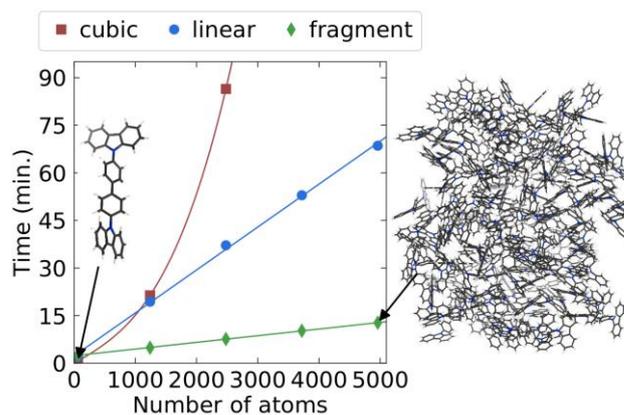


This multiscale approach is showcased in the present work. The involved steps are: (1) the **host-guest structure** is found using the Metropolis Monte Carlo algorithm; (2) the nearest neighbours for each molecule are determined and included into the calculations to create a more **realistic system setup**; (3) the **support functions are optimized** for each “template” fragment, taking into account its chemical environment; (4) the various **template support functions** are rototranslated to be used for all other identical fragments.; (5) the **transport properties** are calculated.



The Results

This approach to calculating key **transport properties in supramolecular architectures** has been implemented and successfully applied to a system based on two organic host and guest molecules. The values of the calculated properties are different from the results obtained using two isolated molecules, showing the **importance of the chemical environment** and demonstrating the usefulness of this **accurate and efficient method to treat larger systems with BigDFT**. It can be used to explore other guest candidates and systems or even to calculate the evolution of these transport properties in time, as different structures are generated using Molecular Dynamics. By using the fragments scheme, which enables the reuse of support functions for other parts of the system, the **computational cost is reduced by an order of magnitude** with respect to the native linear scaling method. This case study demonstrates that BigDFT can be a useful **tool for the design of novel organic electronic devices**.



For more information:

Publication: ***Toward Fast and Accurate Evaluation of Charge On-Site Energies and Transfer Integrals in Supramolecular Architectures Using Linear Constrained Density Functional Theory (CDFT)-Based Methods***

Laura E. Ratcliff, Luca Grisanti, Luigi Genovese, Thierry Deutsch, Tobias Neumann, Denis Danilov, Wolfgang Wenzel, David Beljonne, and Jérôme Cornil

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Video: [Simulating OLED with BigDFT](#)