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EVO—Evolutionary algorithm for crystal structure prediction☆

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ABSTRACT

We present EVO—an evolution strategy designed for crystal structure search and prediction. The concept and main features of biological evolution such as creation of diversity and survival of the fittest have been transferred to crystal structure prediction.

EVO successfully demonstrates its applicability to find crystal structures of the elements of the 3rd main group with their different spacegroups. For this we used the number of atoms in the conventional cell and multiples of it. Running EVO with different numbers of carbon atoms per unit cell yields graphite as the lowest energy structure as well as a diamond-like structure, both in one run. Our implementation also supports the search for 2D structures and was able to find a boron sheet with structural features so far not considered in literature.

Program summary

Program title: EVO

Catalogue identifier: AEOZ_v1_0

Program summary URL: http://cpc.cs.qub.ac.uk/summaries/AEOZ_v1_0.html

Program obtainable from: CPC Program Library, Queen's University, Belfast, N. Ireland

Licensing provisions: GNU General Public License version 3

No. of lines in distributed program, including test data, etc.: 23488

No. of bytes in distributed program, including test data, etc.: 1830122

Distribution format: tar.gz

Programming language: Python.

Computer: No limitations known.

Operating system: Linux.

RAM: Negligible compared to the requirements of the electronic structure programs used

Classification: 7.8.

External routines: Quantum ESPRESSO (http://www.quantum-espresso.org/), GULP (https://projects.ivec.org/gulp/)

Nature of problem:
Crystal structure search is a global optimisation problem in $3N + 3$ dimensions where N is the number of atoms in the unit cell. The high dimensional search space is accompanied by an unknown energy landscape.

Solution method:
Evolutionary algorithms transfer the main features of biological evolution to use them in global searches. The combination of the "survival of the fittest" (deterministic) and the randomised choice of the parents and normally distributed mutation steps (non-deterministic) provides a thorough search.

☆ This paper and its associated computer program are available via the Computer Physics Communication homepage on ScienceDirect (http://www.sciencedirect.com/science/journal/00104655).

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Restrictions:
The algorithm is in principle only restricted by a huge search space and simultaneously increasing calculation time (memory, etc.), which is not a problem for our piece of code but for the used electronic structure programs.

Running time:
The simplest provided case runs serially and takes 30 minutes to one hour. All other calculations run for significantly longer time depending on the parameters like the number and sort of atoms and the electronic structure program in use as well as the level of parallelism included.

1. Introduction

The crystal structure contains the main information of a material and can be used to calculate many physical properties. Therefore, it is one aim in materials design to find so far unknown crystal structures and ideally predict new compounds that have desirable properties. Discovering stable structures is an optimisation problem where one has to find the structure with the minimal free energy for a given atomic composition. Quantum mechanical methods can provide free energies for most of the structures but the remaining problem is locating the global minimum in a high-dimensional energy landscape.

Most optimisation methods suffer from the danger to get stuck in a local minimum but there exist several methods that have the ability to leave local minima. This is essential in crystal structure prediction where one has to deal with a high dimensional search space ($3N + 3$ dimensions, where N is the number of atoms) and the assumption of many local minima.

There are several methods that are used in crystal structure prediction such as evolutionary algorithms [1–6], simulated annealing [7], minima/basin hopping [8,9], random structure search [10,11] and particle swarm optimisation [12]. An overview of these and other approaches for crystal structure prediction can be found in [13] and references therein.

We apply an evolutionary algorithm to crystal structure prediction, which uses the most significant features of natural evolution for global optimisation—recombination, mutation, selection, and survival of the fittest. In the case of the use of a real space representation of the individuals, e.g. crystal structures, one refers to the algorithm as an evolution strategy. Historically, the term genetic algorithm differs from evolution strategies mainly in the binary encoding of the individuals. For further distinction see [14].

In this paper, we present our newly developed evolution strategy for the purpose of crystal structure prediction. A special feature of our implementation is the possibility to search also for 2-dimensional structures, which is applied to boron sheets as a test case. We also introduced the concept of ageing of the individuals and implemented several termination criteria to discharge the user from ending EVO. Our evolution strategy is implemented in python and currently using the external programs Quantum ESPRESSO [15] and GULP [16] but can easily be extended to use other programs for total energy evaluation of the structures.

The paper is organised as follows. After describing the method and its characteristics in detail we illustrate the modification of the code to deal with sheet structures before we present some examples to demonstrate the general applicability of our algorithm. We also show a boron sheet structure so far not reported in the literature. The last section is a more detailed description of the implementation including input and output files, and requirements.

2. Method

Evolution strategies as a branch of evolutionary algorithms [14] use a real space representation of the parameters of the individual. The operators recombination and mutation that are known from natural evolution are applied to individuals to form new ones. These offspring are possibly superior (fitter) according to the target function.

Our algorithm for crystal structure prediction (EVO) uses crystal structures as individuals and the Gibbs free energy as the fitness function that has to be minimised. At 0 K the free energy reduces to the enthalpy $H = E + PV$ which we use as the fitness value (which accounts for different unit cell volumes).

Basic terms in evolutionary algorithms are *individual* and *population*. An individual I containing the crystal structure information in our evolution strategy is characterised by

$$I = (cell, atoms, age, fitness)$$

where *cell* denotes the form of the unit cell. *Atoms* stands for the atomic positions linked to the respective atom species and *fitness* is the calculated free energy of the individual. Cell parameters and atom positions of the structures are represented in the usual way. Since we do not assume any symmetry, the cell is defined by the three cell axes (in Å) and the angles between them. The atom positions are stored in fractional coordinates (between 0 and 1) with respect to the cell vectors.

The *age* parameter (maximal life span) allows a certain individual to reproduce only over a finite number of generations [17], which encorporates the two traditional selection strategies (see [14, p. 78]) as special cases.

There is also an option to rotate the unit cell so that the axes are ordered according to their length which enhances comparability and recombination behaviour of the evolutionary algorithm. The comparison of two individuals is done as described in Section 2.4.

A population P in our scheme is a number μ of individuals I that is allowed to mate and to create new individuals.

$$P = (I_1, \ldots, I_\mu).$$

Fig. 1 shows a schematic overview of the evolution strategy. In the following sections we will explain the single steps of the algorithm in more detail.

2.1. Initial population

The structures that build the starting population are initialised in a random way respecting several global constraints:

- The length of the unit cell vectors are restricted as follows: The lower limiting value is the largest covalent diameter of all atoms whereas the upper limit is defined by the sum of covalent diameters of the atoms in the unit cell. In addition, the minimal height of the parallelepiped of the unit cell is restricted to the same lower limit.
- The angles between the axes of the unit cell are free to vary between 45° and 135°, which is enough to represent all forms of crystal structures.
- The volume of the unit cell is limited to values between the volume of a close packed structure and the quadruple of it. If the