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Sinergia Objective

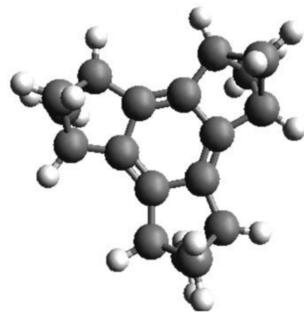
The objective of this initiative is to transform current heuristic approaches to analyzing the total, three-dimensional (3D) diffraction pattern of solid-state materials into a unified, efficient and conceptually revolutionized computational method that capitalizes on

- Advances in synchrotron and neutron physics,
- A new crystallographic description of disordered structures,
- Progress in modeling molecular and other materials, and
- New technologies in large-scale computing.

The project is a multi-disciplinary and international team effort between the University of Zürich, ETH Zürich and Oak Ridge National Laboratory (ORNL).

Finding a Model Structure

Tetracyclic benzene derivative, tris(bicyclo[2.1.1]hexeno)benzene, exhibits



- Molecule shows threefold symmetry
- Molecules arrange in equal layers with various stacking options leading to streaks of diffuse scattering perpendicular to the stacking axis (Fig. 1)

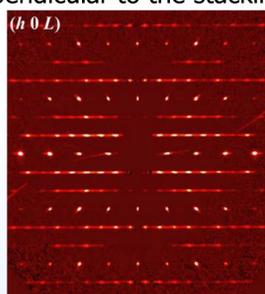


Fig. 1. Bragg peaks and diffuse streaks parallel to the stacking axis c^* in tris(bicyclo[2.1.1]hexeno)benzene.

Developing a Disordered Stacking Model

We build a model crystal of faulted stacking of translationally equivalent layers in Fig.2.

- Stacking probabilities (4)
- c
 - del (angle conformation)
 - e_2
 - tilt
 - $t=c$
 - $e_1=1-2*c$

- Atomic displacement parameters (3)
- U_1, U_2, U_3

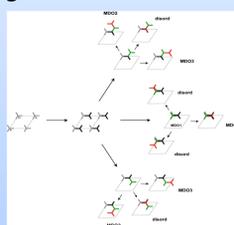


Fig. 2. Stacking model options of 4 symmetric layers

Computing Resources

- Presently hundreds of cores of TeraGrid Ranger or the Oak Ridge Institutional Cluster are used for hours to days to optimize the model parameters
- Since evaluating the fitness of a genetic population and its clones can be done in parallel and each evaluation takes over five minutes on current processors, this should be an embarrassingly parallel application.



Modeling Diffuse Scattering with a Probabilistic Crystal Builder

- Conventional structure analysis only models the average structure contributing to Bragg intensities. To construct a model that simulates the diffuse scattering contribution we are using a combination of Monte Carlo and Differential Evolution modelling techniques.
- Parameters: stacking probabilities, geometrical parameters, ADPs
- Simultaneous construction of 40 crystals (= layer stackings with 10^5 layers), each with a different set of parameters (= genes). Calculation of intensities
- Different model crystals produced from a given set of disorder parameters (clones) differ, the fit to the data of such clones differs too
- Improved model fitness using clones during optimization
- Gene mutation and recombination by Differential Evolution (Fig. 3). Select against experimental intensities (R) to optimize parameters.

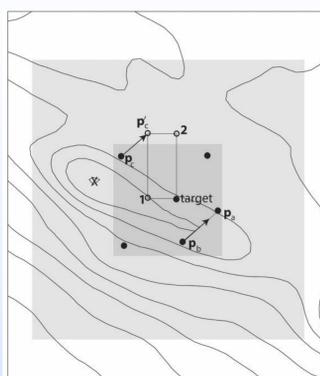


Fig. 3. Differential Evolution: p_a, p_b, p_c are randomly chosen from parent generation.
 $p_c' = p_c + 0.7(p_a - p_b)$
 Child (1 or 2) is uniform crossover of p_c' and target.

Clones

- Best phenotype (Fig. 4) of a suboptimal genotype will bias the refinement
- Refinement converges faster (Fig. 5) with clones
- More scalable with clones

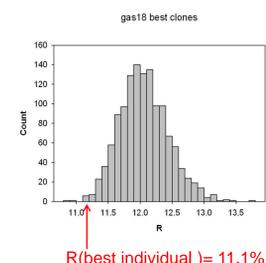


Fig. 4. Histogram of 1280 clones of best fit after 287 generations.

	gas18	gas18 20 clones (gap1)
mean R(%)	11.5(1)	11.4(2)
R(I _{mean}) (%)	11.8	11.25
Generations	287	137

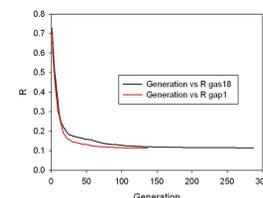


Fig. 5. History of Differential Evolution without clones (blue) and with clones (red).

Lessons Learned

- Think of scaling when developing the code (Fig. 6)

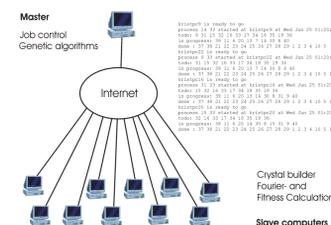


Fig. 6. As a starting point, a code used on distributed workstations to study the stacking disorder of a tetracyclic benzene derivative was ported to the TeraGrid's largest parallel computers.

- Use computer languages available on high performance computers: C++, C, or Fortran.
- Minimize input and output (Fig. 7)

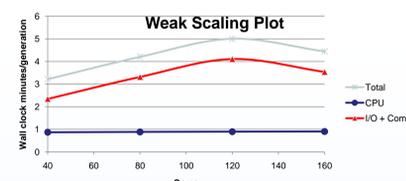


Fig. 7. Distributed workstations do lots of I/O better than high performance computers.

- Use MPI or hybrid MPI/OpenMP for parallelism
- Test if using FFTs calculated from CUDA libraries or FFTW is faster than Fourier Transforms
- Make code development more synergistic with SVN and tickets
- Use standard software for genetic optimization

New Code: ZODS Zürich Oak Ridge Disorder Simulation

- A next generation parallel code is in development by this collaboration to simulate disordered crystals
- Optimizes parameters describing the disorder on the basis of diffuse scattering data
- Provides tools for analyzing the disordered crystals
- Designed to scale on high performance computers
- Flexible to simulate a broad range of disorder models

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