Some new directions in random structure search

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Over the last decade, high pressure research has been transformed by the ability to predict both the structures and properties of materials under extreme compression from first principles. In many cases these predictions have been later confirmed by experiment. In others they have provided fruitful new directions to explore.

This progress has been achieved through the combination of stochastic approaches with reliable and efficient first principles methods. Diverse ensembles of initial structures can be generated, and structurally optimized. The resulting low energy structures are candidates for stable, and metastable, phases and/or defects and interfaces that might be experimentally realized. Success, of course, depends on a sufficiently broad and thorough sampling of configuration space.

A purely random strategy, as employed by Ab Initio Random Structure Searching (AIRSS), [1,2] is entirely *parallel,* and a natural fit to the high throughput computation (HTC) paradigm. Challenging cases can be tackled by designing the initial random structures so that they focus the search in regions of configuration space that are anticipated to yield success.

The design of these random "sensible" structures will be explored, along with some new directions [3] which promise to accelerate random search, and recent applications to high pressure research.

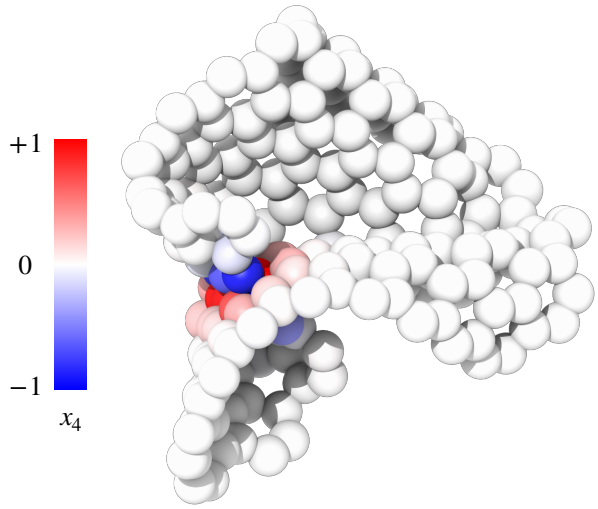

Figure 1. A graphene mat, part way through a hyperspatial optimization. The sheet apparently passes through itself in three dimensions, but in four dimensions it can untangle, and a low energy structure is obtained. The atoms are coloured according to how much they enter the fourth dimension.

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[1] C. J. Pickard, and R. J. Needs, *Phys. Rev. Lett.*, **97** (4), 045504 (2006) & JPCM, **23**(5), 053201 (2011)
[2] Released under the GPL2 license: `http://www.mtg.msm.cam.ac.uk/Codes/AIRSS`
[3] C. J. Pickard, "Hyperspatial optimization of structures", *Phys. Rev. B*, **99**, 054102 (2019)