MS14 O1

The blind tests of crystal structure prediction. 15 years of development: where are we now? Graeme M. Day, *Department of Chemistry, University of Cambridge, E-mail: gmd27@cam.ac.uk*

Keywords: crystal structure prediction, molecular crystals, computer simulation

The ambitious goal of predicting the crystal structure of a molecule, given only a molecular diagram, is a great challenge for the molecular modelling community. The problem has led to the development of a variety of methods, both for sampling the space of all possible close-packed crystal structures and for assessing the most likely structure(s) from the subsequent list of possibilities.

Blind tests of crystal structure prediction have been organised by the Cambridge Crystallographic Data Centre as an objective evaluation of existing methodologies. For these challenges, molecules were chosen whose crystal structures were previously unknown. Their crystal structures were determined and withheld by an independent referee for the duration of the test. Diagrams of the molecules were presented to the community and participants asked to submit their three most likely crystal structures for each; these predictions were then compared to the experimentally determined structures.

Previous blind tests [1] have highlighted the main challenges in crystal structure prediction. For example, there has only been one successful prediction for a conformationally flexible molecule in all three earlier challenges. Some results from the recently completed fourth blind test are presented here, in the context of previous blind tests and recent advances in methodologies.

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MS14 O2

Molecular structure and properties: Looking at hydrogen bonds. Enrique Espinosa[a], Ignasi Mata[b], Ibon Alkorta[c], Elies Molins[b], [a]*LCM3B, University Henri-Poincaré, Nancy, France.* [b]*ICMAB-CSIC, Barcelona, Spain,* [c]*IQM-CSIC, Madrid, Spain.* E-mail: enrique.espinosa@lcm3b.uhp-nancy.fr

Keywords: Hydrogen bonds, Topological analysis of the electron density, Energetic properties

Molecular structure is determined by nuclei positions. From the Hellmann-Feynman theorem [1] we know that the exact ground state electron density distribution $\rho(\mathbf{r})$ depends on the nuclei positions only. Furthermore, the Hohenberg-Kohn theorem [2] states that the total energy of a system can be written in terms of this distribution. In that context, the relationship between the structure of a molecular system and its physical and chemical properties, in particular energetics, should be reflected by the electron distribution. Accordingly, hydrogen bonding interactions described in terms of the electron density distribution can be considered as a fundamental subject of study to get insight on their structure-properties relationship, as the former, $\rho(\mathbf{r})$, is a conceptual bridge between the latter. On the other hand, the topological analysis of $\rho(\mathbf{r})$ developed by Bader and co-workers [3] is a very useful tool for characterizing atomic interactions in internuclear regions. Thus, within this framework, the structure-properties relationship of hydrogen bonds will be discussed in terms of the topological analysis of experimental and theoretically calculated electron density distributions that have been characterized in hydrogen bonding regions [4-9].

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MS14 O3

Crystal structures of moderately complex organic molecules are predictable Marcus A. Neumann, *Avant-garde Materials Simulation, 30 bis rue du vieil Abreuvoir, 78100 St-Germain-en-Laye, France.* E-mail: sarlams@aol.com

Keywords: crystal structure prediction, lattice energy ranking, force field parameterization

A comprehensive computational strategy for the prediction of crystal structures with an unprecedented success rate is presented. Key components are a hybrid method [1] for the accurate calculation of lattice energies, a robust procedure for the parameterization of tailor-made force fields and a novel approach for crystal structure generation. The hybrid method combines DFT calculations by means of the VASP program [2-4] with an empirical van der Waals correction. It is used for the final lattice energy ranking and acts as a reference standard for force field parameterization. A tailor-made force field is derived for each molecule for which the crystal structure is to be predicted. The apparently simple force field involves bond increments, isotropic van der Waals potentials and essentially uncoupled intramolecular energy terms, but the mathematical framework of the force field and the fitting procedure have been designed to allow for a maximum amount of customizability while avoiding redundancy. Hundreds of force field parameters can be optimized simultaneously and typical disagreements between the tailor-made force field and the hybrid method lie in the range of 0.015-0.03 kcal/mol per atom. The tailor-made force field is used for crystal structure generation and for the preparation of Hessian matrices for the final lattice energy optimization with the hybrid method. Based on the known statistical deviation between the tailor-made force field and the hybrid method, a shortlist of crystal structures from a small energy window is selected for the final optimization.

Energy ranking results for 15 organic molecules are presented, including ethane, ethylene, acetylene, methanol, urea, acetic acid, cyclohexane-1,4-dione, paracetamol, CCDC blind test molecules I-VI and a pharmaceutical compound for which crystal structures have been predicted in a blind test fashion. In 14 out of 15 cases, the experimental crystal structure is found among the two most stable predicted crystal structures. The planned implementation of a free energy correction is expected to improve the success rate even further.

All calculations have been carried out with the development version of a novel software package, GRACE (Generation, Ranking And Characterization Engine), which uses VASP as an external component.

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MS14 O4

Stationary state equilibrium conditions for the electron density distribution Julian Henn, *Department of Inorganic Chemistry, University of Göttingen, 37077 Göttingen, Germany* . E-mail: julianhenn@web.de

Keywords: mem, multipole refinement, accuracy

Stationary state equilibrium conditions describing the electron density distribution in equilibrium (not necessarily the ground state) as derived from Levy and Perdew [1] *via* the Hellmann-Feynman theorem [2] may be employed to put the experimental electron density reconstruction process on a self-consistency scheme. This should increase the accuracy in present day multipole refinements [3] and maximum entropy methods [4].

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MS14 O5

Refinement of a one electron density matrix from Compton and Bragg pseudo data : the diatomic molecule case J.-M. Gillet, *Ecole Centrale Paris, laboratoire SPMS UMR8580, Grande Voie des Vignes, 92295 Chatenay-Malabry,France.*
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Keywords: Density, Diffraction, Compton

High resolution x-ray diffraction, convergent beam electron diffraction, deep inelastic x-ray scattering (Compton scattering), e-2e and γ-e-γ spectroscopies are all directly related to the One-electron Reduced Density Matrix (1-RDM [1] On the other hand, it is well known [2][3] that the 1-RDM contains all the information about the electronic structure available at the one-electron level. Unfortunately, and to our best knowledge, few attempts for refining 1-RDM models have been carried out [3]-[6]. With the exceptions of Schmider [7][8] for atomic systems, and Schulke and co-workers [9]} only x-ray diffraction data were employed as experimental references.

The purpose of this talk is to discuss to what extent the successfull decomposition of the electron density into aspherical pseudo-atomic contributions [10] can be adapted to the 1-RDM case. Furthermore, we intend to show that, with such a model, the complementarities between very different experiments can be better exploited through a joint refinement.

Emphasis will be put on the difficulties which are specific to a joint refinement of 1-matrices from different experimental data as opposed to usual "mono-experimental" refinements..

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