Theory & Simulation at the CCMMP

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CCMMP

- Organic semiconductors
- Functional materials
- Carbon, nanomaterials, biomaterials
- Disordered materials
- Method development

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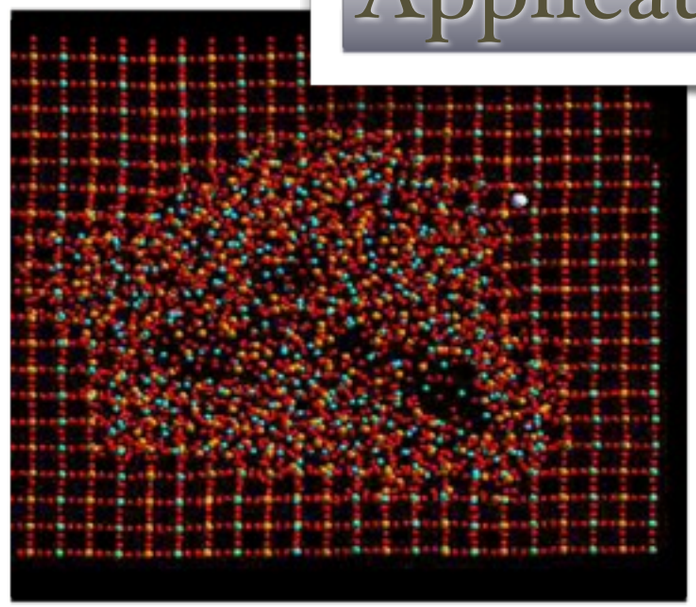
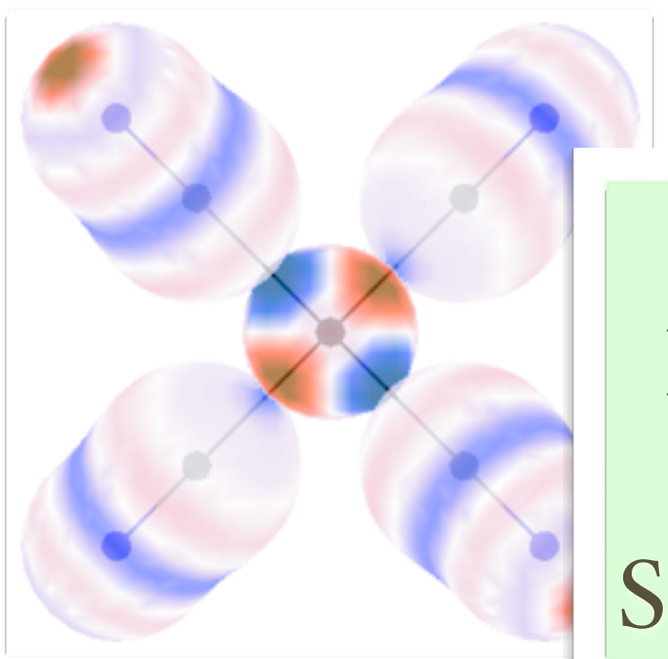
Simulation Tools

GULP
DL_POLY

Experiment

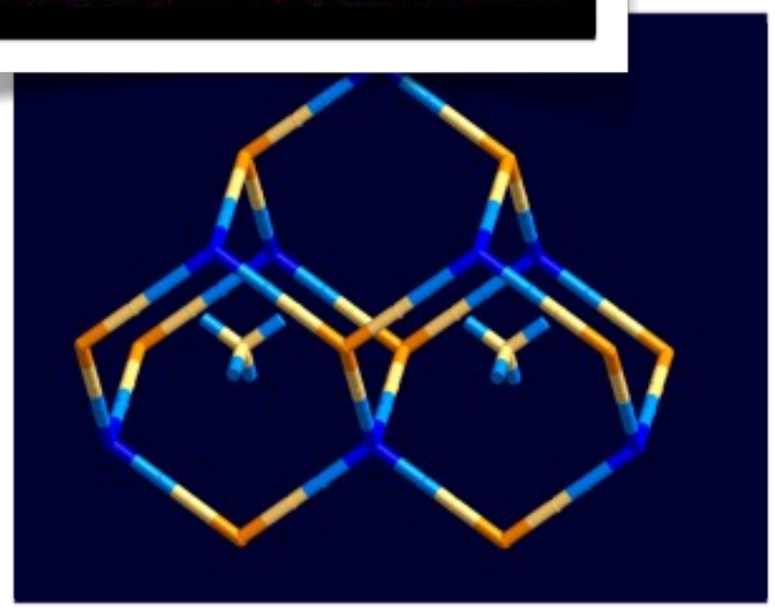
Applications





Methods

SAPT(DFT)
Interaction models
CamCASP
Statistical Mechanics



Large-scale simulations

Effect of radiation-induced amorphization on diffusion

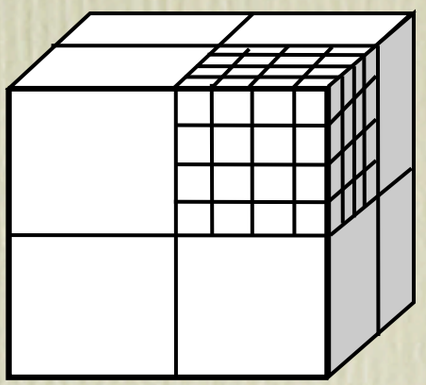
What are long-term (millions of years) effects of irradiation on the performance of waste forms?

Case study: zircon $ZrSiO_4$ found minerals are ~1 billion years old, completely amorphous yet intact

Absorbs large ions like Pu on Zr site

Molecular dynamics simulation of radiation damage

1. Empirical potentials and short-range ZBL potential at short <~1 Å distances



2. Almost perfectly scalable MD code based on domain decomposition strategy (DL_POLY 3 MD package)

3. Parallel computers (Cambridge HPC, HPCx, HECToR)

4. Adapted MD code to handle out-of-equilibrium conditions (variable time step, boundary scaling) and to analyze radiation damage on the fly

High-energy U recoils in $ZrSiO_4$

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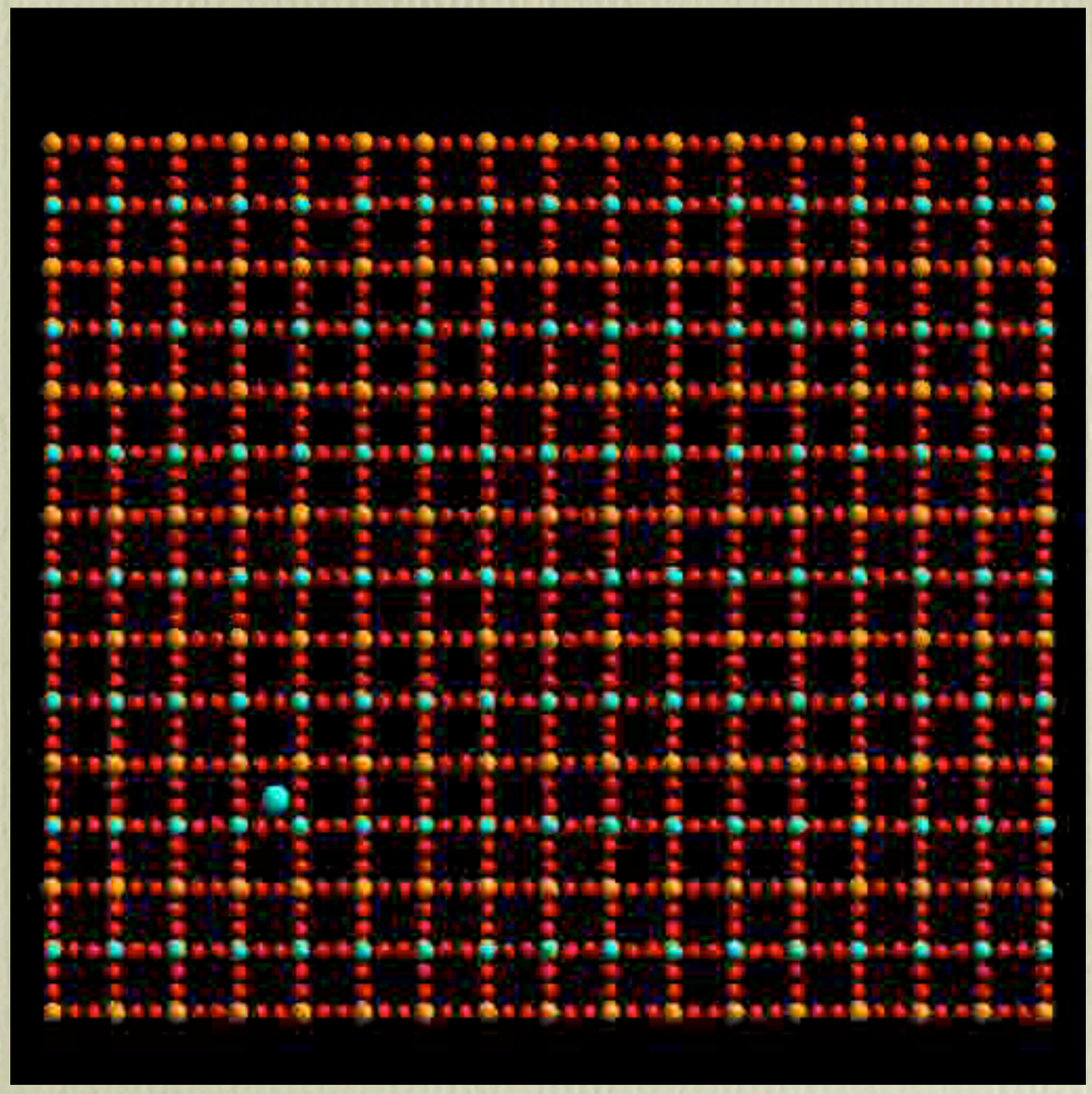

High-energy U recoils in $ZrSiO_4$

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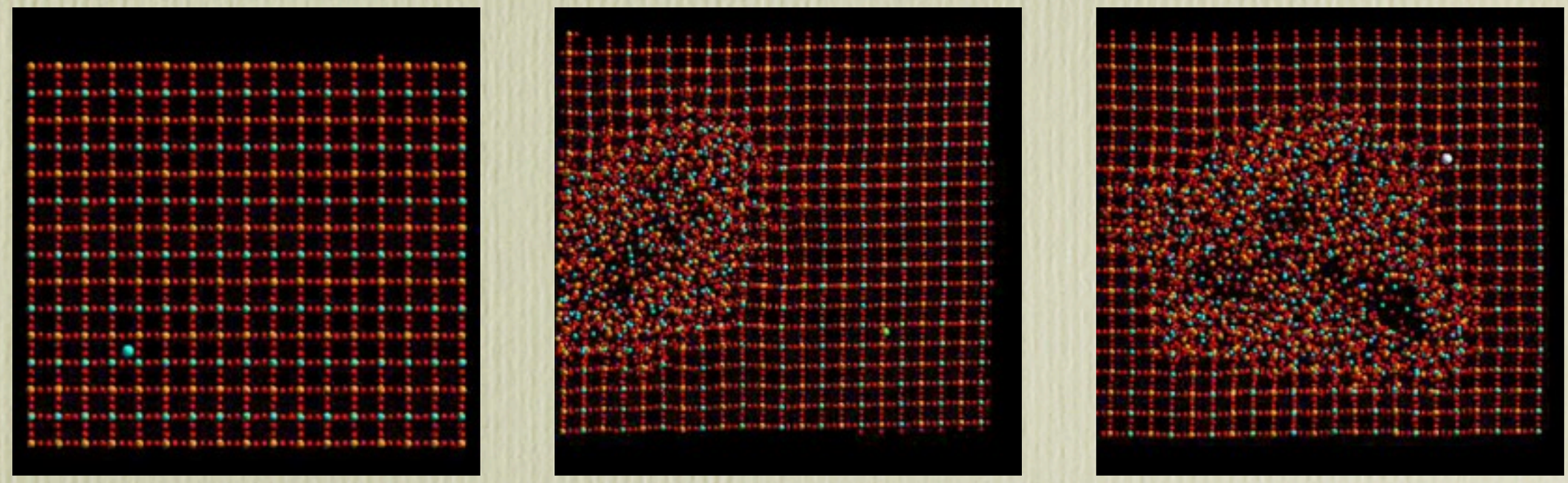
70 keV U recoil and their overlap



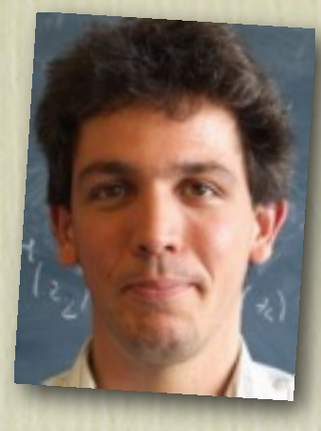
Channels of low density appearing along the track

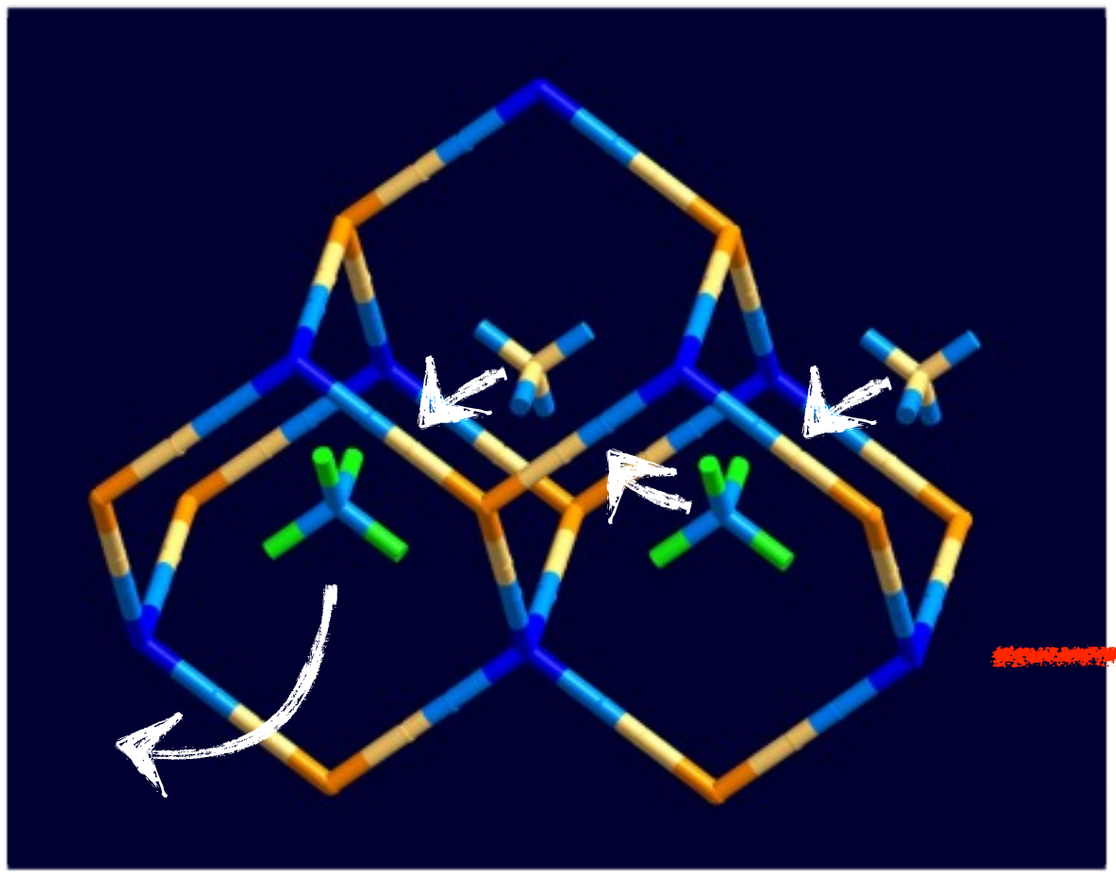
Frameworks

Local structure

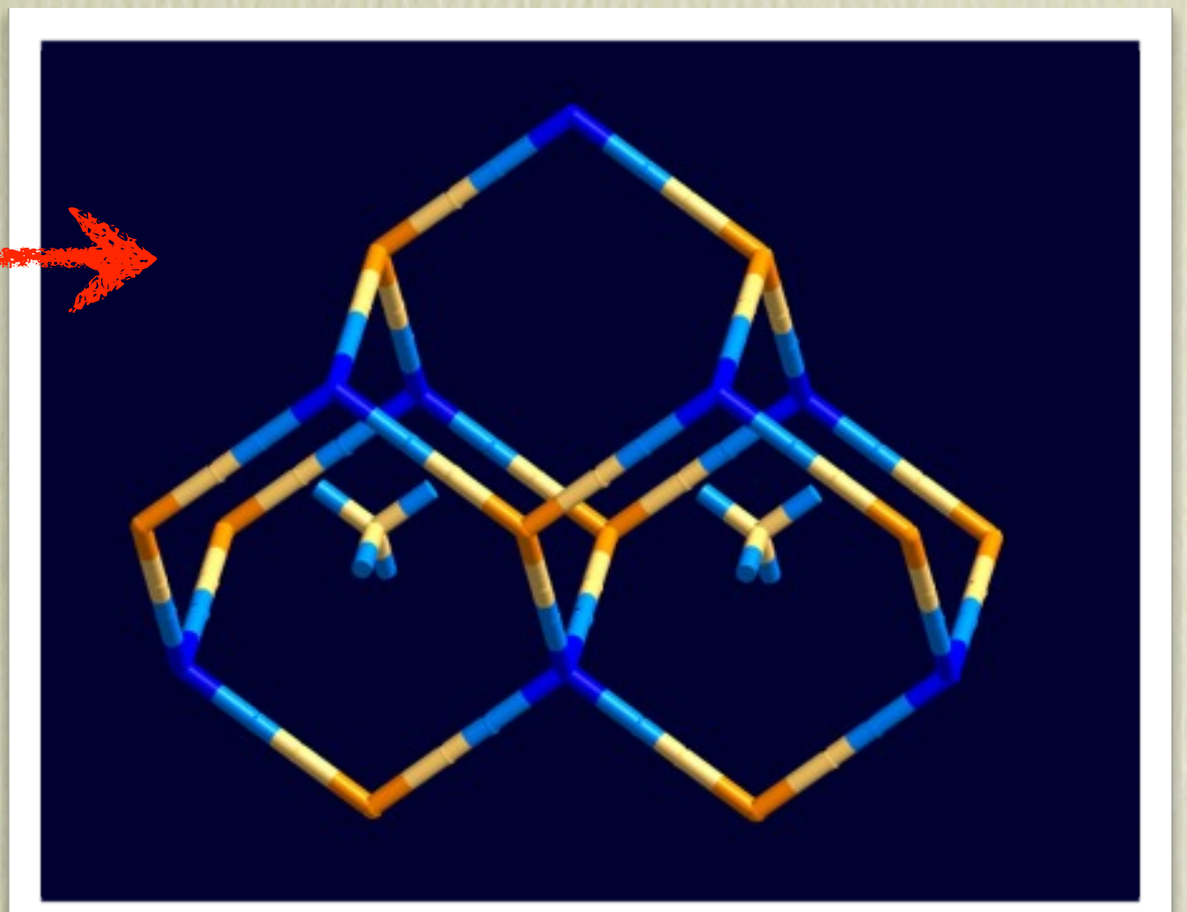
Organo-Metallic frameworks:

Guest rearrangement



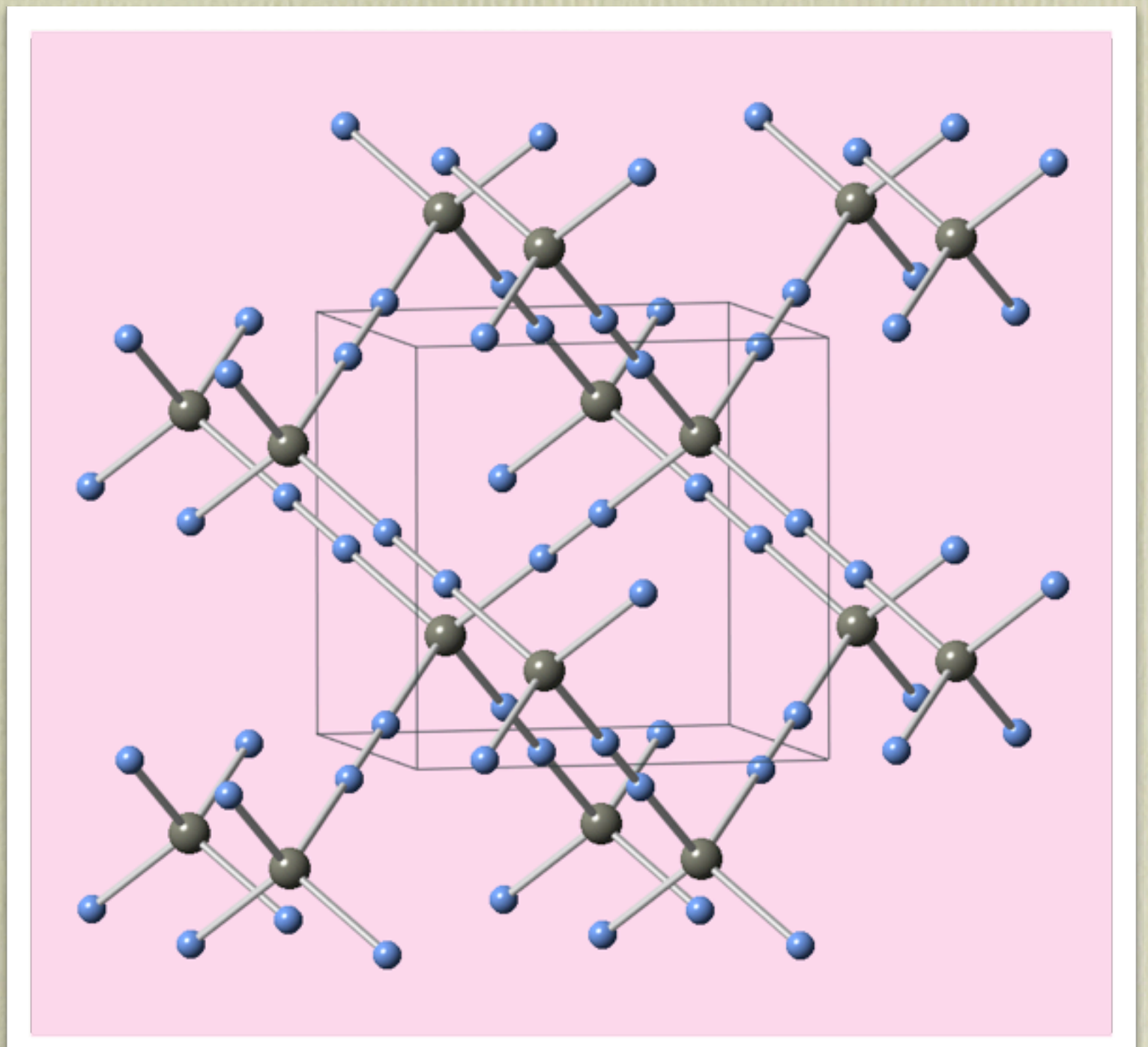


Enthalpic as well as entropic contribution to guest desorption

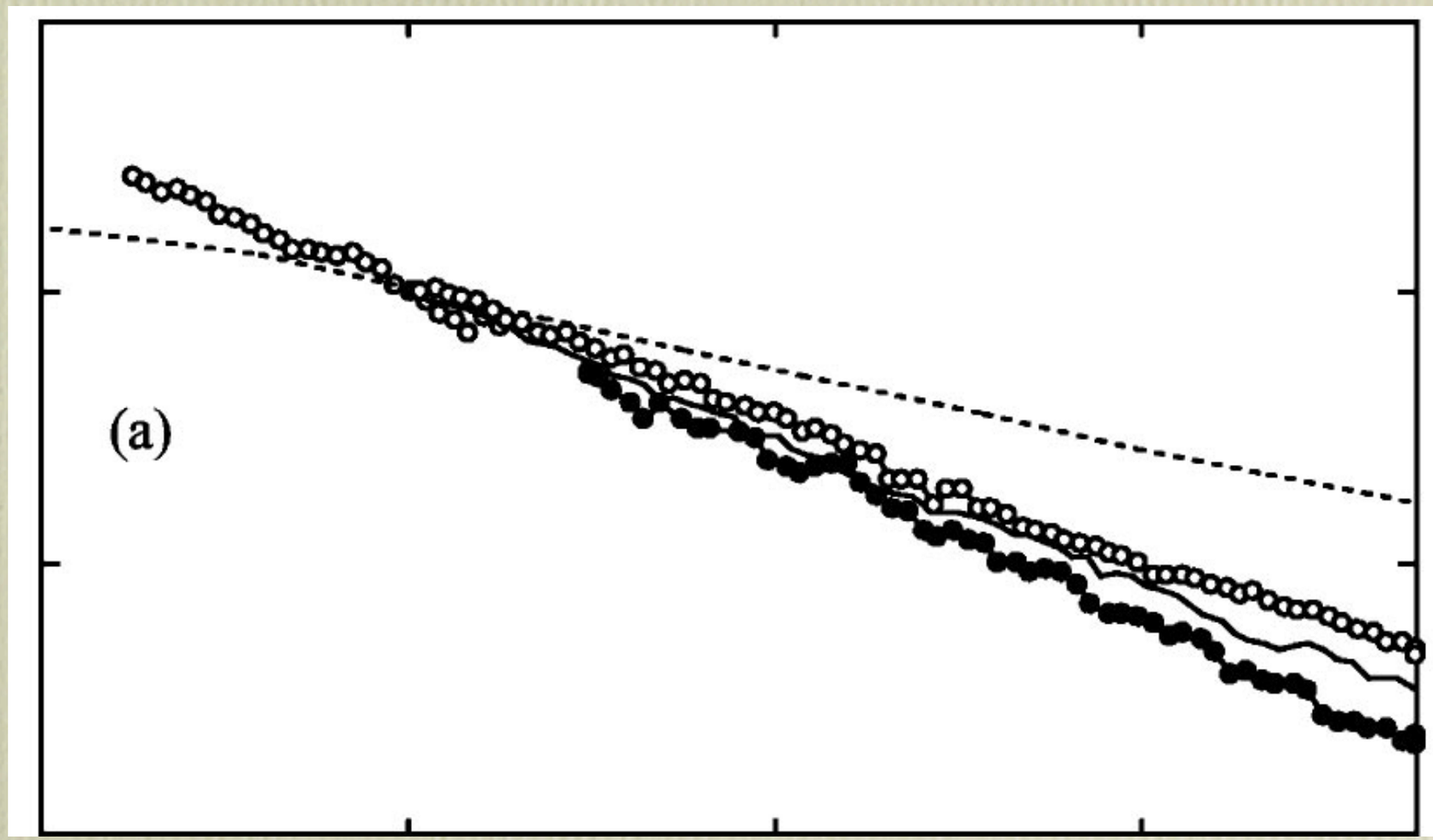


$CuZn(CN)_4 \cdot N(CH_3)_4 \cdot CCl_4$

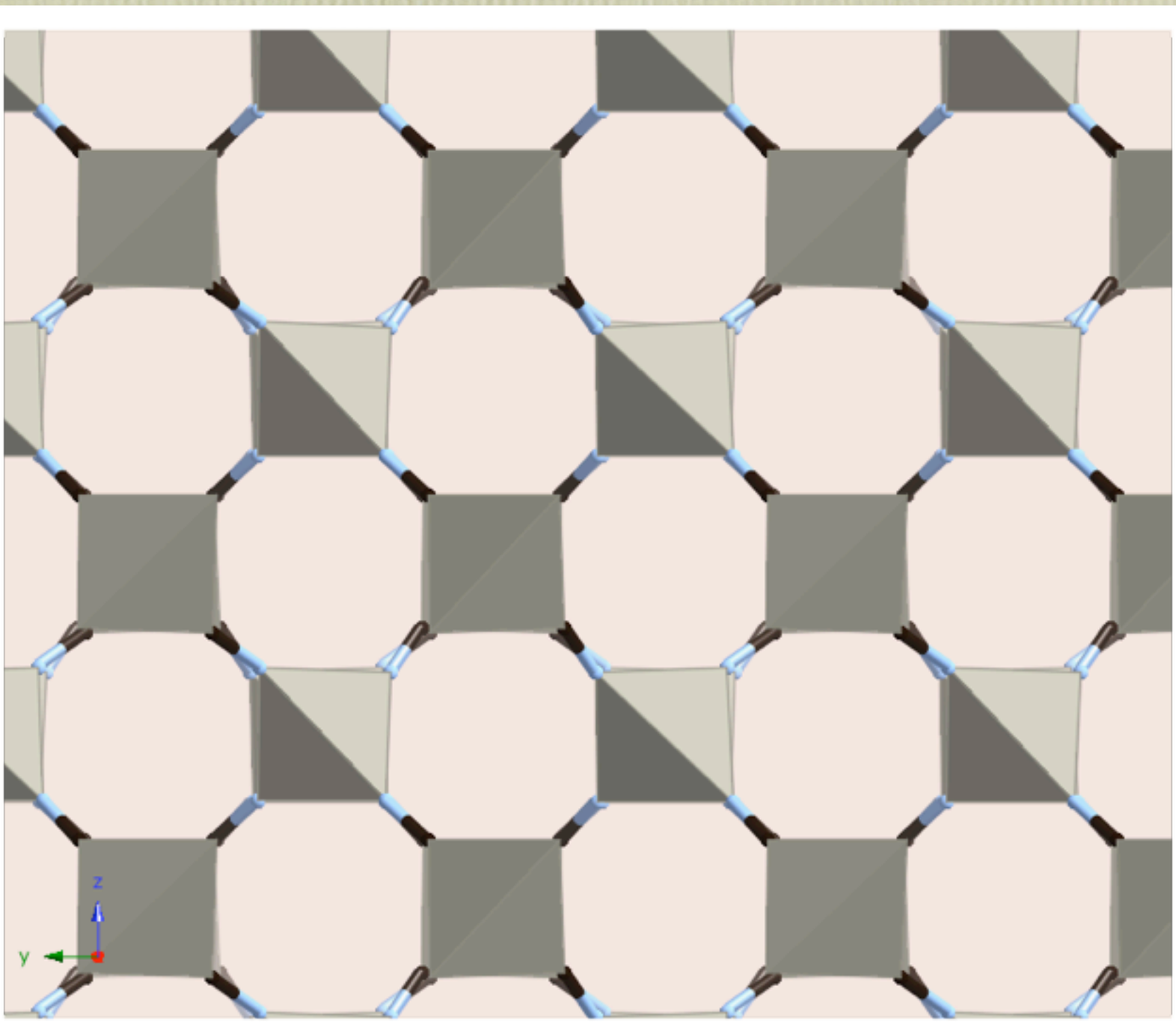
Zinc cyanide framework structure



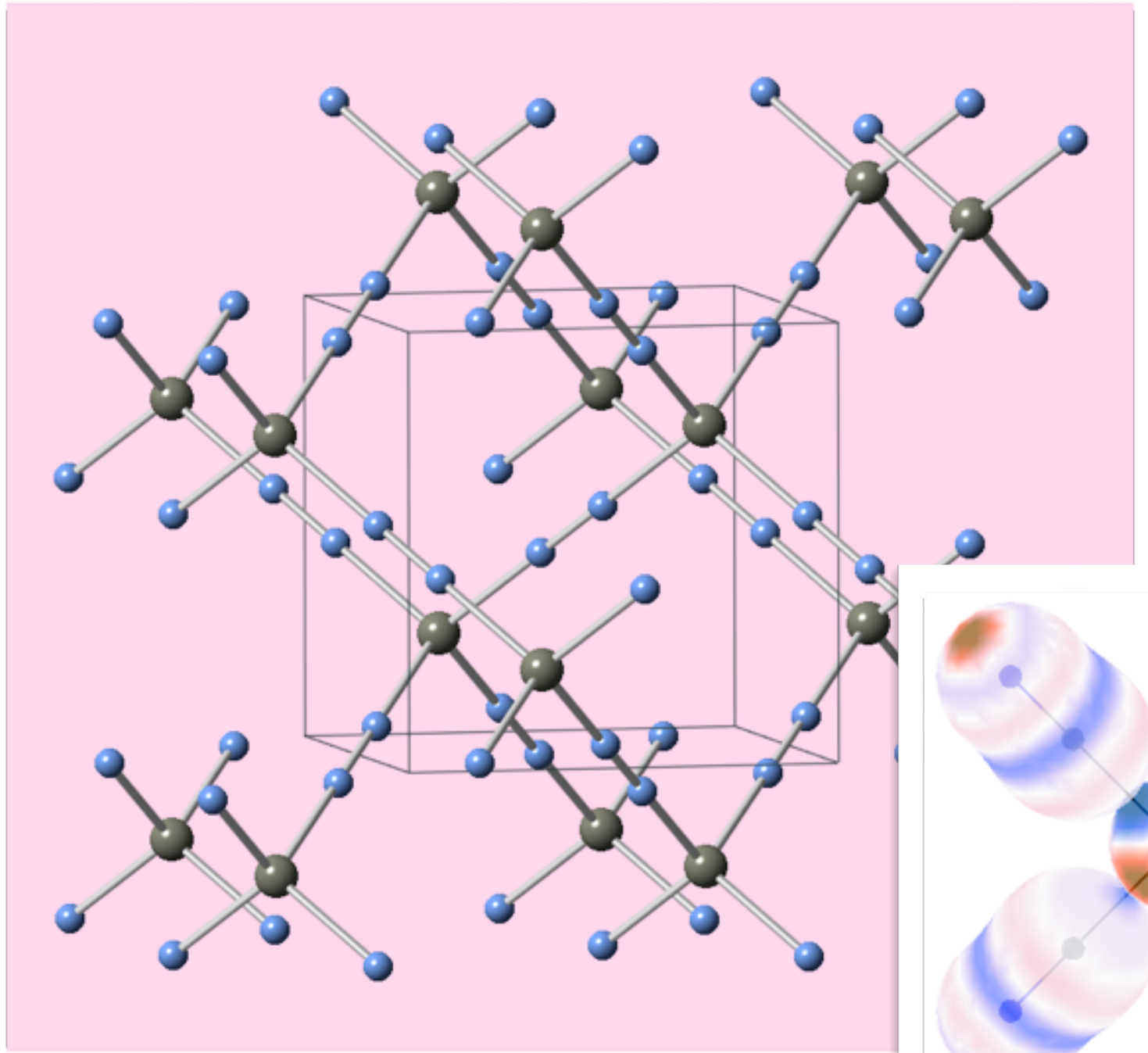
Negative thermal expansion in zinc cyanide, $Zn(CN)_2$



Important vibrations for negative thermal expansion

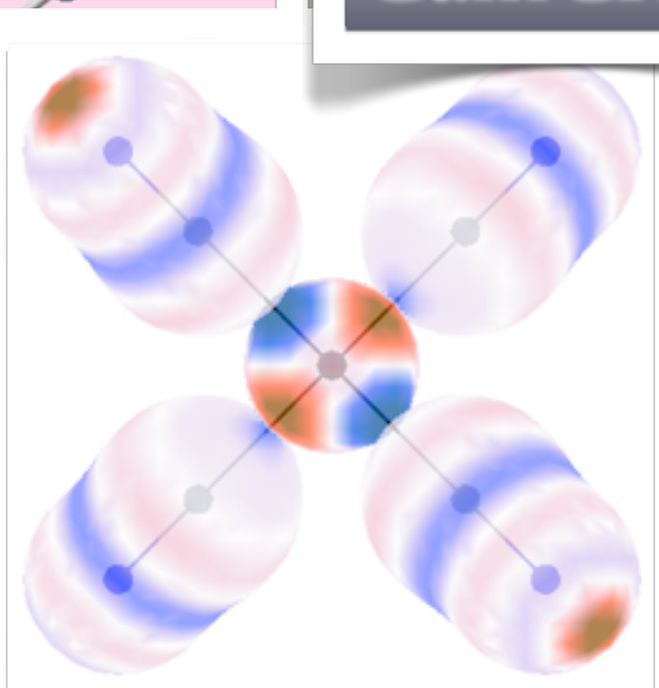


Accurate models : multipoles



GDMA

CamCASP



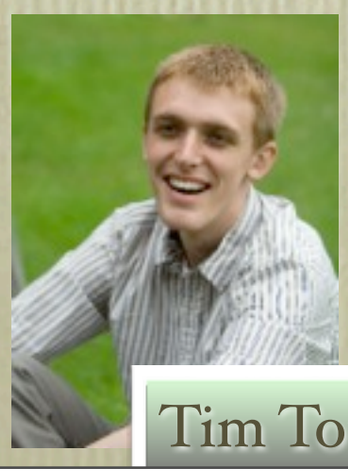
Nucleation

400,000 deaths in India alone

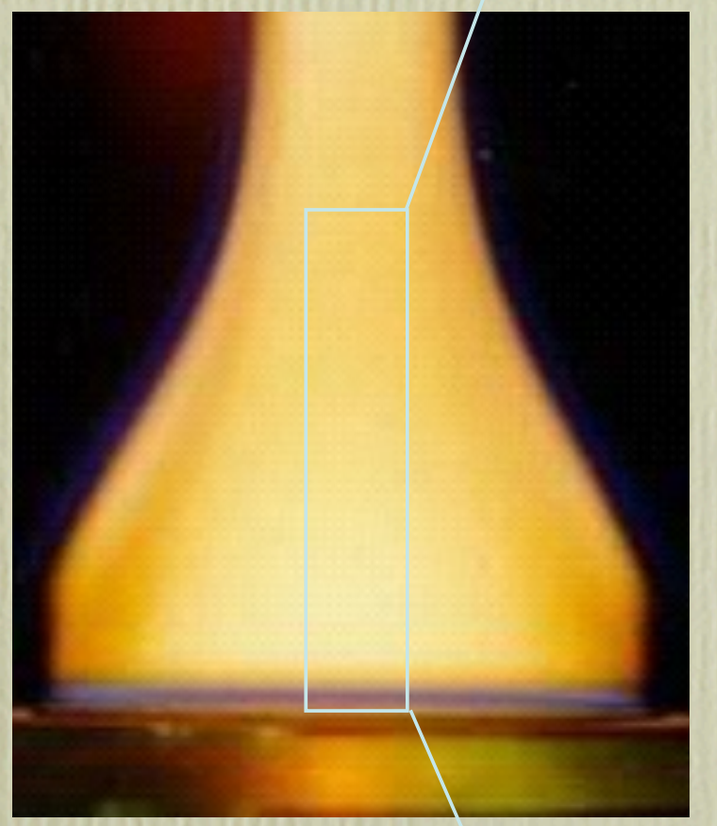
Smog at the Taj

Soot

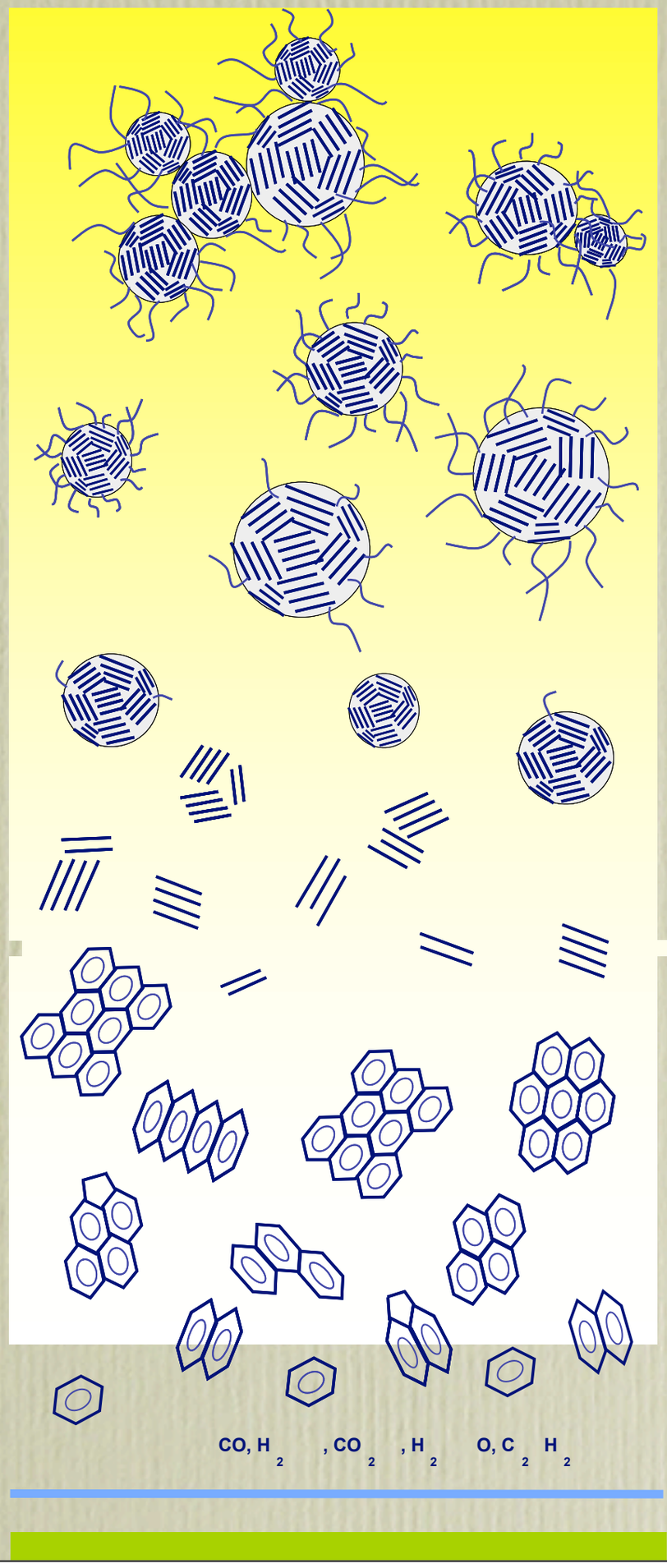
Black carbon particulates warm the upper atmosphere. (Myhre, Science 2009)



Tim Totton



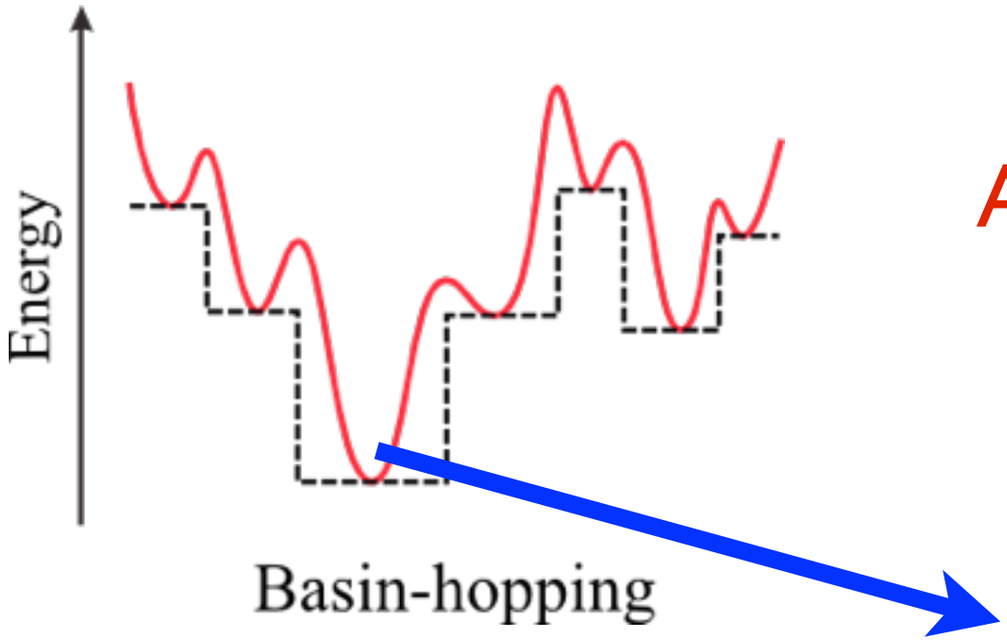
H. Wang, Proceedings of the Combustion Institute 33 (2011) 41-67



- Gas phase nucleation
- 1500K
- Soot inception thought to occur by pyrene dimerization
- PAHs thought to aggregate through van der Waals forces
- Structure of soot nuclei needed for combustion models
- Large uncertainty in models

PAHAP : Totton et al. (2010,2011)

CamCASP



Are coronene clusters stable at 1500K?

MD simulation of a
50-coronene cluster

Sublimation at 798.2K

Totton, Misquitta & Kraft, PCCP 2012

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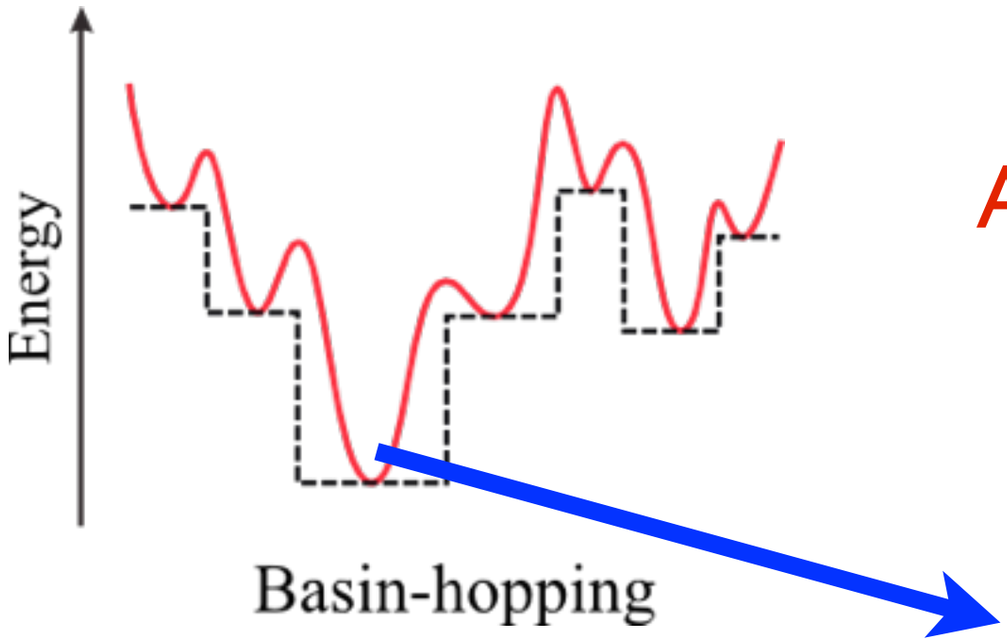

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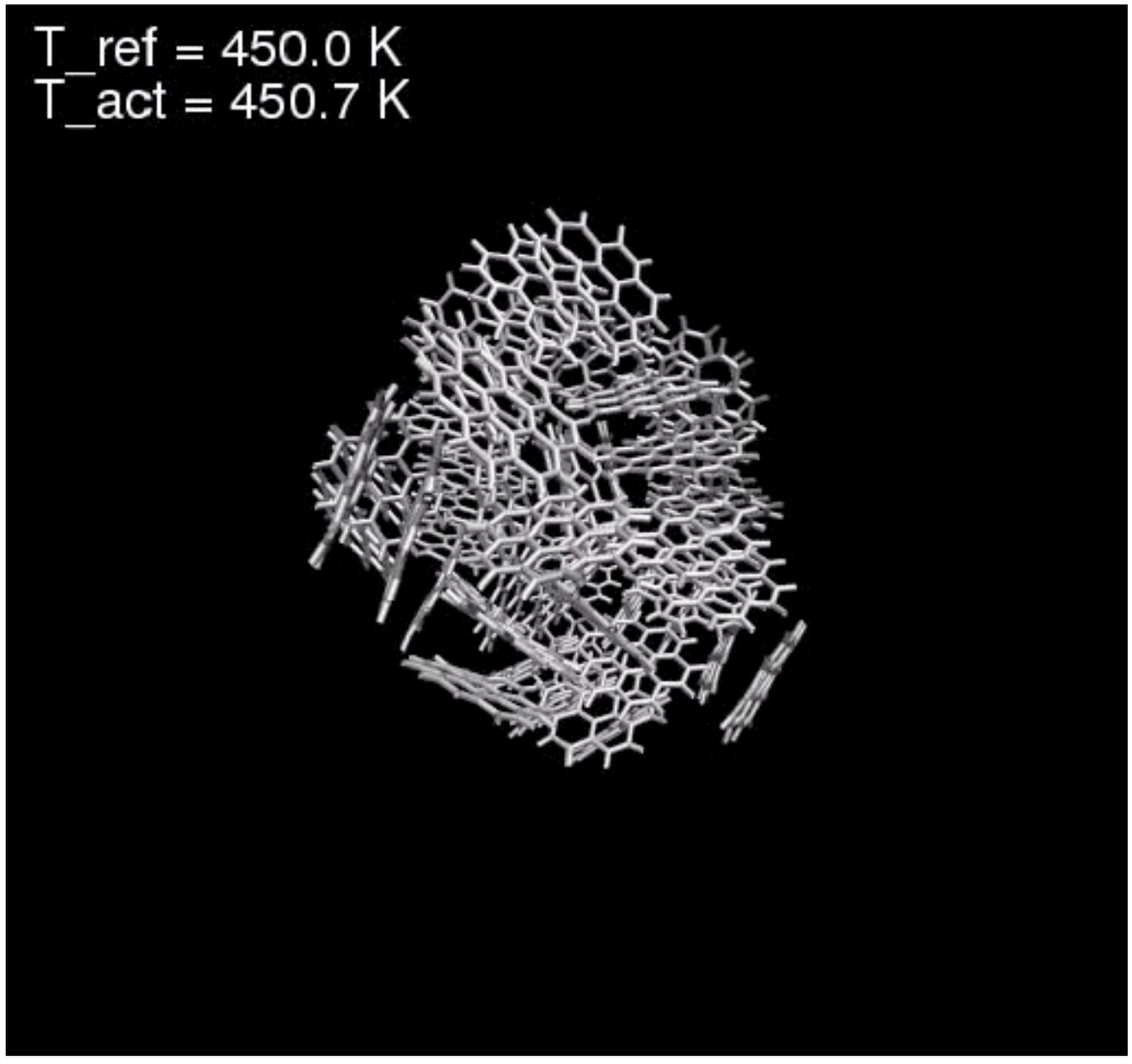

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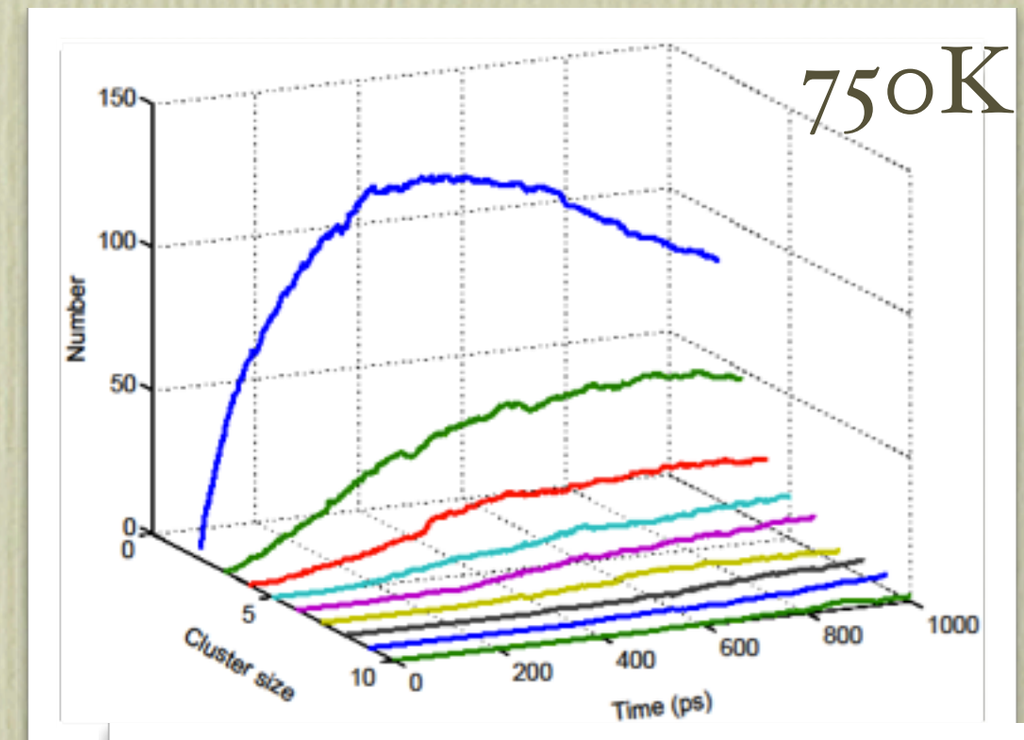
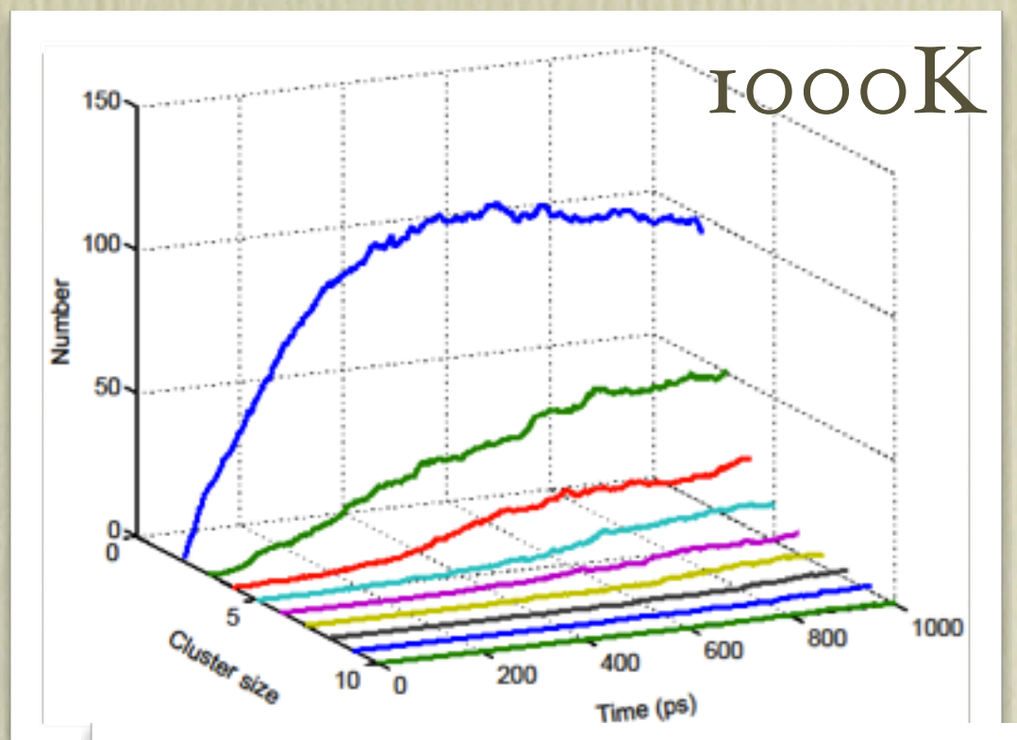
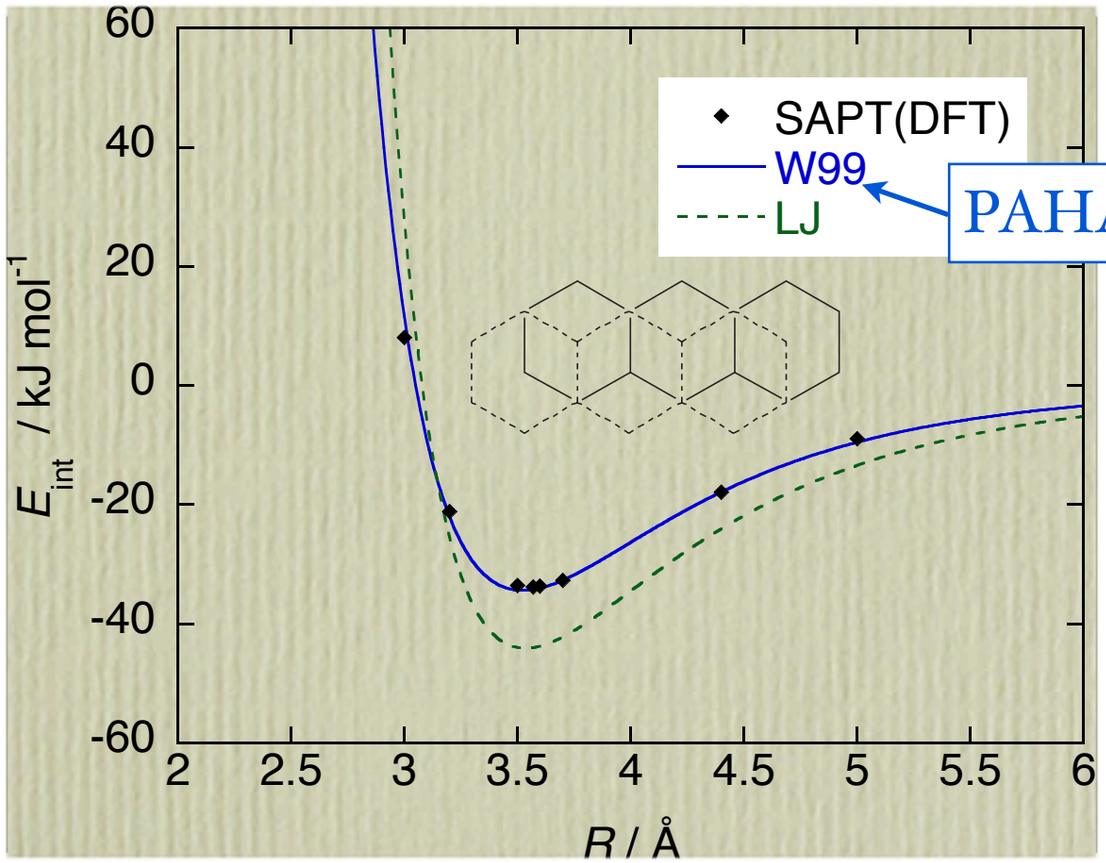
Circumcoronene ($C_{54}H_{18}$)

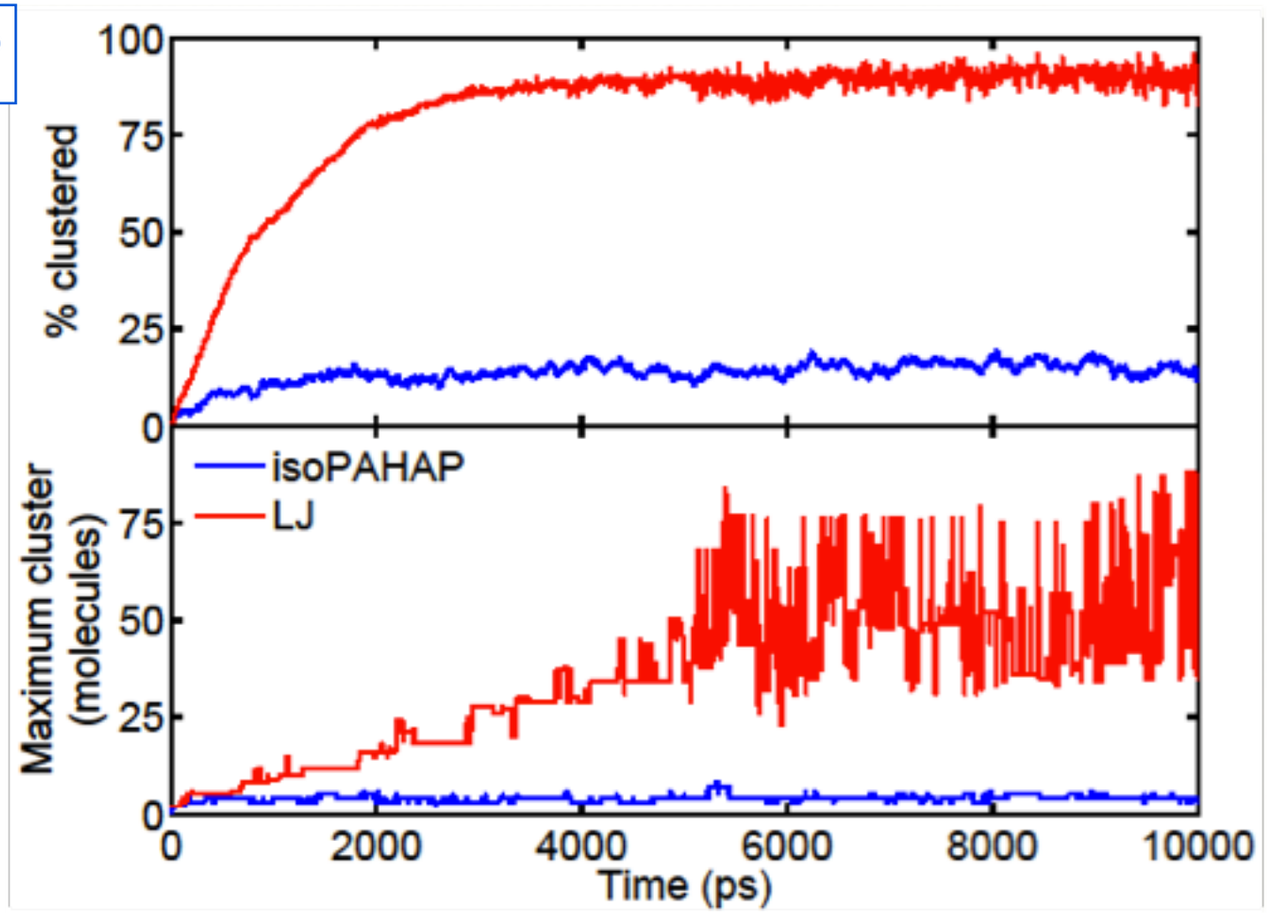




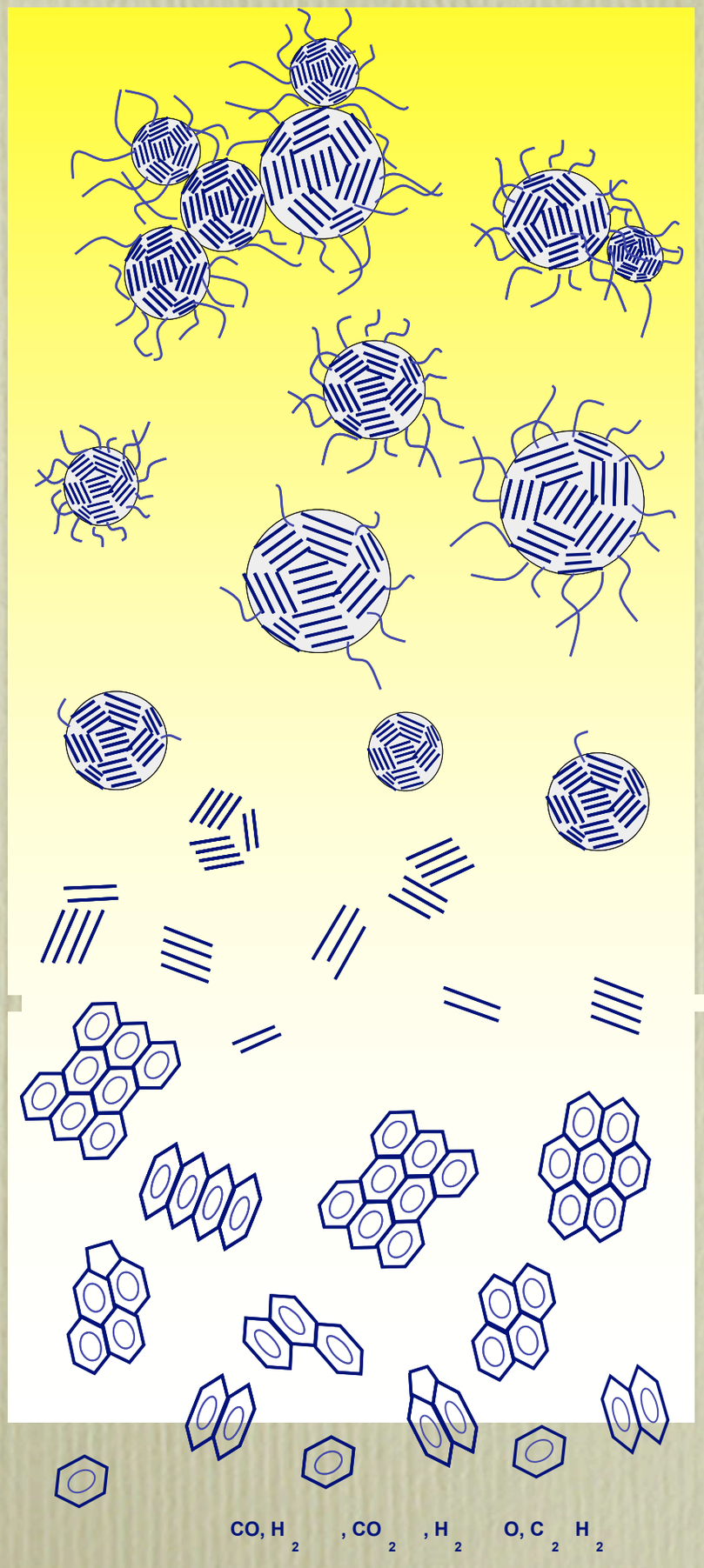
Totton, Misquitta & Kraft, hot article in PCCP (2012)

LJ versus (iso)PAHAP





The apparently small differences lead to an order of magnitude increase in clustering.



- Supported by experimental evidence from Sabbah et al. (2010) and Happold et al. (2007)
- BUT soot *does* form easily!
- Other kinds of mechanisms?
- Are the interactions ionic? Chemical?
- Side-chains could buffer collisions.
- Heterogenous nucleation? How?

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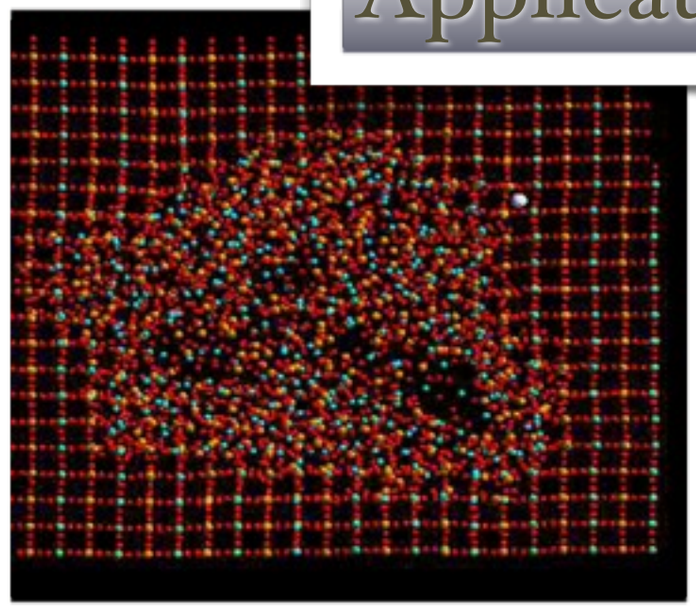

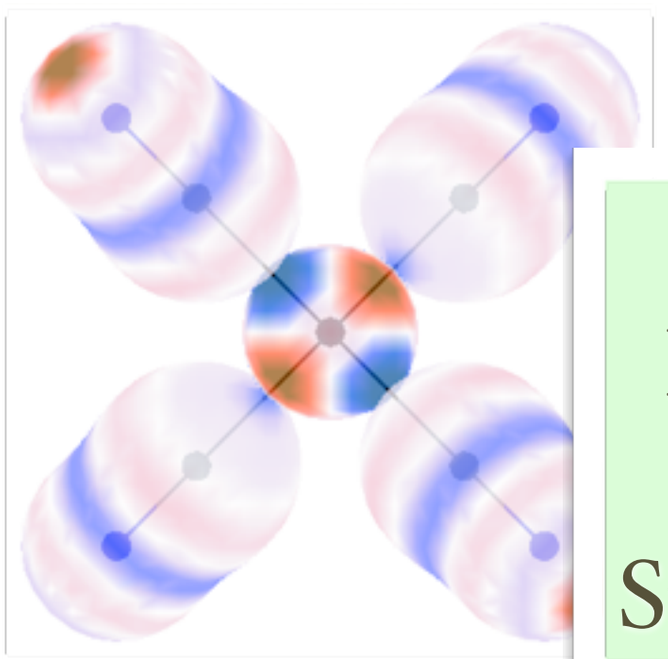

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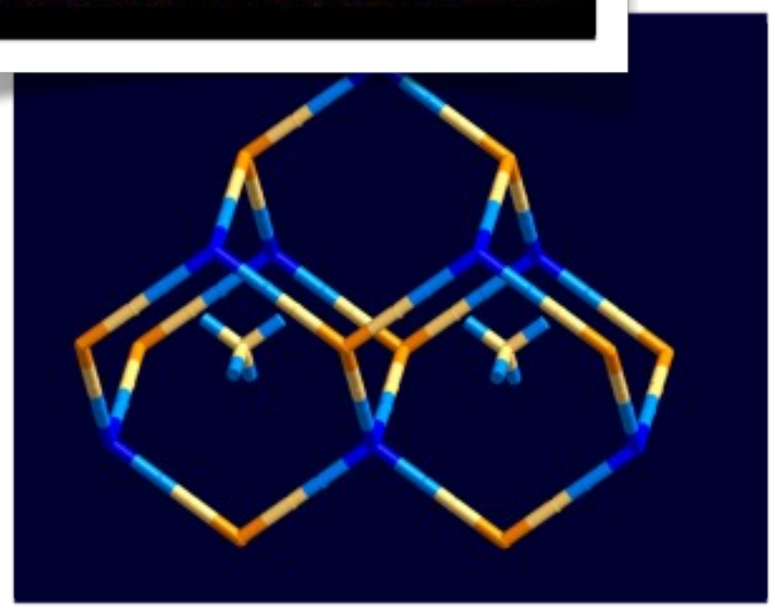

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- Within the CCMMP
- QMUL
- TYC
- ...and beyond!

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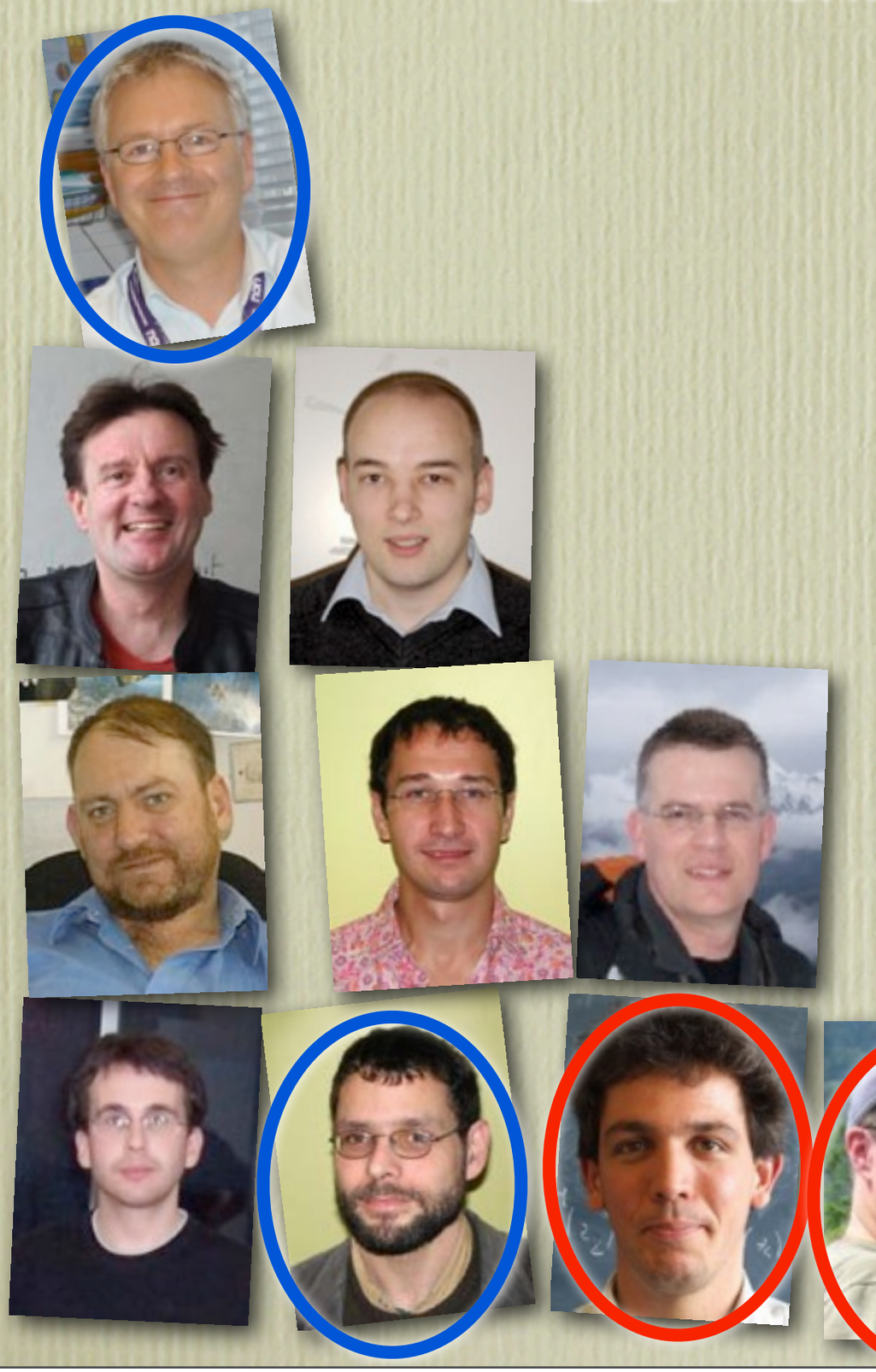

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