Getting started with UQ for plasma-material interactions

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National Science Foundation
WHERE DISCOVERIES BEGIN

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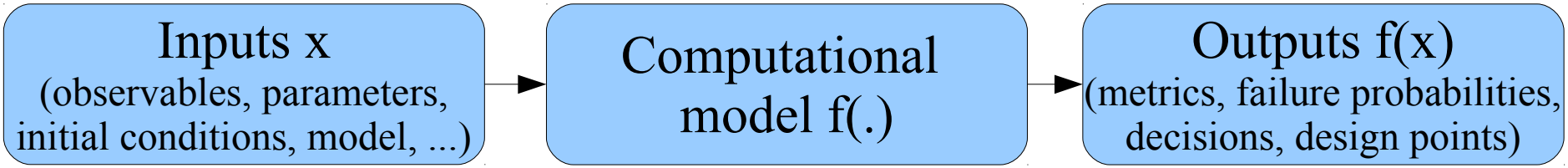
Outline

- Community agitation
- Overview of UQ
- Plasma-materials questions of focus
- Initial foray into the W-H MD potential

What is Uncertainty Quantification?

- Propagate uncertainties in input variables, parameters and models to quantify effects on output metrics
  - Essential for incorporating outputs of physical models into engineering design/decision processes
  - Guides research activities and investments
  - Rigorous derivation of coarse-graining schemes



Total uncertainty: modeling error + numerical error + statistical error

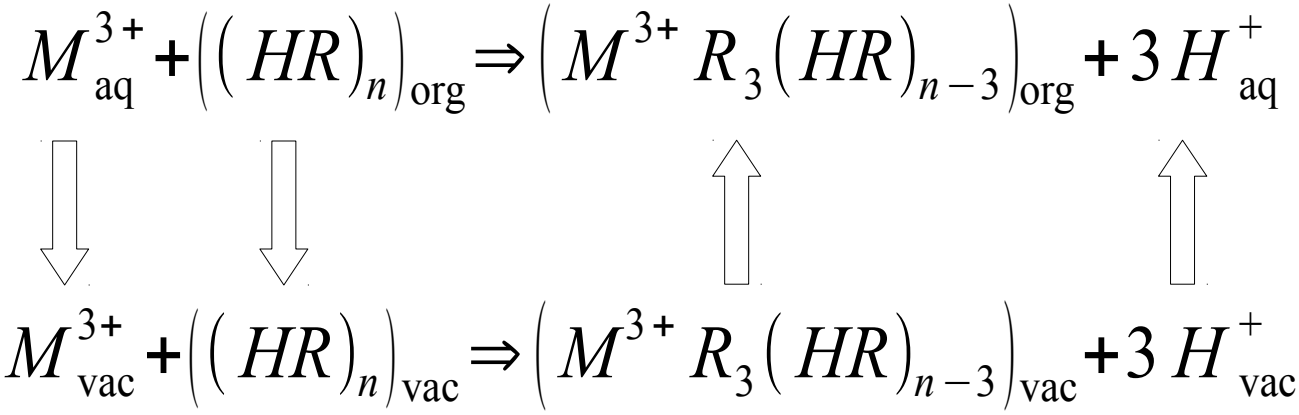
Chemical examples

- Kinetics

$$\kappa = A\, e^{-\Delta G/kT}$$

- Equilibria

$$K = \frac{A_f}{A_b} e^{-\Delta G/kT}$$



Community Agitation

- UQ is widely applied in engineering
  - e.g., nuclear reactor design, construction, ...
- and a few science domains
  - e.g., climate
- but is largely absent in the physical sciences
  - chemistry, materials, ...
  - a few groups are pursuing this, but it needs to become pervasive
- We are trying to help kick start this

Community Agitation

- Spoken with NSF and DOE program managers
  - Warm support (math, physics, fusion, chemistry)
- Workshop on UQ in physics/chemistry (or perhaps multiscale physics?)
  - Organizing committee so far: Gordon Drake, Petr Plechac, Daren Stottler, PK, RJH
  - Bring together mathematicians + scientists
  - Proposed for late spring 2015 in/near NYC
  - More info soon

A. Aleatory uncertainty

- Inherent or irreducible
  - E.g., weather, radioactive decay of atom, ...
  - Must be addressed for physically complete model
  - Often modeled with probability distributions

B. Epistemic uncertainty

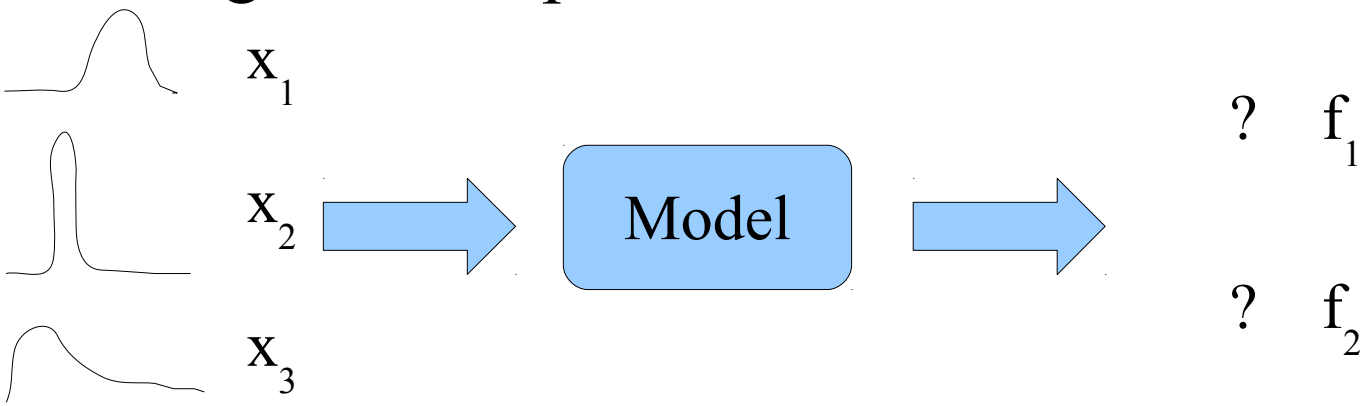
- Reducible (in principle)
  - State of knowledge
  - E.g., incomplete models, imprecise constants, ...
  - Must be addressed for uncertainty, validity regimes, model sensitivity, ...
  - Often modeled with intervals

Epistemic UQ techniques

- Interval analysis
  - Various statistical sample methods, non-intrusive
  - Accelerated by approximate models
  - Important to not interpret resulting distributions
- Evidence theory (e.g., Dempster-Shafer)
  - Combine belief and plausbilty
  - Treats imprecise, ambiguous, conflicting data
  - Potentially very expensive to apply?
- Other
  - (imprecise) probability theory, fuzzy sets, ...

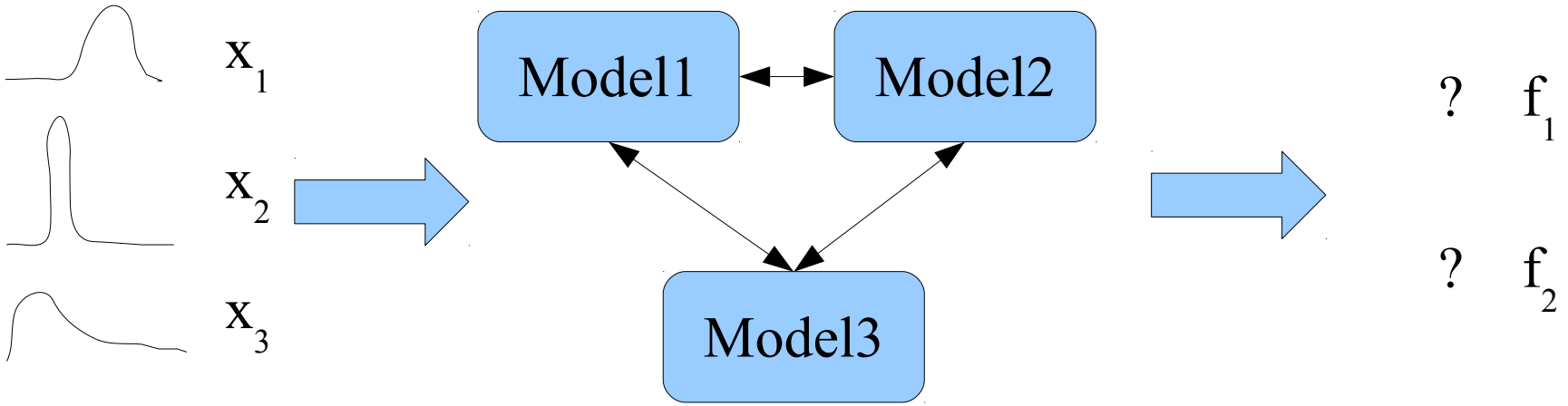
Aleatory UQ techniques

- Random sampling
  - Non-intrusive, but potentially expensive
  - Orthogonal/Latin sampling avoid exponential growth in number of samples; adaptive techniques
  - Correlation must be addressed
  - Means, variances and even full output PDF
  - Challenges: small probabilities, rare events, tails



Challenge with multiscale sampling

- Propagation of uncertainties within model
  - Tight coupling vs. parameter passing
  - Possibly massive increase in expense
  - Intrusive approaches possibly helpful



Initial plasma material foci

- How robust is the simulation observation that H deposition on W forms a passivation layer under certain operating conditions and time scales?

- Bound the distribution function for depth penetration of H in W as a function of energy, preparation, structure, etc.

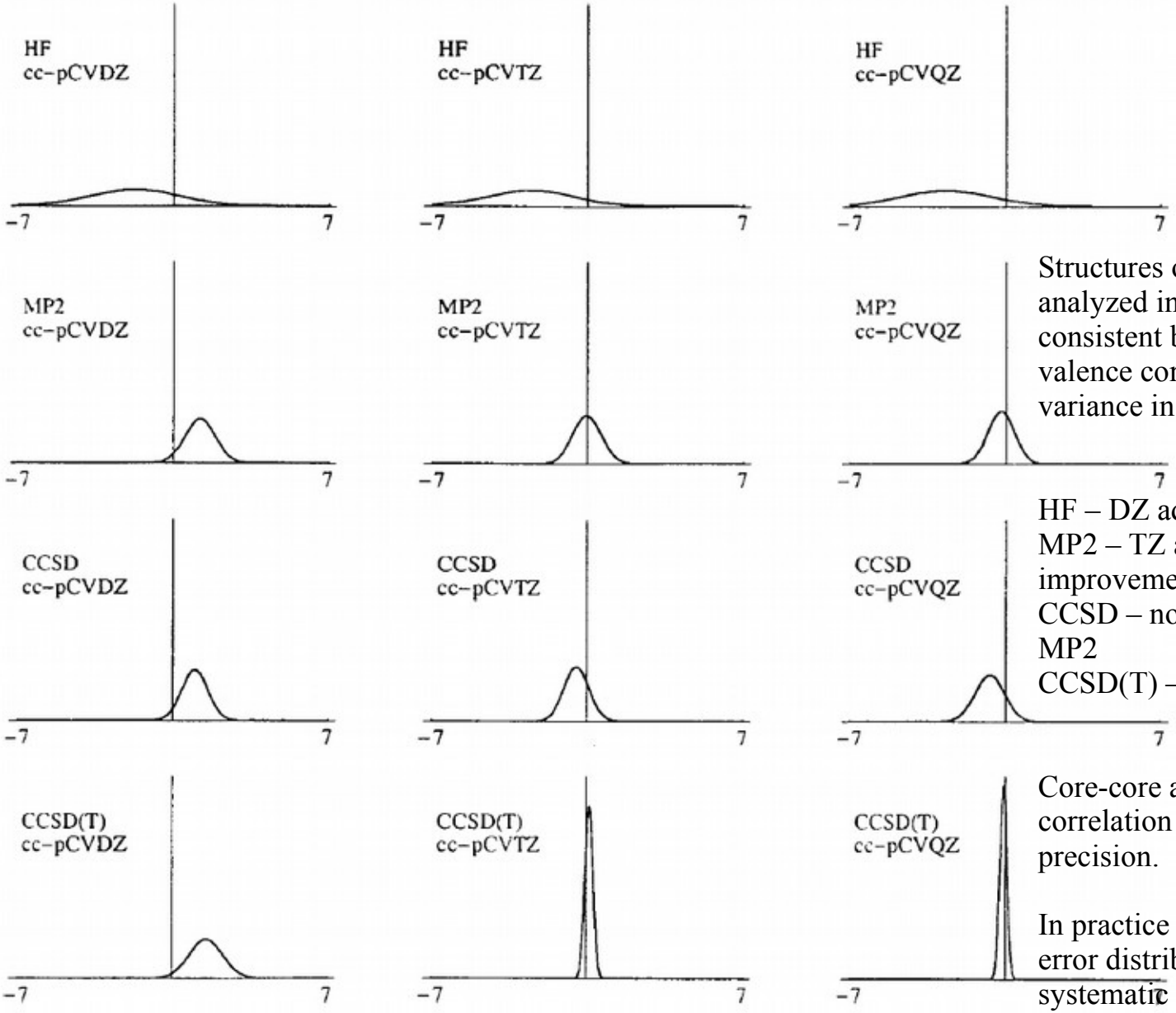
- How does H distribute and recombine within a cavity and how sensitive is this to the potential?

Sources of uncertainty

- Scales: time (fs to years), space (fm to m)
- Multiphysics/models: electronic, atomic, quantum, classical, continuum, ZPE, many-body, ...
- Material: history, chemical composition, ...
- ...
- How does the form/detail of the pair potential affect our conclusions?

W-H potential

- PK using Tersoff form due to
  - Juslin et al., J.App.Phys. 98 (2005) 123520
  - Tour de force paper synthesizing multiple scales of simulation with experiment
- Explore comparison with various ab initio forms and model systems
  - For smallish finite systems we know we can get the right answer with ZPE, excited states, etc.
  - The right answer for the right reasons



Structures of 19 molecules analyzed in correlation consistent basis sets with core-valence correlation. Mean and variance in normal form

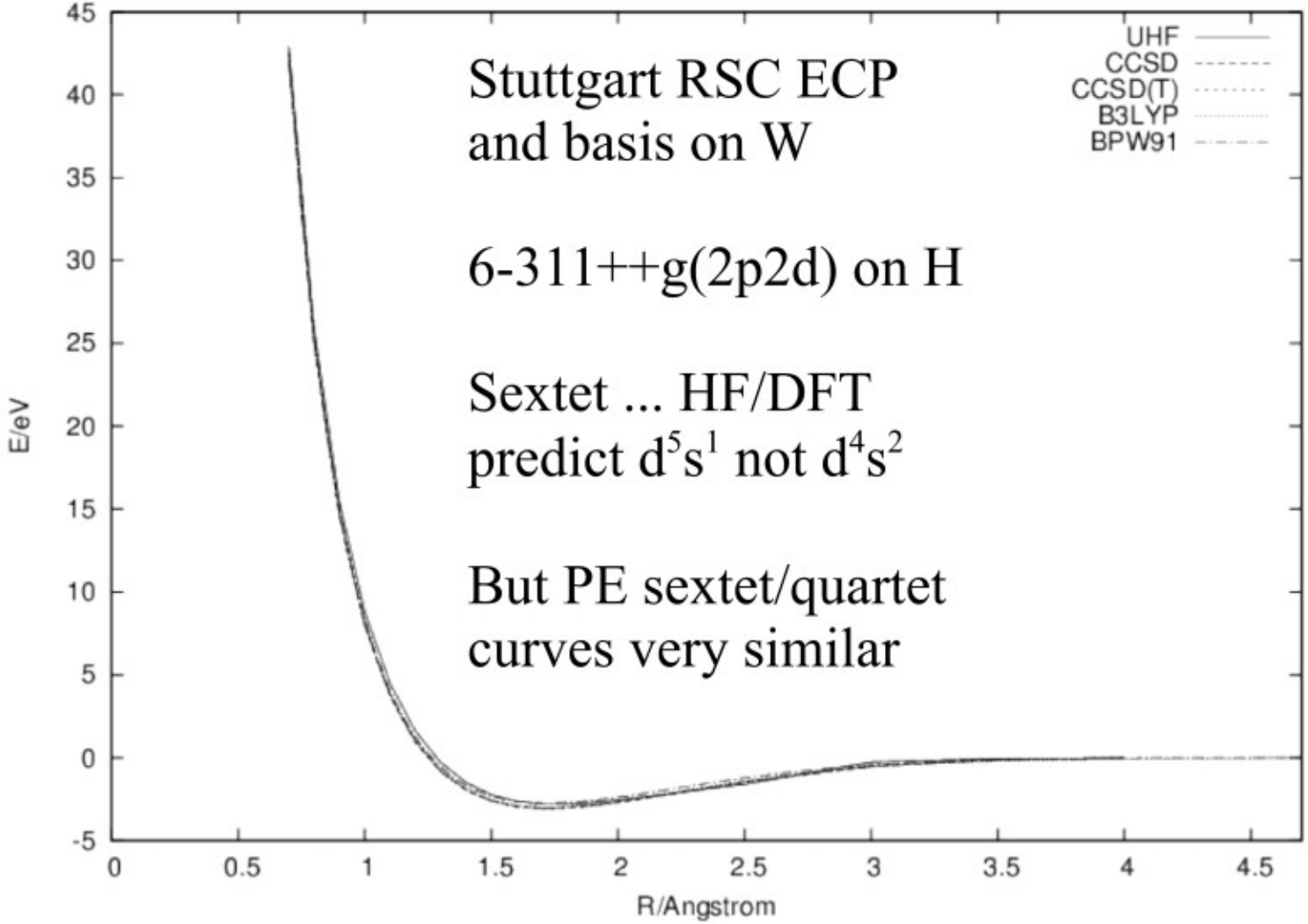
HF – DZ adequate
MP2 – TZ adequate, big improvement over HF
CCSD – no improvement over MP2
CCSD(T) – DZ inadequate

Core-core and core-valence correlation important for high-precision.

In practice this oversimplifies error distribution and ignores systematic errors: need Gn etc.

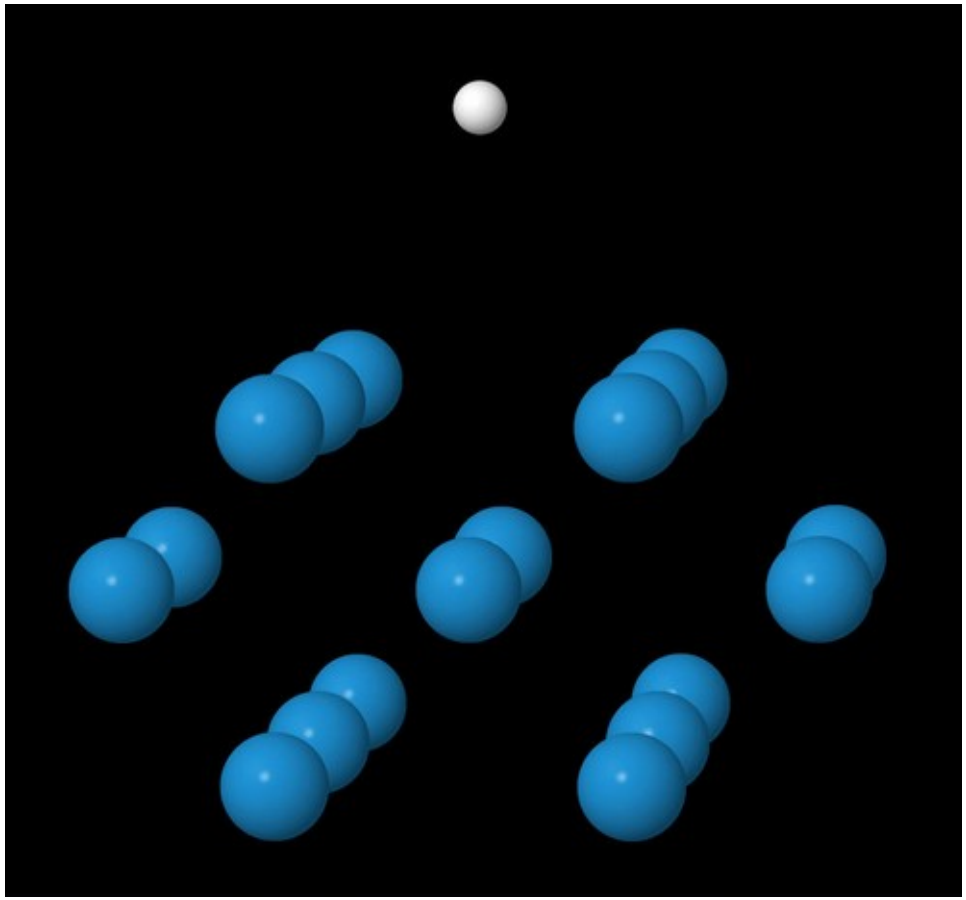
FIG. 2. Normal distributions of errors in the calculated bond distances (pm). For ease of comparison, all distributions have been normalized to one and plotted against the same horizontal and vertical scales.

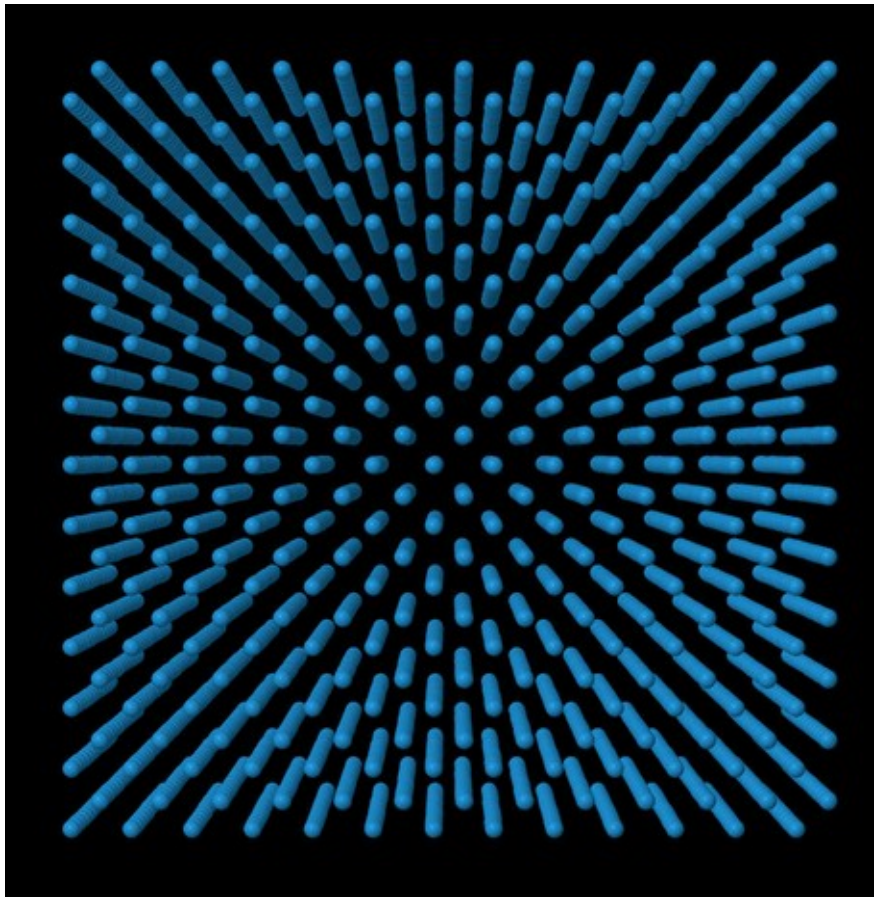
W-H ab initio potentials



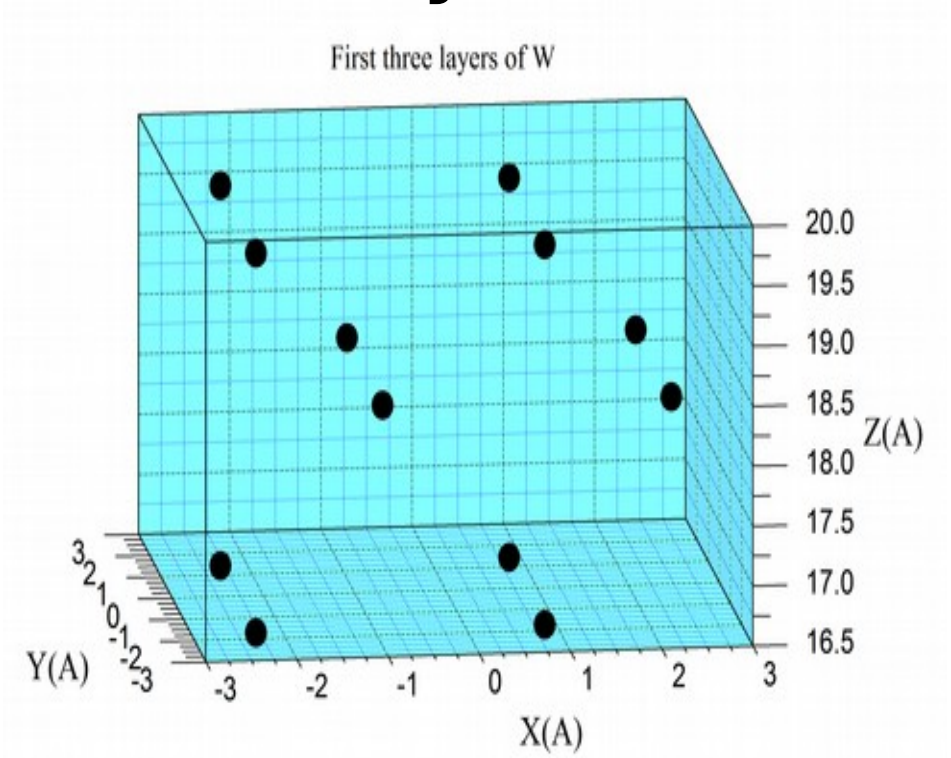
Cluster model

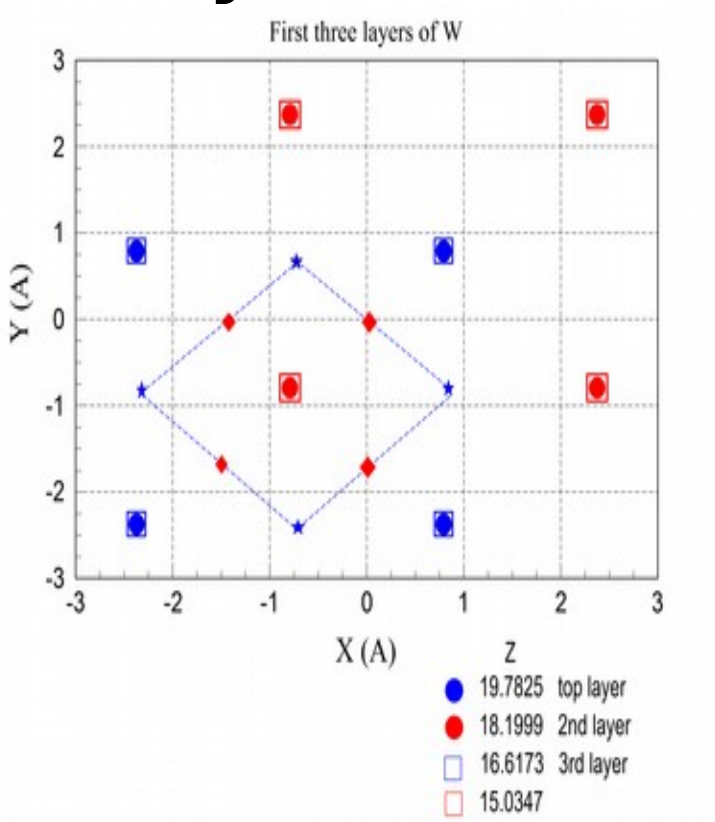
- 18 W atom cluster cut from KP's periodic





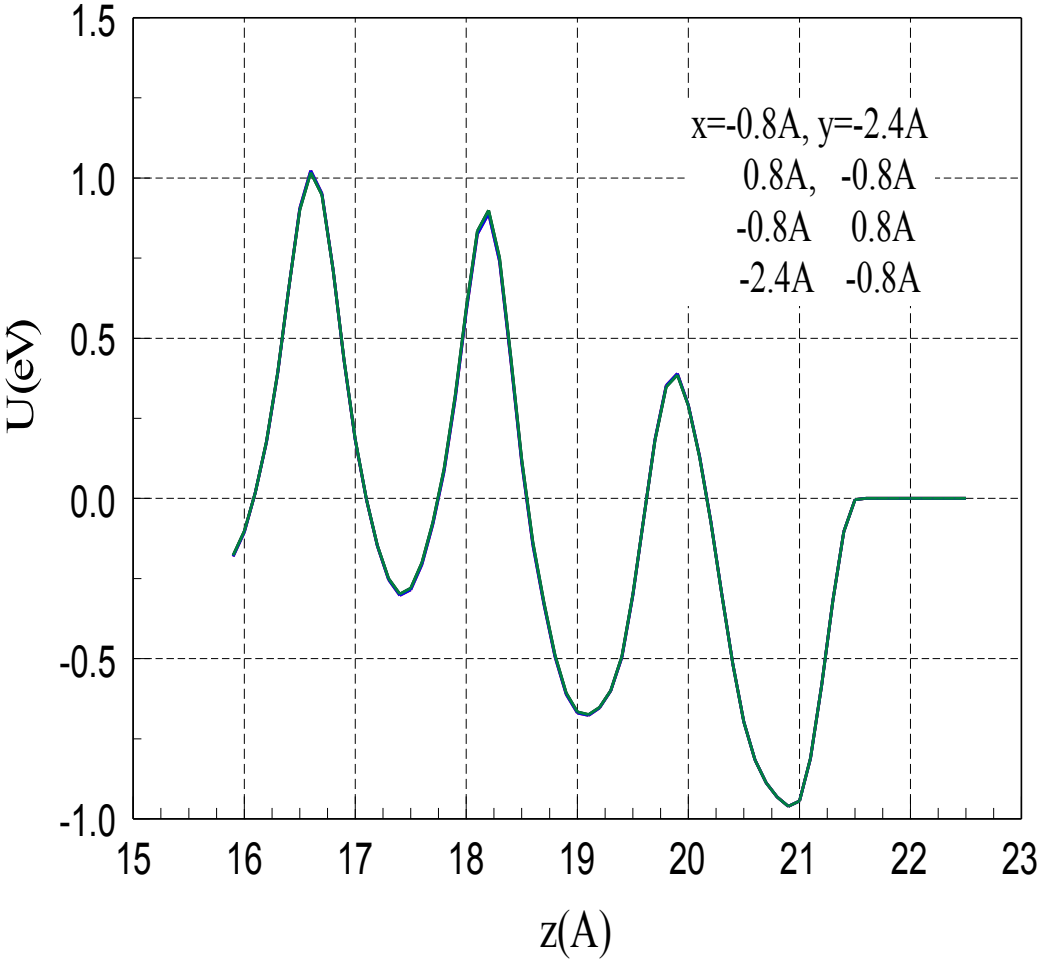
Potential: Nondamaged TUNGSTEN monoCrystal, first three layers shown





The surface interface (top layer) is at Z=19.7825 Å. Simulation box is 4x4x4 nm (~5000 atoms. Shown is just a central piece of the surface, form the top (center of the box is app at (0,0,0).
X-Y is periodic even in the potential scan that follows.
At the right figure are the layers form the left figure presented in plane X-Y (view form the top), layers are color coded for various Z.
Meaning of the blue stars and red rhomboids will be clear in the next slides.

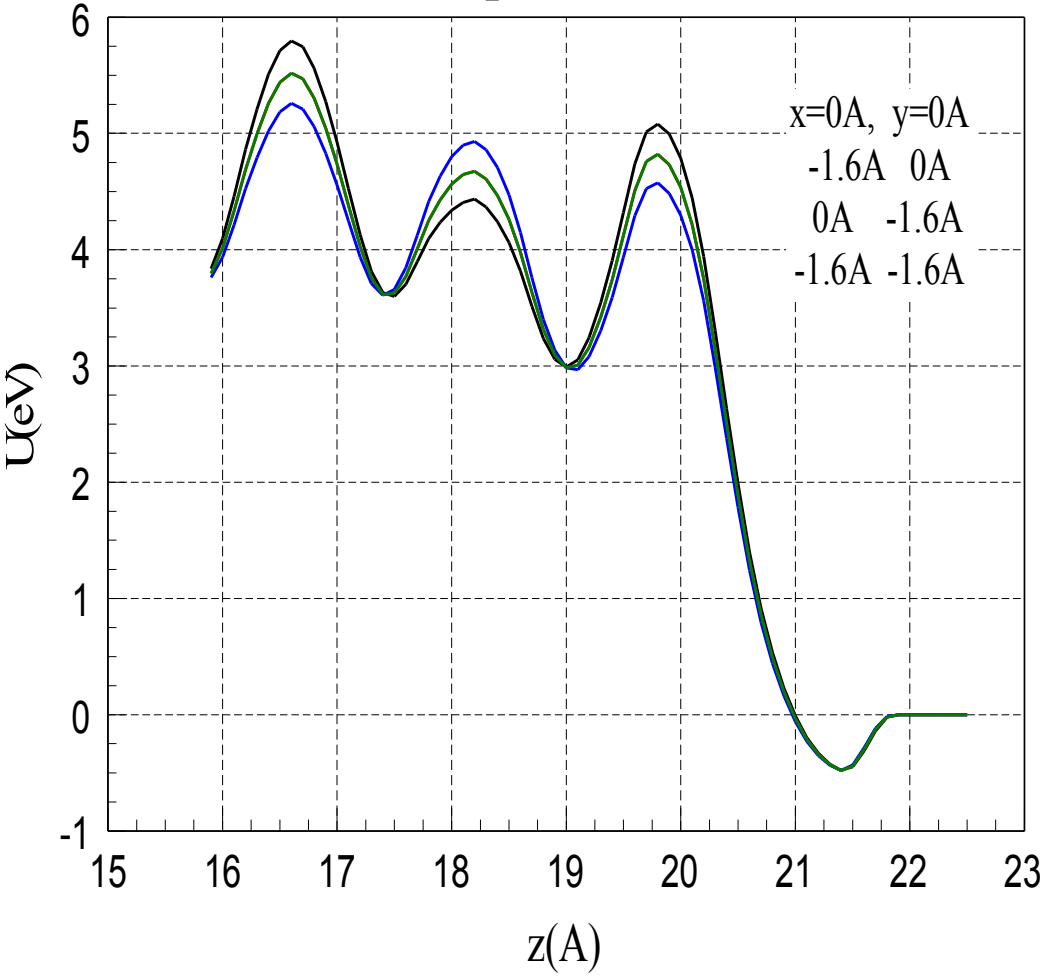
Potential as function of Z for a fixed (X,Y) (line of “impact”) I



Potential at four (X,Y) points (blue stars in slide 1) is identical.

1.Notice chemisorption well of depth of 1 eV in front of the first layer (the first layer is at Z~19.8Å)
2.About 2 A above the surface, no more Influence of the potential (short-range)
3. First barrier is at about first layer Z (height here <0.5 eV), than there is a second bonding sight between first and second layer
4. Next barrier is at Z of the 2nd layer of The height close to 1 eV
5. Next potential well, bonding (~-0.25 eV) is between 2nd and 3rd layer.
6. Next barrier is at about the third layer and is of 1 eV height….

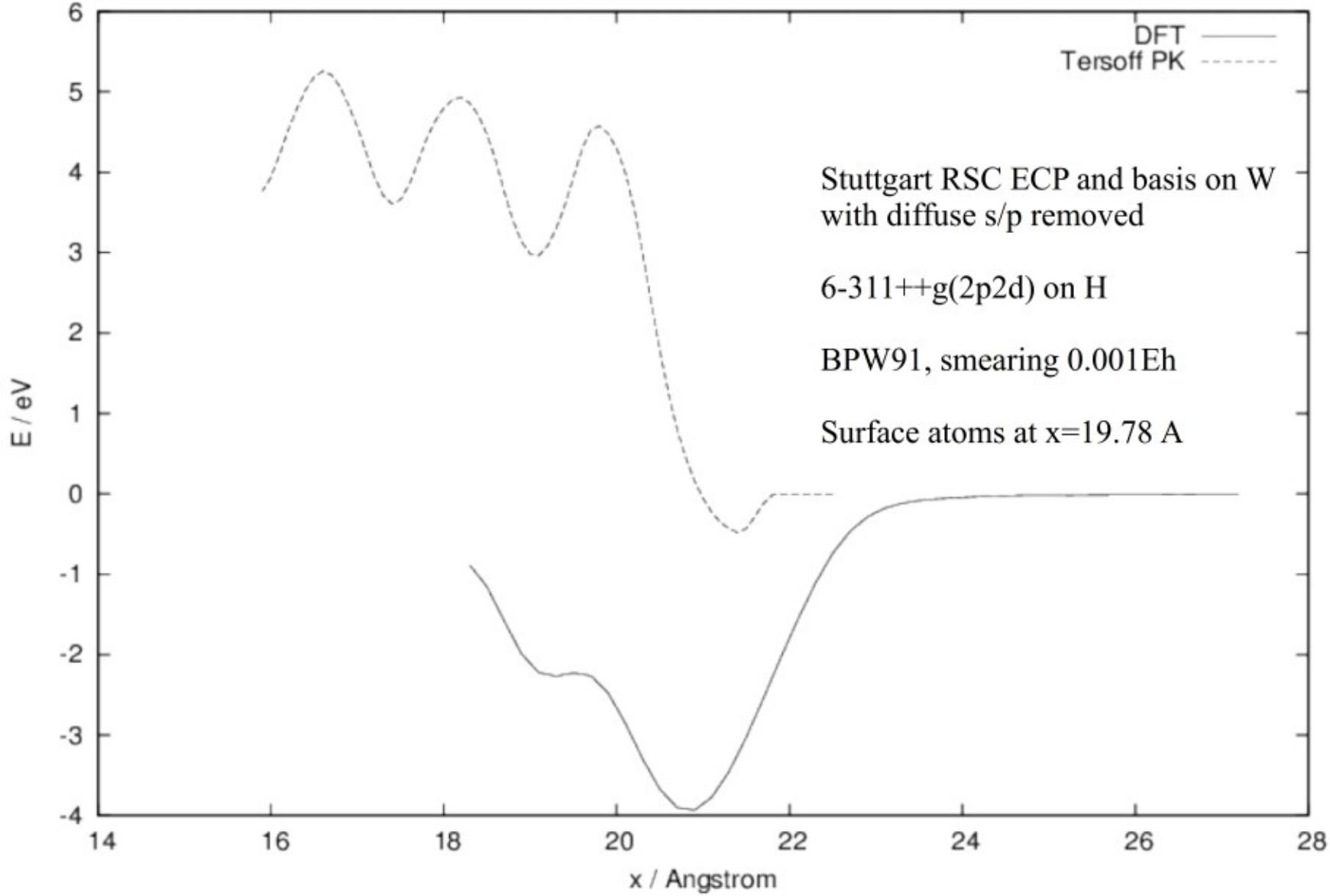
Potential as function of Z for a fixed (X,Y) (line of “impact”) II



Potential at four (X,Y) points (red rhomboids in slide 1) is almost Identical (deviation comes because positions of having the potential calculated are a bit shifted form the rhomboid points.

1.Notice chemisorption well of depth of ~0.5 eV in front of the first layer (the first layer is at Z~19.8Å)
2.About 2+ A above the surface, no more Influence of the potential (short-range)
3. First barrier is at about first layer Z (height here about 5 eV),
4. There is a second potential well (depth 3 eV) between first and second layer
5. Next barrier is at Z of the 2nd layer of the height close to 5 eV
6. Next potential well, bottom pot. Is 3.5 eV is between 2nd and 3rd layer.
7. Next barrier is at about the third layer and is of 5.5 eV height....

DFT BPW91 vs. Tersoff



Next steps

- Convergence w.r.t basis and many-body
  - Explicitly correlated wave functions
  - Excited states, spin-coupling, $H/H_2/WH/WH_2/...$
- Transfer accurate many-body data to solid state
  - Explore error in/via embedding methods
- Quantify remaining uncertainty
  - Geometry, defects, finite temp, local electronic variation, confinement effects, ...