BOSS Demo 3
3D Molecular Adsorption Energy Landscapes

1 Problem

Molecule-surface interactions critically (but not exclusively) depend on the molecular position above the surface, which can be described by an (x,y,z) position vector of the molecule. A common task in BOSS is to minimise adsorption energy with respect to the position vector. This demo will introduce such a mixed-boundary 3D simulation. We will use pre-computed DFT data to perform a single surrogate model fit to the data and compute 3D adsorption energy landscapes.

Given periodic boundary conditions of the simulation cell, x and y are periodic variables. Their bounds are defined by supercell lattice parameters. It is most convenient to choose the dimensions of the smallest repeating surface unit and employ a lateral offset (this is not essential). The non-periodic z variable is typically chosen to sample a region of vacuum immediately above the surface slab. It can be defined as the centre of the molecule, the height of the molecular axis, or the height of the atom closest to the surface. When defining the z variable, it is important to consider molecule dimensions and tilt in simulations to avoid probing variable domains where the molecule overlaps with the surface. Interactions in z direction are dominated by dispersion, in which case adsorption strength is usually scaled without altering the landscape variation, but it is also possible to detect the onset of chemical binding as a function of molecular registry. z domain is chosen to span several Å from the closest surface approach (approximately the length of a chemical bond 1.5Å).

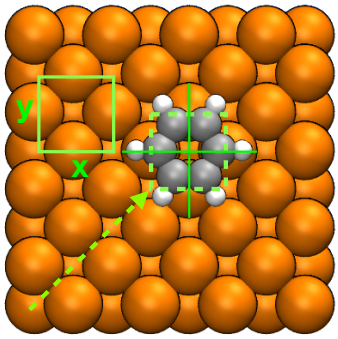

Figure 1: Benzene/Cu(100).

2 Run instructions

This demo involves a restart with a (confidential) DFT dataset. The adsorption of benzene on Cu(100) was simulated with FHI-aims, as illustrated in Fig. 1. The input file features a list of datapoints in the form (x,y,z,E_{ads}). You can compare your BOSS results with the enclosed output files in the `solutions` directory. The xyz file illustrates the structures corresponding to data acquisitions.

Run tasks

1. Read the input file and consider variable boundaries. The kernel type for each dimension is defined as a list of string parameters in the `kernel` line. Expected variation of adsorption energy is specified as `yrange`. The number of `initpts` should match dataset size (110). We will not make further data acquisitions, so `iterpts` is 0.
2. Note the dataset under the `RESULTS` keyword. How many data points were acquired at the boundaries of the non-periodic z variable?
3. Load the data acquisition trajectory and consider the location of the acquisition points.
4. Run `boss` on the input file to fit a single 3D GP surrogate model to this data. Check `boss.out` to note the position and value of the global energy minimum. Is there an energy value in the dataset that is lower than the predicted global minimum of adsorption? Compare your global minimum against the precomputed result in the enclosed `boss.out` file.

5. Check the `postprocessing` directory for the model slice images in x-y (dimensions 1-2). Are they chemically meaningful in comparison to the Cu(100) surface spanned by the x-y domain? Which surface site corresponds to the global minimum of adsorption?
6. Change the input file to plot x-z or y-z adsorption energy. Does the molecule physisorb or chemisorb?
7. Compare your results with images in section 3.

3 Results and Analysis

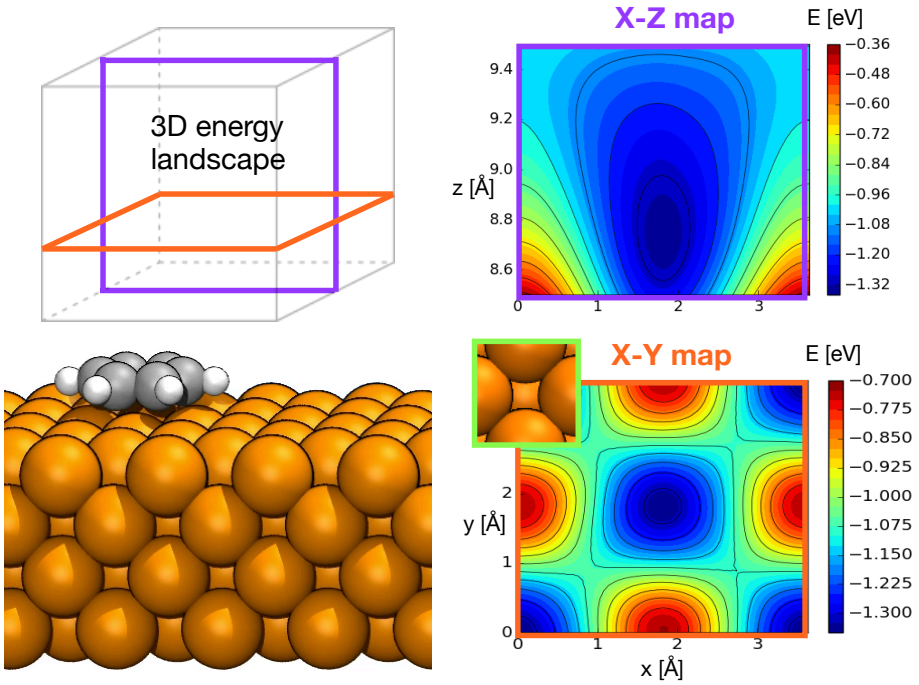

Figure 2: 3D lanscape for adsorption energy of benzene/Cu(100): model slices in x-y and x-z are taken through the $\hat{x}$ global minimum.