Ab-initio Study of Lithium Intercalation into a Graphite Nanoparticle
Supporting Information

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1 Simulation Cell

We include an image of our nanoparticle inside its large simulation cell.

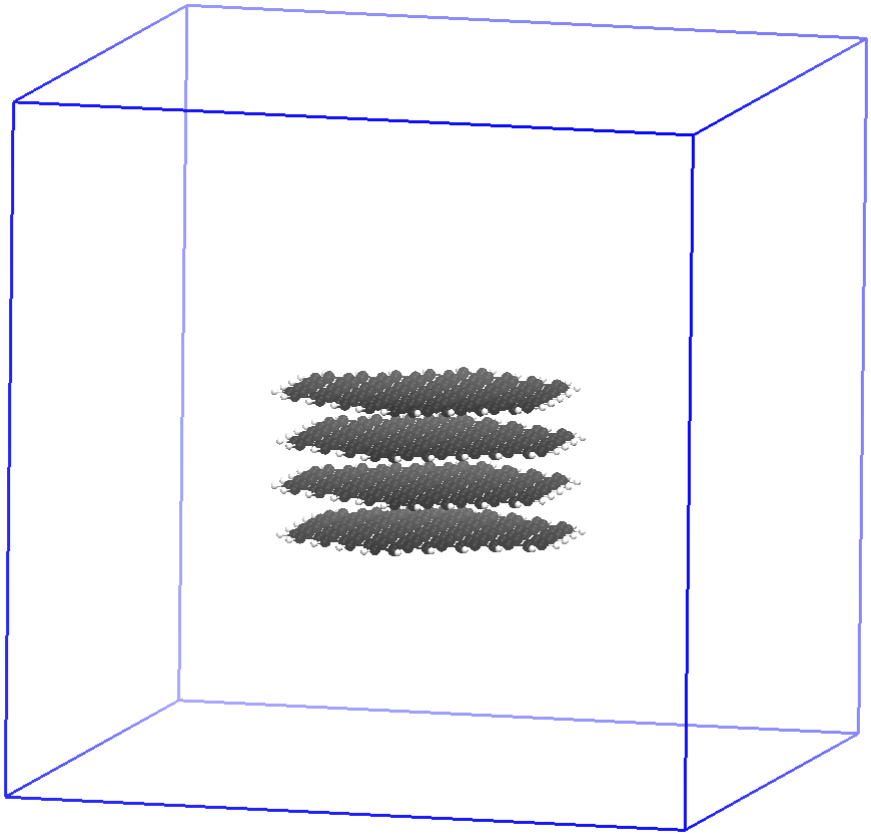
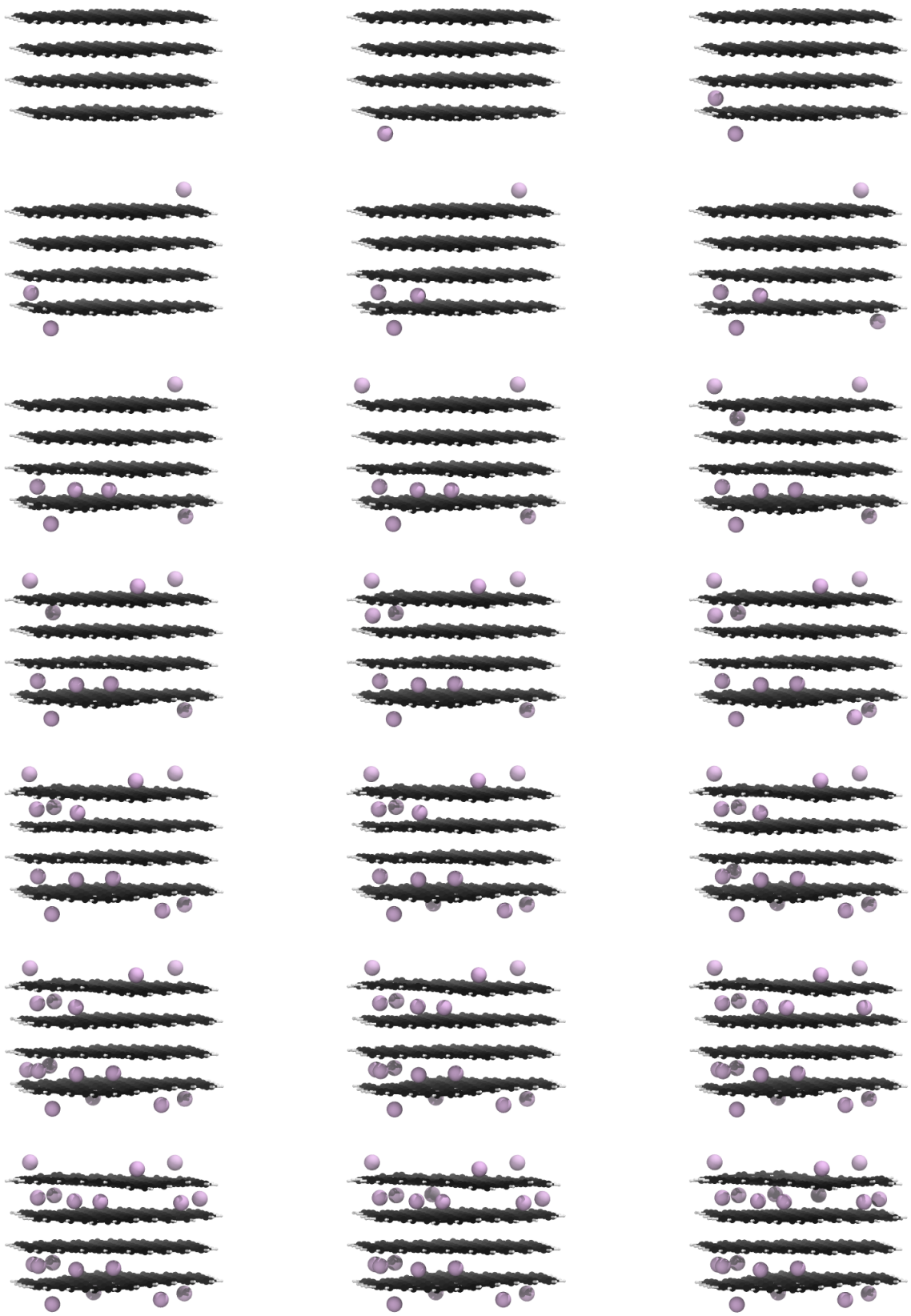
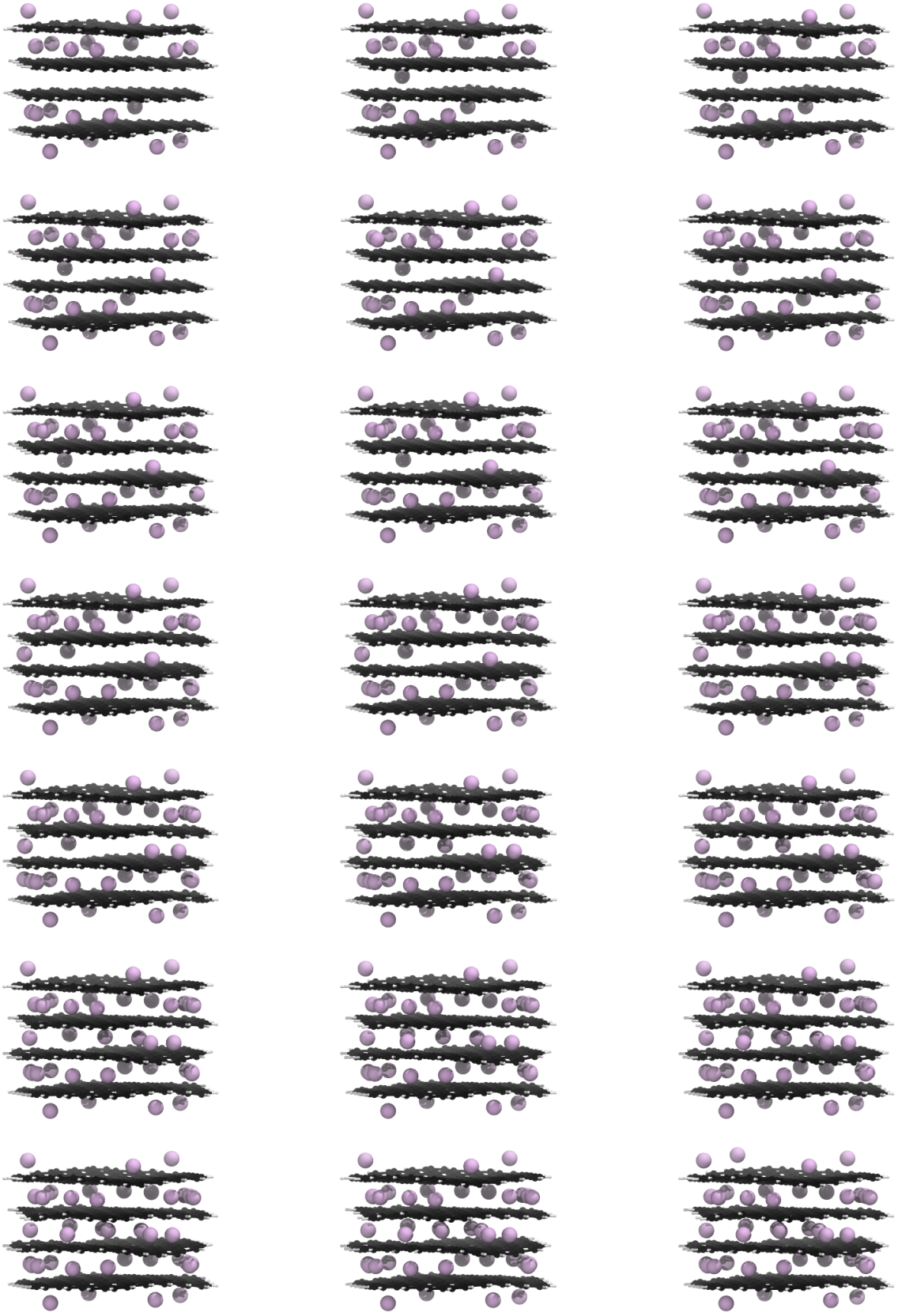


Figure S1: Our nanoparticle in a 50Å × 50Å × 50Å simulation cell

2 Structures

2.1 No Restrictions PBE-D2 structures





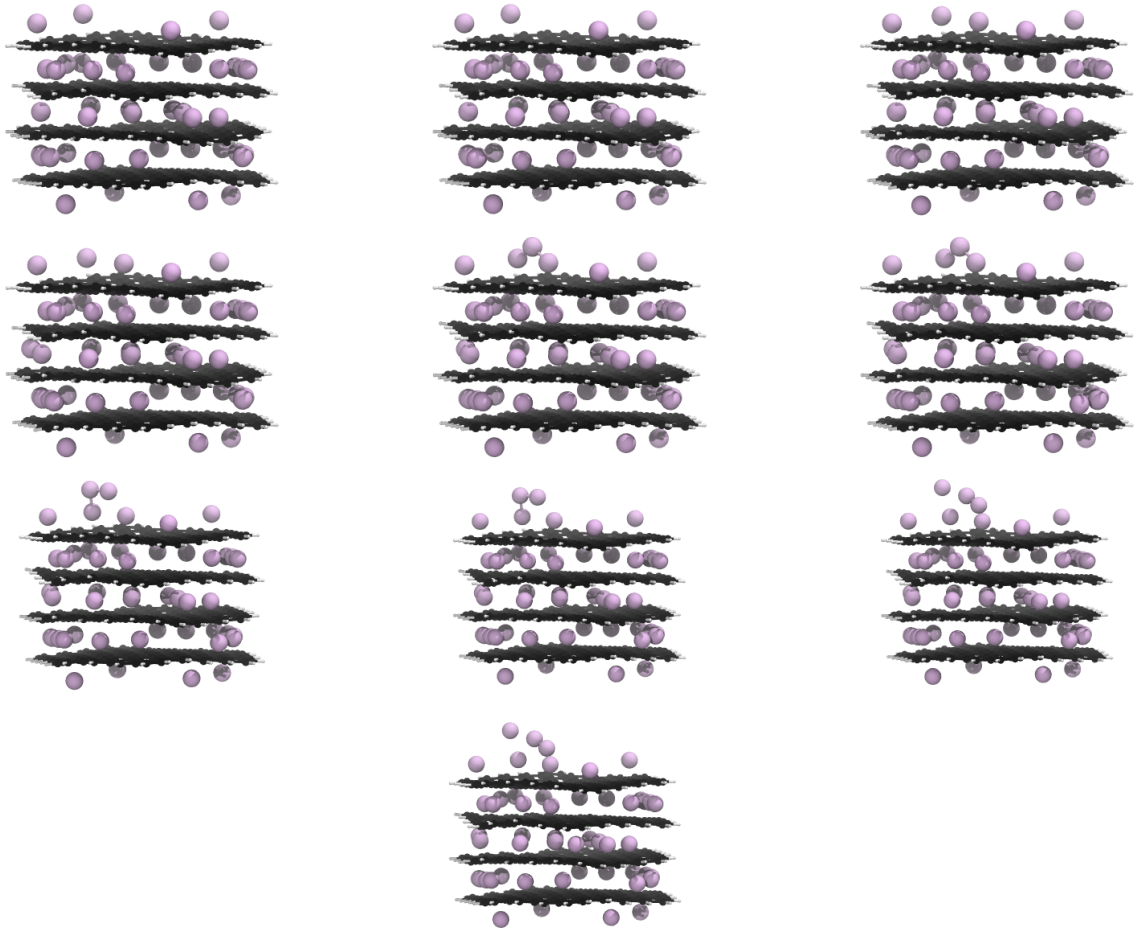
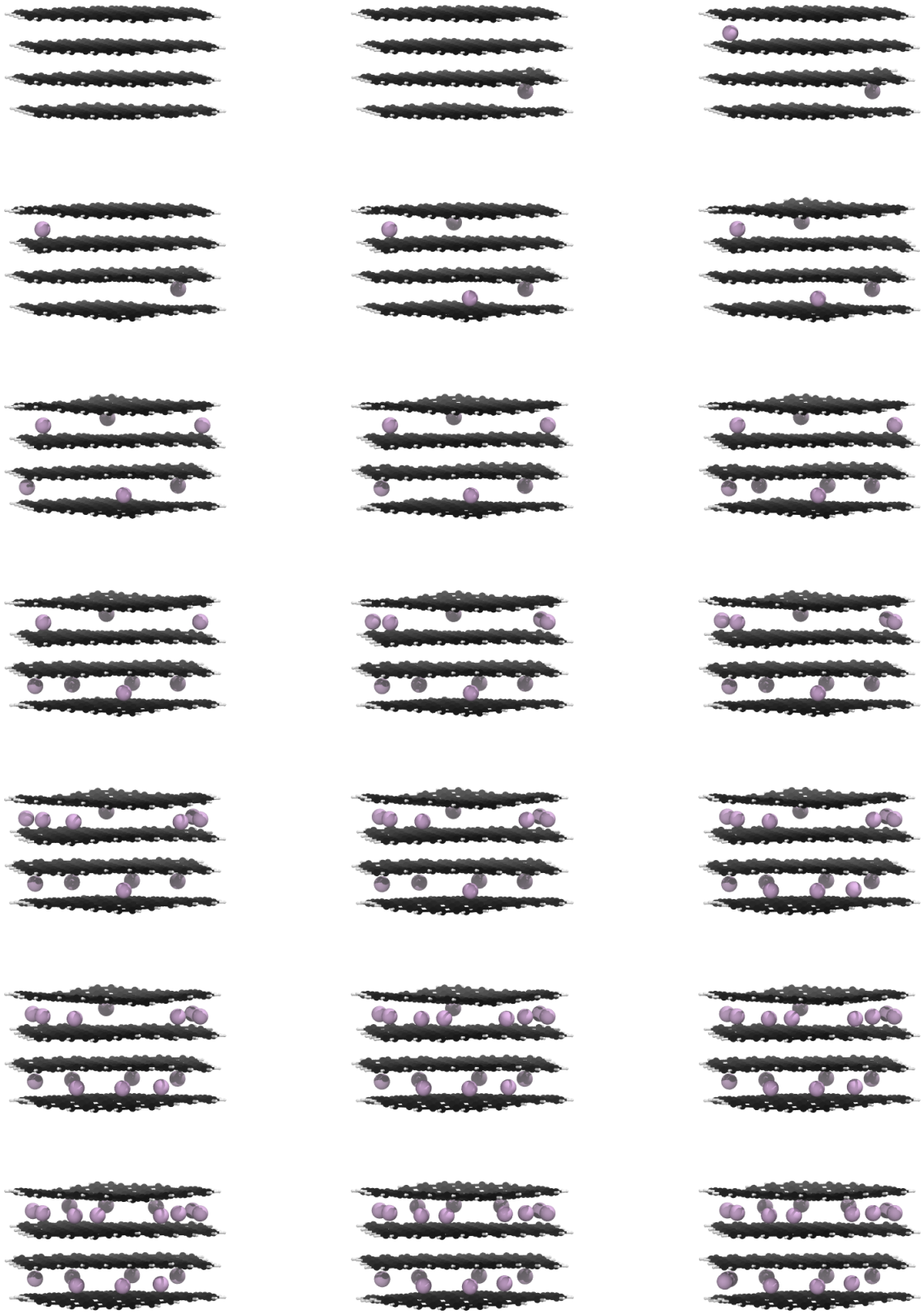
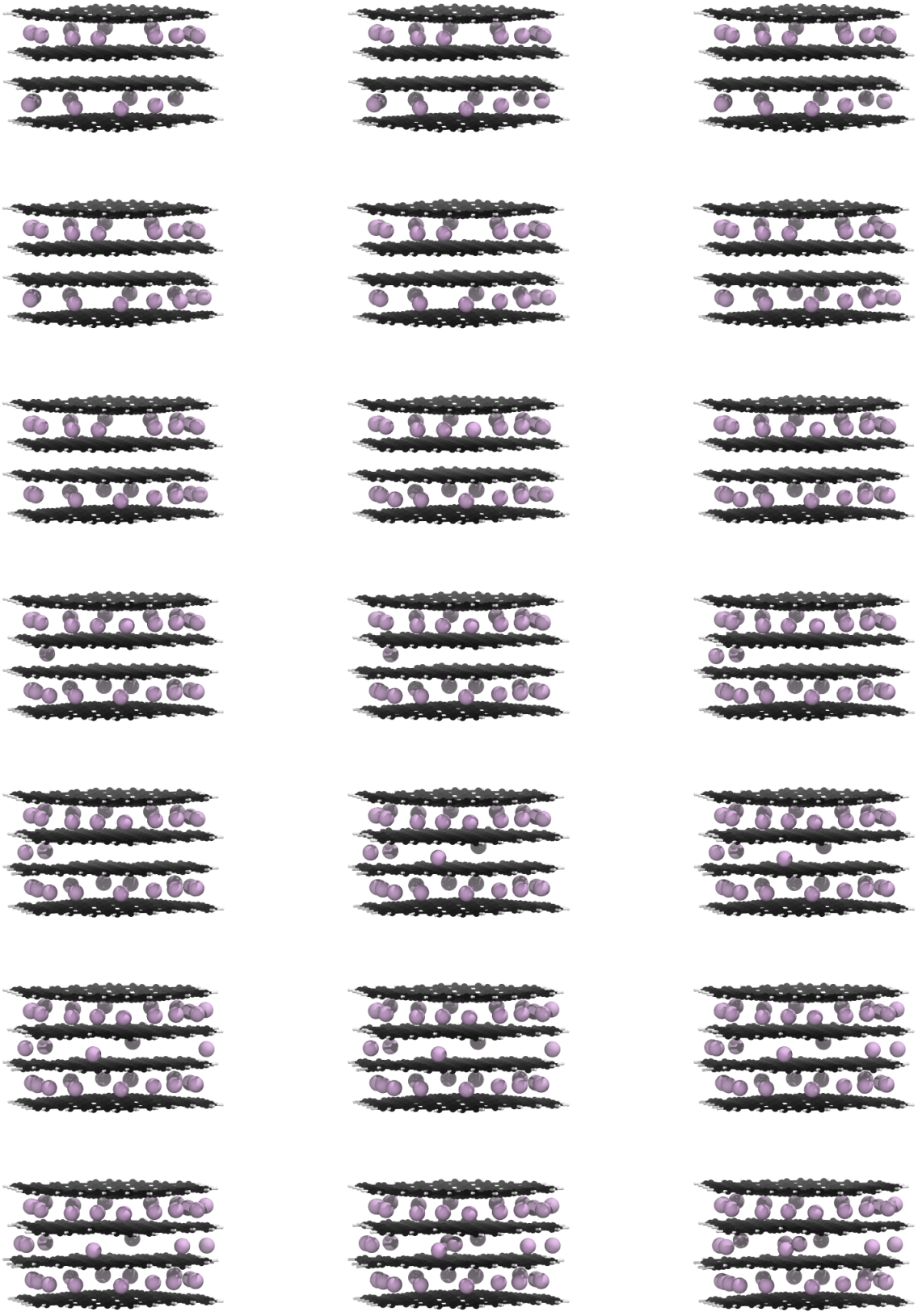


Figure S2: First 51 structures made using the PBE-D2 functional without any restriction on atom placement.

2.2 PBE-D2 structures





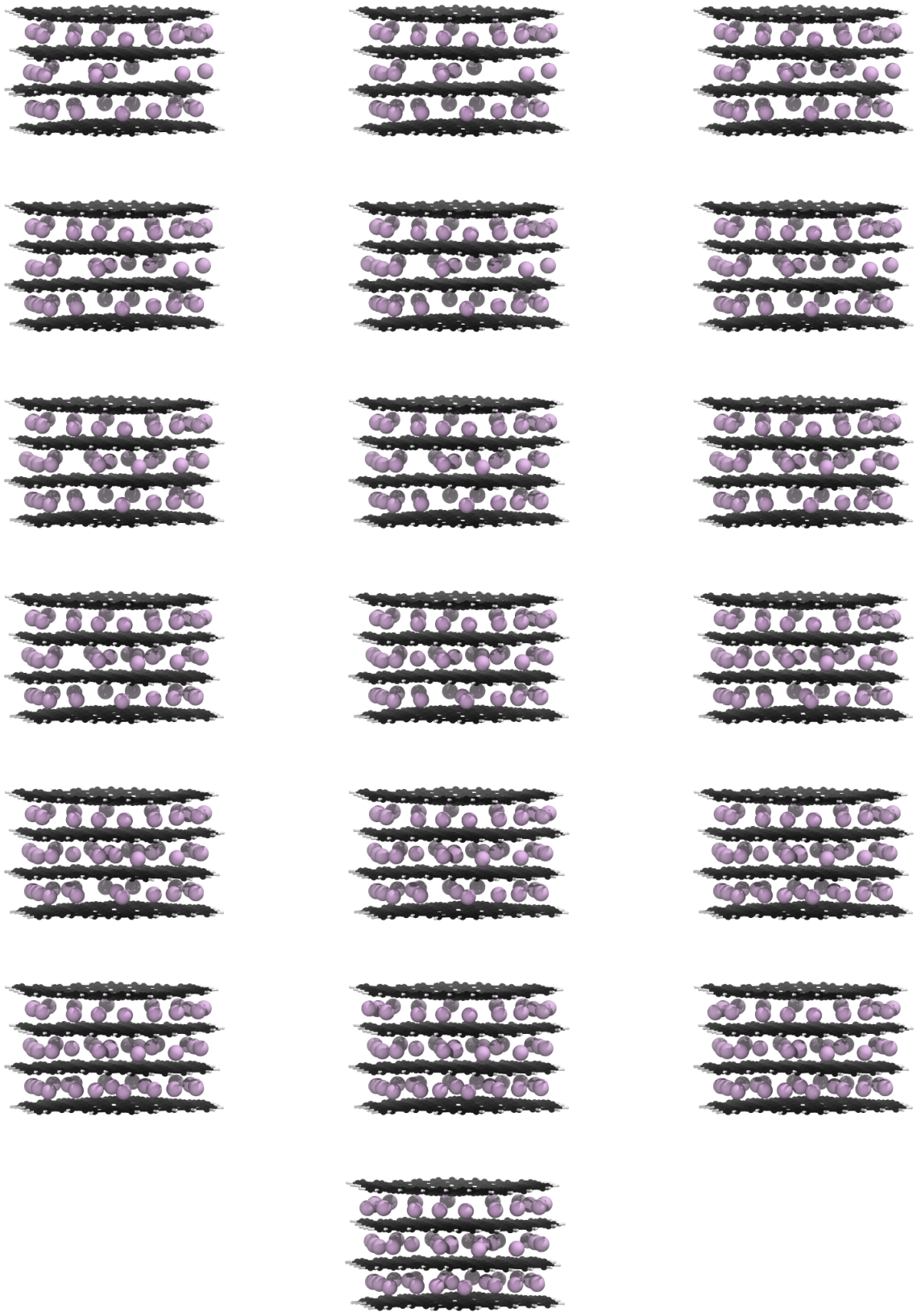
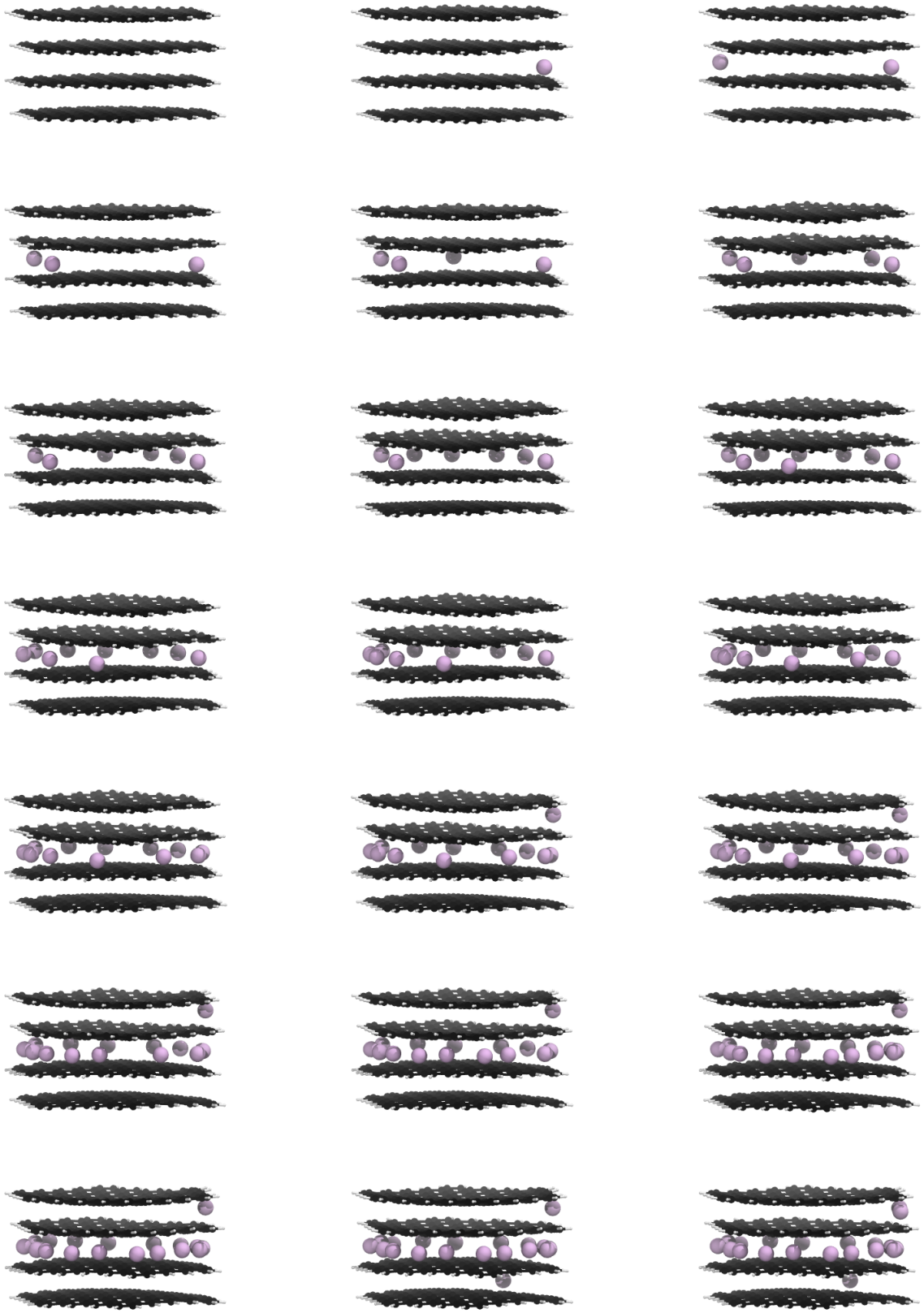
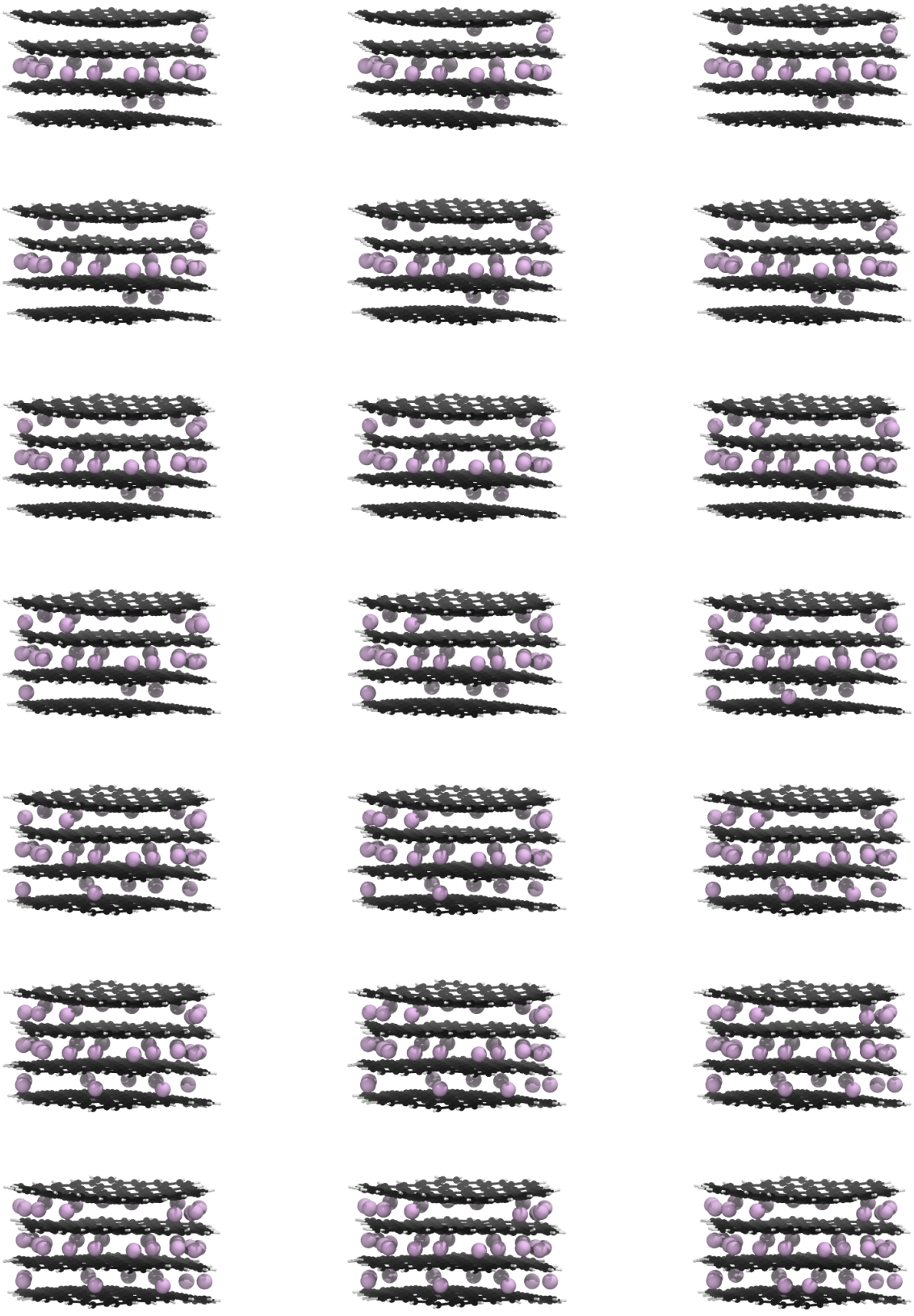


Figure S3: The 60 structures made using the PBE-D2 functional with restriction on atom placement between graphite layers.

2.3 optPBE structures





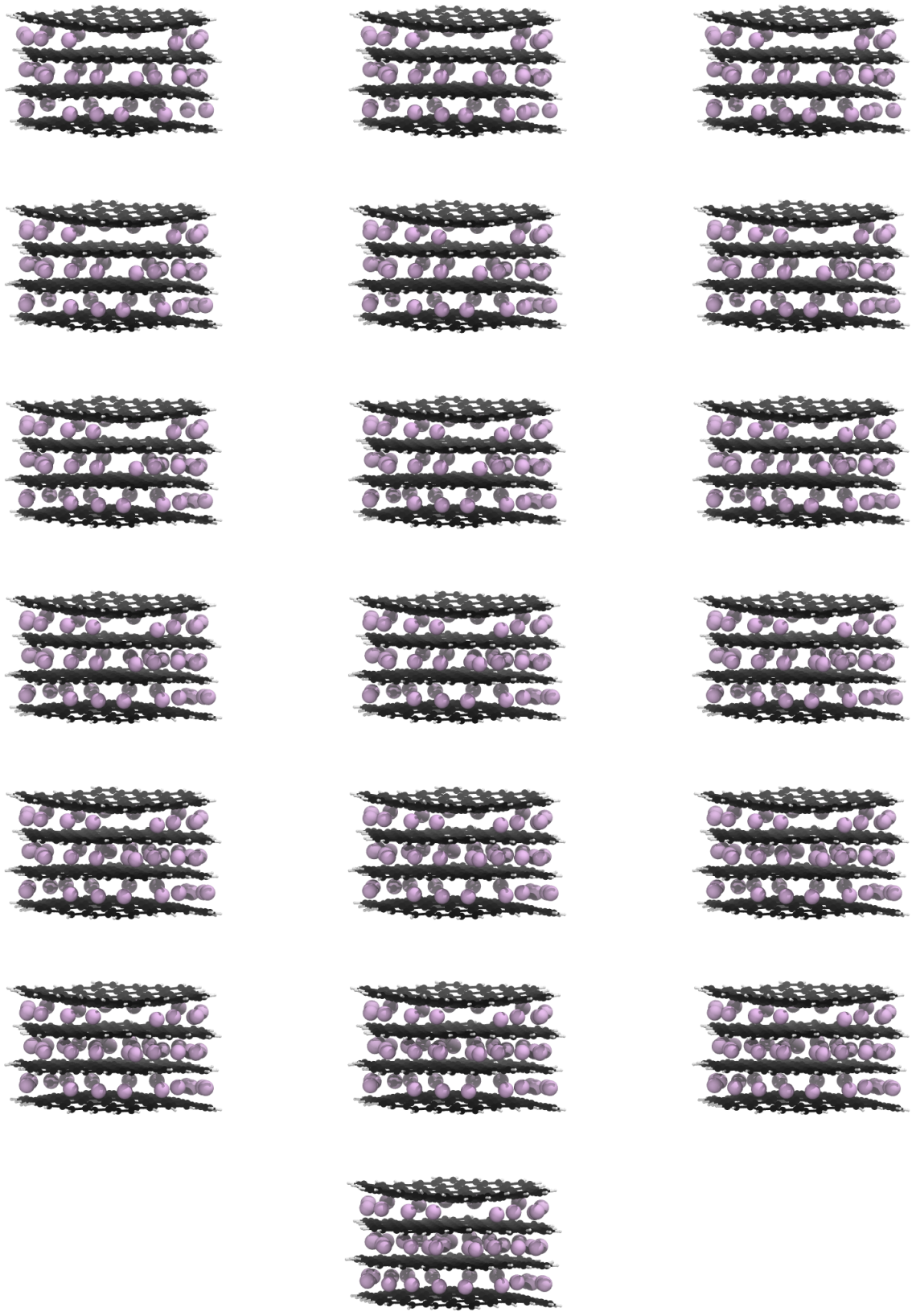
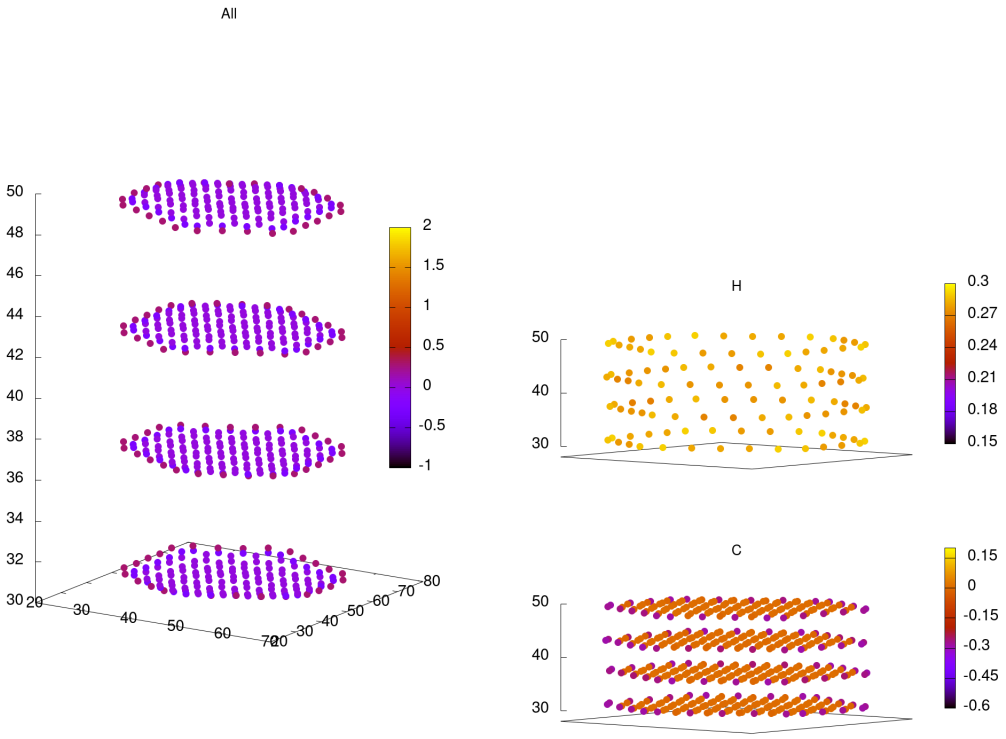
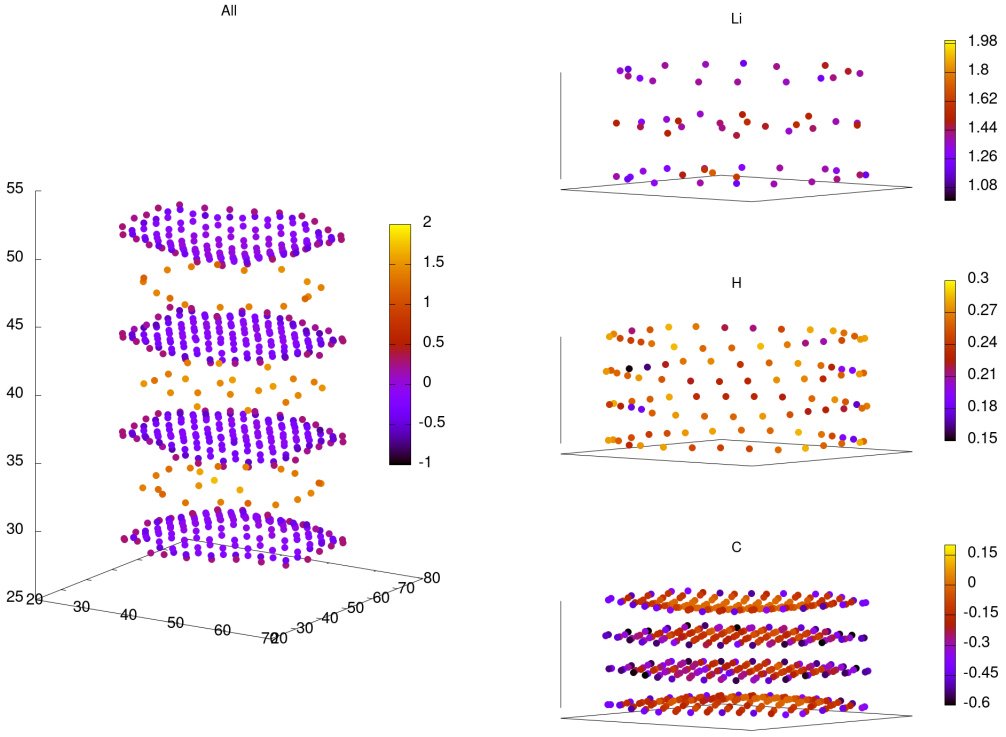


Figure S4: The 60 structures made using the optPBE functional with restriction on atom placement between graphite layers.

3 Mulliken Population Analysis

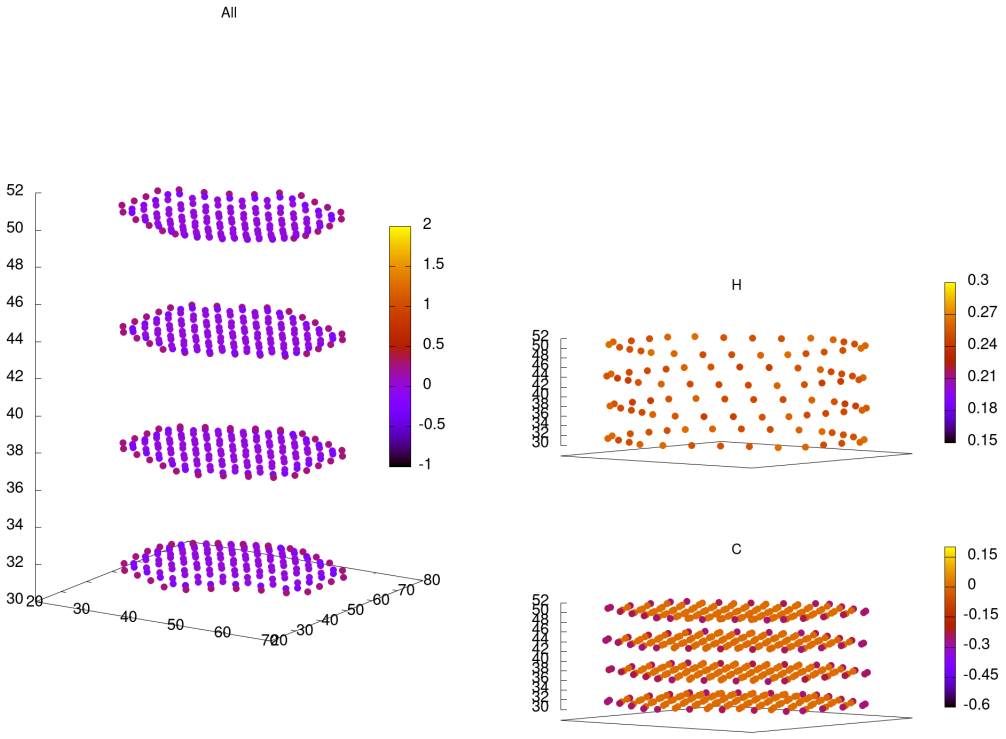


(a) 0 Li PBE-D2

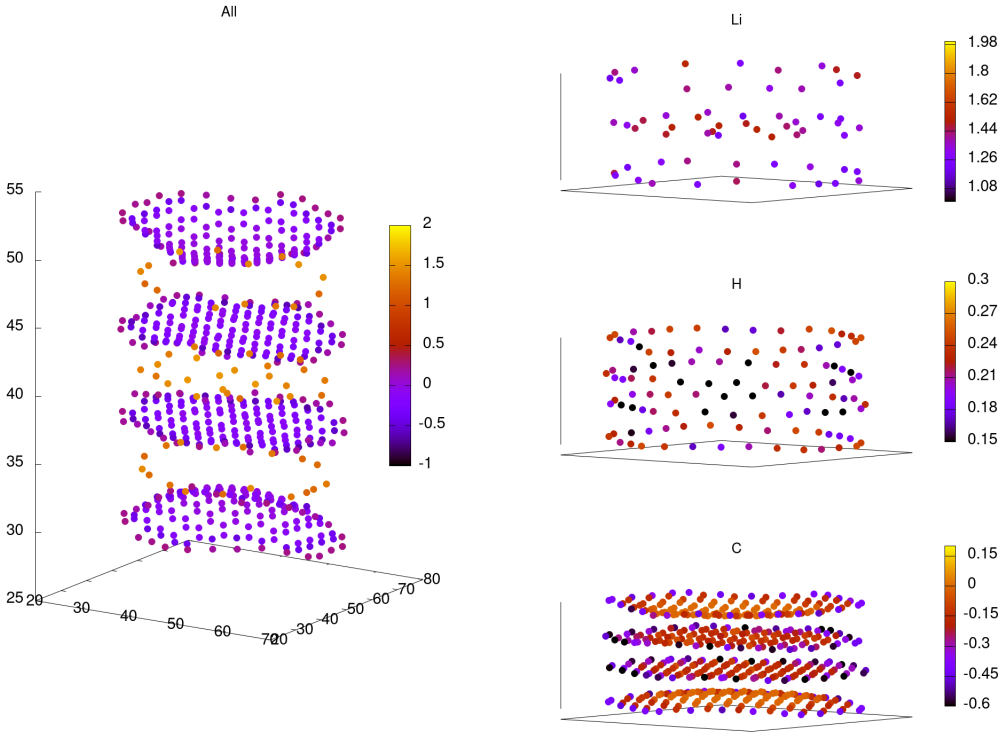


(b) 60 Li PBE-D2

Figure S5



(a) 0 Li optPBE



(b) 60 Li optPBE

Figure S6

4 Insertion Energy

We calculate the insertion energy as a slight reformulation of equation 5:

$$E_{ins} = \frac{\Delta E}{\Delta n} = \frac{E_{\mathrm{Li}_{n_2}\mathrm{G}} - E_{\mathrm{Li}_{n_1}\mathrm{G}} - (n_2 - n_1)E_{\mathrm{Li(BCC)}}}{(n_2 - n_1)} \tag{1}$$

for all on hull sturctures. This provides the values for the graph shown in Fig. S7.

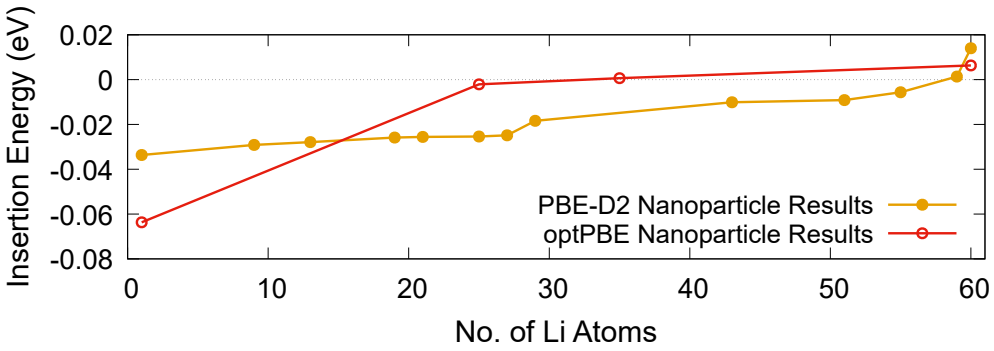


Figure S7: Insertion energy (eV) for on-hull structures with increasing lithiation for optPBE (red) and PBE+D2 (orange)

5 Data

Data related to the individual calculations performed in this paper, such as the ONETEP .out files, are provided at the following github repository. Any questions/request for more data can be asked for on the github repository by opening an issue.

`https://github.com/julianholland/Ab_initio_Study_of_Lithium_Intercalation_into_a_Graphite_Nanoparticle_SI/`