OAK RIDGE National Laboratory

07/17/2025

Session 6: Real world calculations of 2D Materials

QMCPACK Summer School, 2025

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U.S. DEPARTMENT of ENERGY

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Goals of this tutorial

- How to reach publication quality results
- Calculate the binding energy of bilayer hBN
- How to write a reusable script in Nexus
- Finite size extrapolation in 2D systems

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DMC Convergence for publication quality results

Statistical convergence

- Equilibration period
- Sampling length
- Autocorrelation

Session 3 (**S3**) has in-depth information. **Should always be controlled.** Mainly post-processing and can be **system dependent**. Nexus is very helpful.

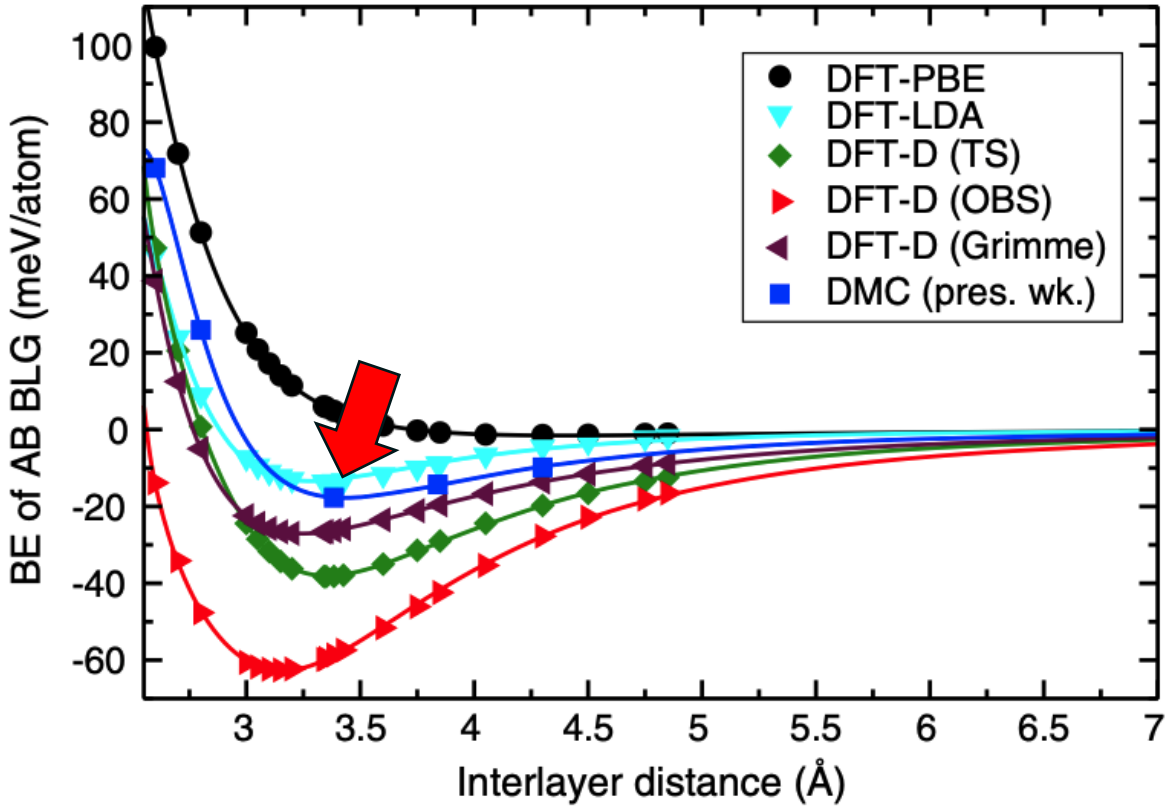
Systematic convergence

- Time step convergence (**S3**)
- Population control (walker count) (**S4**)
- Pseudopotentials (**S3**)
- Trial wavefunction quality and representation (**S3**, **S4, S5**)
- Localization approximation (**S3**)
- Finite-size effects (1 and 2 body) (**S5**)

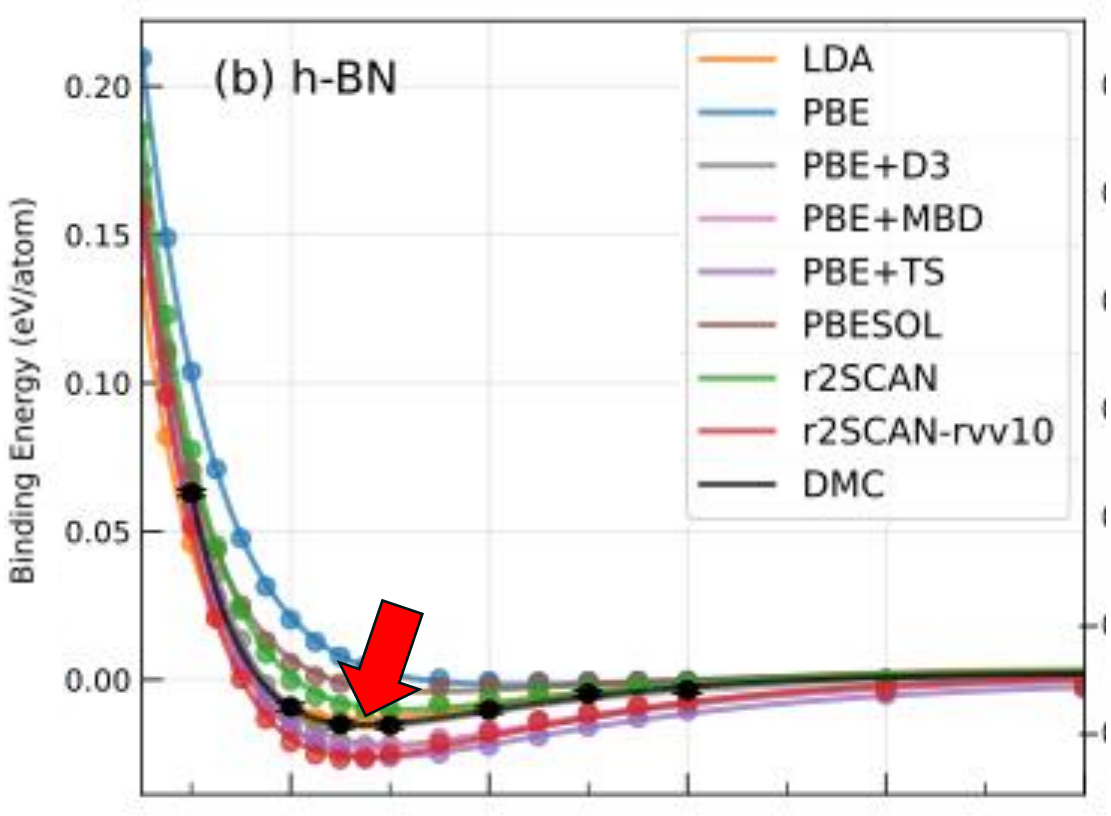
Requires an intuition and understanding of how DMC works.

For workshop files: https://github.com/QMCPACK/qmc_summer_school_2025

Accuracy of interlayer binding energy depends on the DFT functional

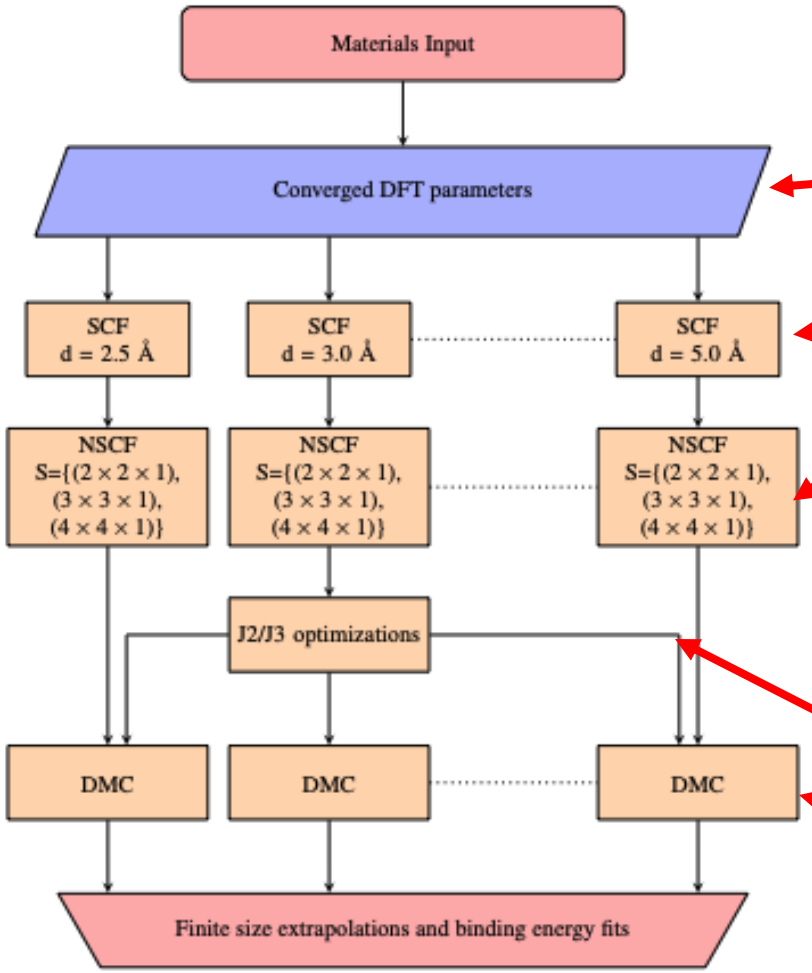


Mostaani et al. Phys. Rev. Lett. **115**, 115501 (2015)



How to write reusable scripts in Nexus

run.py



run_library.py

```python
#!/usr/bin/env python
from run_library import get_dft_settings, get_qmc_settings
from nexus import obj

dft_shared = obj(
    kgrid    = (8,8,1),  # K-point grid for DFT calculations
    ecutwfc  = 400,      # Plane-wave cutoff energy in Rydberg
    pseudos  = 'B.ccECP.upf N.ccECP.upf'.split()  # DFT PP files for Boron and Nitrogen
)
# SCF, NSCF and PW2QMCPACK settings
scf_shared, nscf_shared, conv_shared = get_dft_settings(**dft_shared)
```

```python
qmc_shared = obj(
      # Hybrid representation cutoff radius for Boron and Nitrogen in atomic units (a.u.)
      hybrid_rcut  = obj(B=1.1, N=1.1),
      # Maximum angular momentum for hybrid representation for Boron and Nitrogen
      hybrid_lmax  = obj(B=5, N=5),
      # Blip-spline Mesh factor for QMC calculations
      meshfactor   = 0.5,
      # Pseudopotential files for QMC calculations
      pseudos      = 'B.ccECP.xml  N.ccECP.xml'.split()
  )

j2_settings, j3_settings, dmc_settings  = get_qmc_settings(system = tiled_system,
                                                           **qmc_shared)
```

Contents of tutorial directory

```
/BN_tutorial
├── pseudos/
│   ├── B.ccECP.upf
│   ├── B.ccECP.xml
│   ├── N.ccECP.upf
│   └── N.ccECP.xml
├── README
├── run_library.py
├── run.py
└── structures/
    ├── hBN_d_2500.xsf
    ├── hBN_d_3000.xsf
    ├── hBN_d_3250.xsf
    ├── hBN_d_3500.xsf
    ├── hBN_d_4000.xsf
    ├── hBN_d_4500.xsf
    ├── hBN_d_5000.xsf
    └── hBN_mono.xsf
```

Supercell vectors

Corresponding twist grids

run.py

```
#!/usr/bin/env python
# user library imports
from run_library import get_dft_settings, get_qmc_settings
# nexus imports
from nexus import run_project, read_structure, obj
from nexus import generate_physical_system
from nexus import generate_pwscf
from nexus import generate_pw2qmcpack
from nexus import generate_qmcpack

# structure files and interlayer separations in Angstroms
structures = {3.0:  'structures/hBN_d_3000.xsf',
              2.5:  'structures/hBN_d_2500.xsf',
              3.25: 'structures/hBN_d_3250.xsf',
              3.5:  'structures/hBN_d_3500.xsf',
              4.0:  'structures/hBN_d_4000.xsf',
              4.5:  'structures/hBN_d_4500.xsf',
              5.0:  'structures/hBN_d_5000.xsf',
              'mono' : 'structures/hBN_mono.xsf'}
interlayer_separations  = list(structures.keys())

# Supercell tiling vectors and respective kgrids
tiling_vectors          = [(2,2,1), (3,3,1), (4,4,1)]
tiling_kgrids           = {(2,2,1):(4,4,1),
                           (3,3,1):(2,2,1),
                           (4,4,1):(2,2,1)}

# DFT and QMC settings shared across all calculations
system_shared = obj(
    B        = 3,        # Boron PP valency
    N        = 5,        # Nitrogen PP valency
    net_spin = 0         # Net spin of the system
)
```

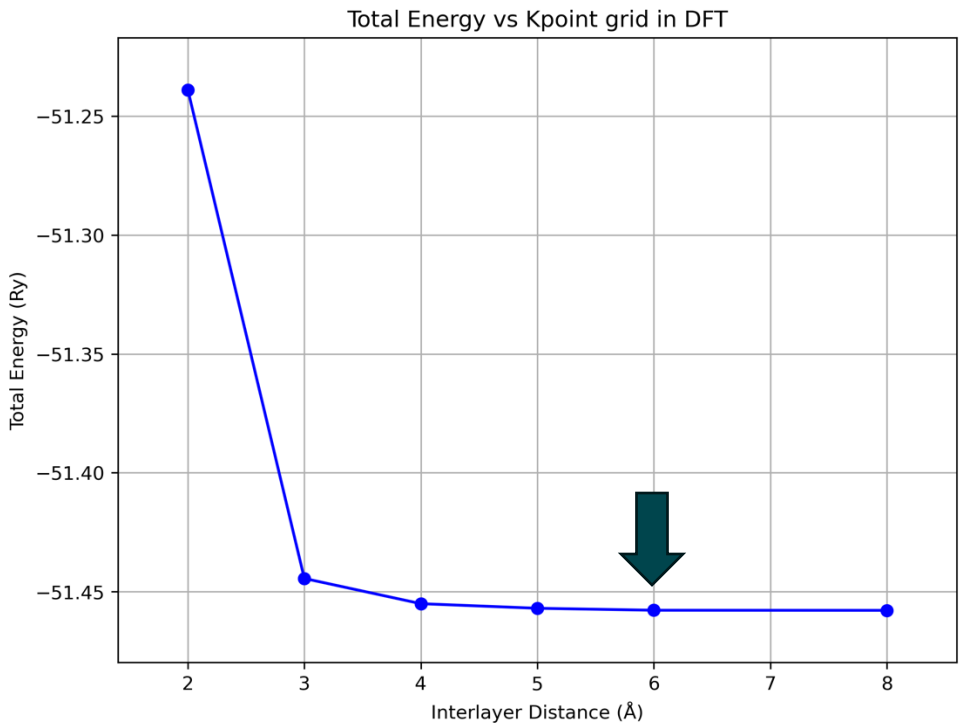
DFT convergence guides twist grid

run.py

```
#!/usr/bin/env python
# user library imports
from run_library import get_dft_settings, get_qmc_settings
# nexus imports
from nexus import run_project, read_structure, obj
from nexus import generate_physical_system
from nexus import generate_pwscf
from nexus import generate_pw2qmcpack
from nexus import generate_qmcpack

# structure files and interlayer separations in Angstroms
structures = {3.0:  'structures/hBN_d_3000.xsf',
              2.5:  'structures/hBN_d_2500.xsf',
              3.25: 'structures/hBN_d_3250.xsf',
              3.5:  'structures/hBN_d_3500.xsf',
              4.0:  'structures/hBN_d_4000.xsf',
              4.5:  'structures/hBN_d_4500.xsf',
              5.0:  'structures/hBN_d_5000.xsf',
              'mono' : 'structures/hBN_mono.xsf'}
interlayer_separations  = list(structures.keys())

# Supercell tiling vectors and respective kgrids
tiling_vectors          = [(2,2,1), (3,3,1), (4,4,1)]
tiling_kgrids           = {(2,2,1):(4,4,1),
                           (3,3,1):(2,2,1),
                           (4,4,1):(2,2,1)}

# DFT and QMC settings shared across all calculations
system_shared = obj(
    B        = 3,        # Boron PP valency
    N        = 5,        # Nitrogen PP valency
    net_spin = 0         # Net spin of the system
)
```

Supercell vectors

Corresponding twist grids



Settings common to all DFT and QMC calculations

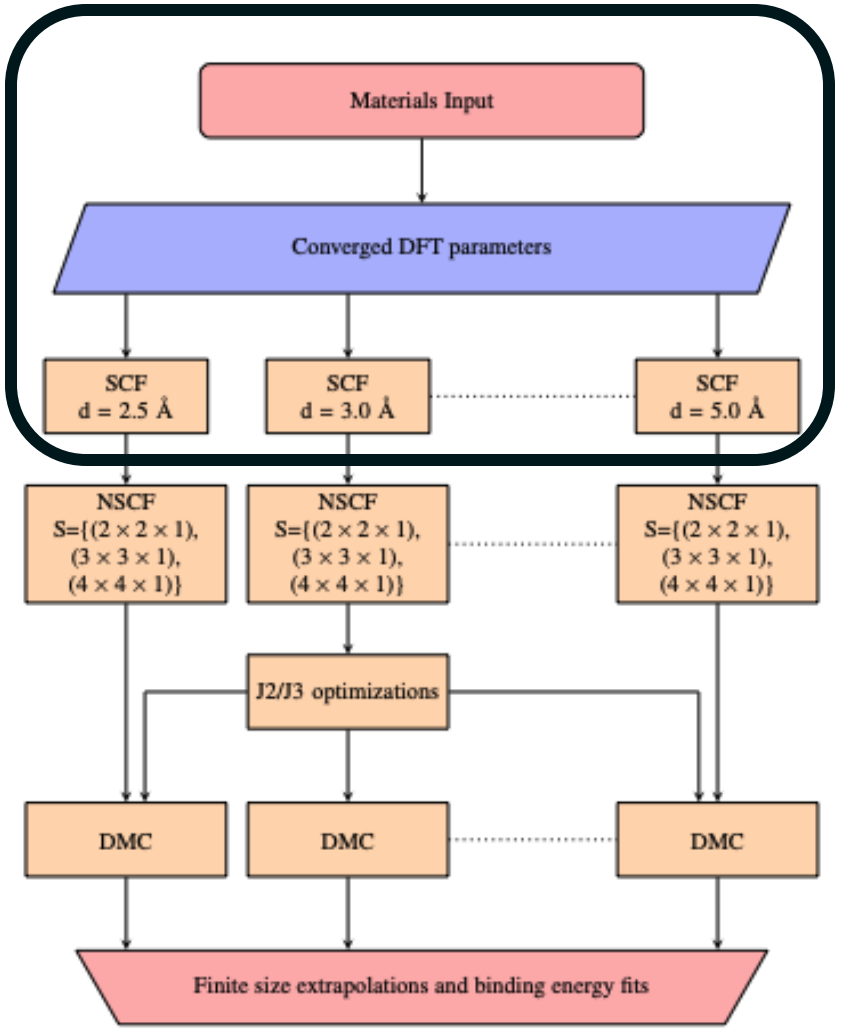
run.py

```
dft_shared = obj(
    kgrid    = (6,6,1),  # K-point grid for DFT calculations
    ecutwfc  = 400,      # Plane-wave cutoff energy in Rydberg
    pseudos  = 'B.ccECP.upf N.ccECP.upf'.split()  # DFT PP files for Boron and Nitrogen
)

qmc_shared = obj(
    # Hybrid representation cutoff radius for Boron and Nitrogen in atomic units (a.u.)
    hybrid_rcut  = obj(B=1.1, N=1.1),
    # Maximum angular momentum for hybrid representation for Boron and Nitrogen
    hybrid_lmax  = obj(B=5, N=5),
    # Blip-spline Mesh factor for QMC calculations
    meshfactor   = 0.5,
    # Pseudopotential files for QMC calculations
    pseudos      = 'B.ccECP.xml  N.ccECP.xml'.split()
)
```

SCF calculations

run.py

```
52  # SCF, NSCF and PW2QMCPACK settings
53  scf_shared, nscf_shared, conv_shared = get_dft_settings(**dft_shared)
54
55  # Binding energy workflow start
56  for d in interlayer_separations:
57      # Convert interlayer separation to an int for file naming
58      if isinstance(d, (int, float)):
59          d_name = int(d*1000)
60      else:
61          d_name = d
62
63      scf_path = 'scf_{}'.format(d_name)
64
65      # Generate the primitive cell system
66      prim_system = generate_physical_system(
67          structure = read_structure(structures[d]),
68          **system_shared
69          )
70      # SCF calculation
71      scf_run = generate_pwscf(
72          system = prim_system,
73          path = scf_path,
74          **scf_shared
75          )
```

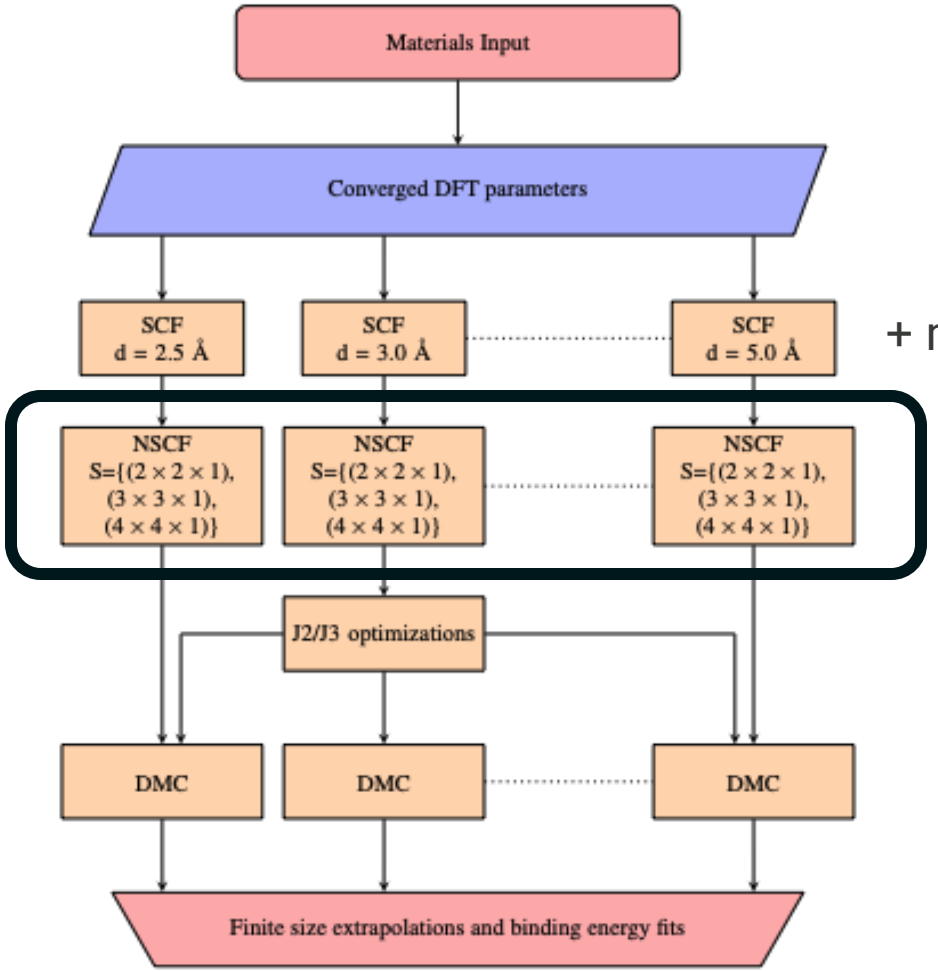


+ mono

NSCF/CONV calculations

run.py

```
76      for t in tiling_vectors:
77          # Directory for the NSCF calculation
78          nscf_path = 'nscf_{}_{}'.format(d_name, t[0])
79
80          # Generate the supercell system
81          tiled_system = generate_physical_system(
82              structure = read_structure(structures[d]),
83              tiling    = t,
84              kgrid     = tiling_kgrids[t],
85              **system_shared
86          )
87          # NSCF calculation
88          nscf_run = generate_pwscf(
89              system = tiled_system,
90              path = nscf_path,
91              **nscf_shared
92          )
93          # PW2QMCPACK conversion calculation
94          conv_run = generate_pw2qmcpack(
95              path         = nscf_path,    # Use the same path as the NSCF calculation
96              dependencies = (nscf_run, 'orbitals'),
97              **conv_shared
98          )
```



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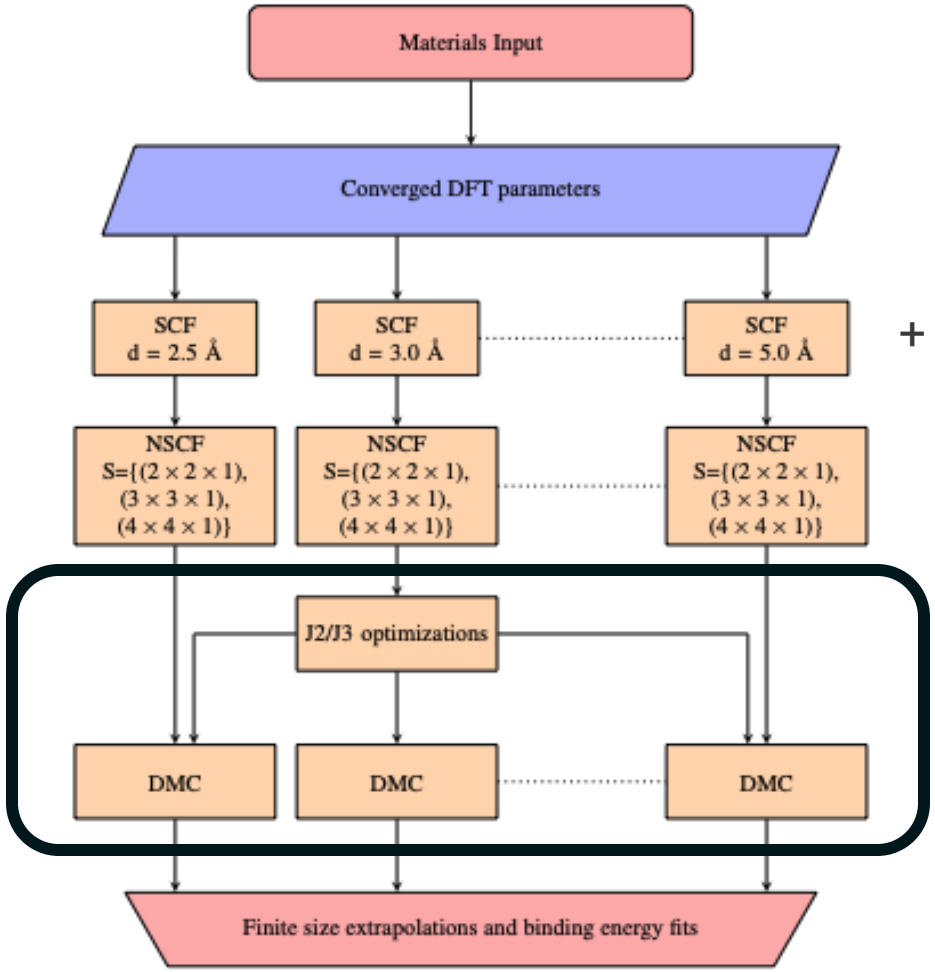
VMC/DMC calculations

run.py

```
99
100         dmc_path = 'dmc_{}_{}'.format(d_name, t[0])
101
102         # Optimize jastrows using the first structure listed in interlayer_separations
103         # In this example, this is d == 3.0 since dictionary keys are always ordered in Python 3.7+
104         j2_settings, j3_settings, dmc_settings  = get_qmc_settings(system = tiled_system,
105                                                                    **qmc_shared)
106         if d == interlayer_separations[0]:
107             j2_path = 'j2_{}_{}'.format(d_name, t[0])
108             j3_path = 'j3_{}_{}'.format(d_name, t[0])
109             # J2, J3 optimizations and DMC calculation settings
110             # Here each "settings" object is specific to the system size
111
112
113             # J2 optimization calculation
114             j2_run = generate_qmcpack(path = j2_path,
115                                       dependencies = (conv_run, 'orbitals'),
116                                       **j2_settings)
117             # J3 optimization calculation
118             j3_run = generate_qmcpack(path = j3_path,
119                                       dependencies = [(j2_run, 'jastrow'), (conv_run, 'orbitals')],
120                                       **j3_settings)
121         # DMC calculation
122         dmc_run = generate_qmcpack(path = dmc_path,
123                                       dependencies = [(j3_run, 'jastrow'),(conv_run, 'orbitals')],
124                                       **dmc_settings)
125 run_project()
```



+ mono

DFT settings

run_library.py

```python
#!/usr/bin/env python
# nexus imports
from nexus import Job, obj
from nexus import settings
from nexus import linear, loop, vmc, dmc
from qmcpack_input import spindensity
# general settings for nexus
settings(
    pseudo_dir    = './pseudos',
    status_only   = 0,                  # only show status of runs
    generate_only = 0,                  # only make input files
    sleep         = 3,                  # check on runs every 3 secondsa
    machine       = 'perlmutter',       # Perlmutter NERSC machine
    account       = '<account_name>',   # User account name
)

def get_dft_settings(kgrid     = None,
                     ecutwfc   = None,
                     pseudos   = None,
                     start_mag = None,
                     hubbard   = None,
                     tot_magnetization = None):


    if settings.machine == 'perlmutter':
        qe_modules = '' # Modules used to build QE
        qe_bin     = '' # QE build directory
        dft_job = Job(cores  = 4,
                      threads= 1,
                      hours  = 12,
                      app    = qe_bin+'/pw.x',
                      constraints = 'cpu', # Default
                      presub= qe_modules)
        conv_job = Job(cores=1,
                       hours=1,
                       app='/pw2qmcpack.x',
                       constraints = 'cpu',
                       presub=qe_modules)
    else:
        print('Error: Unknown computer for DFT, using {}'.format(settings.machine))
        exit()
```

```python

    qe_shared = obj(
        job          = dft_job,
        input_type   = 'generic',
        ecutwfc      = ecutwfc,          # DFT planewave energy cutoff
        input_DFT    = 'PBE',            # DFT functional
        conv_thr     = 1e-8,             # SCF convergence threshold
        wf_collect   = True,             # write orbitals
        pseudos      = pseudos,          # QE Pseudopotentials
        start_mag    = start_mag,        # Starting magnetization
        hubbard      = hubbard,          # Hubbard U parameters
        occupations  = 'smearing',       # Occupation scheme
        smearing     = 'gauss',          # Smearing type
        degauss      = 0.001,            # Smearing widt
        tot_magnetization = tot_magnetization
    )

    scf_shared = obj(
        nosym        = False,            # use symmetry
        identifier   = 'scf',            # identifier/file prefix
        calculation  = 'scf',            # perform scf calculation
        kgrid        = kgrid,            # Converged DFT k-grid
        **qe_shared
    )

    nscf_shared = obj(
        nosym        = True,             # don't use symmetry
        identifier   = 'nscf',           # identifier/file prefix
        calculation  = 'nscf',           # perform nscf calculation
        diagonalization = 'cg',          # Diagonalization method
        **qe_shared
    )

    conv_shared = obj(
            identifier   = 'conv',           # identifier/file prefix
            job          = conv_job,         # Job object for PW2QMCPACK
            write_psir   = False,            # Don't write psir
    )

    return scf_shared, nscf_shared, conv_shared
```

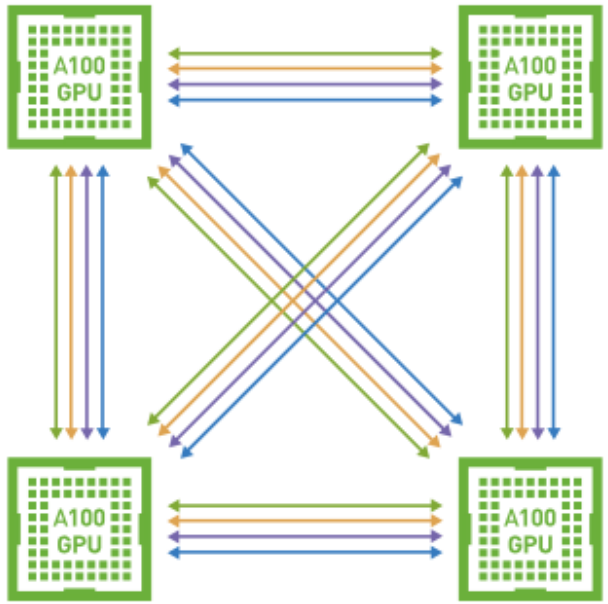
DMC is more sensitive to phase factor than DFT (**S5**)

QMC settings

run_library.py

```
83  def get_qmc_settings(system      = None,
84                       hybrid_rcut = None,
85                       hybrid_lmax = None,
86                       meshfactor  = 1.0,
87                       pseudos     = None):
88
89      if settings.machine == 'perlmutter':
90          qmcpack_modules = ''    # Modules used to build QMCPACK
91          qmcpack_bin     = ''    # QMCPACK build directory
92          qmcpack_exec    = qmcpack_bin+'/qmcpack_complex'
93          opt_job = Job(nodes    = 12,
94                        threads = 16,
95                        hours    = 12,
96                        constraint = 'gpu',
97                        app      = qmcpack_exec,
98                        presub   = qmcpack_modules)
99          dmc_job = Job(nodes    = 24,
100                       threads = 16,
101                       hours    = 12,
102                       constraint = 'gpu',
103                       app      = qmcpack_exec,
104                       presub   = qmcpack_modules)
105     else:
106         print('Error: Unknown computer for QMC, using {}'.format(settings.machine))
107         exit()
```

Perlmutter architecture



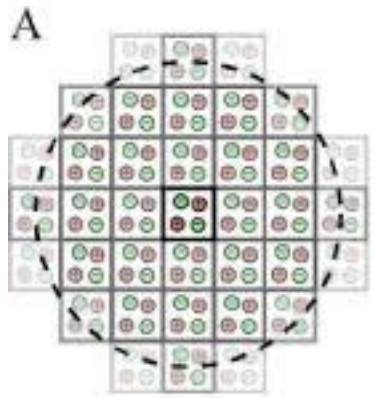
- Single AMD EPYC 7763 (Milan) CPU
- 64 cores per CPU
- Four NVIDIA A100 (Ampere) GPUs

https://docs.nersc.gov/systems/perlmutter/architecture/

QMC settings

run_library.py

```
109     system.structure.change_units('B')
110     rwigner = system.structure.rwigner()
111
112     qmc_settings = obj(
113         system          = system,        # PhysicalSystem object containing structural info
114         input_type      = 'basic',       # Simple input format for QMCPACK
115         pseudos         = pseudos,       # Pseudopotential files for QMC
116         driver          = 'batched',     # Use batched driver in QMCPACK
117         hybrid_rcut     = hybrid_rcut,   # Cutoff radius for hybrid orbital representation
118         hybrid_lmax     = hybrid_lmax,   # Max angular momentum for hybrid orbitals
119         meshfactor      = meshfactor,    # Controls fineness of real-space (Spline) grid
120         lr_handler      = 'ewald',       # Use Ewald summation for long-range interactions
121         lr_dim_cutoff   = 30,            # Cutoff for long-range Ewald sums
122         spin_polarized  = True,          # Enable spin-polarized calculations
123     )
124
125     opt_parameters = obj(
126         num_varmin_j2   = 12,     # Number of variance minimization iterations for 2-body Jastrow
127         num_emin_j2     = 8,      # Number of energy minimization iterations for 2-body Jastrow
128         num_emin_j3     = 6,      # Number of energy minimization iterations for 3-body Jastrow
129         j2_init         = "rpa",  # Initialize 2-body Jastrow with Random Phase Approximation
130         num_j1_jastrows = 10,     # Number of 1-body Jastrow parameters to optimize
131         num_j2_jastrows = 10,     # Number of 2-body Jastrow parameters to optimize
132         num_j3_jastrows = 3,      # Number of 3-body Jastrow parameters to optimize
133         j3_rcut         = 4.0 if rwigner > 4.0 else rwigner,  # 3-body Jastrow cutoff radius (min of 4.0 or Wigner radius)
134         timestep        = 1.0     # VMC timestep for optimization
135     )
136
137     opt_settings = obj(
138         job             = opt_job,
139         twistnum        = 0,     # Twist index
140     )
141     opt_settings = opt_settings.set(qmc_settings)
```



J. Chem. Phys. 124, 234104 (2006)

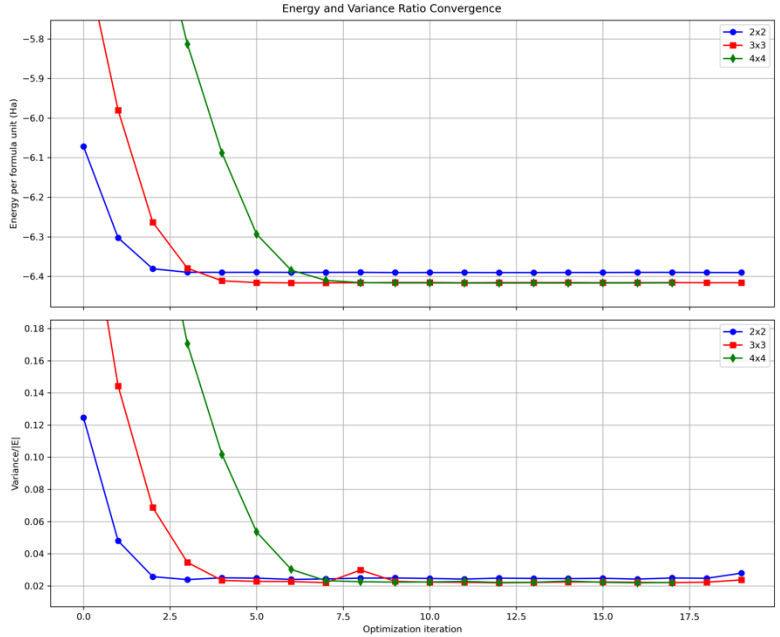
1. J2-varmin
2. J2-emin
3. J2+J3-emin

QMC settings

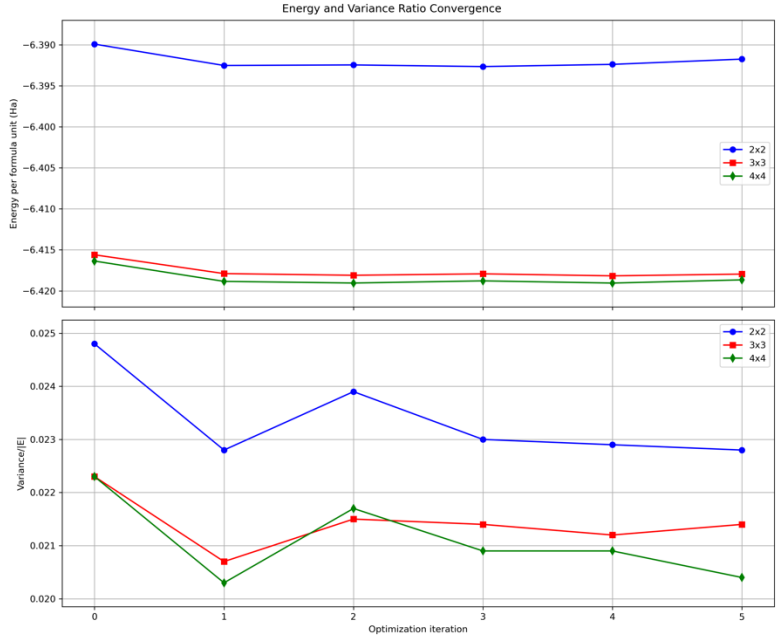
run_library.py

```
    # Variance minimization settings
    varmin = linear(
        energy               = 0.0,              # Weight for energy minimization (0 = pure variance min)
        unreweightedvariance = 1.0,              # Weight for unreweighted variance minimization
        reweightedvariance   = 0.0,              # Weight for reweighted variance minimization
        minwalkers           = 1e-4,             # Lower bound of the effective walker weight
        shift_i              = 0.05,             # (OneShiftOnly Optimizer) Direct stabilizer shift
        shift_s              = 1.0,              # (OneShiftOnly Optimizer) Stabilizer shift based on overlap matrix
        warmupsteps          = 200,              # Number of steps before measurements begin
        blocks               = 100,              # Number of statistical measurement blocks
        steps                = 1,                # Steps per block
        timestep             = 1.0,              # VMC timestep
        minmethod            = "OneShiftOnly",   # Minimization algorithm to use
        substeps             = 10,               # Number of MC steps between parameter updates
    )

    # Energy minimization settings
    emin = varmin.copy() # Copy from varmin
    emin.minwalkers             = 0.5  # Use larger minwalkers, since varmin provides a better starting point
    emin.energy                 = 0.95 # Mixed cost function 0.95 energy / 0.05 variance
    emin.unreweightedvariance   = 0.0
    emin.reweightedvariance     = 0.05
    emin.shift_i                = 0.01 # Reduced shift_i, since we are closer to the minimum

    j2_settings     = obj(
        calculations = [loop(max=opt_parameters.num_varmin_j2, qmc=varmin),
                        loop(max=opt_parameters.num_emin_j2,   qmc=emin)],
        J1_size = opt_parameters.num_j1_jastrows,
        J2_size = opt_parameters.num_j2_jastrows,
        J1_rcut = rwigner,
        J2_rcut = rwigner,
        J2_init = opt_parameters.j2_init,
        **opt_settings
    )

    j3_settings     = obj(
        calculations = [loop(max=opt_parameters.num_emin_j3, qmc=emin)],
        J3=True,
        J3_isize = opt_parameters.num_j3_jastrows,
        J3_esize = opt_parameters.num_j3_jastrows,
        J3_rcut  = opt_parameters.j3_rcut,
        **opt_settings
    )
```

J2-opt



J3-opt

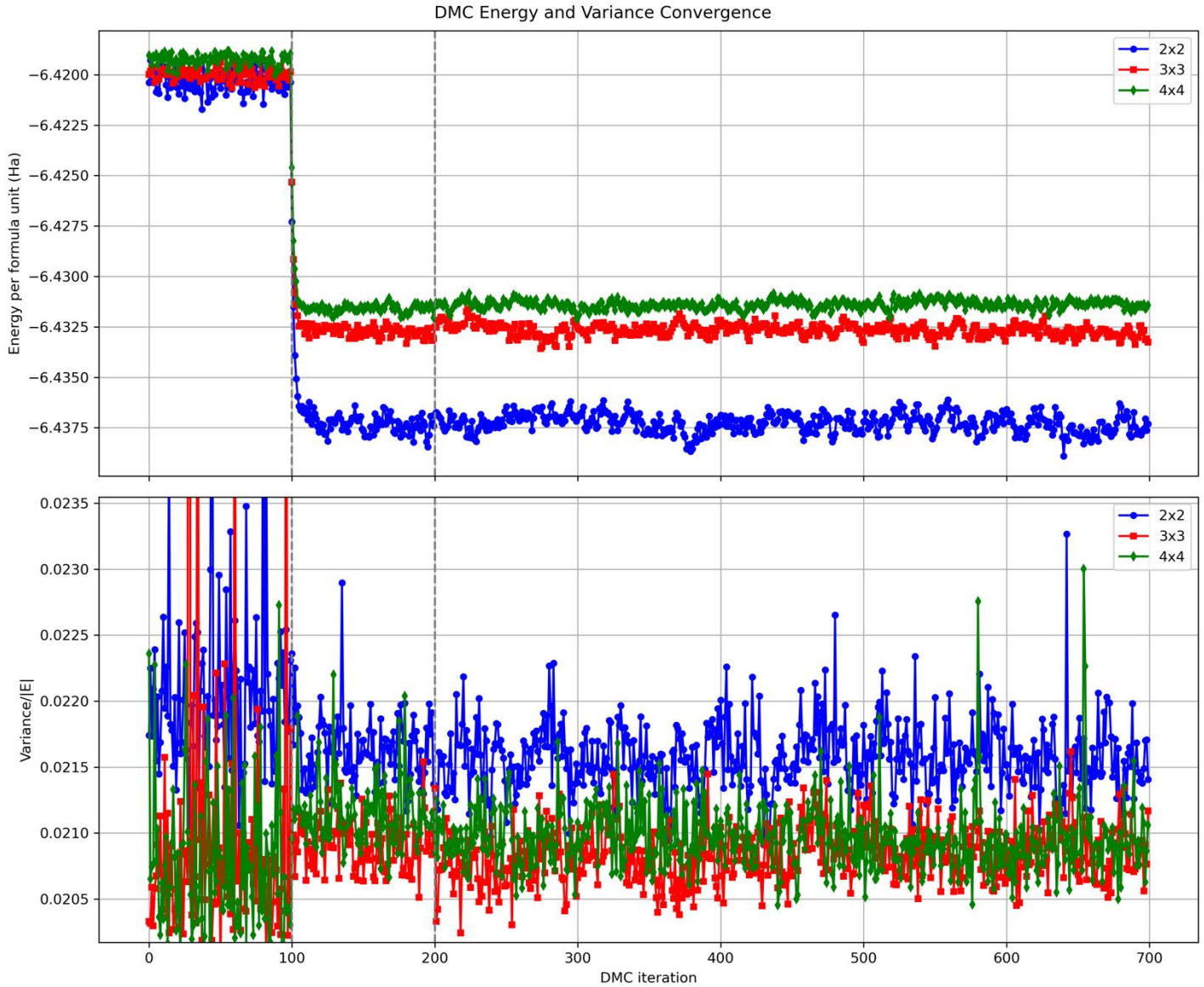


QMC settings

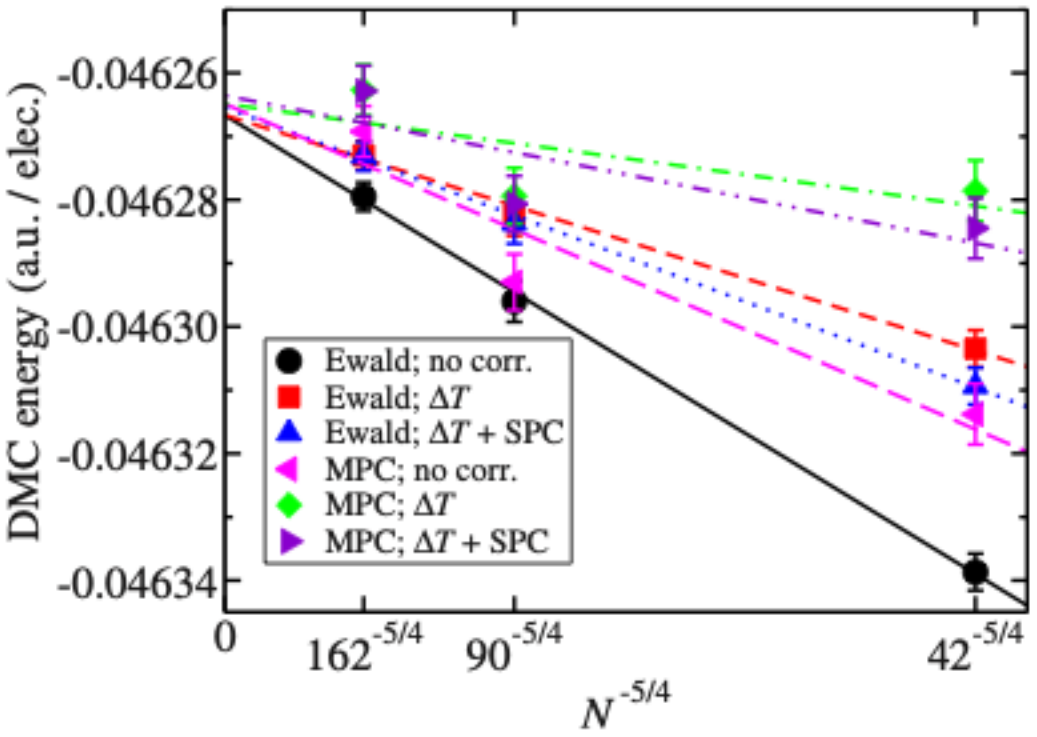
run_library.py

```
    dmc_parameters = obj(
        vmcdt                = 0.3,     # VMC timestep in atomic units
        vmcwarmup            = 25,      # Number of VMC blocks to equilibrate
        vmcblocks            = 100,     # Number of VMC measurement blocks
        vmcsubsteps          = 4,       # VMC steps between measurements
        dmc_eq_dt            = 0.02,    # DMC equilibration timestep
        dmc_eq_blocks        = 100,     # Number of DMC equilibration blocks
        dmcdt                = 0.005,   # DMC production timestep
        dmcblocks            = 500,     # Number of DMC production blocks
        dmcwarmup            = 100,     # Number of DMC blocks to equilibrate
        dmcsteps             = 10,      # Steps per DMC block
        vmc_walkers_per_rank = 240,     # Number of VMC walkers per MPI rank
        dmc_walkers_per_rank = 240,     # Number of DMC walkers per MPI rank
        nonlocalmoves        = False,   # Use T-moves for non-local pseudopotentials
    )
    vmc_dmc = obj(
        warmupsteps = dmc_parameters.vmcwarmup,
        blocks      = dmc_parameters.vmcblocks,
        steps       = 1,
        timestep    = dmc_parameters.
        substeps    = dmc_parameters.vmcsubsteps,
        walkers_per_rank = dmc_parameters.vmc_walkers_per_rank
    )
    dmc_eq  = obj(
        warmupsteps = dmc_parameters.dmcwarmup,
        blocks      = dmc_parameters.dmc_eq_blocks,
        steps       = dmc_parameters.dmcsteps,
        timestep    = dmc_parameters.dmc_eq_dt,
        walkers_per_rank = dmc_parameters.dmc_walkers_per_rank,
        nonlocalmoves = dmc_parameters.nonlocalmoves,
    )
    dmc_stat = obj(
        warmupsteps = dmc_parameters.dmcwarmup,
        blocks      = dmc_parameters.dmcblocks,
        steps       = dmc_parameters.dmcsteps,
        timestep    = dmc_parameters.dmcdt,
        walkers_per_rank = dmc_parameters.dmc_walkers_per_rank,
        nonlocalmoves = dmc_parameters.nonlocalmoves,
    )

    dmc_settings = obj(
        job           = dmc_job,
        calculations  = [vmc(**vmc_dmc), dmc(**dmc_eq), dmc(**dmc_stat)],
        estimators    = [spindensity(dr=3*[0.3])],
        **qmc_settings
    )

    return j2_settings, j3_settings, dmc_settings
```

(function) vmcsubsteps: Any

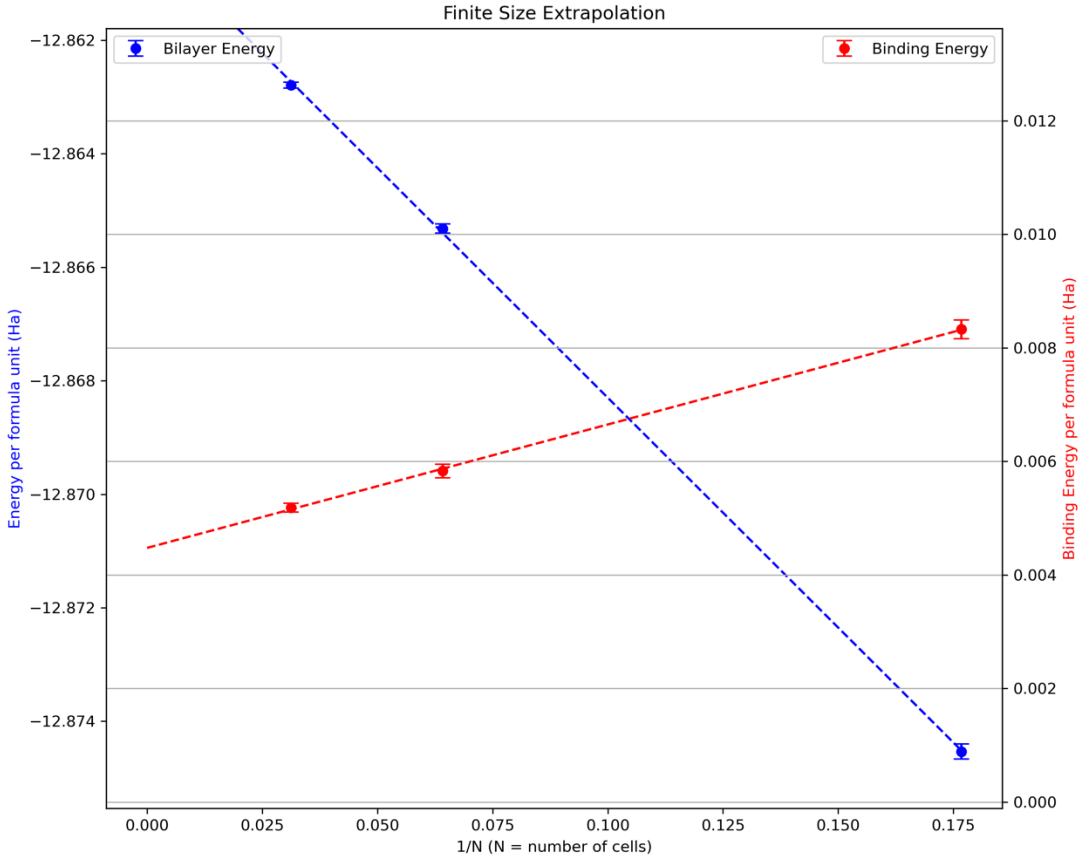


Finite size extrapolation in 2D



Drummond et al, PRB **78**, 125106 (2008)

hBN d = 2.5 A



Next session: Running on GPUs and Surrogate Hessian Geom. Opt.