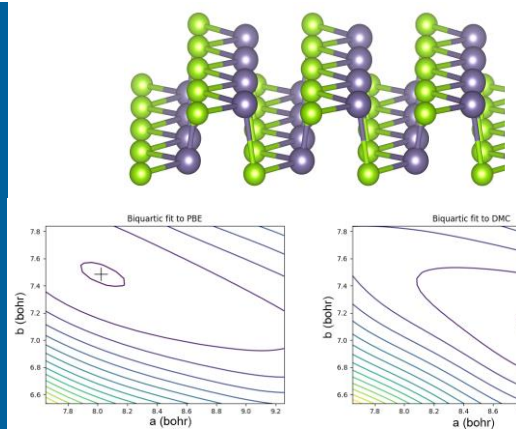
REAL-WORLD CALCULATION : STRUCTURAL AND OPTICAL PROPERTIES OF GESE MONOCHALCOGENIDE FROM QUANTUM MONTE CARLO SIMULATIONS



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CPSFM | Center for Predictive Simulation of Functional Materials

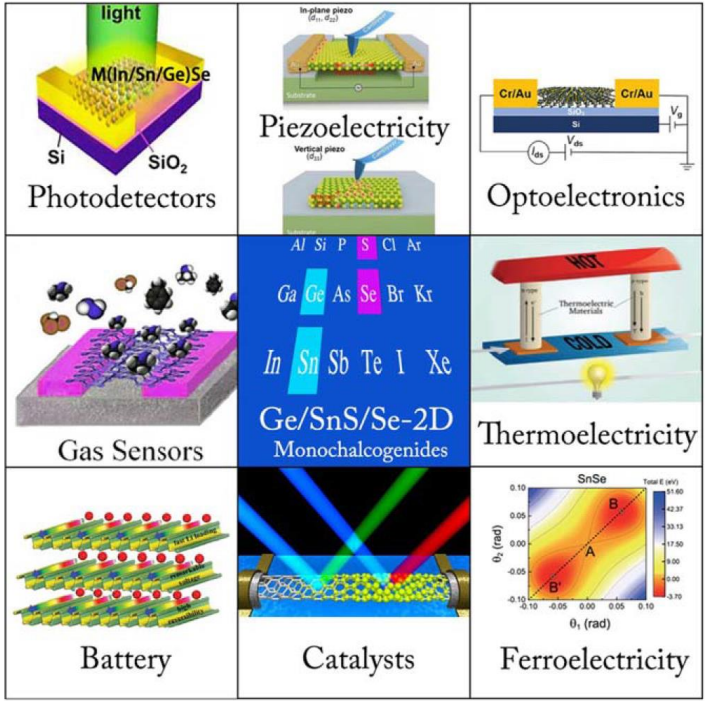
OUTLINES

1. Introduction
   - 2D monochalcogenide
   - Sensitivity of electronic & optical properties
   - First-principle study for 2D monochalcogenides
2. DFT and QMC studies for bulk GeSe
   - Equation of states
   - Excited states
3. DFT and QMC studies for monolayer GeSe
   - Geometry optimization
   - Excited states

INTRODUCTION

- 2D monochalcogenides

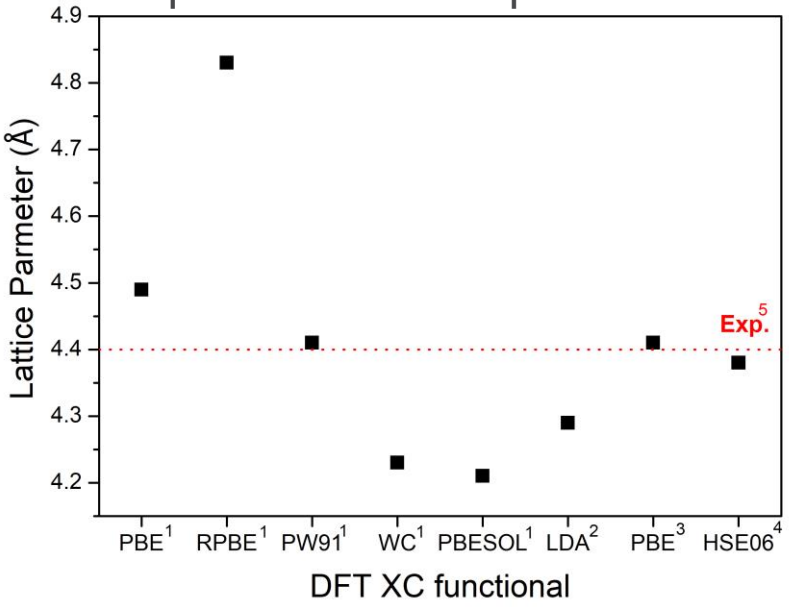
- Replace graphene that is hard difficult to create semiconductor.
- Puckered layered orthorhombic structure (Pnma-D^{16}_{2h})
- Various polytypes & electronic structures
- **Strong vdW interaction**
  : layer dependent electronic & optical properties
  : wider band gap with monolayer than bulk
- **Tunable electronic properties**
  : with strain, electric field, defects/doping etc.
- Various applications
  : photodetector, gas sensors, catalysts, etc.



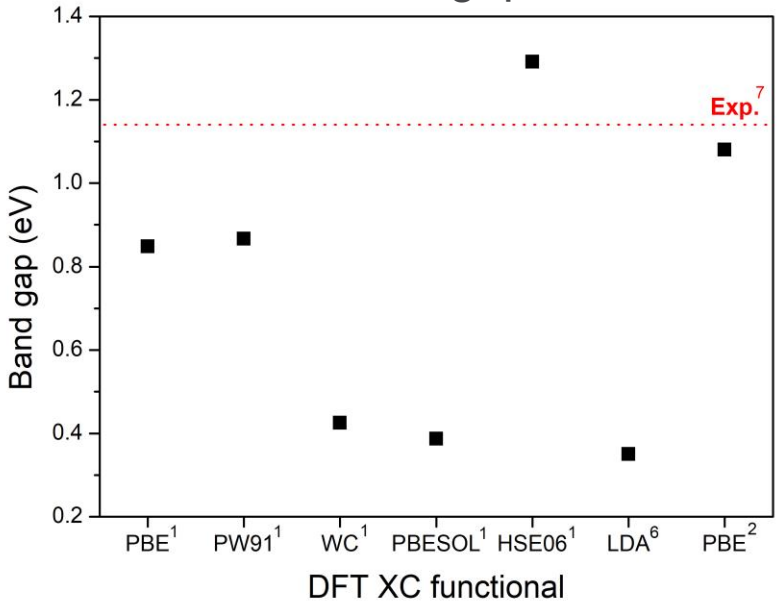
Z. Hu *et al.*, Nanotechnology 30, 252001 (2019).

DFT STUDIES ON BULK GeSe

Equilibrium lattice parameter



Band gap



DFT results are very scattered and no XC functional satisfying accurate structural and optical properties at same time.

[1] Y. Hu et al., Appl. Phys. Lett. 107, 122107 (2015).
[2] L. Makinistian and E. A. Albanesi, J. Phys.: Condens. Matter 19, 186211 (2007).
[3] A. K. Singh and R. G. Henning, Appl. Phys. Lett. 105, 042103 (2014).
[4] G. Valiukonis et al., Phys. Status Solidi b 135, 299 (1986).
[5] R. W. G. Wyckoff, Crystal Structures, 2nd Edition, Interscience Publishers, New York (1963).
[6] H. C. Hsueh, et al., Phys. Rev. B 51, 16750 (1995).
[7] D. D. Vaughn II, et al., J. Am. Chem. Soc. 132, 15170 (2010).

FIRST-PRINCIPLE STUDY FOR GESE

- Monochalcogenides (including GeSe) are difficult to study with DFT

1. accurate description of vdW interaction is required due to the strong dependencies of electronic and optical properties on number of layers.

2. Electronic and optical properties are very sensitive with the geometry
-> Accurate geometry estimation is required if their experimental result of equilibrium geometry is not known. (e.g. δ- and ε-InSe, monolayer GeSe)

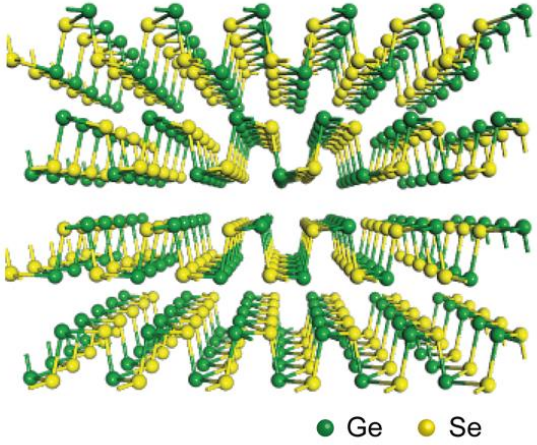
3. DFT electronic and optical properties, and optimized geometry are very scattered depending on exchange-correlation functional.
-> hard to obtain accurate observables at the same time.

➡ **Quantum Monte Carlo method**

DFT AND QMC STUDY ON BULK GESE

BULK GESE

- high thermodynamic stability
- low toxicity, environmental friendliness
- high oxidation resistivity
- potential applications in random access memory, etc.

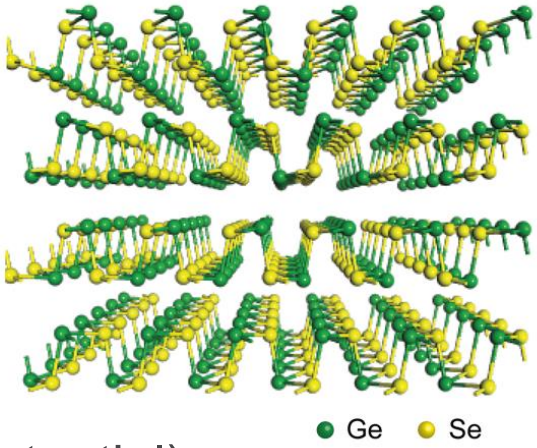


S.-C. Liu *et al*., Nat. Commun. 12, 670 (2021).

- geometry and electronic properties are well-known.

: Known as an “Indirect gap” semiconductor (1.07 ~ 1.29 eV) with different experimental methods.
(energy-electron loss, diffuse reflectance, UV photoemission)
: Recently claimed possibility of a direct gap semiconductor (1.3 eV) with adsorption spectra measurement.
-> high sensitivity of the experimentally measured optical properties

COMPUTATIONAL DETAILS



- Experimental result of GeSe geometry
  : a = 4.38, b = 3.82, and c = 10.79 Å (Pnma)

- Plane wave basis set
  : PBE with 350 Ry kinetic energy cut-off
  (Burkatzki-Filippi-Dolg energy-consistent pseudopotential)
  : 12x12x12 Monkhorst-Pack grids

- DMC calculations
  : 0.005 Ha^{-1} time step (fully converged)
  : One-body finite size effects are controlled by twist-averaged boundary condition (TABC).
  : Two-body finite size effect is reduced by the extrapolation of total energies.

EQUATION OF STATE FOR BULK GeSe

```
a0 = 4.38

scales = [0.8, 0.9, 1.0, 1.1, 1.2]

for scale in scales:
 a = a0 * scale
 system = generate_physical_system(
    units  = 'A',
    axes   = [[a,0.,0.   ],
              [0.   ,a*0.872146118721461,0.],
              [0.,0.   ,a*2.463470319634703]],
    elem   = ['Ge','Ge','Ge','Ge','Se','Se','Se','Se'],
    posu   = [[0.106    ,0.25    ,0.879    ],
              [0.394    ,0.25    ,0.379    ],
              [0.894    ,0.75    ,0.121    ],
              [0.606    ,0.75    ,0.621    ],
              [0.503    ,0.25    ,0.148    ],
              [0.997    ,0.25    ,0.648    ],
              [0.497    ,0.75    ,0.852    ],
              [0.003    ,0.75    ,0.352    ]],
    net_spin   = 0,
    net_charge = 0,
    tiling     = [[1, 0, 0],[0, 1, 0],[0, 0, 1]],
    symm_kgrid = True,
    kgrid  = (1,1,1),
    kshift = (0,0,0),
    Ge      = 4,
    Se      = 6,
 )
```

Lattice vectors

Atomic coordinates

```
dft_shared = obj(
    input_type       = 'generic',
    input_dft        = 'pbe',
    ecutwfc          = 350,
    occupations      = 'smearing',
    smearing         = 'fermi-dirac',
    degauss          = 1e-3,
    mixing_beta      = 0.5,
    nspin            = 2,
    start_mag        = obj(Ge=0.2),
    kshift           = (0,0,0),
    pseudos          = ['Ge.ccECP.upf','Se.ccECP.upf'],
    )


 basepath = '{}/'.format(scale)

  scf = generate_pwscf(
      identifier        = 'scf',
      path              = basepath + 'scf',
      job               = job(cores=cores, app='pw.x'),
      calculation       = 'scf',
      system            = system,
      kgrid             = (12,12,12),
      **dft_shared
      )
```

Pwscf SCF

```
nscf = generate_pwscf(
    identifier        = 'nscf',
    path              = basepath + 'nscf',
    job               = job(cores=cores, app='pw.x'),
    calculation       = 'nscf',
    system            = system,
    nosym             = True,
    nogamma           = True,
    dependencies      = (scf,'charge_density'),
    **dft_shared
    )
```

Pwscf NSCF

```
p2q = generate_pw2qmcpack(
    identifier        = 'p2q',
    path              = basepath + 'nscf',
    job               = job(cores=cores,app='pw2qmcpack.x'),
    write_psir        = False,
    dependencies      = (nscf,'orbitals'),
    )
```

Pwscf p2q

Argonne NATIONAL LABORATORY

EQUATION OF STATE FOR BULK GeSe

```
qmc_shared = obj(
    driver          = 'batched',
    input_type      = 'basic',
    meshfactor      = 1.00,
    spin_polarized = True,
    pseudos         = ['Ge.ccECP.xml','Se.ccECP.xml'],
    )
```

```
optJ12 = generate_qmcpack(
    identifier          = 'optJ12',
    path                = basepath + 'optJ12',
    job                 = job(cores=cores,app='qmcpack'),
    system              = system,
    twistnum            = 0,
    qmc                 = 'opt',
    J2                  = True,
    minmethod           = 'oneshift',
    warmupsteps         = 10,
    init_cycles         = 5,
    cycles              = 5,
    blocks              = 10,
    samples             = 51200,
    substeps              = 20,
    minwalkers          = 0.3,
    corrections         = [],
    dependencies        = (p2q,'orbitals'),
    **qmc_shared
    )
```

J12 opt

```
J3_rcut = system.structure.rwigner()
```

```
optJ123 = generate_qmcpack(
    identifier          = 'optJ123',
    path                = basepath + 'optJ123',
    job                 = job(cores=cores,app='qmcpack'),
    system              = system,
    twistnum            = 0,
    qmc                 = 'opt',
    J2                  = True,
    J3                  = True,
    J3_rcut             = J3_rcut,
    minmethod           = 'oneshift',
    warmupsteps         = 10,
    init_cycles         = 0,
    cycles              = 10,
    blocks              = 10,
    samples             = 51200,
    substeps              = 20,
    minwalkers          = 0.5,
    corrections         = [],
    dependencies        = [(p2q,'orbitals'), (optJ12,'jastrow')],
    **qmc_shared
    )
```

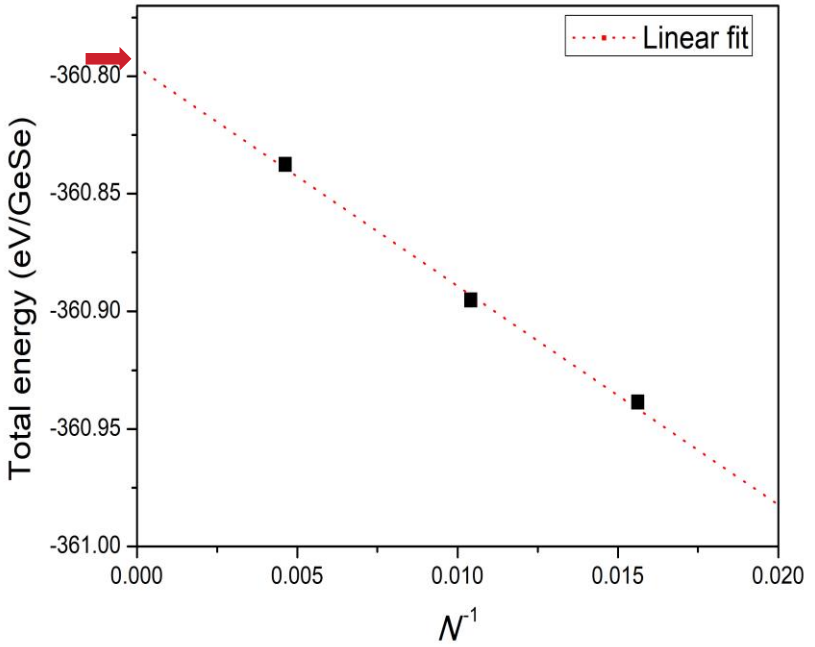
J3 opt

```
qmc = generate_qmcpack(
    identifier          = 'qmc',
    path                = basepath + 'qmc',
    job                 = job(cores=cores,app='qmcpack'),
    system              = system,
    calculations        = [
    vmc(
        total_walkers = 2048,     # Total walkers are small. Need to increase.
        warmupsteps     = 100,
        blocks          = 20,
        steps           = 20,
        timestep        = 0.3,
        ),
    dmc( # main dmc
        total_walkers = 2048,
        timestep        = 0.005,
        warmupsteps     = 1000,
        blocks          = 200,
        steps           = 20,
        nonlocalmoves   = 'v3',
        ),
    ],
    dependencies        = [(p2q,'orbitals'), (optJ123,'jastrow')],
    **qmc_shared
    )
```

DMC

FINITE SIZE ANALYSIS

Finite size extrapolation




```
a0 = 4.38

scales = [0.8, 0.9, 1.0, 1.1, 1.2]

 or scale in scales:
 a = a0 * scale
 system = generate_physical_system(
    units  = 'A',
    axes   = [[a,0.,0.   ],
              [0.   ,a*0.872146118721461,0.],
              [0.,0.   ,a*2.463470319634703]],
    elem   = ['Ge','Ge','Ge','Ge','Se','Se','Se','Se'],
    posu   = [[0.106    ,0.25    ,0.879    ],
              [0.394    ,0.25    ,0.379    ],
              [0.894    ,0.75    ,0.121    ],
              [0.606    ,0.75    ,0.621    ],
              [0.503    ,0.25    ,0.148    ],
              [0.997    ,0.25    ,0.648    ],
              [0.497    ,0.75    ,0.852    ],
              [0.003    ,0.75    ,0.352    ]],
    net_spin   = 0,
    net_charge = 0,
    tiling     = [[1, 0, 0],[0, 1, 0],[0, 0, 1]],
    symm_kgrid = True,
    kgrid  = (1,1,1),
    kshift = (0,0,0),
    Ge      = 4,
    Se      = 6,
)
```

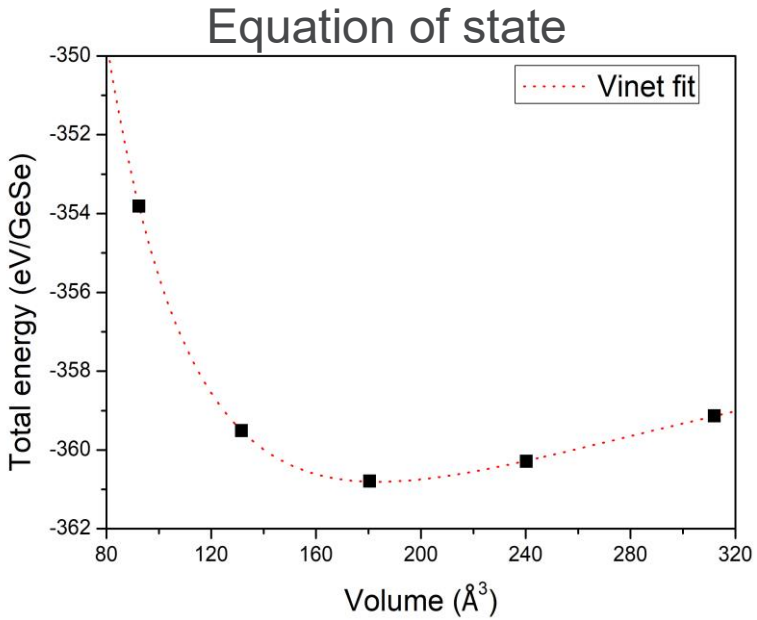
Finite size extrapolation

S8 tiling = [[2, 2, 0],[1, 0, 1],[2, -2, 0]],
S12 tiling = [[0, 2, 1],[3, 0, 0],[0, 2, -1]],
S18 tiling = [[3, 0, 0],[0, 3, 0],[0, 0, 2]],
S27 tiling = [[3, 0, 0],[0, 3, 0],[0, 0, 3]],
:

Twist-averaged boundary condition

kgrid = (2,2,2),
kgrid = (3,3,3),
kgrid = (4,4,4),
:

EQUATION OF STATE FOR BULK GeSe



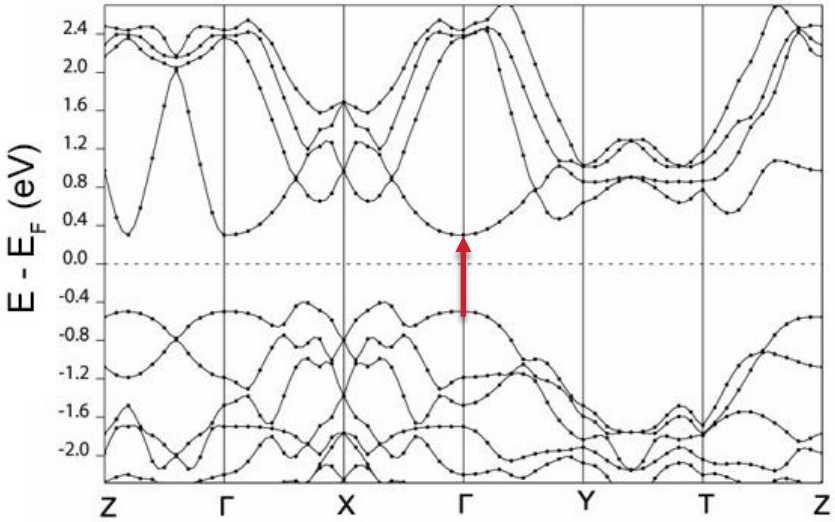
	PBE	LDA	SCAN	PBE0	B3LYP	HSE06	DMC	Exp.
a (Å)	4.43	4.38	4.43	4.38	4.48	4.38	**4.40(1)**	**4.40**[1]
B_0 (GPa)	28.9	32.8	30.0	33.2	27.1	32.4	**32.1(3)**	**37.9**[2]
E_{coh} (eV)	6.89	8.54	-	6.96	6.06	5.97	**6.91(2)**	-

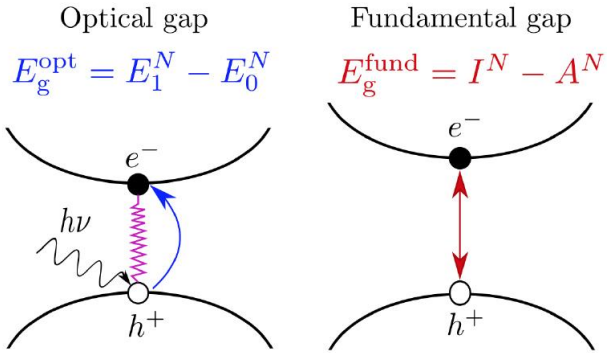
[1] R. W. G. Wyckoff, Crystal Structures, 2nd Edition, Interscience Publishers, New York (1963).
[2] H. C. Hsueh and J. Crain, Phys. Stat. Sol. (b) 211, 365 (1999).

H. Shin *et al*., Phys. Rev. Materials 5, 024002 (2021).

OPTICAL PROPERTIES FOR BULK GeSe

PBE band structure





X. Blase *et al.*, J. Phys. Chem. Lett. 11, 7371 (2020).

1. Excitonic(optical) gap
(bound electron-hole pair)

2. Quasiparticle(fundamental) gap
(unbound electron-hole pair)

H. Shin *et al.*, Phys. Rev. Materials 5, 024002 (2021).

EXCITONIC GAP

1. Excitonic(optical) gap (bound electron-hole pair)

$$\Delta_{EX}(K \to K') = E_{ex}(K \to K') - E_{gr}$$

```
<sposet name="spo-up" size="20">
  <occupation mode="ground" spindataset="0"/>
</sposet>
<sposet name="spo-dn" size="20">
  <occupation mode="ground" spindataset="1"/>
</sposet>
```

```
<sposet name="spo-up" size="20">
  <occupation mode="excited" spindataset="0">
     -20 +21
  </occupation>
</sposet>
<sposet name="spo-dn" size="20">
  <occupation mode="ground" spindataset="1"/>
</sposet>
```

```
<sposet name="spo-up" size="20">
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     0 19 0 20
  </occupation>
</sposet>
<sposet name="spo-dn" size="20">
  <occupation mode="ground" spindataset="1"/>
</sposet>
```

einspline.tile_100010001.spin_0.tw_0.g0.bandinfo.dat

# Band	State	TwistIndex	BandIndex	Energy	Kx	Ky	Kz	K1	K2	K3	KmK
0	0	0	0	-0.302241	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
1	1	0	1	-0.288033	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
2	2	0	2	-0.247845	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
3	3	0	3	-0.224796	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
4	4	0	4	-0.062419	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
5	5	0	5	-0.036102	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
6	6	0	6	0.019171	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
7	7	0	7	0.037928	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
8	8	0	8	0.045392	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
9	9	0	9	0.087316	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
10	10	0	10	0.095284	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
11	11	0	11	0.100719	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
12	12	0	12	0.105450	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
13	13	0	13	0.122192	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
14	14	0	14	0.155849	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
15	15	0	15	0.162660	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
16	16	0	16	0.181928	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
17	17	0	17	0.194166	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
18	18	0	18	0.202680	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
19	19	0	19	0.231322	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
20	20	0	20	0.258623	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
21	21	0	21	0.329408	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
22	22	0	22	0.334187	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
23	23	0	23	0.337607	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1

# Band	State	TwistIndex	BandIndex	Energy	Kx	Ky	Kz	K1	K2	K3	KmK
0	0	0	0	-0.302241	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
1	1	0	1	-0.288033	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
2	2	0	2	-0.247845	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
3	3	0	3	-0.224796	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
4	4	0	4	-0.062419	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
5	5	0	5	-0.036102	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
6	6	0	6	0.019171	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
7	7	0	7	0.037928	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
8	8	0	8	0.045392	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
9	9	0	9	0.087316	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
10	10	0	10	0.095284	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
11	11	0	11	0.100719	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
12	12	0	12	0.105450	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
13	13	0	13	0.122192	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
14	14	0	14	0.155849	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
15	15	0	15	0.162660	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
16	16	0	16	0.181928	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
17	17	0	17	0.194166	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
18	18	0	18	0.202680	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
19	19	0	20	0.258623	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
20	20	0	19	0.231322	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
21	21	0	21	0.329408	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
22	22	0	22	0.334187	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
23	23	0	23	0.337607	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1

QUASI-PARTICLE GAP

2. Quasiparticle gap (unbound electron-hole pair)

$$\Delta_{QP} = EA - IP$$
$$= \left(E_{N+1}^{CBM} - E_N^{K'}\right) - \left(E_N^{K'} - E_{N-1}^{VBM}\right)$$
$$= E_{N+1}^{CBM} + E_{N-1}^{VBM} - 2E_N^{K'}$$

```
<particleset name="e" random="yes" randomsrc="i">
  <group name="u" size="21">
    <parameter name="charge">-1</parameter>
  </group>
  <group name="d" size="20">
    <parameter name="charge">-1</parameter>
  </group>
</particleset>
<wavefunction name="psi0" target="e">
  <sposet_collection type="einspline" href="../GeSe.pwscf.h5" tilematrix="1 0 0 0 1 0 0 0 1" source="i"  twistnum="0" gpu="yes" meshfactor="1.0" precision="single">
    <sposet name="spo-up" size="21">
      <occupation mode="ground" spindataset="0"/>
    </sposet>
    <sposet name="spo-dn" size="20">
      <occupation mode="ground" spindataset="1"/>
    </sposet>
```

```
<particleset name="e" random="yes" randomsrc="i">
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    <parameter name="charge">-1</parameter>
  </group>
  <group name="d" size="20">
    <parameter name="charge">-1</parameter>
  </group>
</particleset>
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      <occupation mode="ground" spindataset="1"/>
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```
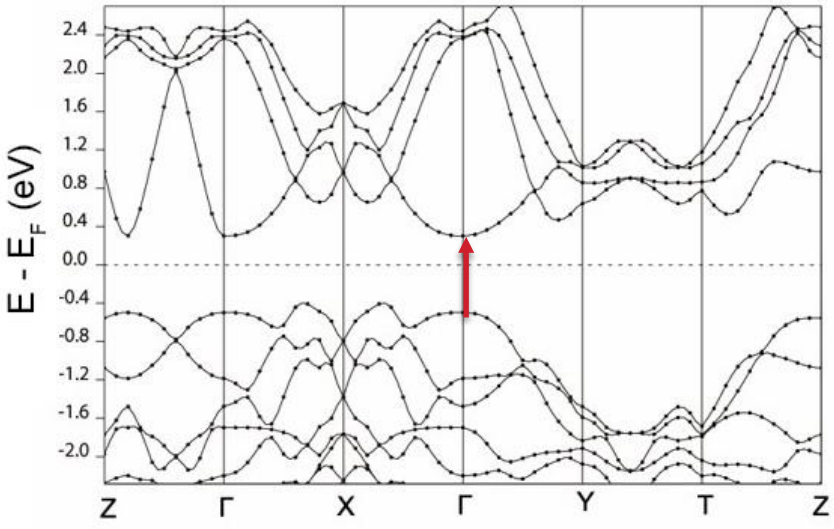
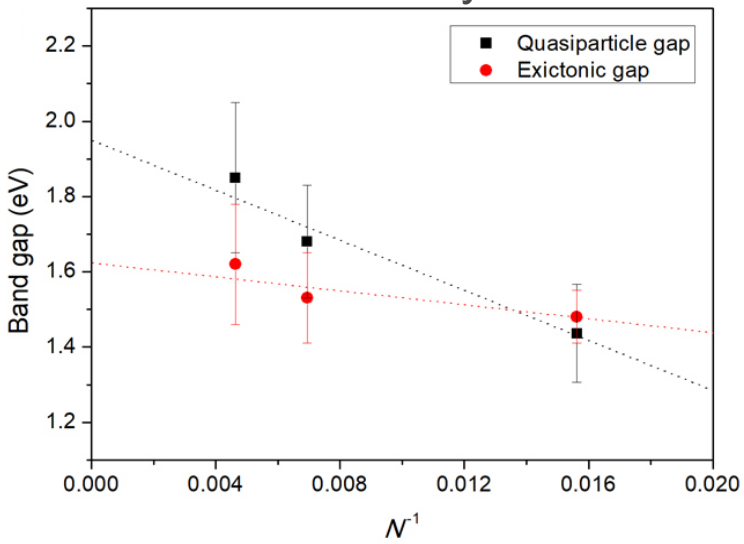

OPTICAL PROPERTIES FOR BULK GeSe

PBE band structure



Size analysis



	PBE	LDA	SCAN	PBE0	B3LYP	HSE06	DMC	Exp.
Gap (eV)	0.68	0.58	0.88	1.88	1.77	1.21	**1.62(16) : ex** **1.95(21) : qp**	**1.53**[1]

DMC excitonic gap is in the good agreement with the experimental results.

[1] E. P. O. O'Reailly, J. Phys. C Solid State Phys. 15, 1449 (1982).

H. Shin *et al.*, Phys. Rev. Materials 5, 024002 (2021).

SUMMARY

- **DMC can provide accurate structural and optical properties for 2D GeSe simultaneously.**

- As bulk GeSe cohesive energy is not experimentally reported yet, DMC cohesive energy (6.91(2) eV/f.u.) can give good guidance to assess the energetic stability of GeSe-based materials.

- The excitonic gap (1.62(16) eV) is good agreement with the experimental result of 1.53 eV.

- The quasiparticle gap (1.95(21) eV) is seemingly larger than the excitonic gap.

: one would expect the excitonic gap to be smaller by an amount equal to the exciton binding energy.

-> weak exciton binding energy of 0.33(26) eV

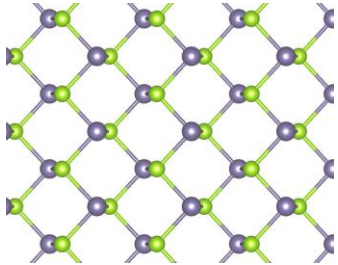
DFT AND QMC STUDY ON MONOLAYER GESE

MONOLAYER GESE

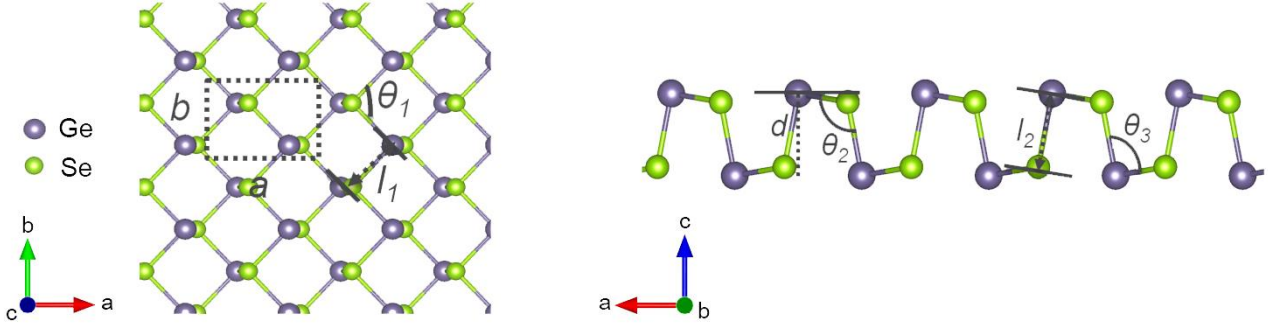
: the most attractive akali metal-ion anode materials
: better electrical conductivity than a black phosphorus and graphene (high P & T)
: expected to possess direct band gap via DFT studies
: gap can be tuned using uniaxial or biaxial strains
-> strain sensitivity clearly shows the importance of obtaining an accurate monolayer chemical structure.

Detailed geometry & optical properties for monolayer are not experimentally known.

: DFT-GGA shows huge variation of relaxed latter parameters
(a = 3.99 – 4.83 Å, b = 3.78 – 3.97 Å)
-> difficult to ascertain the monolayer chemical structure
-> difficult to assess the accuracy of reported DFT band gap (0.8 – 1.8 eV)



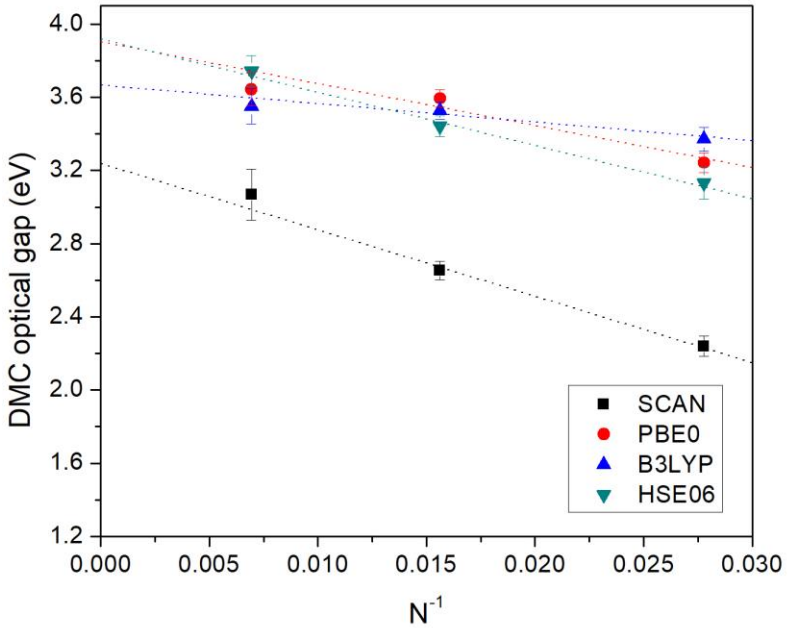
OPTIMIZED DFT GEOMETRY FOR MONOLAYER



d	a (Å)	b (Å)	d (Å)	l_1 (Å)	l_2 (Å)	θ_1 (deg)	θ_2 (deg)	θ_3 (deg)
PBE	4.26	3.95	2.50	2.65	2.52	96.5	93.9	97.2
SCAN	4.71	3.76	2.46	2.57	2.54	94.2	103.6	95.4
PBE0	4.21	3.85	2.48	2.58	2.51	96.5	100.3	91.8
B3LYP	4.47	3.89	2.51	2.56	2.51	96.9	98.5	96.7
HSE06	4.38	3.99	2.46	2.63	2.50	98.8	101.3	93.2

H. Shin *et al.*, Phys. Rev. Materials 5, 024002 (2021).

DMC OPTICAL GAP ON DFT GEOMETRIES

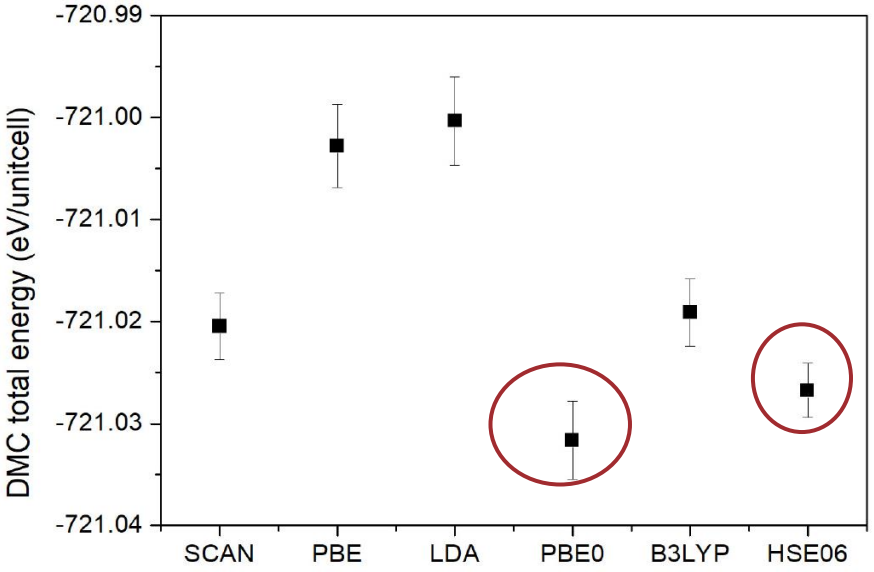


Geometry	SCAN	PBE0	B3LYP	HSE06
Direct gap (Γ) (eV)	3.24(11)	3.90(9)	3.67(10)	3.92(10)

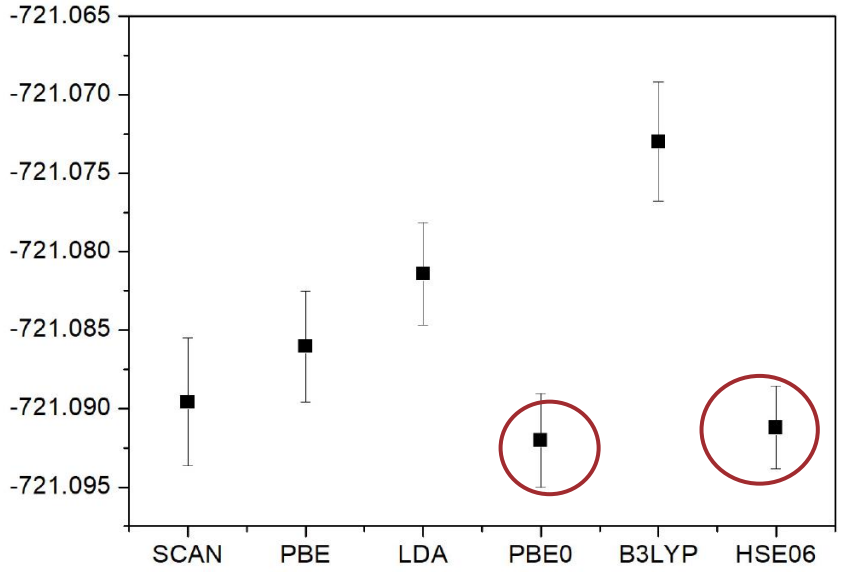
Accurate benchmark calculation of the monolayer GeSe is needed

DMC ON RELAXED DFT ATOMIC COORDINATES

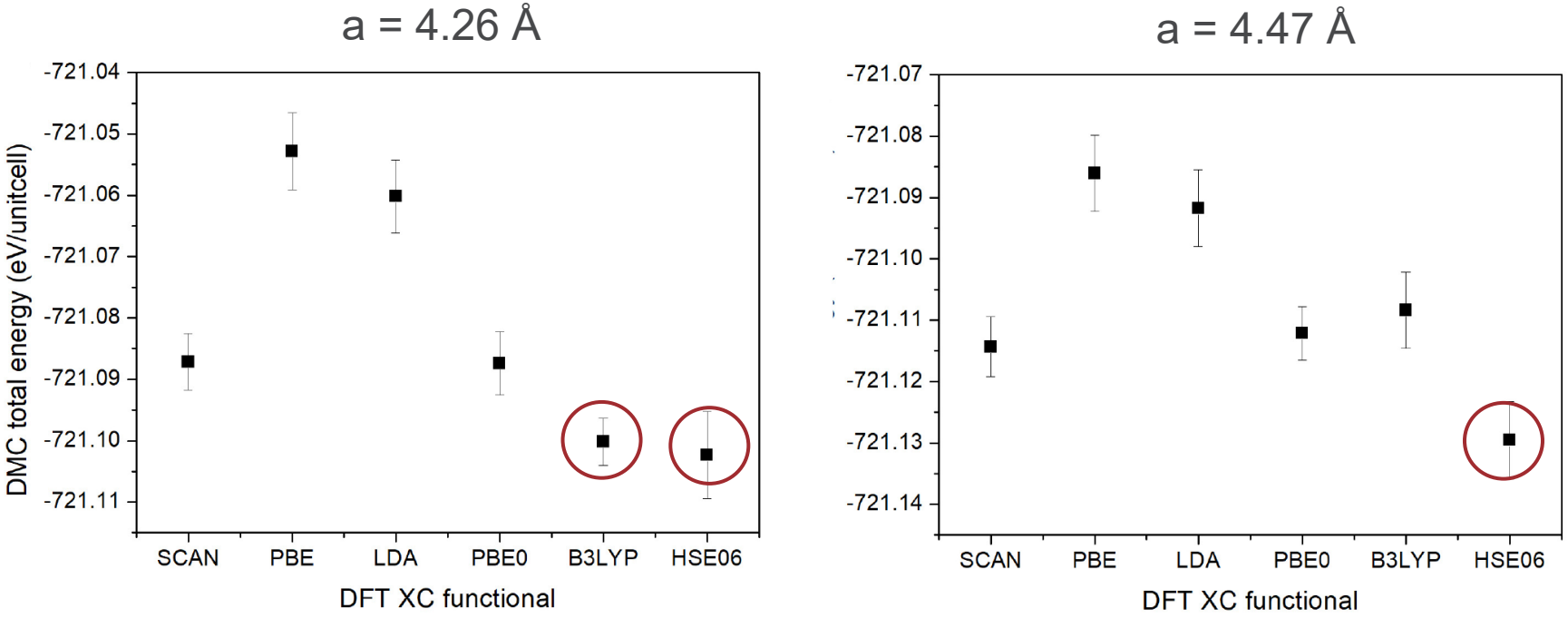
a = 4.05 Å



a = 4.15 Å



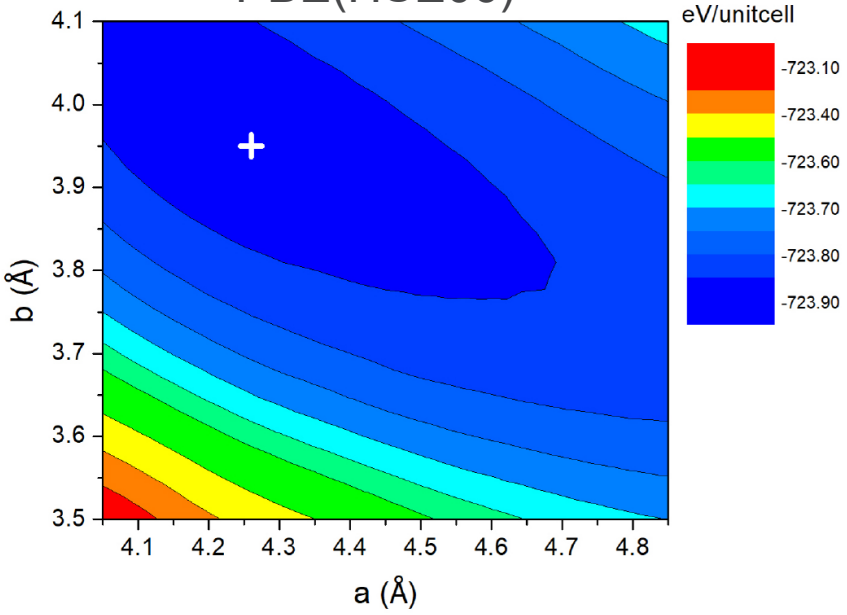
DMC ON RELAXED DFT GEOMETRIES



HSE06 can provide relatively stable GeSe atomic coordinates as compared to the other XC functionals

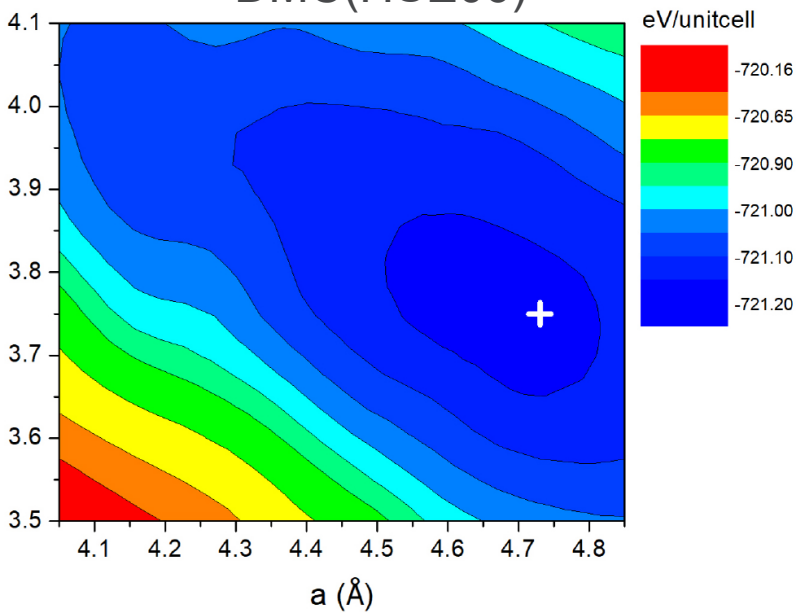
OPTIMIZED GEOMETRY WITH DMC(HSE06)

PBE(HSE06)



a = 4.26 Å, b = 3.95 Å

DMC(HSE06)



a = 4.73(1) Å, b = 3.76(1) Å

- Optimized DMC and DFT geometries are rather different.
  : atomic force evaluation is underestimated in DFT relaxation process.

GEOMETRY OPTIMIZATION

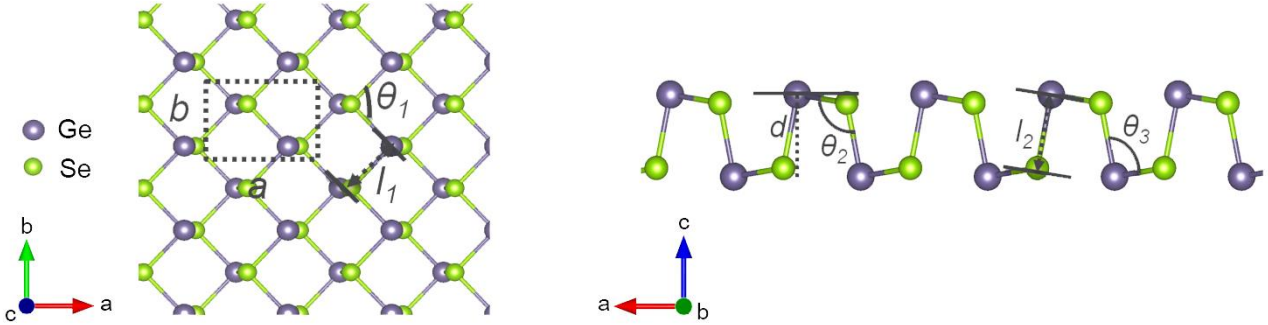
How to find chemical geometry for GeSe monolayer ?

Surrogate Hessian-based parallel line search method
- accelerate the search for the DMC potential energy surface optimum by incorporating approximate from the DFT-energy Hessian

Details are in session 7
“Surrogate Hessian Geometry optimization with STALK”
July 22

H. Shin *et al*., Phys. Rev. Materials 5, 024002 (2021).

GEOMETRY OPTIMIZATION



Total 5 parameters

Lattice parameters (a, b)

Ge (x, 0.25, z_1), Ge (x + 0.5, 0.75, 1 – z_1), Se (0.5, 0.25 , 1 - z_2), Se (0, 0.75, z_2)

$$l_1^2 = (xa)^2 + (0.5b)^2 + [(z_1 - z_2)c]^2$$

$$l_2^2 = [(x - 0.5)a]^2 + \{[(z_1 + z_2) - 1]c\}^2$$

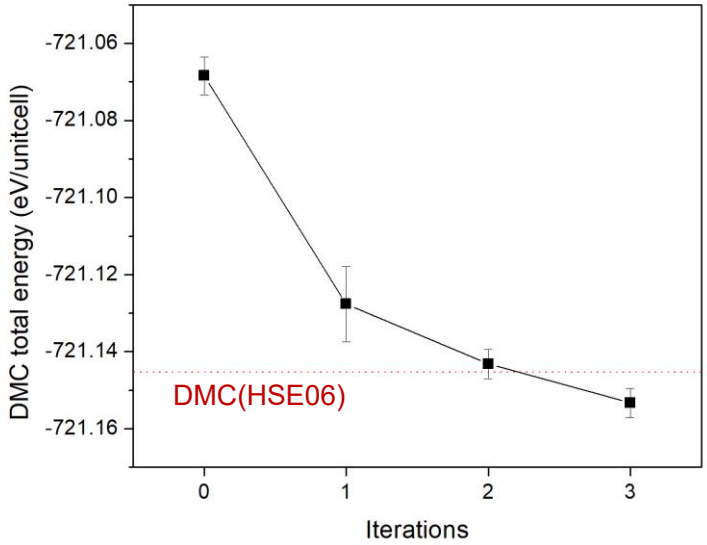
$$d = [(z_1 + z_2) - 1]c$$

$$\theta_1 = 2\tan^{-1}\left[\frac{0.5b}{\sqrt{x^2a^2 + (z_1 - z_2)^2c^2}}\right]$$

$$\theta_2 = \cos^{-1}\left[\frac{(2x-1)xa^2 + 2\left(z_1 - z_2 - z_1^2 + z_2^2\right)c^2}{2\sqrt{(0.5-x)^2a^2 + (1 - z_2 - z_1)^2c^2}\sqrt{x^2a^2 + 0.25b^2 + (z_1 - z_2)^2c^2}}\right]$$

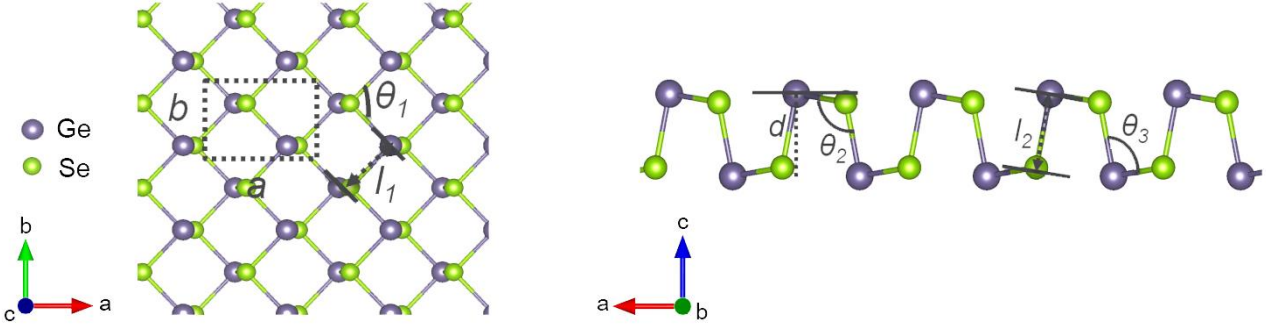
$$\theta_3 = \cos^{-1}\left[\frac{(2x-1)xa^2 + 2\left(z_2 - z_1 - z_2^2 + z_1^2\right)c^2}{2\sqrt{(0.5-x)^2a^2 + (1 - z_2 - z_1)^2c^2}\sqrt{x^2a^2 + 0.25b^2 + (z_1 - z_2)^2c^2}}\right],$$

GEOMETRY OPTIMIZATION



	E_{DMC} (eV/unitcell)	a (Å)	b (Å)	x	z_1	z_2
Step0	-721.0684(49)	4.26	3.95	0.4140	0.5560	0.5600
Step1	-721.1277(98)	4.27(2)	3.95(1)	0.402(6)	0.5603(3)	0.5536(4)
Step2	-721.1432(38)	4.40(1)	3.89(1)	0.399(3)	0.5607(3)	0.5528(4)
Step3	**-721.1533(38)**	**4.40(2)**	**3.89(1)**	**0.400(3)**	**0.5604(2)**	**0.5532(2)**

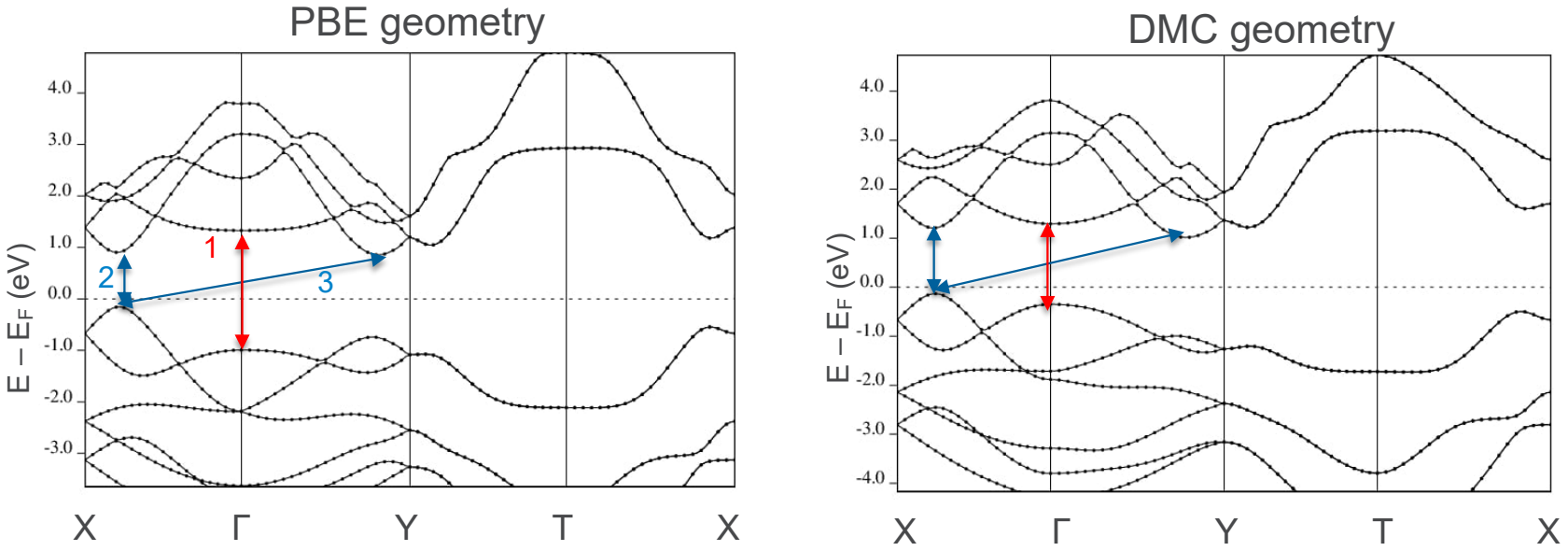
OPTIMIZED GEOMETRY FOR MONOLAYER



	a (Å)	b (Å)	d (Å)	l_1 (Å)	l_2 (Å)	θ_1 (deg)	θ_2 (deg)	θ_3 (deg)
PBE	4.26	3.95	2.50	2.65	2.52	96.5	93.9	97.2
SCAN	4.71	3.76	2.46	2.57	2.54	94.2	103.6	95.4
PBE0	4.21	3.85	2.48	2.58	2.51	96.5	100.3	91.8
B3LYP	4.47	3.89	2.51	2.56	2.51	96.9	98.5	96.7
HSE06	4.38	3.99	2.46	2.63	2.50	98.8	101.3	93.2
DMC(HSE06)	4.73(1)	3.76(1)	2.44	2.56	2.53	94.2	103.1	96.4
DMC	4.40(2)	3.89(1)	2.45(1)	2.63(1)	2.49(1)	95.4(5)	100.2(2)	93.4(2)
Exp. (Bulk)	4.40	3.85	2.49	2.58	2.54	95.4	103.6	90.8

H. Shin *et al.*, Phys. Rev. Materials 5, 024002 (2021).

PBE BAND STRCUTURE (DMC VS. PBE GEOMETRY)

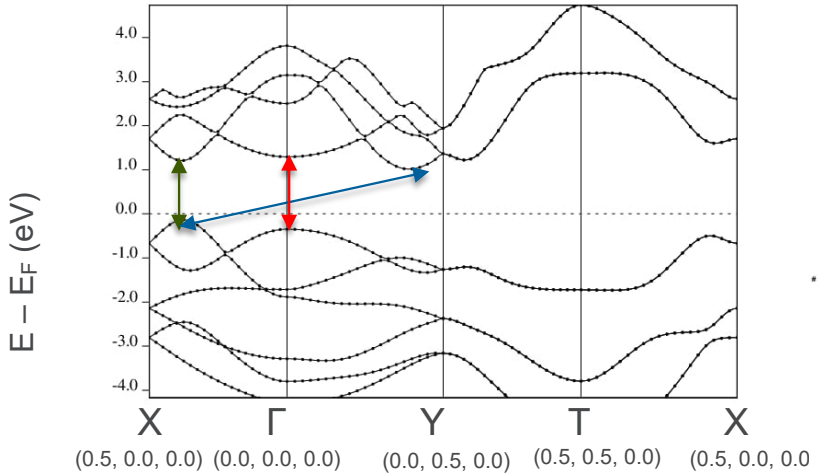


1. PBE Geometry
- PBE gives the indirect band gap (1.01 eV) but difference between indirect & direct gap (X-Γ) is very small. (0.05 eV)

2. DMC geometry
- Larger direct (1.34 eV) and indirect gap (1.16 eV) than PBE geometry
- **Additional direct gap (1.64 eV) is shown in DMC geometry (smaller than PBE geometry (2.32 eV))**
  : band structure is very sensitive to the precise structure.

H. Shin *et al.*, Phys. Rev. Materials 5, 024002 (2021).

EXCITED STATE CALCULATION (DIRECT & INDIRECT GAP)



2x2x1

#	Band	State	TwistIndex	BandIndex	Energy	Kx	Ky	Kz	K1	K2	K3	KmK
	0	0	0	0	-0.591710	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	1	1	2	0	-0.556408	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	2	2	2	1	-0.556408	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	3	3	1	0	-0.543909	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	4	4	1	1	-0.543909	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	5	5	3	0	-0.536346	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	6	6	3	1	-0.536346	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	7	7	0	1	-0.533014	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	8	8	1	2	-0.365203	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	9	9	1	3	-0.365203	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	10	10	2	2	-0.361029	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	11	11	2	3	-0.361029	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	12	12	0	2	-0.349276	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	13	13	3	2	-0.349030	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	14	14	3	3	-0.349030	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	15	15	0	3	-0.266109	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	16	16	3	4	-0.247844	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	17	17	3	5	-0.247844	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	18	18	0	4	-0.224857	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	19	19	0	5	-0.206730	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	20	20	3	6	-0.206525	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	21	21	3	7	-0.206525	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	22	22	0	6	-0.188185	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	23	23	1	4	-0.183413	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	24	24	1	5	-0.183413	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	25	25	2	4	-0.170398	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	26	26	2	5	-0.170398	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	27	27	1	6	-0.154558	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	28	28	1	7	-0.154558	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	29	29	2	6	-0.146125	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	30	30	2	7	-0.146125	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	31	31	0	7	-0.136993	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	32	32	3	8	-0.131258	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	33	33	3	9	-0.131258	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	34	34	0	8	-0.130929	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	35	35	1	8	-0.114123	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	36	36	1	9	-0.114123	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	37	37	2	8	-0.092243	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	38	38	2	9	-0.092243	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	39	39	0	9	-0.081440	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	40	40	0	10	-0.019258	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	41	41	1	10	-0.015895	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	42	42	1	11	-0.015895	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	43	43	2	10	-0.004087	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	44	44	2	11	-0.004087	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	45	45	1	12	0.004332	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	46	46	1	13	0.004332	0.0000	0.4274	0.0000	0.0000	0.5000	0.0000	1
	47	47	0	11	0.027011	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	48	48	2	12	0.028131	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	49	49	2	13	0.028131	0.3776	0.0000	0.0000	0.5000	0.0000	0.0000	1
	50	50	0	12	0.047689	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	51	51	3	10	0.050356	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	52	52	3	11	0.050356	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	53	53	0	13	0.070761	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	54	54	3	12	0.104637	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1
	55	55	3	13	0.104637	0.3776	0.4274	0.0000	0.5000	0.5000	0.0000	1

3x3x1

#	Band	State	TwistIndex	BandIndex	Energy	Kx	Ky	Kz	K1	K2	K3	KmK
	0	0	0	0	-0.591791	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	1	1	2	0	-0.573989	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	2	3	6	0	-0.566789	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	3	5	7	0	-0.553562	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	4	7	8	0	-0.553562	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	5	9	2	1	-0.541536	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	6	11	7	1	-0.533932	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	7	13	8	1	-0.533932	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	8	15	0	1	-0.533047	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	9	16	6	1	-0.530447	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	10	18	6	2	-0.373743	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	11	20	2	2	-0.370439	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	12	22	7	2	-0.361365	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	13	24	8	2	-0.361365	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	14	26	0	2	-0.349437	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	15	27	7	3	-0.337416	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	16	29	8	3	-0.337416	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	17	31	2	3	-0.331522	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	18	33	6	3	-0.328945	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	19	35	0	3	-0.266207	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	20	36	7	4	-0.251081	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	21	38	8	4	-0.251081	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	22	40	0	4	-0.224926	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	23	41	7	5	-0.211308	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	24	43	8	5	-0.211308	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	25	45	0	5	-0.206821	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	26	46	6	4	-0.201526	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	27	48	2	4	-0.195796	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	28	50	7	6	-0.194911	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	29	52	8	6	-0.194911	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	30	54	6	5	-0.194316	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	31	56	0	6	-0.188273	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	32	57	6	6	-0.171951	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	33	59	2	5	-0.166667	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	34	61	2	6	-0.159677	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	35	63	7	7	-0.157697	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	36	65	8	7	-0.157697	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	37	67	6	7	-0.143331	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	38	69	7	8	-0.142048	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	39	71	8	8	-0.142048	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	40	73	0	7	-0.137047	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	41	74	2	7	-0.132186	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	42	76	0	8	-0.130984	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	43	77	2	8	-0.114403	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	44	79	7	9	-0.108267	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	45	81	8	9	-0.108267	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	46	83	6	8	-0.108017	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	47	85	6	9	-0.105411	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	48	87	0	9	-0.081480	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	49	88	2	9	-0.076930	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	50	90	6	10	-0.024286	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	51	92	0	10	-0.019337	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	52	93	2	10	-0.017243	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	53	95	6	11	0.007886	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	54	97	7	10	0.010101	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	55	99	8	10	0.010101	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	56	101	2	11	0.010513	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	57	103	6	12	0.011201	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	58	105	2	12	0.024216	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	59	107	0	11	0.026886	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	60	108	6	13	0.030051	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	2
	61	110	2	13	0.033761	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	2
	62	112	7	11	0.043316	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	63	114	8	11	0.043316	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	64	116	0	12	0.047558	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	65	117	7	12	0.069981	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	66	119	8	12	0.069981	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2
	67	121	0	13	0.070624	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	68	122	7	13	0.083256	0.2517	0.5699	0.0000	0.3333	0.6667	0.0000	2
	69	124	8	13	0.083256	0.5035	0.5699	0.0000	0.6667	0.6667	0.0000	2

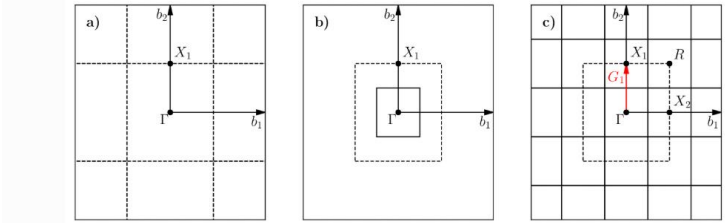


Fig. 27 *a) First BZ of the primitive cell centered on* Γ*. Dashed lines indicate zone boundaries. b) First BZ of the* 2×2 *supercell inside the first BZ of the primitive cell. First BZ boundaries of the supercell are shown using solid lines. c) Periodic translations of the first BZ of the supercell showing that* Γ *and* X_1 *are periodic images of each other given the supercell BZ.*

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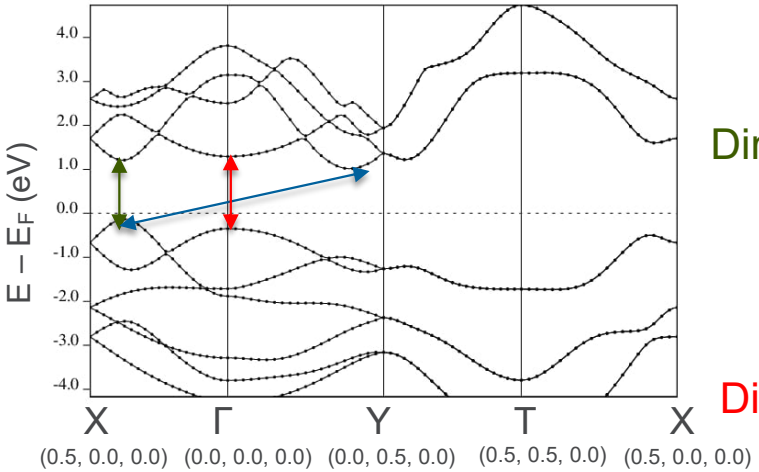
EXCITED STATE CALCULATION (DIRECT & INDIRECT GAP)



Direct gap

#	Band	State	TwistIndex	BandIndex	Energy	Kx	Ky	Kz	K1	K2	K3	KmK
	86	86	6	9	-0.105411	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1
	87	87	0	9	-0.081480	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	88	88	1	9	-0.076930	0.2517	0.0000	0.0000	0.3333	0.0000	0.0000	1
	89	89	2	9	-0.076930	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	1
	90	90	3	10	-0.024286	0.0000	0.2850	0.0000	0.0000	0.3333	0.0000	1
	91	91	6	10	-0.024286	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1
	92	92	0	10	-0.019337	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	93	93	1	10	-0.017243	0.2517	0.0000	0.0000	0.3333	0.0000	0.0000	1
	94	94	2	10	-0.017243	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	1
	95	95	3	11	0.007886	0.0000	0.2850	0.0000	0.0000	0.3333	0.0000	1
	96	96	6	11	0.007886	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1

Direct gap

#	Band	State	TwistIndex	BandIndex	Energy	Kx	Ky	Kz	K1	K2	K3	KmK
	86	86	6	9	-0.105411	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1
	87	87	0	9	-0.081480	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	88	88	1	9	-0.076930	0.2517	0.0000	0.0000	0.3333	0.0000	0.0000	1
	89	89	2	9	-0.076930	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	1
	90	90	3	10	-0.024286	0.0000	0.2850	0.0000	0.0000	0.3333	0.0000	1
	91	91	6	10	-0.024286	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1
	92	92	0	10	-0.019337	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	93	93	1	10	-0.017243	0.2517	0.0000	0.0000	0.3333	0.0000	0.0000	1
	94	94	2	10	-0.017243	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	1
	95	95	3	11	0.007886	0.0000	0.2850	0.0000	0.0000	0.3333	0.0000	1
	96	96	6	11	0.007886	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1

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  </determinant>
</slaterdeterminant>
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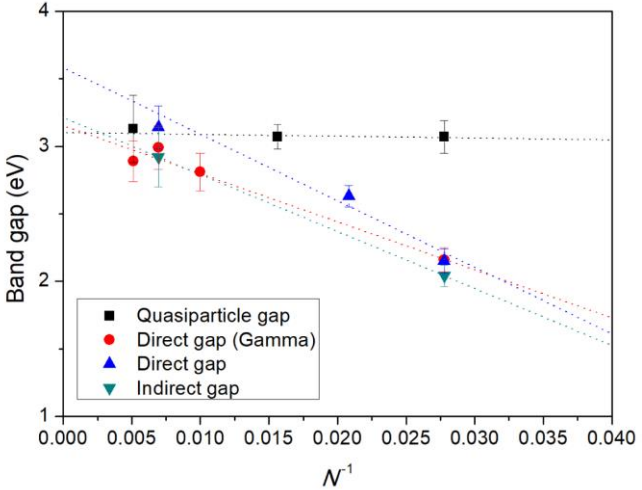
Indirect gap

#	Band	State	TwistIndex	BandIndex	Energy	Kx	Ky	Kz	K1	K2	K3	KmK
	86	86	6	9	-0.105411	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1
	87	87	0	9	-0.081480	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	88	88	1	9	-0.076930	0.2517	0.0000	0.0000	0.3333	0.0000	0.0000	1
	89	89	2	9	-0.076930	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	1
	90	90	3	10	-0.024286	0.0000	0.2850	0.0000	0.0000	0.3333	0.0000	1
	91	91	6	10	-0.024286	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1
	92	92	0	10	-0.019337	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	1
	93	93	1	10	-0.017243	0.2517	0.0000	0.0000	0.3333	0.0000	0.0000	1
	94	94	2	10	-0.017243	0.5035	0.0000	0.0000	0.6667	0.0000	0.0000	1
	95	95	3	11	0.007886	0.0000	0.2850	0.0000	0.0000	0.3333	0.0000	1
	96	96	6	11	0.007886	0.0000	0.5699	0.0000	0.0000	0.6667	0.0000	1

H. Shin *et al.*, Phys. Rev. Materials 5, 024002 (2021).

OPTICAL PROPERTIES FOR MONOLAYER (DMC OPTIMIZED GEOMETRY)

Size analysis



	Direct (Γ) gap (eV)	Direct gap (X-Γ)	Indirect gap	Quasiparticle gap
LDA	1.50	1.24	1.02	-
PBE	1.64	1.34	1.16	-
SCAN	1.90	1.61	1.38	-
PBE0	2.97	2.45	2.27	-
B3LYP	2.71	2.36	2.21	-
HSE06	2.29	1.87	1.67	-
DMC (bulk)	1.62(16)	-	-	1.95(21)
DMC (monolayer)	3.2(1)	3.6(2)	3.2(2)	3.1(2)

H. Shin *et al*., Phys. Rev. Materials 5, 024002 (2021).

1. Monolayer has substantially larger band gaps (3.2 ~ 3.6 eV) than bulk GeSe (1.6 ~ 2 eV).
   : Monolayer GeSe is a highly tunable wide band-gap semiconductor using strain.
2. Very small difference between direct & indirect gap
   : not easy to confirm between direct & indirect gap for monolayer GeSe.
3. Large difference on DFT band structure & band gap in different geometries
   : The strain sensitivity of the electronic structure
   -> monolayer GeSe has a tunable direct-to-indirect gap transition.

CONCLUSIONS

- GeSe monolayer exhibits optical characteristic of a wide-gap semiconductor with tunable band gap.

- Very small difference of direct & indirect gap and the sensitivity of electronic structure to the geometry tells that transition can be induced from a direct gap to an indirect gap semiconductor.

- Very shallow minimum in potential energy surface and coupling of electronic structure to strain makes for difficult problem with DFT – may be more general to 2D materials than just for GeSe (Wines, Saritas and Ataca on GaSe J Chem Phys 2020).

- Simple examples of GeSe work (gap calculations) can be found in the repo.

CPSFM | Center for Predictive Simulation of Functional Materials

This work was supported by
the Center for Predictive Simulation of Functional Materials,
a DOE funded Computational Sciences Center,
where in addition to this research we develop and make available
the open source QMCPACK code (https://qmcpack.org/) &
ccECPs in the pseudopotential library
(https://pseudopotentiallibrary.org/).

Supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division, as part of the Computational Materials Sciences Program.