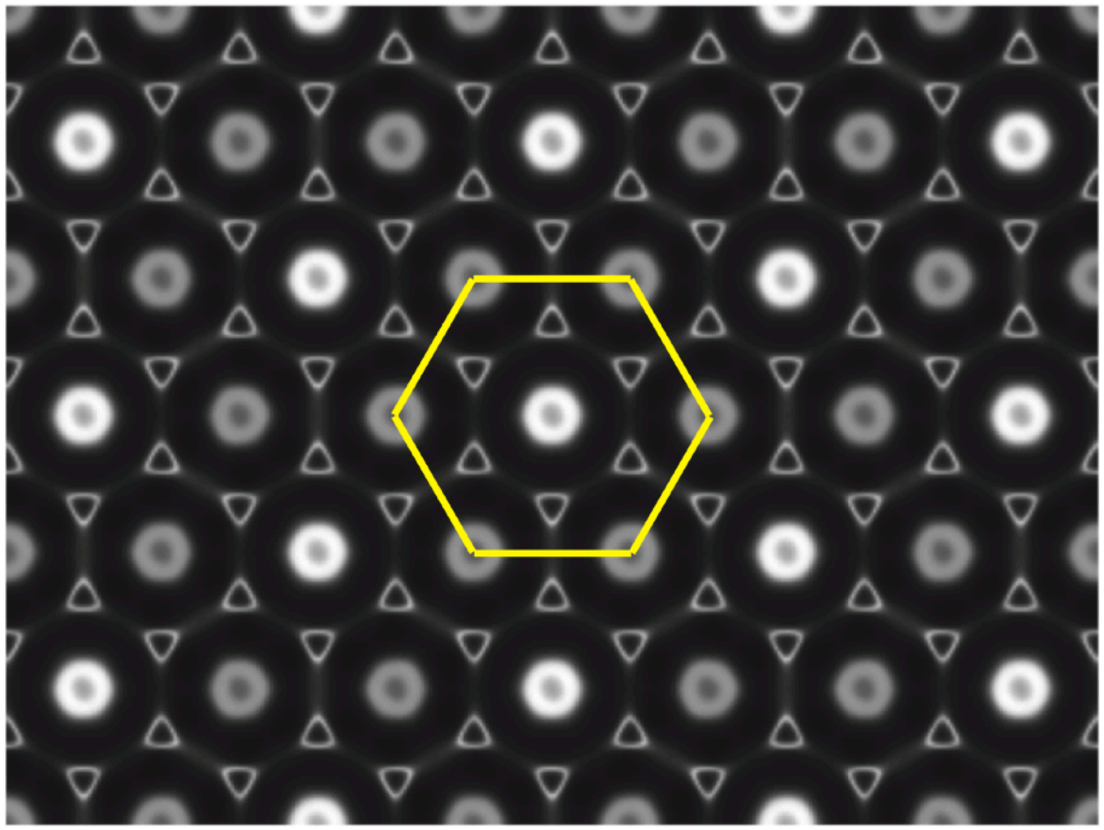
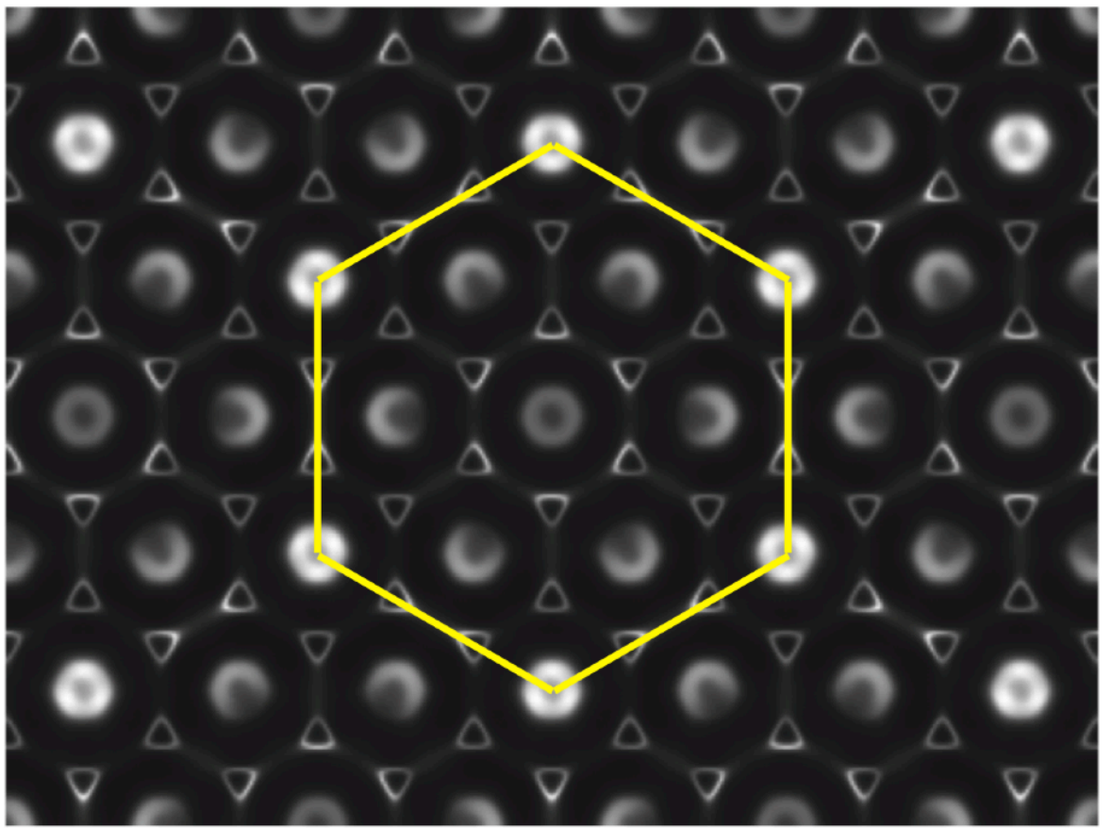
Unfolding band structure into a conceptual Brillouin zone





Chi-Cheng Lee

Institute for Solid State Physics, The University of Tokyo, Japan

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cclee.physics@gmail.com

What is band structure?

What is conceptual Brillouin zone?

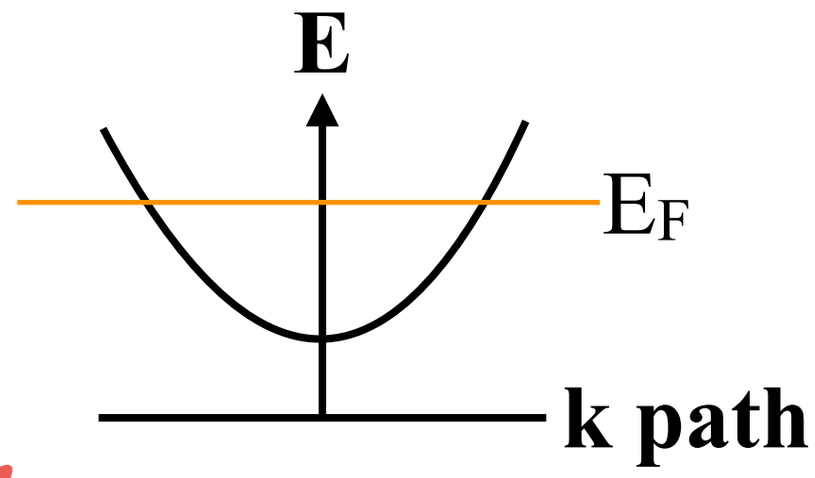
What is unfolding?

Band structure: Bloch theory and Kohn-Sham Hamiltonian

$$\hat{H}\psi=\left(-\frac{\hbar^2}{2m}\nabla^2+U(r)\right)\psi=\varepsilon\psi$$

$$U(r+R)=U(r)$$

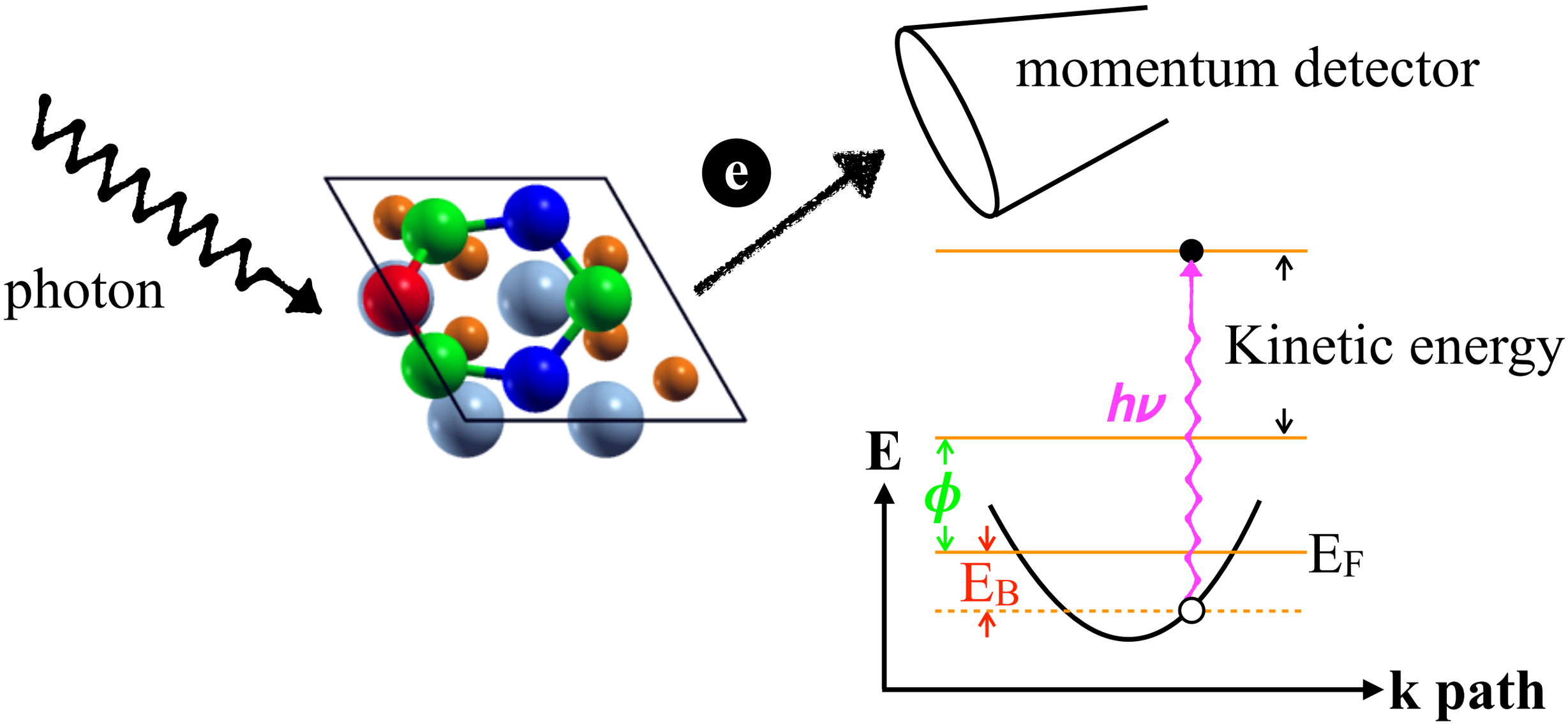
$$\psi_{nk}(r)=e^{ikr}u_{nk}(r) \quad with \quad eigenvalue \ \varepsilon_{nk}$$



This is called
Band structure

- The eigenstates of single-particle Hamiltonian can be compared with experiments that measure single-particle properties, for example, the ARPES measurement.
- Note that Kohn-Sham Hamiltonian is a single-particle Hamiltonian although the delivered charge density is many-body charge density.

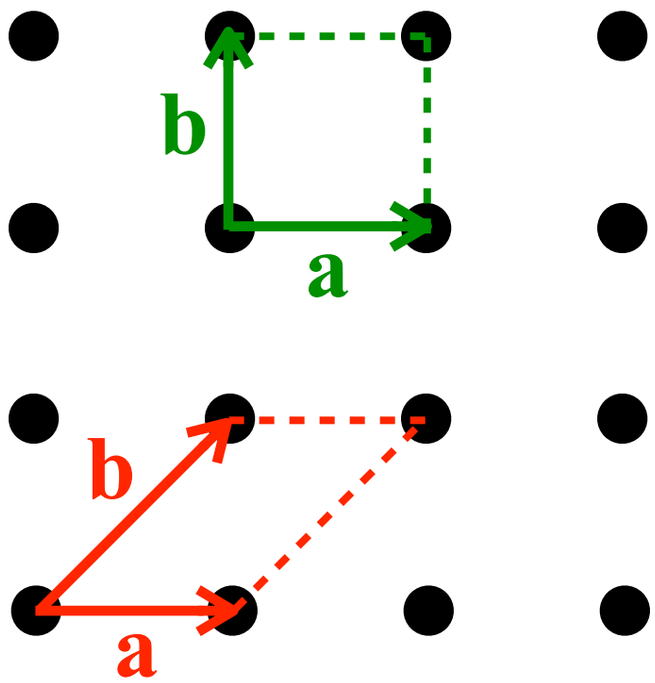
Band structure: Angle-Resolved Photoelectron Spectroscopy



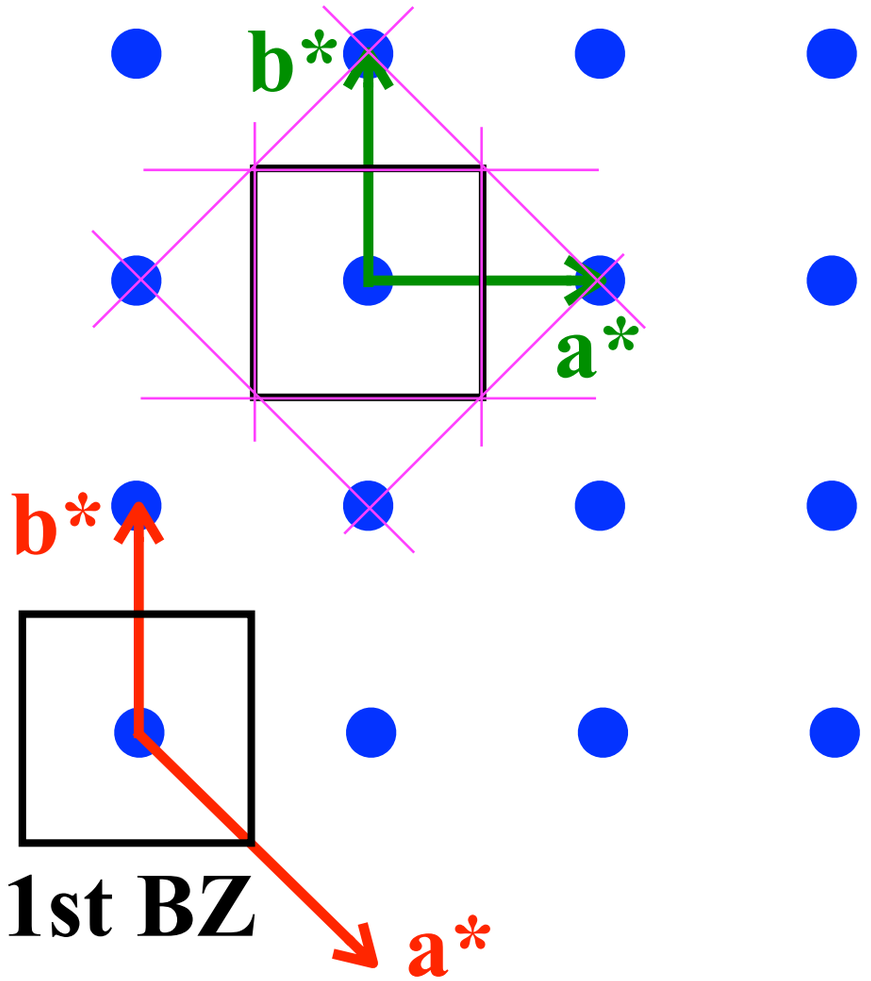
- ARPES experiment can directly measure the kinetic energy of the outgoing electron, and therefore, the momentum (the angle is known). Having the work function ϕ, the relationship between binding energies and momenta of the electrons can be plotted as the **band structure**.

First Brillouin zone: Primitive unit cell in reciprocal space

Real space



Reciprocal space



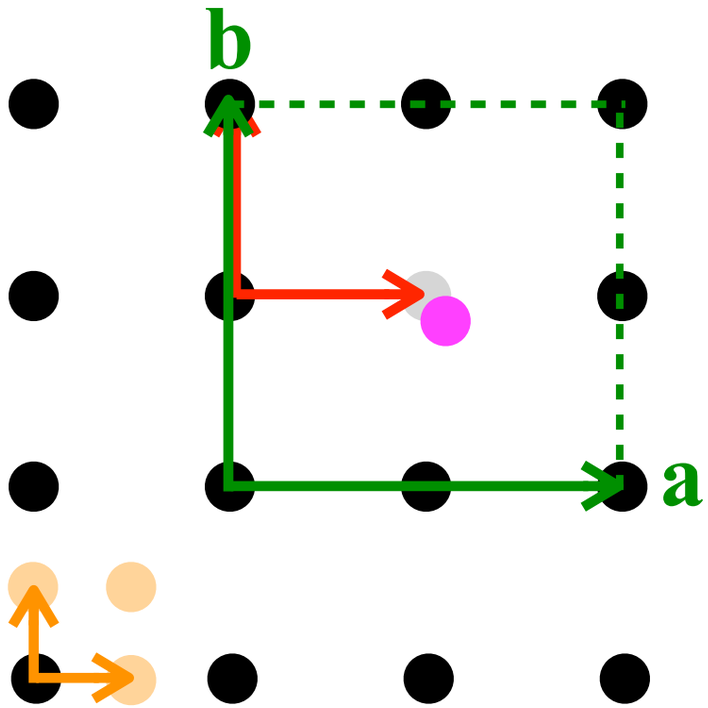
- Once the real-space primitive unit cell is determined, the reciprocal lattice vectors are also determined via

$$\vec{a}^* = 2\pi \frac{\vec{b}\times\vec{c}}{\vec{a}\cdot\vec{b}\times\vec{c}}, \quad \vec{b}^* = 2\pi \frac{\vec{c}\times\vec{a}}{\vec{b}\cdot\vec{c}\times\vec{a}}, \quad \vec{c}^* = 2\pi \frac{\vec{a}\times\vec{b}}{\vec{c}\cdot\vec{a}\times\vec{b}}$$

- Once the reciprocal lattice vectors are known, first Brillouin zone can be obtained using perpendicular bisectors.

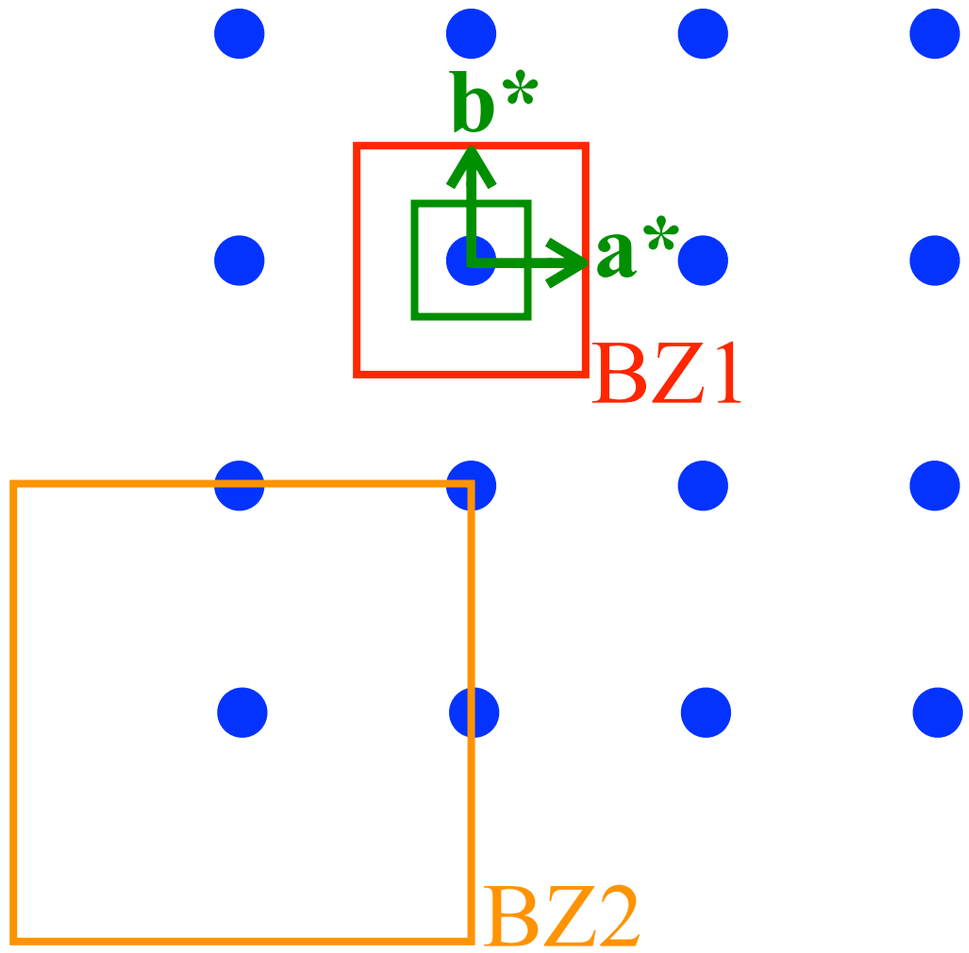
Conceptual Brillouin zone: BZ not restricted by geometry

Real space



many ***commensurate*** *unit cells*

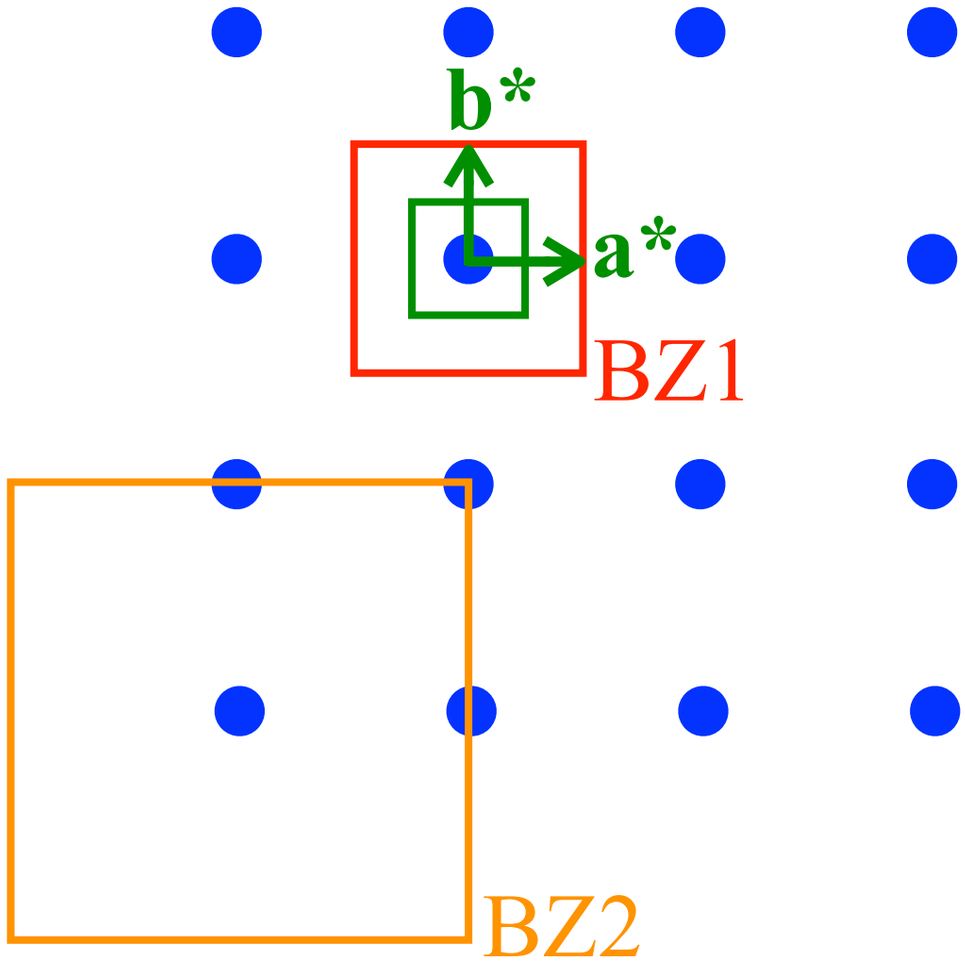
Reciprocal space



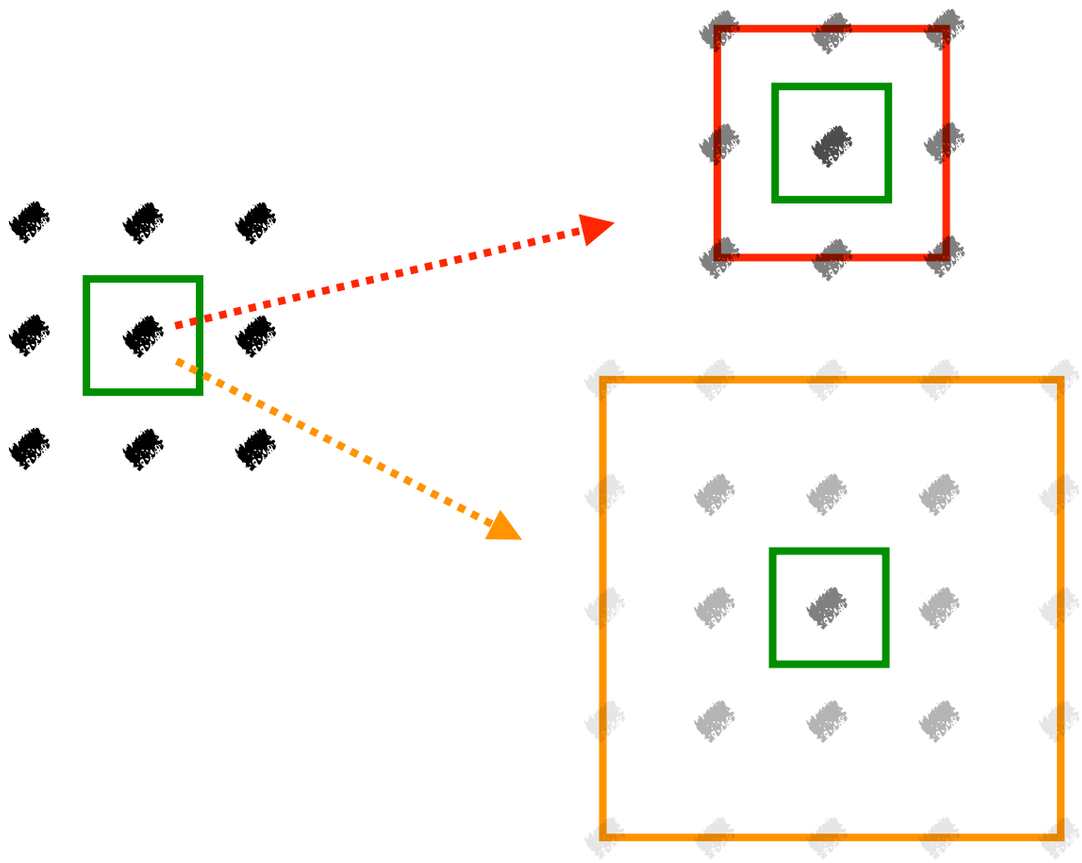
- For the choice of BZ1, the supercell is needed due to the dislocated atom at the center (see the plot in real space). The conceptual BZ is chosen as the the original BZ without considering the dislocation.
- For the choice of BZ2, a smaller unit cell (smaller than the primitive one) is chosen as the conceptual unit cell. The corresponding BZ is called the conceptual Brillouin zone.

Unfolding bands: Redistribute the weight from small to big BZs

Reciprocal space



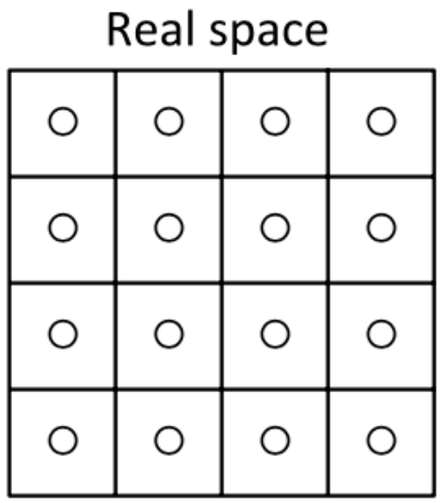
Constant energy contour

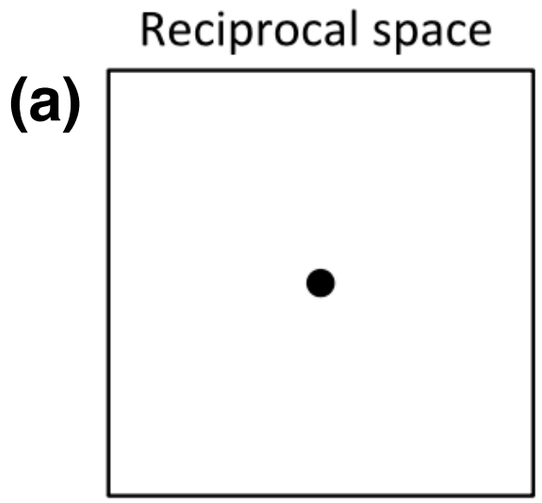


- Unfolding band structure can be considered as the calculation of new weight of each “supercell” eigenstate in the conceptual BZ.

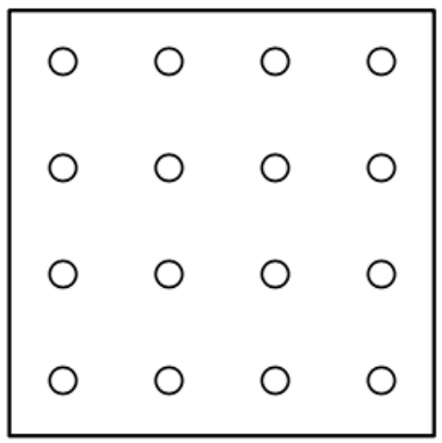
Why do we want to unfold bands?

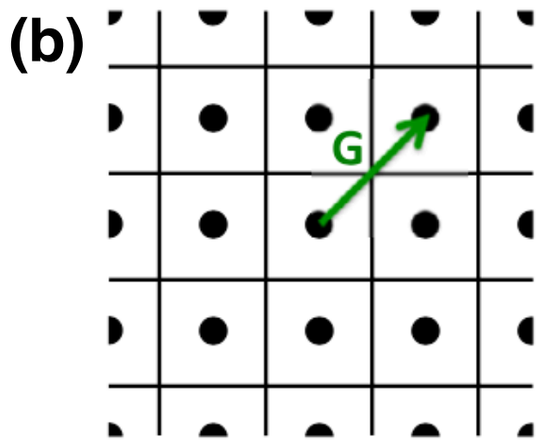
Compare with ARPES experiments: Elaboration 1



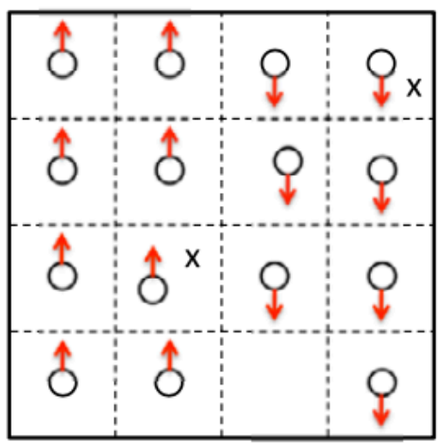


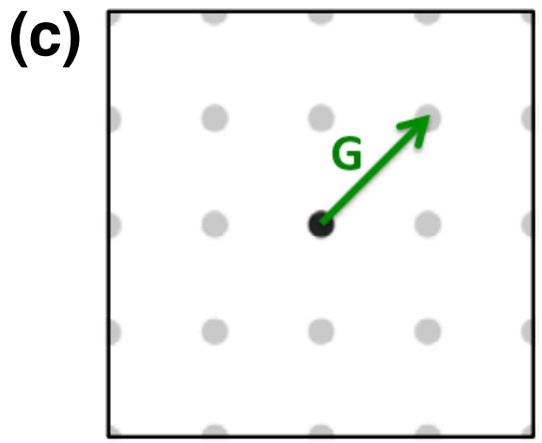
Assume the Fermi point (constant energy contour) in the BZ of primitive unit cell is measured by ARPES experiments in the first place.





In theoretical calculation, we can perform the calculation for the same *system* using a large supercell. The supercell BZ is much smaller than the one shown in (a). Note that the weight is periodic and is the same by shifting a **G** vector.





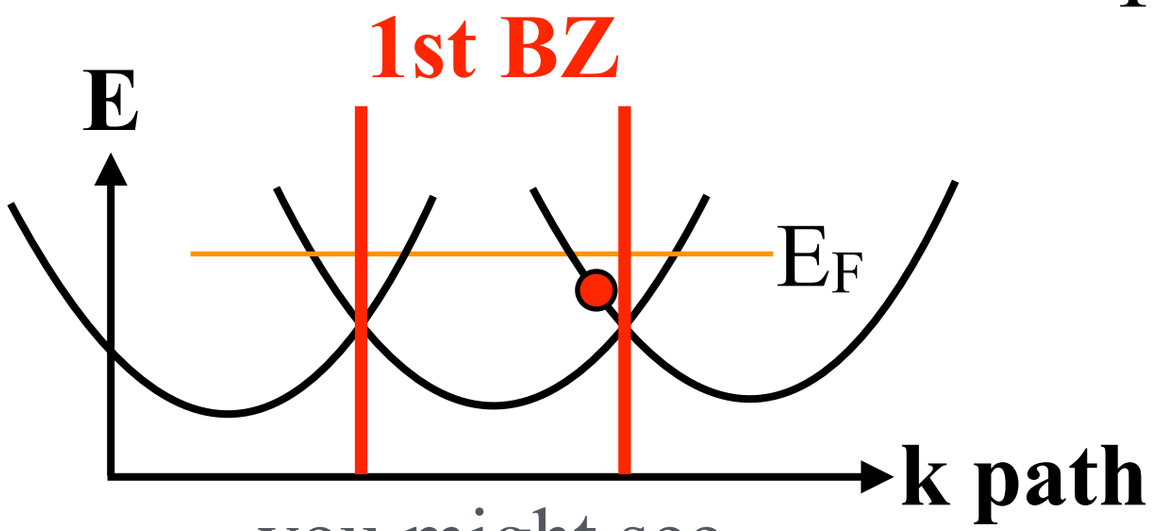
In the case the translational symmetry is broken, we must adopt a supercell for the calculation. We can ask a question: is the measured spectral weight similar to (a) or (b)? The answer is in (c). Reason: Measured intensity cannot experience a drastic change via a tiny perturbation. So we want to represent the weight in the BZ shown in (a).

Chi-Cheng Lee *et al.*, J. Phys.: Condens. Matter **25**, 345501 (2013).

Compare with ARPES experiments: Elaboration 2

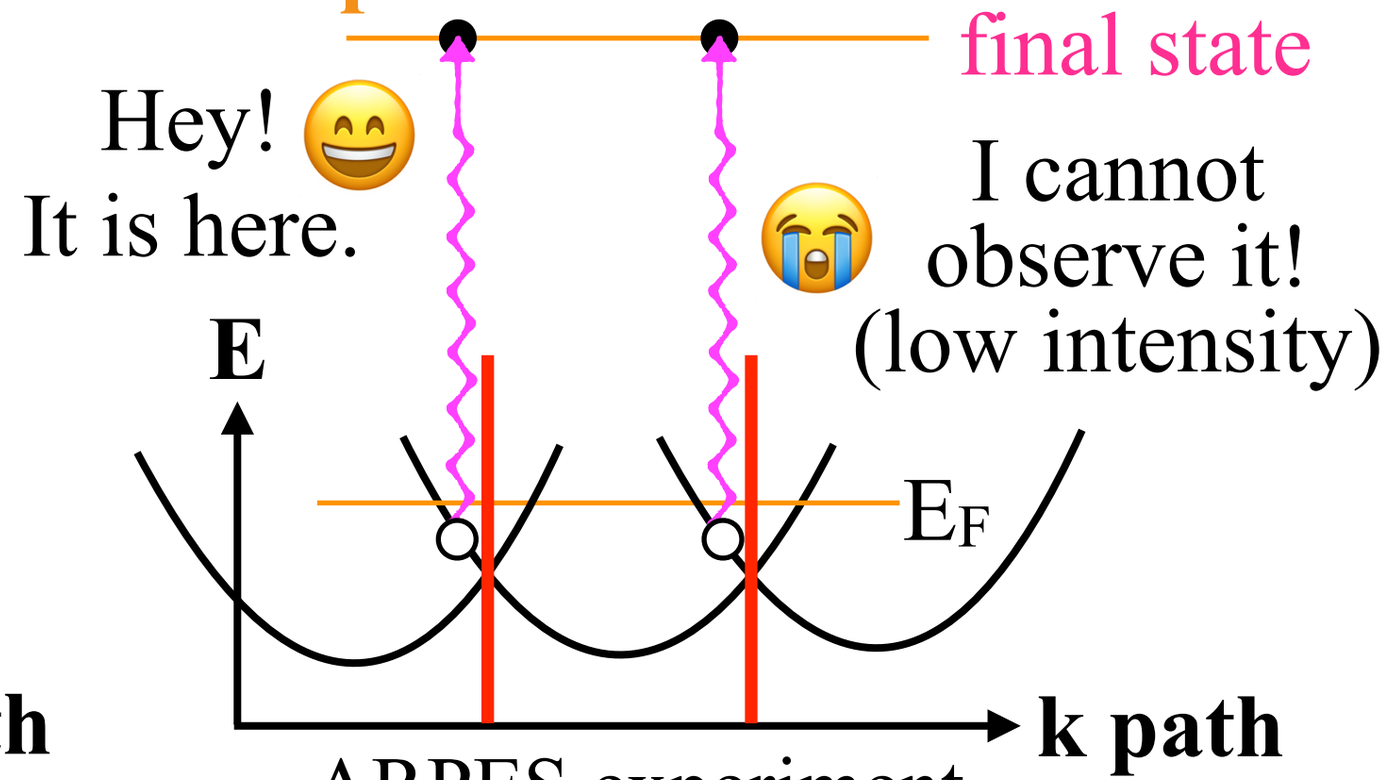
Periodic-zone representation

periodic-zone scheme allows us to discuss everything in 1st BZ



you might see larger gap-opening at zone boundary

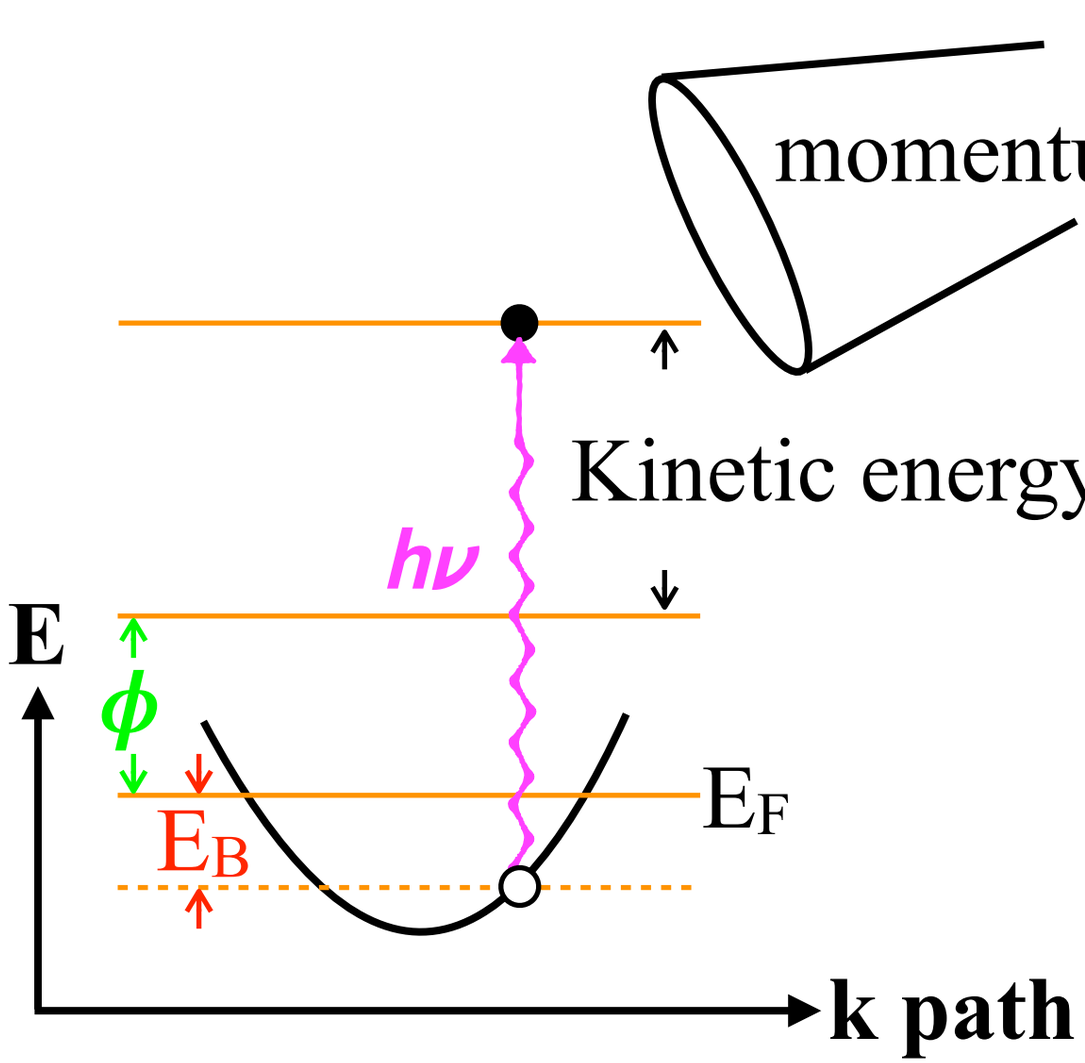
Experiments



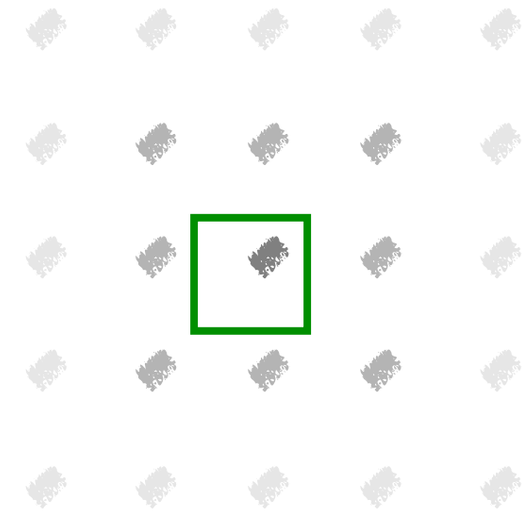
ARPES experiment prefers the extended-zone representation

- In DFT calculations, there is no difference between the eigenstates inside and outside the BZ as long as they differ by a **G** vector since we have periodicity. However, ARPES cannot observe all the states.
- Which state can be observed can be analyzed by carefully considering the matrix elements between the relevant initial and finial states.

Compare with ARPES experiments: Momentum distribution



Individual Fourier component



$$|\vec{k}n\rangle = \sum_{\vec{G}} C^{n}_{\vec{k}+\vec{G}} \, |\vec{k}+\vec{G}\rangle$$

plane wave coefficient gives us momentum distribution

$$|\vec{k}n\rangle = |\vec{k}+\vec{G}\, n\rangle$$

quantum number

Note that

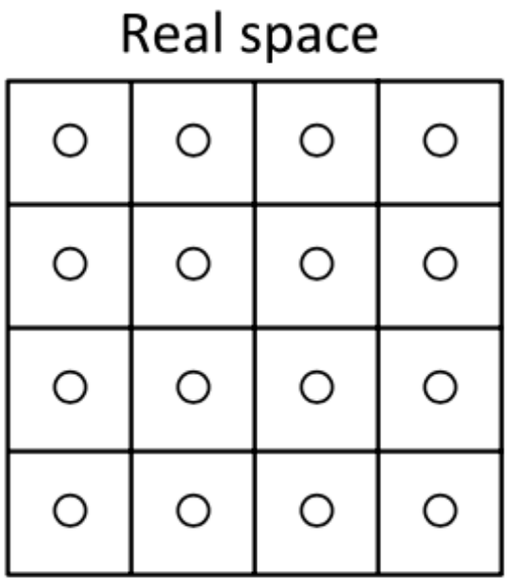
$$\langle \vec{k}n | \hat{\vec{p}} | \vec{k}n \rangle = \frac{m}{\hbar} \frac{\partial \epsilon_{\vec{k}n}}{\partial \vec{k}}$$

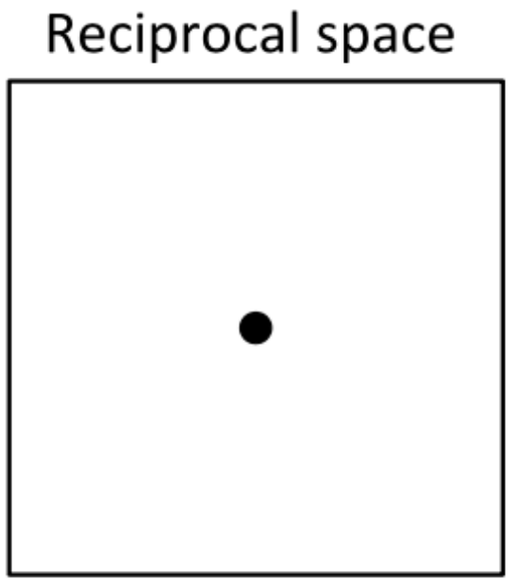
$$= \sum_{\vec{G}} \hbar(\vec{k}+\vec{G}) \left| C^{n}_{\vec{k}+\vec{G}} \right|^2$$

$$I(\vec{p}, |\vec{k}n\rangle) \approx |\vec{A}_{\vec{q}\to 0} \cdot \hat{\vec{p}}|^2 \, |\langle \vec{p} | \vec{k}n \rangle|^2$$

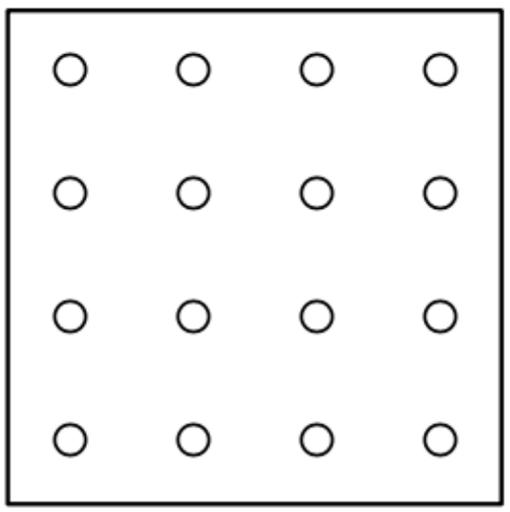
Chi-Cheng Lee *et al.*, arXiv: 1707.02525 (2018), JPCM in press.

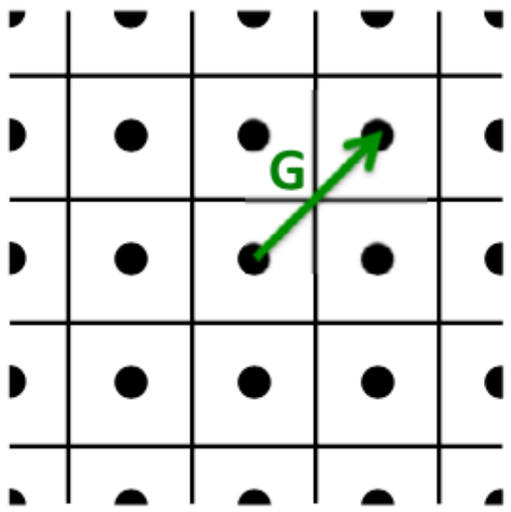
Theoretical interest: Degree of symmetry breaking



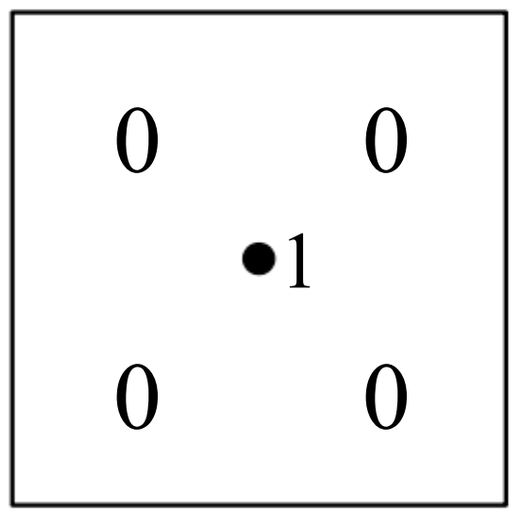


the same



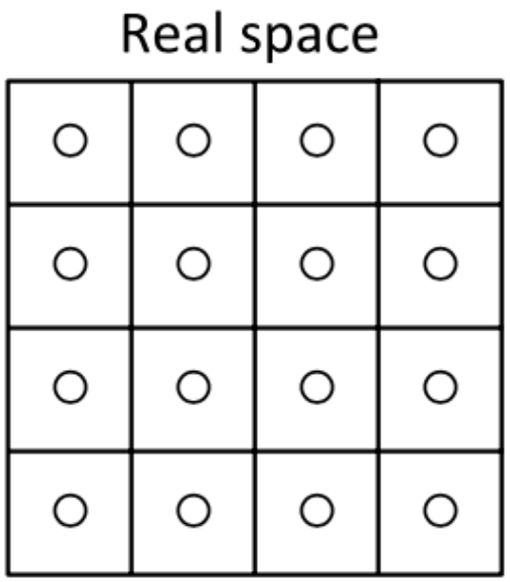


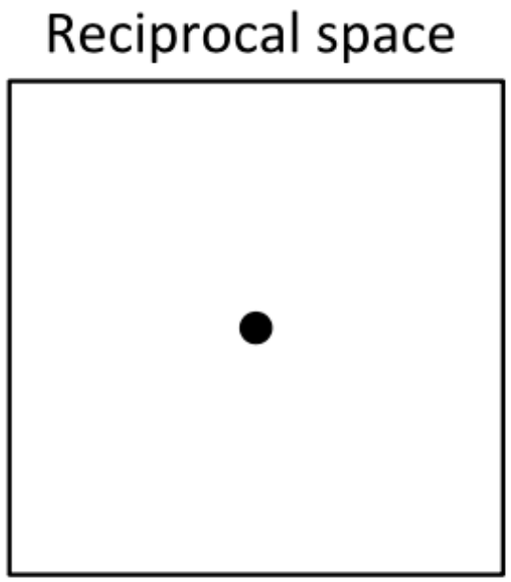
unfold



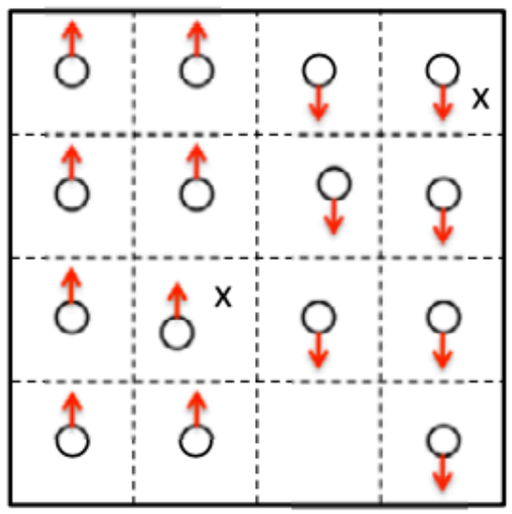
(no symmetry breaking)

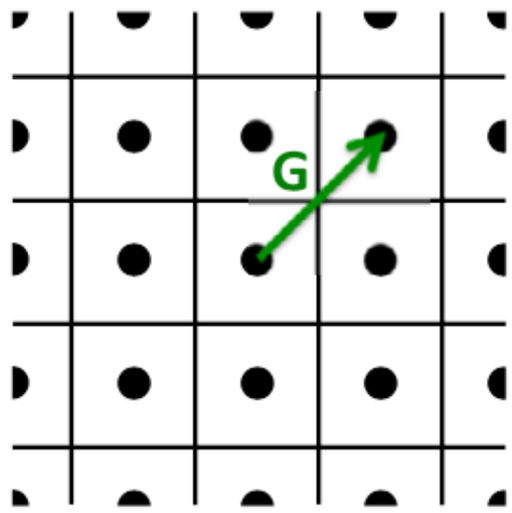
Theoretical interest: Degree of symmetry breaking



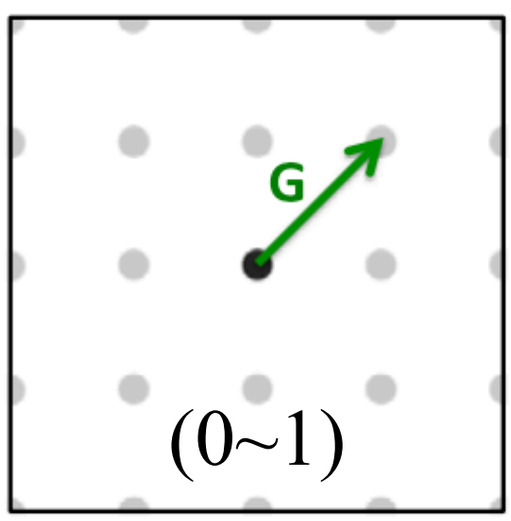


not the same





unfold



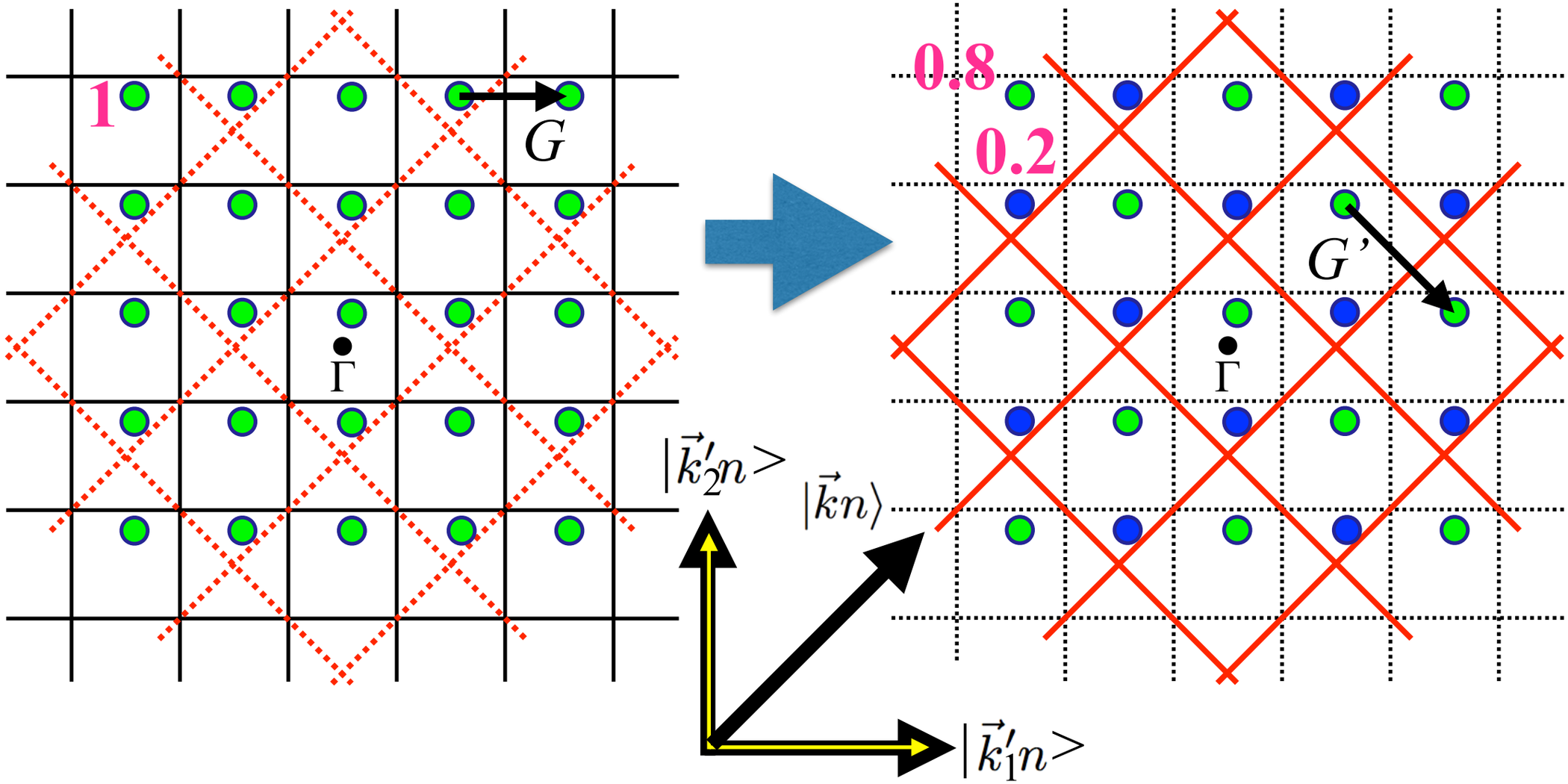
(translational symmetry is broken with respect to the original unit cell)

How to unfold bands? (change basis)

Plane wave: Change basis from |*kn*> to |*k'n*>

$$|\vec{k}n\rangle = \sum_{\vec{G}} C^n_{\vec{k}+\vec{G}} |\vec{k}+\vec{G}\rangle$$

$$= \sum_{\vec{k}'} \sum_{\vec{G}'} C^n_{\vec{k}'+\vec{G}'} |\vec{k}'+\vec{G}'\rangle$$



◉ Unfolded weight $|\ll \vec{k}'n \parallel \vec{k}n\rangle|^2 = \sum_{\vec{G}'} |C^n_{\vec{k}'+\vec{G}'}|^2$

Plane wave: Change basis from |*kn*> to |*k'n*>

$$|\vec{k}n\rangle = \sum_M C^{LCAO}_{\vec{k}n,M}|\vec{k}M\rangle$$

$$= \sum_M C^{LCAO}_{\vec{k}n,M} \sum_{\vec{G}} |\vec{k}+\vec{G}\rangle\langle\vec{k}+\vec{G}|\vec{k}M\rangle$$

$$= \sum_{\vec{G}} (\sum_M C^{LCAO}_{\vec{k}n,M}\langle\vec{k}+\vec{G}|\vec{k}M\rangle)|\vec{k}+\vec{G}\rangle$$

$$\langle\vec{k}+\vec{G}|\vec{k}M\rangle = \frac{1}{\sqrt{v}} e^{-i(\vec{k}+\vec{G})\cdot\vec{r}_M} \times$$

$$\int d\vec{r}\langle\vec{r}-\vec{r}_M|M\rangle e^{-i(\vec{k}+\vec{G})\cdot(\vec{r}-\vec{r}_M)}$$

- Spherical harmonics
- Spherical Bessel functions

- More freedom to choose the conceptual unit cell for performing the unfolding and the completeness of plane waves is not essential.
- But need caution because of the **pseudo** wave functions (pseudopotential)

Chi-Cheng Lee *et al*., arXiv: 1707.02525 (2018), JPCM in press.

Change basis from |*KJ*> to |*kj*> in real space

- Another way is to change the basis of spectral function in real space.
- By assuming we have an eigenstate |kj> and its corresponding LCAO basis |kn> of the conceptual system, we can insert the identify operator composed of the supercell eigenstates |KJ>.

$$A_{kj,kj} = \sum_{mnK} S^{-1}_{nm}(k)\langle km|KJ\rangle A_{KJ,KJ}\langle KJ|kn\rangle$$

100% spectral weight at SC ε_{KJ}

$$A_{kj,kj}(\omega) = \frac{L}{l}\sum_{KG}\delta_{k-G,K} W_{KJ} A_{KJ,KJ}(\omega)$$

Overlap matrix elements for non-orthogonal basis set

$$W_{KJ} = \sum_{MNr} \mathrm{e}^{\mathrm{i}k(r-r'(M))} C^{KJ}_{M} C^{KJ*}_{N} S_{0N,rm(M)}$$

The derivation detail can be found in
Chi-Cheng Lee *et al*., J. Phys.: Condens. Matter **25**, 345501 (2013).

Change basis from |*KJ*> to |*kj*> in real space

This is the one currently available in OpenMX (v3.8)

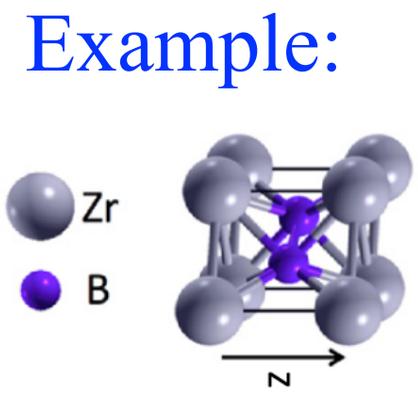
$$A_{kj,kj} = \sum_{mnK} S^{-1}_{nm}(k)\langle km|KJ\rangle A_{KJ,KJ}\langle KJ|kn\rangle$$

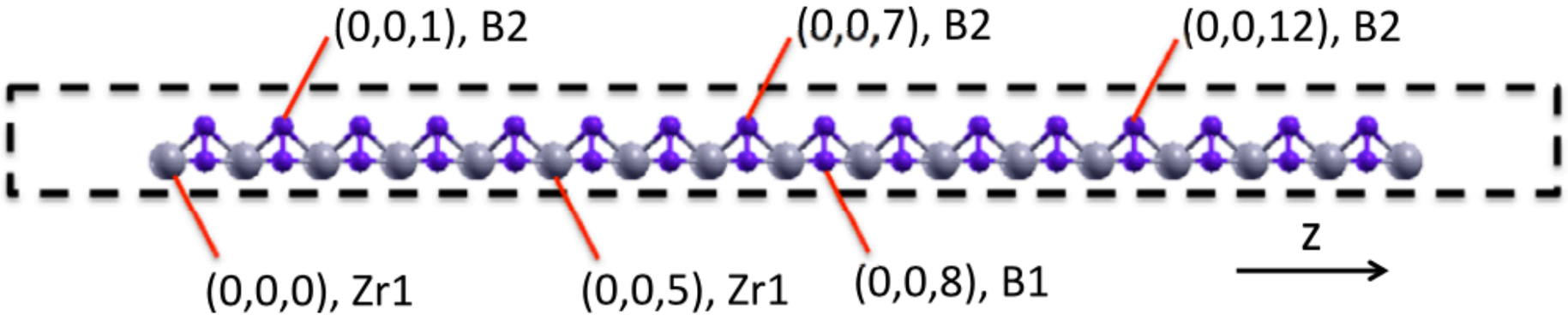
$$A_{kj,kj}(\omega) = \frac{L}{l}\sum_{KG}\delta_{k-G,K}W_{KJ}A_{KJ,KJ}(\omega)$$

$$W_{KJ} = \sum_{MNr} \mathrm{e}^{\mathrm{i}k(r-r'(M))}\, C^{KJ}_{M} C^{KJ*}_{N} S_{0N,rm(M)}$$

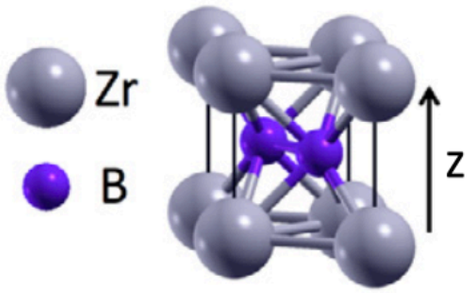
- The essential part is to relabel SC lattice vector by conceptual lattice vector and relabel the SC orbital in terms of the conceptual-cell orbital:
  $R \rightarrow R + r_0(M)$ and $M \rightarrow m'(M)$

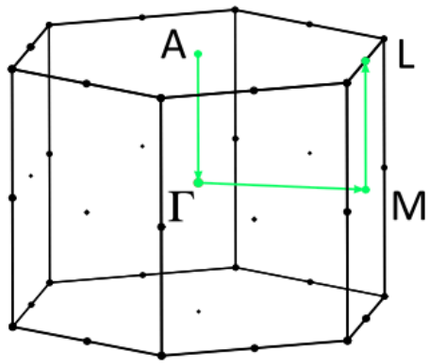
- Example:

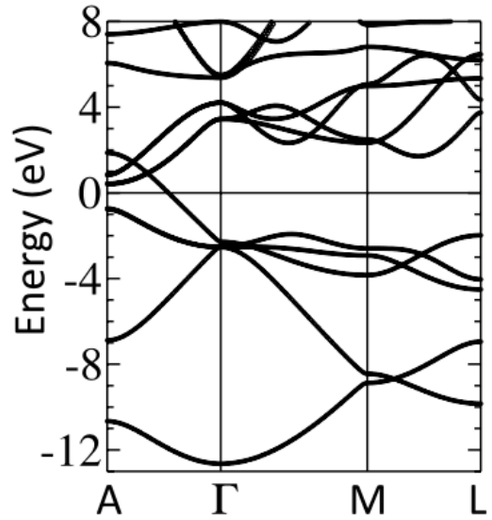




ZrB_2 slab

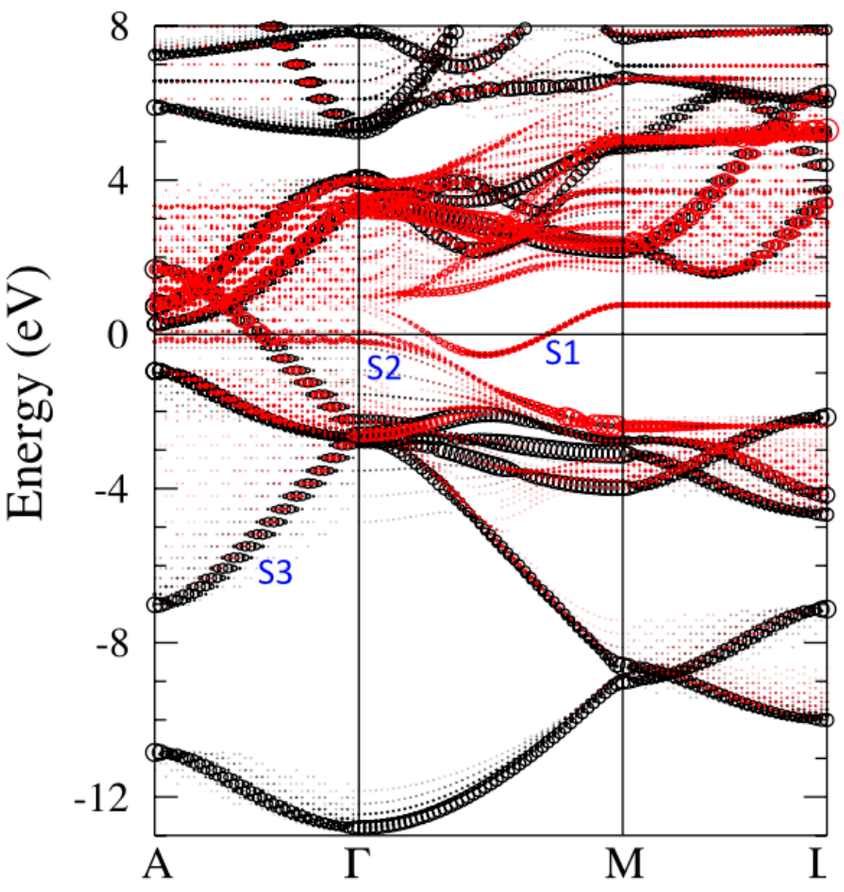


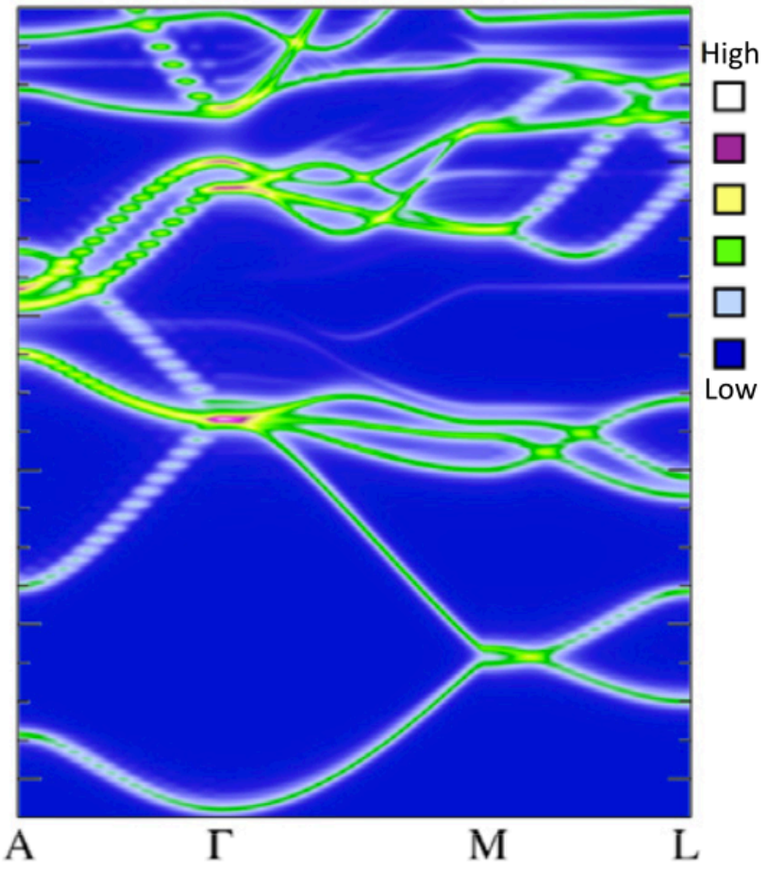




(bulk bands)

(unfolded slab bands)



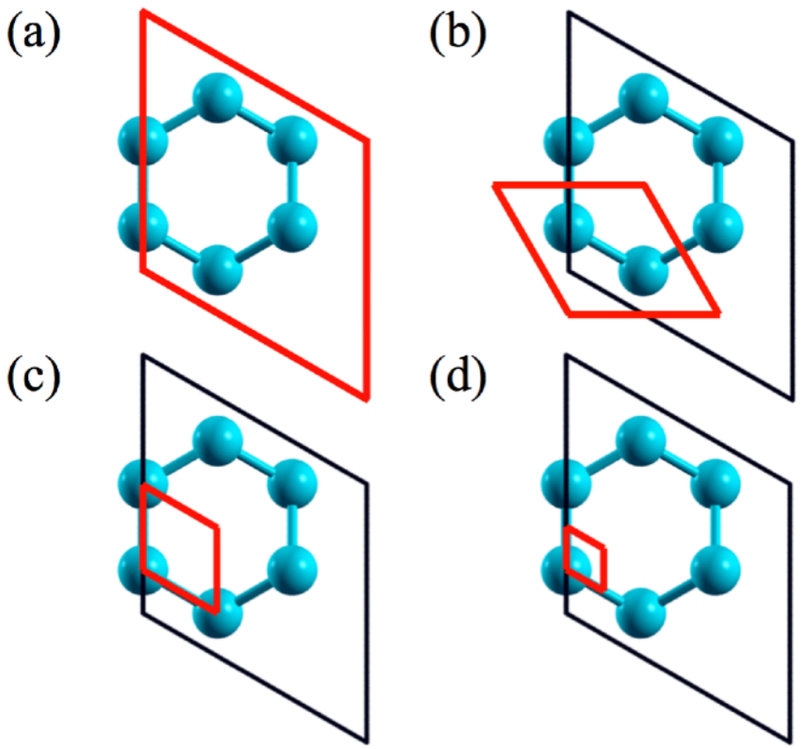


Chi-Cheng Lee *et al.*, J. Phys.: Condens. Matter **25**, 345501 (2013).

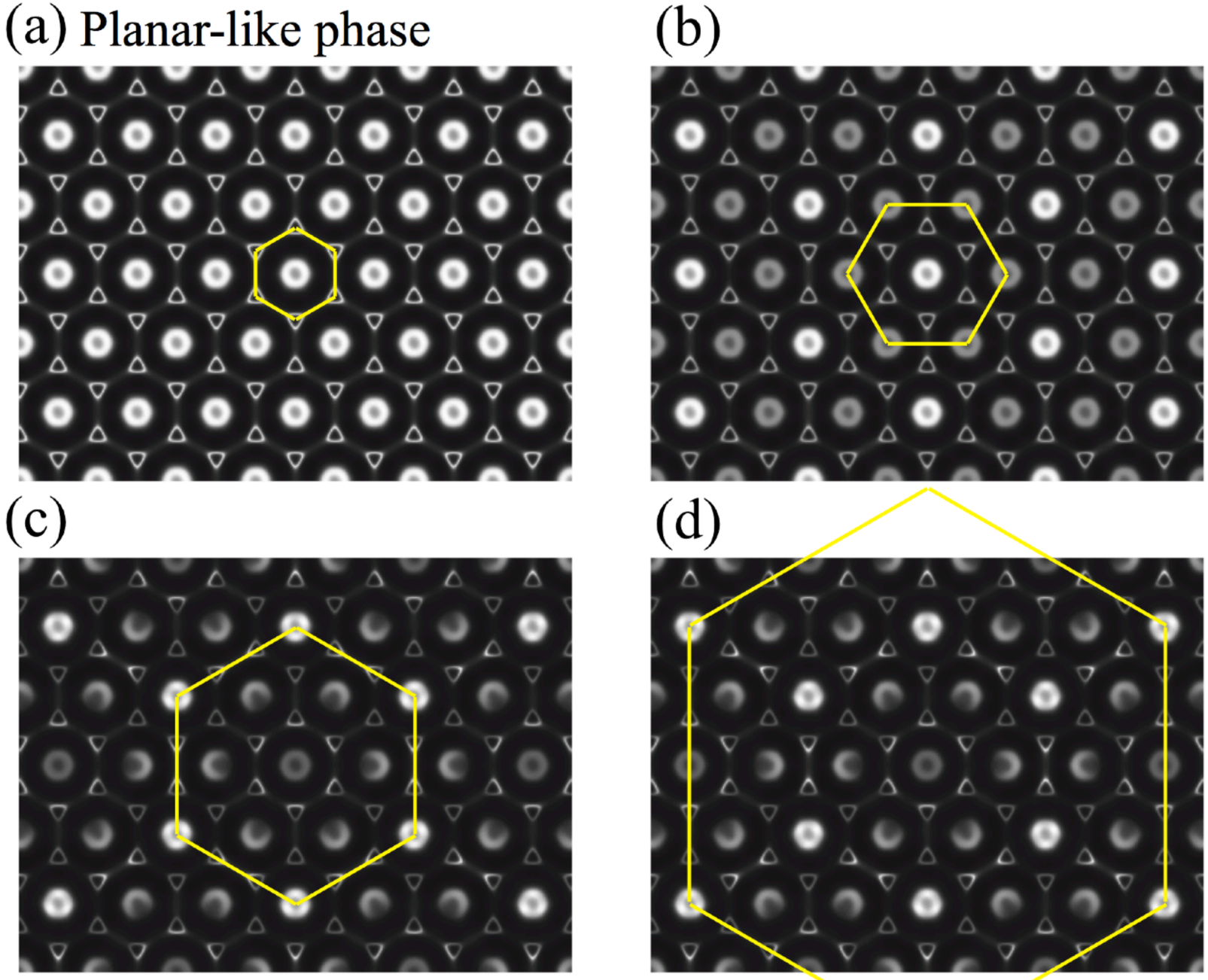
Example: Missing spectral weight

Silicene: Missing spectral weight

Free-standing planar-like silicene



Energy = 1eV below the Fermi energy

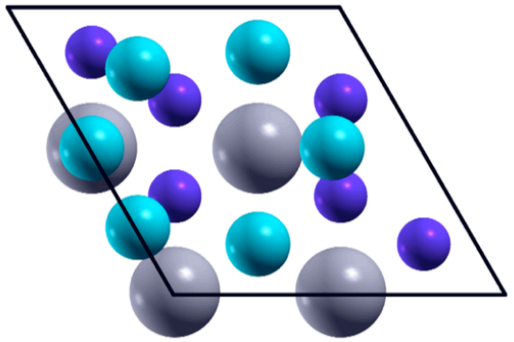


◉ Iso-energy surface, for example, Fermi surface, could be disconnected!

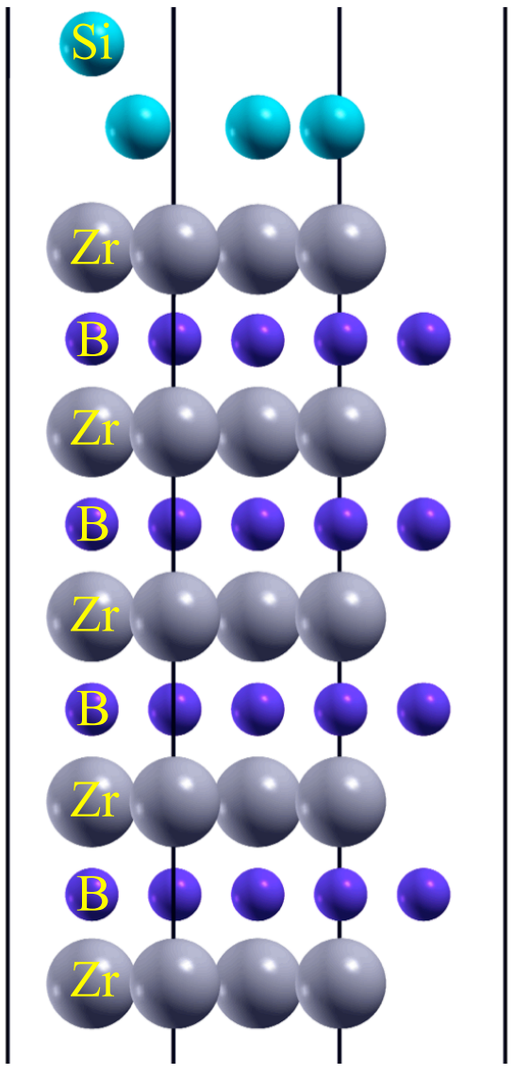
Chi-Cheng Lee *et al.*, Phys. Rev. B **90**, 075422 (2014).

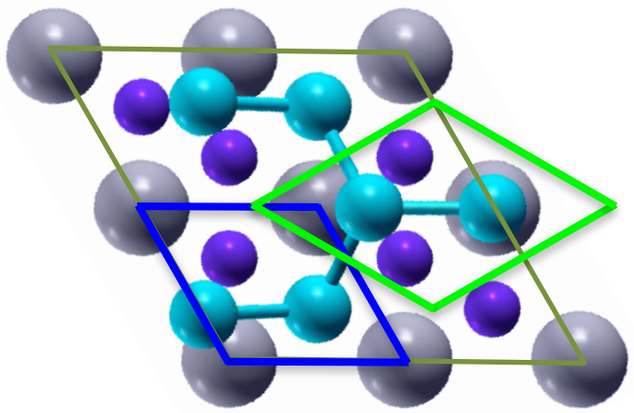
Silicene on ZrB_2: Choice of a good conceptual unit cell

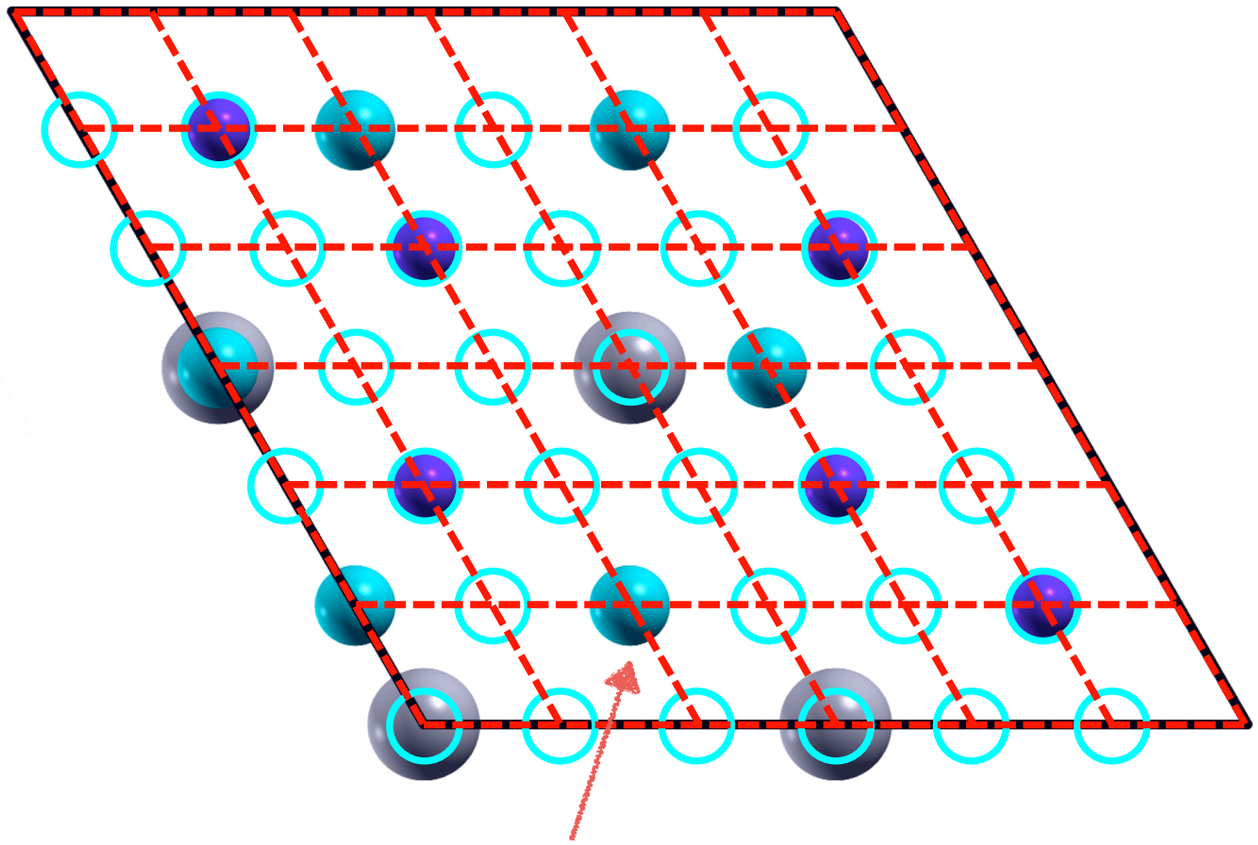
Planar-like silicene



$\sqrt{3} \times \sqrt{3}$ - reconstructed silicene



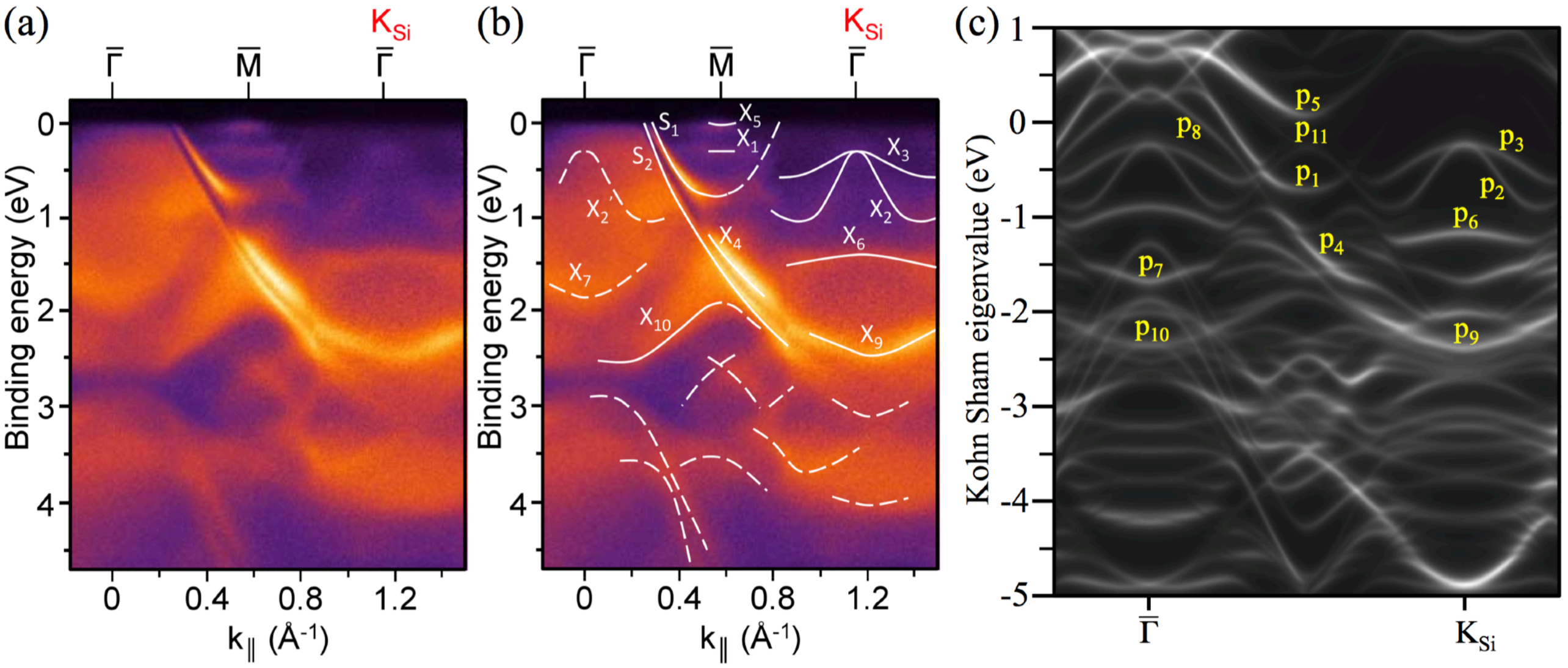




commensurate unit cell

Chi-Cheng Lee *et al.*, Phys. Rev. B **90**, 075422 (2014).

Silicene on ZrB_2: Unfolded spectral weight

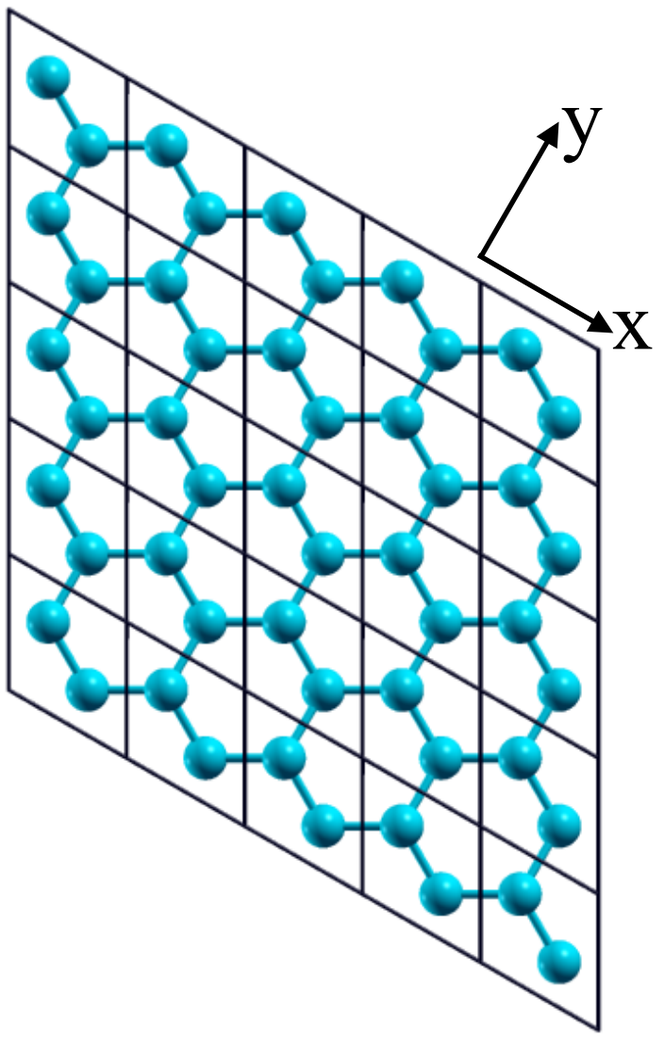


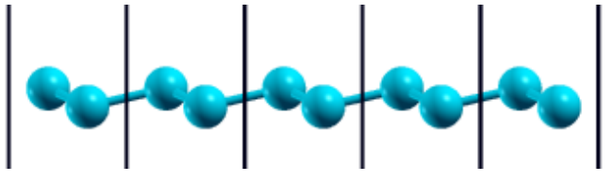
Chi-Cheng Lee *et al.*, Phys. Rev. B **90**, 075422 (2014).

How to run unfolding in OpenMX?

Free-standing silicene

Step 1: Choose a system to study

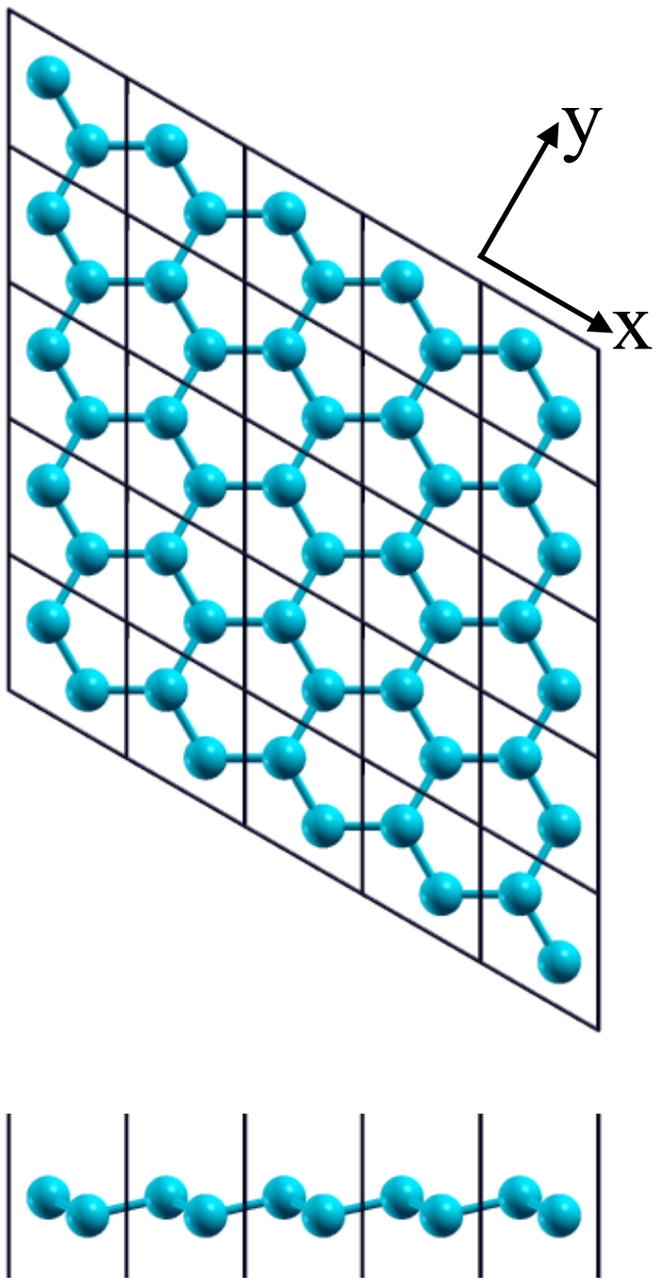




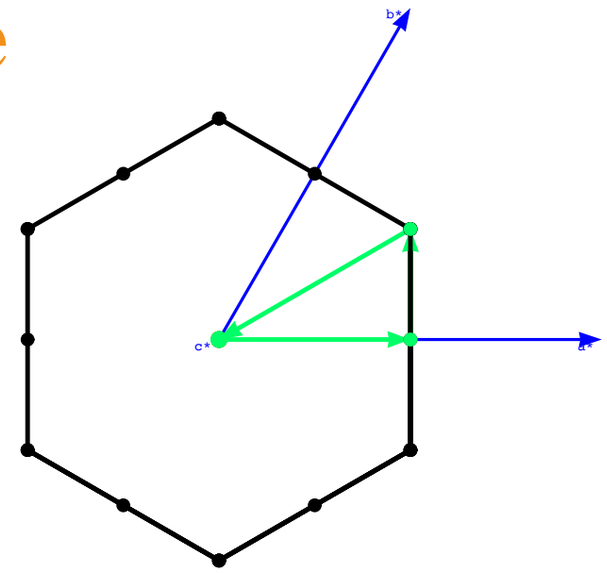
```
System.CurrrentDirectory         ./
System.Name                      Silicene
DATA.PATH                /provide_your_path/DFT_DATA13

Species.Number       1
<Definition.of.Atomic.Species
  Si   Si7.0-s2p2d1  Si_PBE13
Definition.of.Atomic.Species>

Atoms.Number         2
Atoms.SpeciesAndCoordinates.Unit   FRAC # Ang|AU
<Atoms.SpeciesAndCoordinates
     1     Si     0.33333   0.66666  0.4871  2. 2.
     2     Si     0.66666   0.33333  0.5128  2. 2.
Atoms.SpeciesAndCoordinates>

Atoms.UnitVectors.Unit  Ang
<Atoms.UnitVectors
  3.8577926 0 0
 -1.9288963 3.3409463939 0
  0 0 20
Atoms.UnitVectors>

scf.XcType                GGA-PBE      # LDA|LSDA-CA|LSDA-PW|GGA-PBE
scf.SpinPolarization       Off         # On|Off
scf.energycutoff           250.0       # default=150 (Ry)
scf.maxIter                100         # default=40
scf.EigenvalueSolver      band         # Recursion|Cluster|Band
scf.Kgrid                  8 8 1       # means n1 x n2 x n3
scf.Mixing.Type            rmm-diisk   # Simple|Rmm-Diis|Gr-Pulay
scf.criterion             1.0e-8       # default=1.0e-6 (Hartree)
```

Free-standing silicene

Step 2: Choose *k* paths for your band structure



We can plot band structure using unfolding keyword



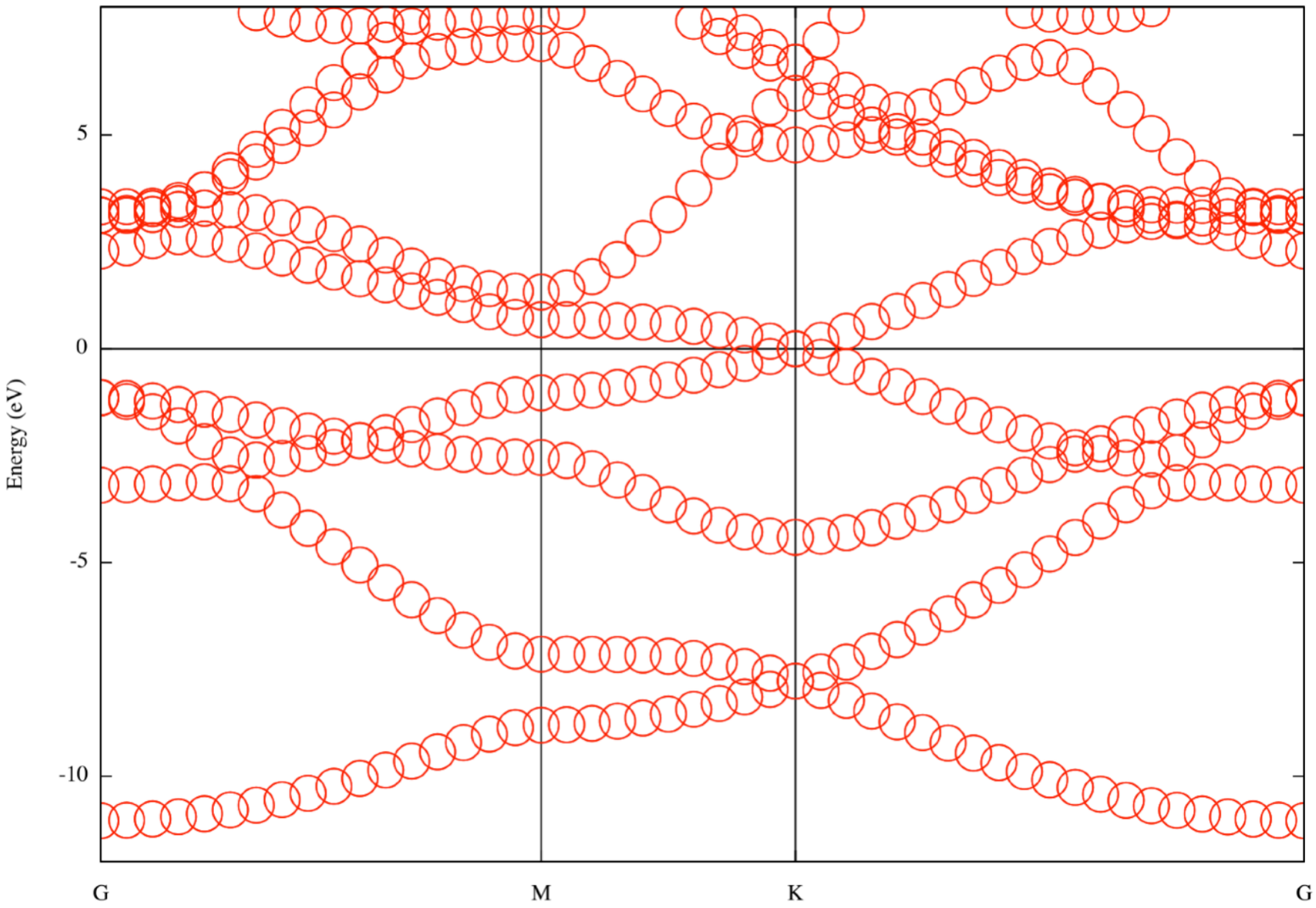

```
Unfolding.Electronic.Band       on          # on|off, default=off
Unfolding.LowerBound         -12.0          # default=-10 eV
Unfolding.UpperBound           8.0          # default= 10 eV

Unfolding.Nkpoint                4

<Unfolding.kpoint
G 0 0 0
M 0.5 0 0
K 0.3333333 0.3333333 0
G 0 0 0
Unfolding.kpoint>

Unfolding.desired_totalnkpt    50
```

- Although the keyword is “unfolding”, the *unfolding* is not performed! More precisely, the band is unfolded to itself (the same zone).

Free-standing silicene

Step 3: Looking for the output files: Silicene.unfold_totup Silicene.unfold_orbup Silicene.unfold_plotexample

gnuplot
gnuplot> load ‘Silicene.unfold_plotexample’
plot 'Silicene.unfold_totup' using 1:2:($3)*0.02 notitle with circles lc rgb 'red'

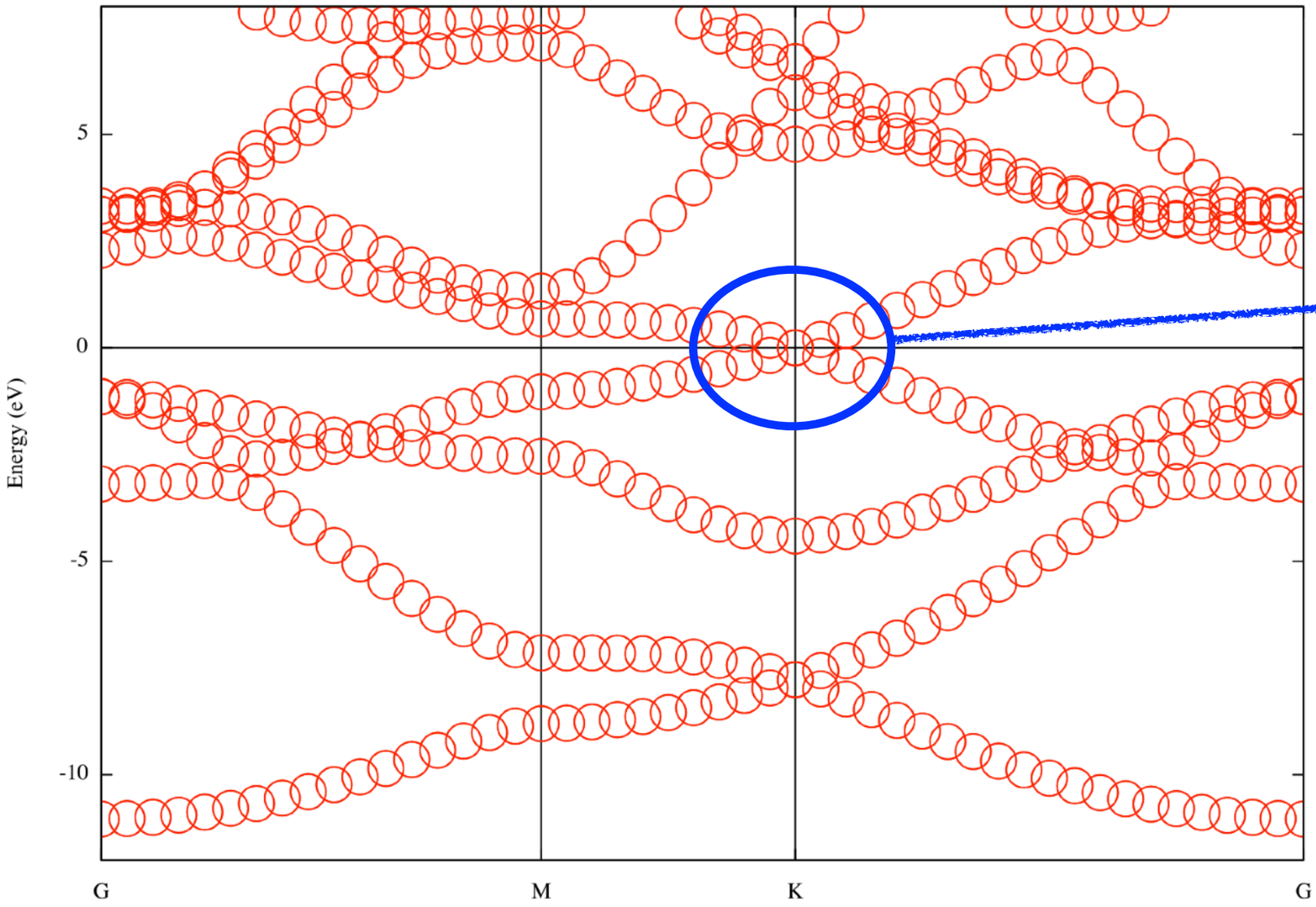



```
Silicene.unfold_totup

0.000000 -11.038613  1.0000000
0.000000 -3.189627  1.0000000
0.000000 -1.146319  1.0000000
0.000000 -1.140370  1.0000000
0.000000 2.287865  1.0000000
0.000000 3.125793  1.0000000
0.000000 3.130633  1.0000000
0.000000 3.321159  1.0000000
0.029271 -11.028500  1.0000000
0.029271 -3.182389  1.0000000
0.029271 -1.233445  1.0000000
0.029271 -1.161229  1.0000000
0.029271 2.363152  1.0000000
...
```

weight is 1

Free-standing silicene

Step 4: Try to find something interesting



We have a Dirac cone. What is the orbital contribution?

- The orbital contribution can be found in Silicene.unfold_orbup and the format is k_dis (Bohr^{-1}), energy(eV), and weight. The sequence of the orbital weights can be found in Silicene.out.

Free-standing silicene

Step 5: Read output file for picking up the investigated orbitals

In Silicene.out, we can find

The sequence for the orbital weights in System.Name.unfold_orbup(dn) is given below.

```
1     1  Si 0 s
2           1 s
3           0 px
4           0 py
5           0 pz
6           1 px
7           1 py
8           1 pz
9           0 d3z^2-r^2
10          0 dx^2-y^2
11          0 dxy
12          0 dxz
13          0 dyz
14    2  Si 0 s
15          1 s
16          0 px
17          0 py
18          0 pz
19          1 px
20          1 py
21          1 pz
22          0 d3z^2-r^2
23          0 dx^2-y^2
24          0 dxy
25          0 dxz
26          0 dyz
```

3
4
5
6
7

k_dis (Bohr^{-1}), energy(eV)

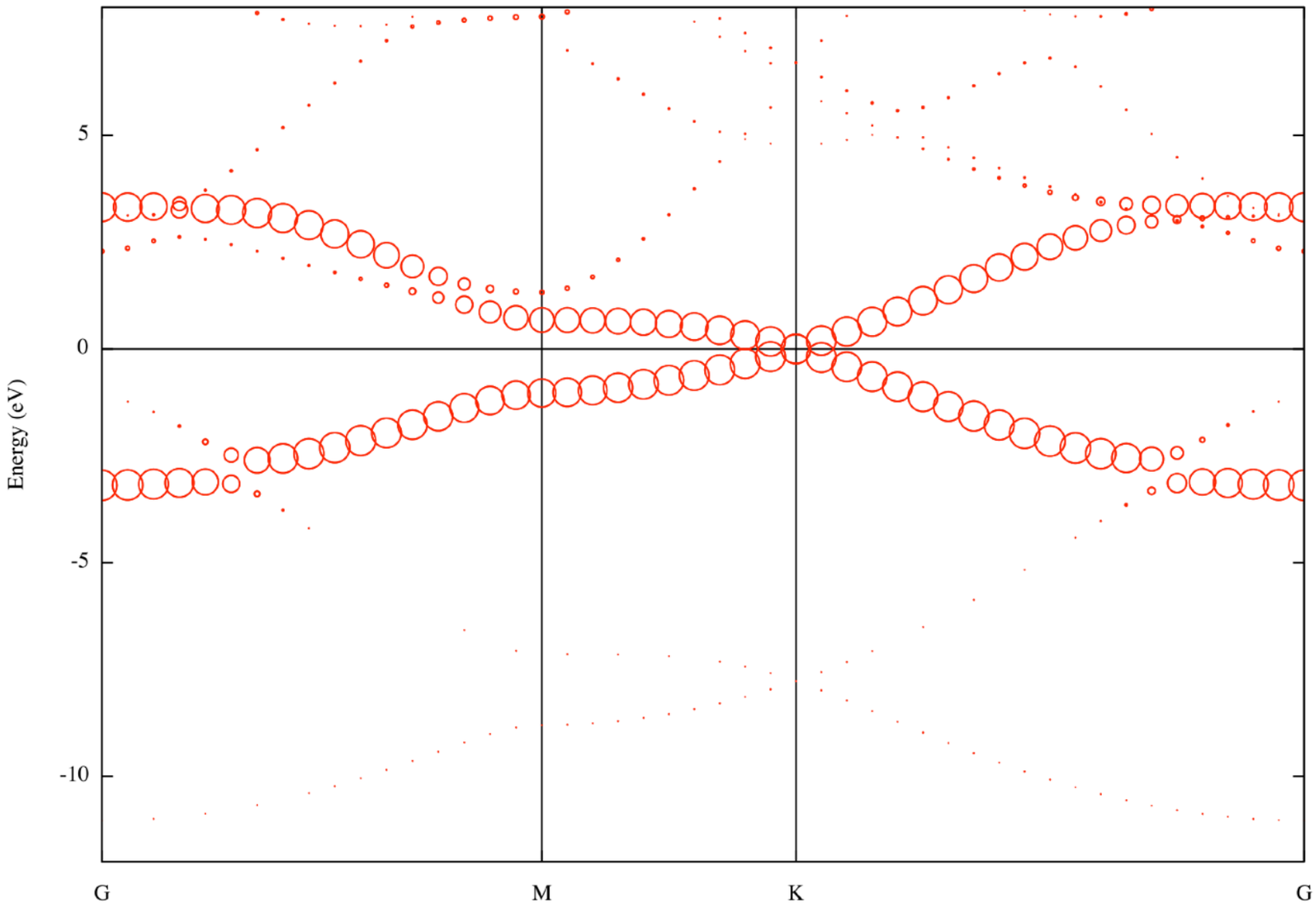
1 2

p_z: \$7+\$10+\$20+\$23

Free-standing silicene

Step 6: Examine orbital contribution

gnuplot
gnuplot> load ‘Silicene.unfold_plotexample’
plot 'Silicene.unfold_orbup' using 1:2:($7+$10+$20+$23)*0.02 notitle with circles lc rgb 'red'

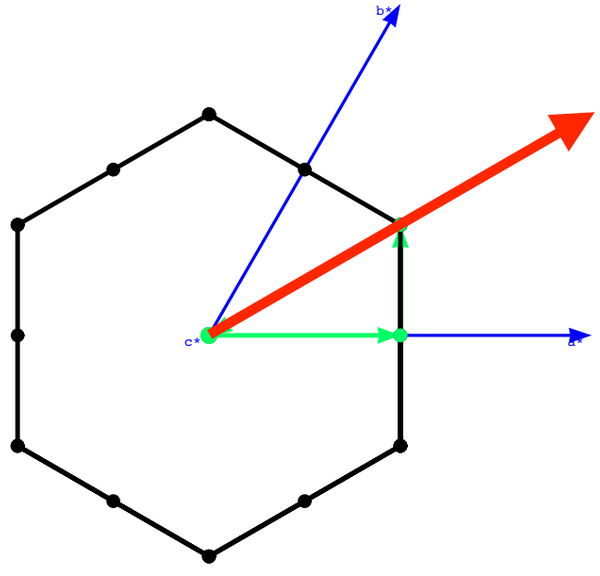


◉ The Dirac cone is composed of p_z orbitals

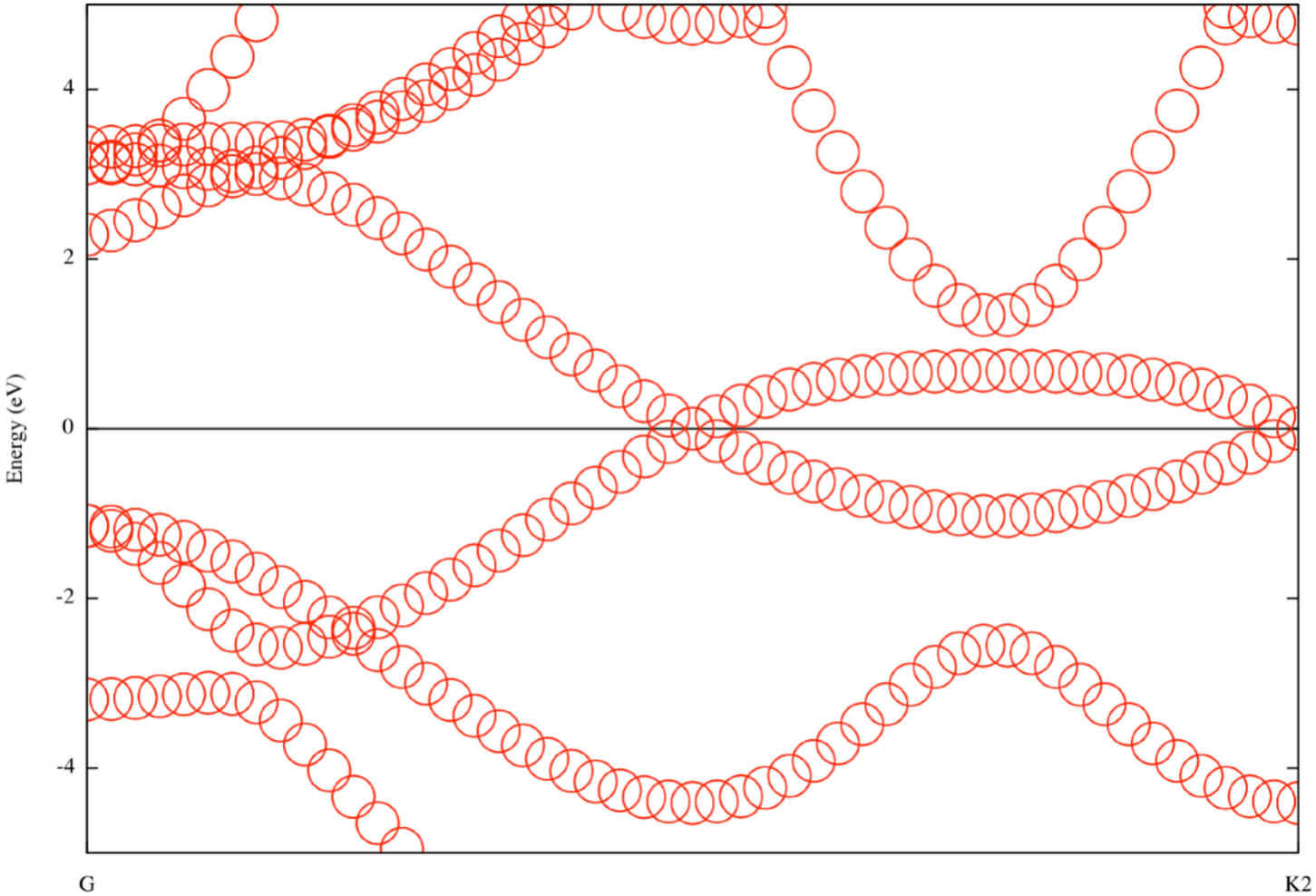
Free-standing silicene

Step 7: Find something even more interesting
Preparation for real unfolding

```
Unfolding.Electronic.Band       on        # on|off, default=off
Unfolding.LowerBound          -5.0        # default=-10 eV
Unfolding.UpperBound           5.0        # default= 10 eV
Unfolding.Nkpoint                2
<Unfolding.kpoint
G 0 0 0
K2 0.6666666 0.6666666 0
Unfolding.kpoint>
Unfolding.desired_totalnkpt     51
```



Let us focus on the red path

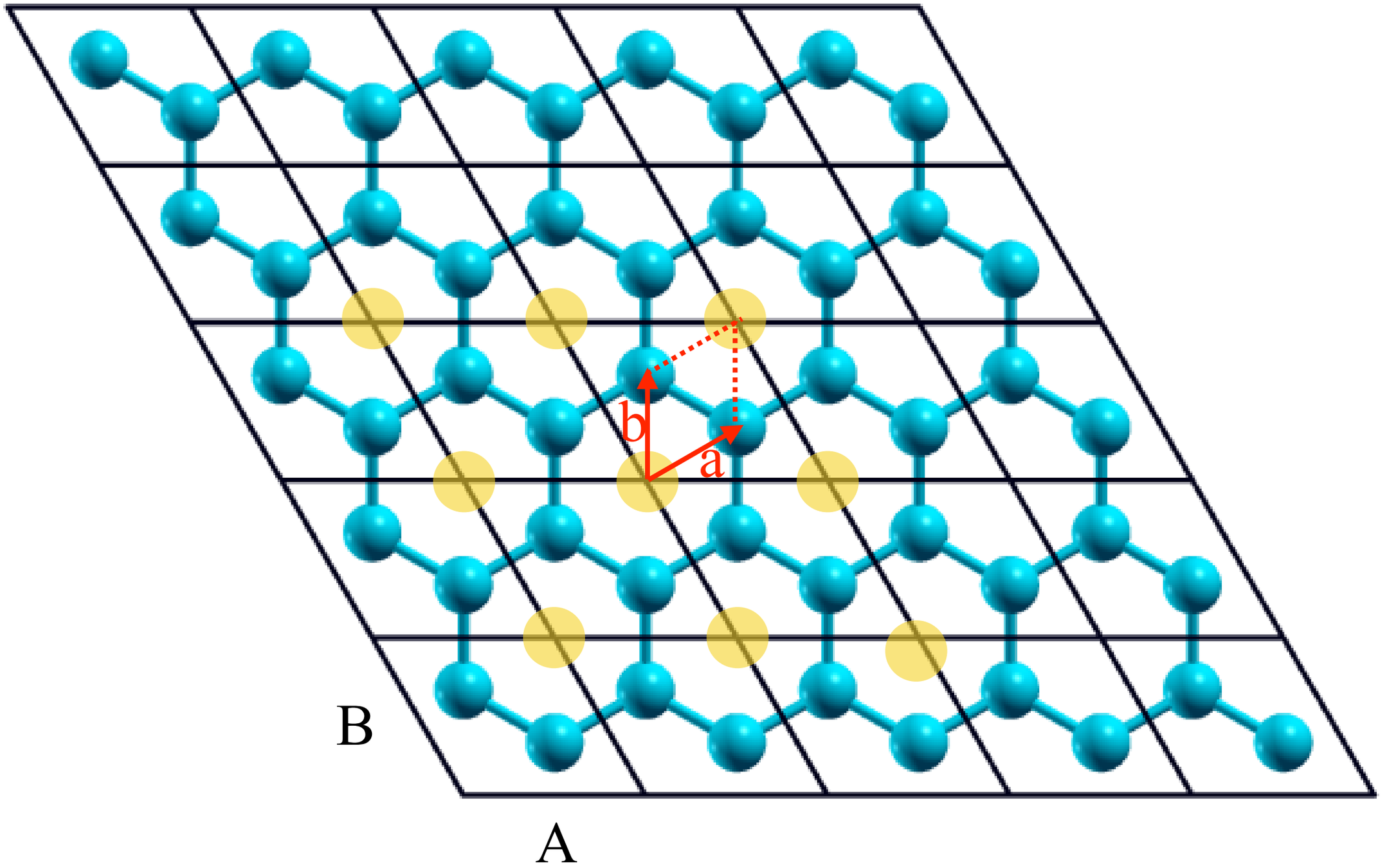


Will the result be different if we choose a larger BZ and unfold the bands to that zone?

The choice of a new *conceptual* unit cell seems to be important.

Free-standing silicene

Step 8: Find a conceptual unit cell for your purpose



◉ The conceptual unit cell is still commensurate with the primitive unit cell

Free-standing silicene

Step 9: Unfolding: Provide the information of conceptual cell

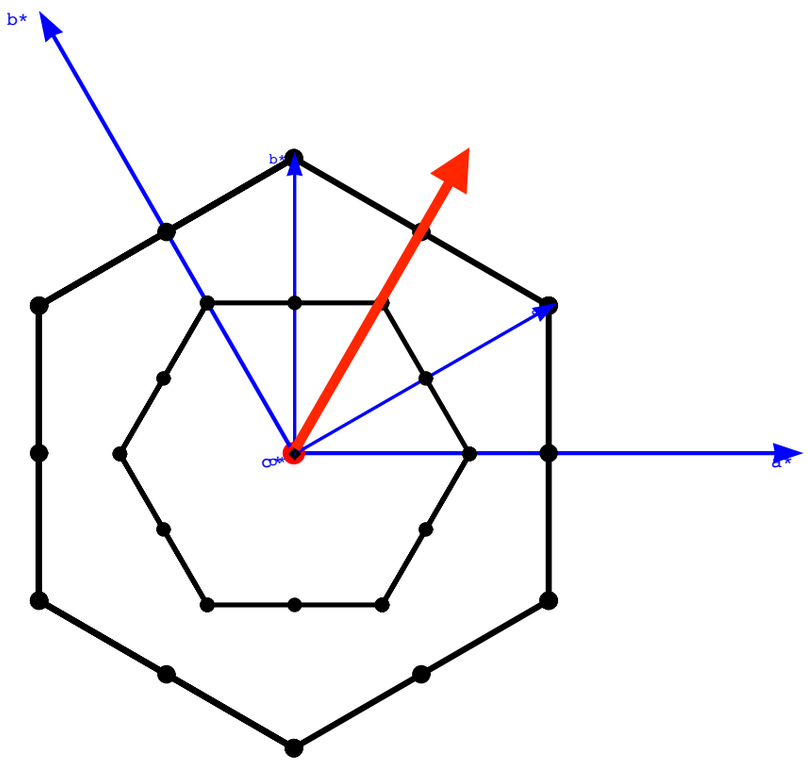
Calculated unit cell (supercell)

```
Atoms.UnitVectors.Unit  Ang
<Atoms.UnitVectors
  3.8577926 0 0
 -1.9288963 3.3409463939 0
  0 0 20
Atoms.UnitVectors>
```

Conceptual unit cell that also defines the new Brillouin zone

```
<Unfolding.ReferenceVectors
  1.9288963033 1.1136488 0
  0 2.2272976 0
  0 0 20
Unfolding.ReferenceVectors>

Unfolding.Electronic.Band      on
Unfolding.LowerBound         -5.0
Unfolding.UpperBound          5.0
Unfolding.Nkpoint               2
Unfolding.desired_totalnkpt    51

<Unfolding.kpoint
G 0 0 0
K2 0.6666666 0.6666666 0
Unfolding.kpoint>

<Unfolding.Map
1 1
2 1
Unfolding.Map>
```

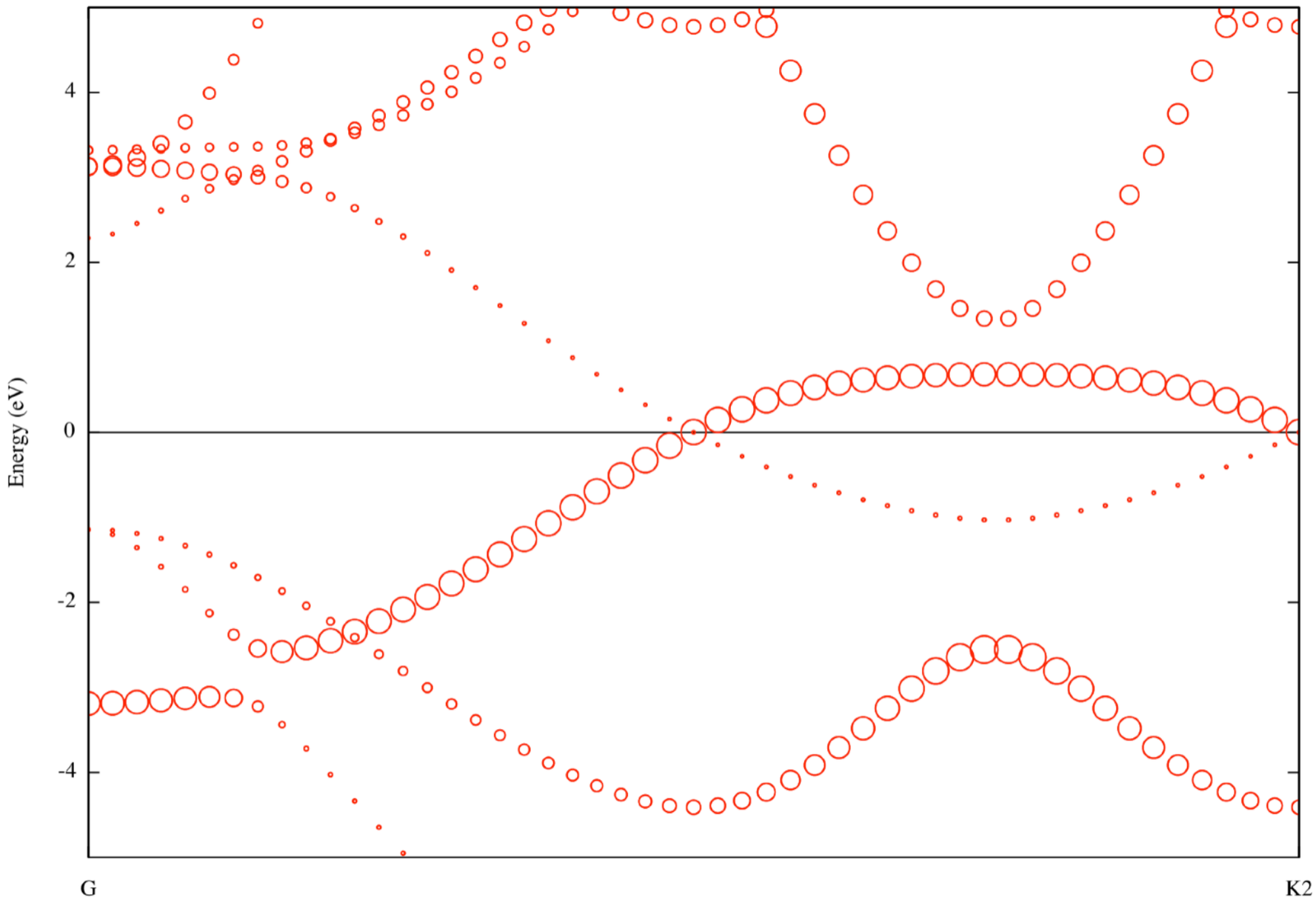
using the reciprocal lattice vectors of the conceptual unit cell as the basis vectors



Free-standing silicene

Step 10: Plot unfolded spectral weight

gnuplot
gnuplot> load ‘Silicene.unfold_plotexample’
plot 'Silicene.unfold_totup' using 1:2:($3)*0.02 notitle with circles lc rgb 'red'



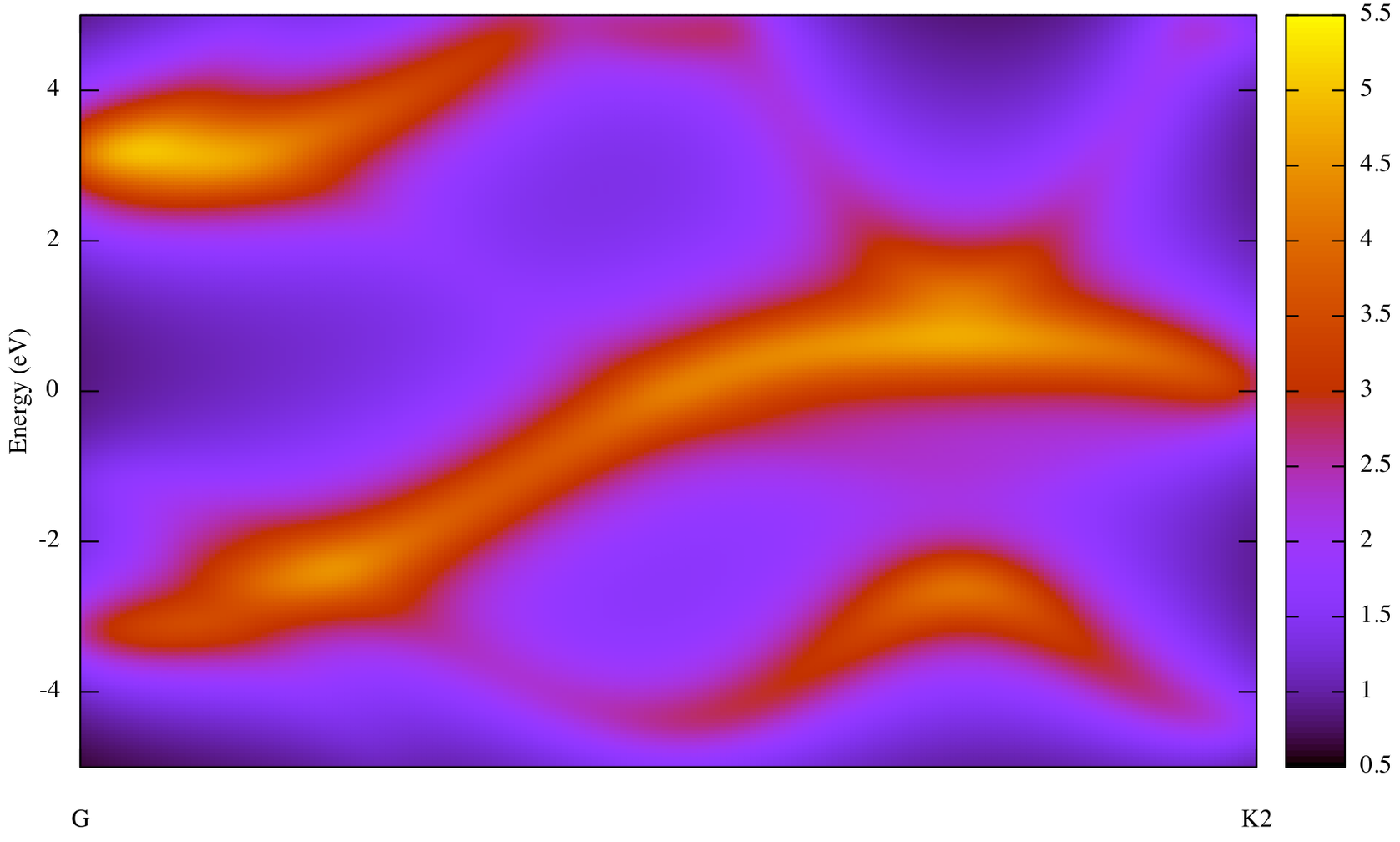
Free-standing silicene

Step 10: Or using intensity map

gcc intensity_map.c -lm -o intensity_map (do this in the “source” directory)
./intensity_map Silicene.unfold_totup -c 3 -k 0.5 -e 0.05 -l -5 -u 5 > map.txt
modify the Silicene.unfold_plotexample as the following:

```
set yrange [-5.000000:5.000000]
set ylabel 'Energy (eV)'
set xtics('G' 0.000000,'K2' 1.149161)
set xrange [0:1.149161]
set arrow nohead from 0,0 to 1.149161,0
set pm3d map
sp ‘map.txt
```

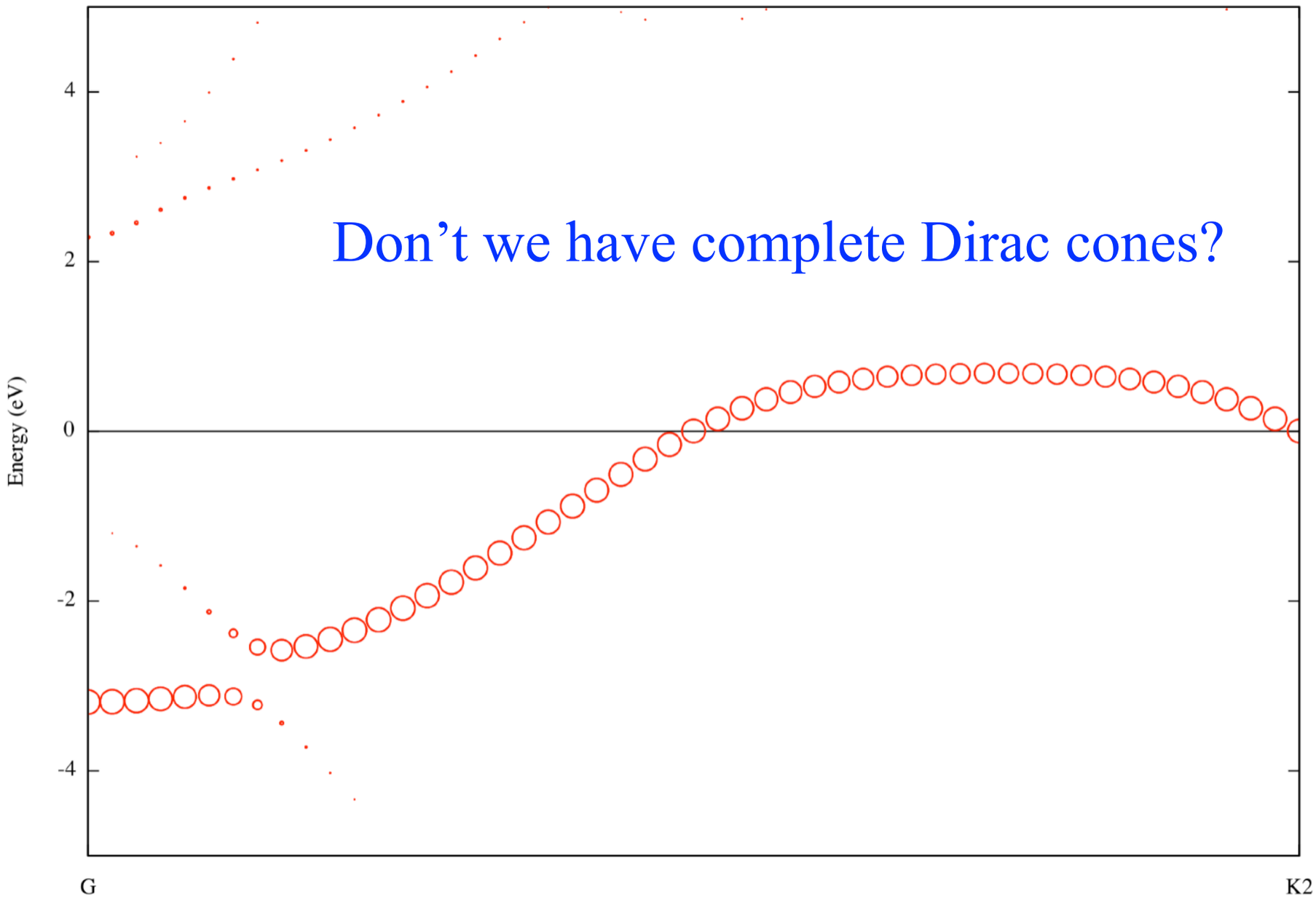
gnuplot> load 'Silicene.unfold_plotexample'



Free-standing silicene

Step 10: Or plot unfolded spectral weight of certain orbitals

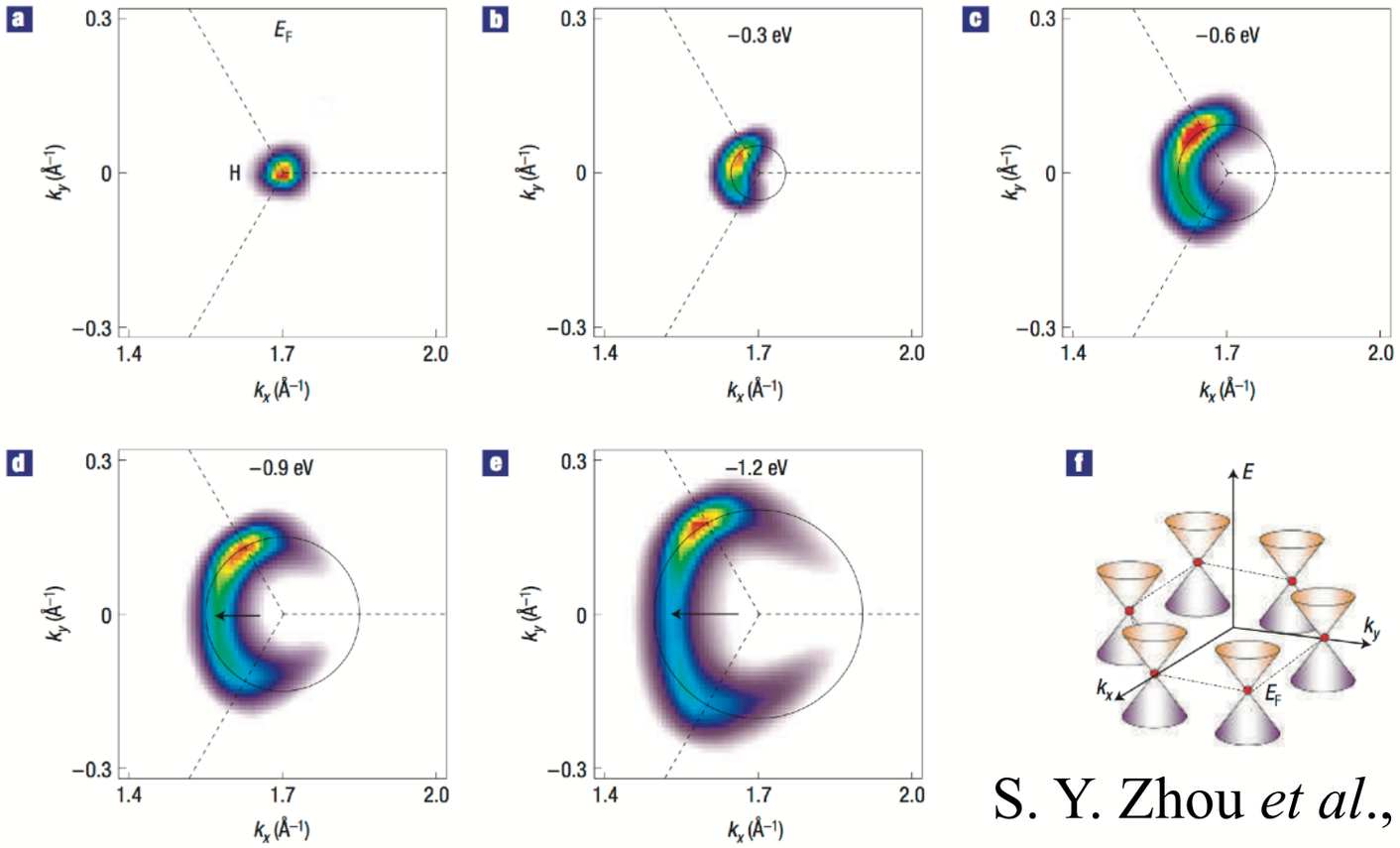
gnuplot
gnuplot> load ‘Silicene.unfold_plotexample
plot 'Silicene.unfold_orbup' using 1:2:($7+$10+$20+$23)*0.02 notitle with circles lc rgb 'red'



Free-standing silicene and graphene

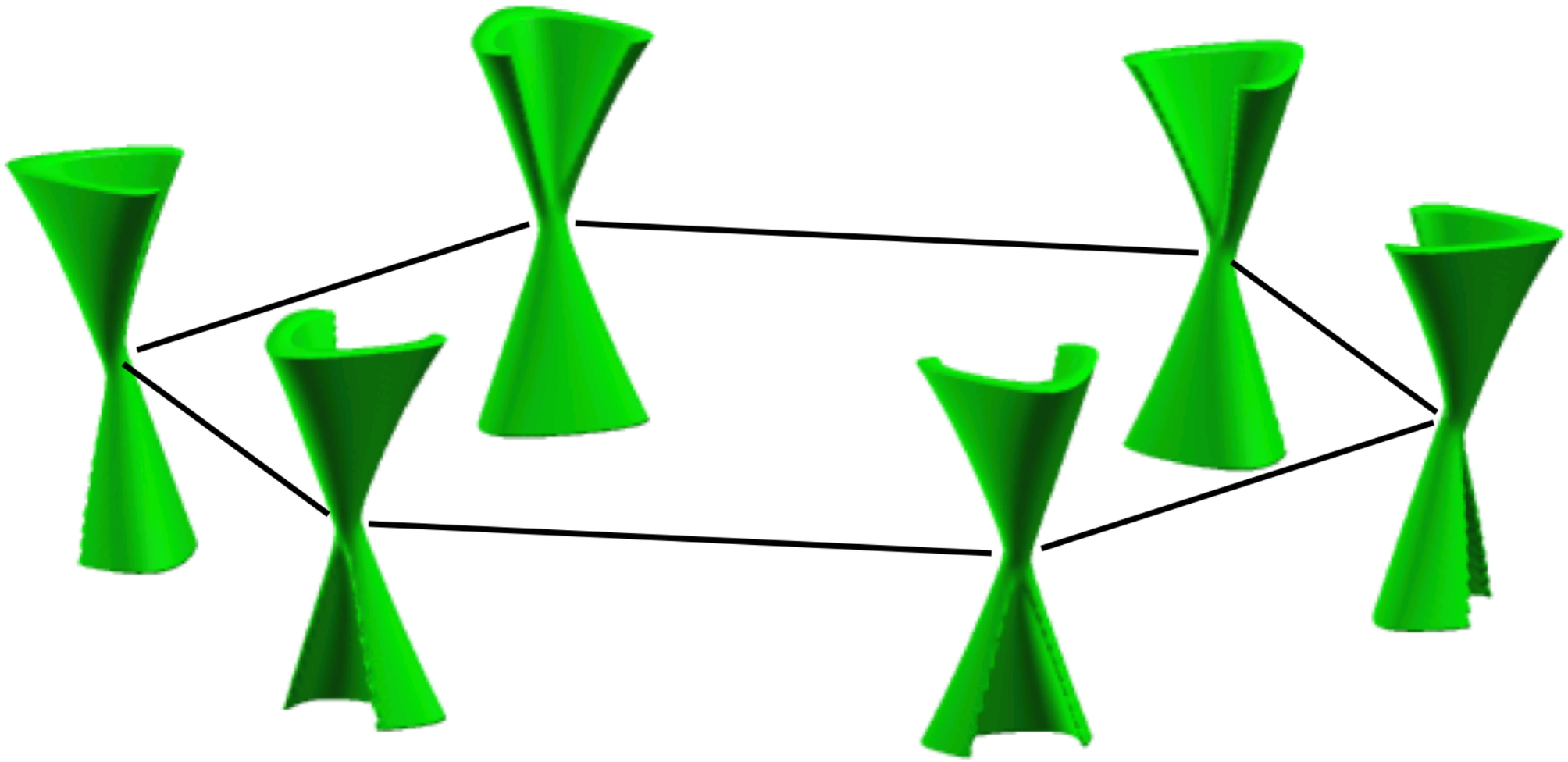
Step 11: Think about what is going on

- The translational symmetry breaking seen by the conceptual unit cell is two-fold. One is the periodic Si vacancies and the other is the dislocation due to the buckling. We can see the Si_A-Si_B interference because we label them as the same atom. In the map, we define 1 and 1 (not 2).
- The missing spectral weight indicates an incomplete loop of Dirac cone at constant Energy.
- Incomplete constant-energy contour of Dirac cone observed in graphene:



S. Y. Zhou *et al*., Nature Physics **2**, 595 (2006)

Dirac cone represented by extended-zone scheme



◉ Dirac cone is intrinsically broken in momentum space

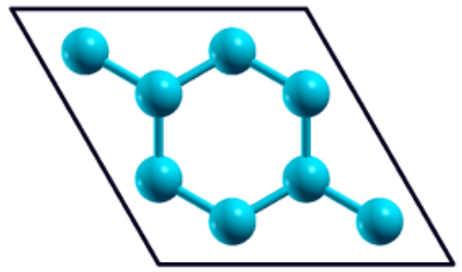
Chi-Cheng Lee *et al*., arXiv: 1707.02525 (2018), JPCM in press.

Another case for running unfolding

Free-standing silicene

Step 1: Build a perfect supercell but unfold to primitive zone

```
System.CurrrentDirectory         ./
System.Name                      Silicene
DATA.PATH                /provide_your_path/DFT_DATA13

Species.Number       1
<Definition.of.Atomic.Species
  Si   Si7.0-s2p2d1  Si_PBE13
Definition.of.Atomic.Species>

Atoms.Number         8
Atoms.SpeciesAndCoordinates.Unit   FRAC # Ang|AU
<Atoms.SpeciesAndCoordinates
     1     Si     0.166667  0.333333  0.4871  2. 2.
     2     Si     0.333333  0.166667  0.5128  2. 2.
     3     Si     0.666667  0.333333  0.4871  2. 2.
     4     Si     0.833333  0.166667  0.5128  2. 2.
     5     Si     0.166667  0.833333  0.4871  2. 2.
     6     Si     0.333333  0.666667  0.5128  2. 2.
     7     Si     0.666667  0.833333  0.4871  2. 2.
     8     Si     0.833333  0.666667  0.5128  2. 2.
Atoms.SpeciesAndCoordinates>

Atoms.UnitVectors.Unit  Ang
<Atoms.UnitVectors
  7.7155852 0 0
 -3.8577926 6.6818927878 0
  0 0 20
Atoms.UnitVectors>

scf.XcType               GGA-PBE      # LDA|LSDA-CA|LSDA-PW|GGA-PBE
scf.SpinPolarization      Off         # On|Off
scf.energycutoff          250.0       # default=150 (Ry)
scf.maxIter               100         # default=40
scf.EigenvalueSolver     band         # Recursion|Cluster|Band
scf.Kgrid                 4 4 1       # means n1 x n2 x n3
scf.Mixing.Type           rmm-diisk   # Simple|Rmm-Diis|Gr-Pulay
scf.criterion            1.0e-8       # default=1.0e-6 (Hartree)
```



```
Unfolding.Electronic.Band      on
Unfolding.LowerBound         -12.0
Unfolding.UpperBound          8.0

Unfolding.Nkpoint              4

Unfolding.desired_totalnkpt    50

<Unfolding.ReferenceVectors
3.8577926 0 0
-1.9288963 3.3409463939 0
0 0 20
Unfolding.ReferenceVectors>

<Unfolding.kpoint
G 0 0 0
M 0.5 0 0
K 0.3333333 0.3333333 0
G 0 0 0
Unfolding.kpoint>

<Unfolding.Map
1 1
2 2
3 1
4 2
5 1
6 2
7 1
8 2
Unfolding.Map>
```

Free-standing silicene

Step 2: Plot unfolded band structure with SC bands

To do that, we need to turn on band dispersion

```
Band.dispersion                  on
<Band.KPath.UnitCell
  3.8577926 0 0
 -1.9288963 3.3409463939 0
  0 0 20
 Band.KPath.UnitCell>
Band.Nkpath                    3
<Band.kpath
17 0 0 0  0.5 0 0 G M
10 0.5 0 0 0.333333 0.333333 0 M K
20 0.333333 0.333333 0 0 0 0 K G
Band.kpath>
```

We also need to compile

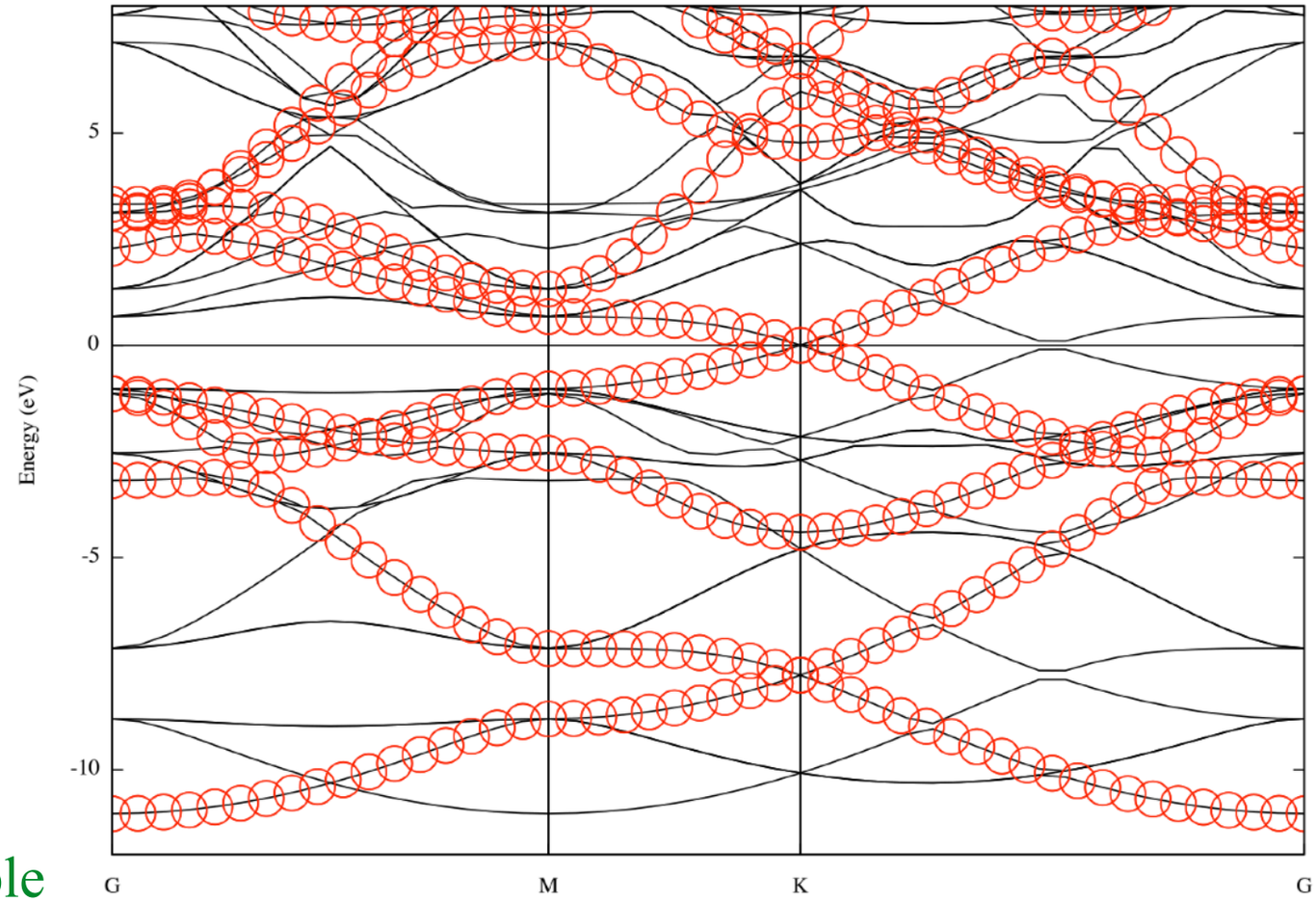
```
gcc bandgnu13.c -lm -o bandgnu13
```

and perform (to get Silicene.BANDDAT1)

```
./bandgnu13 Silicene.Band
```

We can modify the Silicene.unfold_plotexample

```
set yrange [-12.000000:8.000000]
set ylabel 'Energy (eV)'
set xtics('G' 0.000000,'M' 0.497601,'K' 0.784892,'G' 1.359472)
set xrange [0:1.359472]
set arrow nohead from 0,0 to 1.359472,0
set arrow nohead from 0.497601,-12.000000 to 0.497601,8.000000
set arrow nohead from 0.784892,-12.000000 to 0.784892,8.000000
set style circle radius 0
set style data lines
p "Silicene.BANDDAT1" notitle lc rgb 'black',"Silicene.unfold_totup" u 1:2:($3)*0.02 notitle w circles lc rgb 'red'
```



We have 1 and 0 in the weight

```
0.000000 -11.038611  1.0000000
0.000000 -8.808899  0.0000000
0.000000 -8.804652  0.0000000
0.000000 -8.804652  0.0000000
...
```

(Silicene.unfold_totup)

We can run gnuplot and load 'Silicene.unfold_plotexample'

Free-standing silicene

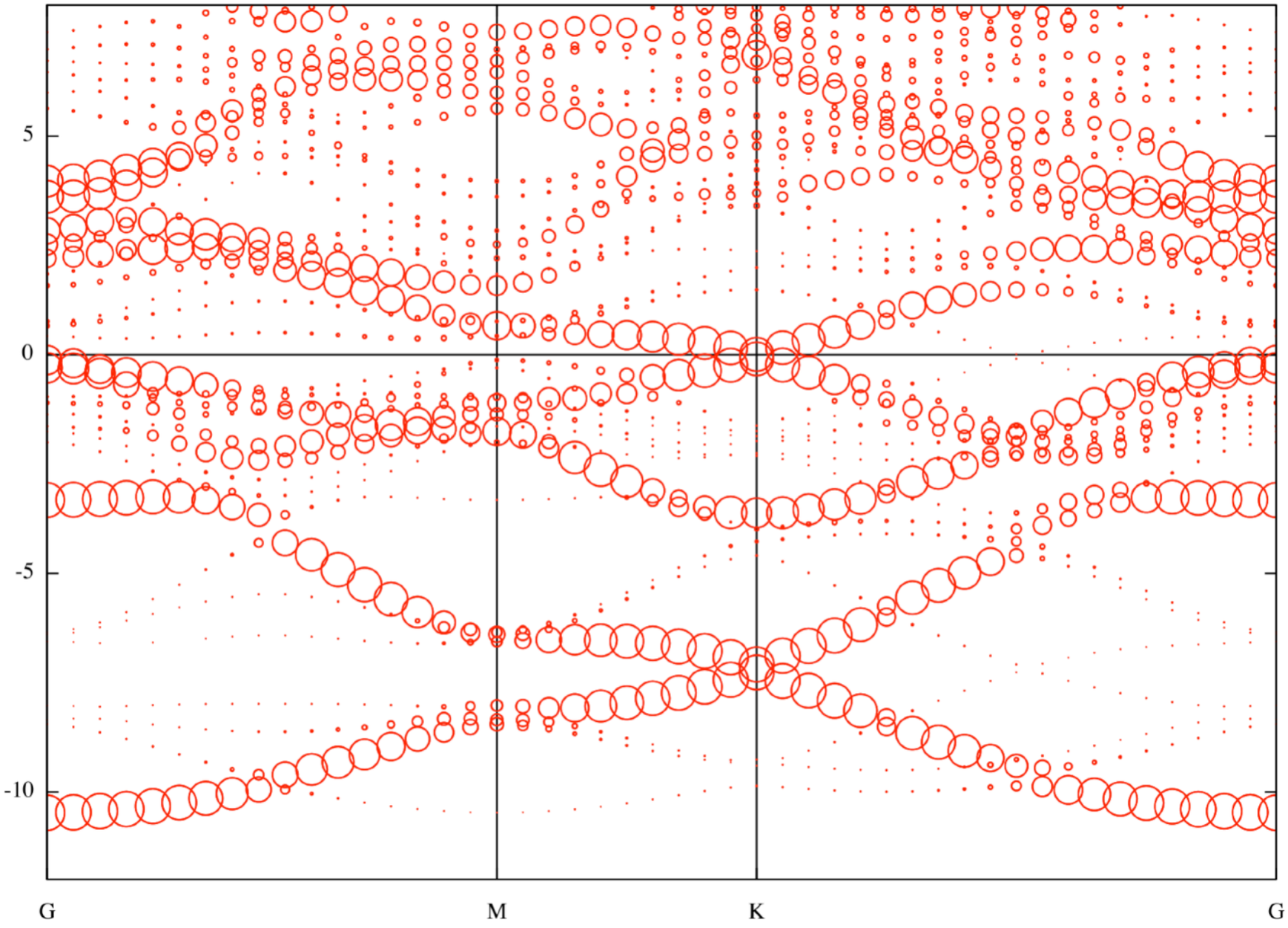
Step 3: Perturb the atomic positions by hand

```
System.CurrrentDirectory         ./
System.Name                      Silicene
DATA.PATH              /provide_your_path/DFT_DATA13

Species.Number       1
<Definition.of.Atomic.Species
  Si   Si7.0-s2p2d1  Si_PBE13
Definition.of.Atomic.Species>

Atoms.Number         8
Atoms.SpeciesAndCoordinates.Unit   FRAC # Ang|AU
<Atoms.SpeciesAndCoordinates
     1     Si     0.186667  0.353333  0.4971  2. 2.
     2     Si     0.313333  0.146667  0.5228  2. 2.
     3     Si     0.656667  0.313333  0.4871  2. 2.
     4     Si     0.823333  0.156667  0.5128  2. 2.
     5     Si     0.156667  0.853333  0.4771  2. 2.
     6     Si     0.343333  0.686667  0.5328  2. 2.
     7     Si     0.656667  0.813333  0.4671  2. 2.
     8     Si     0.843333  0.686667  0.5128  2. 2.
Atoms.SpeciesAndCoordinates>

Atoms.UnitVectors.Unit  Ang
<Atoms.UnitVectors
  7.7155852 0 0
 -3.8577926 6.6818927878 0
  0 0 20
Atoms.UnitVectors>

scf.XcType                GGA-PBE      # LDA|LSDA-CA|LSDA-PW|GGA-PBE
scf.SpinPolarization       Off         # On|Off
scf.energycutoff           250.0       # default=150 (Ry)
scf.maxIter                100         # default=40
scf.EigenvalueSolver      band         # Recursion|Cluster|Band
scf.Kgrid                  4 4 1       # means n1 x n2 x n3
scf.Mixing.Type            rmm-diisk   # Simple|Rmm-Diis|Gr-Pulay
scf.criterion             1.0e-8       # default=1.0e-6 (Hartree)
```

```
Unfolding.Electronic.Band      on
Unfolding.LowerBound         -12.0
Unfolding.UpperBound          8.0

Unfolding.Nkpoint              4

Unfolding.desired_totalnkpt    50

<Unfolding.ReferenceVectors
3.8577926 0 0
-1.9288963 3.3409463939 0
0 0 20
Unfolding.ReferenceVectors>

<Unfolding.kpoint
G 0 0 0
M 0.5 0 0
K 0.3333333 0.3333333 0
G 0 0 0
Unfolding.kpoint>

<Unfolding.Map
1 1
2 2
3 1
4 2
5 1
6 2
7 1
8 2
Unfolding.Map>
```

Free-standing silicene

Step 4: Plot the unfolded band structure



We have 0 ~ 1 in the weight

```
0.000000 -10.475841   0.9734492
0.000000 -8.449300   0.0185308
0.000000 -8.348240   0.0048797
0.000000 -8.022265   0.0074129
...
```

(Silicene.unfold_totup)

Free-standing silicene

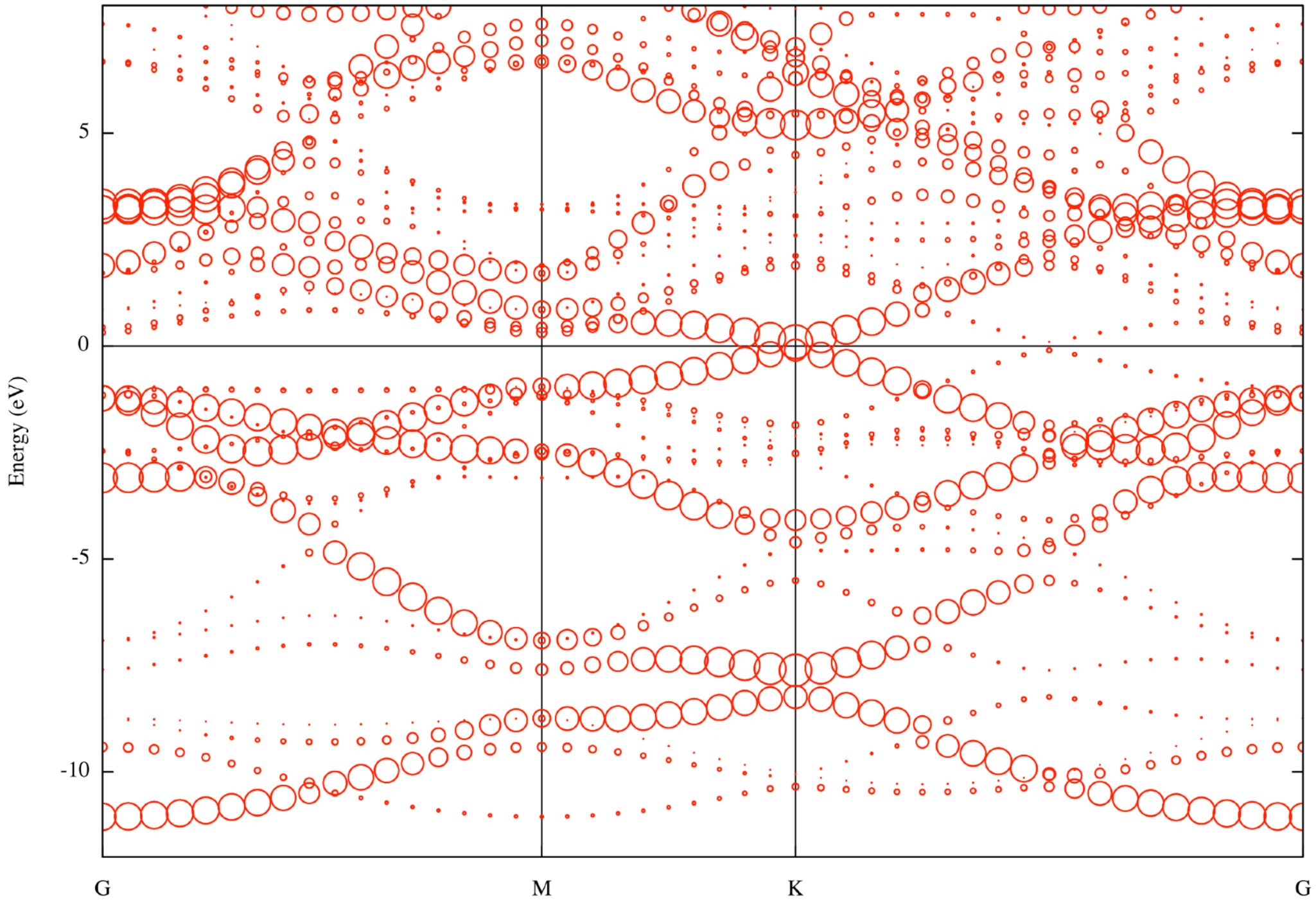
Step 5: How about change one Si atom to C atom as an impurity

```
System.CurrrentDirectory         ./
System.Name                      Silicene
DATA.PATH                /provide_your_path/DFT_DATA13

Species.Number       2
<Definition.of.Atomic.Species
  Si   Si7.0-s2p2d1  Si_PBE13
  C    C7.0-s2p2d1  C_PBE13
Definition.of.Atomic.Species>

Atoms.Number         8
Atoms.SpeciesAndCoordinates.Unit   FRAC # Ang|AU
<Atoms.SpeciesAndCoordinates
     1     Si     0.166667  0.333333  0.4871  2. 2.
     2     Si     0.333333  0.166667  0.5128  2. 2.
     3     C      0.666667  0.333333  0.4871  2. 2.
     4     Si     0.833333  0.166667  0.5128  2. 2.
     5     Si     0.166667  0.833333  0.4871  2. 2.
     6     Si     0.333333  0.666667  0.5128  2. 2.
     7     Si     0.666667  0.833333  0.4871  2. 2.
     8     Si     0.833333  0.666667  0.5128  2. 2.
Atoms.SpeciesAndCoordinates>

Atoms.UnitVectors.Unit  Ang
<Atoms.UnitVectors
  7.7155852 0 0
 -3.8577926 6.6818927878 0
  0 0 20
Atoms.UnitVectors>

scf.XcType                GGA-PBE      # LDA|LSDA-CA|LSDA-PW|GGA-PBE
scf.SpinPolarization       Off         # On|Off
scf.energycutoff           250.0       # default=150 (Ry)
scf.maxIter                100         # default=40
scf.EigenvalueSolver      band         # Recursion|Cluster|Band
scf.Kgrid                  4 4 1       # means n1 x n2 x n3
scf.Mixing.Type            rmm-diisk   # Simple|Rmm-Diis|Gr-Pulay
scf.criterion             1.0e-8       # default=1.0e-6 (Hartree)
```

```
Unfolding.Electronic.Band      on
Unfolding.LowerBound         -12.0
Unfolding.UpperBound          8.0

Unfolding.Nkpoint              4

Unfolding.desired_totalnkpt    50

<Unfolding.ReferenceVectors
3.8577926 0 0
-1.9288963 3.3409463939 0
0 0 20
Unfolding.ReferenceVectors>

<Unfolding.kpoint
G 0 0 0
M 0.5 0 0
K 0.3333333 0.3333333 0
G 0 0 0
Unfolding.kpoint>

<Unfolding.Map
1 1
2 2
3 3
4 2
5 1
6 2
7 1
8 2
Unfolding.Map>
```

Free-standing silicene

Step 6: Plot the unfolded band structure



Reference

◉ The theoretical discussions can be found in

Chi-Cheng Lee *et al*., J. Phys.: Condens. Matter **25**, 345501 (2013).
Chi-Cheng Lee *et al*., arXiv: 1707.02525 (2018), JPCM in press.

◉ All other details can be found in

http://www.openmx-square.org/openmx_man3.8/openmx.html

- Unfolding method for band structures
  - Analysis of band structures
  - Unfolding of band structures
  - The origin of the reference unit cell
  - Intensity map of unfolded spectral weight
  - In case of non-collinear DFT calculations
  - Examples

For example, you can also define the origin of the conceptual unit cell by yourself via "<Unfolding.ReferenceOrigin".