Band unfolding calculations compared with ARPES experiments: Examples of $Ni_{1/3}TiS_2$ and twisted-bilayer graphene

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PRB **95**, 085420 (2017).

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Contents

Part I, Great success of the band unfolding technique

- Brief review of the band unfolding technique
- Direct comparison with ARPES results for TiS_2

Part II, Novel band unfolding technique for multi-periodicity materials

- Serious problem in the unfolding scheme
- New unfolding scheme and its application to twisted-bilayer graphene (tBLG)

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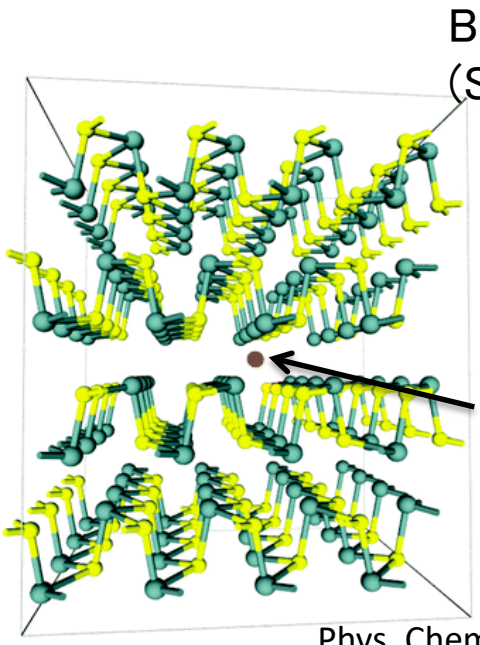
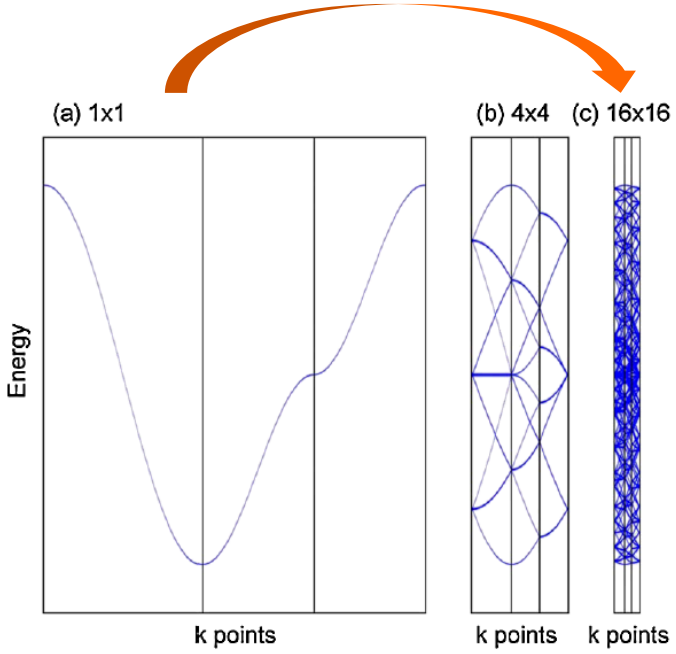
Why band unfolding?

- Impurity doping, substitution, and so on are technologically important.
- To treat imperfection in crystal, supercell scheme is often used.

Very complicated!



Phys. Chem. Chem. Phys., **16**, 26176



Unfolding needed!

- The electronic bands are folded into the small supercell BZ. (Bad point)

Two-dimensional one-band tight-binding model

PRL **104**, 216401 (2010).

Brief review of band unfolding method

Start from the supercell(SC) Green's func. $\hat{G}^{\mathrm{SC}}(z) = \sum_{n\mathbf{k}_{\mathrm{SC}}} \frac{\left|\Psi^{\mathrm{SC}}_{n\mathbf{k}_{\mathrm{SC}}}\right\rangle \left\langle\Psi^{\mathrm{SC}}_{n\mathbf{k}_{\mathrm{SC}}}\right|}{z - \epsilon_{n\mathbf{k}_{\mathrm{SC}}}}$

Projection to the primitive-cell(PC) BZ $\hat{G}_{\mathrm{unfold}}(\mathbf{k}, z) = \hat{P}^{\dagger}_{\mathbf{k}} \hat{G}^{\mathrm{SC}}(z) \hat{P}_{\mathbf{k}}$

$\hat{P}_{\mathbf{k}} = \sum_n \left|\Psi^{\mathrm{PC}}_{n\mathbf{k}}\right\rangle \left\langle\Psi^{\mathrm{PC}}_{n\mathbf{k}}\right|$

Energy spectrum $A(\mathbf{k}, \epsilon) = -\frac{1}{\pi}\mathrm{Im}\left[\mathrm{tr}\left(\hat{G}_{\mathrm{unfold}}(\mathbf{k}, \epsilon + i\delta)\right)\right]$

Notable things:

- Assumption that the system has single PC (Single PC representation)
- The unfolded bands have the periodicity of the primitive cell

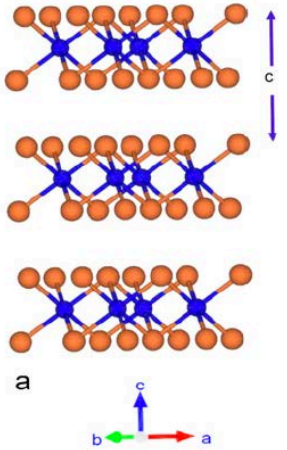
It is easily understood by considering the relation: $\hat{P}_{\mathbf{k}} = \hat{P}_{\mathbf{k}+\mathbf{G}}$

$\because \quad \hat{P}_{\mathbf{k}} = \sum_n \left|\Psi^{\mathrm{PC}}_{n\mathbf{k}}\right\rangle \left\langle\Psi^{\mathrm{PC}}_{n\mathbf{k}}\right|$

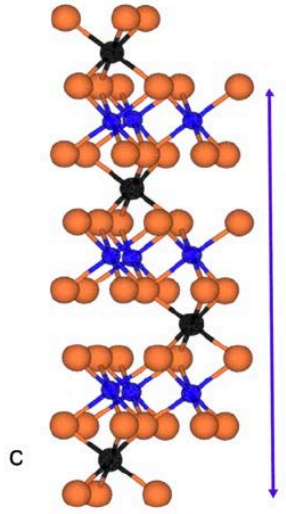
$\hat{G}_{\mathrm{unfold}}(\mathbf{k}, z) = \hat{P}^{\dagger}_{\mathbf{k}} \hat{G}^{\mathrm{SC}}(z) \hat{P}_{\mathbf{k}}$ has the periodicity of the PC.

Direct comparison with ARPES measurements for whole BZ

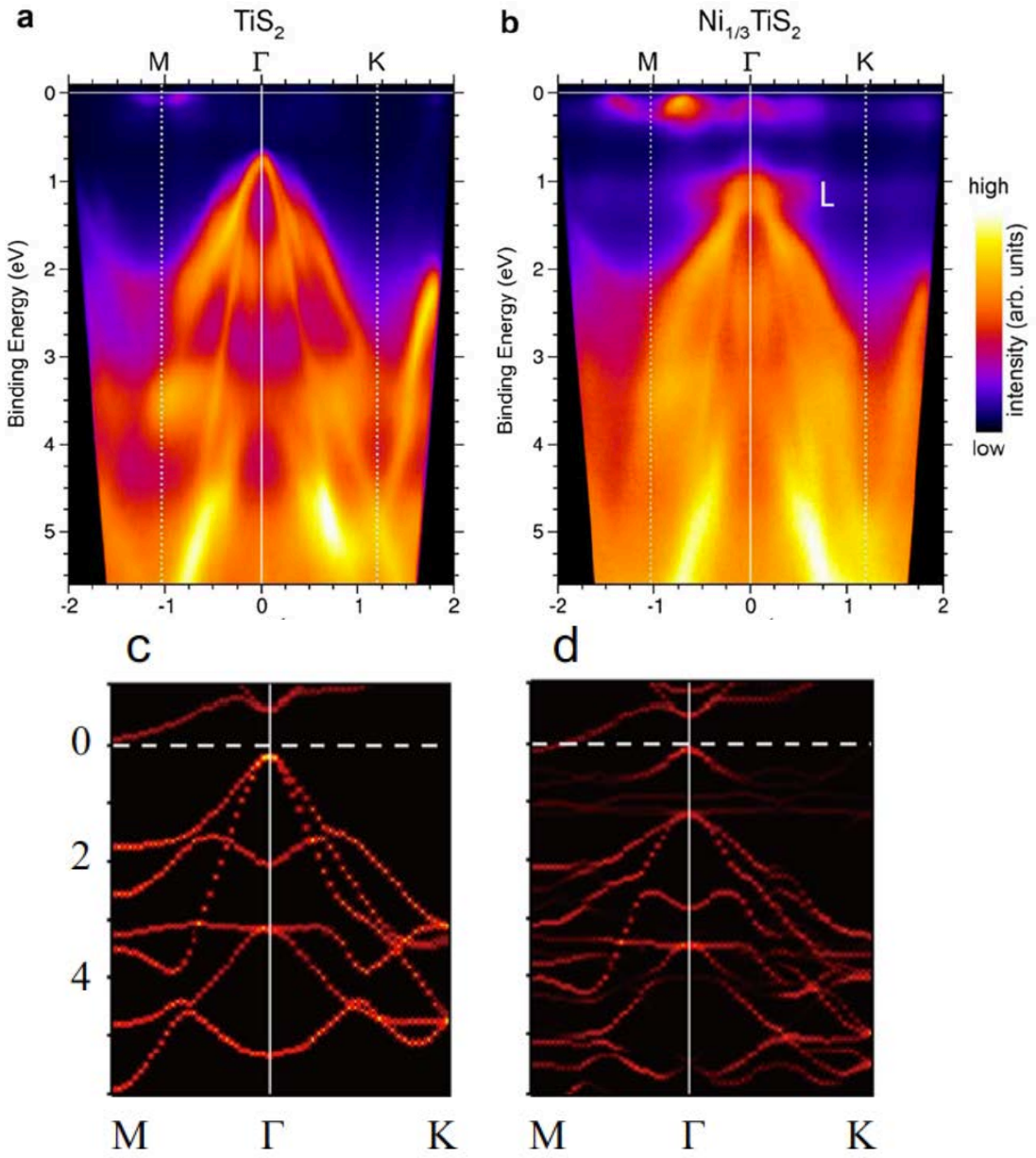
Target: Layered semiconductor



TiS_2



$Ni_{1/3}TiS_2$



Excellent agreement!

New J. Phys, **17** 083010 (2015).

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- Practical application to TiS_2

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Our results clearly show the usefulness of the band unfolding method

It's a happy story so far…

Contents

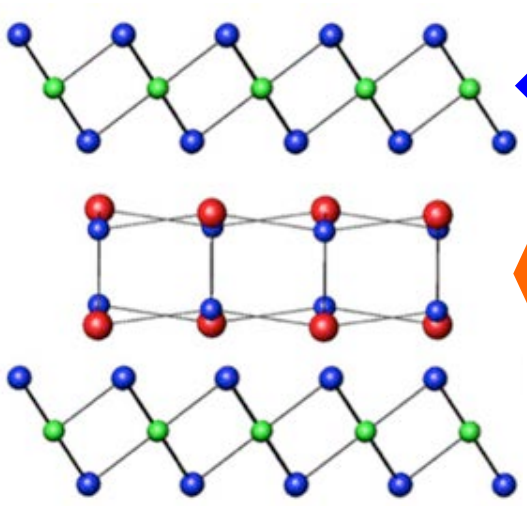
Serious problem for multi-periodicity matter

Breakdown of the assumption of the conventional unfolding scheme:

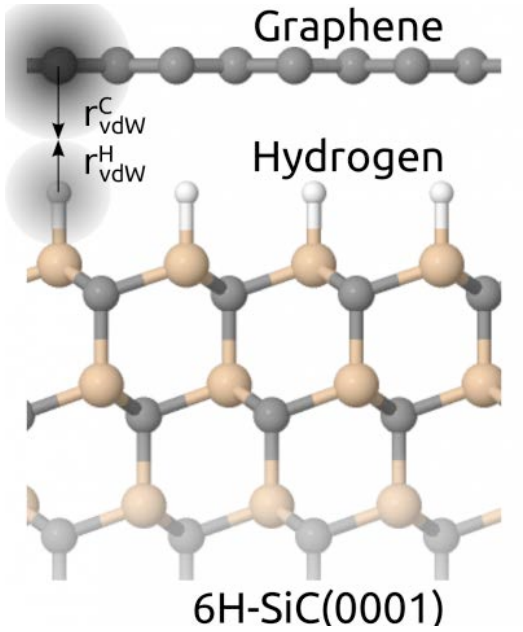
- The assumption of single PC representation collapses.

Misfit compounds

Thin films on a substrate



Each consisting material has each periodicity.



Materials, **8** 2000 (2015).

PRL, **114** 106804 (2015).

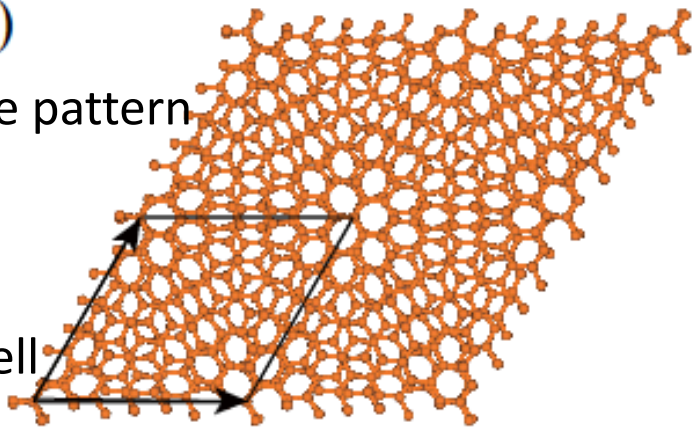
Multi-periodicity materials are ubiquitous!

What happens, if we apply the unfolding scheme to multi-periodicity matter?

Target: tBLG

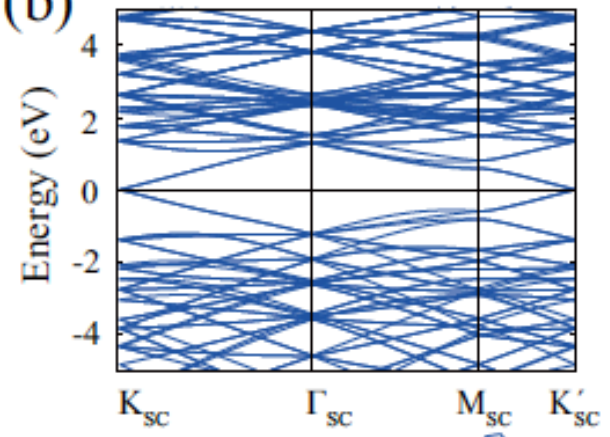
(a)

Moire pattern

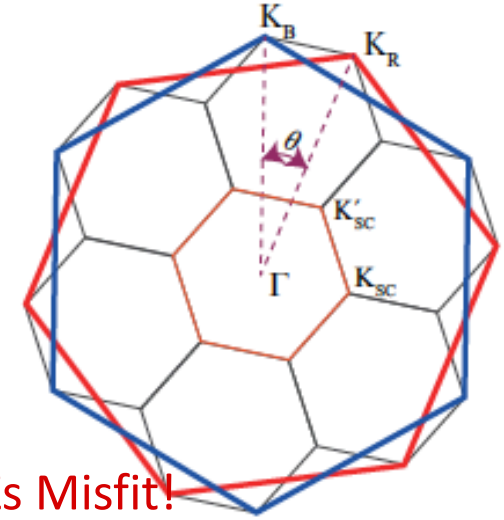


Simulation cell

(b)

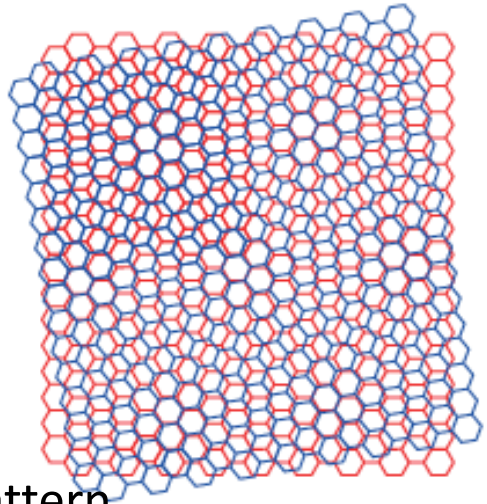


(c)



Two PC BZs Misfit!

(d)

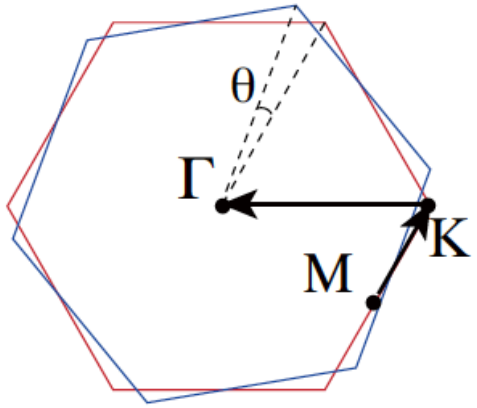


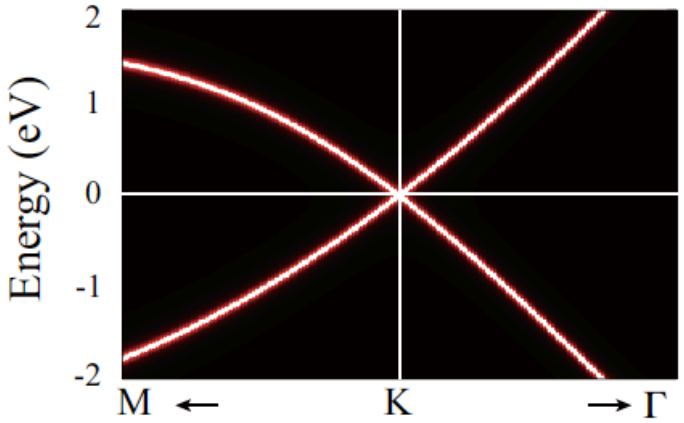
Moire pattern

tBLG is a good example of multi-periodicity materials

What happens, if we apply the unfolding scheme to multi-periodicity matter?

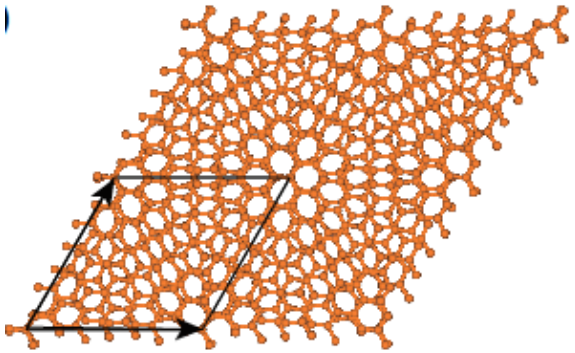
Most prominent result is no interlayer-interacting tBLG. (∵The band is much simpler)
For the no interacting system, Two independent unfolding calculations were performed for each monolayer.

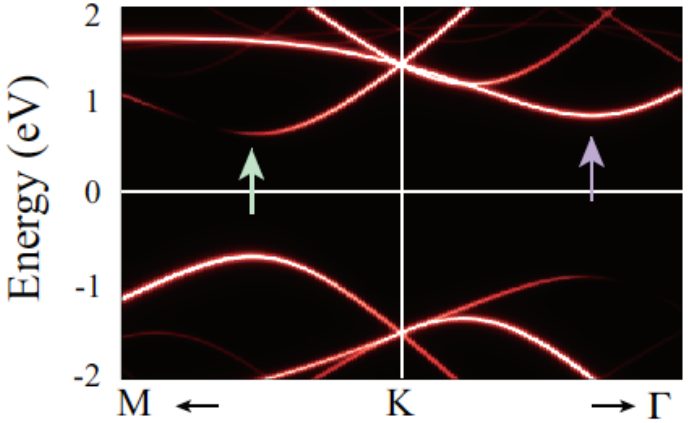




Unfolded bands of the upper monolayer graphene onto the upper layer PC BZ.

Uniform intensity





Unfolded bands of the lower monolayer graphene onto the upper layer PC BZ.

Nonuniform intensity

Ghost bands appear!

New band unfolding method for multi-periodicity materials
- Multi-space representation-

Start from the supercell(SC) Green's func. $\hat{G}^{\mathrm{SC}}(z) = \sum_{n\mathbf{k}_{\mathrm{SC}}} \frac{\left|\Psi^{\mathrm{SC}}_{n\mathbf{k}_{\mathrm{SC}}}\right\rangle \left\langle\Psi^{\mathrm{SC}}_{n\mathbf{k}_{\mathrm{SC}}}\right|}{z - \epsilon_{n\mathbf{k}_{\mathrm{SC}}}}$

Projection to the primitive-cell(PC) BZ $\hat{G}_{\mathrm{unfold}}(\mathbf{k}, z) = \hat{P}^{\dagger}_{\mathbf{k}} \hat{G}^{\mathrm{SC}}(z) \hat{P}_{\mathbf{k}}$

$$\hat{P}_{\mathbf{k}} = \hat{P}^{(\mathrm{A})}_{\mathbf{k}} + \hat{P}^{(\mathrm{B})}_{\mathbf{k}}$$

Decomposition to two subsystems, (A) and (B), of each consisting material

$$\hat{P}^{(i)}_{\mathbf{k}} = \sum_n \left|\Psi^{\mathrm{PC}(i)}_{n\mathbf{k}}\right\rangle \left\langle\Psi^{\mathrm{PC}(i)}_{n\mathbf{k}}\right|$$

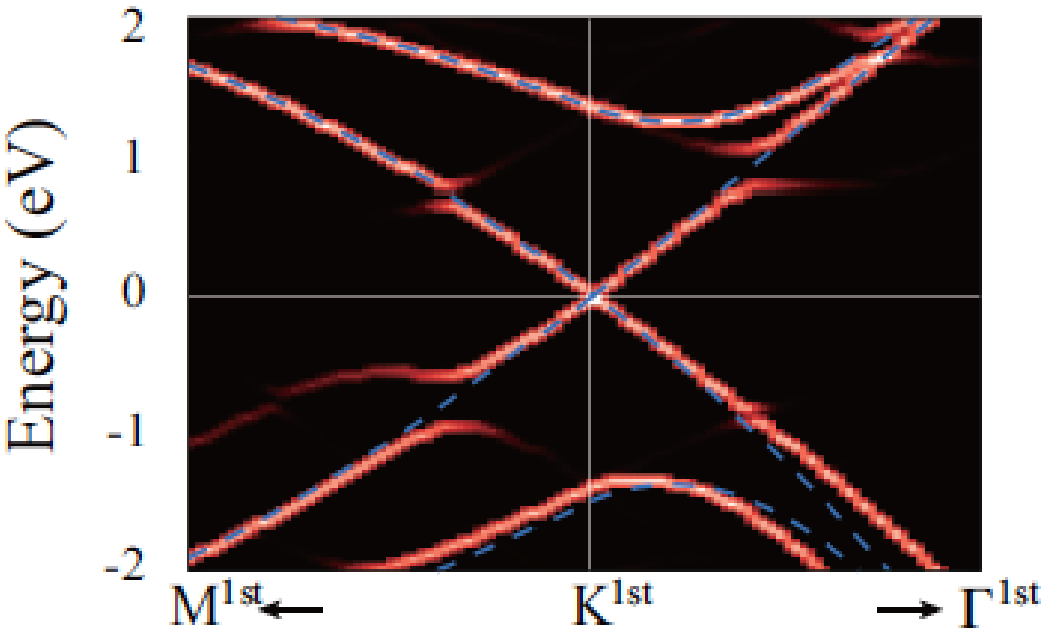
$$(i = A \text{ or } B)$$

Each projection operator shows each periodicity

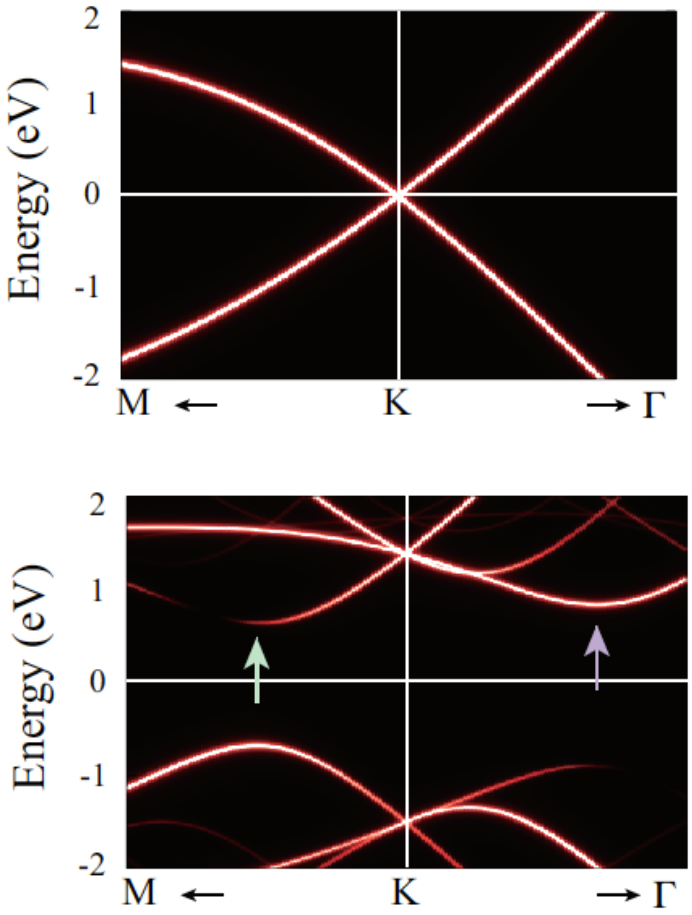
Energy spectrum $A(\mathbf{k}, \epsilon) = -\frac{1}{\pi}\mathrm{Im}\left[\mathrm{tr}\left(\hat{G}_{\mathrm{unfold}}(\mathbf{k}, \epsilon + i\delta)\right)\right]$

Novel band unfolding method for tBLG

New unfolding



Conventional unfolding



Ghost bands are much clear than the real ones

Summary

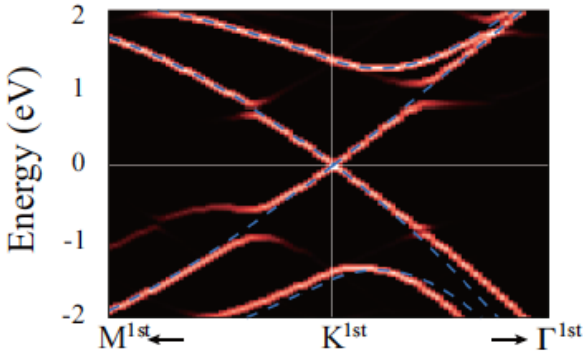
Part I, Great success of the band unfolding technique

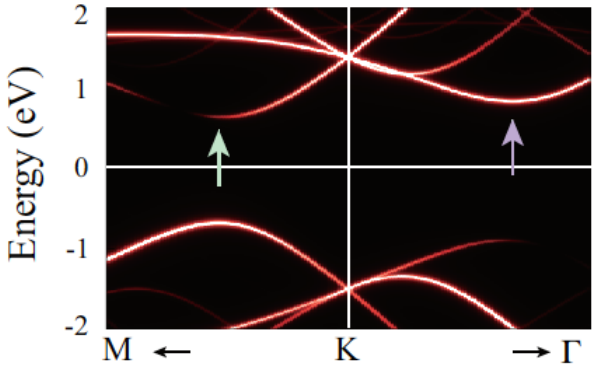
- Unfolded bands give good agreement with the ARPES measurements for TiS_2 and $Ni_{1/3}TiS_2$

New J. Phys, **17** 083010 (2015).

Part II, Novel band unfolding technique for multi-periodicity materials

- Ghost bands appear for multi-periodicity materials by the conventional unfolding scheme
- New unfolding scheme proposed and clarified its validity





PRB **95**, 085420 (2017). arXiv: 1706.05921