Development of the photoemission simulator SPADExp and its application to an ARPES study of nodal line materials

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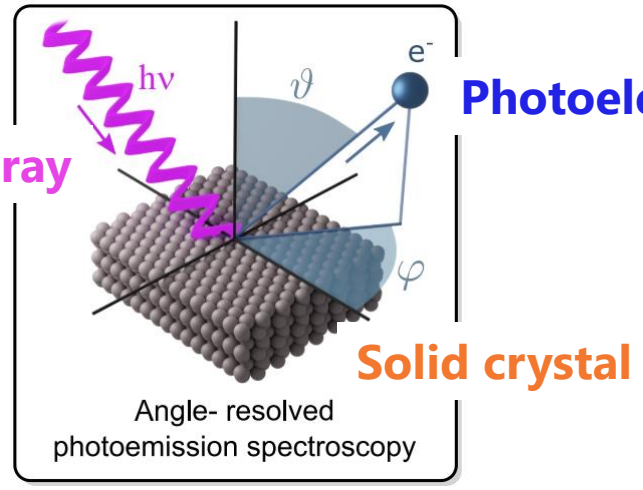
2 Angle-resolved photoemission spectroscopy (ARPES)

Observation of the **band dispersion** using the **photoelectric effect**



UV, X-ray

Photoelectron

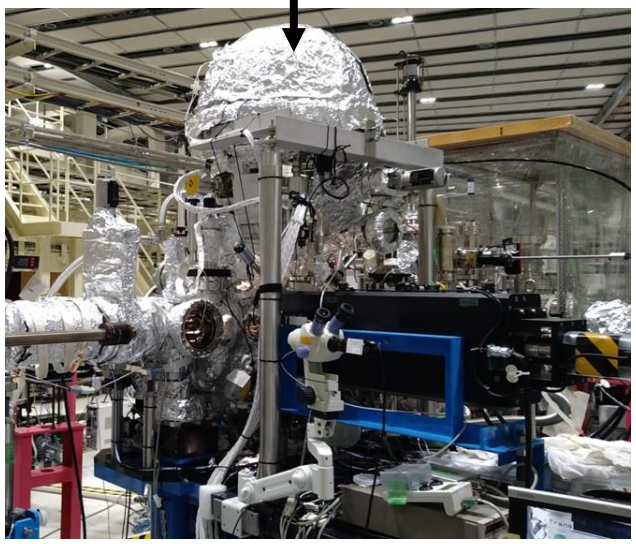
Solid crystal

Schematic of the photoemission process

J. A. Sobota *et al.*, Rev. Mod. Phys. **93**, 025006 (2021).

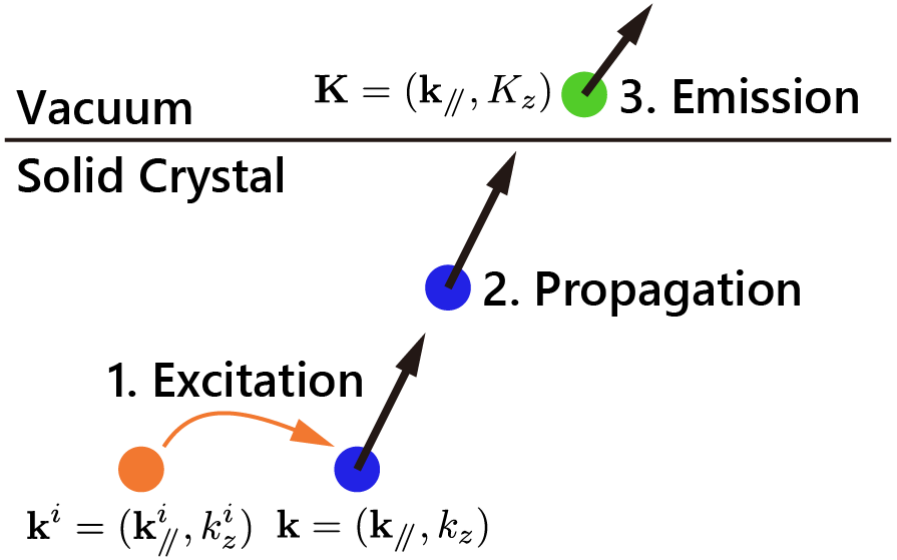
Photoelectron analyzer

X-ray→



Synchrotron ARPES endstation @SPring-8 BL25SU

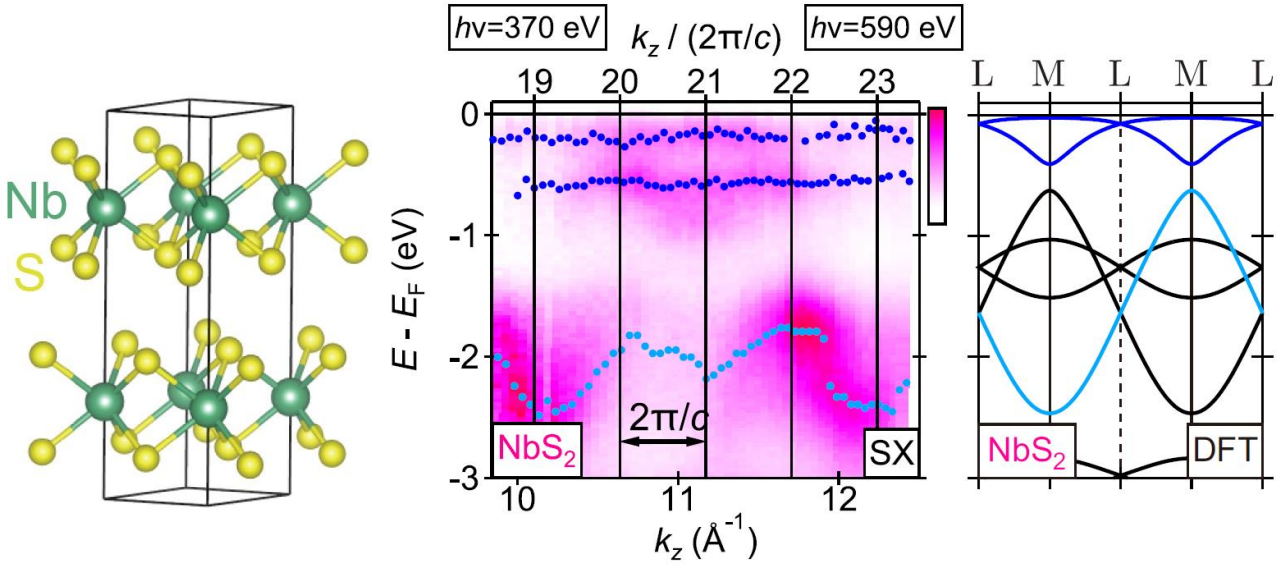
3 Photoemission 3-step model



- In-plane momentum ($\mathbf{k}_{//}$) is conserved in the whole process
- Out-of-plane momentum (k_z) is conserved in the crystal
- Photoelectron energy E_{kin} = Ground state energy + $h\nu$
  - $E_{\mathrm{kin}} = \frac{\hbar^2 k^2}{2m} + V_0 = \frac{\hbar^2 K^2}{2m}$ determines k_z ($V_0 < 0$ is a parameter)

4 Starting point of our study

- **Bulk band degeneration** at the *L* point was not observed.
- **Surface sensitivity** (a few nm)
  → Momentum selection rule along *z* may not be rigorous?

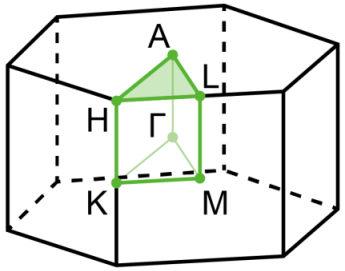


ARPES spectra of the transition metal dichalcogenide 2*H*-NbS_2

H. Tanaka, S. Okazaki *et al.*, Phys. Rev. B **105**, L121102 (2022).

5 Overview of this talk

- Development of the photoemission simulator SPADExp
  [H. Tanaka *et al.*, J. Electron Spectrosc. **264**, 147297 (2023)]
  - Tight-binding model (previous studies) → DFT with localized basis sets



- Application to an ARPES study of nodal line materials
  [H. Tanaka *et al.*, *under review*. (arXiv:2308.00999)]
  - Line node at the *AL* path protected by the 2_1 screw rotation symmetry

- Photoemission calculations beyond the free-electron approximation
  [H. Tanaka *et al.*, *in preparation*.]
  - Wave function calculation using the Kohn-Sham potential

Development of the photoemission simulator SPADExp

H. Tanaka *et al.*, J. Electron Spectrosc. **264**, 147297 (2023)
https://github.com/Hiroaki-Tanaka-0606/SPADExp

7 Photoemission probability

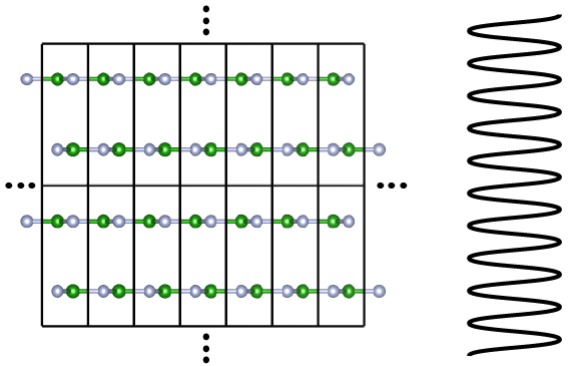
Electric field

Fermi's golden rule $$w_{fi} = \frac{2\pi}{\hbar}\left|\langle\psi_f|H_{\mathrm{int}}|\psi_i\rangle\right|^2 \delta(E_f - E_i - h\nu)$$

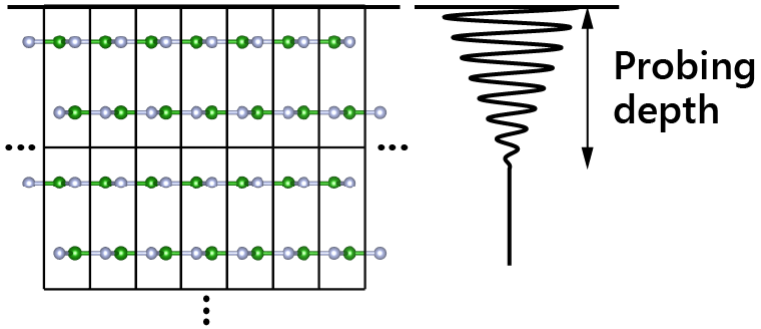
Energy conservation

Final state Initial state

Final state: **Free-electron approximation** is applied



Bulk system: Plane wave



Slab system: Plane wave × Decay

8 Calculation formulae

Initial state: Linear combination of (pseudo-)atomic orbitals

$$\psi_i(\mathbf{r}) = \frac{1}{\sqrt{N}} \sum_n^N e^{\mathbf{i}\mathbf{R}_n \cdot \mathbf{k}} \sum_{i\alpha\sigma} c_{\mu,i\alpha}^{\sigma(\mathbf{k})} \phi_{i\alpha}(\mathbf{r} - \tau_i - \mathbf{R}_n)|\sigma\rangle$$

$\mathbf{R}_n$=lattice vector, τ_i=atom position, ϕ=orbital wave function, α=(plm)

Final state: Partial wave expansion at each atom position

$$\psi_f(\mathbf{r}) = 4\pi e^{i\mathbf{k}\cdot\mathbf{t}_i} \sum_{lm} i^l Y_{lm}^*(\hat{\mathbf{k}}) Y_{lm}(\theta,\varphi) j_l(kr)$$

Electric field: Dipole approximation

$$H_{\text{int}} \sim \mathbf{r}\cdot\mathbf{e} \sim \sum_j e_j r Y_{1j}(\theta,\phi)$$

e=electric field vector

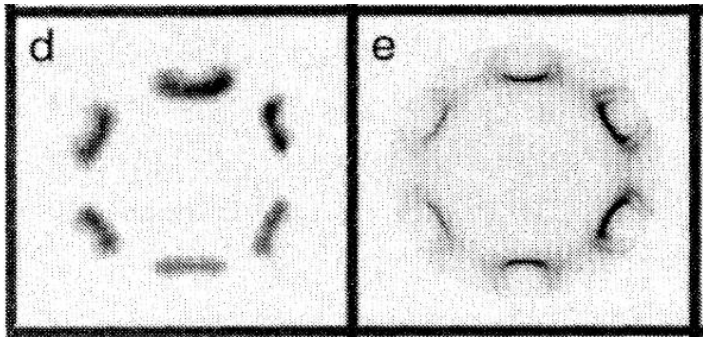
Integration over angles → excitation **selection rule**

$$l_f = l_i \pm 1, m_f = m_i + j$$

9 Previous study of graphene

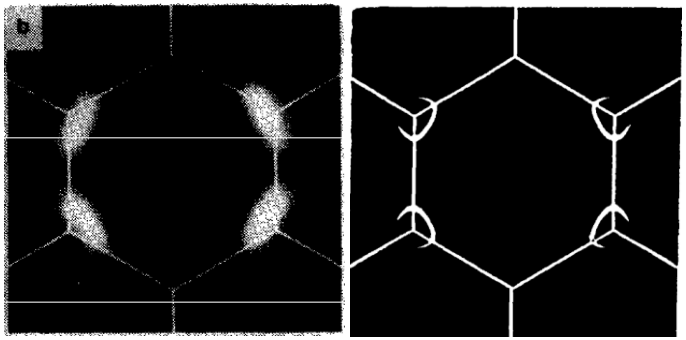
- A tight-binding model (carbon p_z orbital) is used for initial states
- Graphene Dirac cones are excited only within the Brillouin zone

ARPES **Simulation**



50 eV polarization not specified, E_F-2.1 eV [E. L. Shirley *et al.*, Phys. Rev. B **51**, 13614 (1995)]

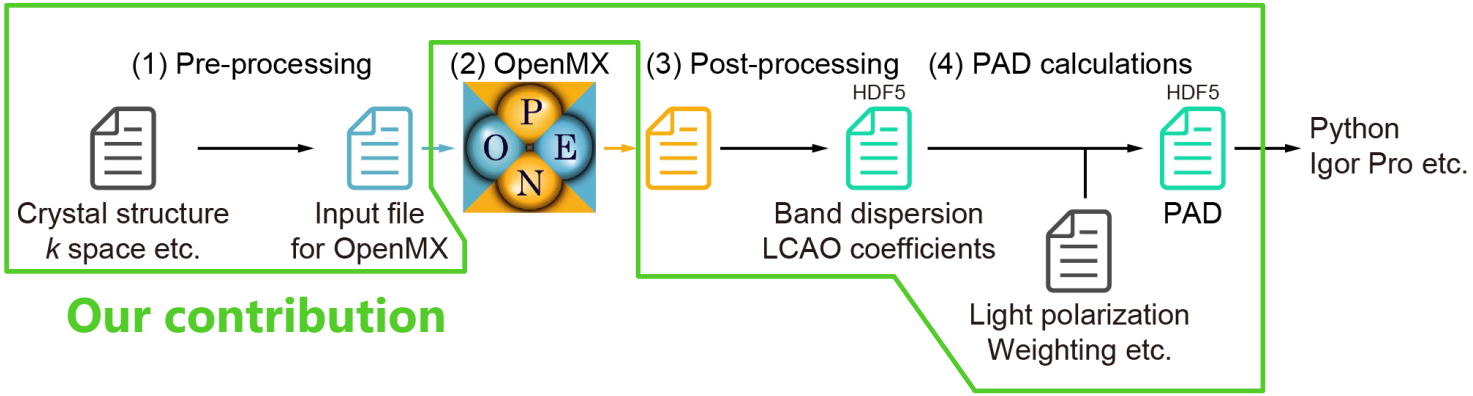
ARPES **Simulation**



54 eV horizontal linear polarization, E_F-1.7 eV [H. Nishimoto *et al.*, Solid State Commn. **98**, 671 (1996)]

10 Our software

Simulator of Photoemission Angular Distribution for Experiments (SPADExp)

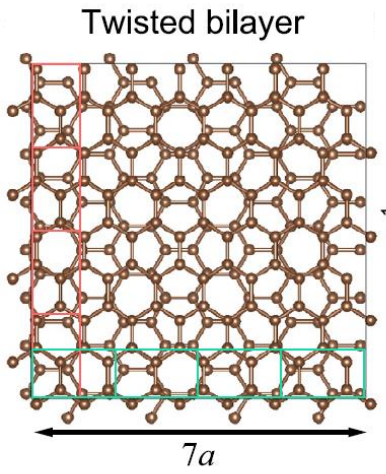


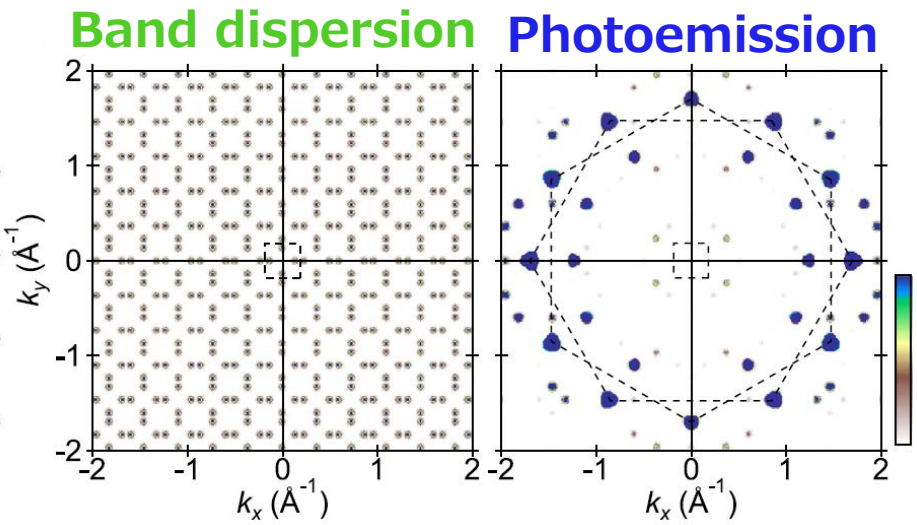
- Use MO.fileout / MO.Nkpoint / <MO.kpoints> to extract LCAO coefficients
- C++ / OpenMP parallelization / Lapack / HDF5
- Slab calculations were performed on ISSP Supercomputer System B

11 Calculation example

30°-twisted bilayer graphene: ARPES spectra with 12-fold symmetry

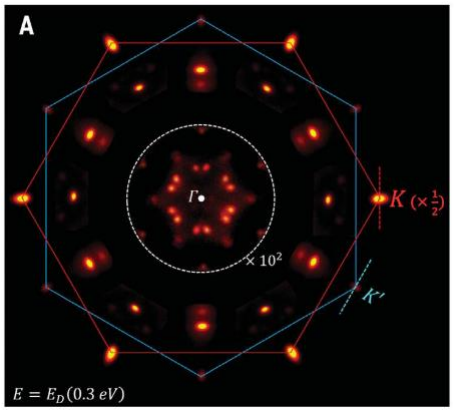
- Approximating the quasicrystal by a supercell (224 carbon atoms)





21 eV, horizontal polarization
constant-energy maps at the Fermi level

ARPES



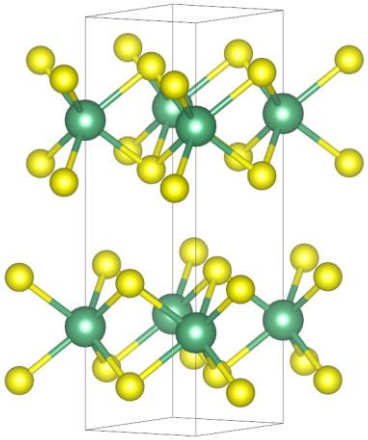
21 eV no polarization

Ahn *et al.*, Science **361**, 782 (2018).

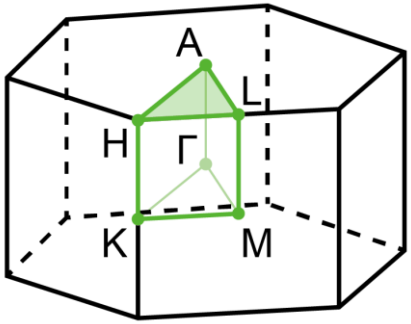
Application of SPADExp to an ARPES study of nodal line materials

Hiroaki Tanaka *et al.*, *under review.*
(arXiv:2308.00999)

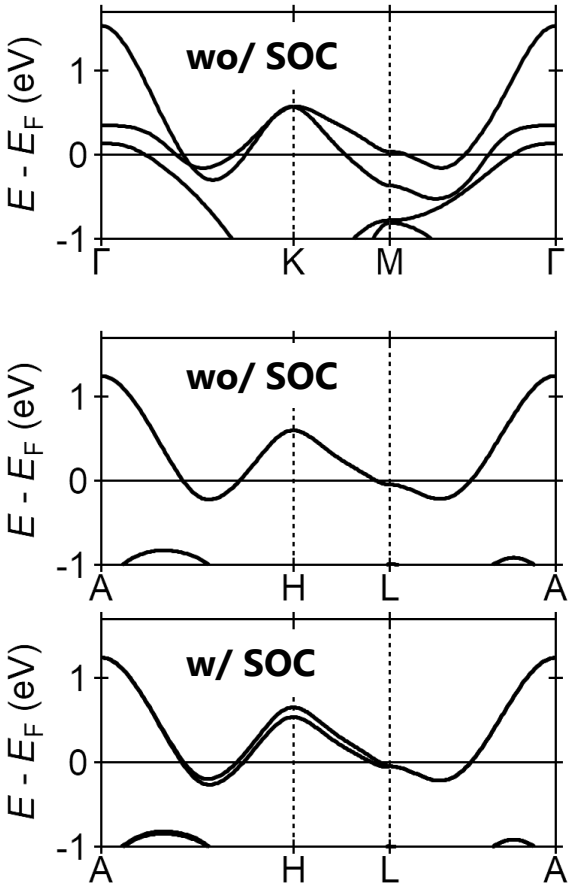
13 Bulk nodal lines in 2*H*-NbS_2



2*H*-NbS_2



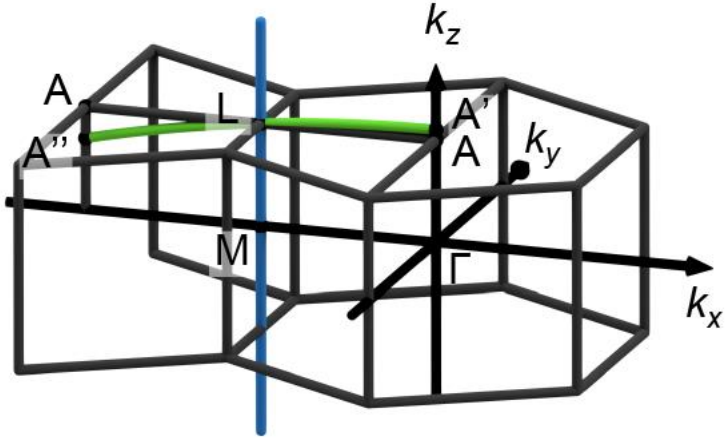
Hexagonal Brillouin zone



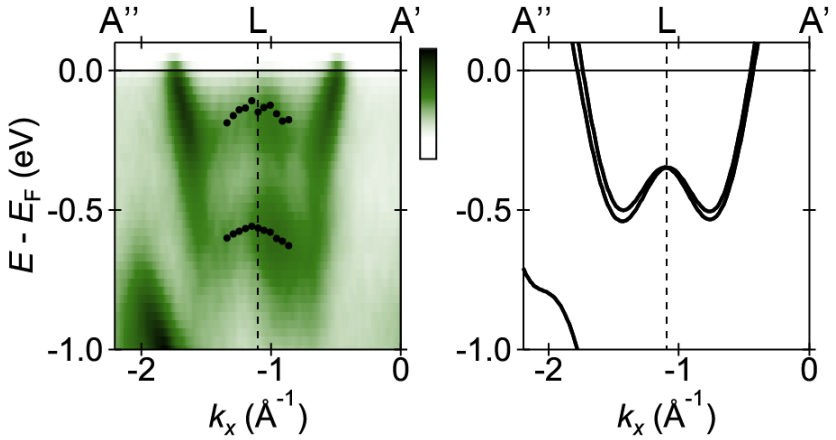
Bulk band dispersion of 2*H*-NbS_2

- Without SOC, a **nodal plane** on $k_z = \frac{\pi}{c}$
  - Due to 2_1 screw and time-reversal
- With SOC, a **nodal line** remains on *AL*

14 ARPES experiments

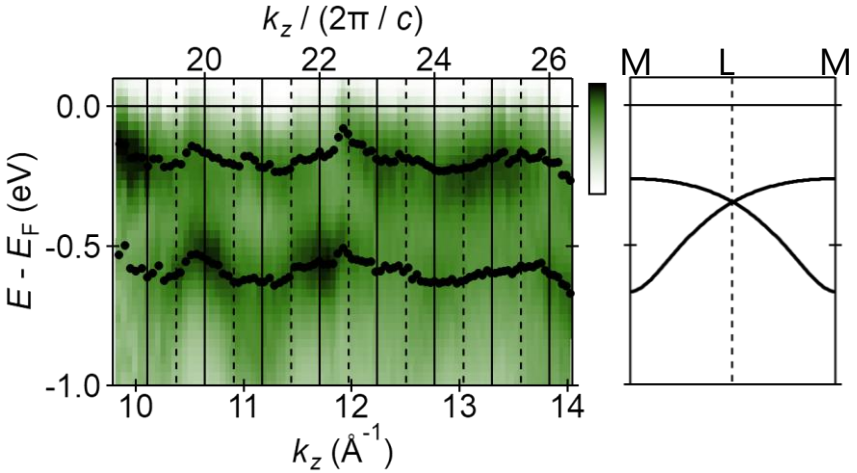


Hexagonal Brillouin zone



Band dispersion around *L* ($h\nu = 525$ eV)

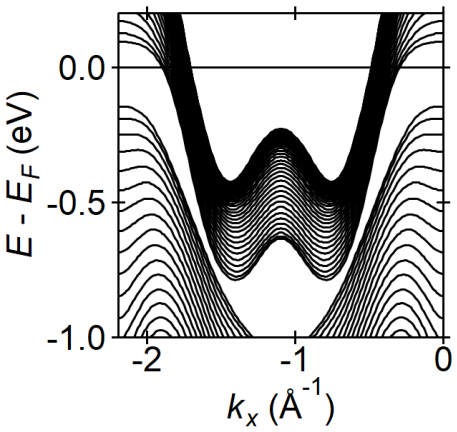
- Two bands were **gapped** (0.5 eV)
- All the observed *L* points had gapped dispersions
- 2_1 screw rotation symmetry is broken in the near-surface electronic structure?



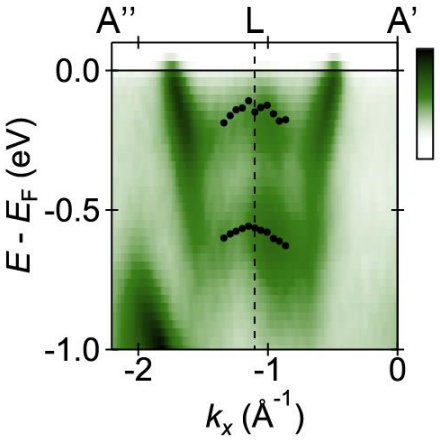
Band dispersion along *ML*
($h\nu = 370 \sim 750$ eV)

15 Slab calculations

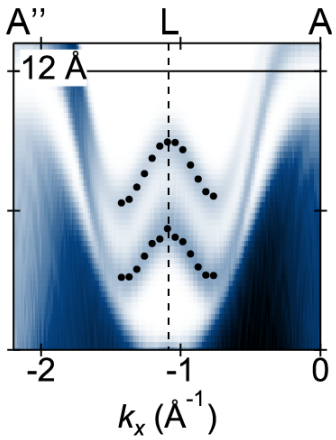
- 20 bilayers (120 atoms, 1000 electrons) for no spin calculations
- 10 bilayers for noncollinear calculations
- Reproduced gapped spectra by photoemission intensity calculations
  - Probing depth parameter ~ 1 nm



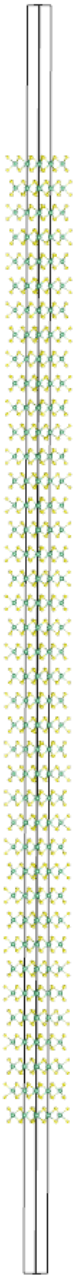
Slab band dispersion (spin off)



ARPES spectra



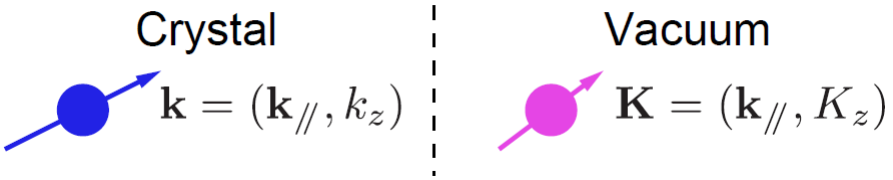
Simulation spectra



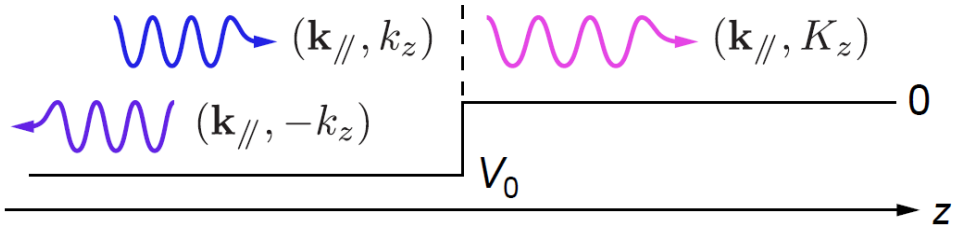
20 bilayers
2*H*-NbS_2

Future work and summary

Beyond the free-electron approximation

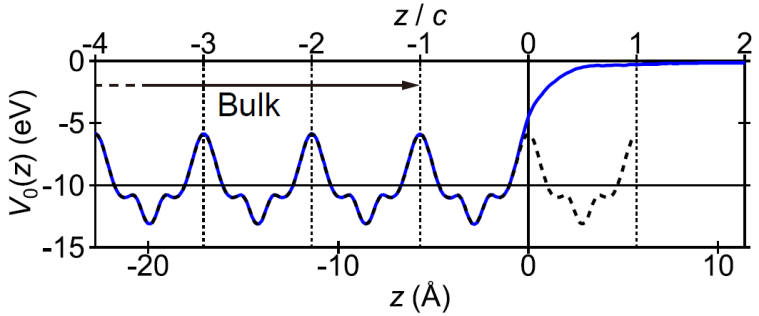


Three-step model / Free-electron approximation



Precise description of photoelectron wave function

→ Photoelectron wave function from the local Kohn-Sham potential



Local Kohn-Sham potential of 1*T*-TiS_2 slab

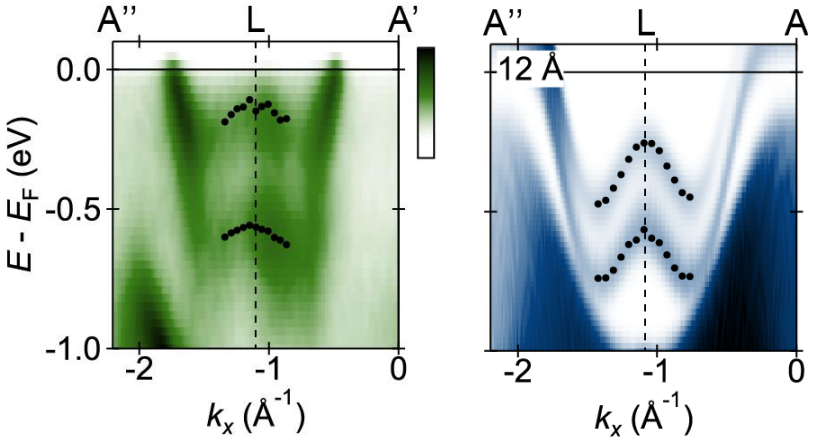
Summary

Photoemission simulator SPADExp

- Directly use the ground state wave function exported from OpenMX
- Able to handle large slab systems and surface electronic structure

ARPES study of nodal line materials

- Bulk band degeneracy due to nonsymmorphic symmetry was not absent
- Photoemission intensity calculations considering surface sensitivity successfully reproduced gapped spectra



ARPES Simulation

References

20 References

Photoelectric effect, photoemission spectroscopy

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Band degeneration due to symmetry

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- C. J. Bradley and A. P. Cracknell, *The Mathematical Theory of Symmetry in Solids* (Oxford, 2010). (Textbook in English)
- Supplementary Section E in G. Chang *et al.*, Nat. Mater. **17**, 978 (2018). (Example of 2_1 screw rotation symmetry)
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21 References

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- A. F. Starace, Phys. Rev. A **3**, 1242 (1971); A. J. Read and R. J. Needs, Phys. Rev. B **44**, 13071 (1991). (position operator vs. momentum operator when nonlocal potential exists)

Photoemission intensity calculations (tight-binding model)

- E. L. Shirley *et al.*, Phys. Rev. B **51**, 13614 (1995); H. Nishimoto *et al.*, Solid State Commn. **98**, 671 (1996). (graphene)

First-principles calculations

- P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964); W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965). (Density functional theory)
- T. Ozaki, Phys. Rev. B **67**, 155108 (2003). (Theory for OpenMX)
- https://www.openmx-square.org/ (OpenMX software)

22 References

SPADExp

- H. Tanaka, K. Kuroda, and T. Matsushita, J. Electron Spectrosc. **264**, 147297 (2023). (Paper about the software development)
- https://github.com/Hiroaki-Tanaka-0606/SPADExp (Repository)

Inelastic mean free path

- H. Kanter, Phys. Rev. B **1**, 522 (1970). (Electron beam transmittance)
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- M. P. Seah and W. A. Dench, Surf. Interf. Anal. **1**, 2 (1979). (so-called the universal curve)
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23 References

ARPES

- H. Tanaka, S. Okazaki *et al.*, Phys. Rev. B **105**, L121102 (2022). (NbS_2 & 3*d*-intercalated system)
- D. Huang *et al.*, Phys. Rev. B **105**, 245145 (2022) (NbS_2)
- H. Tanaka *et al.*, *under review*. (arXiv:2308.00999) (NbS_2 & *h*BN)