Optical Spectroscopy of 2-Dimensional Antiferromagnetic Materials

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Seoul, Korea

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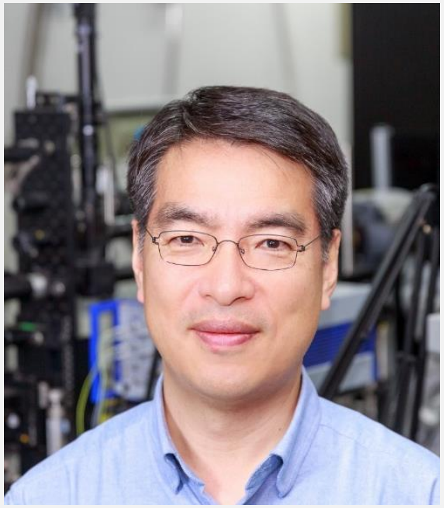

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Zur Theorie der Metalle.

I. Eigenwerte und Eigenfunktionen der linearen Atomkette.

Von **H. Bethe** in Rom.

(Eingegangen am 17. Juni 1931.)

No magnetic ordering in 1 dimension.

Z. Physik **71,** 205 (1931).

$$H = -J_{XY} \sum_{j\delta} \left(S_j^x S_{j+\delta}^x + S_j^y S_{j+\delta}^y \right) - J_I \sum_{j\delta} S_j^z S_{j+\delta}^z$$

Magnetic Hamiltonian

VOLUME 17, NUMBER 22 PHYSICAL REVIEW LETTERS 28 NOVEMBER 1966

ABSENCE OF FERROMAGNETISM OR ANTIFERROMAGNETISM IN ONE- OR TWO-DIMENSIONAL ISOTROPIC HEISENBERG MODELS*

N. D. Mermin† and H. Wagner‡

PHYSICAL REVIEW VOLUME 65, NUMBERS 3 AND 4 FEBRUARY 1 AND 15, 1944

Crystal Statistics. I. A Two-Dimensional Model with an Order-Disorder Transition

LARS ONSAGER
Sterling Chemistry Laboratory, Yale University, New Haven, Connecticut

(Received October 4, 1943)

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Ferromagnetism in 2-Dimensional materials

LETTER

doi:10.1038/nature22060

Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals

Cheng Gong[1]*, Lin Li[2]*, Zhenglu Li[3,4]*, Huiwen Ji[5], Alex Stern[2], Yang Xia[1], Ting Cao[3,4], Wei Bao[1], Chenzhe Wang[1], Yuan Wang[1,4], Z. Q. Qiu[3], R. J. Cava[5], Steven G. Louie[3,4], Jing Xia[2] & Xiang Zhang[1,4]

$Cr_2Ge_2Te_6$: Ferromagnetic ordering down to 2L

Nature **546**, 265 (2017).

LETTER

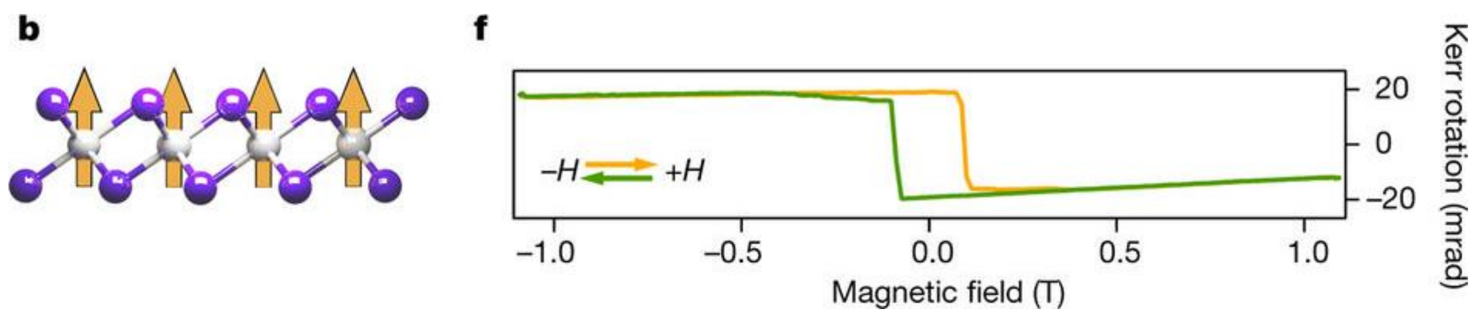
doi:10.1038/nature22391

Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit

Bevin Huang[1]*, Genevieve Clark[2]*, Efrén Navarro-Moratalla[3]*, Dahlia R. Klein[3], Ran Cheng[4], Kyle L. Seyler[1], Ding Zhong[1], Emma Schmidgall[1], Michael A. McGuire[5], David H. Cobden[1], Wang Yao[6], Di Xiao[4], Pablo Jarillo-Herrero[3] & Xiaodong Xu[1,2]

CrI_3: Ferromagnetic ordering down to 1L

Nature **546**, 270 (2017).

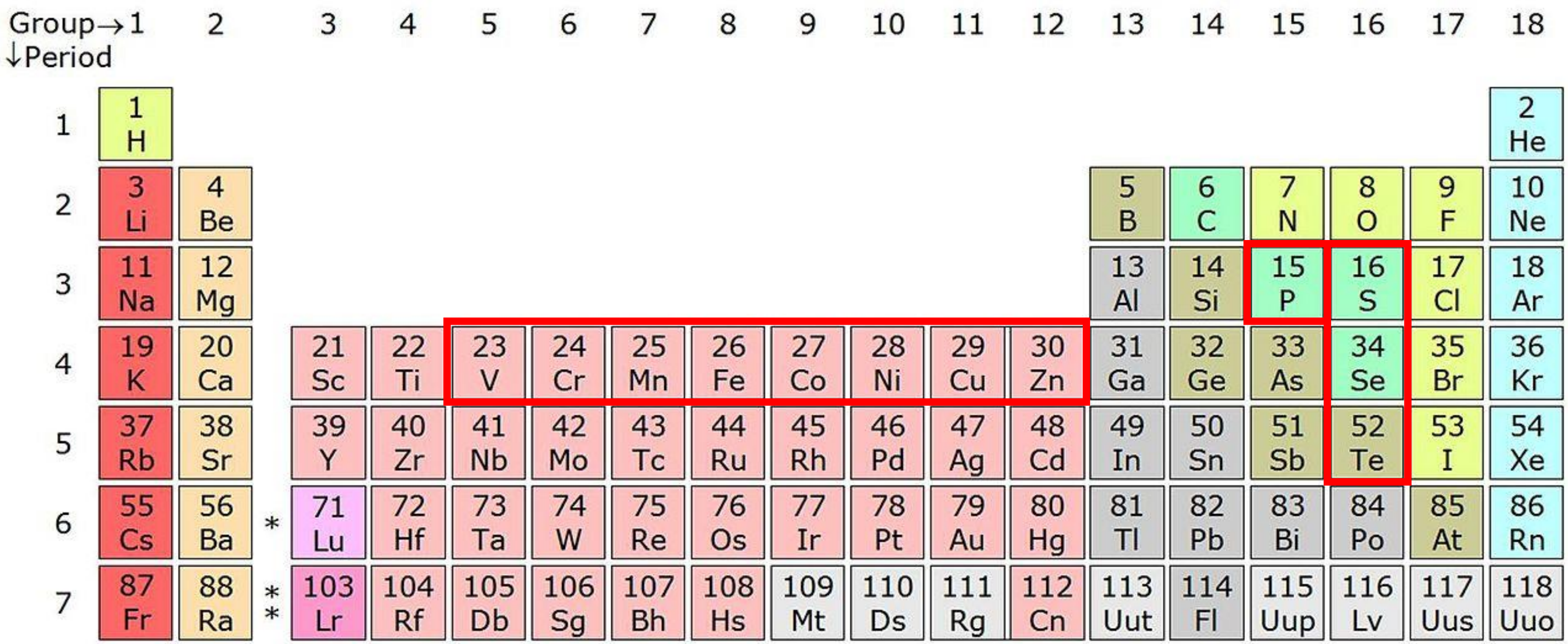


Antiferromagnetism in 2-Dimensional materials

- Experimental Difficulties
  - No net magnetic moment
  - Conventional tools (e.g. neutron scattering) are inadequate due to extremely small sample volume.

- Indirect measurements of magnetic ordering are needed.
  → Raman scattering

Transition metal phosphorus trichalcogenides ($TMPX_3$)



https://en.wikipedia.org/wiki/Periodic_table

- Ternary compound semiconductor with layered structure
- Magnetic phase transitions (Antiferromagnetism for TM=Mn, Fe, Co, Ni)

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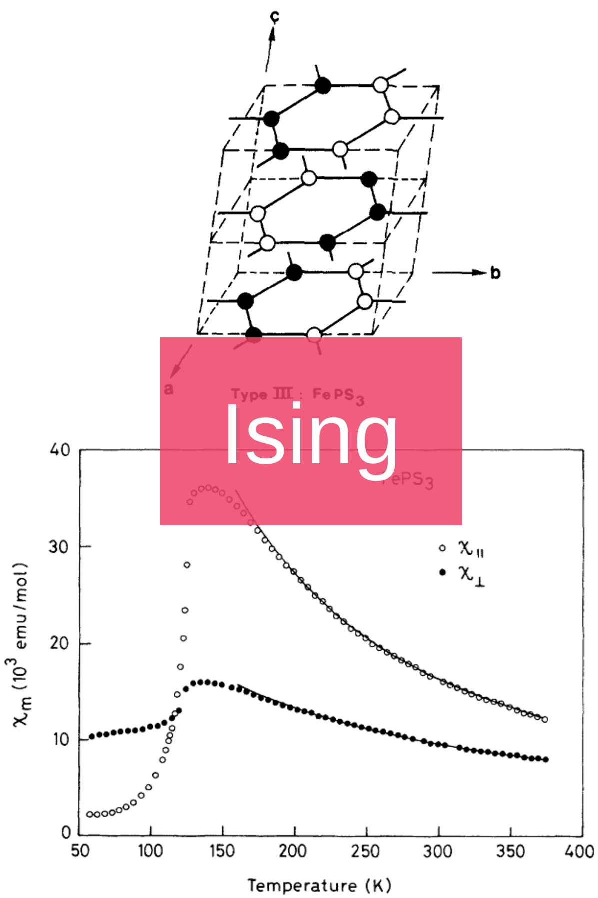
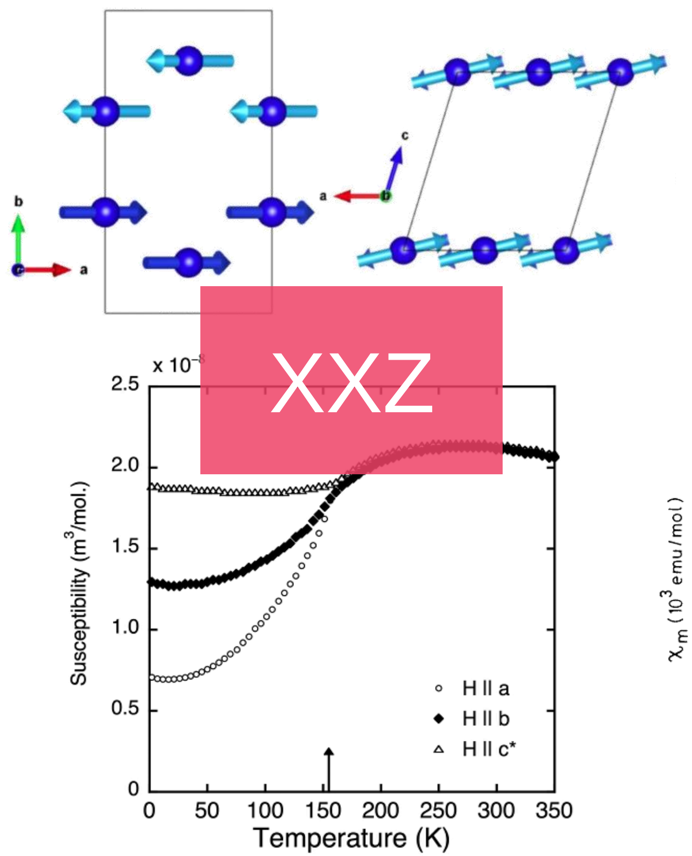
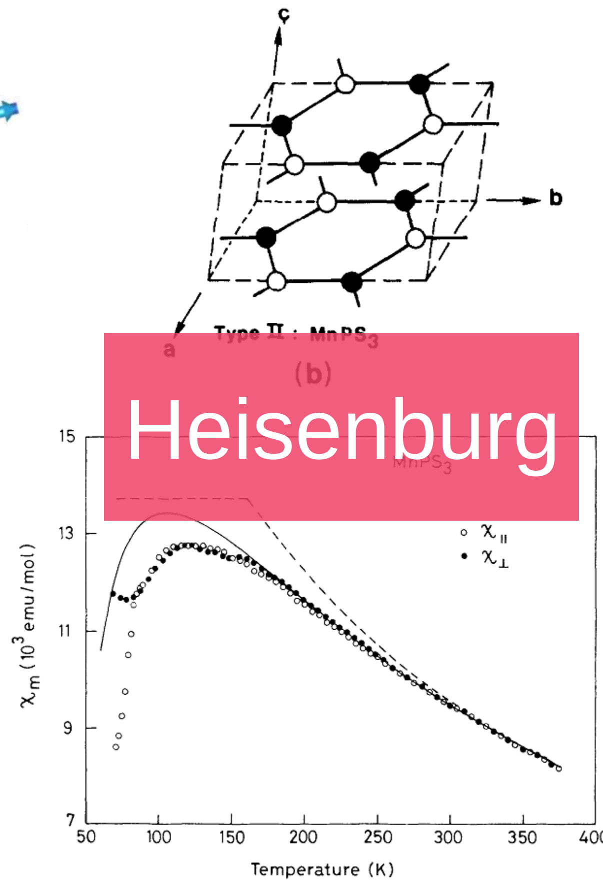
Magnetism depending on TM elements







R. Brec, *Solid State Ionics* **22**, 3-30 (1986). P. A. Joy *et al.*, *Phys. Rev. B* **46**, 5425 (1992).
A. R. Wildes *et al.*, *Phys. Rev. B* **92**, 224408 (2015).

Contents

- $FePS_3$ (Ising type antiferromagnet)

  Nano Letters **16**, 7433 (2016).

- $NiPS_3$ (XXZ-type antiferromagnet)

  Nature Communications **10**, 345 (2019).

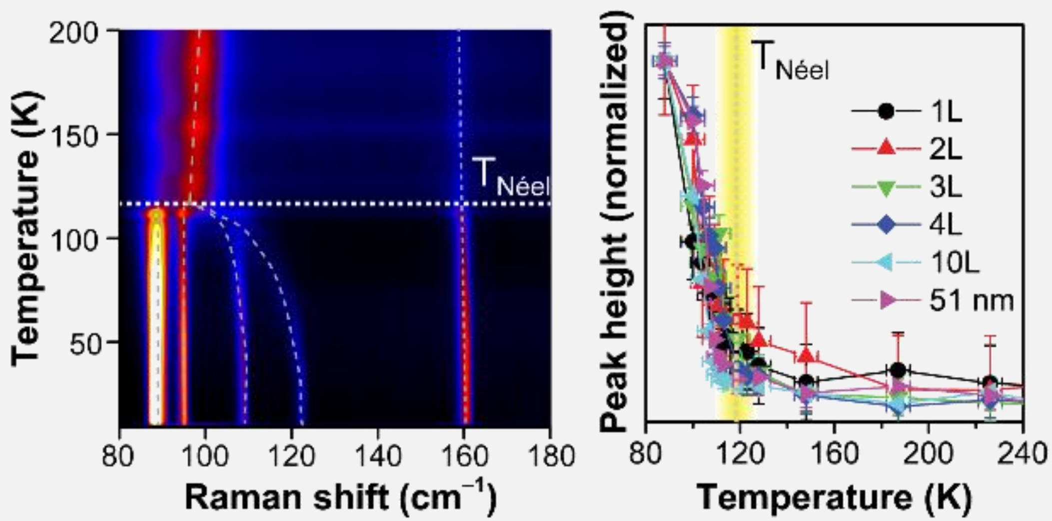
- $MnPS_3$ (Heisenberg-type antiferromagnet)

  2D Materials **6**, 041001 (2019).
  Current Applied Physics, submitted.

- Observation of coherent many-body exciton in $NiPS_3$

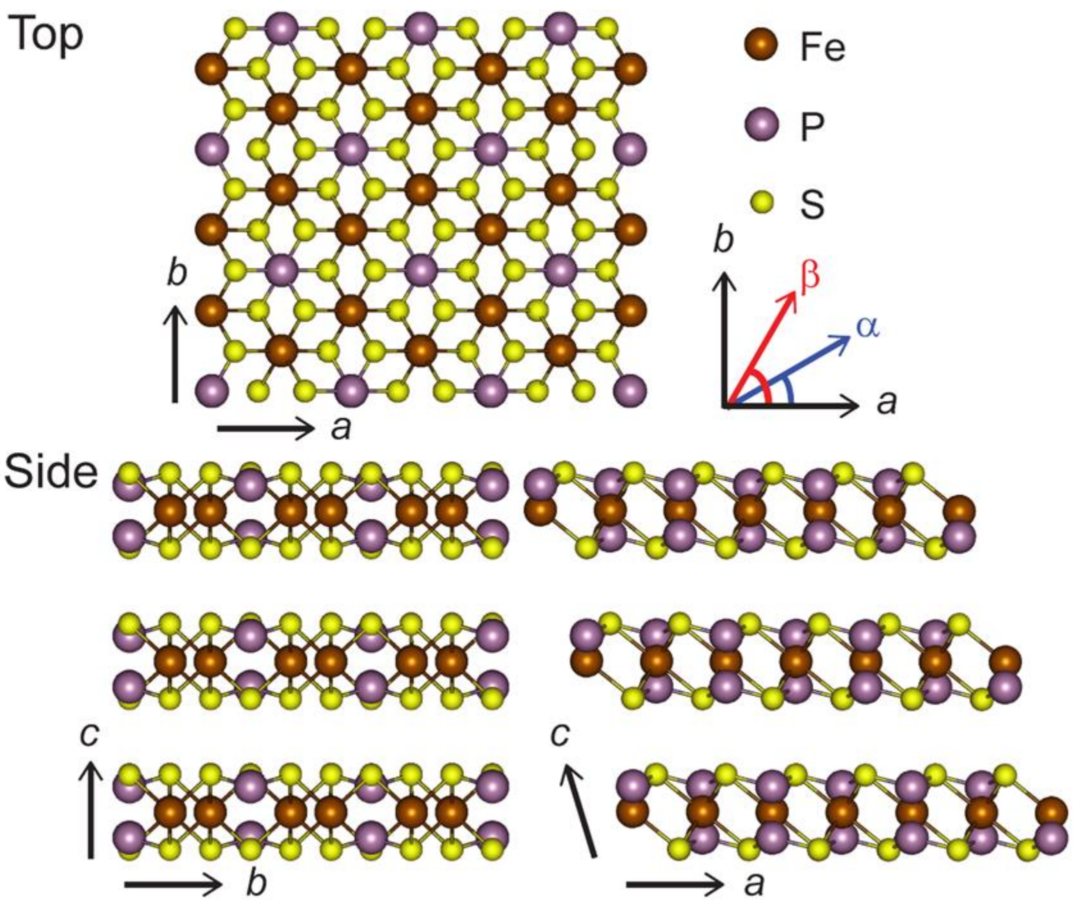
  Nature **583**, 785 (2020).

1. Ising type antiferromagnet: $FePS_3$



Nano Letters **16**, 7433 (2016)
Also, Wang *et al.*, *2D Mater*. (2016).

Iron phosphorus trisulfide ($FePS_3$)



- Honeycomb arrangement of Fe atoms surrounded by six S atoms.
- These S atoms are connected to two P atoms above and below Fe plane.

- Bulk : monoclinic (C_{2h})
- Monolayer : hexagonal (D_{3d})

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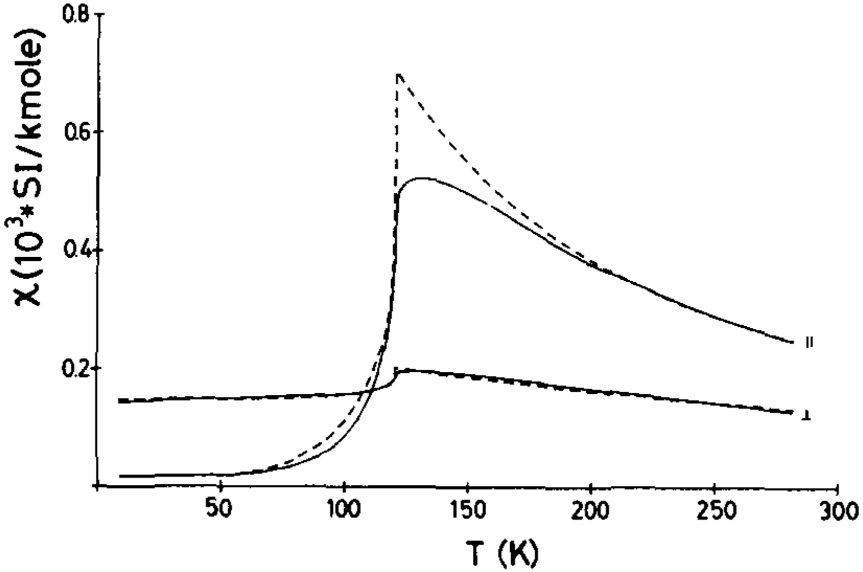
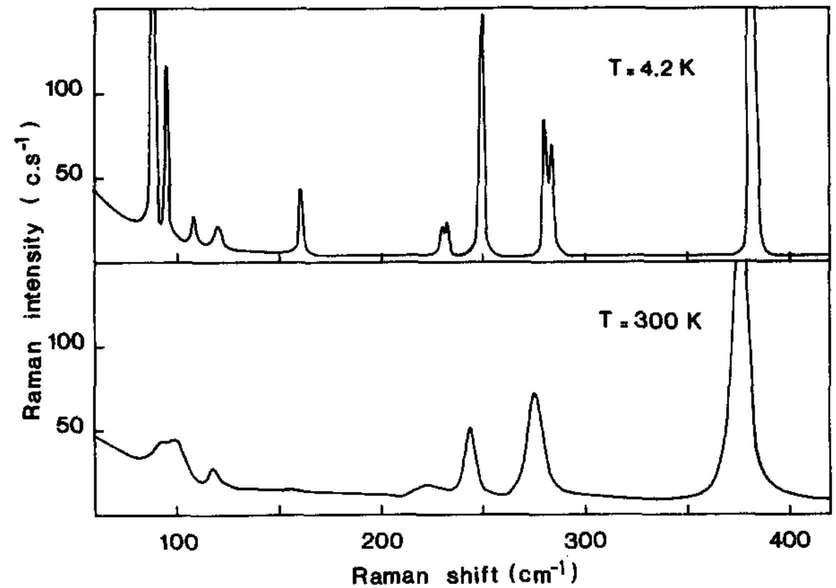
Iron phosphorus trisulfide ($FePS_3$)

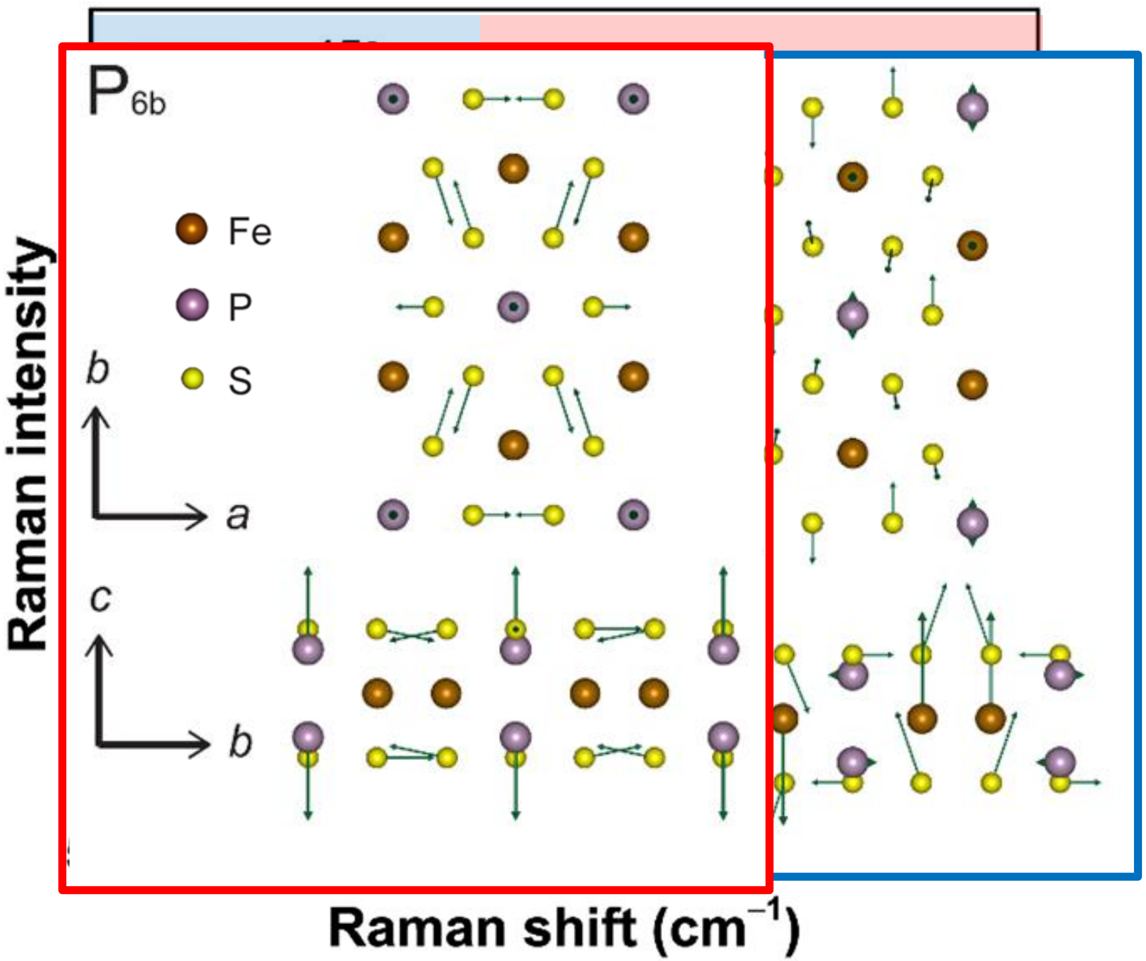


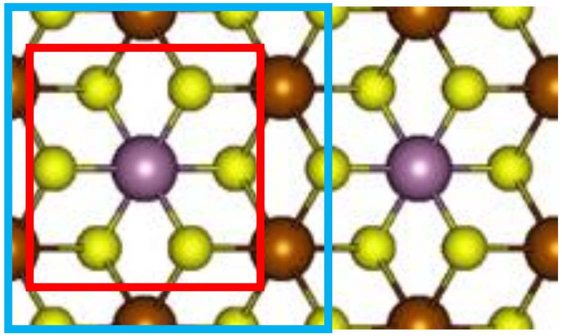


- Antiferromagnetic phase transition at 118K in bulk
- Dramatic changes of Raman spectra due to magnetic ordering
- Raman spectroscopy can be used as a probe for magnetic ordering.

Jernberg *et al.*, J. Magn. Magn. Mater. **46**, 178 (1984)
Scagliotti et al., Solid State Commun. **54**, 291 (1985)

Raman modes of bulk $FePS_3$





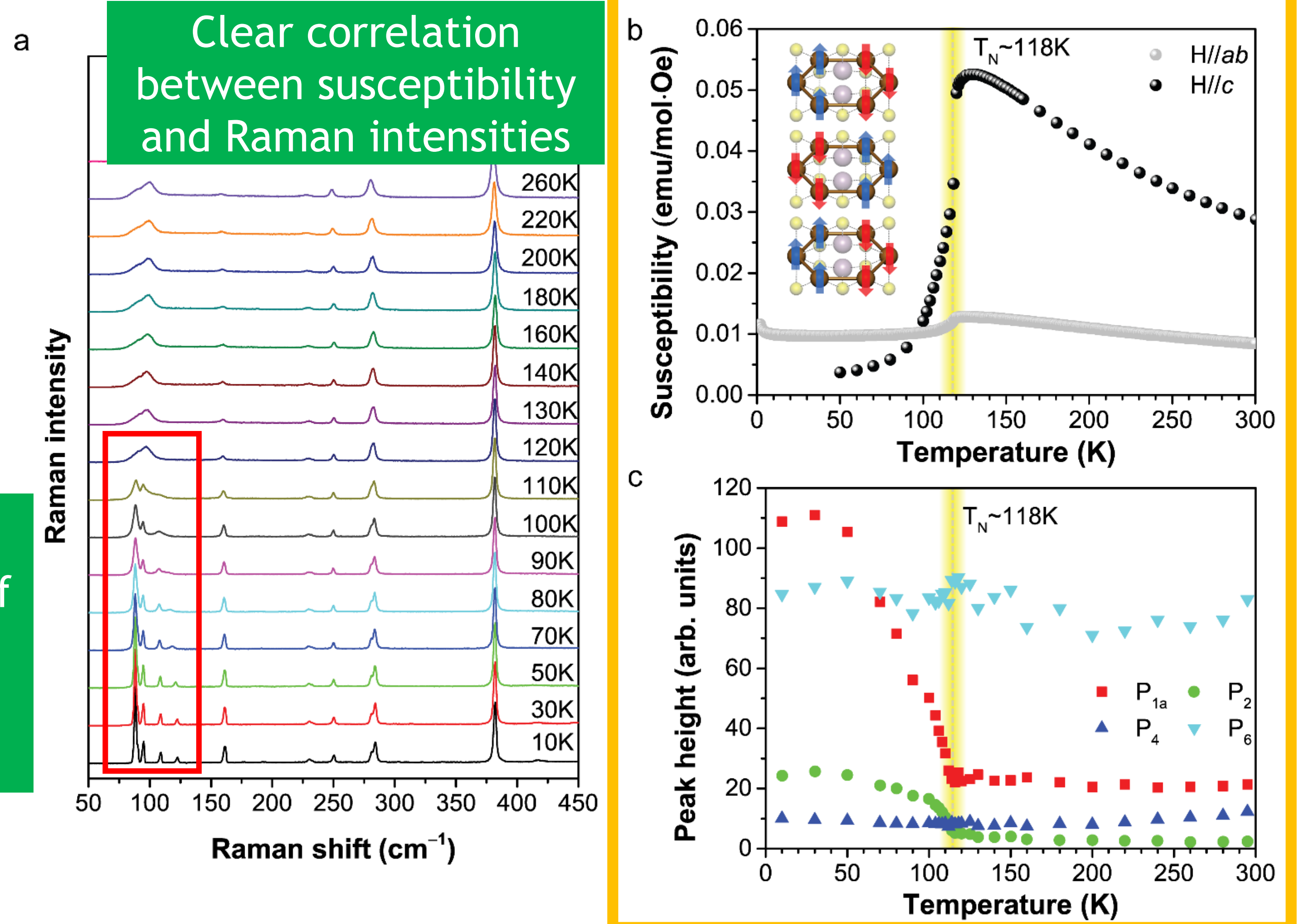
P_3 to P_6 : Vibrations of P and S atoms are dominant.

P_1 and P_2 : Vibrations including Fe atoms

Temperature dependence of Raman modes in bulk $FePS_3$

Clear correlation between susceptibility and Raman intensities

Dramatic changes of P_1 after phase transition

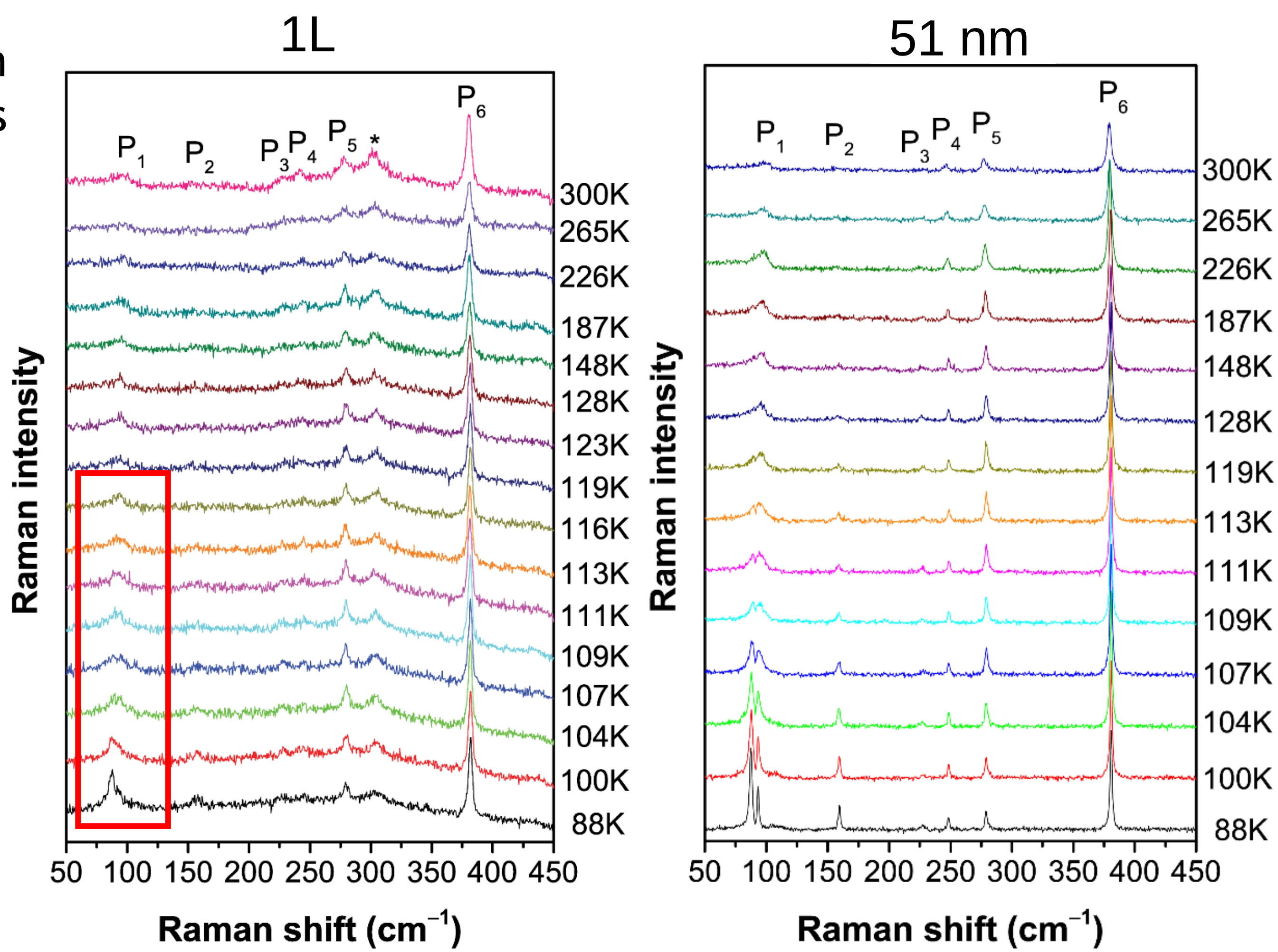


cf. PRB **101**, 064416 (2020).

Raman spectra of few-layer $FePS_3$

Incident polarization is fixed where P_1 has the maximum intensity.

Signatures of antiferromagnetic phase transition in 1L similar to bulk



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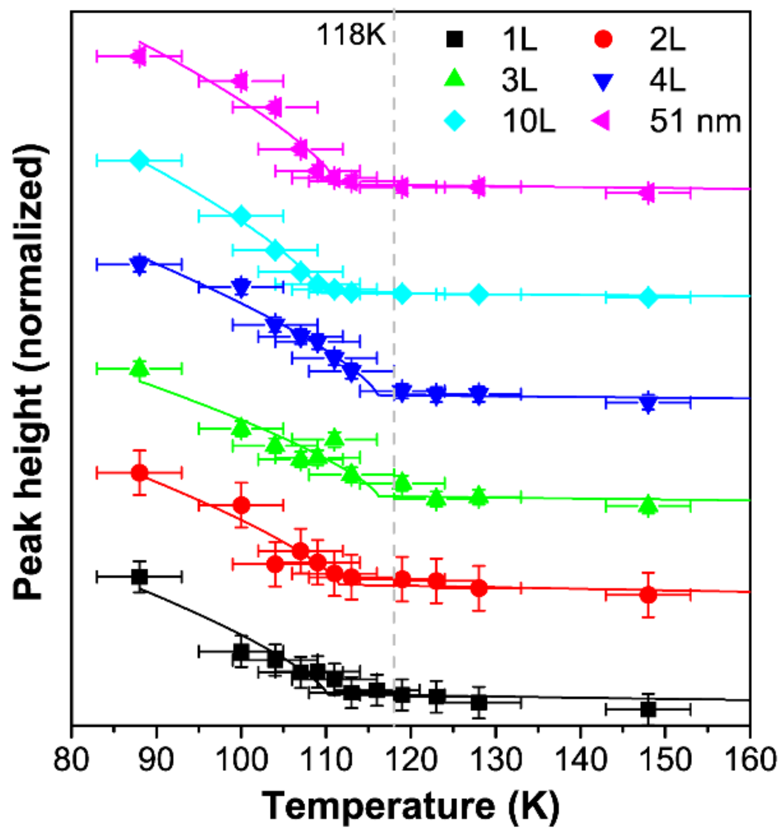
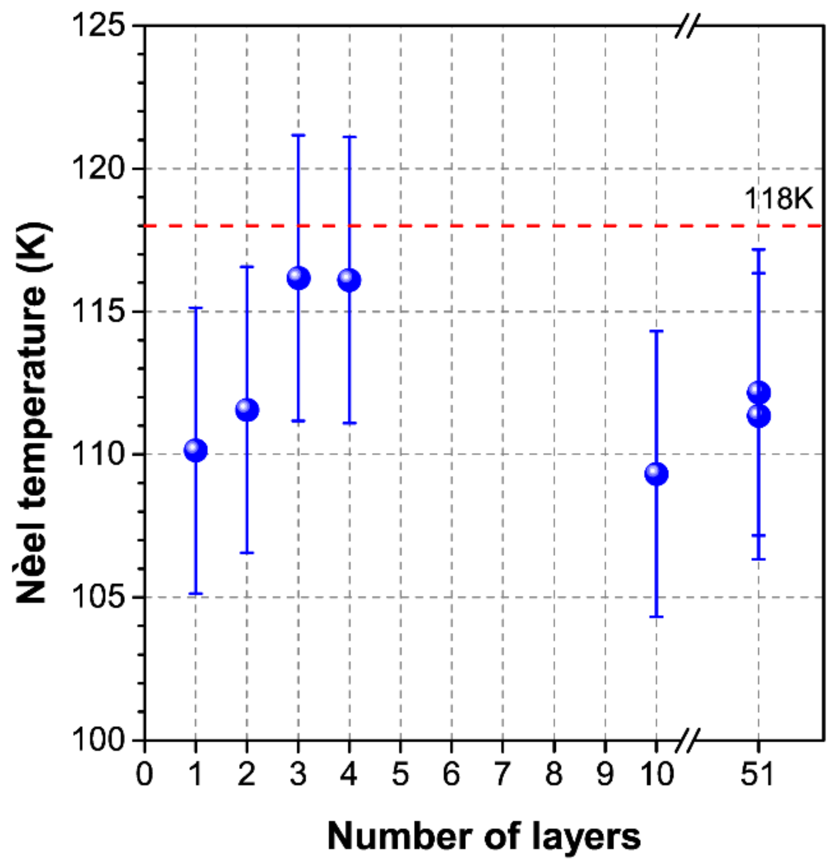
Thickness dependence of transition temperature (T_N)





- No apparent thickness dependence of Néel temperature

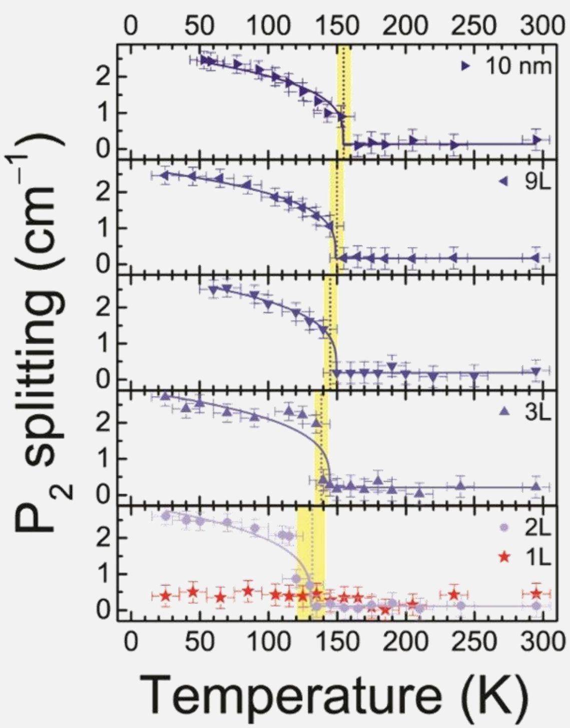
Summary: $FePS_3$

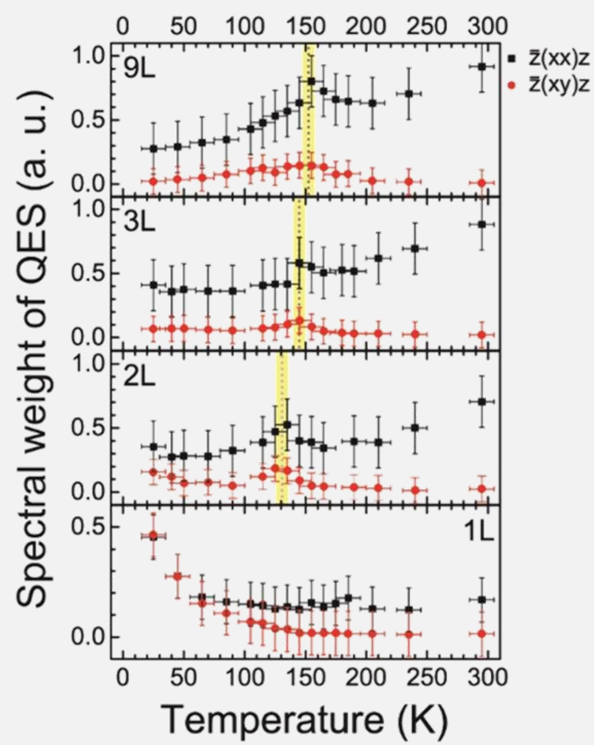
◆ $FePS_3$ exhibits an **Ising-type antiferromagnetic ordering down to the monolayer limit.**

◆ The transition temperature is almost independent of the thickness, indicating that the **interlayer interaction has little effect on the magnetic ordering.**

Nano Letters **16**, 7433 (2016)
Also, Wang *et al.*, *2D Mater.* (2016).

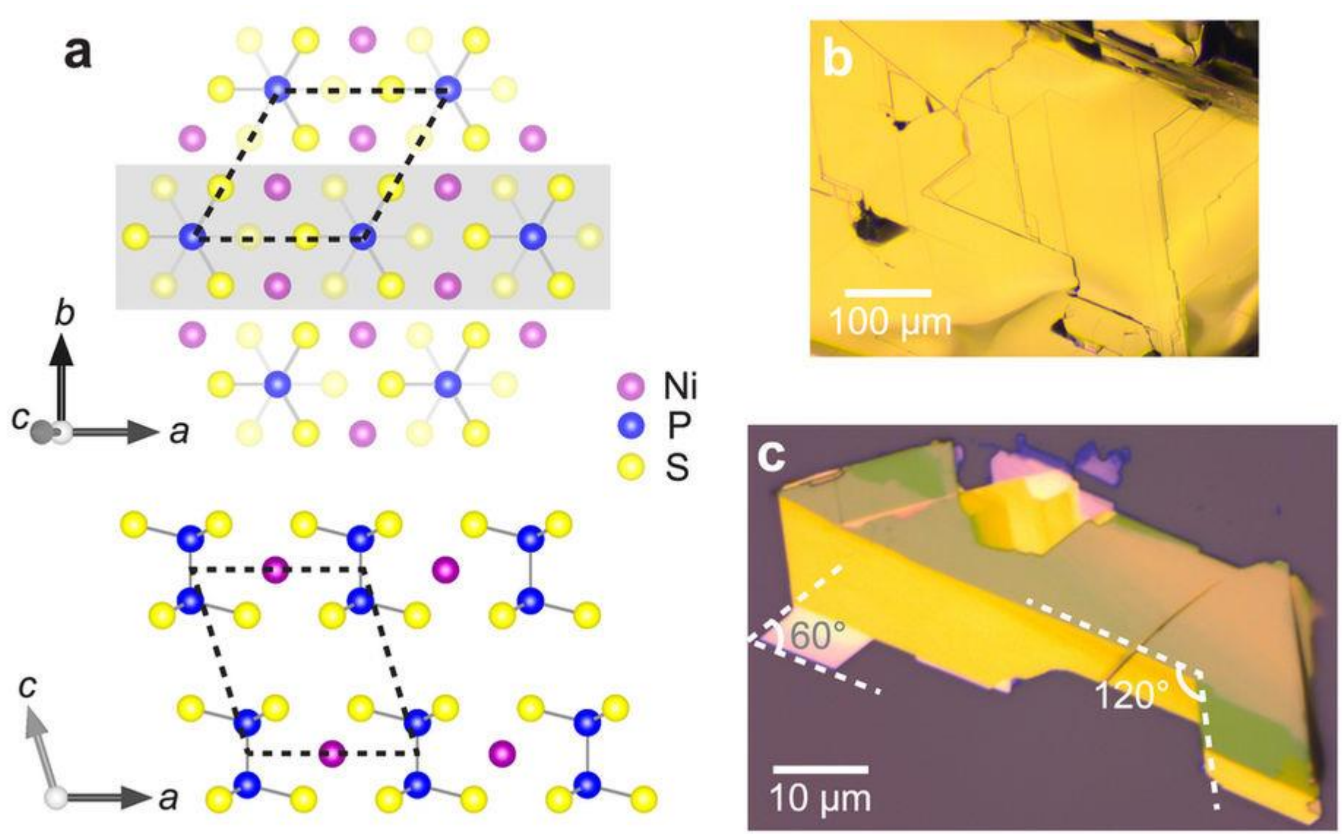
2. XXZ type antiferromagnet: $NiPS_3$





Nature Comm. **10**, 345 (2019).

Nickel phosphorus trisulfide ($NiPS_3$): Crystal structure



Je-Geun Park *et al.*, *Sci. Rep.* **6**, 20904 (2016).

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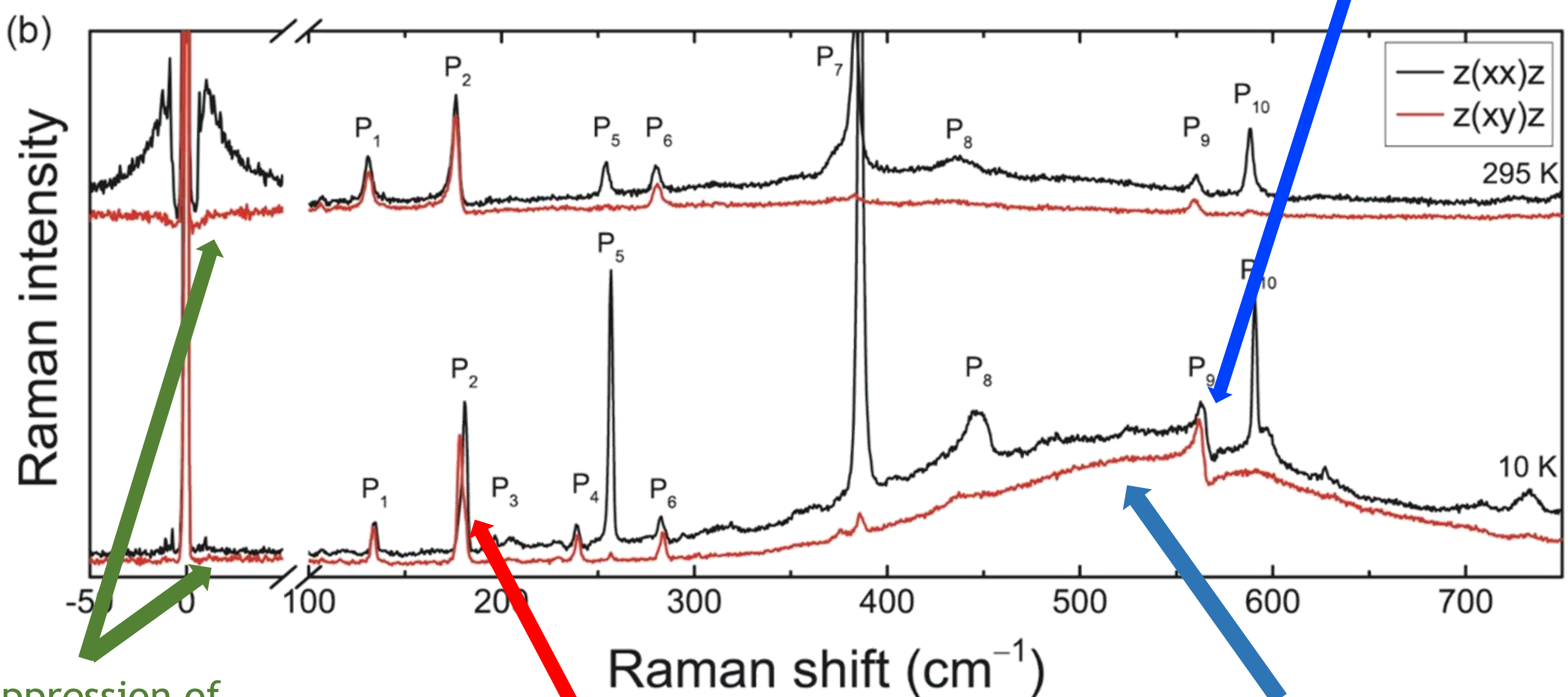
Raman spectra of bulk $NiPS_3$

Fano resonance



Suppression of quasi-elastic scattering due to spin fluctuation

Splitting of P_2

2-magnon scattering

Nature Comm. 10, 345 (2019).

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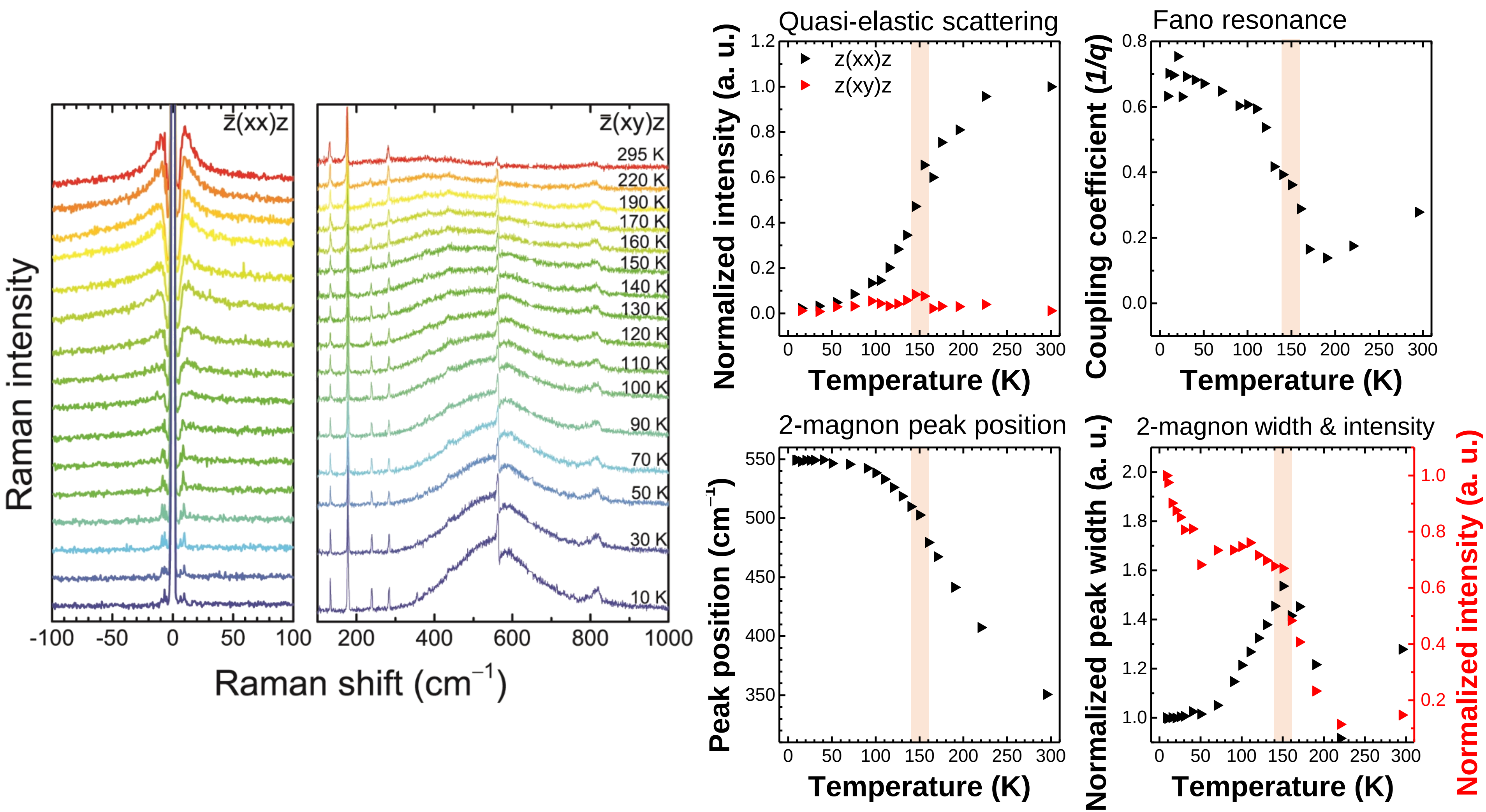
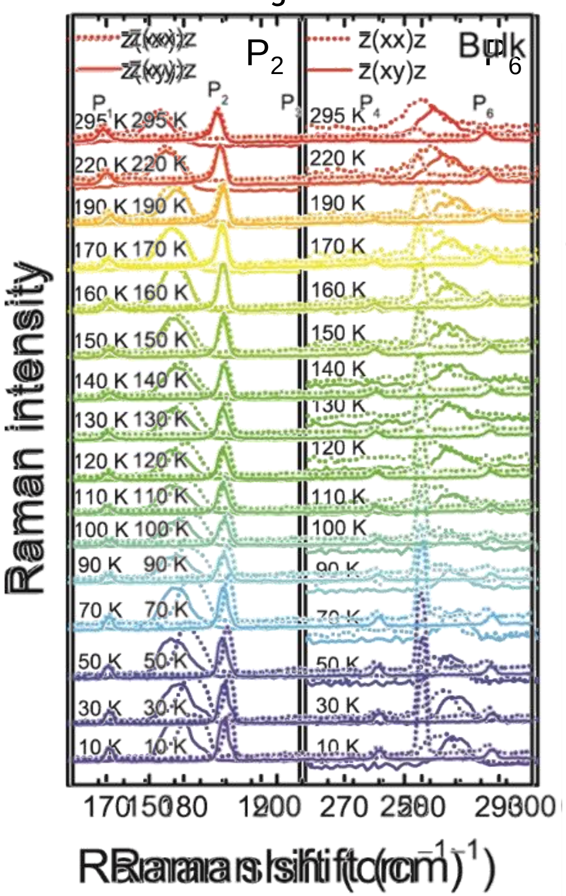
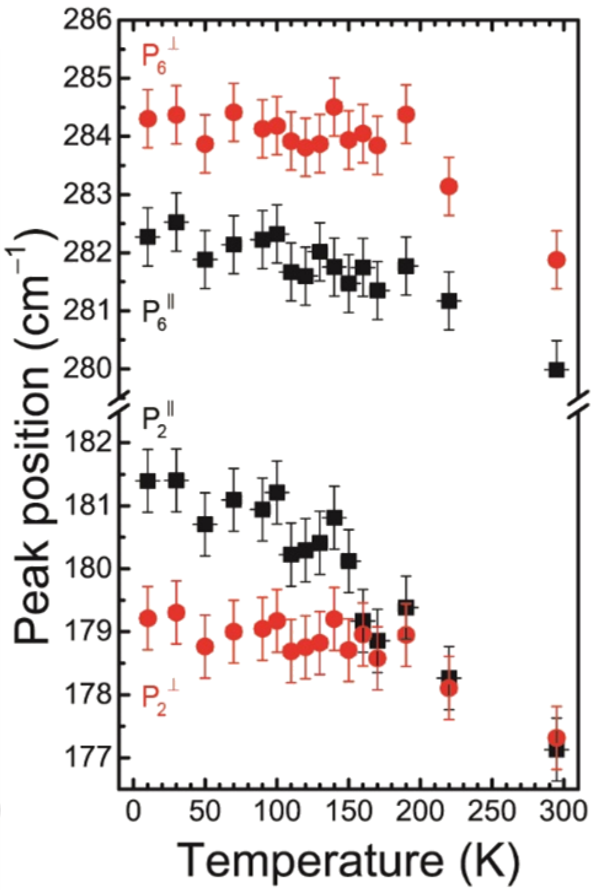
Raman signatures of magnetic ordering in bulk $NiPS_3$



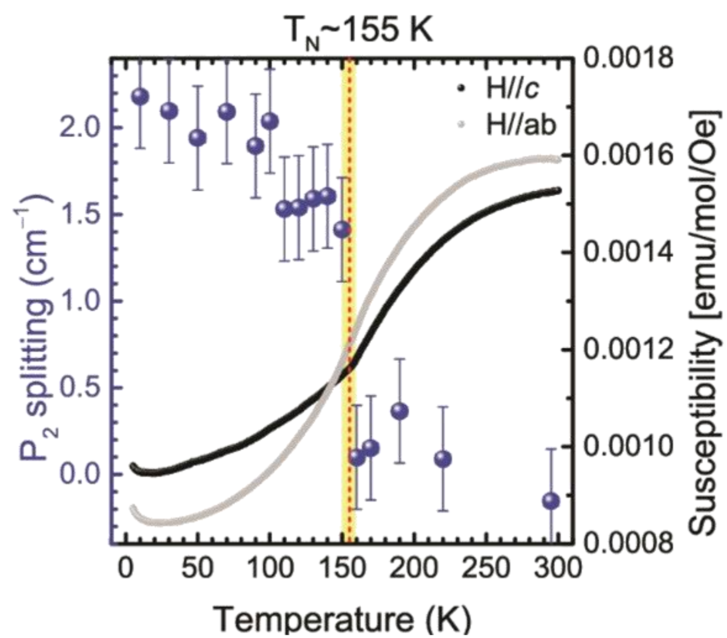
Raman signatures of magnetic ordering in **bulk** $NiPS_3$

Bulk $NiPS_3$





Splitting of P_2



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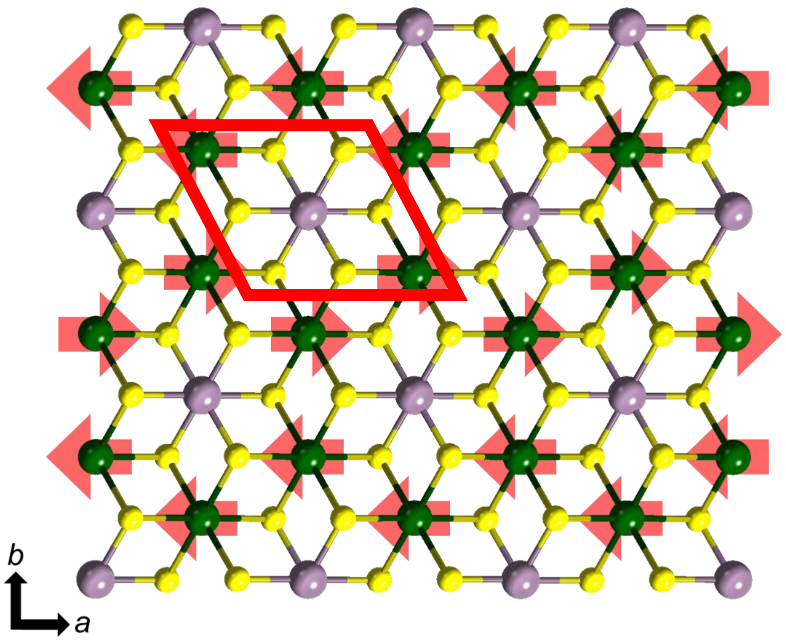
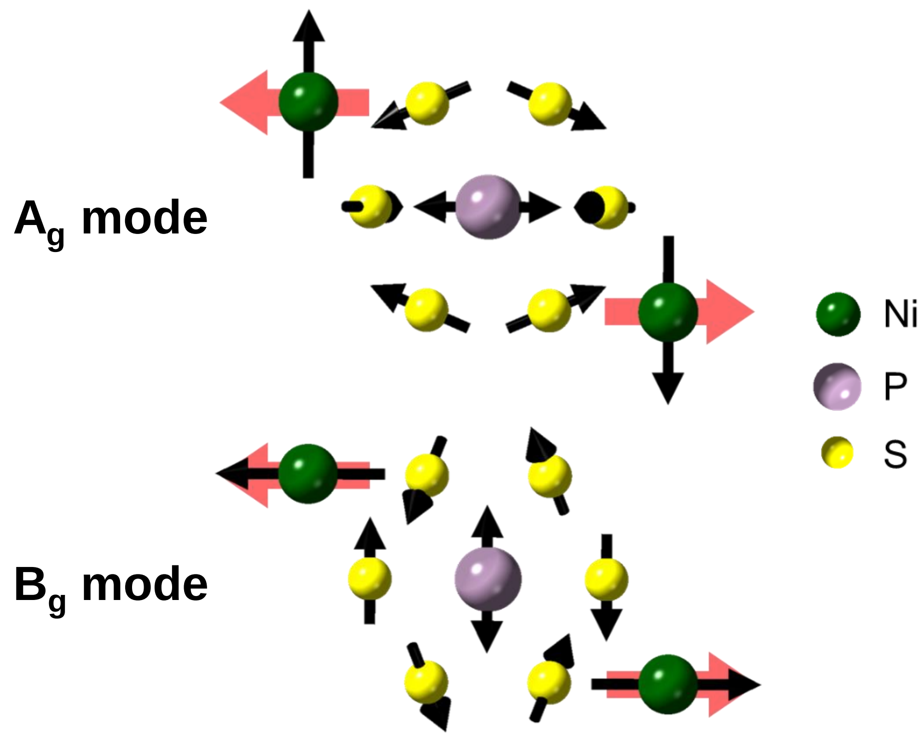
Splitting of phonon P_2 due to antiferromagnetic ordering

Magnetic Ordering in $NiPS_3$



Nearly Degenerate Modes (P_2)



- **Red arrows**: distribution of spin polarization below T_N
- **Black** arrows: vibration direction

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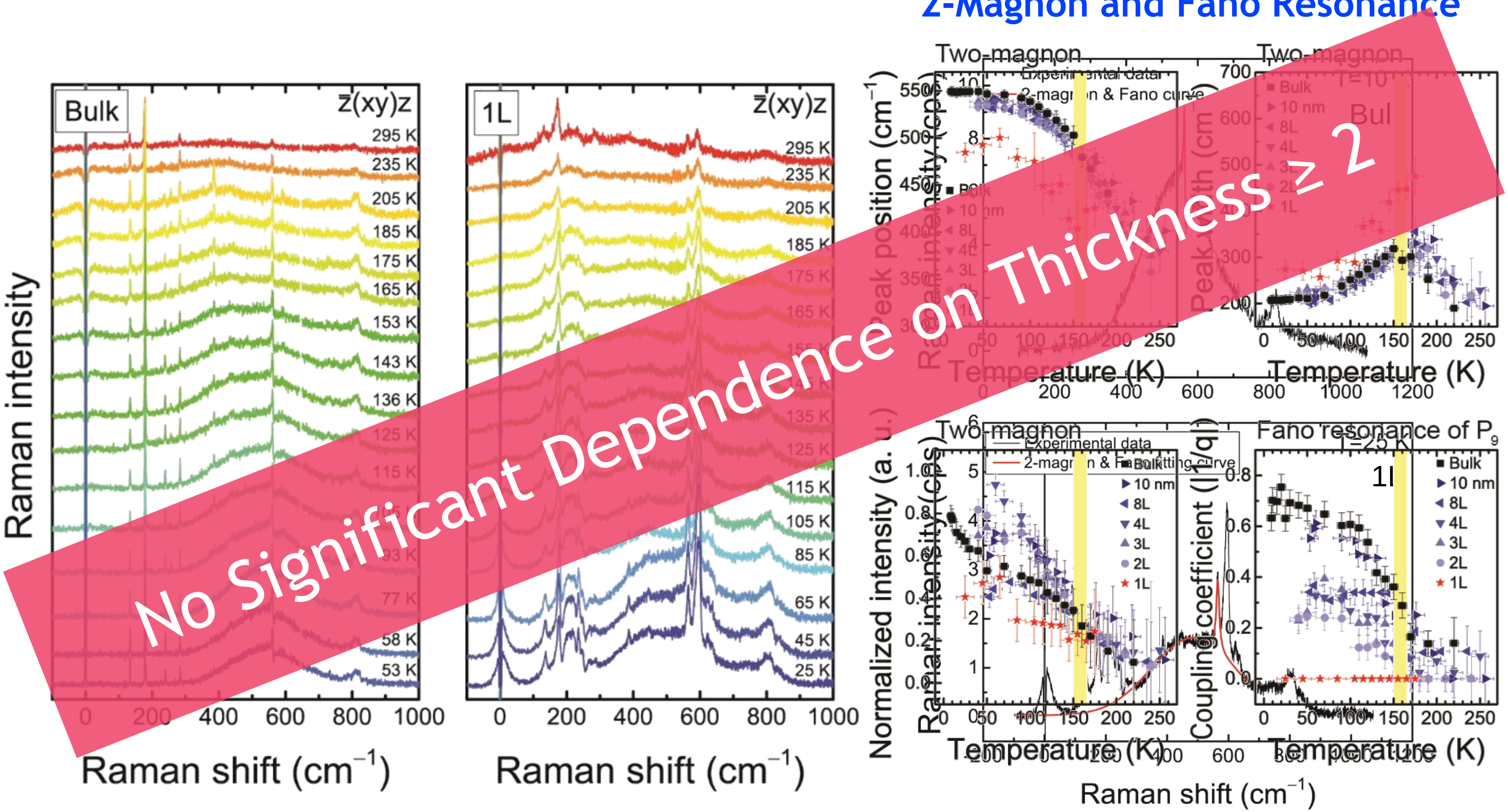
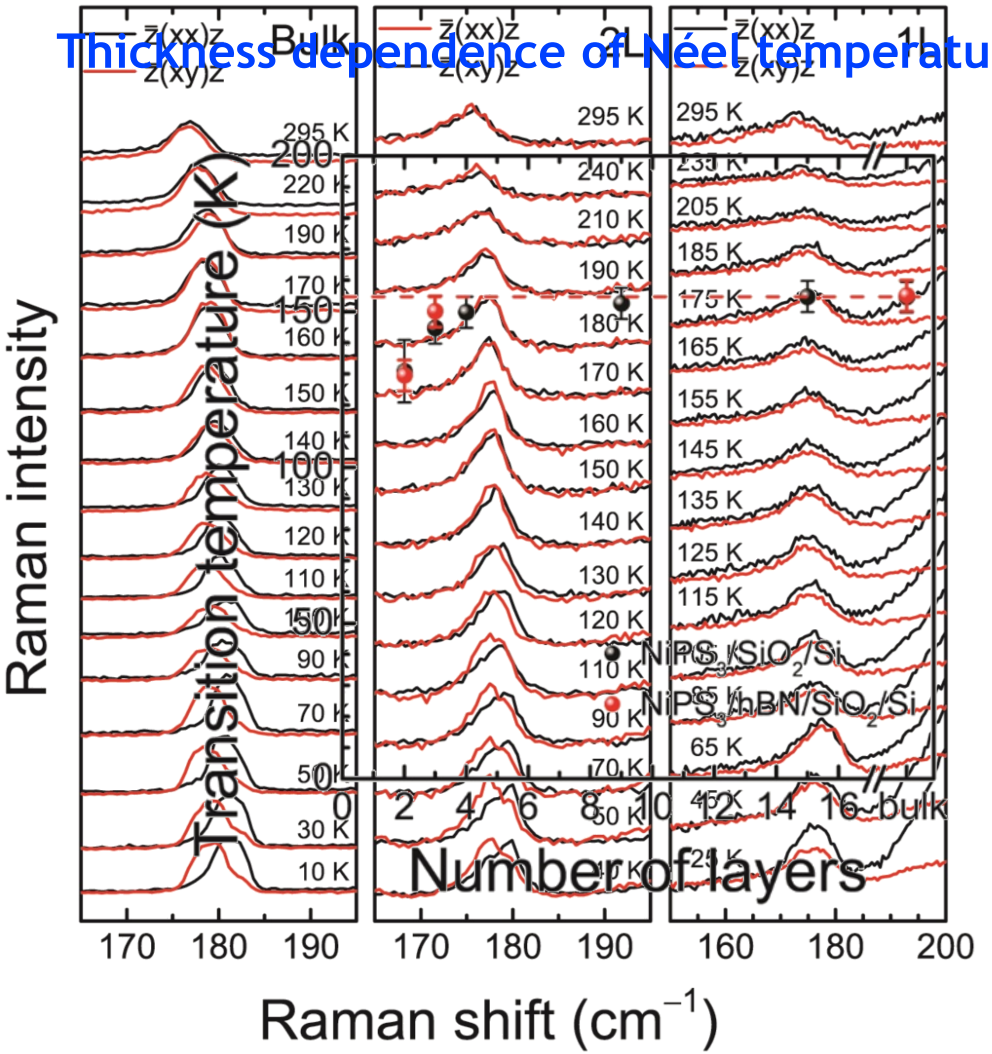
Thickness Dependence for Few-layer $NiPS_3$

2-Magnon and Fano Resonance

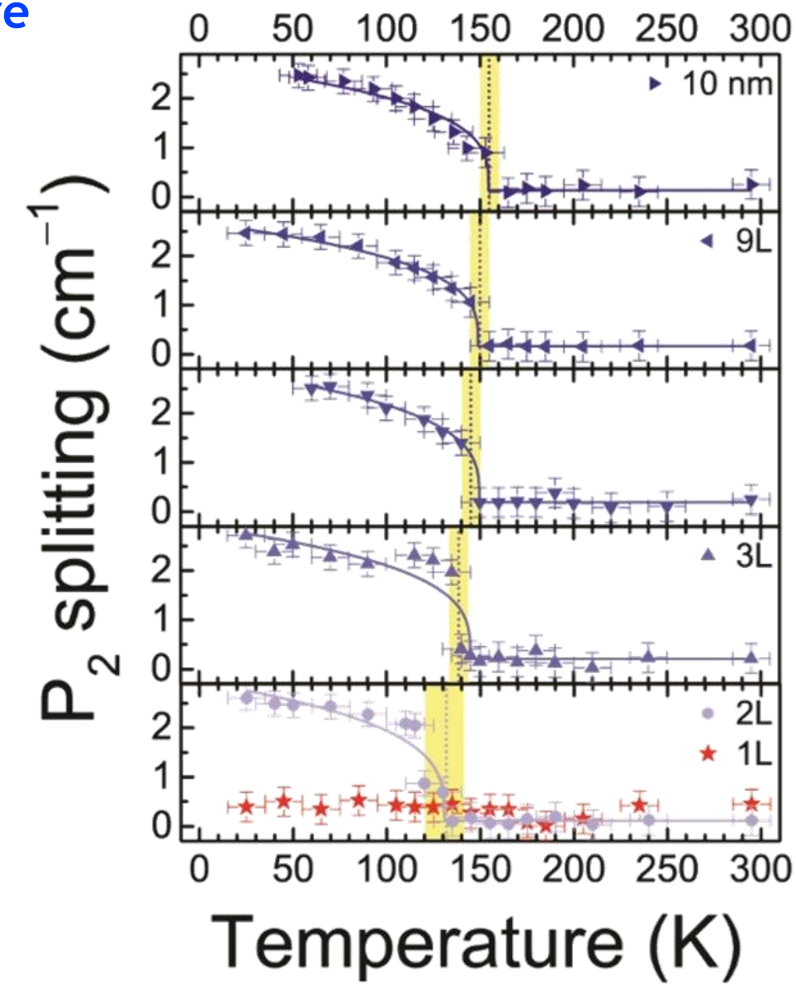


Thickness dependence for few-layer $NiPS_3$

Thickness dependence of Néel temperature

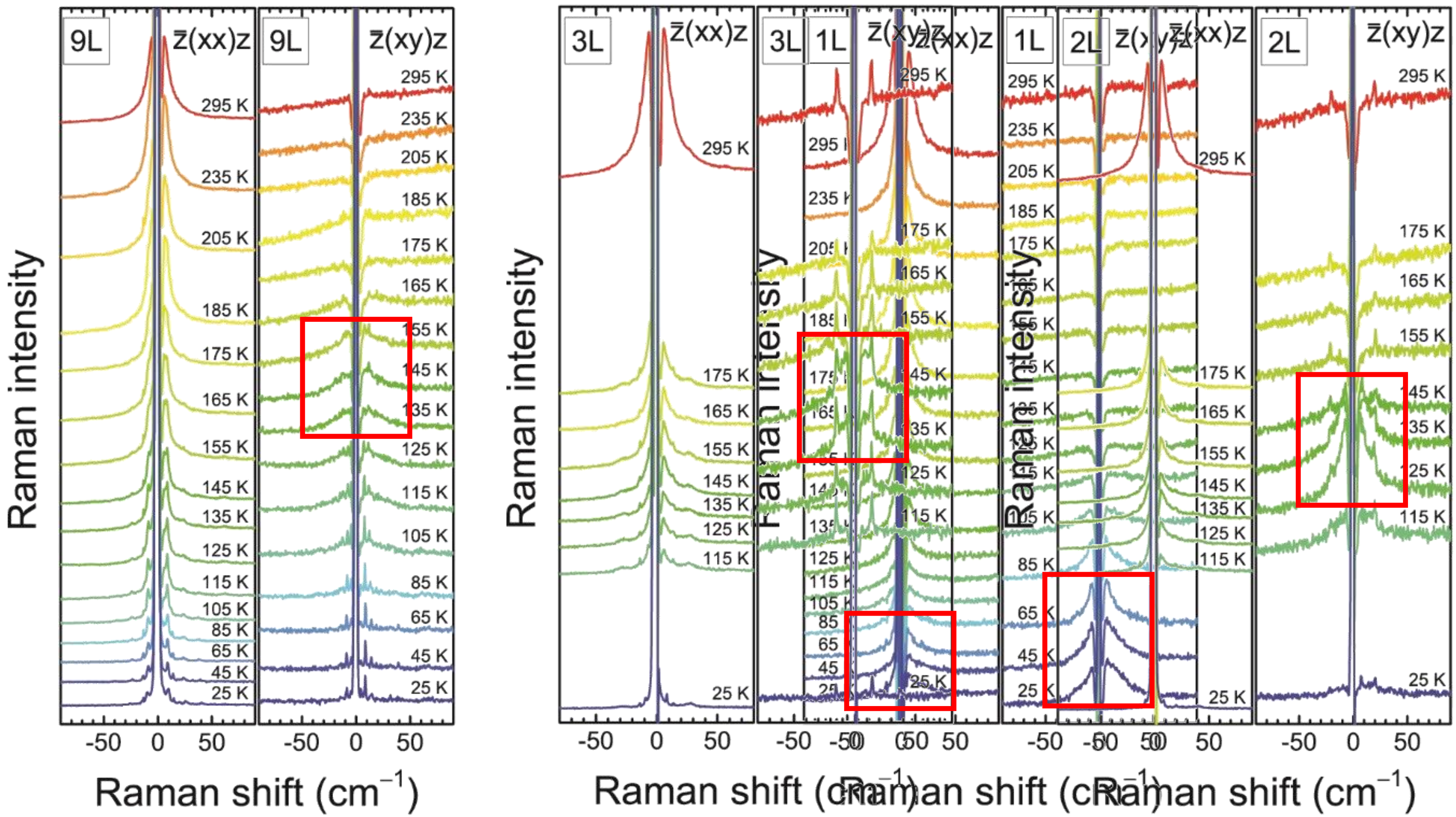


Splitting of P_2



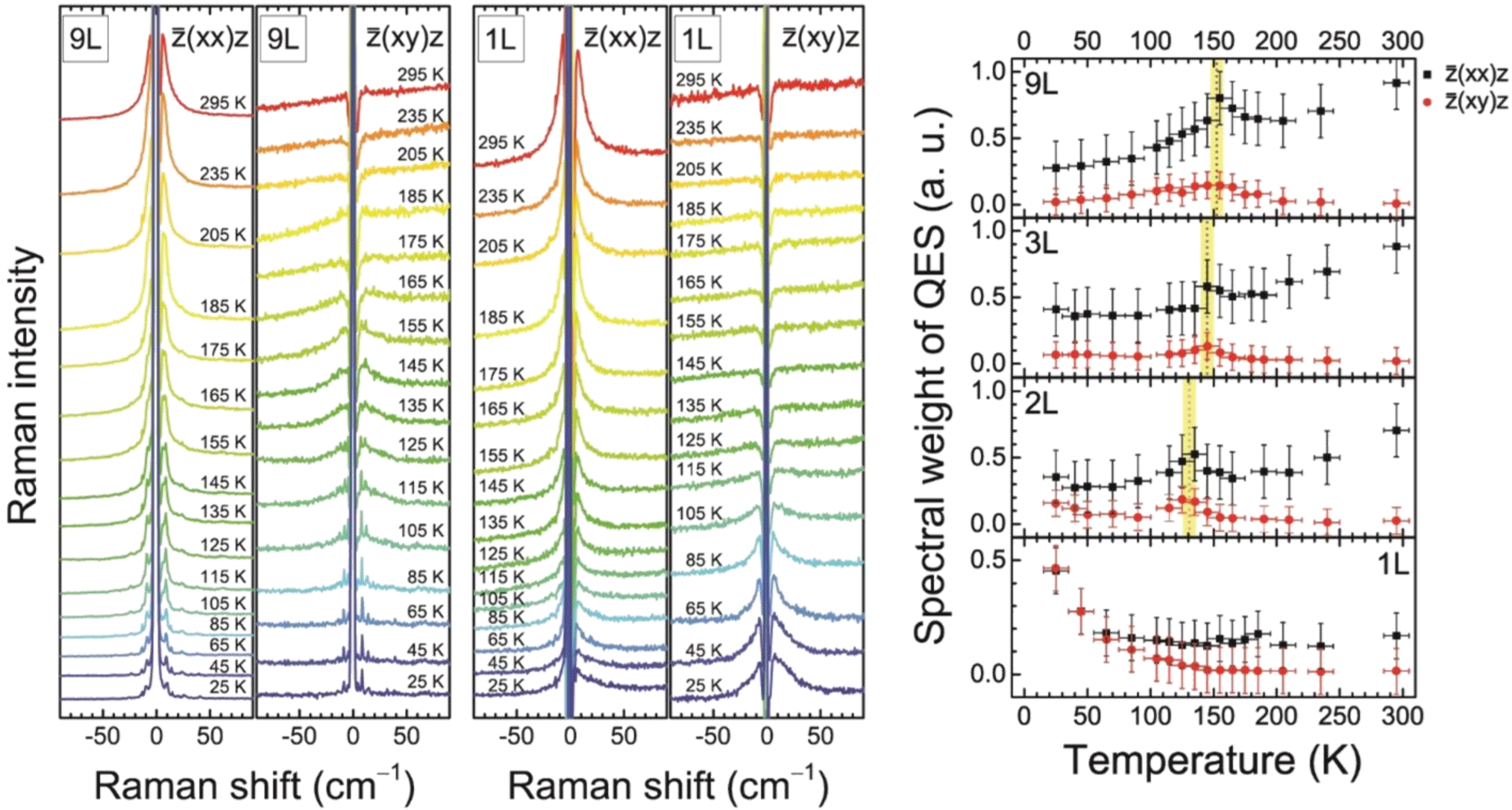
Thickness dependence for few-layer $NiPS_3$

Low-Frequency Raman spectra: Quasi-elastic scattering



Thickness dependence for few-layer $NiPS_3$

Temperature dependence of spectral weight of QES

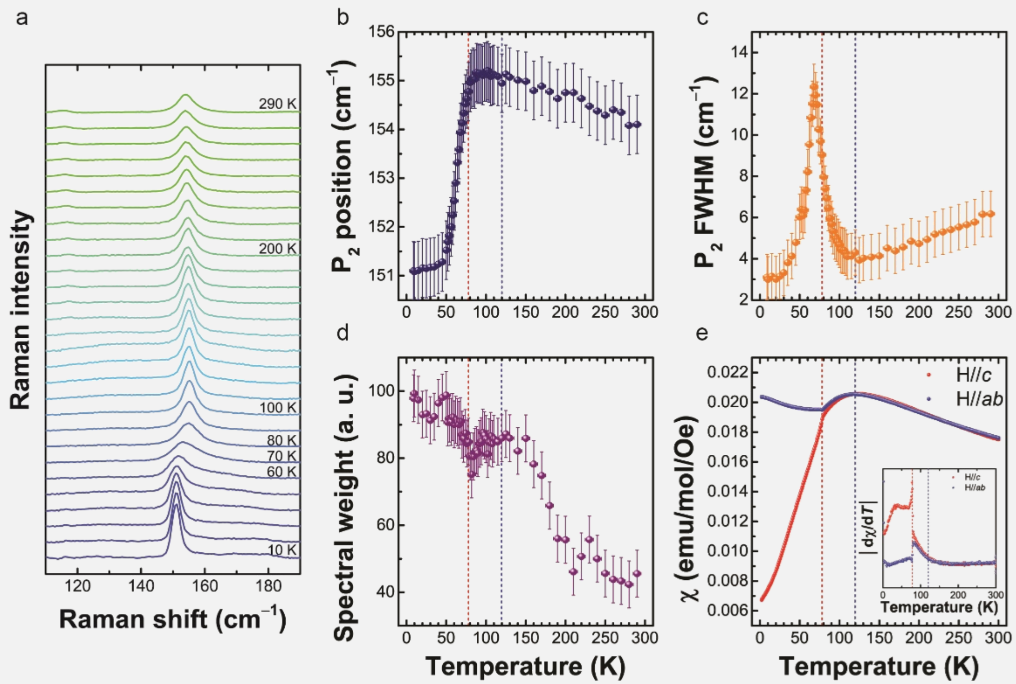


Summary: $NiPS_3$

- $NiPS_3$ exhibits an **antiferromagnetic ordering down to 2-layer**.
- The transition temperature is **almost independent of the thickness down to 2L** but seems to be **suppressed significantly for 1L**.
- In XXZ antiferromagnet, the magnetic ordering becomes very fragile in the 2D limit of 1-layer.

Nature Comm. 10, 345 (2019).

3. Heisenberg type antiferromagnet: $MnPS_3$

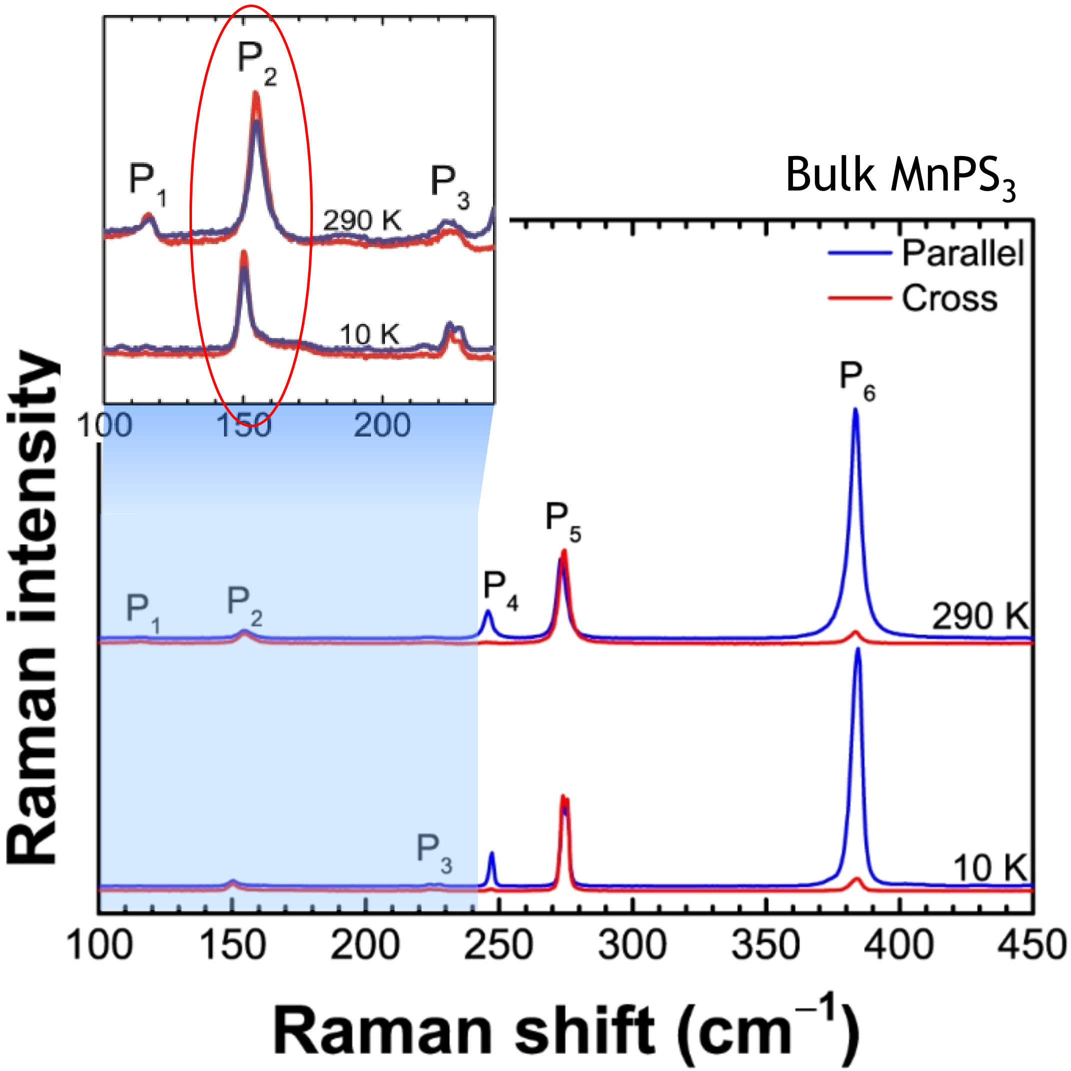


2D Materials **6**, 041001 (2019).

Current Applied Physics, submitted.

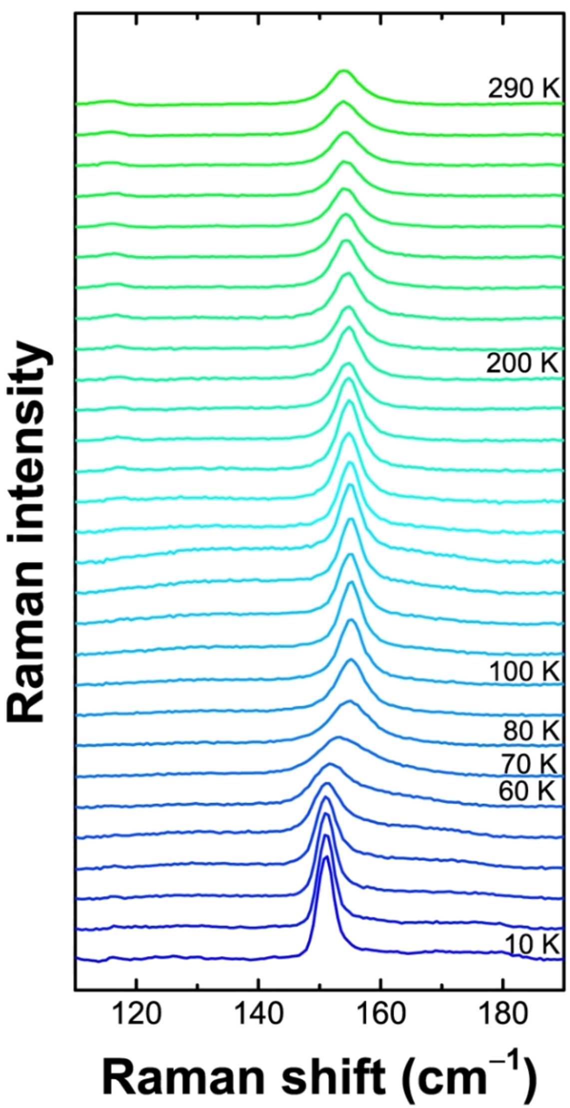
Poster 15, TODAY

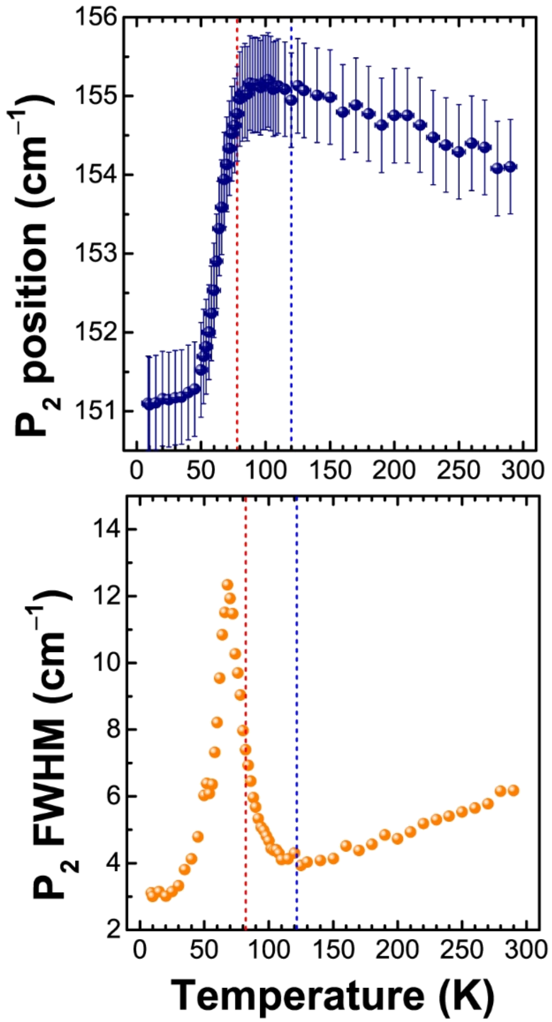
Raman spectra of $MnPS_3$ (Heisenberg type)

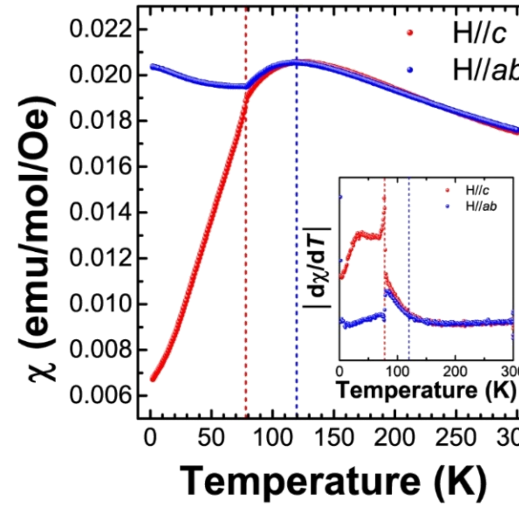


2D Mat. **6**, 041001 (2019).

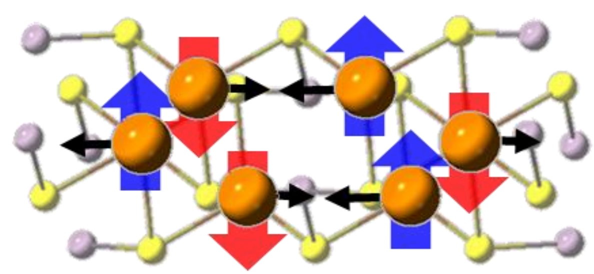
Bulk $MnPS_3$ (Heisenberg type)







P$_2$ vibration mode



2D Mat. **6**, 041001 (2019).

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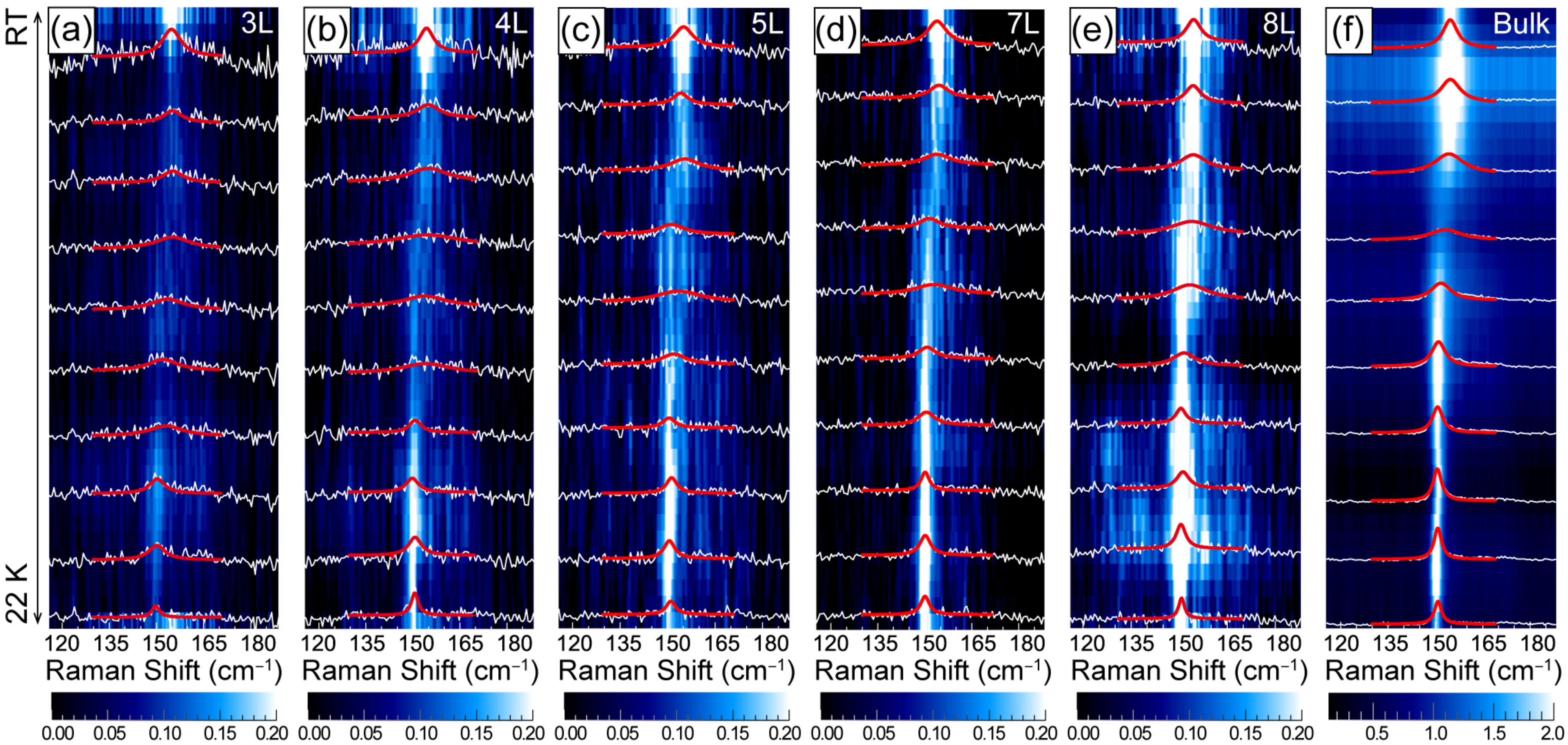
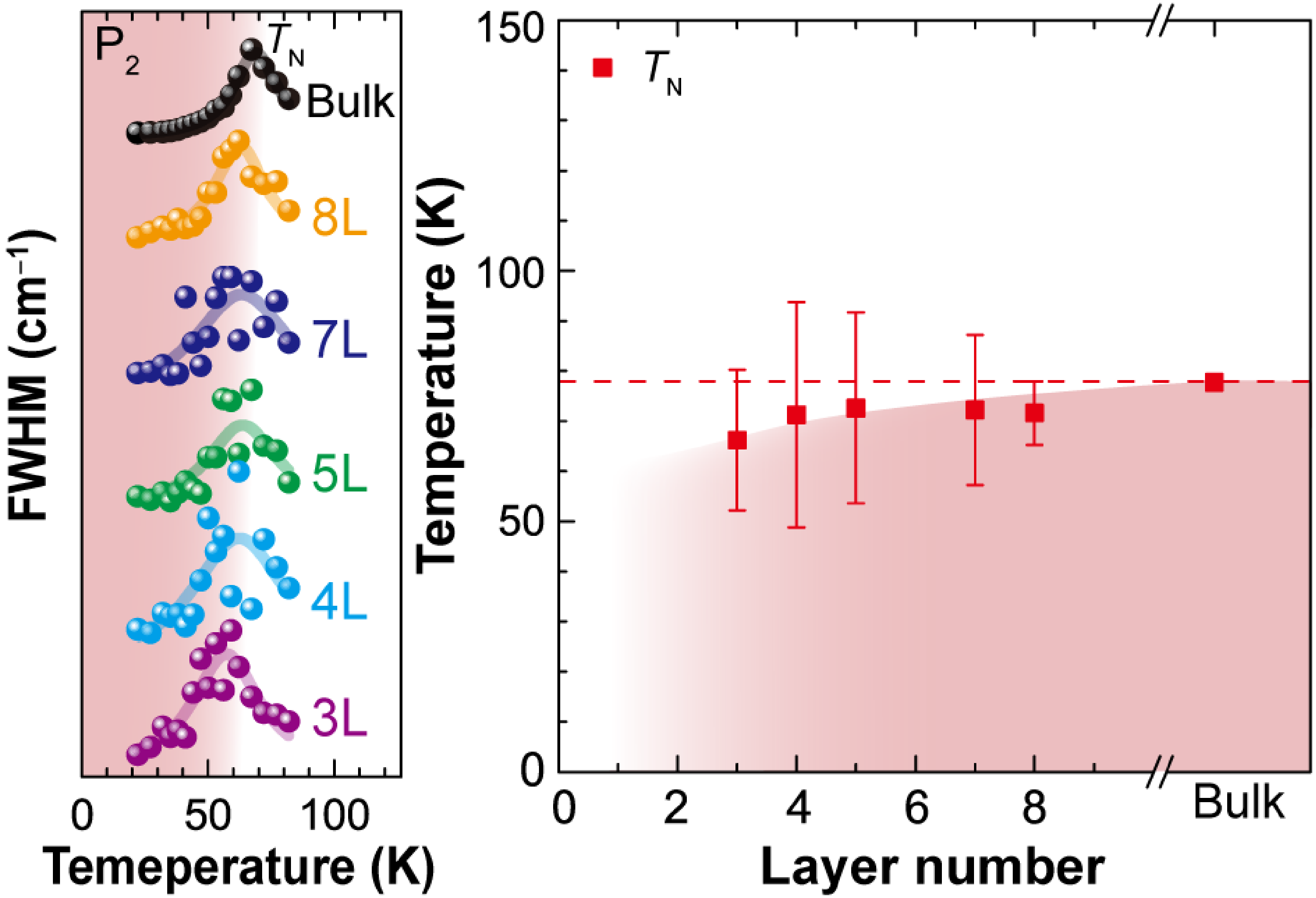
Temperature dependence of P_2



Current Applied Physics, submitted.

Poster 15, TODAY

Temperature dependence of FWHM of P_2



Current Applied Physics, submitted.

Poster 15, TODAY

Summary: $MnPS_3$

- $MnPS_3$ exhibits an **antiferromagnetic ordering down to 2-layer**.
- In Heisenberg antiferromagnet, the magnetic ordering can be sustained down to 2 layer.
- The transition temperature decreases slightly for thinner layers.

2D Materials **6**, 041001 (2019).
Current Applied Physics, submitted.
Poster 15, TODAY
Also, Sun, et al., J. Phys. Chem. Lett. **10**, 3087 (2019).

IOP Publishing Nanotechnology

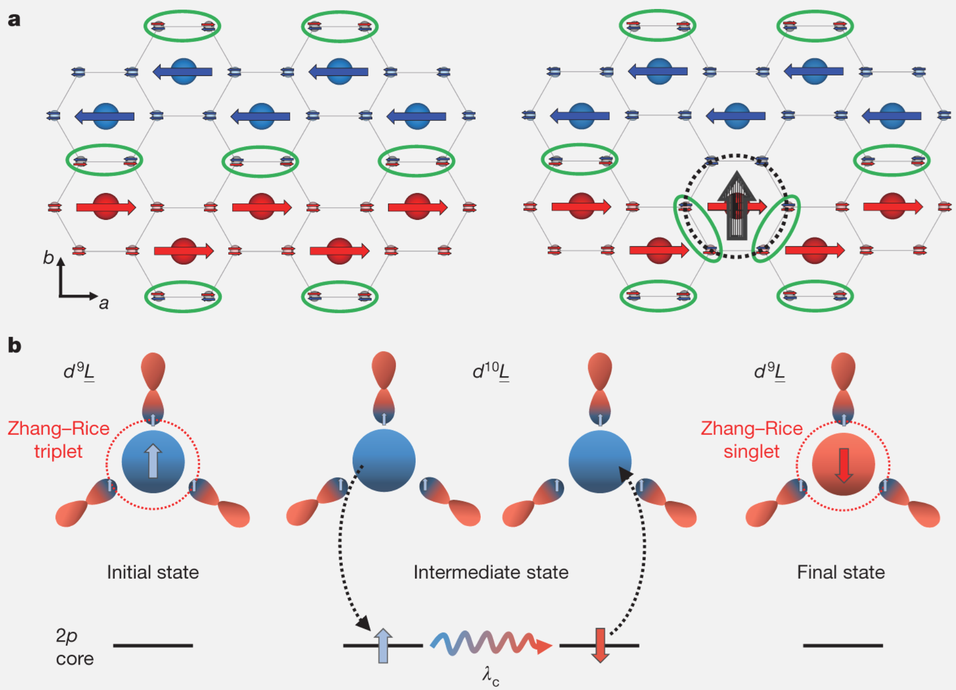
Nanotechnology **30** (2019) 452001 (10pp) https://doi.org/10.1088/1361-6528/ab37a4

Topical Review

Raman spectroscopy of two-dimensional magnetic van der Waals materials

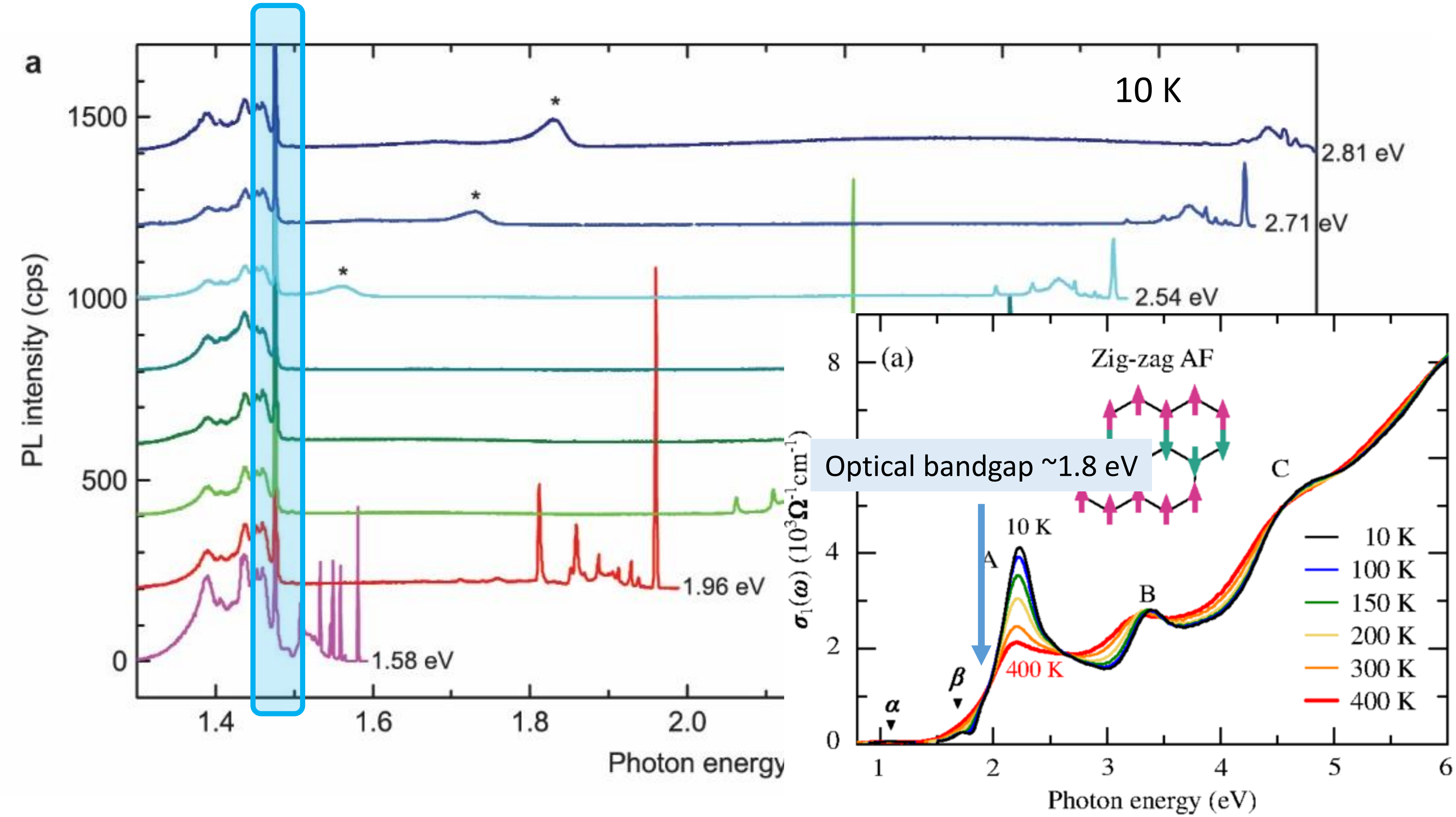
Kangwon Kim[1], Jae-Ung Lee[2] and Hyeonsik Cheong[1]

4. Coherent many-body exciton in $NiPS_3$



Nature **583**, 785 (2020).

Low-temperature photoluminescence



Kim, *et al.*, PRL **120**, 136402 (2018).

Coherent many-body exciton in $NiPS_3$

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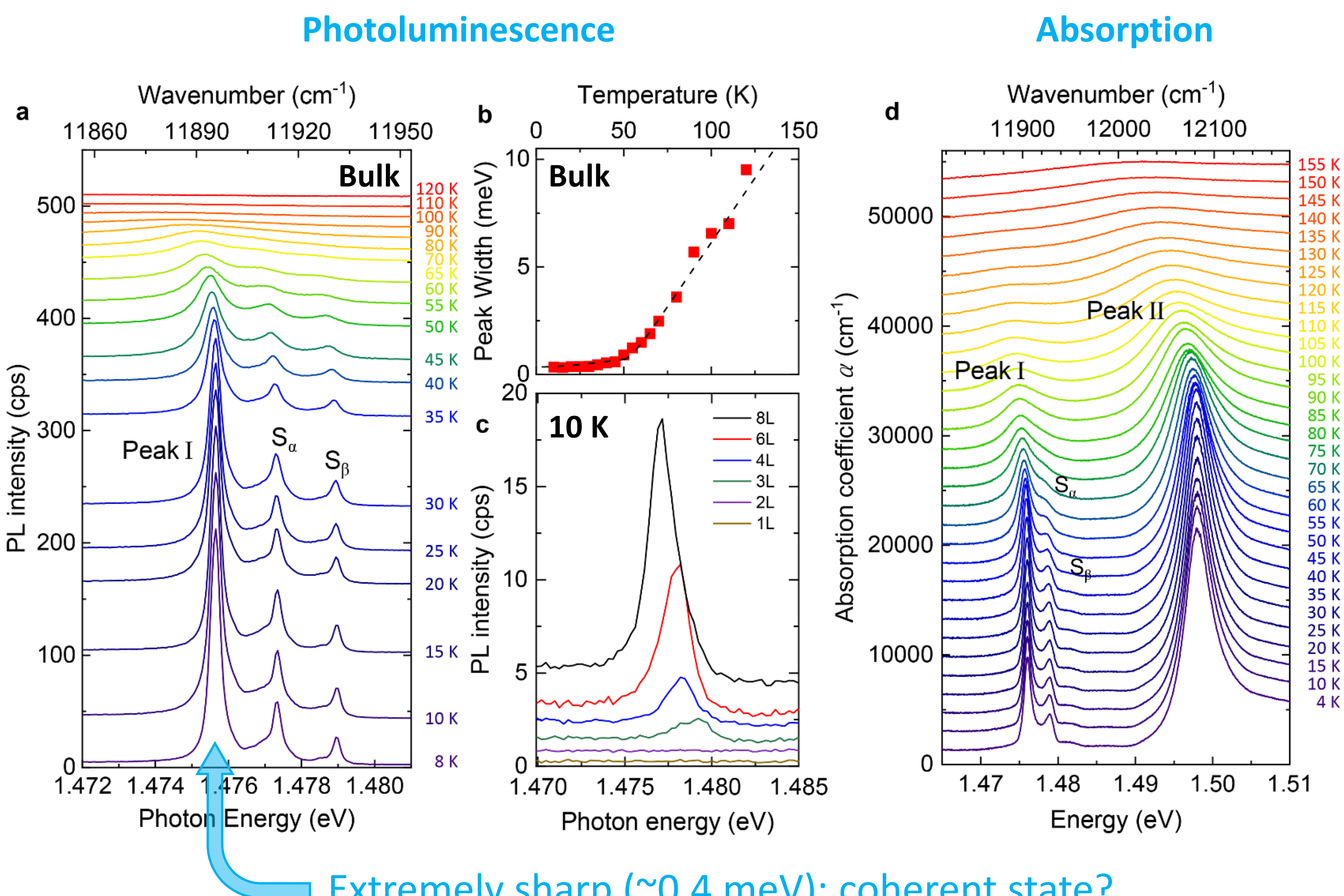
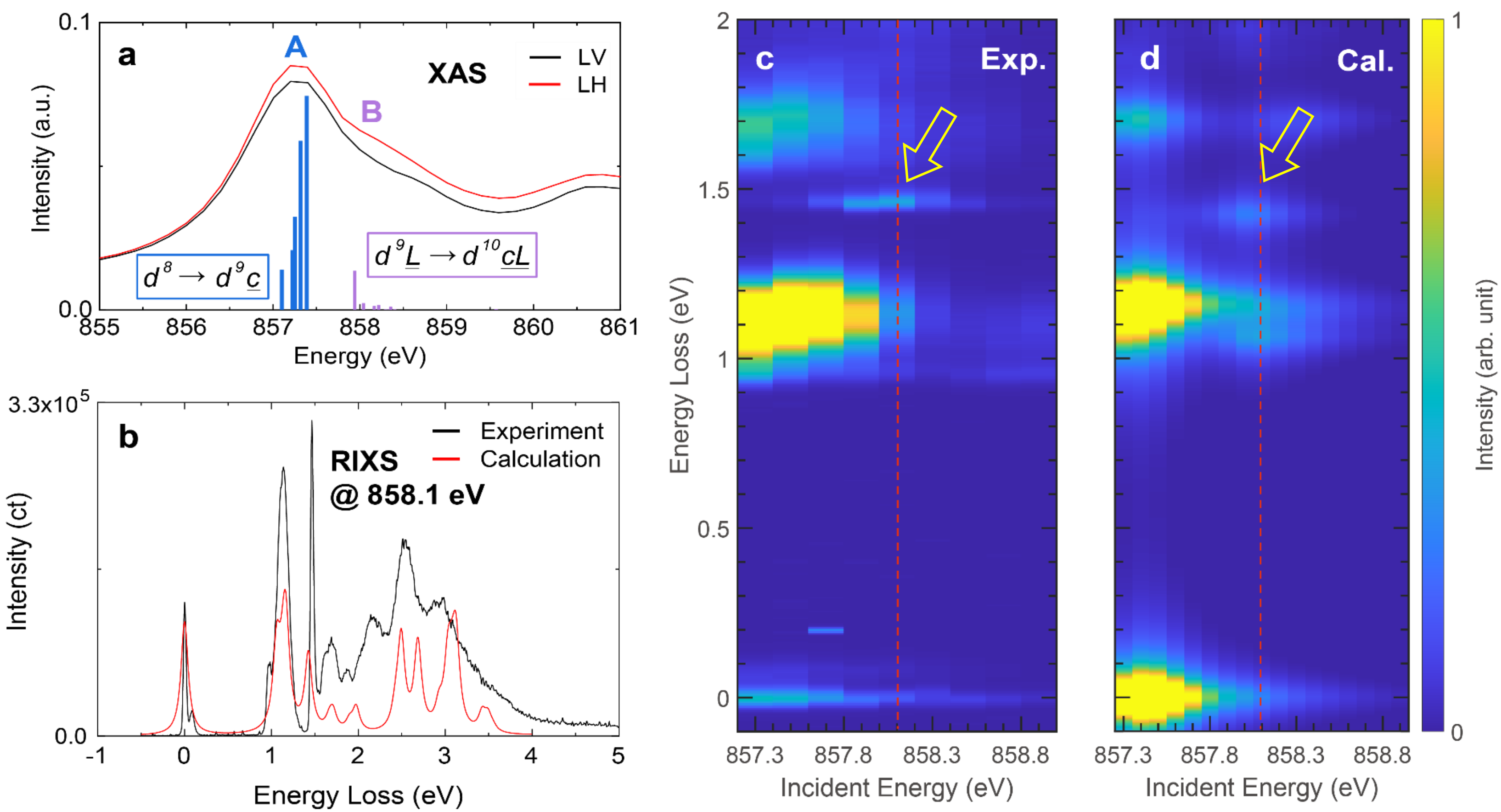
Temperature dependence of PL & absorption



Extremely sharp (~0.4 meV): coherent state?

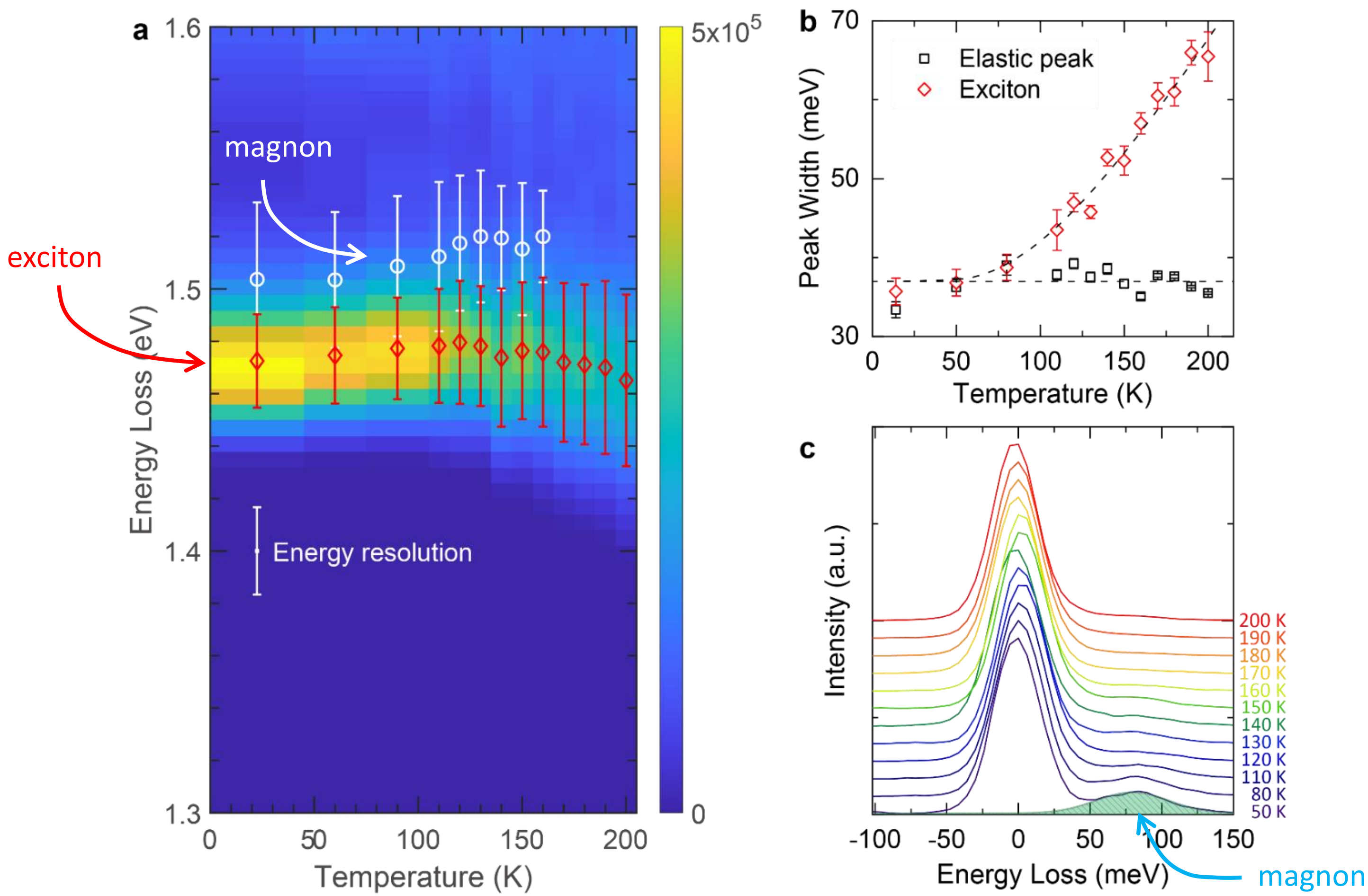
Nature **583**, 785 (2020).

Resonant inelastic x-ray scattering (RIXS)



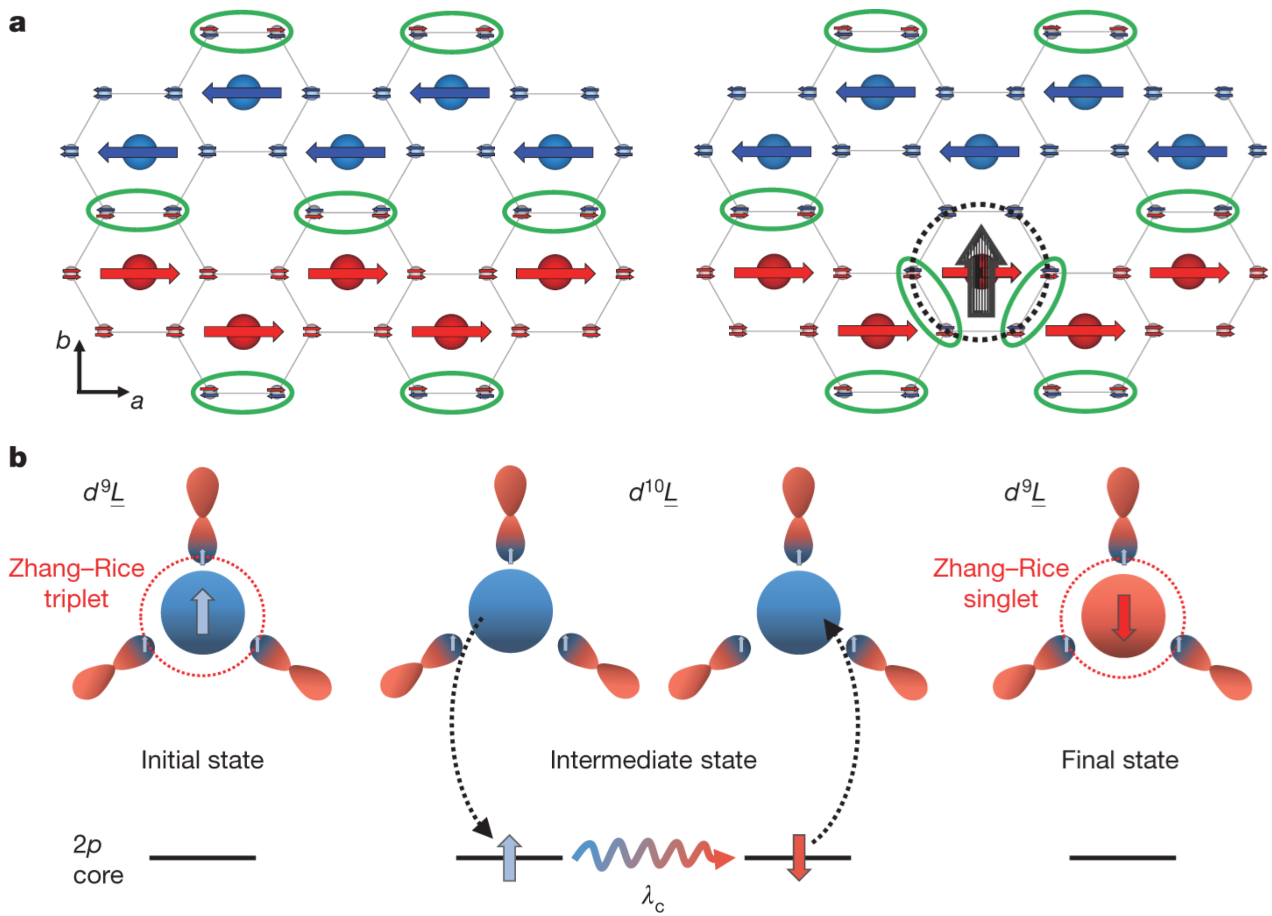
Nature **583**, 785 (2020).

Temperature dependence of RIXS



Nature **583**, 785 (2020).

Theoretical model



Nature **583**, 785 (2020).

Collaborators

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- Je-Geun Park (Seoul National Univ.): Materials growth
- Cheol-Hwan Park (Seoul National Univ.): Phonon calculations
- Gun Sang Jeon (Ewha Womans Univ.): Monte Carlo simulation
- Jae Hoon Kim (Yonsei Univ.): Optical Absorption
- Young-Woo Son (KIAS): Theoretical calculations

- Thanks for helpful discussions: H.C. Lee (Sogang Univ.), S. Yoon (Ewha Womans Univ.), K. Burch (Boston College), A.R. Wildes (Grenoble) and K.-Y. Choi (Chung-Ang Univ.)

Thank you!