Raman studies of the antiferromagnetic phase transition in atomically thin $NiPS_3$

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01 Introduction & Motivation

Magnetism in low dimension

- **XXZ Hamiltonian**

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

- J_{XY} & J_I : spin-exchange energies on the basal plane and along the *c* axis

$J_{XY} = 0$	$J_I = 0$	$J_{XY} = J_I$
Ising	XY	Isotropic Heisenberg

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)

➔ Phase transition at a finite temperature in 2D-Ising systems

VOLUME 17, NUMBER 22 PHYSICAL REVIEW LETTERS 28 NOVEMBER 1966

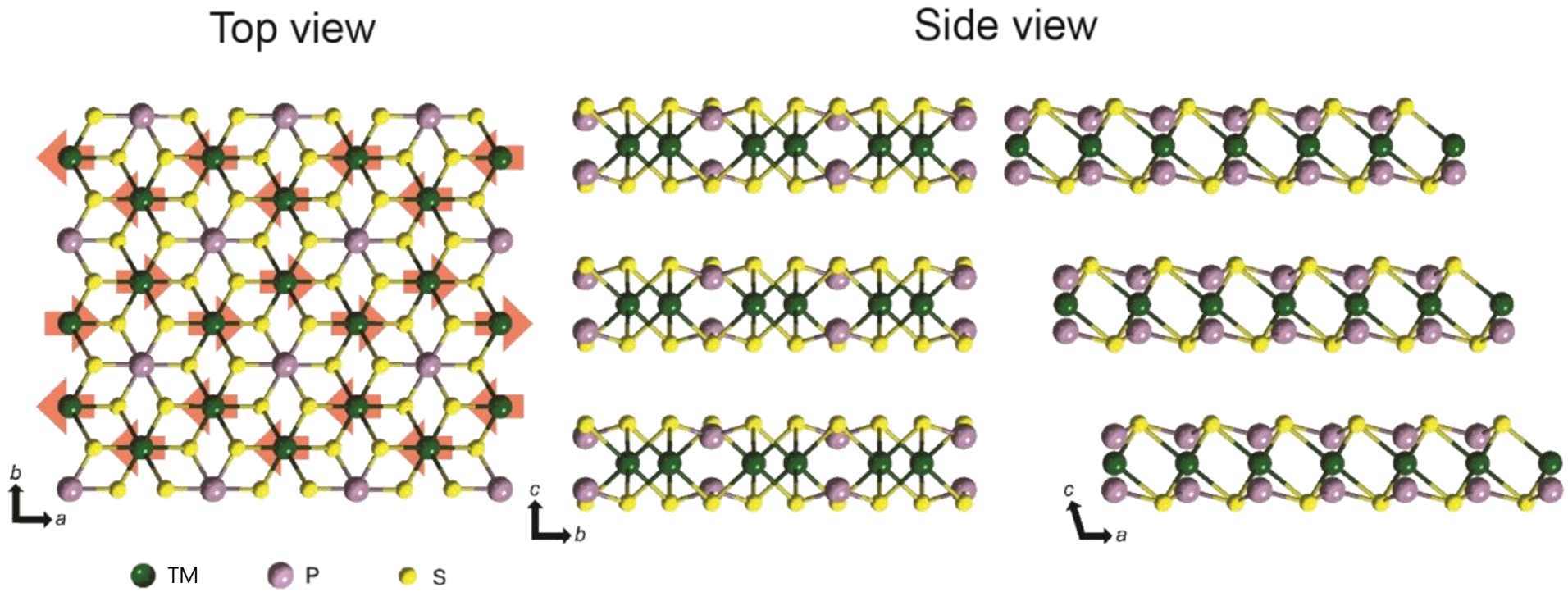
ABSENCE OF FERROMAGNETISM OR ANTIFERROMAGNETISM IN ONE- OR TWO-DIMENSIONAL ISOTROPIC HEISENBERG MODELS*

N. D. Mermin† and H. Wagner‡
Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York
(Received 17 October 1966)

➔ No magnetic order at finite temperature for 1D- or 2D isotropic Heisenberg systems

01 Introduction & Motivation

Transition metal phosphorous trisulfides ($TMPS_3$)

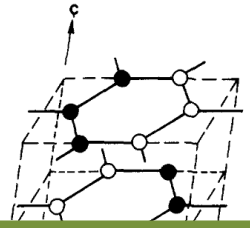


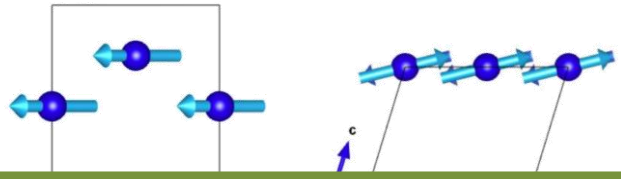
- $TMPS_3$ (TM = Fe, Ni, or Mn): monoclinic structure (C_{2h})
- Because of the weak van der Waals interaction along the *c* axis, $TMPS_3$ can be easily exfoliated to atomically thin films.

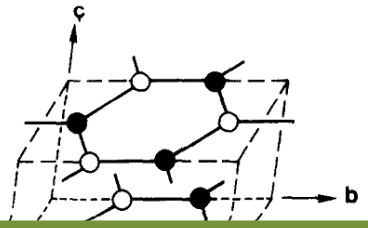
01 Introduction & Motivation

Magnetism of $TMPS_3$ depending on TM elements

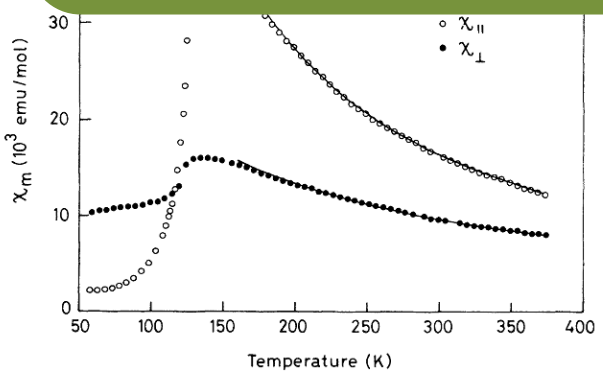
$FePS_3$ **(Ising)** $NiPS_3$ **(XXZ)** $MnPS_3$ **(Heisenberg)**

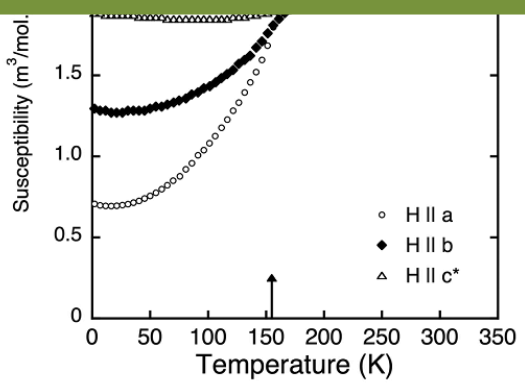


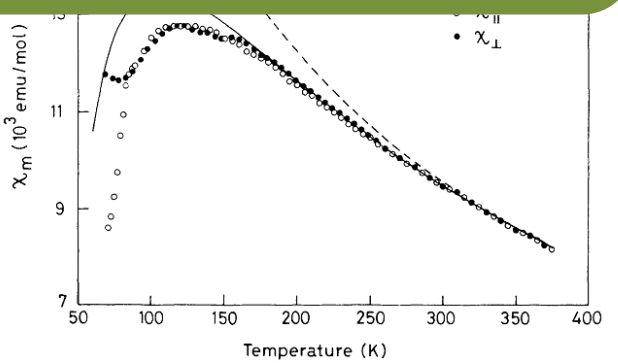




$TMPS_3$ is a suitable material to study antiferromagnetism in the 2D limit.







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).

01 Introduction & Motivation

Ferromagnetism in low dimension

서강대학교 SOGANG UNIVERSITY

LETTER

doi:10.1038/nature22060

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

$Cr_2Ge_2Te_6$: Ferromagnetic ordering down to 2L

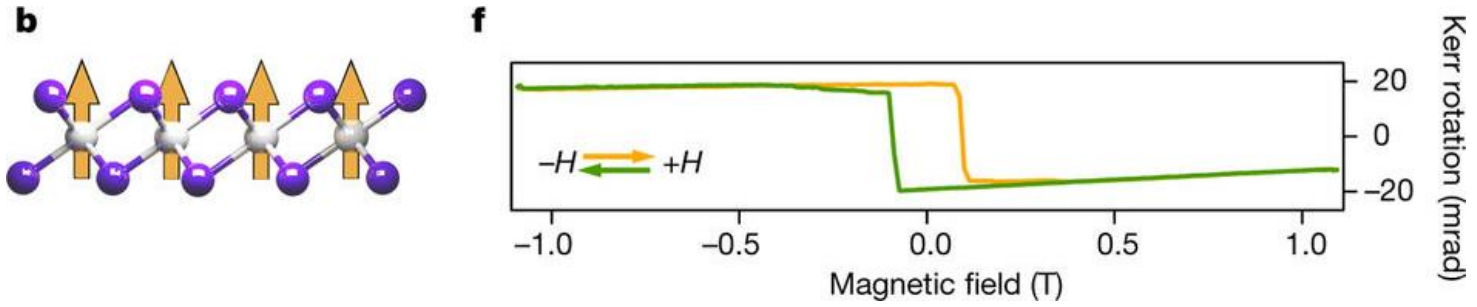
Nature **546**, 265 (2017).

Antiferromagnetism is more difficult to measure!

Waals crystal down to the monolayer limit

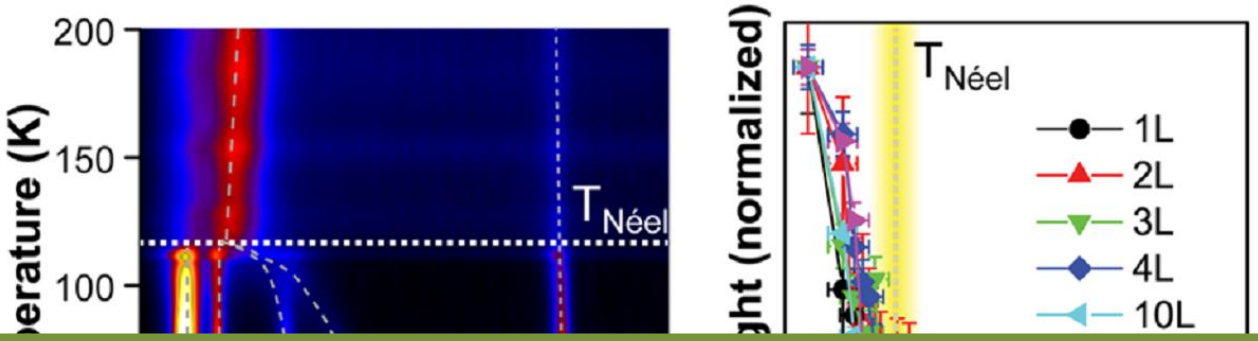
Bevin Huang[1]*, Genevieve Clark[2]*, Efrén Navarro-Moratalla[3]*, Dahlia R. Klein[3], Ran Cheng[4], Kyle L. Seyler[1], Ding Zhong[1], Emma Schmidgall[1], Michael A. McGuire[5], David H. Cobden[1], Wang Yao[6], Di Xiao[4], Pablo Jarillo-Herrero[3] & Xiaodong Xu[1,2]

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



01 Introduction & Motivation

Estimating the Néel temperature by Raman spectroscopy



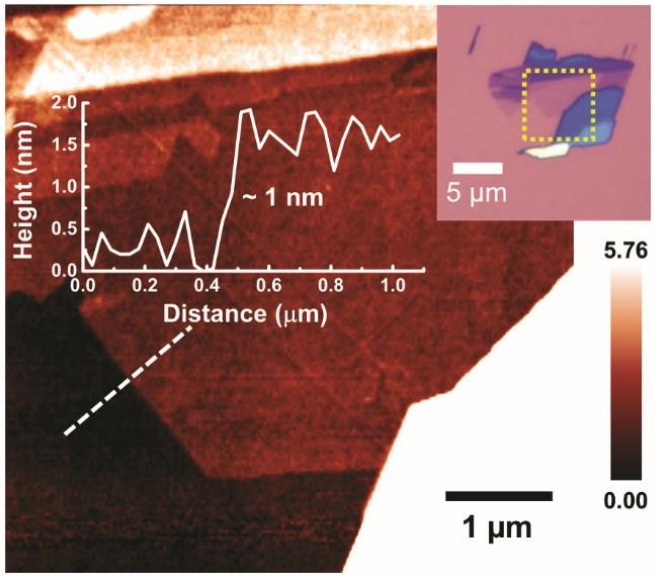
How about XXZ-type antiferromagnet, $NiPS_3$, in the 2D limit?

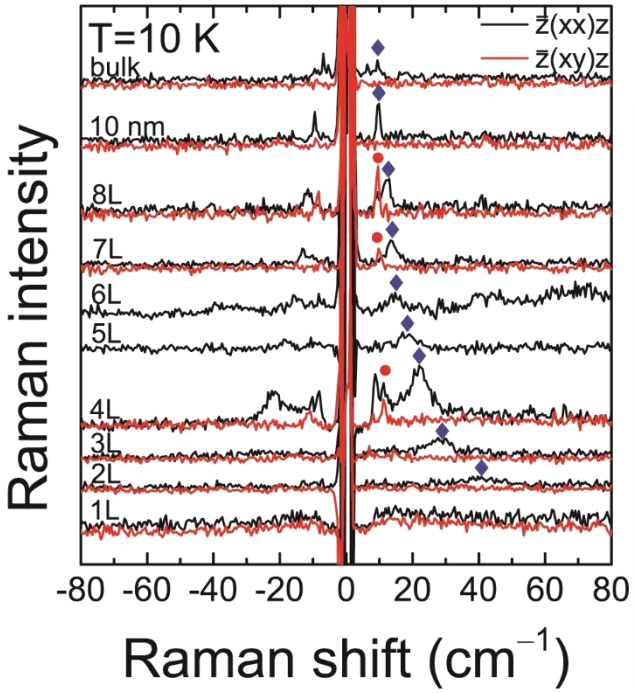
J.-U. Lee *et al., Nano Lett.* **16**, 7433 (2016).
Also, Wang *et al.*, 2D Mater. (2016).

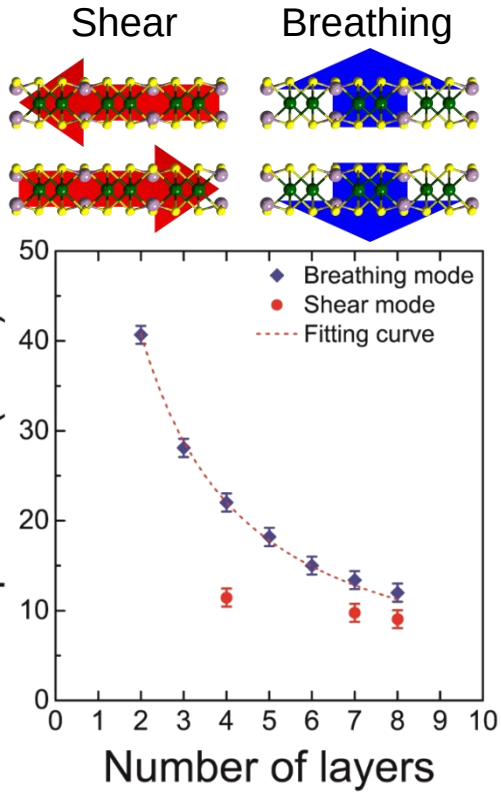
- Ising-type antiferromagnet $FePS_3$ exhibits magnetic ordering even down to the monolayer limit and the transition temperature is almost independent of the thickness.
- This result is in good agreement with the Onsager solution for 2-dimensional order−disorder transition

02 Method

Sample preparations & Raman measurements



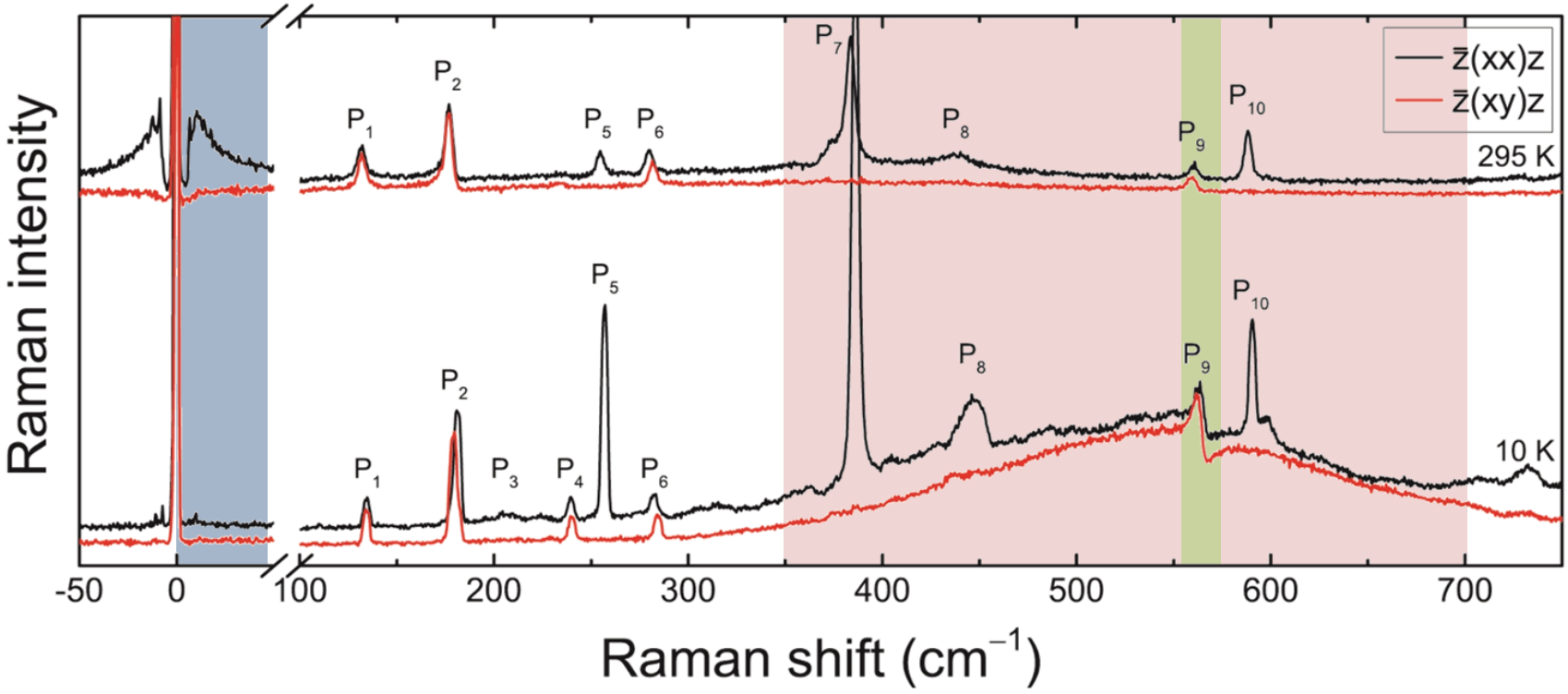




- Bulk single-crystal $NiPS_3$ is synthesized by vapor transport method (CVT).
- Mechanically exfoliated on typical SiO_2/Si substrate.
- Temperature dependent polarized Raman measurements are performed with home-built Raman systems with 514.5 nm excitation source.

03 Results & Discussion

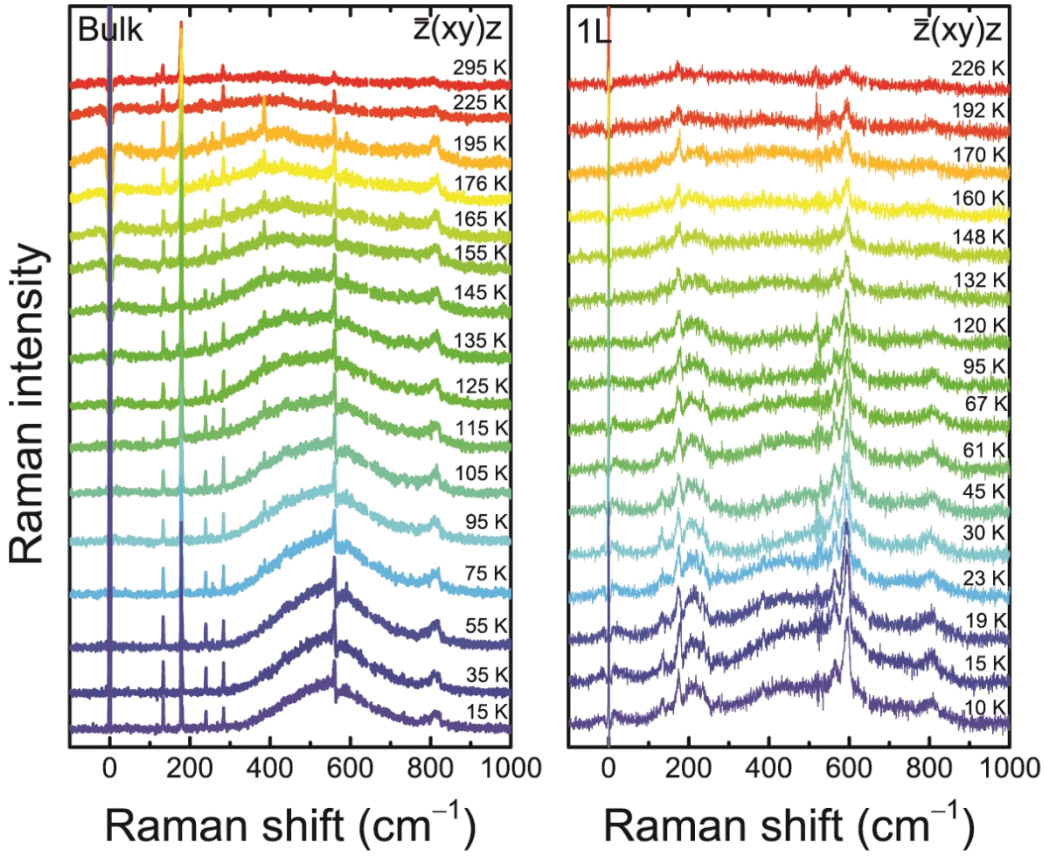
Signatures of magnetic ordering in Raman spectra of bulk $NiPS_3$

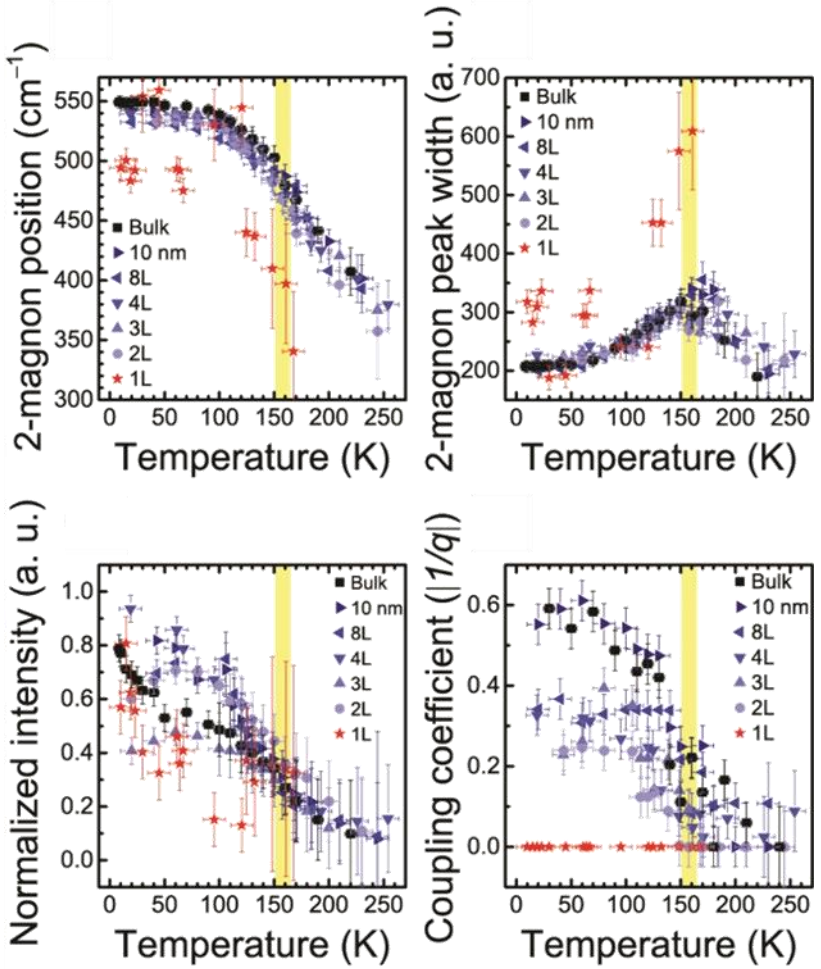


- Appearance of **2-magnon scattering**
- Appearance of **Fano resonance**
- Suppression of low-frequency **quasi-elastic scattering** due to spin fluctuation

03 Results & Discussion

Temperature dependence of the 2-magnon & Fano resonance



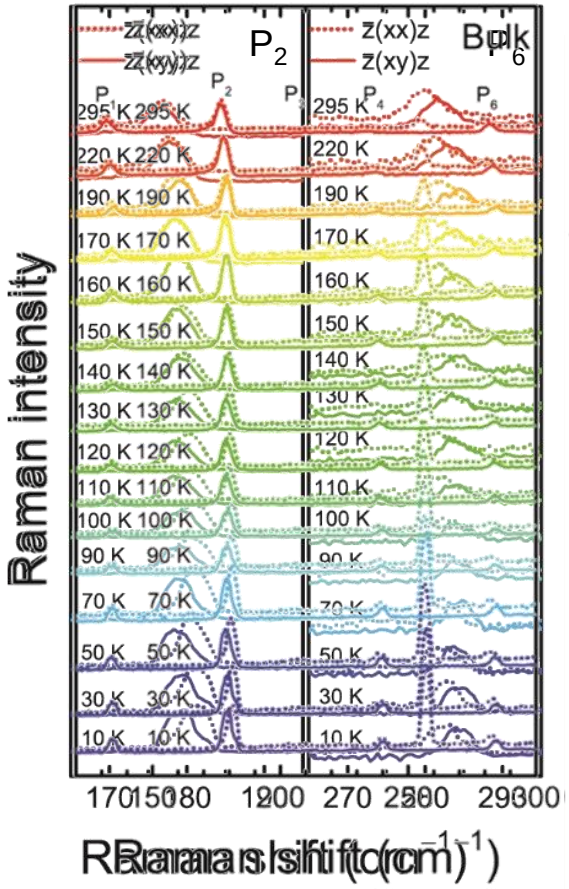


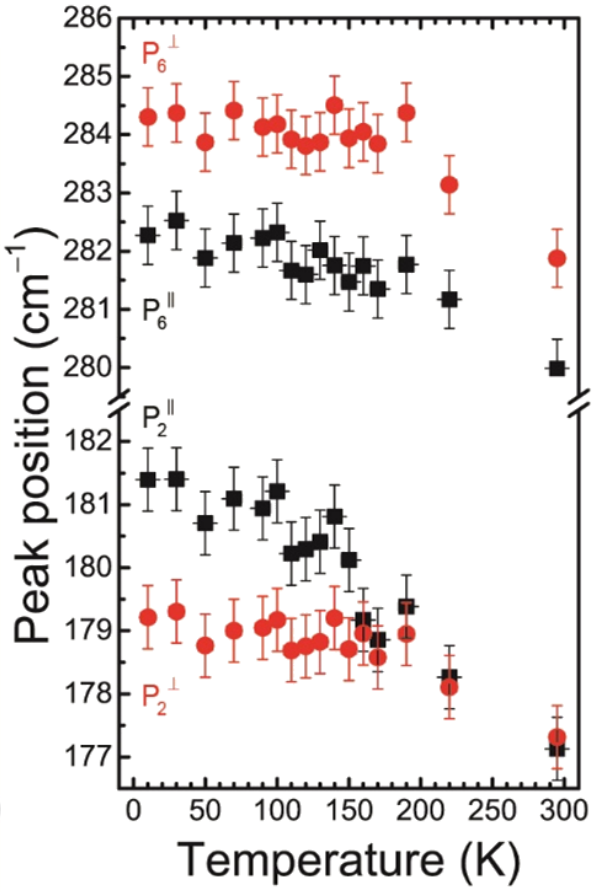
- Few-layer $NiPS_3$ showed similar trend as bulk $NiPS_3$, except 1L $NiPS_3$.

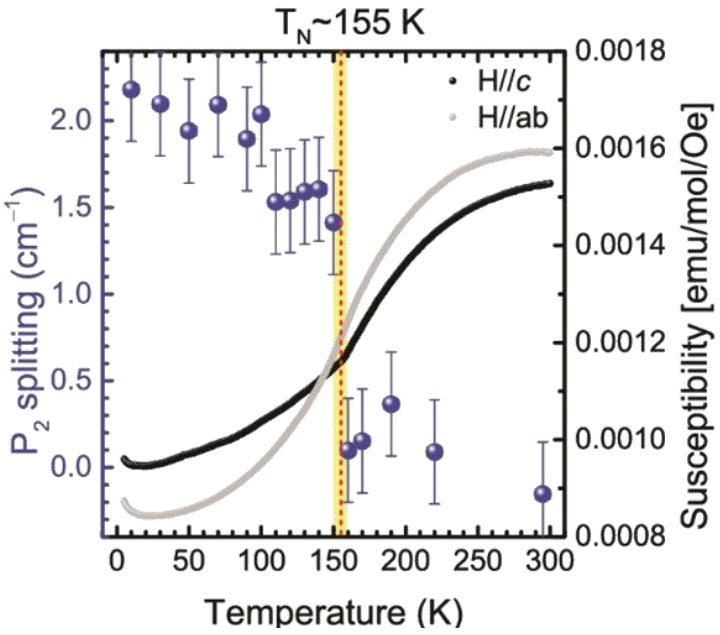
03 Results & Discussion

Phonon splitting due to antiferromagnetic phase transition

Bulk $NiPS_3$



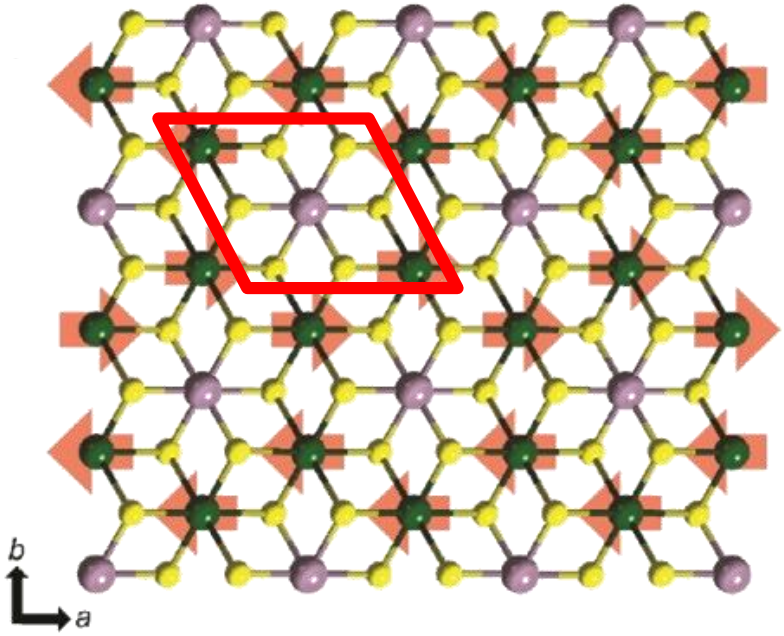


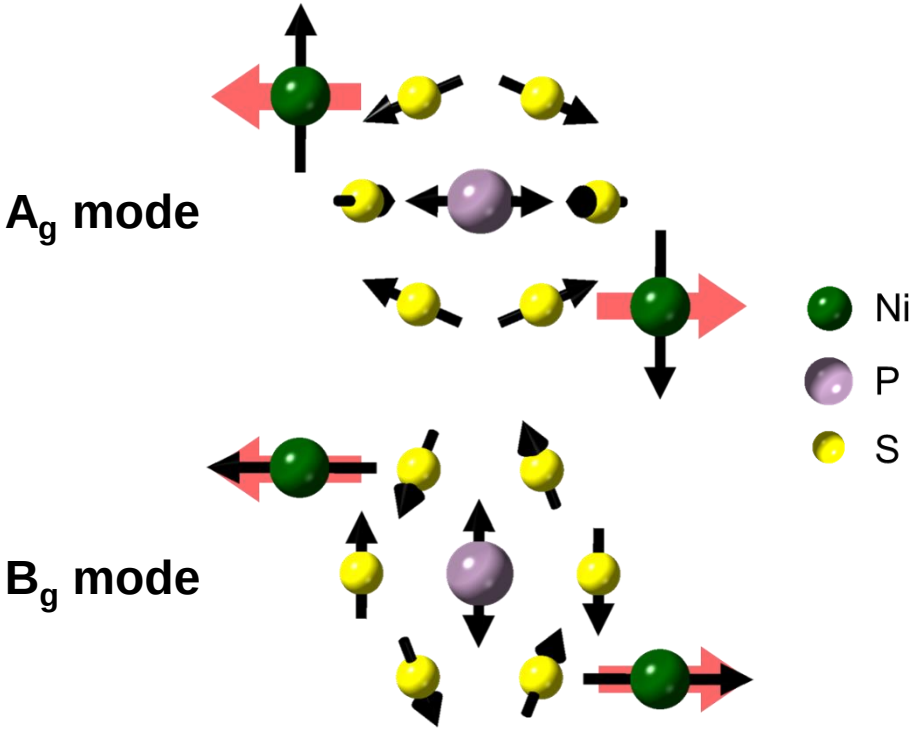


➔ By using the phonon splitting of P_2, Néel temperature of $NiPS_3$ can be estimated.

03 Results & Discussion

Phonon splitting due to antiferromagnetic phase transition



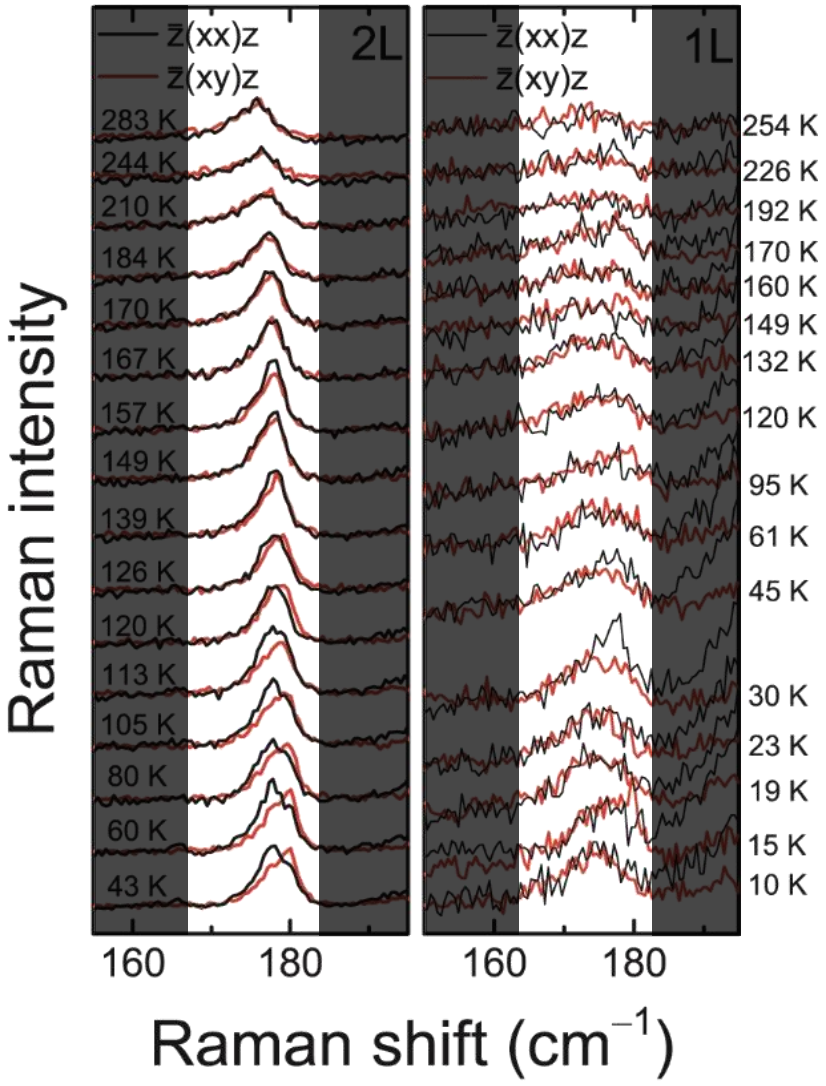


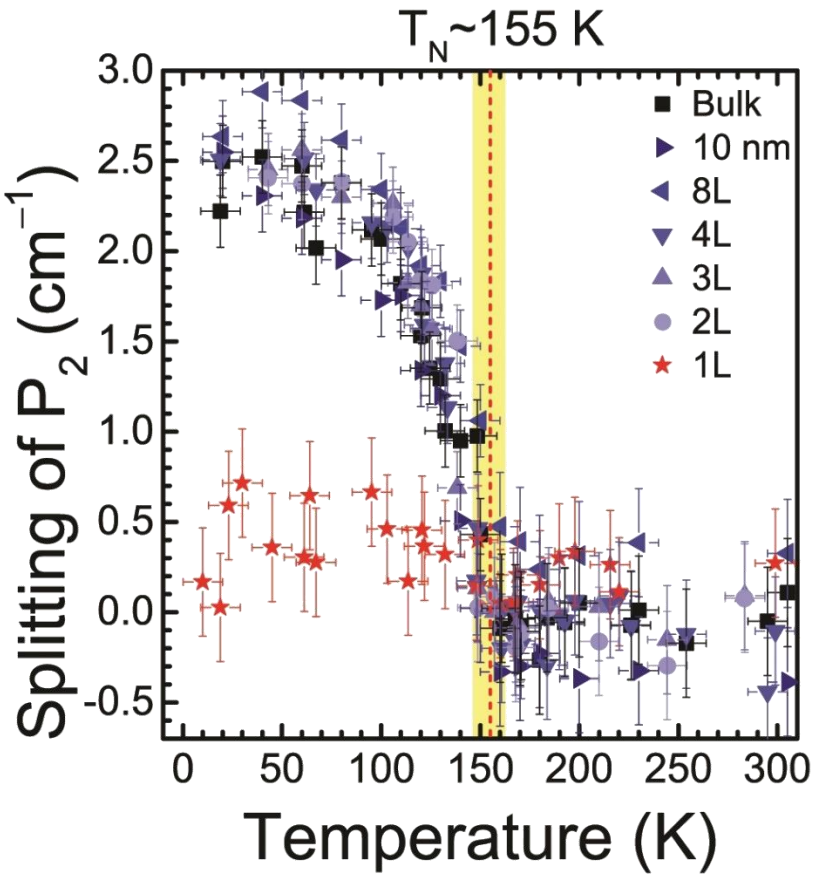
- **Black** arrows: vibration direction
- **Red** arrows: distribution of spin polarization below T_N

➔ Phonon splitting of P_2 is due to relative directions of the vibration and the distribution of the spin polarizations.

03 Results & Discussion

Thickness dependence of Néel temperature





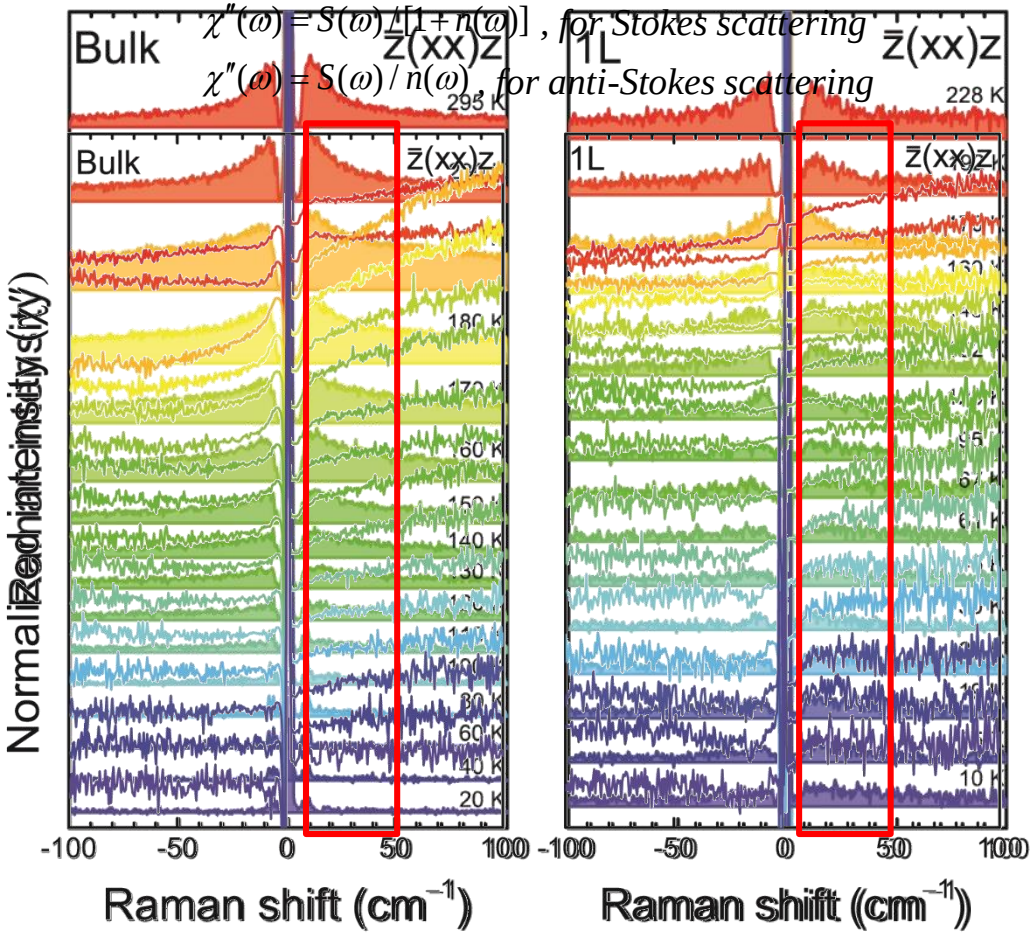
➔ T_N is almost constant for thicknesses down to 2L, but no magnetic phase transition is observed for 1L.

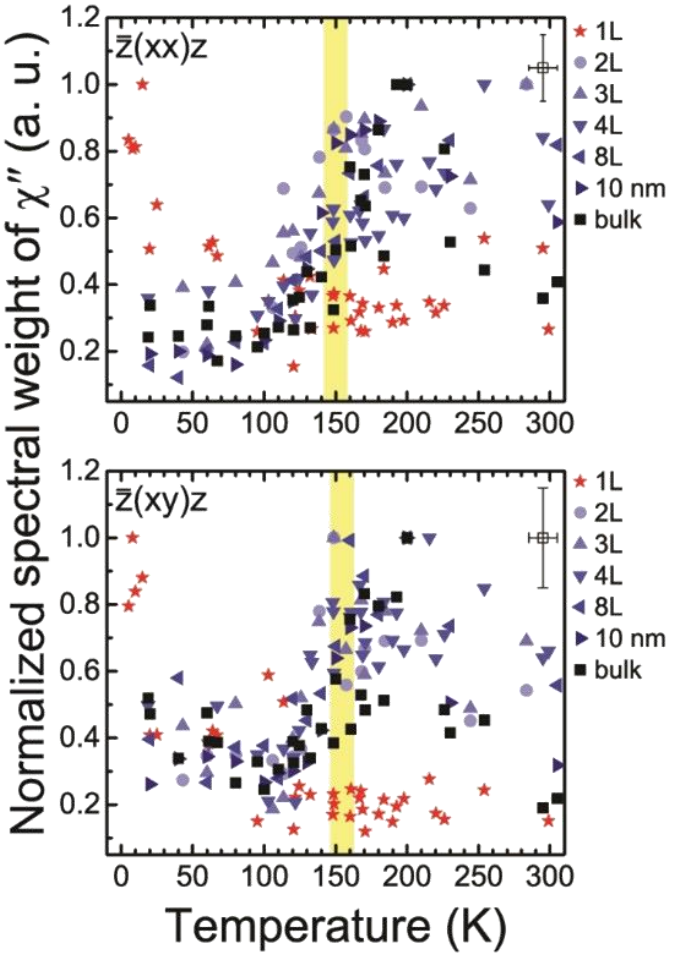
03 Results & Discussion

Temperature dependence of the quasi-elastic scattering (QES)

$\chi''(\omega) = S(\omega)/[1+n(\omega)]$, *for Stokes scattering*

$\chi''(\omega) = S(\omega)/n(\omega)$, *for anti-Stokes scattering*





➔ It also supports that T_N of few-layer $NiPS_3$ is almost constant down to 2L, but there is no magnetic phase transition for 1L.

04 Summary

- $NiPS_3$ 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-type antiferromagnet, the magnetic ordering becomes very fragile in the true 2D limit of 1-layer.

Thank you for your attention