北京理工大学
BEIJING INSTITUTE OF TECHNOLOGY

Dysurf: A program for simulating four-dimensional dynamical structure factor for inelastic neutron scattering measurement

Jiawang Hong

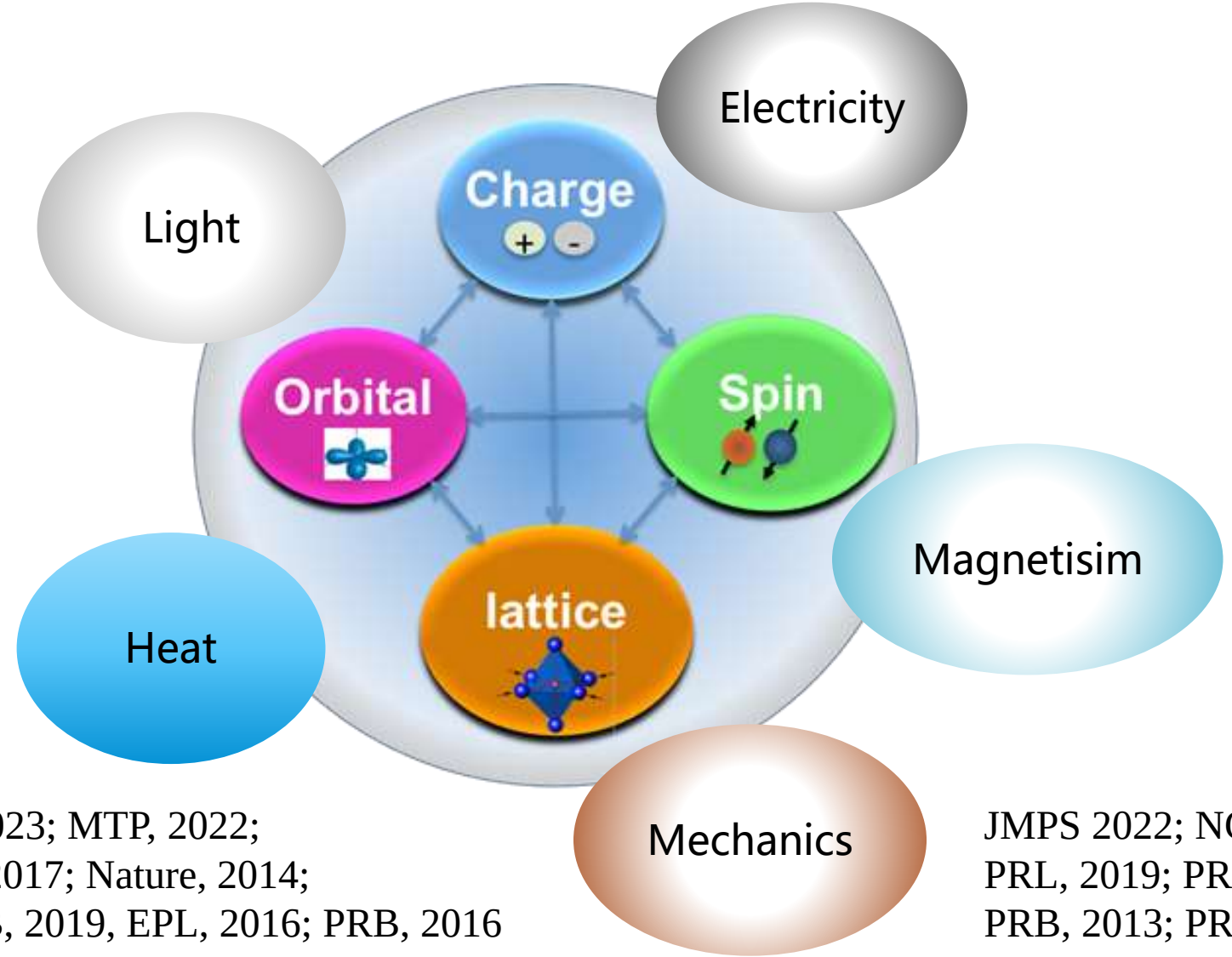
Beijing Institute of Technology

Bin Wei

Henan Polytechnic University

2023.11.01

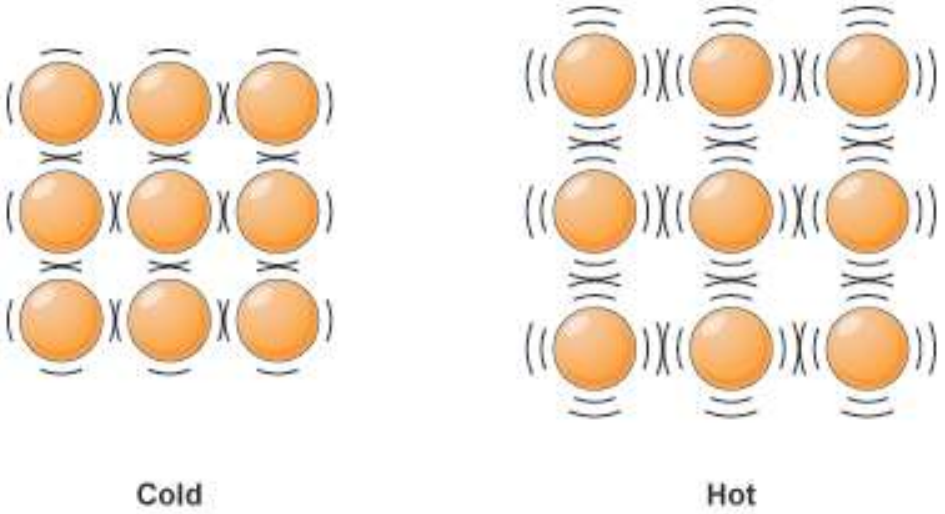
Multi-field couplings in advanced materials (IXS/INS + DFT + AFM)

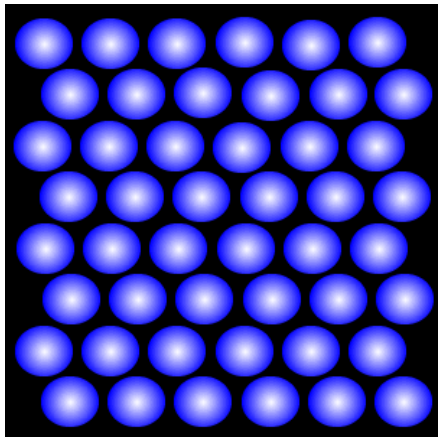


npj Compt. Mater., 2023; MTP, 2022;
PRB 2021; Science, 2017; Nature, 2014;
Nat. Phys. 2015; PRB, 2019, EPL, 2016; PRB, 2016

JMPS 2022; NC 2022; Nat. Mater. 2020;
PRL, 2019; PRL, 2015; APL, 2016;
PRB, 2013; PRB, 2012;

Atomic vibrations in solids





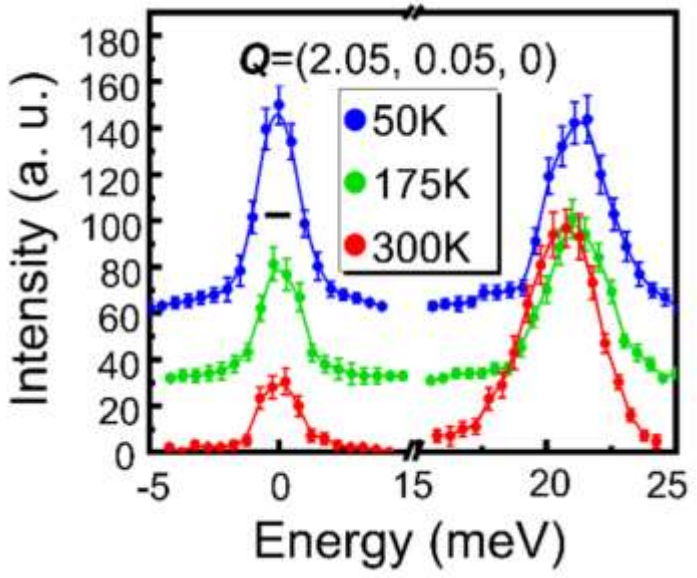
- **Lattice dynamics is essential for fundamental understanding of the structure, thermodynamics, phase stability, and thermal transport properties of solids.**

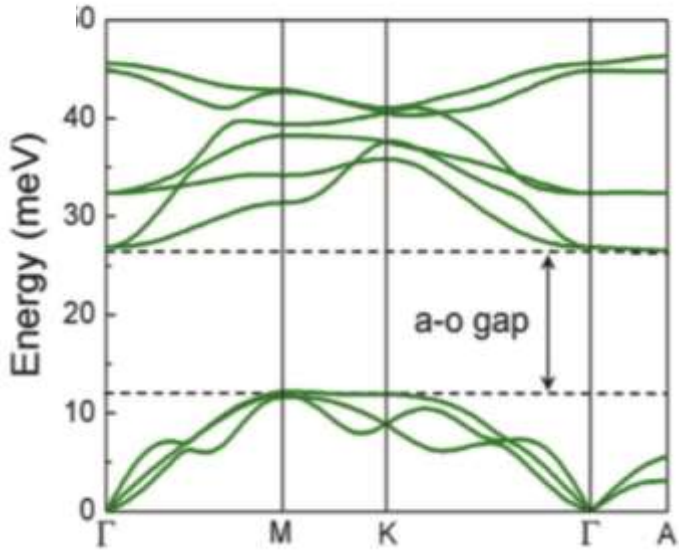
- **Electronic, optical, magnetic, structure phase transition, superconductivity etc.**

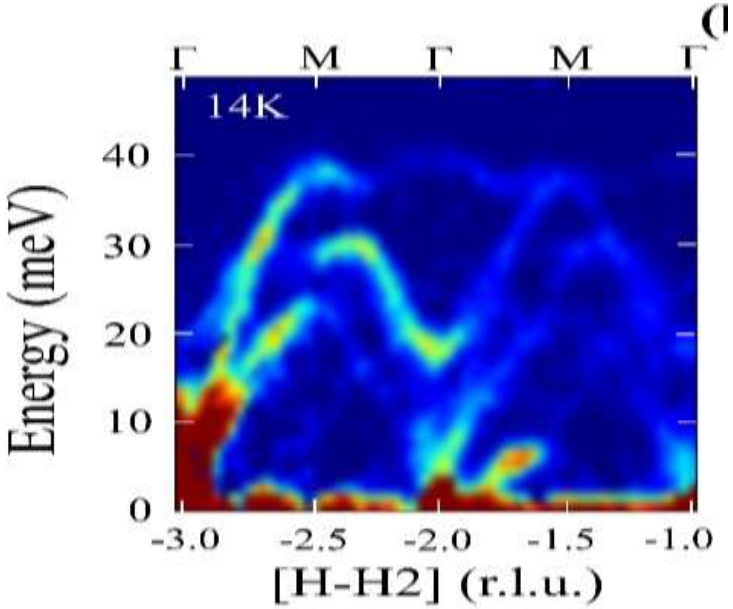
M. Born and K. Huang, Dynamical Theory of Crystal Lattices, 1954

Phonon measurements from IXS and INS

- IXS and INS are powerful to measure phonons
- Time consuming
- Challenge to find suitable BZ, specific phonon branch; huge data





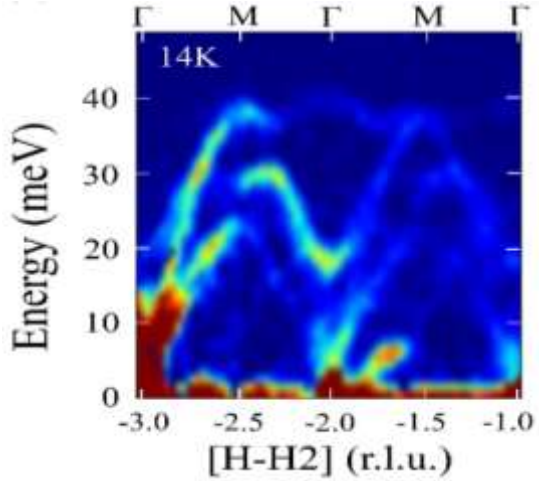


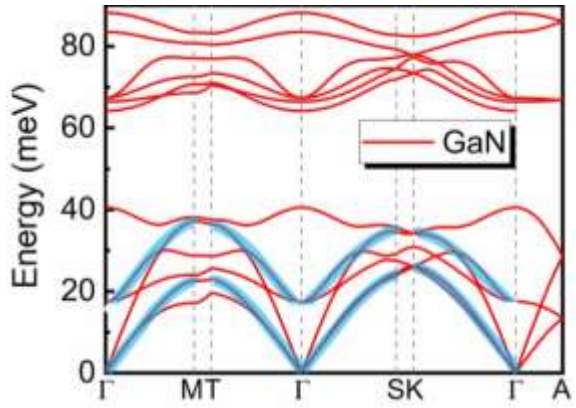
small single crystal : 10^{-3} gram (HP)
time : ~days

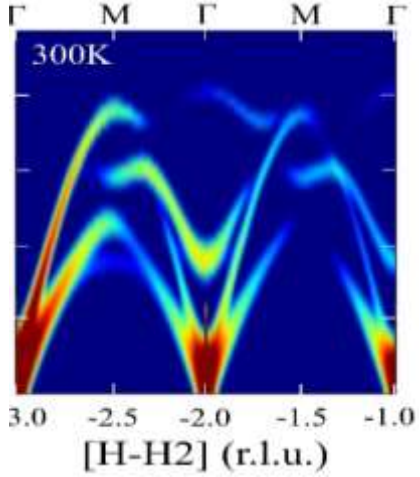
Large single crystal : ~ gram
huge data: ~GB
time : ~days

Development of Dysurf for S(Q,E) simulation

- Fortran code, open source, detailed manual
- Support multi-platform: Windows, Linux, Mac; multi-interface: VASP, QE, Abinit, etc.
- Deal with 1D，2D，3D, 4D INS and IXS data







- Develop Dysurf to calculate Dynamic Structure Factors S(**Q**,E)) to directly compare with measurement :

$$S(\mathbf{Q},E) \propto \sum_s \sum_\tau \frac{1}{E_s} \left| \sum_d \frac{\bar{b}_d}{\sqrt{M_d}} e^{(-W_d + i\,\mathbf{Q}\cdot\mathbf{r}_d)} (\mathbf{Q}\cdot\epsilon_{ds}) \right|^2 \times \langle n_s + 1 \rangle \, \delta(E - E_s) \, \delta(\mathbf{Q} - \mathbf{q} - \tau) \,,$$

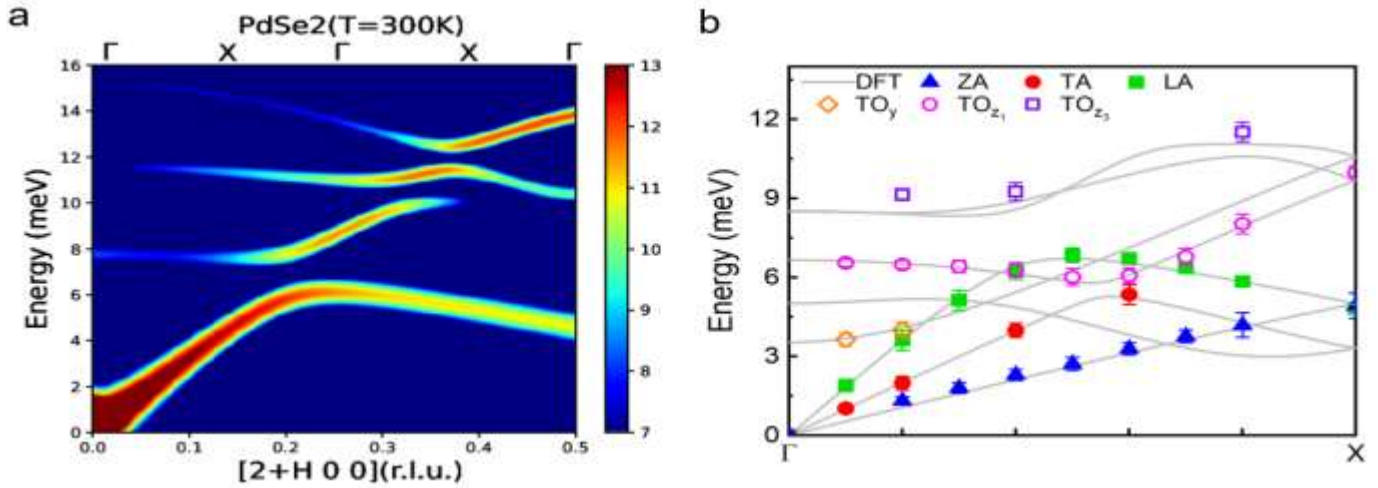
$$I_{calculated}(\bar{Q}_0, E_0) = \int_{\Delta\bar{Q}=-\infty}^{\infty} \int_{\Delta E=-\infty}^{\infty} S(\bar{Q}_0 + \Delta\bar{Q}, E_0 + \Delta E) \times R(\Delta\bar{Q}, \Delta E) d\Delta\bar{Q} d\Delta E$$

Instrument resolution

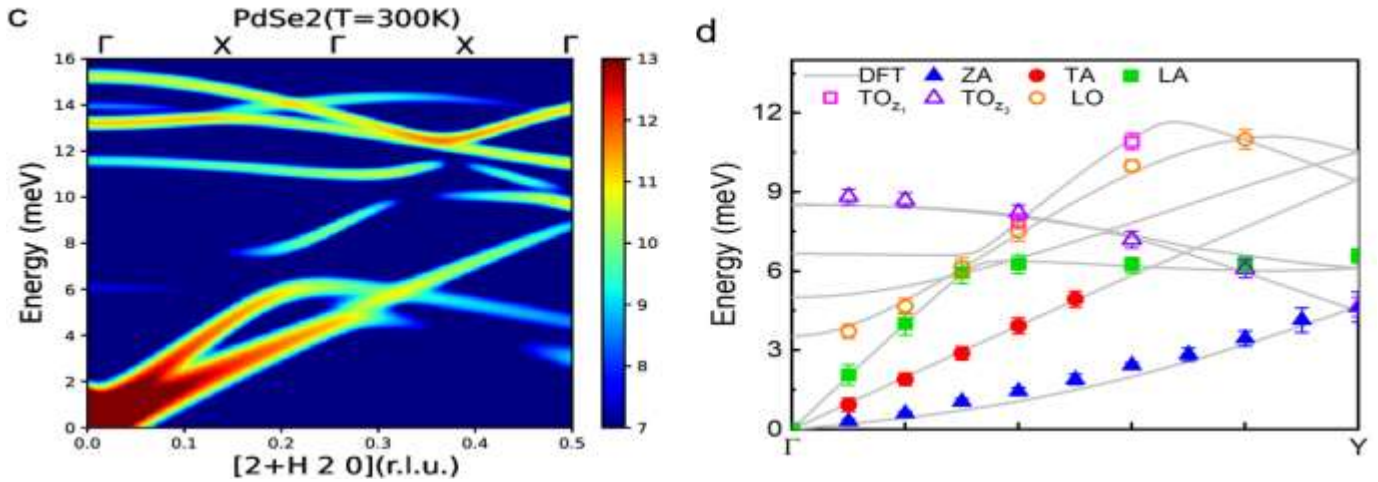
BZ and phonon branch selection(I) -1D

- **Measure phonon branches as many as possible in one scan to save beamtime**

[2+k,0,0]



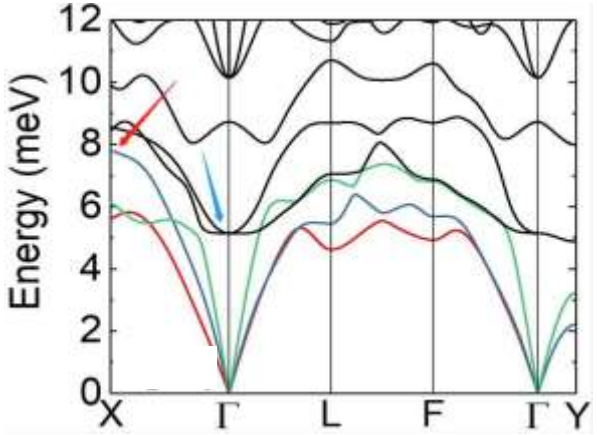
[2+k,0,2]



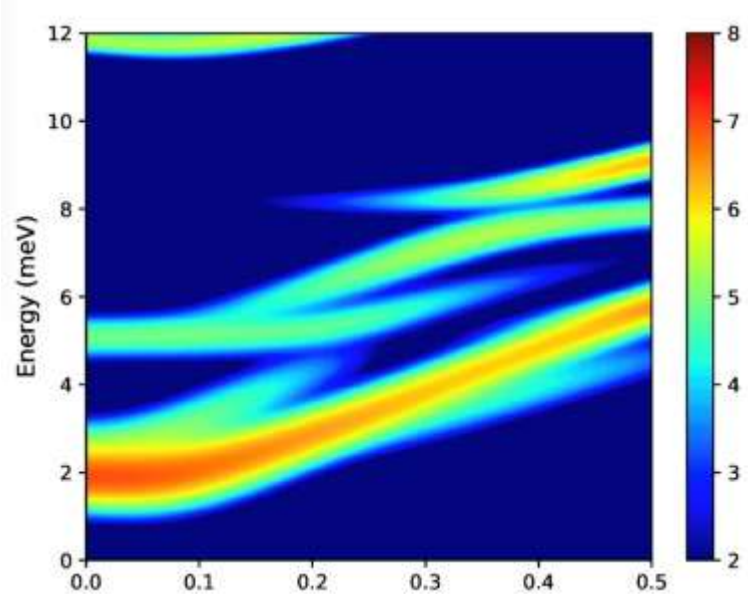
save up to 60% beamtime

BZ and phonon branch selection (II) -1D

- Measure LA phonons
- Q range [100] or [101]

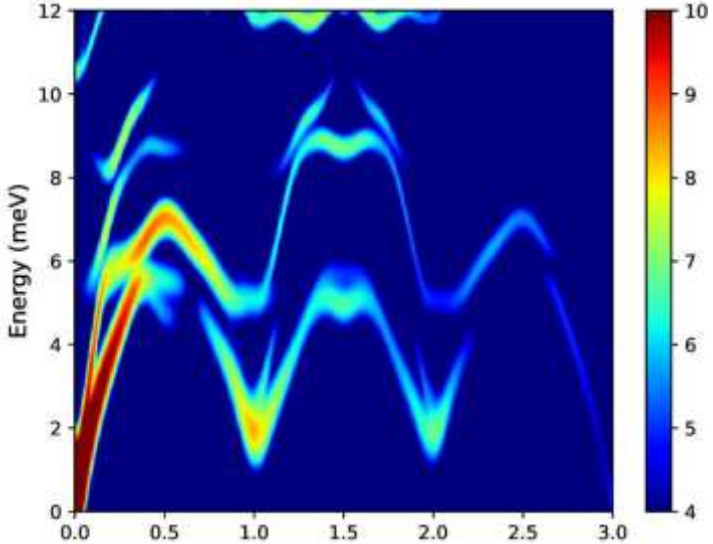


[1+k, 0, 0]



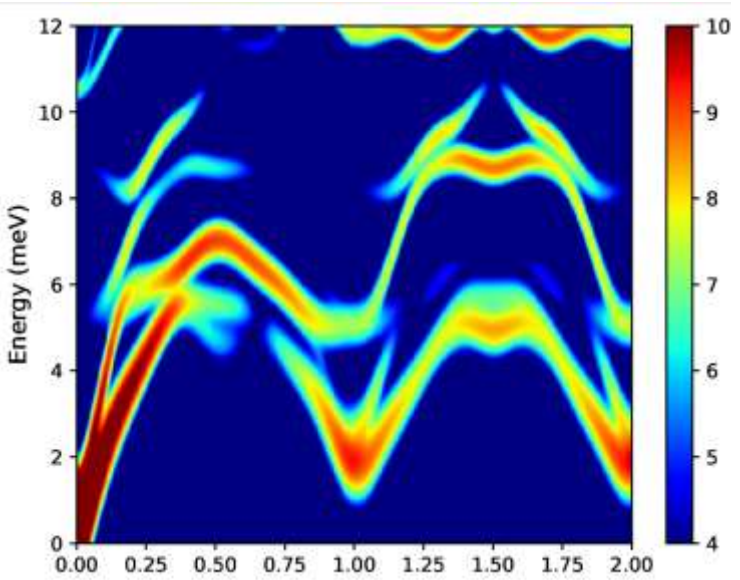
[1 0 0] [1.5 0 0]

[3-k, 0, 0]



[3 0 0] [2 0 0] [1 0 0] [0 0 0]

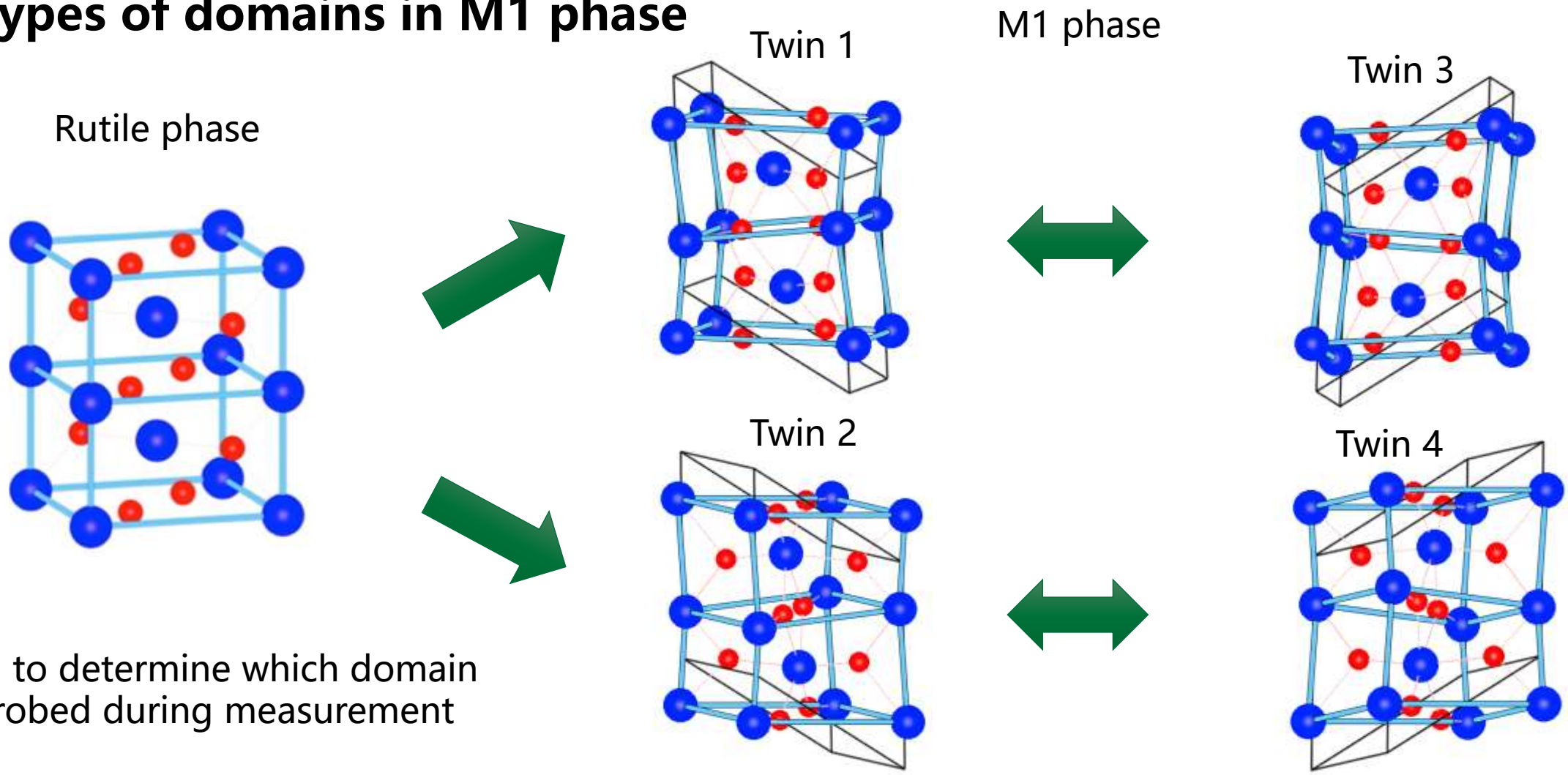
[1+k, 0,1]



Domain twins in M1 phase -2D

北京理工大学 BEIJING INSTITUTE OF TECHNOLOGY

4 types of domains in M1 phase



Need to determine which domain we probed during measurement

TDS in {111} plane -2D

北京理工大學 BEIJING INSTITUTE OF TECHNOLOGY

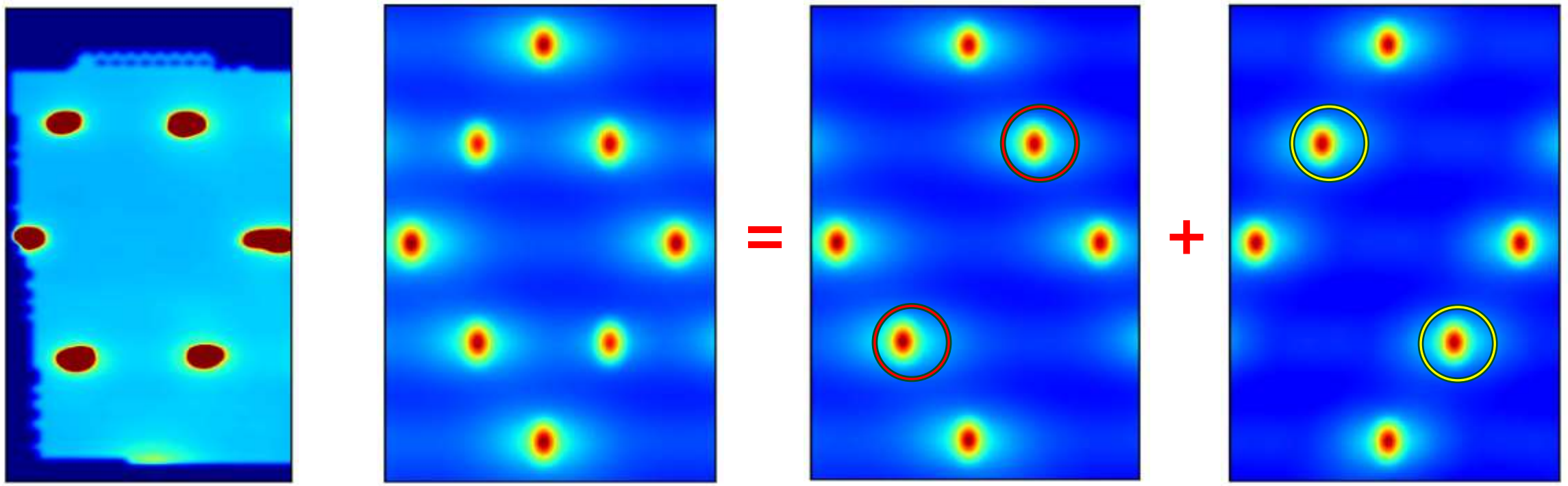
- **4 types of domains have to be considered to obtain full TDS in M1 phase**

Experiment
beamline 33-BM @APS

Theory
full

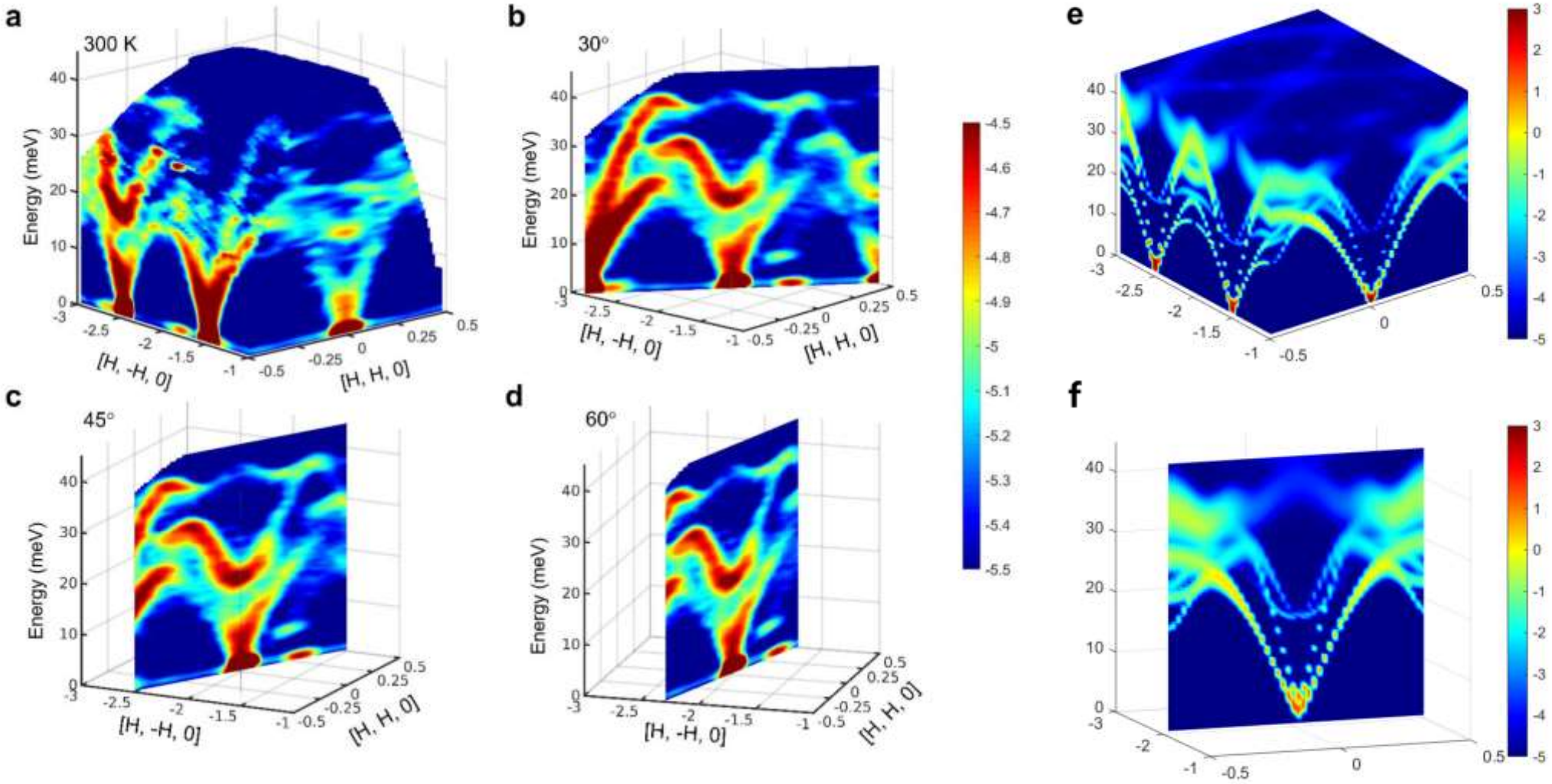
Theory
Twins 1&3

Theory
Twins 2&4



Exp. data from E. D. Specht, J. D. Budai, O. Delaire et al.

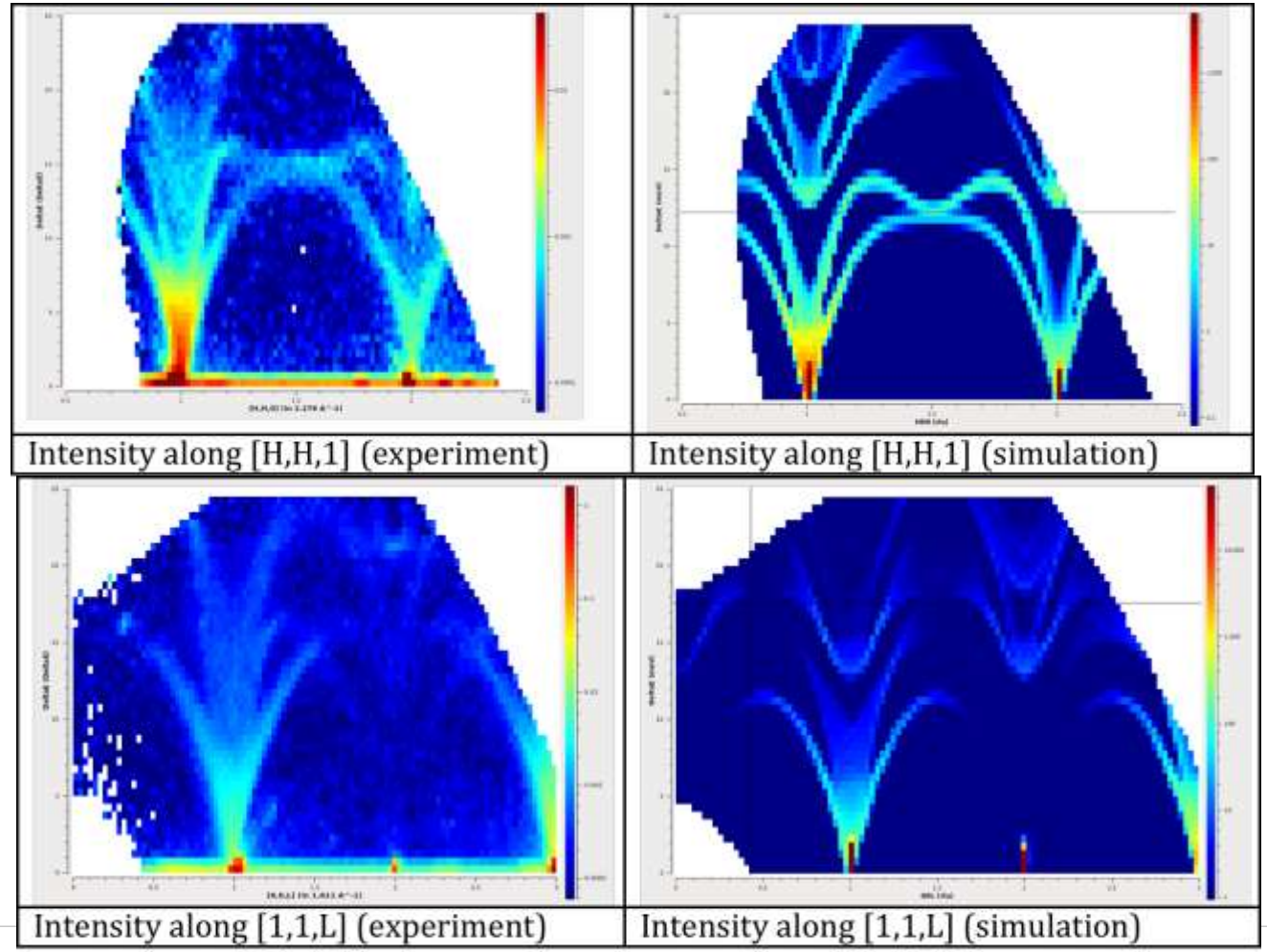
Matryoshka Phonon Twinning- 3D



Synchronization of INS and simulations

Measurements

Calculations



Intensity along [H,H,1] (experiment)

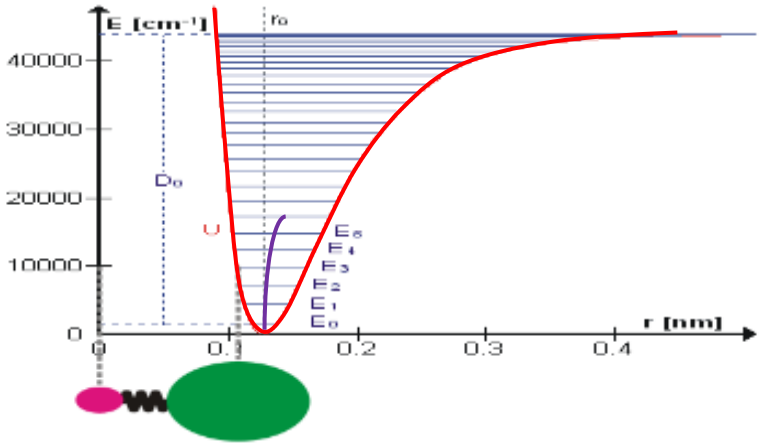
Intensity along [H,H,1] (simulation)

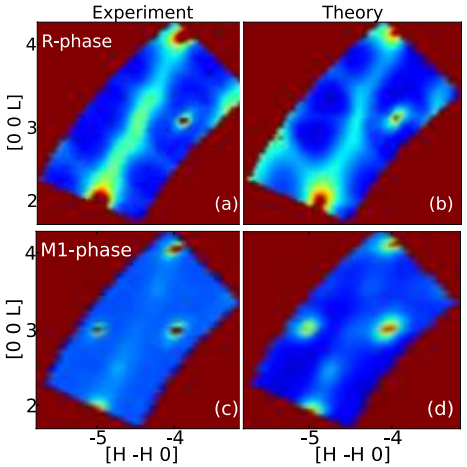
Intensity along [1,1,L] (experiment)

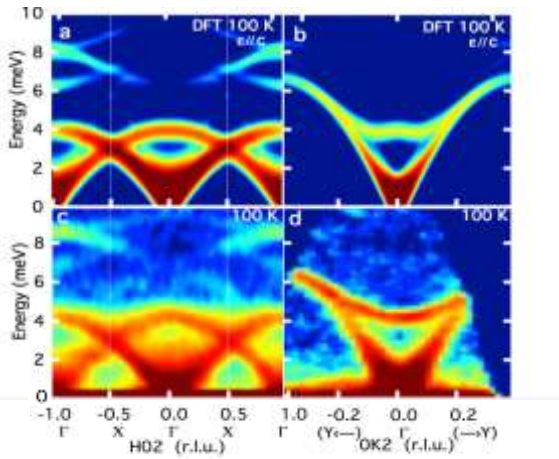
Intensity along [1,1,L] (simulation)

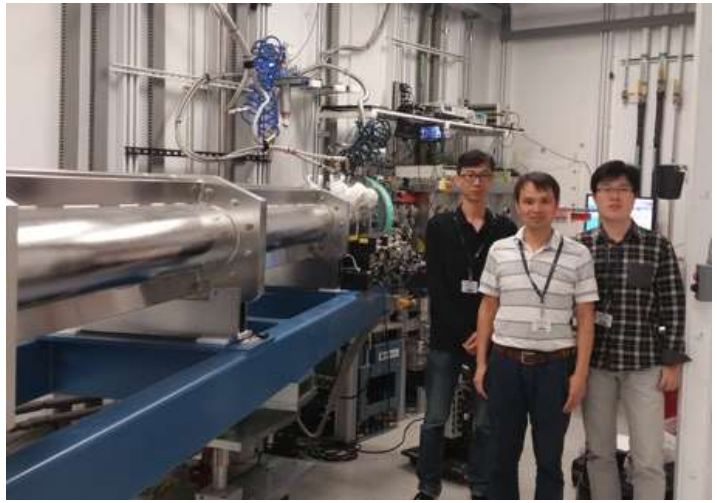
Some examples

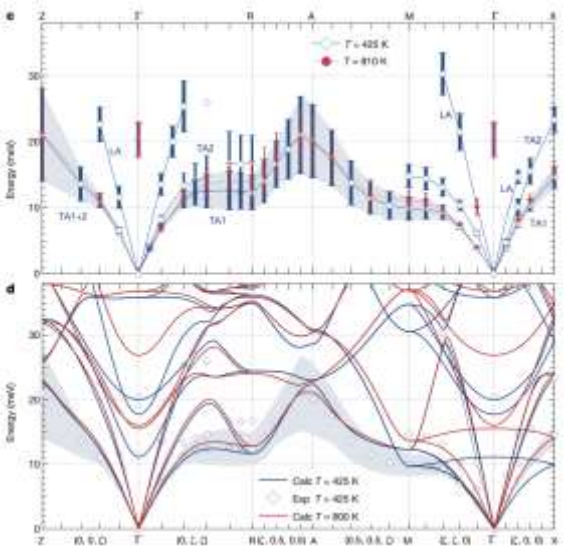
Anharmonic Phonons in Advanced Materials

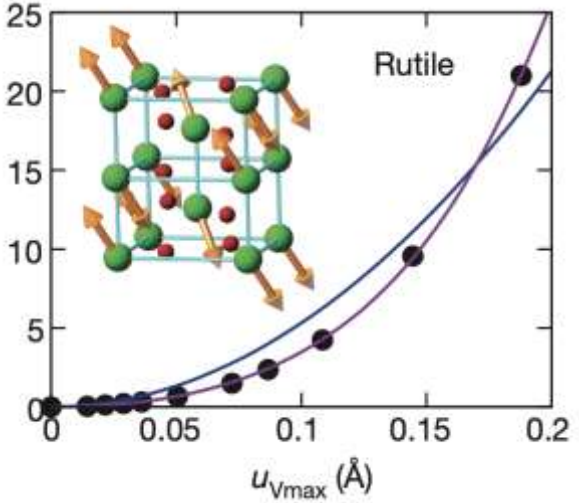












德以明理　学以精工

Science 355, 371 (2017); Nature, 515, 535 (2014); Nature Physics, 11, 1603 (2015); Materials Today Physics, (2019, 2021,2022), Advanced Science (2021)

Thermal transport in GaN

- GaN, 3rd generation semiconductor
- Thermal conductivity anisotropy is unclear

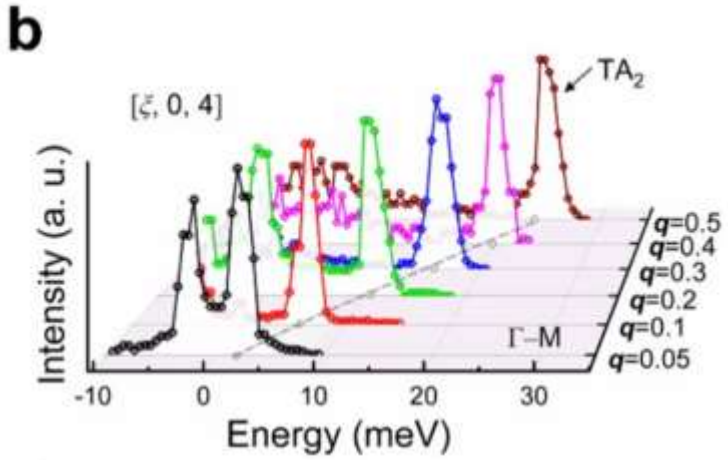
Anisotropic thermal conductivity in GaN

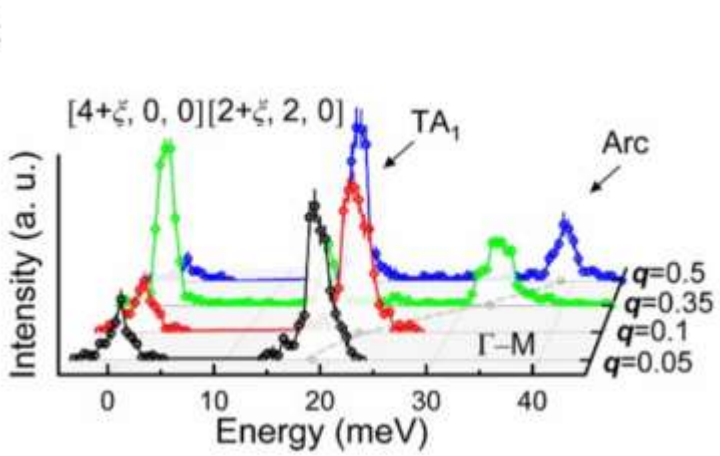
Work	In-plane κ		Out-of-plane κ	
Lindsay L	242		239	
Yang J	278	>	342	
Wu R	228	?	260	
Yang X	228	223	< 261	243

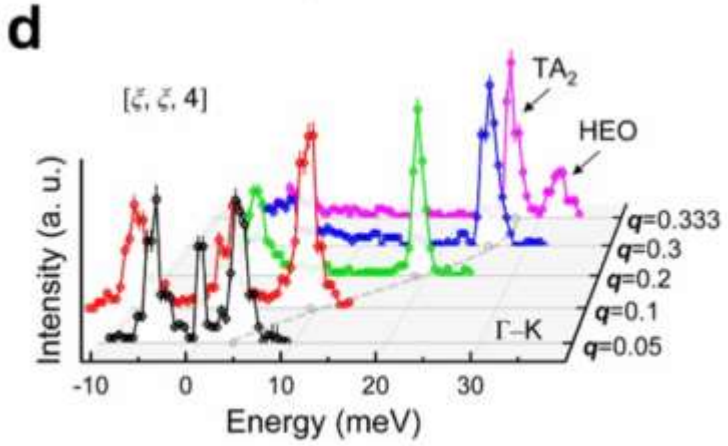
PRL 109, 095901 (2012); **APL** 109, 242103 (2016)
JAP 119, 145706 (2016); **PRB** 100, 245203 (2019)

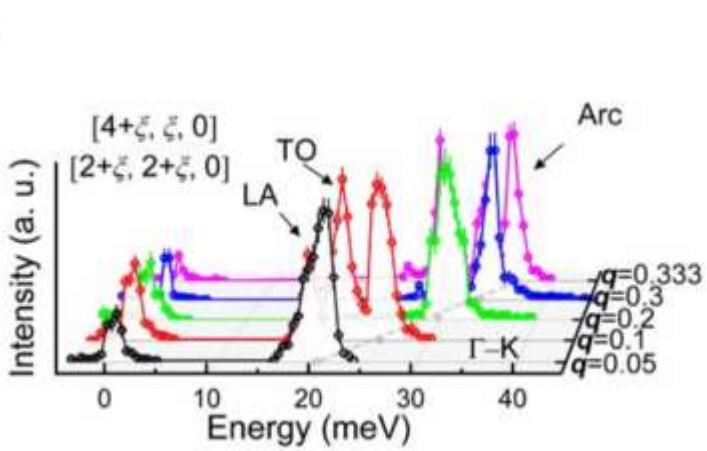
IXS measurement of GaN

- Calculate S(Q,E) to select specific BZ
- Good statistics

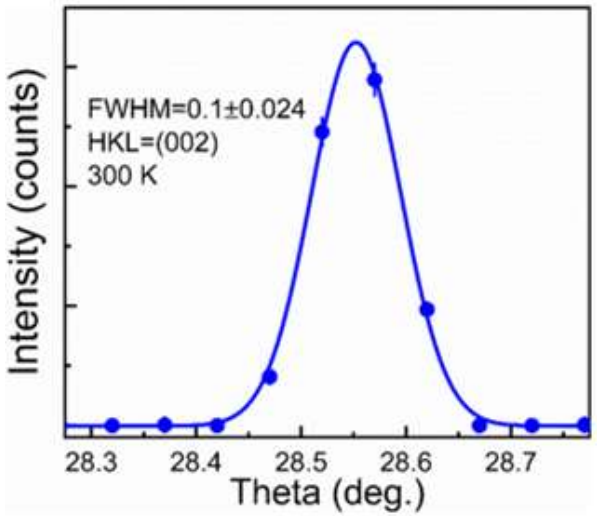






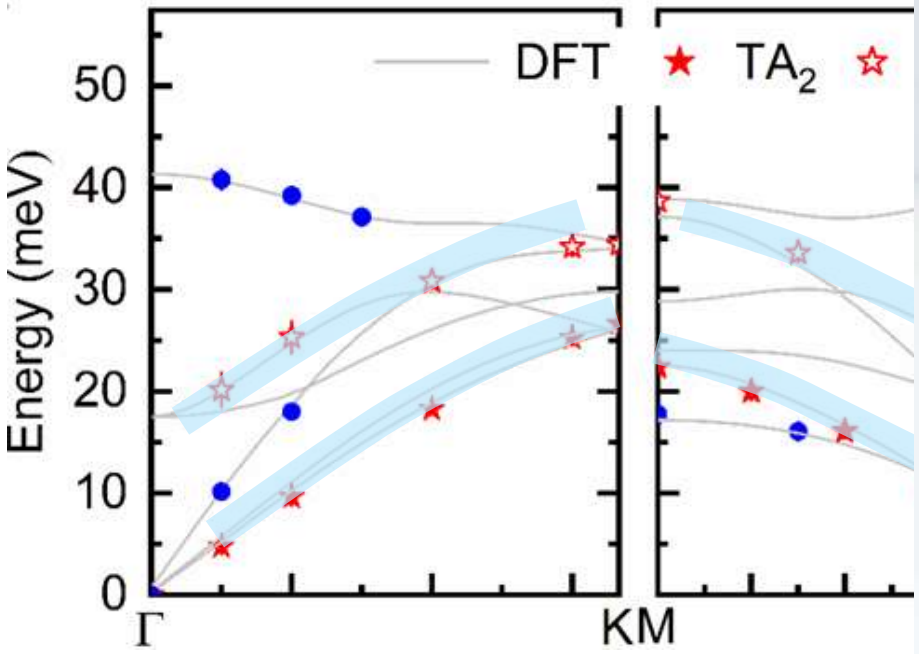


High sample quality



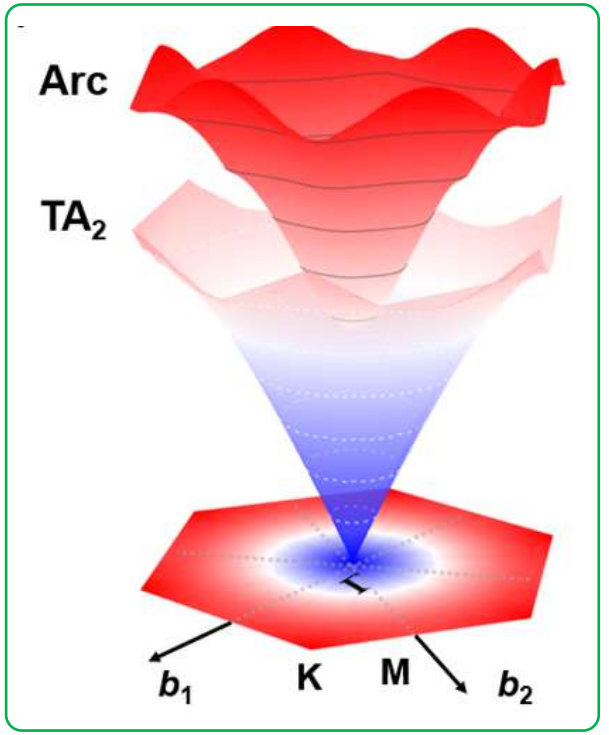
Matryoshka twinning from IXS and DFT

- Phonon nesting along Γ–M and Γ–K
- DFT shows surface nesting in 3D



In-plane

DFT

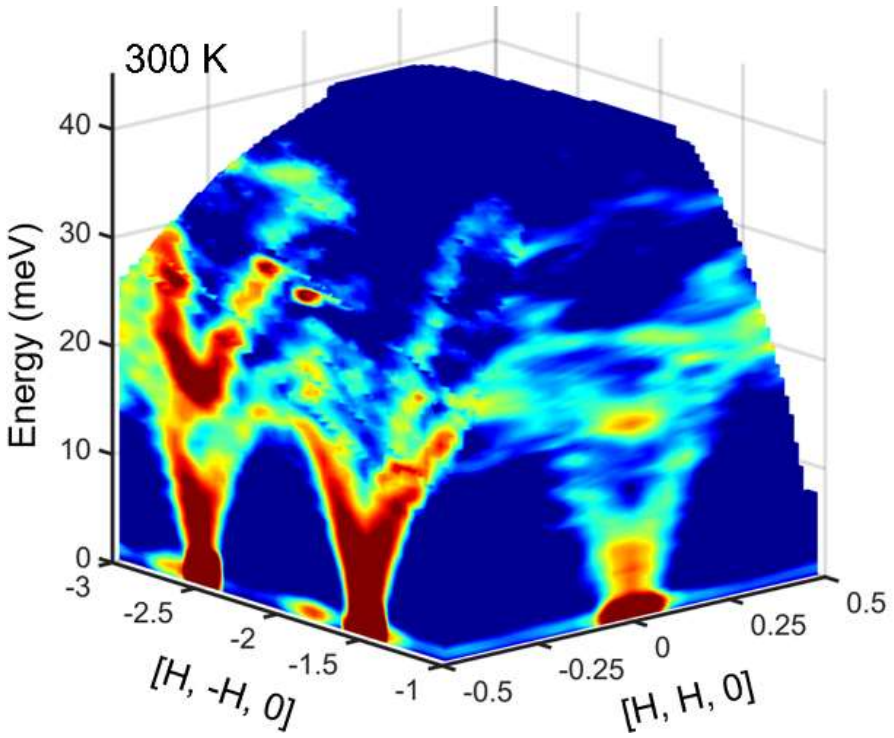


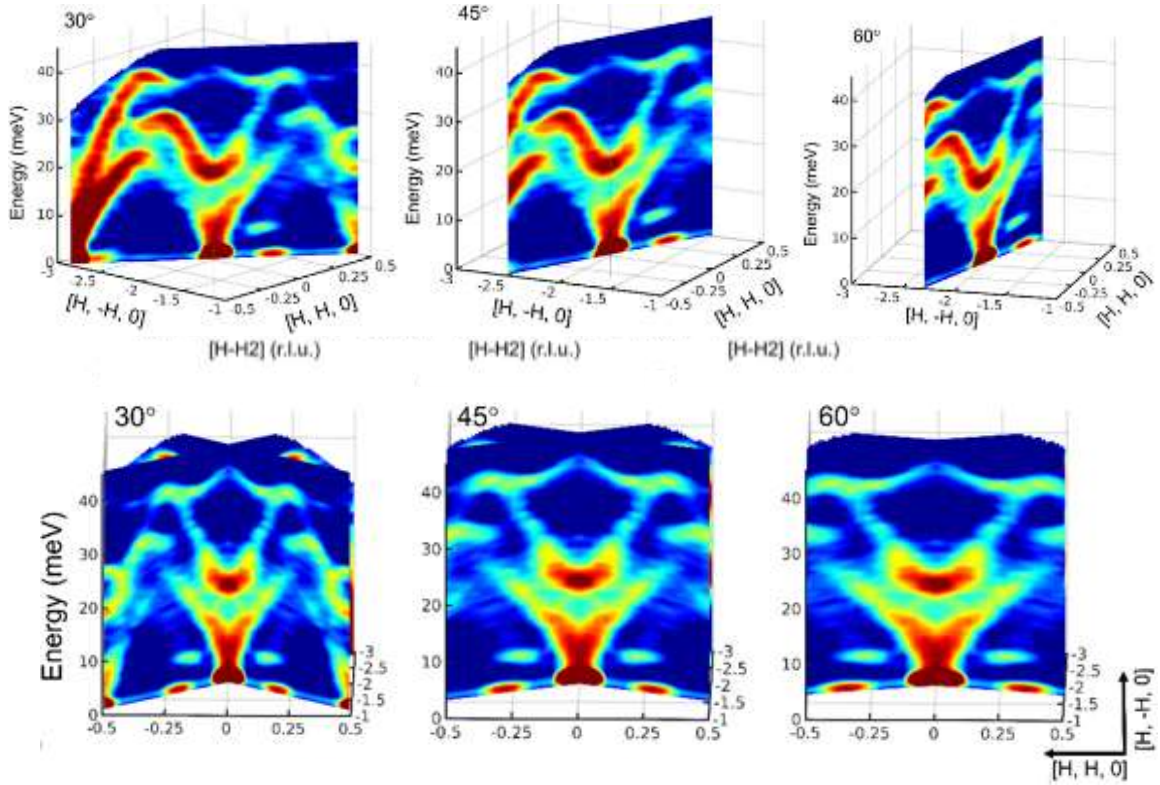
德以明理 学以精工

Wei, Hong et al, Communications Physics 4, 227 (2021)

Matryoshka twinning from 4D INS data

- **4D INS data from TOF confirms the Matryoshka twins**





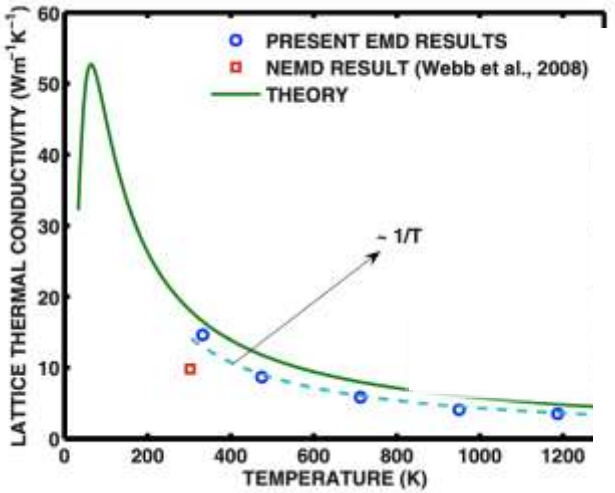
德以明理 学以精工

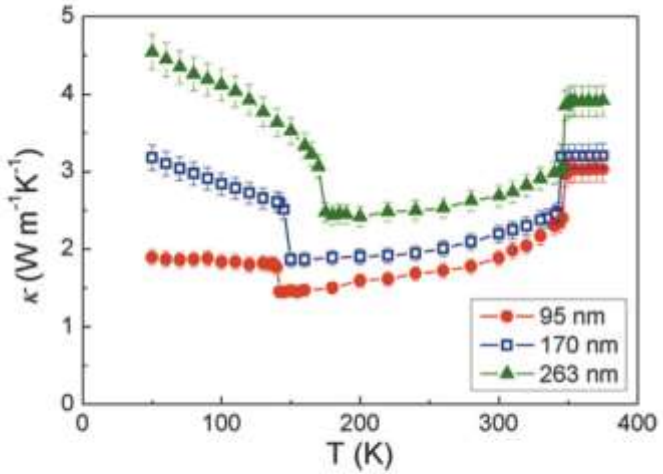
Wei, Hong et al, Communications Physics 4, 227 (2021)

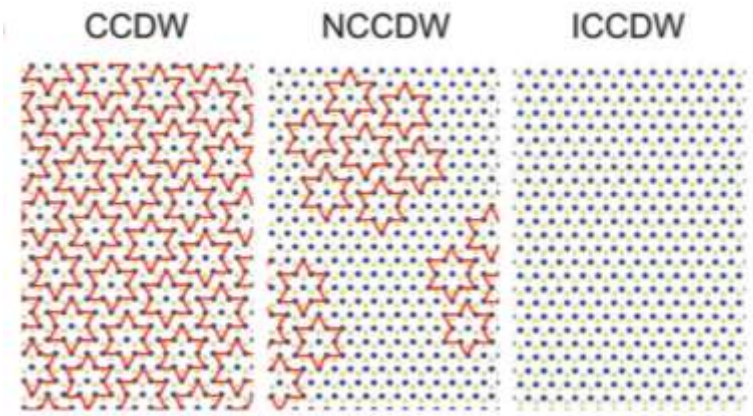
T independence of lattice thermal conductivity

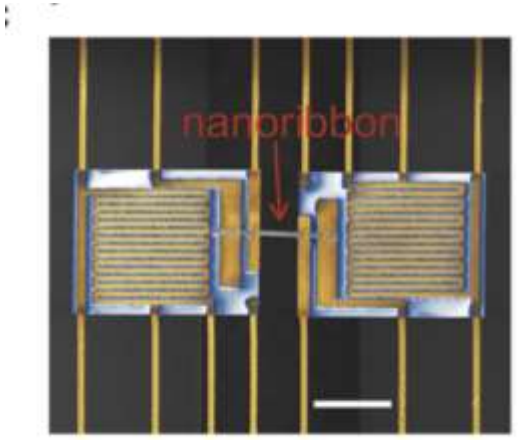
- CDW materials TaS2, showing commensurate structure
- Nearly T independence of lattice thermal conductivity

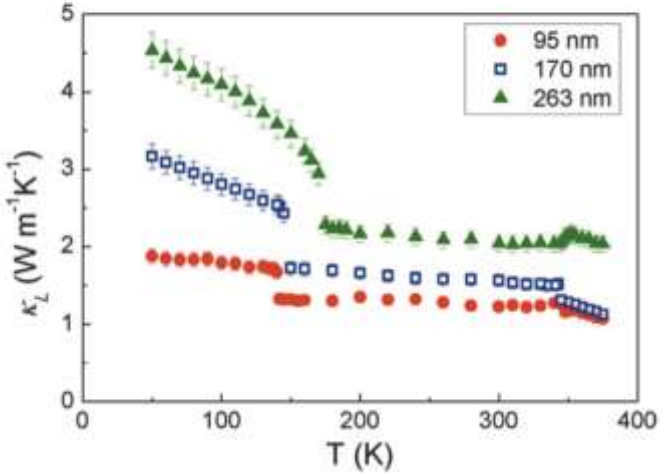
Why?









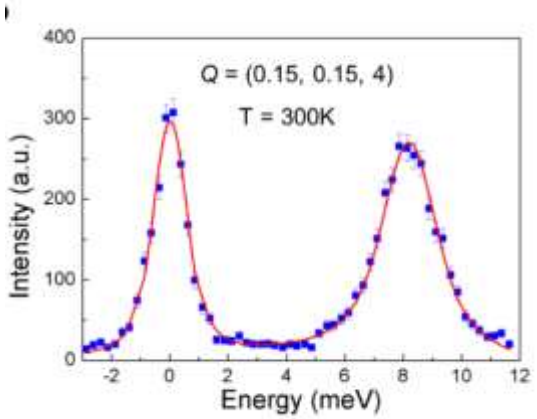


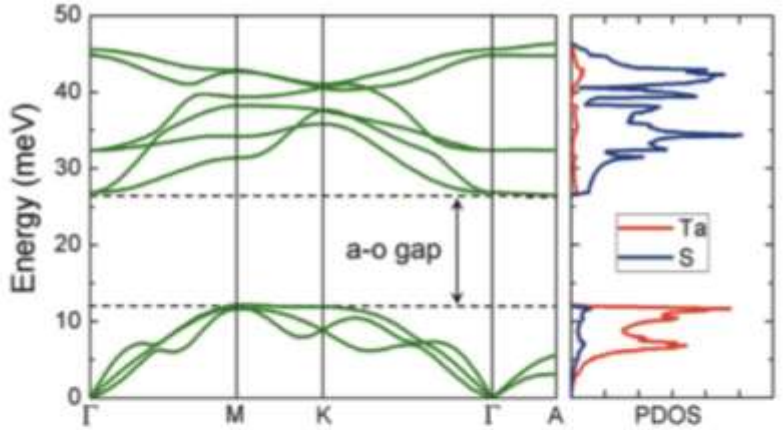
Feng et al, JAP (2009)

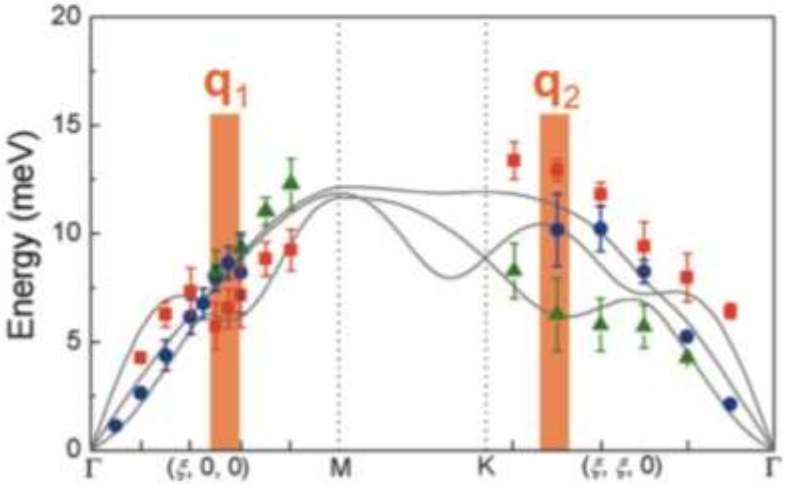
Liu and Wu, UC Berkeley

Phonon dispersion of TaS_2

- Densed individual TA and LA measurement
- Bunched TA, LA dispersions, reducing aaa
- Large a-o gap, reducing aao scattering
- Limitted phonon-phonon scattering
- Kohn anomaly at q1 and q2

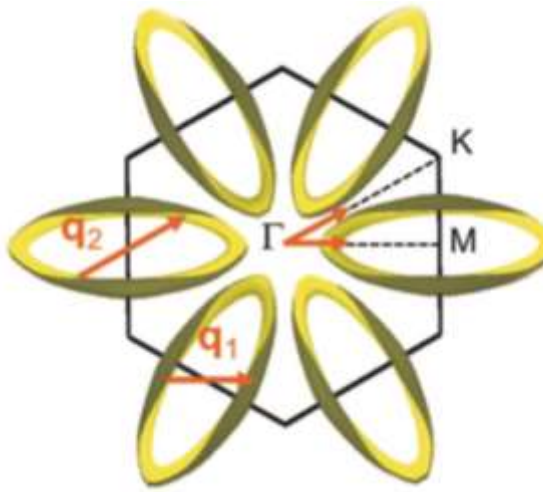


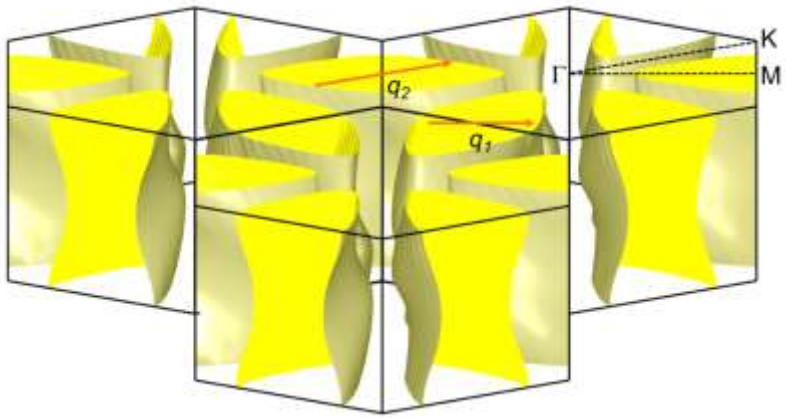


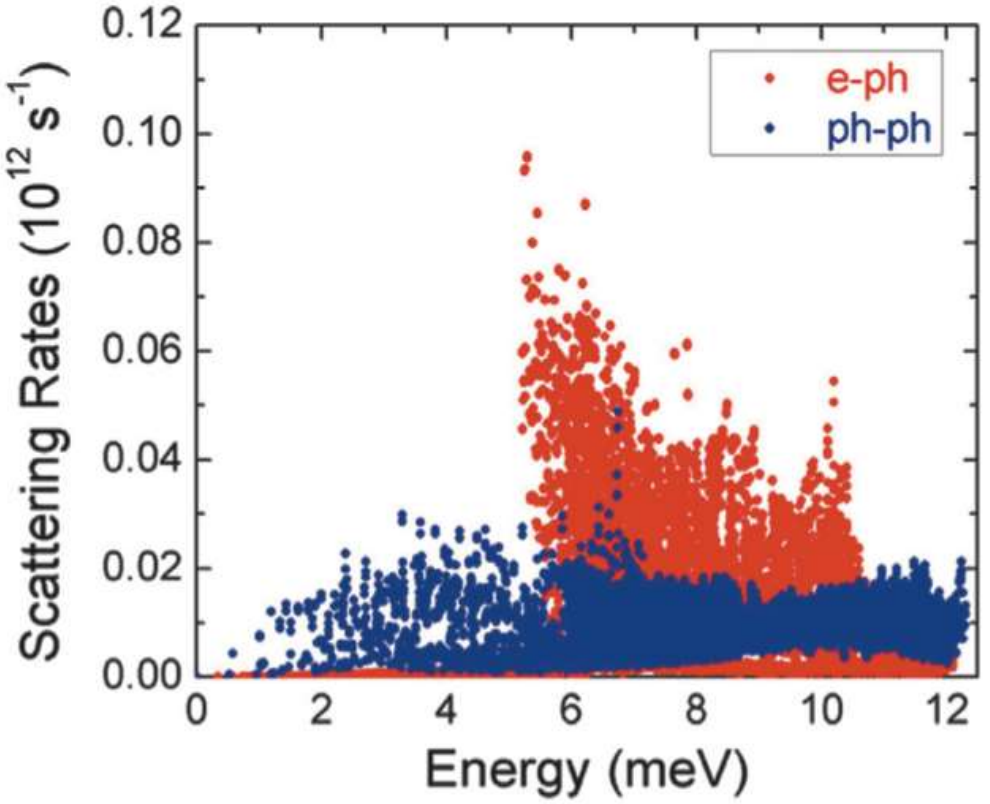


Strong e-p scattering

- Fermi surface nesting at q1 and q2, inducing Kohn anomaly
- Strong e-p scattering + limmited p-p scattering
- T independence of thermal conductivity



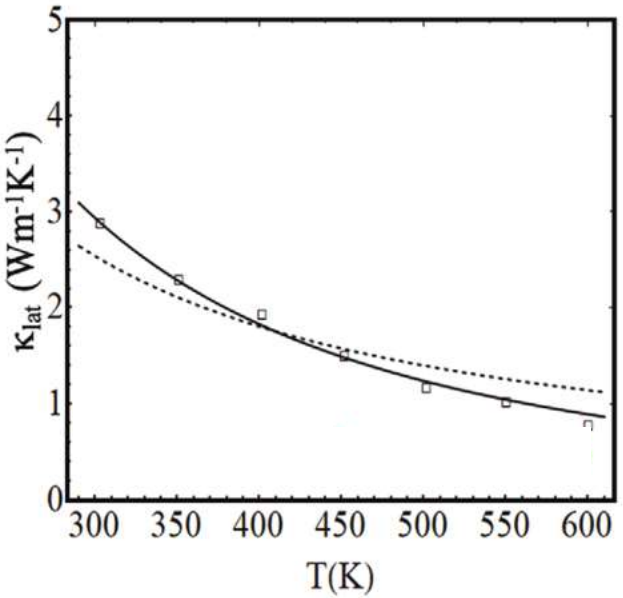


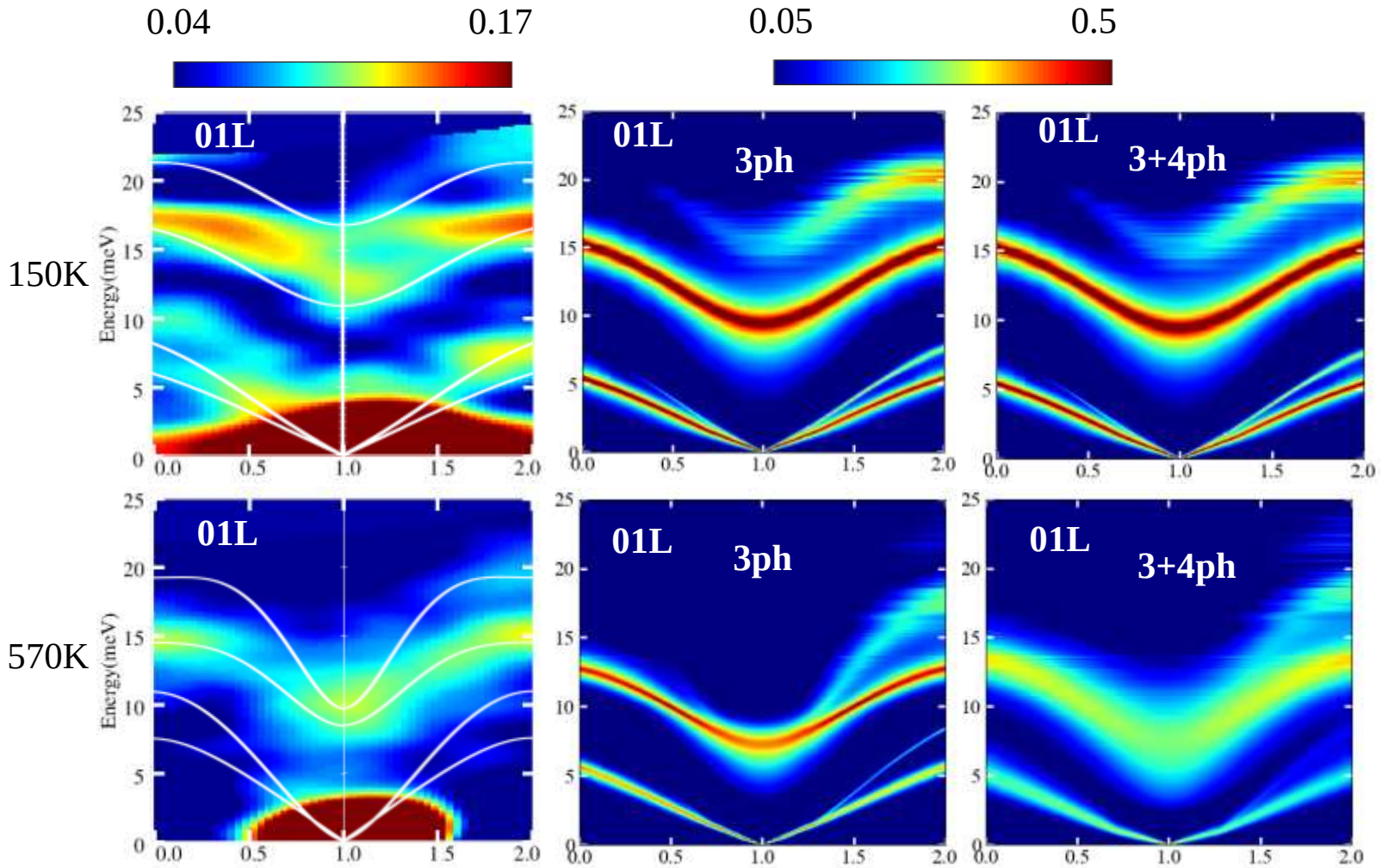


H Liu# , C Yang#, Bin Wei, Lei Jin, Ahmet Alatas, Ayman Said, Sefaattin Tongay, Fan Yang, Ali Javey, J Hong*, and Junqiao Wu*, Adv. Sci., 7, 1902071 (2020)

Four phonon scatterings inducing anomalous κ

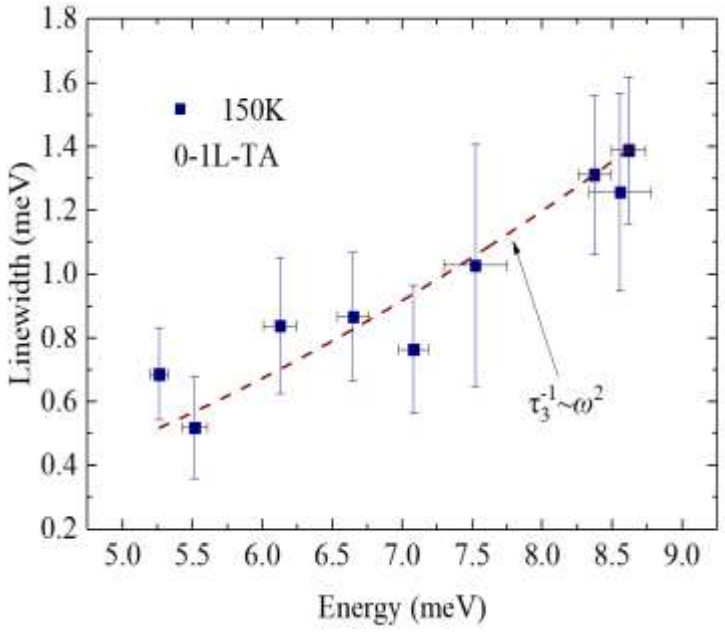
- **Thermal conductivity away from 1/T**
- **Four phonon scatterings is weak at low T but strong at high T**

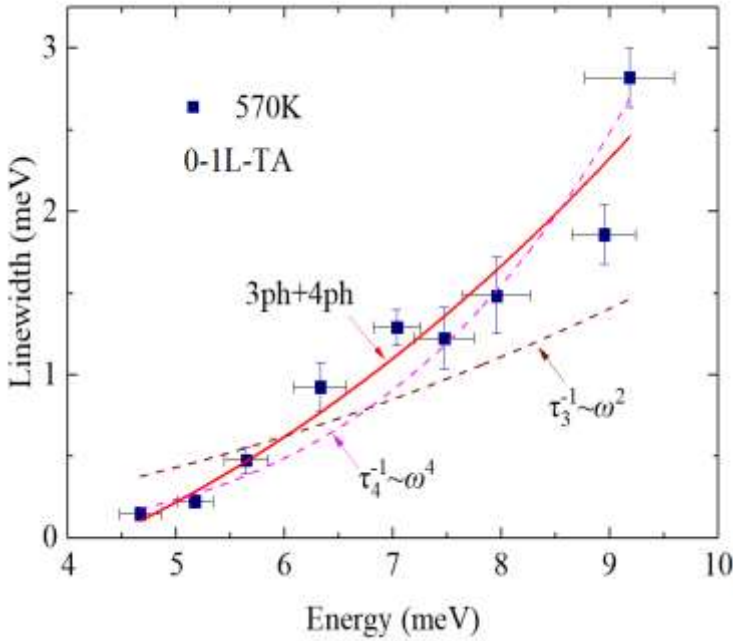


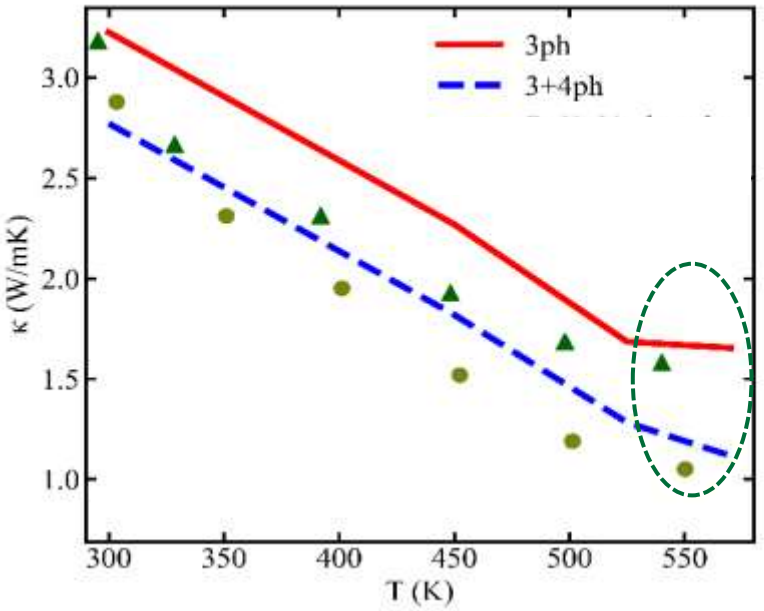


Four phonon scatterings inducing anomalous κ

- Four phonon scatterings broaden the phonon linewidth at high T
- Four phonon scatterings induce anomalous κ

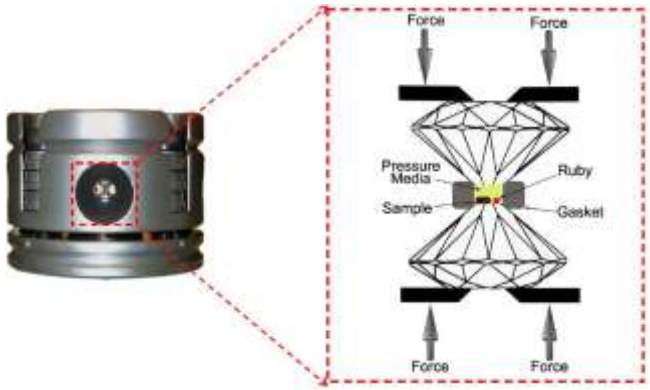




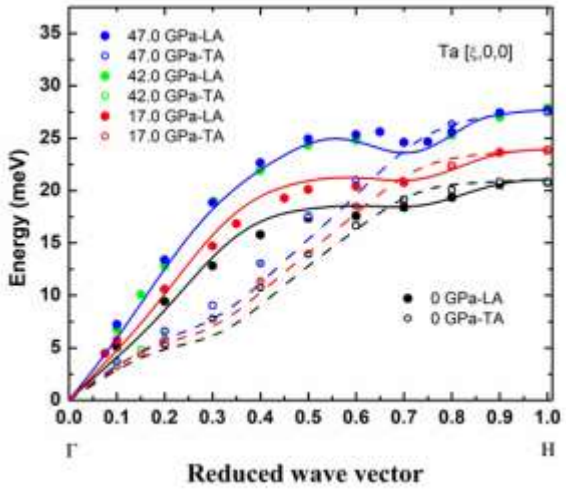


Yongheng Li, Wei, Hong et al (unpublished)

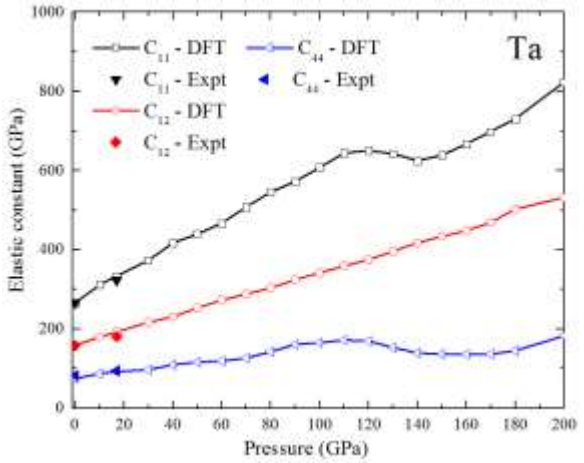
Elasticity of Ta and EET under pressure



Kohn anomaly

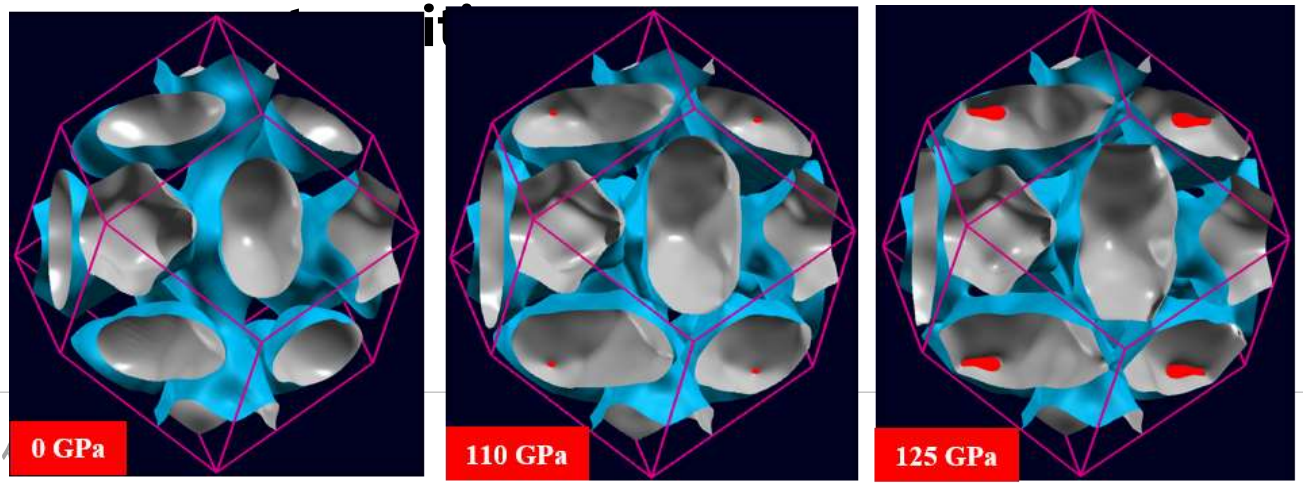


stiffness soften



HERIX @ APS

Electronic topological



J. Hong*, J. Lin* et al, PRB (2019,2020)

Conclusions

- Developed a code to simulate dynamic structure factor S(Q,E)
- Bunched phonon branches and large a-o gap suppresses p-p scattering in TaS_2, strong e-p scattering induces T independence of κ
- Matryoshka twinning is found in GaN, inducing strong in-plane p-p scattering and anisotropic κ
- Experimentally confirm the four-phonon scattering

Acknowledgements

Qi Ren, Yongheng Li, Changpeng Lin, Bin Wei (BIT)

Junqiao Wu (UC Berkeley)

Chen Li (UC Riverside)

Jung-Fu Lin (UT Austin)

Olivier Delaire (Duke Univ.)

Ayman Said, Ahmet Alatas (ANL, APS)

Baron Alfred, Yuiga Nakamura, Hiroshi Fukui, Cong Lu, (Spring-8)

John Budai, Raphael Hermann, Michael Manley (ORNL)

Dehong Yu, Guochu Deng (ANSTO)

Kazuya KAMAZAWA (J-PARC)

Thanks for your attention!