

# Understanding Spin–Phonon Interactions in Antiferromagnets

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# Acknowledgements



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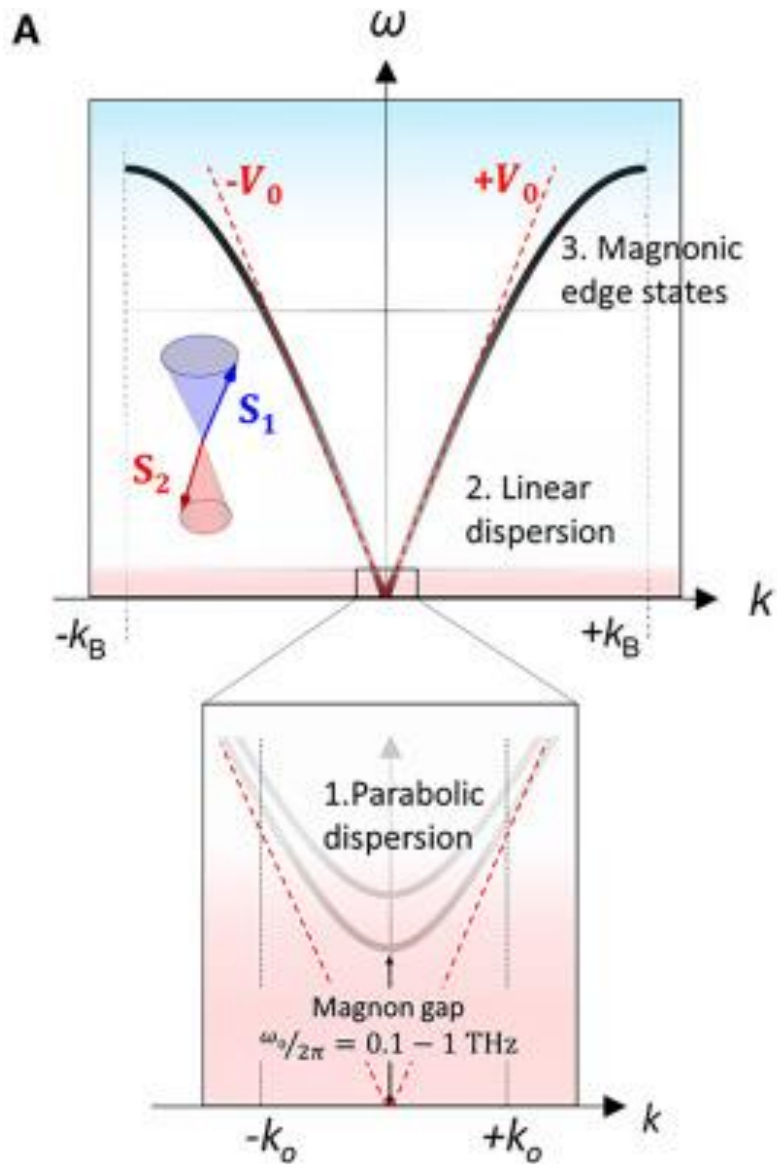
Funded by European Union



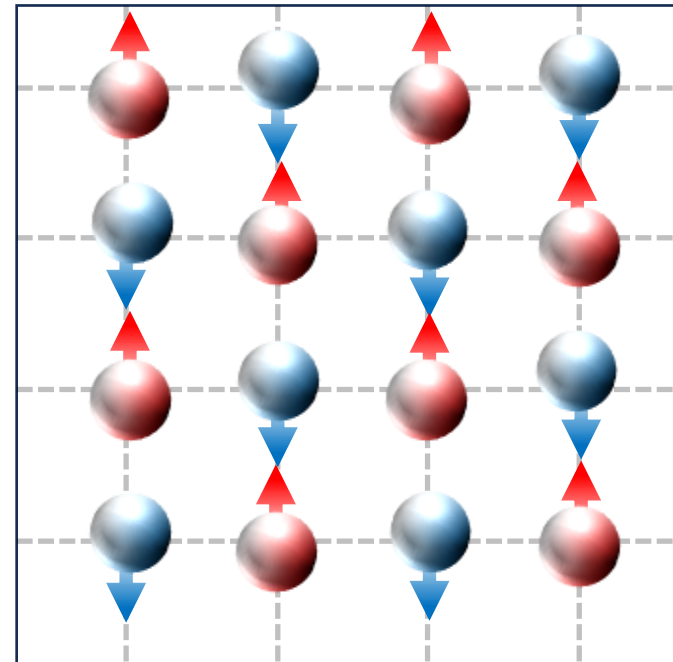
Radboud Universiteit



# Ferromagnets vs Antiferromagnet

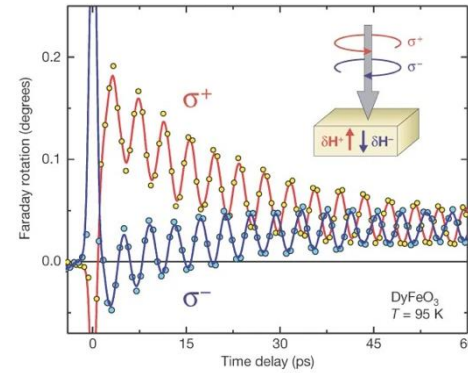
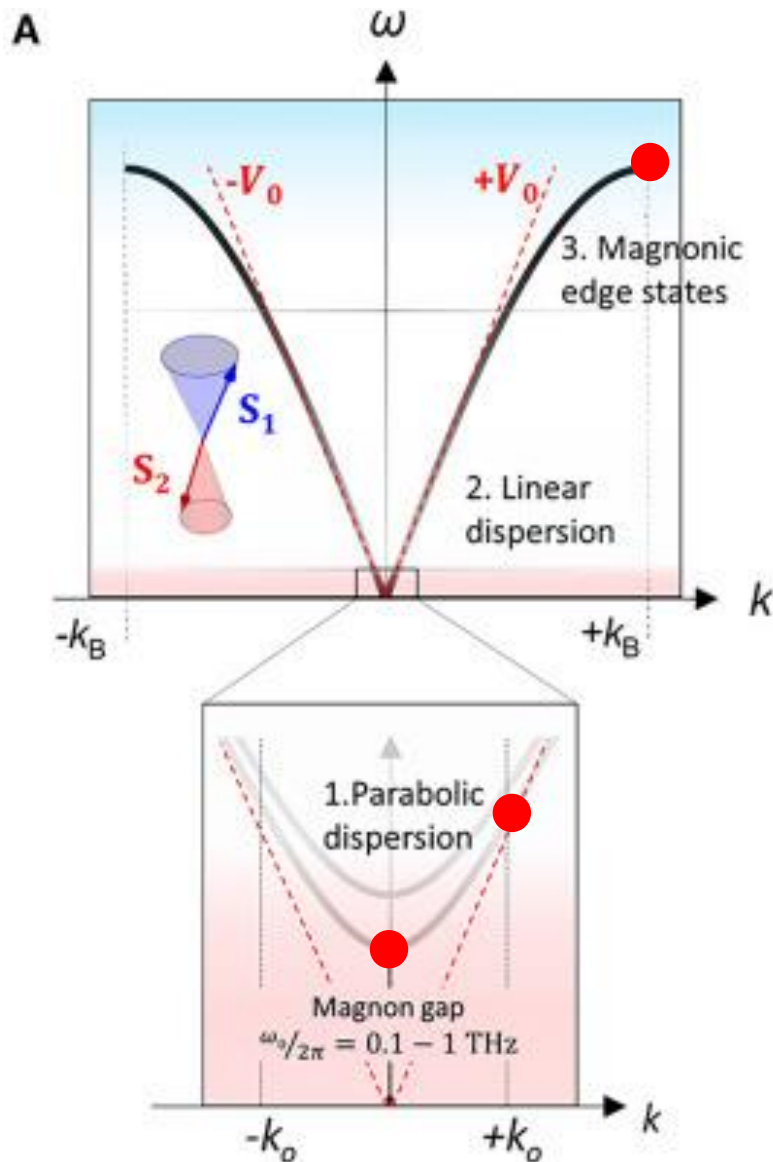


Antiferromagnet  
 $M = M_1 + M_2 = 0$ ;  $L = M_1 - M_2 \neq 0$   
 Two sublattice

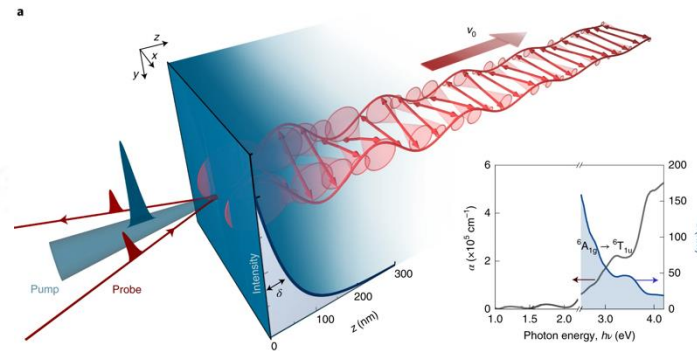


$$\omega_{\text{AFM}} \propto \sqrt{\omega_{\text{anisotropy}} \omega_{\text{Exchange}}}$$

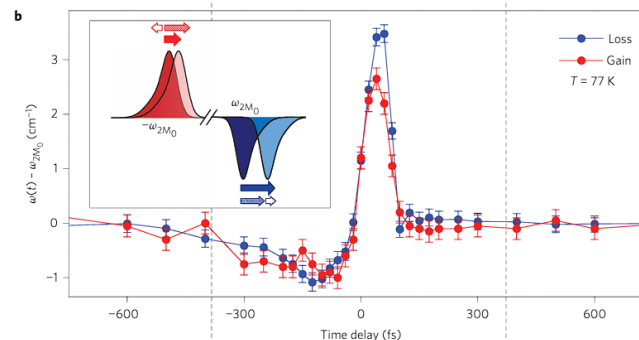
# Antiferromagnetic Terahertz Spin waves



Nature 435,  
655 (2005)

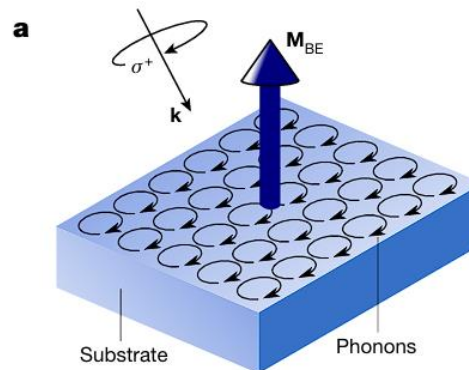
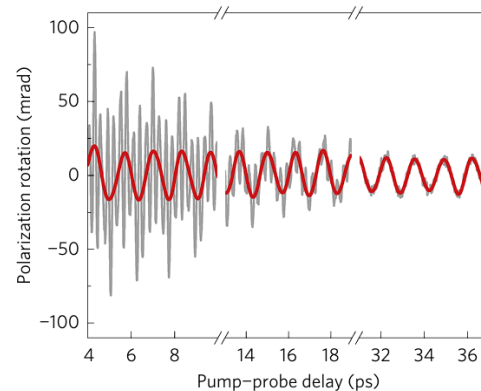
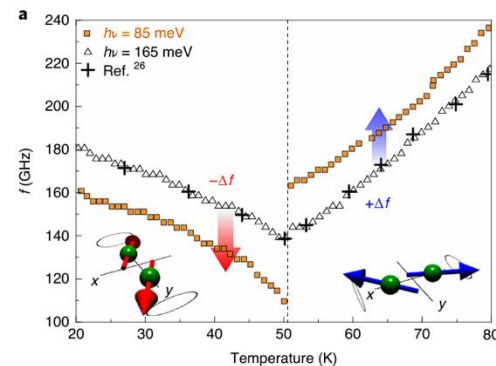
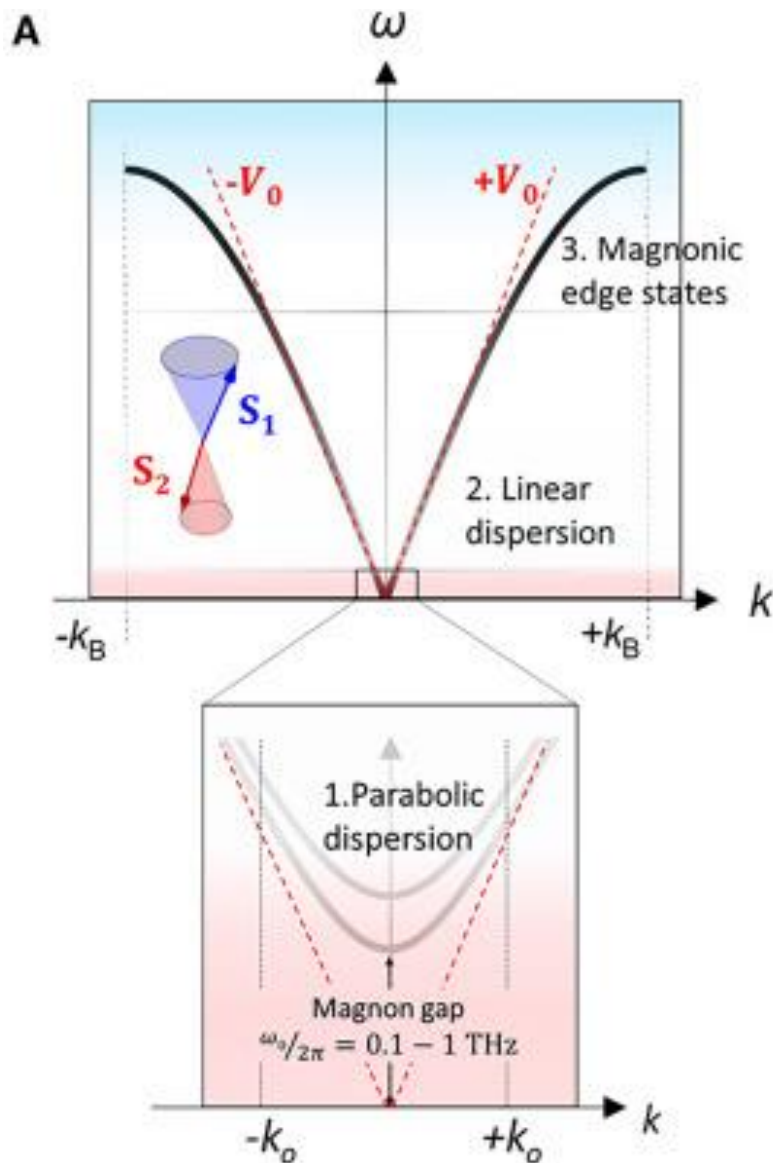


Nat. Phys. 17,  
1001, (2021)



Nature  
Photonics 9,  
506, (2015)

# Antiferromagnetic Terahertz Spin waves



Phonon driven  
Control of  
exchange interactions  
Nat. Mat. 20, 607,  
2021

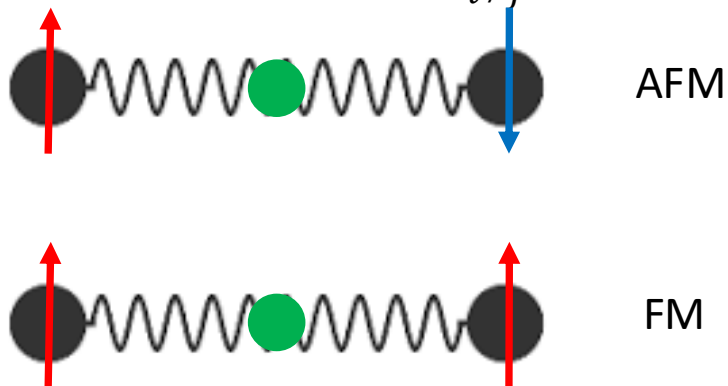
Phonon-magnetic  
field-induced  
excitation of Spin  
waves  
Nat. Phys. 13, 132,  
(2017)

A chiral phonon  
causes a magnetic  
field, which  
induces switching  
Nature 628, 540–  
544 (2024)

# Role of Lattice to Magnetic Interactions

$H_{\text{magnetic Interactions}} = \text{Exchange Interactions} + \text{DM Interaction} + \text{Magnetic Anisotropy}$

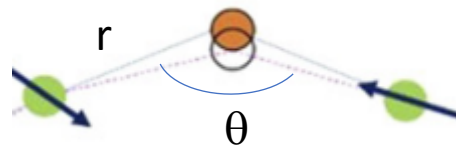
$$H_{\text{exchnage}} = - \sum_{i \neq j} J_{ij} S_i S_j$$



$$H_{DM} = - \sum_{i \neq j} D_{ij} (S_i \times S_j)$$



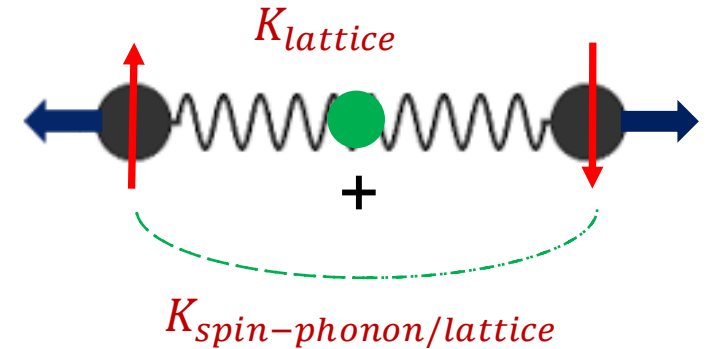
$$J_{ij} \rightarrow J_{ij}(r, \theta)$$



$$J_{ij} \rightarrow J_{ij} + \frac{\partial J_{ij}}{\partial Q_i}$$

$$D_{ij} \rightarrow D_{ij} + \frac{\partial D_{ij}}{\partial Q_i}$$

# Spin-phonon/lattice coupling



Harmonic Oscillator

$$\omega = \sqrt{\frac{K}{m}}$$

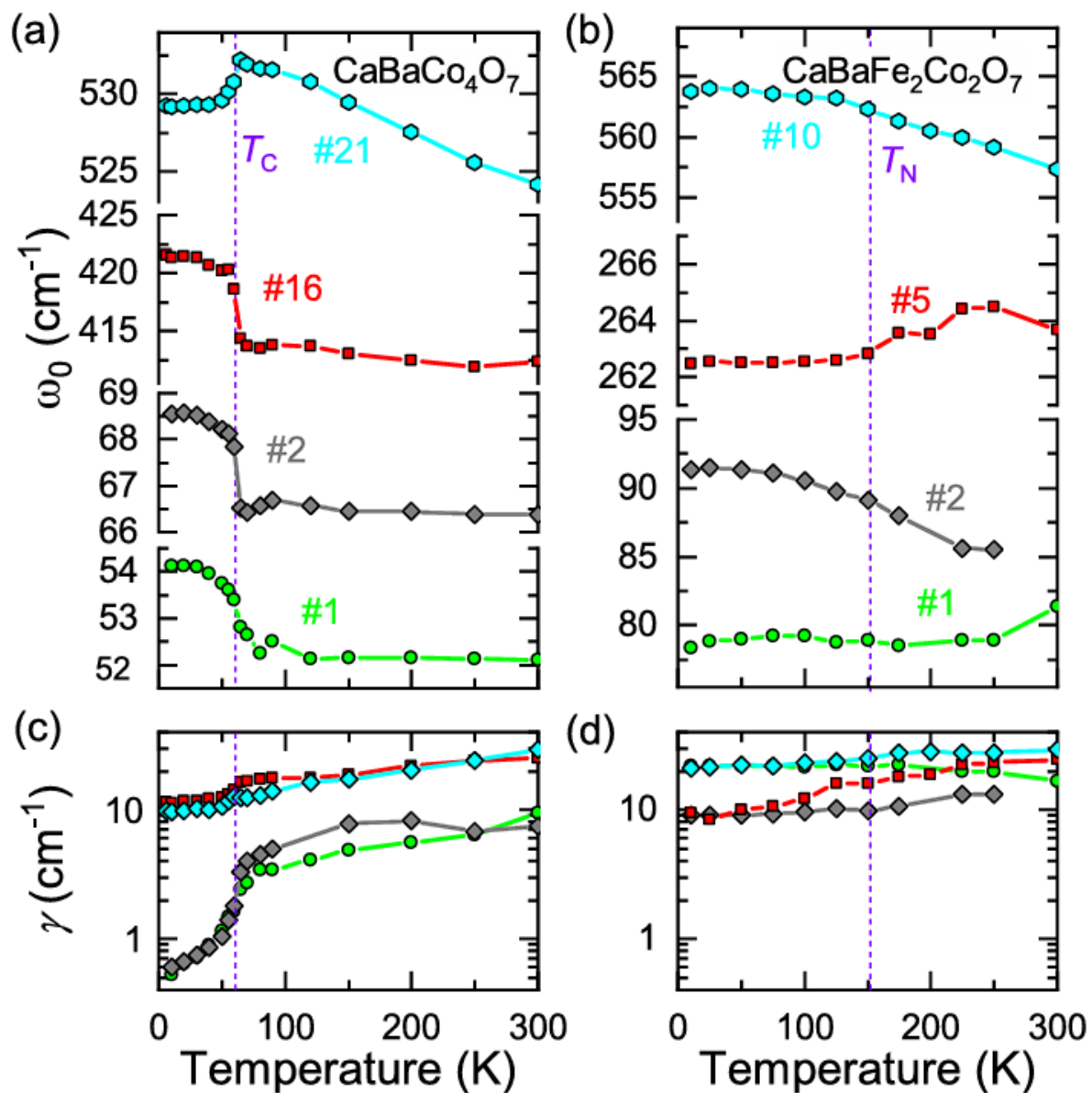
Coupled Oscillator

$$\omega = \sqrt{\frac{K_{eff}}{\mu}}$$

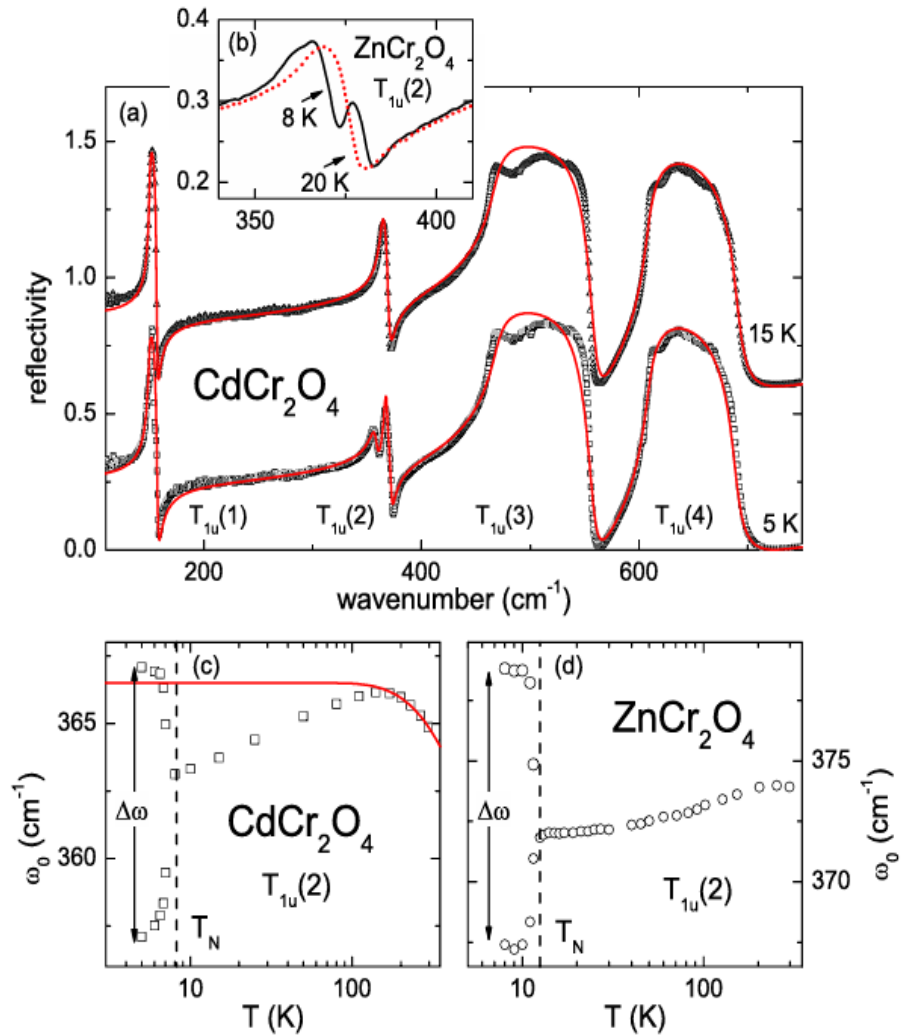
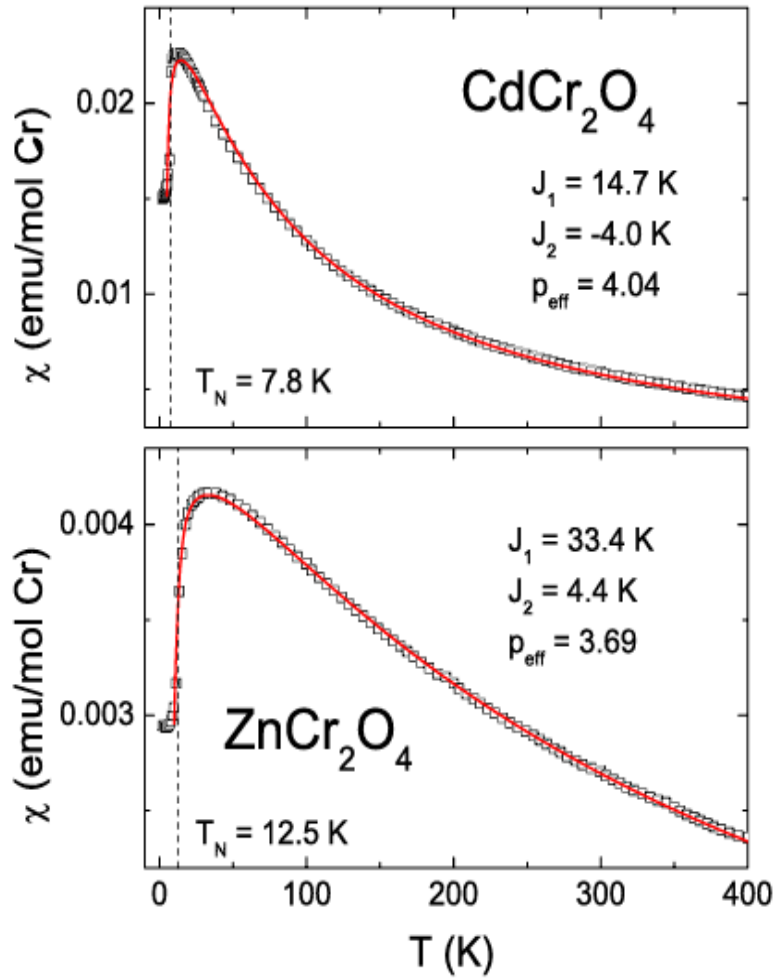
$$K_{eff} = K_{lattice} + K_{spin-phonon/lattice} + K_{electron-phonon} + K_{anharmonic}$$

$$\Delta\omega_{sp} = \frac{2}{\omega\mu} \frac{\delta^2 J}{\delta u^2} \langle S_i \cdot S_j \rangle = \lambda_{sp} \langle S_i \cdot S_j \rangle$$

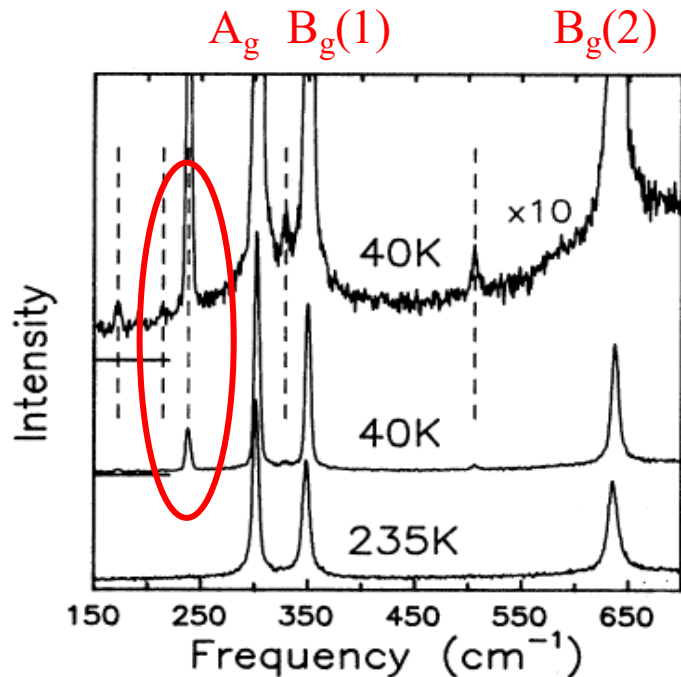
# Signatures of the Spin-phonon coupling



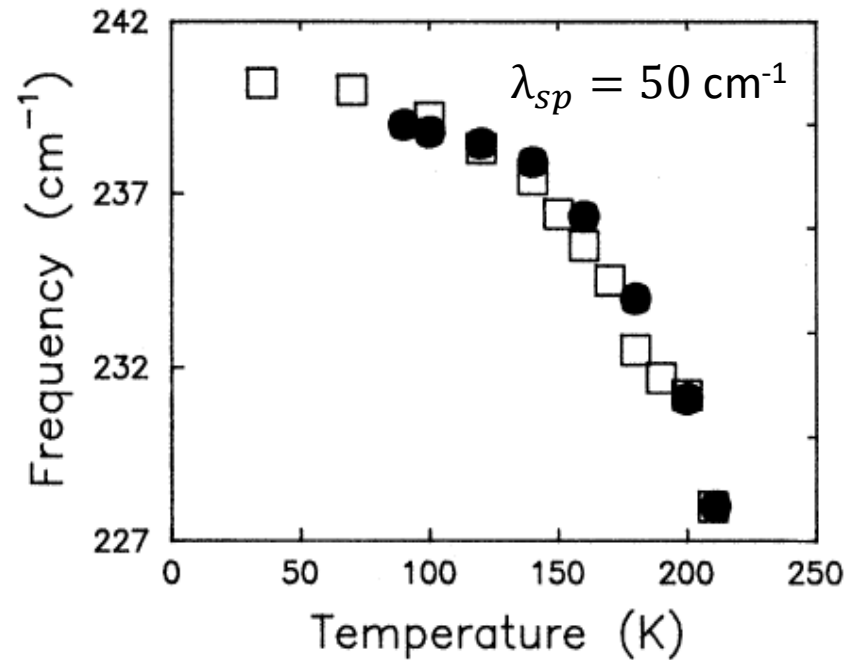
# Signatures of the Spin-phonon coupling



# Spin-Phonon coupling in CuO



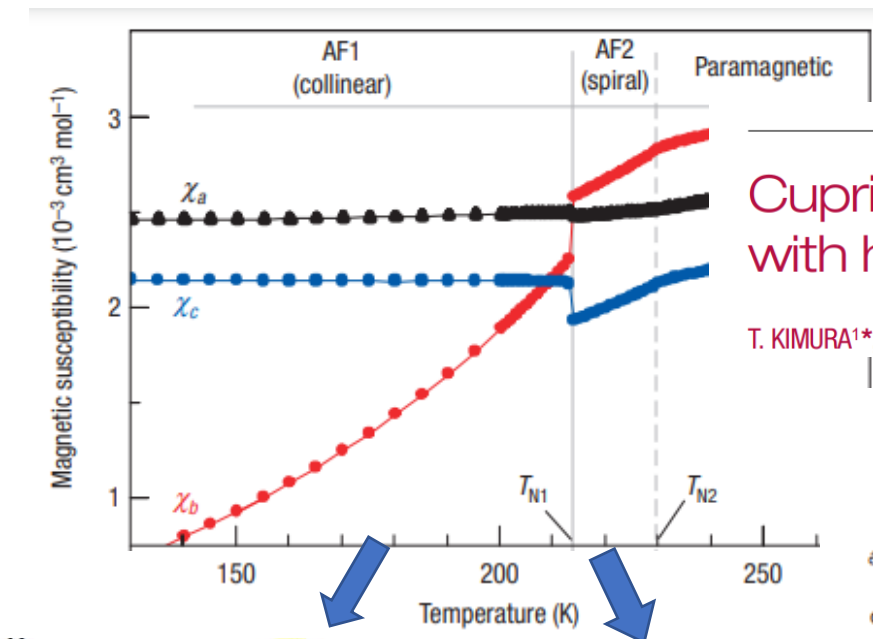
Zone boundary modes are appearing in a commensurate magnetic state because of the zone folding effect.



Spin-Phonon coupling constant is estimated to be  $40 \text{ cm}^{-1}$ .

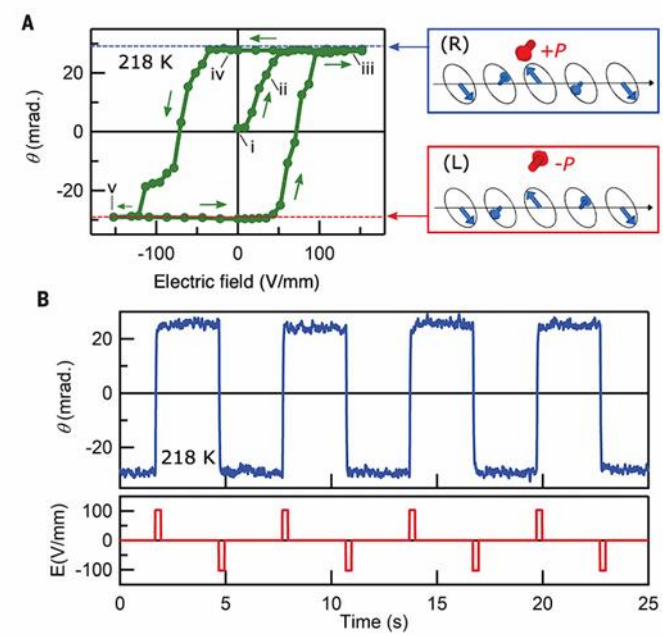
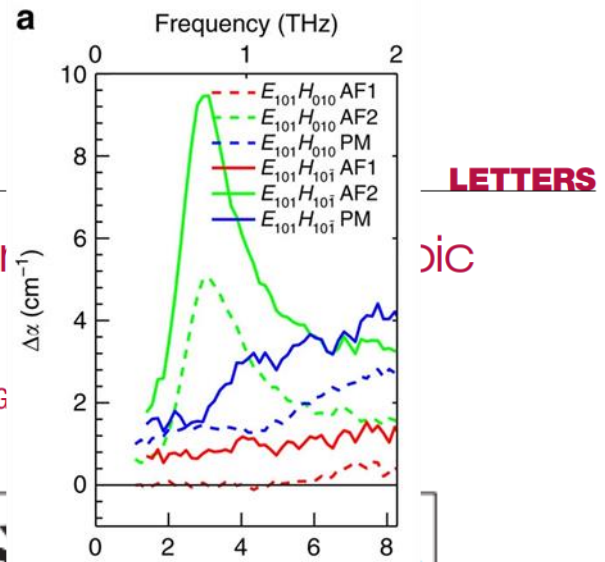
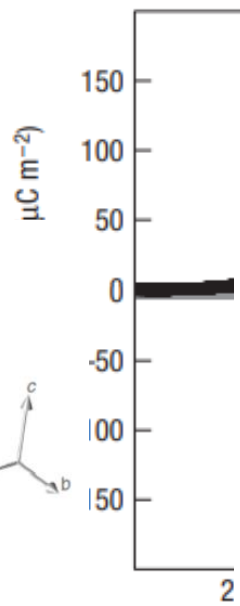
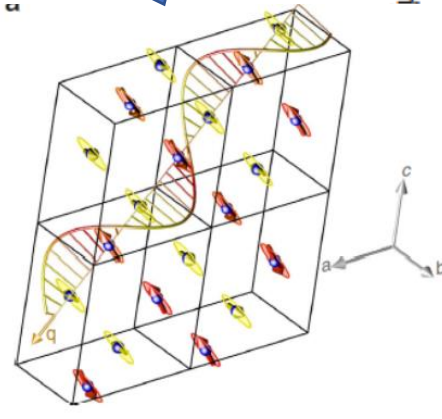
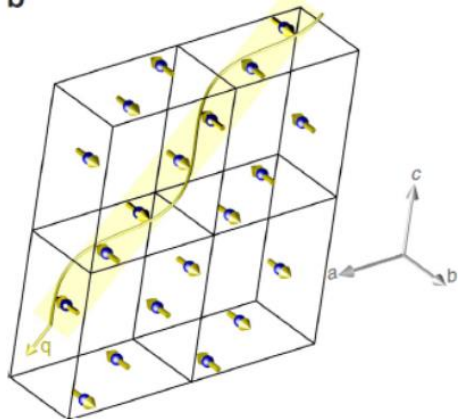
# Spin-induced polarization of CuO

# Spin-Induced High Temperature Multiferroicity of CuO



Cupric oxide as an *im* with high- $T_C$

T. KIMURA<sup>1\*</sup>, Y. SEKIO<sup>1</sup>, H. NAKAMURA<sup>1</sup>, T. SIEG



# Spin-Induced Room Temperature Multiferroicity of CuO

ARTICLE

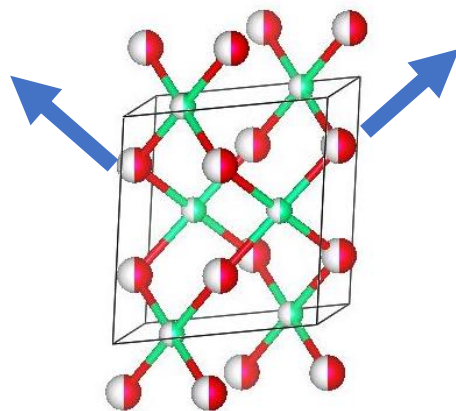
Received 8 May 2013 | Accepted 27 Aug 2013 | Published 23 Sep 2013

DOI: 10.1038/ncomms3511

## Room-temperature spin-spiral multiferroicity in high-pressure cupric oxide

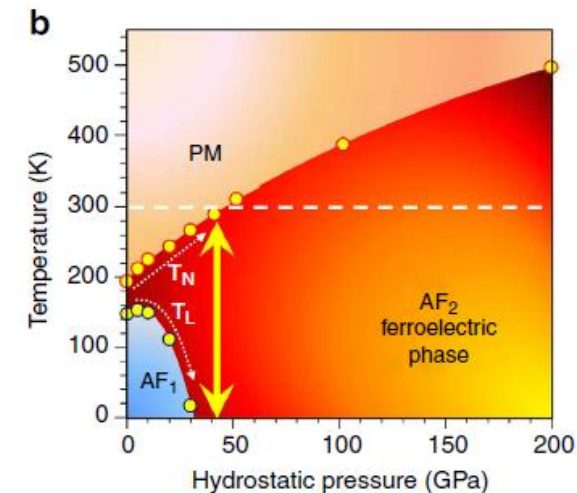
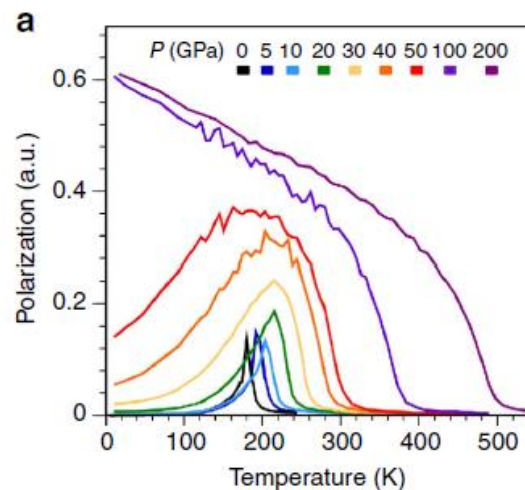
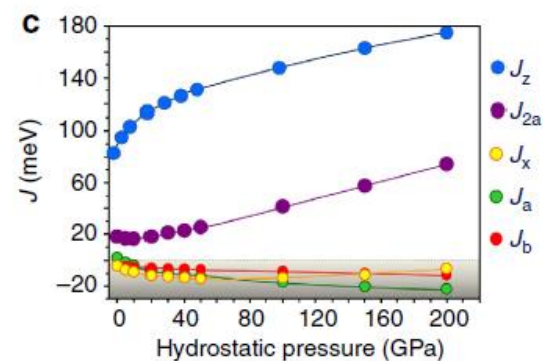
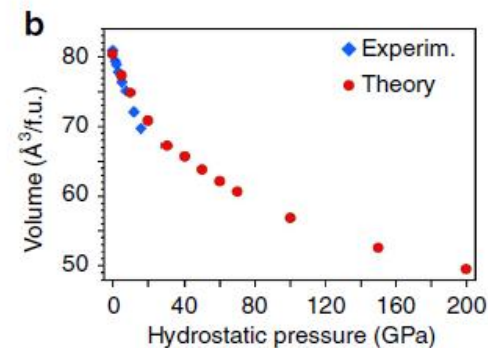
Xavier Rocquefelte<sup>1</sup>, Karlheinz Schwarz<sup>2</sup>, Peter Blaha<sup>2</sup>, Sanjeev Kumar<sup>3</sup> & Jeroen van den Brink<sup>4,5</sup>

(101)  
107°  
FM  
Interaction

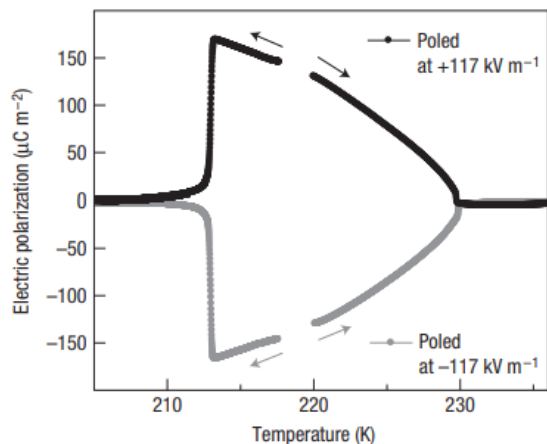


(10-1)  
146°  
AFM  
Interaction

Change in Cu-O-Cu bonds along (10-1) is responsible for enhancing the transition temperature ( $J_z=80$  meV)



# Spin induced Polarization of CuO



How can the  $T_C$  be *tuned* to room temperature?



What is the origin?  
Needed for functional engineering

**Theoretical prediction**



Which one



DM interaction  
*Lattice displa*

Spin current

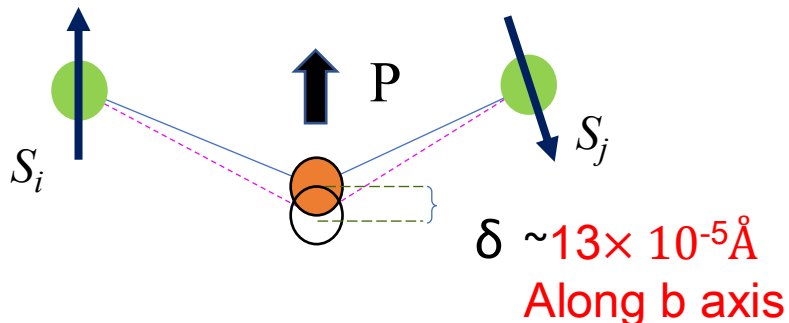
**Femtosecond Magnetically Induced Lattice Distortions in Multiferroic TbMnO<sub>3</sub>**

H. C. WALKER, F. FABRIZI, L. PAOLASINI, F. DE BERGEMIN, J. HERRERO-MARTIN, A. T. BOOTHROYD, D. PRABHAKARAN, AND D. F. MCMORROW [Authors Info & Affiliations](#)

SCIENCE · 2 Sep 2011 · Vol 333, Issue 6047 · pp. 1273-1276 · DOI: 10.1126/science.1208085

XRD, EXAFS sensitivity  $\sim 10^{-3}$  Å

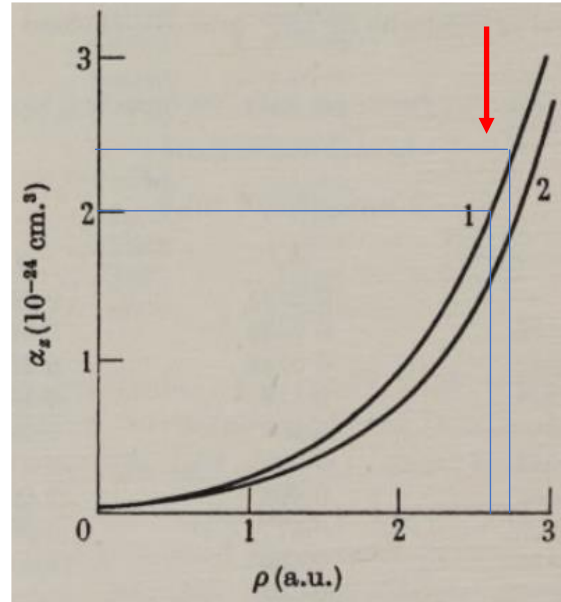
Require a new approach



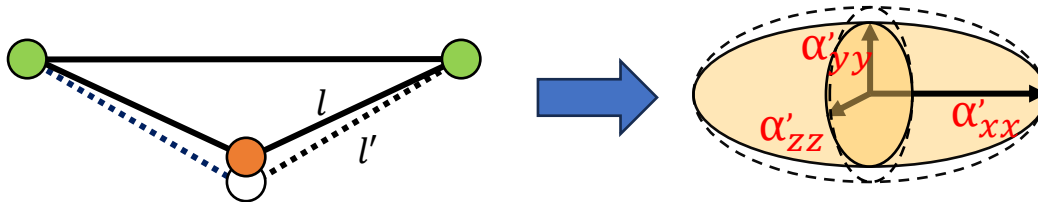
# How to identify the signature of atomic displacements?



Polarizability is more highly sensitive to interatomic distances



1 a.u. = 0.529 Angstrom



Raman polarizability

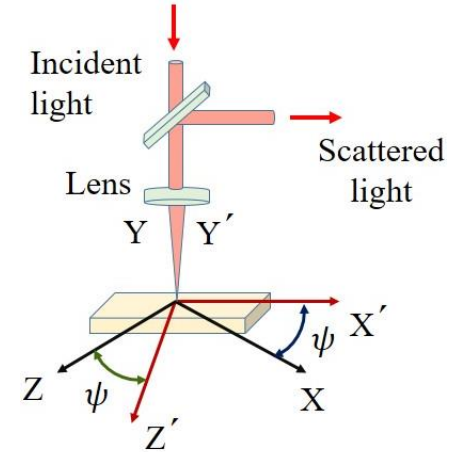
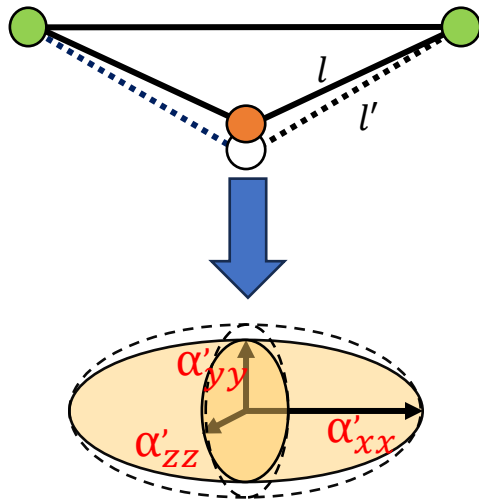
$$R = \begin{pmatrix} \alpha_{xx} & \alpha_{xy} & \alpha_{xz} \\ \alpha_{yx} & \alpha_{yy} & \alpha_{yz} \\ \alpha_{zx} & \alpha_{zy} & \alpha_{zz} \end{pmatrix}$$

**Phonon Renormalization**

$$\Delta\omega = K_{ii}\delta x_{ii} = K_{11}\delta x_{11} + K_{22}\delta x_{22} + K_{33}\delta x_{33}$$

Where and How..?

# Angle-resolved polarized Raman spectroscopy



$$\text{Raman intensity } I \propto (\mathbf{e}_i \phi R \phi^T \mathbf{e}_s)^2$$

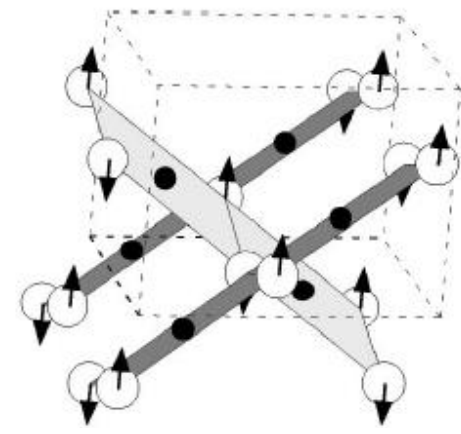
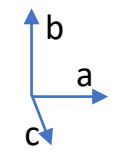
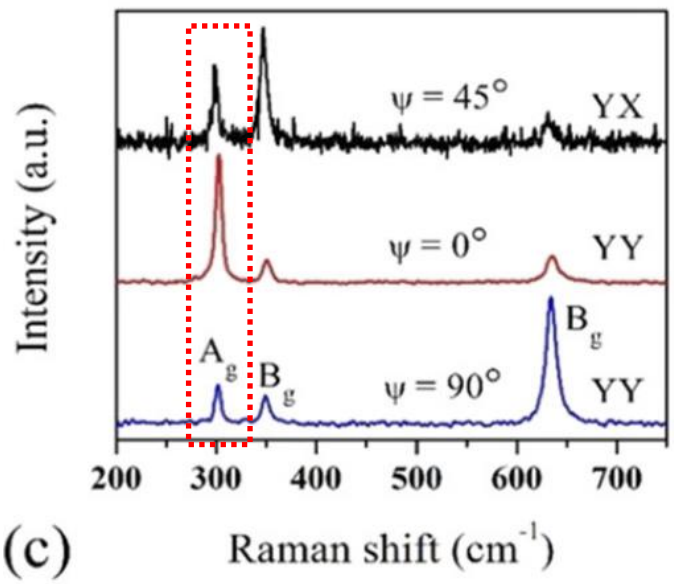
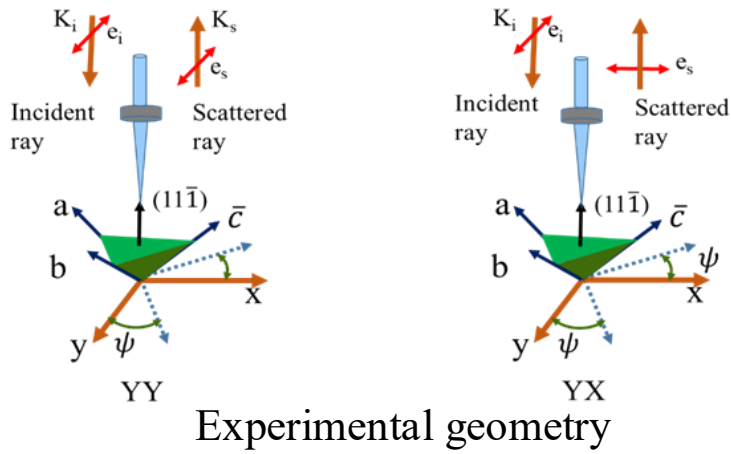
$$R = \begin{pmatrix} \alpha_{xx} & \alpha_{xy} & \alpha_{xz} \\ \alpha_{yx} & \alpha_{yy} & \alpha_{yz} \\ \alpha_{zx} & \alpha_{zy} & \alpha_{zz} \end{pmatrix}$$

Raman tensor for  $\text{BaTiO}_3$

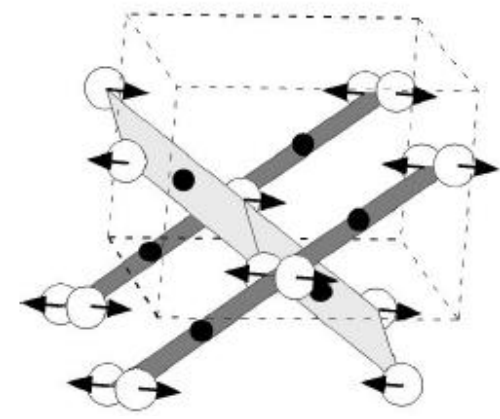
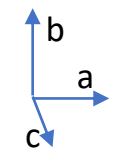
$$A_1 = \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix} \quad B_1 = \begin{pmatrix} c & 0 & 0 \\ 0 & -c & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

$$I_{A_1}^{\parallel} \propto [a \cos^2 \Psi + b \sin^2 \Psi]^2 \quad I_{B_1}^{\parallel} \propto [c \sin^2 \Psi]^2$$

# Polarized Raman spectroscopy of CuO

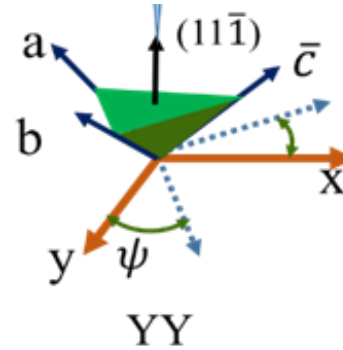
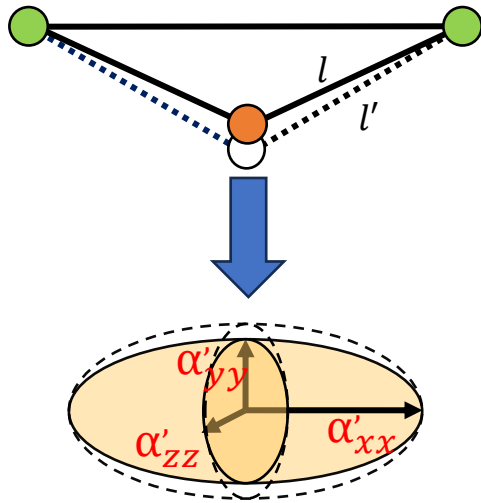


$A_g$



$B_g(1)$

# Polarized Raman spectroscopy of CuO



$$A_g = \begin{pmatrix} a & 0 & d \\ 0 & b & 0 \\ d & 0 & c \end{pmatrix} \quad B_g = \begin{pmatrix} 0 & e & 0 \\ e & 0 & f \\ 0 & f & 0 \end{pmatrix}$$

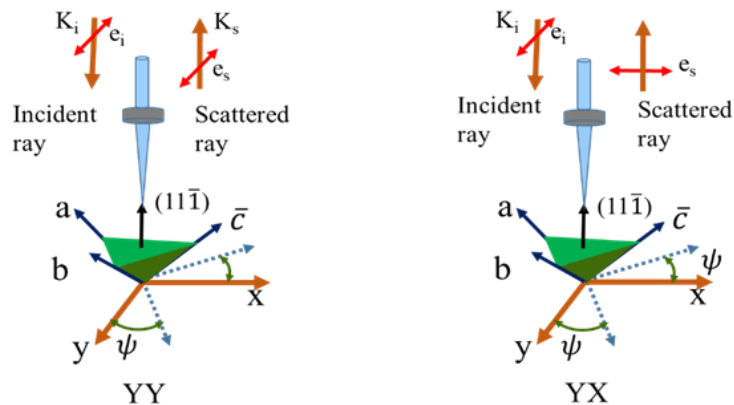
$$I_{A_g}^{\parallel} \propto ((0.64a + 0.34b)\cos^2\psi + (0.15a + 0.28b + 0.54c + 0.58d)\sin^2\psi + (0.31(b - a) - 0.58d)\sin 2\psi)^2$$

$$I_{B_g}^{\parallel} \propto ((-0.93e)\cos^2\psi + (0.41e + 0.79f)\sin^2\psi + (-0.20e + 0.43f)\sin 2\psi)^2$$

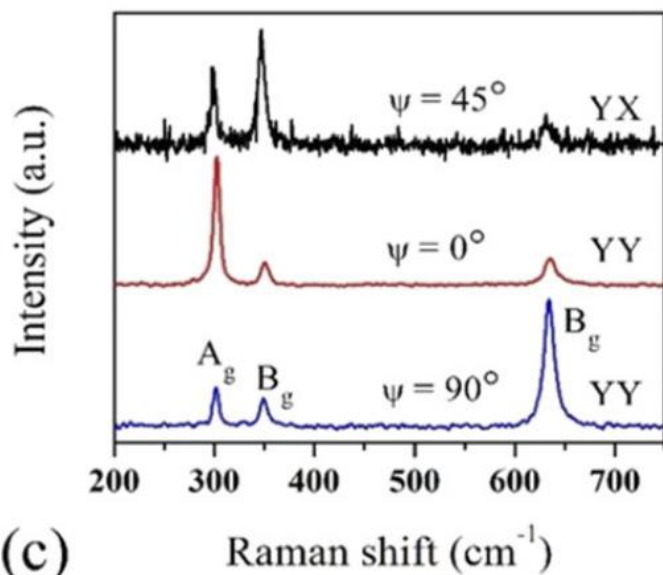
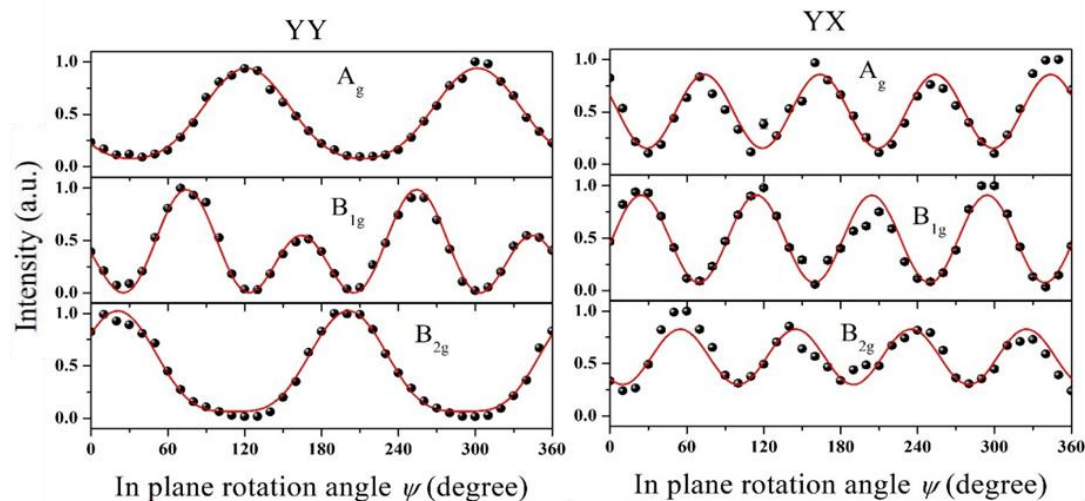
$$I_{A_g}^{\perp} \propto \left( (0.31(a - b) + 0.58d)\cos 2\psi + \frac{(0.49a + 0.05b - 0.54c - 0.58d)}{2}\sin 2\psi \right)^2$$

$$I_{B_g}^{\perp} \propto ((0.20e - 0.43f)\cos 2\psi + (-0.67e - 0.39f)\sin 2\psi)^2$$

# Polarized Raman spectroscopy of CuO



Experimental geometry



(c)

Modes	Tensor elements	mean square error
$A_g$	$a = -0.12$	$\pm 0.02$
	$b = 0.30$	$\pm 0.04$
	$c = -0.34$	$\pm 0.01$
$B_g$	$d = 0.23$	$\pm 0.02$
	$e = -0.23$	$\pm 0.06$
	$f = 0.77$	$\pm 0.10$

# Determination of ferroelectric polarization direction

Raman tensor

$$A_g = \begin{pmatrix} a & 0 & d \\ 0 & b & 0 \\ d & 0 & c \end{pmatrix}$$

Raman intensity

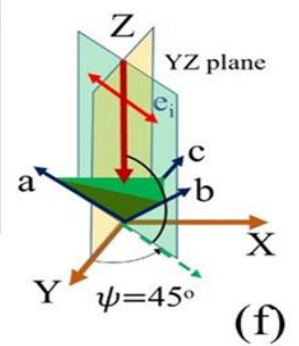
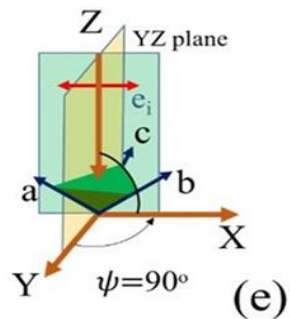
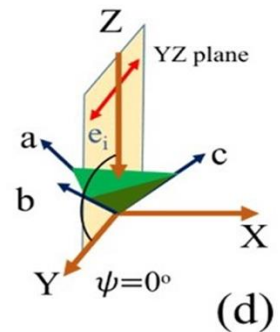
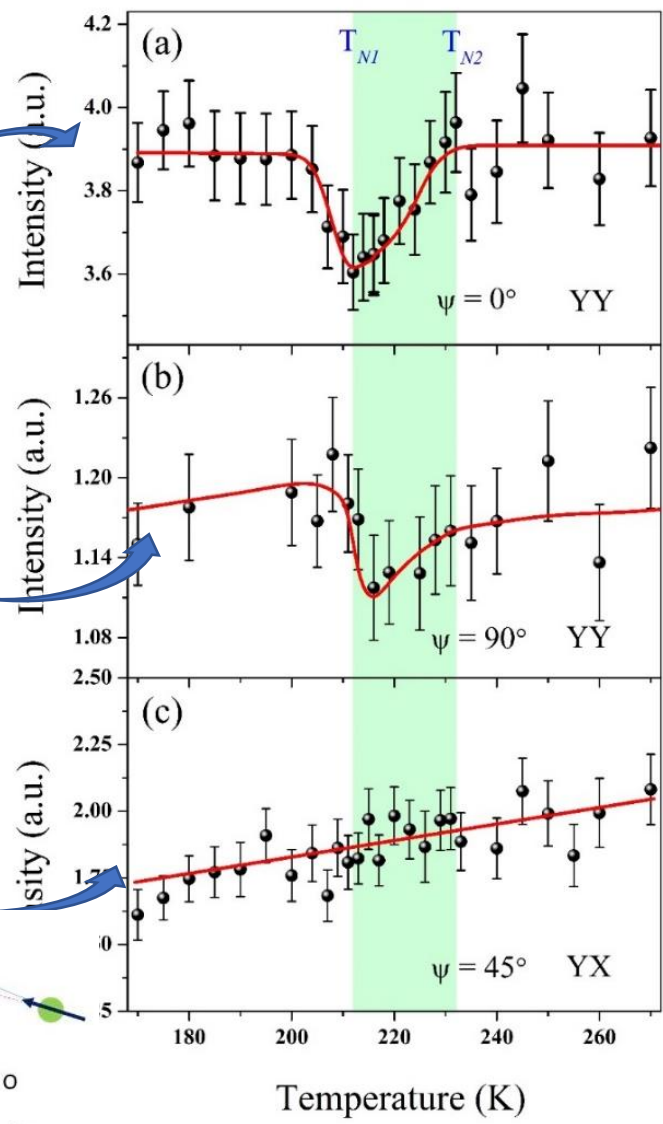
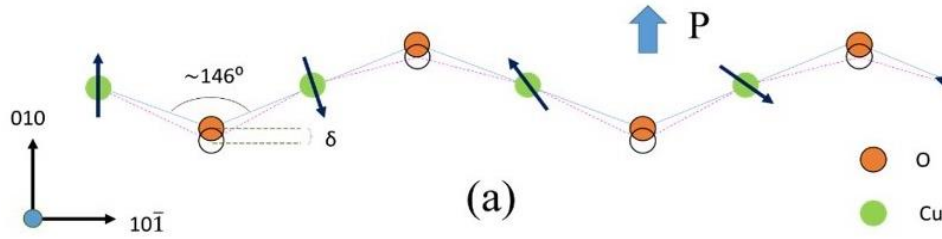
$$I_{A_g}^{\parallel} \propto ((0.64a + 0.34b)\cos^2\psi + (0.15a + 0.28b + 0.54c + 0.58d)\sin^2\psi + (0.31(b - a) - 0.58d)\sin 2\psi)^2$$

$$I_{A_g}^{\perp} \propto \left( (0.31(a - b) + 0.58d)\cos 2\psi + \frac{(0.49a + 0.05b - 0.54c - 0.58d)}{2}\sin 2\psi \right)^2$$

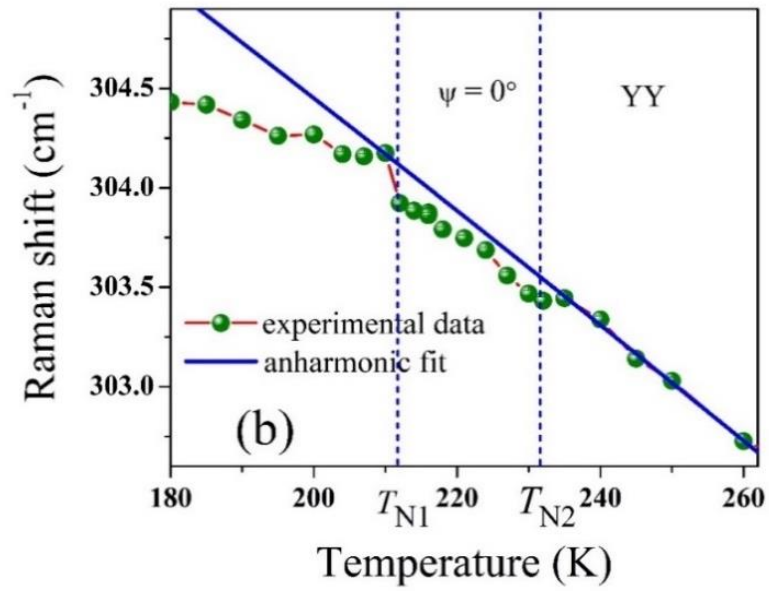
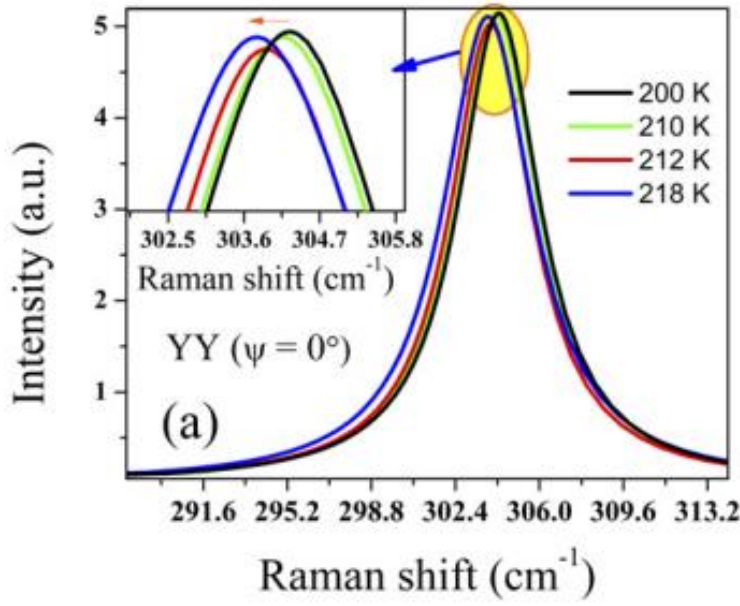
57% from b

27% from b

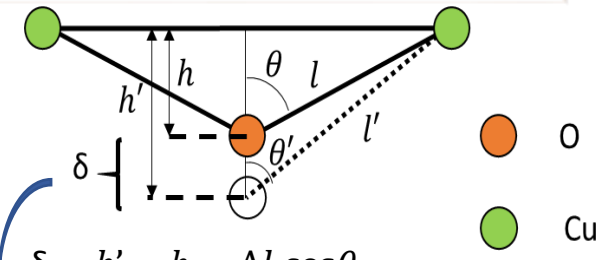
3% from b



# Phonon renormalization and finding atomic displacement

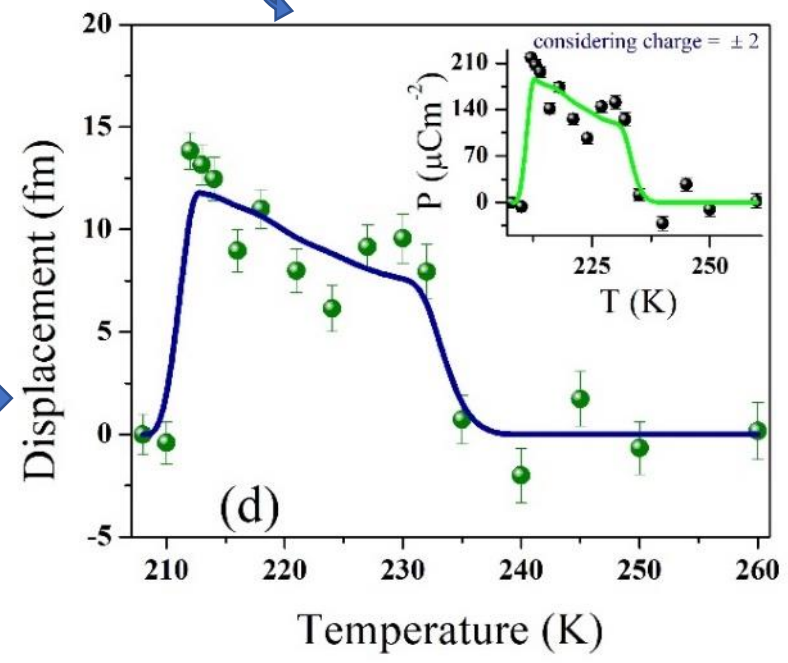


Quantification of displacement using the mode Gruneisen parameter



$$|\Delta l| = l \frac{\Delta\omega}{3Y\omega}$$

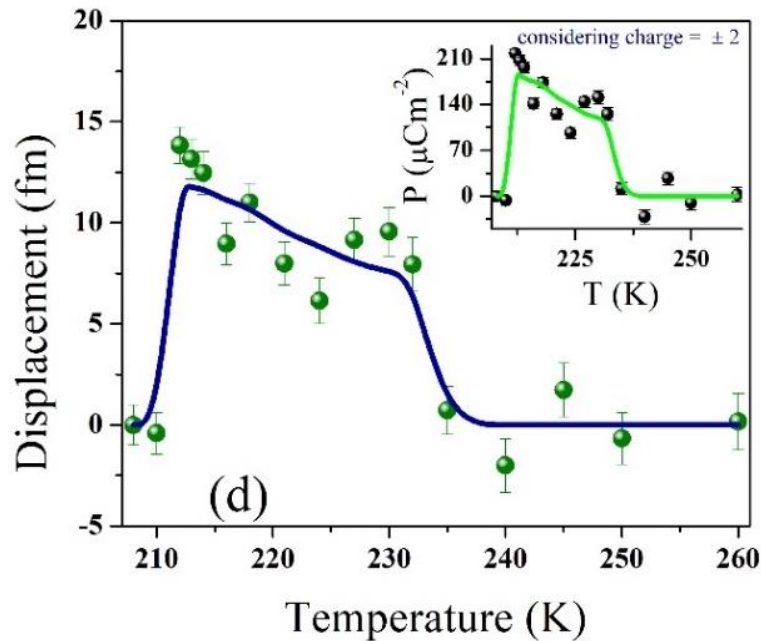
$$\delta = h' - h = \Delta l \cos\theta$$



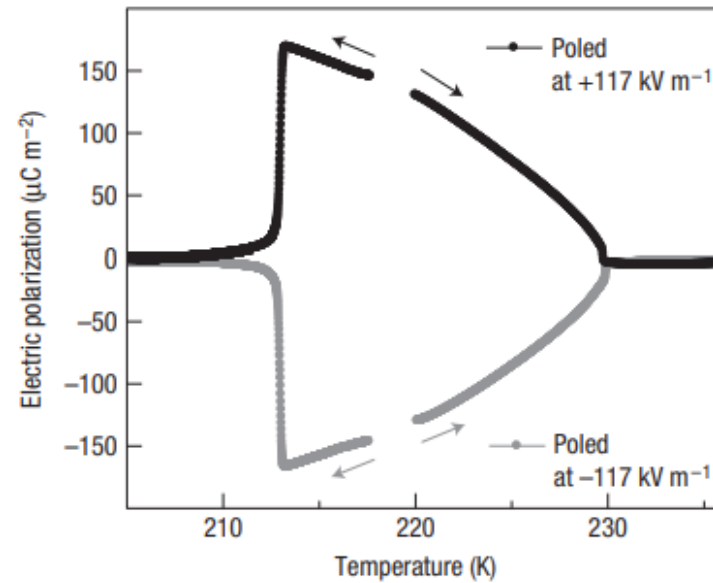
# Phonon renormalization and finding atomic displacement



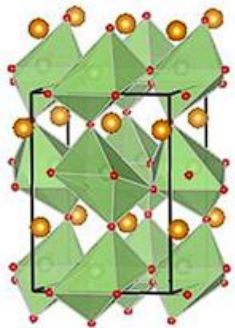
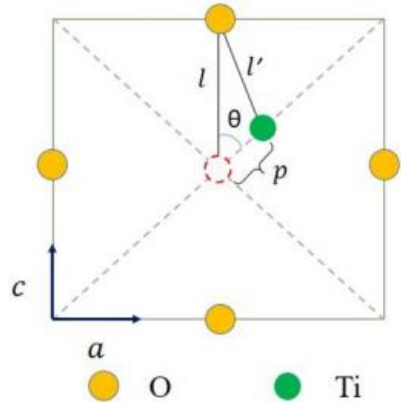
## From microscopic study (Raman)



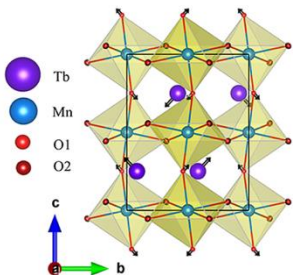
## From a macroscopic study (Pyroelectric)



# Tracking Atomic displacements



$Pbn2_1$



PAPER

## Tracing microscopic atomic displacements using polarized Raman spectroscopy: a case study on BaTiO<sub>3</sub>

Binoy Krishna De, Vivek Dwij, Hemant Singh Kunwar, Sumesh Rana and V G Sathe

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[Journal of Physics D: Applied Physics](#), Volume 55, Number 3

Citation Binoy Krishna De *et al* 2022 *J. Phys. D: Appl. Phys.* 55 035304

DOI 10.1088/1361-6463/ac2c39



PHYSICAL REVIEW B **103**, 054106 (2021)

## Breaking of inversion symmetry in NdGaO<sub>3</sub>

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ACCEPTED PAPER

## Direct observation of femtometer-scale atomic displacement driving multiferroicity in TbMnO<sub>3</sub>: A polarized Raman study

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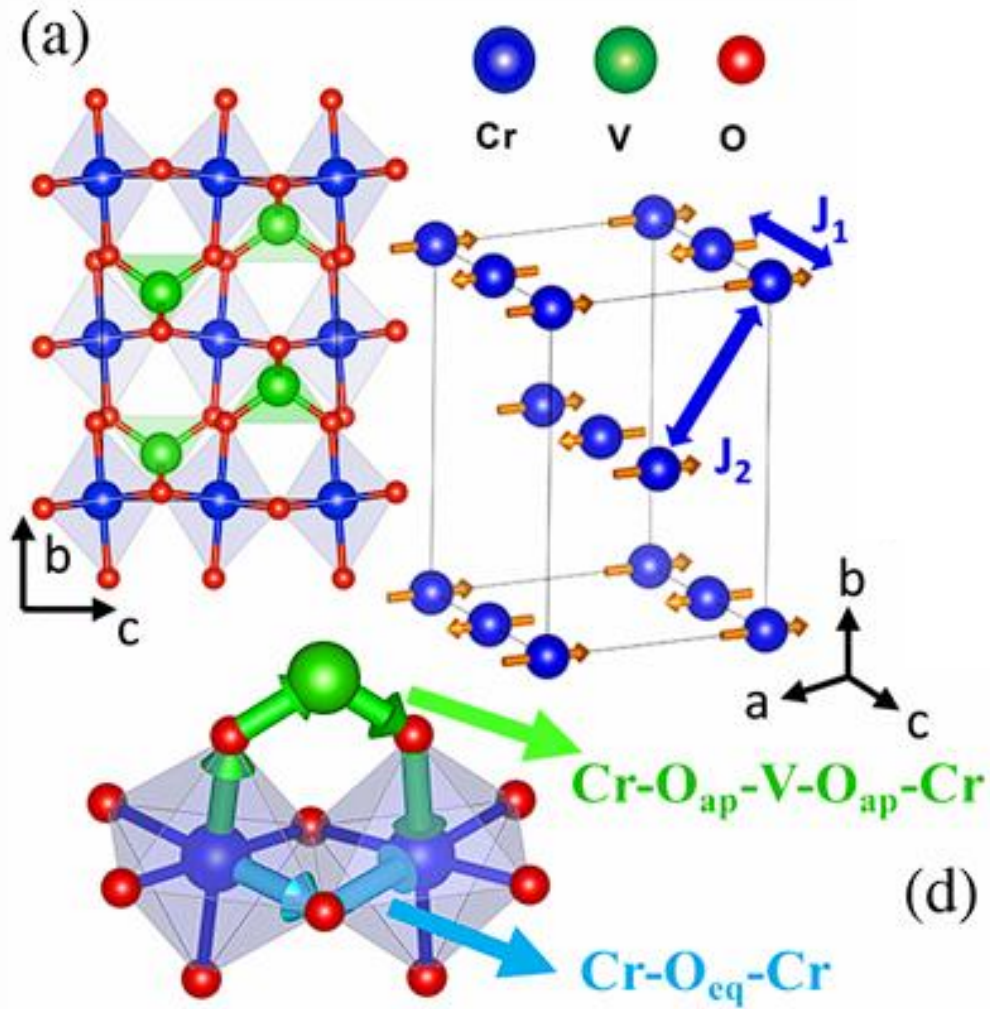
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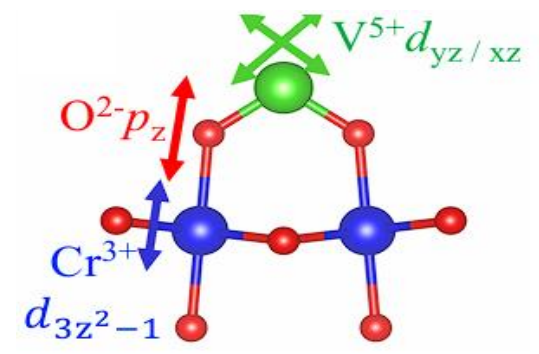
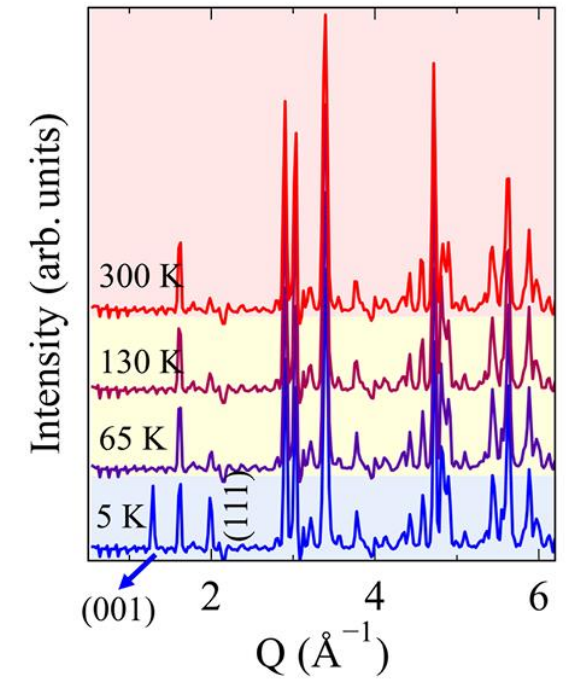


# Spin-Orbital-Phonon Coupling

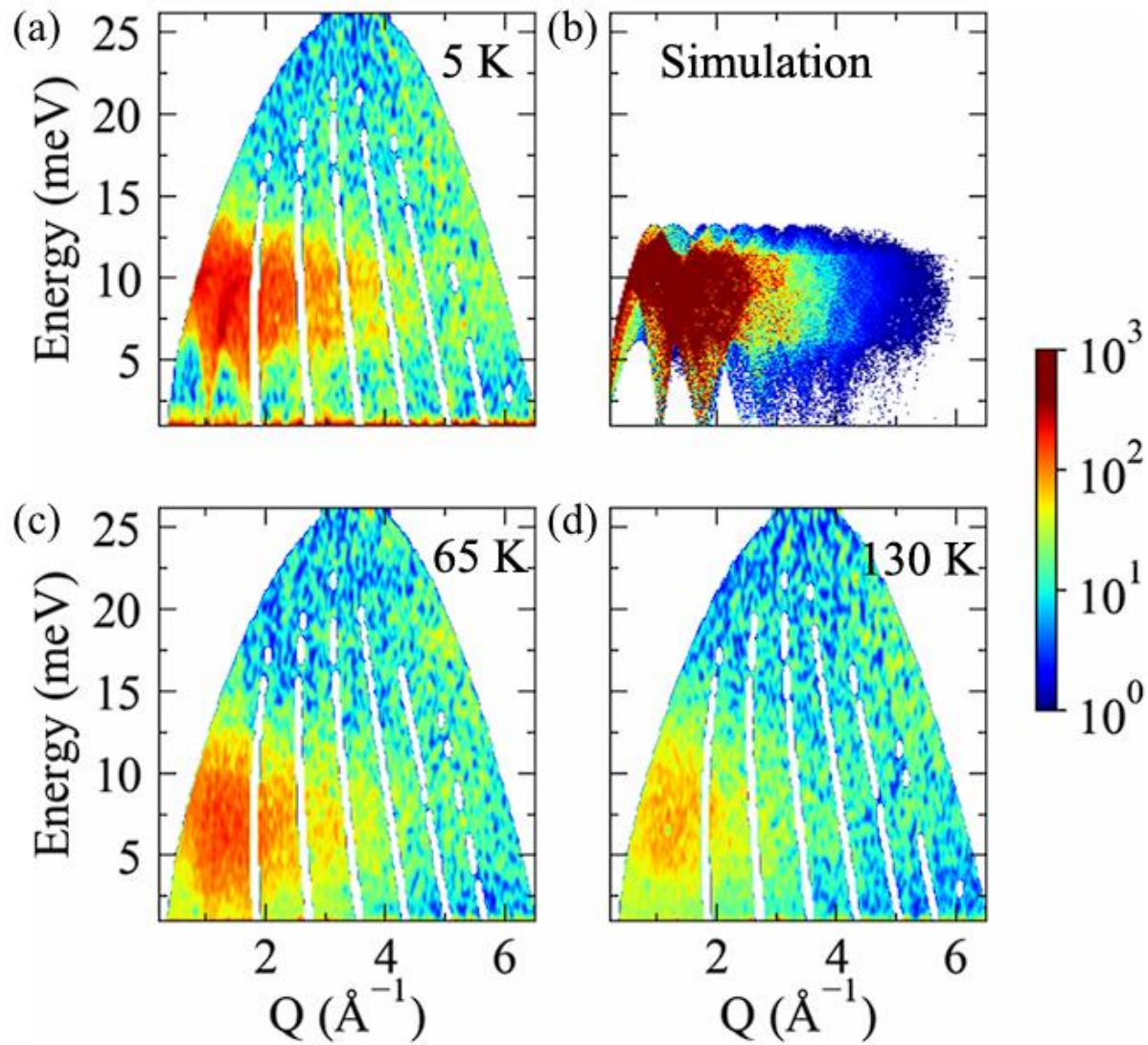
# Magnetism in $\text{CrVO}_4$



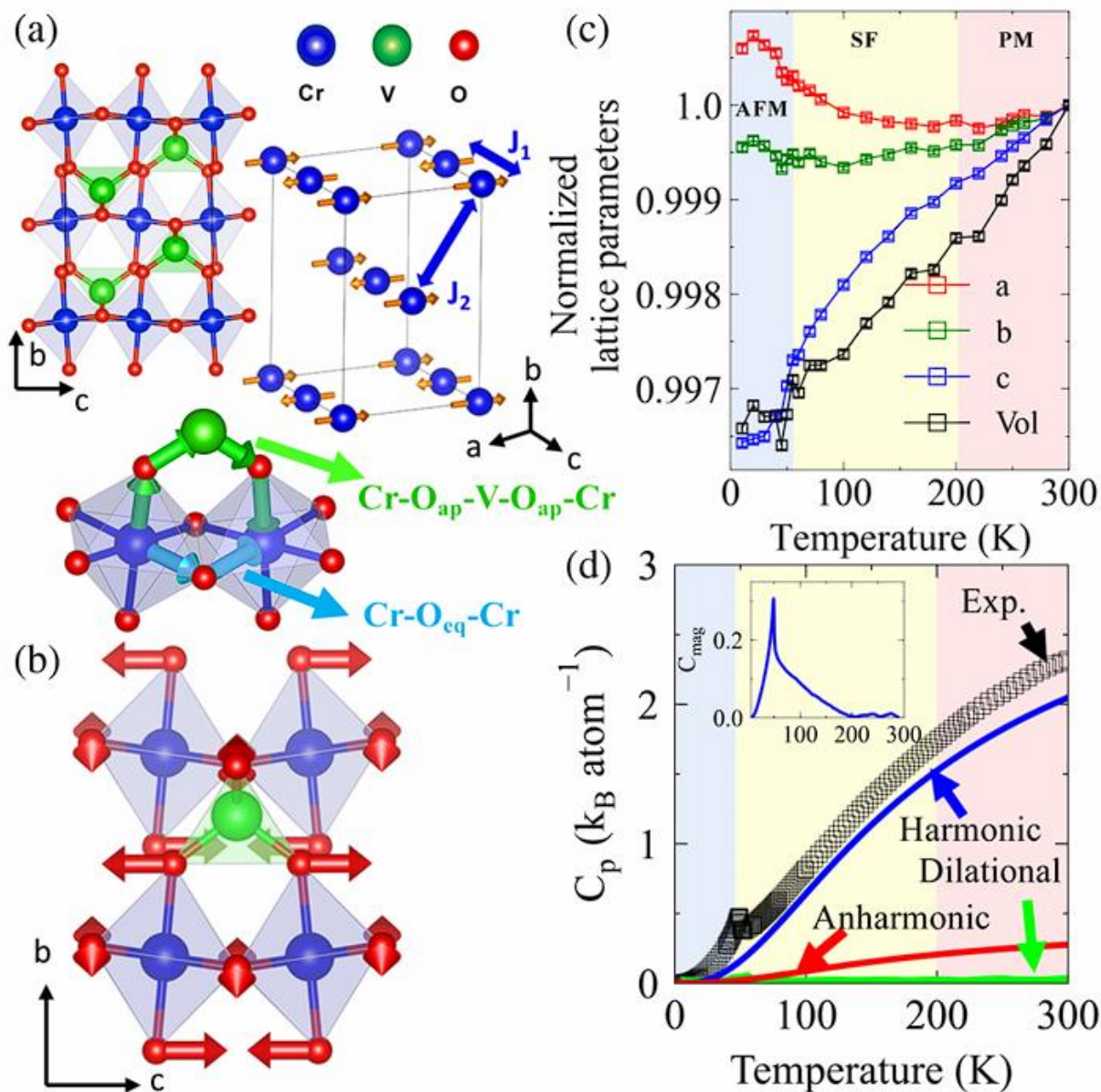
## Neutron Scattering Evidence of A-type AFM



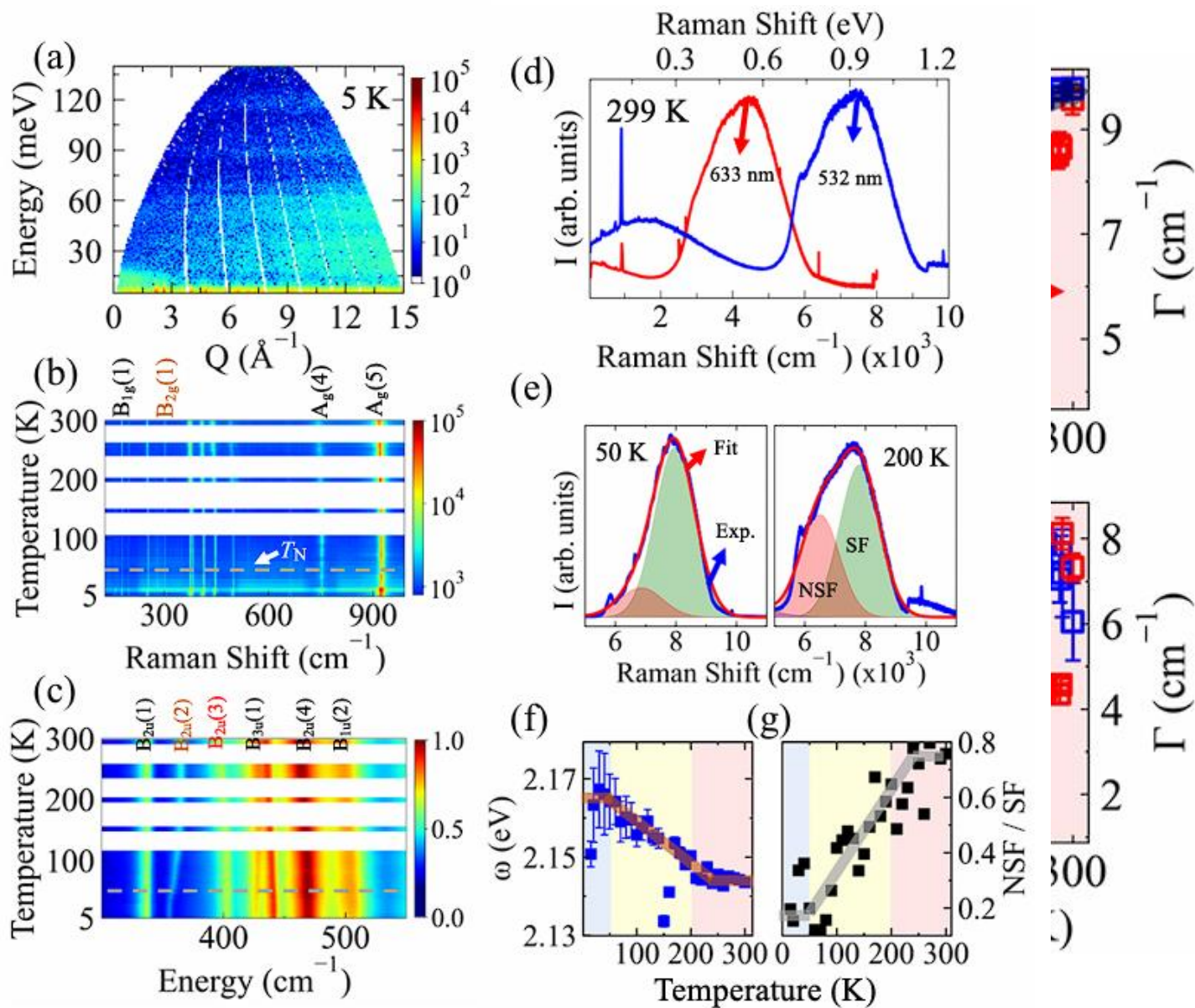
# Magnons in $\text{CrVO}_4$



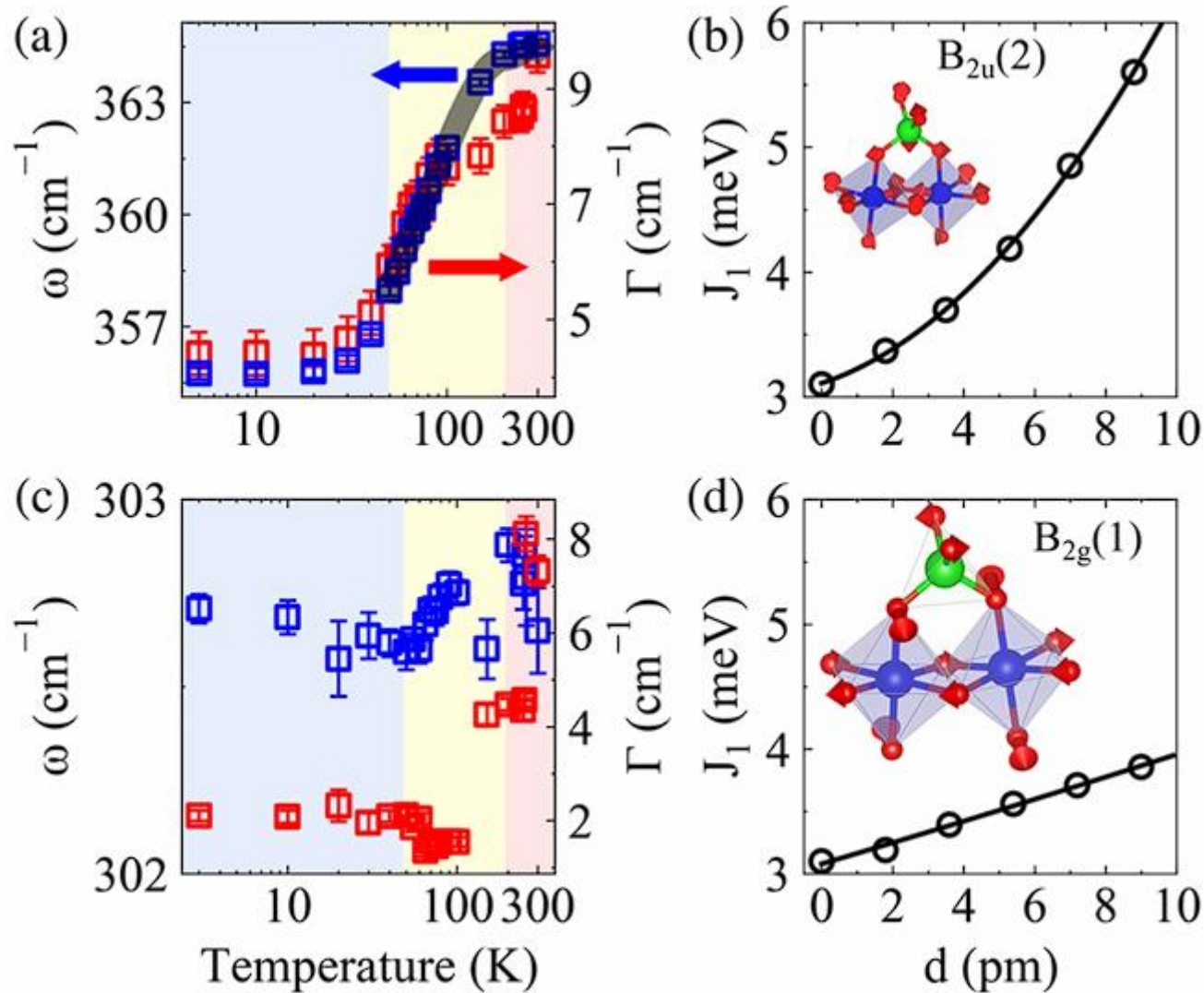
# Magnetism in $\text{CrVO}_4$



# Phonons and Electronic renormalization in $\text{CrVO}_4$

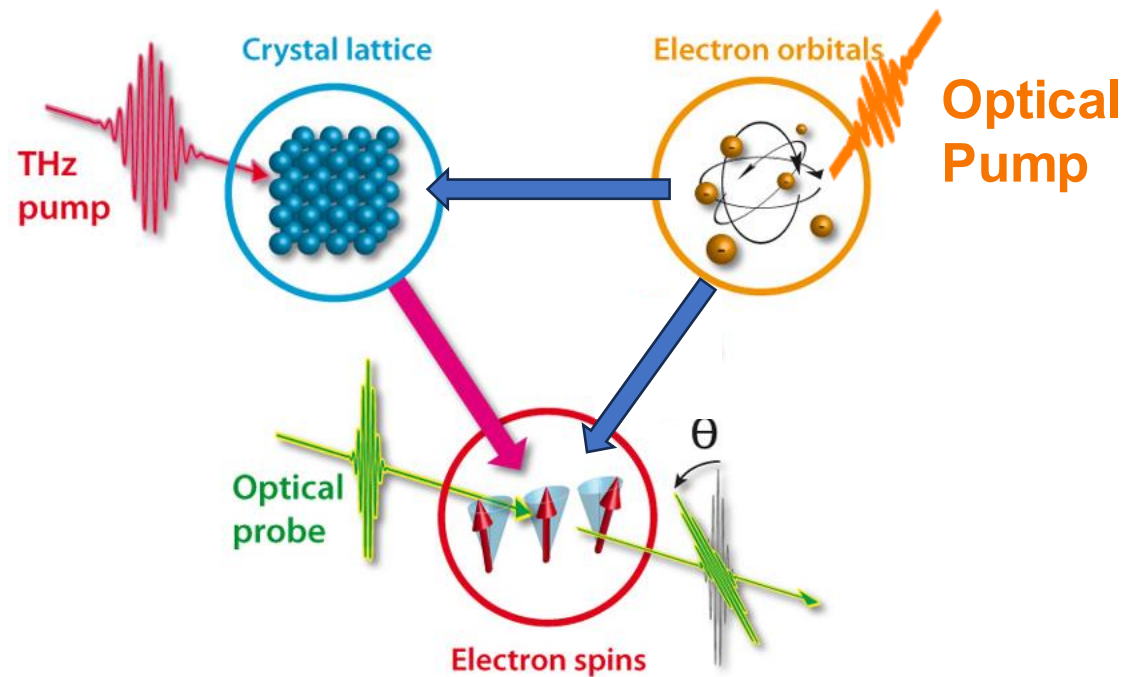


# Phonon Selectivity of the Exchange Interactions in $\text{CrVO}_4$

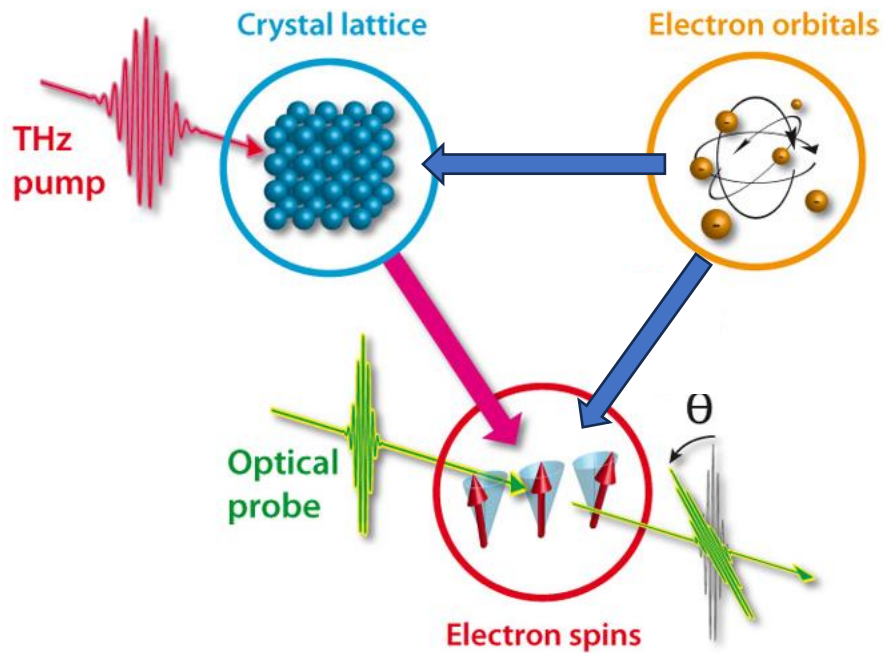


# Summary

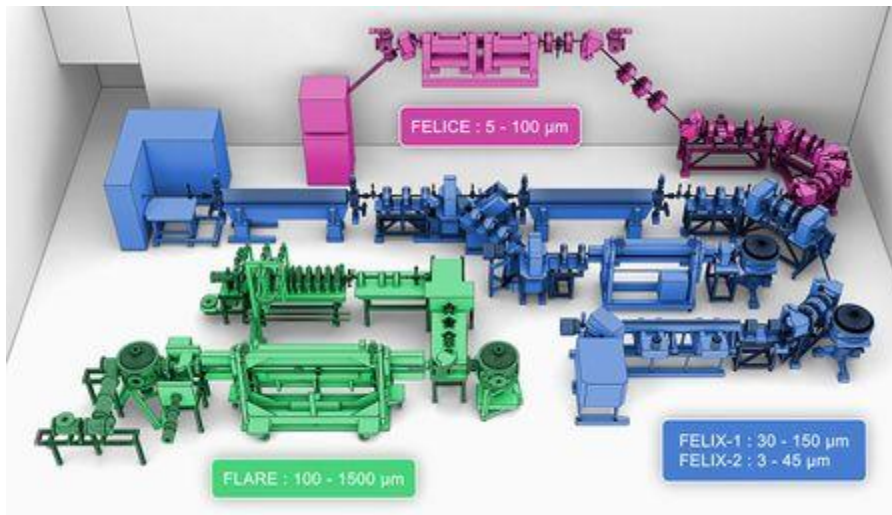
- Demonstrate the role of coupled interaction in deducing the physical properties of the material.
- We selectively deduce the directional contribution to the overall response through angle-resolved polarized Raman spectroscopy.
- Finally, the approach allows us to disentangle the Phonon specific contributions to magnetism which can help to understand the nonlinear phononic pathway for controlling the magnetism.



# Future Perspective with HFML-FELIX



- $\lambda$  tunable between 2.7 $\mu\text{m}$  and 1.5mm or between 0.2 – 90 THz
- 10s  $\mu\text{J}$ 's per (sub)ps pulse
- 0.4-4 % bandwidth, Fourier-transform limited



FELIX Pump Optical Imaging Probe

FELIX Pump Raman as probe

FELIX Pump Optical Probe