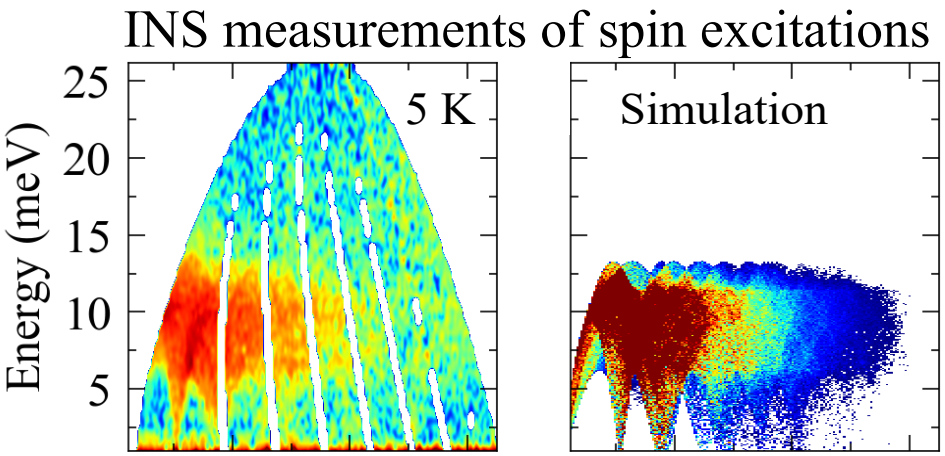
Probing spin-phonon coupling in antiferromagnetic oxides



Dipanshu Bansal
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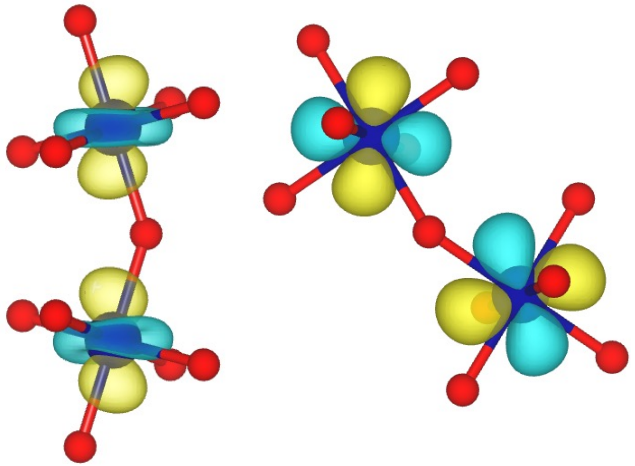
https://sites.google.com/view/spectroscopy-iitb/



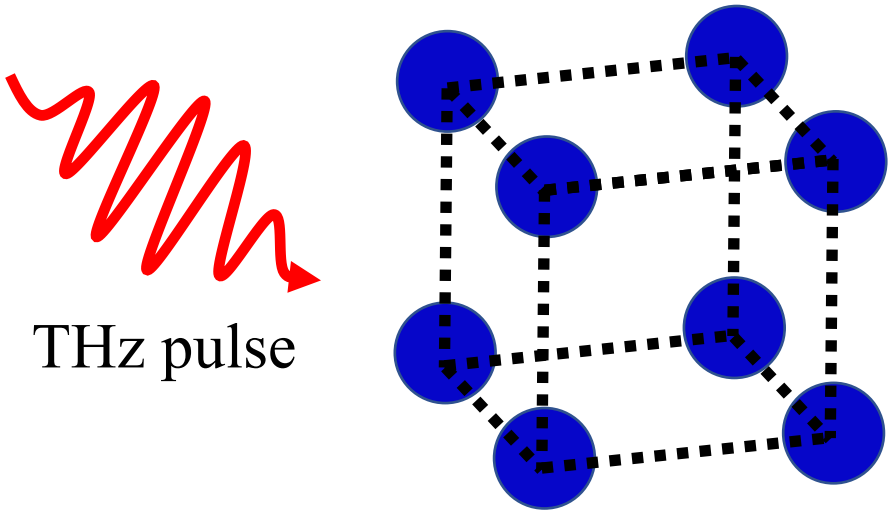
1. A.P. Roy, Jayakrishnan SS, …, and D. Bansal. **"Evidence of strong orbital-selective spin-orbital-phonon coupling in $CrVO_4$."** PRL, Vol. 132, 026701, 2024.
2. Jayakrishnan SS and D. Bansal. **"Effect of spin-phonon coupling on phonons and magnons in the antiferromagnet NiO."** PRB, Vol. 111, 104306, 2025.
3. Jayakrishnan SS and D. Bansal. **"Coherent phonon excitation induced evolution of spin dynamics and spin-phonon coupling in yttrium orthochromite."** PRB, Vol. 112, 214419, 2025.

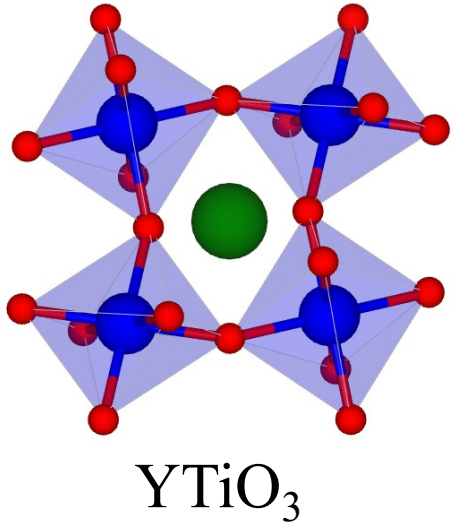
https://sites.google.com/view/spectroscopy-iitb/

Transient evolution of spin-phonon coupling

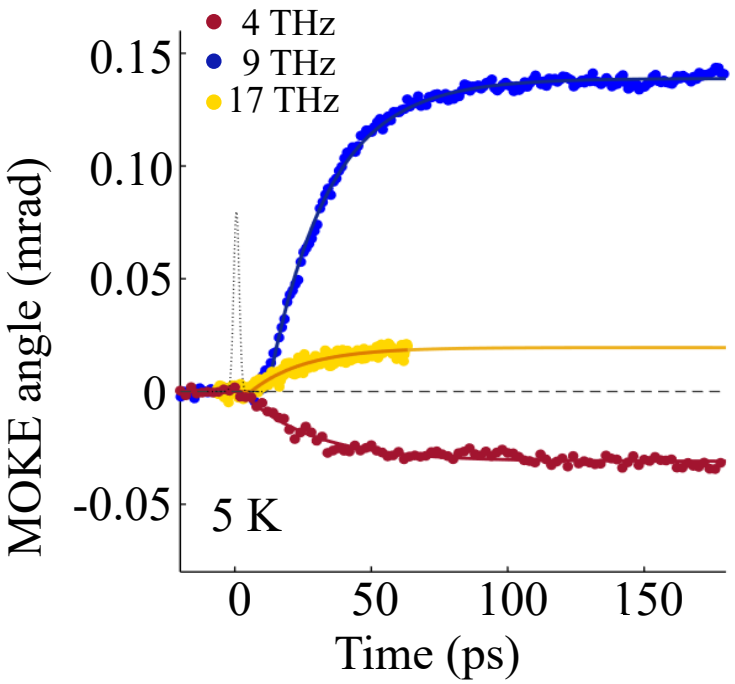
[1]Disa, *et al.*, Nature 617.7959 (2023): 73-78
[2]Disa, *et al.*, Nature 16.9 (2020): 937-941
[3]Ilyas, *et al.*, Nature 636.8043 (2024): 609-614

Transient evolution of spin-phonon coupling





Strengthening of FM order in $YTiO_3$[1]



[1]Disa, *et al.*, Nature 617.7959 (2023): 73-78
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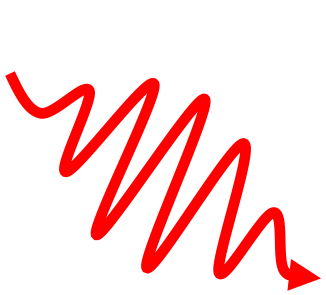
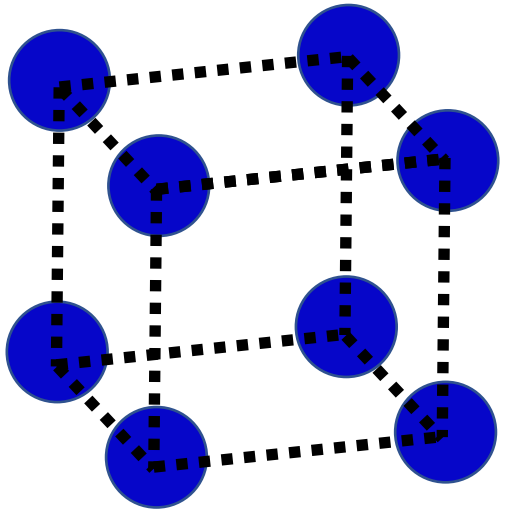
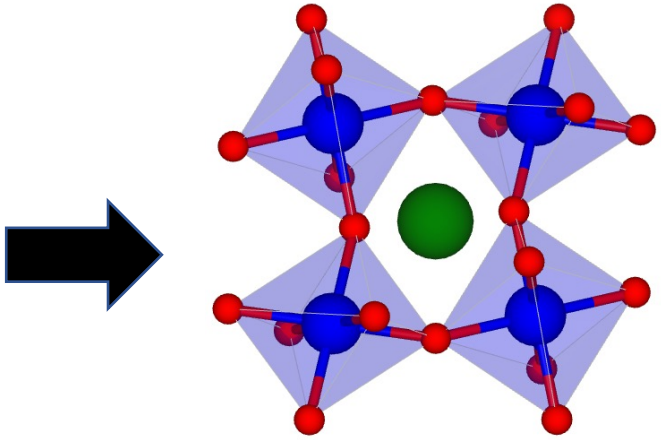
Transient evolution of spin-phonon coupling



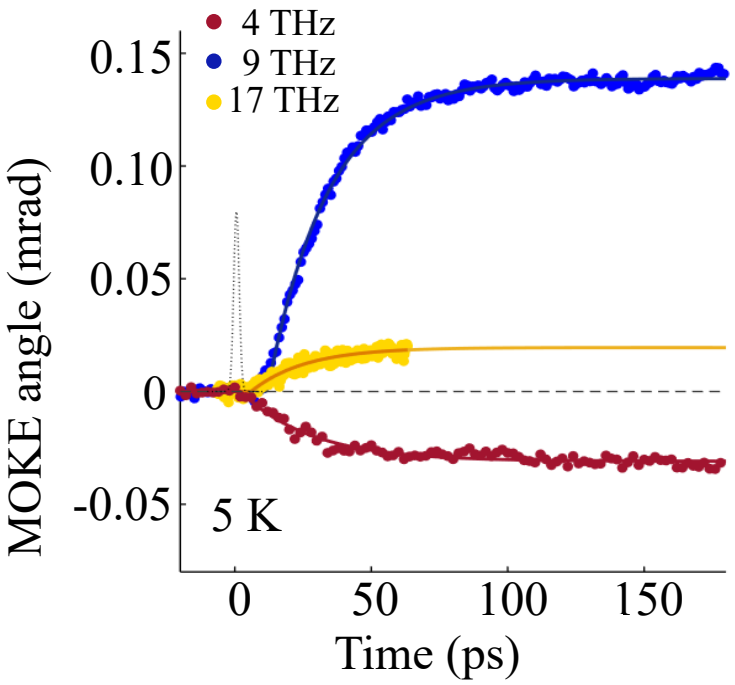
THz pulse

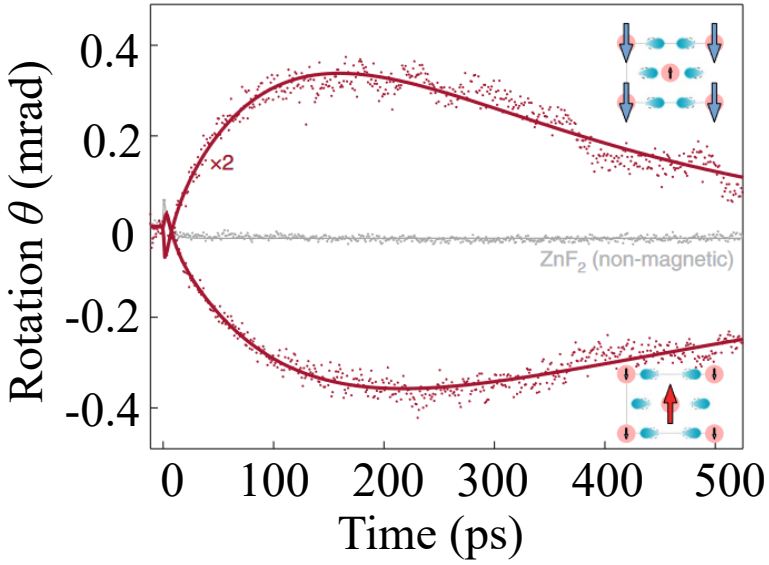




$YTiO_3$

Strengthening of FM order in $YTiO_3$[1]



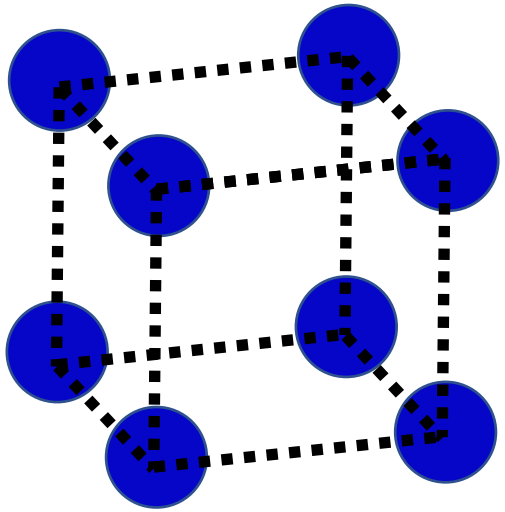


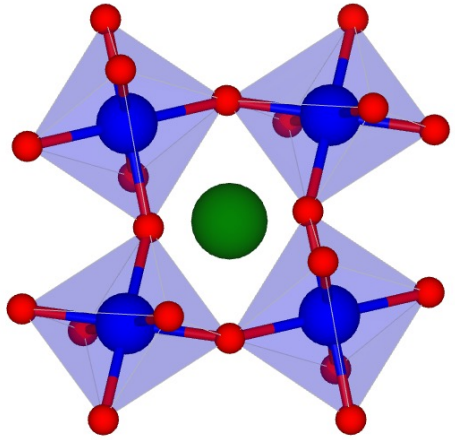
AFM to FM in CoF_2[2]

[1]Disa, *et al.*, Nature 617.7959 (2023): 73-78
[2]Disa, *et al.*, Nature 16.9 (2020): 937-941
[3]Ilyas, *et al.*, Nature 636.8043 (2024): 609-614

Transient evolution of spin-phonon coupling

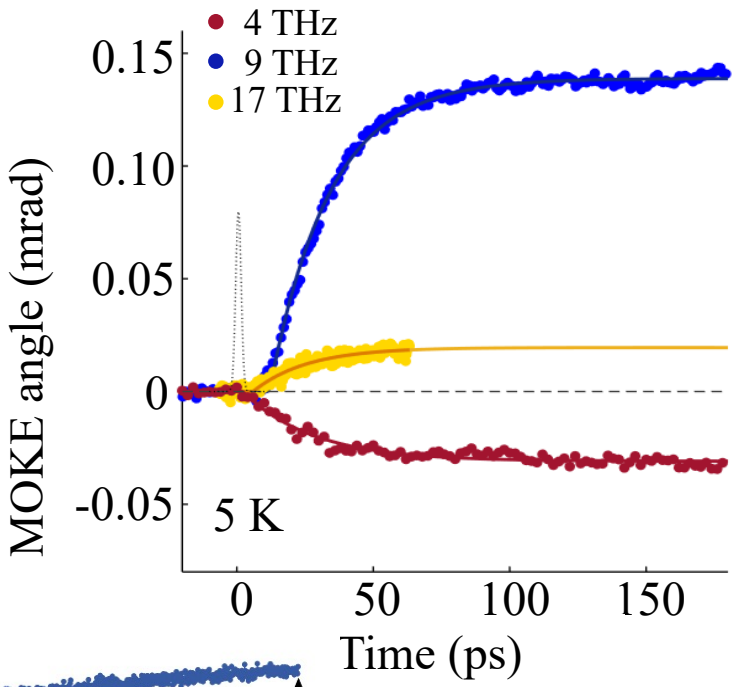
THz pulse

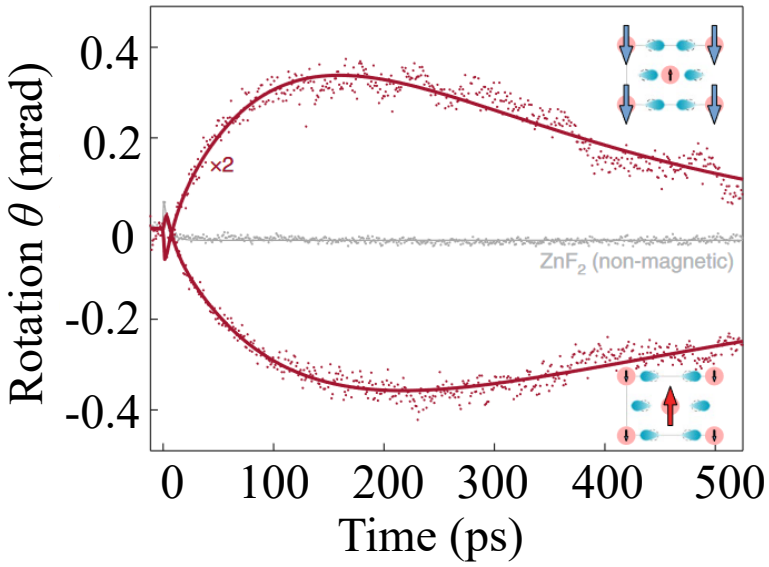




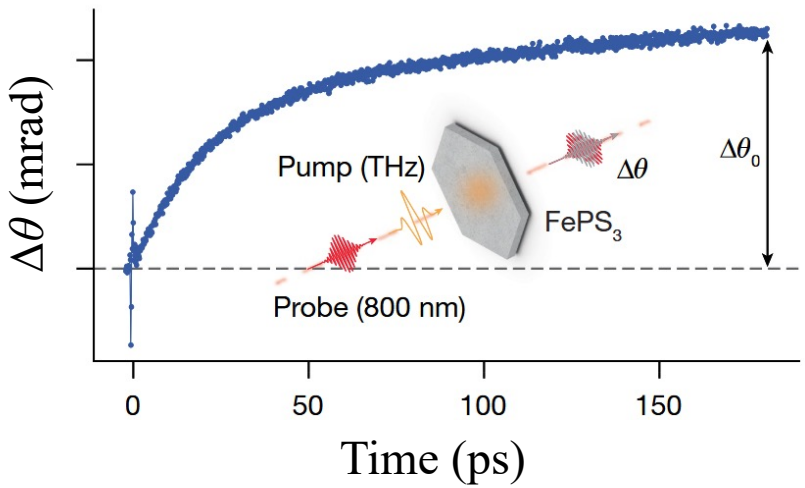
$YTiO_3$

Strengthening of FM order in $YTiO_3$[1]





AFM to FM in CoF_2[2]

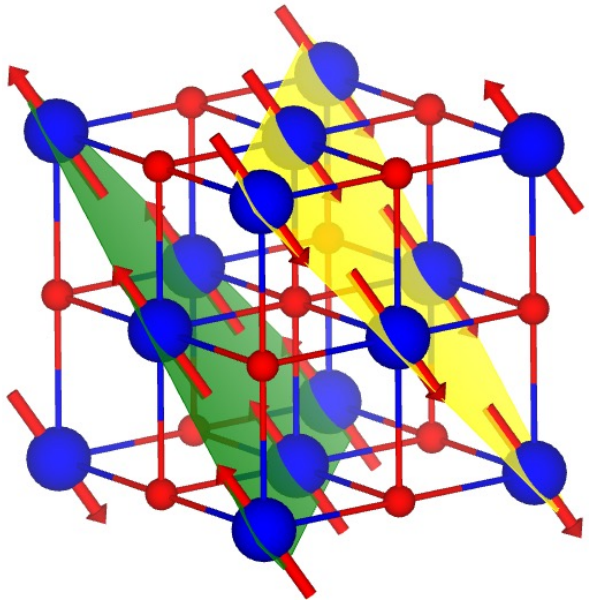


Magnetization in $FePS_3$[3]

[1]Disa, *et al.*, Nature 617.7959 (2023): 73-78
[2]Disa, *et al.*, Nature 16.9 (2020): 937-941
[3]Ilyas, *et al.*, Nature 636.8043 (2024): 609-614

Nickel Oxide

Nickel Oxide



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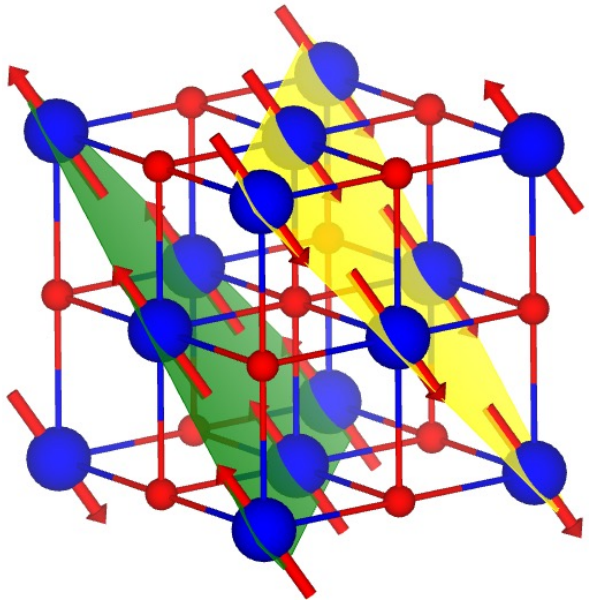
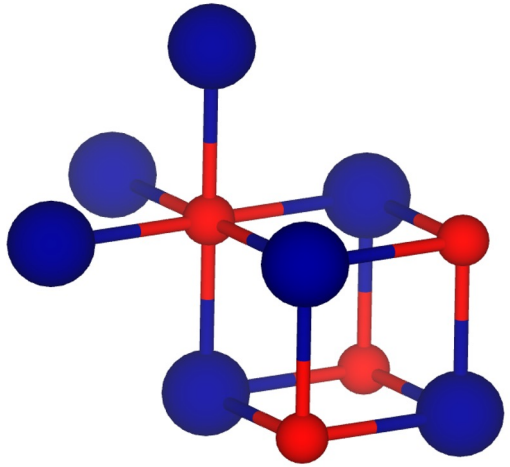
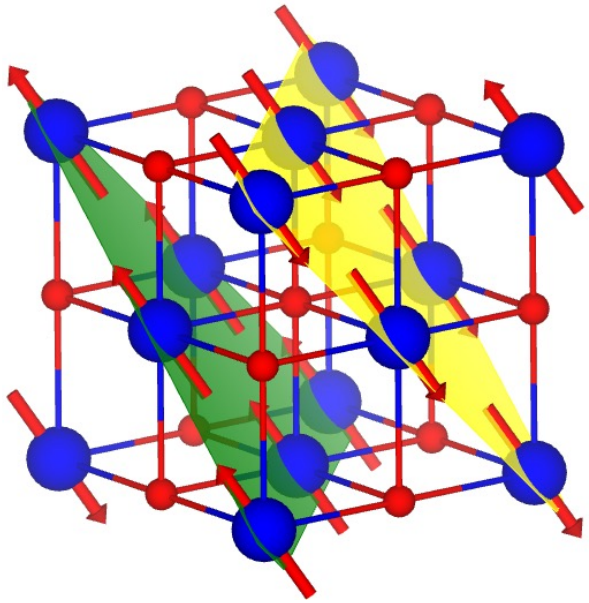
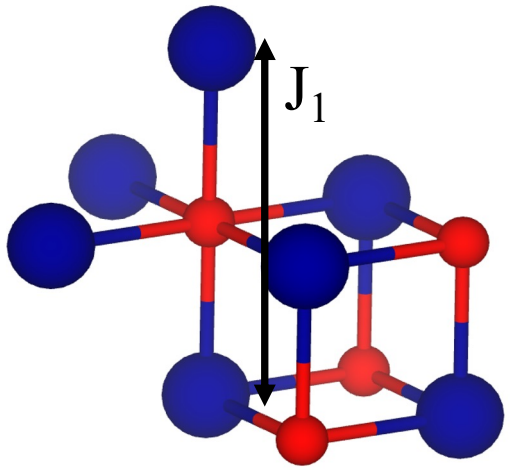




Ni O

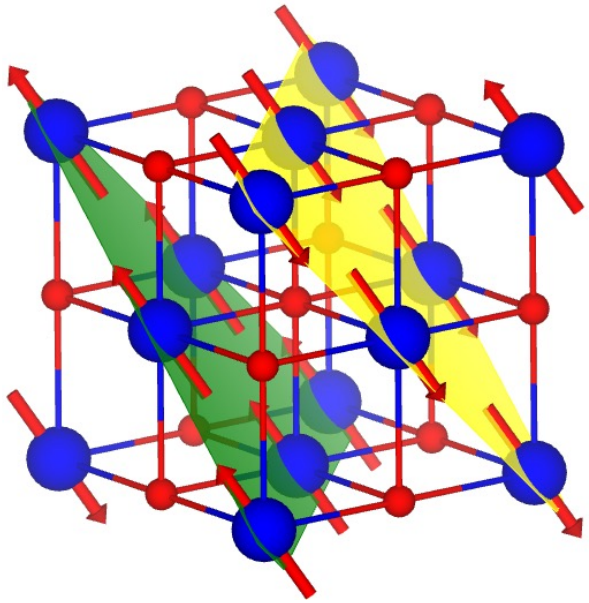
Nickel Oxide

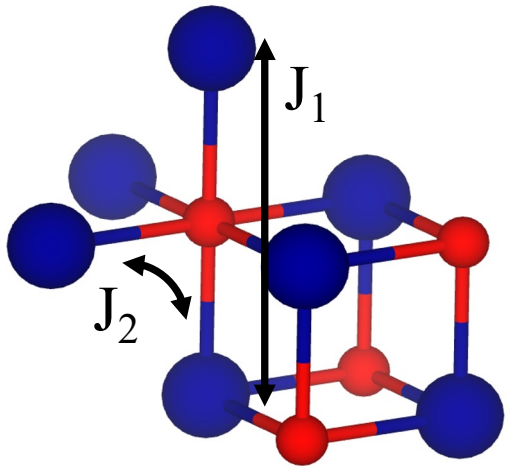




Ni O

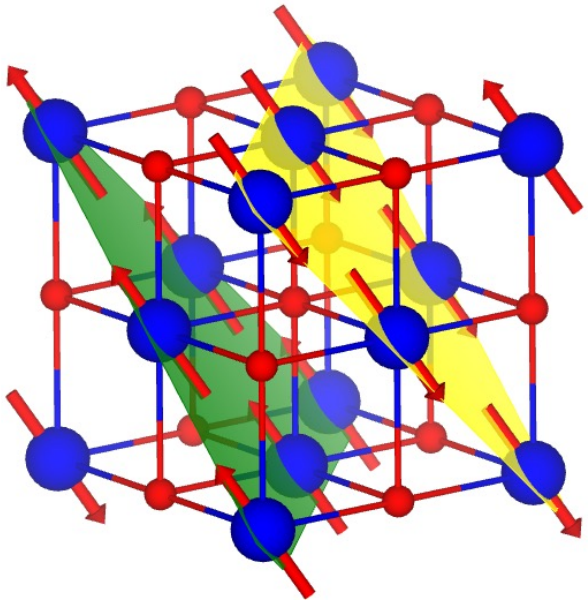
Nickel Oxide

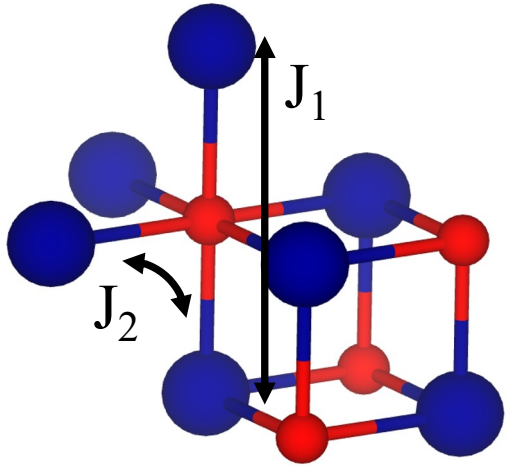


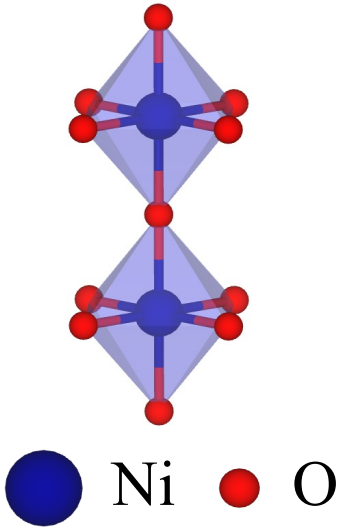


Ni O

Nickel Oxide

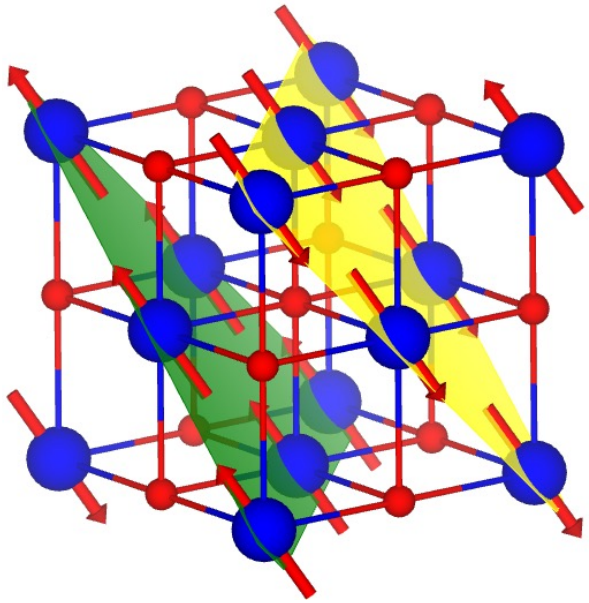


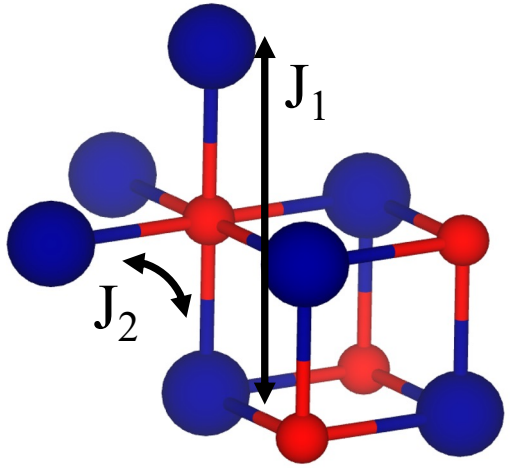


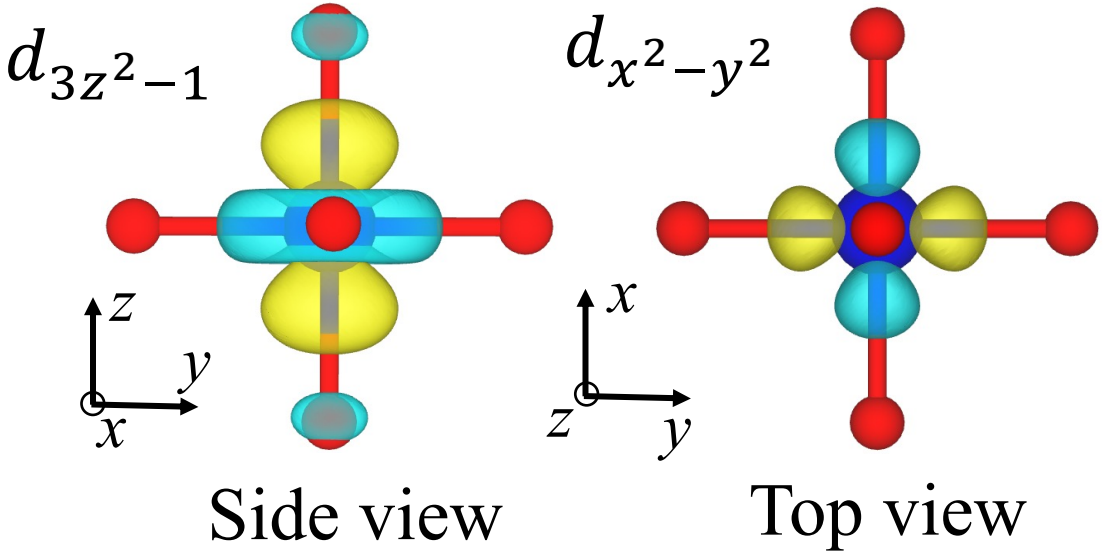


Ni O

Nickel Oxide

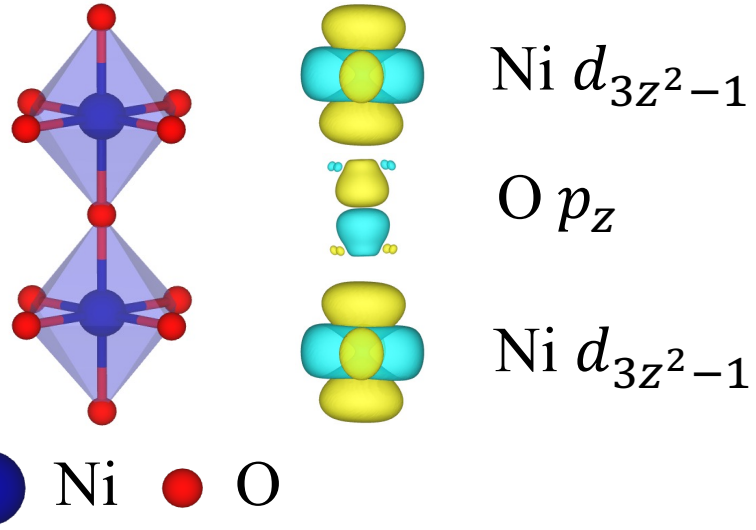






Side view

Top view



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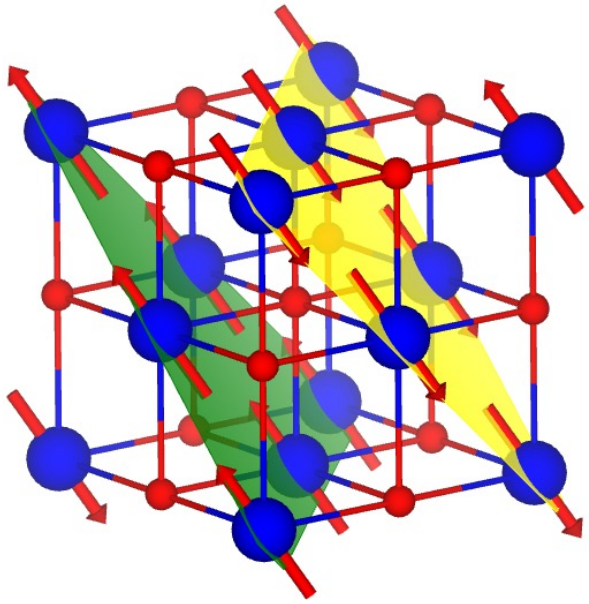
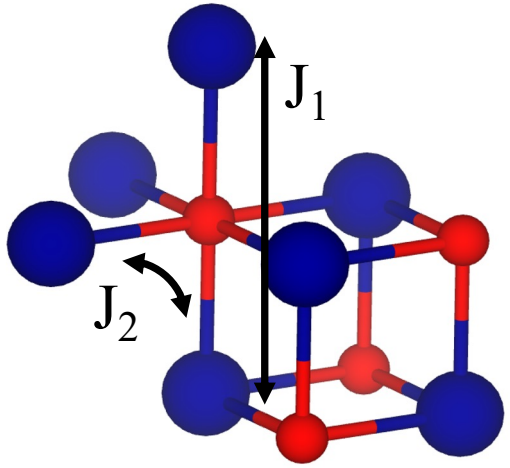
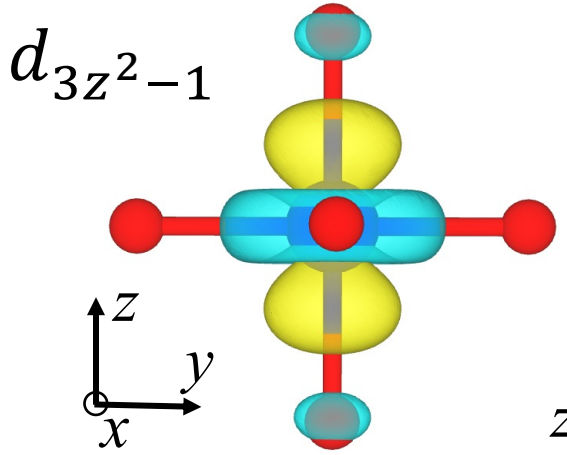
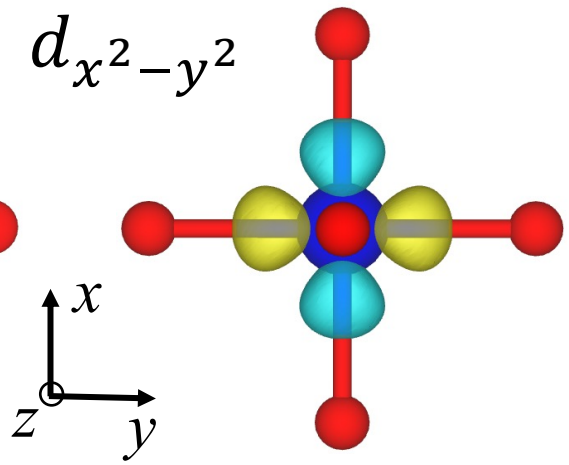






Side view



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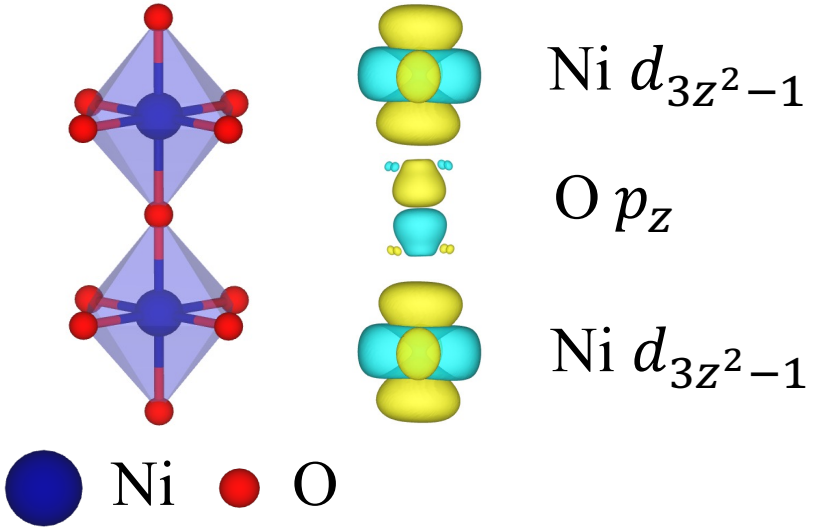

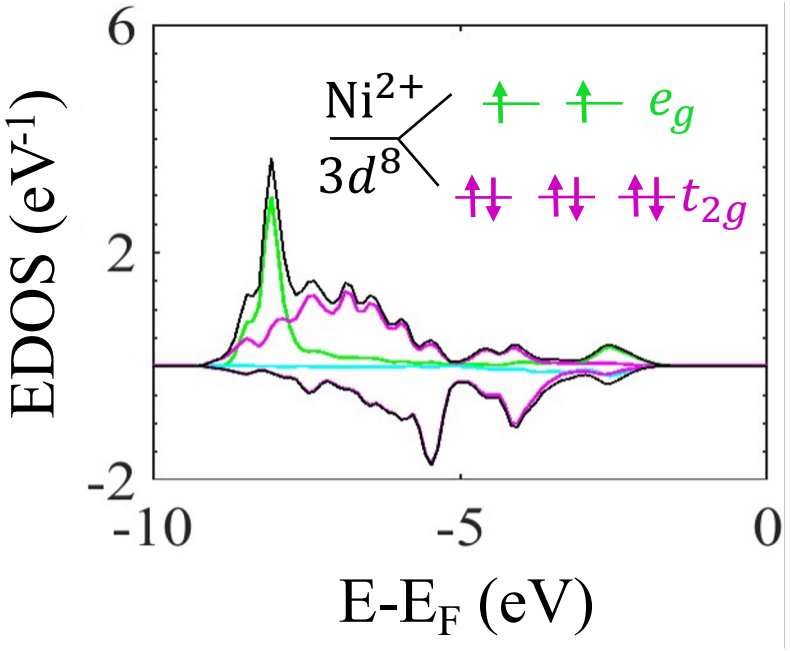

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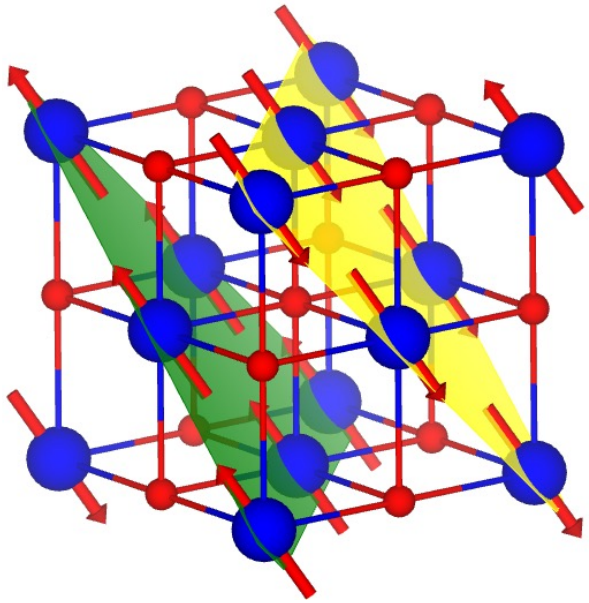
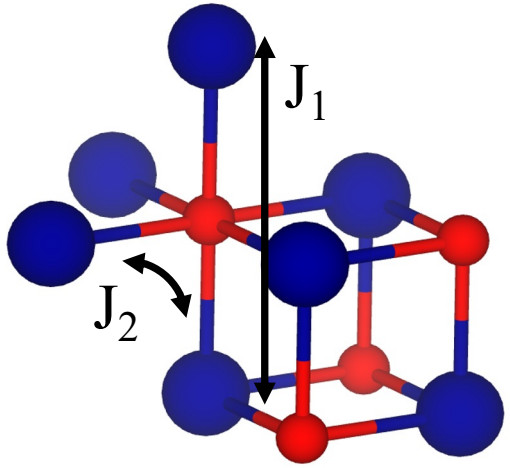
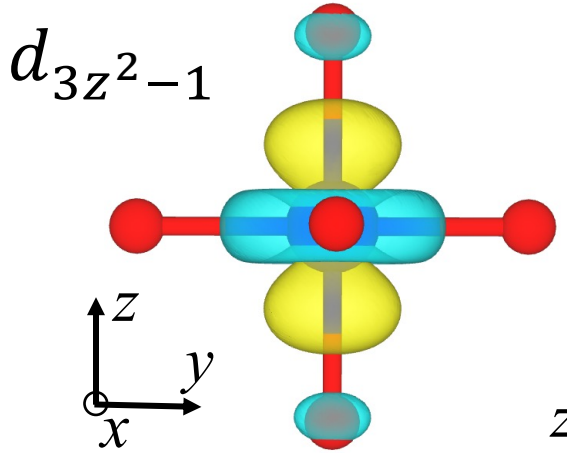
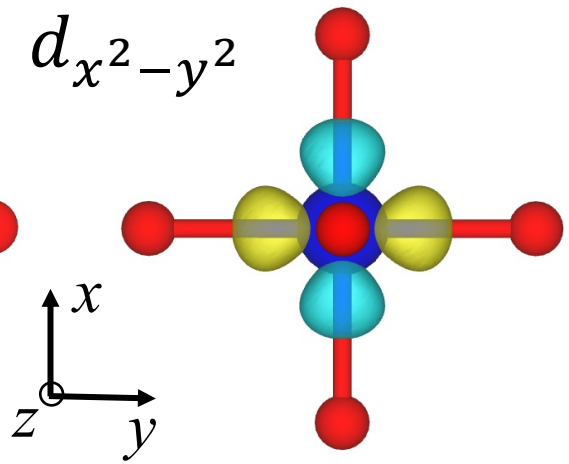


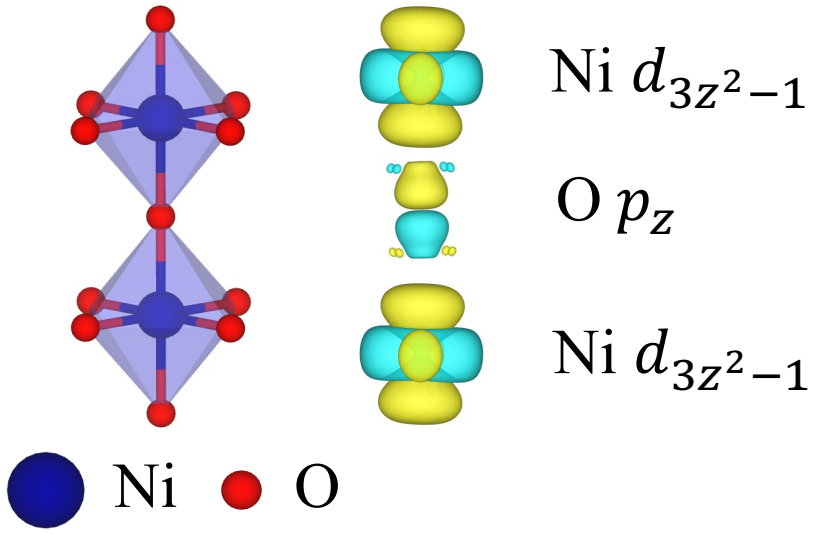


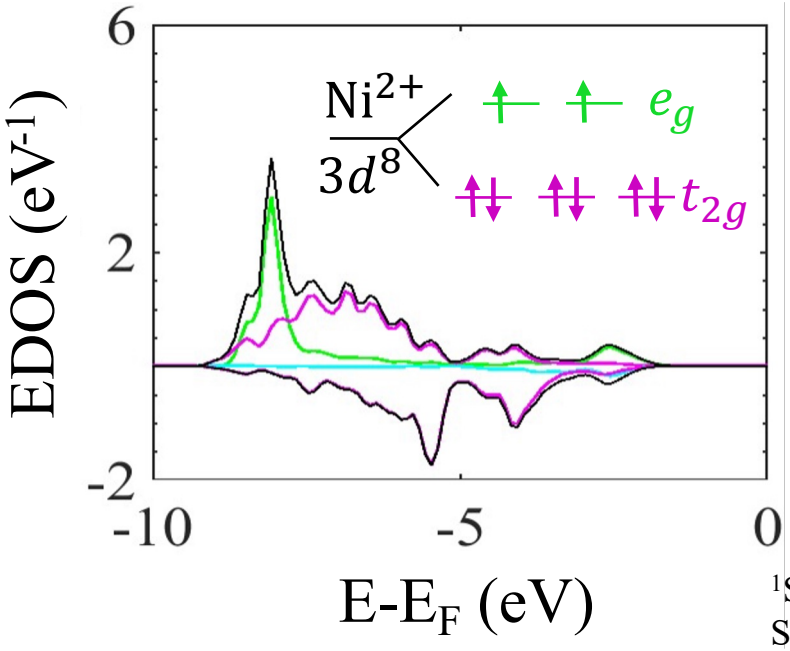


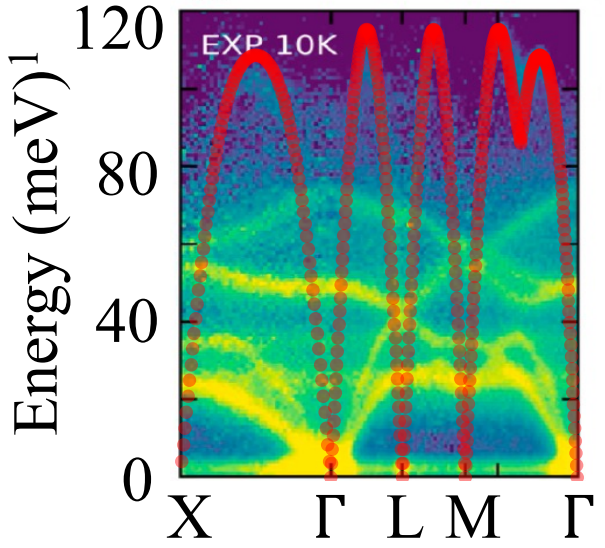
Side view



Top view

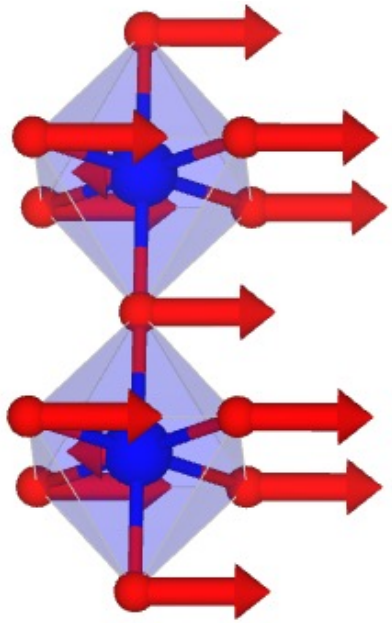






[1]Sun, *Materials Today Physics* 35, 101094 (2023)
SS, *PRB* 111, 104306 (2025)

Spin wave renormalization for TO mode perturbation



Jayakrishnan SS and D. Bansal. **"Effect of spin-phonon coupling on phonons and magnons in the antiferromagnet NiO."** PRB, Vol. 111, 104306, 2025.

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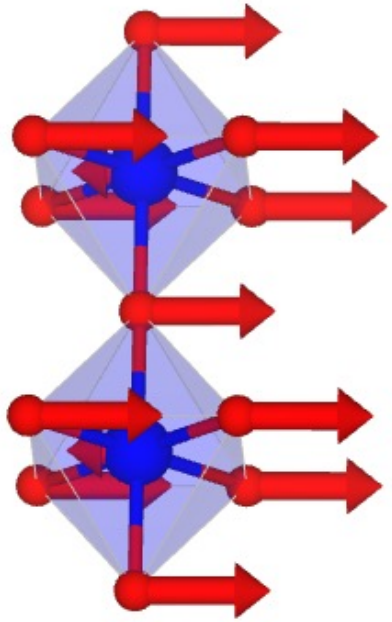

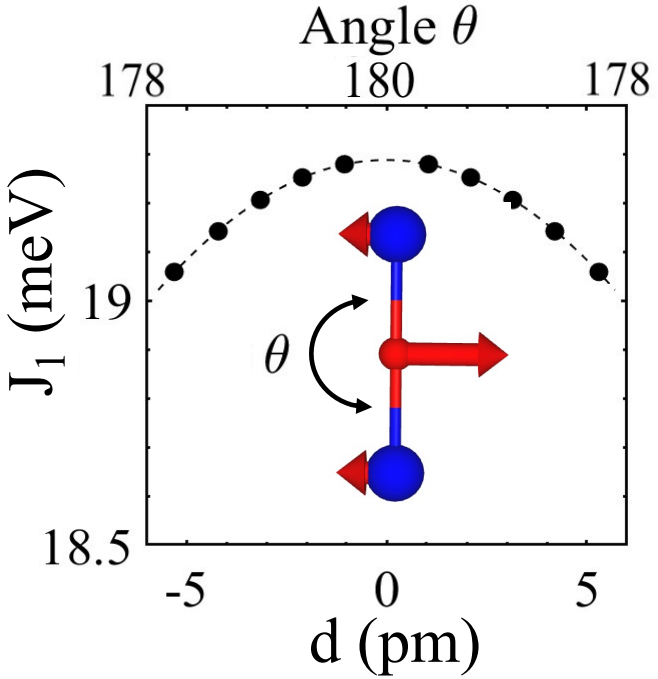

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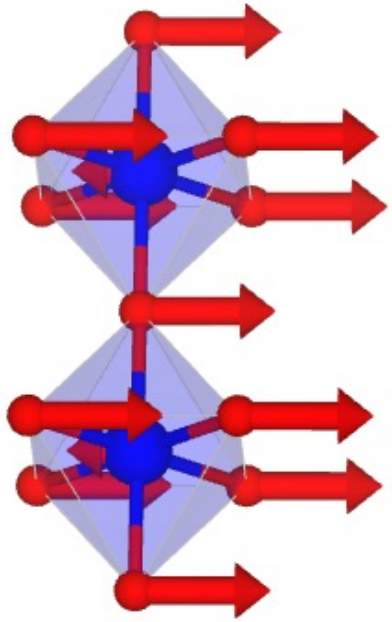

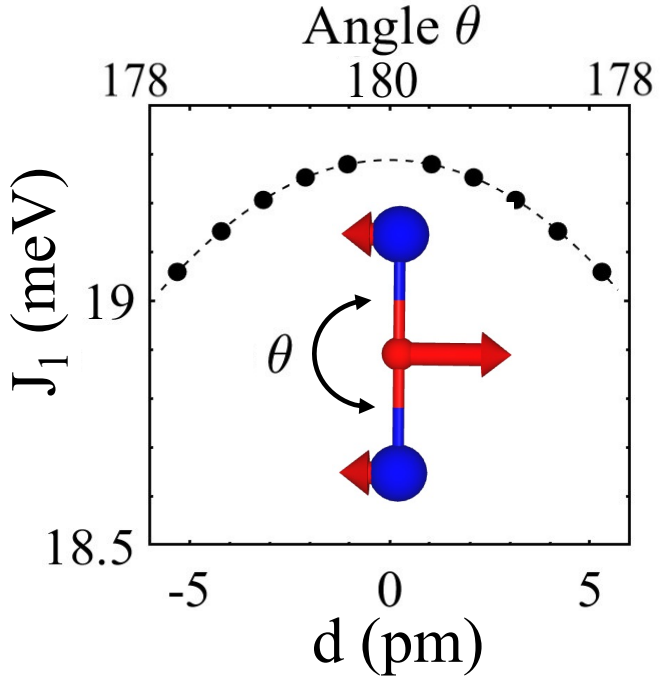

Quadratic dependency with perturbation

$$J_{ij} = -\frac{1}{2\pi}\int_{-\infty}^{E_F} d\epsilon \sum_{mm'm''m'''} \mathrm{Im}(\Delta_i^{mm'} G_{ij,\downarrow}^{mm'} \Delta_j^{m''m'''} G_{ji,\uparrow}^{m''m'''})$$

PRB **91**, 224405, 2015

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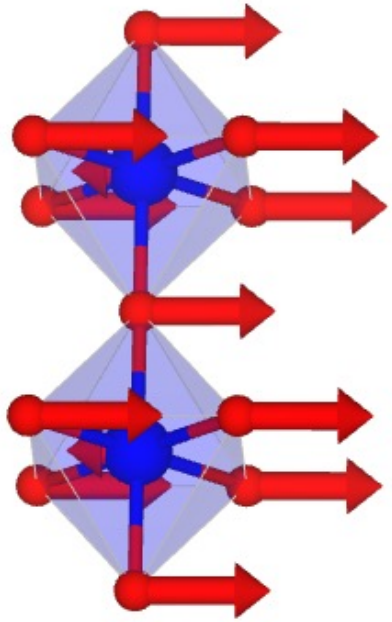
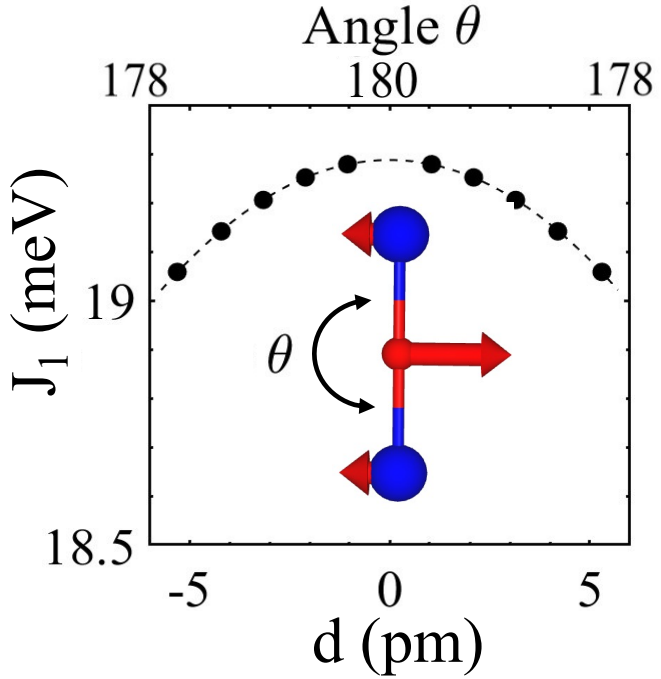
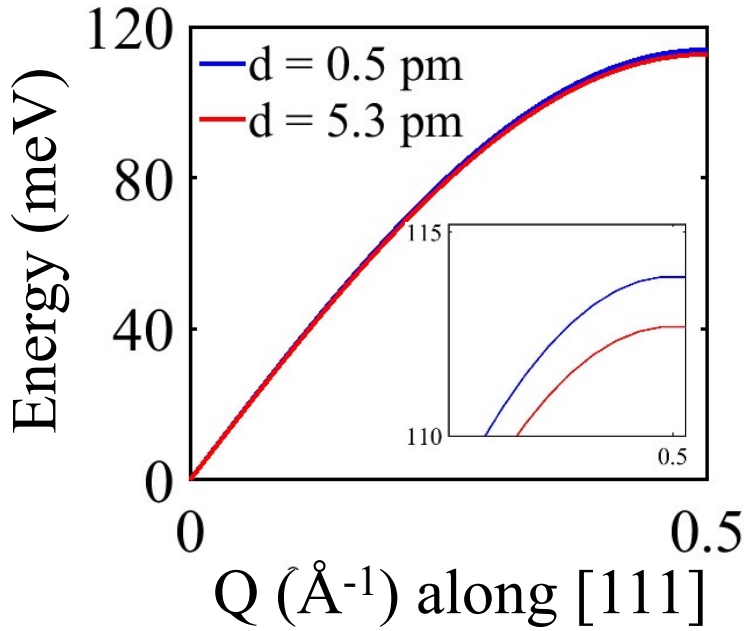
Spin wave renormalization for TO mode perturbation





Quadratic dependency with perturbation



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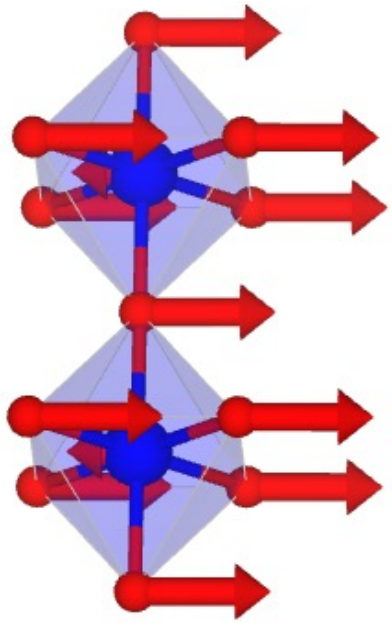
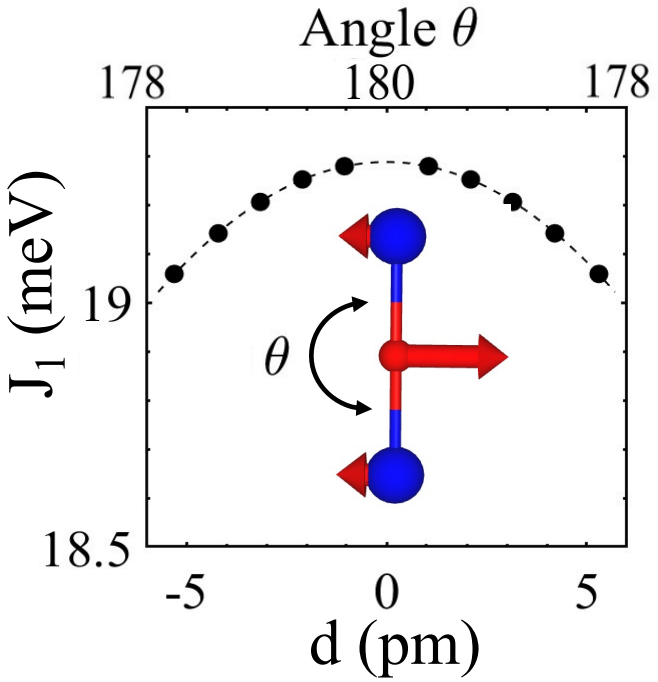
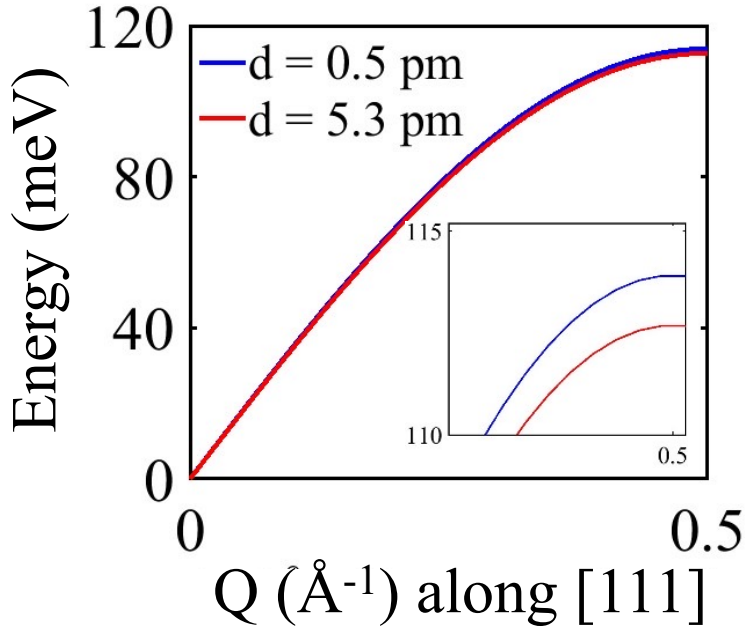
Spin wave renormalization for TO mode perturbation

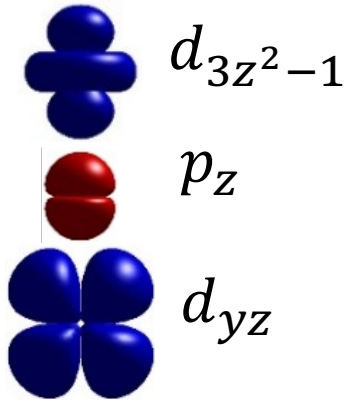


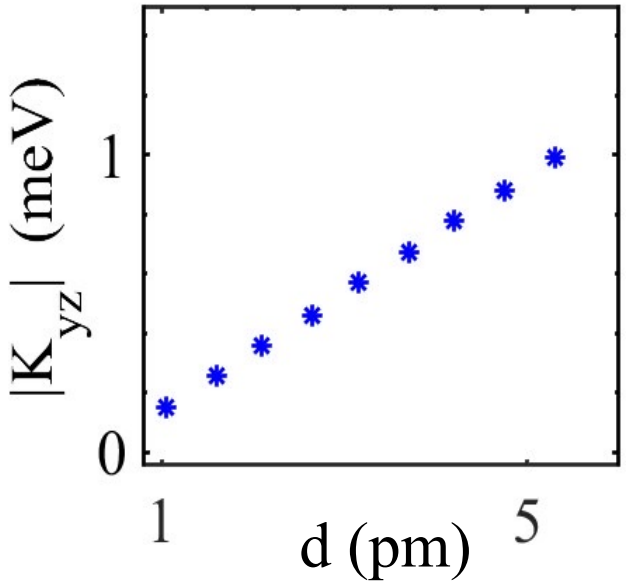


Quadratic dependency with perturbation



Off-diagonal interaction terms

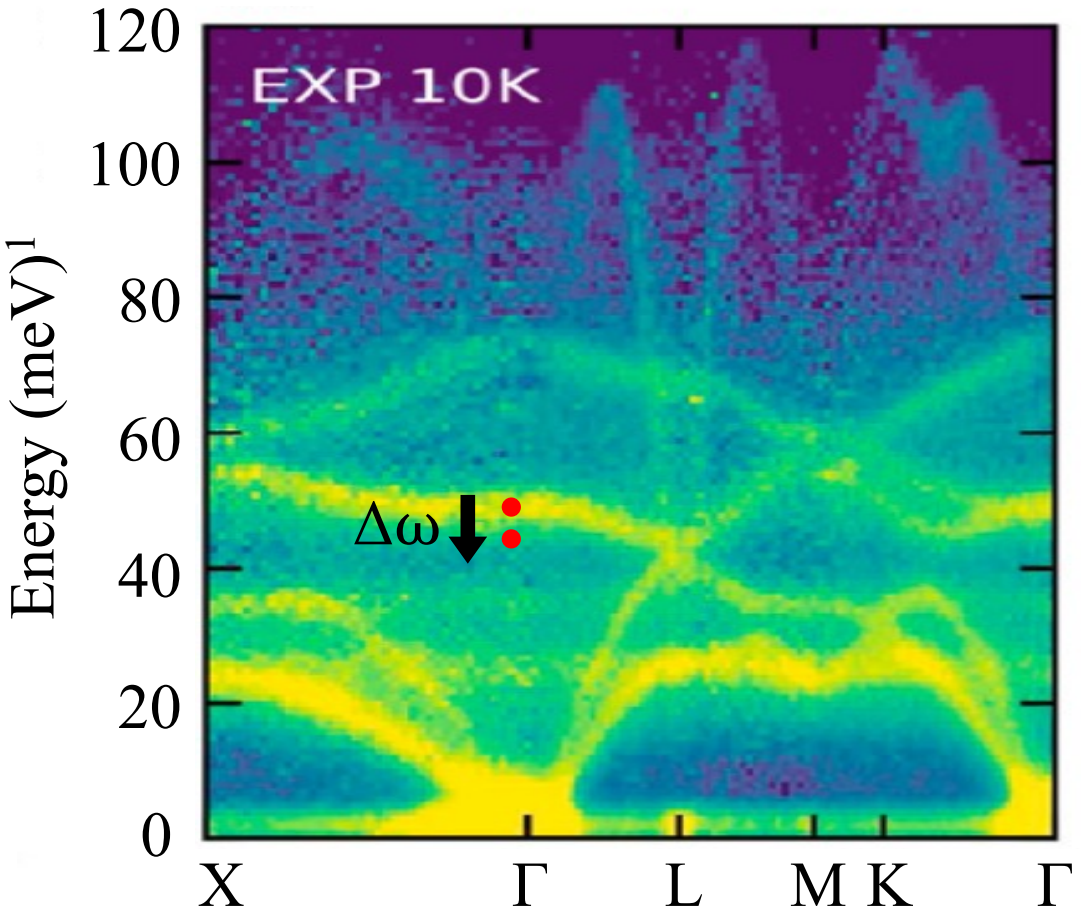




Jayakrishnan SS and D. Bansal. **"Effect of spin-phonon coupling on phonons and magnons in the antiferromagnet NiO."** PRB, Vol. 111, 104306, 2025.

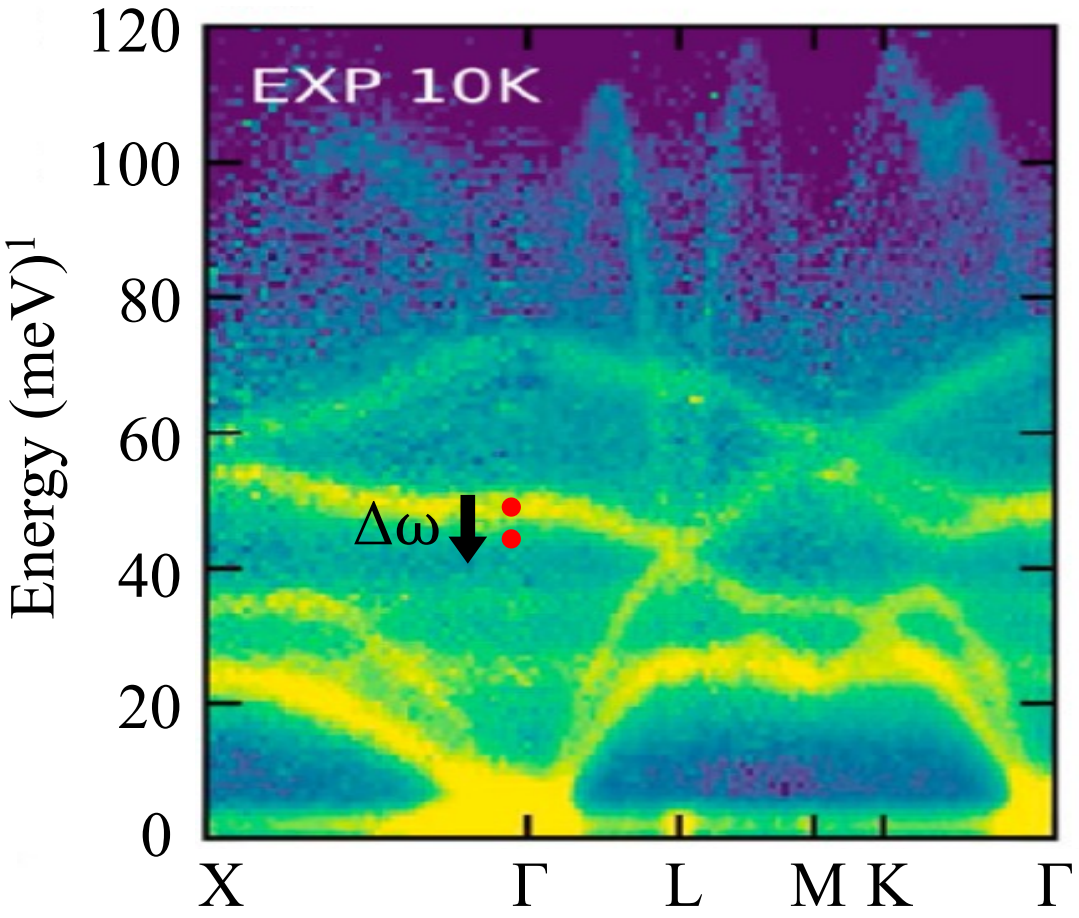
Phonon renormalization in NiO

Phonon renormalization in NiO



Sun, *Materials Today Physics* 35, 101094 (2023)

Phonon renormalization in NiO



Sun, *Materials Today Physics* 35, 101094 (2023)

Coupled spin-lattice Hamiltonian

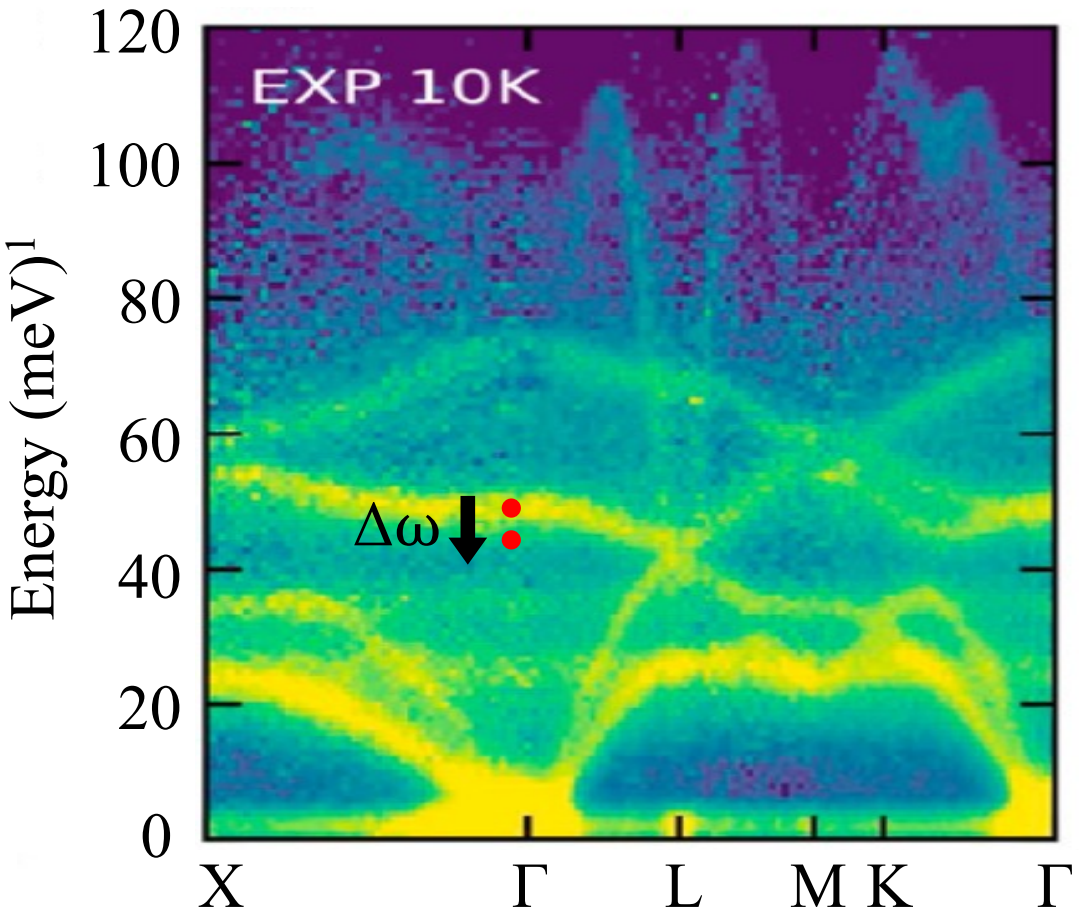
$$\hat{H}_{SLD} = \hat{H}_S + \hat{H}_L + \hat{H}_{SL} + \hat{H}_{LS}$$

$$\Delta\omega = \frac{2.09}{m_r \omega} \frac{\partial^2 J_{ij}}{\partial d^2} m_i m_j$$

Phonon shift due to magnon renormalization

$\Delta\omega = 0.4$ meV (3 cm^{-1})

Phonon renormalization in NiO



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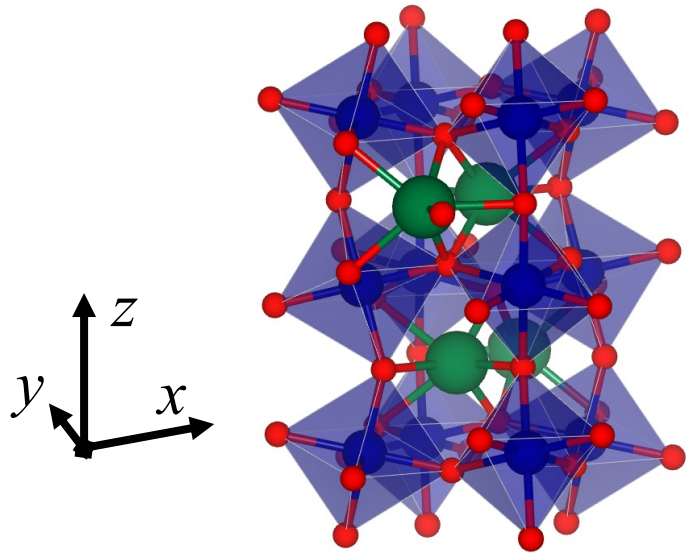
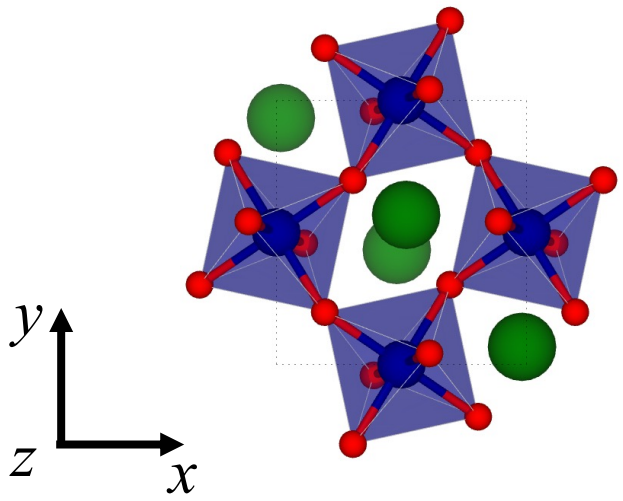
Summary

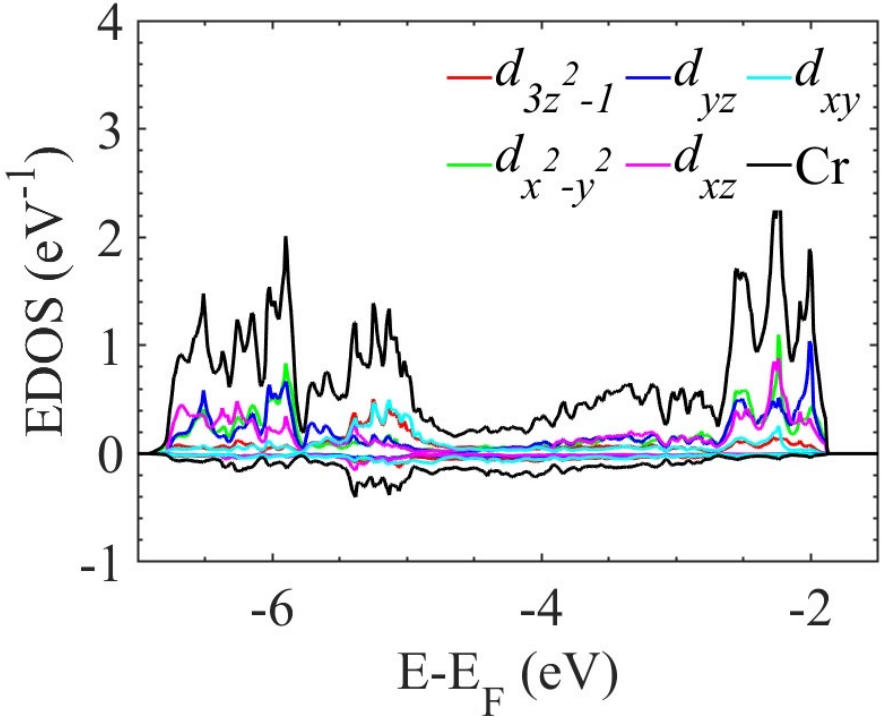
- Exchange interaction changes by 0.3 meV.
- Spin wave changes by 1 meV.
- Phonon changes by 0.4 meV.

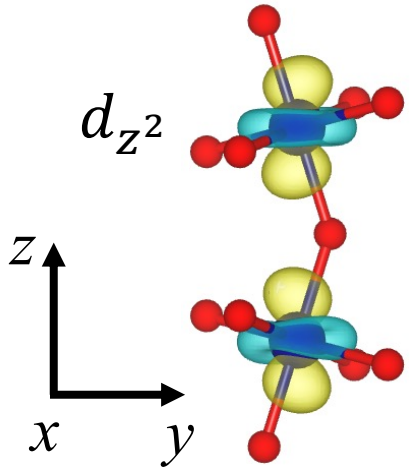
Jayakrishnan SS and D. Bansal. **"Effect of spin-phonon coupling on phonons and magnons in the antiferromagnet NiO."** PRB, Vol. 111, 104306, 2025.

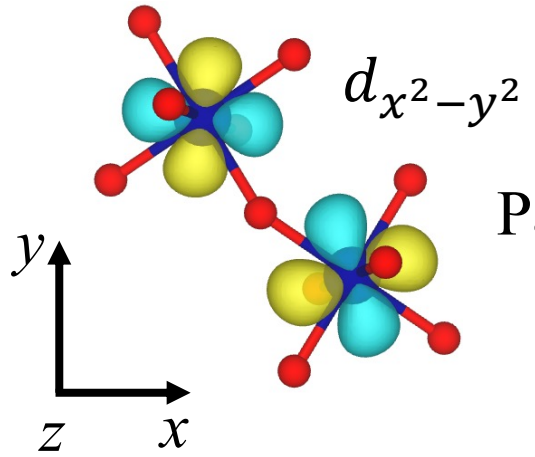
Yttrium orthochromite ($YCrO_3$)











Partial orbital occupancies for Cr d orbitals

Exchange interaction in unperturbed $YCrO_3$

Jayakrishnan SS and D. Bansal*. **"Coherent phonon excitation induced evolution of spin dynamics and spin-phonon coupling in yttrium orthochromite."** Physical Review B, 112, 214419, 2025.

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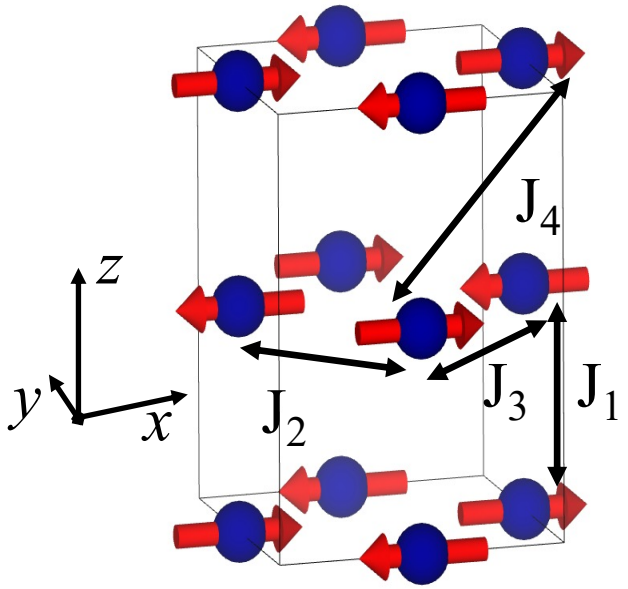

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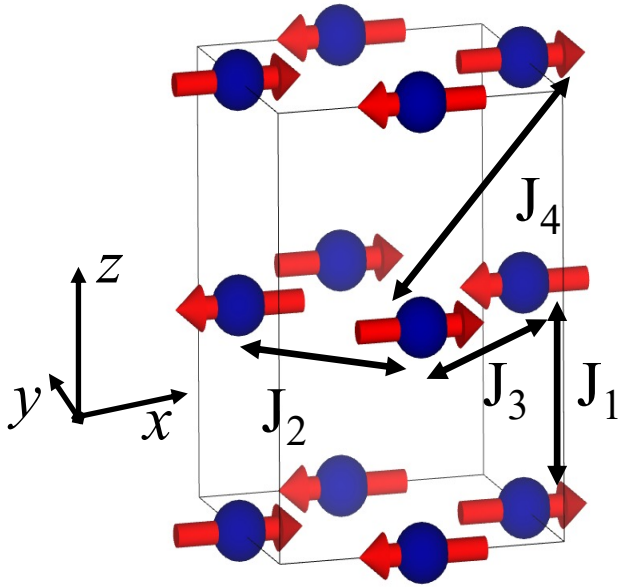

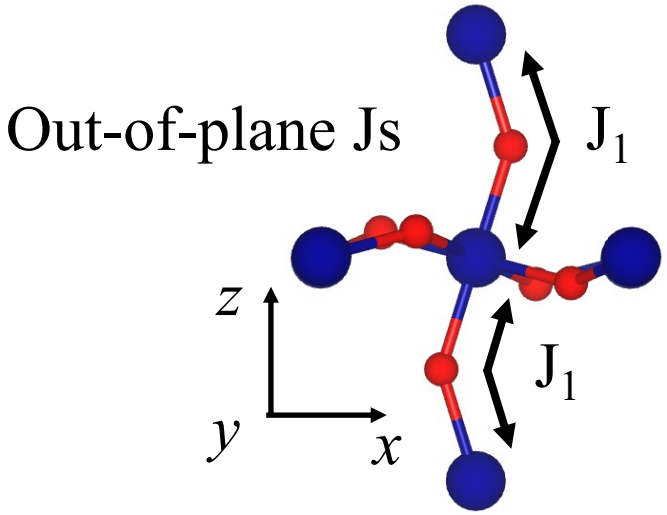

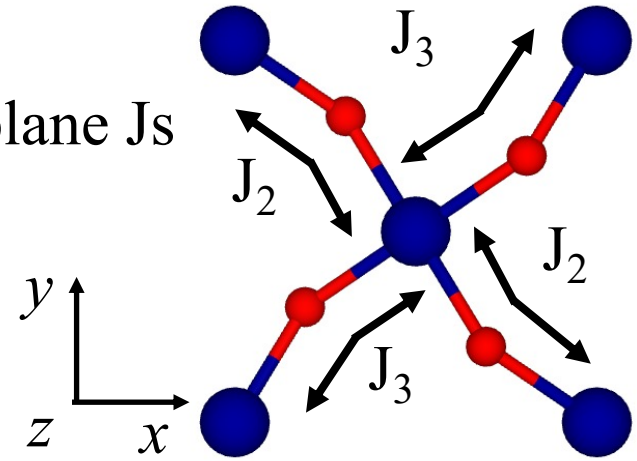

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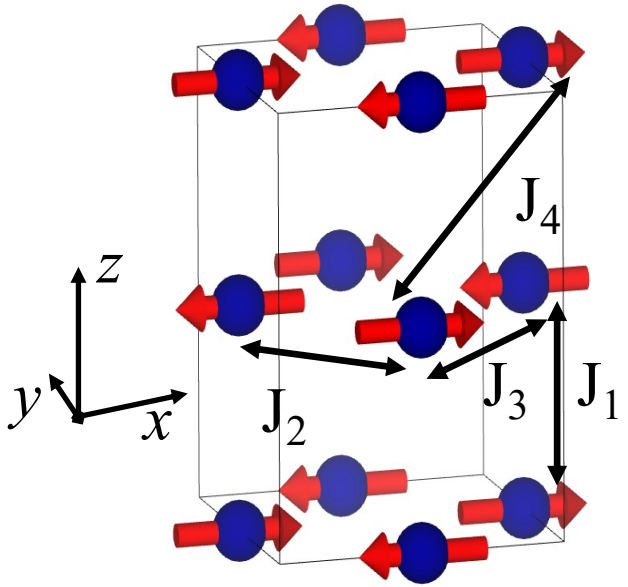
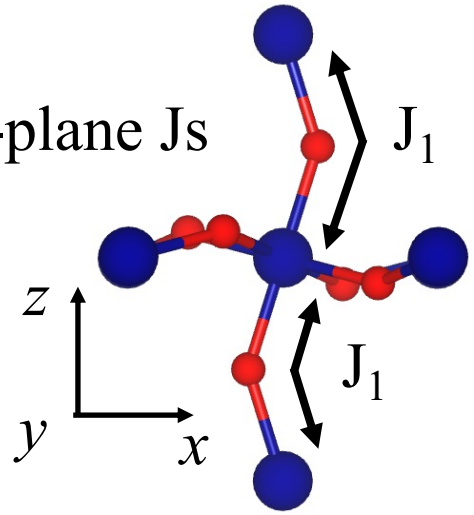


Out-of-plane Js



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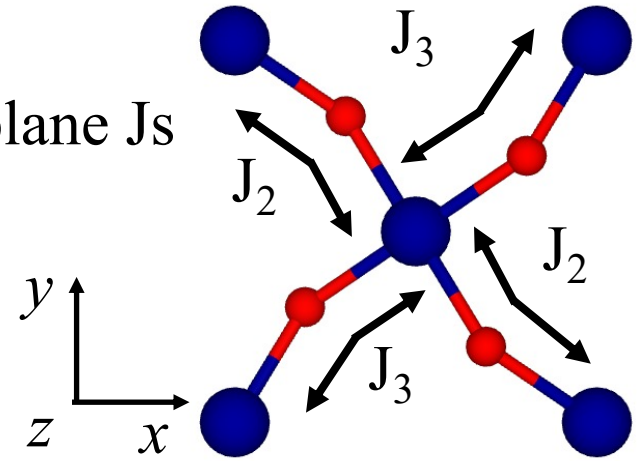

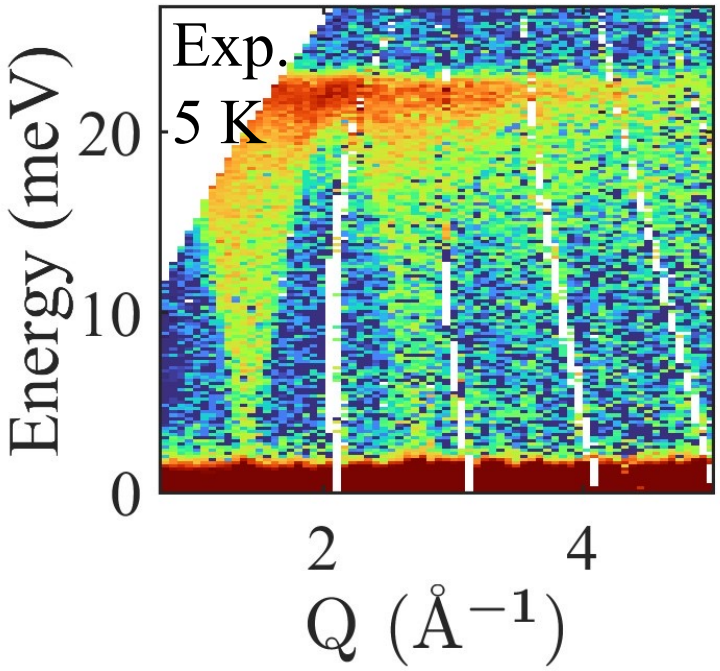

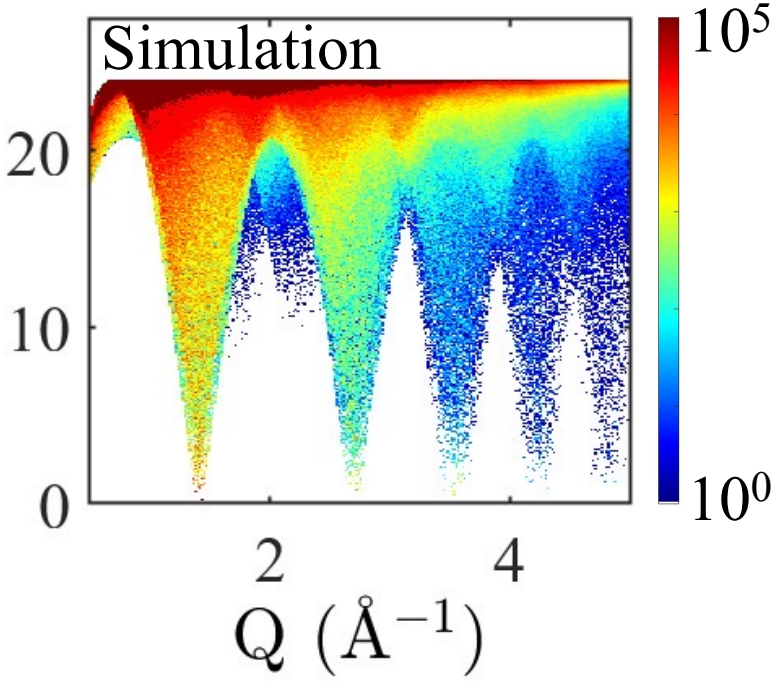

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Spin wave renormalization in $YCrO_3$

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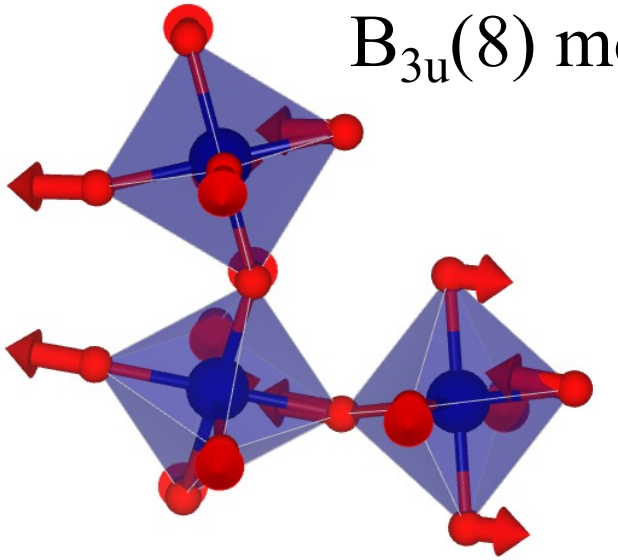
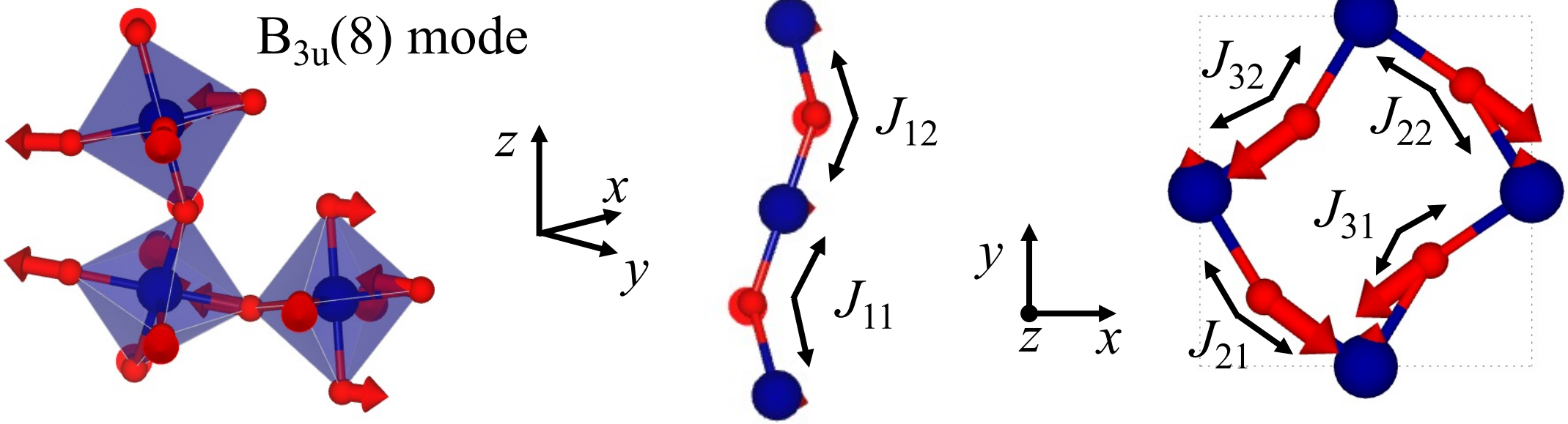


$B_{3u}(8)$ mode

Spin wave renormalization in $YCrO_3$



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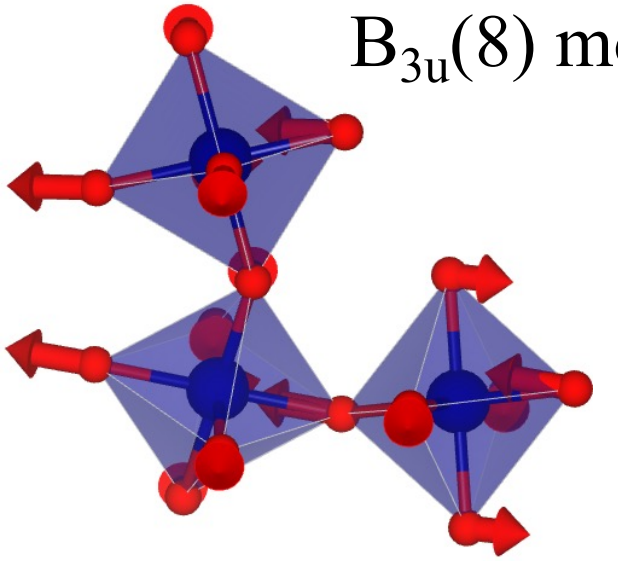

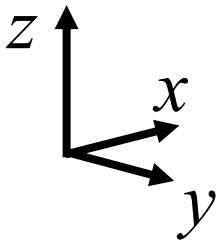

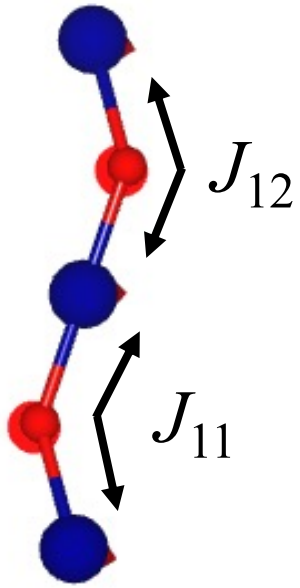

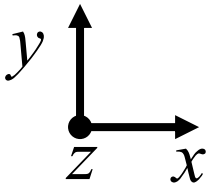

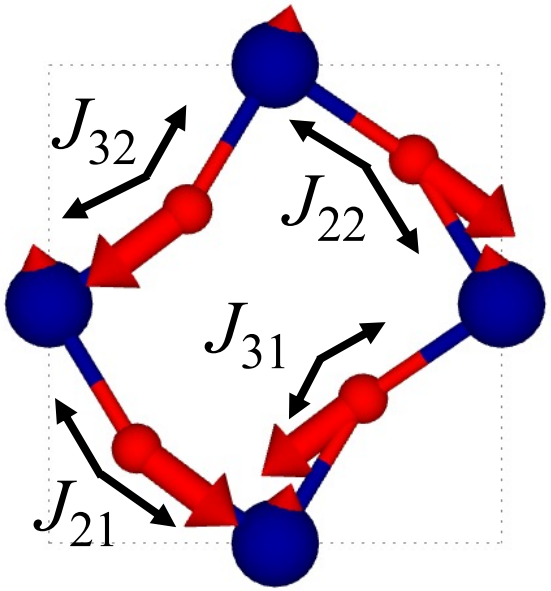

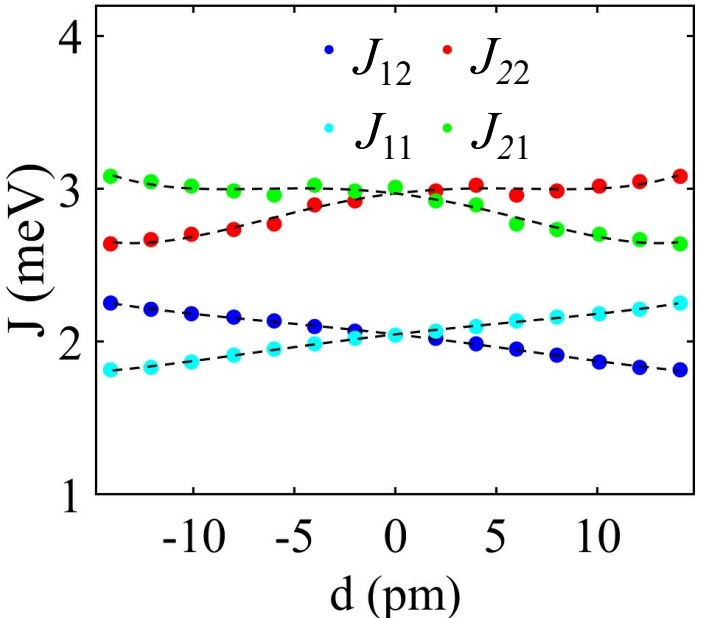

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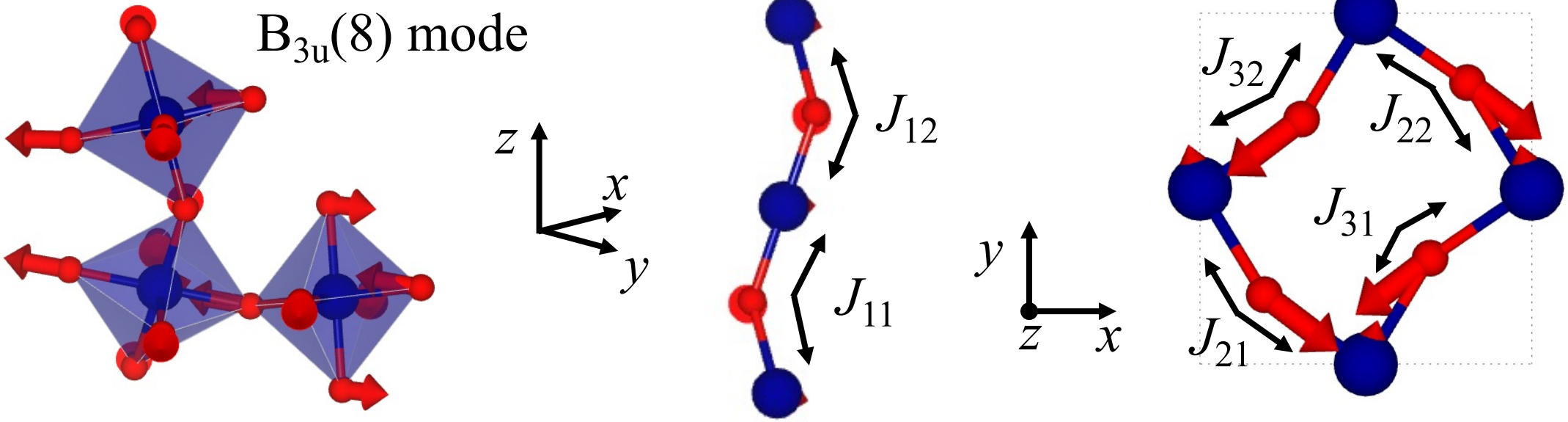

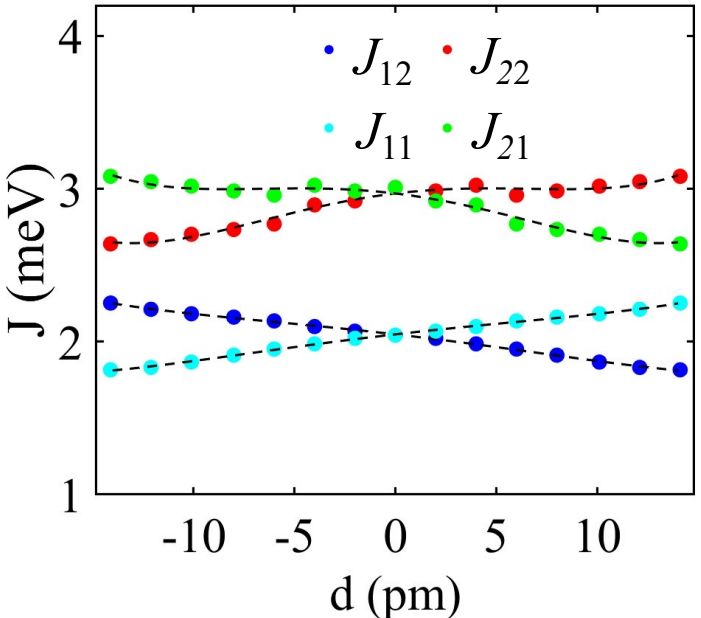

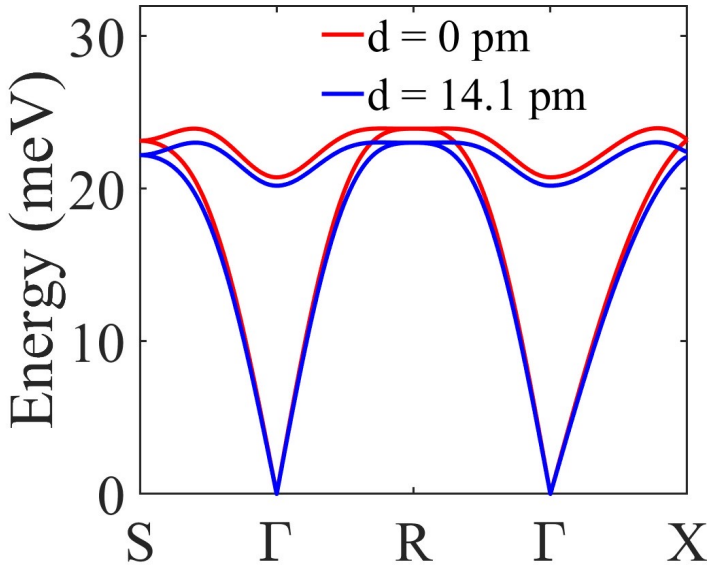

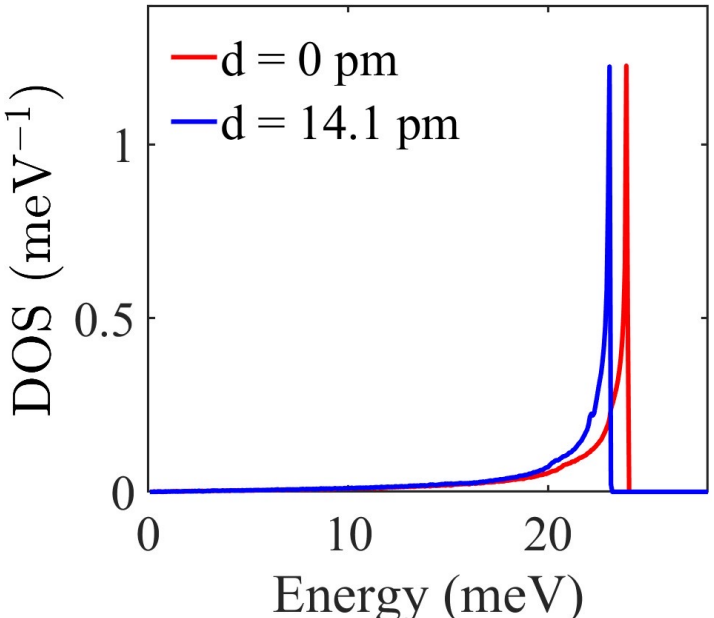

Phonon renormalization in $YCrO_3$

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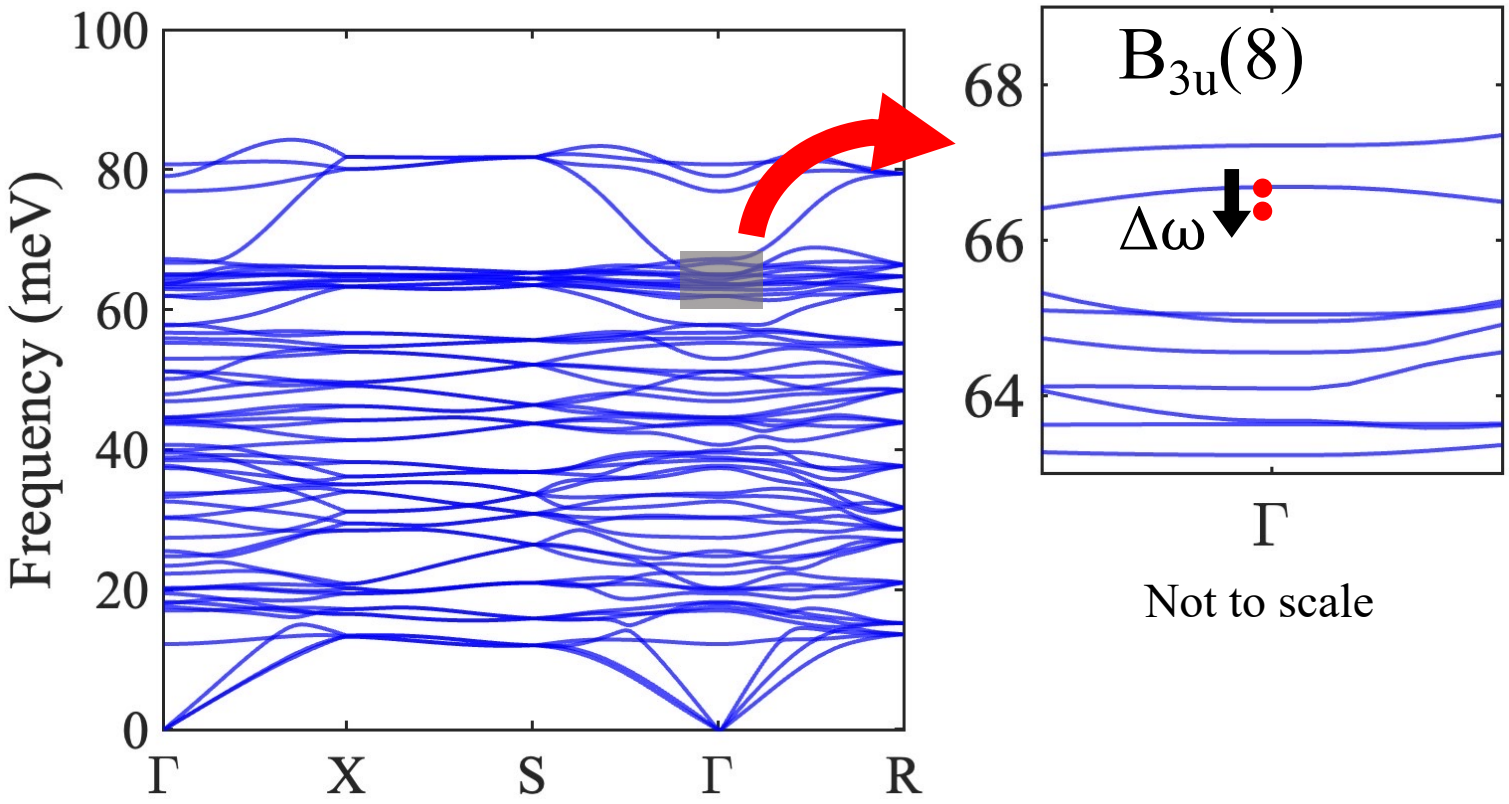

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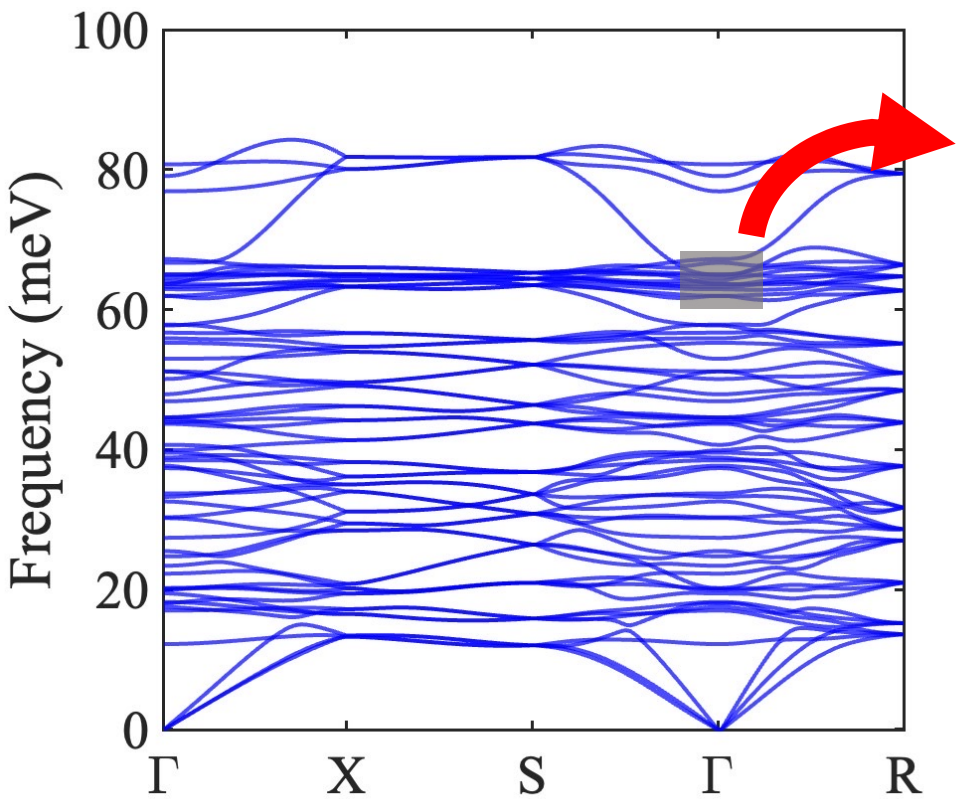
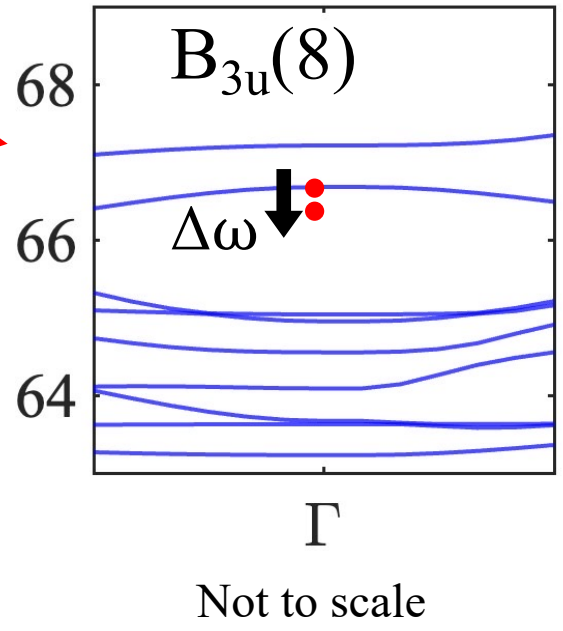
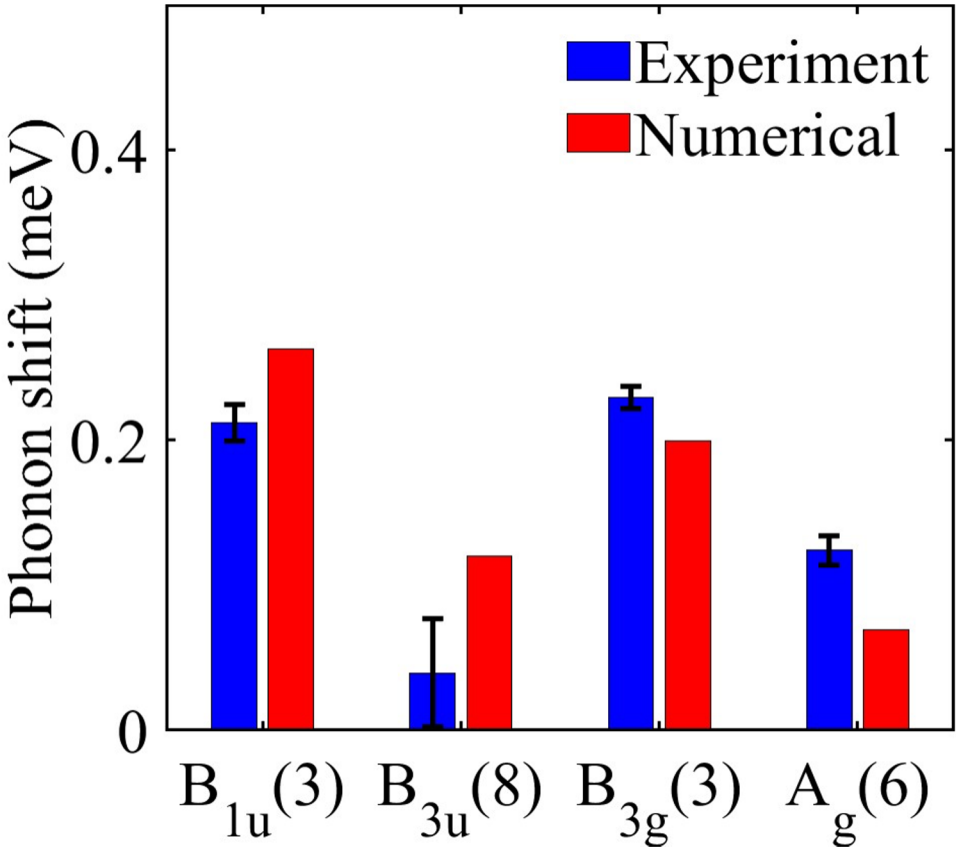




Not to scale



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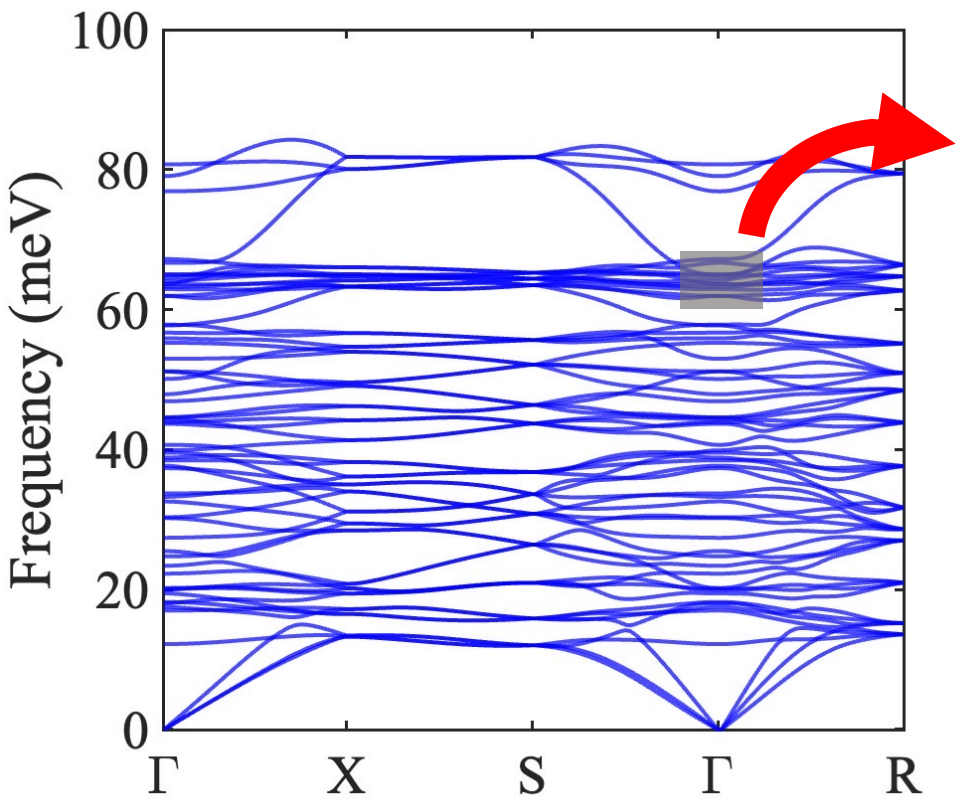

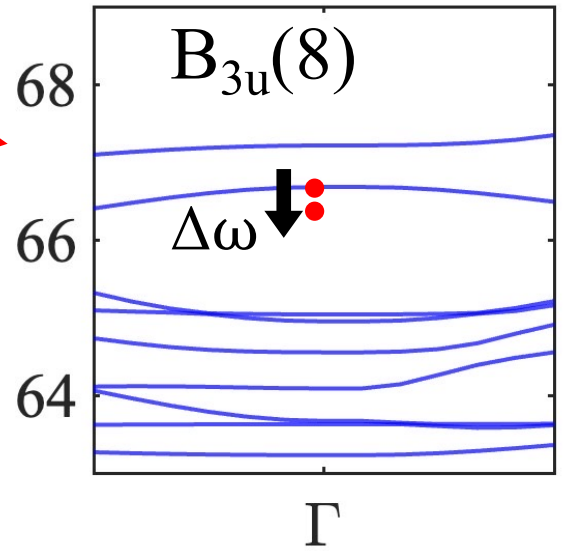

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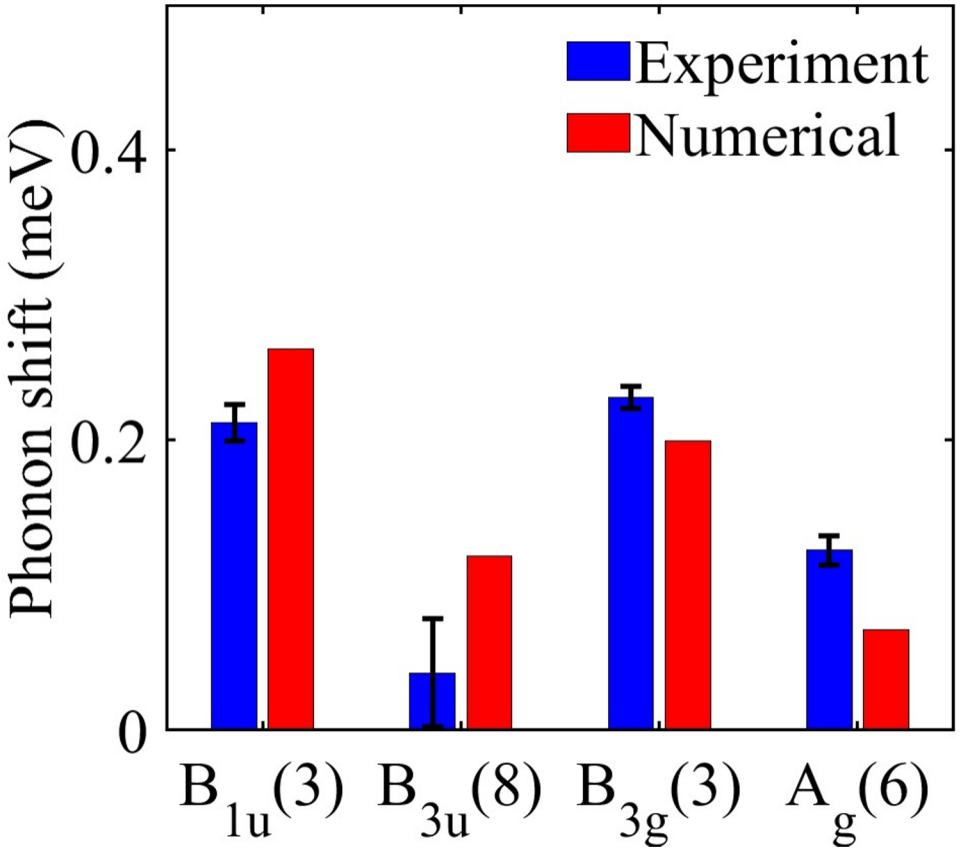

- Exchange interaction changes by 0.1 to 0.6 meV.
- Spin wave changes by 0.5 to 1.5 meV.
- Phonon changes by 0.1 to 0.25 meV.

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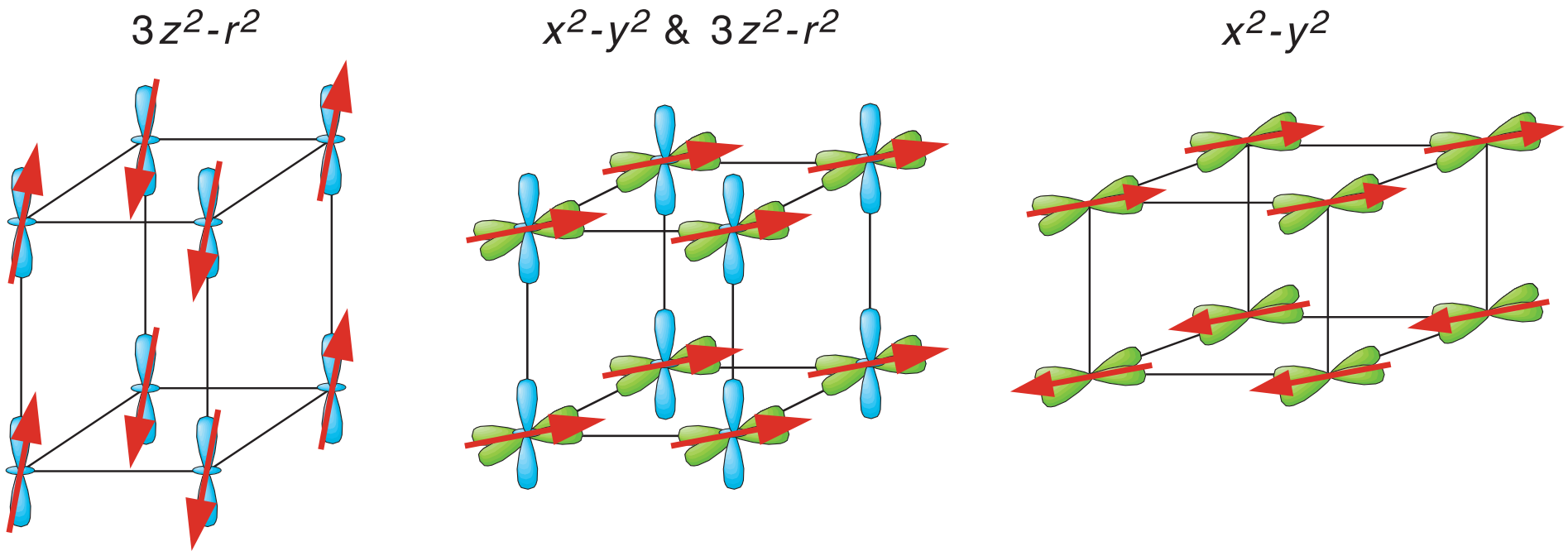
Kugel-Khomskii-type interaction

- Coupling of orbital degree of freedom with a spin exchange
  - responsible for colossal magnetoresistance, enhanced magnetoelectric response, and photoinduced high-temperature magnetism



K.I. Kugel and D. Khomskii, Sov. Phys. Usp. 25, 231 (1982)
Science **288**, 462-468 (2000)
PRL **130**, 036801 (2023)

Kugel-Khomskii-type interaction induced enhanced magnetoelectric response

Spin-Lattice and Magnetoelectric Couplings Enhanced by Orbital Degrees of Freedom in Polar Multiferroic Semiconductors

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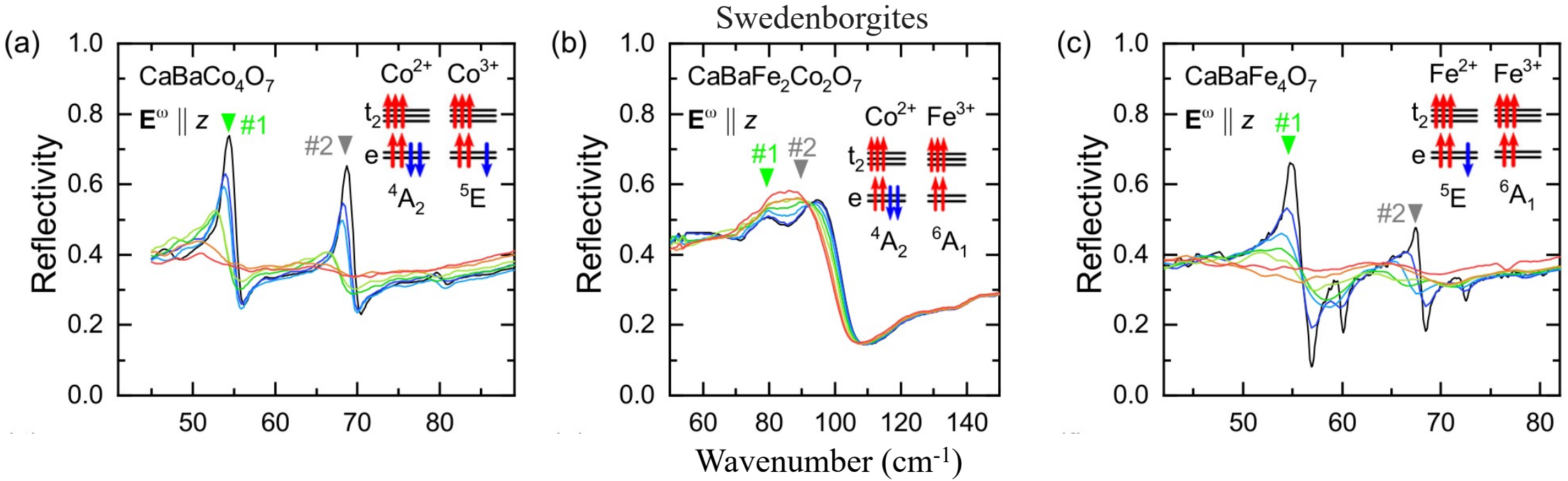

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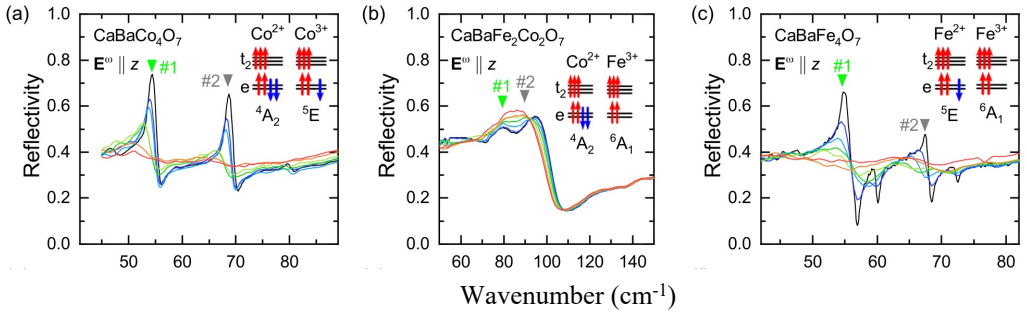
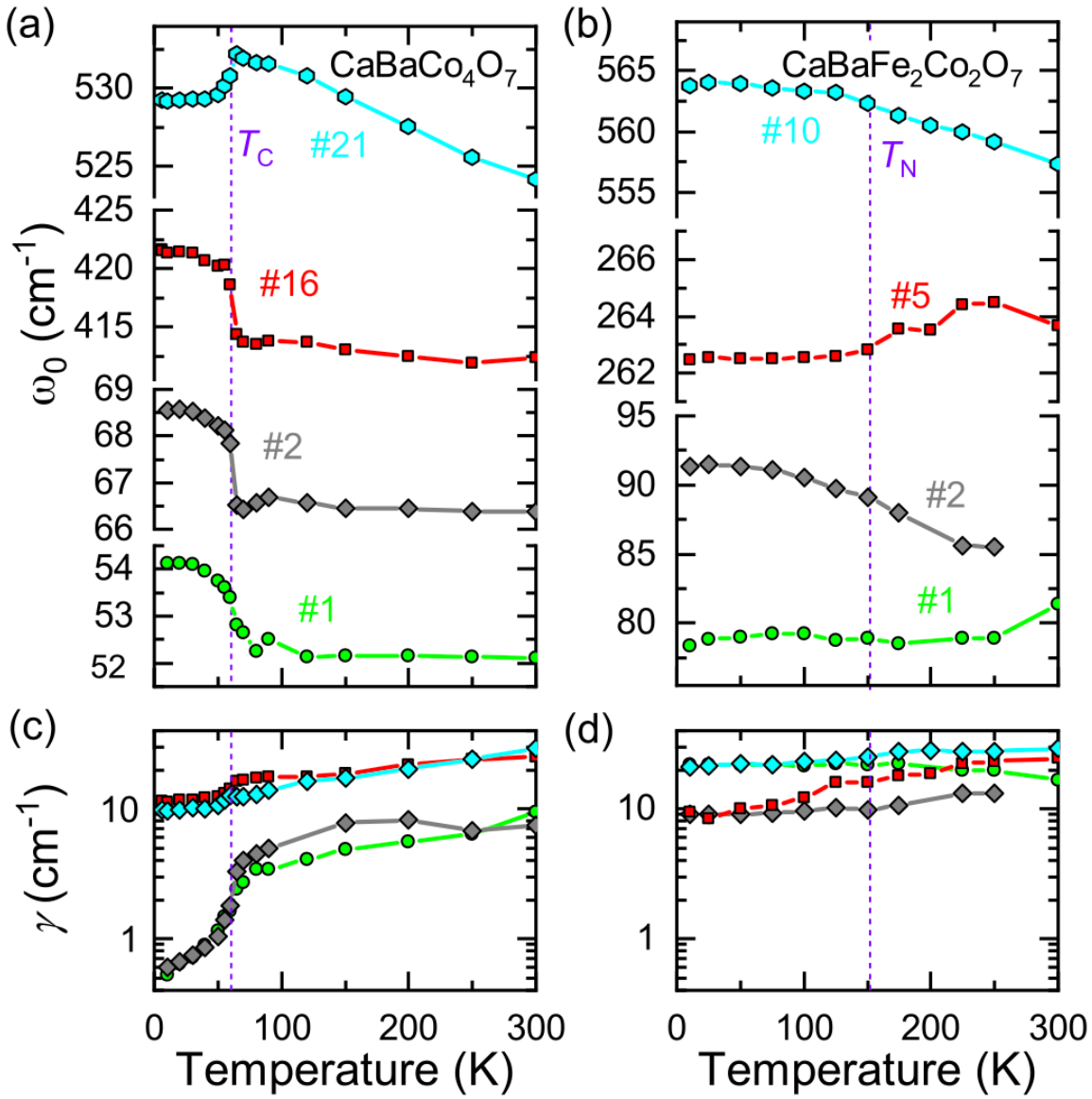
Kugel-Khomskii-type interaction: Effect on phonon lifetimes





PRL **130**, 036801 (2023)

Evidence of Kugel-Khomskii-type interaction in $CrVO_4$ at $4T_N$

A.P. Roy *et al.* **"Evidence of strong orbital-selective spin-orbital-phonon coupling in $CrVO_4$."** PRL 132, 026701, 2024.

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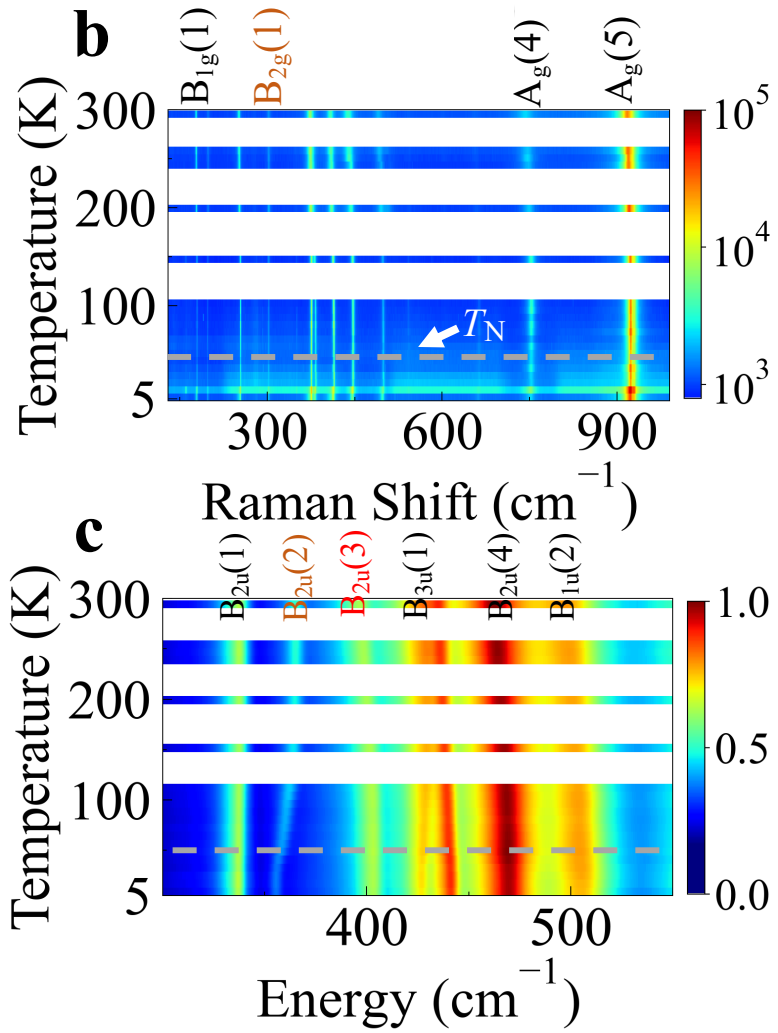

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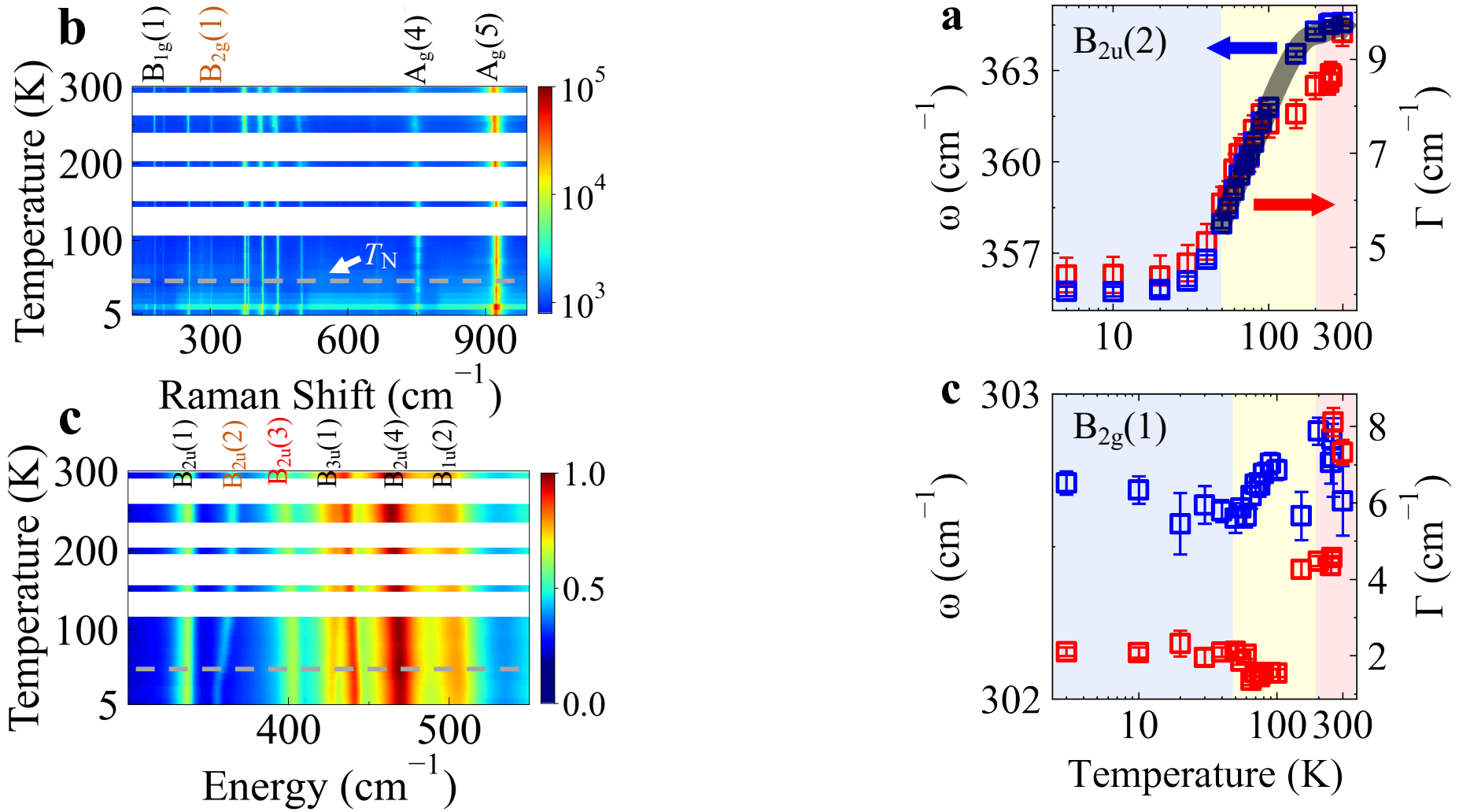

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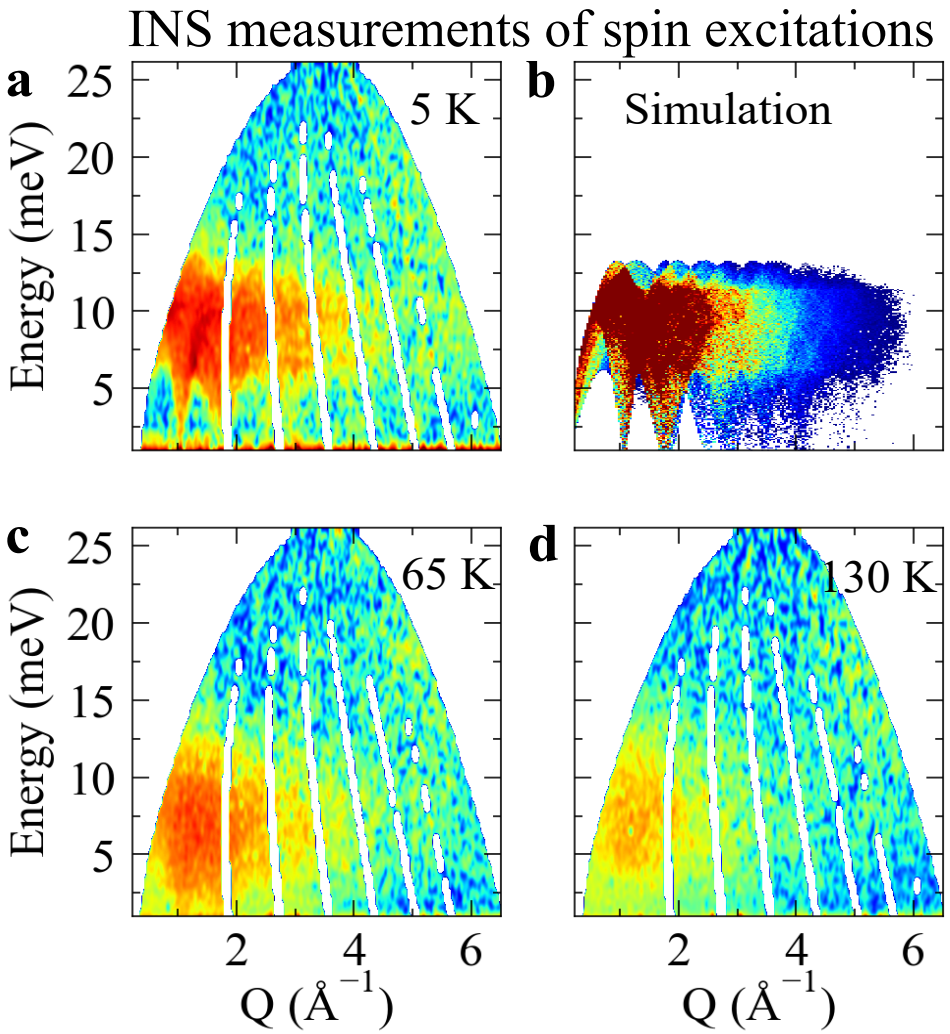
Evidence of Kugel-Khomskii-type interaction in $CrVO_4$ at $4T_N$: But Why?



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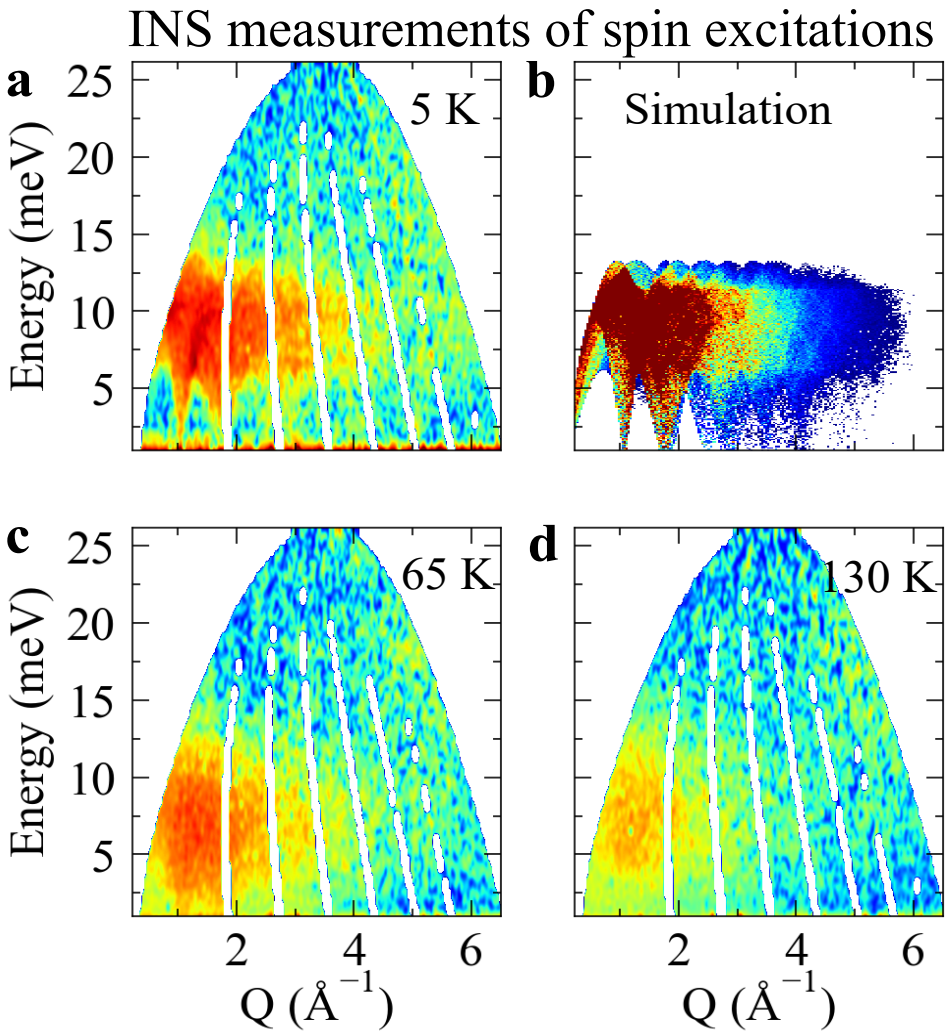

Short-range spin correlations could not be the reason for anomalies seen at $4T_N$ as they start right above T_N

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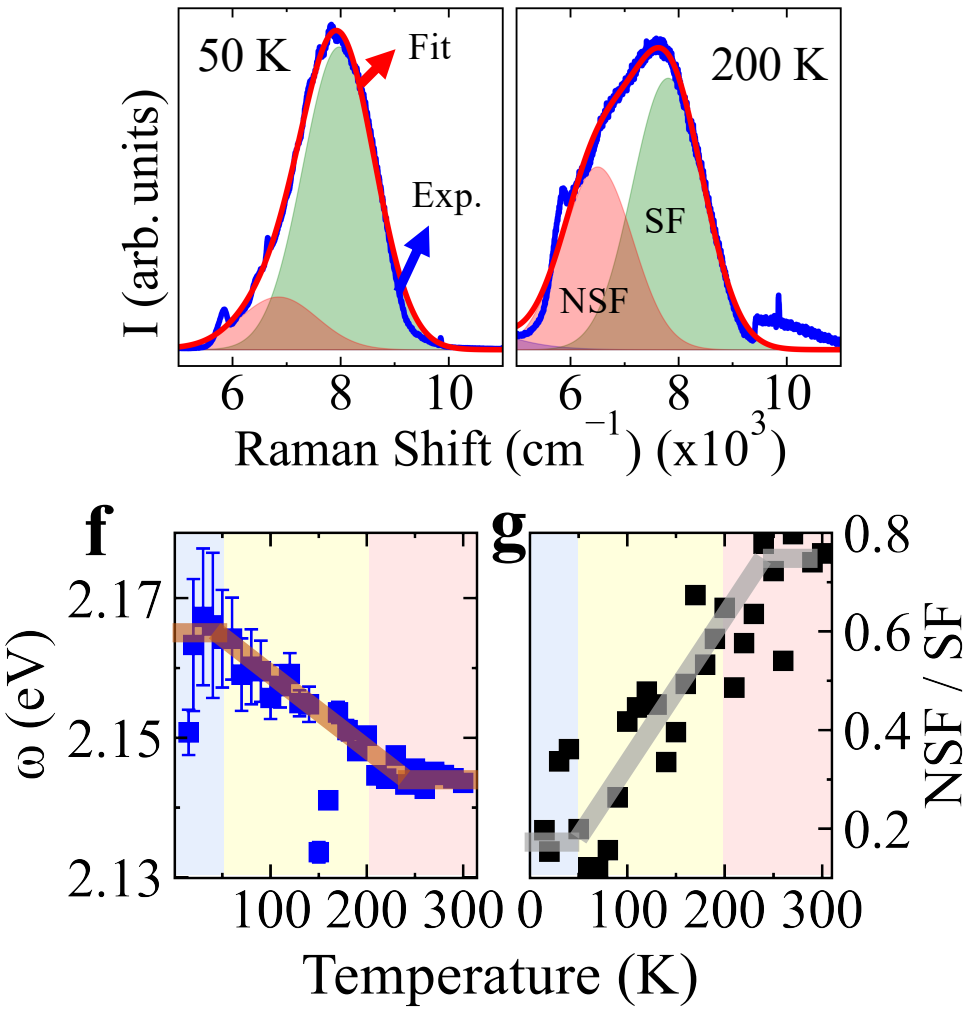

But Short-range spin correlations initiate orbital polarization (facilitate exchange interactions, particularly d_{x2-y2} and d_{3z2-1}), and the continuous orbital fluctuations decrease

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Evidence of Kugel-Khomskii-type interaction in $CrVO_4$ at $4T_N$: But Why?

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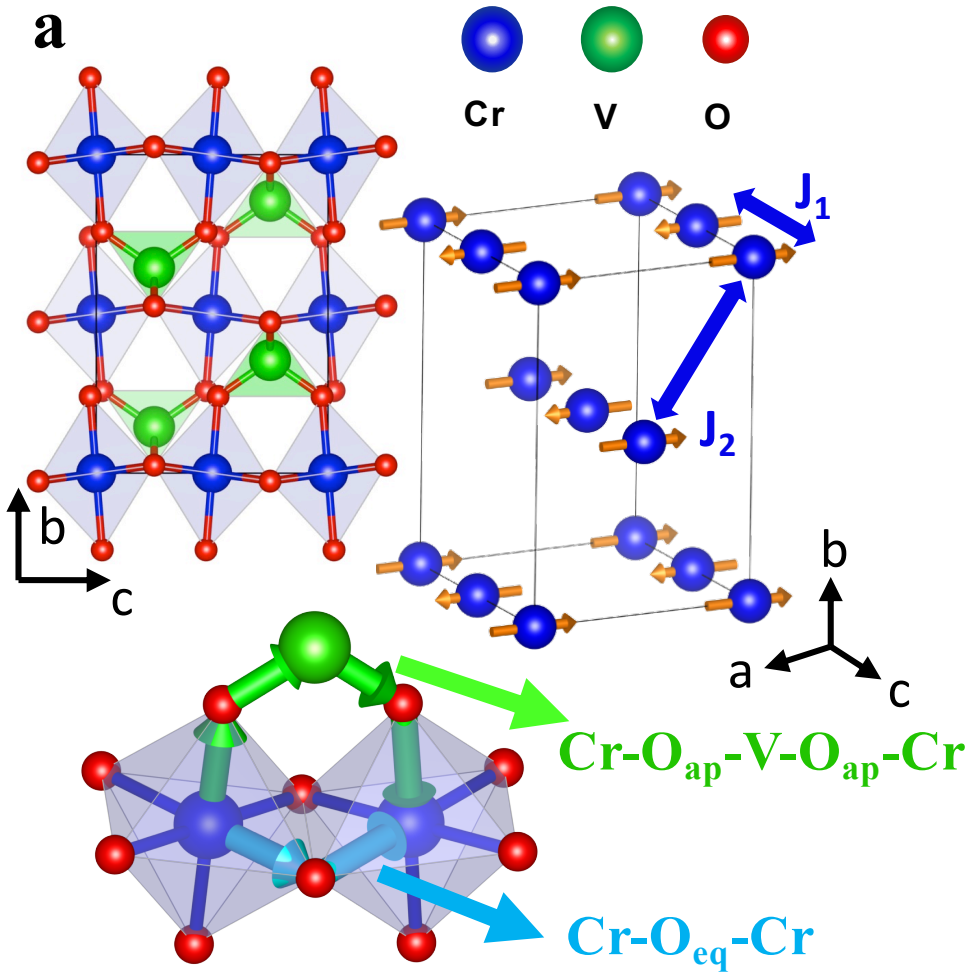

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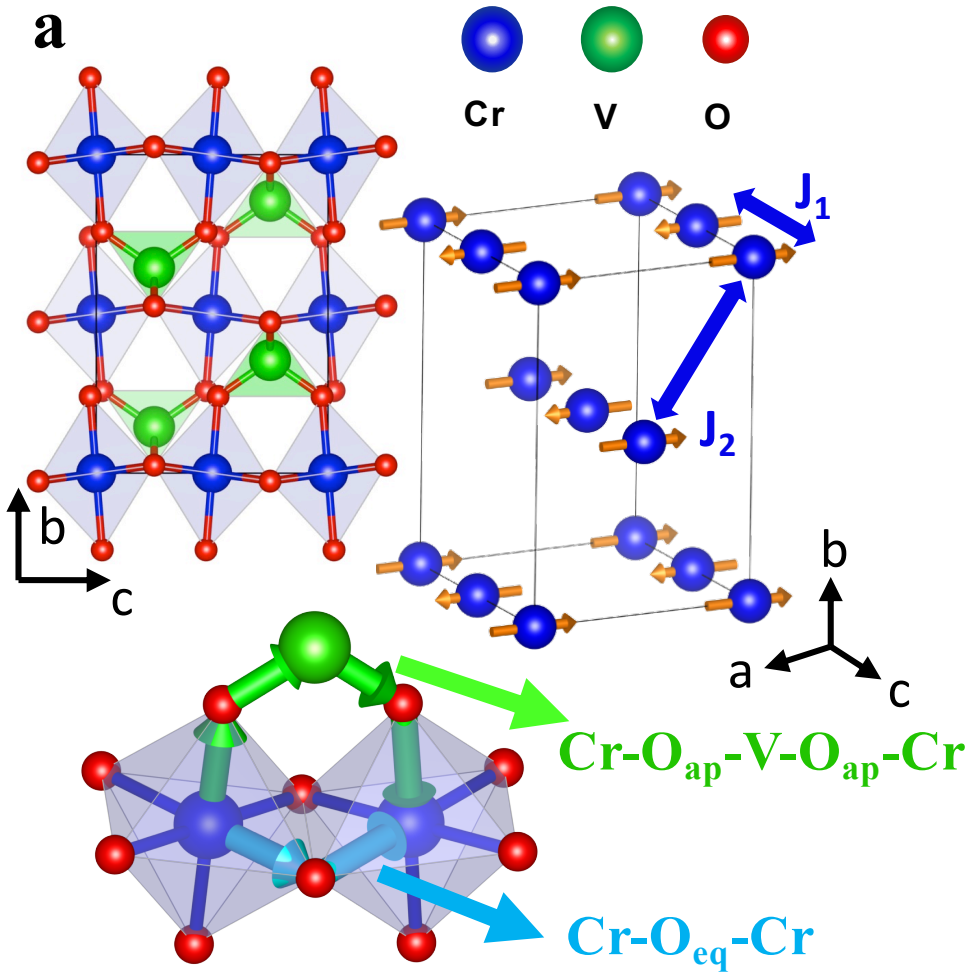

$$J_{ij} = -\frac{1}{2\pi}\int_{-\infty}^{E_F} d\epsilon \sum_{mm'm''m'''} \mathrm{Im}(\Delta_i^{mm'} G_{ij,\downarrow}^{mm'} \Delta_j^{m''m'''} G_{ji,\uparrow}^{m''m'''})$$

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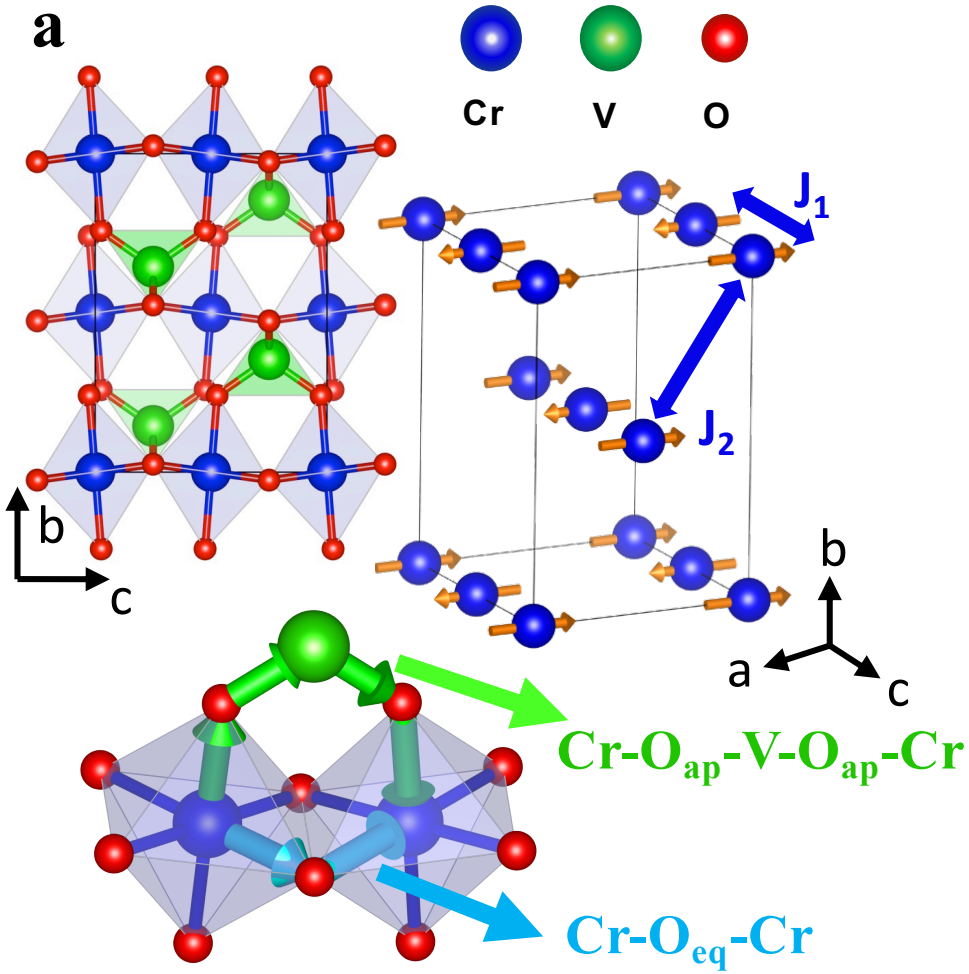

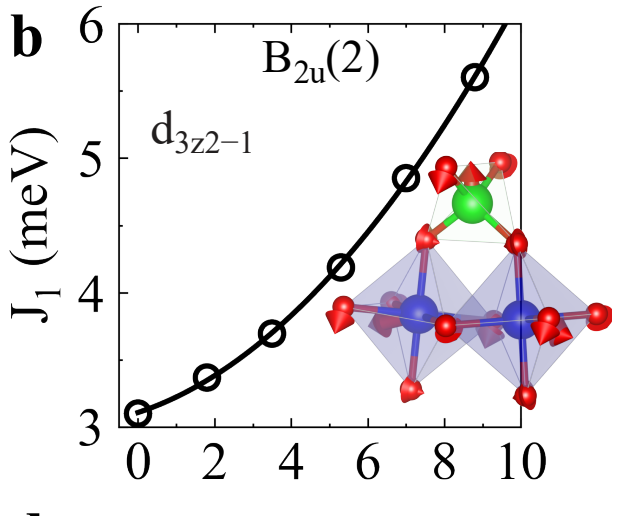

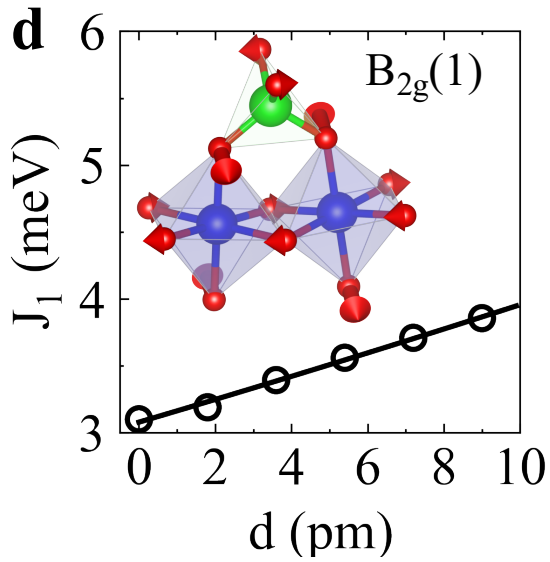

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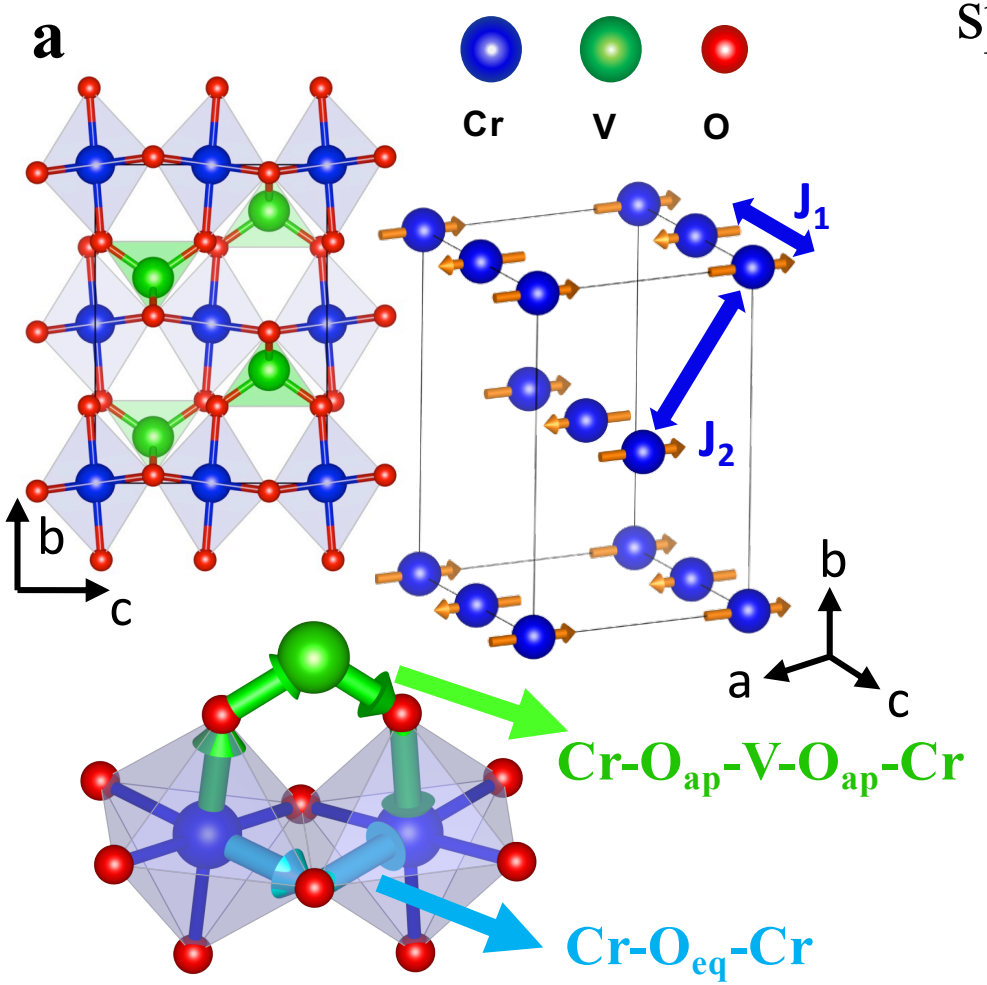
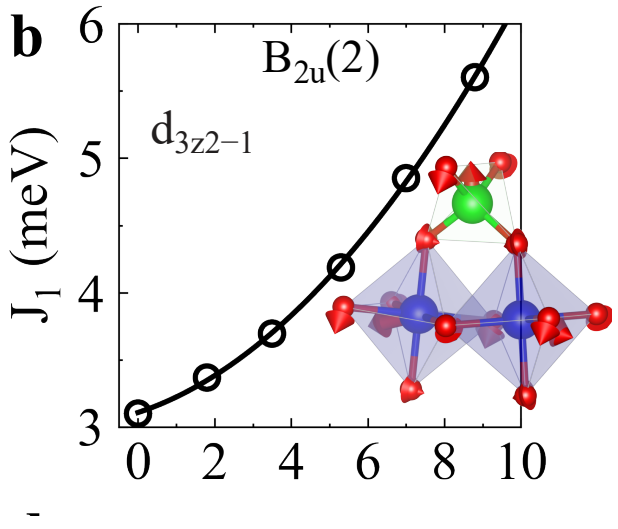
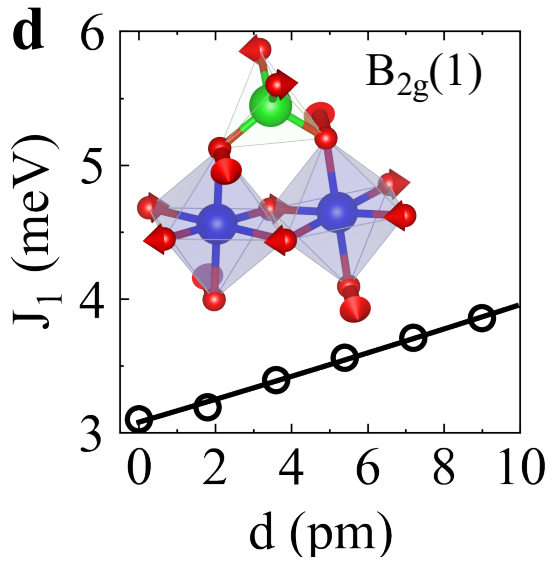


Due to extremely strong orbital-dependent spin-phonon coupling ($d^2J/du^2 \sim 432$ meV/A^2)





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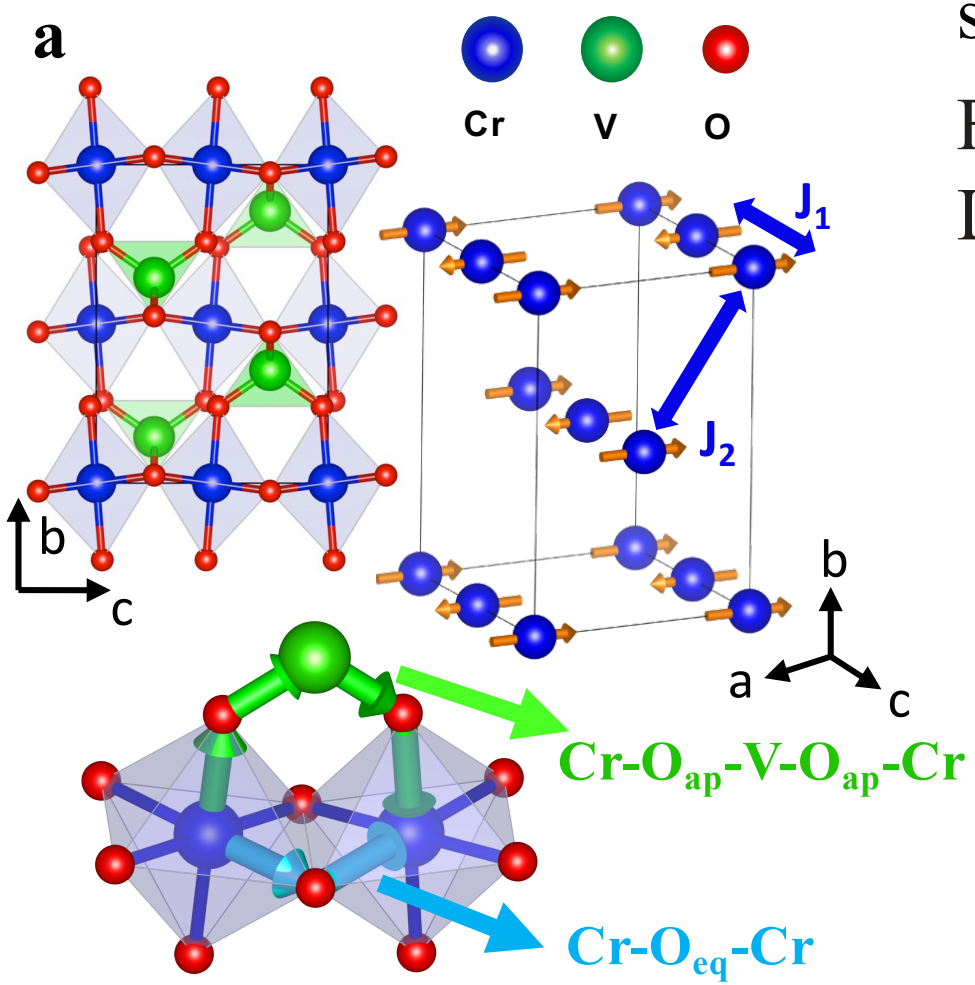

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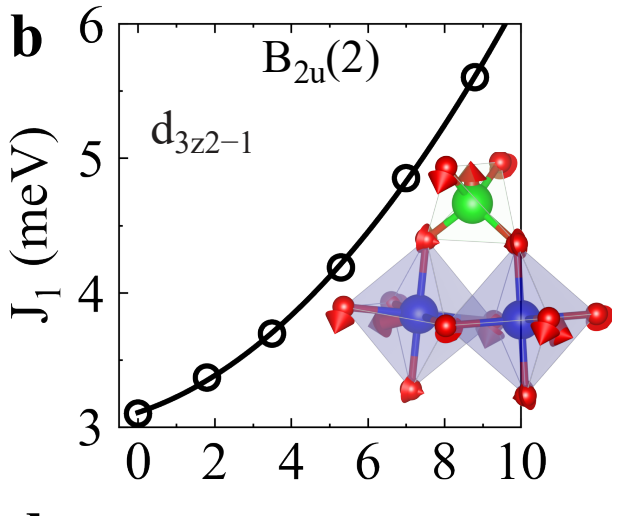
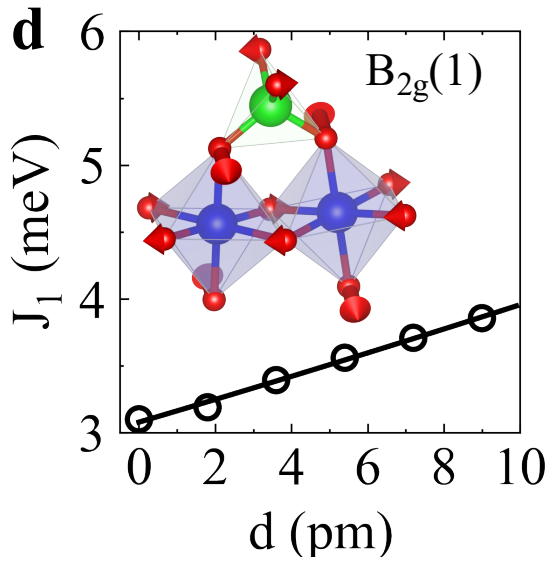
For $PrMnO_3$, $NdMnO_3$, $SmMnO_3$ and $LaMnO_3$ ranges between 175 to 220 meV/A^2





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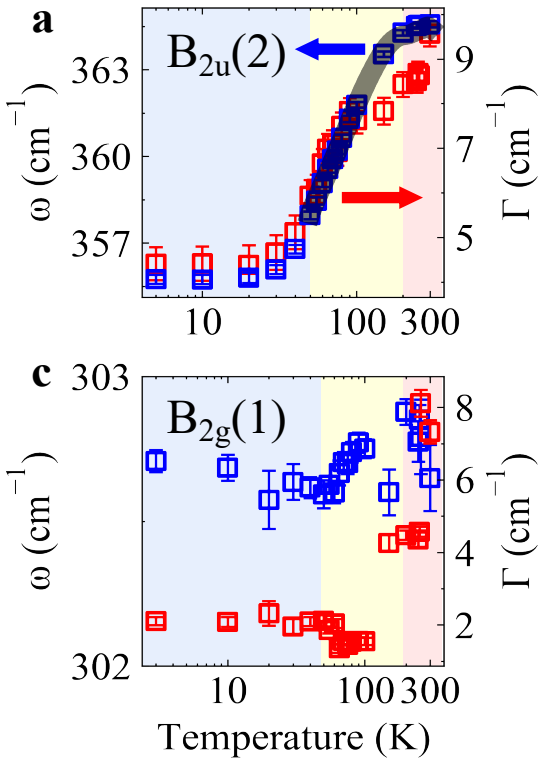
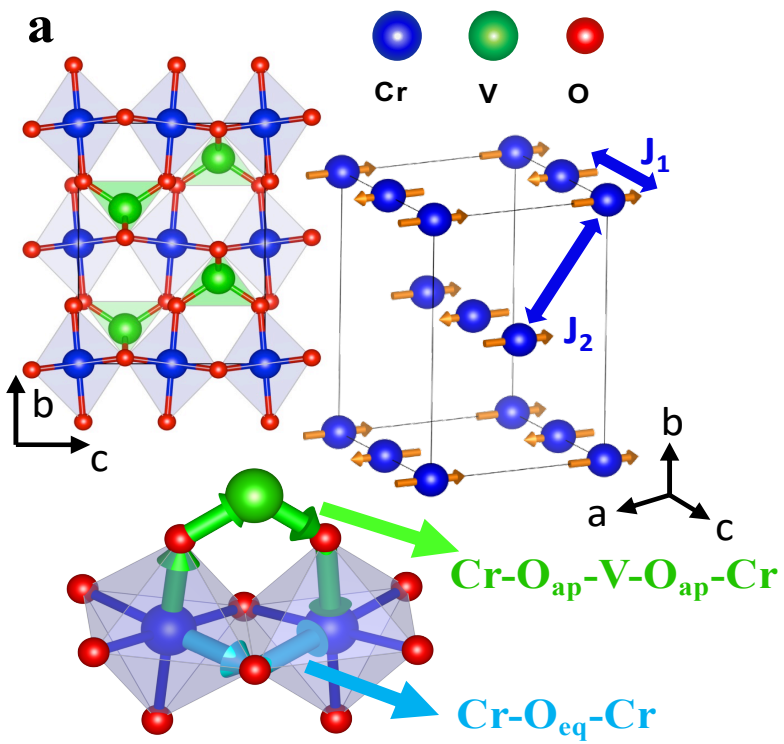
In a nutshell

- Extremely strong Orbital-Selective Spin-Orbital-Phonon Coupling (two exchange pathways) – controls anomaly in phonon energy
- Continuous orbital fluctuations of the Cr^{+3} orbitals are suppressed at $4T_N$ on cooling – controls anomaly in phonon linewidth





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In a nutshell

- Transient evolution of spin-phonon coupling

Jayakrishnan SS and D. Bansal. **"Coherent phonon excitation induced evolution of spin dynamics and spin-phonon coupling in $YCrO_3$."** PRB, 112, 214419, 2025.
Jayakrishnan SS and D. Bansal. **"Effect of spin-phonon coupling on phonons and magnons in the antiferromagnet NiO."** PRB, Vol. 111, 104306, 2025.

In a nutshell

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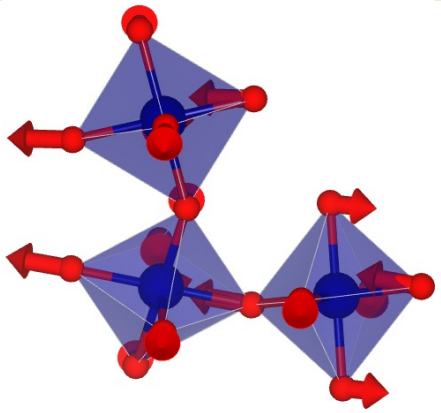

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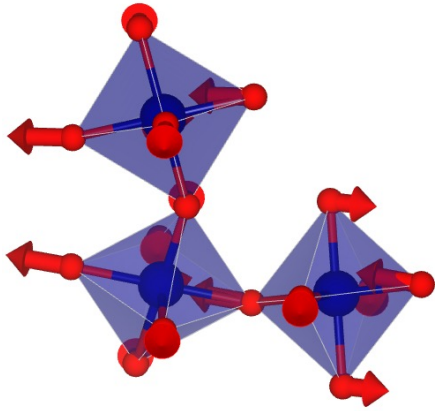

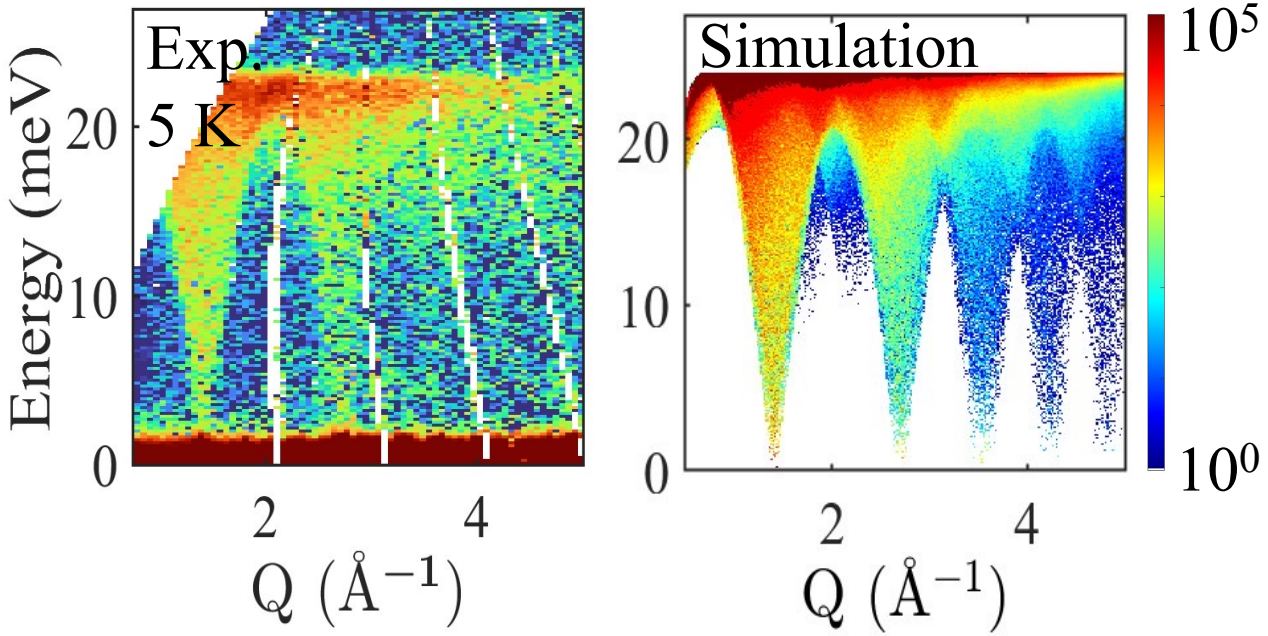

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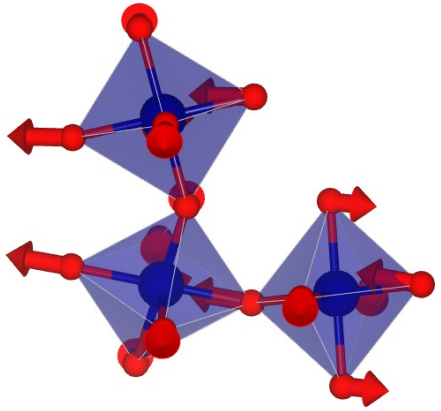

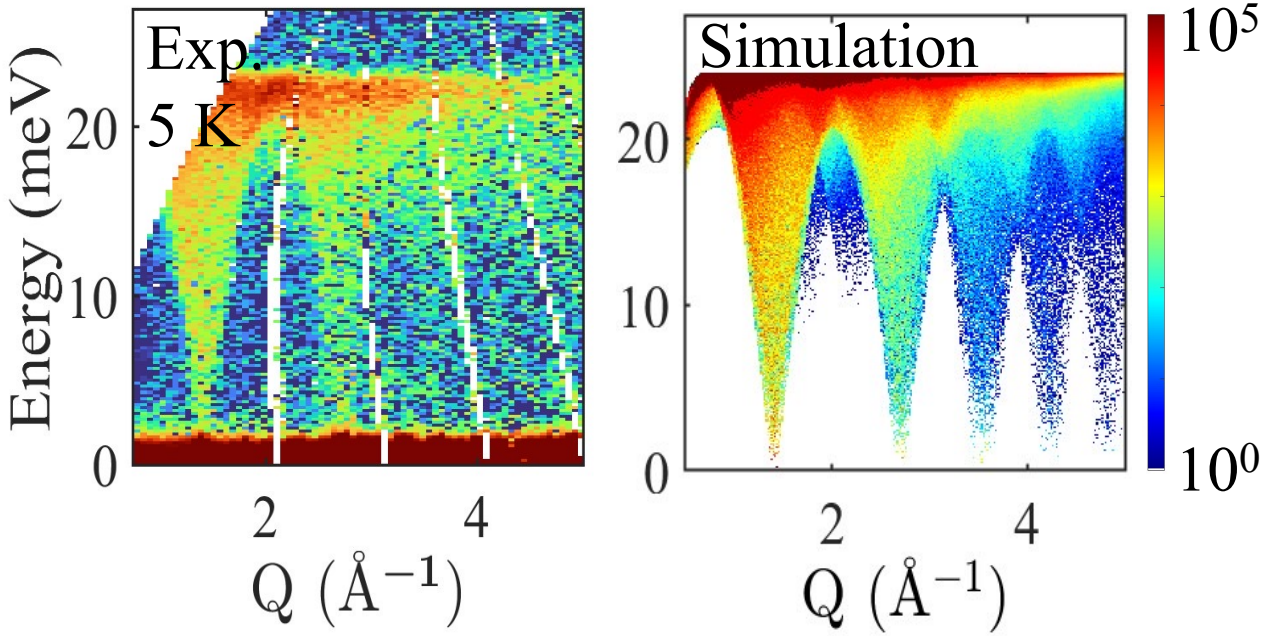

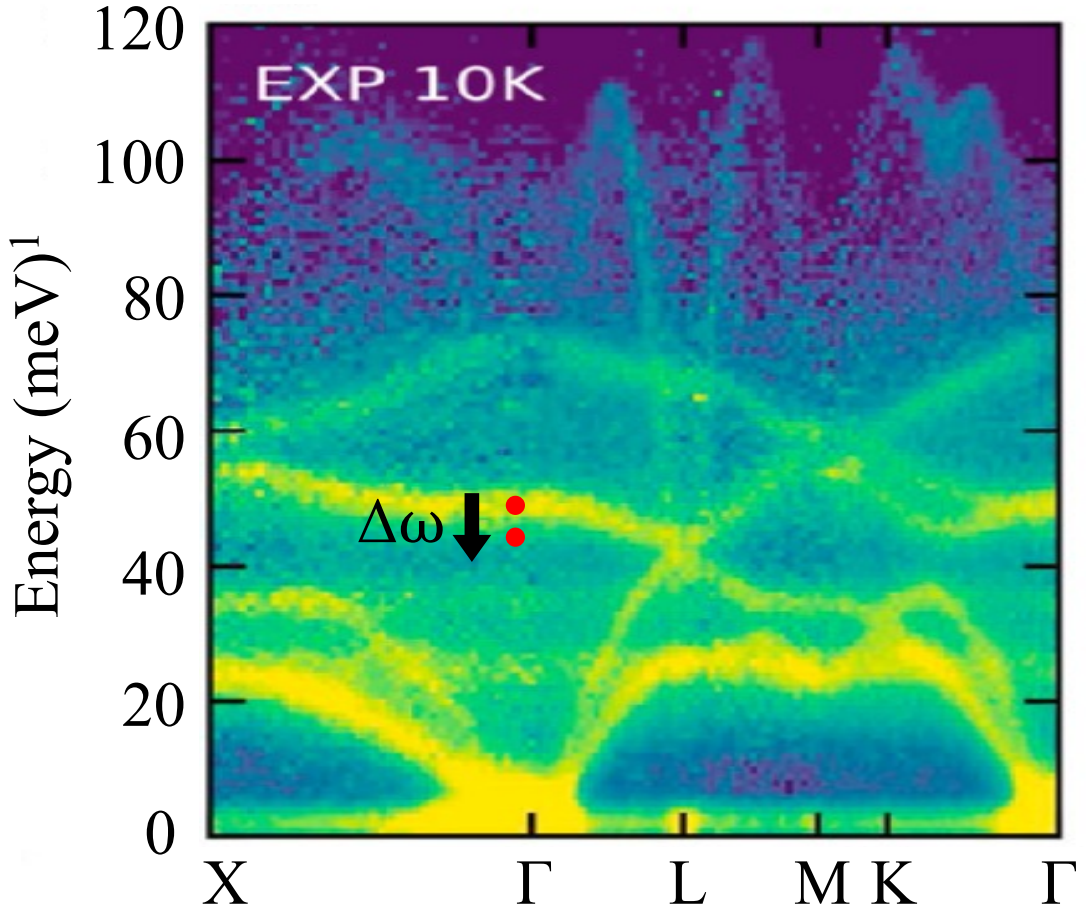

Sun, *Materials Today Physics* 35, 101094 (2023)

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$$H_L = \frac{1}{2}\sum_{k,l>k} \phi_{kl}^{\mu\nu} u_k^\mu u_l^\nu,$$

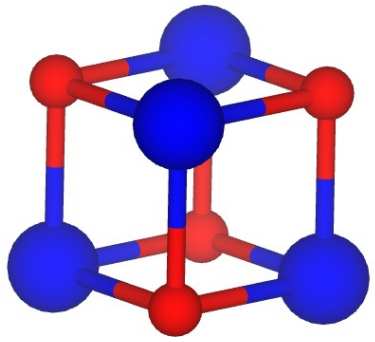
$$H_{LS} = \frac{1}{2}\sum_{k,l>k} \phi_{kl}^{\mu\nu} u_k^\mu u_l^\nu + \frac{1}{2}\sum_{k,k>l,i} \frac{\partial \phi_{kl}^{\mu\nu}}{\partial m_i^\alpha} m_i^\alpha u_k^\mu u_l^\nu + \frac{1}{4}\sum_{\substack{k,l>k \\ i,j>i}} \frac{\partial^2 \phi_{kl}^{\mu\nu}}{\partial m_i^\alpha \partial m_j^\beta} m_i^\alpha m_j^\beta u_k^\mu u_l^\nu,$$

$$H_{SL} = \sum_{i,j>i} J_{ij}^{\alpha\beta} m_i^\alpha m_j^\beta + \sum_{i,j>i,k} \frac{\partial J_{ij}^{\alpha\beta}}{\partial u_k^\mu} u_k^\mu m_i^\alpha m_j^\beta + \frac{1}{2}\sum_{\substack{k,l>k \\ i,j>i}} \frac{\partial^2 J_{ij}^{\alpha\beta}}{\partial u_k^\mu \partial u_l^\nu} u_k^\mu u_l^\nu m_i^\alpha m_j^\beta.$$

$$H_S = \sum_{i,j>i} \mathcal{J}_{ij}^{\alpha\beta} m_i^\alpha m_j^\beta.$$

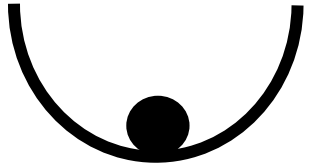
$$H_{SLD} = \sum_{i,j>i} J_{ij}^{\alpha\beta} m_i^\alpha m_j^\beta + \sum_{i,j>i,k} J_f u_k^\mu m_i^\alpha m_j^\beta + \frac{1}{2}\sum_{\substack{k,l>k \\ i,j>i}} J_s u_k^\mu u_l^\nu m_i^\alpha m_j^\beta + \frac{1}{2}\sum_{k,l>k} \phi_{kl}^{\mu\nu} u_k^\mu u_l^\nu.$$

Methodology



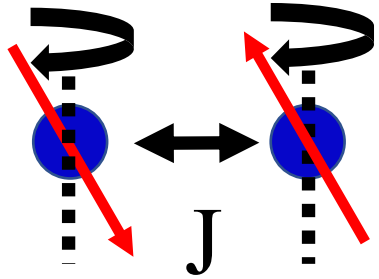
Crystal structure

DFT



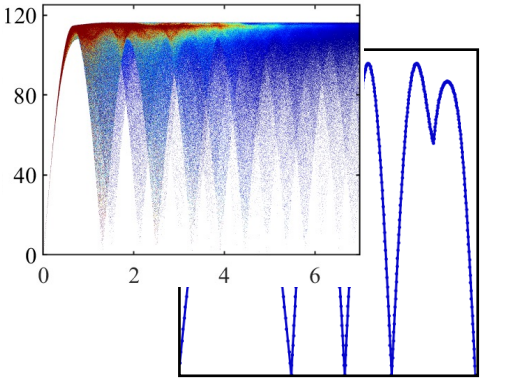
Ground state energy and forces

Wannier projection
Green's function

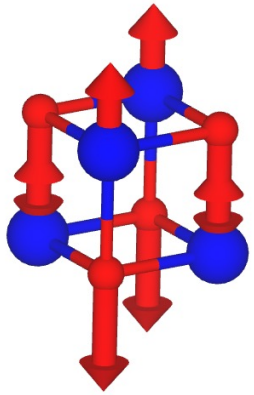


Exchange interactions

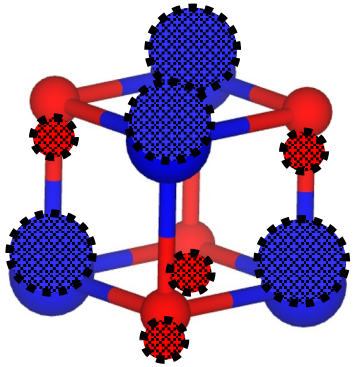
LSWT



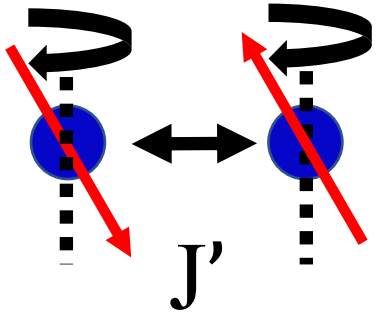
Spin waves



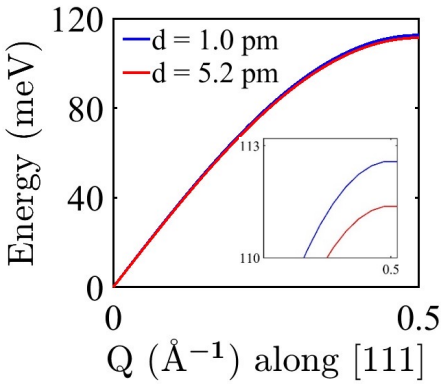
Phonon eigen vectors



Perturbed system

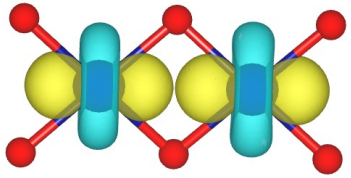


Renormalized exchange interactions

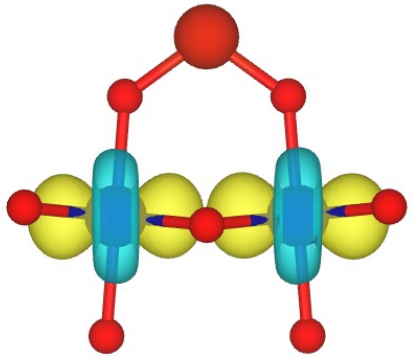


Renormalized spin/phonon waves

Top view b

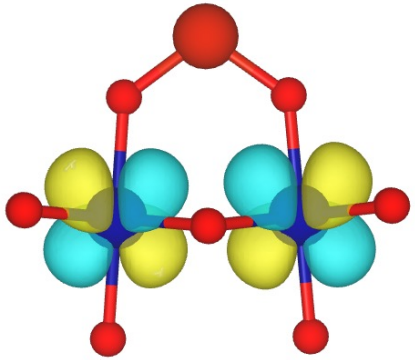


Side view a



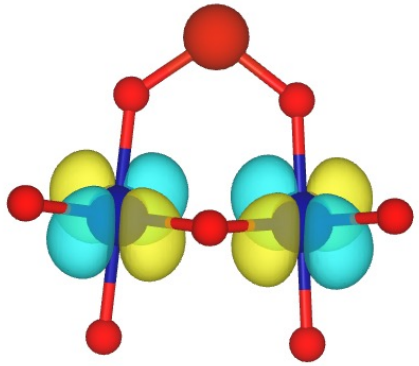
d_{z^2}

Side view a



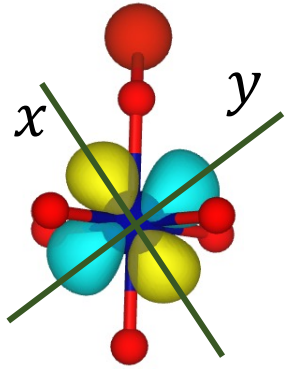
d_{xz}

Side view a



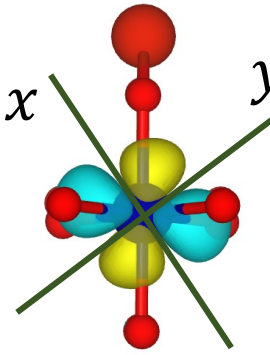
d_{yz}

Side view c



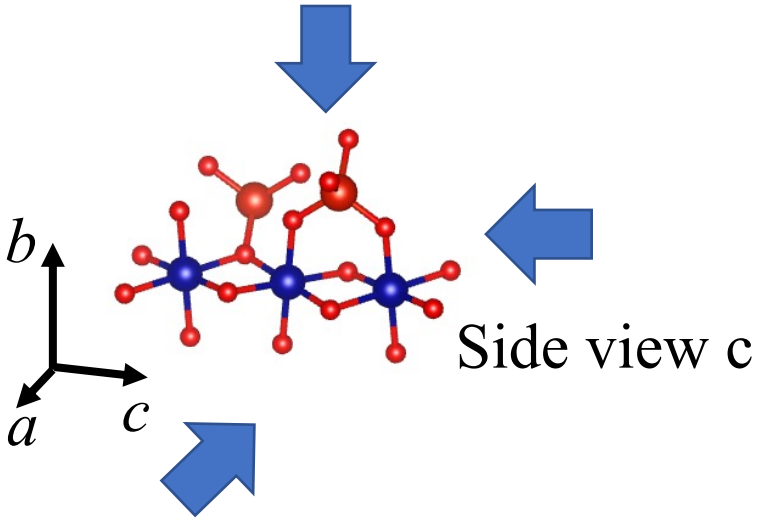
$d_{x^2-y^2}$

Side view c



d_{xy}

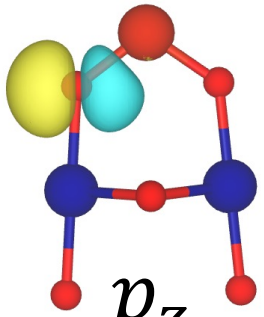
Top view b



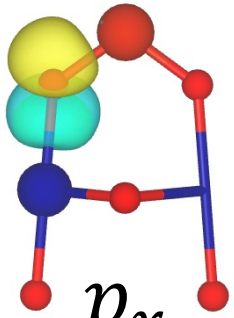
Side view c

Side view a

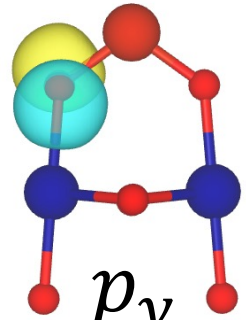
Apical O
(side view a)



p_z

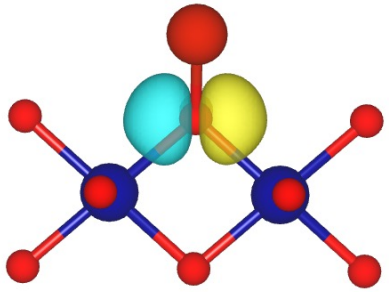


p_x

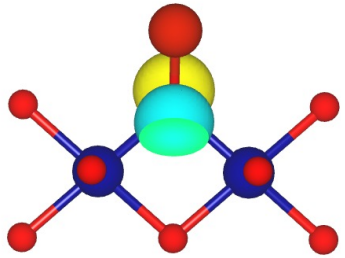


p_y

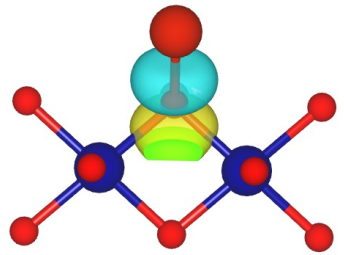
Equatorial O
(top view b)



p_z



p_x



p_y