Magnetoelectricity in polar antiferromagnets

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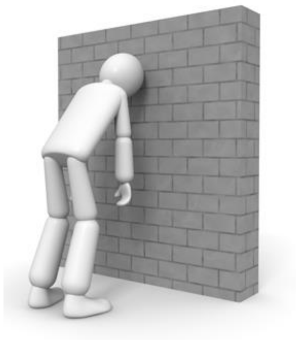
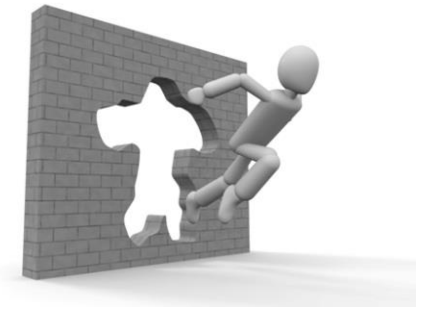
07-11-2018, IT4Innovations, Ostrava, Czech Republic

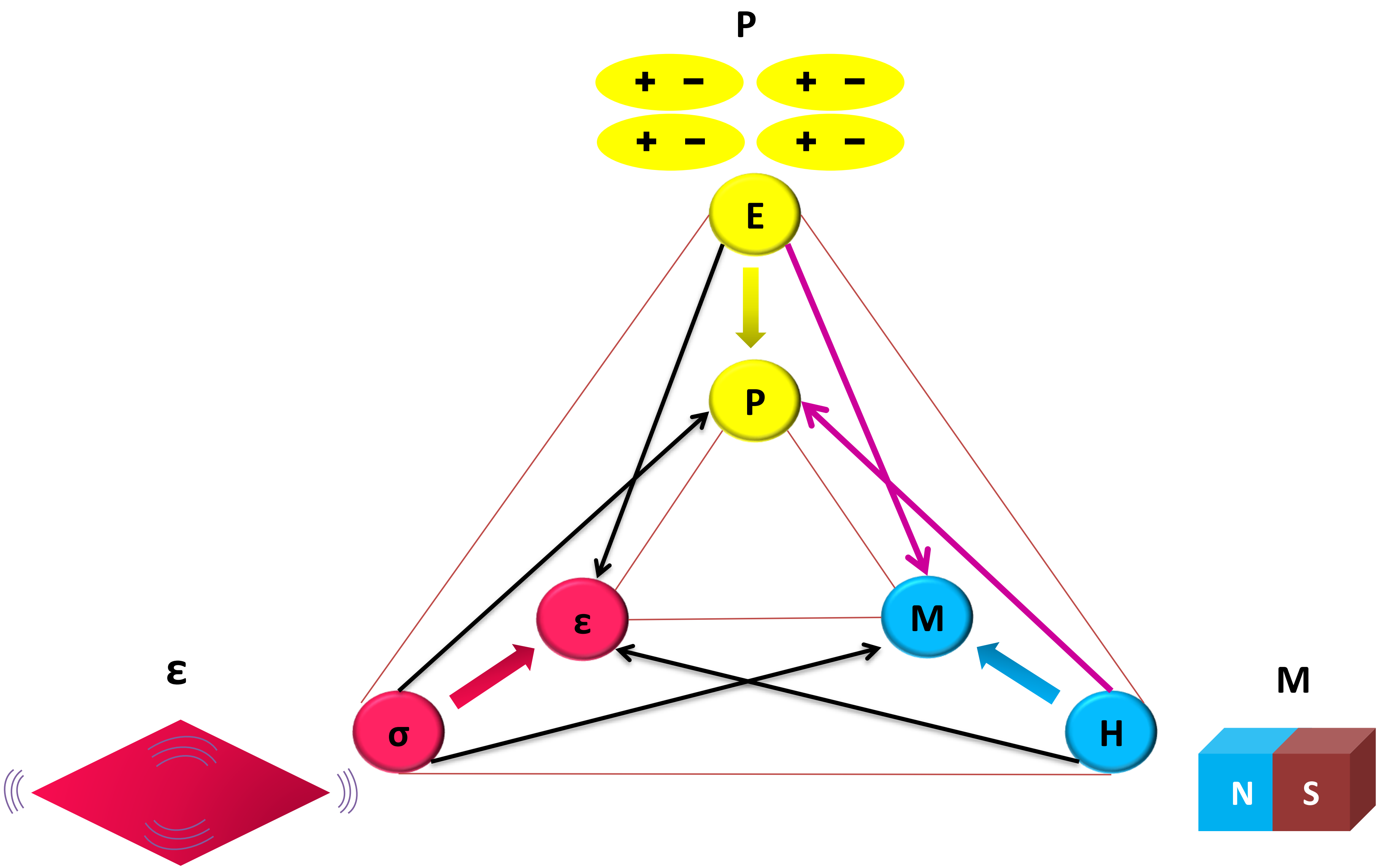
The future microelectronics

Moore's Law	**More than Moore**	**Beyond Moore**
Scaling Down	Functional Diversification (RF, optical, sensing tech)	"New" physics (electron spin, magnetic dipoles, photons…)

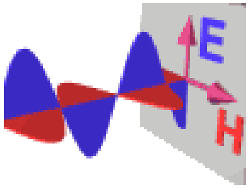




Multiferroics



Dynamical ME coupling



http://physics-animations.com

electric and magnetic components of light

$\boldsymbol{E}^{\omega}$ and $\boldsymbol{H}^{\omega}$

Non-multiferroic

$$\boldsymbol{P} = \chi^{\mathrm{e}}\boldsymbol{E}$$

$$\boldsymbol{M} = \chi^{\mathrm{m}}\boldsymbol{H}$$

BUT

oscillating magnetization and polarization

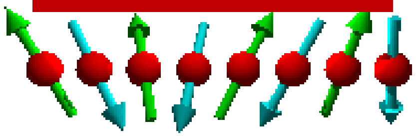
$\boldsymbol{M}^{\omega}$ and $\boldsymbol{P}^{\omega}$

$$\boldsymbol{P}_i^{\omega} = \chi_{ii}^{\mathrm{e}}(\omega)\boldsymbol{E}_i^{\omega} + \alpha_{ij}^{\omega}\boldsymbol{H}_j^{\omega}$$

$$\boldsymbol{M}_j^{\omega} = \chi_{jj}^{\mathrm{m}}(\omega)\boldsymbol{H}_j^{\omega} + \alpha_{ji}^{\omega}\boldsymbol{E}_i^{\omega}$$

$$\mathrm{N}_{\pm}(\omega) \approx \sqrt{\varepsilon_{ii}(\omega)\mu_{jj}(\omega)} \pm \frac{1}{2}\left[\chi_{\mathrm{ij}}^{\mathrm{me}}(\omega) + \chi_{\mathrm{ij}}^{\mathrm{em}}(\omega)\right]$$

Electromagnons


O. Waldmann, Freiburg University

Kezsmarki *et al.*, *Nature Communications* **5** (2014)

Electromagnon

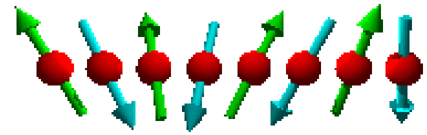
Experimental Methods

Time-domain THz Transmission → Phonons, Magnons, Electromagnons

Fourier Transform Infrared Reflectivity → Phonons

Raman → Phonons, Magnons, Electromagnons

Electromagnons ?



O. Waldmann, Freiburg University

Simultaneous detection of magnetic excitations in both IR and Raman

Computational Methods

VASP (Vienna Ab-initio Simulation Package)

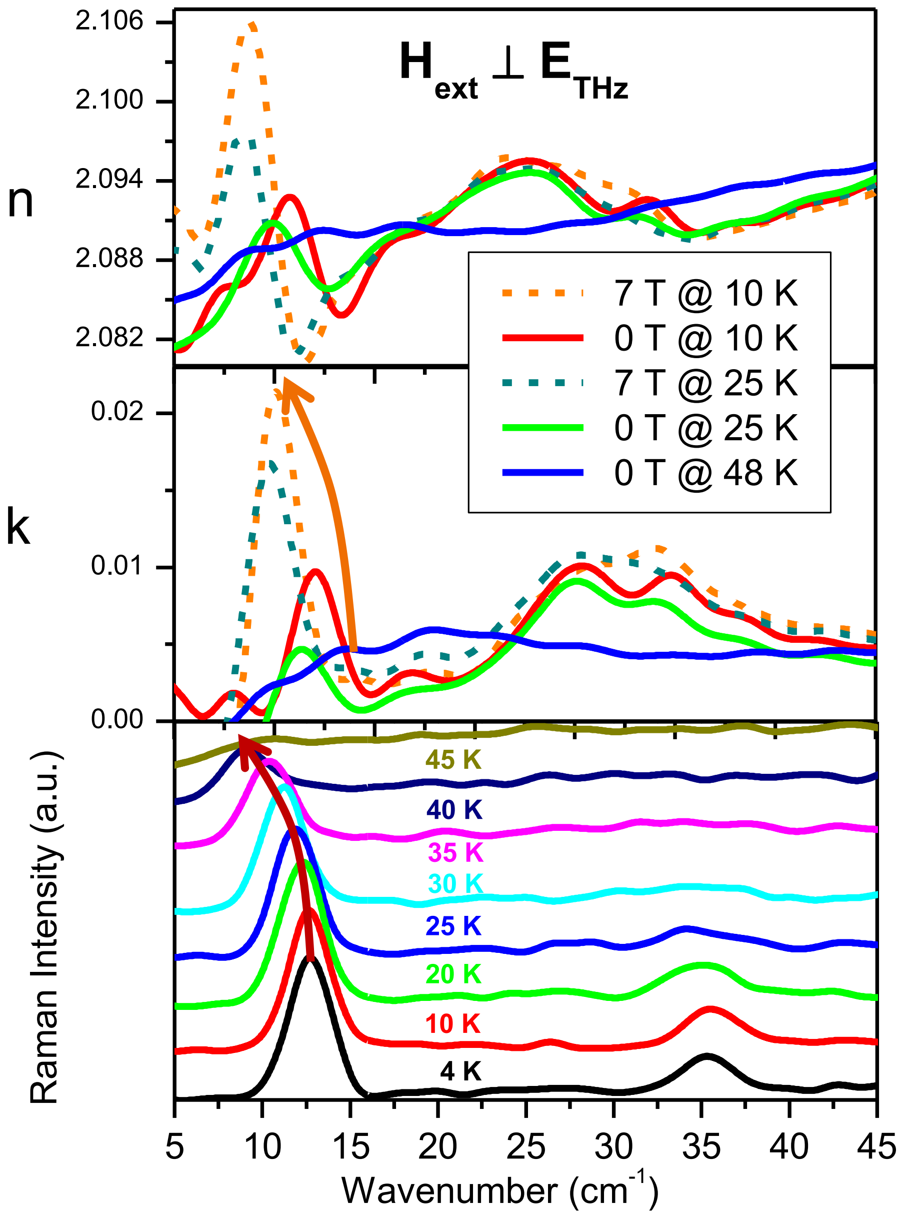
phonopy 1.11.6

Density Functional Theory (DFT)
Spin-polarized
Spin-orbit coupling
GGA + U

+

Phonon calculation

Ni_3TeO_6: Raman and THz active modes



PHYSICAL REVIEW B **95**, 184435 (2017)

Magnetoelectric excitations in multiferroic Ni_3TeO_6

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(Received 24 February 2017; published 30 May 2017)

2 excitations in THz range

Detected simultaneously at Raman and THz spectra

+

Tuned by H_{ext}

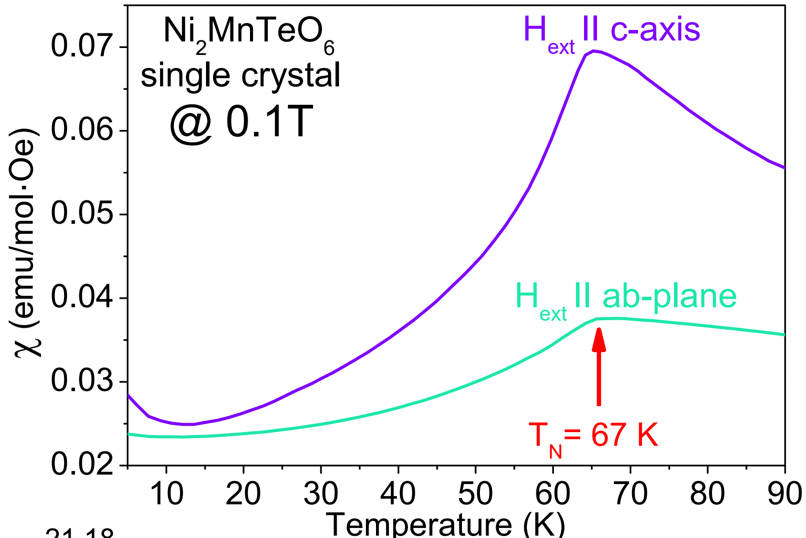
electromagnons

Skiadopoulou *et al.*, PRB **95** (2017)

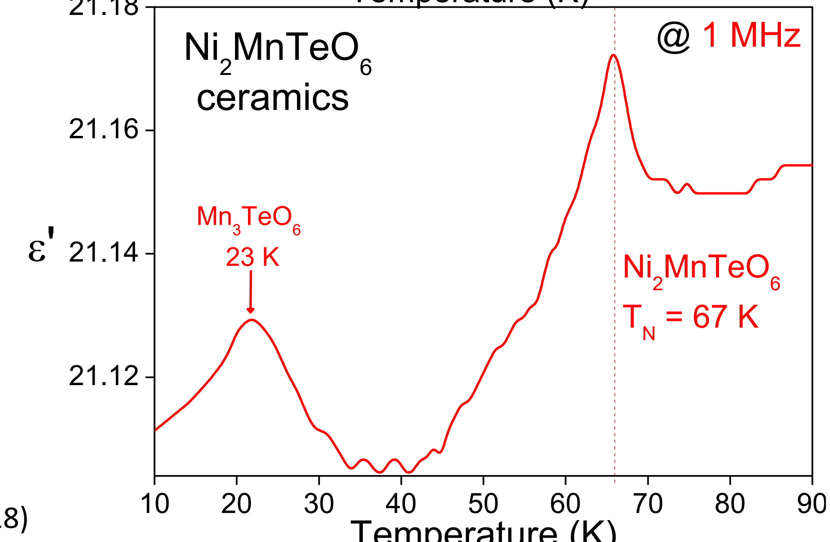
Ni_2MnTeO_6: magnetic and dielectric properties

Ni substitution by **Mn** ➡ ⬆ T_N by ~20 K, **T_N = 67 K**

AFM phase transition
T_N = 67 K



dielectric anomaly
@ T_N = 67 K

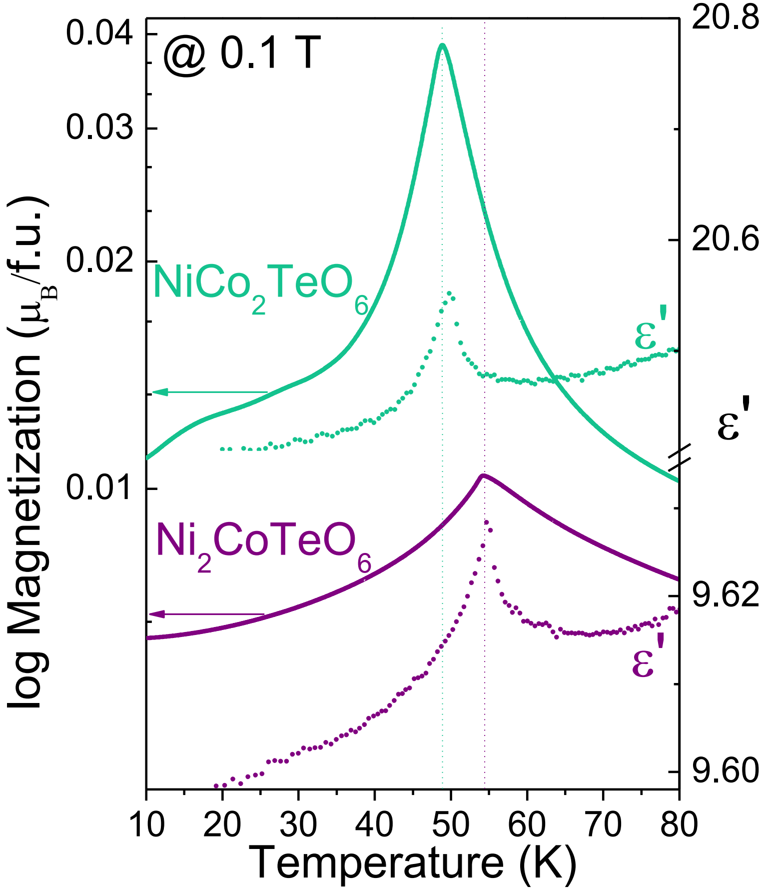


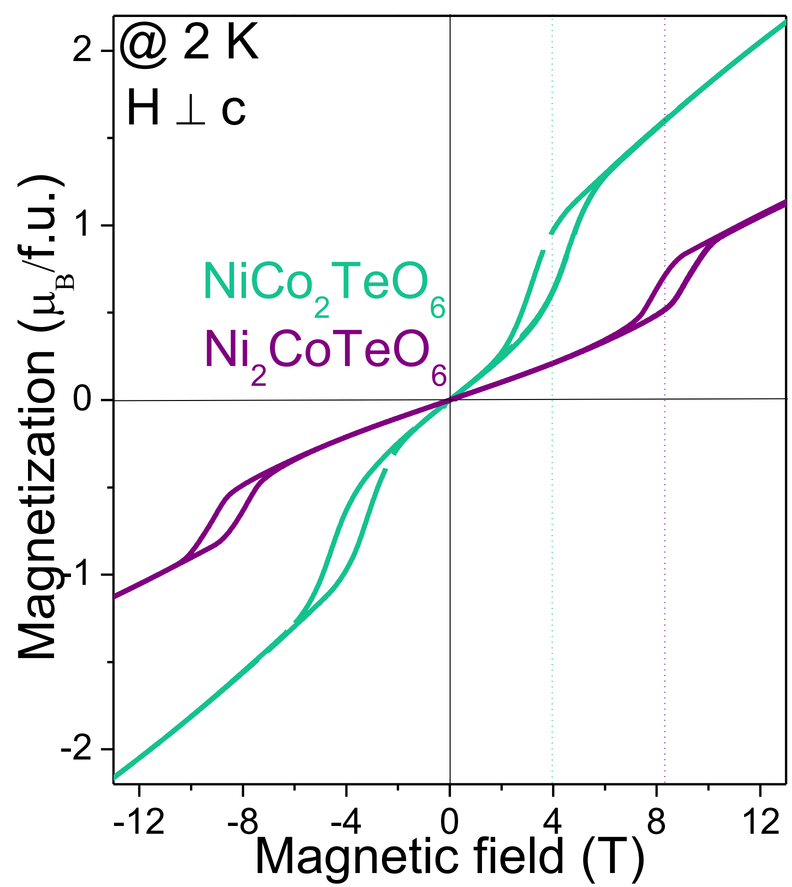
Retuerto, Skiadopoulou *et al.*, *PRB* **97**, 144418 (2018)

$Ni_{3-x}Co_xTeO_6$: magnetic properties

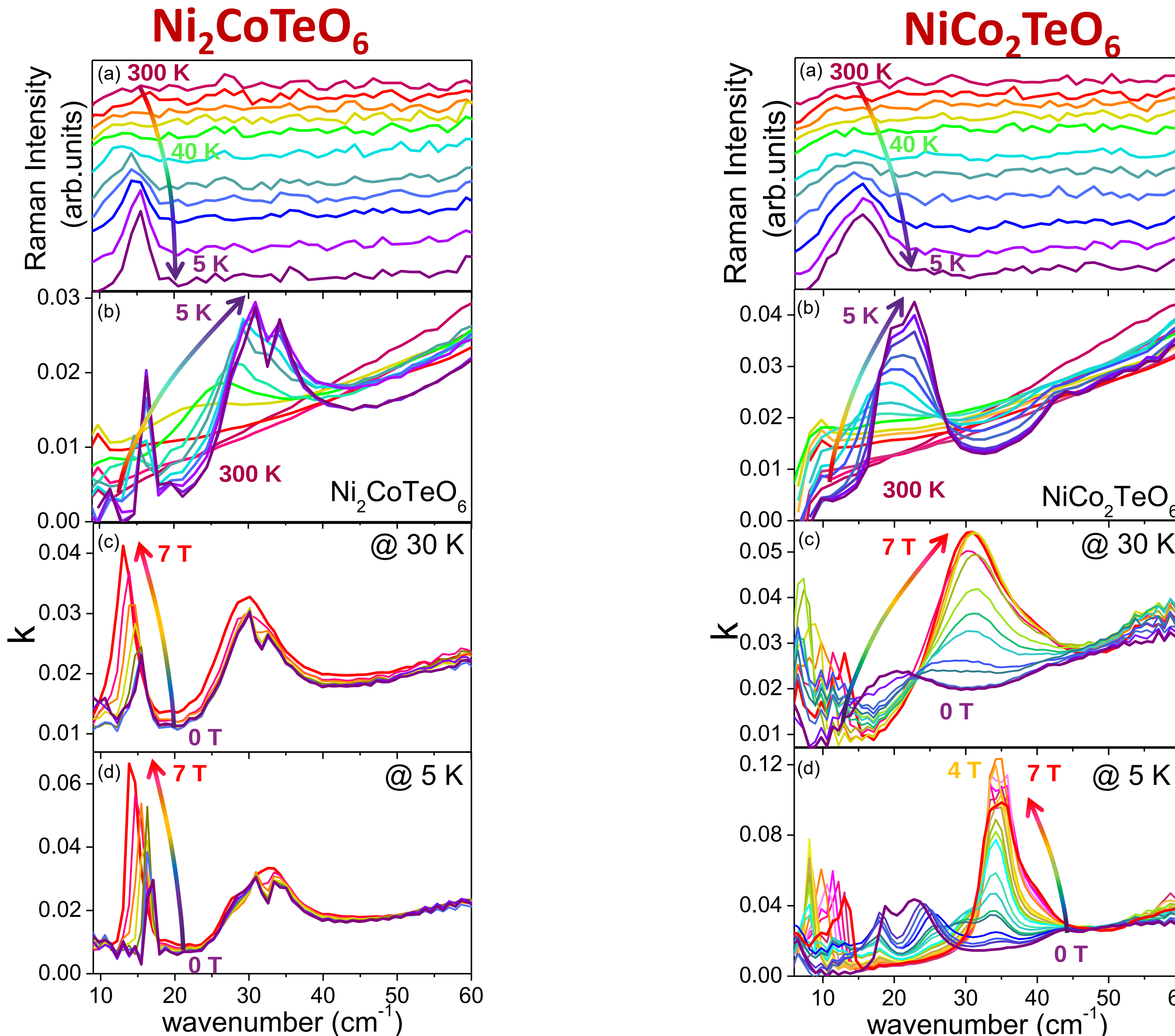
Ni substitution by **Co** ➡

Ni_2CoTeO_6 : $T_N \approx 49$ K, $H_{sf} \approx 8$ T, *R*3
$NiCo_2TeO_6$: $T_N \approx 54$ K, $H_{sf} \approx 4$ T, *R*3





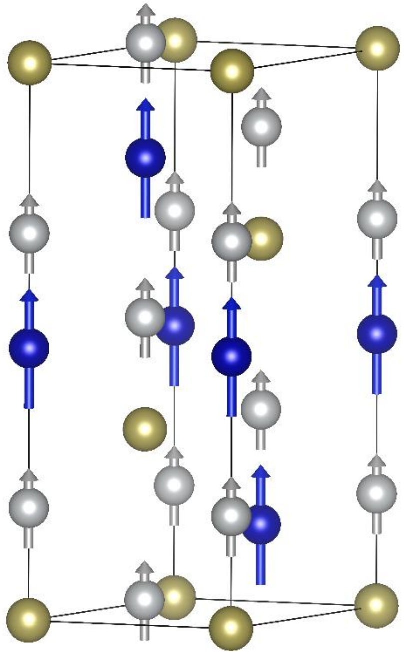
$Ni_{3-x}Co_xTeO_6$: Raman and THz active modes

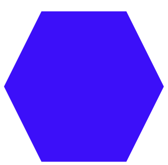


$Ni_{3-x}Co_xTeO_6$: DFT calculations

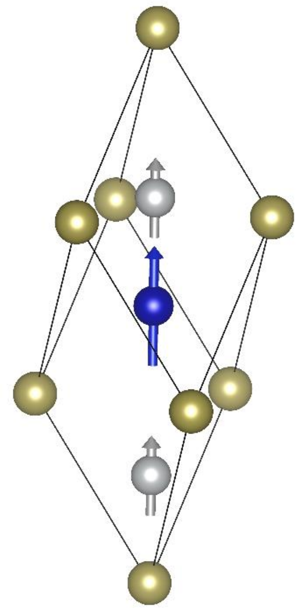
Ni_2CoTeO_6
$NiCo_2TeO_6$

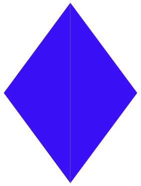
Spin-polarized
Spin-orbit coupling
GGA + U[1]
U_{eff} = 7 eV/Ni,Co





Hexagonal setting
3*rhombohedral (30 atoms)
Double cell along c-axis (60 atoms)





Rhombohedral setting
10 atoms

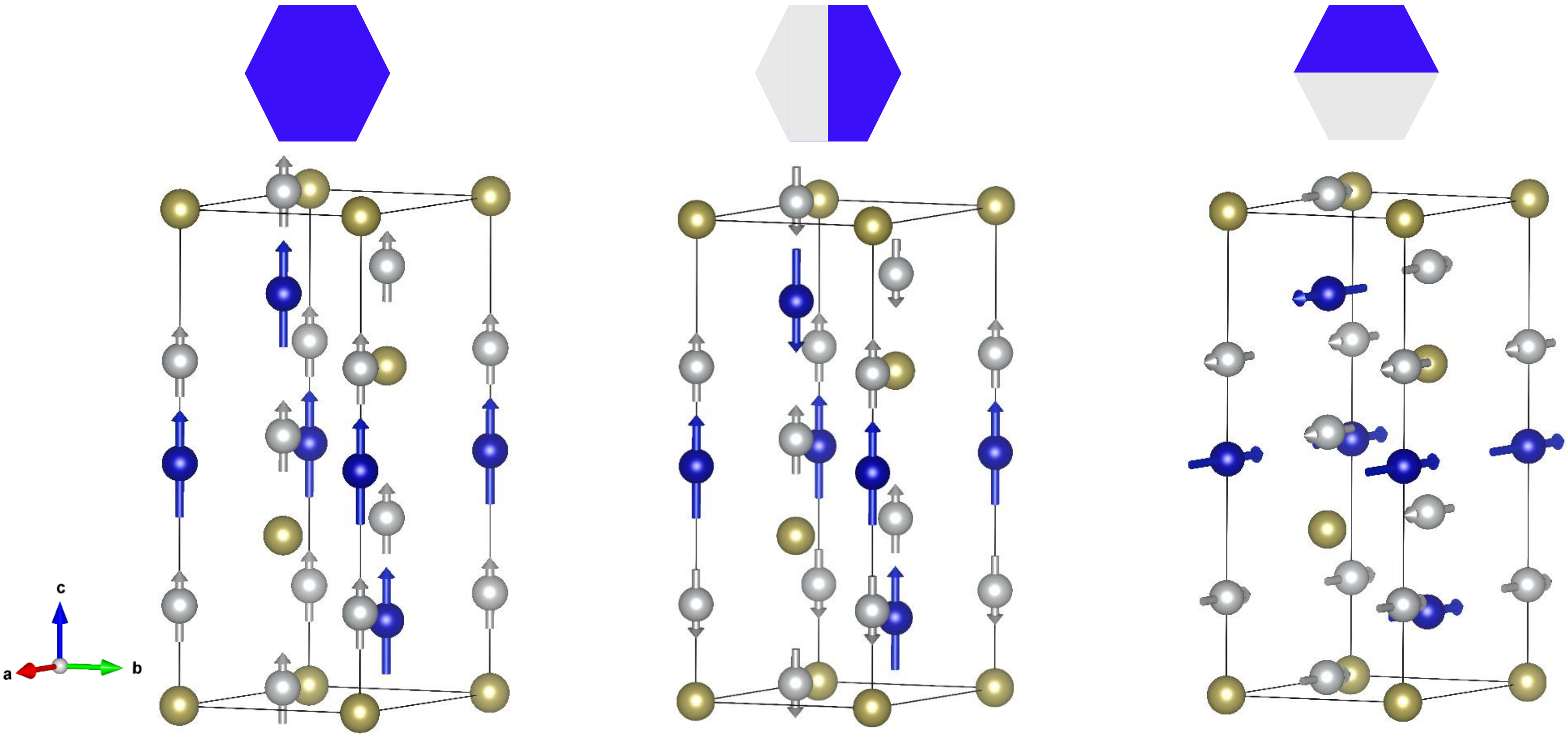
1. **S. L. Dudarev,** *et al.*, Phys. Rev. B 57, 1505 (1998).

$Ni_{3-x}Co_xTeO_6$: DFT+U calculations

	Ni_2CoTeO_6		$NiCo_2TeO_6$	
	Experiment	**DFT+U**	**Experiment**	**DFT+U**
μ_{Ni} (μ_B)	**2**[1] (Ni_3TeO_6)	**1.8**	**2**[1] (Ni_3TeO_6)	**1.8**
μ_{Co} (μ_B)	**2-2.7**[2] (Co_3TeO_6)	**2.9**	**2-2.7**[2] (Co_3TeO_6)	**2.9**
Volume (Å^3)	**313.75**	**322.42**	**316.14**	**327.33**
Lattice parameters (Å)	a = **5.1257** c = **13.7874**	a = **5.1649** c = **13.9561**	a = **5.1606** c = **13.8372**	a = **5.2106** c = **13.9237**
Band Gap (eV)	**2.2**[3] (Ni_3TeO_6)	**2.5**	**2.2**[3] (Ni_3TeO_6)	**2.3**

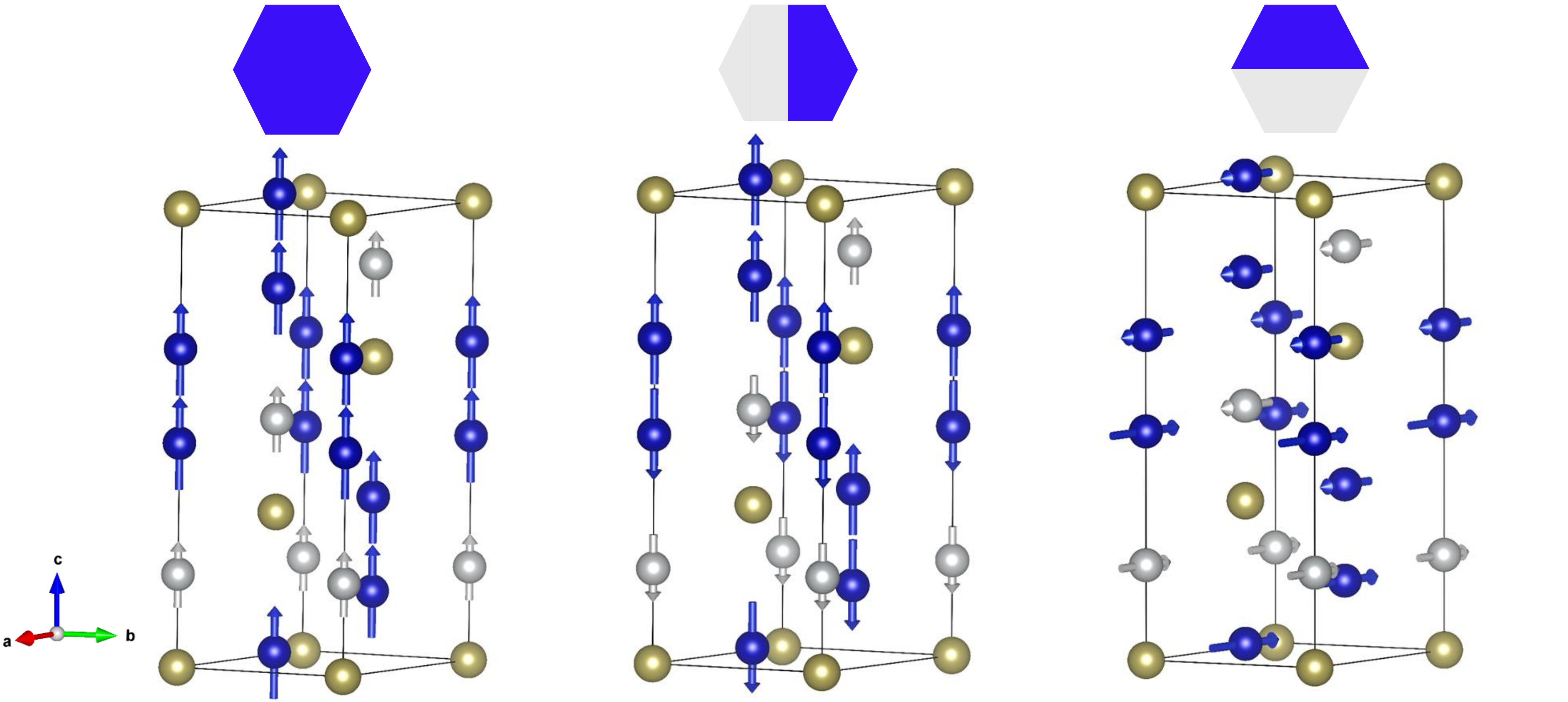
1. I. Živković, *et al.*, *J. Phys. Condens. Matter* **22**, 56002 (2010).
2. M. Rössle, *et al.*, *Phys. Rev. B* **88**, 104110 (2013).
3. Yokosuk *et al.*, *PRL* **117**, 147402 (2016)

Ni_2CoTeO_6: magnetic anisotropy



ΔE (meV/atom)		
1.2	0.3	0
FM: $M_{tot} \approx 42\ \mu_B$	AFM: $M_{tot} = 0$	
	Ni: $m_L \approx 0.18\ \mu_B$, Co: $m_L \approx 0.18\ \mu_B$	

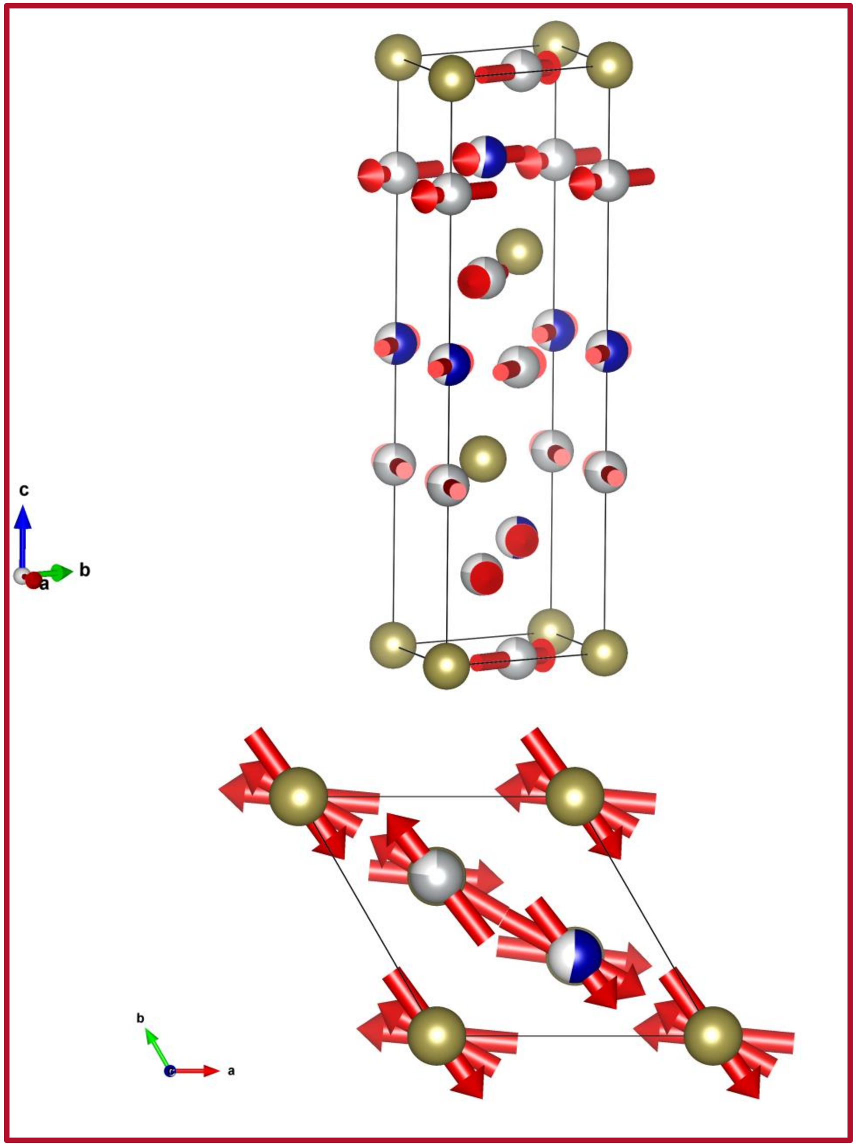
$NiCo_2TeO_6$: magnetic anisotropy



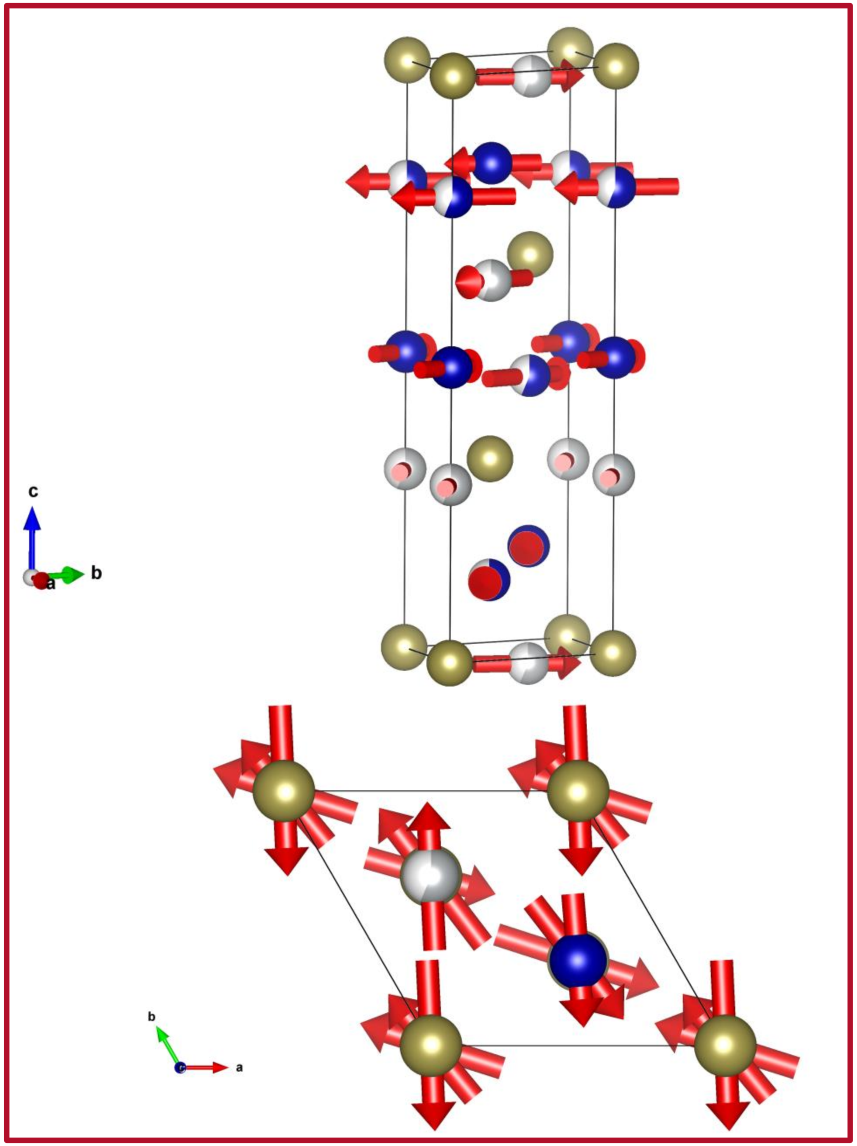
ΔE (meV/atom)		
6.6 (?)	6.5 (?)	0
	M≠ 0, 1 Ni flipped	
	Ni: $m_L \approx 0.18\ \mu_B$, Co: $m_L \approx 1.4\ \mu_B$	

$Ni_{3-x}Co_xTeO_6$: noncollinear in-plane (PND)

Ni_2CoTeO_6



$NiCo_2TeO_6$



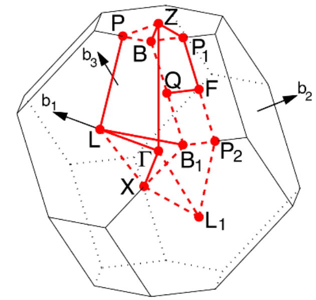
Ni_2CoTeO_6: Phonons

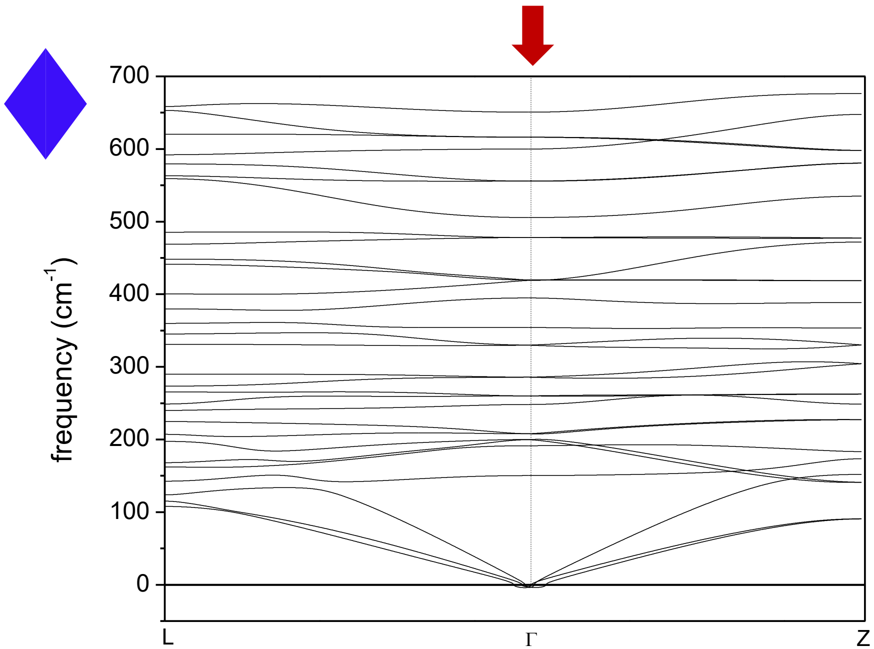
***R*3 (No, 146)**

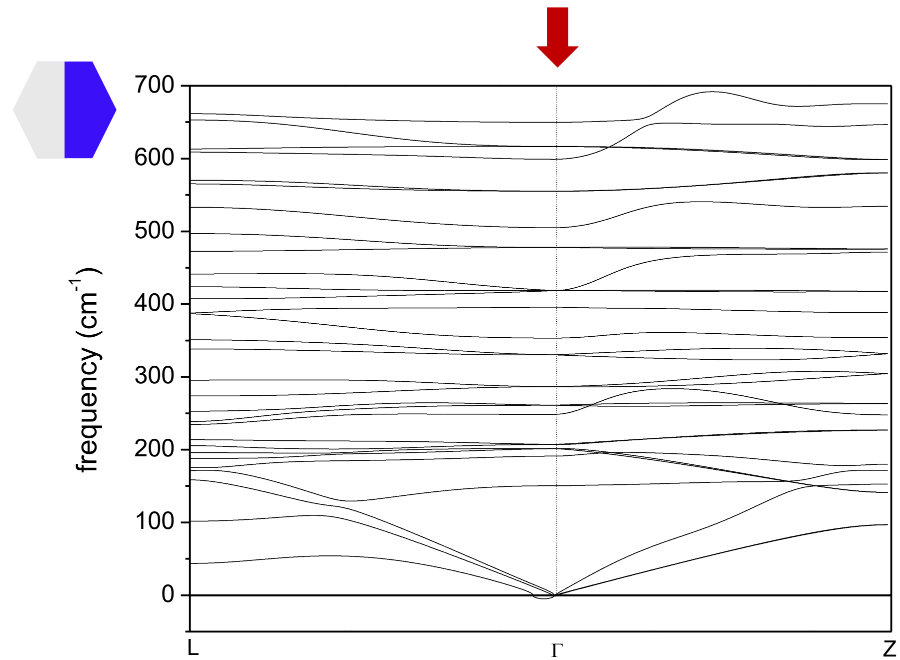
@ Γ-point: 18 IR- and Raman-active modes

9 *E*(x,y,x^2-y^2,xy,xz,yz) + 9 *A*(x^2+y^2,z^2,z)

in agreement with experiment





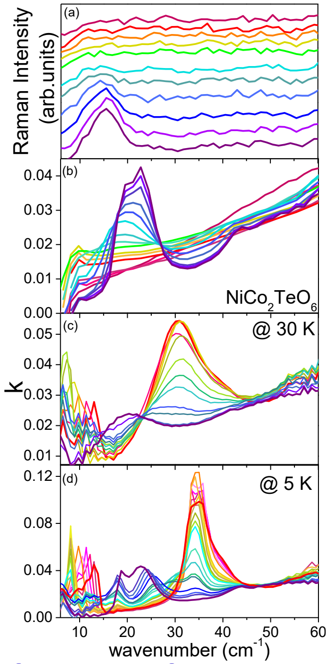


phonopy 1.11.6

Ni-based tellurates: polar antiferromagnets

$Ni_{3-x}Co_xTeO_6$

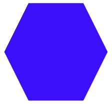
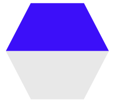
H_{ext}-tunable spin excitations



1 simultaneously seen in Raman

↓

ME excitation

Magnetic structure/anisotropy ΔE (meV/atom)			
			
Ni_2CoTeO_6	1.2	0.3	0
		Ni: $m_L \approx 0.18\ \mu_B$, Co: $m_L \approx 0.18\ \mu_B$	
$NiCo_2TeO_6$	6.6 (?)	6.5(?)	0
		Ni: $m_L \approx 0.18\ \mu_B$, Co: $m_L \approx 1.4\ \mu_B$	

- Examine in-plane non-collinear spin-order
- Are the lattice vibrations affected by the magnetic structure…?

Future →

Experiment
magnetoelectric coupling
directional dichroism

+

Calculations
exchange interactions
Phonons
TO-LO splitting

Thank you for your attention!

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