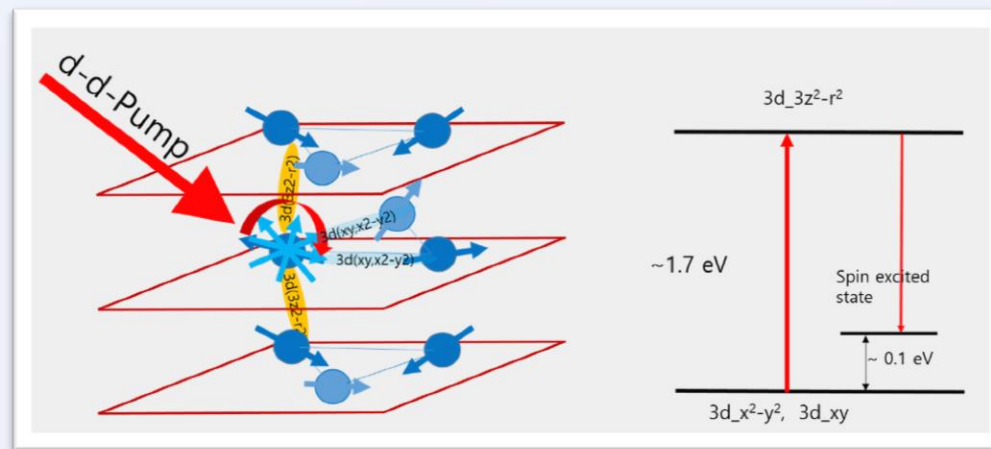


Raman study of spin excitations in hexagonal $RMnO_3$

October 16, 2019
고려대학교 물리학과



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Education

1988.8 PhD, University of Illinois at Urbana-Champaign (A.C. Anderson)

1986.5 MS, University of Illinois at Urbana-Champaign (A.C. Anderson)

1983.2 BS, Seoul National University, Department of Physics

PhD Thesis:

Phonon-dislocation interaction in LiF and KCl crystals

Professional Experience:

2000. 3 - present : Professor, Department of Physics and Division of nano-sciences, Ewha Womans University

2009.3 – 2010. 2 : Director, Basic Science Research Institute, Ewha Womans University

2005. 5 - 2008. 4 : Editor, Journal of the Korean Physical Society.

2002. 3 – 2004. 2: Chief Secretary, Condensed Matter Division, Korean Physical Society.

1998. 8 – 2002. 2 : Visiting Research Associate Professor, Physics, University of Illinois at Urbana-Champaign

1994.9 - 1999.8: Associate Professor, Department of Physics, Ewha Womans University

1990.9 - 1994.8 : Assistant Professor, Department of Physics, Ewha Womans University

1988.9 - 1990.8 : IBM Thomas J. Watson Research Center, Post-Doctoral

Awards:

Korean Government Fellowship for Studying Abroad, 1983

Award for Prospective Young Scientists, Korean Physical Society, 1996

Ewha Best Lecturer Award, 2006



CONTRIBUTORS

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Gwangju Institute of Science and Technology, Gwangju, Korea

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Rutgers Center for Emergent Materials and Department of Physics, New Jersey, USA

Young Mee Jung

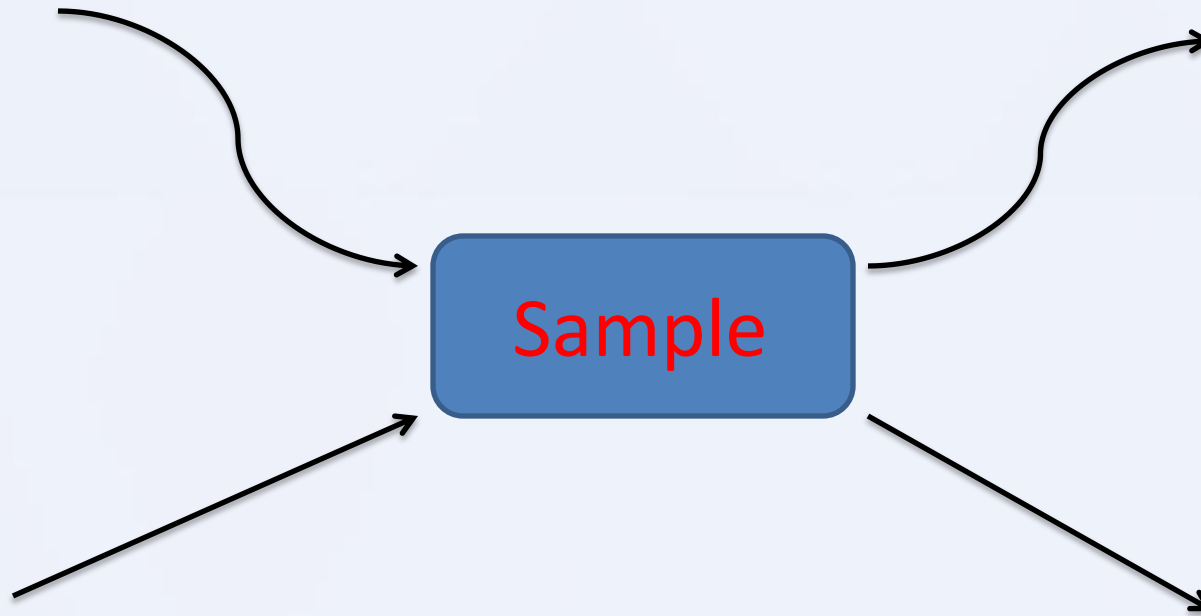
Department of Chemistry, Kangwon National University, Chunchon, Korea



Probing methods

Light (X-ray, UV, Visible, IR)

Light



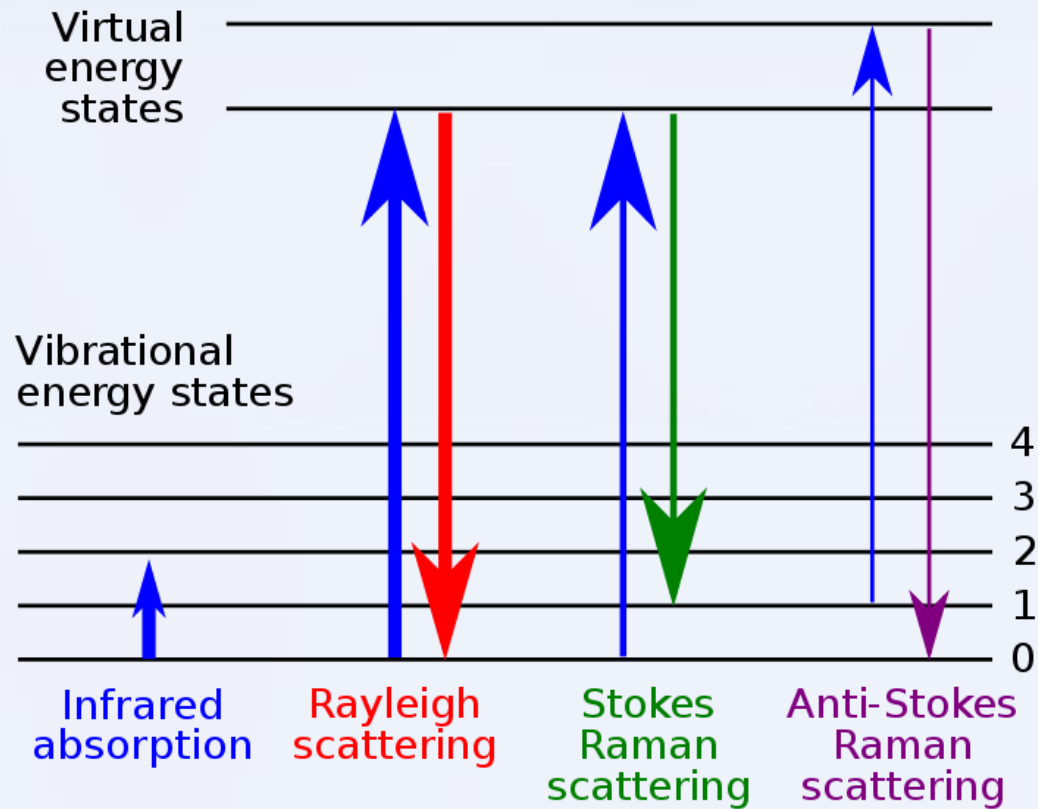
Particle (electron, neutron)

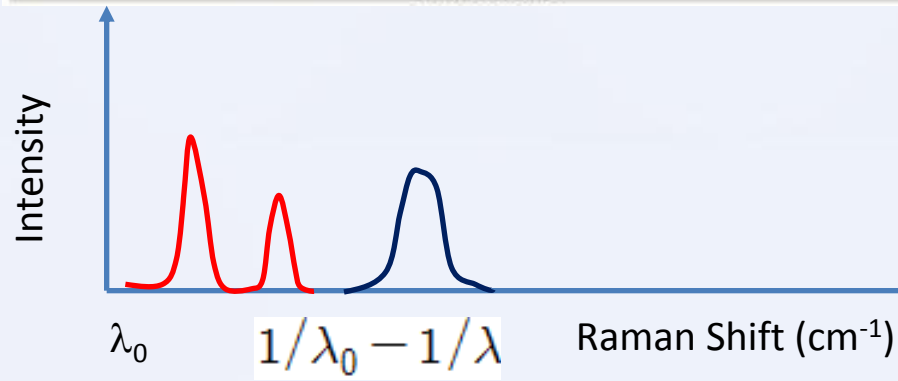
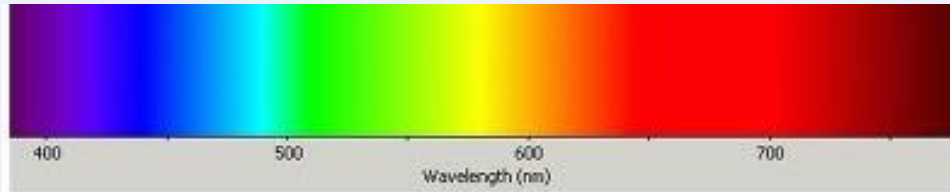
Particle

Raman Spectroscopy

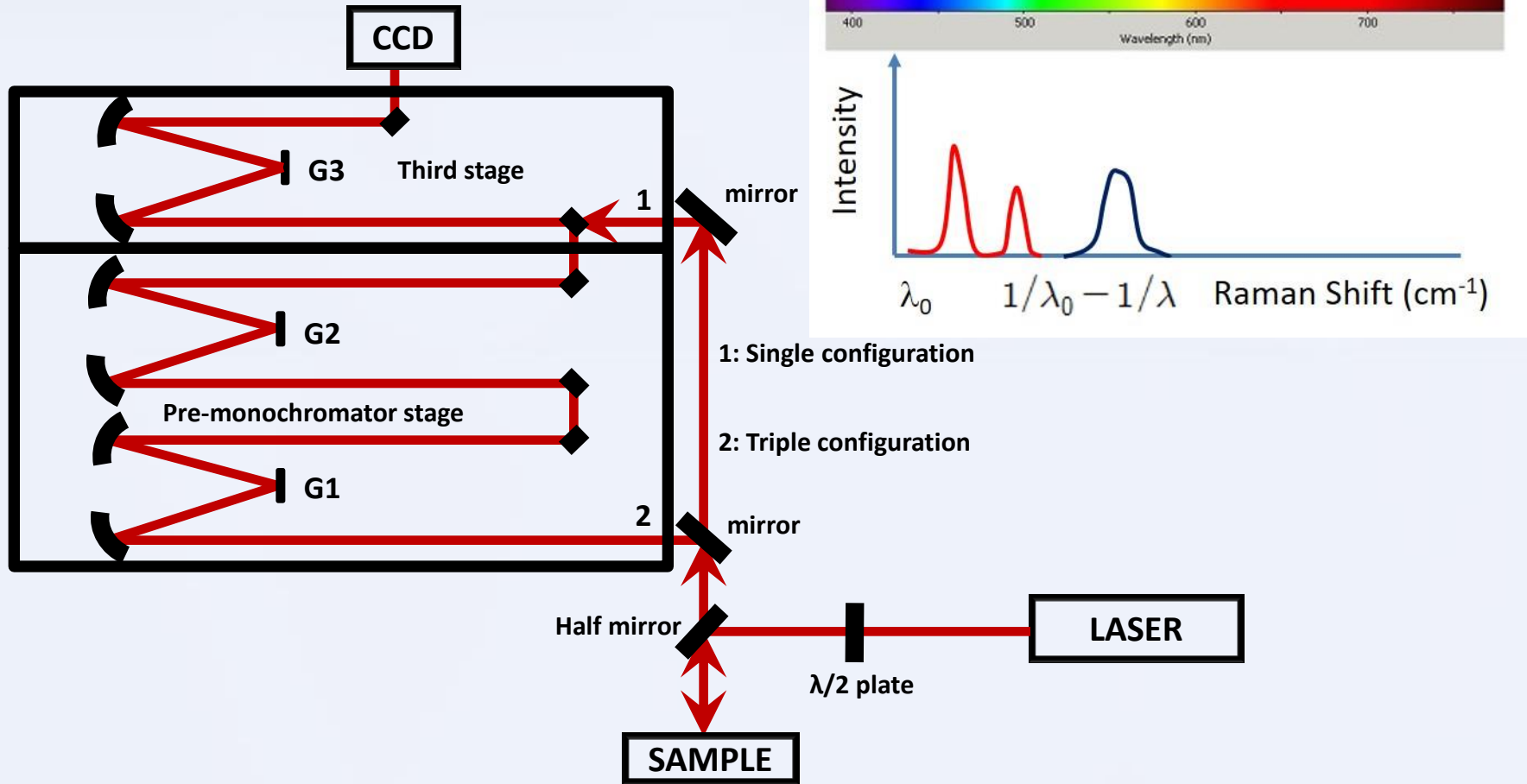


Low energy excitation = $h(\nu - \nu')$





Raman Spectroscopy



Jobin Yvon T64000 spectrometer

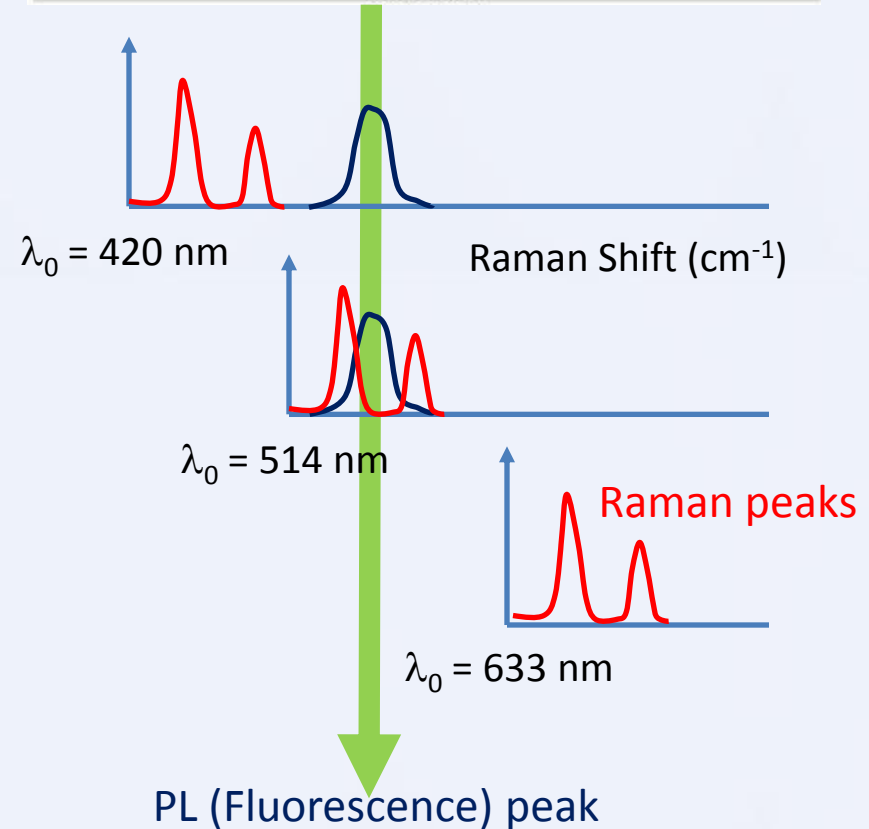
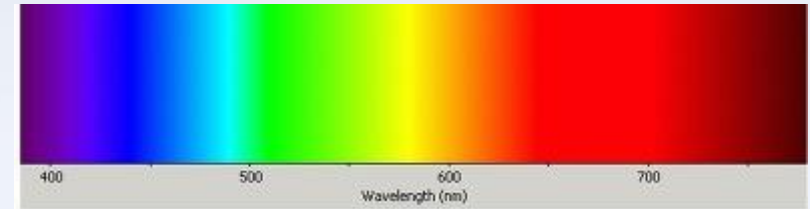
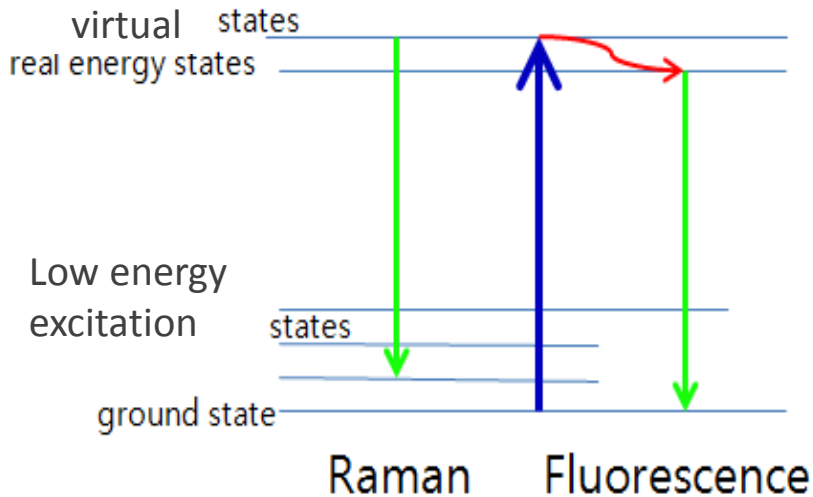
Raman Effect vs Fluorescence

Raman spectroscopy

$$h\nu \rightarrow h\nu' ; \text{excitation} = h(\nu - \nu')$$

Fluorescence spectroscopy

$$h\nu \rightarrow h\nu' ; \text{excitation} = h\nu'$$



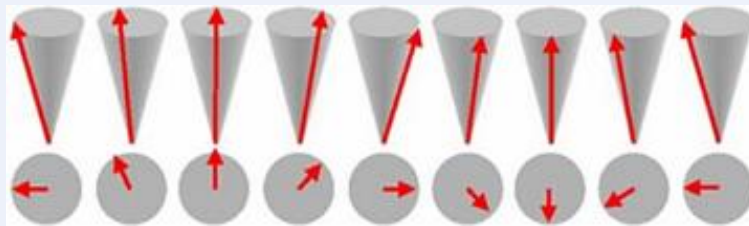
Raman spectroscopy can probe :

- Vibrational states
- Rotational states
- Electronic states
- Charge ordering (Superconducting gap, 2Δ)
- Spin ordering (spin waves)
- ...

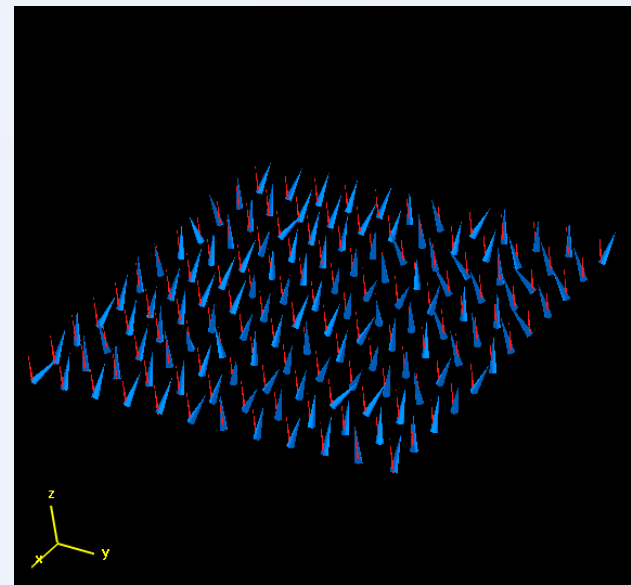
Quantized Spin Waves

A **magnon** is a [quasiparticle](#), a [collective excitation](#) of the [electrons spin](#) structure in a [crystal lattice](#). In the equivalent wave picture of quantum mechanics, a magnon can be viewed as a quantized [spin wave](#). Magnons carry a fixed amount of [energy](#) and [lattice momentum](#), and are spin-1, indicating they obey boson behavior.

- From Wikipedia, the free encyclopedia -



1-D Analogy



2-D Analogy

Quantum Memories

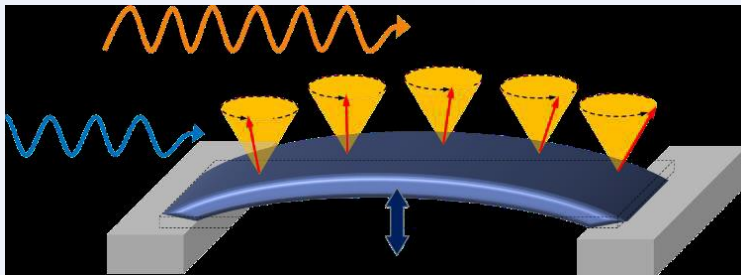
- Superconducting Nano devices
 - Nuclear spin - decoherence time
 - Atomic states - natural candidates
- ➔ Require strong light-matter interaction

- Solid state systems = scalability, integration into existing technology

$\text{Pr}^{3+}:\text{Y}_2\text{SiO}_5$ crystal : solid-state **spin-wave** optical quantum memory

-Mustafa Gündoğan et al., Phys. Rev. Lett. 114, 230501 (2015)

Precession of Magnetic Moments – **Spin Wave**



Magneto-Mechanical Resonator

Adopted from the Talk of Cho, Sung Un, Department of Physics and Astronomy, Seoul National University (July 4, 2017)

Spin Wave

a reliable channel of the light-matter interaction:

excitation/detection
(write/read) in
the Quantum Memories

0. Brief history of Raman studies of spin excitations in hexagonal $RMnO_3$ system

Spin excitations in hexagonal $RMnO_3$ (R = rare earths)

+ First found in hexagonal $HoMnO_3$ thin films, using 647 nm excitation source.

Raman scattering studies of the magnetic ordering in hexagonal $HoMnO_3$ thin films, Nguyen Thi Minh Hien, Xiang-Bai Chen, Luc Huy Hoang, D, Lee, S.-Y. Jang, T. W. Noh, and In-Sang Yang, Journal of Raman Spectroscopy 41, 983-988 (2010)

+ The spin excitations were resonant in very narrow range of excitation energy.

Resonant A(1) phonon and four-magnon Raman scattering in hexagonal $HoMnO_3$ thin film, Xiang-Bai Chen, Nguyen Thi Minh Hien, D Lee, S-Y Jang, T W Noh, and In-Sang Yang, New Journal of Physics 12, 073046 (2010)

+ The spin excitations were found in other $RMnO_3$ (R = rare earths).

Raman scattering studies of hexagonal rare-earth $RMnO_3$ (R = Tb, Dy, Ho, Er) thin films, Nguyen Thi Minh Hien, Su-Young Oh, Xiang-Bai Chen, D. Lee, S.-Y. Jang, T. W. Noh, and In-Sang Yang, Journal of Raman Spectroscopy 42, 1774-1779 (2011)

+ We could get the spin-exchange interaction integral J values from Raman!

Spin exchange interactions in hexagonal manganites $RMnO_3$ (R = Tb, Dy, Ho, Er) epitaxial thin films, Xiang-Bai Chen, Nguyen Thi Minh Hien, D. Lee, S.-Y. Jang, T. W. Noh, and In-Sang Yang, App.Phys.Lett. 99, 052506 (2011)

Magnons in hexagonal $RMnO_3$ ($R = \text{rare earths}$)

+ Spin excitations in Mn^{+3} sublattices of hexagonal $RMnO_3$ can be selectively excited!
-> Raman study is much more useful in studying the magnetic transitions due to changes in Mn-spin interactions than magnetization measurements.

"Study of spin-ordering and spin-reorientation transitions in hexagonal manganites through Raman spectroscopy", Xiang-Bai Chen, Nguyen Thi Minh Hien, Kiok Han, Ji-Yeon Nam, Nguyen Thi Huyen, Seong-Il Shin, Xueyun Wang, S. W. Cheong, D. Lee, T. W. Noh, N. H. Sung, B. K. Cho, and In-Sang Yang, Scientific Reports 5, 13366 (2015).

+ Spin excitations tell us about the spin-structural phase transition in hexagonal $RMnO_3$.

"Correlation between magnon and magnetic symmetries of hexagonal $RMnO_3$ ($R = \text{Er, Ho, Lu}$), Thi Minh Hien Nguyen, Thi Huyen Nguyen, Xiang-Bai Chen, Yeonju Park, Young Mee Jung, D. Lee, T.W. Noh, Sang-Wook Cheong, and In-Sang Yang, Journal of Molecular Structure 1124, 103-109 (2016).

+ We suggested the **spin flip** mechanism in hexagonal LuMnO₃ single crystal.

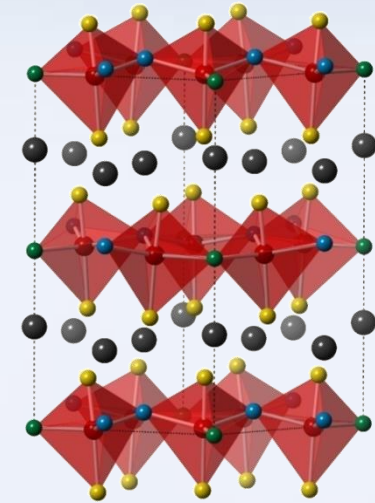
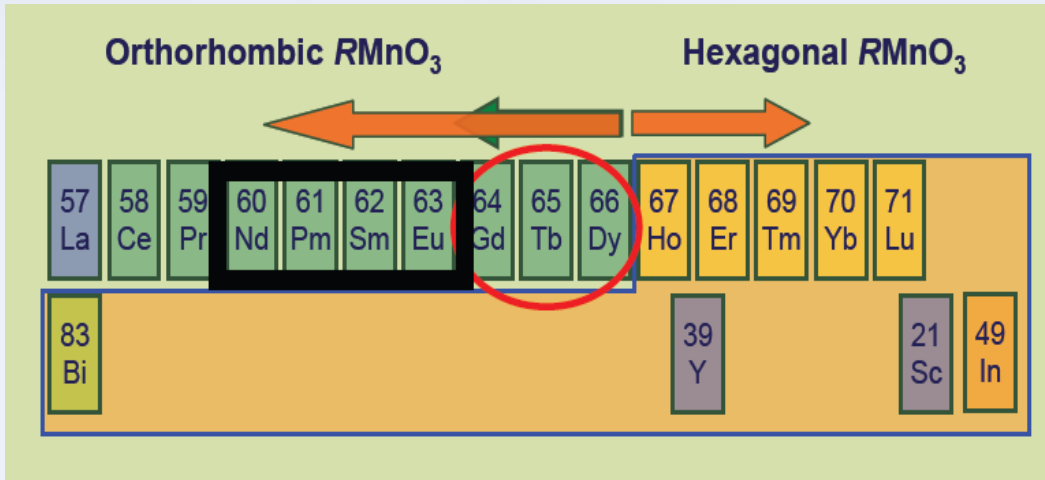
“Spin wave and spin flip in hexagonal LuMnO₃ single crystal”, Xiang-Bai Chen, Peng-Cheng Guo, Nguyen Thi Huyen, Seung Kim, **In-Sang Yang**, Xueyun Wang, Sang-Wook Cheong, Appl. Phys. Lett. **110**, 122405 (2017).

+ Dr. Hien investigated two-magnons in other AIAO material (Cd₂Os₂O₇)

“Two-Magnon Scattering in 5d All-In-All-Out Pyrochlore Magnet Cd₂Os₂O₇”, Nguyen Thi Minh Hien, Luke J. Sandilands, C.H. Sohn, C. H. Kim, Aleksander L Wysocki, **In-Sang Yang**, S. J. Moon, Jae-Hyeon Ko, Z. Hiroi, J. Yamaura, and Tae Won Noh, Nature Communications 8, 251 (2017).

-> The spin excitations are very important, useful quantity !

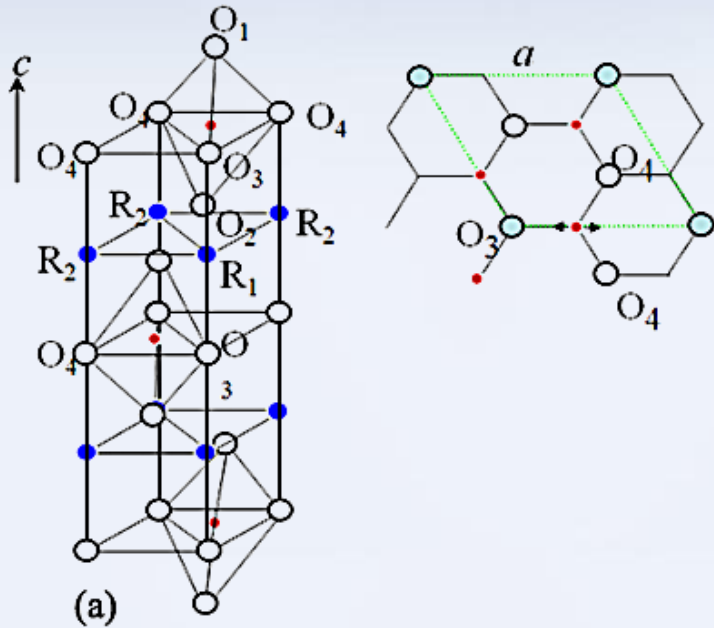
I. Review of RMnO_3 : why hexagonal RMnO_3 ?



	Ferroelectric T_{FE} (K)	P_r ($\mu\text{C}/\text{cm}^2$)	Anti-FM T_N (K)
Ortho – RMnO_3 ($R = \text{Dy, Tb, Gd}$)	~ 24	~ 0.2	~ 40
Hexa – RMnO_3 ($R = \text{Ho, Y, ...}$)	> 900	~ 5.6	~ 100

<i>Ferroelectric</i>	<i>Ferroelectric</i>	<i>Antiferroelectric</i>	<i>Paraelectric</i>
<i>Antiferromagnetic</i>	<i>Paramagnetic</i>	<i>Paramagnetic</i>	<i>Paramagnetic</i>
	T_N	T_{FE}	T_{AFE}

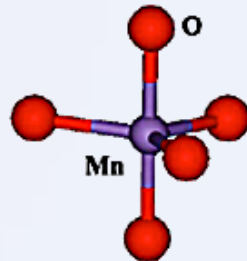
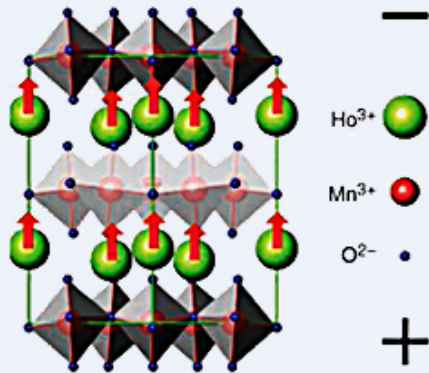
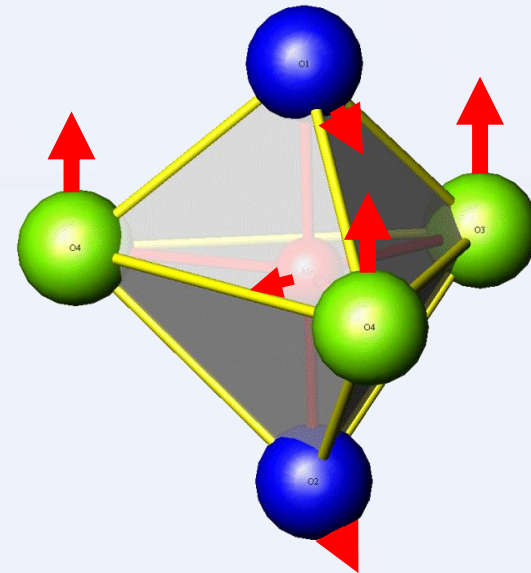
I. Review of RMnO_3 : Structure & Raman active modes



Space group: $P6_3cm$

The unit cell contains

- + Layers of corner sharing MnO_5 bipyramids
- + O3 and O4 : triangular base
- + O1 and O2 : apical sites.



$$\Gamma = 10A_1 + 5A_2 + 10B_1 + 5B_2 + 15E_1 + 15E_2$$

Raman active phonon : $9A_1 + 14E_1 + 15E_2$

(b)

(c)

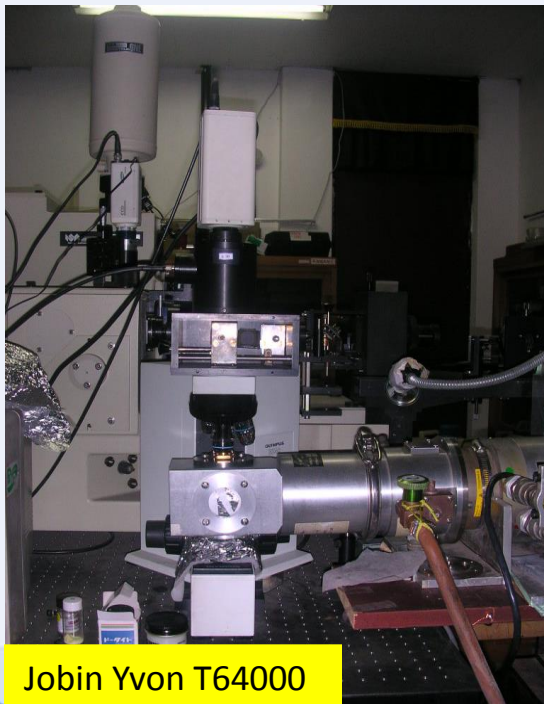
II. Samples and Experiments: *Raman experiments*

Hexagonal HoMnO_3 and ErMnO_3 thin films: were grown on Pt (111)// Al_2O_3 (0001) substrates by laser ablation method. All the thin films were grown epitaxially with their c axis perpendicular to the film surface

Hexagonal LuMnO_3 single crystal was grown using the traveling floating zone method. Platelet single crystal sample was cleaved perpendicular to the c axis.

Raman experiments

- + Objective lenses: $\times 50$ ultra long working distance (ULWD)
- + Focus length: 8mm.



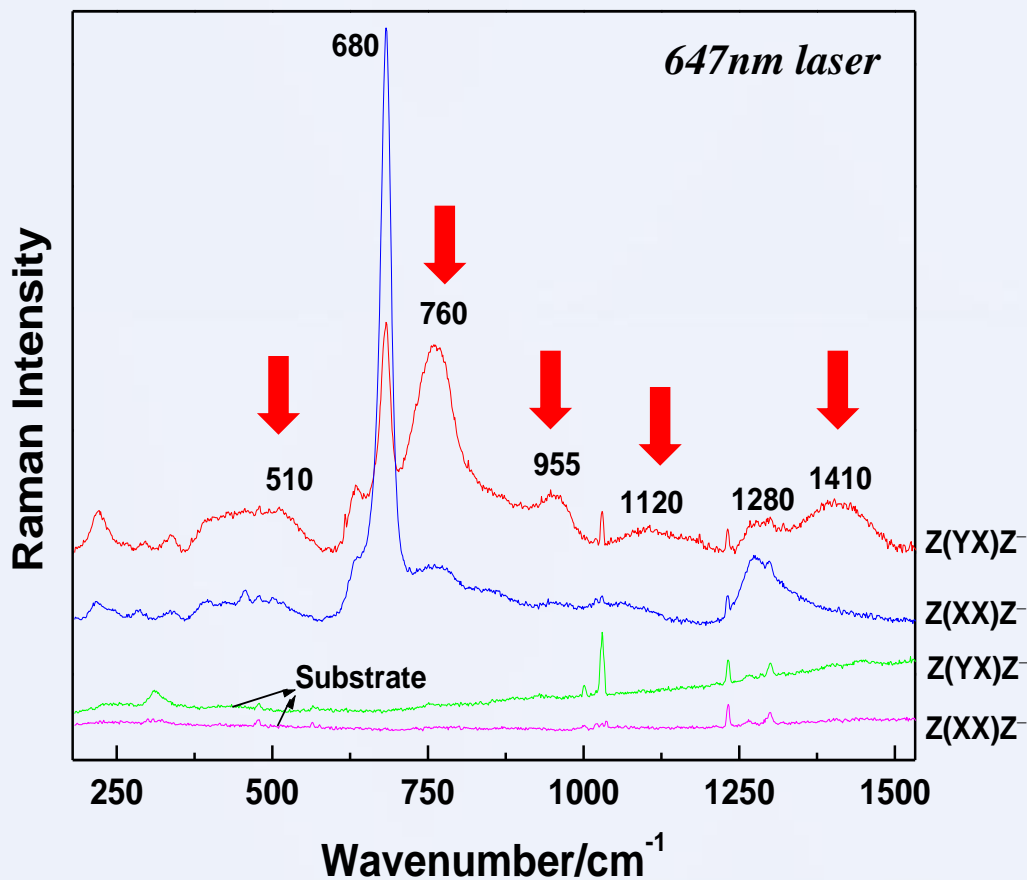
A closed-cycle helium cryostat was used to vary the sample temperatures from 13 to 300K.

1. The magnetic ordering in hexagonal HoMnO_3 thin film

1. Raman scattering studies of the magnetic ordering in hexagonal HoMnO_3 thin films, Nguyen Thi Minh Hien, Xiang-Bai Chen, Luc Huy Hoang, D, Lee, S.-Y. Jang, T. W. Noh, and In-Sang Yang, Journal of Raman Spectroscopy 41, 983-988 (2010)
2. Resonant $A(1)$ phonon and four-magnon Raman scattering in hexagonal HoMnO_3 thin film, Xiang-Bai Chen, Nguyen Thi Minh Hien, D Lee, S-Y Jang, T W Noh, and In-Sang Yang, New Journal of Physics 12, 073046 (2010).

III. Raman results: Spin excitation peaks

Raman spectra of hexagonal HoMnO₃ and Pt(111)//Al₂O₃ (0001) substrate at 13 K in different configurations.



+ 680cm⁻¹: A₁ phonon

+ Broad peak 1280 cm⁻¹:

2nd order of A₁ phonon

+ Several broad peaks :

~510, ~760, ~ 955, ~ 1120

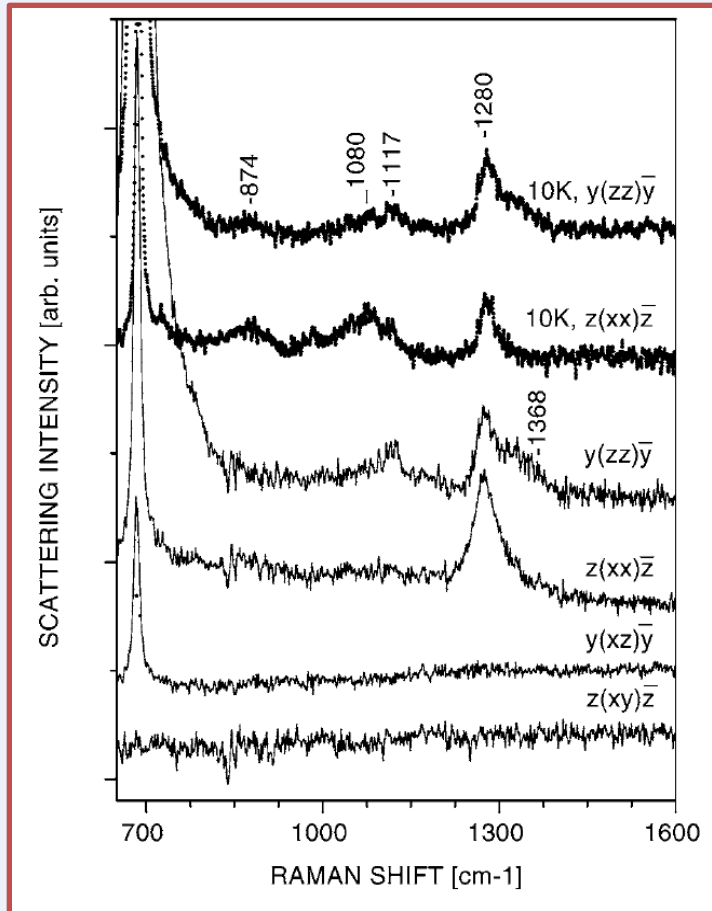
and ~1410 cm⁻¹

- cross polarization only!

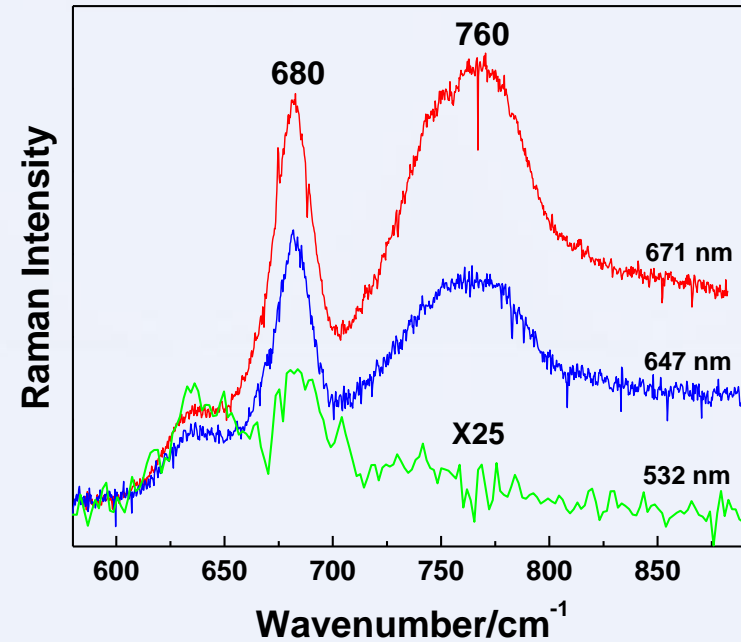
Magnon ??

HoMnO₃, $\lambda_{exc}=514.5\text{nm}$

J. Phys.: Condens. Matter, 16, 809 (2004).



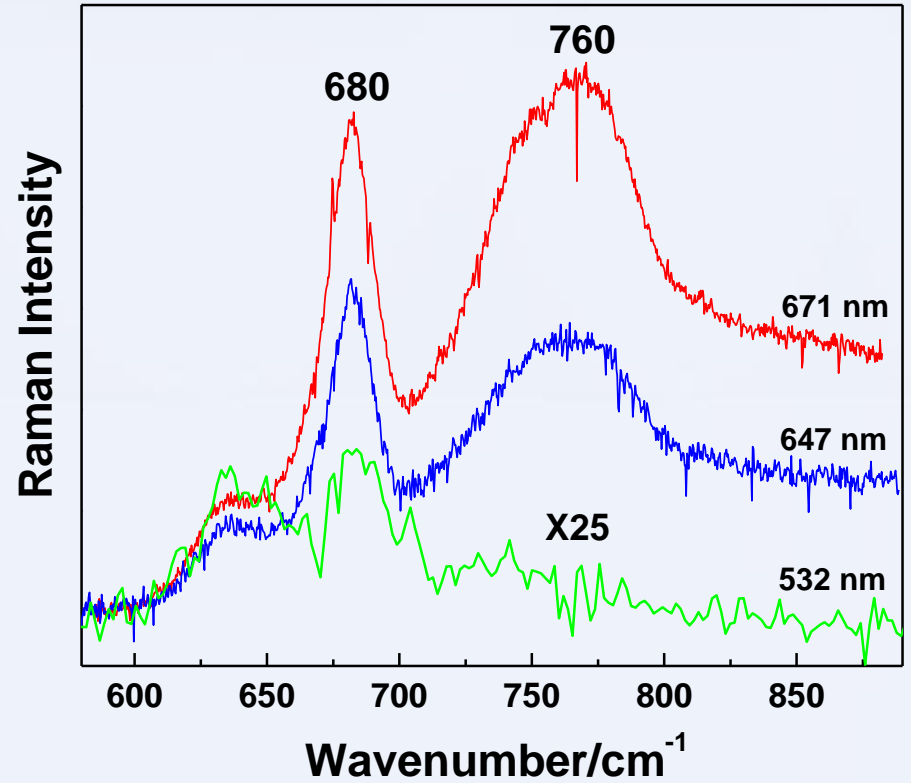
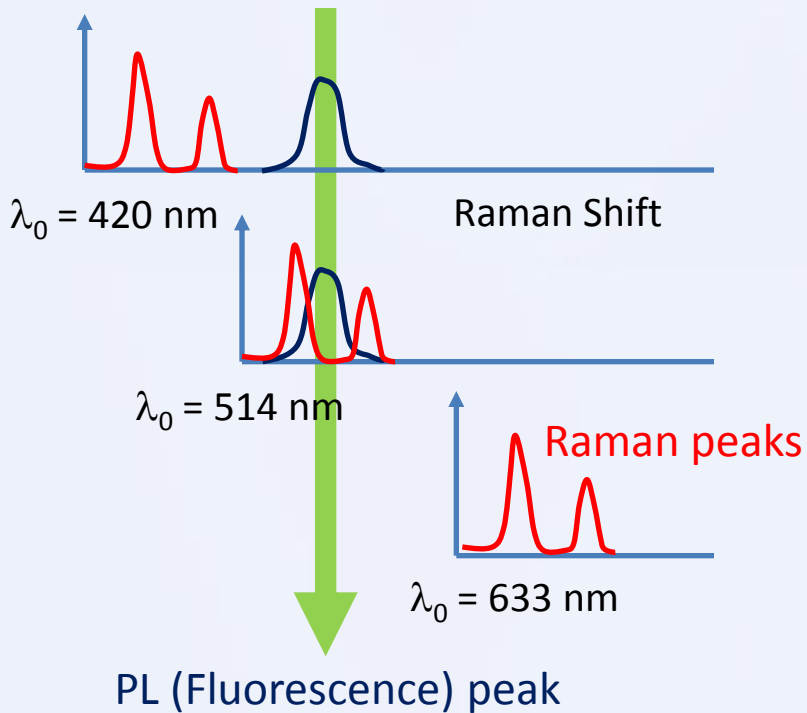
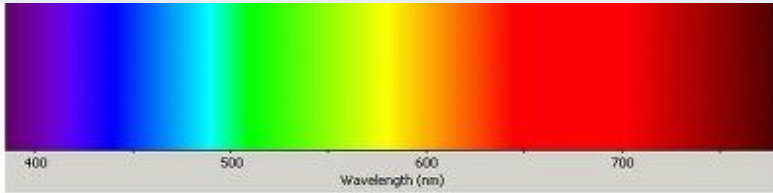
In cross polarization scattering @ 13 K



Confirms:

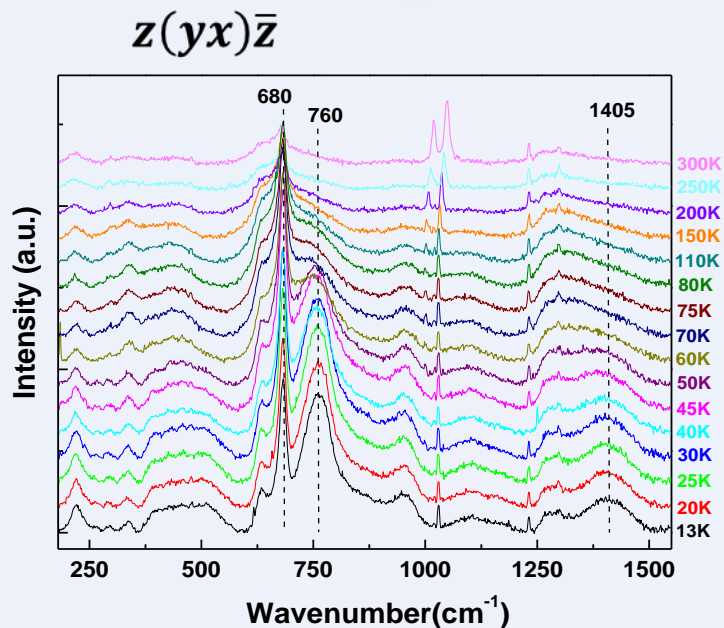
+ Raman origin, not PL!

+ Strongly resonant at 671nm (~1.8 eV)



III. Raman results: Spin excitation peaks

T dependence-Raman spectra of a hexagonal HoMnO_3 – cross polarization configurations

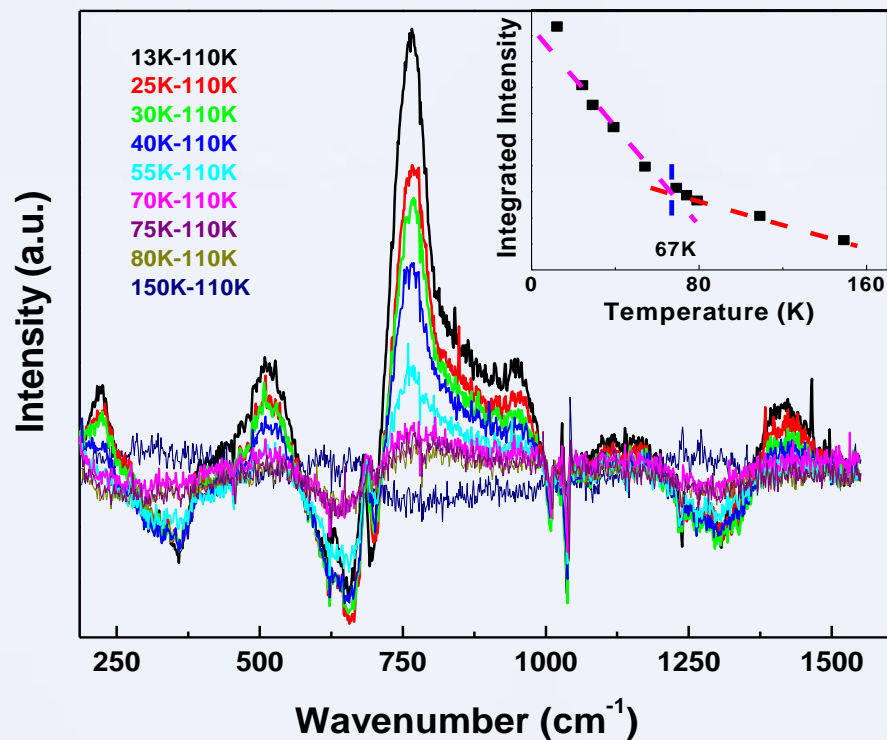


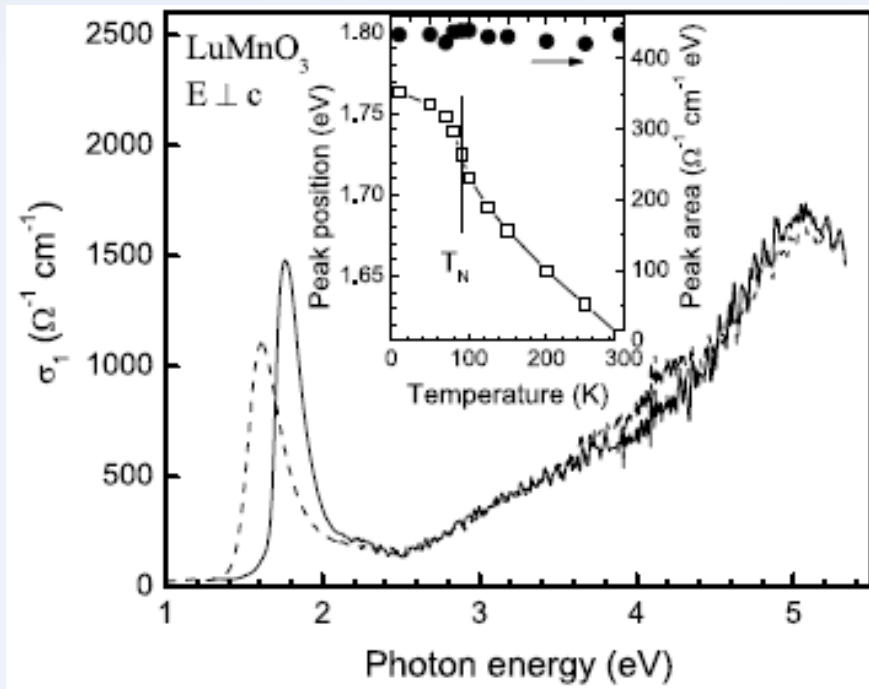
\Rightarrow shows an inflection point at
 $T=67\text{ K} \sim T_N$

\Rightarrow **Related with Magnetic Ordering**

$T \nearrow$: these broad peaks decrease $>$ the phonon modes, and are disappearing above T_N

T dependence of the difference spectra

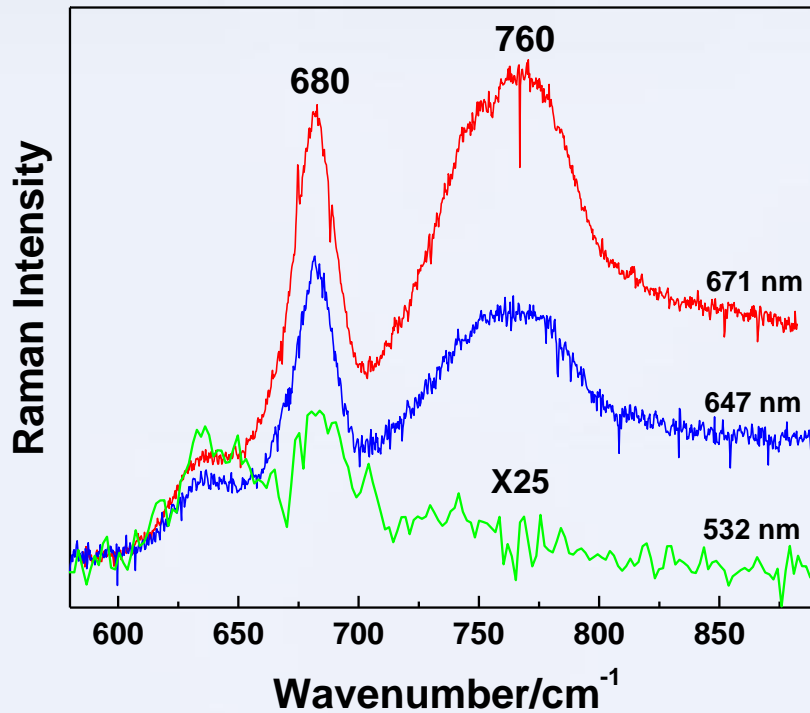




The electronic conductivity of LuMnO₃.
 Phys. Rev. Lett. **91**, 027203 (2003)

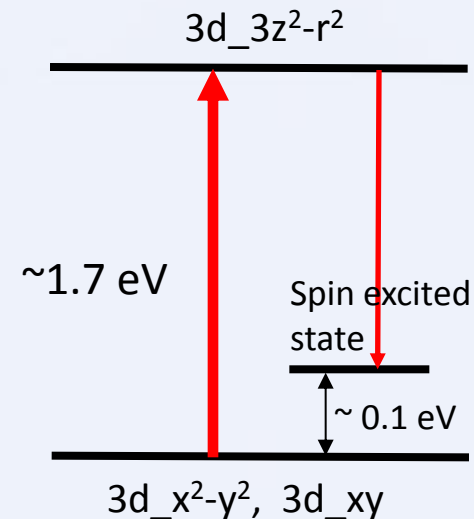
- Peak at ~ 1.7 eV: on-site Mn $d-d$ transition.
- Broad band centered at ~ 5 eV: charge transfer transition from the hybridized oxygen p levels to the Mn d levels.
- The on-site Mn $d-d$ transition at ~ 1.7 eV shifts ~ 0.15 eV with temperature.
- The shift is mainly caused by the effects of the exchange interaction between the Mn ions.

Resonant effect



Spin excitation peak (760 cm⁻¹) resonant effect is related to Mn d-d transition.

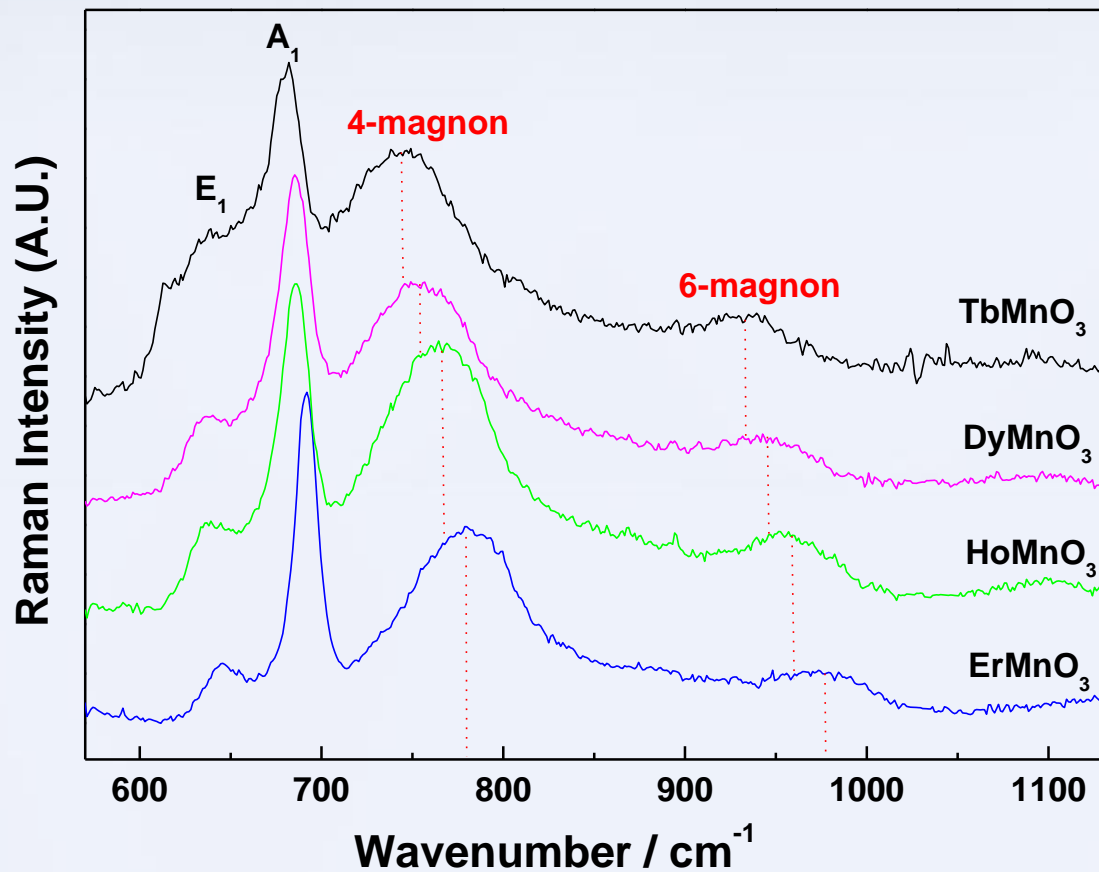
671nm	1.85eV
647nm	1.92eV
532nm	2.33eV



2. Rare-earth R dependence of $RMnO_3$ ($R=Tb, Dy, Ho, Er$) thin films

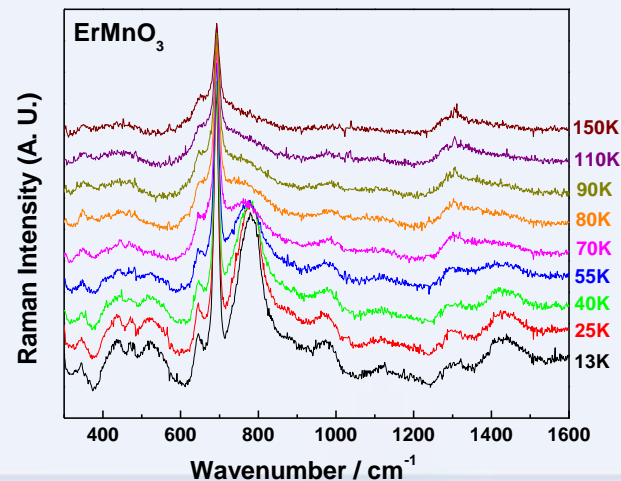
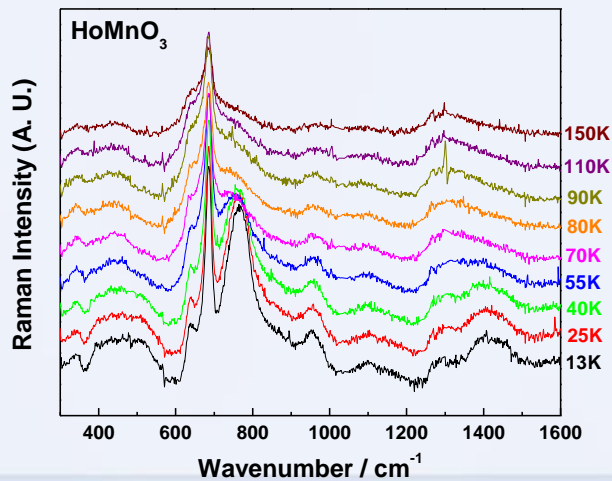
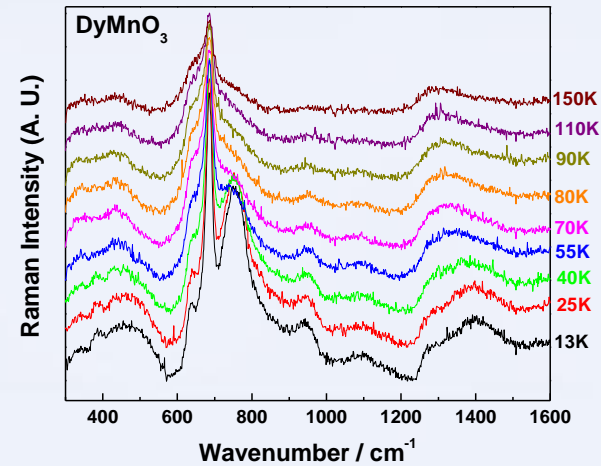
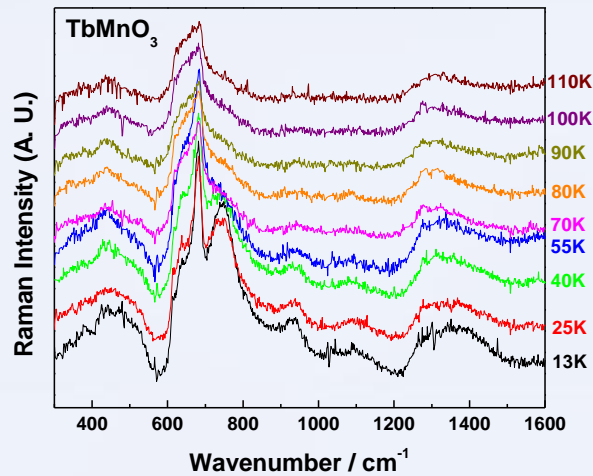
3. Raman scattering studies of hexagonal rare-earth $RMnO_3$ ($R = Tb, Dy, Ho, Er$) thin films, Nguyen Thi Minh Hien, Su-Young Oh, Xiang-Bai Chen, D. Lee, S.-Y. Jang, T. W. Noh, and In-Sang Yang, *Journal of Raman Spectroscopy* 42, 1774-1779 (2011).

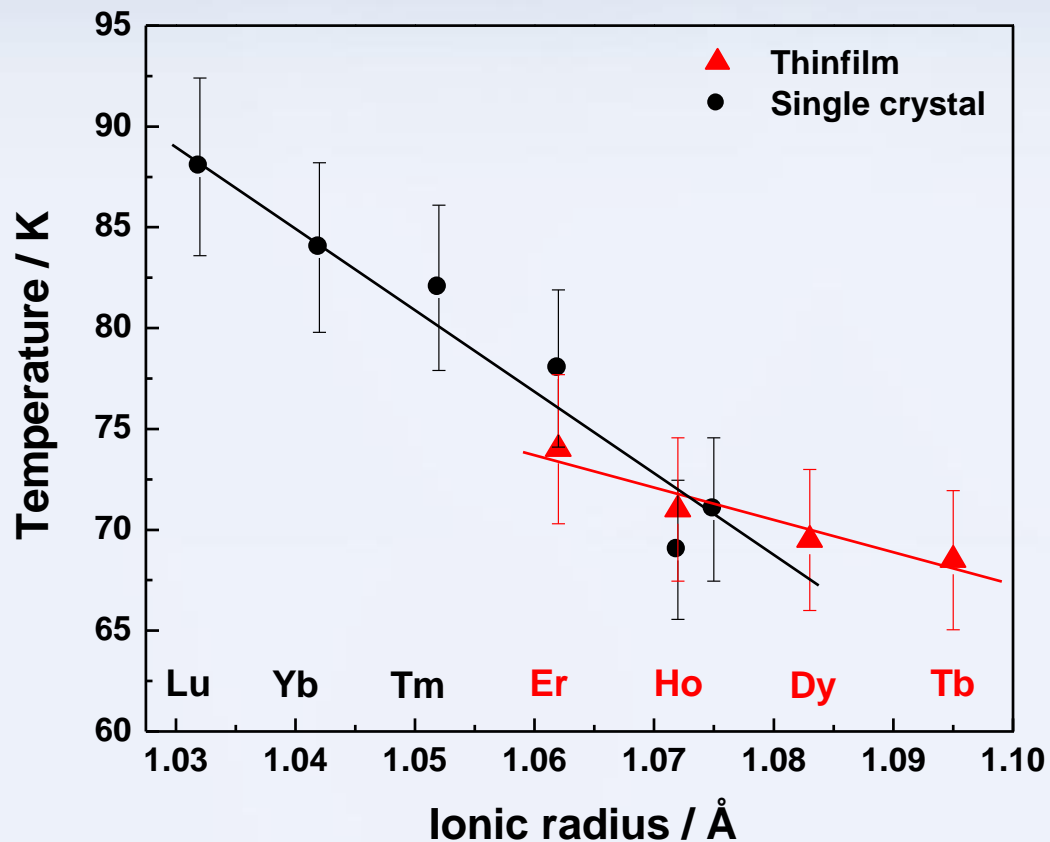
4. Spin exchange interactions in hexagonal manganites $RMnO_3$ ($R = Tb, Dy, Ho, Er$) epitaxial thin films, Xiang-Bai Chen, Nguyen Thi Minh Hien, D. Lee, S.-Y. Jang, T. W. Noh, and In-Sang Yang, *App.Phys.Lett.* 99, 052506 (2011).



Polarized Raman spectra of hexagonal RMnO_3 ($R = \text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$) thin films at 13 K obtained in the $Z(\text{XY})Z$ -configuration.

T dependent (13K – 150K) Raman spectra of hexagonal RMnO₃ (R = Tb, Dy, Ho, Er) thin films obtained in the Z(XY)Z- configuration.

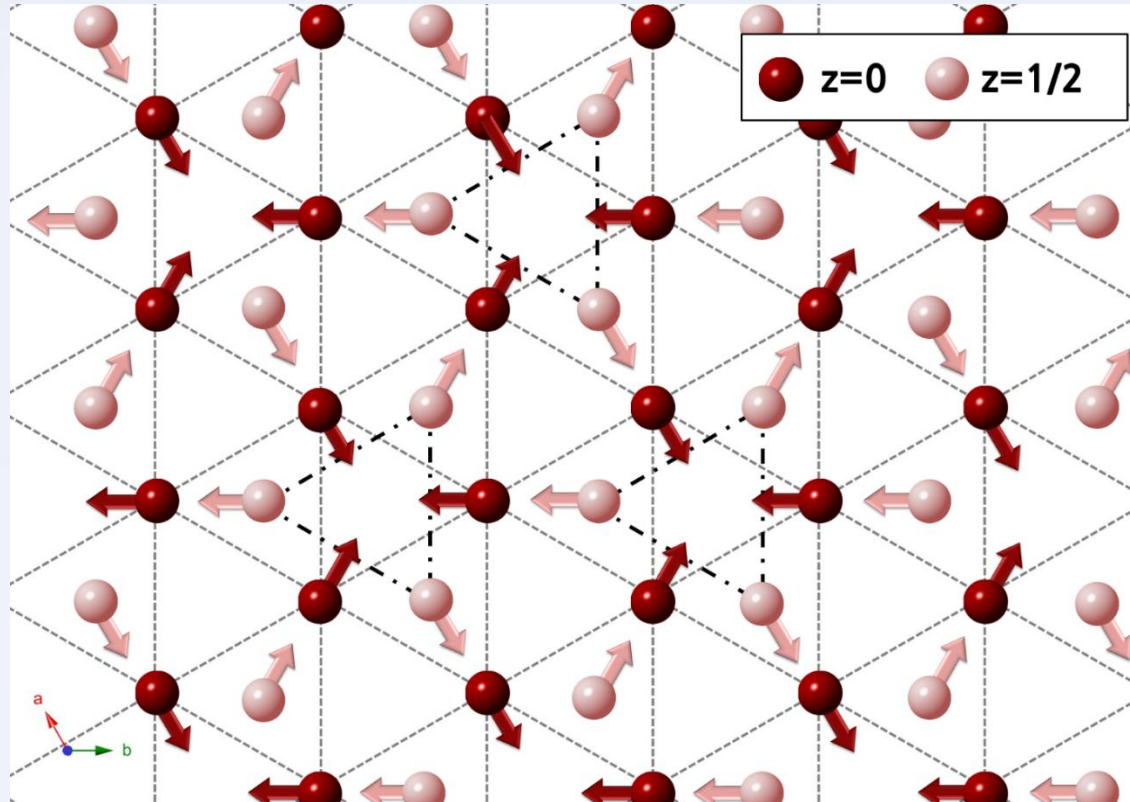




T_N - R dependence: our thin films
 $<$ single crystals.
 \Rightarrow the lattice constant a varies
 slower with the radius of R ion
 comparing with the single crystals

Fig 8: The R ionic radius dependence of T_N for our thin films and the single crystals

Mn-plane (@ $T < T_N$)



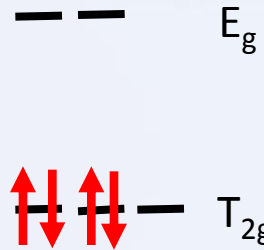
Below T_N the Mn spins order antiferromagnetically in a noncollinear 120° spin structure. (All-In-All-Out structure)

Spin states of some transition ions

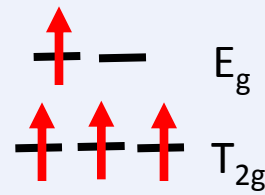
3d level electron	$3d^4$	$3d^5$	$3d^6$	$3d^7$
TM ions	Mn^{3+}	Mn^{2+}, Fe^{3+}	Fe^{2+}	Co^{2+}, Ni^{3+}
Weak Crystal Field High Spin State	$S=2$	$S=5/2$	$S=2$	$S=3/2$
Strong Crystal Field Low Spin State	$S=0$	$S=1/2$	$S=0$	$S=1/2$

Mn³⁺ (3d⁴)

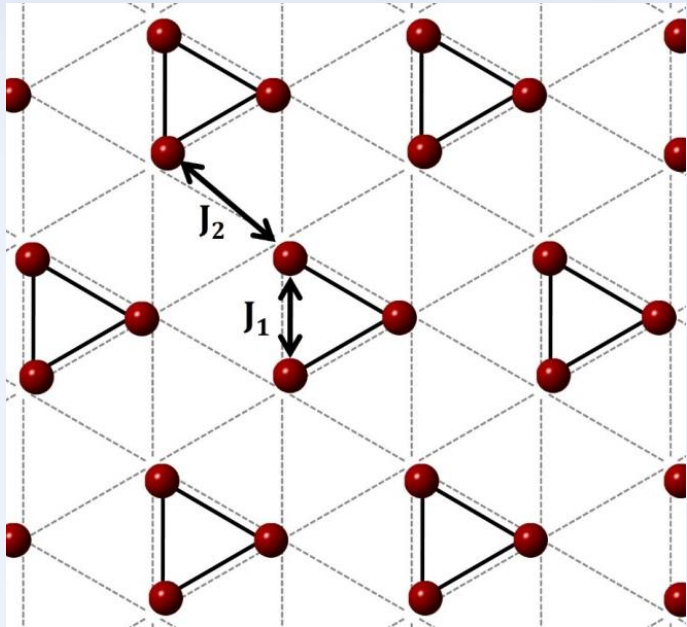
Strong Crystal Field
Low Spin State
S=0



Weak Crystal Field
High Spin State
S=2



III. Raman results: Spin excitation peaks



$$T < T_N$$

$$H = J_1 \sum_{\langle i,j \rangle} (S_i \cdot S_j) + J_2 \sum_{\langle i,k \rangle} (S_i \cdot S_k)$$

J_1 : intratrimer Mn-Mn spin interaction

J_2 : intertrimer Mn-Mn spin interaction

The 4-spin flipping magnon

4-magnon

spin flipping of 3 Mn³⁺ ions in 1 triangle and 1 Mn³⁺ ion in the neighboring triangle

$$13J_1 + 26J_2: 96.2\text{meV} \\ \sim 770 \text{ cm}^{-1}$$

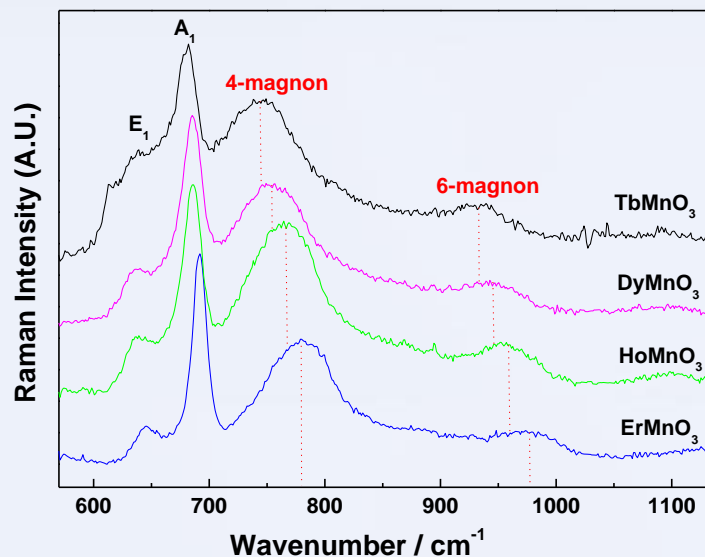
The 6-spin flipping magnon

6-magnon

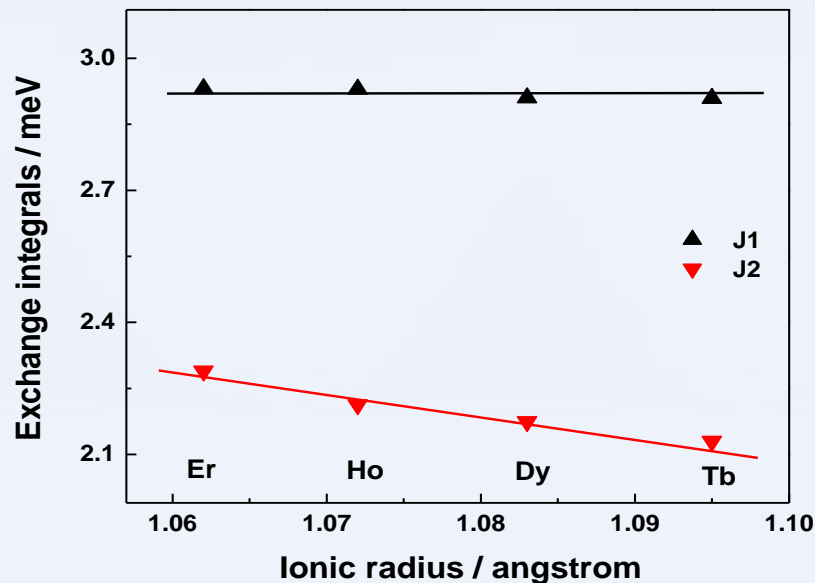
spin flipping of 6 Mn³⁺ ions in 2 neighboring triangle

$$18J_1 + 34J_2: 129.2\text{meV} \\ \sim 1000 \text{ cm}^{-1}$$

Raman spectra of hexagonal $RMnO_3$ ($R = Tb, Dy, Ho, Er$) thin films-13 K- cross configuration.



The R dependence J_1 and J_2 of hexagonal $RMnO_3$.



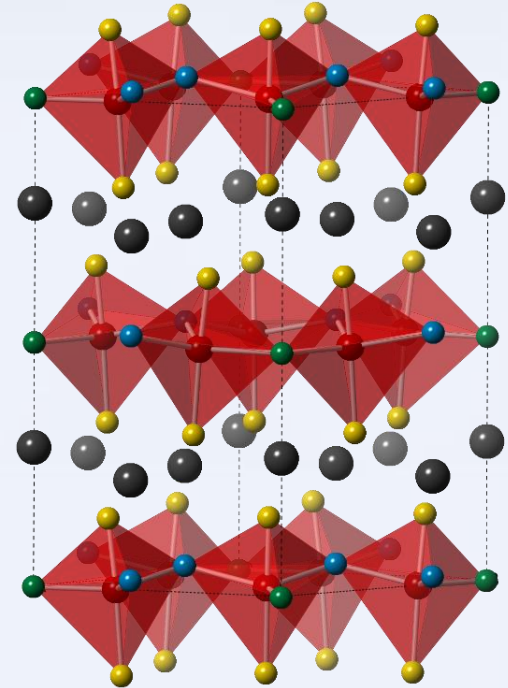
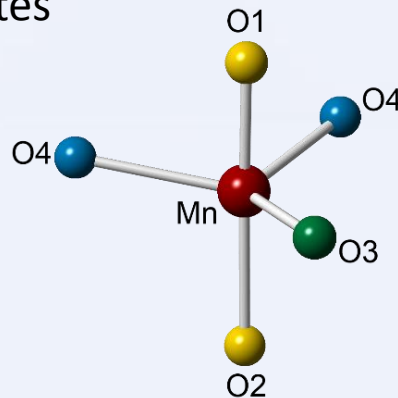
J_2 decreases systematically when the R ionic radius increases
 J_1 is nearly independent of R ionic radius.

➔ Raman Spectroscopy is an easier method than Neutron Scattering !

3. Spin excitations in Ga-doped hexagonal HoMnO_3 thin films

Sample - Hexagonal HoMnO_3

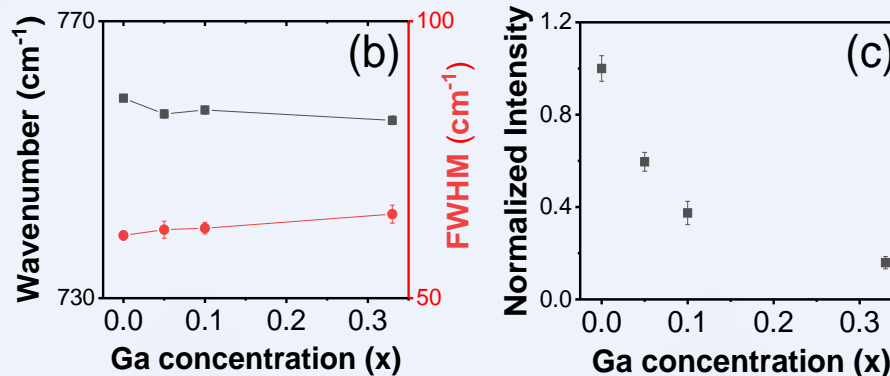
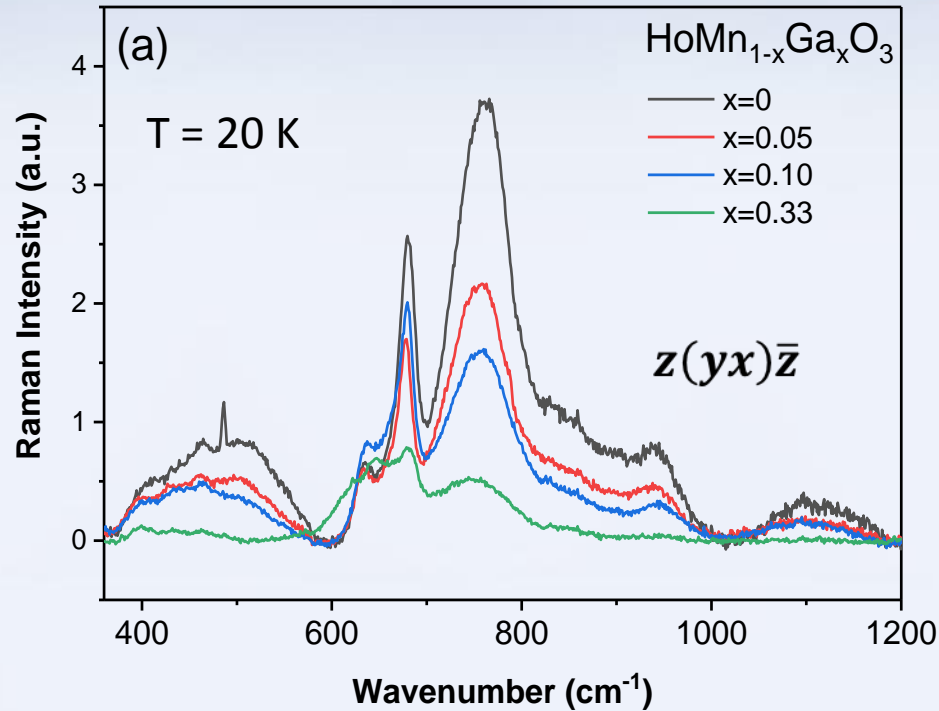
- Hexagonal HoMnO_3
 - Space group: $P6_3cm$
 - Layers of corner-sharing MnO_5 bipyramids
 - O3 and O4 : triangular base
 - O1 and O2 : apical sites



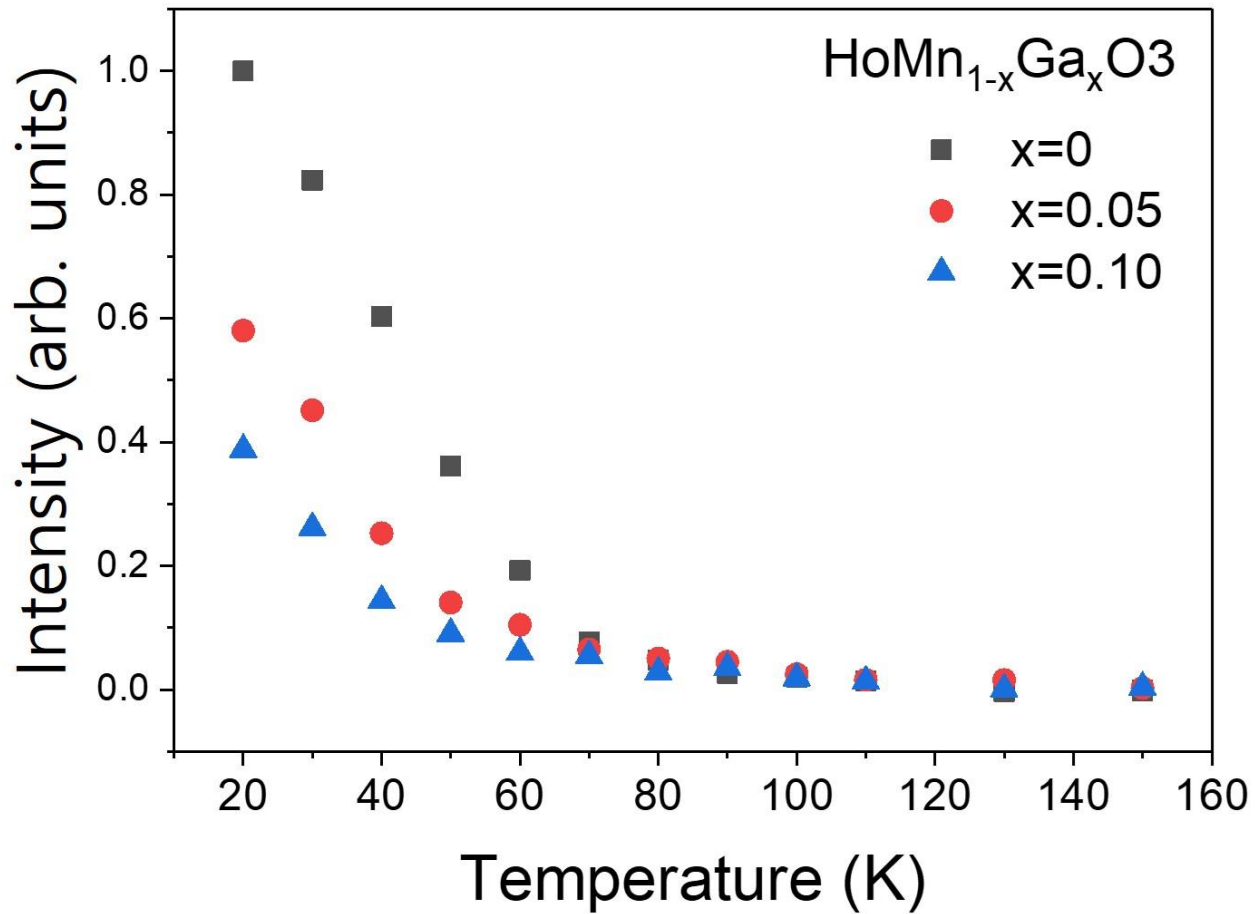
- $\text{HoMn}_{1-x}\text{Ga}_x\text{O}_3$ thin film ($x=0, 0.05, 0.1$ and 0.33)
- The samples were grown on $\text{Pt}(111)//\text{Al}_2\text{O}_3(0001)$ substrates by pulsed laser deposition techniques.

Results

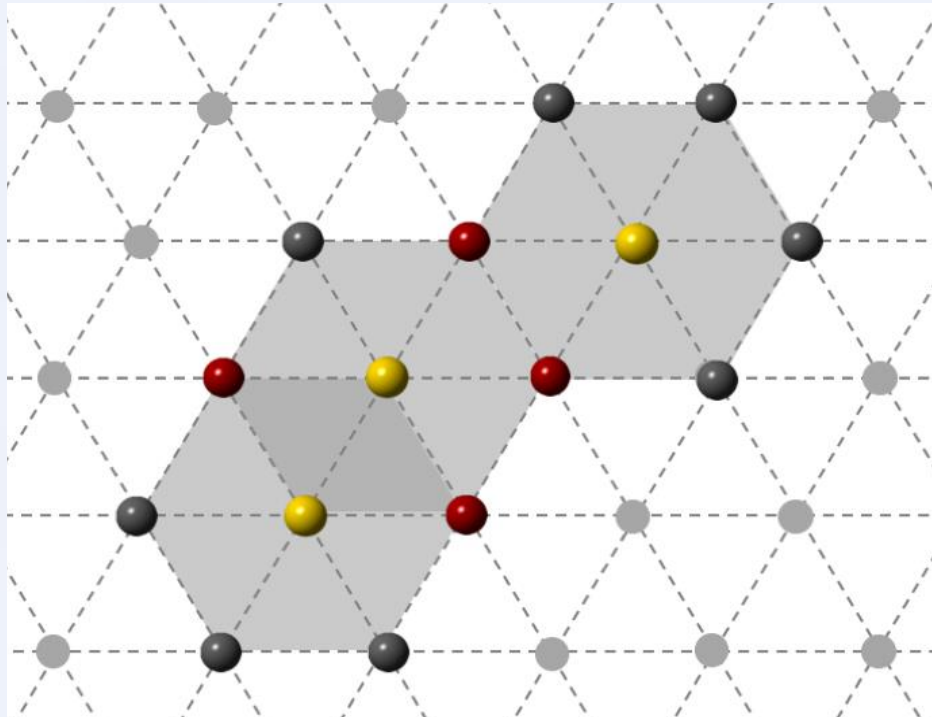
- Spin excitation energy (peak position) and FWHM values are weakly affected.
- Spin excitation intensity has $\sim 40\%$ and 60% decrease for 5% and 10% Ga doping, respectively.



Results



A Mn-Mn triangular network :



● Ga ion

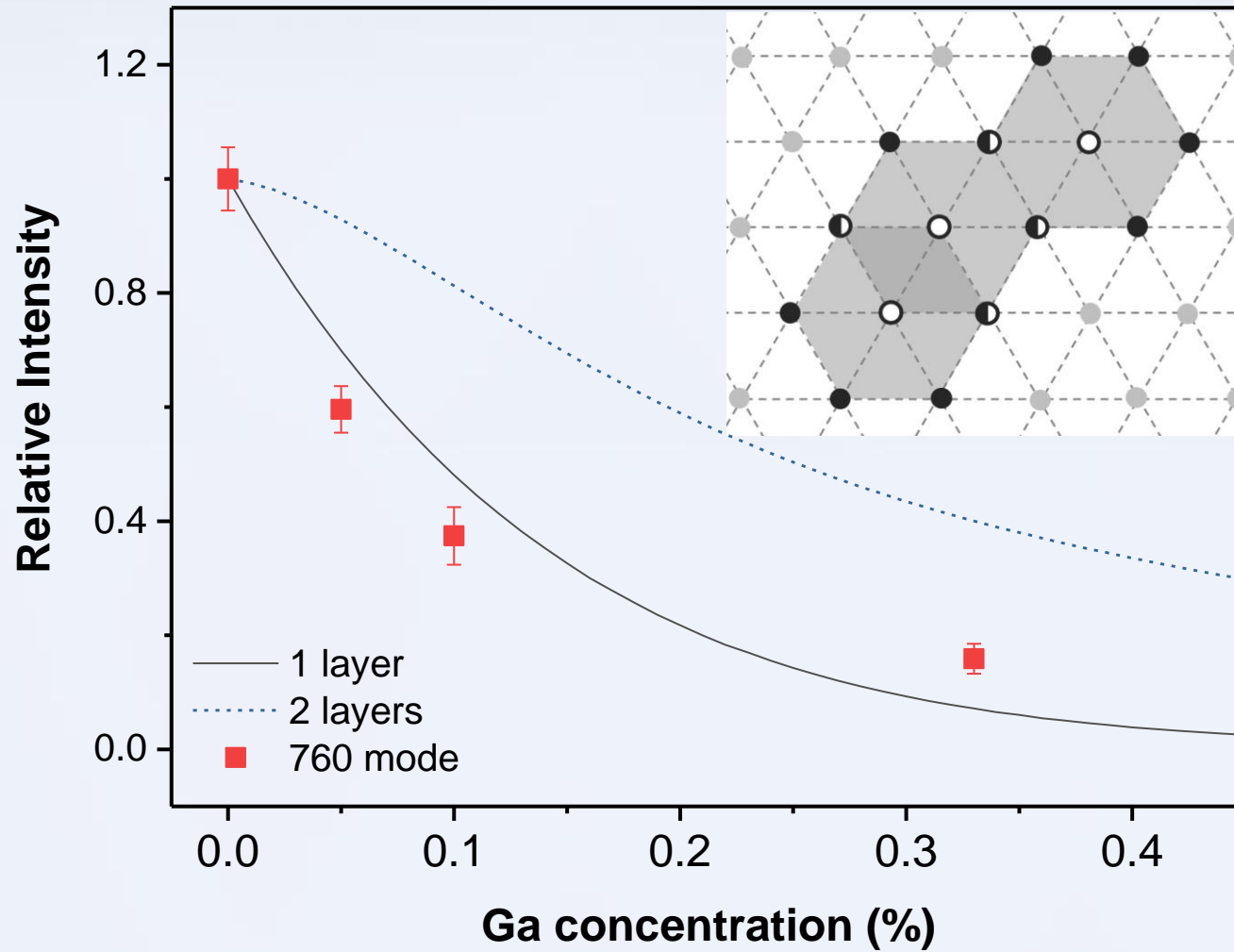
● Mn ion

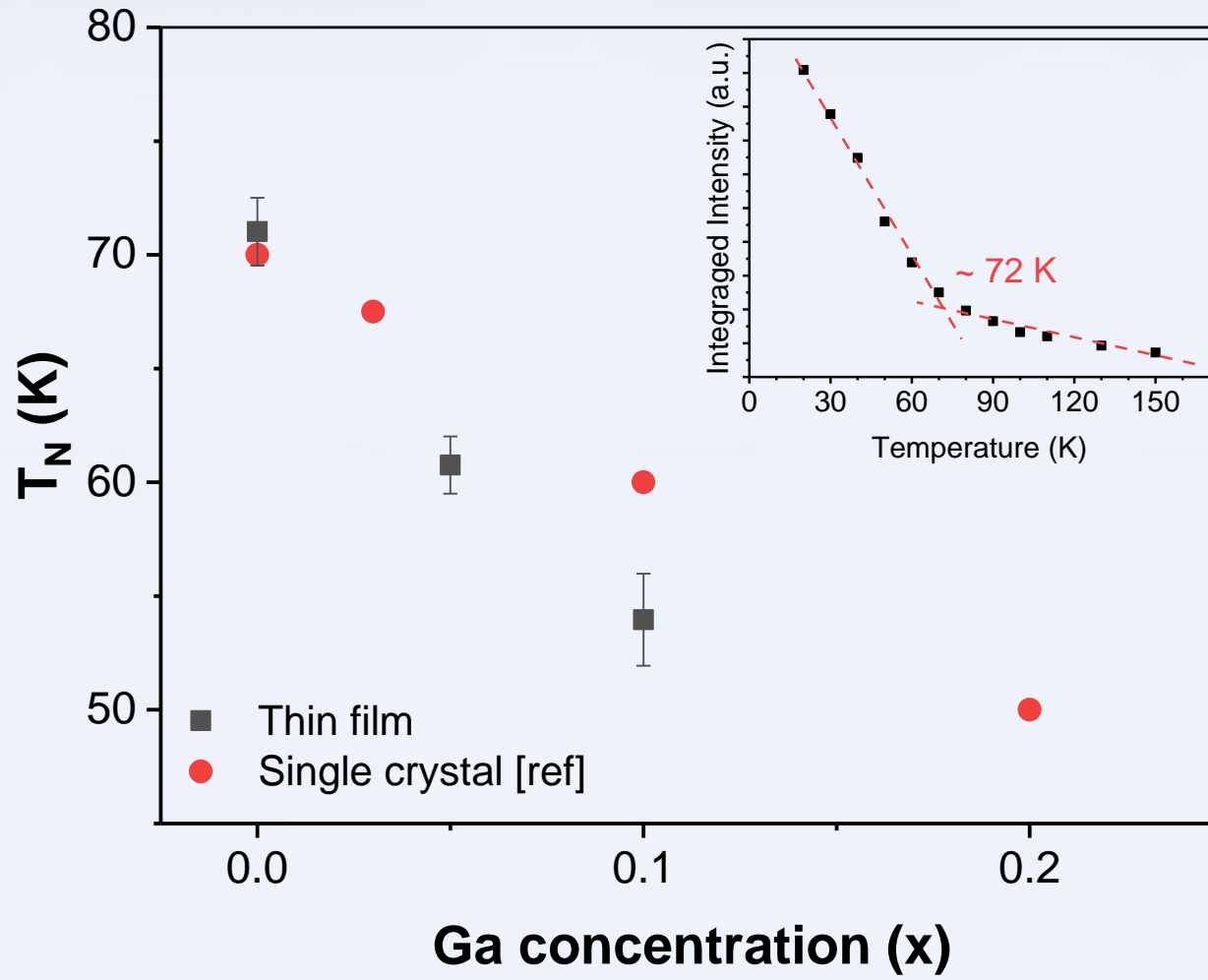


Analysis

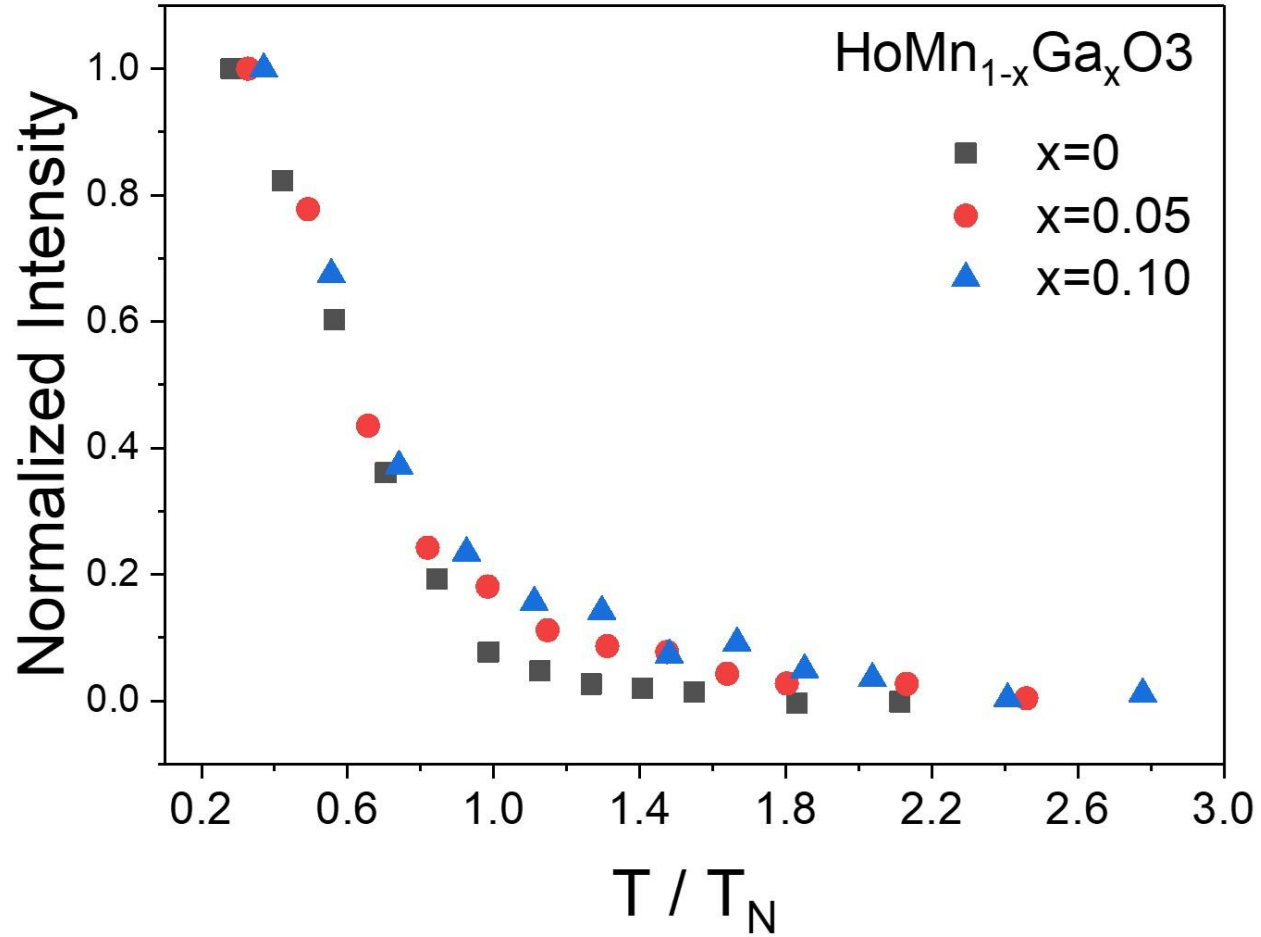
1. Consider 60×60 Mn ion lattices. (X)
2. Give each ion a number from 1 to 3600.
3. Choose 5%, 10%, and 33% random numbers.
4. Assume that the Ga ions enter a position corresponding to a random number.
5. Remove Mn ions “**affected**” by Ga ions.-Previous page
6. Count the number of remaining Mn ions. (Y)
7. Get the ratio Y/X .

Analysis





Results



Universal curve !

-> Spin excitation mechanism is not influenced by the Ga-substitution.

Substituting Ga ions **affect the neighboring Mn ions only.**

The spin excitations are **limited** in the Mn triangular network.

Summary:

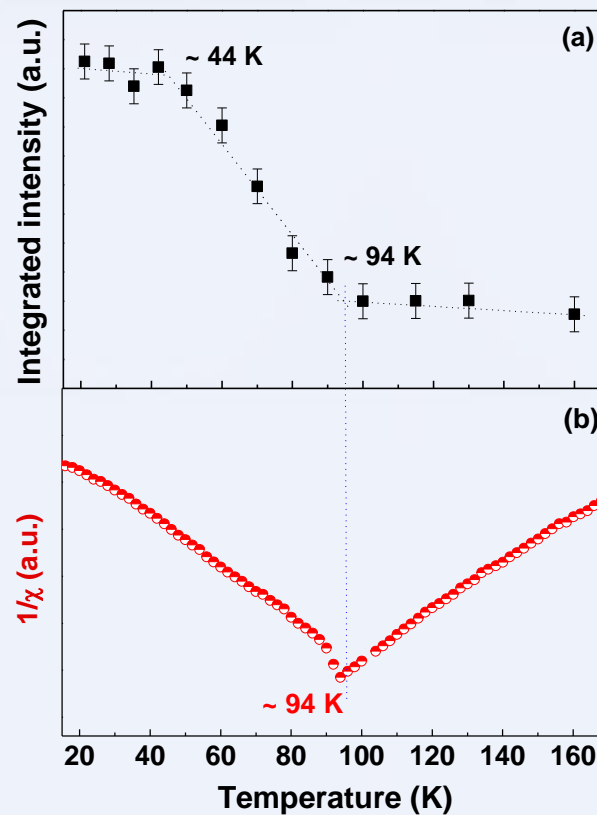
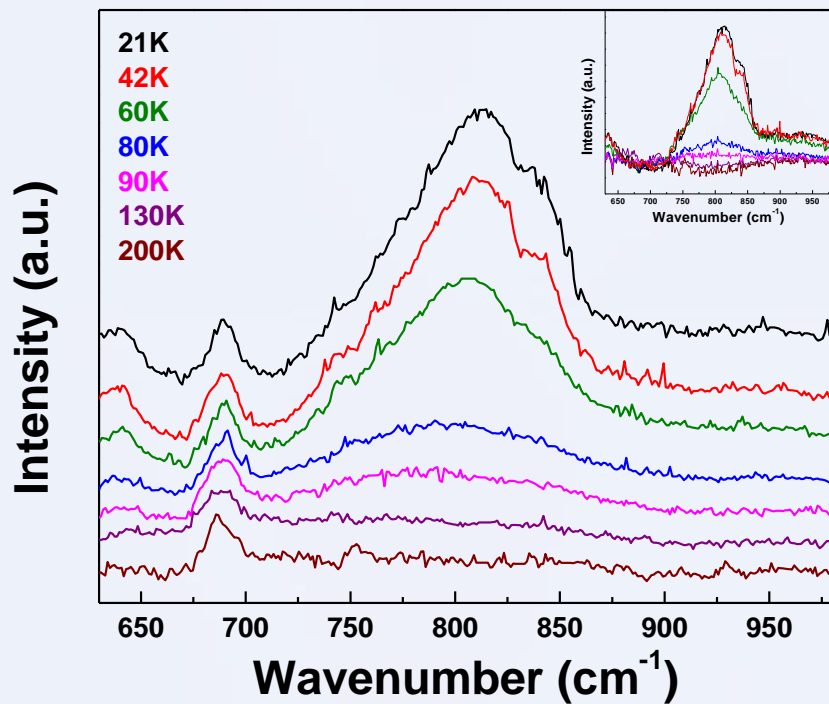
- We presented magnon scattering studies in $\text{HoMn}_{1-x}\text{Ga}_x\text{O}_3$ ($x = 0, 0.05, 0.10$ and 0.33) thin films.
- The spin-wave intensity has strong decrease by Ga substitution.
- We suggest a model of how Ga-ion substitution affects neighboring Mn ions.
- The effect of Ga-ion substitution is limited to the nearest Mn ions.
- The spin excitations seem to be **localized** in the Mn triangular network.

4. Raman study of magnetic phase transitions of hexagonal manganites single crystal LuMnO_3

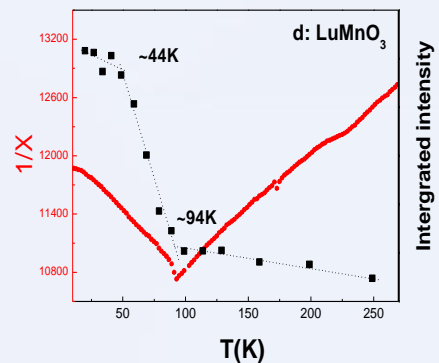
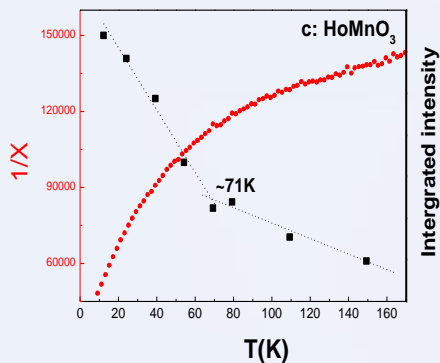
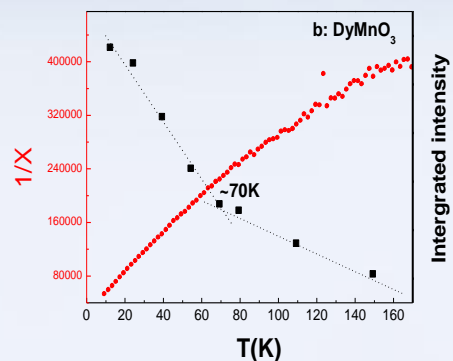
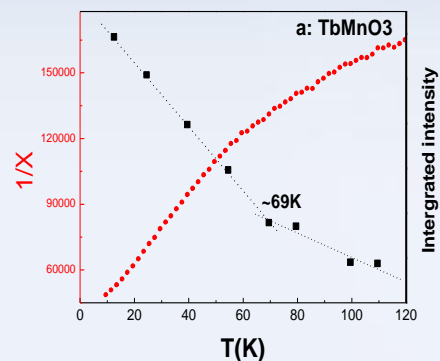
5. “Spin wave and spin flip in hexagonal LuMnO_3 single crystal”,
Xiang-Bai Chen, Peng-Cheng Guo, Nguyen Thi Huyen, Seung Kim,
In-Sang Yang, Xueyun Wang, Sang-Wook Cheong,
Appl. Phys. Lett. **110**, 122405 (2017).

III. Raman results: Spin excitation peaks

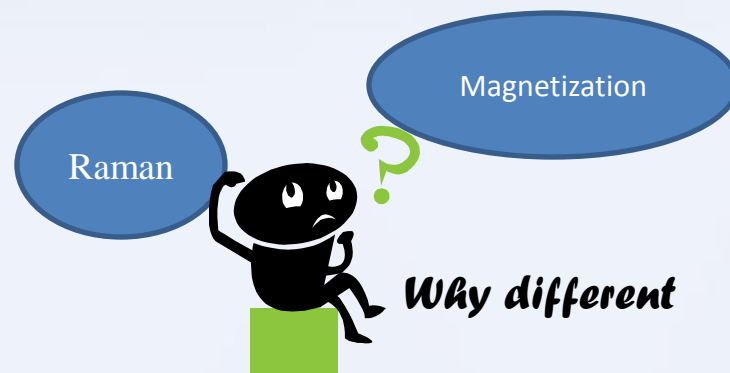
T dependent (20K – 200K) Raman spectra of hexagonal LuMnO_3 single crystal in the cross configuration



Raman vs. magnetization experiment of hexagonal $RMnO_3$



Thin films: $R=Tb, Dy, Ho$
Single crystal: $R=Lu$



Raman: probes the magnons in Mn^{3+} sublattices. Measure the skin-depth of the thin film, thus, it investigate the intrinsic properties of the thin film.

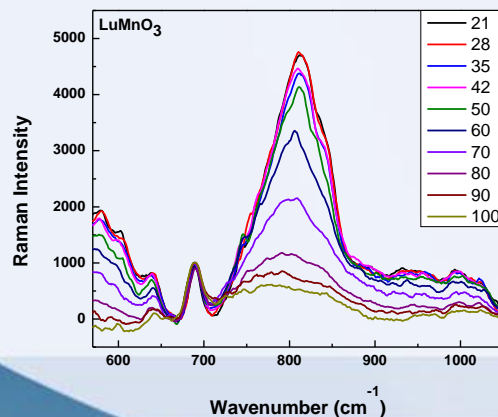
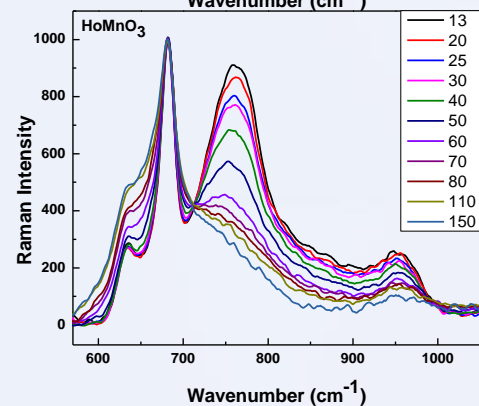
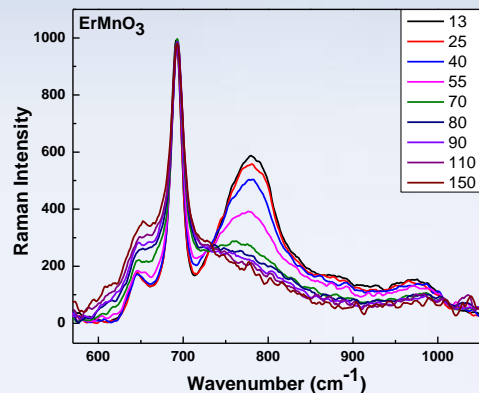
Magnetization: Strong paramagnetic contribution from the f-electron of the rare-earths dominate the magnetization data. Substrate also measured at the same time.

Magnon Raman spectra selectively probes magnons in the Mn^{+3} sublattices.
-> Effective to study the magnetic phase transitions due to Mn-spin ordering.

5. Two-Dimensional Correlation Analysis and Principal Component Analysis

6. Correlation between magnon and magnetic symmetries of hexagonal $RMnO_3$ ($R = Er, Ho, Lu$), Thi Minh Hien Nguyen, Thi Huyen Nguyen, Xiang-Bai Chen, Yeonju Park, Young Mee Jung, D. Lee, T.W. Noh, Sang-Wook Cheong, and In-Sang Yang, *Journal of Molecular Structure* 1124, 103-109 (2016).

II. 2D COS & PCA



PRETREATMENT OF DATA

Normalize the reference peak at $\sim 680\text{cm}^{-1}$ to 1000cts
Noise reduction: Savitzky- Golay smoothing method (13 points)

PCA

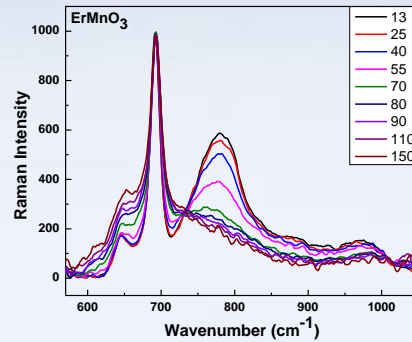
The PCA was performed using the PLS Toolbox ver. 7.3 (Eigenvector Research, Inc.) for Matlab (The MathWorks Inc.)

2D COS

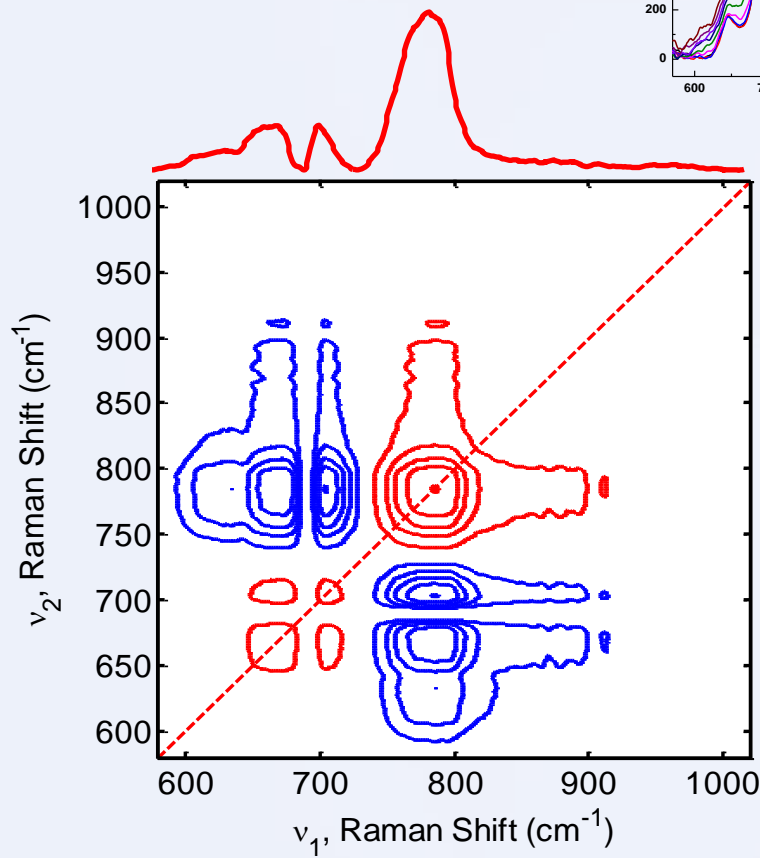
The 2D correlation spectra were obtained in Matlab R2013b

IV. Support from 2D COS & PCA: ErMnO_3

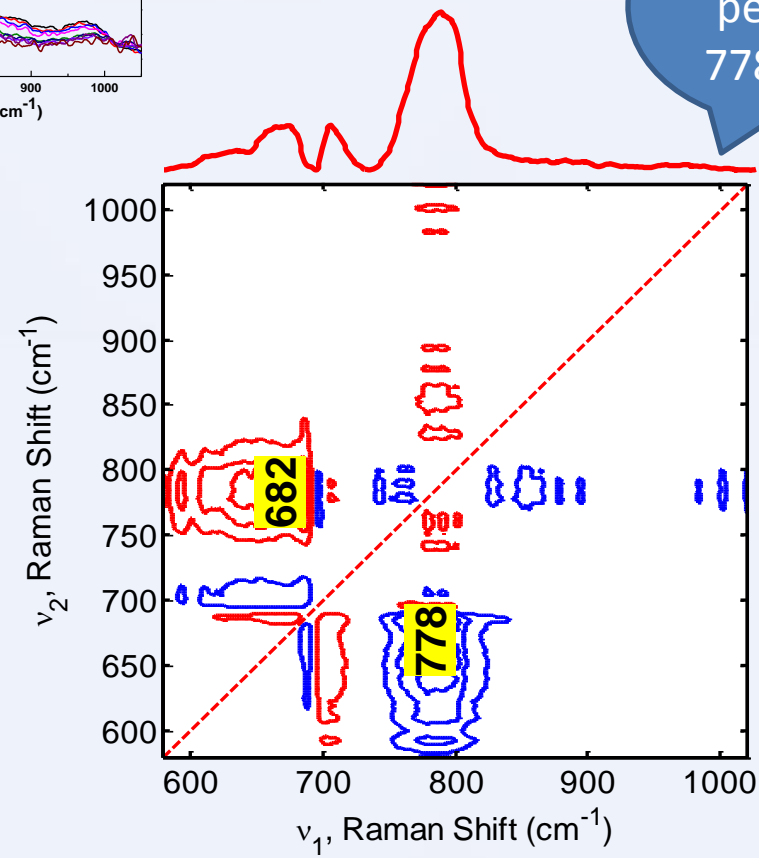
ErMnO_3



Only one peak at 778 cm⁻¹

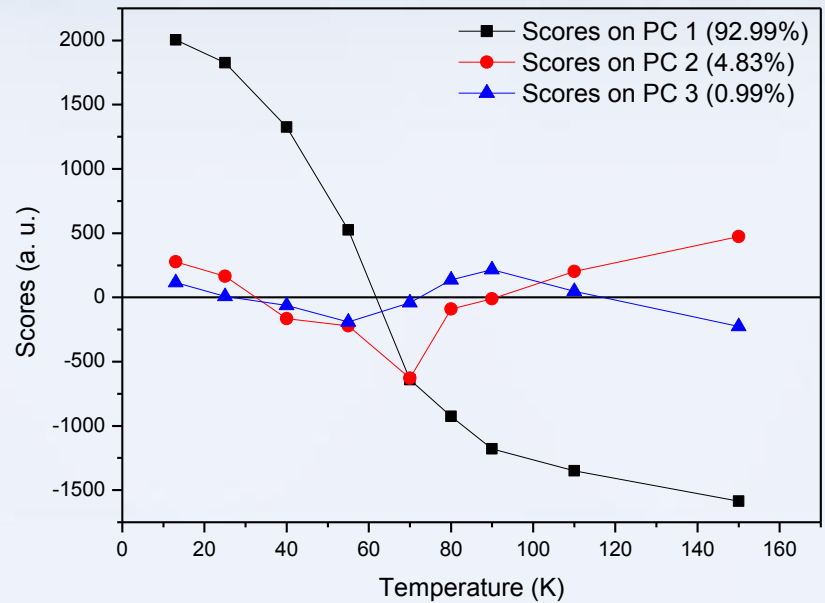
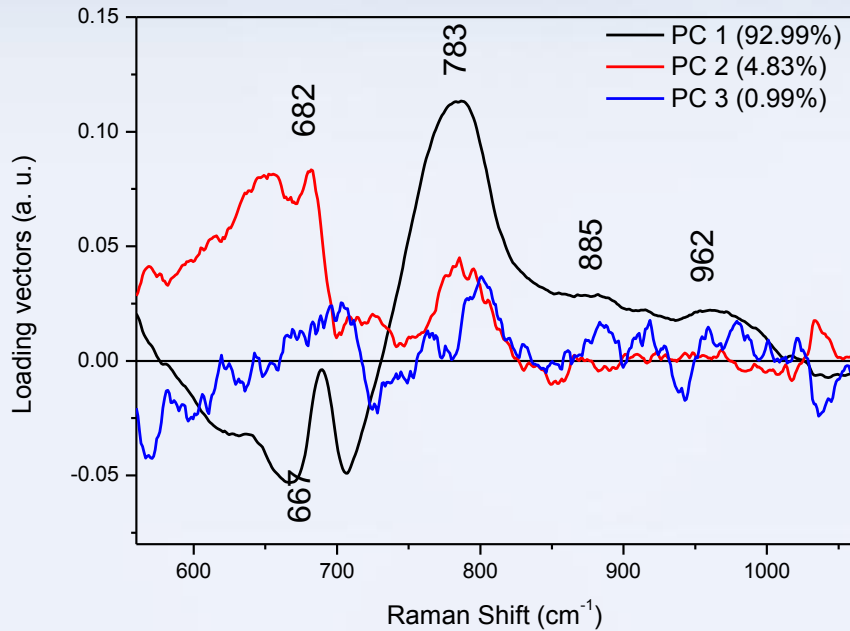


Synchronous



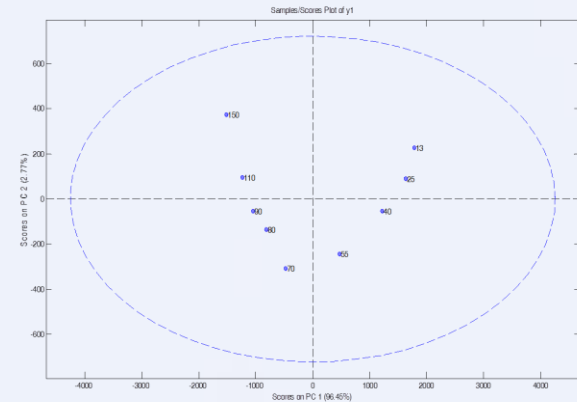
Asynchronous

IV. Support from 2D COS & PCA: ErMnO_3

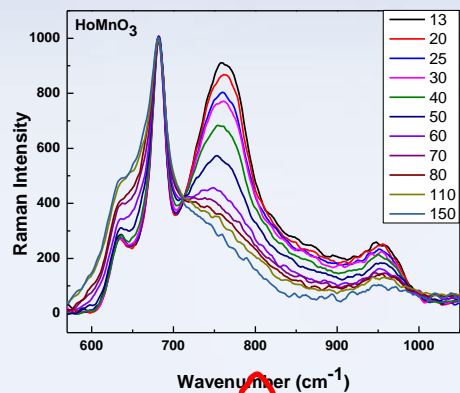


PC3: too noisy

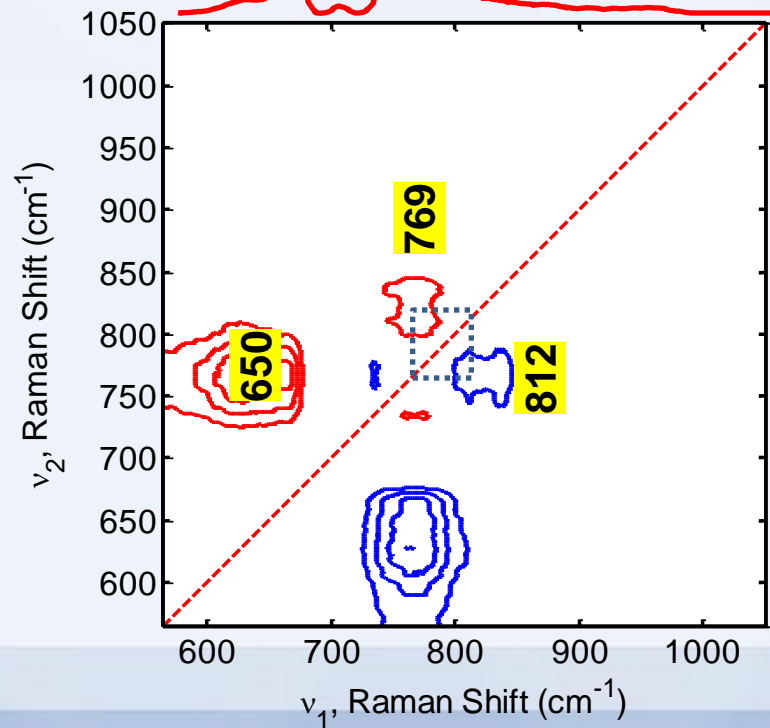
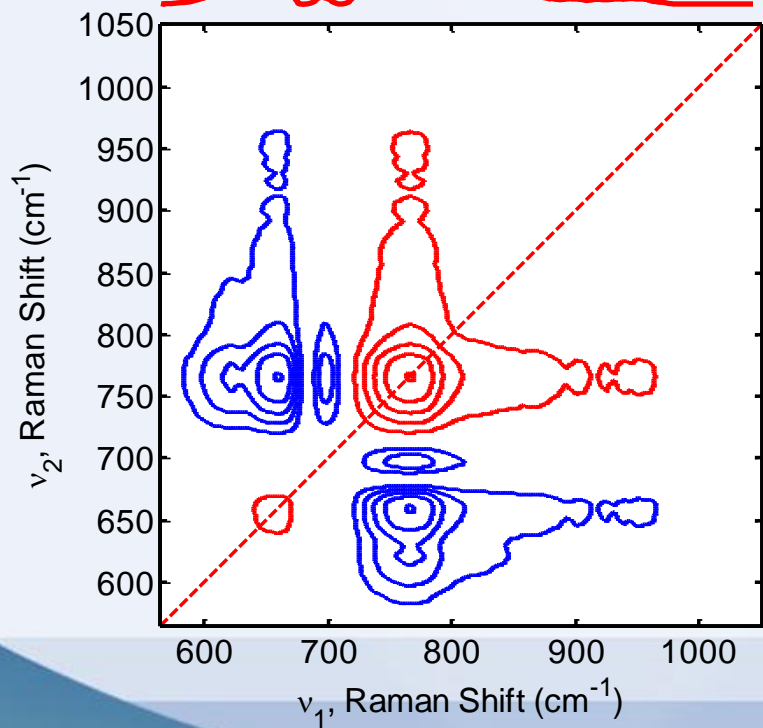
From PC1 & PC2: Broad magnon band of ErMnO_3 at $\sim 778\text{cm}^{-1}$ consists of a single peak



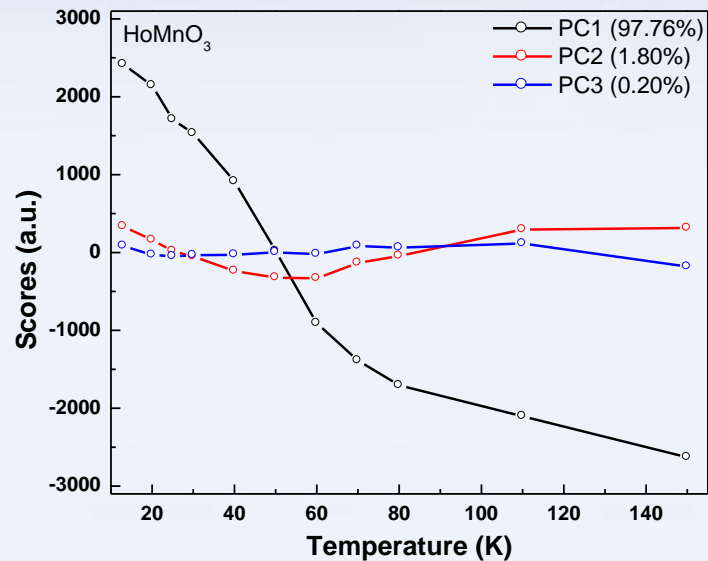
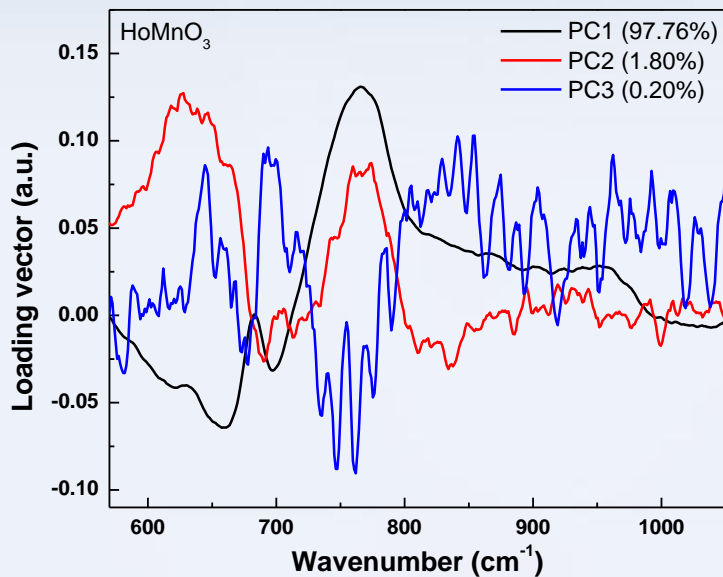
IV. Support from 2D COS & PCA: HoMnO_3



Broad band consist of 2 overlap peaks at 769 & 812 cm^{-1}

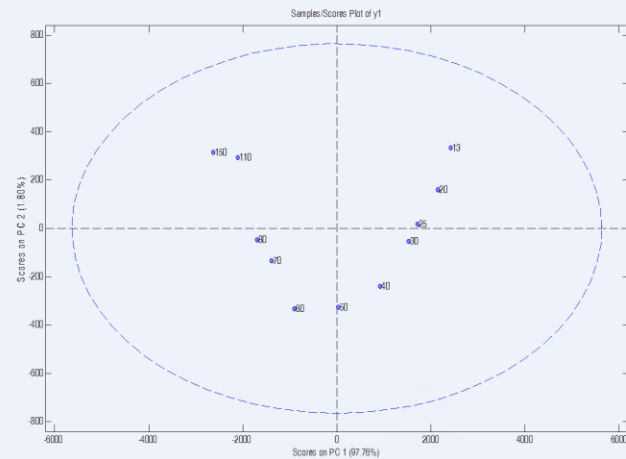


IV. Support from 2D COS & PCA: HoMnO_3

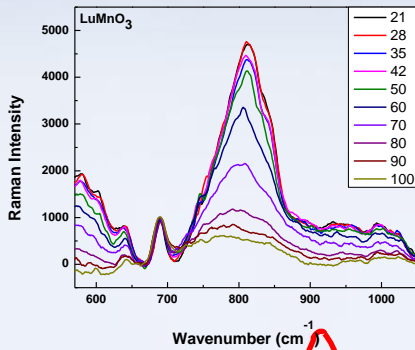


PC3: too noisy

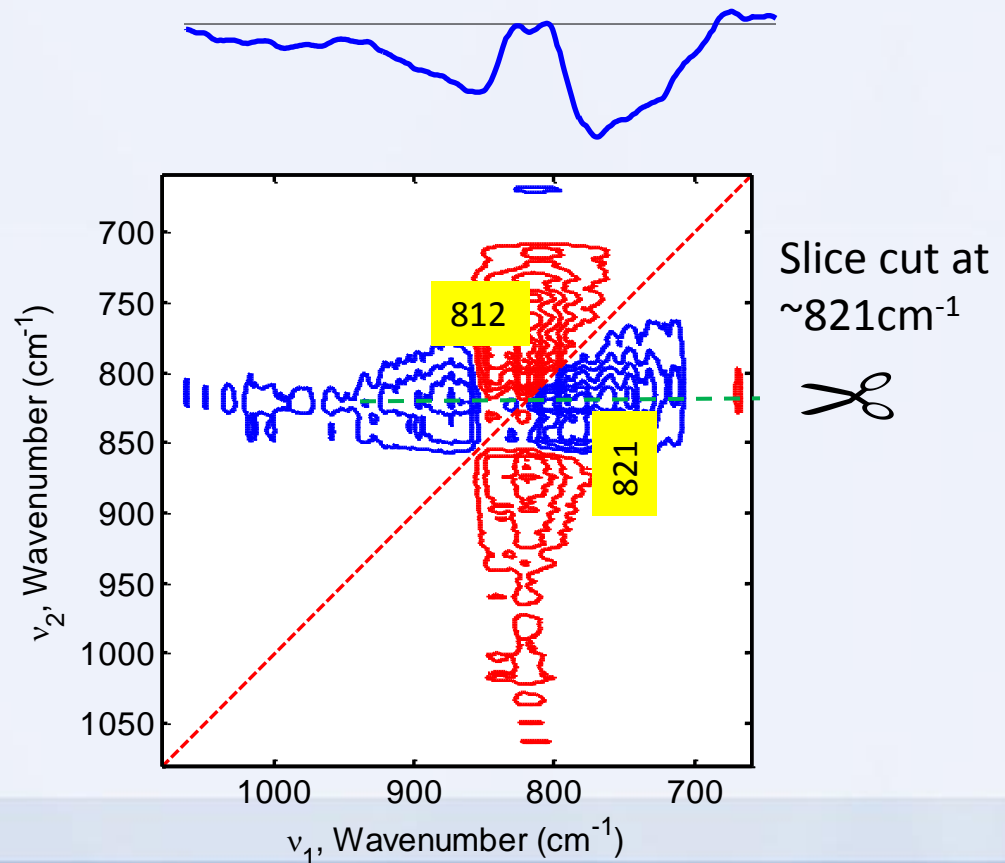
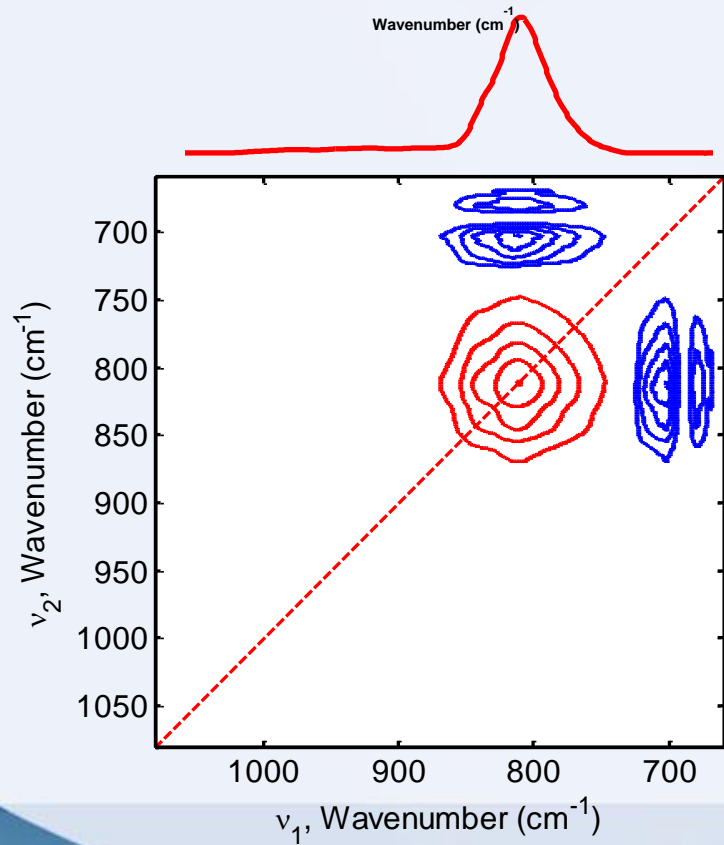
From PC1 & PC2: Difficult to realize the broad band of HoMnO_3 at $\sim 760\text{cm}^{-1}$ consists of 1 or 2 overlap peaks



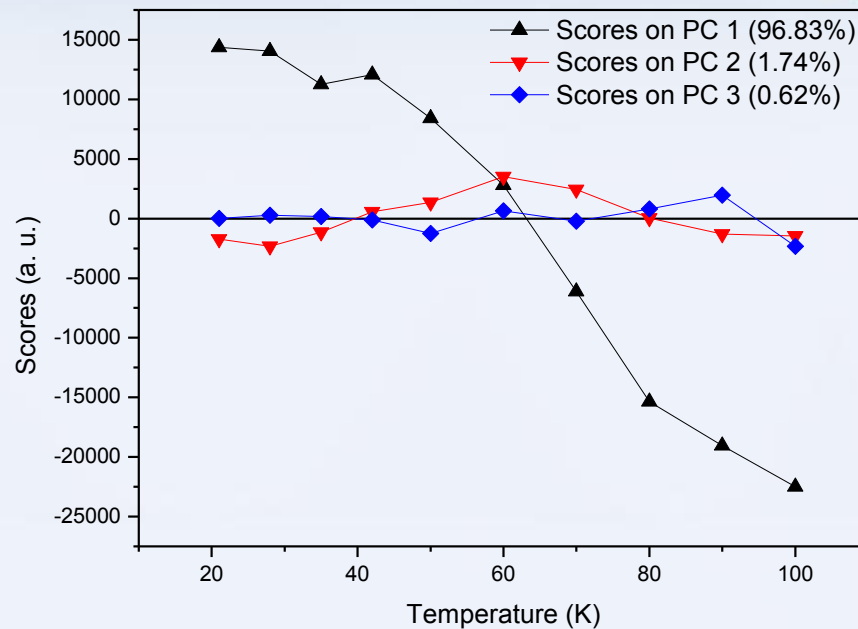
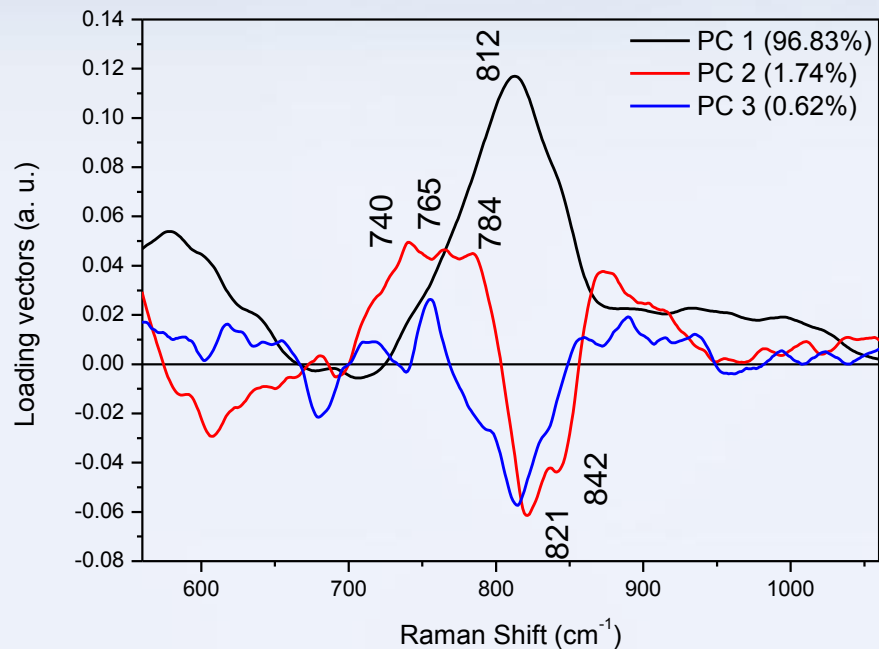
IV. Support from 2D COS & PCA: LuMnO_3



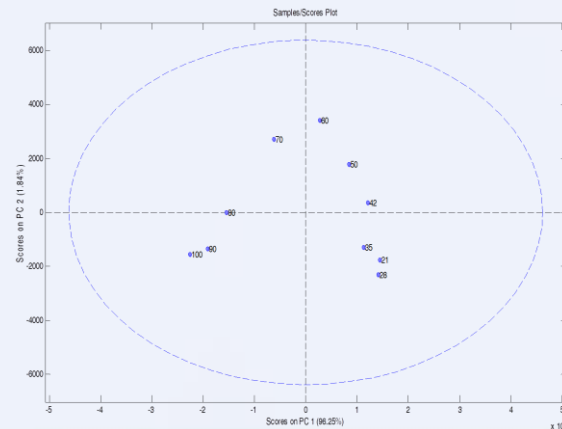
Asynchronous spectra is too noisy, only realize 2 peaks at 812 & 821cm⁻¹



IV. Support from 2D COS & PCA: LuMnO_3

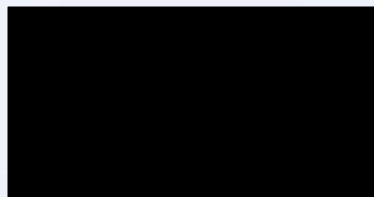
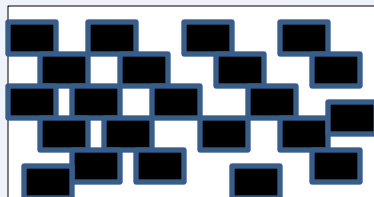
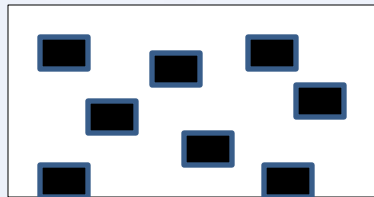


From PC1 and PC2: observed 6 peaks overlap in the broad magnon peak.

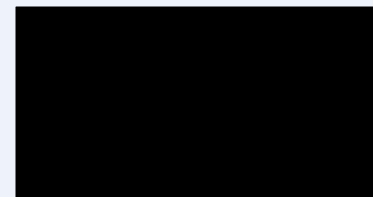
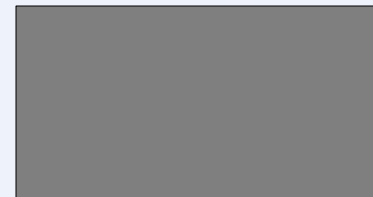
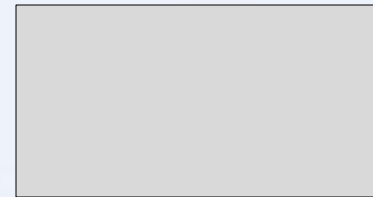


Overlap peaks vs Single peak

- Overlapped peaks -

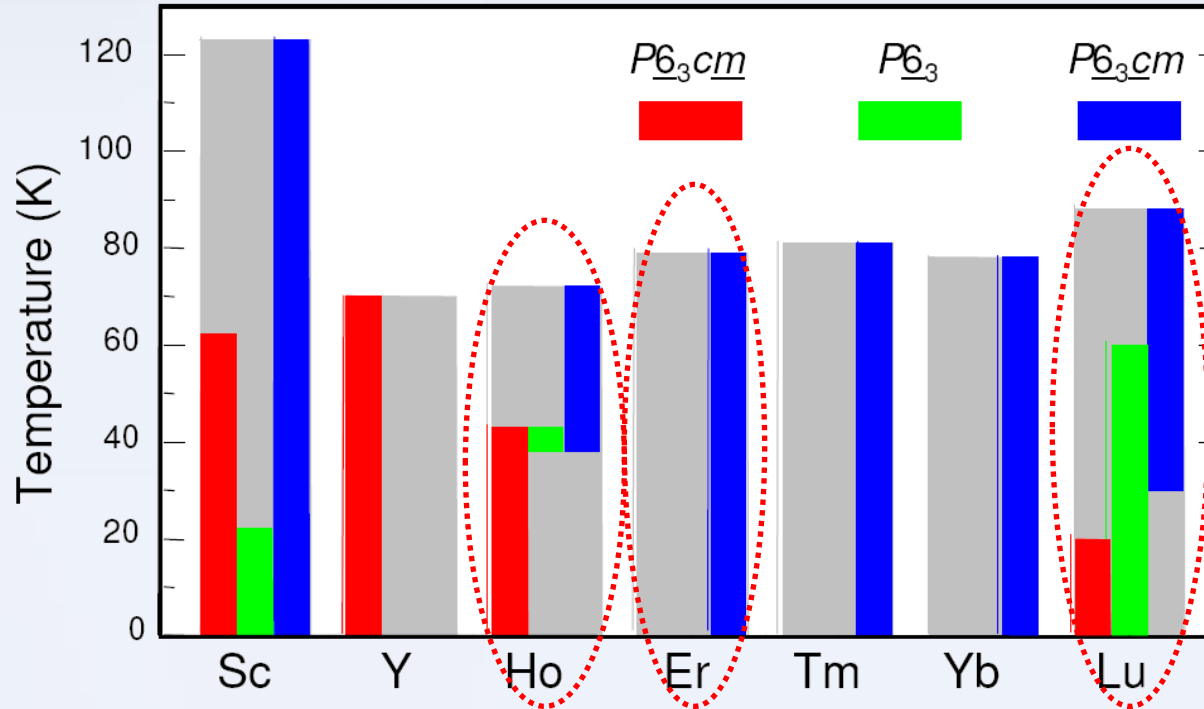


- Single peak -



DISCUSSION

Symmetry of the hexagonal manganites $RMnO_3$

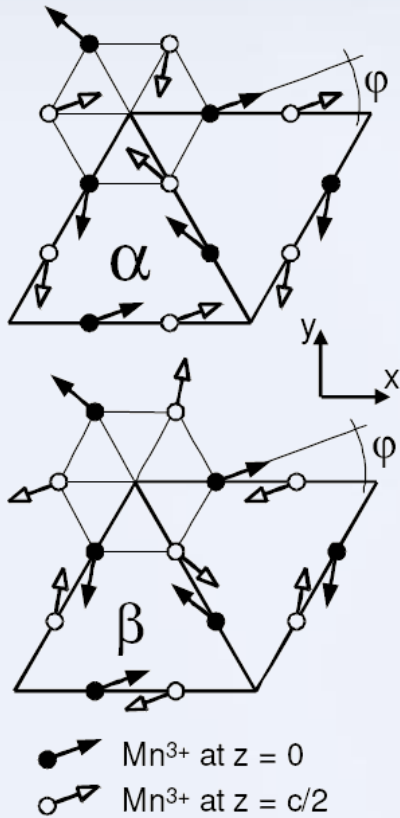


Should be correlated with the number of overload peaks in the broad magnon peak?



The symmetry of $LuMnO_3$ and $HoMnO_3$ are more complicated than that of $ErMnO_3$

RMnO₃ : Magnetic structure



$T < T_C$:
 ferroelectric
 ↓
 Symmetry :
 $P6_3cm$

 $T < T_N$:
 antiferromagnetic
 ↓

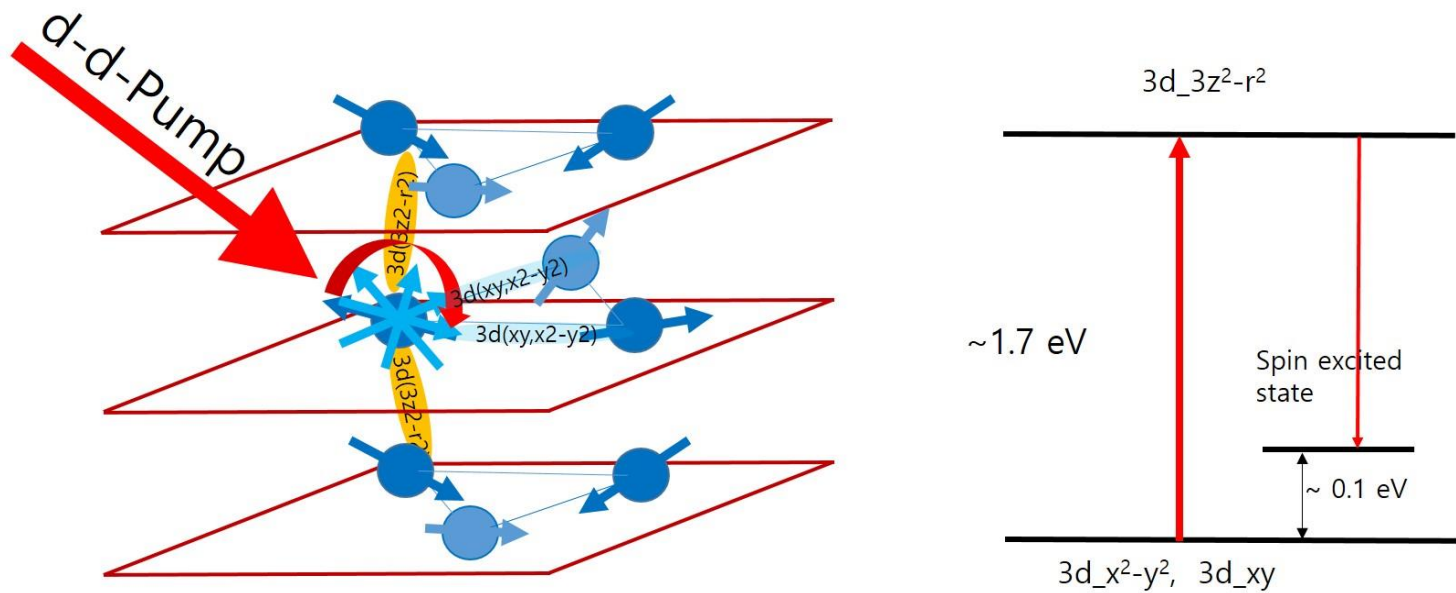
$\alpha_1 (\varphi = 0^\circ)$: $\chi_{yyy} = -\chi_{yxx} = -\chi_{xyx} = -\chi_{xxy}$	$P\bar{6}_3cm$	$\beta_1 (\varphi = 0^\circ)$: $\chi_{xyz} = \chi_{xzy} = -\chi_{yxz} = -\chi_{yzx}$	$P6_3cm$
$\alpha_2 (\varphi = 90^\circ)$: $\chi_{xxx} = -\chi_{xyy} = -\chi_{yxy} = -\chi_{yyx}$	$P\bar{6}_3cm$	$\beta_2 (\varphi = 90^\circ)$: $\chi_{zxx} = \chi_{zyy}, \chi_{xxz} = \chi_{xzx} = \chi_{yyz} = \chi_{yzy}, \chi_{zzz}$	$P6_3cm$
$\alpha_\varphi (0^\circ < \varphi < 90^\circ)$: $\chi(\alpha_1) \oplus \chi(\alpha_2)$	$P\bar{6}_3$	$\beta_\varphi (0^\circ < \varphi < 90^\circ)$: $\chi(\beta_1) \oplus \chi(\beta_2)$	$P6_3$

Summary:

- ◆ 2DCOS and PCA are performed on the temperature-dependent Raman spectra of hexagonal $RMnO_3$ ($R=Ho, Er$) thin films and $LuMnO_3$ single crystal.
- ◆ The difference in the magnon scattering of hexagonal $RMnO_3$ ($R=Lu, Ho, Er$) is correlated with the different magnetic symmetries of these materials.

Conclusion

- ◆ Spin excitations near 0.1 eV are found in hexagonal $RMnO_3$ by Raman spectroscopy.
- ◆ They are strongly resonant with 1.8 eV, near the Mn d-d transition.
- ◆ They correlate with the magnetic ordering of Mn^{3+} ions (T_N).



A scenario of spin-flip assisted by Mn d-d transition

**THANK YOU FOR YOUR
ATTENTION**

