



# ORIGIN OF FERROMAGNETISM IN PYROCHLORE AND CHALCOSPINELS

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# MOTIVATION

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Important to understand origin of ferromagnetism in

- ▶ Mn-based Pyrochlores, for e. g.  $\text{Tl}_2\text{Mn}_2\text{O}_7$
- ▶ (Cd,Cu) Chalcospinels

## WHY ?

- ▶ scope of application in magnetic data storage devices
- ▶ rich physics: magnetic and transport properties



**PART I**

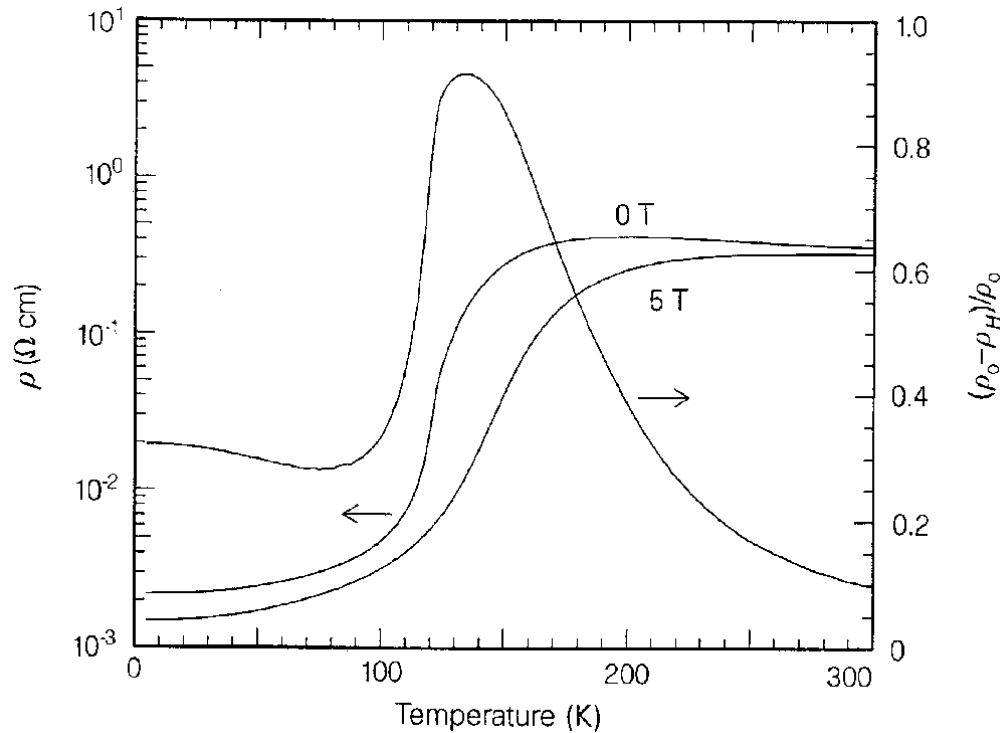
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**Phys. Rev. Lett. 96, 087205 (2006)**

**Phys. Rev. B 75, 014443( 2007)**

# MOTIVATION



Hwang  
Cheong,  
(1997)

and  
Nature

Magnetoresistance ratio at  $T_c = 135-140 \text{ K}$  & 7 T reaches -86 %  
(Shimakawa et. al., Nature (1996))

**Main issue - ORIGIN OF FERROMAGNETISM**



# MANGANITES & $Tl_2Mn_2O_7$ ?

## SIMILARITIES WITH MANGANITES

- ▶ Magnetoresistance ratio at 135 K & 7 T reaches -86 %, comparable with -90 % at 15 T in  $La_{1-x}Sr_xMnO_3$
- ▶ Both are oxides
- ▶ Both have strong local moment magnetism arising from octahedrally coordinated Mn
- ▶ Both exhibit dramatic decrease in resistivity associated with transition from High Temperature (HT) Paramagnetic (PM) to Low Temperature (LT) Ferromagnetic (FM) states

## DISSIMILARITIES WITH MANGANITES

- ▶ Jahn-Teller distortion (JTD) is absent
- ▶  $Mn^{4+}$  - A  $t_{2g}^3$  system, No mixed valency
- ▶ Both HT and LT Phases - METALLIC



## Double exchange and Polaronic mechanism

- ▶ Absence of mixed valency
- ▶ No Jahn-Teller effect
- ▶ No anomalous spin diffusion



## Ferromagnetic Superexchange

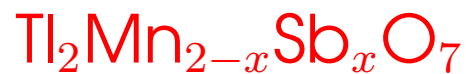
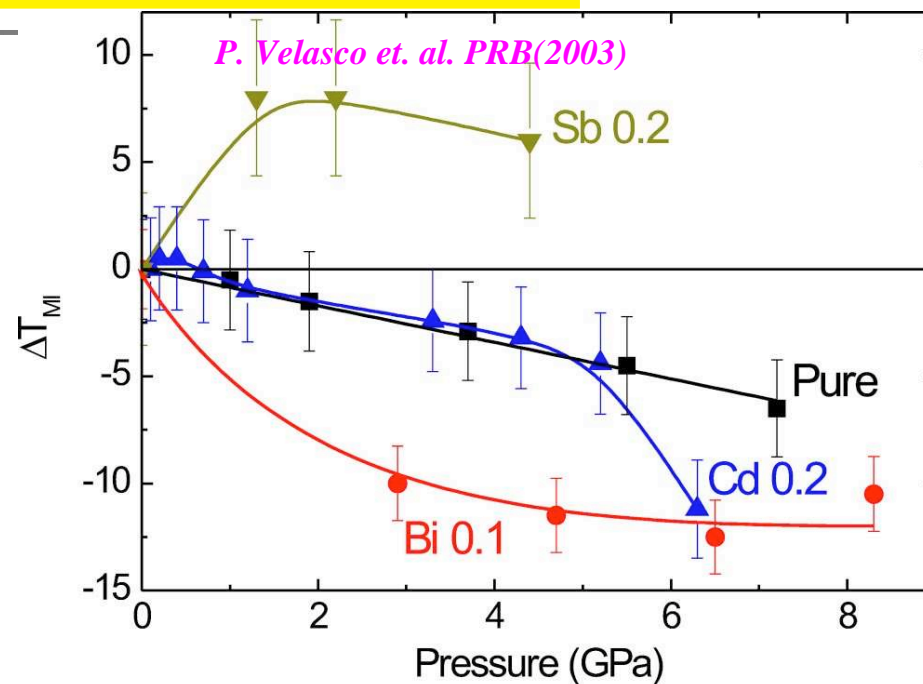
- ▶ Mn-O-Mn angle is about  $134^\circ$
- ▶  $T_c$  decreases with pressure
- ▶ Doping with Sb at the Mn site enhances ferromagnetism



NN AFM superexchange coupling suppressed by longer range FM due to geometric frustration

*Nunez-Regueiro et. al., PRB, Vol. 63, 14417 (2000)*

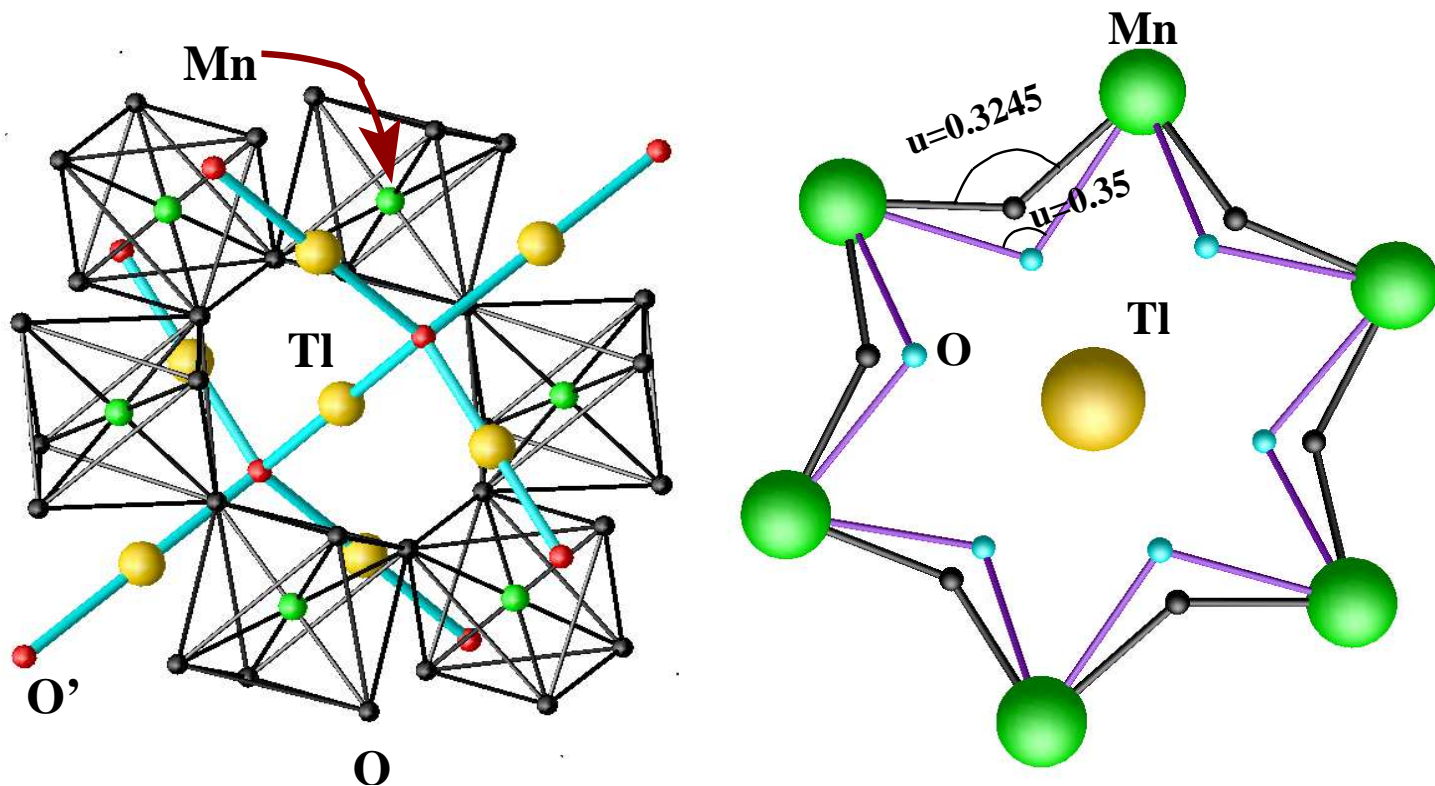
# EXPTS SAY NO TO FM SE



*Alonso et. al, PRB(1999)*

$x$	$T_C$ (K)	$\mu_s(\mu_B)$	$\mu_{eff}(\mu_B)$	$\Theta_{Weiss}$ (K)
0	135	5.30	5.37	147
0.05	190	4.80	4.91	204
0.1	192	4.25	4.84	208
0.2	195	3.18	4.91	205

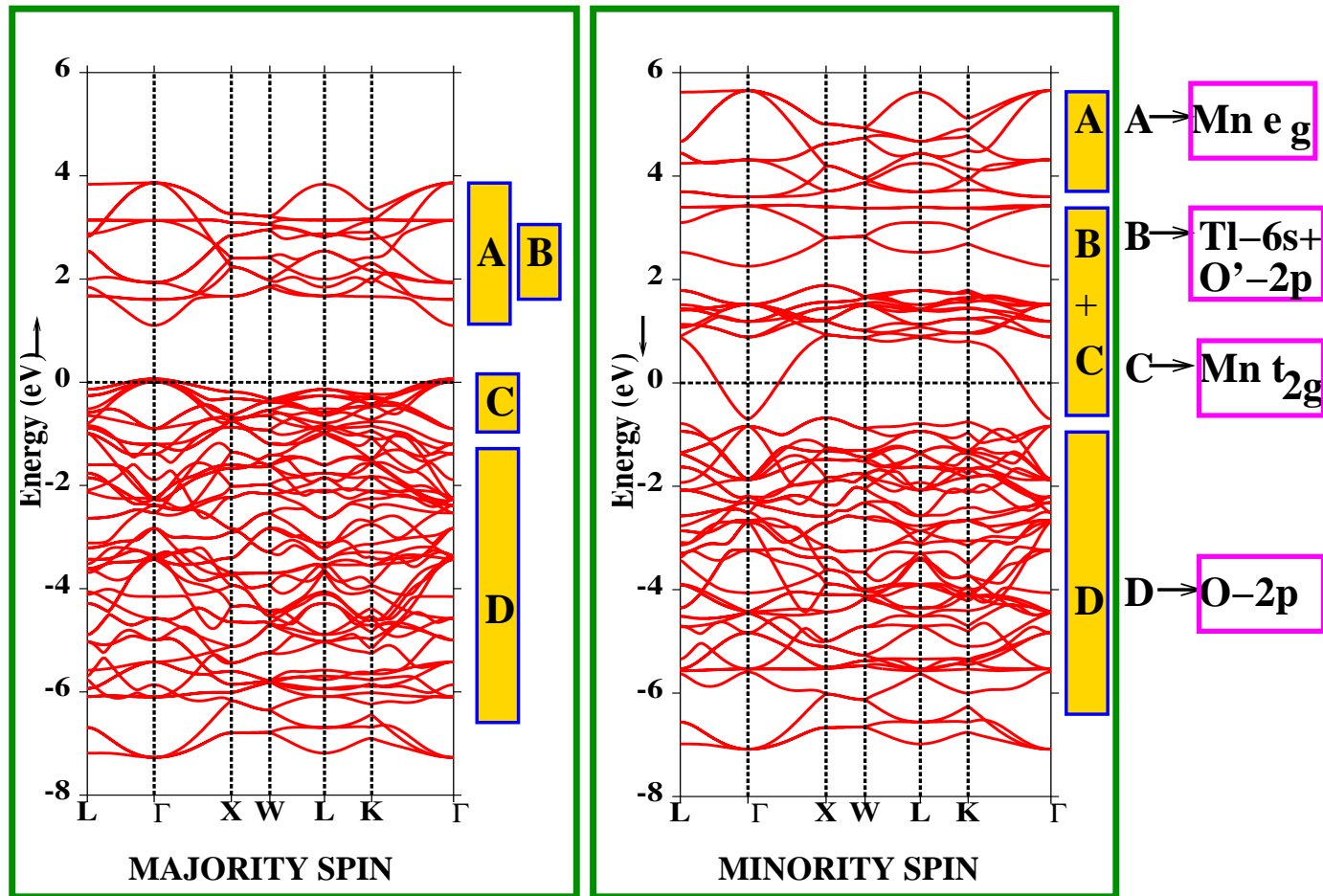
# Structure of $Tl_2Mn_2O_7$



Atom	Tl	Mn	O'	O
Position	.5 .5 .5	0 0 0	.375 .375 .375	$u$ .125 .125

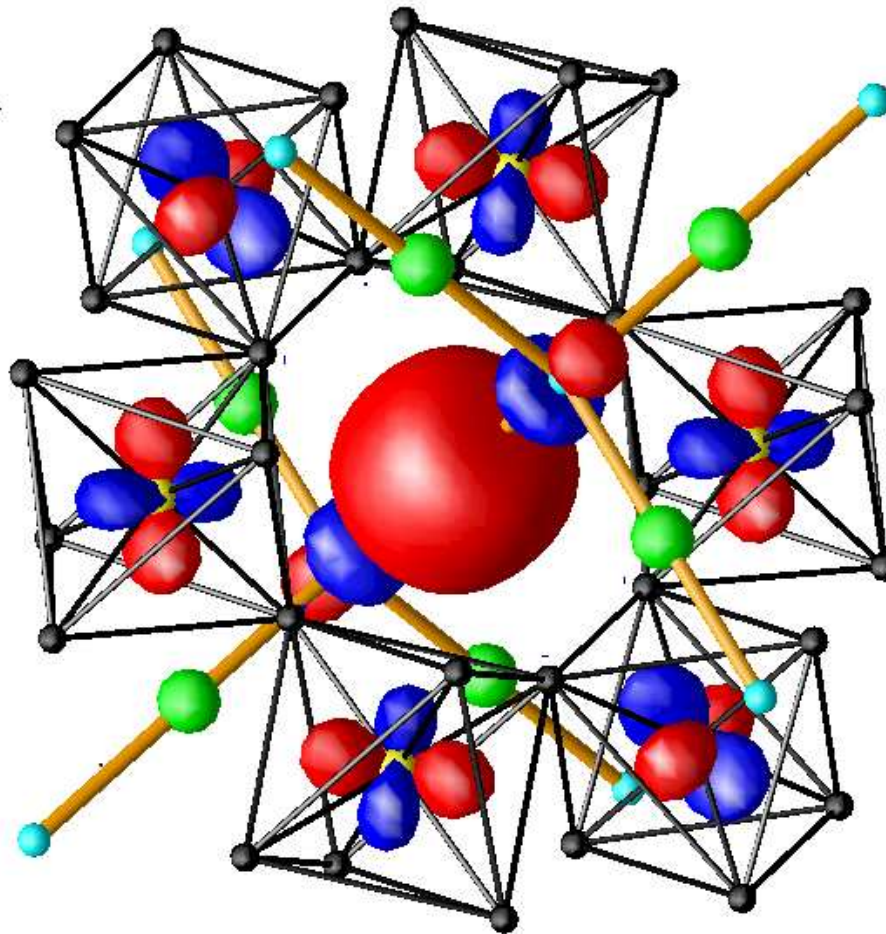
$$u=0.3254$$

# LSDA BAND STRUCTURE UNDER AMBIENT CONDITION



- ▶ **ALMOST HALF-METALLIC**
- ▶ **UNUSUAL HYBRIDISATION BETWEEN THE TI-6s, O'-2p and Mn  $t_{2g}$  STATES IN THE MINORITY SPIN CHANNEL**

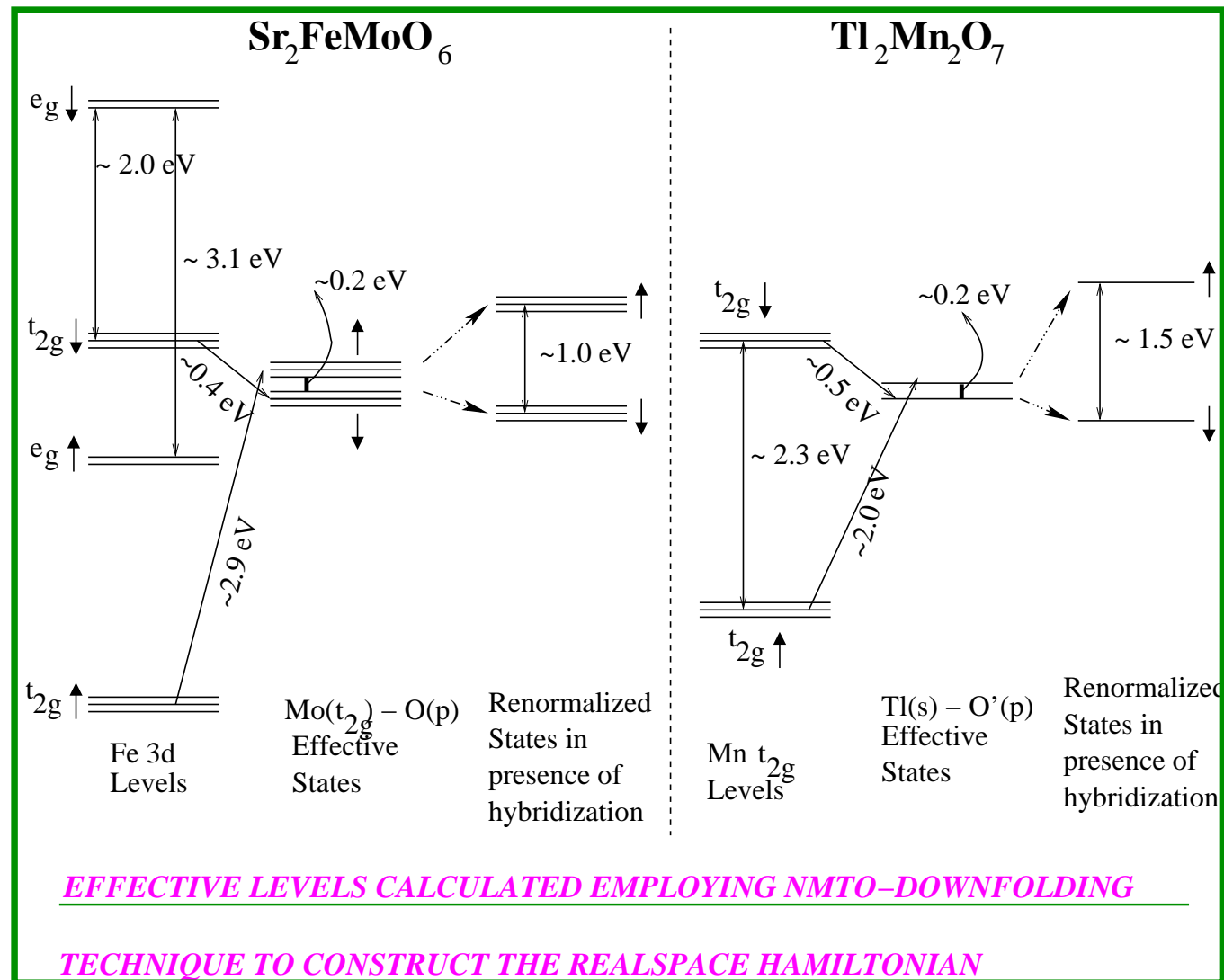
# WANNIER ORBITAL



*NMTO-DOWNFOLDING-O. K. Andersen and T. Saha-Dasgupta,(PRB Rapid  
2000)*

# $Tl_2Mn_2O_7$ and $Sr_2FeMoO_6$

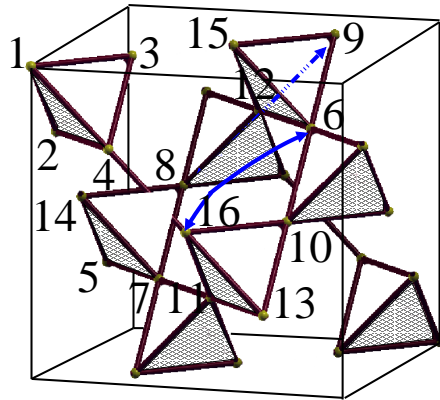
$Sr_2FeMoO_6$  -  
Sarma et. al.  
(PRL 2000)



► Half-filled  $t_{2g}^3$  state of Mn  $\leftrightarrow$  Half-filled  $d^5$  state of Fe

► Delocalised Tl-O'  $\leftrightarrow$  Delocalised Mo-O

# MAGNETIC EXCHANGE INTERACTIONS



*The Mn ions  
in the Eight Formula  
Unit Supercell*

	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	$\Delta E$
FM	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+	0
AFM1	+	-	+	+	+	-	+	+	+	-	+	+	+	-	+	+	9.55
AFM2	-	-	-	+	-	+	+	-	-	-	-	-	+	-	-	-	13.98
AFM3	+	+	-	-	+	+	-	-	+	+	-	-	+	+	-	-	12.24
AFM4	-	-	+	+	-	+	+	+	+	-	-	+	-	-	-	-	17.59
AFM5	-	-	+	+	-	-	+	+	-	-	+	+	-	+	-	-	18.39

*Spin Configurations & total energies  
Considered to calculate J.*

Total Energy differences for five AFM configurations relative to FM mapped onto effective Heisenberg Hamiltonian, considered till 3NN

$$(1) \quad H = J_1 \sum_{nn} S_i \cdot S_j + J_2 \sum_{2nn} S_i \cdot S_j + J_3 \sum_{3nn} S_i \cdot S_j$$

$$T_c = \frac{S(S+1)}{3k_B} J_0; \quad J_0 = 6J_1 + 12J_2 + 12J_3; \quad S_i = \frac{3}{2}$$

↓

$$J_1 = -2.52 \text{ meV}, \quad J_2 = -0.11 \text{ meV} \text{ and } J_3 = +0.33 \text{ meV}$$



**Mean Field  $T_c = 181 \text{ K}$  (exptal. = 142 K)**

**NN INTERACTION IS NOT AFM**



# PRESSURE AND DOPING EFFECTS -POSTER

## PRESSURE

- ▶ Structural issue is resolved - Negligible decrease in  $\angle \text{Mn-O-Mn}$
- ▶ Movement of the energy levels causes changes in hybridisation strengths  $\Rightarrow$  suppression in the kinetically driven magnetic interactions  $\Rightarrow$  reduction of  $T_c$



**Suppression of ferromagnetism**

## DOPING WITH Sb

Following perturbative treatment (Kanamori, 2001),  $J \approx t^4 D_p(E_F) / \Delta^2 \Rightarrow$   
Increase of magnetic interaction on increase of density of states at  $E_F$  !



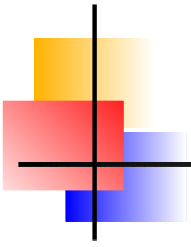
**Signature of enhancement of ferromagnetism due to increase  
in the concentration of the delocalised charge carriers**



## CONCLUSIONS OF PART I

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- ▶ **Kinetic energy driven spin polarisation of the delocalised charge carriers derived from Tl-6s and O'-2p states leads to robust Ferromagnetism**
- ▶ **The anomalous pressure and doping effect can be explained within the same mechanism**



**THANK YOU FOR YOUR KIND ATTENTION**