

Title: Low Spin State Mott Insulators and Emergence of novel Quantum Spin Liquids

Date: Feb 14, 2014 11:00 AM

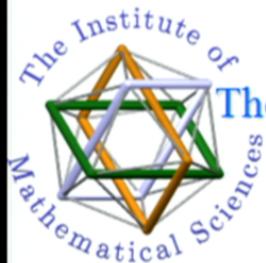
URL: <http://pirsa.org/14020128>

Abstract: <span>In a conventional Mott insulator, magnitude of local spin moments remain fixed. They are 'fixed spin Mott insulators'. We suggest that, in a multi orbital Hubbard model, when local Hund coupling is won over by inter-orbital superexchange couplings between neighboring sites, local spin moment can decrease its value in a cooperative fashion, through a first order phase transition. These are 'Low spin state Mott insulators' (LSSMI). The minimal value of spins that can be reached are zero (half), for even (odd) number of electrons per site. We show that in LSSMI, depending on orbital degeneracy and electron number per site, novel quantum spin liquids can emerge. We discuss systems such as Fe arsenide, Fe selenide family and La<sub>2</sub>CoO<sub>3</sub> in the light of our proposal. Certain long standing puzzles, including absence of any magnetic phase transition in La<sub>2</sub>CoO<sub>3</sub> is explained in terms of a novel quantum spin liquid. Some properties of this spin liquid, a liquid of 'quantum strings' will be discussed and some predictions made.</span>

# **Low Spin State Mott Insulators and Novel Quantum Spin Liquids**

**Emergence in Complex Systems**

February 10-14, 2013



G Baskaran

The Institute of Mathematical Sciences  
Chennai 600 113, India

**PERIMETER  
INSTITUTE**  
for Theoretical Physics

## Down to Earth Materials and Emergence

### Spin State Transition in Complexes ...

Competing Hund coupling, crystal field splitting, spin orbit coupling, covalency ...

### A walk through the Garden of Mott Insulators

Transition Metal Oxides, chalcogenides, pnictides, organics ...

### Soft Spin Mott Insulators

### A old Puzzle in $\text{LaCoO}_3$ and a New Resolution

A novel 3 dimensional quantum spin liquid

### Fe Pnictides, NiGa compound, $\text{La}_2\text{Ni O}_4$ , $\text{Li}_2\text{Ru O}_3$ ...

An unexpectedly Rich Quantum World created by  
down to earth Materials

Emergence of New Notions & Entities

More is much more different in Quantum World

Niels Bohr's remark to Ernst Myer, an evolutionary Biologist  
All of Physics is Emergence



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Metals .. Cu, Al ...

Fermi Sea, Pauli Pressure Chandrasekhar Mass limit of stars

Specific Heat of Solids ...

Phonon, magnon, free Bose gas  
First Quantum Field Theory

Superconductivity in Hg ...

Macroscopic Wave Function  
Flux Quantization, Josephson Effect  
Nambu, Anderson-Higgs Phenomenon

Insulators, Semiconductors ...

Bloch States, Band Gaps, holes,  
Impurity States, Anderson localization

2D Electrons, Graphene  
in magnetic Fields

... Integer Quantum Hall Effect

Topological Band Insulator ... Topologically protected Quantum States



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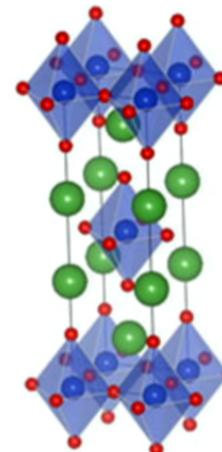
## THE QUANTUM FIELD THEORY

- Superconductivity in Hg ... Macroscopic Wave Function
- Flux Quantization, Josephson Effect
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# Mott Insulators

## Quantum Magnetism, Multiferroics



**Quantum Spin Liquids** Anderson  
Seat of High Tc Superconductivity, Non-Fermi Liquids ...

Emergent Fermions with Fermi Surface Anderson-GB

Emergent Gauge Fields U(1) Anderson-GB , SU(2) Affleck,Zou,Hsu, Anderson

Spin-Charge Decoupling, Spinon, Holon Kivelson, Rokhsar, Sethna

Quantum Order, Projective Symmetry Group ... Wen

Topological Order Haldane-Rezayi, Rokhsar-Kivelson Vison Read, Senthil, Fisher

Haldane Gap, AKLT Chain, Haldane-Shastry Model

Kitaev Toric Code, Levin-Wen Models, Dimer model Rokhsar-Kivelson

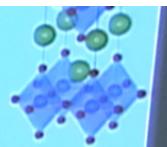
Spin liquid & Majorana Fermion in Kitaev Honey Comb lattice Model

Majorana Liquid Biswas, Fu, Laumann, Sachdev

Quantum String Liquid in Pnictides GB



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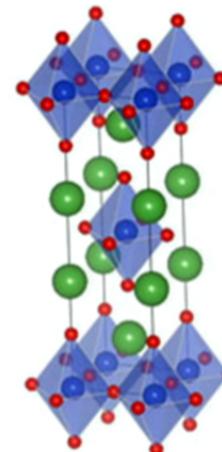


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## Hund Coupling Hubbard U

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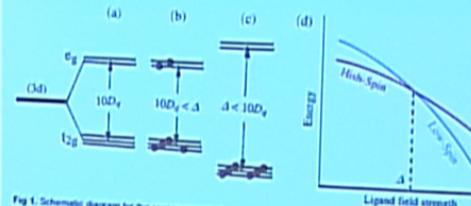
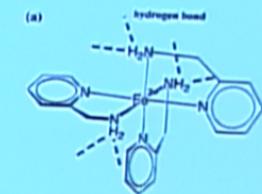


Fig 1. Schematic diagram for the spin crossover phenomena: (a) the energy scheme for one d-electron in octahedral ligand field symmetry; (b) six d-electrons in a weak ligand field; (c) six d-electrons in a strong ligand field; and (d) relationship between the spin state and the ligand field strength.



Pirsa: 14020128

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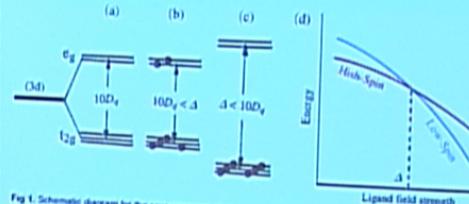
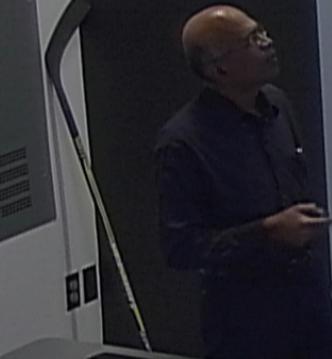
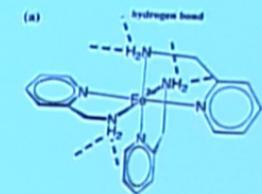


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# Spin State Transition in Transition Metal Complexes

Octahedrally coordinated transition metals  $\text{Fe}^{2+}$   $\text{Fe}^{3+}$   $\text{Co}^{2+}$   $\text{Co}^{3+}$  ....

Crystal Field Splitting (charge and covalency)

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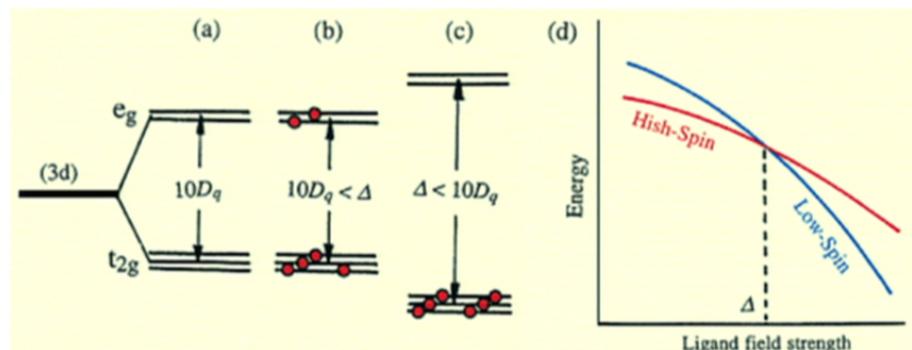
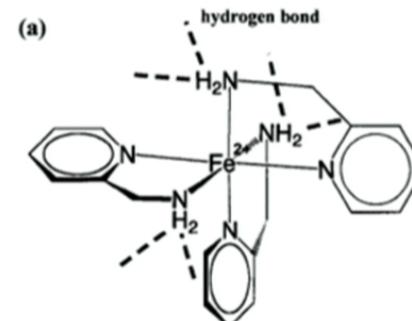


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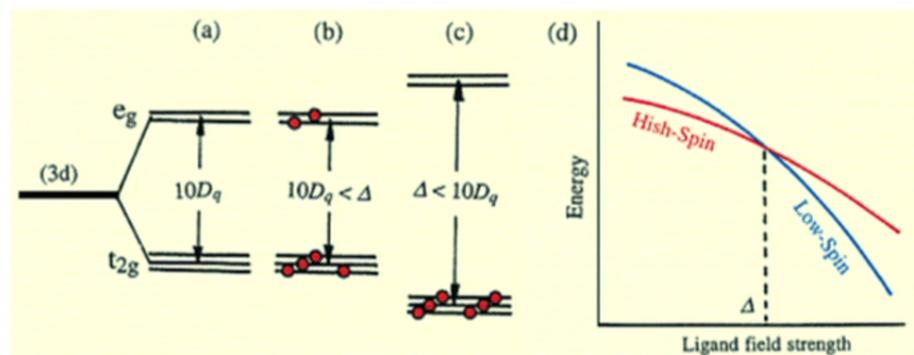
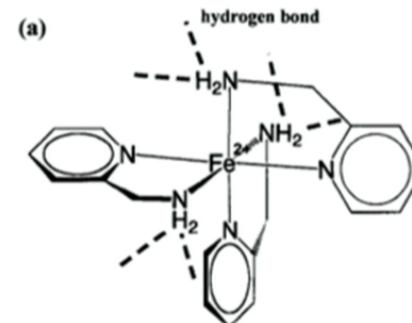


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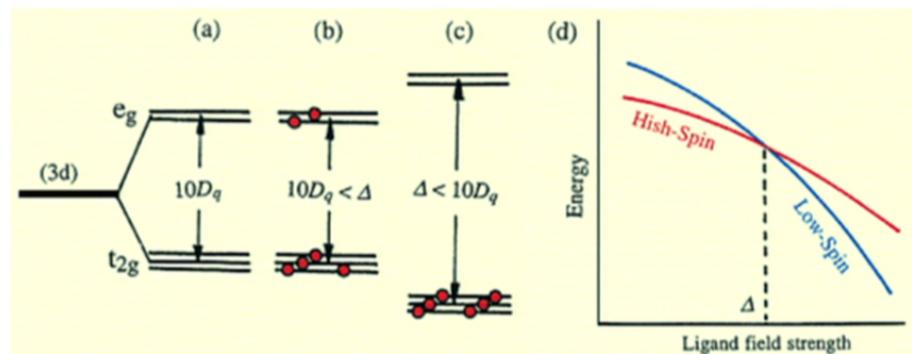
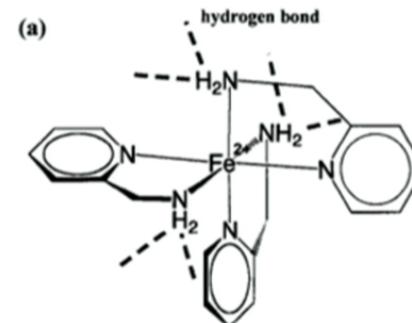


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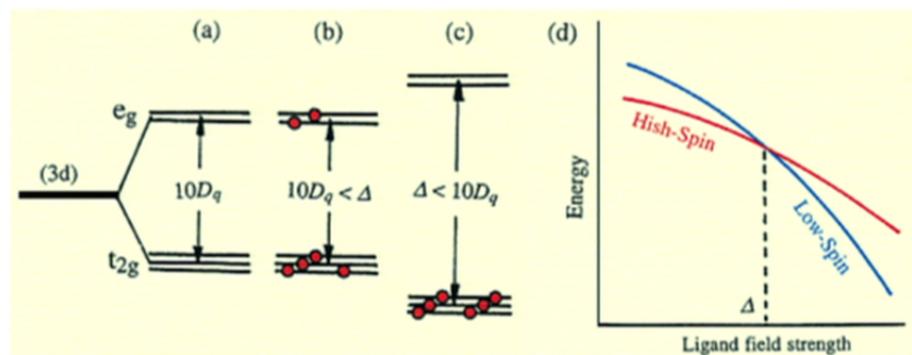
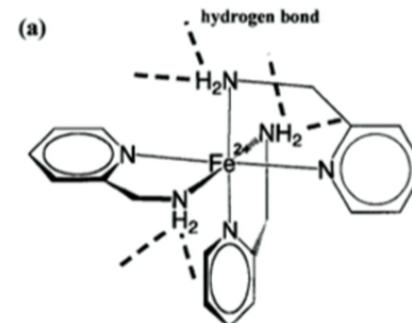


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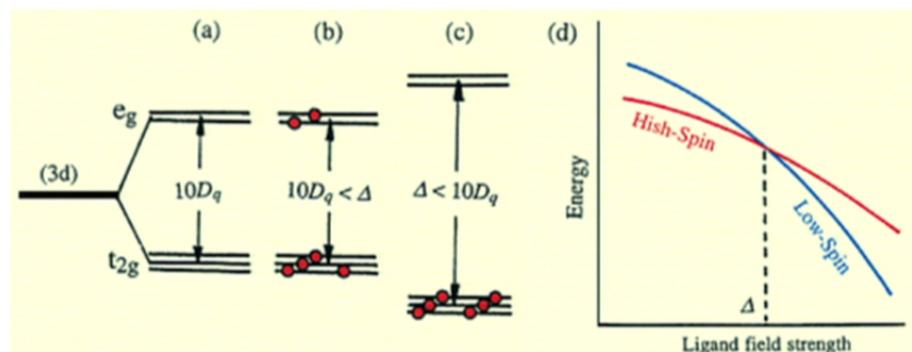
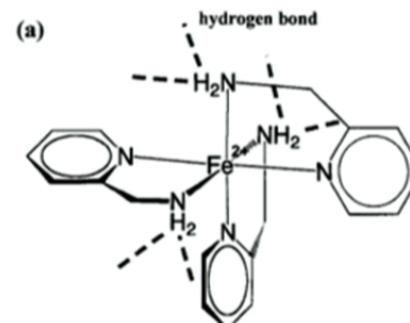


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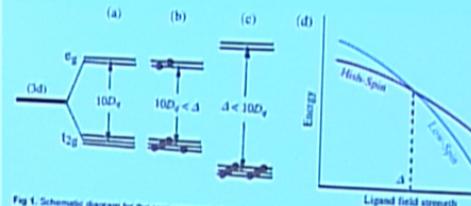
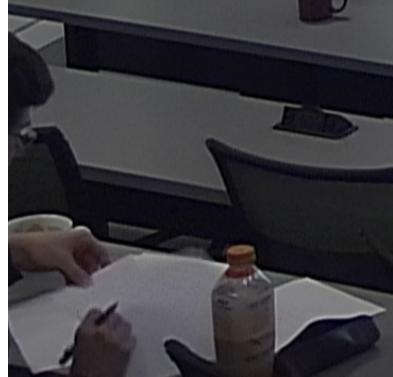
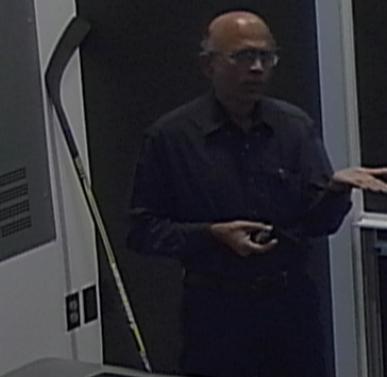
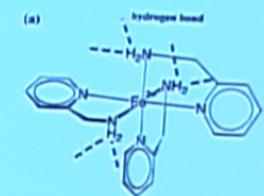


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Friednau Coupling  
Entropy favors high spin state at high T

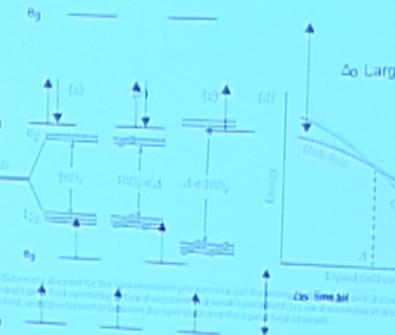
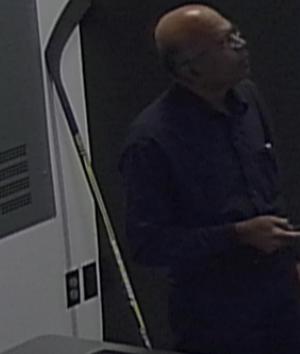
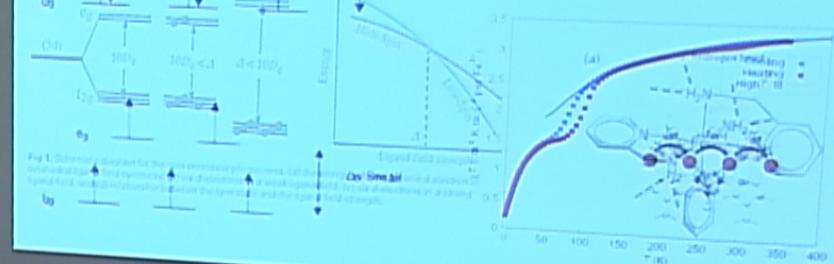


Fig. 1. Schematic diagram for the octahedral complexes of transition metals. (a) the strong ligand field case of a large ligand field splitting  $\Delta_0$  and a low temperature favours the low spin state; (b) intermediate ligand field case where  $\Delta_0$  is intermediate and the two states are degenerate; (c) weak ligand field case where  $\Delta_0$  is small and the high spin state is favoured; (d) very weak ligand field case where  $\Delta_0$  is negligible and the high spin state is favoured.

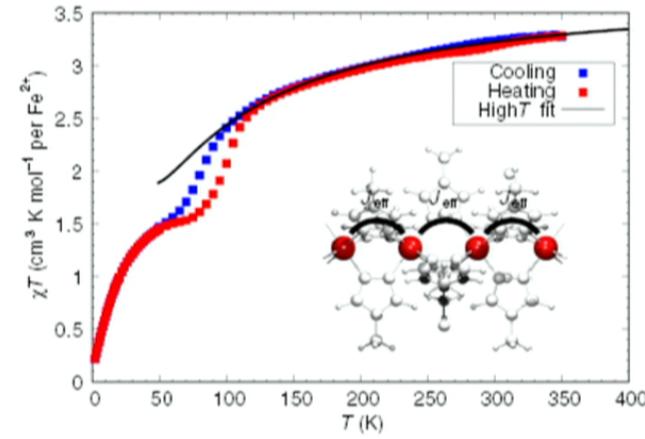
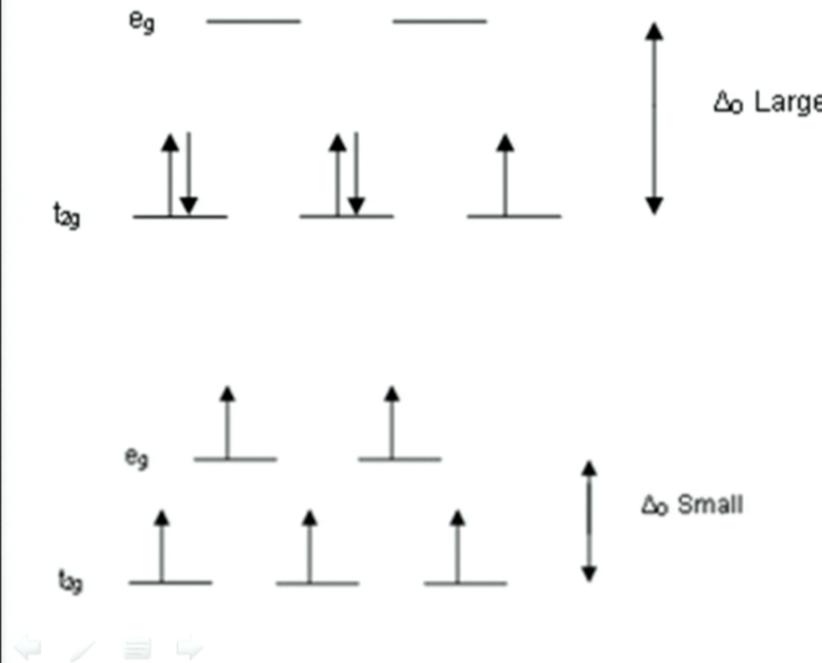
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The effective moment varies from a typical d<sup>5</sup> low-spin value of 2.25  $\mu_B$  at 80 K to more than 4  $\mu_B$  above 300 K

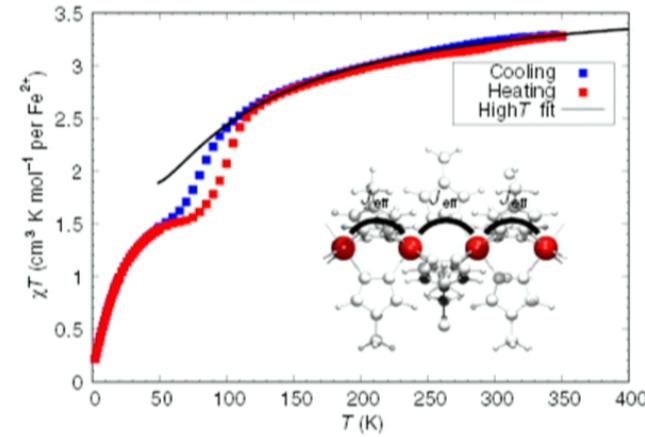
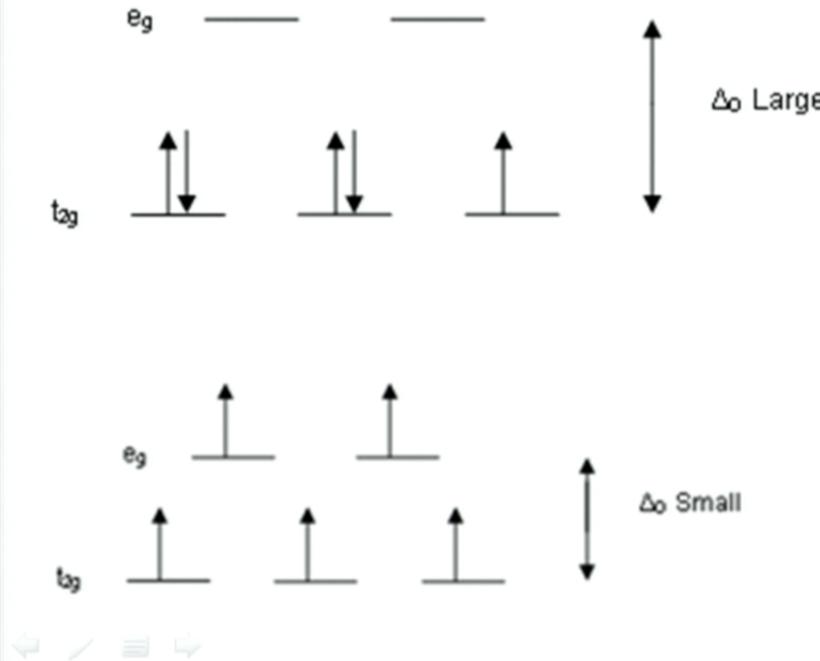
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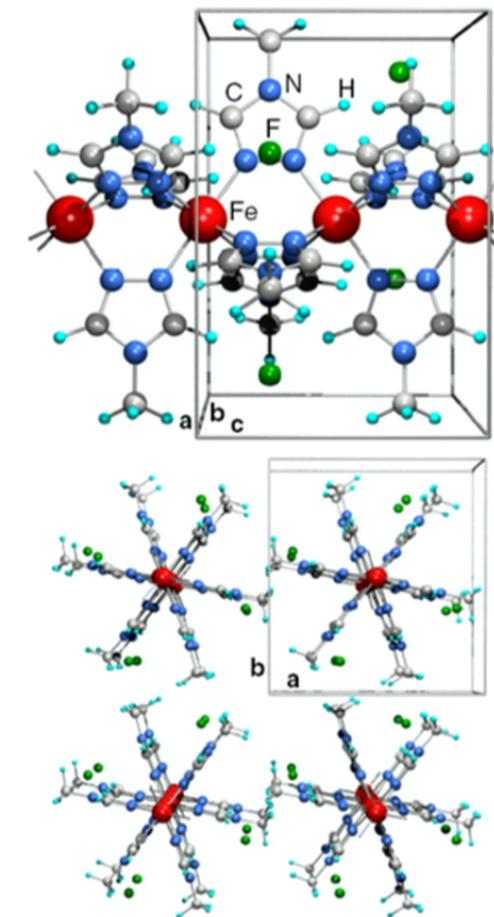
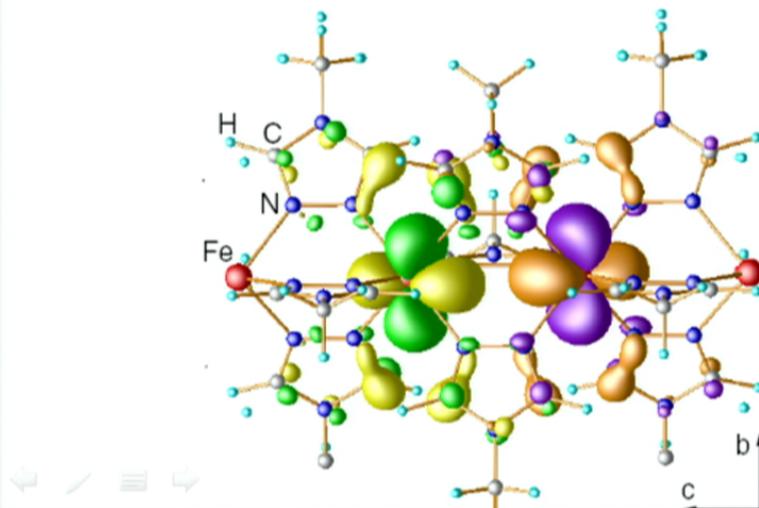


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**Superexchange interaction between Fe-Fe  
starts influencing spin state transitions**

**4-R-1,2,4-triazole ligands**

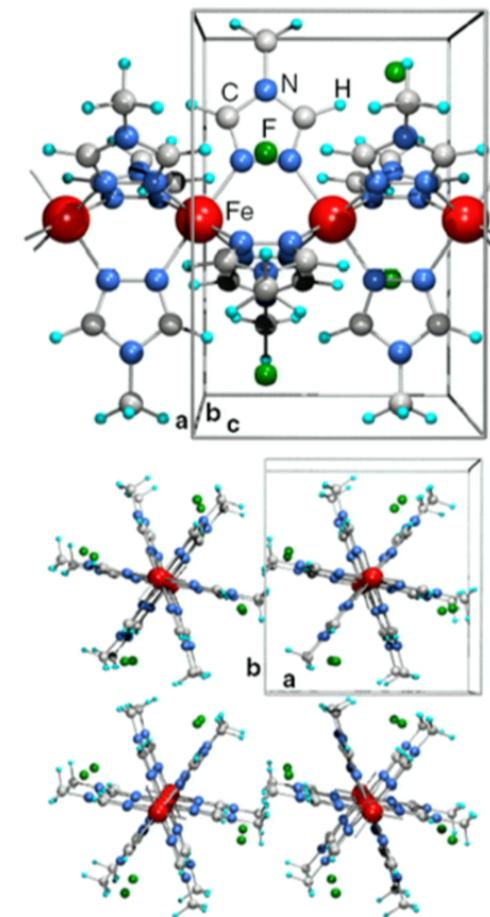
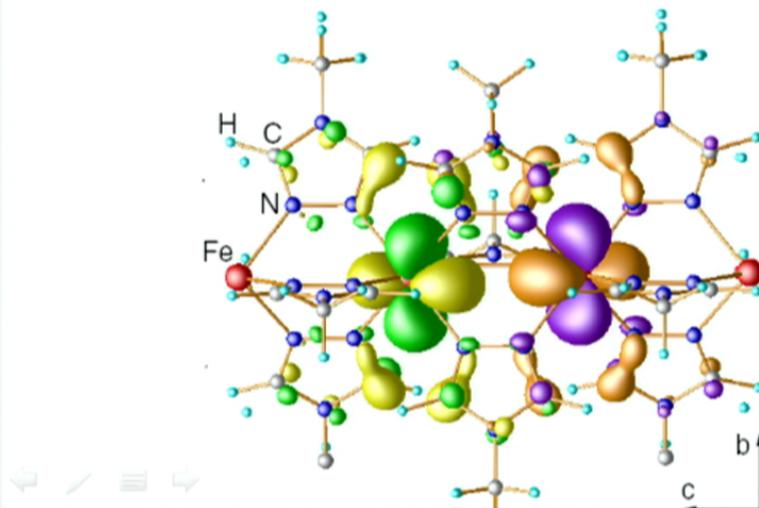


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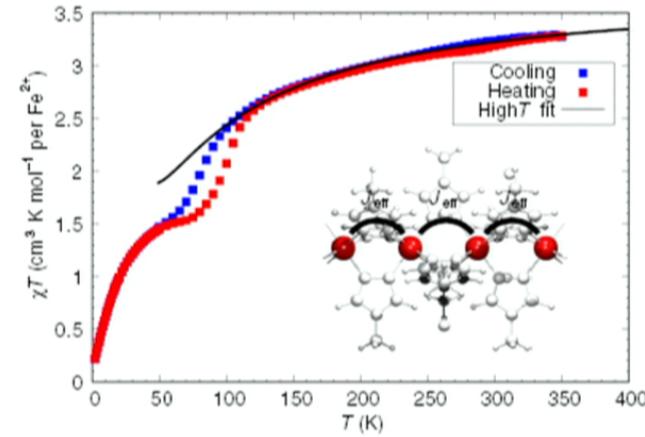
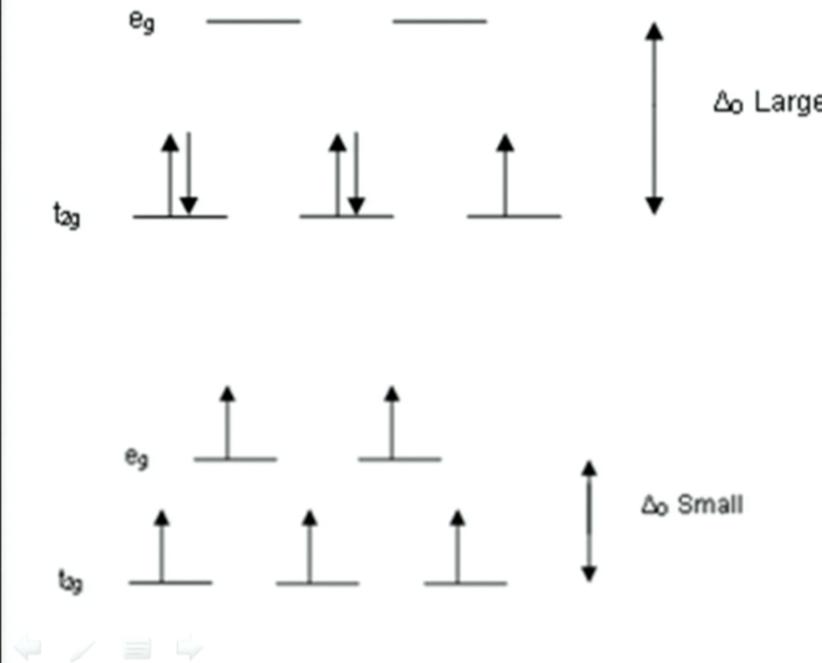
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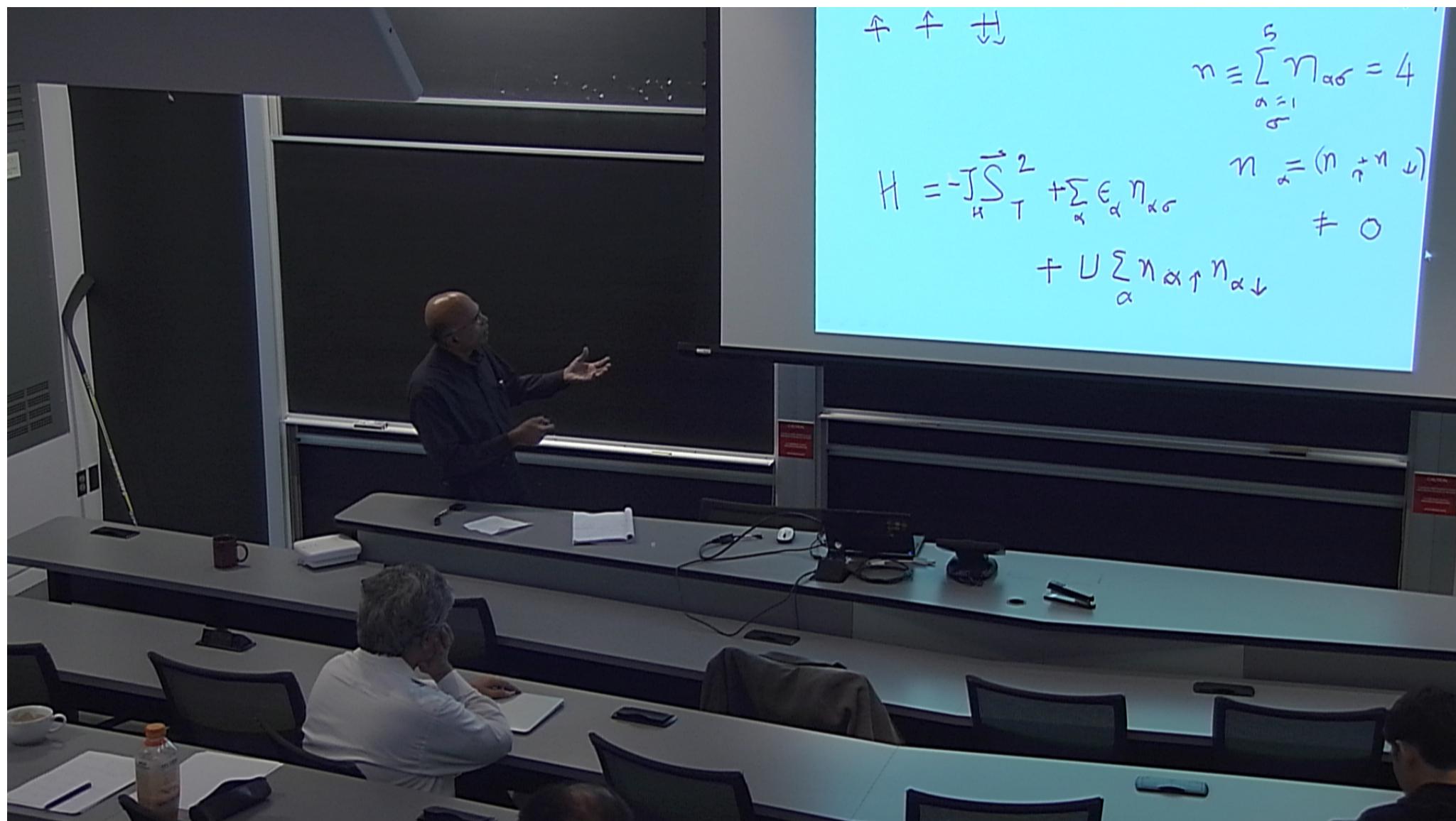
$\uparrow \uparrow \downarrow \downarrow$

$$n = \sum_{\alpha=1}^5 n_{\alpha\sigma} = 4$$

$$H = -J \vec{S}_T^2 + \sum_{\alpha} \epsilon_{\alpha} n_{\alpha\sigma}$$

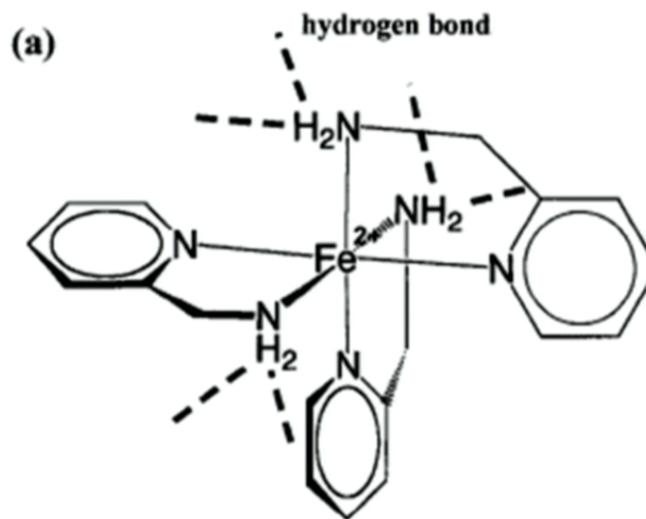
$$n_{\alpha} = (n_{\alpha\uparrow} + n_{\alpha\downarrow}) \neq 0$$

$$+ U \sum_{\alpha} n_{\alpha\uparrow} n_{\alpha\downarrow}$$



In a molecular solid of Fe complexes a first order structural phase transition occurs accompanied by jump in Spin value at a  $T_c \sim 110\text{ K}$

A spin state change causes local charge redistribution and/or Jahn Teller Distortions. The cooperative structural change is a response of the molecular lattice to these changes.



P. Gütlich, A. Hauser, and H. Spiering, Angew. Chem. 33, 2024 (1994).

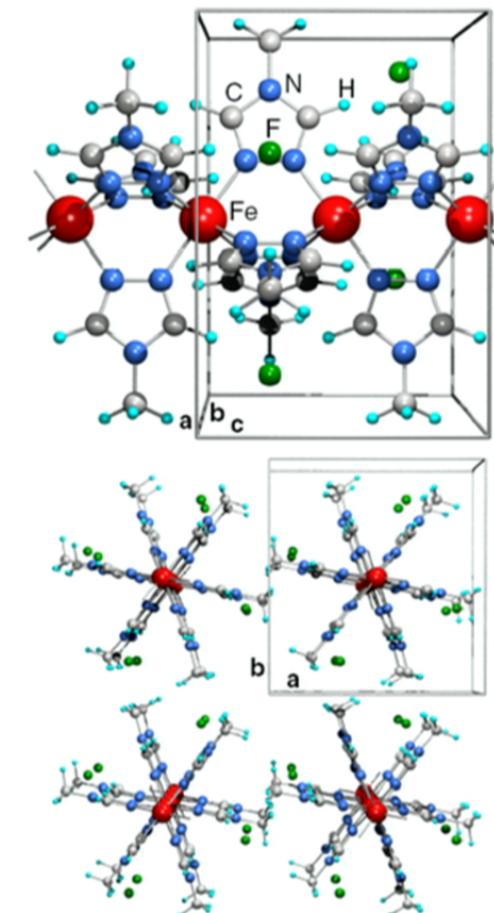
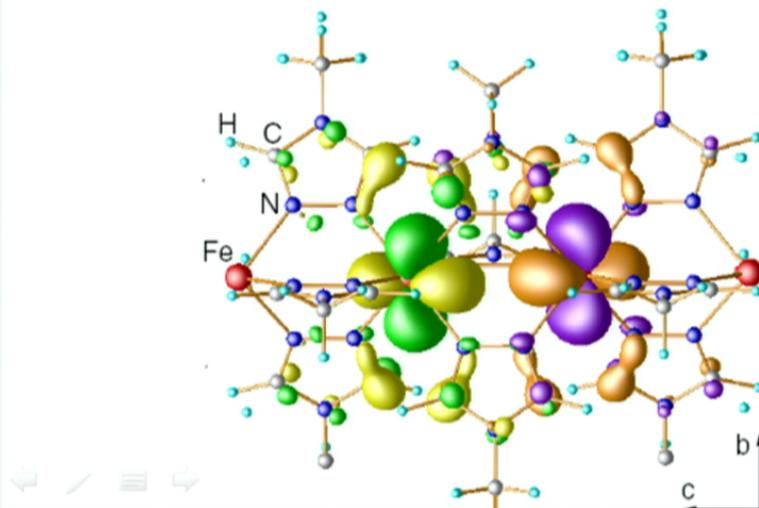


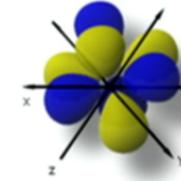
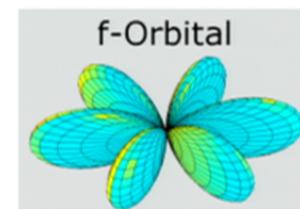
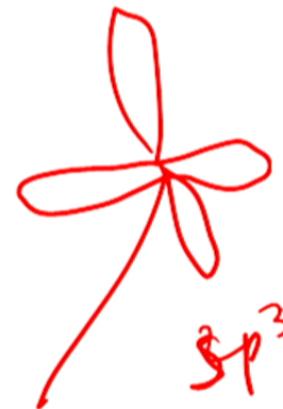
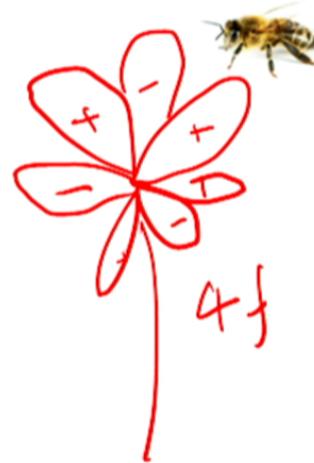
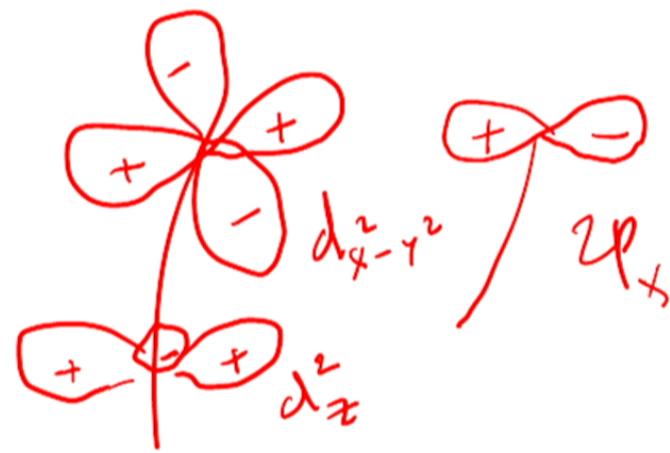
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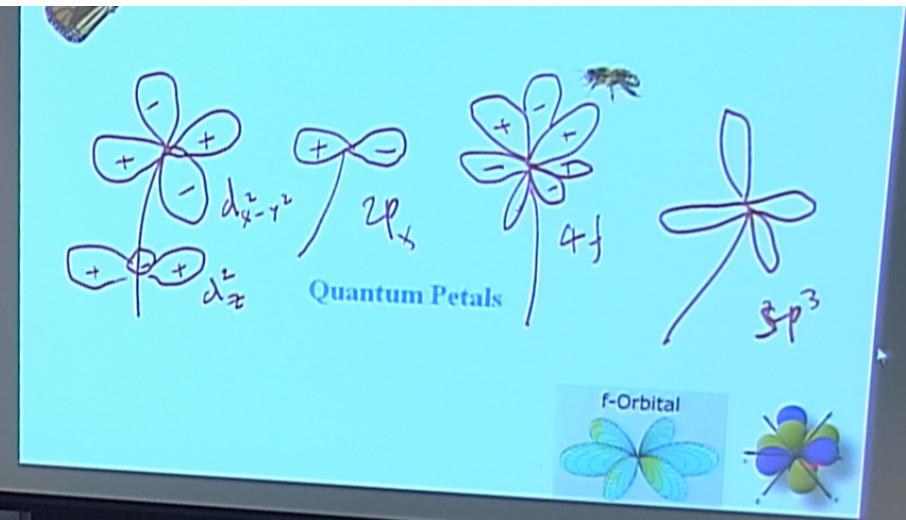
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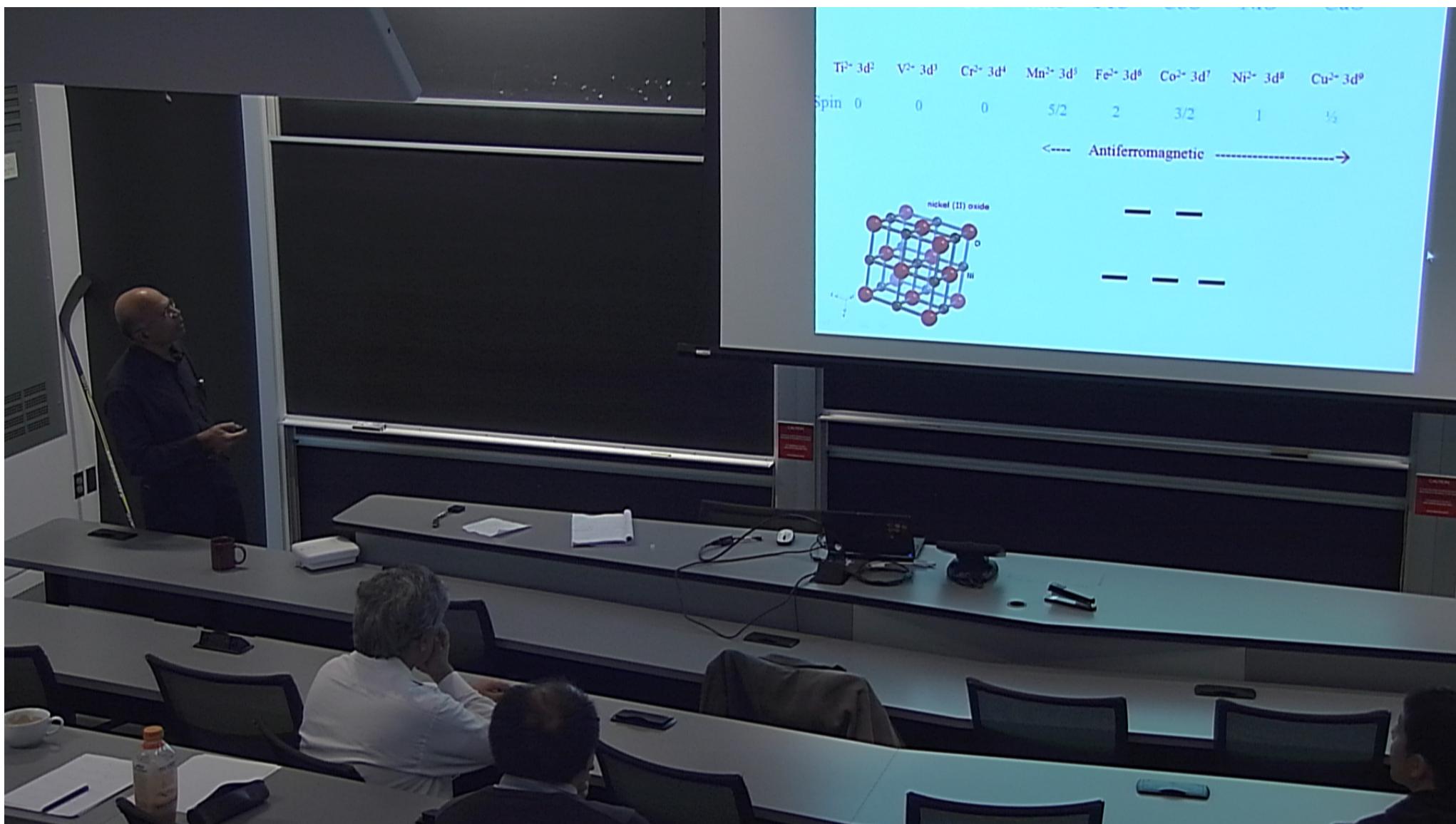
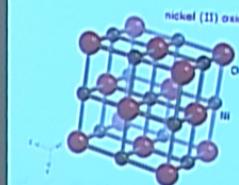






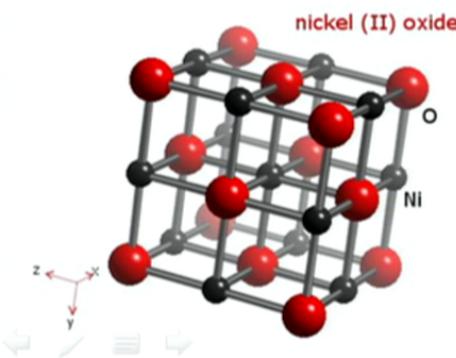
Ti <sup>2+</sup> 3d <sup>2</sup>	V <sup>2+</sup> 3d <sup>1</sup>	Cr <sup>2+</sup> 3d <sup>4</sup>	Mn <sup>2+</sup> 3d <sup>5</sup>	Fe <sup>2+</sup> 3d <sup>6</sup>	Co <sup>2+</sup> 3d <sup>7</sup>	Ni <sup>2+</sup> 3d <sup>8</sup>	Cu <sup>2+</sup> 3d <sup>9</sup>
Spin 0	0	0	5/2	2	3/2	1	1/2

<---- Antiferromagnetic ----->



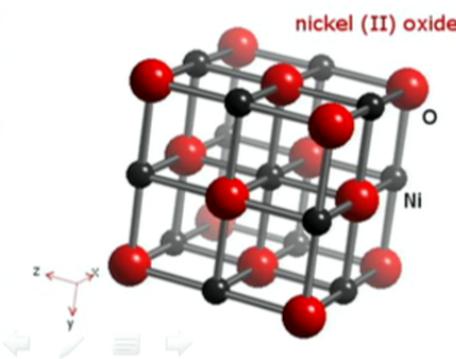
## Simplest Transition Metal Oxide Mott Insulators (NaCl structure)

TiO	VO	CrO	MnO	FeO	CoO	NiO	CuO
$\text{Ti}^{2+} \ 3d^2$	$\text{V}^{2+} \ 3d^3$	$\text{Cr}^{2+} \ 3d^4$	$\text{Mn}^{2+} \ 3d^5$	$\text{Fe}^{2+} \ 3d^6$	$\text{Co}^{2+} \ 3d^7$	$\text{Ni}^{2+} \ 3d^8$	$\text{Cu}^{2+} \ 3d^9$
Spin 0	0	0	5/2	2	3/2	1	1/2
←---- Antiferromagnetic -----→							



## Simplest Transition Metal Oxide Mott Insulators (NaCl structure)

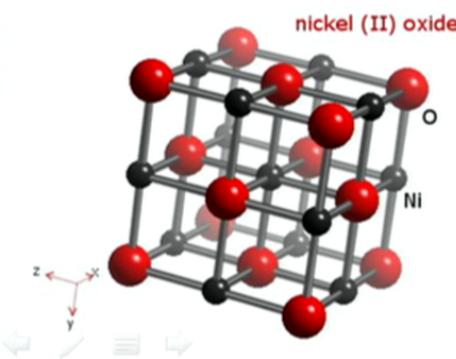
TiO	VO	CrO	MnO	FeO	CoO	NiO	CuO
$\text{Ti}^{2+} \ 3d^2$	$\text{V}^{2+} \ 3d^3$	$\text{Cr}^{2+} \ 3d^4$	$\text{Mn}^{2+} \ 3d^5$	$\text{Fe}^{2+} \ 3d^6$	$\text{Co}^{2+} \ 3d^7$	$\text{Ni}^{2+} \ 3d^8$	$\text{Cu}^{2+} \ 3d^9$
Spin 0	0	0	5/2	2	3/2	1	1/2
<---- Antiferromagnetic ----->							



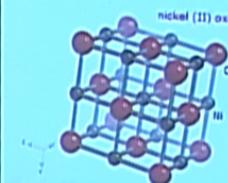
## Simplest Transition Metal Oxide Mott Insulators (NaCl structure)

TiO	VO	CrO	MnO	FeO	CoO	NiO	CuO
$\text{Ti}^{2+} \ 3d^2$	$\text{V}^{2+} \ 3d^3$	$\text{Cr}^{2+} \ 3d^4$	$\text{Mn}^{2+} \ 3d^5$	$\text{Fe}^{2+} \ 3d^6$	$\text{Co}^{2+} \ 3d^7$	$\text{Ni}^{2+} \ 3d^8$	$\text{Cu}^{2+} \ 3d^9$
Spin 0	0	0	5/2	2	3/2	1	1/2

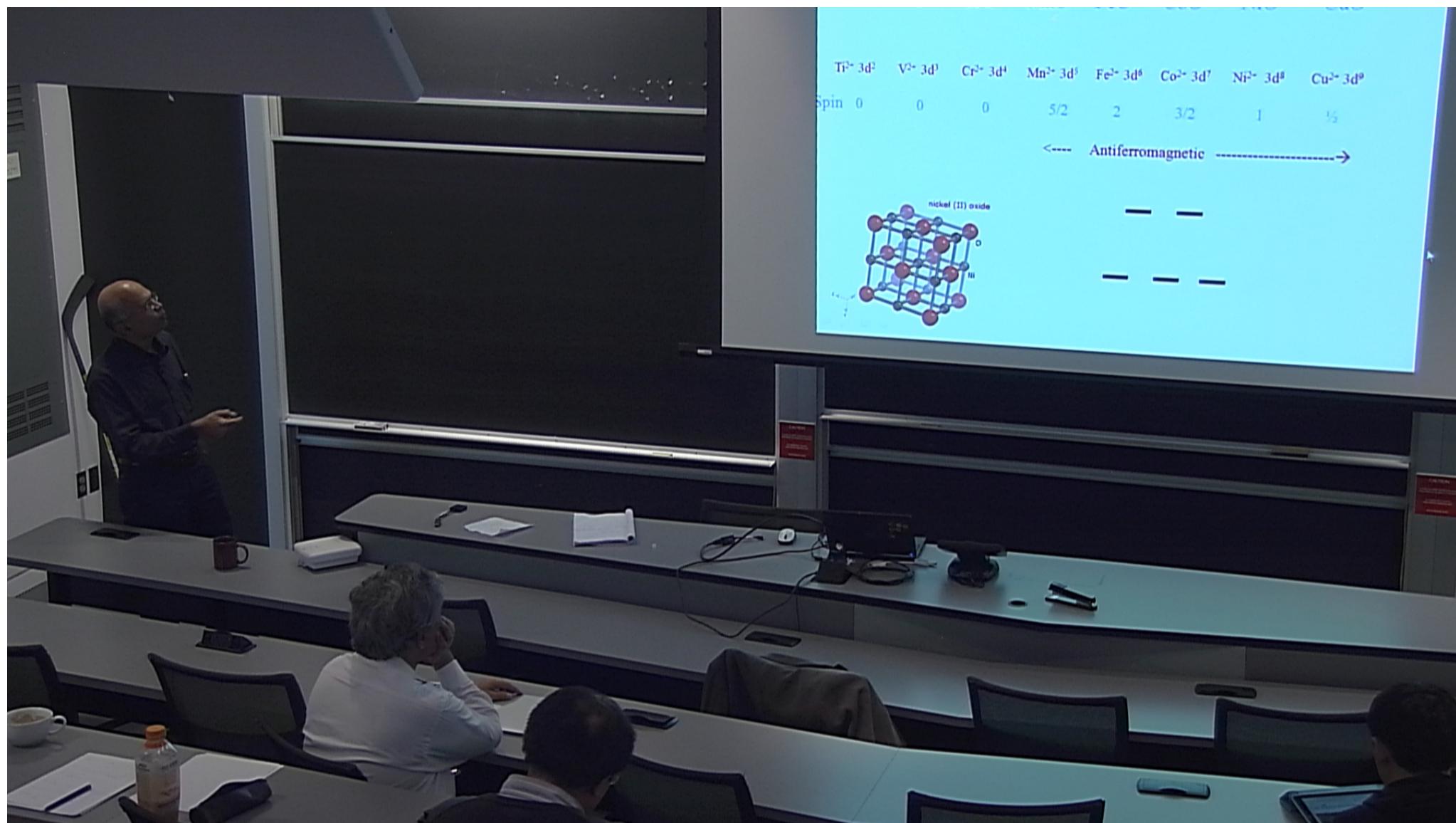
<---- Antiferromagnetic ----->

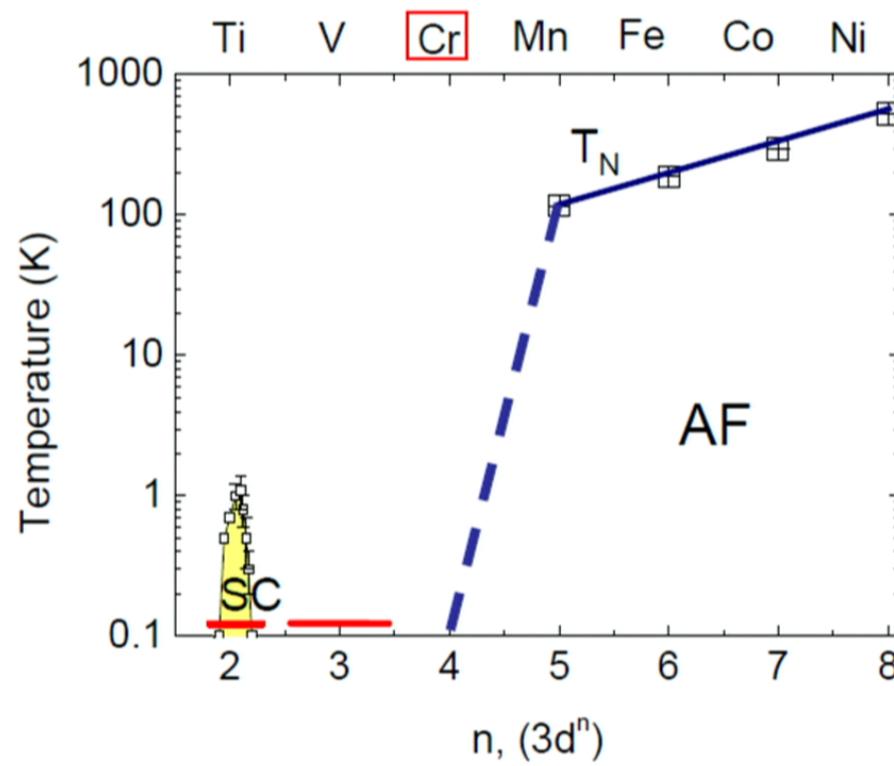


Ti <sup>2+</sup> 3d <sup>2</sup>	V <sup>2+</sup> 3d <sup>1</sup>	Cr <sup>2+</sup> 3d <sup>4</sup>	Mn <sup>2+</sup> 3d <sup>5</sup>	Fe <sup>2+</sup> 3d <sup>6</sup>	Co <sup>2+</sup> 3d <sup>7</sup>	Ni <sup>2+</sup> 3d <sup>8</sup>	Cu <sup>2+</sup> 3d <sup>9</sup>
Spin 0	0	0	5/2	2	3/2	1	1/2

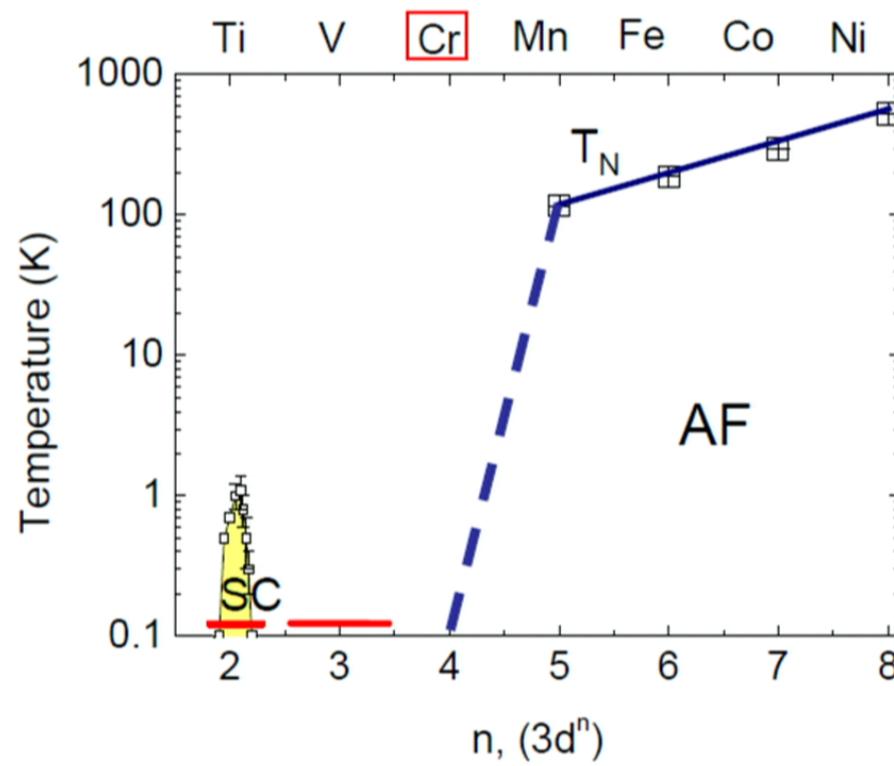


<---- Antiferromagnetic ----->

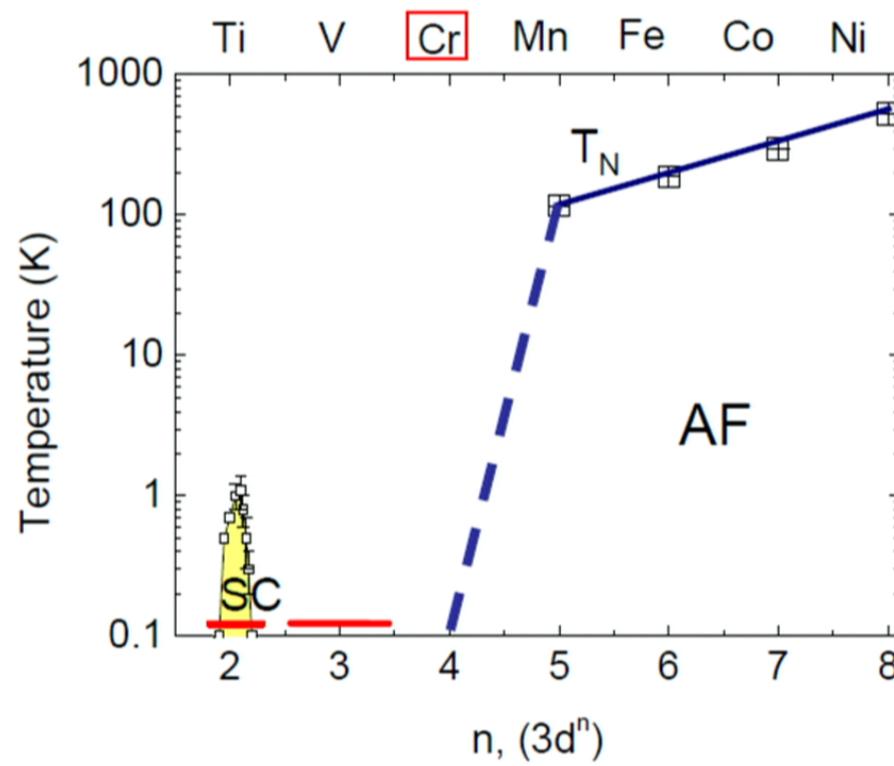




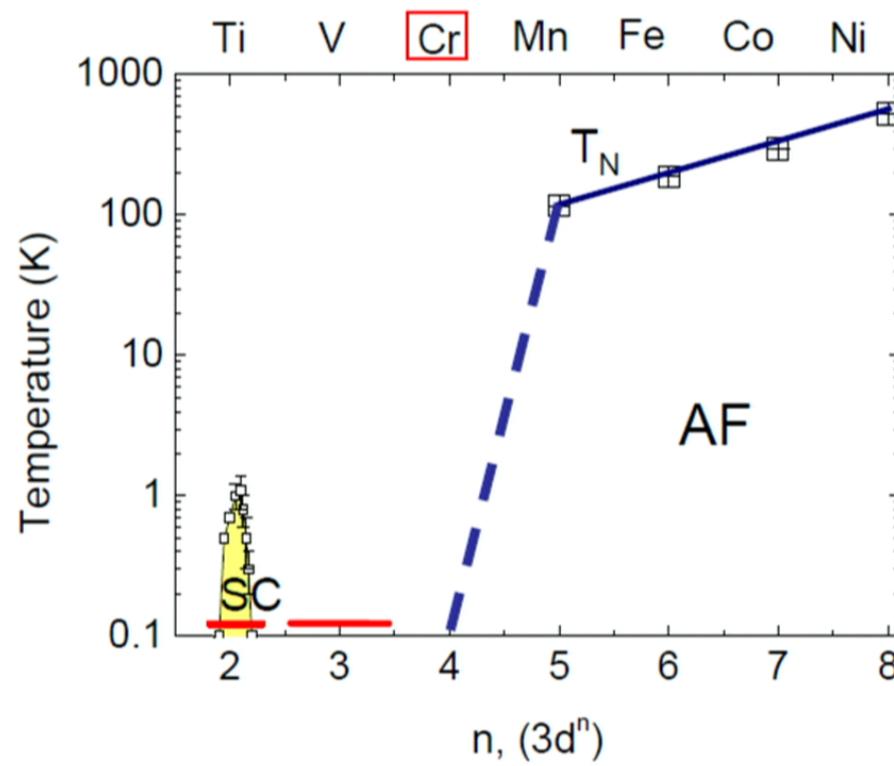
F. Rivadulla et al. Phys. Rev. B 76, 205110 (2007)



F. Rivadulla et al. Phys. Rev. B 76, 205110 (2007)



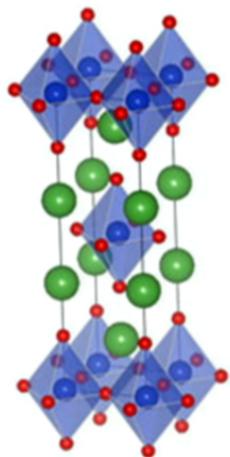
F. Rivadulla et al. Phys. Rev. B 76, 205110 (2007)



F. Rivadulla et al. Phys. Rev. B 76, 205110 (2007)



$\text{La}_2 \text{Ni O}_4$  A layered perovskite similar to cuprate

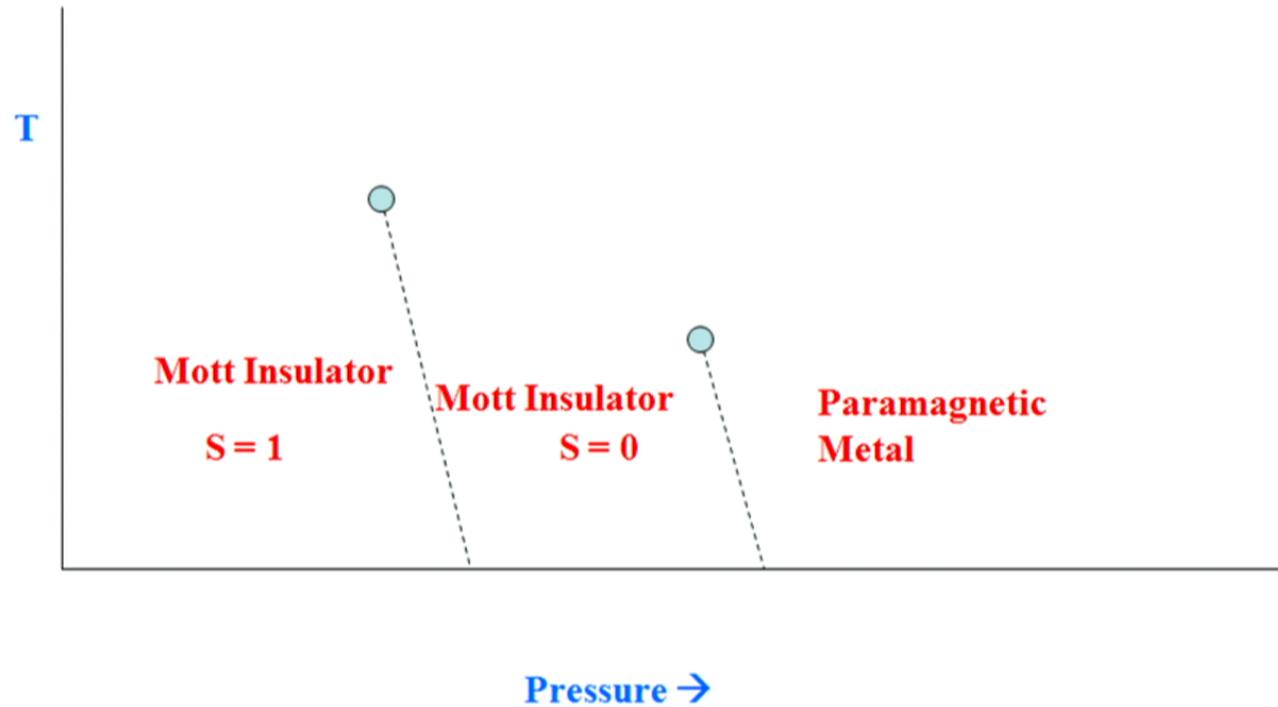


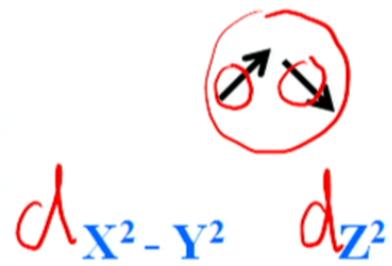
$\text{Ni}^{2+} : 3d^8$   
 $t_{2g}^4 t_{2g}^3$   
 $t_{2g}^3 t_{2g}^2$   
 $t_{2g}^2 t_{2g}^1$

$$H = -J_H \sum (\vec{S}_{i1} + \vec{S}_{i2})^2 + J_c \sum_{\langle ij \rangle} \vec{S}_{i1} \cdot \vec{S}_{j1} + J_Z \sum_{\langle ij \rangle} \vec{S}_{i2} \cdot \vec{S}_{j2}$$



## Mott Transition for $\text{La}_2 \text{Ni O}_4$ Schematic





**Spin-1**    Escape from Antiferromagnetic order  
and Hund coupling through spin singlet formation



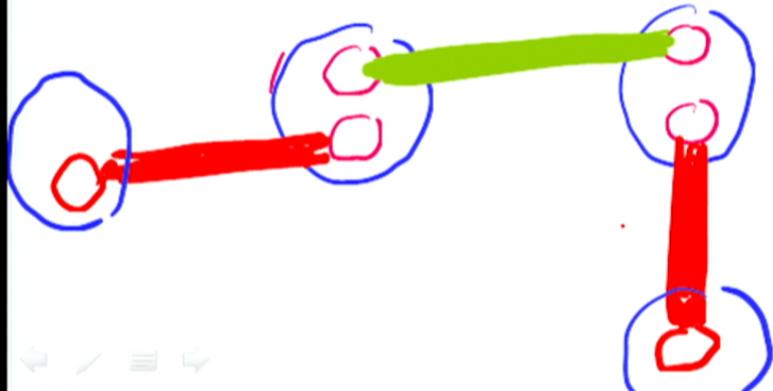
◀ Haldane mechanism    AKLT Chain

# Bond Charge Repulsion

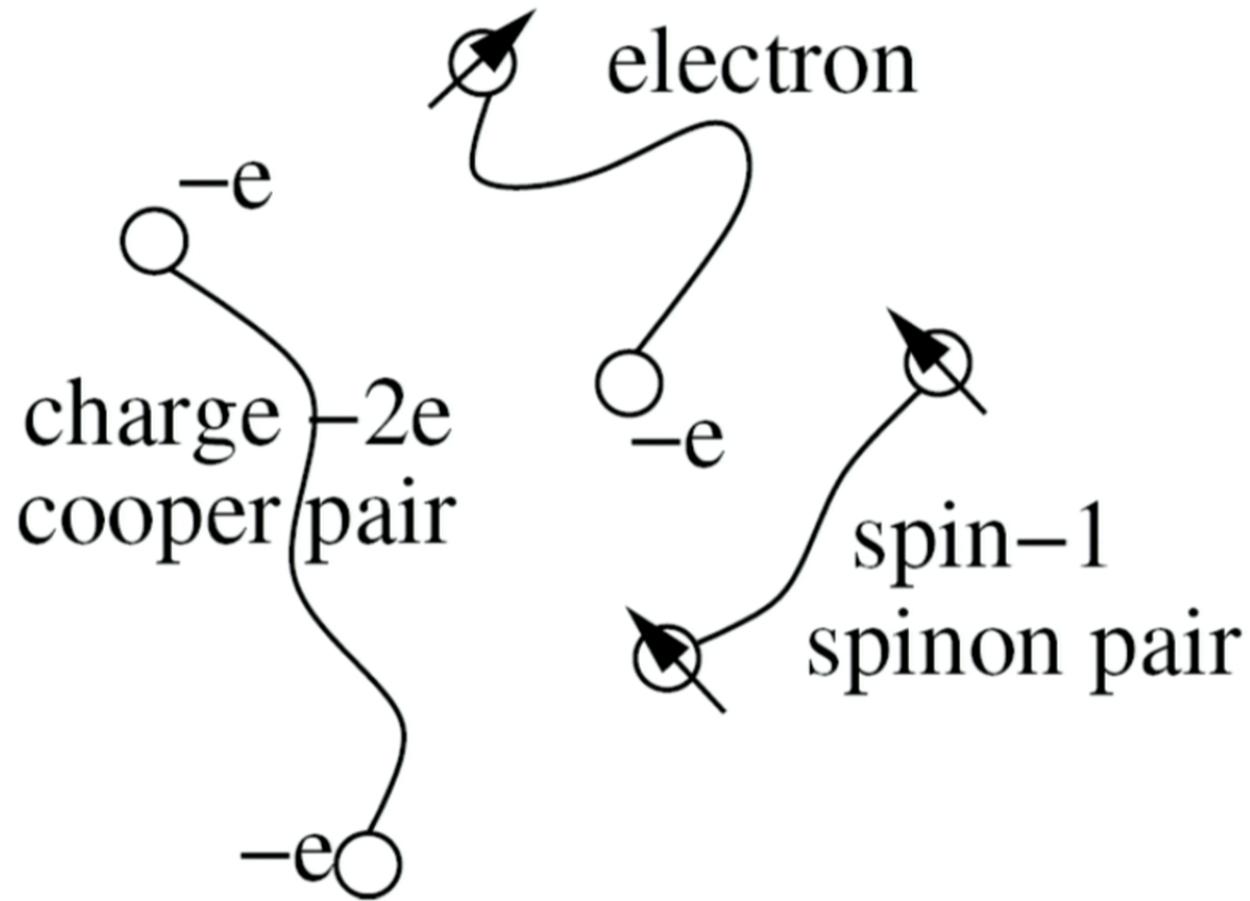
GB  
2008



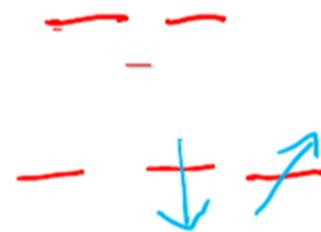
Not allowed  
by electrostatic  
repulsion



Allowed

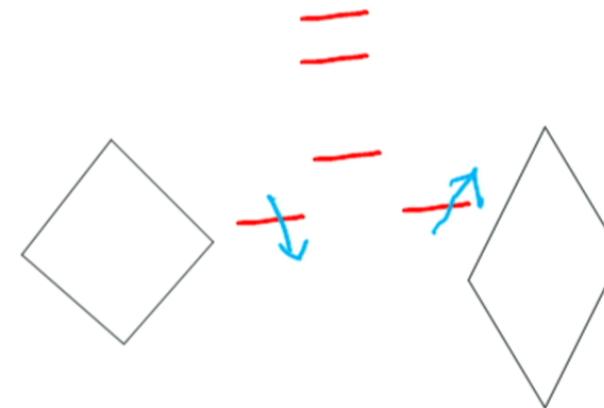


TiO



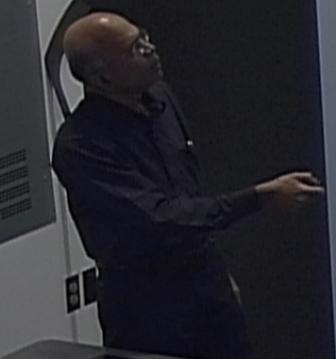
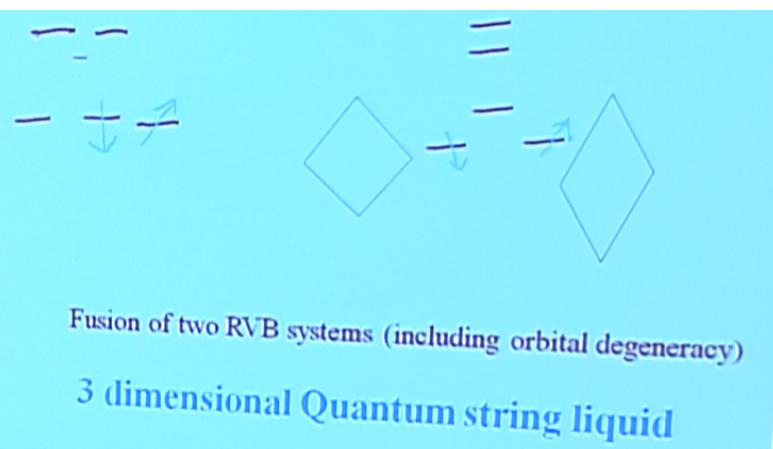
orbital degeneracy

How to avoid Jahn Teller distortion ?



Fusion of two RVB systems (including orbital degeneracy)

**3 dimensional Quantum string liquid**



## Theoretical Possibilities

$S = 1$

Quantum string liquid

Every site has two singlet bonds 2 of the 6 distinct neighbors



TiO

orbital degeneracy

How to avoid Jahn Teller distortion ?



orbital degeneracy

How to avoid JT distortion

$$|+\rangle = |xy\rangle + e^{\frac{i2\pi}{3}} |yz\rangle + e^{\frac{i4\pi}{3}} |zx\rangle$$

Chiral Spin liquid

TiO

orbital degeneracy

How to avoid Jahn Teller distortion ?



orbital degeneracy

How to avoid JT distortion

$$|+\rangle = |xy\rangle + e^{\frac{i\pi}{3}} |yz\rangle + e^{\frac{i\pi}{3}} |zx\rangle$$

Chiral Spin liquid

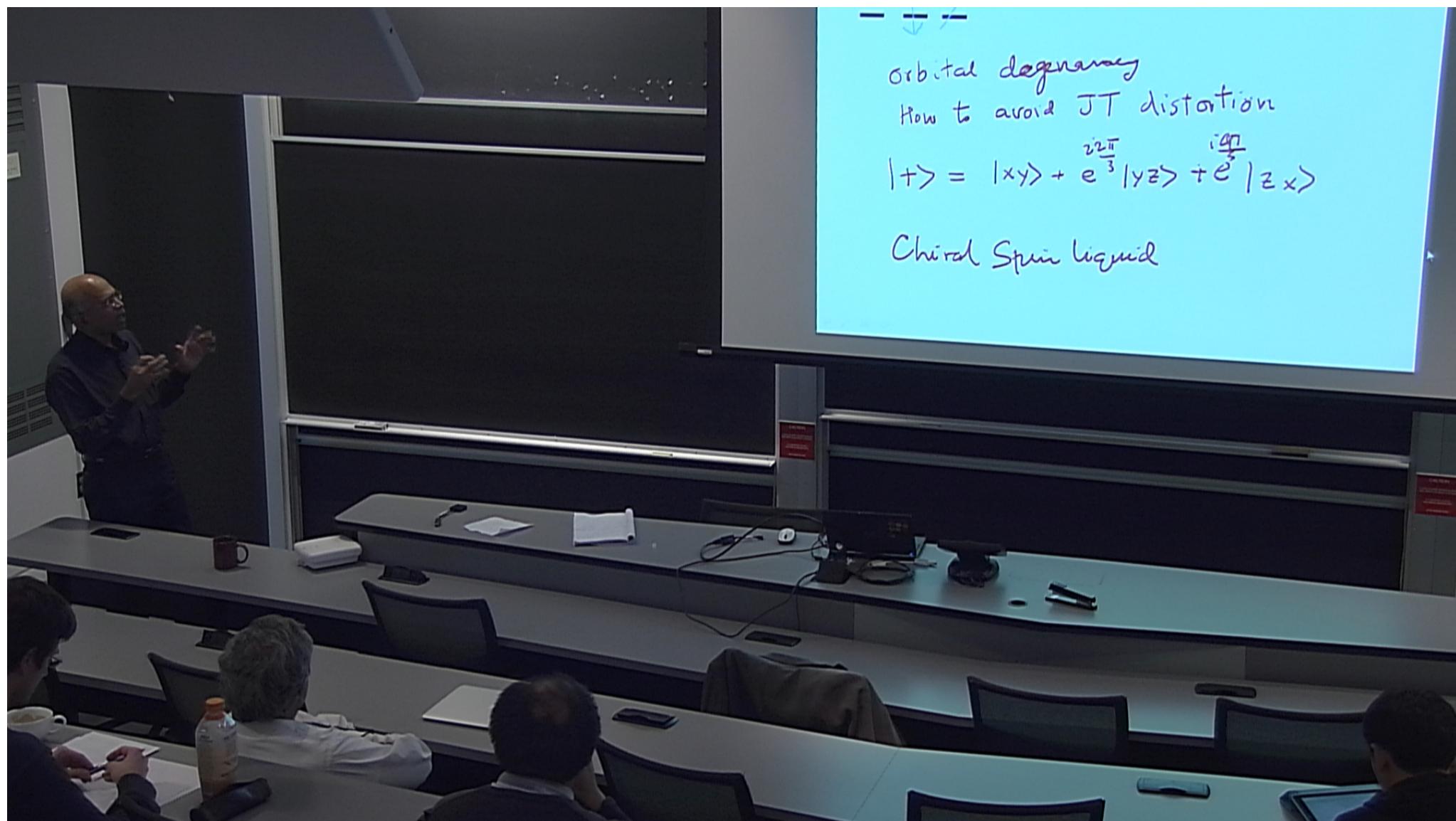
- ↓ /

orbital degeneracy

How to avoid JT distortion

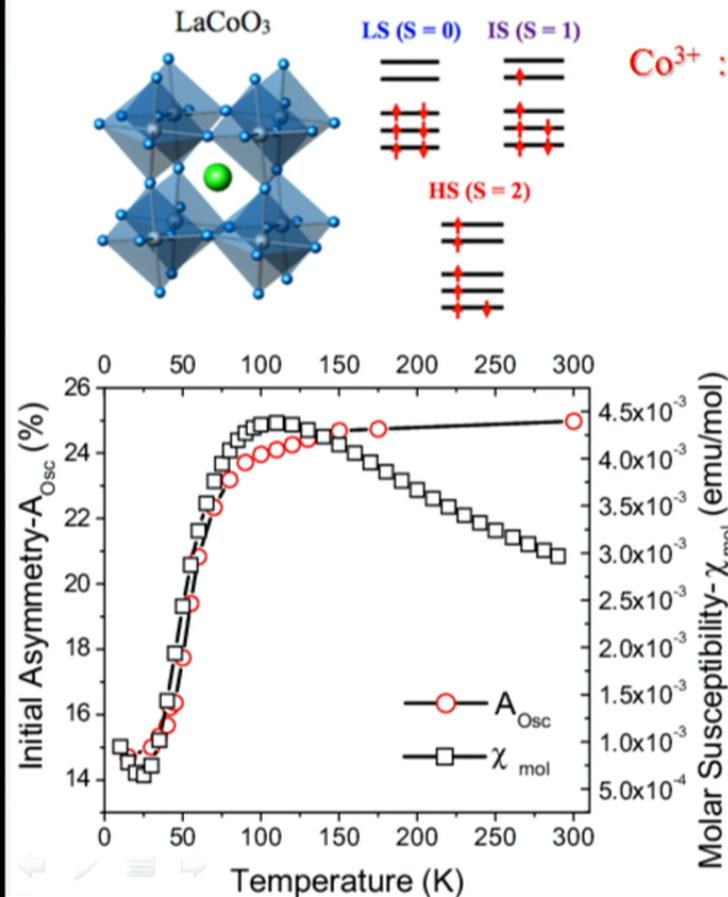
$$|+\rangle = |xy\rangle + e^{\frac{i2\pi}{3}}|yz\rangle + e^{\frac{i\pi}{3}}|zx\rangle$$

Chiral Spin liquid



## LaCoO<sub>3</sub>

## A cubic Perovskite and an old puzzle



No magnetic or structural phase transition  
At very high temperature a gradual metal insulator transition is observed

Local probes see below 100 K  
a predominantly S = 0 state  
S = 1 and S = 2 states grow gradually

Controversial theories

Doping and strain induce ferromagnetism

Bhatia, CNR Rao, Goodenough ...

## Theoretical Possibilities

$S = 1$

Quantum string liquid

Every site has two singlet bonds to 2 of the 6 distinct neighbors

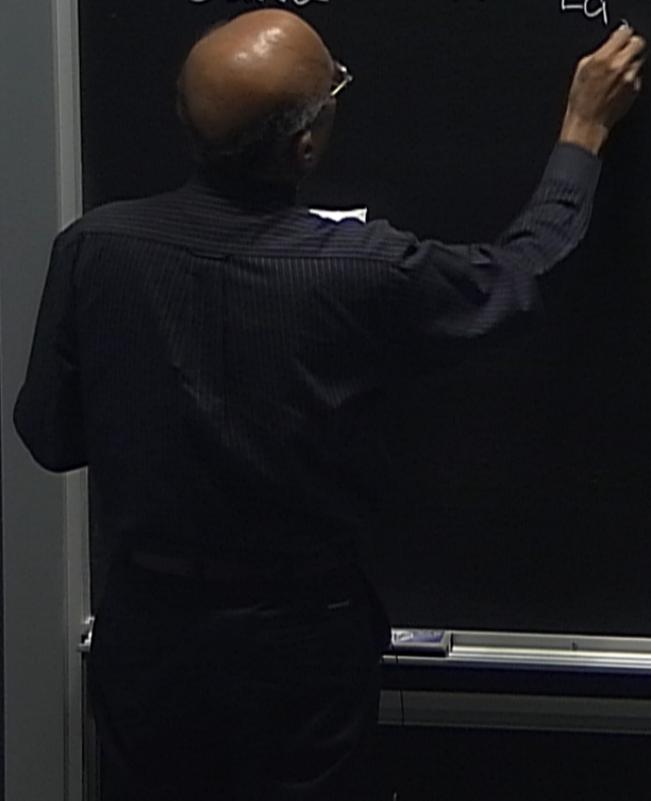
$S = 2$

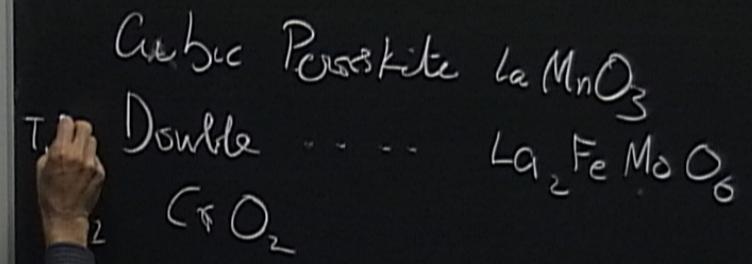
Quantum Branched Polymer Liquid

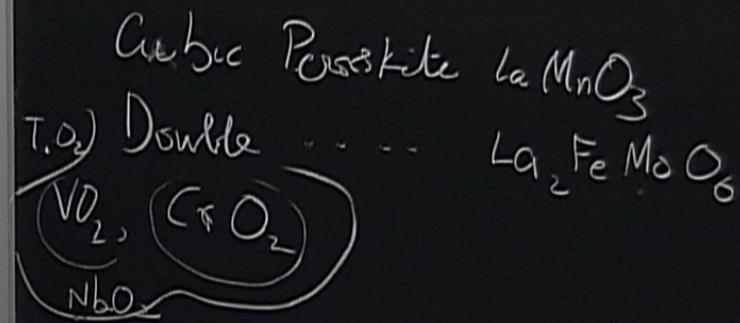
Every site four singlet bonds to 4 of the 6 distinct neighbors

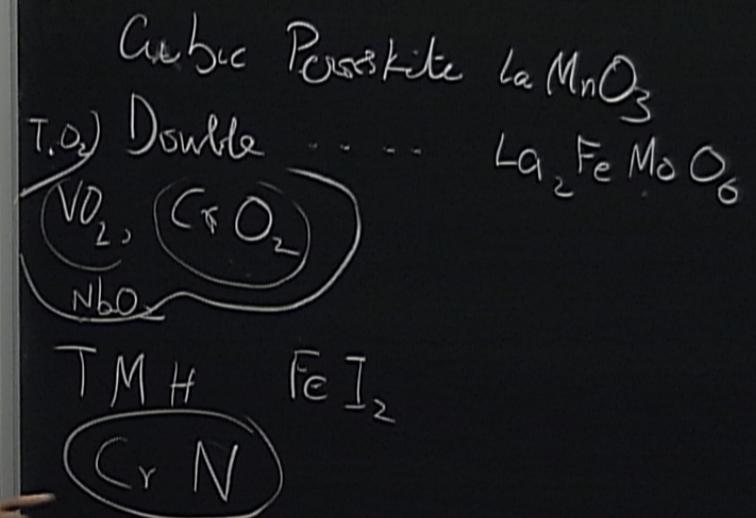


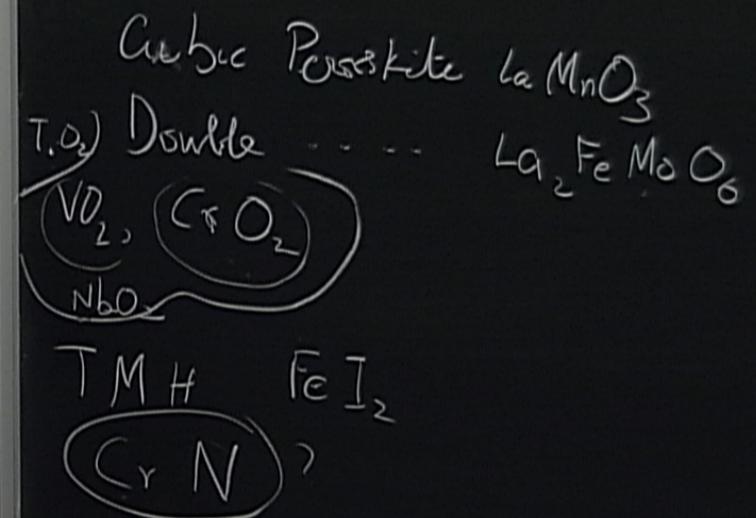
Cubic Perovskite  $\text{LaMnO}_3$   
Double . . . La











Professor speaking at a chalkboard:

- Cubic Perovskite  $\text{La}_3\text{MnO}_6$
- T.O. Double  $\text{La}_2\text{FeMoO}_6$
- $(\text{VO}_2, \text{CrO}_2)$   $\text{H}_2\text{O}$
- $\text{TM} + \text{FeI}_2$   $\text{N}$
- $\text{Hg}_2\text{Ru}_2\text{O}_7$

Diagram illustrating  $\text{Co}^{2+} : 3d^9$  states:

HS ( $S=2$ )

Local probes see see below 100 K  
a predominantly  $S=0$  state  
 $S=1$  and  $S=2$  states grow gradually

Controversial theories

Doping and strain induce ferromagnetism

Bhatia, CNR Rao, Goodenough ...

Graph showing Initial Asymmetry  $A_{\text{osc}}$  (%) and Molar Susceptibility  $\chi_{\text{mol}}$  (emu/mol) versus Temperature (K):

Temperature (K)	$A_{\text{osc}}$ (%)	$\chi_{\text{mol}}$ (emu/mol)
0	14.0	5.0e-4
50	16.0	1.0e-3
100	24.0	4.0e-3
150	24.5	4.5e-3
200	23.5	3.5e-3
250	21.0	2.5e-3
300	19.0	2.0e-3