Interplay of electronic, spin and orbital degrees of freedom

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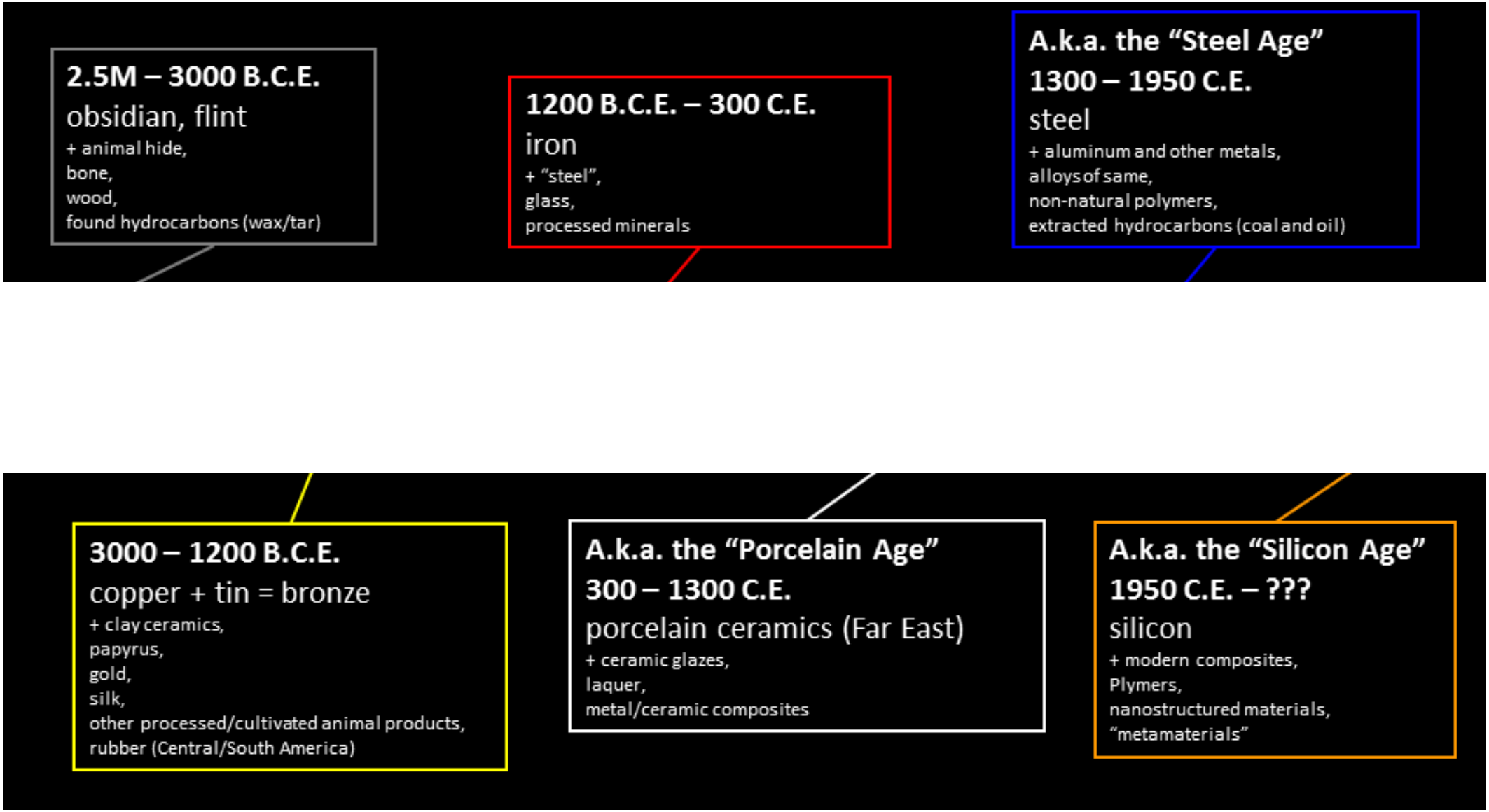
Nussinov and JvdB, RMP 87, 1 (2015) & arXiv:1303.5922

International School of Oxide Electronics
Cargese
25.06.2019

Materials in Time



Materials have defined Time

Outline

Interplay of electronic, spin and orbital degrees of freedom

PART 1

- *Partially filled electronic shells*
- *Atomic wavefunctions — orbitals*
- *Electron-electron interactions U and J_H*
- *Splitting of e_g and t_{2g} manifolds*
- *Spin vs. orbital degrees of freedom*
- *Mott-Hubbard and magnetism*

Outline

Relativistic oxide materials

PART 2

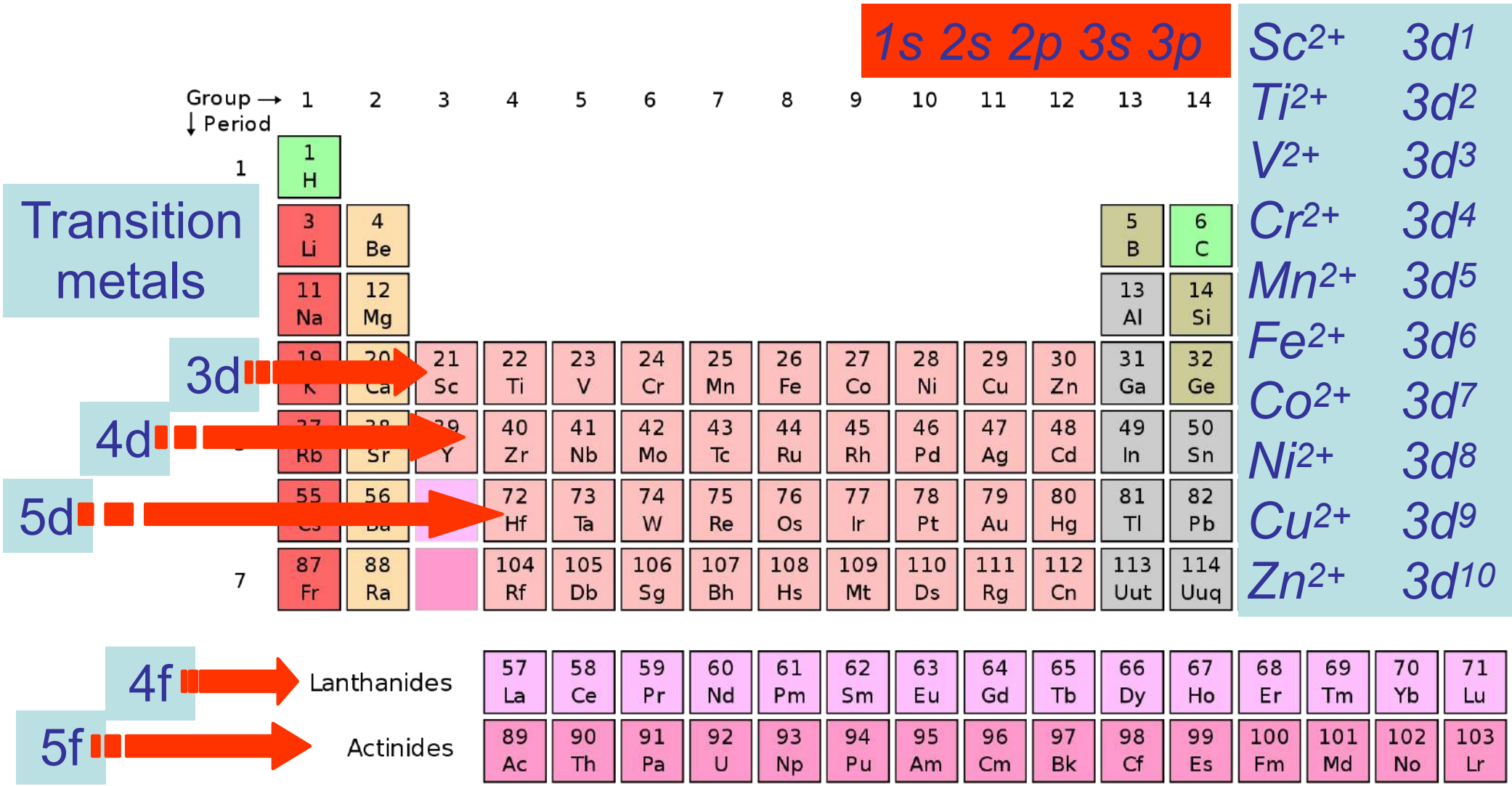
- *Superexchange with orbital d.o.f.'s*
- *The e_g Kugel-Khomskii Hamiltonian*
- *Goodenough-Kanamori-Anderson rules for superexchange*
- *Relativistic spin-orbit coupling*
- *Super exchange in iridates*
- *Honeycomb Kitaev model - spin liquid*
- *Topological quantum computing*

PART 1

INTRODUCTION

Partially filled atomic shells

Periodic Table of Elements



Sc^{2+} $3d^1$
Ti^{2+} $3d^2$
V^{2+} $3d^3$
Cr^{2+} $3d^4$
Mn^{2+} $3d^5$
Fe^{2+} $3d^6$
Co^{2+} $3d^7$
Ni^{2+} $3d^8$
Cu^{2+} $3d^9$
Zn^{2+} $3d^{10}$

Ti: $3d^2\ 4s^2$

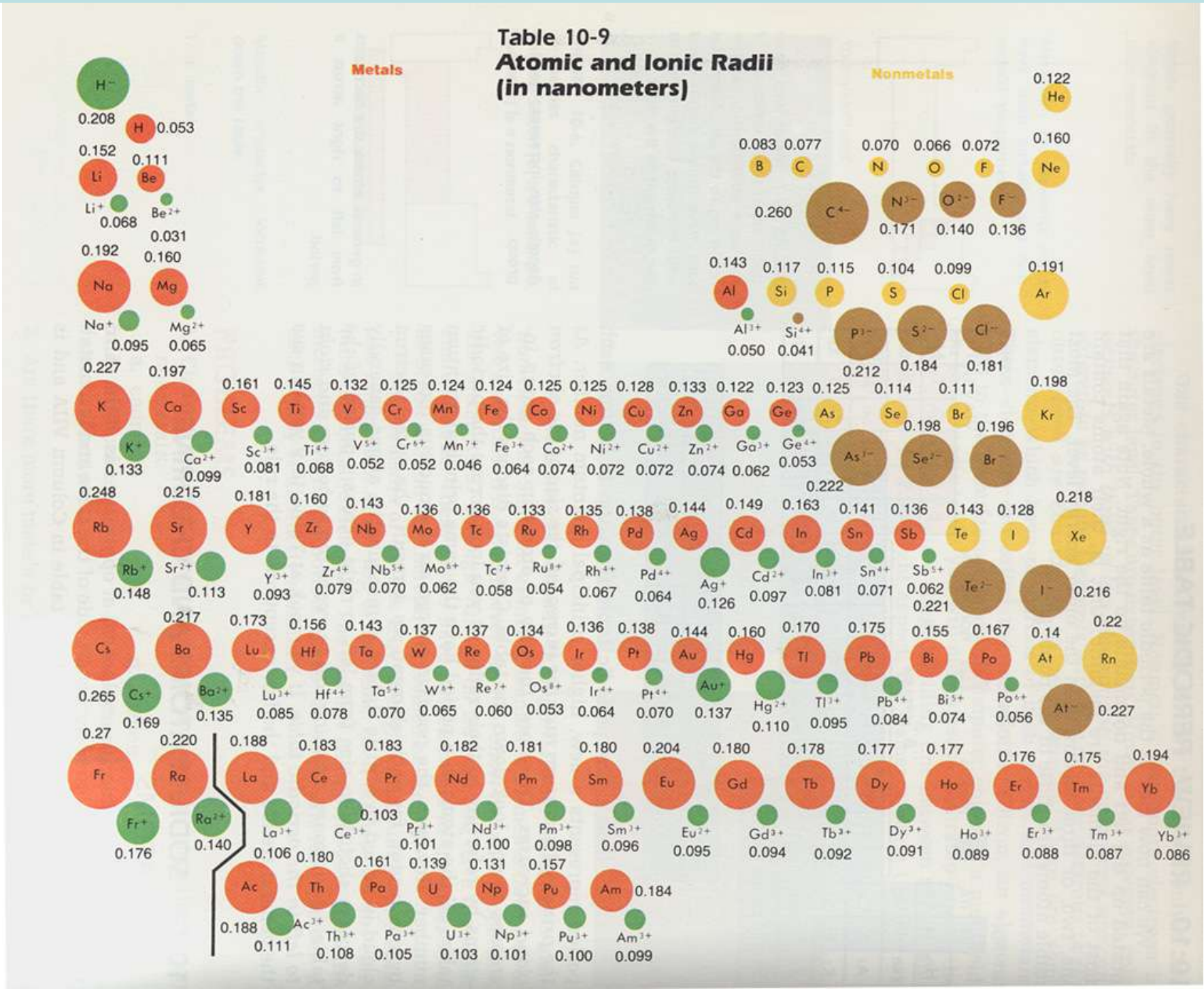
Ce: $4f^1\ 5d^1\ 6s^2$

radius wavefunction

$R(3d) << R(4s)$

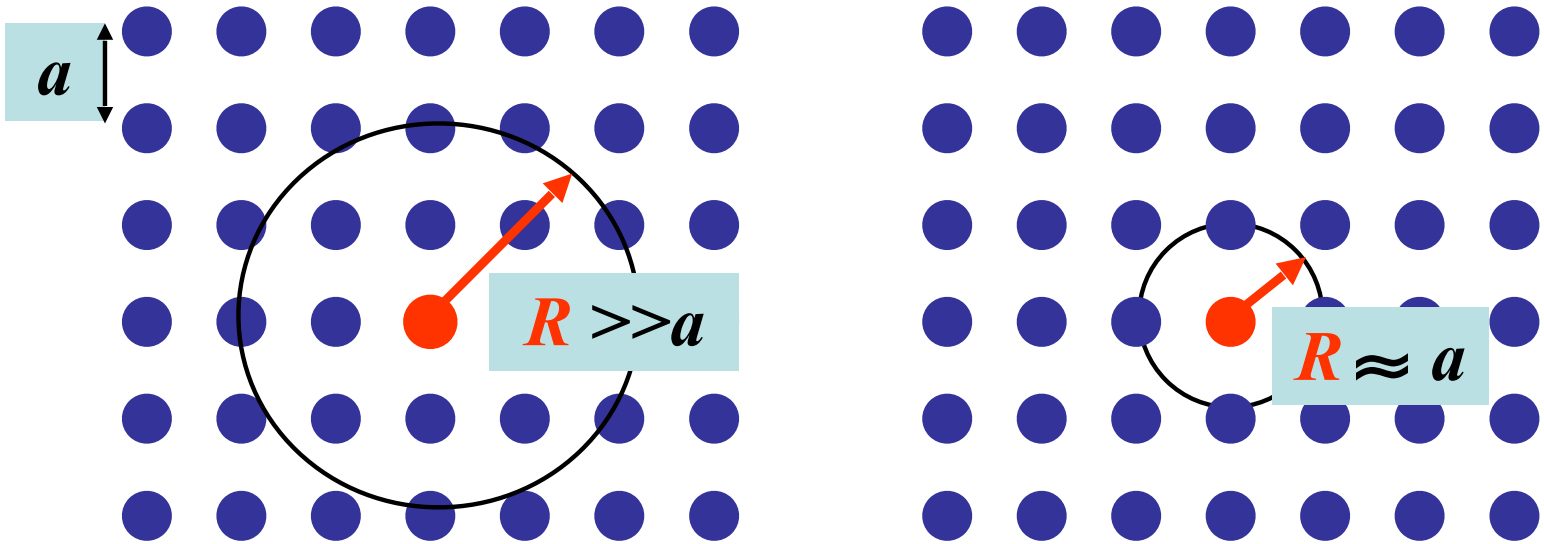
$R(4f) << R(6s)$

Atomic & Ionic Radii of Elements



Localized orbitals

High Tc copper oxides, manganites, iron-, chromium-, nickel-oxides…..



R >> a conventional metals, semiconductors
R ≈ a correlated electron systems

Do atomic physics first, include translation symmetry later

Small overlap of neighboring atomic wave functions:
'Electrons spend a long time on one atom and hop around infrequently'

atomic wavefunctions — orbitals

Wavefunction

The normalized position wavefunctions, given in spherical coordinates are:[5]

$$\psi_{n\ell m}(r,\vartheta,\varphi) = \sqrt{\left(\frac{2}{na_0}\right)^3 \frac{(n-\ell-1)!}{2n[(n+\ell)!]^3}} e^{-\rho/2} \rho^{\ell} L_{n-\ell-1}^{2\ell+1}(\rho) \cdot Y_{\ell}^{m}(\vartheta,\varphi)$$

where:

$$\rho = \frac{2r}{na_0},$$

a_0 is the Bohr radius,

$L_{n-\ell-1}^{2\ell+1}(\rho)$ are the generalized Laguerre polynomials of degree $n - \ell - 1$, and

$Y_{\ell}^{m}(\vartheta,\varphi)$ is a spherical harmonic function of degree ℓ and order m.

$$\psi_{nlm} = R_{nl} Y_l^m \qquad E_n = \frac{-13.6\ eV}{n^2}$$

Hydrogen atom

$$\psi_{n\ell m}(r,\vartheta,\varphi) = \sqrt{\left(\frac{2}{na_0}\right)^3 \frac{(n-\ell-1)!}{2n[(n+\ell)!]^3}} e^{-\rho/2} \rho^\ell L_{n-\ell-1}^{2\ell+1}(\rho) \cdot Y_\ell^m(\vartheta,\varphi)$$

The quantum numbers can take the following values:

$$n = 1, 2, 3, \ldots$$
$$\ell = 0, 1, 2, \ldots, n-1$$
$$m = -\ell, \ldots, \ell.$$

Additionally, these wavefunctions are orthogonal:

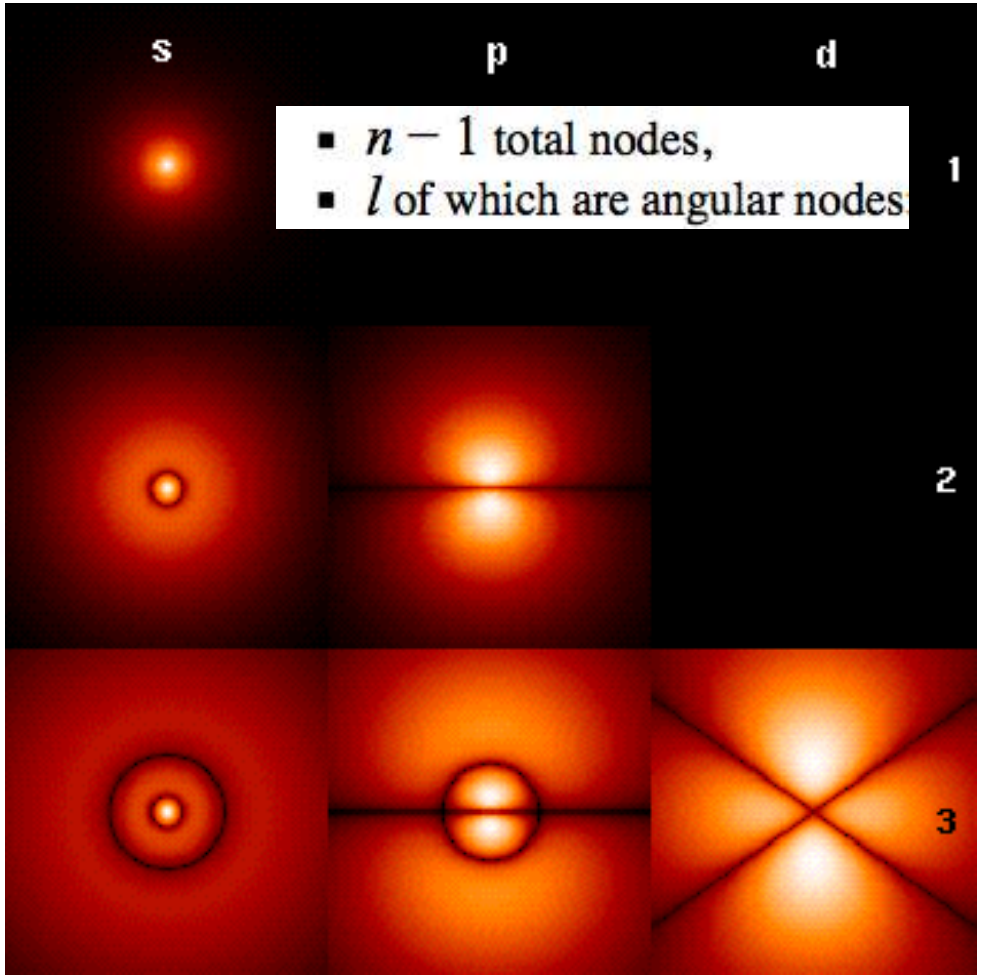
$$\langle n, \ell, m | n', \ell', m' \rangle = \delta_{nn'} \delta_{\ell\ell'} \delta_{mm'},$$

where $|n, \ell, m\rangle$ is the representation of the wavefunction $\psi_{n\ell m}$ in Dirac notation, and δ is the Kronecker delta function. [6]

Angular momentum

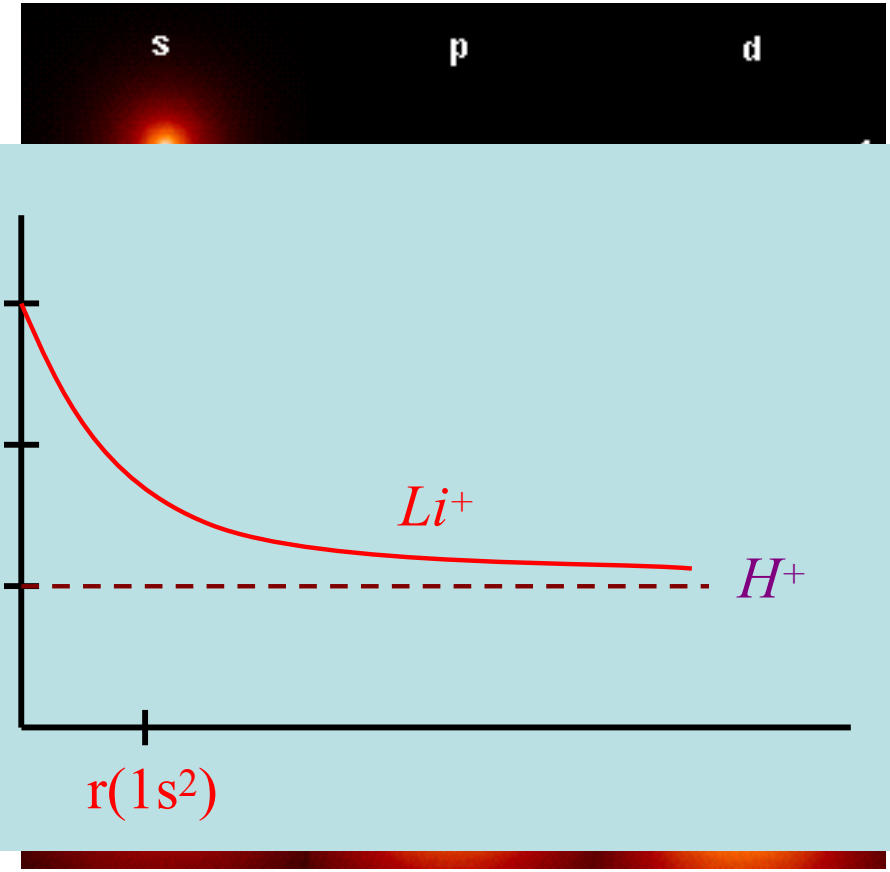
The eigenvalues for Angular momentum operator:

$$L^2 |n, \ell, m\rangle = \hbar^2 \ell(\ell+1) |n, \ell, m\rangle$$
$$L_z |n, \ell, m\rangle = \hbar m |n, \ell, m\rangle.$$



In multi-electron atoms degeneracy of s, p ,d , f states with same n is lifted

Screening of nuclear charge



Fill 1s with two electrons

next electron into 2s or 2p orbital?

we know the answer: lithium $1s^2 2s$

why?

the $1s^2$ core screens nuclear charge

2p orbital mostly outside $1s^2$ core → *attractive nuclear charge is well-screened* (Lithium 3+ → ~1+)

2s: considerable weight inside $1s^2$ core → *attractive nuclear charge screened less efficiently*

Aufbau principle: 1s 2s 2p 3s 3p 3d 4s

For 3d screening is so efficient that in TM atoms 4s already filled while 3d partially empty

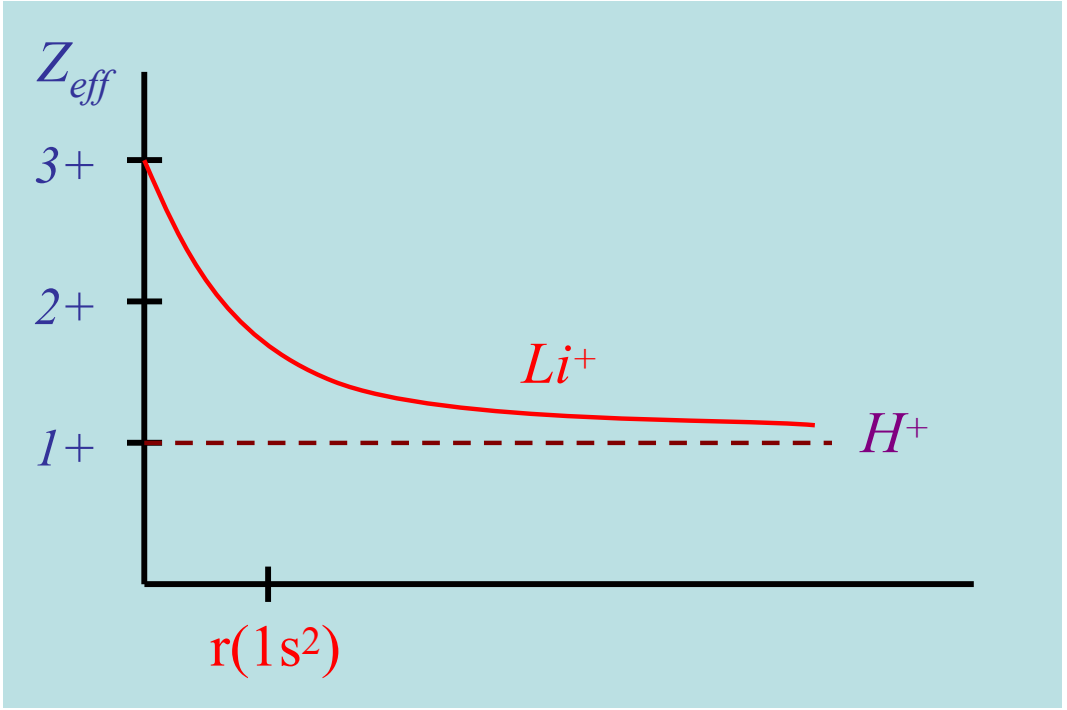
Contraction of orbitals

How can 3d electrons profit from large effective attractive potential close to the nucleus?

3d orbitals contract!

Can 3s orbitals contract too?

No, because radial nodes of 3s are fixed by orthogonality to 1s and 2s

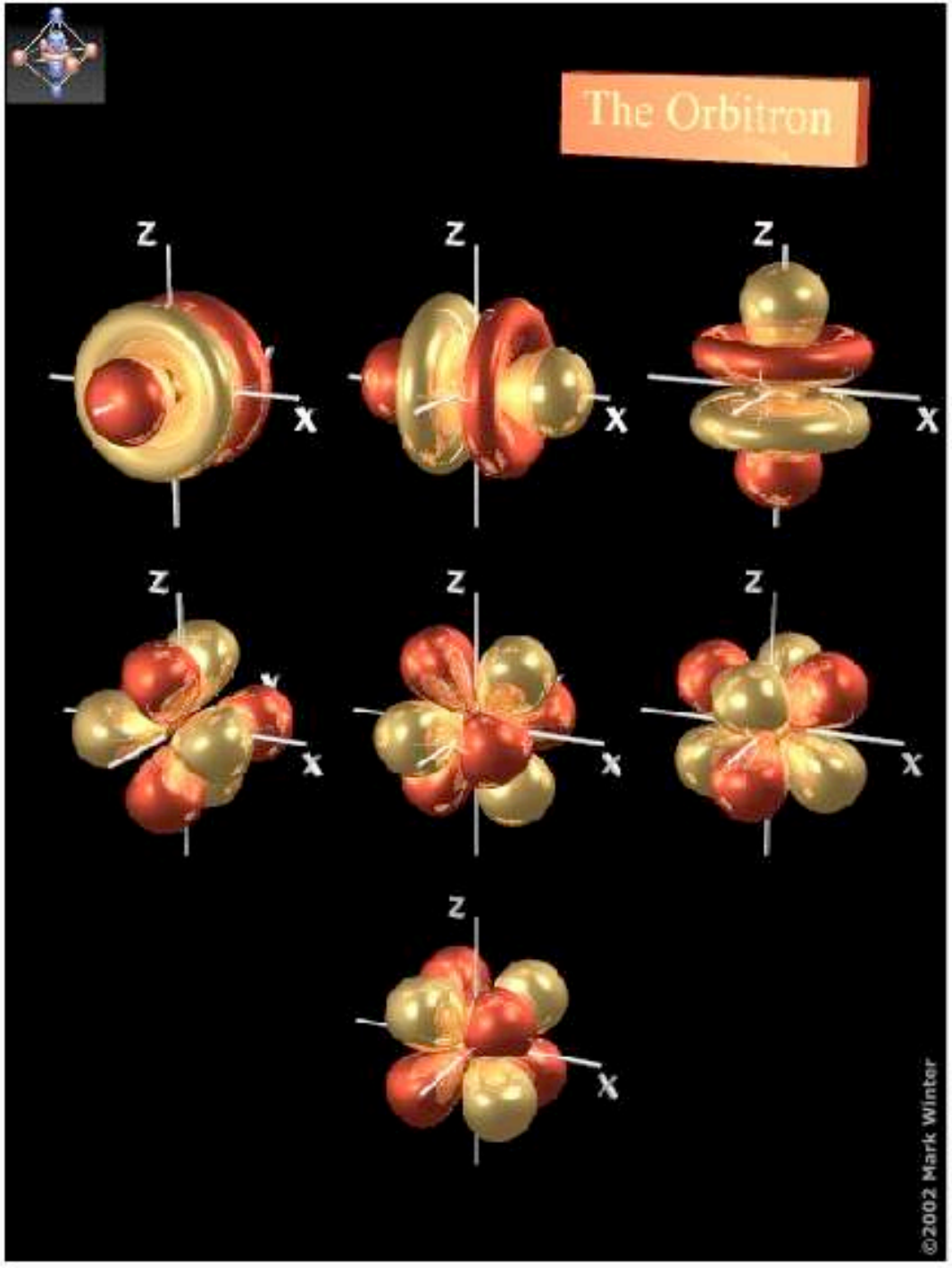


3d can contract because **angular** *wavefunction is orthogonal to filled orbitals*

4d & 5d therefore cannot contract much further....

...but 4f orbitals contract very much (can even be inside the core)

4f: (lanthanides)	very localized	Kondo-lattice models
3d: (row 4 transition metals) 5f: (actinides)	between localized and delocalized	Mott-Hubbard physics
4d & 5d TM's	rather delocalized	Mott-Hubbard	strong **L•S**



These orbitals are
NOT
the spherical harmonics
Y_l^m

1s	*2s*	*3s*	*4s*	*2p*	*3p*	*3d*	*4d*	*4f*

l=2

d-orbitals

$$Y_2^0 = \sqrt{\tfrac{5}{16\pi}}\left(3\cos^2\Theta - 1\right)$$

$$Y_2^1 = -\sqrt{\tfrac{15}{8\pi}}\sin\Theta\cos\Theta e^{i\phi}$$

$$Y_2^2 = \sqrt{\tfrac{15}{32\pi}}\sin^2\Theta e^{2i\phi}$$

$$Y_l^{-m} = (-1)^m\left(Y_l^m\right)^*$$

real wavefunctions:

$$Y_2^2 + Y_2^{-2} = \sqrt{\tfrac{15}{8\pi}}\sin^2\Theta\cos 2\phi$$

spherical coordinates:

$$x = r\sin\Theta\cos\phi$$

$$y = r\sin\Theta\sin\phi$$

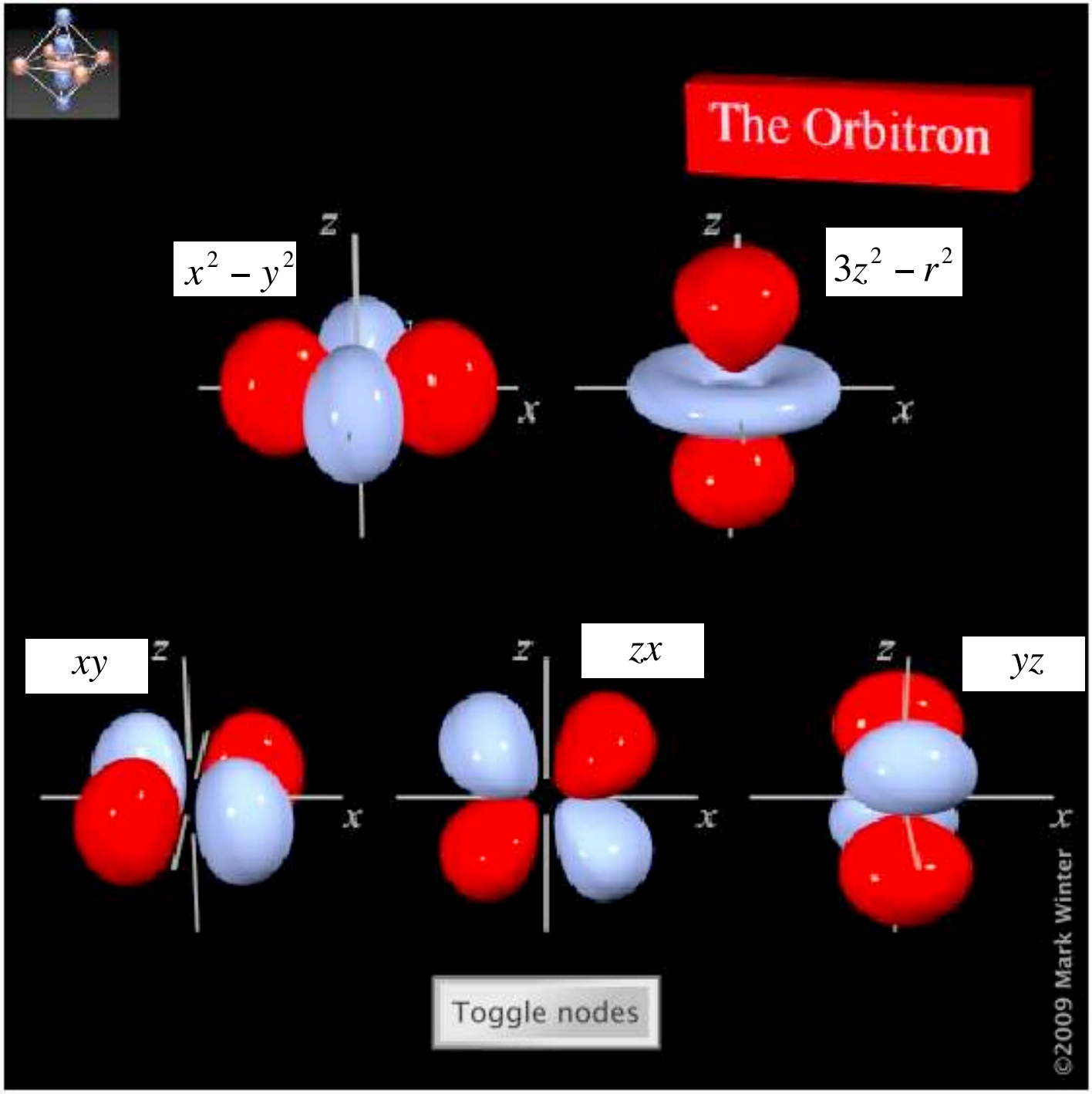
$$z = r\cos\Theta$$

$$\frac{Y_2^2 + Y_2^{-2}}{\sqrt{2}} = \sqrt{\tfrac{15}{16\pi}}\sin^2\Theta\cos 2\phi = \sqrt{\tfrac{15}{16\pi}}\sin^2\Theta\left(\cos^2\phi - \sin^2\phi\right) = \frac{\sqrt{\frac{15}{16\pi}}}{r^2}\left(x^2 - y^2\right)$$

$$Y_2^0 = \sqrt{\tfrac{5}{16\pi}}\left(3\cos^2\Theta - 1\right) = \frac{\sqrt{\frac{5}{16\pi}}}{r^2}\frac{1}{\sqrt{3}}\left(3z^2 - r^2\right)$$

e_g *orbitals*: $x^2 - y^2, \frac{1}{\sqrt{3}}\left(3z^2 - r^2\right)$ ***orbital doublet***

t_{2g} *orbitals*: $xy,\ yz,\ zx$ ***orbital triplet***


The Orbitron
$x^2 - y^2$
$3z^2 - r^2$
xy
zx
yz
z
x
Toggle nodes
©2009 Mark Winter

3d

electron-electron interactions

many-electron states *electron-electron interactions*

Full Hamiltonian: $\hat{H} = \hat{H}_{KE} + \hat{H}_2$

$$\hat{H}_2 = \frac{1}{2}\int d^3r d^3r' \sum_{\sigma,\sigma'} \psi^\dagger(\mathbf{r},\sigma)\psi^\dagger(\mathbf{r}',\sigma')v(|\mathbf{r}-\mathbf{r}'|)\psi(\mathbf{r}',\sigma')\psi(\mathbf{r},\sigma)$$

Coulomb interaction $v(|\mathbf{r}-\mathbf{r}'|) = \dfrac{e^2}{|\mathbf{r}-\mathbf{r}'|}$

where the $\psi(\mathbf{r})$ operators are the usual annihilation operators for an electron at position $\mathbf{r}$.

single - particle basisfunctions $\psi_{nlm} = R_{nl}Y_l^m$

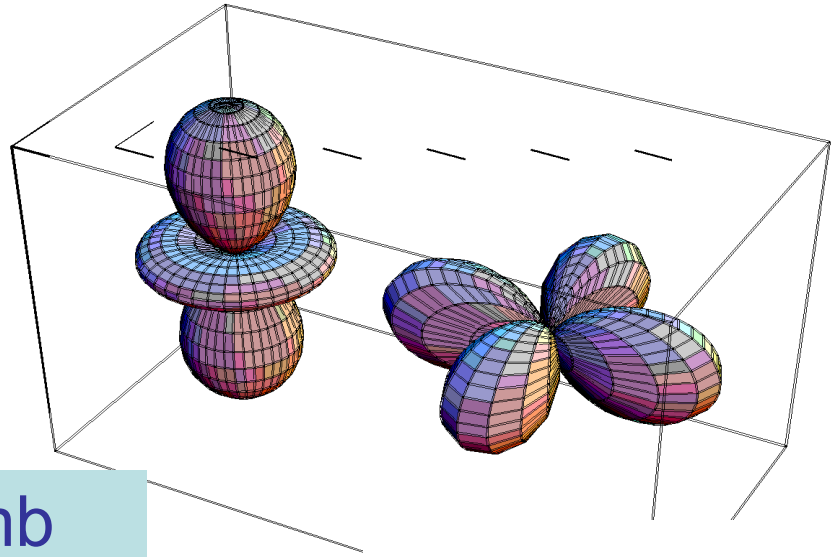
d - d interactions matrix elements $Y_2^{m1}(\sigma)Y_2^{m2}(\sigma')Y_2^{m3}(\sigma')Y_2^{m4}(\sigma)$ ➡

Coulomb ***U***

exchange J_H

3d orbitals of a TM-ion

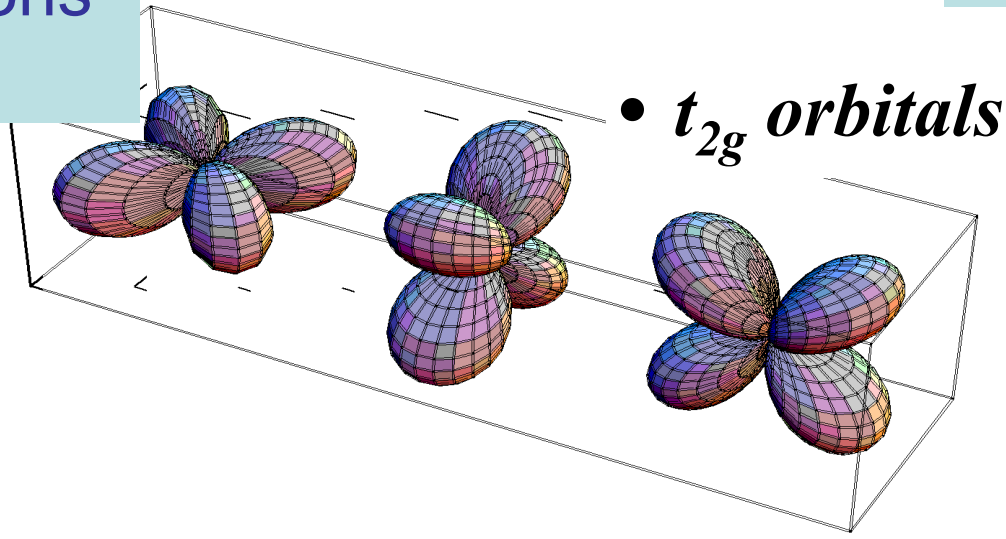
Large Atomic Hund's rule exchange J_H



- e_g *orbitals*

Electron Spins Parallel

Large Coulomb interaction ***U*** between electrons on the ion

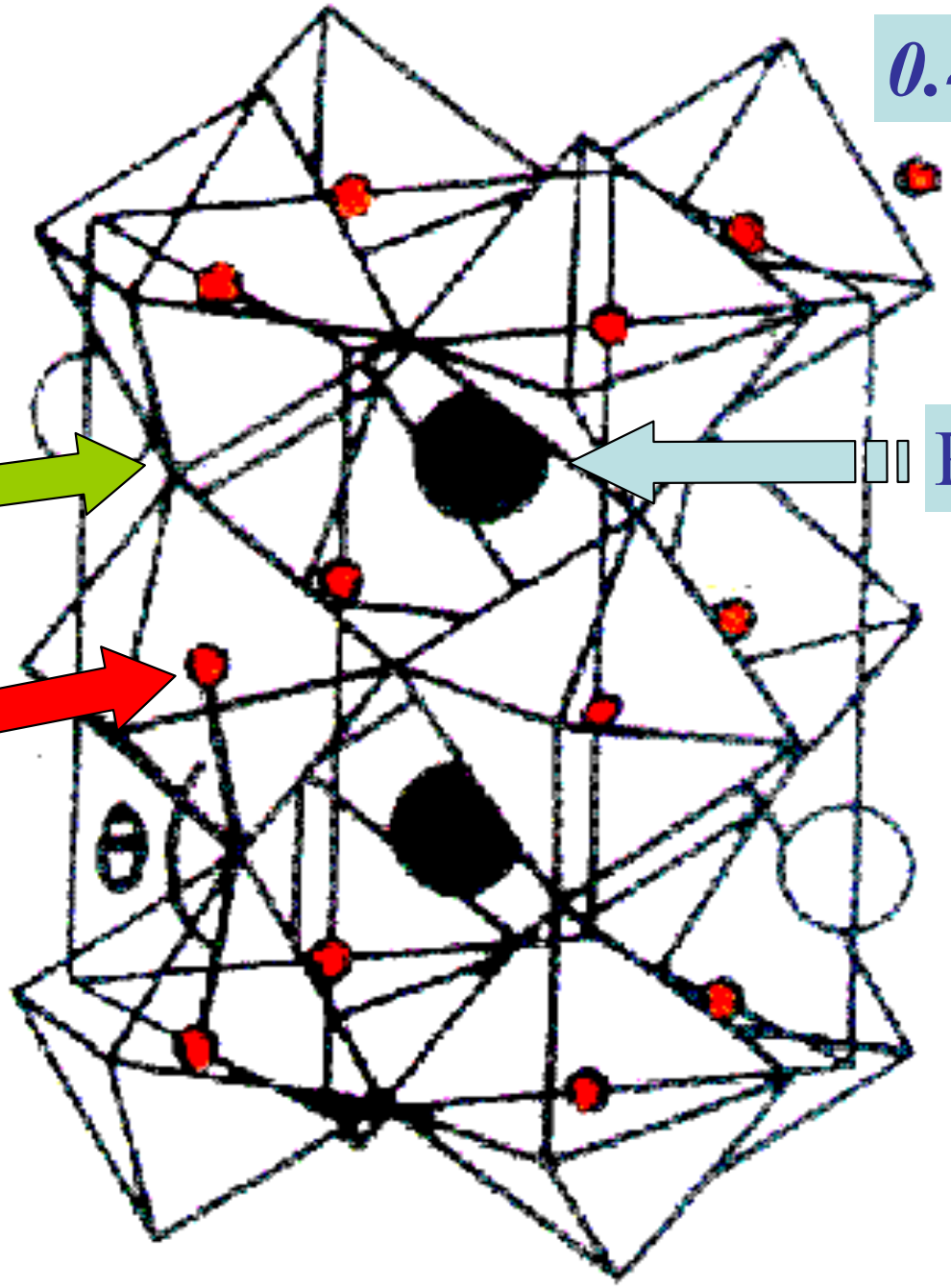


- t_{2g} *orbitals*

Splitting of e_g and t_{2g} manifolds: the crystal-field

Perovskite crystal structure of $Pr_{1-x}Ca_xMnO_3$

0.4 < x < 0.5



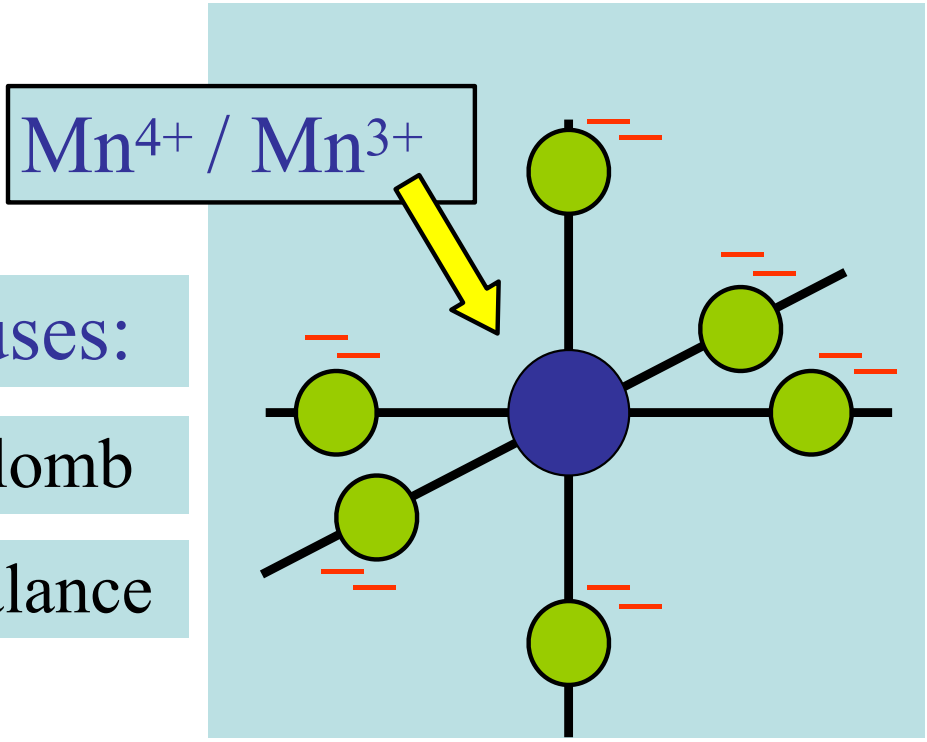
Oxygen^{2-}

Pr^{3+}/Ca^{2+}

Mn^{4+} / Mn^{3+}

Local considerations

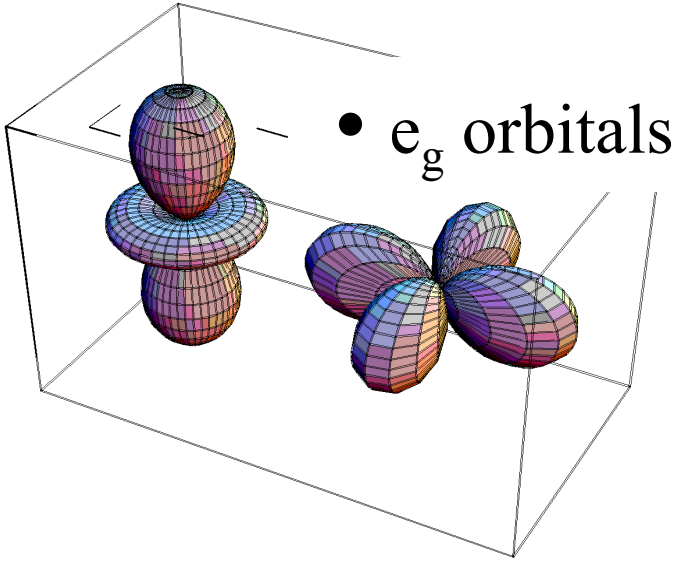
Cubic Crystal field splitting: 10 Dq



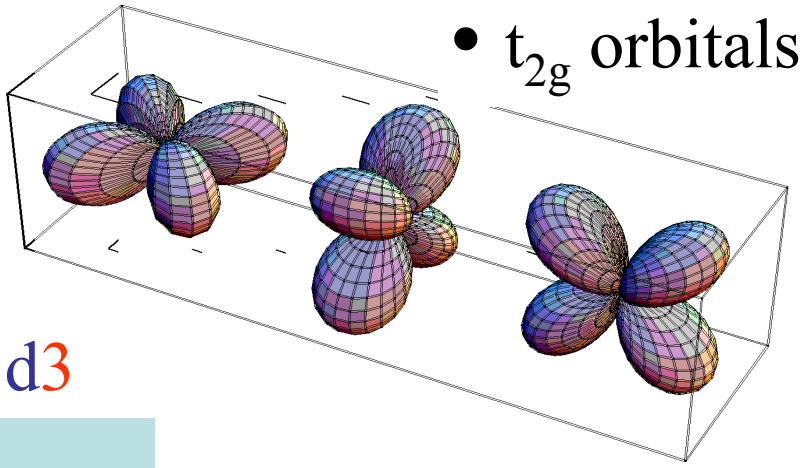
Causes:

Coulomb

Covalance

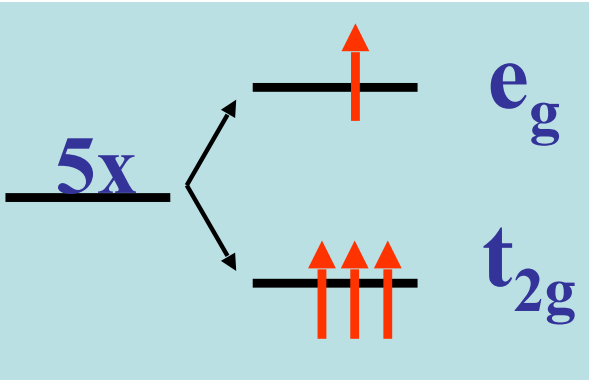


- e_g orbitals

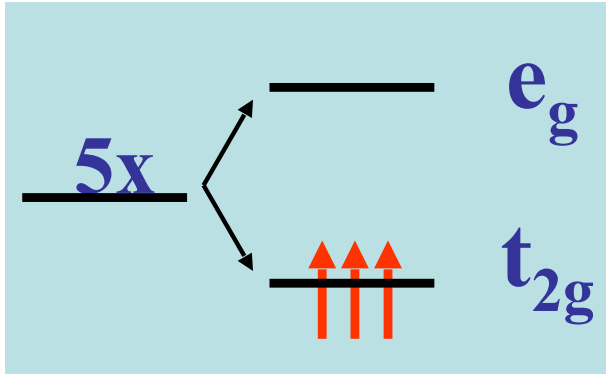


- t_{2g} orbitals

Mn (3+) = 3d4

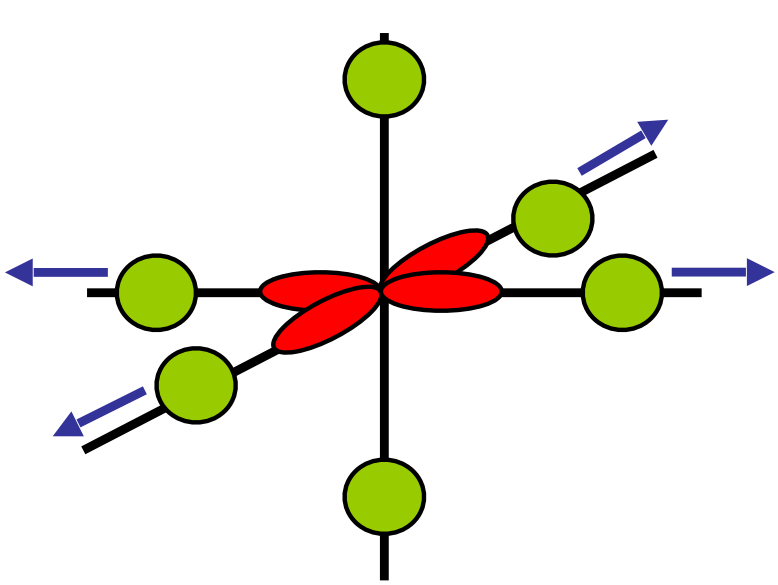


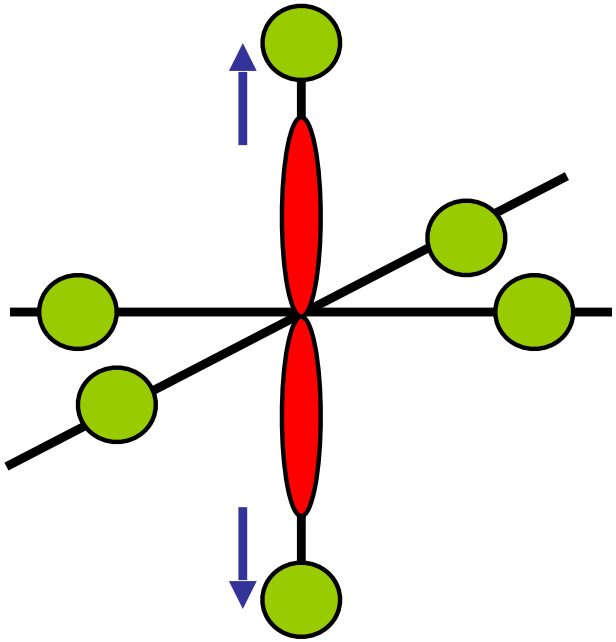
Mn (4+) = 3d3

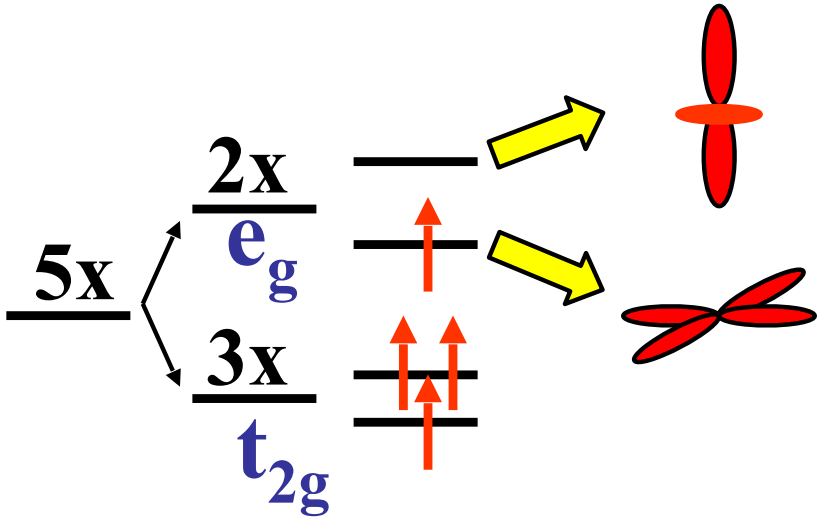


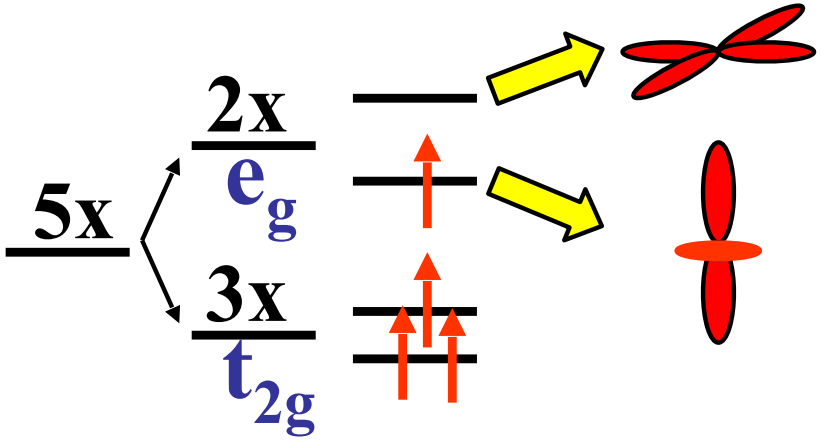
Lifting of degeneracy: lattice

Crystal field splitting of e_g levels → Jahn-Teller distortion









Spin vs. orbital degrees of freedom

Orbitals are extra degree of freedom
Impact on physical properties

- *Order-disorder*
- *Thermodynamics*
- *Magnetism*
- *Lattice distortions*

Orbitals behave like electron spins

Compare orbitals and spins....

Orbitals and spins

Similarities

Localized moment
emergent from electron-electron interactions

Angular momentum SU(2) algebra: $[S^x, S^y] = iS^z$

Possibility of long range ordering

Spin-spin and orbital-orbital interaction due to superexchange

Orbitals and spins

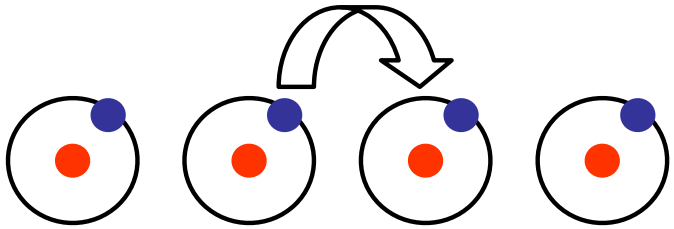
Differences

Spins		*Orbitals*
Weak	*coupling to lattice*	*Strong*
High	*Symmetry of Hamiltonian*	*Low*
Gapless	*Excitations*	*Gaped*
Sometimes	*Frustration of order*	*Always*

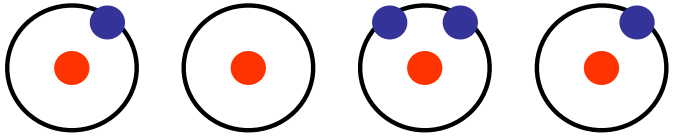
Non-local correlation effects: Mott-Hubbard and magnetism

Hubbard model

Consider array of Hydrogen atoms



Hopping amplitude: ***t***

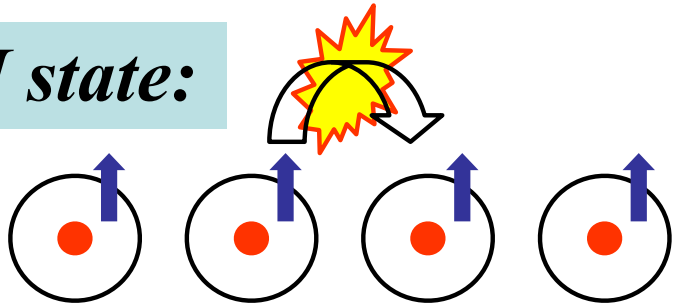


Coulomb interaction: ***U***

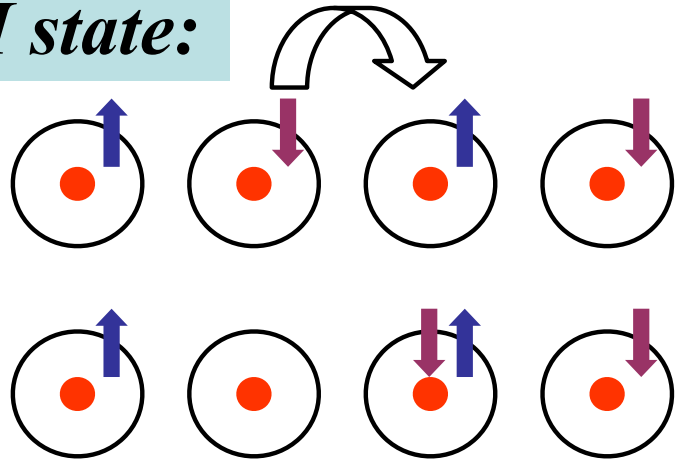
U = 0 ***Bands: Metallic behaviour***

U >> t ***Mott-Hubbard Insulator*** ➡ Antiferromagnetism

FM state:



AFM state:

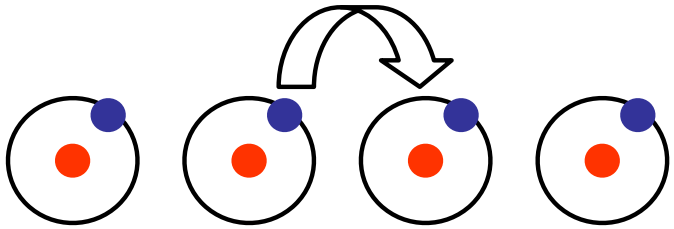


E = 0

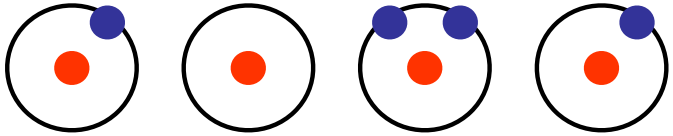
E = -t²/U

$$H_{Heisenberg} = J \sum_{ij} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \tfrac{1}{4} \right); \qquad J = \frac{4t^2}{U}$$

Hubbard model ***Consider array of Hydrogen atoms***



Hopping amplitude: ***t***



Coulomb interaction: ***U***

U = 0 ***Bands: Metallic behaviour***

U >> t ***Mott-Hubbard Insulator*** ➡ Antiferromagnetism

Heisenberg Hamiltonian

$$H_{Heis} = J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j \qquad [S^x, S^y] = iS^z$$

⬇

Rotational invariant

In real materials beyond 1s: orbital d.o.f.'s

(easy axis) exchange anisotropy

Outline

Relativistic oxide materials

PART 2

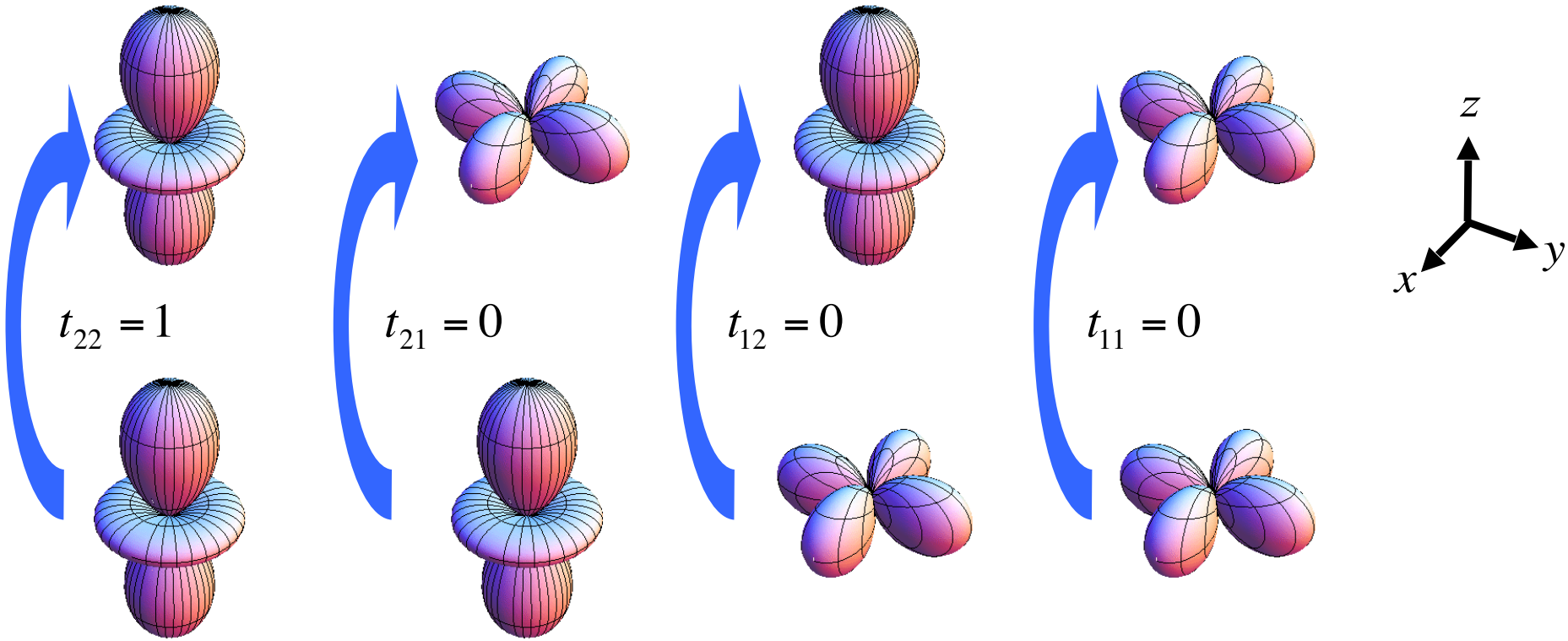
- *Superexchange with orbital d.o.f.'s*
- *The e_g Kugel-Khomskii Hamiltonian*
- *Goodenough-Kanamori-Anderson rules for superexchange*
- *Relativistic spin-orbit coupling*
- *Super exchange in iridates*
- *Honeycomb Kitaev model - spin liquid*
- *Topological quantum computing*

PART 2

Superexchange with orbital d.o.f.'s

The e_g Kugel-Khomskii Hamiltonian

electronic orbital-orbital hopping $t_{\alpha\beta}$ *for e_g orbitals*



hopping along **z**-axis

$$t^{\gamma}_{\alpha\beta} = \begin{pmatrix} & \\ & 1 \end{pmatrix};\ \gamma = z \qquad t^{x} = \hat{R}_2^{T} t^{z} \hat{R}_2;\ t^{y} = \hat{R}_2^{T} t^{y} \hat{R}_2$$

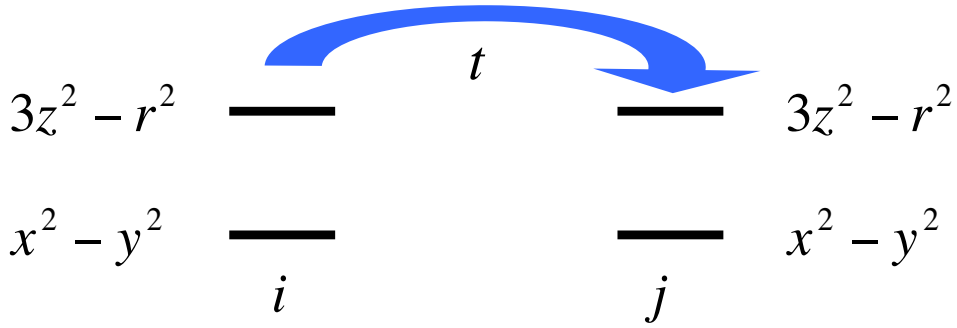
R_2: rotates e_g orbitals by $\varphi/2=2\pi/3$

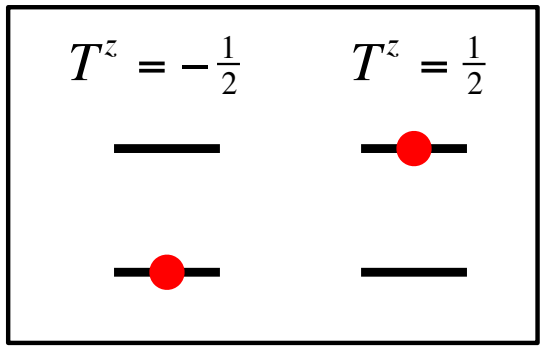
$$t^{x} = \tfrac{1}{4}\begin{pmatrix} -1 & -\sqrt{3} \\ \sqrt{3} & -1 \end{pmatrix}\begin{pmatrix} & \\ & 1 \end{pmatrix}\begin{pmatrix} -1 & \sqrt{3} \\ -\sqrt{3} & -1 \end{pmatrix} = \tfrac{1}{4}\begin{pmatrix} 3 & \sqrt{3} \\ \sqrt{3} & 1 \end{pmatrix}$$

$$t^{y} = \tfrac{1}{4}\begin{pmatrix} 3 & -\sqrt{3} \\ -\sqrt{3} & 1 \end{pmatrix}$$

Superexchange in presence of e_g orbitals

consider 2 sites (*i* and *j*) with each two e_g orbitals and one spin-less fermion





when 2 electrons on same site (and by definition in different orbitals): energy U

energy gain $-\frac{t^2}{U}$ possible if $T_i^z = \frac{1}{2}$ and $T_j^z = -\frac{1}{2}$

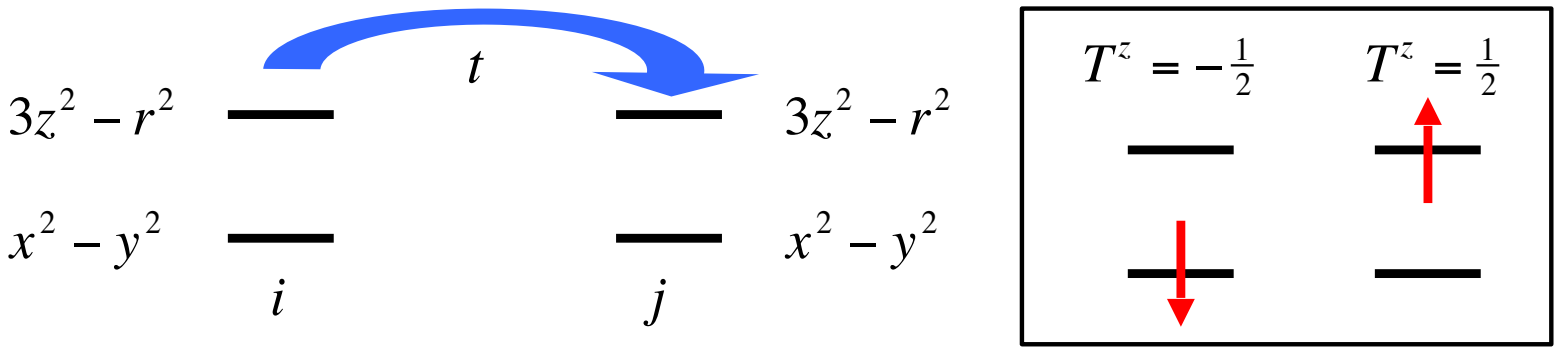
or $T_i^z = -\frac{1}{2}$ and $T_j^z = \frac{1}{2}$

$$H_{ij}^z = -\frac{t^2}{U}\left[\left(\frac{1}{2}+T_i^z\right)\left(\frac{1}{2}-T_j^z\right)+\left(\frac{1}{2}-T_i^z\right)\left(\frac{1}{2}+T_j^z\right)\right] \text{ with } J=\frac{4t^2}{U} \text{ this is } \boxed{H_{ij}^z = \frac{J}{2}\left(T_i^z T_j^z - \frac{1}{4}\right)}$$

Superexchange in presence of e_g orbitals *with spin d.o.f.'s*

Kugel-Khomskii Hamiltonians

consider 2 sites (*i* and *j*) with each two e_g orbitals and one spin-full fermion



2 electrons on same site: energy U — for the moment do not consider J_H

2 electrons in different orbitals: $H_{ij}^z = \frac{J}{2} T_i^z T_j^z$ (spin independent)

2 electrons in $3z^2$-r^2 orbital: regular spin superexchange $J\left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4}\right);\ J = \frac{4t^2}{U}$

$$H_{ij}^z = J\left(\mathbf{S}_i \cdot \mathbf{S}_j - \tfrac{1}{4}\right)\left(\tfrac{1}{2} + T_i^z\right)\left(\tfrac{1}{2} + T_j^z\right) + \tfrac{J}{2} T_i^z T_j^z$$

$$= J\left(\mathbf{S}_i \cdot \mathbf{S}_j - \tfrac{1}{4}\right)\left(\tfrac{1}{2} + T_i^z\right)\left(\tfrac{1}{2} + T_j^z\right) + \tfrac{J}{2}\left[\left(\tfrac{1}{2} + T_i^z\right)\left(\tfrac{1}{2} + T_j^z\right) - \tfrac{1}{2}\left(\tfrac{1}{2} + T_i^z + T_j^z\right)\right]$$

$$= J\left(\mathbf{S}_i \cdot \mathbf{S}_j + \tfrac{1}{4}\right)\left(\tfrac{1}{2} + T_i^z\right)\left(\tfrac{1}{2} + T_j^z\right) - \tfrac{J}{4}\left(\tfrac{1}{2} + T_i^z + T_j^z\right)$$

Superexchange in presence of spins in e_g orbitals

Kugel-Khomskii Hamiltonians

$$H^z = J\sum_{ij}\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{1}{4}\right)\left(\tfrac{1}{2}+T_i^z\right)\left(\tfrac{1}{2}+T_j^z\right)-\tfrac{1}{4}\left(\tfrac{1}{2}+T_i^z+T_j^z\right)$$ and H^x, H^y by rotation

defines the e_g Kugel-Khomksii model Hamiltonian:

$$H_{e_g}^{KK} = J\sum_{i\gamma}\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{1}{4}\right)\left(\tfrac{1}{2}+T_i^{\gamma}\right)\left(\tfrac{1}{2}+T_{i+\mathbf{e}_\gamma}^{\gamma}\right)$$

upto a constant

because $\sum_{\gamma} T_i^{\gamma} = 0$

with: $T^{\gamma} = T^z\cos\Theta_\gamma + T^x\sin\Theta_\gamma$ and $\{\Theta_\gamma\} = \left\{0, \frac{2\pi}{3}, \frac{4\pi}{3}\right\}$

$\gamma = 1,2,3$ and $\{\mathbf{e}_\gamma\} = \{\mathbf{e}_x, \mathbf{e}_y, \mathbf{e}_z\}$ the cubic unit vectors

Kugel & Khomskii, Sov. Phys. Usp. 25, 231 (1982)

How do spin and orbital order?

$$H_{e_g}^{KK} = J \sum_{i\gamma} \left(\mathbf{S}_i \cdot \mathbf{S}_j + \tfrac{1}{4}\right)\left(\tfrac{1}{2} + T_i^{\gamma}\right)\left(\tfrac{1}{2} + T_{i+\mathbf{e}_\gamma}^{\gamma}\right)$$

≥ 0

Consider perfect Neel order $\mathbf{S}_i \cdot \mathbf{S}_j = -\frac{1}{4}$ → *H* vanishes!

→ spin ordering must be antiferromagnetic

(theoretical) solution: orbital ordering such that "1D" spin chains form, T^z

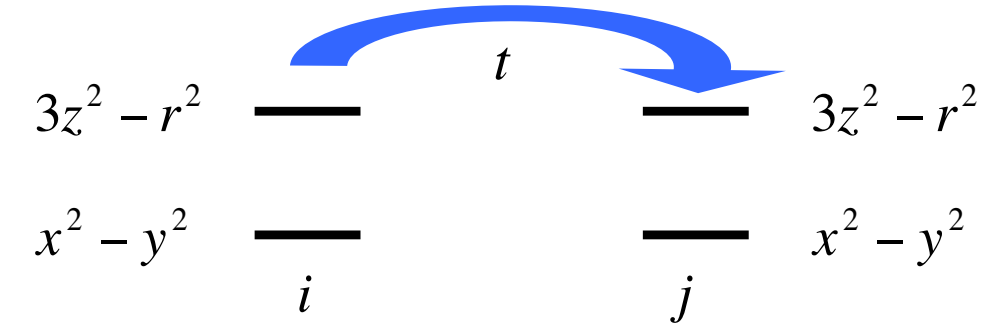
along such a chain $\left\langle \mathbf{S}_i \cdot \mathbf{S}_j \right\rangle = \frac{1}{4} - \ln 2$

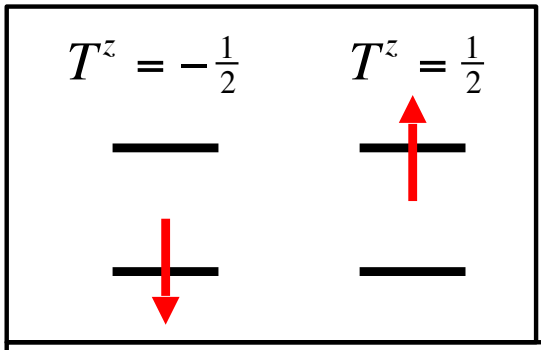
→ ordering arises from interplay of spin and orbital *fluctuations*

Khaliullin & Oudovenko, PRB 56, R14243 (1997)

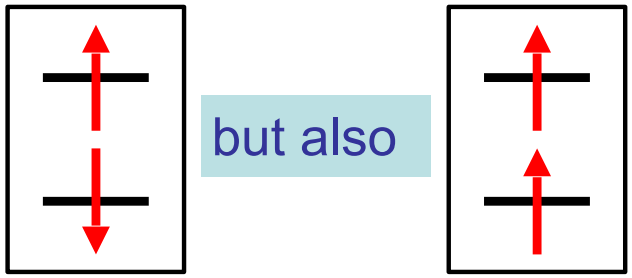
Finite J_H superexchange with spins and *e_g orbitals*

consider 2 sites (*i* and *j*) with each two e_g orbitals and one spin-full fermion





$$H^{KK}_{e_g} = J\sum_{i\gamma}\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{1}{4}\right)\left(\tfrac{1}{2}+T^{\gamma}_i\right)\left(\tfrac{1}{2}+T^{\gamma}_{i+\mathbf{e}_\gamma}\right)$$



take care of ηJ part

$$\tilde{H}^z_{ij} = \frac{\eta J}{2}\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{3}{4}\right)\left(T^z_iT^z_j-\tfrac{1}{4}\right)$$

exchange:

$$J=\frac{4t^2}{U} \qquad \frac{4t^2}{U-J_H}\approx\left(1-\frac{J_H}{U}\right)\frac{4t^2}{U}=(1-\eta)J$$

$$H^{KK} = J\sum_{i\gamma}\left[\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{1}{4}\right)\left(\tfrac{1}{2}+T^{\gamma}_i\right)\left(\tfrac{1}{2}+T^{\gamma}_{i+\mathbf{e}_\gamma}\right)+\eta\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{3}{4}\right)\left(T^{\gamma}_iT^{\gamma}_{i+\mathbf{e}_\gamma}-\tfrac{1}{4}\right)\right]$$

spin and *orbitals order*

$$H^{KK} = J\sum_{i\gamma}\left[\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{1}{4}\right)\left(\tfrac{1}{2}+T_i^{\gamma}\right)\left(\tfrac{1}{2}+T_{i+\mathbf{e}_\gamma}^{\gamma}\right)+\eta\left(\mathbf{S}_i\cdot\mathbf{S}_j+\tfrac{3}{4}\right)\left(T_i^{\gamma}T_{i+\mathbf{e}_\gamma}^{\gamma}-\tfrac{1}{4}\right)\right]$$

different orbitals occupied $T_i^z T_j^z - \frac{1}{4} < 0$ ➡ spin exchange is ferromagnetic

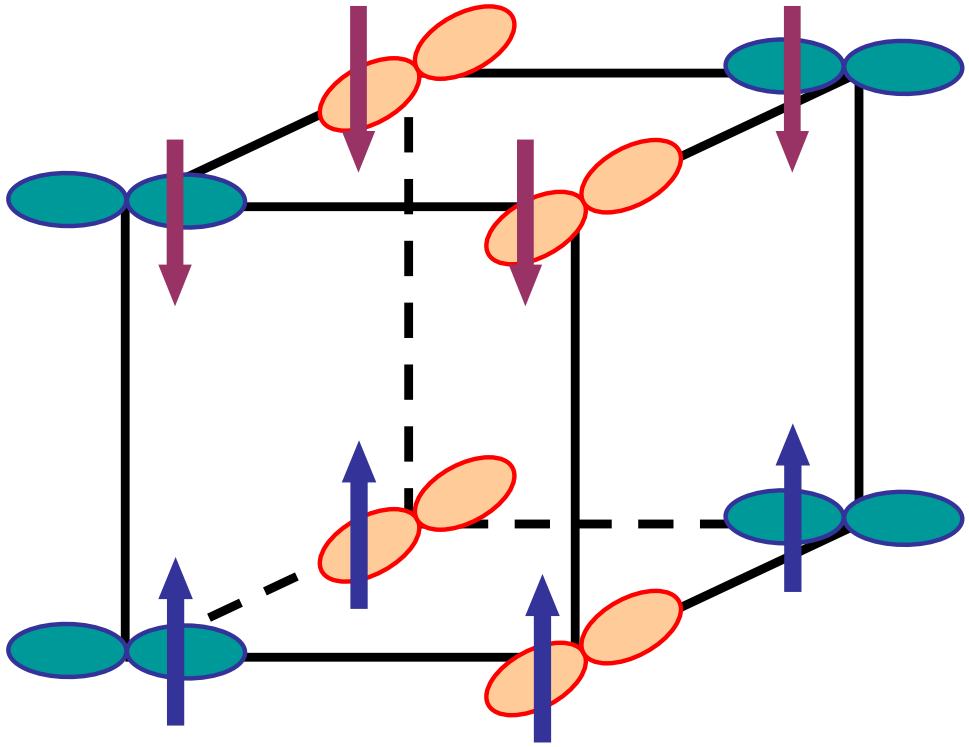
same orbitals occupied $T_i^z T_j^z - \frac{1}{4} = 0$ ➡ spin exchange is antiferro

very general result

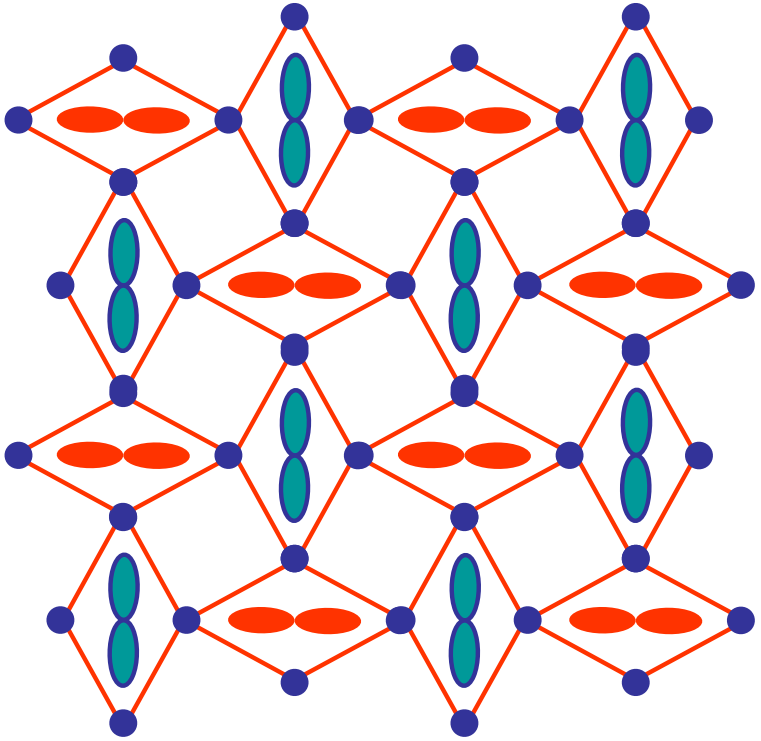
Goodenough-Kanamori-Anderson rules for superexchange

Goodenough, *Magnetism and the Chemical Bond*, Interscience, New York (1963)

Spin and Orbital order in $LaMnO_3$



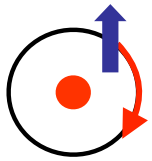
Goodenough (1963)



Orbital order in plane

Relativistic spin-orbit coupling

Magnetic anisotropy



$$\vec{B} = \frac{\vec{v} \times \vec{E}}{c^2}, \quad \vec{E} = -\nabla V$$

$$Zeeman : \vec{B} \cdot \vec{S} \sim \vec{L} \cdot \vec{S}$$

spin-orbit coupling

1. When $c \to \infty$ anisotropy $\to 0$

2. Total angular momentum $\vec{J} = \vec{L} + \vec{S}$

3. ∇V large when Z large $\to$ heavy elements $\to$ 4d, 5d

Ru, Mo

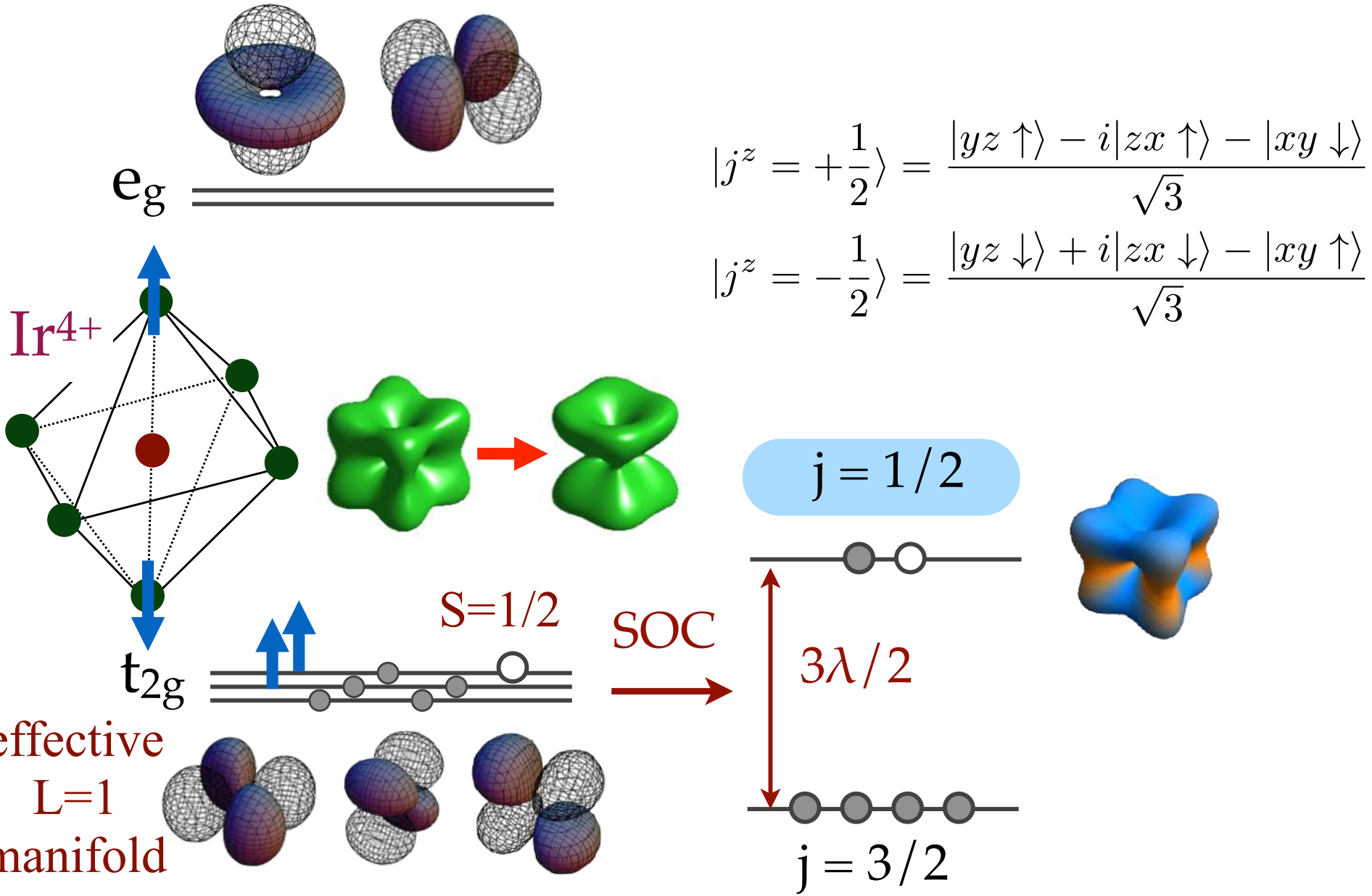
Ir, Os

4. $\vec{J}$ has direction & breaks rotational invariance of H

$S_i^z S_j^z$ instead of $\vec{S}_i \cdot \vec{S}_j$

(for $S = 1/2$ we have $(S_i^z)^2 = 1/4$)

Kitaev Materials: Magnetic Iridium Oxides



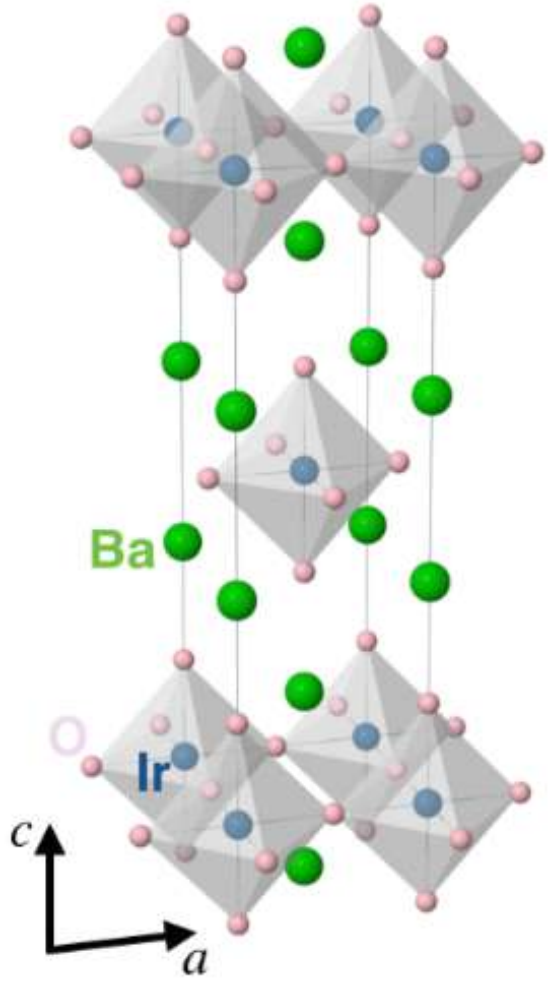
$$|j^z = +\frac{1}{2}\rangle = \frac{|yz\uparrow\rangle - i|zx\uparrow\rangle - |xy\downarrow\rangle}{\sqrt{3}}$$

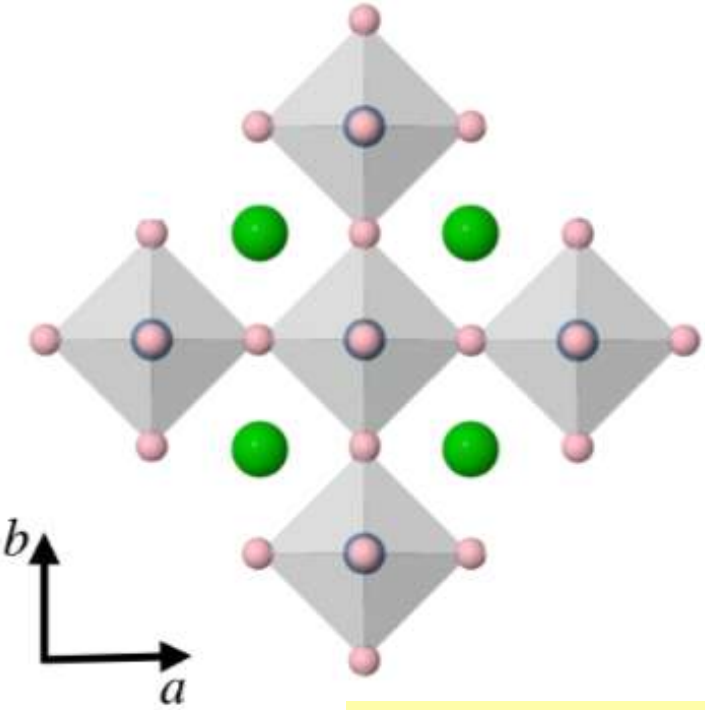
$$|j^z = -\frac{1}{2}\rangle = \frac{|yz\downarrow\rangle + i|zx\downarrow\rangle - |xy\uparrow\rangle}{\sqrt{3}}$$

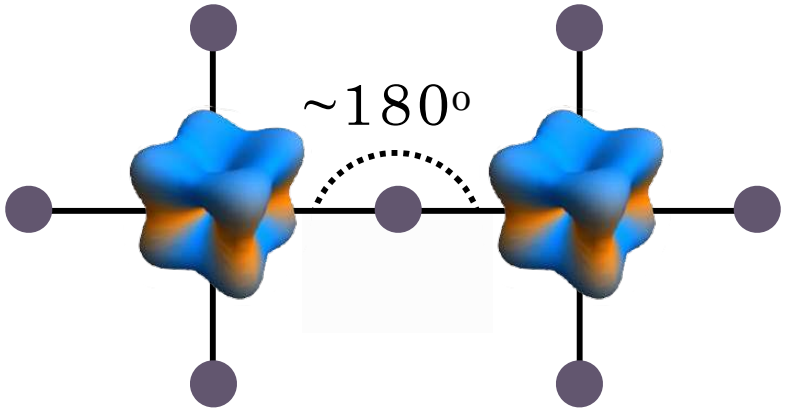
214 Magnetic Iridium Oxides: corner sharing

Sr_2IrO_4: equivalent of cuprate La_2CuO_4

j=1/2 moments
instead of S=1/2



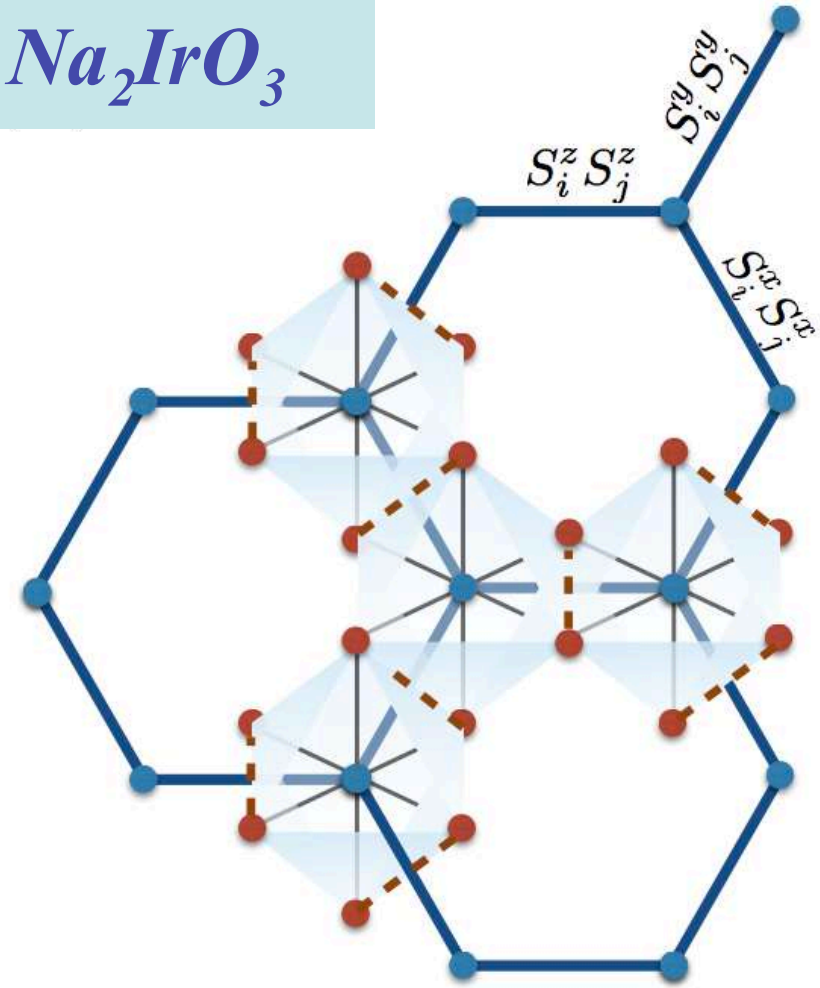




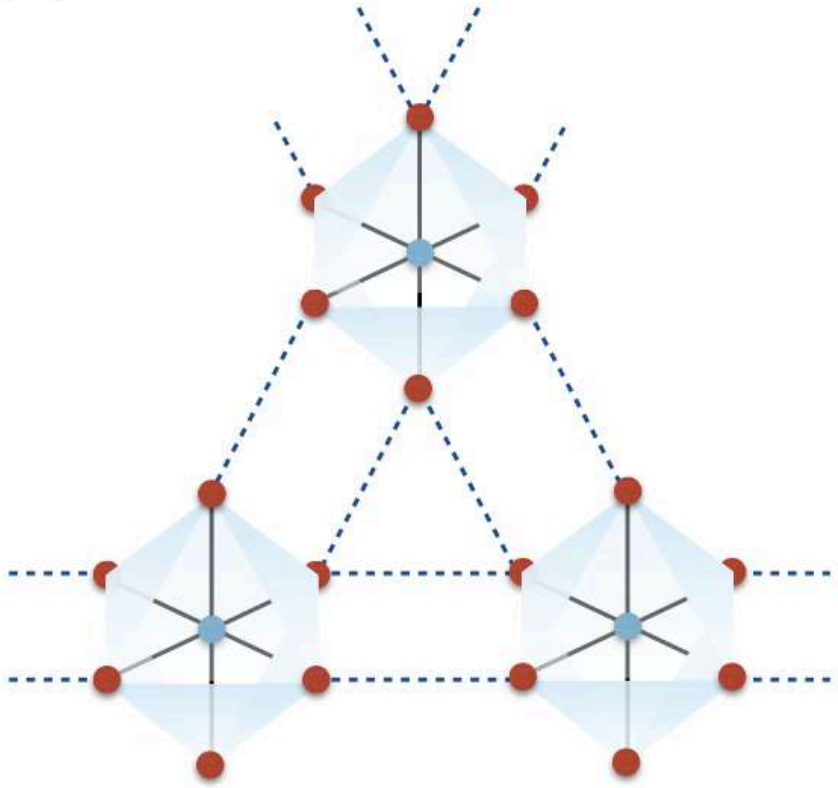
Jackeli & Khaliullin, PRL 102, 017205 (2009)

Edge sharing Iridium Oxides

Na_2IrO_3

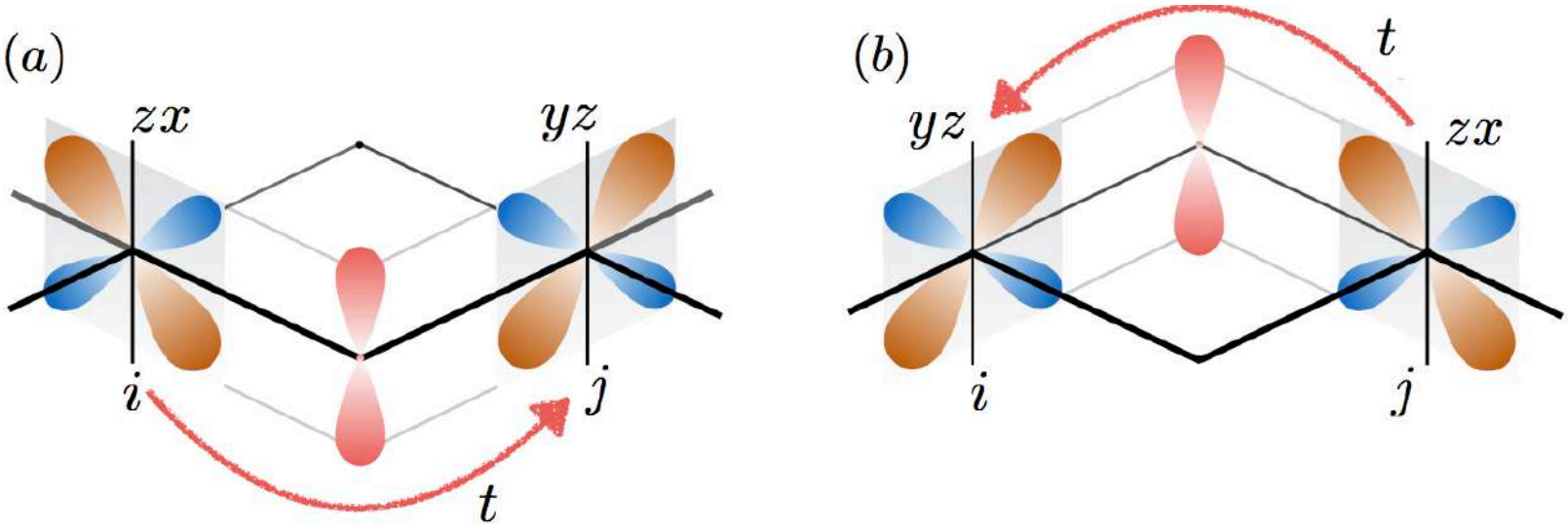


$Ba_3IrTi_2O_9$



Edge sharing Iridium Oxides

orbital dependent hopping



$$|j^z = +\frac{1}{2}\rangle = \frac{|yz\uparrow\rangle - i|zx\uparrow\rangle - |xy\downarrow\rangle}{\sqrt{3}}$$

$$|j^z = -\frac{1}{2}\rangle = \frac{|yz\downarrow\rangle + i|zx\downarrow\rangle - |xy\uparrow\rangle}{\sqrt{3}}$$

Exchange Hamiltonian flux phases

exchange interaction $H^{M,0}_{\langle ij \rangle} = J_0 \sin^2 \phi/2 \left(\mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{4} \right)$

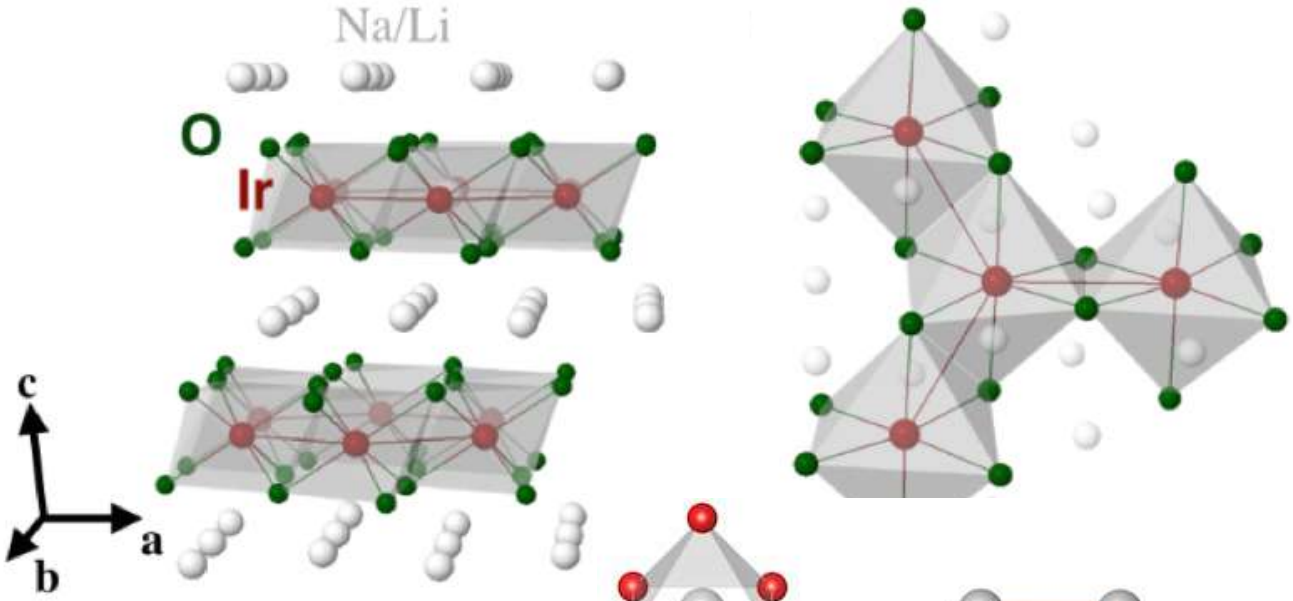
exchange interaction | order $J_H/U\ t^2/U = \eta\ t^2/U$

$$H^{M}_{\langle ij \rangle_\gamma} = \left(1 + \frac{\eta}{2}\right) H^{M,0}_{\langle ij \rangle} + \eta H^{K}_{\langle ij \rangle_\gamma}$$

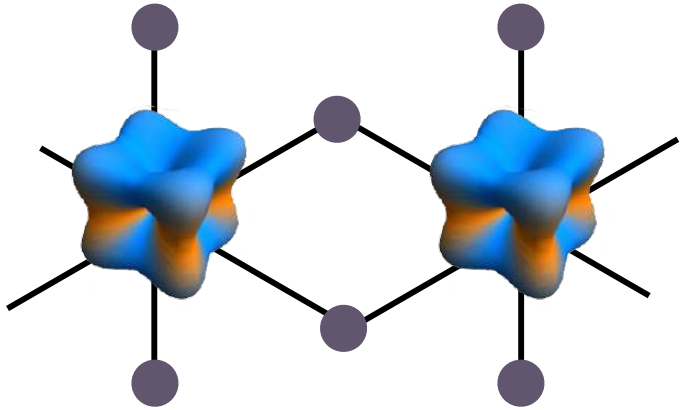
plus symmetry allowed residual interactions (further exchange anisotropies, and/or longer range interactions)

213 Magnetic Iridium Oxides

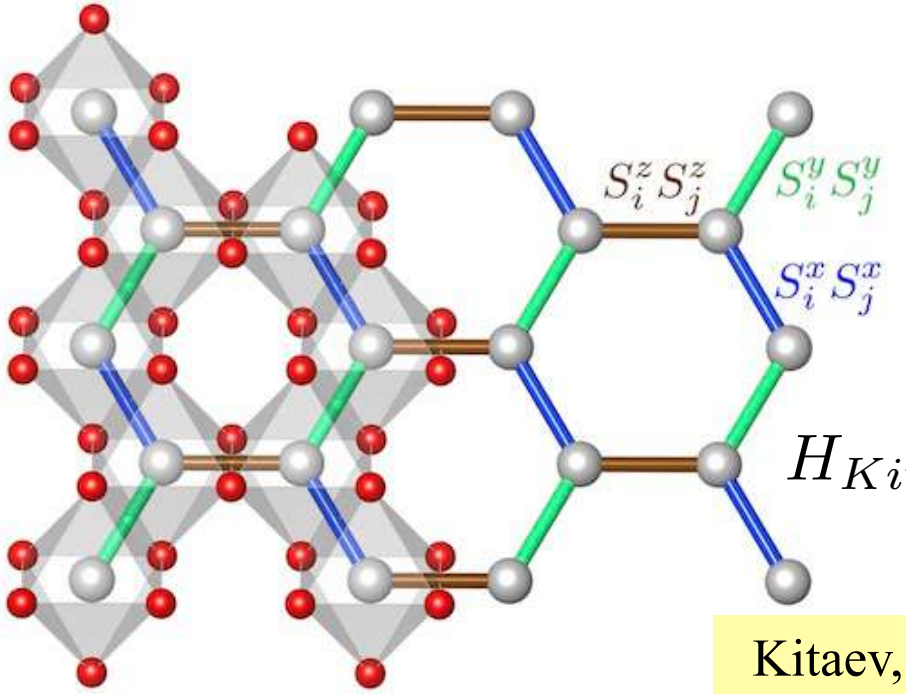
Na_2IrO_3 : honeycomb structure



$\angle\ Ir - O - Ir \sim 90^{o}$



$$H_{ij}^{(1)} = K S_i^z S_j^z$$

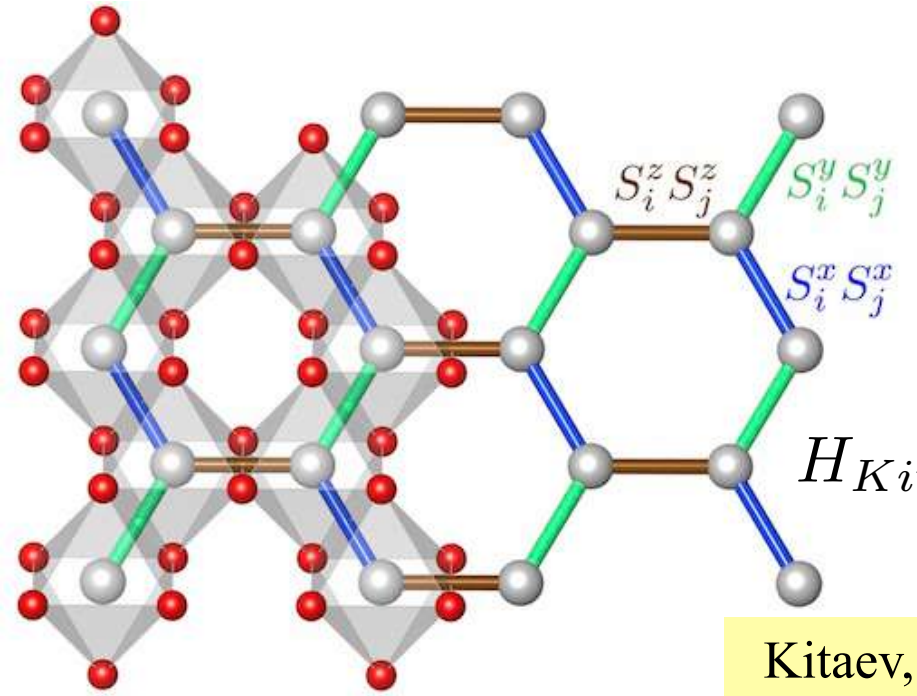


$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

Jackeli & Khaliullin, PRL 102, 017205 (2009)

Kitaev, Ann. Phys. 321, 2 (2006)

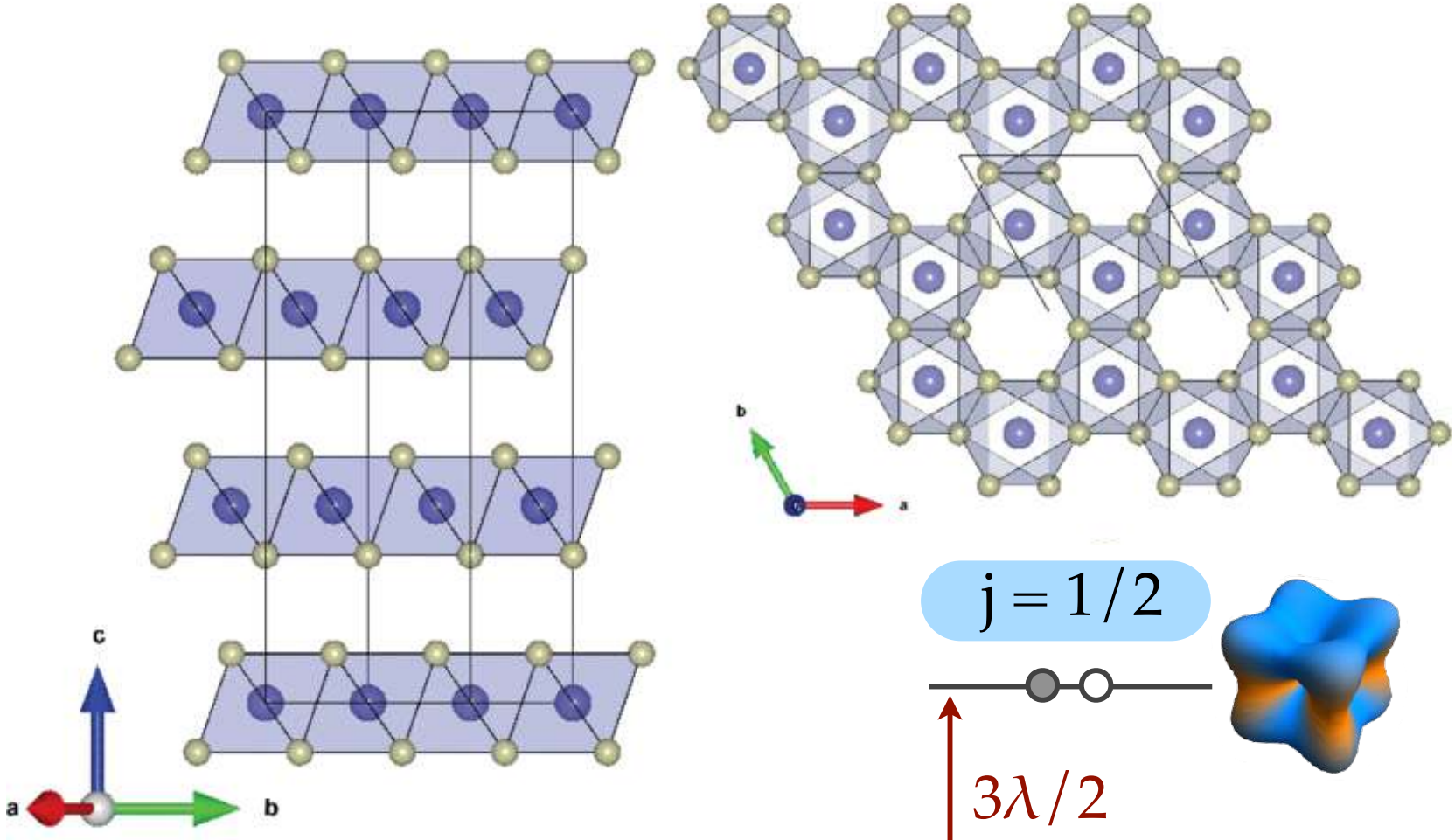
Honeycomb Kitaev model



$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

Kitaev, Ann. Phys. 321, 2 (2006)

Ru^{3+} $4d^5$ in honeycomb α-$RuCl_3$



$$H = KH\sum_{\langle ij\rangle_\gamma} S_i^\gamma S_j^\gamma + J\sum_{\langle ij\rangle_\gamma}\sum_{\langle ij\rangle} \vec{S}_i \vec{S}_j + \ldots.$$

Kitaev

Plumb, Clancy, Sandilands, Shankar, Hu, Burch, H-Y Kee & Y-J Kim, PRB 90, 041112 (2014)

Honeycomb Kitaev model I

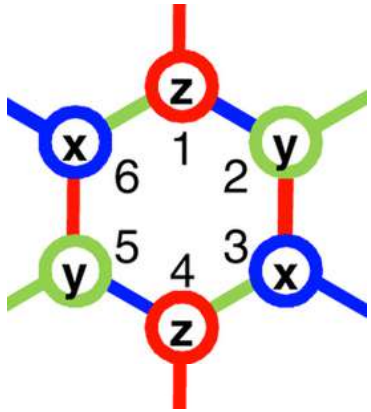
$$H = \sum_{\langle ij \rangle_\gamma} S_i^\gamma S_j^\gamma$$

A 1. Introduce flux on each hexagon $\hat{O}_i = S_1^z S_2^y S_3^x S_4^z S_5^y S_6^x$

2. $[H_K, \hat{O}_i] = 0 \ \forall i$

3. $[\hat{O}_i, \hat{O}_j] = 0 \ \forall i, j$

4. $\hat{O}_i^{\,2} = 1 \rightarrow O_i = \pm 1$



Flux on each hexagon: quantum number

System decomposes into 2^{Nh} sectors (Nh=N/2)

B Algebra of bond operators $b_{\mathbf{r}\gamma}$:

bonds *without* common sites ***commute***

bonds *with* common sites ***anti-commute***

Honeycomb Kitaev model II

$$H = \sum_{\langle ij \rangle_\gamma} S_i^\gamma S_j^\gamma = \sum_{\mathbf{r}\gamma} b_{\mathbf{r}\gamma}$$ bond operators $b_{\mathbf{r}\gamma}$

B Algebra of bond operators $b_{\mathbf{r}\gamma}$:

bonds *without* common sites ***commute***

bonds *with* common sites ***anti-commute***

→ related to algebra of majorana fermions:

$$b_{\mathbf{r}\gamma} = 2i\eta_{\mathbf{r}\gamma}\ c_{\mathbf{r}} c_{\mathbf{r}+\mathbf{e}_\gamma}$$

C 1. anticommutator $\{c_i, c_j\} = 0\ \forall\ i \neq j$

2. constant $\eta_{\mathbf{r}\gamma} = \pm 1$ depending on fluxes

3. $c_i^\dagger = c_i$ and $c_i^2 = 1/2$

4. groundstate is “flux free”: $O_i = 1\ \forall i$

5. “real fermion” $f^\dagger = (c_1 + ic_2)/2$

Honeycomb Kitaev model *III*

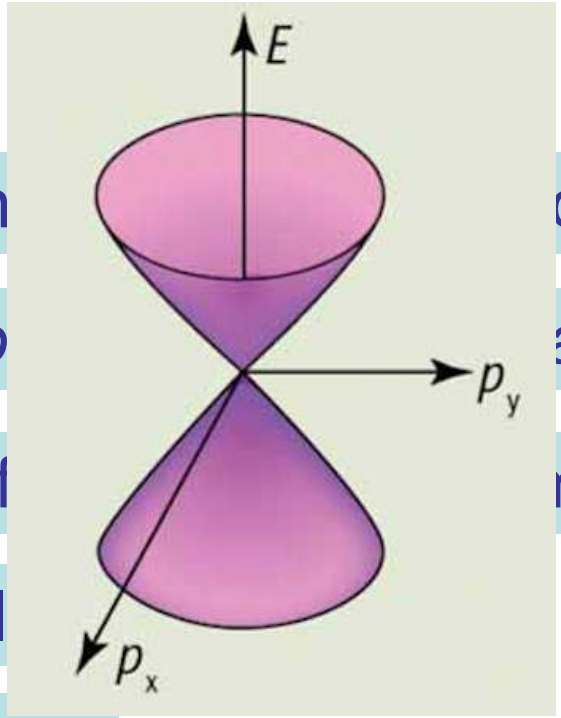
$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma = \sum_{\mathbf{r}\gamma} K_\gamma b_{\mathbf{r}\gamma}$$

bond operators $b_{\mathbf{r}\gamma}$

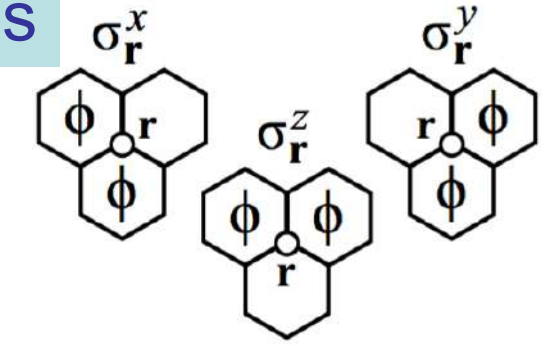
$$b_{\mathbf{r}\gamma} = 2i\eta_{\mathbf{r}\gamma}\ c_{\mathbf{r}} c_{\mathbf{r}+\mathbf{e}_\gamma}$$

- majoranas on honeycomb lattice with nearest neighbor hopping

"majorana graphene"



- static flux distribution of hopping $\eta_{\mathbf{r}\gamma} = \pm 1$
- *"spins breaks up into anas"* fractionalization
- spin excitation = flip f majoranas
- ground state is spin-l
- spins → spin statistics



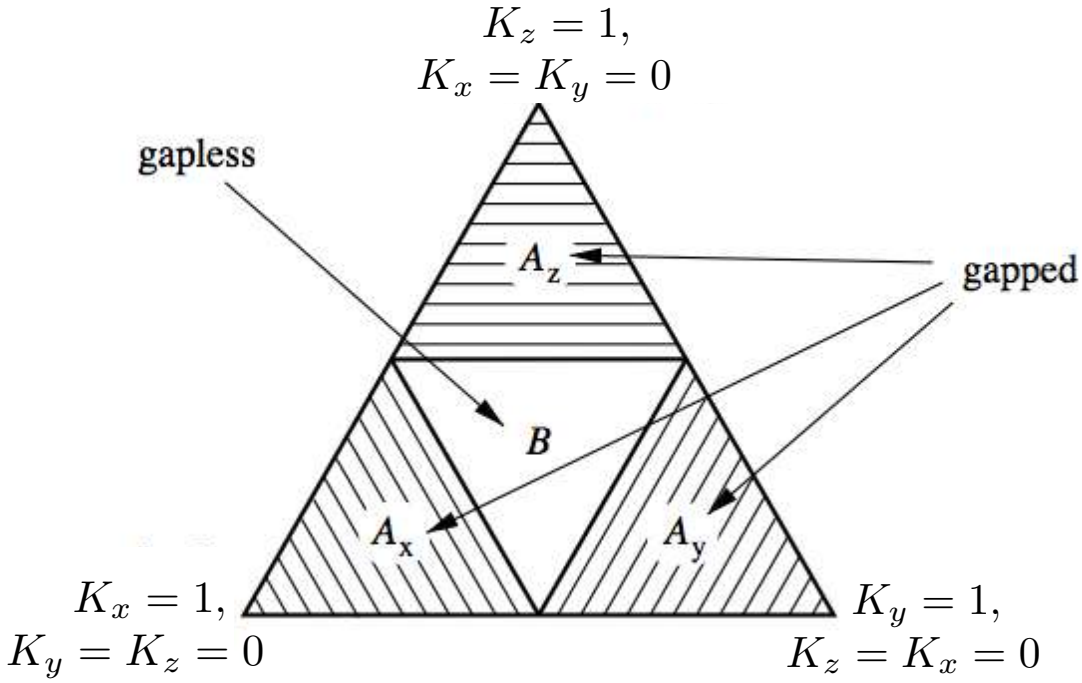
- majoranas → fermi statistics
- fluxes → anyon statistics

Honeycomb Kitaev model *IV*

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

- phase diagram

Abelian spin-liquid phases



- in magnetic field $H_{K-B} = K \sum_{\langle ij \rangle_\gamma} S_i^\gamma S_j^\gamma + B \sum_{i\gamma} S_i^\gamma$

gapped non-Abelian spin-liquid phase

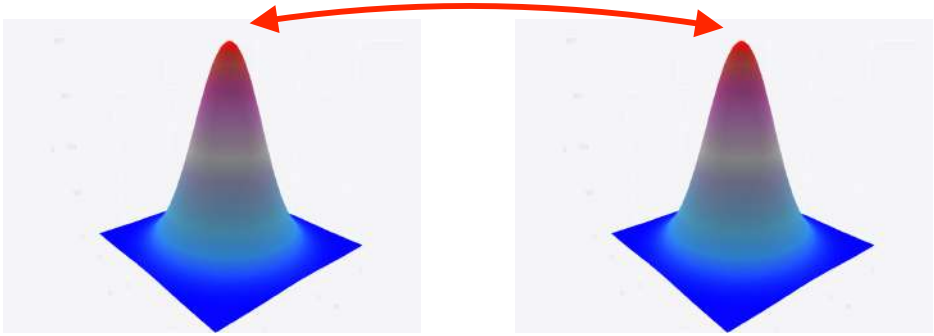
(perturbative in B/K)

Kitaev, Ann. Phys. 321, 2 (2006)

Quantum statistics of 2 particles in 3D

exchange operator of the two particles

P_{12}



wavefunction $\psi(\mathbf{r}_1, \mathbf{r}_2)$

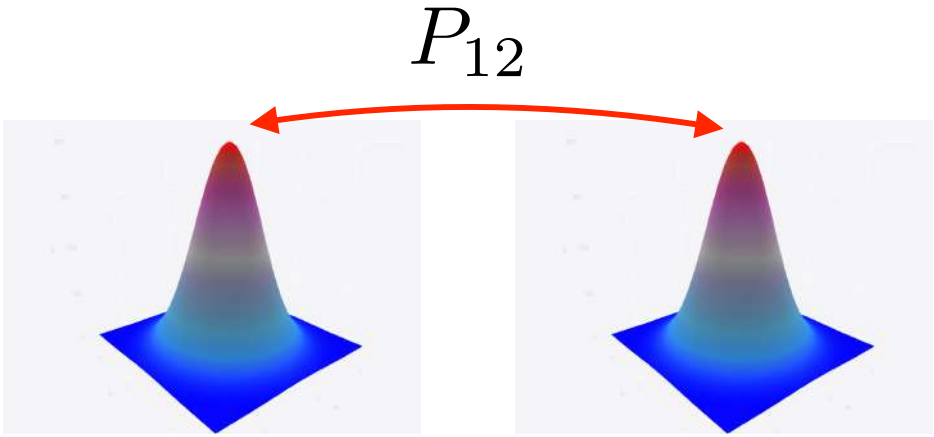
as $P_{12}^2 \psi(\mathbf{r}_1, \mathbf{r}_2) = \psi(\mathbf{r}_1, \mathbf{r}_2)$

it follows that $P_{12}\psi(\mathbf{r}_1, \mathbf{r}_2) = \pm\psi(\mathbf{r}_2, \mathbf{r}_1)$

$$= e^{i\gamma}\psi(\mathbf{r}_2, \mathbf{r}_1) \qquad \gamma = 0, \pi$$

Quantum statistics of 2 particles in 3D

exchange operator of the two a particles



bosons $P_{12} = +1$ integer intrinsic angular momentum

fermions $P_{12} = -1$ half integer intrinsic angular momentum

spin statistics theorem

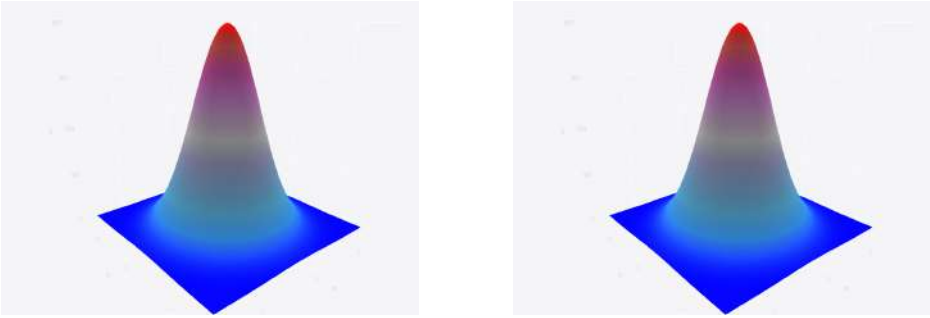
Markus Fierz

Wolfgang Pauli

Quantum statistics of 2 particles in 3D

rotate one particle around the other one

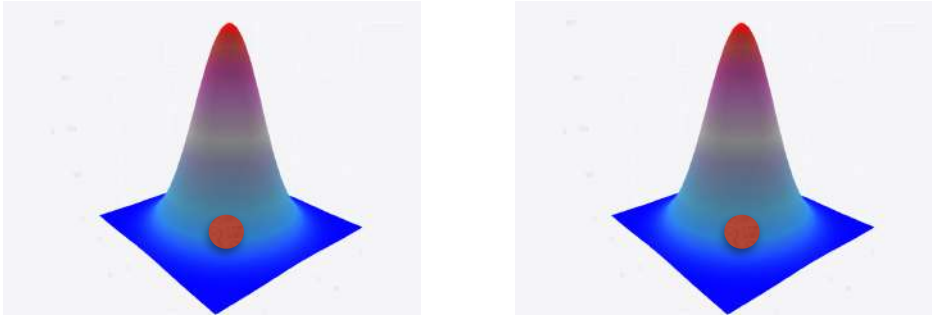
= exchange them twice



after a rotation loop $R\psi(\mathbf{r}) = e^{i\gamma}\psi(\mathbf{r})$ can $\gamma \neq 0, \pi$?

Not in 3D because all loops are topologically equivalent

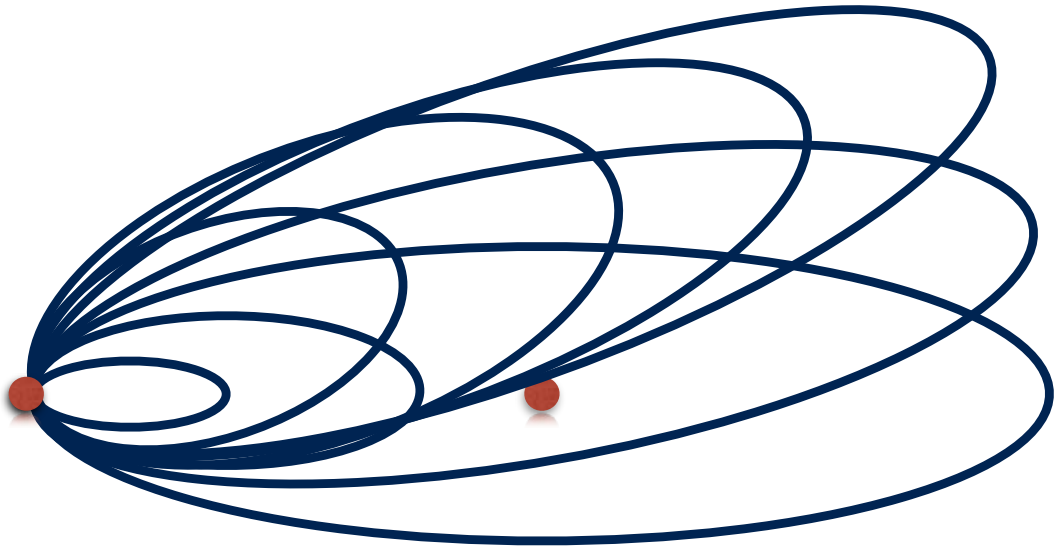
Quantum statistics of 2 particles in 3D



after a rotation loop $R\psi(\mathbf{r}) = e^{i\gamma}\psi(\mathbf{r})$ can $\gamma \neq 0, \pi$?

Not in 3D because all loops are topologically equivalent

Quantum statistics of 2 particles in 3D



after a rotation loop $R\psi(\mathbf{r}) = e^{i\gamma}\psi(\mathbf{r})$ can $\gamma \neq 0, \pi$?

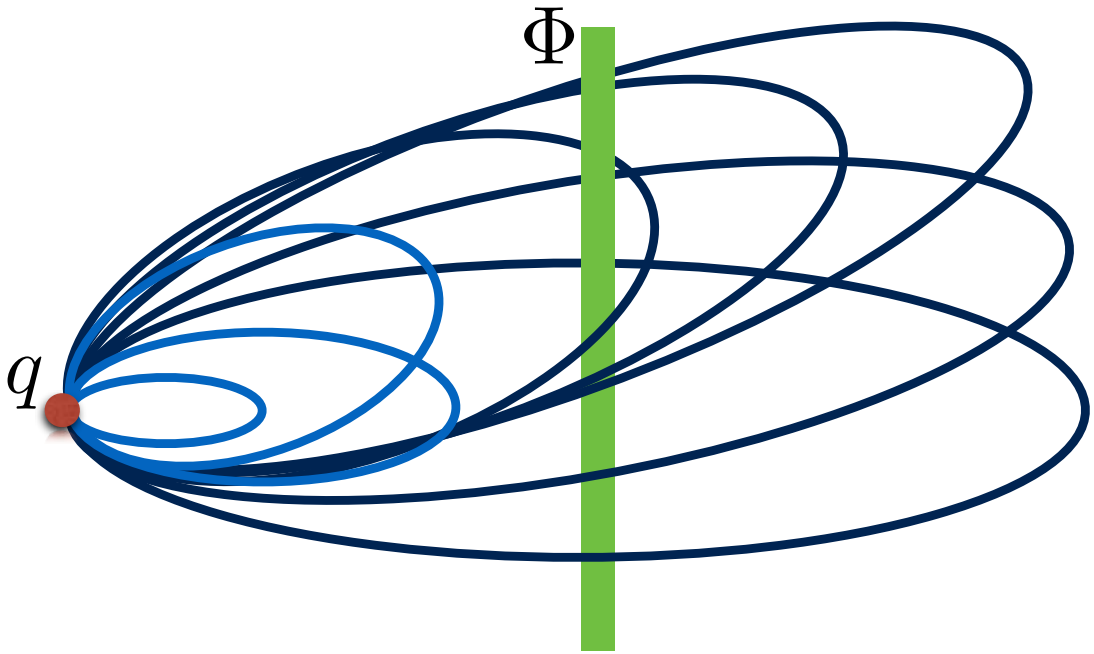
Not in 3D because all loops are topologically equivalent

and can be contracted to a rotation around its own axis

For a similar topological reason one cannot tie shoelaces in 4D

But now a particle in quasi-2D

rotate charged particle around a magnetic flux



now there are topologically distinct loops

Aharanov-Bohm phase $\gamma = \frac{q\Phi}{\hbar}$ for enclosed flux Φ

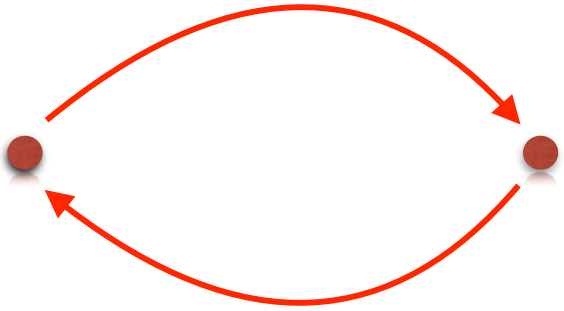
For encircled elementary flux quantum $\Phi_0 = \frac{h}{2e} \rightarrow \gamma = \pi$

"Exchanging" q and Φ produces phase difference

Exchange two particles 2D

"exchange" corresponds to 1/2 full rotation

$$P_{12}\psi(\mathbf{r}_1,\mathbf{r}_2) = e^{i\gamma/2}\psi(\mathbf{r}_2,\mathbf{r}_1)$$



Leinaas & Myrheim
Nuovo Cimento B. 37, 1 (1977)

statistical angle γ can take any value

$\rightarrow$ anyon

rather than exchanged (permuted), anyons are braided

"exchange" also -1/2 full rotation

$$P_{12}\psi(\mathbf{r}_1,\mathbf{r}_2) = e^{-i\gamma/2}\psi(\mathbf{r}_2,\mathbf{r}_1)$$

extremely robust

topologically protected

Generalise to non-Abelian (noncommutative) anyons

Suppose the anyon has an internal degree of freedom

label it by α so that $\psi_\alpha(\mathbf{r}_1, \mathbf{r}_2)$

wavefunction in degenerate subspace

More than one state: store (quantum) information qubit

Braiding produces $\psi_\alpha(\mathbf{r}_1, \mathbf{r}_2) \to e^{-i\gamma T_{\alpha\beta}} \psi_\beta(\mathbf{r}_2, \mathbf{r}_1)$

where $T_{\alpha\beta}$ is a matrix

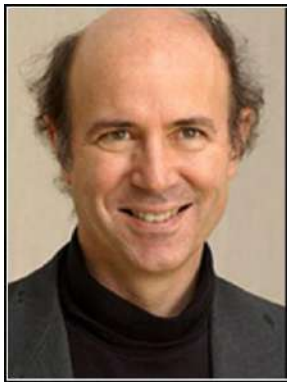
Braiding anyons rotates the qubit

By braiding anyons one can perform topologically protected non-commuting operations on qubits

How to construct anyons?

introduce charged particles with attached magnetic flux

Φ can take any value $\rightarrow$ anyon



Unfortunately does not work for Maxwell's electromagnetic fields

Jackiw & Redlich PRL 555 (1983)

Wilczek PRL 957 (1982)

Need emergent fluxes

= fluxes generated by the interactions between electrons

that act on the wavefunctions just like magnetic fluxes

Recipe: take interacting electrons, break them up in charged and fluxed particles, reassemble them

How to do that?

Fractional Quantum Hall

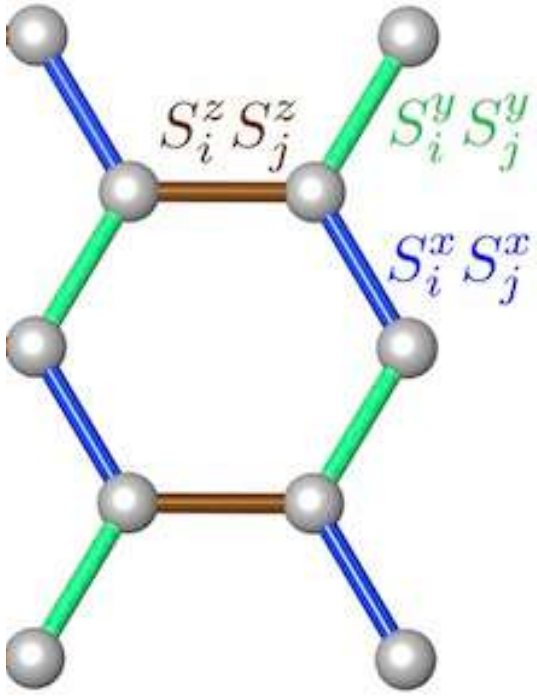
closing in but not there yet

Willett, Nayak, Shtengel, Pfeiffer & West, PRL **111**, 186401 (2013)

von Keyserlingk, Simon & Rosenow, PRL **115**, 126807 (2015)

Topological spin liquids
Kitaev Materials…

the race just started…



Kitaev model

$$H_{Kitaev} = \sum_{\langle ij \rangle_\gamma} K_\gamma S_i^\gamma S_j^\gamma$$

spins 1/2 on honeycomb lattice

spatially anisotropic interactions

Magnetic nearest neighbor interactions in α-$RuCl_3$

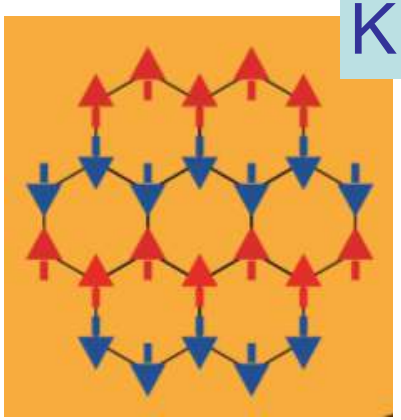
Quantum chemistry calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\tilde{S}_i^z\tilde{S}_j^z + \sum_{\alpha\neq\beta}\Gamma_{\alpha\beta}(\tilde{S}_i^\alpha\tilde{S}_j^\beta + \tilde{S}_i^\beta\tilde{S}_j^\alpha)$$

Structure	∠Ru-Cl-Ru	K	J	Γ_{xy}	$\Gamma_{zx}=-\Gamma_{yz}$
$C2/m$ [30]	94°	−5.6	1.2	−1.2	−0.7
$C2/m$ [29]					
Link 1 (×2)	94°	−5.3	1.2	−1.1	−0.7
Link 2 (×1)	93°	−4.8	−0.3	−1.5	−0.7
$P3_112$ [28]	89°	−1.2	−0.5	−1.0	−0.4

K large FM, J small AFM

Experimentally: zig-zag order below ~8K



However INS: K AFM

Banerjee et al., Nat. Mater. 4604 (2016)

Sears, Songvilay, Plumb, Clancy, Qiu, Zhao, Parshall & Y-J Kim, PRB 91, 144420 (2015)

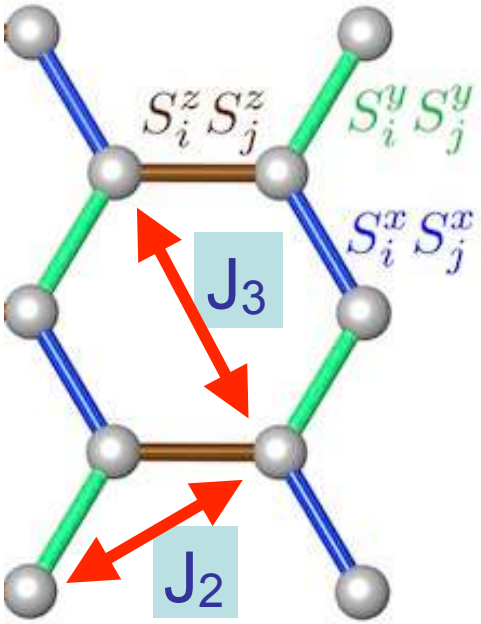
Yadav, Bogdanov, Katukuri, Nishimoto, JvdB & Hozoi, Sci. Rep. 6, 37508 (2016)

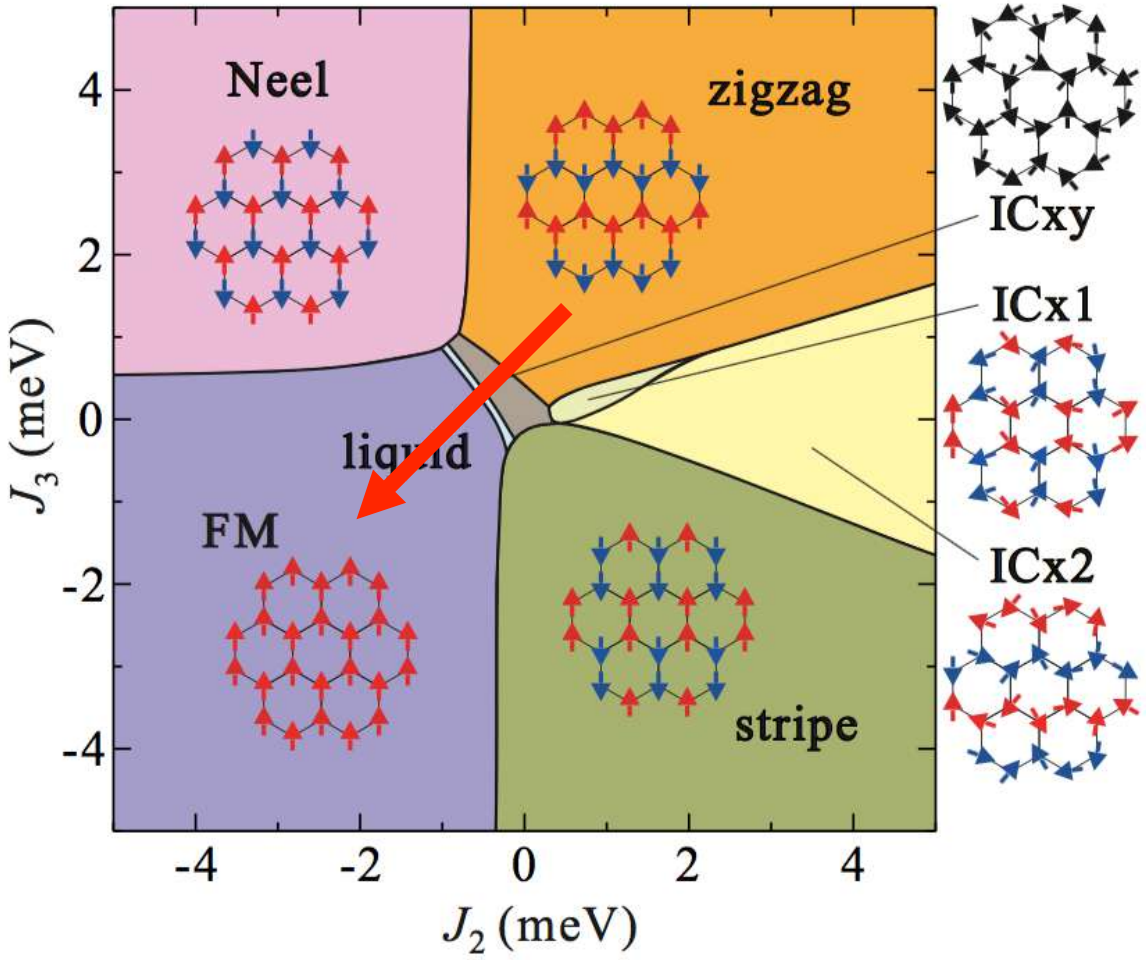
Magnetic nearest neighbor interactions in α-$RuCl_3$

Exact diagonalization calculations

$$\mathcal{H}_{i,j} = J\,\tilde{\mathbf{S}}_i \cdot \tilde{\mathbf{S}}_j + K\tilde{S}_i^z\tilde{S}_j^z + \sum_{\alpha\neq\beta}\Gamma_{\alpha\beta}(\tilde{S}_i^\alpha\tilde{S}_j^\beta + \tilde{S}_i^\beta\tilde{S}_j^\alpha)$$

+ longer range Heisenberg J_2 and J_3





zig-zag order driven by J_2 & J_3

Summarizing

fractionalizing quantum particles, transmuting even their statistics, is fun

in theory new quantum liquid states can appear

in practise:

ruthenium trichloride: $|K/J| \sim 5$, K ferro, J antiferro

other residual interactions O(J)

magnetic field of ~10T stabelizes spin liquid?