Reconciling spin 1 and orbital degeneracy in V_2O_3

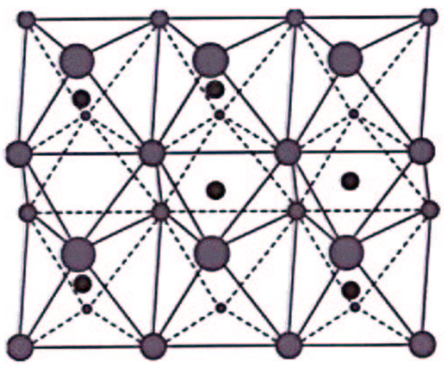
Frédéric MILA
Institut de Physique Théorique
Université de Lausanne (Switzerland)

Collaborators: R. Shiina, F.-C. Zhang, A. Joshi, M. Ma, V. A. Anisimov, T. M. Rice

Scope

- **Introduction to V_2O_3**
  - Phase diagram
  - New experiments
- **Vertical pair**
  - Level crossings
  - Orbital degeneracy
- **Pair model**
  - In-plane coupling
  - Role of spin-orbit coupling
  - Open issues
- **Conclusions**

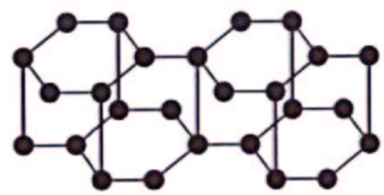
Structure of V_2O_3



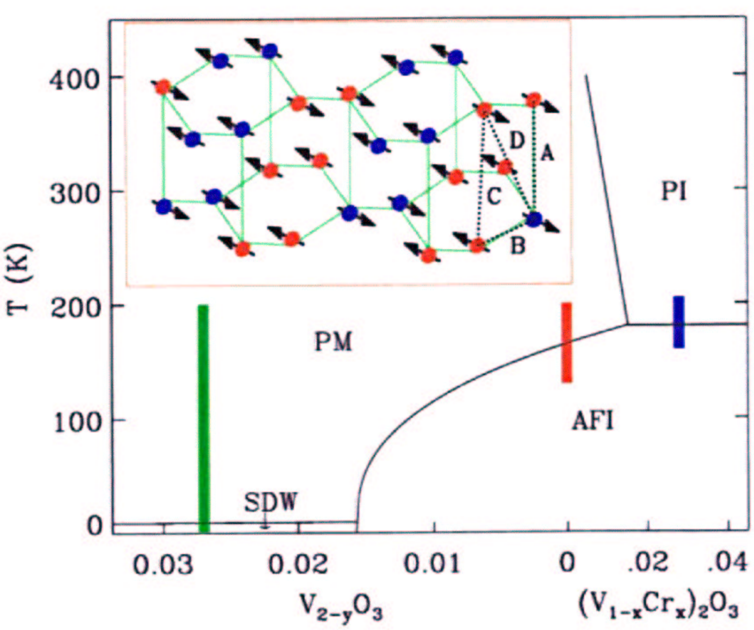
Corundum structure

$V^{3+} \Rightarrow 3d_2 \Rightarrow S = 1$

Vanadium lattice $\Longleftrightarrow$ coupled hexagonal lattices



Phase diagram of V_2O_3



New experiments

Bao *et al* (PRB '98) Inelastic neutron scattering

Fluctuations in PI phase **not related** to magnetic order in AFI
(1, 0, 1) $(\frac{1}{2}, \frac{1}{2}, 0)$

Park *et al* (PRB 2000) Polarized X-ray absorption

Both $e_g e_g$ **and** $a_{1g} e_g$ + Spin 1

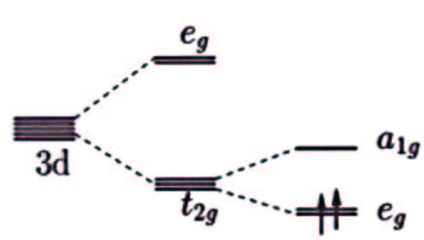
Paolasini *et al* (PRL '99) Resonant X-ray scattering

Additional Bragg peaks **not directly related** to magnetic order

Goulon *et al* (PRL 2000) Nonreciprocal X-ray gyrotropy

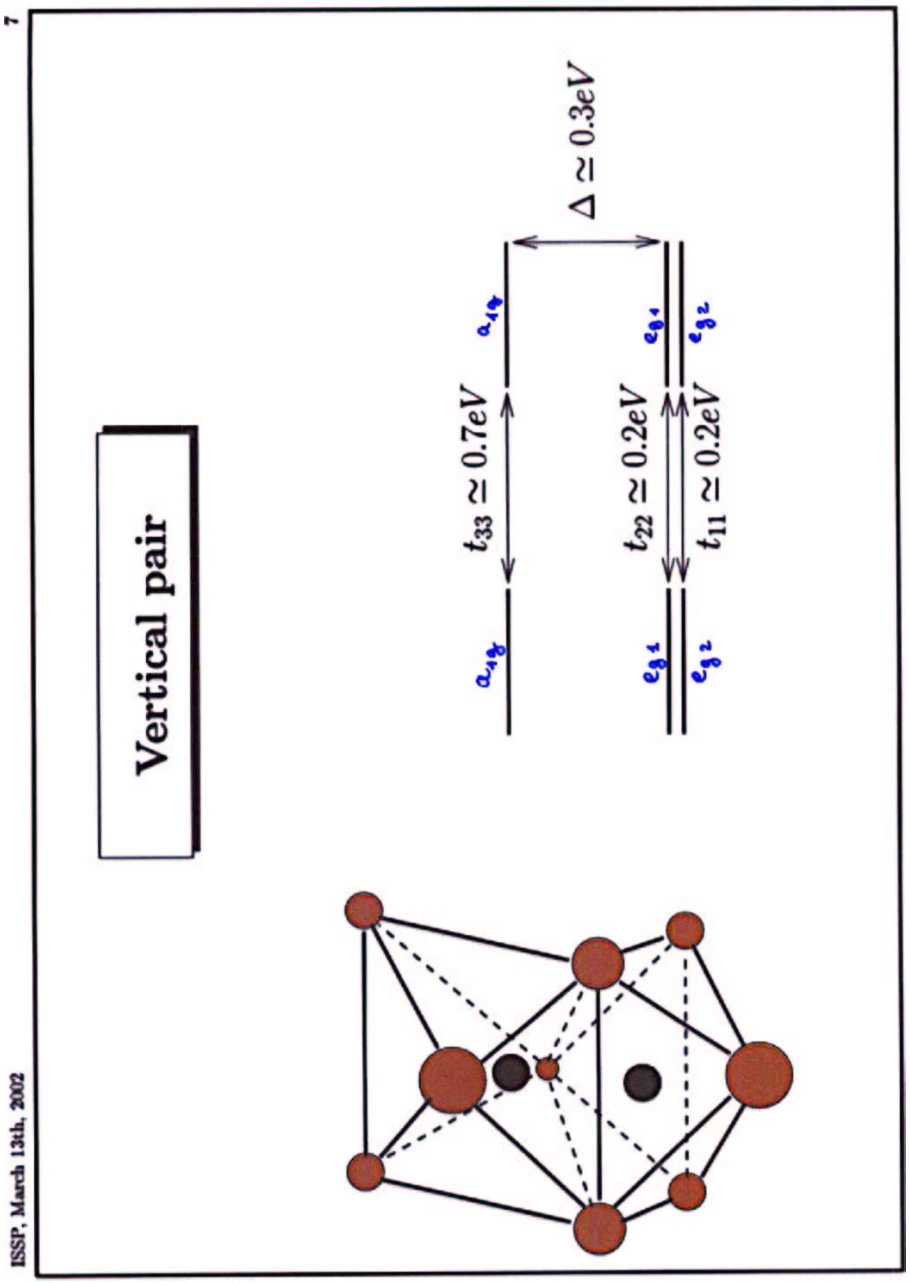
Magnetic group **non** centrosymmetric

Single site



Spin 1 but **NO ORBITAL DEGENERACY!**

→ CONSIDER VERTICAL PAIRS (Castellani et al, '79)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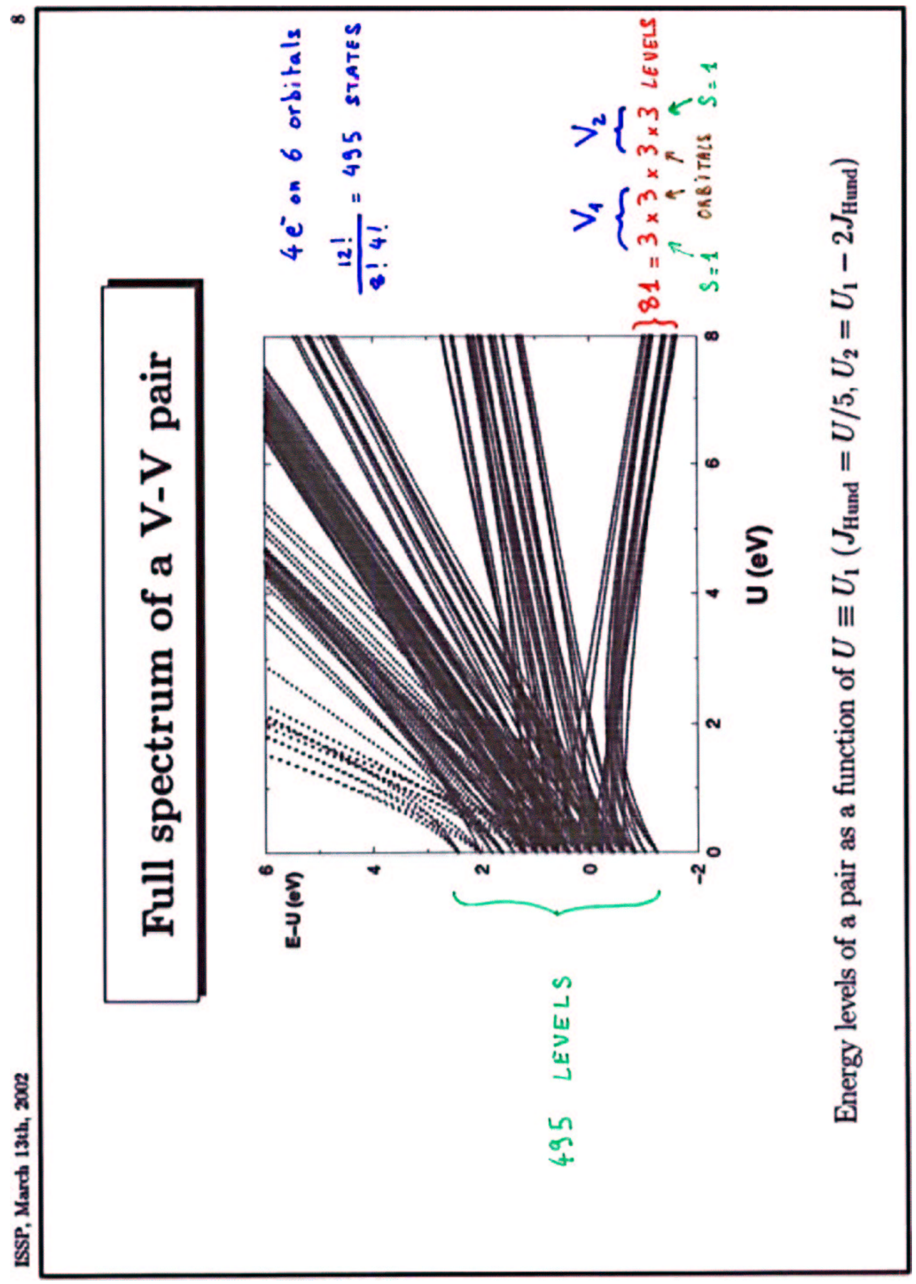
Vertical pair
t33 ≃ 0.7eV
t22 ≃ 0.2eV
t11 ≃ 0.2eV
Δ ≃ 0.3eV
a1g
eg1
eg2
ISSP, March 13th, 2002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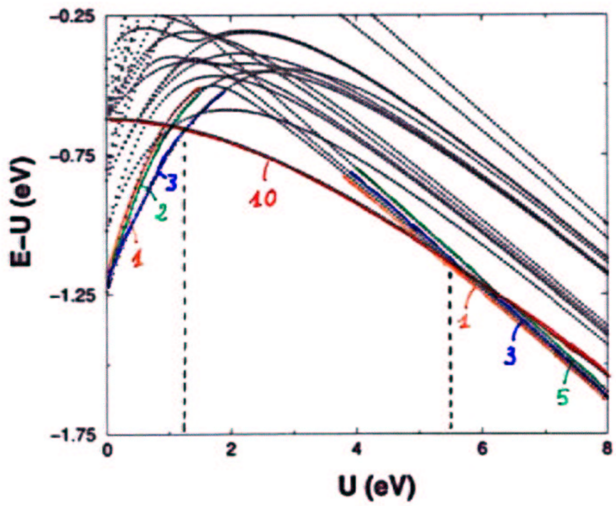
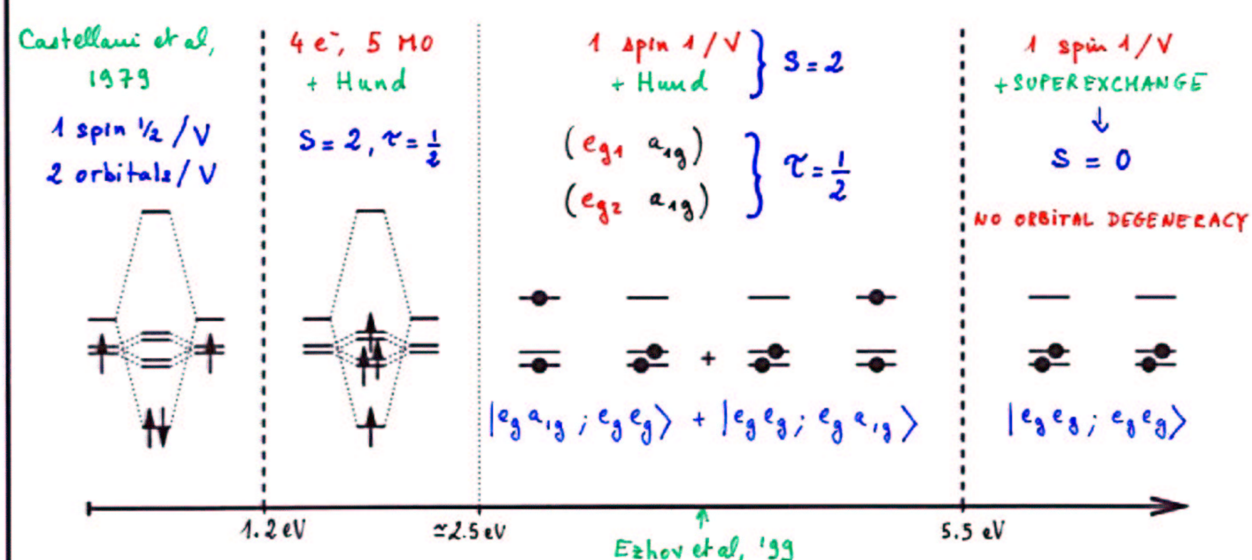
Full spectrum of a V-V pair
4 e⁻ on 6 orbitals
12! / 8! 4! = 495 STATES
81 = 3 x 3 x 3 x 3 LEVELS
V1 V2
S=1 ORBITALS S=1
495 LEVELS
E−U (eV)
U (eV)
Energy levels of a pair as a function of U ≡ U1 (J_Hund = U/5, U2 = U1 − 2J_Hund)
ISSP, March 13th, 2002

Level crossings in the low-energy pair spectrum



Lowest energy levels of a pair as a function of $U \equiv U_1$ ($J_{\text{Hund}} = U/5$, $U_2 = U_1 - 2J_{\text{Hund}}$)

Effect of e^--e^- interactions



Theory of V_2O_3: Basic Ingredients

Relevant states of a vertical pair

$|e_g a_{1g}; e_g e_g\rangle$

or/and

$|e_g e_g; e_g e_g\rangle$

Residual interactions

– Inter-pair couplings

– Spin-orbit coupling

– Coupling to the lattice

Pair model

Effect of interpair coupling on $|e_g a_{1g}; e_g e_g\rangle$ configurations

F. Mila et al, PRL 2000; R. Shiina et al, PRB 2001

- Spin 1 on each site + Ferromagnetic coupling $\Rightarrow S = 2$ $(\vec{\sigma})$
- Choice for empty e_g orbital $\Rightarrow$ 2-fold orbital degeneracy $(\vec{\tau})$

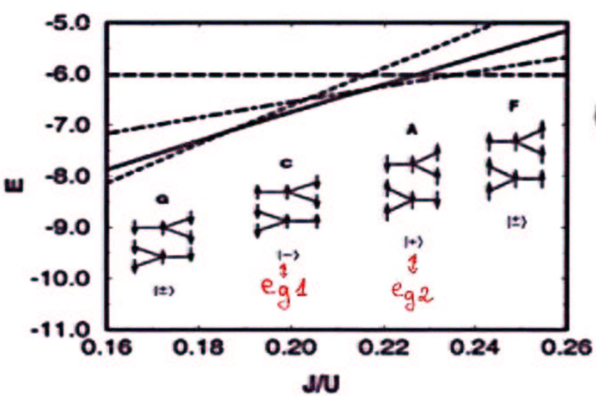
$$H_{\text{eff}}^{\text{bond}} = \left(G + \tfrac{1}{4}G_3(\tau_l^z + \tau_m^z)\right) \vec{\sigma}_l . \vec{\sigma}_m$$

+ appropriate rotation by $2\pi/3$ for other bonds.

$$G = -\tfrac{1}{3}G_1 + \tfrac{1}{3}G_2 + \tfrac{3}{4}G_3, \quad G_1 = \frac{t^2}{4(U'-J)}, \; G_2 = \frac{t^2}{4(U'+2J)}, \; G_3 = \frac{t^2}{4(U+J)}$$

Magnetic Structure

Mean-field decoupling of spin and orbital ⇒ Effective spin Hamiltonian

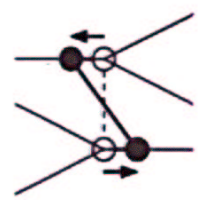


C-phase: real spin structure

+ ferroorbital ordering of vertical pairs

(e_{g1}) consistent with monoclinic distortion

(Dernier and Marezio, 1970).



Polarized X-ray Absorption

($e_g e_g$) and ($a_{1g} e_g$) both present (Park et al, 2000).

Simple pair model

⇓

50% ($e_g e_g$) - 50% ($a_{1g} e_g$)

- Qualitatively: correct

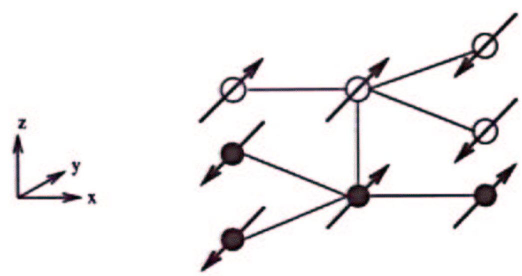
- Quantitatively: one should include some excited states of the pairs.

$|e_g e_g ; e_g e_g\rangle$

Resonant X-ray scattering (Paolasini et al, PRL 1999)

- Bragg peaks consistent with magnetic struture
- Additional Bragg peaks → orbital ordering?

Pair model



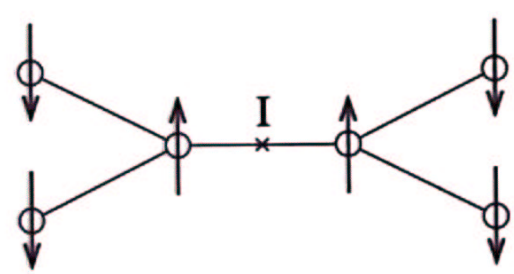
○ $(=e_{g1}) = (\sqrt{2}\, d_{xy} + d_{xz})/\sqrt{3}$

$\neq$

● $(=e_{g1}) = (\sqrt{2}\, d_{xy} - d_{xz})/\sqrt{3}$

Additional Bragg peaks consistent with magnetic scattering

Nonreciprocal X-Ray Gyrotropy (Goulon et al, PRL 2000)

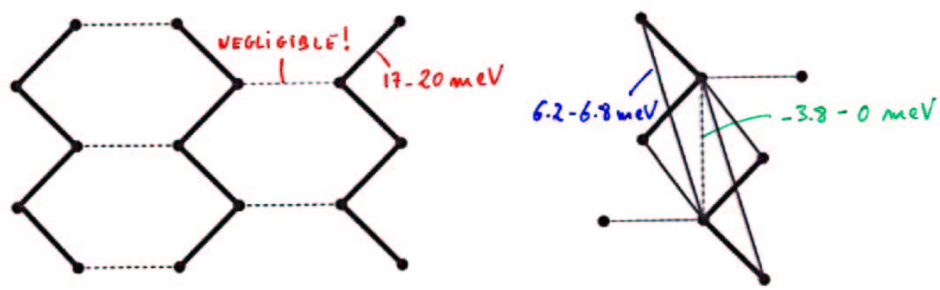


Nonreciprocal X-Ray gyrotropy ⇒ Inversion center is lost

First Proposal (Di Matteo et al, unpublished)

Different e_g orbitals on different vertical pairs

Alternative explanation: Spin canting



Ferromagnetic bonds **very weak** (Bao et al, unpublished)

Dzyaloshinskii-Moriya interaction ⇒ small canting

NB: EXCHANGE CONSTANTS CONSISTENT WITH PAIR MODEL IF SOME MIXING WITH $|e_g e_g; e_g e_g\rangle$ IS INCLUDED (Shiina et al, PRB '01).

Effect of spin-orbit coupling

- Magnetic resonant scattering
- Orbital moments → Additional Bragg peaks (Lovesey and Knight; Tanaka)
- Tilting of magnetic moments away from z-axis if $(e_g e_g; e_g e_g)$ not too far (Shiina et al, PRB 2001; see also Tanaka)

More radical point of view — A. Tanaka, cond-mat/0201407

- Degeneracy between $|e_{g1} a_{1g}; e_g e_g\rangle$ and $|e_{g2} a_{1g}; e_g e_g\rangle$ lifted

→ NO ORBITAL DEGENERACY!

– Why 1st order transition? (Pair model OK, see Joshi et al, PRL '00).
– Why J_{Ferro} and J_{AF} so different?
– Why treat $t_{in-plane}$ as second-order effect? ($t_{in-plane} \gg \Delta_{spin-orbit}$)

Conclusions

- **V_2O_3 still resists!**
- **Orbital degeneracy**
  - Plays an essential role.
  - No orbital ordering at atomic level.
  - Possible orbital ordering of vertical pairs → { GOOD CASE FOR FERROORBITAL ORDERING OF VERTICAL PAIRS
- **Resonant X-Ray scattering**
  - Excellent technique to probe orbitals in V_2O_3 → FINAL ANSWER ?