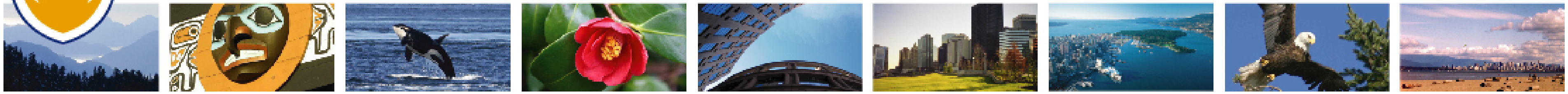
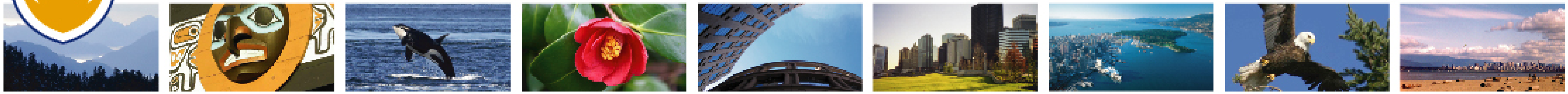
UBC UNIVERSITY OF BRITISH COLUMBIA

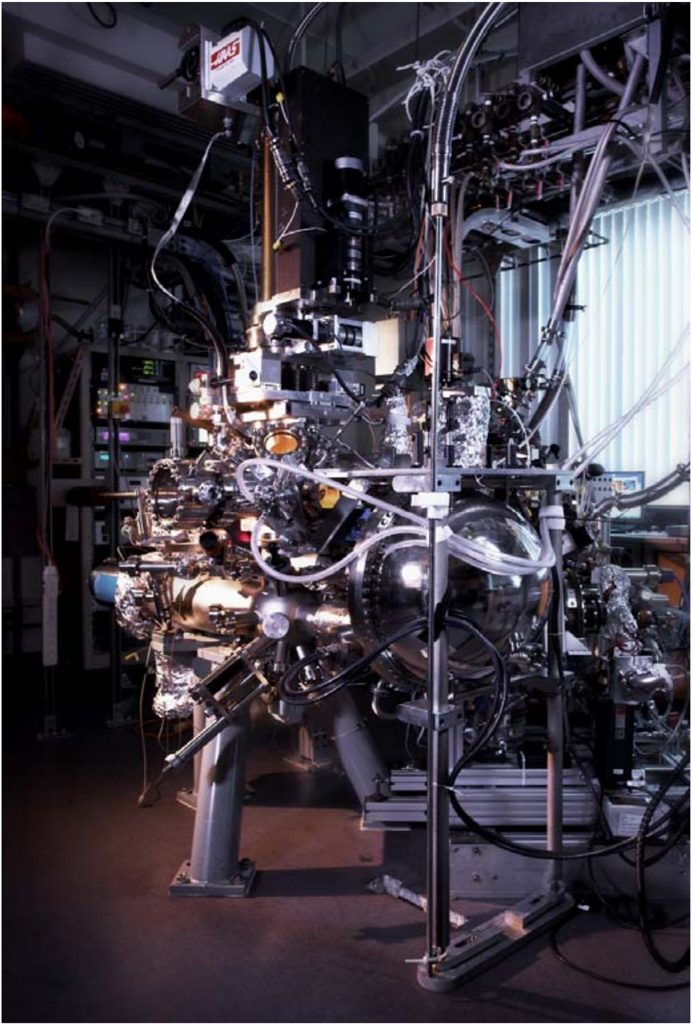


Andrea Damascelli

Spin-orbital entanglement and spin-triplet pairing in Sr_2RuO_4

Max Planck – UBC
Quantum Matter Institute





C.N. Veenstra, Z.-H. Zhu,
G. Levy, B. Ludbrook,
J.A. Rosen, R. Comin

M.H. Haverkort, I.S. Elfimov

A. Damascelli

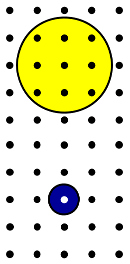
Sr_2RuO_4

S. Kittaka, Y. Maeno
Kyoto University

Strongly Correlated Electron Systems

Comin & Damascelli
arXiv:1303.1438

d - f
open shells materials

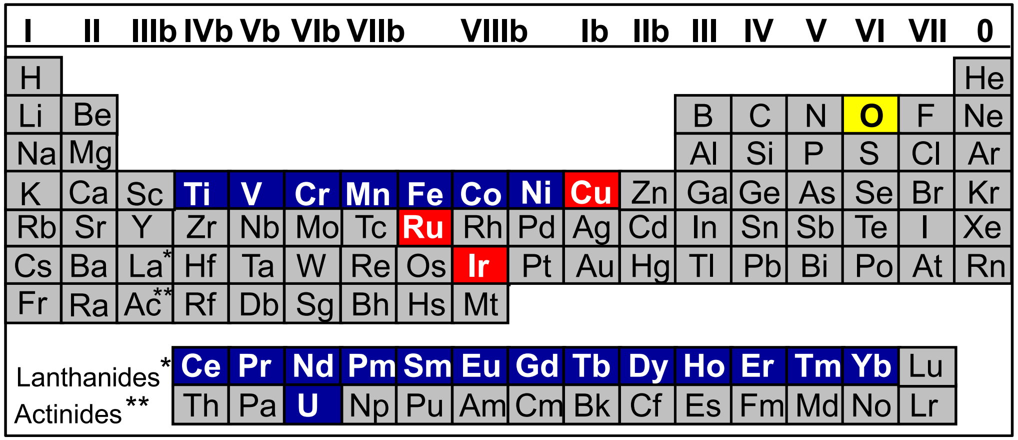


U<<W
Charge fluctuations

U>>W
Spin fluctuations

Control parameters

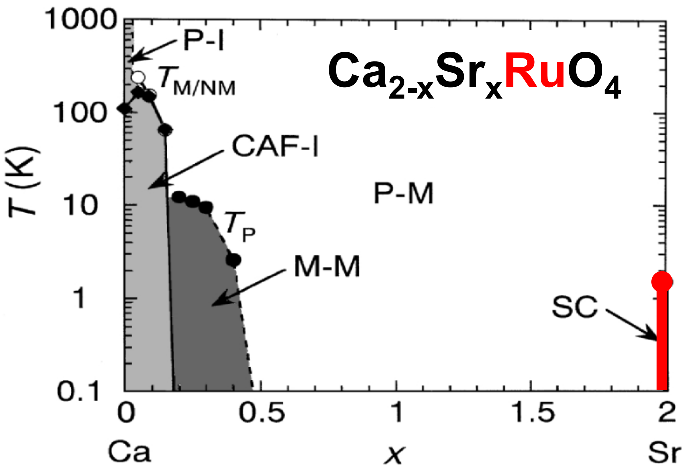
Bandwidth (U/W)
Band filling
Dimensionality



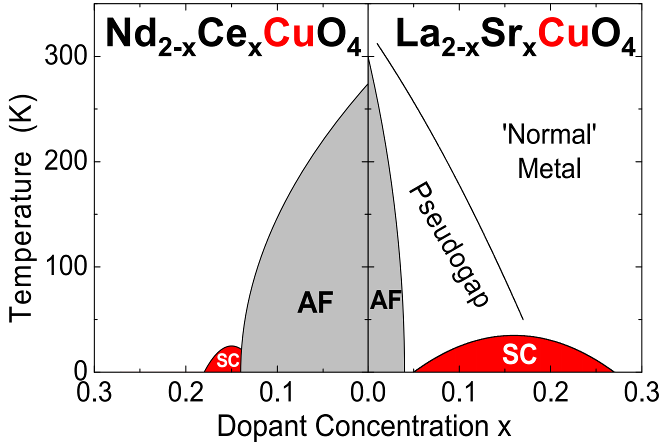
I	II	IIIb	IVb	Vb	VIb	VIIb	VIIIb			Ib	IIb	III	IV	V	VI	VII	0
H																	He
Li	Be											B	C	N	O	F	Ne
Na	Mg											Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**	Rf	Db	Sg	Bh	Hs	Mt									
Lanthanides*			Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	
Actinides**			Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr	

Degrees of freedom

Charge / Spin
Orbital
Lattice



- Kondo
- Mott-Hubbard
- Heavy Fermions
- Unconventional SC
- Spin-charge order
- Colossal MR



Strongly Correlated Electron Systems

Comin & Damascelli
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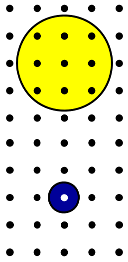
d - f
open
shells
materials



U<<W
Charge fluctuations

U>>W
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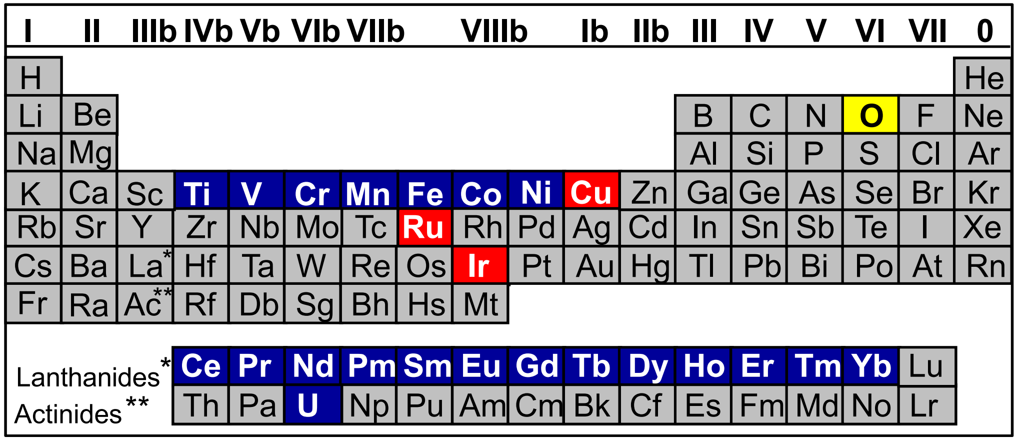

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K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
Cs	Ba	La*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
Fr	Ra	Ac**	Rf	Db	Sg	Bh	Hs	Mt									
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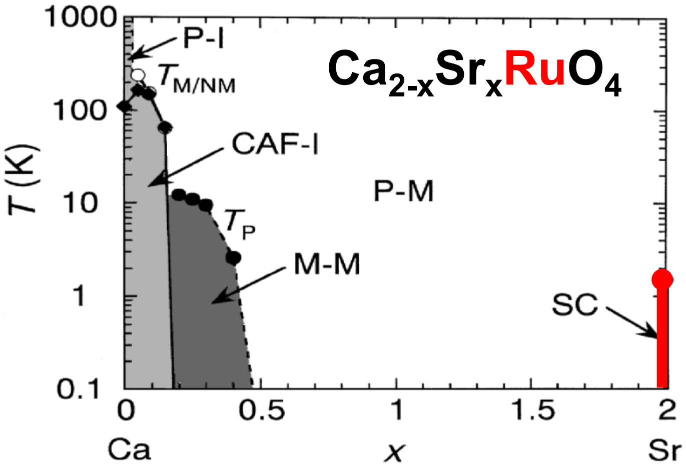
Degrees of freedom

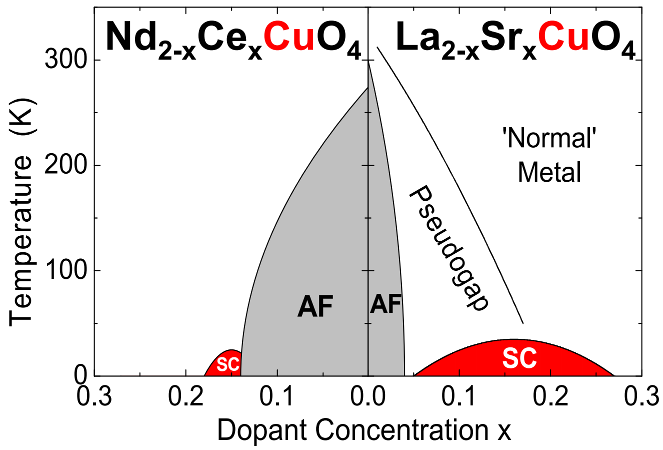
Charge / Spin
Orbital
Lattice



d-orbitals
radial extent

↔

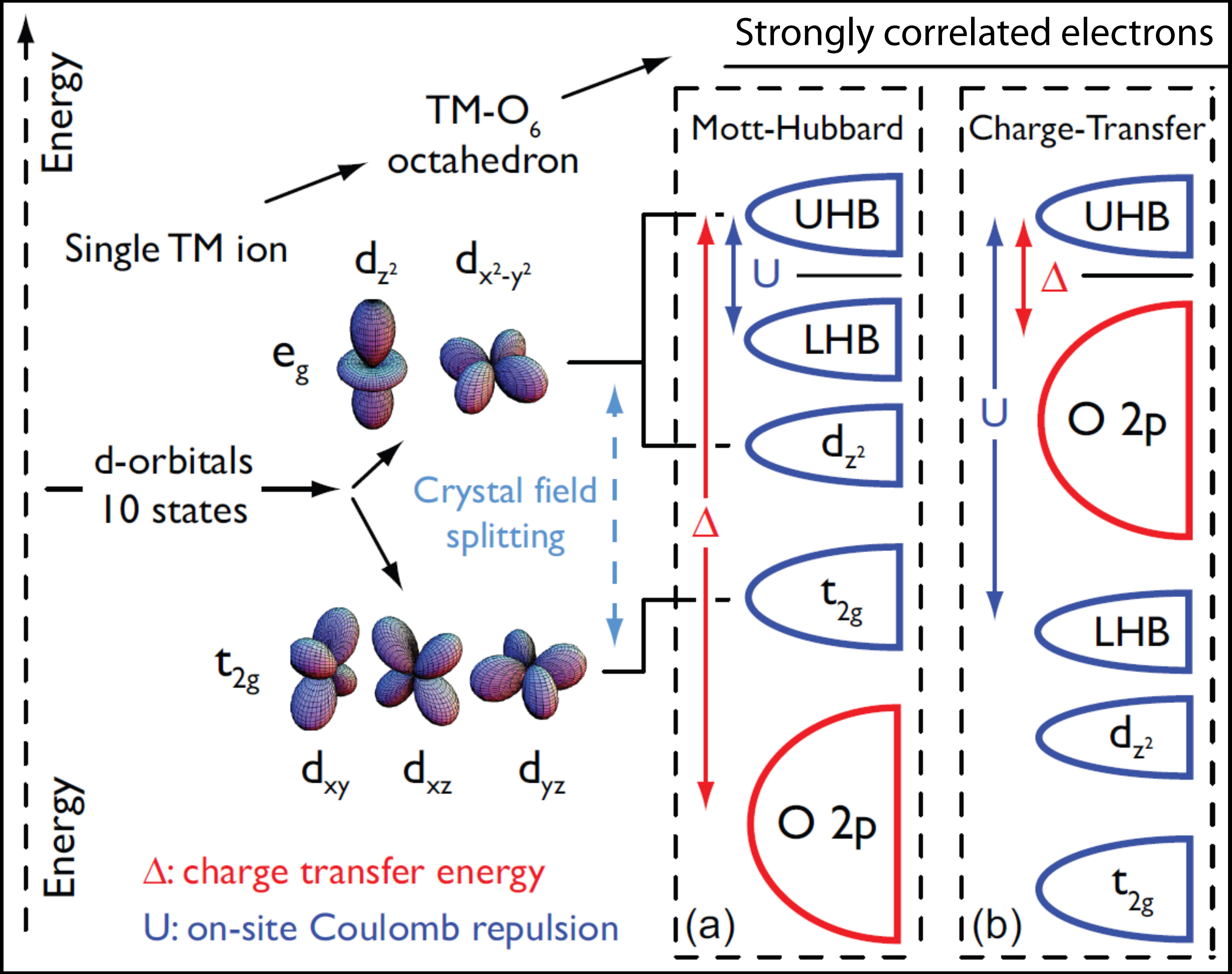
Spin-orbit
coupling



Mott criterion U>W

Early
3d TMO

Late
3d TMO

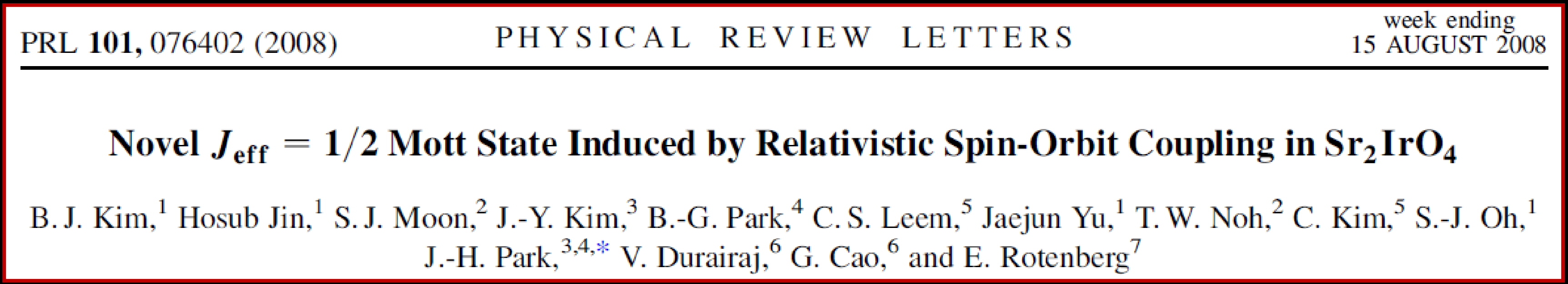


Δ: charge transfer energy

U: on-site Coulomb repulsion

Comin & Damascelli, arXiv:1303.1438

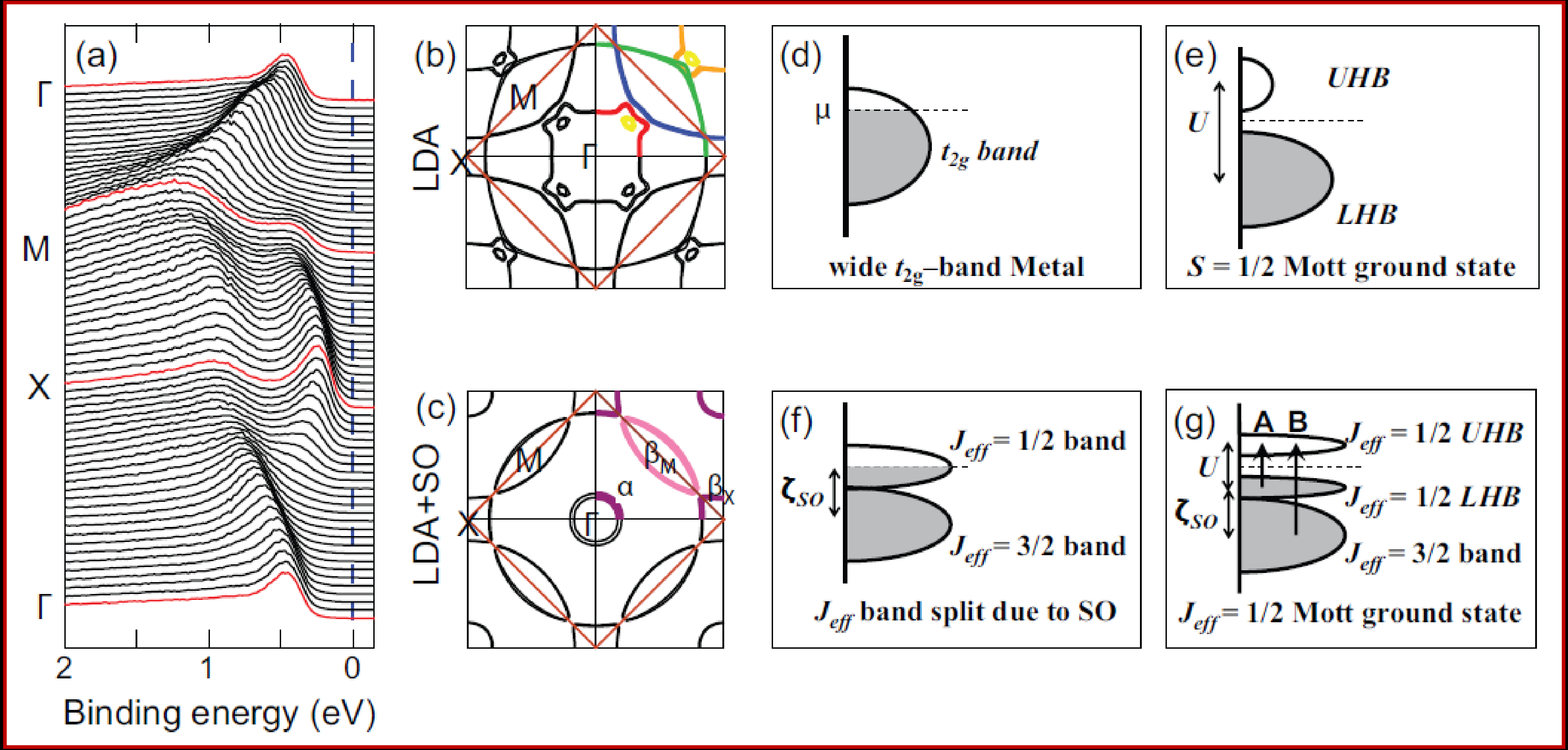
Interplay of Coulomb U and SO in 5d iridates



PRL **101,** 076402 (2008) PHYSICAL REVIEW LETTERS week ending 15 AUGUST 2008

Novel $J_{\text{eff}} = 1/2$ Mott State Induced by Relativistic Spin-Orbit Coupling in Sr_2IrO_4

B. J. Kim,[1] Hosub Jin,[1] S. J. Moon,[2] J.-Y. Kim,[3] B.-G. Park,[4] C. S. Leem,[5] Jaejun Yu,[1] T. W. Noh,[2] C. Kim,[5] S.-J. Oh,[1]
J.-H. Park,[3,4,*] V. Durairaj,[6] G. Cao,[6] and E. Rotenberg[7]



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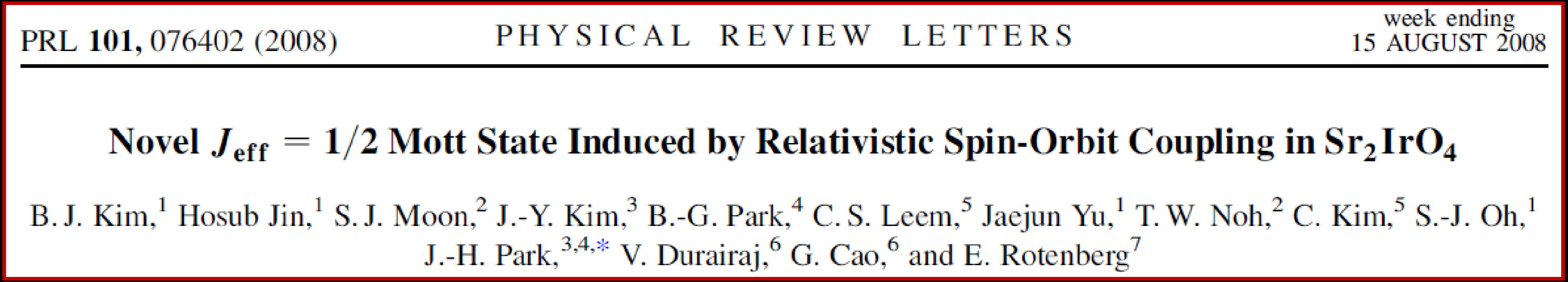

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PRL **108,** 0864 PHYSICAL REVIEW LETTERS week ending 24 FEBRUARY 2012

***Ab initio* Studies on the Interplay between Spin-Orbit Interaction and Coulomb Correlation in Sr_2IrO_4 and Ba_2IrO_4**

R. Ar[1,2,3] J. Kuneš,[4] A. V. Kozhevnikov,[5] A. G. Eguiluz,[6] and M. Imada[1,3]

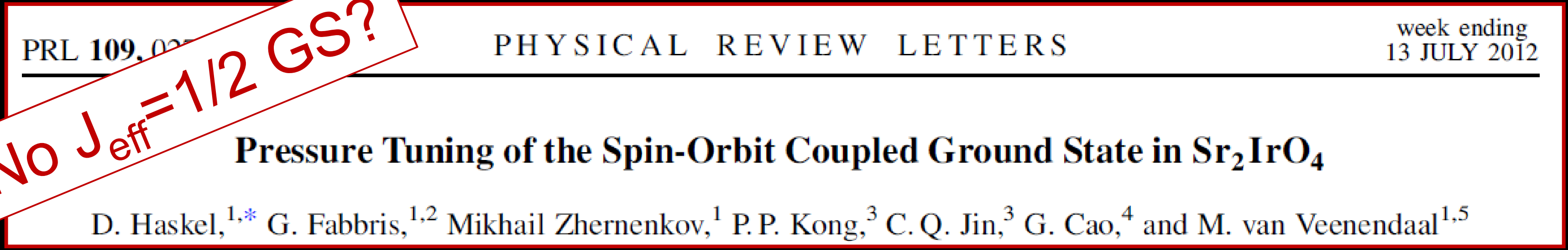
Slater Insulator?

PHYSICAL REVIEW B **86**, 035128 (2012)

Ol a metal-to-insulator transition with both Mott-Hubbard and Slater characteristics in Sr_2IrO_4 from time-resolved photocarrier dynamics

D. Hsieh,[1] F. Mahmood,[1] D. H. Torchinsky,[1] G. Cao,[2,3] and N. Gedik[1]

Both Mott-Slater?



PRL **109,** 0 PHYSICAL REVIEW LETTERS week ending 13 JULY 2012

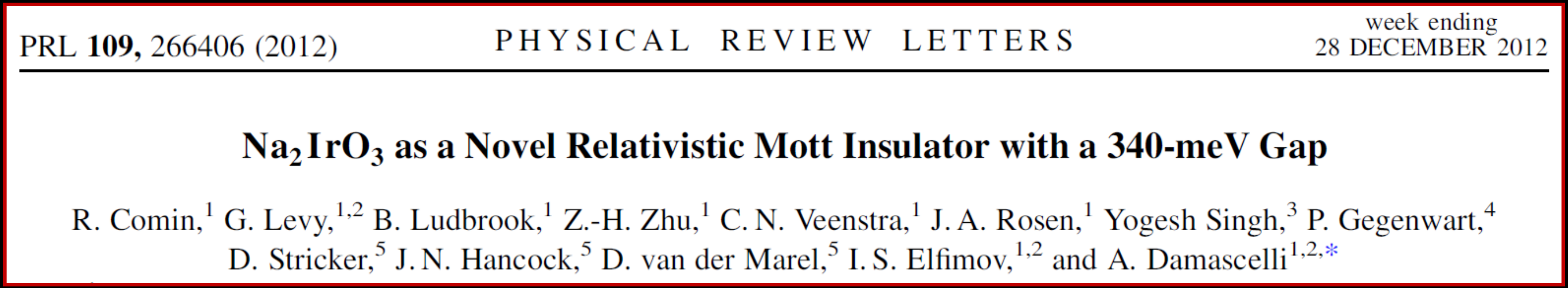
Pressure Tuning of the Spin-Orbit Coupled Ground State in Sr_2IrO_4

D. Haskel,[1,*] G. Fabbris,[1,2] Mikhail Zhernenkov,[1] P. P. Kong,[3] C. Q. Jin,[3] G. Cao,[4] and M. van Veenendaal[1,5]

No J_{eff}=1/2 GS?

Interplay of Coulomb U and SO in 5d iridates

Could be Na_2IrO_3 a clear cut case?



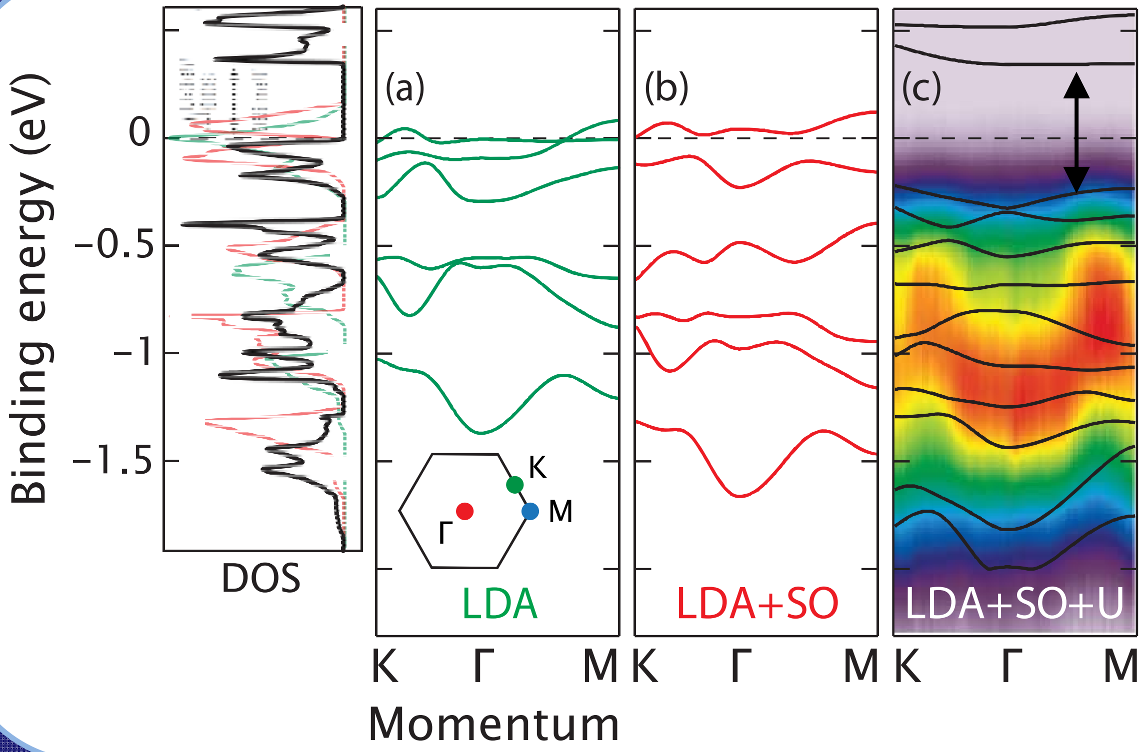
PRL **109**, 266406 (2012) PHYSICAL REVIEW LETTERS week ending 28 DECEMBER 2012

Na_2IrO_3 as a Novel Relativistic Mott Insulator with a 340-meV Gap

R. Comin,[1] G. Levy,[1,2] B. Ludbrook,[1] Z.-H. Zhu,[1] C. N. Veenstra,[1] J. A. Rosen,[1] Yogesh Singh,[3] P. Gegenwart,[4] D. Stricker,[5] J. N. Hancock,[5] D. van der Marel,[5] I. S. Elfimov,[1,2] and A. Damascelli[1,2,*]

T_N = 15K ⟶ distinguish Mott from Slater

Including Correlations: LDA+SO+U



We must include on-site correlations via U to get the right gap and local ½ moments

Would this work without SO?

Not really..

Na_2IrO_3 is a Relativistic Mott Insulator:

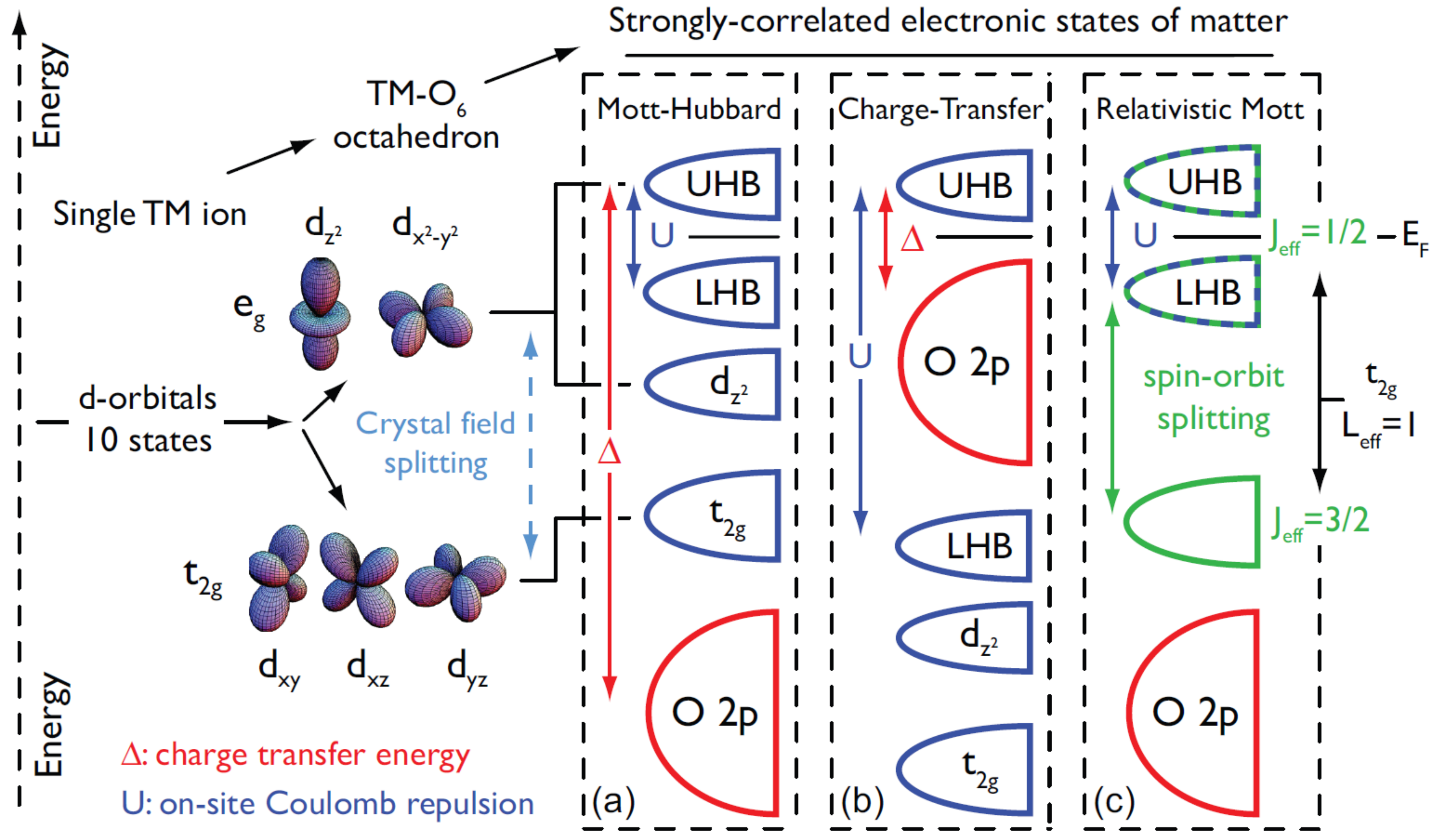
Coulomb U and SO coupling cannot be decoupled

Mott criterion U>W

Early 3d TMO

Late 3d TMO

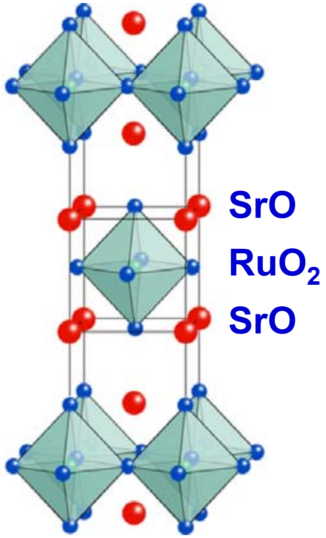
5d TMO



What about 4d oxides? Let’s consider Sr_2RuO_4

Sr_2RuO_4: p-wave Superconductivity

2D perovskite

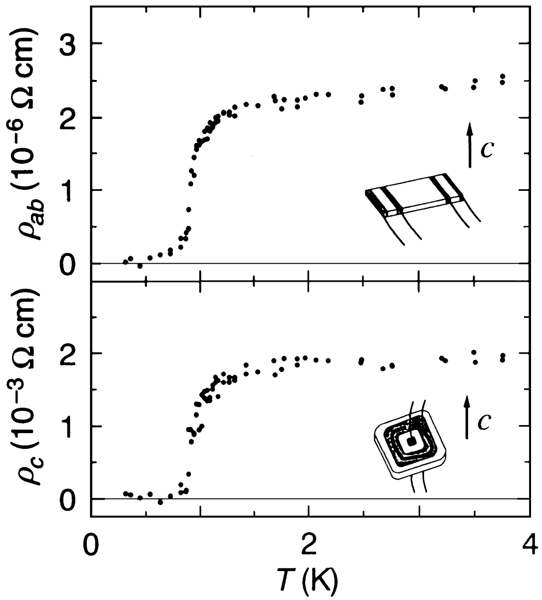


Unconventional superconductivity

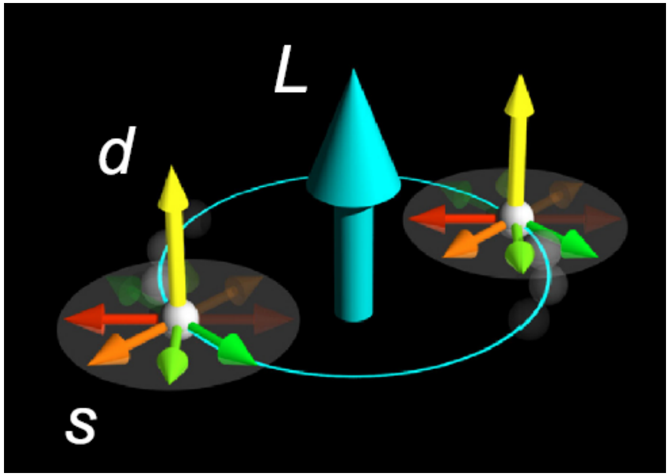
- Pairing mechanism ?
- Order parameter ?
- FM-AF fluctuations ?

Rice & Sigrist, JPCM **7**, L643 (1995)

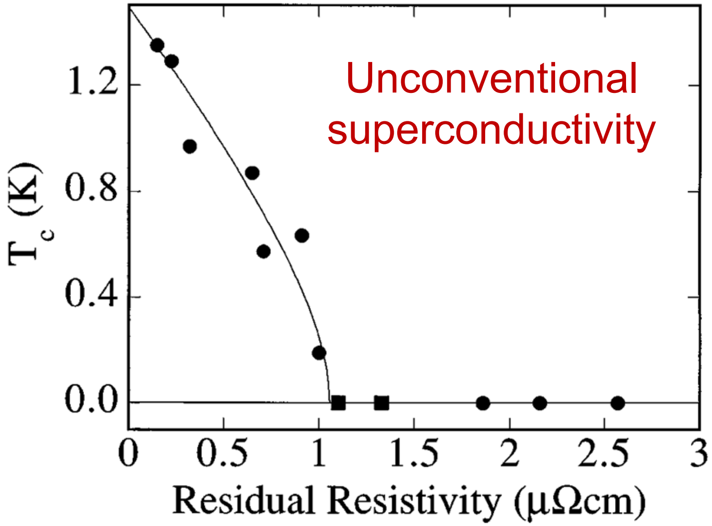
Maeno *et al*., Nature **372**, 532 (1994)



Mackenzie & Maeno, RMP **75**, 657 (2003)



Mackenzie *et al*., PRL **80**, 161 (1998)



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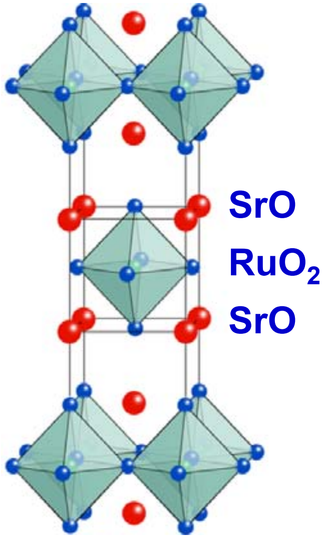

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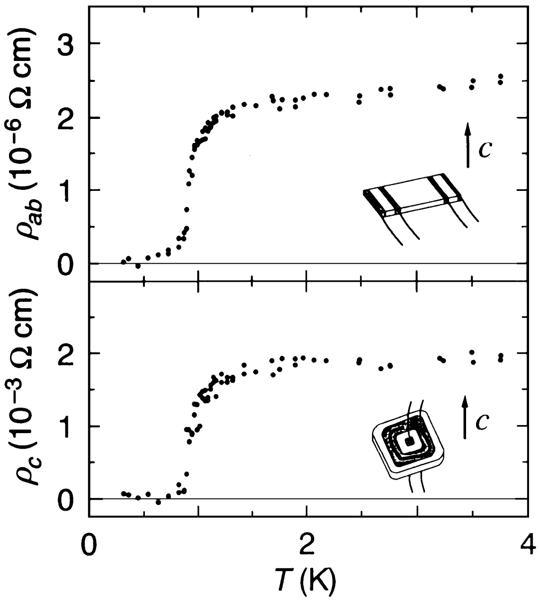

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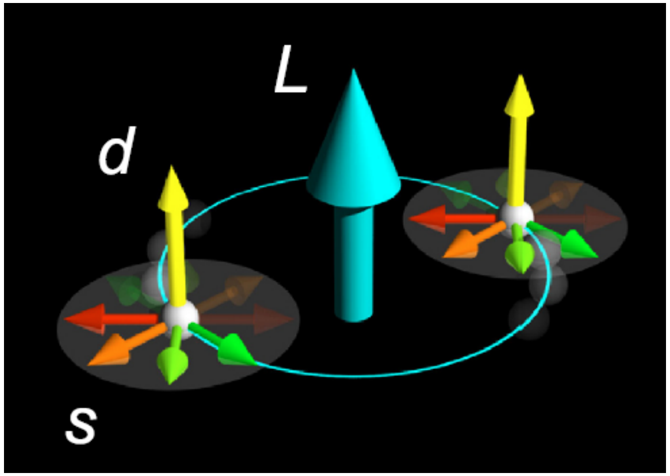
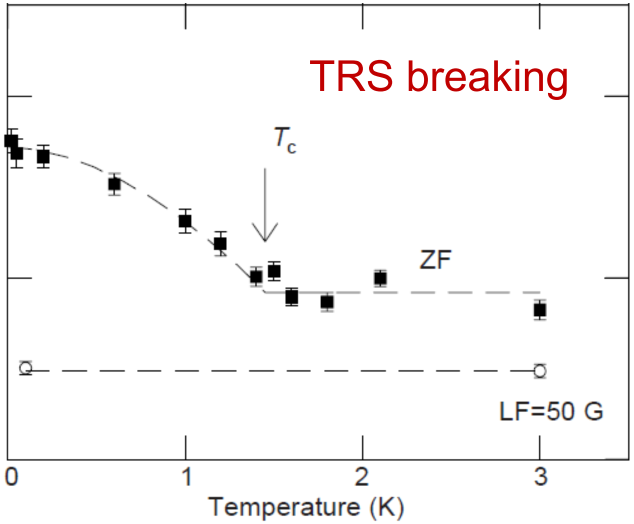


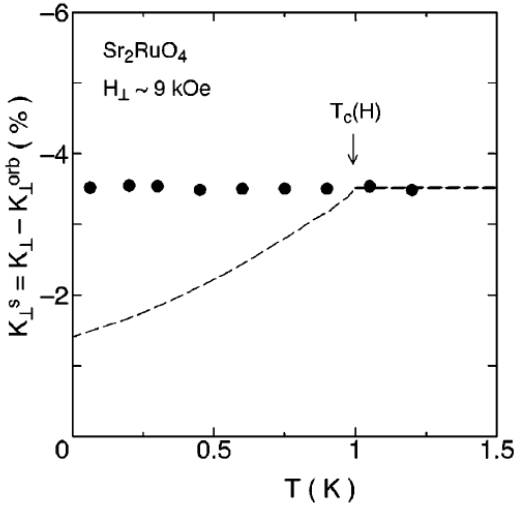
Luke *et al*., Nature **394**, 558 (1998)



Sr_2RuO_4: Evidence for Spin-triplet Pairing

Knight shift

Microscopic spin susceptibility



Ishida et al., Nature (1998)

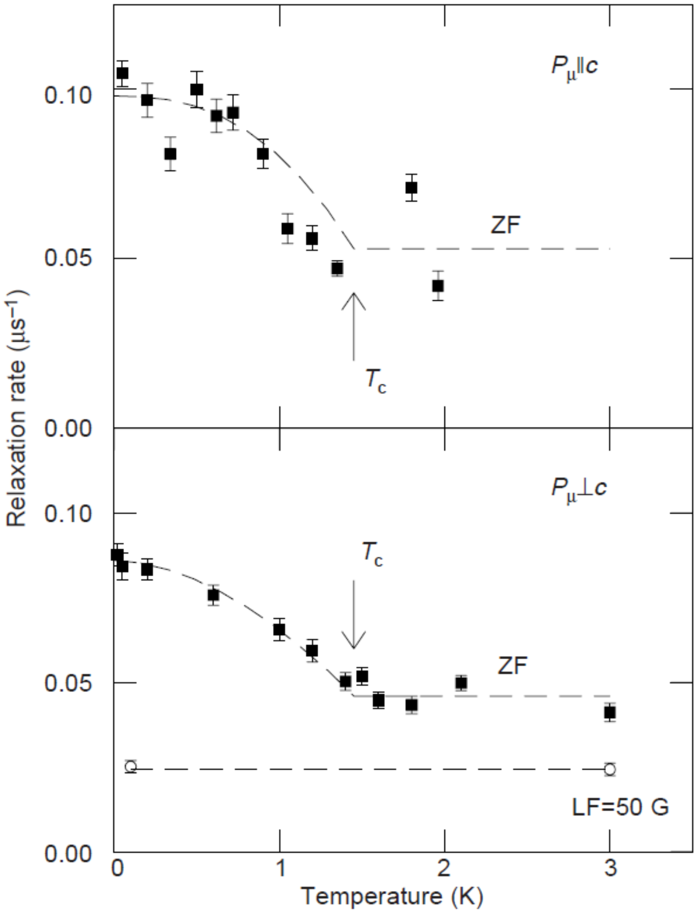
Phase sensitive measurements

verified the odd-parity orbital pairing symmetry due to the formation of spin-triplet Cooper pairs

Nelson et al., Science (2004)

μSR Relaxation rate

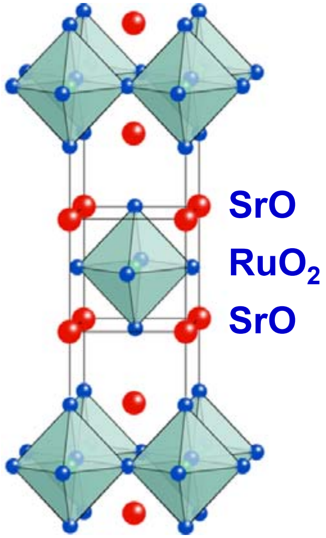
Time-reversal symmetry breaking



Luke et al., Nature (1998)

Sr_2RuO_4: p-wave Superconductivity

2D perovskite



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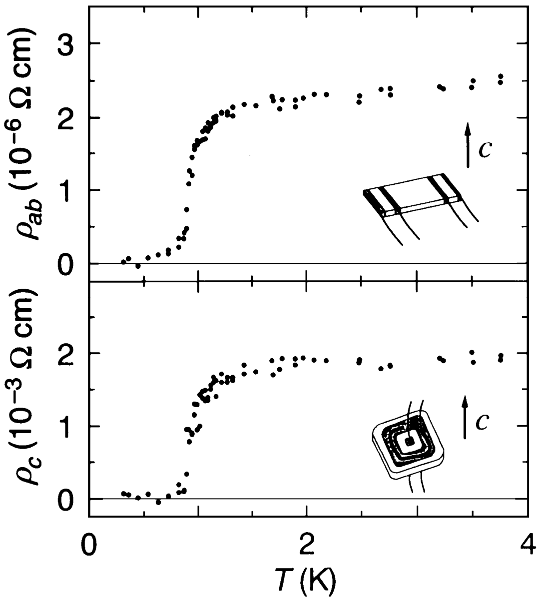

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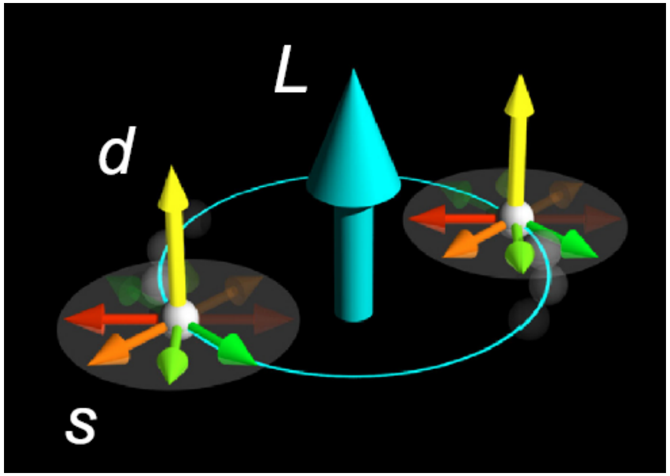
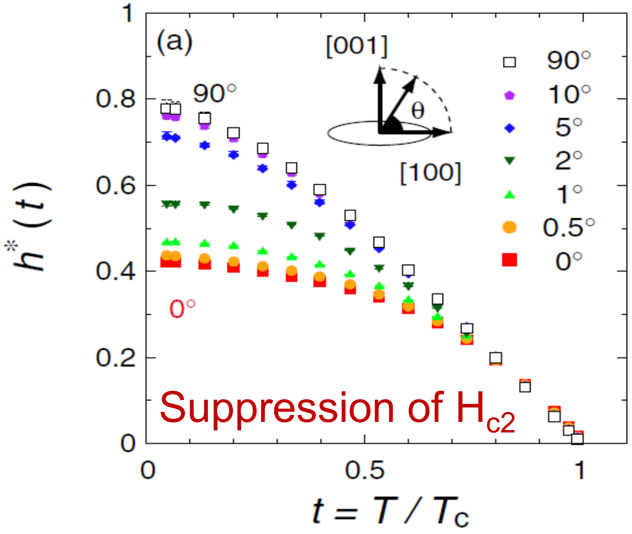
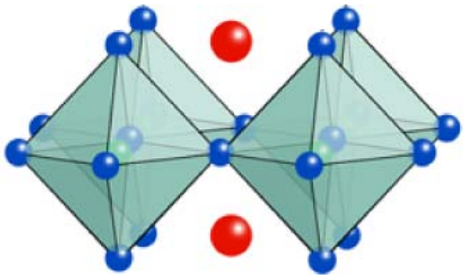


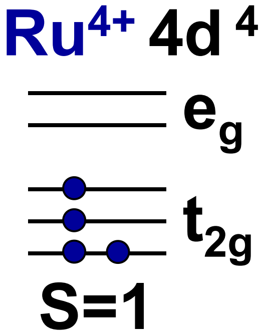
Maeno et al., JPSJ (2012)

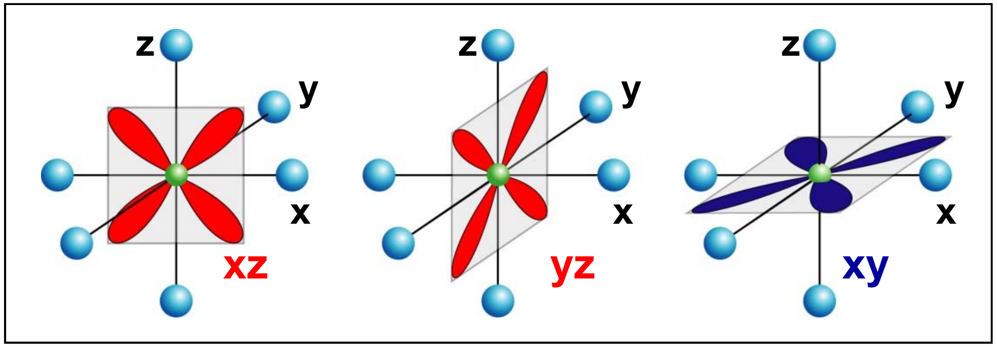


Suppression of H_{c2}

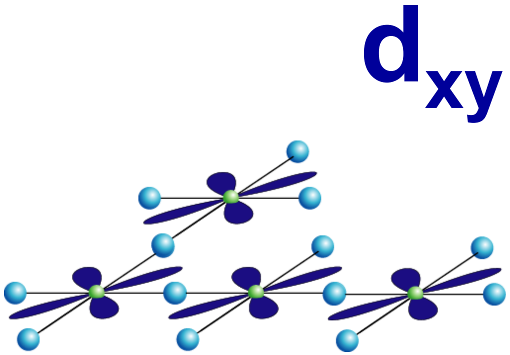
1D ($d_{xz,yz}$) versus 2D (d_{xy}) Superconductivity ?

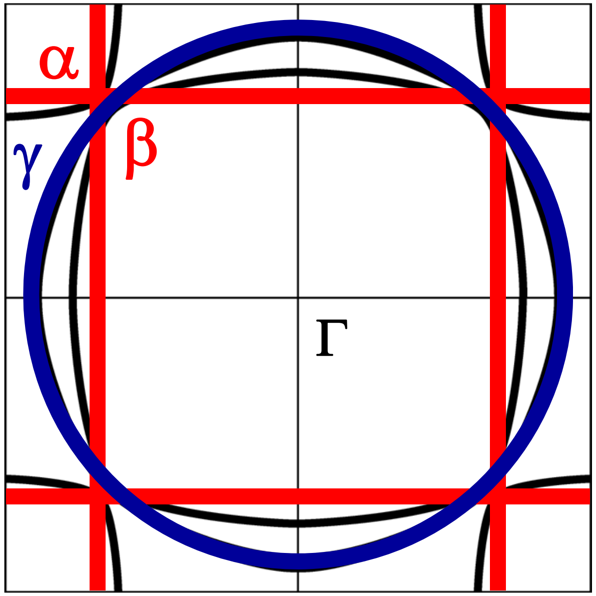


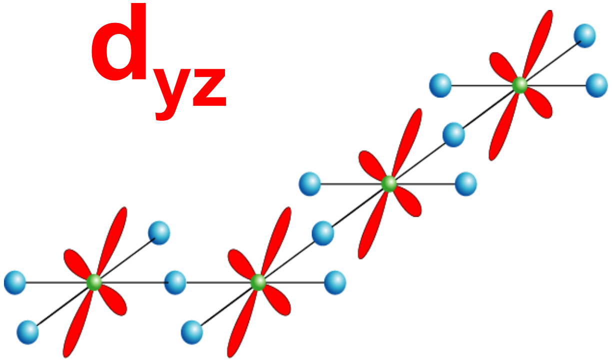




- Band structure calculation: **3 t_{2g}** bands crossing **E_F**



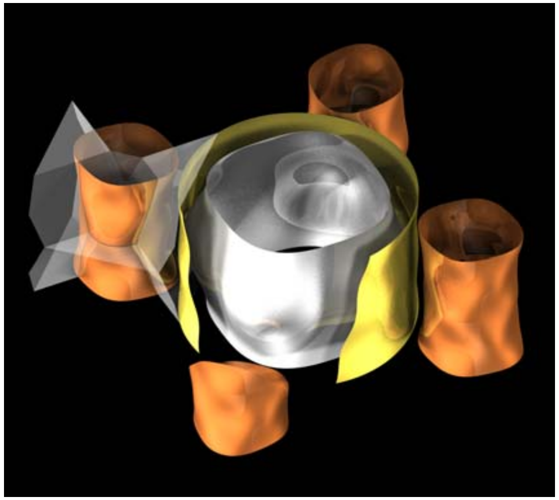




Mazin *et al.*, PRL **79**, 733 (1997)

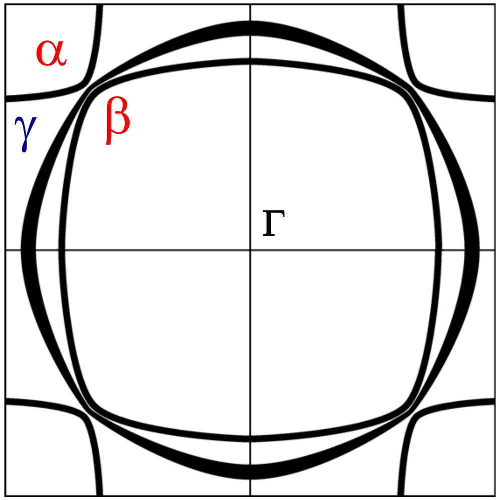
The Fermi Surface of Sr_2RuO_4

de Haas-van Alphen



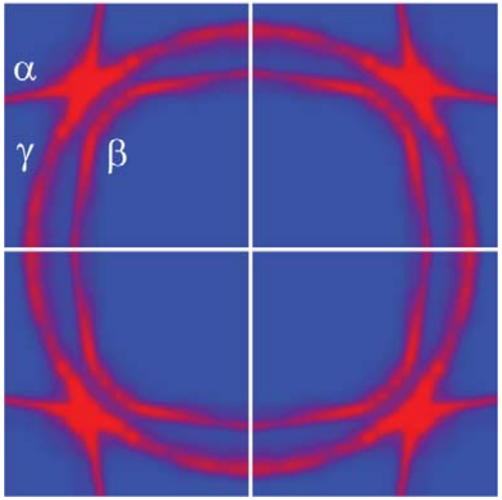
Bergemann *et al.*, PRL **84**, 2662 (2000)

LDA



Mazin *et al.*, PRL **79**, 733 (1997)

ARPES



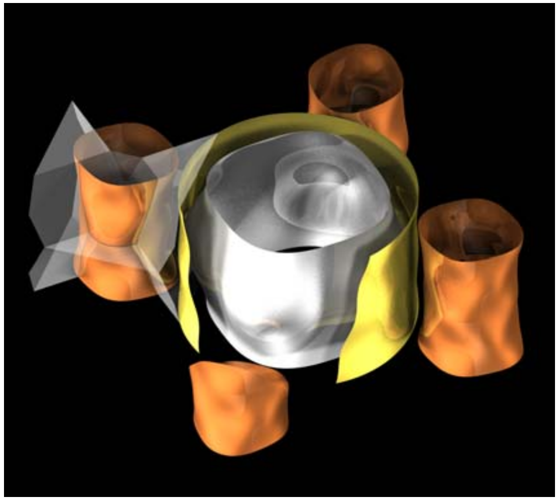
Damascelli *et al*, PRL **85**, 5194 (2000)

The Fermi Surface of Sr_2RuO_4

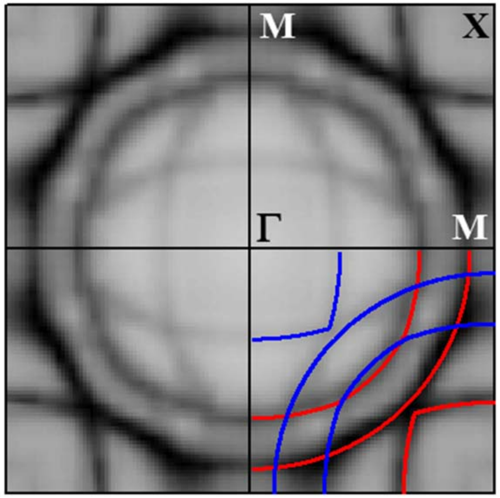
de Haas-van Alphen

Surface

Bulk

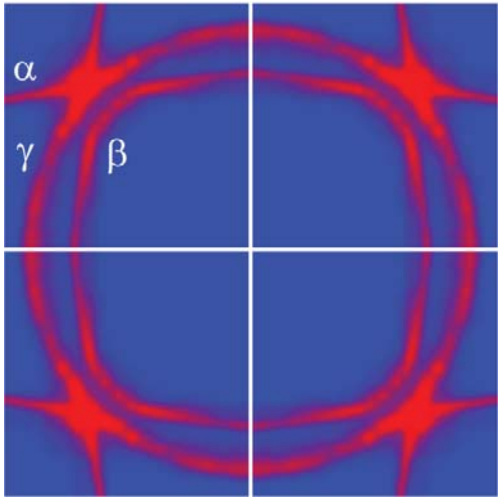


Bergemann *et al.*, PRL **84**, 2662 (2000)



Damascelli *et al*, PRL **85**, 5194 (2000)

Cleaved at 10K



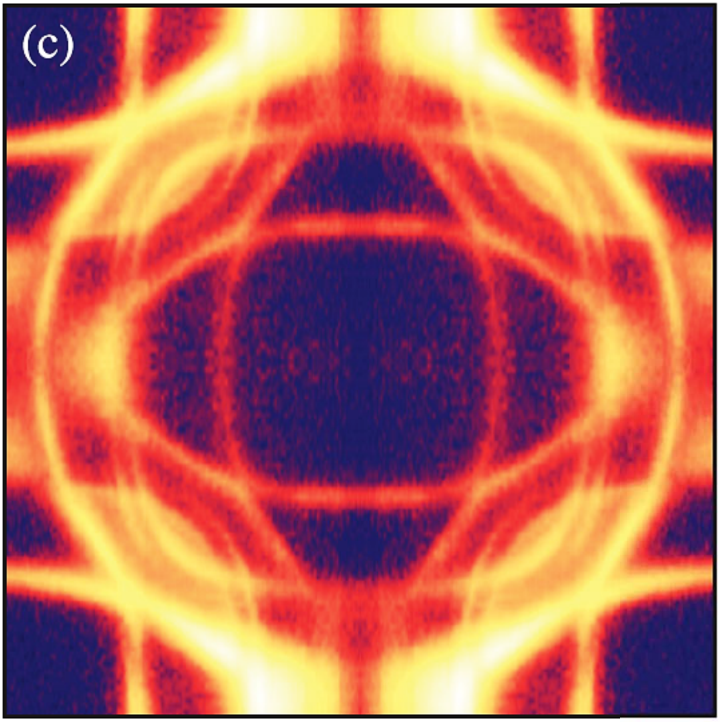
Damascelli *et al*, PRL **85**, 5194 (2000)

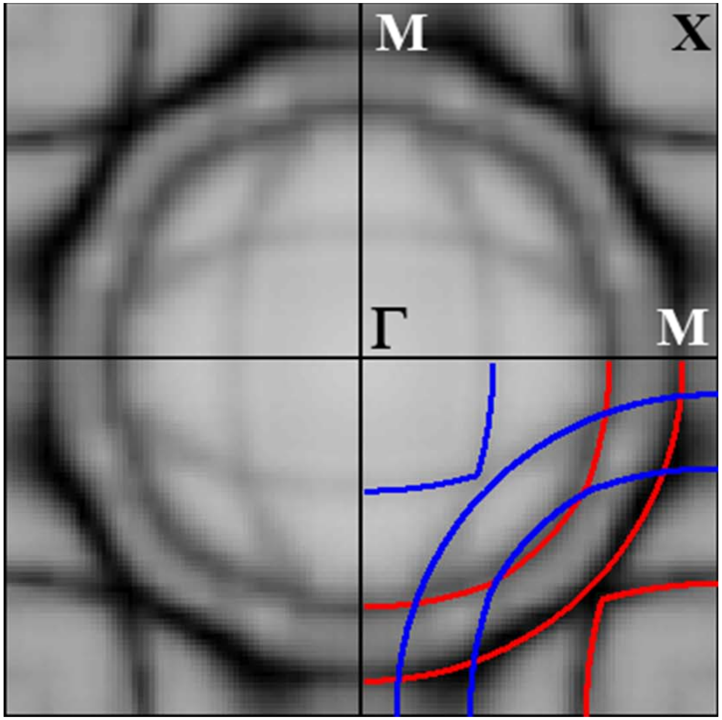
Cleaved at 200K

Structural surface reconstruction: rotation of RuO_6 octahedra

R. Matzdorf *et al.*, Science **289**, 746 (2000)

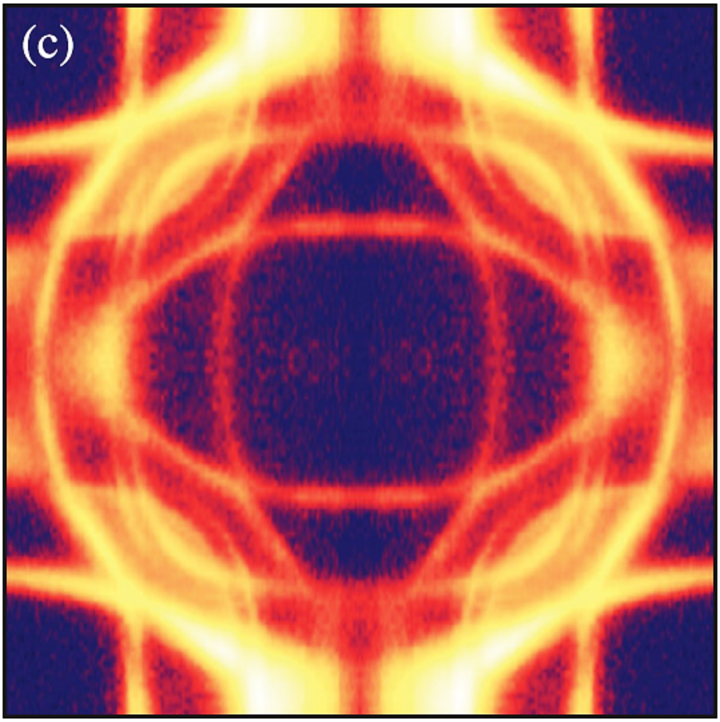
Determining the surface-to-bulk progression of the normal-state electronic structure of Sr_2RuO_4

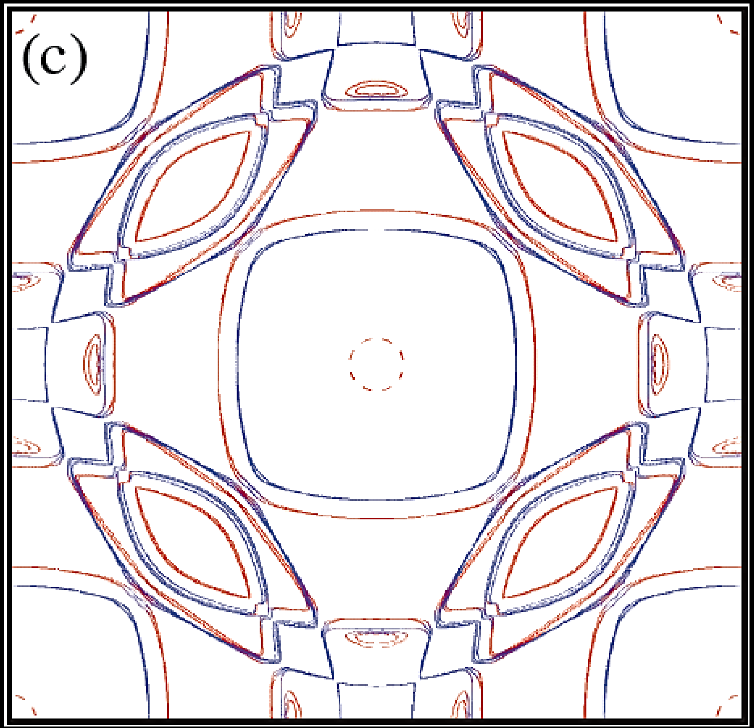




C.N. Veenstra et al. PRL **110**, 097004 (2013)

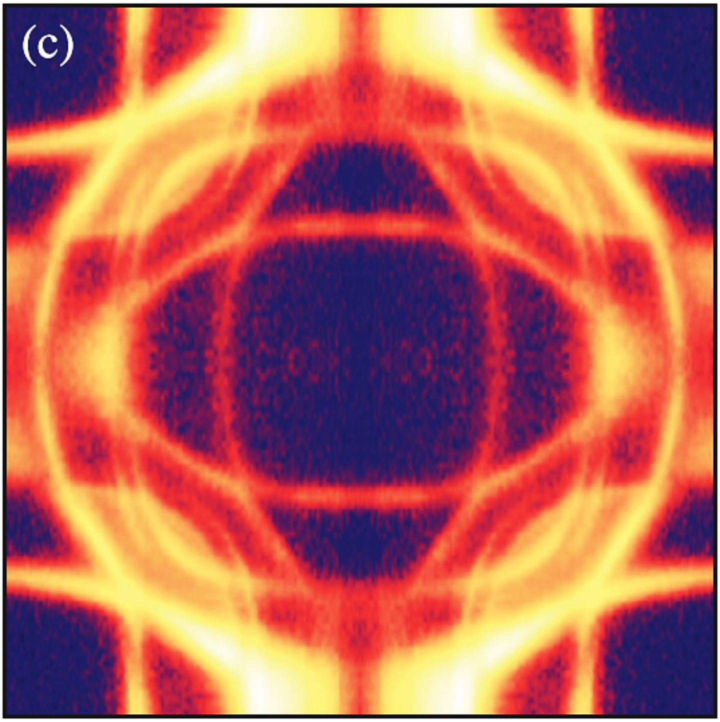
Determining the surface-to-bulk progression of the normal-state electronic structure of Sr_2RuO_4

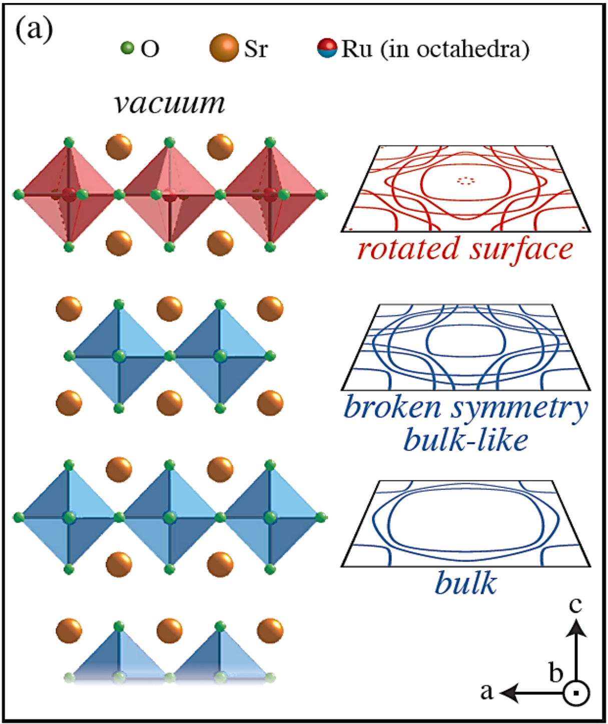




C.N. Veenstra et al. PRL **110**, 097004 (2013)

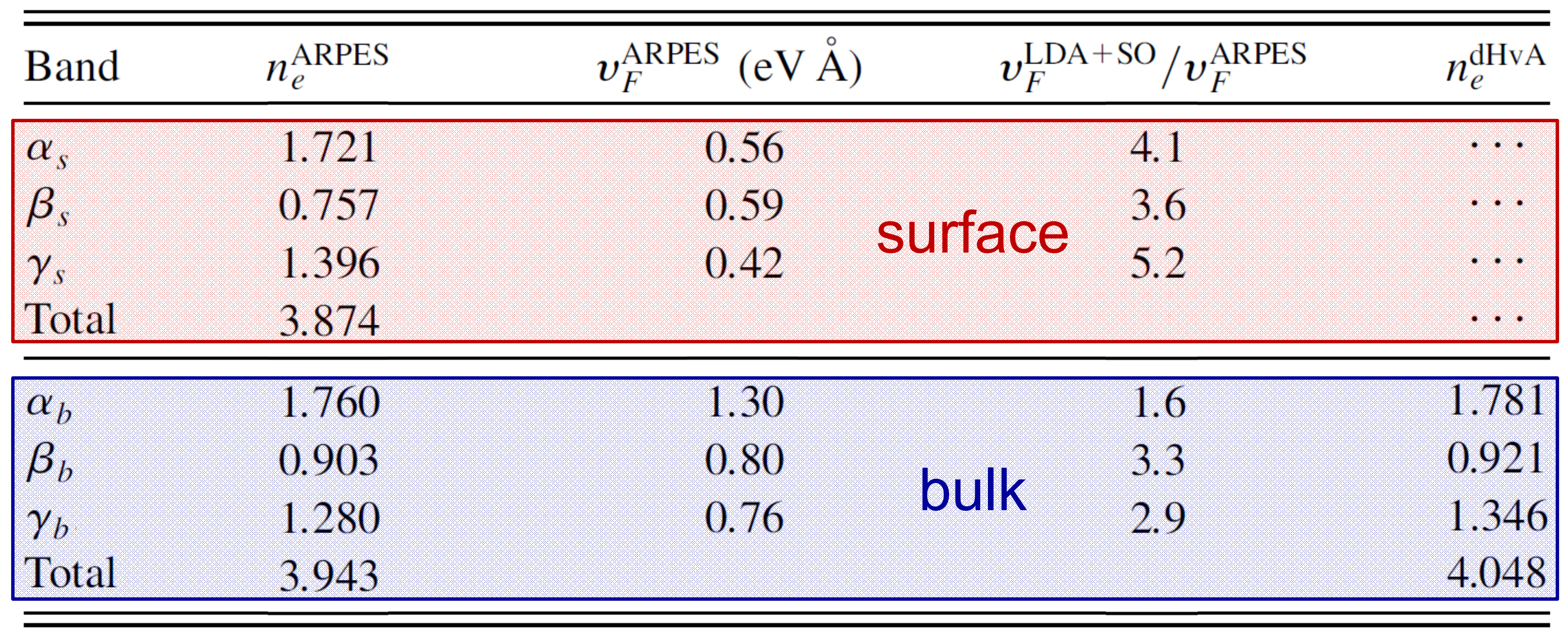
Determining the surface-to-bulk progression of the normal-state electronic structure of Sr_2RuO_4





C.N. Veenstra et al. PRL **110**, 097004 (2013)

Determining the surface-to-bulk progression of the normal-state electronic structure of Sr_2RuO_4



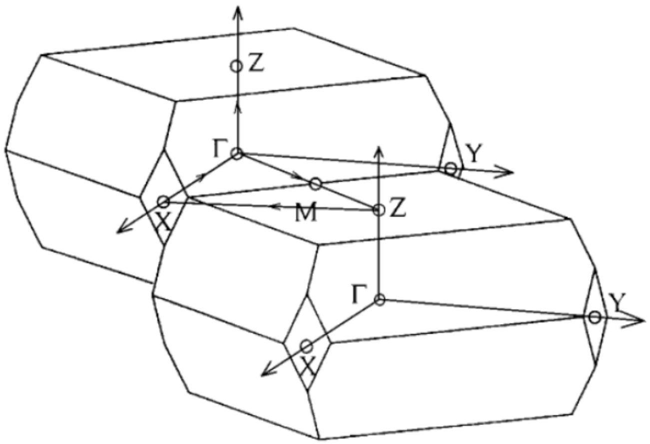
Band	n_e^{ARPES}	v_F^{ARPES} (eV Å)	$v_F^{\mathrm{LDA+SO}}/v_F^{\mathrm{ARPES}}$	n_e^{dHvA}
α_s	1.721	0.56	4.1	$\cdots$
β_s	0.757	0.59	3.6	$\cdots$
γ_s	1.396	0.42	5.2	$\cdots$
Total	3.874			$\cdots$
α_b	1.760	1.30	1.6	1.781
β_b	0.903	0.80	3.3	0.921
γ_b	1.280	0.76	2.9	1.346
Total	3.943			4.048

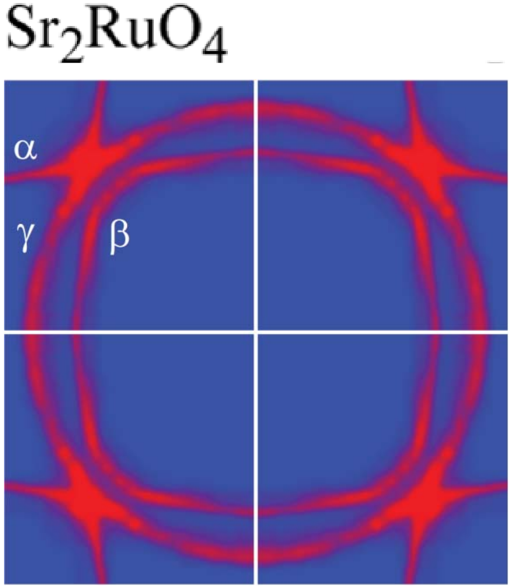
surface

bulk

C.N. Veenstra et al. PRL **110**, 097004 (2013)

Importance of Spin-Orbit Coupling in 4d Oxides





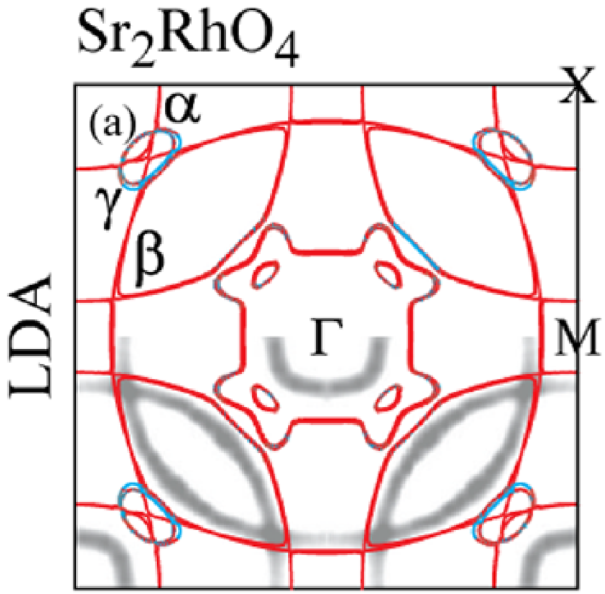
Damascelli *et al*, PRL **85**, 5194 (2000)

Why LDA for Rh214 does not work as well?
Something is missing in LDA!

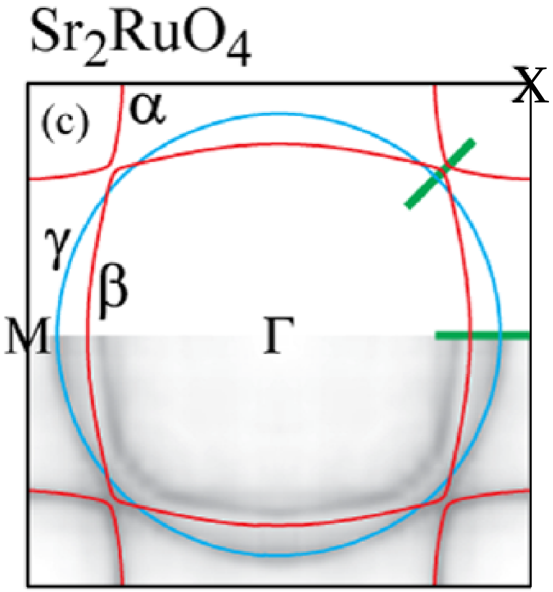
Correlations? Structure? Surface?

M.W. Haverkort, I.S. Elfimov, L.H. Tjeng, G.A. Sawatzky, A. Damascelli, PRL **101**, 026406 (2008)

Importance of Spin-Orbit Coupling in 4d Oxides



Kim *et al*, PRL **97**, 106401 (2006)
Baumberger *et al*, PRL **96**, 246402 (2006)



Damascelli *et al*, PRL **85**, 5194 (2000)

Orbital character
xy
xz
yz

Why LDA for Rh214 does not work as well?
Something is missing in LDA!

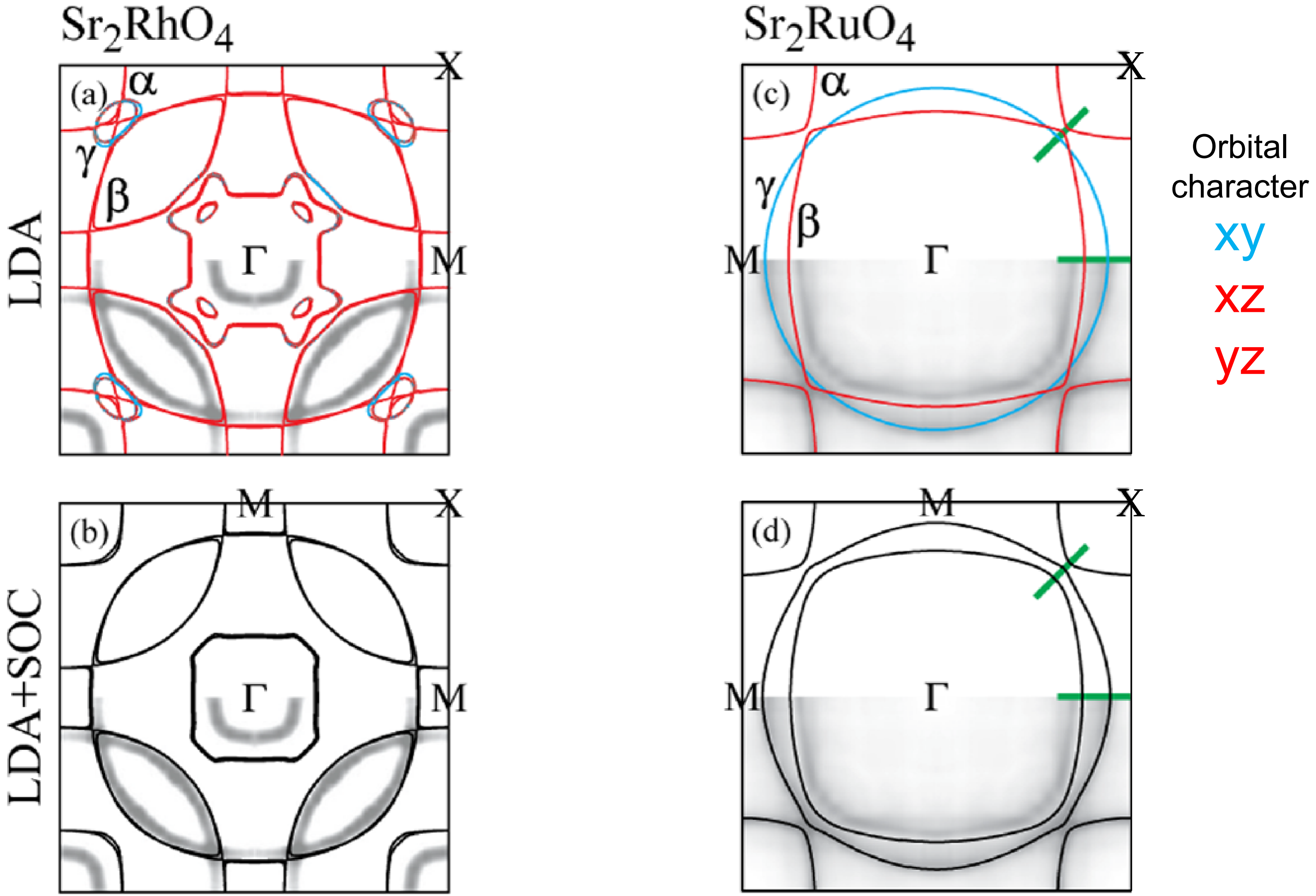
$$H = \zeta \sum_i l_i \cdot s_i$$

Spin-Orbit Coupling

Atomic relativistic SOC
Ru^{4+} ζ=161 meV
Rh^{4+} ζ=191 meV

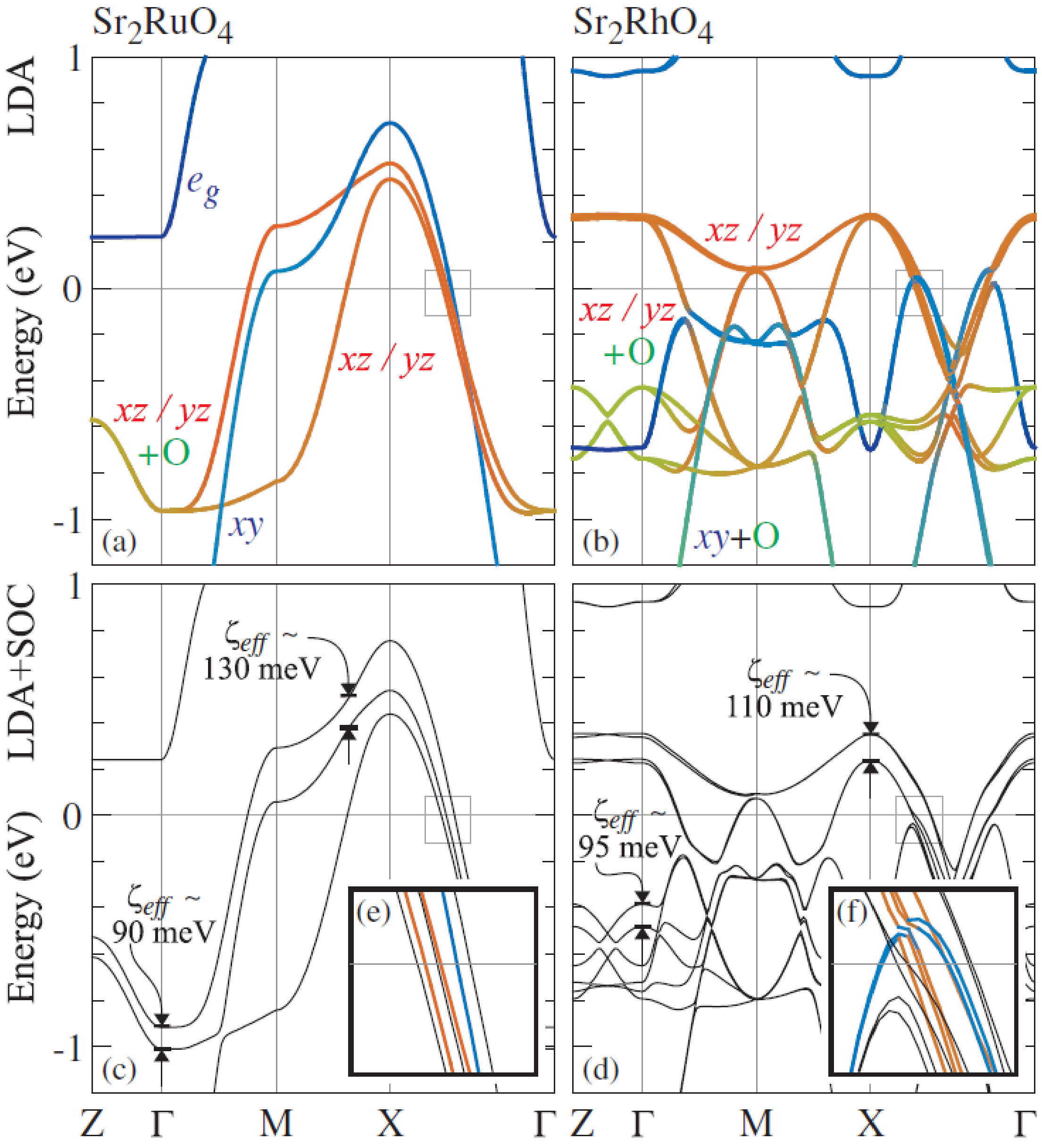
M.W. Haverkort, I.S. Elfimov, L.H. Tjeng, G.A. Sawatzky, A. Damascelli, PRL **101**, 026406 (2008)

Importance of Spin-Orbit Coupling in 4d Oxides



M.W. Haverkort, I.S. Elfimov, L.H. Tjeng, G.A. Sawatzky, A. Damascelli, PRL **101**, 026406 (2008)

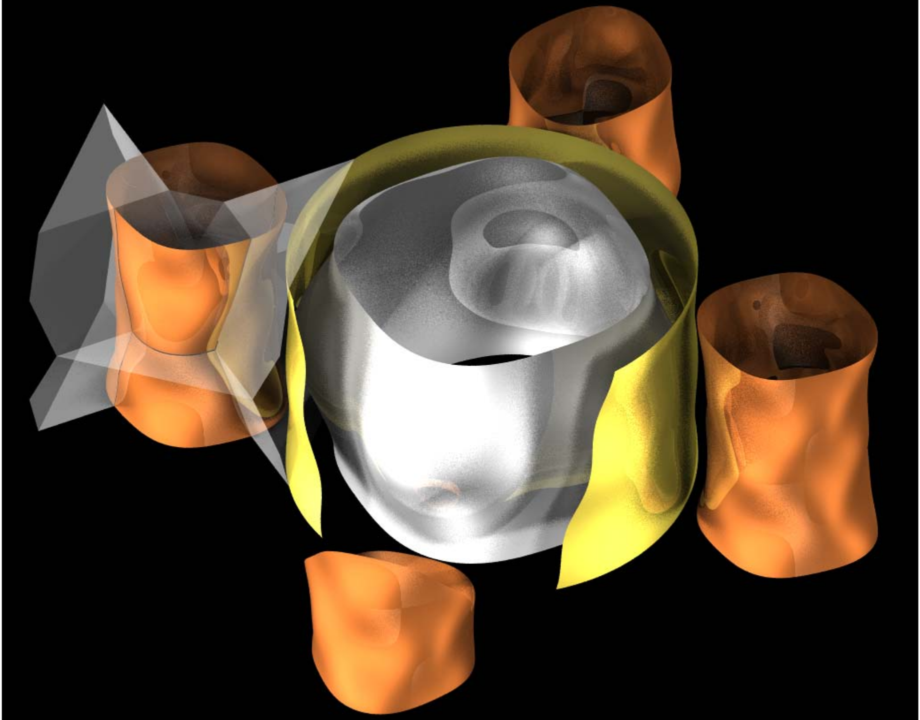
When is Spin-Orbit important? Degeneracy at E_F !



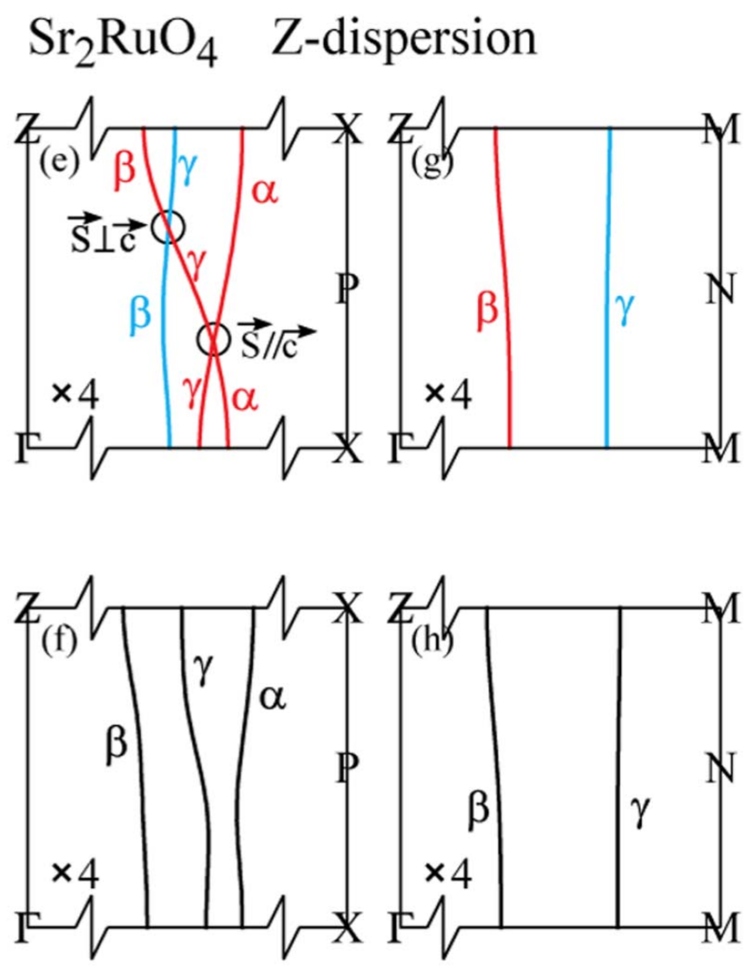
M.W. Haverkort, I.S. Elfimov, L.H. Tjeng, G.A. Sawatzky, A. Damascelli, PRL **101**, 026406 (2008)

SO coupling suppresses the band dispersion along c axis

Weak k_z dispersion in dHvA



Bergemann *et al.*, PRL **84**, 2662 (2000)



No SO coupling

With SO coupling

M.W. Haverkort, I.S. Elfimov, L.H. Tjeng, G.A. Sawatzky, A. Damascelli, PRL **101**, 026406 (2008)

SO coupling suppresses the band dispersion along c axis

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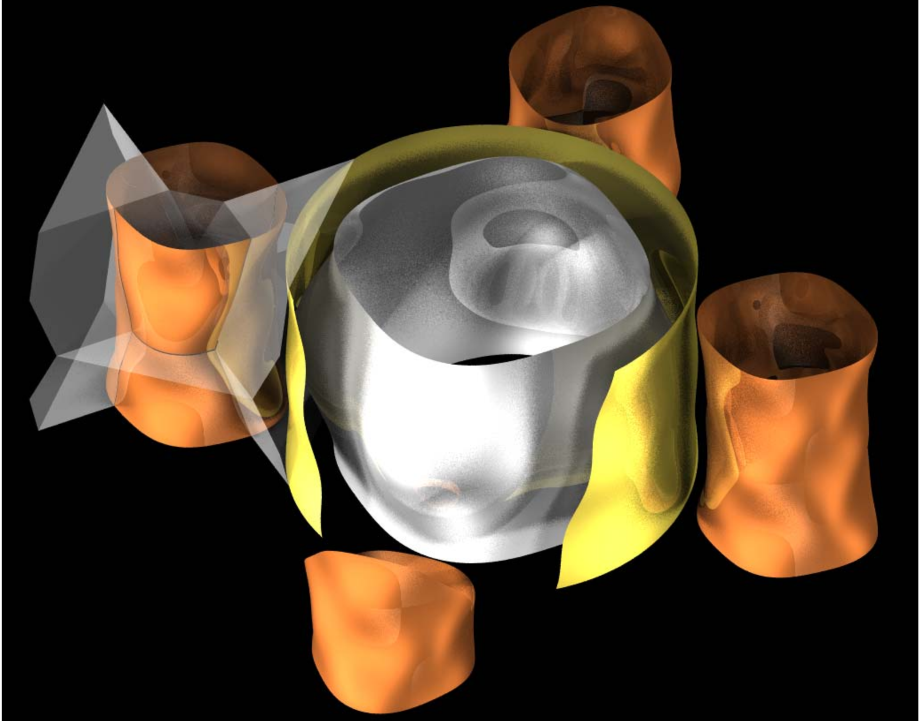
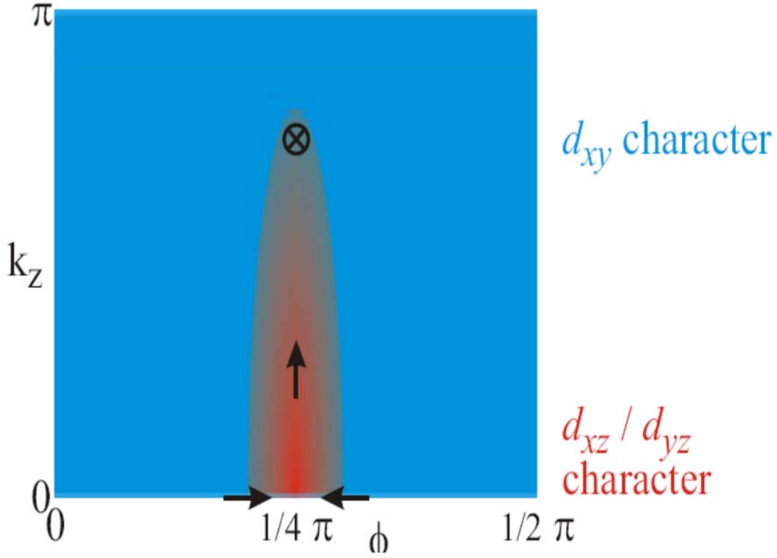
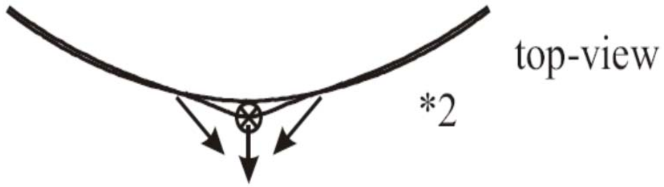


Bergemann *et al.*, PRL **84**, 2662 (2000)



K-dependent Spin Anisotropy

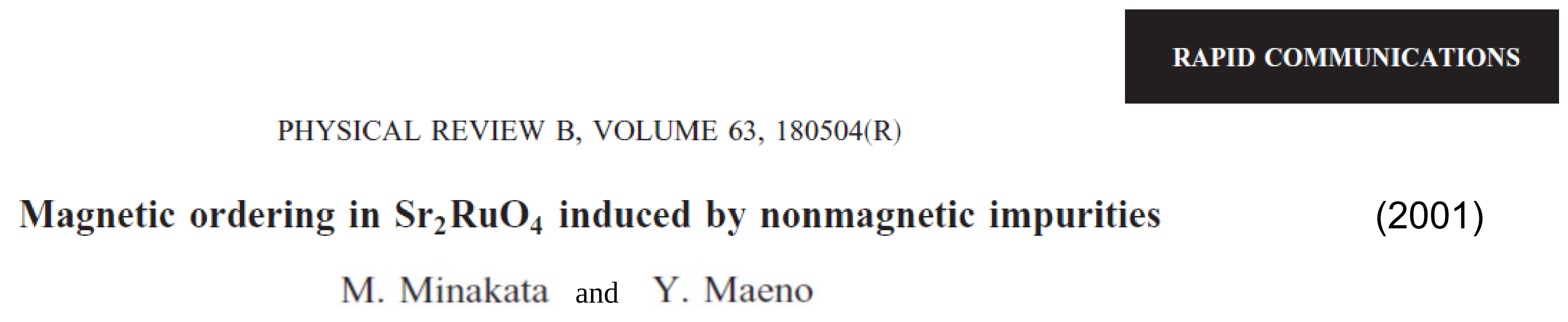


Spin-Triplet Superconductivity?

SO coupling leads to strong k_z dispersion of quantization axis

M.W. Haverkort, I.S. Elfimov, L.H. Tjeng, G.A. Sawatzky, A. Damascelli, PRL **101**, 026406 (2008)

Magnetic Anisotropy and Spin Fluctuations in Sr_2RuO_4



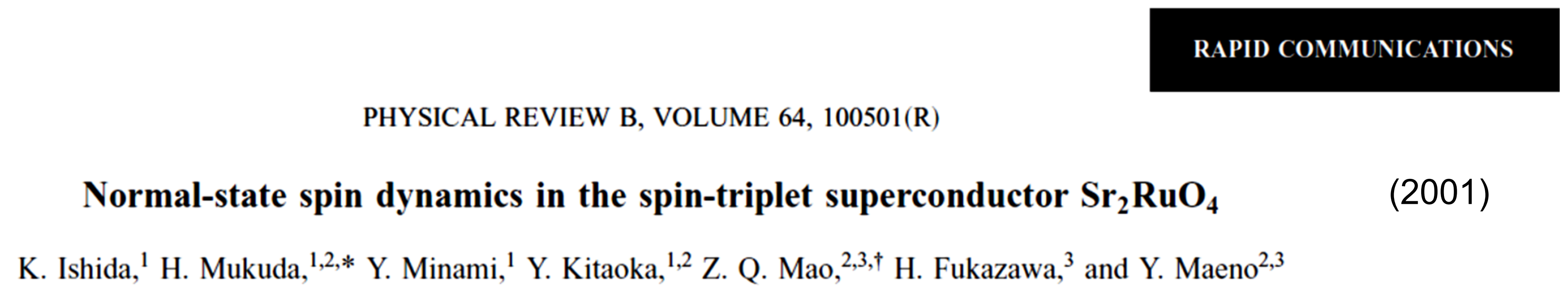
RAPID COMMUNICATIONS

PHYSICAL REVIEW B, VOLUME 63, 180504(R)

Magnetic ordering in Sr_2RuO_4 induced by nonmagnetic impurities

(2001)

M. Minakata and Y. Maeno



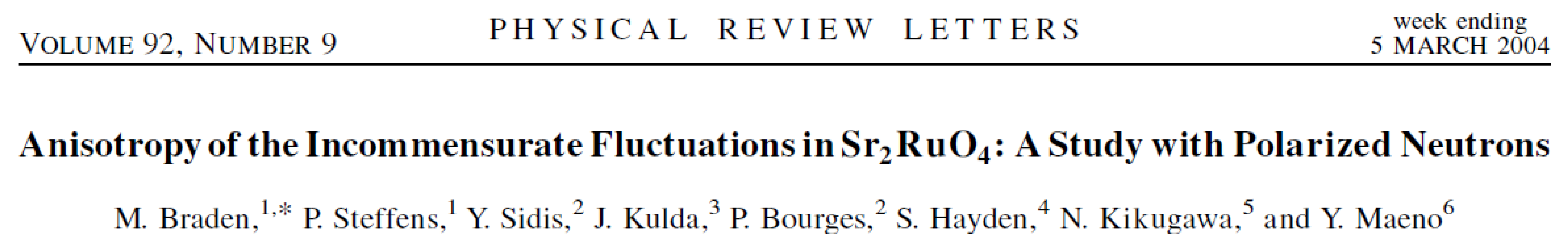
RAPID COMMUNICATIONS

PHYSICAL REVIEW B, VOLUME 64, 100501(R)

Normal-state spin dynamics in the spin-triplet superconductor Sr_2RuO_4

(2001)

K. Ishida,[1] H. Mukuda,[1,2,*] Y. Minami,[1] Y. Kitaoka,[1,2] Z. Q. Mao,[2,3,†] H. Fukazawa,[3] and Y. Maeno[2,3]



VOLUME 92, NUMBER 9 PHYSICAL REVIEW LETTERS week ending 5 MARCH 2004

Anisotropy of the Incommensurate Fluctuations in Sr_2RuO_4: A Study with Polarized Neutrons

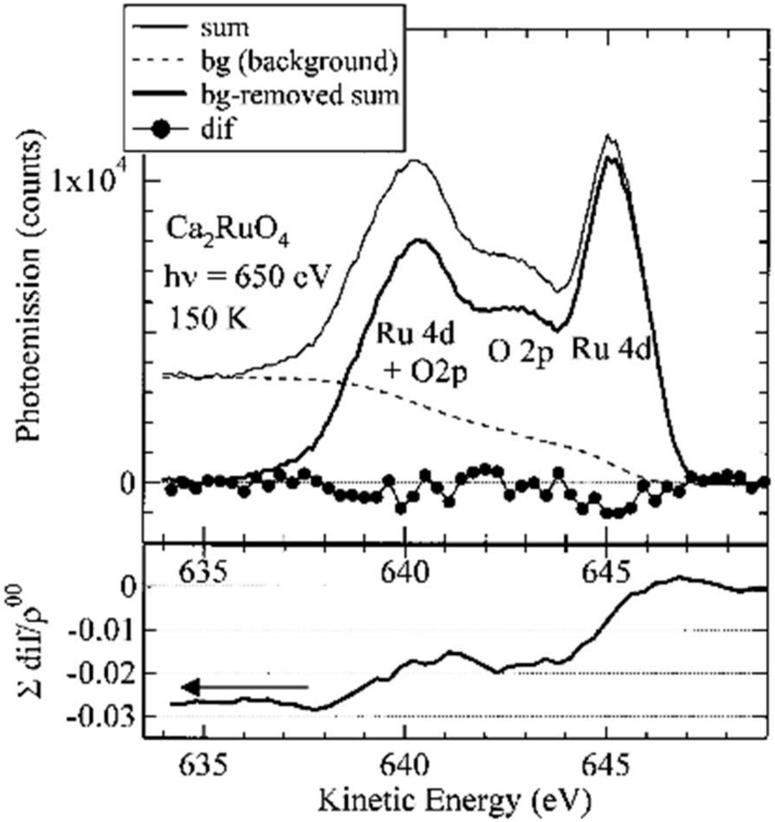
M. Braden,[1,*] P. Steffens,[1] Y. Sidis,[2] J. Kulda,[3] P. Bourges,[2] S. Hayden,[4] N. Kikugawa,[5] and Y. Maeno[6]

k-dependent enhanced out-of-plane dynamic susceptibility

Spin-resolved Circularly-polarized Photoemission

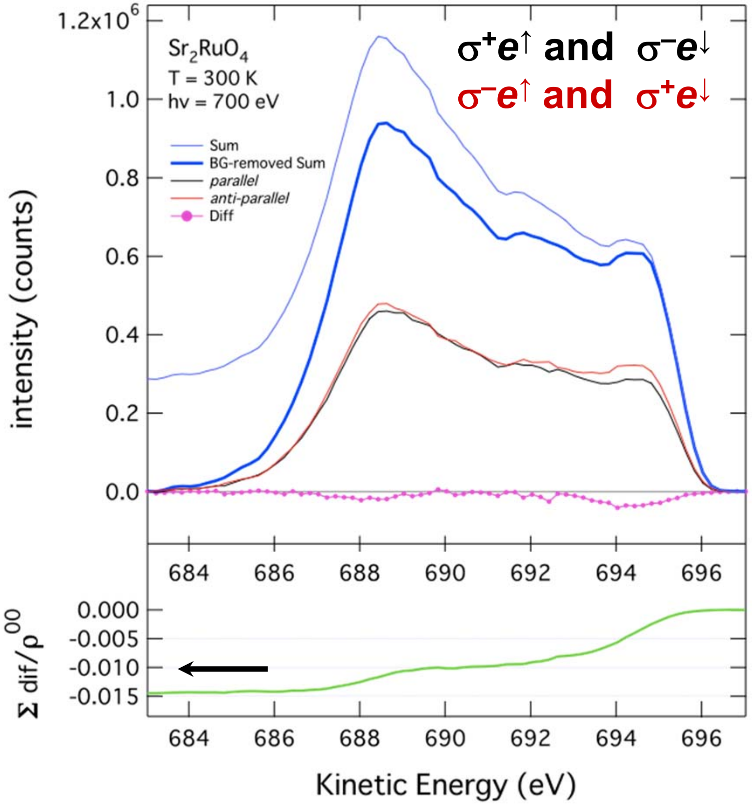
Ca_2RuO_4

T. Mizokawa, L.H.Tjeng *et al.* PRL **87**, 077202 (2001)



Sr_2RuO_4

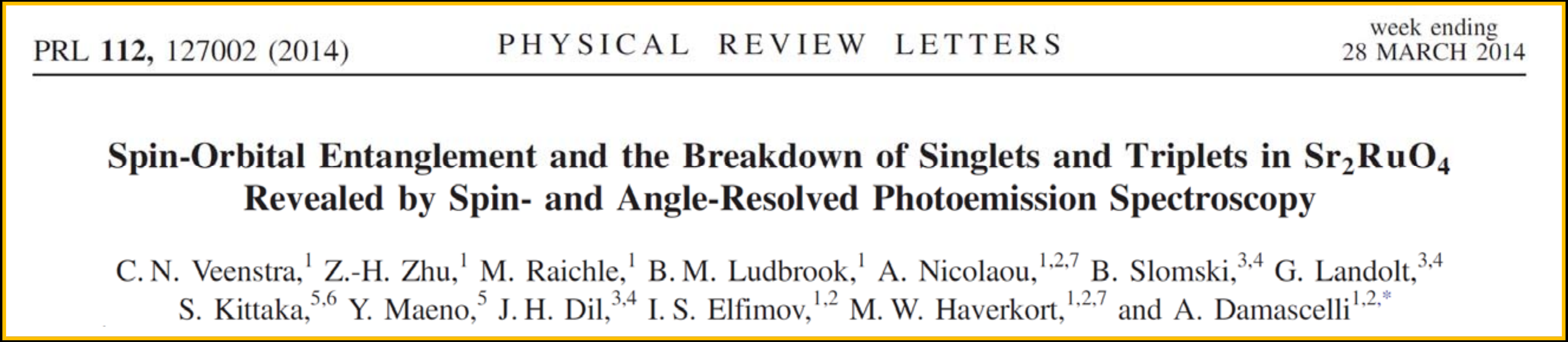
H. Fujiwara, L.H.Tjeng *et al.* (unpublished)



The expectation value of the Spin-Orbit coupling can be experimentally determined by sum rule : $\Sigma diff / \rho^{00} \propto \langle \Sigma_i l_{z,i} \cdot s_{z,i} \rangle$. [G. Van der Laan and B. T. Thole PRB **48**, 210 (1993)]

Spin-orbit coupling is important for both Ca_2RuO_4 and Sr_2RuO_4

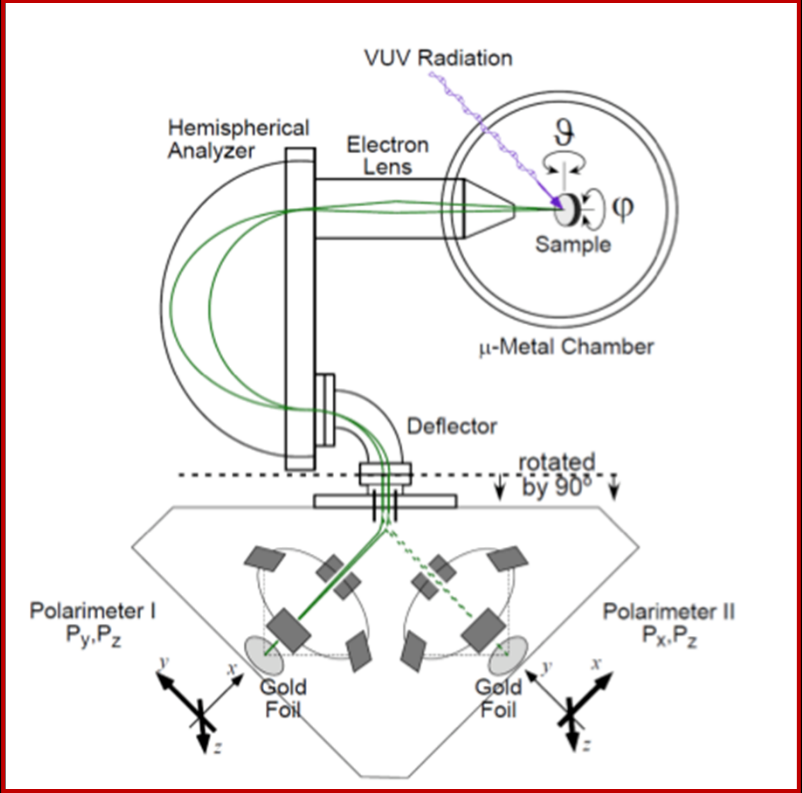
Direct measurement of SO: Spin-resolved ARPES



PRL **112,** 127002 (2014) PHYSICAL REVIEW LETTERS week ending 28 MARCH 2014

Spin-Orbital Entanglement and the Breakdown of Singlets and Triplets in Sr_2RuO_4 Revealed by Spin- and Angle-Resolved Photoemission Spectroscopy

C. N. Veenstra,[1] Z.-H. Zhu,[1] M. Raichle,[1] B. M. Ludbrook,[1] A. Nicolaou,[1,2,7] B. Slomski,[3,4] G. Landolt,[3,4] S. Kittaka,[5,6] Y. Maeno,[5] J. H. Dil,[3,4] I. S. Elfimov,[1,2] M. W. Haverkort,[1,2,7] and A. Damascelli[1,2,*]

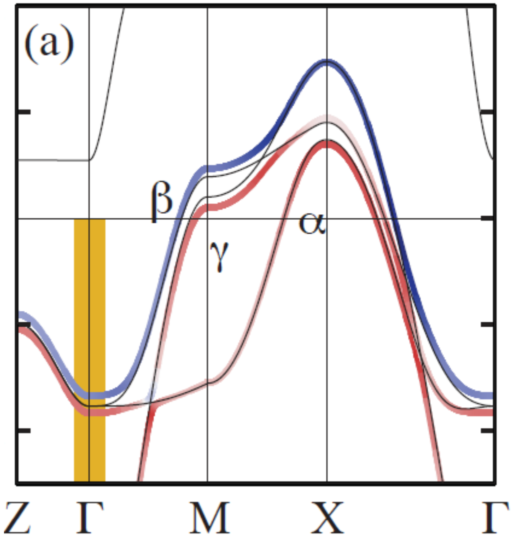


Conventional ARPES

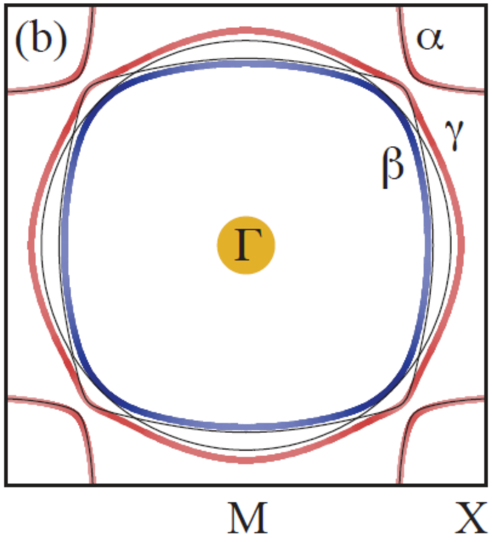
No SO-split states in spin-integrated ARPES

Direct measurement of SO: Spin-resolved ARPES

Band structure +
spin-orbit coupling



Fermi surface +
spin-orbit coupling

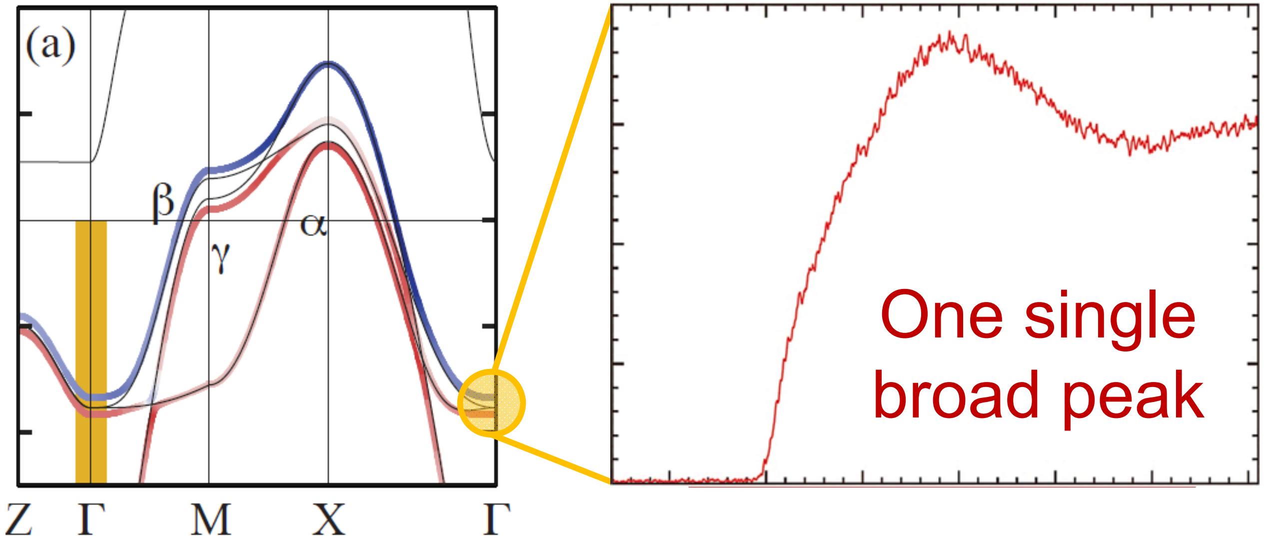


C.N. Veenstra et al. PRL **112**, 127002 (2014)

Direct measurement of SO: Spin-resolved ARPES

Band structure +
spin-orbit coupling

Spin-integrated
ARPES at Γ

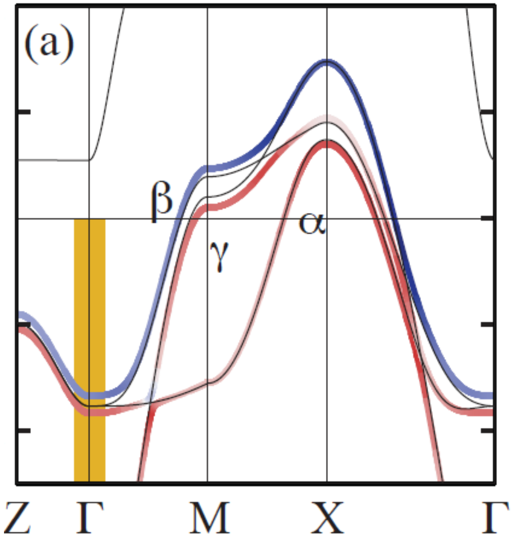


One single
broad peak

C.N. Veenstra et al. PRL **112**, 127002 (2014)

Direct measurement of SO: Spin-resolved ARPES

Band structure + spin-orbit coupling

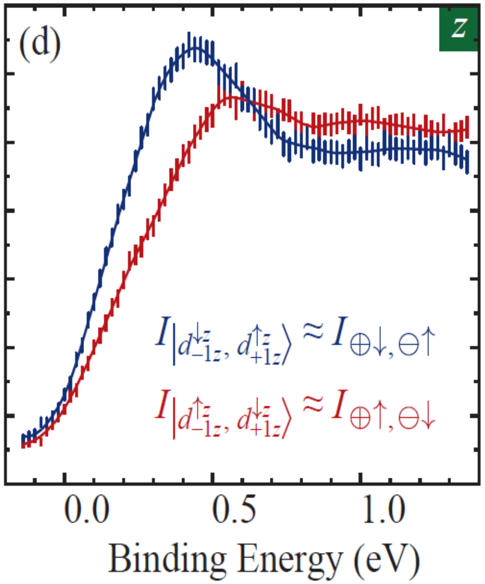


New SO-split eigenstates

$$\left| d^{\downarrow z}_{-1_z}, d^{\uparrow z}_{+1_z} \right\rangle$$

$$\left| d^{\uparrow z}_{-1_z}, d^{\downarrow z}_{+1_z} \right\rangle$$

Circular polarization Spin-resolved ARPES



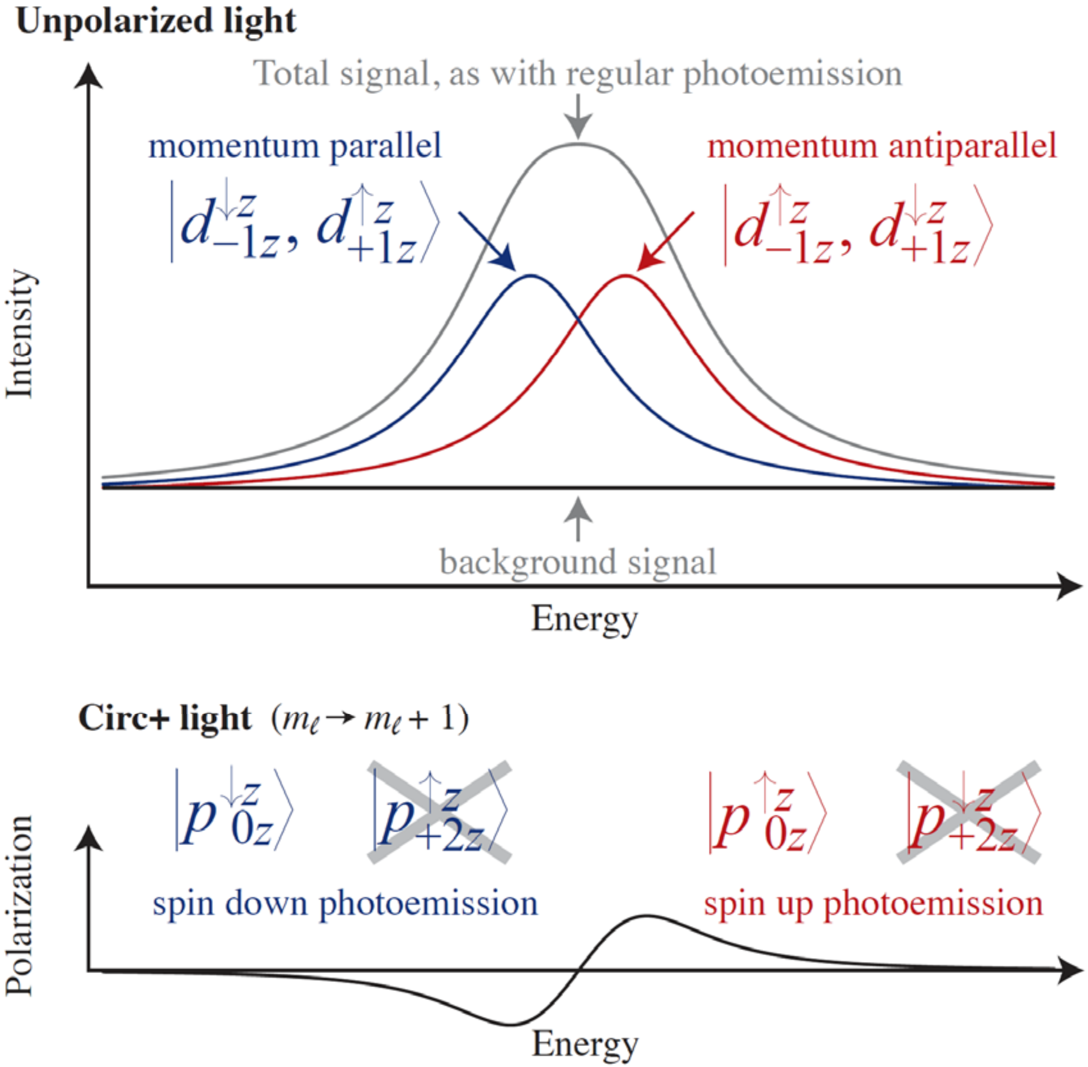
Spin-orbit splitting 130±30 meV

Spin-ARPES confirms SO entanglement

C.N. Veenstra et al. PRL **112**, 127002 (2014)

Direct measurement of SO: Spin-resolved ARPES

Spin-polarized photoemission using selection rules



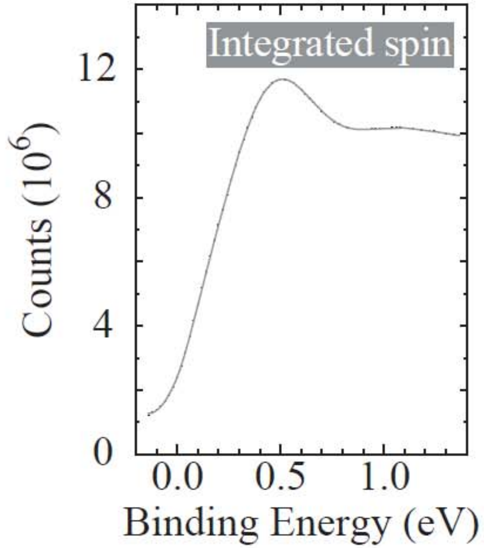
C.N. Veenstra et al. PRL **112**, 127002 (2014)

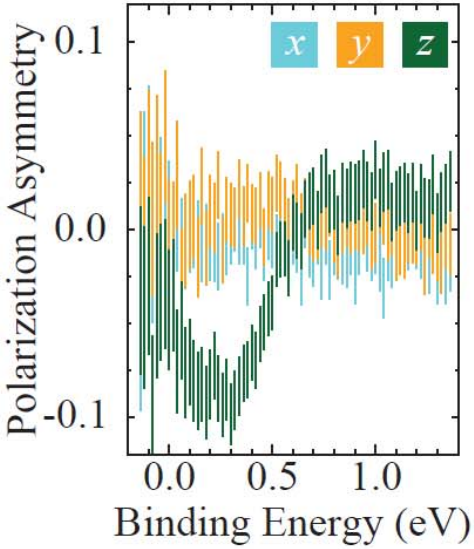
Direct measurement of SO: Spin-resolved ARPES

Confirming with spin-ARPES

- Initial states **not spin polarized**
- Repeat experiment with both helicities
- Measure a clear polarization asymmetry

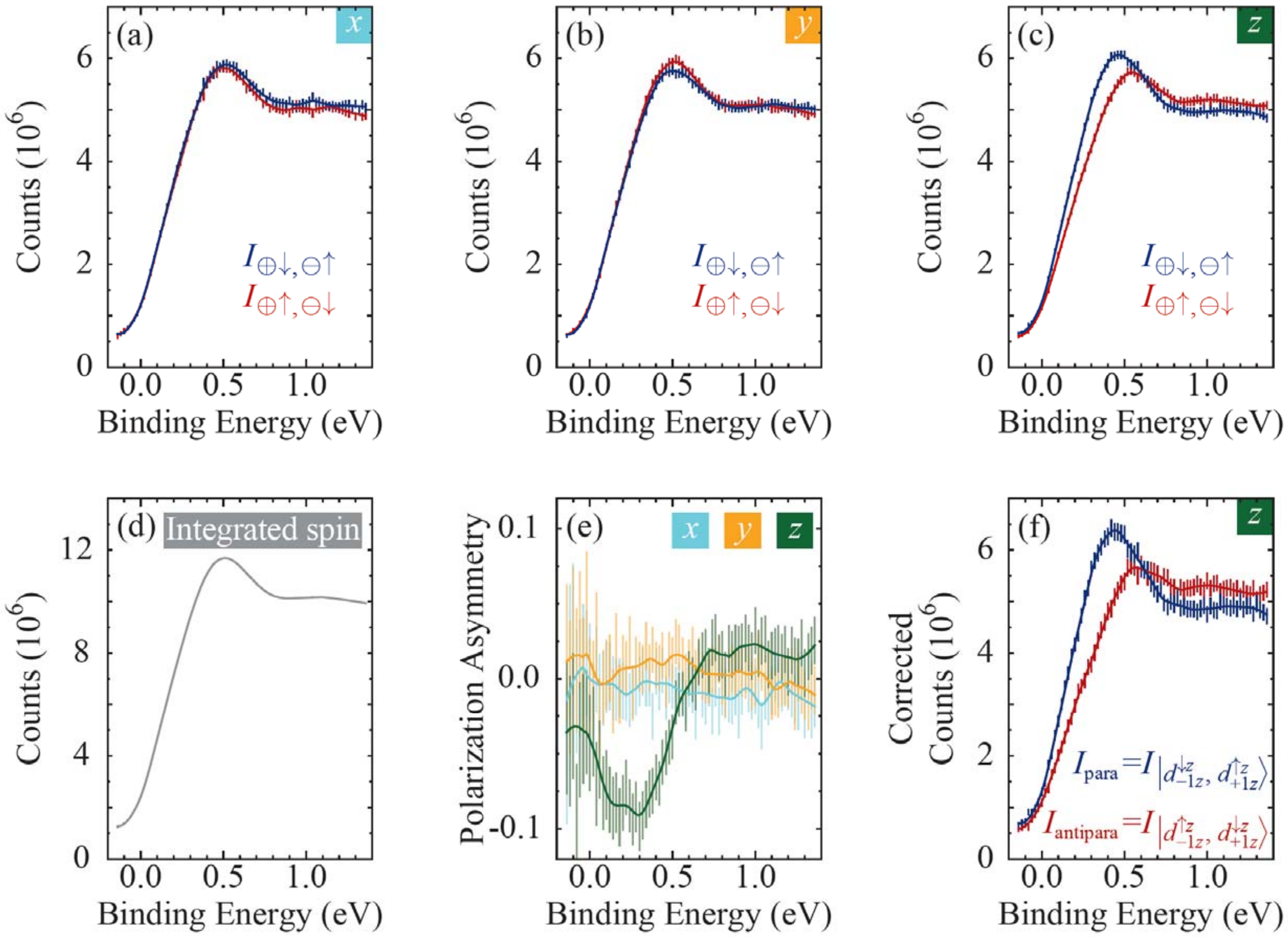
$$P^{\otimes} S_{\text{Motts}} = \frac{\sqrt{I_U^{\oplus} I_D^{\ominus}} - \sqrt{I_D^{\oplus} I_U^{\ominus}}}{\sqrt{I_U^{\oplus} I_D^{\ominus}} + \sqrt{I_D^{\oplus} I_U^{\ominus}}}$$





C.N. Veenstra et al. PRL **112**, 127002 (2014)

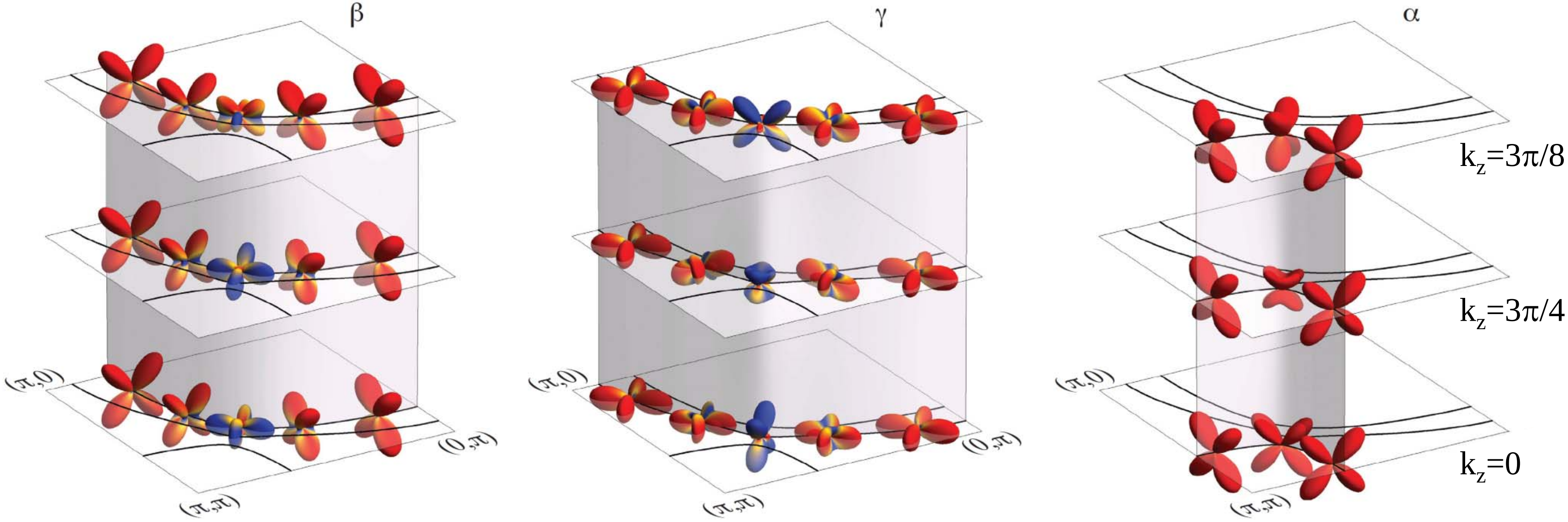
Direct measurement of SO: Spin-resolved ARPES



C.N. Veenstra et al. PRL **112**, 127002 (2014)

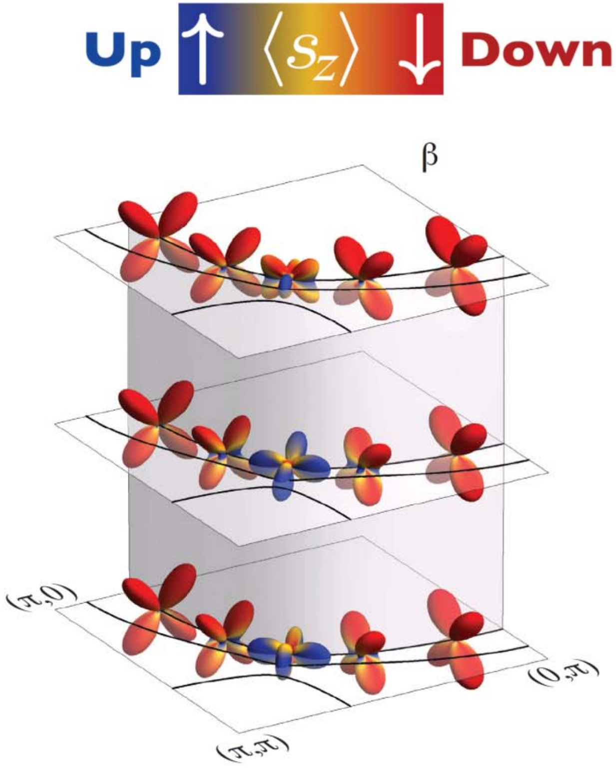
Spin-Orbit Coupling: K-dependent Entanglement

K-dependent entanglement of orbital and spin quantum numbers



C.N. Veenstra et al. PRL **112**, 127002 (2014)

Spin-Orbit Coupling: K-dependent Entanglement



K-dependent entanglement of orbital and spin quantum numbers

$$\xcancel{\psi(\mathbf{k},\sigma) = \varphi(\mathbf{k})\,\phi_{\sigma}^{\mathrm{spin}}}$$

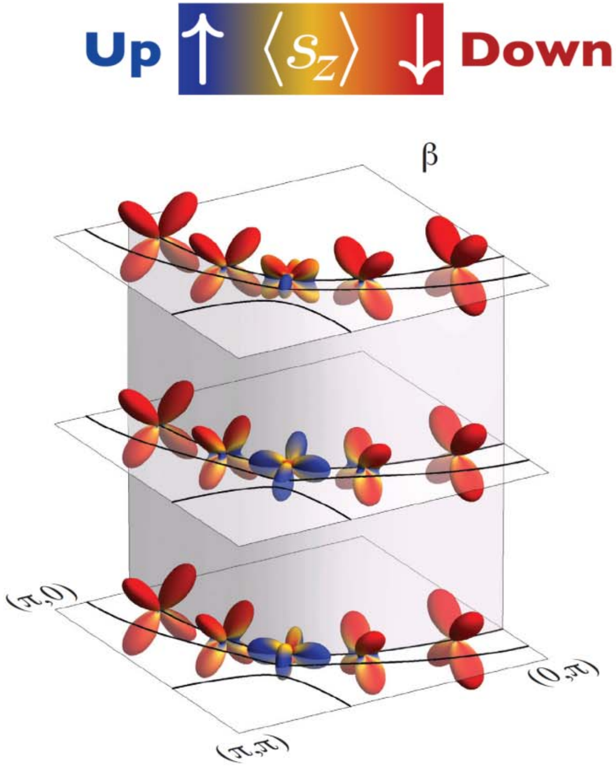
$$\boxed{\psi(\mathbf{k},\sigma) = \alpha\varphi_{\uparrow}(\mathbf{k})\phi_{\uparrow}^{\mathrm{spin}} + \beta\varphi_{\downarrow}(\mathbf{k})\phi_{\downarrow}^{\mathrm{spin}}}$$

What is the fate of spin-triplet superconductivity?

? $\Psi(\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2) = \varphi(\mathbf{r}_1 - \mathbf{r}_2)\phi_{\sigma_1,\sigma_2}^{\mathrm{spin}}$?

C.N. Veenstra et al. PRL **112**, 127002 (2014)

Spin-Orbit Coupling: K-dependent Entanglement



K-dependent entanglement of orbital and spin quantum numbers

$$\xcancel{\psi(\mathbf{k},\sigma) = \varphi(\mathbf{k})\,\phi_{\sigma}^{\text{spin}}}$$

$$\boxed{\psi(\mathbf{k},\sigma) = \alpha\varphi_{\uparrow}(\mathbf{k})\phi_{\uparrow}^{\text{spin}} + \beta\varphi_{\downarrow}(\mathbf{k})\phi_{\downarrow}^{\text{spin}}}$$

Breakdown of singlet-triplet Cooper pairs!

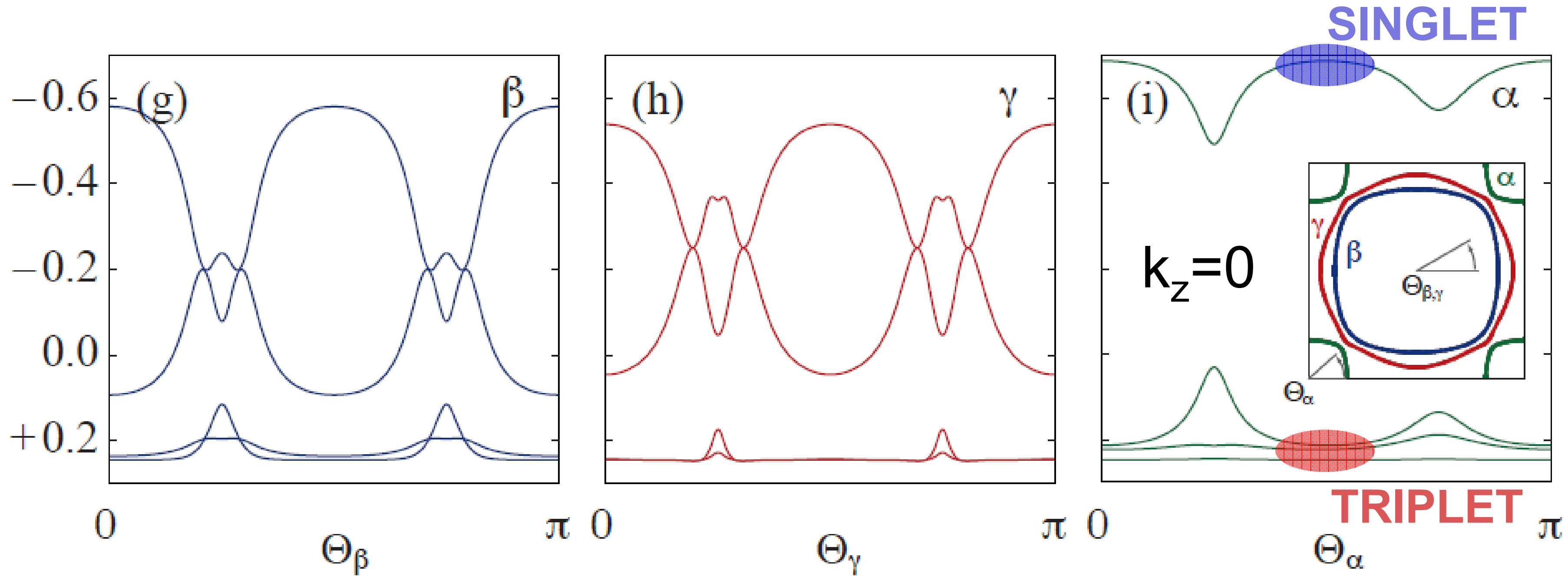
? $\xcancel{\Psi(\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2) = \varphi(\mathbf{r}_1-\mathbf{r}_2)\phi_{\sigma_1,\sigma_2}^{\text{spin}}}$?

C.N. Veenstra et al. PRL **112**, 127002 (2014)

Spin-Orbit Coupling: What is the Fate of Cooper Pairs?

$$\left\langle \vec{S}_k \cdot \vec{S}_{-k} \right\rangle$$

Spin-eigenstates for Cooper pairs

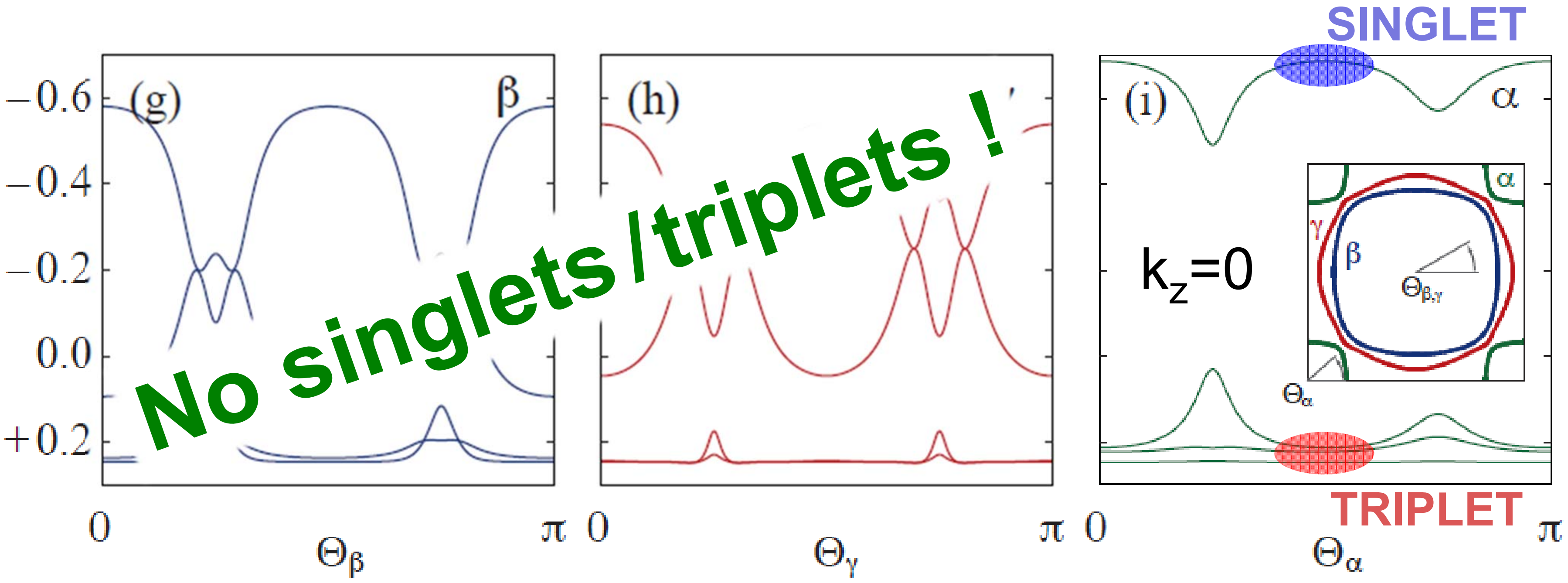


$$\xcancel{\Psi(\mathbf{r}_1, \sigma_1, \mathbf{r}_2, \sigma_2) = \varphi(\mathbf{r}_1 - \mathbf{r}_2)\phi^{\text{spin}}_{\sigma_1, \sigma_2}}$$

C.N. Veenstra et al. PRL **112**, 127002 (2014)

Spin-Orbit Coupling: What is the Fate of Cooper Pairs?

$\langle \vec{S}_k \cdot \vec{S}_{-k} \rangle$ Spin-eigenstates for Cooper pairs



SINGLET

No singlets/triplets !

TRIPLET

$$\xcancel{\Psi(\mathbf{r}_1,\sigma_1,\mathbf{r}_2,\sigma_2)=\varphi(\mathbf{r}_1-\mathbf{r}_2)\phi^{\text{spin}}_{\sigma_1,\sigma_2}}$$

C.N. Veenstra et al. PRL **112**, 127002 (2014)

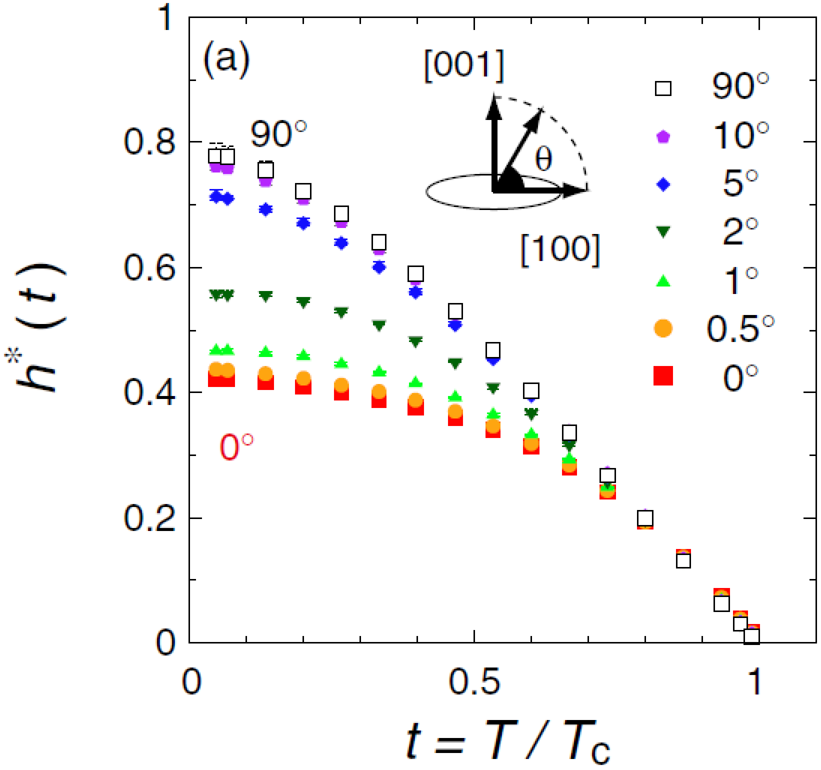
Sr_2RuO_4: Challenges to Spin-triplet Pairing

In plane suppression of H_{c2}

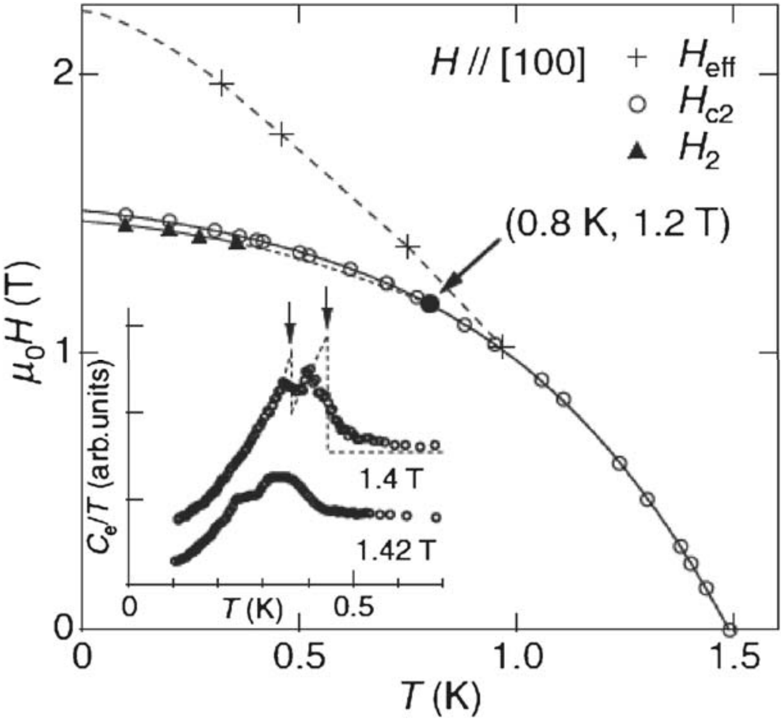
Strong for field angle θ<5°

Additional SC phase transition

Suppressed for field angle θ~1°



Kittaka et al., PRB (2009)
Maeno et al., JPSJ (2012)



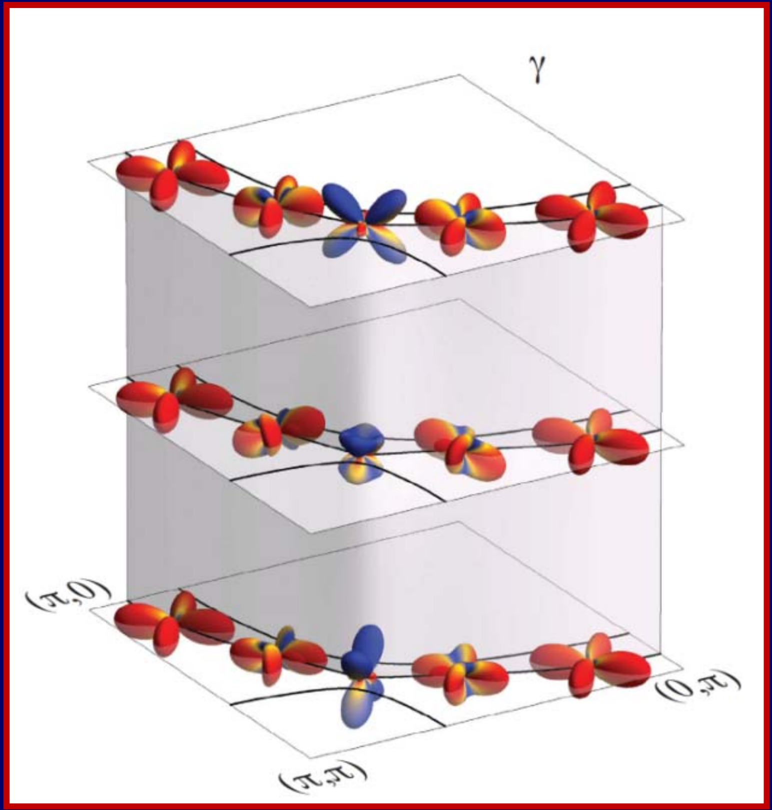
Deguchi et al., JPSJ (2002)

Magnetic anisotropy driven by spin-orbit coupling?

Direct measurement of SO: Spin-resolved ARPES

Spin-Orbital Entanglement and the Breakdown of Singlets and Triplets in Sr_2RuO_4 Revealed by Spin- and Angle-Resolved Photoemission Spectroscopy

C. N. Veenstra,[1] Z.-H. Zhu,[1] M. Raichle,[1] B. M. Ludbrook,[1] A. Nicolaou,[1,2,7] B. Slomski,[3,4] G. Landolt,[3,4] S. Kittaka,[5,6] Y. Maeno,[5] J. H. Dil,[3,4] I. S. Elfimov,[1,2] M. W. Haverkort,[1,2,7] and A. Damascelli[1,2,*]



$$\xcancel{\psi(\mathbf{k},\sigma) = \varphi(\mathbf{k})\;\phi^{\text{spin}}_{\sigma}}$$

$$\xcancel{\Psi = \varphi(\mathbf{r}_1 - \mathbf{r}_2)\phi^{\text{spin}}_{\sigma_1,\sigma_2}}$$

NO singlets and triplets
on most of Fermi surface

C.N. Veenstra et al. PRL **112**, 127002 (2014)