A Hidden Magnetic Phase in the Sr_2IrO_4 System Revealed Using Nonlinear Optical Measurements

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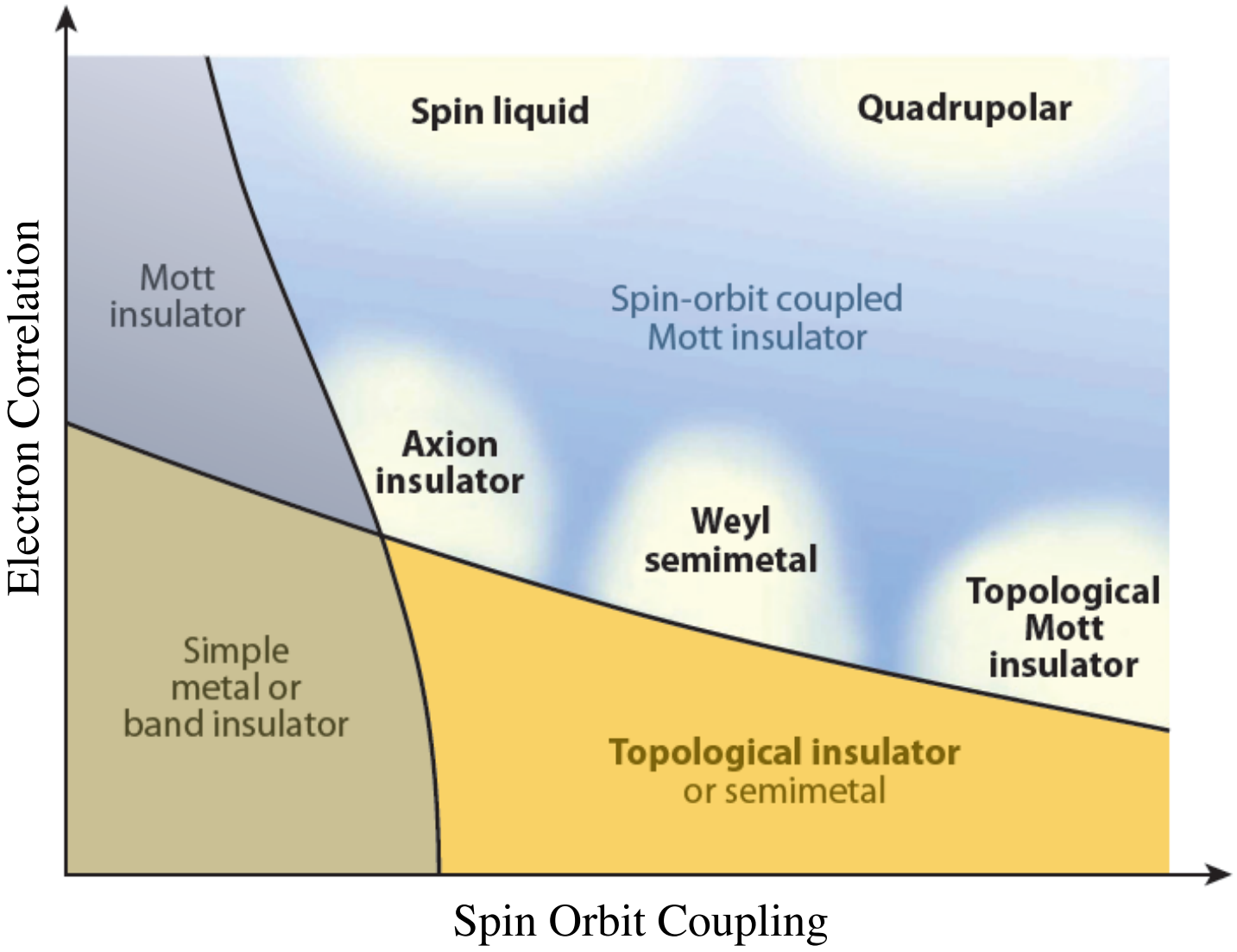
Prize Fellowship

Outline

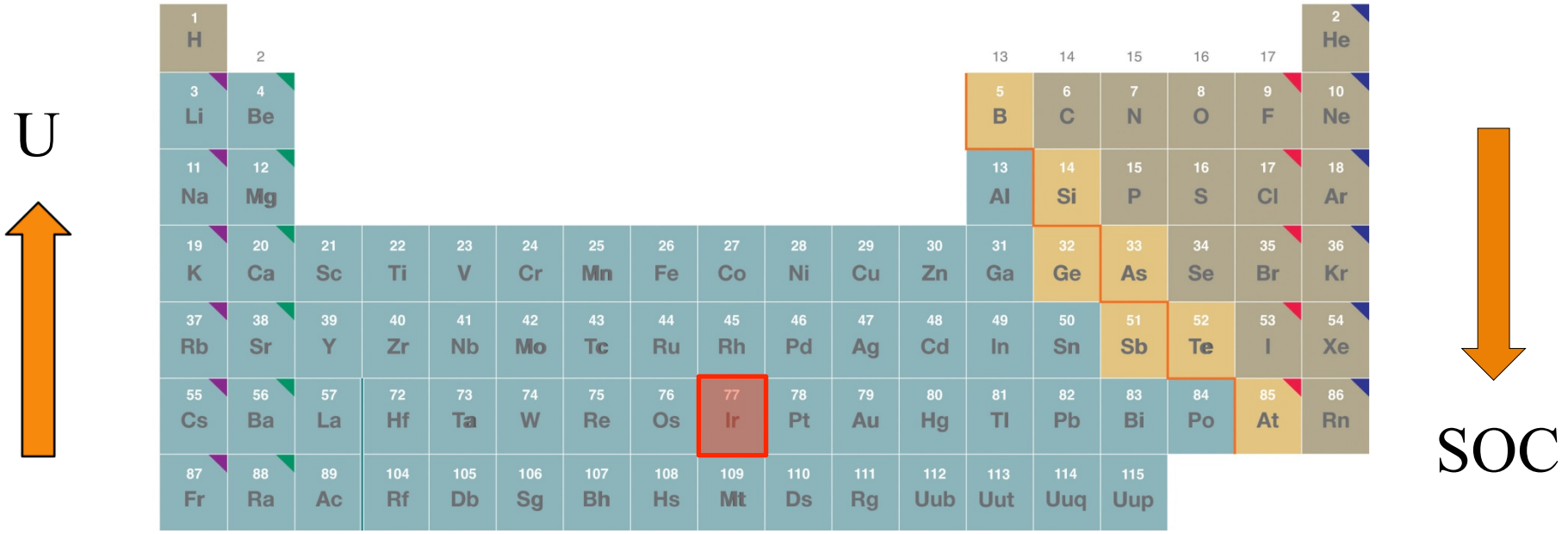
- Motivation of Searching for Symmetry Broken States in Sr_2IrO_4
- Nonlinear Optical Spectroscopy to Probe Symmetry Properties
- A Non-dipolar Magnetic Order Revealed in Sr_2IrO_4

Electron Correlation & Spin Orbit Coupling



W. Witczak-Krempa et al., Ann. Rev. Condens. Mat. Phys. 5, 57 (2014)

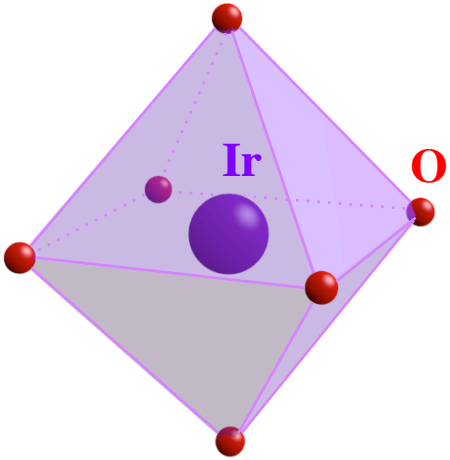
5d Transition Metal Oxides

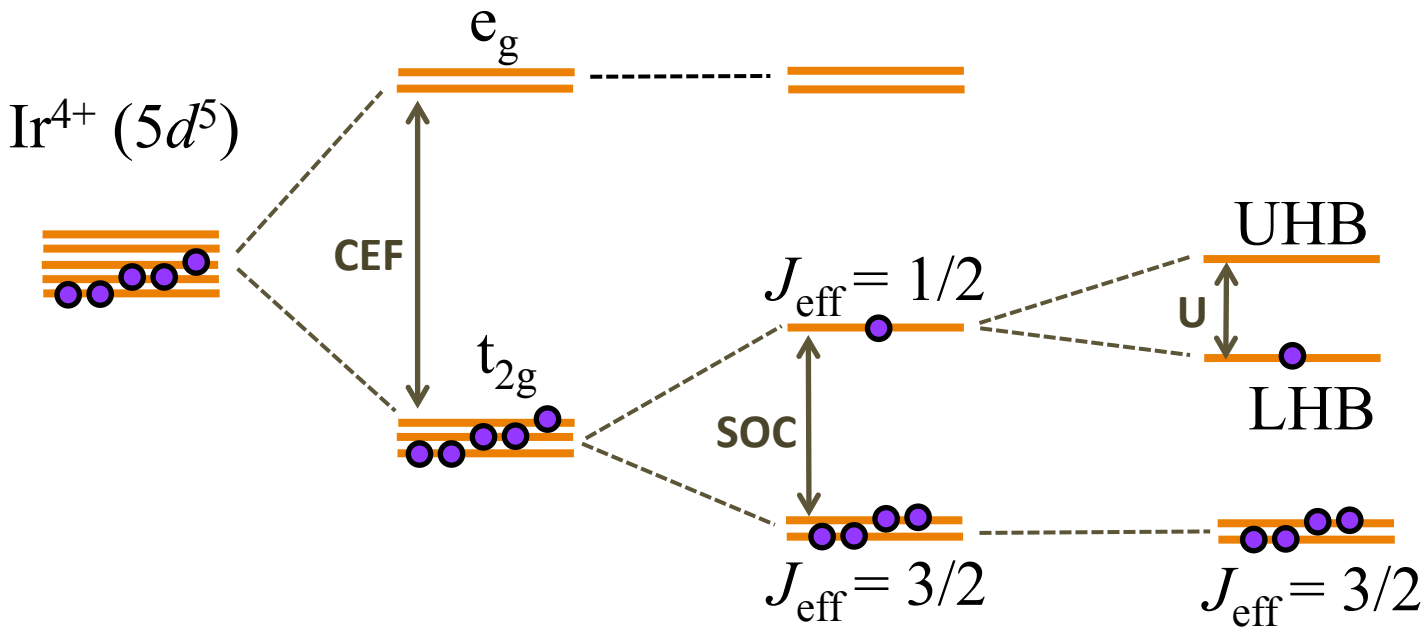


- Interplay among electron correlations, spin-orbit coupling and crystal electric field (U ~ SOC ~ CEF)
- Spin orbit coupled Mott insulator → exotic quantum phases

B.J. Kim *et al.* PRL **101**, 076402 (2012)
W. Witczak-Krempa et al., Ann. Rev. Condens. Mat. Phys. 5, 57 (2014)

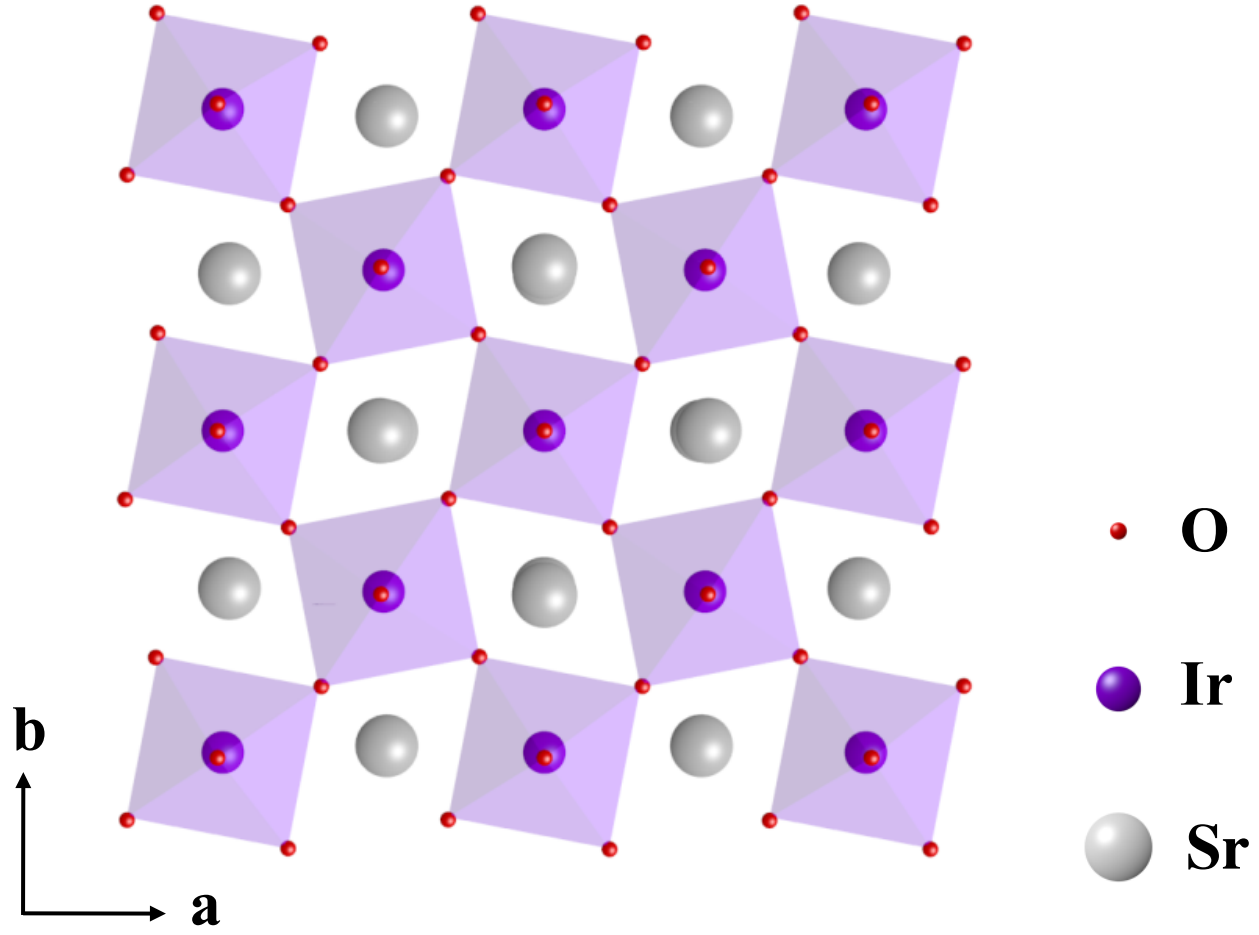
Iridates – J_{eff} = ½ Mott Insulator





B.J. Kim *et al.* PRL **101**, 076402 (2012)

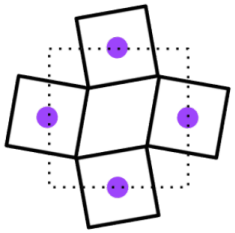
Sr_2IrO_4 – Single Layer Perovksite Structure



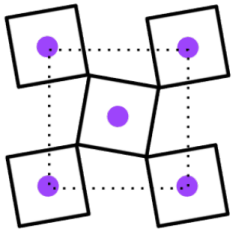
Sr_2IrO_4 – Single Layer Perovksite Structure

Sr_2IrO_4

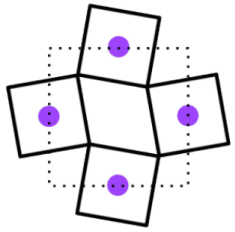
z = 7/8



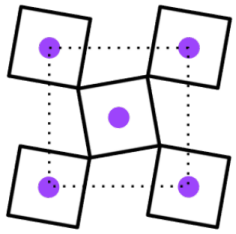
z = 5/8



z = 3/8



z = 1/8



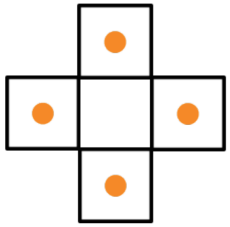
● Ir

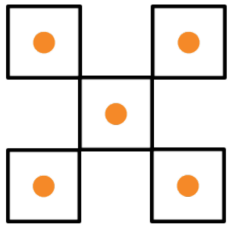
Crystal structure:

tetragonal point group 4/mmm

inversion symmetric

La_2CuO_4



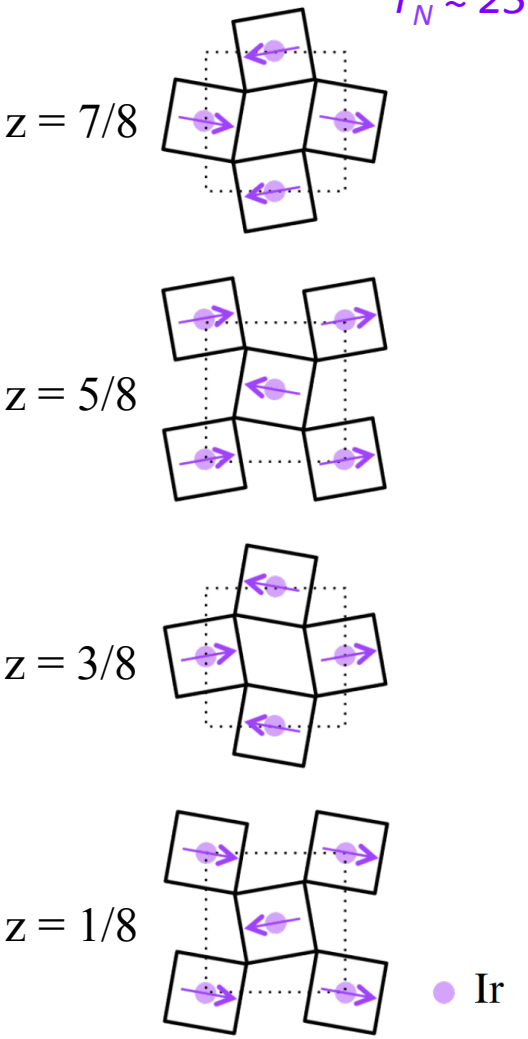


● Cu

Sr_2IrO_4 – Orthorhombic Magnetic (J_{eff} = ½) Structure

Sr_2IrO_4 J_{eff} = ½

$T_N \approx 230K$



Crystal structure:

tetragonal point group 4/mmm

inversion symmetric

Magnetic structure:

in-plane canted dipolar AFM

orthorhombic mmm1'

inversion symmetric

<u>Neutron diffraction</u>

Q. Huang *et al.*, J. Sol. State. Chem. 112, 355 (1994)

M. K. Crawford *et al.*, PRB 49, 9198 (1994)

F. Ye *et al.*, PRB 87, 140406(R) (2013)

C. Dhital *et al.*, PRB 87, 144405 (2013)

<u>Resonant x-ray diffraction</u>

B. J. Kim *et al.*, Science, 323, 1329 (2009)

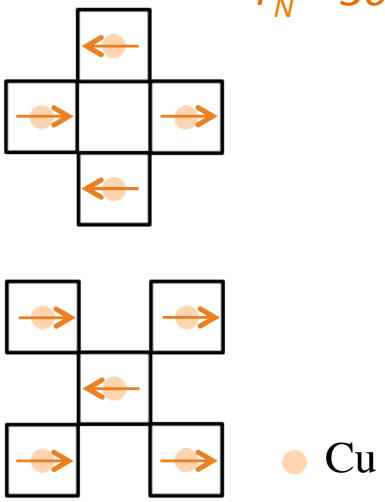
S. Boseggia *et al.*, J. Phys. CM 25, 422202 (2013)

S. Boseggia *et al.*, PRL 110, 117207 (2013)

M. Moretti Sala *et al.*, PRL 112, 026403 (2014)

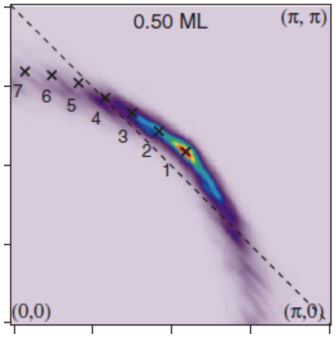
La_2CuO_4 S = ½

$T_N \approx 300K$

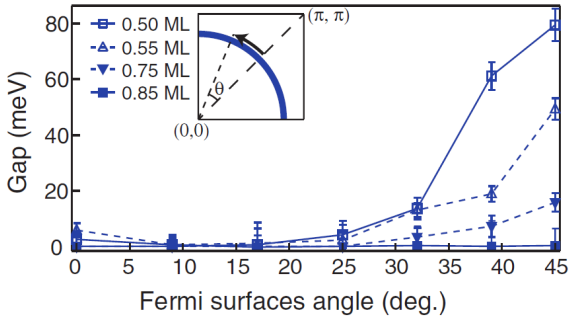


Doped Sr_2IrO_4 – Fermi Arcs (*Pseudogap* Behavior)

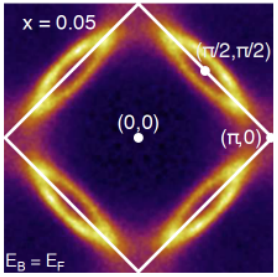
Surface K doped Sr_2IrO_4 (electron doping)

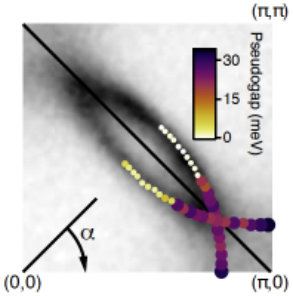


Y. K. Kim *et al.*, Science 345, 187 (2014)

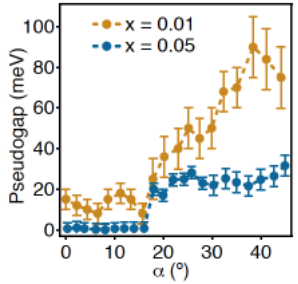


$(Sr_{1-x}La_x)_2IrO_4$ (electron doping)

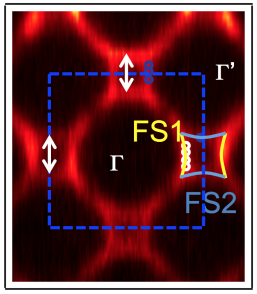




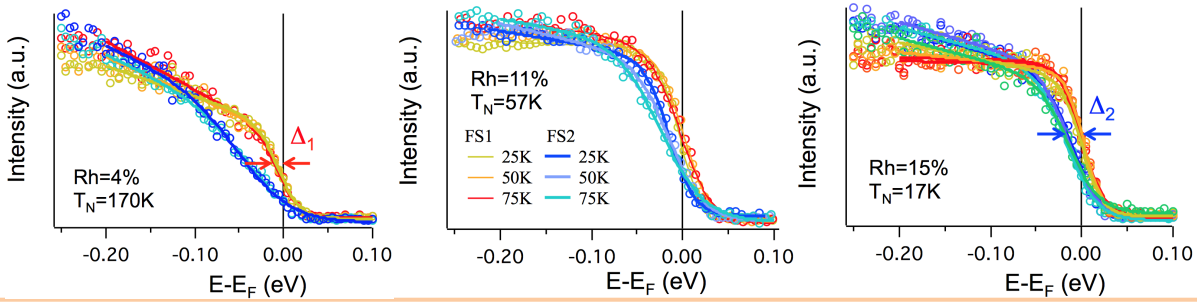
A. De la Torre *et al.*, http://arxiv.org/1506.00616 (2015)



$Sr_2Ir_{1-x}Rh_xO_4$ (hole doping)



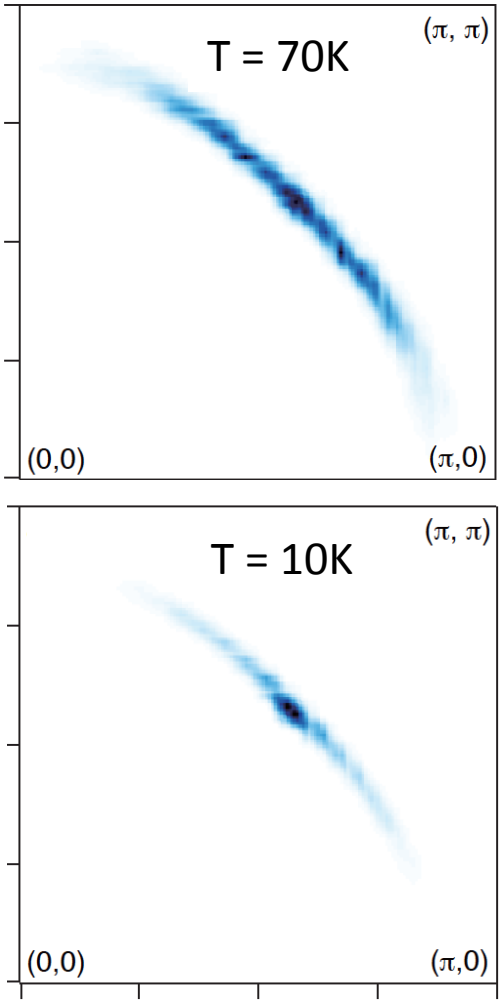
Y. Cao *et al.*, http://arxiv.org/1406.4978 (2014)



Surface K Doped Sr_2IrO_4 – d-wave Gap

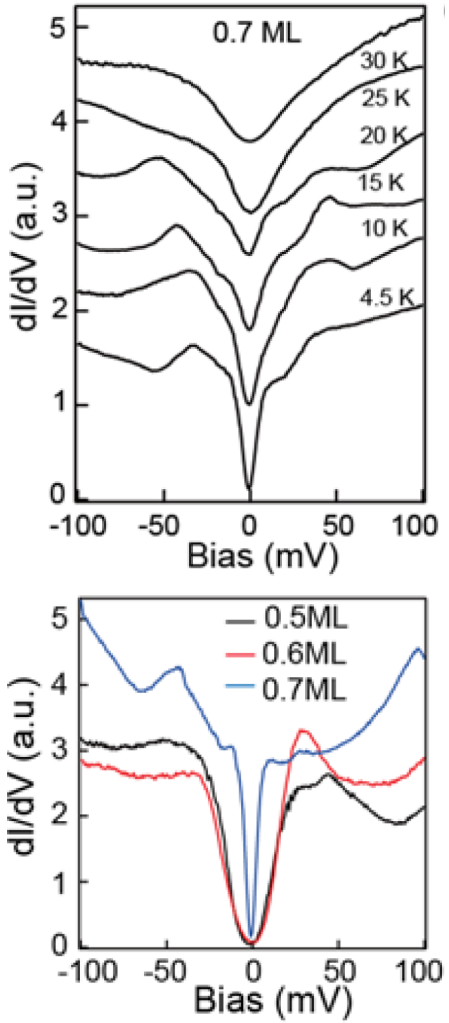
ARPES results

Y. K. Kim *et al.,* Nature Physics DOI: 10.1038 (2015)

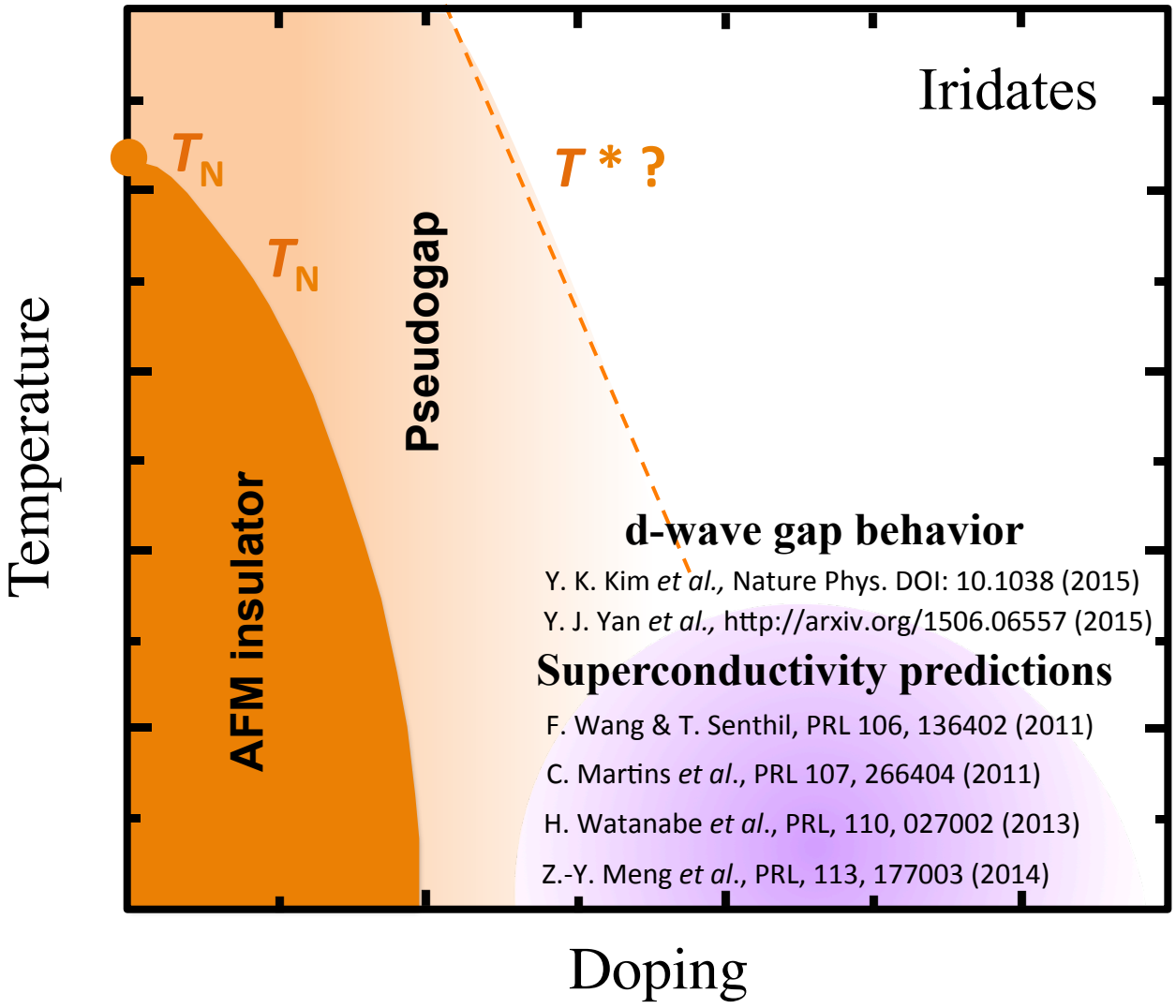


STM/S results

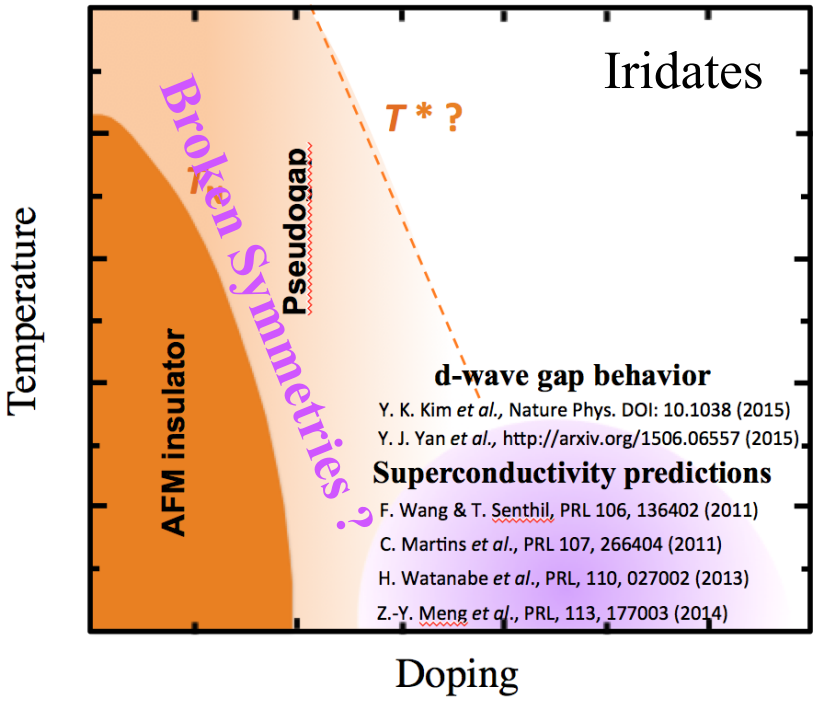
Y. J. Yan *et al.,* http://arxiv.org/1506.06557 (2015)

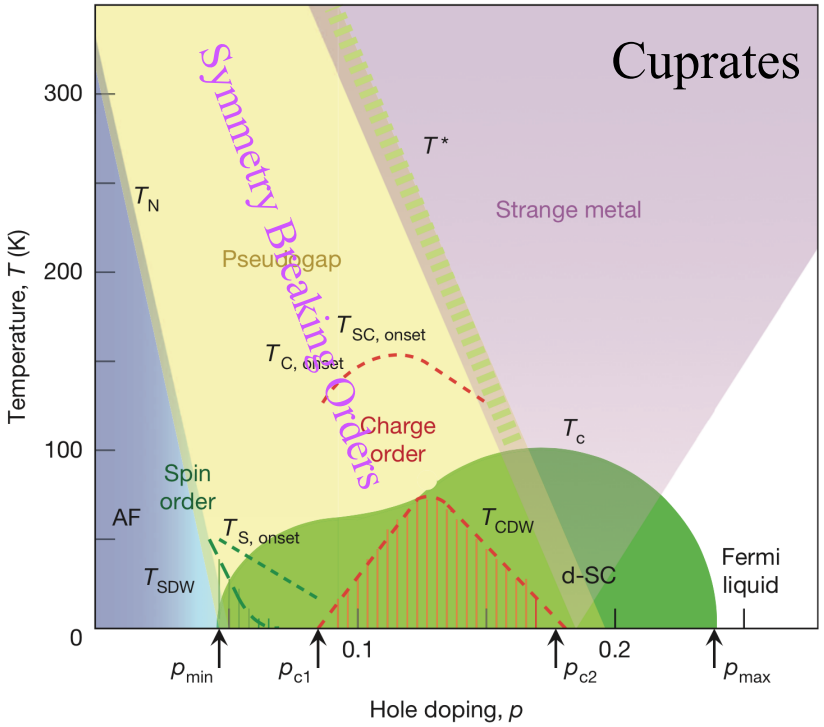


Constructing A Phase Diagram?



Analogy to Cuprates Phase Diagram





B. Keimer *et. al.* Nature, 518, 179 (2015)

Broken symmetries in Cuprates pseudogap:

A. Kaminski *et al.* Nature, 416, 610 (2002)
S. V. Borisenko *et al.* PRL, 92, 207001 (2004)
B. Fauqué *et al.* PRL, 96, 197001 (2006)
H. A. Mook *et al.* PRB, 78, 020506 (R) (2008)
J. Xia *et al.* PRL, 100, 127002 (2008)
Y. Lubashevsky *et al.* PRL, 112, 147001 (2014)
M. J. Lawler *et al.* Nature, 466, 347
K. Fujita *et al.* PNAS, 2015 112 (8) 2367
……

Nonlinear Optics

- Multipole expansion of radiation source term

Electric Dipole Magnetic Dipole Electric Quadruple

$$\vec{S} \propto \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2} + \mu_0 \left(\vec{\nabla} \times \frac{\partial \vec{M}}{\partial t} \right) - \mu_0 \left(\vec{\nabla} \frac{\partial^2 \hat{Q}}{\partial t^2} \right) + \ldots$$

M. Fiebig *et al.* J. O. S. A. (B) **22**, 96118 (2005)

- Expansion of electric dipolar (**P**) contribution

$$P_i(\omega, 2\omega, 3\omega \ldots) = \underbrace{\chi_{ij}^{pe} E_j(\omega)}_{1^{st}\text{ order}} + \underbrace{\chi_{ijk}^{pee} E_j(\omega) E_k(\omega)}_{2^{nd}\text{ order}} + \underbrace{\chi_{ijkl}^{peee} E_j(\omega) E_k(\omega) E_l(\omega)}_{3^{rd}\text{ order}} + \ldots$$

  - χ_{ij}^{pe}, χ_{ijk}^{pee} and χ_{ijkl}^{peee} $\longleftrightarrow$ Properties Tensors

Property Tensor & Symmetry Properties

- ***Neumann's principle*** : A property tensor of a crystal is invariant under the symmetry operation of the crystal.
- Symmetry group → independent nonzero tensor elements
- Higher rank response tensor → greater symmetry resolution

$$\chi_{ij}^{pe}$$

	# elem.
Tetragonal Trigonal Hexagonal	2

$$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$$

$$\chi_{ijk}^{pee}$$

$4 = C_4$	7
$\bar{4} = S_4$	6
$422 = D_4$	3
$4mm = C_{4v}$	4
$\bar{4}2m = D_{2d}$	3*
$4/m = C_{4h}$	0
$4/mmm = D_{4h}$	0

Second Harmonic Generation (SHG)

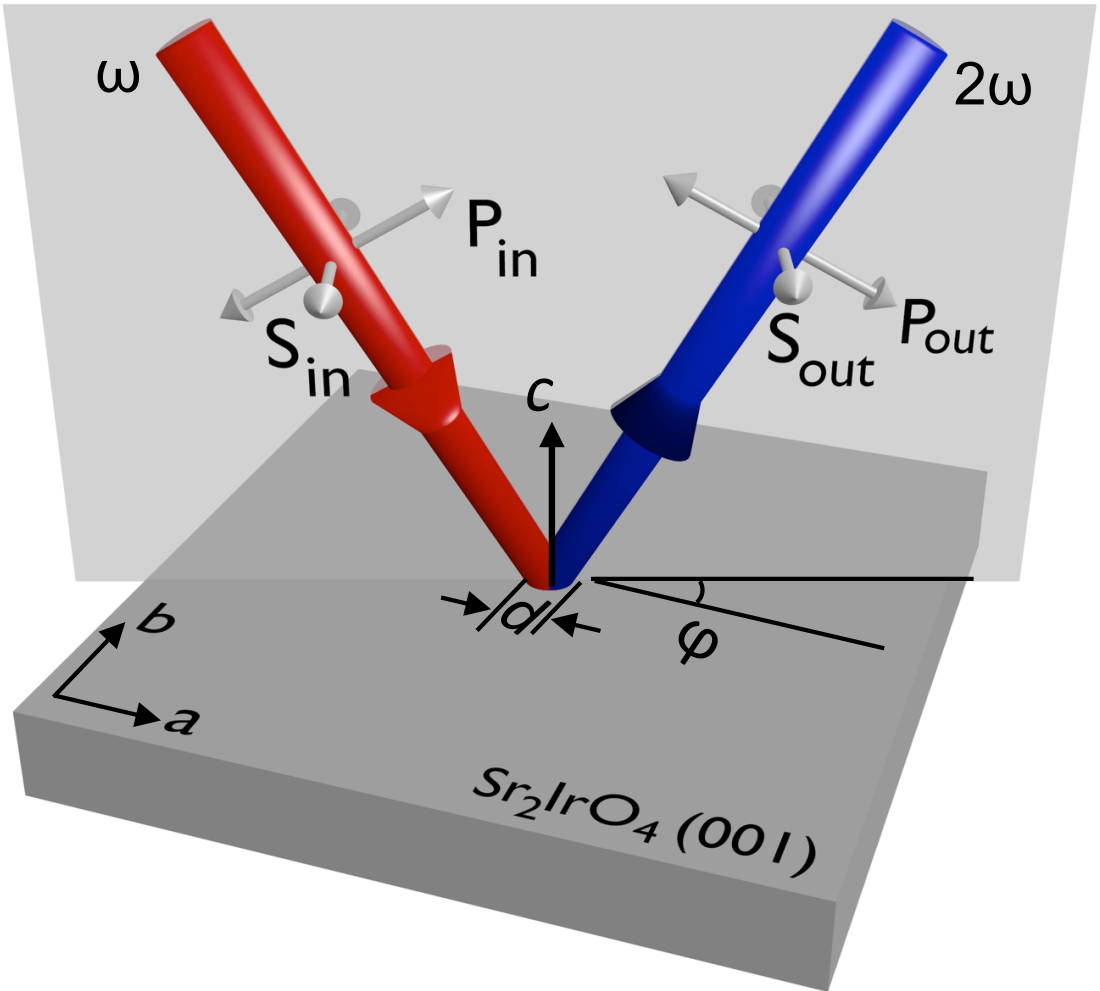
Inversion Symmetry: $(x, y, z) \rightarrow (-x, -y, -z)$

	χ^{pee}_{ijk}	χ^{pe}_{ij}
	$\mathbf{P}(2\omega) = \chi^{pee}\,\mathbf{E}(\omega)\mathbf{E}(\omega)$	$\mathbf{P}(\omega) = \chi^{pe}\,\mathbf{E}(\omega)$
I. operat.	$-\mathbf{P}(2\omega) = \mathbf{I}(\chi^{pee})\,(-\mathbf{E}(\omega))(-\mathbf{E}(\omega))$	$-\mathbf{P}(\omega) = \mathbf{I}(\chi^{pe})\,(-\mathbf{E}(\omega))$
	$-\mathbf{P}(2\omega) = \mathbf{I}(\chi^{pee})\,\mathbf{E}(\omega)\mathbf{E}(\omega)$	$\mathbf{P}(\omega) = \mathbf{I}(\chi^{pe})\,\mathbf{E}(\omega)$
I. symm.	$\mathbf{I}(\chi^{pee}) = \chi^{pee}$	$\mathbf{I}(\chi^{pe}) = \chi^{pe}$
	$-\mathbf{P}(2\omega) = \chi^{pee}\,\mathbf{E}(\omega)\mathbf{E}(\omega)$	$\mathbf{P}(\omega) = \chi^{pe}\,\mathbf{E}(\omega)$
$\rightarrow$	$\chi^{pee} = 0$	$\chi^{pe} \neq 0$

$$\vec{S} \propto \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2} + \mu_0 \left(\vec{\nabla} \times \frac{\partial \vec{M}}{\partial t} \right) - \mu_0 \left(\vec{\nabla} \frac{\partial^2 \hat{Q}}{\partial t^2} \right) + \ldots$$

Rotation Anisotropy SHG (RA-SHG)

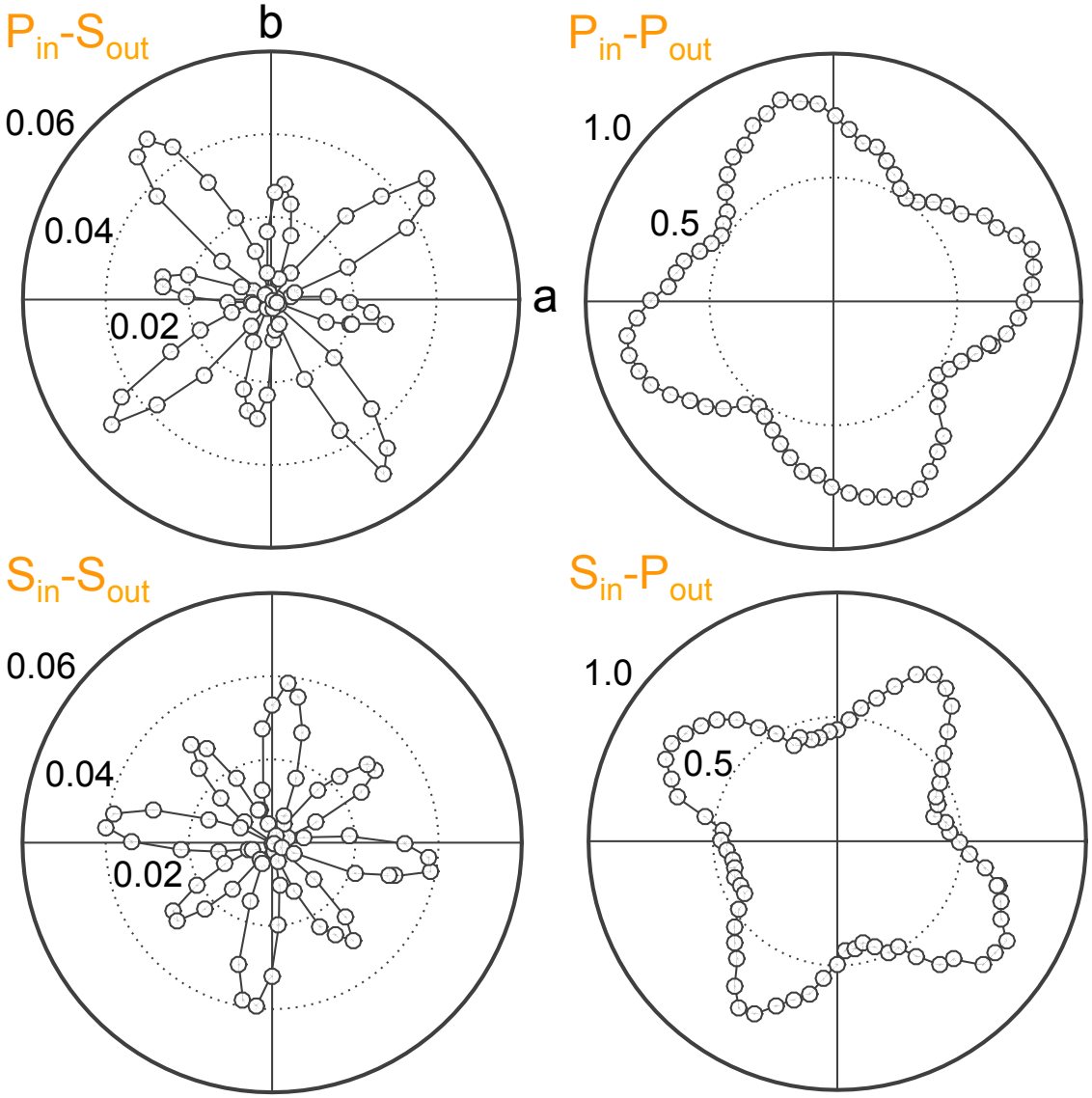
$I^{2\omega}(\varphi, T, r)$



- Four polarization geometries $P/S_{in} - P/S_{out}$
- φ dependence RA-SHG
- *T* dependence
- ***r*** dependence SHG imaging

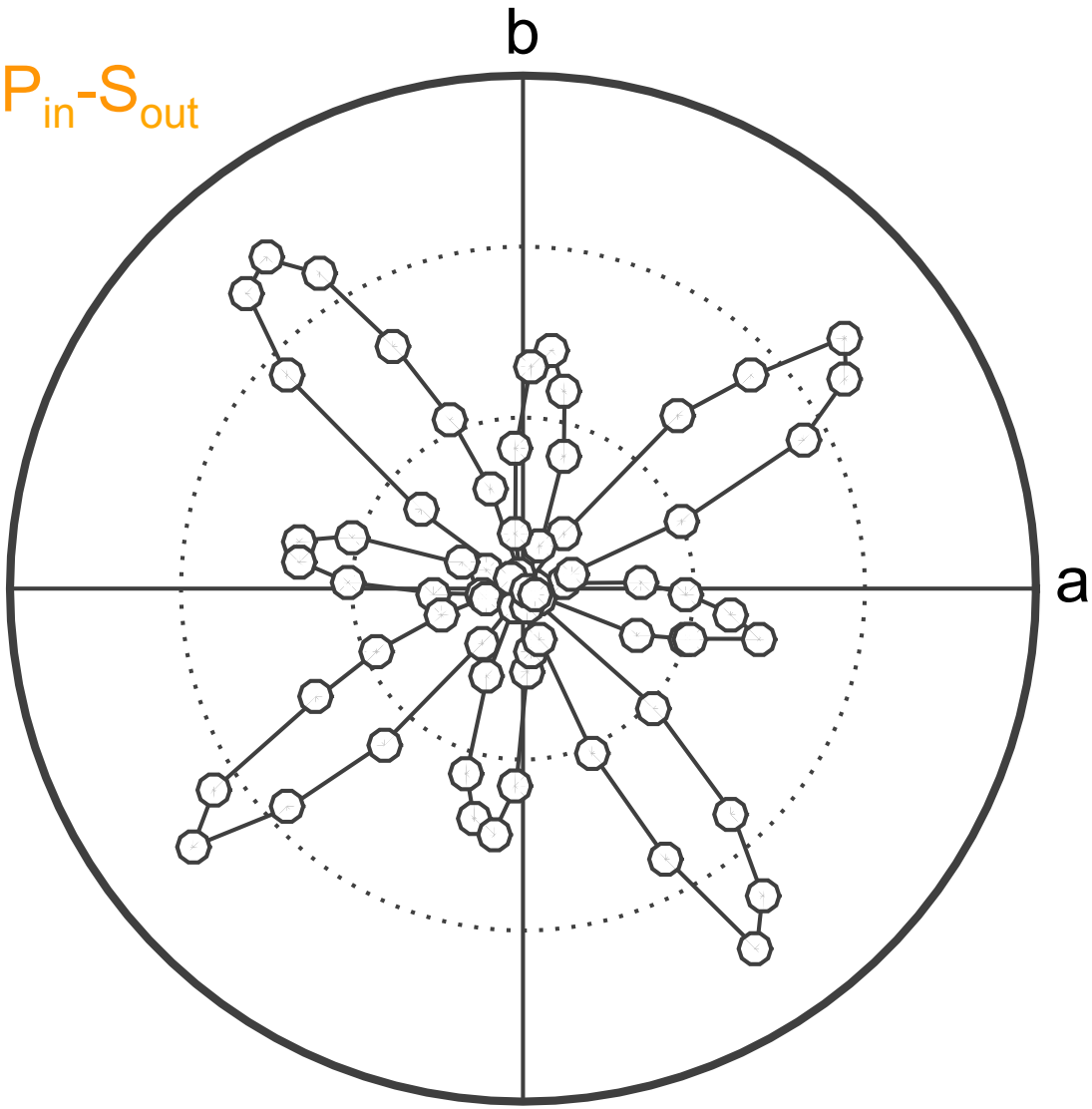
D. Torchinsky *et al.* Rev. Sci. Inst. 85, 083102 (2014)

RA – SHG on Sr_2IrO_4 at 295K



L. Zhao *et al.* to appear in Nature Physics (2015)
D. Torchinsky, H. Chu, L. Zhao *et al.* PRL, 114, 096404 (2015)

RA – SHG on Sr_2IrO_4 at 295K

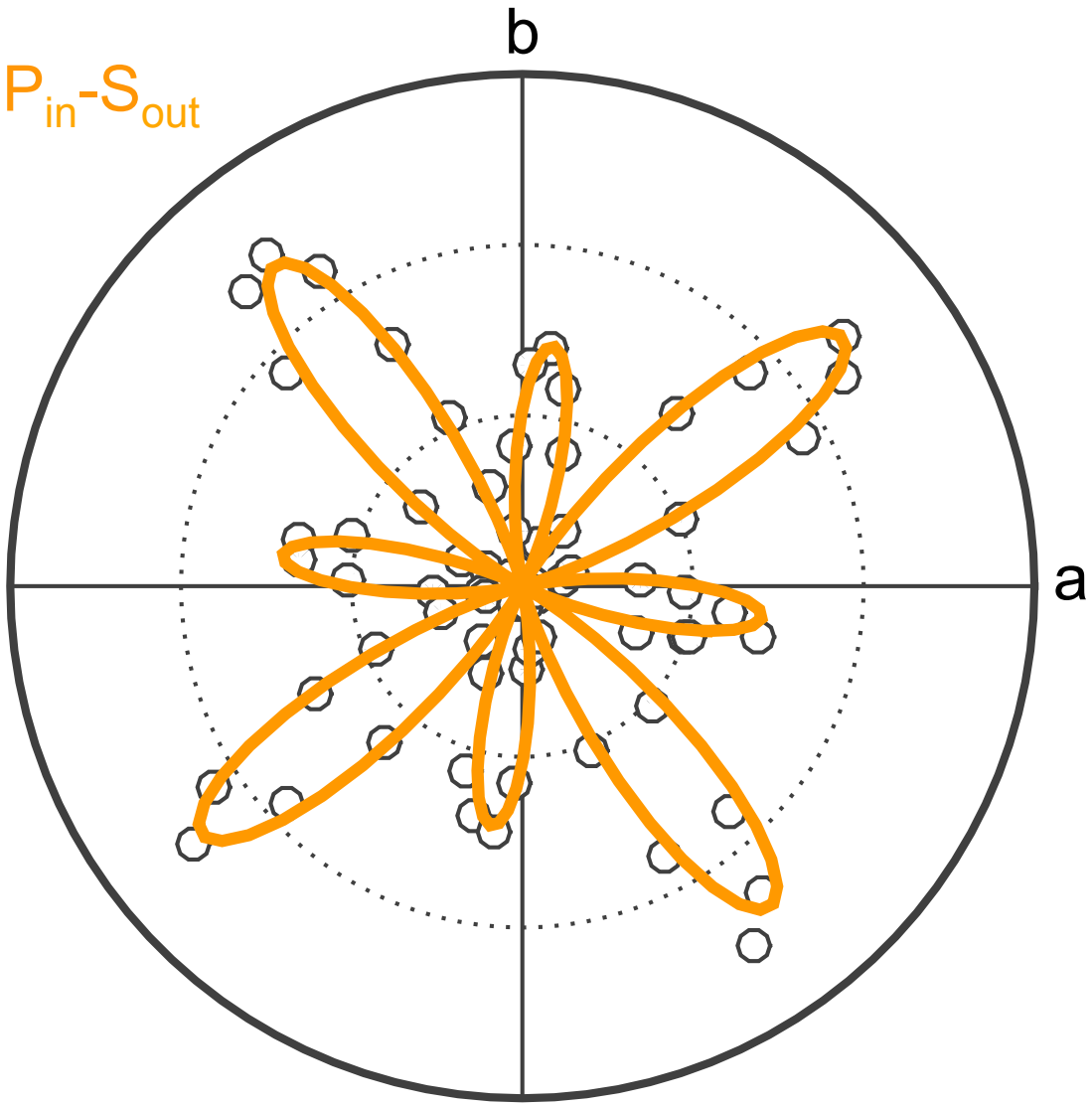


- C_4 rotational symmetry
- Rotated ~12° off **a** axis
  Broken *ac*, *bc* mirrors
- Refined structure ***4/m***
  Inversion symmetric
- Bulk electric quadruple

$$Q = \chi^{qee}\, E\, E$$

$$P_{eff} = \chi^{qee}\, E\, \nabla E$$

RA – SHG on Sr_2IrO_4 at 295K



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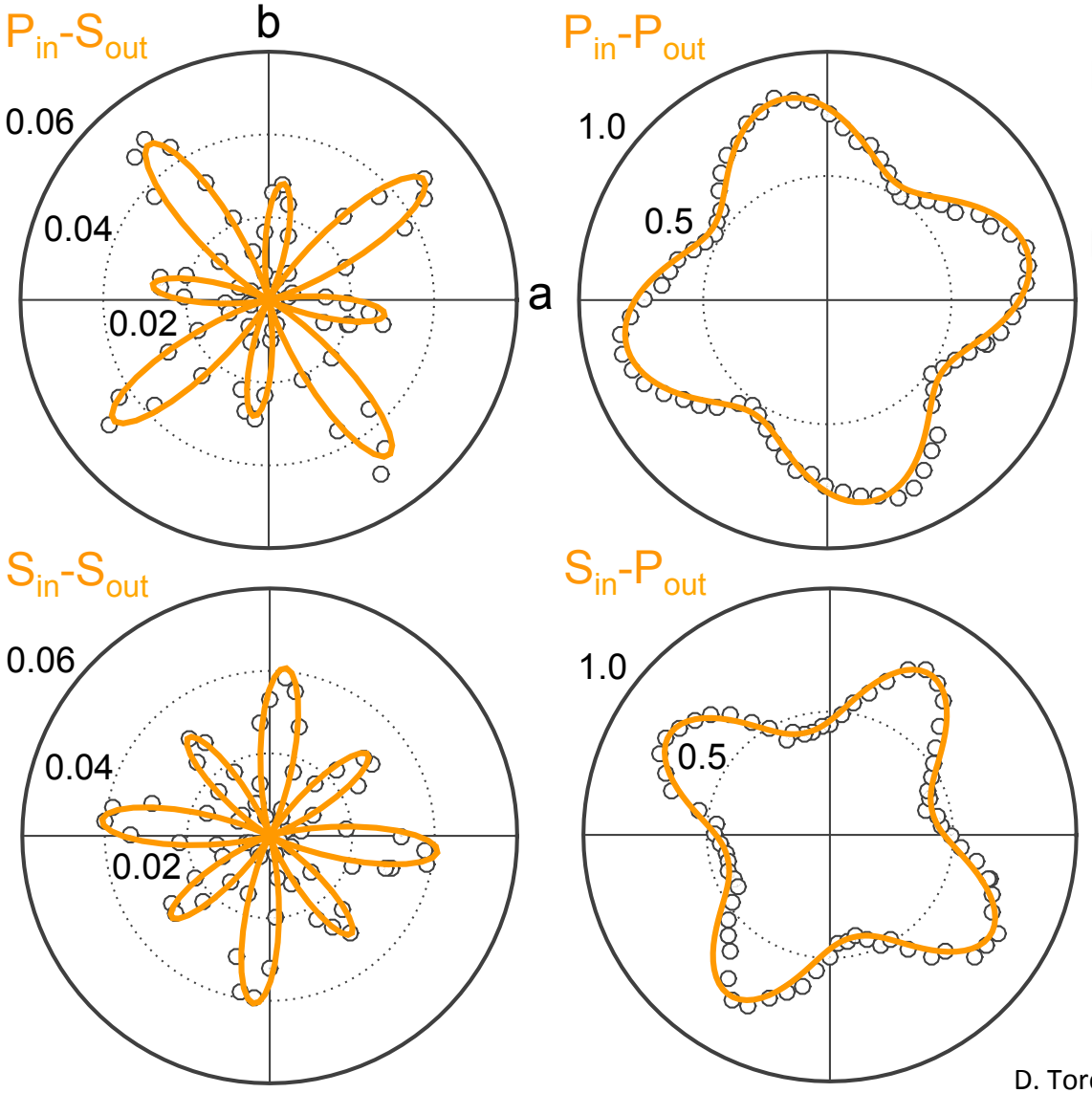
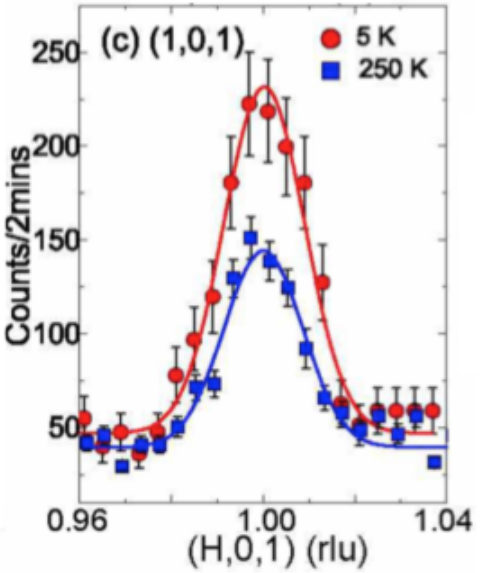
RA – SHG on Sr_2IrO_4 at 295K



- Bulk E. Q. contribution from ***4/m*** structure.
- Consistent with recent findings of forbidden neutron peaks if ***4/mmm***



C. Dhital *et al.*, PRB 87, 144405 (2013)

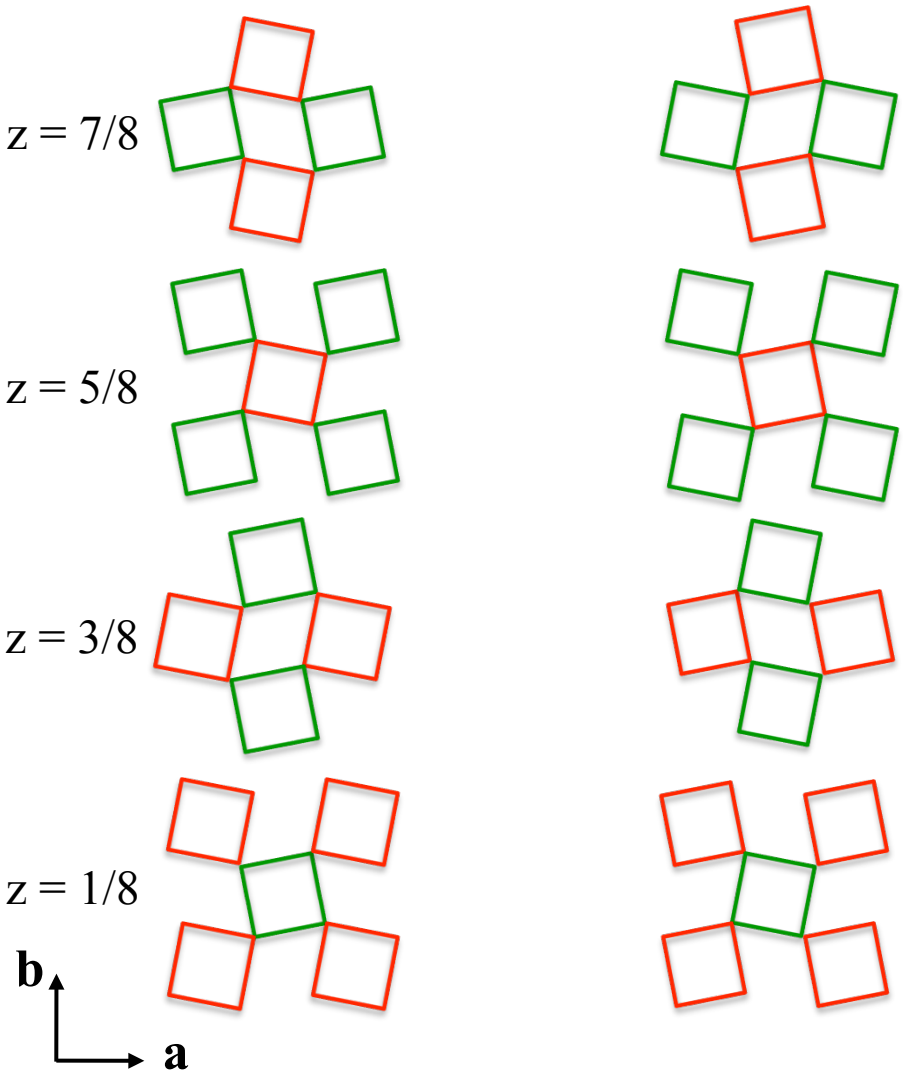
F. Ye *et al.*, PRB 87, 140406(R) (2013)

D. Torchinsky, H. Chu, L. Zhao *et al.* PRL, 114, 096404 (2015)

L. Zhao *et al.* to appear in Nature Physics (2015)

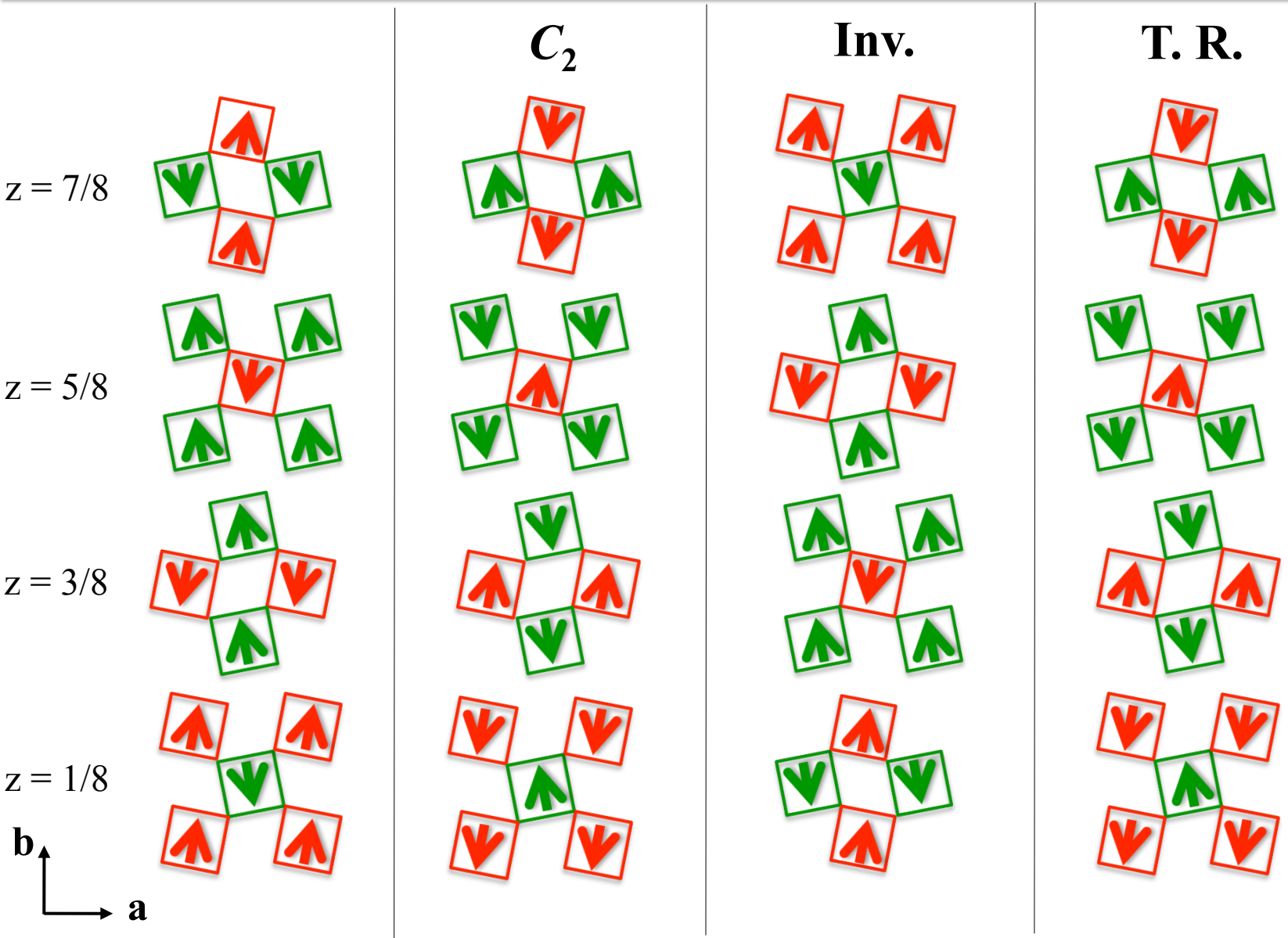
4/*m* Crystal Structure

ac, *bc* mirrors

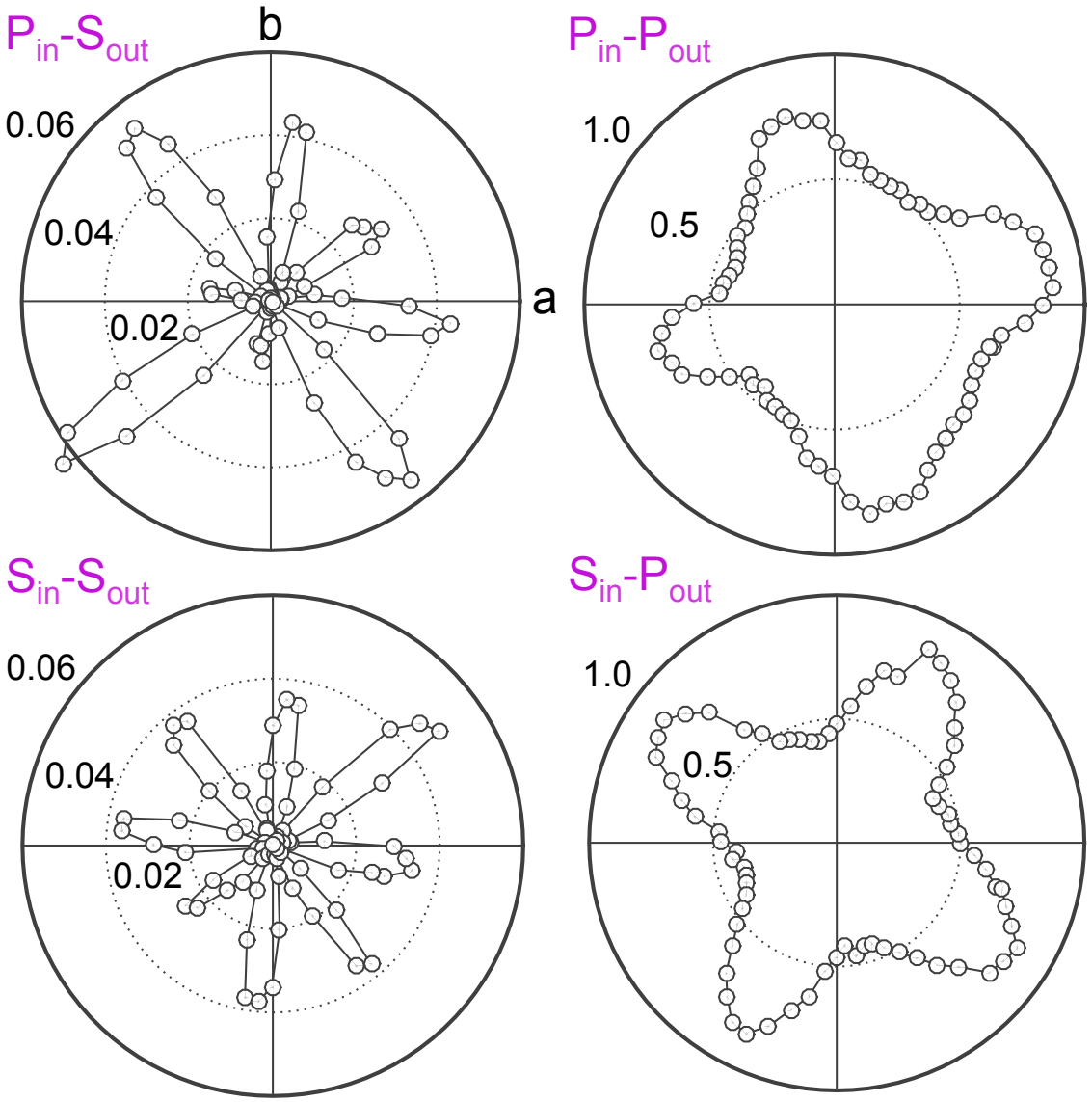


D. Torchinsky, H. Chu, L. Zhao *et al.* PRL, 114, 096404 (2015)

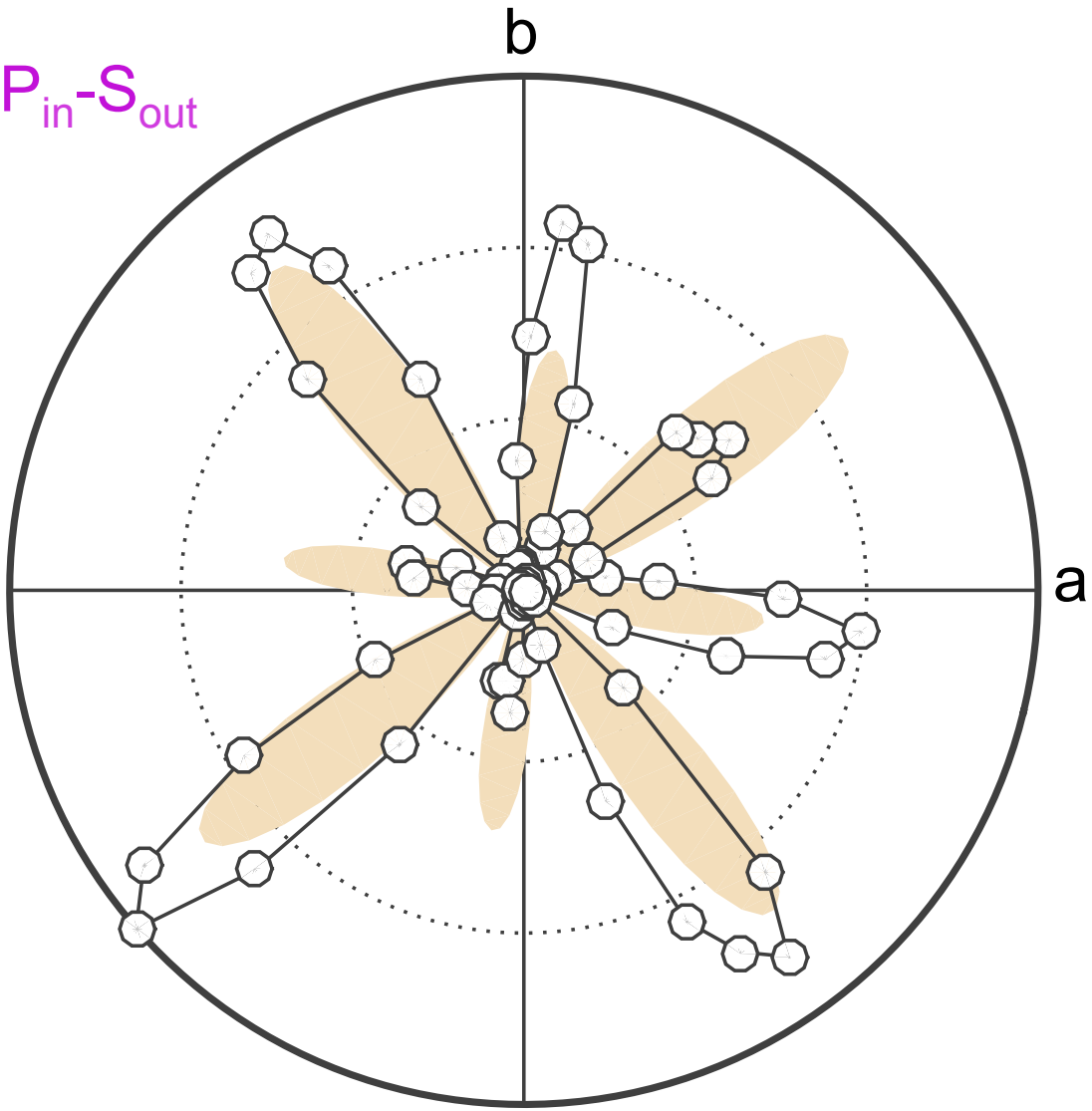
2/*m*1' AFM Structure



RA – SHG on Sr_2IrO_4 at 175K



RA – SHG on Sr_2IrO_4 at 175K



- C_1 rotational symmetry
- Structural contribution from E.Q. under ***4/m***

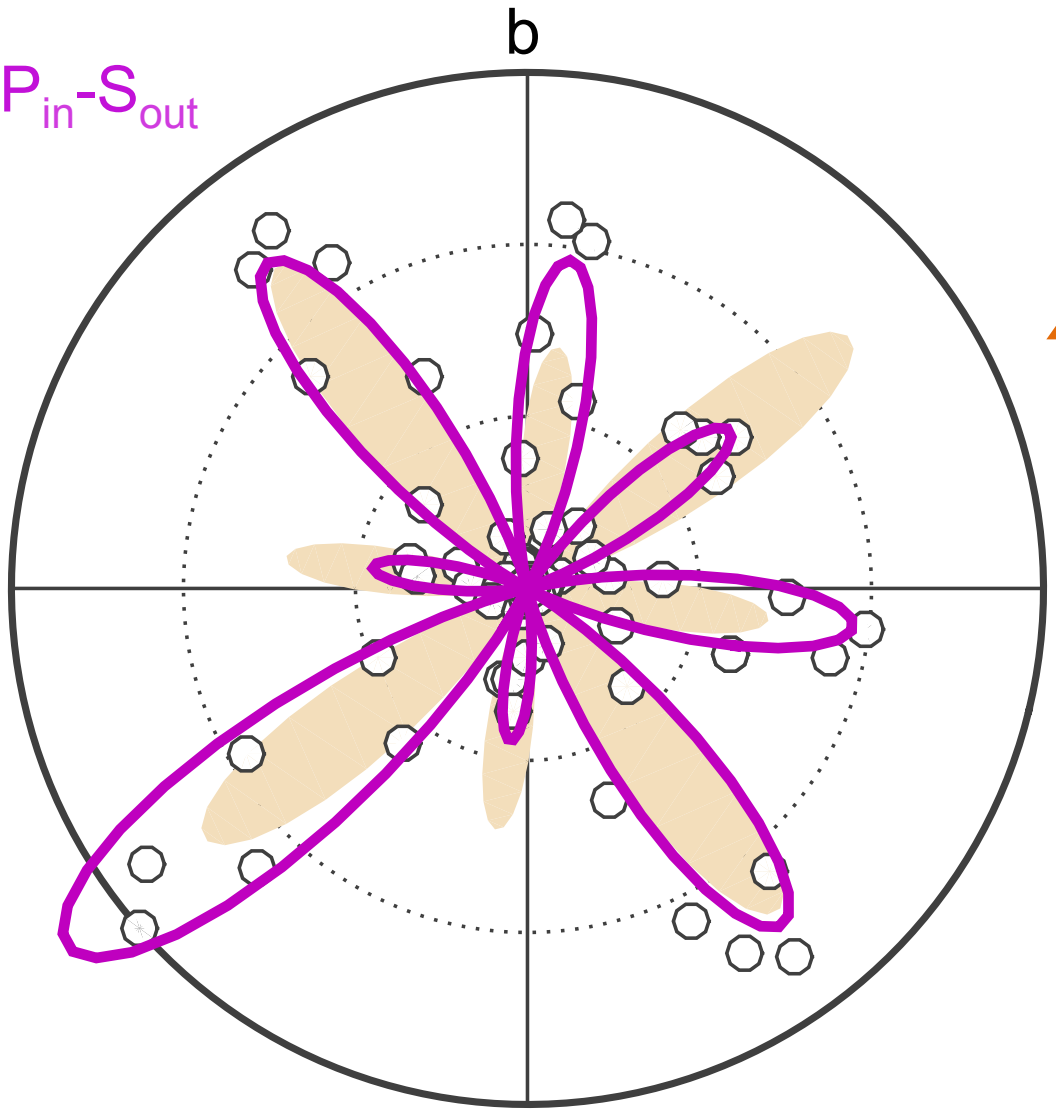
  &

  New contribution from E.D. under ***2'/m***

  Broken Inversion

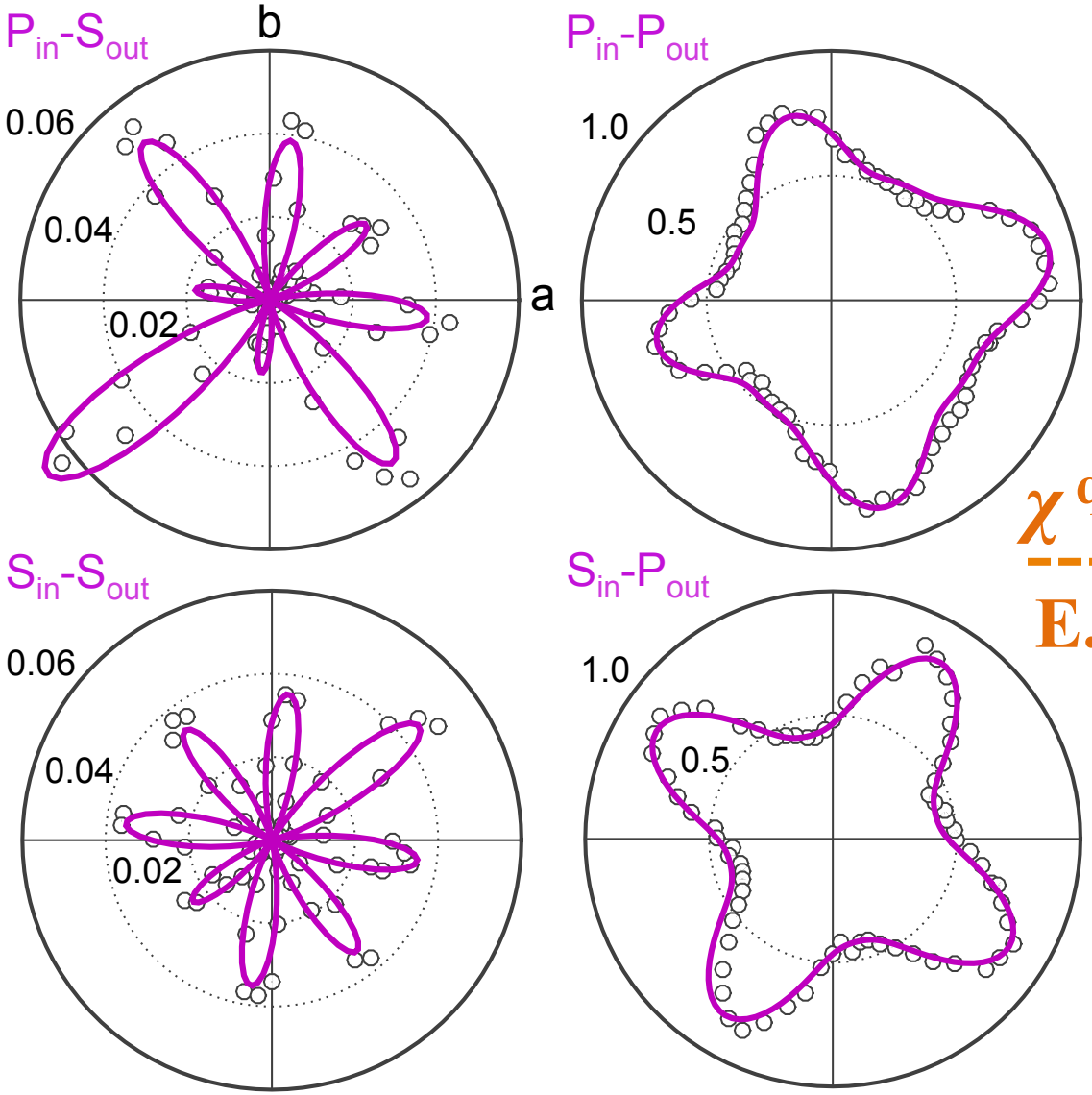
  Broken Time Reversal
- NOT AFM (***2/m*1'**)

RA – SHG on Sr_2IrO_4 at 175K



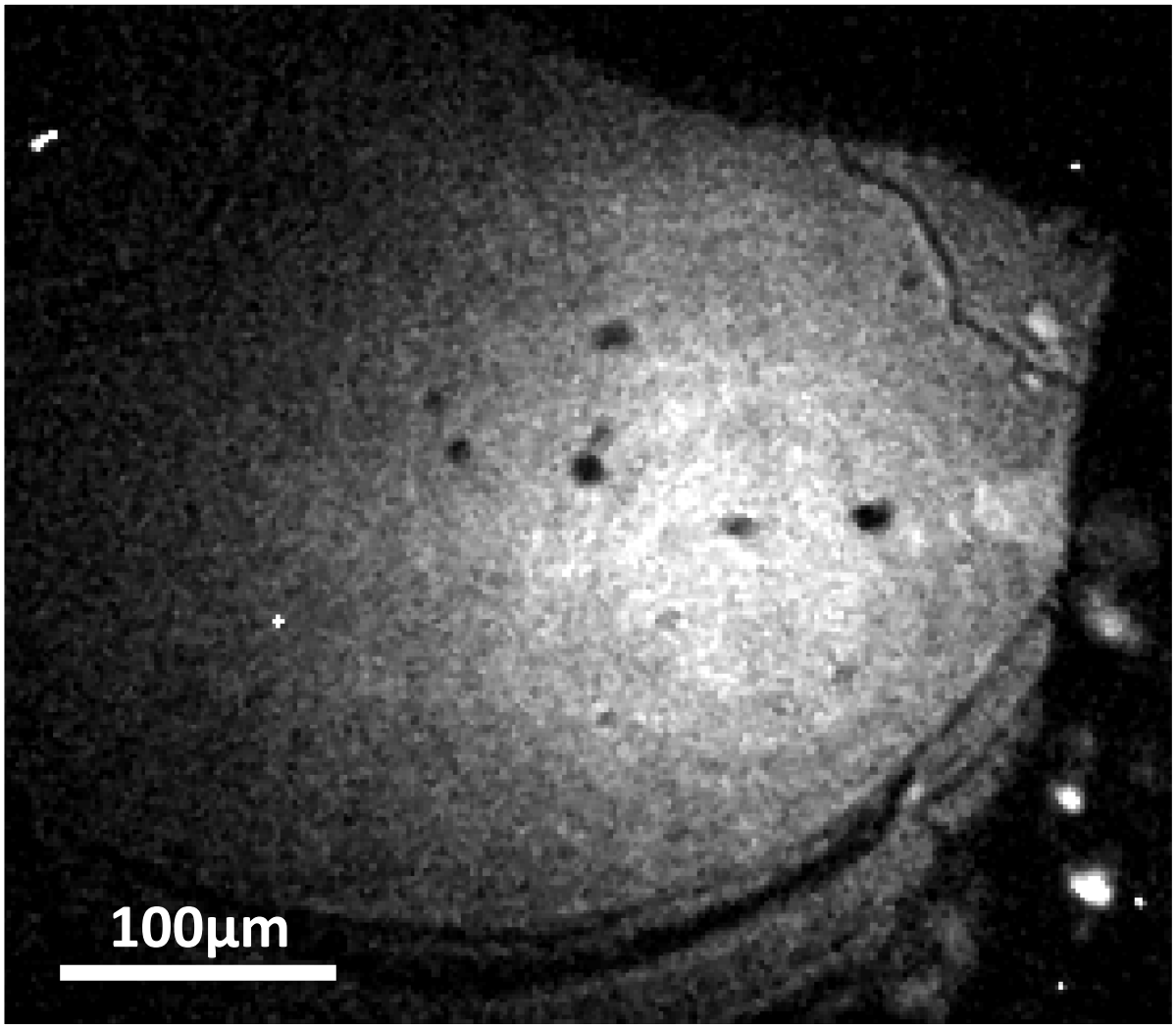
$$\frac{\chi^{qee}\,E\nabla E}{\text{E. Q. } (4/m)} + \frac{\chi^{pee}\,EE}{\text{E. D. } (2'/m)}$$

RA – SHG on Sr_2IrO_4 at 175K

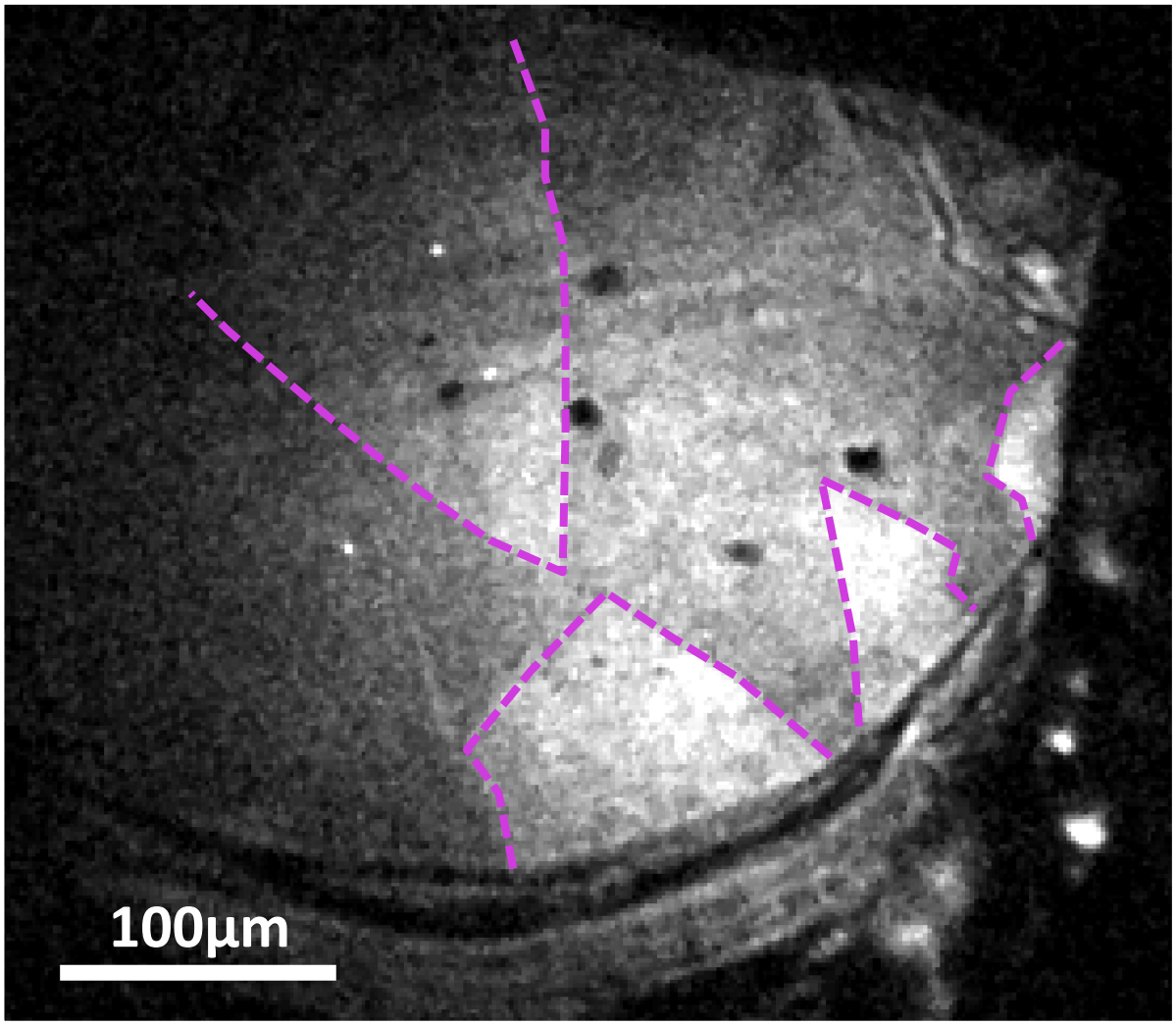


$$\frac{\chi^{qee}\,E\nabla E}{\text{E. Q. } (4/m)} + \frac{\chi^{pee}\,EE}{\text{E. D. } (2'/m)}$$

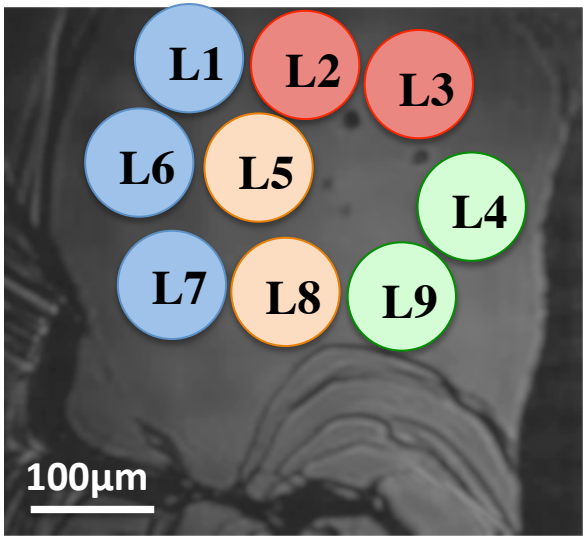
SHG Imaging on Sr_2IrO_4 at 295K

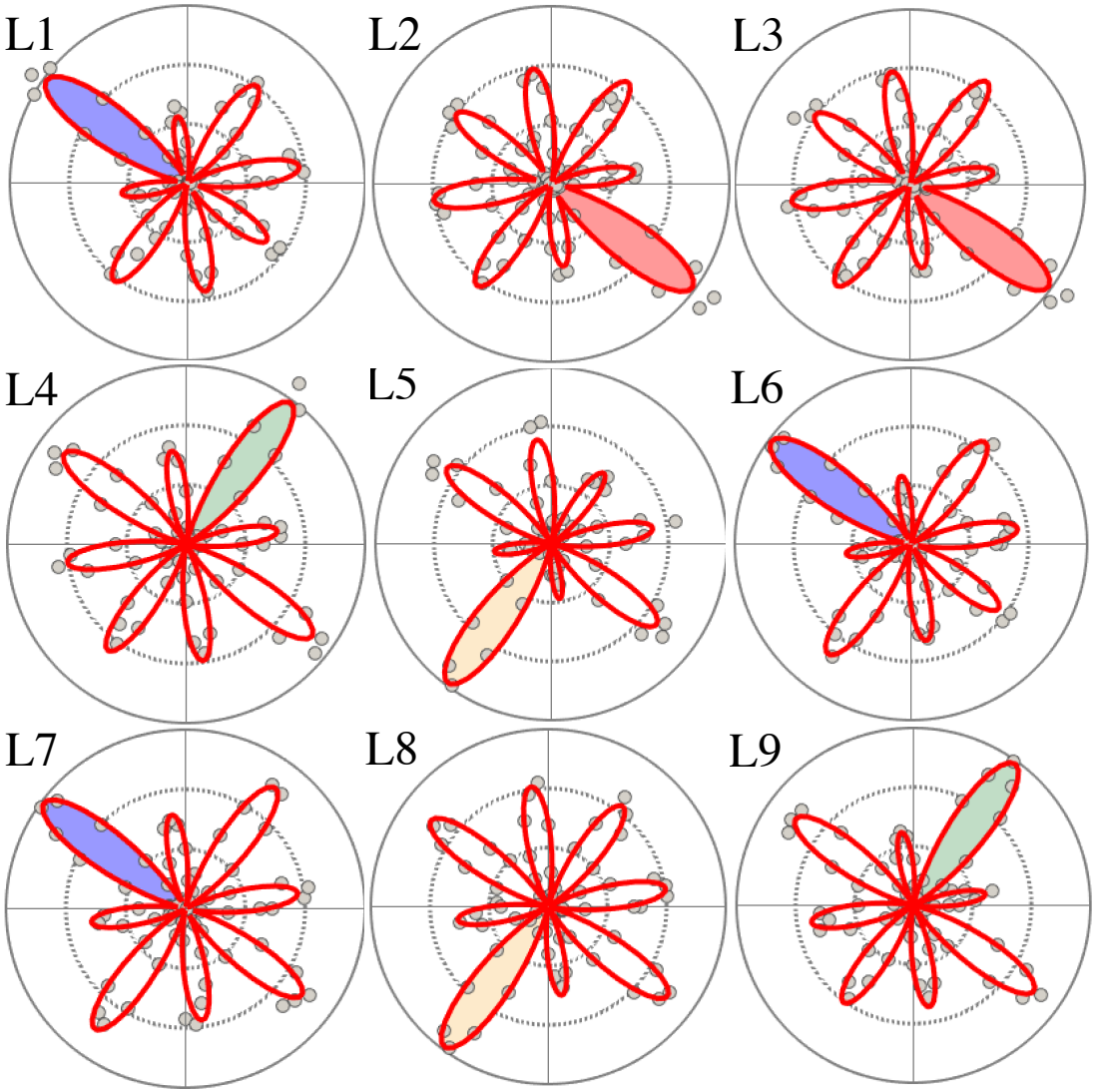


SHG Imaging on Sr_2IrO_4 at 175K

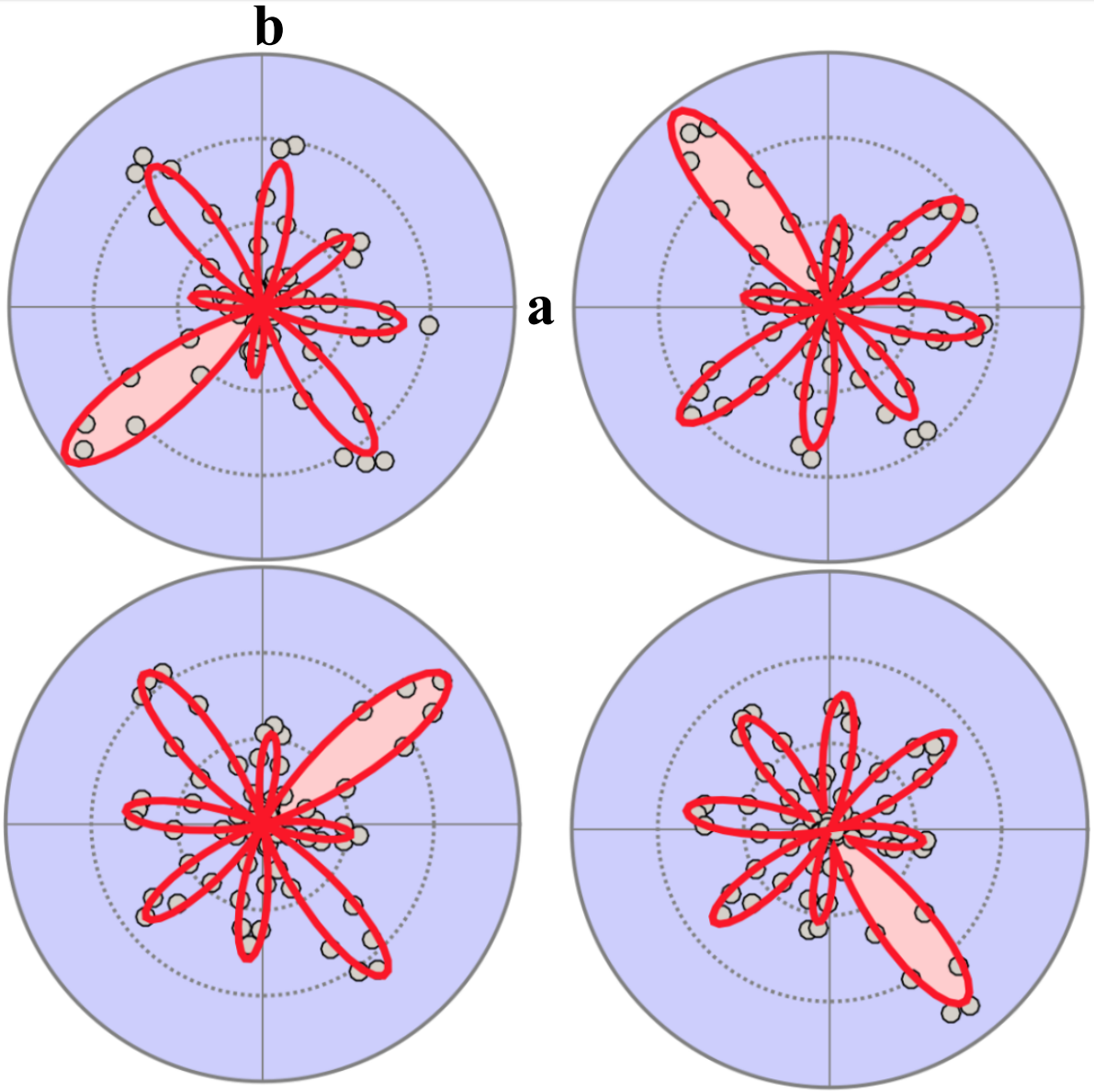


Scanning RA–SHG on Sr_2IrO_4 at 175K



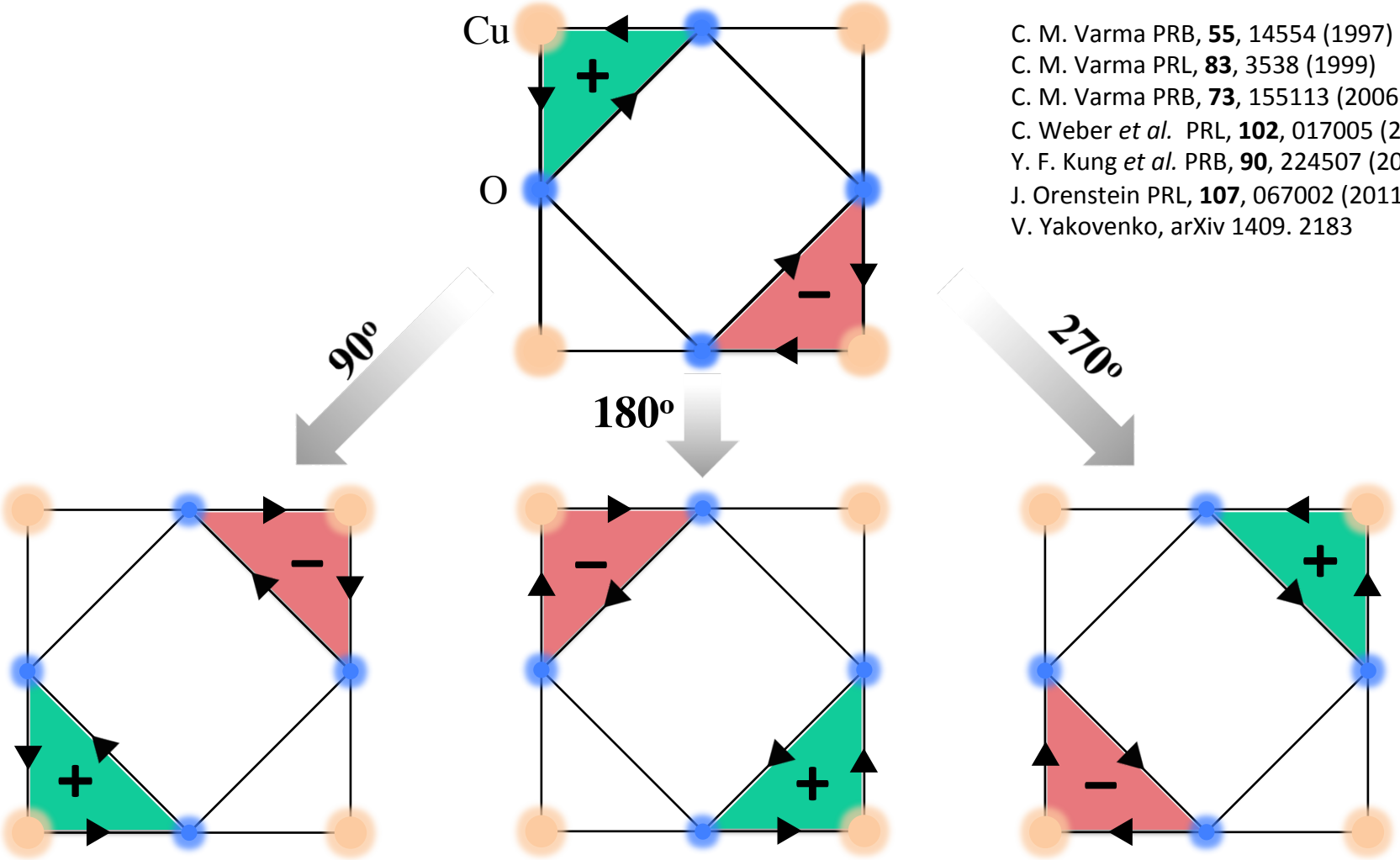


Four Domains of the Hidden Order in Sr_2IrO_4



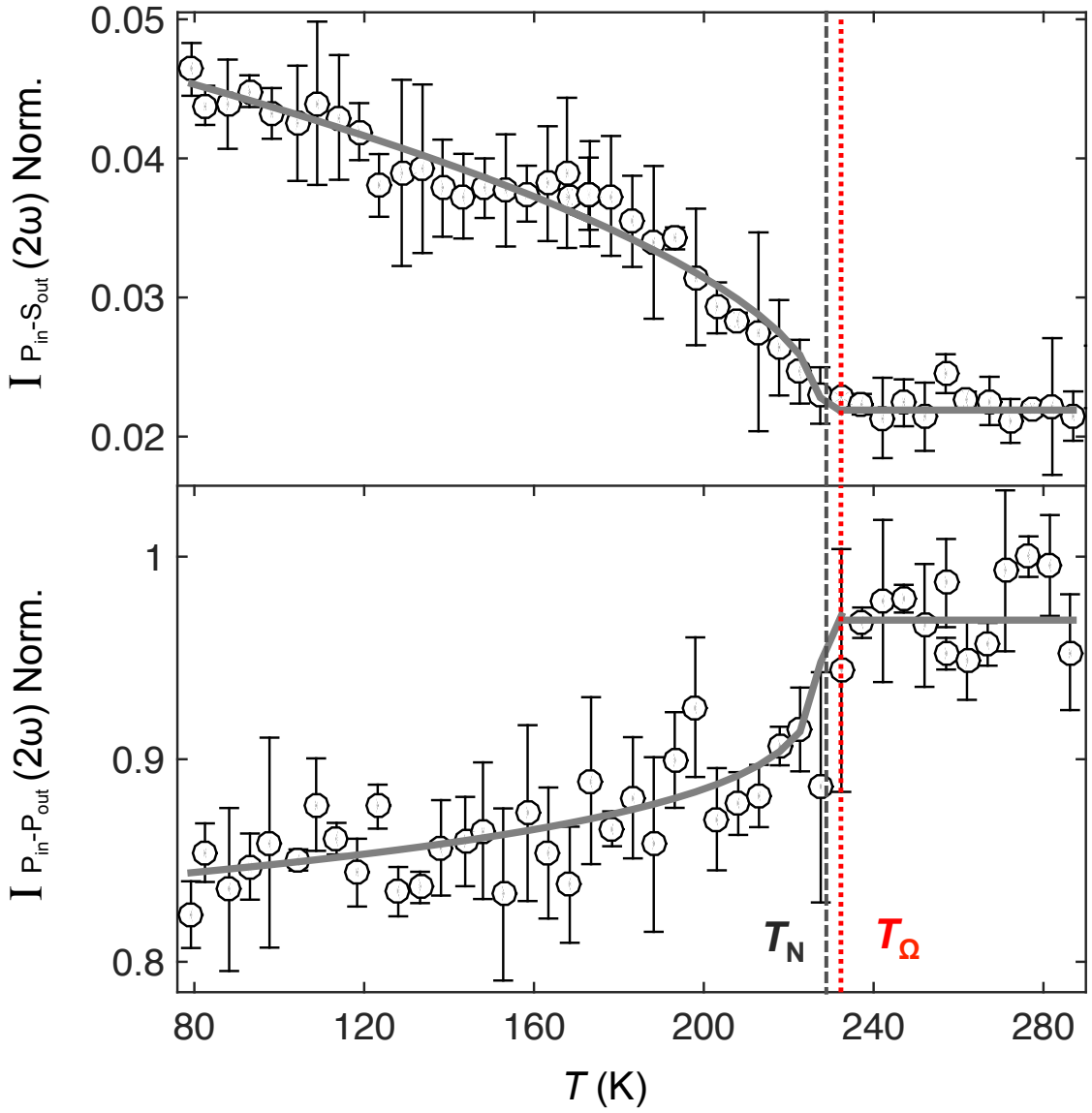
Consistency with Θ_{II} Loop Current Order (magneto-electric)

C_4 **C_1 & Broken Inv. & T. R.**



C. M. Varma PRB, **55**, 14554 (1997)
C. M. Varma PRL, **83**, 3538 (1999)
C. M. Varma PRB, **73**, 155113 (2006)
C. Weber *et al.* PRL, **102**, 017005 (2009)
Y. F. Kung *et al.* PRB, **90**, 224507 (2014)
J. Orenstein PRL, **107**, 067002 (2011)
V. Yakovenko, arXiv 1409. 2183

Relation between Hidden Magnetic Order and AFM in Sr_2IrO_4

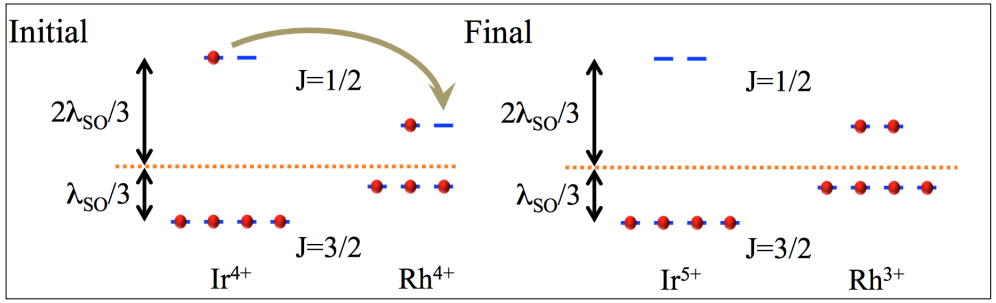


- Ordering temperature
- T_Ω ~ 232 K
- T_N ~ 230 K

$Sr_2Ir_{1-x}Rh_xO_4$

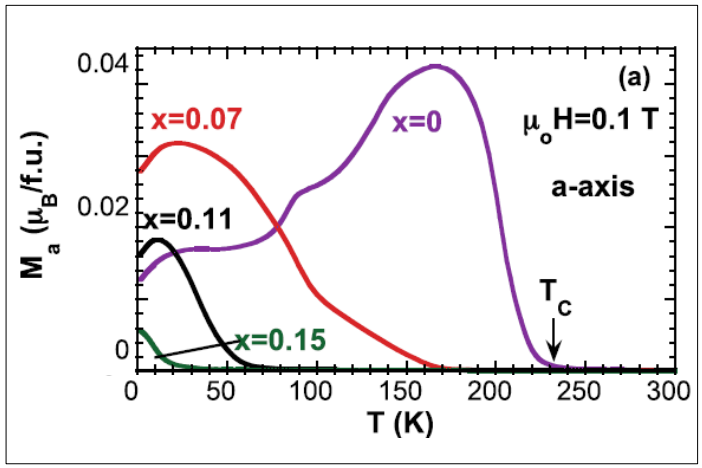
- Hole doping

  Y. Cao *et al.*, http://arxiv.org/14406.4978
  J. P. Clancy *et al.*, PRB 89, 054409



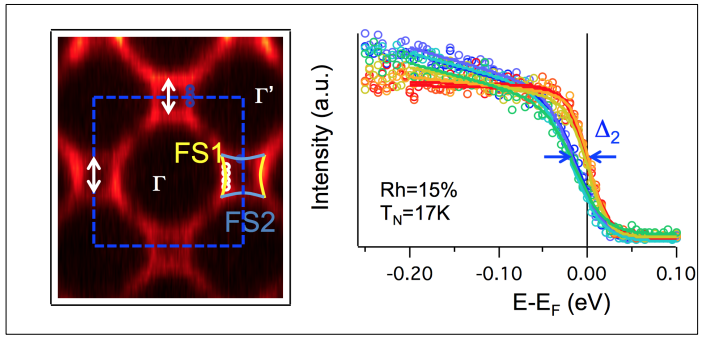
- Suppressed AFM

  J. P. Clancy *et al.*, PRB 89, 054409
  T. F. Qi *et al.*, PRB, 86, 125105

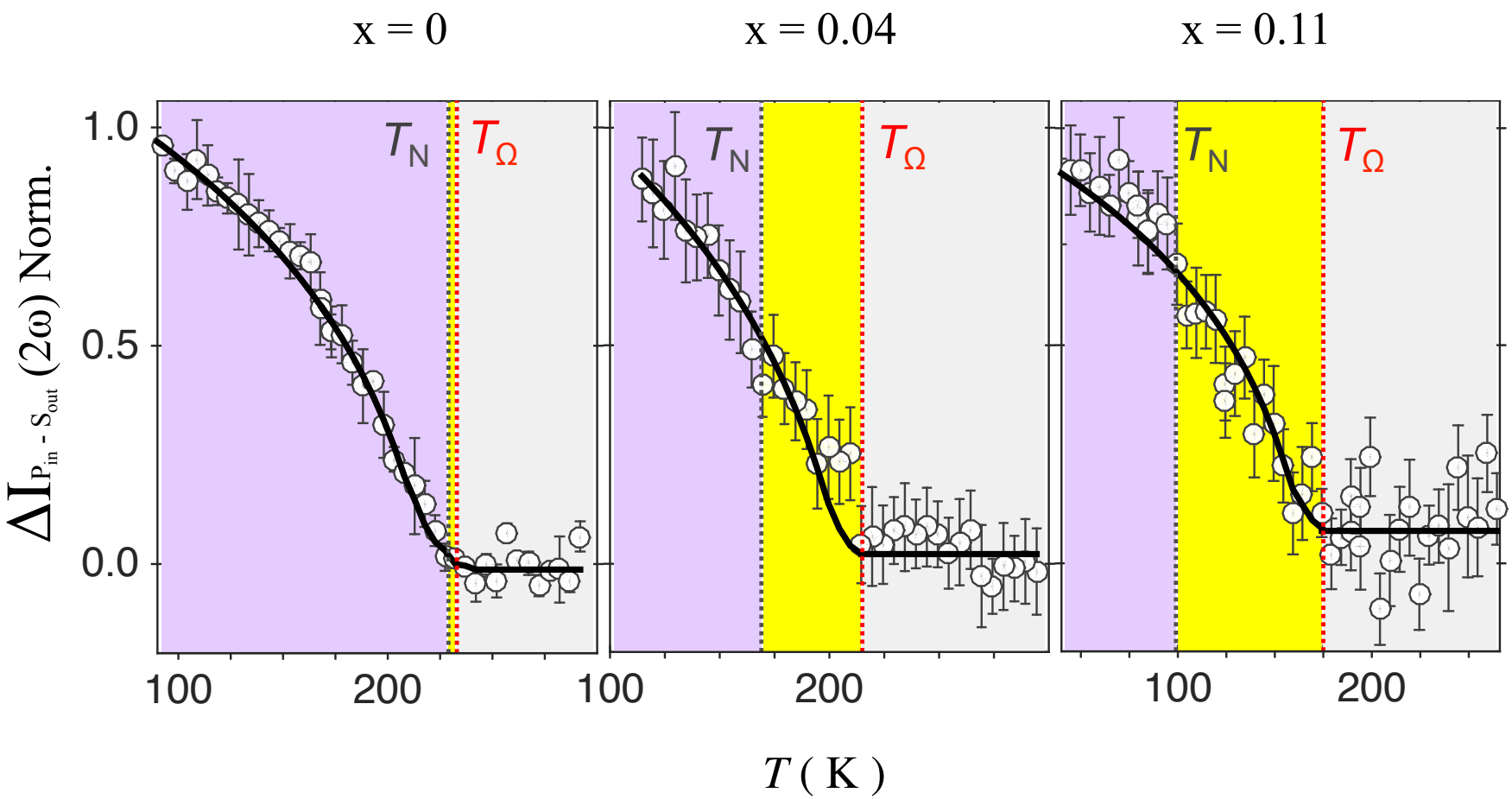


- Pseudogap

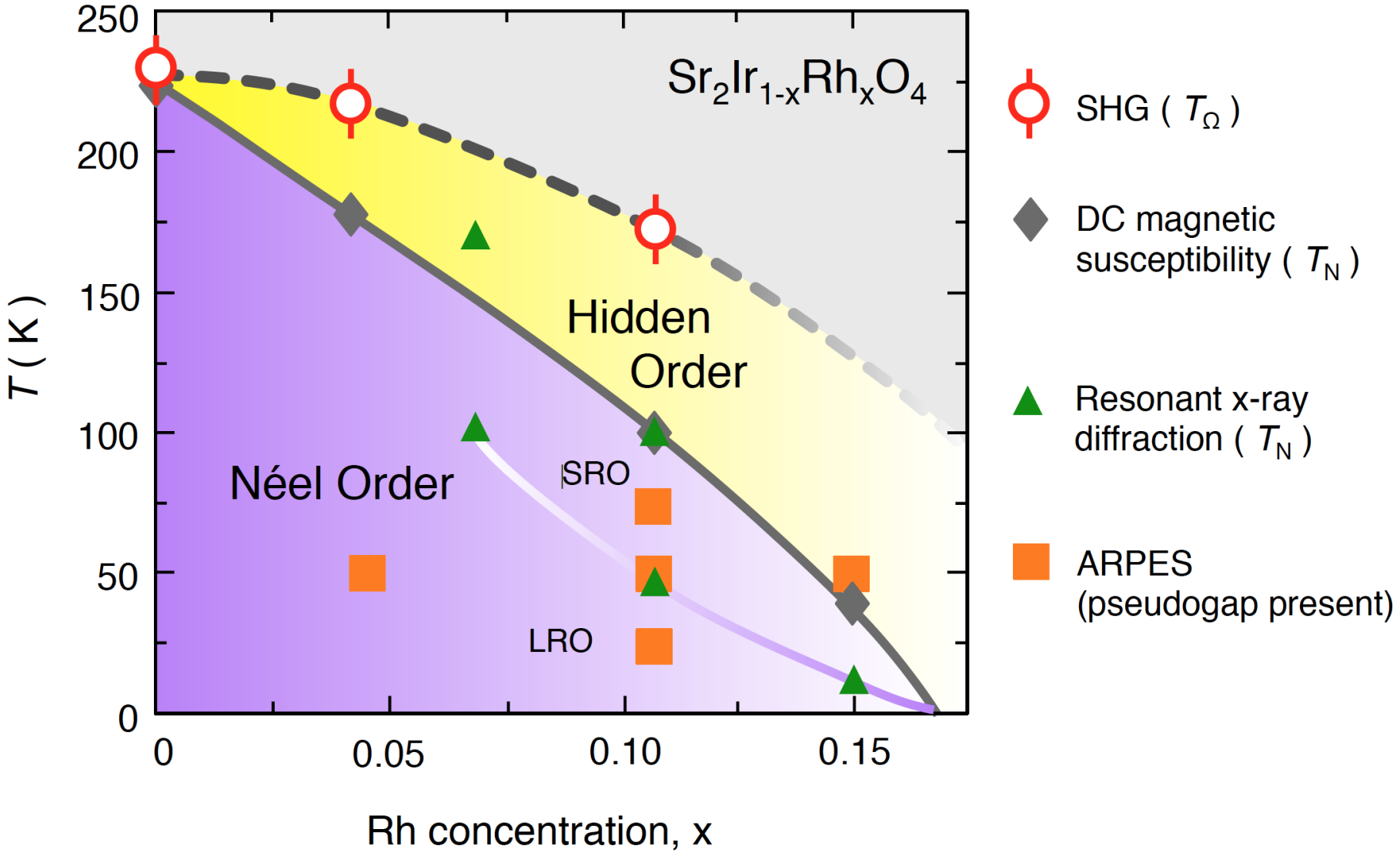
  Y. Cao *et al.*, http://arxiv.org/14406.4978



T_Ω Evolution upon Rh Doping in $Sr_2Ir_{1-x}Rh_xO_4$



Hidden Magnetic Order Region in $Sr_2Ir_{1-x}Rh_xO_4$



Conclusions and Outlook

- A hidden magnetic order: broken rotation, inversion, time reversal symmetries.
- Four domains.
- Not trivially tied to AFM
- Possible relation with Pseudogap behavior ?

Acknowledgements

David Hsieh

Darius Torchinsky

Hao Chu

Tongfei Qi

Gang Cao

Natalia Perkins

Rebecca Flint

Ron Lifshitz