Hidden Order in a Perovskite Iridate Revealed by Nonlinear Optics

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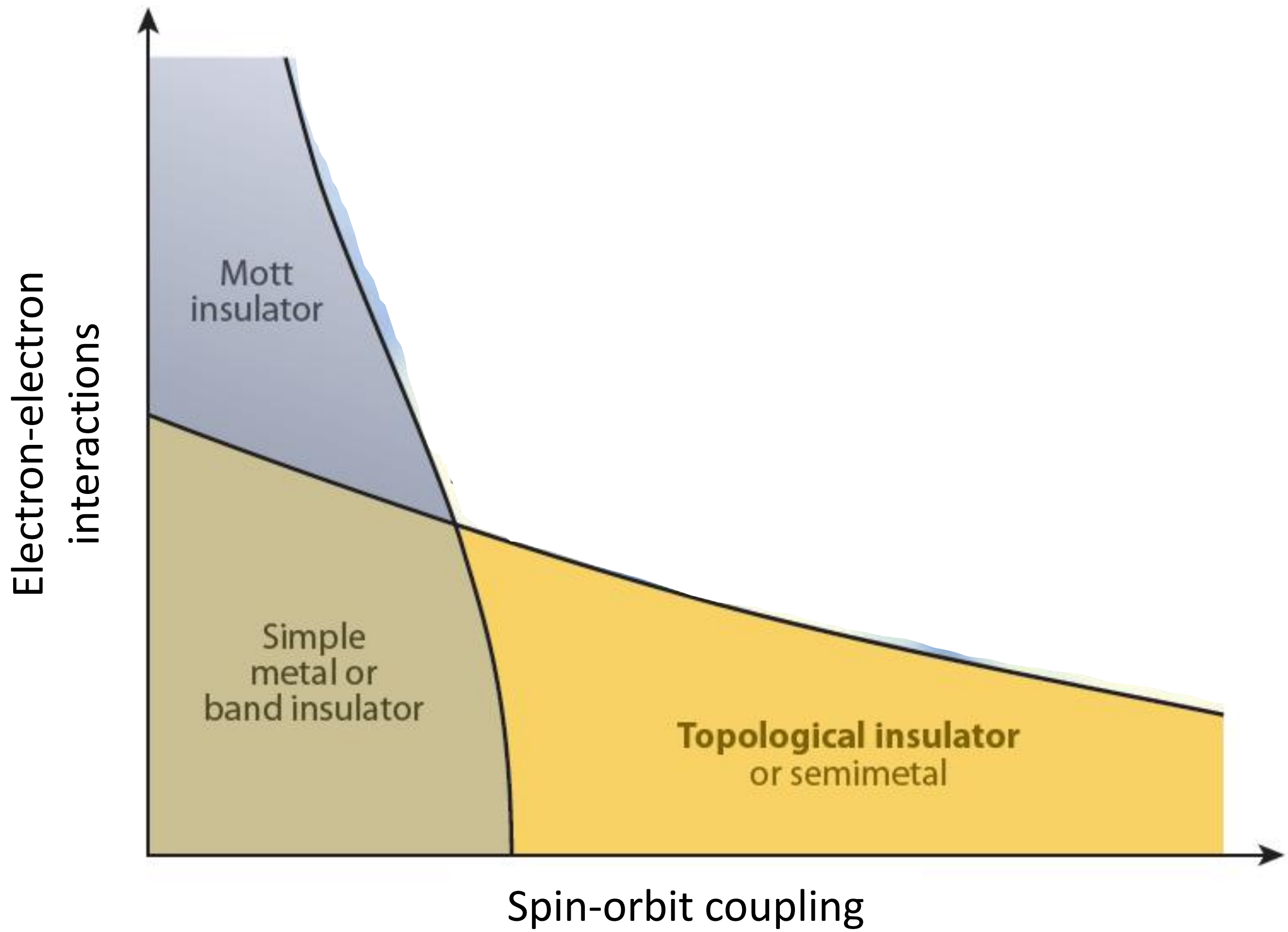
KITP Program: New Phases and Emergent Phenomena in Correlated Materials with Strong Spin-Orbit Coupling
9/15/2015

Electron correlations + spin-orbit coupling



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

Outline

- Sr_2IrO_4 : A spin-orbit coupled Mott insulator

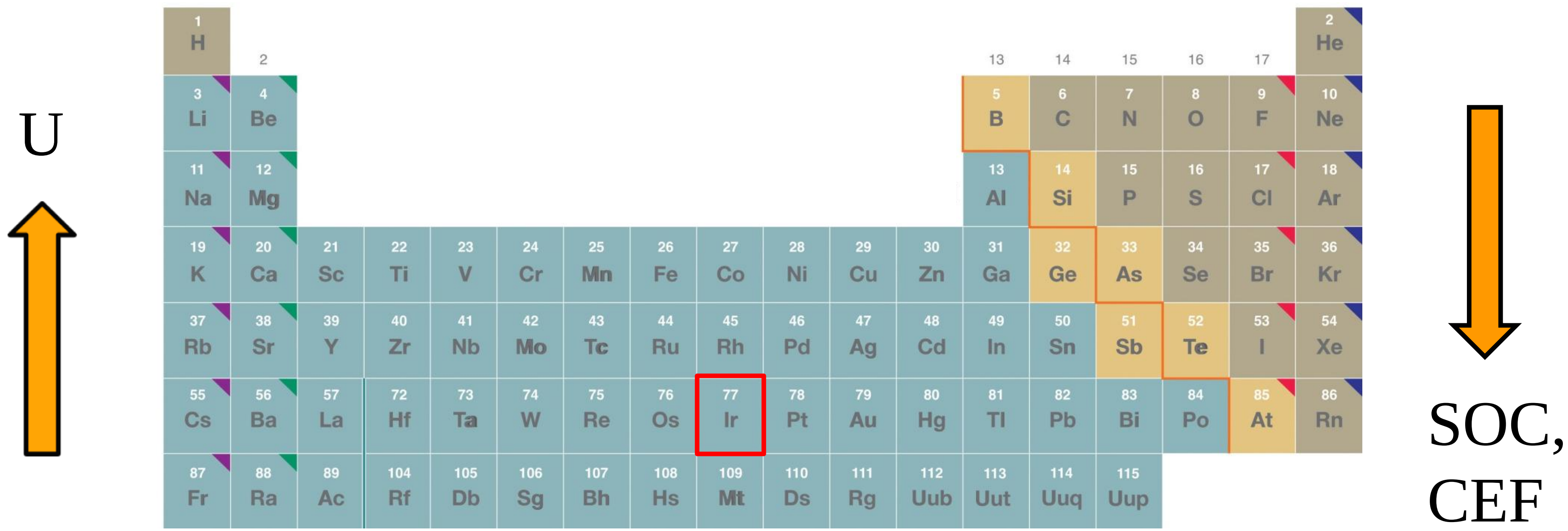
- Nonlinear optical harmonic generation

  - A tool for detecting electronic symmetry breaking
  - Principles of operation
  - High precision rotational anisotropy

- Physics of Sr_2IrO_4 revealed through nonlinear optics

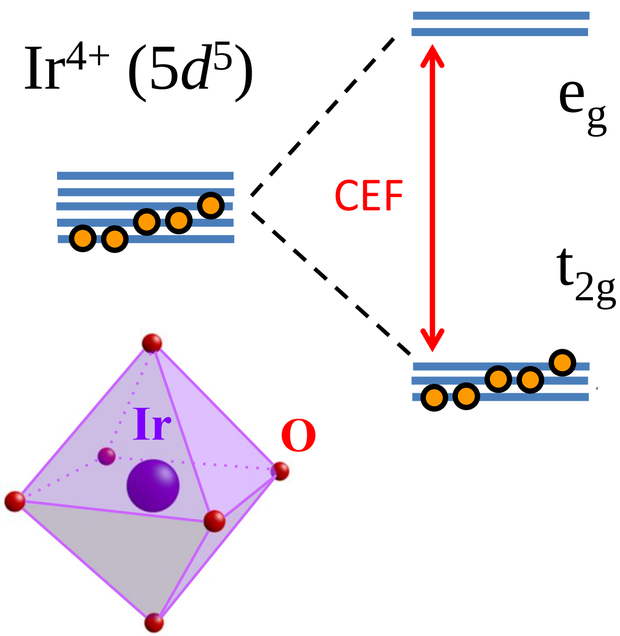
  - Perfect magneto-elastic locking via structural distortion
  - A hidden odd-parity magnetic order

5*d* transition metal oxides



- Interplay between electron correlations, crystal electric field and spin-orbit coupling and (U ~ SOC ~ CEF)
- Potential for exotic physics driven by strong SOC (~0.5eV)

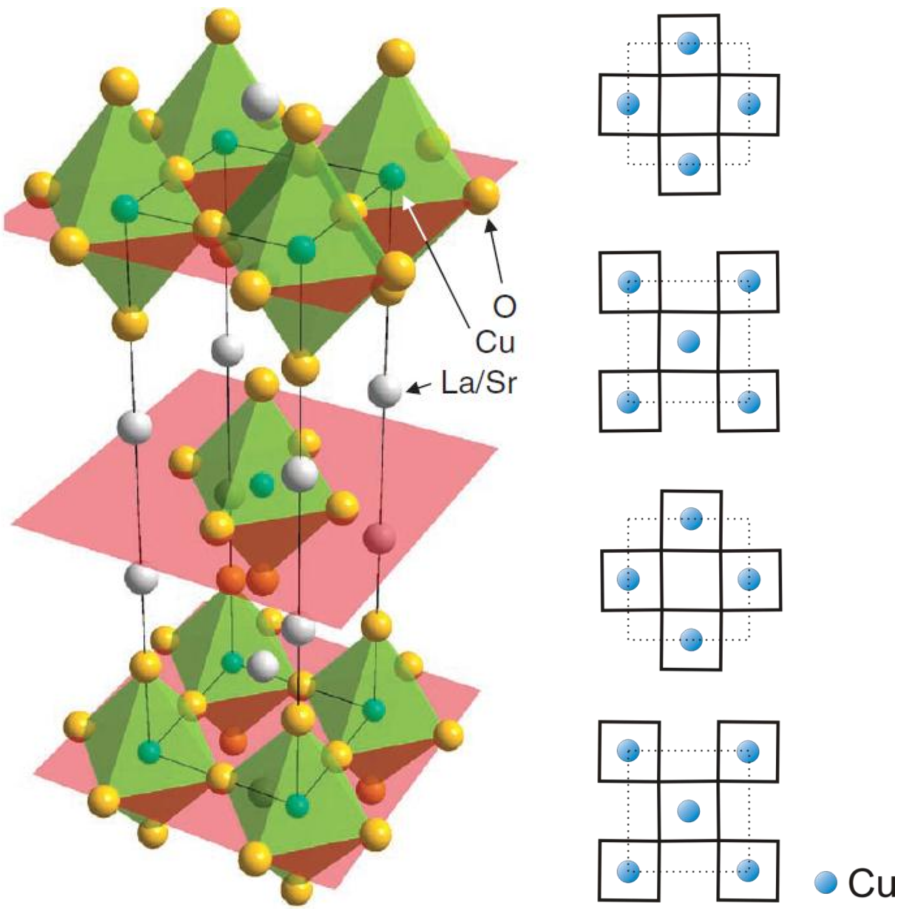
J_{eff} = ½ Mott insulators in 5*d* systems



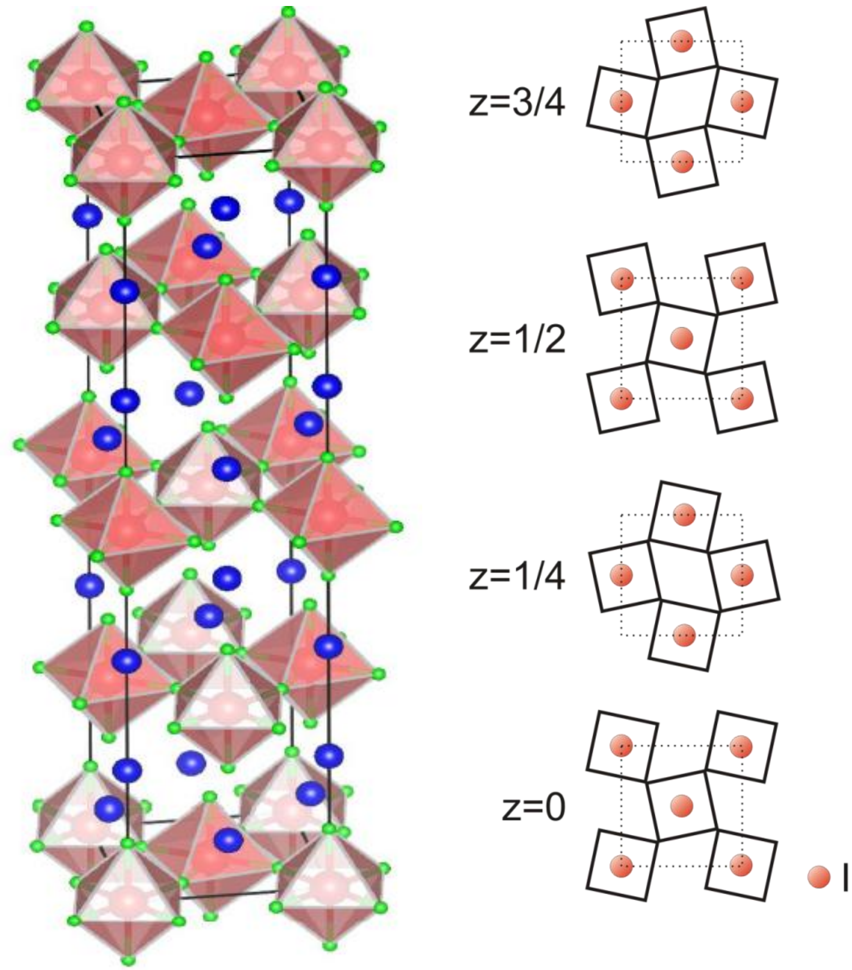
Kim *et al*., PRL 101, 076402 (2008)

Sr_2IrO_4 (Single-layer perovskite structure)

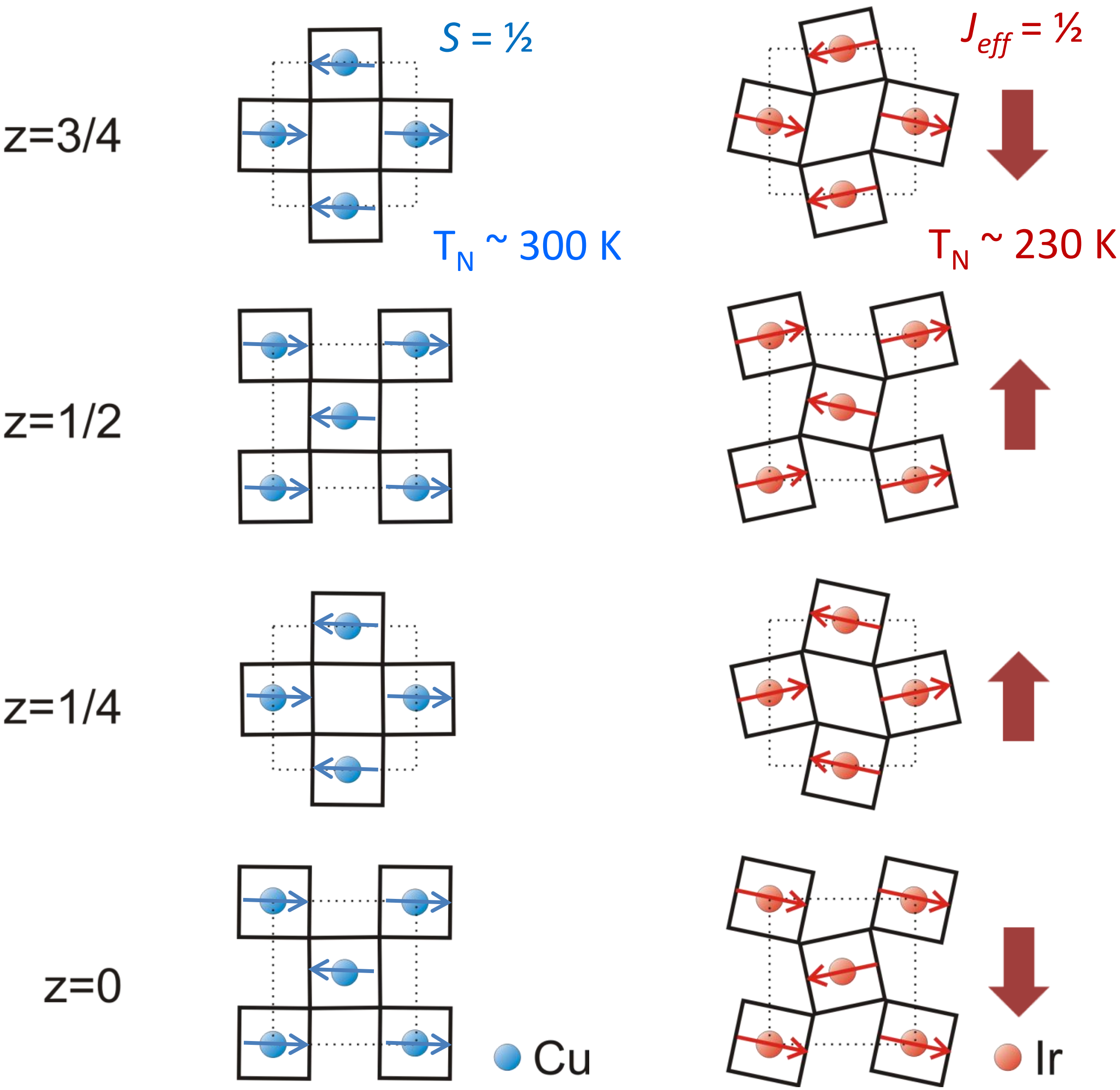
La_2CuO_4



Sr_2IrO_4



Sr_2IrO_4 (Orthorhombic magnetic structure)



In-plane canted dipolar AFM
Centrosymmetric *mmm1'*

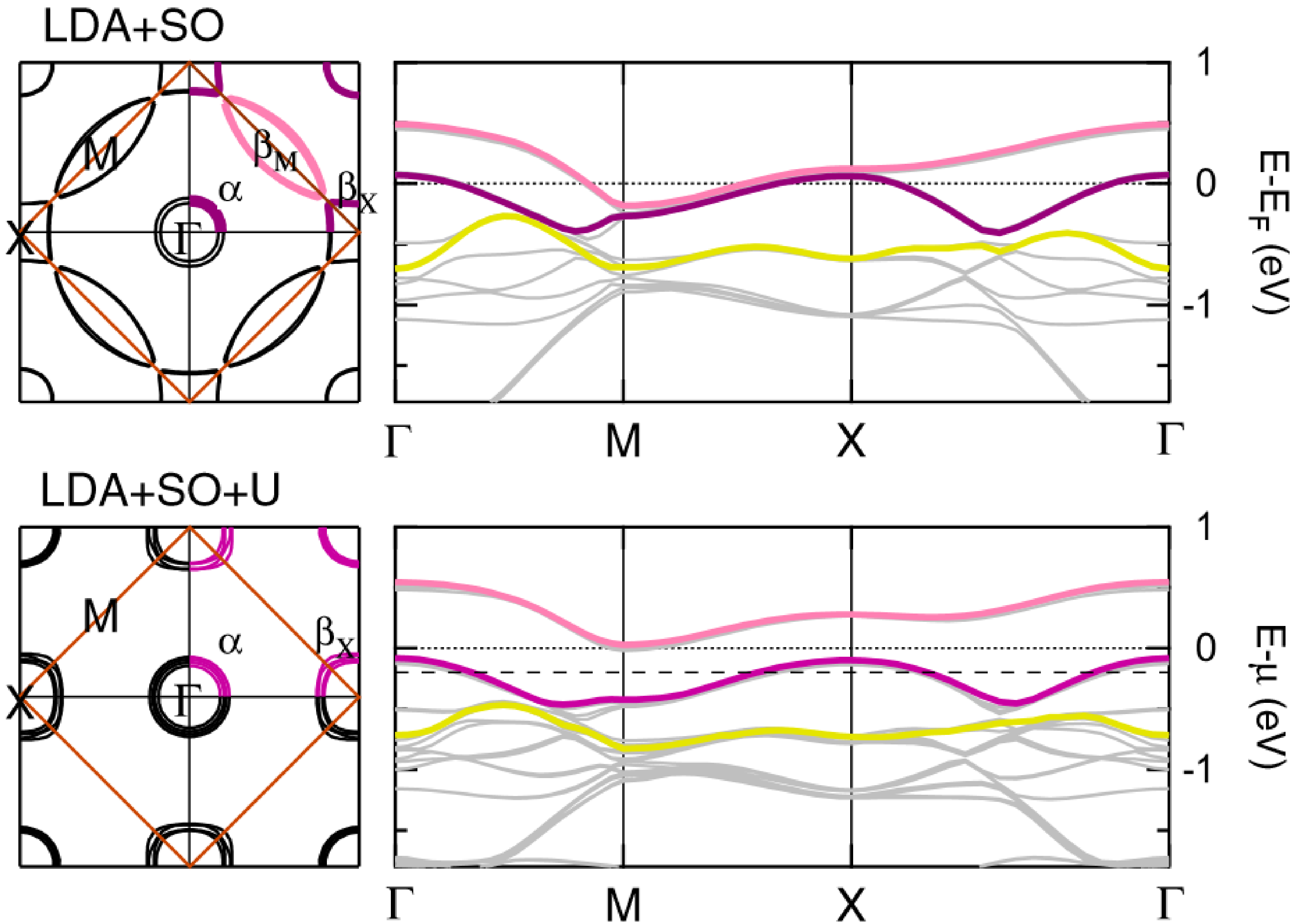
Neutron diffraction

Q. Huang *et al.*, J. Sol. State. Chem. 112, 355 (1994)
C. Dhital *et al.*, PRB 87, 144405 (2013)
F. Ye *et al.*, PRB 87, 140406(R) (2013)

Resonant x-ray diffraction

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)

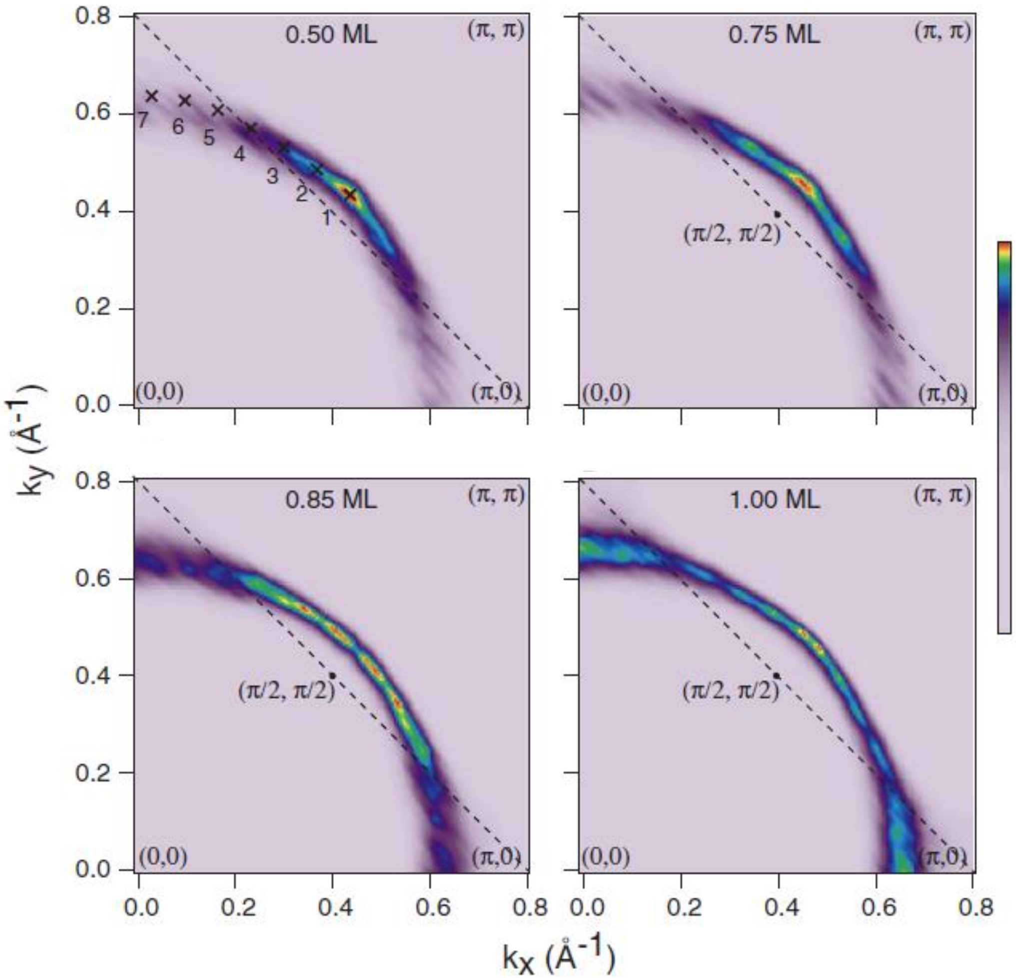
Sr_2IrO_4 (Electronic structure)

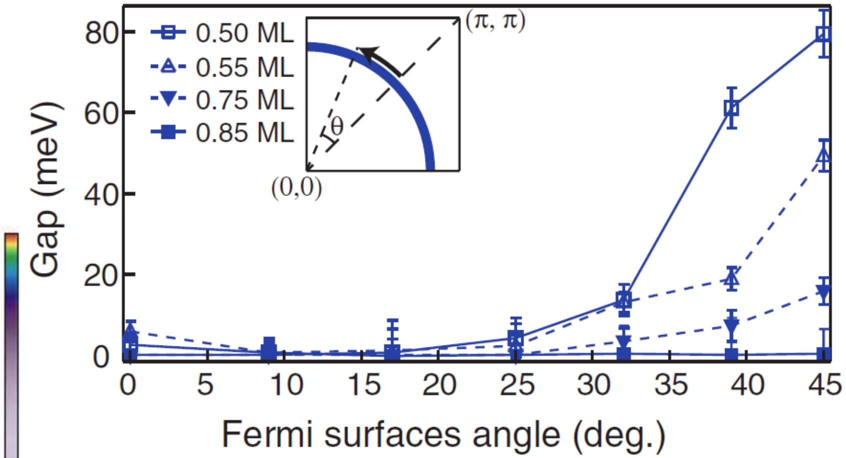


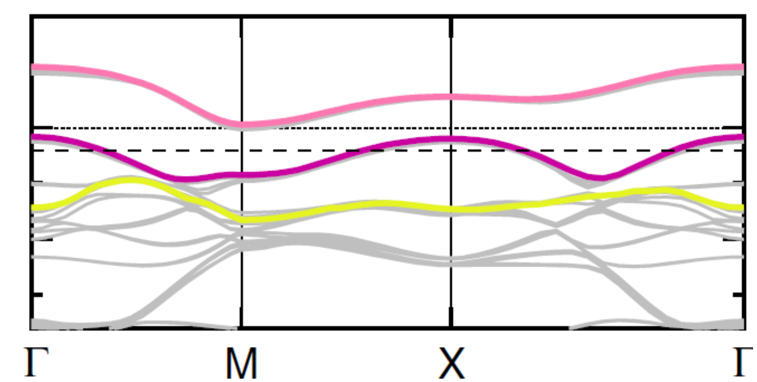
Kim *et al.*, PRL 101, 076402 (2008)

Fermi arcs in surface K doped Sr_2IrO_4 (electron doping)

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





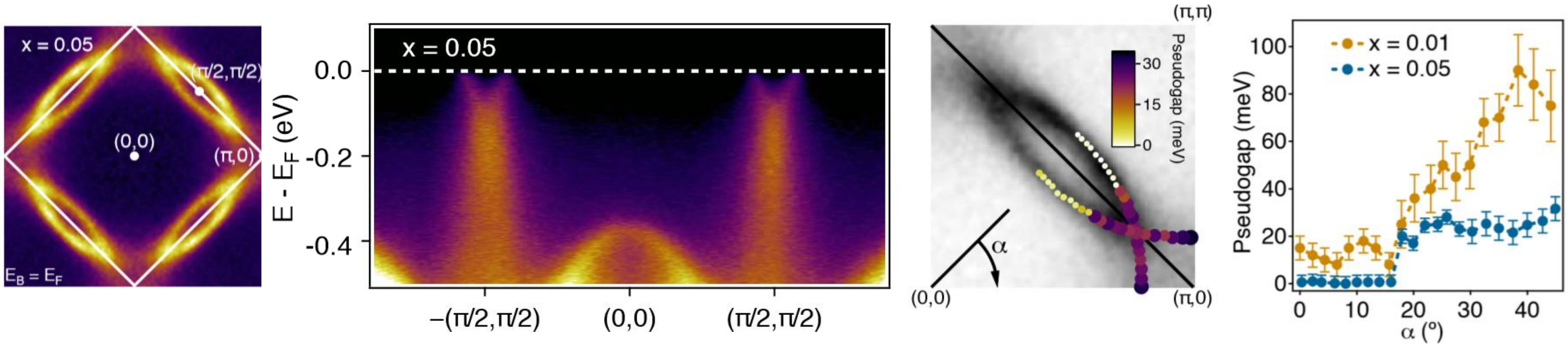


Fermi arcs and pseudogap in bulk doped Sr_2IrO_4

Observation of Fermi Arcs in ARPES measurements

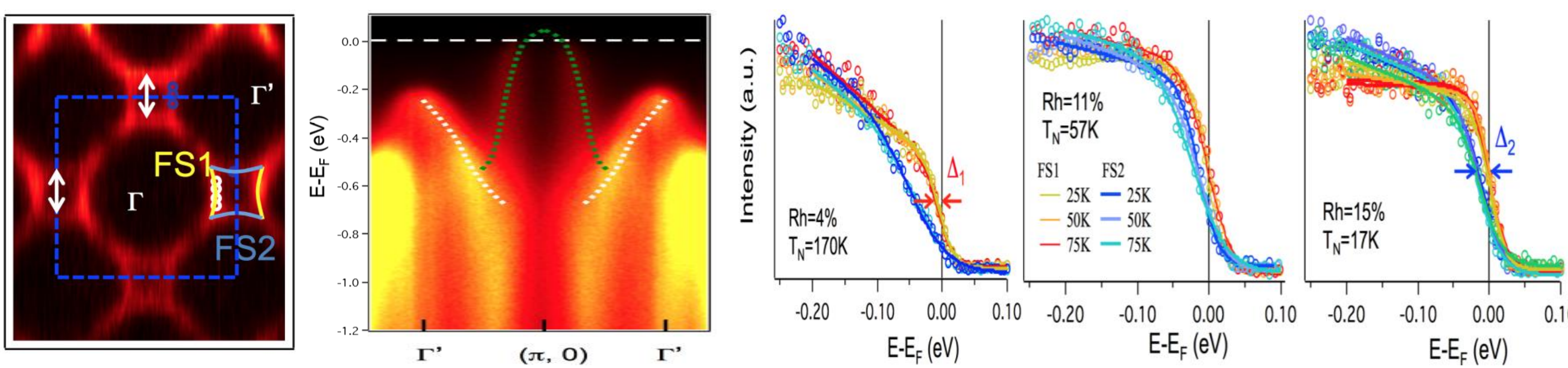
$(Sr_{1-x}La_x)_2IrO_4$ (electron doping)

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

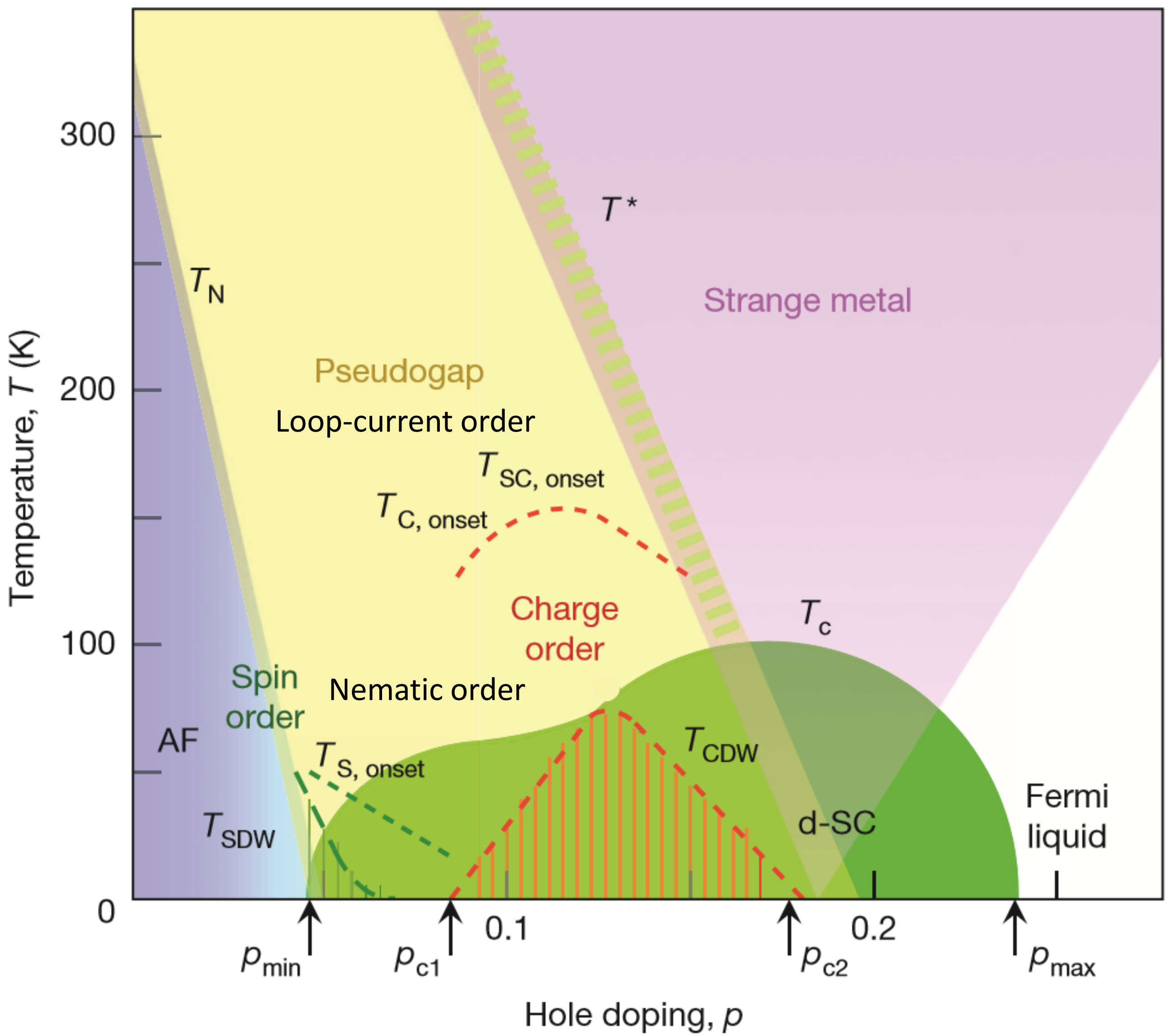


$Sr_2Ir_{1-x}Rh_xO_4$ (hole doping)

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

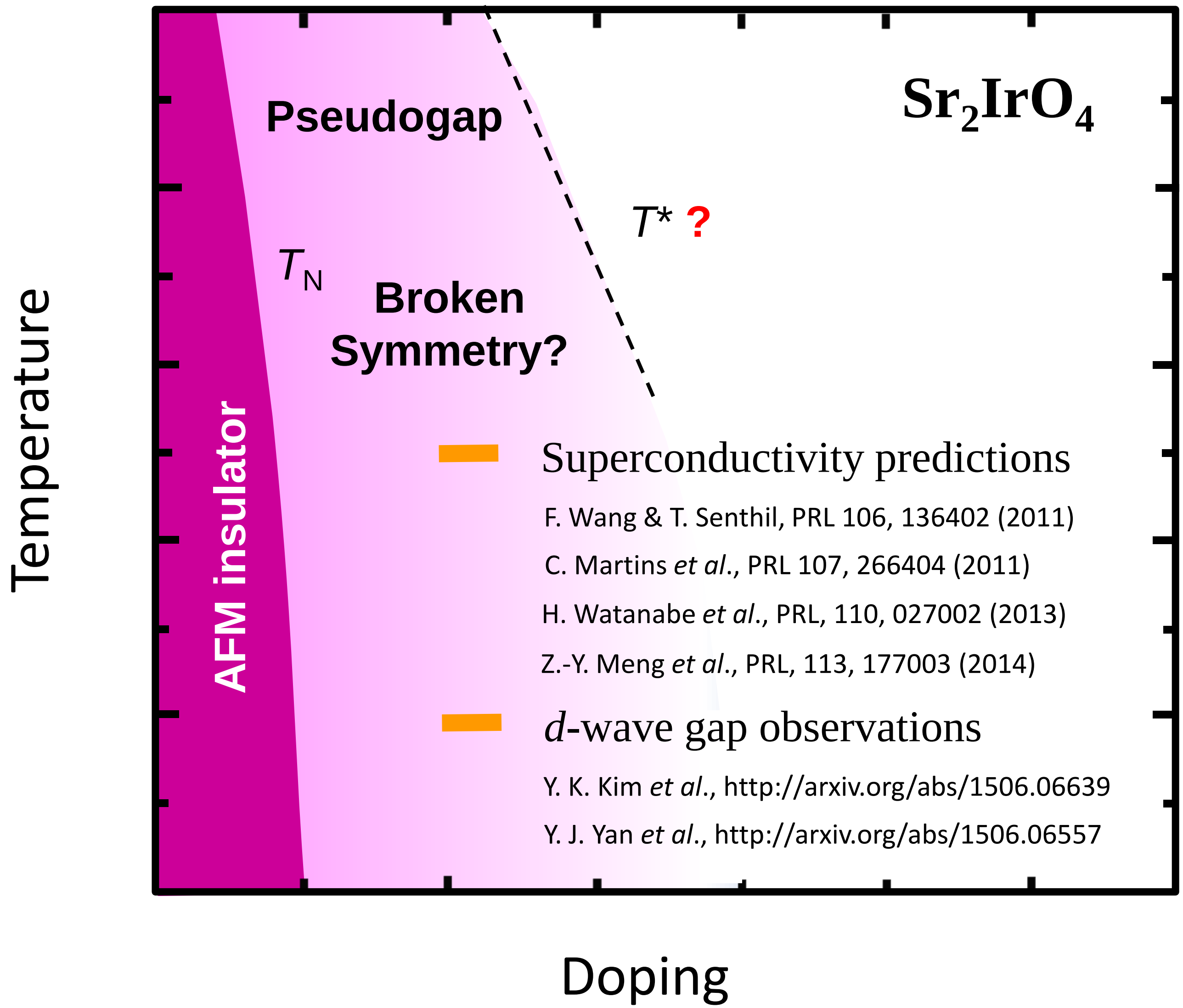


Broken symmetry phases proximate to AF order in cuprates



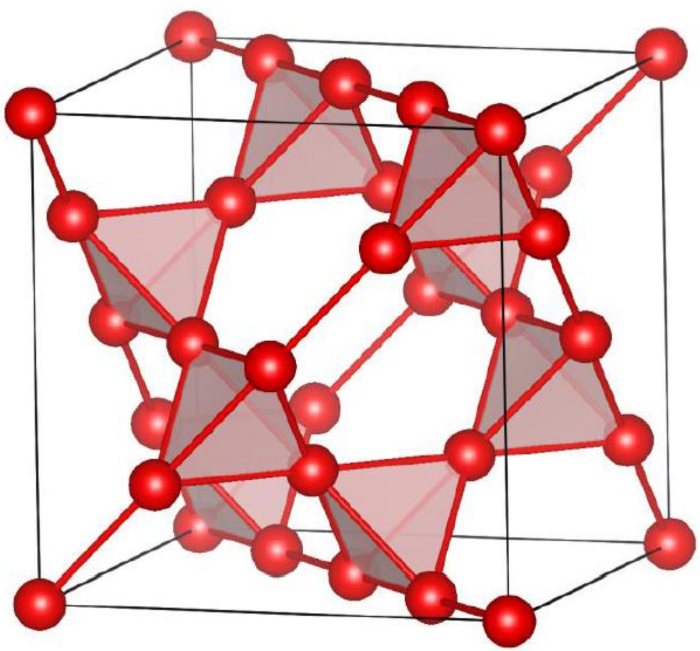
B. Keimer *et al*., Nature Review 518, 179 (2015)

Exotic phases in iridates?



Neumann's principle

A tensor describing any physical property of a crystal must be invariant under all symmetry operations of the crystal



- If $\chi_{\mathrm{ijk}\ldots\mathrm{n}}$ is a property tensor
- and T_{ip} is an element in the symmetry group of the crystal
- then:

$$\chi_{ijk\ldots n} = T_{ip}T_{jq}T_{kr}\ldots T_{nu}\chi_{pqr\ldots u}$$

- The set of relationships between χ's greatly reduced the number of non-zero independent tensor components

R. R. Birss Symmetry & Magnetism (1966)

Higher rank tensors → greater symmetry resolution

$\chi^{(1)}_{ij}$		$\chi^{(2)}_{ijk}$		$\chi^{(3)}_{ijkl}$	
Tetragonal Trigonal Hexagonal	# elem. 2	$4 = C_4$	7	$4 = C_4$	21
		$\bar{4} = S_4$	6	$\bar{4} = S_4$	21
		$422 = D_4$	3	$422 = D_4$	11
$\begin{bmatrix} xx & 0 & 0 \\ 0 & xx & 0 \\ 0 & 0 & zz \end{bmatrix}$		$4mm = C_{4v}$	4	$4mm = C_{4v}$	11
		$\bar{4}2m = D_{2d}$	3*	$\bar{4}2m = D_{2d}$	11
		$4/m = C_{4h}$	0	$4/m = C_{4h}$	21
		$4/mmm = D_{4h}$	0	$4/mmm = D_{4h}$	11

Nonlinear optics

- Multipole expansion of radiation source term

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

- Expansion of electric dipole (P), magnetic dipole (M) and electric quadrupole (Q) contributions

$$P_i = \chi_{ij}^{ee} E_j(\omega) + \chi_{ij}^{em} H_j(\omega) + \chi_{ijk}^{eee} E_j(\omega) E_k(\omega) + \chi_{ijk}^{eem} E_j(\omega) H_k(\omega) + \chi_{ijk}^{emm} H_j(\omega) H_k(\omega) + \ldots$$

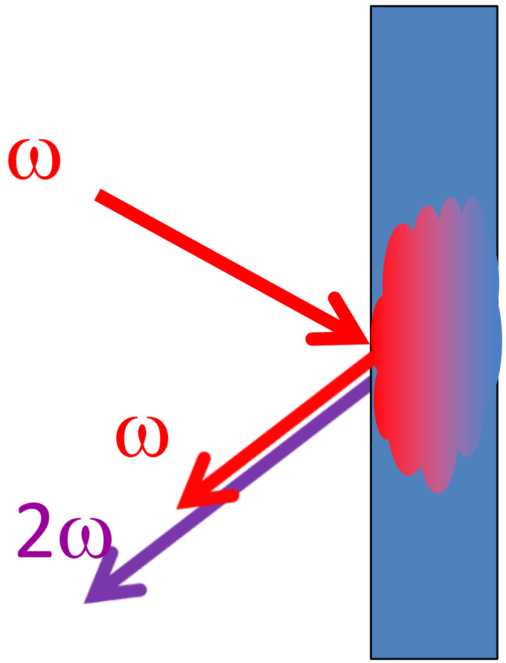
$$M_i = \chi_{ij}^{me} E_j(\omega) + \chi_{ij}^{mm} H_j(\omega) + \chi_{ijk}^{mee} E_j(\omega) E_k(\omega) + \chi_{ijk}^{mem} E_j(\omega) H_k(\omega) + \chi_{ijk}^{mmm} H_j(\omega) H_k(\omega) + \ldots$$

$$\hat{Q}_{ij} = \underbrace{\chi_{ijk}^{qe} E_k(\omega) + \chi_{ijk}^{qm} H_k(\omega)}_{1^{st} \text{ order responses}} + \underbrace{\chi_{ijkl}^{qee} E_k(\omega) E_l(\omega) + \chi_{ijkl}^{qem} E_k(\omega) H_l(\omega) + \chi_{ijkl}^{qmm} H_k(\omega) H_l(\omega) + \ldots}_{2^{nd} \text{ order responses}}$$

1st order responses

2nd order responses

Optical second harmonic generation (SHG)



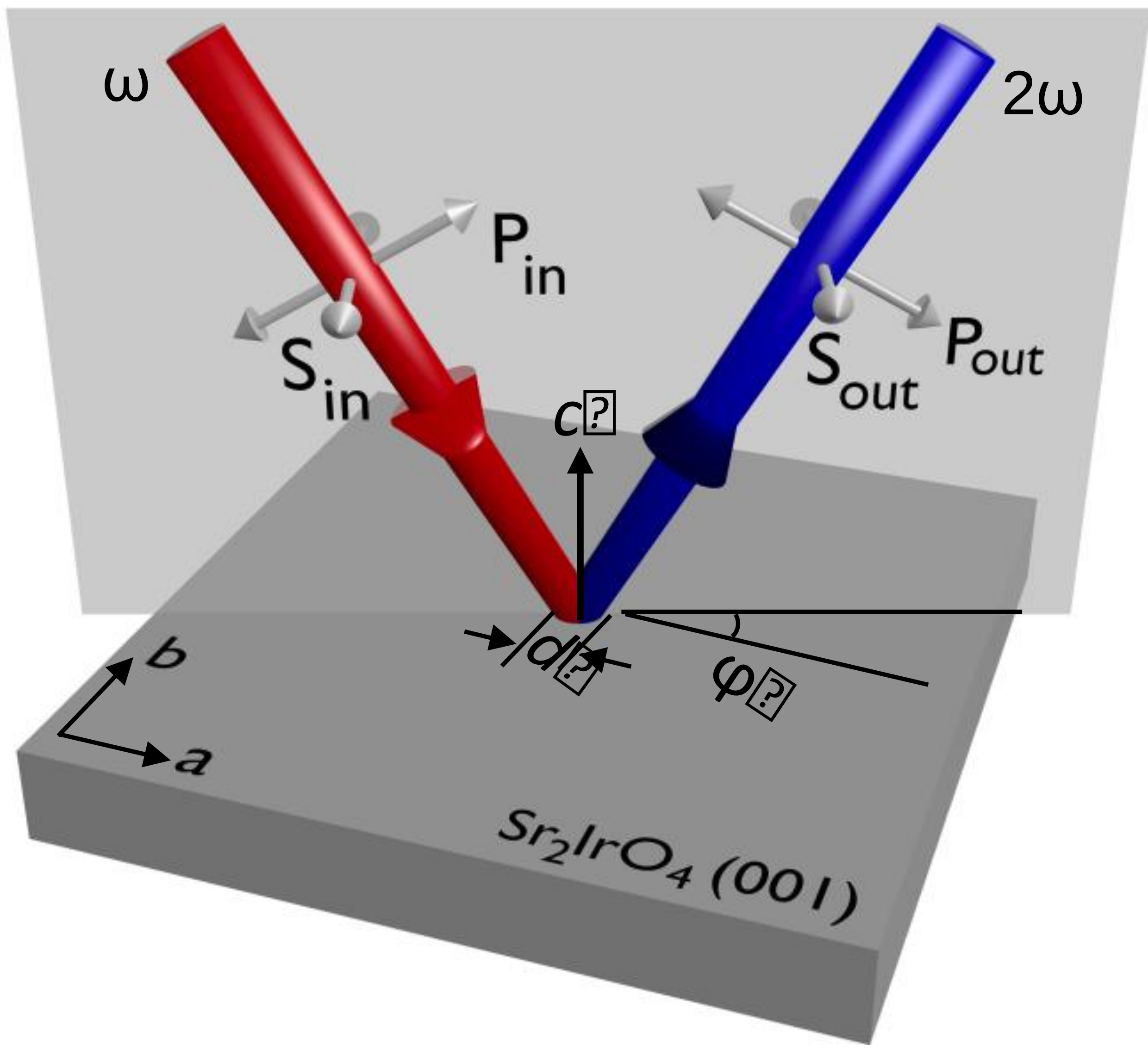
$$P_i(\omega) = \chi_{ij}^{ED} E_j(\omega)$$

$$P_i(2\omega) = \chi_{ijk}^{ED} E_j(\omega) E_k(\omega) + \chi_{ijkl}^{EQ} E_j(\omega) \nabla_k E_l(\omega) + \ldots$$

$\chi_{ijk}^{ED} = 0$ if system has inversion symmetry

$\chi_{ijkl}^{EQ} \neq 0$ even if system has inversion symmetry
much weaker than ED contribution (~λ/a)

Rotational anisotropy



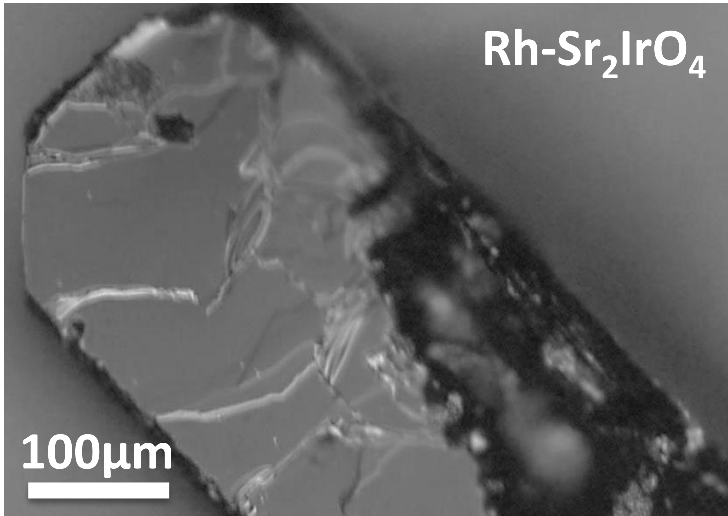
Technical limitations of conventional rotational anisotropy

Alignment problems:

- Beam walk on sample
- Precession of reflected light
- Need large area flat single crystals (e.g. thin-film, polishing)

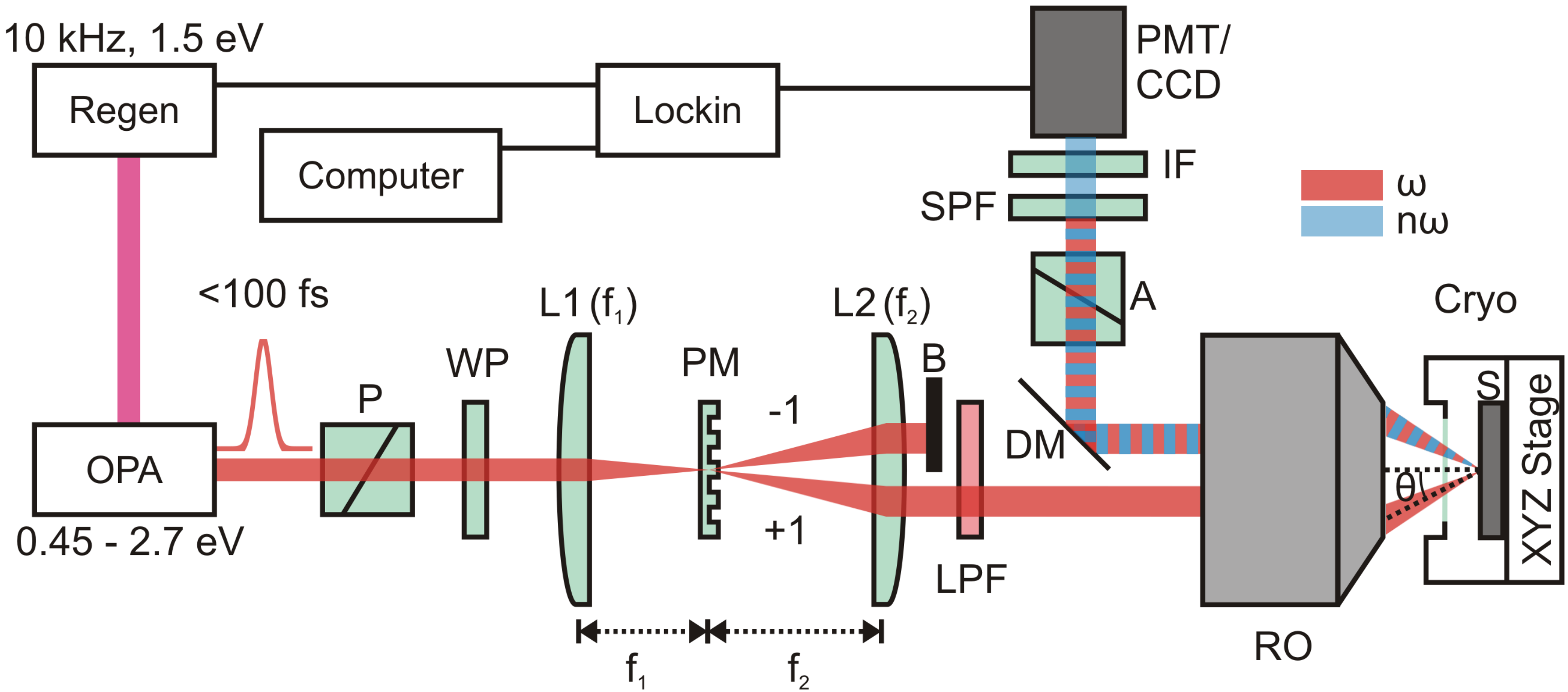
Need for rotating sample creates additional challenges for:

- Cryogenic measurements
- Magnetic field measurements
- Strained samples
- Imaging measurements



Rotating scattering plane based RA-SHG

Instead of rotating the sample, the scattering plane is rotated.

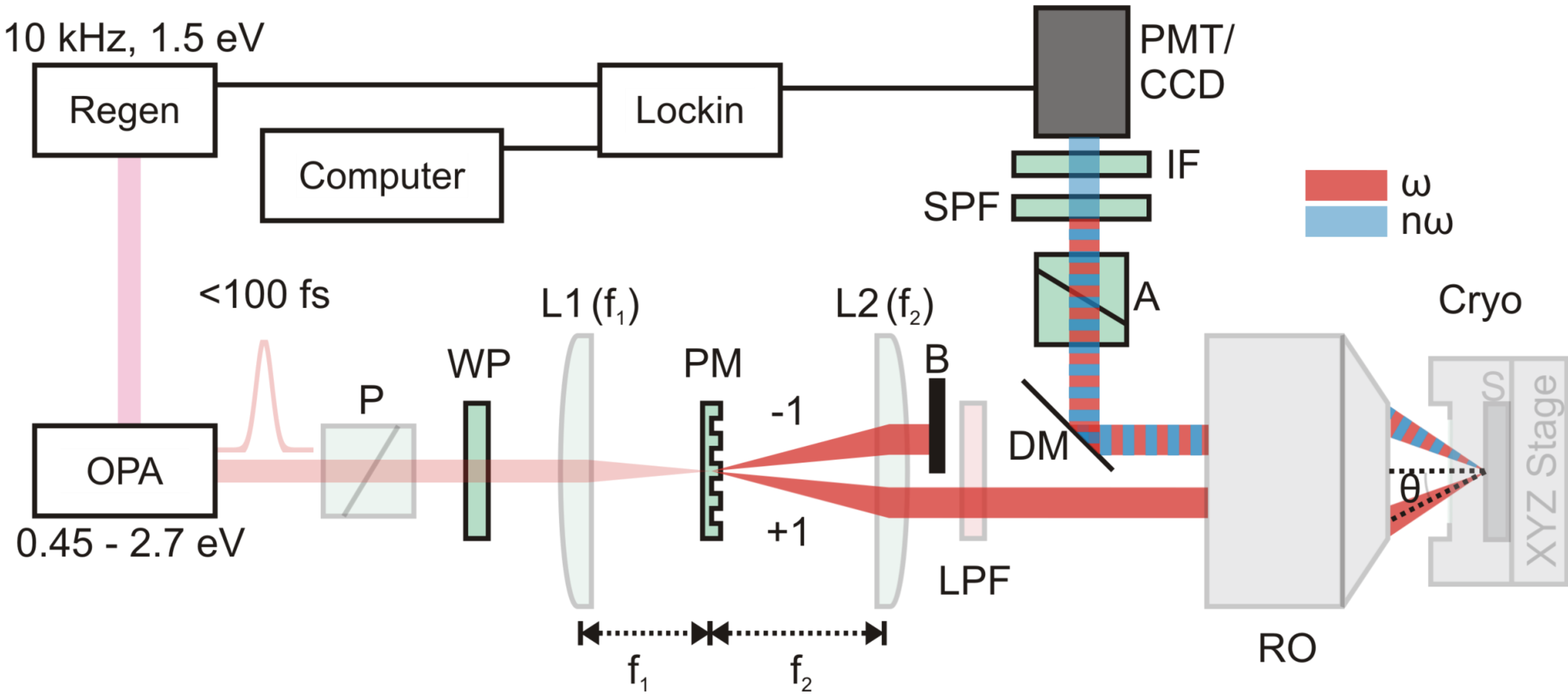


Measurement is performed at oblique angle θ✓

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

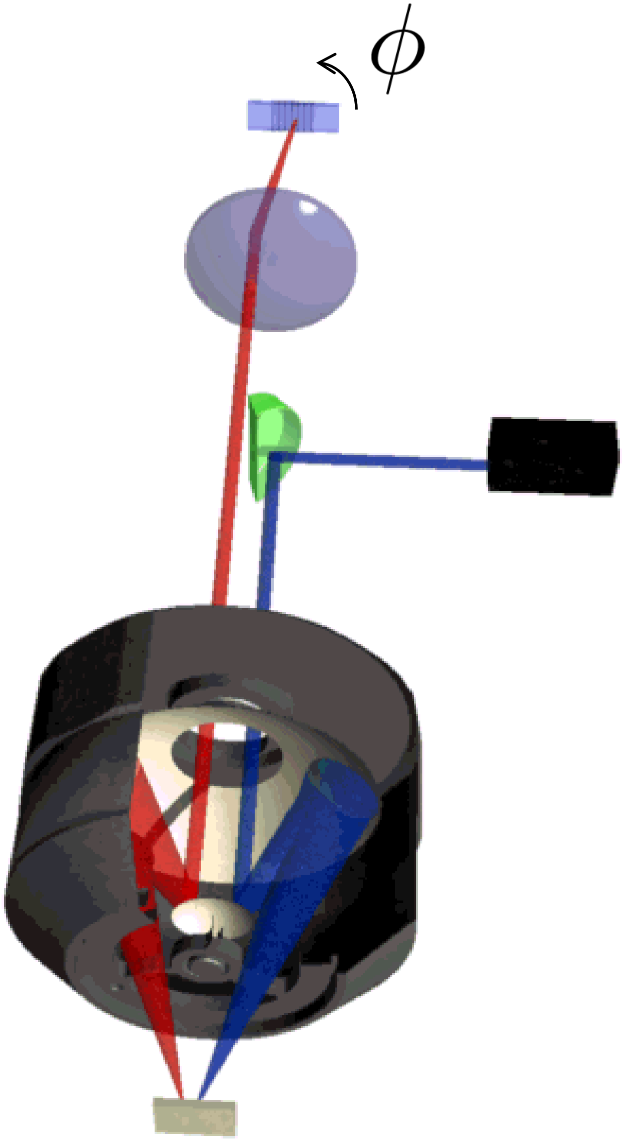
Rotating scattering plane based RA-SHG

Only a handful of optics rotate.



D. H. Torchinsky et al., Rev. Sci. Instrum. 85, 083102 (2014)

Rotating scattering plane based RA-SHG

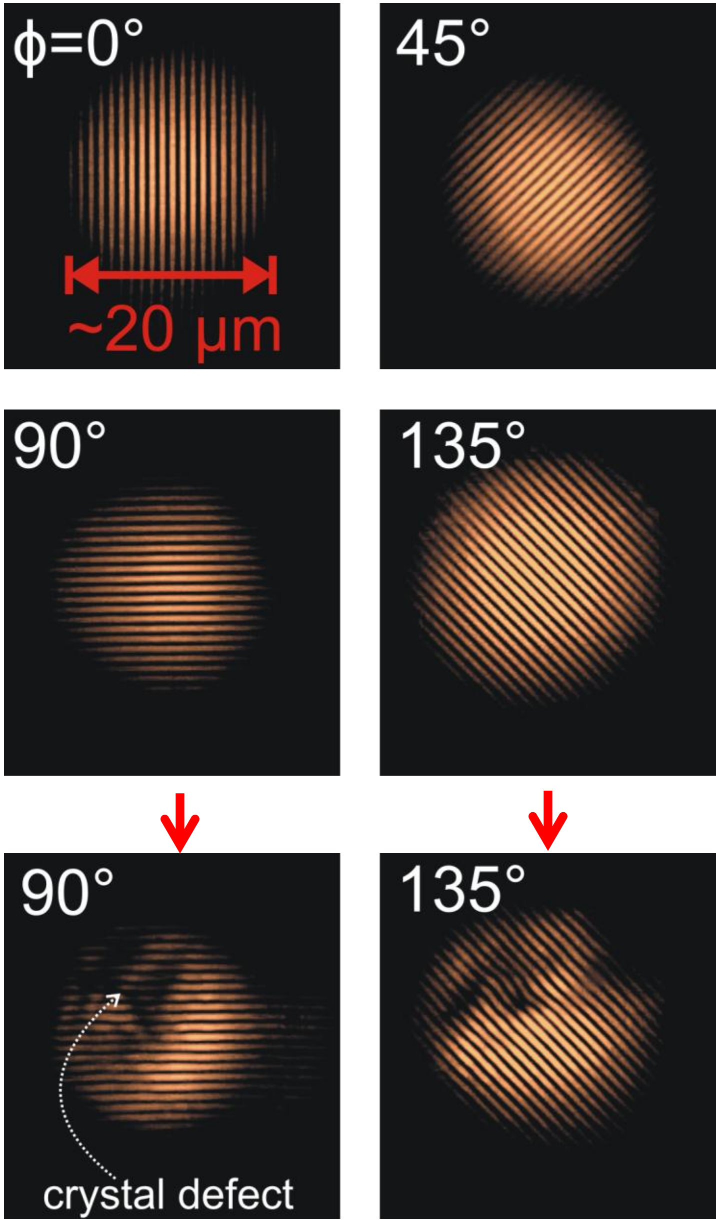


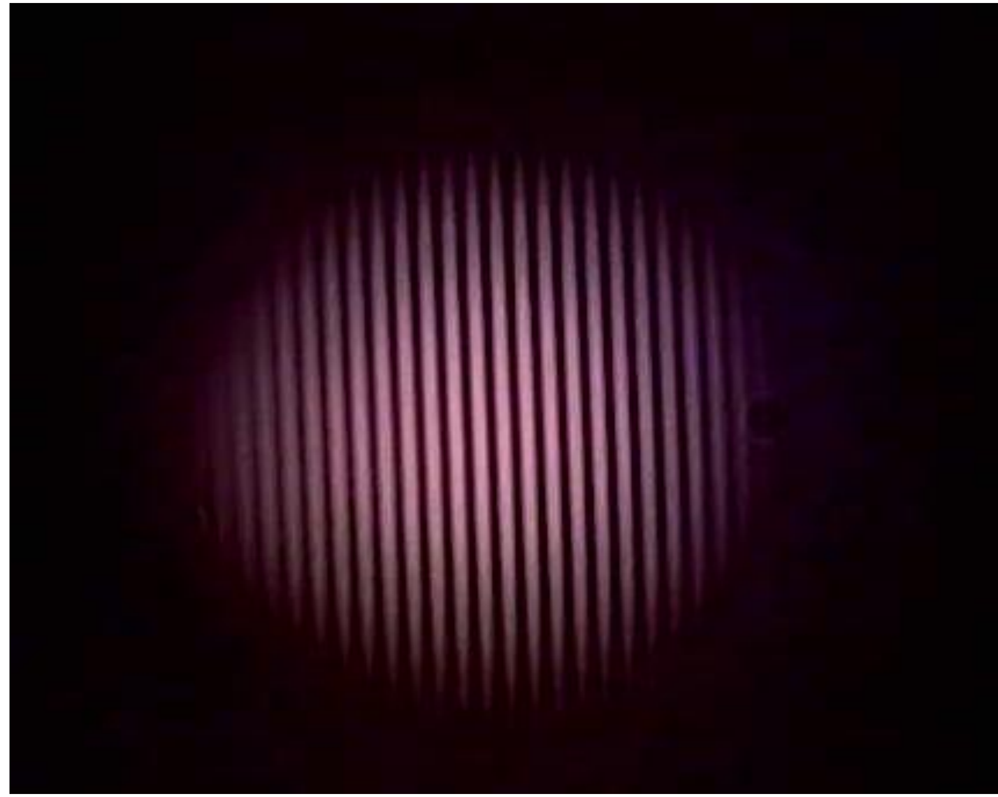
Sample is totally stationary.
Thus, we may perform measurements:

- in ultracold environments ✓
- in a magnetic field ✓
- on small single crystals ✓
- as a scanning experiment ✓

D. H. Torchinsky et al., Rev. Sci. Instrum. 85, 083102 (2014)

Characterization of spot size and local repeatability

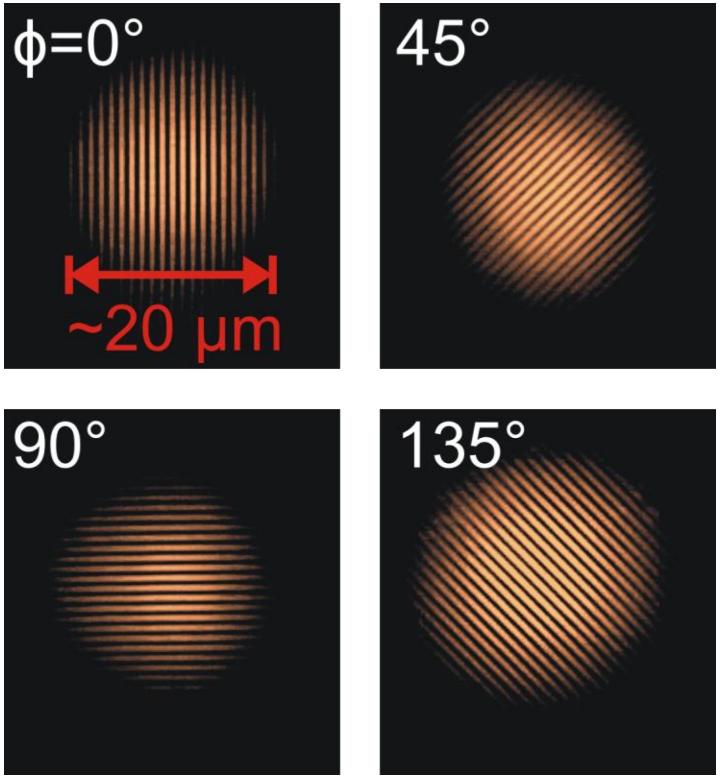




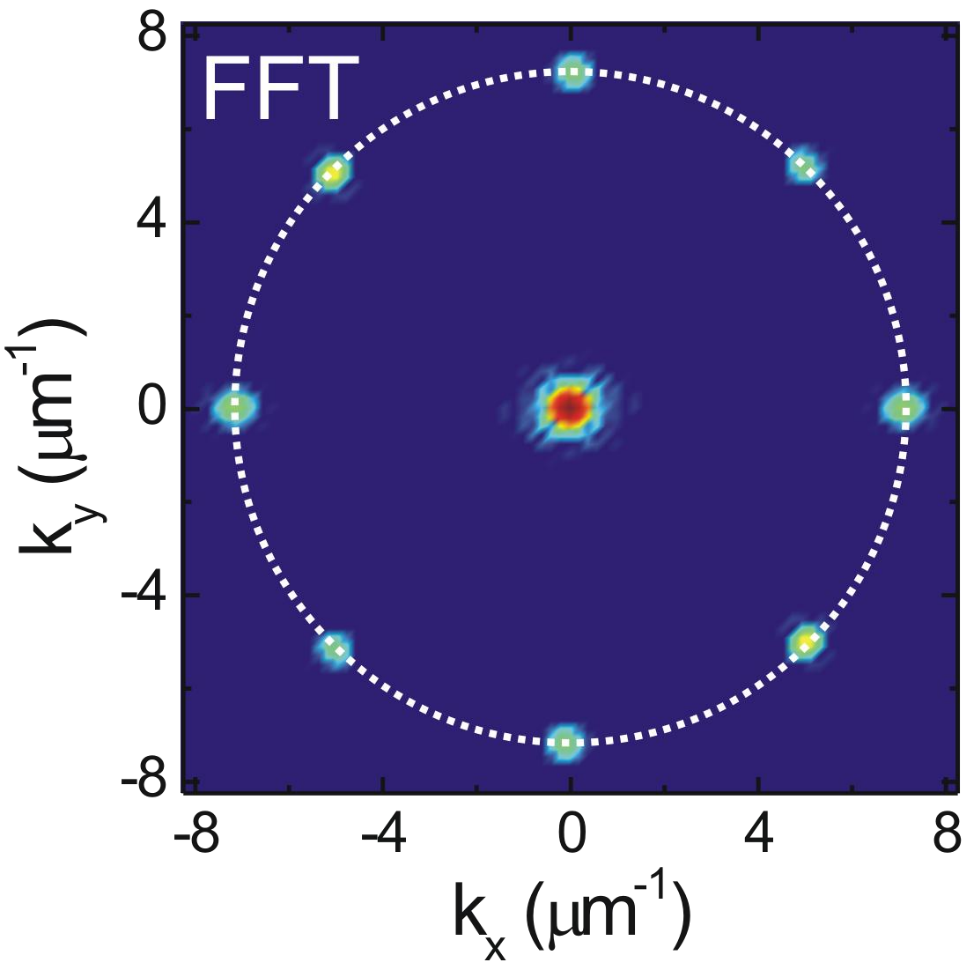
- Beam walking on sample ~ 1 μm
- Always measure the same portion of the sample ✓

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

Characterization of scattering angle precession



$$\Lambda = \frac{\lambda}{2\sin(\theta)} \sim 900 \text{ nm}$$

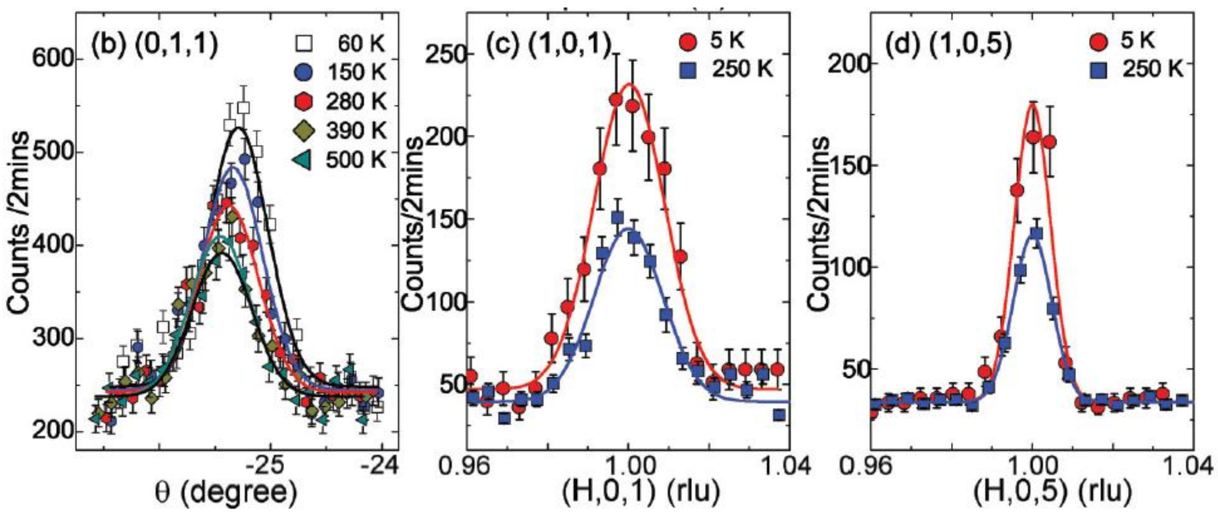


Precession of scattering plane with respect to surface normal < 0.05° ✓

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

Structural symmetry

Unexplained forbidden neutron peaks



- (1994 – 2013) space group $I4_1/acd$

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

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

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

- (2013) (h = 2n, 0, l = 2n) peaks observed

  Violation of $I4_1/acd$

  Structural defects?

  Spatial inhomogeneity?

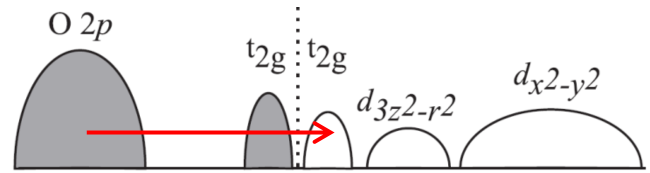
  Lower crystal symmetry?

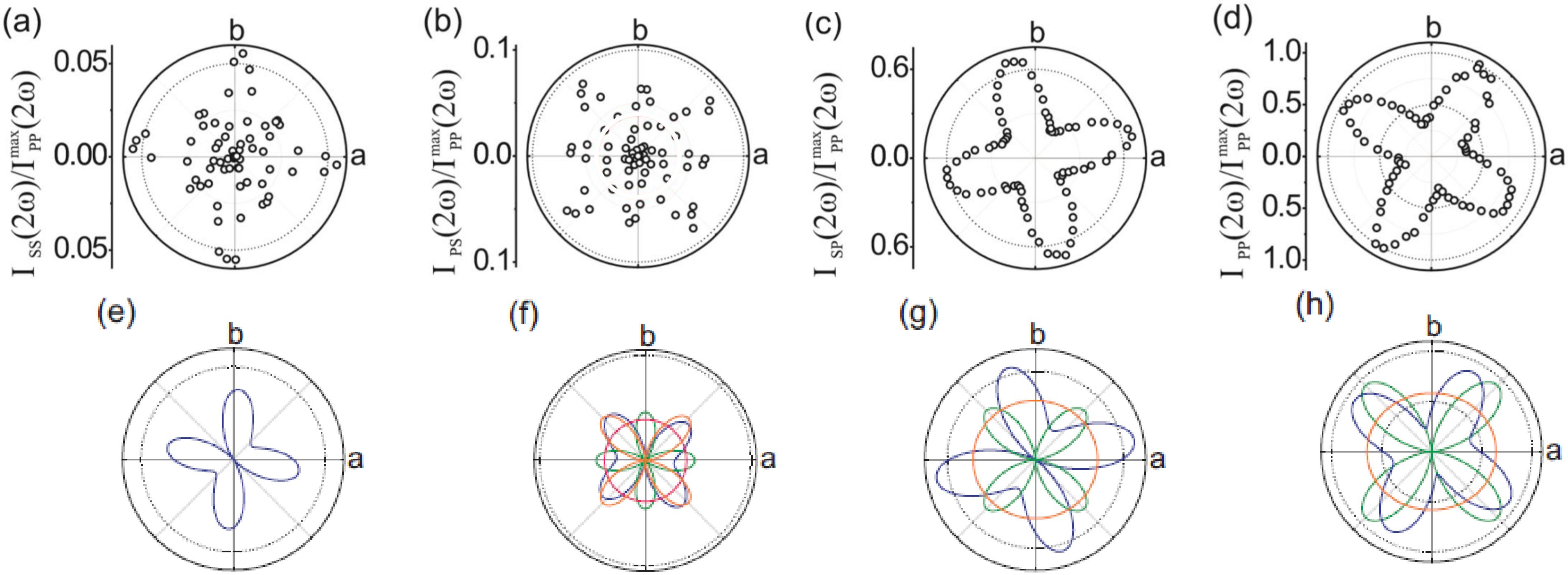
C. Dhital *et al*., PRB 87, 144405 (2013)

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

Second harmonic generation from Sr_2IrO_4 (001)

λ_{in} = 800 nm
λ_{out} = 400 nm





Proposed

Non-centrosymmetric (bulk ED)

C_s D_2 D_4

D. H. Torchinsky et al. PRL 114, 096404 (2015)

Second harmonic generation from Sr_2IrO_4 (001)

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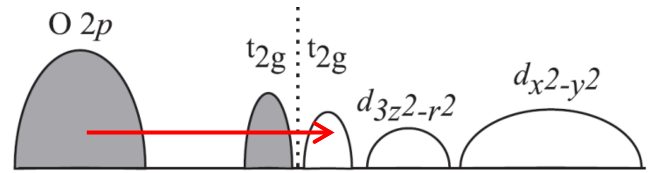
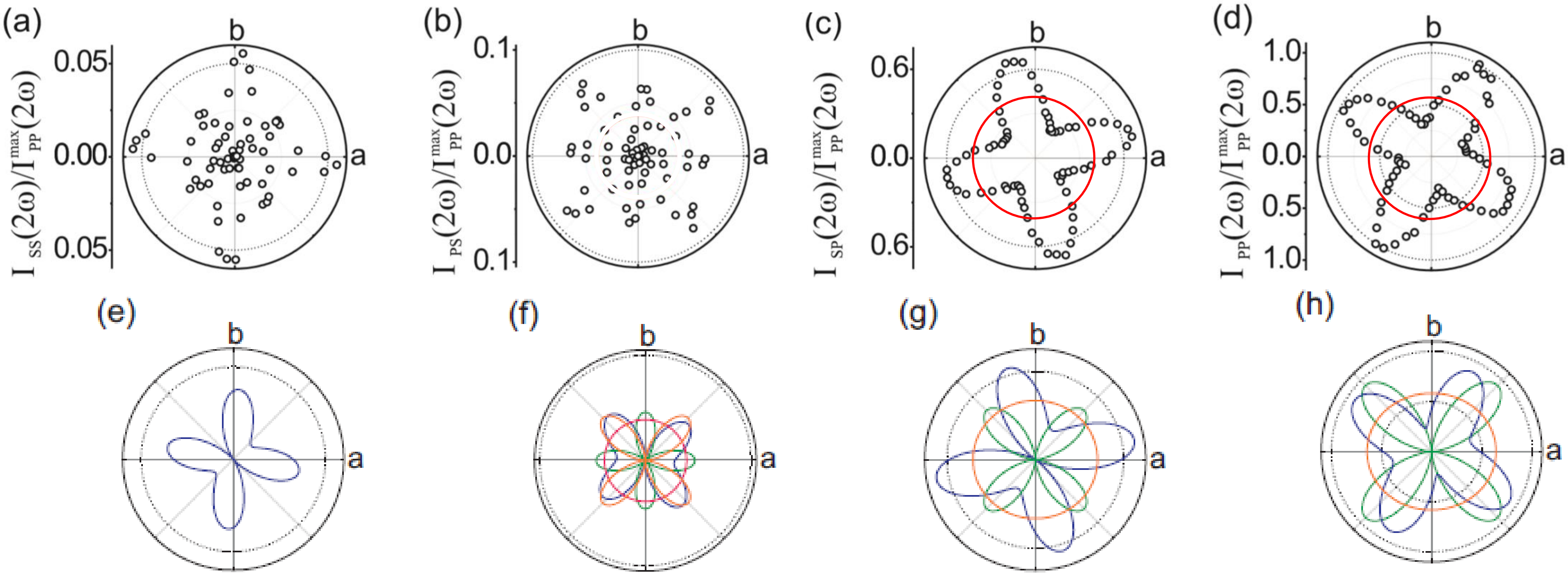




Centrosymmetric (surface ED) | Non-centrosymmetric (bulk ED)

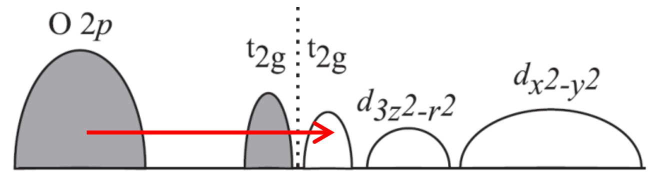
Proposed — D_{4h} — C_s — D_2 — D_4

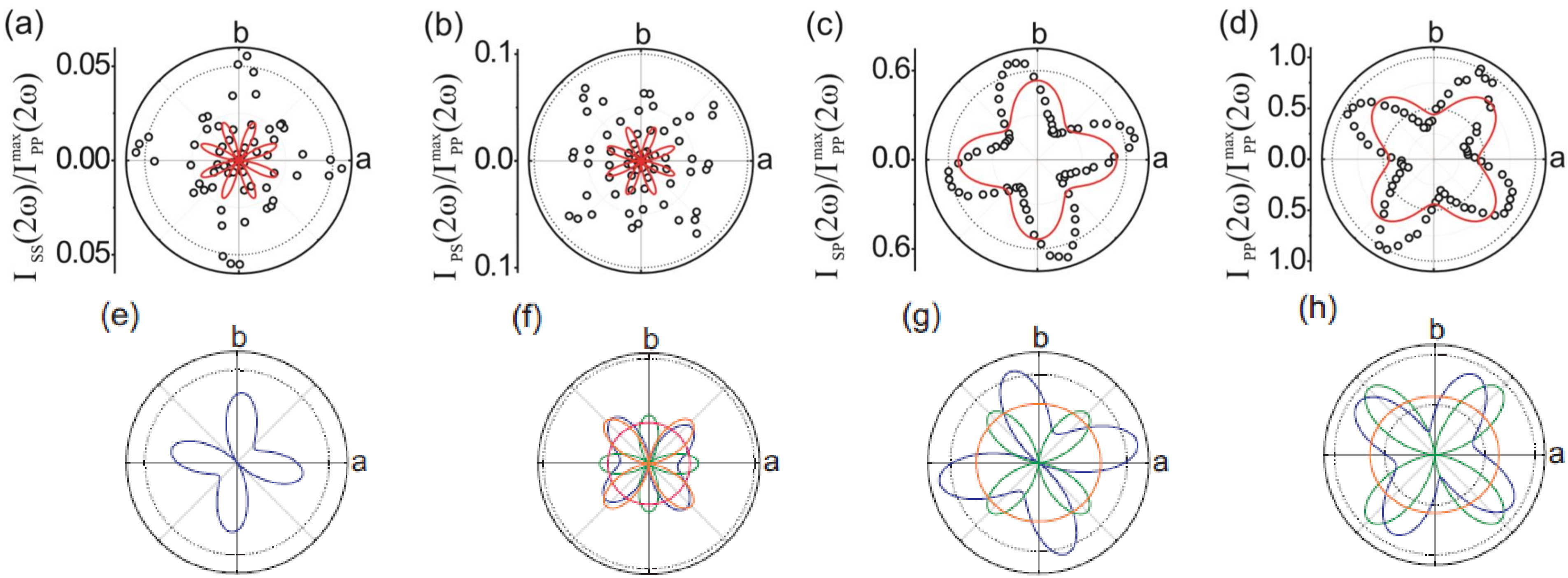
$I4_1/acd$

D. H. Torchinsky et al. PRL 114, 096404 (2015)

Second harmonic generation from Sr_2IrO_4 (001)

λ_{in} = 800 nm
λ_{out} = 400 nm





Centrosymmetric (bulk EQ)

Non-centrosymmetric (bulk ED)

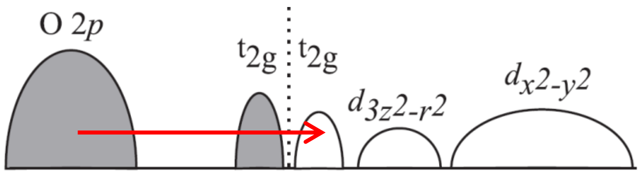
Proposed — D_{4h} — C_s — D_2 — D_4

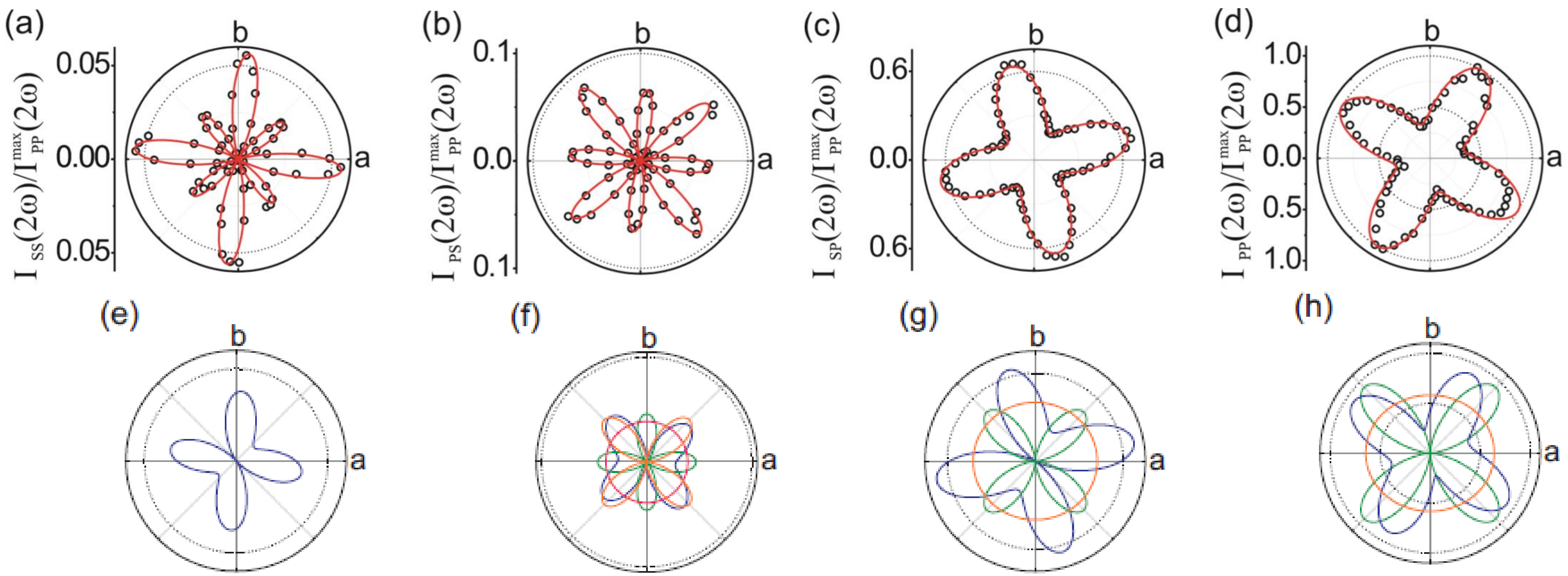
$I4_1/acd$

D. H. Torchinsky et al. PRL 114, 096404 (2015)

Second harmonic generation from Sr_2IrO_4 (001)

λ_{in} = 800 nm
λ_{out} = 400 nm





Centrosymmetric (bulk EQ)

Non-centrosymmetric (bulk ED)

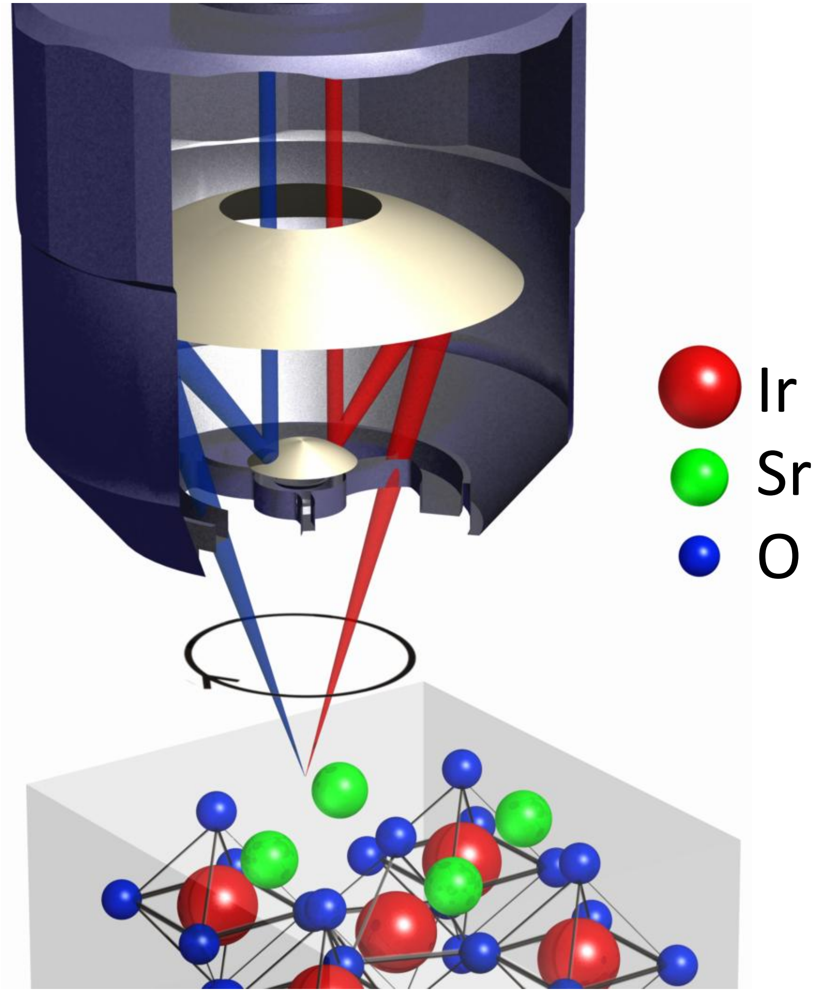
Proposed —— C_{4h}

—— C_s —— D_2 —— D_4

$I4_1/a$

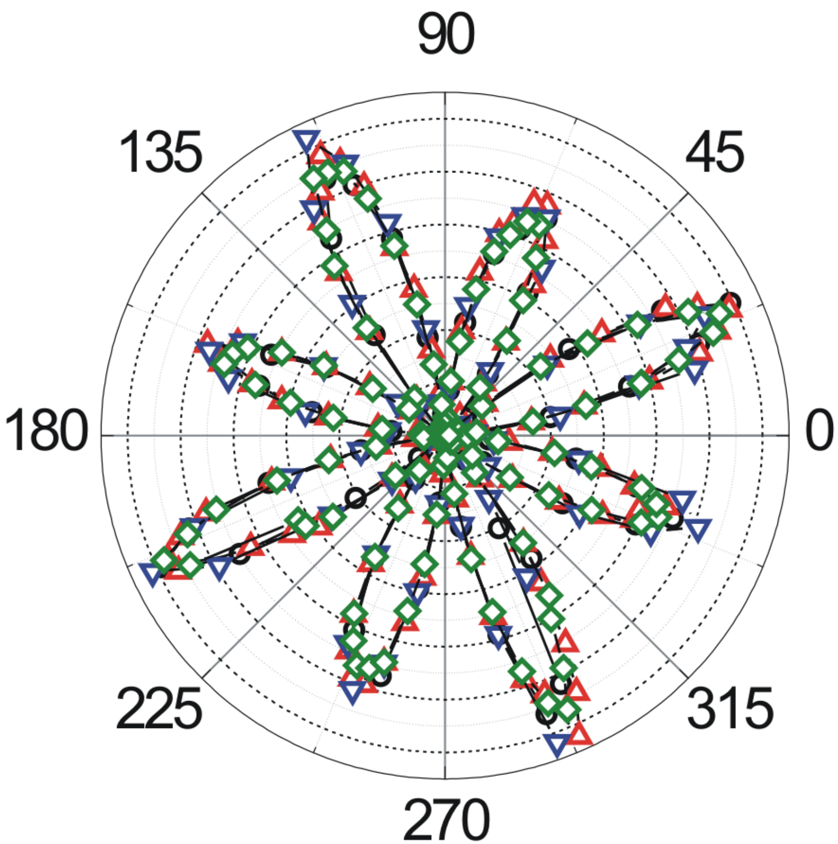
D. H. Torchinsky et al. PRL 114, 096404 (2015)

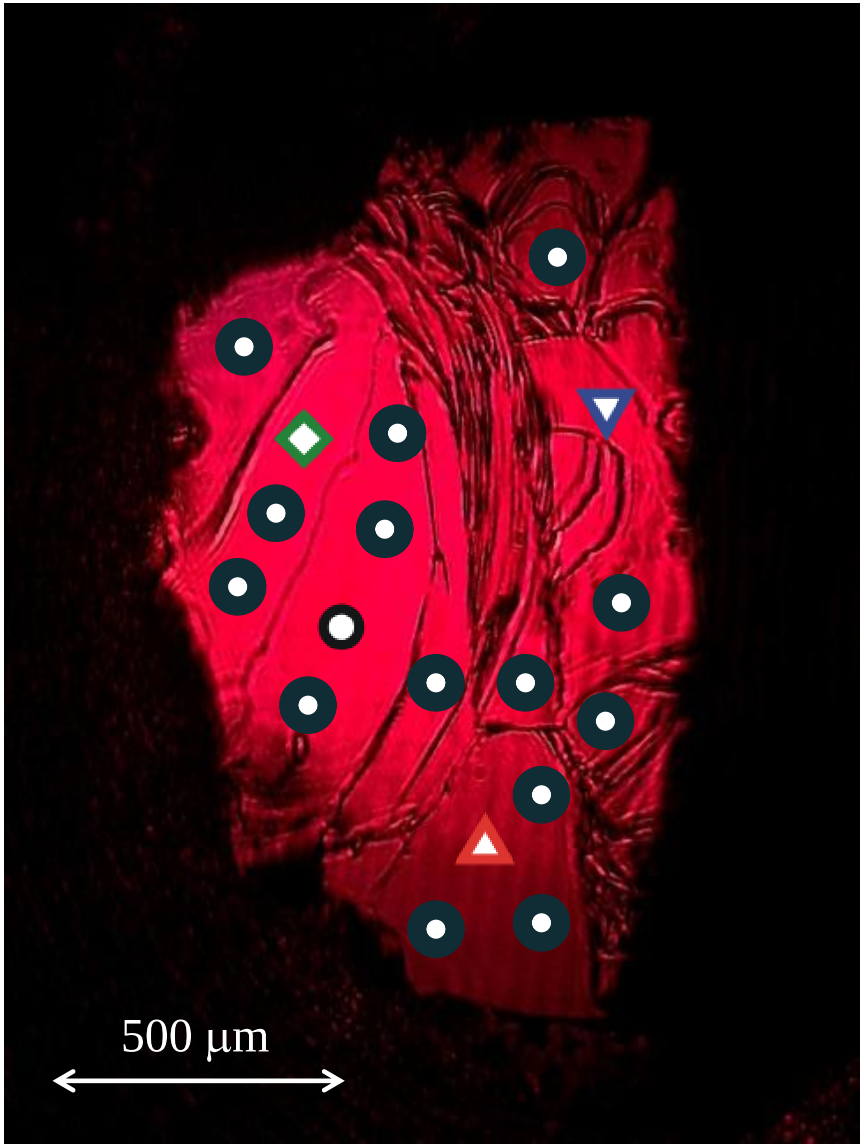
Nonlinear optical microscopy



D. H. Torchinsky et al. PRL 114, 096404 (2015)

Spatially resolved symmetry mapping

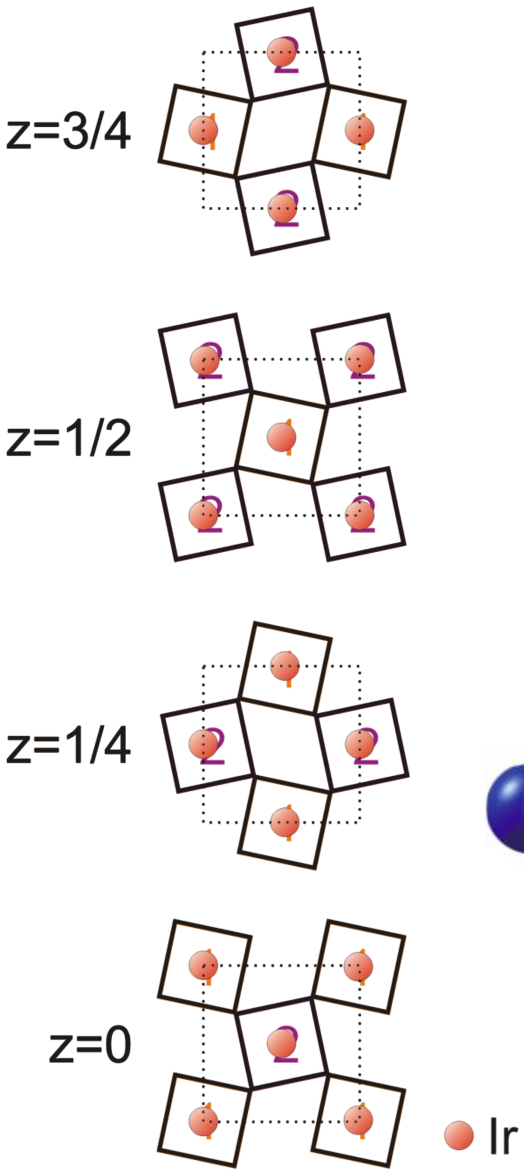




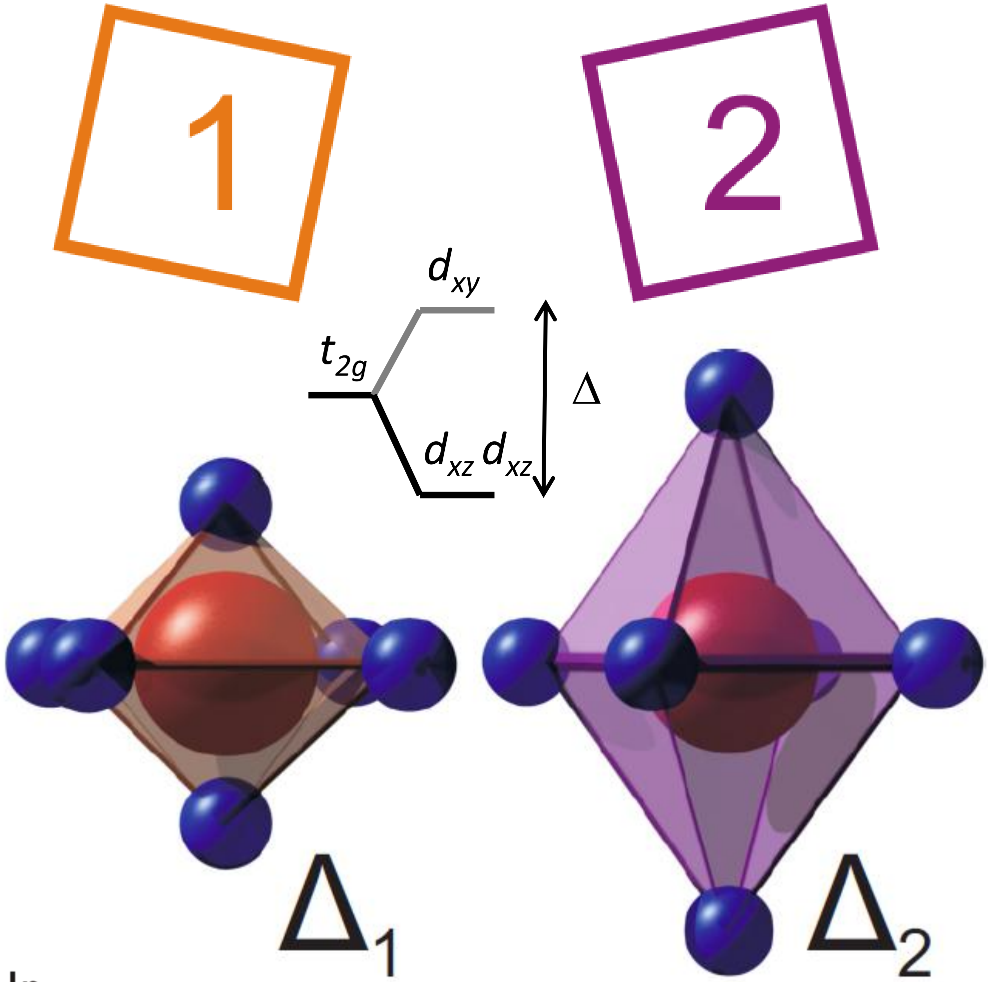
- No evidence of spatial inhomogeneity or parasitic phases

D. H. Torchinsky et al. PRL 114, 096404 (2015)

Lowered global symmetry $I4_1/acd \rightarrow I4_1/a$



Loss of c, d glide planes from $I4_1/acd \rightarrow I4_1/a$ occurs via staggered tetragonal distortion

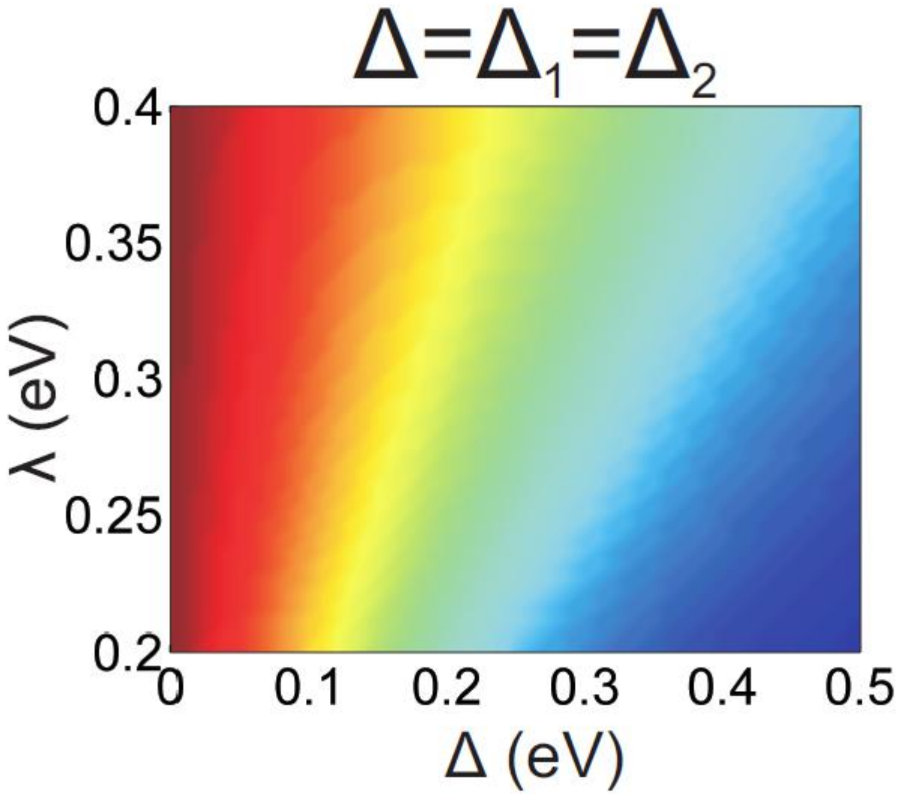


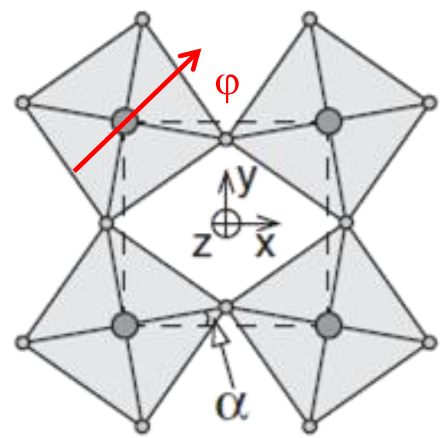
D. H. Torchinsky et al. PRL 114, 096404 (2015)

Consequences of lowered global symmetry

D. H. Torchinsky et al. PRL 114, 096404 (2015)

$$H = \Sigma_{n,n'} J \mathbf{S}_n \mathbf{S}_{n'} - D(S_n^x S_{n'}^y - S_n^y S_{n'}^x)$$
$$+\delta J_z S_n^z S_{n'}^z + \delta J_{xy} \left(\mathbf{S}_n \cdot \mathbf{r}_{n,n'}\right)\left(\mathbf{S}_{n'} \cdot \mathbf{r}_{n,n'}\right)$$



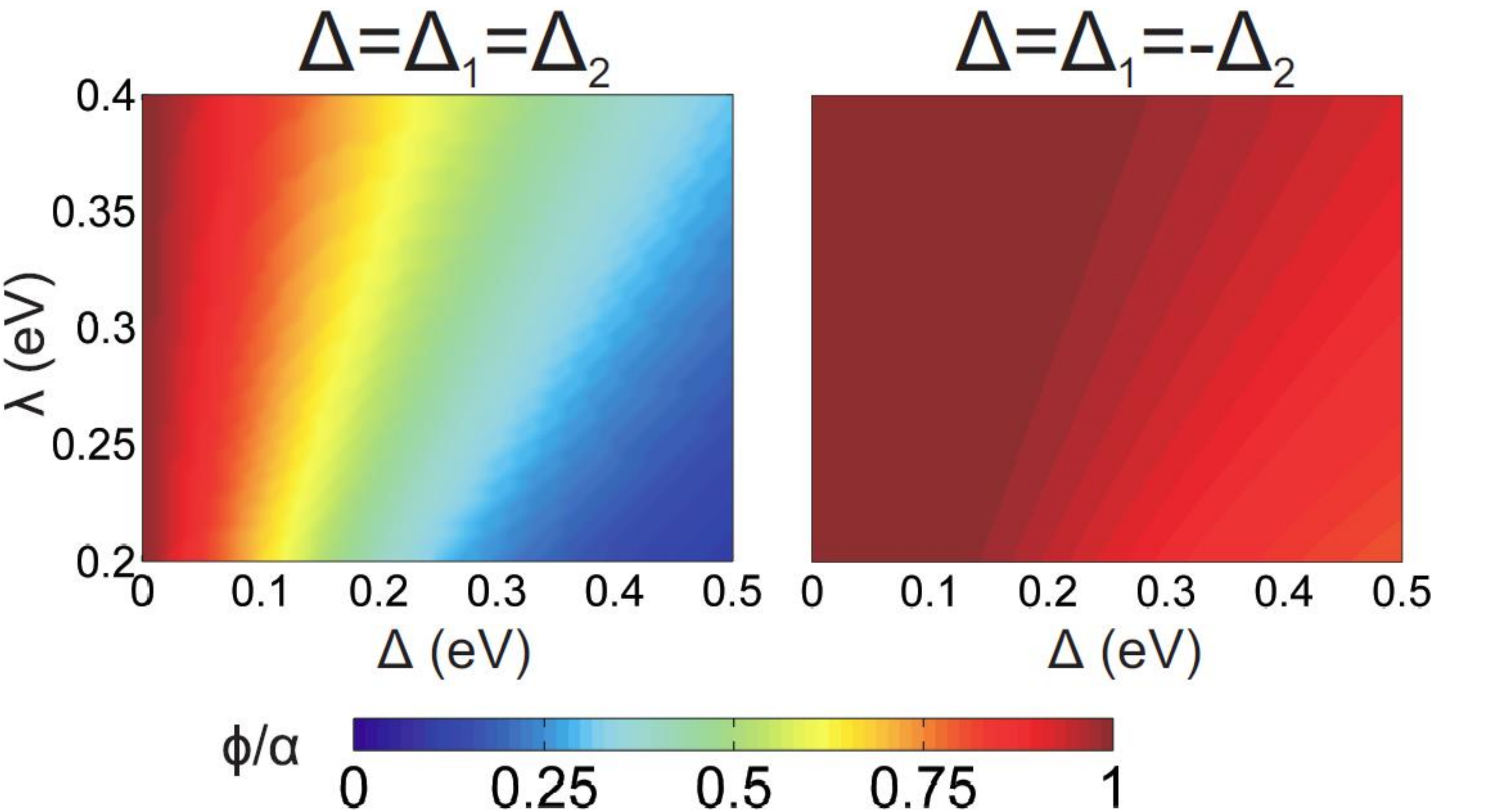


G. Jackeli and G. Khaliullin PRL 102, 017205 (2009)

Consequences of lowered global symmetry

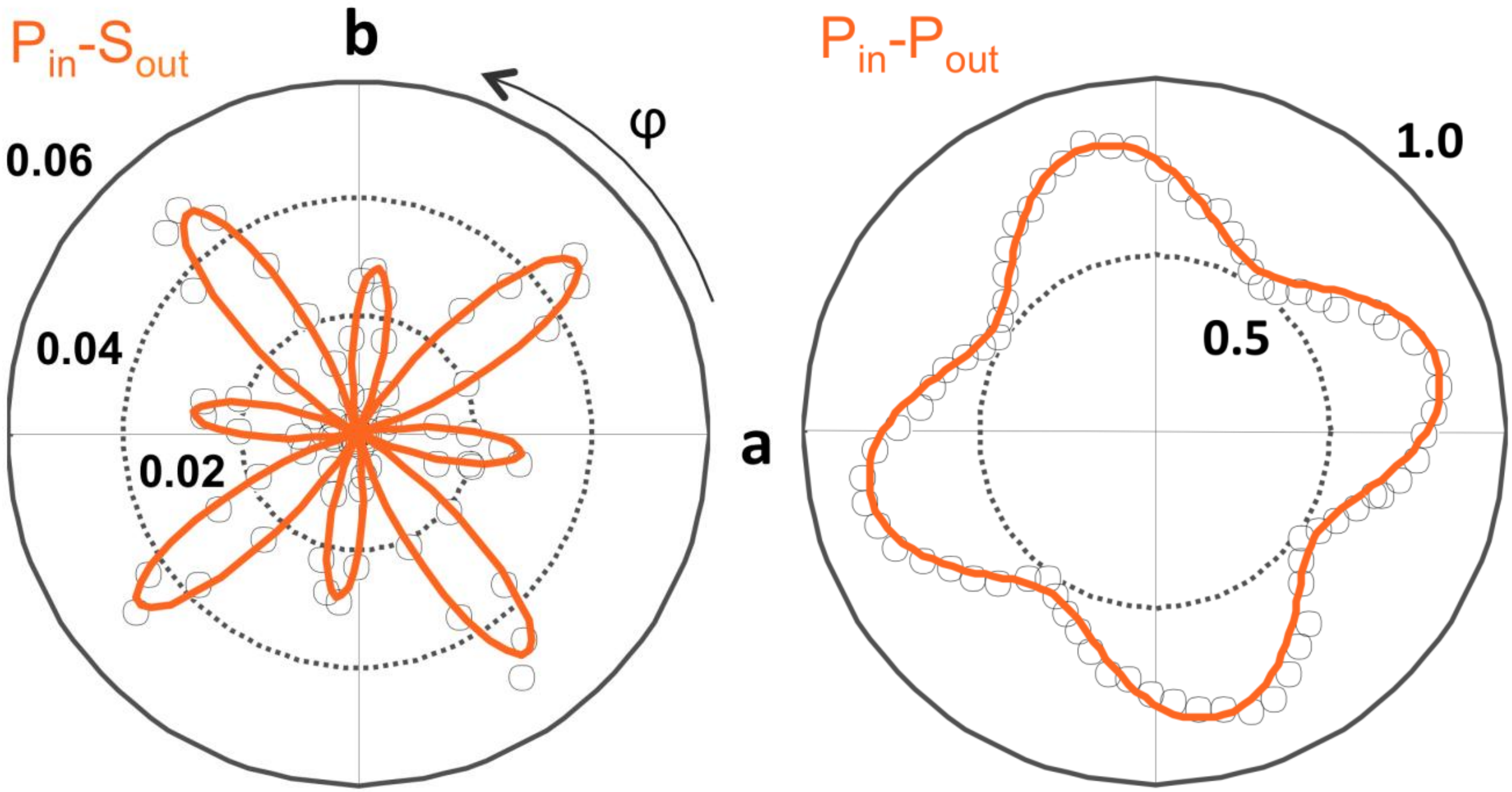
D. H. Torchinsky et al. PRL 114, 096404 (2015)

$$H = \Sigma_{n,n'} J \mathbf{S}_n \mathbf{S}_{n'} - D(S_n^x S_{n'}^y - S_n^y S_{n'}^x)$$
$$+\delta J_z S_n^z S_{n'}^z + \delta J_{xy} \left(\mathbf{S}_n \cdot \mathbf{r}_{n,n'}\right)\left(\mathbf{S}_{n'} \cdot \mathbf{r}_{n,n'}\right)$$



RA-SHG data at T = 295 K

L. Zhao et al. in review



χ^{EQ} E ∇E

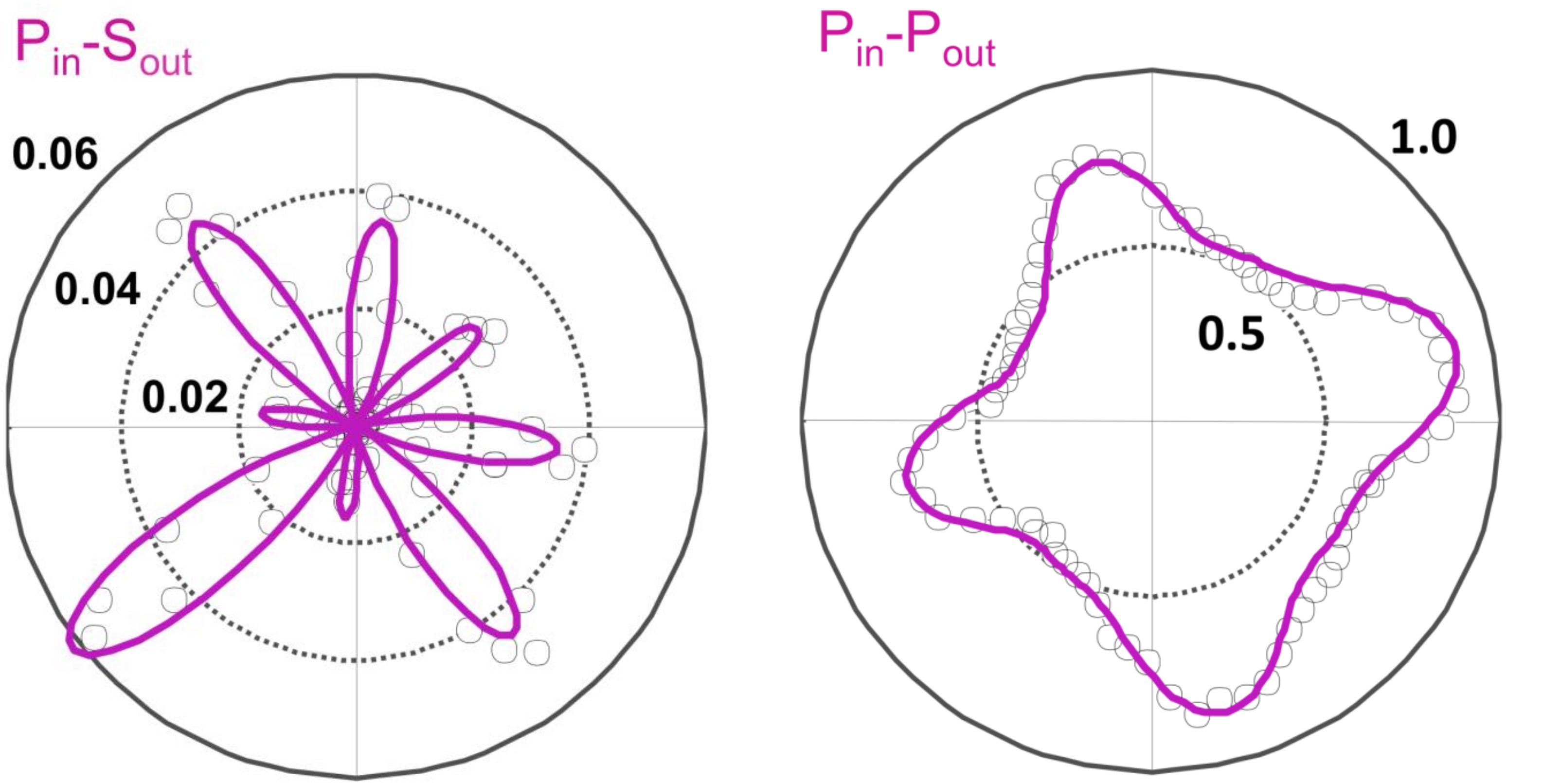
cryst. 4/m (C_{4h})

Inversion sym. ✓

Rotational sym. C_4

RA-SHG data at T = 170 K

L. Zhao et al. in review



χ^{EQ} E ∇E + χ^{ED} E E

cryst. 4/m

magn. 2′/m (m1′)

Inversion sym.

Rotational sym.

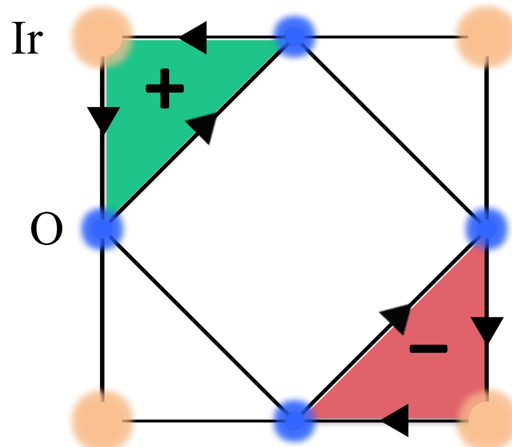
C_1

Symmetries of Θ_{II} loop current order (magneto-electric)

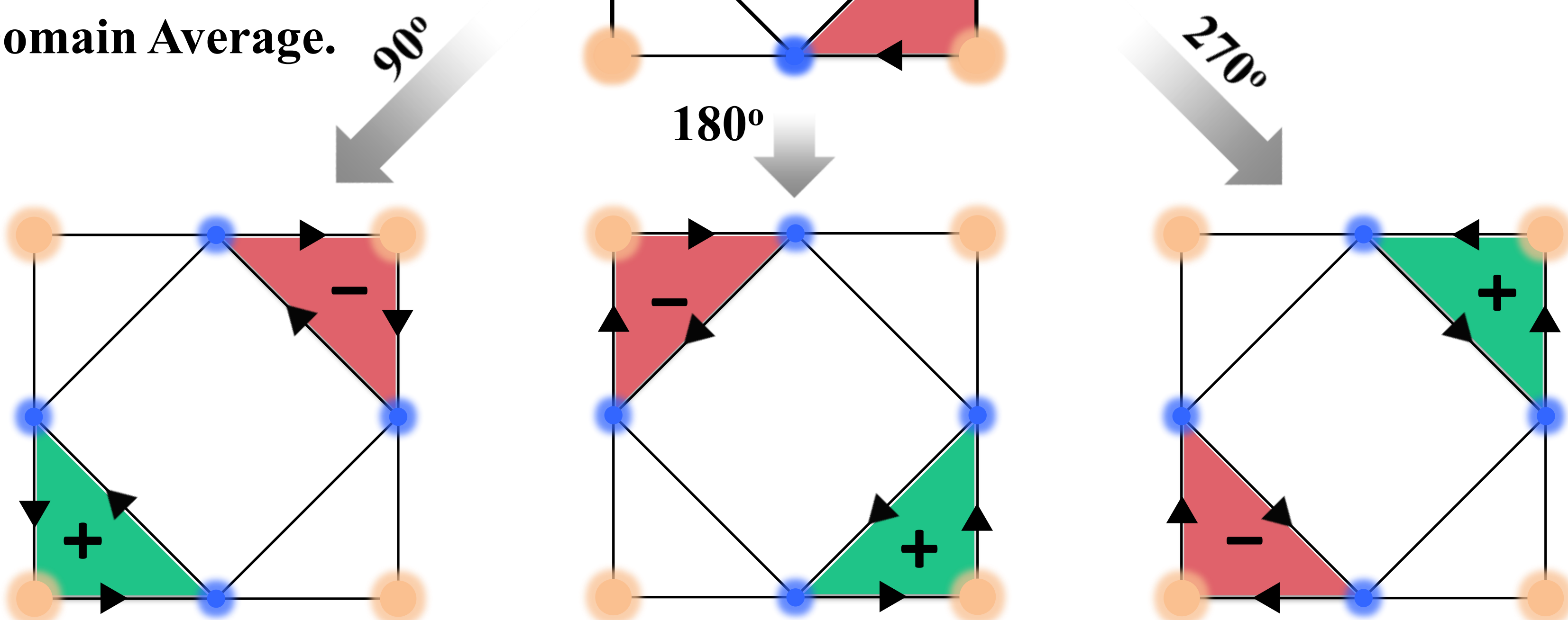
Properties: $\mathbf{C_4}$

- **Q = 0 ;**
- **No Net Magnetization per unit cell;**
- **Non-Dipolar;**
- **Domain Average.**

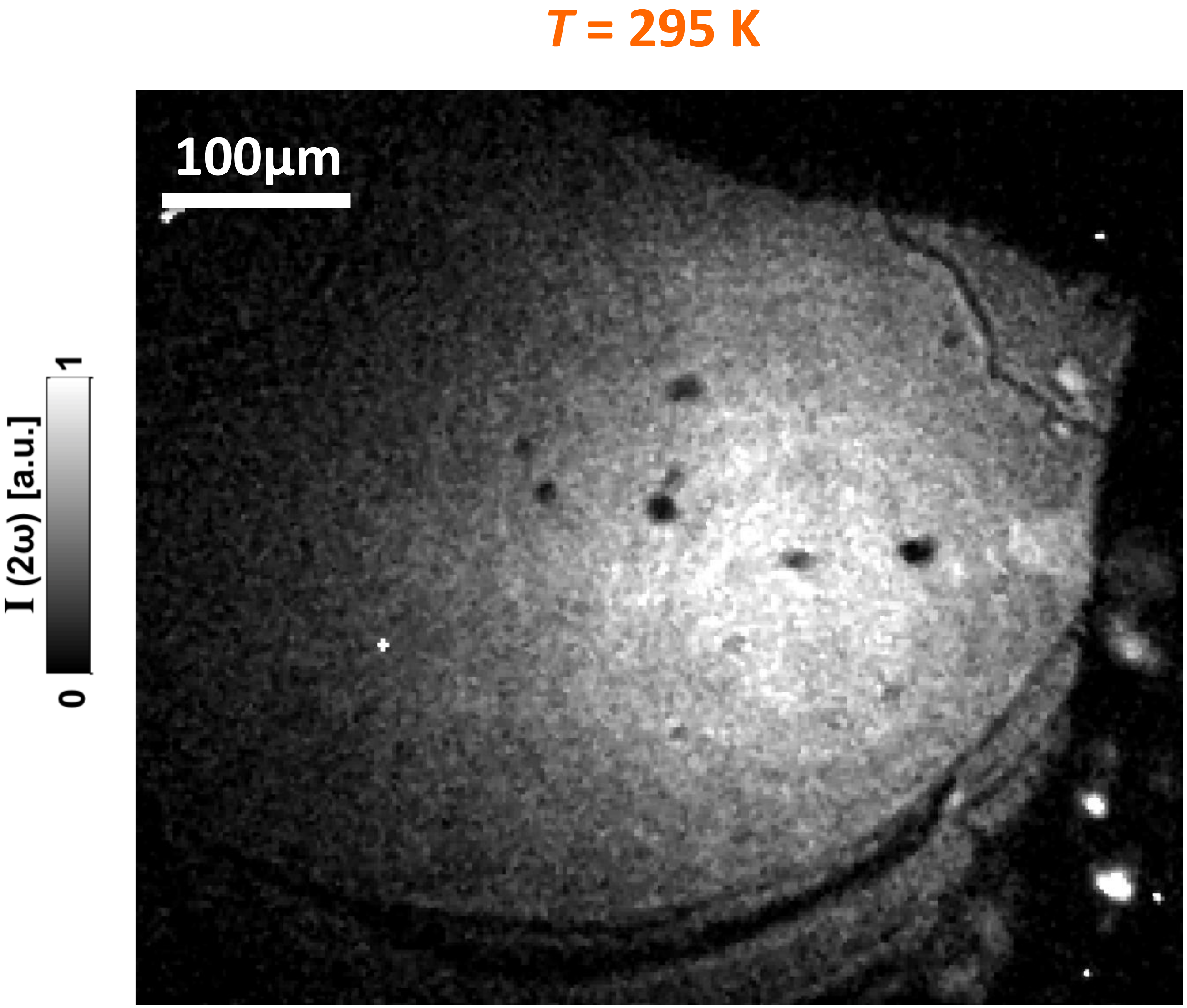
C_1 & Broken Inv. & T. R.



Ref:
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 Physica B, **460**, 159 (2015)



Nonlinear optical microscopy images



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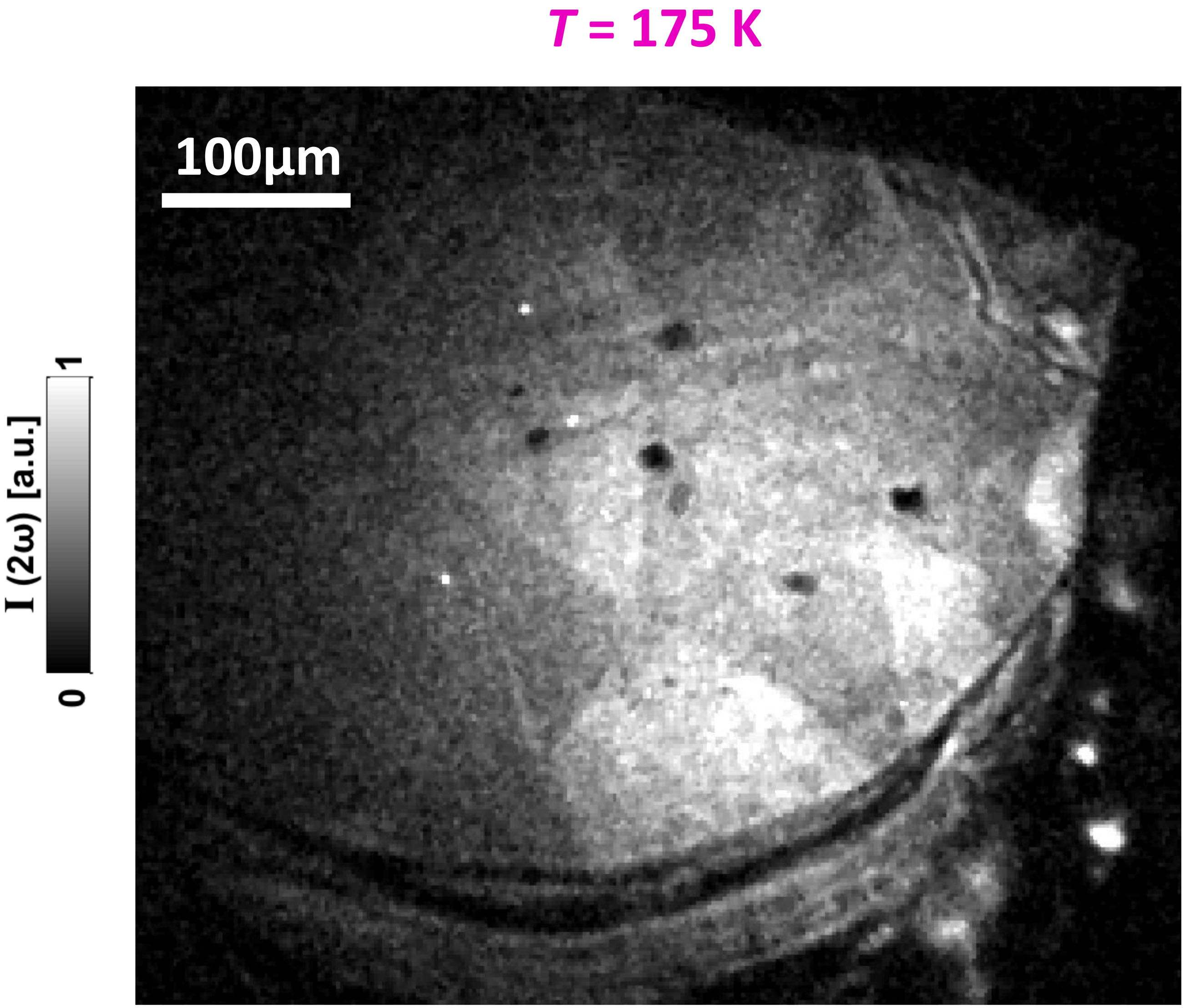
Nonlinear optical microscopy images



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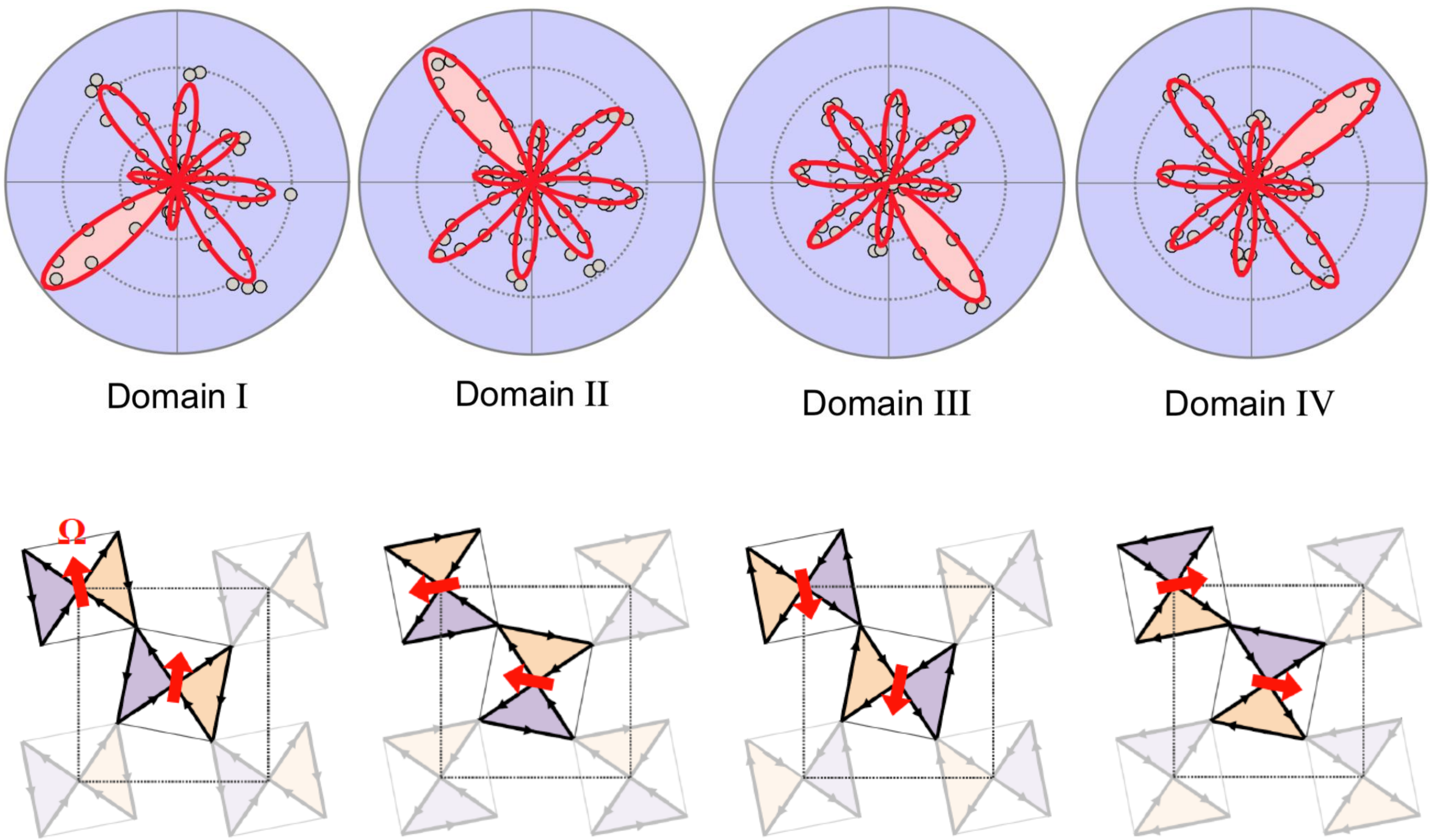
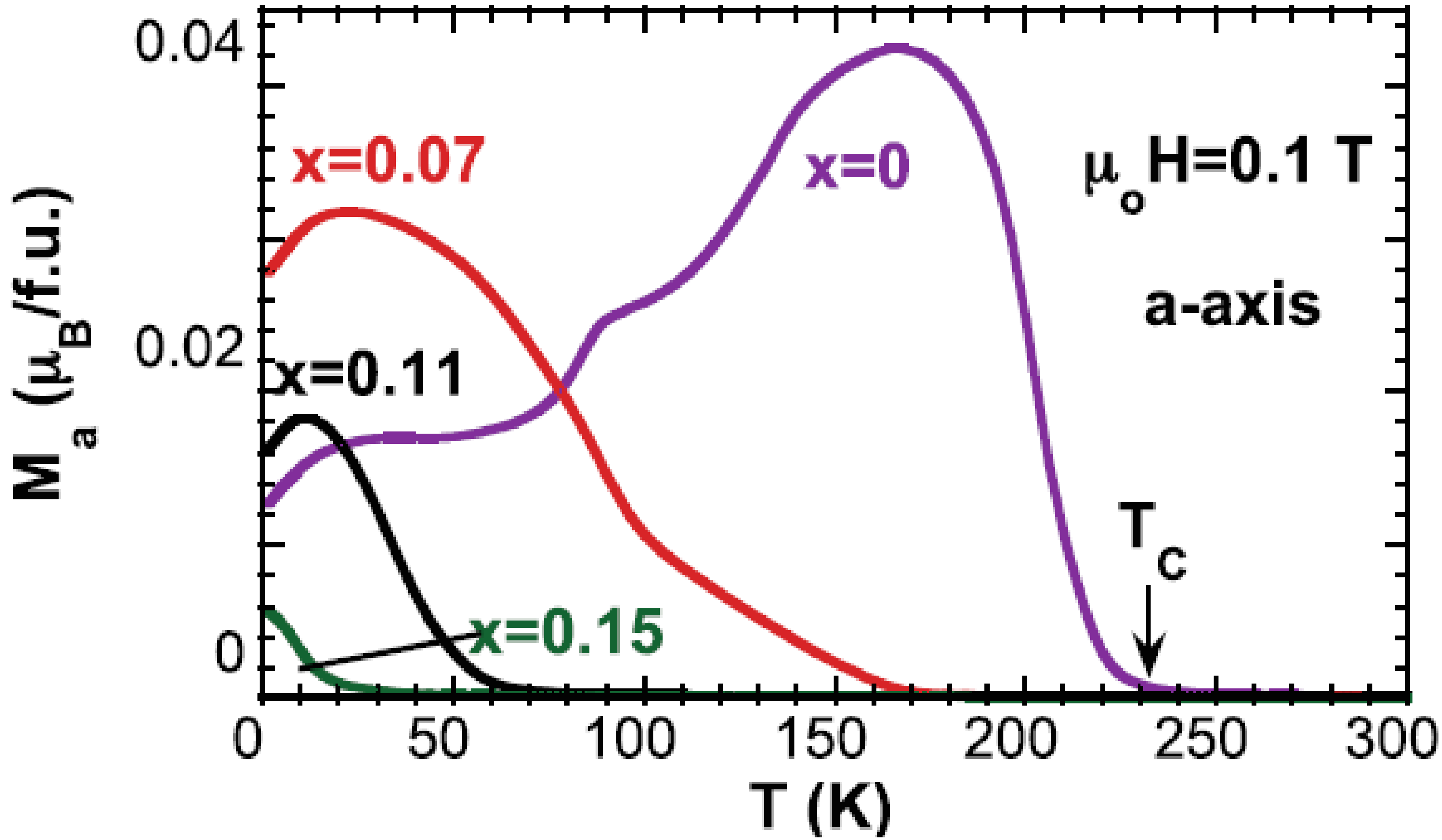
Hidden order domain orientations in Sr_2IrO_4

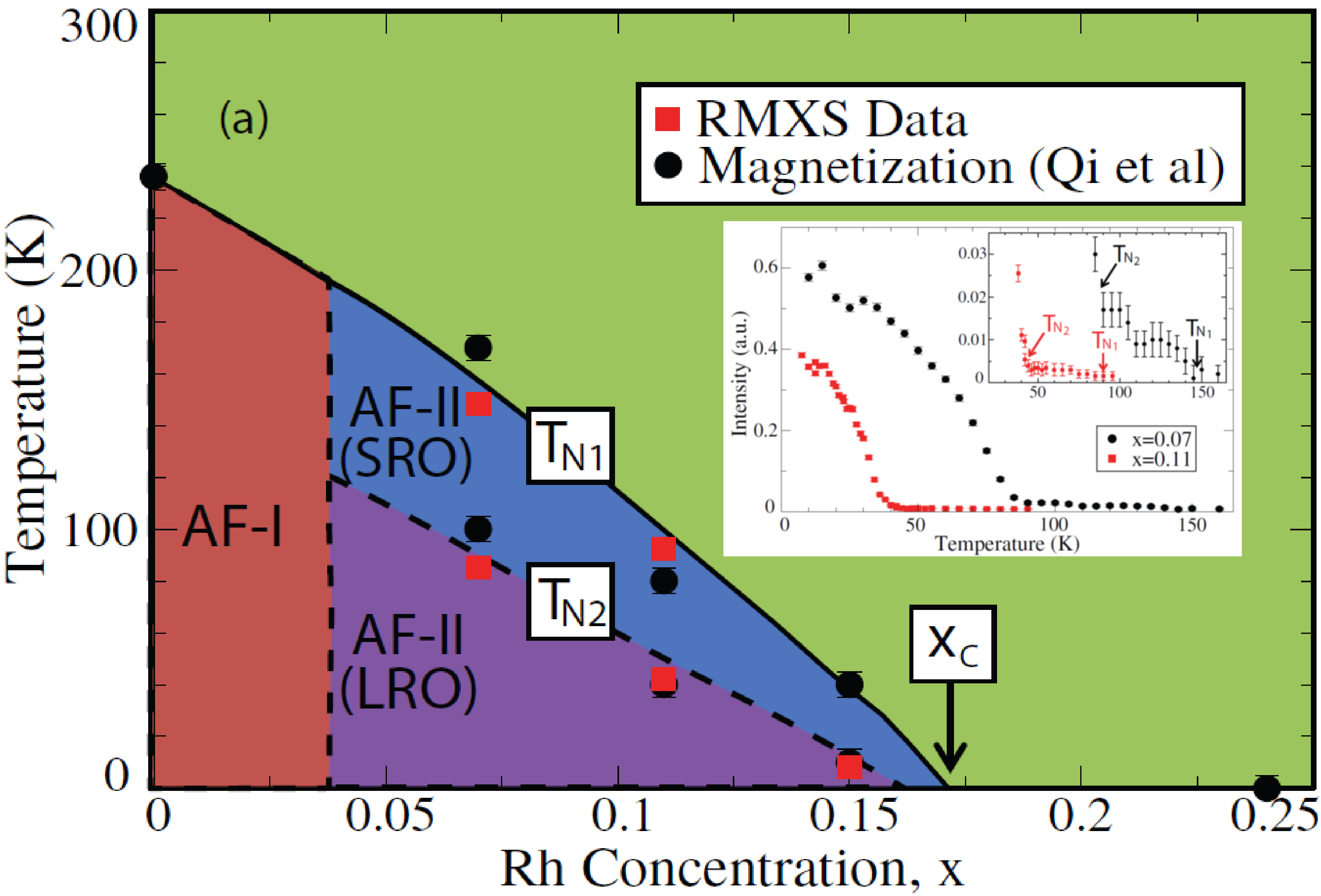


Hole doped $Sr_2Ir_{1-x}Rh_xO_4$



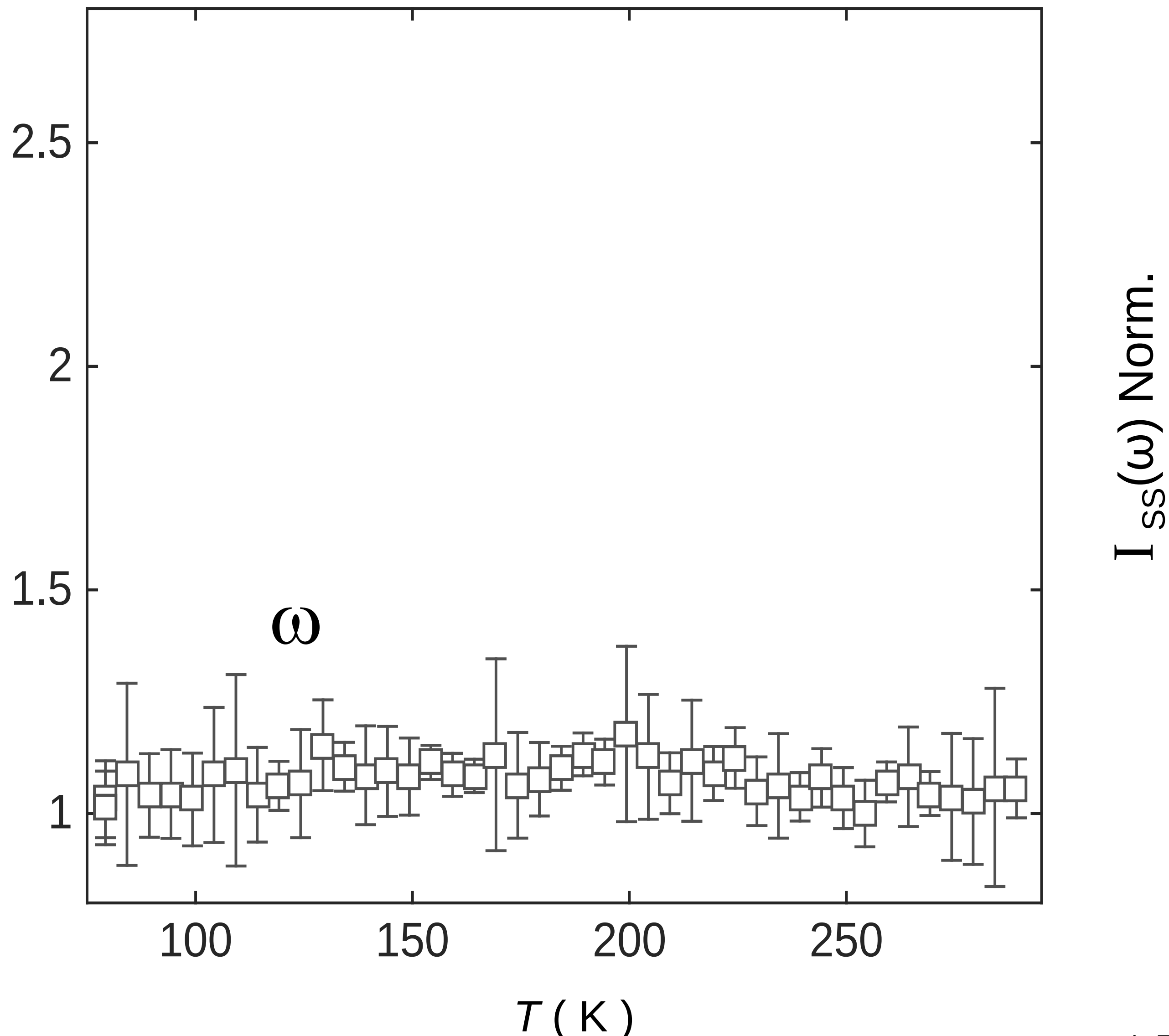
T. Qi *et al.*, PRB 86, 125105 (2012)

Hole doped $Sr_2Ir_{1-x}Rh_xO_4$



J. P. Clancy *et al.*, PRB 89, 054409 (2014)

Hidden symmetry breaking below T_Ω



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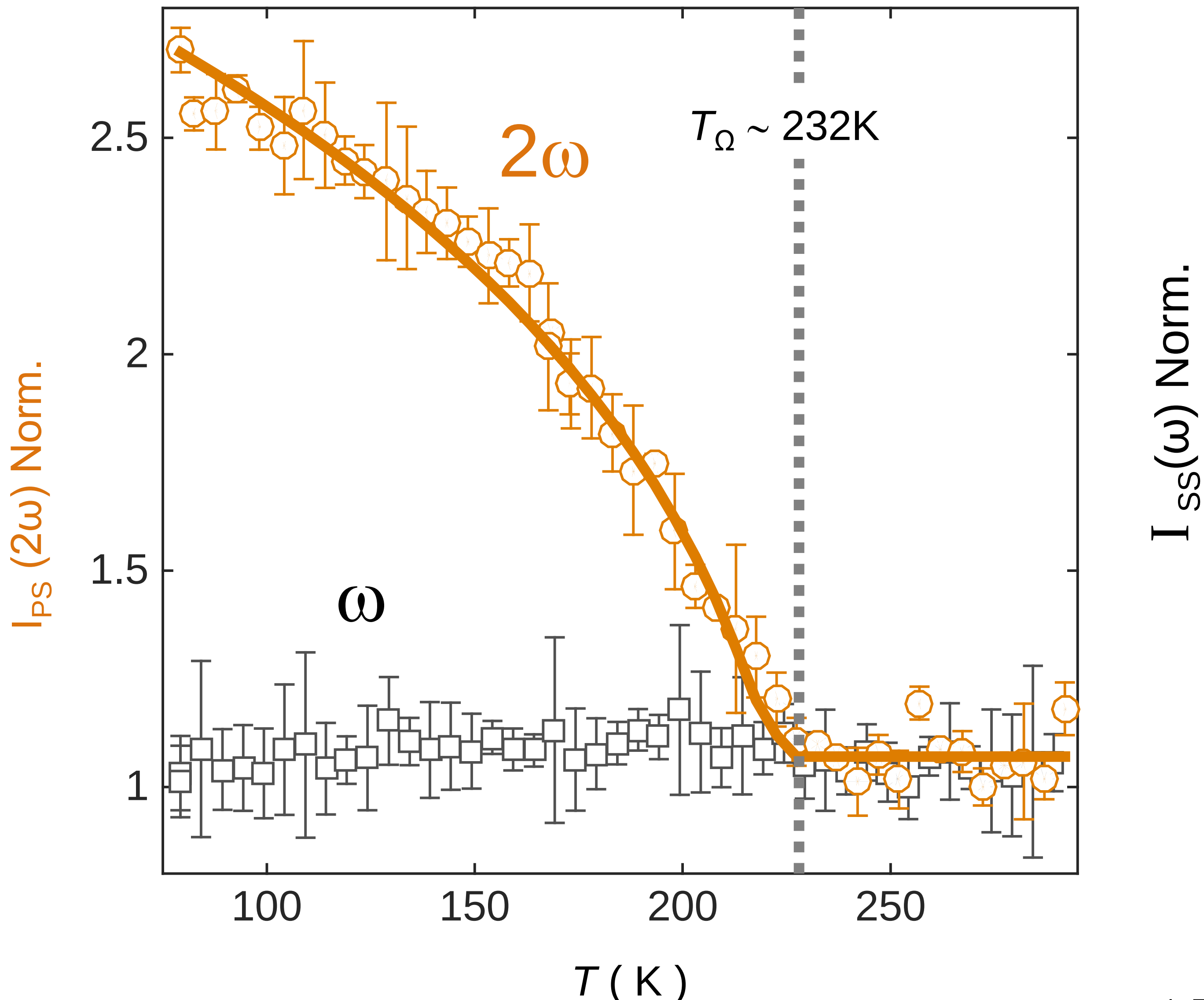
Hidden symmetry breaking below T_Ω



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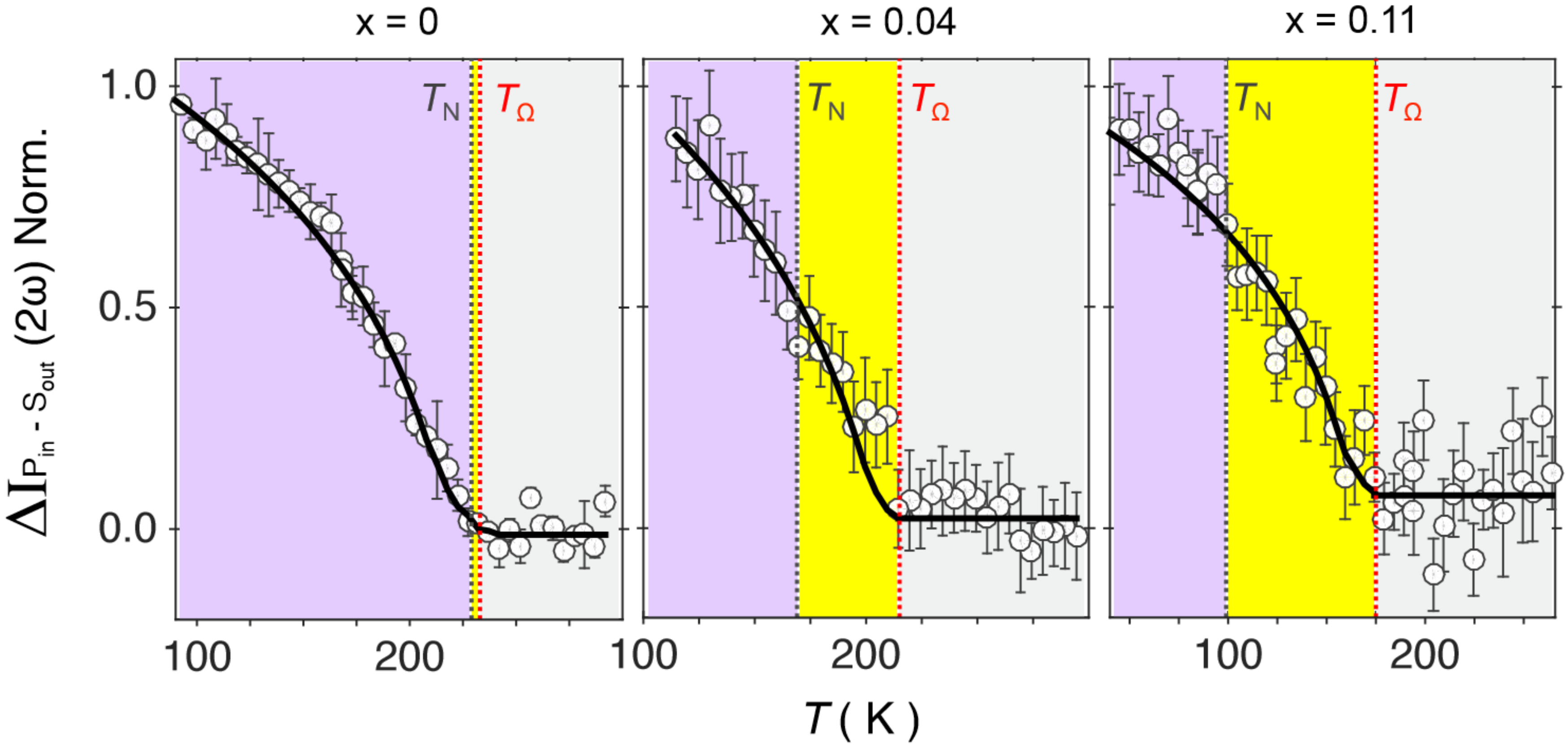
Doping dependence of T_Ω in $Sr_2Ir_{1-x}Rh_xO_4$



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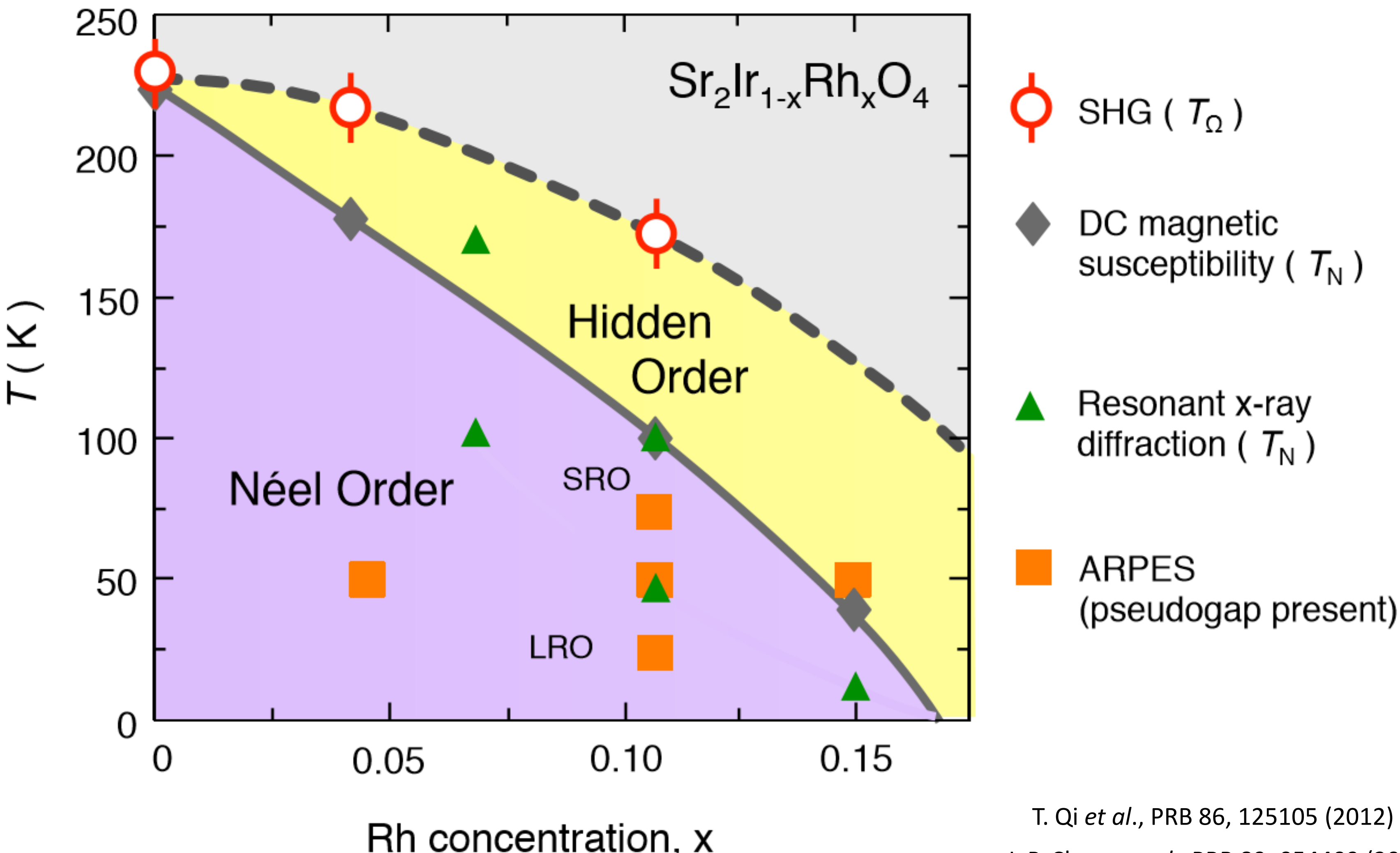
Exotic phases in iridates?



T. Qi *et al.*, PRB 86, 125105 (2012)

J. P. Clancy *et al.*, PRB 89, 054409 (2014)

L. Zhao et al. in review

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

Conclusions and Outlook

- Nonlinear optical response is an effective probe of bulk structural and electronic symmetry breaking.

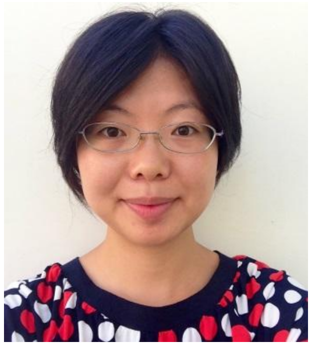
- Complementary to neutron and (non-) resonant x-ray diffraction. Small crystals ✓ Spatial resolution ✓ Multipolar order parameters ✓ Strong neutron absorbers (e.g. Ir) ✓

- Lower global structural symmetry revealed in Sr_2IrO_4. Supports perfect magneto-elastic locking.

- Hidden parity-odd magnetic phase revealed in parent and doped Sr_2IrO_4 consistent with Θ_{II} loop-current symmetry. Not trivially tied to Neel order.

- Possible relationship to pseudogap temperature?

Acknowledgements



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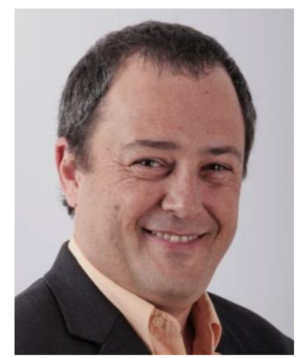

Prof. Ron Lifshitz