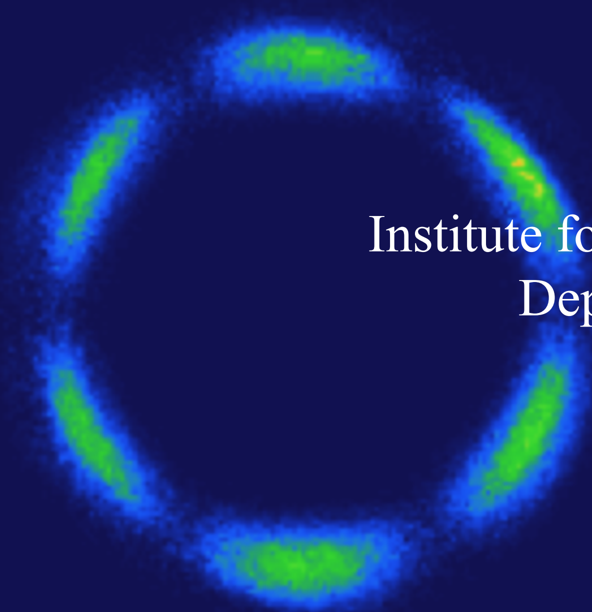
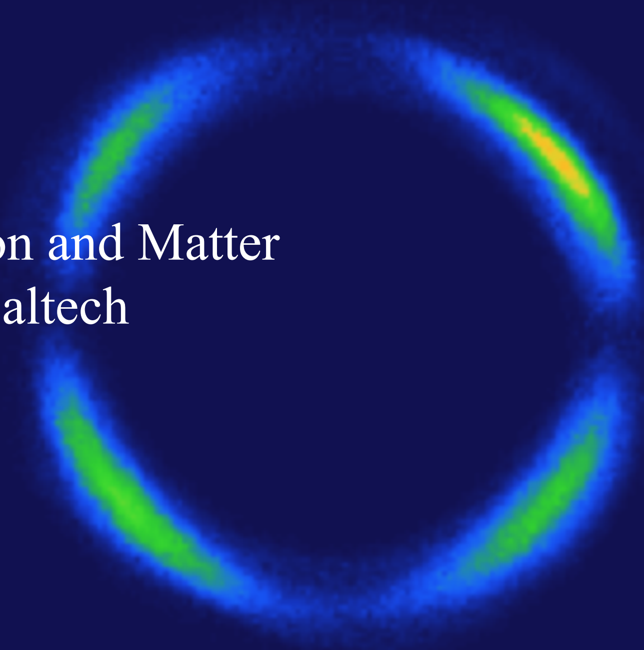
Signatures of a Multipolar Nematic Fluid in $Cd_2Re_2O_7$





David Hsieh
Institute for Quantum Information and Matter
Department of Physics, Caltech

KITP Conference
7/31/2017

Outline

Part One

Second harmonic optical rotational anisotropy

Part Two

Hidden order in the pseudogap region of doped Sr_2IrO_4 and $YBa_2Cu_3O_y$

Part Three

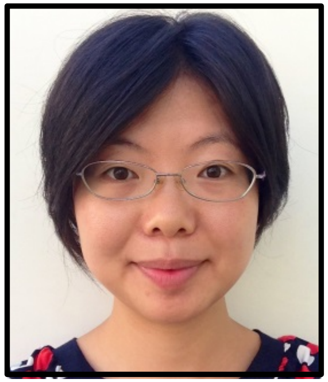
A parity-breaking multipolar nematic phase in $Cd_2Re_2O_7$

Part Four

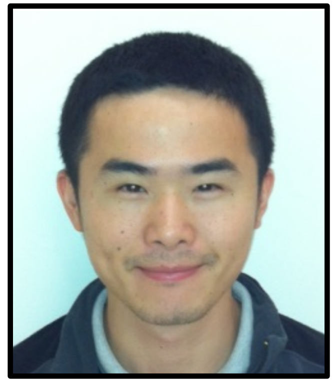
Evidence of an improper displacive phase transition in $Cd_2Re_2O_7$

Acknowledgements

John Harter
(now UCSB)



Liuyan Zhao
(now U. Michigan)



Hao Chu

Alberto de la Torre

Darius Torchinsky
(now Temple U.)

Sr_2IrO_4

C. U. Boulder
Gang Cao

Iowa State U.
Rebecca Flint

U. Minnesota
Natalia Perkins
Yuriy Sizyuk

U. Tel Aviv
Ron Lifshitz

$YBa_2Cu_3O_y$

UBC
Ruixing Liang
Doug Bonn
Walter Hardy

Johns Hopkins
Peter Armitage

$Cd_2Re_2O_7$

U. T. Knoxville
David Mandrus
Jiaqiang Yan
Zhiying Zhao

Columbia U.
Dante Kennes
Andy Millis

Caltech
Carina Belvin, Lauren Niu
Anthony Woss, Seva Ivanov

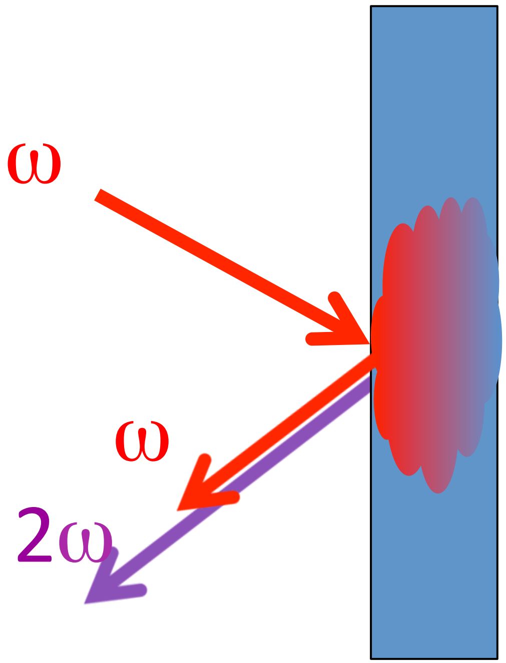
Part One

Second harmonic optical rotational anisotropy

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

J. Harter *et al*., Opt. Lett. **40**, 4671 (2015)

Optical second harmonic generation (SHG)



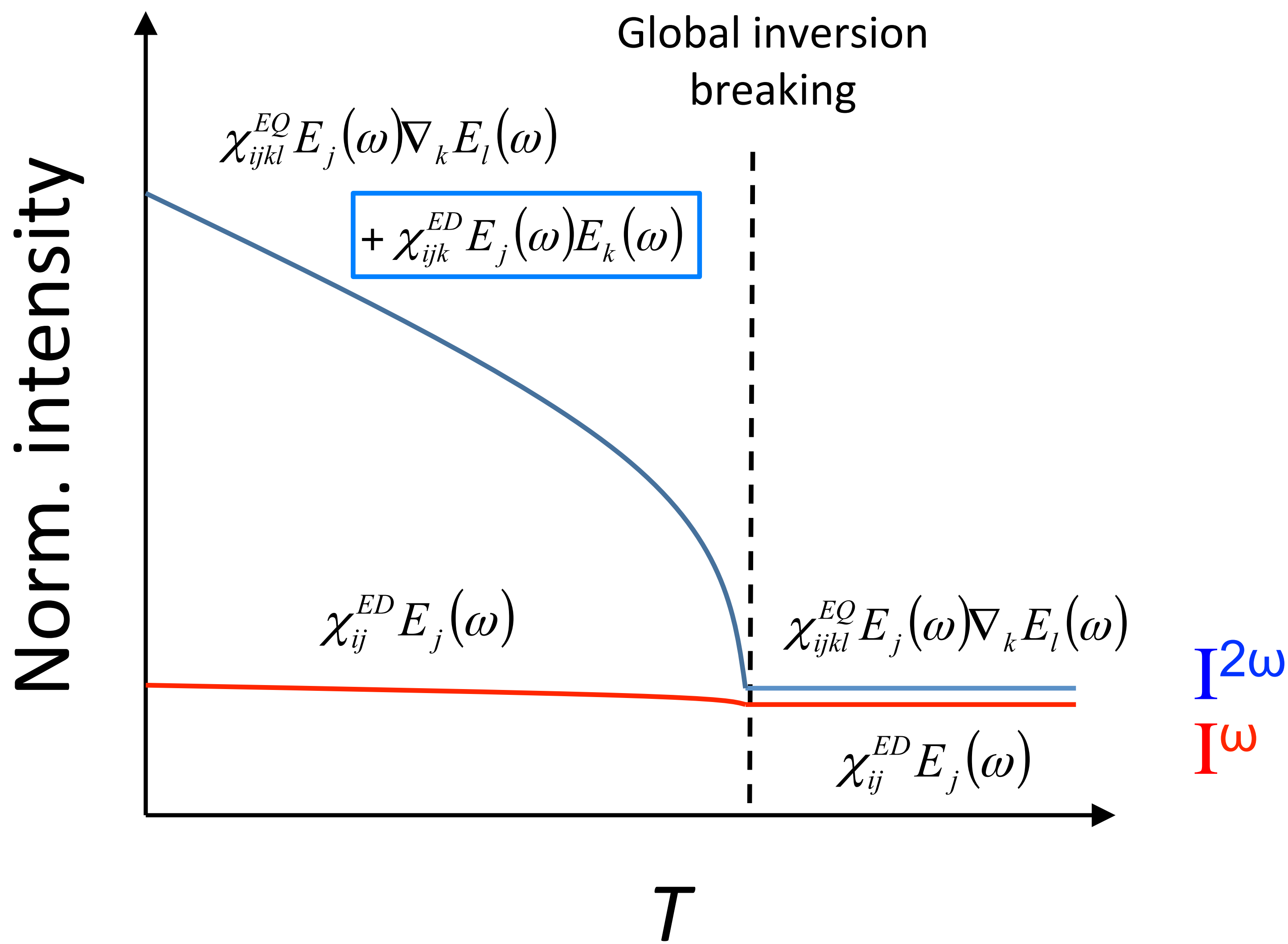
$$P_i(\omega) = \chi_{ij}^{ED} E_j(\omega)$$

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

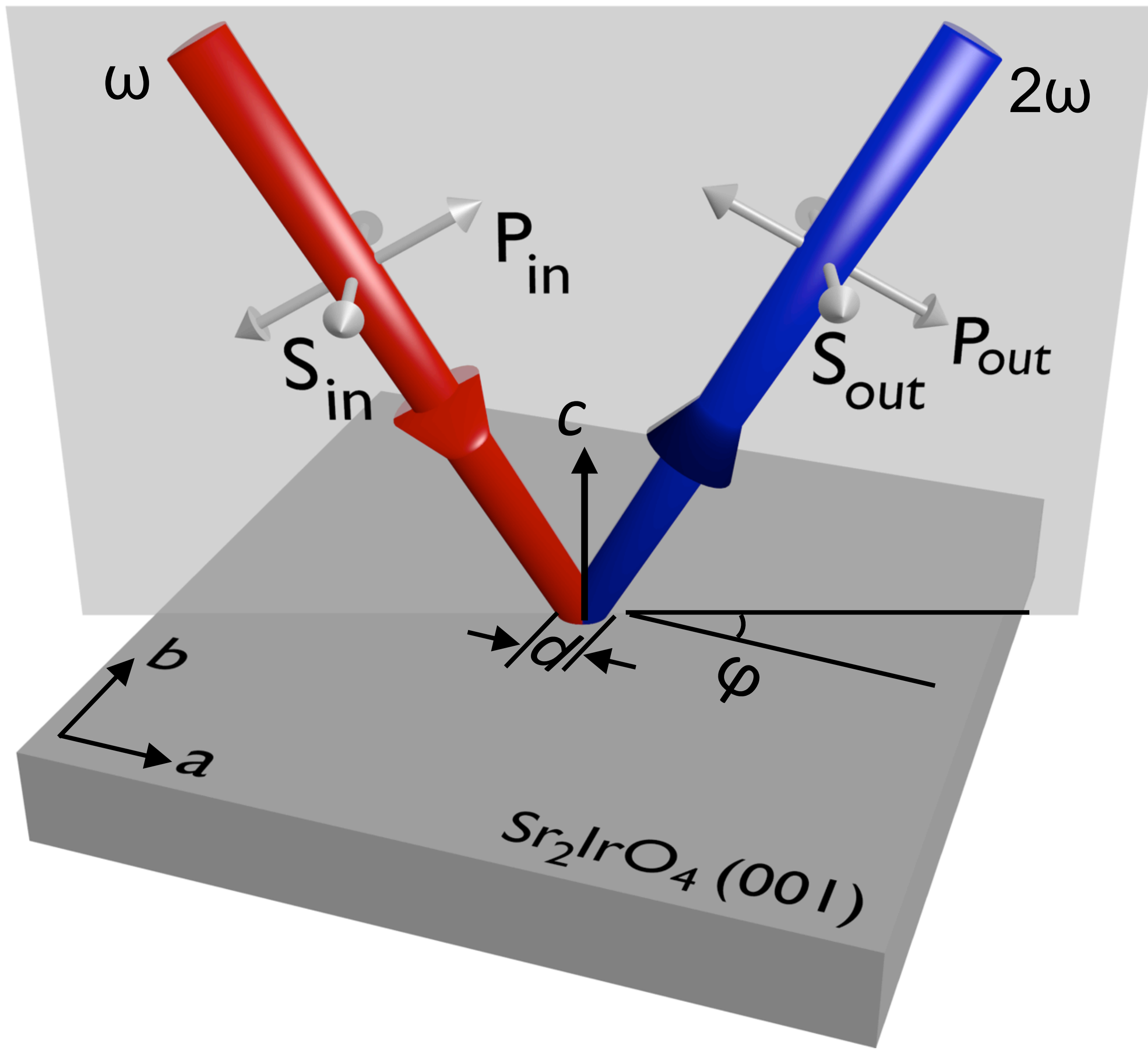
$\chi_{ijk}^{ED} = 0$ if system has inversion symmetry (except at surface)

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

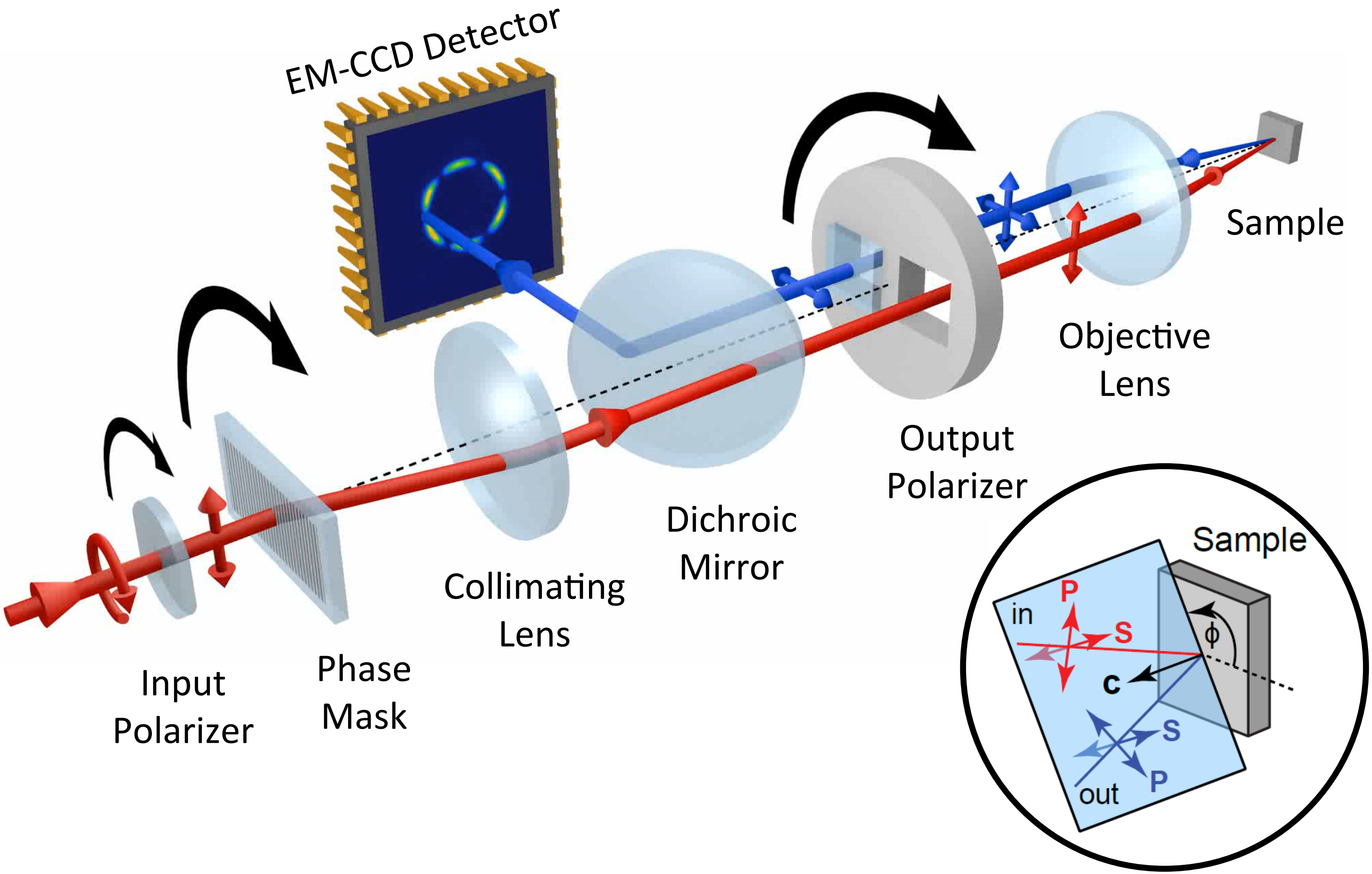
Signature of global inversion symmetry breaking



Rotational anisotropy to measure χ



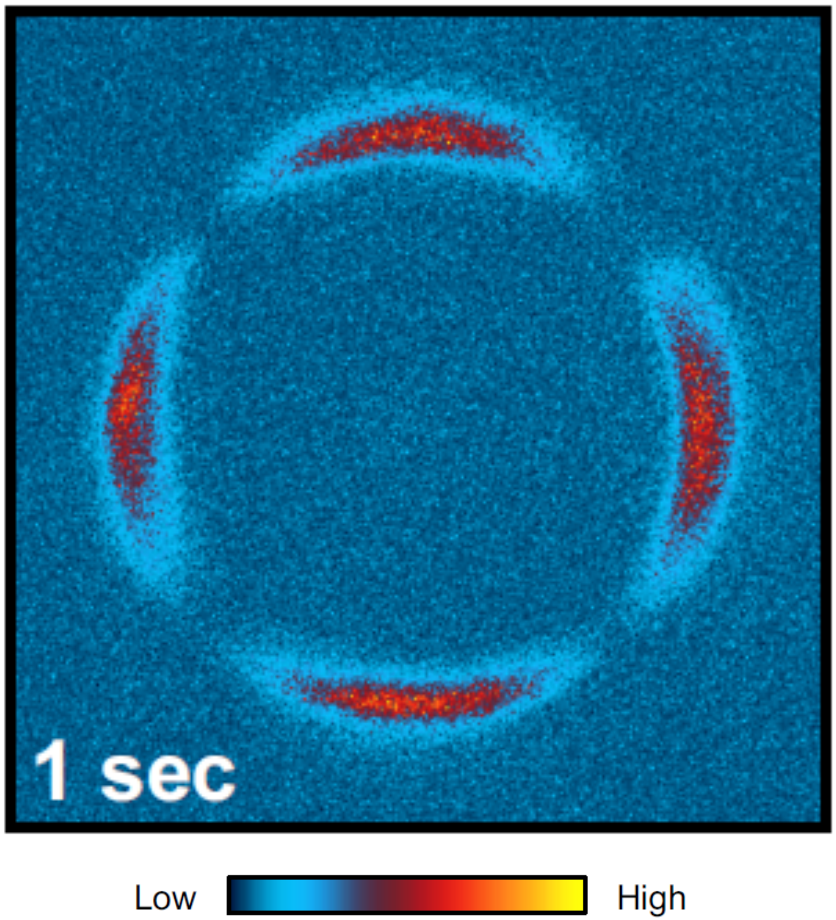
Rotating scattering plane based RA technique

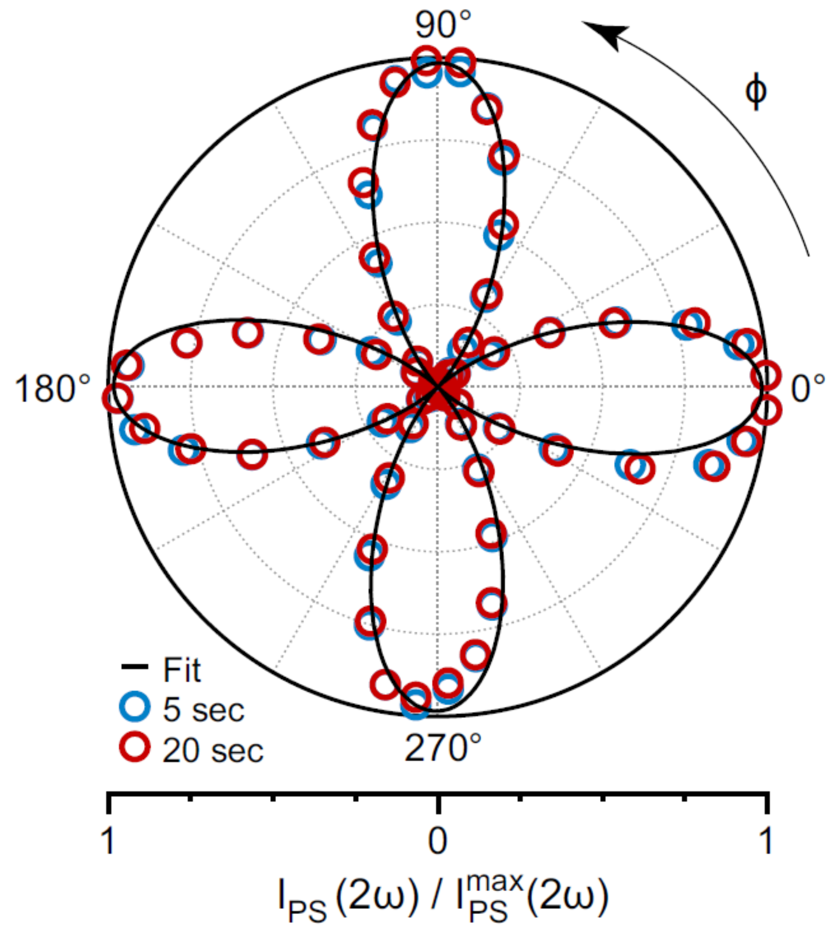


High speed / high sensitivity RA data acquisition

$$I^{2\omega}(\varphi) \propto |e_i^{2\omega}(\varphi)\chi_{ijk}^{ED} e_{j,0}^{\omega}(\varphi) e_{k,0}^{\omega}(\varphi)|^2 I_0^2$$

RA-SHG from GaAs(001)





Part Two

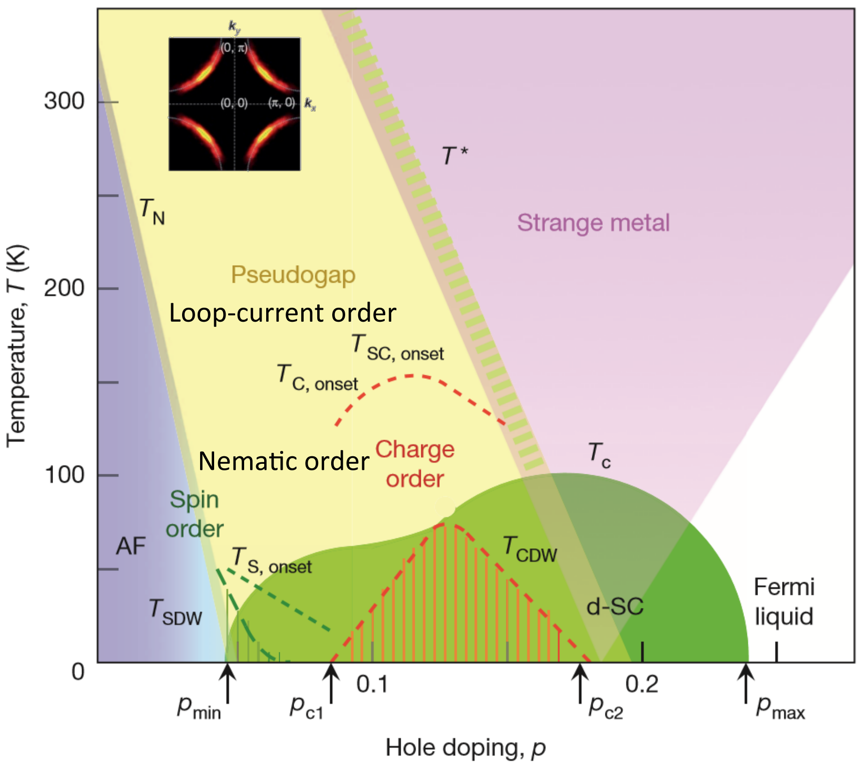
Hidden order in the pseudogap region of doped Sr_2IrO_4 and $YBa_2Cu_3O_y$

D. H. Torchinsky *et al*. Phys. Rev. Lett. **114**, 096404 (2015)

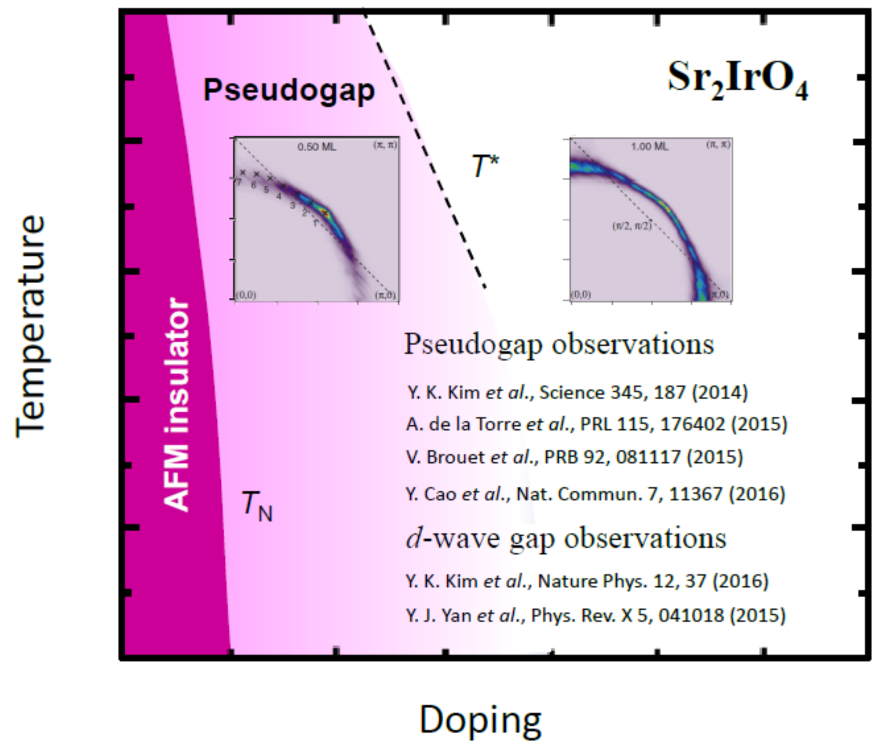
L. Zhao *et al*. Nature Phys. **12**, 32 (2016)

L. Zhao *et al*. Nature Phys. **13**, 250 (2017)

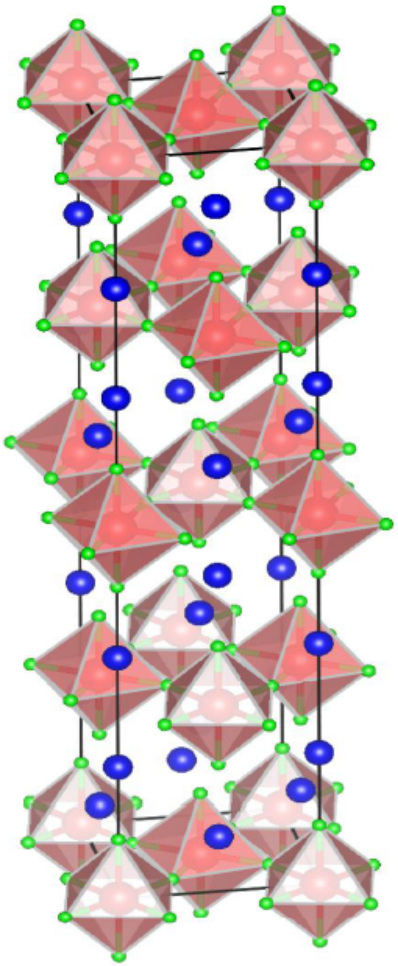
The cuprate/iridate phase diagram

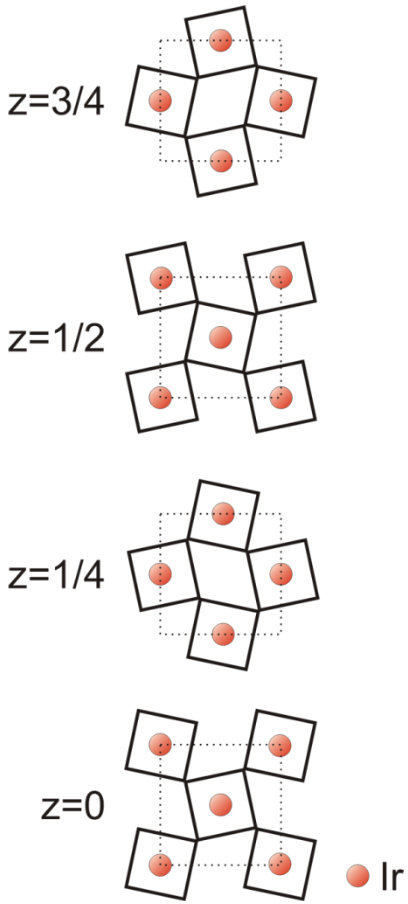


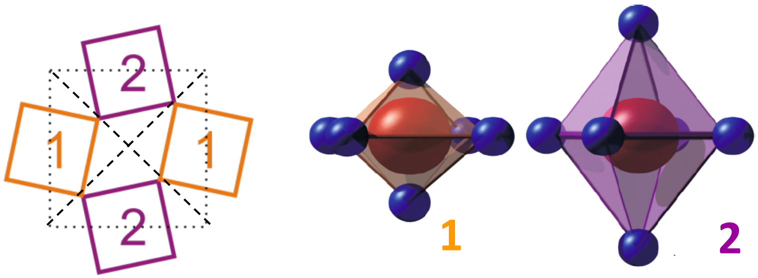
B. Keimer *et al.*, Nature Review 518, 179 (2015)



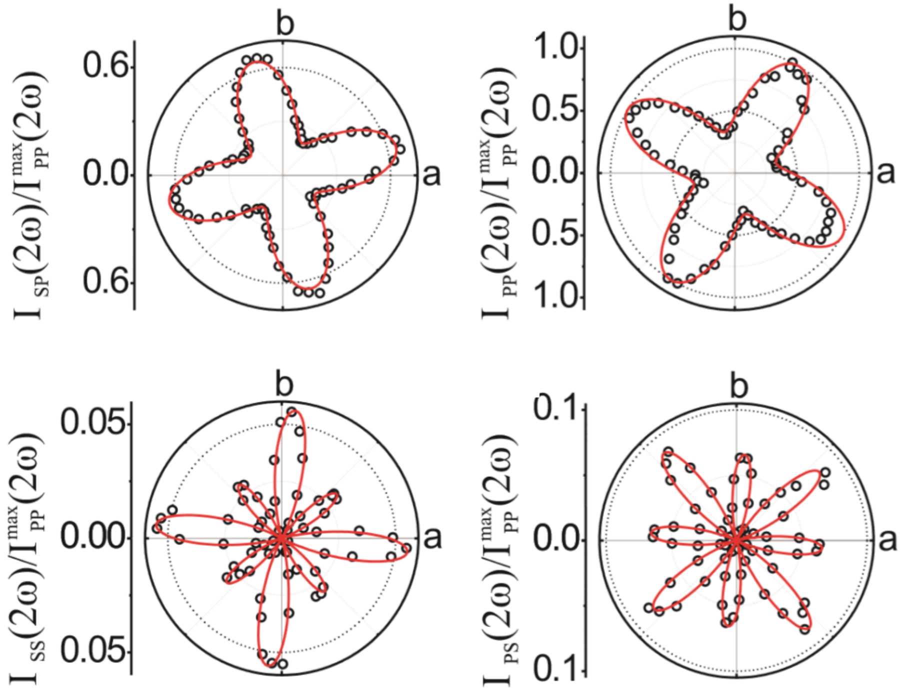
RA-SHG data from Sr_2IrO_4 (001) at $T = 295$ K







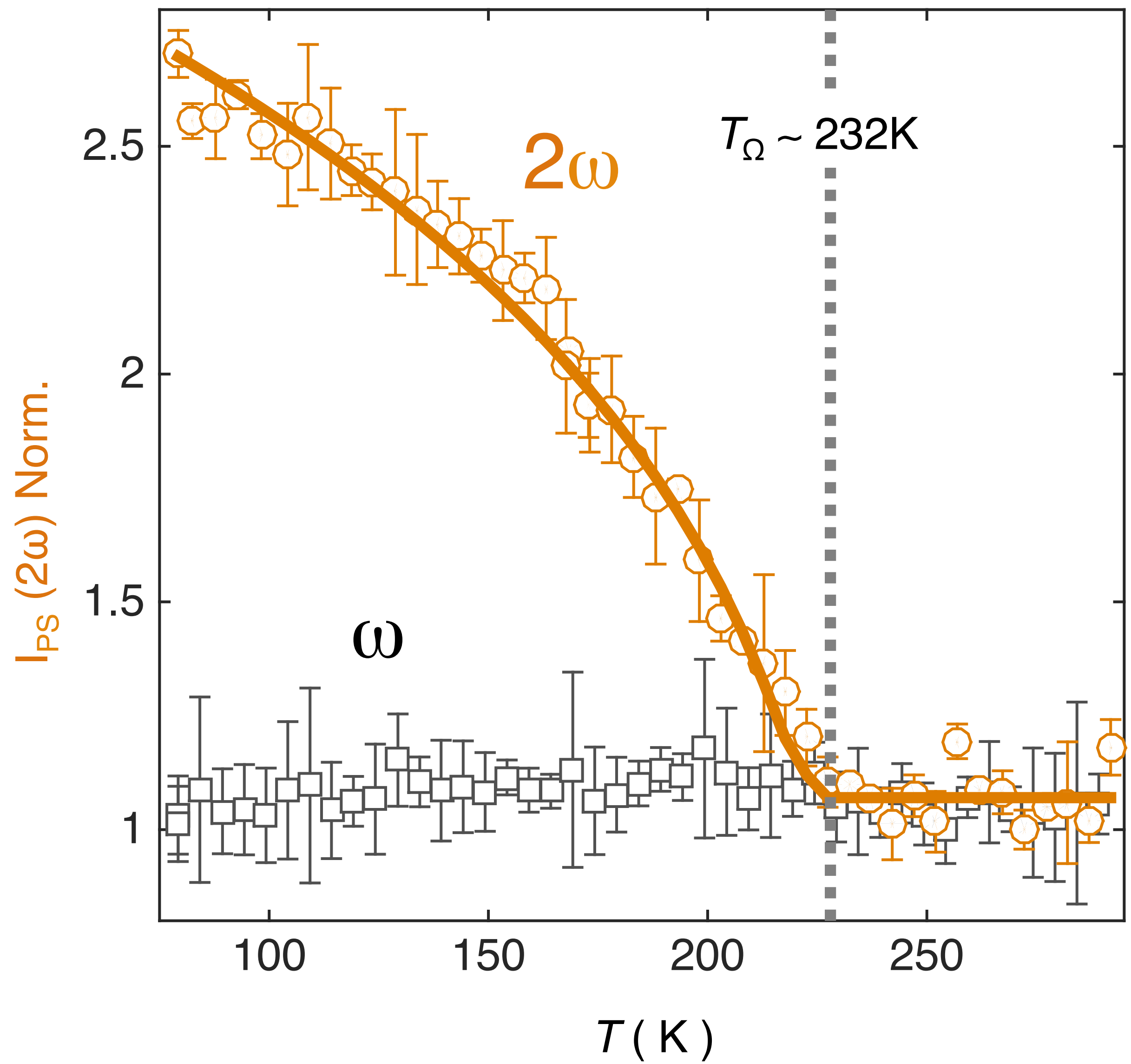
— $4/m$ centrosymmetric (bulk EQ)

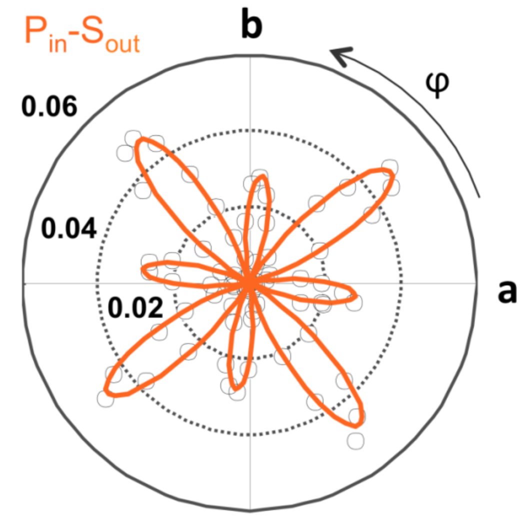


D. H. Torchinsky et al. PRL 114, 096404 (2015)

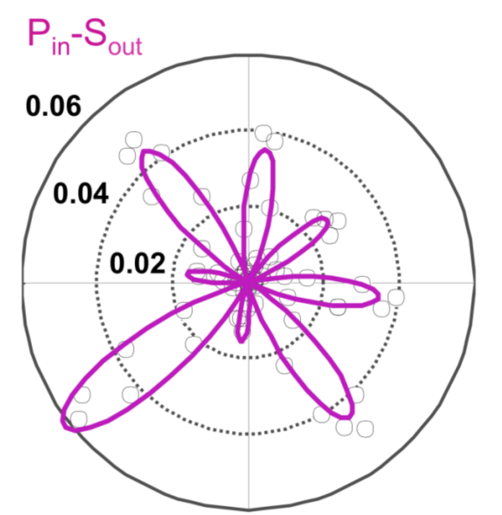
corroborated by neutron: Ye et al. PRB 92, 201112(R) (2015)

Hidden symmetry breaking below T_Ω



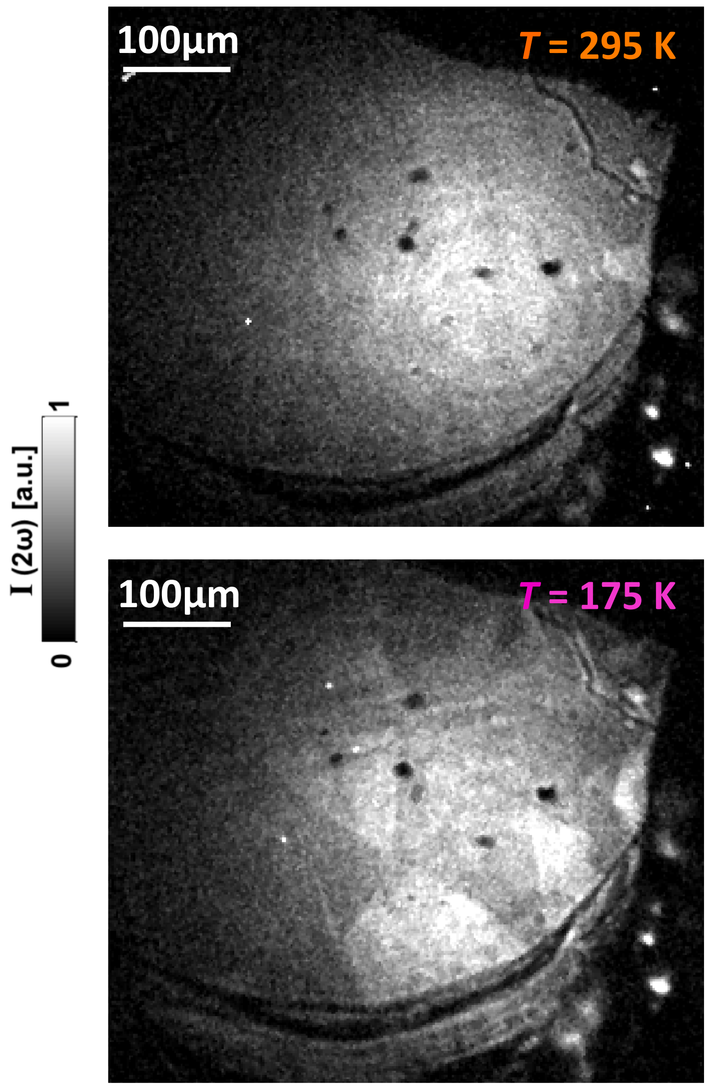


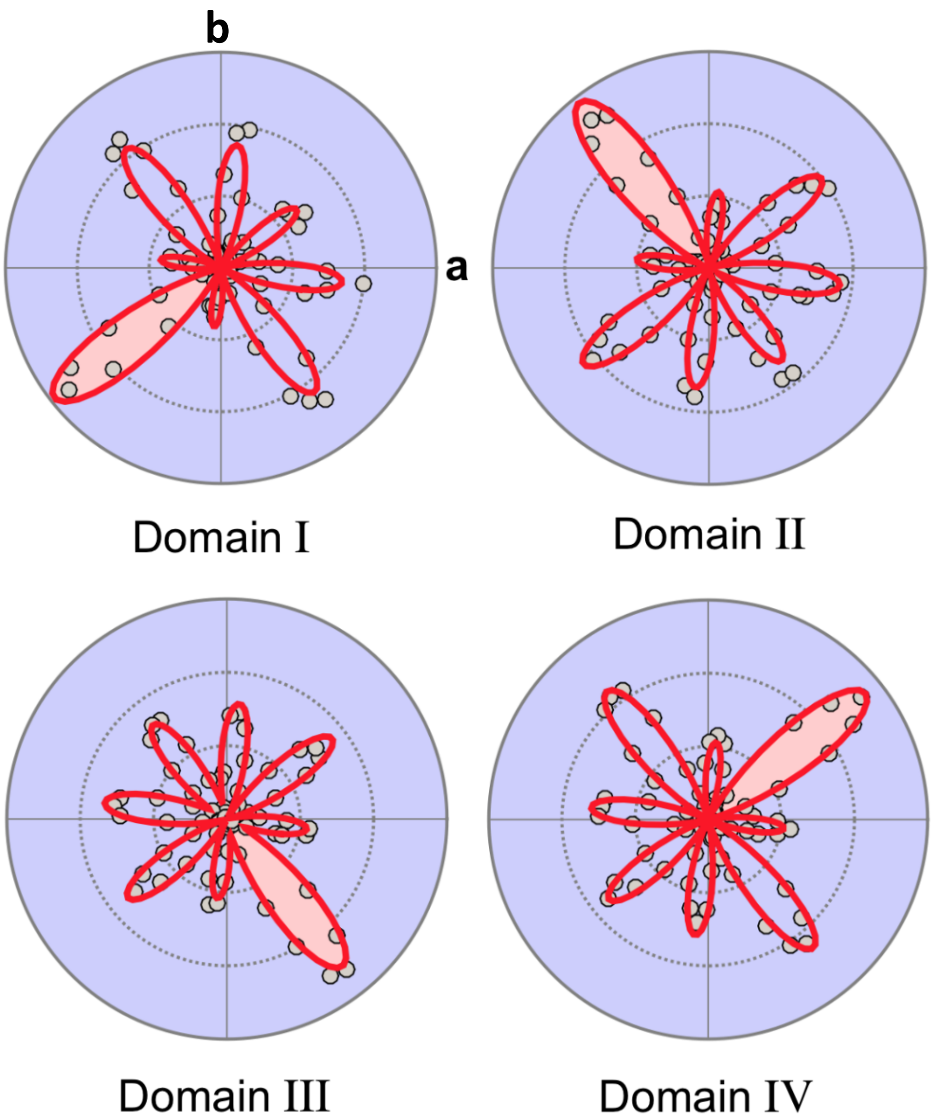
Bulk EQ (4/m)



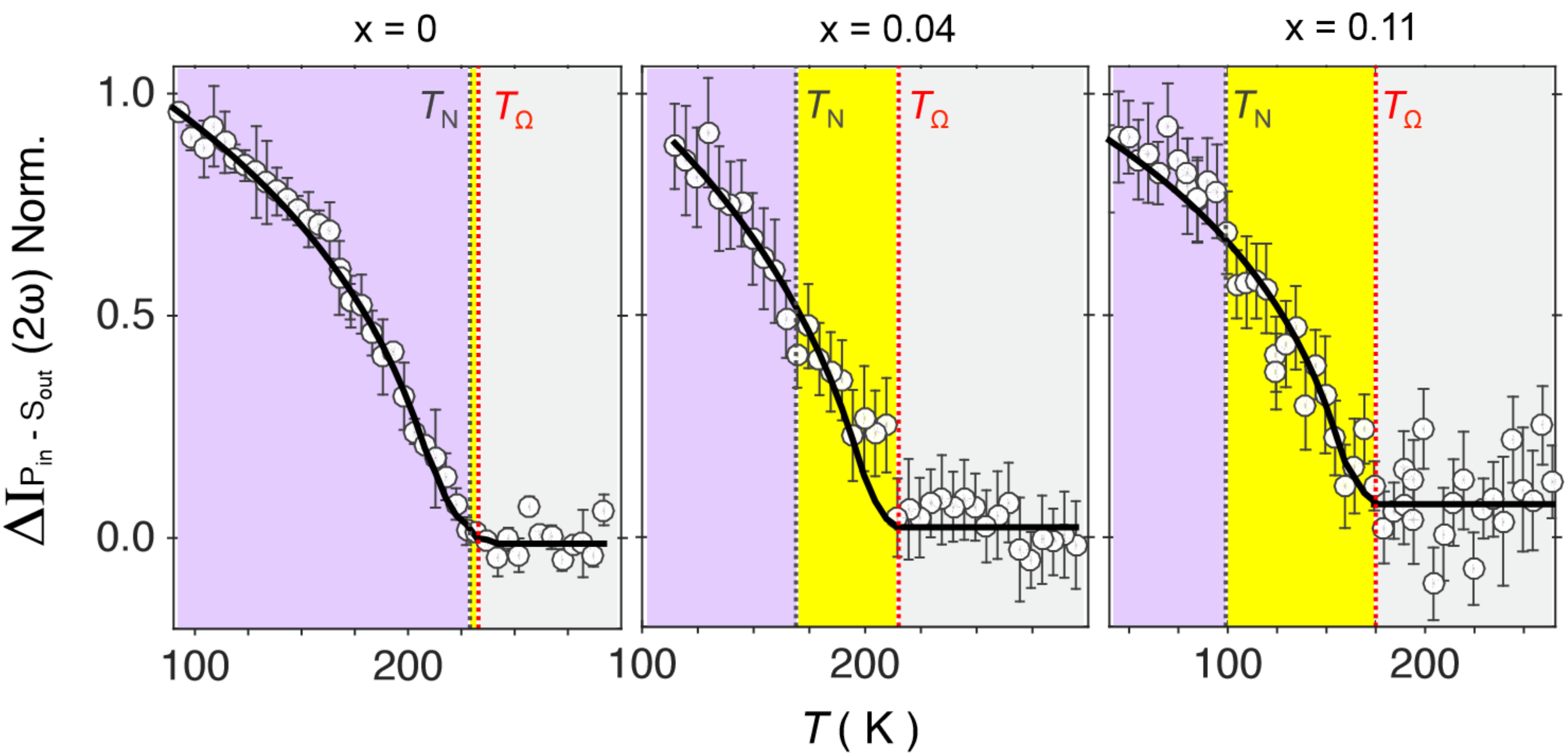
Bulk EQ (4/m) + bulk ED (2′/m)

Nonlinear optical microscopy

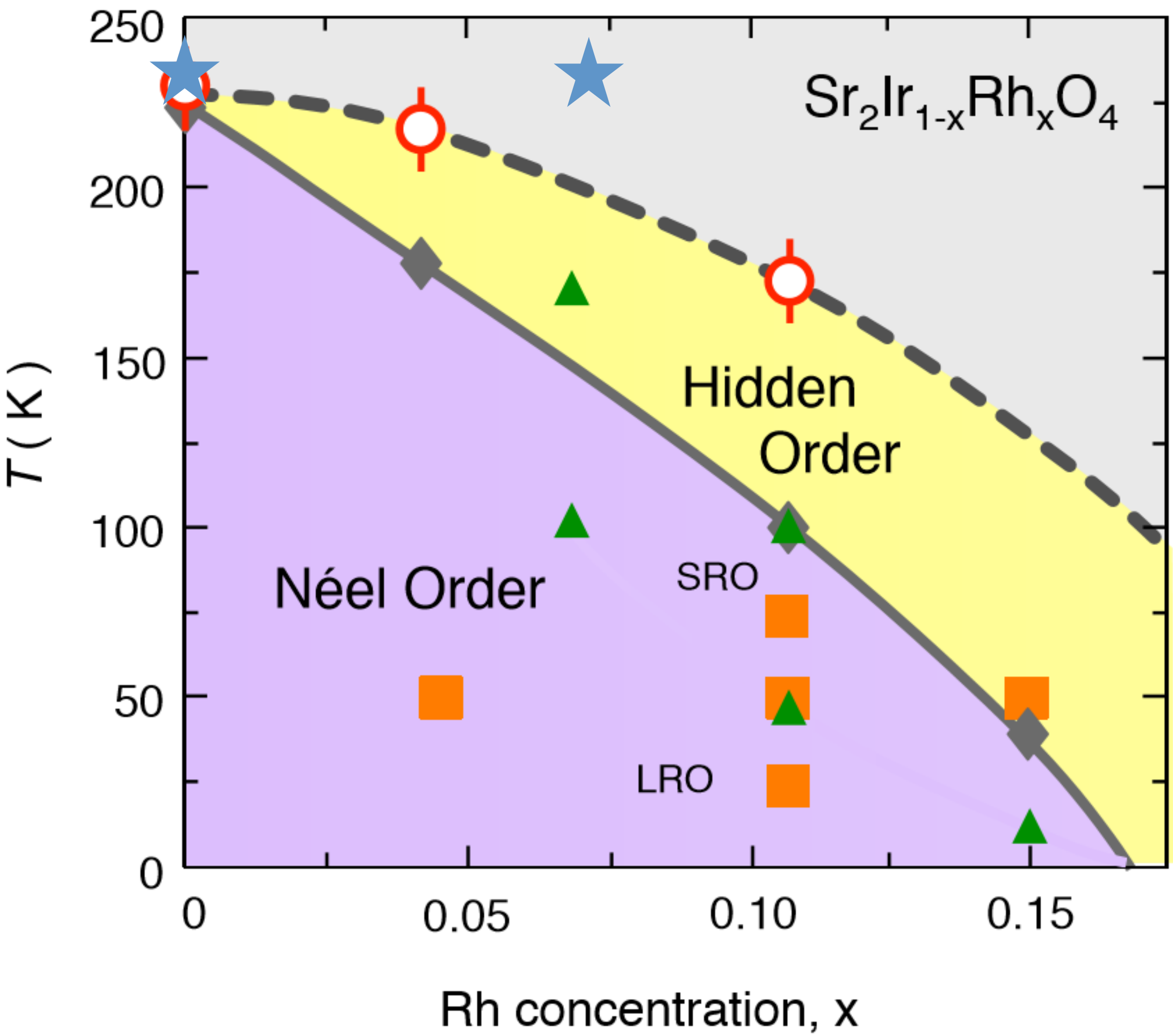




Doping dependence of T_Ω in $Sr_2Ir_{1-x}Rh_xO_4$



Phase diagram of $Sr_2Ir_{1-x}Rh_xO_4$



SHG (T_Ω)

DC magnetic susceptibility (T_N)

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

Resonant x-ray diffraction (T_N)

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

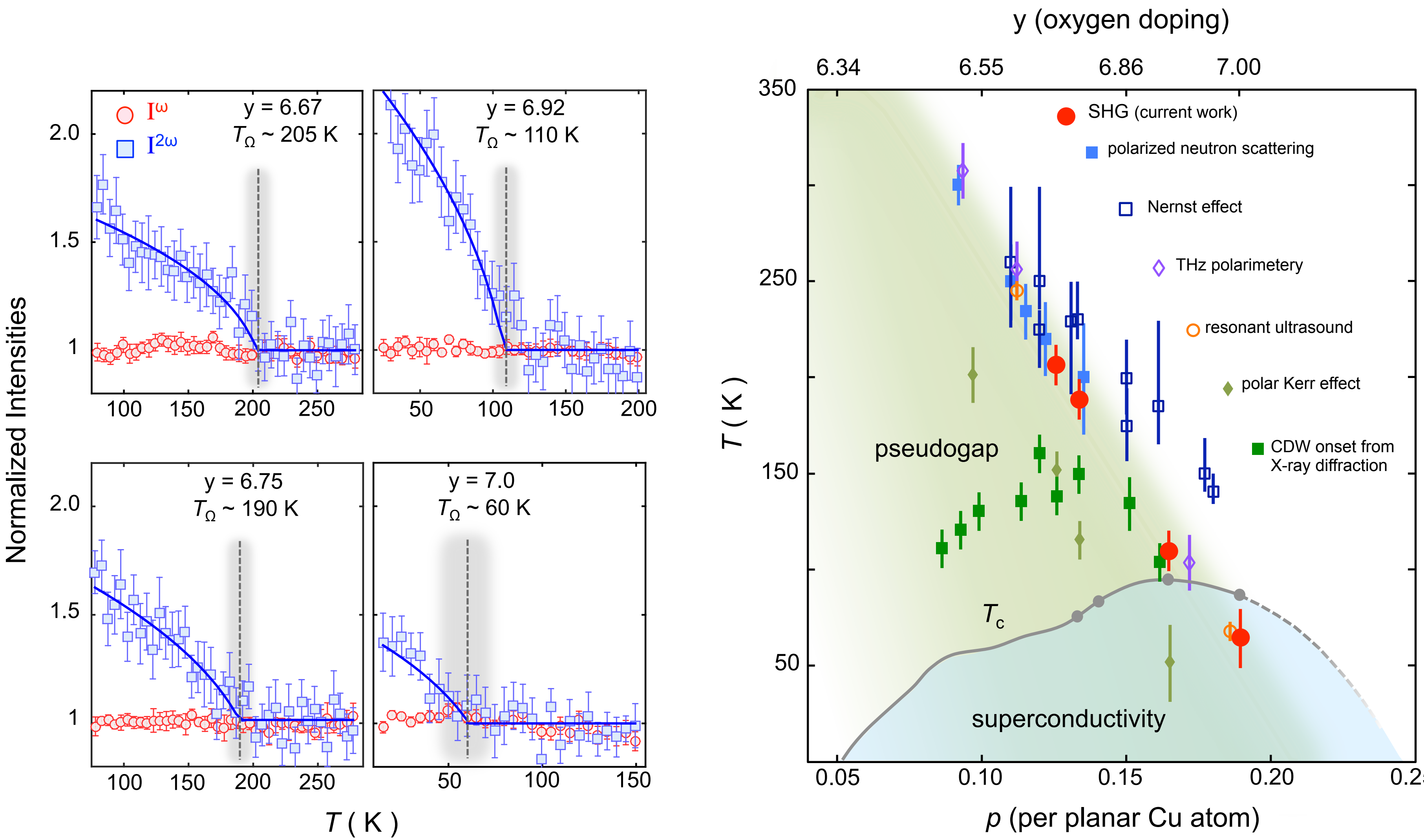
ARPES (pseudogap present)

Y. Cao *et al.*, Nat. Commun. 7, 11367 (2016)

Polarized neutron diffraction

J. Jeong *et al.*, Nat. Comm. 8, 15119 (2017)

Broken inversion symmetry below T^* in $YBa_2Cu_3O_y$



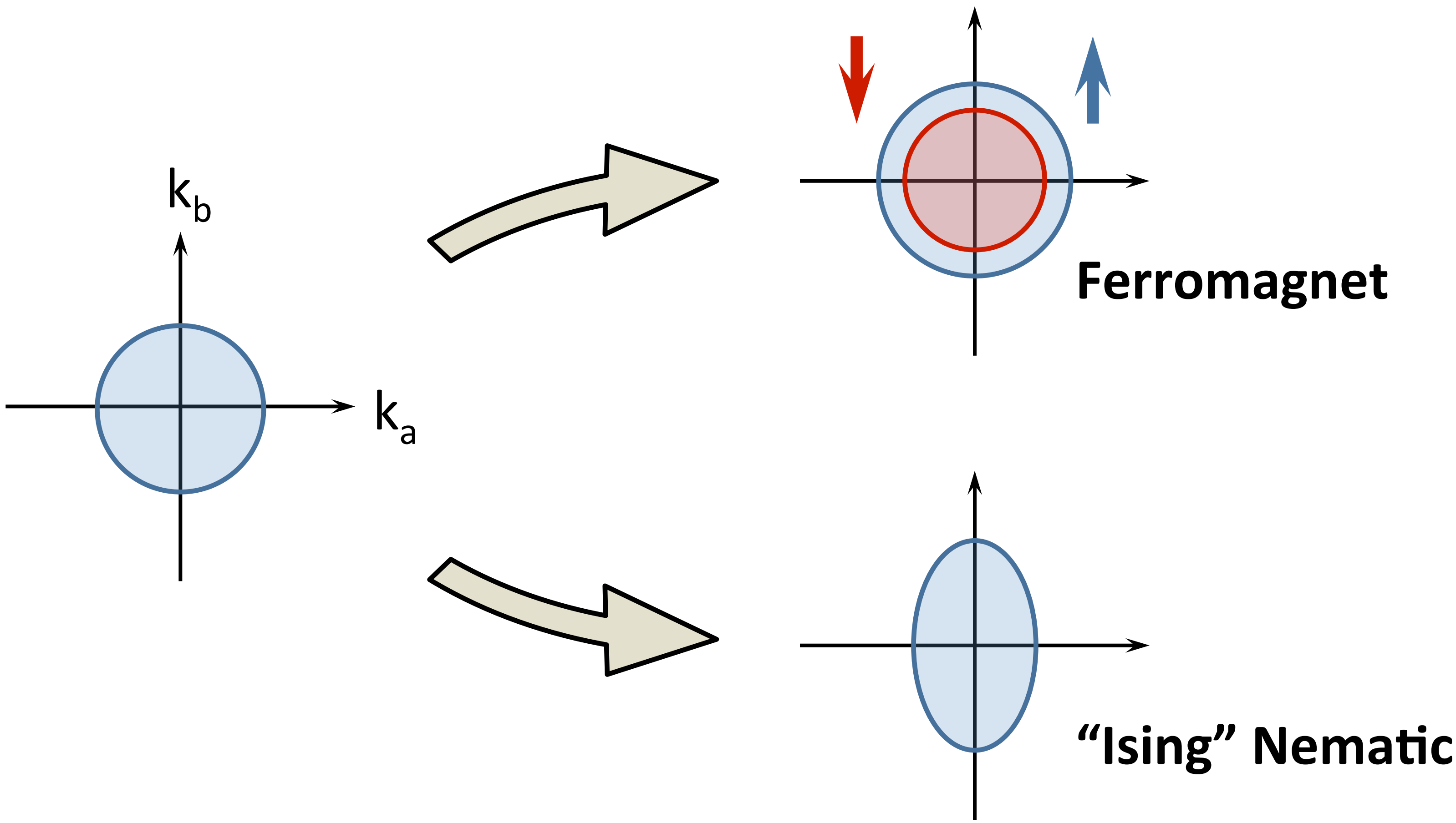
Part Three

A parity-breaking multipolar nematic phase in $Cd_2Re_2O_7$

J. Harter *et al*., Science **356**, 295 (2017)

Pomeranchuk instabilities in metals

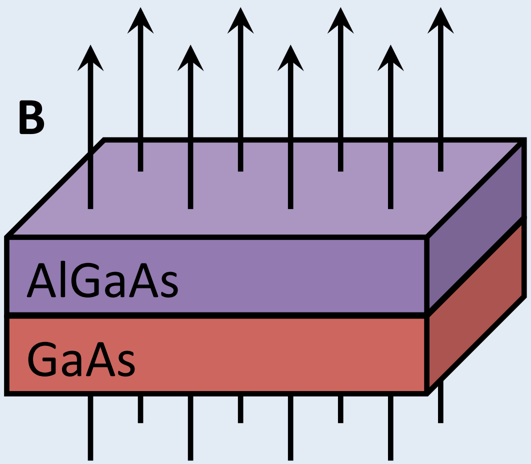
The Fermi surface of a metal can become thermodynamically unstable against a distortion that spontaneously breaks some symmetries of the lattice.

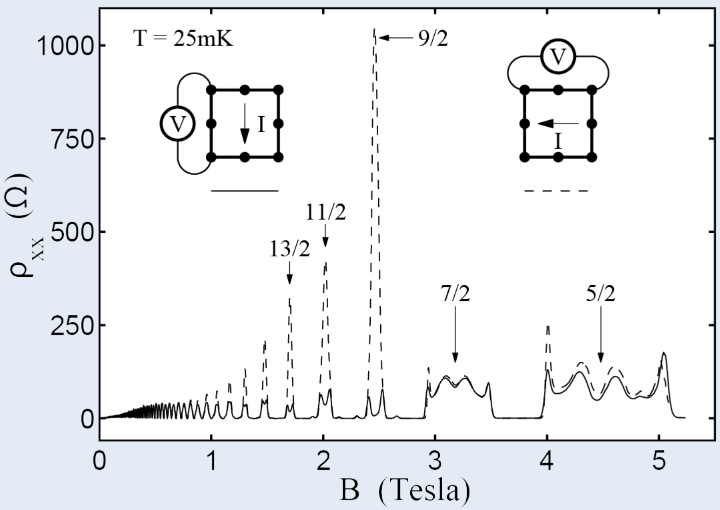


Pomeranchuk, *Sov. Phys. JETP* **8**, 361 (1959).

2D "Ising" electronic nematics

1. GaAs/AlGaAs Heterostructures





Lilly *et al.*, *Phys. Rev. Lett.* **82**, 394 (1999).

2D “Ising” electronic nematics

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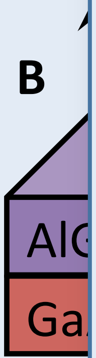
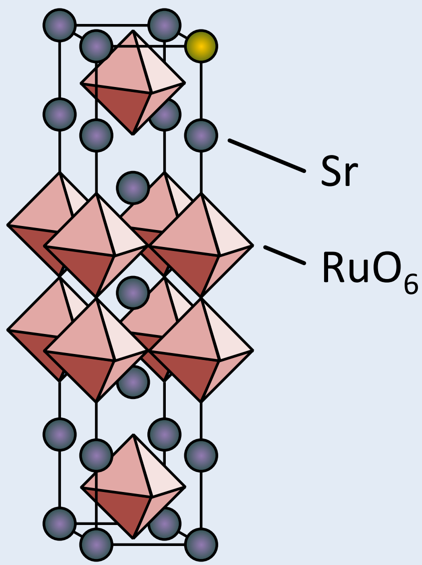
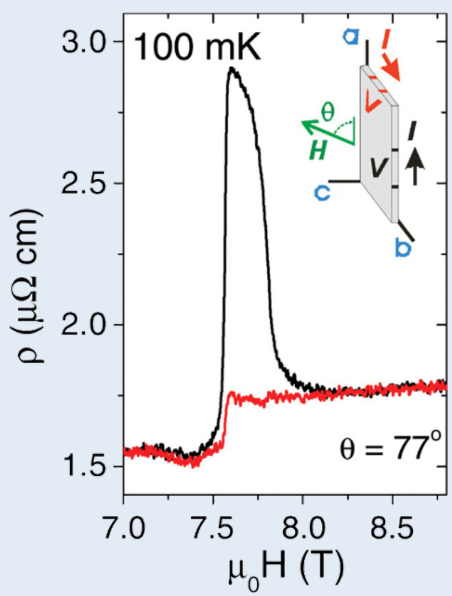


2. Metamagnetic $Sr_3Ru_2O_7$





Borzi *et al.*, *Science* **315**, 214 (2007).

2D “Ising” electronic nematics

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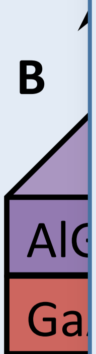
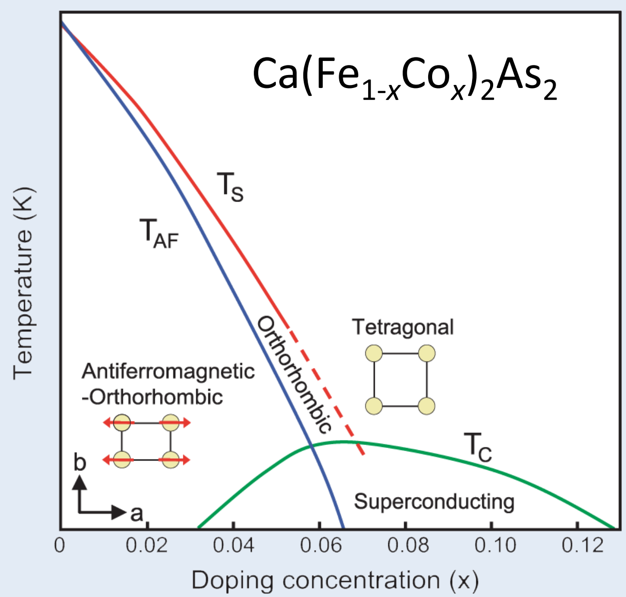
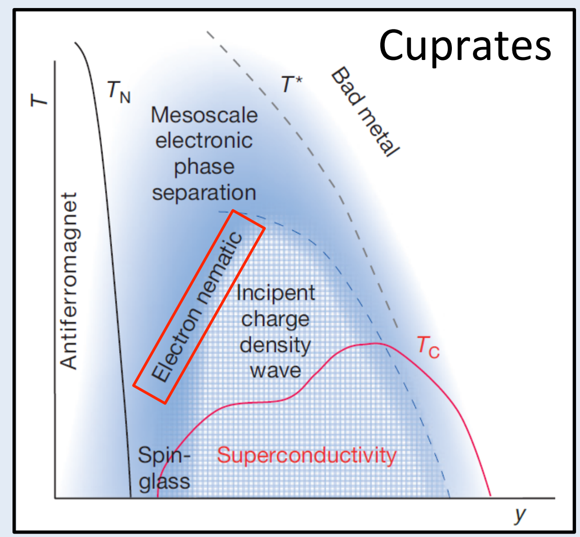


2. Metamagnetic $Sr_3Ru_2O_7$

3. High-T_c Superconductors



Chuang *et al.*, *Science* **327**, 181 (2010).



Fradkin & Kivelson, *NPhys*. **8**, 864 (2012).

Spin-textured Fermi surface (generalized ferromagnetism)

TR-invariant
Inv. broken

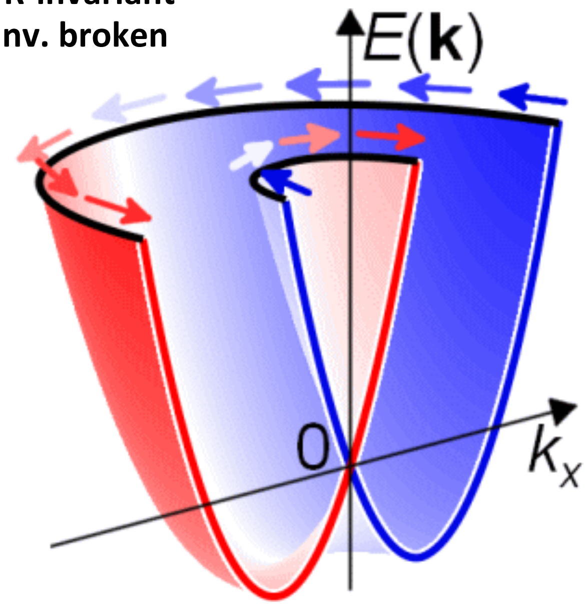


Image from Ishizaka group website

SOC + inversion broken structural host

- Asymmetric semiconductor quantum wells
- Topological insulator surfaces (e.g. Bi_2Se_3)
- Layered polar semiconductors (e.g. BiTeI)

Can it be spontaneously driven by electronic interactions?

- Examples of theoretical predictions:

Hirsch, *PRB* **41**, 6820 (1990)

Wu & Zhang, *PRL* **93**, 036403 (2004)

Varma & Zhu, *PRL* **96**, 036405 (2006)

Wu, Sun, Fradkin & Zhang, *PRB* **75**, 115103 (2007)

Li & Wu, *PRB* **85**, 205126 (2012)

Kunes & Geffroy, *PRL* **116**, 256403 (2016)

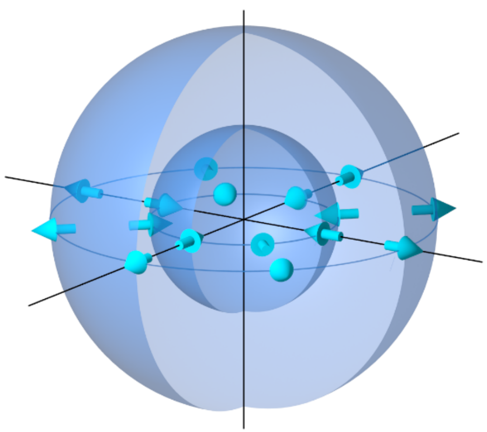
Parity-breaking phases of relativistic SO coupled metals

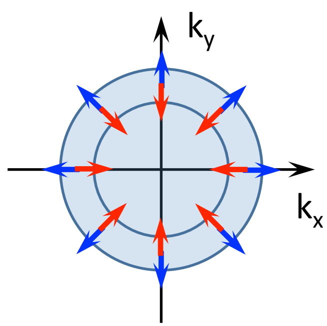
Fu, *Phys. Rev. Lett.* **115**, 026401 (2015):

Gyrotropic

(pseudoscalar)

$$\eta=\sum_{\mathbf{k}} \mathbf{k}\cdot\mathbf{s}(\mathbf{k})$$





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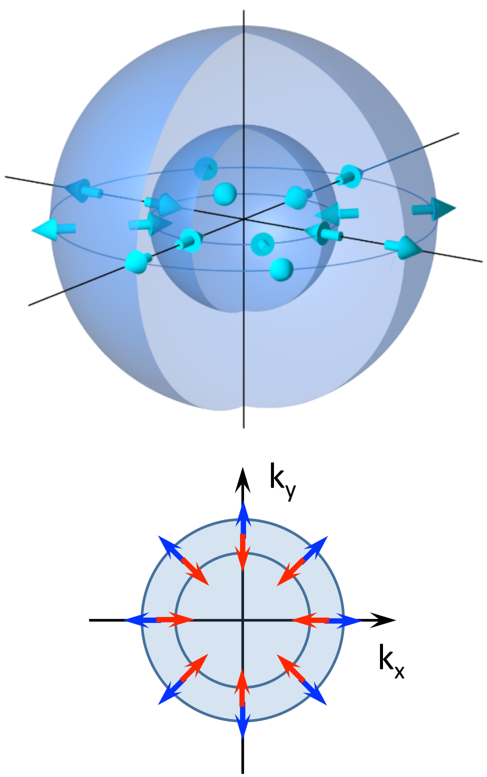
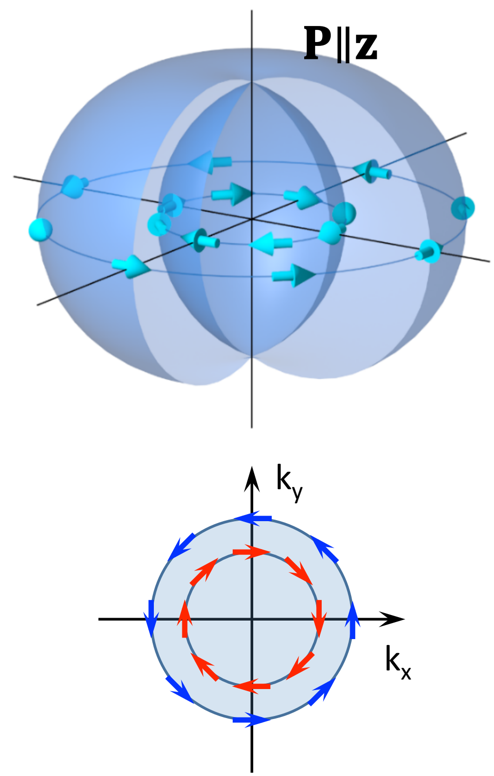


Ferroelectric
(vector)

$$\mathbf{P} = \sum_{\mathbf{k}} \mathbf{k} \times \mathbf{s}(\mathbf{k})$$

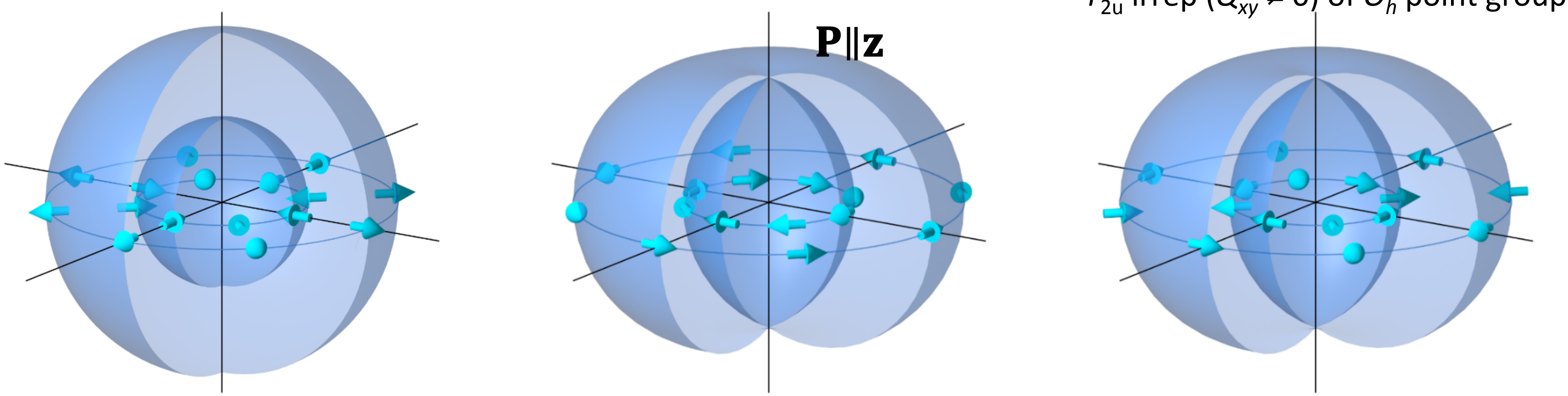


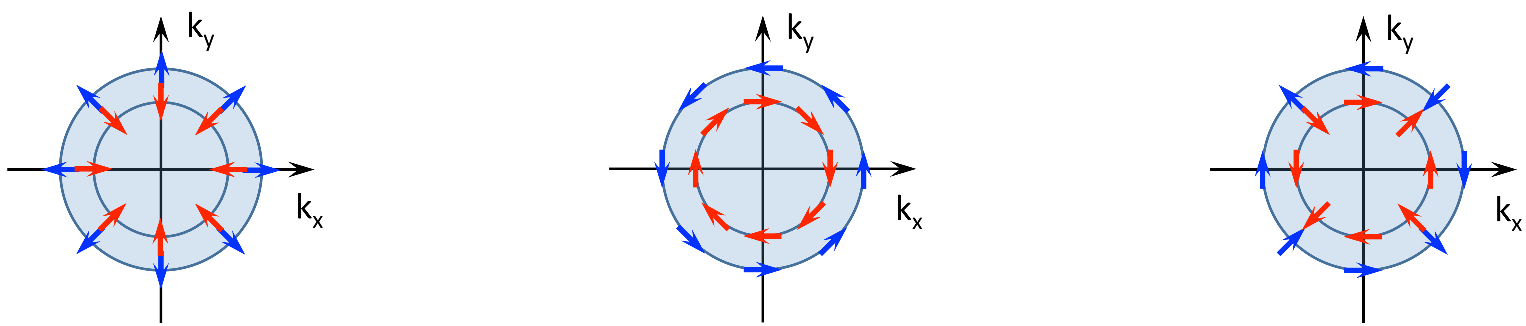
Parity-breaking phases of relativistic SO coupled metals

Fu, *Phys. Rev. Lett.* **115**, 026401 (2015):

Gyrotropic	**Ferroelectric**	**Multipolar Nematic**
(pseudoscalar)	(vector)	(rank-2 pseudotensor)
$\eta = \sum_{\mathbf{k}} k \cdot \mathbf{s}(\mathbf{k})$	$\mathbf{P} = \sum_{\mathbf{k}} k \times \mathbf{s}(\mathbf{k})$	$Q_{ij} = \sum_{\mathbf{k}} k_i \, s_j(\mathbf{k}) + k_j \, s_i(\mathbf{k})$

T_{2u} irrep ($Q_{xy} \neq 0$) of O_h point group



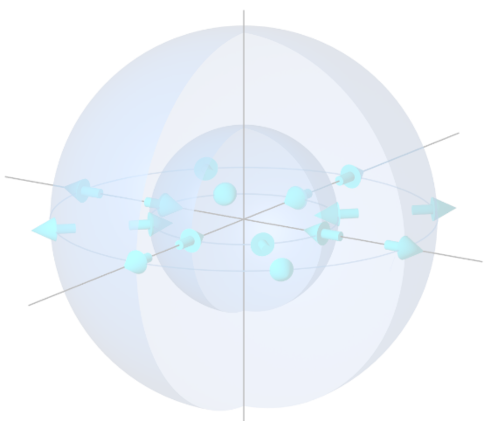


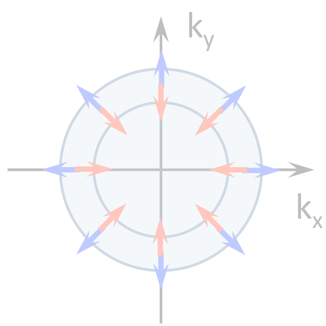
Parity-breaking phases of relativistic SO coupled metals

Fu, *Phys. Rev. Lett.* **115**, 026401 (2015):

Gyrotropic
(pseudoscalar)

$\eta=\sum_{\mathbf{k}} \mathbf{k}\cdot\mathbf{s}(\mathbf{k})$

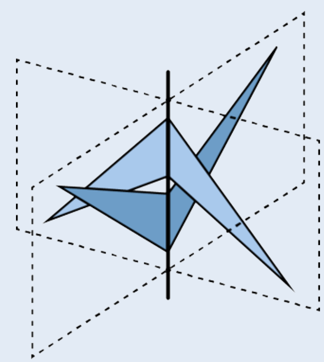




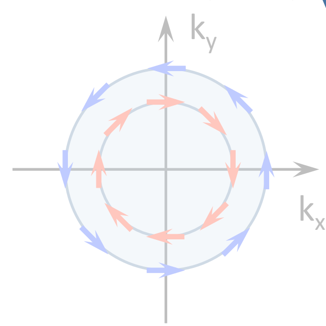
Ferroelectric
(vector)

$(\mathbf{k})$

N_T Phase



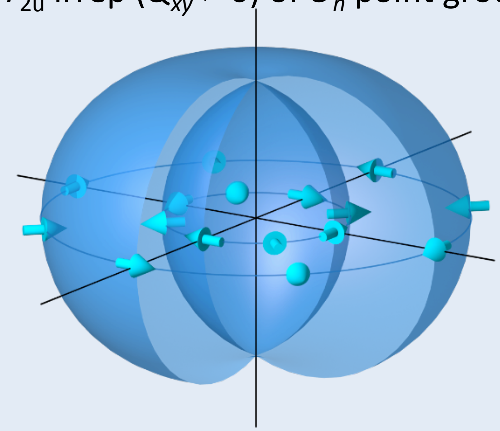
Lubensky & Radzihovsky,
Phys. Rev. E **66**, 031704 (2002).

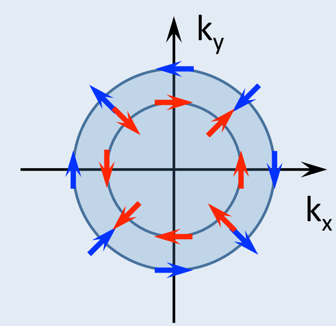


Multipolar Nematic
(rank-2 pseudotensor)

$Q_{ij}=\sum_{\mathbf{k}} k_i\, s_j(\mathbf{k})+ k_j\, s_i(\mathbf{k})$

T_{2u} irrep ($Q_{xy} \neq 0$) of O_h point group

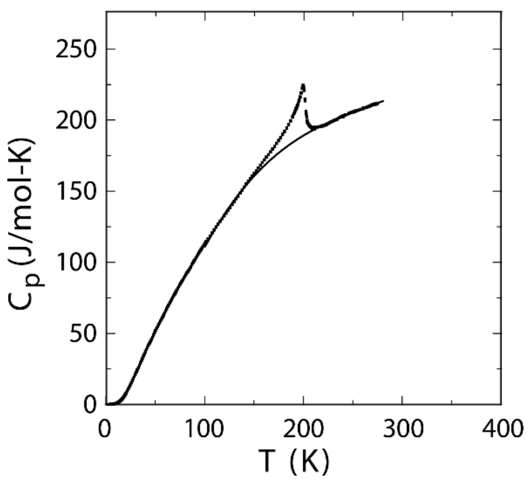




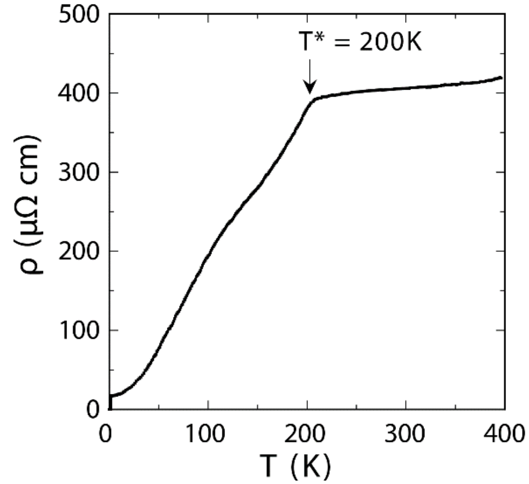
Superconducting pyrochlore $Cd_2Re_2O_7$

- At 200 K, a very weak structural distortion occurs, with dramatic electronic changes

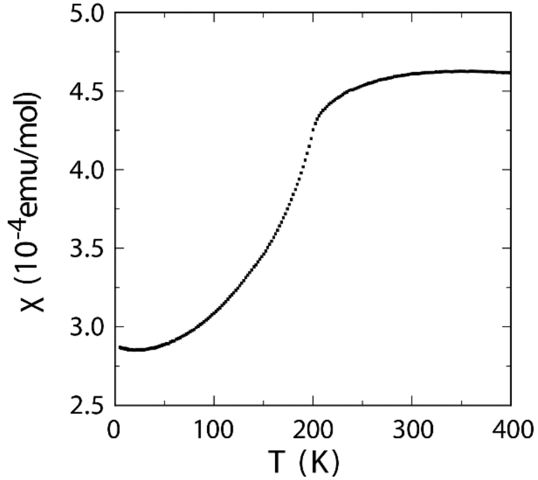
Heat capacity



Resistivity



Susceptibility



Jin *et al.*, *J. Phys.: Condens. Matter* **14**, L117 (2002).

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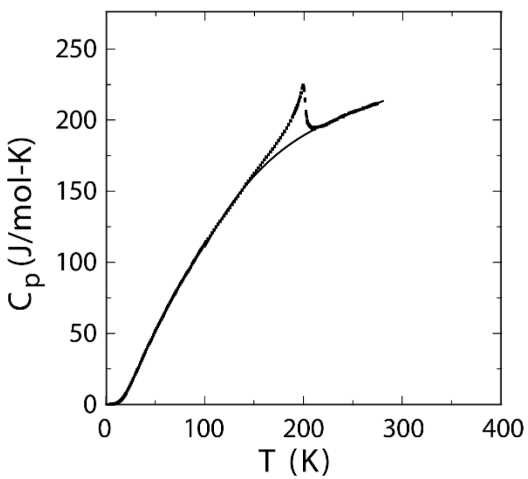
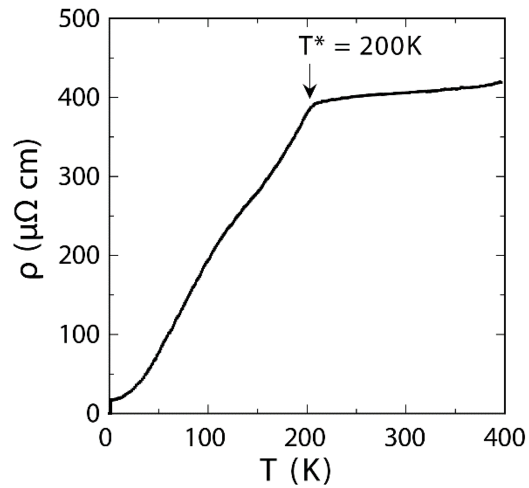
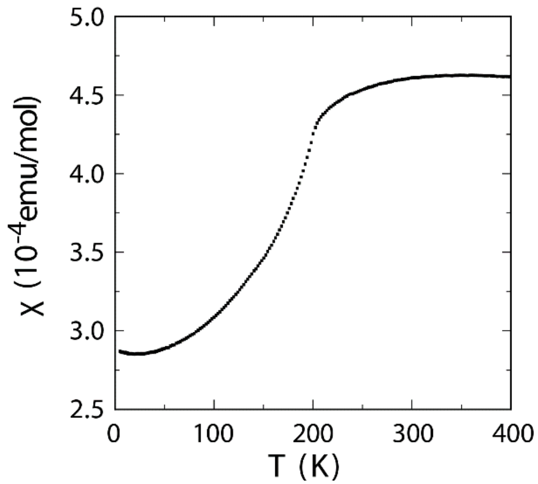


Resistivity

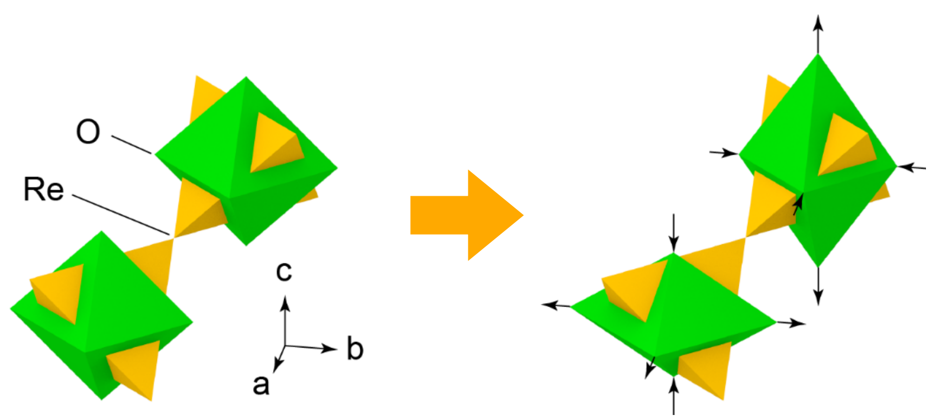


Susceptibility

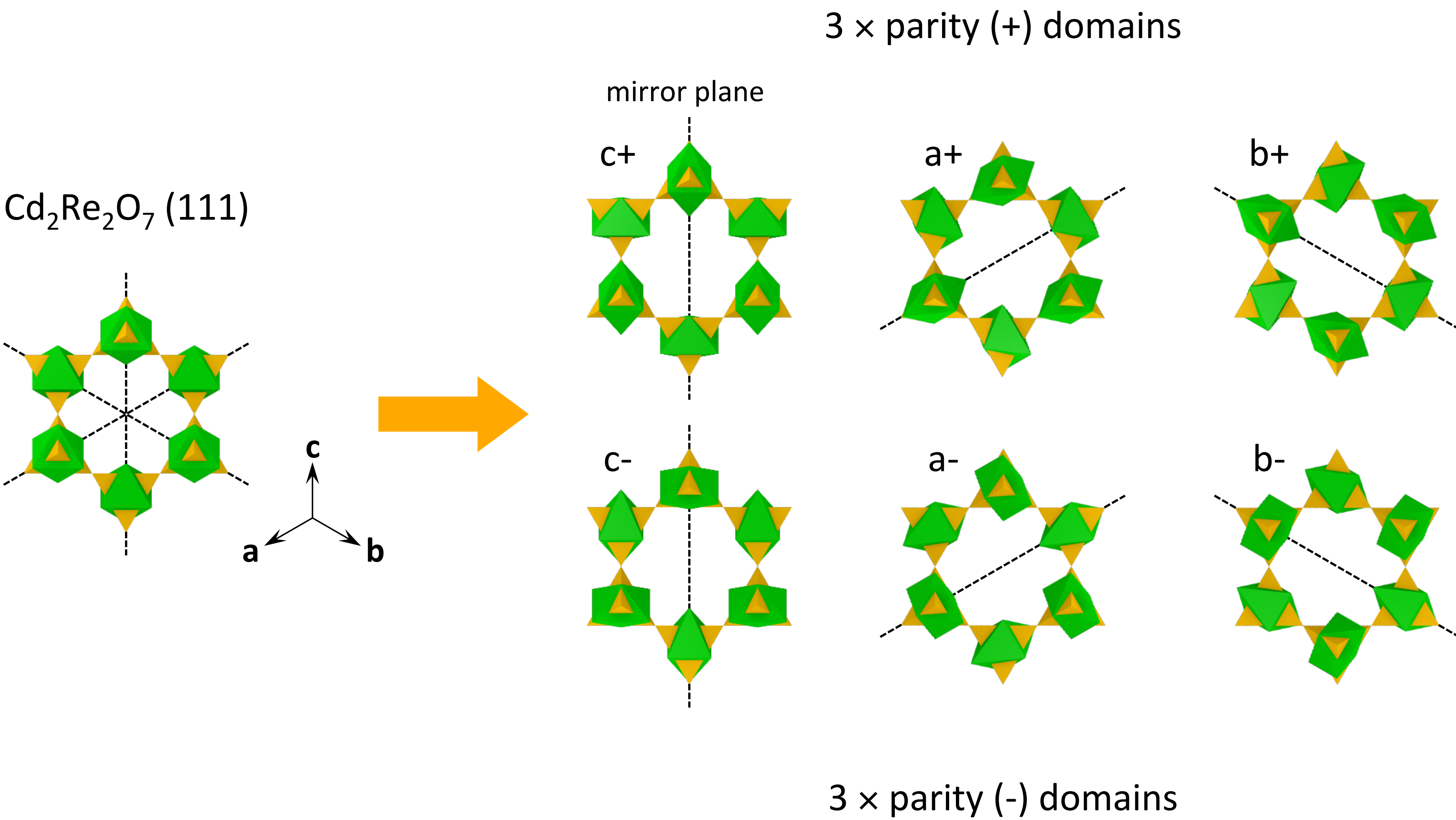


Jin *et al.*, *J. Phys.: Condens. Matter* **14**, L117 (2002).

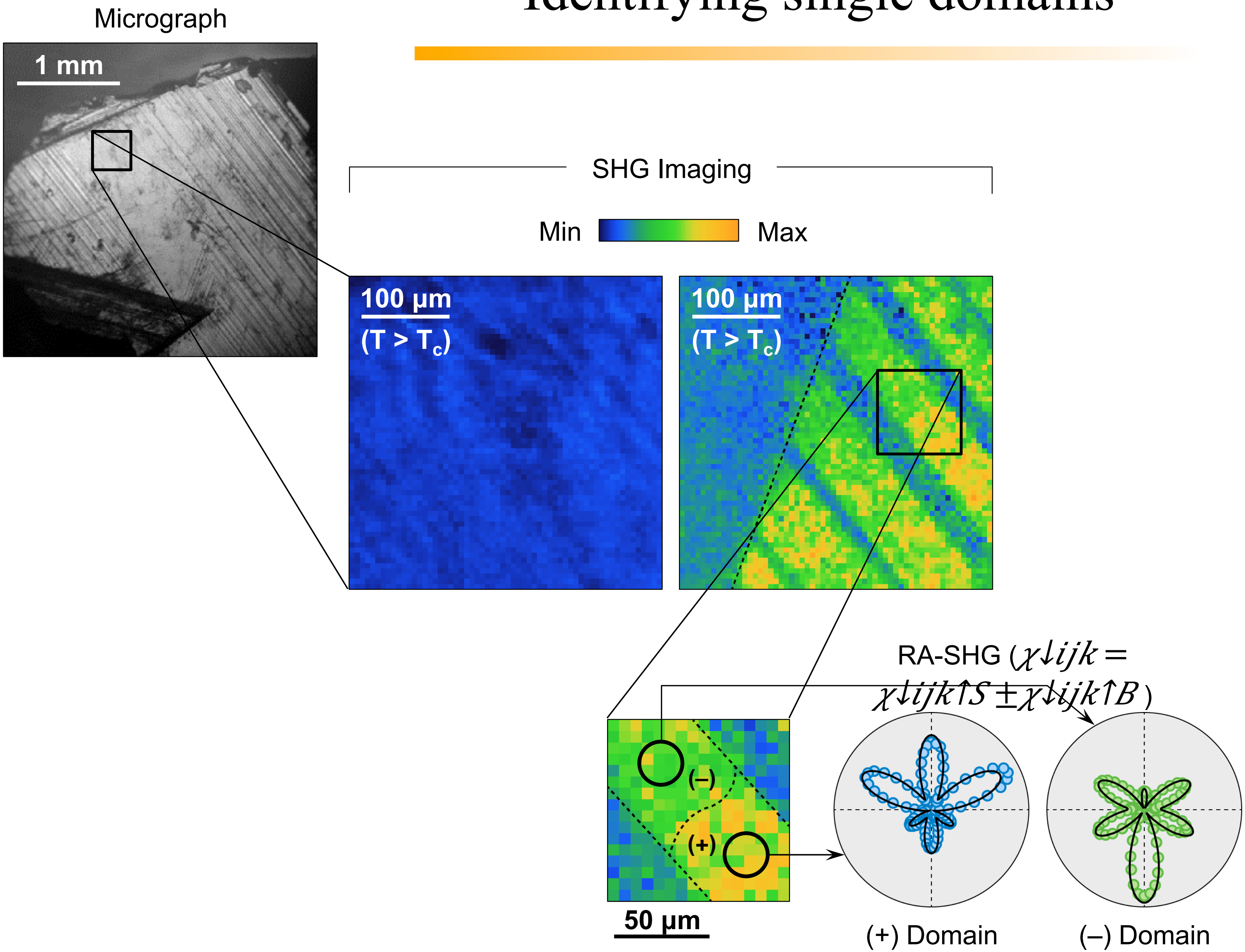
- Transition is cubic-to-tetragonal, with loss of inversion symmetry
- Structural order parameter has E_u symmetry, associated with distortion of O atoms



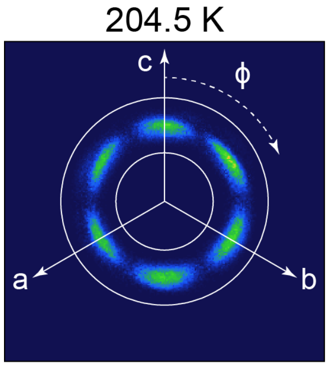
Structural domain formation



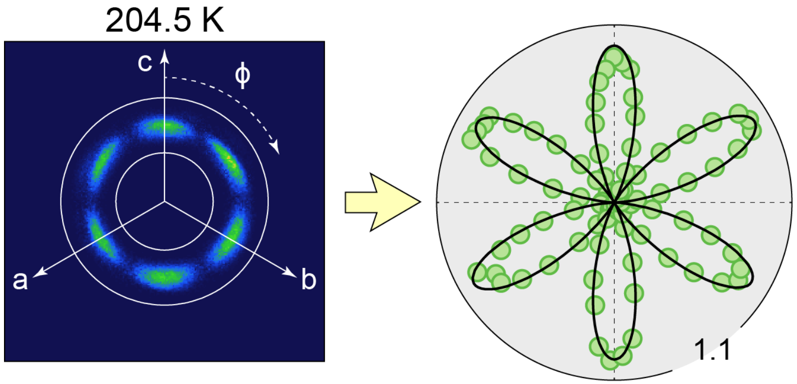
Identifying single domains

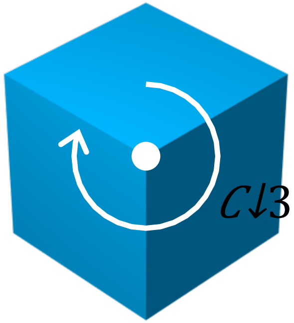


Rotational anisotropy measurements

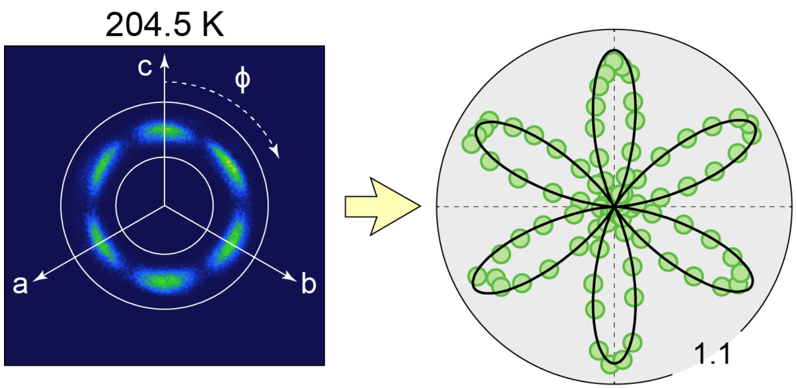


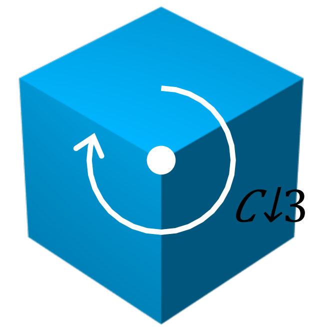
Rotational anisotropy measurements

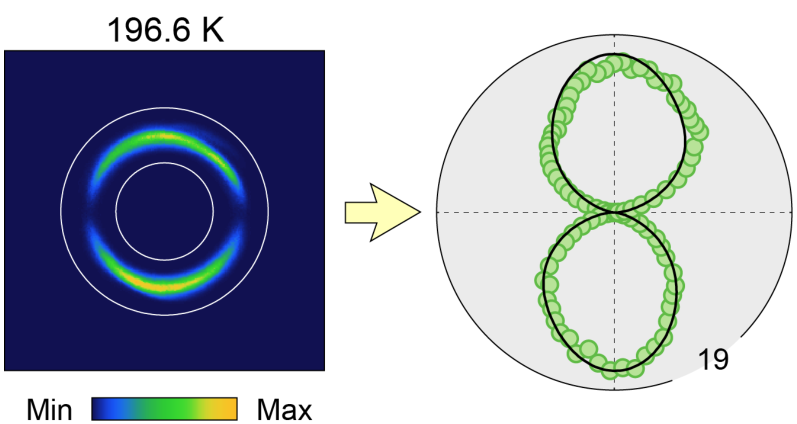


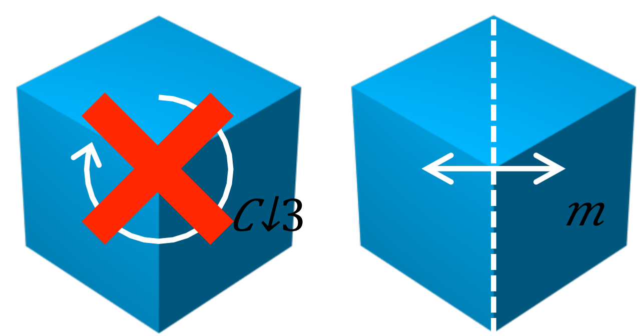


Rotational anisotropy measurements

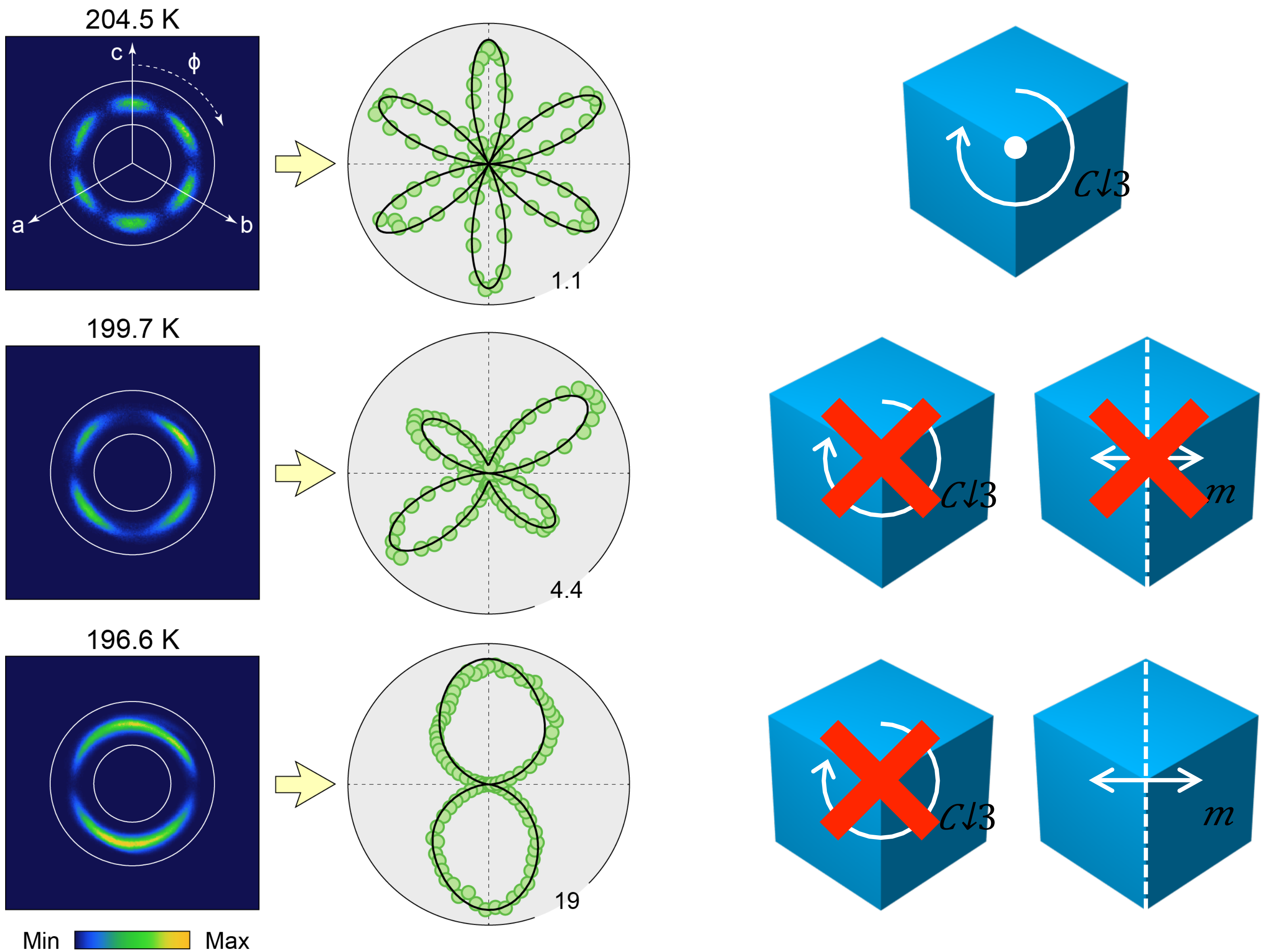




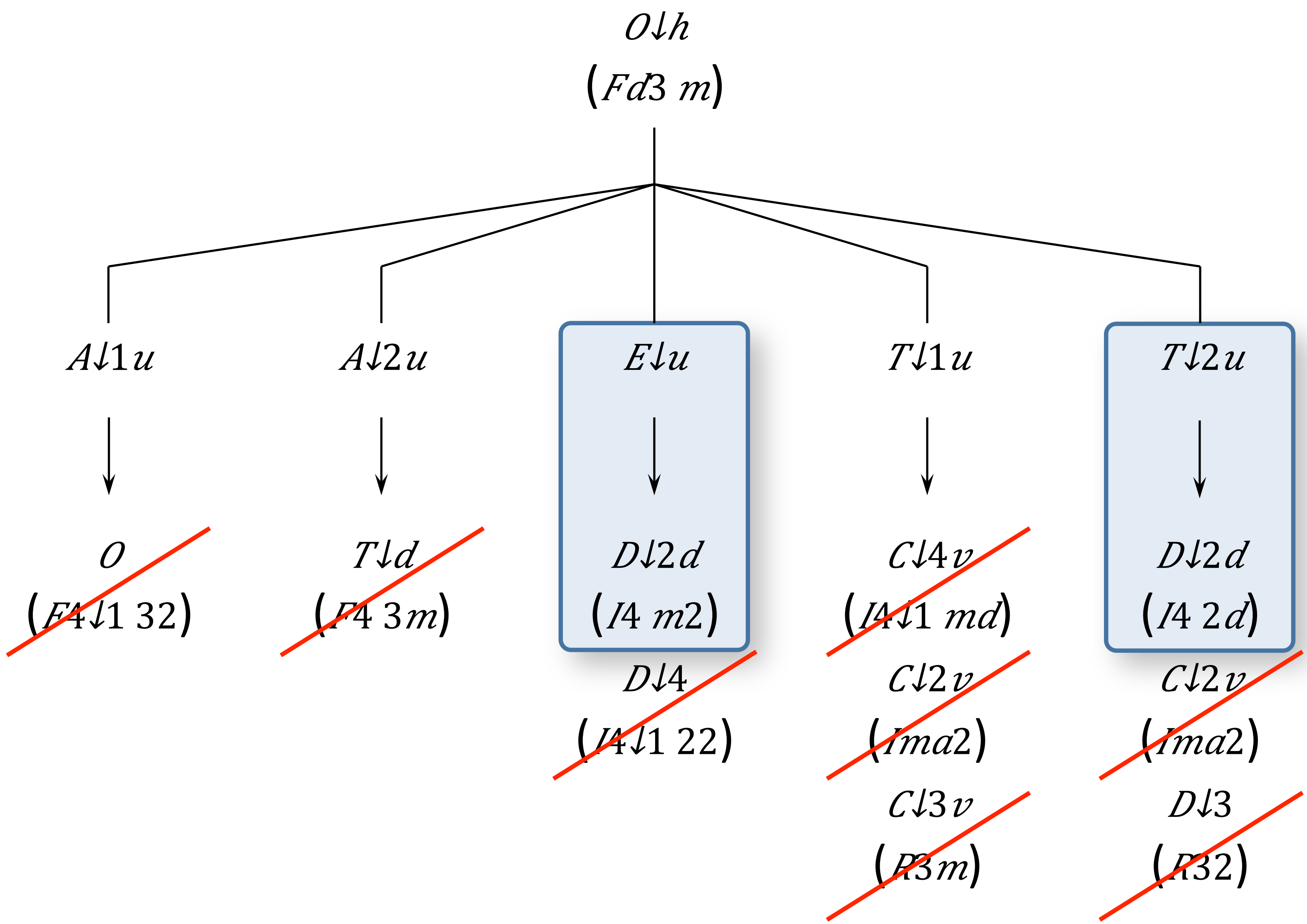




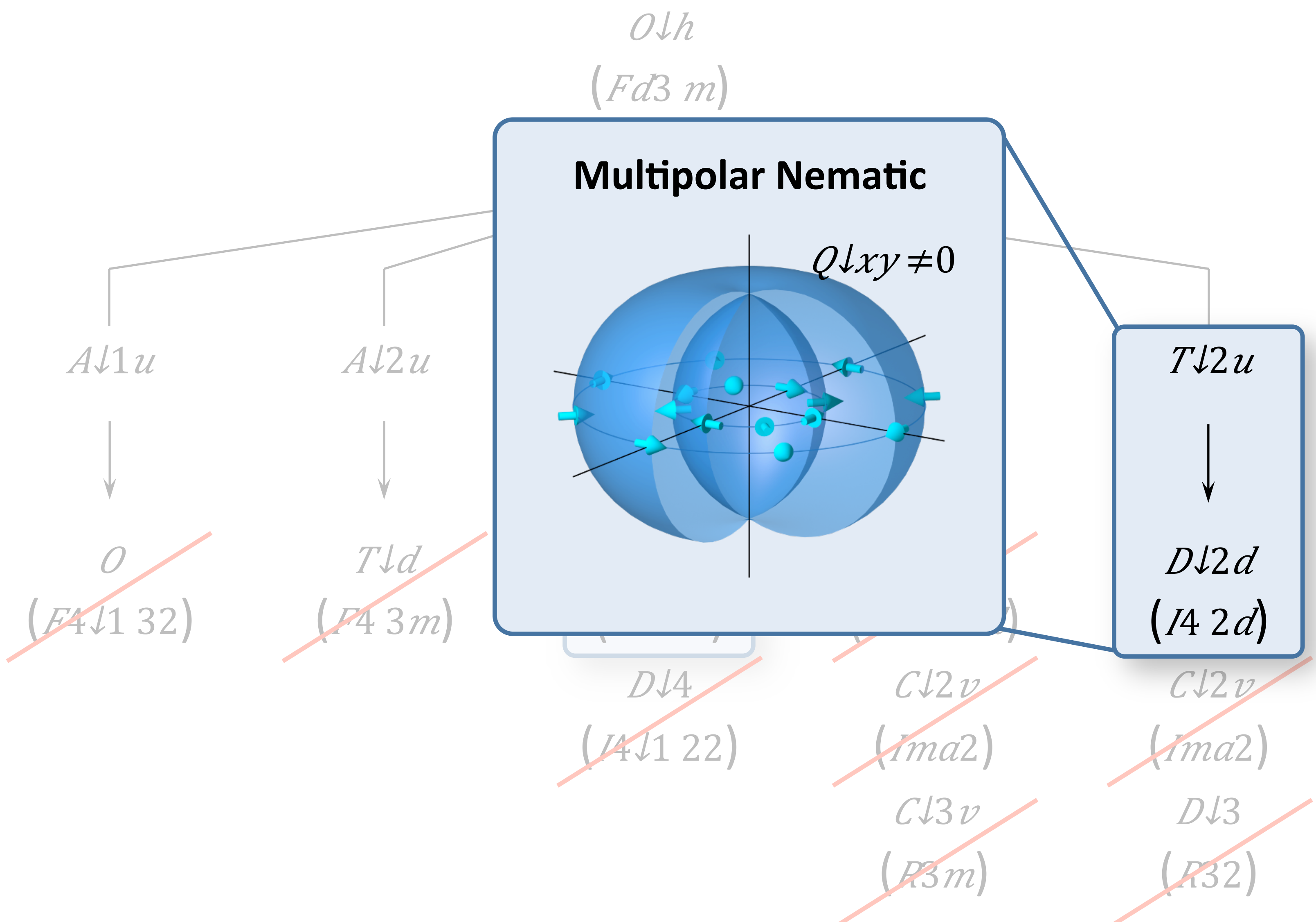
Rotational anisotropy measurements



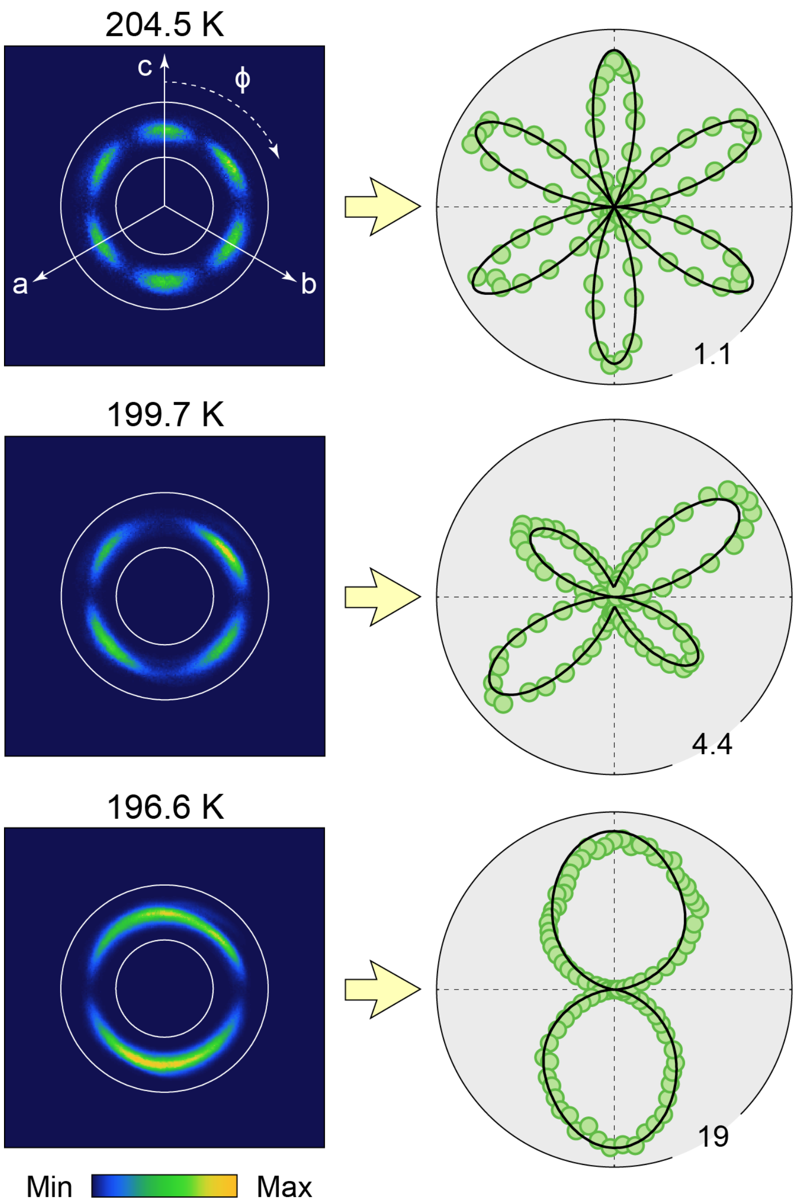
Irreducible representations of O_h



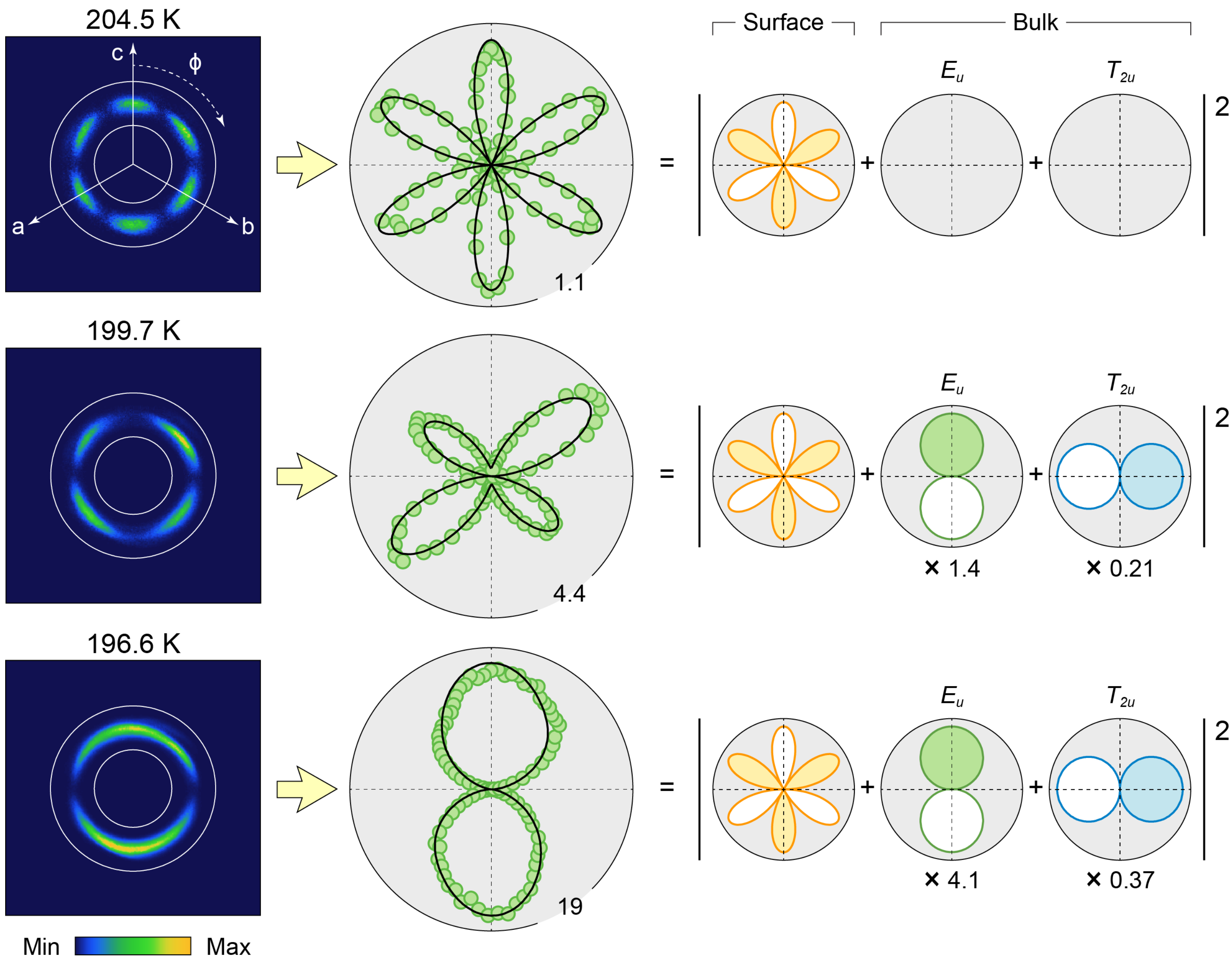
Irreducible representations of O_h



Rotational anisotropy measurements

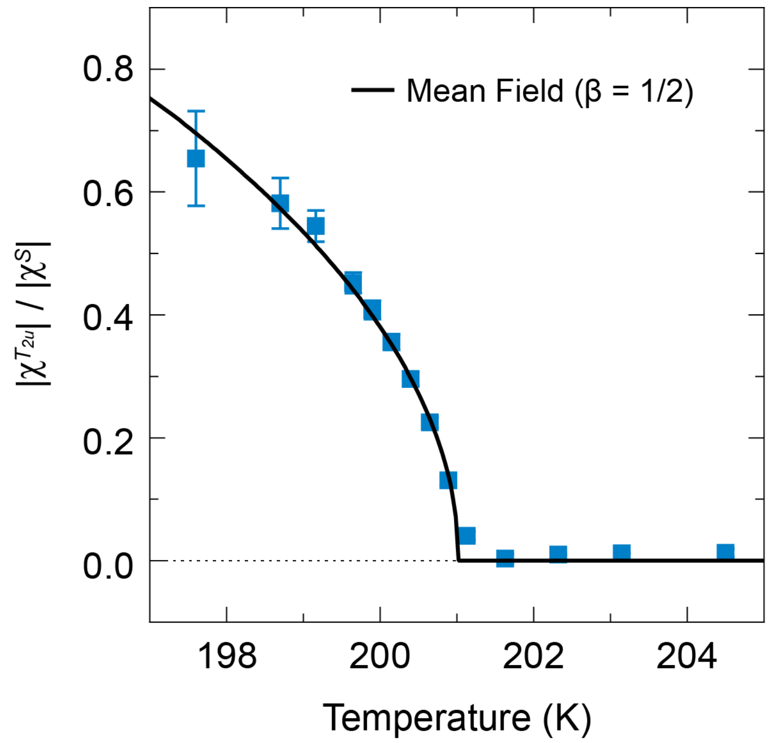


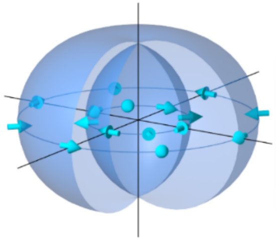
Rotational anisotropy measurements



Temperature dependence of order parameters

T_{2u} (Electronic)

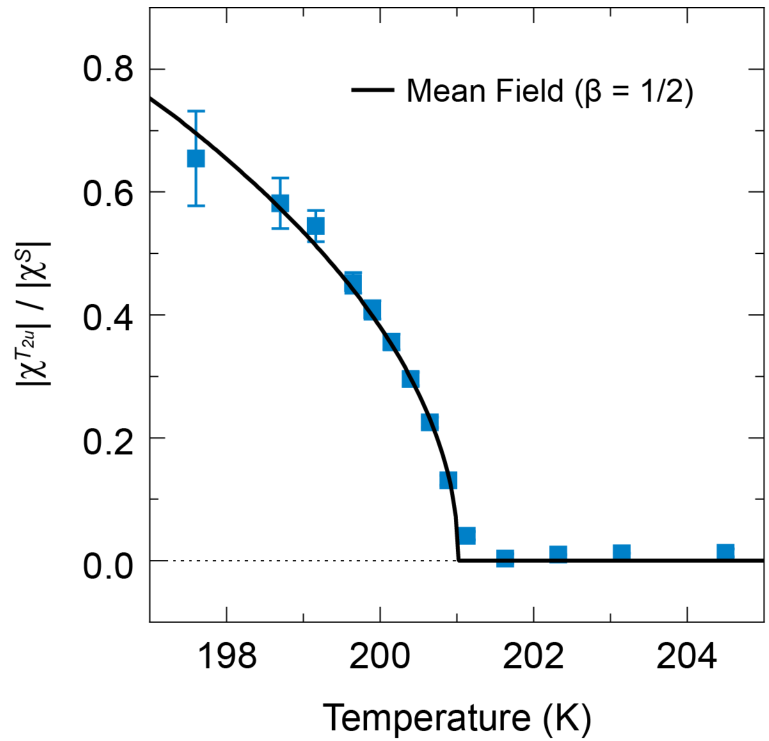




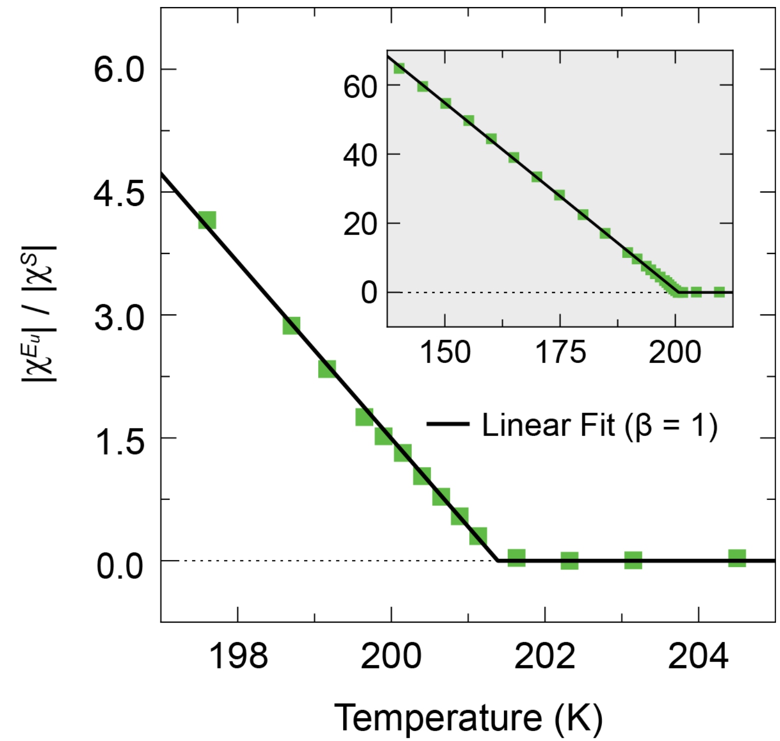
$$\Phi_{T_{2u}} \propto |1 - T/T_c|^{1/2}$$

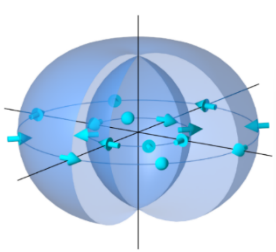
Temperature dependence of order parameters

T_{2u} (Electronic)

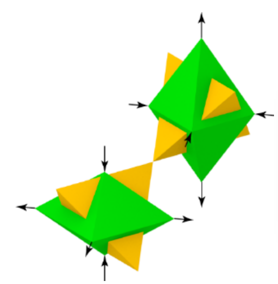


E_u (Structural)





$$\Phi_{T_{2u}} \propto |1-T/T_c|^{1/2}$$



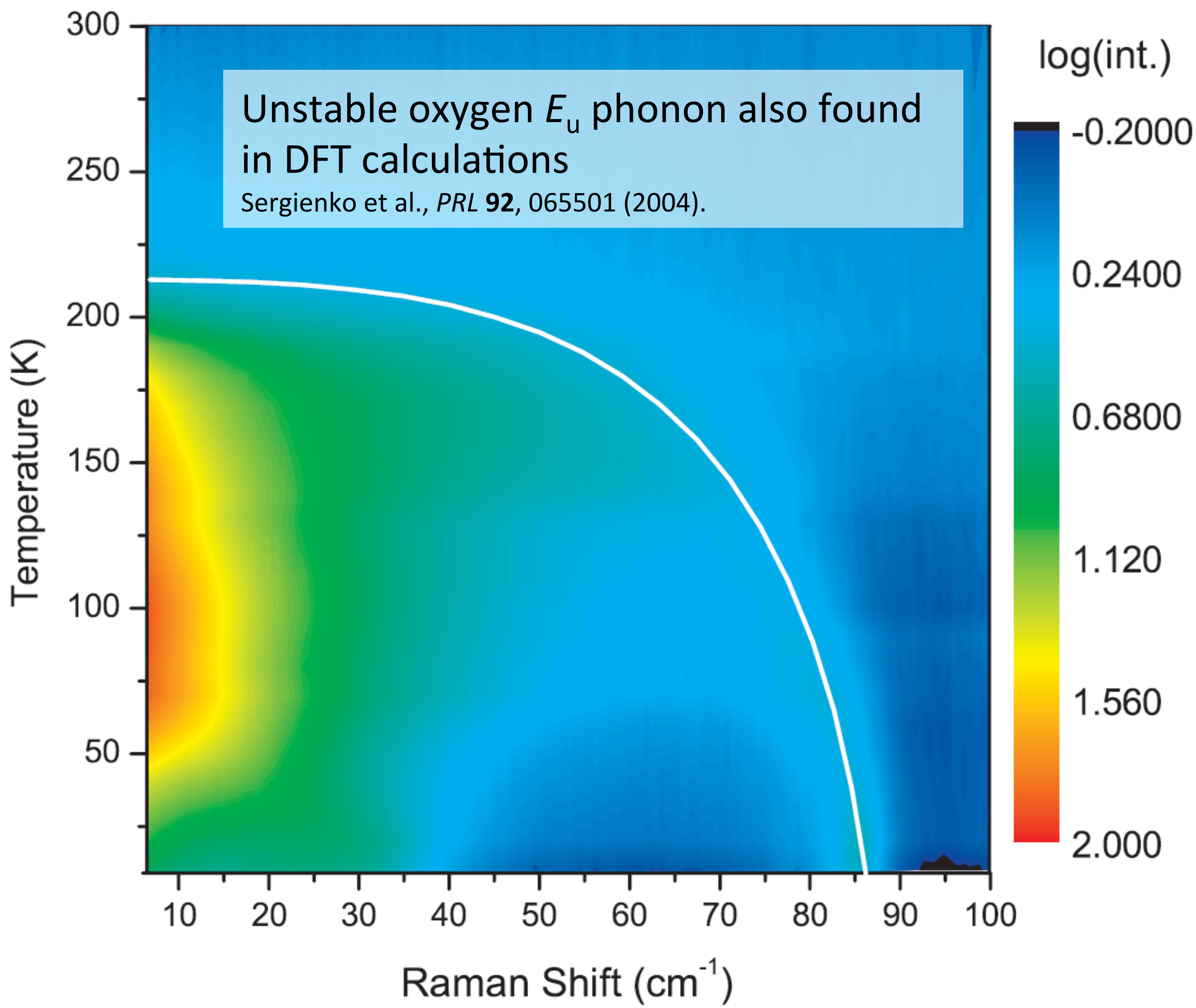
$$\Phi_{E_u} \propto |1-T/T_c|$$

Part Four

Evidence of an improper displacive phase transition in $Cd_2Re_2O_7$

J. Harter *et al.*, (*in preparation*)

Evidence for a soft mode from Raman spectroscopy



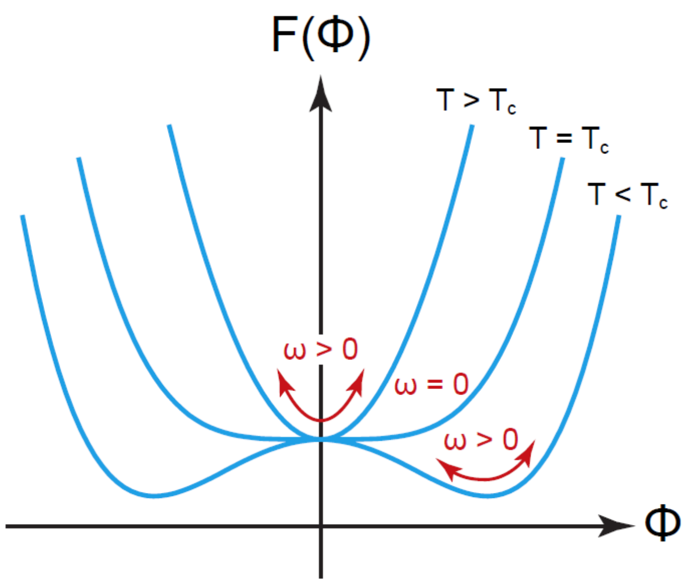
Kendziora *et al.*, Phys. Rev. Lett. **95**, 125503 (2005).

Two competing interpretations

Interpretation 1 (Raman, DFT…)

The E_u structural order parameter (Φ) is soft and freezes at T_c

$$F(\Phi)=a(T/T_c-1)\Phi^2+b\Phi^4$$

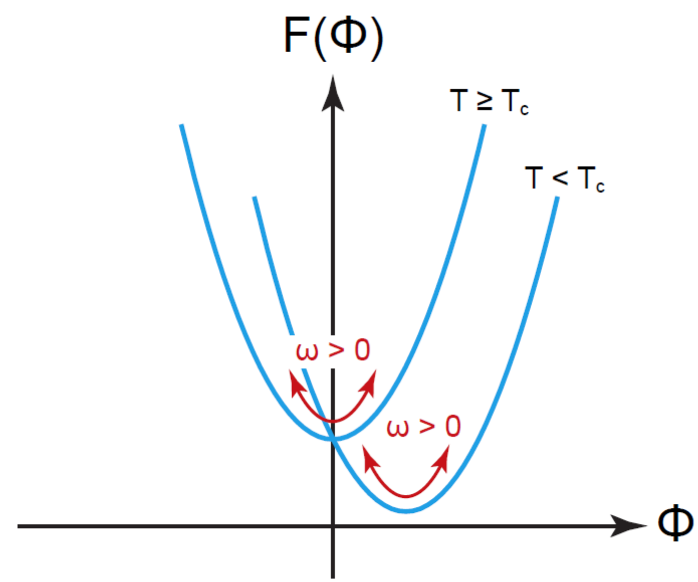


Soft Mode

Interpretation 2 (SHG, x-ray…)

The E_u structural order parameter (Φ) is secondary, carried along by (two) primary electronic order parameters (Ψ)

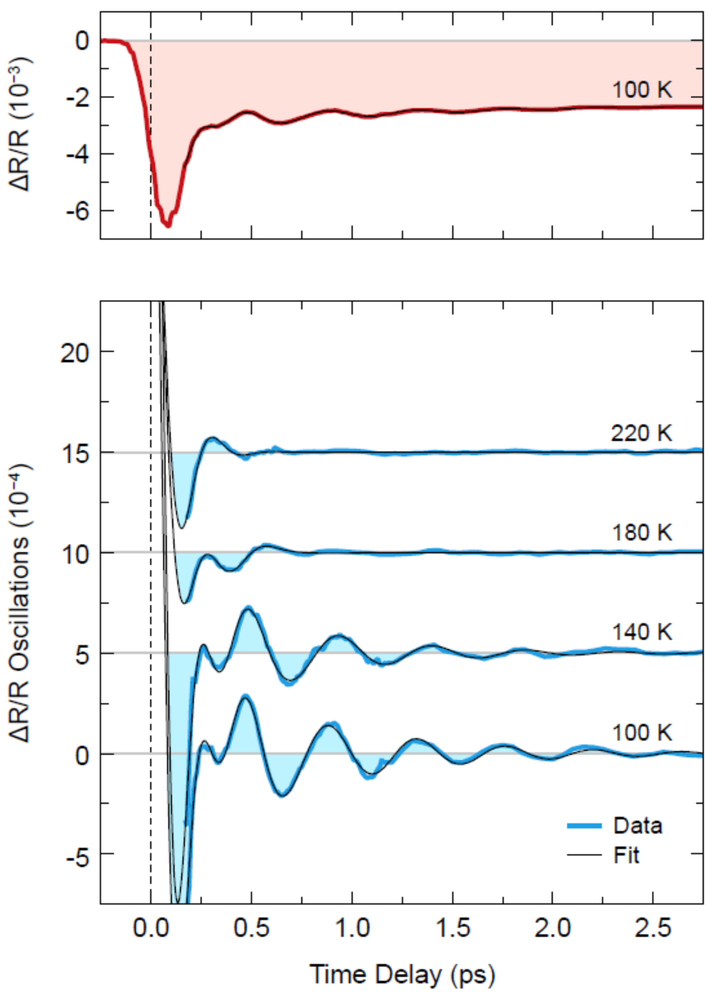
$$F(\Phi)=a\Phi^2-g\Psi_{T_{1g}}\Psi_{T_{2u}}\Phi$$



Secondary Order

J. W. Harter, D.H. et al., in preparation

Time-resolved optical reflectivity



- Data can be fit to the equation:

$$\frac{\Delta R(t)}{R} = B_0 + B_1 e^{-\gamma_1 t} + B_2 e^{-\gamma_2 t} + \sum_{i=1,2} A_i x_i(t)$$

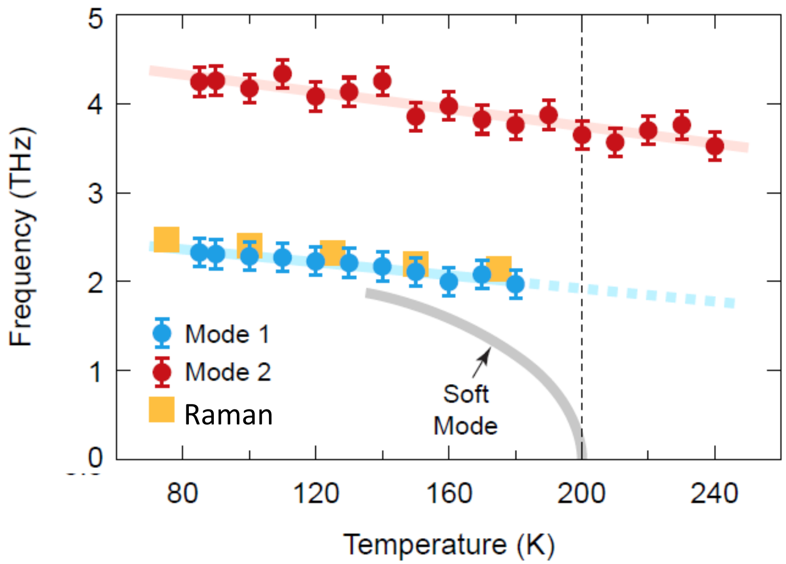
- $x_i(t)$ is the response of a damped harmonic oscillator:

$$x_i(t) = e^{-t/\tau_i} \left[\cos(\Omega_i t) + \frac{\sin(\Omega_i t)}{\Omega_i \tau_i} \right]$$

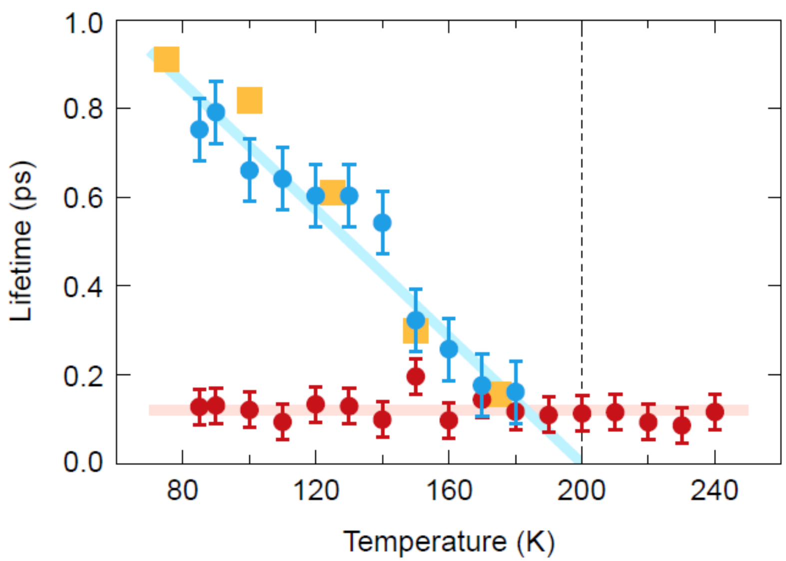
$$\Omega_i = \sqrt{\omega_i^2 - 1/\tau_i^2}$$

J. W. Harter, D.H. et al., in preparation

Time-resolved optical reflectivity



- Two modes observed
  Mode 1 $\rightarrow E_u$
  Mode 2 $\rightarrow A_{1g}$
- **No mode softening!**

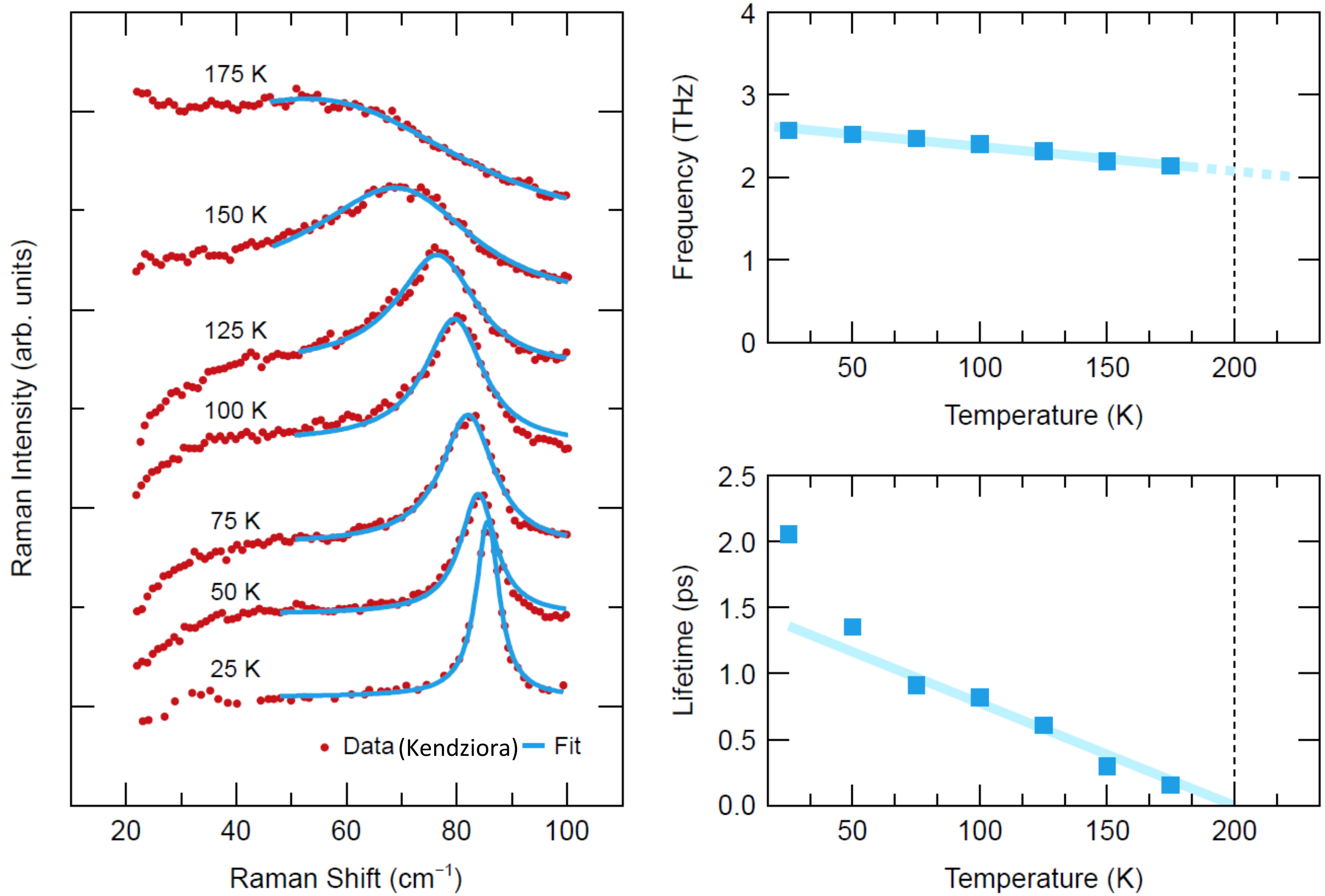


- Dramatic suppression of Mode 1 lifetime approaching T_c
- Explains spurious appearance of soft phonon in Raman data

J. W. Harter, D.H. et al., in preparation

Re-analysis of Raman data with damped SHO

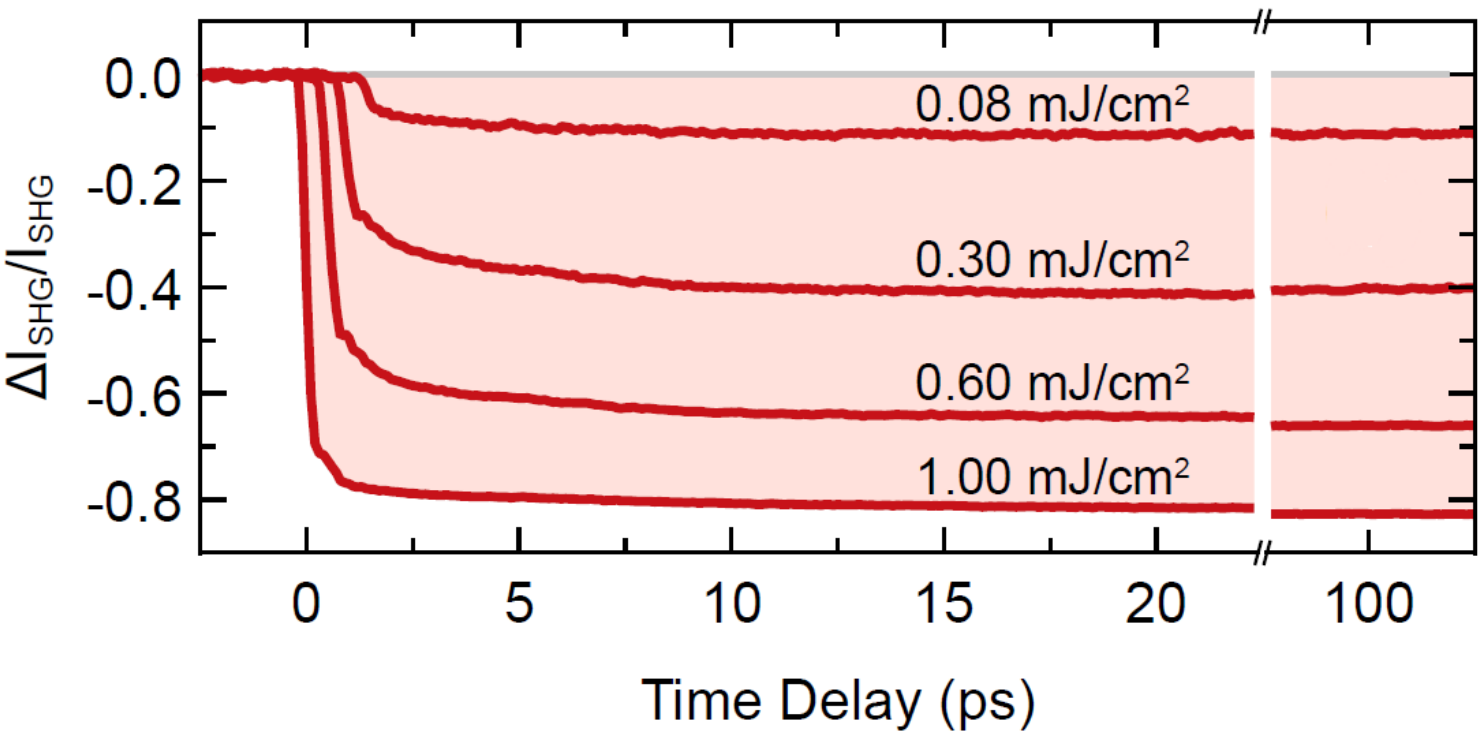
- If the Raman data is reinterpreted within a damped harmonic oscillator model ($\Omega = \sqrt{\omega^2 - 1/\tau^2}$), it agrees with our data:



J. W. Harter, D.H. et al., in preparation

Why is the E_u phonon lifetime non-trivial?

- Time-resolved SHG (ultrafast thermometry) shows strong e-ph coupling.
- Time-dependent Ginzburg-Landau theory (Kennes & Millis) shows coupling of the form $F(\Phi) = a\Phi^2 - g\Psi_{T_{1g}}\Psi_{T_{2u}}\Phi$ naturally leads to phonon lifetime scaling as $\tau \propto 1 - T/T_c$.



J. W. Harter, D.H. et al., in preparation

Conclusions

- Evidence of a T_{2u} electronic order observed in $Cd_2Re_2O_7$ observed by RA-SHG below T_c = 200 K, consistent with parity-breaking multipolar nematic order.

- Time-resolved coherent phonon spectroscopy show that the E_u phonon in $Cd_2Re_2O_7$ does ***not*** soften and therefore does ***not*** drive the phase transition, as originally thought, supporting our static RA-SHG work.

- The E_u phonon lifetime nevertheless has a nontrivial temperature dependence (approaching zero at T_c). Time-resolved SHG and time-dependent Ginzburg-Landau theory shows this is due to strong e-ph coupling and a linear coupling between the electronic and structural order parameter.

Outlook

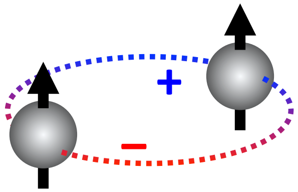
- Our Landau theory analysis indicates an additional T_{1g} (inversion symmetric) electronic order parameter coupled to the T_{2u} order.

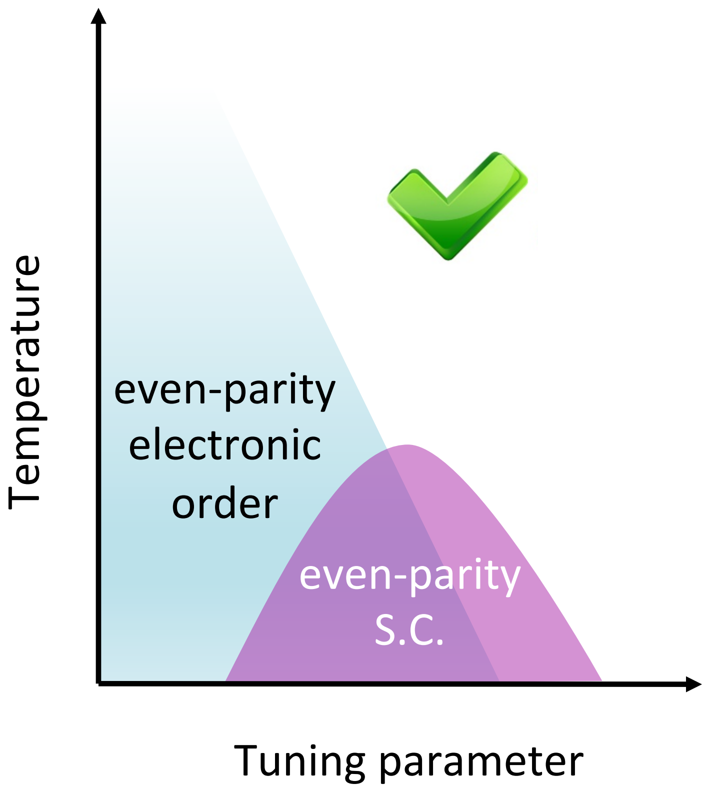
- Recent theory shows possible magnetic ferro-quadrupolar order below T_c that couples to SHG (Di Matteo & Norman https://arxiv.org/abs/1707.03807).

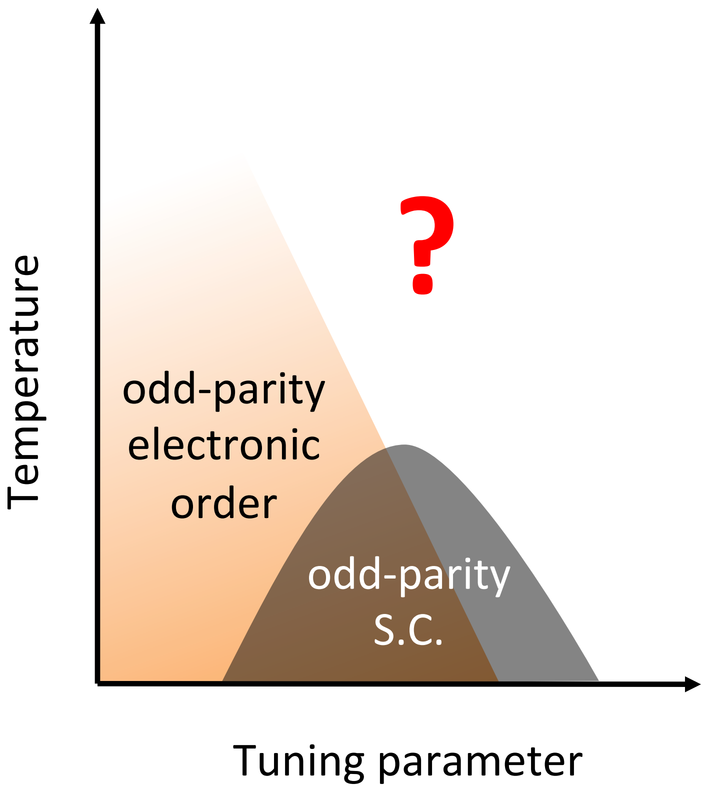
Outlook: a possible route to odd-parity superconductivity?

Odd-parity fluctuations can induce triplet *p*-wave Cooper pairing, providing a route to topological superconductivity.

- Kozii & Fu, *Phys. Rev. Lett.* **115**, 207002 (2015).
- Wang *et al.*, *Phys. Rev. B* **93**, 134512 (2016).







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