Determination of Magnetic Symmetry by Optical Second Harmonic Generation

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The Idea

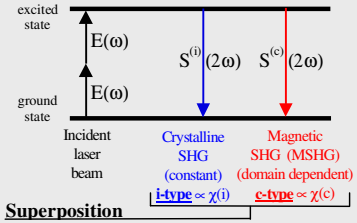
Magnetic symmetry: access by neutron or res. x-ray diffraction:

- Large-scale (n or x-ray source)
- Large samples required
- Not unambiguous
- Low spatial resolution (>100 μm)

Other techniques with access to magnetic structures?

The Method:

Second harmonic generation (SHG)



Source term for SHG: $P_i(2\omega) = \varepsilon_0 \chi^{SH}_{ijk} E_j(\omega) E_k(\omega)$

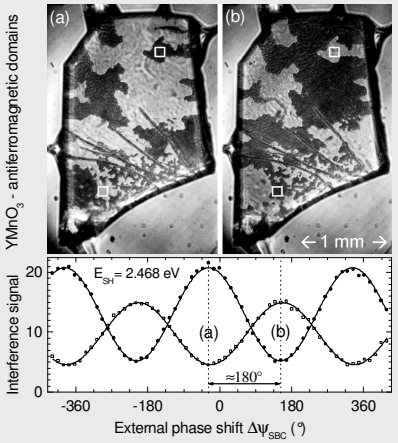
Intensity of SH signal:

$$I_{SH} \propto |P(c) + P(i)|^2$$
$$\propto |\chi(c) + A e^{i\varphi} \chi(i)|^2 I^2(\omega)$$
$$= \underbrace{(\chi^2(c) + A^2\chi^2(i)}_{\text{always} > 0} + \underbrace{2A\chi(c)\chi(i)\cos\varphi)}_{\text{interference term}} I^2(\omega)$$

A : amplitude ratio of i-type and c-type terms
φ : phase shift between complex contributions
A and φ can be fully controlled in experiment

Susceptibility $\chi_{ijk}(c)$ couples linearly to magnetic symmetry!

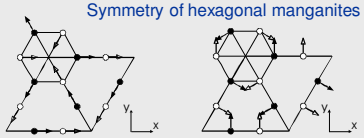
Phase Control



Novel Degrees of Freedom by MSHG

Spectroscopy

Symmetry of hexagonal manganites



Possible spin structures of Mn spins

Space group $P6_3cm$ $\chi_{yyy} \neq 0$, $\chi_{xxx} = 0$

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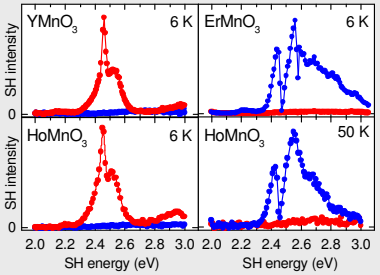
SH selection rules

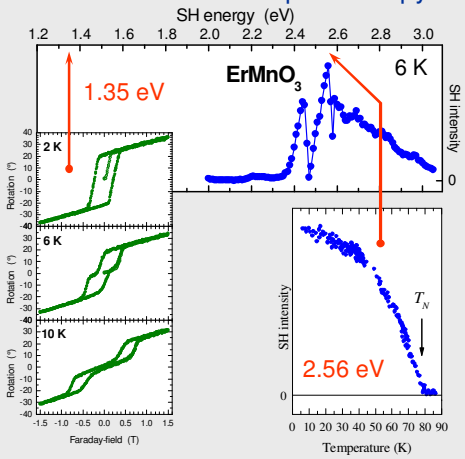


Spectra
- SH(χ_{xxx})
- SH(χ_{yyy})

Poster 1T-05

Symmetry, not substance, determines the SH spectrum!
Determination of symmetry alone from the spectra!

Sublattice-selective spectroscopy:



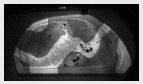
Twofold magnetic ordering in rare-earth manganites:
Mn - antiferromagnetic (79 K), observed at > 2.35 eV
Er - ferrimagnetic (4.9 K), observed at < 1.50 eV

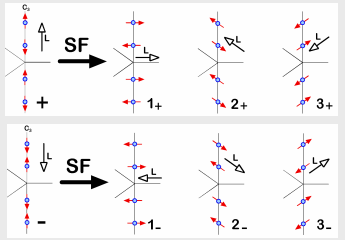
Spatial Resolution

Distinction between domains in spin-flopped Cr_2O_3

Cr_2O_3

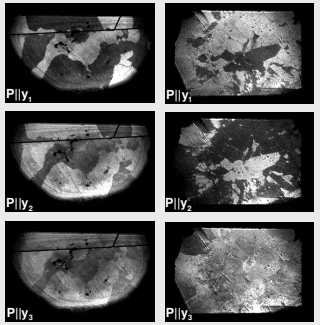
in zero magnetic field with circularly pol. light:



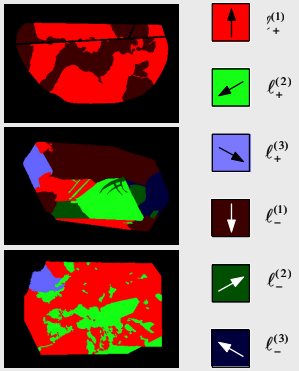


Two 180° domains

Six spin-flop domains



SH intensity with light polarized along the three y axes in the trigonal crystal



2 180° domains
1 orientation (y_1)

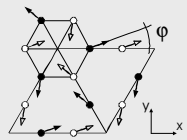
1 180° domain (+)
3 orientations

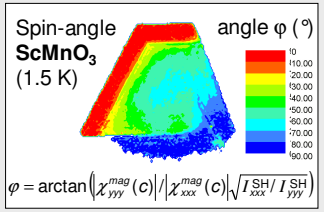
2 180° domains
3 orientations

Spins oriented along y axis (C_2 axis) in the spin-flop phase
All six domains can be clearly distinguished by MSHG
Integral techniques average over domains ⇒ wrong symmetry!

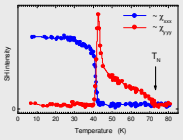
Spin Topography

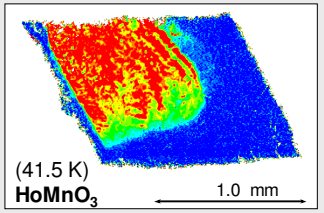
Frustrated Mn^{3+} lattice:





Spin reorientation in $HoMnO_3$ below T_N:





- Phase coexistence due to very small *xy*-plane anisotropy
- Average spin angle isn $ScMnO_3$ is 70.2° ($P6_3$ symmetry)
- This is not the correct local symmetry which may be higher ($P6_3cm$ or $P6_3cm$)

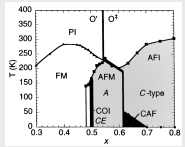
⇒ Wrong symmetry by techniques without spatial resolution

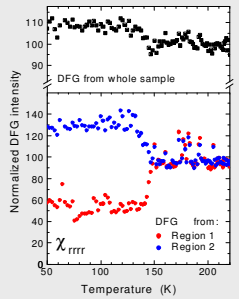
The Next Step:

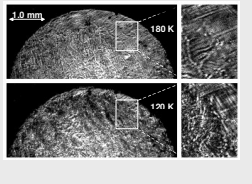
Magnetic 3-Photon Processes

Colossal magnetoresistive manganites:
SHG forbidden due to inversion symmetry of perovskite lattice
Magnetic 3-photon difference frequency generation in reflection

$Nd_{0.50}Sr_{0.50}MnO_3$: antiferromagnetic, charge and orbital ordering below 150 K







Below 150 K interference with **MDFG** contribution and formation of very small domains

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