Symmetry and coupling of magnetic and electric order parameters in $YMnO_3$

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Ferroelectromagnetic $R MnO_3$

Ferroelectromagnetism:
Simultaneous electric *and* magnetic ordering

Hexagonal maganites $RMnO_3$ (R = Sc, Y, Ho, Er, Tm, Yb, Lu)

$T < T_C \approx$ 600-1000 K ⇨ ferroelectric (FE) + paramagnetic (PM)

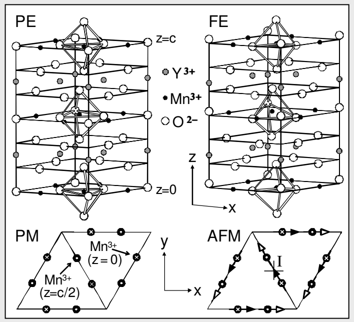
$T < T_N \approx$ 70-130 K ⇨ ferroelectric (FE) + antiferromagnetic (AFM)

Question: Nature of the coupling between the ferroelectric and antiferromagnetic order?

Experimental method: Second harmonic generation (SHG)

Polarization dependent spectroscopy
+
Nonlinear phase sensitive domain topography

Electric and magnetic ordering of $YMnO_3$



Ferroelectric phase transition:

Breaking of inversion symmetry I!

Order parameter: P

Antiferromagnetic phase transition of the Mn^{3+} sublattice:

Breaking of time-reversal symmetry T, but *not* of inversion symmetry I!

Order parameter: ℓ

Optical second harmonic generation (SHG)

In general: Multipole expansion of source term $\vec{S}$ for SHG:

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

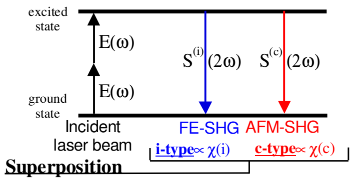
⇨ Three nonlinear contributions:

Electric dipole (ED): $\vec{P}^{NL}(2\omega) \propto \hat{\chi}^{ED} : \vec{E}(\omega)\vec{E}(\omega)$

Magnetic dipole (MD): $\vec{M}^{NL}(2\omega) \propto \hat{\chi}^{MD} : \vec{E}(\omega)\vec{E}(\omega)$

Electric quadrupole (EQ): $\hat{Q}^{NL}(2\omega) \propto \hat{\chi}^{EQ} : \vec{E}(\omega)\vec{E}(\omega)$

SHG of electric-dipole type:



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

Intensity of SH signal: $I_{SH} \propto |P(c) + P(i)|^2$

$$\propto |\chi(c) + A e^{i\varphi} \chi(i)|^2 I^2(\omega)$$

$$= (\chi^2(c) + A^2 \chi^2(i) + 2A\chi(c)\chi(i)\cos\varphi) I^2(\omega)$$

always > 0 interference term

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 χ couples linearly to symmetry!

Order parameter O ⇨ $\chi \equiv \chi(\mathcal{O}) \propto \mathcal{O}$

Observation of ferro-electromagnetic SHG

Order	Space group	Symmetry operation	Order parameter
PE + PM	$P6_3/mmc$	I, T, IT	---
FE + PM	$P6_3cm$	T	P
PE + AFM	$P\underline{6}_3/\underline{m}c\underline{m}$	I	ℓ
FE + AFM	$P\underline{6}_3c\underline{m}$	---	$P \cdot \ell$

Expansion of coupling of SHG to the order parameters:

$$\vec{P}^{NL}(2\omega) = \epsilon_0(\hat{\chi}^{ED}(0) + \hat{\chi}^{ED}(\mathcal{P}) + \hat{\chi}^{ED}(\ell) + \hat{\chi}^{ED}(\mathcal{P}\ell))\vec{E}(\omega)\vec{E}(\omega)$$

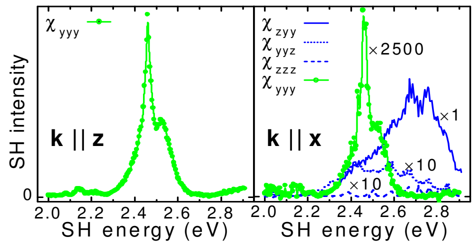
Lowest order non-zero contributions to SHG:

Zero order: Electric dipole (ED): $\hat{\chi}^{ED}(P) = i_1, i_2, i_3$ and $\hat{\chi}^{ED}(P \cdot \ell) = e_1$

First order: Magnetic dipole (MD): $\hat{\chi}^{MD}(\ell) = m_1$

First order: Electric quadrupole (EQ): $\hat{\chi}^{EQ}(\ell) = q_1, q_2, q_3$

		$S^{ED}(\mathcal{P})$	$S^{ED}(\mathcal{P}\cdot\ell)$	$S^{MD}(\ell)$	$S^{EQ}(\ell)$
k ∥ x	S_y	$2i_1E_yE_z$	$e_1E_y^2$	---	---
	S_z	$i_2E_y^2 + i_3E_z^2$	---	---	---
k ∥ y	S_x	$2i_1E_xE_z$	---	---	$-2q_1E_xE_z$
	S_z	$i_2E_x^2 + i_3E_z^2$	---	$m_1E_x^2$	$-q_2E_x^2$
k ∥ z	S_x	---	$-2e_1E_xE_y$	$-2m_1E_xE_y$	$-2q_3E_xE_y$
	S_y	---	$e_1(E_y^2 - E_x^2)$	$m_1(E_y^2 - E_x^2)$	$q_3(E_y^2 - E_x^2)$



Identical magnetic spectra for k||z and k||x

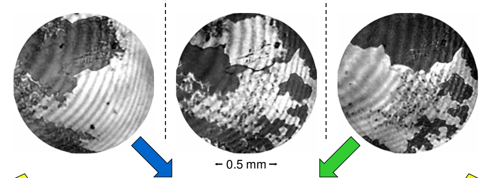
⇨ **bilinear coupling to P · ℓ.**

First observation of ferroelectromagnetic (FEM) SHG!

Observation of ferro-electromagnetic domains

Simultaneous investigation of *coexisting* electric & magnetic domains in the *same* sample!

FE domains | AFM domains | FEM domains



Independent FE and FEM domain structures!

Introduction of a FEM order parameter F ≡ P · ℓ

Clamping of order parameters:

⇨ Ferroelectric (P) and ferroelectromagnetic (F) order parameters form *independent* domains.

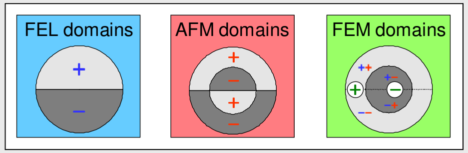
⇨ Antiferromagnetic order parameter (ℓ) does ***not*** form independent domains ⟶ AFM state is "unphysical"!

⇨ Ferroelectromagnetic domains:

F = +1 for P = +1, ℓ = +1 or P = −1, ℓ = −1

F = −1 for P = +1, ℓ = −1 or P = −1, ℓ = +1

⇨ Any reversal of the FE order parameter is **clamped** to a reversal of the AFM order prameter.

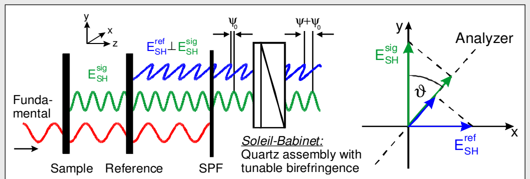


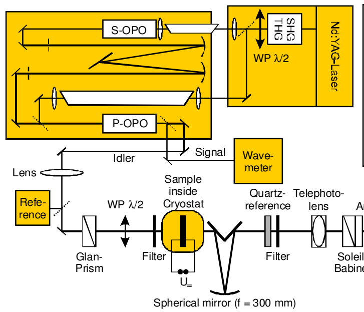
Nonlinear phase sensitive domain topography

Domain topography with external reference and tuneabal light source ⇨ problems:

(i) Propagation effects due to distance between sample ⟷ reference
(ii) Inhomogeneous beam profile
} ⇨ **Loss of contrast!**

Solution: Achromatic beam imaging of fundamental light *and* second harmonic light!





Laser system:
Frequency tripeled Nd:YAG-Laser (3 ns, max. 100 Hz)
+
Optical-parametric-oscillator (OPO)
Spectral range: 400-2800 nm

SHG/THG: Second/third harmonic generation, **OPO:** Optical-parametric oscillator, **S/P-OPO:** Seed/power OPO, **WP:** Waveplate, **$U_=$:** DC sourcemeter **MC:** Monochromator, **PM:** Photomultiplier, **CCD:** Camera, **PC:** Computer

Microscopic theory

Origin of the SH signal:

- Mn^{3+} in trigonal bipyramidal field of oxygen ligands
- Local field distorted by ferroelectric ordering and spin-orbit interaction
- Exchange between adjacent Mn^{3+} ions

⇨ Calculated spectral dependence of SH suszeptibilities:

$$\chi_{yyy} \propto \mathcal{P}\ell\left(\frac{1}{E_2 - 2\hbar\omega} + \frac{\gamma}{E_1 - 2\hbar\omega}\right) \qquad \chi_{zyy} \propto \mathcal{P}\frac{v_x}{E_1 - 2\hbar\omega}$$

E_1 = 2.46 eV, E_2 = 2.7 eV, $\gamma \ll 1$,
v_x: expectation value of crystal field induced by FEL ordering

⇨ Coupling of order parameters for the magneticaly induced SHG!

T. Iizuka-Sakano, E. Hanamura, Y. Tanabe: J. Phys.: Cond. Matt. **13**, 3031 (2001)

Origin of the clamping of FE and AFM order parameters:

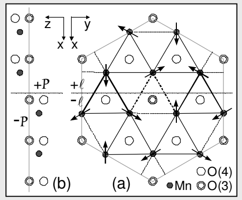
- Antiferromagnetic Mn-O-Mn superexchange
- Across FE domain wall decrease of the Mn-O(4)-Mn bond angle (3%)

⇨ Exchange energy at a FE domain wall:

$$E = -(J_{(3)} + 2J_{(4)}) \pm (J'_{(3)} + 2J'_{(4)})$$

$J_{(3,4)}, J'_{(3,4)} > 0$: Exchange integrals

$J'_{(3)} - J'_{(4)} < 0$ ⇨ Ferromagnetic coupling across FE wall!



Alternatively: Magneto-elastic effects at FE domain wall.