Magnetic Structure of Hexagonal Manganites $RMnO_3$ (R = Sc, Y, Ho, Er, Tm, Yb, Lu)

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Motivation

- Hexagonal manganites $RMnO_3$ (R = Sc, Y, Ho, Er, Tm, Yb, Lu).
- Phase transition into frustrated antiferromagnetic ordering of the Mn^{3+} ions at T_N = 70 – 130 K.
- With neutron or magnetic x-ray diffraction experiments an unambiguous determination of the antiferromagnetic structure is not possible.

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⇒ Introduction of a new optical method:

Polarization dependent spectroscopy of magnetic second harmonic (MSHG)

Method

Second-order nonlinear polarisation:

$$P_i(2\omega)=\varepsilon_0\sum_{j,k=1}^{3}\chi^{(2)}_{ijk}E_j(\omega)\,E_k(\omega)$$

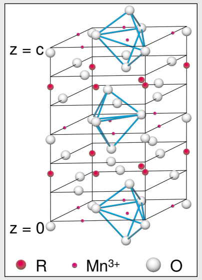
The components of the second-order nonlinear susceptibility $\chi^{(2)}_{ijk}$ are unambiguously determined by the magnetic symmetrie of the crystal!

⇒ The nonvanishing components of $\chi^{(2)}_{ijk}$ are given by measurements of the

spectral, polarization, and temperature dependence

of the nonlinear polarization $P_i(2\omega)$.

Crystal and spin structure



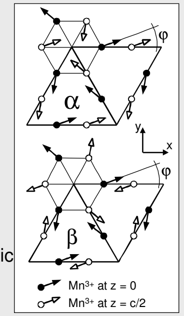
$T < T_C$: ferroelectric

↓

Symmetry: $P6_3cm$

$T < T_N$: antiferromagnetic

↓



Some magnetic symmetry and SH selection rules:

α_1 (φ = 0°): $P\underline{6_3}\underline{cm}$
$\chi_{yyy} = -\chi_{yxx} = -\chi_{xyx} = -\chi_{xxy}$

β_1 (φ = 0°): $P\underline{6_3}c\underline{m}$
$\chi_{xyz} = \chi_{xzy} = -\chi_{yxz} = -\chi_{yzx}$

α_2 (φ = 90°): $P\underline{6_3}c\underline{m}$
$\chi_{xxx} = -\chi_{xyy} = -\chi_{yxy} = -\chi_{yyx}$

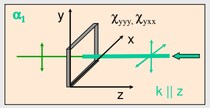
β_2 (φ = 90°): $P6_3cm$
$\chi_{zxx} = \chi_{zyy}$, $\chi_{xzx} = \chi_{xxz} = \chi_{yyz} = \chi_{yzy}$, χ_{zzz}

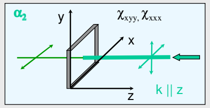
α_φ (0° < φ < 90°): $P\underline{6_3}$
$\chi(\alpha_1) \oplus \chi(\alpha_2)$

β_φ (0° < φ < 90°): $P6_3$
$\chi(\beta_1) \oplus \chi(\beta_2)$

⇒ MSHG allowed for k||z!

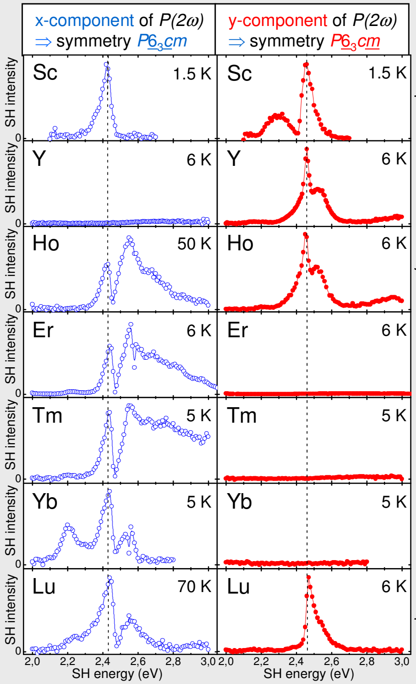
⇒ No MSHG allowed for k||z!



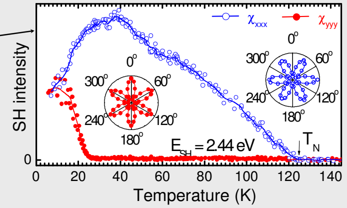


Symmetry determination of $RMnO_3$

MSHG spectroscopy:



Additional magnetic phase transitions below the Néel temperature T_N:

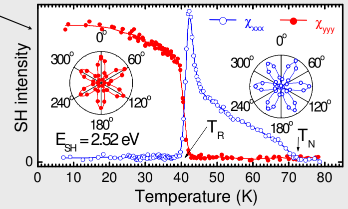


$ScMnO_3$:

$T < T_N$: Symmetry $P\underline{6_3}c\underline{m}$

$T \to 0$ K: Transfer from $P\underline{6_3}c\underline{m}$ to $P\underline{6_3}\underline{cm}$ symmetry

⇒ coexistance of magnetic phases

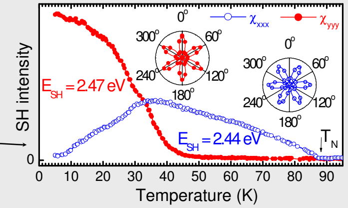


$HoMnO_3$:

90° spin rotation at T_R = 41 K

$T < T_R$: Symmetry $P\underline{6_3}\underline{cm}$

$T > T_R$: Symmetry $P\underline{6_3}c\underline{m}$



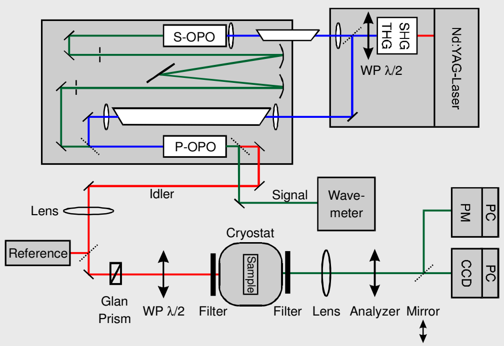
$LuMnO_3$:

$T < T_N$: Symmetry $P\underline{6_3}c\underline{m}$

$T \to 0$ K: Transfer from $P\underline{6_3}c\underline{m}$ to $P\underline{6_3}\underline{cm}$ symmetry

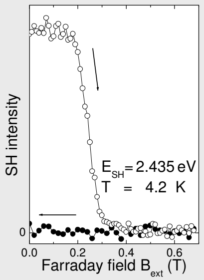
⇒ coexistance of magnetic phases

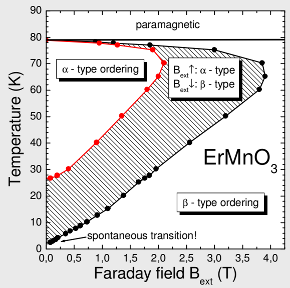
Experimental setup



SHG/THG: second/third harmonic generation, **OPO:** optical parametric oscillator, **S/P-OPO:** seed/power OPO, **WP:** waveplate, **PM:** photomultiplier, **CCD:** camera, **PC:** computer

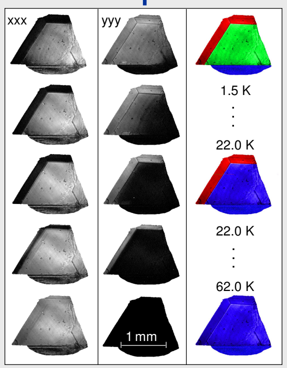
Field induced change of symmetry in $ErMnO_3$

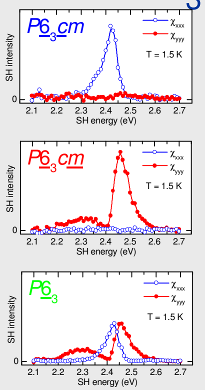




The quenching of SH signal is due to an antiferromagnetic transition from α- to β-type ordering in a magnetic field!

Coexistence of magnetic phases in $ScMnO_3$





Conclusions

- α-type ordering for all hexagonal $RMnO_3$ compounds.
- Temperature dependent phase coexistance ⇒ small in-plane anisotropy.
- Phase transition to β-type ordering for magnetic field in Faraday configuration.

Symmetry of the hexagonal manganites $RMnO_3$