# Determination of the Magnetic Symmetry of Hexagonal Manganites by Second Harmonic Generation


Th. Lottermoser[1], C. Degenhardt[1], M. Fiebig[1], D. Fröhlich[1], K. Kohn[2], St. Leute[1], V. V. Pavlov[3], and R. V. Pisarev[3]

1) Institut für Physik, Universität Dortmund, 44221 Dortmund, Deutschland
2) Department of Physics, Waseda University, 3-4-1 Okubo, Shinjuku-ku, Tokyo 169-8555, Japan
3) Ioffe Physical Technical Institute of the Russian Academy of Sciences, 194021 St. Petersburg, Russia


## Motivation

In the hexagonal manganites $RMnO_3$ (R = Y, Sc, Ho, Er, Tm, Yb, Lu), a phase transition from the paramagnetic into the antiferromagnetic phase occurs at $T_N$ = 70 – 130 K. With neutron or magnetic x-ray diffraction experiments it is *not* possible to determine unambiguously the antiferromagnetic structure of the frustrated $Mn^{3+}$ spins.

⇒ Introduction of a new optical method:

Polarisation dependent spectroscopy of magnetic second harmonic (MSHG)

## Method

Second-order nonlinear polarisation:

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

Second-order nonlinear susceptibility in magnetically ordered crystals:

$$\chi_{ijk}^{SH}=\chi_{ijk}^{cr}(i)+\chi_{ijk}^{mag}(c)$$

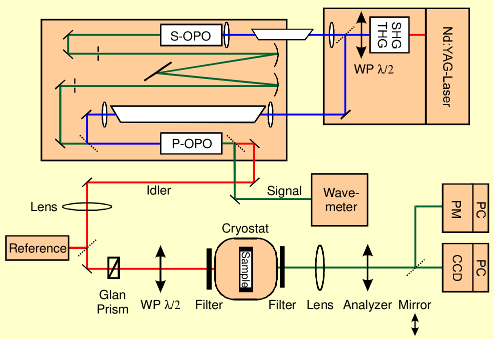
determined by **crystal structure** — determined by **magnetic structure**

Studying the structure of the nonlinear susceptibillity $\chi_{ijk}^{mag}(c)$ allows the determination of the magnetic structure. The nonvanishing components of $\chi_{ijk}^{mag}(c)$ are given by measurements of the

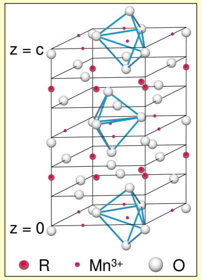
spectral, polarization, and temperature dependence.

Spatially resolved measurements lead to additional information.

## Experimental setup




## Crystal and spin structure



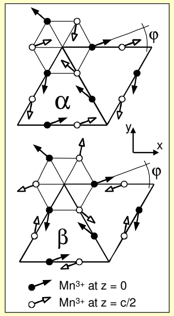

$T < T_C$ : ferroelectric

↓

Symmetry : $P6_3cm$

$T < T_N$ : antiferromagnetic

↓




Some magnetic symmetry and SH selection rules :

$\alpha_1$ ($\varphi$ = 0°): $P\underline{6_3}c\underline{m}$

$\chi_{yyy} = -\chi_{yxx} = -\chi_{xyx} = -\chi_{xxy}$

$\alpha_2$ ($\varphi$ = 90°): $P\underline{6_3}\underline{c}m$

$\chi_{xxx} = -\chi_{xyy} = -\chi_{yxy} = -\chi_{yyx}$

$\alpha_\varphi$ ($\varphi$ = 0°...90°): $P\underline{6_3}$

$\chi(\alpha_1) \oplus \chi(\alpha_2)$

⇒ MSHG allowed for k||z!

$\beta_1$ ($\varphi$ = 0°): $P6_3\underline{cm}$

$\chi_{xyz} = \chi_{xzy} = -\chi_{yxz} = -\chi_{yzx}$

$\beta_2$ ($\varphi$ = 90°): $P6_3cm$

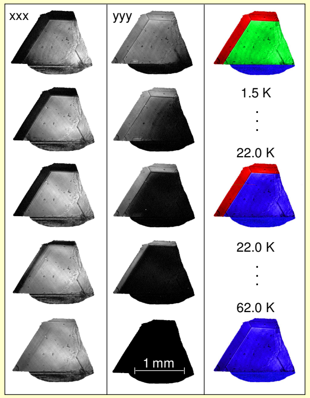
$\chi_{zxx} = \chi_{zyy}$, $\chi_{xxz} = \chi_{xzx} = \chi_{yyz} = \chi_{yzy}$, $\chi_{zzz}$

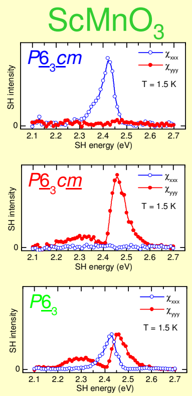
$\beta_\varphi$ ($\varphi$ = 0°...90°): $P6_3$

$\chi(\beta_1) \oplus \chi(\beta_2)$

⇒ No MSHG allowed for k||z!

## Spin-angle topography



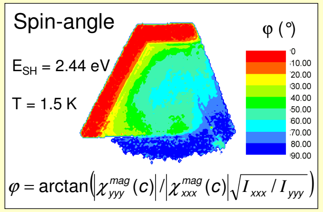




Magnetic symmetry:

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

T < 62 K: $P\underline{6_3}\underline{c}m$ + $P\underline{6_3}c\underline{m}$

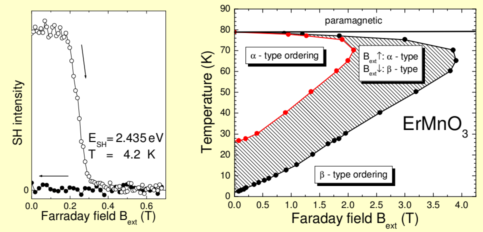
T < 22 K: $P\underline{6_3}\underline{c}m$ + $P\underline{6_3}c\underline{m}$ + $P\underline{6_3}$



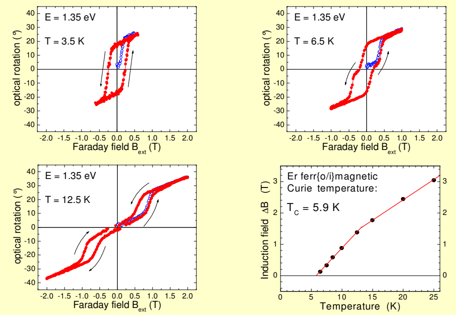

$$\varphi=\arctan\left(\left|\chi_{yyy}^{mag}(c)\right|/\left|\chi_{xxx}^{mag}(c)\right|\sqrt{I_{xxx}/I_{yyy}}\right)$$

## Interaction of Mn and Er magnetic sublattices

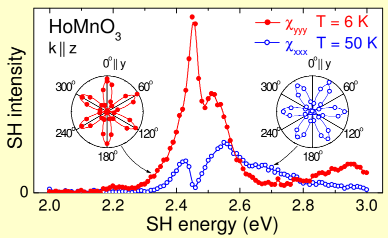
Quenching of Mn-induced SH signal due to an antiferromagnetic transition from α- to β-type ordering

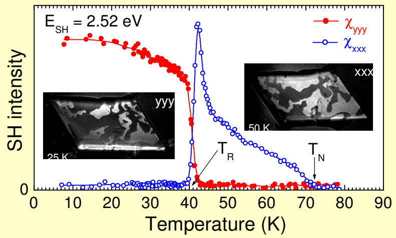



Linear optical rotation induced by Er ferro- or ferrimagnetic moment




## Phase transitions in $HoMnO_3$ and $LuMnO_3$




**$HoMnO_3$:**

Additional magnetic phase transition at 41 K:

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

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

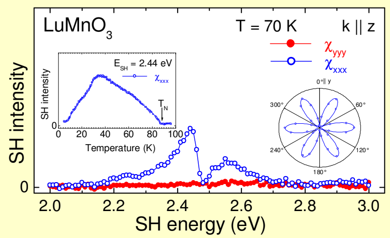
90° spin rotation at $T_R$ = 41 K

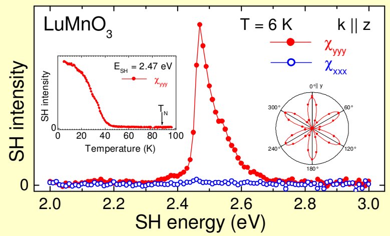
**$LuMnO_3$:**

Temperature dependence of the symmetry:

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

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




## Conclusions

- MSHG is an important complement to neutron and x-ray diffraction experiments for symmetry analysis
- Additional degrees of freedom:
  - Spectroscopy
  - Topography
- α-type ordering of hexagonal $RMnO_3$ compounds
- Phase coexistance ⇒ small in plane anisotropy

Symmetry of the hexagonal manganites $RMnO_3$