Nonlinear magneto-optics

Linear and nonlinear optical susceptibilities

Tensor formalism

Second harmonic generation:

SHG and a symmetry of a medium

SHG in magnetically-ordered media

SHG of the electric- and magnetic-dipole origins

Literature:

M. Fiebig, V. V. Pavlov, R. V. Pisarev, J. Opt. Soc. Am. B **22**, 96 (2005)

A. Kirilyuk, Th. Rasing, J. Opt. Soc. Am. B **22**, 148 (2005)

Linear and nonlinear (magneto-)optical susceptibility

Free energy expressions for a **linear nonmagnetic** medium

$$F = -1/2[\chi_{ij}(\omega)E_i(\omega)E_j^*(\omega) + \chi_{ij}^*(\omega)E_i^*(\omega)E_j^*(\omega)]$$

Polarization in a medium: $$P_j(\omega) = -\frac{\partial F}{\partial E_j^*} = \chi_{ij}(\omega)E_i(\omega)$$

Dielectric tensor: $$\varepsilon_{ij}(\omega) = 1 + \chi_{ij}(\omega)$$

Properties of linear optical susceptibility (transparent medium)

Hermitian tensor $$\chi_{ij} = \chi_{ji}^*$$

in nonmagnetic medium $$\chi_{ij} = \chi_{ij}^*$$

$\hat{\chi}(\omega)$ Real and symmetric tensor

Linear and nonlinear (magneto-)optical susceptibility

Free energy expressions for a **linear magnetic** medium

$$F = -\mathrm{Re}[\chi_{ij}(\omega)E_i(\omega)E_j^*(\omega) + \chi_{ijk}(\omega)E_i(\omega)E_j^*(\omega)M_k(0) + \cdots]$$

Crystallographic contribution

Magnetic contribution contribution
(Faraday and MO Kerr effects)

Properties of linear optical and magneto-optical susceptibility

$$\chi_{ij} = \chi_{ji}^*$$

$$\chi_{ij} = \chi_{ij}^*$$

Real symmetric

$$\chi_{ijk} = \chi_{jik}^*$$

$$\chi_{ijk} = -\chi_{ijk}^*$$

Imaginary antisymmetric

$$\begin{bmatrix} \varepsilon_0 & -\varepsilon_1 m_z & 0 \\ \varepsilon_1 m_z & \varepsilon_0 & 0 \\ 0 & 0 & \varepsilon_0 \end{bmatrix} \quad \begin{bmatrix} \varepsilon_0 & 0 & \varepsilon_1 m_y \\ 0 & \varepsilon_0 & 0 \\ -\varepsilon_1 m_y & 0 & \varepsilon_0 \end{bmatrix} \quad \begin{bmatrix} \varepsilon_0 & 0 & 0 \\ 0 & \varepsilon_0 & -\varepsilon_1 m_x \\ 0 & \varepsilon_1 m_x & \varepsilon_0 \end{bmatrix}$$

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Linear and nonlinear (magneto-)optical susceptibility

Free energy expressions for a **nonlinear nonmagnetic** medium

$$F = -\mathrm{Re}[\chi_{ijk}(\omega_3;\omega_2,\omega_1)E_i^*(\omega_3)E_j(\omega_2)E_k(\omega_1) \quad]$$

$$\omega_3 = \omega_2 + \omega_1$$

Properties of nonlinear optical susceptibility (transparent medium)

$$\chi_{ijk} - \chi^*_{jik} - \chi^*_{kij}$$

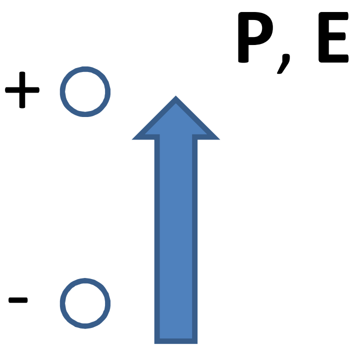
in nonmagnetic medium

$$\chi_{ijk} = \chi^*_{ijk}$$

$\hat{\chi}^{i,3}$

- Polar i-tensor of 3rd rank
- Nonzero only in noncentrosymmetric medium
- Real

Electric field/ polarization



Polar i-vector

Time-invariant

Non-invariant under the spatial inversion operation

Second harmonic generation

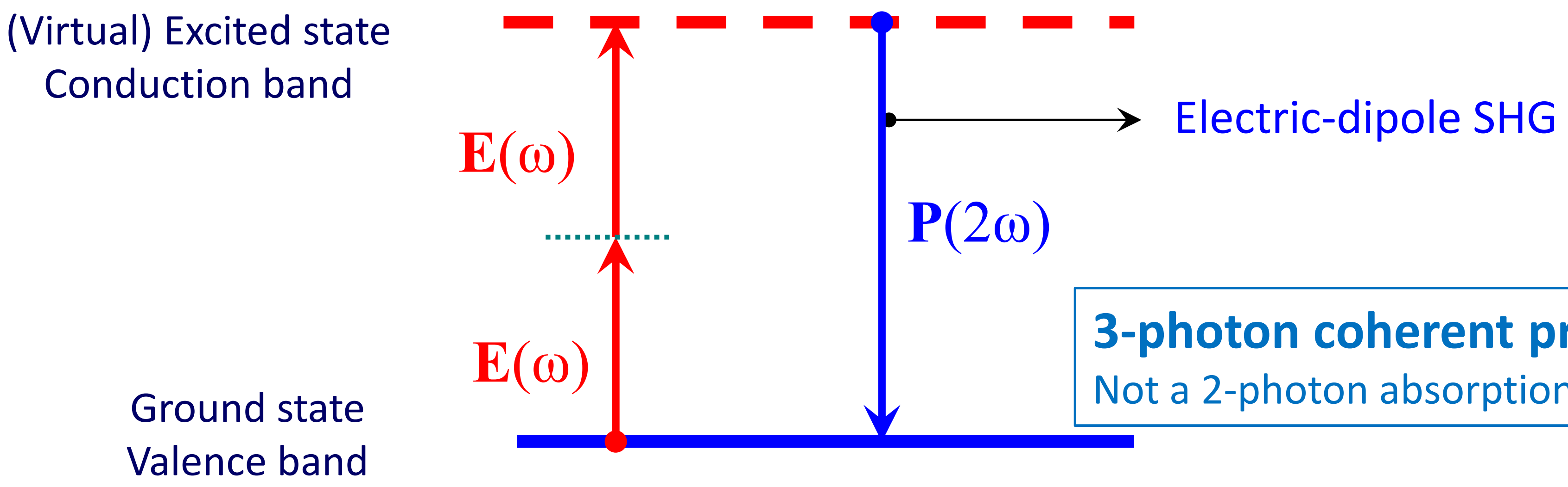
Free energy expressions for a **nonlinear nonmagnetic** medium

$$F = -\mathrm{Re}[\chi_{ijk}(\omega_3;\omega_2,\omega_1)E_i^*(\omega_3)E_j(\omega_2)E_k(\omega_1) \quad]$$

$$\omega_1 = \omega_2; \omega_3 = 2\omega$$

Nonlinear polarization in a medium:

$$P_i(2\omega) = -\frac{\partial F}{\partial E_i^*} = \chi_{ijk}(2\omega;\omega,\omega)E_i(\omega)E_j(\omega)$$



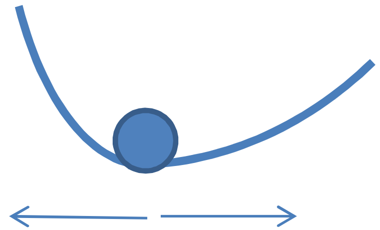
3-photon coherent process
Not a 2-photon absorption!

Origin of SHG

No inversion symmetry

⇩

Asymmetric potential for an electron



$$\frac{\partial^2 x(t)}{\partial t^2} \qquad + \omega_0^2 x(t) \qquad = \frac{e}{2m} E_o e^{-i\omega t}$$

Dissipation

Anharmonic
restoring force

Solution:

Linear response

$$x(\omega, t) = \left(q_1 e^{-i\omega t} + q_2 e^{-i2\omega t} \right)$$

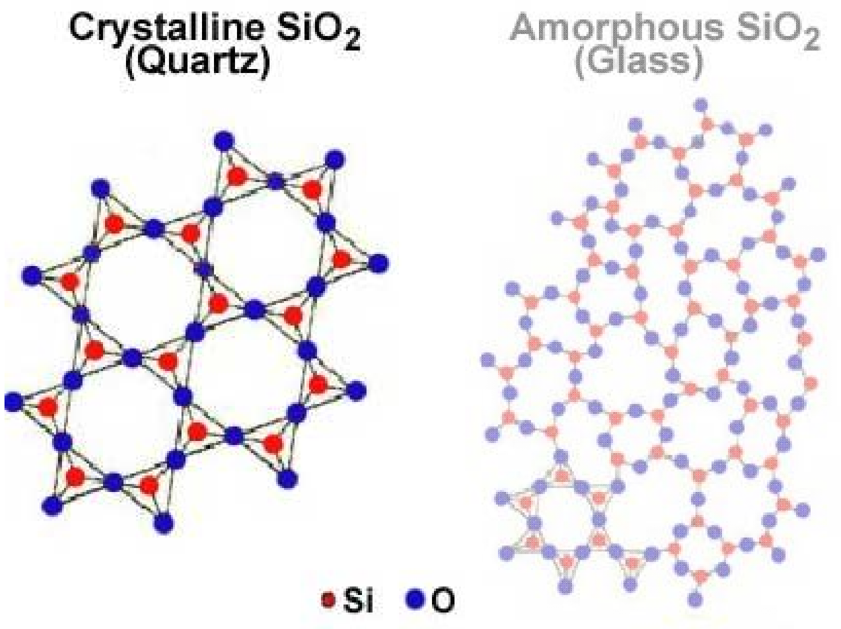
SHG

$$q_1 = \frac{eE_0}{m} \cdot \frac{1}{\omega_0^2 - \omega^2 + i\gamma \cdot \omega}$$

Resonance conditions

$$q_2 = \frac{-De^2 E_0^2}{2m^2 \left[\omega_0^2 - \omega^2 + i\gamma \cdot \omega \right]^2 \left[\omega_0^2 - (2\omega)^2 + i\gamma \cdot 2\omega \right]}$$

SHG in crystalline quartz



Polarization at a frequency 2ω

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega)$$

Refer to the Birss tables
[R. R. Birss,
Reports on Progress in Physics ***26****, 307 (1963)]*

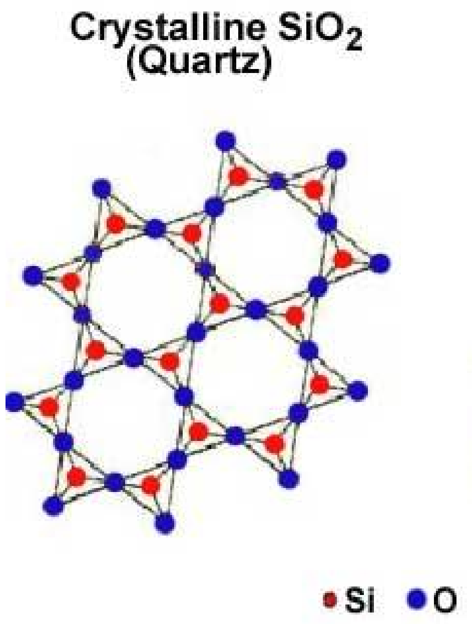
Trigonal crystal
Point group: 32

Symmetry operators:

$$1,\ 3_z,\ 2_{\perp}(3)$$

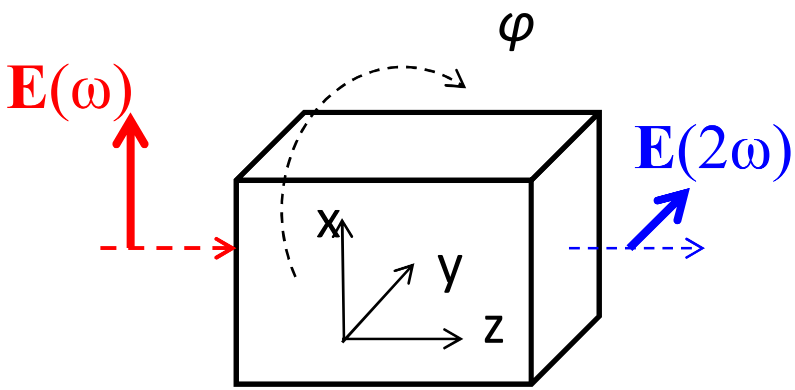
$$\hat{\chi}^{i,3}: \quad \begin{aligned} &xxx = a \\ &yyx = yxy = xyy = -xxx = -a \\ &xyz = -yxz; xzy = -yzx; zxy = -zyx \end{aligned}$$

SHG in crystalline quartz



Trigonal crystal

Point group: 32



Polarization at a frequency 2ω

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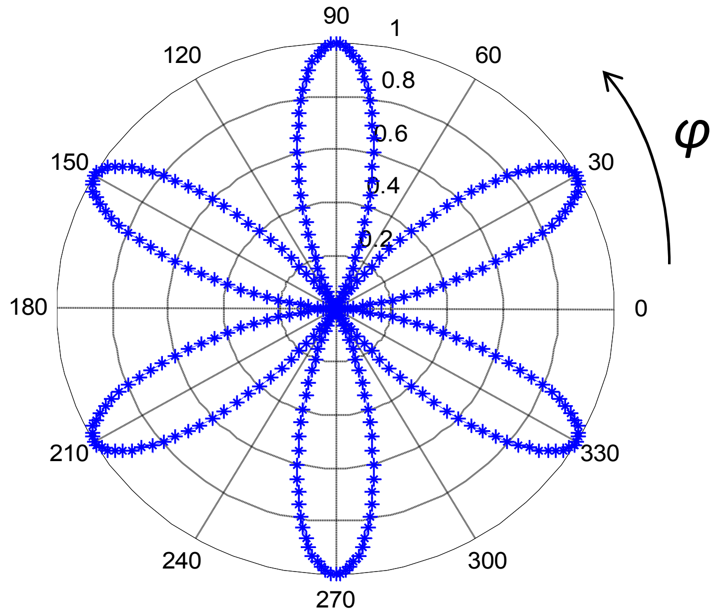
Reports on Progress in Physics ***26****, 307 (1963)]*



3-fold axis → **6-fold** polarization dependence of SHG

First experimental observation of SHG in crystalline quartz

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

pp. 118-119

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

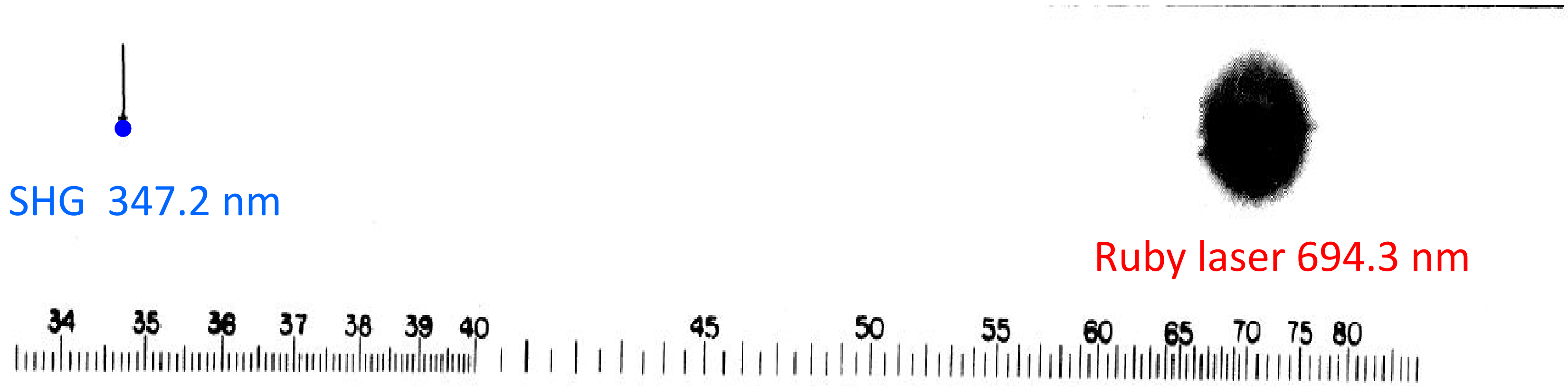


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

Magnetic SHG

- SHG in magnetically-ordered medium

  Electric-dipole SHG in noncentrosymmetryc magnetic media

- Magnetic-dipole SHG

  Beyond the electric-dipole approximation

- SHG in antiferromagnetic domains

SHG in a magnetic medium

In **noncentrosymmetric** medium only!

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega) + \hat{\chi}^{i,4}\mathbf{E}(\omega)\mathbf{E}(\omega)\mathbf{M}(0)$$

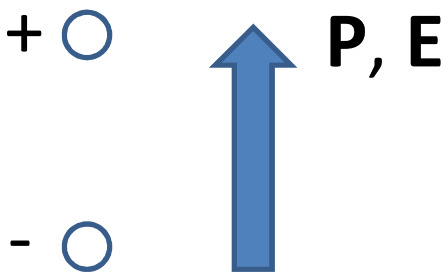
polar i-tensor

axial i-tensor

polar c-tensor

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega) + \hat{\chi}^{c,3}(\mathbf{M})\mathbf{E}(\omega)\mathbf{E}(\omega)$$

Electric field/ polarization

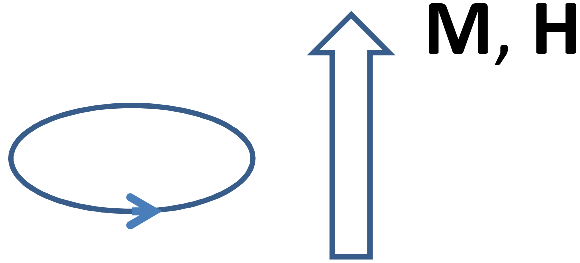


Polar i-vector

Time-invariant

Non-invariant under
the spatial inversion operation

Magnetic field/ magnetization



Axial c-vector

Time-noninvariant

Invariant under
the spatial inversion operation

SHG in magnetic medium

In **noncentrosymmetric** medium only!

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega) + \hat{\chi}^{i,4}\mathbf{E}(\omega)\mathbf{E}(\omega)\mathbf{M}(0)$$

polar i-tensor ⇕ axial i-tensor

polar c-tensor

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega) + \hat{\chi}^{c,3}(\mathbf{M})\mathbf{E}(\omega)\mathbf{E}(\omega)$$

	Crystallographic SHG	Magnetic SHG
Transparent medium:	$\hat{\chi}^{i}$ is real	$\hat{\chi}$ is imaginary

$$\mathrm{I}(2\omega) = [(\hat{\chi}^{i,3})^2 + [\hat{\chi}^{i,4}\mathbf{M}(0)]^2]\mathrm{I}^2$$

There is **no interference** between crystallographic and magnetic contributions

No sensitivity to the sign of the (anti-)ferromagnetic vector

SHG in magnetic medium

In **noncentrosymmetric** medium only!

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega) + \hat{\chi}^{i,4}\mathbf{E}(\omega)\mathbf{E}(\omega)\mathbf{M}(0)$$

polar i-tensor ⇕ axial i-tensor

polar c-tensor

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega) + \hat{\chi}^{c,3}(\mathbf{M})\mathbf{E}(\omega)\mathbf{E}(\omega)$$

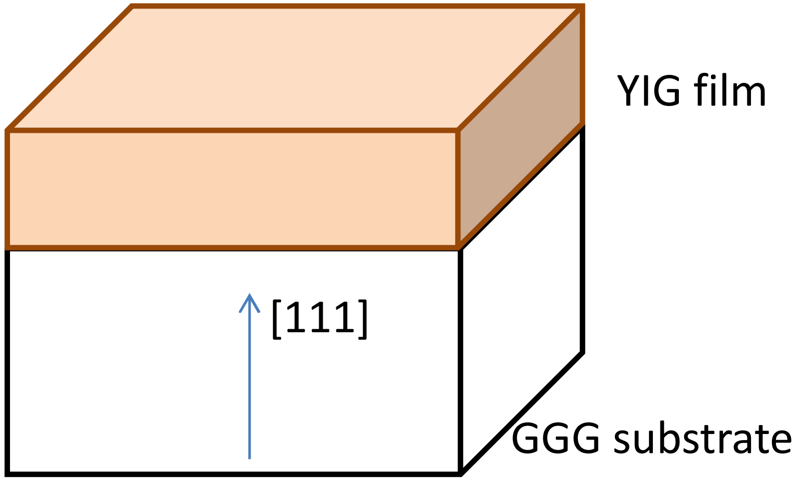
	Crystallographic SHG	Magnetic SHG
Absorbing medium:	Both $\hat{\chi}$	are complex

$$\mathrm{I}(2\omega) = [(\hat{\chi}^{i,3})^2 + (\hat{\chi}^{i,4}\mathrm{M}(0))^2 + \hat{\chi}^{i,3}\hat{\chi}^{i,4}\mathrm{M}(0)\cos(\Delta\varphi)]\mathrm{I}^2$$

Interference term

Sensitivity to the reversal of magnetization!

SHG in a ferrimagnetic iron garnet film

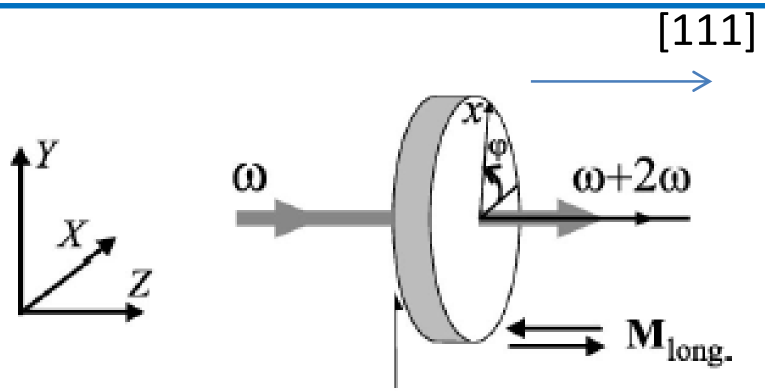


Cubic in a bulk form (no SHG)

The thin (111) film:

Trigonal system

Point group: 3*m* (z||[111])

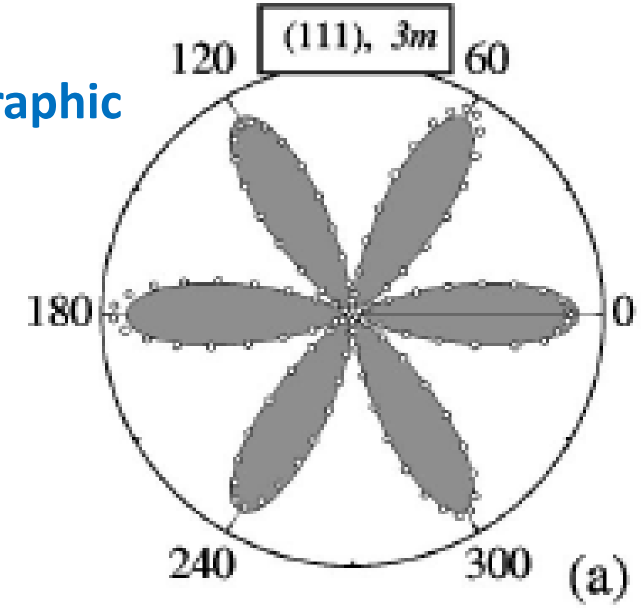


E (ω)|| y

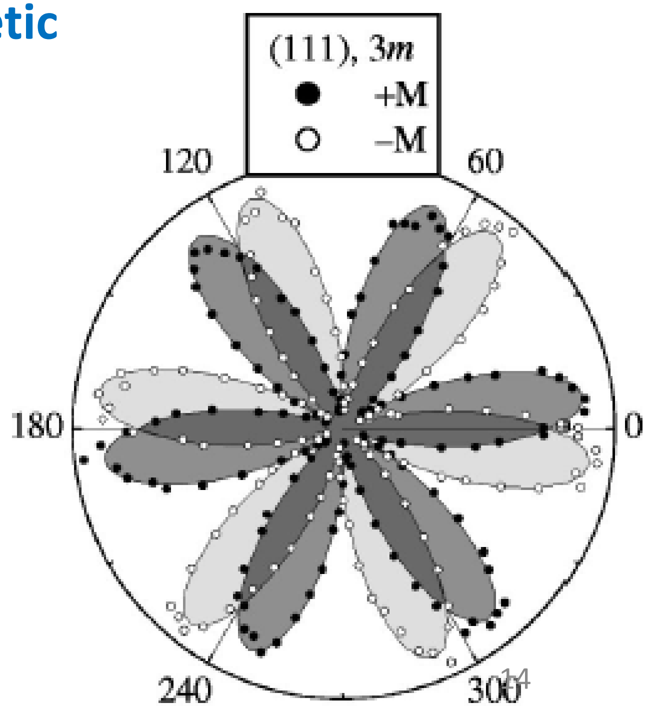
P (2ω)|| y

*[PRB **63**, 184407 (2002)]*

Crystallographic SHG

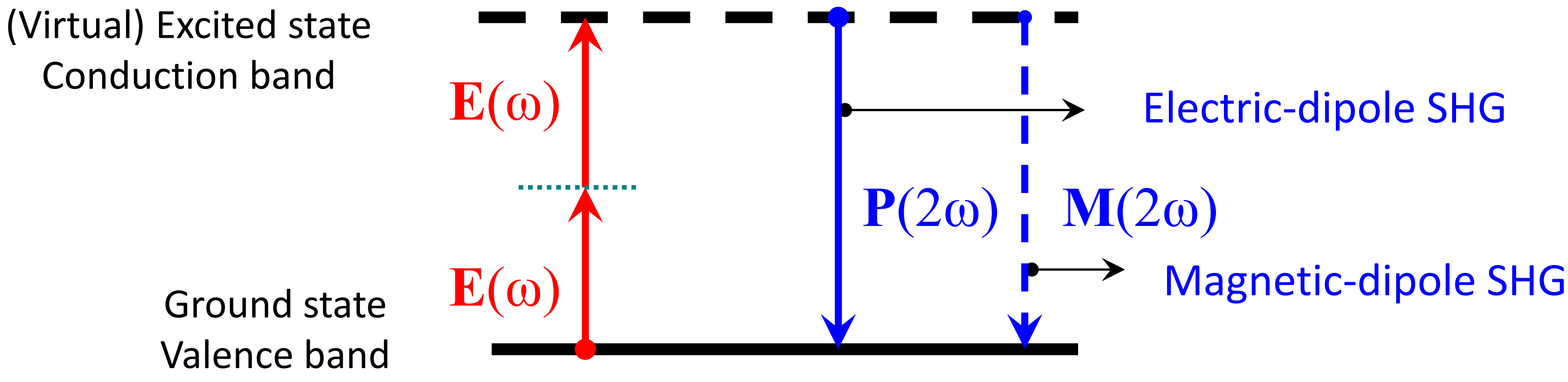


+magnetic SHG



SHG in centrosymmetric media

$$\mathbf{P}(2\omega) = \hat{\chi}^{i,3}\mathbf{E}(\omega)\mathbf{E}(\omega) + \hat{\chi}^{c,3}(\mathbf{M})\mathbf{E}(\omega)\mathbf{E}(\omega)$$

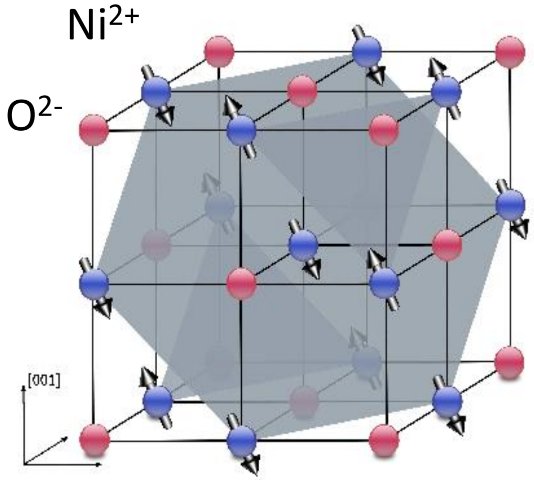


$$\mathbf{M}(2\omega) = \hat{\chi}^{c,3}\mathbf{E}(\omega)\mathbf{E}(\omega)$$

i-tensor ED MD c-tensor

$$\begin{pmatrix} \mathbf{P} \\ \mathbf{M} \\ \mathbf{Q} \end{pmatrix}^{(2\omega)} \propto \begin{pmatrix} \hat{\chi}^{eee} & \hat{\chi}^{eem} & \hat{\chi}^{emm} \\ \hat{\chi}^{mee} & \hat{\chi}^{mem} & \hat{\chi}^{mmm} \\ \hat{\chi}^{qee} & \hat{\chi}^{qem} & \hat{\chi}^{qmm} \end{pmatrix} \begin{pmatrix} \mathbf{EE} \\ \mathbf{EH} \\ \mathbf{HH} \end{pmatrix}^{(\omega)}$$

SHG in centrosymmetric antiferromagnet NiO



Cubic crystal (NaCl sturcture)

Point group: *m*3*m*

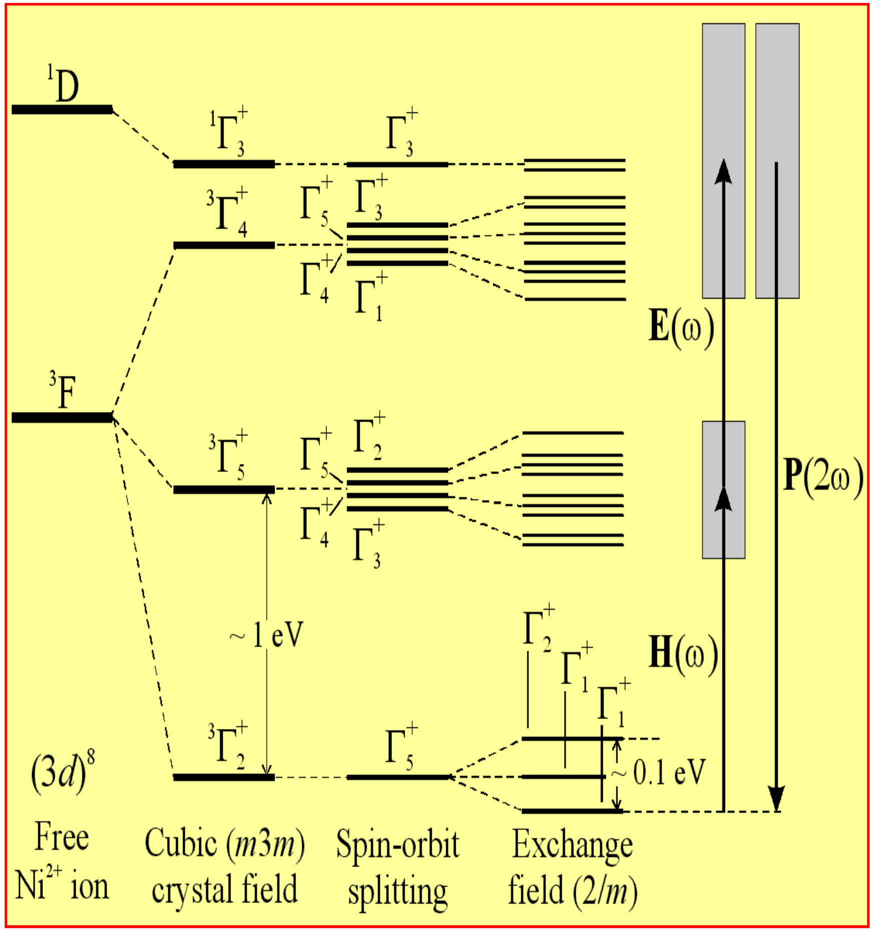
Polar tensor (ED-SHG):

$$\hat{\chi}^{i,3} = 0$$

Axial tensor (MD-SHG):

$$\hat{\chi}^{c,3}: \begin{matrix} xyz = zxy = yzx = a \\ xzy = yxz = zyx = -a \end{matrix}$$

3*d*-transitions of Ni $^{2+}$ ion in the crystal field

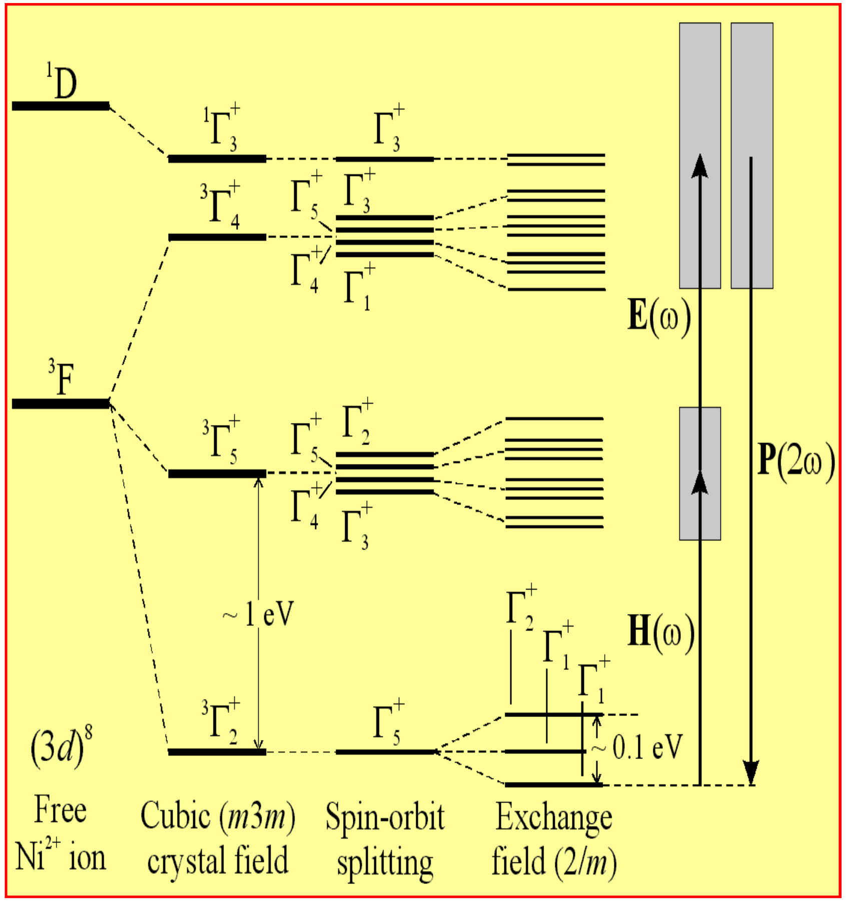


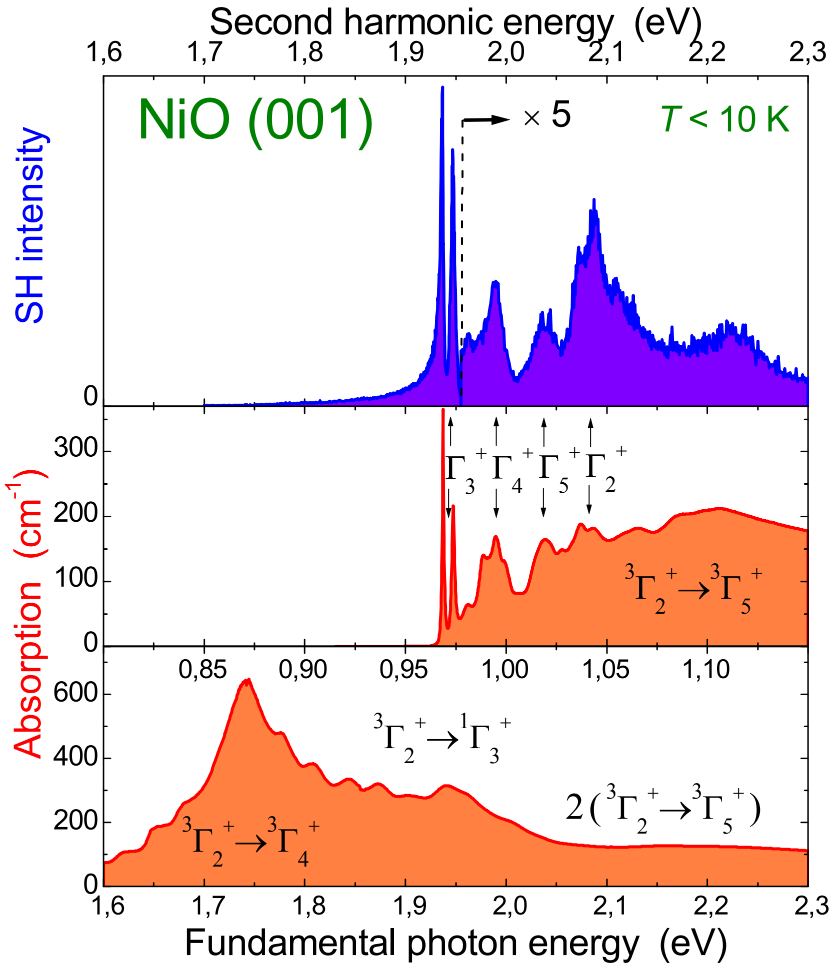
$^3\Gamma_2 \rightarrow {}^3\Gamma_5$

is the magnetic-dipole transition

SHG in centrosymmetric antiferromagnet NiO

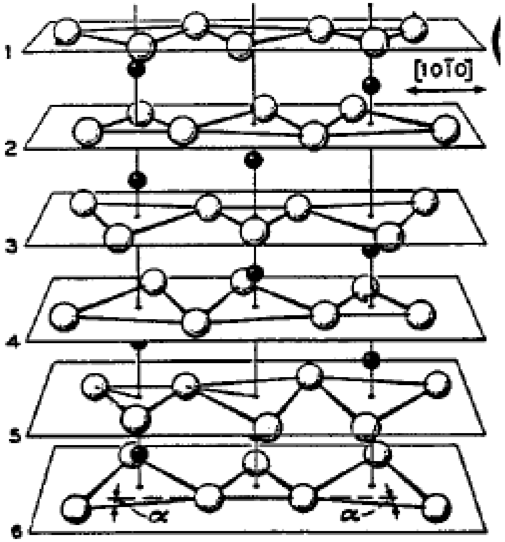
3d-transitions of Ni $^{2+}$ ion in the crystal field

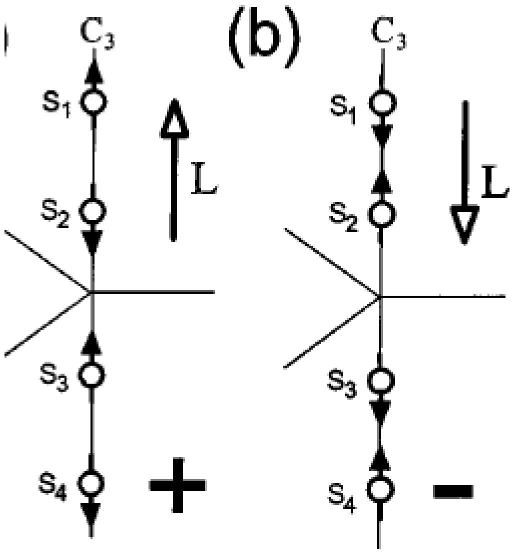




[PRL **87**, 137202 (1994)]

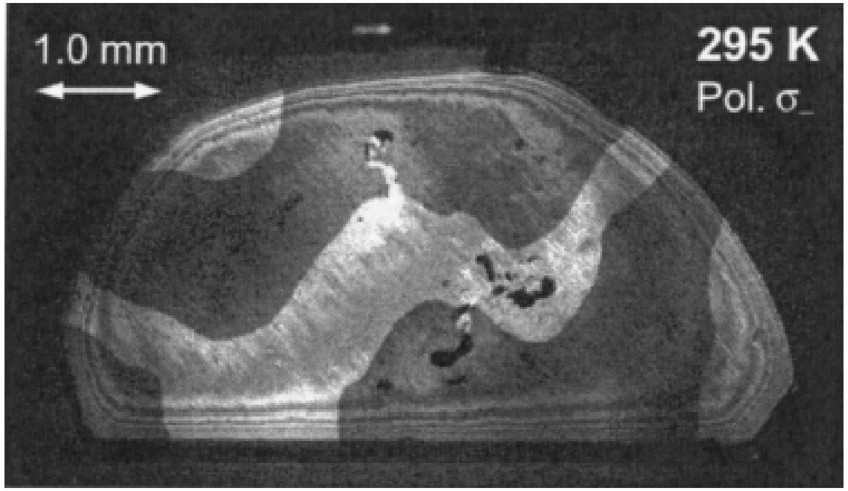
SHG in an antiferromagnet Cr_2O_3





Magnetic point group is $\bar{3}m$

SHG image



180° antiferromagnetic domains!

Breaking symmetry due
to antiferromagnetic order
Magnetic ED-SHG
+
Crystallographic MD-SHG

*[PRL **73**, 2127(1994)]*