Optical Second Harmonic Generation

Jean-Yves Chauleau
SPEC CEA Saclay

3 review articles and 1 book:

J. Opt. Soc. Am. B/Vol. 22, No. 1/January 2005

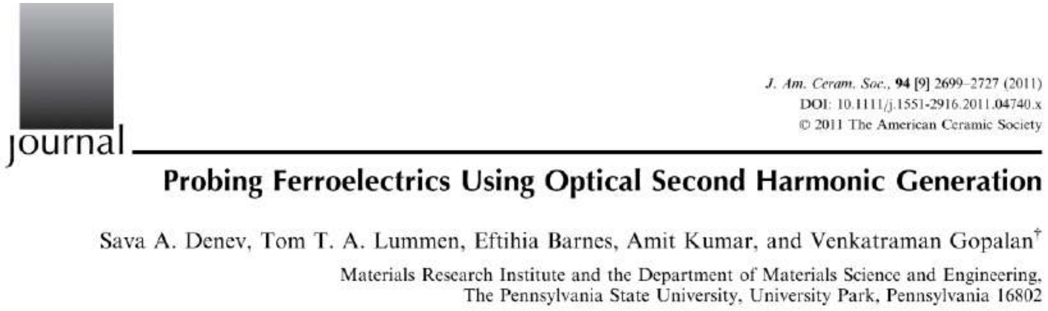
Second-harmonic generation as a tool for studying electronic and magnetic structures of crystals: review

Manfred Fiebig

Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin, Germany

Victor V. Pavlov and Roman V. Pisarev

A. F. Ioffe Physical Technical Institute, 26 Politekhnicheskaya Street, 194021 St. Petersburg, Russia



journal

J. Am. Ceram. Soc., **94** [9] 2699–2727 (2011)
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Probing Ferroelectrics Using Optical Second Harmonic Generation

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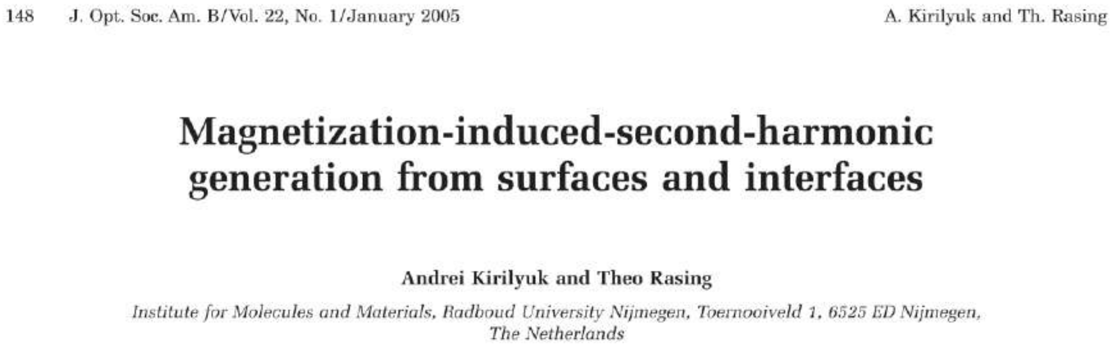
Materials Research Institute and the Department of Materials Science and Engineering,
The Pennsylvania State University, University Park, Pennsylvania 16802



148 J. Opt. Soc. Am. B/Vol. 22, No. 1/January 2005 A. Kirilyuk and Th. Rasing

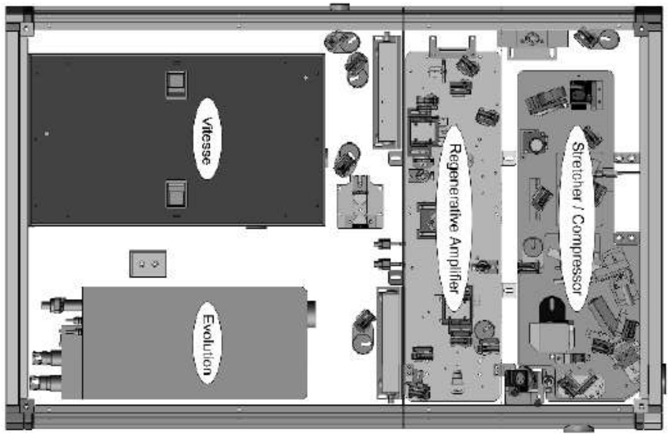
Magnetization-induced-second-harmonic generation from surfaces and interfaces

Andrei Kirilyuk and Theo Rasing

Institute for Molecules and Materials, Radboud University Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands

R. R. Birss, Symmetry and Magnetism, 1964 Ed.

Second Harmonic Generation (SHG):



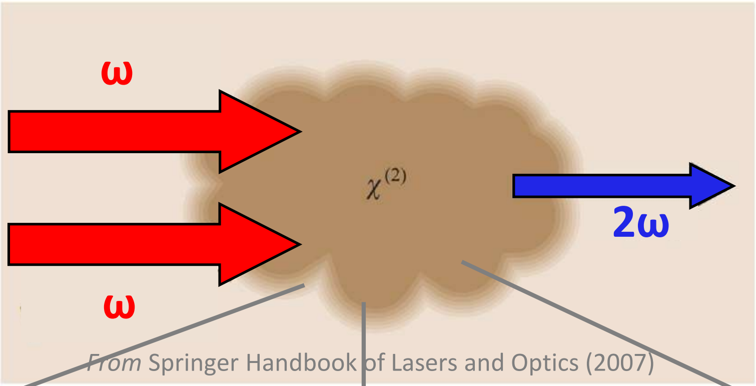
LIBRA from Coherent©

Light source at ω

Leading order SHG terms allowed only if:

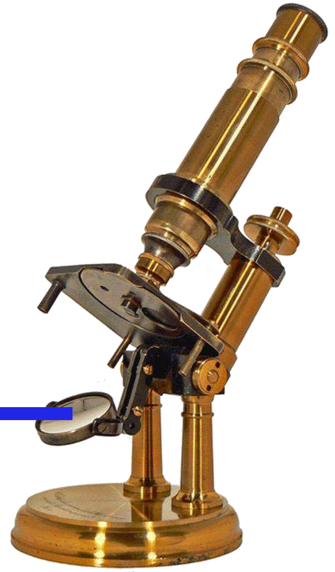
- Spatial centro-symmetry breaking (ferroelectricity)
- Time-inversion symmetry breaking ((anti)-ferromagnetism)

System presenting 2nd order non-linearities



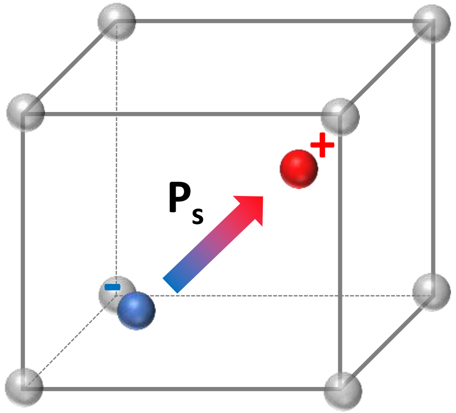
From Springer Handbook of Lasers and Optics (2007)

Analysis of the light at 2ω

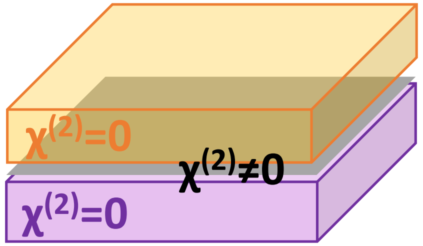


From www.lecompendium.com

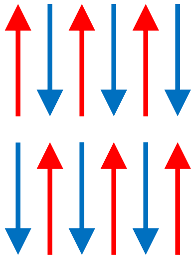
Ferroelectricity



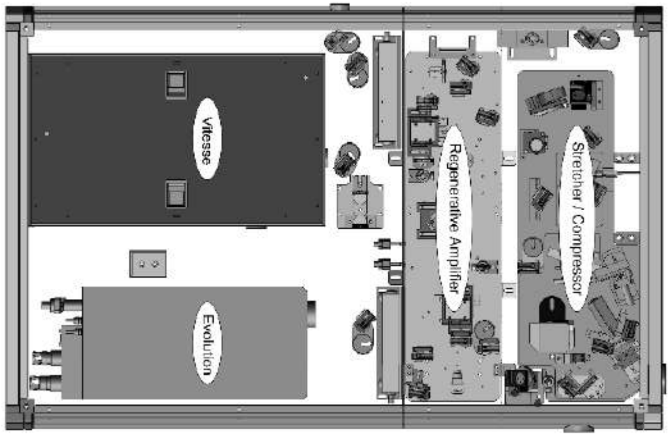
Interfaces



Magnetism



Second Harmonic Generation (SHG):



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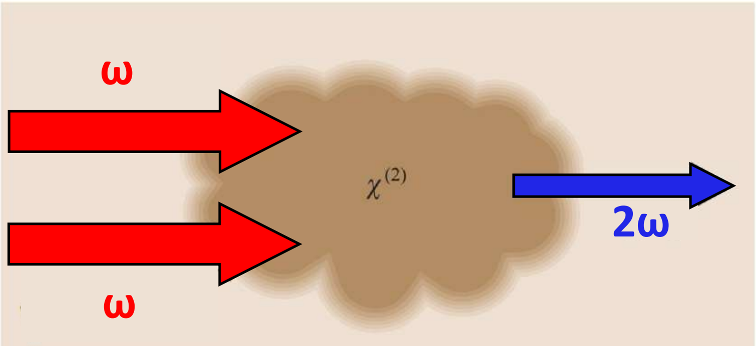

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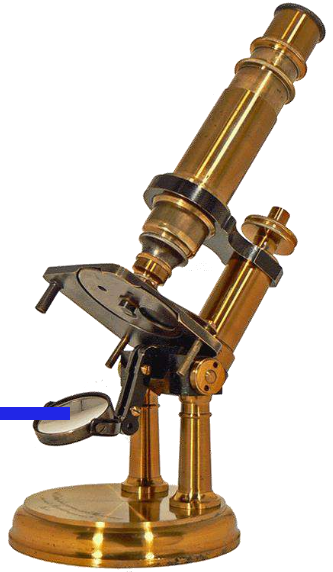


From www.lecompendium.com

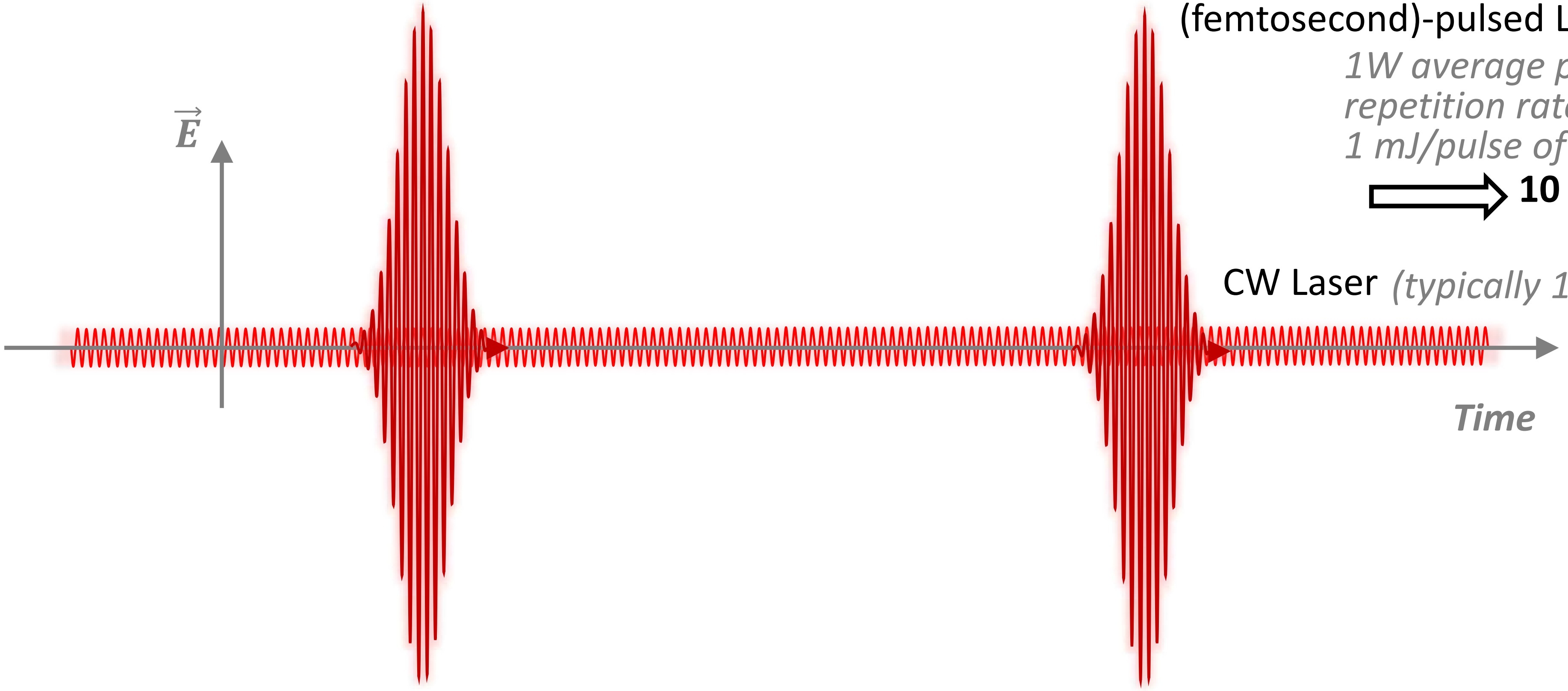
Outline:

1) ***Intense electric field laser pulses***: example of Kerr-lens mode locking
2) ***Basic formalism***: how can we tackle an SHG experiment/study
3) ***SHG illustrated by some studies:***
   a) Ferroelectrics
   b) Multiferroics
   c) Antiferromagnets
   d) Time-resolved and ultrafast abilities

Second Harmonic Generation (SHG): Non-linear optical process

Non-linear optics : *"S*tudy of the interaction of light with matter when the response of the system (or material) is nonlinear with the amplitude of the electric field of light "

⟹ Booming field of research since the apparition of intense pulsed laser sources



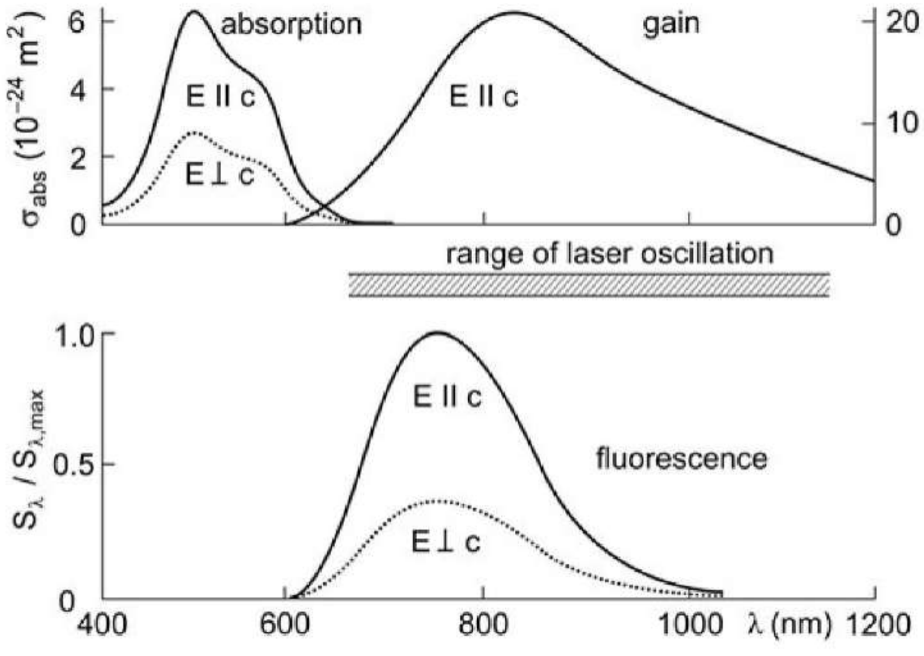
(femtosecond)-pulsed Laser

1W average power
repetition rate: 1kHz
1 mJ/pulse of 100 fs

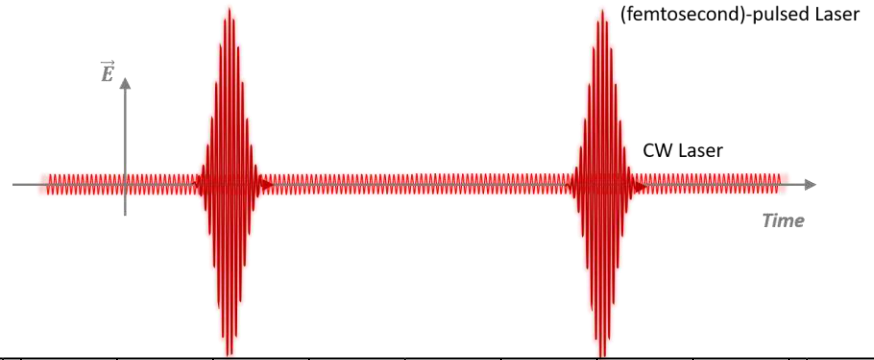
⟹ **10 GW peak power**

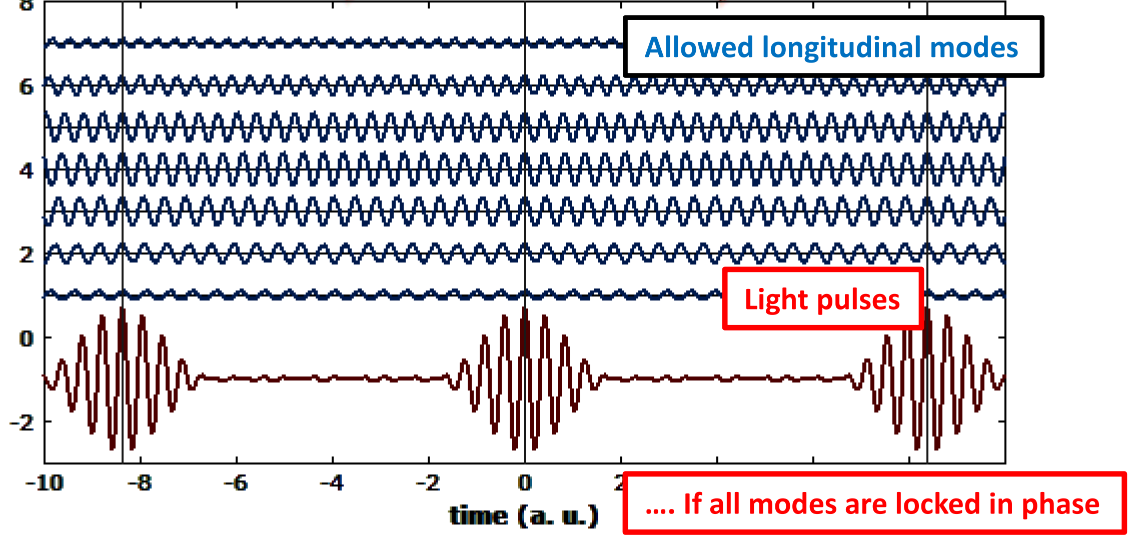
CW Laser *(typically 1W)*

Femtosecond Laser pulses: Kerr lens Mode-locking



For Ti:Al$_2$O$_3$ Lasers



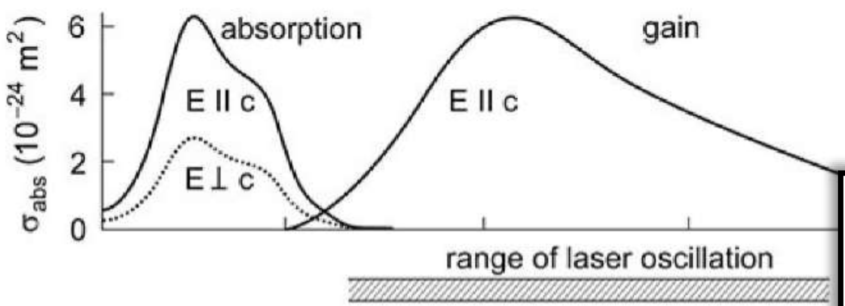


Allowed longitudinal modes

Light pulses

.... If all modes are locked in phase

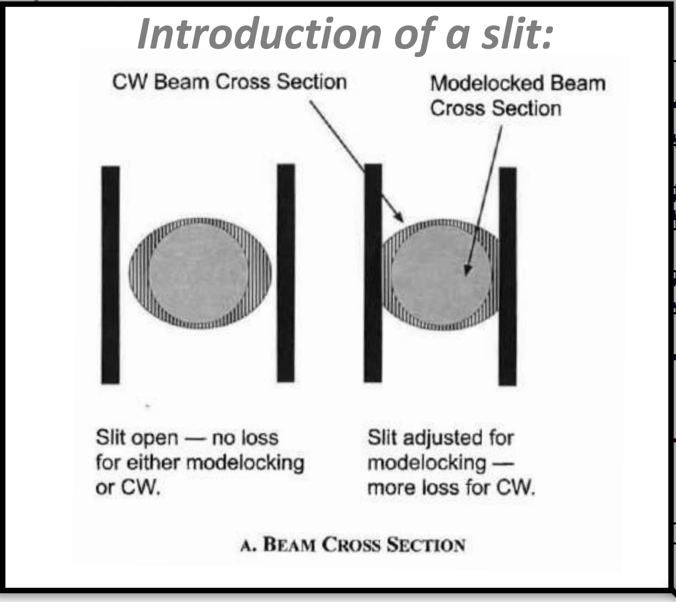
Femtosecond Laser pulses: Kerr lens Mode-locking



Non-linear Kerr effect:

$$n = n_0 + n_2 \cdot I$$

Introduction of a slit:



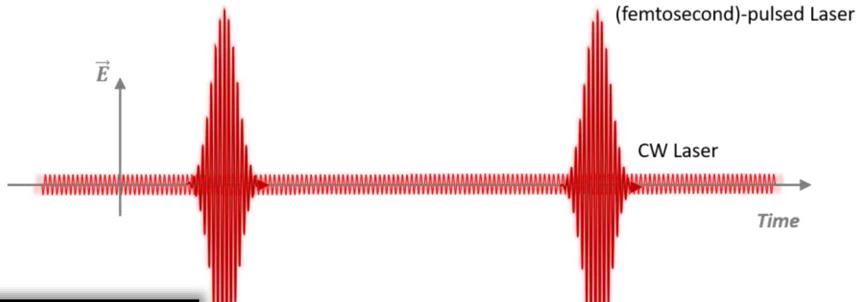
CW Beam Cross Section

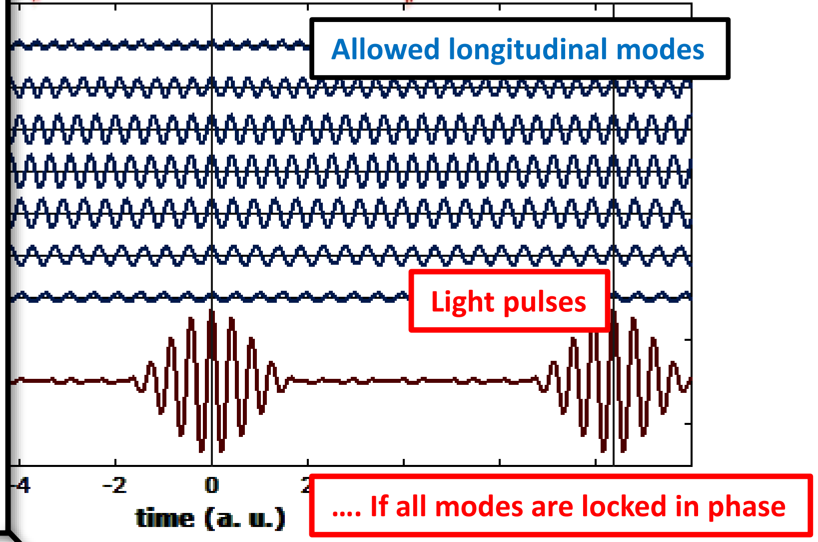
Modelocked Beam Cross Section

Slit open — no loss for either modelocking or CW.

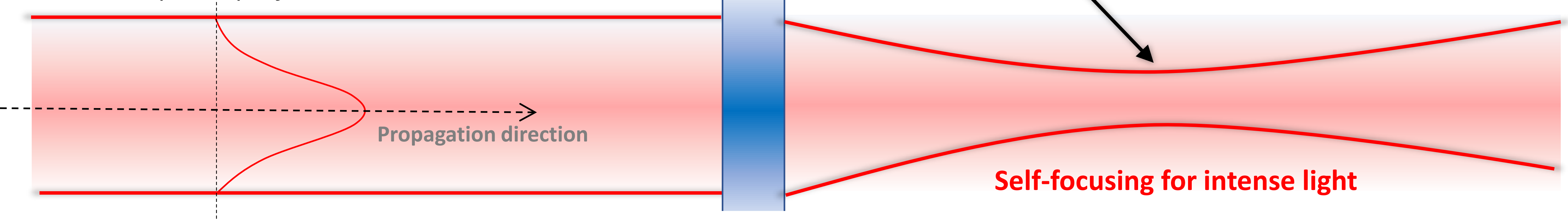
Slit adjusted for modelocking — more loss for CW.

A. BEAM CROSS SECTION



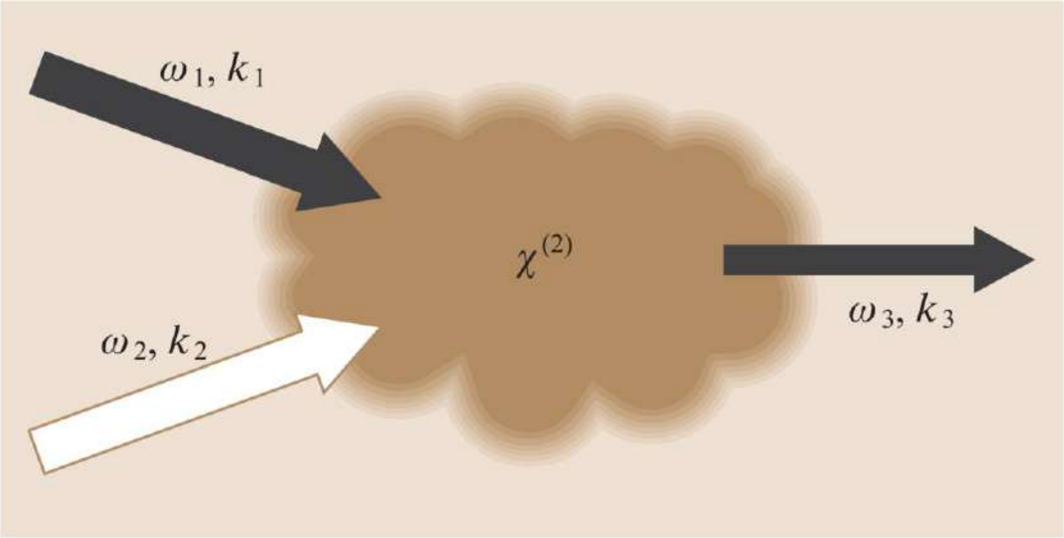


Gaussian spatial profil:

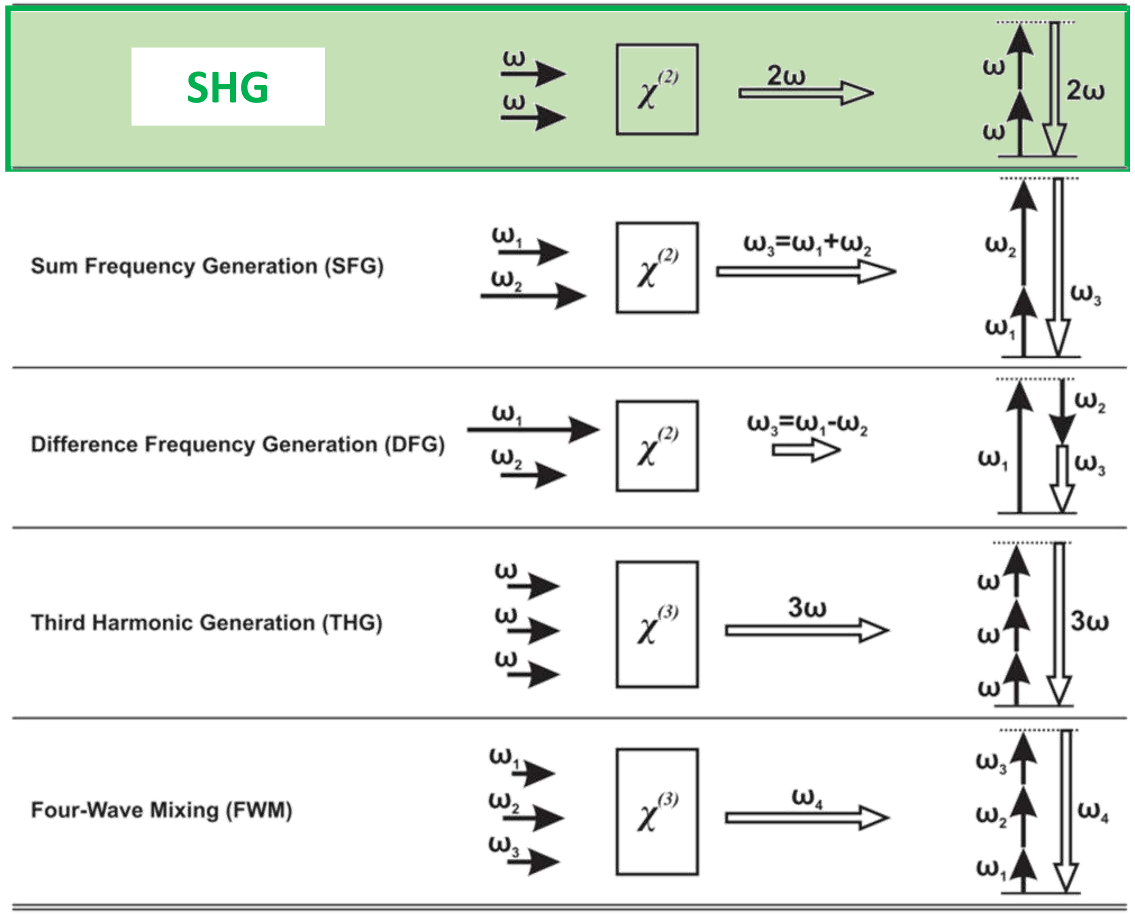


Gradient of refractive index (= lens)

Second Harmonic Generation (SHG): Non-linear optical process

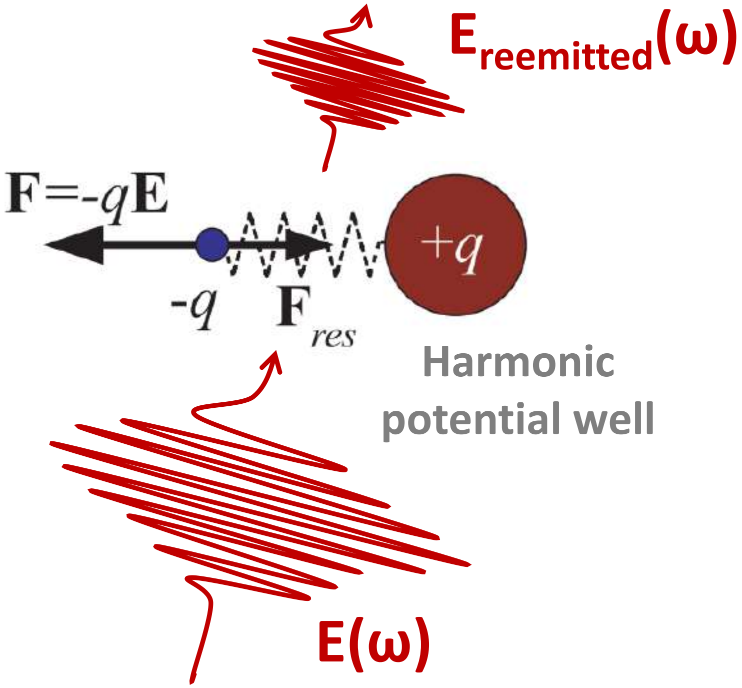


From Springer Handbook of Lasers and Optics (2007)



SHG from classical NL electron spring model :

Described in : Denev et al. *J. Am. Ceram. Soc.* **94** 2699 (2011)



Oscillating dipole

$$\mathbf{P}(t) = N_{\mathrm{e}} e\mathbf{x}(t) = \varepsilon_0 \chi_{\mathrm{e}} \mathbf{E}(t).$$

Reemit an electromagnetic field at ***ω***

$$\nabla^2 \mathbf{E} - \mu\varepsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu \frac{\partial^2 \mathbf{P}}{\partial t^2}.$$

SHG from classical NL electron spring model :

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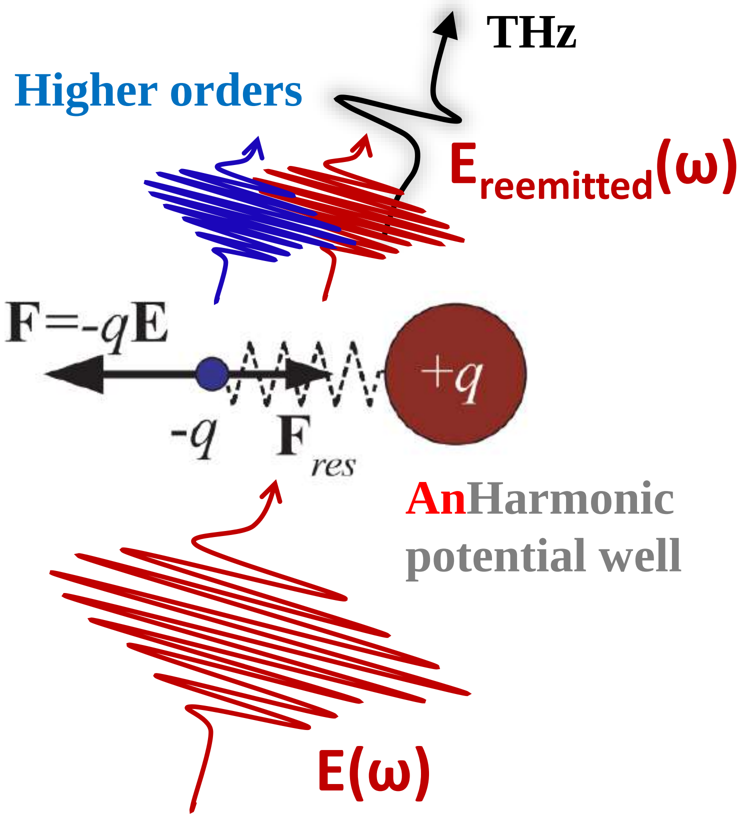


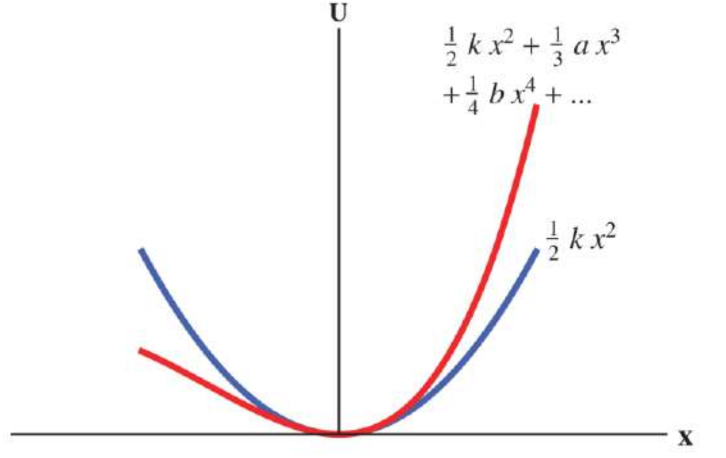


Fig. 3. Harmonic (blue) and anharmonic (red) potential wells.

Oscillating dipole

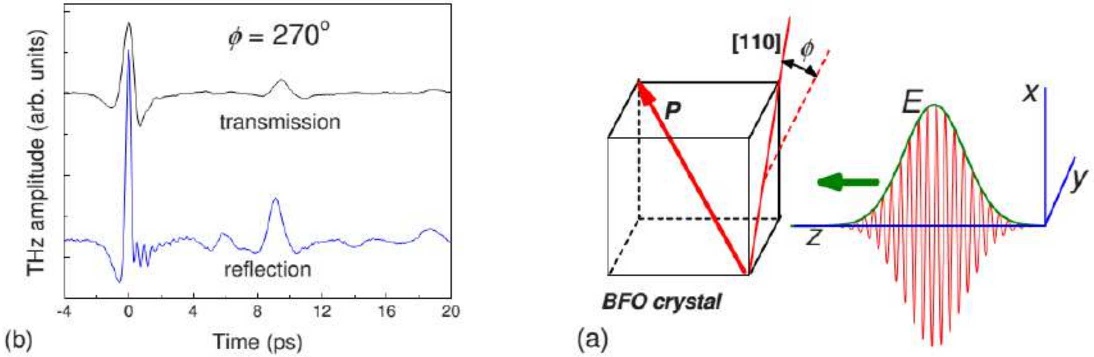
$$\mathbf{P}(t) = \mathbf{P}_0 + \varepsilon_0\chi_e\mathbf{E}(t) + \chi^{(2)}\mathbf{E}(t)^2 + \chi^{(3)}\mathbf{E}(t)^3 + \cdots$$

Sources of non-linear optical phenomena

Optical rectification

Linear susceptibility

2nd order non-linear susceptibility



N.B.: $\cos\omega t \times \cos\omega t \rightarrow 1 + \cos 2\omega t$

SHG more general tensorial approach:

Additional source terms…

$$\nabla^2\mathbf{E} - \mu\varepsilon_0 \frac{\partial^2\mathbf{E}}{\partial t^2} = \mu\frac{\partial^2\mathbf{P}}{\partial t^2} + \mu\left(\nabla\times\frac{\partial\mathbf{M}}{\partial t}\right) - \mu\left(\nabla\frac{\partial^2\hat{Q}}{\partial t^2}\right).$$

Electric dipole *Magnetic dipole* *Electric quadrupole*

$$\begin{aligned}
\mathbf{P} &\propto \chi^{ee}:\mathbf{E} + \chi^{em}:\mathbf{H} + \chi^{eee}:\mathbf{EE} + \chi^{eem}:\mathbf{EH} + \chi^{emm}:\mathbf{HH} + \mathcal{O}[(\mathbf{E},\ \mathbf{H})^3]\\
\mathbf{M} &\propto \chi^{me}:\mathbf{E} + \chi^{mm}:\mathbf{H} + \chi^{mee}:\mathbf{EE} + \chi^{mem}:\mathbf{EH} + \chi^{mmm}:\mathbf{HH} + \mathcal{O}[(\mathbf{E},\ \mathbf{H})^3],\\
\hat{Q} &\propto \chi^{qe}:\mathbf{E} + \chi^{qm}:\mathbf{H} + \chi^{qee}:\mathbf{EE} + \chi^{qem}:\mathbf{EH} + \chi^{qmm}:\mathbf{HH} + \mathcal{O}[(\mathbf{E},\ \mathbf{H})^3].
\end{aligned}$$

Linear optics

Non-Linear optics

$\mathbf{E}$: electric field of light
$\mathbf{H}$: magnetic field of light

SHG more general tensorial approach:

Additional source terms…

$$\nabla^2\mathbf{E} - \mu\varepsilon_0 \frac{\partial^2 \mathbf{E}}{\partial t^2} = \mu\frac{\partial^2\mathbf{P}}{\partial t^2} + \mu\left(\nabla\times\frac{\partial\mathbf{M}}{\partial t}\right) - \mu\left(\nabla\frac{\partial^2\hat{Q}}{\partial t^2}\right).$$

Electric dipole *Magnetic dipole* *Electric quadrupole*

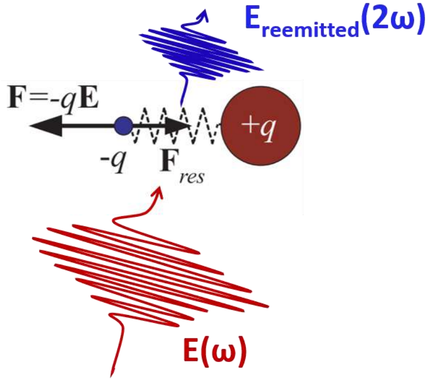
Tensorial expression of the possible SHG terms :

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

***9** possible 3rd rank tensors (**27** components each)*

N.B.: $I^{2\omega}_{SHG} \propto \left|\vec{P}^{2\omega}\right|^2$ in the E.D. approximation

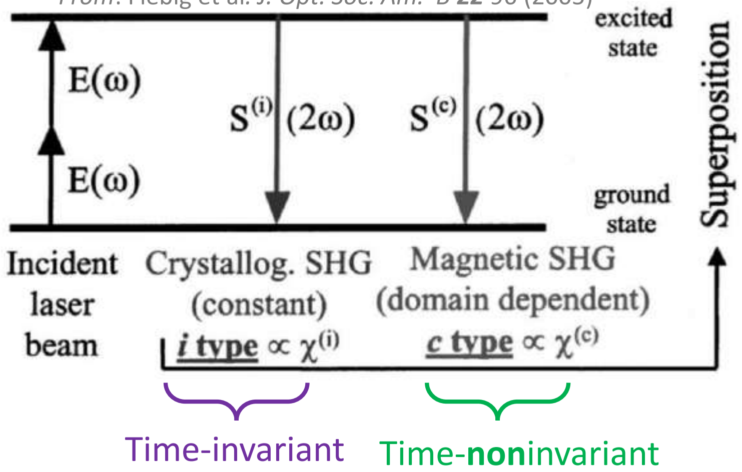
Leading term



$\chi^{eee} = 0$

in centrosymmetric systems

From: Fiebig et al. *J. Opt. Soc. Am. B* ***22*** 96 (2005)



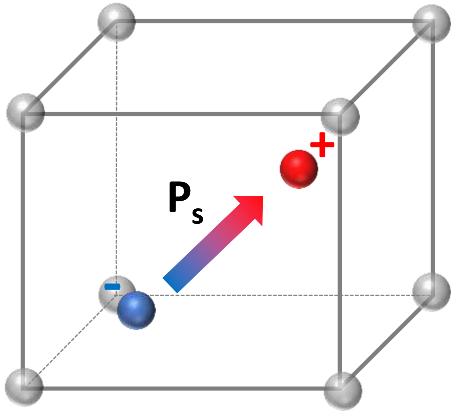
Some symmetry considerations:

Neumann's principle : "any type of symmetry which is exhibited by the point group of the crystal is possessed by every physical property of the crystal"

All components of **axial** tensors of **even rank** and **polar** tensors of **odd rank** must vanish identically for centrosymmetrical classes (symmetry operator $\bar{1}$)

SHG in ferroelectric materials:

Protypical bulk systems presenting spatial centrosymmetry breaking



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

$$\vec{P}(2\omega) \propto \chi^{eee(i)} : \vec{E}(\omega) \otimes \vec{E}(\omega))$$

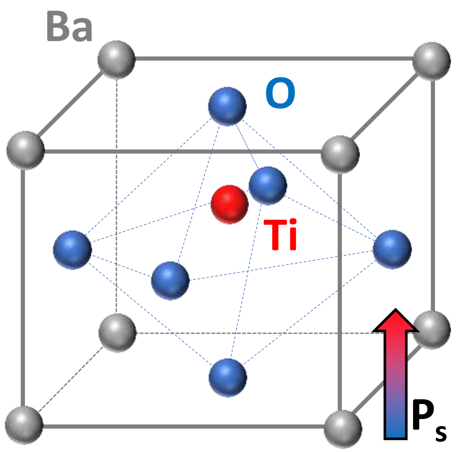
$$\begin{bmatrix} P_1\\ P_2\\ P_3\end{bmatrix} \propto \begin{bmatrix} \chi_{111}{}^{(i)} & \chi_{122}{}^{(i)} & \chi_{133}{}^{(i)} & \chi_{123}{}^{(i)} & \chi_{113}{}^{(i)} & \chi_{112}{}^{(i)}\\ \chi_{211}{}^{(i)} & \chi_{222}{}^{(i)} & \chi_{233}{}^{(i)} & \chi_{223}{}^{(i)} & \chi_{213}{}^{(i)} & \chi_{212}{}^{(i)}\\ \chi_{311}{}^{(i)} & \chi_{322}{}^{(i)} & \chi_{333}{}^{(i)} & \chi_{323}{}^{(i)} & \chi_{313}{}^{(i)} & \chi_{312}{}^{(i)}\end{bmatrix}\begin{bmatrix} E_1^2\\ E_2^2\\ E_3^2\\ 2E_2E_3\\ 2E_1E_3\\ 2E_1E_2\end{bmatrix}$$

(1,2,3) : crystal frame

If no symmetry : 27 different components

SHG in ferroelectric materials:

Protypical bulk systems presenting spatial centrosymmetry breaking



Example : *tetragonal barium titanate* **$BaTiO_3$**

Non-centrosysmetric Point Group **4mm**

Symmetry operations:

$\cancel{\bar{1}}$ but $1, 2_z, \bar{2}_x, \bar{2}_y, \bar{2}_{xy}, \bar{2}_{-xy}, \pm 4_z$

$$\begin{bmatrix} P_1 \\ P_2 \\ P_3 \end{bmatrix} \propto \begin{bmatrix} \chi_{111}^{(i)} & \chi_{122}^{(i)} & \chi_{133}^{(i)} & \chi_{123}^{(i)} & \chi_{113}^{(i)} & \chi_{112}^{(i)} \\ \chi_{211}^{(i)} & \chi_{222}^{(i)} & \chi_{233}^{(i)} & \chi_{223}^{(i)} & \chi_{213}^{(i)} & \chi_{212}^{(i)} \\ \chi_{311}^{(i)} & \chi_{322}^{(i)} & \chi_{333}^{(i)} & \chi_{323}^{(i)} & \chi_{313}^{(i)} & \chi_{312}^{(i)} \end{bmatrix} \begin{bmatrix} E_1^2 \\ E_2^2 \\ E_3^2 \\ 2E_2E_3 \\ 2E_1E_3 \\ 2E_1E_2 \end{bmatrix}$$

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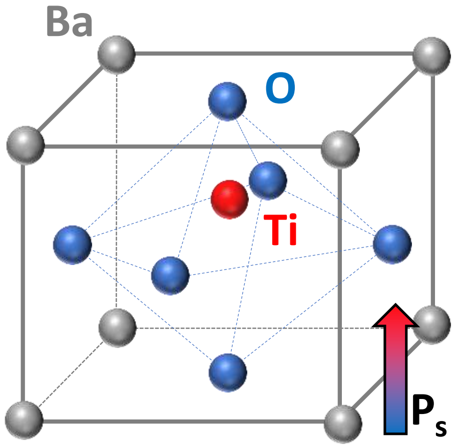

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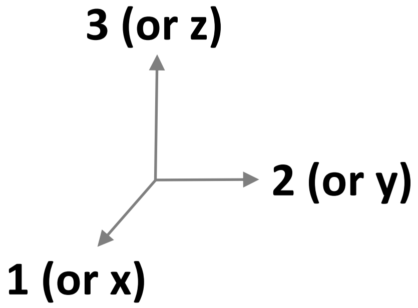
$$\begin{bmatrix}P_1\\P_2\\P_3\end{bmatrix} \propto \begin{bmatrix} 0&0&0&0&\chi_{113}{}^{(i)}&0\\ 0&0&0&\chi_{113}{}^{(i)}&0&0\\ \chi_{311}{}^{(i)}&\chi_{311}{}^{(i)}&\chi_{333}{}^{(i)}&0&0&0\end{bmatrix}\begin{bmatrix}E_1^2\\E_2^2\\E_3^2\\2E_2E_3\\2E_1E_3\\2E_1E_2\end{bmatrix}$$

(1,2,3) : crystal frame

3 independent components



SHG in ferroelectric materials:

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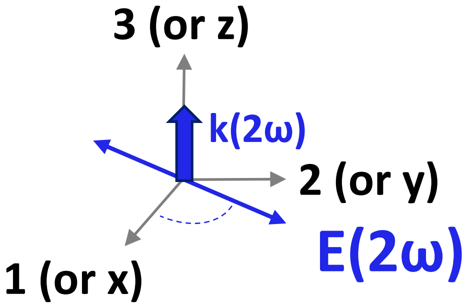
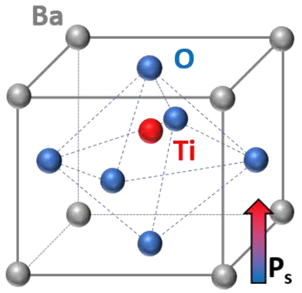
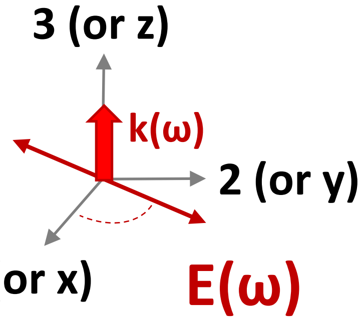


$$\begin{bmatrix} P_1 \\ P_2 \\ P_3 \end{bmatrix} \propto \begin{bmatrix} 0 & 0 & 0 & 0 & {\chi_{113}}^{(i)} & 0 \\ 0 & 0 & 0 & {\chi_{113}}^{(i)} & 0 & 0 \\ {\chi_{311}}^{(i)} & {\chi_{311}}^{(i)} & {\chi_{333}}^{(i)} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_1^2 \\ E_2^2 \\ E_3^2 \\ 2E_2E_3 \\ 2E_1E_3 \\ 2E_1E_2 \end{bmatrix}$$

(1,2,3) : crystal frame

If:



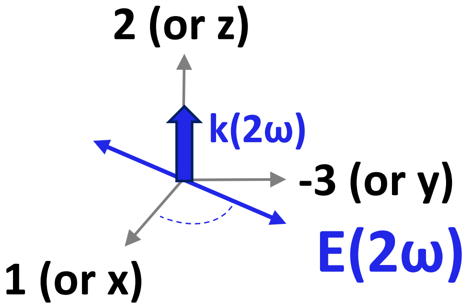


$$\mathbf{E(\omega)} = \begin{bmatrix} E_x = E_0 \cos\varphi \\ E_y = -E_0 \sin\varphi \\ 0 \end{bmatrix} \Rightarrow \mathbf{P(2\omega)} = \begin{bmatrix} 0 \\ 0 \\ {\chi_{311}}^{(i)} E_0^2 \end{bmatrix} \Rightarrow \mathbf{E(2\omega)} = \begin{bmatrix} 0 \\ 0 \\ E_z \end{bmatrix}$$

Experimentally not accessible: ***no SHG contrast is expected for this configuration…***

SHG in ferroelectric materials:

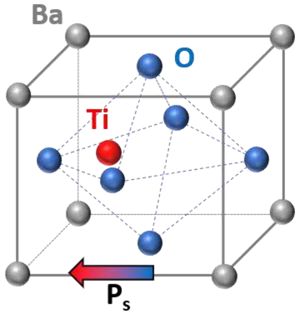
Protypical bulk systems presenting spatial centrosymmetry breaking



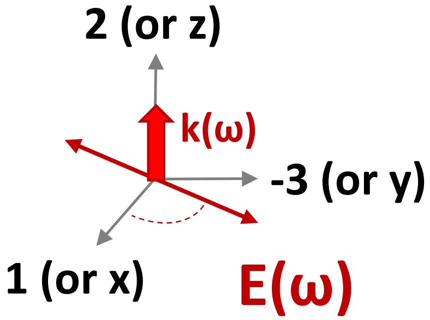
$$\begin{bmatrix} P_1 \\ P_2 \\ P_3 \end{bmatrix} \propto \begin{bmatrix} 0 & 0 & 0 & 0 & 0 & \chi_{113}{}^{(i)} \\ \chi_{311}{}^{(i)} & \chi_{333}{}^{(i)} & \chi_{311}{}^{(i)} & 0 & 0 & 0 \\ 0 & 0 & 0 & \chi_{113}{}^{(i)} & 0 & 0 \end{bmatrix} \begin{bmatrix} E_1^2 \\ E_2^2 \\ E_3^2 \\ 2E_2E_3 \\ 2E_1E_3 \\ 2E_1E_2 \end{bmatrix}$$

(1,2,3) : crystal frame

If:



$$\mathbf{E(\omega)} = \begin{bmatrix} E_1 = E_0 \cos\varphi \\ E_2 = -E_0 \sin\varphi \\ 0 \end{bmatrix} \Rightarrow \mathbf{P(2\omega)} = \begin{bmatrix} \chi_{333}{}^{(i)} E_0^2 \cos^2\varphi + \chi_{311}{}^{(i)} E_0^2 \sin^2\varphi \\ \chi_{113}{}^{(i)} E_0^2 \sin 2\varphi \\ 0 \end{bmatrix}$$



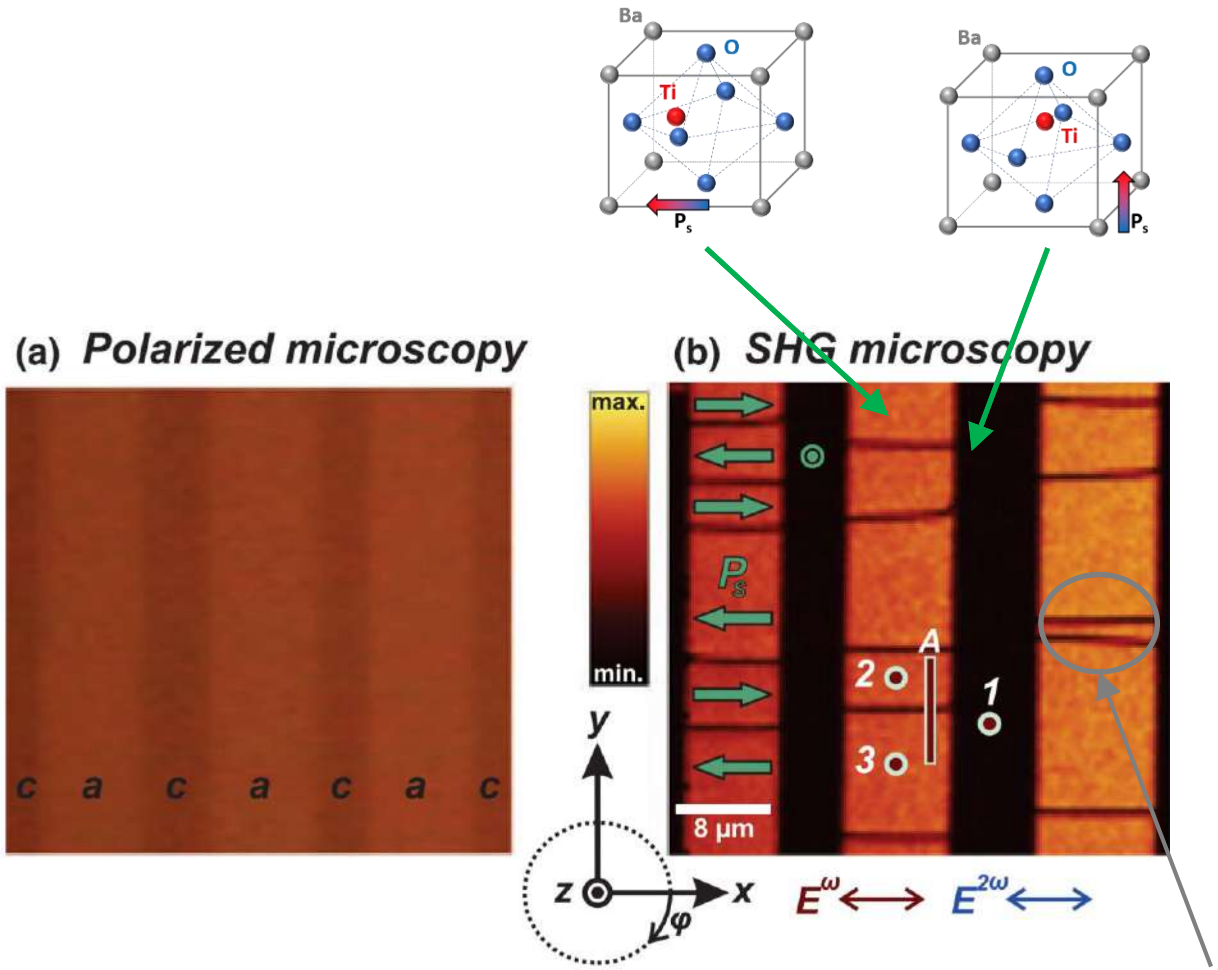
Experimentally accessible: *SHG contrast*

Tensor rotation:

$$\chi_{ijk}^{new} = \sum_{lmn} a_{il} a_{jm} a_{kn} \ \chi_{ijk}^{old}$$

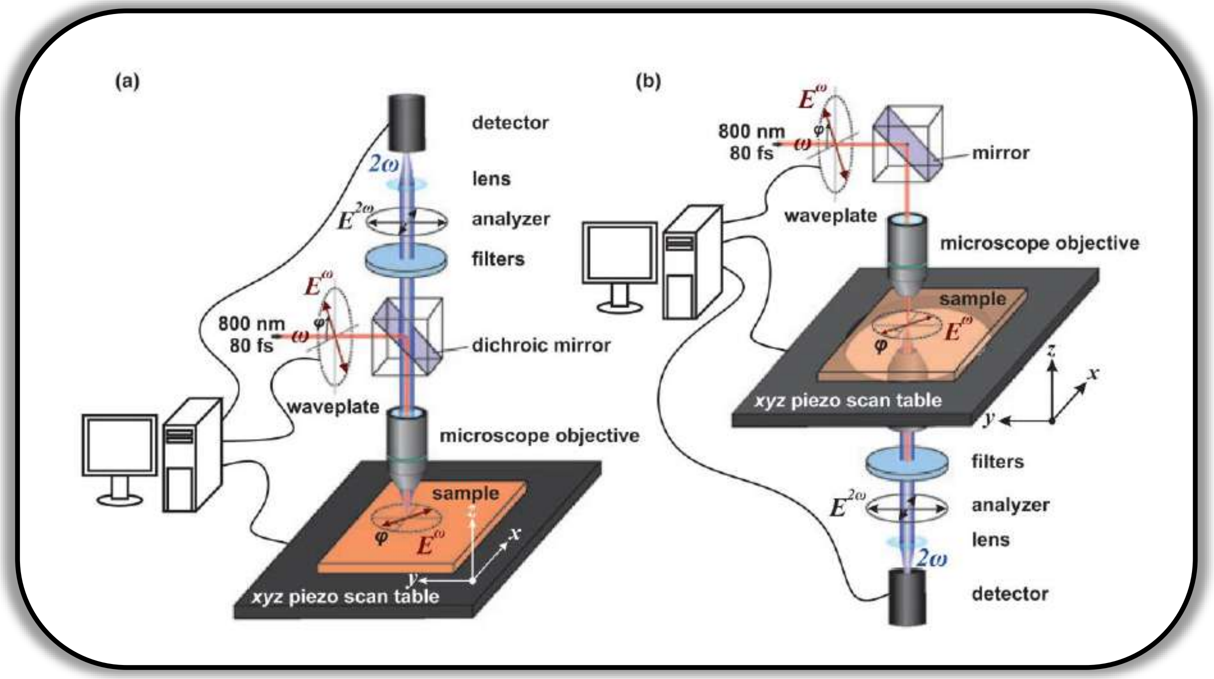
SHG in ferroelectric materials:

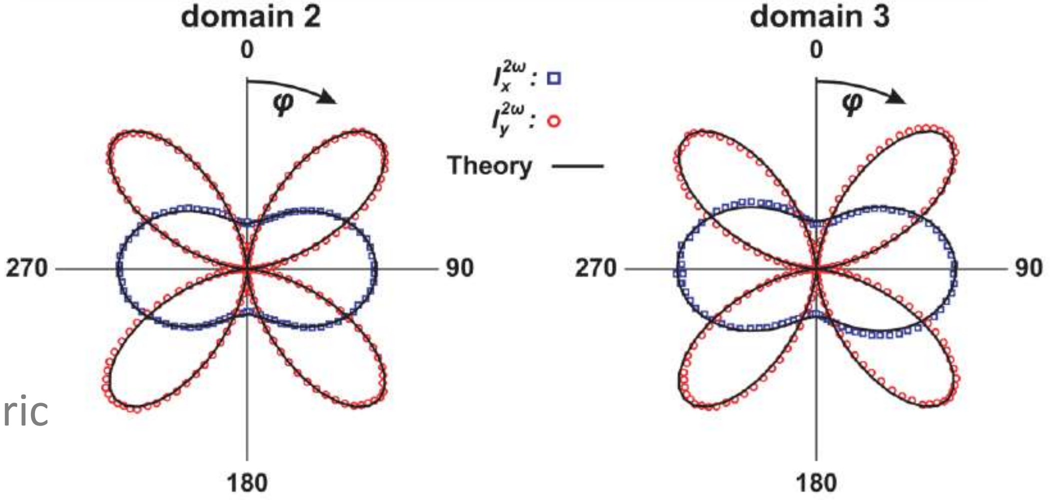
Protypical bulk systems presenting spatial centrosymmetry breaking



From Denev et al. *J. Am. Ceram. Soc.* 94 2699 (2011)

180° Ferroelectric domain walls





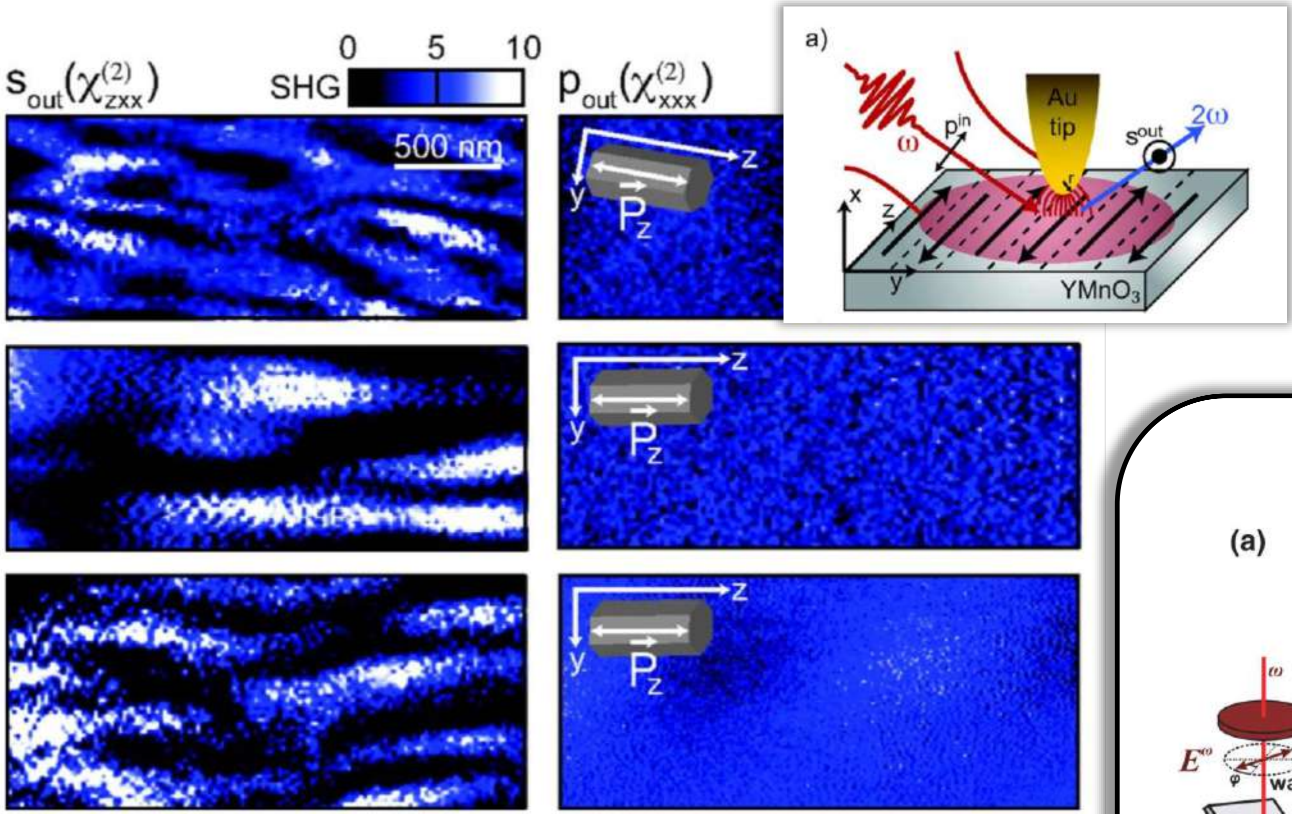
SHG in ferroelectric materials: Near-field imaging in $YMnO_3$

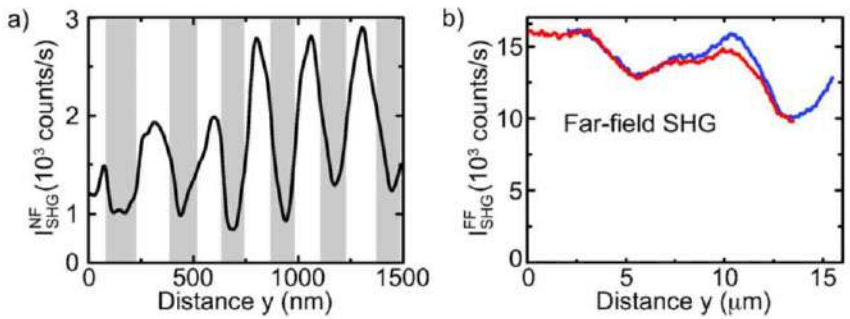
Neacsu et al. Phys. Rev. B. 79, 100107(R) (2009)

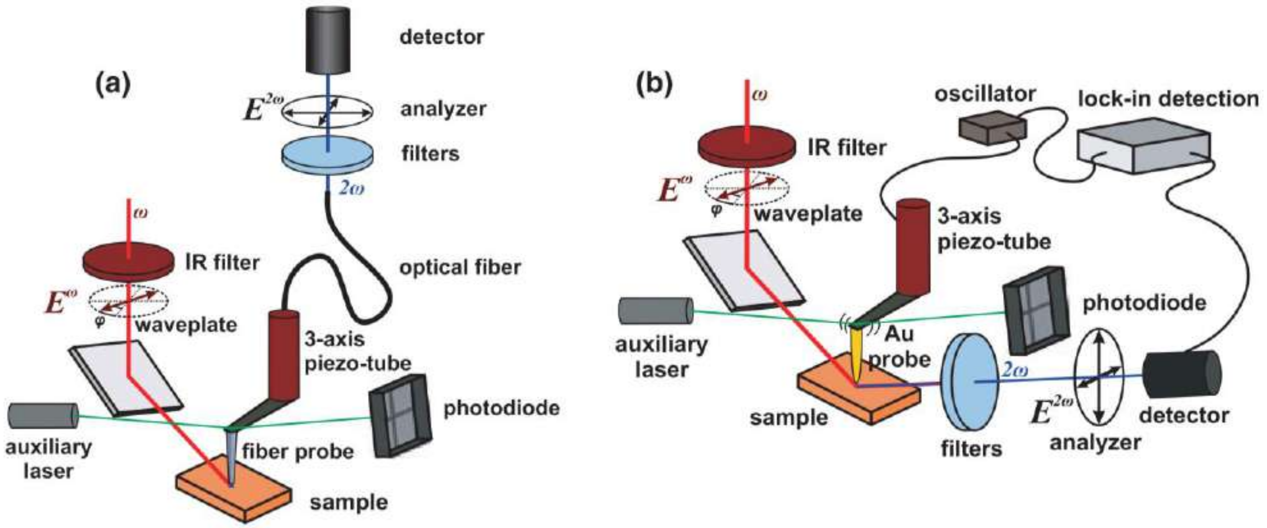
$$\chi^{(2)}_{6mm} = \begin{bmatrix} 0 & 0 & 0 & 0 & \chi_{113}{}^{(i)} & 0 \\ 0 & 0 & 0 & \chi_{113}{}^{(i)} & 0 & 0 \\ \chi_{311}{}^{(i)} & \chi_{311}{}^{(i)} & \chi_{333}{}^{(i)} & 0 & 0 & 0 \end{bmatrix}$$

$$I_{SHG}^{TOTAL} \propto \left| \pm \vec{P}_z^{Near-Field} + \vec{P}_z^{Far-Field} e^{i\Phi_{FF}} \right|^2$$

Fiebig et al. Phys. Rev. B. 66, 144102 (2002)





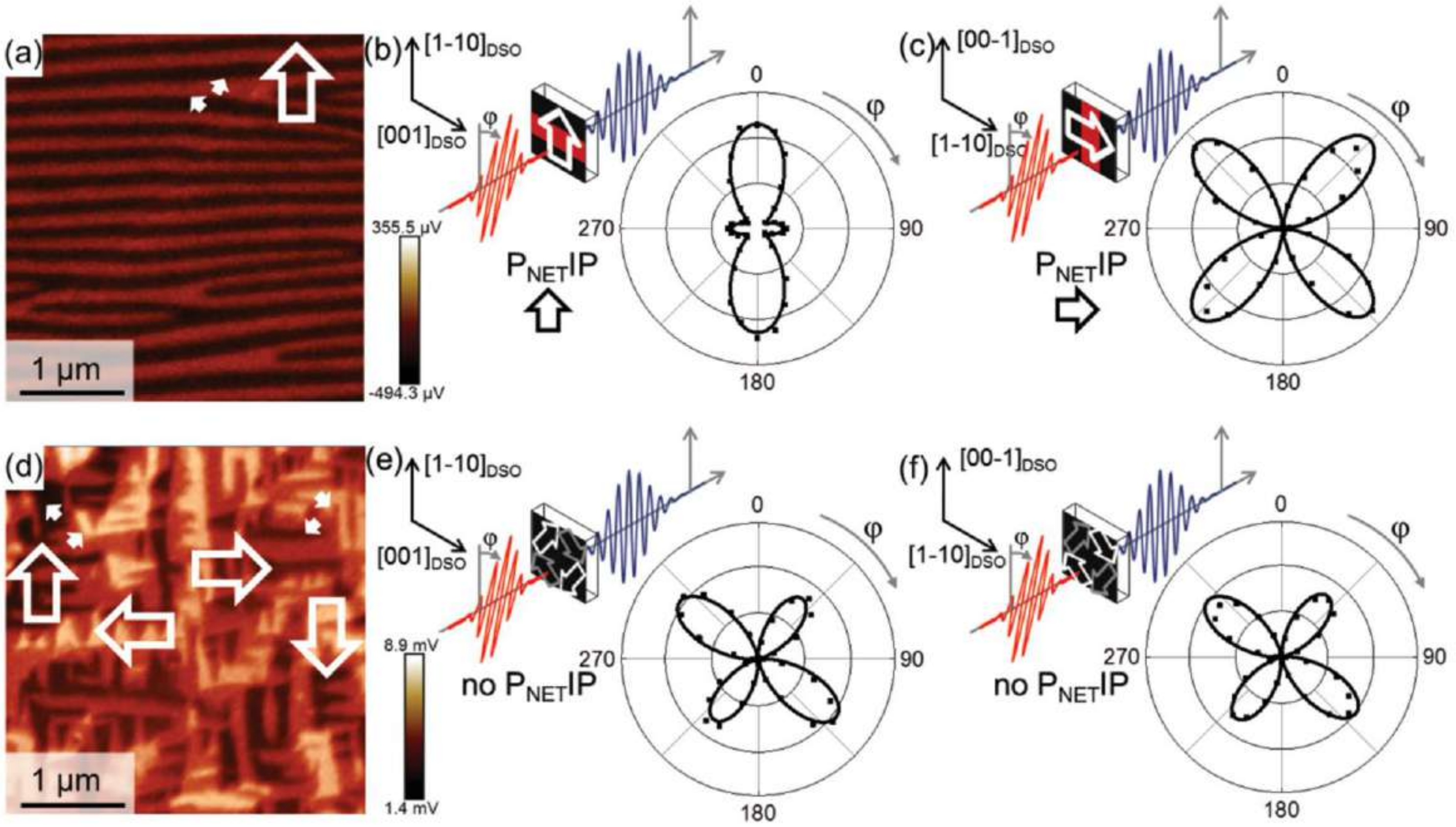


From Denev et al. *J. Am. Ceram. Soc.* 94 2699 (2011)

Visualization of 180° ferroelectric domain below the optical diffraction limit

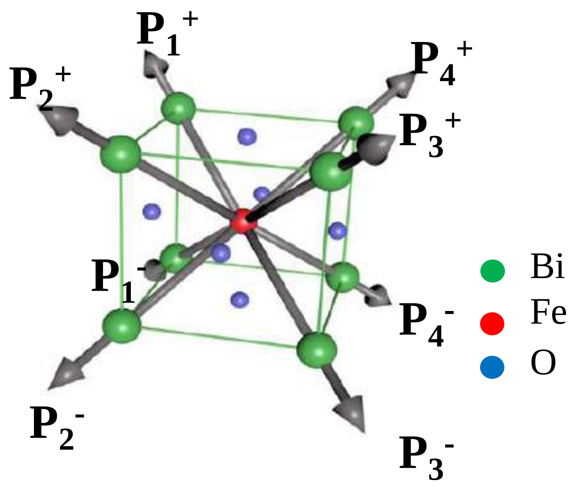
SHG in ferroelectric materials:

Trassin et al. Adv. Materials 27, 4871 (2015)



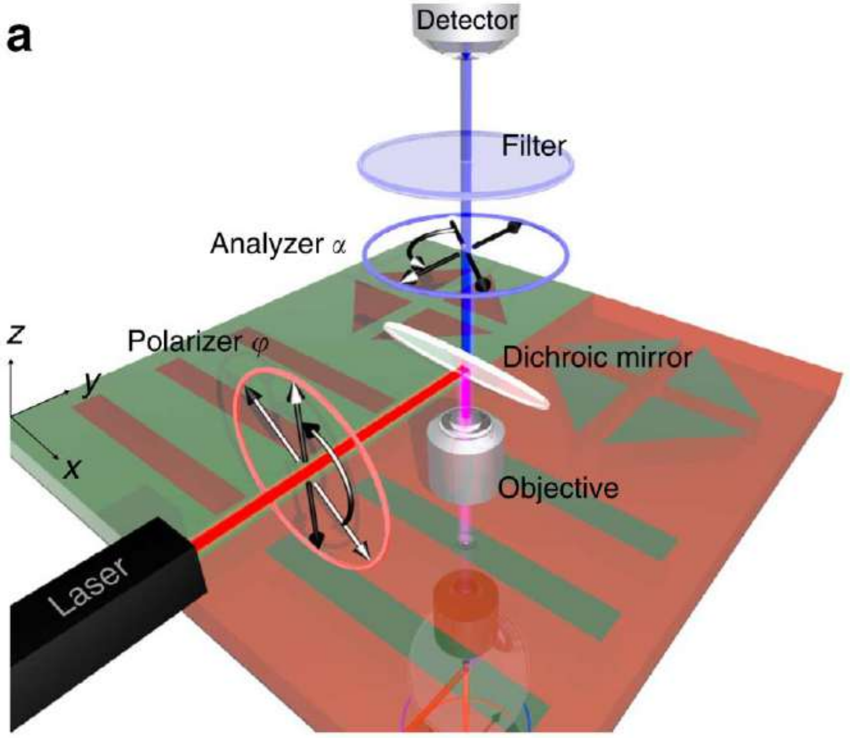
Ferroelectric configuration:

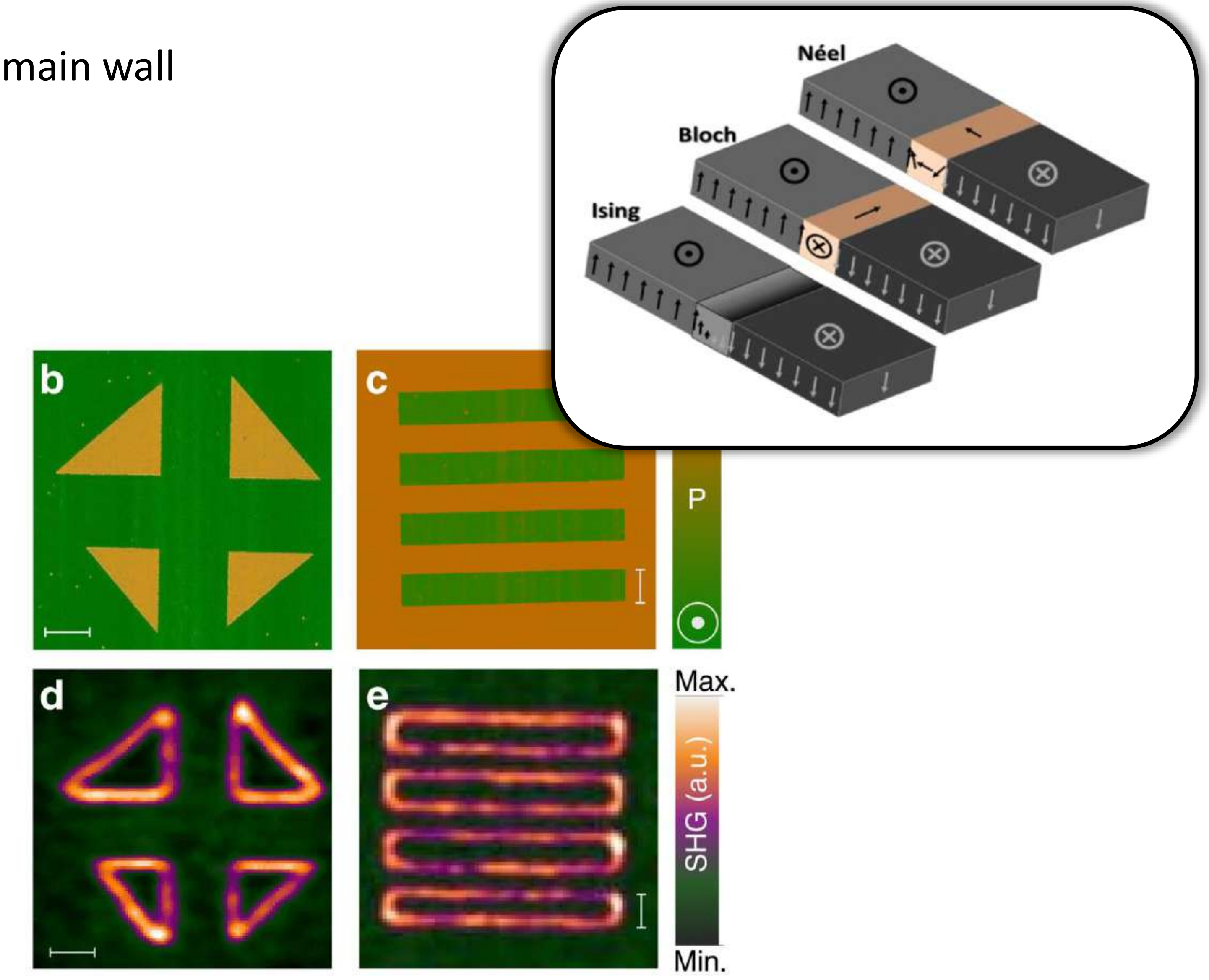
8 different ferroelectric polarization directions [111]



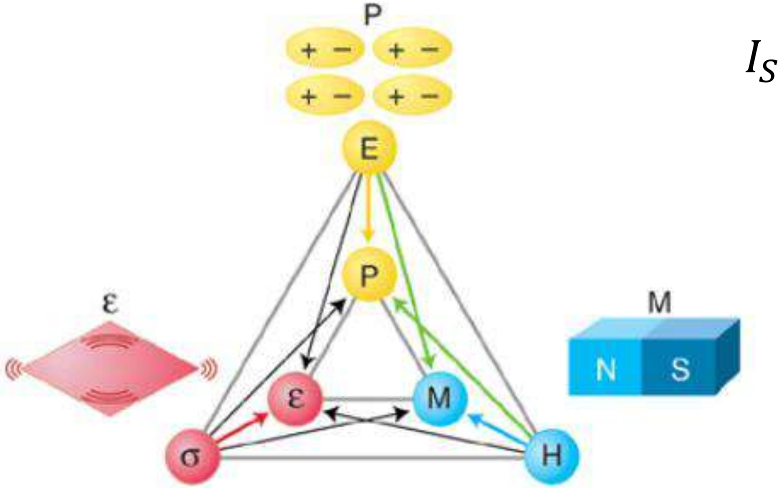
SHG in ferroelectric materials: Non-Ising FE domain wall

Cherifi et al. Nat. Comm. 8, 15768 (2017)





SHG in ferroelectric and magnetic materials: Multiferroics

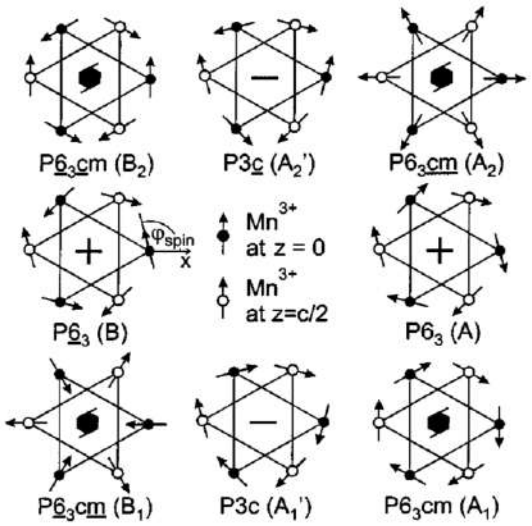


Spalding & Fiebig, Science, **309** *391* (2005)

Prototypical multiferroic materials:

$BiFeO_3$, $RMnO_3$…

$$I_{SHG} \propto \left|\vec{P}\right|^2 \quad \text{with} \quad \vec{P} = \varepsilon_0\left(\chi^{(i)} + \chi^{(c)}\right) : \vec{E}(\omega) \otimes \vec{E}(\omega)$$



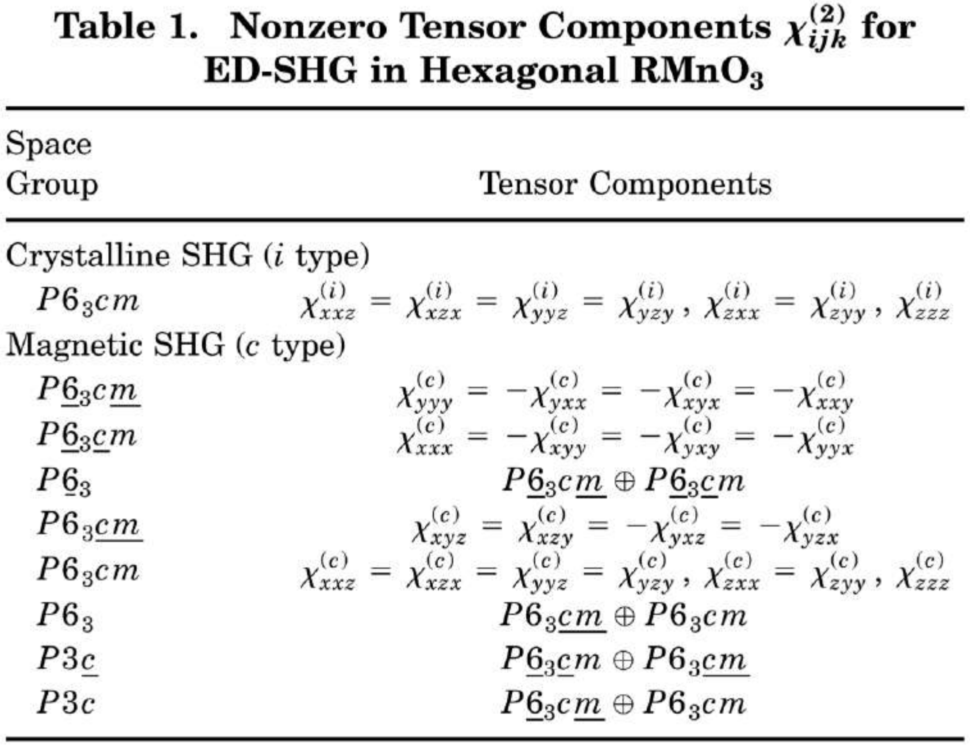
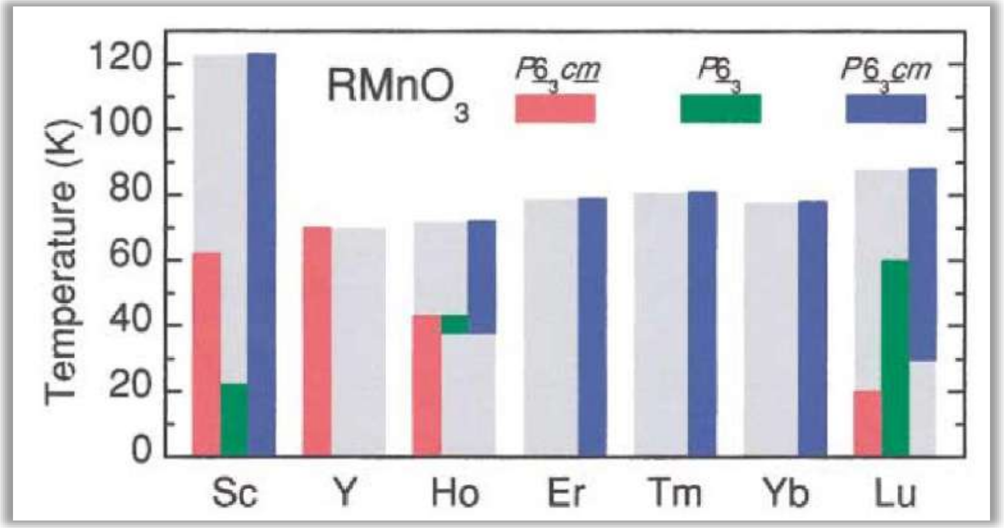


Table 1. Nonzero Tensor Components $\chi^{(2)}_{ijk}$ for ED-SHG in Hexagonal $RMnO_3$

Space Group	Tensor Components
Crystalline SHG (i type)	
$P6_3cm$	$\chi^{(i)}_{xxz} = \chi^{(i)}_{xzx} = \chi^{(i)}_{yyz} = \chi^{(i)}_{yzy}, \chi^{(i)}_{zxx} = \chi^{(i)}_{zyy}, \chi^{(i)}_{zzz}$
Magnetic SHG (c type)	
$P\underline{6}_3c\underline{m}$	$\chi^{(c)}_{yyy} = -\chi^{(c)}_{yxx} = -\chi^{(c)}_{xyx} = -\chi^{(c)}_{xxy}$
$P\underline{6}_3\underline{c}m$	$\chi^{(c)}_{xxx} = -\chi^{(c)}_{xyy} = -\chi^{(c)}_{yxy} = -\chi^{(c)}_{yyx}$
$P\underline{6}_3$	$P\underline{6}_3c\underline{m} \oplus P\underline{6}_3\underline{c}m$
$P6_3\underline{cm}$	$\chi^{(c)}_{xyz} = \chi^{(c)}_{xzy} = -\chi^{(c)}_{yxz} = -\chi^{(c)}_{yzx}$
$P6_3cm$	$\chi^{(c)}_{xxz} = \chi^{(c)}_{xzx} = \chi^{(c)}_{yyz} = \chi^{(c)}_{yzy}, \chi^{(c)}_{zxx} = \chi^{(c)}_{zyy}, \chi^{(c)}_{zzz}$
$P6_3$	$P6_3\underline{cm} \oplus P6_3cm$
$P3\underline{c}$	$P\underline{6}_3\underline{c}m \oplus P6_3\underline{cm}$
$P3c$	$P\underline{6}_3c\underline{m} \oplus P6_3cm$

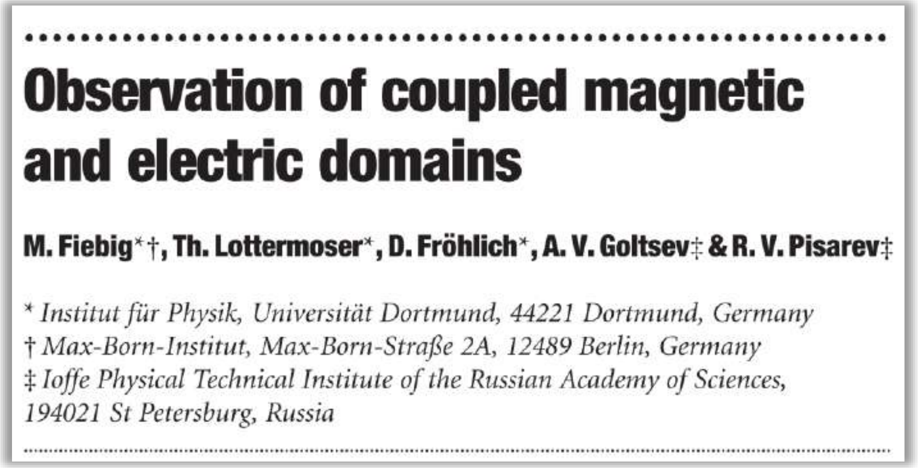


⇨ **Access to microscopic details of the magnetic structure**

Fiebig et al. *Phys. Rev. Lett.* 84, 5620 (2000)

Fiebig et al. *J. Opt. Soc. Am. B 22* 96 (2005)

SHG in ferroelectric and magnetic materials: Multiferroics



Observation of coupled magnetic and electric domains

M. Fiebig*†, Th. Lottermoser*, D. Fröhlich*, A. V. Goltsev‡ & R. V. Pisarev‡

** Institut für Physik, Universität Dortmund, 44221 Dortmund, Germany*
† Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin, Germany
‡ Ioffe Physical Technical Institute of the Russian Academy of Sciences, 194021 St Petersburg, Russia

NATURE | VOL 419 | 24 OCTOBER 2002 | www.nature.com/nature

$$\mathbf{S}(2\omega) = \epsilon_0(\hat{\chi}(0) + \hat{\chi}(P) + \hat{\chi}(l) + \hat{\chi}(Pl))\mathbf{E}(\omega)\mathbf{E}(\omega).$$

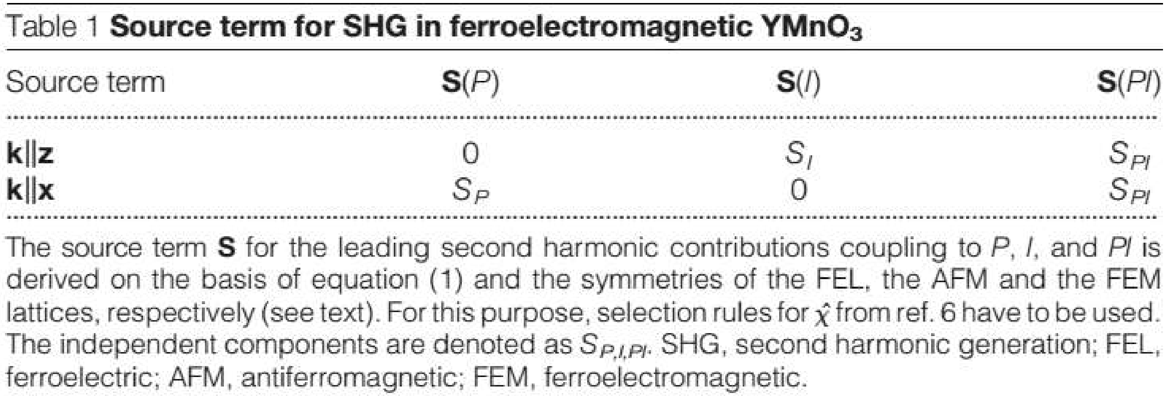
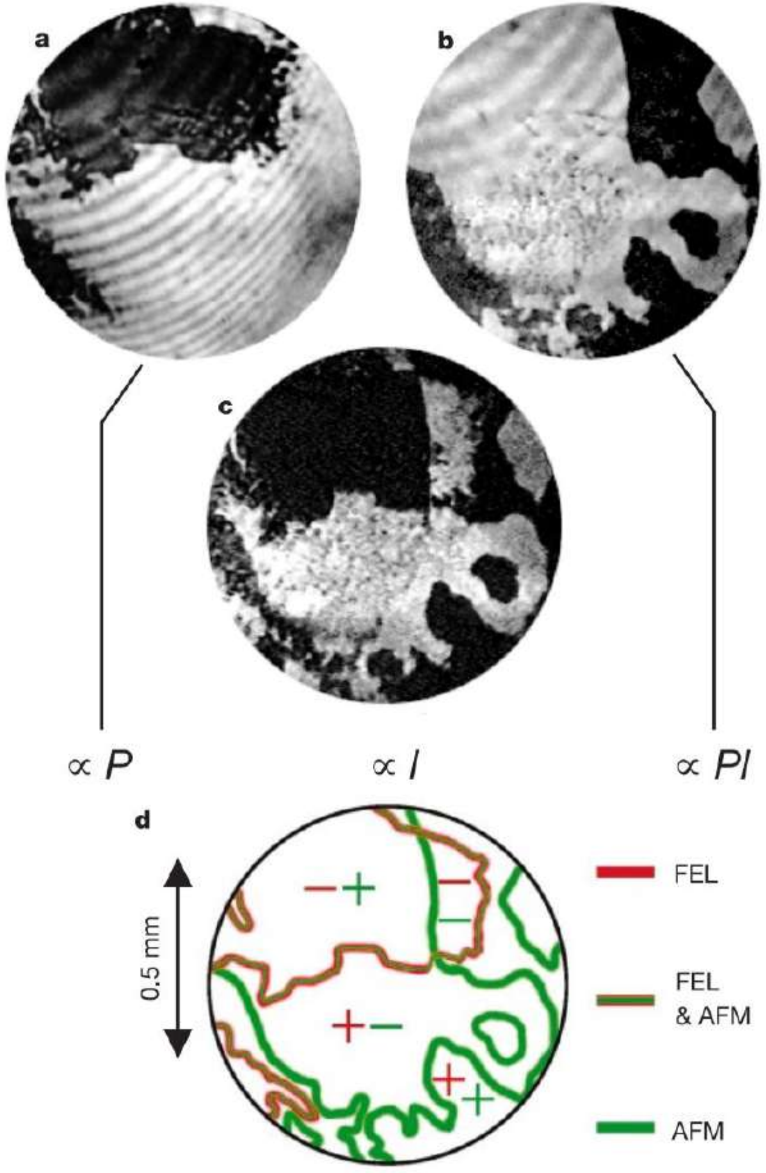


Table 1 **Source term for SHG in ferroelectromagnetic YMnO$_3$**

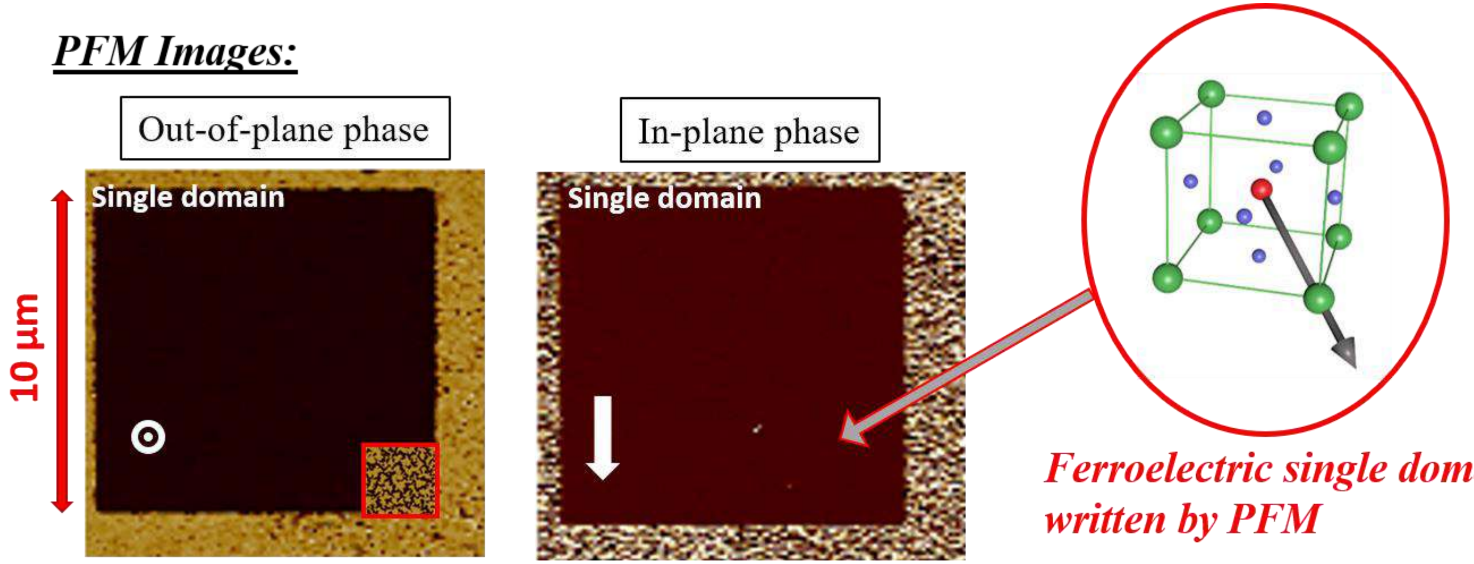
Source term	**S**(P)	**S**(l)	**S**(Pl)
k||**z**	0	S_l	S_{Pl}
k||**x**	S_P	0	S_{Pl}

The source term **S** for the leading second harmonic contributions coupling to P, l, and Pl is derived on the basis of equation (1) and the symmetries of the FEL, the AFM and the FEM lattices, respectively (see text). For this purpose, selection rules for $\hat{\chi}$ from ref. 6 have to be used. The independent components are denoted as $S_{P,l,Pl}$. SHG, second harmonic generation; FEL, ferroelectric; AFM, antiferromagnetic; FEM, ferroelectromagnetic.



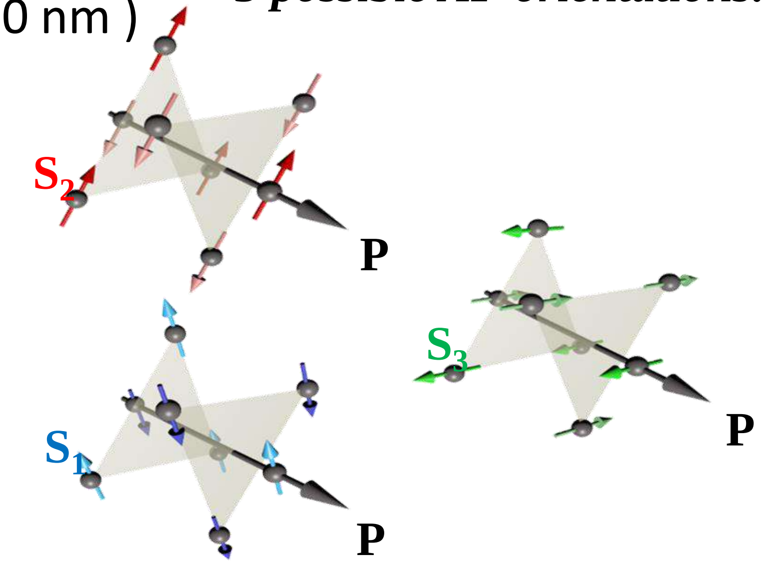
SHG in ferroelectric and magnetic materials: in BiFeO$_3$ 001 epitaxial layer (100 nm)

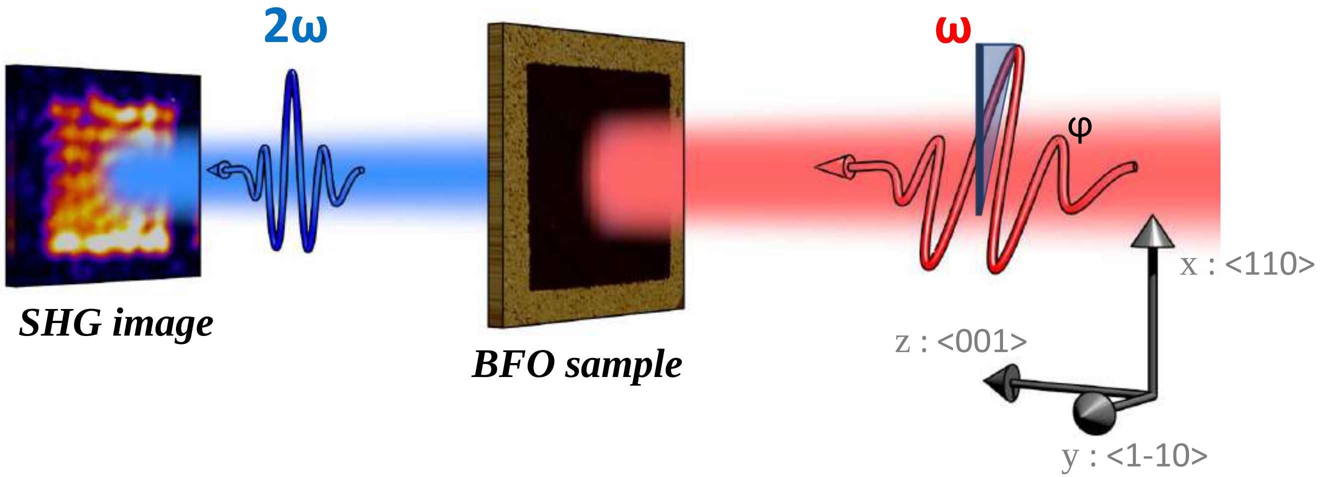
PFM Images:



Ferroelectric single domain written by PFM

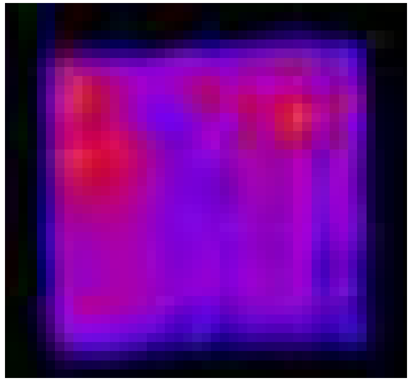
3 possible AF orientations:





SHG image

BFO sample

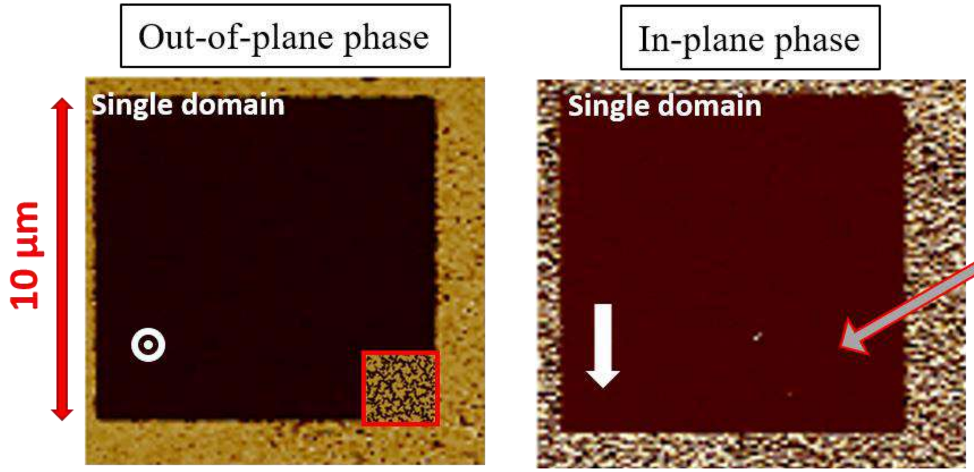


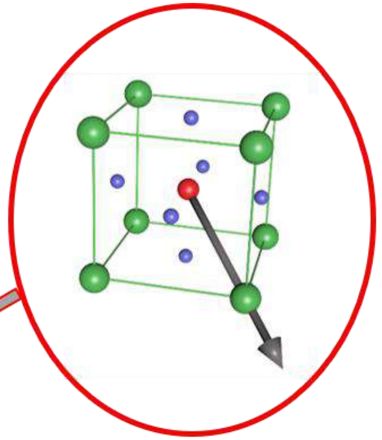
Full angular polarization dependence

Chauleau et al. *Nat. Materials* 16, 803 (2017)

SHG in ferroelectric and magnetic materials: in BiFeO$_3$ 001 epitaxial layer (100 nm)

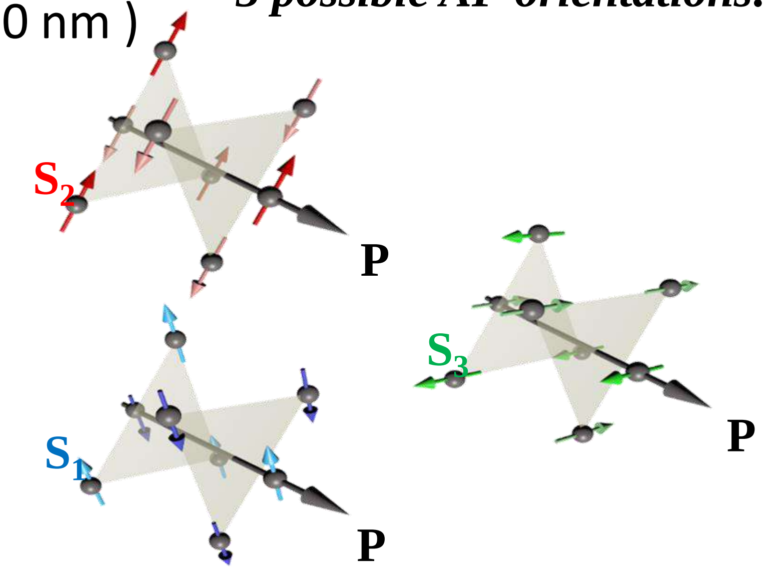
PFM Images:

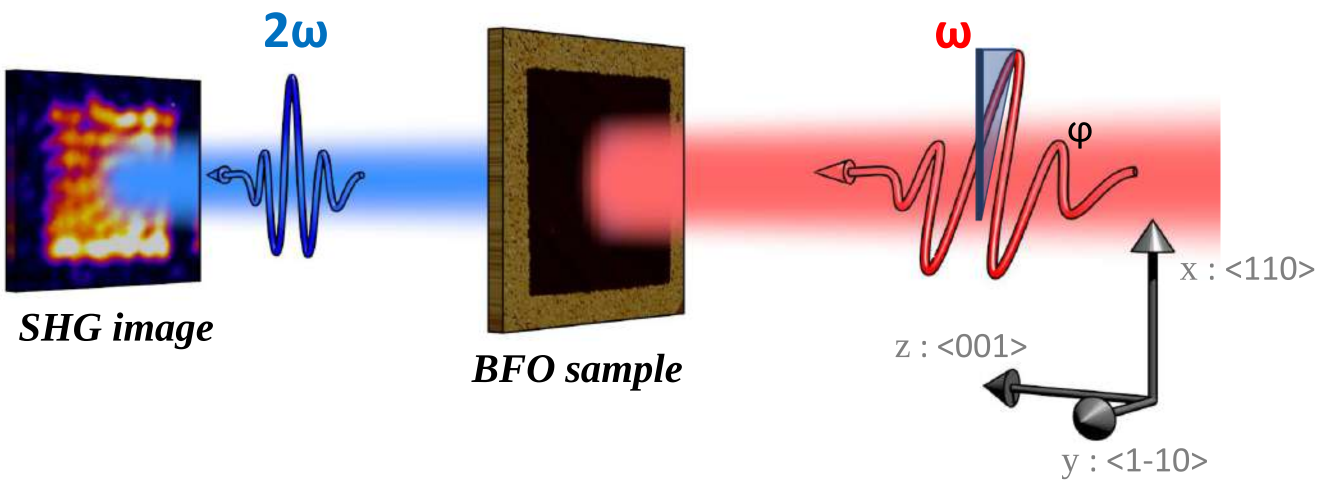




Ferroelectric single domain written by PFM

3 possible AF orientations:

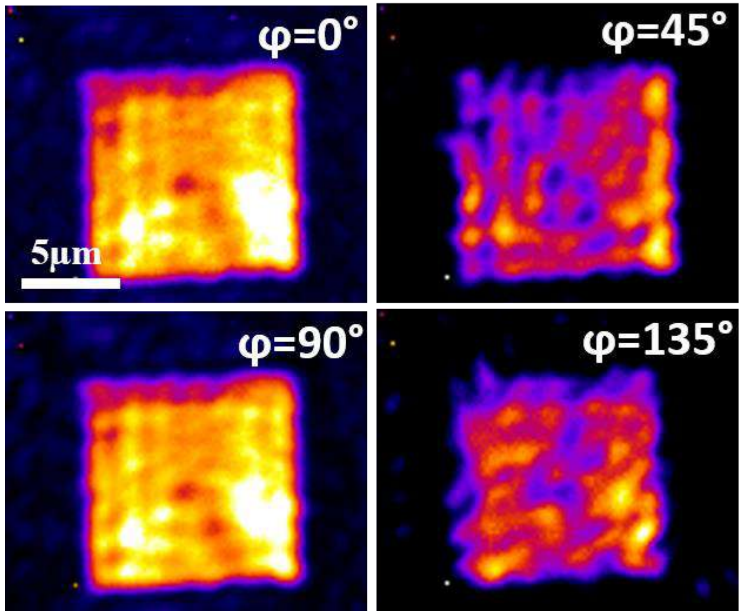




SHG image

BFO sample

Set of SHG images

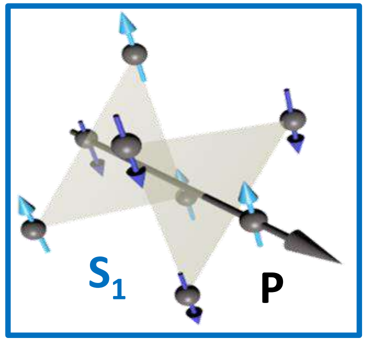


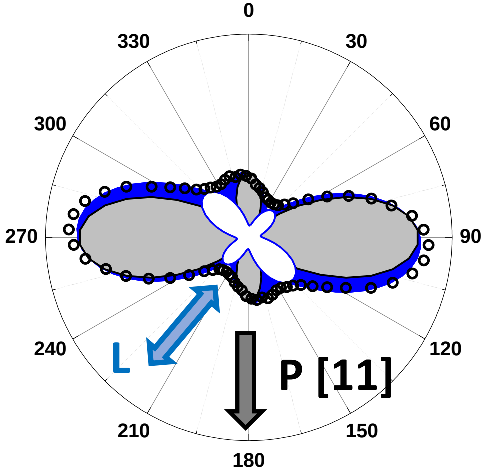
Chauleau et al. *Nat. Materials* 16, 803 (2017)

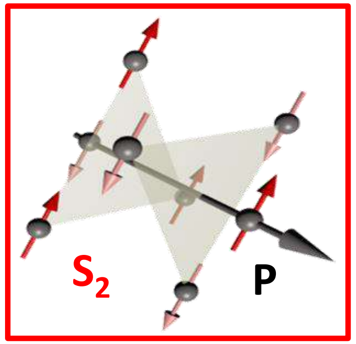
SHG in ferroelectric and magnetic materials: in BiFeO$_3$ 001 epitaxial layer (100 nm)

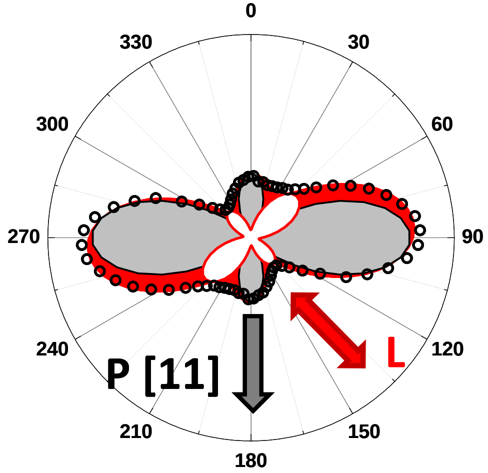
$$\begin{pmatrix}\mathbf{P}\\ \mathbf{M}\\ \mathbf{Q}\end{pmatrix}^{(2\omega)} \propto \begin{pmatrix} \boxed{\hat{\chi}^{eee}} & \hat{\chi}^{eem} & \hat{\chi}^{emm}\\ \hat{\chi}^{mee} & \hat{\chi}^{mem} & \hat{\chi}^{mmm}\\ \hat{\chi}^{qee} & \hat{\chi}^{qem} & \hat{\chi}^{qmm}\end{pmatrix}\begin{pmatrix}\mathbf{EE}\\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

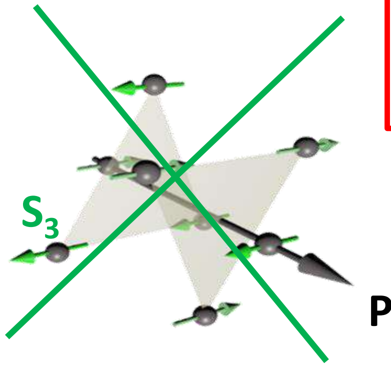
Antiferromagnetic configuration:











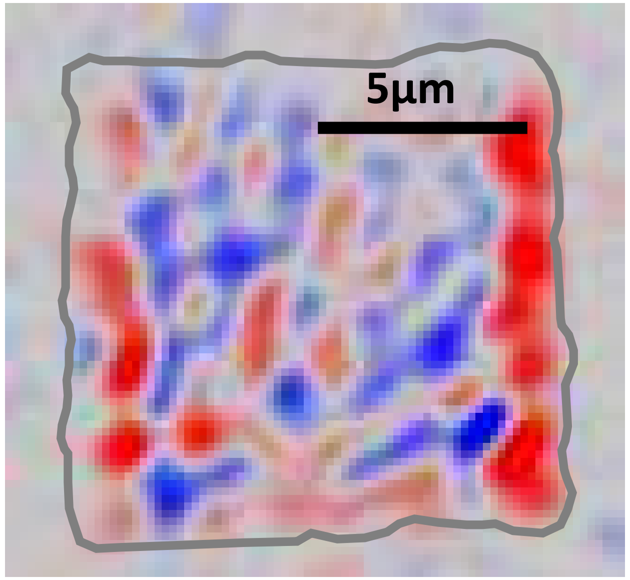
Chauleau et al. *Nat. Materials* 16, 803 (2017)

$$\vec{P} = \varepsilon_0\left(\chi^{(i)} + \chi^{(c)}\right): \vec{E}(\omega) \otimes \vec{E}(\omega)\Big)$$

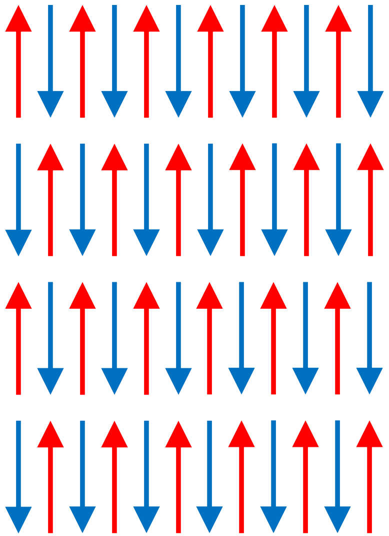
$$\swarrow \qquad \searrow$$

$$\propto \vec{P} \qquad\qquad \propto \vec{P}\cdot\vec{L}$$

Reconstruction of the image depending on the asymmetry



SHG in "pure" antiferromagnets:

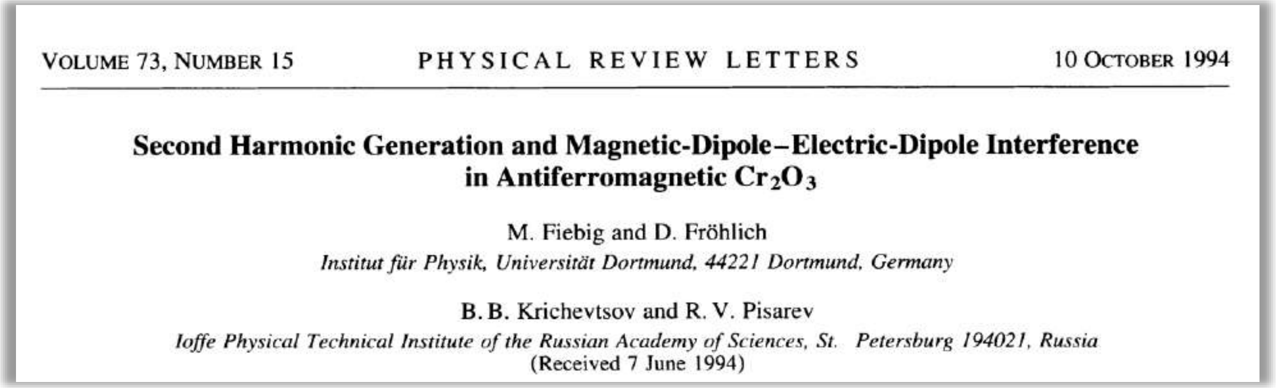


- **Antiparallel arrangement of spins**

(No dipolar field, No net Magnetization)

⇨ **Fairly difficult to access/image**

First demontration in Cr_2O_3:



VOLUME 73, NUMBER 15 PHYSICAL REVIEW LETTERS 10 OCTOBER 1994

Second Harmonic Generation and Magnetic-Dipole–Electric-Dipole Interference in Antiferromagnetic Cr_2O_3

M. Fiebig and D. Fröhlich
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B. B. Krichevtsov and R. V. Pisarev
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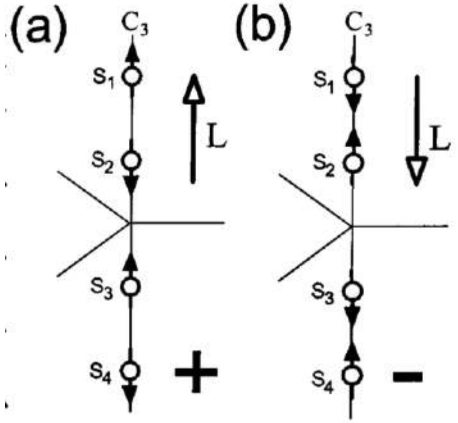
(Received 7 June 1994)

T>T$_N$ $\bar{3}m$ ⇨ $\cancel{\chi^{eee(i)}}$ but $\chi^{mee(i)}$

T<T$_N$ $\underline{\boldsymbol{\bar{3}m}}$ ⇨ $\chi^{eee(c)}$

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

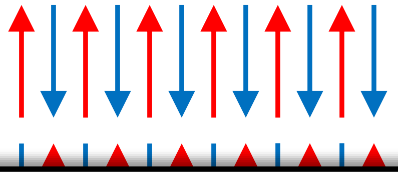


$$\mathbf{S} = \mu_0\left(\nabla \times \frac{\partial \mathbf{M}_{\mathrm{NL}}}{\partial t} + \frac{\partial^2 \mathbf{P}_{\mathrm{NL}}}{\partial t^2}\right) = 4\frac{\omega^2}{c^2}\begin{pmatrix} \chi_m(i)(E_x^2 - E_y^2) + 2\chi_e(c)E_xE_y \\ -2\chi_m(i)E_xE_y + \chi_e(c)(E_x^2 - E_y^2) \\ 0 \end{pmatrix},$$

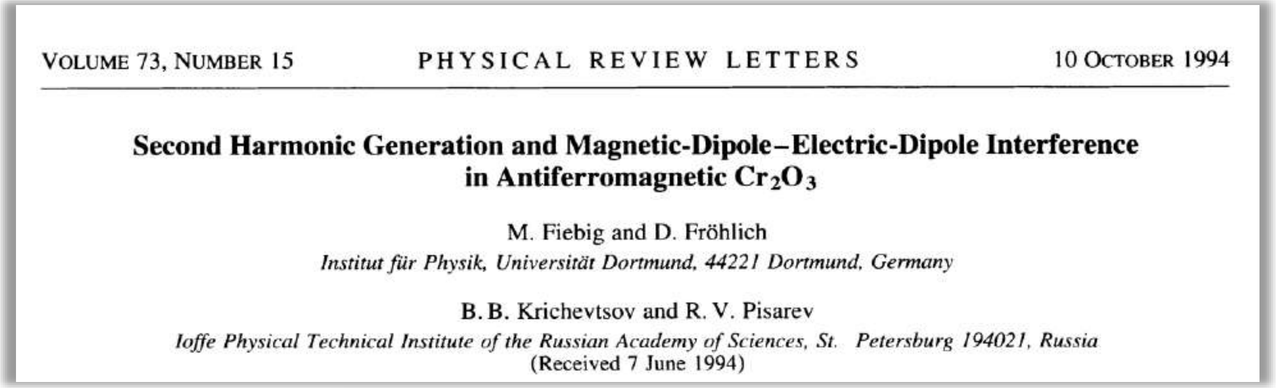
Interference term

$$\Rightarrow \quad I(l,\sigma) \propto I_\sigma^2[|\chi_m(i)|^2 + |\chi_e(c)|^2 \boxed{- \Delta(l)\sigma}],$$

SHG in “pure” antiferromagnets:



First demontration in Cr_2O_3:



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(Received 7 June 1994)

Fiebig et al. *Phys. Rev. Lett. 66*, 2906 (1995)

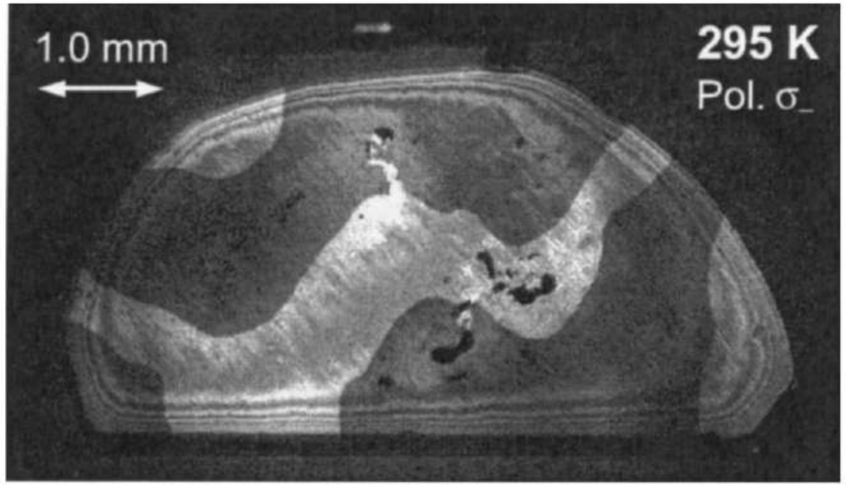


Fig. 6. Antiferromagnetic 180° domains in Cr_2O_3 exposed to circularly polarized light for SHG at 2.1 eV. Exposure time was 35 min but was reduced to 1–5 min in subsequent experiments.

Fiebig et al. *Phys. Rev. B 54*, 12681 (1996)

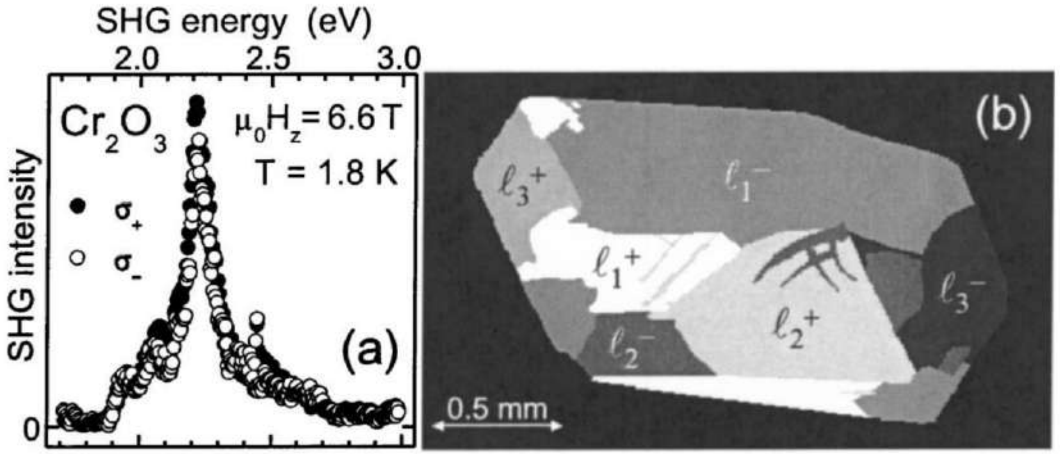
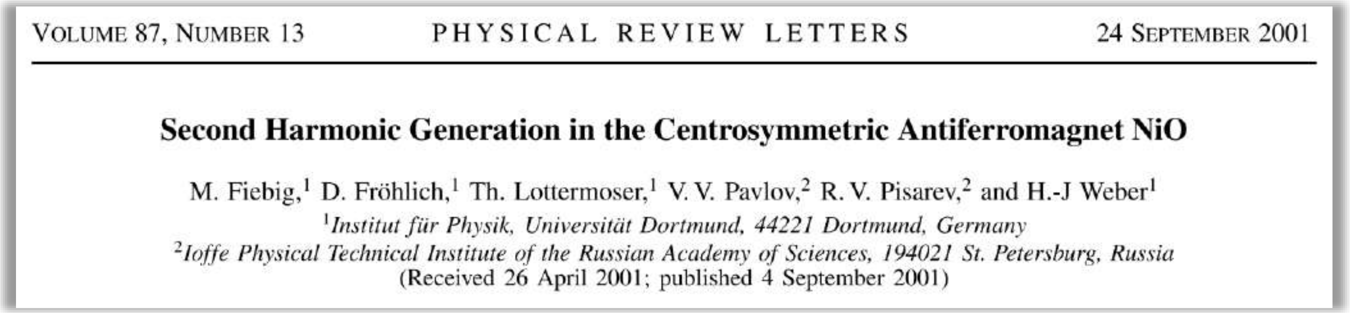


Fig. 7. (a) SHG spectrum of Cr_2O_3 in the spin-flop phase taken with circularly polarized light incident along the z axis. (b) Distribution of the six orientational domains, $\ell^{\pm}_{1,2,3}$, that exist in the spin-flop phase.

SHG in “pure” antiferromagnets:



Volume 87, Number 13 PHYSICAL REVIEW LETTERS 24 September 2001

Second Harmonic Generation in the Centrosymmetric Antiferromagnet NiO

M. Fiebig,[1] D. Fröhlich,[1] Th. Lottermoser,[1] V. V. Pavlov,[2] R. V. Pisarev,[2] and H.-J Weber[1]

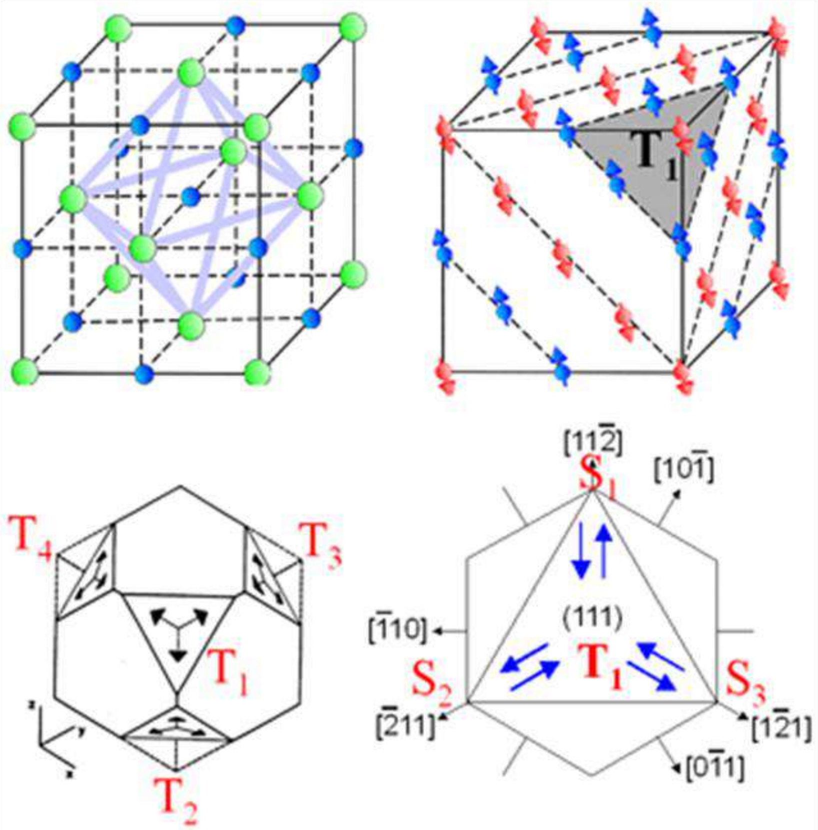
[1]*Institut für Physik, Universität Dortmund, 44221 Dortmund, Germany*

[2]*Ioffe Physical Technical Institute of the Russian Academy of Sciences, 194021 St. Petersburg, Russia*

(Received 26 April 2001; published 4 September 2001)

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

Antiferromagnetic order in NiO (G-type):



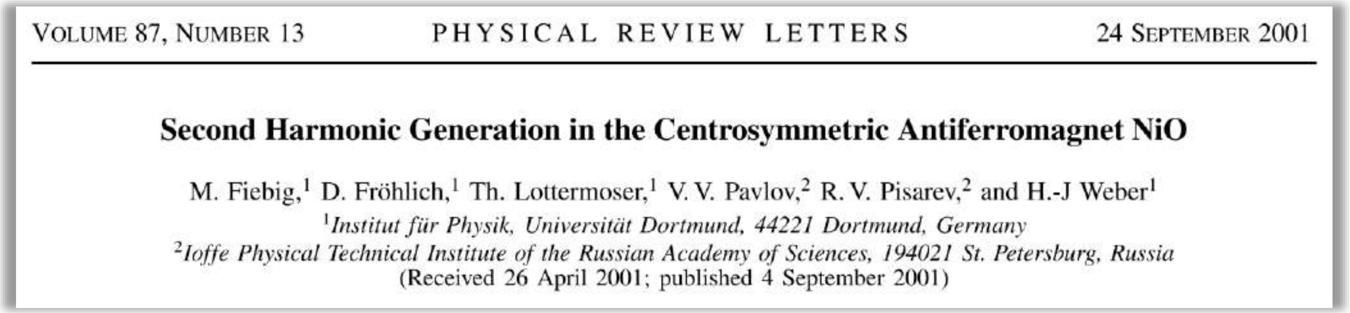
Centrosymmetric cristal structure

$T<T_N$ 2/m

Centrosymmetric magnetic structure

$T<T_N$ C_c2/c magnetic space group

SHG in "pure" antiferromagnets:



VOLUME 87, NUMBER 13 PHYSICAL REVIEW LETTERS 24 SEPTEMBER 2001

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[1] *Institut für Physik, Universität Dortmund, 44221 Dortmund, Germany*
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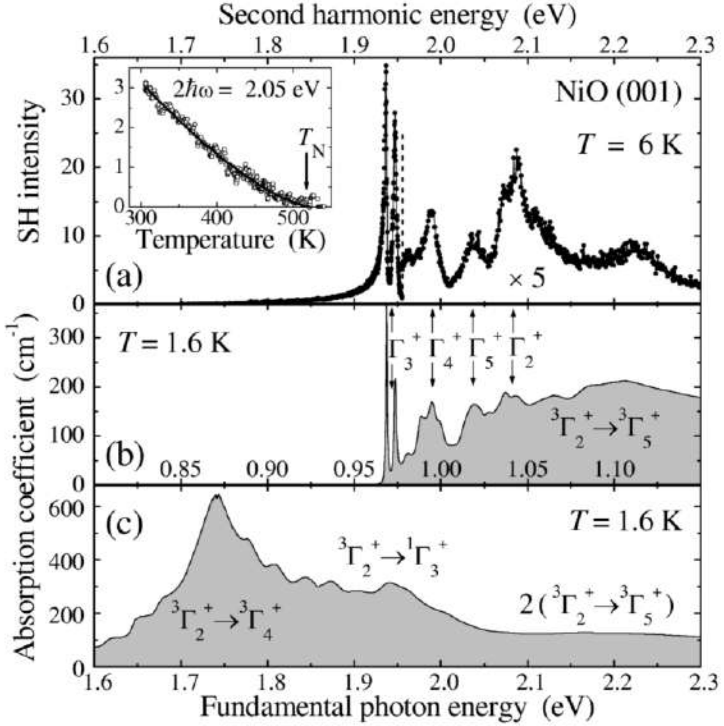
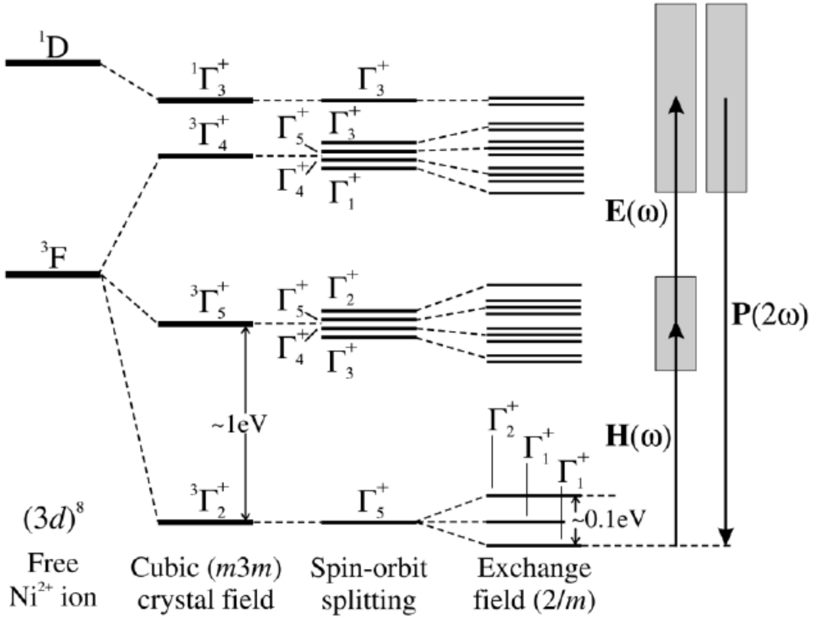
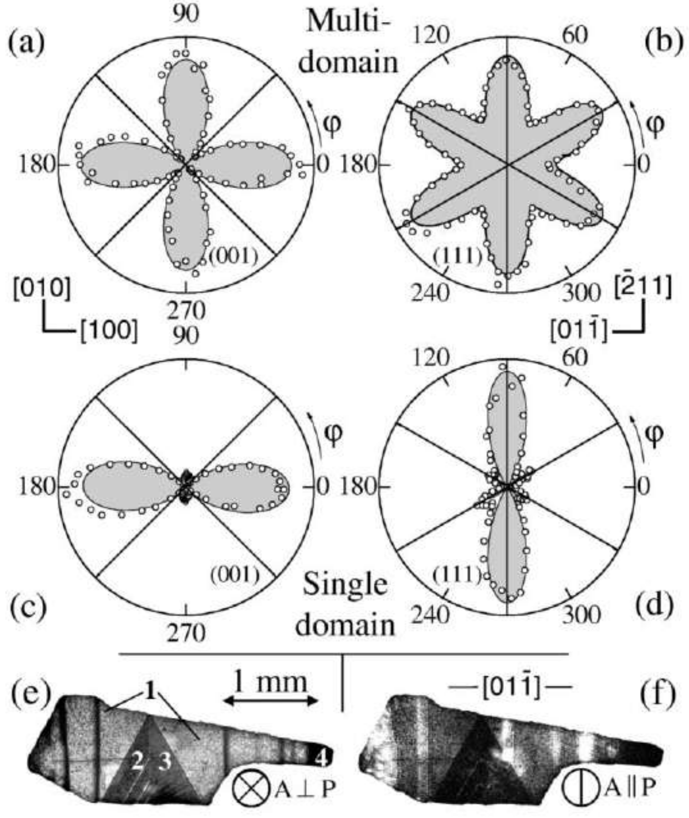
(Received 26 April 2001; published 4 September 2001)

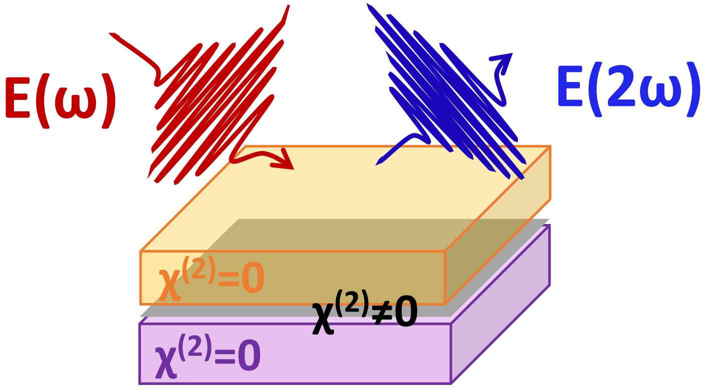
$$\begin{pmatrix} \mathbf{P} \\ \mathbf{M} \\ \mathbf{Q} \end{pmatrix}^{(2\omega)} \propto \begin{pmatrix} \cancel{\hat{\chi}^{eee}} & \hat{\chi}^{eem} & \cancel{\hat{\chi}^{emm}} \\ \hat{\chi}^{mee} & \cancel{\hat{\chi}^{mem}} & \hat{\chi}^{mmm} \\ \hat{\chi}^{qee} & \cancel{\hat{\chi}^{qem}} & \hat{\chi}^{qmm}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







SHG at interfaces:

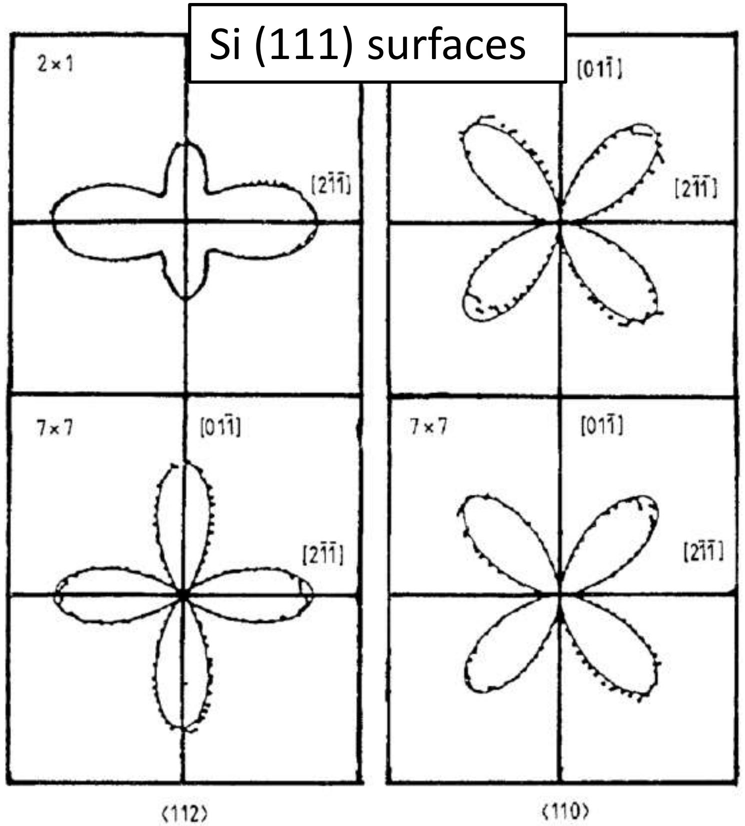


Symmetry is intrinsically broken at the interface/surface

$$\vec{P}(2\omega) \propto \chi_S{}^{eee(i)} : \vec{E}(\omega) \otimes \vec{E}(\omega)$$

$\chi_S{}^{eee(i)}$: Transform with the point group symmetry of the surface

Si (111) surfaces

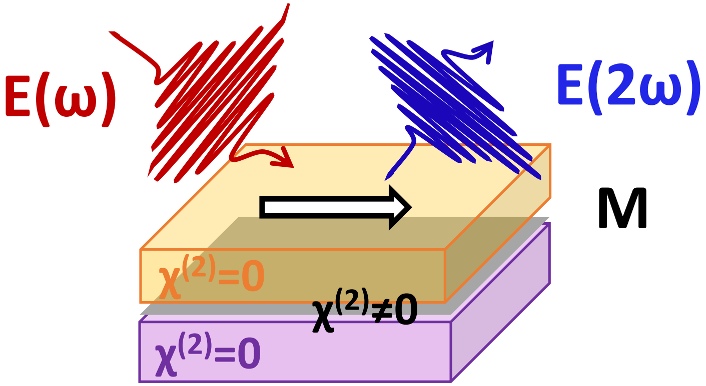


⇨ Single m symmetry

⇨ 3m symmetry

Heinz et al. *Phys. Rev. Lett. 54*, 63 (1985)

SHG at interfaces: Magneto-Optics



Breaking **time-reversal** symmetry and **space-inversion** symmetry are needed to observed MO SHG in the **electron-dipole approximation**

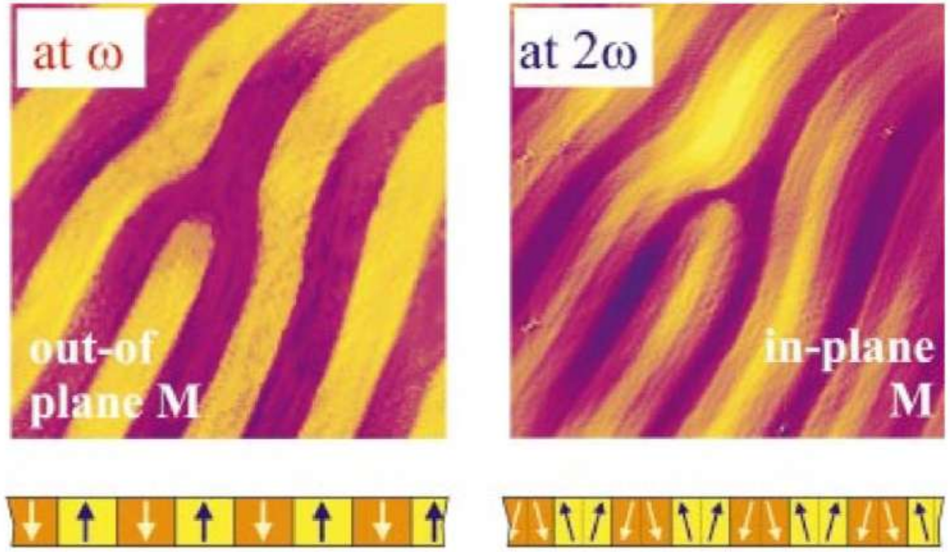
$$\vec{P}(2\omega) \propto \left(\chi_S{}^{eee(i)} + \chi_S{}^{eee(c)}\right) : \vec{E}(\omega) \otimes \vec{E}(\omega)$$

Could be written as:

Axial 4th rank

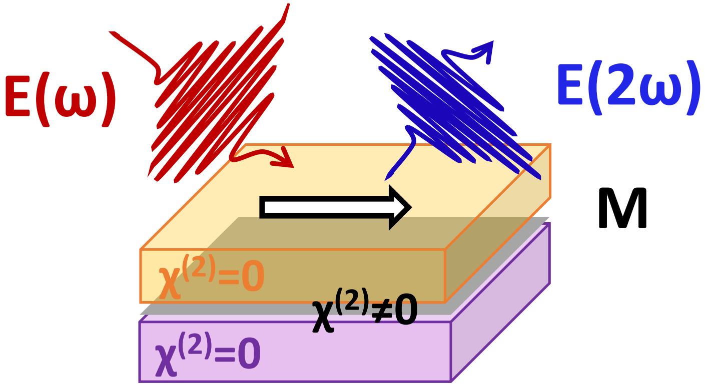
$$\mathbf{P}^{\mathrm{nl}}(2\omega) = \chi^{\mathrm{cr}}\mathbf{E}(\omega)\mathbf{E}(\omega) + \boxed{\chi^{\mathrm{magn}}}\mathbf{E}(\omega)\mathbf{E}(\omega)\mathbf{M},$$

$$\chi^{(2)} = \begin{pmatrix} \chi_{xxx}(M_y) & \chi_{xyy}(M_y) & \chi_{xzz}(M_y) & \chi_{xzy}(M_z) & \chi_{xzx}^{\mathrm{cr}} & \chi_{xxy}(M_x) \\ \chi_{yxx}(M_x) & \chi_{yyy}(M_x) & \chi_{xzz}(M_x) & \chi_{yzy}^{\mathrm{cr}} & \chi_{yzx}(M_z) & \chi_{yxy}(M_y) \\ \chi_{zxx}^{\mathrm{cr}} & \chi_{zyy}^{\mathrm{cr}} & \chi_{zzz}^{\mathrm{cr}} & \chi_{zzy}(M_x) & \chi_{zzx}(M_y) & \chi_{zxy}(M_z) \end{pmatrix}.$$



From: Kirilyuk and Raising et al. *J. Opt. Soc. Am. B* ***22*** 148 (2005)

SHG at interfaces: Magneto-Optics



Breaking **time-reversal** symmetry and **space-inversion** symmetry are needed to observed MO SHG in the **electron-dipole approximation**

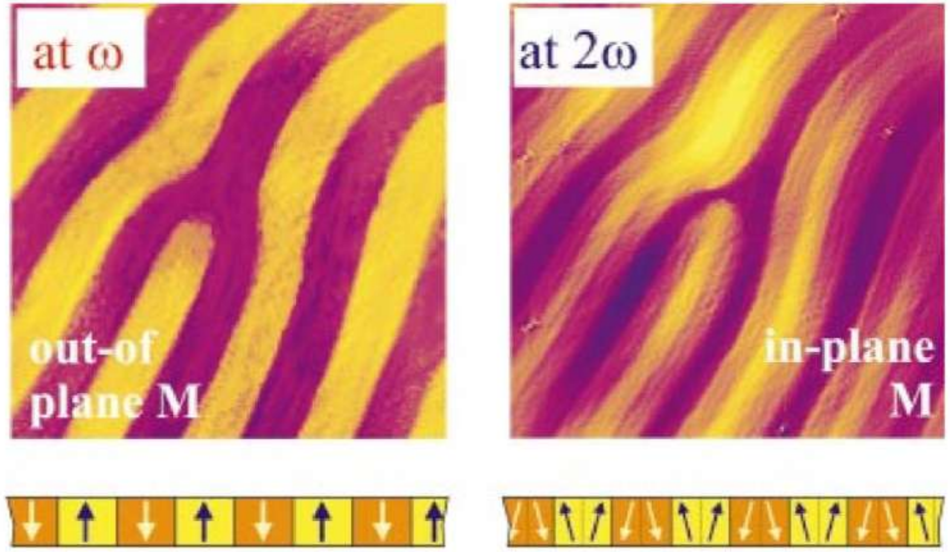
$$\vec{P}(2\omega) \propto \left(\chi_S{}^{eee(i)} + \chi_S{}^{eee(c)}\right) : \vec{E}(\omega) \otimes \vec{E}(\omega)$$

Could be written as:

Axial 4th rank

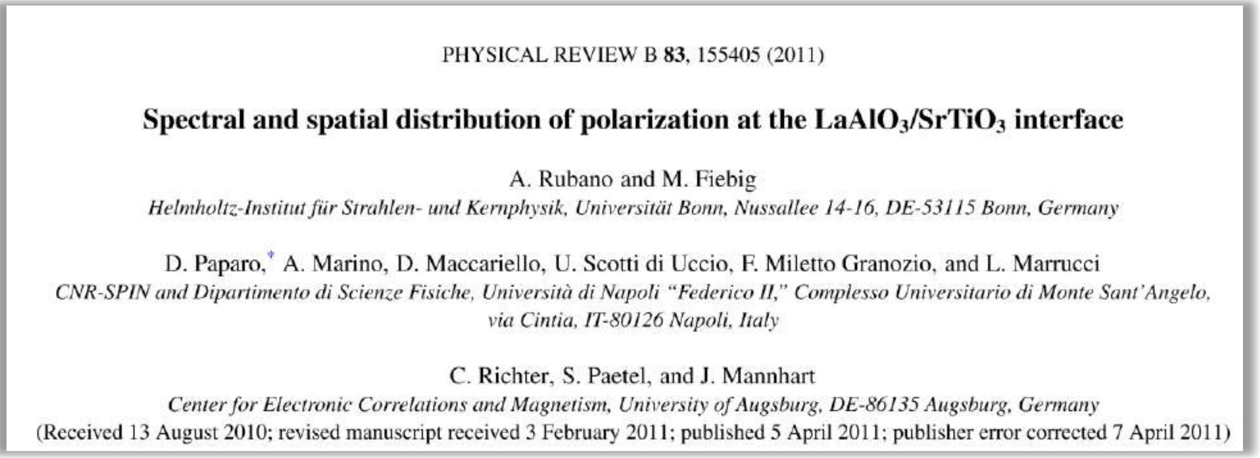
$$\mathbf{P}^{\mathrm{nl}}(2\omega) = \chi^{\mathrm{cr}}\mathbf{E}(\omega)\mathbf{E}(\omega) + \chi^{\mathrm{magn}}\mathbf{E}(\omega)\mathbf{E}(\omega)\mathbf{M},$$

$$\chi^{(2)} = \begin{pmatrix} \chi_{xxx}(M_y) & \chi_{xyy}(M_y) & \chi_{xzz}(M_y) & \chi_{xzy}(M_z) & \chi^{\mathrm{cr}}_{xzx} & \chi_{xxy}(M_x) \\ \chi_{yxx}(M_x) & \chi_{yyy}(M_x) & \chi_{xzz}(M_x) & \chi^{\mathrm{cr}}_{yzy} & \chi_{yzx}(M_z) & \chi_{yxy}(M_y) \\ \chi^{\mathrm{cr}}_{zxx} & \chi^{\mathrm{cr}}_{zyy} & \chi^{\mathrm{cr}}_{zzz} & \chi_{zzy}(M_x) & \chi_{zzx}(M_y) & \chi_{zxy}(M_z) \end{pmatrix}.$$



From: Kirilyuk and Raising et al. *J. Opt. Soc. Am. B* ***22*** 148 (2005)

SHG at interfaces (case of $LaAlO_3/SrTiO_3$):



PHYSICAL REVIEW B **83**, 155405 (2011)

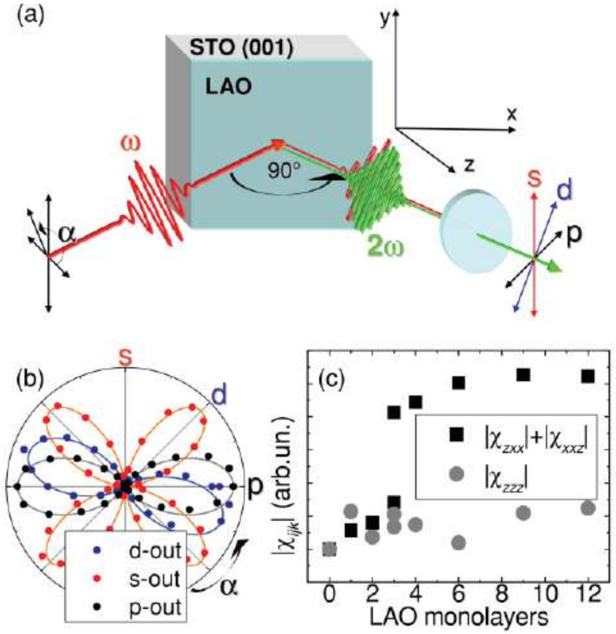
Spectral and spatial distribution of polarization at the $LaAlO_3/SrTiO_3$ interface

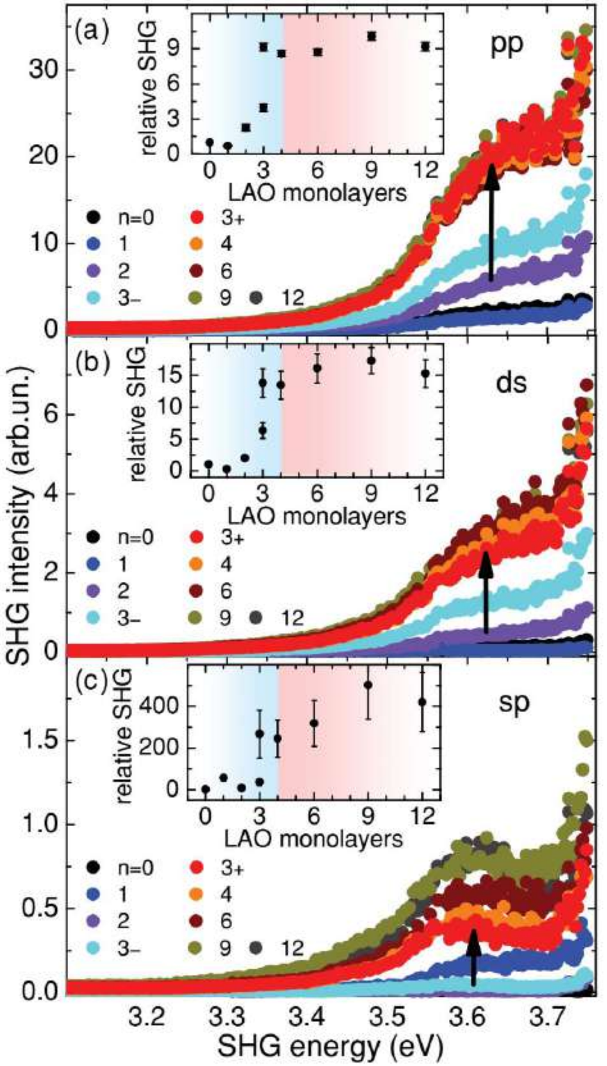
A. Rubano and M. Fiebig
Helmholtz-Institut für Strahlen- und Kernphysik, Universität Bonn, Nussallee 14-16, DE-53115 Bonn, Germany

D. Paparo,* A. Marino, D. Maccariello, U. Scotti di Uccio, F. Miletto Granozio, and L. Marrucci
CNR-SPIN and Dipartimento di Scienze Fisiche, Università di Napoli "Federico II," Complesso Universitario di Monte Sant'Angelo, via Cintia, IT-80126 Napoli, Italy

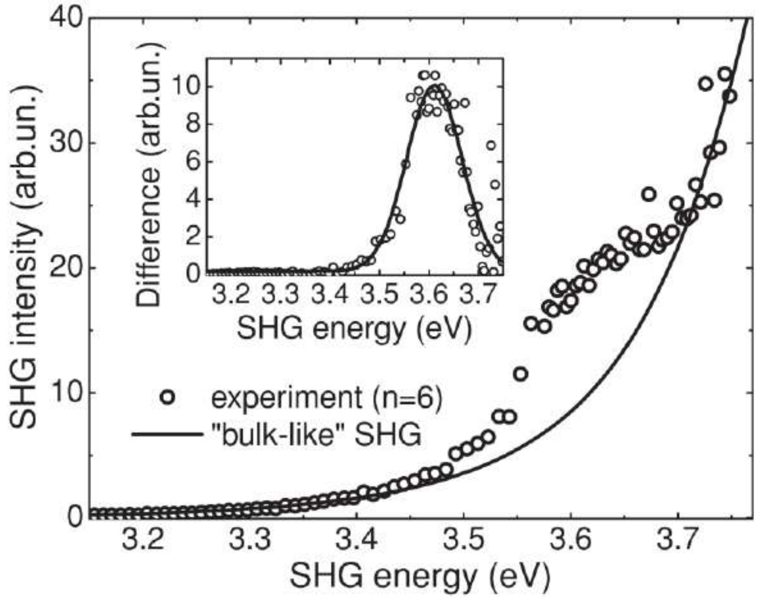
C. Richter, S. Paetel, and J. Mannhart
Center for Electronic Correlations and Magnetism, University of Augsburg, DE-86135 Augsburg, Germany

(Received 13 August 2010; revised manuscript received 3 February 2011; published 5 April 2011; publisher error corrected 7 April 2011)





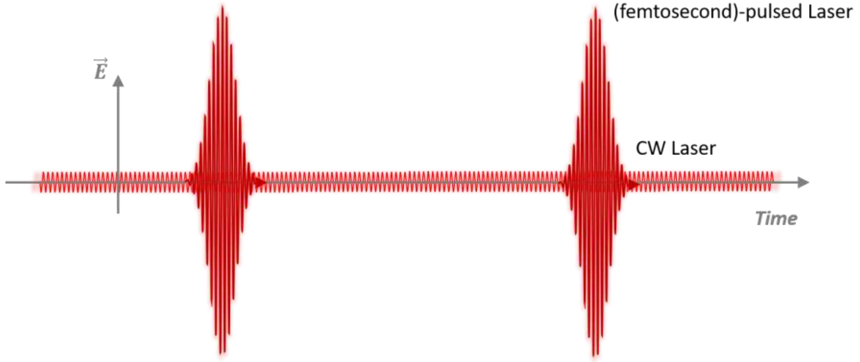
Polarization configuration		SHG
Ingoing	Outgoing	susceptibility
s	p	$\chi_{zxx} = \chi_{zyy}$
d	s	$\chi_{xxz} = \chi_{xzx} = \chi_{yyz} = \chi_{yzy}$
p	p	$a\chi_{zzz} + a'\chi_{zxx} + a''\chi_{xxz}$

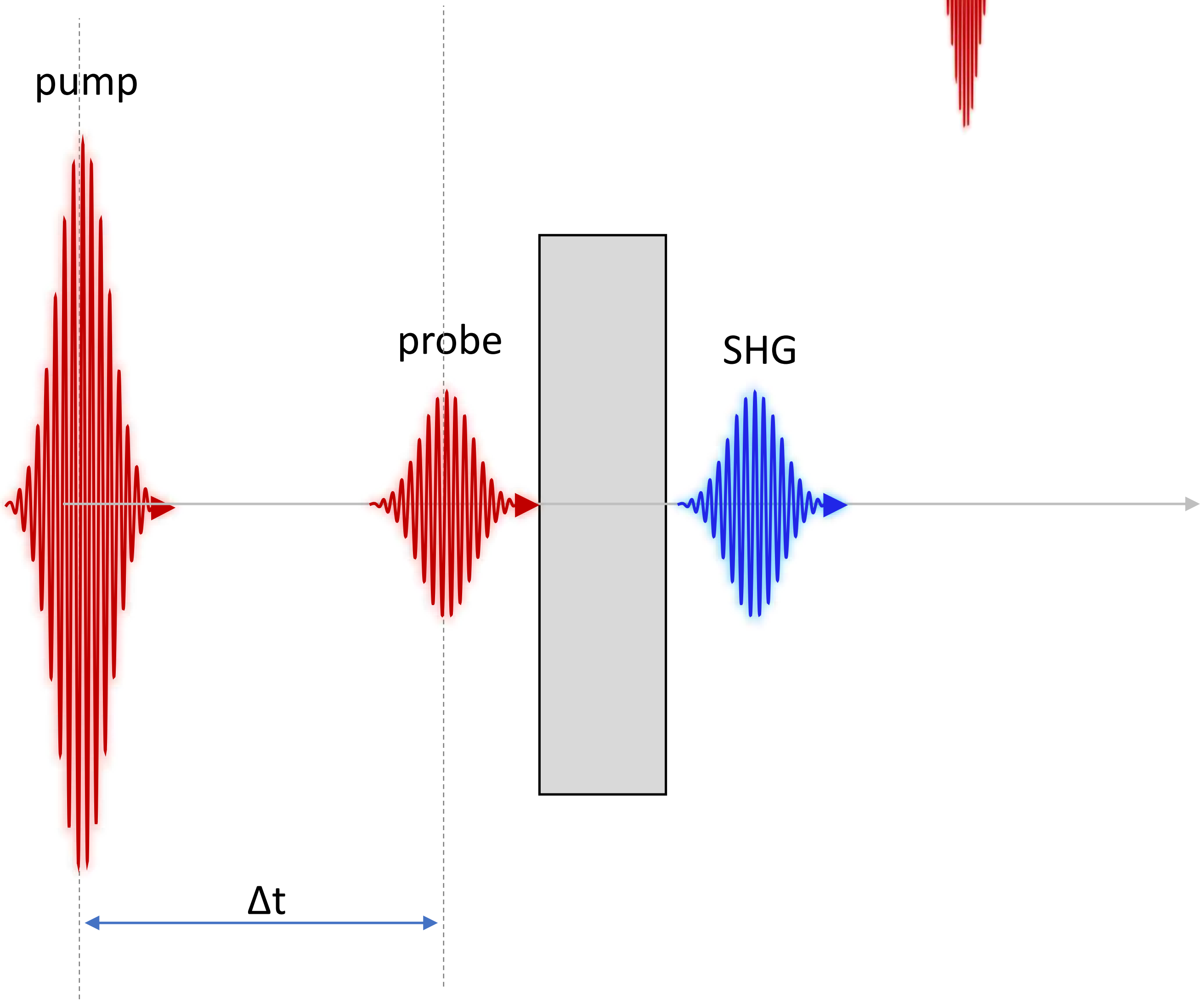


See also:
Savoia et al. *Phys. Rev. B* **80** 075110 (2009)
Ogawa et al. *Phys. Rev. B* **80** 081106R (2009)
Paparo et al. *J. Opt. Soc. Am. B* **30** 2452 (2013)

SHG: ultrafast capabilities

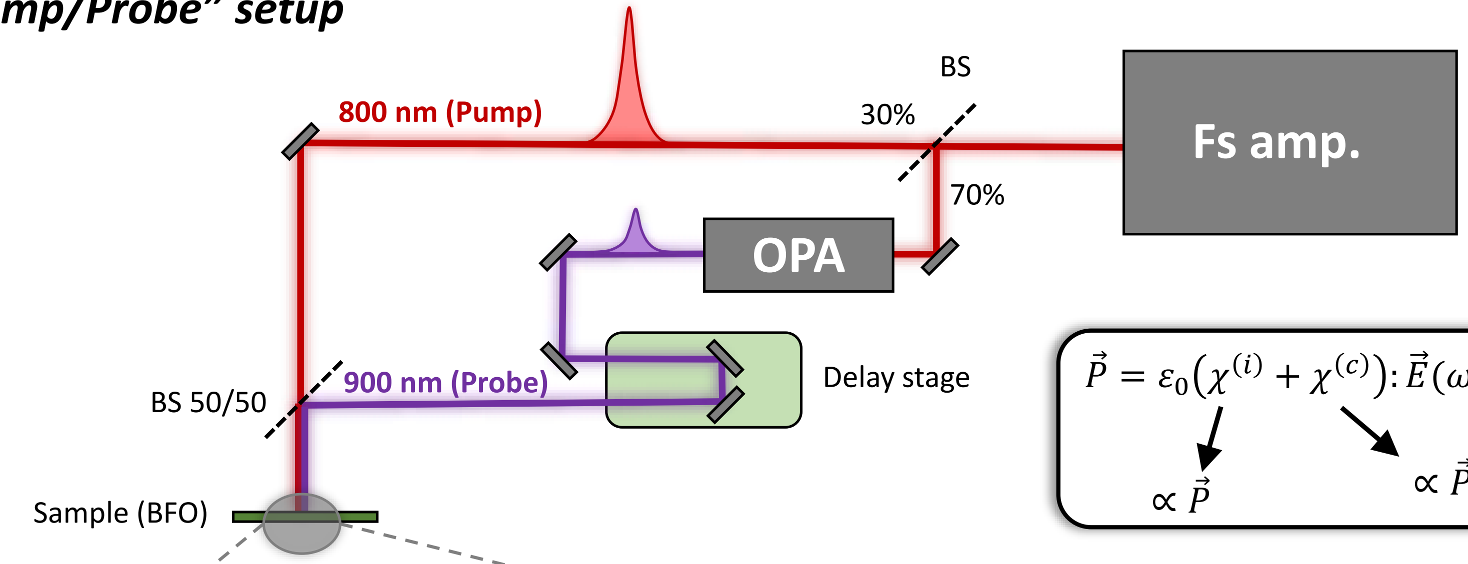
Time resolved Pump-probe approach

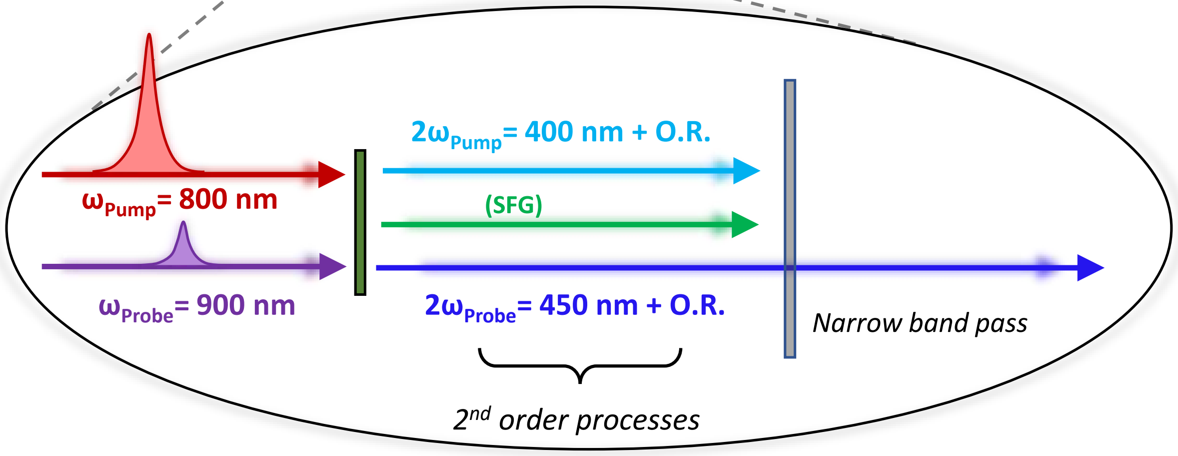




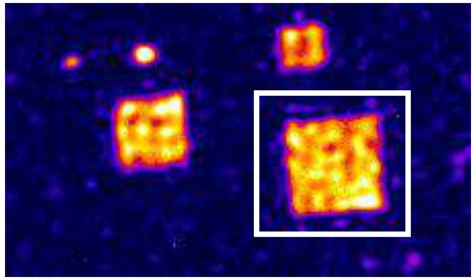
Time-resolved SHG:

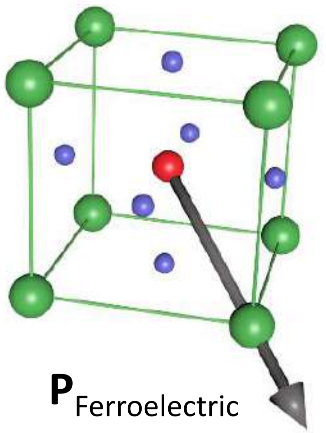
"Pump/Probe" setup





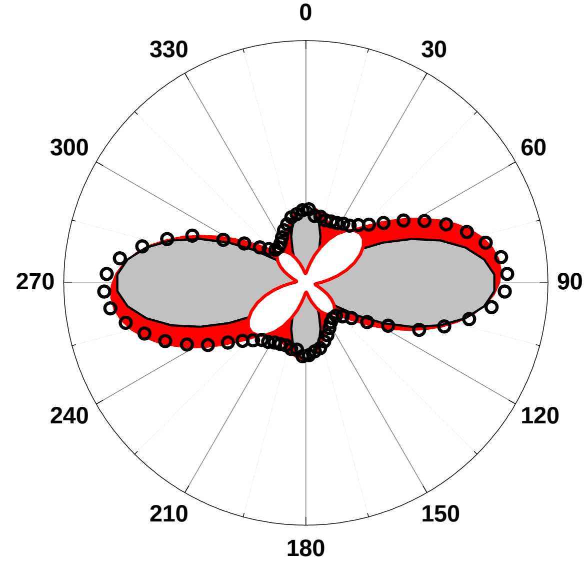
Single FE domain in $BiFeO_3$





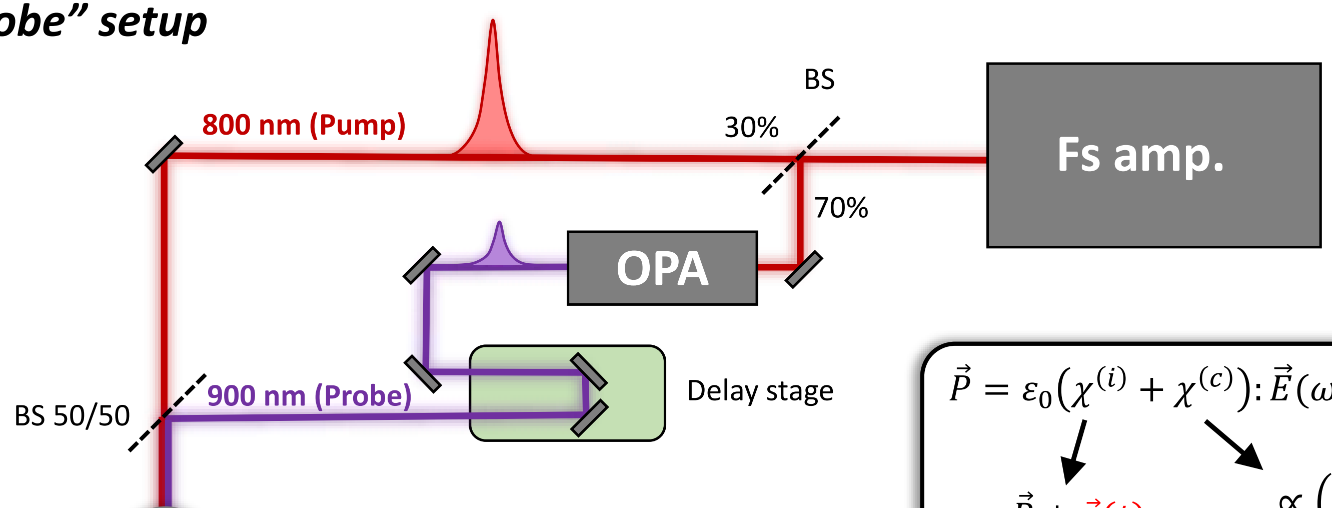
$$\vec{P} = \varepsilon_0\left(\chi^{(i)} + \chi^{(c)}\right):\vec{E}(\omega)\otimes\vec{E}(\omega)$$

$\propto \vec{P}$ $\qquad \propto \vec{P}\cdot\vec{L}$

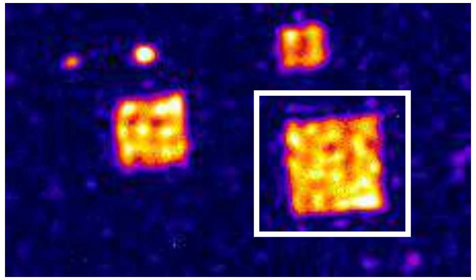


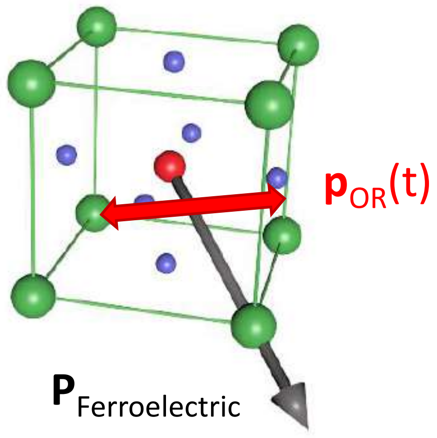
Time-resolved SHG:

"Pump/Probe" setup



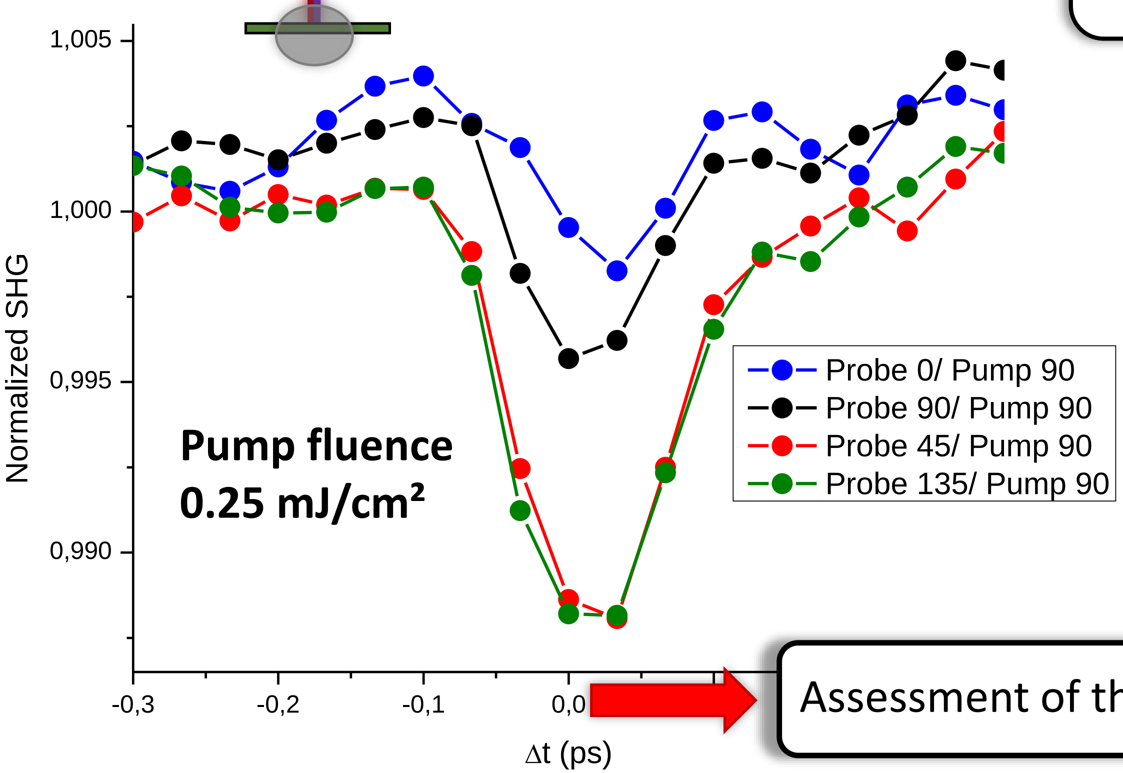
Single FE domain in $BiFeO_3$

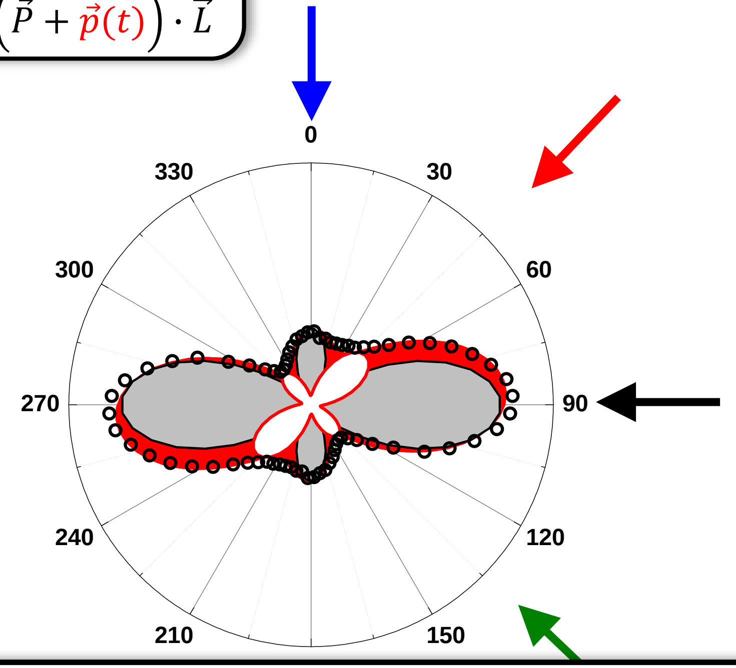




$$\vec{P} = \varepsilon_0\left(\chi^{(i)} + \chi^{(c)}\right) : \vec{E}(\omega) \otimes \vec{E}(\omega)$$

$\propto \vec{P} + \vec{p}(t)$ $\qquad$ $\propto \left(\vec{P} + \vec{p}(t)\right) \cdot \vec{L}$





Assessment of the ultrafast electrical polarization induced by optical rectification

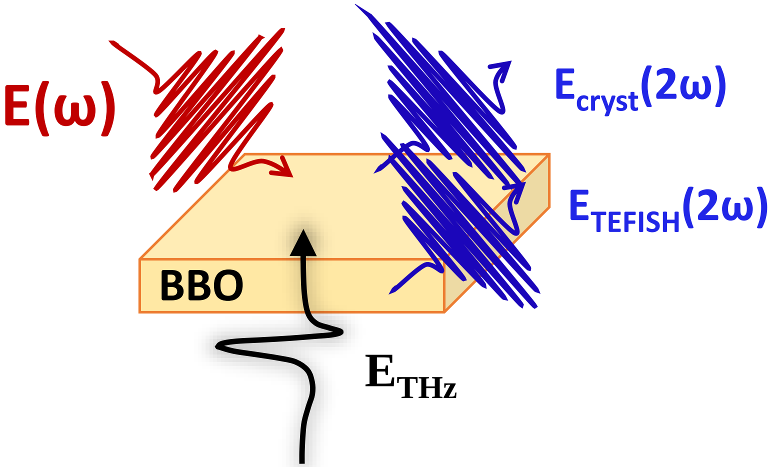
Time-resolved SHG:

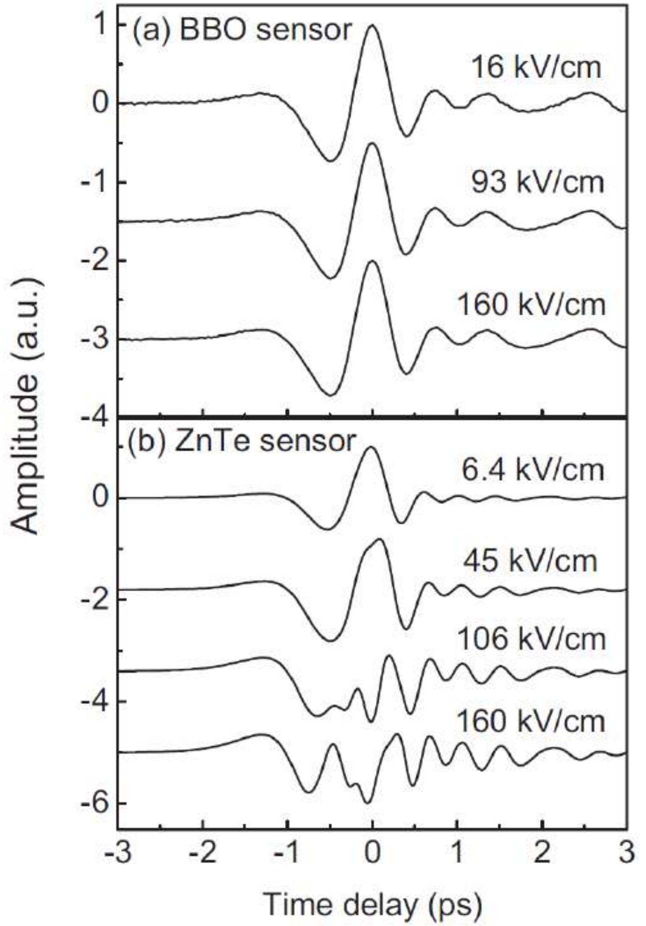
APPLIED PHYSICS LETTERS **95**, 011118 (2009)

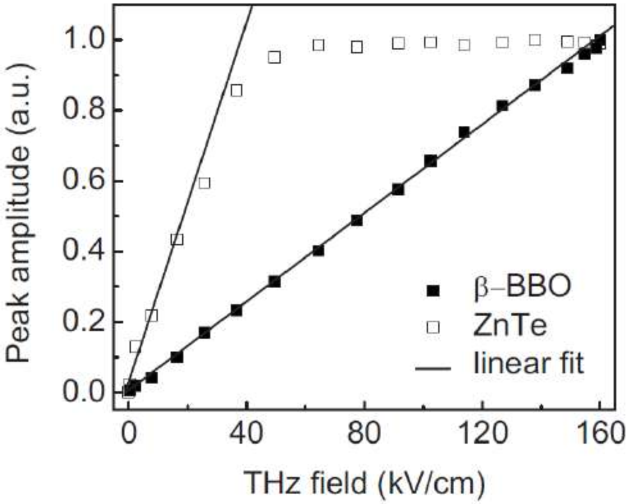
Terahertz-field-induced second-harmonic generation in a beta barium borate crystal and its application in terahertz detection

Jian Chen, Pengyu Han, and X.-C. Zhang[a]
Center for Terahertz Research, Rensselaer Polytechnic Institute, Troy, New York 12180, USA

(Received 28 April 2009; accepted 21 June 2009; published online 10 July 2009)

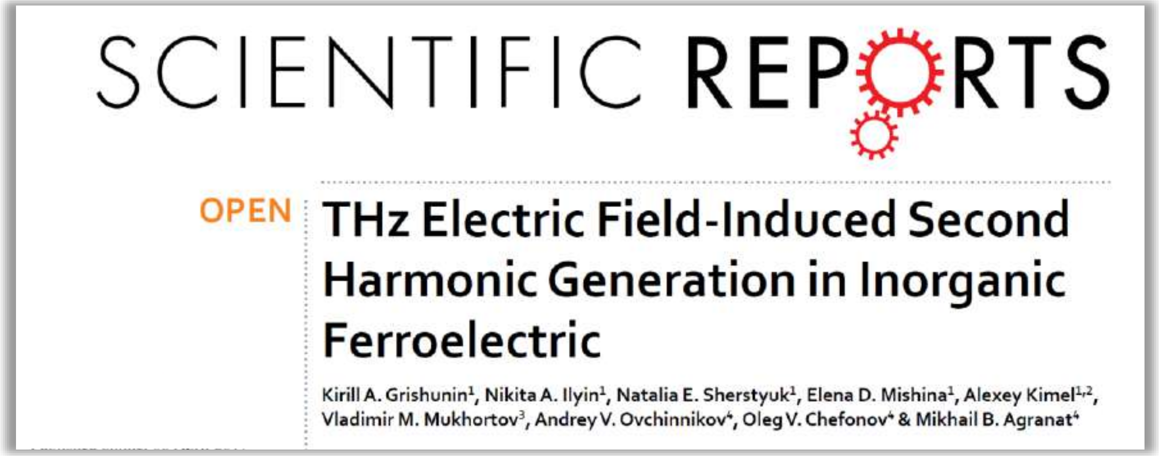






$$\vec{P}(2\omega) = \vec{P}^{\,cryst}(2\omega) + \vec{P}^{\,TEFISH}(2\omega) = \hat{\chi}^{(2)}\vec{E}_\omega\vec{E}_\omega + \hat{\chi}^{(3)}\vec{E}_\Omega\vec{E}_\omega\vec{E}_\omega.$$

Time-resolved SHG:

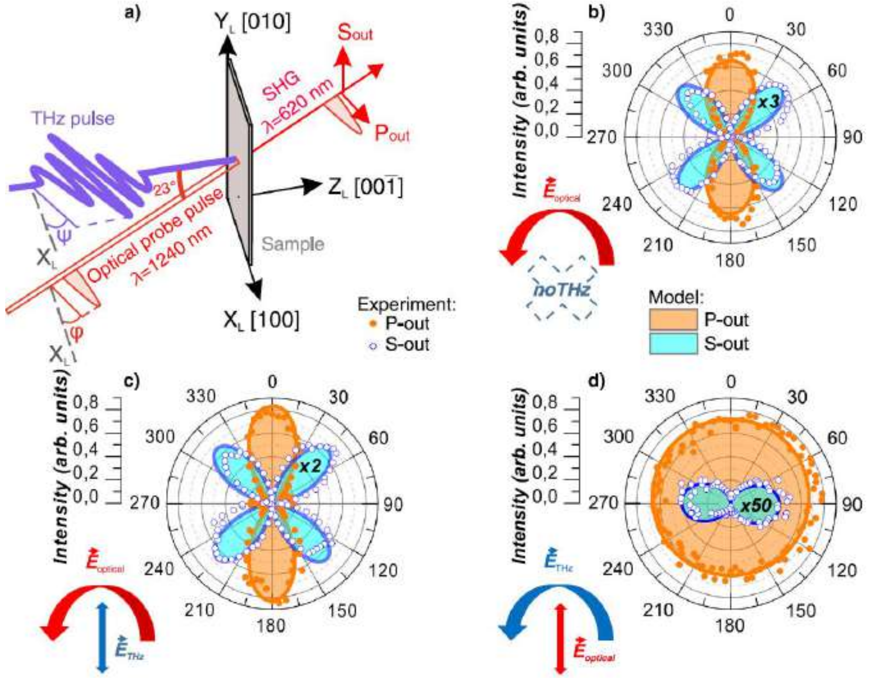


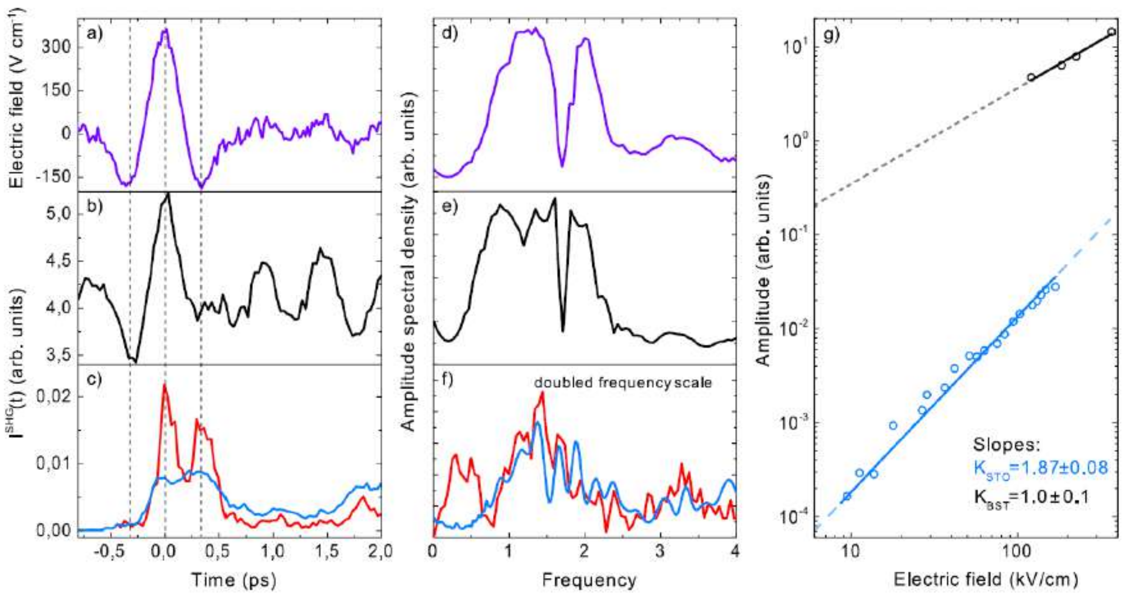
SCIENTIFIC REPORTS

OPEN

THz Electric Field-Induced Second Harmonic Generation in Inorganic Ferroelectric

Kirill A. Grishunin[1], Nikita A. Ilyin[1], Natalia E. Sherstyuk[1], Elena D. Mishina[1], Alexey Kimel[1,2], Vladimir M. Mukhortov[3], Andrey V. Ovchinnikov[4], Oleg V. Chefonov[4] & Mikhail B. Agranat[4]





$$\vec{P}(2\omega) = \vec{P}^{\text{cryst}}(2\omega) + \vec{P}^{\text{TEFISH}}(2\omega) = \hat{\chi}^{(2)}\vec{E}_\omega\vec{E}_\omega + \hat{\chi}^{(3)}\vec{E}_\Omega\vec{E}_\omega\vec{E}_\omega.$$

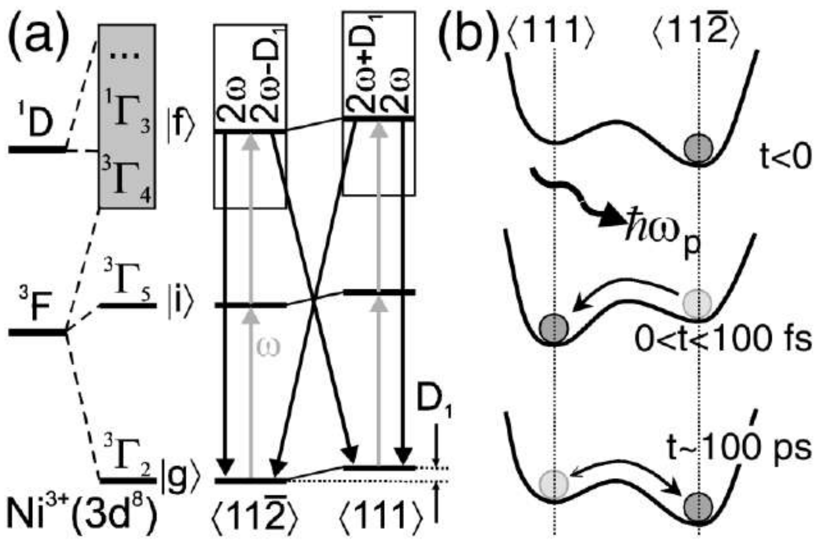
Time-resolved SHG:

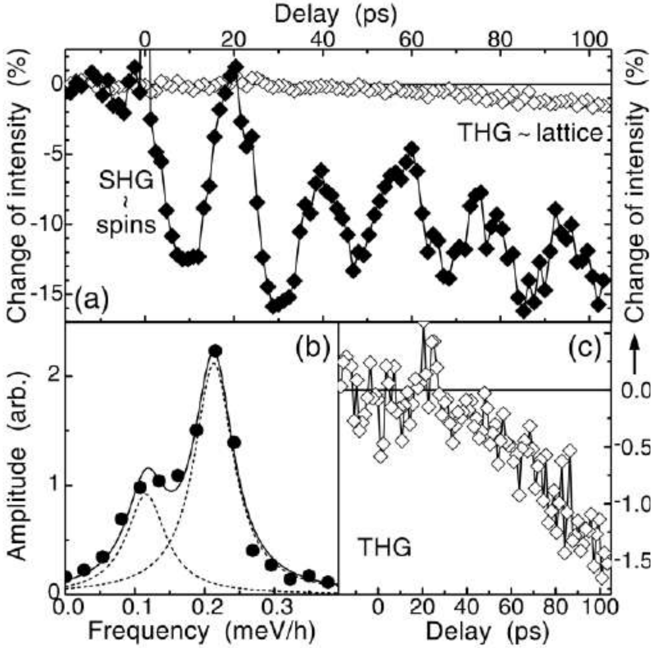
VOLUME 93, NUMBER 11 PHYSICAL REVIEW LETTERS week ending 10 SEPTEMBER 2004

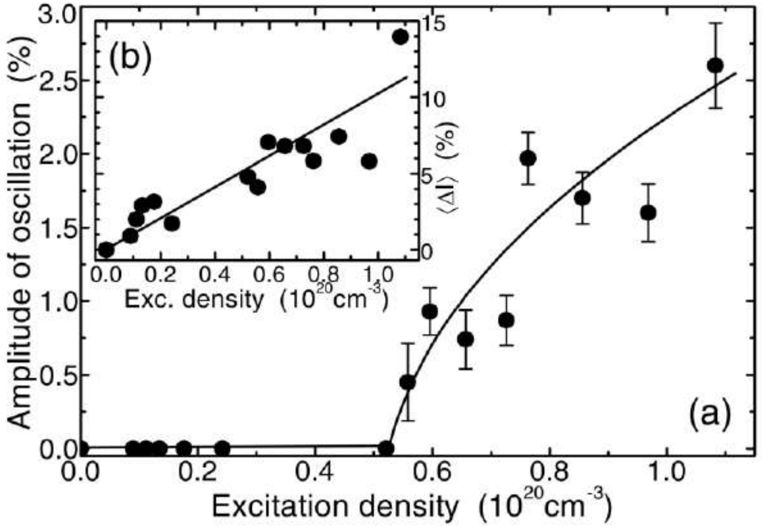
Ultrafast Manipulation of Antiferromagnetism of NiO

N. P. Duong,[1] T. Satoh,[1,2] and M. Fiebig[1,*]
[1]*Max-Born-Institut, Max-Born-Straße 2A, 12489 Berlin, Germany*
[2]*Department of Applied Physics, The University of Tokyo, Tokyo 113-8656, Japan*

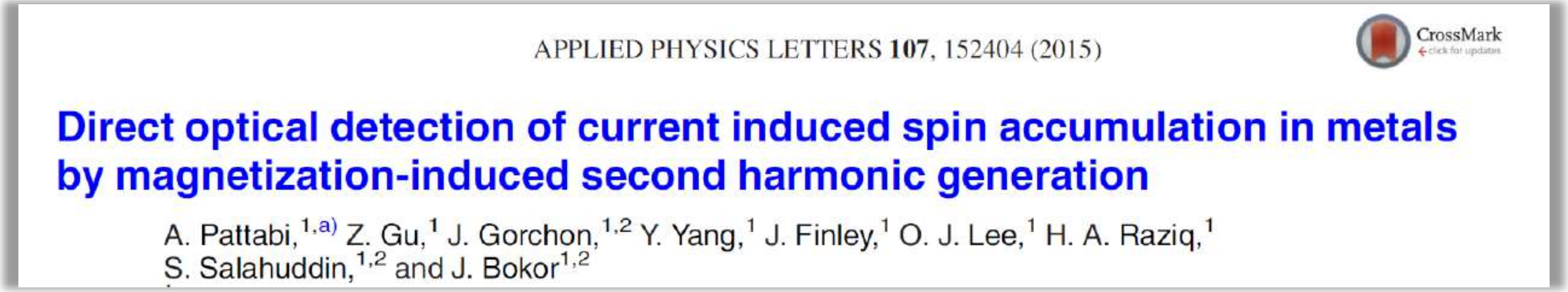
(Received 12 May 2004; published 10 September 2004)







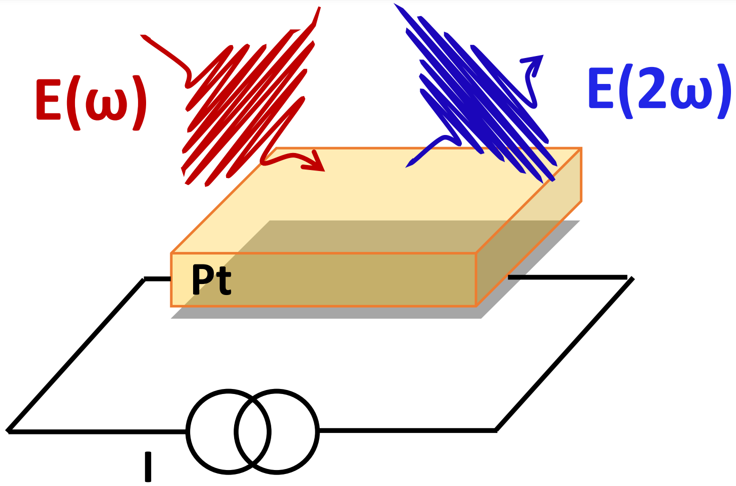
Time-resolved SHG:



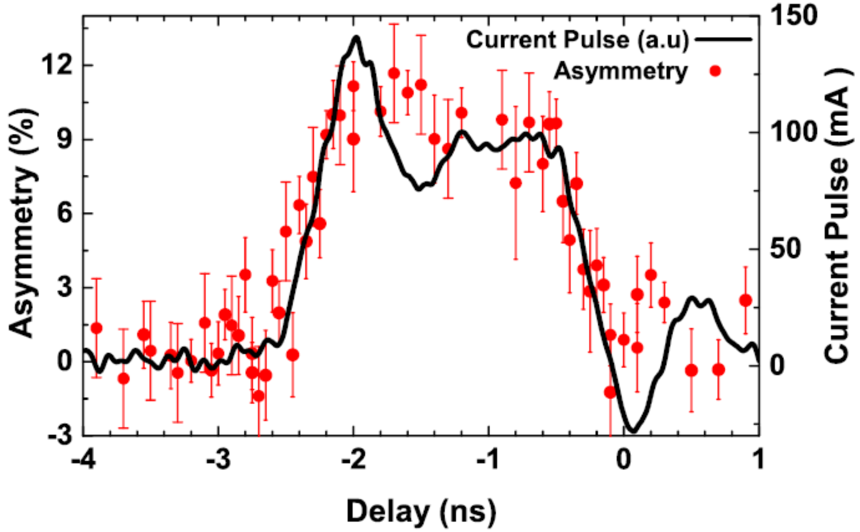
APPLIED PHYSICS LETTERS **107**, 152404 (2015)

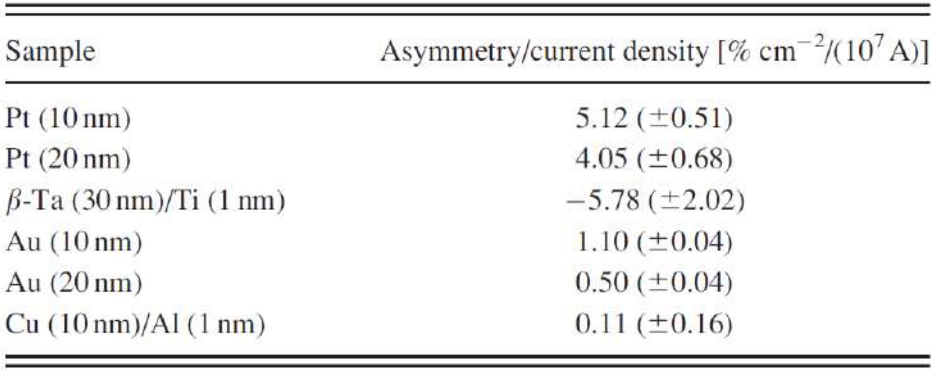
Direct optical detection of current induced spin accumulation in metals by magnetization-induced second harmonic generation

A. Pattabi,[1,a)] Z. Gu,[1] J. Gorchon,[1,2] Y. Yang,[1] J. Finley,[1] O. J. Lee,[1] H. A. Raziq,[1]
S. Salahuddin,[1,2] and J. Bokor[1,2]



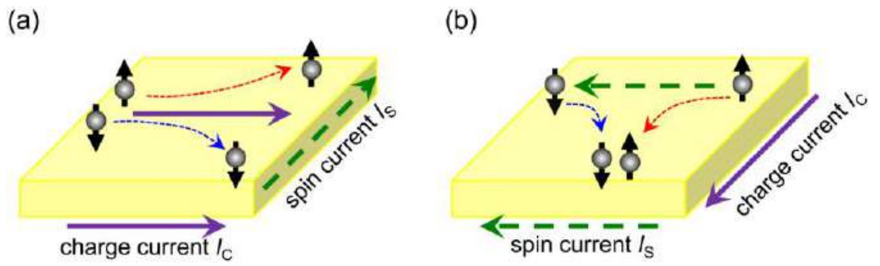
Spin accumulation at interfaces



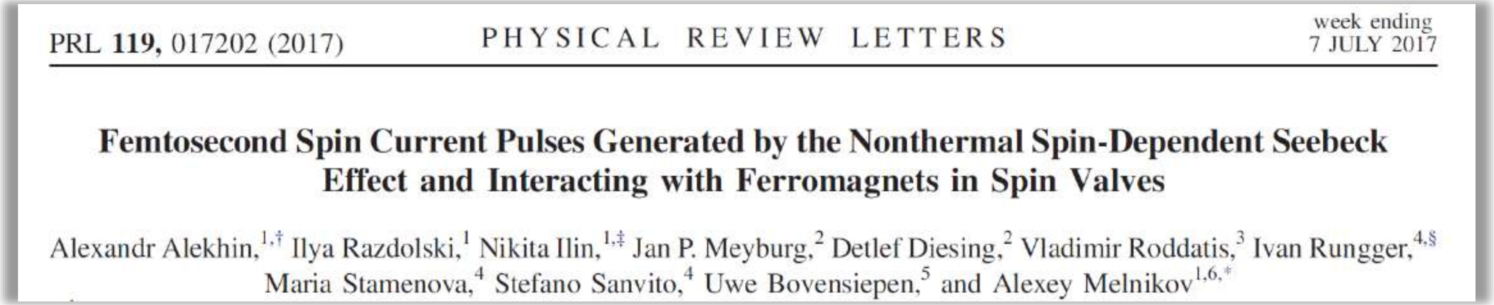


Sample	Asymmetry/current density [% cm^{-2}/(10^7 A)]
Pt (10 nm)	5.12 (±0.51)
Pt (20 nm)	4.05 (±0.68)
β-Ta (30 nm)/Ti (1 nm)	−5.78 (±2.02)
Au (10 nm)	1.10 (±0.04)
Au (20 nm)	0.50 (±0.04)
Cu (10 nm)/Al (1 nm)	0.11 (±0.16)

Spin Hall effect



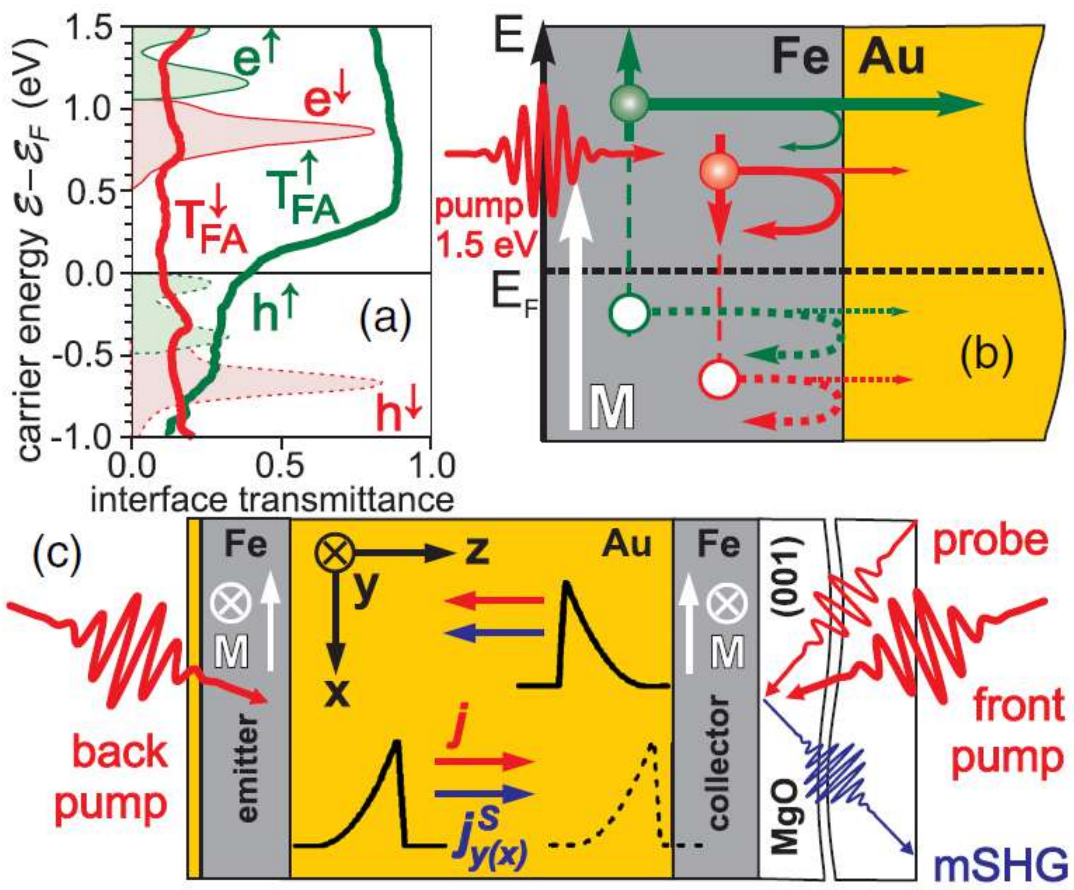
from Niimi et al. *Rep. Prog. Phys.* **78** 124501 (2015)

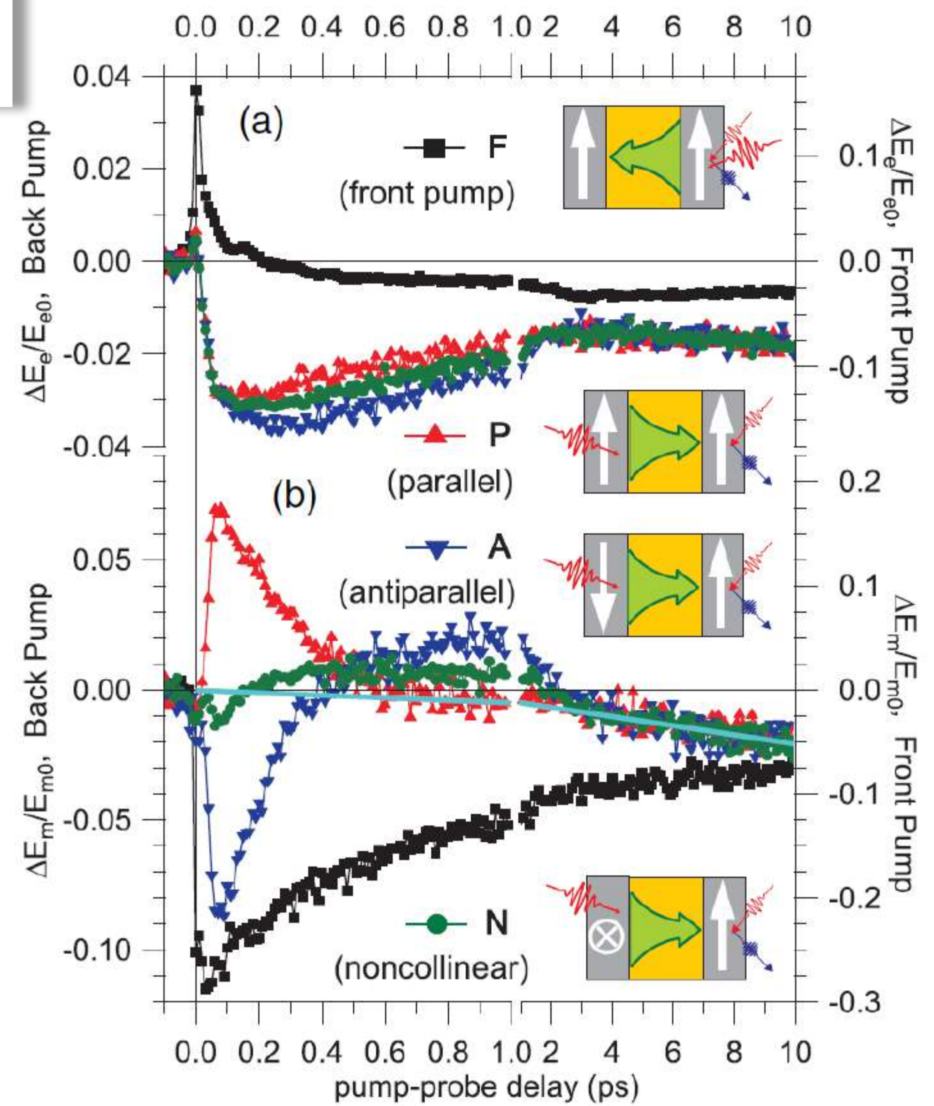


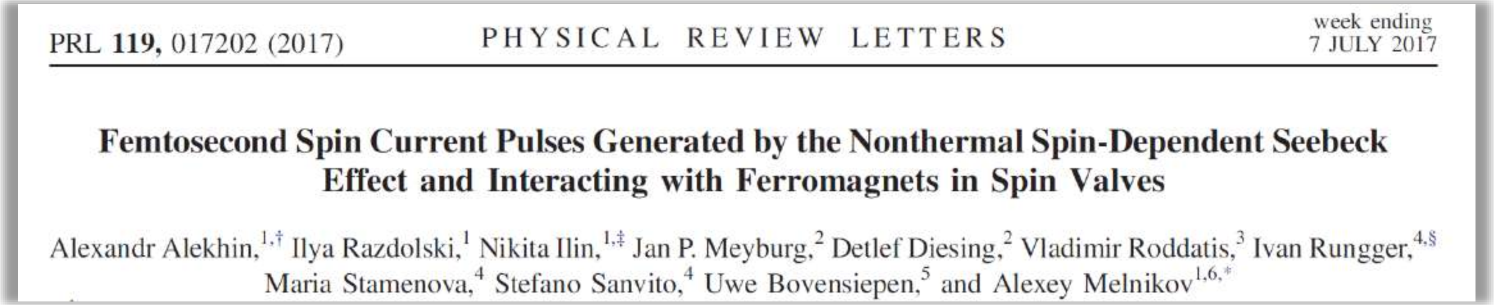
PRL **119,** 017202 (2017) PHYSICAL REVIEW LETTERS week ending 7 JULY 2017

Femtosecond Spin Current Pulses Generated by the Nonthermal Spin-Dependent Seebeck Effect and Interacting with Ferromagnets in Spin Valves

Alexandr Alekhin,[1,†] Ilya Razdolski,[1] Nikita Ilin,[1,‡] Jan P. Meyburg,[2] Detlef Diesing,[2] Vladimir Roddatis,[3] Ivan Rungger,[4,§] Maria Stamenova,[4] Stefano Sanvito,[4] Uwe Bovensiepen,[5] and Alexey Melnikov[1,6,*]







PRL **119,** 017202 (2017) PHYSICAL REVIEW LETTERS week ending 7 JULY 2017

Femtosecond Spin Current Pulses Generated by the Nonthermal Spin-Dependent Seebeck Effect and Interacting with Ferromagnets in Spin Valves

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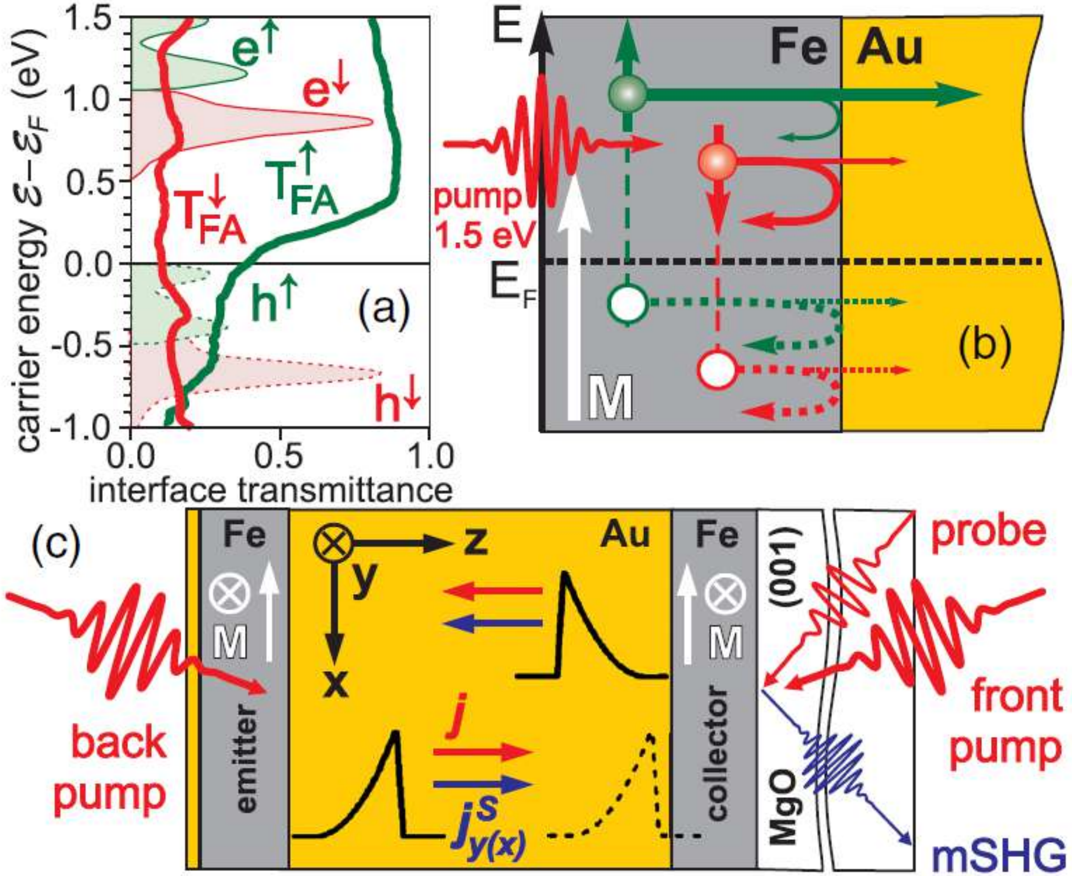
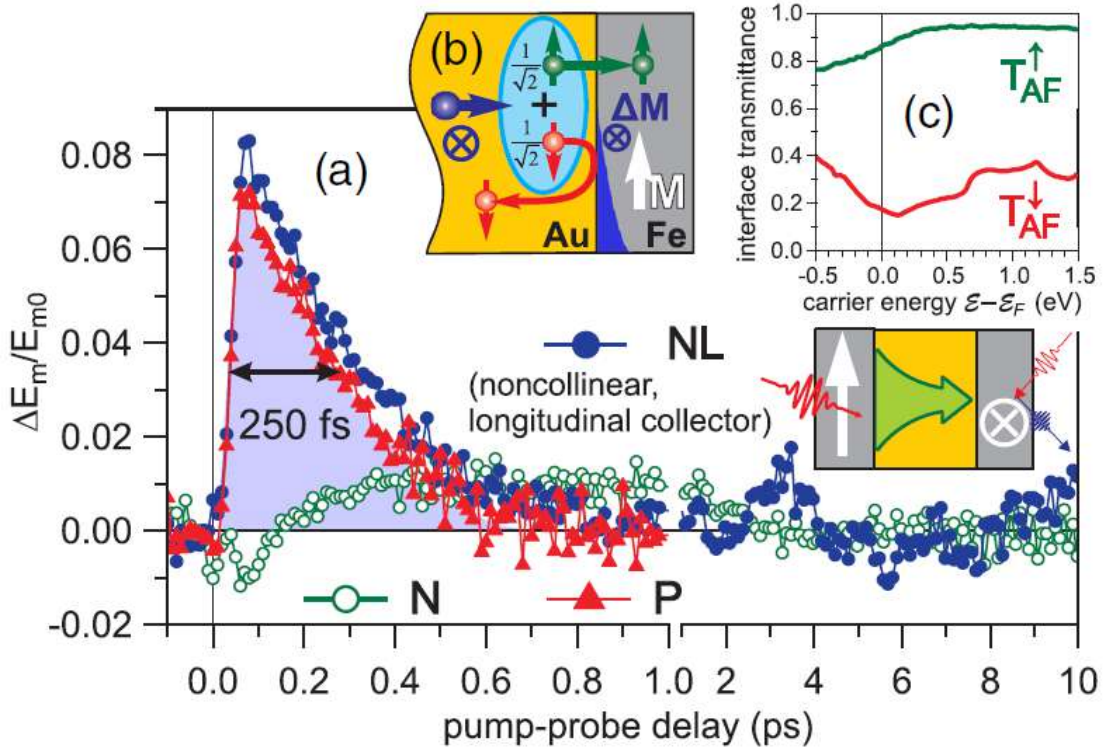
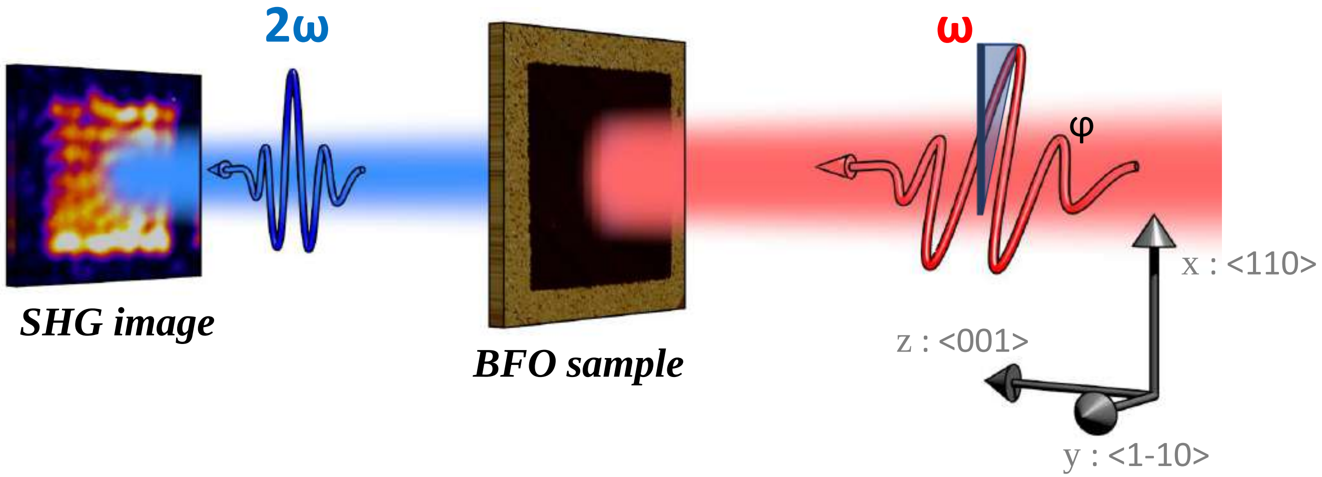




SHG: Conclusions and Summary



- 2nd order Non-linear optical process
- Powerful tool to access and investigate complex ferroic distributions (microscopy, spectroscopy, polarimetry…)
- Despite its macroscopic signature, access to microscopic detail of the magnetic and multiferroic strucutures
- Surface, interface sensitivity
- To fully master SHG, symmetry argumentation and group theory are required
- Ultrafast time-resolved capabilities