# Noncollinear Frequency Conversion

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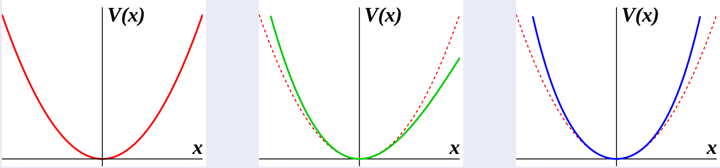
Fachbereich Physik

UNIVERSITÄT OSNABRÜCK

Ring Lecture PhD Program — Summer 2004

## Outline

# Part I: Introduction – Nonlinear Optics




1. Linear and Nonlinear Response
   - Linearity
   - Nonlinearity
   - Light and Matter
2. Nonlinear Optical Susceptibility
   - Linear Polarization
   - Nonlinear Polarization
   - Crystal Symmetry

## Linear Response

- Our basic experience in physics and life:

  **We are living in a linear world !**

- Mechanics: Doubled Force $\Rightarrow$ Doubled Impact
- Electricity: Doubled Voltage $\Rightarrow$ Doubled Current
- Measurements rely on linearity:

  Length, Weight, Intensity, ...

- Prices usually add up linearly:

  Two apples are twice the price of one.

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"Physics would be dull and life most unfulfilling if all physical phenomena around us were linear. Fortunately, we are living in a **nonlinear** world. While linearization beautifies physics, nonlinearity provides excitement in physics."

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- What about 100 apples ?
- You get discount.
- That's sort of **nonlinear response** by the salesman.

- And in physics ?

- Hooke's law is only valid in a limited range.
- Audio signals get distorted at high intensities.
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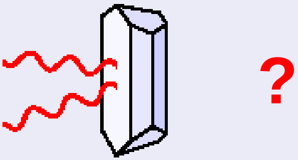

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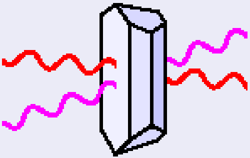

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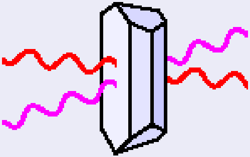

**No Interaction of Light Beams**

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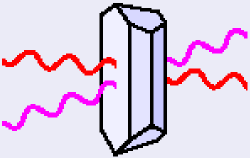

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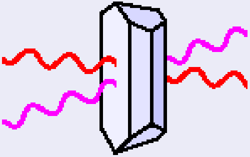

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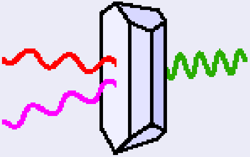

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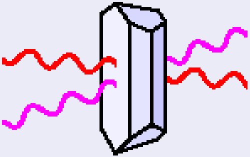

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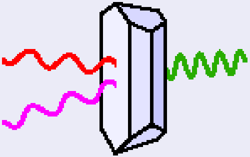

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## **Linear Polarization** — we start with the simple case

Electric Field $\overset{\text{Susceptibility}}{\Longrightarrow}$ Polarization

$$\mathbf{P}(\mathbf{r},t) = \epsilon_0 \int_{-\infty}^{\infty} \chi^{(1)}(\mathbf{r}-\mathbf{r}',t-t') \cdot \mathbf{E}(\mathbf{r}',t') d\mathbf{r}' dt'$$

Linear susceptibility $\chi^{(1)}$ usually strictly local ($\delta(\mathbf{r}-\mathbf{r}')$).

Plane waves $\mathbf{E}(\mathbf{k},\omega) = \boldsymbol{E}(\mathbf{k},\omega)\exp(i\mathbf{k}\mathbf{r} - i\omega t)$,

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## Nonlinear Polarization

**P** to be expanded into a power series of **E**

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$$+ \epsilon_0 \int_{-\infty}^{\infty} \chi^{(2)}(\mathbf{r}-\mathbf{r}_1, t-t_1; \mathbf{r}-\mathbf{r}_2, t-t_2)$$
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$$+ \epsilon_0 \int_{-\infty}^{\infty} \chi^{(3)}(\mathbf{r}-\mathbf{r}_1, t-t_1; \mathbf{r}-\mathbf{r}_2, t-t_2; \mathbf{r}-\mathbf{r}_3, t-t_3) * \mathbf{E}(\mathbf{r}_1, t_1)\mathbf{E}(\mathbf{r}_2, t_2)\mathbf{E}(\mathbf{r}_3, t_3) d\mathbf{r}_1 dt_1 d\mathbf{r}_2 dt_2 d\mathbf{r}_3 dt_3$$

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## Nonlinear Polarization – Fourier Transform

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## Nonlinear Polarization – Fourier Transform

Electric field $\mathbf{E}(\mathbf{r}, t) = \sum_i \mathbf{E}(\mathbf{k}_i, \omega_i)$ ,

Polarization $\mathbf{P}(\mathbf{k}, \omega) = \mathbf{P}^{(1)}(\mathbf{k}, \omega) + \mathbf{P}^{(2)}(\mathbf{k}, \omega) + \mathbf{P}^{(3)}(\mathbf{k}, \omega) \ldots$

$$\mathbf{P}^{(1)}(\mathbf{k}, \omega) = \epsilon_0 \chi^{(1)}(\mathbf{k}, \omega) \cdot \mathbf{E}(\mathbf{k}, \omega) ,$$

$$\mathbf{P}^{(2)}(\mathbf{k}, \omega) = \epsilon_0 \chi^{(2)}(\mathbf{k} = \mathbf{k}_i + \mathbf{k}_j, \omega = \omega_i + \omega_j) * \mathbf{E}(\mathbf{k}_i, \omega_i)\mathbf{E}(\mathbf{k}_j, \omega_j) ,$$

$$\mathbf{P}^{(3)}(\mathbf{k}, \omega) = \epsilon_0 \chi^{(3)}(\mathbf{k} = \mathbf{k}_i + \mathbf{k}_j + \mathbf{k}_k, \omega = \omega_i + \omega_j + \omega_k)$$

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# Crystal Symmetry

- $\chi^{(n)}$ are polar tensors describing certain material properties.
- These tensors have to be invariant with regard to all symmetry operations of the crystal (point) symmetry.
- From symmetry one thus can deduce which tensor elements are equal and which are zero.
- In crystals of centric symmetry all tensors of odd rank vanish ($\chi^{(2)}$, e. g., is a third-rank tensor).

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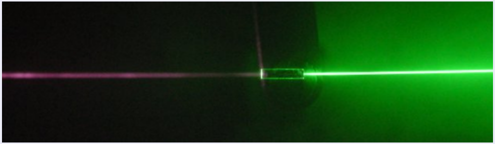
## Outline

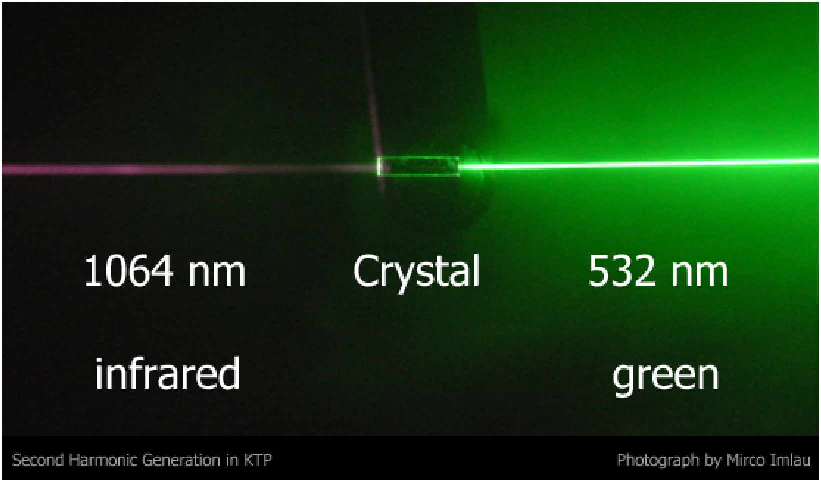
Part I: Introduction – Nonlinear Optics

Part II: Harmonic Generation

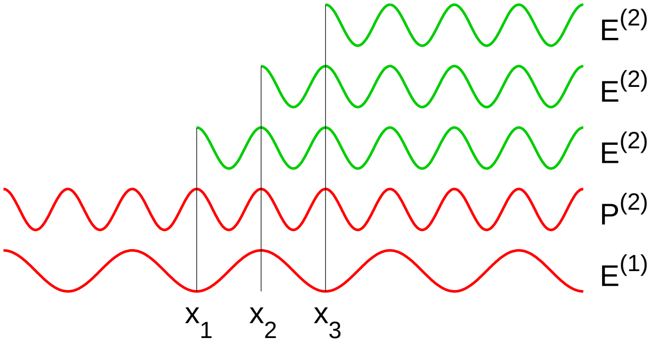
Part III: Noncollinear Harmonic Generation

## Part II: Harmonic Generation

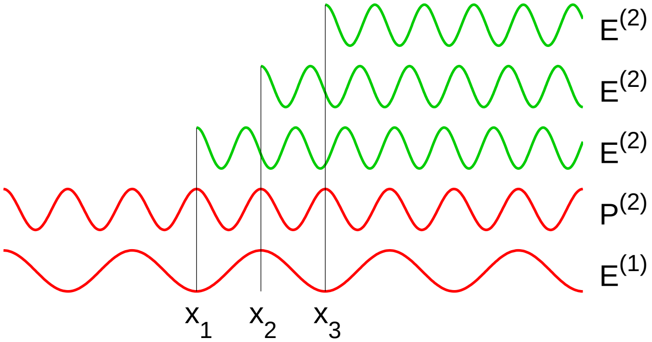



1064 nm
Crystal
532 nm
infrared
green
Second Harmonic Generation in KTP
Photograph by Mirco Imlau

## Idealized



## Reality



## Spatial Dependence of Field and Polarization

Fundamental Field

$$E^{(1)}(x) \quad = \quad E^{(1)}(0) \cdot e^{-ik_1 x}$$

Second Harmonic Polarization

$$P^{(2)}(x) \quad = \quad \chi E^{(1)}(x) E^{(1)}(x) = \chi E^{(1)}(0) E^{(1)}(0) \cdot e^{-i2k_1 x}$$

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## Second Harmonic Field at $x'$

### Harmonic Field at Position $x$

$$E^{(2)}(x) = K \cdot E^{(1)}(0)E^{(1)}(0) \cdot e^{-i2k_1x}$$

$E^{(2)}$ travels through the material with a velocity characteristic for the frequency $\omega_2 = 2\omega_1$ and wave vector $k_2$

$$\begin{aligned} E^{(2)}(x') &= E^{(2)}(x) \cdot e^{-ik_2(x'-x)} \\ &= K \cdot E^{(1)}(0)E^{(1)}(0) \cdot e^{-ik_2x'} e^{-i(2k_1-k_2)x} \end{aligned}$$

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E^{(2)}_{\text{total}}(x') &= K \cdot E^{(1)}_{(0)} E^{(1)}_{(0)} \cdot e^{-ik_2 x'} \int_0^L e^{-i(2k_1 - k_2)x} dx \\
&= K \cdot E^{(1)}_{(0)} E^{(1)}_{(0)} \cdot e^{-ik_2 x'} \frac{1}{i\Delta k} \left[ e^{i\Delta k L} - 1 \right] \\
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with

$$\Delta k = k_2 - 2k_1 = \frac{2\pi}{\lambda_2} n(\omega_2) - 2\frac{2\pi}{\lambda_1} n(\omega_1) = \frac{4\pi}{\lambda_1}(n(\omega_2) - n(\omega_1))$$

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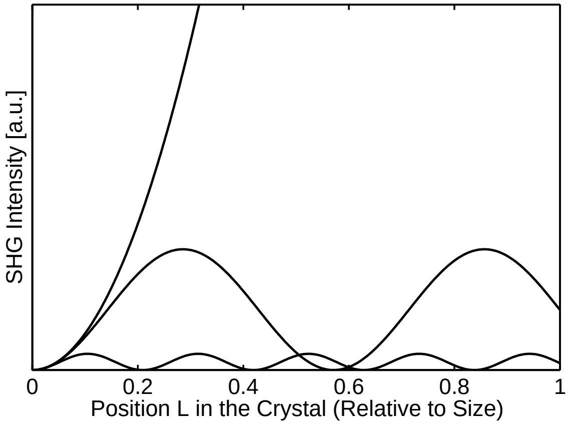

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$$I^{(2)} = C \cdot d_{\text{eff}}^2 \cdot I^{(1)\,2} \cdot \frac{\sin^2(\Delta k\, L/2)}{(\Delta k/2)^2}$$ maximized for $\Delta k = 0$

Normal dispersion $n(\omega_2) > n(\omega_1)$ $\Delta k > 0$

## Birefringent Crystals

Different refractive indices for different polarization directions

Property connected with crystal symmetry

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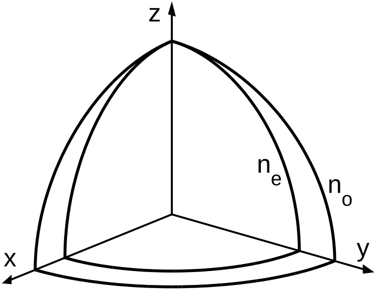
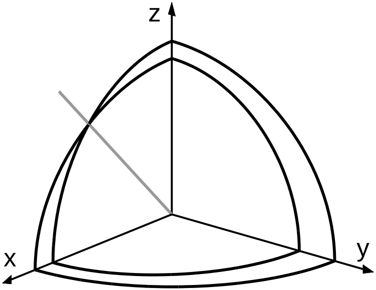
## Birefringence – Index Surfaces

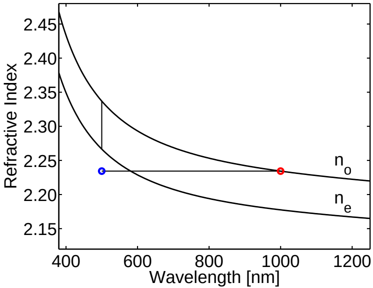



Uniaxial

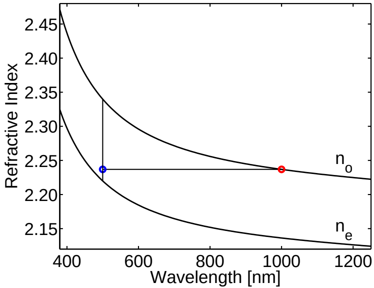



Biaxial

## Birefringence – Phase Matching



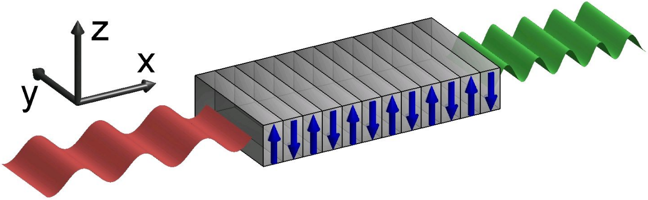

Too small




Sufficient

## Quasi Phase Matching

Example: Periodically Poled Lithium Niobate




$$k_2 = k_1 + k_1' + K$$

# Quasi Phase Matching

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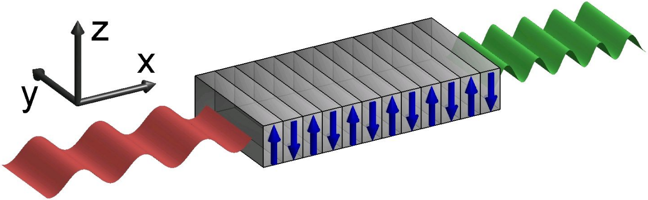


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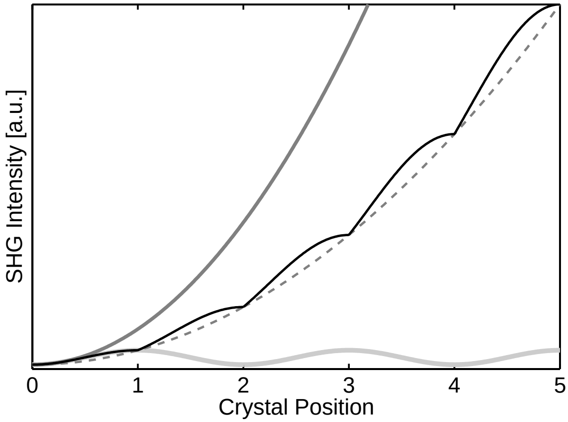
## QPM: Second Harmonic Intensity, same Tensor Element




$$d \implies d \cdot 2/\pi$$

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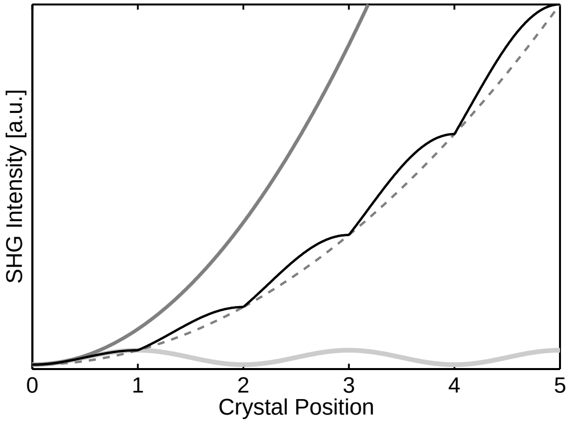


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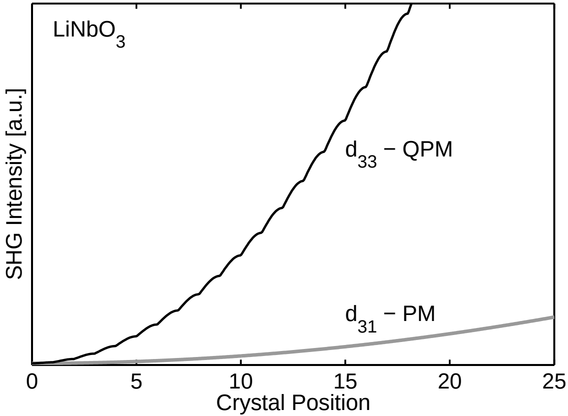
# QPM: Lithium Niobate, $d_{33}$ instead of $d_{31}$




$d_{31} = 4.3\,\text{pm/V} \quad d_{33} = 27\,\text{pm/V} \implies d_{\text{eff}} = 17\,\text{pm/V}$

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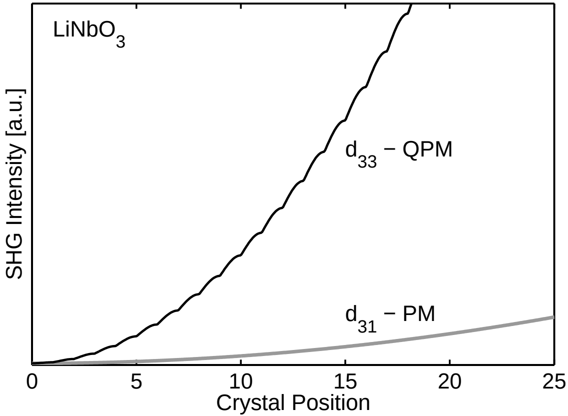


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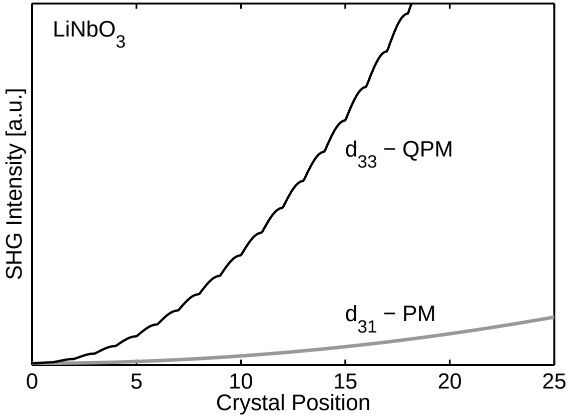


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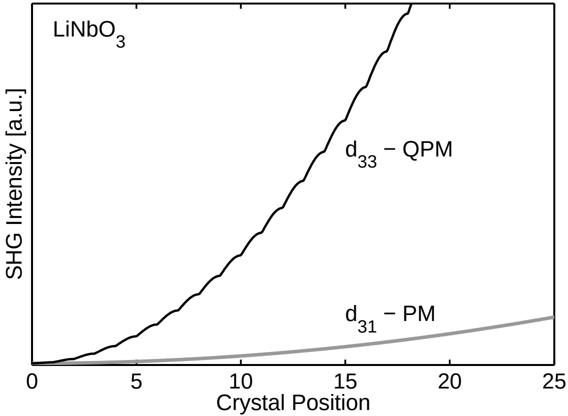


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# High-Order Harmonic Generation

- Atoms in High Laser Fields
- Centric Symmetry $\Longrightarrow$ Odd Harmonics
- Refractive Index near to 1
- Phase Matching by Gas Mixing
- *Below* and *Above* Resonance

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- Centric Symmetry $\Longrightarrow$ Odd Harmonics
- Refractive Index near to 1
- Phase Matching by Gas Mixing
- *Below* and *Above* Resonance

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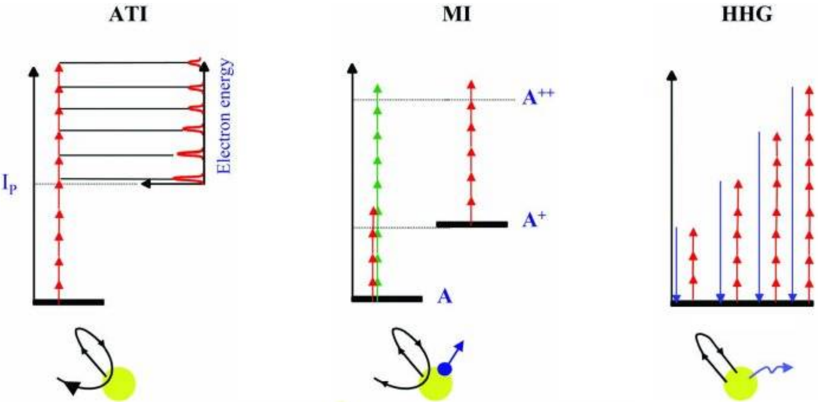
## Processes in High Laser Fields



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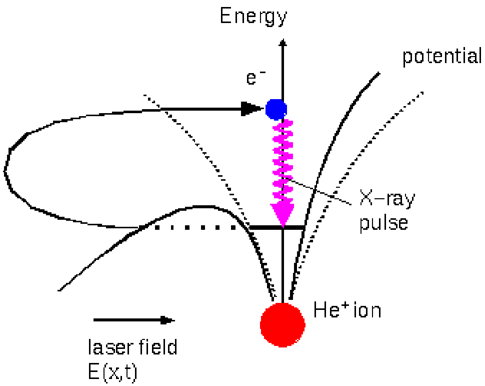
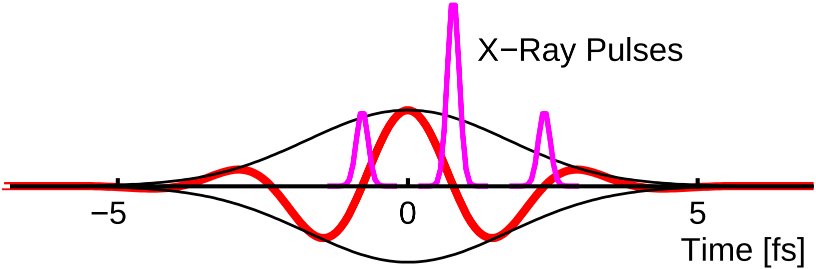


## HHG: Timing



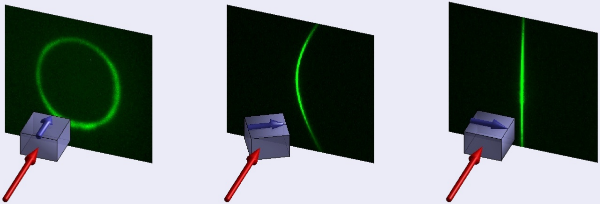
## Outline

Part I: Introduction – Nonlinear Optics

Part II: Harmonic Generation

Part III: Noncollinear Harmonic Generation

# Part III: Noncollinear Harmonic Generation



5. Noncollinear Frequency Doubling
   - Induced Noncollinear Frequency Doubling
   - Spontaneous Noncollinear Frequency Doubling
6. Conical Harmonic Generation
7. Domain-Induced Noncollinear Second-Harmonic Generation
   - Experiment
   - Model
   - Cylindrically Polarized Light

## Collinear and Noncollinear Harmonic Generation

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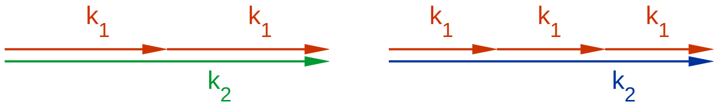
- Collinear Case: $\sum \boldsymbol{k_i} = \mathbf{0}$



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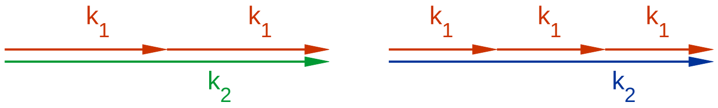
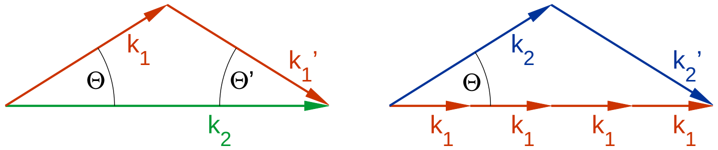
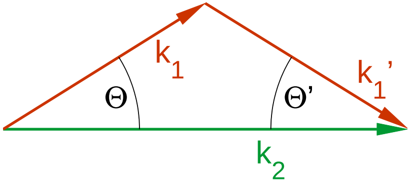



- Noncollinear Case: $\sum \boldsymbol{k_i} \neq \mathbf{0}$ yet $\sum \overrightarrow{\boldsymbol{k_i}} = \mathbf{0}$



# Induced Noncollinear Frequency Doubling: Principle




$$|\mathbf{k}_2| = |\mathbf{k}_1|\cos\Theta + |\mathbf{k}_1'|\cos\Theta' \quad \text{and} \quad |\mathbf{k}| = \frac{\omega}{c} n_p(\omega, \mathbf{k})$$

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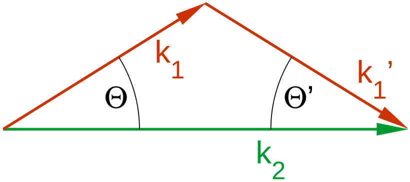


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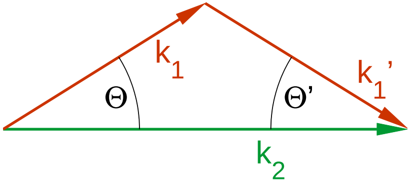


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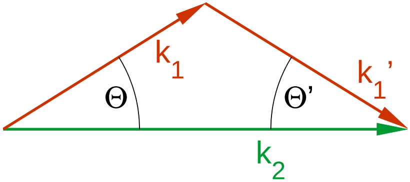


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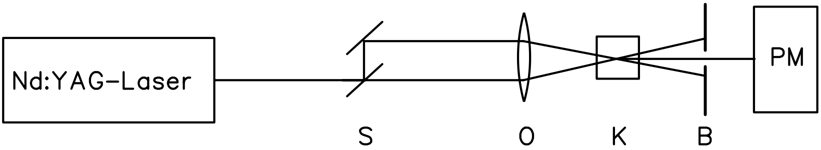


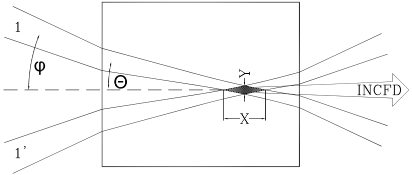

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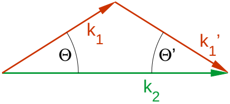


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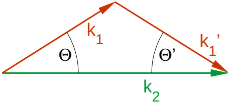


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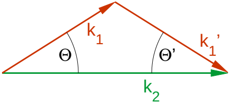


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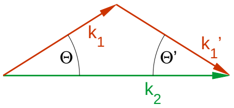


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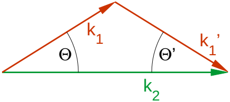


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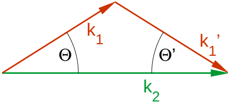


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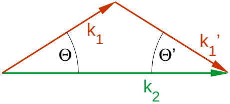


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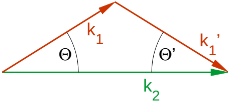


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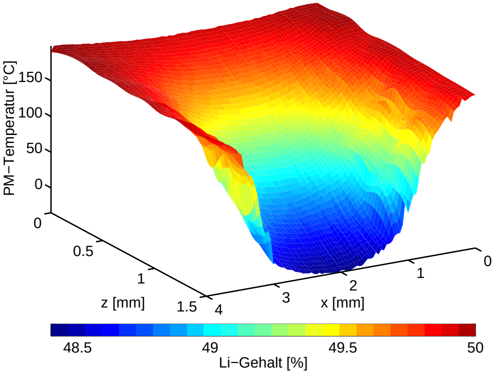
# Induced Noncollinear Frequency Doubling: Results 1

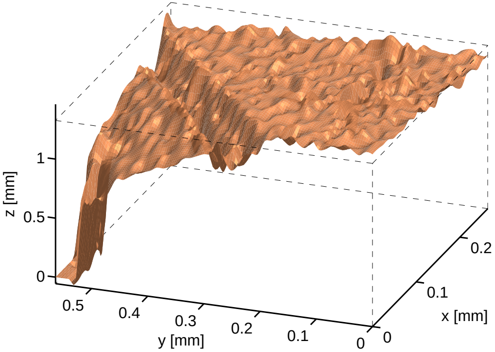
Vapor Transport Equilibration (VTE) on Lithium Niobate



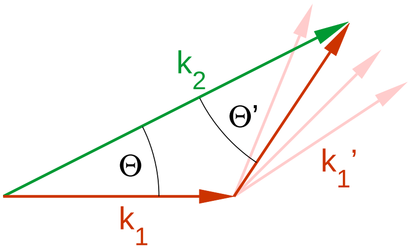

A. Reichert, K. Betzler: *Induced noncolinear frequency doubling: A new characterization technique for electrooptic crystals*. J. Appl. Phys. **79**, 2209–2212 (1996).

# Induced Noncollinear Frequency Doubling: Results 2

Domain Boundaries in Potassium Niobate



## Spontaneous Noncollinear Frequency Doubling: Principle

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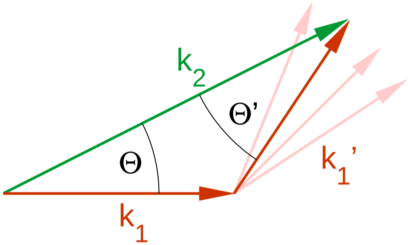


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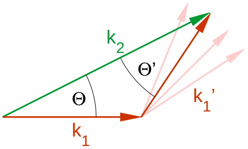


- Again: Refractive Indices Determine Geometry
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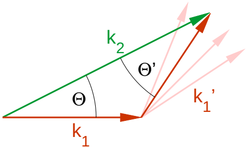


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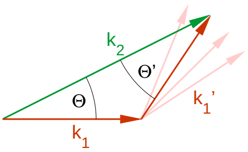


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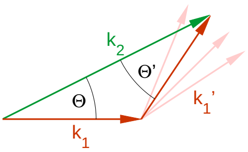


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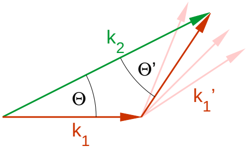


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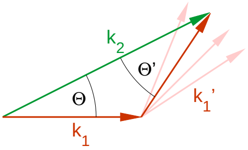


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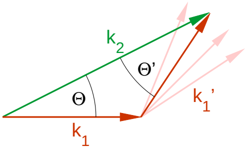


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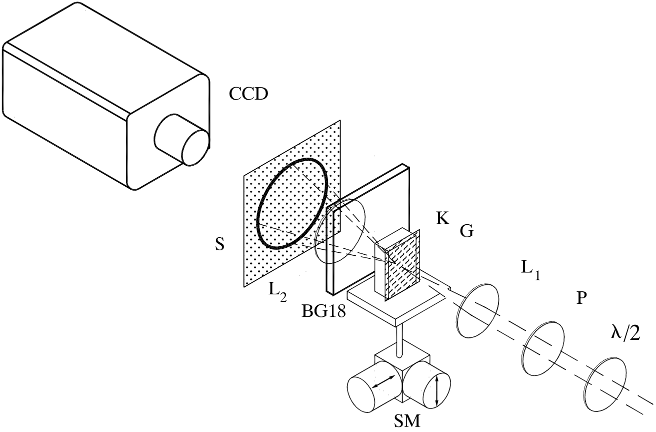
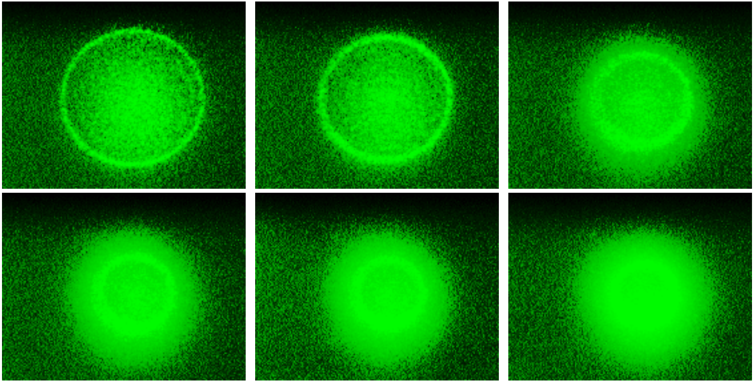
# Spontaneous Noncollinear Frequency Doubling: Experiment



# Spontaneous Noncollinear Frequency Doubling: Evaluation



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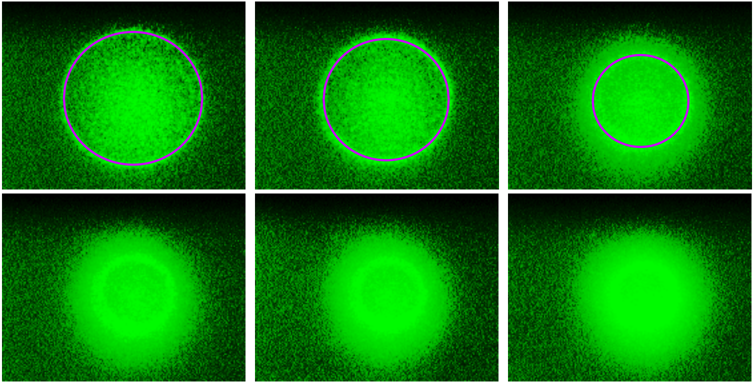

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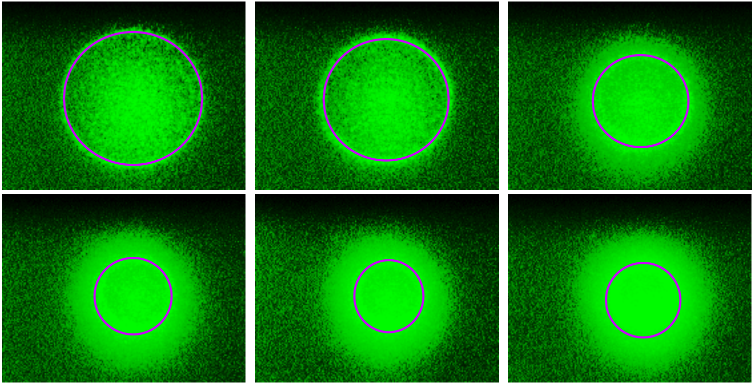
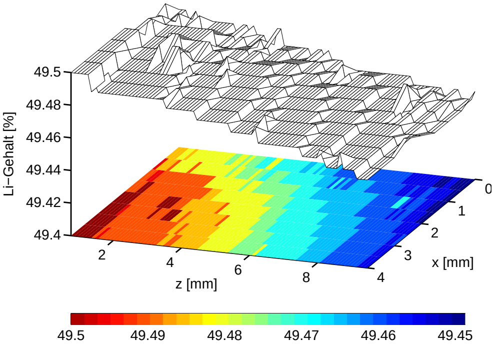


K.-U. Kasemir, K. Betzler: *Detecting Ellipses of Limited Eccentricity in Images with High Noise Levels.* Image and Vision Computing **21**, 221 (2003).

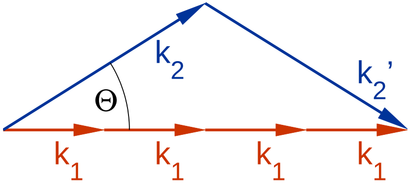
# Spontaneous Noncollinear Frequency Doubling: Results

## Crystal Growth of Lithium Niobate – Homogeneity




K.-U. Kasemir, K. Betzler: *Characterization of photorefractive materials by spontaneous noncolinear frequency doubling.* Applied Physics **B 68**, 763 (1999).

# Conical Harmonic Generation: Principle

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$\omega_2 = 2\omega_1$ , general case: $\omega_m = m\omega_1$

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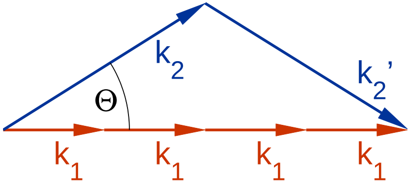


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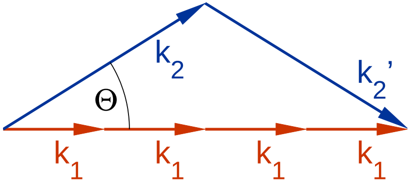


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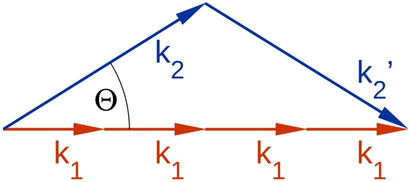


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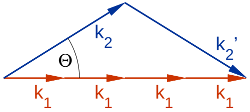


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- *Parametric* Harmonic Generation
- Odd-Order Process $\Longrightarrow$ Always Allowed
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- Compatible with *Normal* Dispersion

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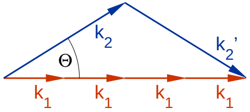


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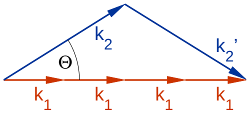


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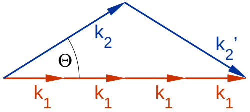


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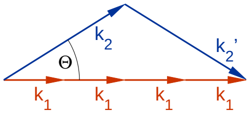
## Conical Harmonic Generation




- General Case: $\mathbf{k}_m = 2m \cdot \mathbf{k}_1 - \mathbf{k}'_m$
- $\omega_m = m \cdot \omega_1$
- *Parametric* Harmonic Generation
- Odd-Order Process $\Longrightarrow$ Always Allowed
- $\cos\Theta = n(\omega_1)/n(\omega_m)$
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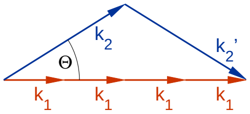


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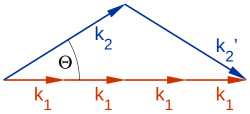


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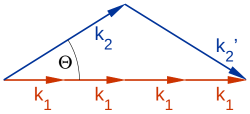


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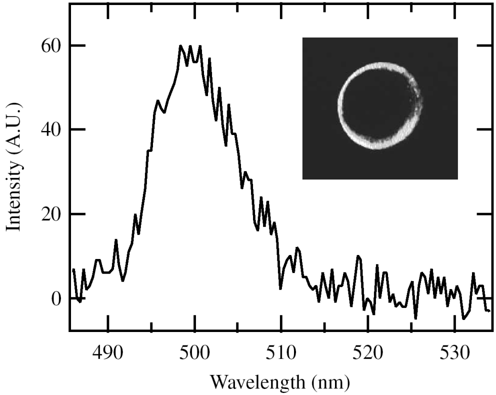
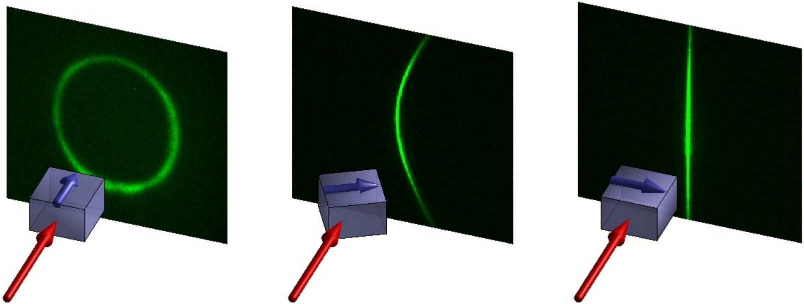
# Conical Harmonic Generation: Result




K. D. Moll, D. Homoelle, Alexander L. Gaeta, Robert W. Boyd: *Conical Harmonic Generation in Isotropic Materials*. Phys. Rev. Lett. **88**, 153901 (2002).

## Domain-Induced Noncollinear Second-Harmonic Generation

Strontium Barium Niobate (SBN)



Arthur R. Tunyagi, Michael Ulex, Klaus Betzler: *Non-collinear optical frequency doubling in Strontium Barium Niobate*. Physical Review Letters **90**, 243901 (2003)

## Domain-Induced Noncollinear Second-Harmonic Generation

To be explained by a Model:

- Strontium Barium Niobate – Low Birefringence
- Weak in Poled, Strong in Unpoled Samples
- Circle – Ellipse – Hyperbola – Straight Line
- Ring is Radially Polarized (Cylindric Polarization)
- $$d_{ij} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{15} & 0 \\ 0 & 0 & 0 & d_{24} & 0 & 0 \\ d_{31} & d_{32} & d_{33} & 0 & 0 & 0 \end{pmatrix}$$

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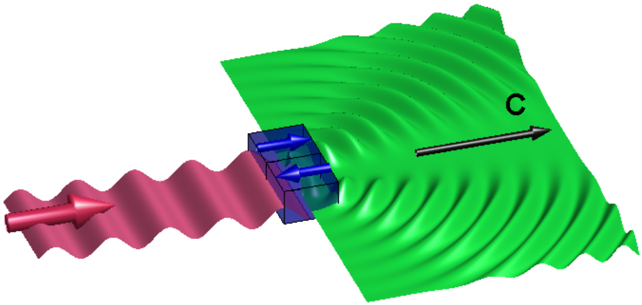
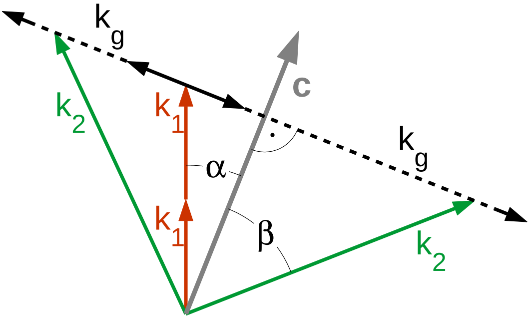


## Domain-Induced Noncollinear Second-Harmonic Generation

### Momentum Diagram:




$$\mathbf{k}_2 = 2\mathbf{k}_1 + \mathbf{k}_g$$

$\mathbf{k}_g \perp \mathbf{c}$ : $\quad 2\mathbf{k}_1 \cos\alpha = \mathbf{k}_2 \cos\beta, \quad n_1(\alpha)\cos\alpha = n_2(\beta)\cos\beta$

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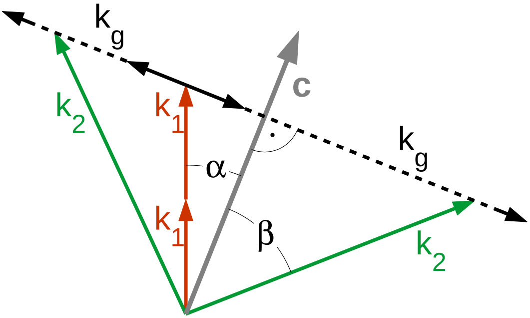


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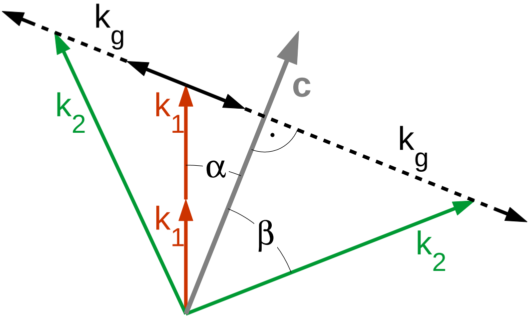


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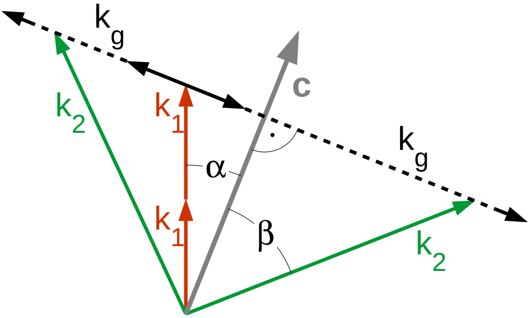


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- SBN: Needle-like Domains aligned in c-Direction
- $\mathbf{k}_g \perp \mathbf{c} \implies \cos\beta = \text{const.} \implies$ Circular Cone
- Cone Sections on Screen:
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- Intensity Distribution reflects Density Distribution of $\mathbf{k}_g$
- k–Space Spectroscopy of Domains

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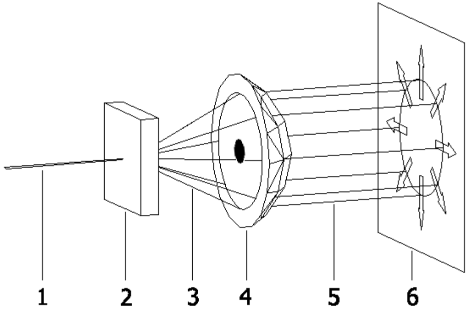
# Cylindrically Polarized Light




**1** Laser
**2** SBN Crystal
**3** Noncollinear SHG Light
**4** Collimation Optic
**5** Cylindric Parallel Beam
**6** with radial Polarisation