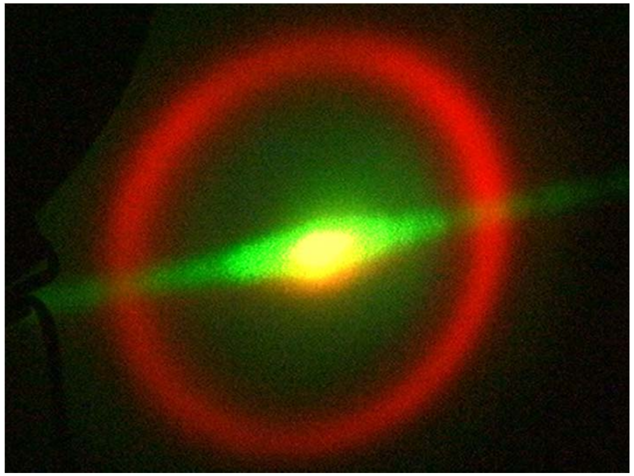
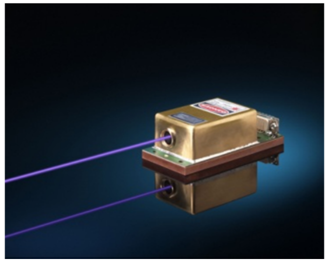
Nonlinear optics

Fredrik Laurell



Laser Physics Group
Department of Applied Physics, KTH

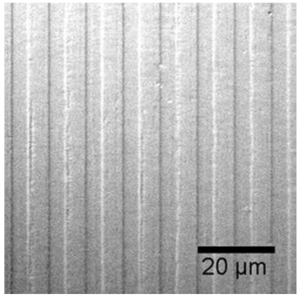
Our research profile



Lasers

Nonlinear optics

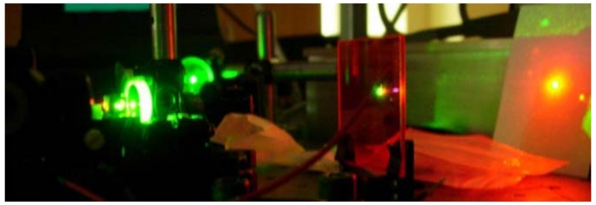
Optical materials and novel components



Investigate fundamental optical
and material physics problems

Build efficient diode-pumped lasers and light sources

Use them in applications



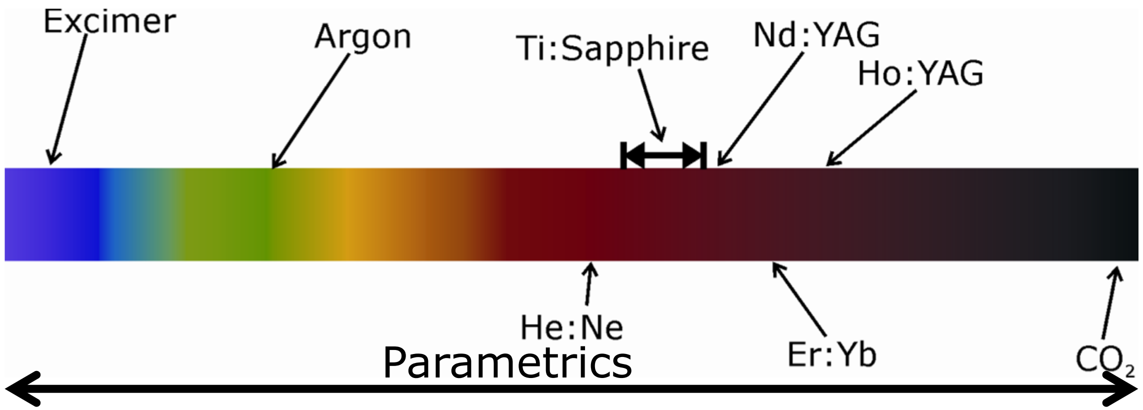
Start-ups and industrial collaboration

Outline

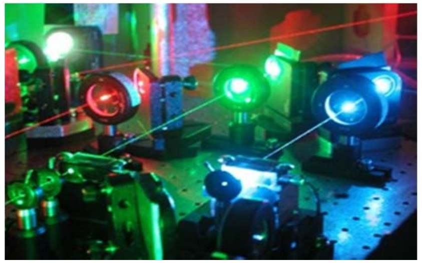
- Introduction to nonlinear optics
- Wave treatment
- Second order effects
- Phasematching and quasi-phase-matching
- Second harmonic generation, visible generation
- Material limitations and damage
- Applications
- Summary

Why nonlinear optics?

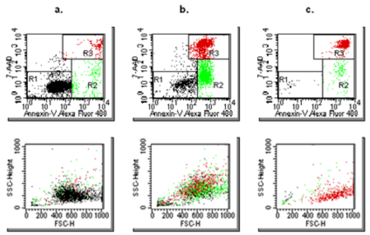
- Second order effects gives a natural extension of lasers
  - Reach the UV and the mid-IR + THz
  - Facilitates tuning
  - Many applications
- Efficient amplifiers – low thermal load and high gain
- Third order nonlinear phenomena
  - Kerr lensing, Self-phase modulation
  - Cascading effects



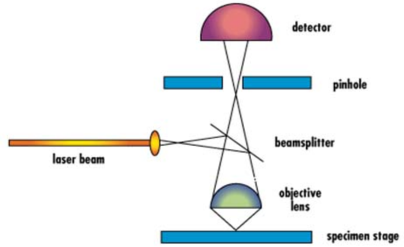
Motivation - upconversion



Laser display

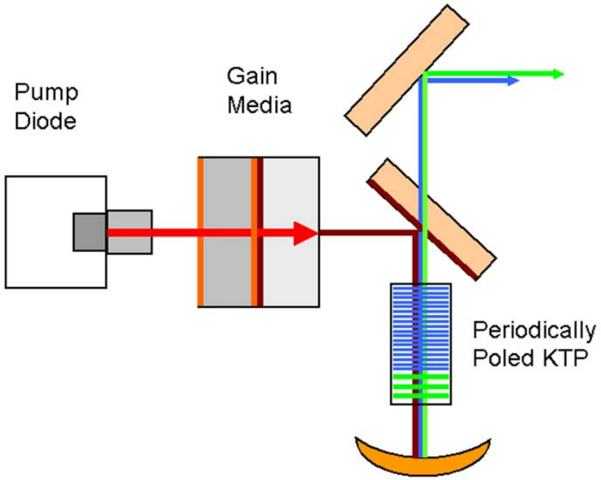


Bio-analysis



Confocal Microscopy

- Needs for visible and UV lasers
- Technology
- Material issues



Wafer inspection

Printing

The birth of nonlinear optics

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

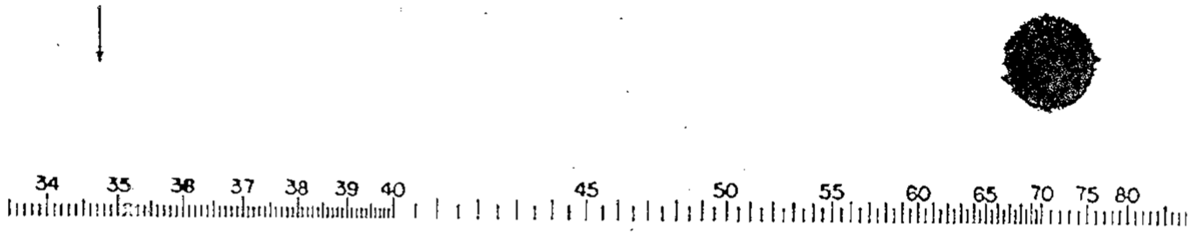
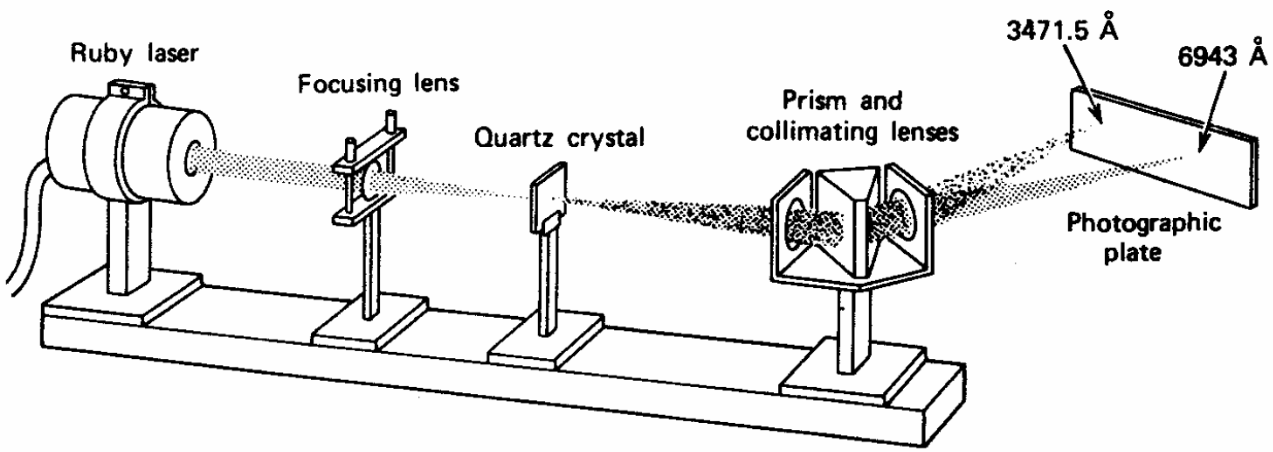


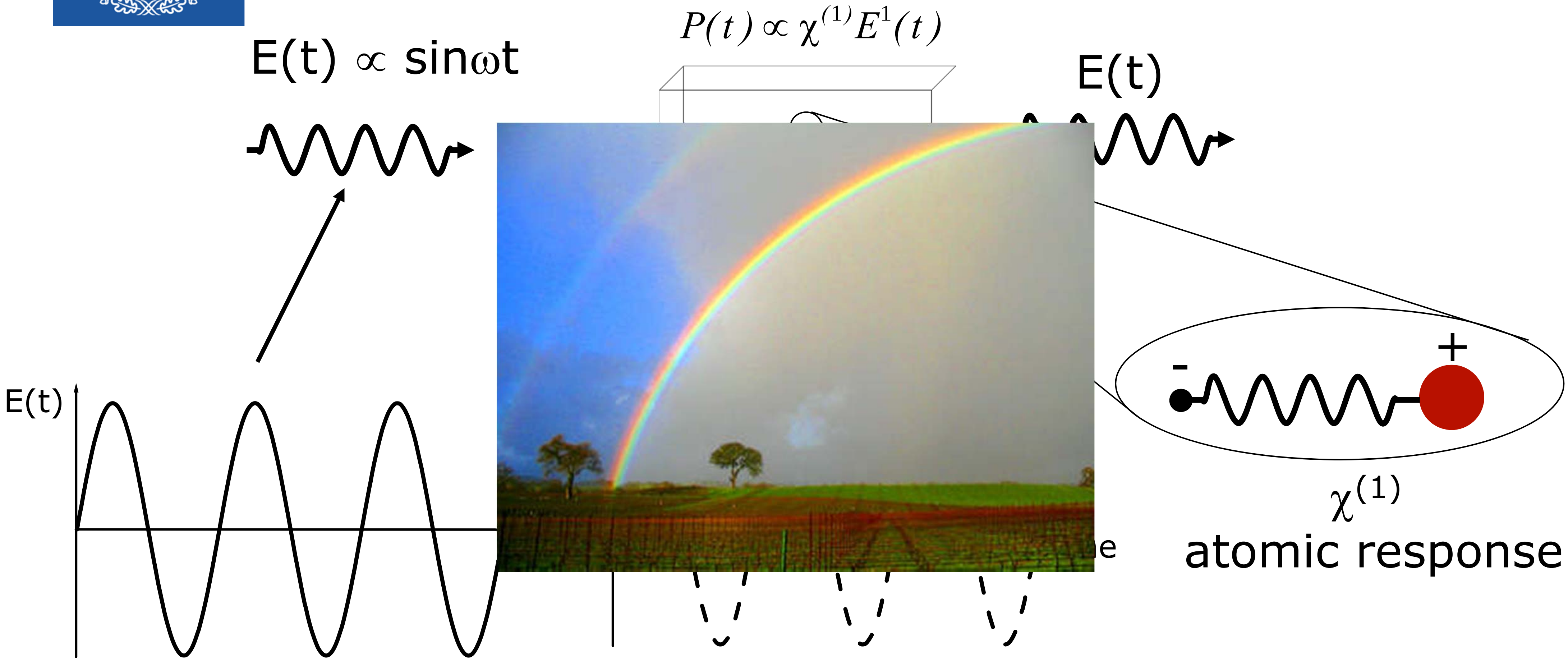
FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.



P.A. Franken, A.E. Hill, C.W. Peters and G. Weinreich, Phys. Rev. Lett. **7** (1961) 118

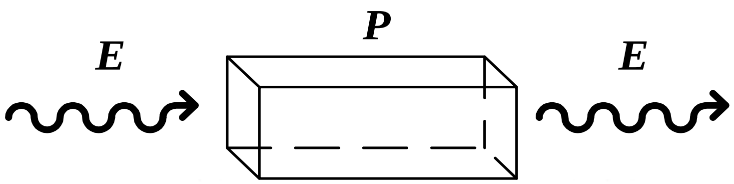
Atomic response on electromagnetic radiation

Linear optical susceptibility

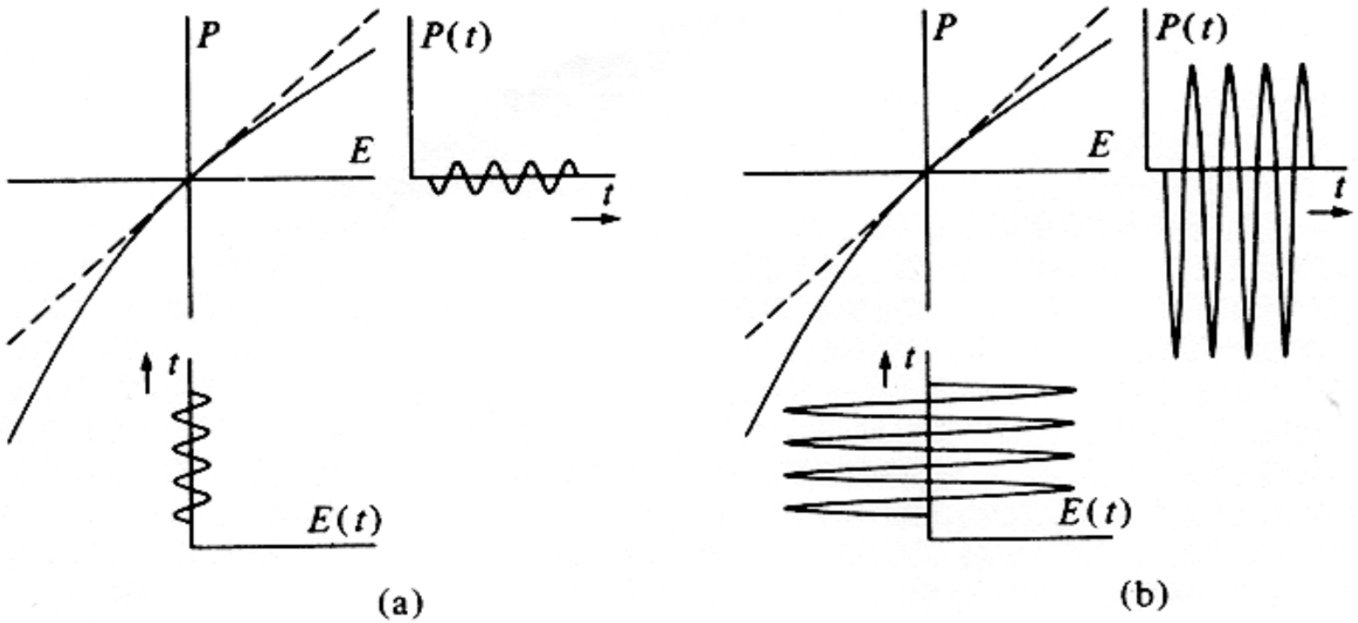


Nonlinear polarisation

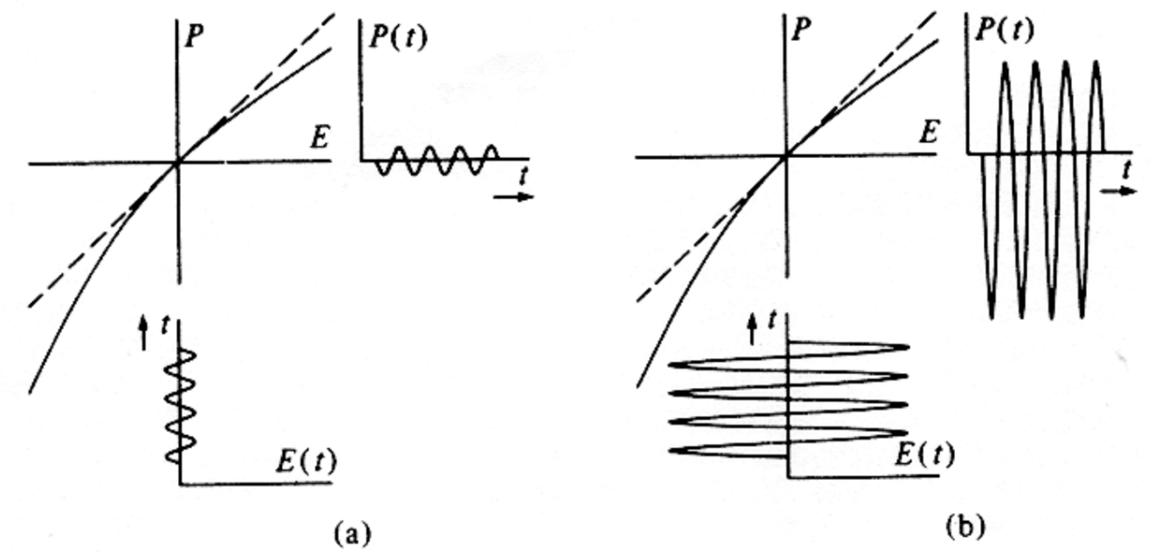
Field induces a polarisation
Driver for a new wave

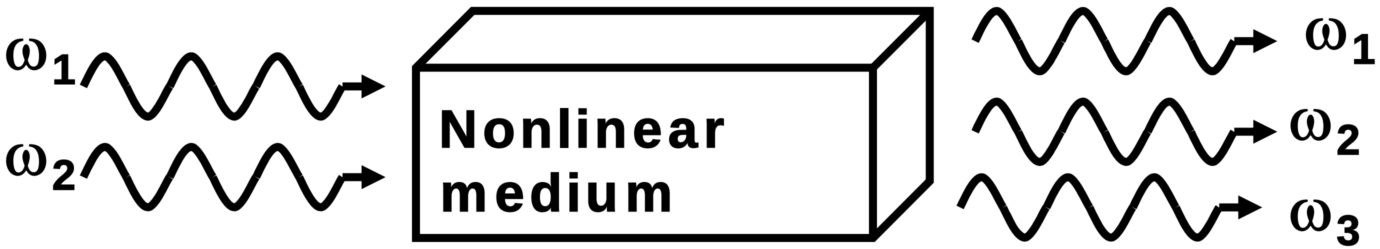


$$\boldsymbol{P} = \varepsilon_0 \chi^{(1)} \boldsymbol{E} + \varepsilon_0 \chi^{(2)} \boldsymbol{E}^2 + \varepsilon_0 \chi^{(3)} \boldsymbol{E}^3 + \ldots = \boldsymbol{P}^L + \boldsymbol{P}^{NL}$$



2^{nd} order effects in noncentrosymmetric materials – three wave mixing





$$2\omega_1 = \omega_3$$

$$2\omega_2 = \omega_3$$

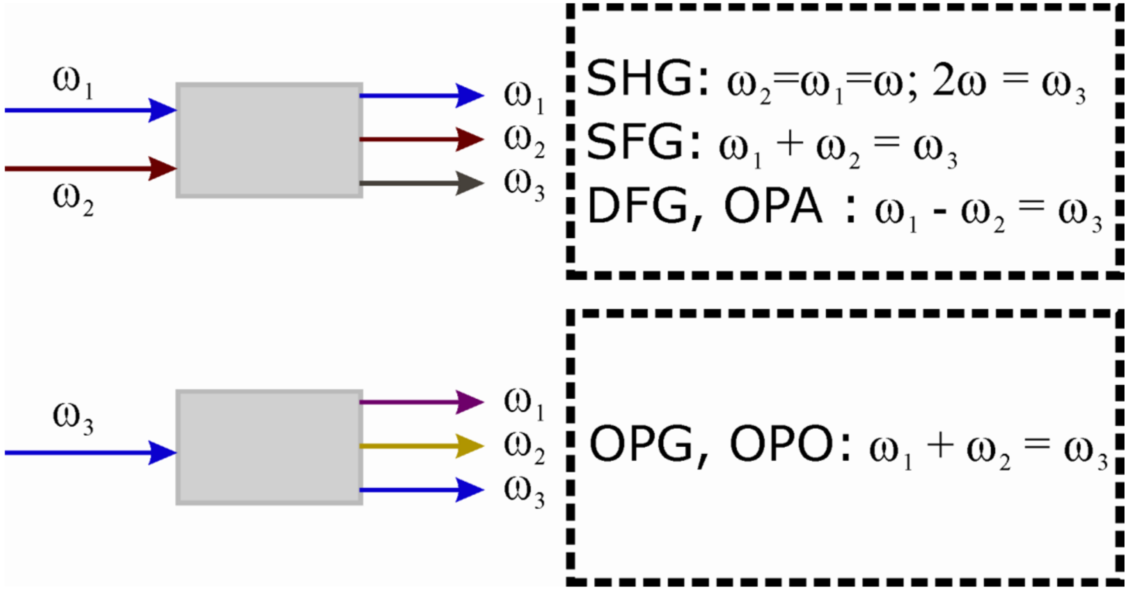
$$\omega_1 \pm \omega_2 = \omega_3$$

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

$$P = \varepsilon_0\, \chi^{(1)} E + \varepsilon_0\, (\chi^{(2)} E^2 + \dots) = P_L + P_{NL}$$

The second order nonlinear processes

$$P^{(2\omega)} \propto \varepsilon_0 \chi^{(2)} E_1 E_2$$



- second harmonic generation
- sum-frequency generation
- difference-frequency generation
- optical rectification
- optical parametric generation
- optical parametric amplification
- optical parametric oscillation

Frequency conversion

$$P = \varepsilon_0 \chi^L E + 2\varepsilon_0 d_{eff} EE$$

$$d_{il} \equiv \frac{1}{2}\chi^{(2)}_{ijk}$$

Maxwell's equations

=>

Nonlinear wave equation

=>

Three coupled equations

$$\frac{dE_s}{dx} = -\alpha_s E_s + \frac{i\omega_s^2}{k_s c^2} K d_{eff} E_p E_i^* \exp(i\Delta k x)$$

$$\frac{dE_i}{dx} = -\alpha_i E_i + \frac{i\omega_i^2}{k_i c^2} K d_{eff} E_p E_s^* \exp(i\Delta k x)$$

$$\frac{dE_p}{dx} = -\alpha_p E_p + \frac{i\omega_p^2}{k_p c^2} K d_{eff} E_s E_i \exp(-i\Delta k x)$$

Solution to the coupled wave equation

Example: Sum-frequency generation

$$\frac{\partial E_1}{\partial z} = i\frac{\omega_1 d_{eff}}{cn_1}E_2E_3\exp(i\Delta kz))$$

$$\frac{\partial E_2}{\partial z} = i\frac{\omega_2 d_{eff}}{cn_2}E_1E_3^*\exp(-i\Delta kz))$$

$$\frac{\partial E_3}{\partial z} = i\frac{\omega_3 d_{eff}}{cn_3}E_1E_2^*\exp(-i\Delta kz))$$

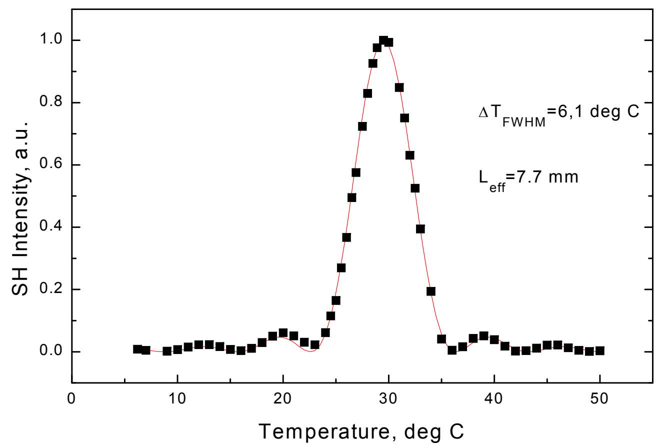
plane-waves and lossless media

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

$$\Delta k = k_1 - k_2 - k_3$$

In small conversion efficiency limit: $\frac{\partial E_2}{\partial z} = 0, \quad \frac{\partial E_3}{\partial z} = 0.$

$$I_1(L) = \frac{2\omega_1^2 d_{eff}^2}{\varepsilon_0 c^3 n_1 n_2 n_3} I_2 I_3 L^2 \operatorname{sinc}^2(\Delta kL/2)$$



General solution to the coupled wave equation

- can be solved analytically
- Solutions only valid for plane-waves and lossless media

General solutions in the case of $\Delta k=0$

SHG

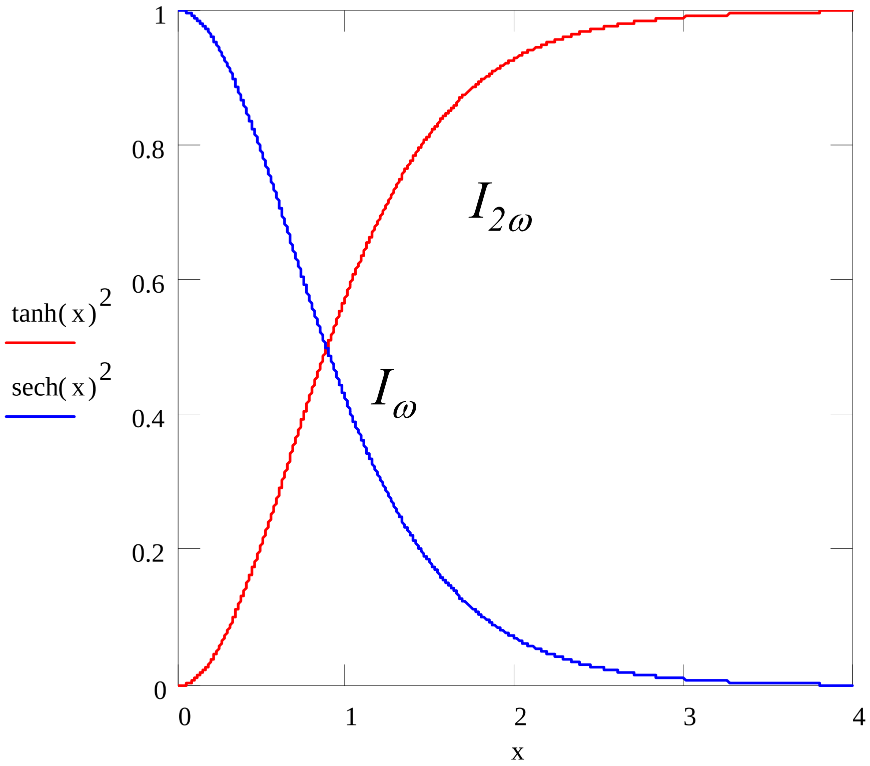
$$I_{2\omega}(L) = I_\omega(0)\tanh^2(\Gamma L)$$

$$I_\omega(L) = I_\omega(0)\,\mathrm{sech}^2(\Gamma L)$$

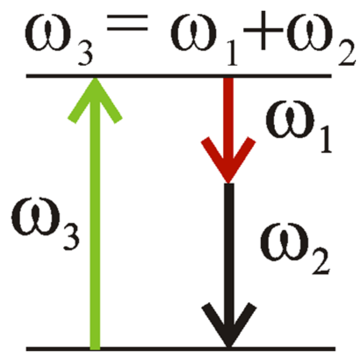
where $$\Gamma = \frac{4\pi d_{eff}\sqrt{I_\omega(0)}}{\sqrt{2cn_\omega^2 n_{2\omega}\varepsilon_0\lambda_\omega^2}}$$

Conversion efficiency is defined as:

$$\eta(L) = I_{2\omega}(L)/I_\omega(0) = \tanh^2(\Gamma L)$$



Physical constraints



- Energy conservation

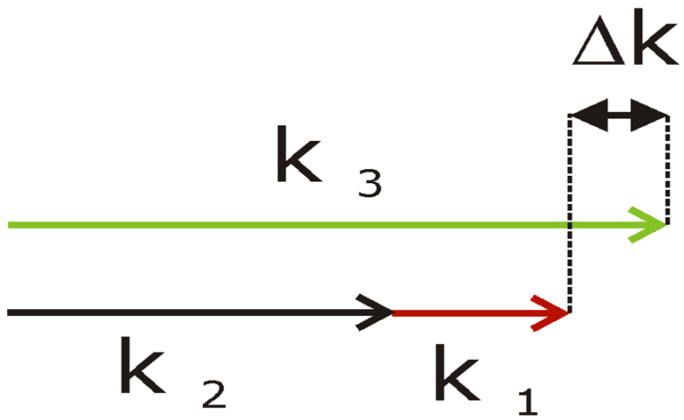
  $\omega_1 + \omega_2 = \omega_3$

- Momentum conservation

  $\Delta k = k_3 - k_2 - k_1 =$

  $= 1/c \times [n_3\ \omega_3 - n_2\ \omega_2 - n_1\ \omega_1]$

  n_i is the refractive index

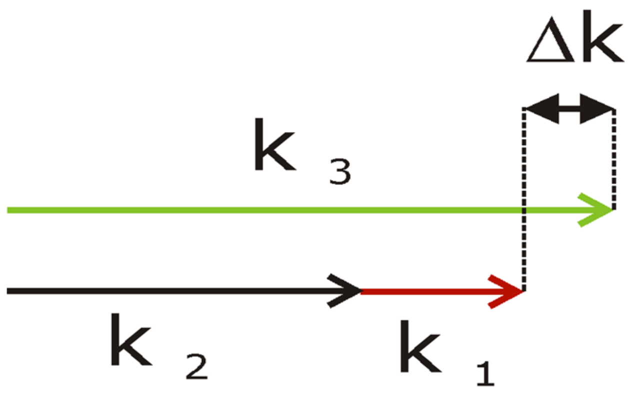


Coherence length: $L_c = \left| \frac{\pi}{\Delta k} \right|$

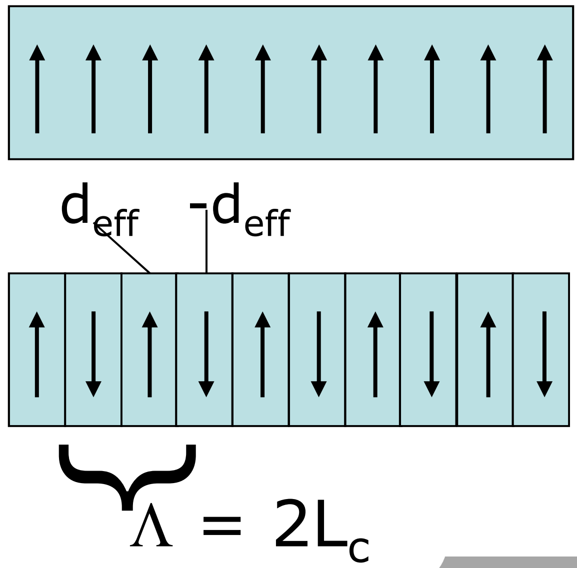
Phasematching

Momentum conservation

$$\Delta k = k_3 - k_2 - k_1$$

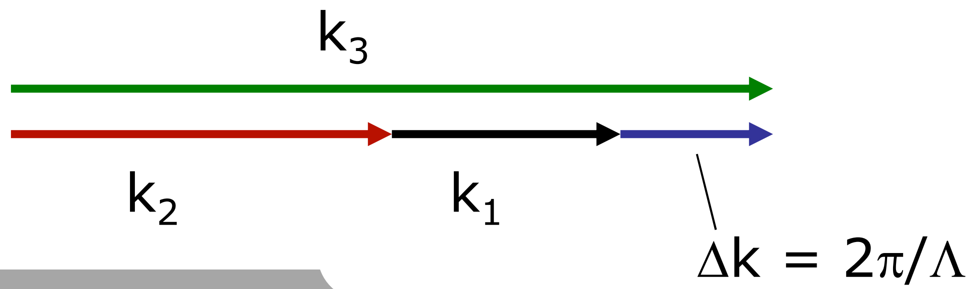


Modulation of the nonlinearity

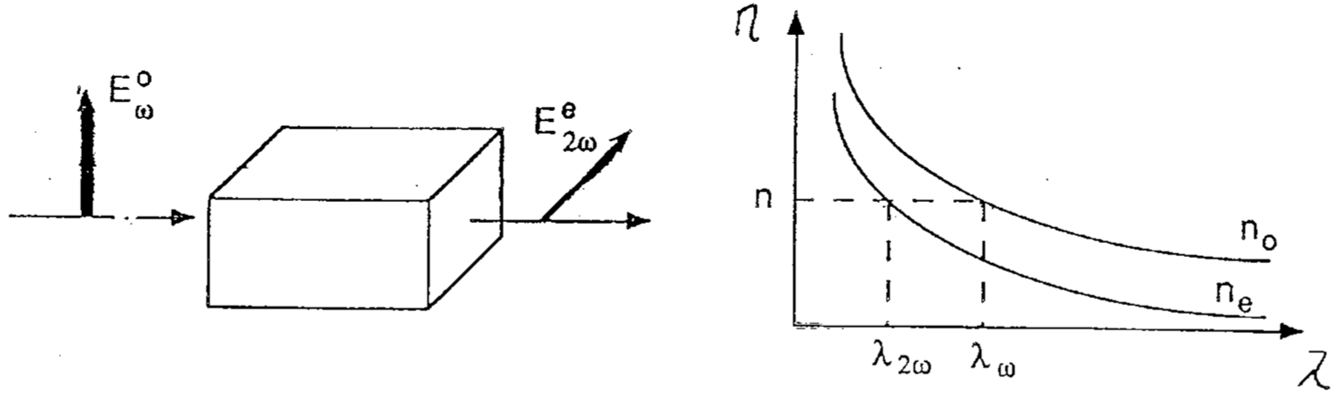


Birefringent phasematching $\quad \Delta k = 0$

Quasi-phase-matching $\quad \Delta k \neq 0$



Birefringent phasematching

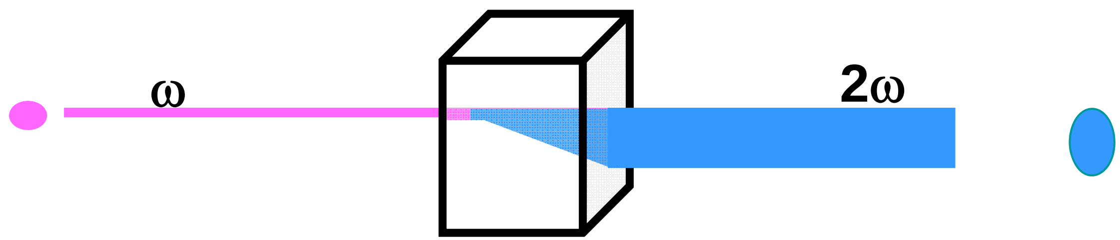


- need birefringent crystal
- $\chi^{(2)}$ must couple orthogonal polarizations
- temperature constraints
- cannot use full transparency range

Limited phasematching range

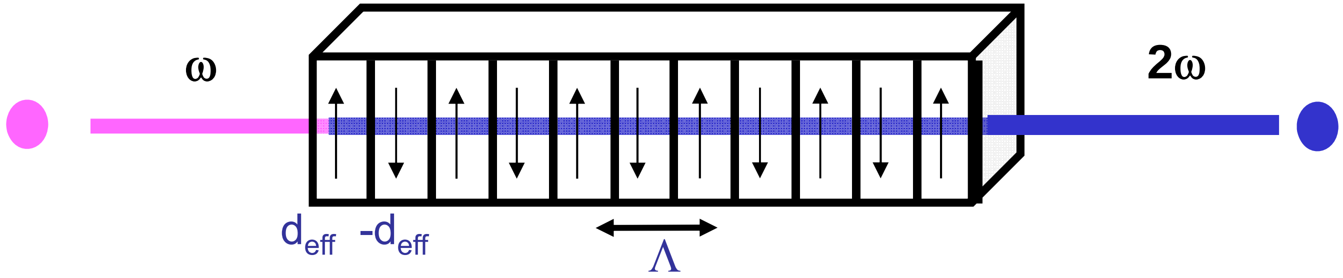
Walk-off – short interaction length

Low nonlinearity



Quasi-phase matching (QPM)

Proposed by Bloembergen, *et al.* 1962



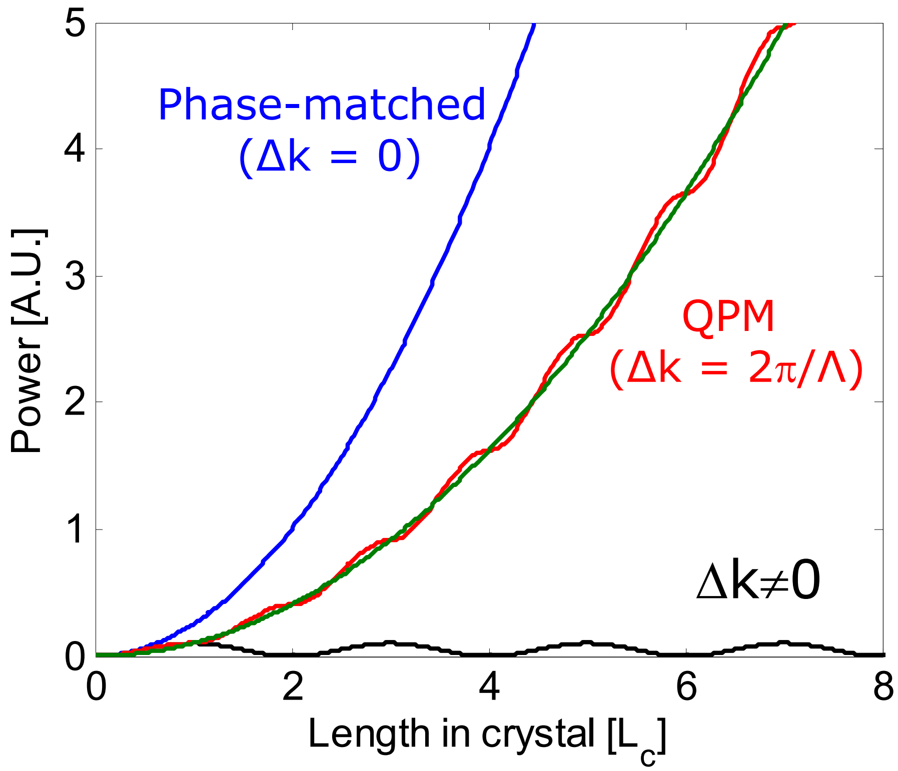
Periodic inversion of domains in ferroelectrics

Modulation of the nonlinear susceptibility

to compensate the phase mismatch

- Arbitrary wavelength within transparency
- Walk-off free interaction – long lengths
- Highest nonlinearity accessible

$$L_c = \left| \frac{\pi}{\Delta k} \right|$$



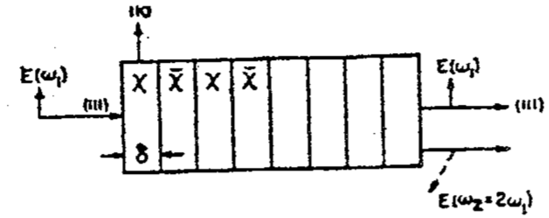
The main advantages of QPM

- Phasematching at any wavelength within transparency
- Free choice of polarization of interacting waves
- Largest nonlinear coefficient accesible, d_{33}
- Noncritical phasematching, walk-off free interaction
- Enigineering of the phasematching condition

Implementations of QPM

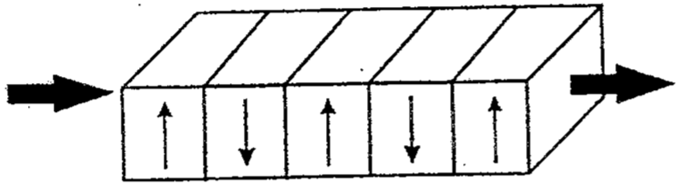
- Stacked plates with alternating layers rotated.

[Bloembergen 1962]



(a) ZnS

- In ferroelectric materials, sign of d_{eff} is reversed with reversal of domain polarity.



Monolithic crystal with grating structure

- Electric-field poling with lithographic electrodes can achieve near-ideal structure for QPM.

[Yamada 1993]

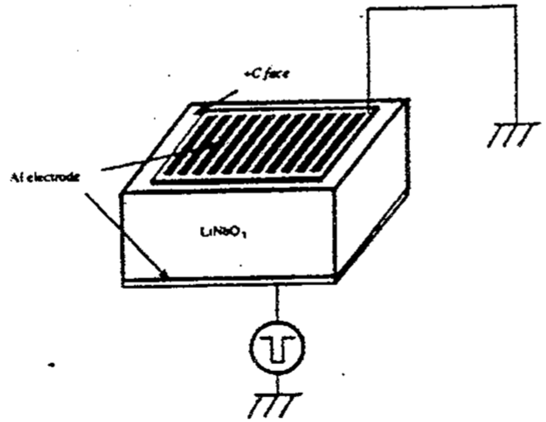


FIG. 1. Schematic of applying voltage for periodically domain inversion.

Types of phasematching

Type 0 SHG two photons having extraordinary polarization will combine to form a single photon with double the frequency/energy and extraordinary polarization.

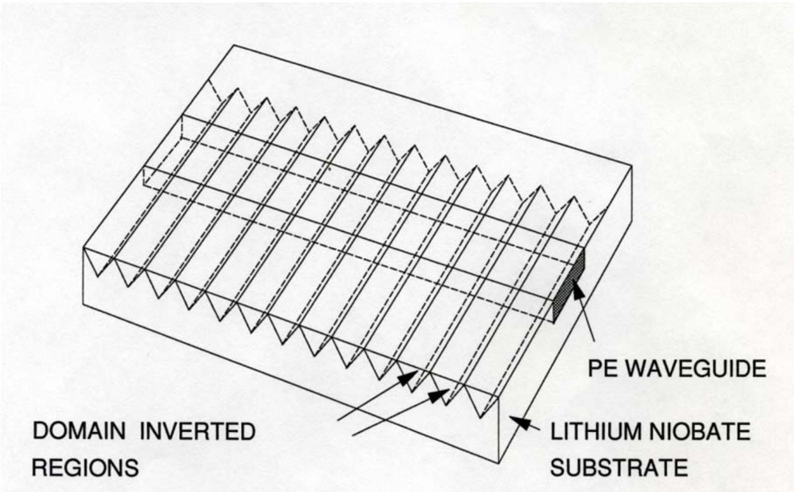
In **Type I SHG** two photons having ordinary polarization will combine to form one photon with double the frequency and extraordinary polarization.

In **Type II SHG**, two photons having orthogonal polarizations will combine to form one photon with double the frequency and extraordinary polarization

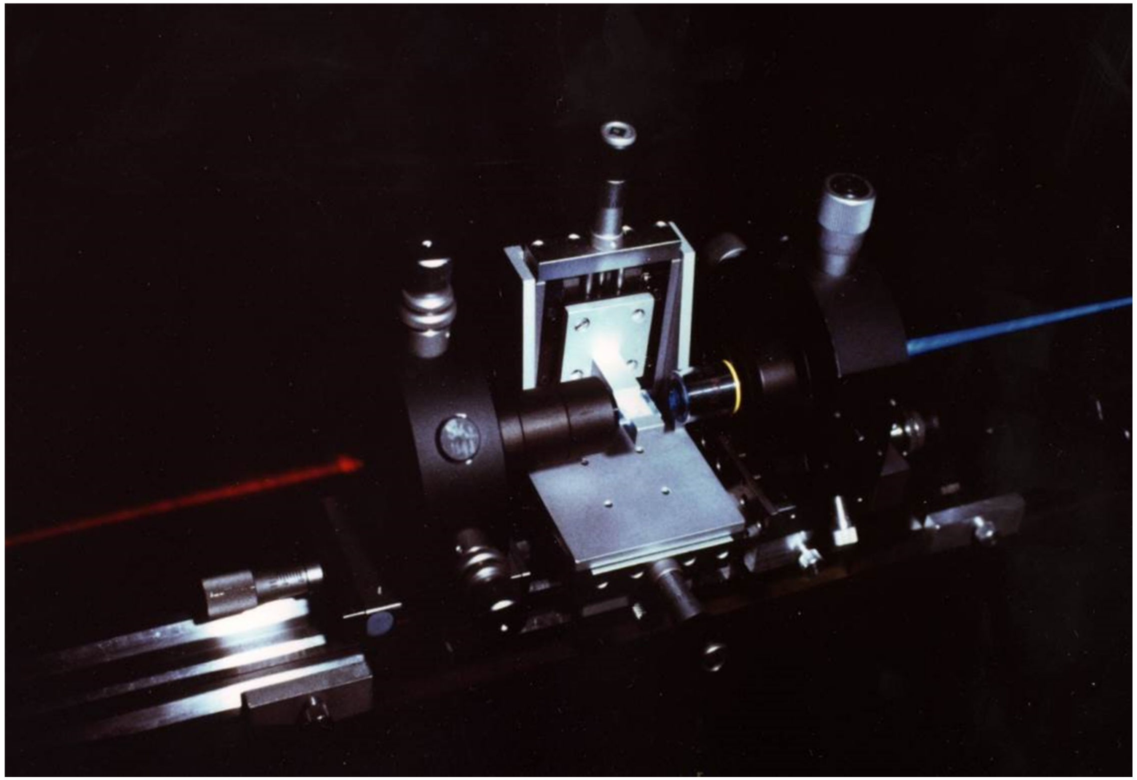
In moat cases Quasi-phase-matching is used with a **Type 0** interaction

Quasi-phase matching (QPM)

First efficient demonstration with $LiNbO_3$ waveguides



Domain structure fabricated
by diffusion techniques

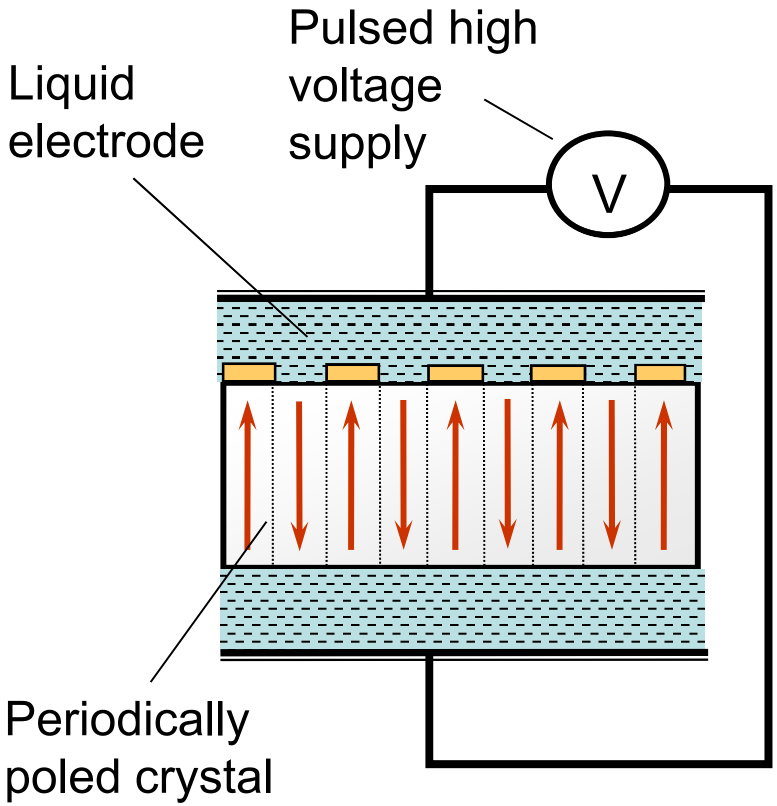


Blue light generated with AlGaAs diode laser

J Webjorn, F. Laurell, and G. Arvidsson, IEEE Photon. Technol. Lett., 1, 316, 1989

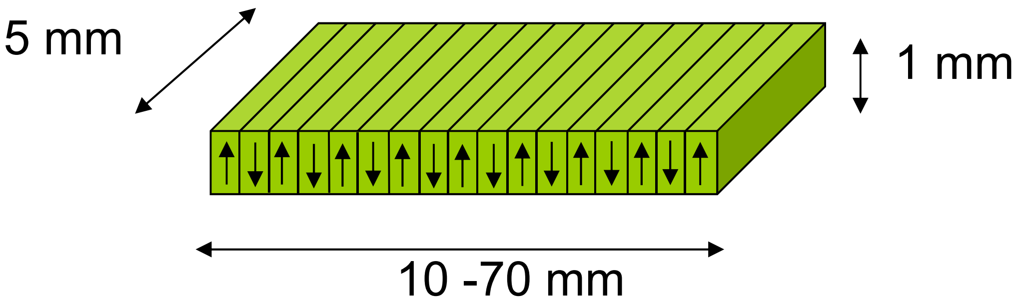
Electric field poling of bulk crystals

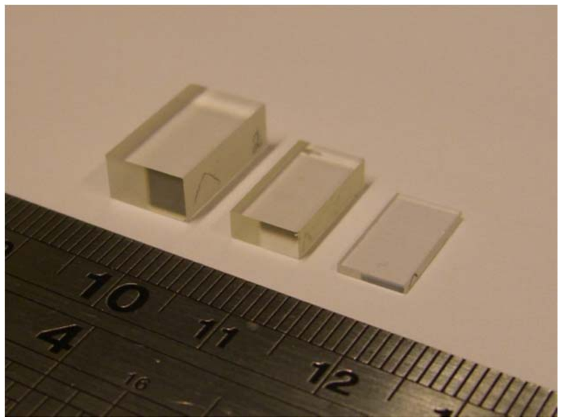
- $LiNbO_3$
- $LiTaO_3$
- $KTiOPO_4$



Bandwidth tailored for specific interaction

Damage resistant and efficient material





sub-μm to 100 μm periods

M. Yamada, N. Nada, M. Saitoh, and K. Watanabe, Appl. Phys. Lett. **62**, 435 (1993)

$LiNbO_3$, the first QPM material

- Periodically poled in 1993*
- Available in large, homogeneous and inexpensive wafers
- High nonlinearity
- “Straight forward” poling

•High coercive field (congruent crystal ~21 kV/mm)

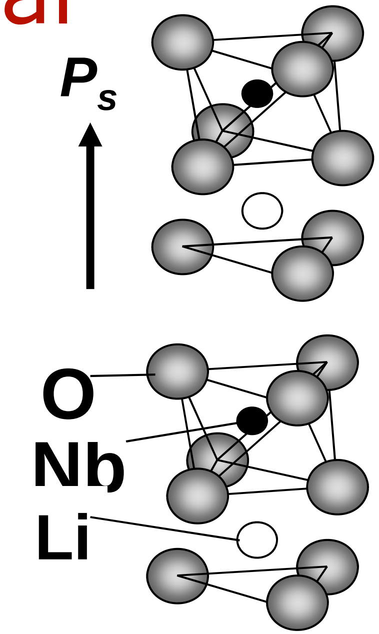
➔ Limited thickness (~0.5 mm)

➔ Hexagonal domains -severe broadening

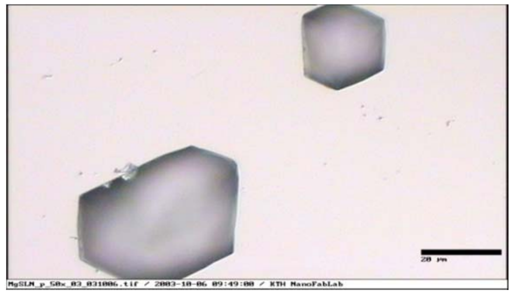
difficult to fabricate dense gratings (< 10 μm)

•Optical damage (Photorefraction, light-induced absorption)

➔Limited power handling, life time issues



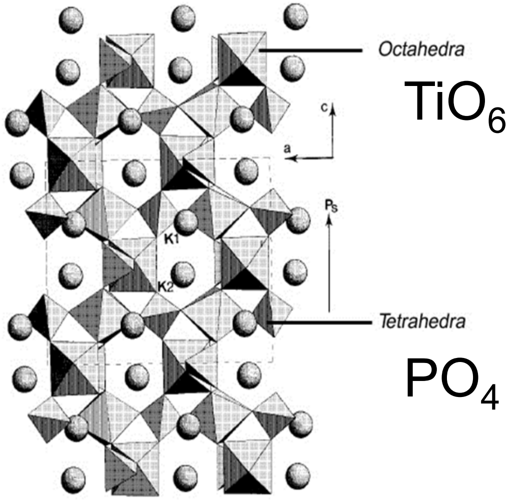
Trigonal 3m



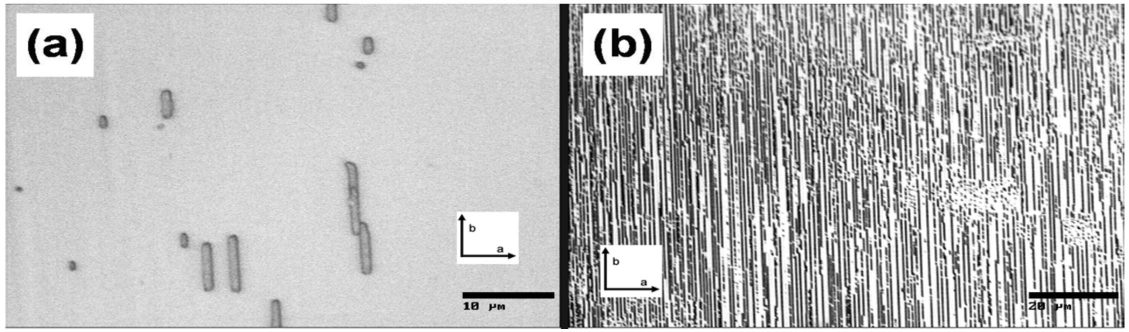
Solution MgO-doped Lithium niobate – optical damage ”resistant”
suitable for waveguide fabrication
commercially available

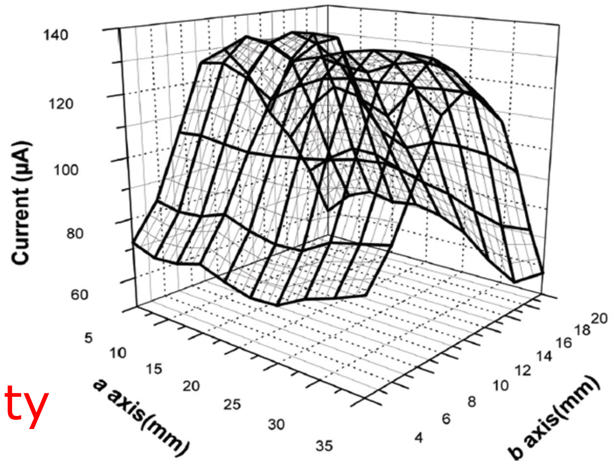
*M. Yamada, Appl. Phys. Lett. **62**, 435 (1993).

Flux grown $KTiOPO_4$



Domain morphology





High nonlinearity
High resistance to optical damage
Low coercieve field (2 kV/mm)
dense gratings, ”thick” samples (3mm)

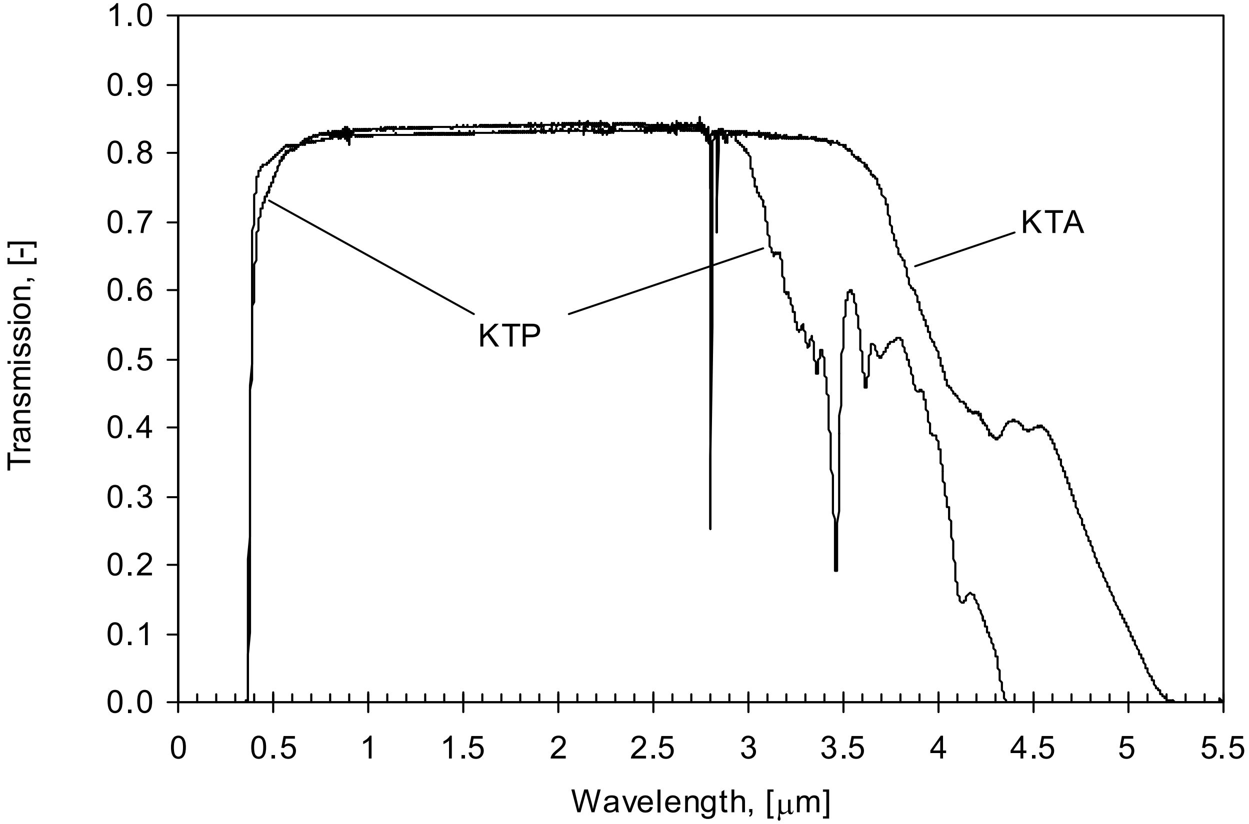
Difficult to grow – 1 inch wafers
Non-stoichiometric - inhomogeneous – highly varying conductivity
Expensive

Requirement for NLO materials for frequency conversion

- **Phasematching --** limits useful interactions & applicability
- **Transmission --** interacting waves not absorbed or scattered
- **Nonlinearity --** non-centrosymmetric materials for $\chi^{(2)}$
- **Homogeneity --** uniformity 1 part in 10^{-5}
- **Damage --** absolute & relative to operating point
- **Mechanical properties --** growing, polishing, coating
- **Thermal properties --** dn/dT, thermal conductivity
- **Lifetime --** chemical stability, hygroscopic, aging in use
- **Lack of "weirdness" --** photorefractive, gray tracking
- **Availability --** size, cost, uniformity of properties

All requirements must be simultaneously satisfied!

$KTiOPO_4$ Transparency



$LiNbO_3$

Transparency 0.35 - 5.4 μm

Figure of merit for nonlinear materials

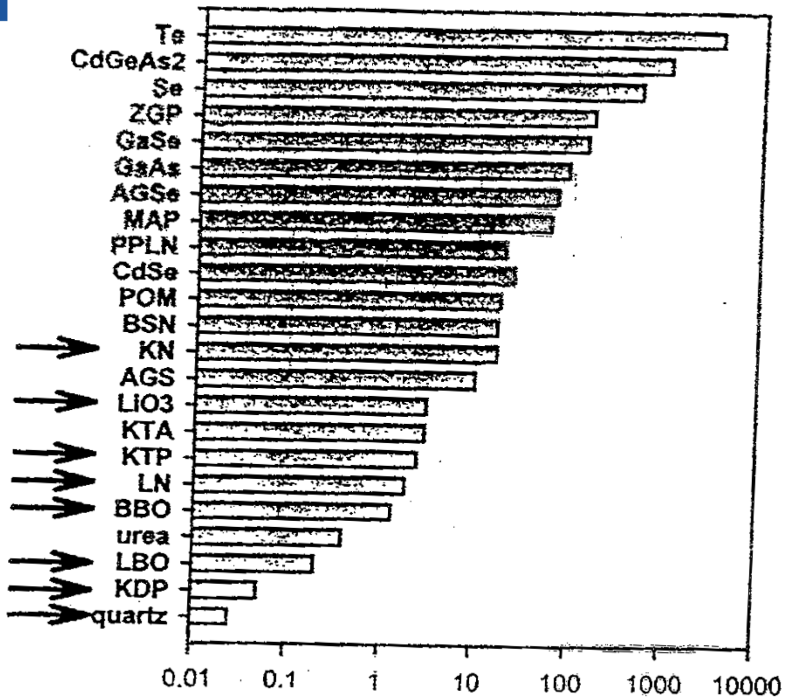


Figure of Merit (d^2 / n^3)

→ = used in commercial device

- Value of d_{eff} depends on angles of propagation & polarization and on wavelengths

Units of d_{eff} ~ pm/V

$$\text{Figure of Merit} = \frac{d_{eff}^2}{n^3}$$

- Commercially used materials have lower nonlinear coefficients
- High nonlinear coefficient does not necessarily make material useful

Comparison of PP crystals

- $KTiOPO_4$ family ($KTiOPO_4$, $RbTiOPO_4$, $KTiOAsO_4$, $RbTiOAsO_4$)
- $LiNbO_3$ family
  - stoichiometric $LiTaO_3$ (SLT)
  - 1-5 mol% MgO doped stoichiometric $LiTaO_3$ - (MgO:SLT)
  - 1-5 mol% MgO doped stoichiometric $LiNbO_3$ - (MgO:SLN)
- $KNbO_3$

Low coercive field (<5 kV/mm)

Higher optical damage threshold

Figure of merit for SHG of 1064 nm

$$\frac{d^2}{n_{\omega_1} \cdot n_{\omega_2} \cdot n_{\omega_3}}$$

	LN	KN	KTP	LT
FOM	61.1	38.8	33.7	18.8

Artifically structured ferroelectrics

Domain gratings with µm periodicity

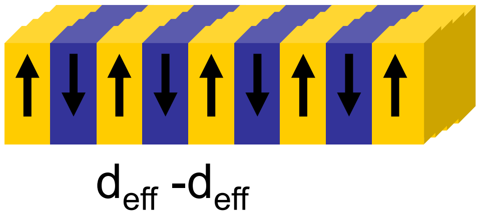
Frequency converters: from UV to mid-IR

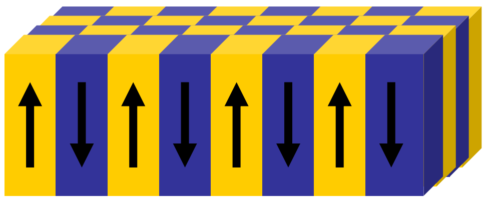
Domain gratings with nm periodicity

New devices:

electrically controlled Bragg reflectors, BSHG, BOPO,…

1D – gratings





2D structures - Nonlinear photonic crystals

Second-harmonic generation

- Energy conservation:

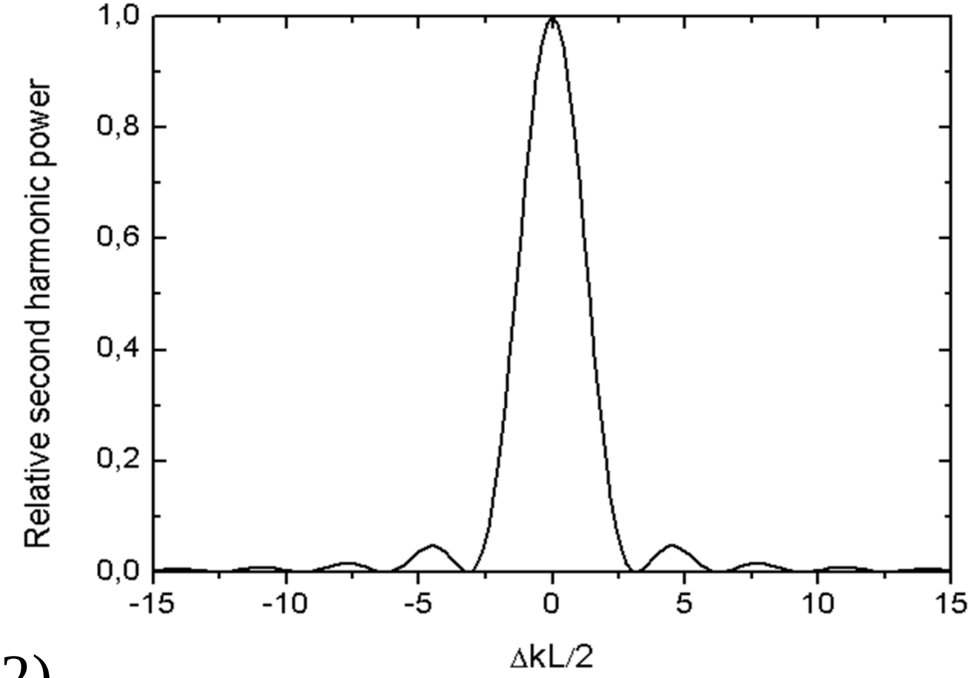
$$\omega_3 = 2\omega_1 = 2\omega_2$$

- Momentum conservation:

$$\Delta k = k_{SH} - 2k_F$$

- SH power for Gaussian beam:

$$P_{SH} = \left(\frac{2\omega_F^2 d_{eff}^2 k_F P_F^2}{\pi n_F^2 n_{SH} \varepsilon_0 c^3} \right) Lh(B,\xi)\ \mathrm{sinc}^2(\Delta k L/2)$$



$$d_{eff}^{the} = \frac{2d_{il}}{m\pi}$$

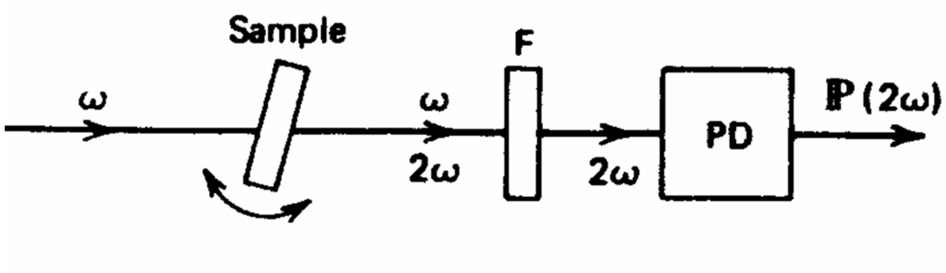
- Phasematching:

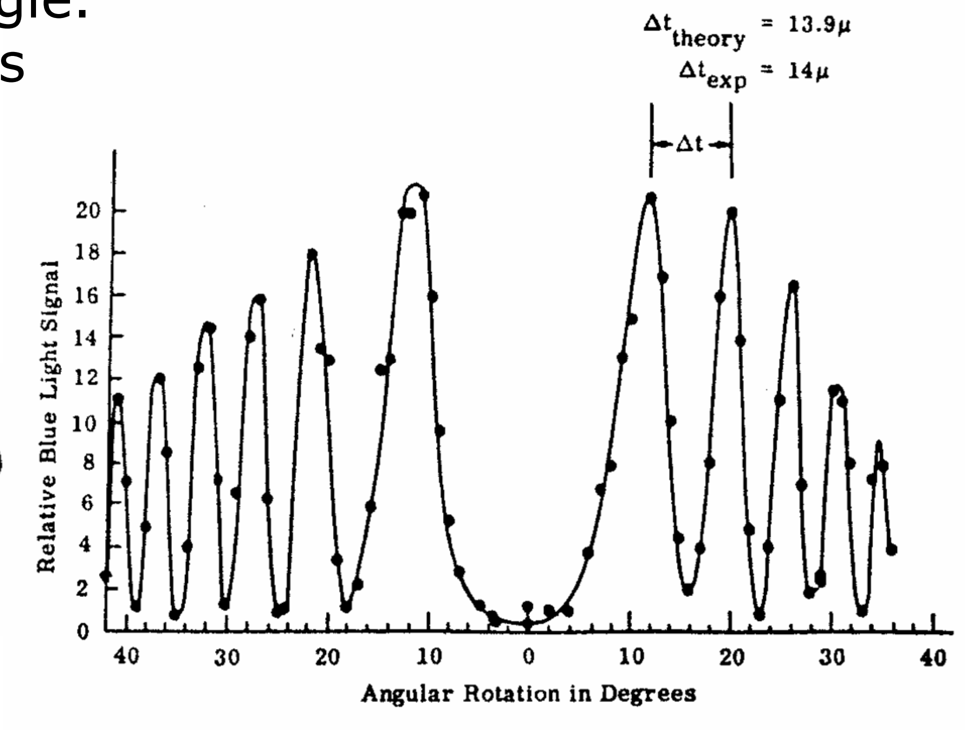
$$\Delta k \begin{cases} = 0: & \text{Perfect phase matching} \\ \neq 0: & \text{SH} \leftrightarrow \text{Fundamental after a distance, Lc} = \pi / \Delta k \end{cases}$$

Maker fringe method

Useful method to characterize a nonlinear material

For given polarisation,
measure the SHG as function of angle.
Determine the nonlinear coefficients
and coherence length

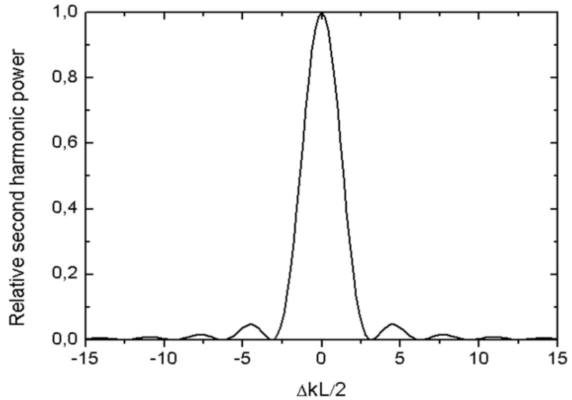




P.D. Maker, R.W. Terhune, M. Nisenoff, and C. M. Savage, Phys. Rev. Lett. **8**, 19 (1962).

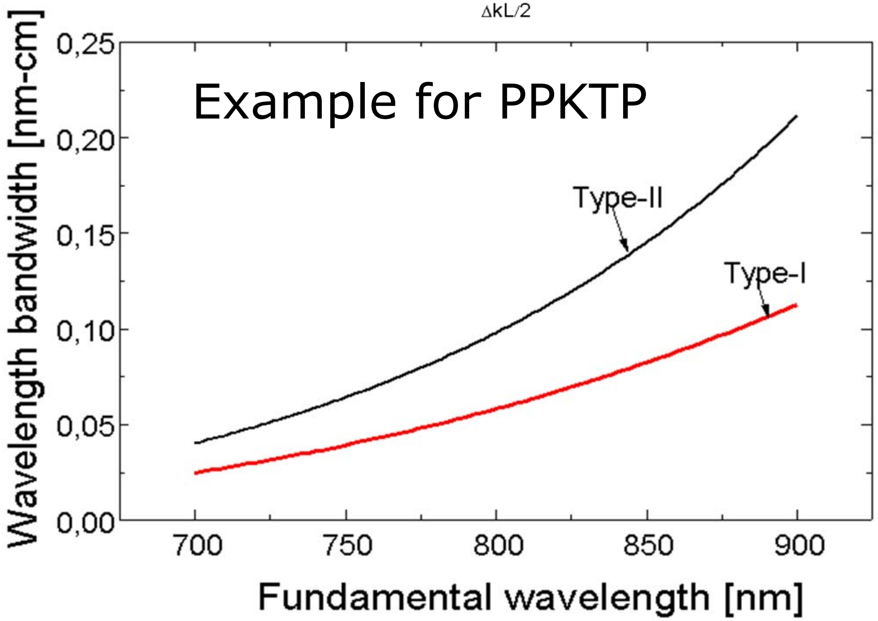
Phasematching bandwidth

The mismatch Δk depends on λ, T and polarization of waves



- Wavelength acceptance bandwidth

$$\Delta\lambda_{FWHM} = \frac{0.4429\lambda}{L}\left|\frac{n_{SH}-n_F}{\lambda}+\frac{\partial n_F}{\partial\lambda}-\frac{1}{2}\frac{\partial n_{SH}}{\partial\lambda}\right|^{-1}$$



- Temperature acceptance bandwidth

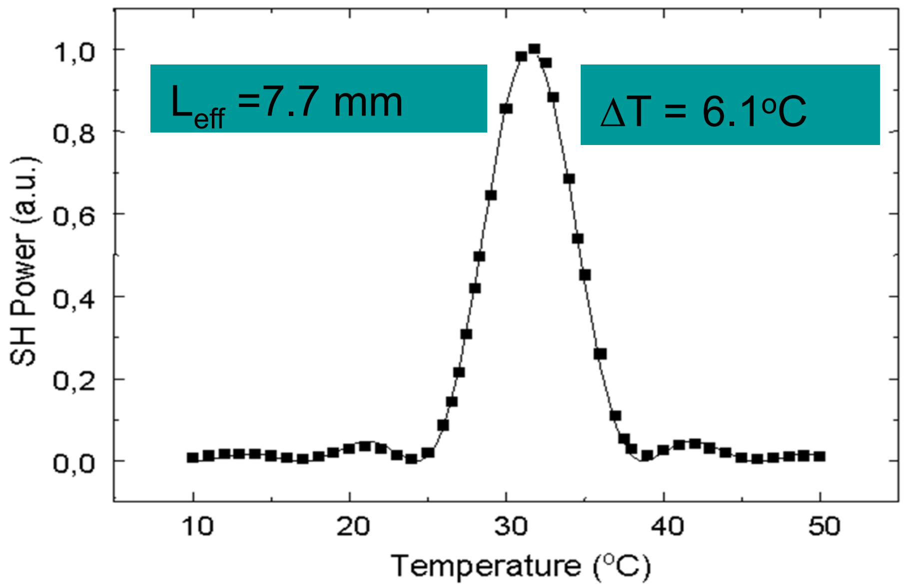
$$\Delta T_{FWHM} = \frac{0.4429\lambda_F}{L}\left|\left.\frac{\partial n_{SH}}{\partial T}\right|_{T_0}-\left.\frac{\partial n_F}{\partial T}\right|_{T_0}+\alpha(n_{SH}-n_F)\right|^{-1}$$

Tuning characteristics

Quality of a PP structure: $L_{eff} \propto 1/\Delta T_{FWHM}$ or $L_{eff} \propto 1/\Delta\lambda_{FWHM}$

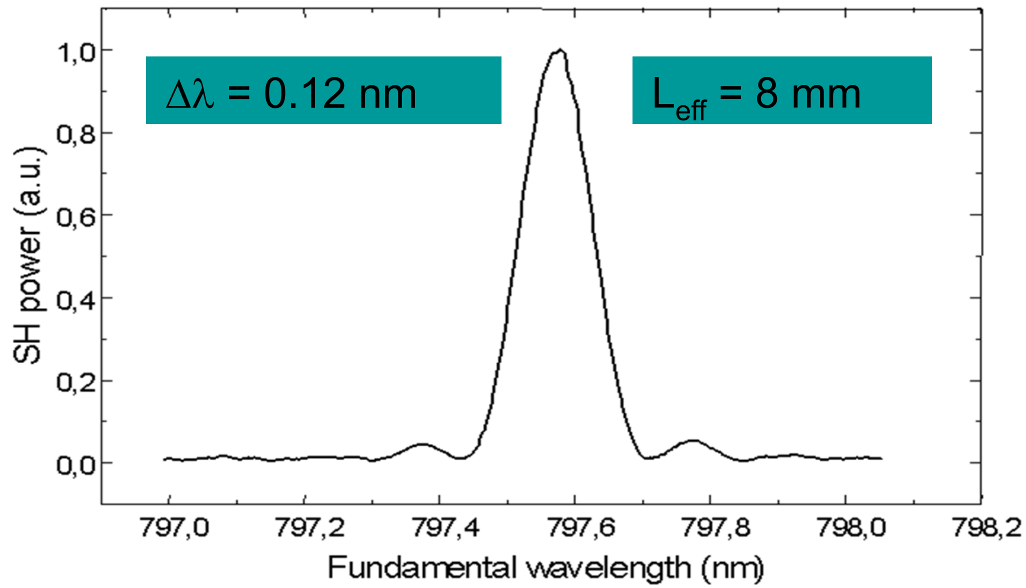
Temperature tuning

λ = 1064 nm
Type-I SHG @ 532 nm
PPKTP: Λ = 9.01μm, $L_{physical}$ = 8 mm



Wavelength tuning

λ = 797.6 nm
Type-II SHG @ 398.8 nm
PPKTP: Λ = 9.01μm, $L_{physical}$ = 8.5mm



Effective QPM nonlinear coefficient

- Theoretical value of effective nonlinear coefficient:

$$d_{eff}^{the} = \frac{2d_{il}}{m\pi}$$

KTP @ 1064 nm:
d_{33}= 16.9 pm/V; d_{24} = 3.64 pm/V

- Experimental value of effective nonlinear coefficient:

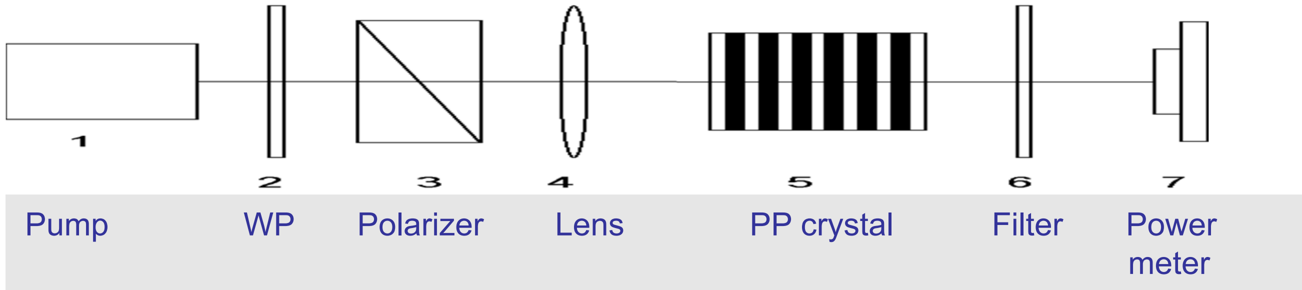
$$d_{eff}^{\exp} = \sqrt{\frac{\pi n_F^2 n_{SH} \varepsilon_0 c^3}{2\omega_F^2 k_F L h(B,\xi)} \times \frac{P_{SH}}{P_F^2}}$$

- Useful technique to screen quality of PP crystals

CW SHG

low power regime

Single-pass SHG set-up

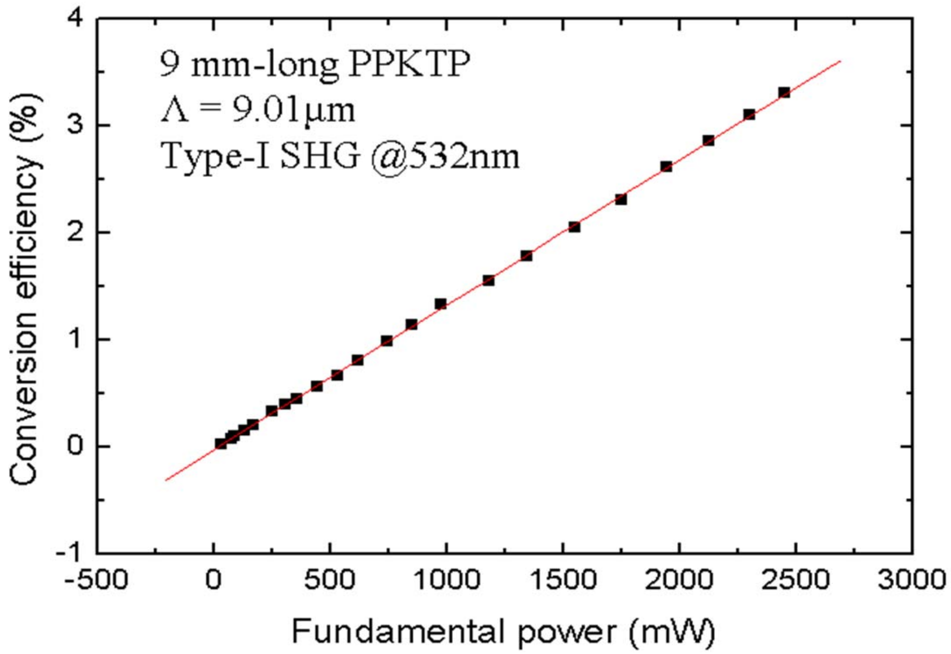


Power and conversion efficiency

$$P_{SH} \propto P_F^2 \qquad \eta = \frac{P_{SH}}{P_F}$$

Normalized conversion efficiency

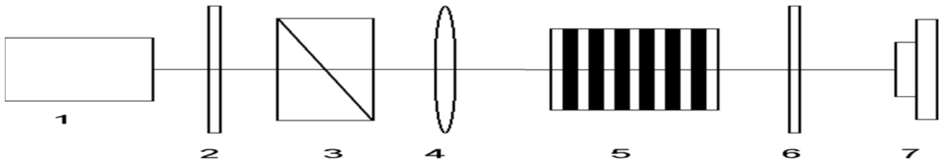
$$\eta_{norm} = \frac{P_{SH}}{P_F^2 L}$$



CW SHG

high power regime

Single-pass SHG set-up

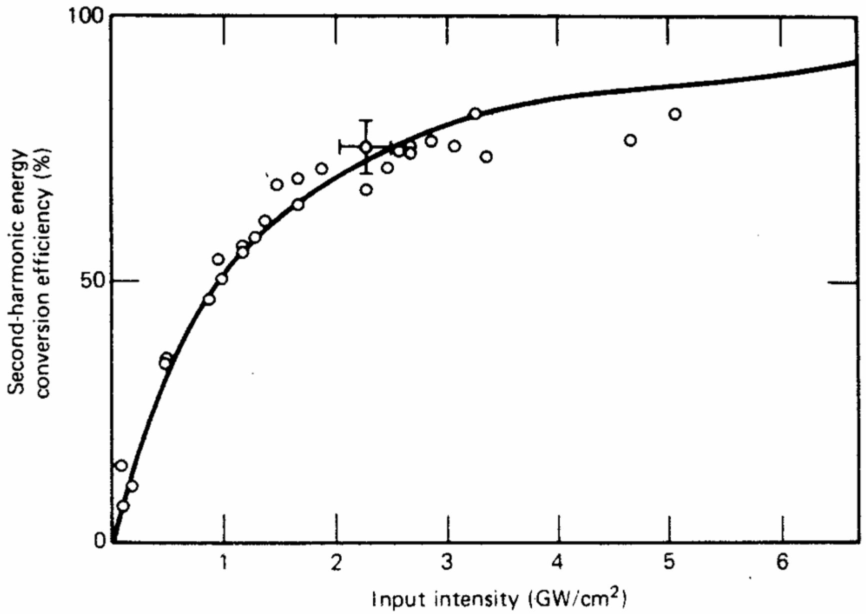


conversion efficiency

$$\eta(L) = I_{2\omega}(L) / I_{\omega}(0) = \tanh^2(\Gamma L)$$

$$\Gamma = \frac{4\pi d_{eff}\sqrt{I_{\omega}(0)}}{\sqrt{2cn_{\omega}^2 n_{2\omega}\varepsilon_0\lambda_{\omega}^2}}$$

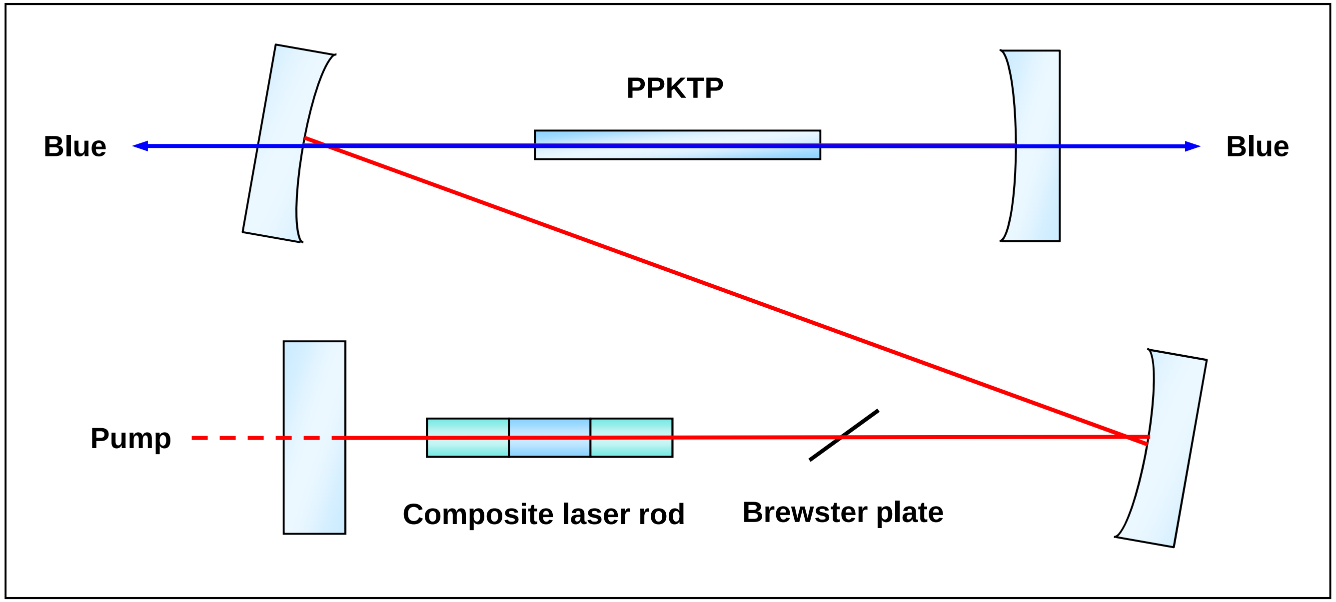
SHG in KDP crystal

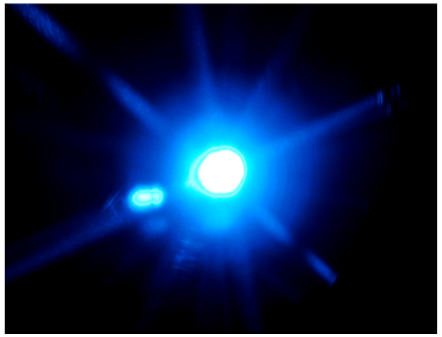


From Yariv: Quantum Electronics

Intra-cavity frequency doubling

- Much higher power available in the cavity
- Often associated with instabilities, the so called “green problem”
- 946 nm → 473 nm

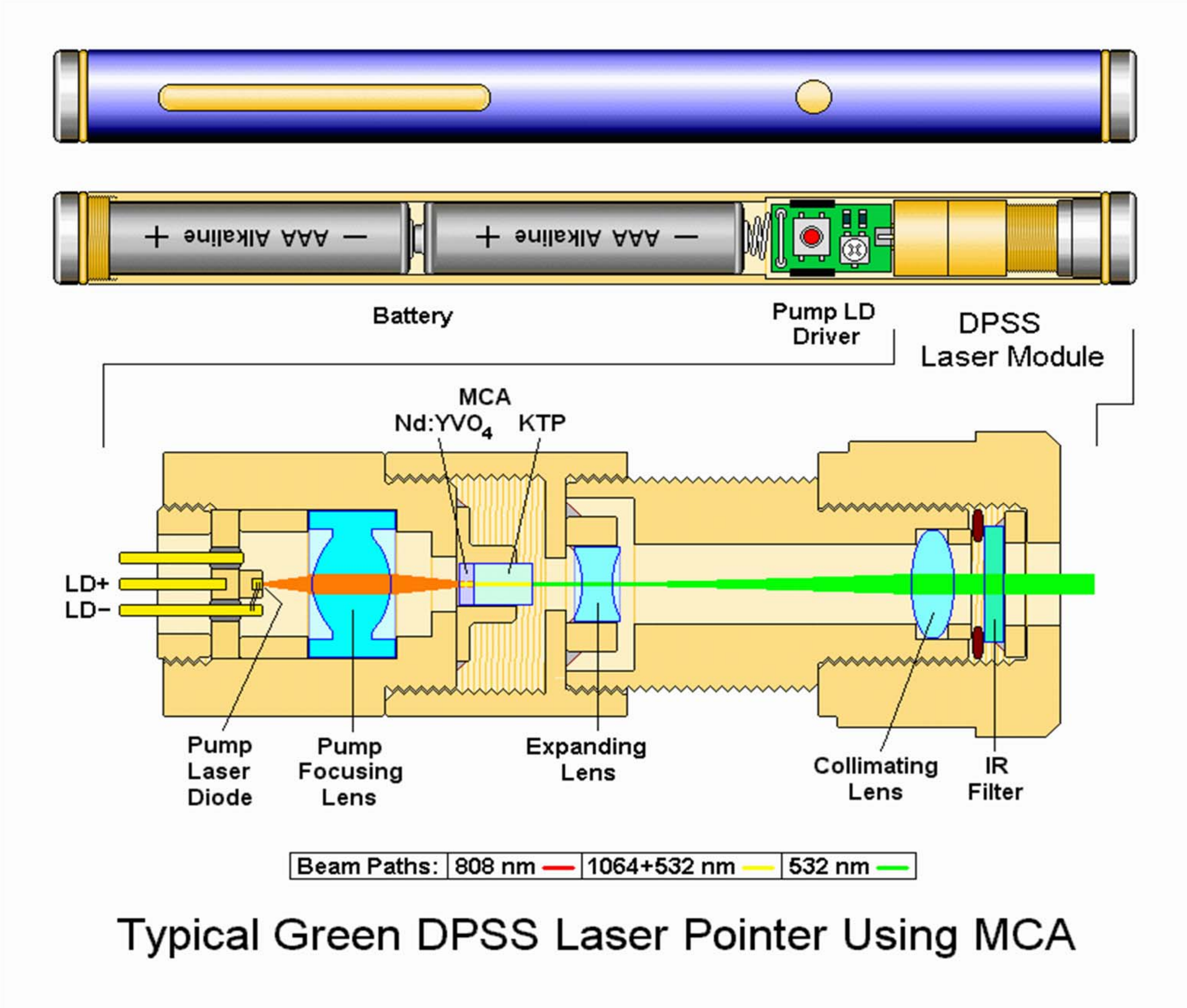




> 500 mW

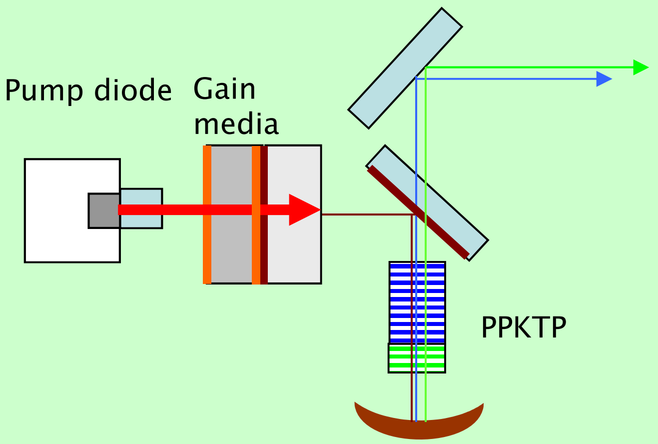
The laser pointer

A diode-pumped IC frequency doubled solid-state laser

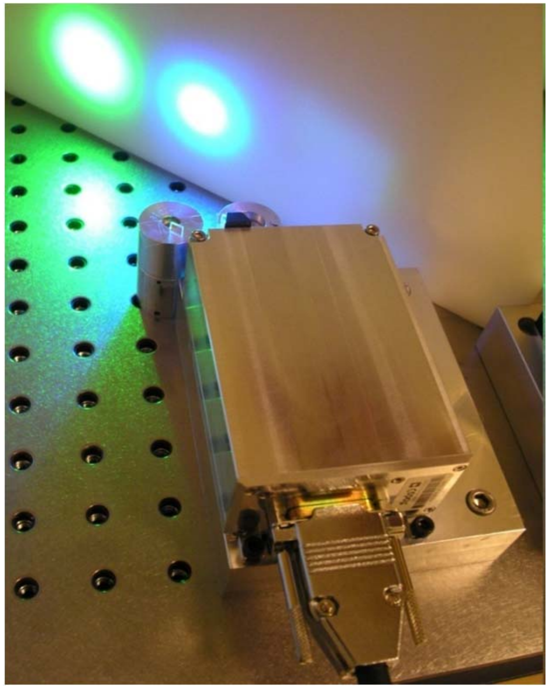


IC SHG and SFG lasers with PPKTP

Mixing of two laser lines combined with SHG and SFG



Frequency Mixing of two DPSSLs in PPKTP



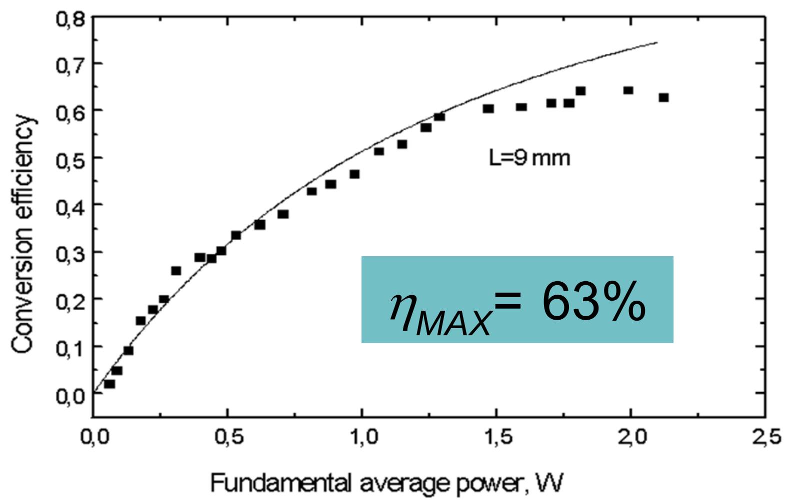
Lasers for biotech, display and graphics

Pico- and nano-second SHG

- high peak power - take pump depletion under consideration
- Absorption of SH light can limit the efficiency

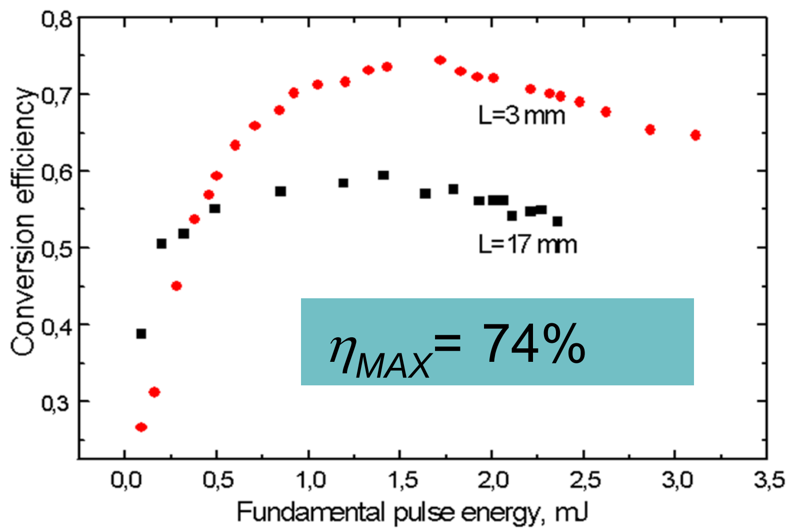
Mode-locked Nd:YAG

τ = 100 ps, f = 100 MHz
Type-I SHG @ 532 nm
PPKTP: Λ =9.01 μm



Q-switched Nd:YAG

τ = 5 ns, f = 20 Hz
Type-I SHG @ 532 nm
PPKTP: Λ =9.01 μm



Ultrashort pulse SHG

- Group-velocity mismatch limits the effective interaction length
- SH pulse spreading
- SH spectral broadening due to GVM
- Intensity-dependent SH spectral broadening

τ = 100 fs, f = 80 MHz

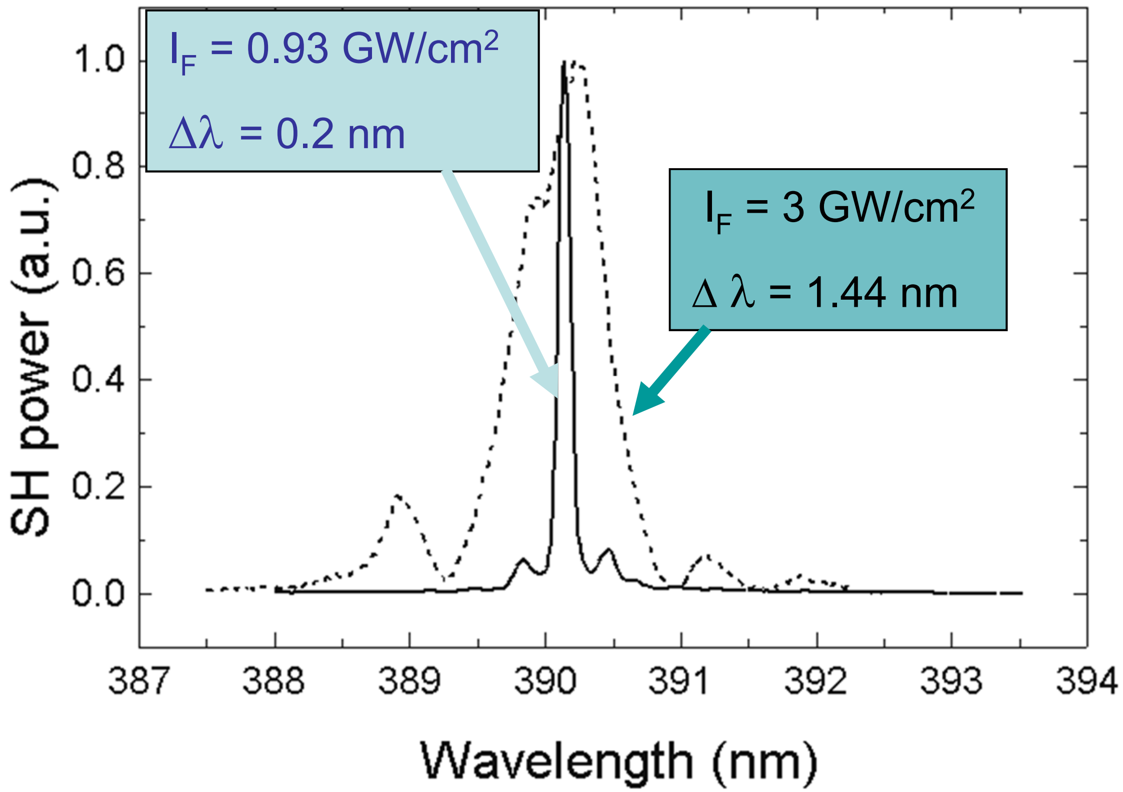
Type-I SHG @ 390 nm

PPKTP: Λ = 2.95 μm, L = 9 mm

L_{eff} = 5.6 mm

L_W = 57 μm

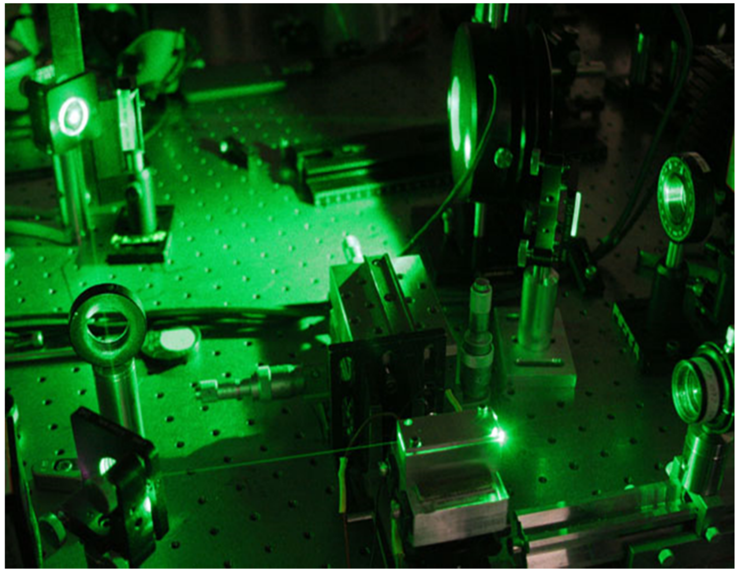
τ_{SH} = 10 ps

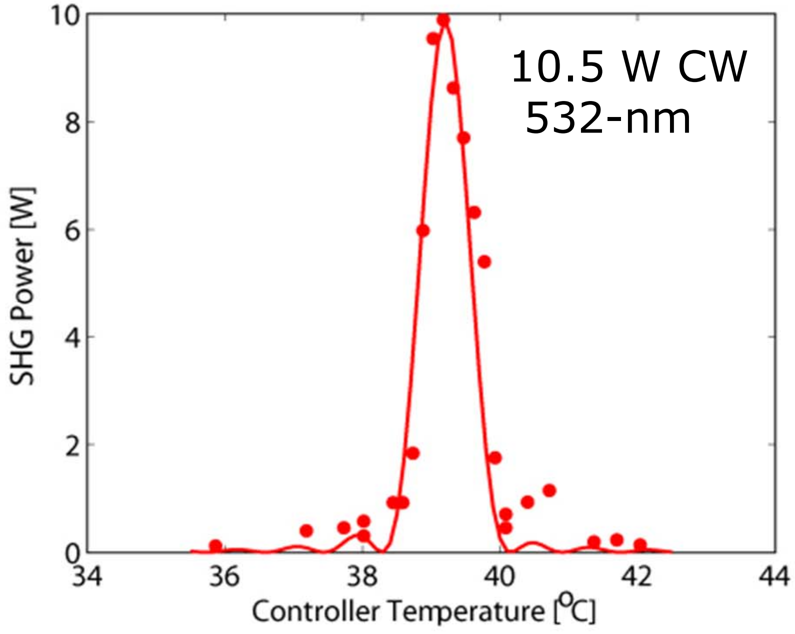


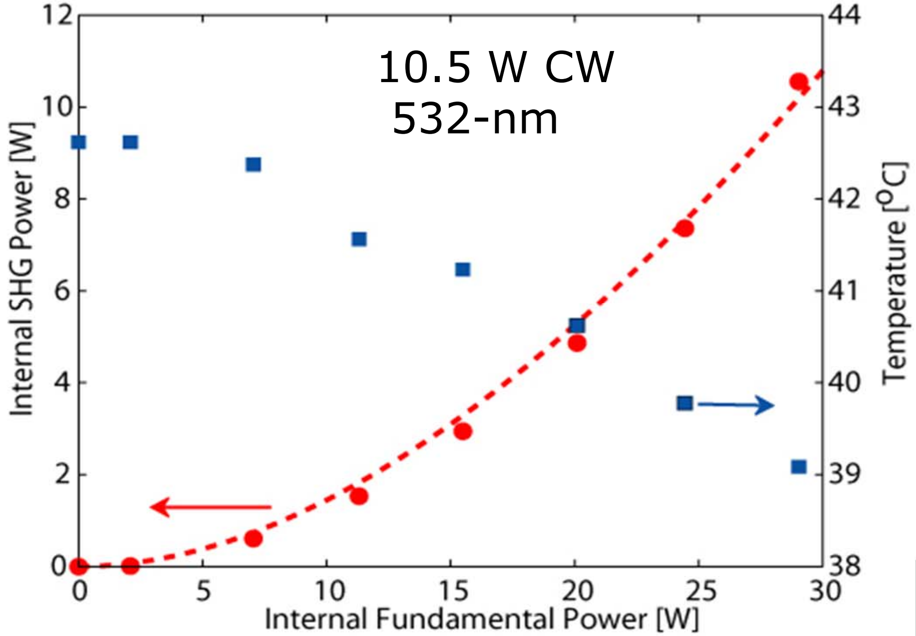
10 W CW 532 nm SHG in VTE PPSLT

Stoichiometry control in PPSLT by VTE
- increased photoconductivity
- reduced photorefraction
- eliminated GRIIRA
- visible absorption ~ 0.1 %/cm

Estimate ~100 W of CW SHG before material problem





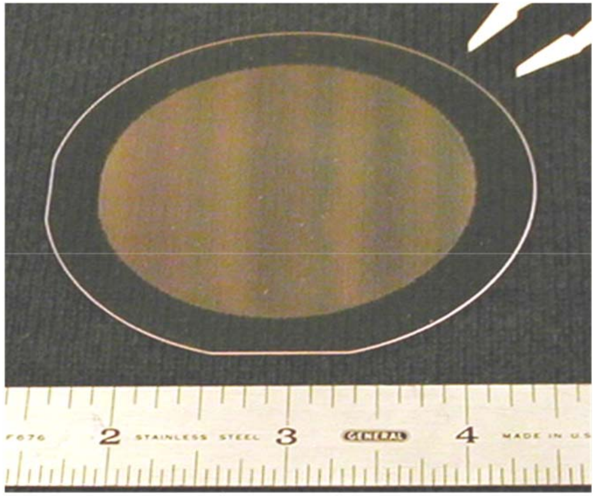


High energy conversion - tilted PP-samples

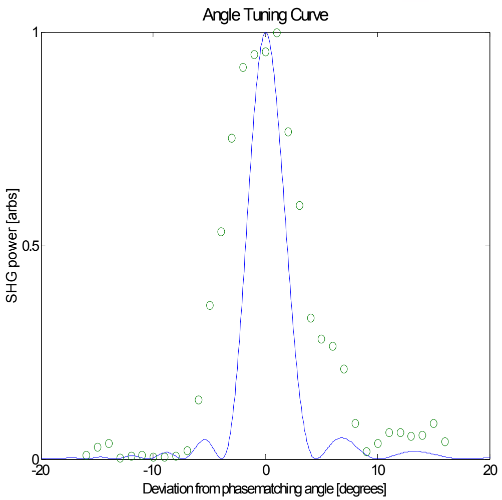
Damage threshold ~ 1 J/cm^2 @ 10 ns in LT
⇒ scaling to large fluence requires large apertures

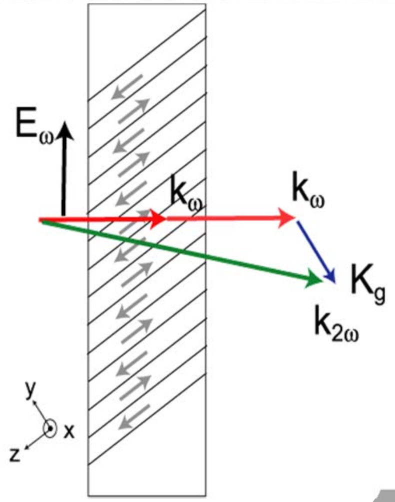
Large wafers commercial (100 mm)
⇒ scalable pulse energy
(100 mm wafer ~ 100 J @ 10 ns)

Rotated Y-cut SAW lithium tantalate



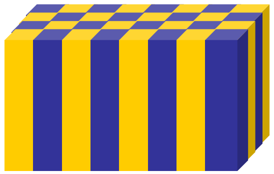
Surface period = 41 μm
thickness = 180 μm (after polishing)
4th order
4 periods total
PSH = 14.5μW
PFH = 6.2W
(100kHz, 150ns)

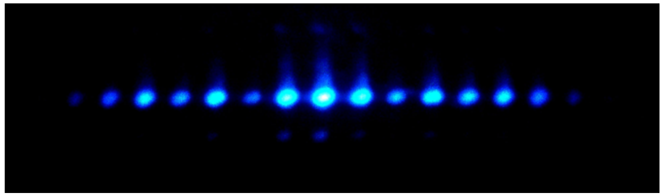


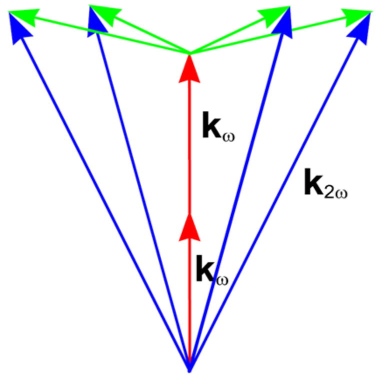


Nonlinear photonic crystals

Periodic variation in $\chi^{(2)}$ but no variation in $\chi^{(1)}$





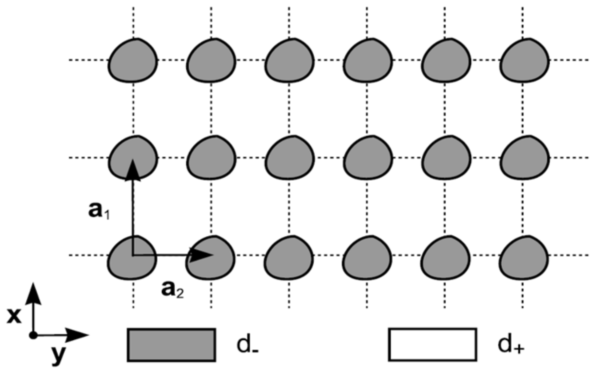


+ Multiple wavelengths
+ Simultaneous phase matching processes
+ Broader bandwidth in non-collinear SHG

- Low efficiency for non-collinear SHG

2D SH generator

Real lattice

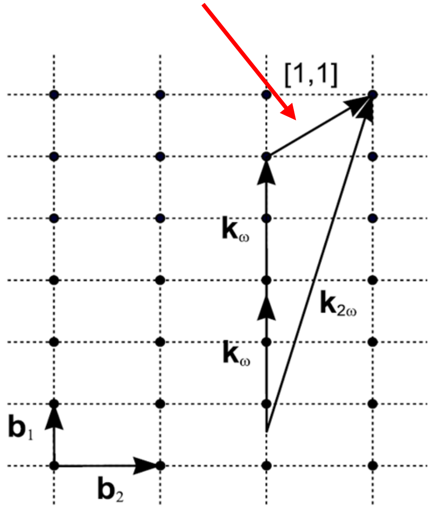


Reciprocal lattice

$$\mathbf{b}_1 = \frac{2\pi}{a_1}\hat{\mathbf{x}}$$

$$\mathbf{b}_2 = \frac{2\pi}{a_2}\hat{\mathbf{y}}$$

Grating vector



[m,n]

Phase-matching condition:

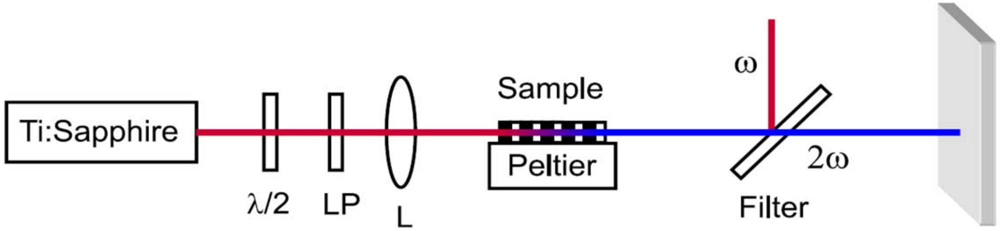
$$\vec{K}_{mn} - \vec{k}_{2\omega} + 2\vec{k}_{\omega} = 0$$

$$\vec{K}_{mn} = mb_1 + nb_2$$

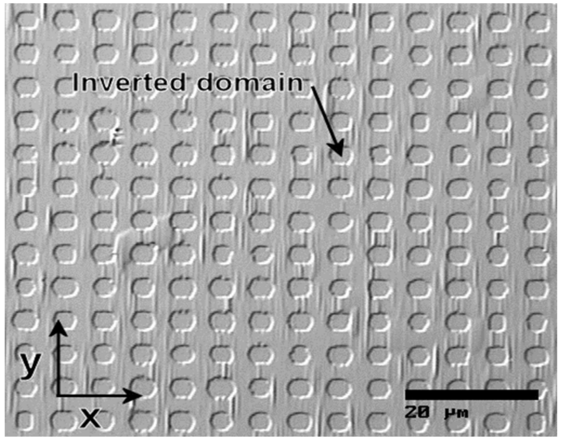
Walk-off angle 2θ between fundamental and second harmonic:

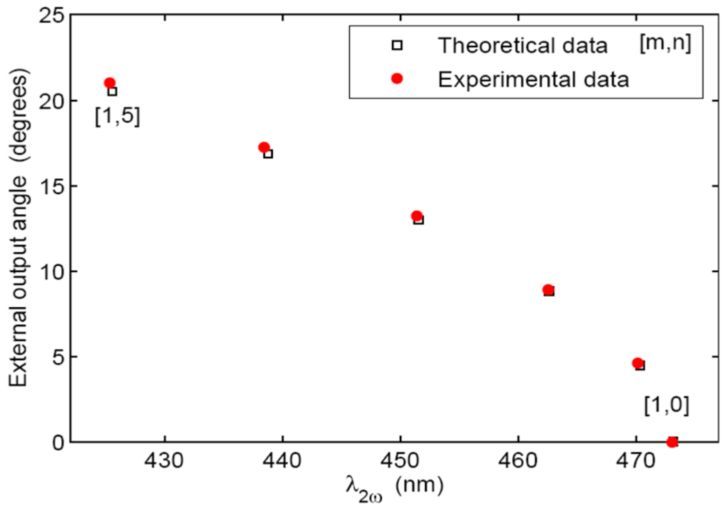
$$\frac{\lambda_{2\omega}}{n_{2\omega}} = \frac{2\pi}{|\mathbf{K}_{mn}|}\sqrt{\left(1 - \frac{n_\omega}{n_{2\omega}}\right)^2 + 4\frac{n_\omega}{n_{2\omega}}\sin^2\theta}$$

CW SHG in 2D PPKTP

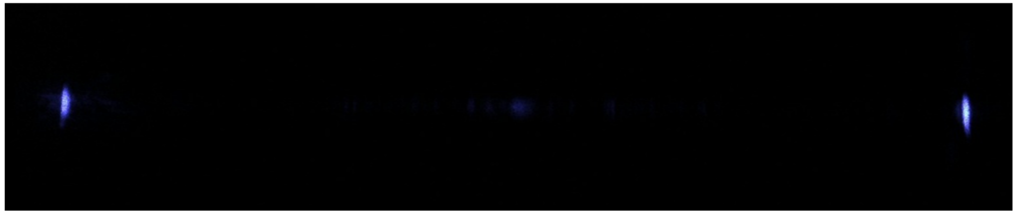


6.09 x 6 μm²





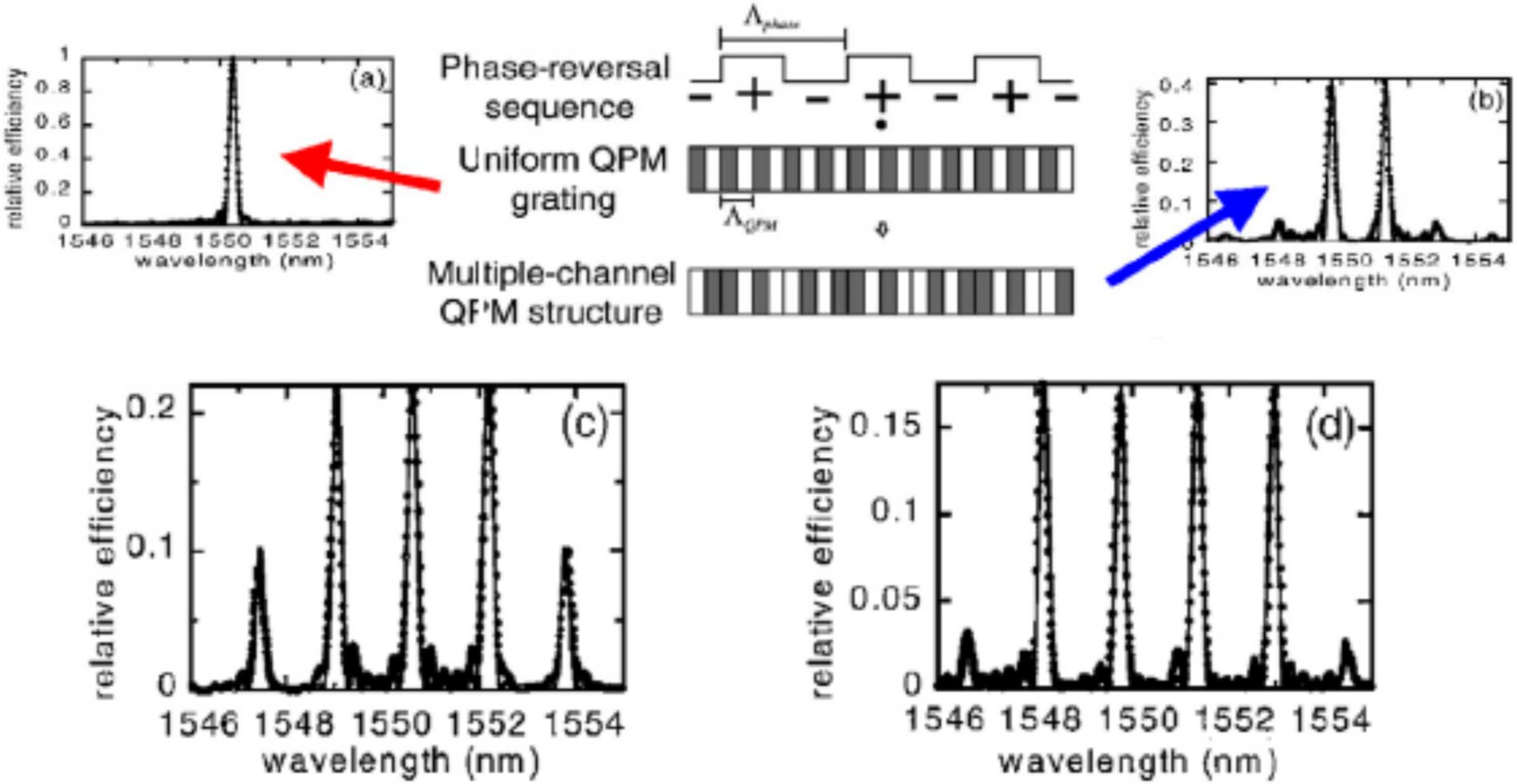
Patterned area: 8x4 mm²
Domain depth: 400μm



Spectral Tailoring

spatially modifying the grating

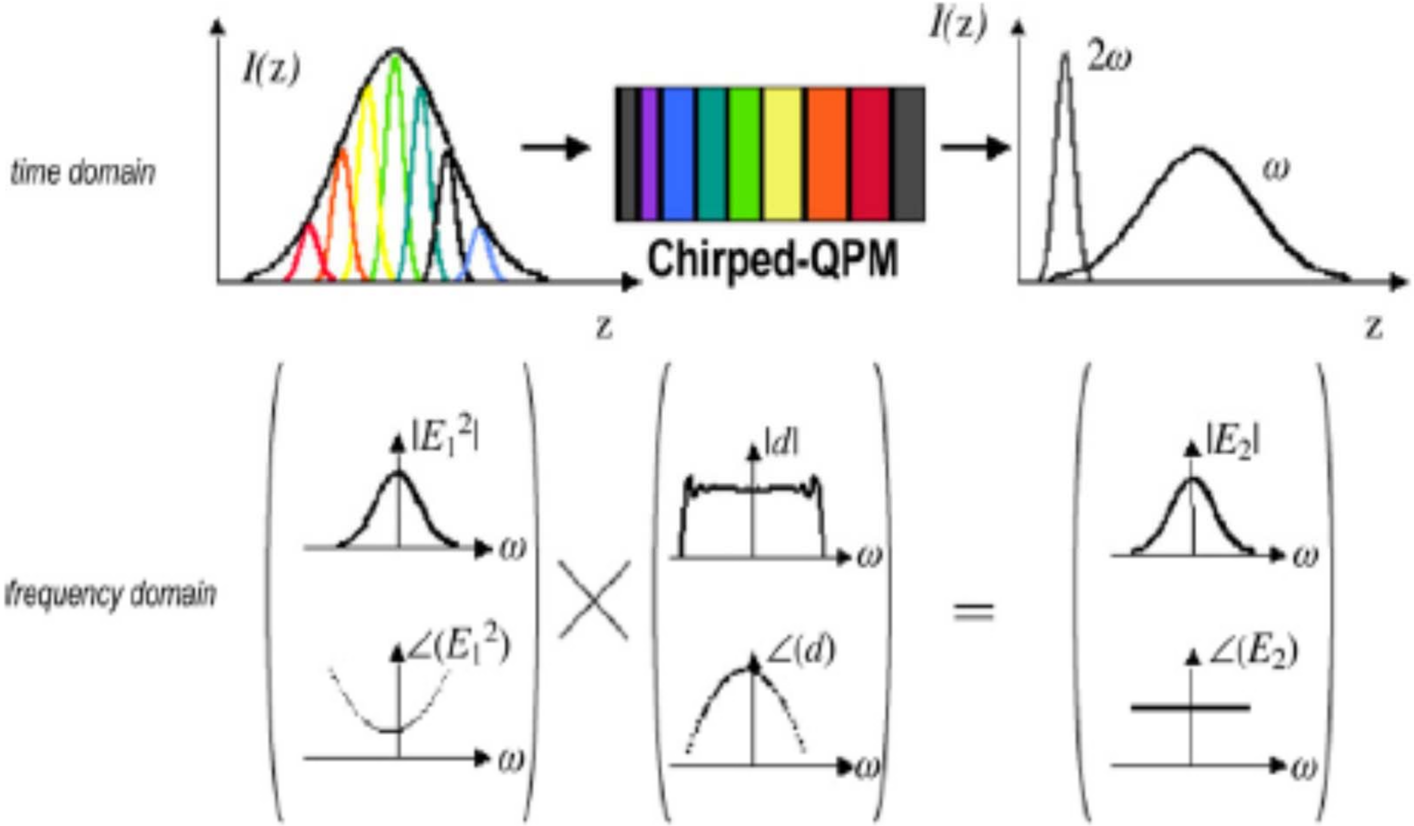
Fourier synthesis used to get the frequency response



Bortz, Stanford University

fs-manipulation with chirped gratings

SH conversion with chirped pulse



Each spectral component is phasematched in a different section of the grating

Summary – part one

- Second order nonlinear optics extends the possibilities for lasers – from the UV to the mid-IR and the THz region
- Quasi-phase matching adds flexibility to nonlinear optics
  - Spectral, spatial, temporal shaping of light
- The material aspects have to be considered