



The Abdus Salam
International Centre
for Theoretical Physics



2443-8

Winter College on Optics: Trends in Laser Development and Multidisciplinary Applications to Science and Industry

4 - 15 February 2013

Nonlinear optics - introduction

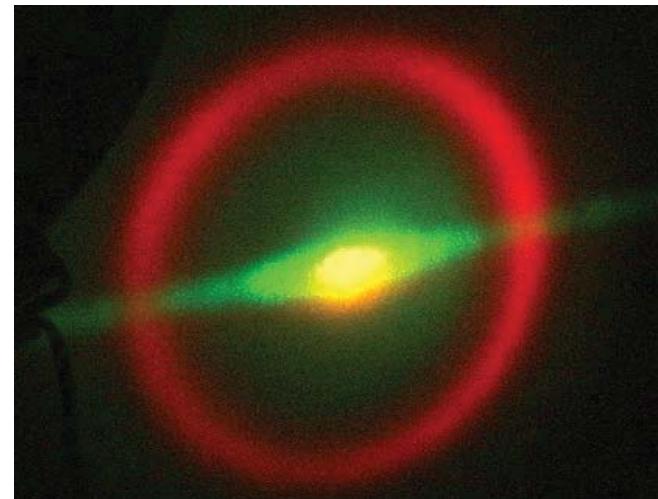
F. Laurell
KTH
Sweden



Nonlinear optics

introduction

Fredrik Laurell

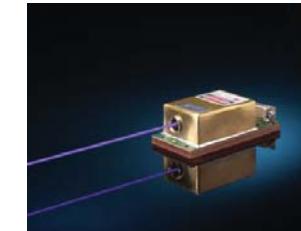


Department of Applied Physics
KTH – Royal Institute of Technology
Stockholm, Sweden

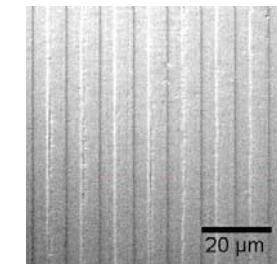


Our research profile

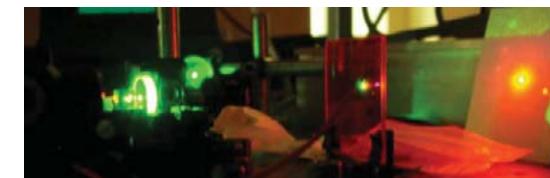
Lasers
Nonlinear optics
Optical materials and novel components



Investigate fundamental optical
and material physics problems
Build efficient diode-pumped lasers and light sources
Use them in applications



Start-ups and industrial collaboration





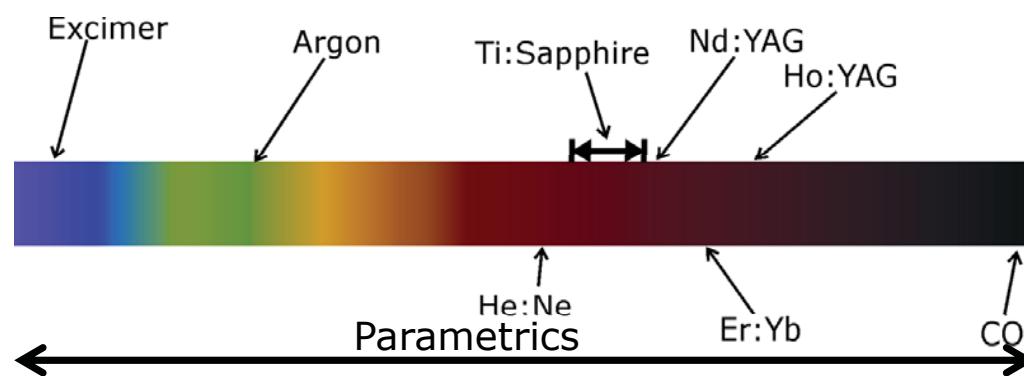
Outline

- 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



Motivation - Why nonlinear optics?

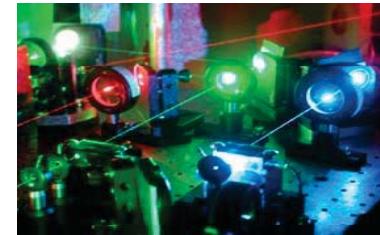
- Second order effects gives a natural extension of lasers
 - Reach the UV and the mid-IR + THz
 - Facilitates tuning
 - Many applications
- Efficient amplifiers – low thermal load and high gain
- Third order nonlinear phenomena
 - Kerr lensing, Self-phase modulation
 - Cascading effects



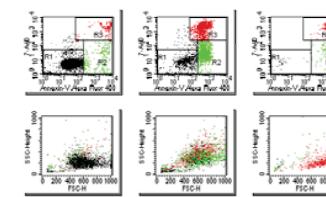
Motivation - upconversion



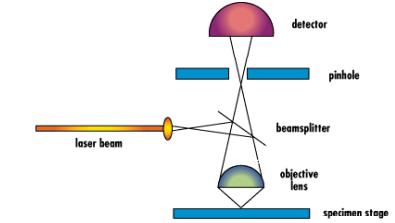
- Needs for visible and UV lasers
- Technology
- Material issues



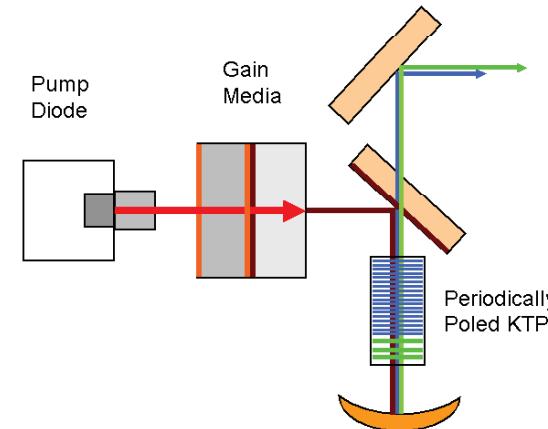
Laser display



Bio-analysis



Confocal Microscopy



Wafer inspection



Printing



The birth of nonlinear optics

VOLUME 7, NUMBER 4

PHYSICAL REVIEW LETTERS

AUGUST 15, 1961

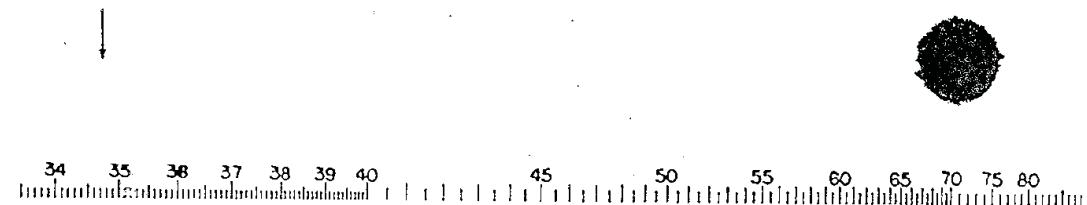
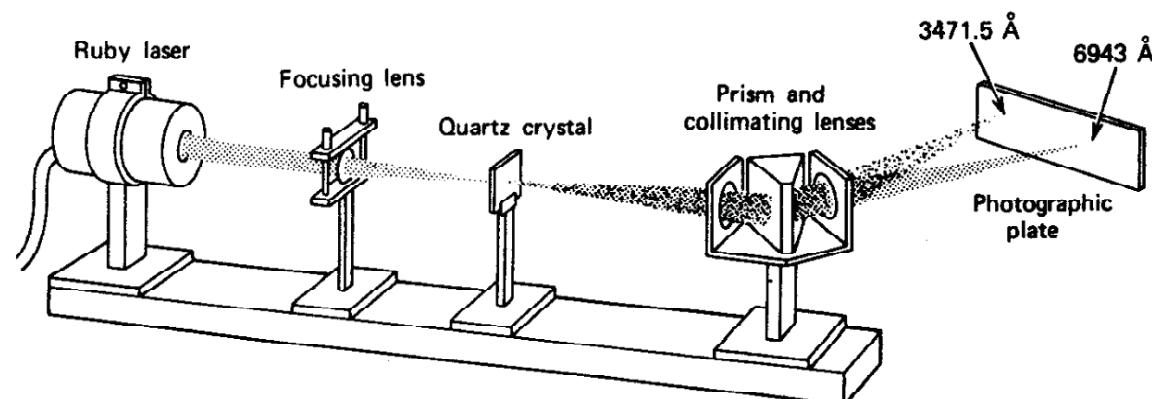


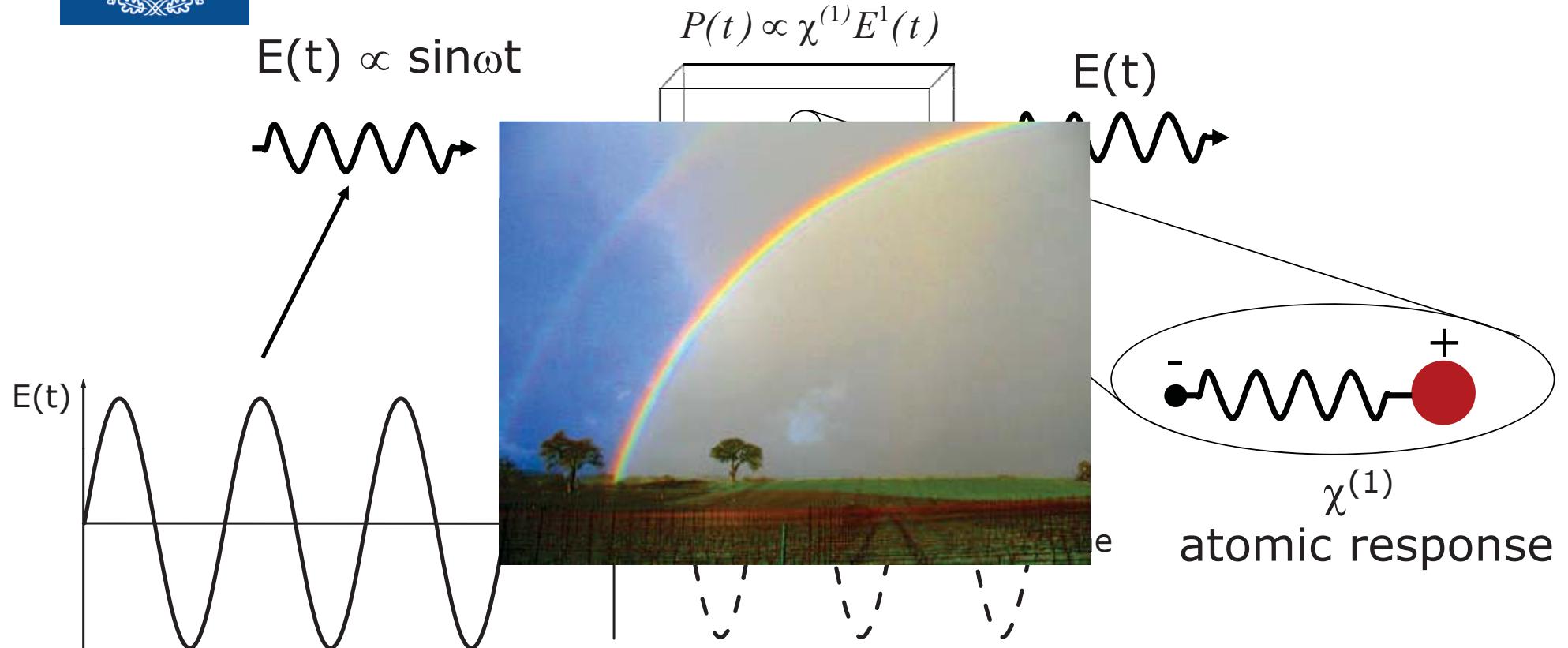
FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 Å. The arrow at 3472 Å indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 Å is very large due to halation.





Atomic response on electromagnetic radiation

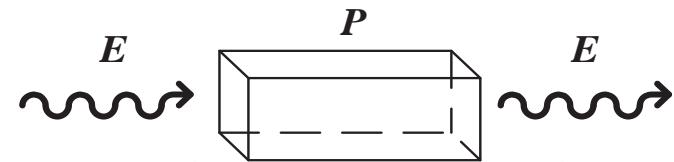
Linear optical susceptibility



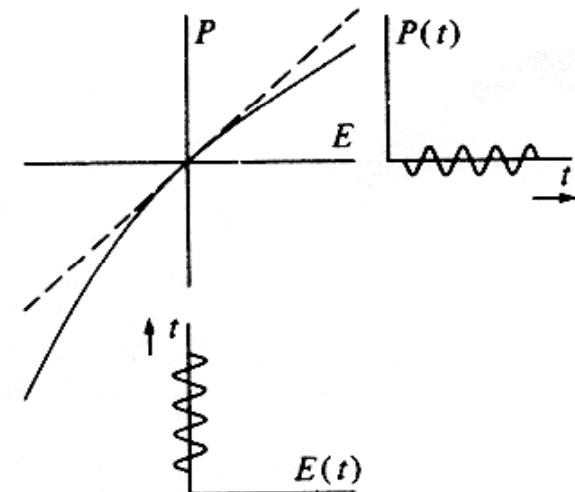
Nonlinear polarisation



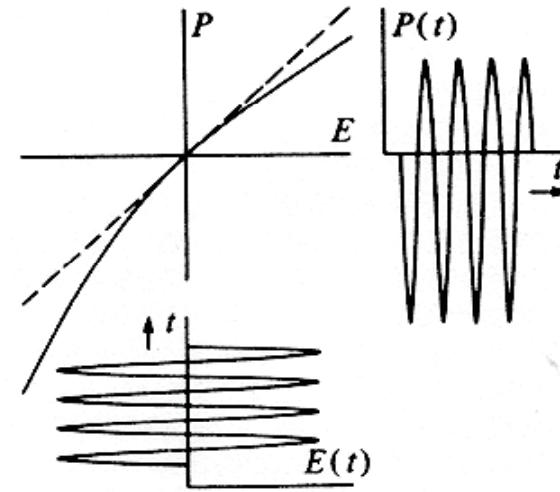
Field induces a polarisation
Driver for a new wave



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



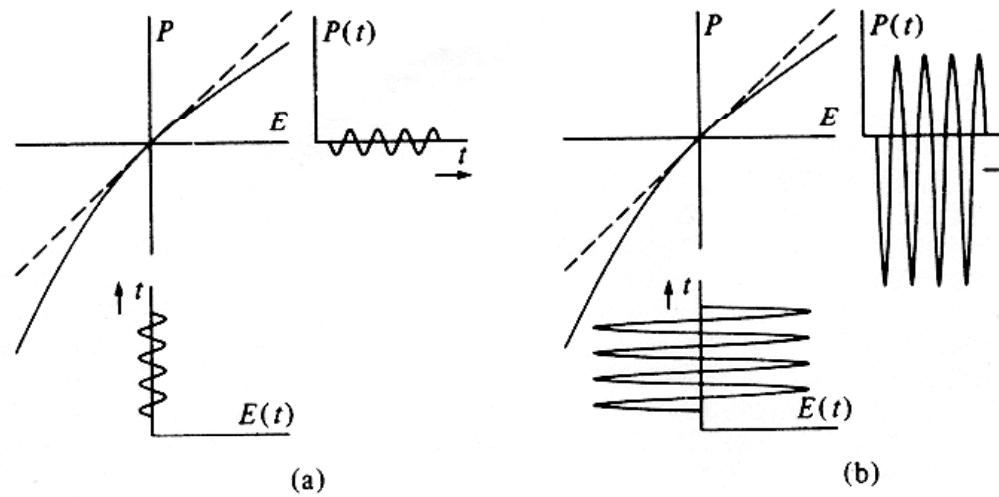
(a)



(b)



2nd order effects in noncentrosymmetric materials – three wave mixing



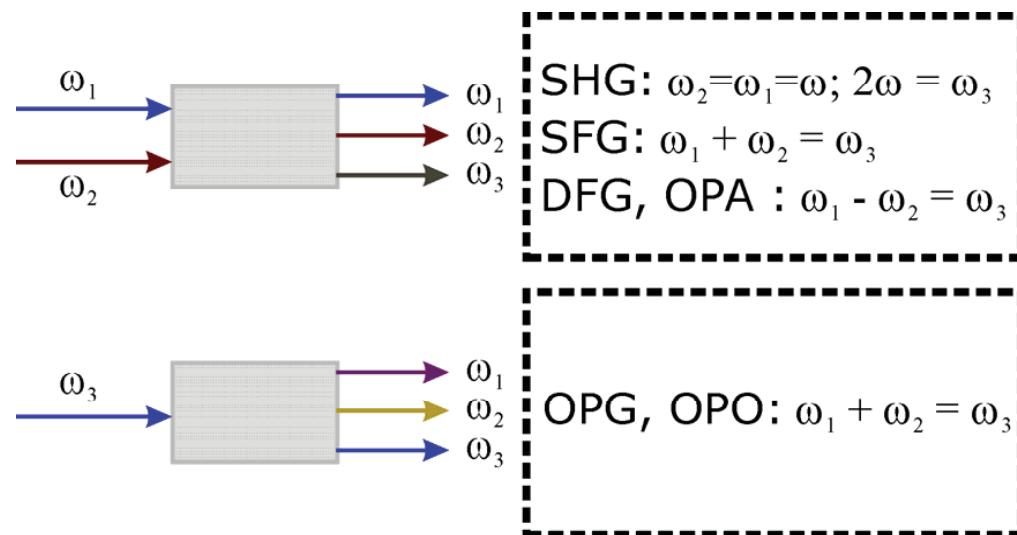
$$P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 (\chi^{(2)} E^2 + \dots) = P_L + P_{NL}$$

$$\begin{aligned}2\omega_1 &= \omega_3 \\2\omega_2 &= \omega_3 \\\omega_1 \pm \omega_2 &= \omega_3 \\\omega_1 &= \omega_2 + \omega_3\end{aligned}$$

The second order nonlinear processes



$$P^{(2\omega)} \propto \epsilon_0 \chi^{(2)} E_1 E_2$$



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



Frequency conversion

$$P = \varepsilon_0 \chi^L E + 2\varepsilon_0 d_{eff} EE \quad d_{il} \equiv \frac{1}{2} \chi_{ijk}^{(2)}$$

Maxwell's equations

=>

Nonlinear wave equation

=>

Three coupled equations

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

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

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



Solution to the coupled wave equation

Example: Sum-frequency generation

$$\frac{\partial E_1}{\partial z} = i \frac{\omega_1 d_{eff}}{cn_1} E_2 E_3 \exp(i\Delta kz))$$

plane-waves and
lossless media

$$\frac{\partial E_2}{\partial z} = i \frac{\omega_2 d_{eff}}{cn_2} E_1 E_3^* \exp(-i\Delta kz))$$

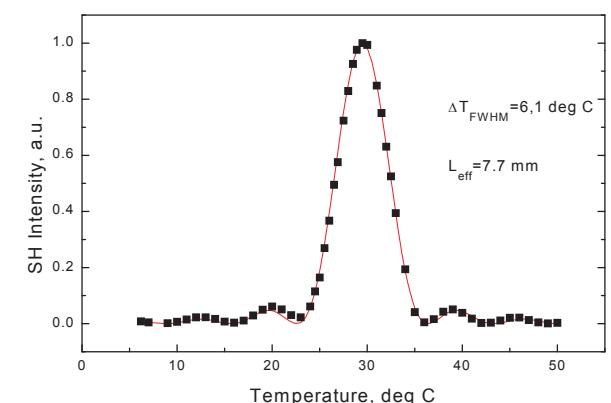
$$\omega_1 = \omega_2 + \omega_3$$

$$\frac{\partial E_3}{\partial z} = i \frac{\omega_3 d_{eff}}{cn_3} E_1 E_2^* \exp(-i\Delta kz))$$

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

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

$$I_1(L) = \frac{2\omega_1^2 d_{eff}^2}{\epsilon_0 c^3 n_1 n_2 n_3} I_2 I_3 L^2 \text{sinc}^2(\Delta k L / 2)$$





General solution to 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

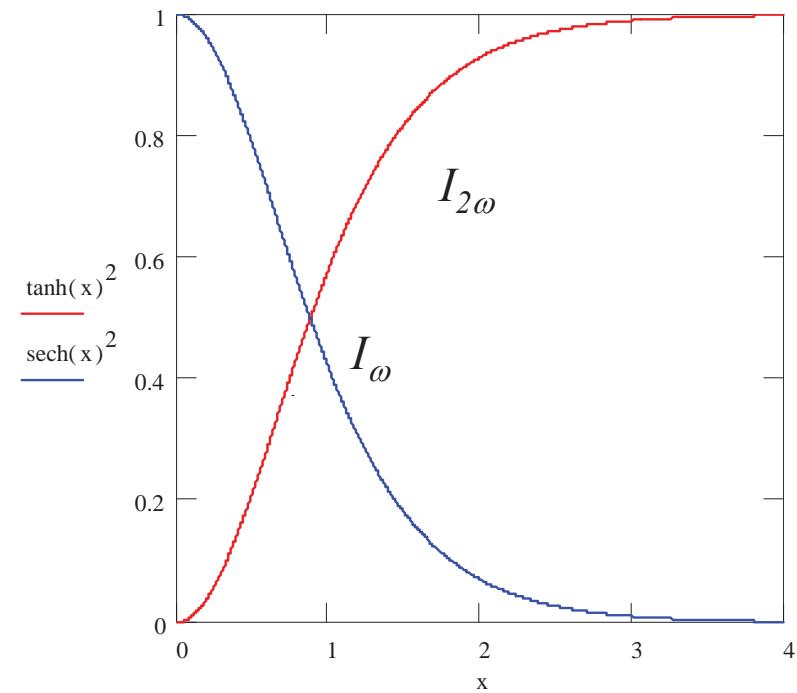
$$I_{2\omega}(L) = I_\omega(0) \tanh^2(\Gamma L)$$

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

where $\Gamma = \frac{4\pi d_{eff} \sqrt{I_\omega(0)}}{\sqrt{2c n_\omega^2 n_{2\omega} \epsilon_0 \lambda_\omega^2}}$

Conversion efficiency is defined as:

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

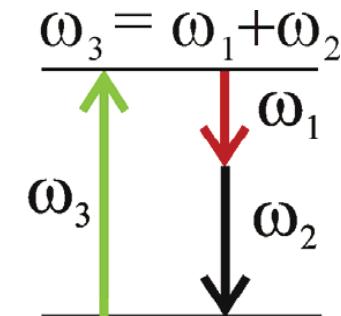




Physical constraints

- Energy conservation

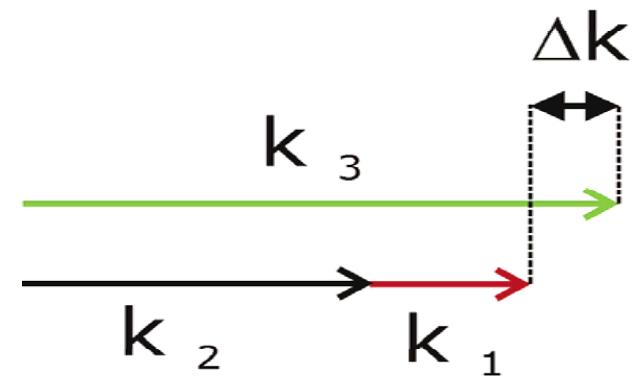
$$\omega_1 + \omega_2 = \omega_3$$



- Momentum conservation

$$\begin{aligned}\Delta k &= k_3 - k_2 - k_1 = \\ &= 1/c \times [n_3 \omega_3 - n_2 \omega_2 - n_1 \omega_1]\end{aligned}$$

n_i is the refractive index



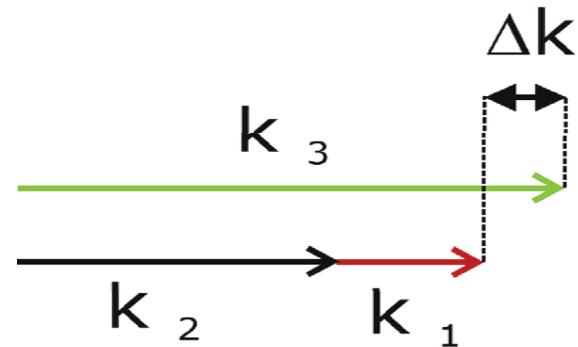
Coherence length: $L_c = \left| \frac{\pi}{\Delta k} \right|$

Phasematching



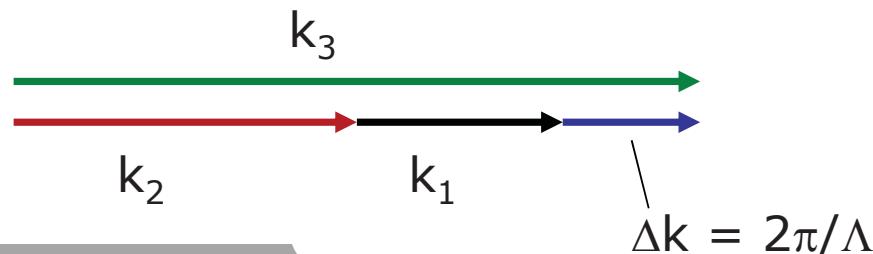
Momentum conservation

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

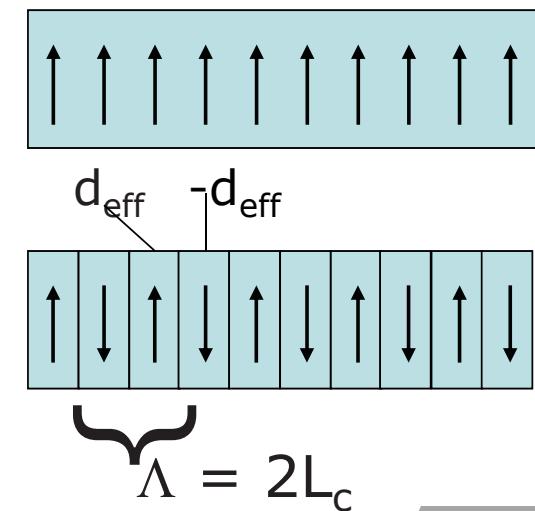


Birefringent phasematching $\Delta k = 0$

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

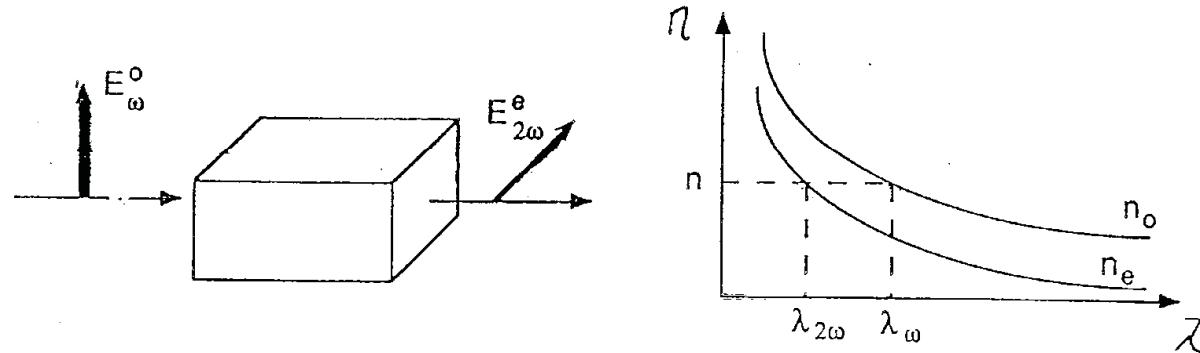


Modulation of the nonlinearity





Birefringent phasematching

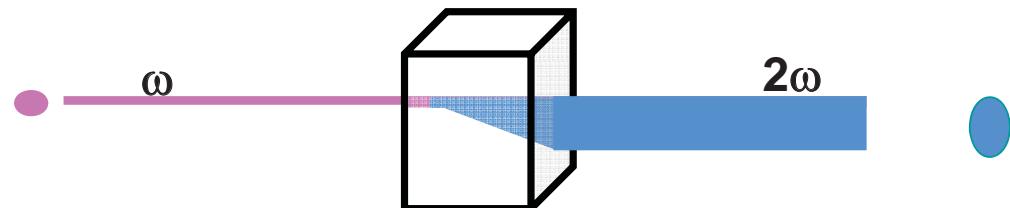


- need birefringent crystal
- $\chi^{(2)}$ must couple orthogonal polarizations
- temperature constraints
- cannot use full transparency range

Limited phasematching range

Walk-off – short interaction length

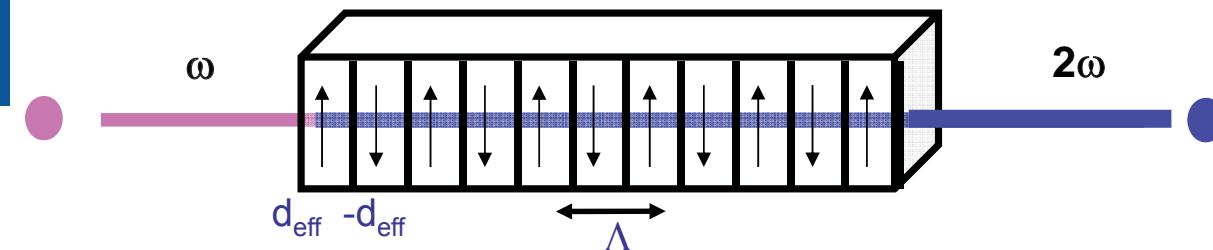
Low nonlinearity





Quasi-phasematching (QPM)

Proposed by Bloembergen, et al. 1962

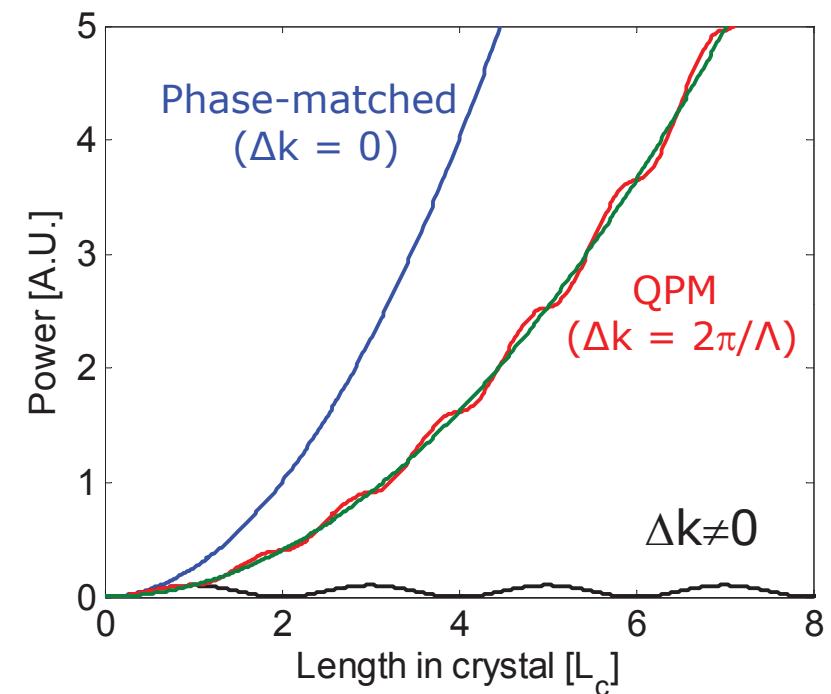


Periodic inversion of domains in ferroelectrics

Modulation of the nonlinear susceptibility
to compensate the phase mismatch

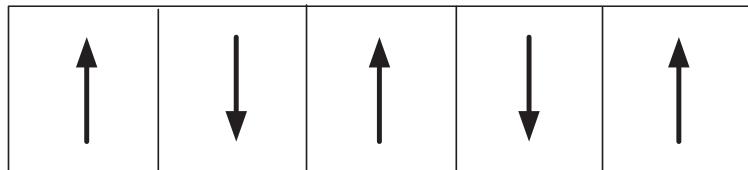
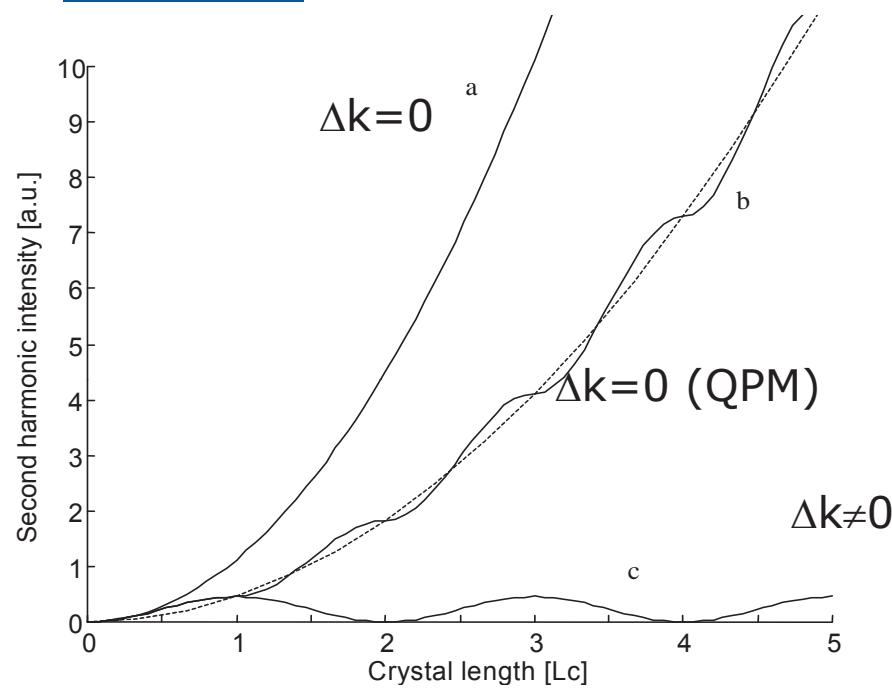
- Arbitrary wavelength within transparency
- Walk-off free interaction – long lengths
- Highest nonlinearity accessible

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





Quasi-phase matching



Advantages:

- Access to highest nonlinear coefficient
- Walk-off free interaction
- Phasematching over entire transparency
- Engineering of phasematching condition

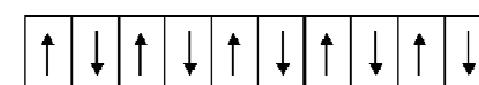
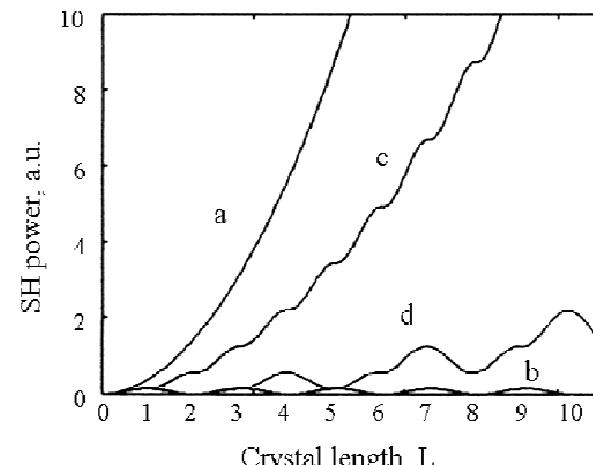
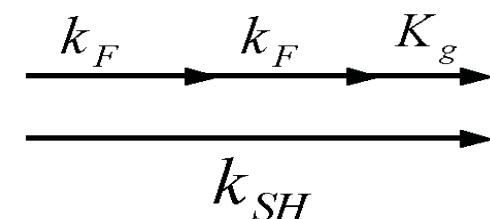
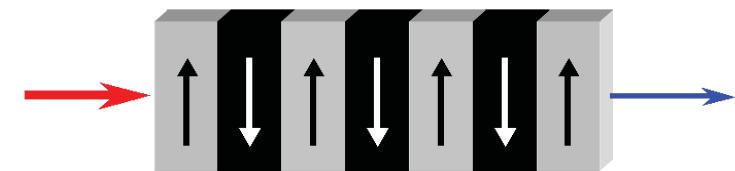
Disadvantages:

- Extra processing steps (time = cost)
- Parasitic processes



Quasi-phasematching (QPM)

- Accumulated phase mismatch prevented by a spatical modulation of the sign of the nonlinear susceptibility
- In ferroelectrics, periodically alter domain orientation by electric field poling
- First-order QPM: $m = 1$
 d coefficient reversed each L_c
- m -order QPM:
 d coefficient reversed each mL_c





The main advantages of QPM

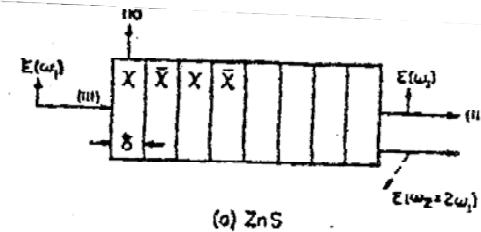
- Phasematching at any wavelength within transparency
- Free choice of polarization of interacting waves
- Largest nonlinear coefficient accessible, d_{33}
- Noncritical phasematching, walk-off free interaction
- Reduced sensitivity to the photorefractive effect



Implementations of QPM

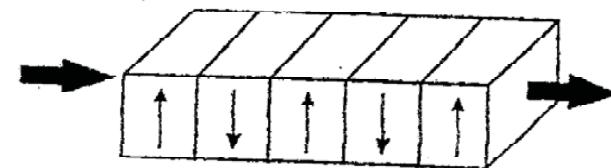
- Stacked plates with alternating layers rotated.

[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]

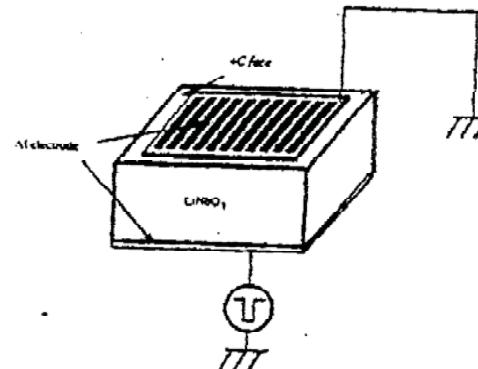


FIG. 1. Schematic of applying voltage for periodically domain inversion.



Types of phasematching

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

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

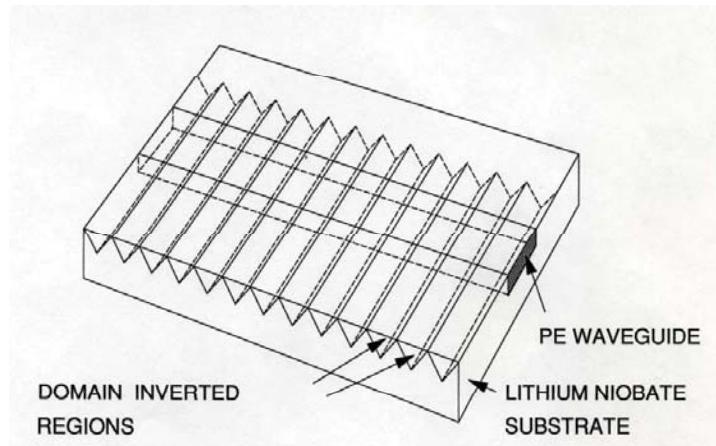
In **Type II SHG**, two photons having orthogonal polarizations will combine to form one photon with double the frequency and extraordinary polarization

In general to utilize **Type O** interactions a Quasi-phase-matching crystal type will be required, for example periodically poled lithium niobate (PPLN).

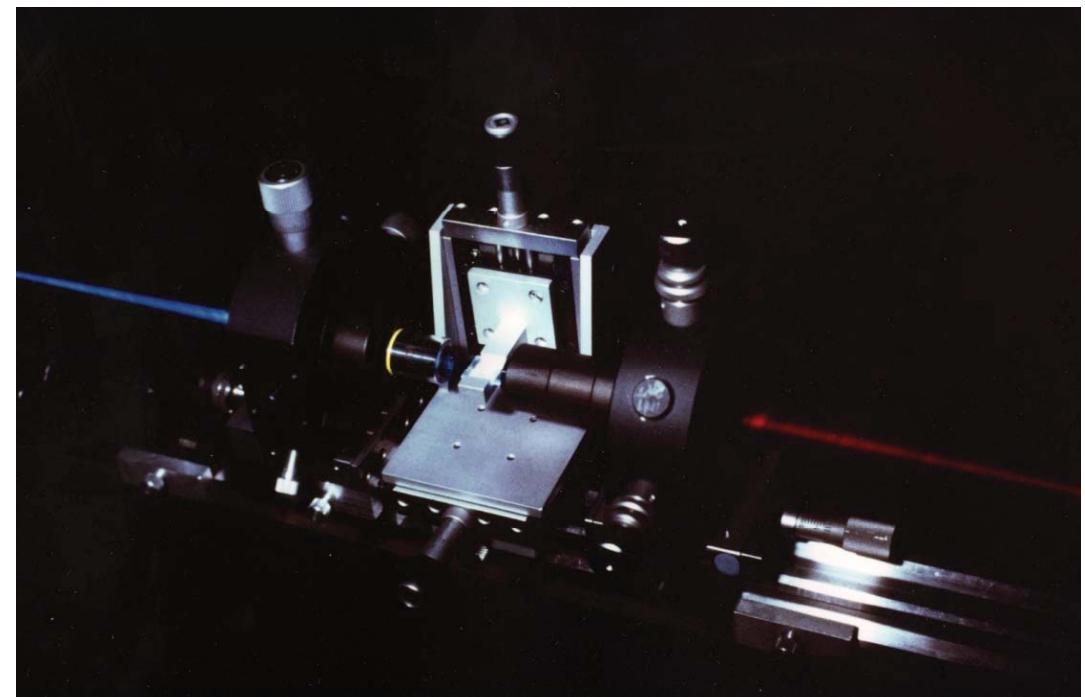
Quasi-phase matching (QPM)



First efficient demonstration with LiNbO_3 waveguides



Domain structure fabricated
by diffusion techniques



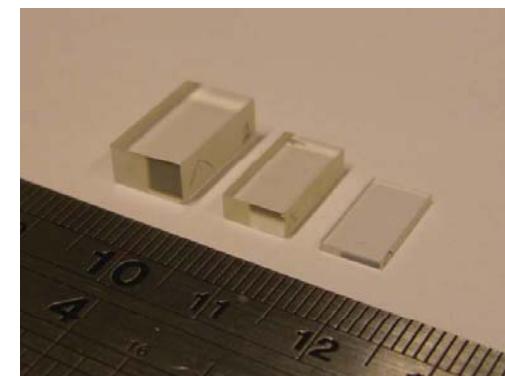
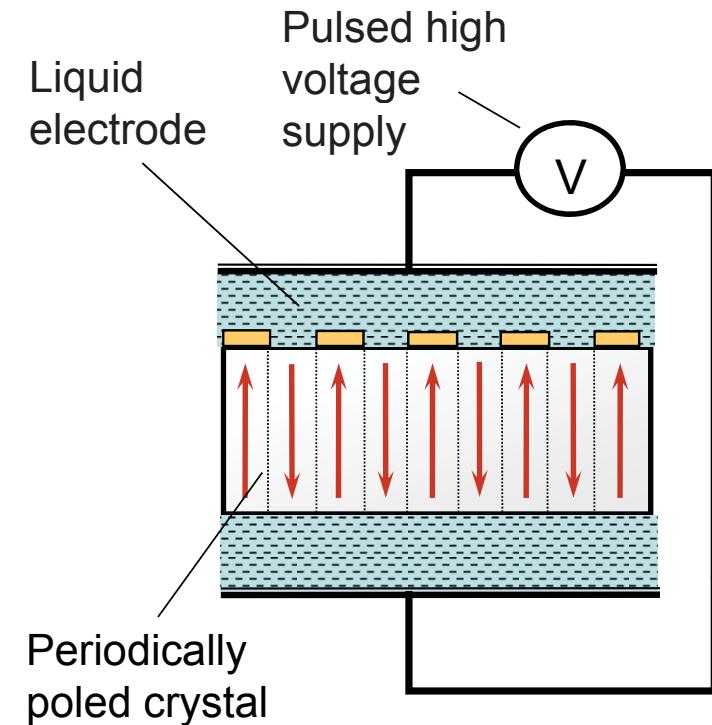
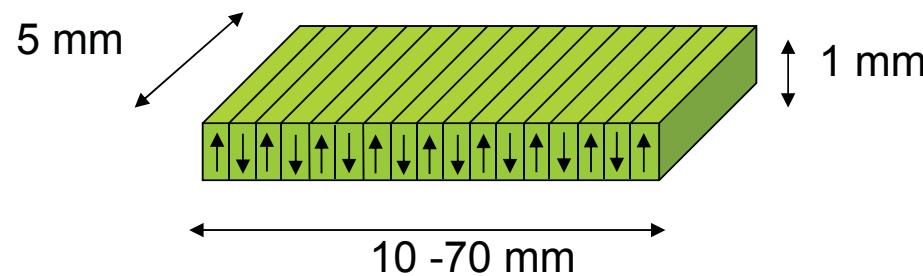
Blue light generated with AlGaAs diode laser

Electric field poling of bulk crystals



- LiNbO₃
- LiTaO₃
- KTiOPO₄

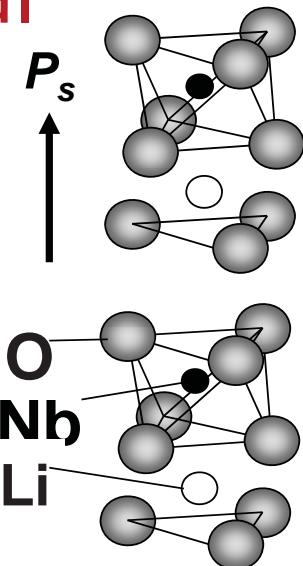
Bandwidth tailored for specific interaction
Damage resistant and efficient material



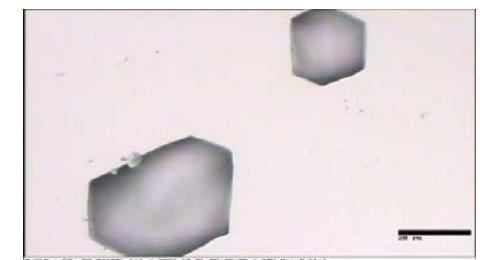


LiNbO₃, the first QPM material

- 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



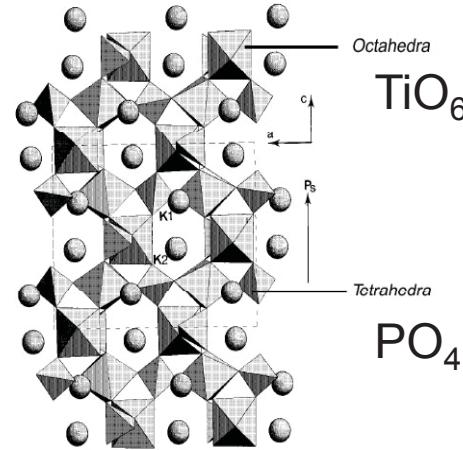
Trigonal 3m



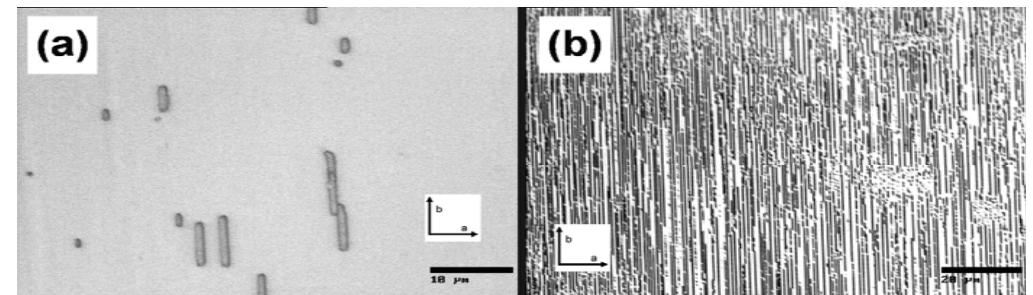
Solution MgO-doped Lithium niobate – optical damage “resistant”
suitable for waveguide fabrication
commercially available

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

Flux grown KTiOPO₄



Domain morphology



High nonlinearity

High resistance to optical damage

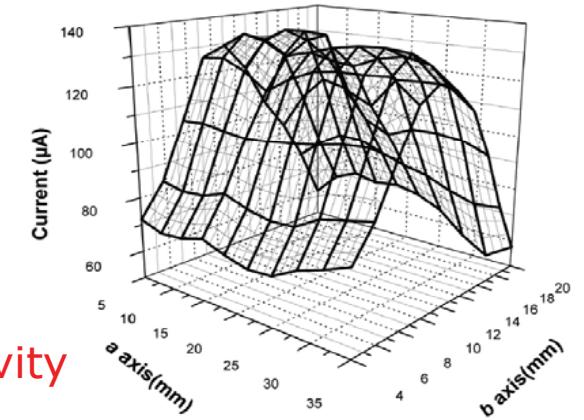
Low coercive field (2 kV/mm)

dense gratings, "thick" samples (3mm)

Difficult to grow – 1 inch wafers

Non-stoichiometric - inhomogeneous – highly varying conductivity

Expensive





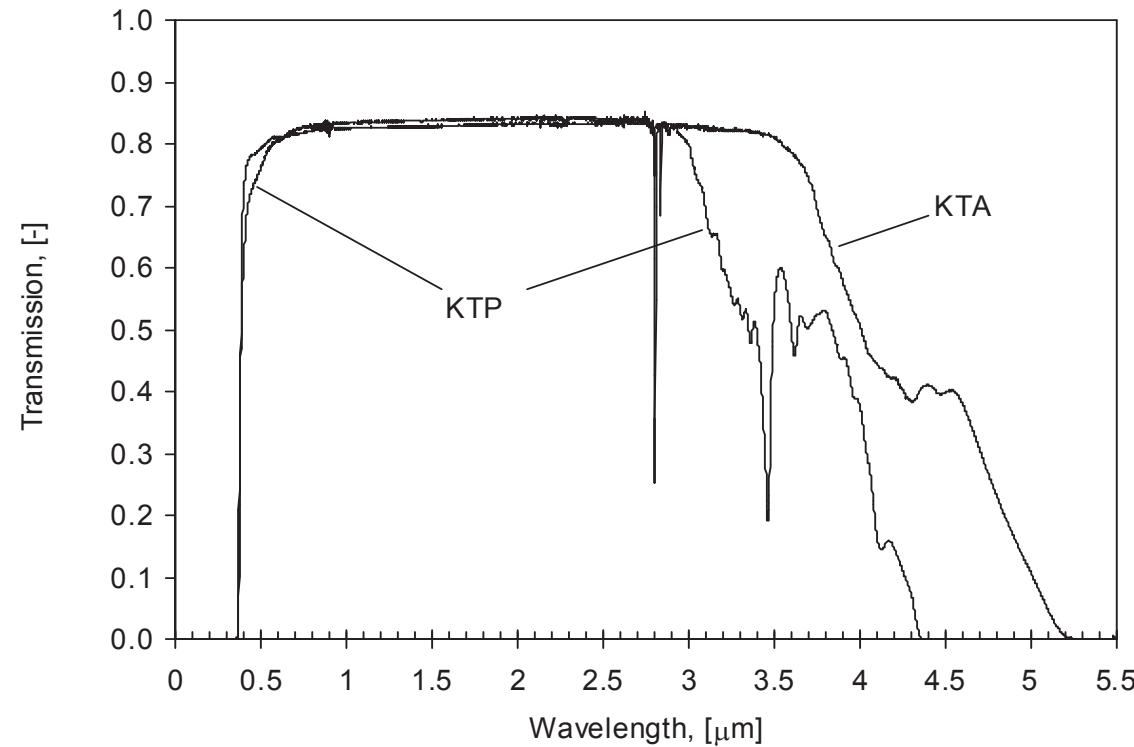
Requirement for NLO materials for frequency conversion

- **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!



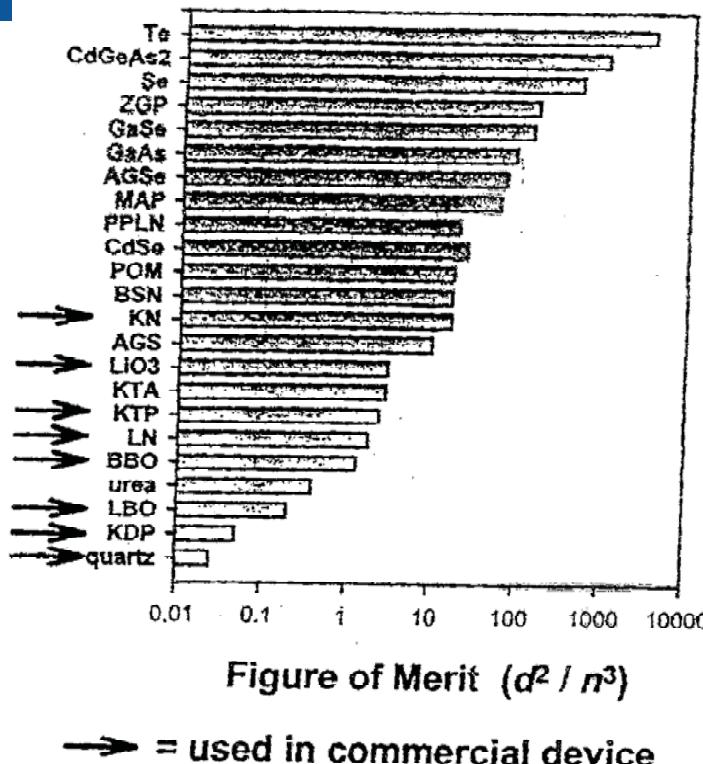
KTiOPO₄ Transparency



LiNbO₃
Transparency 0.35 - 5.4 μm



Figure of merit for nonlinear materials



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

Units of $d_{eff} \sim \text{pm/V}$

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

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

Comparison of PP crystals



- KTiOPO₄ family (KTiOPO₄, RbTiOPO₄, KTiOAsO₄, RbTiOAsO₄)
- LiNbO₃ family
 - stoichiometric LiTaO₃ (SLT)
 - 1-5 mol% MgO doped stoichiometric LiTaO₃ - (MgO:SLT)
 - 1-5 mol% MgO doped stoichiometric LiNbO₃ - (MgO:SLN)
- KNbO₃

Low coercive field (<5 kV/mm)

Higher optical damage threshold

Figure of merit for SHG of 1064 nm

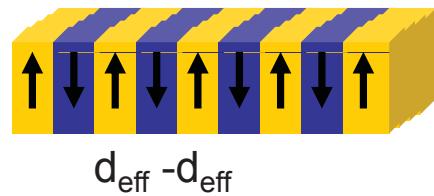
$$\frac{d^2}{n_{\omega_1} \cdot n_{\omega_2} \cdot n_{\omega_3}}$$

	LN	KN	KTP	LT
FOM	61.1	38.8	33.7	18.8



Artificially structured ferroelectrics

1D – gratings

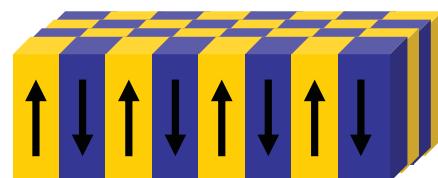


Domain gratings with μm periodicity

Frequency converters: from UV to mid-IR

Domain gratings with nm periodicity

New devices:
electrically controlled Bragg reflectors, BSHG, BOPO,



2D structures - Nonlinear photonic crystals

Second-harmonic generation



- Energy conservation:

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

- Momentum conservation:

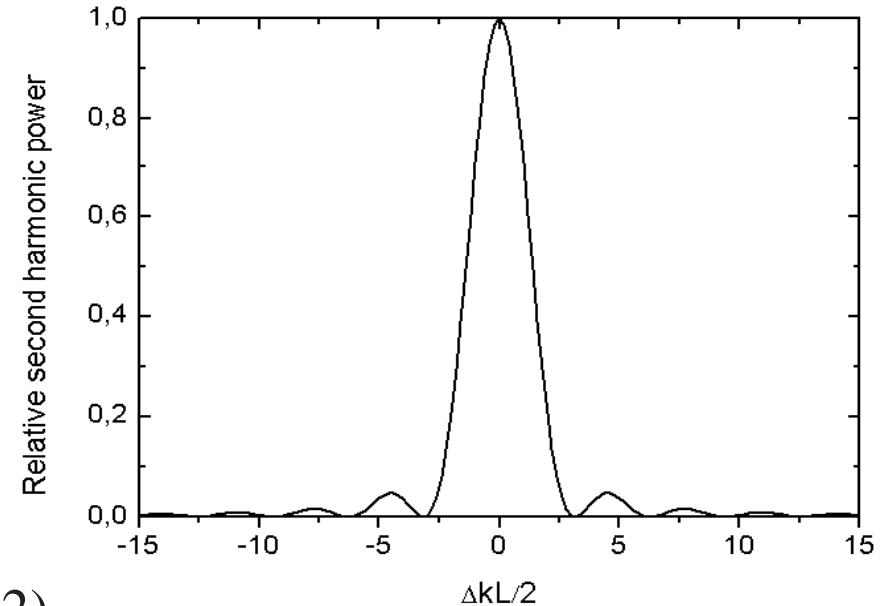
$$\Delta k = k_{SH} - 2k_F$$

- SH power for Gaussian beam:

$$P_{SH} = \left(\frac{2\omega_F^2 d_{eff}^2 k_F P_F^2}{\pi n_F^2 n_{SH} \epsilon_0 c^3} \right) L h(B, \xi) \operatorname{sinc}^2(\Delta k L / 2)$$

- Phasematching:

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



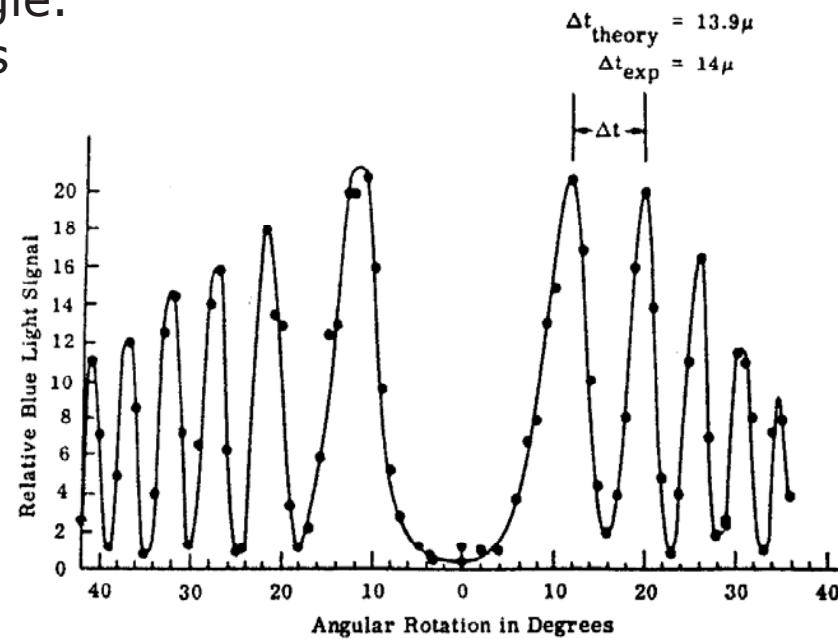
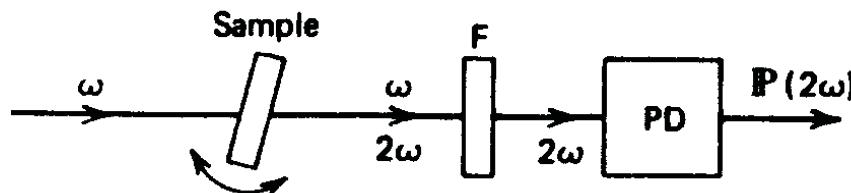
$$d_{eff}^{the} = \frac{2d_{il}}{m\pi}$$



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



Phasematching bandwidth



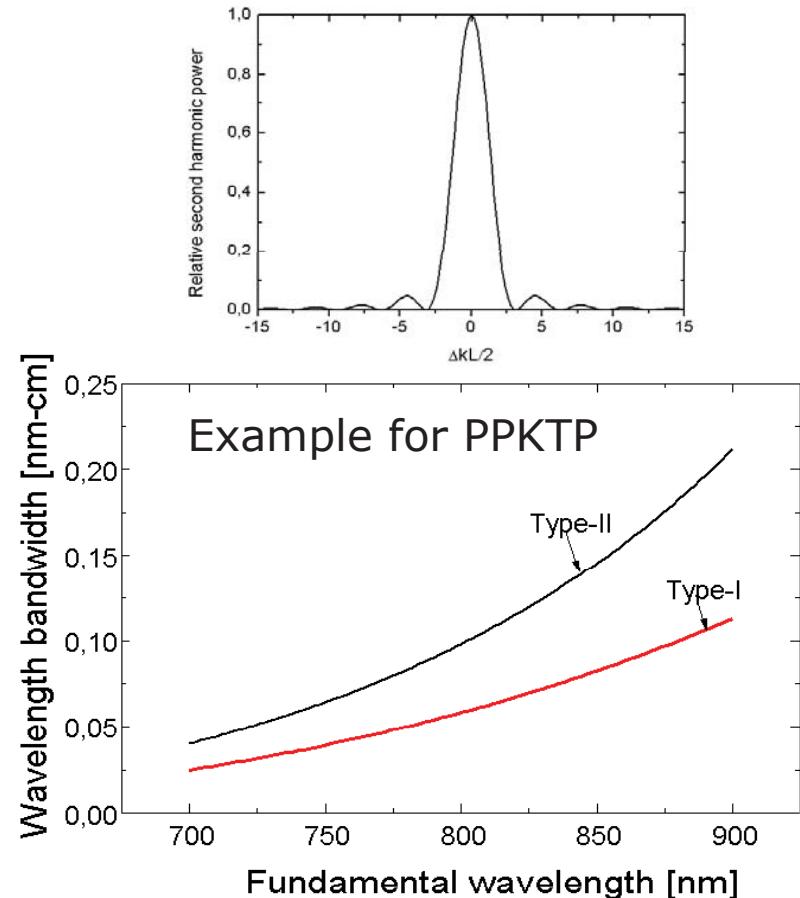
The mismatch Δk depends on λ , T and polarization of waves

- 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

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



Tuning characteristics



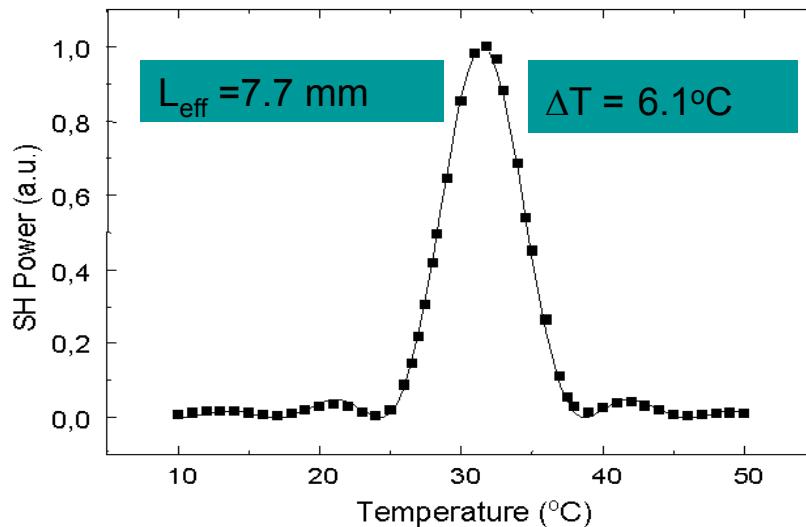
Quality of a PP structure: $L_{\text{eff}} \propto 1/\Delta T_{\text{FWHM}}$ or $L_{\text{eff}} \propto 1/\Delta \lambda_{\text{FWHM}}$

Temperature tuning

$$\lambda = 1064 \text{ nm}$$

Type-I SHG @ 532 nm

PPKTP: $\Lambda = 9.01 \mu \text{m}$, $L_{\text{physical}} = 8 \text{ mm}$

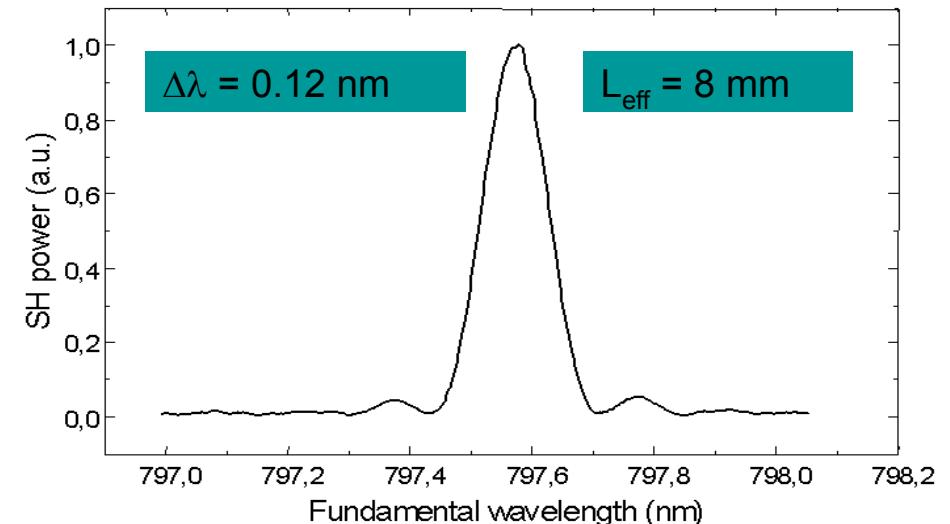


Wavelength tuning

$$\lambda = 797.6 \text{ nm}$$

Type-II SHG @ 398.8 nm

PPKTP: $\Lambda = 9.01 \mu \text{m}$, $L_{\text{physical}} = 8.5 \text{ mm}$





Effective QPM nonlinear coefficient

- Theoretical value of effective nonlinear coefficient:

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

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

- Experimental value of effective nonlinear coefficient:

$$d_{\text{eff}}^{\text{exp}} = \sqrt{\frac{\pi n_F^2 n_{SH} \epsilon_0 c^3}{2\omega_F^2 k_F L h(B, \xi)} \times \frac{P_{SH}}{P_F^2}}$$

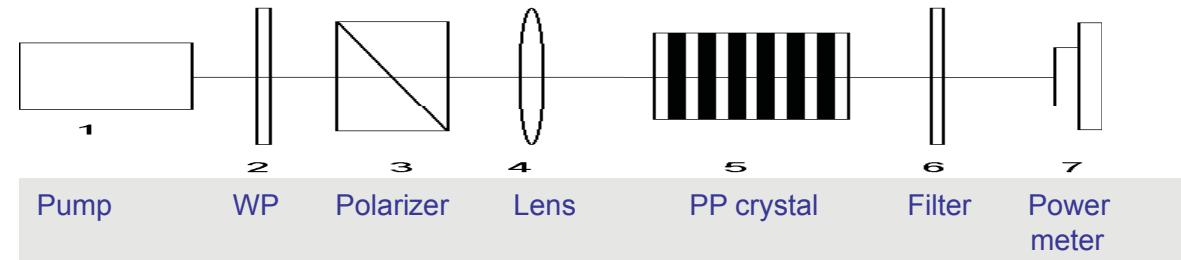
- Useful technique to screen quality of PP crystals



CW SHG

low power regime

Single-pass SHG set-up

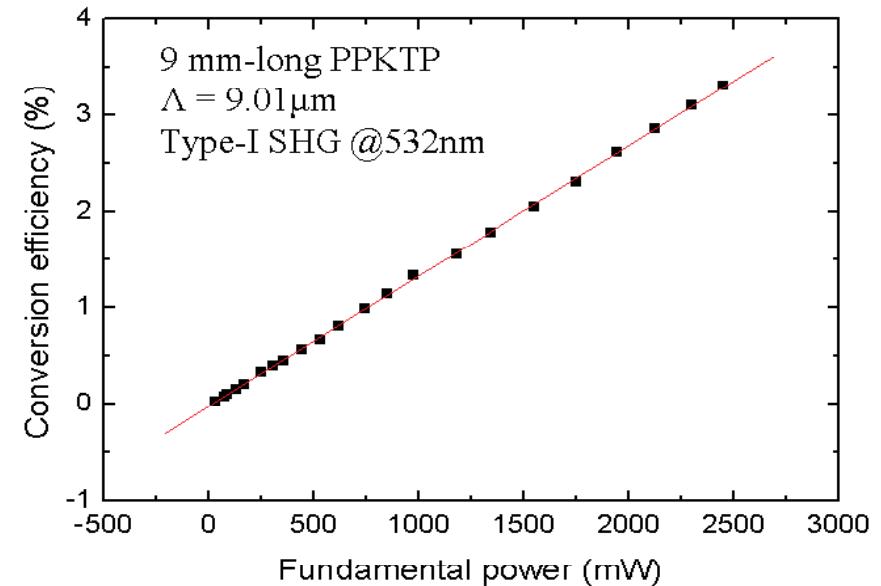


Power and conversion efficiency

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

Normalized conversion efficiency

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

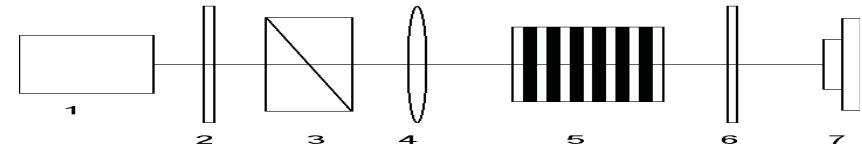




CW SHG

high power regime

Single-pass SHG set-up

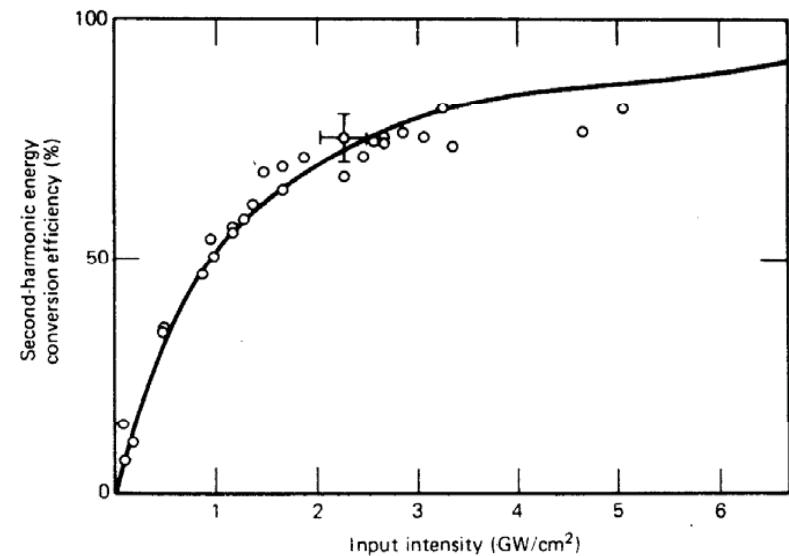


conversion efficiency

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

$$\Gamma = \frac{4\pi d_{eff} \sqrt{I_\omega(0)}}{\sqrt{2c n_\omega^2 n_{2\omega} \epsilon_0 \lambda_\omega^2}}$$

SHG in KDP crystal

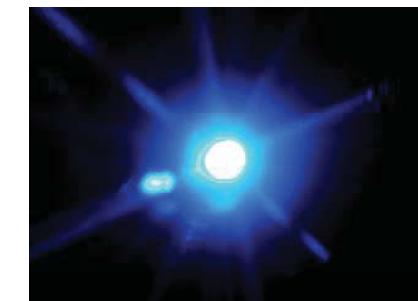
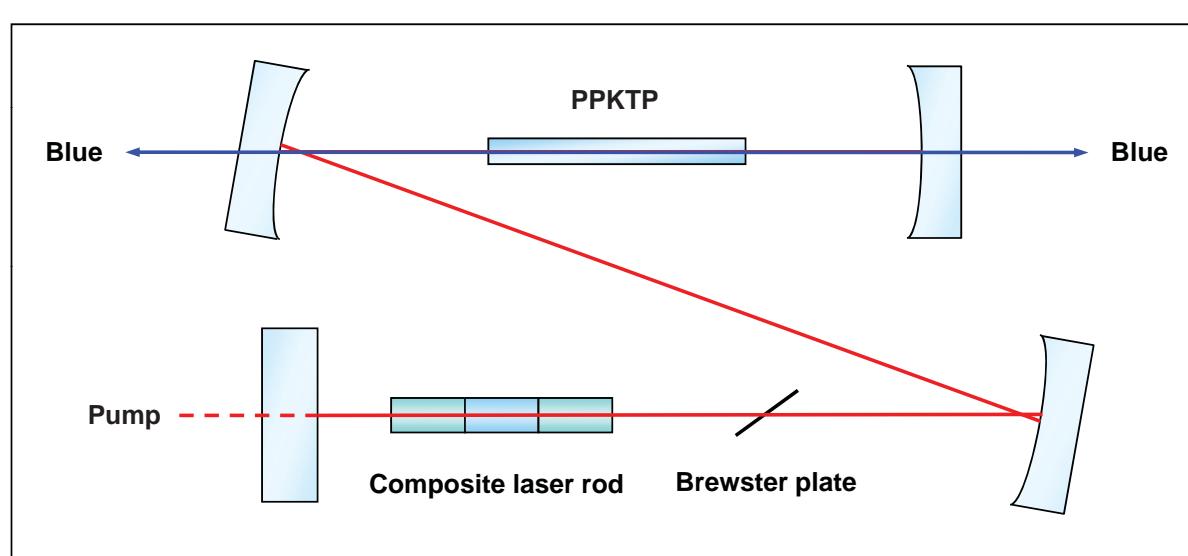


From Yariv: Quantum Electronics

Intra-cavity frequency doubling



- Much higher power available in the cavity
- Often associated with instabilities, the so called “green problem”
- $946 \text{ nm} \rightarrow 473 \text{ nm}$

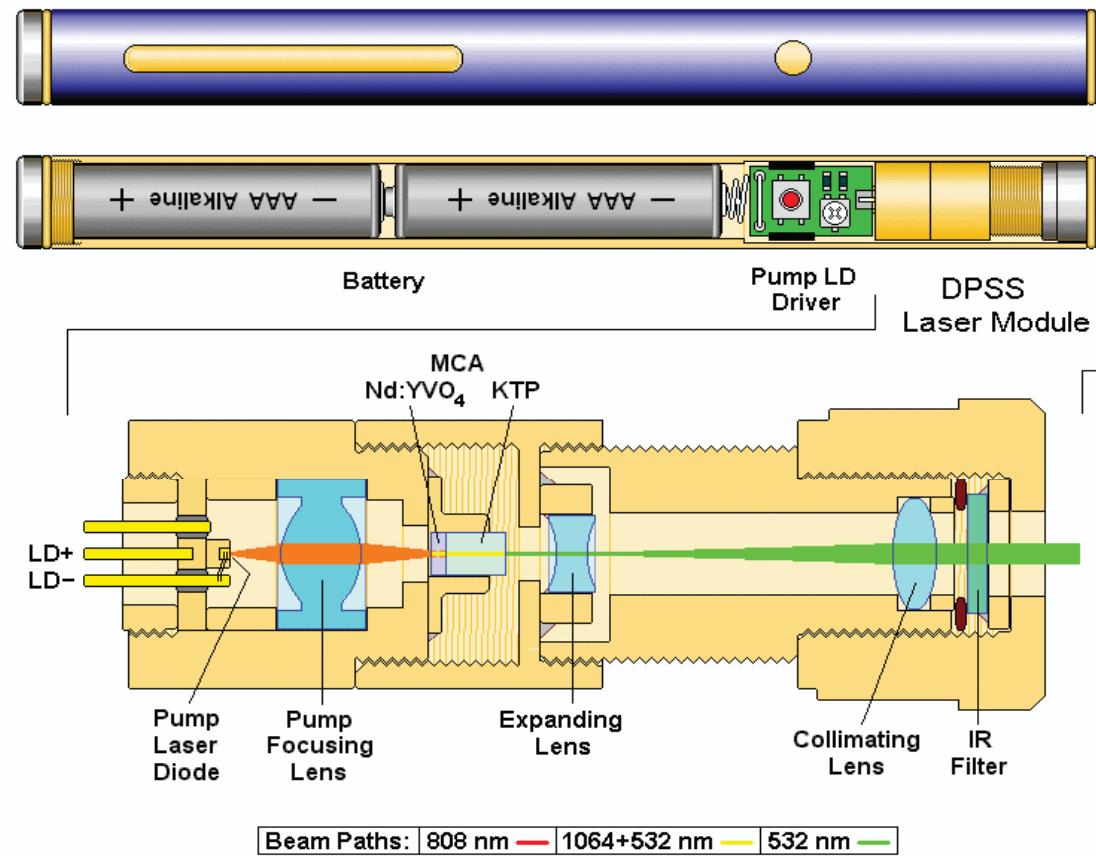


> 500 mW

The laser pointer



A diode-pumped IC frequency doubled solid-state laser

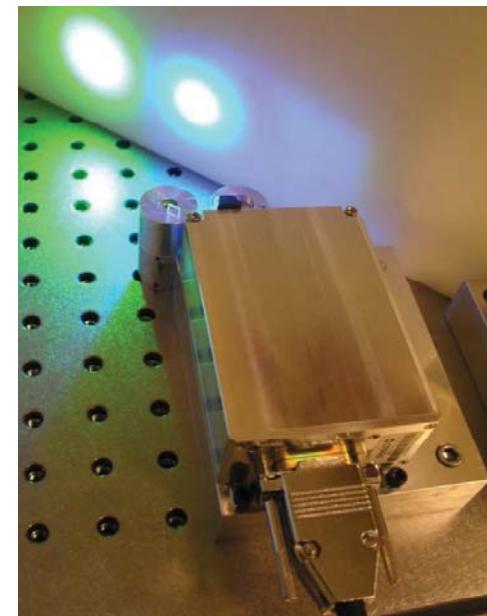
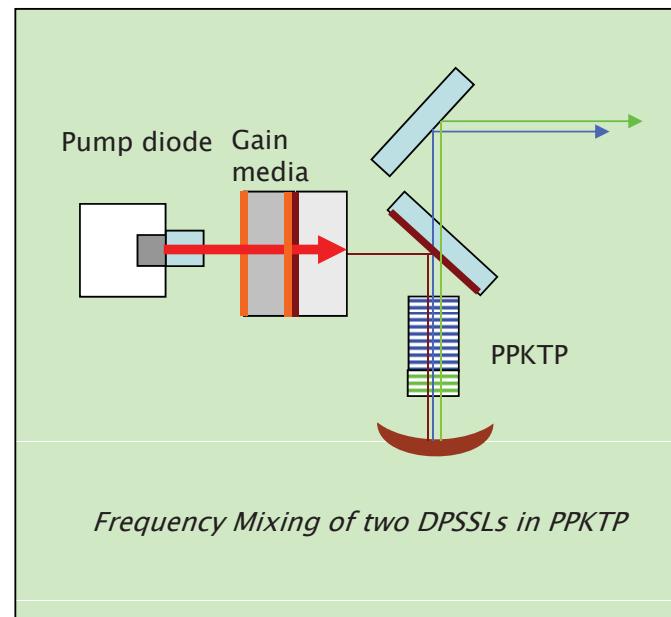


Typical Green DPSS Laser Pointer Using MCA

IC SHG and SFG lasers with PPKTP

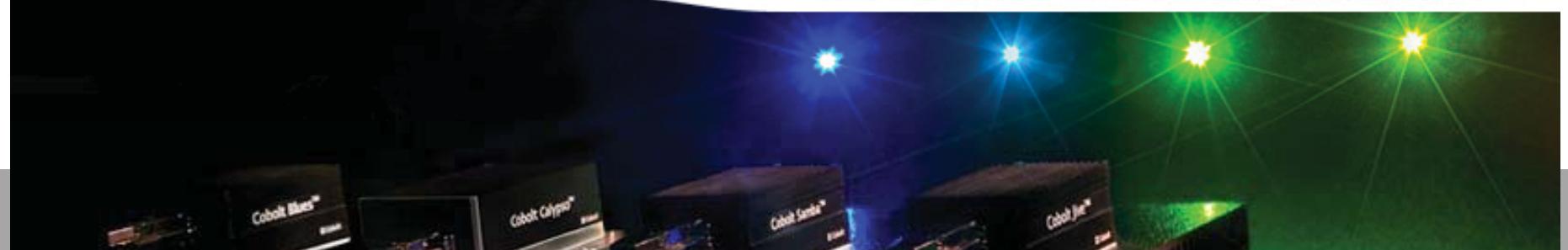


Mixing of two laser lines combined with SHG and SFG



Lasers for biotech, display and graphics

Phone +46 8 545 91 230 Fax +46 8 545 91 231 E-mail info@cobolt.se



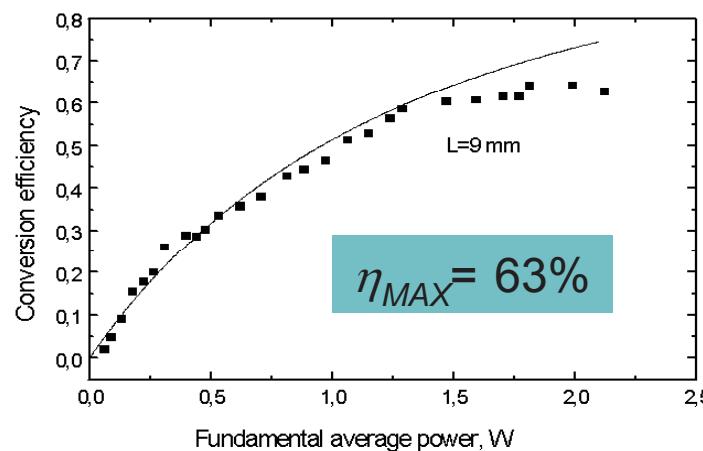
Pico- and nano-second SHG



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

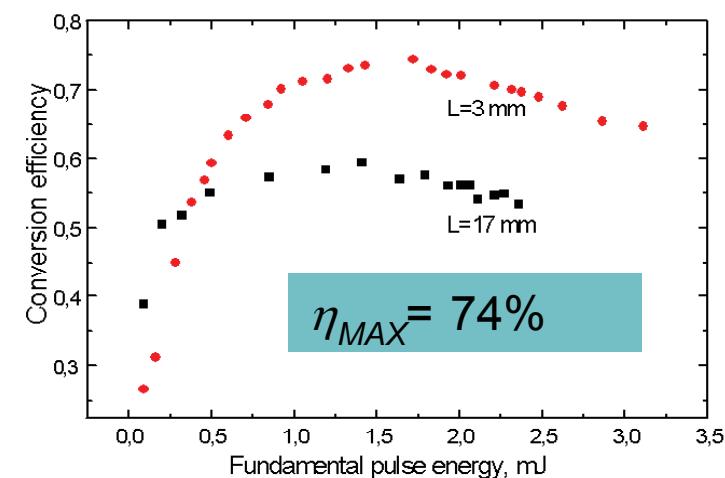
Mode-locked Nd:YAG

$\tau = 100 \text{ ps}$, $f = 100 \text{ MHz}$
Type-I SHG @ 532 nm
PPKTP: $\Lambda = 9.01 \mu \text{m}$



Q-switched Nd:YAG

$\tau = 5 \text{ ns}$, $f = 20 \text{ Hz}$
Type-I SHG @ 532 nm
PPKTP: $\Lambda = 9.01 \mu \text{m}$

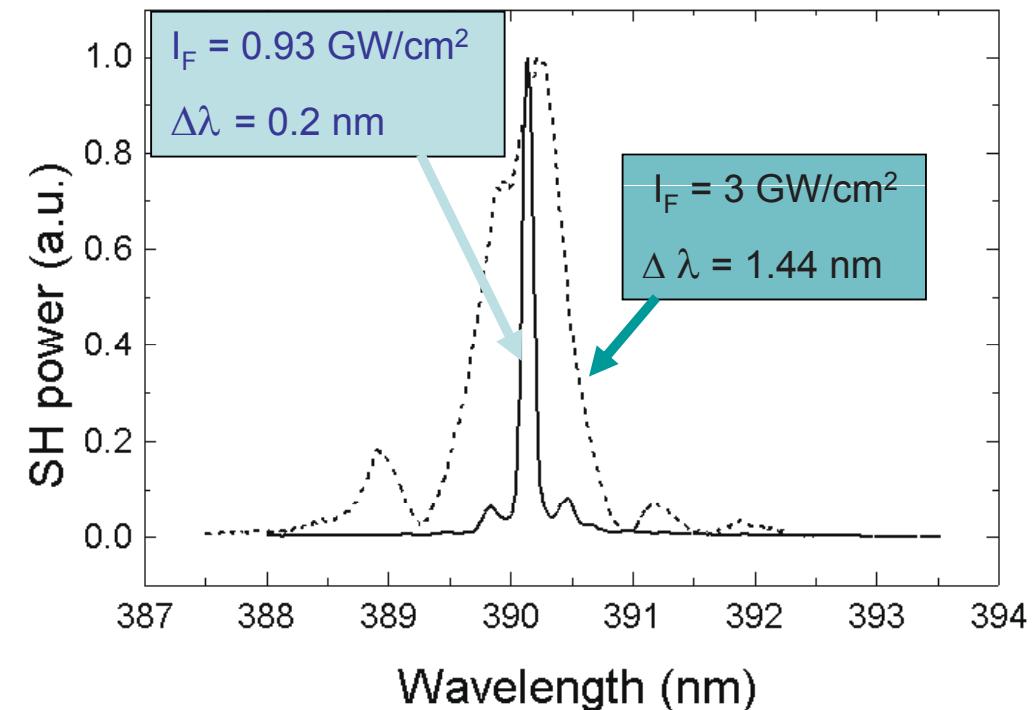


Ultrashort pulse SHG



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

$\tau = 100 \text{ fs}$, $f = 80 \text{ MHz}$
Type-I SHG @ 390 nm
PPKTP: $\Lambda = 2.95 \mu\text{m}$, $L = 9 \text{ mm}$
 $L_{\text{eff}} = 5.6 \text{ mm}$
 $L_w = 57 \mu\text{m}$
 $\tau_{SH} = 10 \text{ ps}$

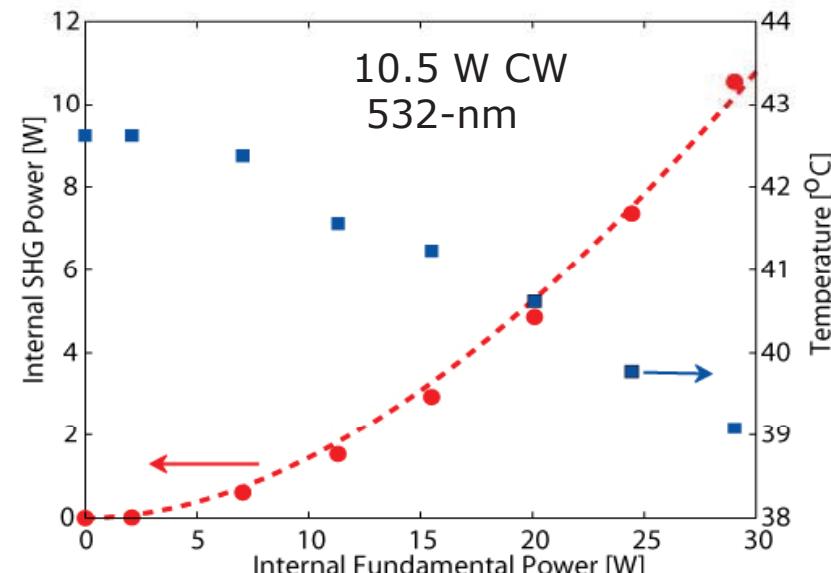
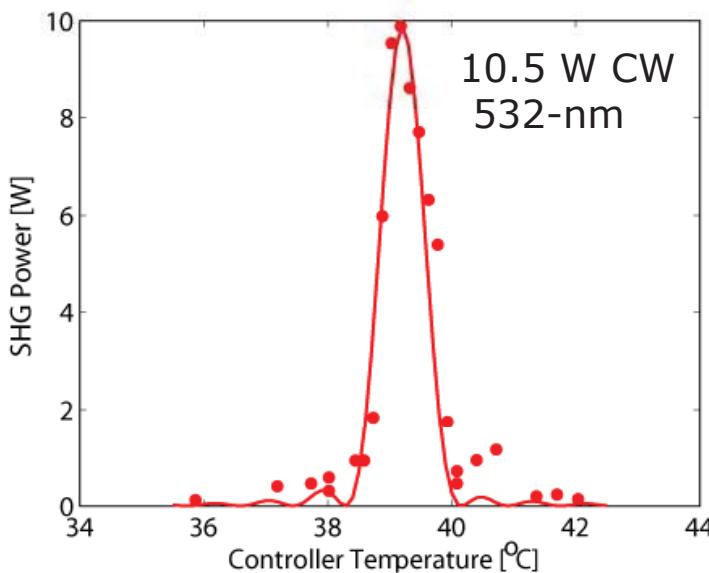
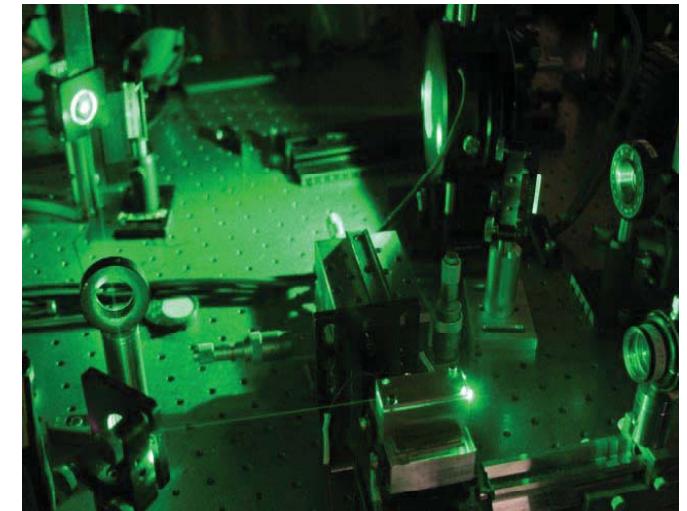


10 W CW 532 nm SHG in VTE PPSLT



Stoichiometry control in PPSLT by VTE
increased photoconductivity
reduced photorefraction
eliminated GRIIRA
visible absorption $\sim 0.1 \text{ \%}/\text{cm}$

Estimate $\sim 100 \text{ W}$ of CW SHG
before material problem





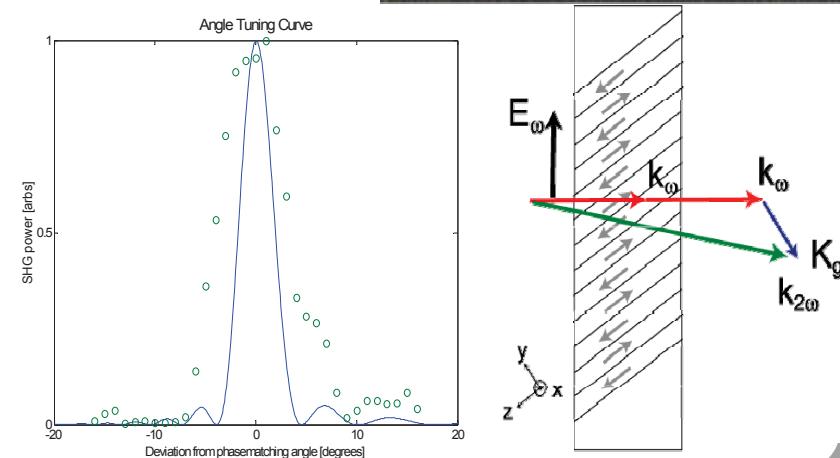
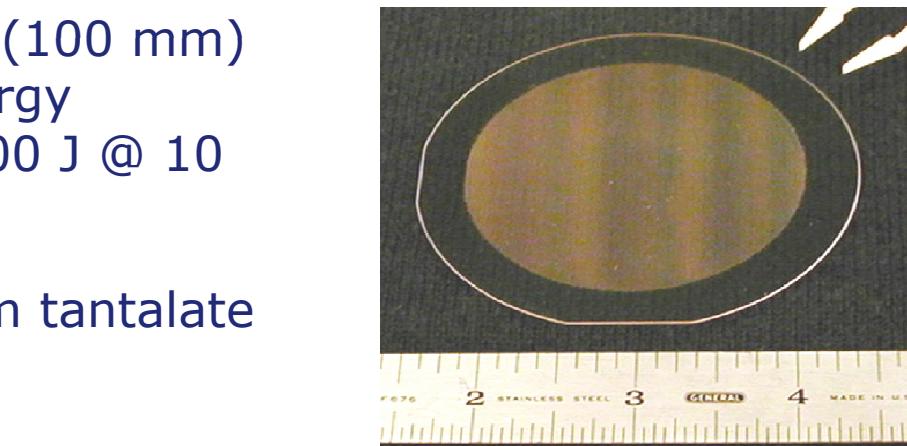
High energy conversion - tilted PP-samples

Damage threshold $\sim 1 \text{ J/cm}^2$ @ 10 ns in LT
⇒ scaling to large fluence requires large apertures

Large wafers commercial (100 mm)
⇒ scalable pulse energy
(100 mm wafer $\sim 100 \text{ J}$ @ 10 ns)

Rotated Y-cut SAW lithium tantalate

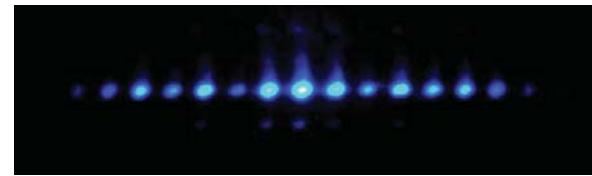
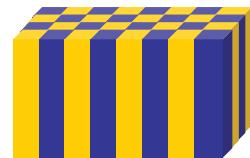
Surface period = 41 μm
thickness = 180 μm (after polishing)
4th order
4 periods total
PSH = 14.5 μW
PFH = 6.2 W
(100kHz, 150ns)



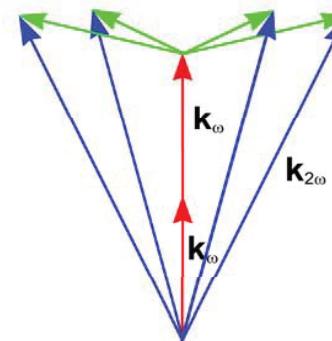
Nonlinear photonic crystals



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



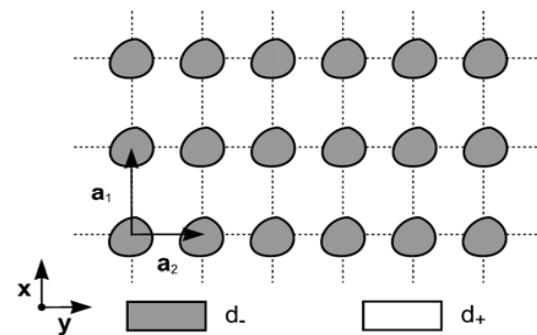
- + Multiple wavelengths
- + Simultaneous phase matching processes
- + Broader bandwidth in non-collinear SHG
- Low efficiency for non-collinear SHG





2D SH generator

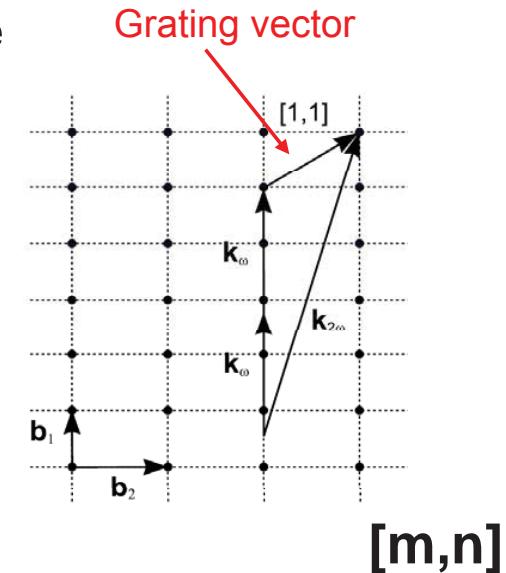
Real lattice



Reciprocal lattice

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

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



Phase-matching condition:

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

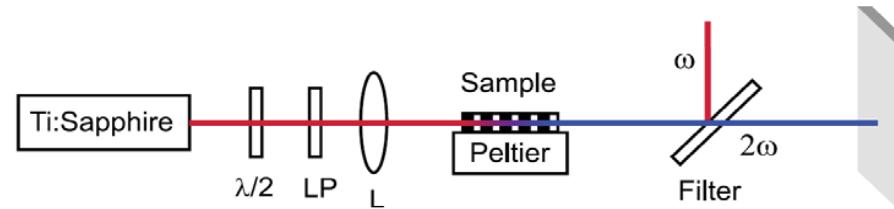
$$\vec{K}_{mn} = mb_1 + nb_2$$

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

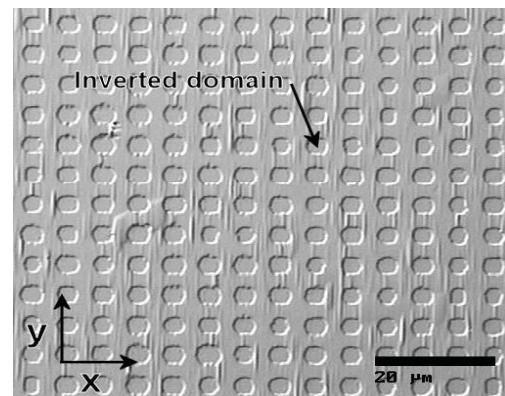
$$\frac{\lambda_{2\omega}}{n_{2\omega}} = \frac{2\pi}{|\mathbf{K}_{mn}|} \sqrt{\left(1 - \frac{n_\omega}{n_{2\omega}}\right)^2 + 4 \frac{n_\omega}{n_{2\omega}} \sin^2 \theta}$$



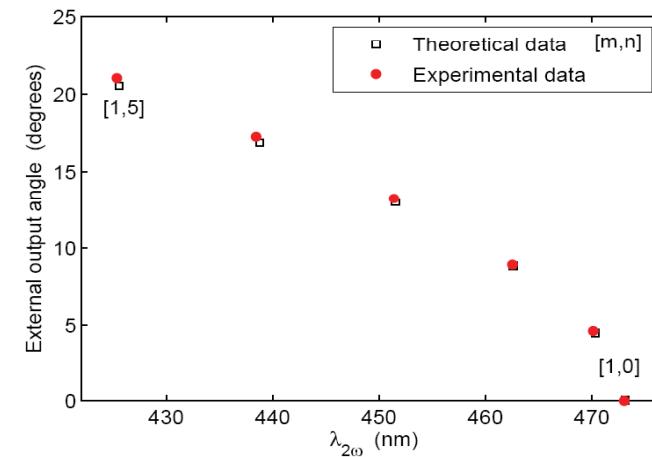
CW SHG in 2D PPKTP



$6.09 \times 6 \mu\text{m}^2$



Patterned area: $8 \times 4 \text{ mm}^2$
Domain depth: $400 \mu\text{m}$

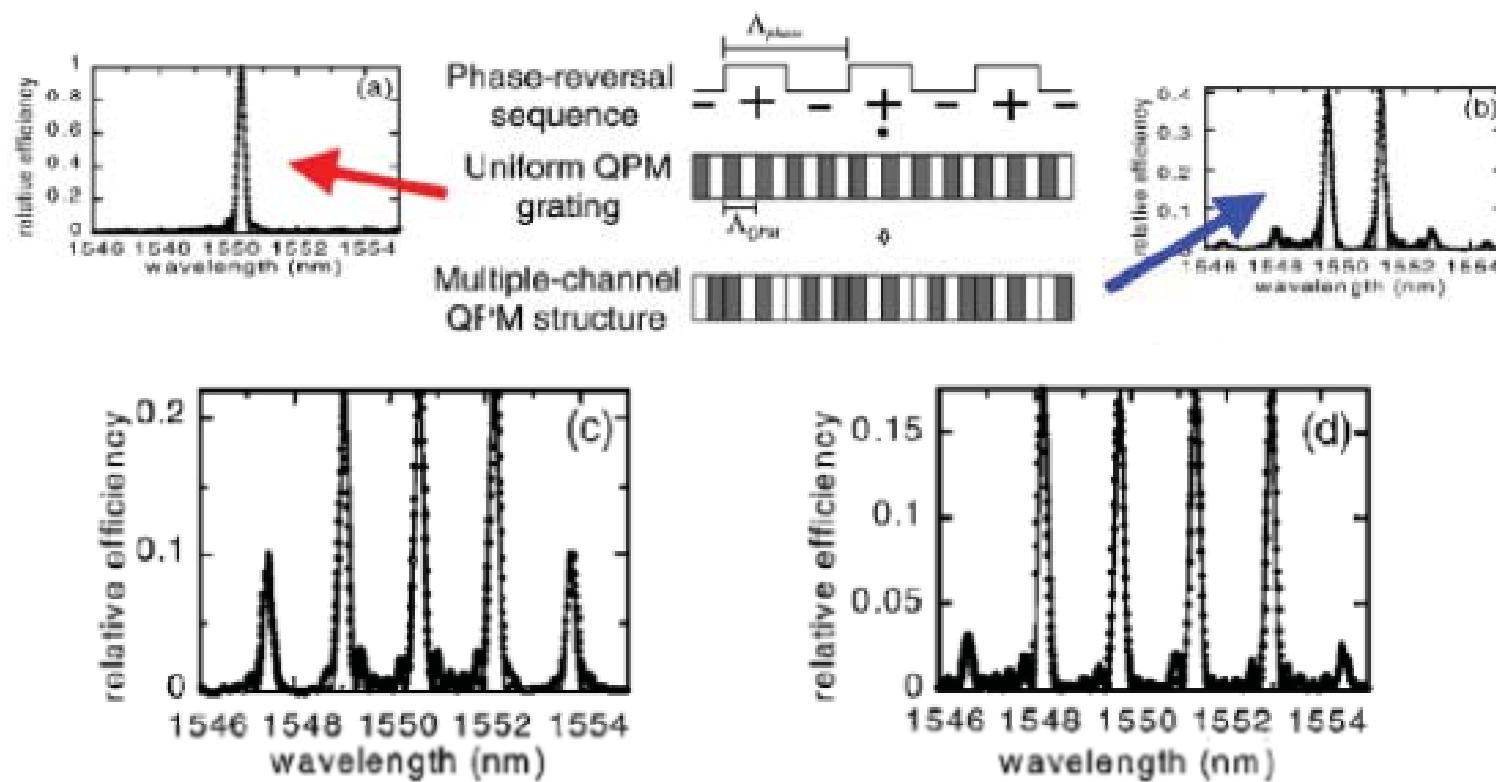




Spectral Tailoring

spatially modifying the grating

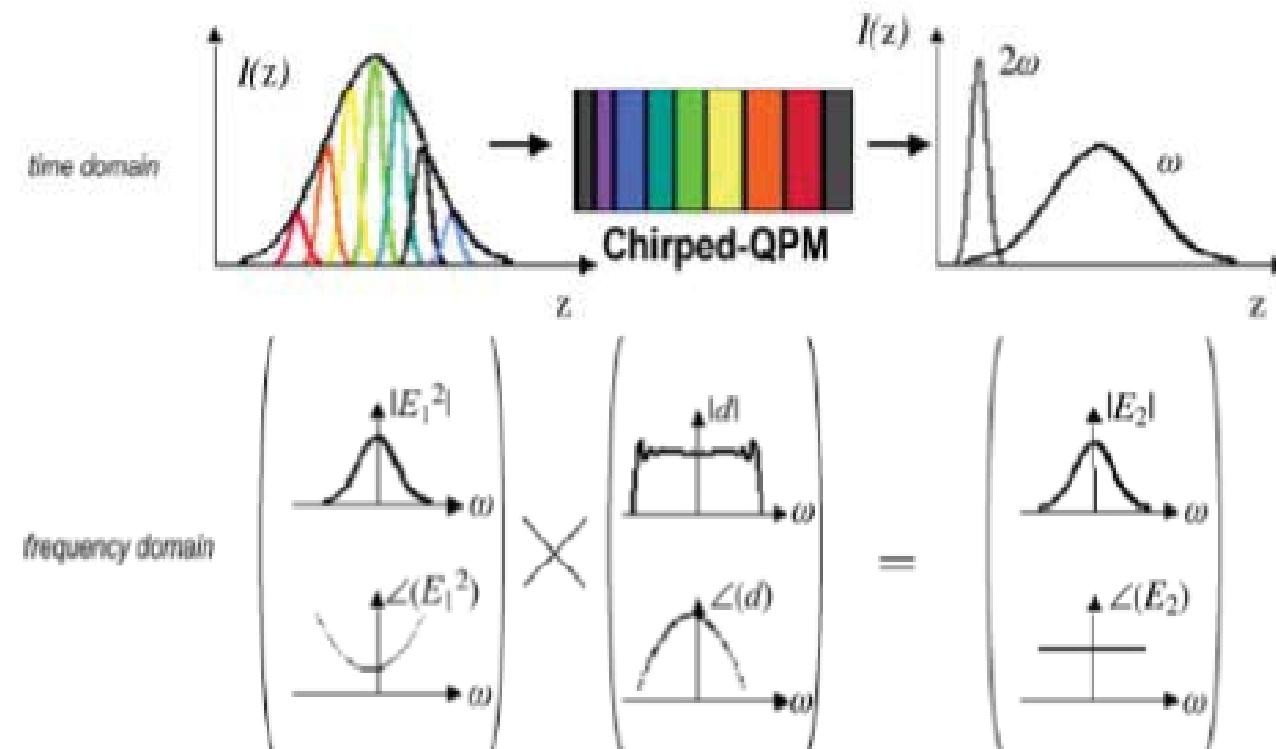
Fourier synthesis used to get the frequency response





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