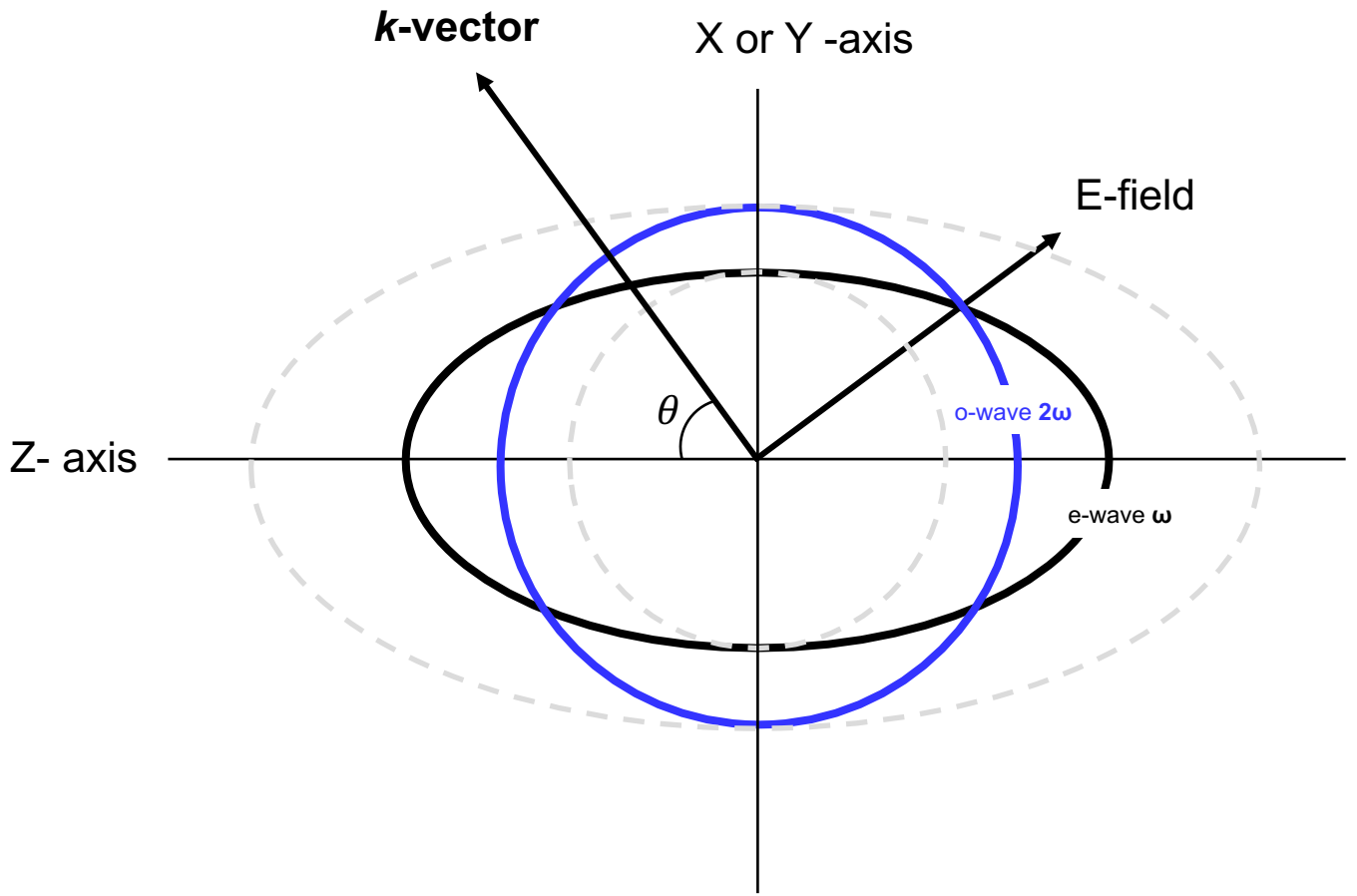
Lecture 9

Phase mismatch & coherence length. Quasi-phase matching (QPM) via domain inversion in ferroelectrics and other media.

Two problems with angular phase matching

Angular phase matching

refractive index ellipsoid



Angular phase matching: spatial walkoff effect problem

If the angle θ between the propagation direction and the optic axis is not 0 or 90 degrees, the Poynting vector (**S**) and the propagation vector ***k*** are not parallel for extraordinary rays. The ordinary and extraordinary rays with parallel propagation vectors diverge from one another as they propagate through the crystal.

plain wave

$$\boldsymbol{E} \rightarrow \boldsymbol{E}e^{i(\omega t-\boldsymbol{kr})},$$
$$\boldsymbol{D} \rightarrow \boldsymbol{D}e^{i(\omega t-\boldsymbol{kr})},$$
$$\boldsymbol{H} \rightarrow \boldsymbol{H}e^{i(\omega t-\boldsymbol{kr})},$$

$$\boldsymbol{\nabla} \cdot \boldsymbol{D} = 0$$

for a plain wave $\boldsymbol{\nabla} \rightarrow = -i\mathbf{k}$

hence $\mathbf{k} \cdot \boldsymbol{D} = 0$

D is orthogonal to **k**

Anisotropic crystal

$$\boldsymbol{D} = \boldsymbol{\varepsilon}\boldsymbol{E}$$

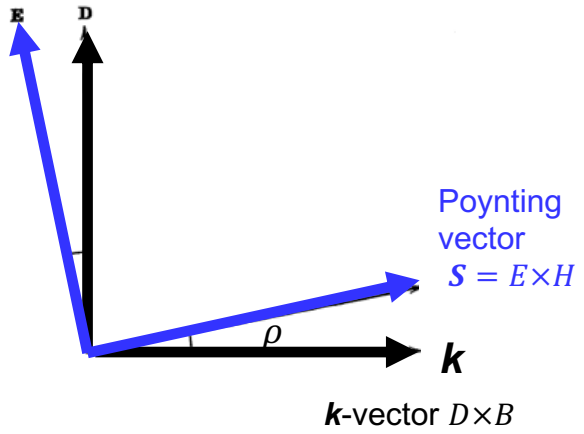
$$\varepsilon = \varepsilon_0 \begin{bmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{bmatrix}$$

in general ***D*** is not parallel to ***E***

Pointing vector $\boldsymbol{S} = \boldsymbol{E} \times \boldsymbol{H}$

k- vector $\boldsymbol{k} = \boldsymbol{D} \times \boldsymbol{B}$

Extraordinary wave

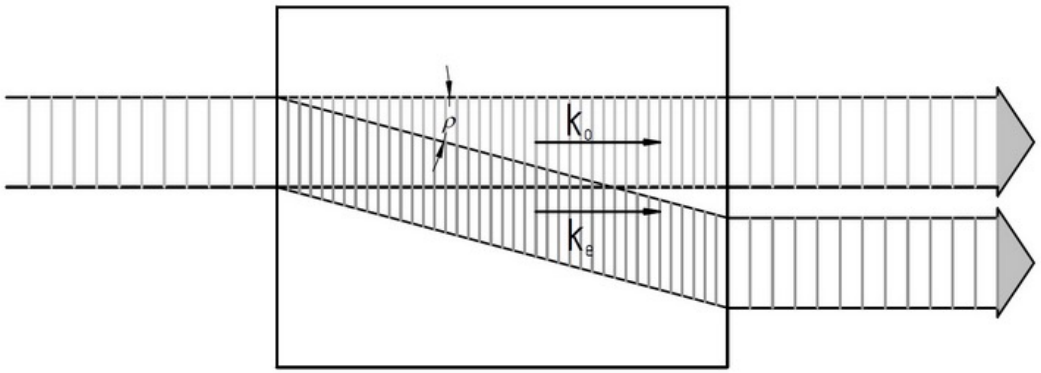


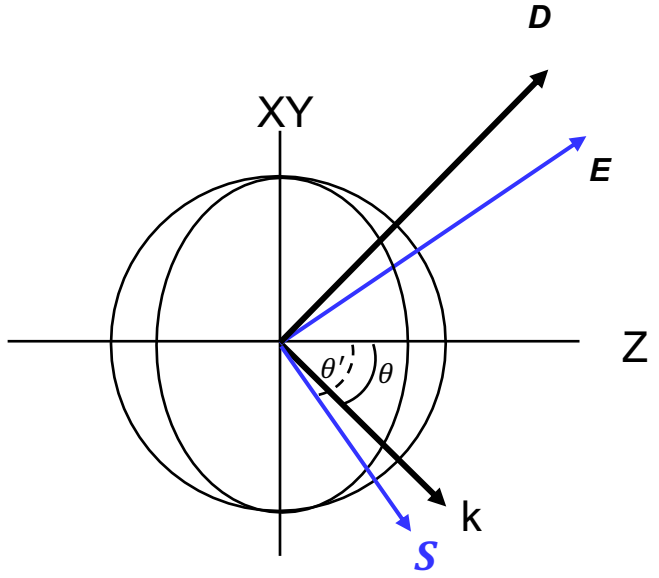
Energy flows along Poynting vector S

In anisotropic crystals, Pointing vector (the power flow) and k-vector generally do not coincide.

Angular phase matching: spatial walkoff effect problem

Walk-off between ordinary and extraordinary beams in a birefringent medium





Walkoff angle:

$$\tan(\theta') = (\frac{n_o}{n_e})^2 \tan(\theta)$$

$$\rho = \theta' - \theta \approx \frac{n_e - n_o}{n_o} \sin(2\theta) = \boxed{\frac{\Delta n}{n_o} \sin(2\theta)} \qquad (9.1)$$

This effect limits the spatial overlap of the two waves and decreases the efficiency of any nonlinear mixing process.

Lithium niobate

$LiNbO_3$ point group 3m uniaxial crystal $n_e < n_o$

from lecture 7:

$$d_{ij} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

Dmitriev et al., Handbook of NLO crystals (1997), p.125

$$d_{22} = 2.1 \frac{pm}{V}$$

$$d_{31} = -4.35 \frac{pm}{V}$$

$$d_{33} = 27.2 \frac{pm}{V}$$

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = 2\epsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{31} & -d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E_x^2 \\ E_y^2 \\ E_z^2 \\ 2E_yE_z \\ 2E_xE_z \\ 2E_xE_y \end{bmatrix}$$

Only two scenarios for SFG: 1) Type-I SFG $\omega_1 + \omega_2 = \omega_3$

$o + o = e$

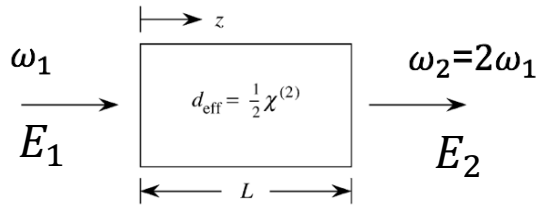
2) Type-II SFG $\omega_1 + \omega_2 = \omega_3$

$e + o = e$

Second problem: using birefringence, one cannot have access to the highest tensor elements like **d_{33}** in lithium niobate, because ω and 2ω polarizations need to be the same!

Coherence length

Use the example of second harmonic generation (SHG).



A plane wave field of amplitude E_1 at frequency ω_1 and wavevector $k_1 = \frac{n_1\omega_1}{c}$, passes through a medium with quadratic nonlinear susceptibility d, generating a nonlinear polarization wave proportional to dE_1^2 at frequency ω_2=$2\omega_1$ and wavevector $2k_1$.

The polarization wave radiates a free second harmonic wave with wave vector $k_2 = \frac{n_2\omega_2}{c}$.

The *forced* and *free* waves accumulate a phase shift

$$\Delta\varphi = \Delta k l = (k_2 - 2k_1)l = \left(\frac{n_2\omega_2}{c} - 2\frac{n_1\omega_1}{c}\right)l = \frac{2\omega_1}{c}(n_2 - n_1)l$$

$$\Delta\varphi = l_c\,\Delta k = \pi \qquad \boxed{l_c = \frac{\pi}{\Delta k}} \qquad (9.2)$$

definition of the coherence length: phase changes by π

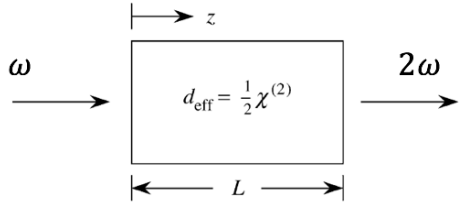
or $$l_c = \frac{\pi}{k_2 - 2k_1} = \frac{\pi c}{2\omega_1(n_2 - n_1)} = \frac{\lambda_\omega}{4(n_2 - n_1)} = \frac{\lambda_\omega}{4\Delta n} = \frac{\lambda_{2\omega}}{2\Delta n} \qquad (9.3)$$

where λ_ω is the vacuum wavelength (at ω_1)

The direction of power flow between the fundamental and harmonic depends on the relative phase of the forced and free waves, and hence changes sign every coherence length.

Non-phase-matched SHG

Second harmonic generation



ΔkL>>1

The slowly-varying envelope equation (low conversion limit) from Lecture 6:

$$\frac{dA_2}{dz} = -i\frac{g}{2}A_1{}^2 e^{i\Delta kz} = -i\gamma e^{i\Delta kz} \qquad (9.4)$$

where $\gamma = \frac{g}{2}A_1{}^2 = \frac{d}{2c}\sqrt{\frac{\omega_1^2\omega_2}{n_1^2 n_2}}A_1{}^2$

$$\Delta k = k_2 - 2k_1$$

$$A_i = \sqrt{\frac{n_i}{\omega_i}}E_i$$

$$g = \frac{d}{c}\sqrt{\frac{\omega_1^2\omega_2}{n_1^2 n_2}}$$

NL coupling coeff.

$$I = \frac{c\varepsilon_0}{2}\omega|A|^2$$

Now integrate:

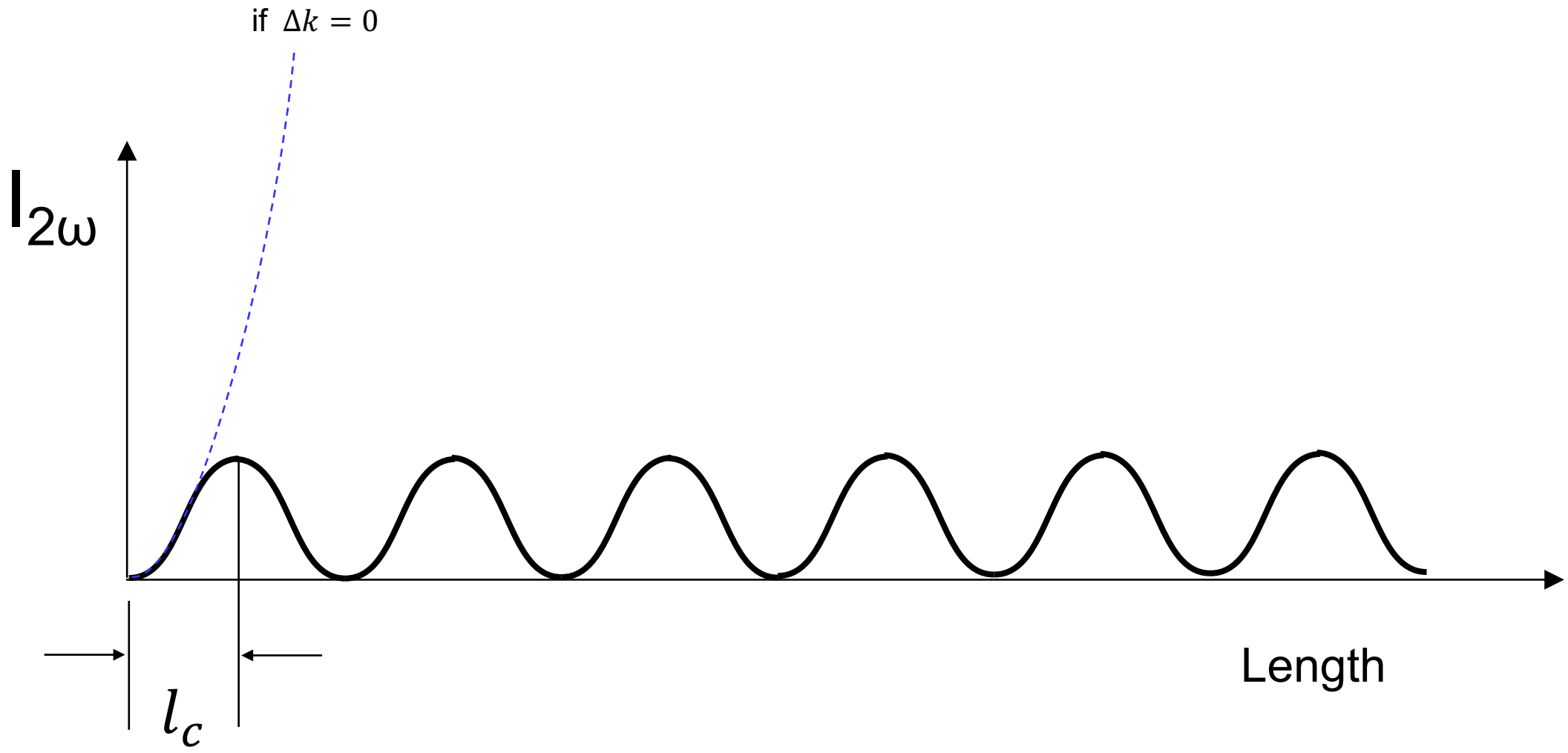
$$A_2 = -i\gamma\int_0^L e^{i\Delta kz}dz = -i\gamma\frac{1}{i\Delta k}(e^{i\Delta kL} - 1) = -i\gamma\frac{1}{i\Delta k}e^{\frac{i\Delta kL}{2}}2i\sin\left(\frac{\Delta kL}{2}\right) =$$

$$= \underbrace{-i\,e^{\frac{i\Delta kL}{2}}}_{\text{phase term}}\underbrace{\gamma\frac{2}{\Delta k}}_{\text{amplitude}}\sin\left(\frac{\Delta kL}{2}\right); \qquad (9.5)$$

SHG intensity $\sim |A_2|^2 = (\frac{2\gamma}{\Delta k})^2 sin^2\left(\frac{\Delta kL}{2}\right)$ (9.6)

Coherence length

ΔkL>>1



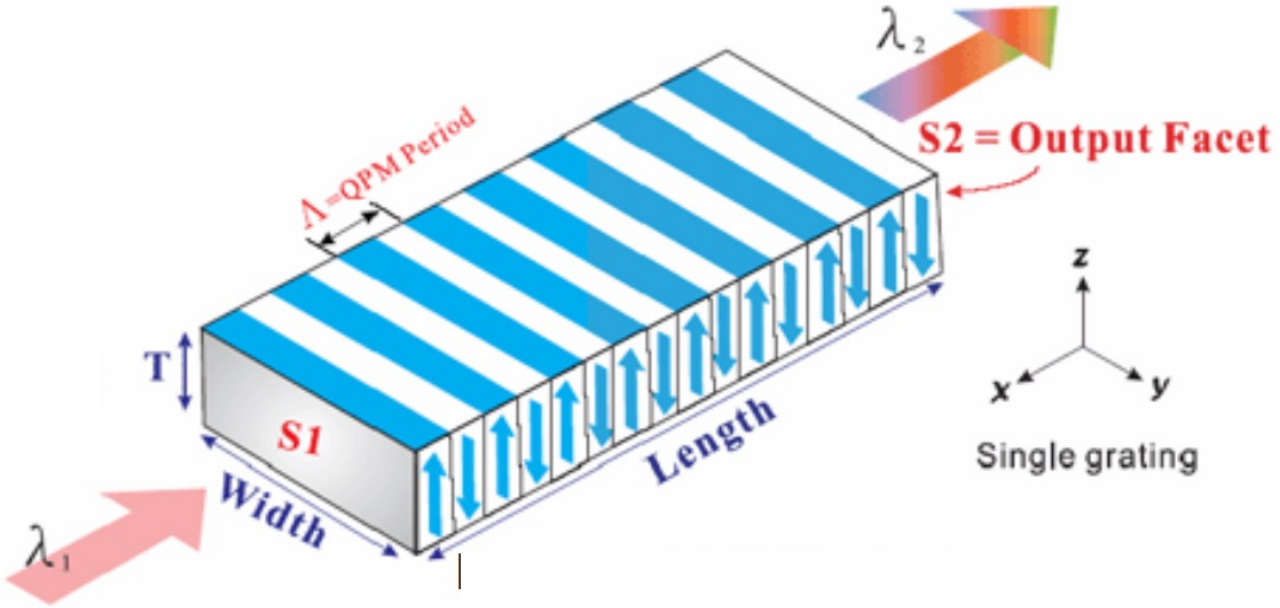
Quasi-phase matching (QPM)

The other method satisfying momentum conservation is quasi-phase-matching (QPMP).

A nonlinear material with spatially modulated nonlinear properties; the crystal axis is flipped at a regular interval $\Lambda = 2l_c$

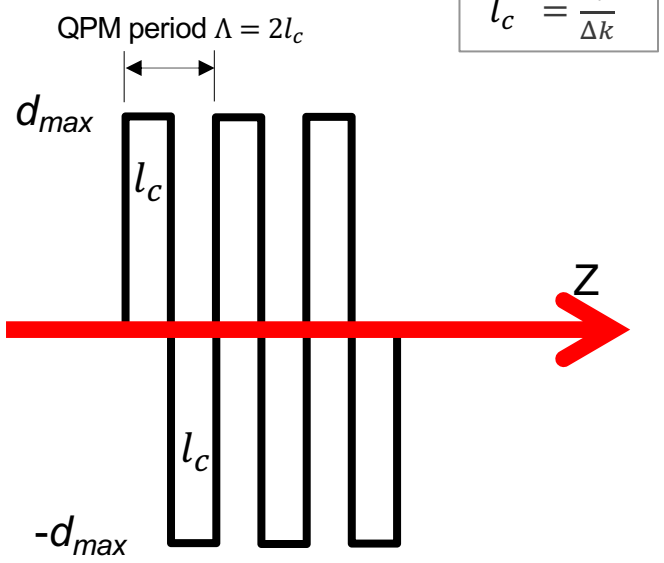
Typically Λ =10–100 μm.

Example: QPM lithium niobate



Quasi-phase matching (QPM): theory

$l_c = \frac{\pi}{\Delta k}$



Imagine we flip the sign of the nonlinear coefficient *d(z)* with the period $\Lambda = 2l_c$

Now integrate (9.2), taking into account that $\gamma = \gamma(z)$ will also flip from (+) to (-)

$$A_2 = -i \int_0^L \gamma(z)\, e^{i\Delta k z} dz$$

Let us first integrate this over an interval from 0 to $2l_c = \frac{2\pi}{\Delta k}$

$$(-i)\int_0^{\frac{2\pi}{\Delta k}} \gamma(z)\, e^{i\Delta k z} dz = (-i\gamma)\{\int_0^{\frac{\pi}{\Delta k}} e^{i\Delta k z} dz - \int_{\frac{\pi}{\Delta k}}^{\frac{2\pi}{\Delta k}} e^{i\Delta k z} dz\} = \frac{(-i\gamma)}{i\Delta k}\{-2-2\} = \frac{4\gamma}{\Delta k}$$

Since both $\gamma(z)$ and $e^{i\Delta k z}$ are periodic with the period $\frac{2\pi}{\Delta k}$

and there are $\mathrm{L}/(2\pi/\Delta k)$ such periods, the sought-for integral is:

$$A_2 = -i \int_0^L \gamma(z)\, e^{i\Delta k z} dz = \frac{\mathrm{L}}{\frac{2\pi}{\Delta k}} \cdot \frac{4\gamma}{\Delta k} = \left(\frac{2}{\pi}\right)\gamma L$$

Quasi-phase matching (QPM): theory

from $A_2 = (\frac{2}{\pi})\gamma L$ (9.7)

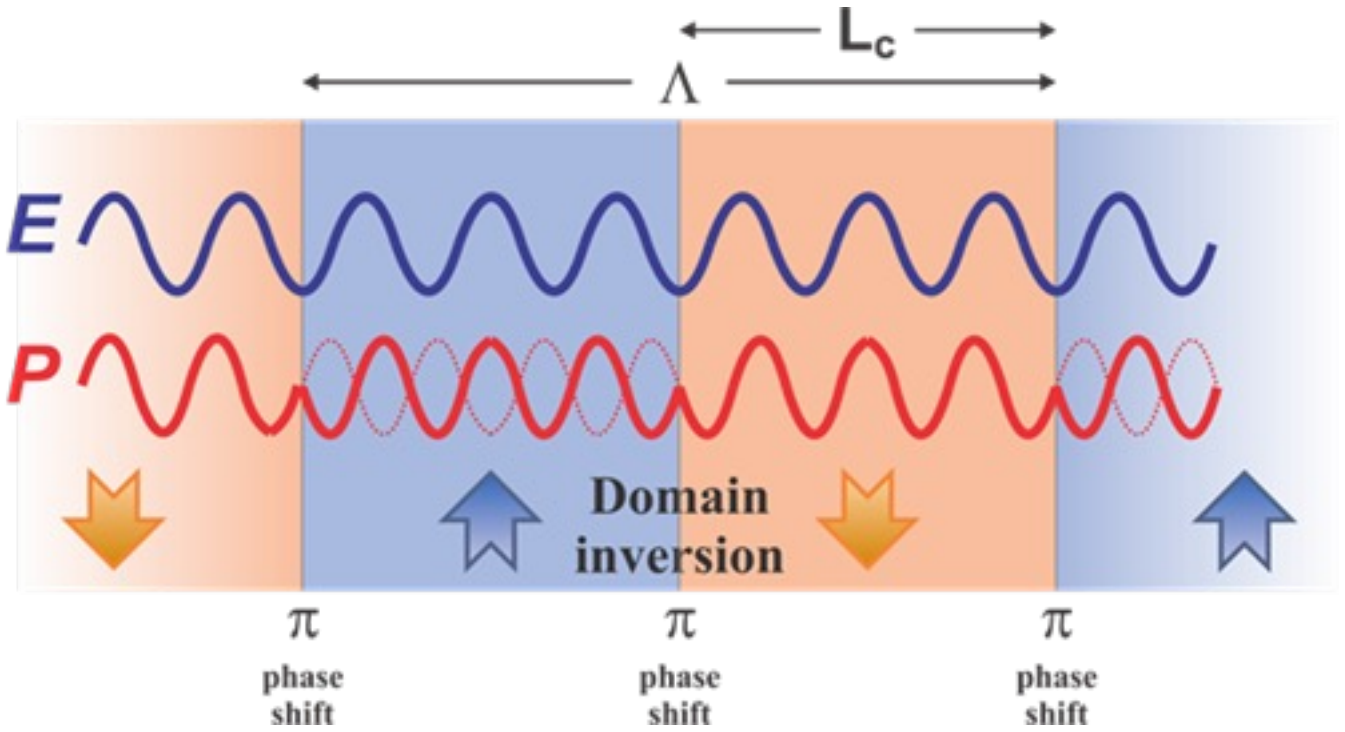
we get: $|A_2|^2 = (\frac{2}{\pi})^2 \gamma^2 L^2$ (9.8)

$$I_{2\omega} = \frac{2\omega^2}{\epsilon_0 c^3}(\frac{d_{QPM}^2}{n^3})\, I_\omega{}^2 L^2 \quad (9.9)$$

$$\boxed{d_{QPM} = \frac{2}{\pi} d} \quad (9.10)$$

We see that the largest QPM nonlinear coefficient is reduced below that of a truly phase-matched medium by a factor $2/\pi$.

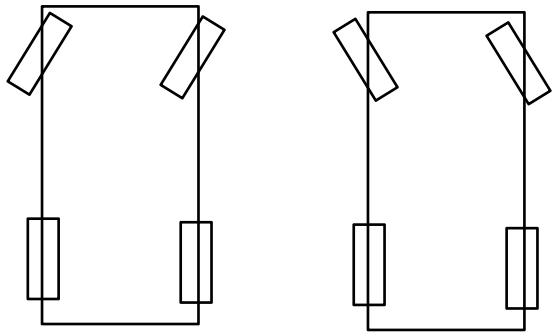
Quasi-phase matching (QPM)

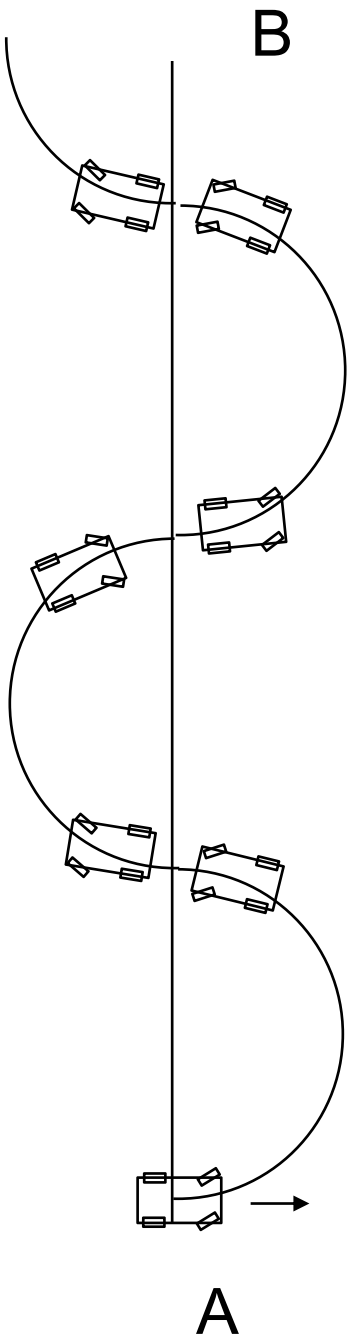


Changing the direction of z or the direction of the domain alignment, changes the sign of the nonlinear coupling and reverses the direction of energy flow.

By changing the sign of the nonlinear susceptibility every coherence length, the phase of the polarization wave is shifted by π, effectively rephasing the interaction and leading to monotonic power flow into the harmonic wave.

QPM analogy





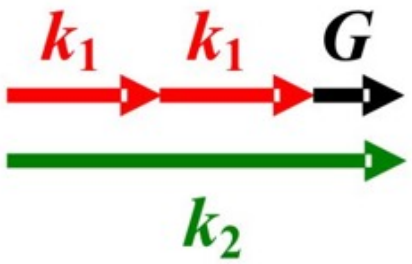
your gas mileage will decrease by a coefficient of

$$\frac{2}{\pi}$$

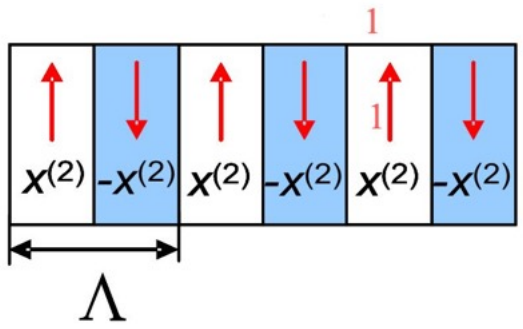
Quasi-phase matching (QPM) - another view

SHG

Momentum conservation:



$$k_1 + k_1 + G = k_3$$

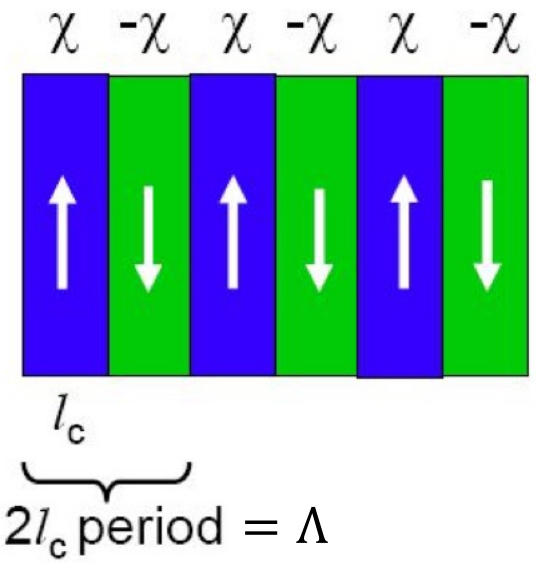


$G=2\pi/\Lambda$

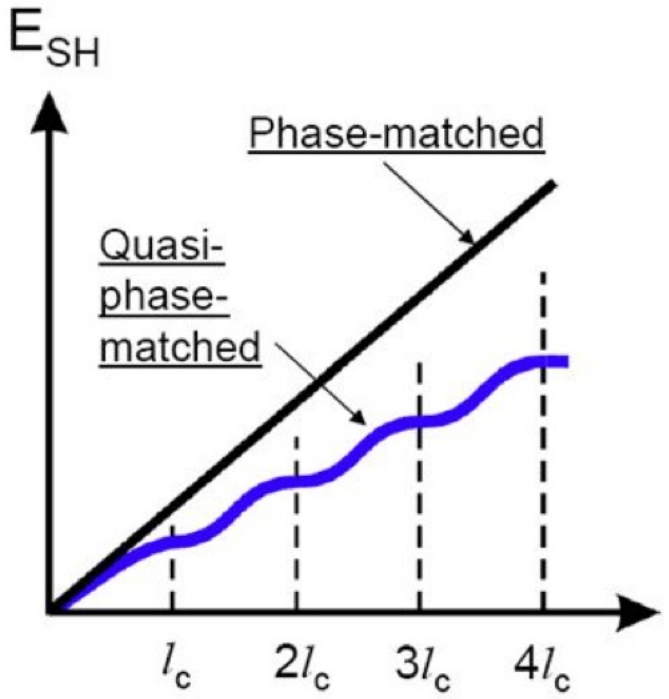
The QPM crystal itself provides the additional wavevector $k = 2\pi/\Lambda$ (and hence momentum) to satisfy the phase-matching condition.

Quasi-phase matching (QPM)

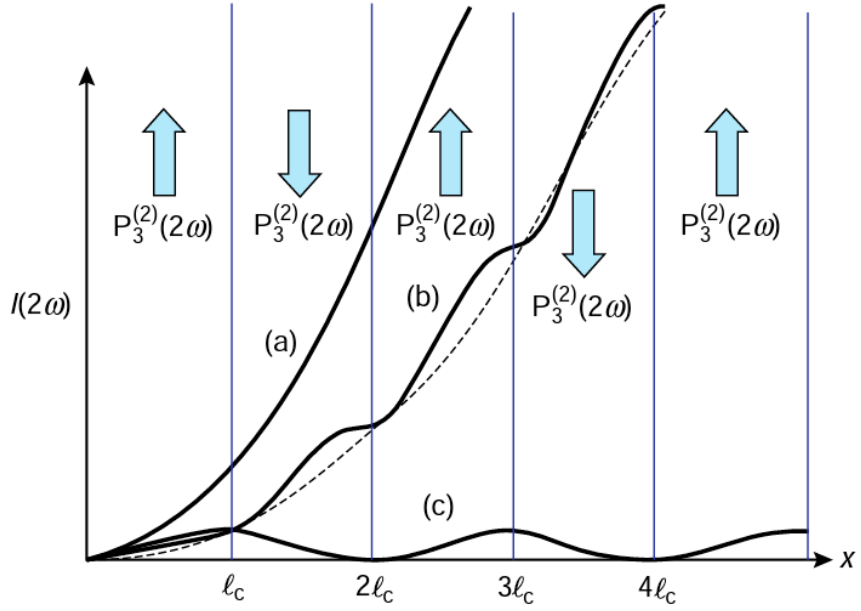
Period = $2l_c$
1st order
phase-matching

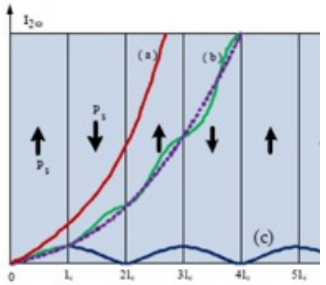


SH field



SH intensity

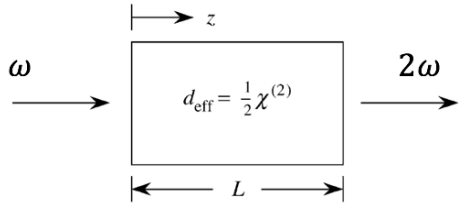




Phase matched SHG

Second harmonic generation

$\Delta kL \sim 1$ *or less*



The slowly-varying envelope equation (low conversion limit) from (9.4) is:

$$\frac{dA_2}{dz} = -i\frac{g}{2}A_1{}^2\, e^{i\Delta kz} = -i\gamma\, e^{i\Delta kz}$$

where $\gamma = \frac{g}{2}A_1{}^2 = \frac{d}{2c}\sqrt{\frac{\omega_1^2\omega_2}{n_1^2 n_2}}A_1{}^2$

$= \frac{d}{2c}\sqrt{\frac{2\omega^3}{n^3}}A_1{}^2$

$\Delta k = k_2 - 2k_1$

$A_i = \sqrt{\frac{n_i}{\omega_i}}E_i$

$g = \frac{d}{c}\sqrt{\frac{\omega_1^2\omega_2}{n_1^2 n_2}}$ NL coupling coeff.

$I = \frac{c\varepsilon_0}{2}\omega|A|^2$

Now integrate:

$$A_2 = -i\gamma\int_0^L e^{i\Delta kz}dz = -i\gamma\frac{1}{i\Delta k}(e^{i\Delta kL} - 1) =$$

$$= -i\gamma\frac{1}{i\Delta k}\, e^{\frac{i\Delta kL}{2}} 2i\sin\left(\frac{\Delta kL}{2}\right) = -ie^{i(\frac{\Delta kL}{2})}\gamma L\sin\left(\frac{\Delta kL}{2}\right)/\left(\frac{\Delta kL}{2}\right) = -ie^{i(\frac{\Delta kL}{2})}\gamma L\, sinc\left(\frac{\Delta kL}{2}\right) \quad (9.11)$$

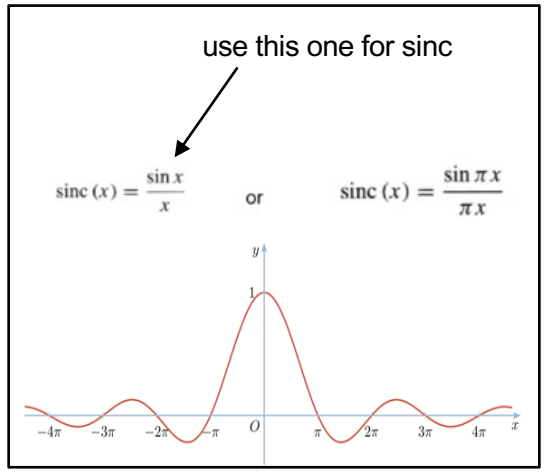
phase term

flow of photons at 2ω

$$|A_2|^2 = (\gamma L)^2 sinc^2\left(\frac{\Delta kL}{2}\right) = \left(\frac{d}{2c}\right)^2 \frac{2\omega^3}{n^3}|A_1|^4 L^2\, sinc^2\left(\frac{\Delta kL}{2}\right)$$

use $|A_1|^2 = \frac{2I_1}{c\varepsilon_0\omega}$ $|A_2|^2 = \frac{2I_1}{c\varepsilon_0(2\omega)}$

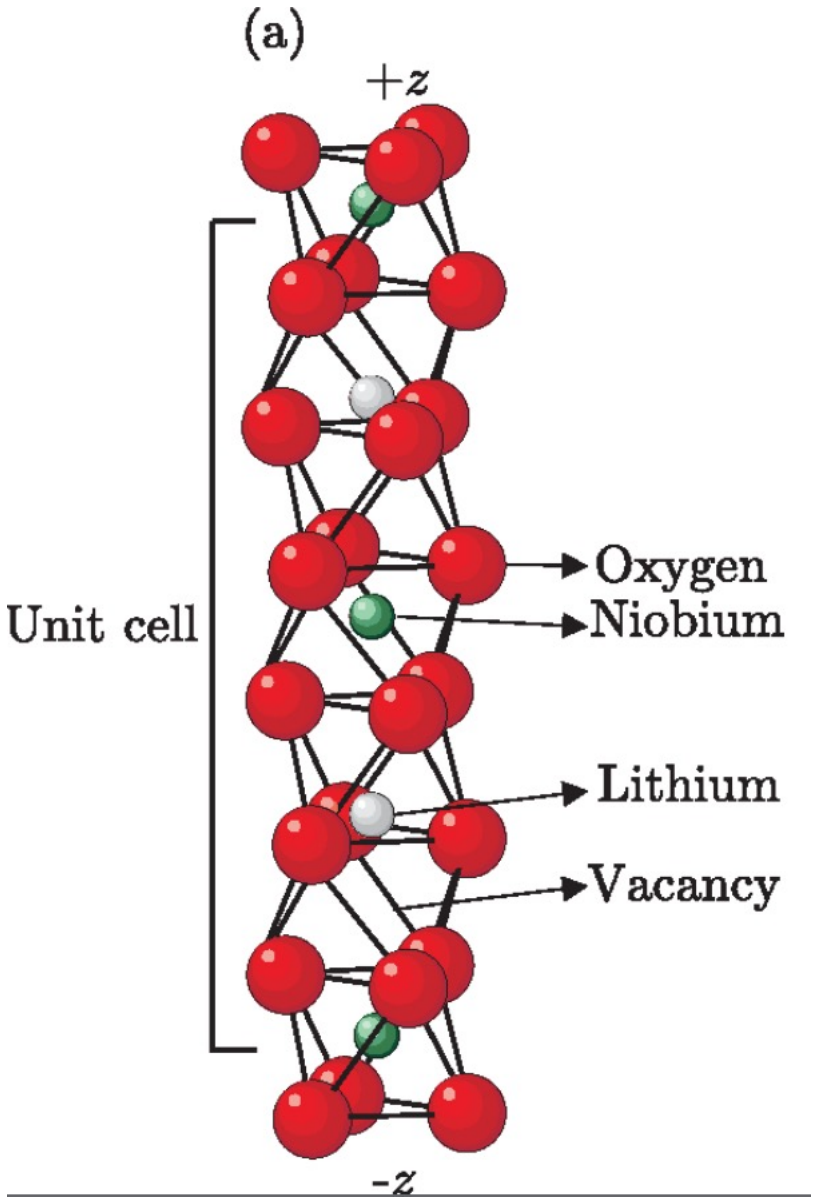
$$\boxed{I_{2\omega} = \frac{2\omega^2}{\epsilon_0 c^3}\left(\frac{d^2}{n^3}\right) I_\omega{}^2 L^2 sinc^2\left(\frac{\Delta kL}{2}\right)} \quad (9.12)$$



self-test : Δk=0 → get formula (6.10a) for low conv. eff-cy approx.

also: recall uncertainty relation $\Delta kL \sim 1$

Quasi-phase matching (QPM)



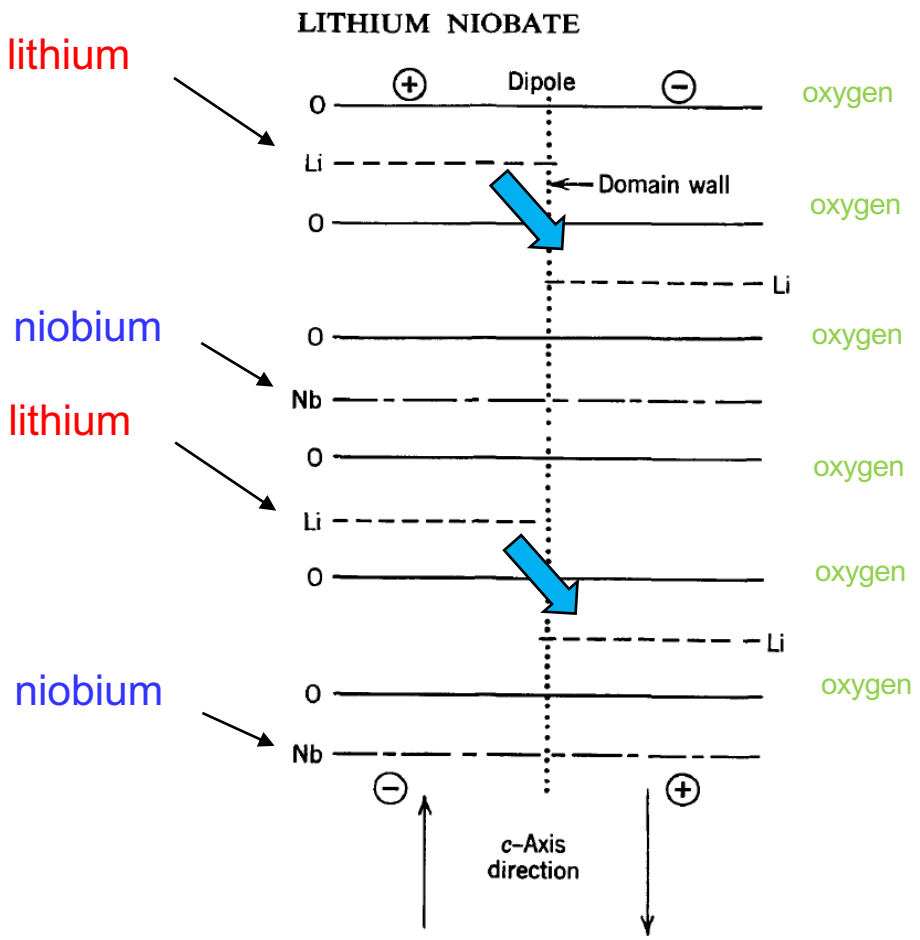
Lithium niobate – a ferroelectric material

Ferroelectric effect: phenomenon where a crystal exhibits a permanent and spontaneous dipole moment; analogous to ferromagnetism of permanent magnets

Periodically-poled lithium niobate (PPLN) is a domain-engineered crystal. The ferroelectric domains point alternatively to the +c and the −c direction, with a period of typically between 5 and 35 μm. The shorter periods of this range are used for second harmonic generation, while the longer ones for optical parametric oscillation. Domain reversal is achieved by electrical poling .

Quasi-phase matching (QPM)

Lithium niobate: ferroelectric domain reversal

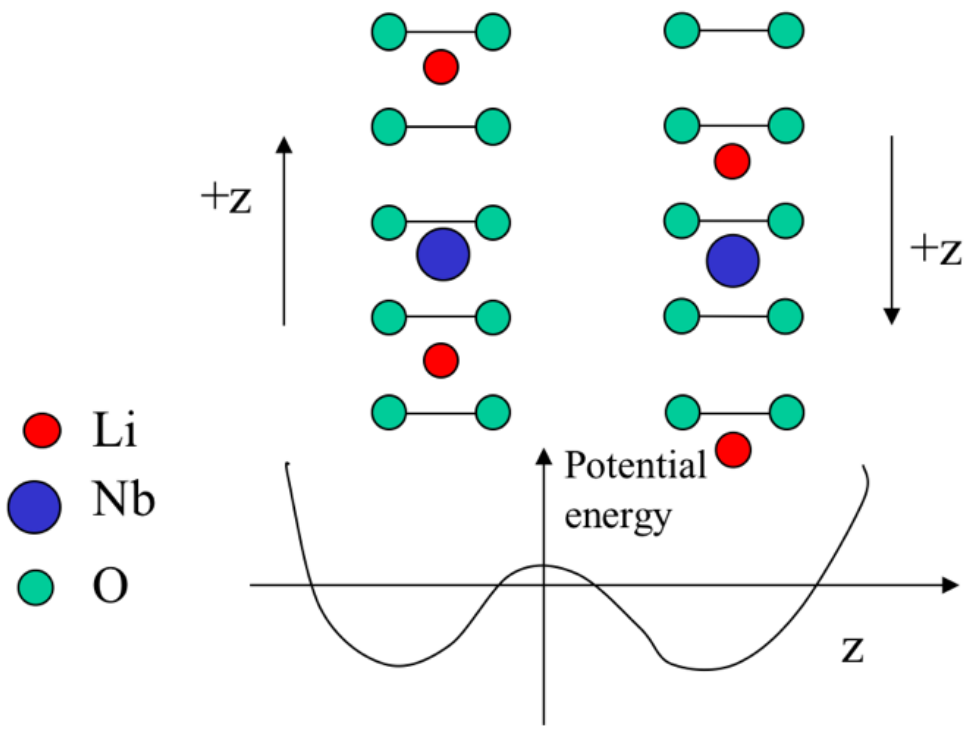


Schematic diagram of the structure of lithium niobate showing the effects of poling.

The lithium, niobium, and oxygen atoms lie in layers.

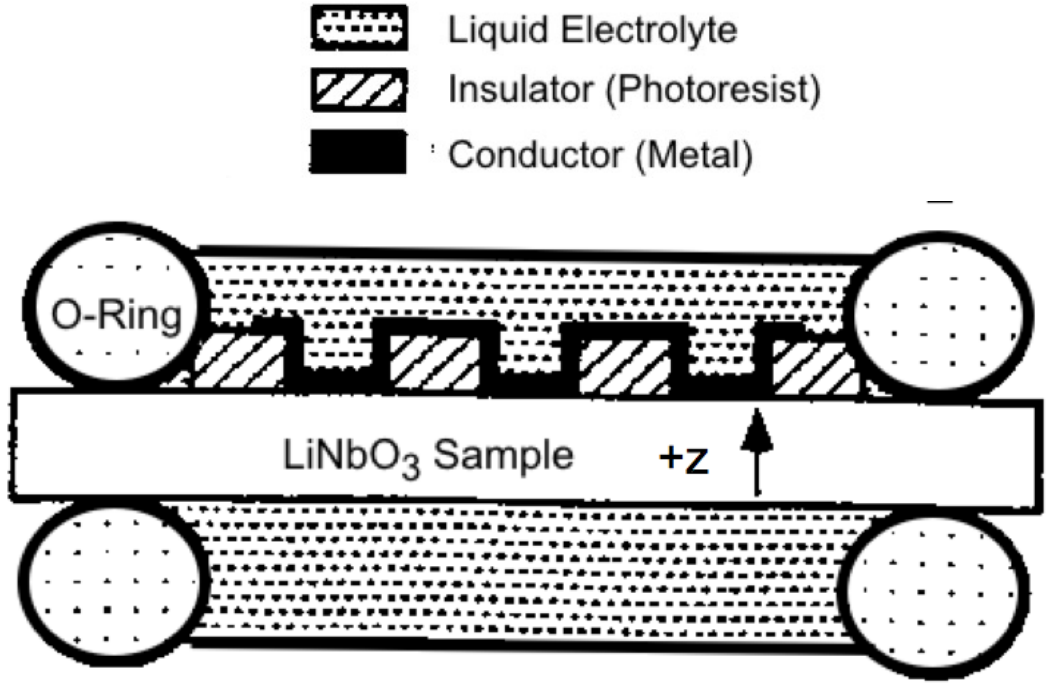
The positive z direction, is determined by the position of the layer of lithium atoms in the structure. Poling causes this layer of **lithium atoms to move through the oxygen layer into the adjacent space**, reversing the direction of the crystal structure.

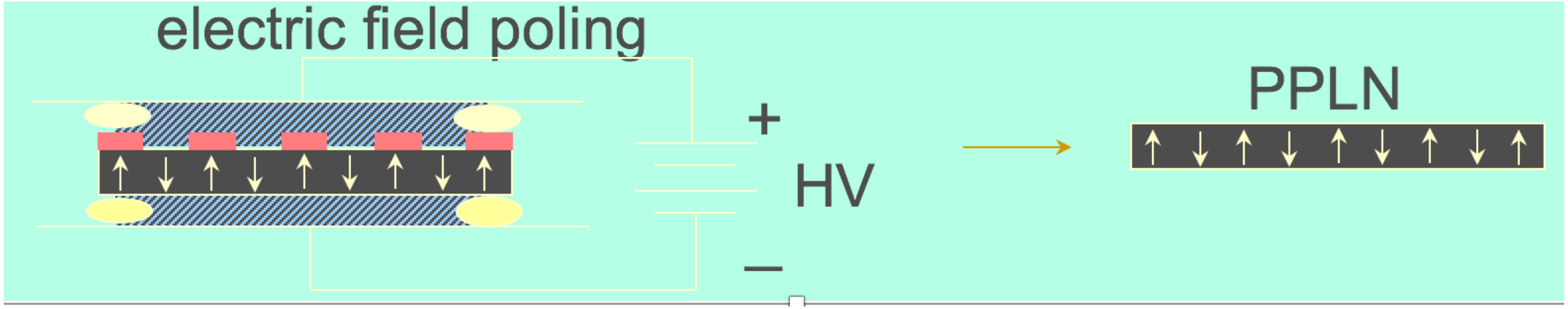
Ferroelectric Domain Reversal



If an applied field is larger than the potential energy barrier, then the Li ions will move through the potential energy barrier reversing the domain.

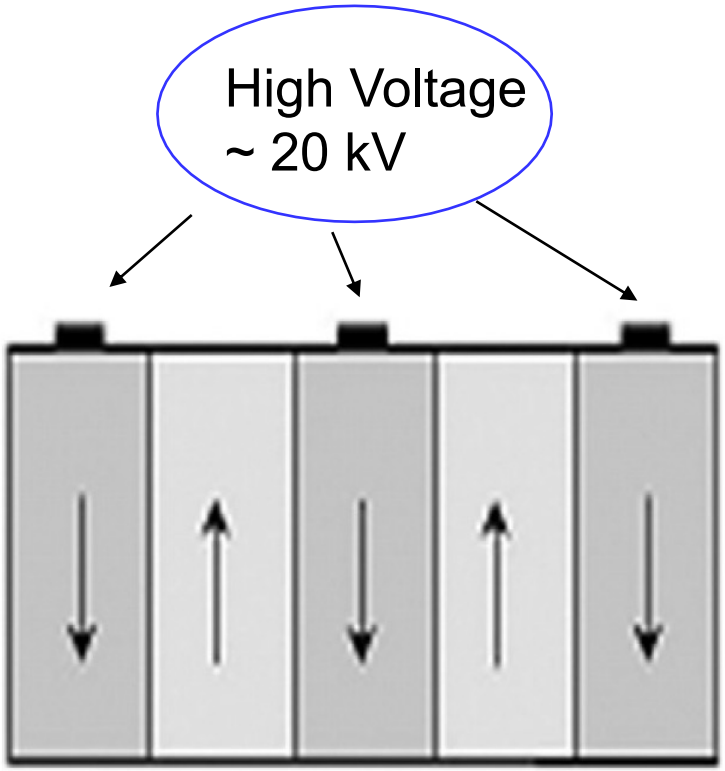
Quasi-phase matching (QPM)



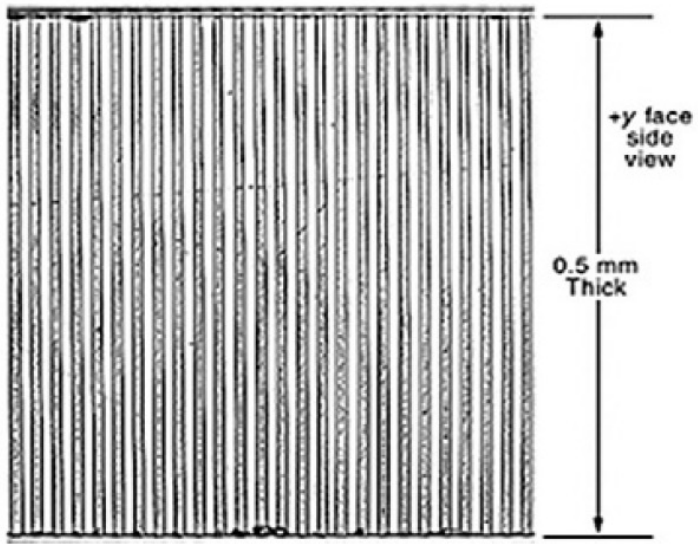


Quasi-phase matching (QPM)

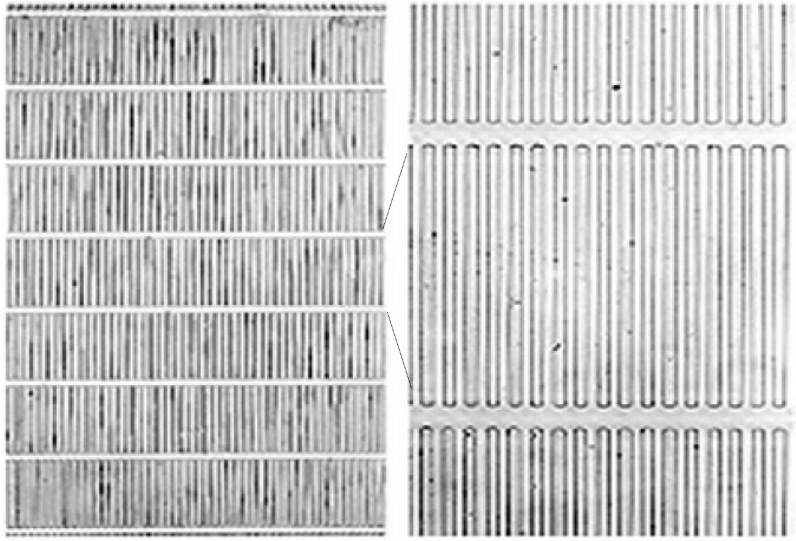
Lithium niobate (Stanford Univ.)



Domain patterns expand underneath the electrode region when electric fields larger than the coercive field are applied.



These 15-μm-period domains propagate through 0.5-mm-thick substrates; aspect ratios greater than 100:1 are observed.



Using photolithography allows patterning of multi-grating devices. Here the +z surface of a 0.5-mm-thick PPLN chip is revealed by etching with hydrofluoric acid. Each grating is 500-μm wide. The QPM periods are 29–30.5 μm

D.S. Hum, M.M. Fejer / C. R. Physique 8 (2007) 180–198

Quasi-phase matching (QPM)

The first, and still the most widely exploited of the PP ferroelectrics are the lithium niobate ($LiNbO_3$) and lithium tantalate ($LiTaO_3$) family.

Both are readily available in three- and four-inch diameter substrates, convenient for lithographic patterning and processing, and have established waveguide technologies compatible with periodic domain structures.

The d_{33} coefficient in LN, 28 pm/V, is the largest among commonly used ferroelectrics, while that of LT is somewhat smaller, 16 pm/V.

Both materials offer transparency from the near-UV (~ 300 nm) to the IR 4–5 μm

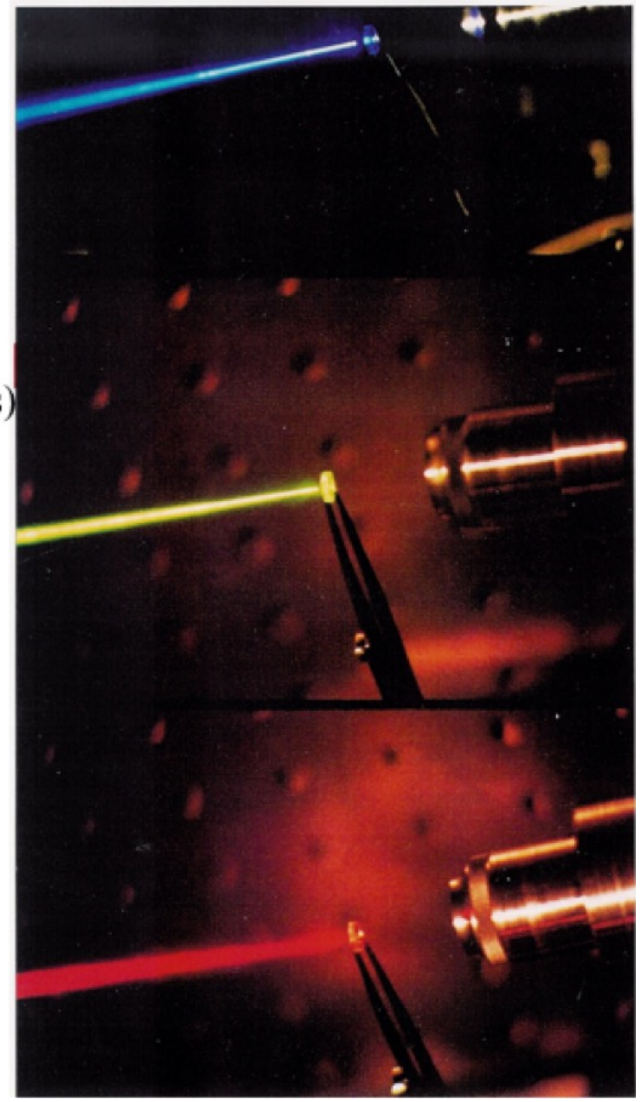
Quasi-phase matching (QPM)

State-of-the-art QPM $LiNbO_3$

-fundamental and harmonic co-polarized

- $\mathbf{d^{(2)}_{eff} \approx 16}$ **pm/V** (p=1) – (could be 25)
- samples up to **8 cms long**
- conversion efficiency → 1000%/W (waveguides)
- commercially available from many sources
- still some damage issues

Right-hand side picture shows blue, green-yellow and red beams obtained by doubling 0.82, 1.06 and 1.3 μm compact lasers in QPM $LiNbO_3$ or "PPLN"



Quasi-phase matching (QPM)

Benefits of QPM:

- utilize a large nonlinear coefficient (d_{33} in LN and LT)
- can work with crystals with weak or no birefringence at all (GaAs)
- no spatial walk-off
- all beams can be co-polarized (*type 0* phase matching)

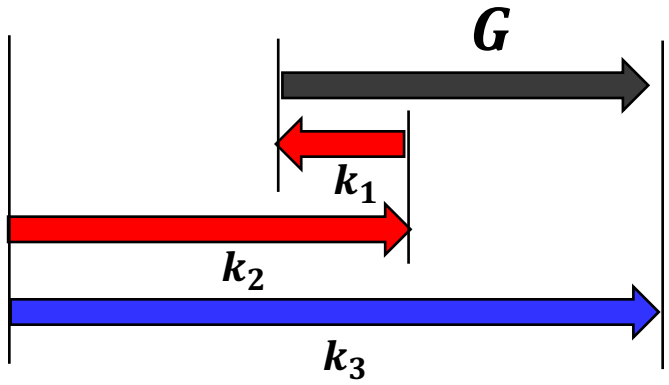
Limitations of QPM:

- possible with only certain crystal materials (LN, LT, KTP and its family, GaAs, GaP)
- limited thickness – excludes high power applications
- parasitic higher-order processes can generate light at additional frequencies

Quasi-phase matching (QPM)

Producing backward waves

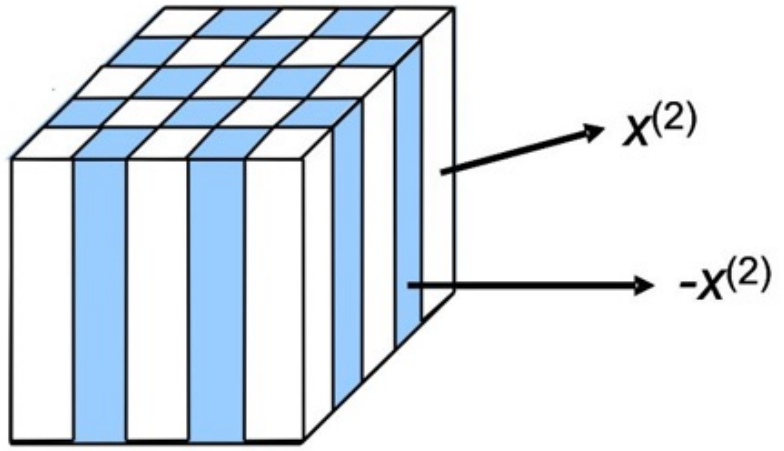
DFG



$$k_1 + k_2 + G = k_3$$

Quasi-phase matching (QPM)

Even more exotic scenarios: 2D QPM



2D Nonlinear Photonic Crystal

Other QPM Materials

Ferroelectrics [poled]

Semiconductors[patterned growth]

$LiTaO_3$

KTP

RTA

GaAs

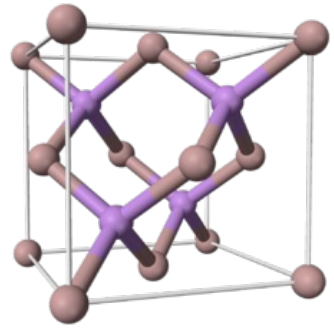
GaP

GaN

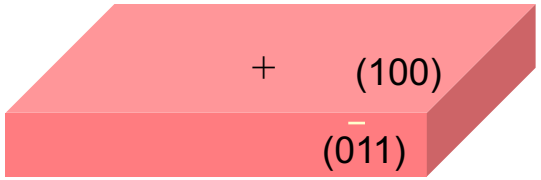
ZnSe ← early stage

First Practical QPM Semiconductor: Orientation-Patterned GaAs (OP-GaAs)

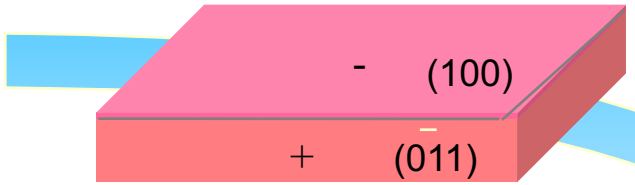
All-epitaxial growth pioneered at Stanford University[1,2]/University of Tokyo[3,4]



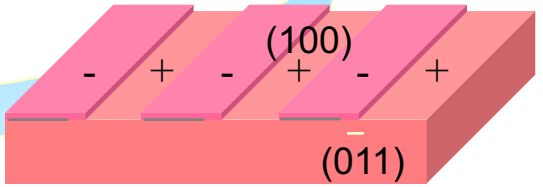
1) GaAs wafer



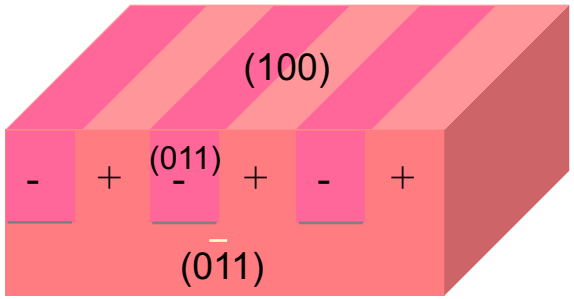
2) MBE growth of inverted GaAs on nonpolar Ge layer



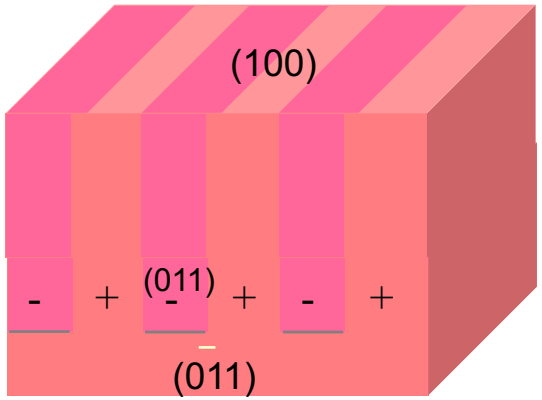
3) Photolithography & Etching

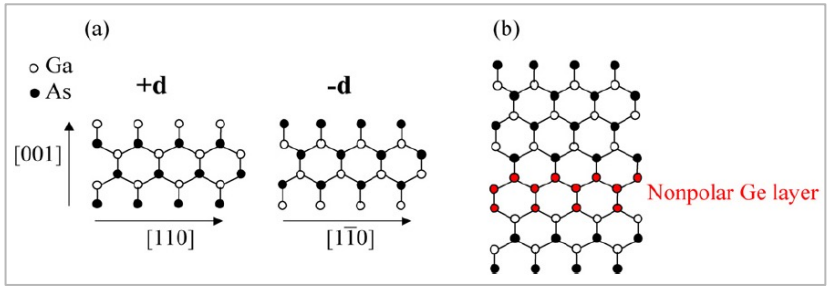


4) MBE regrowth of orientation-patterned film (2-3μm thick)



5) HVPE growth of thick-film device (>1000μm)





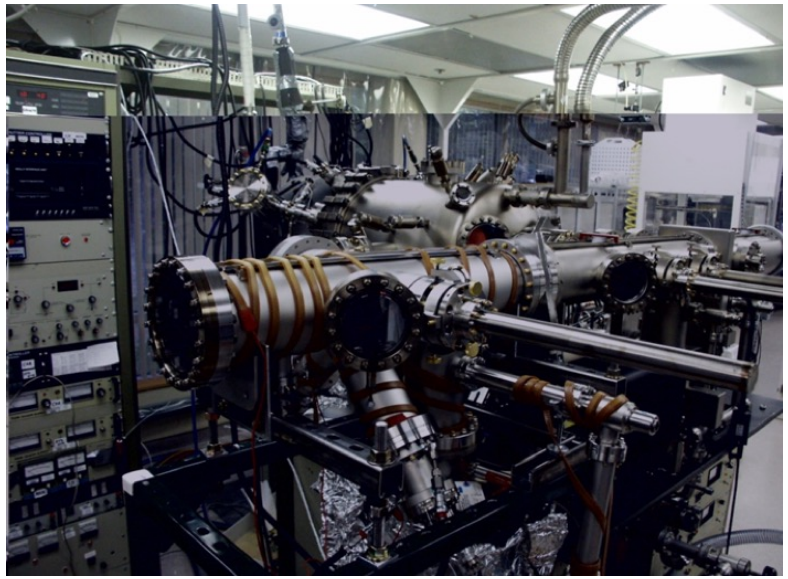
[1] C.B. Ebert, L.A. Eyres, M.M. Fejer, and J.S. Harris, J. Crystal Growth 201/202, 187 (1999).

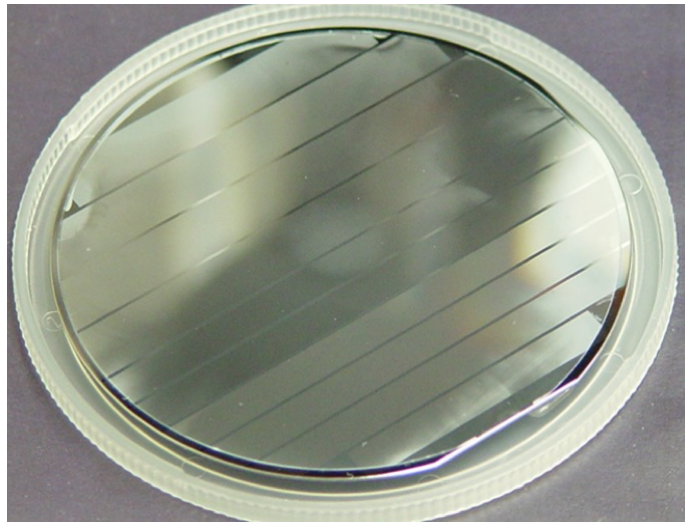
*[2] T. J. Pinguet et al., in OSA Trends in Optics and Photonics Vol. **56**, (Optical Society of America, Washington DC, 1998) pp. 226-229.*

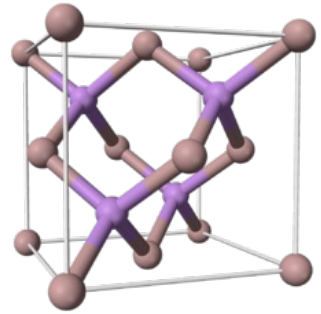
[3] S. Koh, T. Kondo, T. Ishiwada, C. Iwamoto, H. Ichinose, H. Yamaguchi, T. Usami, Y. Shiraki, and R. Ito, Jpn. J. Appl. Phys. 37 (1998) L1493.

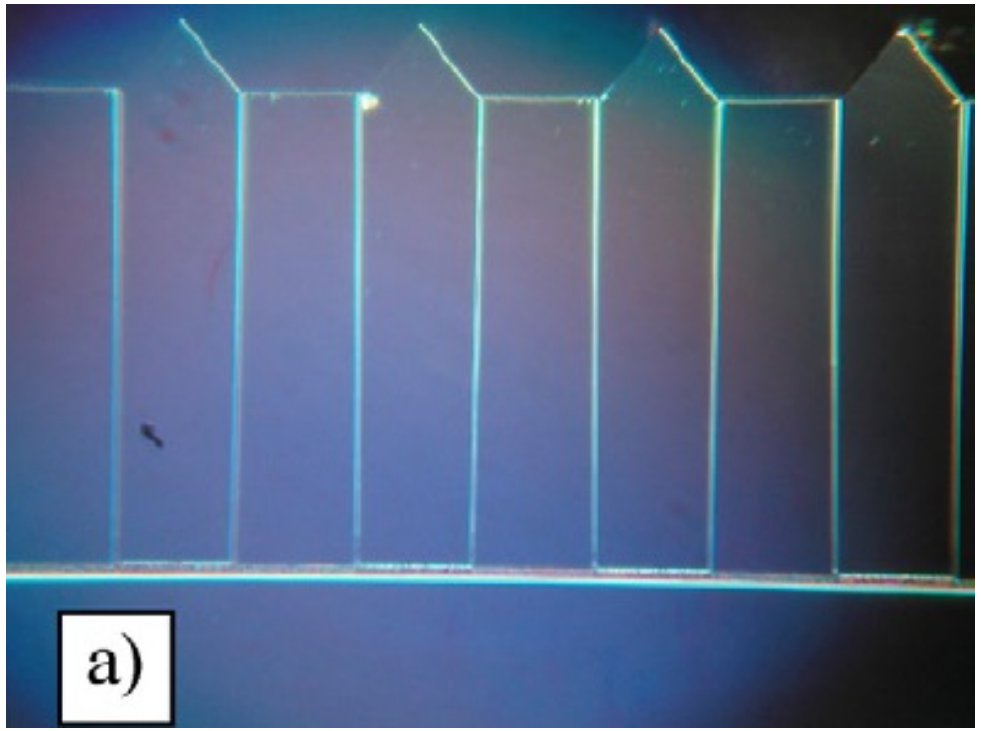
[4] S. Koh, T. Kondo, M. Ebihara, T. Ishiwada, H. Sawada, H. Ichinose, I. Shoji, and R. Ito, Jpn. J. Appl. Phys. 38 (1999) L508.

QPM GaAs: MBE and HVPE Growth





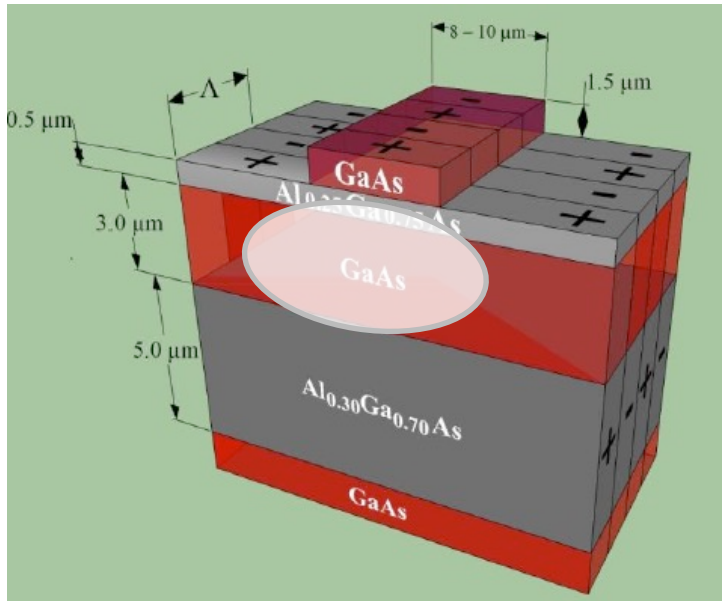




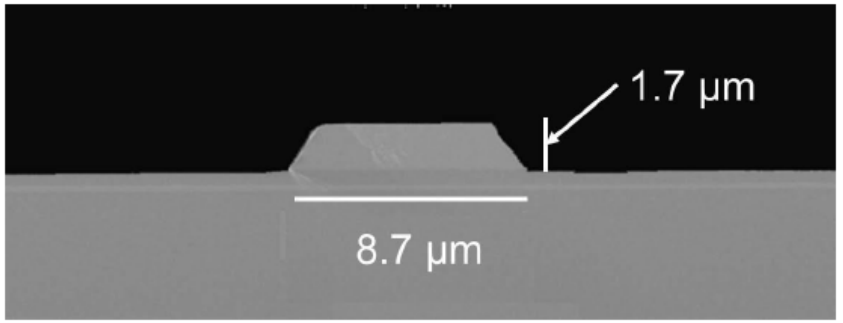
0.5–1 mm

Stain-etched cross-sections of OP-GaAs with 80-μm period

Waveguides in QPM Materials based on GaAs



MOCVD & Chemical Polish



Why use waveguides?

- Offers compatibility with fiber pump laser or QCL pump laser
  [no free-space coupling]
- Beam is tightly confined resulting in high intensity – thus high nonlinear gain
- high nonlinear gain over long lengths
- Thick material fabrication not needed

M.B. Oron, S. Shusterman and P. Blau, "Periodically oriented GaAs templates and waveguide structures for frequency conversion", SPIE Proceedings, Vol. 6875, 68750F-1 (2008).

QPM History

QPM was actually suggested [Bloembergen] as a phasematching technique before BPM.

The first demonstration* of QPM was done by cutting up crystals into coherence lengths and stacking them [1]. But there were disadvantages:

- thicknesses involved are a few microns so the slabs were difficult to fabricate
- added surfaces added significant loss making nonlinear conversion efficiencies poor [even a loss of 0.1% in 1000 interfaces yields a total loss of $1-0.999^{1000} = 63\%$!!]
- assembling a QPM device was labor intensive

Attempts were made to grow crystals with alternating domains. There was some success with $LiNbO_3$ but it was difficult to maintain the exact same periodicity for hundreds of domain thicknesses.

The **breakthrough** came with the discovery ~1995 that ferroelectric crystals can have their domains flipped by applying an electric field.

[1] *Szilagyi, Hordvik and Schlossberg, "A quasi-phase-matching technique for efficient optical mixing and frequency doubling," J of Appl Phys, **47** (5) 2025-2032 (1976).