# <u>6</u>

# calculation of $d_{\text{eff}}$
# $2^{\text{nd}}$ order NL mixing

SFG, DFG, SHG, OPA, OR…

Reading for this section:

1.5.6, 1.5.7, 1.5.11, 1.5.12

2.2, 2.5

Next time: 2.6-2.8

# Contracted notation: non-dispersive, non-absorbing medium

Non-dispersive: can permute any spatial index (Kleinmann symm)
- like colors have the same value, except black terms are unique

$$\begin{pmatrix} d_{xxx} & d_{xxy} & d_{xxz} \\ & d_{xyy} & d_{xyz} \\ & & d_{xzz} \end{pmatrix}$$

$$\begin{pmatrix} d_{xxy} & d_{xyy} & d_{xyz} \\ & d_{yyy} & d_{yyz} \\ & & d_{yzz} \end{pmatrix}$$

$$\begin{pmatrix} d_{xxz} & d_{xyz} & d_{xzz} \\ & d_{yyz} & d_{yzz} \\ & & d_{zzz} \end{pmatrix}$$

$$\begin{pmatrix} d_{xxx} & d_{xyy} & d_{xzz} & d_{xyz} & d_{xxz} & d_{xxy} \\ d_{xxy} & d_{yyy} & d_{yzz} & d_{yyz} & d_{xyz} & d_{xyy} \\ d_{xxz} & d_{yyz} & d_{zzz} & d_{yzz} & d_{xzz} & d_{xyz} \end{pmatrix}$$

$$\begin{pmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{pmatrix}$$

# Symmetry in d-matrices

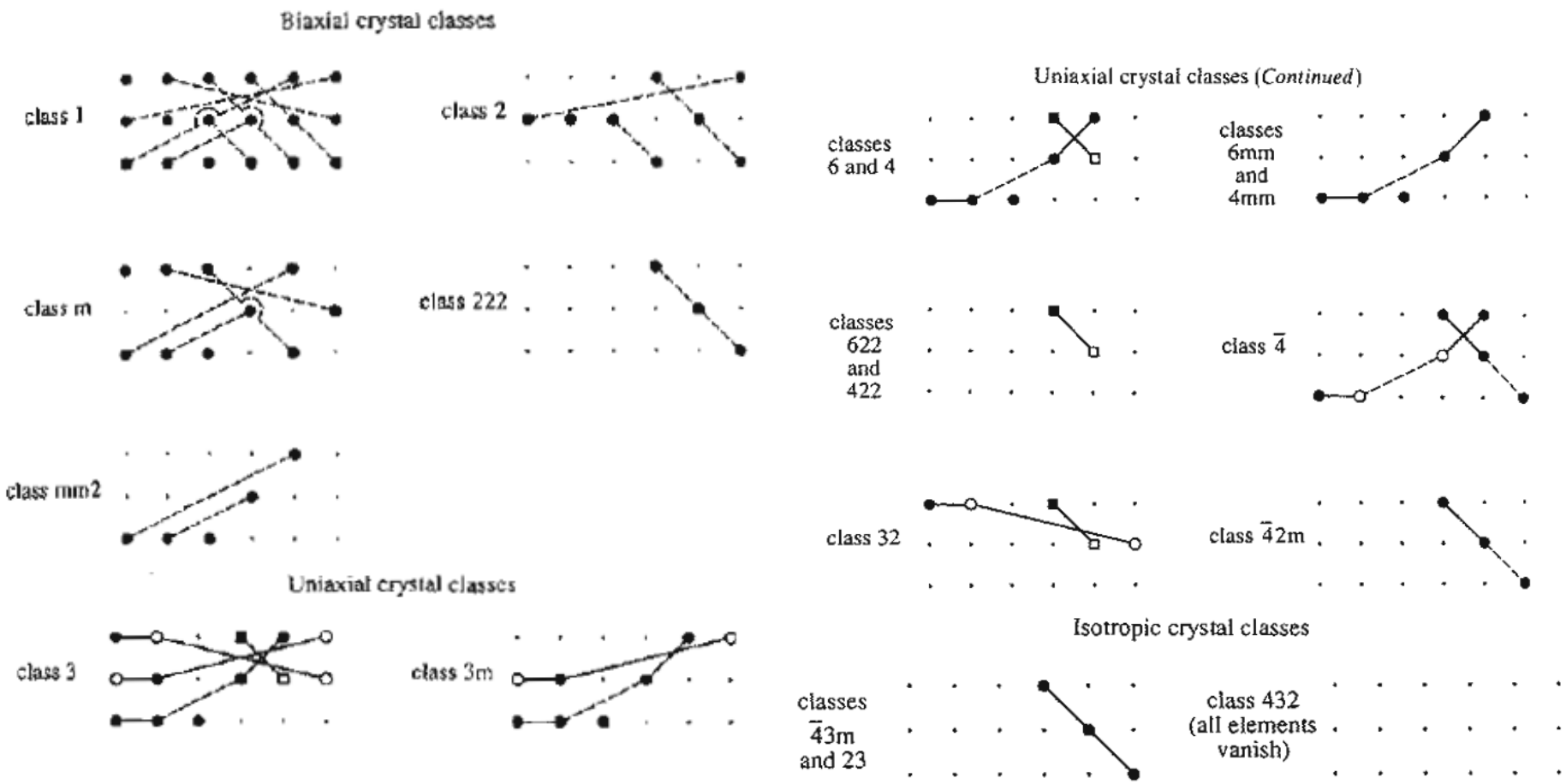



FIGURE 1.5.3 Form of the $d_{ij}$ matrix for the 21 crystal classes that lack inversion symmetry. Small dot: zero coefficient; large dot: nonzero coefficient; square: coefficient that is zero when Kleinman's symmetry condition is valid; connected symbols: numerically equal coefficients, but the open-symbol coefficient is opposite in sign to the closed symbol to which it is joined. Dashed connections are valid only under Kleinman's symmetry conditions. (After Zernike and Midwinter, 1973.)

# Effective NL coefficient

- *d*-matrix contains NL tensor coefficients for each crystal type
- Orientation of crystal affects effective strength
- Example: BBO, 3m symmetry

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_{1x}E_{2x} \\ E_{1y}E_{2y} \\ E_{1z}E_{2z} \\ E_{1z}E_{2y} + E_{1y}E_{2z} \\ E_{1z}E_{2x} + E_{1x}E_{2z} \\ E_{1y}E_{2x} + E_{1x}E_{2y} \end{pmatrix}$$

# Example calculation of $d_{\text{eff}}$

- For type I phase matching:
  - $\mathbf{E_1}$ and $\mathbf{E_2}$ are polarized in the same direction
  - Both are o-waves, so no projection on z-axis (optic axis)
  - Define **s** (= unit vector for **k**) in spherical coords,

$$\mathbf{s} = \hat{\mathbf{k}} = \{\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta\}$$

  - Find direction of **E** consistent with **k**:

$$\mathbf{E}\cdot\mathbf{s} = 0$$

$$E_1 \cos\phi_E \sin\theta\cos\phi + E_1 \sin\phi_E \sin\theta\sin\phi = 0$$

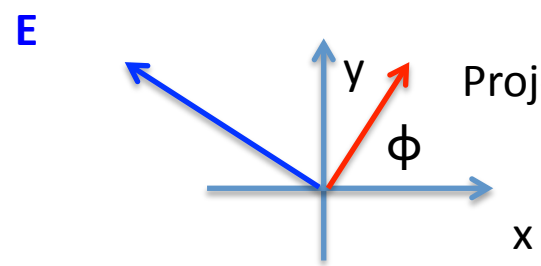
$$\cos\phi_E \cos\phi + \sin\phi_E \sin\phi = 0$$

$$\cos(\phi_E - \phi) = 0$$

$$\phi_E - \phi = \pi/2$$

$$E_1 = E_{10}\{-\sin\phi, \cos\phi, 0\}$$

$$E_2 = E_{20}\{-\sin\phi, \cos\phi, 0\}$$




Projection of **s** onto x-y

# Induced polarization

- Compose the $E_1E_2$ vector:
- The d-matrix is used to calculate the induced NL polarization

$$\begin{pmatrix} E_{1x}E_{2x} \\ E_{1y}E_{2y} \\ E_{1z}E_{2z} \\ E_{1z}E_{2y}+E_{1y}E_{2z} \\ E_{1z}E_{2x}+E_{1x}E_{2z} \\ E_{1y}E_{2x}+E_{1x}E_{2y} \end{pmatrix} = \begin{pmatrix} E_1E_2\sin^2\phi \\ E_1E_2\cos^2\phi \\ 0 \\ 0 \\ 0 \\ -E_1E_2\sin\phi\cos\phi \end{pmatrix}$$

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \begin{pmatrix} 0 & 0 & 0 & 0 & d_{31} & d_{22} \\ -d_{22} & d_{22} & 0 & d_{31} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} E_1E_2\sin^2\phi \\ E_1E_2\cos^2\phi \\ 0 \\ 0 \\ 0 \\ -E_1E_2\sin\phi\cos\phi \end{pmatrix} = \begin{pmatrix} d_{22}E_1E_2\sin 2\phi \\ d_{22}E_1E_2\cos 2\phi \\ d_{32}E_1E_2 \end{pmatrix}$$

# Finding the e-wave vector

- Note that NL polarization is induced at 2ω in all 3 directions

$$\begin{pmatrix} P_x \\ P_y \\ P_z \end{pmatrix} = \begin{pmatrix} d_{22}E_1E_2\sin 2\phi \\ d_{22}E_1E_2\cos 2\phi \\ d_{32}E_1E_2 \end{pmatrix}$$

- The generated e-wave unit vector **a** must be perpendicular to $\mathbf{E_1}$ and **s**

$$\mathbf{E_1}\cdot\mathbf{a} = 0 \rightarrow -E_{10}\sin\phi\, a_0 \sin\theta_a \cos\phi_a + E_{10}\cos\phi\, a_0 \sin\theta_a \sin\phi_a = 0$$

$$\sin\phi\cos\phi_a = \cos\phi\sin\phi_a \rightarrow \phi_a = \phi$$

$$\mathbf{a}\cdot\mathbf{s} = 0 \rightarrow a_0\cos\theta_a\cos\theta + a_0\sin\theta = 0$$

$$\cos\theta_a = -\tan\theta$$

- **a** is a unit vector, so from **a.a** = 1

$$a_0 = \cos\theta \qquad \rightarrow \mathbf{a} = \left\{ \cos\theta\cos\phi \quad \cos\theta\sin\phi \quad -\sin\theta \right\}$$

# Component of P that drives e-wave

- Pick out component that will drive a wave propagating in the direction of **k** ( = **s** ) and that is an e-wave

$$\mathbf{P}\cdot\mathbf{a} = d_{eff}E_1E_2 \rightarrow d_{eff} = d_{31}\sin\theta - d_{22}\cos\theta\sin 3\phi \qquad (1.5.30a)$$

- For type II,

$$d_{eff} = d_{22}\cos^2\theta\cos 3\phi \qquad (1.5.30b)$$

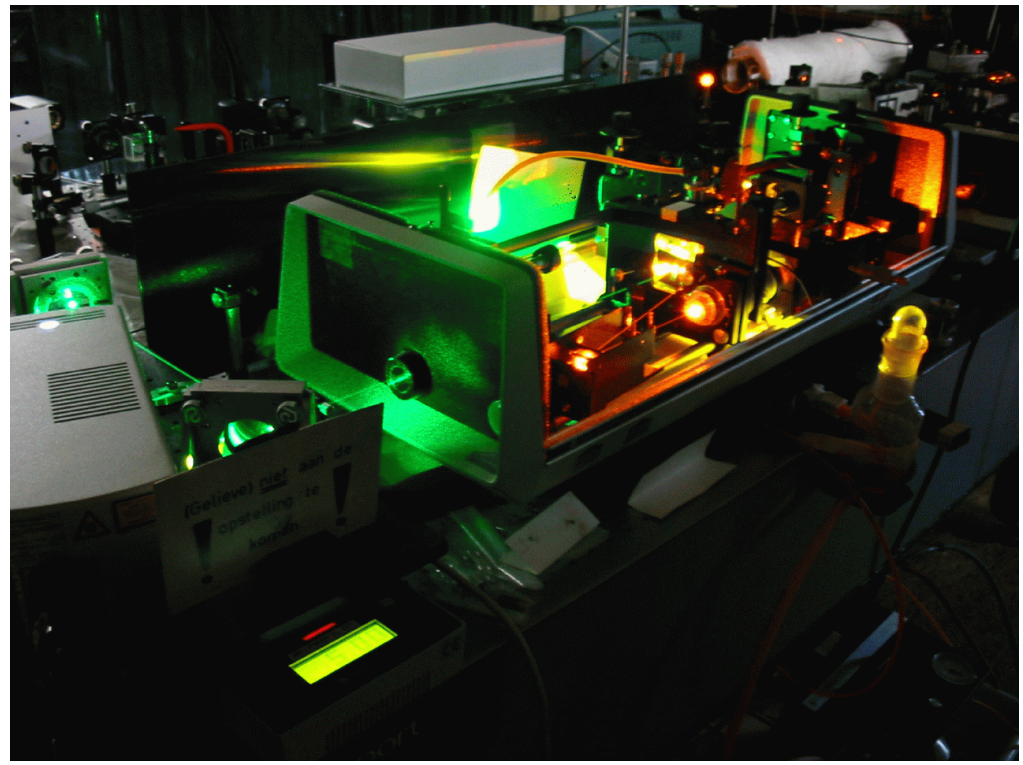
- Different relations for crystals with different symmetry
- These equations are used to optimize the orientation of the crystal for maximum signal
- Some directions that could be phase-matched don't have an induced polarization in the right direction

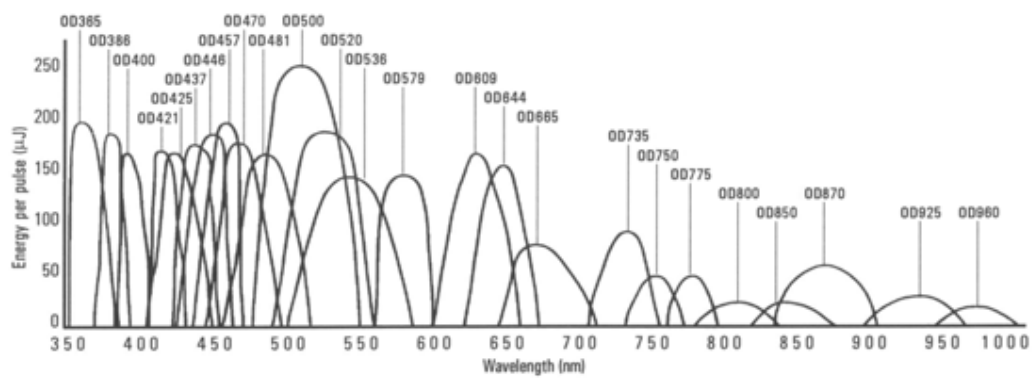
# Laser sources

- Many applications (industrial, commercial, scientific) require affordable, efficient lasers at particular wavelengths.
- But lasers aren't available at all wavelengths.
- [Laser wavelength chart]
- Dye lasers can make most wavelengths:

- messy, not compact, unhealthy, hard to change dye

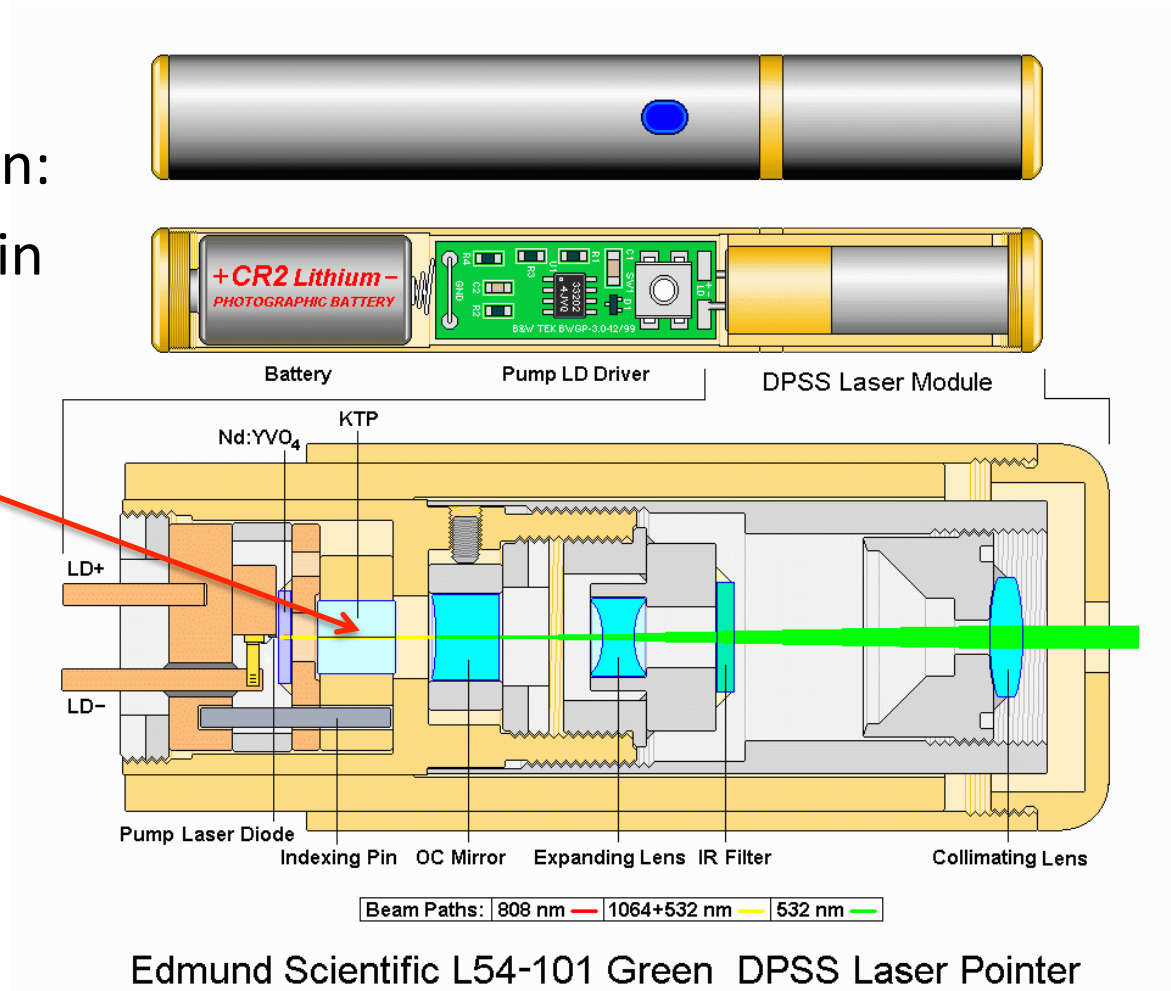
- Harmonic conversion and parametric amplification allow a solid-state alternative.





# Intracavity doubling: Green laser pointer

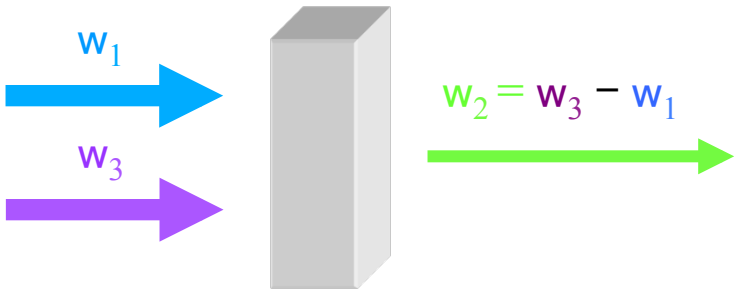
- Pump: laser diode
- Laser: Nd:vanadate
- Frequency conversion:
- Intracavity doubling in KTP
- Type II allow use of both polarization components of IR
- HR reflects IR and green
- OC reflects most IR, passes green



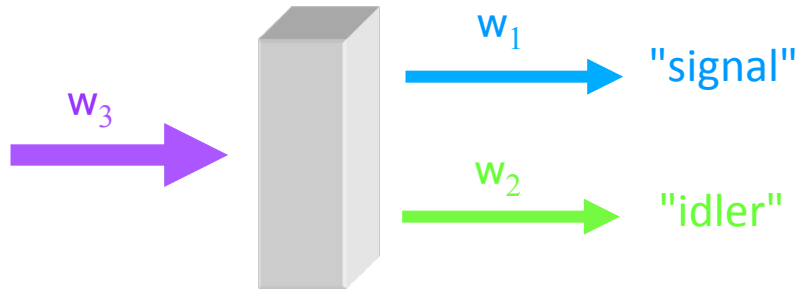

Edmund Scientific L54-101 Green DPSS Laser Pointer

# Difference-Frequency Generation: Optical Parametric Generation, Amplification, Oscillation

Difference-frequency generation takes many useful forms.

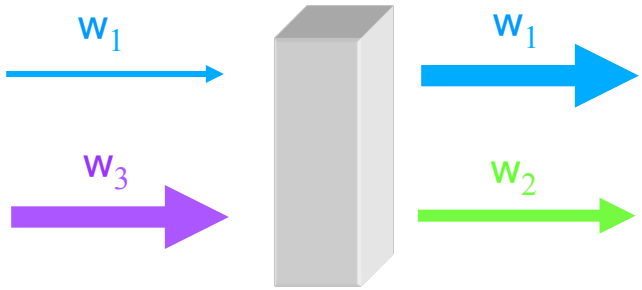



Parametric Down-Conversion
(Difference-frequency generation)

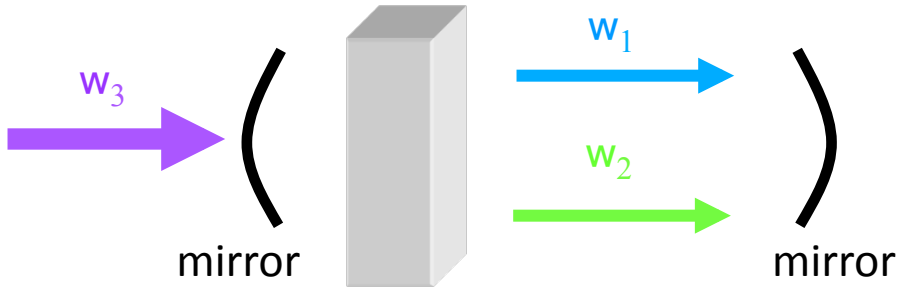



Optical Parametric Generation (OPG),
spontaneous down conversion

By convention:
$w_{signal} > w_{idler}$




Optical Parametric
Amplification (OPA)




Optical Parametric
Oscillation (OPO)

# Ultrafast rainbow: tunable ultrashort pulses from a solid-state kilohertz system

Kent R. Wilson and Vladislav V. Yakovlev

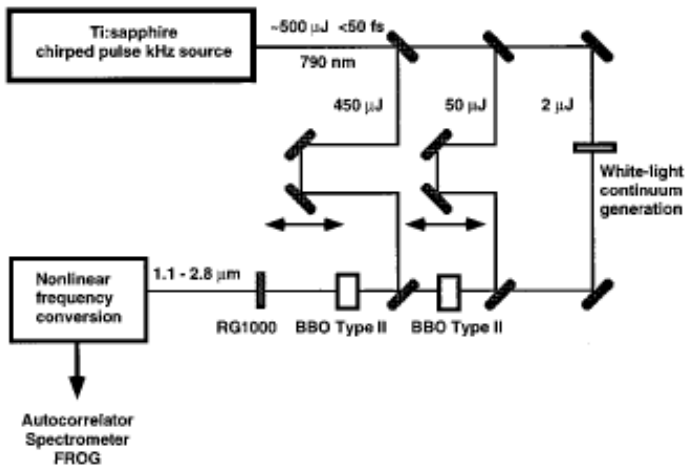



Fig. 3. Schematic of the experimental apparatus.

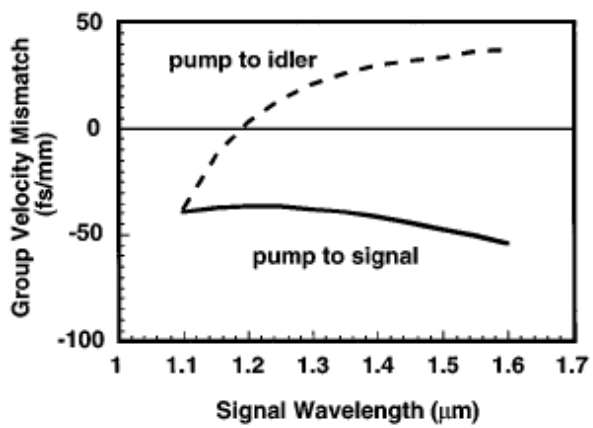



Fig. 1. Calculated group-velocity mismatch between pump and signal waves (solid curve) and pump and idler waves (dashed curve) for the 790-nm pump wavelength.

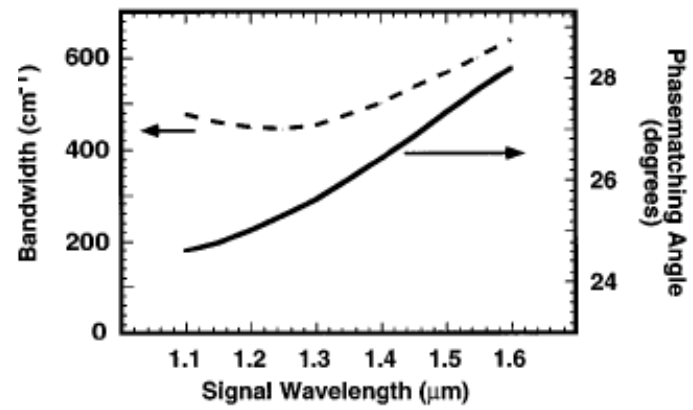



Fig. 2. Calculated external phase-matching angle (solid curve) and amplified bandwidth (dashed curve) for a 3-mm-long BBO crystal with a pump intensity of ~100 GW/cm$^2$.

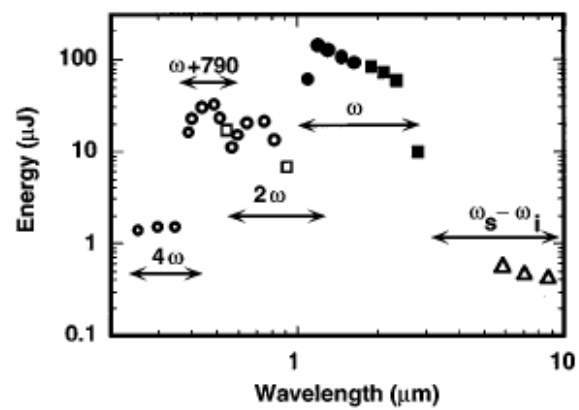


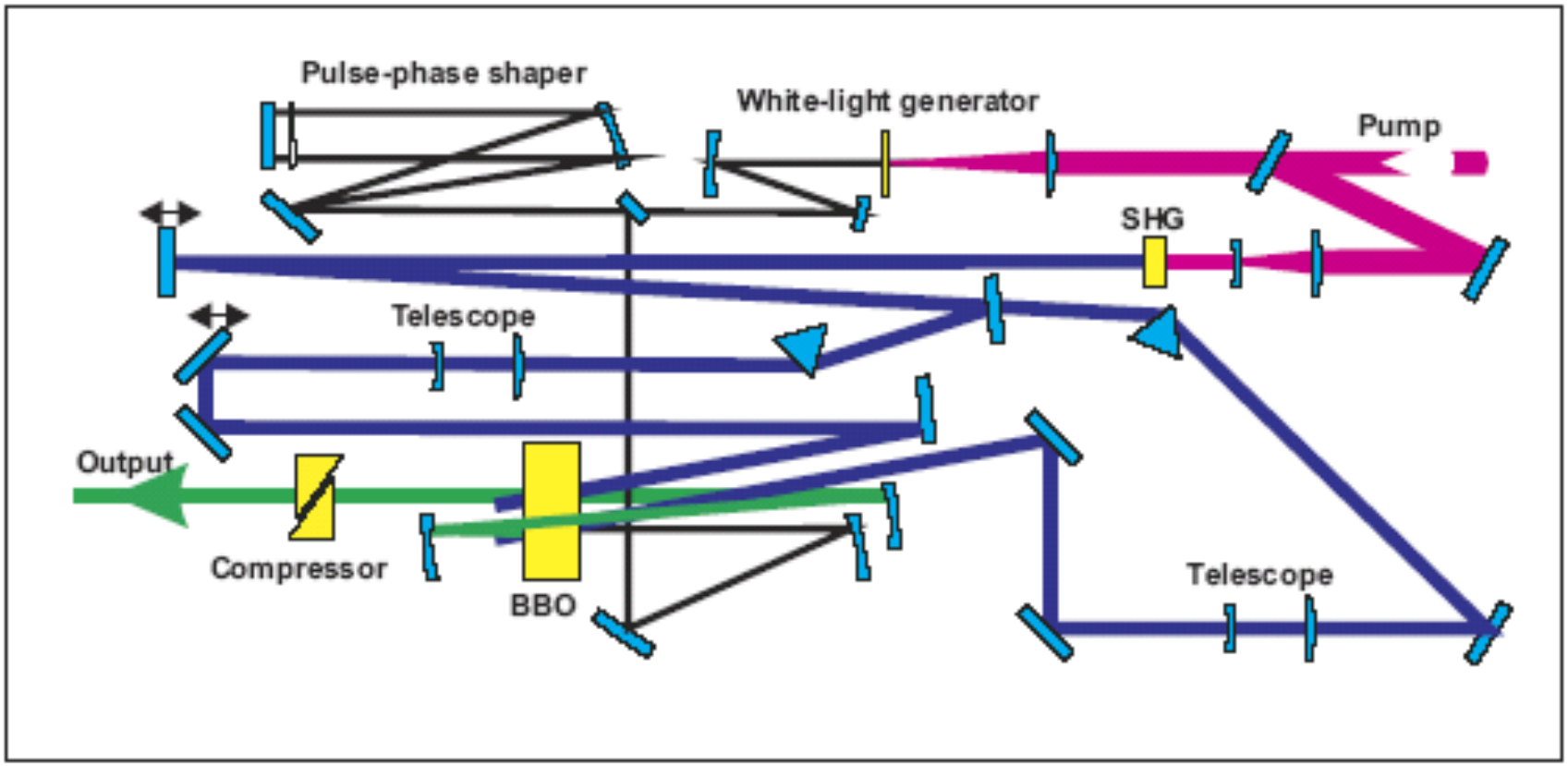

Fig. 7. Filled circles and filled squares show the measured energy of the amplified signal and idler waves, respectively. Open circles and open squares show the measured energy of the pulses generated by different nonlinear processes. $2\omega$, second harmonic of the signal (circles) and the idler (squares) (type I phase matching in 0.25-mm-thick BBO); $\omega + 790$: sum-frequency generation from the residual 790 nm and signal (type II phase matching in 0.2-mm-thick BBO); $4\omega$, fourth harmonic ($2\omega + 2\omega$) of the signal pulse (type II in 0.2-mm-thick BBO); open triangles, $\omega_S - \omega_I$: difference frequency generation (Type I in 1-mm-thick $AgGaS_2$).

# An ultrafast noncollinear OPA (NOPA)

LIGHT CONVERSION

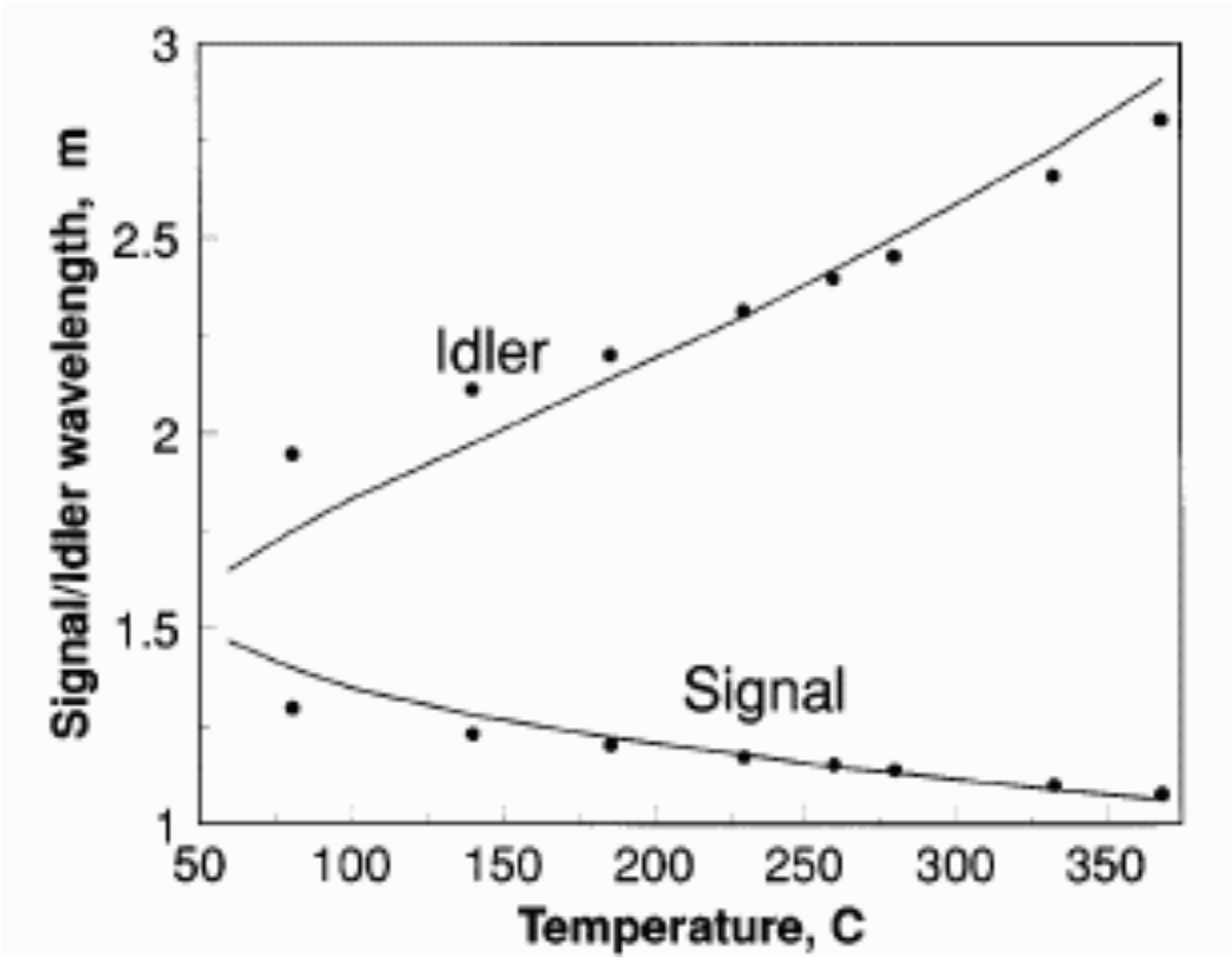
Continuum generates an arbitrary-color seed pulse.

TOPAS-white



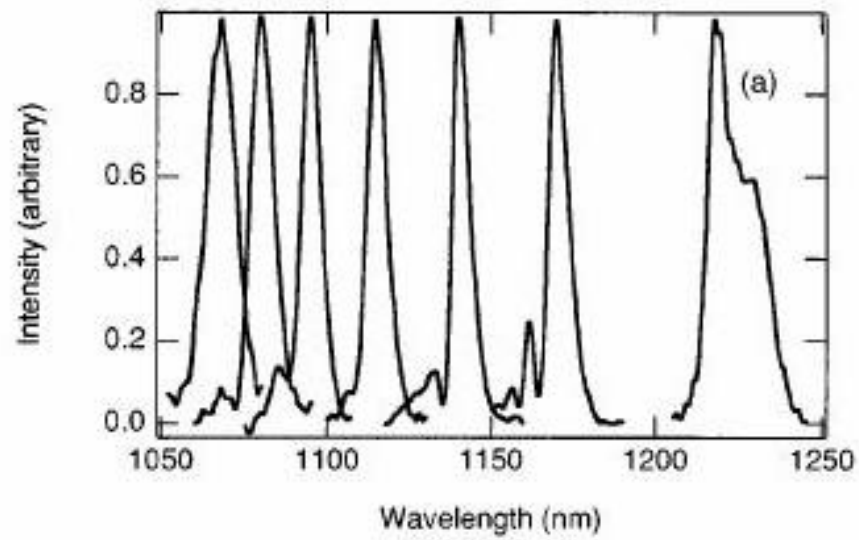
# Phase-matching applies.

We can vary the crystal angle in the usual manner, or we can vary the crystal temperature (since n depends on T).



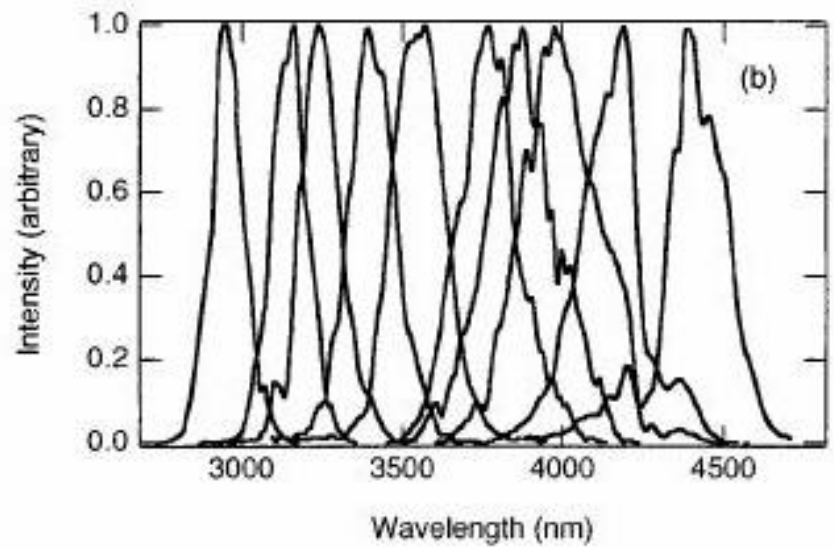
# Optical Parametric Generation

signal:




OPG with periodically poled $RbTiOAsO_4$

idler:

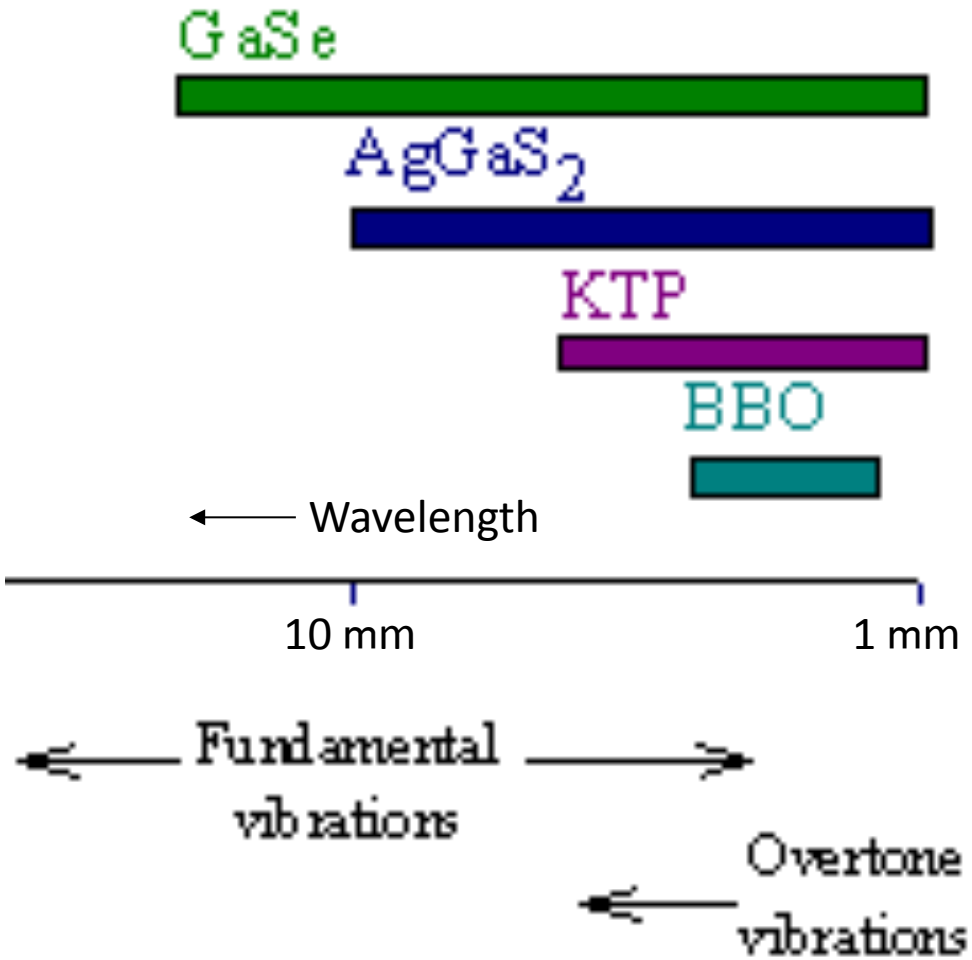



Sibbett, et al., Opt. Lett., **22**, 1397 (1997).

# Crystals for far-IR generation

With unusual crystals, such as $AgGaS_2$, $AgGaSe_2$ or GaSe, one can obtain radiation to wavelengths as long as 20 µm.

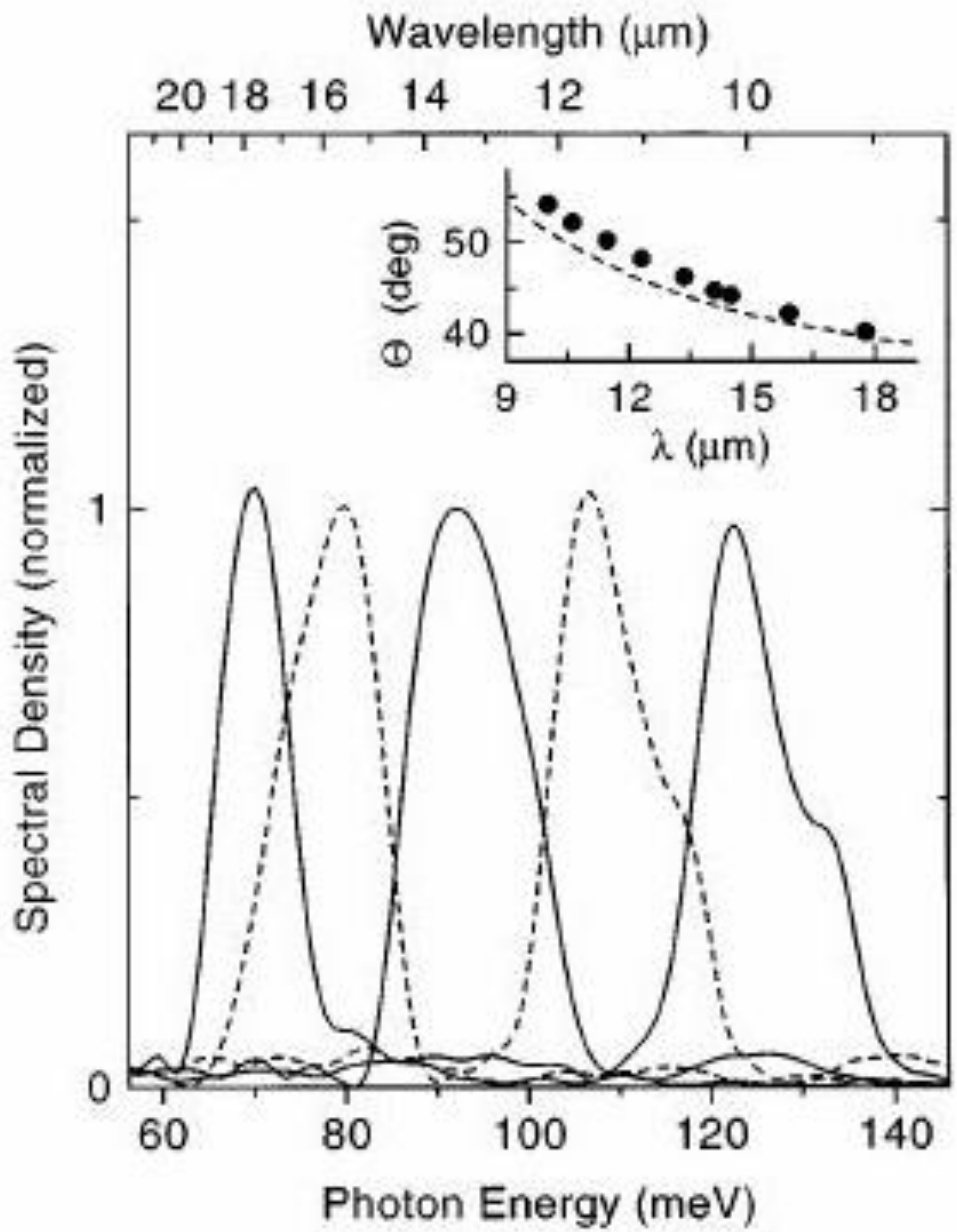
These long wavelengths are useful for vibrational spectroscopy.




Gavin D. Reid, University of Leeds, and Klaas Wynne, University of Strathclyde

# Difference-frequency generation in GaSe

Angle-tuned wavelength




Elsaesser, et al., *Opt. Lett.*, **23**, 861 (1998)

# NL coupled equations for 2nd order mixing

- NL equations valid for
  - Sum frequency mixing
  - Difference frequency mixing (OPA, OPO, SPDC)
  - Define by choosing initial conditions, assumptions about which waves are the strongest

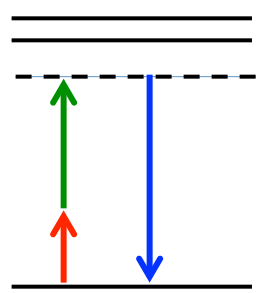
$$\frac{dA_1}{dz} = \frac{2\,i\,d_{eff}\,\omega_1^2}{k_1 c^2} A_3 A_2^* e^{-i\,\Delta k\,z}$$

$$\frac{dA_2}{dz} = \frac{2\,i\,d_{eff}\,\omega_2^2}{k_2 c^2} A_3 A_1^* e^{-i\,\Delta k\,z}$$

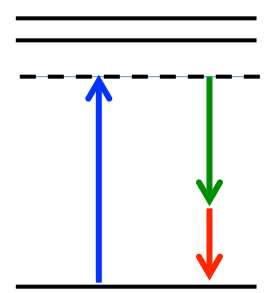
$$\frac{dA_3}{dz} = \frac{2\,i\,d_{eff}\,\omega_3^2}{k_3 c^2} A_1 A_2 e^{+i\,\Delta k\,z}$$

$$\Delta k = k_1 + k_2 - k_3 \qquad \omega_3 = \omega_1 + \omega_2$$

Sum frequency mixing



Difference frequency mixing



# Phase matching for parametric mixing

- For negative uniaxial: $n_e$ is lowest, so place $\omega_3$ as *e*-wave
- Type I: $\omega_1$ and $\omega_2$ parallel polarization, along *o*-direction
  - Generally broader bandwidth for OPA
- Type II: one of lower frequencies is along *e*-direction
  - can separate signal and idler with polarizer
- Quasi-phasematching (2.4), periodic poling

$$\Delta k = k_1 + k_2 - k_3$$
$$= \frac{1}{c}\left(\omega_1 n_1 + \omega_2 n_2 - \omega_3 n_3\right)$$

*this generalizes to a vector relation*

Note: for wave mixing, phase matching isn't just matching phase velocities or ref. indices. For SHG:

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

# Power conservation

- Manley-Rowe relations (2.5)

$$\frac{d}{dz}\left(\frac{I_1}{\omega_1}\right)=\frac{d}{dz}\left(\frac{I_2}{\omega_2}\right)=-\frac{d}{dz}\left(\frac{I_3}{\omega_3}\right)$$

- Can use these to reduce the number of coupled equations
- Helpful to understand saturated conversion limits in parametric processes