Second-harmonic generation, Phase-matching bandwidth, and Group velocity mismatch (GVM)

Phase-matching in SHG
Phase-matching bandwidth
Group-velocity mismatch
Nonlinear-optical crystals
Practical numbers for SHG
Electro-optics
Phase-matching in Second-Harmonic Generation

How does phase-matching affect SHG? It’s a major effect, one of the most important reasons why you just don’t see SHG every day.
First Demonstration of Second-Harmonic Generation


Figure 12.1. Arrangement used in the first experimental demonstration of second-harmonic generation [1]. A ruby-laser beam at $\lambda = 0.694 \, \mu m$ is focused on a quartz crystal, causing the generation of a (weak) beam at $\frac{1}{2}\lambda = 0.347 \, \mu m$. The two beams are then separated by a prism and detected on a photographic plate.

The second-harmonic beam was very weak because the process was not phase-matched.
First demonstration of SHG: The Data

The actual published results...

Note that the very weak spot due to the second harmonic is missing. It was removed by an overzealous Physical Review Letters editor, who thought it was a speck of dirt.
Sinusoidal Dependence of SHG Intensity on Length

Large $\Delta k$  

Small $\Delta k$

The SHG intensity is sharply maximized if $\Delta k = 0$. 
Phase-matching Second-Harmonic Generation

So we’re creating light at $\omega_{\text{sig}} = 2\omega$.

The k-vector of the second-harmonic is:

$$k_{\text{sig}} = \frac{\omega_{\text{sig}}}{c_0} n(\omega_{\text{sig}}) = \frac{(2\omega)}{c_0} n(2\omega)$$

And the k-vector of the polarization is:

$$k_{\text{pol}} = 2k = 2\frac{\omega}{c_0} n(\omega)$$

The phase-matching condition is:

$$k_{\text{sig}} = k_{\text{pol}}$$

which will only be satisfied when:

$$n(2\omega) = n(\omega)$$

Unfortunately, dispersion prevents this from ever happening!
Phase-matching Second-Harmonic Generation using Birefringence

Birefringent materials have different refractive indices for different polarizations. Ordinary and extraordinary refractive indices can be different by up to $\sim 0.1$ for SHG crystals.

We can now satisfy the phase-matching condition.

Use the extraordinary polarization for $\omega$ and the ordinary for $2\omega$.

$$n_o(2\omega) = n_e(\omega)$$

$n_e$ depends on propagation angle, so we can tune for a given $\omega$. Some crystals have $n_e < n_o$, so the opposite polarizations work.
Noncollinear SHG phase-matching

\[ \vec{k} = k \cos \theta \hat{z} - k \sin \theta \hat{x} \quad \rightarrow \quad \hat{G} \]

\[ \vec{k}' = k \cos \theta \hat{z} + k \sin \theta \hat{x} \quad \rightarrow \quad \omega \]

\[ \vec{k}_{pol} = \vec{k} + \vec{k}' = 2k \cos \theta \hat{z} \]

\[ \Rightarrow k_{pol} = 2 \frac{\omega}{c_0} n(\omega) \cos \theta \]

But:

\[ k_{sig} = \frac{2\omega}{c_0} n(2\omega) \]

So the phase-matching condition becomes:

\[ n(2\omega) = n(\omega) \cos \theta \]
Phase-Matching Bandwidth

Recall that the intensity out of an SHG crystal of length $L$ is:

$$I_{\text{sig}}(L) \propto \left(\frac{L}{\lambda}\right)^2 \text{sinc}^2\left(\frac{\Delta k L}{2}\right)$$

where:

$$\Delta k(\lambda) = \frac{4\pi}{\lambda} \left[n(\lambda) - n(\lambda/2)\right]$$

Phase-matching only works exactly for one wavelength, say $\lambda_0$. Since ultrashort pulses have lots of bandwidth, achieving approximate phase-matching for all frequencies is a big issue.

The range of wavelengths (or frequencies) that achieve approximate phase-matching is the phase-matching bandwidth.
Phase-matching efficiency vs. wavelength for the nonlinear-optical crystal, beta-barium borate (BBO), for different crystal thicknesses:

Note the huge differences in phase-matching bandwidth and efficiency with crystal thickness.

These curves also take into account the \((L/\lambda)^2\) factor.

While the curves are scaled in arbitrary units, the relative magnitudes can be compared among the three plots. (These curves don’t, however, include the nonlinear susceptibility, \(\chi^{(2)}\).)
Phase-matching efficiency vs. wavelength for KDP

Phase-matching efficiency vs. wavelength for the nonlinear-optical crystal, potassium dihydrogen phosphate (KDP), for different crystal thicknesses:

The huge differences in phase-matching bandwidth and efficiency with crystal thickness occur for all crystals.

The curves for the thin crystals don’t fall to zero at long wavelengths because KDP simultaneously phase-matches for two wavelengths, that shown and a longer (IR) wavelength, whose phase-matching ranges begin to overlap when the crystal is thin.
Calculation of Phase-Matching Bandwidth

The phase-mismatch is:

$$\Delta k(\lambda) = \frac{4\pi}{\lambda} \left[ n(\lambda) - n(\lambda / 2) \right]$$

Assuming the process is phase-matched at $\lambda_0$, let's see what the phase-mismatch will be at $\lambda = \lambda_0 + \delta\lambda$

$$\Delta k(\lambda) = \frac{4\pi}{\lambda_0} \left[ 1 - \frac{\delta\lambda}{\lambda_0} \right] \left[ n(\lambda_0) + \delta\lambda n'(\lambda_0) - n(\lambda_0 / 2) - \frac{\delta\lambda}{2} n'(\lambda_0 / 2) \right]$$

but the process is phase-matched at $\lambda_0$

because, when the input wavelength changes by $\delta\lambda$, the second-harmonic wavelength changes by only $\delta\lambda/2$.

$$\Delta k(\lambda) = \frac{4\pi \delta\lambda}{\lambda_0} \left[ n'(\lambda_0) - \frac{1}{2} n'(\lambda_0 / 2) \right] \quad \text{to first order in } \delta\lambda$$
The $\text{sinc}^2$ curve will decrease by a factor of 2 when $\Delta k L/2 = \pm 1.39$.

So solving for the wavelength range that yields $|\Delta k| < 2.78/L$ yields the phase-matching bandwidth.

\[-2.78/L < \frac{4\pi \delta \lambda}{\lambda_0} \left[ n'(\lambda_0) - \frac{1}{2} n'(\lambda_0 / 2) \right] < 2.78/L\]

\[\delta \lambda_{\text{FWHM}} = \frac{0.44 \lambda_0 / L}{|n'(\lambda_0) - \frac{1}{2} n'(\lambda_0 / 2)|}\]
Phase-Matching Bandwidth

The phase-matching bandwidth is usually too small, but it increases as the crystal gets thinner or the dispersion decreases (i.e., the wavelength approaches ~1.5 microns).

The theory breaks down, however, when the bandwidth approaches the wavelength.
Group-Velocity Mismatch

Inside the crystal the two different wavelengths have different group velocities.

Define the Group-Velocity Mismatch (GVM):

\[ GVM \equiv \frac{1}{v_g(\lambda_0/2)} - \frac{1}{v_g(\lambda_0)} \]

As the pulse enters the crystal:
- Second harmonic created just as pulse enters crystal (overlaps the input pulse)

As the pulse leaves the crystal:
- Second harmonic pulse lags behind input pulse due to GVM
Group-Velocity Mismatch

Calculating GVM:

\[ v_g(\lambda) = \frac{c_0}{n(\lambda)} \left( 1 - \frac{\lambda}{n(\lambda)} n'(\lambda) \right) \]

So:

\[ \frac{1}{v_g(\lambda)} = \frac{n(\lambda)}{c_0} \left[ 1 - \frac{\lambda}{n(\lambda)} n'(\lambda) \right] \]

\[ GVM \equiv \frac{1}{v_g(\lambda_0/2)} - \frac{1}{v_g(\lambda_0)} \]

\[ = \frac{n(\lambda_0/2)}{c_0} \left[ - \frac{\lambda_0}{n(\lambda_0/2)} n'(\lambda_0/2) \right] - \frac{n(\lambda_0)}{c_0} \left[ - \frac{\lambda_0}{n(\lambda_0)} n'(\lambda_0) \right] \]

But we only care about GVM when \( n(\lambda_0/2) = n(\lambda_0) \)

\[ GVM = \frac{\lambda_0}{c_0} \left[ n'(\lambda_0) - \frac{1}{2} n'(\lambda_0/2) \right] \]
Group-velocity mismatch lengthens the SH pulse.

Assuming that a very short pulse enters the crystal, the length of the SH pulse, $\delta t$, will be determined by the difference in light-travel times through the crystal:

$$\delta t = \frac{L}{v_g(\lambda_0/2)} - \frac{L}{v_g(\lambda_0)} = L \ GVM$$

We always try to satisfy: $L \ GVM \ll \tau_p$
Group-velocity mismatch pulse lengthening

Second-harmonic pulse shape for different crystal lengths:

\[ L_D \equiv \frac{\tau_p}{GVM} \]

\( L_D \) is the crystal length that doubles the pulse length.

It’s best to use a very thin crystal. Sub-100-micron crystals are common.
### Group-velocity mismatch numbers

<table>
<thead>
<tr>
<th>Crystal</th>
<th>$\lambda$ [nm]</th>
<th>$\theta$ [$^\circ$]</th>
<th>$(v_2^{-1} - v_1^{-1})$ [fs/mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>KDP</td>
<td>550</td>
<td>71</td>
<td>266</td>
</tr>
<tr>
<td></td>
<td>620</td>
<td>58</td>
<td>187</td>
</tr>
<tr>
<td></td>
<td>800</td>
<td>45</td>
<td>77</td>
</tr>
<tr>
<td></td>
<td>1000</td>
<td>41</td>
<td>9</td>
</tr>
<tr>
<td>LiIO$_3$</td>
<td>620</td>
<td>61</td>
<td>920</td>
</tr>
<tr>
<td></td>
<td>800</td>
<td>42</td>
<td>513</td>
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<td></td>
<td>1000</td>
<td>32</td>
<td>312</td>
</tr>
<tr>
<td>BBO</td>
<td>500</td>
<td>52</td>
<td>680</td>
</tr>
<tr>
<td></td>
<td>620</td>
<td>40</td>
<td>365</td>
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<tr>
<td></td>
<td>1000</td>
<td>24</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td>1500</td>
<td>20</td>
<td>5</td>
</tr>
</tbody>
</table>
Group-velocity mismatch limits bandwidth.

Let’s compute the second-harmonic bandwidth due to GVM.

Take the SH pulse to have a Gaussian intensity, for which \( \delta t \delta \nu = 0.44 \).

Rewriting in terms of the wavelength,

\[
\delta t \delta \lambda = \delta t \delta \nu \left[ \frac{dn}{d\lambda} \right]^{-1} = 0.44 \left[ \frac{dn}{d\lambda} \right]^{-1} = 0.44 \frac{\lambda^2}{c_0}
\]

where we’ve neglected the minus sign since we’re computing the bandwidth, which is inherently positive. So the bandwidth is:

\[
\delta \lambda_{FWHM} \approx \frac{0.44 \lambda_0^2 / c_0}{\delta t} = \frac{0.44 \lambda_0^2 / c_0}{L \ GVM}
\]

where

\[
GVM = \frac{\lambda_0}{c_0} \left[ n'(\lambda_0) - \frac{1}{2} n'(\frac{\lambda_0}{2}) \right]
\]

Calculating the bandwidth by considering the GVM yields the same result as the phase-matching bandwidth!
The second-harmonic field is given by:

\[ E^{2\omega}(L,t) = -i \frac{\mu_0 \omega^2 L}{2k} P \exp(i\Delta kL/2) \text{sinc}(\Delta kL/2) \]

The irradiance will be:

\[ I^{2\omega} = \frac{\eta_0 \omega^2 (\chi^{(2)})^2 (I^{\omega})^2 L^2}{2c_0^2 n^3} \text{sinc}^2(\Delta kL/2) \]

Dividing by the input irradiance to obtain the SHG efficiency:

\[ \frac{I^{2\omega}}{I^{\omega}} = \frac{2\eta_0 \omega^2 d^2 I^{\omega} L^2}{c_0^2 n^3} \]

Take \( \Delta k = 0 \)

\( d \propto \chi^{(2)} \), and includes crystal additional parameters.

Substituting in typical numbers:

\[ \frac{I^{2\omega}}{I^{\omega}} \approx \left[ 5 \times 10^{-8} / W \right] I^{\omega} L^2 \]
Amazing second-harmonic generation

Frequency-doubling KDP crystals at Lawrence Livermore National Laboratory

These crystals convert as much as 80% of the input light to its second harmonic. Then additional crystals produce the third harmonic with similar efficiency!

These guys are serious!
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)

By convention: \( \omega_{\text{signal}} > \omega_{\text{idler}} \)

Optical Parametric Amplification (OPA)

Optical Parametric Oscillation (OPO)
Optical Parametric Generation

Equations are just about identical to those for SHG:

\[
\begin{align*}
\left( \frac{\partial}{\partial z} + \frac{1}{v_{g1}} \frac{\partial}{\partial t} \right) \tilde{E}_1 &= -i \chi^{(2)} \frac{\omega_1^2}{2c^2k_1} \tilde{E}_2^* \tilde{E}_3 e^{i \Delta k \cdot z} \\
\left( \frac{\partial}{\partial z} + \frac{1}{v_{g2}} \frac{\partial}{\partial t} \right) \tilde{E}_2 &= -i \chi^{(2)} \frac{\omega_2^2}{2c^2k_2} \tilde{E}_1^* \tilde{E}_3 e^{i \Delta k \cdot z} \\
\left( \frac{\partial}{\partial z} + \frac{1}{v_{g3}} \frac{\partial}{\partial t} \right) \tilde{E}_3 &= -i \chi^{(2)} \frac{\omega_3^2}{2c^2k_3} \tilde{E}_1 \tilde{E}_2 e^{-i \Delta k \cdot z}
\end{align*}
\]

where:
- \( k_i \) = wave vector of \( i^{th} \) wave
- \( \Delta k = k_1 + k_2 - k_3 \)
- \( v_{g i} \) = group velocity of \( i^{th} \) wave

The solutions for \( E_1 \) and \( E_2 \) involve exponential gain!

OPA’s etc. are ideal uses of ultrashort pulses, whose intensities are high.
Phase-matching applies.

We can vary the crystal angle in the usual manner, or we can vary the crystal temperature (since \( n \) depends on \( T \)).
Free code to perform OPO, OPA, and OPG calculations
Optical Parametric Generation

Recent results using the nonlinear medium, periodically poled RbTiOAsO$_4$

An Ultrafast Noncollinear OPA (NOPA)

Continuum generates an arbitrary-color seed pulse.
PERFORMANCE SPECIFICATIONS WITH 0.5 mJ PUMP PULSES

**SIGNAL OUTPUT (all modifications)**

<table>
<thead>
<tr>
<th>Modification</th>
<th>Tuning ranges (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>TOPAS-white</td>
<td>490-750, 850-1000</td>
</tr>
<tr>
<td>TOPAS-white-SH</td>
<td>245-375, 425-490</td>
</tr>
<tr>
<td></td>
<td>490-750, 850-1000</td>
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<tr>
<td>TOPAS-white-IR</td>
<td>490-750</td>
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<tr>
<td></td>
<td>850-1700</td>
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<tr>
<td>TOPAS-white-IR-SH</td>
<td>245-375, 425-490</td>
</tr>
<tr>
<td></td>
<td>490-750</td>
</tr>
<tr>
<td></td>
<td>700-850</td>
</tr>
<tr>
<td></td>
<td>850-1700</td>
</tr>
</tbody>
</table>

**Tuning range**

- 490 - 750 nm
- 850 - 1000 nm

**Pulse energy**

- $\geq 30 \, \mu J \, @ \, 550 \, nm$
- $\geq 20 \, \mu J \, @ \, 700 \, nm$

**Pulse duration, \text{sech}^2**

- $\leq 20 \, fs \, @ \, 530 - 720 \, nm$
- $\leq 60 \, fs \, @ \, 490 - 530 \, nm$
- $\leq 60 \, fs \, @ \, 720 - 1000 \, nm$

**Energy instability**

- $\leq 1.5\% \, \text{rms} - 5\% \, \text{rms}$
  (depending on the wavelength and the input stability)

**Instability of pulse duration**

- $\leq 2\% \, \text{rms}$

**Pre-pulse contrast**

- $\leq 10^{-3}$

**Spatial profile**

- Super-gaussian, $M^2 \leq 1.5$
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

Another 2\textsuperscript{nd}-order process: Electro-optics

Applying a voltage to a crystal changes its refractive indices and introduces birefringence. In a sense, this is sum-frequency generation with a beam of zero frequency (but not zero field!).

A few kV can turn a crystal into a half- or quarter-wave plate.

If $V = 0$, the pulse polarization doesn’t change.

If $V = V_p$, the pulse polarization switches to its orthogonal state.

Abruptly switching a Pockels cell allows us to switch a pulse into or out of a laser.
The Pockels’ Cell (Q-Switch)

The Pockels effect is a type of second-order nonlinear-optical effect.

The Pockels effect involves the simple second-order process:

\[ \omega_{\text{sig}} = \omega + 0 \]

The signal field has the orthogonal polarization, however.