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phase matching

time domain representation of 2nd order response

ideal SHG output spectrum

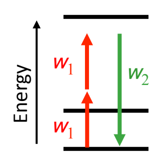
Phase matching: an important application for the the angle-dependent refractive index

Recall that for SHG:

$$\frac{\partial A_2}{\partial z} e^{i(k_2 z - \omega_2 t)} = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 e^{i(2k_1 z - 2\omega_1 t)}$$

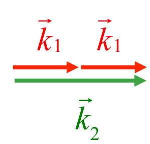
Energy *must* be conserved:

$$\omega_1 + \omega_1 = \omega_2 \quad \Rightarrow \quad \omega_2 = 2\omega_1$$



Momentum ***may or may not*** be conserved:

$$\frac{\partial A_2}{\partial z} = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 e^{i\Delta k z} \quad \text{where} \quad \Delta k = 2k_1 - k_2$$



Conversion will be most efficient if $\Delta k = 0$

$$\Rightarrow \quad 2\frac{\omega_1}{c_0} n(\omega_1) = \frac{2\omega_1}{c_0} n(2\omega_1)$$

$\boxed{n(\omega_1) = n(2\omega_1)}$ This is the ***phase-matching condition*** for SHG

Real crystal dispersion data

- Best resource: refractiveindex.info
- Others: crystal manufacturers, Handbook of Optics

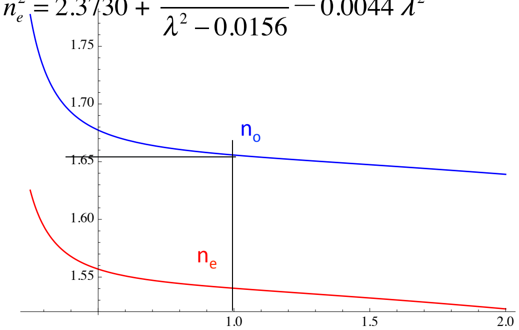
Example: β-BBO = barium borate, BaB_2O_4

$$n_o^2 = 2.7405 + \frac{0.0184}{\lambda^2 - 0.0179} - 0.0155\ \lambda^2$$

λ is in micrometers!

$$n_e^2 = 2.3730 + \frac{0.0128}{\lambda^2 - 0.0156} - 0.0044\ \lambda^2$$

$n_e < n_o$ everywhere, so we need to angle tune



λ (µm)

Type I phase matching
Input at 1µm, SH at 0.5µm
Input along n_o
Output can be tuned
$n_e < n_e(\theta) < n_o$

Types of phase matching

- Type 1:
  - 2ω on low index (n_e)
  - ω on high (n_o)
  - Opposite polarizations ($\chi^{(2)}$ tensor allows this)

$$\Delta k = 2\frac{\omega_1}{c} n_o(\omega_1) - \frac{\omega_2}{c} n_e(\omega_2,\theta) = 2\frac{\omega_1}{c}\left(n_o(\omega_1) - n_e(\omega_2,\theta)\right)$$

- Type 2:
  - 2ω on low index (n_e)
  - Project E_1 equally on both axes (n_o and n_e)

$$\Delta k = \frac{\omega_1}{c} n_o(\omega_1) + \frac{\omega_1}{c} n_e(\omega_1,\theta) - \frac{\omega_2}{c} n_e(\omega_2,\theta)$$

- Type 3:
  - "non-critical" or "90°" phase matching
  - Temperature-tuned
  - Only for particular crystals and wavelengths

$$\Delta k = 2\frac{\omega_1}{c}\left(n_o(\omega_1,T) - n_e(\omega_2,90,T)\right)$$

SHG without phase-matching

Non-depleted pump approximation: treat A_1 as constant

$$\frac{\partial A_2}{\partial z} = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 e^{i\Delta k z}$$ Integrate: $$A_2(L) = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 \int_0^L e^{i\Delta k z}\, dz$$

$$A_2(L) = i\frac{\omega_2^2 d}{k_2 c^2} A_1^2 L \frac{\left(e^{i\Delta k L} - 1\right)}{i\Delta k L}$$

Convert to intensity $I_2 = 2\varepsilon_0 n_2 c |A_2|^2$

$$\rightarrow \frac{1}{2\varepsilon_0 n_2 c} I_2(z) = \left(\frac{1}{2\varepsilon_0 n_1 c}\right)^2 I_1^2 \left(\frac{\omega_2 d}{n_2 c}\right)^2 L^2 \left(\frac{\sin(\Delta k L/2)}{\Delta k L/2}\right)^2$$

$$\rightarrow I_2(L) = \frac{\omega_2^2 d^2}{2\varepsilon_0 n_1^2 n_2 c^3} I_1^2 L^2 \,\mathrm{sinc}^2(\Delta k L/2)$$

As a function of L and fixed |Δk|>0: $$I_2(L) = \frac{\omega_2^2 d^2}{2\varepsilon_0 n_1^2 n_2 c^3} I_1^2 \frac{4}{\Delta k^2} \sin^2(\Delta k L/2)$$

Yield oscillates:

- Period = "coherence length" $L_{coh} = 2\pi/\Delta k$
- Amplitude proportional to $\max(I_2) \propto 1/\Delta k^2$

Practical issues

- Phase matching bandwidth
  - Type 1 has more BW, choose L of crystal
- Group velocity walk-off (for short pulses)
- Angular acceptance
- Birefringent beam walk-off
- Strength of nonlinearity
- Crystal damage threshold
- Thermal stability:
  - typically angle-tuned, temperature stabilized
- Available size of crystals, $$

Inversion symmetry

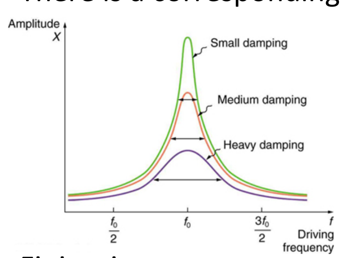
- If a material has inversion symmetry, then $\chi^{(2)}$ (and all even orders) must be zero.
  - Suppose we have a 2nd order response, $P^{(2)}(t) = \varepsilon_0 \chi^{(2)} E^2(t)$
  - Driven by a wave: $E(t) = E_0 \cos\omega t$
  - With inversion symmetry, changing the sign of E(t) should change sign of $P^{(2)}(t)$

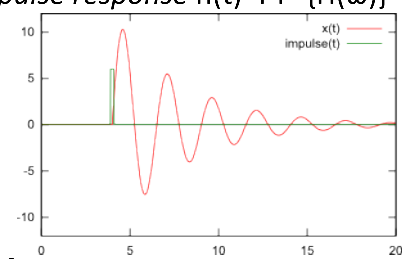
$$-P^{(2)}(t) = \varepsilon_0 \chi^{(2)} \left(-E(t)\right)^2 = \varepsilon_0 \chi^{(2)} E^2(t)$$

  - This cannot be true, so such a material can't have $\chi^{(2)}$

Time-domain representation of linear response

- For linear response, we can calculate response of medium as fcn of ω. This is the *frequency response*, H(ω)
- There is a corresponding *impulse response* $h(t) = FT^{-1}\{H(\omega)\}$





- Finite time response results from resonance
- ***Causal***: h(t)=0 for t<0
- Linear response: $R^{(1)}(t) = \mathcal{F}^{-1}\left\{\chi^{(2)}(\omega)\right\} = \frac{1}{2\pi}\int_{-\infty}^{\infty} \chi^{(2)}(\omega) e^{-i\omega t} \, d\omega$

Time-domain response to general input

- Convolve impulse response with input

$$f_{out}(t)=\int_0^\infty h(\tau)f_{in}(t-\tau)d\tau$$

  - Start integration at τ=0: h(τ)=0 for τ<0 (causality)
  - Output can only depend on previous history
- For EM:

$$P^{(1)}(t)=\varepsilon_0\int_0^\infty R^{(1)}(\tau)E(t-\tau)d\tau$$

- By convolution thm: we can evaluate in ω-domain

$$P^{(1)}(\omega)=\varepsilon_0\chi^{(1)}(\omega)E(\omega)$$

Time dependent NL response

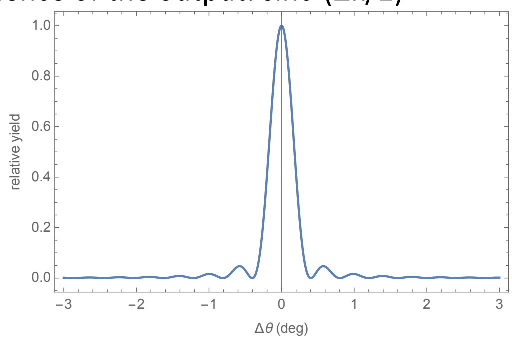
- By extension, we can define the 2nd order NL response in the time domain:

$$P^{(2)}(t)=\varepsilon_0\int_0^\infty d\tau_1\int_0^\infty d\tau_2 R^{(2)}(\tau_1,\tau_2)E_1(t-\tau_1)E_2(t-\tau_2)$$

  - Causality: $R^{(2)}(\tau_1,\tau_2)=0 \text{ for } \tau_1<0 \text{ or } \tau_2<0$
- Again, finite time response results from frequency response of $\chi^{(2)}$.
  - If we ignore frequency dependence, we also assume instantaneous response.
  - OK for non-resonant electronic response
  - Not OK: Raman response, slow nonlinearities (molecular, thermal, …)

Frequency response of phase matched SHG

- Even if we assume the NL response is instantaneous at the microscopic level, phase mismatch leads to a frequency dependence of the output: $\mathrm{sinc}^2(\Delta k/2)$



- For pulsed input, this limits the duration of the output
- In time domain, this leads to a rectangular output pulse

Short pulse harmonic conversion

- Although the phase matching is best described in the frequency domain, the nonlinear interaction takes place in the time domain.
  – For perfect phase matching across the bandwidth, non-depleted conversion for the q^{th} harmonic is:

$$\frac{dE_q(z,t)}{dz} \propto \chi^{(q)} E_1{}^q(t)$$

  – This leads to an output pulse shorter than the input

$$E_1(t) = E_0 e^{-t^2/\tau_1{}^2} \qquad E_q(t) \propto e^{-qt^2/\tau^2} \qquad \tau_q = \tau_1/\sqrt{q}$$

Ideal output bandwidth

- For a transform-limited pulse

  $$\tau\,\Delta\omega = 2$$

  - τ, $\Delta\omega$ are $1/e^2$ half-widths
  - Independent of the central frequency or wavelength
- In terms of wavelength:

  $$\frac{\Delta\omega}{\omega_0} = \frac{\Delta\lambda}{\lambda_0} \qquad \Delta\omega = \frac{2}{\tau} = \frac{\Delta\lambda}{\lambda_0}\omega_0 \qquad \boxed{\Delta\lambda = \frac{\lambda_0^{\ 2}}{\pi c \tau}}$$

- In the lab, we measure wavelength

  $$\tau_q = \tau_1/\sqrt{q} \qquad \boxed{\Delta\lambda_q = \frac{\lambda_q^{\ 2}}{\pi c \tau_q} = \frac{\lambda_1^{\ 2}}{q^2}\frac{\sqrt{q}}{\pi c \tau_1} = \frac{1}{q^{3/2}}\Delta\lambda_1}$$

Generalized spectral shape for SHG

- Since the output (non-depleted, phase-matched) is

  $$E_2(t) \propto \chi^{(2)} E_1^{\ 2}(t)$$

- The spectrum is obtained with the Fourier transform

  $$E_2(\omega) \propto \chi^{(2)} \mathcal{F}\left\{E_1(t)E_1(t)\right\}$$

- Use convolution theorem:

  $$E_2(\omega) \propto \chi^{(2)} E_1(\omega) \otimes E_1(\omega) = \chi^{(2)} \int E_1(\omega')E_1(\omega-\omega')d\omega'$$

  - The ideal output spectrum is the autoconvolution of the input spectrum