Alex Filin

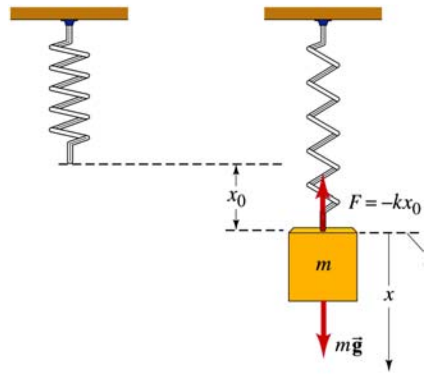
Phase Matching

Everything you always wanted to know about it
but were afraid to ask

Outline

- Introduction: Origin of Optical Nonlinearity
- Phase Matching in SHG
- Phase Matching in CARS
- Conclusion

Origin of optical nonlinearity: mechanical analog



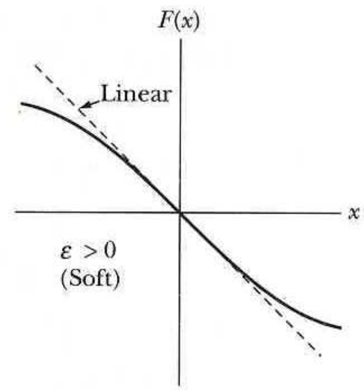
<u>Linear conditions</u>

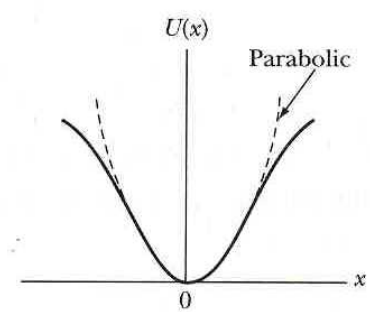
Force:

$$F(x) = -kx$$

Potential:

$$U(x) = \frac{1}{2}kx^2$$





<u>Nonlinear conditions</u>

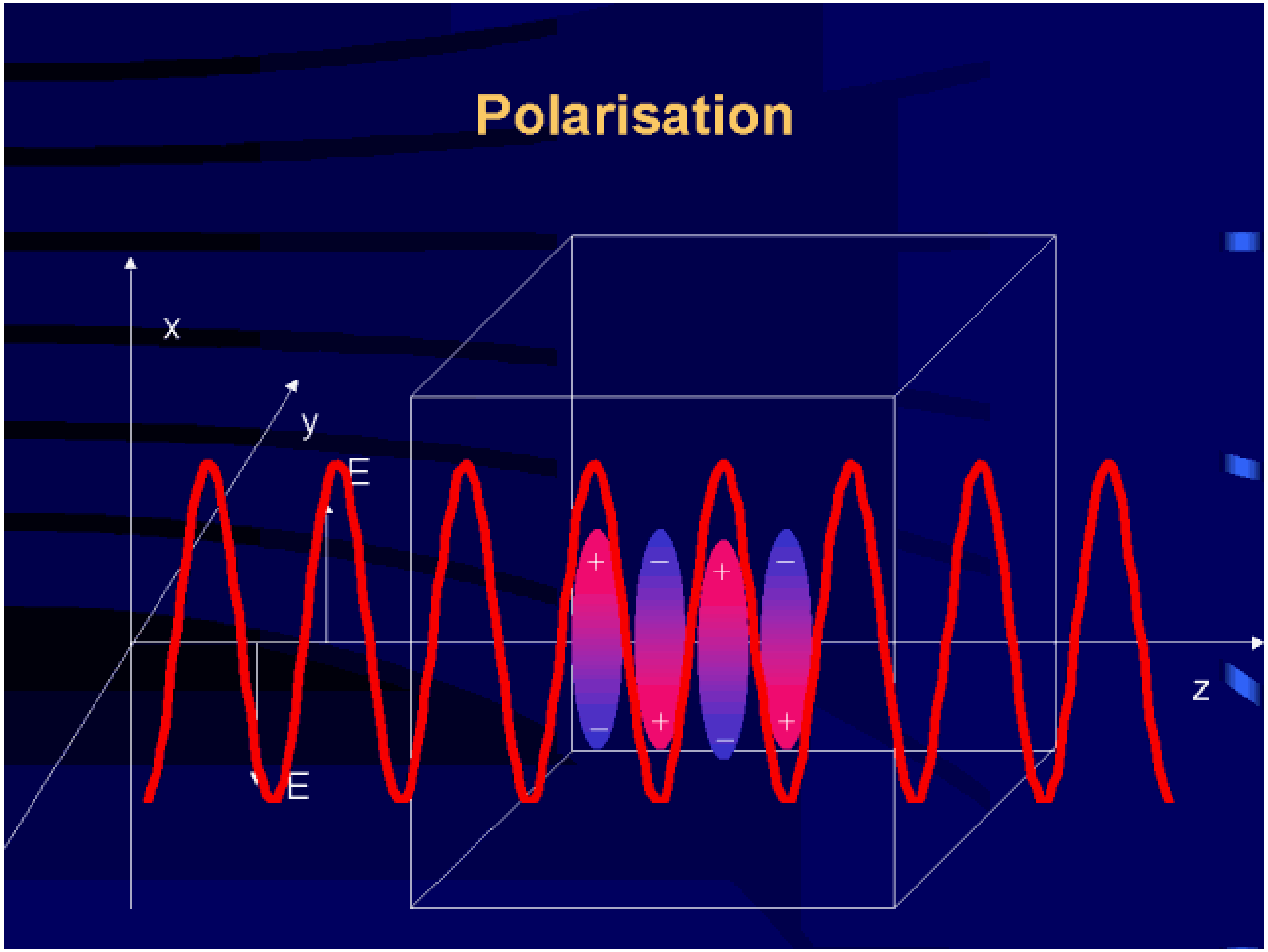
Force:

$$F(x) \cong -kx + \varepsilon x^3$$

Potential:

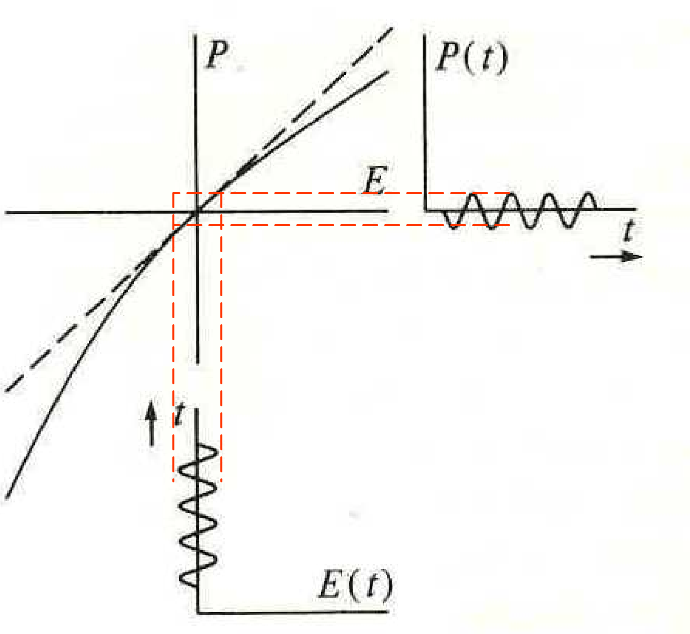
$$U(x) = \frac{1}{2}kx^2 - \frac{1}{4}\varepsilon x^4$$

Origin of optical nonlinearity: Polarization

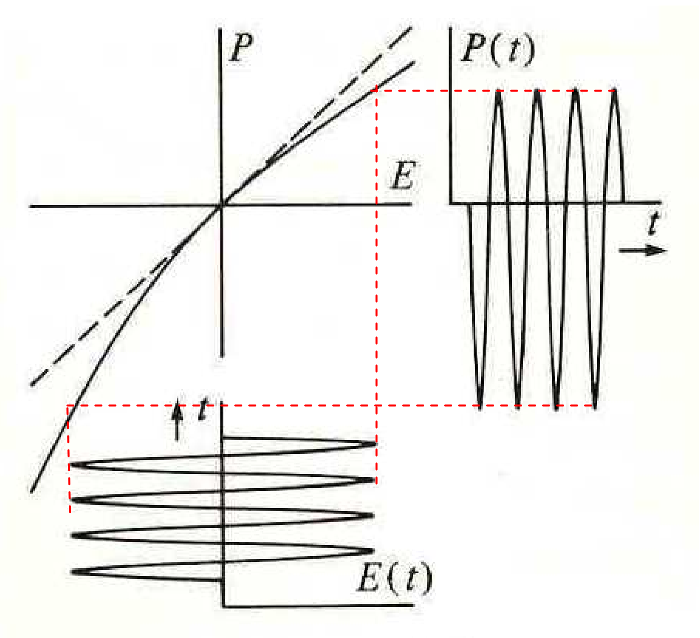


Origin of optical nonlinearity

Linear conditions



Nonlinear conditions



$$P = \varepsilon_0 \chi E$$

Where P is polarization
ε_o is free-space permittivity
χ is susceptibility
E is electric field

$$P = \varepsilon_0 (\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \ldots)$$

Where
$\chi^{(i)}$ is nonlinear susceptibility
of i^{th} order

Origin of optical nonlinearity

$$P = \varepsilon_0(\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \ldots)$$

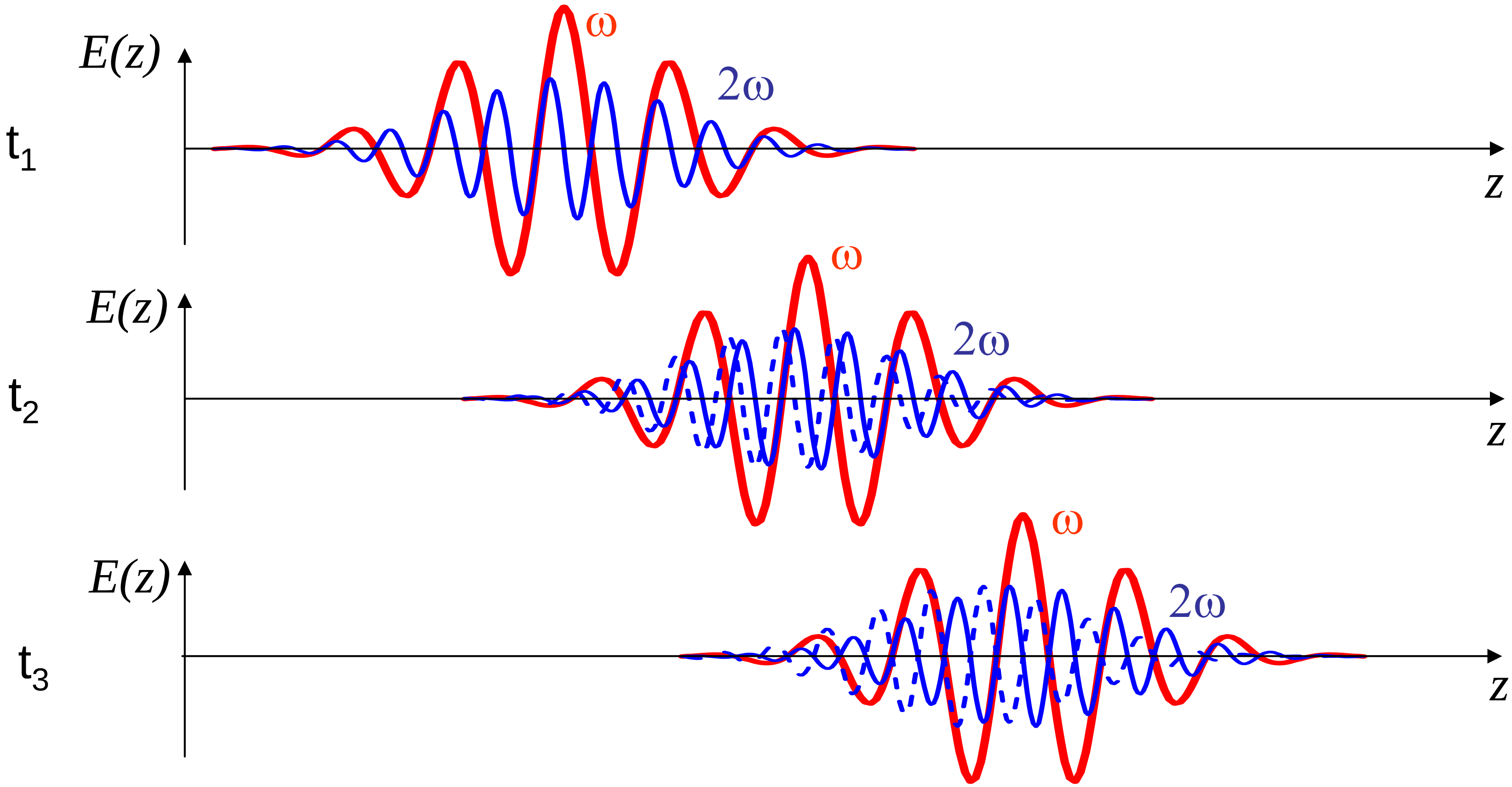
- All mixing phenomena, involving generation of sum and difference frequencies (SHG, parametric amplification)
- Pockels' effect
- Optical rectification

$\chi^{(2)}$ vanishes in media with inversion symmetry

- Third Harmonic Generation
- Kerr effect
- All types of FWM phenomena, including CARS

Second Harmonic Generation

Why does phase mismatching happen?

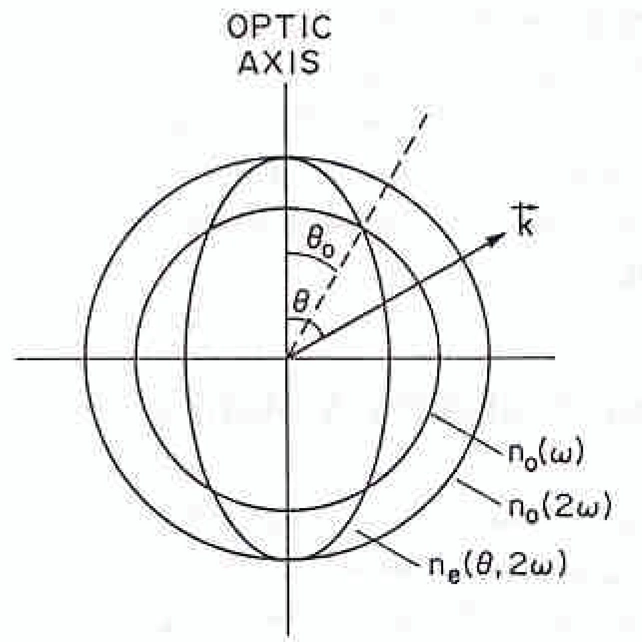


Second Harmonic Generation

In an uniaxial crystal

$$\frac{1}{n_e^2(\omega,\theta)} = \frac{\cos^2\theta}{n_o^2(\omega)} + \frac{\sin^2\theta}{n_e^2(\omega)}$$

where n_e and n_o are indexes of refraction for extraordinary and ordinary rays, respectively, θ is angle between $\boldsymbol{k}$ and optic axis of the crystal



Phase matching conditions: $\theta = \theta_0$ and $n_e(\omega,\theta) = n_o(\omega)$

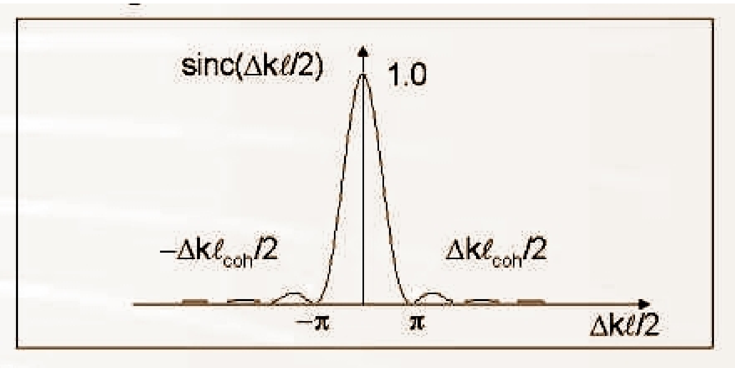
Or $n_{2\omega} = n_{\omega}$, but $n_{\omega} = \lambda k_{\omega}/2\pi$ and $n_{2\omega} = (\lambda/2)k_{2\omega}/2\pi$

So, $2k_{\omega} = k_{2\omega}$, or $\boxed{\Delta k = k(2\omega) - 2k(\omega) \Rightarrow 0}$

Second Harmonic Generation

One can show, that electric field

$$E(\omega_2, z) \propto e^{i\Delta kz/2} \frac{\sin(\Delta kz/2)}{\Delta k}$$



And Poynting vector

$$S(\omega_2, z) \propto \frac{\sin^2(\Delta kz/2)}{(\Delta k)^2}$$

Because

$$\lim_{\Delta k \to 0} \frac{\sin(\Delta kz/2)}{\Delta k} = \frac{z}{2} \quad =>$$

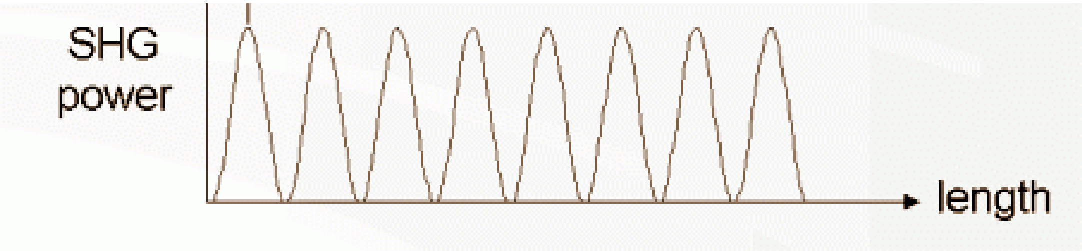
In ideal case ($\Delta k = 0$)

$$E(\omega_2, z) \propto z$$

$$S(\omega_2, z) \propto z^2$$

Second Harmonic Generation

In real case Δk never is equal to 0,
So, SHG power oscillates with z



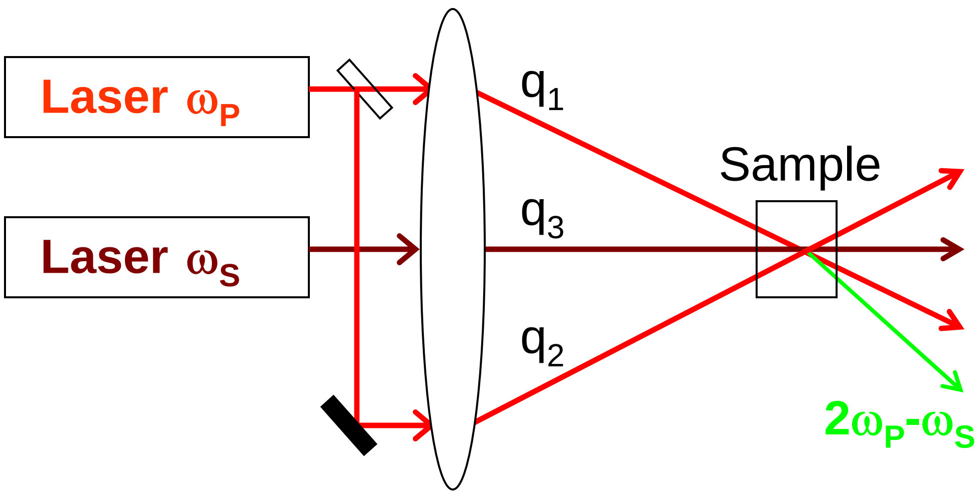
Finally, phase matching for SHG requires 2 conditions:

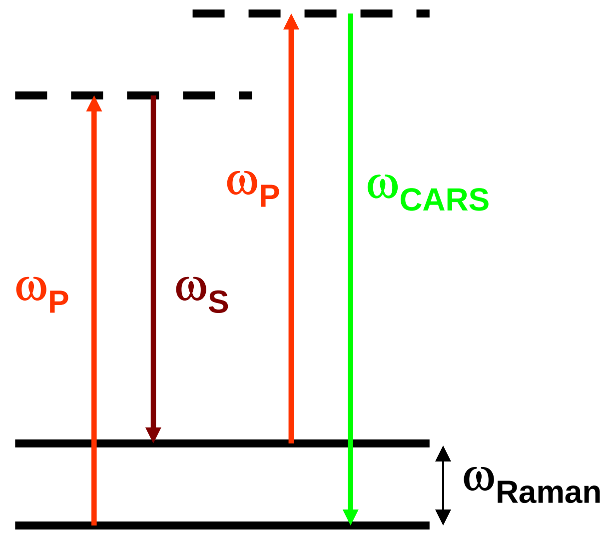
a) Correct angle between k and crystal axis to reach $n_{2\omega} = n_{\omega}$

or $\Delta k = k(2\omega) - 2k(\omega) \Rightarrow 0$

b) Correct crystal length to reach maximum SHG power

Coherent Anti-Stokes Raman Spectroscopy (CARS)





$2\omega_P = \omega_S + \omega_{CARS}$

- q_1 and q_2 correspond to ω_P
- q_3 corresponds to ω_S
- $\omega_P - \omega_S = \omega_{Raman}$ is the Raman shift (Raman active vibrational mode)

Coherent Anti-Stokes Raman Spectroscopy (CARS)

Intensity:

$$I_{CARS} = \frac{\omega_{CARS}^2}{n_P^2 n_S n_{CARS} \varepsilon_0^2 c^4} \left| \chi^{(3)} \right|^2 I_P^2 I_S L^2 \frac{\sin^2(\Delta k L/2)}{(\Delta k L/2)^2}$$

Where:

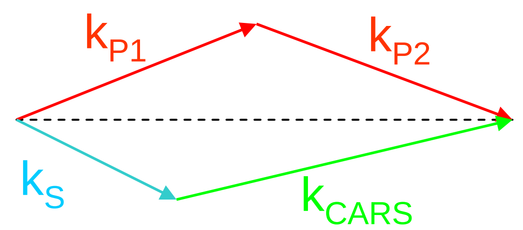
n_i is the refractive index at frequency ω_i

I_i is the intensity of *i*-th signal

L is the interaction length

$$\Delta k = \left| 2\vec{k}_P - \vec{k}_S - \vec{k}_{CARS} \right|$$

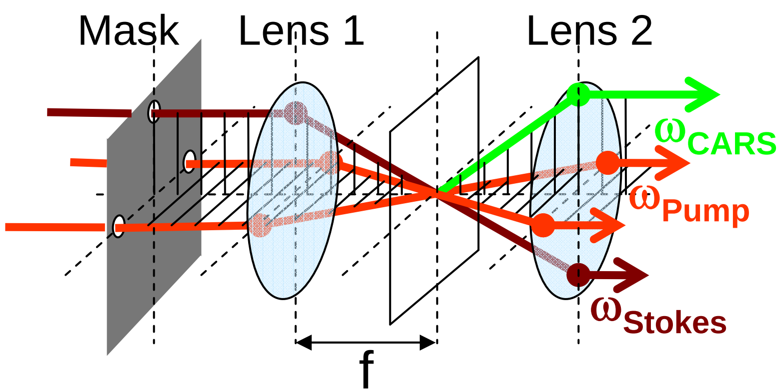
Phase matching for BOXCARS



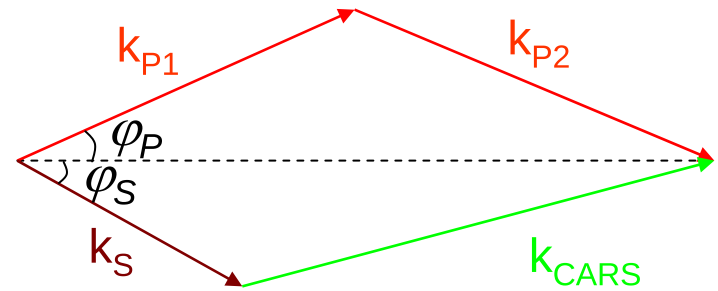
After Maker and Terhune (1989)

Principles of BOXCARS Method

Geometry of laser beams for BOXCARS

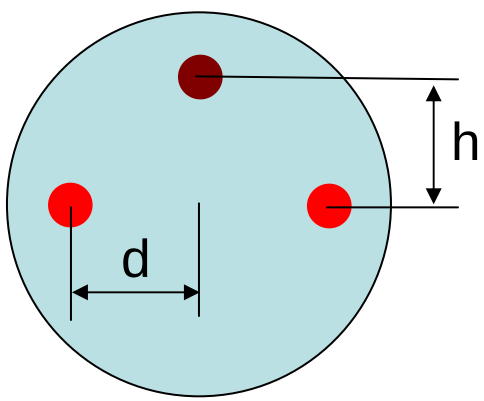


Phase matching for BOXCARS



$$2\,|k_{P1}| = |k_S| + |k_{CARS}|$$

$$2\omega_{Pump} = \omega_{Stokes} + \omega_{CARS}$$



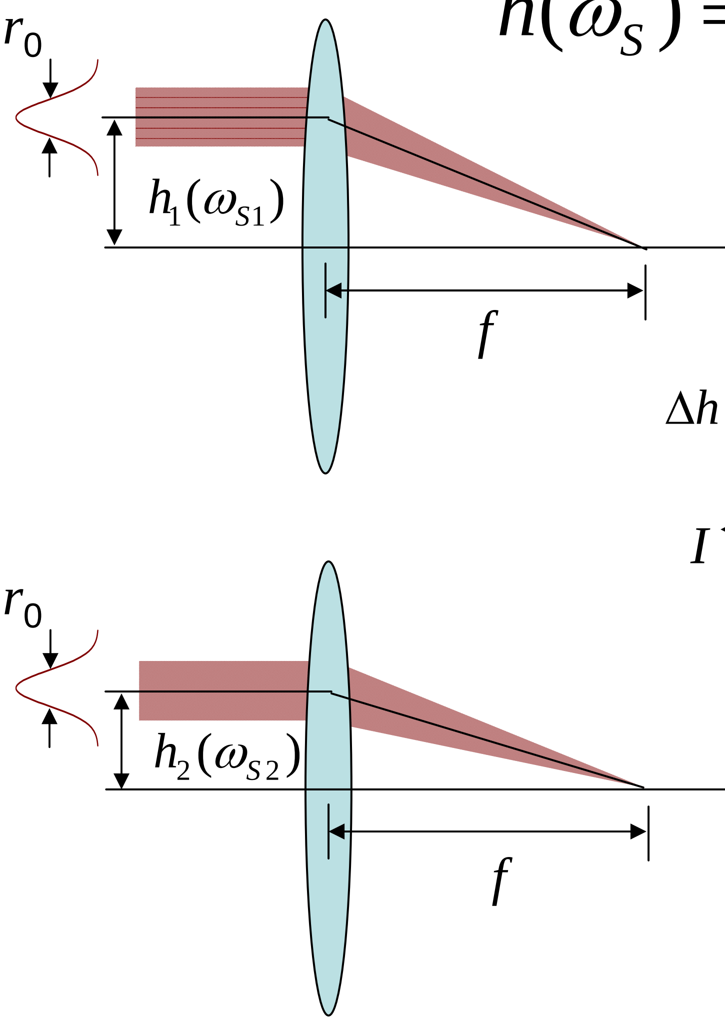
For h << f $\quad \sin\varphi_S = \sin\varphi_P \sqrt{2\frac{\omega_{Pump}}{\omega_{Stokes}} - 1}$

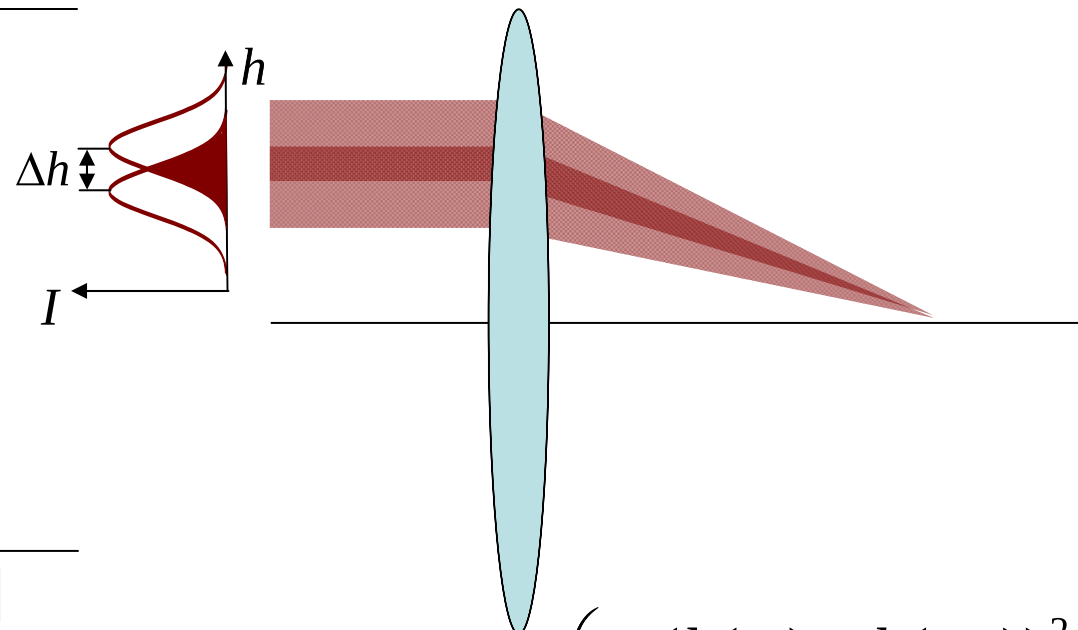
or

$$h(\omega_{Stokes}) = d \times \sqrt{2\frac{\omega_{Pump}}{\omega_{Stokes}} - 1}$$

Phase Matching in fs-BOXCARS

$$h(\omega_S) = d \times \sqrt{2\frac{\omega_P}{\omega_S} - 1}$$





$$I(\omega) = I(\omega_0)\exp\left(-\frac{(h(\omega) - h(\omega_0))^2}{2r_0^2}\right)$$

fs-CARS: Theory

$$I_{CARS}(\Delta_C,\Delta_S)=e^{-\frac{1+\alpha}{(2+\alpha)^2}\Delta_C^2}\left|B+Aie^{-\frac{1}{2}\left(\Delta_S-\frac{1+\alpha}{2+\alpha}\Delta_C\right)^2}\left(1+\mathrm{Erf}\left(\frac{i\left(\Delta_S-\frac{1+\alpha}{2+\alpha}\Delta_C\right)}{\sqrt{2}}\right)\right)\right|^2$$

Where: $\Delta_C=\dfrac{\omega+\omega_S-2\omega_P}{\sqrt{\sigma}};\quad \Delta_S=\dfrac{\omega_S+\omega_R-\omega_P}{\sqrt{\sigma}}$ normalized CARS frequency and normalized Stokes detuning

$$\sigma=\sigma_P\frac{1+\alpha}{2+\alpha};\quad \alpha=\frac{\sigma_S}{\sigma_P}=\left(\frac{FWHM_S}{FWHM_P}\right)^2$$

$$\sigma_P=\frac{(FWHM_P)^2}{4\ln 2}$$

Phase Matching in fs-BOXCARS

ps-CARS: $$I_{CARS} = A I_P^2 I_S L^2 \frac{\sin^2(\Delta k L/2)}{(\Delta k L/2)^2}$$

fs-BOXCARS: $$I_{CARS}^{REAL} = I_{CARS}(\Delta_C, \Delta_S) \times G\left(\frac{\Delta_C}{\omega_C}, \frac{\Delta_S}{\omega_S}, \frac{r_0}{h}, \frac{r_0}{d}, \frac{r_0}{f}\right)$$

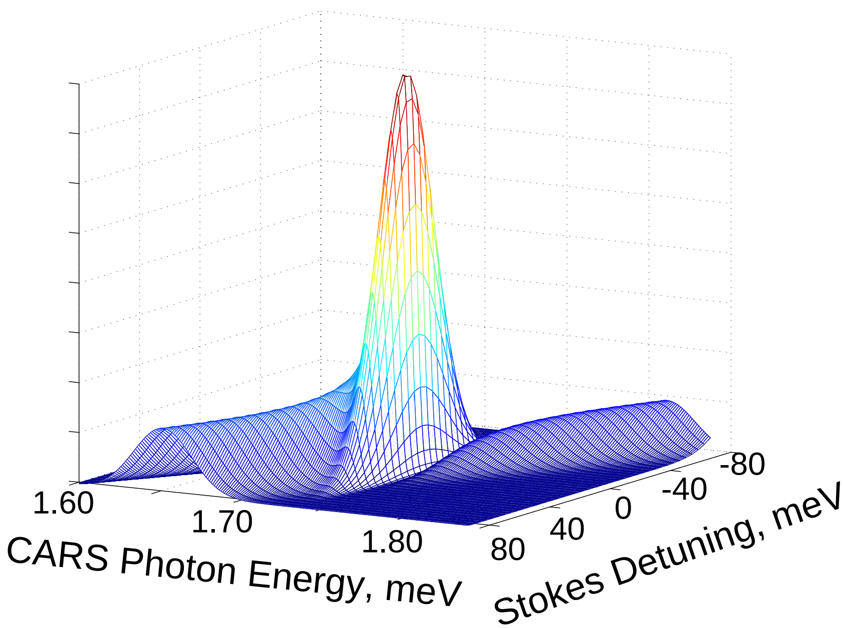
So far:

$$G\left(\frac{\Delta_C}{\omega_C}, \frac{\Delta_S}{\omega_S}, \frac{r_0}{h}, \frac{r_0}{d}\right) = \exp\left(-\frac{2}{3}\left(\left(\frac{\Delta_C}{\omega_C}\right)^2\left(\frac{d}{r_0}\right)^2 + \left(\frac{\Delta_S}{\omega_S}\right)^2\left(\frac{h}{r_0}\right)^2\right)\right)$$

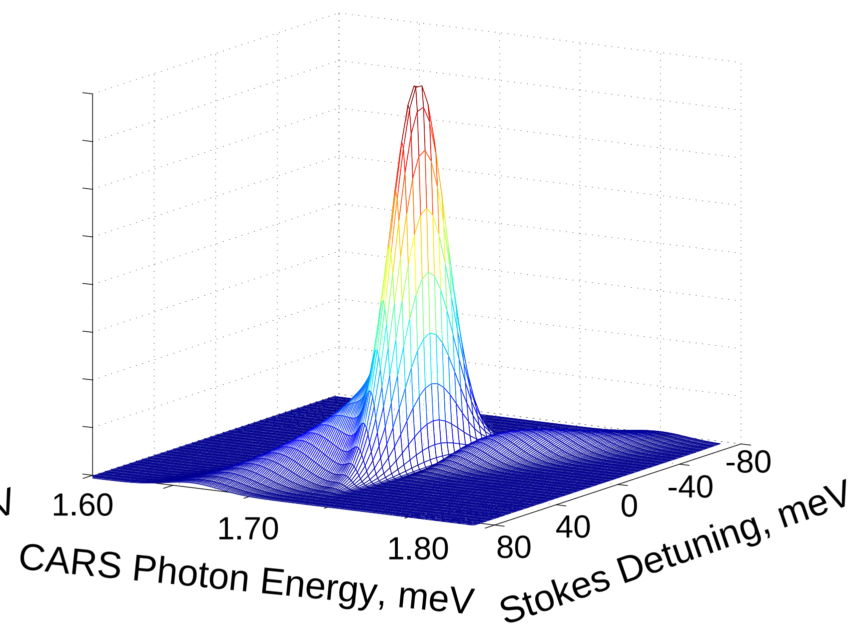
Phase Matching in fs-BOXCARS

Our results

Without G-correction

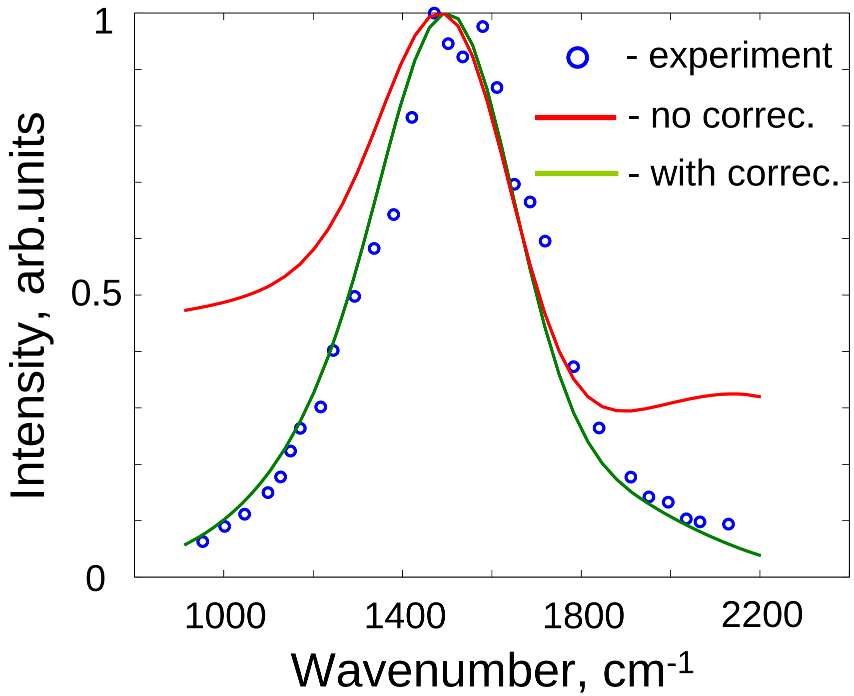


With G-correction



Phase Matching in fs-BOXCARS

Comparison theory and experiment



Conclusion

- Every nonlinear optical phenomenon requires it's own unique approach to understand the phase matching conditions
- Understanding of phase matching is crucially important to run a nonlinear optical experiment correctly and for interpretation of it's results.