A brief introduction to nonlinear optical materials, mode-locked oscillators, and ultrashort laser pulses

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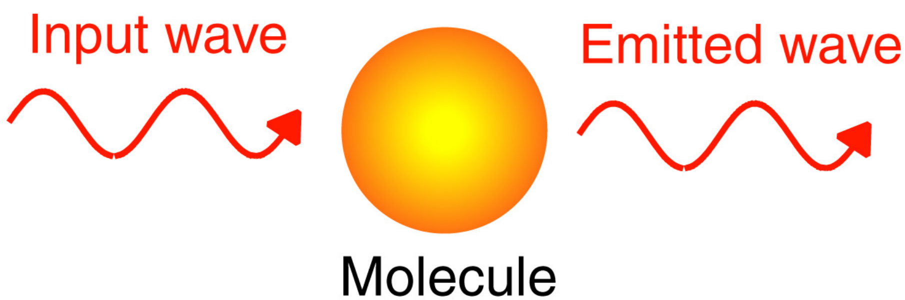
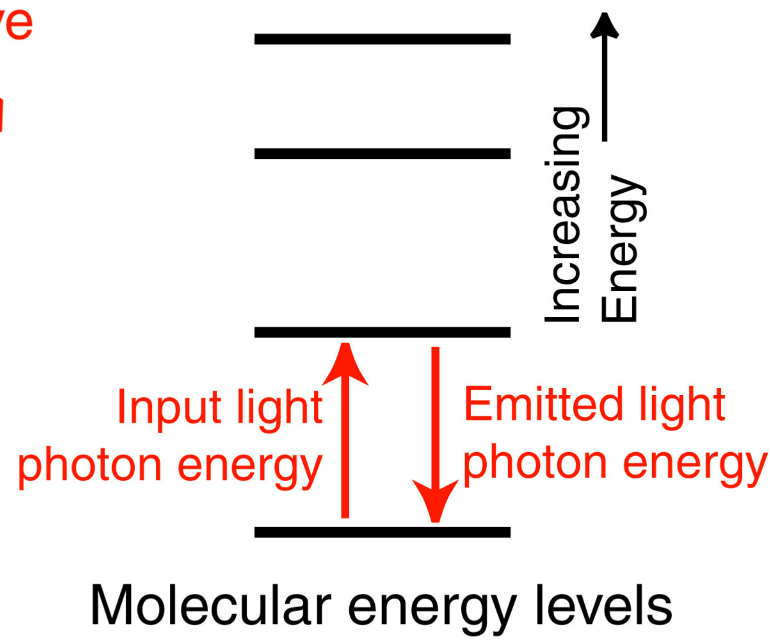
(based on lectures by Rick Trebino)

Why do nonlinear-optical effects occur?

- Recall that, in normal linear optics, a light wave acts on a molecule,
- which vibrates and then emits its own light wave that interferes
- with the original light wave.

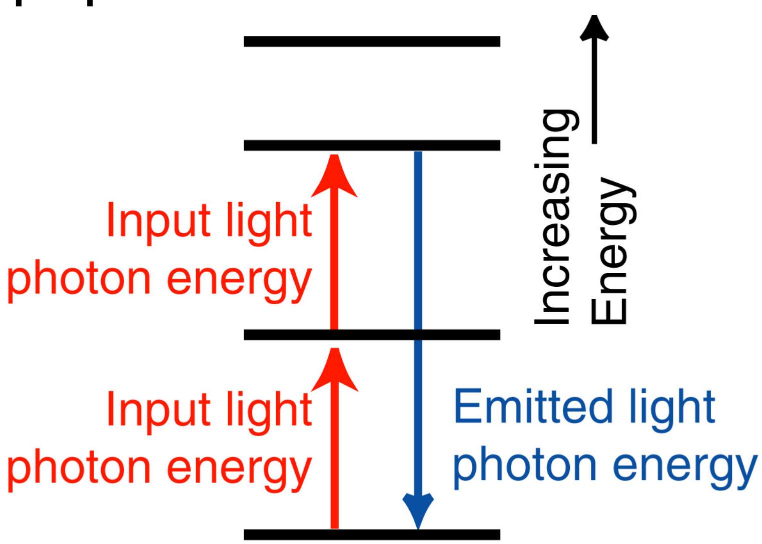


We can also imagine this process in terms of the molecular energy levels, using arrows for the photon energies:

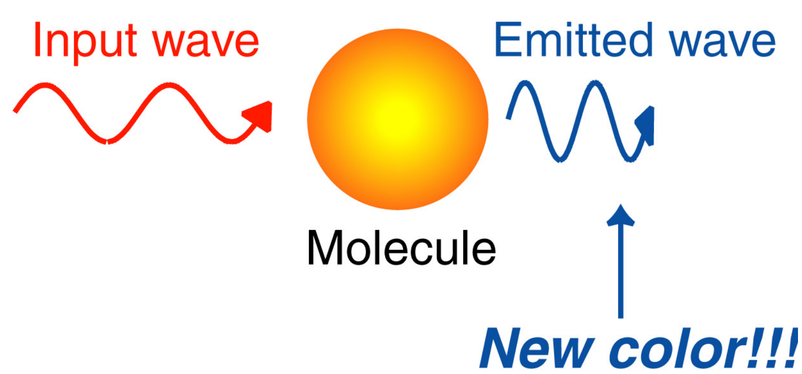


Why do nonlinear-optical effects occur? (continued)

•Now, suppose the irradiance is high enough that many molecules are excited to the higher-energy state. This state can then act as the lower level for additional excitation. This yields vibrations at all frequencies corresponding to all energy differences between populated states.

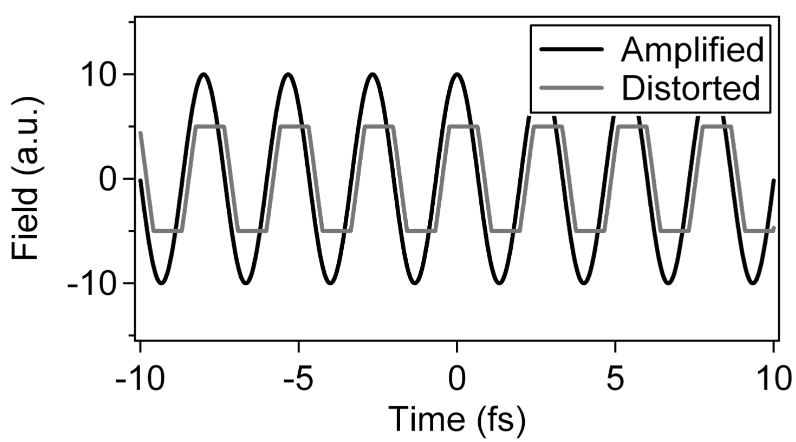


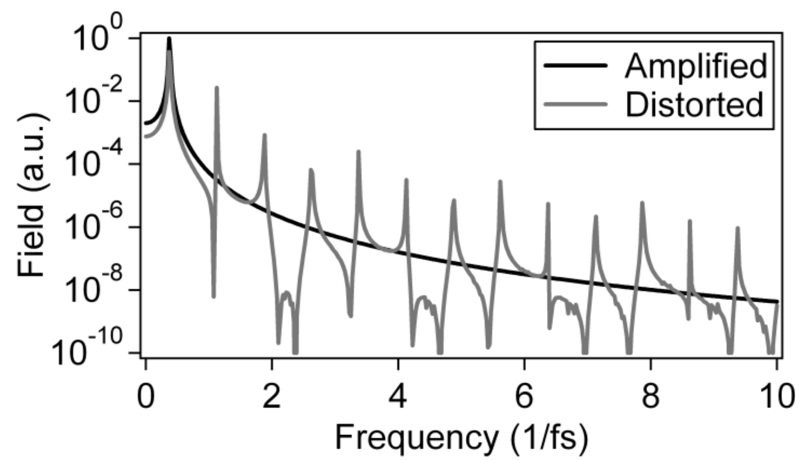
Molecular energy levels



Nonlinear optics is analogous to nonlinear electronics, which we can observe easily.

Sending a high-volume sine-wave ("pure frequency") signal into cheap speakers yields a truncated output signal, more of a square wave than a sine. This square wave has higher frequencies.



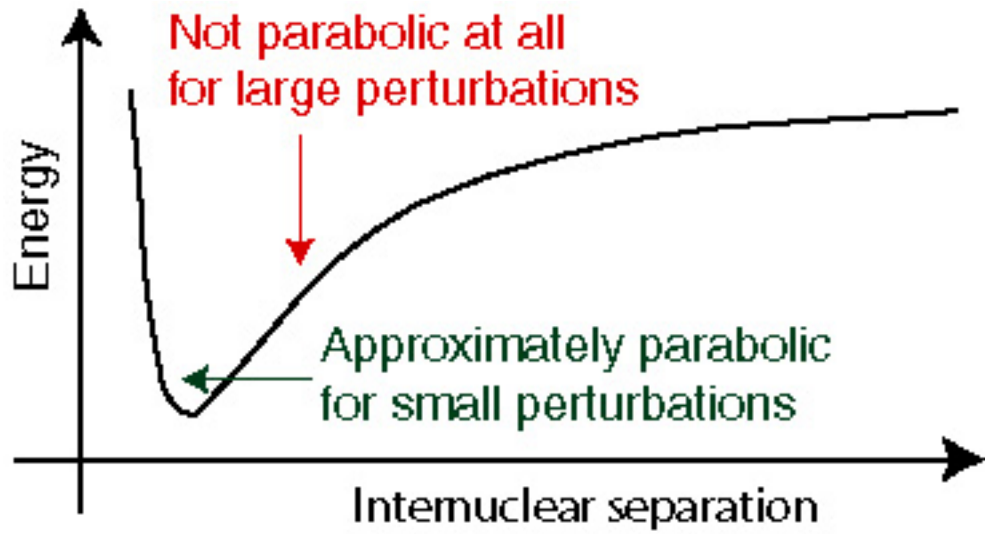


We hear this as distortion.

Nonlinear optics and anharmonic oscillators

Another way to look at nonlinear optics is that the potential of the electron or nucleus (in a molecule) is not a simple harmonic potential.

Example:



For weak fields, motion is harmonic, and linear optics prevails.
For strong fields (i.e., lasers), anharmonic motion occurs, and higher harmonics occur, both in the motion and the light emission.

Maxwell's Equations in a Medium

- The induced polarization, $\mathscr{P}$, contains the effect of the medium:

$$\vec{\nabla} \cdot \vec{\mathscr{E}} = 0 \qquad \vec{\nabla} \times \vec{\mathscr{E}} = -\frac{\partial \vec{\mathscr{B}}}{\partial t}$$

$$\vec{\nabla} \cdot \vec{\mathscr{B}} = 0 \qquad \vec{\nabla} \times \vec{\mathscr{B}} = \frac{1}{c_0^2} \frac{\partial \vec{\mathscr{E}}}{\partial t} + \mu_0 \frac{\partial \vec{\mathscr{P}}}{\partial t}$$

These equations reduce to the (scalar) wave equation:

$$\frac{\partial^2 \mathscr{E}}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 \mathscr{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \mathscr{P}}{\partial t^2}$$

"Inhomogeneous Wave Equation"

Sine waves of all frequencies are solutions to the wave equation; it's the polarization that tells which frequencies will occur.

The polarization is the driving term for the solution to this equation.

Maxwell's Equations in a *Nonlinear* Medium

Nonlinear optics is what happens when the polarization is the result of higher-order (nonlinear!) terms in the field:

$$\mathscr{P} = \varepsilon_0 \left[\chi^{(1)} \mathscr{E} + \chi^{(2)} \mathscr{E}^2 + \chi^{(3)} \mathscr{E}^3 + \ldots \right]$$

What are the effects of such nonlinear terms? Consider the second-order term:

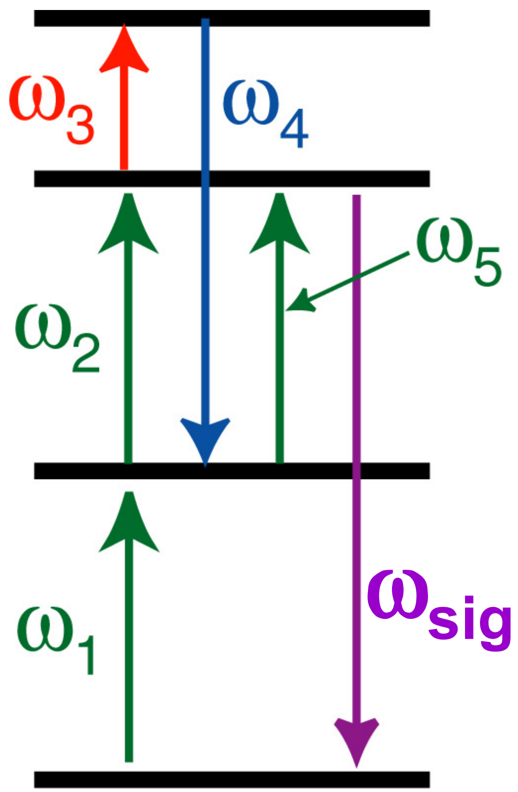
Since $\mathscr{E}(t) \propto E \exp(i\omega t) + E^* \exp(-i\omega t)$,

$$\mathscr{E}(t)^2 \propto E^2 \exp(2i\omega t) + 2|E|^2 + E^{*2} \exp(-2i\omega t)$$

2ω = 2nd harmonic!

Harmonic generation is one of many exotic effects that can arise!

Complicated nonlinear-optical effects can occur.



Emitted-light frequency

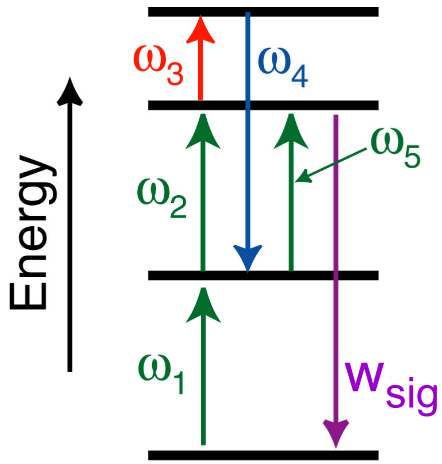
Nonlinear-optical processes are often referred to as:

"N-wave-mixing processes"

where N is the number of photons involved (including the emitted one). This is a six-wave-mixing process.

- The more photons (i.e., the higher the order) the weaker the effect, however. Very-high-order effects can be seen, but they require very high irradiance. Also, if the photon energies coincide with the medium's energy levels as above, the effect will be stronger.

Phase-matching = Conservation laws for photons in nonlinear optics

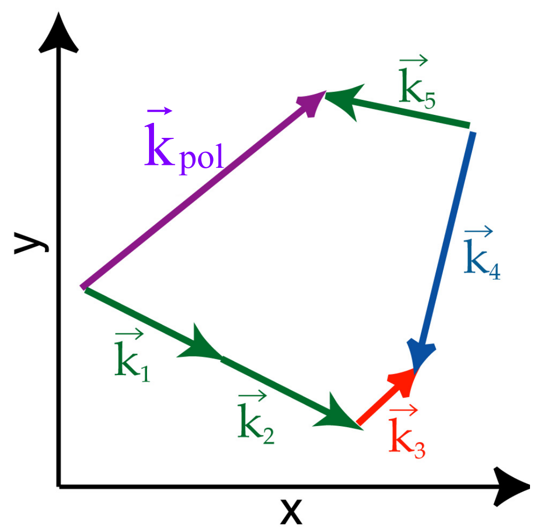


Adding the frequencies:

$$\omega_1 + \omega_2 + \omega_3 - \omega_4 + \omega_5 = \omega_{sig}$$

is the same as energy conservation if we multiply both sides by $\hbar$:

$$\hbar\omega_1 + \hbar\omega_2 + \hbar\omega_3 - \hbar\omega_4 + \hbar\omega_5 = \hbar\omega_{sig}$$



Adding the k's conserves momentum:

$$\vec{k}_1 + \vec{k}_2 + \vec{k}_3 - \vec{k}_4 + \vec{k}_5 = \vec{k}_{sig}$$

$$\hbar\vec{k}_1 + \hbar\vec{k}_2 + \hbar\vec{k}_3 - \hbar\vec{k}_4 + \hbar\vec{k}_5 = \hbar\vec{k}_{sig}$$

So phase-matching is equivalent to conservation of energy and momentum!

Group-velocity mismatch

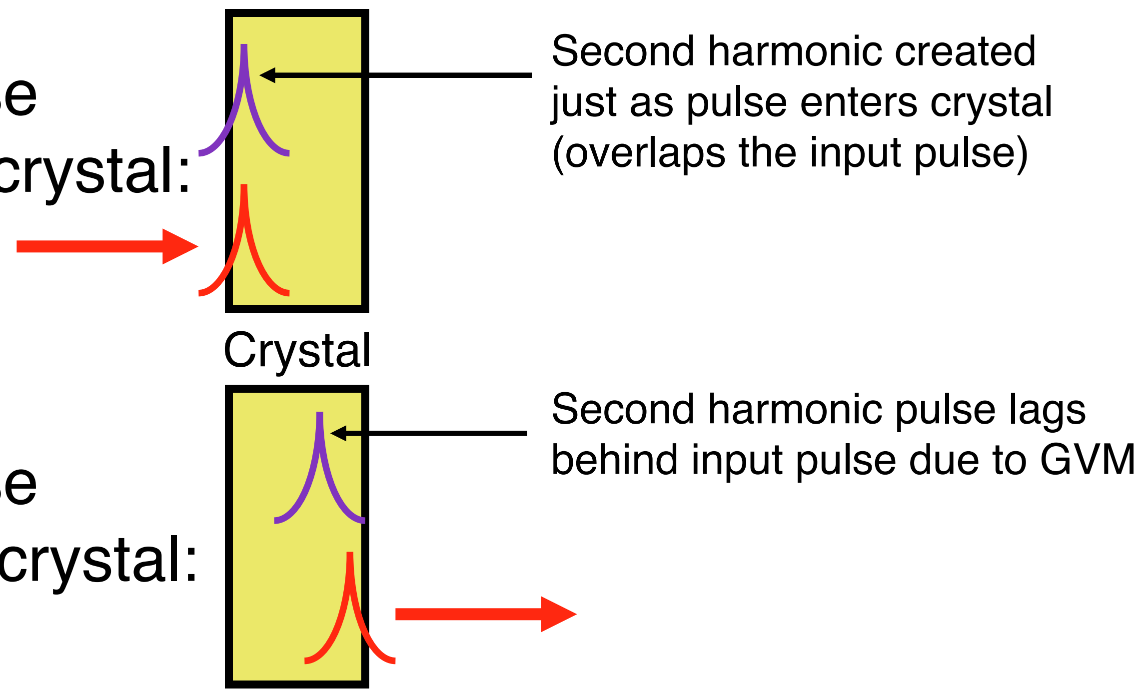
Inside the crystal the two different wavelengths have different group velocities.

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:

As the pulse leaves the crystal:



Phase-matching second-harmonic generation

So we're creating light at $w_{sig} = 2w$.

The k-vector of the second-harmonic is: $k_{sig} = \frac{\omega_{sig}}{c_0} n(\omega_{sig}) = \frac{(2\omega)}{c_0} n(2\omega)$

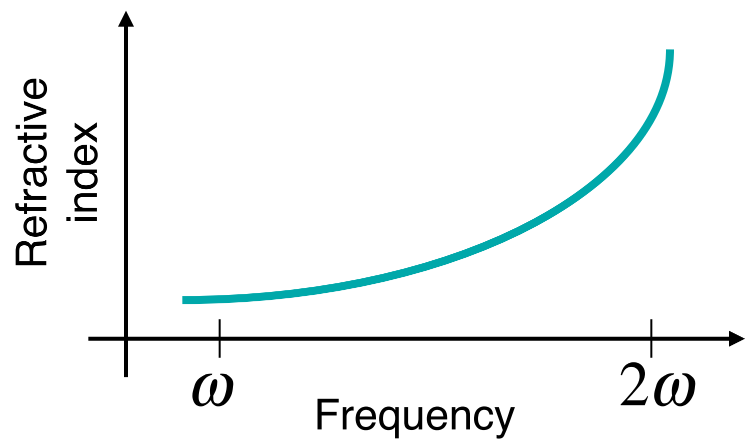
And the k-vector of the polarization is: $k_{pol} = 2\,k = 2\frac{\omega}{c_0} n(\omega)$

The phase-matching condition is: $k_{sig} = k_{pol}$

which will only be satisfied when:

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

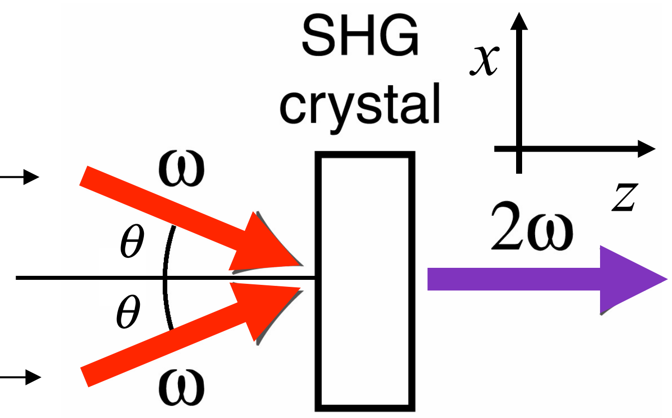
Unfortunately, dispersion prevents this from ever happening!



Noncollinear SHG phase-matching

$$\vec{k} = k\cos\theta\,\hat{z} - k\sin\theta\,\hat{x} \longrightarrow$$

$$\vec{k}' = k\cos\theta\,\hat{z} + k\sin\theta\,\hat{x} \longrightarrow$$



$$\vec{k}_{pol} = \vec{k} + \vec{k}' = 2\,k\cos\theta\,\hat{z}$$

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

But:

$$k_{sig} = \frac{2\omega}{c_o}\,n(2\omega)$$

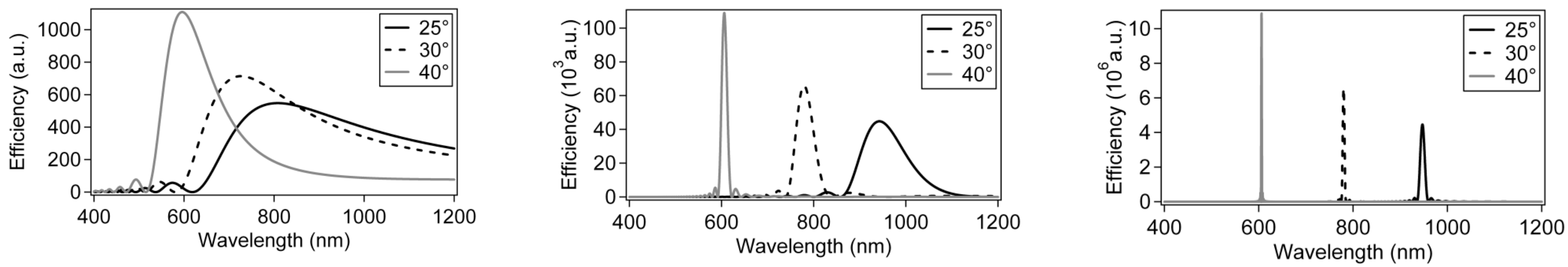
So the phase-matching condition becomes:

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

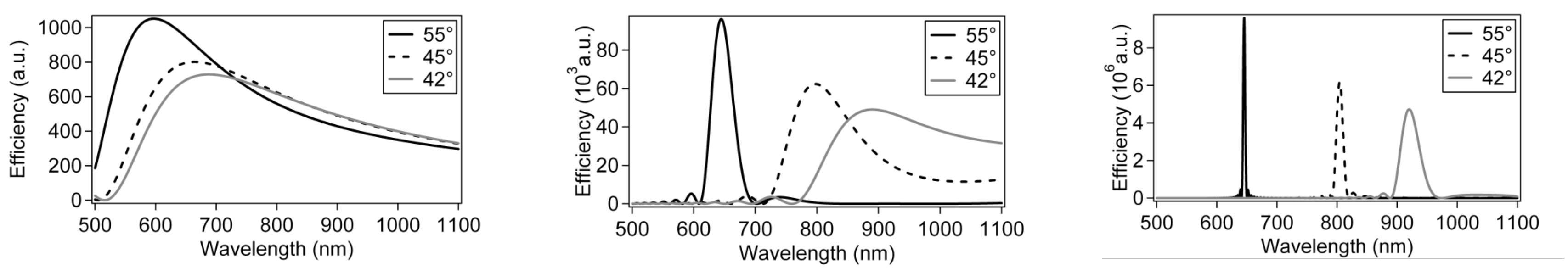
Phase-matching efficiency vs. wavelength for BBO and KDP

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

10 micron | 100 micron | 1000 micron



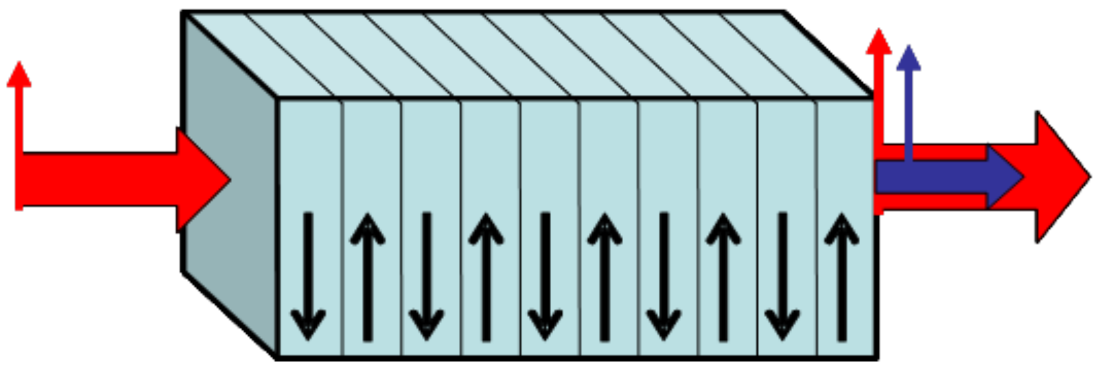
Note the huge differences in phase-matching bandwidth and efficiency with crystal thickness.



Alternative method for phase-matching: periodic poling

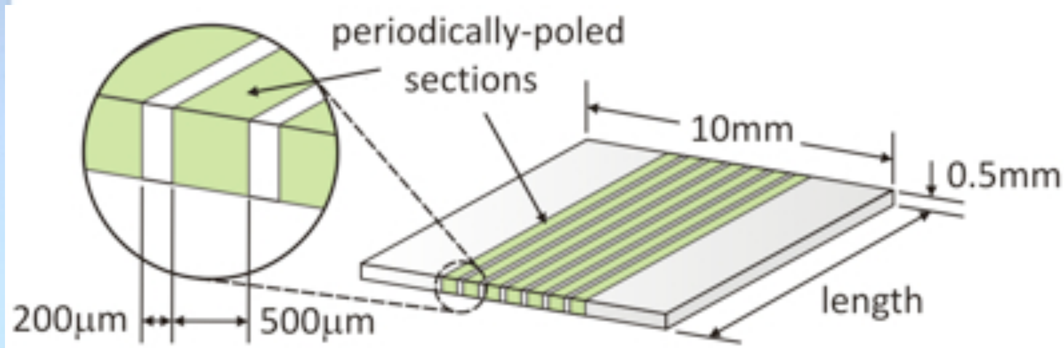
Recall that the second-harmonic phase alternates every coherence length when phase-matching is not achieved, which is always the case for the same polarizations—whose nonlinearity is much higher.

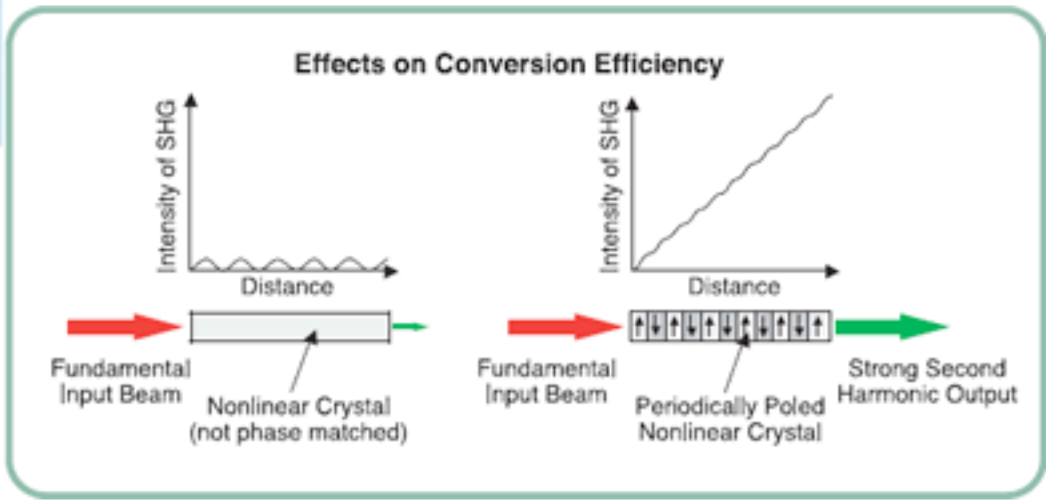
Periodic poling solves this problem. But such complex crystals are hard to grow and have only recently become available.



Example Products

- Covesion MgO:PPLN for Second Harmonic Generation

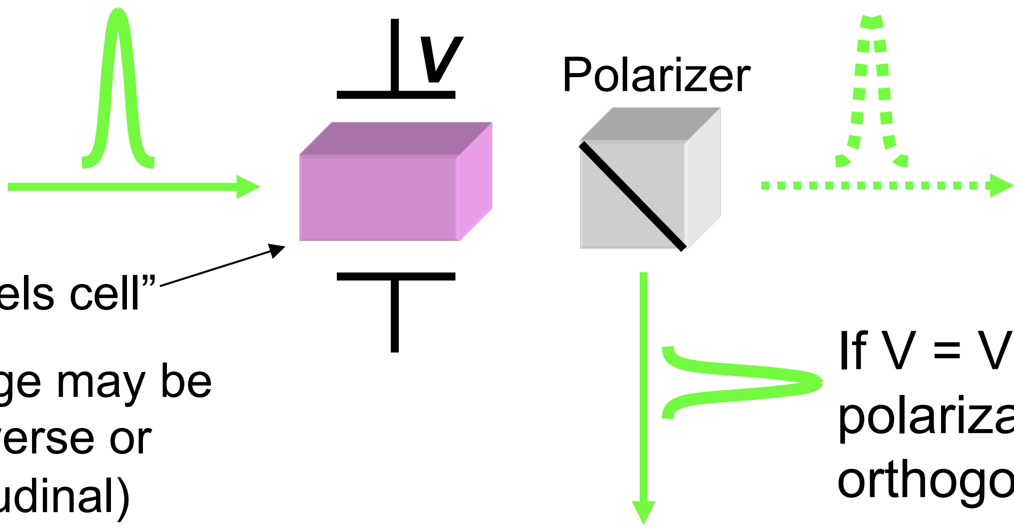




Another 2nd-order process: Electro-optics

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.



"Pockels cell"

(voltage may be transverse or longitudinal)

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

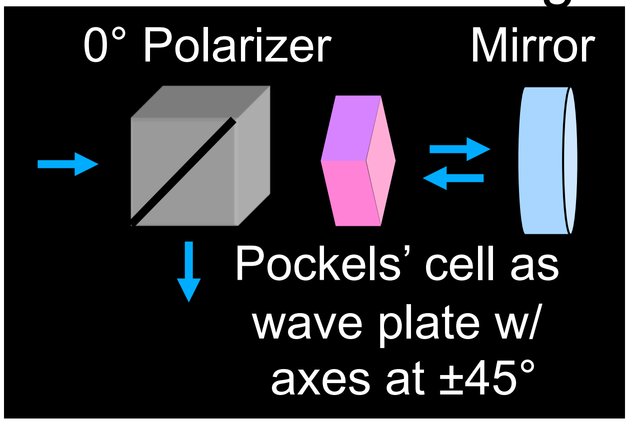
If $V = V_p$, the pulse polarization switches to its orthogonal state.

Abruptly switching a Pockels cell allows us to switch a pulse into or out of a laser.

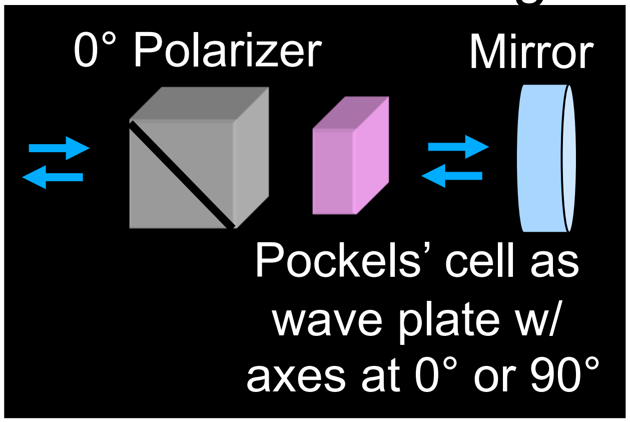
The Pockels' Cell (Q-Switch)

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

Before switching



After switching



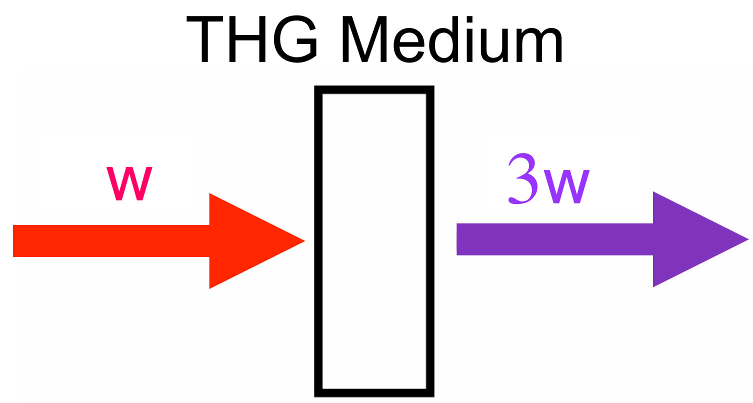
The Pockels effect involves the simple second-order process:

$$\omega_{sig} = \omega + 0$$

dc field

The signal field has the orthogonal polarization, however.

Third-harmonic generation



We must now cube the input field:

$$\mathscr{E}(\vec{r},t) = E\exp[i(\omega t - kz)] + E^{*}\exp[-i(\omega t - kz)]$$

$$\mathscr{E}(\vec{r},t)^3 = E^3\exp[i(3\omega t - 3kz)] + E^{*3}\exp[-i(3\omega t - 3kz)]$$

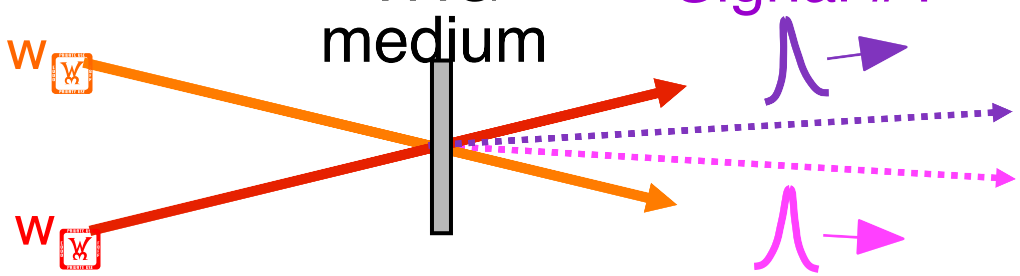
$$+ \text{other terms}$$

Third-harmonic generation is weaker than second-harmonic and sum-frequency generation, so the third harmonic is usually generated using SHG followed by SFG, rather than by direct THG.

Noncollinear third-harmonic generation

We can also allow two different input beams, whose frequencies can be different.

So in addition to generating the third harmonic of each input beam, the medium will generate interesting sum frequencies.



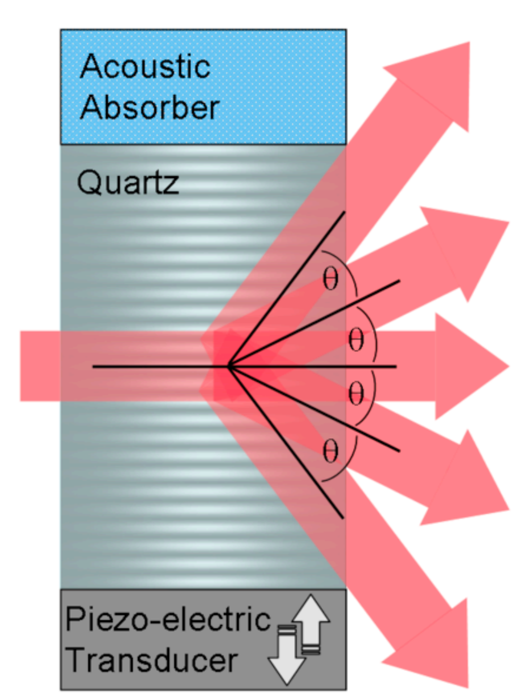
$$\mathscr{E}(\vec{r},t)^3 = E_1^2 E_2 \exp\{i[(2\omega_1+\omega_2)t-(2\vec{k}_1+\vec{k}_2)\cdot\vec{r})]\} +$$

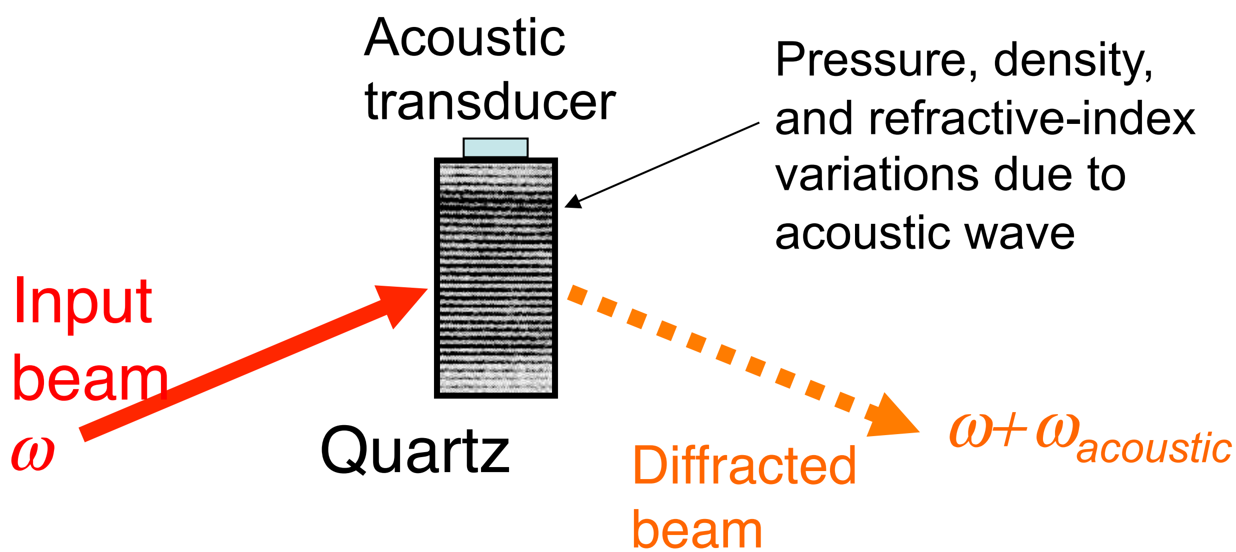
$$+ E_2^2 E_1 \exp\{i[(2\omega_2+\omega_1)t-(2\vec{k}_2+\vec{k}_1)\cdot\vec{r})]\} +$$

$$+ \text{other terms}$$

Acousto-optics involves diffracting light off a grating induced by an acoustic wave.

An acoustic wave induces sinusoidal density, and hence sinusoidal refractive-index, variations in a medium.



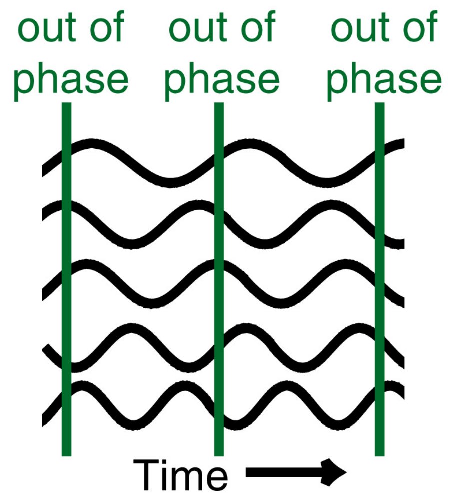


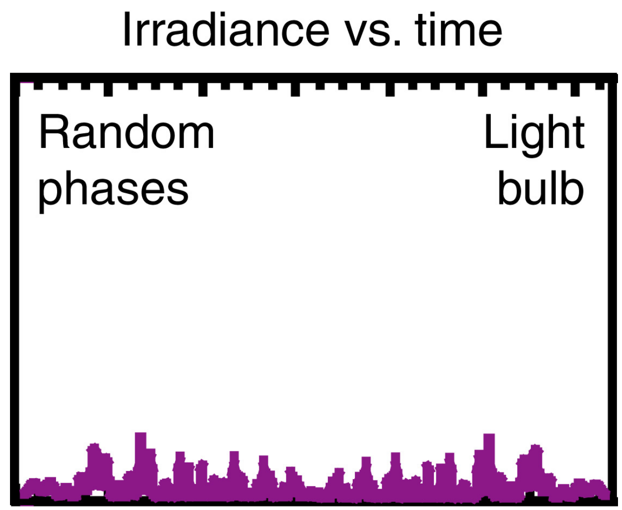
Acousto-optics works because acoustic waves have about the same wavelengths as (visible) light waves. Such diffraction can be quite strong: ~70%. Acousto-optics is the basis of useful devices.

Generating short pulses = "mode-locking"

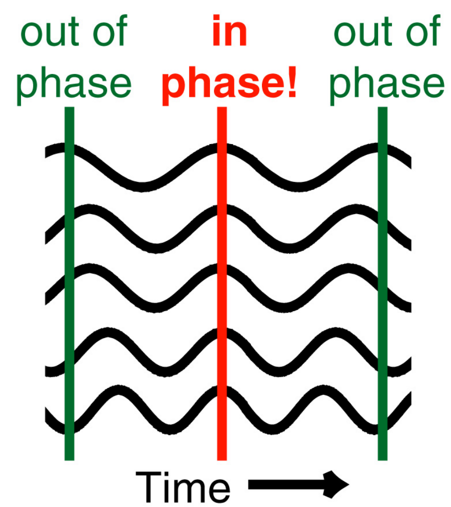
Locking the phases of the laser frequencies yields an ultrashort pulse.

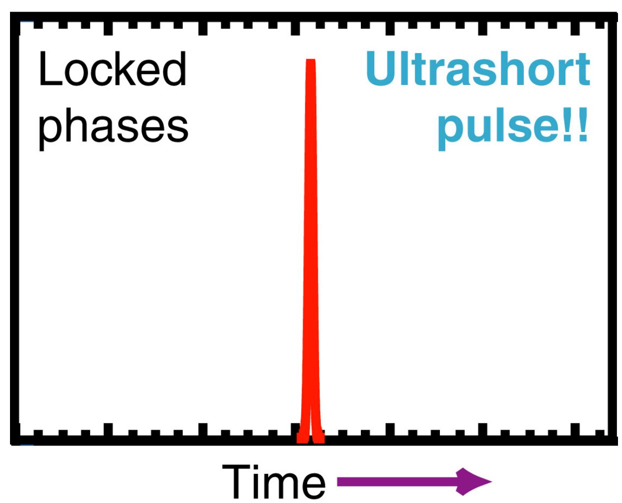
Random phases of all laser modes





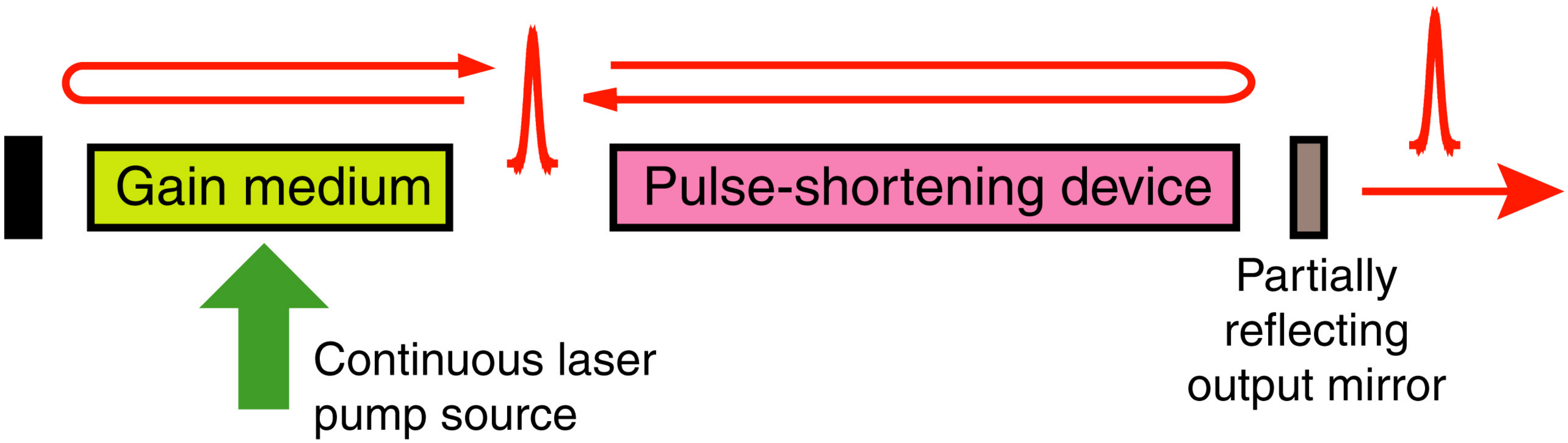
Locked phases of all laser modes





A generic ultrashort-pulse laser

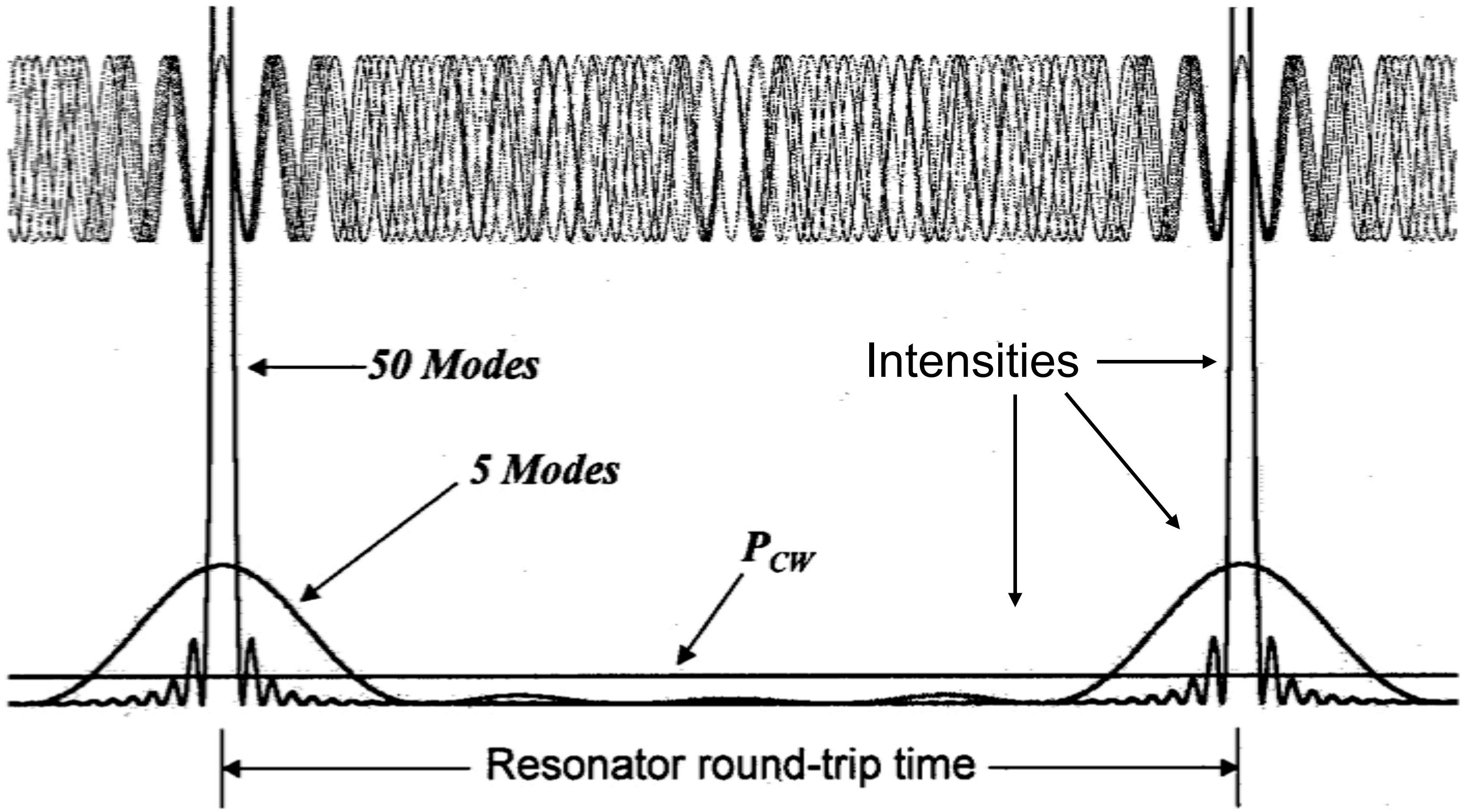
A generic ultrafast laser has a broadband gain medium,a pulse-shortening device, and two or more mirrors:



Pulse-shortening devices include:

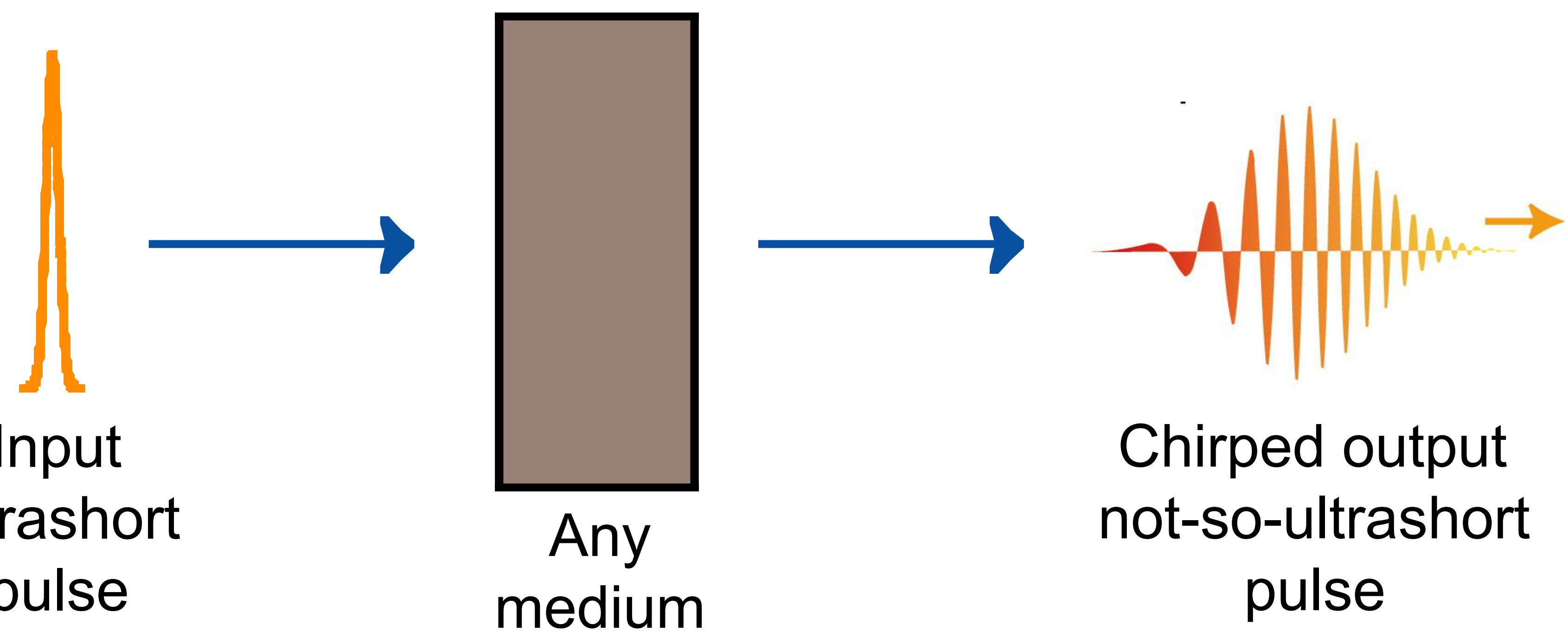
- Saturable absorbers
- Phase modulators
- Dispersion compensators
- Optical-Kerr media

Locking modes



Group velocity dispersion broadens ultrashort laser pulses

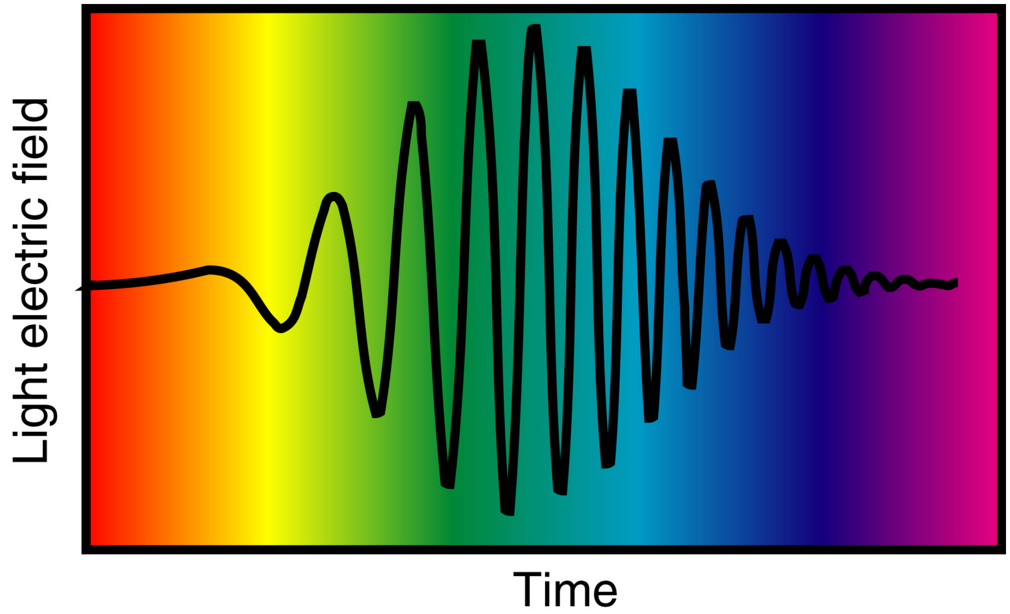
Different fquencies travel at different group velocities in materials, causing pulses to expand to highly "chirped" (frequency-swept) pulses.



Longer wavelengths almost always travel faster than shorter ones.

The Linearly Chirped Pulse

Group velocity dispersion produces a pulse whose frequency varies in time.

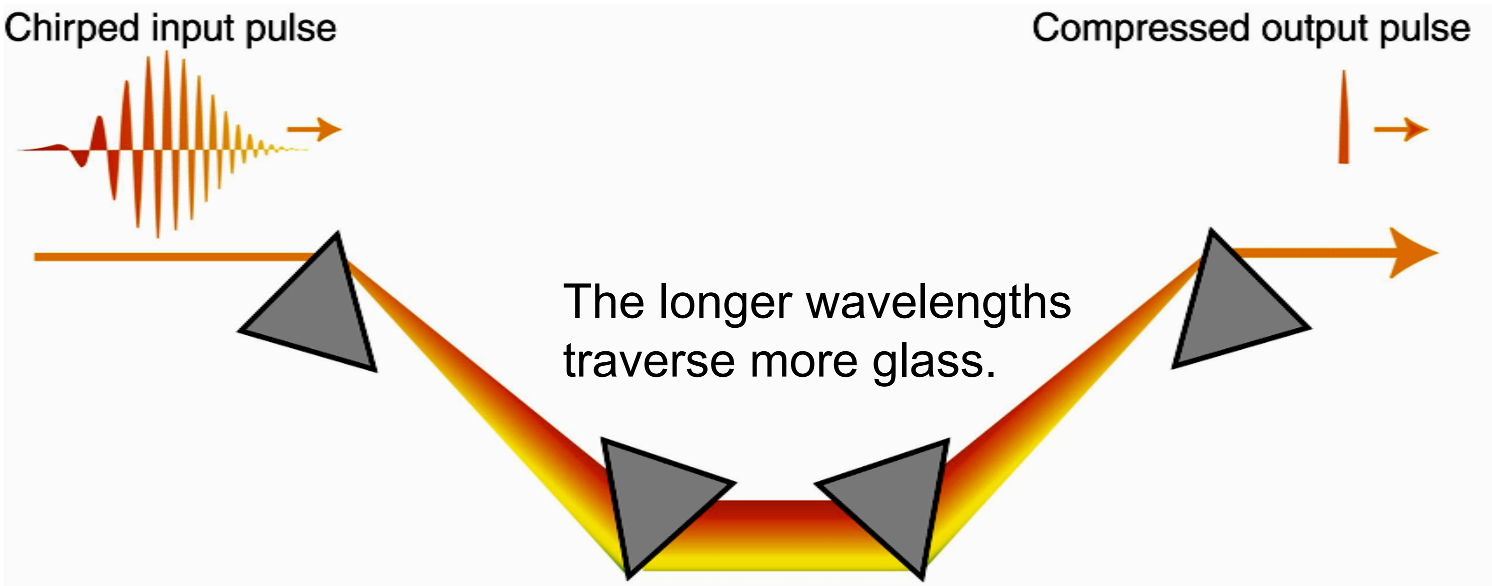


This pulse increases its frequency linearly in time (from red to blue).

In analogy to bird sounds, this pulse is called a "chirped" pulse.

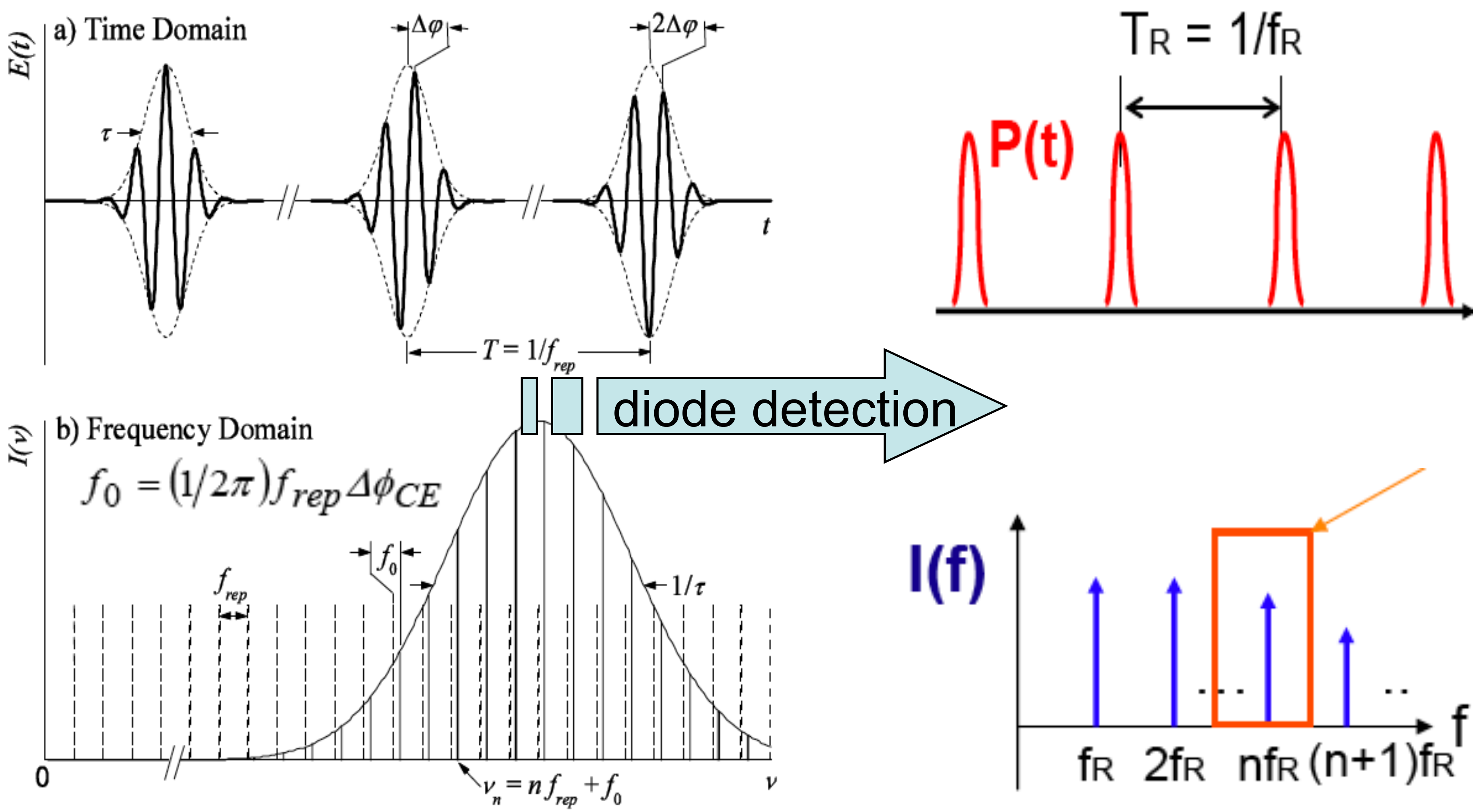
Pulse Compressor

This device has negative group-velocity dispersion and hence can compensate for propagation through materials (i.e., for positive chirp).

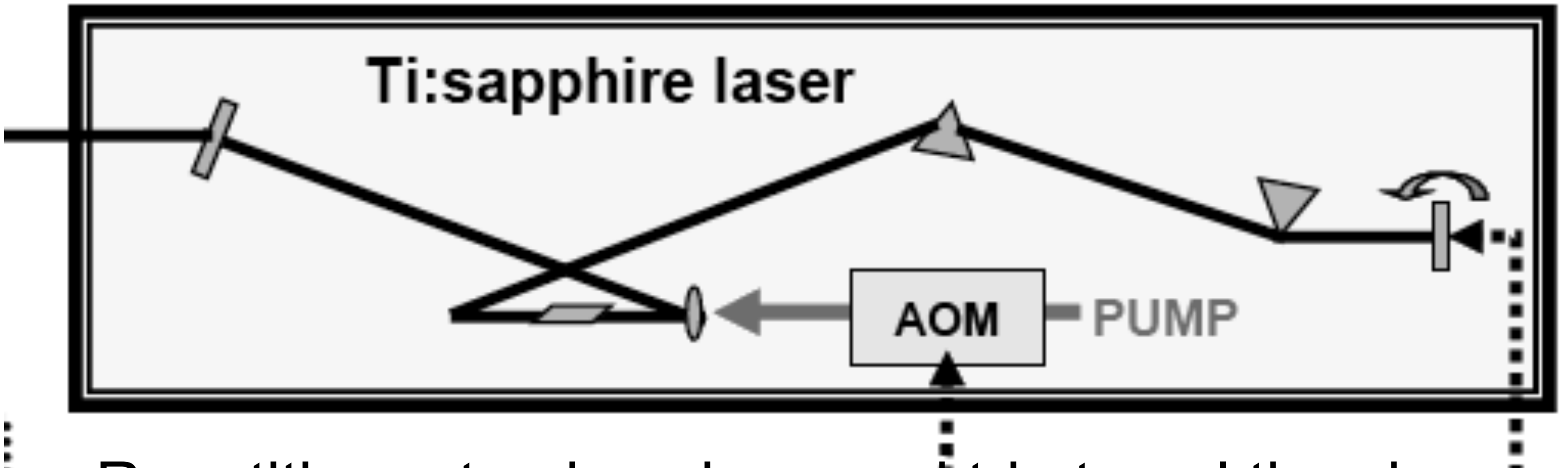


It's routine to stretch and then compress ultrashort pulses by factors of >1000

Femtosecond combs



Example:Ti:Sapph MLL



Repetition rate given by round trip travel time in cavity. Modulated by piezo adjustment of cavity mirror.

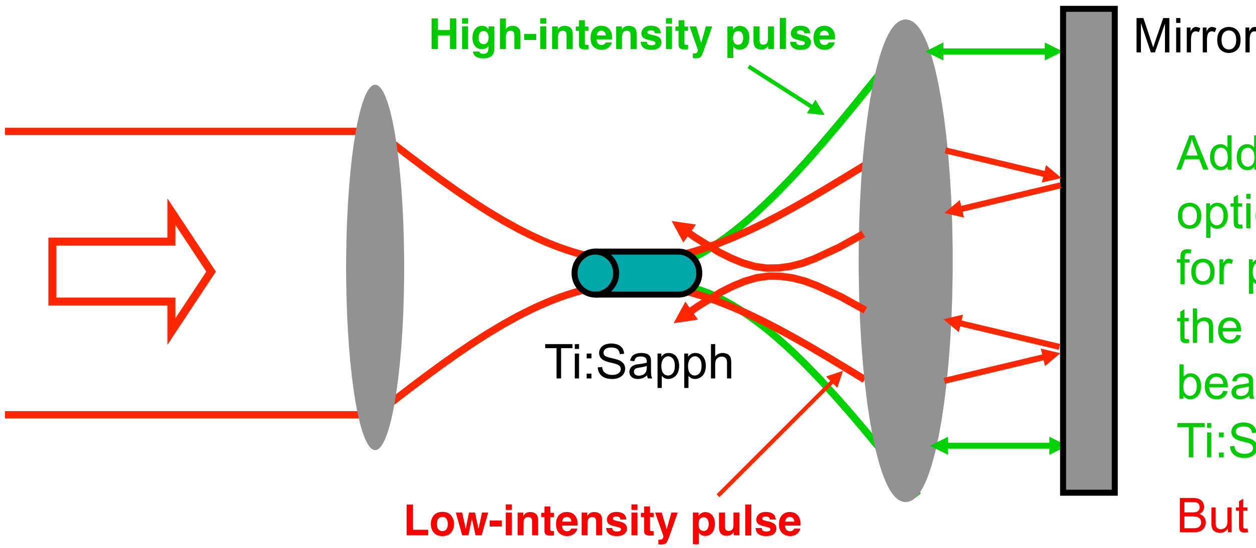
Passive mode locking achieved by properties of nonlinear crystal

Modern commercial designs include dispersion compensation in optics

Comb spectrum allows direct link of microwave frequencies to optical frequencies

Kerr-lensing is a type of saturable absorber.

If a pulse experiences additional focusing due to high intensity and the nonlinear refractive index, and we align the laser for this extra focusing, then a high-intensity beam will have better overlap with the gain medium.



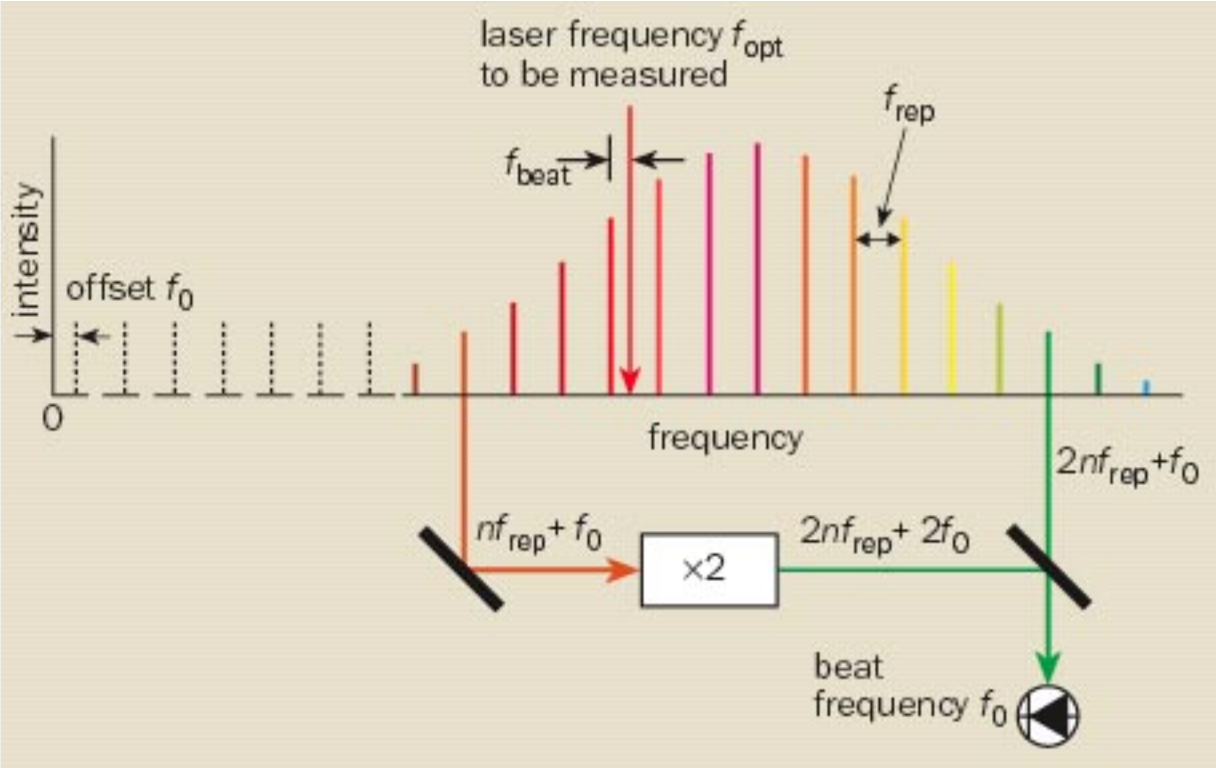
Additional focusing optics can arrange for perfect overlap of the high-intensity beam back in the Ti:Sapphire crystal.

But not the low-intensity beam!

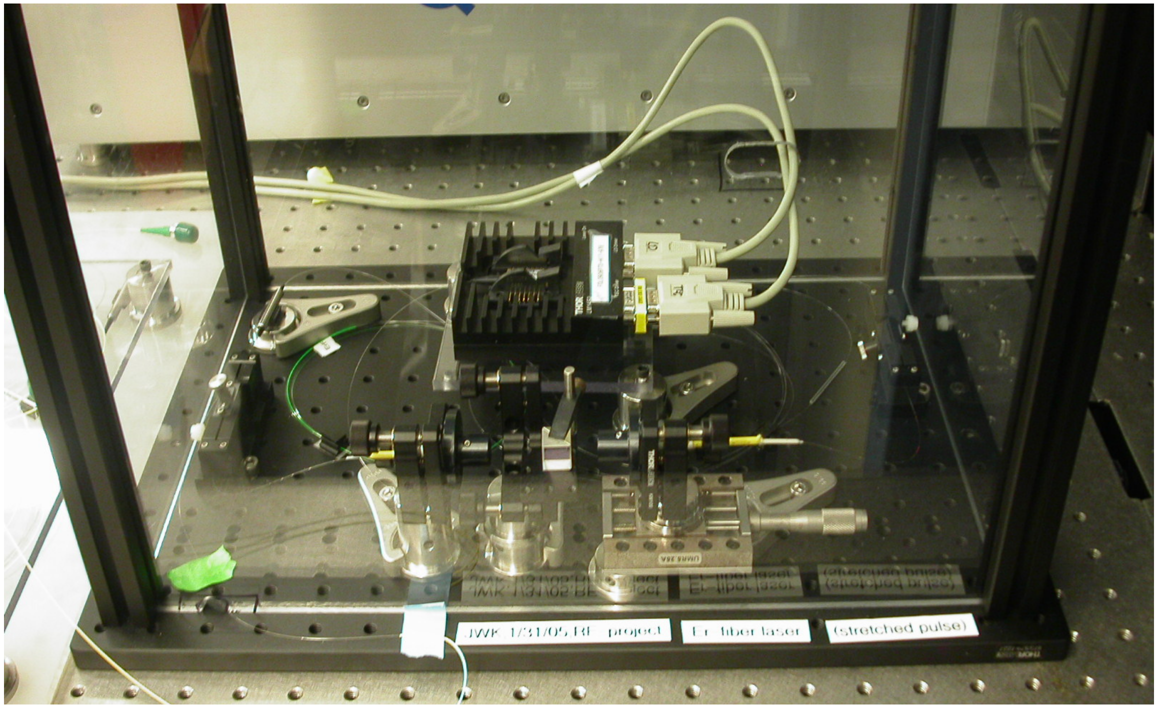
This is a type of saturable absorption.

Self-referencing stabilizer

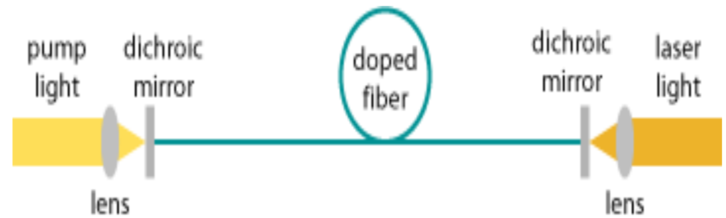
CEO frequency can be directly measured with an octave spanning spectrum and stabilized in a feedback loop. This allows direct comparision (and or locking) with optical frequency standards.



Master Oscillator: Passively Mode-Locked Er-fiber lasers



Ippen et al. Design: Opt. Lett. 18, 1080-1082 (1993)



- diode pumped
- sub-100 fs to ps pulse duration
- 1550 nm (telecom) wavelength for fiber-optic component availability
- repetition rate 30-100 MHz