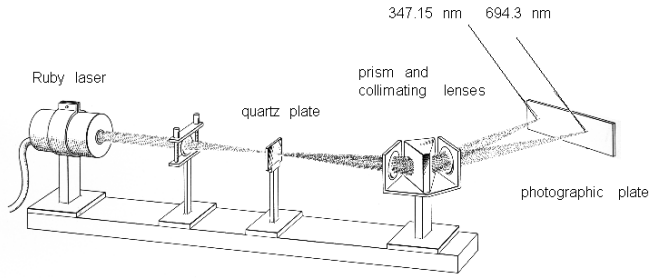
3C43

LASERS
&
MODERN OPTICS

5 Non-linear optics

## In the beginning

Second-harmonic generation of Ruby laser light in quartz (Franken, 1961)




Conversion efficiency $10^{-4}$ to $10^{-6}$ %

Nowadays efficiency ≈ 50% is achievable exploiting resonant enhancement and phase-matching.

## The anharmonic oscillator

Origin of non-linear effects:

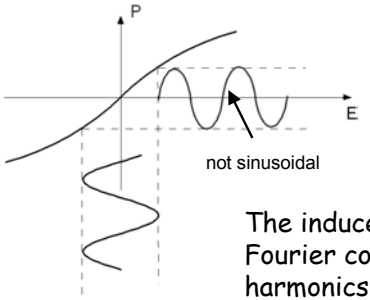
For large driving amplitude, the induced polarization is no longer a linear function of E:
e.g. The r.m.s. electric field in a 10 mJ Nd:YAG laser pulse lasting 10ns is, using $P = \frac{1}{2}\varepsilon_0 c\left|E_{r.m.s.}\right|^2$

is $E_{r.m.s.} \approx 5\times10^7\ V/m$

For comparison, the electric field in the lowest Bohr orbit of a hydrogenic atom

$$E_{atomic} = \frac{e}{4\pi\varepsilon_0 a_0^2} \approx 3\times10^8\ V/m$$

(For sunlight $\mathrm{P} \approx 20\ \mathrm{W/m^2} \Rightarrow E \approx 1 V/cm$ )




The induced polarisation has Fourier components at harmonics of the frequency of the incident light

## The non-linear polarisation

We can write the induced polarisation as the sum of a linear and a non-linear term

$$\mathrm{P} = \mathrm{P}^{\mathrm{L}} + \mathrm{P}^{\mathrm{NL}}$$

where $\mathrm{P}_i^{\mathrm{L}} = \varepsilon_0 \chi_{ij}^{(1)} \mathrm{E}_j$ the linear polarisation

$$\mathrm{P}_i^{\mathrm{NL}} = \sum_{n=2}^{\infty} \mathrm{P}_i^{(n)}$$ the non-linear polarisation

and $\mathrm{P}_i^{(n)} = \varepsilon_0 \chi_{ijk\ldots m}^{(n)} E_j E_k \ldots E_m$ n amplitudes

- $P^{(n)}$ generally has a number of frequency components (for n even(odd) all linear combinations of an even(odd) number of the incident frequencies up to n).

e.g. For n=2 and $E = E_1 \cos\omega_1 t + E_2 \cos\omega_2 t$

there are frequency components at

$$\omega = 0,\ 2\omega_1,\ 2\omega_2,\ \omega_1 - \omega_2,\ \omega_1 + \omega_2$$

- The second-order susceptibility at frequency $w_p$ is a function of three frequencies:

$$\chi^{(2)}(\omega_{p;}\omega_1, \omega_2)$$

Under suitable conditions, one particular frequency-component can dominate.

## The non-linear wave-equation

Maxwell's equations lead to:

$$\nabla^2\vec{E} - \frac{1}{c^2}\frac{\partial^2\vec{E}}{\partial t^2} = \mu_0\frac{\partial^2\vec{P}}{\partial t^2}$$

where P is the induced polarisation.
(no free charges, non-magnetic medium)

With $\vec{P} = P_L + P_{NL}$

$$= \varepsilon_0\chi^{(1)}\vec{E} + P_{NL}$$

The wave equation becomes:

$$\nabla^2\vec{E} - \frac{n^2}{c^2}\frac{\partial^2\vec{E}}{\partial t^2} = \mu_0\frac{\partial^2\overrightarrow{P_{NL}}}{\partial t^2}$$

known as the non-linear wave-equation.

## Symmetry considerations

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

Under reversal of the coordinate system: $\chi^{(2)} \rightarrow \tilde{\chi}^{(2)}$

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

Even-order terms
observed in media without inversion symmetry
(anisotropic crystalline materials)

e.g. second-harmonic generation,
(frequency components are linear combinations of an even number of frequencies)

Odd-order terms
observed in media inversion symmetry
(gases, liquids and isotropic crystals)

e.g. third-harmonic generation,
(frequency components are linear combinations of an odd number of frequencies)

## Solving the non-linear wave equation

e.g. Second-harmonic generation

- Assumptions:
  - P parallel to E, so can use a scalar equation
  - Incident plane-wave propagating along Oz
  - only one frequency of $P^{(2)}$ is important, the second-harmonic

Incident wave: $E_1 = \mathcal{E}_1 \exp i(\omega t - k_\omega z)$

Generated wave: $E_2 = \mathcal{E}_2(z) \exp i(2\omega t - k_{2\omega} z)$

Induced polarisation: $P^{(2)} = \varepsilon_0 \chi^{(2)} E_1^2$

$$= \varepsilon_0 \chi^{(2)} \mathcal{E}_1^2 \exp 2i(\omega t - k_\omega z)$$

Substituting in the non-linear wave-equation, writing for E the total electric field:

$$\frac{\partial^2 E_1}{\partial z^2} + \frac{\partial^2 E_2}{\partial z^2} - \frac{n_\omega^2}{c^2}\frac{\partial^2 E_1}{\partial t^2} - \frac{n_{2\omega}^2}{c^2}\frac{\partial^2 E_2}{\partial t^2} = \mu_0 \varepsilon_0 \chi^{(2)} \frac{\partial^2 (E_1^2)}{\partial t^2}$$

Work out all the terms:

$$\frac{\partial^2 E_1}{\partial z^2} = -k_\omega^2 \mathcal{E}_1 \exp i(\omega t - k_\omega z)$$

$$\frac{\partial^2 E_2}{\partial z^2} = -k_{2\omega}^2 \mathcal{E}_2 \exp i(2\omega t - k_{2\omega} z) - 2i\, k_{2\omega} \frac{\partial \mathcal{E}_2(z)}{\partial z} \exp i(2\omega t - k_{2\omega} z) + \cancel{\frac{\partial^2 \mathcal{E}_2(z)}{\partial z^2} \exp i(2\omega t - k_{2\omega} z)}$$

$$k_{2\omega} \frac{\partial \mathcal{E}_2(z)}{\partial z} >> \frac{\partial^2 \mathcal{E}_2(z)}{\partial z^2}$$

since $E_2$ varies little on the scale of a wavelength (slowly-varying envelope approximation).

$$\frac{n_\omega^2}{c^2}\frac{\partial^2 E_1}{\partial t^2} = -k_\omega^2 \mathcal{E}_1 \exp i(\omega t - k_\omega z)$$

$$\frac{n_{2\omega}^2}{c^2}\frac{\partial^2 E_2}{\partial t^2} = -k_{2\omega}^2 \mathcal{E}_2(z) \exp i(2\omega t - k_{2\omega} z)$$

$$\mu_0 \frac{\partial^2 P^{(2)}}{\partial t^2} = \frac{-4\omega^2}{c^2} \chi^{(2)} \mathcal{E}_1^2 \exp 2i(\omega t - k_\omega z)$$

Substitute everything into the non-linear wave equation and compare coefficients of $\exp 2i\omega t$ on both sides of the equation:

$$\frac{\partial \mathcal{E}_2(z)}{\partial z} = -2i \frac{\omega^2/c^2}{k_{2\omega}} \chi^{(2)} \mathcal{E}_1^2 \exp 2i\,\Delta k\, z$$

where $\Delta k \equiv \frac{1}{2} k_{2\omega} - k_\omega$

For $\mathcal{E}_2(0) = 0$ the solution gives:

$$\mathcal{E}_2(L) = -i \frac{2\omega^2/c^2}{k_{2\omega}} \chi^{(2)} \mathcal{E}_1^2 \exp(i\Delta k\, L)\; L \frac{\sin \Delta k\, L}{\Delta k\, L}$$

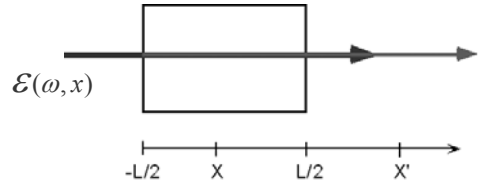
So the intensity of the second-harmonic wave is proportional to

$$I_2(L) = \left|\chi^{(2)}\right|^2 I_1^2 \;\; L^2 \; \frac{\sin^2(\Delta k\, L)}{(\Delta k\, L)^2}$$

The coherence length, Lc, is given by $L_c = \pi / \Delta k$

The second-harmonic intensity increases for L up to $L_c$, then decreases.

Alternative (less rigourous) derivation of S.H.G. amplitude




$$\mathcal{E}(\omega, x) = \mathcal{E}_1 \exp i(\omega t - k_\omega x)$$

$$\mathcal{E}^2(\omega, x) = \mathcal{E}_1^2 \exp 2i(\omega t - k_\omega x)$$

Propagation term

$$\delta\mathcal{E}(2\omega, x) = \mathcal{E}_1^2 \exp 2i(\omega t - k_\omega x)\; \exp - i\, k_{2\omega}(x' - x) =$$
$$\mathcal{E}_1^2 \exp i(2\omega t - k_{2\omega} x')\; \exp i(k_{2\omega} - 2k_\omega)x$$

Integrating over $x$

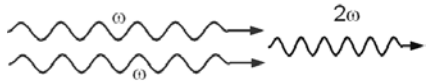
$$\mathcal{E}(2\omega, L/2) = \mathcal{E}_1^2 \exp i(2\omega t - i\, k_{2\omega} x') \int_{-L/2}^{+L/2} \exp i(k_{2\omega} - 2k_\omega)x \cdot dx$$

$$\left|\mathcal{E}(2\omega, x')\right|^2 \propto L^2 \; \frac{\sin^2(k_{2\omega}/2 - k_\omega)L}{(k_{2\omega}/2 - k_\omega)^2 L^2}$$

## The photon picture

The energy of a photon of angular frequency $\omega$ is $\hbar\omega$ and its momentum $\hbar k = \frac{h}{\lambda}$

Second-harmonic generation conserves energy because two photons of frequency $\omega$ are destroyed for every one at frequency $2\omega$ created:




The condition for maximum efficiency in the second-harmonic generation process is

$$\Delta k \equiv \tfrac{1}{2} k_{2\omega} - k_{\omega} = 0$$

Multiplying by $2\hbar$, we get $\hbar k_{2\omega} - 2\hbar k_{\omega} = 0$ , which is just an expression of momentum conservation.

Since $k_{\omega} = \frac{n_{\omega}\omega}{c}$ and $k_{2\omega} = \frac{n_{2\omega}(2\omega)}{c}$

the condition for momentum to be conserved is

$$n_{2\omega} = n_{\omega}$$

The art of arranging for $n_{2\omega} = n_{\omega}$ is known as phase-matching or index-matching.

## Phase-matching in second-harmonic generation

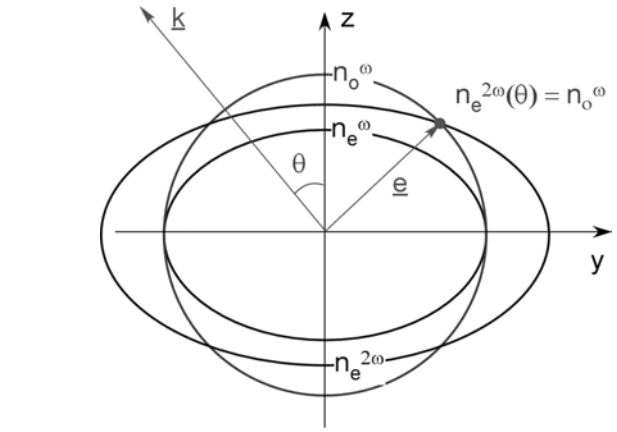
• SHG requires a medium without inversion symmetry, an anisotropic crystalline material (e.g. KDP)

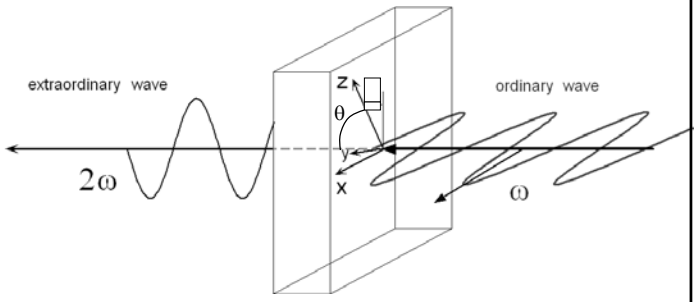
• An anisotropic crystal is also birefringent, so we can profit from the frequency dependence of the birefringence to achieve phase-matching.

e.g. second-harmonic generation in a negative uniaxial crystal (KDP) with normal dispersion ($n_{2w} > n_w$)

Phase-matching is achieved for an appropriate propagation direction and polarisation (e.g. Type 1 phase-matching).

## Type 1 phase-matching




Calculating the phase-matching angle

The index ellipsoid (at frequency 2ω) is:

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

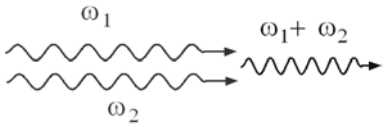
so the phase-matching condition

$$n_e^{2\omega}(\theta_m) = n_o^{\omega}$$

gives $$\sin^2\theta_m = \frac{\{n_o^{\omega}\}^{-2} - \{n_o^{2\omega}\}^{-2}}{\{n_e^{2\omega}\}^{-2} - \{n_o^{2\omega}\}^{-2}}$$ !

## Other second-order processes

Second-harmonic generation is just a special case of sum-frequency generation

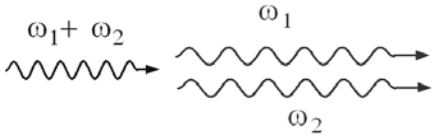



Energy conservation: $\hbar\omega_1 + \hbar\omega_2 = \hbar(\omega_1 + \omega_2)$

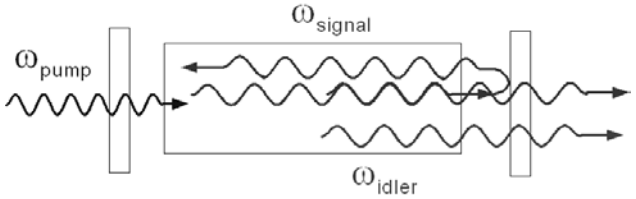
The phase-matching condition is a bit more complicated:

$$\hbar\vec{k}_1 + \hbar\vec{k}_2 = \hbar\vec{k}_{sum}$$

Parametric amplification is sum-frequency generation in reverse:




## The optical parametric oscillator




The build up of light in the cavity at $\omega_{signal}$ stimulates the process:

$$\hbar\omega_{pump} \Rightarrow \hbar\omega_{signal} + \hbar\omega_{idler}$$

whilst the escape from the cavity of the idler beam prevents the reverse process:

$$\hbar\omega_{signal} + \hbar\omega_{idler} \Rightarrow \hbar\omega_{pump}$$

The result is a widely-tunable source of coherent radiation. Tune by varying cavity length (can be achieved by changing temperature of crystal).

e.g. Titanium-Sapphire laser (Ar+ laser pumped) temperature tuning range 700-1100 nm (for 10°C)

e.g. $LiNbO_3$ (doubled Nd:YAG pumped) tuning range 980-1150 nm

## The 3rd Order Polarization:

$$\mathrm{P}_i^{(3)}(\omega_4) = \varepsilon_0\ \chi^{(3)}_{ipqr}(\omega_4;\omega_1,\omega_2,\omega_3)\times \mathrm{E}_{1p}(\omega_1)\,\mathrm{E}_{2q}(\omega_2)\,\mathrm{E}_{3r}(\omega_3)$$

$$\chi^{(3)}_{ipqr}(\omega_4;\omega_1,\omega_2,\omega_3)$$

number of independent elements reduced by structural symmetries of medium

components at 25 distinct frequencies

Off-resonance $\mathrm{P}^{(3)} / \mathrm{P}^{(2)} \approx E / E_{at}$

but:

- we can exploit resonant enhancement
- gaseous media allow higher power density

## The Kerr Effect

- The optical frequency Kerr effect

For an incident monochromatic optical field, the expression for the polarization becomes:

$$\vec{\mathrm{P}}^{(3)}(\omega) = \varepsilon_0\ \chi^{(3)}(\omega)\ \left|E\ (\omega)\right|^2\ \vec{E}(\omega)$$

(Since the medium is isotropic, ***P*** is necessarily in the direction of ***E***)

Equivalent to an intensity-dependent refractive index:

$$n = n_0 + n_2 I, \qquad n_2 = \tfrac{1}{\varepsilon_0 c}\,\chi^{(3)}$$

Gives rise to:

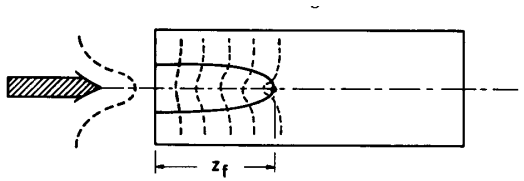
- self-focussing, -defocussing
- self phase-modulation

## Self-focusing

- Solids

Mechanism usually electrostriction ⟶ $n_2 > 0$

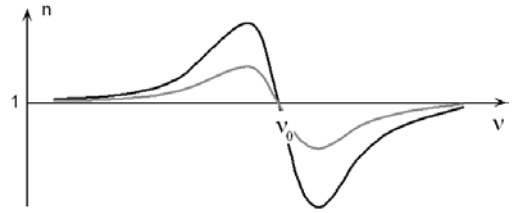
An incident Gaussian beam is focussed onto the beam axis




… often catastrophically

Damage caused by self-focusing

- Gases

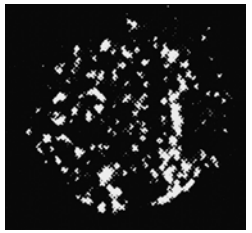



—— $I < I_{sat}$

—— $I > I_{sat}$

Near an atomic resonance we get
self-focussing for blue-detuning
self-defocussing for red detuning

(the situation is reversed if there is a population-inversion)



Self-focussing causes filamentation of a laser beam

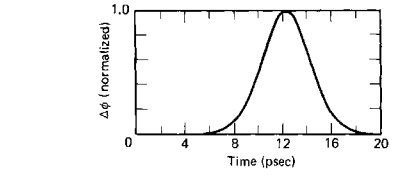
- Self-phase modulation

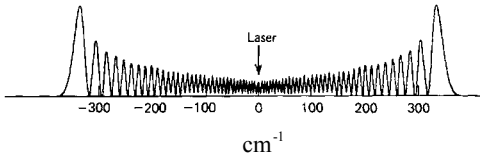
The changing refractive index during a laser pulse modulates the frequency of the pulse:

$$\Delta n(t) = n_2\, I(t)$$

$$\Delta\varphi(t) = \frac{\omega}{c}\ell\ \Delta n(t) = \frac{\omega}{c}\ell\ n_2\, I(t)$$

$$\Delta\omega(t) = \frac{\omega}{c}\ell\ n_2\ \frac{\partial}{\partial t} I(t)$$




An incident nearly monochromatic optical pulse emerges with a broad frequency spectrum.

## Third-harmonic Generation

$$\vec{P}^{(3)}(3\omega) = \varepsilon_0\, \chi^{(3)}(3\omega;\omega,\omega,\omega)\ \left|E\ (\omega)\right|^2\ \vec{E}(\omega)$$

Can't use birefringence to ensure phase-matching in an isotropic medium, but need

$$n_{3\omega} = n_{\omega}$$

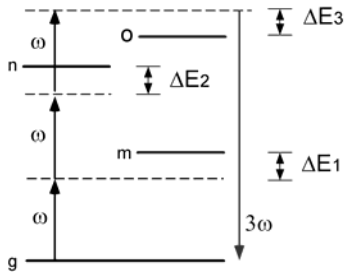
Solution:

1. Use gases e.g. alkali-metal vapour
2. Exploit resonant enhancement

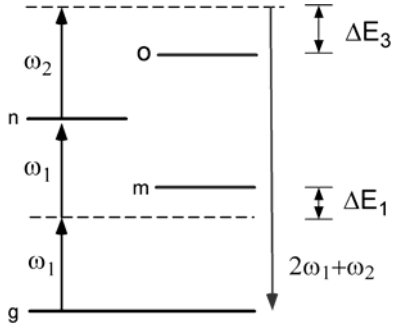
Perturbation theory:

$$\chi^{(3)}(3\omega) \propto \sum_{pqr} \frac{\langle g|V|p\rangle\langle p|V|q\rangle\langle q|V|r\rangle}{\Delta E_1\, \Delta E_2\, \Delta E_3}$$

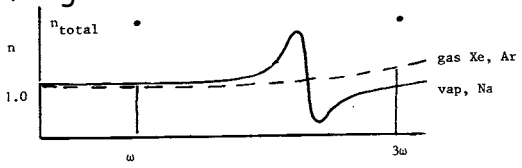
$$V = -er \cdot E_{total}$$



2-photon resonant THG gives less absorption of $\omega$, $3\omega$ and tunable VUV radiation




3. Phase-match by exploiting anomalous dispersion of the active gas near resonance and the normal dispersion of an inert buffer gas




$$n_{vap}(\omega) + n_{buffer}(\omega) = n_{vap}(3\omega) + n_{buffer}(3\omega)$$

Efficiency up to 30%