

## An Introduction to Nonlinear Optics for Accelerator Diagnostics

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# What do we mean by nonlinear?



Examples:

Cheap stereos turned up too loud Electric guitar distortions (clipping)

# **Linear Optics**

- Properties independent of light intensity
  - Linear refractive index Snell's Law
  - Linear absorption Beer-Lambert Law
- Super position principle
  - Can look at inputs singularly and sum at the end
- Frequency of light is constant
  - $v_{in} = v_{out}$
- Beams do not interact
  - You can cross the beams with no effect
  - (not counting interference effects...)

# **Nonlinear Optics**

- Refractive index and absorption can be a function of intensity
  - Two photon absorption
  - Self focussing
- Superposition no longer true
  - Sum of responses to two inputs not the same as response to both inputs simultaneously
- Optical frequency can be changed
  - Mixing processes
  - Harmonic generation
- Beams can interact
  - Crossing the beams makes interesting things happen

# Which Media are Nonlinear?

#### All of them!

- Solids But it's only observable if you
- pump them hard enough! Liquids
- Gasses
- Plasmas

In general, lasers are required to observe nonlinear effects

Vacuum

(via generation of vitual e- e+ pairs)

# The Birth of Nonlinear Optics

# The field was kick started in 1961, by the first demonstration of second harmonic generation



FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

# This was made possible by the realisation of the ruby laser the previous year

# Origin of the Nonlinear Response

The polarisation of atoms and molecules is affected by an incident EM wave



# Origin of the Nonlinear Response



Incident light affects the polarisation of atoms/molecules throughout the medium, setting up a polarisation field.

This field reradiates EM waves, and in the linear regime is the origin of the refractive index.

# Origin of the Nonlinear Response

The expression for the material polarisation field can include terms for the nonlinear responses



Note – for second order effects the material must not have inversion symmetry  $P(E) = \varepsilon_0 \chi^{(2)} E^2$ If P(E) = -P(-E) then  $\chi^{(2)} = 0$ 

#### Effects of the Nonlinearity

Consider an electric field,  $E = E_1 \cos(kz - \omega t)$ 

$$P = \varepsilon_0 \chi^{(1)} E_1 \cos(kz - \omega t) + \varepsilon_0 \chi^{(2)} E_1^2 \cos^2(kz - \omega t)$$

$$P = \varepsilon_0 \chi^{(1)} E_1 \cos(kz - \omega t) \\ + \varepsilon_0 \chi^{(2)} E_1^2 \frac{1}{2} (1 + \cos(2kz - 2\omega t))) \\ \uparrow \qquad \uparrow \qquad \uparrow \\ DC \text{ offset} \qquad Doubled frequency} \\ Optical \qquad Second Harmonic \\ Rectification \qquad Generation \\ \end{bmatrix}$$

## Effects of the Nonlinearity

Hang on, we now have two waves

$$E = E_1 \cos(kz - \omega t) + E_2 \cos(2kz - 2\omega t)$$

Only considering second order response:

$$P_{NL} = \varepsilon_0 \chi^{(2)} E^2$$

$$P_{NL} = \varepsilon_0 \chi^{(2)} [E_1^2 \cos^2(kz - \omega t) + 2E_1 E_2 \cos(kz - \omega t) \cos(2kz - 2\omega t) + E_2^2 \cos^2(2kz - 2\omega t)]$$
Original
Original

$$P_{NL} = \varepsilon_0 \chi^{(2)} [E_1^2 \frac{1}{2} (1 + \cos(2kz - 2\omega t)) + E_1 E_2 (\cos(3kz - 3\omega t) + \cos(kz - \omega t)) + E_2^2 \frac{1}{2} (1 + \cos(4kz - 4\omega t))]$$

Original frequency again!

These new frequencies would also mix to generate even more frequencies! Which processes will occur?

#### **Sum- and difference-frequency generation**

Suppose there are two different-color beams present:

 $\mathscr{C}(t) \propto E_1 \exp(i\omega_1 t) + E_1^* \exp(-i\omega_1 t) + E_2 \exp(i\omega_2 t) + E_2^* \exp(-i\omega_2 t)$ So:

$$\mathscr{C}(t)^{2} \propto E_{1}^{2} \exp(2i\omega_{1}t) + E_{1}^{*2} \exp(-2i\omega_{1}t)$$

$$+ E_{2}^{2} \exp(2i\omega_{2}t) + E_{2}^{*2} \exp(-2i\omega_{2}t)$$

$$+ 2E_{1}E_{2} \exp[i(\omega_{1} + \omega_{2})t] + 2E_{1}^{*}E_{2}^{*} \exp[-i(\omega_{1} + \omega_{2})t]$$

$$+ 2E_{1}E_{2}^{*} \exp[i(\omega_{1} - \omega_{2})t] + 2E_{1}^{*}E_{2} \exp[-i(\omega_{1} - \omega_{2})t]$$

$$Diff-freq gen$$

$$+ 2|E_{1}|^{2} + 2|E_{2}|^{2}$$

$$dc rectification$$

Note also that, when  $\omega_i$  is negative inside the exp, the *E* in front has a \*.

# Phasematching

- It seems that a there is potentially an awful lot going on
- Especially consider the cascading of processes that appears unavoidable
- How can we make practical use of these effect?
- Well, all the processes happen, but in general with low efficiencies
- We can drastically enhance efficiency via "phasematching"

# Phasematching

Let's consider SHG again. Throughout the medium, the polarisation field is oscillating at

 $\cos(2k_1z - 2\omega_1t)$ Propagation constant  $k = \frac{\omega n(\omega)}{c}$ 

This is radiating into an EM field at  $\omega_2 = 2\omega_1$ , with  $k_2 = \frac{2\omega \cdot n(2\omega)}{c}$  $\cos(k_2 z - \omega_2 t)$ 

Dispersion in a material cause a phase slippage between generated and propagating waves at  $2\omega_1$ 

#### **Phasor Representation**





# Phasematching

The phase-matching condition for SHG:

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

Unfortunately, dispersion prevents this from happening!



Except in very rare cases near absorption features.

# **Birefringent Phasematching**

#### Some materials exhibit Birefringence

i.e. there are two (or three) orthogonal, principle refractive indicies



# The Index Ellipsoid

Define a "uniaxial" crystal as ordinary axis with

$$n_o = n_x = n_y$$

extraordinary axis

 $n_z$ 

The **optic axis**, is the direction in which both polarisations of light experience the same refractive index.





The polarisation must lie in a direction awhich is perpendicular to k0 = a.k $0 = {a_x \choose a_y}.{sin \theta \choose cos \theta}$  $a = \pm {-cos \theta \choose sin \theta}$  $\frac{1}{n_e^2} = \left(\frac{a_x}{n_y}\right)^2 + \left(\frac{a_z}{n_z}\right)^2$ 

This method can be extended to into 3D for "biaxial" crystals  $n_z > n_y > n_x$ 

# **Birefringent Phasematching**

**Birefringent** materials have different refractive indices for different polarizations. **Ordinary** and **Extraordinary** refractive indices can be different by up to 0.1 for SHG crystals.

We can now satisfy the phase-matching condition.

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

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



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

# Light created in real crystals



Note that SH beam is brighter as phase-matching is achieved.

# Walk-Off



In a real crystal the electrons experience stronger restoring forces in some directions than in others.

Generally *E* and *P* are not parallel (except along principle axes – this is a consequence of how they are defined)

A result of this is that direction of energy flow, given by the Poynting Vector  $(s = E \times H)$ , is not parallel to the k vector of the wave. i.e. the wave will undergo "walk-off"

## The Nonlinear Tensor

Conventionally the susceptibility is replaced by the nonlinear tensor  $2d_{ijk}$ 

$$\mathbf{P}_{i} = \varepsilon_{0} \boldsymbol{\chi}_{ijk}^{(2)} \mathbf{E}_{j} \mathbf{E}_{k} = \varepsilon_{0} (2\mathbf{d}_{ijk}) \mathbf{E}_{j} \mathbf{E}_{k}$$

 $\mathbf{d}_{ijk}$  is a 3x3x3 tensor, and maps all components of the vector  $\mathbf{E}$  to vector  $\mathbf{P}$ 

Consider: d(i,j,k) describes the nonlinear coupling of fields E<sup>a</sup> and E<sup>b</sup> along j and k into the polarisation along i



Note that the order of the E fields is not important  $d_{232} = d_{223}$ 

## Simplifications of the Nonlinear Tensor

Using the "piezoelectric contraction" the nonlinear response is now (order of fields not important)  $E_X^a E_X^b$ 

$$\begin{bmatrix} P_{X} \\ P_{Y} \\ P_{Z} \end{bmatrix} = 2\varepsilon_{o} \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_{Y}^{a}E_{Y}^{b} \\ E_{Z}^{a}E_{Z}^{b} \\ E_{Y}^{a}E_{Z}^{b} + E_{Z}^{a}E_{Y}^{b} \\ E_{X}^{a}E_{Z}^{b} + E_{Z}^{a}E_{X}^{b} \\ E_{X}^{a}E_{Y}^{b} + E_{Y}^{a}E_{X}^{b} \end{bmatrix}$$
  
lements i k have been replaced with L

Where elements j,k have been replaced w

jk	11	22	33	23, 32	13, 31	12, 21
I	1	2	3	4	5	6

We can go one step further in lossless media, the Kleinman's contraction For given *i,j,k* then  $d_{ijk} = d_{ikj} = d_{kji} = d_{kji} = d_{jik} = d_{jki}$ 

$$d_{i,l} = \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{bmatrix}$$

Now there are only 10 independent values!

The actual elements present depends on the crystal symmetry.

# The Effective Nonlinear Coefficient

An example:

LBO crystal, E-fields along (1,1,0) and (1,-1,0)



 $P = [0, 0, \varepsilon_o(d_{15} - d_{24})]$ 

Polarisation response is along z

$$\frac{(d_{15}-d_{24})}{2}$$
 is the **effective nonlinear coefficient**,  $d_{eff}$ 

# Maxwell's Equations

Evaluating the effect of an induced polarization on a wave requires solving Maxwells equations, with an induced polarization term.

This gives rise to an extra term in the wave equation:

$$\nabla^2 \mathbf{E} - \mu_0 \varepsilon \frac{\partial^2 \mathbf{E}}{\partial t^2} = \left( \mu_0 \frac{\partial^2 \mathbf{P}_{NL}}{\partial t^2} \right)$$

(Normally = 0)

This is the Inhomogeneous Wave Equation.

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

## **Coupled Wave Equations**

If we consider the interaction of 3 waves propagating in the z direction

$$\begin{split} E_{i}^{(\omega_{1})}(z,t) &= \frac{1}{2} \left( E_{1i} \exp[i(\omega_{1}t - k_{1}z)] + \text{ c.c.} \right) \\ E_{k}^{(\omega_{2})}(z,t) &= \frac{1}{2} \left( E_{2k} \exp[i(\omega_{2}t - k_{2}z)] + \text{ c.c.} \right) \\ E_{j}^{(\omega_{3})}(z,t) &= \frac{1}{2} \left( E_{3j} \exp[i(\omega_{3}t - k_{3}z)] + \text{ c.c.} \right) \end{split}$$

i,j,k are the wave polarisation, and can be x or y

Substituting the three waves to the inhomogeneous wave equation we get a set of 3 <u>coupled wave equations</u>

E is complex as it contains	$\frac{dE_{1i}}{dz} = -\frac{i\omega_1}{cn_1} d_{ijk} E_{3j} E_{2k}^* \exp(-i\Delta kz)$ $\frac{dE_{2k}^*}{dz} = -\frac{i\omega_2}{cn_2} d_{kij} E_{1i} E_{3j}^* \exp(-i\Delta kz)$	Phase mismatch
a static phase term	$\frac{dE_{3j}}{dz} = -\frac{i\omega_3}{cn_3} d'_{jik} E_{1i} E_{2k} \exp(+i\Delta kz)$	Conjugate terms arise from dropping $\omega_2 + \omega_3$ components

#### **Manley-Rowe Relations**

$$\frac{n_1 d|E_1|^2}{\omega_1 dz} = \frac{n_2 d|E_2|^2}{\omega_2 dz} = -\frac{n_3 d|E_3|^2}{\omega_3 dz} = i \frac{d_{ijk}}{c} E_1^* E_2^* E_3 \exp(-i\Delta kz)$$
 "Photon Splitting" Picture  

$$I = nc\varepsilon_o |E|^2 / 2$$

$$\frac{1}{\omega_1} \frac{dI_1}{dz} = \frac{1}{\omega_2} \frac{dI_2}{dz} = -\frac{1}{\omega_3} \frac{dI_3}{dz}$$

"Parametric" – no absorption

# **Example Analysis of SHG**

Using the coupled wave equations we can now analyse second order processes.

All we need to do is apply appropriate assumptions and boundary conditions, and then solve the equations.

Quick example for SHG:  $\omega_3 = 2\omega_2 = 2\omega_1$ 

Assume negligible depletion

$$\frac{dE_1}{dz} = \frac{dE_2}{dz} = 0 \qquad \qquad \frac{dE_3}{dz} = -\frac{i\omega_3}{cn_3}d'_{jik}E_1E_2\exp(+i\Delta kz)$$

Trial solution

$$E_3 = -a \exp(+i\Delta kz) + b$$
  $\frac{dE_3}{dz} = -i\Delta ka \exp(+i\Delta kz)$ 

$$a = -\frac{\omega_3 d'_{jik} E_1 E_2}{cn_3 \Delta k} \qquad E_3 = 0 \ @z = 0 \Rightarrow \qquad b = -a = \frac{\omega_3 d'_{jik} E_1 E_2}{cn_3 \Delta k}$$

### SHG Analysis Continued...

$$I_{3} = E_{3}E_{3}^{*} = \frac{4\omega^{2}d_{jik}^{*}^{2}I_{\omega}^{2}}{c^{2}n_{3}^{2}\left(\frac{\Delta k}{2}\right)^{2}} \left(1 - \exp(+i\Delta kz)\right) \left(1 - \exp(+i\Delta kz)\right)^{*}$$

Gain depends on wavelength  $\$ 

Phasematching is important!

$$I_{3} = \frac{4\omega^{2}d_{jik}^{2}I_{\omega}^{2}z^{2}}{c^{2}n_{3}^{2}} \times \frac{\sin^{2}\left(\frac{\Delta kz}{2}\right)}{\left(\frac{\Delta kz}{2}\right)^{2}}$$

Signal grows quadratically

with length when phasematched

#### Don't worry, we're not going to derive all the $\chi^{(2)}$ processes!

# Quasiphasematching

Highest nonlinear coefficients don't generally match with allowed phasematching conditions



Power oscillates between second harmonic and fundamental

#### Quasiphasematching



$$\frac{dE_{1i}}{dz} = -\frac{i\omega_1}{cn_1} d_{ijk} E_{3j} E_{2k}^* \exp(-i\Delta kz) \times \exp(i\pi)$$

$$\frac{dE_{2k}^*}{dz} = -\frac{i\omega_2}{cn_2} d_{kij} E_{1i} E_{3j}^* \exp(-i\Delta kz) \times \exp(i\pi)$$

$$\frac{dE_{3j}}{dz} = -\frac{i\omega_3}{cn_3} d_{jik} E_{1i} E_{2k} \exp(+i\Delta kz) \times \exp(i\pi)$$

$$\exp(i\pi) = -1$$

Change the sign of  $d_{ijk}$ ?

#### Quasiphasematching



This is possible in ferro-electric materials, e.g. lithium niobate, by applying a periodic high electric field which flips the material domains. Efficiency is improved by an order of magnitude!

# Non-colinear phasematching

The wave vector, k, is actually a vector property.

This allows us to have non-colinear waves and still achieve phasematching.

One application is the compensation of "walk-off"



If not compensation for walk-off, this has the effect of limiting the interaction length

In an appropriate material, it can also be arranged that multiple wavelengths phasematch with the same propagation angle, leading to a very large bandwidth for the nonlinear process.



# Sum Frequency Generation

SHG was a special case of SFG

SFG is the condition of a weak wave at  $\omega_1$  mixing with a strong (undepleted) wave at  $\omega_2$  to generate a wave at  $\omega_3$ 



In the general, low conversion efficiency case

$$I_3 = \frac{2\omega_3 d_{ijk}^2 I_1 I_2 z^2}{n_1 n_2 n_3 c^3 \varepsilon_0} \operatorname{sinc}^2 \left(\frac{\Delta kz}{2}\right)$$

Otherwise, energy is swapped back and forth between the weak wave  $\omega_1$  and the sum frequency  $\omega_3$ , the "pump" at  $\omega_2$ changes little

 $\omega_1$ 

# **Difference Frequency Generation**

Here we consider a strong (undepleted) pump wave at  $\omega_3$ , which mixes with a weak wave at  $\omega_1$  to give rise to a wave at  $\omega_2$ 



i.e. both waves experience gain at the expense of the "pump"

\*Starting from low conversion conditions, it takes the quadratic form of the SFG solution

# Optical Parametric Generation/Oscillation

- For DFG, both of the down-converted (lower frequency) waves experience gain
- Additionally, there exists spontaneously emitted photons (thermal, quantum noise)
- If the gain is sufficiently high, the few photons that are generated with the correct polarisation, frequency, and direction, will be amplified
  - i.e. you can get new wavelengths without the need to seed the process!



Broadly tuneable laser-like sources

## Pockel's Effect

The second order nonlinearity can also be exploited to modulate the phase, amplitude or frequency of light by applying a suitable electric field

Consider the polarisation response:  $P = \varepsilon_o(\chi^{(1)}E + \chi^{(2)}E^2 + ...)$ 

And define the total susceptibility: 
$$\chi = \frac{P}{\varepsilon_o E}$$
  
Now, the refractive index is:  $n = \sqrt{1 + \chi} = (1 + \chi^{(1)} + \chi^{(2)}E + ...)^{1/2} \approx (1 + \frac{\chi^{(2)}E}{n_o^2})^{1/2} n_o$   
Using aTaylor series approx.  $n = n_o + \frac{\chi^{(2)}E}{2n_o}$ 

This effect was observed >50 years before the invention of the laser!

# **Important Considerations**

- Temperature range for phasematching
- Angular acceptance for phasematching
- Which element of d<sub>eff</sub> to use
- Focussing trade-off





## **Ultrashort Pulses**

Need to consider Group Velocities

$$v_g = \frac{\partial \omega}{\partial k}$$

Starting at t=0 Say all beams overlap



3 pulses begin to separate Conversion still happens in overlap Pulses are separate No more conversion Generated pulse "stretched"

#### Now, Some Applications

# Photo Injector System

A short, intense, pulse of high energy photons is required at the photocathode in order to liberate electrons for acceleration.

Lasers can produce the synchronised, high intensity pulses, except they are at **much longer wavelengths**!

e.g. Ti:Sapphire 800nm, Nd 1064nm, Er 1550nm

The solution, as we know, is nonlinear optics.

What if we want UV light, as required by some photocathodes.

Try 3<sup>rd</sup> harmonic of Ti:Sapphire?

# **Third Harmonic Generation**

Can be achieved via a 3<sup>rd</sup> order process but a very high pump field is required due to the weak susceptibility

Thankfully there is a better way – cascaded second order processes!

$$\lambda_{SHG} = \left(\frac{1}{\lambda_p} + \frac{1}{\lambda_p}\right)^{-1}, \lambda_{SFG} = \left(\frac{1}{\lambda_{SHG}} + \frac{1}{\lambda_p}\right)^{-1} = \left(\frac{1}{3\lambda_p}\right)^{-1} = 3\lambda_p$$
Can be 50%+ efficient Also quite efficient
emtosecond
Ti:Sapphire Second Harmonic Generation Generation Generation Generation Generation Generation Generation Generation Also Generation Gen

# **Electron Bunch Length Diagnostics**

- A number of techniques are being refined for the optical characterisation of the bunch temporal profile.
- Generally, this is achieved by shifting characteristics of the coulomb field onto an optical frequency carrier beam
- This optical signal is then characterised in order to infer properties of the electron bunch

The field of THz generation and detection by modelocked lasers is well developed, and as the coulomb field is similar in nature to a THz pulse, similar techniques are used.

Typically, THz detection is performed via the electro-optic effect.

#### Physics of EO encoding ... standard description

Refractive index modified by external (quasi)-DC electric field



quasi-DC description ok if  $\tau_{laser} \ll$  time scale of  $E_{DC}$  variations (basis for Pockels cells, sampling electro-optic THz detection, ...)

**N.B.** Time-varying refractive index is a restricted approximation to the physics (albeit a very useful and applicable formalism for majority of situations )

# **Electron Bunch Length Diagnostics**

#### **Spectral Decoding**





- $\ensuremath{\circ}$  Chirped optical input
- Spectral readout
- $\circ$  Use time-wavelength relationship
- O Ultrashort optical input
  O Spatial readout (EO crystal)
  O Use time-space relationship

#### **Temporal Decoding**



- Spectral upconversion\*\*
- Long pulse + ultrashort pulse gate
   Spatial readout (cross-correlator crystal)
- $\circ$  Use time-space relationship
- monochomatic optical input (long pulse)
   Spectral readout
- $\circ$  \*\*Implicit time domain information only

#### Electro-optic detection bandwidth

description of EO detection as sum- and difference-frequency mixing



This is "Small signal" solution. High field effects c.f. Jamison Appl Phys B 91 241 (2008)

# GRENOUILLE



- Grating Eliminated No-nonsense Observation of Ultrafast Laser Light Efields...
- Spectrometer is replaced by a thick nonlinear crystal and a CCD the phasematching condition now determines the angular spread
- Delay stage has been replaced by a Fresnel Biprism, thereby mapping the delay across the CCD and allowing single shot pulse characterisation

## Thank you for listening