Generation of Ultrashort Pulses:

Basic concepts and nonlinear optics techniques

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OSA Ultrafast Optical Phenomena Technical Group Online Workshop

Why short pulses?



Blurry picture measured using an insufficiently short



When detectors are too slow...

... you need a light pulse which is faster than the process to be recorded

Fastest incoherent light pulse: **10-10**⁻⁹ s



- Lasers pulses: duration down to 10.10⁻¹⁸ s
- They are the **shortest artificial events** ever generated
- They are the ruler to measure ultrafast events in nature 5



Photograph taken by Harold Edgerton, MIT, 1964



Properties of a light pulse





Representation of optical pulses



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Examples of chronocyclic functions

Ludwig van Beethoven Symphony No. 5 C minor op. 67 Allegro con brio $\widehat{}$ ff audio frequency 0 0.5 1.0 1.5 2.0 2.5 time [s]

The musical score lives in the **time-frequency** domain



https://www.gw-openscience.org/GWTC-1/ DOI: 10.7935/82H3-HH23 - https://arxiv.org/abs/1811.12907

Examples of pulses



















Outline

Turn-key sources of ultrashort pulses

- Nonlinear processes: wave equations
 - General equation
 - Second order processes
 - Coupled nonlinear equations
 - Meaning of phase matching (I)
- Corpuscular view of second order processes
 - ✓ Manley-Rowe equations
 - Meaning of phase matching (II)
- Fulfilling phase matching
- Second order processes with pulses
 - ✓ Temporal overlap
 - Broadband phase matching

Laser mode locking

- A laser is an optical oscillator which consists of an optical amplifying medium enclosed between two high reflectance mirrors
- A laser allows longitudinal modes which are spaced in frequency by the resonance condition: $\Delta v = c/2L$.





In a mode-locked laser all the modes oscillate with a constant phase difference.
 How is this locking obtained?

 A mode-locked laser incorporates an ultrafast optical switch, which drives the ultrashort pulse generation



 The <u>saturable absorber</u> transmits the intense laser pulse and absorbs the less intense CW radiation

Kerr-lens mode-locking



A Kerr medium has a refractive index that depends on intensity, so it acts as a lens for the high intensity (pulsed) beam

Together with an aperture it induces a transmission that increases with intensity \rightarrow saturable absorber

Chirped pulse amplification (CPA)



- Ti:sapphire pumped by green laser: wavelength 800 nm Pulse energy 1-10 mJ Repetition rate 1-10 kHz Pulsewidth: 30-100 fs
- Diode-pumped Yb laser: wavelength 1040 nm Pulse energy 0.01-10 mJ Repetition rate 1kHz-2 MHz Pulsewidth: 150-300 fs

For both these sources the wavelength is fixed We need nonlinear optics to achieve frequency tunability

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Tuning by nonlinear optics

Spectral broadening
 third order effect -



Parametric amplification
 Second order effect -



Nonlinear interactions



C. Manzoni and G. Cerullo, "Design criteria for ultrafast optical parametric amplifiers", J. Opt. 18, 103501 (2016)

Nonlinear processes: wave equations

• Starting from Maxwell's equation for wave propagation:

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

High-intensity fields
Polarization:
$$P(z, t) = \underbrace{P_L(z, t)}_{\text{Weak fields}} + P_{\text{NL}}(z, t)$$

Weak fields

 $\frac{\partial^2 E}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_L}{\partial t^2} + \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}$

Nonlinear processes: wave equations

$$\frac{\partial^2 E}{\partial z^2} - \frac{1}{c_0^2} \frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P_L}{\partial t^2} + \mu_0 \frac{\partial^2 P_{NL}}{\partial t^2}$$

$$D(z, t) = \varepsilon_0 E(z, t) + P_{\rm L}(z, t)$$

General wave equation with nonlinear processes:

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \mu_0 \frac{\partial^2 D(z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 P_{\rm NL}(z,t)}{\partial t^2}$$

In the following: second order nonlinear processes:

$$P_{\rm NL}(z, t) = \varepsilon_0 \chi^{(2)} E^2(z, t) = 2\varepsilon_0 d_{\rm eff} E^2(z, t)$$

Second order processes



Let's start from 2 oscillating fields:

 $E(t) = A_1 \exp(-i\omega_1 t) + A_2 \exp(-i\omega_2 t) + cc$



Sum frequency generation (SFG)

Source of second order processes

3 interacting waves:

$$E(z, t) = \frac{1}{2} \{A_1(z)e^{j(\omega_1 t - k_1 z)} + A_2(z)e^{j(\omega_2 t - k_2 z)} + A_3(z)e^{j(\omega_3 t - k_3 z)}\} + \text{c.c.}$$

with:
$$\omega_1 + \omega_2 = \omega_3$$



Forcing term for Maxwell's equation:

$$\frac{\partial^2 P_{\mathrm{NL}}(z, t)}{\partial t^2} = -\varepsilon_0 d_{\mathrm{eff}} \omega_1^2 A_2^*(z) \cdot A_3(z) \cdot \mathrm{e}^{j[\omega_1 t - (k_3 - k_2)z]}$$
$$-\varepsilon_0 d_{\mathrm{eff}} \omega_2^2 A_1^*(z) \cdot A_3(z) \cdot \mathrm{e}^{j[\omega_2 t - (k_3 - k_1)z]}$$
$$-\varepsilon_0 d_{\mathrm{eff}} \omega_3^2 A_1(z) \cdot A_2(z) \cdot \mathrm{e}^{j[\omega_3 t - (k_1 + k_2)z]} + \mathrm{c.c}$$

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \mu_0 \frac{\partial^2 D(z,t)}{\partial t^2} = \mu_0 \frac{\partial^2 P_{\rm NL}(z,t)}{\partial t^2}$$

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Coupled nonlinear equations

Slowly varying envelope approximation :

$$\frac{\partial^2 A}{\partial z^2} \ll 2k \left| \frac{\partial A}{\partial z} \right|$$

$$\begin{cases} \frac{\partial A_1}{\partial z} = -j\sigma_1 A_2^* A_3 \cdot e^{-j\Delta kz}, \\ \frac{\partial A_2}{\partial z} = -j\sigma_2 A_1^* A_3 \cdot e^{-j\Delta kz}, \\ \frac{\partial A_3}{\partial z} = -j\sigma_3 A_1 A_2 \cdot e^{j\Delta kz}, \end{cases}$$

$$\omega_1 + \omega_2 = \omega_3$$

Where:

$$\sigma_i = d_{\rm eff} \, \omega_i / c_0 n_i$$

$$\Delta k = k_3 - k_2 - k_1$$

Phase mismatch

Example 1: Sum frequency generation

• Boundary conditions:

✓ Negligible depletion of
$$A_I(z)$$
: $\frac{\partial A_1(z)}{\partial z} = 0$
✓ No A_3 field: $A_3(0)=0$



$$I_3(z) = \Gamma^2 \frac{\omega_3}{\omega_2} I_1 \cdot I_{20} \cdot \left| \frac{\sin(gz)}{g} \right|^2$$

• Largest efficiency \implies Smallest $g \implies \Delta k = 0$ Phase matching

Example 2: Parametric amplification

• Boundary conditions:

✓ Negligible depletion of
$$A_3(z)$$
: $\frac{\partial A_3(z)}{\partial z} = 0$
✓ No A_2 field: $A_2(0) = 0$



$$I_1(L) = I_{10} \left\{ 1 + \left[\frac{\Gamma}{g} \sinh(gL) \right]^2 \right\} \simeq I_{10} \left(\frac{\Gamma}{g} \right)^2 \frac{e^{2gL}}{4} \qquad I_2(L) = I_{10} \frac{\omega_2}{\omega_1} \left[\frac{\Gamma}{g} \sinh(gL) \right]^2$$

$$\Gamma^2 = \frac{2d_{\rm eff}^2 \omega_1 \omega_2}{c_0^3 \varepsilon_0 n_1 n_2 n_3} I_3 \qquad \qquad g = \sqrt{\Gamma^2 - \frac{\Delta k^2}{4}}$$

• Largest efficiency \longrightarrow Biggest $g \longrightarrow \Delta k = 0$ Phase matching

Signal intensity:

$$I_1(L) = I_{10} \left\{ 1 + \left[\frac{\Gamma}{g} \sinh(gL) \right]^2 \right\} \simeq I_{10} \left(\frac{\Gamma}{g} \right)^2 \frac{e^{2gL}}{4} \qquad g = \sqrt{\Gamma^2 - \frac{\Delta k^2}{4}}$$





Meaning of Phase matching (I)

$$E(z, t) = \frac{1}{2} \{A_1(z)e^{j(\omega_1t-k_1z)} + A_2(z)e^{j(\omega_2t-k_2z)} + A_3(z)e^{j(\omega_3t-k_3z)}\} + \text{c.c.}$$

$$Propagation \text{ velocity: } v_3 = \frac{\omega_3}{k_3}$$

$$\frac{\partial^2 P_{\text{NL}}(z, t)}{\partial t^2} = -\varepsilon_0 d_{\text{eff}} \omega_1^2 A_2^*(z) \cdot A_3(z) \cdot e^{j[\omega_1t-(k_3-k_2)z]}$$

$$-\varepsilon_0 d_{\text{eff}} \omega_2^2 A_1^*(z) \cdot A_3(z) \cdot e^{j[\omega_2t-(k_3-k_1)z]}$$

$$-\varepsilon_0 d_{\text{eff}} \omega_3^2 A_1(z) \cdot A_2(z) \cdot e^{j[\omega_3t-(k_1+k_2)z]} + \text{c.c}$$

$$Propagation \text{ velocity: } v_{P_{\text{NL}}} = \frac{\omega_3}{k_1+k_2}$$

$$Propagation \text{ velocity: } v_{P_{\text{NL}}} = \frac{\omega_3}{k_1+k_2}$$

• $P_{\rm NL}$ efficiently deposits energy into ω_3 when they propagate with the same velocity:

$$v_{P_{\text{NL}}} = v_3 \implies k_3 = k_1 + k_2 \implies \Delta k = k_3 - k_2 - k_1 = 0$$
 Phase matching

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Manley Rowe



the sum of the energies of the three waves is conserved (in a lossless medium)

Intensity $I_i(z)$ corresponds to number of photons $N_i(z)/\Delta t$: $I_i(z) = \frac{N_i(z) \cdot \hbar \omega_i}{S \Delta t} c_0$

$$\underbrace{\mathbf{II}}_{\partial z} = \frac{\partial N_1(z)}{\partial z} = \frac{\partial N_2(z)}{\partial z} = -\frac{\partial N_3(z)}{\partial z}$$

PHOTON CONSERVATION:

when one photon at ω_3 is created, two photons at ω_1 and ω_2 are simultaneously annihilated

Corpuscular view of second order processes

SFG - Sum Frequency Generation

<u>DFG</u> - Difference Frequency Generation



Meaning of Phase matching (II)



Nonlinear interaction as a collision of collinear photons:

$$\omega_3 = \omega_1 + \omega_2 \qquad \hbar \omega_3 = \hbar \omega_2 + \hbar \omega_1 \qquad \text{Energy conservation}$$

$$k_3 = k_1 + k_2 \qquad \hbar k_3 = \hbar k_2 + \hbar k_1 \qquad \text{Momentum conservation}$$

$$\square \text{ Can be also applied to noncollinear interactions:} \qquad \hbar \overline{k_3} = \hbar \overline{k_2} + \hbar \overline{k_1}$$

Extension to non-collinear interactions



Why exponential gain?

 $I_1(L) = I_{10} \left\{ 1 + \left[\frac{\Gamma}{g} \sinh(gL) \right]^2 \right\} \simeq I_{10} \left(\frac{\Gamma}{g} \right)^2 \frac{e^{2gL}}{4}$

Role of the idler beam...



... which gives rise to a positive loop.



Tuning range of an OPA



Energy conservation

 $\hbar\omega_{\rm s} + \hbar\omega_{\rm i} = \hbar\omega_{\rm p}$

✓ Momentum conservation $\hbar \vec{k}_s + \hbar \vec{k}_i = \hbar \vec{k}_p$

The tuning range of an OPA can be evaluated considering:

Energy conservation:

$$\omega_s + \omega_i = \omega_p \rightarrow \boxed{\frac{\omega_s + \omega_i}{2} = \frac{\omega_p}{2}} \rightarrow \text{Signal and idler: symmetric with respect to } \frac{\omega_p}{2}$$

if $\omega_s = \omega_i \rightarrow \omega_s = \omega_i = \frac{\omega_p}{2}$ Degeneracy point

Transmission range of the nonlinear crystal

Example:

 Nonlinear Crystal: BBO: absorbs at λ >3 μm (v <100 THz)
 Laser: Ti:sa (800 nm) or Yb-doped (1030 nm)



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How to get phase matching?

Phase matching requires $k_1 + k_2 = k_3$ equivalent to $\omega_1 n_1 + \omega_2 n_2 = \omega_3 n_3$

In a medium with normal dispersion $(dn/d\omega > 0)$:

 $\omega_1 < \omega_2 < \omega_3$ have refractive index $n_1 < n_2 < n_3$

Phase matching can be written as

no phase matching in isotropic bulk materials

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan (Received July 21, 1961)

The development of pulsed ruby optical masers^{1,2} has made possible the production of monochromatic (6943 A) light beams which, when focussed, exhibit electric fields of the order of 10^5 volts/cm. Table I. The square of the total p perpendicular to the direction of propagation of light through crystalline quartz.



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.

that second harmonic intensities as high as a fraction of a percent of the fundamental could be achieved.

In the experiments we have used a commercially available ruby optical maser⁴ which produces approximately 3 joules of 6943A light in a onemillisecond pulse. This light is passed through us to believe that the order of 10^{11} second harmonic photons were generated within the quartz sample per pulse.

The production of a second harmonic should be observable in isotropic materials such as glass if a strong bias field were applied to the sample. This bias could be oscillatory, thus

Solution: Birefringent crystals



o: ordinary axis n_o, n_{go}, v_{go}

e: extraordinary axis

 n_e, n_{ge}, v_{ge}

• n_e depends on θ :





• $\omega_1 < \omega_2 < \omega_3$ can have different polarizations:

Туре	ω_3 extraordinary	ω_3 ordinary
Type 0	eee	000
Type I	ooe	eeo
Type II	eoe	eoo
	oee	oeo



Finding the phase-matching condition means:

calculating, for a given Type, the angle θ that satisfies $\omega_1 n_1 + \omega_2 n_2 = \omega_3 n_3$

• When 2 fields are extraordinary: θ to be found <u>numerically</u>

Dmitriev V G, Gurzadyan G G, Nikogosyan D N and Lotsch H K V, Optics of nonlinear crystals: Handbook of Nonlinear Optical Crystals, Springer Series in Optical Sciences vol 64 (1999)

Example 1: Phase matching curves of a visible OPA



Example 2: Phase matching curves of an IR OPA



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Broadband gain: general calculation



Broadband gain: general calculation



OPA pumped by Ti:Sapphire laser



C. Manzoni, D. Polli and G. Cerullo, Rev. Sci. Instr. 77, 023103 (2006)

D. Brida et al., J. Opt. A 13, 013001 (2010)

Ti:sa - Generation of UV pulses

The idea: **sum frequency generation** between **broadband** and **narrowband** pulses



R. Borrego Varillas et al., Opt. Lett. 39, 3849-3852 (2014)



R. Borrego-Varillas et al., Appl. Sci. 8, 989 (2018)

OPA pumped by Yb laser (2ω seeded 3ω pumped)



- Tuning from **395-650nm**
- **1.2 µJ** pulse energy in the maximum
 500 nJ over the tuning range
- Sub-20 fs pulses and sub-10 fs Fourier limits

"Sub-20 fs µJ-energy pulses tunable down to the near-UV from a 1 MHz Yb-fiber laser system," M. Bradler and E. Riedle, Opt. Lett. **39**, 2588 (2014).

Courtesy of prof. E. Riedle

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