INTRO TO NONLINEAR OPTICS

presented by

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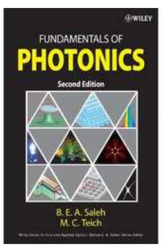
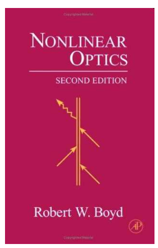
2012 Biophotonics Summer School

University of Illinois at Urbana-Champaign

SOURCE MATERIAL

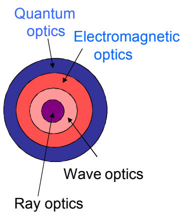




LECTURE OUTLINE

- Background
- Theory
- Second-order nonlinear optics
- Third-order nonlinear optics

OPTICS: THEORIES



Ray optics-Limit of wave optics when wavelength is infinitesimally small

Wave optics-provides a description of optical phenomena using scalar wave theory

Electromagnetic optics- provides most complete treatment of light within classical optics

Quantum optics-provides a quantum mechanical description of the electromagnetic theory

LINEAR OPTICS

- Optical properties of materials (n and α) are independent of light intensity
- Principle of superposition applies
- Frequency of light does not change by passing through a medium
- Two beams of light do not interact in a medium (i.e., light cannot control light)

NONLINEAR OPTICS

- Refractive index depends on optical intensity
- Principle of superposition does not apply in a nonlinear optical medium
- Optical frequency changes by passing through a nonlinear optical medium
- Two beams of light can interact within a nonlinear medium (i.e., light can control light)

SOME CONSEQUENCES OF NONLINEARITY

- Optical frequency conversion – sum-frequency generation, second harmonic generation, third-harmonic generation, frequency-difference generation
- Optical parametric amplification
- Spontaneous parametric downconversion
- Optical Kerr effect
- Self-focusing
- Multiphoton absorption

WHERE IS THE NONLINEARITY?

- In the medium, not in the light
- Medium serves as a mediator for light interaction
- Not observed for free space propagation

WHERE IS THE NONLINEARITY?

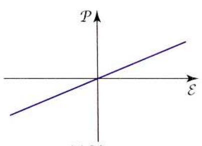
- Polarization density $\mathscr{P} = Np$

Case:

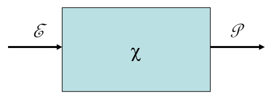
1) Dipole moment *p* could depend nonlinearly on applied electric field

2) Number of atoms (or molecules) that occupy the various energy levels (number density N) depends on the photon-flux density (intensity)

THEORY



(a) Linear



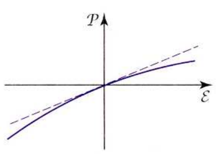
$$\mathcal{P} = \varepsilon_o \chi \mathcal{E}$$

Polarization density

electric susceptibility

Image Source: Fundamentals of Photonics

THEORY



(b) Nonlinear

$$\mathcal{P} = \varepsilon_o \chi^{(1)} \mathcal{E} + \underbrace{\chi^{(2)} \mathcal{E}^2 + \chi^{(3)} \mathcal{E}^3 + \ldots}_{\mathcal{P}_{NL}}$$

- Begins to deviate from linearity when $\mathcal{E}$ becomes large
- Became realizable with invention of ruby laser in 1960
- Still, externally applied e-field is small in comparison to interatomic field (~10^5-10^8 V/m), and thus nonlinearity is weak

Image Source: Fundamentals of Photonics

THEORY

- Recall, that wave equation in a homogeneous and isotropic medium is

$$\nabla^2 \mathcal{E} - \frac{1}{c_o^2} \frac{\partial^2 \mathcal{E}}{\partial t^2} = \mu_o \frac{\partial^2 \mathcal{P}}{\partial t^2}$$

- True, even if medium is dispersive

THEORY: NONLINEAR WAVE EQUATION

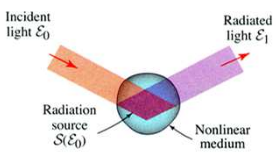
- For nonlinear, homogeneous, and isotropic medium the wave equation is

$$\nabla^2 \mathcal{E} - \frac{1}{c_o^2}\frac{\partial^2 \mathcal{E}}{\partial t^2} = -\mathcal{S}$$

$$\mathcal{S} = -\mu_o \frac{\partial^2 \mathcal{P}_{NL}}{\partial t^2}$$

- $\mathcal{S}$ is a “source” that radiates in a linear medium

THEORY: BORN APPROXIMATION



- Nonlinear wave equation is solved iteratively
- Use 1st Born approximation
- Assumes scattered field is sufficiently weak to consider nonlinearity small
- Incident field determines $\mathcal{P}_{NL}$ (or $\mathcal{S}$)
- $\mathcal{S}$ gives rise to scattered field, which can be determined by adding contribution of each spherical waves

Image Source: Fundamentals of Photonics

2nd-ORDER NONLINEAR OPTICS

- Interaction between 3 waves→ Three-wave mixing

$$\mathcal{P}_{NL} = \chi^{(2)}\mathcal{E}^2 + \underbrace{\chi^{(3)}\mathcal{E}^3 + ...}_{\text{negligible}}$$

- 2 waves of frequencies ω_1 and ω_2 can interact in $\chi^{(2)}$ medium to generate a third wave of frequency ω_3; the three waves then interact in a variety of ways
- Scattering process

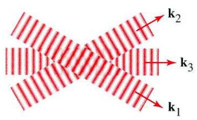
PHASE MATCHING

- Frequency (energy)-matching condition

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

- Phase (momentum)-matching condition

$$\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3$$



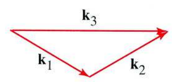
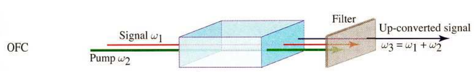


Image Source: Fundamentals of Photonics

SECOND-ORDER NONLINEAR OPTICS

- Optical Frequency Conversion

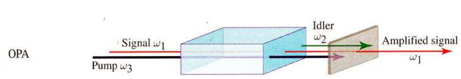


- Waves 1 and 2 are either added to produce a new wave 3, a.k.a., sum-frequency generation (SFG)
- Second-harmonic generation (SHG) is a special case of SFG
- Wave 3 can interact with wave 1 to produce wave 2 (via subtracting), a.k.a., downconversion or frequency-difference generation

Image Source: Fundamentals of Photonics

SECOND-ORDER NONLINEAR OPTICS

- Optical Parametric Amplification

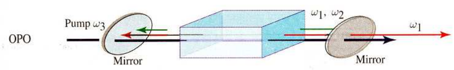


- Waves 1 interacts with wave 3 so that wave 1 is amplified; wave 1 is referred to as signal and wave 3 as the pump
- Wave 2 is auxiliary wave that is created and is called is the idler
- Useful for detection of weak light

Image Source: Fundamentals of Photonics

SECOND-ORDER NONLINEAR OPTICS

- Optical Parametric Oscillator

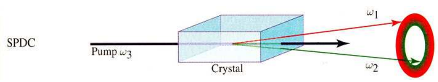


- Coherent oscillator by adding feedback (mirrors) and pump (wave 3)
- Waves 1 and 2 are generated (often only one is coupled out)
- Useful for generating light at wavelengths not available with current laser

Image Source: Fundamentals of Photonics

SECOND-ORDER NONLINEAR OPTICS

- Spontaneous Parametric Downconversion



- Wave 3 is input to NLC
- Waves 1 and 2 are created spontaneously
- Multiple possibilities for generating wave 3 leads to ring of wavelengths/wavevectors
- Useful source of entangled photons

Image Source: Fundamentals of Photonics

SECOND-HARMONIC GENERATION

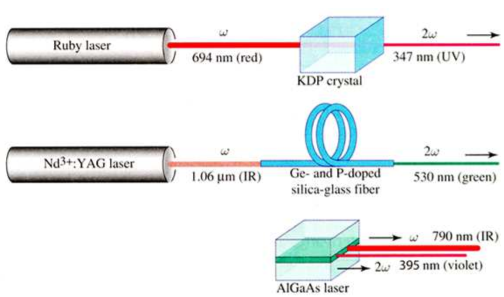


Image Source: Fundamentals of Photonics

SECOND-HARMONIC GENERATION PERFECT PHASE MATCHING

- Frequency (energy)-matching condition

$$\omega_1 + \omega_1 = \omega_3 = 2\omega_1$$

- Phase (momentum)-matching condition

$$\mathbf{k}_1 + \mathbf{k}_1 = \mathbf{k}_3 = 2\mathbf{k}_1$$

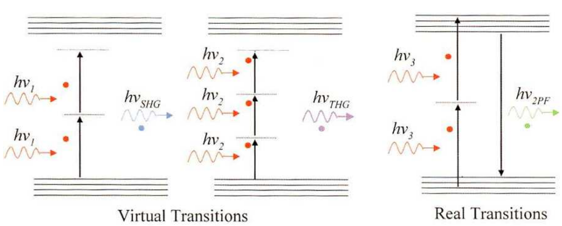
- Perfect phase matching

$$2\mathbf{k}_1 - \mathbf{k}_3 = \Delta\mathbf{k} = 0$$

- Cannot be achieved in dispersive media without introducing birefringence

Image Source: Fundamentals of Photonics

QUANTUM PICTURE



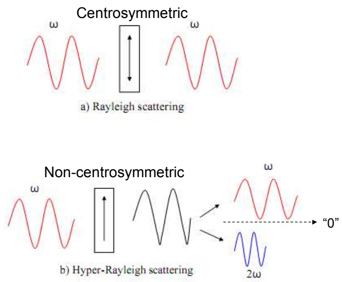
- SHG, THG → "no energy" is absorbed
- 2PF → energy is absorbed

Image Source: Handbook of Biological Confocal Microscopy

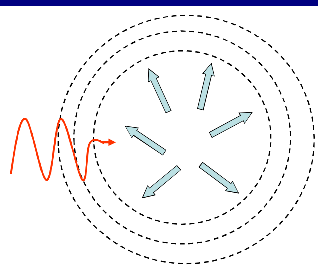
BACKWARD SHG

- Generally, due to phase matching, SHG is generated in the forward direction
- For molecules (scatterers) spaced with an axial period of $\lambda/4$, then up to 25% of SHG is in backward direction
- % in backward direction depends on whether distribution is homogenous/inhomogenous, and whether excitation is focused/unfocused
- Backward SHG can also be generated from multiple scattering of forward SHG (backward SHG is incoherent)

HYPER-RAYLEIGH SCATTERING VS. SECOND-HARMONIC GENERATION



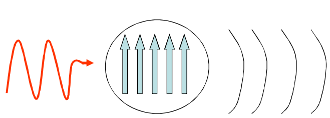
HRS: RANDOMLY-ORIENTED MOLECULES



- HRS emission is incoherent

HRS: ALIGNED MOLECULES

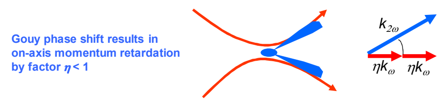
- HRS emission is coherent
- HRS is now called Second-Harmonic Generation (SHG)



- At scales < λ a non-centrosymmetric molecule can emit HRS, but it may not be ordered at the scale of λ to produce SHG

EFFECT OF FOCUSING

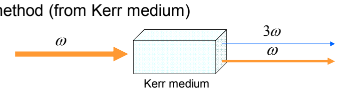
- Typically, high-NA focusing is employed to get high spatial resolution
- Recall, focused Gaussian experiences Gouy phase shift (in comparison to plane wave)
- Gouy phase has effect on directionality of SHG b/c momentum along focal center has decreased (been retarded)
- Results in two off-axis symmetric lobes



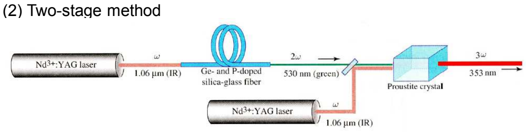
THIRD-ORDER NONLINEAR OPTICS

•Third-Harmonic Generation

(1) Direct method (from Kerr medium)



(2) Two-stage method

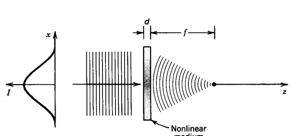


• Can be achieved via SHG followed by SFG of the fundamental and second-harmonic waves

Image Source: Fundamentals of Photonics

THIRD-ORDER NONLINEAR OPTICS

•Self Focusing



• Optical Kerr effect (Δn is proportion to I) results in self-phase modulation, i.e., phase shift experienced by wave depends on its own intensity

• Intense beam impinging on a Kerr medium could be focused as if medium were a graded-index material (imparting nonuniform phase shift)

• Useful for mode locking in pulsed lasers

Image Source: Fundamentals of Photonics

BREAK

Intro to Microscopy

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