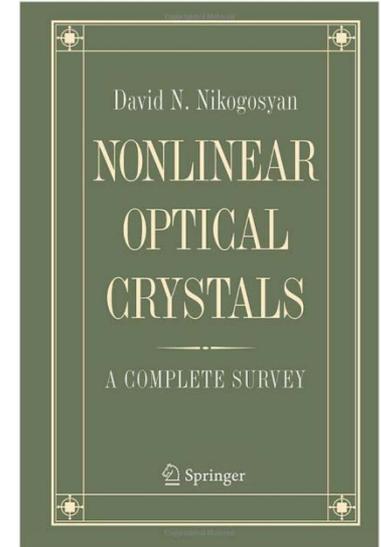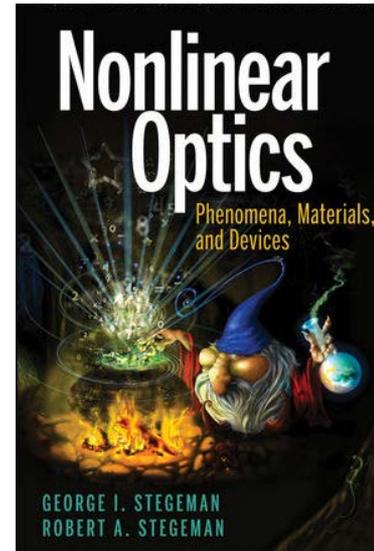Guoqing (Noah) Chang, October 09, 2015

Nonlinear optics: a back-to-basics primer

Lecture 1: linear optics

Suggested references

Robert W. Boyd, *Nonlinear optics* (2008)

Geoffrey New, *Introduction to nonlinear optics* (2011)

George Stegeman and Robert Stegeman, *Nonlinear optics: phenomena, materials, and devices* (2012)

David N. Nikogosyan, *Nonlinear optical crystals: a complete survey* (2005)

Rick Trebino's course slides on optics (http://frog.gatech.edu/lectures.html)

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Contents

If you want to talk to a nonlinear optics person, you need to speak his language; that is, you should understand the jargon in this field.

Basic Nonlinear Optical Crystals

This chapter contains information on the four most widely used nonlinear optical crystals: beta-barium borate (BBO), lithium triborate (LBO), lithium niobate (LN), and potassium titanyl phosphate (KTP). Together with their periodically poled derivatives, periodically poled lithium niobate (PPLN) and periodically poled potassium titanyl phosphate (PPKTP), these materials are employed in at least 75% of all today's practical applications.

All the values of the angular, temperature and spectral acceptances, given in this and in the following chapters, correspond to a 1 cm length of the considered nonlinear crystal.

2.1 β-BaB$_2$O$_4$, Beta-Barium Borate (BBO)

Negative uniaxial crystal: $n_o > n_e$
Molecular mass: 222.950
Specific gravity: 3.84 g/cm^3 [1]; 3.849 g/cm^3 [2]; 3.85 g/cm^3 at $T = 293$ K [3]
Point group: $3m$
Lattice constants:
$a = 12.532$ Å [4]; 12.532 ± 0.001 Å [2]; 12.547 Å [5]
$c = 12.717$ Å [4]; 12.726 ± 0.001 Å [2]; 12.736 Å [5]
Mohs hardness: 4 [6], [7]; 4.5 [2]
Melting point: 1368 K [2], [8]

Linear thermal expansion coefficient α_t [3]

T [K]	$\alpha_t \times 10^6$ [K^{-1}], $\parallel c$	$\alpha_t \times 10^6$ [K^{-1}], $\perp c$
293	0.36	-2.54

Mean value of linear thermal expansion coefficient [5]

T [K]	$\alpha_t \times 10^6$ [K^{-1}], $\parallel c$	$\alpha_t \times 10^6$ [K^{-1}], $\perp c$
298–1173	36	4.0

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David N. Nikogosyan, Nonlinear optical crystals: a complete survey (2005)

Specific heat capacity c_p at $P = 0.101325$ MPa

T [K]	c_p [J/kgK]	Ref.
298	490	[2]
	496	[9]

Thermal conductivity coefficient

κ [W/mK], $\|c$	κ [W/mK], $\perp c$	Ref.
0.8	0.08	[5]
1.6	1.2	[10]

Direct band-gap energy at room temperature: $E_g = 6.2$ eV [11], 6.43 eV [12]
Transparency range:
at 0.5 level: 0.198–2.6 μm for 0.8-cm-long crystal [13]; 0.196–2.2 μm for 0.3-cm-long crystal [2]
at "0" transmittance level: 0.189–3.5 μm [8], [14]
at 0.5 transmittance level: 0.198–2.6 μm [1]

Linear absorption coefficient α

λ [μm]	α [cm^{-1}]	Ref.	Note
0.1934	1.39	[15]	$T = 295$ K
	0.29	[15]	$T = 91$ K
0.213	<0.21	[1]	best crystals
0.264	0.04 ± 0.01	[16]	$\|c$
	0.06 ± 0.003	[16]	$\perp c$, o-wave
	0.10 ± 0.003	[16]	$\perp c$, e-wave
0.2661	<0.17	[1]	best crystals
	0.04–0.15	[2]	
0.5321	0.01	[17]	
	<0.01	[9]	
1.0	0.001–0.002	[2]	
1.0642	<0.001	[9]	
2.09	0.0085	[2]	e-wave
	0.07	[2]	o-wave
2.55	0.5	[18]	

Two-photon absorption coefficient β

λ [μm]	τ_p [ns]	$\beta \times 10^{11}$ [cm/W]	Ref.	Note
0.211	0.0009	243 ± 85	[19]	$\theta = 30°, \phi = 0°$
0.264	0.0008	93 ± 33	[19]	$\theta = 30°, \phi = 0°$
	0.00022	68 ± 6	[20]	$\|c$

λ [μm]	τ_p [ns]	$\beta \times 10^{11}$ [cm/W]	Ref.	Note
		66 ± 7	[20]	$\perp c$, o-wave
		47 ± 5	[20]	$\perp c$, e-wave
	0.0002	61	[21]	$\theta = 48°$
0.2661	0.015	90 ± 10	[11]	$\|c$
0.3547	0.017	1.0 ± 0.2	[11]	$\|c$

Experimental values of refractive indices [5]

λ [μm]	n_o	n_e
0.40466	1.69267	1.56796
0.43583	1.68679	1.56376
0.46782	1.68198	1.56024
0.47999	1.68044	1.55914
0.50858	1.67722	1.55691
0.54607	1.67376	1.55465
0.57907	1.67131	1.55298
0.58930	1.67049	1.55247
0.64385	1.66736	1.55012
0.81890	1.66066	1.54589
0.85212	1.65969	1.54542
0.89435	1.65862	1.54469
1.01400	1.65608	1.54333

Temperature derivative of refractive indices for temperature range 293–353 K [5]

λ [μm]	$dn_o/dT \times 10^6$ [K^{-1}]	$dn_e/dT \times 10^6$ [K^{-1}]
0.4–1.0	−16.6	−9.3

Best set of dispersion relations (λ in μm, $T = 293$ K) [13]:

$$n_o^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\,\lambda^2$$

$$n_e^2 = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\,\lambda^2$$

Sellmeier equations with better accuracy near infrared absorption edge (λ in μm, $T = 293$ K) [22]:

$$n_o^2 = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01471\,\lambda^2 + 0.0006081\,\lambda^4 - 0.00006740\,\lambda^6$$

$$n_e^2 = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01627\,\lambda^2 + 0.0005716\,\lambda^4 - 0.00006305\,\lambda^6$$

Other sets of Sellmeier equations are given in [1], [5], [8], [23], [24], [25], [26].

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David N. Nikogosyan, Nonlinear optical crystals: a complete survey (2005)

Nonlinear refractive index γ

λ [μm]	$\gamma \times 10^{15}$ [cm^2/W]	Ref.	Note
0.2661	0.025 ± 0.008	[11]	$\parallel c$
0.3547	0.36 ± 0.08	[11]	$\parallel c$
0.5321	0.55 ± 0.10	[11]	$\parallel c$
0.780	0.40 ± 0.05	[27]	[100] direction
	0.32 ± 0.05	[27]	[010] direction
0.850	0.37 ± 0.06	[28]	$\theta = 29.2°, \phi = 0°$
1.0642	0.29 ± 0.05	[11]	$\parallel c$

Linear electrooptic coefficients measured at low frequencies (well below the acoustic resonances of BBO crystal, i.e., for the "free" crystal) at room temperature

λ [μm]	r_{22}^T [pm/V]	r_{51}^T [pm/V]	Ref.	Note
0.5145	2.5 ± 0.1		[29]	$T = 296$ K
0.6328	2.5	<0.04	[30]	
	2.2 ± 0.1		[31]	

Linear electrooptic coefficient measured at high frequencies (well above the acoustic resonances of BBO crystal, i.e., for the "clamped" crystal)

λ [μm]	r_{22}^S [pm/V]	Ref.	Note
0.5145	2.1 ± 0.3	[29]	$T = 296$ K
0.6328	2.1 ± 0.1	[31]	

Expressions for the effective second-order nonlinear coefficient in general case (Kleinman symmetry) conditions are valid, $d_{31} = d_{15}$) [32]:

$$d_{ooe} = d_{31} \sin(\theta + \rho) - d_{22} \cos(\theta + \rho) \sin 3\phi$$

$$d_{eoe} = d_{oee} = d_{22} \cos^2(\theta + \rho) \cos 3\phi$$

Simplified expressions for the effective second-order nonlinear coefficient (approximation of small birefringence angle, Kleinman symmetry conditions are valid, $d_{31} = d_{15}$) [33]:

$$d_{ooe} = d_{31} \sin \theta - d_{22} \cos \theta \sin 3\phi$$

$$d_{eoe} = d_{oee} = d_{22} \cos^2 \theta \cos 3\phi$$

Absolute values of second-order nonlinear coefficients [32]:

$$|d_{22}(0.532\,\mu m)| = 2.6\,pm/V$$

$$|d_{22}(0.852\,\mu m)| = 2.3\,pm/V$$

$$|d_{22}(1.064\,\mu m)| = 2.2\,pm/V$$

$$|d_{22}(1.313\,\mu m)| = 1.9\,pm/V$$

$$|d_{15}(1.064\,\mu m)| = 0.03\,pm/V$$

Experimental values of internal angular, temperature, and spectral bandwidths at $T = 293$ K

Interacting wavelengths [μm]	θ_{pm} [deg]	$\Delta\theta^{int}$ [deg]	ΔT [°C]	$\Delta\nu$ [cm^{-1}]	Ref.
SHG, $o + o \Rightarrow e$					
$1.0642 \Rightarrow 0.5321$	22.8	0.021	37	9.7	[4]
	21.9	0.028			[23]
	22.7	0.030	51		[5]
$0.5321 \Rightarrow 0.26605$	47.3	0.010	4		[5]
$0.53 \Rightarrow 0.265$	47.6 (298 K)	0.006			[80]
SFG, $o + o \Rightarrow e$					
$1.0641 + 0.53205 \Rightarrow 0.3547$	31.3	0.011			[77]
$1.0642 + 0.5321 \Rightarrow 0.35473$	31.1	0.015	16		[5]
$2.44702 + 0.5712 \Rightarrow 0.4631$	22.1	0.026			[25]
$2.68823 + 0.5712 \Rightarrow 0.4711$	21.8	0.028			[25]
SHG, $e + o \Rightarrow e$					
$1.0642 \Rightarrow 0.5321$	32.7	0.034		8.8	[4]
	32.4	0.046	37		[5]
SFG, $e + o \Rightarrow e$					
$1.0642 + 0.5321 \Rightarrow 0.35473$	38.4	0.020	13		[5]
SFG, $o + e \Rightarrow e$					
$1.0642 + 0.5321 \Rightarrow 0.35473$	58.4	0.050	12		[5]

Temperature variation of phase-matching angle at $T = 293$ K [5]

Interacting wavelengths [μm]	θ_{pm} [deg]	$d\theta_{pm}/dT$ [deg/K]
SHG, $o + o \Rightarrow e$		
$0.5321 \Rightarrow 0.26605$	47.3	0.00250
$1.0642 \Rightarrow 0.5321$	22.7	0.00057
SFG, $o + o \Rightarrow e$		
$1.0642 + 0.5321 \Rightarrow 0.35473$	31.1	0.00099
SHG, $e + o \Rightarrow e$		
$1.0642 \Rightarrow 0.5321$	32.4	0.00120
SFG, $e + o \Rightarrow e$		
$1.0642 + 0.5321 \Rightarrow 0.35473$	38.4	0.00150
SFG, $o + e \Rightarrow e$		
$1.0642 + 0.5321 \Rightarrow 0.35473$	58.4	0.00421

Calculated values of inverse group-velocity mismatch for SHG process in BBO

Interacting wavelengths [μm]	θ_{pm} [deg]	β [fs/mm]
SHG, $o + o \Rightarrow e$		
$1.2 \Rightarrow 0.6$	21.18	54
$1.1 \Rightarrow 0.55$	22.28	76
$1.0 \Rightarrow 0.5$	23.85	104
$0.9 \Rightarrow 0.45$	26.07	141

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David N. Nikogosyan, Nonlinear optical crystals: a complete survey (2005)

A timeline of classical (<u>linear</u>) optics

Willibrord Snell (1591-1626), Snell's law

Johannes Kepler (1571-1630), Total internal reflection

Pierre de Fermat (1601-1665), Fermat principle

Christiaan Huygens (1629-1695), Wave theory of optics

Isaac Newton (1642-1727), Particle theory of light

Thomas Young (1773-1829), Interference of optical waves

Augustin Fresnel (1788-1827), Fresnel coefficients

James Clerk Maxwell (1831-1879), Maxwell's equations

Operators used in Maxwell's Equations

The "Del" operator:
$$\nabla \equiv \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z} \right)$$

The "Gradient" of a scalar function:
$$\nabla f \equiv \left(\frac{\partial f}{\partial x}, \frac{\partial f}{\partial y}, \frac{\partial f}{\partial z} \right)$$

The "Divergence" of a vector function:
$$\nabla \cdot G \equiv \frac{\partial G_x}{\partial x} + \frac{\partial G_y}{\partial x} + \frac{\partial G_z}{\partial x}$$

The "Laplacian" operator:
$$\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2}$$

Operators used in Maxwell's Equations

The "Laplacian" of a scalar function:

$$\nabla^2 f \equiv \nabla \cdot \nabla f = \nabla \cdot (\frac{\partial f}{\partial x}, \frac{\partial f}{\partial y}, \frac{\partial f}{\partial z})$$

$$= \frac{\partial^2 f}{\partial x^2} + \frac{\partial^2 f}{\partial y^2} + \frac{\partial^2 f}{\partial z^2}$$

The "Laplacian" of a vector function is the same, but for each component:

$$\nabla^2 G \equiv (\frac{\partial^2 G_x}{\partial x^2} + \frac{\partial^2 G_x}{\partial y^2} + \frac{\partial^2 G_x}{\partial z^2}, \frac{\partial^2 G_y}{\partial x^2} + \frac{\partial^2 G_y}{\partial y^2} + \frac{\partial^2 G_y}{\partial z^2}, \frac{\partial^2 G_z}{\partial x^2} + \frac{\partial^2 G_z}{\partial y^2} + \frac{\partial^2 G_z}{\partial z^2})$$

Operators used in Maxwell's Equations

The "Curl" of a vector function:

$$\nabla \times G \equiv (\frac{\partial G_z}{\partial y} - \frac{\partial G_y}{\partial z}, \frac{\partial G_x}{\partial z} - \frac{\partial G_z}{\partial x}, \frac{\partial G_y}{\partial x} - \frac{\partial G_x}{\partial y})$$

The curl can be computed from a matrix determinant:

$$\nabla \times G = \det \begin{bmatrix} \hat{x} & \hat{y} & \hat{z} \\ \dfrac{\partial}{\partial x} & \dfrac{\partial}{\partial y} & \dfrac{\partial}{\partial z} \\ G_x & G_y & G_z \end{bmatrix}$$

Maxwell's Equations of differential form in a medium without free current and free charge

Ampere's law:

$$\nabla \times H = \frac{\partial D}{\partial t}$$

Faraday's law:

$$\nabla \times E = -\frac{\partial B}{\partial t}$$

Gauss's law:

$$\nabla \cdot D = 0$$

Gauss's law of magnetism:

$$\nabla \cdot B = 0$$

Constitutive relations for a nonmagnetic material:

$$D = \varepsilon_0 E + \boxed{P} \longrightarrow$$ **Polarization. It takes into light-matter interaction.**

$$B = \mu_0 H$$

ε_0 **Electric permittivity in vacuum**

μ_0 **Magnetic permeability in vacuum**

Derivation of wave equation

Vector Identity:
$$\nabla \times (\nabla \times E) = \nabla(\nabla \cdot E) - \nabla^2 E$$

$$\nabla \times (\nabla \times E) = -\nabla \times \frac{\partial B}{\partial t} = -\frac{\partial}{\partial t}(\nabla \times B)$$

$$= -\frac{\partial}{\partial t}(\nabla \times (\mu_0 H)) = -\frac{\partial}{\partial t}(\mu_0 \nabla \times H)$$

$$= -\frac{\partial}{\partial t}(\mu_0(\varepsilon_0 \frac{\partial E}{\partial t} + \frac{\partial P}{\partial t}))$$

$$\nabla^2 E - \nabla(\nabla \cdot E) = \mu_0(\varepsilon_0 \frac{\partial^2 E}{\partial t^2} + \frac{\partial^2 P}{\partial t^2})$$

Derivation of wave equation

$$\nabla^2 E - \nabla(\nabla \cdot E) = \mu_0(\varepsilon_0 \frac{\partial^2 E}{\partial t^2} + \frac{\partial^2 P}{\partial t^2})$$

In the linear optics of isotropic source-free media:

$$\nabla \cdot D = 0 \quad \longrightarrow \quad \nabla \cdot E = 0$$

In the nonlinear optics, normally we have: $\quad \nabla(\nabla \cdot E) << \nabla^2 E$

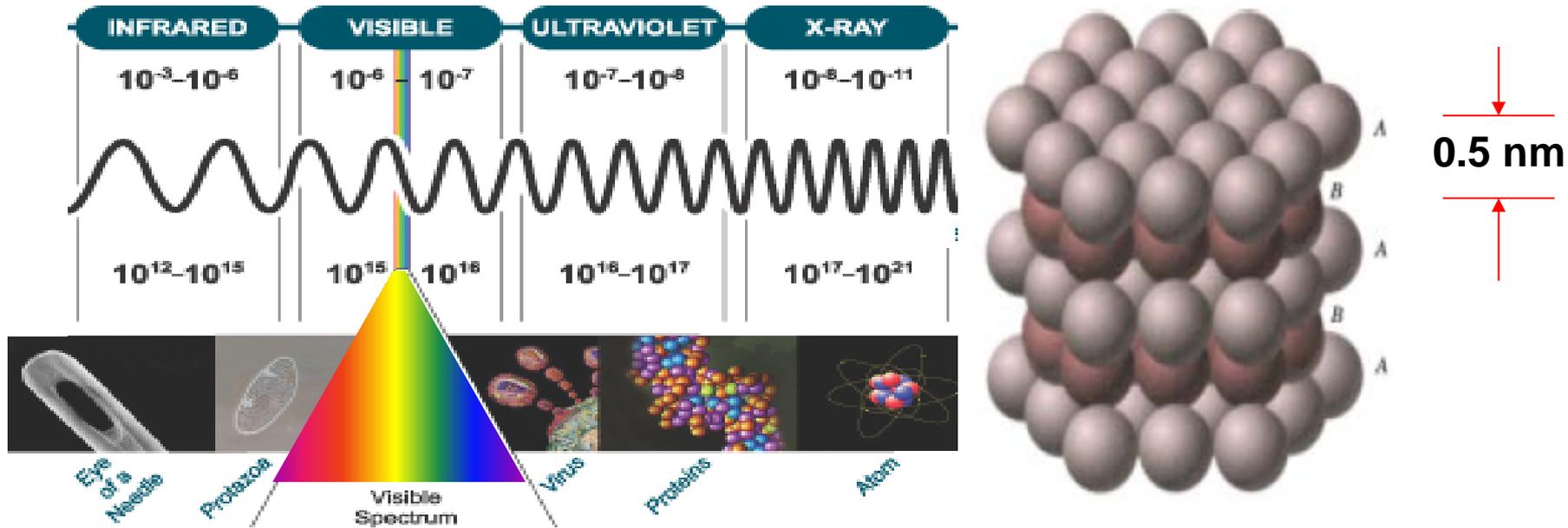**Vacuum speed of light:**

$$c_0 = \sqrt{\frac{1}{\mu_0 \epsilon_0}}.$$

Simplified wave equation:

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

Wave in vacuum   **Source term**

Interaction between EM waves and materials

INFRARED	VISIBLE	ULTRAVIOLET	X-RAY
10^{-3}–10^{-6}	10^{-6} – 10^{-7}	10^{-7}–10^{-8}	10^{-8}–10^{-11}
10^{12}–10^{15}	10^{15} – 10^{16}	10^{16}–10^{17}	10^{17}–10^{21}

Visible Spectrum

Eye of a Needle   Protozoa   Virus   Proteins   Atom

0.5 nm

Wavelength of green light is about 500 nm. So the optical wave experiences an effective homogeneous medium, which is characterized by

Electric permittivity ε **and** **Magnetic permeability** μ

For a nonmagnetic material

$$P = \varepsilon_0 \chi E \quad \varepsilon = \varepsilon_0 (1 + \chi) \quad \mu = \mu_0$$

χ **is the electric susceptibility. The velocity of light is different from the vacuum speed by a factor called the refractive index**

$$n = \sqrt{(\varepsilon \mu)/(\varepsilon_0 \mu_0)} = \sqrt{1 + \chi}$$

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Lorentz model of light-atom interaction

Important assumptions

The atomic core is

-- positively charged
-- static (heavy, fixed within the crystal lattice)
-- with the center of charge at x = 0.

The electrons are

-- light weight
-- elastically bound by a massless spring with spring constant with equilibrium position at x = 0
-- carrying out a damped movement; that is, after removing the force, the movement decreases and finally ends.
-- the electron and atomic core form an oscillator with a resonant frequency w_0.

H. A. Lorentz (1853-1928)

Juergen Popp et al., Handbook of biophotonics (2012).
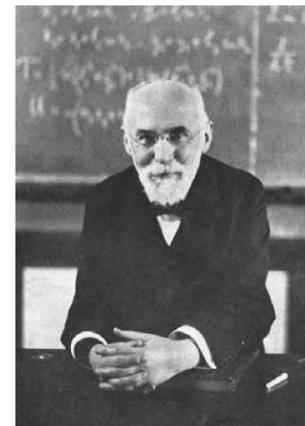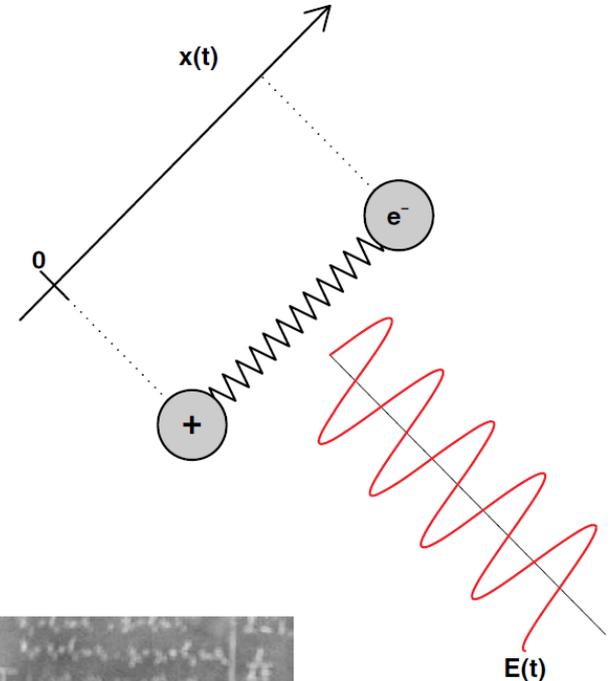
Lorentz model: forced electron harmonic oscillator

Dipole moment is defined as the product of magnitude of charges and the distance of separation between the charges.

Without an applied field, the centers of the negative and the positive charges coincide. The dipole moment is zero.

If a **field constant in time** is applied, the electrons are displaced relative to their position in the absence of an external field. The centers of the positive and negative charges no longer coincide and a static dipole moment is induced.
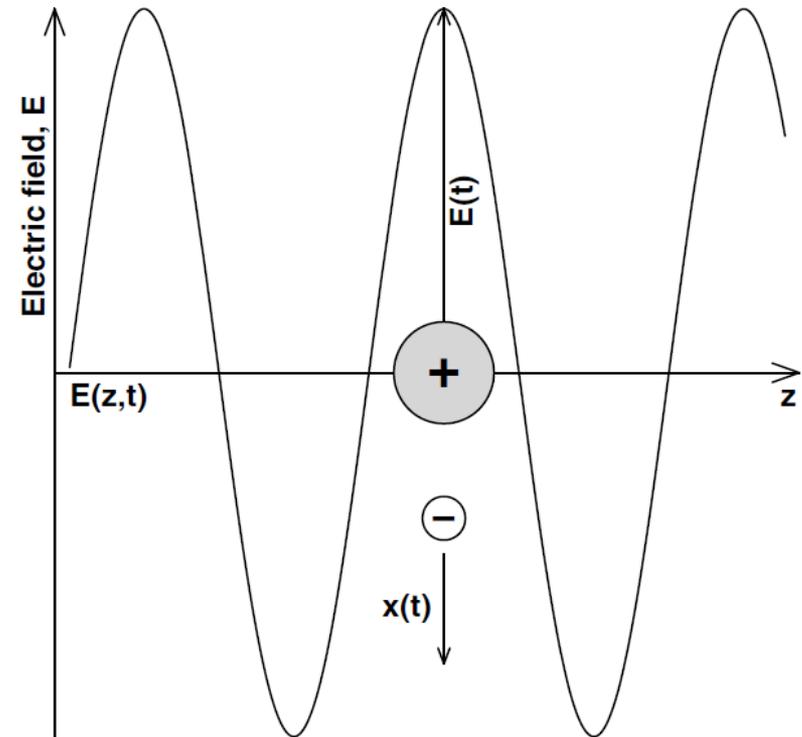
If a **time-dependent electric field** interacts with the atom, then the electron starts to oscillate around its equilibrium position with the same frequency of the electric field. Such an oscillating dipole moment will emit a new electromagnetic wave at the same frequency as well.

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Juergen Popp et al., Handbook of biophotonics (2012).

Lorentz model of light-atom interaction

When light of frequency *w* excites an atom with resonant frequency w_0:

Electric field at atom   Electron   Emitted field

$$\vec{\mathscr{E}}(t) \qquad \vec{x}_e(t) \qquad \vec{\mathscr{E}}'(t)$$

On resonance $(\omega = \omega_0)$

Incident light

+

Emitted light

=
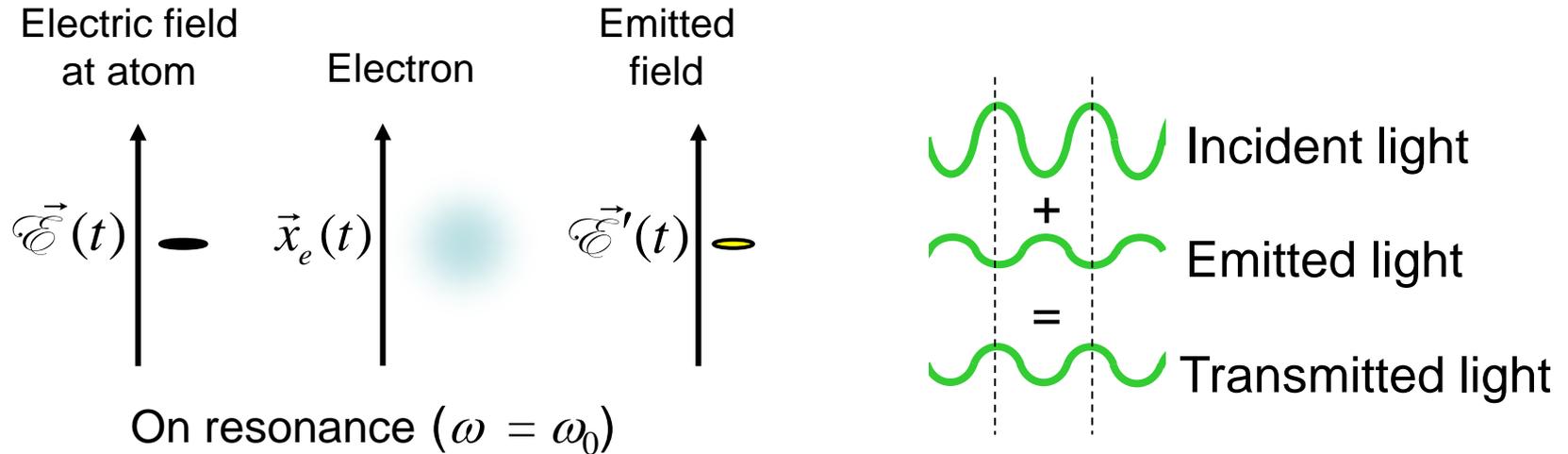
Transmitted light

Incident Light excites electron oscillation → electron oscillation emits new light at the same frequency → incident light interferes with the new light leading to the transmitted light.

The crucial issue is the **relative phase** of the incident light and this emitted light. For example, if these two waves are ~180° out of phase, the beam will be attenuated. We call this absorption.

Adapted from Rick Trebino's course slides

Forced oscillator and resonance

When we apply a periodic force to a natural oscillator (such as a pendulum, spring, swing, or atom), the result is a **forced oscillator**.

Examples:

Child on a swing being pushed

Periodically pushed pendulum

Bridge in wind or an earthquake

Electron in a light wave

Nucleus in a light wave

Tacoma Narrows Bridge oscillating and collapsing because oscillatory winds blew at its resonance frequency. (collapsed under 64 km/h wind conditions the morning of November 7, 1940)

The forced oscillator is one of the most important problems in physics. It is the concept of **resonance**.

Adapted from Rick Trebino's course slides

One more example: child on a swing

LOW FREQUENCY   AT RESONANCE   HIGH FREQUENCY

If you give the swing a push it will swing back and forward. If you just give it one push it will swing back and forth a few times and then come to rest. (That's because of *friction* and *damping*.) To keep the swing moving you have to push again each time the swing reaches the closest point to you. You have to match the *frequency* of the swing to make it swing high.

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The forced oscillator

The amplitude and relative phase of the oscillator motion with respect to the input force depend on the frequencies.

Let the oscillator's resonant frequency be ω_0, and the forcing frequency be ω.

Let the forcing function be a light electric field and the oscillator a (positively charged) nucleus in a molecule.

Electric field at nucleus   Nucleus

Below resonance

$\omega \ll \omega_0$

Weak vibration. **In phase**.

On resonance

$\omega = \omega_0$

Strong vibration. **90° out of phase.**

Above resonance

$\omega \gg \omega_0$

Weak vibration. **180° out of phase.**

Adapted from Rick Trebino's course slides

The forced oscillator

The amplitude and relative phase of the oscillator motion with respect to the input force depend on the frequencies.

The electron charge is **negative**, so there's a **180°** **phase shift** in all cases (compared to the previous slide's plots).

Below
resonance

$\omega \ll \omega_0$

Weak vibration. **180° out of phase.**

On
resonance

$\omega = \omega_0$

Strong vibration. **-90° out of phase**.

Above
resonance

$\omega \gg \omega_0$

Weak vibration. **In phase.**

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Adapted from Rick Trebino's course slides

The forced oscillator

Electric field at atom   Electron   Emitted field

The amplitude and relative phase of the oscillator motion with respect to the input force depend on the frequencies.
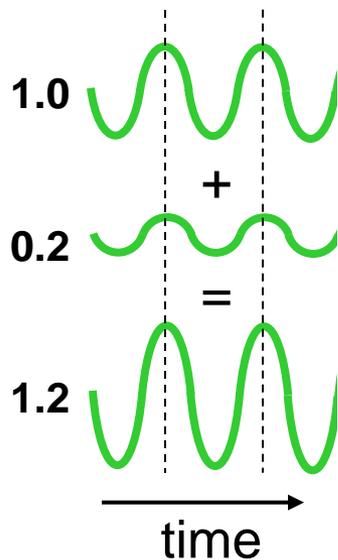
Maxwell's Equations will yield emitted light that's **90° phase-shifted** with respect to the atom's motion.

Below resonance

$$\omega \ll \omega_0$$

Weak emission. **90° out of phase.**

On resonance

$$\omega = \omega_0$$

Strong emission. **180° out of phase.**

Above resonance

$$\omega \gg \omega_0$$

Weak emission. **-90° out of phase.**
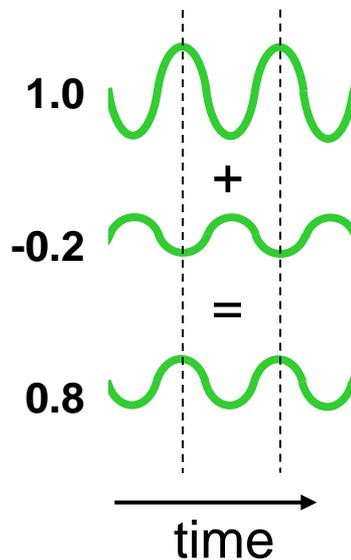
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Interference depends on relative phase

When two waves add together with the same complex exponentials, we add the complex amplitudes, $\underset{\sim}{E}_0 + \underset{\sim}{E}_0{}'$.

Constructive interference:

Destructive interference:

Quadrature phase: $\pm 90°$ interference:

1.0

+

0.2

=

1.2

time

1.0

+

-0.2

=

0.8

time

1.0

+

-0.2i

=

1-0.2i

time

Laser

Absorption
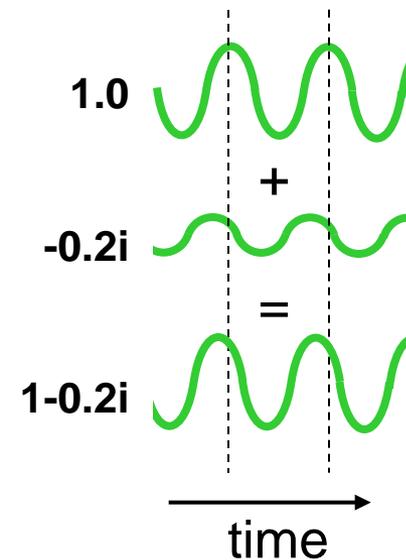
Slower phase velocity (when accumulated over distance)

Adapted from Rick Trebino's course slides

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Dielectric Permittivity: Lorentz model

Density (# of atoms per unit volume)

Dipole moment

$$P(t) = \frac{\text{Dipole moment}}{\text{Volume}} = N \cdot p(t) = \varepsilon_0 \chi E(t)$$

Elementary Dipole

$$\chi = \frac{N \cdot p(t)}{\varepsilon_0 E(t)}$$

Lorentz Model:

Pos.
Charge

Neg.
Charge
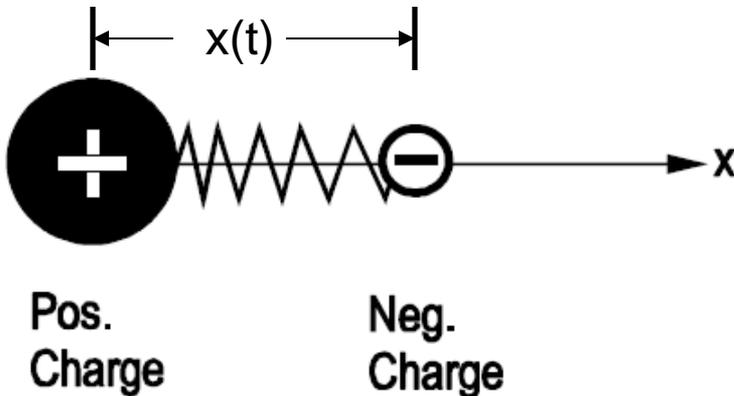
$$p(t) = -ex(t)$$

$x(t)$ is much smaller than the wavelength of electric field. Therefore we can neglect the spatial variation of the E field during the motion of the charge.

$$\vec{E}(\vec{r}, t) = \vec{E}(\vec{r}_A, t) = E(t)\vec{e}_x$$

Response to a monochromatic field: forced electron harmonic oscillator

$$m\frac{d^2x}{dt^2} + 2m\gamma\frac{dx}{dt} + m\omega_0^2 x = -eE(t) \longleftarrow \text{force}$$

mass

damping   frequency of undamped oscillator

electron charge

$$E(t) = \mathrm{E}\, e^{j\omega t} \longrightarrow x(t) = x e^{j\omega t} \longrightarrow p(t) = -ex(t) = \mathrm{p}\, e^{j\omega t}$$

$$\mathrm{p} = \frac{e^2/m}{\omega_0^2 - \omega^2 + 2j\omega\gamma}\mathrm{E}$$

$$\chi(\omega) = \frac{Ne^2/(m\varepsilon_0)}{\omega_0^2 - \omega^2 + 2j\omega\gamma}$$

Sellmeier equation to model refractive index

If the frequency is far away from the absorption resonance $\quad \left| \omega_0^2 - \omega^2 \right| >> 2\omega\gamma$

$$\chi(\omega) = \frac{\omega_p^2}{(\omega_0^2 - \omega^2)} \qquad \omega_p^2 = Ne^2/(m\varepsilon_0)$$

Normally there are multiple resonant frequencies for the electronic oscillators. It means in general the refractive index will have the form

$$n^2(\omega) = 1 + \chi(\omega) = 1 + \sum_i A_i \frac{\omega_i^2}{\omega_0^2 - \omega^2} = 1 + \sum_i a_i \frac{\lambda^2}{\lambda^2 - \lambda_i^2}$$

	Fused Quartz	Sapphire
a_1	0.6961663	1.023798
a_2	0.4079426	1.058364
a_3	0.8974794	5.280792
λ_1^2	$4.679148 \cdot 10^{-3}$	$3.77588 \cdot 10^{-3}$
λ_2^2	$1.3512063 \cdot 10^{-2}$	$1.22544 \cdot 10^{-2}$
λ_3^2	$0.9793400 \cdot 10^2$	$3.213616 \cdot 10^2$

Susceptibility is a tensor

$$P = \varepsilon_0 \chi E$$

$$D = \varepsilon_0 E + P$$

$$\bigg\}\ D = \varepsilon_0 \varepsilon E \qquad \varepsilon = 1 + \chi$$

Dielectric constant

Polarization and electric field are vectors:

$$P = \begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} \qquad E = \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$$

Therefore in general, the susceptibility is a 2nd order tensor (i.e., 3 by 3 matrix):

$$\begin{bmatrix} P_x \\ P_y \\ P_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \chi_{xx} & \chi_{xy} & \chi_{xz} \\ \chi_{yx} & \chi_{yy} & \chi_{yz} \\ \chi_{zx} & \chi_{zy} & \chi_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$$

A more convenient notation:

$$P_i = \varepsilon_0 \chi_{ij} E_j$$

χ_{ij} is the linear susceptibility tensor. Repeated indices imply summation.

$$D_i = \varepsilon_0 \varepsilon_{ij} E_j = \varepsilon_0 (\delta_{ij} + \chi_{ij}) E_j$$

δ_{ij} is the identity matrix.

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Optical anisotropy: birefringence

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$$

We can always select a (x,y,z) axes (i.e., principal dielectric axes) to diagonalize the dielectric matrix to the following form:

$$\begin{bmatrix} D_x \\ D_y \\ D_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix} = \varepsilon_0 \begin{bmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{bmatrix} \begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$$

$n_x = n_y = n_z$ $\longrightarrow$ Isotropic ("the same in all directions") medium (no birefringence)

$n_x = n_y = n_o$ $\quad n_z = n_e \neq n_o$ $\quad$ Uniaxial medium $\begin{cases} n_e > n_o & \text{Positive uniaxial} \\ n_e < n_o & \text{Negative uniaxial} \end{cases}$

$n_x \neq n_y \neq n_z$ $\longrightarrow$ $\quad$ biaxial medium

Crystal symmetry

A **crystal** or crystalline solid is a solid material whose constituents, such as atoms, molecules or ions, are arranged in a highly ordered microscopic structure, forming a **crystal** lattice that extends in all directions.
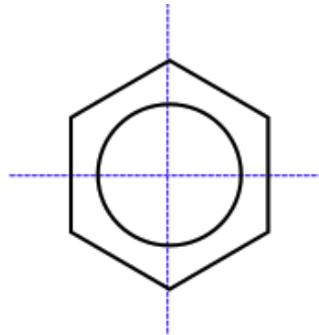
—— *From Wikipedia*

Every crystal belongs to one of 32 point symmetry classes, which are categorized into 7 crystal systems.

If an object is invariant under point reflection through its center, it is said to possess center symmetry or inversion symmetry.
The object is centrosymmeric.
Otherwise it is non-centrosymmetric.

System/Class No.	Symmetry code	Inversion sym.	Examples
Biaxial crystals			
Triclinic system			
1	1	no	
2	$\bar{1}$	yes	Copper sulphate
Monoclinic system			
3	2	no	
4	m	no	
5	$2/m$	yes	
Orthorhombic system			
6	$2\,2\,2$	no	
7	$m\,m\,2$	no	LBO, KTP, KTA
8	$2/m\ 2/m\ 2/m$	yes	
Uniaxial crystals			
Tetragonal system			
9	$\bar{4}$	no	
10	4	no	
11	$\bar{4}\,2\ m$	no	KDP, ADP, CDA
12	$4\,2\,2$	no	Nickel sulphate
13	$4/m$	yes	
14	$4\ m\ m$	no	
15	$4/m\ 2/m\ 2/m$	yes	
Trigonal system			
16	3	no	Sodium periodate
17	$\bar{3}$	yes	
18	$3\,2$	no	α-quartz
19	$3\ m$	no	BBO, Lithium niobate
20	$\bar{3}\ 2/m$	yes	Calcite
Hexagonal system			
21	$\bar{6}$	no	
22	$\bar{6}\,2\ m$	no	Gallium selenide
23	6	no	Lithium iodate
24	$6\,2\,2$	no	β-quartz
25	$6/m$	yes	
26	$6\ m\ m$	no	Cadmium selenide
27	$6/m\ 2/m\ 2/m$	yes	
Optically isotropic crystals			
Cubic system			
28	$2\,3$	no	Sodium chlorate
29	$4\,3\,2$	no	
30	$3m = 2/m\ \bar{3}$	yes	Pyrite
31	$\bar{4}\,3\ m$	no	Gallium arsenide, zinc blende
32	$4/m\ \bar{3}\ 2/m = m3m$	yes	Sodium chloride, diamond

Maxwell's Equations of differential form in a medium without free current and free charge

Ampere's law:

$$\nabla \times H = \frac{\partial D}{\partial t}$$

Faraday's law:

$$\nabla \times E = -\frac{\partial B}{\partial t}$$

Gauss's law:

$$\nabla \cdot D = 0$$

Gauss's law of magnetism:

$$\nabla \cdot B = 0$$

Constitutive relations for a nonmagnetic material:

$$D = \varepsilon_0 E + P \longrightarrow \text{ Polarization}$$

$$B = \mu_0 H$$

ε_0 **Electric permittivity in vacuum**

μ_0 **Magnetic permeability in vacuum**

Light propagation in anisotropic media

Consider plane EM waves of angular frequency ω propagating in the direction of the unit vector $\hat{s}$, that is

$$D = D_0 \exp[j(\omega t - k \cdot r)] \qquad B = B_0 \exp[j(\omega t - k \cdot r)]$$
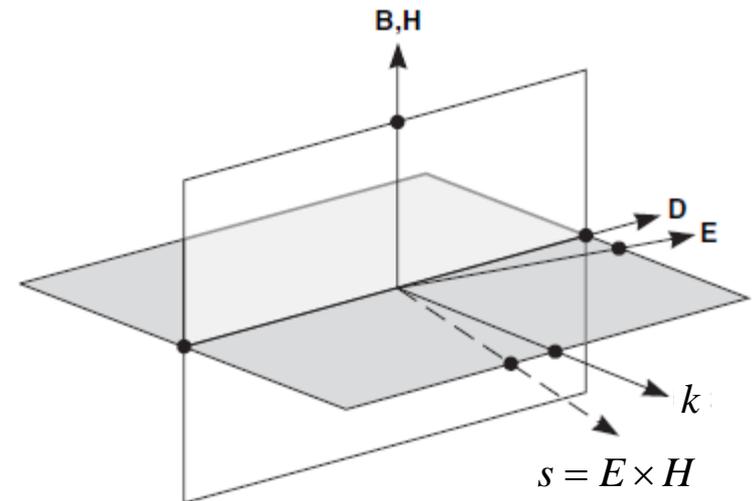
Gauss's law: $\quad \nabla \cdot D = 0 \quad \longrightarrow \quad k \cdot D = 0$

Gauss's law of magnetism: $\quad \nabla \cdot B = 0 \quad \longrightarrow \quad k \cdot B = 0$

$$k \perp D \perp B$$

They form a orthogonal basis.

Ampere's law: $\quad \nabla \times H = \dfrac{\partial D}{\partial t} \quad \longrightarrow \quad H \perp D$

Faraday's law: $\quad \nabla \times E = -\dfrac{\partial B}{\partial t} \quad \longrightarrow \quad E \perp B$

Constitutive relations for a nonmagnetic materi

$$B = \mu_0 H \quad \longrightarrow \quad H /\!/ B$$

$$D = \varepsilon_0 E + P = \varepsilon_0 \varepsilon E \quad \longrightarrow \quad D \not{/\!/} E \quad \text{in an anisotropic medium.}$$

Poynting vector $E \times H$ is NOT in the same direction as k.

$s = E \times H$

Light propagation in a uniaxial crystal

$$\begin{bmatrix} \varepsilon_{xx} & 0 & 0 \\ 0 & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix} = \begin{bmatrix} n_x^2 & 0 & 0 \\ 0 & n_y^2 & 0 \\ 0 & 0 & n_z^2 \end{bmatrix}$$

$$n_x = n_y = n_o \qquad n_z = n_e$$

$$\frac{x^2}{n_o^2} + \frac{y^2}{n_o^2} + \frac{z^2}{n_e^2} = 1$$

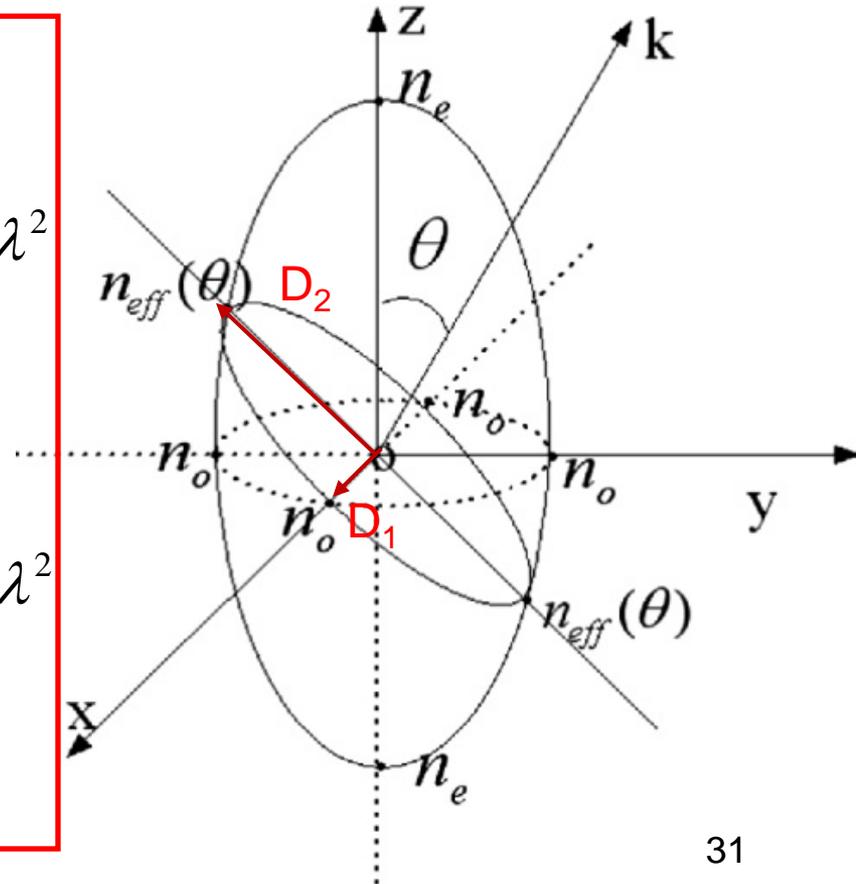Index ellipsoid for uniaxial crystal

Take BBO as an example

$$n_o^2(\lambda) = 2.7359 + \frac{0.01878}{\lambda^2 - 0.01822} - 0.01354\lambda^2$$

$$\frac{dn_o}{dT} = -9.3 \times 10^{-6} / {}^\circ C$$

$$n_e^2(\lambda) = 2.3753 + \frac{0.01224}{\lambda^2 - 0.01667} - 0.01516\lambda^2$$

$$\frac{dn_e}{dT} = -16.6 \times 10^{-6} / {}^\circ C$$

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Light propagation in a uniaxial crystal

(1) Index ellipsoid is used to find the two refractive indices and the two corresponding directions of D associated with the two independent plane waves that can propagate along k direction.

(2) The plane through the origin and perpendicular to k intersects with the index ellipsoid and generates an ellipse.

(3) D_1 lies in the x-y plane and is perpendicular to the optical axis z. D_2 lies in the plane of z-k. D_1 is called ordinary wave and D_2 extraordinary wave.

(4) The two axes of the intersection ellipse are $2n_o$ and $2n_{eff}(\theta)$

(5) n_o is the refractive index for D_1. $n_{eff}(\theta)$ is the refractive index for D_2.
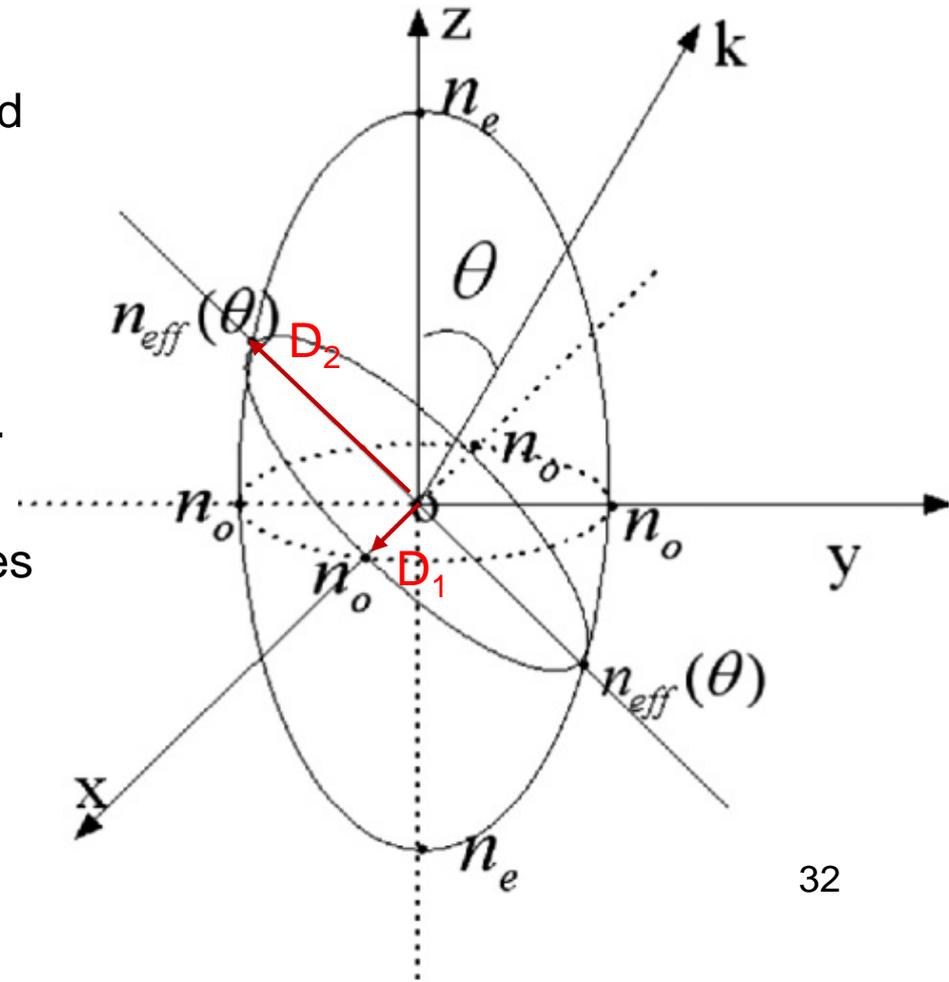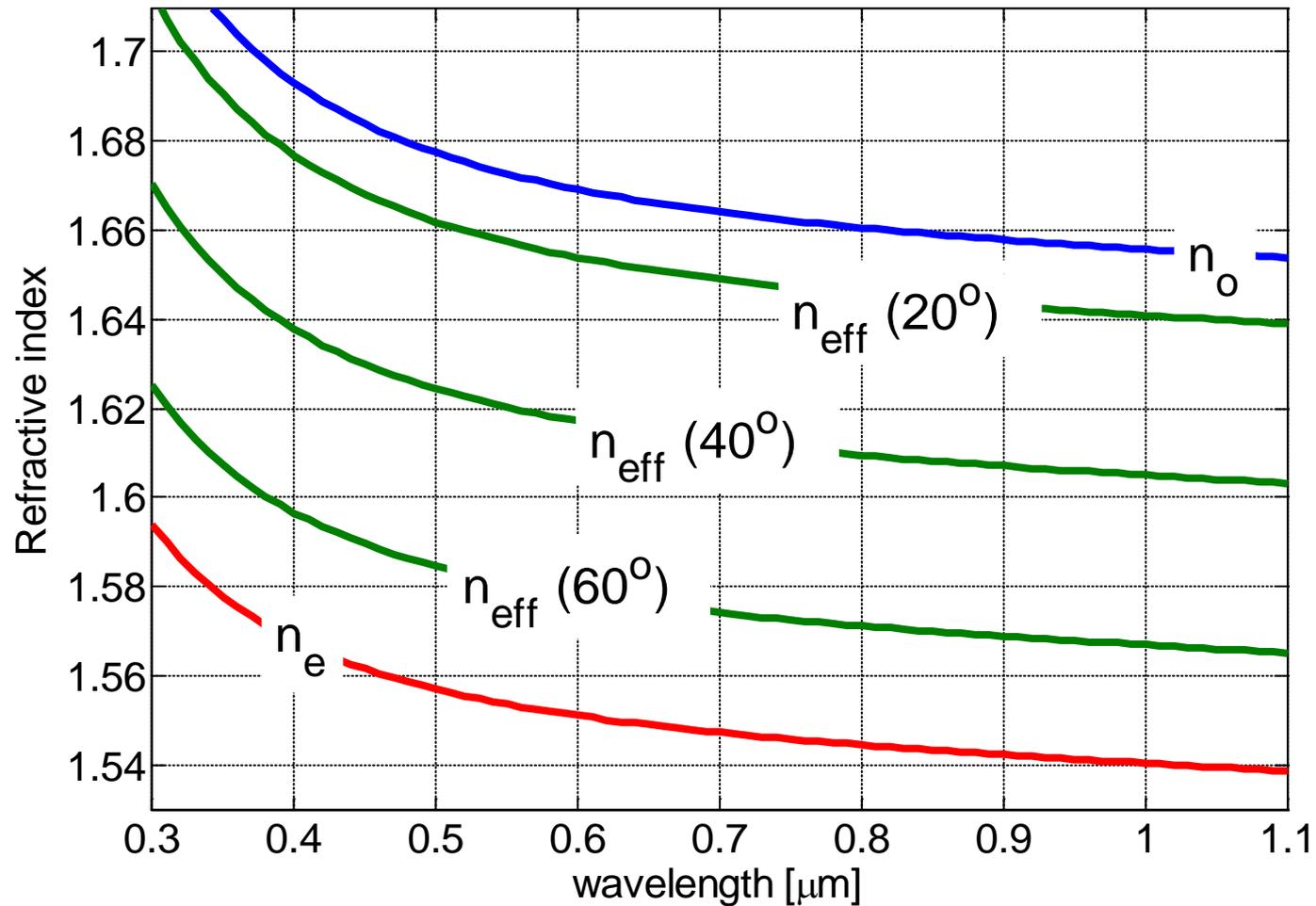
$$\frac{1}{n_{eff}^2(\theta)} = \frac{\cos^2(\theta)}{n_o^2} + \frac{\sin^2(\theta)}{n_e^2}$$

Two special cases:

$$n_e(0^0) = n_o$$
$$n_e(90^0) = n_e$$

Example: BBO at room temperature

Plot of refractive index vs wavelength [μm], showing curves labeled n_o (blue), $n_{eff}(20^o)$, $n_{eff}(40^o)$, $n_{eff}(60^o)$ (green), and n_e (red). X-axis: wavelength [μm] from 0.3 to 1.1. Y-axis: Refractive index from 1.54 to 1.7.

BBO is a negative uniaxial nonlinear crystal.

More on ordinary wave and extraordinary wave

Z (optic axis)

D, E

k, s

θ

$n(\theta) = n_o$

Index surfaces for a positive uniaxial medium.

Z (optic axis)

k

E

D

s

θ

ρ

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

$$\rho(\theta) = \pm \{\tan^{-1}[(\frac{n_o}{n_e})^2 \tan \theta] - \theta\}$$
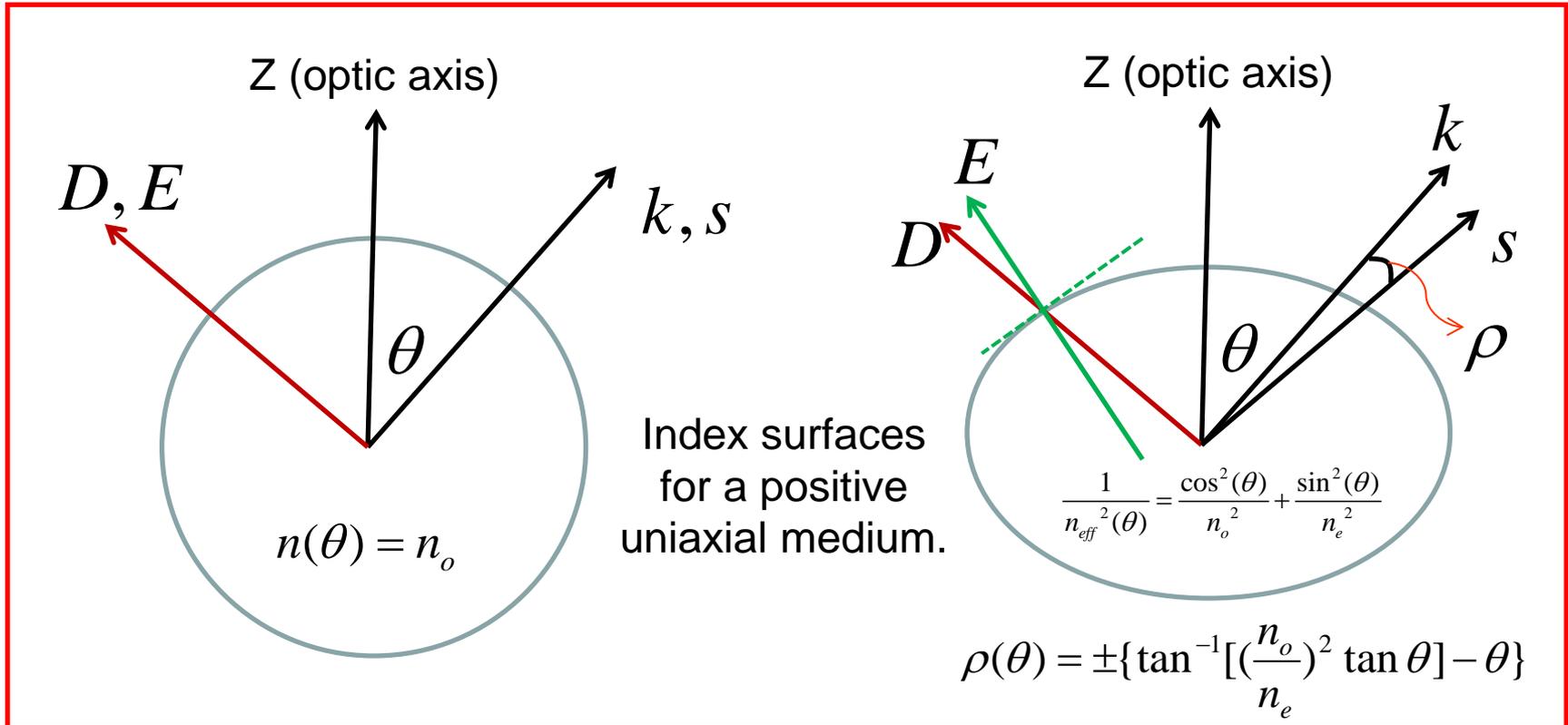
$s = E \times H$ Represents the energy flow.

For o-wave, D and E are in parallel, and k and s are in the same direction.
For e-wave, D and E are NOT in parallel, and the energy flows at the direction different from k.

Calcite Crystal Birefringence
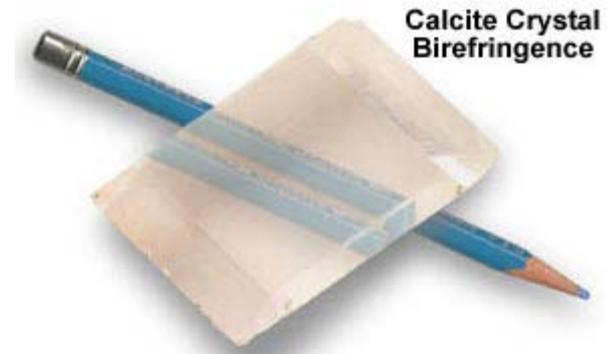
Take-home message

- Constitutive relations describe light-matter interaction.

- Material polarization can be modeled by harmonic electric oscillation.

- In an anisotropic medium, linear susceptibility is a 2nd rank tensor.

- Ordinary wave (o wave) and extraordinary wave (e wave) experience different refractive indices. The refractive index of e wave depends on the incident angle.

- E wave and o wave carry energies, which may flow at different direction causing double refraction.

Suggested reading

Classical harmonic oscillator model

-- Mark Fox, *optical properties of solids*, chapter 2

-- George Stegemann and Robert Stegemann, *Nonlinear optics*, chapter 1

EM wave propagation in anisotropic media

-- Amnon Yariv and Pochi Yeh, *optical waves in crystals*, chapter 1 and 4

-- Geoffrey New, *Introduction to nonlinear optics*, chapter 3