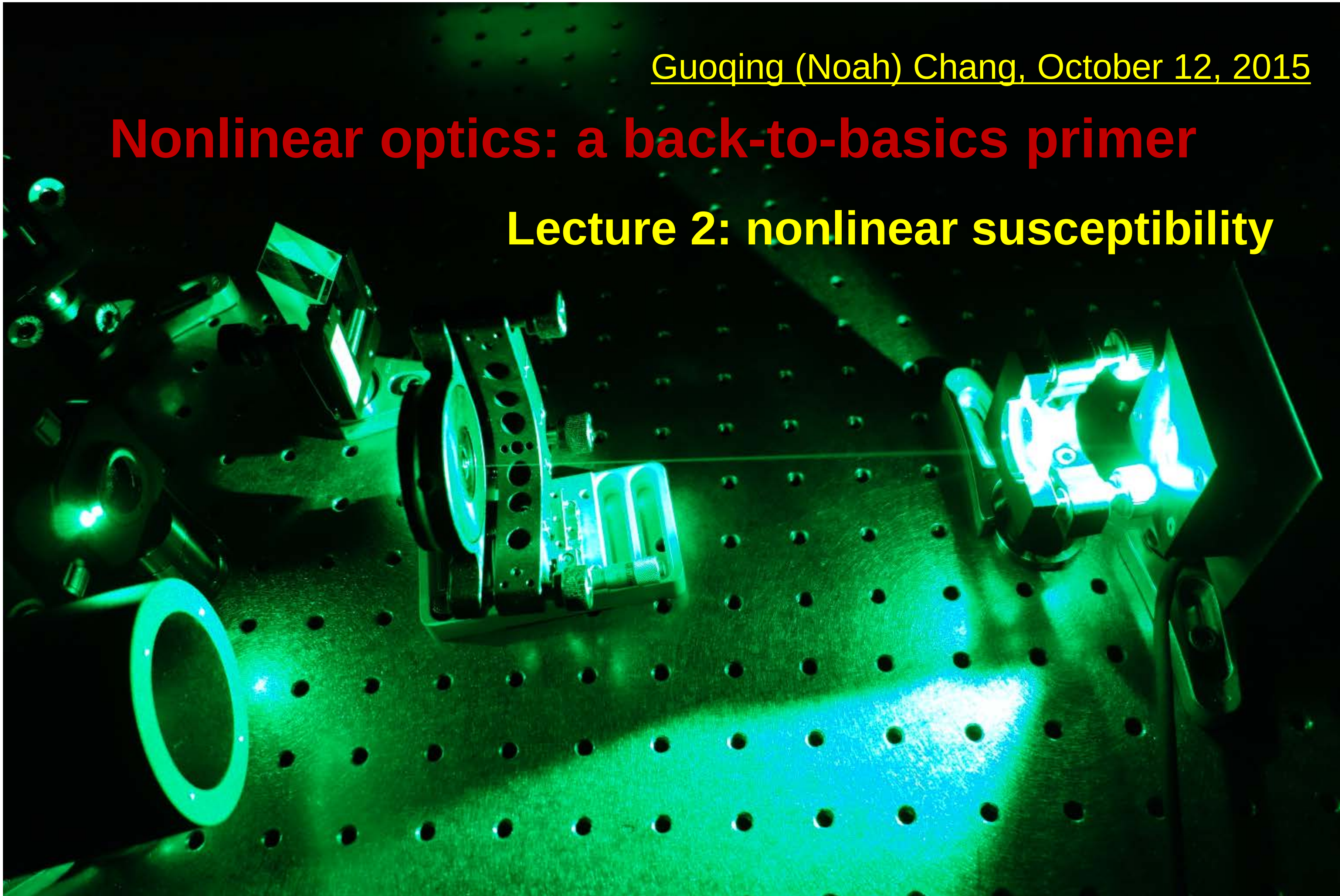
Guoqing (Noah) Chang, October 12, 2015

Nonlinear optics: a back-to-basics primer

Lecture 2: nonlinear susceptibility



Interaction between EM waves and materials

Light wave perturbs material $\longrightarrow$ $P = \varepsilon_0 \chi E$

Perturbed material alters the light wave $\longrightarrow$ $(\nabla^2 - \frac{1}{c_0^2}\frac{\partial^2}{\partial t^2})E = \mu_0 \frac{\partial^2 P}{\partial t^2}$

Examples of changes to light wave:

- Frequency
- Amplitude and phase
- Polarization state
- Direction of propagation
- Transverse profile

Response to a monochromatic field: forced electron harmonic oscillator

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

force

mass

damping

frequency of undamped oscillator

electron charge

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

$$\mathrm{x} = \frac{-e/m}{\omega_0^2 - \omega^2 + 2j\omega\gamma}\mathrm{E} \qquad P = Nex = \frac{Ne^2/m}{\omega_0^2 - \omega^2 + 2j\omega\gamma}E$$

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

Note on complex notation

We live in the "real" world; that is, a real world signal has components of positive frequency and negative frequency.

$$E(t) = 2\mathrm{E}\cos\omega t = \mathrm{E}e^{j\omega t} + \mathrm{E}^* e^{-j\omega t} = \mathrm{E}e^{j\omega t} + c.c.$$

Normally it is safe in calculation to only keep the complex, positive-frequency component. Of course, you may also calculate the susceptibility for the negative-frequency:

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

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

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

In linear optics, susceptibility is independent of the input light field

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

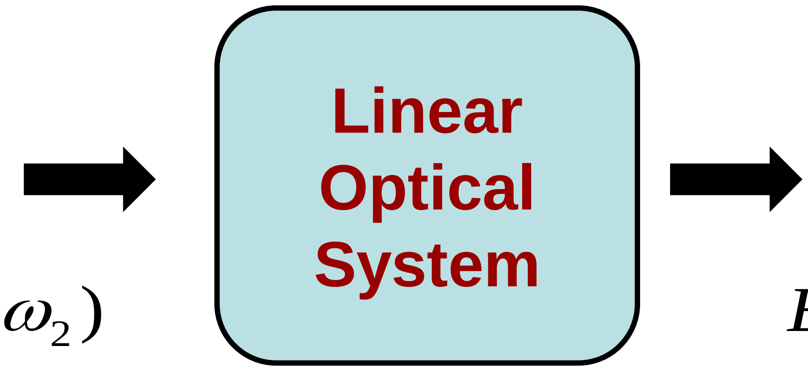
$$P(\omega) = \varepsilon_0 \chi(\omega) E(\omega)$$

Linear optical system

$E_{in}(\omega_1)$

$E_{in}(\omega_2)$

$E_{in}(\omega_1) + E_{in}(\omega_2)$



$E_{out}(\omega_1)$

$E_{out}(\omega_2)$

$E_{out}(\omega_1) + E_{out}(\omega_2)$

Potential energy function

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

Potential energy function for this harmonic oscillator is

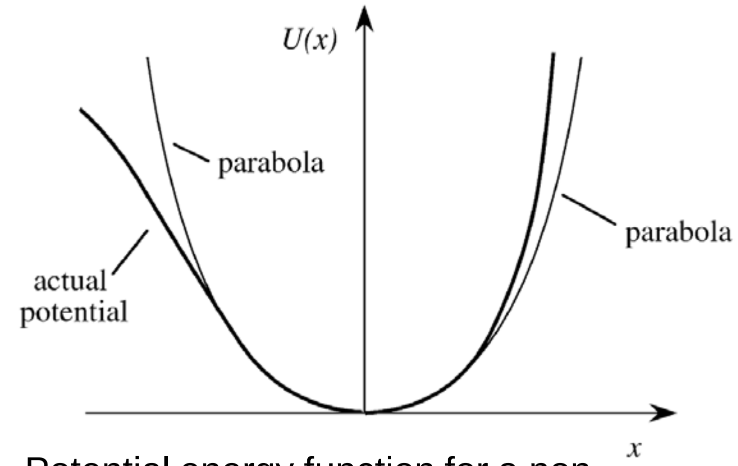
$$U(x)=-\int m\omega_0^2xdx=-\frac{1}{2}m\omega_0^2x^2$$

This is a good parabola approximation when the amplitude of E-field is weak. As E-field becomes large enough, the electron oscillation amplitude proportionally increases to the level that higher-order correction term needs to include:

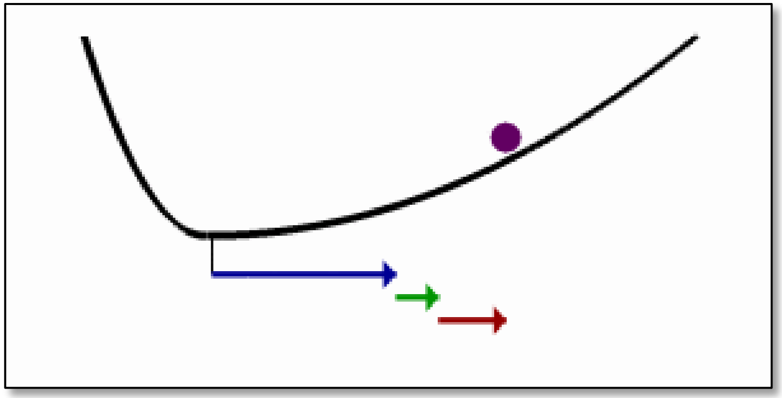
$$m\frac{d^2x}{dt^2}+2m\gamma\frac{dx}{dt}+m\omega_0^2x+\underline{m\eta x^2}=-eE(t)$$

Higher order correction

This equation describes an anharmonic electron oscillator. That is, the oscillation response to a sinusoidal wave is NOT a sinusoidal wave anymore.



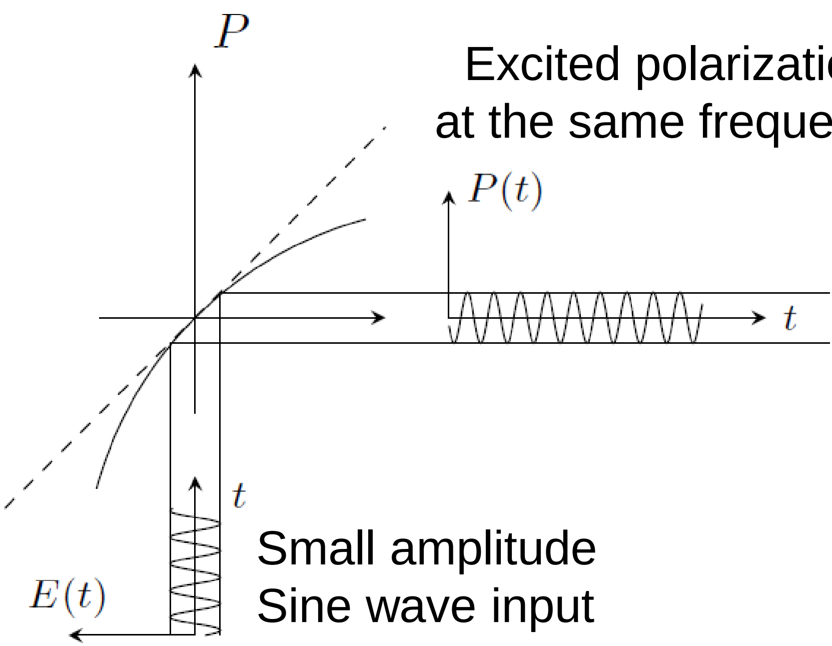
Potential energy function for a non-centrosymmetric medium. (Adapted from *Nonlinear optics, Boyd*)



Color code—blue: linear response; green: frequency doubled; red: DC

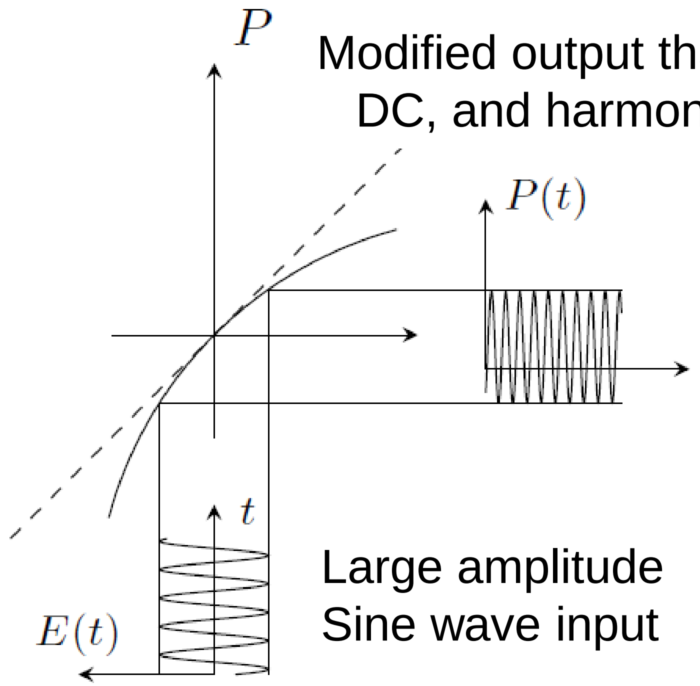
http://physics.stackexchange.com/questions/127531/lack-of-inversion-symmetry-in-crystal

Linear interaction is an approximation for weak field



Excited polarization at the same frequency

Small amplitude Sine wave input



Modified output that contains DC, and harmonic waves

Large amplitude Sine wave input

In general, P is a nonlinear function of E

$$P = \varepsilon_0 [\chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \chi^{(4)} E^4 + \ldots]$$

Linear susceptibility

2nd order susceptibility

3rd order susceptibility

4th order susceptibility

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

New frequencies are generated due to nonlinear light-matter interaction.

Response to an intense field: forced electron anharmonic oscillator

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

Nonlinear response

For $\eta x << \omega_0^2$ we can use perturbation theory to solve the equation by expressing the solution in the form of a power series expansion in the strength of η

$$x(t)=x^{(1)}(t)+\eta x^{(2)}(t)+\eta^2x^{(3)}(t)+\ldots$$

Plugging $x(t)$ into the oscillator equation, we require that the terms proportional to η^0 η^1 η^2 each satisfy the equation separately.

$$\eta^0: \quad \frac{d^2x^{(1)}}{dt^2}+2\gamma\frac{dx^{(1)}}{dt}+\omega_0^2x^{(1)}=\frac{-eE(t)}{m}$$

$$\eta^1: \quad \frac{d^2x^{(2)}}{dt^2}+2\gamma\frac{dx^{(2)}}{dt}+\omega_0^2x^{(2)}=-\eta[x^{(1)}]^2$$

$$\eta^2: \quad \frac{d^2x^{(3)}}{dt^2}+2\gamma\frac{dx^{(3)}}{dt}+\omega_0^2x^{(3)}=-2\eta x^{(1)}x^{(2)}$$

Perturbation theory

$$\frac{d^2x^{(1)}}{dt^2} + 2\gamma\frac{dx^{(1)}}{dt} + \omega_0^2 x^{(1)} = \frac{-eE(t)}{m} \qquad E(t) = \mathrm{E}_1 e^{j\omega_1 t} + \mathrm{E}_2 e^{j\omega_2 t} + c.c.$$

$$x^{(1)}(t) = x^{(1)}(\omega_1)e^{j\omega_1 t} + x^{(1)}(\omega_2)e^{j\omega_2 t} + c.c.$$

$$x^{(1)}(\omega_i) = \frac{-e/m}{\omega_0^2 - \omega_1^2 + 2j\omega_i\gamma}\mathrm{E}_1 = \frac{-e/m}{D(\omega_i)}\mathrm{E}_1$$

$$\boxed{\chi^{(1)}(\omega_i) = \frac{N(e^2/m)}{\varepsilon_0 D(\omega_i)}}$$

$$D(\omega_i) = \omega_0^2 - \omega_i^2 + 2j\omega_i\gamma \qquad i = 1,2$$

$$\frac{d^2x^{(2)}}{dt^2} + 2\gamma\frac{dx^{(2)}}{dt} + \omega_0^2 x^{(2)} = -\eta[x^{(1)}]^2 \qquad x^{(1)}(t) = x^{(1)}(\omega_1)e^{j\omega_1 t} + x^{(1)}(\omega_2)e^{j\omega_2 t} + c.c.$$

$-\eta[x^{(1)}]^2$ contains the frequencies $\pm 2\omega_1$, $\pm 2\omega_2$, $\pm(\omega_1 + \omega_2)$, $\pm(\omega_1 - \omega_2)$, and 0.

Take frequency $(\omega_1 + \omega_2)$ for example: $x^{(2)}(t) = x^{(2)}(\omega_1 + \omega_2)e^{j(\omega_1+\omega_2)t}$

$$x^{(2)}(\omega_1 + \omega_2) = \frac{-2\eta(e/m)^2\mathrm{E}_1\mathrm{E}_2}{D(\omega_1 + \omega_2)D(\omega_1)D(\omega_2)}$$

Perturbation theory

Follow the similar procedure, we get the amplitudes of the response at the other frequencies:

$$x^{(2)}(2\omega_1) = \frac{-\eta(e/m)^2 \mathrm{E}_1^2}{D(2\omega_1)D^2(\omega_1)} \qquad x^{(2)}(2\omega_2) = \frac{-\eta(e/m)^2 \mathrm{E}_2^2}{D(2\omega_2)D^2(\omega_2)}$$

$$x^{(2)}(\omega_1+\omega_2) = \frac{-2\eta(e/m)^2 \mathrm{E}_1 \mathrm{E}_2}{D(\omega_1+\omega_2)D(\omega_1)D(\omega_2)}$$

$$x^{(2)}(\omega_1-\omega_2) = \frac{-2\eta(e/m)^2 \mathrm{E}_1 \mathrm{E}_2^*}{D(\omega_1-\omega_2)D(\omega_1)D(-\omega_2)}$$

$$x^{(2)}(0) = \frac{-2\eta(e/m)^2 \mathrm{E}_1 \mathrm{E}_1^*}{D(0)D(\omega_1)D(-\omega_1)} + \frac{-2\eta(e/m)^2 \mathrm{E}_2 \mathrm{E}_2^*}{D(0)D(\omega_2)D(-\omega_2)}$$

Second-order susceptibility

$$P^{(2)}(\omega_1+\omega_2) = -Nex^{(2)}(\omega_1+\omega_2)$$

$$P^{(2)}(\omega_1+\omega_2) = F\varepsilon_0\chi^{(2)}(\omega_1+\omega_2)E_1(\omega_1)E_2(\omega_2)$$

Degeneracy factor:

$$F=1 \quad \omega_1=\omega_2$$

$$F=2 \quad \omega_1\neq\omega_2$$

Sum-frequency generation (SFG)

$$\chi^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2) = \frac{N(e^3/m^2)\eta}{\varepsilon_0 D(\omega_1+\omega_2)D(\omega_1)D(\omega_2)}$$

$$= \frac{\varepsilon_0^2 m\eta}{N^2e^3}\chi^{(1)}(\omega_1+\omega_2)\chi^{(1)}(\omega_1)\chi^{(1)}(\omega_2)$$

Follow the similar procedure, we get the 2nd-order susceptibility at the other frequencies :

Difference-frequency generation (DFG)

$$\chi^{(2)}(\omega_1-\omega_2,\omega_1,-\omega_2) = \frac{\varepsilon_0^2 m\eta}{N^2e^3}\chi^{(1)}(\omega_1-\omega_2)\chi^{(1)}(\omega_1)\chi^{(1)}(-\omega_2)$$

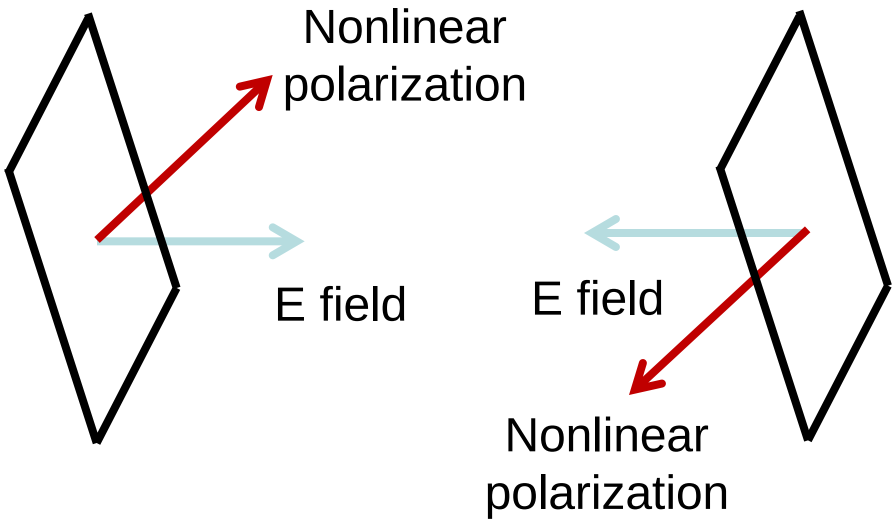
Second-harmonic generation (DFG)

$$\chi^{(2)}(2\omega_1,\omega_1,\omega_1) = \frac{\varepsilon_0^2 m\eta}{N^2e^3}\chi^{(1)}(2\omega_1)[\chi^{(1)}(\omega_1)]^2$$

Optical rectification (OR)

$$\chi^{(2)}(0,\omega_1,-\omega_1) = \frac{\varepsilon_0^2 m\eta}{N^2e^3}\chi^{(1)}(0)\chi^{(1)}(\omega_1)\chi^{(1)}(-\omega_1)$$

Even-order nonlinear effects vanish for centrosymmetric optical crystals



$$P = \varepsilon_0[\chi^{(1)}E + \chi^{(2)}E^2 + \chi^{(3)}E^3 + \chi^{(4)}E^4 + \ldots]$$

For centrosymmetric optical crystals, if we replace *E* by –*E*, according to the symmetry, *P* should become –*P*. This means

$$\chi^{(2N)}(-E)^{2N} = -\chi^{(2N)}E^{2N} \rightarrow \chi^{(2N)} = 0$$

For example, glass is centrosymmetric and therefore the lowest-order nonlinearity arises from the third-order nonlinear susceptibility.

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

Linear susceptibility is a matrix for optically anisotropic media

$$P^{(1)} = \varepsilon_0 \chi^{(1)} E \quad \left\{ \begin{array}{l} P_x^{(1)} = \varepsilon_0 \chi^{(1)} E_x \\ P_y^{(1)} = \varepsilon_0 \chi^{(1)} E_y \\ P_z^{(1)} = \varepsilon_0 \chi^{(1)} E_z \end{array} \right. \quad \Rightarrow$$

Only true for optically isotropic media

For optically anisotropic media, linear susceptibility is a 3X3 matrix (a 2nd-rank tensor):

$$\left. \begin{array}{l} P_x^{(1)} = \varepsilon_0 [\chi_{xx}^{(1)} E_x + \chi_{xy}^{(1)} E_y + \chi_{xz}^{(1)} E_z] \\ P_y^{(1)} = \varepsilon_0 [\chi_{yx}^{(1)} E_x + \chi_{yy}^{(1)} E_y + \chi_{yz}^{(1)} E_z] \\ P_z^{(1)} = \varepsilon_0 [\chi_{zx}^{(1)} E_x + \chi_{zy}^{(1)} E_y + \chi_{zz}^{(1)} E_z] \end{array} \right\}$$

$$P_i^{(1)} = \varepsilon_0 \sum_j \chi_{ij}^{(1)} E_j$$

$$(i, j) = (x, y, z)$$

2^{nd}-order susceptibility is a 3^{rd}-rank tensor

Take sum frequency generation(SFG) $\omega_1 + \omega_2 = \omega_3$ as an example:

$$P_x^{(2)}(\omega_3,\omega_1,\omega_2) = \varepsilon_0[\chi_{xxx}^{(2)}E_x(\omega_1)E_x(\omega_2) + \chi_{xxy}^{(2)}E_x(\omega_1)E_y(\omega_2) + \chi_{xxz}^{(2)}E_x(\omega_1)E_z(\omega_2)$$

$$+ \chi_{xyx}^{(2)}E_y(\omega_1)E_x(\omega_2) + \chi_{xyy}^{(2)}E_y(\omega_1)E_y(\omega_2) + \chi_{xyz}^{(2)}E_y(\omega_1)E_z(\omega_2)$$

$$+ \chi_{xzx}^{(2)}E_z(\omega_1)E_x(\omega_2) + \chi_{xzy}^{(2)}E_z(\omega_1)E_y(\omega_2) + \chi_{xzz}^{(2)}E_z(\omega_1)E_z(\omega_2)]$$

We can represent the lengthy expression using tensor notation:

$$P_i^{(2)}(\omega_3,\omega_1,\omega_2) = \varepsilon_0 \sum_{j,k} \chi_{ijk}^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2)$$

$$(i,j,k) = (x,y,z)$$

$$P_i^{(2)}(\omega_3,\omega_2,\omega_1) = \varepsilon_0 \sum_{k,j} \chi_{ikj}^{(2)}(\omega_1+\omega_2,\omega_2,\omega_1)E_k(\omega_2)E_j(\omega_1)$$

$\chi_{ijk}^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2)$ is a 3^{rd}-order tensor with 27 (3X3X3) elements. According to the crystal symmetry, most of them are zeros.

$$\chi_{ijk}^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2) = \chi_{ikj}^{(2)}(\omega_1+\omega_2,\omega_2,\omega_1)$$

Due to intrinsic permutation symmetry

$$P_i^{(2)}(\omega_3) = 2P_i^{(2)}(\omega_3,\omega_1,\omega_2) = 2\varepsilon_0 \sum_{j,k} \chi_{ijk}^{(2)}(\omega_1+\omega_2,\omega_1,\omega_2)E_j(\omega_1)E_k(\omega_2)$$

Susceptibility is a tensor

Tensor describes linear relations between geometric vectors, scalars, or other tensors. --Wiki

Tensor rank	0 (scalar)	1 (vector)	2 (matrix)	3	4
# of components	$3^0 = 1$	$3^1 = 3$	$3^2 = 9$	$3^3 = 27$	$3^4 = 81$
Examples	a	$\begin{bmatrix} E_x \\ E_y \\ E_z \end{bmatrix}$	$\chi_{ij}^{(1)} = \begin{bmatrix} \chi_{xx}^{(1)} & \chi_{xy}^{(1)} & \chi_{xz}^{(1)} \\ \chi_{yx}^{(1)} & \chi_{yy}^{(1)} & \chi_{yz}^{(1)} \\ \chi_{zx}^{(1)} & \chi_{zy}^{(1)} & \chi_{zz}^{(1)} \end{bmatrix}$	2nd-order nonlinear susceptibility $\chi_{ijk}^{(2)}$	3rd -order nonlinear susceptibility $\chi_{ijkl}^{(3)}$

Linear (the 1st-order) susceptibility is a 2nd -order tensor (i.e., 3 by 3 matrix):

$$\left.\begin{array}{l} P_i^{(1)} = \varepsilon_0 \sum_j \chi_{ij}^{(1)} E_j \\ (i, j) = (x, y, z) \end{array}\right\}$$

A more convenient notation: repeated indices imply summation.

$$P_i^{(1)} = \varepsilon_0 \chi_{ij}^{(1)} E_j$$

2nd-order susceptibility is a 3rd -order tensor with 27 elements:

$$P_i^{(2)}(\omega_3, \omega_1, \omega_2) = \varepsilon_0 \chi_{ijk}^{(2)} E_j(\omega_1) E_k(\omega_2)$$

3rd -order susceptibility is a 4th -order tensor with 81 elements:

$$P_i^{(3)}(\omega_4, \omega_1, \omega_2, \omega_3) = \varepsilon_0 \chi_{ijkl}^{(3)} E_j(\omega_1) E_k(\omega_2) E_l(\omega_3)$$

Kleinmann symmetry reduces number of tensor elements

If all the frequencies involved are far away from the resonance frequencies of the medium, the nonlinear susceptibilities are independent of frequency and ijk indices become equal:

$$\chi_{xyz}^{(2)} = \chi_{xzy}^{(2)} = \chi_{yxz}^{(2)} = \chi_{yzx}^{(2)} = \chi_{zxy}^{(2)} = \chi_{zyx}^{(2)}$$

Under Kleinmann symmetry condition, 27 elements are reduced to 10.

Take sum frequency generation(SFG) $\omega_1 + \omega_2 = \omega_3$ as an example:

$$\begin{aligned}
P_x^{(2)}(\omega_3,\omega_1,\omega_2) = \varepsilon_0[&\chi_{xxx}^{(2)} E_x(\omega_1)E_x(\omega_2) + \underline{\chi_{xxy}^{(2)}} E_x(\omega_1)E_y(\omega_2) + \underline{\chi_{xxz}^{(2)}} E_x(\omega_1)E_z(\omega_2) \\
&+ \underline{\chi_{xyx}^{(2)}} E_y(\omega_1)E_x(\omega_2) + \chi_{xyy}^{(2)} E_y(\omega_1)E_y(\omega_2) + \underline{\chi_{xyz}^{(2)}} E_y(\omega_1)E_z(\omega_2) \\
&+ \underline{\chi_{xzx}^{(2)}} E_z(\omega_1)E_x(\omega_2) + \underline{\chi_{xzy}^{(2)}} E_z(\omega_1)E_y(\omega_2) + \chi_{xzz}^{(2)} E_z(\omega_1)E_z(\omega_2)]
\end{aligned}$$

$$\Downarrow$$

$$\begin{aligned}
P_x^{(2)}(\omega_3,\omega_1,\omega_2) = \varepsilon_0\{ &\chi_{xxx}^{(2)} E_x(\omega_1)E_x(\omega_2) + \chi_{xyy}^{(2)} E_y(\omega_1)E_y(\omega_2) + \chi_{xzz}^{(2)} E_z(\omega_1)E_z(\omega_2) \\
&+ \chi_{xxy}^{(2)}[E_x(\omega_1)E_y(\omega_2) + E_y(\omega_1)E_x(\omega_2)] + \chi_{xyz}^{(2)}[E_y(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_y(\omega_2)] \\
&+ \chi_{xxz}^{(2)}[E_x(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_x(\omega_2)]\}
\end{aligned}$$

Contracted suffix notation

Contracted suffix notation is more commonly used in literature:

$$\chi_{ijk}^{(2)} = 2d_{np} \qquad (i,j,k)=(x,y,z) \qquad i \to n, (jk) \to p$$

i	x	y	z			
n	1	2	3			
jk	xx	yy	zz	yz, zy	xz, zx	xy, yx
p	1	2	3	4	5	6

$$\begin{bmatrix} P_x(\omega_3) \\ P_y(\omega_3) \\ P_z(\omega_3) \end{bmatrix} = 4\varepsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{21} & d_{22} & d_{23} & d_{24} & d_{25} & d_{26} \\ d_{31} & d_{32} & d_{33} & d_{34} & d_{35} & d_{36} \end{bmatrix} \begin{bmatrix} E_x(\omega_1)E_x(\omega_2) \\ E_y(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_z(\omega_2) \\ E_y(\omega_1)E_z(\omega_2)+E_z(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_x(\omega_2)+E_x(\omega_1)E_z(\omega_2) \\ E_x(\omega_1)E_y(\omega_2)+E_y(\omega_1)E_x(\omega_2) \end{bmatrix}$$

Contracted suffix notation

- Under Kleinmann symmetry condition, some of these 18 elements are the same, and there are actually 10 independent elements.

$$d_{21} = d_{16} \quad d_{25} = d_{14} \quad d_{26} = d_{12} \quad d_{31} = d_{15}$$

$$d_{32} = d_{24} \quad d_{34} = d_{23} \quad d_{35} = d_{13} \quad d_{36} = d_{14}$$

$$\begin{bmatrix} P_x(\omega_3) \\ P_y(\omega_3) \\ P_z(\omega_3) \end{bmatrix} = 4\varepsilon_0 \begin{bmatrix} d_{11} & d_{12} & d_{13} & d_{14} & d_{15} & d_{16} \\ d_{16} & d_{22} & d_{23} & d_{24} & d_{14} & d_{12} \\ d_{15} & d_{24} & d_{33} & d_{23} & d_{13} & d_{14} \end{bmatrix} \begin{bmatrix} E_x(\omega_1)E_x(\omega_2) \\ E_y(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_z(\omega_2) \\ E_y(\omega_1)E_z(\omega_2) + E_z(\omega_1)E_y(\omega_2) \\ E_z(\omega_1)E_x(\omega_2) + E_x(\omega_1)E_z(\omega_2) \\ E_x(\omega_1)E_y(\omega_2) + E_y(\omega_1)E_x(\omega_2) \end{bmatrix}$$

- Crystal symmetry causes most of the elements to be zero for most symmetry groups. Take BBO as an example:

$$d_{np} = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & d_{16} \\ d_{16} & -d_{16} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix}$$

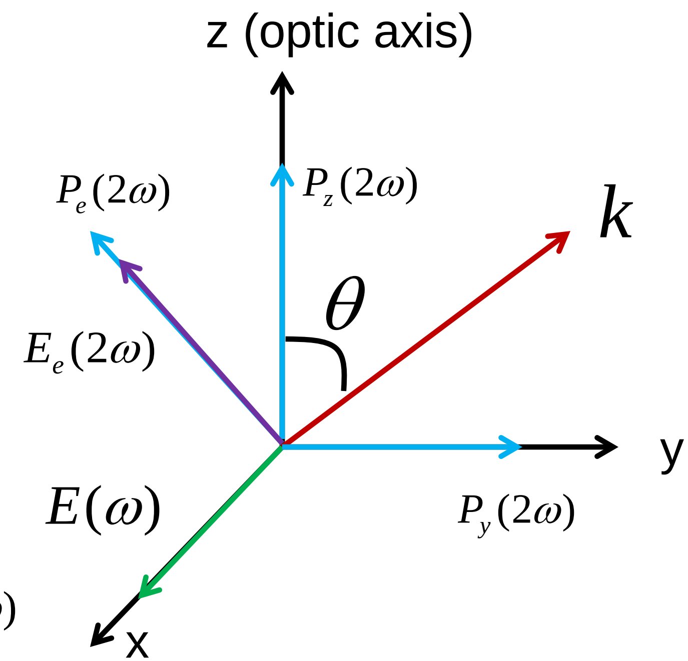
There are only 4 independent elements.

$$d_{16}(1.064um) = 2.2\, pm/V \quad d_{15}(1.064um) = 0.03\, pm/V$$

$$d_{31}(1.064um) = 0.04\, pm/V \quad d_{33}(1.064um) = 0.04\, pm/V$$

Example: SHG of o wave in BBO

One special case: assume the k vector in the yz plane, and the electrical field is along the x-axis; that is, we consider an ordinary wave.



$$\begin{bmatrix} P_x(2\omega) \\ P_y(2\omega) \\ P_z(2\omega) \end{bmatrix} = 2\varepsilon_0 \begin{bmatrix} 0 & 0 & 0 & 0 & d_{15} & d_{16} \\ d_{16} & -d_{16} & 0 & d_{15} & 0 & 0 \\ d_{31} & d_{31} & d_{33} & 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} E^2(\omega) \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{bmatrix}$$

$$P_x(2\omega) = 0 \quad P_y(2\omega) = 2\varepsilon_0 d_{16} E^2(\omega) \quad P_z(2\omega) = 2\varepsilon_0 d_{31} E^2(\omega)$$

We can project the polarization onto k-direction and the direction normal to k:

$$P_k(2\omega) = 2\varepsilon_0 (d_{16}\sin\theta + d_{31}\cos\theta) E^2(\omega) \longrightarrow$$

Diople oscillating along k-direction does not radiate into k direction.

$$P_e(2\omega) = 2\varepsilon_0 (d_{31}\sin\theta - d_{16}\cos\theta) E^2(\omega) = 2\varepsilon_0 d_{eff} E^2(\omega) \longrightarrow$$

In the k-z plane and therefore it radiates e wave

$$\left.\begin{aligned} d_{eff} &= d_{31}\sin\theta - d_{16}\cos\theta \\ |d_{31}(1.064um)| &= 0.04\,pm/V \\ |d_{16}(1.064um)| &= 2.2\,pm/V \end{aligned}\right\} \longrightarrow$$

Seems to suggest choosing $\theta = 0^\circ$ to maximize the nonlinearity. The answer is NO! (we will show why later.)

$$(\nabla^2 - \frac{1}{c_0^2}\frac{\partial^2}{\partial t^2})E = \mu_0 \frac{\partial^2 P}{\partial t^2} = \mu_0 (\frac{\partial^2 P_L}{\partial t^2} + \frac{\partial^2 P_{NL}}{\partial t^2}) \longrightarrow (\nabla^2 - \frac{n_e^2(2\omega)}{c^2}\frac{\partial^2}{\partial t^2})E_e(2\omega) = \mu_0 \frac{\partial^2 P_e(2\omega)}{\partial t^2}$$

Early history of lasers

- 1917: *on the quantum theory of radiation* – Einstein's paper
- 1954: MASER by Charles Townes (1915—2015) *et al*.

If you're a nobel prize winner, and 100 years old, you can comment other winners using harsh words:

Charles Townes

University of California, Berkeley, and 1964 Nobel Prize in Physics recipient

Jim Gordon was a fine person and a great scientist. He was also brave in doing research. When he worked for me as a graduate student trying to build the first maser, the chairman of the physics department and the previous chairman both told him it would not work and that he should stop, because the project was wasting the department's money. Both of them had Nobel Prizes, so presumably weren't stupid physicists. But Jim proceeded with his work and, about four months after they told him it wouldn't work, it did. From the maser also came the laser.

Jim didn't get the Nobel Prize with me, presumably because he was a student when the maser first worked, but I think he deserved it. He went on to do other important work. We should all celebrate him and his contributions.

Optics & Photonics News, 2014

MASER: **M**icrowave **A**mplification by **S**timulated **E**mission of **R**adiation
(**M**eans of **A**cquiring **S**upport for **E**xpensive **R**esearch)

First SHG experiment performed 1 year after laser was invented

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

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)

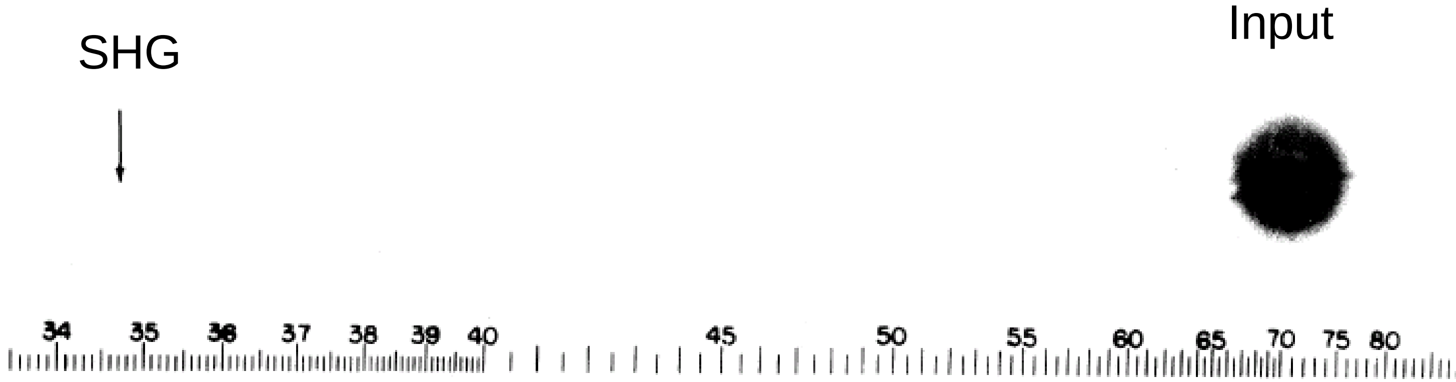
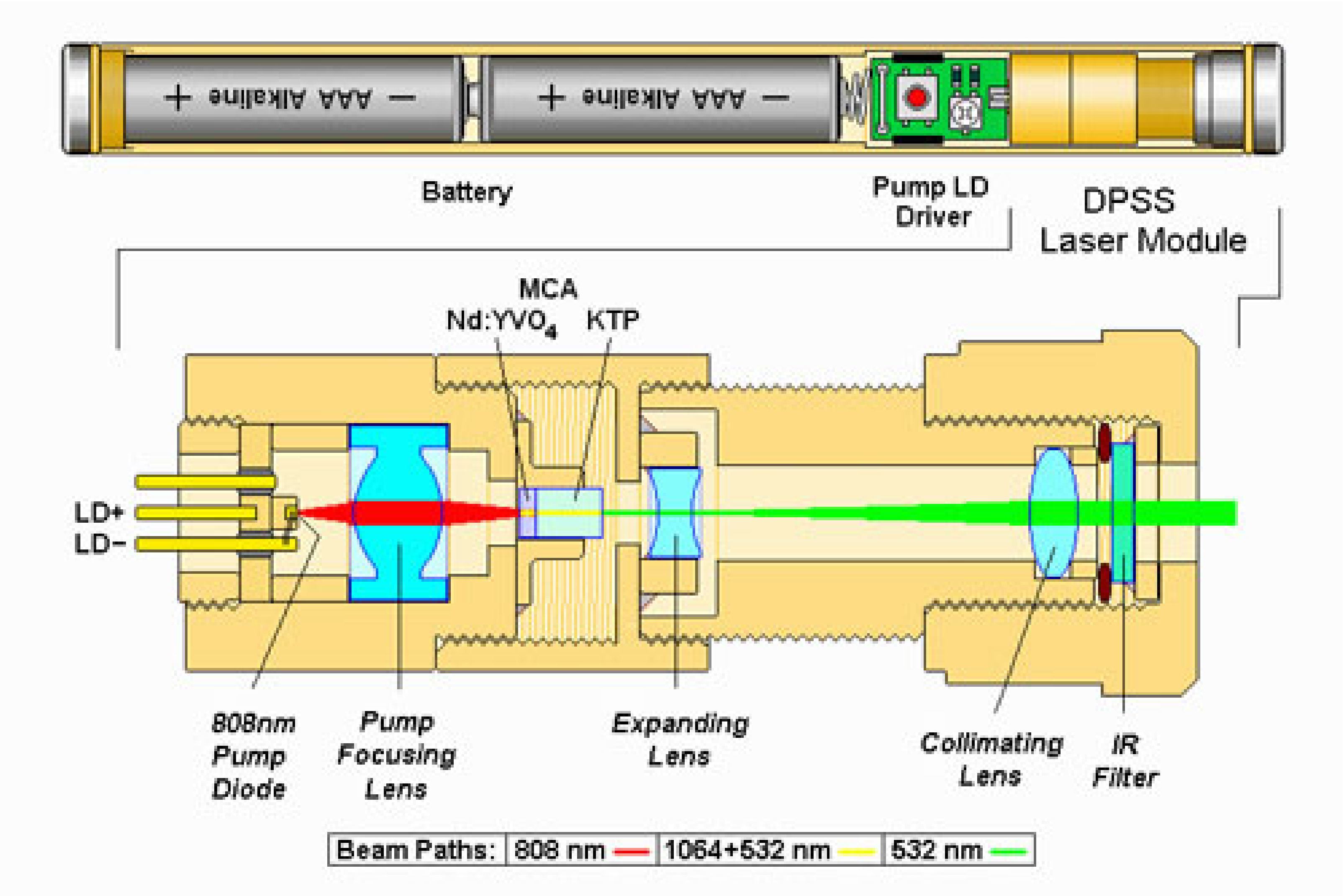


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

The very weak spot due to the second harmonic is missing. It was removed by an overzealous Physical Review Letters editor, who thought it was a speck of dirt and didn't ask the authors first.

SHG in daily life: green laser pointer



Take-home message

- Material polarization at high input E-field can be modeled by anharmonic electron oscillation.
- 2^{nd} –order nonlinear susceptibility is a 3^{rd} rank tensor with 27 elements and 3^{rd} –order nonlinear susceptibility is a 4^{th} rank tensor with 81 elements.
- Most of these tensor elements are zero rendered by Kleinmann symmetry and crystal symmetry.
- Even-order nonlinear effects vanish for centrosymmetric optical crystals.

Suggested reading

Anharmonic oscillator model

-- Robert Boyd, *Nonlinear optics*, chapter 1

Nonlinear susceptibility: tensor and symmetry

-- Geoffrey New, *Introduction to nonlinear optics*, chapter 4

-- George Stegemann and Robert Stegemann, *Nonlinear optics*, chapter 2

-- Robert Boyd, *Nonlinear optics*, chapter 1