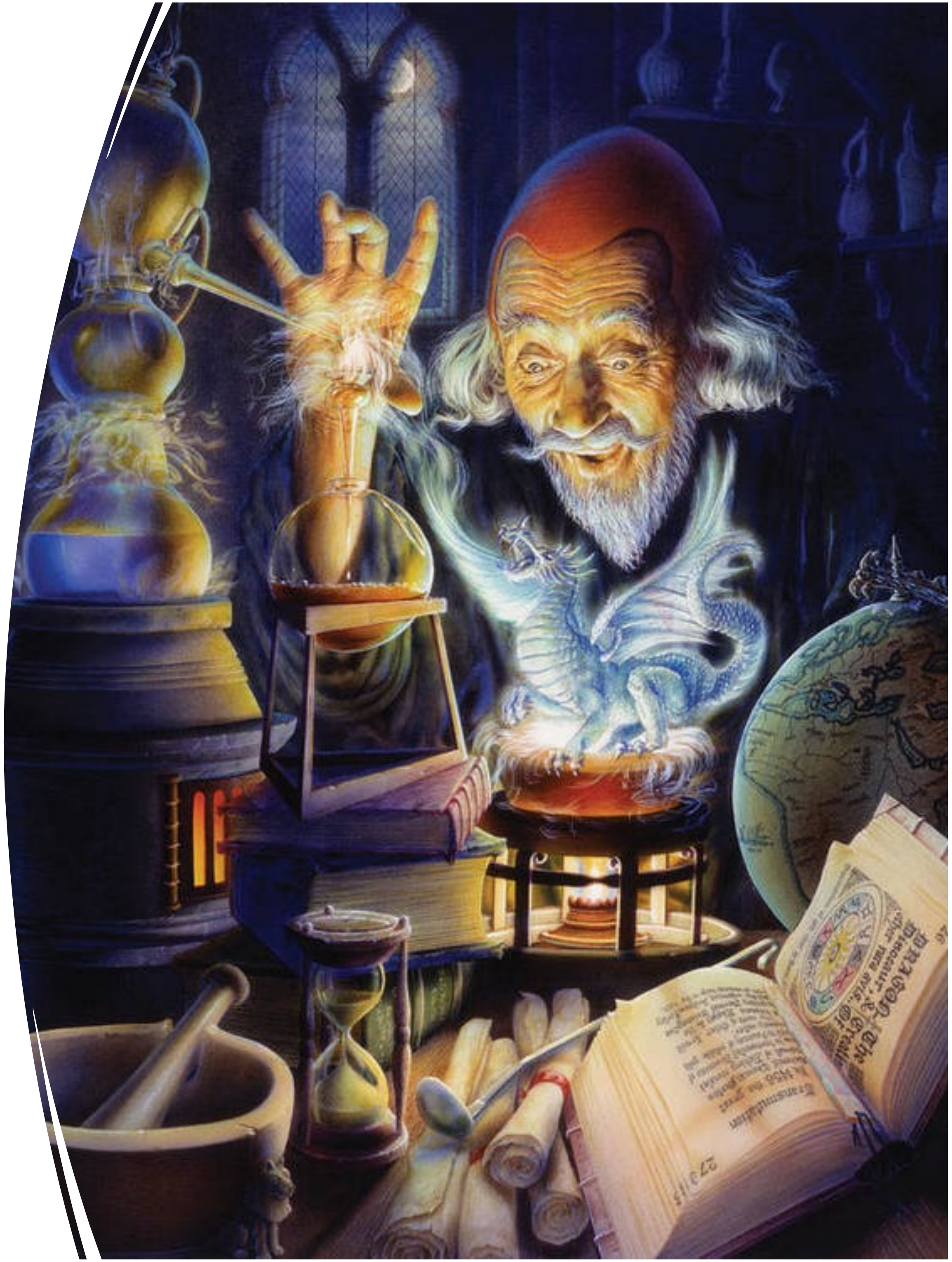
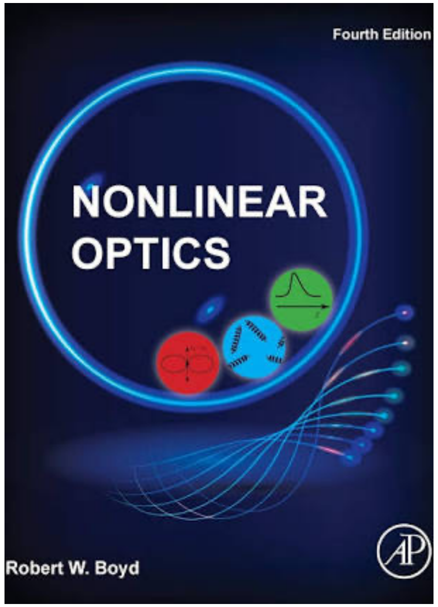
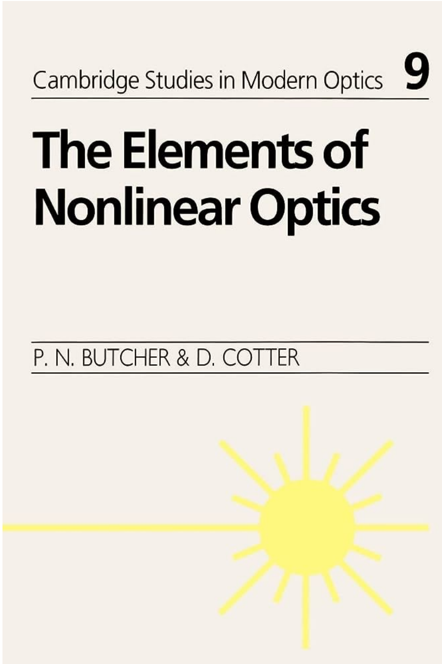
Nonlinear optics: a first look

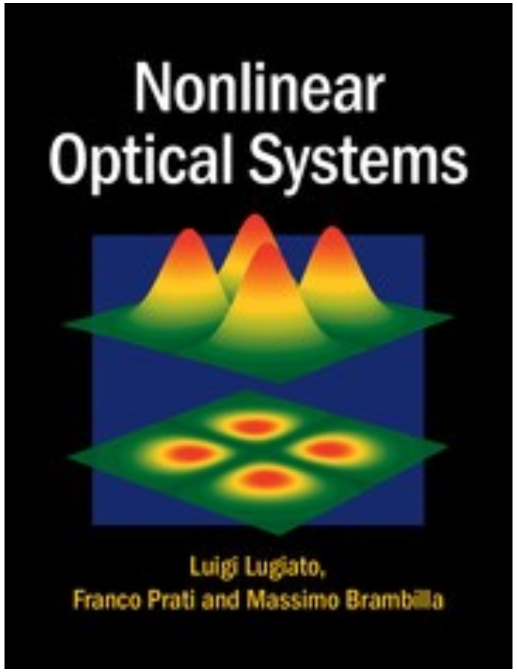
J. E. Sipe
Department of Physics
University of Toronto

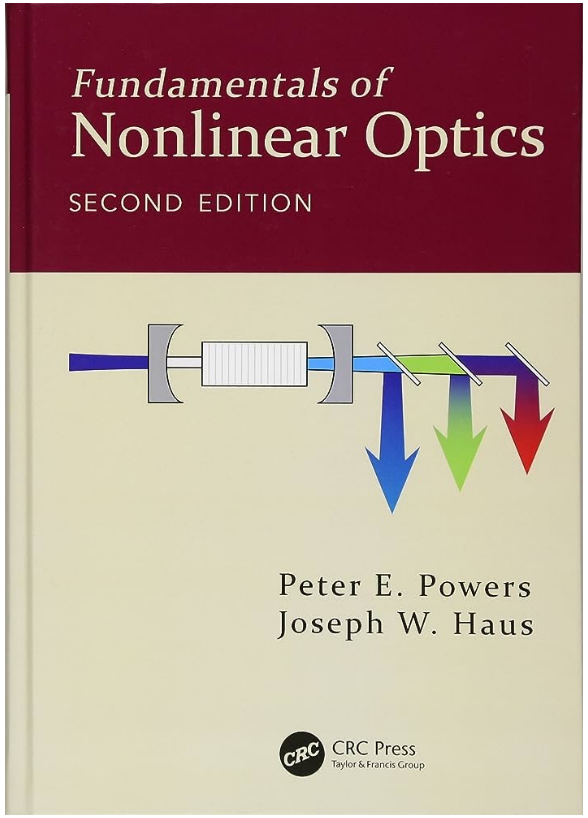
"Optical alchemy"




Fourth Edition
NONLINEAR OPTICS
Robert W. Boyd


Cambridge Studies in Modern Optics 9
The Elements of Nonlinear Optics
P. N. BUTCHER & D. COTTER


Nonlinear Optical Systems
Luigi Lugiato,
Franco Prati and Massimo Brambilla


Fundamentals of
Nonlinear Optics
SECOND EDITION
Peter E. Powers
Joseph W. Haus
CRC Press
Taylor & Francis Group

Susceptibilities

$\chi^{(2)}$ *effects*

$\chi^{(3)}$ *effects*

Quantum nonlinear optics

Nonlinear optics and electronics

Forbidden processes

Susceptibilities

$\chi^{(2)}$ **effects**

$\chi^{(3)}$ **effects**

Quantum nonlinear optics

Nonlinear optics and electronics

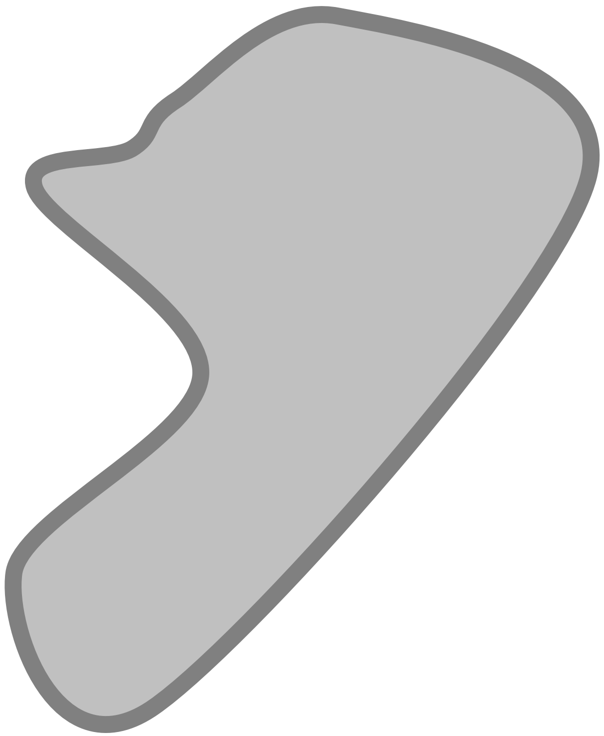
Forbidden processes

$$P(t) = \chi \, E(t)$$

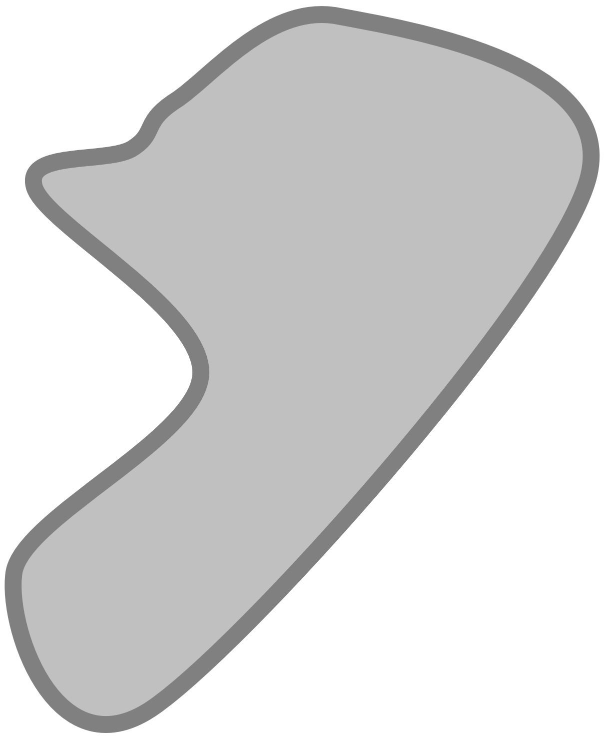
$$P(t) = \chi^{(1)} E(t)$$

$$P(t) = \chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + ...$$

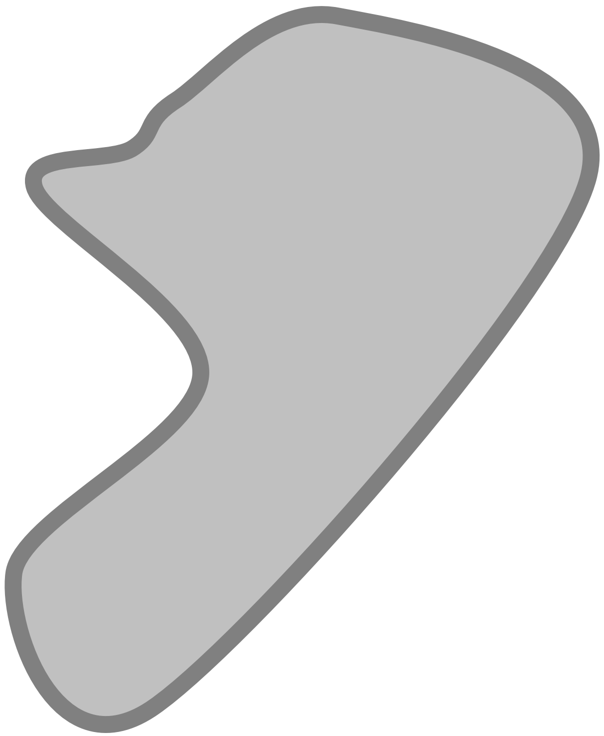
$$P(t) = \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + ...$$



$$P(\boldsymbol{r},t) = \chi^{(1)}(\boldsymbol{r})E(\boldsymbol{r},t) + \chi^{(2)}(\boldsymbol{r})E^2(\boldsymbol{r},t) + \chi^{(3)}(\boldsymbol{r})E^3(\boldsymbol{r},t) + ...$$



$$P(t) = \chi^{(1)}E(t) + \chi^{(2)}E^2(t) + \chi^{(3)}E^3(t) + ...$$



$$P_i(t) = \chi_{ij}^{(1)} E_j(t) + \chi_{ijk}^{(2)} E_j(t) E_k(t) + \chi_{ijkl}^{(3)} E_j(t) E_k(t) E_l(t) + ...$$

$$P_i(t) = \chi^{(1)}_{ij} E_j(t) + \chi^{(2)}_{ijk} E_j(t) E_k(t) + \chi^{(3)}_{ijkl} E_j(t) E_k(t) E_l(t) + ...$$

implicit sum over repeated Cartesian components

$$P_i(t) = \chi^{(1)}_{ij} E_j(t) + \chi^{(2)}_{ijk} E_j(t) E_k(t) + \chi^{(3)}_{ijkl} E_j(t) E_k(t) E_l(t) + ...$$

vanishes if medium has centre-of-inversion symmetry

$$E_i(t) = \sum_{\omega} E_i(\omega) e^{-i\omega t}$$

$$P_i(t) = \chi_{ij}^{(1)} E_j(t) + \chi_{ijk}^{(2)} E_j(t) E_k(t) + \chi_{ijkl}^{(3)} E_j(t) E_k(t) E_l(t) + ...$$

$$E_i(t) = \sum_{\omega} E_i(\omega) e^{-i\omega t}$$

$$\begin{aligned}
P_i(t) &= \sum_{\omega} \chi_{ij}^{(1)}(-\omega;\omega) E_j(\omega) e^{-i\omega t} \\
&+ \sum_{\omega,\omega'} \chi_{ijk}^{(2)}(-\omega-\omega';\omega,\omega') E_j(\omega) E_k(\omega') e^{-i(\omega+\omega')t} \\
&+ \sum_{\omega,\omega',\omega''} \chi_{ijkl}^{(3)}(-\omega-\omega'-\omega'';\omega,\omega',\omega'') E_j(\omega) E_k(\omega') E_l(\omega'') e^{-i(\omega+\omega'+\omega'')t} \\
&+ \ldots\ldots
\end{aligned}$$

$$\chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega')$$

$$
\begin{aligned}
P_i(t) &= \sum_{\omega} \chi^{(1)}_{ij}(-\omega;\omega)E_j(\omega)e^{-i\omega t} \\
&+ \sum_{\omega,\omega'} \chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega')E_j(\omega)E_k(\omega')e^{-i(\omega+\omega')t} \\
&+ \sum_{\omega,\omega',\omega''} \chi^{(3)}_{ijkl}(-\omega-\omega'-\omega'';\omega,\omega',\omega'')E_j(\omega)E_k(\omega')E_l(\omega'')e^{-i(\omega+\omega'+\omega'')t} \\
&+ .....
\end{aligned}
$$

$$\chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega')$$

or $\chi^{(2)}_{ijk}(\omega,\omega';\omega+\omega')$

$$P_i(t)=\sum_{\omega}\chi^{(1)}_{ij}(-\omega;\omega)E_j(\omega)e^{-i\omega t}$$

$$+\sum_{\omega,\omega'}\chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega')E_j(\omega)E_k(\omega')e^{-i(\omega+\omega')t}$$

$$+\sum_{\omega,\omega',\omega''}\chi^{(3)}_{ijkl}(-\omega-\omega'-\omega'';\omega,\omega',\omega'')E_j(\omega)E_k(\omega')E_l(\omega'')e^{-i(\omega+\omega'+\omega'')t}$$

$$+\,.....$$

$$\chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega')$$

or $\chi^{(2)}_{ijk}(\omega,\omega';\omega+\omega')$

or $\chi^{(2)}_{ijk}(\omega,\omega')$

$$P_i(t) = \sum_{\omega} \chi^{(1)}_{ij}(-\omega;\omega)E_j(\omega)e^{-i\omega t}$$

$$+ \sum_{\omega,\omega'} \chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega')E_j(\omega)E_k(\omega')e^{-i(\omega+\omega')t}$$

$$+ \sum_{\omega,\omega',\omega''} \chi^{(3)}_{ijkl}(-\omega-\omega'-\omega'';\omega,\omega',\omega'')E_j(\omega)E_k(\omega')E_l(\omega'')e^{-i(\omega+\omega'+\omega'')t}$$

$$+ \ldots..$$

$$E_i(t) = \sum_{\omega} E_i(\omega) e^{-i\omega t}$$

$$P_i(t) = \sum_{\omega} \chi^{(1)}_{ij}(-\omega;\omega) E_j(\omega) e^{-i\omega t}$$
$$+ \sum_{\omega,\omega'} \chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega') E_j(\omega) E_k(\omega') e^{-i(\omega+\omega')t}$$
$$+ \sum_{\omega,\omega',\omega''} \chi^{(3)}_{ijkl}(-\omega-\omega'-\omega'';\omega,\omega',\omega'') E_j(\omega) E_k(\omega') E_l(\omega'') e^{-i(\omega+\omega'+\omega'')t}$$
$$+ \ldots..$$

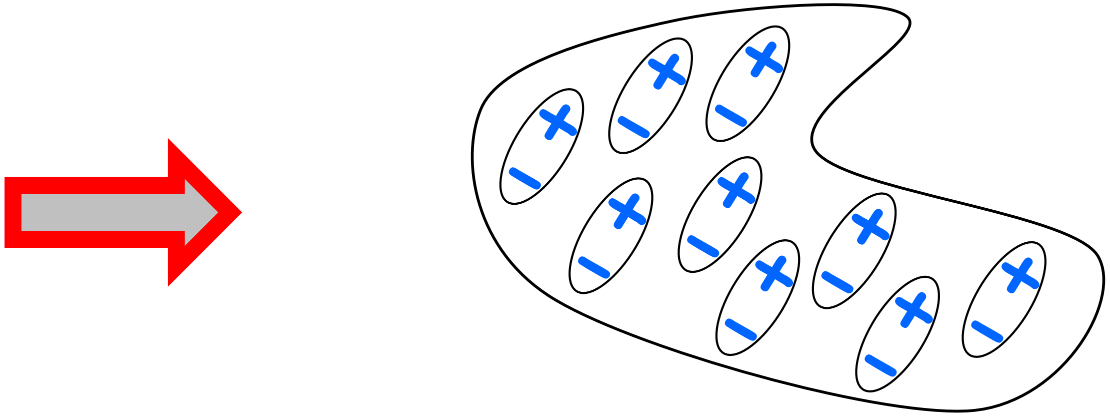
$$E_i(t) = \sum_{\omega} E_i(\omega) e^{-i\omega t}$$

$$\begin{aligned} P_i(t) = &\sum_{\omega} \chi^{(1)}_{ij}(\text{[illegible]}\,\omega) E_j(\omega) e^{-i\omega t} \\ &+ \sum_{\omega,\omega'} \chi^{(2)}_{ijk}(\text{[illegible]}\,\omega,\omega') E_j(\omega) E_k(\omega') e^{-i(\omega+\omega')t} \\ &+ \sum_{\omega,\omega',\omega''} \chi^{(3)}_{ijkl}(\text{[illegible]}\,\omega,\omega',\omega'') E_j(\omega) E_k(\omega') E_l(\omega'') e^{-i(\omega+\omega'+\omega'')t} \\ &+ \ldots.. \end{aligned}$$

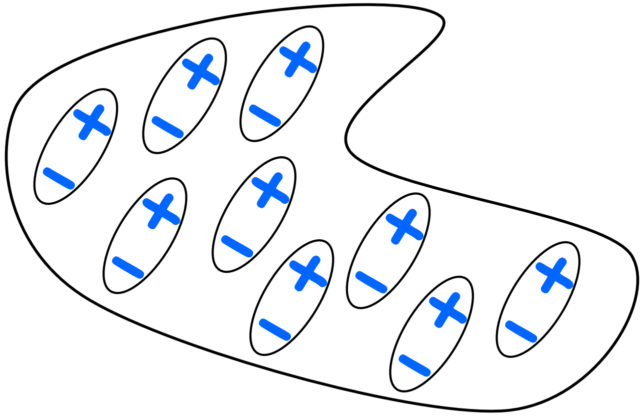
$$E_i(t) = \sum_{\omega} E_i(\omega) e^{-i\omega t}$$

$$\begin{aligned} P_i(t) = & \sum_{\omega} \chi^{(1)}_{ij}(\;\;\;;\omega) E_j(\omega) e^{-i\omega t} \\ & + \sum_{\omega,\omega'} \chi^{(2)}_{ijk}(\;\;\;;\omega,\omega') E_j(\omega) E_k(\omega') e^{-i(\omega+\omega')t} \\ & + \sum_{\omega,\omega',\omega''} \chi^{(3)}_{ijkl}(\;\;\;;\omega,\omega',\omega'') E_j(\omega) E_k(\omega') E_l(\omega'') e^{-i(\omega+\omega'+\omega'')t} \\ & + \ldots\ldots \end{aligned}$$

How calculate?



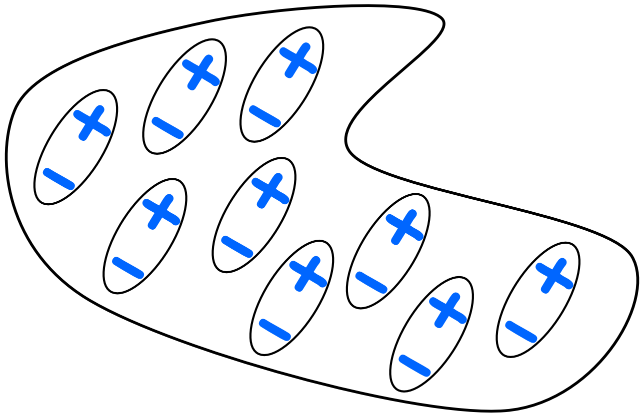
simple molecular models



simple molecular models

Calculate J(t) from minimal coupling Hamiltonian

$$\boldsymbol{J}(t) = \frac{d\boldsymbol{P}(t)}{dt} \iff \boldsymbol{J}(\omega) = -i\omega \boldsymbol{P}(\omega)$$



simple molecular models

Calculate J(t) from minimal coupling Hamiltonian

$$\boldsymbol{J}(t) = \frac{d\boldsymbol{P}(t)}{dt} \iff \boldsymbol{J}(\omega) = -i\omega\boldsymbol{P}(\omega)$$

Adams and Blount

$$\boldsymbol{P} = e\int \frac{d\boldsymbol{k}d\boldsymbol{k}'}{V}\sum_{n,m} a_n^\dagger(\boldsymbol{k}) \langle n\boldsymbol{k}|\boldsymbol{r}|m\boldsymbol{k'}\rangle \, a_m(\boldsymbol{k}')$$

$$\langle n\boldsymbol{k}|\boldsymbol{r}|m\boldsymbol{k'}\rangle = \delta(\boldsymbol{k}-\boldsymbol{k}')\xi_{nm}(\boldsymbol{k}) + i\delta_{nm}\frac{\partial}{\partial \boldsymbol{k}}\delta(\boldsymbol{k}-\boldsymbol{k'})$$

Susceptibilities

$\chi^{(2)}$ effects

$\chi^{(3)}$ effects

Quantum nonlinear optics

Nonlinear optics and electronics

Forbidden processes

Second harmonic generation

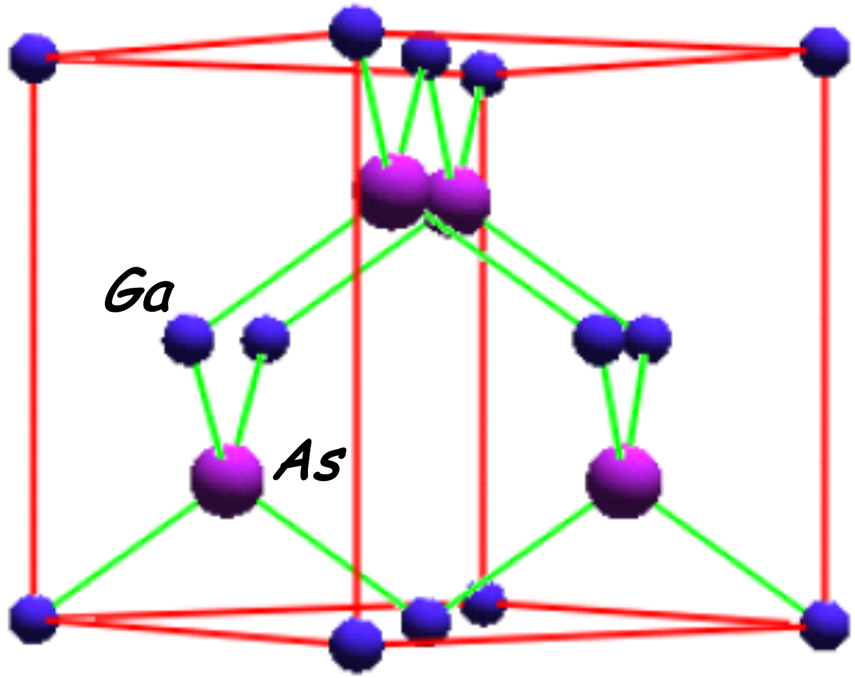
$$P_i(t) = \chi^{(2)}_{ijk}(-2\omega;\omega,\omega)E_j(\omega)E_k(\omega)e^{-2i\omega t} + c.c.$$

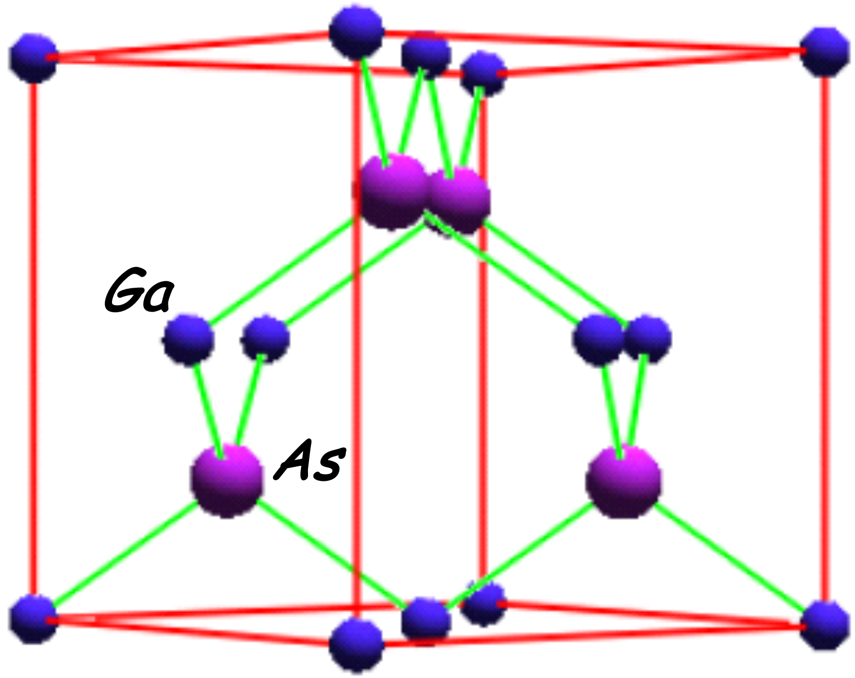
$$P_i(t) = \sum_{\omega} \chi^{(1)}_{ij}(-\omega;\omega)E_j(\omega)e^{-i\omega t}$$

$$+ \sum_{\omega,\omega'} \chi^{(2)}_{ijk}(-\omega-\omega';\omega,\omega')E_j(\omega)E_k(\omega')e^{-i(\omega+\omega')t}$$

$$+ \sum_{\omega,\omega',\omega''} \chi^{(3)}_{ijk}(-\omega-\omega'-\omega'';\omega,\omega',\omega'')E_j(\omega)E_k(\omega')E_l(\omega'')e^{-i(\omega+\omega'+\omega'')t}$$

$$+ \ldots..$$


Ga
As


Ga
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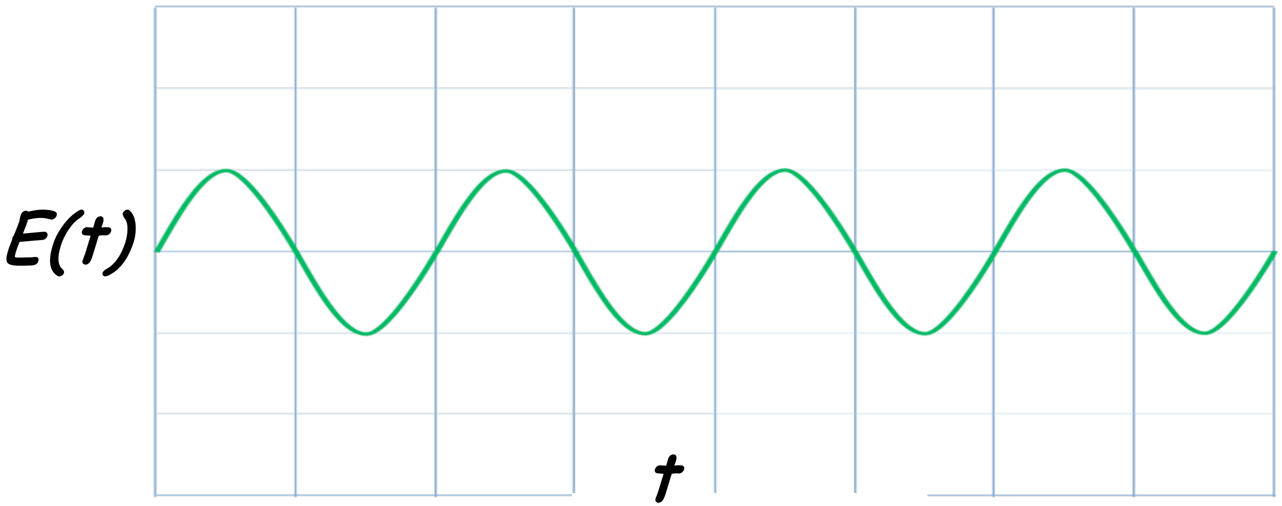
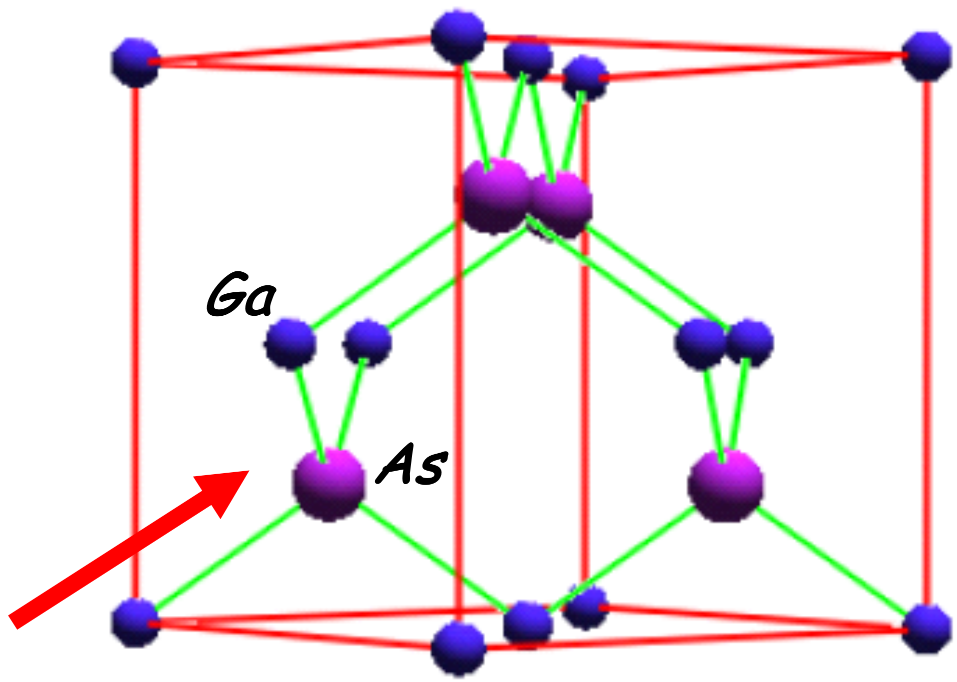

E(t)
t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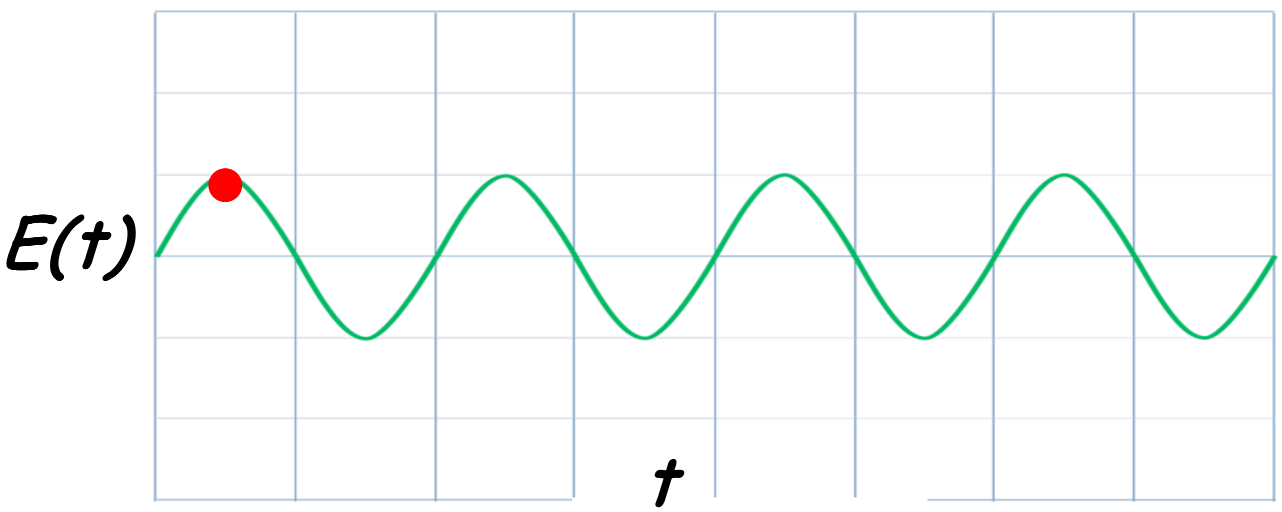
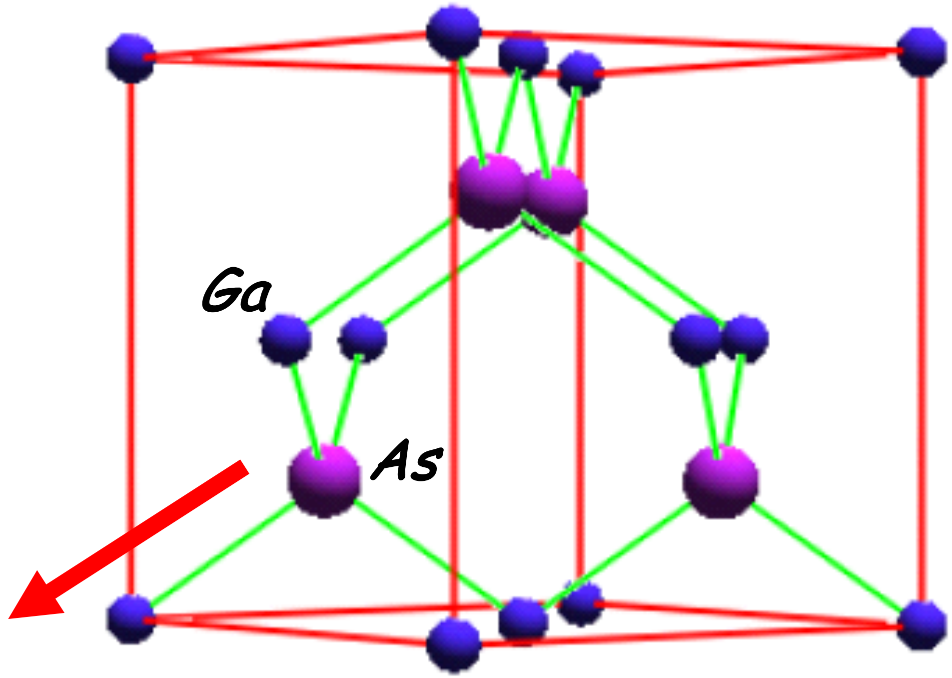
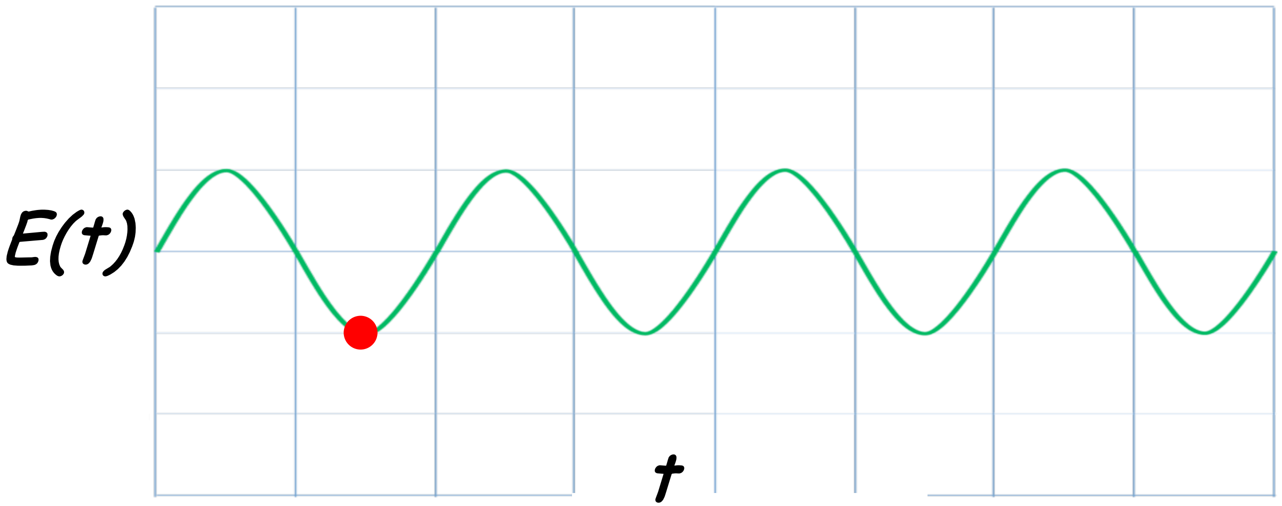
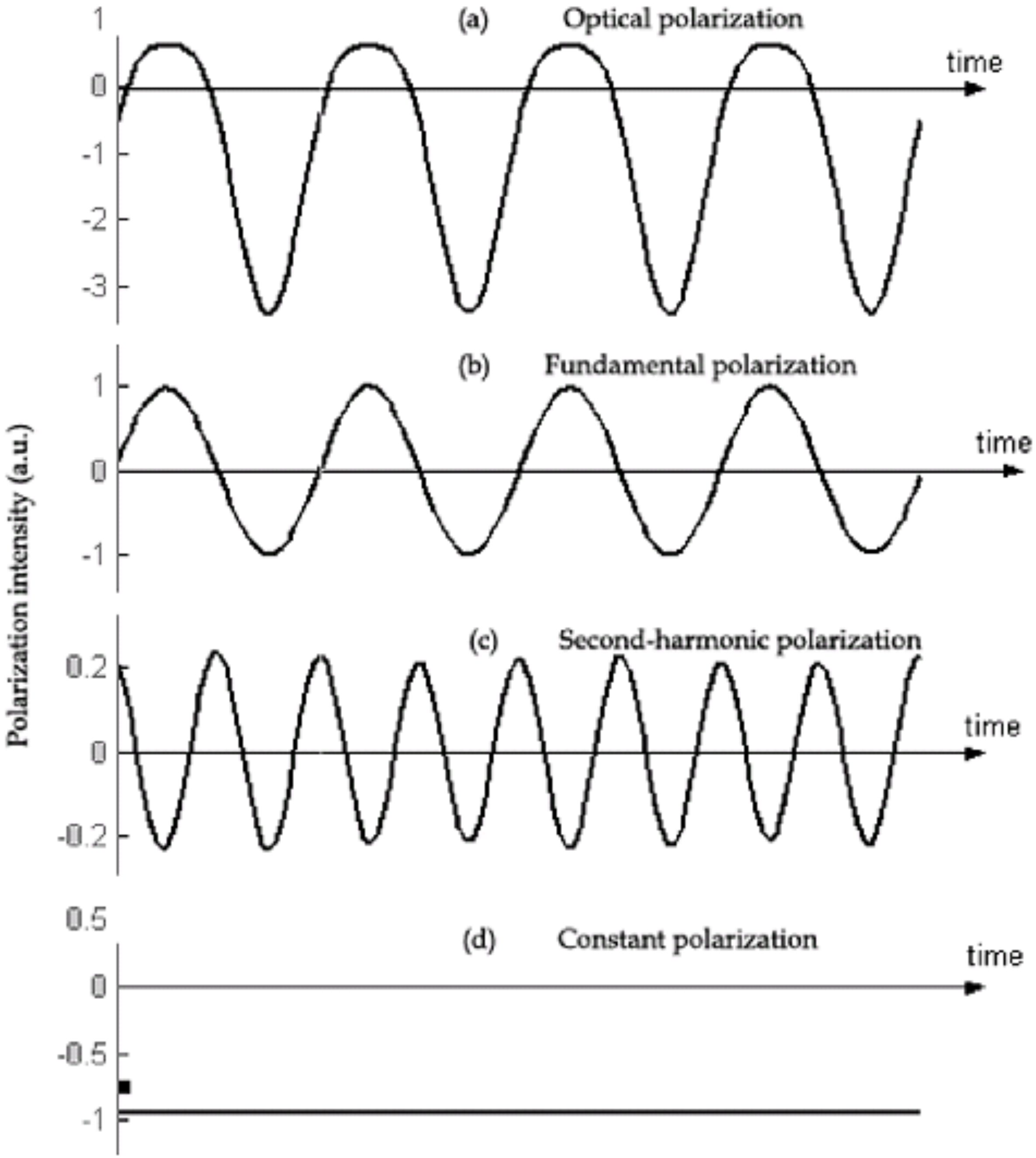
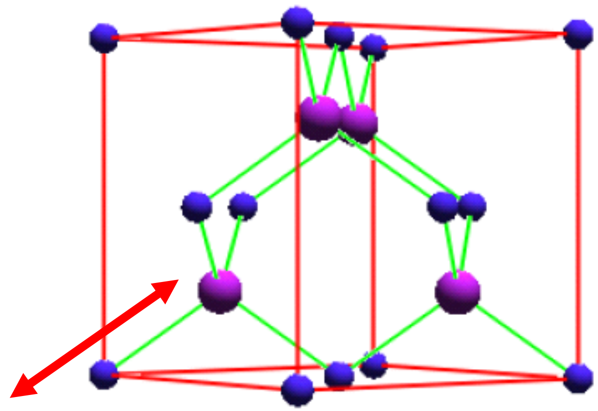
As


E(t)
t

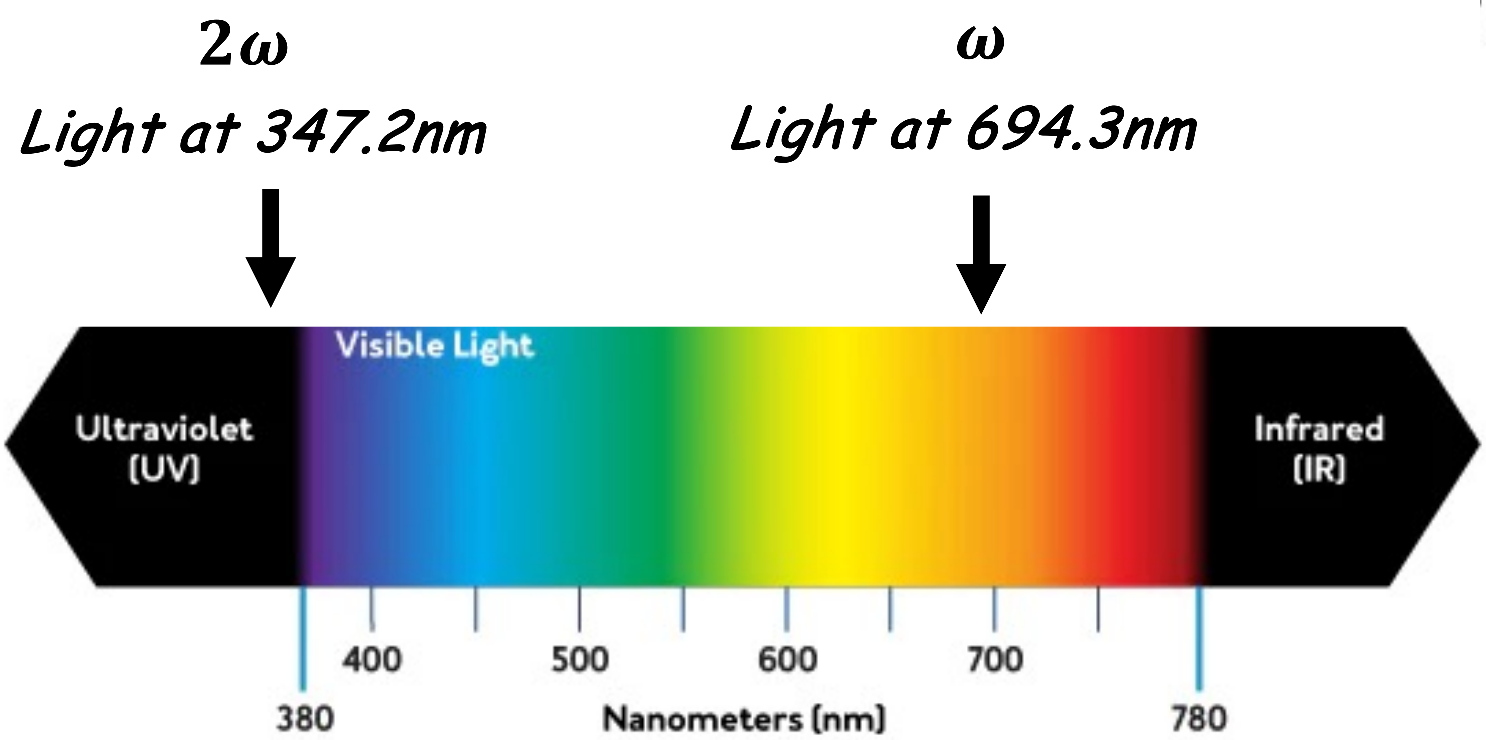

Ga
As


E(t)
t


(a) Optical polarization
(b) Fundamental polarization
(c) Second-harmonic polarization
(d) Constant polarization
Polarization intensity (a.u.)
time
1
0
-1
-2
-3
0.2
-0.2
0.5
-0.5


Second harmonic generation

$$P_i(t) = \chi^{(2)}_{ijk}(-2\omega;\omega,\omega)E_j(\omega)E_k(\omega)e^{-2i\omega t} + c.c.$$



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. The possibility of exploiting this extraordinary intensity for the production of optical harmonics from suitable nonlinear materials is most appealing. In this Letter we present a brief discussion of the requisite analysis and a description of experiments in which we have observed the second harmonic (at ~ 3472 A) produced upon projection of an intense beam of 6943A light through crystalline quartz.

A suitable material for the production of optical harmonics must have a nonlinear dielectric coefficient and be transparent to both the fundamental optical frequency and the desired overtones. Since all dielectrics are nonlinear in high enough fields, this suggests the feasibility of utilizing materials such as quartz and glass. The dependence of polarization of a dielectric upon electric field E may be expressed schematically by

$$P=\chi E\left(1+\frac{E}{E_1}+\frac{E^2}{E_2{}^2}+\cdots\right), \tag{1}$$

where E_1, E_2 ... are of the order of magnitude of atomic electric fields (~10^8 esu). If E is sinusoidal in time, the presence in Eq. (1) of terms of quadratic or higher degree will result in P con-

Table I. The square of the total p perpendicular to the direction of propagation of light through crystalline quartz.

Direction of incident beam	The square of the total p perpendicular to direction of propagation
x ($E_x=0$)	$p_y{}^2+p_z{}^2=0$
y ($E_y=0$)	$p_z{}^2+p_x{}^2=\alpha^2E_x{}^4$
z ($E_z=0$)	$p_x{}^2+p_y{}^2=\alpha^2(E_x{}^2+E_y{}^2)^2$

(z is the threefold, or optic, axis; x a twofold axis). If a light beam traverses quartz in one of the three principal directions, Eqs. (2) predict the results summarized in Table I. The second-harmonic light should be absent in the first case, dependent upon incident polarization in the second case, and independent of this polarization in the third.

If an intense beam of monochromatic light is focussed into a region of volume V, there should occur an intensity I of second harmonic given (in Gaussian units) by

$$I\sim(\omega^4/c^3)(pv)^2(V/v), \tag{3}$$

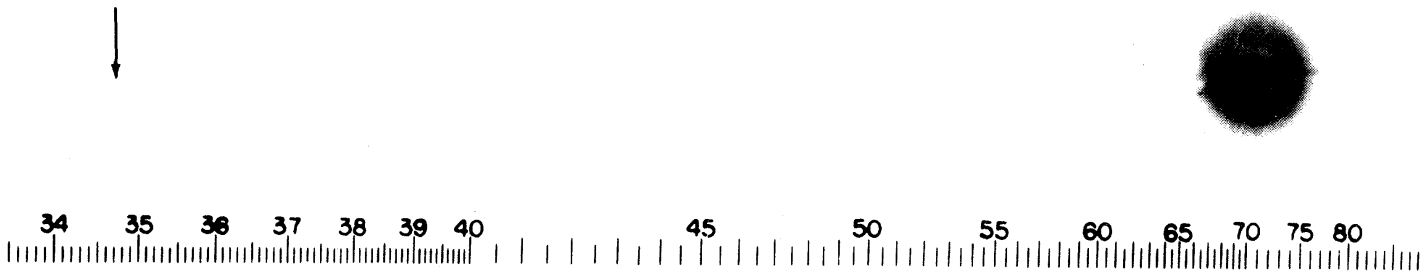
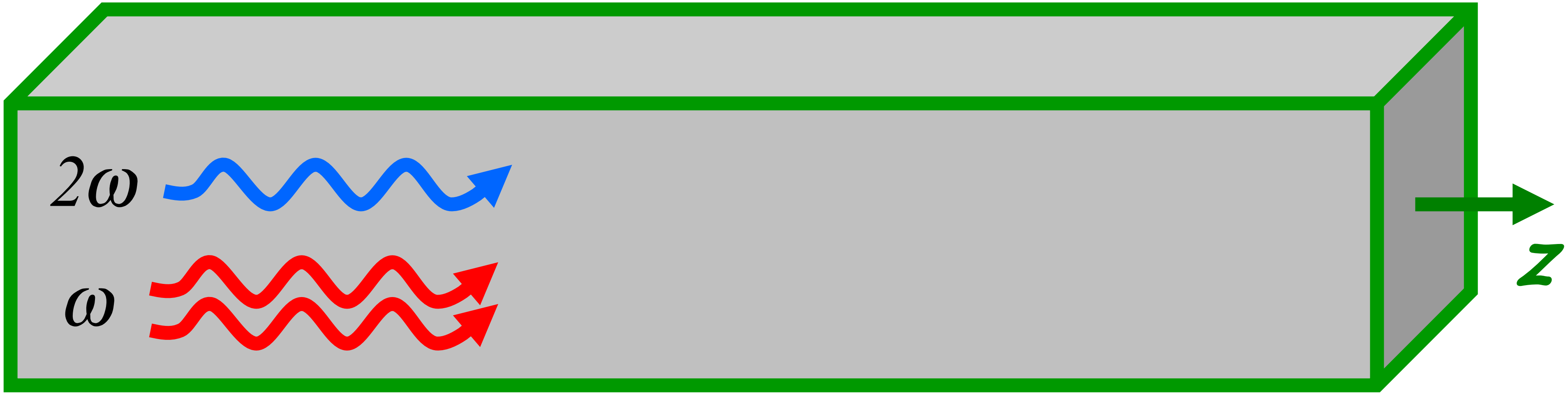


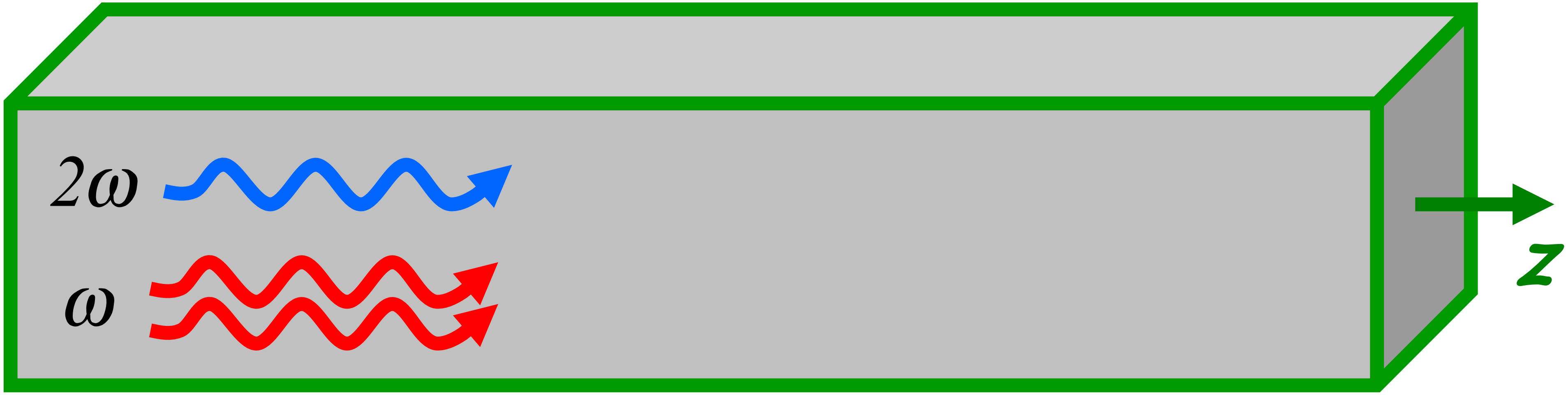
FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.


EASY Pointer

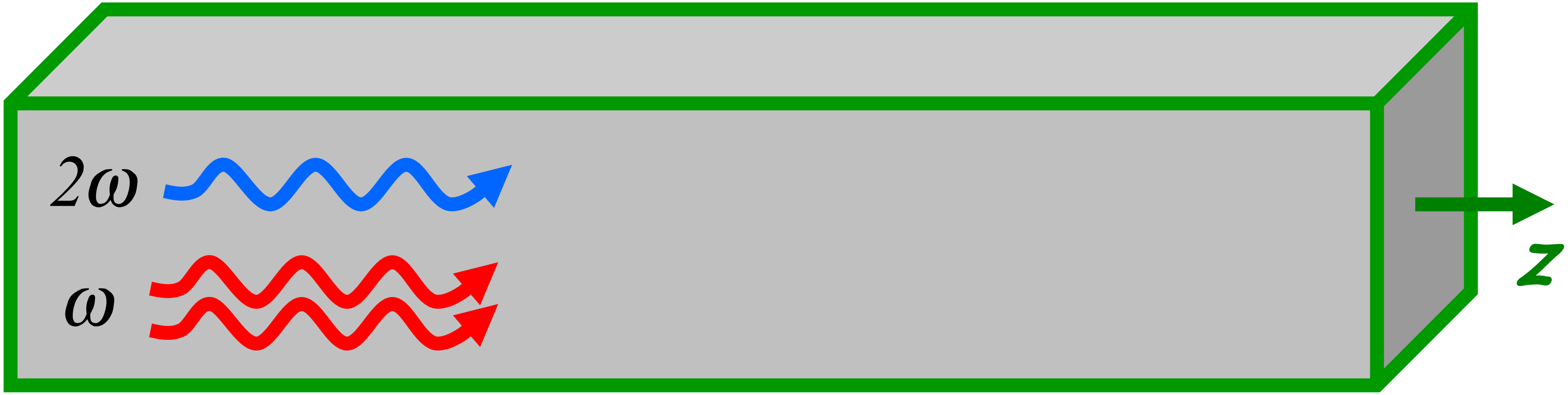
Cap
Battery
Tube
Focusing
Barrel
KTP/YAG crystal
for SHG to 532 nm
Grins lens
IR Pump Diode: 1064 nm
Pump Circuit
Micro Switch
Actuator Tab



$$P(2\omega) = \chi^{(2)} E(\omega) E(\omega)$$

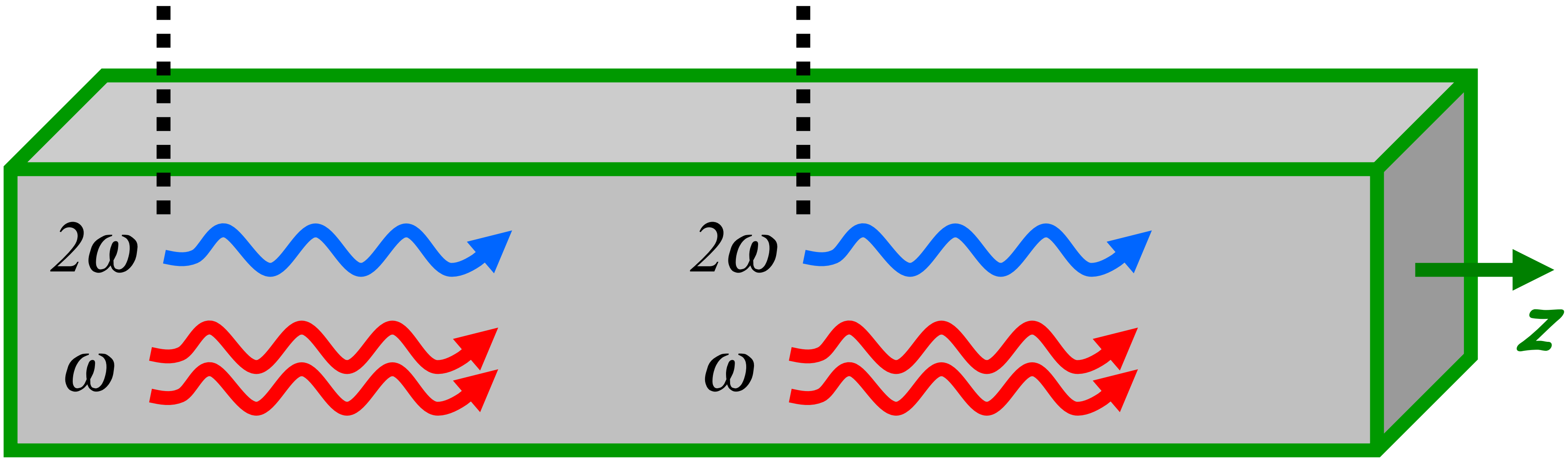


$$P(z, 2\omega) = \chi^{(2)} \left(E(\omega) e^{ik(\omega)z} \right) \left(E(\omega) e^{ik(\omega)z} \right)$$



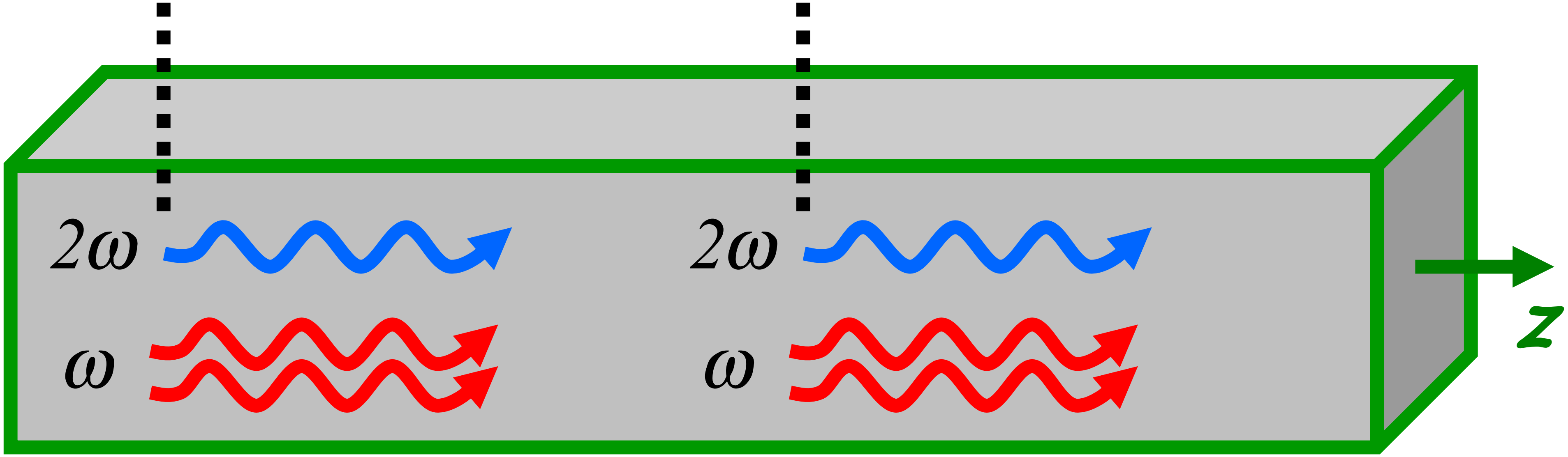
$$P(z, 2\omega) = \chi^{(2)} \left(E(\omega) e^{ik(\omega)z} \right) \left(E(\omega) e^{ik(\omega)z} \right)$$

$$k(\omega) = \frac{\omega}{c} n(\omega)$$



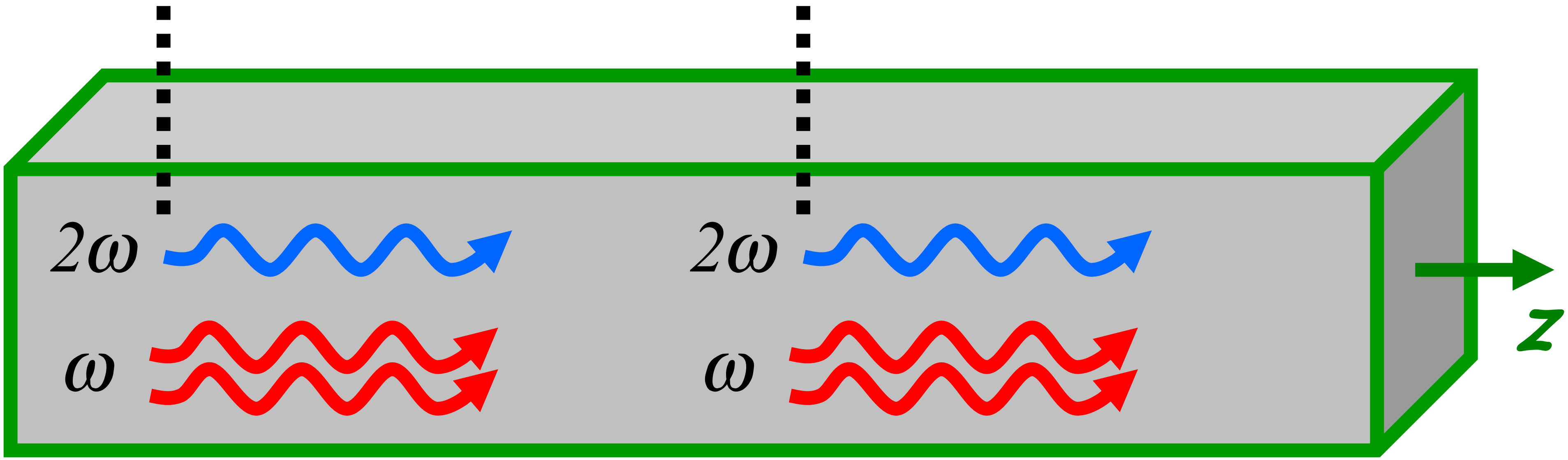
$$P(z, 2\omega) = \chi^{(2)} E(\omega) E(\omega) e^{2ik(\omega)z}$$

$$k(\omega) = \frac{\omega}{c} n(\omega)$$



$$P(z, 2\omega) = \chi^{(2)} E(\omega) E(\omega) e^{2ik(\omega)z}$$

$$k(\omega) = \frac{\omega}{c} n(\omega) \qquad k(2\omega) = \frac{2\omega}{c} n(2\omega)$$



$$P(z, 2\omega) = \chi^{(2)} E(\omega) E(\omega) e^{2ik(\omega)z}$$

$$k(\omega) = \frac{\omega}{c} n(\omega) \qquad k(2\omega) = \frac{2\omega}{c} n(2\omega)$$

for constructive interference require:

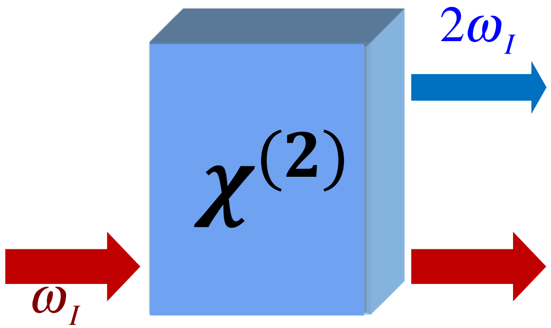
$$k(2\omega) - 2k(\omega) = \frac{2\omega}{c} n(2\omega) - 2\frac{\omega}{c} n(\omega) = 0$$

"phase matching condition"

$$E(t) = \left(E(\omega_S) e^{-i\omega_S t} + E(-\omega_S) e^{i\omega_S t} \right)$$
$$+ \left(E(\omega_I) e^{-i\omega_I t} + E(-\omega_I) e^{i\omega_I t} \right)$$

$$E(t) = \left(E(\omega_S) e^{-i\omega_S t} + E(-\omega_S) e^{i\omega_S t} \right) + \left(E(\omega_I) e^{-i\omega_I t} + E(-\omega_I) e^{i\omega_I t} \right)$$

second harmonic generation

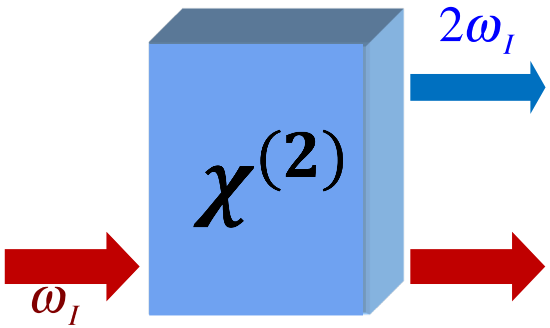


$$\omega_I + \omega_I = 2\omega_I$$

$$P(2\omega_I) = \chi^{(2)} E(\omega_I) E(\omega_I)$$

$$E(t) = \left(E(\omega_S) e^{-i\omega_S t} + E(-\omega_S) e^{i\omega_S t} \right) + \left(E(\omega_I) e^{-i\omega_I t} + E(-\omega_I) e^{i\omega_I t} \right)$$

second harmonic generation



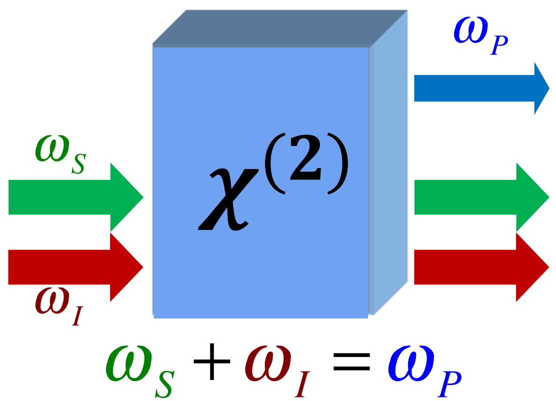
$$\omega_I + \omega_I = 2\omega_I$$

….. and similarly for ω_S

$$E(t) = \left(E(\omega_S)e^{-i\omega_S t} + E(-\omega_S)e^{i\omega_S t} \right)$$

$$+ \left(E(\omega_I)e^{-i\omega_I t} + E(-\omega_I)e^{i\omega_I t} \right)$$

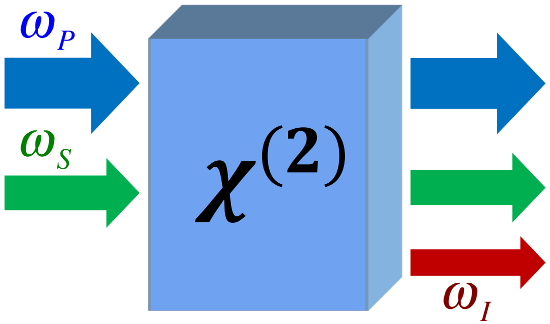
sum frequency generation



$$P(\omega_I + \omega_S) = \chi^{(2)} E(\omega_I) E(\omega_S)$$

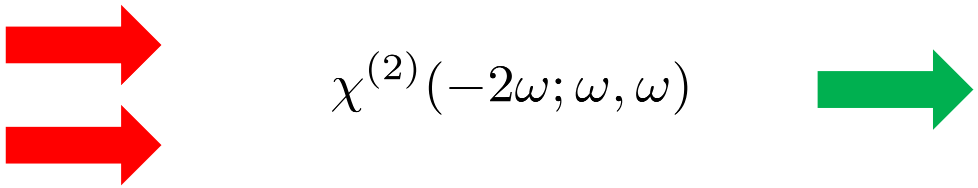
$$E(t) = \left(E(\omega_S) e^{-i\omega_S t} + E(-\omega_S) e^{i\omega_S t} \right) + \left(E(\omega_P) e^{-i\omega_P t} + E(-\omega_P) e^{i\omega_P t} \right)$$

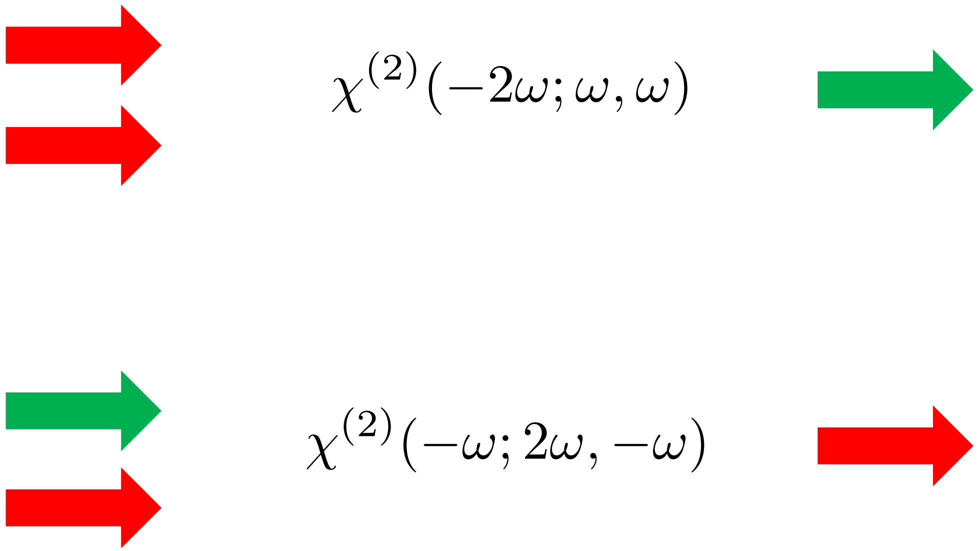
difference frequency generation

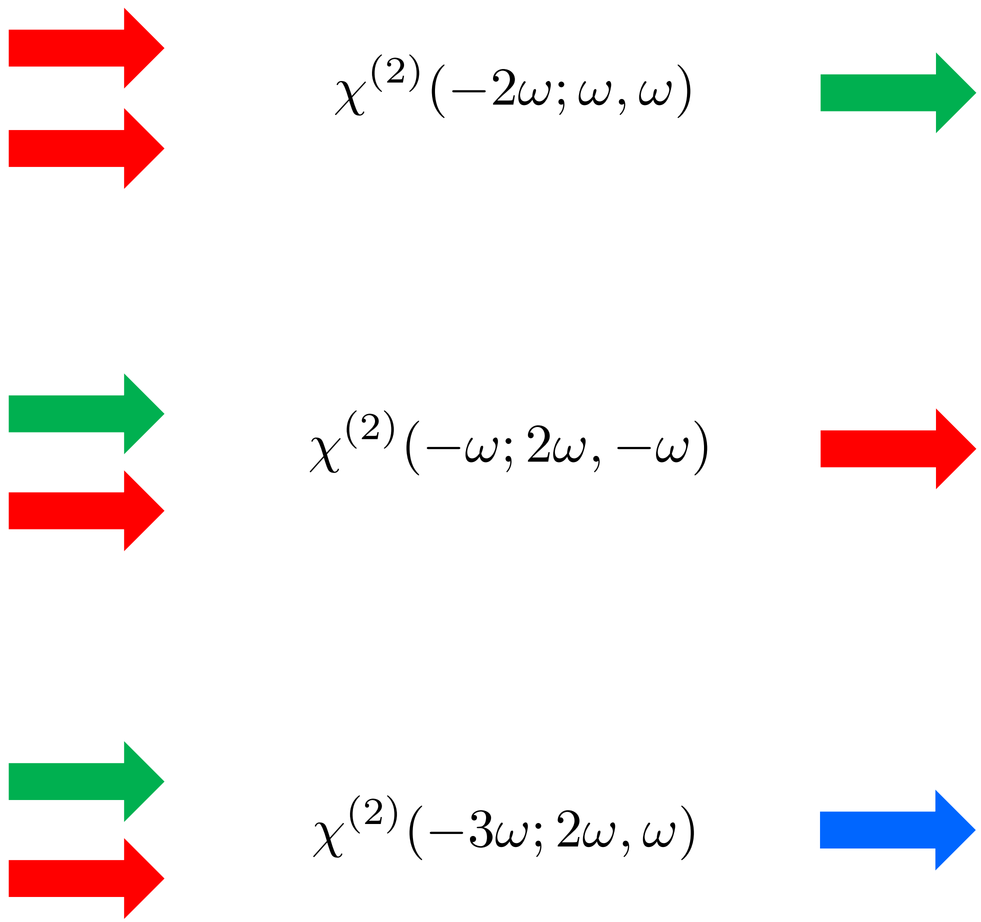


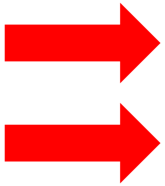
$$\omega_P - \omega_S = \omega_I$$

$$P(\omega_P - \omega_S) = \chi^{(2)} E(\omega_P) E(-\omega_S)$$

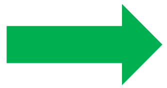

$\chi^{(2)}(-2\omega;\omega,\omega)$


$\chi^{(2)}(-2\omega;\omega,\omega)$
$\chi^{(2)}(-\omega;2\omega,-\omega)$


$\chi^{(2)}(-2\omega;\omega,\omega)$
$\chi^{(2)}(-\omega;2\omega,-\omega)$
$\chi^{(2)}(-3\omega;2\omega,\omega)$



$$\chi^{(2)}(-2\omega;\omega,\omega)$$



undepleted pump approximation

$\chi^{(2)}$ effects

sum frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1-\omega_2;\omega_1,\omega_2)$$

$\chi^{(2)}$ effects

sum frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1 - \omega_2; \omega_1, \omega_2)$$

second harmonic generation

$$\chi^{(2)}_{ijk}(-2\omega; \omega, \omega)$$

$\chi^{(2)}$ effects

sum frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1 - \omega_2; \omega_1, \omega_2)$$

second harmonic generation

$$\chi^{(2)}_{ijk}(-2\omega; \omega, \omega)$$

linear electro-optic effect (Pockels effect)

$$\chi^{(2)}_{ijk}(-\omega; \omega, 0)$$

$$P(\omega) \propto E(\omega) E_{DC}$$

$\chi^{(2)}$ effects

sum frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1-\omega_2;\omega_1,\omega_2)$$

second harmonic generation

$$\chi^{(2)}_{ijk}(-2\omega;\omega,\omega)$$

linear electro-optic effect (Pockels effect)

$$\chi^{(2)}_{ijk}(-\omega;\omega,0)$$

$$P(\omega)\ \propto\ E(\omega)E_{DC}$$

difference frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1+\omega_2;\omega_1,-\omega_2)$$

$\chi^{(2)}$ *effects*

sum frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1-\omega_2;\omega_1,\omega_2)$$

second harmonic generation

$$\chi^{(2)}_{ijk}(-2\omega;\omega,\omega)$$

linear electro-optic effect (Pockels effect)

$$\chi^{(2)}_{ijk}(-\omega;\omega,0)$$

$$P(\omega) \propto E(\omega)E_{DC}$$

difference frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1+\omega_2;\omega_1,-\omega_2)$$

optical rectification

$$\chi^{(2)}_{ijk}(0;\omega,-\omega)$$

$$P_{DC} \propto E(\omega)E(-\omega)$$

$\chi^{(2)}$ effects

sum frequency generation

$$\chi^{(2)}_{ijk}(-\omega_1 - \omega_2; \omega_1, \omega_2)$$

second harmonic generation

$$\chi^{(2)}_{ijk}(-2\omega; \omega, \omega)$$

linear electro-optic effect (Pockels effect)

$$\chi^{(2)}_{ijk}(-\omega; \omega, 0)$$

$$P(\omega) \propto E(\omega) E_{DC}$$

difference frequency generation

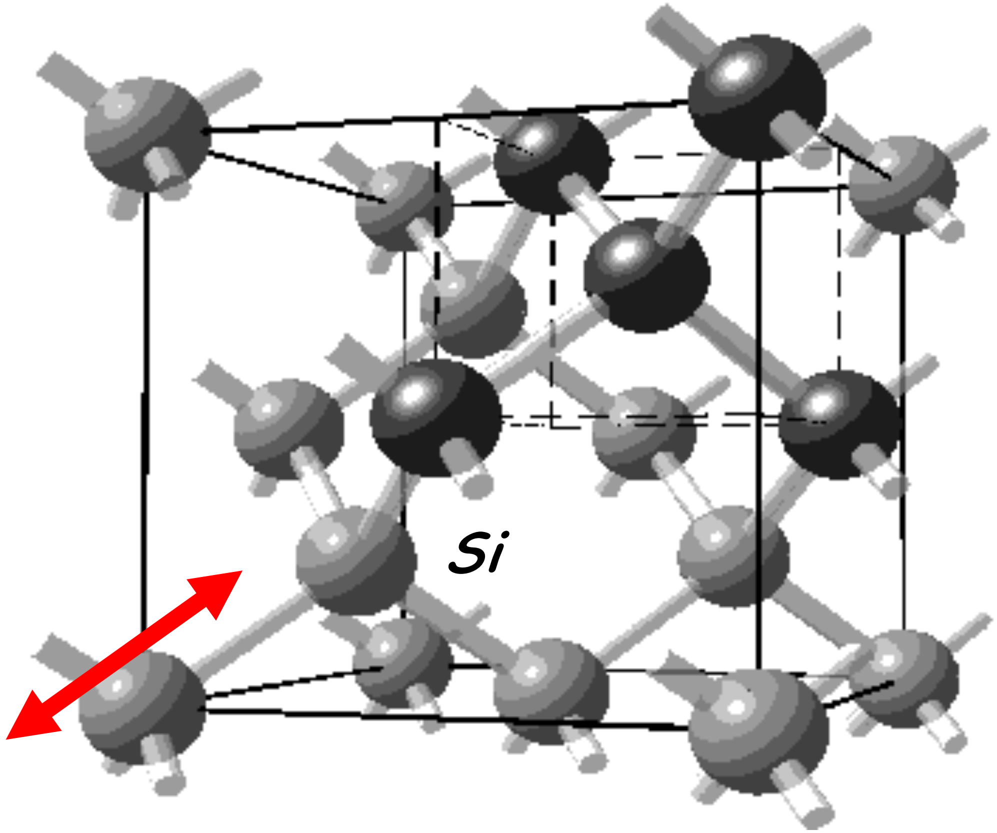
$$\chi^{(2)}_{ijk}(-\omega_1 + \omega_2; \omega_1, -\omega_2)$$

optical rectification

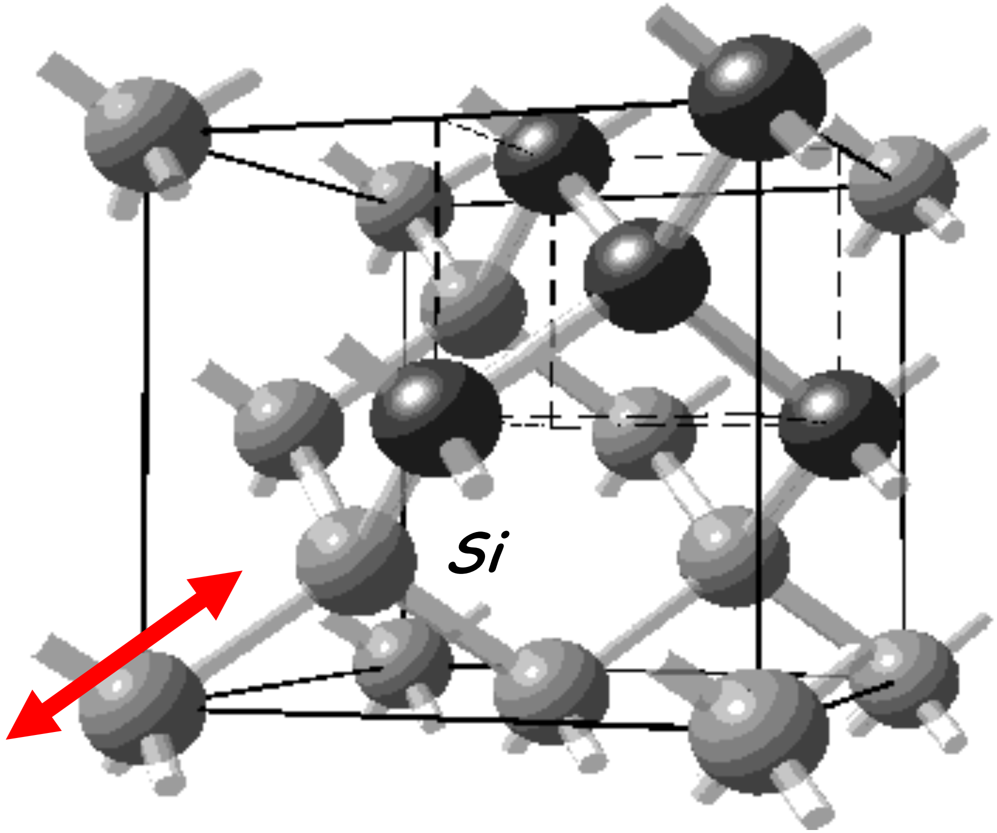
$$\chi^{(2)}_{ijk}(0; \omega, -\omega)$$

$$P_{DC} \propto |E(\omega)|^2$$

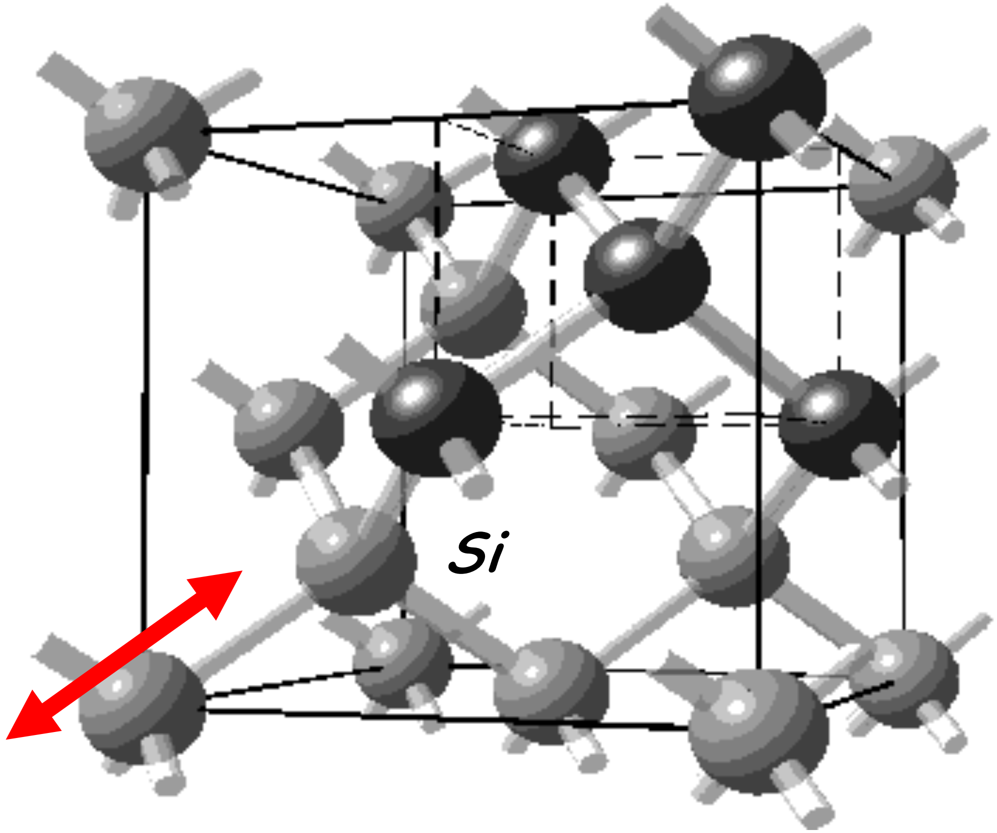
Susceptibilities
$\chi^{(2)}$ effects
$\chi^{(3)}$ effects
Quantum nonlinear optics
Nonlinear optics and electronics
Forbidden processes



$$P(2\omega) = \chi^{(2)} E(\omega) E(\omega)$$



$$P(2\omega) = \chi^{(2)} E(\omega) E(\omega)$$



$$P(2\omega) = \chi^{(2)} E(\omega)E(\omega)$$

$$P(3\omega) = \chi^{(3)} E(\omega)E(\omega)E(\omega)$$

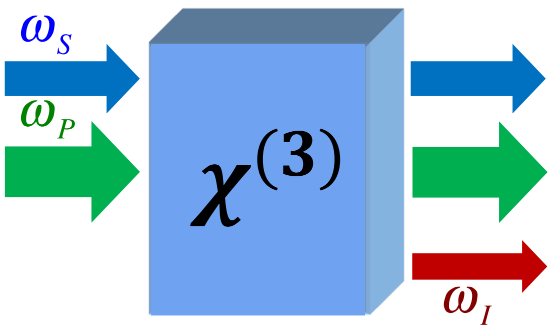
third harmonic generation

$$E(t) = \left(E(\omega_S)e^{-i\omega_S t} + E(-\omega_S)e^{i\omega_S t} \right)$$

$$+ \left(E(\omega_P)e^{-i\omega_P t} + E(-\omega_P)e^{i\omega_P t} \right)$$

$$E(t) = \left(E(\omega_S) e^{-i\omega_S t} + E(-\omega_S) e^{i\omega_S t} \right) + \left(E(\omega_P) e^{-i\omega_P t} + E(-\omega_P) e^{i\omega_P t} \right)$$

four wave mixing

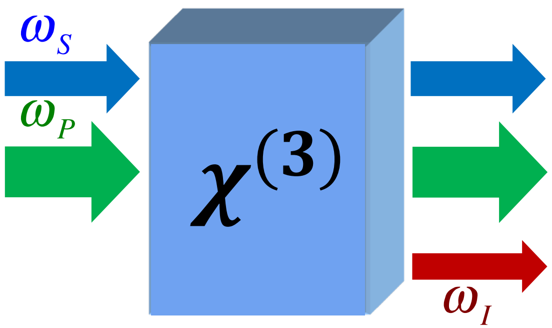


$$2\omega_P - \omega_S = \omega_I$$

$$P(2\omega_P - \omega_S) = \chi^{(3)} E(\omega_P) E(\omega_P) E(-\omega_S)$$

$$E(t) = \left(E(\omega_S)e^{-i\omega_S t} + E(-\omega_S)e^{i\omega_S t}\right) + \left(E(\omega_P)e^{-i\omega_P t} + E(-\omega_P)e^{i\omega_P t}\right)$$

four wave mixing



$$2\omega_P - \omega_S = \omega_I$$

$$P(2\omega_P - \omega_S) = \chi^{(3)} E(\omega_P)E(\omega_P)E(-\omega_S)$$

and many others!

$\chi^{(3)}$ effects

third harmonic generation

$$\chi^{(3)}_{ijkl}(-3\omega;\omega,\omega,\omega)$$

$\chi^{(3)}$ *effects*

third harmonic generation

$$\chi^{(3)}_{ijkl}(-3\omega;\omega,\omega,\omega)$$

four wave mixing

$$\chi^{(3)}_{ijkl}(-2\omega_P+\omega_S;\omega_P,\omega_P,-\omega_S)$$

$\chi^{(3)}$ effects

third harmonic generation

$$\chi^{(3)}_{ijkl}(-3\omega;\omega,\omega,\omega)$$

four wave mixing

$$\chi^{(3)}_{ijkl}(-2\omega_P+\omega_S;\omega_P,\omega_P,-\omega_S)$$

including CARS (coherent anti-Stokes Raman scattering)

$\chi^{(3)}$ *effects*

third harmonic generation

$$\chi^{(3)}_{ijkl}(-3\omega;\omega,\omega,\omega)$$

four wave mixing

$$\chi^{(3)}_{ijkl}(-2\omega_P+\omega_S;\omega_P,\omega_P,-\omega_S)$$

including CARS (coherent anti-Stokes Raman scattering)

self-phase modulation

$$\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)$$

$$P(\omega) \;\propto\; E(\omega)\,|E(\omega)|^2$$

$\chi^{(3)}$ effects

third harmonic generation

$$\chi^{(3)}_{ijkl}(-3\omega;\omega,\omega,\omega)$$

four wave mixing

$$\chi^{(3)}_{ijkl}(-2\omega_P+\omega_S;\omega_P,\omega_P,-\omega_S)$$

including CARS (coherent anti-Stokes Raman scattering)

self-phase modulation

$$\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)$$

$$P(\omega) \propto E(\omega)\,|E(\omega)|^2$$

cross-phase modulation

$$\chi^{(3)}_{ijkl}(-\omega_1;\omega_1,\omega_2,-\omega_2)$$

$$P(\omega_1) \propto E(\omega_1)\,|E(\omega_2)|^2$$

$\chi^{(3)}$ effects

third harmonic generation

$$\chi^{(3)}_{ijkl}(-3\omega;\omega,\omega,\omega)$$

four wave mixing

$$\chi^{(3)}_{ijkl}(-2\omega_P+\omega_S;\omega_P,\omega_P,-\omega_S)$$

including CARS
(coherent anti-Stokes
Raman scattering)

self-phase modulation

$$\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)$$

$$P(\omega) \;\propto\; E(\omega)\,|E(\omega)|^2$$

cross-phase modulation

$$\chi^{(3)}_{ijkl}(-\omega_1;\omega_1,\omega_2,-\omega_2)$$

including SRS
(stimulated Raman
scattering)

$\chi^{(3)}$ effects

third harmonic generation

$$\chi^{(3)}_{ijkl}(-3\omega;\omega,\omega,\omega)$$

four wave mixing

$$\chi^{(3)}_{ijkl}(-2\omega_P+\omega_S;\omega_P,\omega_P,-\omega_S)$$

including CARS (coherent anti-Stokes Raman scattering)

self-phase modulation

$$\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)$$

$$P(\omega) \;\propto\; E(\omega)\,|E(\omega)|^2$$

cross-phase modulation

$$\chi^{(3)}_{ijkl}(-\omega_1;\omega_1,\omega_2,-\omega_2)$$

including SRS (stimulated Raman scattering)

electro-optic effect (Kerr effect)

$$\chi^{(3)}_{ijkl}(-\omega;\omega,0,0)$$

$$P(\omega) \;\propto\; E(\omega)E^2_{DC}$$

Susceptibilities

$\chi^{(2)}$ *effects*

$\chi^{(3)}$ *effects*

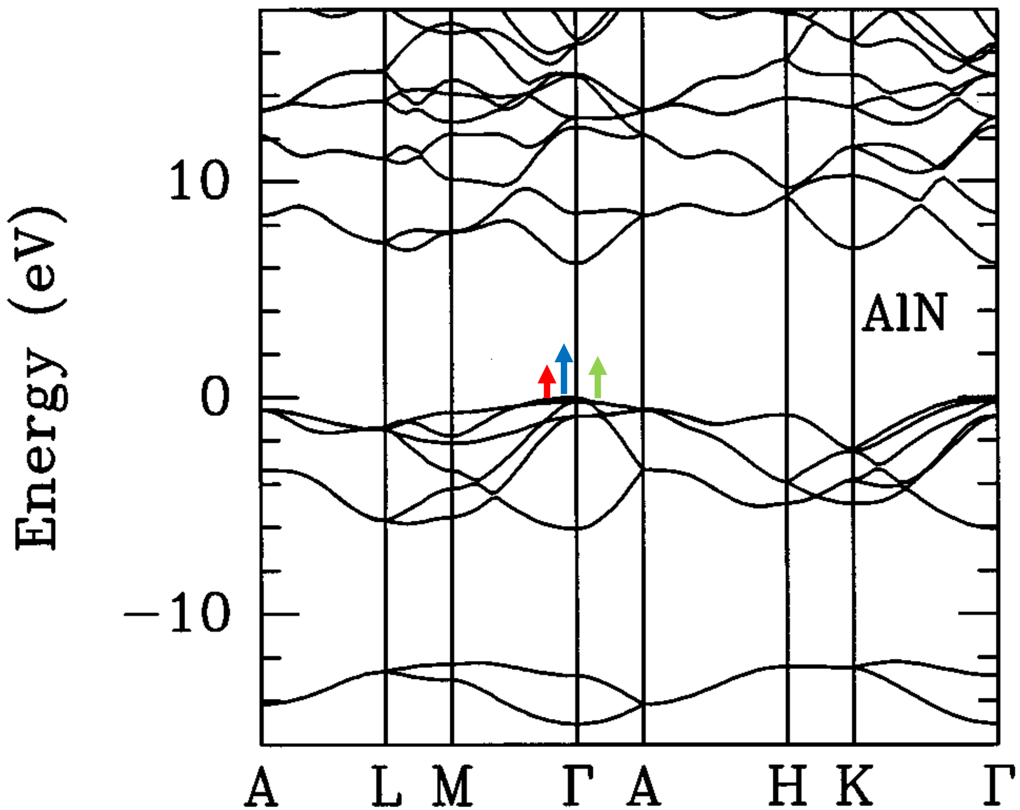
Quantum nonlinear optics

Nonlinear optics and electronics

Forbidden processes

$\hbar\omega_3$

$\hbar\omega_1$ $\hbar\omega_2$



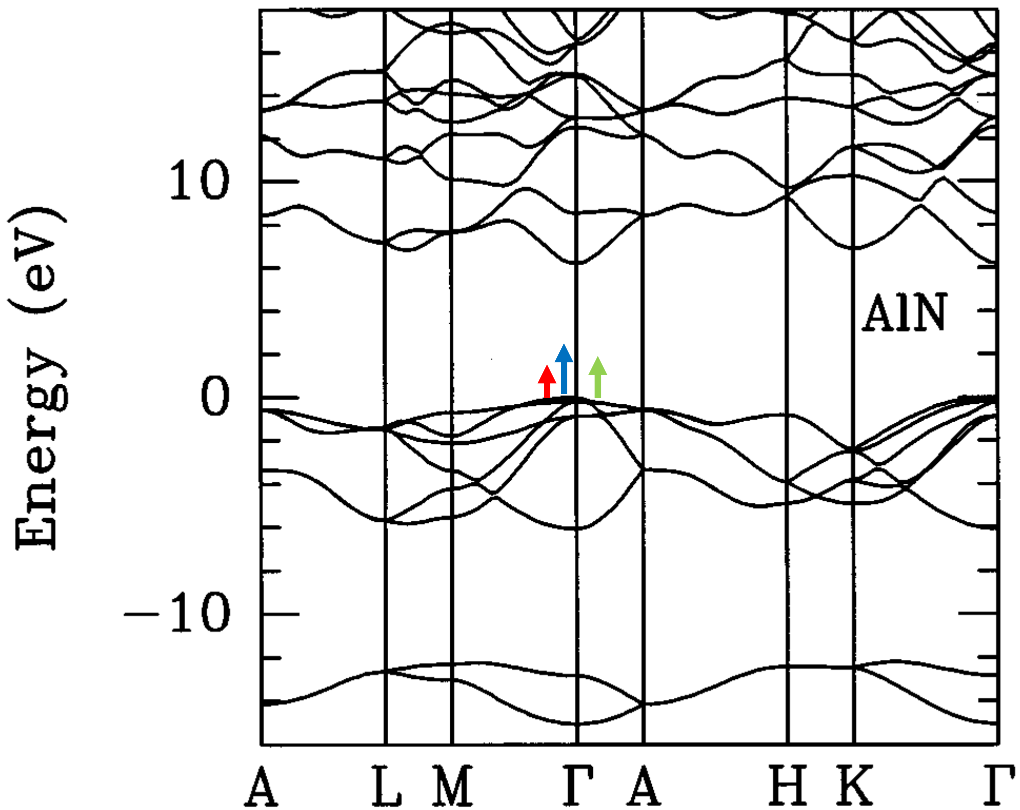
$\hbar\omega_3$

$\hbar\omega_1$ $\hbar\omega_2$

If

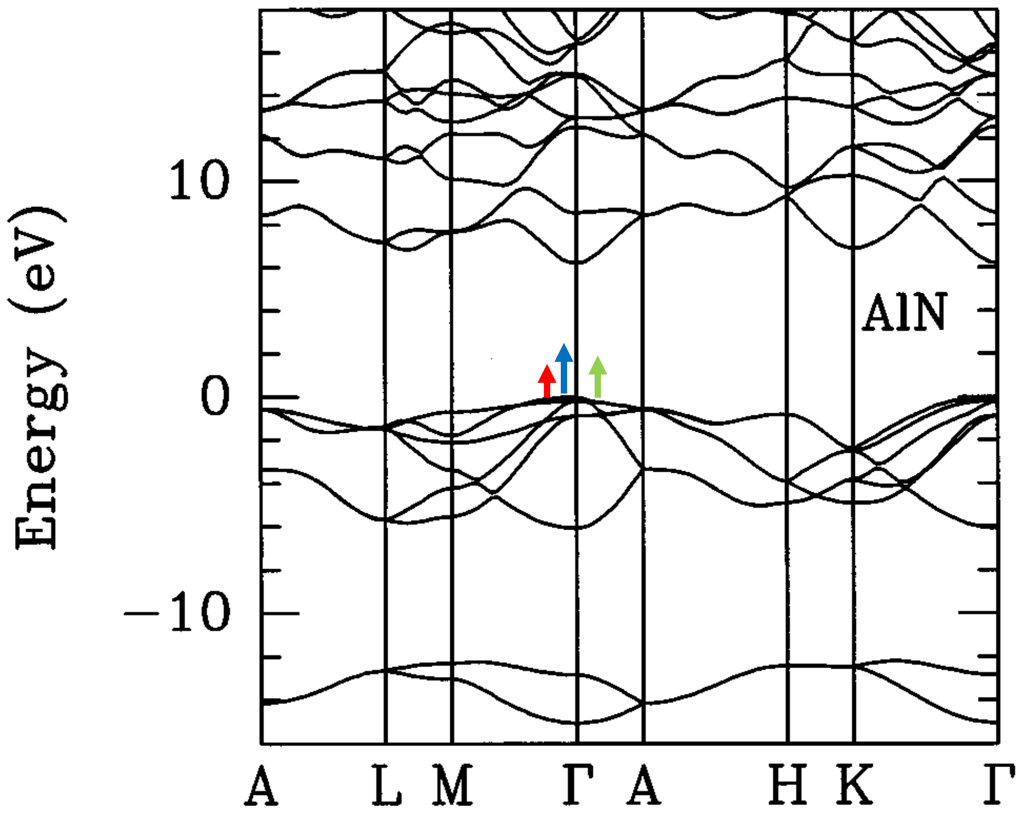
$$\hbar\omega_i \ll E_{gap}$$

$$\sum_i \hbar\omega_i \ll E_{gap}$$



$\hbar\omega_3$

$\hbar\omega_1$ $\hbar\omega_2$



If

$$\hbar\omega_i \ll E_{gap}$$

$$\sum_i \hbar\omega_i \ll E_{gap}$$

$$\chi^{(2)}_{ijk}(-\omega_1-\omega_2;\omega_1,\omega_2) \Rightarrow \chi^{(2)}_{ijk}$$

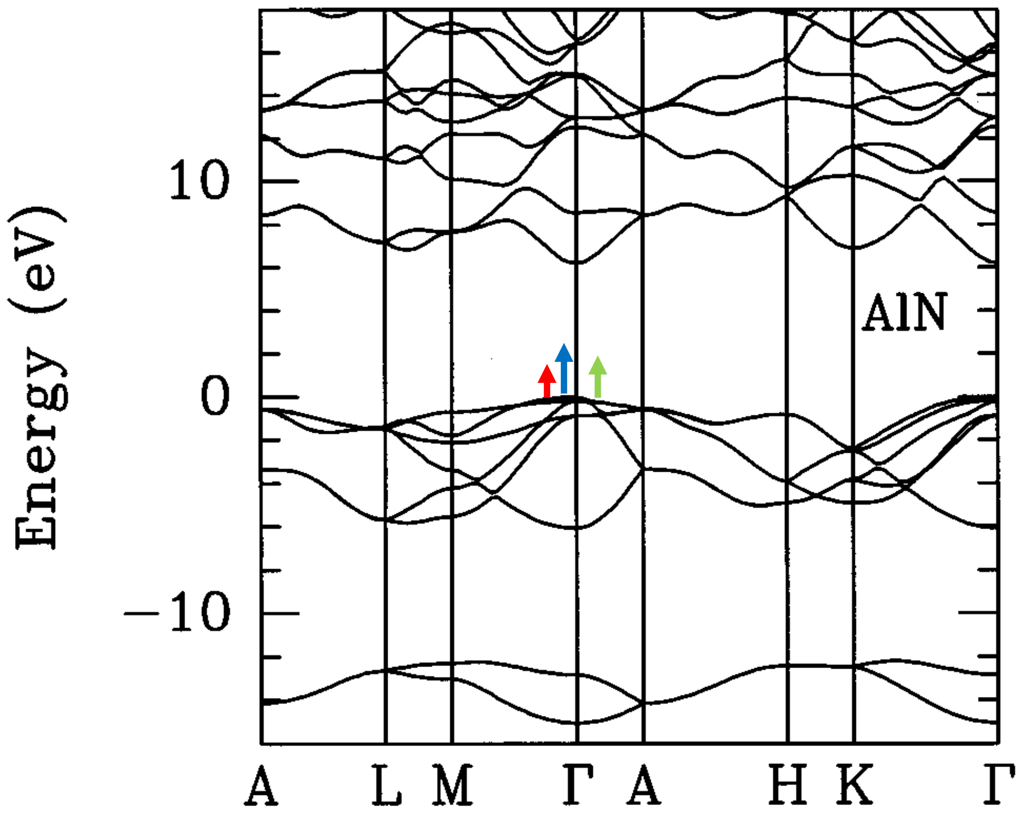
$$\chi^{(3)}_{ijkl}(-\omega_1-\omega_2-\omega_3;\omega_1,\omega_2,\omega_3) \Rightarrow \chi^{(3)}_{ijkl}$$

independent of order of Cartesian indices

real

$n(\omega_i)$ real

Quantum nonlinear optics



If

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$$\sum_i \hbar\omega_i \ll E_{gap}$$

$$\chi^{(2)}_{ijk}(-\omega_1-\omega_2;\omega_1,\omega_2) \Rightarrow \chi^{(2)}_{ijk}$$

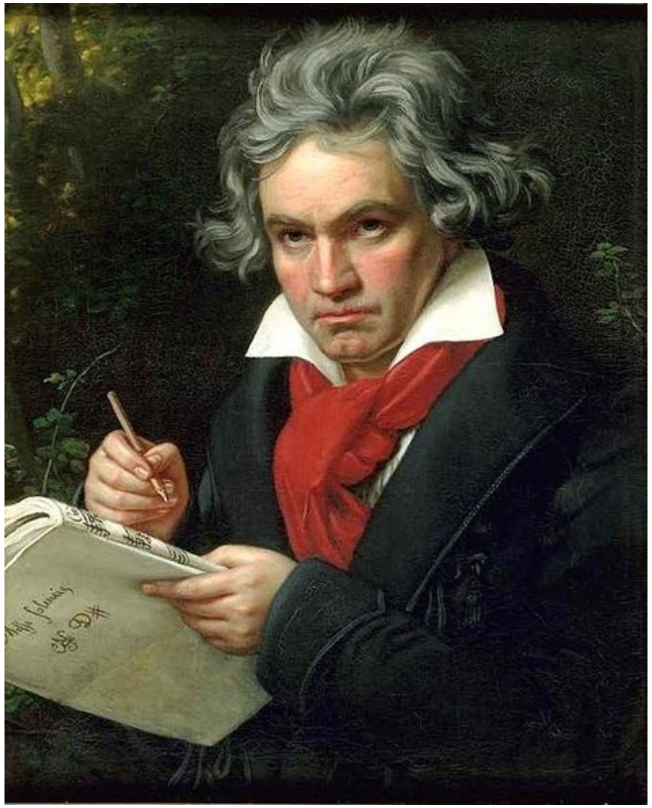
$$\chi^{(3)}_{ijkl}(-\omega_1-\omega_2-\omega_3;\omega_1,\omega_2,\omega_3) \Rightarrow \chi^{(3)}_{ijkl}$$

independent of order of Cartesian indices

real

$n(\omega_i)$ real

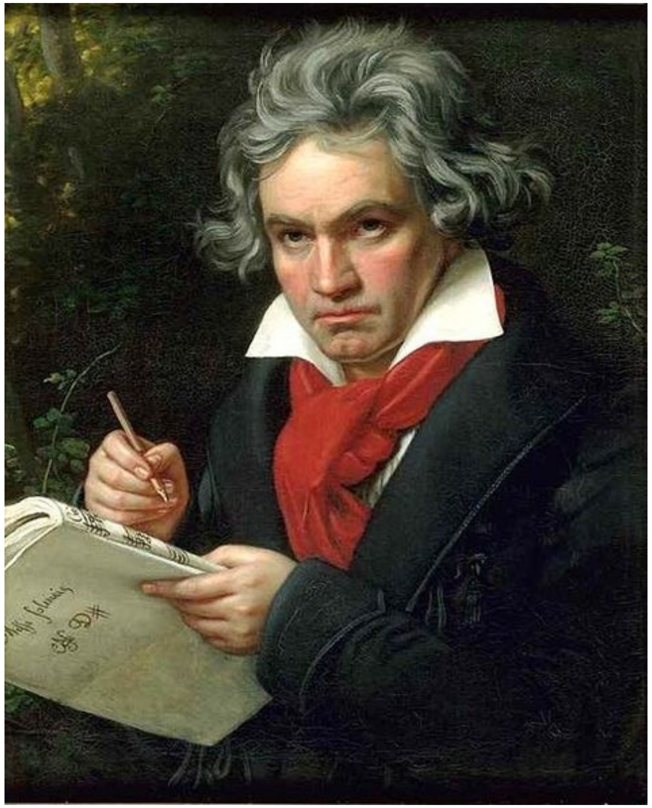
Classical nonlinear optics



nonlinear polarizations,
envelope functions,
coupled mode equations,...

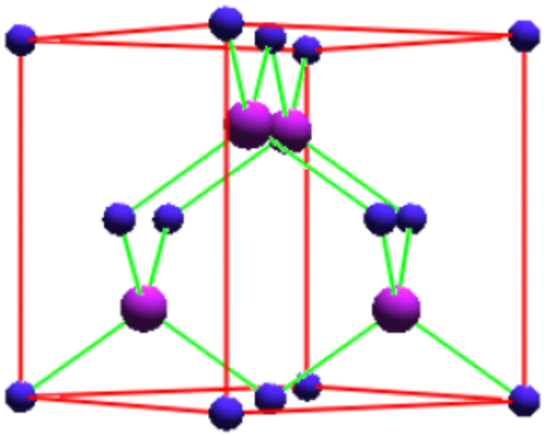
Quantum nonlinear optics

Classical nonlinear optics

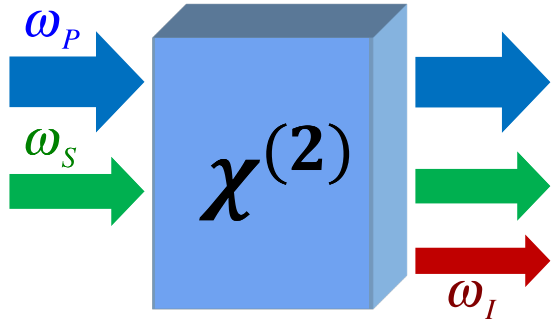


Hamiltonians, raising and lowering operators, Wigner functions,...

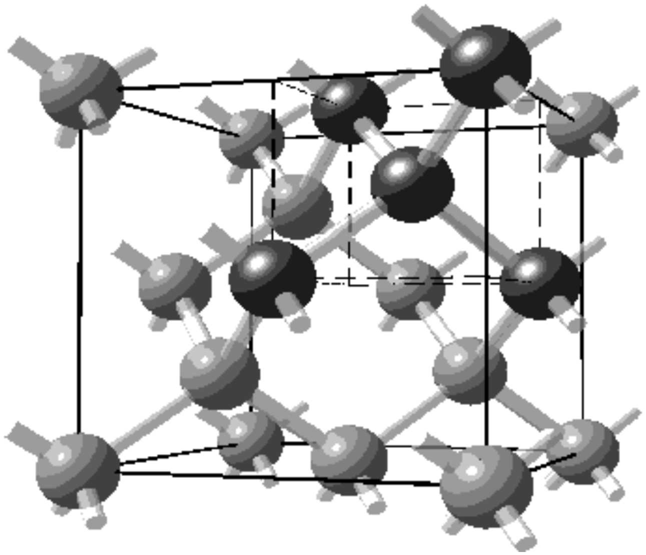
nonlinear polarizations, envelope functions, coupled mode equations,...



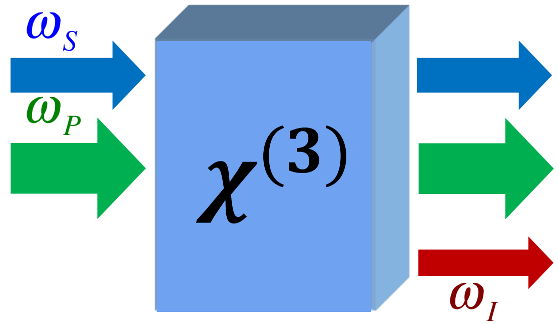
difference frequency generation



$$\omega_P - \omega_S = \omega_I$$

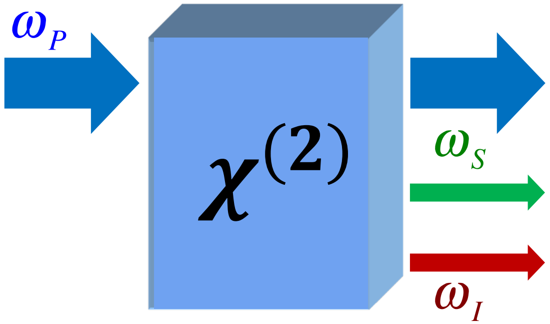


four wave mixing



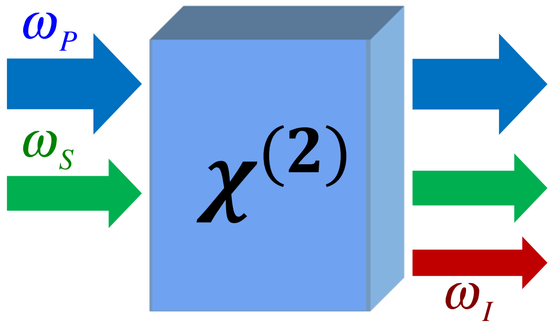
$$2\omega_P - \omega_S = \omega_I$$

spontaneous parametric down conversion



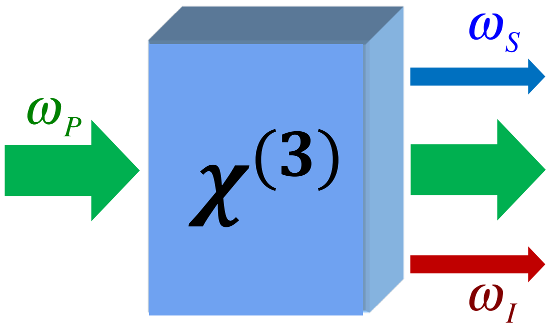
$$\omega_P = \omega_S + \omega_I$$

difference frequency generation



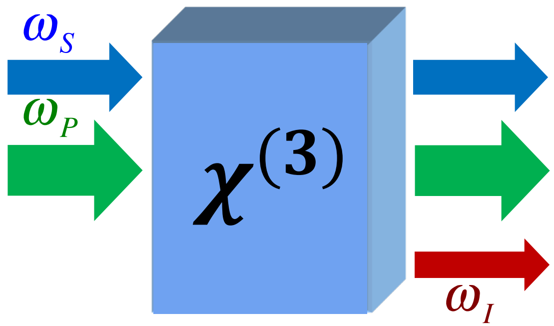
$$\omega_P - \omega_S = \omega_I$$

spontaneous four wave mixing



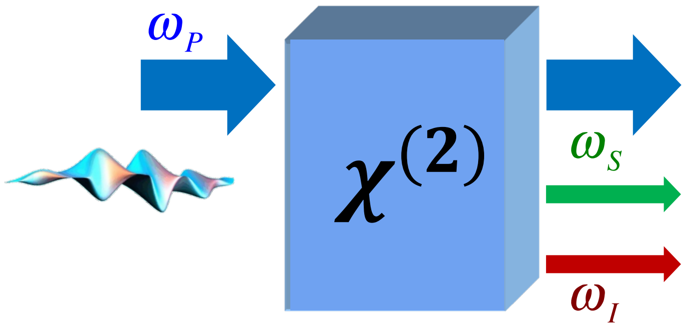
$$2\omega_P = \omega_S + \omega_I$$

four wave mixing



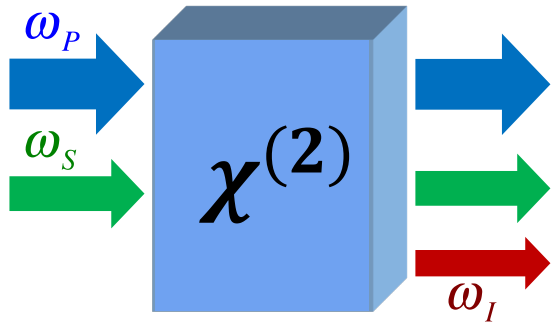
$$2\omega_P - \omega_S = \omega_I$$

spontaneous parametric down conversion



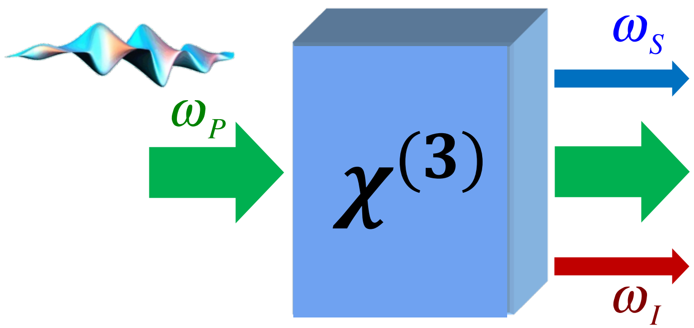
$$\omega_P = \omega_S + \omega_I$$

difference frequency generation



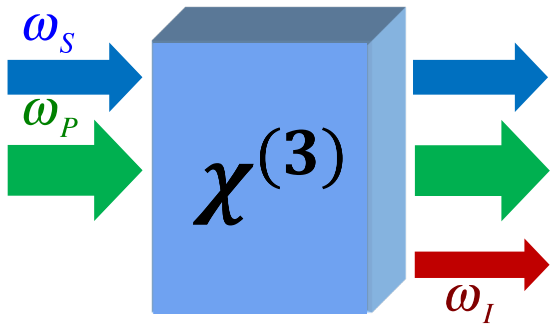
$$\omega_P - \omega_S = \omega_I$$

spontaneous four wave mixing



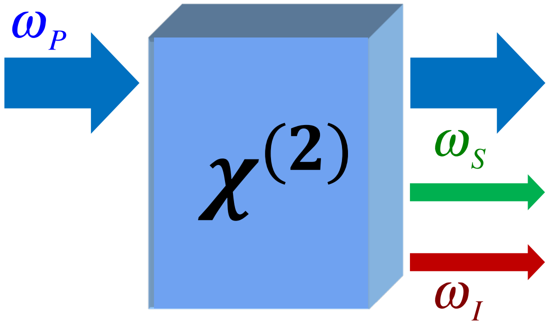
$$2\omega_P = \omega_S + \omega_I$$

four wave mixing



$$2\omega_P - \omega_S = \omega_I$$

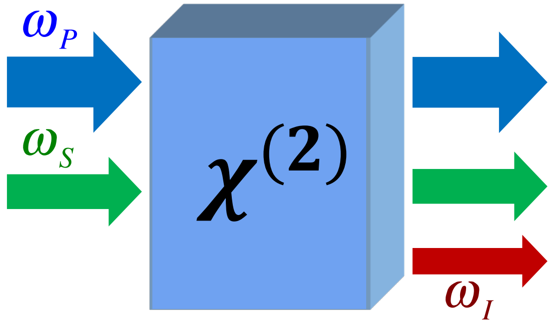
spontaneous parametric down conversion



$$\omega_P = \omega_S + \omega_I$$

$$a_P a_S^\dagger a_I^\dagger$$

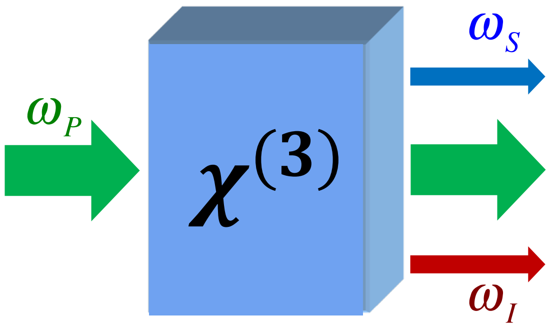
difference frequency generation



$$\omega_P - \omega_S = \omega_I$$

$$a_P a_S^\dagger a_I^\dagger$$

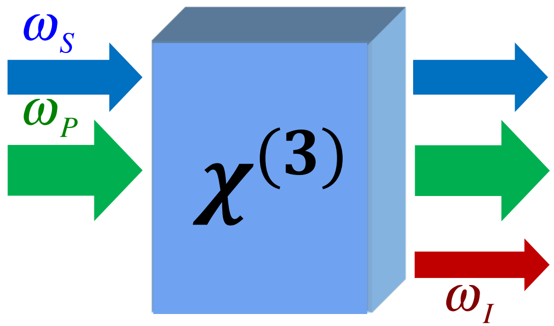
spontaneous four wave mixing



$$2\omega_P = \omega_S + \omega_I$$

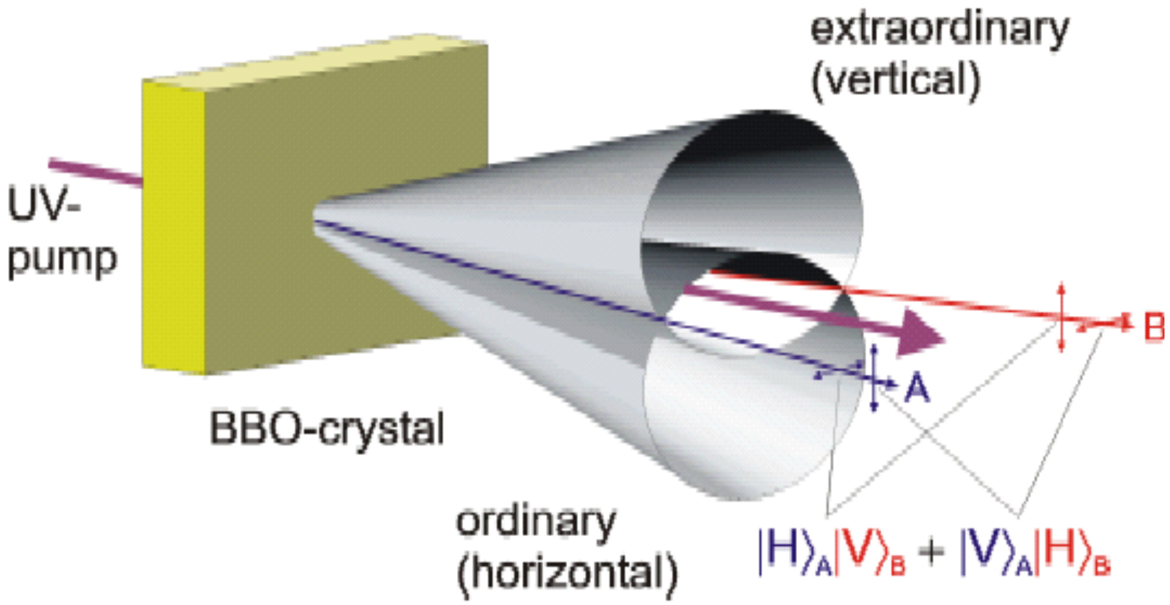
$$a_P a_P a_S^\dagger a_I^\dagger$$

four wave mixing



$$2\omega_P - \omega_S = \omega_I$$

$$a_P a_P a_S^\dagger a_I^\dagger$$

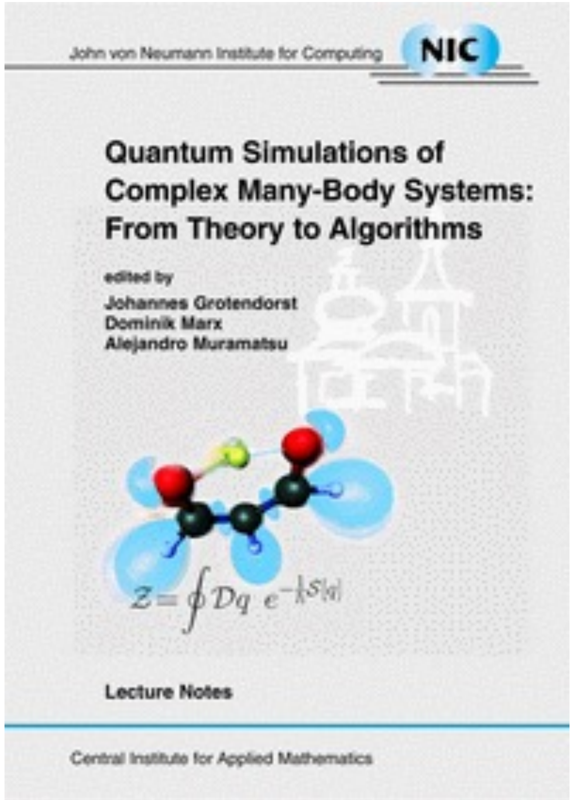


Kwiat et al. PRL 75 4337 (1995)

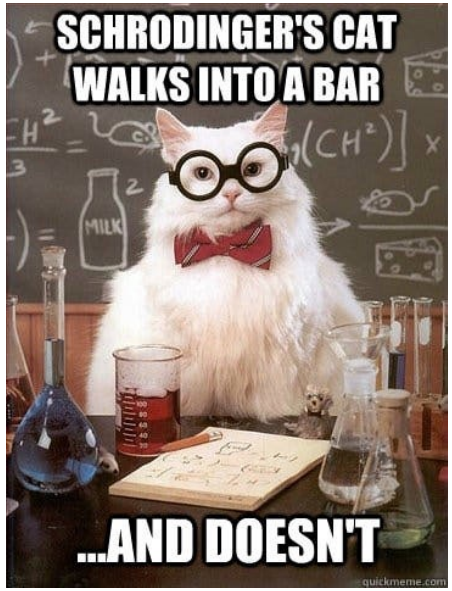
$$|\psi\rangle = \frac{1}{\sqrt{2}}\left(|H\rangle_A |V\rangle_B + |V\rangle_A |H\rangle_B\right)$$

$$|\psi\rangle \neq |\gamma\rangle_A |\sigma\rangle_B$$

quantum information processing



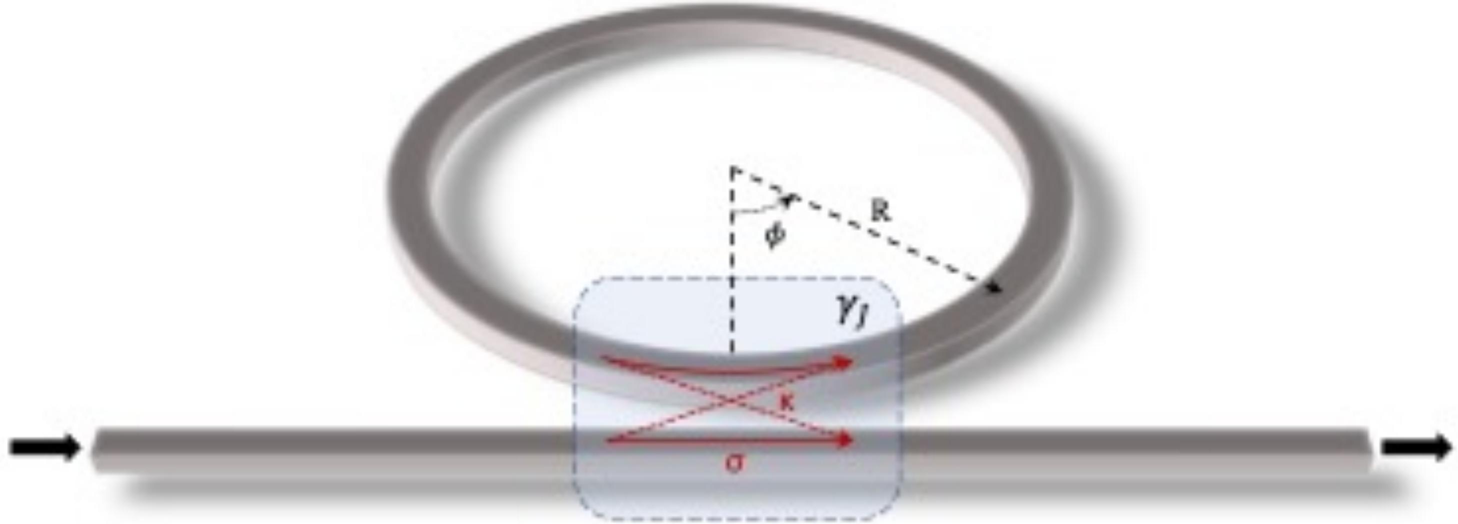
quantum simulation /computing



quantum mechanics

XANADU

Photonic Quantum Computing



"Beyond photon pairs: nonlinear quantum photonics in the high-gain regime"
Advances in Optics and Photonics 14, 291 (2022)

Susceptibilities

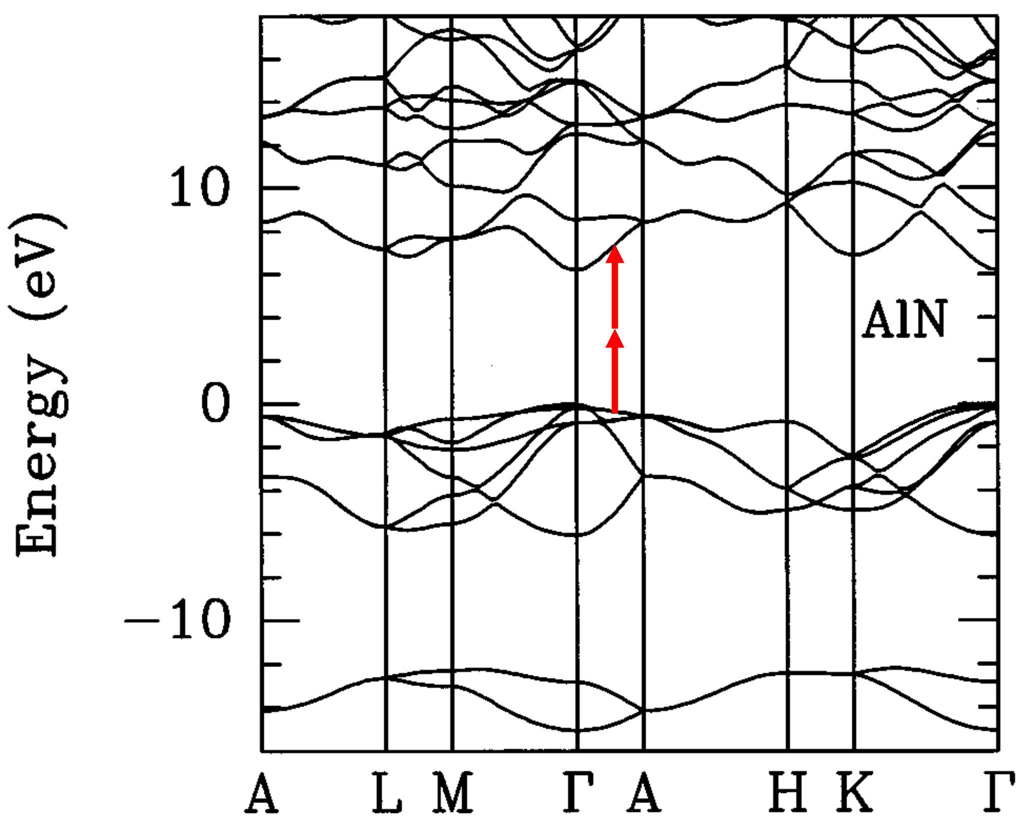
$\chi^{(2)}$ effects

$\chi^{(3)}$ effects

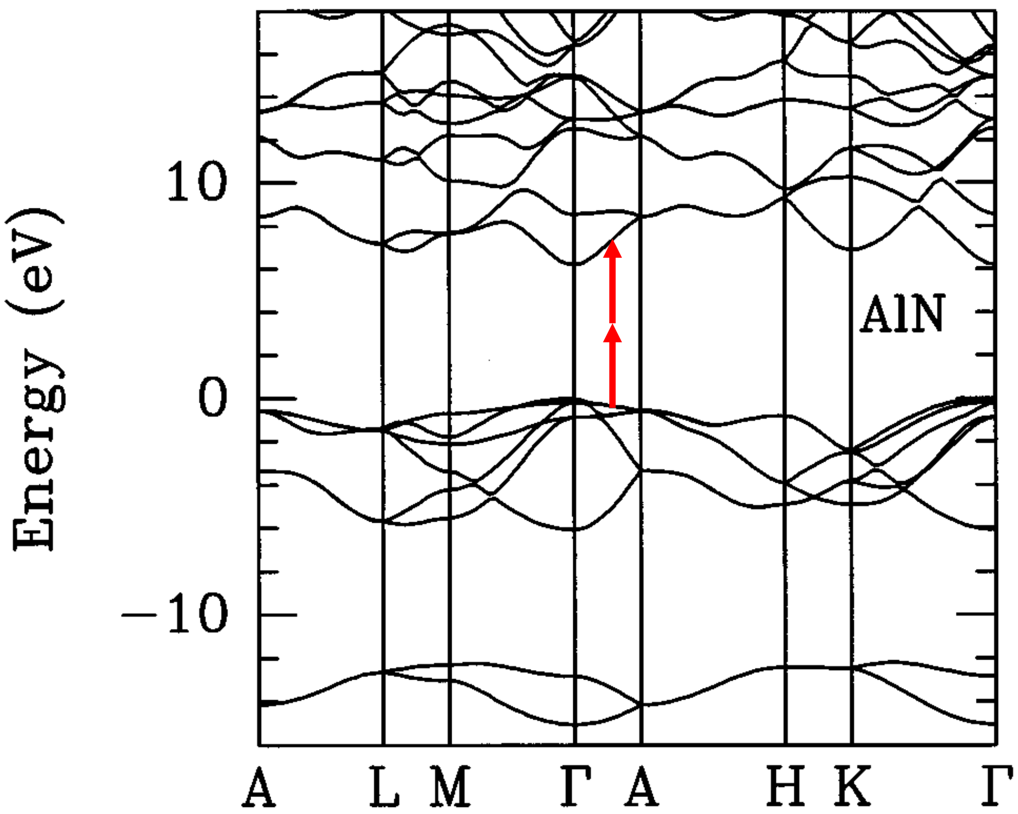
Quantum nonlinear optics

Nonlinear optics and electronics

Forbidden processes

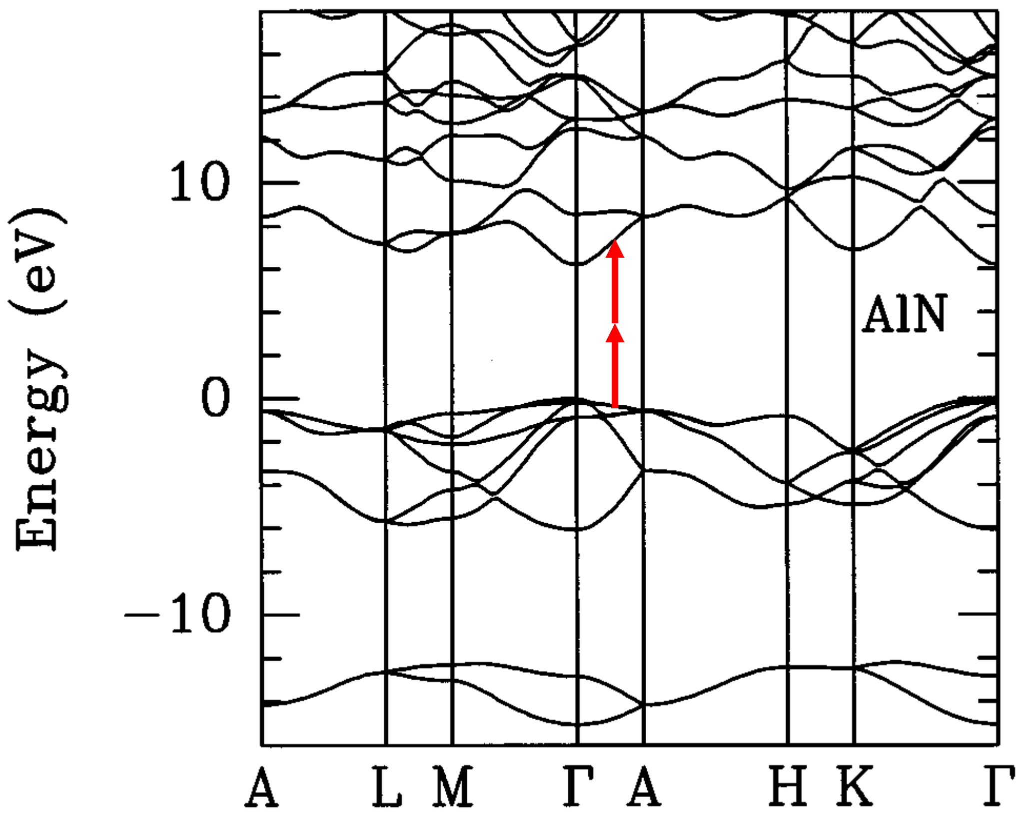


Energy (eV)
10
0
−10
AlN
A
L
M
Γ
A
H
K
Γ



$\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)$

acquires an imaginary part



$$\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)$$

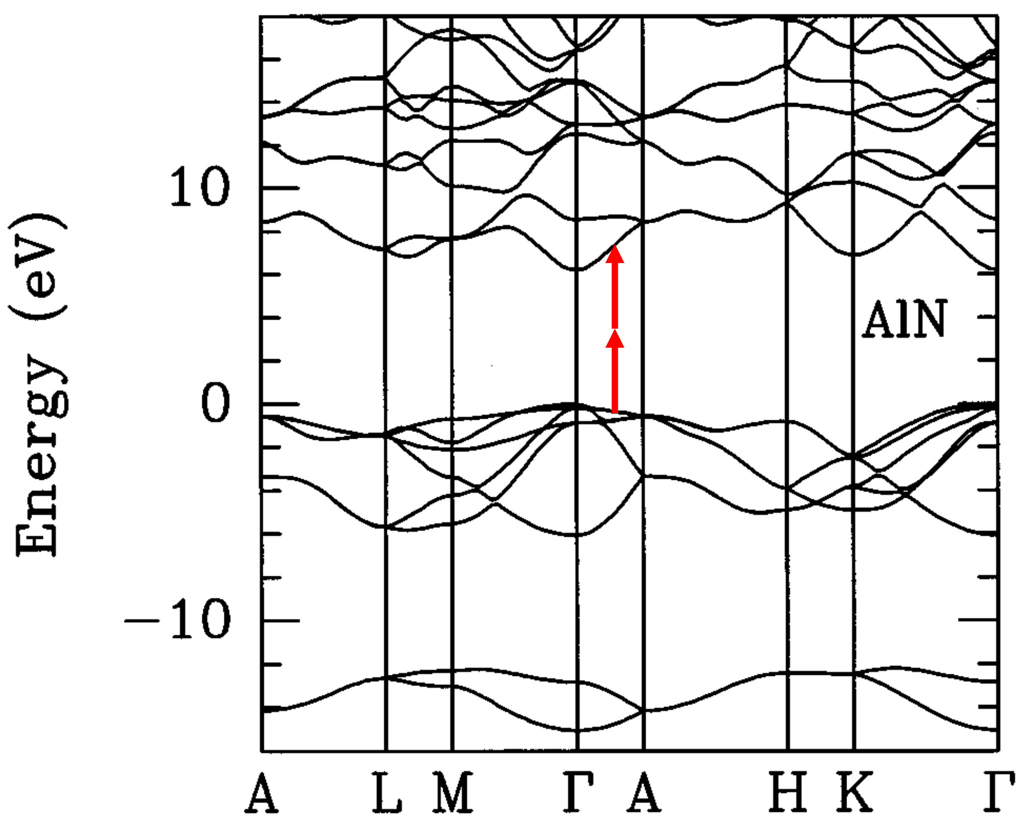
acquires an imaginary part

$$P(\omega) \;\propto\; E(\omega)\,|E(\omega)|^2$$

$$Im\left(\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)\right)$$

⇕

two-photon absorption



$$Im\left(\chi^{(1)}_{ij}(-\omega,\omega)\right)$$

one-photon absorption

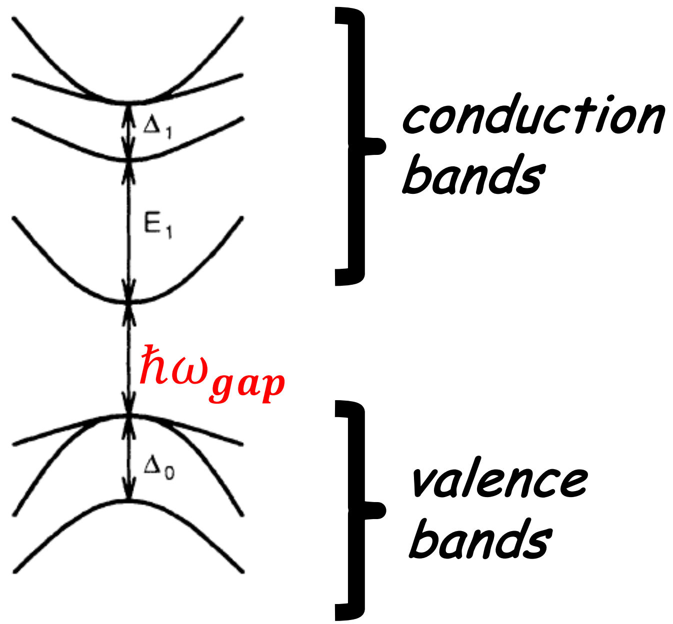
$$\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)$$

acquires an imaginary part

$$P(\omega) \;\propto\; E(\omega)\,|E(\omega)|^2$$

$$Im\left(\chi^{(3)}_{ijkl}(-\omega;\omega,\omega,-\omega)\right)$$

two-photon absorption

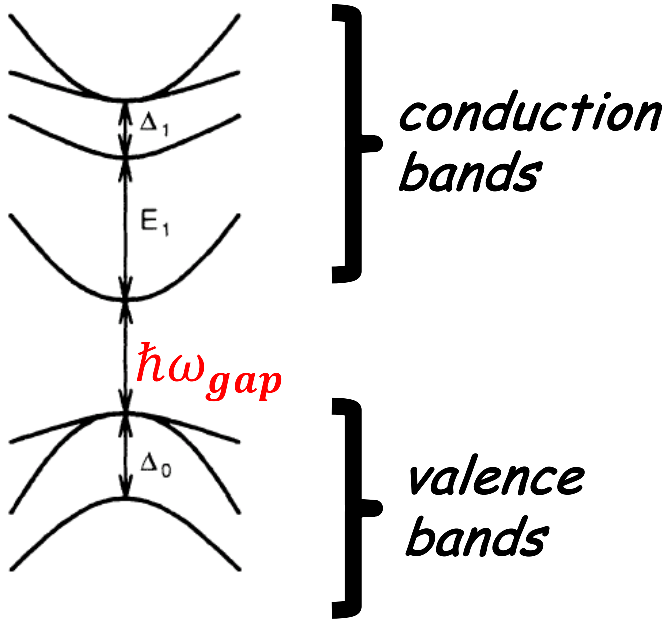


$$\chi^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) =$$

$$\overline{\chi}^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) + \frac{\sigma^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)} + \frac{\eta^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)^2}$$

with

$$\omega_\Sigma = \omega_1 + \omega_2$$



$$\chi^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) =$$

$$\overline{\chi}^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) + \frac{\sigma^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)} + \frac{\eta^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)^2}$$

with

$$\omega_\Sigma = \omega_1 + \omega_2$$

vanish if

$$|\omega_1|\,,|\omega_2| < \omega_{gap}$$

well-behaved at all frequencies

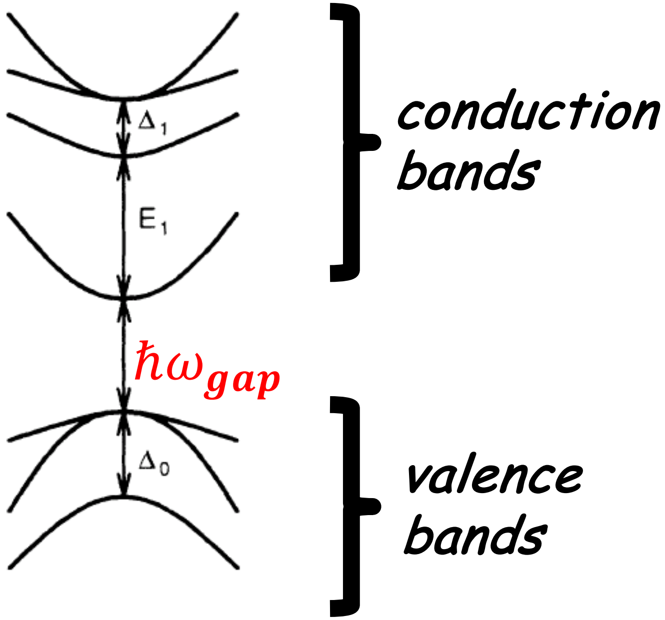
$$\chi^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) = \overline{\chi}^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) + \frac{\sigma^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)} + \frac{\eta^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)^2}$$

with

$\omega_\Sigma = \omega_1 + \omega_2$

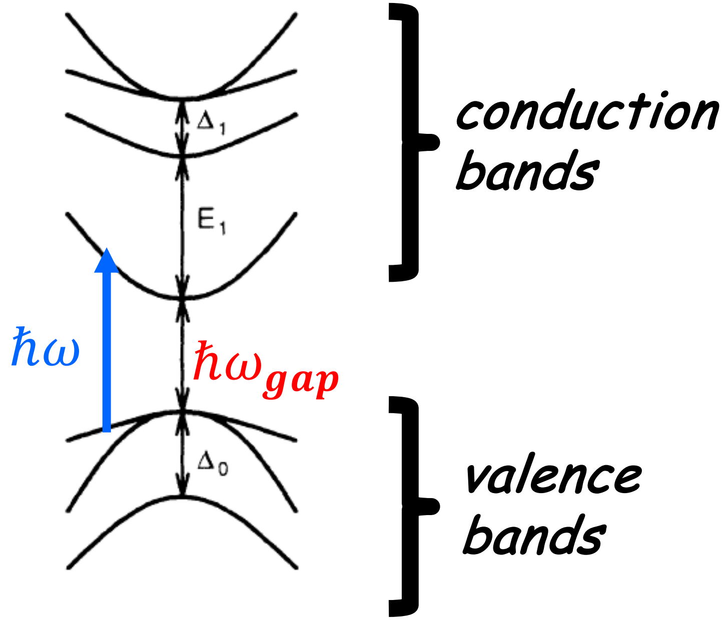
vanish if

$|\omega_1|, |\omega_2| < \omega_{gap}$



Suppose

$$\omega_1 = \omega > \omega_{gap}$$
$$\omega_2 = -\omega$$



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with

$$\omega_\Sigma = \omega_1 + \omega_2$$

Suppose

$$\omega_1 = \omega > \omega_{gap}$$
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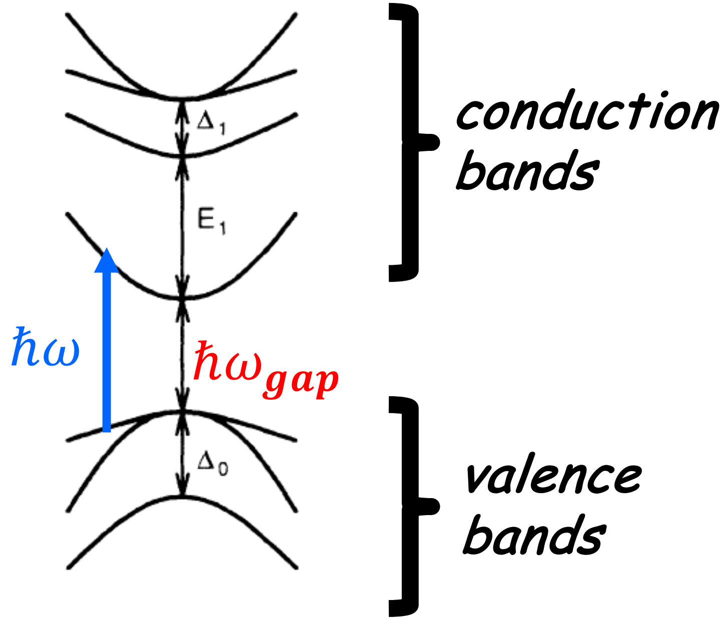

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with

$$\omega_\Sigma = \omega_1 + \omega_2$$

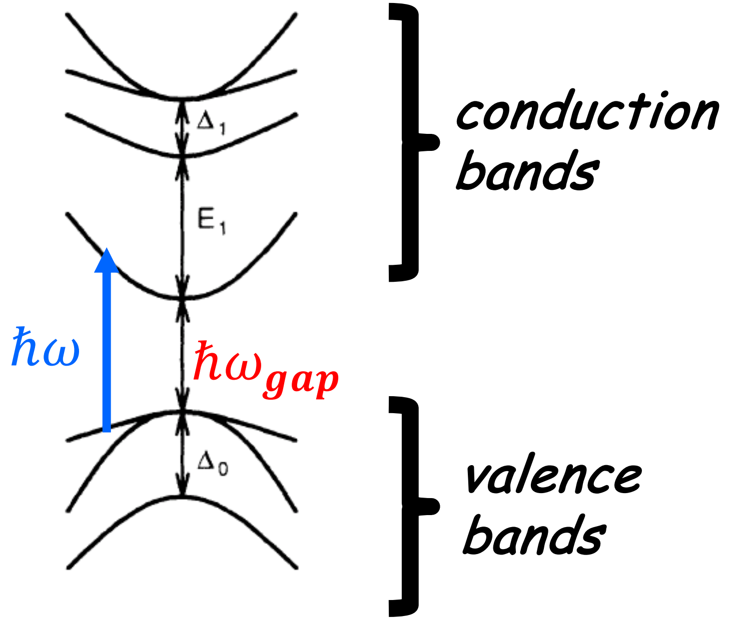
$$\omega_\Sigma \to 0$$

$$\overline{\chi}^{(2)}_{ijk}(0; \omega, -\omega)$$

optical rectification

Suppose

$$\omega_1 = \omega > \omega_{gap}$$
$$\omega_2 = -\omega$$



$$\chi^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) =$$

$$\overline{\chi}^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) + \frac{\sigma^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)} + \frac{\eta^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)^2}$$

?

with

$$\omega_\Sigma = \omega_1 + \omega_2$$

$$\omega_\Sigma \to 0$$

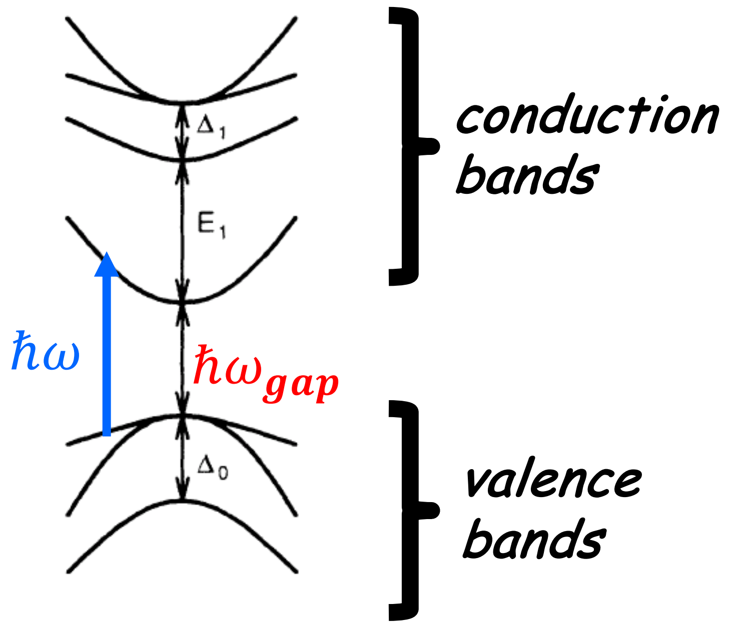
$$\overline{\chi}^{(2)}_{ijk}(0;\omega,-\omega)$$

optical rectification

these terms seem to diverge!

Suppose

$$\omega_1 = \omega > \omega_{gap}$$
$$\omega_2 = -\omega$$



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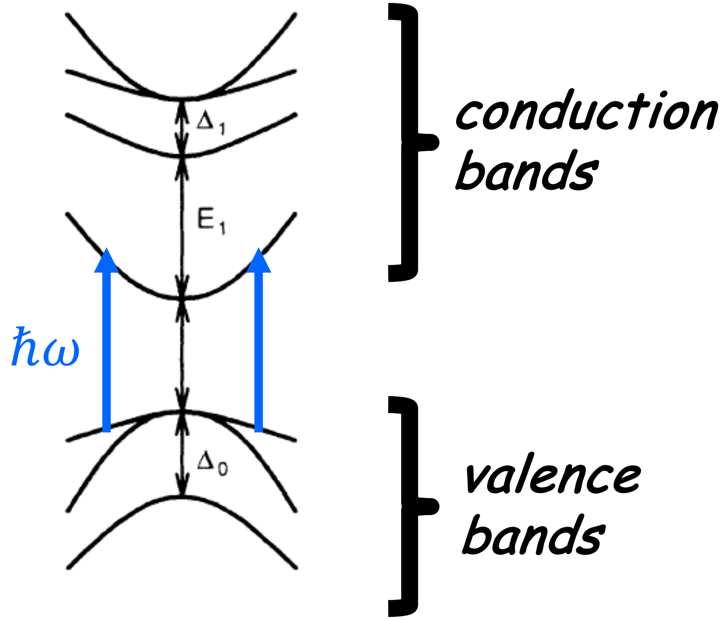
with

$$\omega_\Sigma = \omega_1 + \omega_2$$

$$\omega_\Sigma \to 0$$

$$P_i(\omega_\Sigma) = \frac{\sigma^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)} E_j(\omega_1) E_k(\omega_2)$$



$$\boldsymbol{J}(t) = \frac{d\boldsymbol{P}(t)}{dt}$$

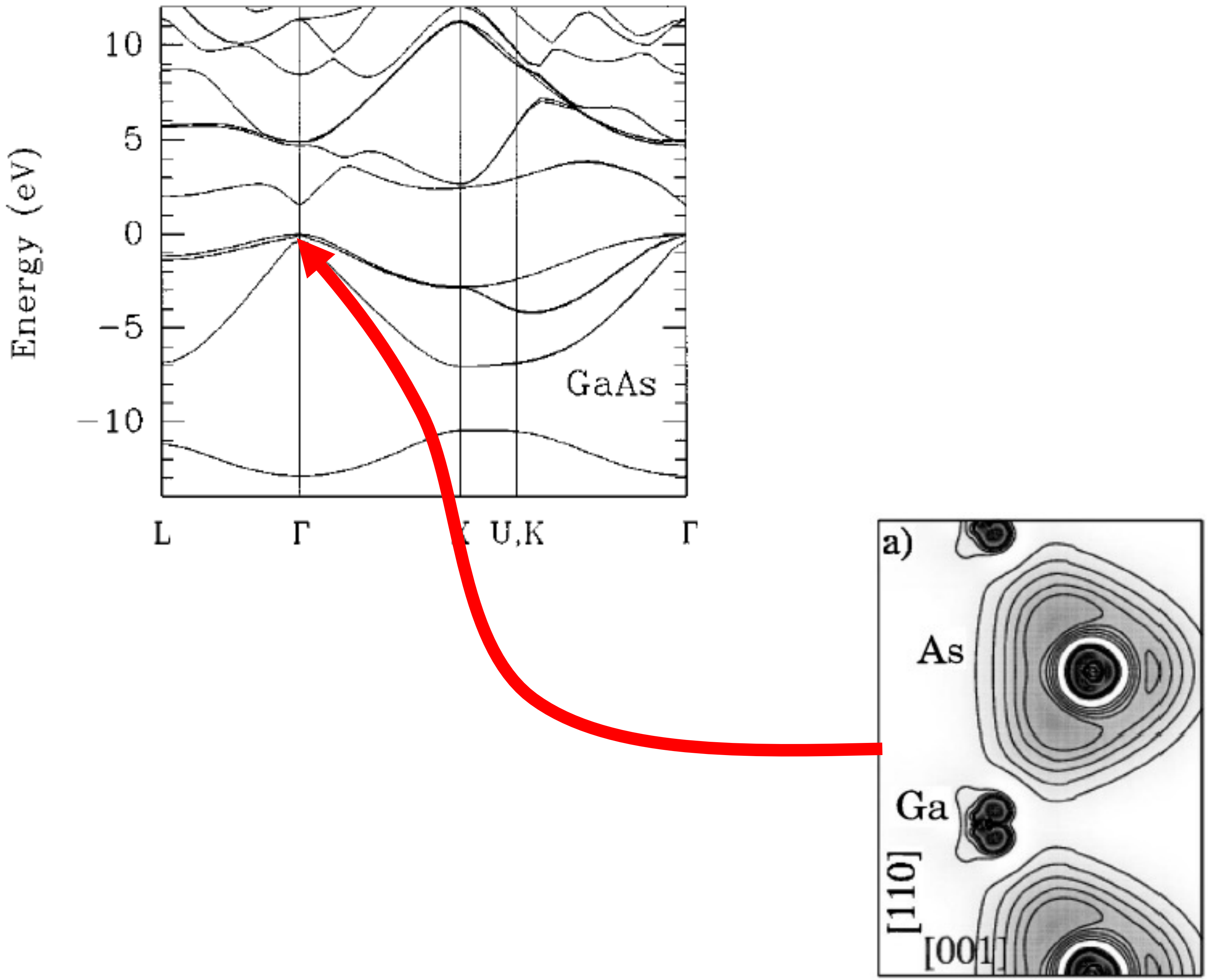
$$(-i\omega_\Sigma) P_i(\omega_\Sigma) = J_i(\omega_\Sigma)$$

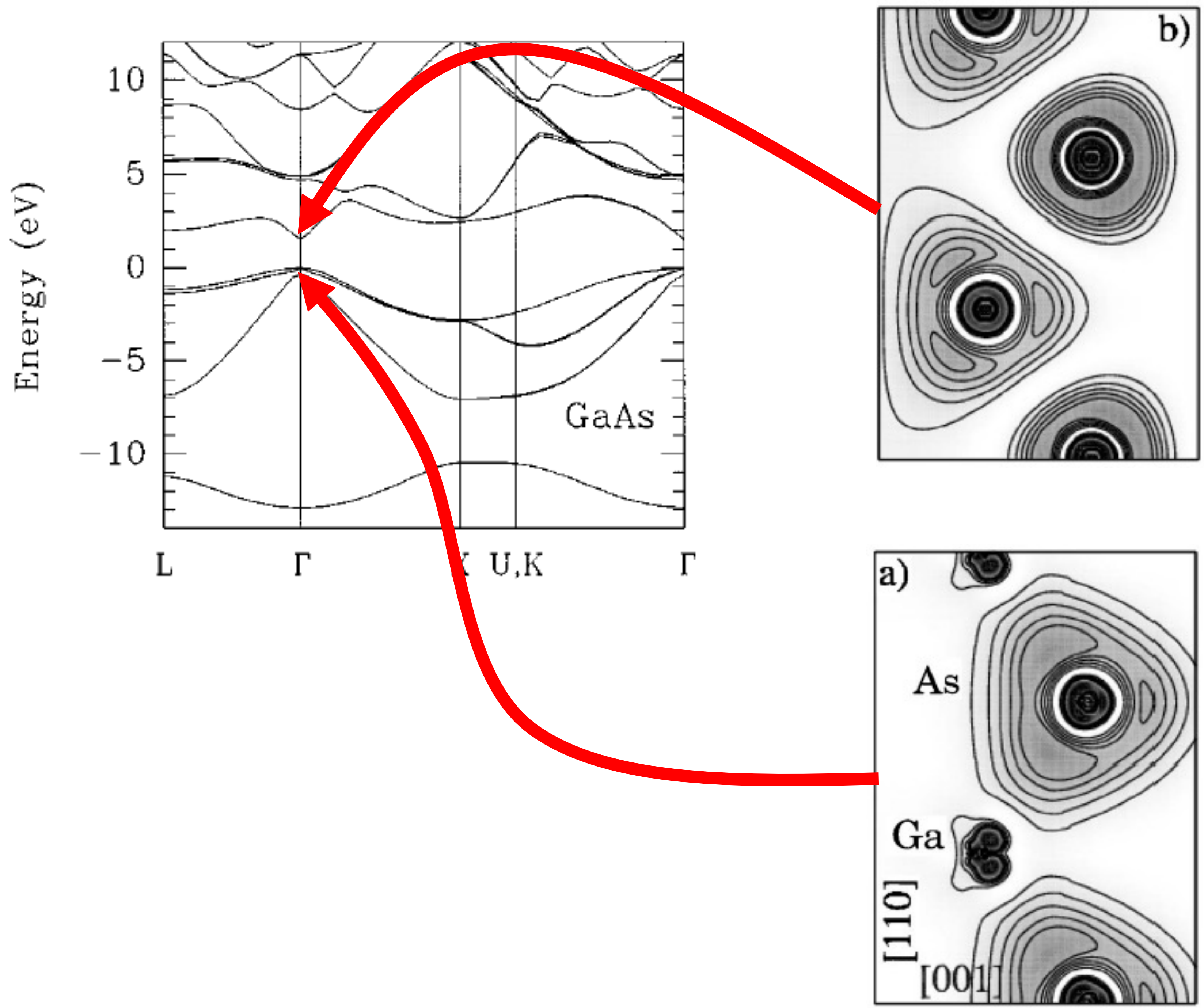
$$J_i(\omega_\Sigma) = \sigma^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) E_j(\omega_1) E_k(\omega_2)$$

CW limit

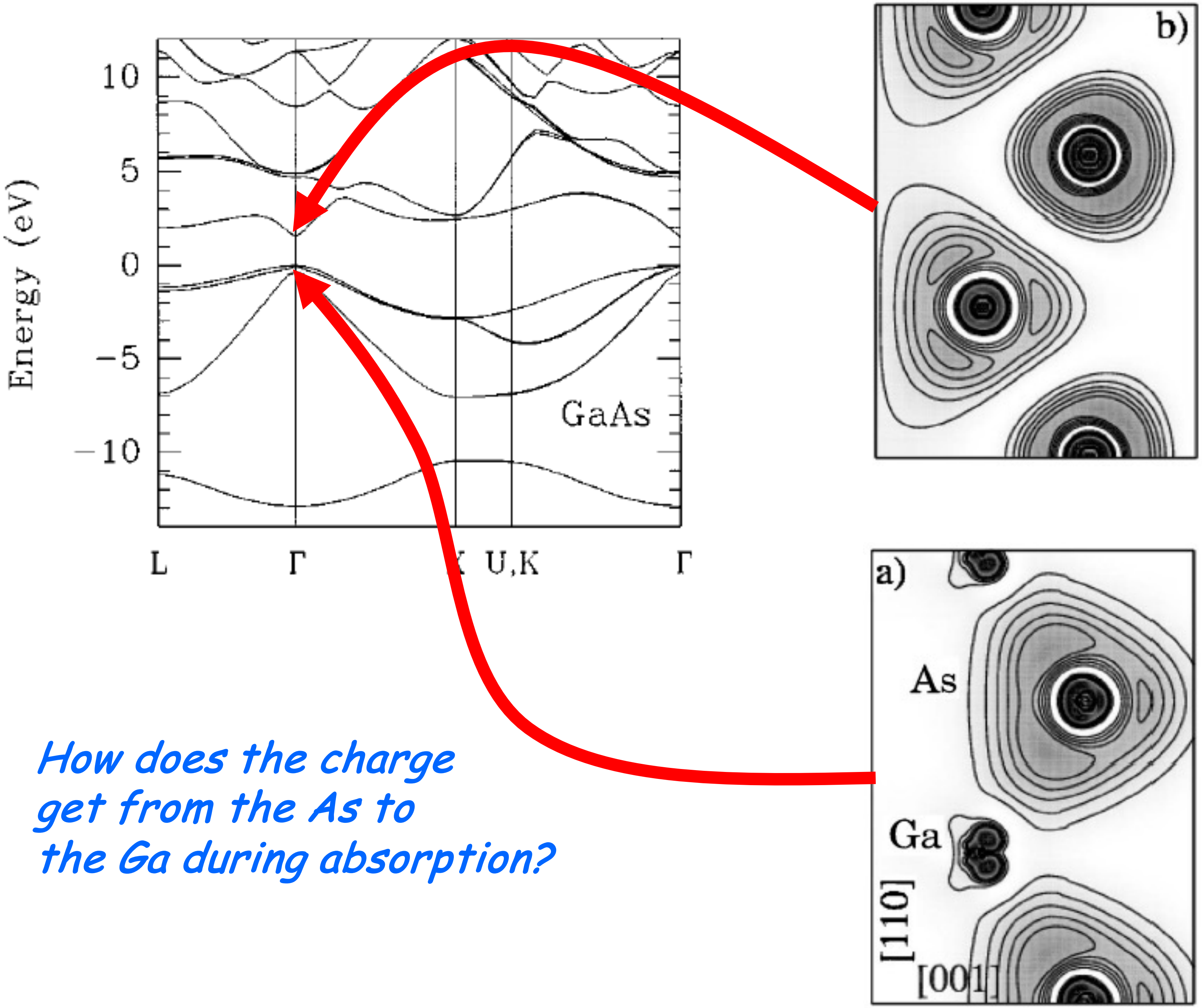
$$J_i = 2\sigma^{(2)}_{ijk}(0;\omega,-\omega) E_j(\omega) E_k(-\omega)$$

"shift current"
"photovoltaic effect"

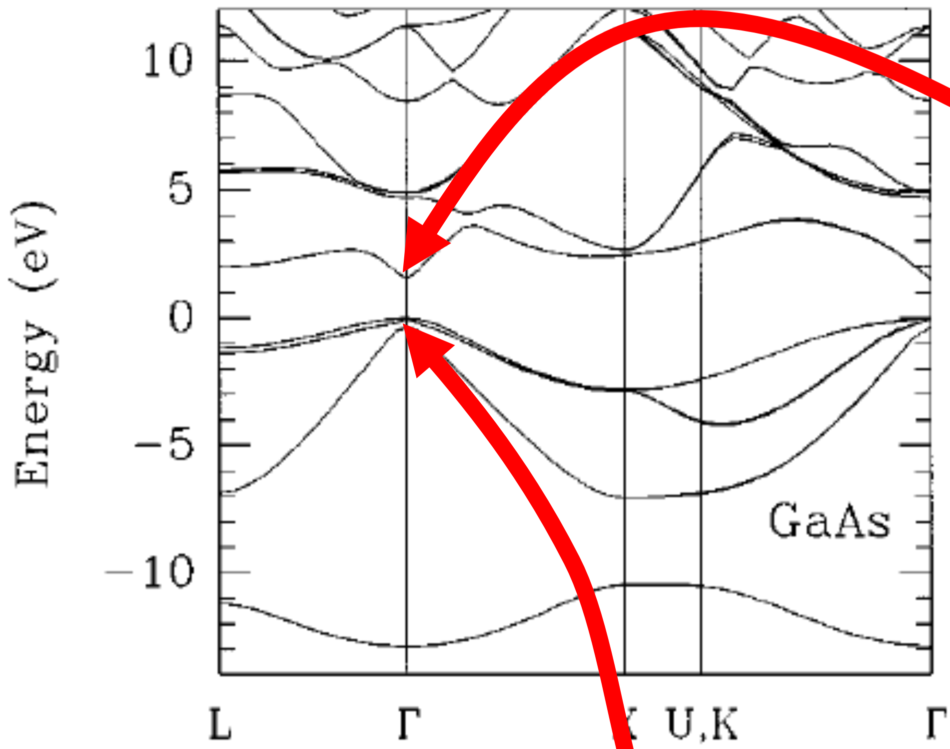

Energy (eV)
10
5
0
−5
−10
GaAs
L
Γ
U,K
Γ
a)
As
Ga
[110]
[001]

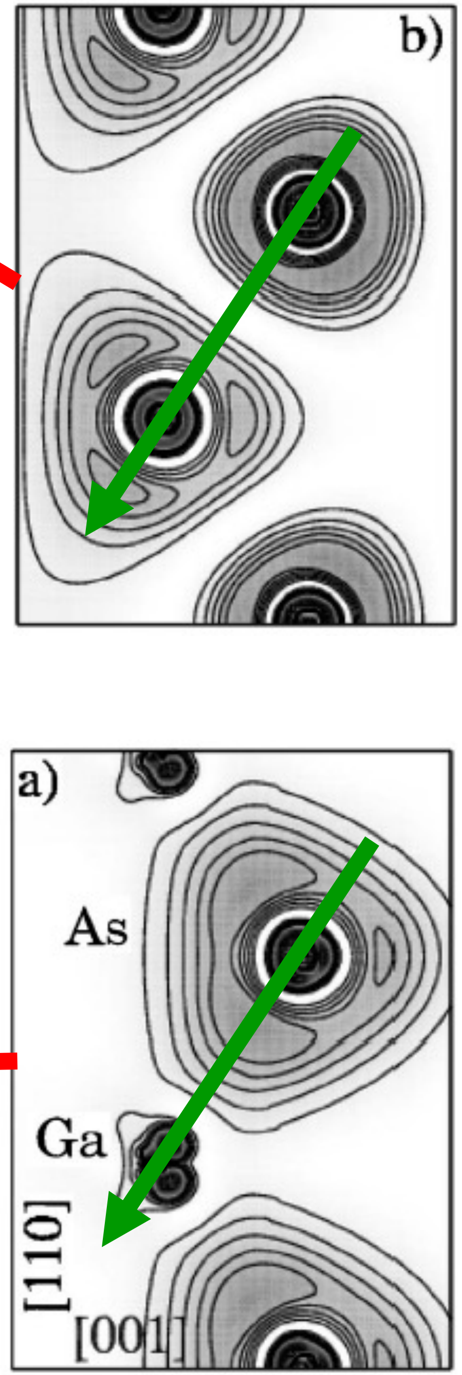


Energy (eV)
10
5
0
−5
−10
GaAs
L
Γ
X
U,K
Γ
b)
a)
As
Ga
[110]
[001]



How does the charge get from the As to the Ga during absorption?



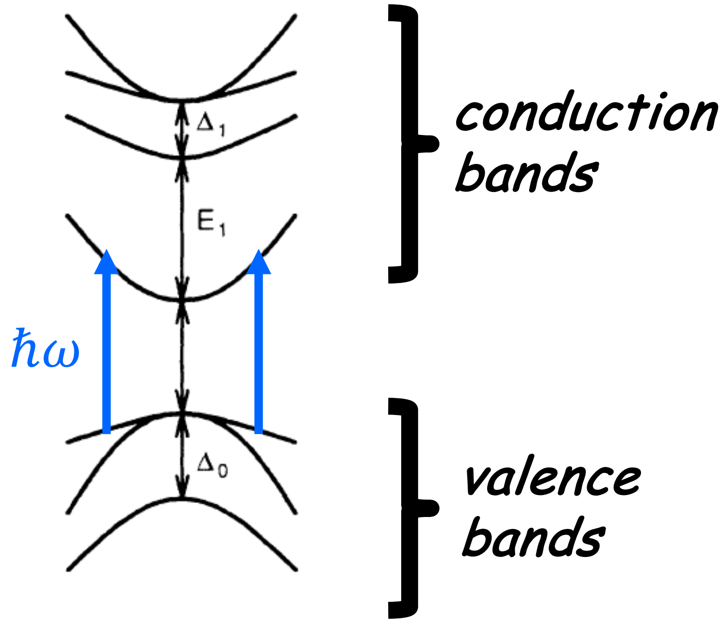


How does the charge get from the As to the Ga during absorption?

For light polarized along [111]

Suppose

$$\omega_1 = \omega > \omega_{gap}$$
$$\omega_2 = -\omega$$



$$\chi^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) =$$

$$\overline{\chi}^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2) + \frac{\sigma^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)} + \frac{\eta^{(2)}_{ijk}(-\omega_\Sigma;\omega_1,\omega_2)}{(-i\omega_\Sigma)^2}$$

with

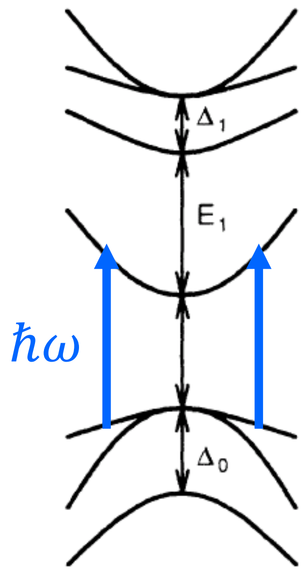
$$\omega_\Sigma = \omega_1 + \omega_2$$

$$\omega_\Sigma \to 0$$

$$P_i(\omega_\Sigma) = \frac{\eta^{(2)}_{ijk}(-\omega_\Sigma; \omega_1, \omega_2)}{(-i\omega_\Sigma)^2} E_j(\omega_1) E_k(\omega_2)$$

$$\frac{d\boldsymbol{J}(t)}{dt} = \frac{d^2 \boldsymbol{P}(t)}{dt^2}$$

$$(-i\omega_\Sigma)^2 P_i(\omega_\Sigma) = (-i\omega_\Sigma) J_i(\omega_\Sigma)$$



conduction bands

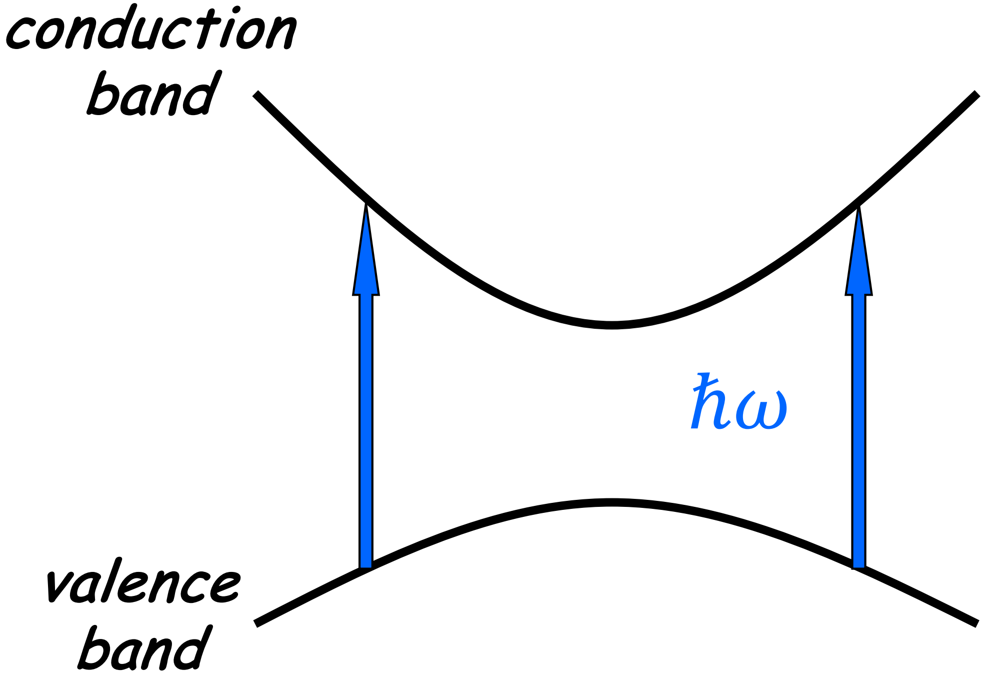
valence bands

$$\frac{dJ_i}{dt}(\omega_\Sigma) = \eta^{(2)}_{ijk}(-\omega_\Sigma; \omega_1, \omega_2) E_j(\omega_1) E_k(\omega_2)$$

CW limit

$$\frac{dJ_i}{dt} = 2\eta^{(2)}_{ijk}(0; \omega, -\omega) E_j(\omega) E_k(-\omega)$$

"injection current"
"circular photocurrent"
"photogalvanic effect"

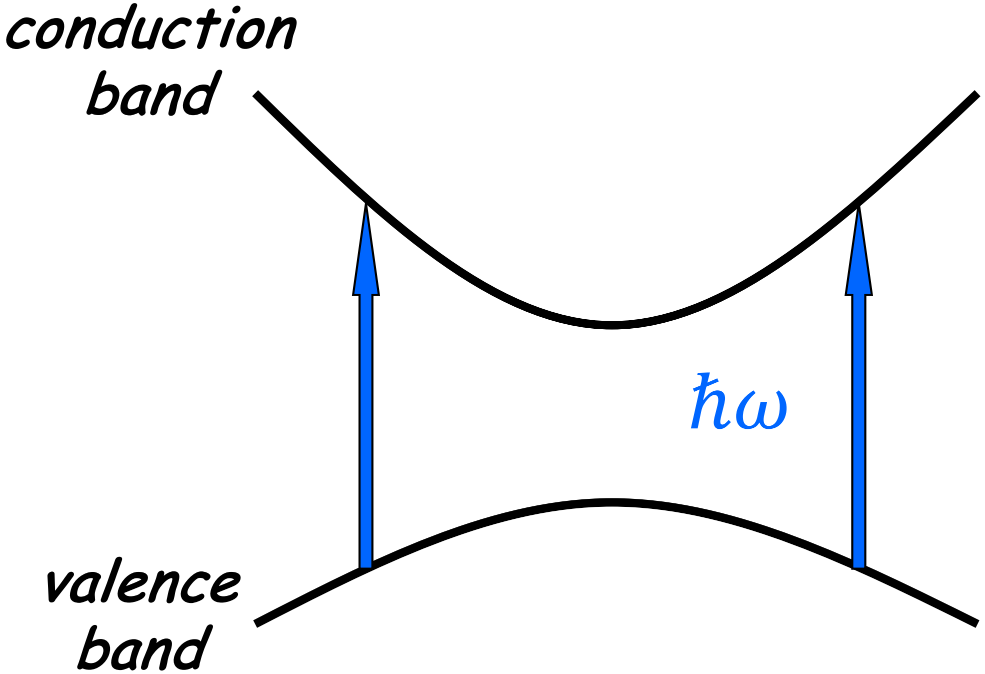


amplitude for transition

$$\propto \quad \mathbf{p}_{cv}(\mathbf{k})\cdot\mathbf{E}(\omega)$$

probability for transition

$$\propto \quad \left|\mathbf{p}_{cv}(\mathbf{k})\cdot\mathbf{E}(\omega)\right|^2$$



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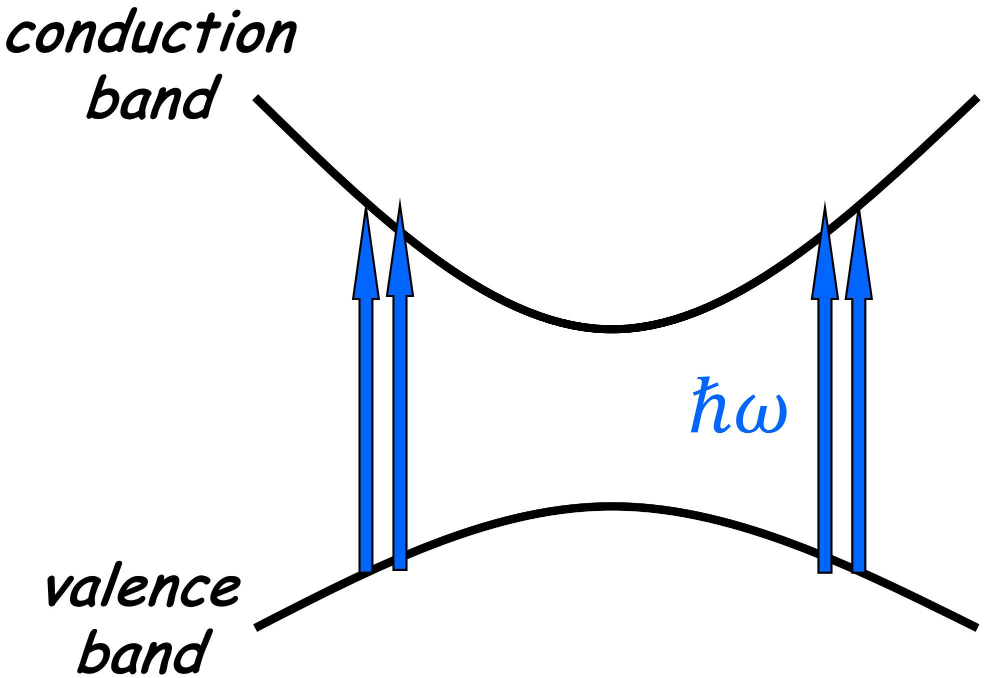
probability for transition

$$\propto \quad \left|\mathbf{p}_{cv}(\mathbf{k})\cdot\mathbf{E}(\omega)\right|^2$$

$$\mathbf{p}_{cv}(\mathbf{k})\cdot\mathbf{E} = p_{cv}^{x}(\mathbf{k})E^{x}(\omega) + p_{cv}^{z}(\mathbf{k})E^{z}(\omega)$$



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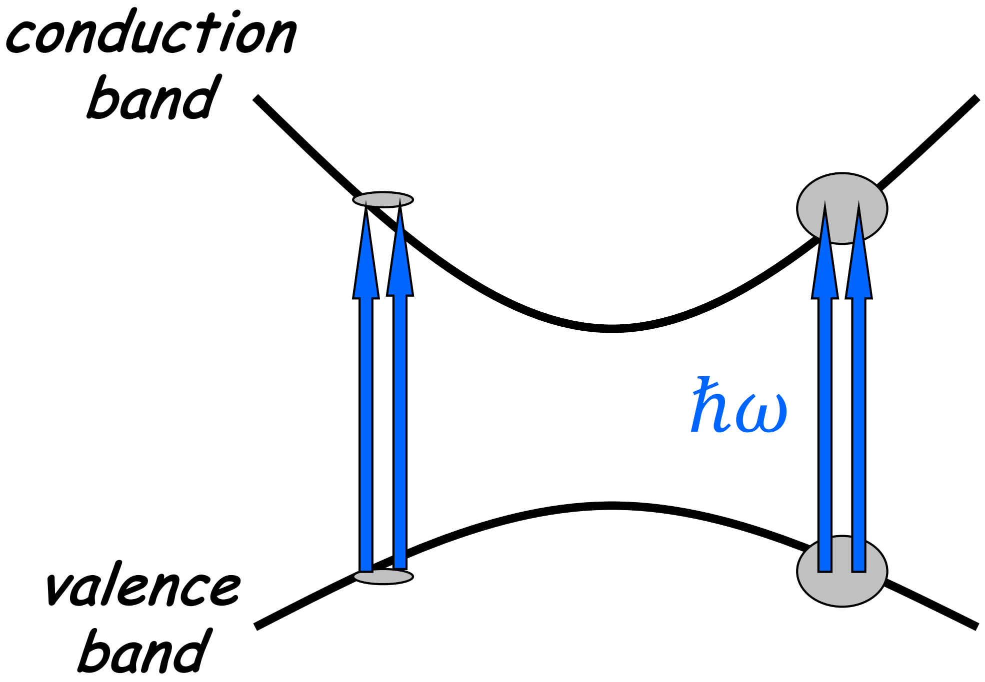


amplitude for transition

$$\propto \quad \mathbf{p}_{cv}(\mathbf{k}) \cdot \mathbf{E}(\omega)$$

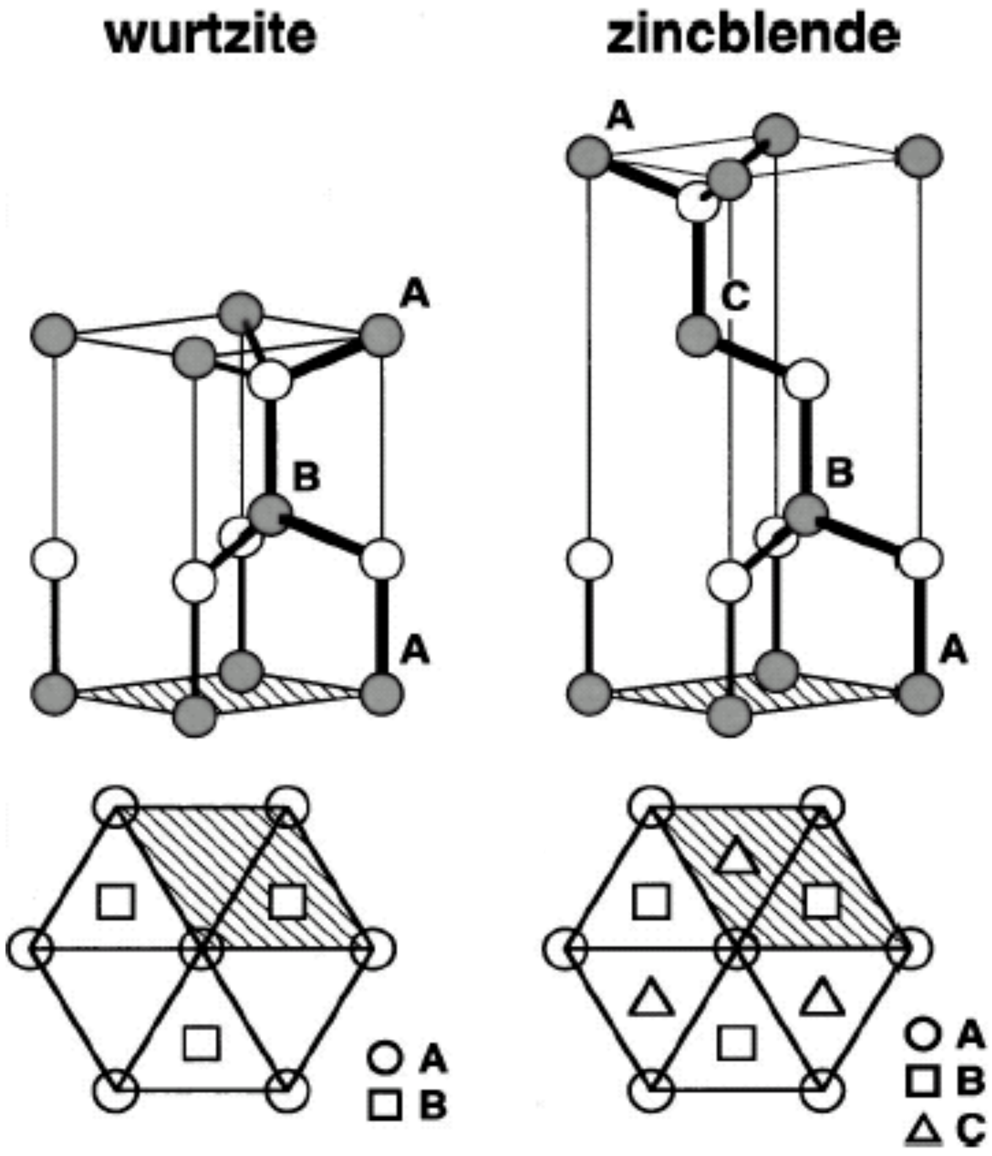
probability for transition

$$\propto \quad \left|\mathbf{p}_{cv}(\mathbf{k}) \cdot \mathbf{E}(\omega)\right|^2$$

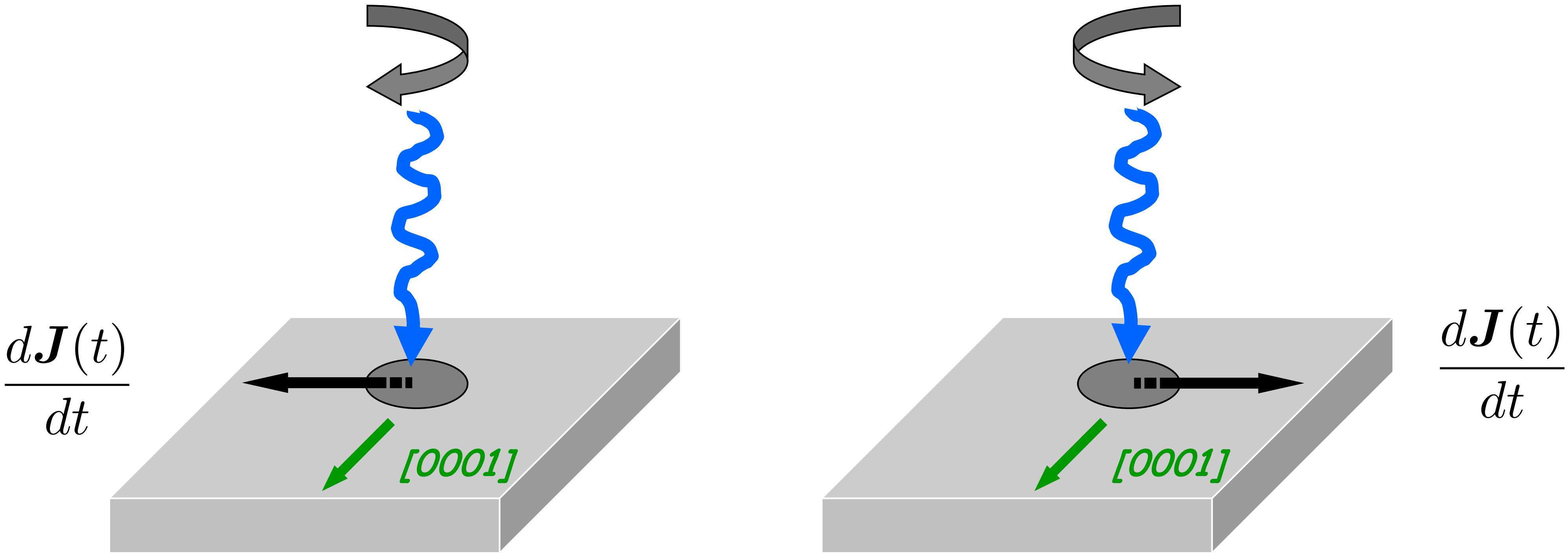
$$\mathbf{p}_{cv}(\mathbf{k}) \cdot \mathbf{E} = p_{cv}^{x}(\mathbf{k}) E^{x}(\omega) + p_{cv}^{z}(\mathbf{k}) E^{z}(\omega)$$

$$\left|p_{cv}^{x}(\mathbf{k}) E^{x}(\omega) + p_{cv}^{z}(\mathbf{k}) E^{z}(\omega)\right|^2$$

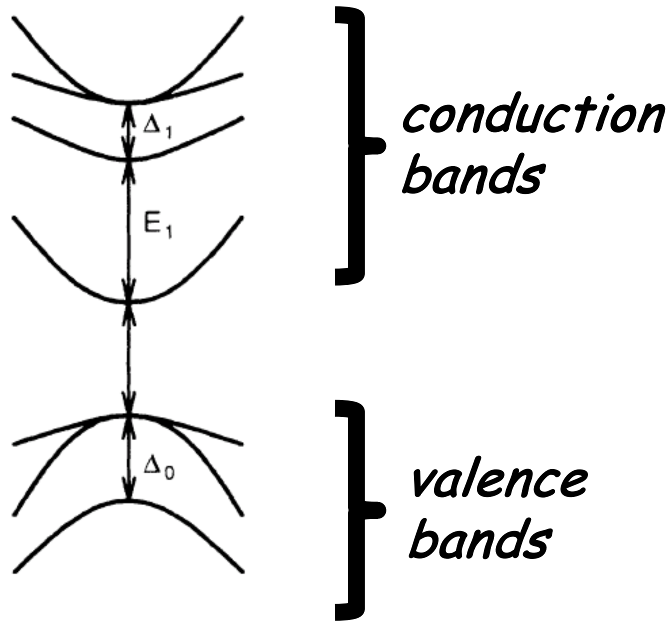
can show interference effects



$\eta^{(2)}_{ijk} \neq 0$ $\qquad$ $\eta^{(2)}_{ijk} = 0$



direction of current from crystal axis depends on helicity of beam



$$\chi^{(3)}_{ijkl}(-\omega_\Sigma;\omega_1,\omega_2,\omega_3) =$$

$$\overline{\chi}^{(3)}_{ijkl}(-\omega_\Sigma;\omega_1,\omega_2,\omega_3) + \frac{\sigma^{(3)}_{ijkl}(-\omega_\Sigma;\omega_1,\omega_2,\omega_3)}{(-i\omega_\Sigma)} + \frac{\eta^{(3)}_{ijkl}(-\omega_\Sigma;\omega_1,\omega_2,\omega_3)}{(-i\omega_\Sigma)^2}$$

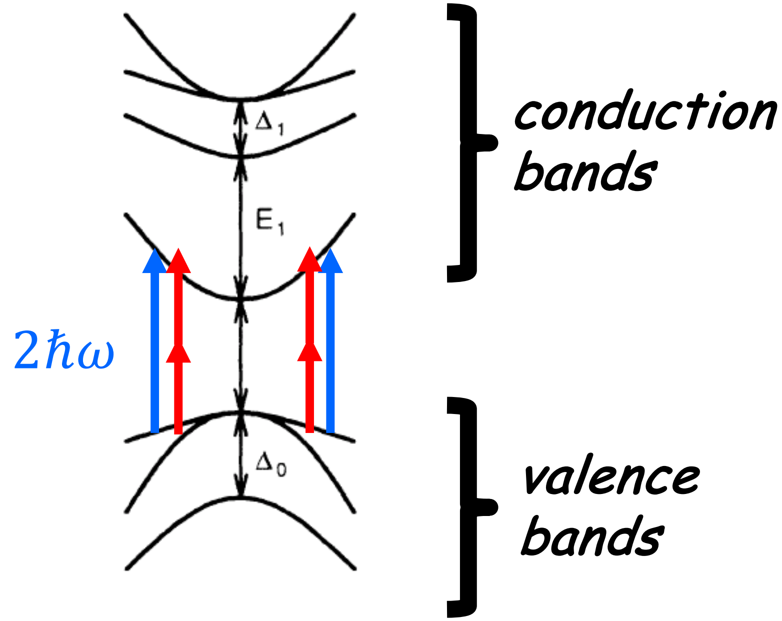
with

$$\omega_\Sigma = \omega_1 + \omega_2 + \omega_3$$

Suppose

$$\omega_1 = 2\omega > \omega_{gap}$$

$$\omega_2 = \omega_3 = -\omega$$



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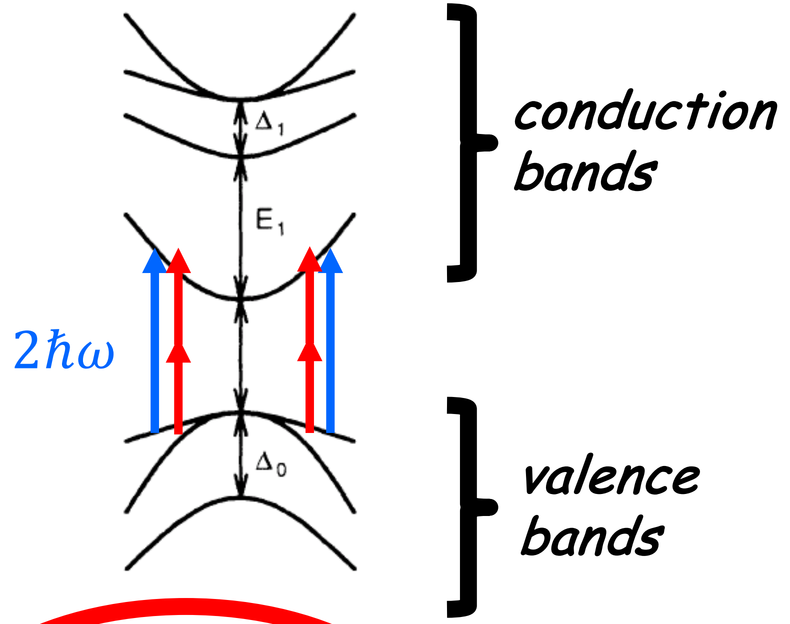
with

$$\omega_\Sigma = \omega_1 + \omega_2 + \omega_3$$

$$\omega_\Sigma \to 0$$

Suppose

$$\omega_1 = 2\omega > \omega_{gap}$$
$$\omega_2 = \omega_3 = -\omega$$



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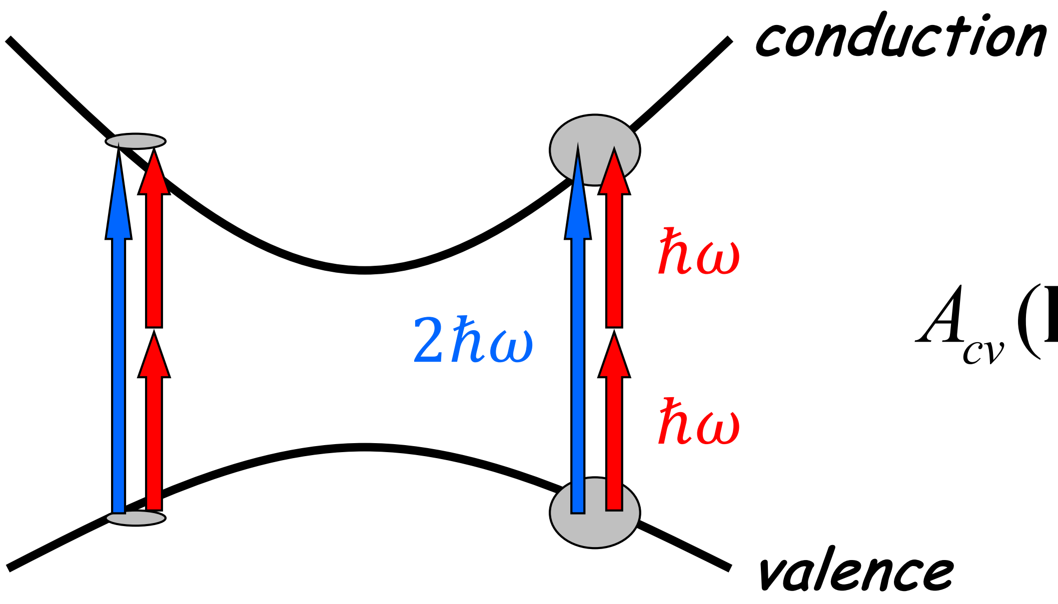
with

$$\omega_\Sigma = \omega_1 + \omega_2 + \omega_3$$

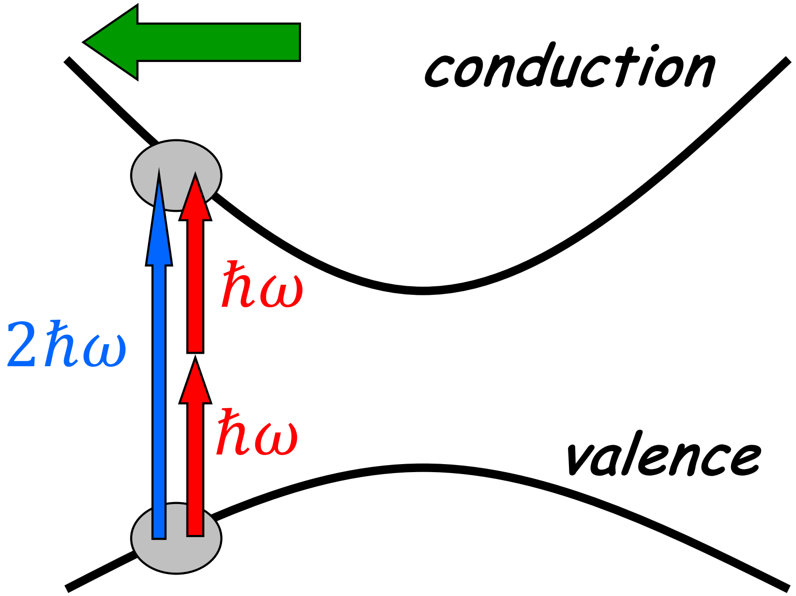
$$\omega_\Sigma \to 0$$

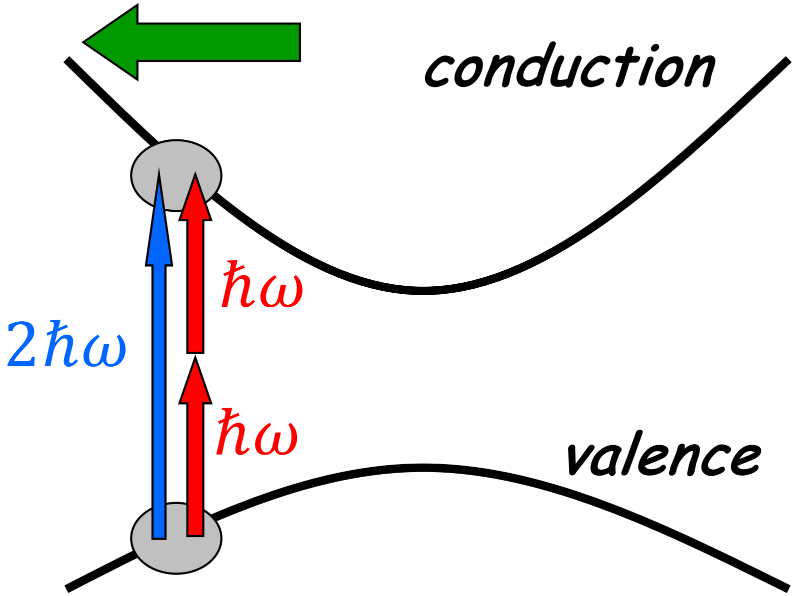


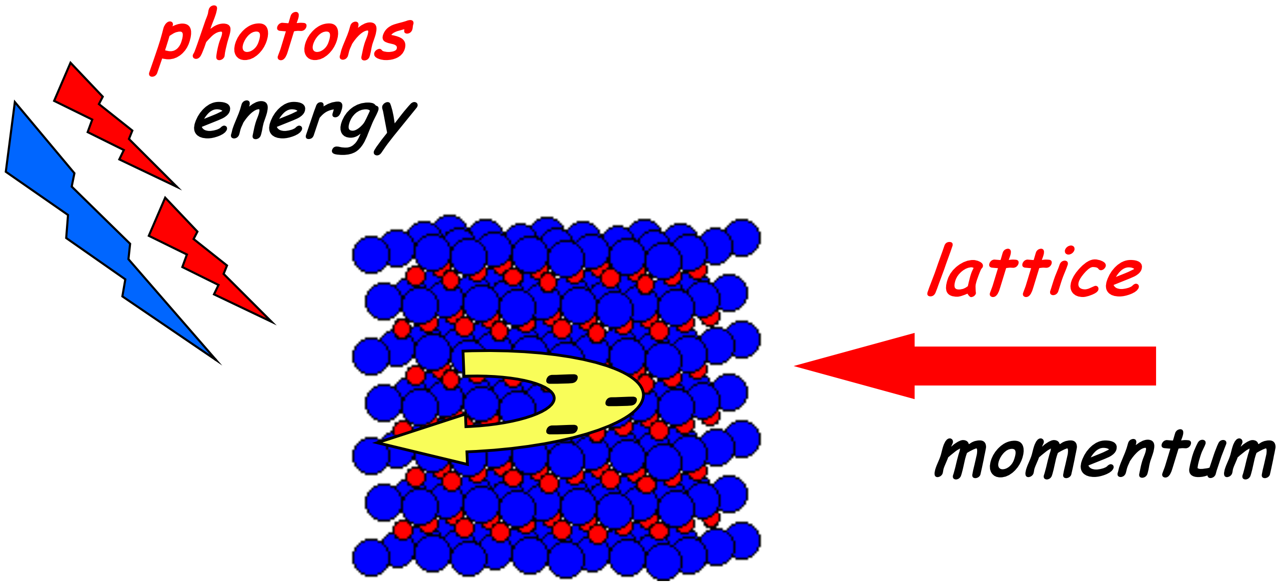
$$A_{cv}(\mathbf{k}) = A_{cv}^{(1)}(\mathbf{k}) + A_{cv}^{(2)}(\mathbf{k})$$

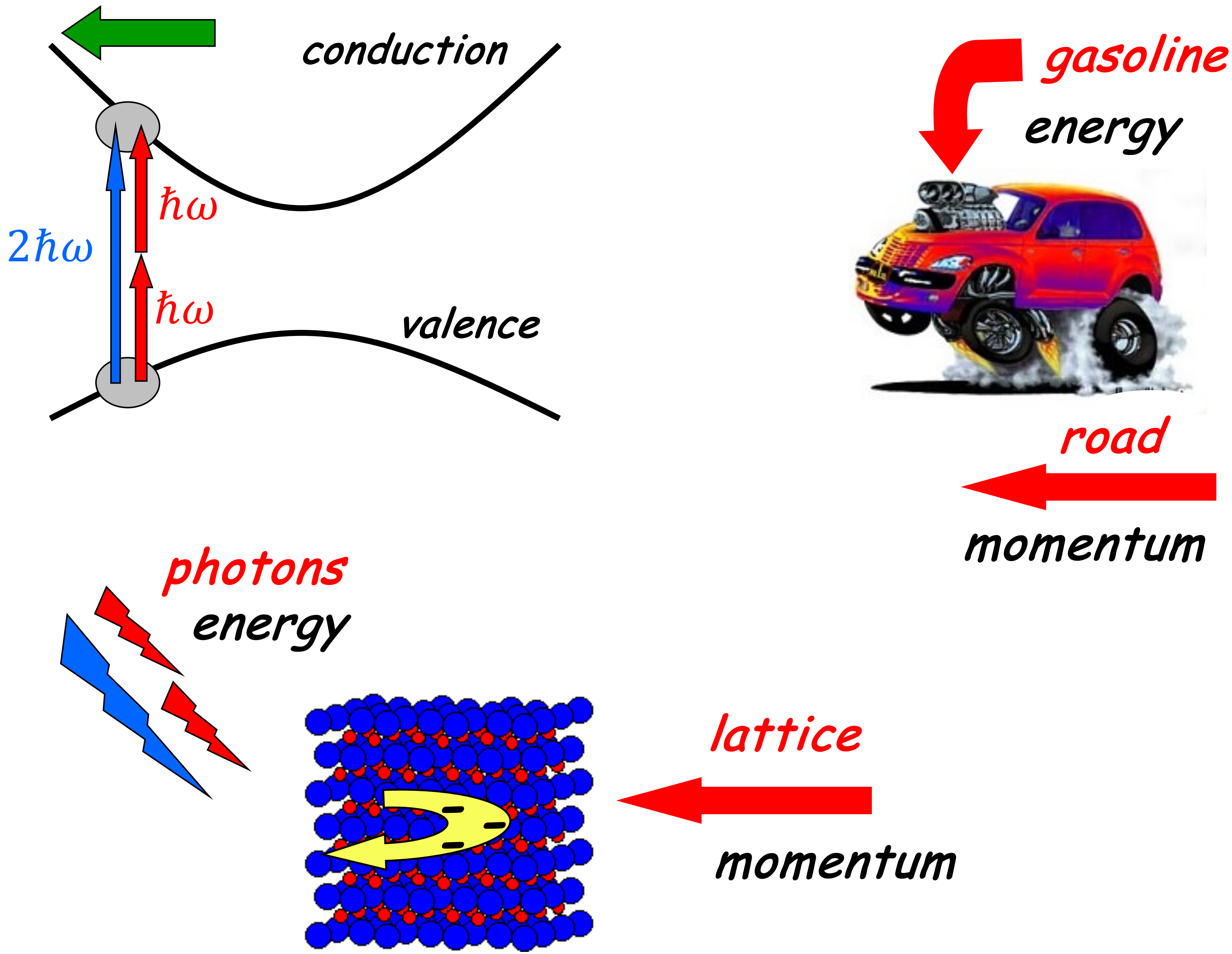
$$A_{cv}^{(1)}(\mathbf{k}) \quad \propto \quad \mathbf{p}_{cv}(\mathbf{k}) \cdot \mathbf{E}(2\omega)$$

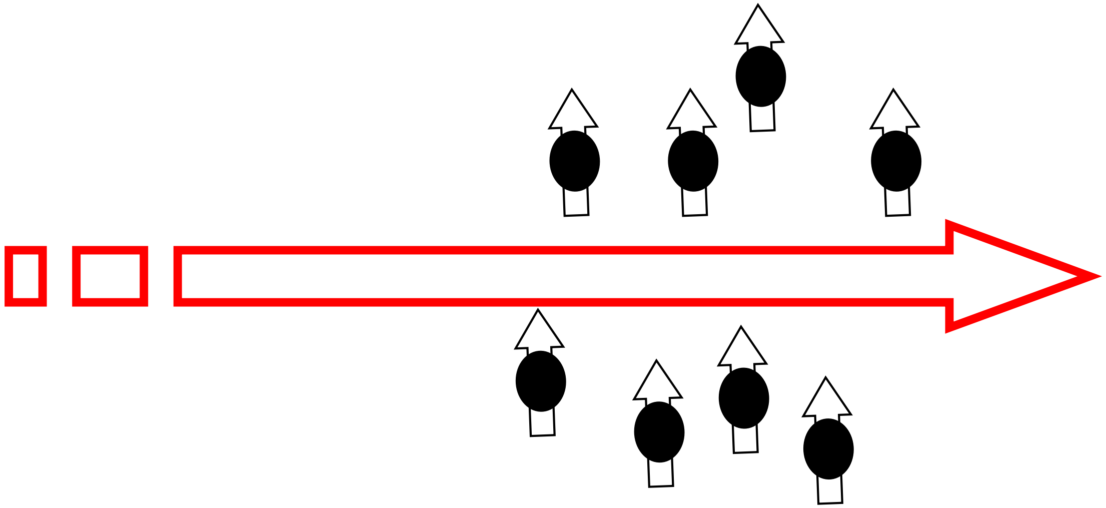
$$A_{cv}^{(2)}(\mathbf{k}) \quad \propto \quad \sum_n \frac{\left[\mathbf{p}_{cn}(\mathbf{k}) \cdot \mathbf{E}(\omega)\right]\left[\mathbf{p}_{nv}(\mathbf{k}) \cdot \mathbf{E}(\omega)\right]}{\left[\omega_c(\mathbf{k}) + \omega_v(\mathbf{k}) - 2\omega_n(\mathbf{k})\right]}$$


conduction
$\hbar\omega$
$2\hbar\omega$
$\hbar\omega$
valence

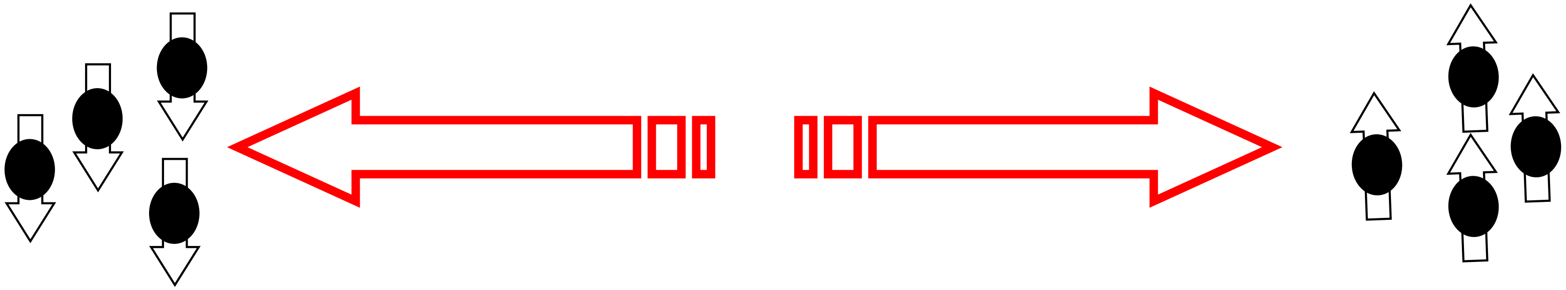

conduction
$\hbar\omega$
$2\hbar\omega$
$\hbar\omega$
valence


photons
energy
lattice
momentum


conduction
valence
$2\hbar\omega$
$\hbar\omega$
$\hbar\omega$
gasoline
energy
road
momentum
photons
energy
lattice
momentum



spin-polarized current



pure spin current

Susceptibilities

$\chi^{(2)}$ *effects*

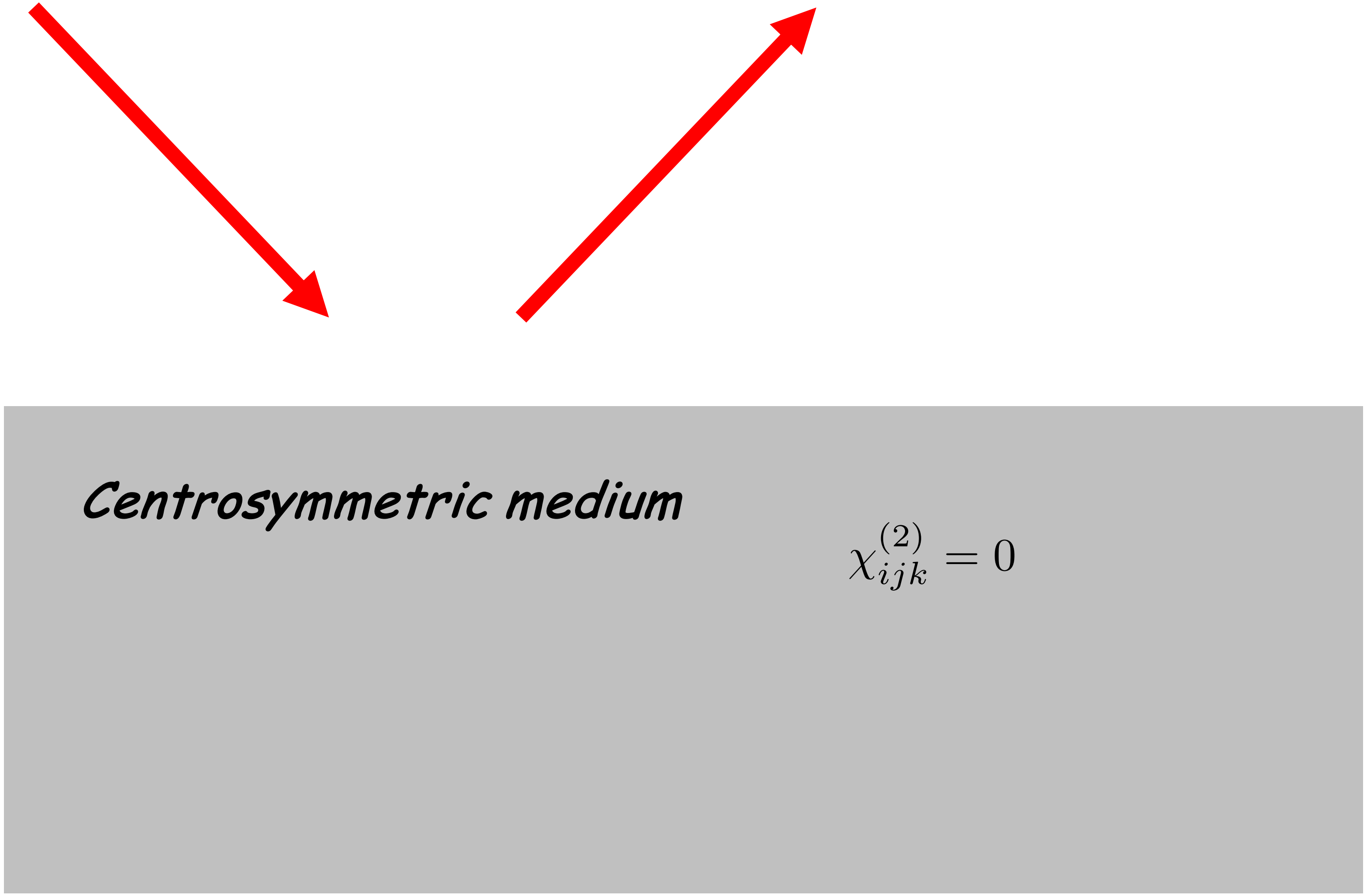
$\chi^{(3)}$ *effects*

Quantum nonlinear optics

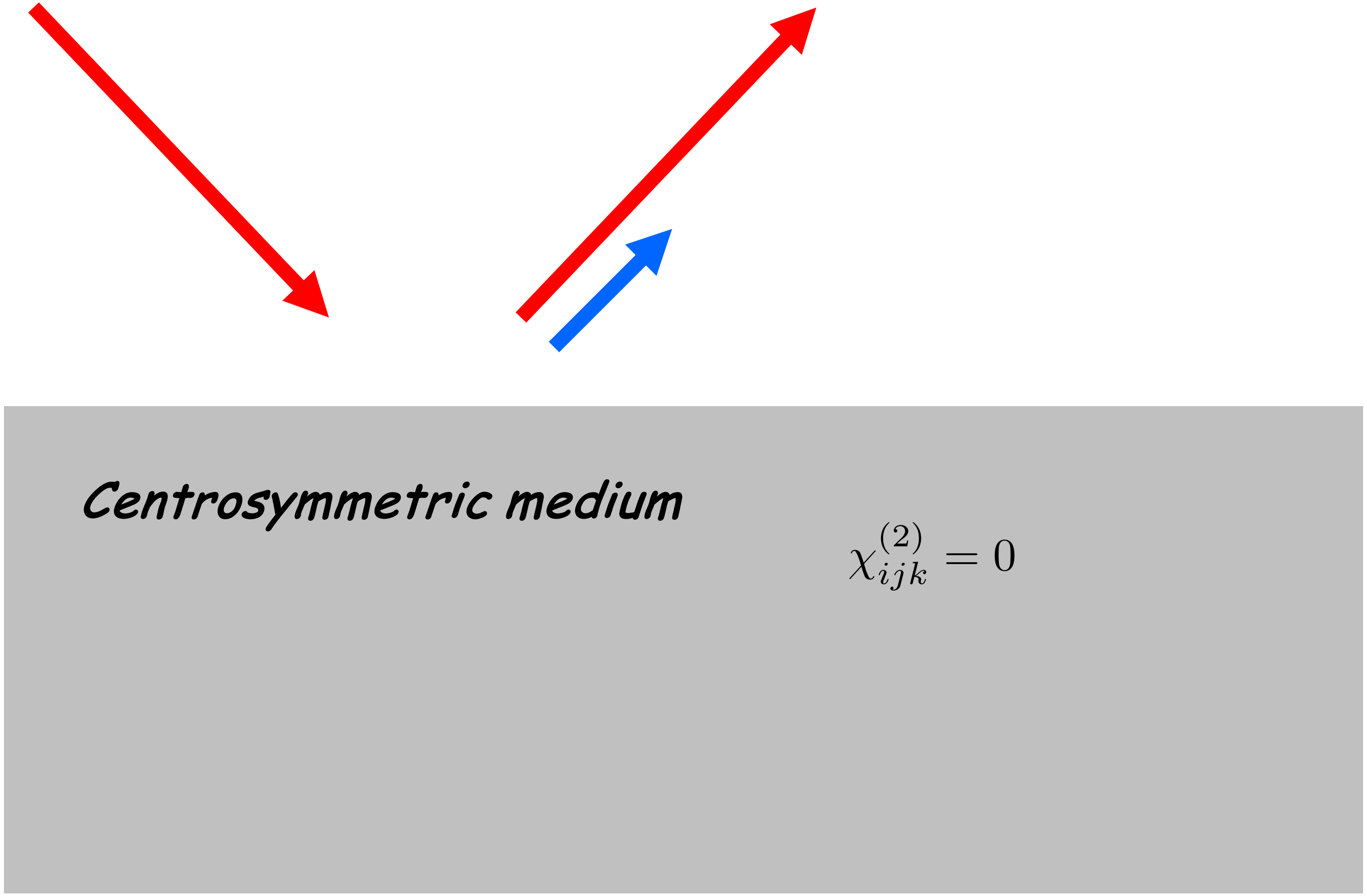
Nonlinear optics and electronics

Forbidden processes

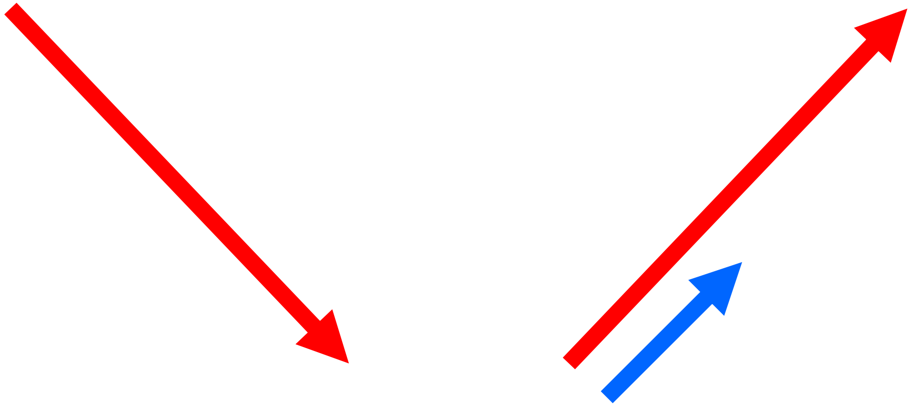
Forbidden processes



Forbidden processes



Forbidden processes



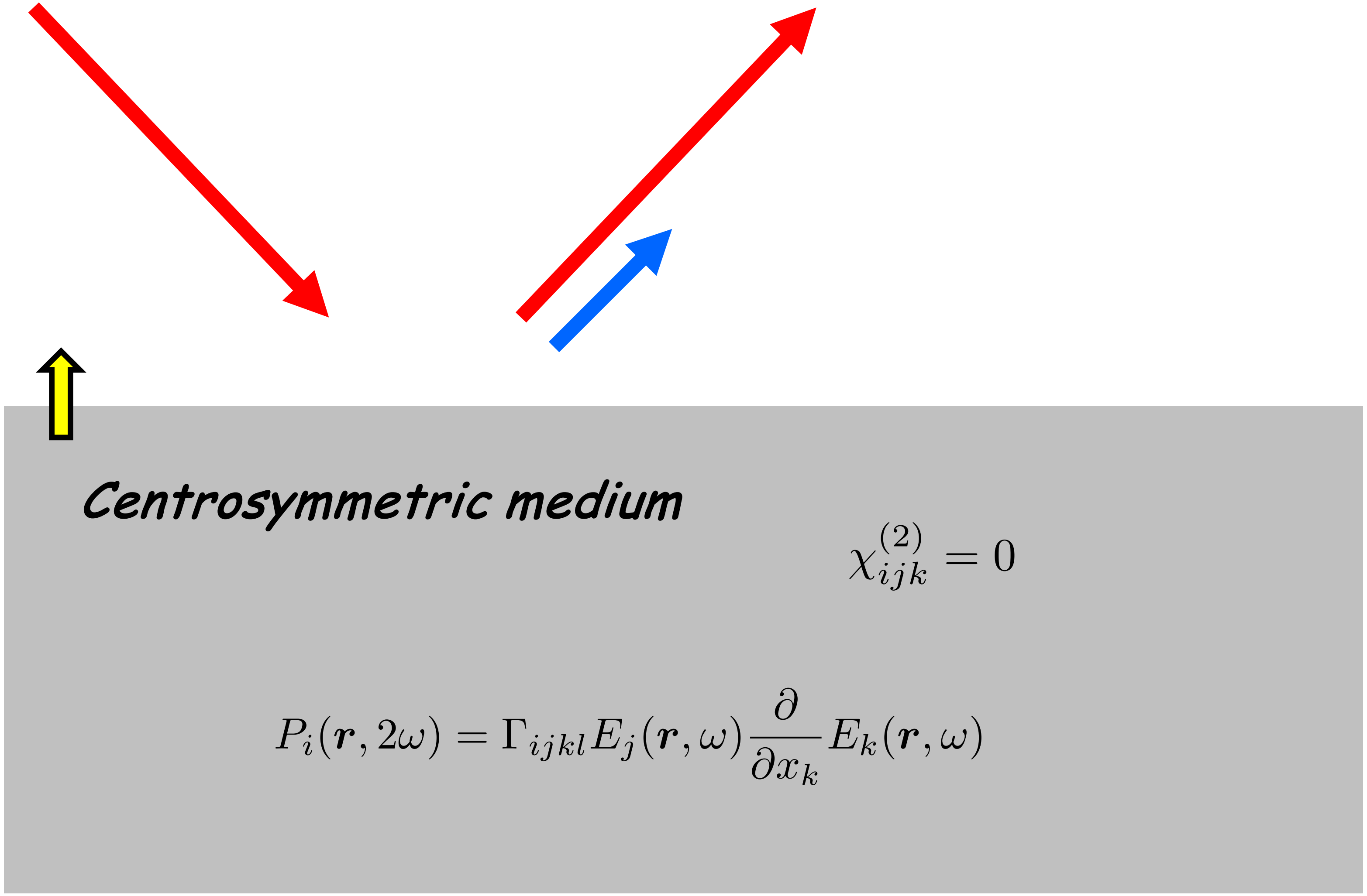
Centrosymmetric medium

$$\chi^{(2)}_{ijk} = 0$$

$$P_i(\boldsymbol{r}, 2\omega) = \Gamma_{ijkl} E_j(\boldsymbol{r}, \omega) \frac{\partial}{\partial x_k} E_k(\boldsymbol{r}, \omega)$$

Forbidden processes

(..and allowed)

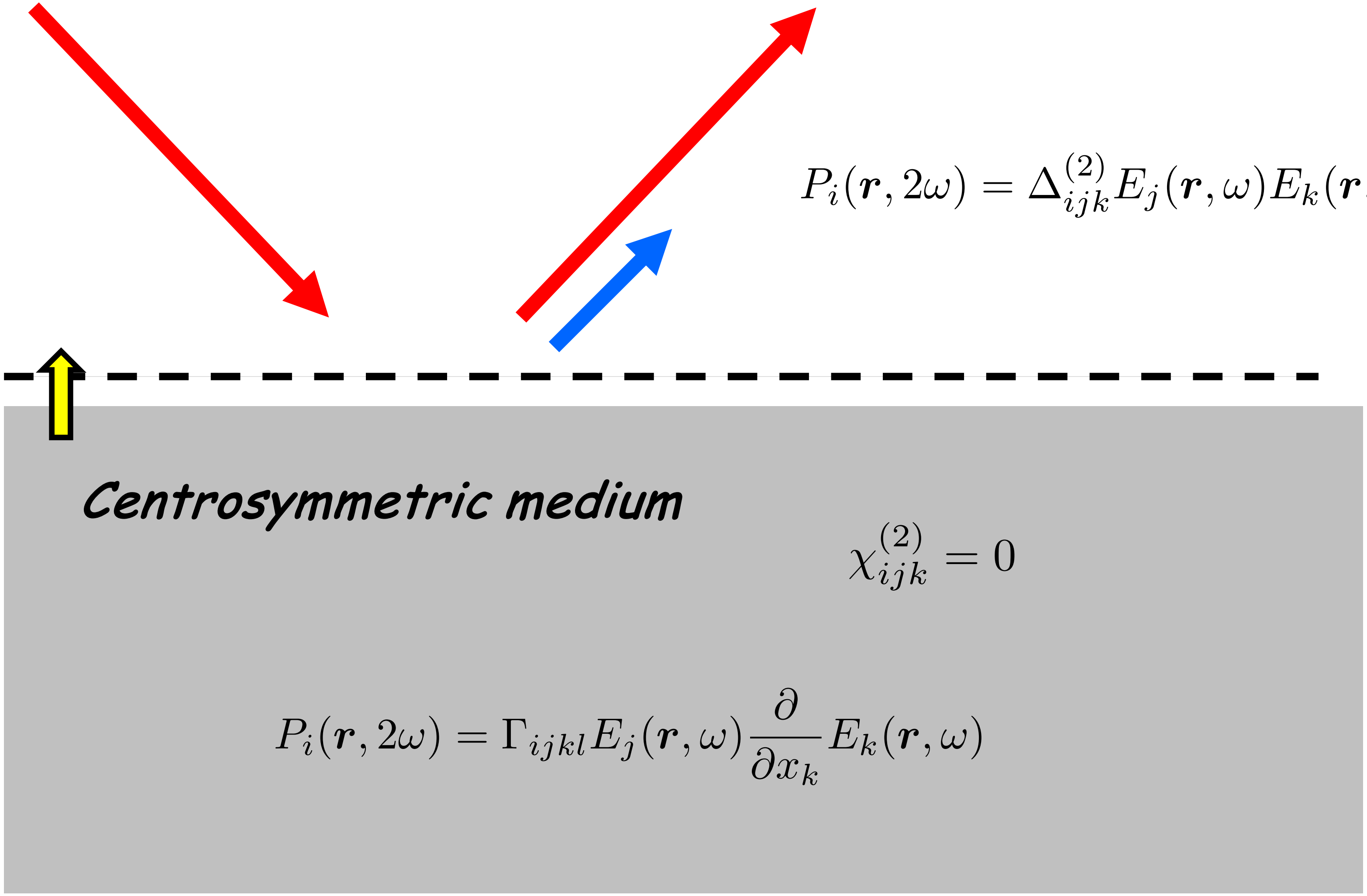


$$\chi^{(2)}_{ijk} = 0$$

$$P_i(\boldsymbol{r}, 2\omega) = \Gamma_{ijkl} E_j(\boldsymbol{r}, \omega) \frac{\partial}{\partial x_k} E_k(\boldsymbol{r}, \omega)$$

Forbidden processes

(..and allowed)



$$P_i(\boldsymbol{r}, 2\omega) = \Delta^{(2)}_{ijk} E_j(\boldsymbol{r}, \omega) E_k(\boldsymbol{r}, \omega) \delta(z - 0^+)$$

$$\chi^{(2)}_{ijk} = 0$$

$$P_i(\boldsymbol{r}, 2\omega) = \Gamma_{ijkl} E_j(\boldsymbol{r}, \omega) \frac{\partial}{\partial x_k} E_k(\boldsymbol{r}, \omega)$$

Susceptibilities
$\chi^{(2)}$ effects
$\chi^{(3)}$ effects
Quantum nonlinear optics
Nonlinear optics and electronics
Forbidden processes

"Optical alchemy"