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The Nonlinear Optical Properties of Semiconductors

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Optical Susceptibility Tensor

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$$\mathbf{P} = \varepsilon_0 \left[\chi^{(1)}\mathbf{E} + \chi^{(2)}\mathbf{EE} + \chi^{(3)}\mathbf{EEE} + \ldots \right]$$

$\chi^{(1)}_{ij}$	ω	Linear refraction and absorption
$\chi^{(2)}_{ijk}$	$\omega_1 + \omega_2$	Sum frequency generation
	$\omega_1 - \omega_2$	Difference frequency generation, rectification
	$\omega + 0$	Electro-optic (Pockel's) effect
$\chi^{(3)}_{ijkl}$	$\omega + \omega + \omega$	Third harmonic generation
	$\omega - \omega + \omega$	Nonlinear refraction and absorption, 4WM
	$\omega + 0 + 0$	Kerr electro-optic effect (DC)

Contents

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- Structure of zincblende semiconductors
- $\chi^{(1)}$: Absorption & Refraction
- $\chi^{(2)}$: Electro-optic effect (Pockels) & Frequency conversion, e.g. SHG
- $\chi^{(3)}$: Two-photon Absorption, Nonlinear Refraction & 4 wave mixing
- Quasi-$\chi^{(3)}$: Carrier effects, Electro-optic effect (Kerr) & Thermal effects

Waveguide geometry

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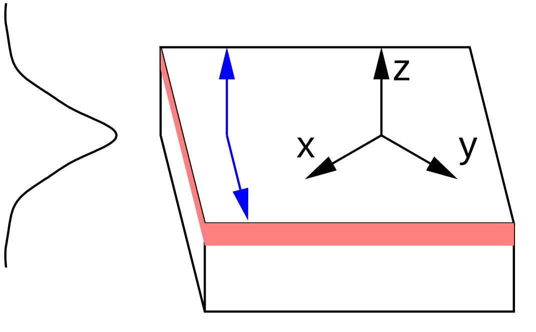
- Slab or rib waveguides
- Assume weakly guiding
- Also assume nonlinearity weak such that transverse guided mode unchanged
- Conventional orientation with $[001]$ growth and $[110]$ cleavage:



Crystal Symmetries

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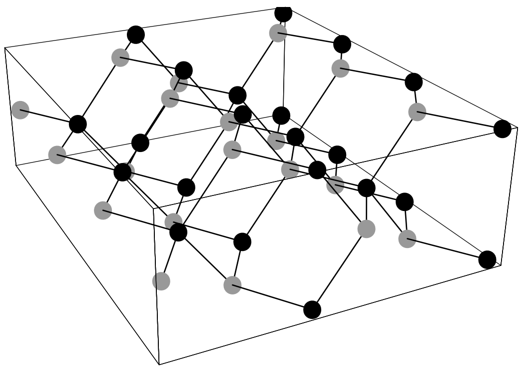

- Common compound semiconductors in photonics have a zinc-blende (cubic) structure $\bar{4}3$m
- Note different layer ordering for 111 and $\overline{111}$
- Introducing heterostructure, e.g. quantum well, breaks translational invariance in one direction

Bandstructure models

- Bloch form for wavefunction with periodic $u_{\mathbf{k}}(\mathbf{r})$

$$\psi(\mathbf{r}) = u_{\mathbf{k}}(\mathbf{r})\mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}$$

- Hamiltonian contains $\mathbf{k}\cdot\mathbf{p}$ term — treat as perturbation
  - 2 parabolic bands: scalar model
  - Kane model with singlet conduction band and triplet valence band: vector but isotropic
  - Kane plus next highest conduction triplet: anisotropic

AlGaAs bandstructure

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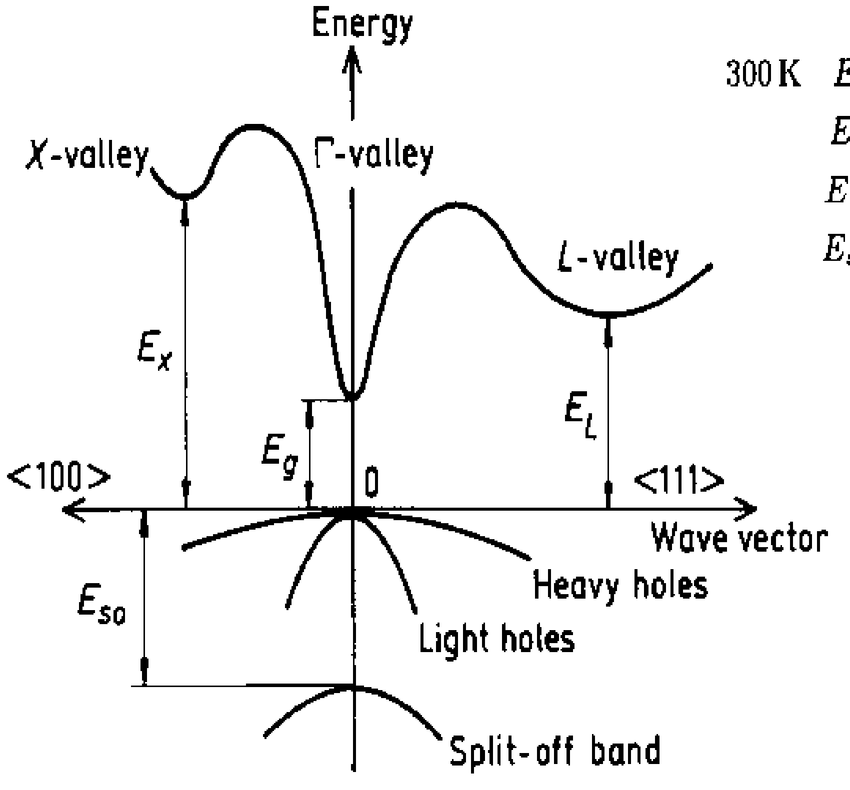
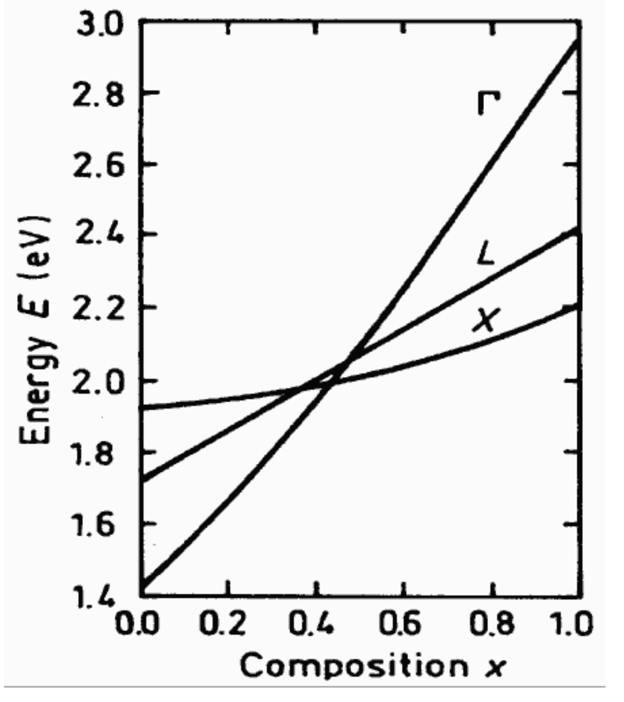


300 K $E_g = 1.42$ eV
$E_L = 1.71$ eV
$E_X = 1.90$ eV
$E_{so} = 0.34$ eV



$Al_xGa_{1-x}As$ direct bandgap for $x < 0.45$

$\mathrm{k} \cdot \mathrm{p}$ *models*

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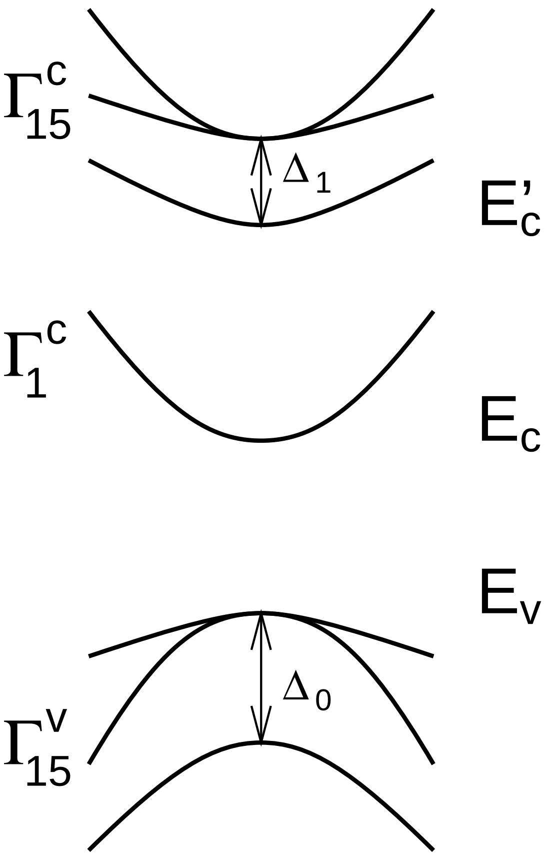

Quantum Theory of $\chi^{(n)}$

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- Susceptibility expressions are derived using the quantum Liouville equation, with Hamiltonian $H = H_0 + (e/m_0)\mathbf{A} \cdot \mathbf{p}$.
- For $\chi^{(n)}$ use sets of $(n+1)$-level systems and sum over $(n+1)!$ time orderings.
- Evaluation of $\chi^{(n)}$ in semiconductors requires:
  - Electronic energies (valence and conduction bands)
  - Momentum matrix elements between electronic states
  - Summation over *all* states

Linear absorption and refraction

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$$\chi_{ii}^{(1)}(\omega) = \frac{e^2}{2m_0^2\hbar\omega^2} \sum_{\mathbf{k},\text{bands}} |\mathbf{e}_i \cdot \mathbf{p}_{vc}(\mathbf{k})|^2$$

$$\times \left(\frac{1}{\Omega_{vc}(\mathbf{k}) - (\omega + \mathrm{i}\delta)} + \frac{1}{\Omega_{vc}(\mathbf{k}) + (\omega + \mathrm{i}\delta)} \right)$$

- take limit $\delta \to 0$:
  - $\alpha \propto \mathrm{Im}\chi^{(1)}$ & $n \propto \mathrm{Re}\chi^{(1)}$
- matrix element $|\mathbf{p}_{vc}(\mathbf{k})|$ is approximately constant.
- for parabolic bands, absorption follows square-root density-of-states

$\chi^{(1)}$ for 2-level system

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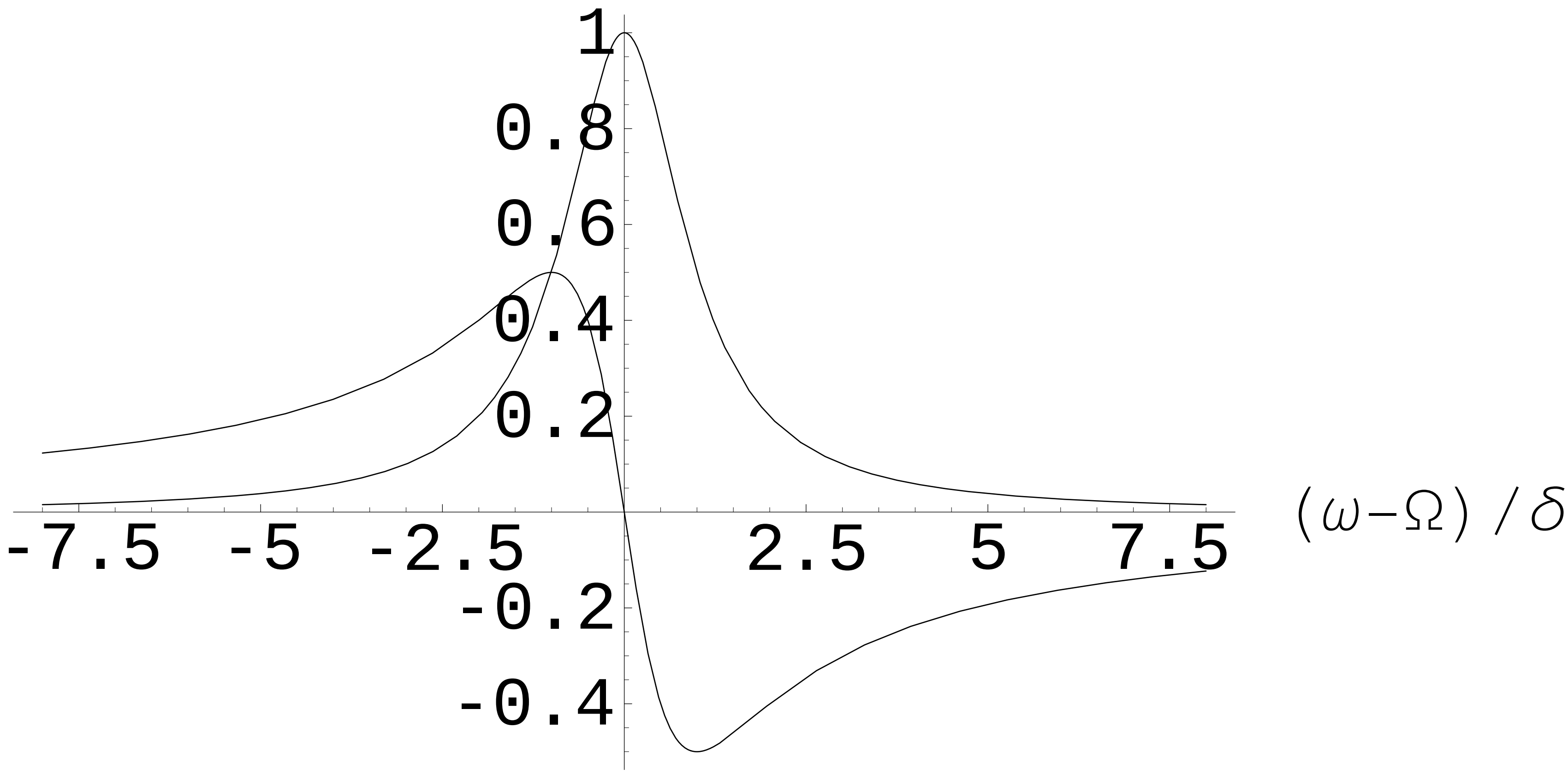

Linear refraction

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Dispersion with parabolic bands gives Adachi formula

$$\varepsilon(\omega) = A\left\{f(X) + \frac{1}{2}\left[\frac{E_g}{E_g+\Delta}\right]^{3/2} f(X_{so})\right\} + B$$

$$f(X) = \mathrm{Re}(2 - \sqrt{1+X} - \sqrt{1-X})/X^2 \qquad n = \sqrt{\varepsilon}$$

$$X = \frac{\hbar\omega}{E_g} = \frac{\lambda_g}{\lambda} \qquad X_{so} = \frac{\hbar\omega}{E_g+\Delta}$$

$$A(x) = 6.3 + 19.0x \qquad B(x) = 9.4 - 10.2x$$

$$E_g = (1.425 + 1.155x + 0.37x^2)\,\text{eV} \qquad \Delta = 0.34\,\text{eV}$$

Linear refraction (300K)

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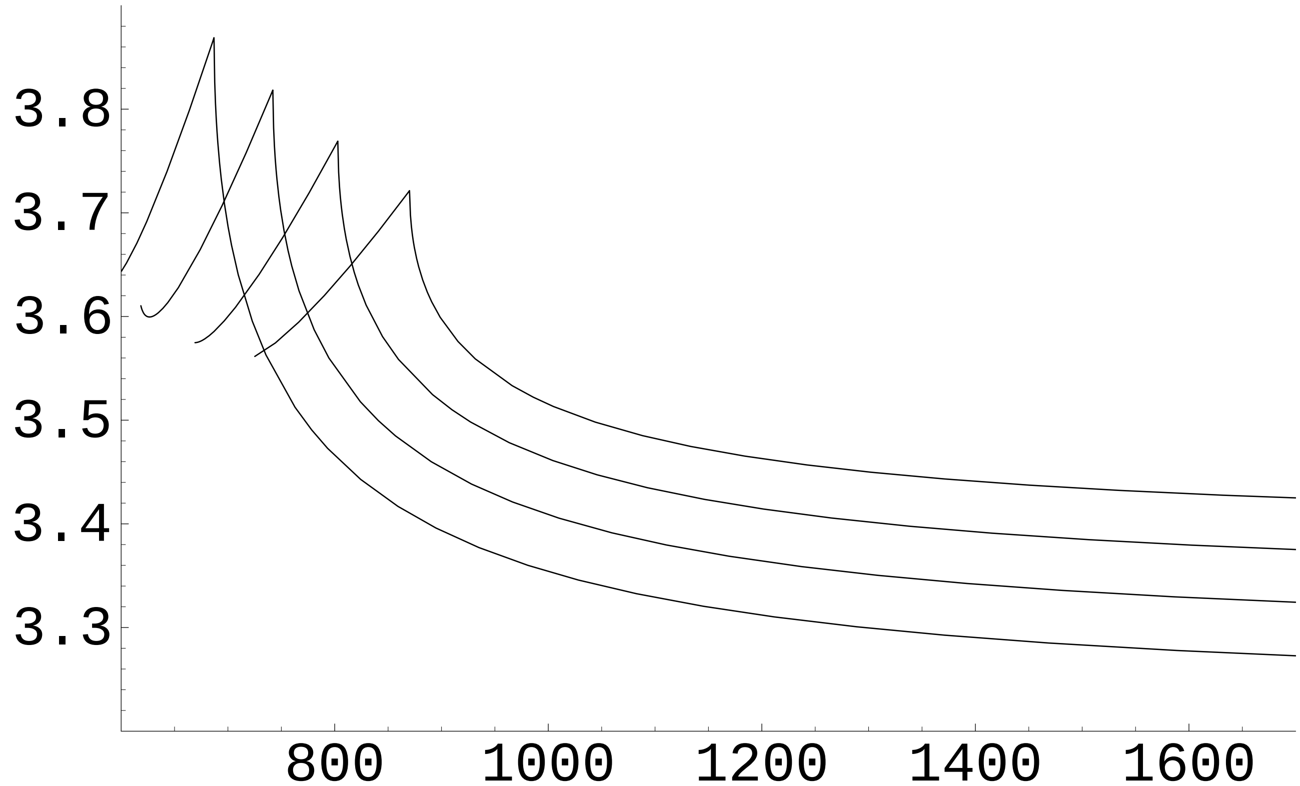

Refractive index vs wavelength (nm)
$x =$0, 0.1, 0.2, 0.3

Slowly varying envelope approx.

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- Combine Maxwell's equations $\rightarrow$ wave equation
- Fourier transform: $t \rightarrow \omega$
- Substitute, with $k = n\omega/c$

$$\mathbf{E}(\omega) = \hat{\mathbf{E}}(\omega, z)\mathrm{e}^{\mathrm{i}kz}$$

- Assume envelope $\hat{\mathrm{E}}(\omega, z)$ varies slowly in comparison to wavelength of light
  - same as paraxial wave equation (BPM)

$$\frac{\mathrm{d}\hat{\mathbf{E}}}{\mathrm{d}z} = -\frac{\alpha}{2}\hat{\mathbf{E}} + \frac{\mathrm{i}\omega^2\mu_0}{2k}\mathbf{P}^{\mathrm{NL}}\mathrm{e}^{-\mathrm{i}kz}$$

Second-order nonlinearities

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- No second-order nonlinearity in media with inversion symmetry
- Crystal symmetry in cubic semiconductors specifies that the only non-zero tensor elements are

$$\chi^{(2)}_{xyz} = \chi^{(2)}_{yzx} = \chi^{(2)}_{zxy}$$

- More independent tensor elements in heterostructures and at surfaces

Pockels Electro-optic effect

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- Pockels Electro-optic effect is $0 + \omega \rightarrow \omega$ and appears as a modification to the refractive index
- transverse geometry, e.g. DC/RF field $\perp$ surface $\rightarrow$ phase modulation for TE optical polarisation
- conventionally use reduced r tensor notation so that optical path length change is

$$\Delta n L = \frac{n^3 r_{41} V L}{2d}$$

- GaAs at $\lambda =$1.5 μm, $r_{41} =$1.36 pmV^{-1} and $n =$3.38

Frequency conversion by $\chi^{(2)}$

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- Optical frequency conversion (define $\omega_3 = \omega_1 + \omega_2$)
  - sum frequency generation: $\omega_1 + \omega_2 \longrightarrow \omega_3$
  - second-harmonic generation: $\omega + \omega \longrightarrow 2\omega$
  - difference frequency generation: $\omega_3 - \omega_1 \longrightarrow \omega_2$
  - parametric amplification: $\omega_3 - \omega_1 \longrightarrow$ amplifies ω_1
- reduced d tensor notation, $d = \chi^{(2)}(\omega, \omega)/2$.
- For epitaxial GaAs at $\lambda =$4.1 μm, $d_{14} =$94 pmV^{-1}.
- For continual forward energy conversion, require *Phase-matching*, i.e. phase velocities of generating and generated waves must be identical.

Second harmonic generation

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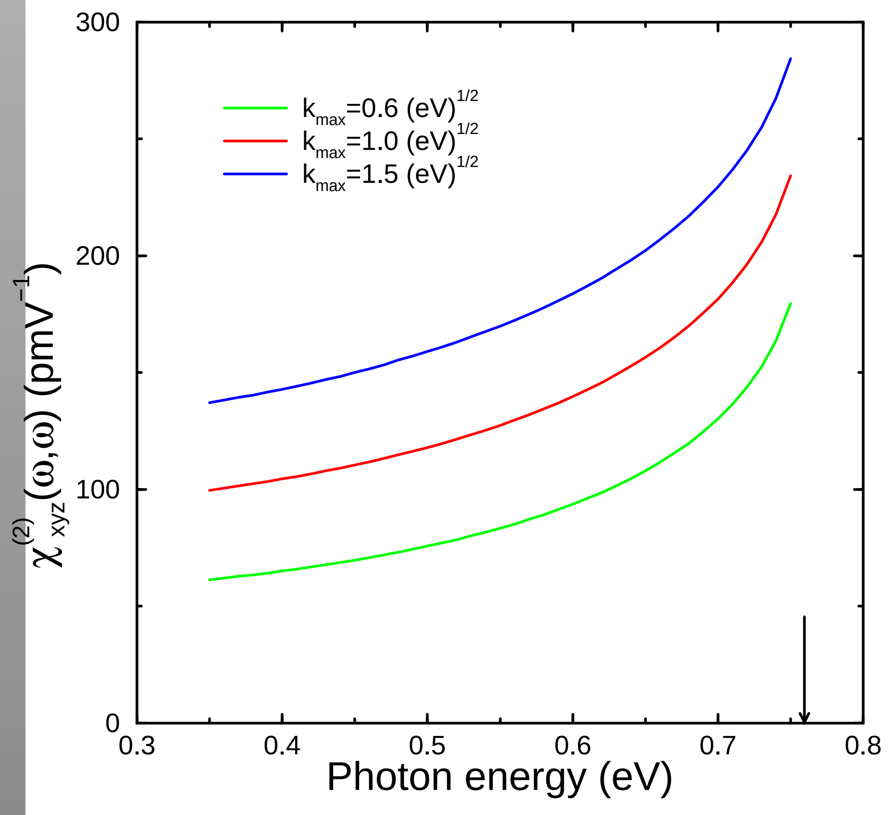

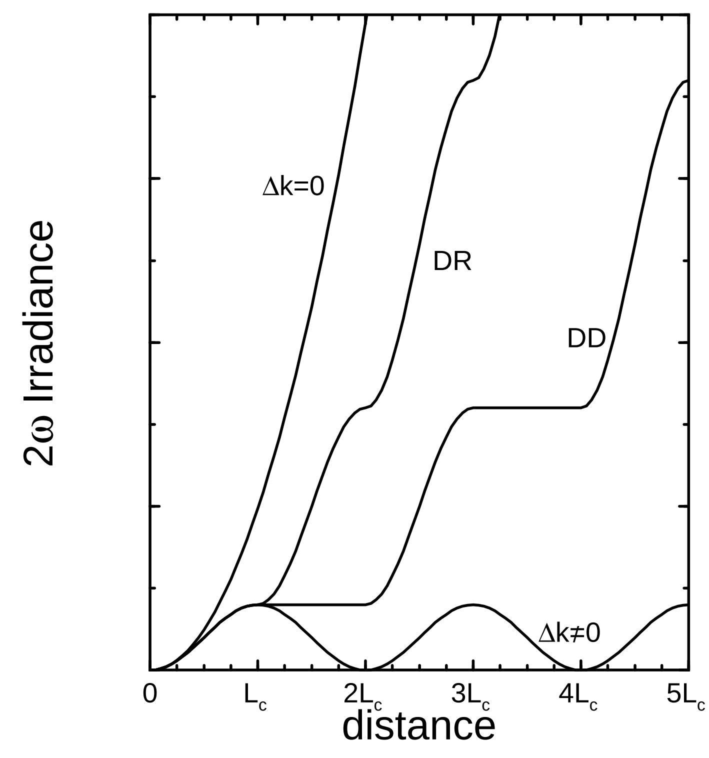

Second harmonic generation

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- Simplest setup to characterise nonlinear interaction
- type-I configuration: TE fundamental → TM SH
- type-II configuration: TE and TM fundamental → TE SH

Difference Frequency Generation

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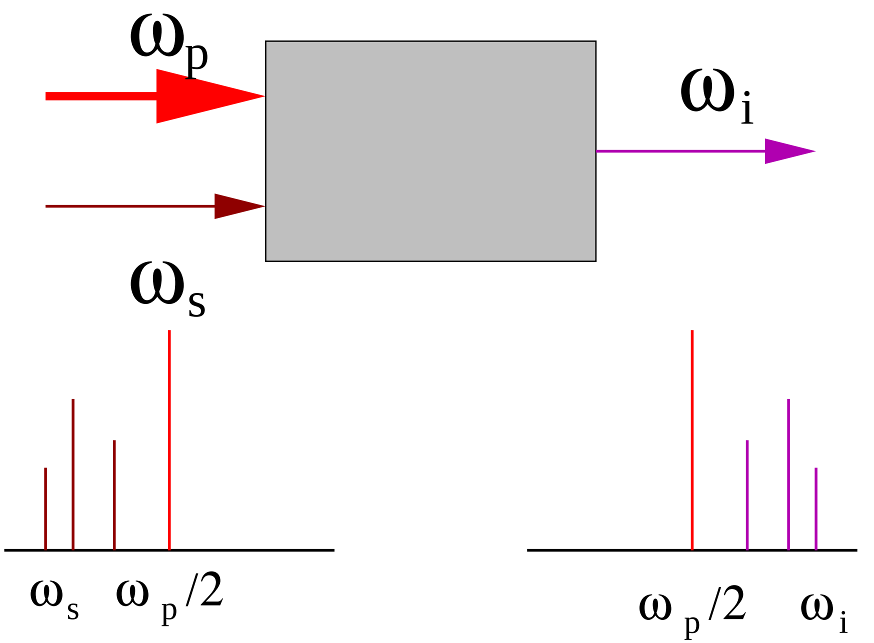

- Channel conversion for WDM
- Potentially integrate pump laser on chip

Parametric Amplification

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- Broad-bandwidth amplifier
- Optical Parametric Oscillator for mid-IR — use cavity and ω_s builds up from noise
- Potentially integrate resonator
- Potentially integrate pump laser on chip

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

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- third-harmonic generation $\propto \chi^{(3)}(\omega,\omega,\omega)$
- two-photon absorption $\Delta\alpha = \beta I$

$$\beta(\omega) = \frac{3\omega}{2\varepsilon_0 n_0^2 c^2} \mathrm{Im}\chi^{(3)}_{\mathrm{eff}}(-\omega,\omega,\omega)$$

- nonlinear refraction $\Delta n = n_2 I$

$$n_2(\omega) = \frac{3}{4\varepsilon_0 n_0^2 c} \mathrm{Re}\chi^{(3)}_{\mathrm{eff}}(-\omega,\omega,\omega)$$

- DC Kerr effect $\propto \chi^{(3)}(0,0,\omega)$

$\chi^{(3)}$ *symmetry considerations*

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- 4 independent nonzero tensor elements for bulk semiconductors
  - 3 for single ω
    $\chi^{(3)}_{xxxx}(-\omega,\omega,\omega)$, $\chi^{(3)}_{xyxy}(-\omega,\omega,\omega)$, $\chi^{(3)}_{xxyy}(-\omega,\omega,\omega)$
- Three measurements required to completely characterise each nonlinear process
- Breaking symmetry, e.g. heterostructure introduces many more independent nonzero tensor elements

Two-photon Absorption

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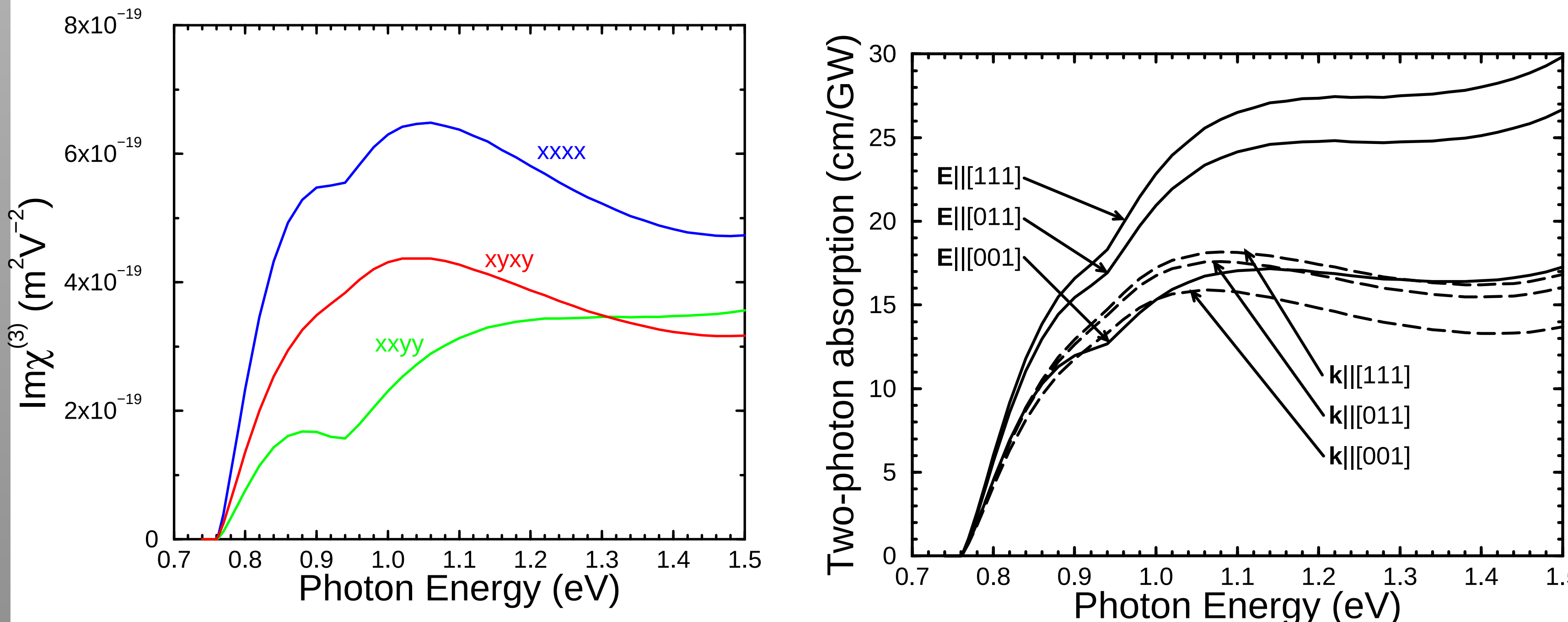

- Calculated $\mathrm{Im}\chi^{(3)}$ tensor elements and spectra of β for GaAs.
- Scales as E_g^{-3}.

Ultrafast Nonlinear Refraction

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- Three measurements completely characterise nonlinear refraction in bulk (cubic) semiconductor

Strength	$n_2^L[001]$	$\Delta n/I\|_{[001]}$
Anisotropy	σ	$2\left\{n_2^L[001] - n_2^L[011]\right\}/n_2^L[001]$
Biref. param.	δ	$\left\{n_2^L[001] - n_2^C(\mathbf{k} \parallel [100])\right\}/n_2^L[001]$

- n_2 scales as E_g^{-4}
- For isotropic Kleinmann: $\sigma = 0$, $\delta = 1/3$
- GaAs at the half-gap (theory): $\sigma = -0.82$, $\delta = 0.08$
- AlGaAs at the half-gap (exper.): $\sigma = -0.54$, $\delta = 0.18$

Coupled Propagation Equations

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- Usual configuration in semiconductor slab waveguides has TE$\|$[110] (cleavage planes) and TM$\|$[001].

$$\mathrm{i}\frac{\partial u}{\partial z}+\frac{\partial^2 u}{\partial x^2}+\gamma u+\left[\left(1-\frac{\sigma}{2}\right)uu^*+\left(1-\delta-\frac{\sigma}{2}\right)vv^*\right]u+\left(\delta-\frac{\sigma}{2}\right)u^*v^2=0$$

$$\mathrm{i}\frac{\partial v}{\partial z}+\frac{\partial^2 v}{\partial x^2}-\gamma v+\left[\left(1-\delta-\frac{\sigma}{2}\right)uu^*+vv^*\right]v+\left(\delta-\frac{\sigma}{2}\right)u^2v^*=0$$

- u and v are the scaled electric field amplitudes for TE and TM.
- γ is proportional to the (structurally induced) birefringence $n_{\mathrm{TM}}-n_{\mathrm{TE}}$.

Calculated $\chi^{(3)}$ *in GaAs*

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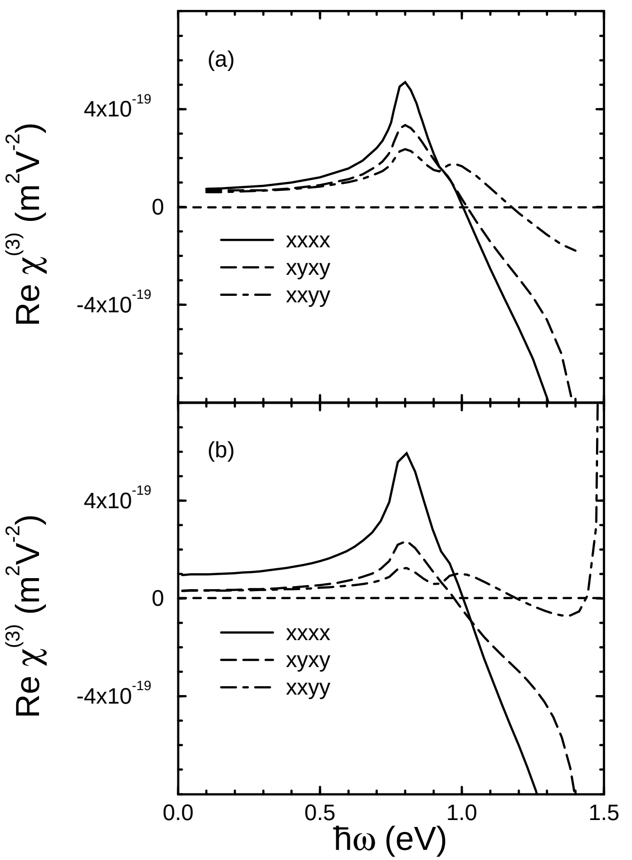

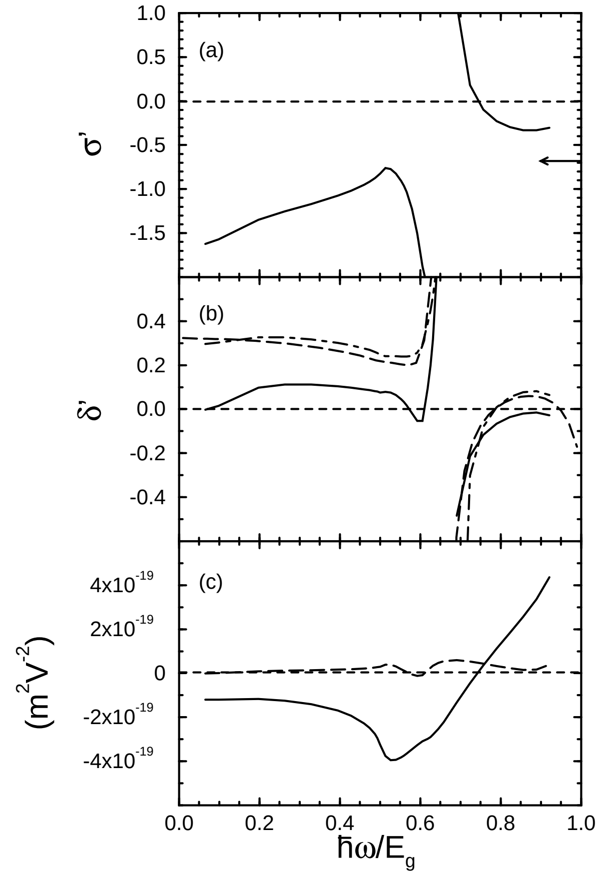

Top: 14-band, bottom: 8-band (isotropic)

n_2 *in Al$_{0.18}$Ga$_{0.82}$As*

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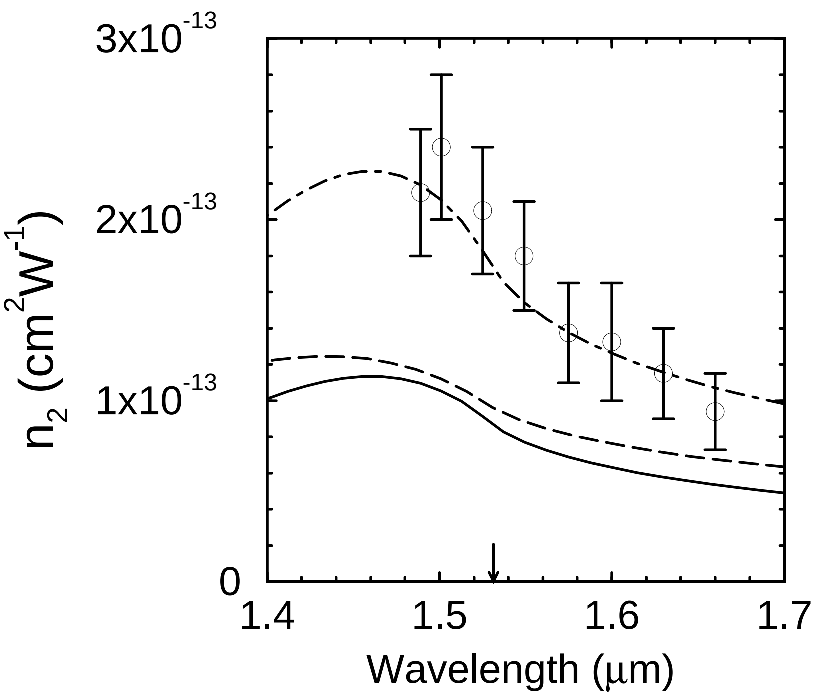

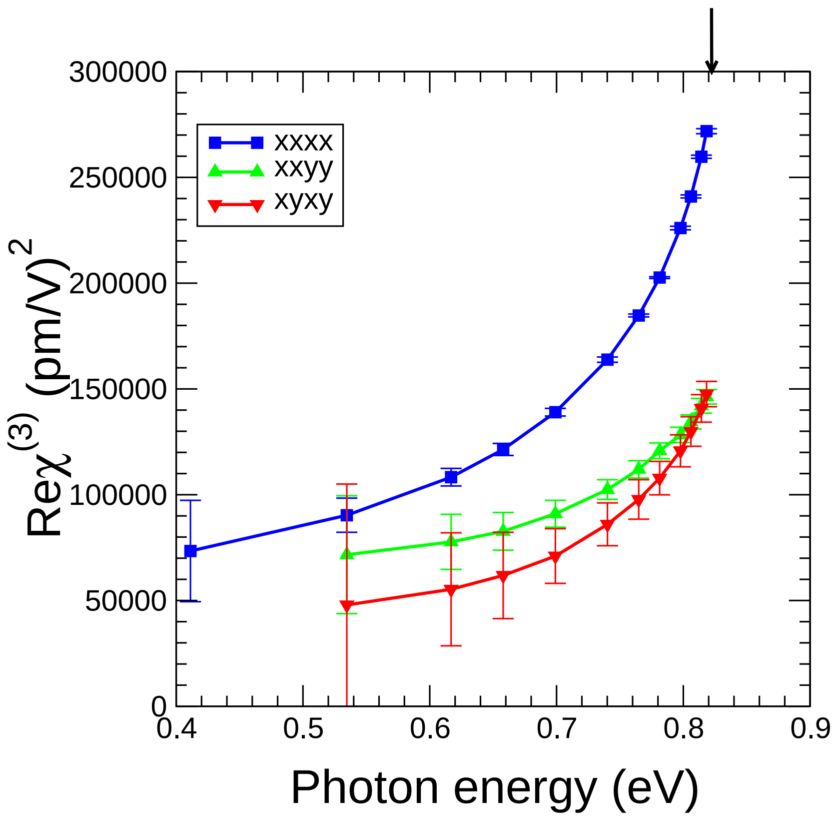

- Measured dispersion of n_2 in Al$_{0.18}$Ga$_{0.82}$As and calculated $\mathrm{Re}\chi^{(3)}$ tensor components

n_2 *in* $Al_{0.18}Ga_{0.82}As$

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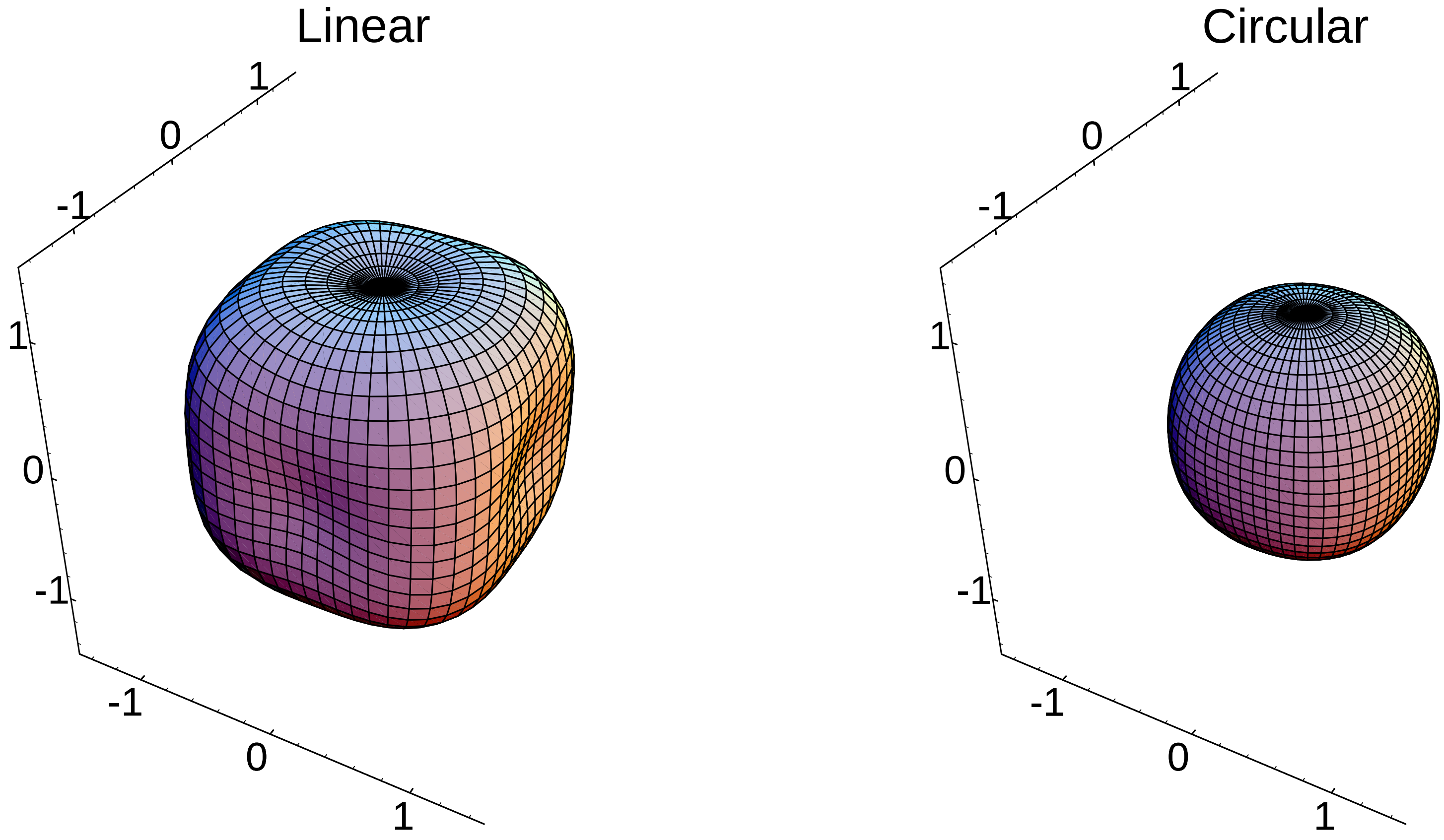

Deduced anistropy from measurements of n_2 in $Al_{0.18}Ga_{0.82}As$ at $\lambda = 1.55\ \mu$m

Figure-of-merit for NLR applications

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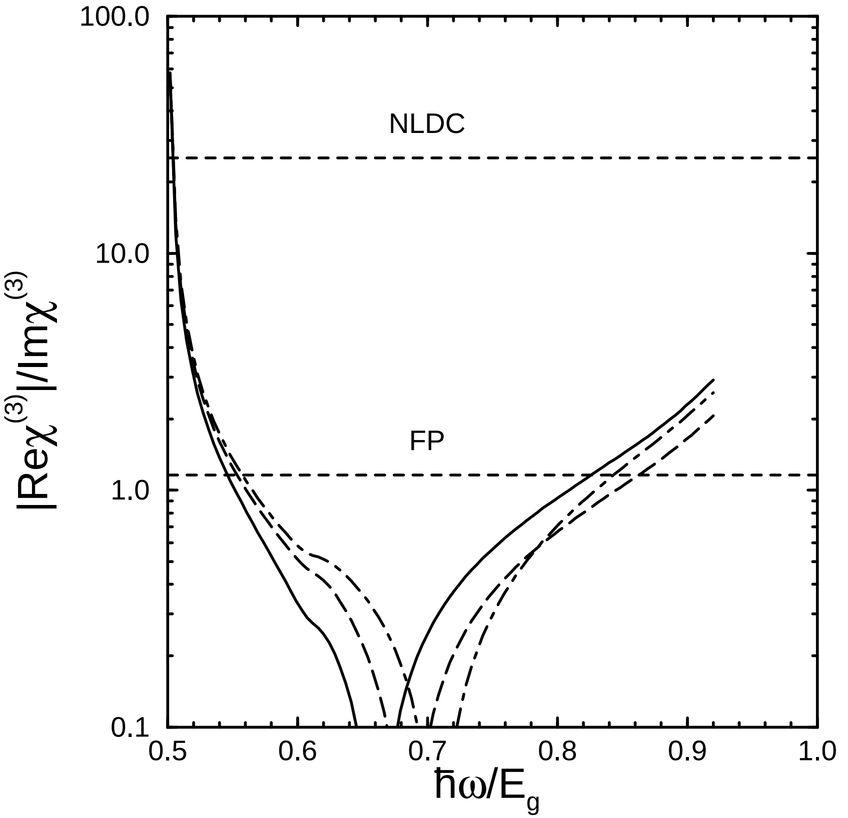
- light absorbed in length α^{-1}
- phase change $2\pi|\Delta n|L/\lambda \sim 2\pi$ for NLO applications
- therefore require figure-of-merit $|\Delta n|/(\alpha\lambda) > 1$
- for $\chi^{(3)}$ only, figure-of-merit $|n_2|/(\beta\lambda) \propto |\mathrm{Re}\chi^{(3)}|/\mathrm{Im}\chi^{(3)}$



Quasi-$\chi^{(3)}$ *processes*

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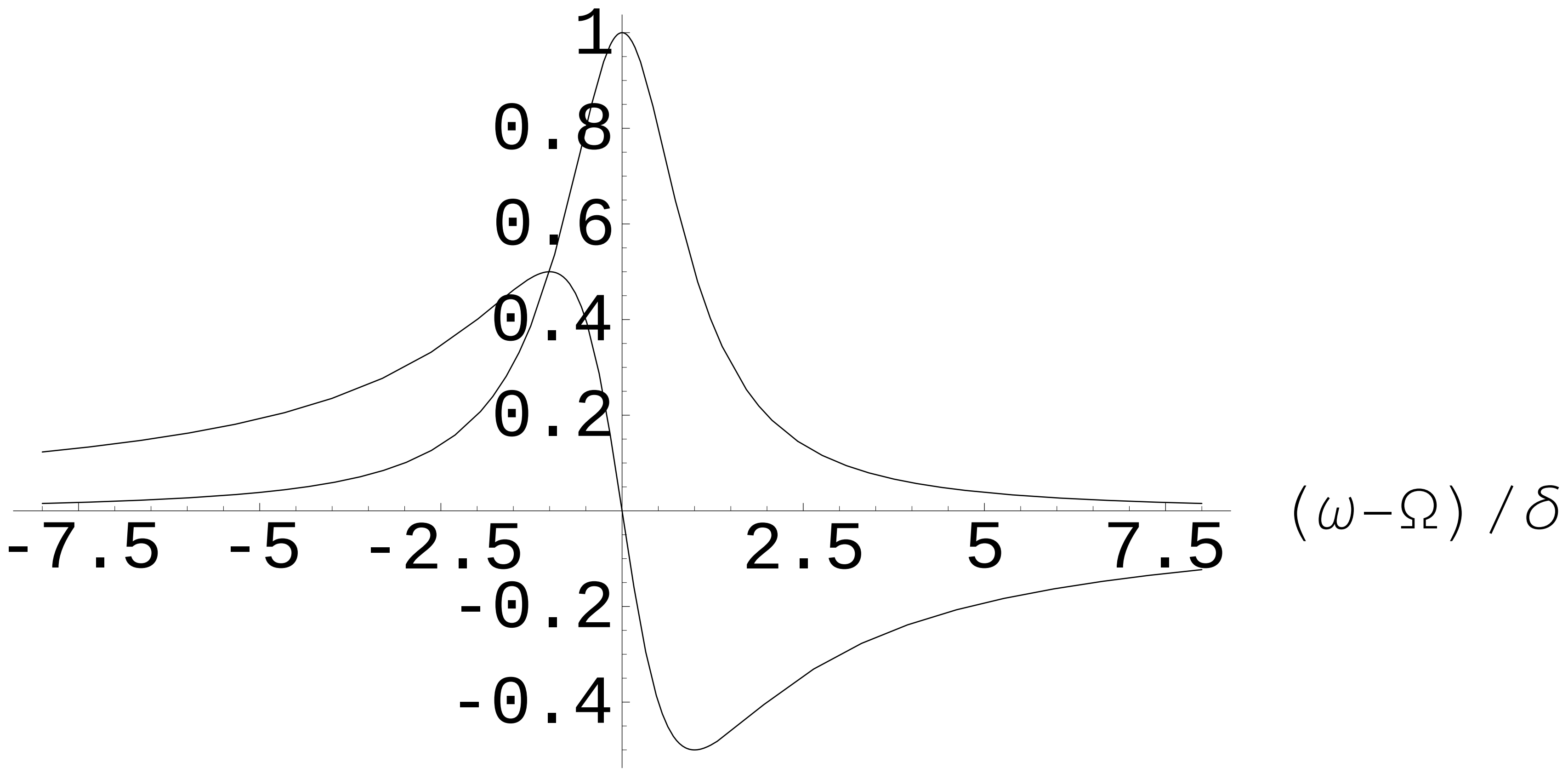

Carrier nonlinearities

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- Free carrier absorption: $N\uparrow$, $\alpha\uparrow$
- Absorption saturation (bandfilling) in passive device: $N\uparrow$, $\alpha\downarrow$, $n\downarrow$
- Gain saturation in active device: $N\downarrow$, gain$\downarrow$, $n\uparrow$
- Exciton absorption saturation (phase-space filling + screening): $N\uparrow$, $\alpha\downarrow$, $n\downarrow$

N.B. effects on n are for below bandgap frequencies

Example: bandfilling

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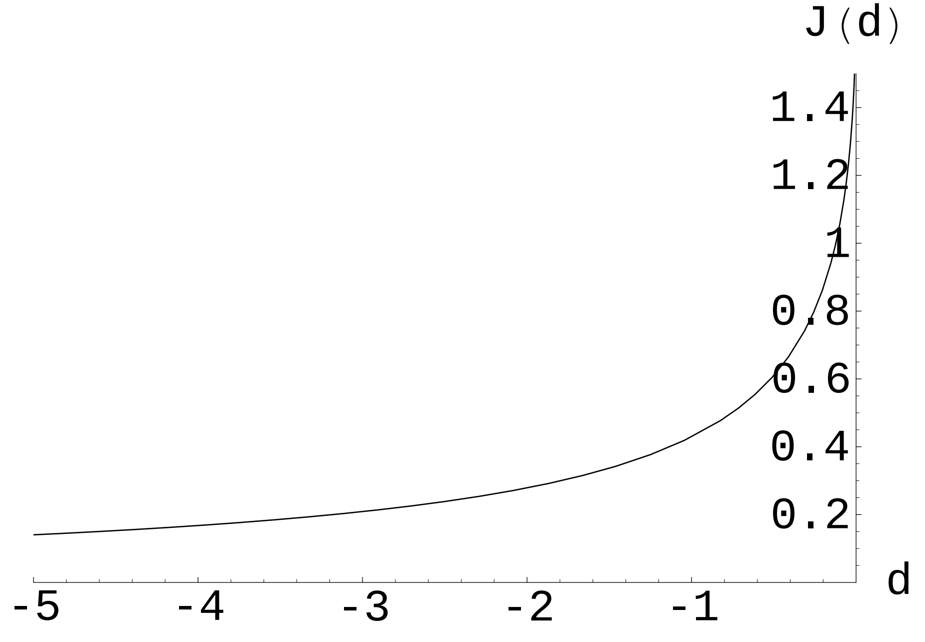
Assuming equal populations N of electrons and heavy-holes which have quasi-equilibrium Boltzmann thermal distribution, we get $\Delta n = \sigma_n N$

$$\sigma_n(\omega) = -\frac{4\sqrt{\pi}}{n_0}\left|\frac{e\mathbf{p}_{vc}}{m_0\omega_g}\right|^2 \frac{1}{k_B T}\sum_{j=hh,lh}\frac{m_{rj}}{m_e} J\left(\frac{m_{rj}}{m_e}\frac{\hbar(\omega-\omega_g)}{k_B T}\right)$$

Carrier nonlinearities

where

$$J(d) = \int_0^{\infty} \frac{\sqrt{x}\mathrm{e}^{-x}\,\mathrm{d}x}{x-d}$$



DC Kerr effects

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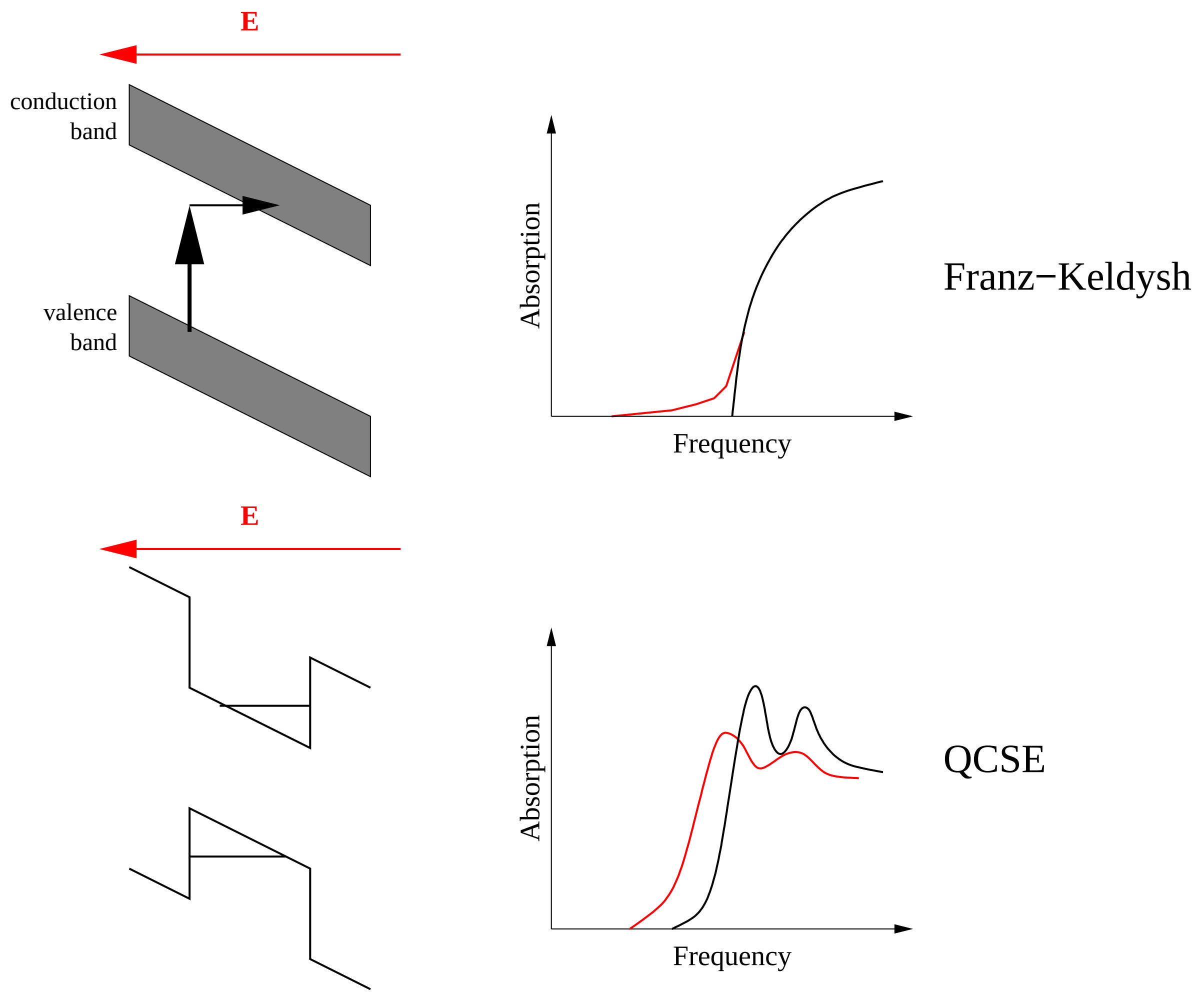
- Franz-Keldysh effect: increases band-tail absorption
- Quantum Confined Stark Effect: shifts exciton resonances to longer wavelength
- Nonlinear absorption as excited carriers screen field (Self electro-optic effect device — SEED)

DC Kerr effects



Thermal effects

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- Absorbed light results in heating of medium and changes linear optical properties

$$\frac{\partial n}{\partial T} = \frac{\partial n}{\partial E_g}\frac{\partial E_g}{\partial T}$$

- For Al$_x$Ga$_{1-x}$As
  $\partial E_g/\partial T = -(3.95 + 1.15x) \times 10^{-4}$ eVK^{-1}.
- Can differentiate Adachi formula for $\partial n/\partial E_g$
  - $\partial n/\partial E_g < 0$ below bandgap
  - giving $\partial n/\partial T > 0$ generally
  - e.g. 6.5×10^{-5} K^{-1} in GaAs at 1.5 μm

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