



Photorefractivity

Nicolas Fressengeas

► **To cite this version:**

| Nicolas Fressengeas. Photorefractivity. DEA. Université Paul Verlaine Metz, 2010. <cel-00520586>

HAL Id: cel-00520586

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Submitted on 23 Sep 2010

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UE SPM-PHO-S09-112

Photorefractivity

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Photorefractive effect

History began in 1966 as *Optical Damage*



Optical Damage in LiNbO_3

- Shine a light on LiNbO_3
- Remove it
- Shine another : **damaged crystal**

Semi-permanent effect

- Leave it in the dark : still damaged
- Leave it under uniform light : sometimes repaired

Today

- Photorefractivity can prove useful
- Some people still call it *optical damage*:
 - Bad for linear optics (electro-optic modulators...)
 - Bad for *instant* Non Linear Optics (SHG, OPA...)

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frame 2

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Band Transport

Harmonic
illumination

Two Wave Mixing

Photorefractivity is attractive

Non linear optics at low optical power



Non Linear Optics

- Dynamic Holography
- Phase conjugation
- All optical computing
- ...
- At milliwatts and below power levels

Observed in many non linear crystals

- Sillenites : $\text{Bi}_{12}\text{SiO}_{20}$, $\text{Bi}_{12}\text{TiO}_{20}$, $\text{Bi}_{12}\text{GeO}_{20}$
- Tungsten-Bronze : $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$
- Ferroelectrics : LiNbO_3 , BaTiO_3
- Semiconductors : $\text{InP}:\text{Fe}$, AsGa

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Usefull reading. . .

[Yeh93, GH88, GH89]




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Band Transport

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Two Wave Mixing

 P. Günter and J. P. Huignard.
Photorefractive materials and their applications I,
volume 61 of *Topics in Applied Physics*.
Springer Verlag, Berlin, 1988.

 P. Günter and J. P. Huignard.
Photorefractive materials and their applications II,
volume 62 of *Topics in Applied Physics*.
Springer Verlag, Berlin, 1989.

 P. Yeh.
Introduction to photorefractive nonlinear optics.
Wiley Interscience, New York, 1993.

1 Band Transport Model

- Schematics
- Carrier Generation
- Charge Transport
- Electro-optic effect

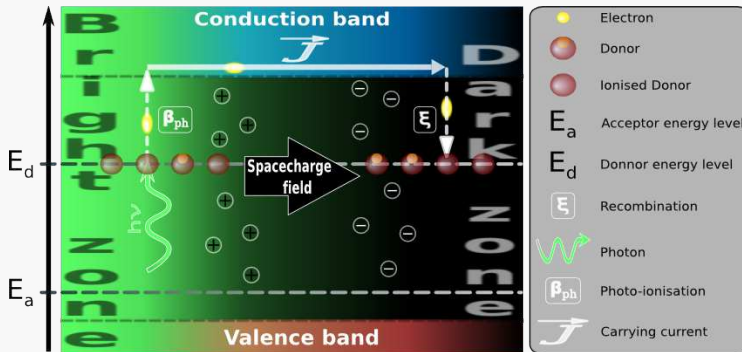
2 Harmonic illumination

- Harmonic framework
- Uniform background: order 0
- Periodic modulation : order 1
- Implications, Simplifications, Diffusion and Saturation

3 Two Wave Mixing

- Gratings graphical view
- Coupled waves
- Two Beam Coupling

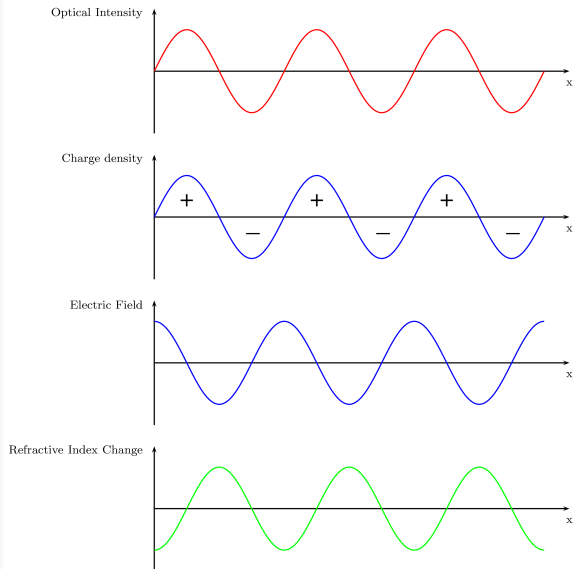
Photorefractive charge transport and trapping



Linear Index Modulation

Space charge electric field generates refractive index variation through electro-optic effect

Periodic illumination of a photorefractive material



Donors wanted

Carriers are generated by donors: no donors, no carriers



Nominally pure crystals

- No in-band-gap level
 - No donor nor acceptor
 - No photorefractive effect
- Structural **defects** often present
- As well as pollutants
- They create **in-band-gap levels**
- **Photorefractivity** can arise from them

More efficient: doping

- Introduce in-band-gap species
- $\text{LiNbO}_3:\text{Fe}$, $\text{InP}:\text{Fe}$...

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Band Transport

Schematics

Carrier Generation

Charge Transport

Electro-optics

Harmonic
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Two Wave Mixing

Introducing Donors and Acceptors



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Introduce Donors of electrons

N_D

- Energy level close to conduction band
- They easily **give electrons** to conduction band

Introducing Acceptors

$N_A \ll N_D$

- Photorefractivity needs traps
- Ionized donors are traps
- Introduce Acceptors close to the valence band
- They **catch Donors electrons**
- Donors are partially ionized $N_D^+ = N_A$

Thermal carrier generation

We assume here that the only carriers are electrons. . . what if not?

n_e



Comes from temperature induced Brownian motion

- Temperature induced
- Electrons are **kicked** into conduction band

Rate proportional to donors-left-to-ionize density

$$\frac{\partial n_e}{\partial t} = \beta (N_D - N_D^+)$$

One generated electron leaves one ionized donor

$$\frac{\partial n_e}{\partial t} = \frac{\partial N_D^+}{\partial t} = \beta (N_D - N_D^+)$$

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Two Wave Mixing

Photo-excitation of carriers

The photoelectric effect at work



Photoelectric effect

- Photon **energy sufficient** to reach conduction band
- Rate proportional to light intensity \mathcal{I}
- And to left-to-ionize donors

Photo-excitation rate

$$\frac{\partial n_e}{\partial t} = \frac{\partial N_D^+}{\partial t} = \sigma \mathcal{I} (N_D - N_D^+)$$

The photo-ionization cross section

σ

- Has the dimensions of a surface
- If \mathcal{I} is given as a number of photon per surface units and time
- Sometimes the case, sometimes not. . .

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Two Wave Mixing

Recombination needs luck, electrons and empty traps

- A luck factor ξ
- Carriers density n_e
- Empty trap density N_D^+

$$\frac{\partial n_e}{\partial t} = \frac{\partial N_D^+}{\partial t} = -\xi n_e N_D^+$$

Carriers rate equation

A combination of generation and recombination



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A combination of generation and recombination

$$\frac{\partial N_D^+}{\partial t} = (\beta + \sigma \mathcal{I}) (N_D - N_D^+) - \xi n_e N_D^+$$

Charge transport in the conduction band

An assumption of the Band Transport Model. . . sometimes untrue



Diffusion

- Due to Temperature and Brownian motion
- Think of it as **sugar in water** (or coffee)
- Depends on concentration **variations**

Drift under electric-field

- Needs electric-field
- Externally applied or diffusion generated
- Depends on **electric field** and **mobility**

Photovoltaic effect

- Sometimes called *photo-galvanic*
- Non-isotropic effect
- Think of solar cells: **light generates current**
- Depends on light intensity

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Diffusion transport

Diffusion current from Fick's first law linked to Einstein relation



Fick's first law

Particle flow

$$\vec{J}_p = -D \text{grad}(p)$$

Einstein relation

- Links diffusion, absolute temperature T and Brownian motion through mobility
- Mobility μ is the velocity to electric field ratio

$$D = \mu_{\tilde{e}} k_B T / e$$

Diffusion Current

$$\vec{J} = -e \vec{J}_{\tilde{e}} = +\mu_{\tilde{e}} k_B T \text{grad}(n_{\tilde{e}})$$

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Drift

A charged particle in an electric field...



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Electric Field



- Externally applied
- Due to charged carrier diffusion

Drift current

- Electrons velocity: $\vec{v} = -\mu_e \vec{E}$
- Drift Current: $\vec{J} = -e \times \vec{v}$
 $\vec{J} = en_e \mu_e \vec{E}$

Band Transport

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illumination

Two Wave Mixing

Photovoltaic current

An non isotropic effect stemming from crystal asymmetry



Origins

- Non centro-symmetric crystal
- Anisotropic photo-electric effect
- Depends on light polarization

e.g. LiNbO_3

Photovoltaic tensor

- Rank 2
- Main component along polar axis
- Often reduced to a scalar

Photovoltaic Current

$$\begin{aligned} [\vec{J}]_i &= (N_D - N_D^+) \sum_{j,k} [\beta^{ph}]_{j,k} [\vec{E}]_j [\vec{E}]_k \vec{u}_i \\ \vec{J} &\approx \beta^{ph} \mathcal{I} (N_D - N_D^+) \vec{c} \end{aligned}$$

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Band Transport

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Band Transport Model

Also known as Kukhtarev model

Published in 1979



Ionized donors rate equation

$$\frac{\partial N_D^+}{\partial t} = (\beta + \sigma \mathcal{I}) (N_D - N_D^+) - \xi n_{\bar{e}} N_D^+$$

Current density expression

$$\vec{\mathcal{J}} = \mu_{\bar{e}} k_B T \text{grad} (n_{\bar{e}}) + e n_{\bar{e}} \mu_{\bar{e}} \vec{E} + \beta^{ph} \mathcal{I} (N_D - N_D^+) \vec{c}$$

Quasi-static Maxwell model

- Continuity : $\text{div}(\vec{\mathcal{J}}) + \frac{\partial \rho}{\partial t} = 0$
- Charge : $\rho = e (N_D^+ - N_A^- - n_{\bar{e}})$
- Maxwell-Gauss : $\text{div}(\vec{D}) = \rho$, with $\vec{D} = \hat{\epsilon} \vec{E}$

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Two Wave Mixing

Refractive index modulation through electro-optics

Space-charge electric field induces refractive index variations



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Electro-optics

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Two Wave Mixing

This is not an electro-optics lesson

Please refer to the electro-optics lesson

Anyhow. . .

- Light generated electric field: the **space charge field** \vec{E}
- In electro-optic materials : creates index modulation

$$\left[\Delta \frac{1}{n^2}\right]_{i,j} = \sum_k [\hat{r}]_{ijk} \left[\vec{E}\right]_k$$

- Local modulation of refractive index
- Local modification of refractive index ellipsoid

Periodic illumination from plane waves interference



Two plane waves interfering

- Same wavelength and coherent
- non collinear wave vectors \vec{k}_1 and \vec{k}_2 : $\vec{K} = \vec{k}_2 - \vec{k}_1$
- Interference pattern : $\mathcal{I}_{(0)} \left(1 + m \cos \left(\vec{K} \cdot \vec{r} \right) \right)$
 - $\mathcal{I}_{(0)} = \mathcal{I}_1 + \mathcal{I}_2$
 - $m = 2 \frac{\sqrt{\mathcal{I}_1 \mathcal{I}_2}}{\mathcal{I}_1 + \mathcal{I}_2}$

Harmonic assumptions

- $m \ll 1$: intensities are very different
- All unknowns are sum of
 - A large uniform background : order 0
 - A small harmonic modulation : order 1
- Linearity : orders can be uncoupled
 - Uniform intensity analysis
 - Followed by **small signal** analysis

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Band Transport

Harmonic
illumination

Harmonic framework

Order 0

Order 1

Simplification and
Consequences

Two Wave Mixing

Simplifying assumptions



One dimension problem

1D

- Plane waves interference
- All phenomena are collinear to \vec{K}

Drift-diffusion transport only assumed

Photovoltaic effect assumed negligible

Photo-generation only assumed

Large intensities: thermal generation can be neglected

Steady state study

All temporal derivatives assumed zero

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Harmonic framework

Order 0

Order 1

Simplification and
Consequences

Two Wave Mixing

Carrier generation–recombination equilibrium

Steady state equilibrium

$$\sigma \mathcal{I} (N_D - N_D^+) = \xi n_{\bar{e}} N_D^+$$

Uniform electric field

- $\vec{D}_{(0)}$ is homogeneous
- $\text{div}(\vec{D}_{(0)}) = \rho_{(0)} = 0$
- $N_{D(0)}^+ - N_A^- - n_{\bar{e}(0)} = 0$

Small illumination

- $n_{\bar{e}} \ll N_A$
- $\sigma \mathcal{I} \ll \xi N_A$

Equilibrium homogeneous densities

- $N_{D(0)}^+ = N_A + n_{\bar{e}(0)}$
- $n_{\bar{e}(0)} = \frac{N_D - N_A}{\xi N_A} \sigma \mathcal{I}_{(0)}$

Order 1 framework

Basic multi-scale modeling



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Band Transport

Harmonic
illumination

Harmonic framework

Order 0

Order 1

Simplification and
Consequences

Two Wave Mixing

All quantities are assumed periodic

- $\operatorname{div}(\vec{X}) = i\vec{K} \cdot \vec{X}$
- ...

Order 0 assumed known

Order 1 assumed small

- $\forall X, X_{(1)} \ll X_{(0)}$
- $(X \times Y)_{(1)} = X_{(0)} Y_{(1)} + X_{(1)} Y_{(0)}$
- Order 0 is independently found
- Order 1 products is Order 2 : assumed negligible

Order one charge and current equilibrium

Steady state equilibrium

- $\sigma \mathcal{I} (N_D - N_D^+) = \xi n_{\bar{e}} N_D^+$
- $\sigma \mathcal{I}_{(1)} (N_D - N_{D(0)}^+) + \sigma \mathcal{I}_{(0)} (-N_{D(1)}^+) = \xi n_{\bar{e}(0)} N_{D(1)}^+ + \xi n_{\bar{e}(1)} N_{D(0)}^+$

Harmonic Current density

$$\vec{\mathcal{J}}_{(1)} = \mu_{\bar{e}} k_B T i n_{\bar{e}(1)} \vec{\mathcal{K}} + e \mu_{\bar{e}} n_{\bar{e}(1)} \vec{E}_{(1)}$$

Harmonic Current density divergence is null $i \vec{\mathcal{K}} \cdot \vec{\mathcal{J}} = 0$

$$i \vec{\mathcal{K}} \cdot \left(\mu_{\bar{e}} k_B T i n_{\bar{e}(1)} \vec{\mathcal{K}} + e \mu_{\bar{e}} n_{\bar{e}(1)} \vec{E}_{(1)} \right) = 0$$

Harmonic Poisson

$$i \vec{\mathcal{K}} \cdot \left(\hat{\epsilon} \cdot \vec{E}_{(1)} \right) = e \left(N_{D(1)}^+ - n_{\bar{e}(1)} \right)$$

Order One Space Charge Field

General Expression

$$\vec{E}_{(1)} = \frac{i\vec{K} \frac{k_B T}{e} - \frac{\vec{K} \cdot \mu E_{(0)}}{\langle \mu \rangle}}{1 + \frac{\|\vec{K}\|^2}{k_D^2} + i \frac{e}{k_B T} \frac{\vec{K} \cdot \mu E_{(0)}}{k_D^2 \langle \mu \rangle}} \frac{\mathcal{I}_{(1)}}{\mathcal{I}_{(0)}}$$

Effective permittivity

$$\langle \varepsilon \rangle = \frac{\vec{K} \cdot \hat{\varepsilon} \vec{K}}{\|\vec{K}\|^2}$$

Effective permeability

$$\langle \mu \rangle = \frac{\vec{K} \cdot \mu \vec{K}}{\|\vec{K}\|^2}$$

Debye vector

$$k_D = \frac{2\pi}{\lambda_D}$$

$$k_D^2 = \frac{e}{\langle \varepsilon \rangle} \frac{e}{k_B T} \frac{N_D}{N_A} (N_D - N_A)$$

Let's simplify this complex expression

General Expression

$$\vec{E}_{(1)} = \frac{i\vec{K} \frac{k_B T}{e} \mathcal{I}_{(1)}}{1 + \frac{\|\vec{K}\|^2}{k_D^2} \mathcal{I}_{(0)}}$$

Simplifying assumptions

- Very often $\vec{E}_{(1)} \parallel \vec{K}$
- When no field is applied : $\vec{E}_{(0)} = 0$
- Quarter period phase shift between Intensity and Space-Charge Field gratings

Space charge field vs. grating spacing

$$\Lambda = 2\pi / \|\vec{K}\|$$



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Band Transport

Harmonic
illumination

Harmonic framework

Order 0

Order 1

Simplification and
Consequences

Two Wave Mixing

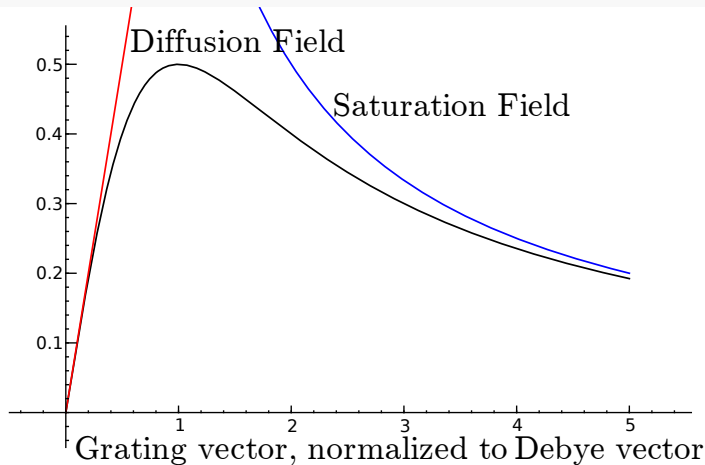
Large grating spacing

- Small \vec{K}
- $\vec{E}_{(1)} = i\vec{K} \frac{k_B T}{e} \frac{\mathcal{I}_{(1)}}{\mathcal{I}_{(0)}}$
- Diffusion field : $\vec{E}_d = \vec{K} \frac{k_B T}{e}$

Small grating spacing

- Large \vec{K}
- $\vec{E}_{(1)} = i\vec{K} \frac{k_B T}{e} \frac{k_D^2}{\|\vec{K}\|^2} \frac{\mathcal{I}_{(1)}}{\mathcal{I}_{(0)}}$
- Saturation Field : $\vec{E}_q = \vec{K} \frac{k_B T}{e} \frac{k_D^2}{\|\vec{K}\|^2}$

Space charge field as a function of grating spacing



Space charge field with externally applied field

No applied field

$$\vec{E}_{(1)} = i \frac{\vec{E}_d}{1 + \frac{E_d}{E_q}} \frac{\mathcal{I}_{(1)}}{\mathcal{I}_{(0)}}$$

Applied field \vec{E}_a

$$\vec{E}_{(1)} = i \frac{\vec{E}_d}{1 + \frac{E_d}{E_q}} \left[\frac{1 + i \frac{E_a}{E_d}}{1 + i \frac{E_a}{E_d + E_q}} \right] \frac{\mathcal{I}_{(1)}}{\mathcal{I}_{(0)}}$$

Band Transport

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Harmonic framework

Order 0

Order 1

Simplification and
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Standard approximations

- For most gratings and materials : $E_d \ll E_q$
- Applied field in the middle : $E_d \ll E_a \ll E_q$

In phase¹ illumination and space charge gratings

$$\vec{E}_{(1)} = - \frac{\vec{E}_a}{1 + \frac{E_d}{E_q}} \frac{\mathcal{I}_{(1)}}{\mathcal{I}_{(0)}}$$

¹Actually, they are π phase shifted. A possible negative sign on the electro-optic coefficient renders in-phase index and illumination gratings.

In phase intensity and index gratings

Beam interference is destructive, owing to reflection sign reversal



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Band Transport

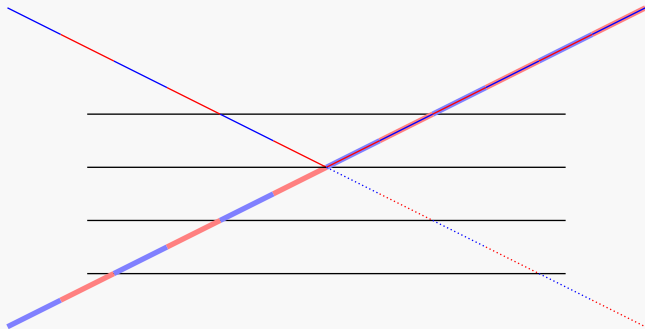
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Two Wave Mixing

Gratings graphical view

Coupled waves

Two Beam Coupling



Quarter period shifted gratings

Beam interference is constructive



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Band Transport

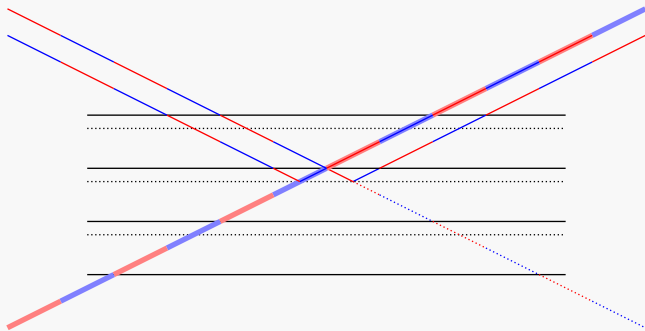
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Two Wave Mixing

Gratings graphical view

Coupled waves

Two Beam Coupling



Two waves and a grating

Two waves make an intensity grating

- Waves are coherent and same wavelength
- Wave vectors are \vec{k}_1 and \vec{k}_2
- Intensity grating vector is $\vec{K} = \vec{k}_2 - \vec{k}_1$
- Waves amplitudes are $A_i = \sqrt{I_i} e^{-i\psi_i}$

Index Grating

- Assume $E_d \ll E_a \ll E_q$
- Index grating $\propto \Phi$ shifted illumination grating

$$n = n_{(0)} + \mathcal{R}e \left[n_{(1)} e^{i\Phi} \frac{\overline{A_1} A_2}{I_{(0)}} e^{\vec{K} \cdot \vec{r}} \right]$$

- $\Phi = \pi/2$ if no applied field and $\Phi = 0$ if field applied

Slow Varying Approximation

Paraxial Framework

- Propagation equation : $\Delta A + \frac{\omega^2}{c^2} n^2 A = 0$
- SVA: $\left\| \frac{\partial^2 A}{\partial z^2} \right\| \ll \left\| \beta \frac{\partial A}{\partial z} \right\|$
- β such as $\beta z = \vec{k} \cdot \vec{r}$

Conventions

- $z = 0$: entrance in the photorefractive material
- Symmetric coupling : $\beta_1 = \beta_2 = \|\vec{k}\| \cos(\theta)$
 θ is the half angle between input beams

After Coupled Mode calculations²

- $\frac{\partial A_1}{\partial z} = -\frac{1}{2L_{(0)}} \Gamma \|A_2\|^2 A_1 - \alpha A_1$
- $\frac{\partial A_2}{\partial z} = -\frac{1}{2L_{(0)}} \bar{\Gamma} \|A_1\|^2 A_2 - \alpha A_2$
- $\Gamma = i \frac{2\pi n_{(1)}}{\lambda \cos(\theta)} e^{-i\Phi}$
- α is absorption

²See lessons on Second Harmonic Generation and Optical Phase Conjugation for details.

Intensity and phase coupling

Diffusion induces intensity coupling

Drift induces phase coupling



Separate Diffusion and Drift influences

$$\Gamma = \gamma + 2i\zeta$$

$$\gamma = \frac{2\pi n_{(1)}}{\lambda \cos(\theta)} \sin(\Phi)$$

$$\zeta = \frac{\pi n_{(1)}}{\lambda \cos(\theta)} \cos(\Phi)$$

Intensity coupling

- $\frac{\partial \mathcal{I}_1}{\partial z} = -\gamma \frac{\mathcal{I}_1 \mathcal{I}_2}{\mathcal{I}_1 + \mathcal{I}_2} - \alpha \mathcal{I}_1$
- $\frac{\partial \mathcal{I}_2}{\partial z} = +\gamma \frac{\mathcal{I}_1 \mathcal{I}_2}{\mathcal{I}_1 + \mathcal{I}_2} - \alpha \mathcal{I}_2$

Phase coupling

- $\frac{\partial \psi_1}{\partial z} = \zeta \frac{\mathcal{I}_2}{\mathcal{I}_1 + \mathcal{I}_2}$
- $\frac{\partial \psi_2}{\partial z} = \zeta \frac{\mathcal{I}_1}{\mathcal{I}_1 + \mathcal{I}_2}$

Energy transfer

- For small absorption α , energy is transferred from one beam to the other
- Transfer direction is given by sign of γ

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Two Wave Mixing

Gratings graphical view

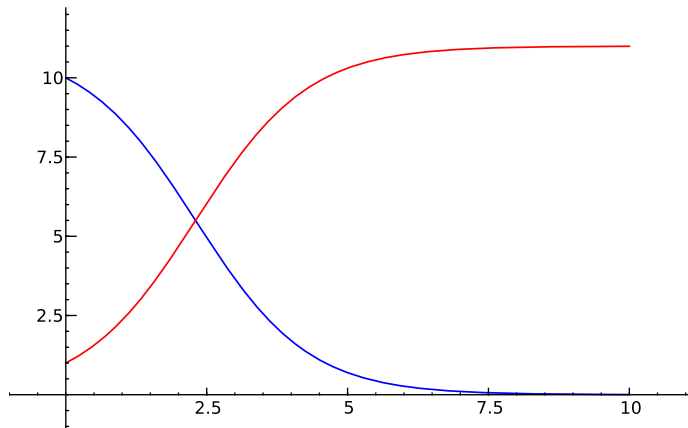
Coupled waves

Two Beam Coupling

Coupled Modes Solution

- Let $m = \frac{\mathcal{I}_1(0)}{\mathcal{I}_2(0)}$
- $\mathcal{I}_1(z) = \mathcal{I}_1(0) \frac{1 + m^{-1}}{1 + m^{-1}e^{\gamma z}} e^{-\alpha z}$
- $\mathcal{I}_2(z) = \mathcal{I}_2(0) \frac{1 + m}{1 + me^{-\gamma z}} e^{-\alpha z}$

Two Wave Mixing Intensity Coupling



Two Wave Mixing Intensity Coupling with Absorption



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Coupled waves

Two Beam Coupling

