13
Mechanisms for the nonlinear refractive index

C. Durfee PHGN 585
Colorado School of Mines

Several mechanisms can lead to a nonlinear refractive index

- Electronic
  - Anharmonic binding potential, fast < fs
- Molecular Rotation
  - Re-orientation of molecule, ~ ps
- Thermal (dn/dT)
  - Heating of lattice, expansion, slow - ~ ms to sec
- Ionization
  - Free electron density changes index
- Semiconductor
  - conduction band population changes fast
- Relativistic
  - Oscillating electron mass shift changes index

Electronic response

- $n_2 \sim 3 \times 10^{-16}$ cm^2/W (e.g. in sapphire)
- Example: for L = 10mm, λ=0.8um,

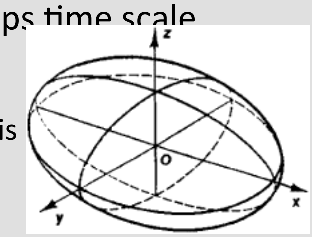
  We need $I = 4.2 \times 10^{10}$ W/cm^2 for B = 1

  If the spot radius = 1mm, duration 100ps:

  max energy = 130mJ
- Note that since potential almost always decreases at large displacement from the nucleus
  - Large amplitude = weaker binding, lower resonance freq
  - Therefore $n_2 > 1$

Molecular electronic response: induced dipole

- This is the dominant response for anisotropic molecular liquids and gases
- Electric field aligns molecules on a ps time scale
- Induced dipole: $p = \alpha E$
  - Polarizability α is larger along long axis
  - Weaker binding, more response
- Polarizability is a tensor $\mathbf{p} = \vec{\alpha} \cdot \mathbf{E}$
  - In molecular coordinates: $\mathbf{p} = \alpha_x E_x \hat{\mathbf{x}} + \alpha_y E_y \hat{\mathbf{y}} + \alpha_z E_z \hat{\mathbf{z}}$
- Permanent dipole is indep of **E**



Molecular alignment in the field

- If applied field is not along an axis, there is a torque

$$\tau = \mathbf{p} \times \mathbf{E}$$

- Use energy instead of force
  - For permanent dipole:

$$U = -\mathbf{p} \cdot \mathbf{E} = -p_{\parallel} E_{\parallel} - p_{\perp} E_{\perp}$$

For uniaxial molecule

  - For induced dipole:

$$dU = -\alpha_{\parallel} E_{\parallel} dE_{\parallel} - \alpha_{\perp} E_{\perp} dE_{\perp} \qquad E_{\perp} = E \sin\theta \qquad E_{\parallel} = E\cos\theta$$

$$U = -\frac{1}{2}\alpha_{\parallel} E_{\parallel}^{2} - \frac{1}{2}\alpha_{\perp} E_{\perp}^{2} = -E^{2}\left(\alpha_{\parallel}\cos^{2}\theta + \alpha_{\perp}\sin^{2}\theta\right)$$

$$U = -\frac{1}{2}\alpha_{\perp} E^{2} - \frac{1}{2}\left(\alpha_{\parallel} - \alpha_{\perp}\right) E^{2}\cos^{2}\theta$$

When $\alpha_{\parallel} > \alpha_{\perp}$,

U is minimized for θ=0

Nonlinear refractive index for molecular gas

- Refractive index built on dipole response

$$n^{2} = 1 + \chi = 1 + N\langle \alpha \rangle$$

- We have to average over all molecular orientations
- What is the distribution of angles?
  - Boltzmann distribution: $e^{-U/kT}$
  - Averaging over distribution:

$$\langle f(\theta) \rangle = \frac{\int f(\theta) e^{-U(\theta)/kT} \, d\Omega}{\int e^{-U(\theta)/kT} \, d\Omega}$$

Thermal averaged molecular response

- Get thermal average for induced dipole
- Angle-dependent energy:
  - Let $U(\theta) = -J\cos^2\theta\, kT$
  - With
    $$J = \frac{1}{2}(\alpha_3 - \alpha_1)\overline{E}^2 / kT$$
  - Use time averaged E^2, since molecule can't respond during one cycle
- Function to average:
  $$f(\theta) = \alpha(\theta) = (\alpha_3\cos^2\theta + \alpha_1\sin^2\theta) = \alpha_1 + (\alpha_3 - \alpha_1)\cos^2\theta$$
  $$\langle\alpha(\theta)\rangle = \alpha_1 + (\alpha_3 - \alpha_1)\frac{\int \cos^2\theta e^{-U(\theta)/kT}\cos\theta\, d\theta}{\int e^{-U(\theta)/kT}\, d\Omega}$$

n_2 calculation for molecular response

- For very low intensity (J~0), $\langle\cos^2\theta\rangle = 1/3$
- For nonlinear response (higher intensity) get 1st order in intensity
  $$\chi^{(3)} = \frac{2}{45}N\frac{(\alpha_3 - \alpha_1)^2}{kT}$$
- Note that there has to be time for response and thermalization
- shorter timescales: "impulsive Raman response"
  - delay in response
  - oscillations in refractive index

Thermal NL effects

- Due to linear absorption the laser can heat the medium
  - Laser pumping in a crystal
  - high average power propagation in low-absorption medium
- Absorption leads to a temperature gradient
- Refractive index depends on temperature

$$n(T) = n_0 + \tfrac{dn}{dT} T(r)$$

  - dn/dT is typically positive, sometimes <0
  - Results from lattice (or gas) expansion

Establishing a thermal distribution

- Heat equation

$$(\rho_0 C)\frac{\partial T}{\partial t} - \kappa \nabla^2 T = Q = \alpha I(r)$$

C = specific heat
ρ_0 = mass density
κ = thermal conductivity
Q = heat source distribution

- Estimate time response: scale equation

$$(\rho_0 C)\frac{T}{\tau} \sim \kappa \frac{T}{R^2} \qquad \rightarrow \tau \approx \frac{\rho_0 C}{\kappa} R^2$$

- Typically ~ 1 sec for macroscopic beams ~mm
- Note: dn/dT, C, and κ all depend on temperature
  - Cryo cooling reduces thermal effects dramatically
  - Smaller thermal gradient, faster cooling response

Thermal lensing

- T(r) leads to n(r), which leads to a lensing effect
- Assume steady state (CW or averaged over many shots) $-\kappa \nabla^2 T = \alpha I(r) = \alpha I_0 e^{-2r^2/w_0^2}$
  - Approximation:

$$\kappa \frac{\Delta T}{w_0^2} \approx \alpha I_0 \rightarrow \Delta T \approx \frac{\alpha I_0}{\kappa} w_0^2$$

$$\Delta n = \tfrac{dn}{dT} \Delta T \approx \left(\tfrac{dn}{dT} \frac{\alpha}{\kappa} w_0^2 \right) I_0$$

Effective n_2

- Other contributions:
  - Bowing out of crystal surfaces, stress birefringence

Plasma frequency

- The plasma frequency is the fundamental collective oscillation
- Consider a cube of plasma (number density n_e) with fixed ion background
  - Displace the electrons by dx
  - Calculate surface charge
  - Use Gauss' law to calculate restoring force
  - Then calculate oscillation frequency

Plasma frequency

- The plasma frequency is the fundamental collective oscillation
- Consider a cube of plasma (number density n_e) with fixed ion background
  - Displace the electrons by +dx

  - Use Gauss' law to calculate field and restoring force
    - Gaussian pill box on one side

$$\varepsilon_0 \int \mathbf{E} \cdot \mathbf{dS} = -\varepsilon_0 E A = \int \left(-e n_e\right) dV = -e n_e A dx$$

    - Equation of motion:

$$F = -eE = -\frac{n_e e^2}{\varepsilon_0} x = m_e \ddot{x} \qquad \rightarrow \ddot{x} = -\frac{e^2 n_e}{\varepsilon_0 m_e} x$$

  - Oscillation frequency

$$\omega_p^2 = \frac{n_e e^2}{\varepsilon_0 m_e}$$

Maxwell's equations in a plasma

- All charges are free. Maxwell's equations:

$$\varepsilon_0 \nabla \cdot \mathbf{E} = Z e n_i - e n_e \qquad \nabla \cdot \mathbf{B} = 0$$

$$\nabla \times \mathbf{E} = -\dot{\mathbf{B}} \qquad \nabla \times \mathbf{B} = \mu_0 \left(\mathbf{J} + \varepsilon_0 \dot{\mathbf{E}}\right)$$

- Current

$$\mathbf{J} = q n \mathbf{u} = Z e n_i \mathbf{u}_i - e n_e \mathbf{u}_e$$

$$\mathbf{J} = -e n_e \mathbf{u}$$ If only electrons are moving

- Charge continuity equation

$$\frac{\partial n_e}{\partial t} + \nabla \cdot \left(n_e \mathbf{u}\right) = 0$$

The force equation and convective derivative

- Equation of motion for a single particle:
  $$m_e \frac{d\mathbf{v}}{dt} = q\left(\mathbf{E} + \mathbf{v}\times\mathbf{B}\right)$$
- For a fluid, multiply by n_e:
  $$m_e n_e \frac{d\mathbf{u}}{dt} = q n_e\left(\mathbf{E} + \mathbf{u}\times\mathbf{B}\right)$$
- We want a fixed reference frame in space, so to account for the flow, we can write this total derivative as $\frac{d}{dt} = \frac{\partial}{\partial t} + \frac{\partial x}{\partial t}\frac{\partial}{\partial x}$
- Or in 3D: $\frac{d}{dt} = \frac{\partial}{\partial t} + \mathbf{u}\cdot\nabla$
  $$\rightarrow m_e n_e\left(\frac{\partial \mathbf{u}}{\partial t} + \left(\mathbf{u}\cdot\nabla\right)\mathbf{u}\right) = q n_e\left(\mathbf{E} + \mathbf{u}\times\mathbf{B}\right)$$

Alternate derivation of plasma frequency

- A plasma oscillation is purely electrostatic B=0

$$m_e n_e\left(\frac{\partial \mathbf{u}}{\partial t} + \left(\mathbf{u}\cdot\nabla\right)\mathbf{u}\right) = q n_e \mathbf{E} \qquad \frac{\partial n_e}{\partial t} + \nabla\cdot\left(n_e \mathbf{u}\right) = 0 \qquad \varepsilon_0 \nabla\cdot\mathbf{E} = Zen_i - en_e$$

- Assume Z=1, and 1D motion in x direction
  $$\nabla\times\mathbf{E} = -\dot{\mathbf{B}} = 0$$

$$m_e n_e\left(\frac{\partial v}{\partial t} + v\frac{\partial v}{\partial x}\right) = -en_e\mathbf{E} \qquad \frac{\partial n_e}{\partial t} + \frac{\partial}{\partial x}\left(n_e v\right) = 0 \qquad \varepsilon_0 \frac{\partial E}{\partial x} = e\left(n_i - n_e\right)$$

Linearization of the equations

- Linearize the equations (1st order perturbation)

$$n_e = n_0 + n_1 \qquad v = v_0 + v_1 \qquad E = E_0 + E_1 \qquad n_i = n_{i0} + n_{i1}$$

- Assume unperturbed plasma is uniform, at rest

$$\nabla n_0 = 0 \qquad v_0 = 0 \qquad E_0 = 0 \qquad \frac{\partial n_0}{\partial t} = \frac{\partial v_0}{\partial t} = \frac{\partial E_0}{\partial t} = 0 \qquad n_i = n_0, \quad n_{i1} = 0$$

$$\frac{\partial v_1}{\partial t} + \cancel{v_1 \frac{\partial v_1}{\partial x}} = -\frac{e}{m_e}\mathbf{E}_1 \qquad \frac{\partial n_1}{\partial t} + \frac{\partial}{\partial x}\left(n_0 v_1 + \cancel{n_1 v_1}\right) = 0 \qquad \varepsilon_0 \frac{\partial E_1}{\partial x} = -e n_1$$

- Input field varies sinusoidally, so does v_1, n_1
  - Note this is a longitudinal wave osc and k in x-direction

$$\mathbf{E_1} = E_1 e^{i(kx-\omega t)}\hat{\mathbf{x}} \qquad \mathbf{v_1} = v_1 e^{i(kx-\omega t)}\hat{\mathbf{x}} \qquad n_1 = n_1 e^{i(kx-\omega t)}$$

Solution for plasma oscillation frequency

- Evaluate derivatives

$$-i\omega v_1 = -\frac{e}{m_e}E_1 \qquad -i\omega n_1 + ikn_0 v_1 = 0 \qquad ik\varepsilon_0 E_1 = -en_1$$

- Eliminate E_1 and n_1

$$n_1 = -i\frac{k\varepsilon_0}{e}E_1 \qquad kn_0 v_1 = \omega n_1 = -i\omega\frac{k\varepsilon_0}{e}E_1 \qquad \rightarrow E_1 = i\frac{n_0 e}{\varepsilon_0 \omega}v_1$$

$$-i\omega v_1 = -\frac{e}{m_e}E_1 = -\frac{e}{m_e}\left(i\frac{n_0 e}{\varepsilon_0 \omega}v_1\right)$$

- End up with plasma frequency $\rightarrow \omega_p{}^2 = \dfrac{n_0 e^2}{\varepsilon_0 m_e}$
  - Note that

$$v_1 = -i\frac{e}{m_e \omega}E_1 \qquad n_1 = -i\frac{k\varepsilon_0}{e}E_1$$

v_1, n_1 90° out of phase with E

EM waves in a free electron, unmagnetized plasma

- Here we are looking for propagating transverse EM waves

$$\nabla n_0 = 0 \qquad v_0 = 0 \qquad \begin{matrix} E_0 = 0 \\ \mathbf{B_0} = 0 \end{matrix} \qquad \frac{\partial n_0}{\partial t} = \frac{\partial v_0}{\partial t} = \frac{\partial E_0}{\partial t} = 0 \qquad n_i = n_0, \quad n_{i1} = 0$$

$$\nabla \times \mathbf{B}_1 = \mu_0 \varepsilon_0 \left(\frac{\mathbf{J}_1}{\varepsilon_0} + \dot{\mathbf{E}}_1 \right) = \frac{1}{c^2} \left(\frac{\mathbf{J}_1}{\varepsilon_0} + \dot{\mathbf{E}}_1 \right)$$

- Take time derivative of 1st eqn

$$\nabla \times \dot{\mathbf{B}}_1 = \frac{1}{c^2} \left(\frac{\dot{\mathbf{J}}_1}{\varepsilon_0} + \ddot{\mathbf{E}}_1 \right)$$

$$\nabla \times \nabla \times \mathbf{E}_1 = \nabla \left(\nabla \cdot \mathbf{E}_1 \right) - \nabla^2 \mathbf{E}_1 = -\nabla \times \dot{\mathbf{B}}_1 = -\frac{1}{c^2} \left(\frac{\dot{\mathbf{J}}_1}{\varepsilon_0} + \ddot{\mathbf{E}}_1 \right)$$

EM waves in a free electron gas

- From previous slide: $\nabla \left(\nabla \cdot \mathbf{E}_1 \right) - \nabla^2 \mathbf{E}_1 = -\frac{1}{c^2} \left(\frac{\dot{\mathbf{J}}_1}{\varepsilon_0} + \ddot{\mathbf{E}}_1 \right)$

- Assume plane waves of form $\exp\left[i \left(\mathbf{k} \cdot \mathbf{r} - \omega t \right) \right]$

$$-\mathbf{k} \left(\mathbf{k} \cdot \mathbf{E}_1 \right) + k^2 \mathbf{E}_1 = -\frac{1}{c^2} \left(-i\omega \frac{\mathbf{J}_1}{\varepsilon_0} - \omega^2 \mathbf{E}_1 \right)$$

- We are looking for transverse waves $\mathbf{k} \cdot \mathbf{E} = 0$

$$\left(\omega^2 - k^2 c^2 \right) \mathbf{E}_1 = -i\omega \frac{\mathbf{J}_1}{\varepsilon_0}$$

  - Note that $\mathbf{J_1}$ can be a source for an EM wave $\mathbf{E_1}$

Dispersion of a free electron gas

- For high frequency waves, only electrons are moving

$$\mathbf{J_1} = -n_0 e \mathbf{v}_1$$

- This velocity is driven by the E-field

$$m_e \frac{d\mathbf{v_1}}{dt} = -e\mathbf{E}_1 \qquad \rightarrow i\omega \mathbf{v_1} = \frac{e}{m_e}\mathbf{E}_1 \qquad \mathbf{J_1} = -n_0 e \frac{e}{im_e\omega}\mathbf{E}_1 = -\frac{n_0 e^2}{im_e\omega}\mathbf{E}_1$$

- Put this into wave equation:

$$\left(\omega^2 - k^2c^2\right)\mathbf{E}_1 = -i\omega\frac{\mathbf{J}_1}{\varepsilon_0} = -i\frac{\omega}{\varepsilon_0}\left(-\frac{n_0 e^2}{im_e\omega}\mathbf{E}_1\right) = \frac{n_0 e^2}{\varepsilon_0 m_e}\mathbf{E}_1 = \omega_p^2\mathbf{E}_1$$

- Finally we have the dispersion relation

$$\omega^2 = \omega_p^2 + k^2c^2 \qquad k^2 \equiv \frac{\omega^2}{c^2}n^2 = \frac{\omega^2}{c^2} - \frac{\omega_p^2}{c^2} = \frac{\omega^2}{c^2}\left(1 - \frac{\omega_p^2}{\omega^2}\right)$$

Cutoff frequency

- Refractive index $n = \sqrt{1 - \frac{\omega_p^2}{\omega^2}}$ $\qquad n^2 = 1 - \frac{N_e}{N_{cr}}$

Critical density $N_{cr} = \frac{\varepsilon_0 m_e \omega^2}{e^2}$

- For $\omega < \omega_p$, $N_e > N_{cr}$ $\qquad n = i\frac{\omega_p}{\omega}\sqrt{1 - \frac{\omega^2}{\omega_p^2}}$

Take +'ve root for energy conservation

- And wave is exponentially damped (reflected)

$$e^{i(k_0 nz - \omega t)} = \exp\left[-\frac{\omega}{c}\frac{\omega_p}{\omega}\sqrt{1 - \frac{\omega^2}{\omega_p^2}}z - i\omega t\right] = \exp\left[-\frac{\omega_p}{c}\sqrt{1 - \frac{\omega^2}{\omega_p^2}}z - i\omega t\right]$$

- 1/coefficient of z is the skin depth
- For low frequency waves, electrons can move to shield out E-field from conductor

Dispersive propagation in plasma

- For high frequency waves (plasma is "underdense")

$$n = \sqrt{1 - \frac{\omega_p^2}{\omega^2}} < 1$$

- Phase velocity is > c: $V_{ph} = \frac{\omega}{k} = \frac{c}{n} = c\left(1 - \frac{\omega_p^2}{\omega^2}\right)^{-1/2} > c$
- But group velocity is < c: $V_{gr} = \left(\frac{dk}{d\omega}\right)^{-1} = c\left(1 - \frac{\omega_p^2}{\omega^2}\right)^{+1/2} < c$
- For low electron density, we often approximate:

$$n = \sqrt{1 - \frac{\omega_p^2}{\omega^2}} \approx 1 - \frac{\omega_p^2}{2\omega^2} = 1 - \frac{N_e}{2N_{cr}}$$

Ionization defocusing

- Refractive index for a free electron gas

$$n^2 = 1 - \frac{\omega_p^2}{\omega^2} \qquad \omega_p^2 = \frac{N_e e^2}{\varepsilon_0 m_e}$$ Plasma frequency

  - Plasma is transparent for $\omega > \omega_p$
  - Alternative representation:

$$n^2 = 1 - \frac{N_e}{N_{cr}} \qquad N_e = \frac{\varepsilon_0 m_e \omega^2}{e^2}$$ Critical density

- As laser ionizes medium, increase in N_e decreases n
  - Leads to a defocusing effect if beam is peaked in center

Ionization mechanisms

- Optical field ionization
  - Multiphoton ionization: $dN_e/dt \sim I^m$ where $m = U_{ion}/h\nu$
  - Tunneling ionization: strong E-field suppresses binding potential. Bound electron can tunnel through barrier
- Avalanche ionization
  - Seed electron is heated in field
  - Collisional ionization frees more electron
  - Exponential build up

The ponderomotive potential

- Consider a free electron responding to an EM wave

$$m_e \frac{d\mathbf{v_1}}{dt} = -e\mathbf{E}_1 \qquad \mathbf{E_1} = E_1(x,y)\cos(k_z z - \omega t)\hat{\mathbf{x}}$$

$$\mathbf{v_1} = -\frac{e}{m_e}\int \mathbf{E}_1\, dt = \frac{e}{m_e\omega}E_1(x,y)\sin(k_z z - \omega t)\hat{\mathbf{x}}$$

- Calculate the time-averaged KE:

$$\left\langle \frac{1}{2} m_e v_1^2 \right\rangle = \left\langle \frac{e^2}{2m_e\omega^2} E_1^{\ 2}(x,y)\sin^2(k_z z - \omega t) \right\rangle = \frac{e^2}{4m_e\omega^2}E_1^{\ 2}(x,y)$$

- This is the ponderomotive potential

$$U_p = \frac{e^2 E_1^{\ 2}}{4m_e\omega^2}$$

Field energy is put into coherent KE of the electrons: High intensity leads to a greater potential energy

Scaling of the ponderomotive potential

- In terms of intensity: $I = \frac{1}{2} n\varepsilon_0 c E_1^{\ 2}$

$$U_p = \frac{e^2 E_1^{\ 2}}{4m_e \omega^2} = \frac{e^2}{4m_e c^2 k^2 \frac{1}{2} n\varepsilon_0 c} I_1 = \frac{r_e \lambda^2}{2\pi n c} I_1$$

$$r_e = \frac{1}{4\pi\varepsilon_0} \frac{e^2}{m_e c^2}$$

classical electron radius
$= 2.82\text{x}10^{-15}$ m

- For numerical estimates:

$$U_p \approx 9\times 10^{-14} I_1 \lambda^2 \quad eV$$

Intensity in W/cm2
Wavelength in microns

The ponderomotive force

- If we treat U_p as a potential, then we expect a force when there is a gradient of U_p:

$$F_p = -\nabla U_p = -\frac{e^2}{4m_e \omega^2} \nabla\left(E_1^{\ 2}\right)$$

- For intensity gradient along the polarization direction, electron moves farther downhill than it returns.
- But force is actually independent of polarization direction.

Derivation of the ponderomotive force

- First order response

$$\mathbf{v}_1 = -\frac{e}{m_e\omega}\mathbf{E}_{10}\sin(\omega t) = \frac{d\mathbf{r}_1}{dt} \qquad \delta\mathbf{r}_1 = \frac{e}{m_e\omega^2}\mathbf{E}_{10}\cos(\omega t)$$

1st order velocity 1st order amplitude

- Expand equations to second order (at z=0)

$$\mathbf{E}(\mathbf{r}) \approx \mathbf{E}_1(r_0) + \delta\mathbf{r}_1 \cdot \nabla\mathbf{E}_1\big|_{r=r_0} + \cdots$$

- Need to add a term to the force equation: $\mathbf{v}_1 \times \mathbf{B}_1$

$$\nabla\times\mathbf{E} = -\dot{\mathbf{B}} \qquad \rightarrow \mathbf{B}_1 = -\frac{1}{\omega}\nabla\times\mathbf{E}_{10}\big|_{r=r_0}\sin\omega t$$

$$\mathbf{v}_1\times\mathbf{B}_1 = \left(-\frac{e}{m_e\omega}\mathbf{E}_{10}\sin(\omega t)\right)\times\left(-\frac{1}{\omega}\nabla\times\mathbf{E}_{10}\sin\omega t\right) = \frac{e}{m_e\omega^2}\mathbf{E}_{10}\times\nabla\times\mathbf{E}_{10}\sin^2\omega t$$

Derivation of the ponderomotive force

- Evaluate 2nd order of force equation

$$m_e\frac{d\mathbf{v}}{dt} = -e(\mathbf{E} + \mathbf{v}\times\mathbf{B}) \qquad \mathbf{v}_1\times\mathbf{B}_1 = \frac{e}{m_e\omega^2}\mathbf{E}_{10}\times\nabla\times\mathbf{E}_{10}\sin^2\omega t$$

$$\mathbf{E}_2(\mathbf{r}) = \delta\mathbf{r}_1\cdot\nabla\mathbf{E}_{10}\cos\omega t \qquad \delta\mathbf{r}_1 = \frac{e}{m_e\omega^2}\mathbf{E}_{10}\cos(\omega t)$$

$$= \frac{e}{m_e\omega^2}\mathbf{E}_{10}\cos(\omega t)\cdot\nabla\mathbf{E}_{10}\cos\omega t$$

$$m_e\frac{d}{dt}\mathbf{v}_2 = -e\left(\frac{e}{m_e\omega^2}\mathbf{E}_{10}\cdot\nabla\mathbf{E}_{10}\cos^2\omega t + \frac{e}{m_e\omega^2}\mathbf{E}_{10}\times\nabla\times\mathbf{E}_{10}\sin^2\omega t\right)$$

- Time average

$$m_e\left\langle\frac{d}{dt}\mathbf{v}_2\right\rangle = -\frac{e^2}{2m_e\omega^2}\left(\mathbf{E}_{10}\cdot\nabla\mathbf{E}_{10} + \mathbf{E}_{10}\times\nabla\times\mathbf{E}_{10}\right) = -\frac{e^2}{4m_e\omega^2}\nabla E_{10}{}^2$$

SHG as a diagnostic of gradients in plasma density and E-fields

A second-harmonic signal can arise in an initally isotropic medium when the symmetry is broken

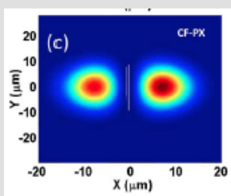
- Third-order response with quasi-DC field (from charge separation)

$$\mathbf{P}(2\omega) = \chi_a^{(3)}(2\omega)\mathbf{E}(\omega)\left[\mathbf{E}_{DC}\cdot\mathbf{E}(\omega)\right] + \chi_b^{(3)}(2\omega)\mathbf{E}_{DC}\left|\mathbf{E}(\omega)\right|^2$$

- Gradient in the electron density

$$\mathbf{P}(2\omega) = -i\frac{e^3}{2m^2\omega^3}(\nabla\delta n\cdot\mathbf{E})\mathbf{E}$$

Example: conventionally focused pulses, the density gradient follows the Gaussian focus: leads to SH signal with lobes:



Gradient points radially outwards
E-field is linearly polarized

Bethune PRA 23 3139 (1981)
Li et al, OptLett 39, 961 (2014)

31

Second harmonic generation in non-uniform plasmas

- Starting equations

$$m_e\left(\frac{\partial\mathbf{v}}{\partial t} + \left(\mathbf{v}\cdot\nabla\right)\mathbf{v}\right) = q\left(\mathbf{E} + \mathbf{v}\times\mathbf{B}\right) \qquad \frac{\partial n_e}{\partial t} + \nabla\cdot\left(n_e\mathbf{v}\right) = 0$$

$$\nabla\times\mathbf{E} = -\dot{\mathbf{B}} \qquad \varepsilon_0\nabla\cdot\mathbf{E} = e\left(n_i - n_e\right) \quad \nabla\times\mathbf{B} = \mu_0\left(\mathbf{J} + \varepsilon_0\dot{\mathbf{E}}\right) \quad \nabla\cdot\mathbf{B} = 0$$

- Expand variables in a perturbation series

$$n_e = n_0 + n_1 + n_2 + \cdots \qquad \mathbf{v} = \mathbf{v}_1 + \mathbf{v}_2 + \cdots \qquad \mathbf{J} = \mathbf{J}_1 + \mathbf{J}_2 + \cdots$$

- We're looking for the current that drives SH

$$\mathbf{J}_1 = -en_0\mathbf{v}_1 \qquad \mathbf{J}_2 = -en_1\mathbf{v}_1 - en_0\mathbf{v}_2$$

  – So we need to expand velocity to 2nd order

First order solutions

- Similar to derivation of EM wave in plasma
- Assume harmonic waves (no assumption on polarization)

  $$\mathbf{E}=\mathbf{E_0}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}+c.c. \qquad \mathbf{v_1}=\mathbf{v}_{10}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}+c.c. \qquad n_1=n_{10}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}+c.c.$$

  - For simplicity, we will assume no gradients in E, B (plane wave) $\nabla\times\mathbf{E}=-\dot{\mathbf{B}}=i\omega\mathbf{B}$
- Solve for 1st order velocity

  $$m_e\left(\frac{\partial\mathbf{v}_1}{\partial t}+\cancel{(\mathbf{v}\cdot\nabla)\mathbf{v}}\right)=q\left(\mathbf{E}+\cancel{\mathbf{v}\times\mathbf{B}}\right)$$

  $$\frac{\partial}{\partial t}\mathbf{v}_1=-i\omega\mathbf{v}_{10}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}=-\frac{e}{m_e}\mathbf{E_0}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} \qquad \rightarrow\mathbf{v_{10}}=-i\frac{e}{m_e\omega}\mathbf{E_0}$$

First order solutions

- Solve for 1st order density variation

  $$\frac{\partial n_1}{\partial t}+\nabla\cdot(n_0\mathbf{v_1})=0 \qquad \mathbf{v_{10}}=-i\frac{e}{m_e\omega}\mathbf{E_0}$$

  $$-i\omega n_{10}+\nabla\cdot(n_0\mathbf{v_{10}})=-i\omega n_{10}+\nabla\cdot\left(n_0\left(-i\frac{e}{m_e\omega}\mathbf{E_0}\right)\right)=0$$

  $$n_{10}=-\frac{e}{m_e\omega^2}\nabla\cdot(n_0\mathbf{E_0})$$

- Gauss' law $\varepsilon_0\nabla\cdot\mathbf{E}=e(n_i-n_e)=-en_{10}$
- Note that we have two expressions for n_{10}

Connect charge perturbation to density gradient

- From previous slide:

$$\nabla\cdot\mathbf{E}=-\frac{e}{\varepsilon_0}n_{10} \qquad n_{10}=-\frac{e}{m_e\omega^2}\nabla\cdot\left(n_0\mathbf{E_0}\right)$$

- Expand div term:

$$\nabla\cdot\mathbf{E}=\frac{e^2}{\varepsilon_0 m_e\omega^2}\nabla\cdot\left(n_0\mathbf{E_0}\right)=\frac{e^2}{\varepsilon_0 m_e\omega^2}\left(n_0\nabla\cdot\mathbf{E_0}+\mathbf{E_0}\cdot\nabla n_0\right)$$

- Gather div E terms

$$\nabla\cdot\mathbf{E_0}-\frac{e^2 n_0}{\varepsilon_0 m_e\omega^2}\nabla\cdot\mathbf{E_0}=\frac{e^2}{\varepsilon_0 m_e\omega^2}\mathbf{E_0}\cdot\nabla n_0 \qquad \frac{e^2 n_0}{\varepsilon_0 m_e\omega^2}=\frac{\omega_p^2}{\omega^2}$$

$$\nabla\cdot\mathbf{E_0}=\frac{e^2}{\varepsilon_0 m_e\omega^2}\frac{\mathbf{E_0}\cdot\nabla n_0}{1-\frac{\omega_p{}^2}{\omega^2}}=-\frac{e}{\varepsilon_0}n_{10} \qquad n_{10}=-\frac{e}{m_e\omega^2}\frac{\mathbf{E_0}\cdot\nabla n_0}{\varepsilon_p}$$

Calculate 2nd order velocity

- From Lorentz equation

$$m_e\left(\frac{\partial\mathbf{v}_2}{\partial t}+\left(\mathbf{v}_1\cdot\nabla\right)\mathbf{v}_1\right)=-e\left(\cancel{\mathbf{E}}+\mathbf{v}_1\times\mathbf{B}\right) \qquad \mathbf{v}_{10}=-i\frac{e}{m_e\omega}\mathbf{E}_{10} \qquad \mathbf{B}_{10}=-\frac{i}{\omega}\nabla\times\mathbf{E}_{10}$$

  - For ponderomotive force, we had 2nd order E from grad E
  - Here we *choose* component oscillating at 2ω

$$\frac{\partial\mathbf{v}_2}{\partial t}=-i2\omega\mathbf{v}_{20}=-\left(\mathbf{v}_{10}\cdot\nabla\right)\mathbf{v}_{10}-\frac{e}{m_e}\mathbf{v}_{10}\times\mathbf{B}_{10}$$

$$\mathbf{v}_{20}=-\frac{i}{2\omega}\left(\left(\left\{-i\frac{e}{m_e\omega}\mathbf{E}_{10}\right\}\cdot\nabla\right)\left\{-i\frac{e}{m_e\omega}\mathbf{E}_{10}\right\}+\frac{e}{m_e}\left\{-i\frac{e}{m_e\omega}\mathbf{E}_{10}\right\}\times\left\{-\frac{i}{\omega}\nabla\times\mathbf{E}_{10}\right\}\right)$$

$$\mathbf{v}_{20}=\frac{i}{4}\frac{e^2}{m_e{}^2\omega^3}\nabla\left(\mathbf{E}_{10}\cdot\mathbf{E}_{10}\right)$$

Calculate 2nd order current

- This current can be a source term for SH radiation

$$\mathbf{J}_2 = -en_0\mathbf{v}_2 - en_1\mathbf{v}_1 \qquad \mathbf{v}_{10} = -i\frac{e}{m_e\omega}\mathbf{E}_{10}$$

$$\mathbf{v}_{20} = \frac{i}{4}\frac{e^2}{m_e^2\omega^3}\nabla\left(\mathbf{E}_{10}\cdot\mathbf{E}_{10}\right) \qquad n_{10} = -\frac{e}{m_e\omega^2}\frac{\mathbf{E}_{10}\cdot\nabla n_0}{\varepsilon_p}$$

$$-en_0\mathbf{v}_2 = -en_0\frac{i}{4}\frac{e^2}{m_e^2\omega^3}\nabla\left(\mathbf{E}_0\cdot\mathbf{E}_0\right)$$

$$-en_1\mathbf{v}_1 = -e\left\{-\frac{e}{m_e\omega^2}\frac{\mathbf{E}_{10}\cdot\nabla n_0}{\varepsilon_p}\right\}\left\{-i\frac{e}{m_e\omega}\mathbf{E}_{10}\right\} = -i\frac{e^3}{m_e^2\omega^3}\frac{\mathbf{E}_{10}\left(\mathbf{E}_{10}\cdot\nabla n_0\right)}{\varepsilon_p}$$

$$\mathbf{J}_{20} = -i\frac{e^3}{m_e^2\omega^3}\left(\frac{n_0}{4}\nabla\left(\mathbf{E}_{10}\cdot\mathbf{E}_{10}\right) + \frac{\mathbf{E}_{10}\left(\mathbf{E}_{10}\cdot\nabla n_0\right)}{\varepsilon_p}\right)$$

We assumed plane waves, so first term = 0

Properties of the SH signal

- The electric field of the SH is calculated from current
  - From wave equation,

$$\left(\omega_2^2 - k_2^2c^2 + 2ik_2\partial_z\right)\mathbf{E}_{20} = -i\omega_2\frac{\mathbf{J}_{20}}{\varepsilon_0} \qquad \omega_2 = 2\omega$$

$$\partial_z\mathbf{E}_{20} = -\frac{\omega_2}{2k_2}\frac{\mathbf{J}_{20}}{\varepsilon_0} \qquad \mathbf{J}_{20} = -i\frac{e^3}{m_e^2\omega^3}\left(\frac{n_0}{4}\nabla\left(\mathbf{E}_{10}\cdot\mathbf{E}_{10}\right) + \frac{\mathbf{E}_{10}\left(\mathbf{E}_{10}\cdot\nabla n_0\right)}{\varepsilon_p}\right)$$

- SH is the same polarization as the input
- Amplitude is modulated by E.grad n_e
  - For circular polarization in, we get vortex phase out!

$$\mathbf{E}_{10}\left(\mathbf{E}_{10}\cdot\nabla n_0\right) = E_{10}^2\left|\nabla n_0\right|\left(\hat{\mathbf{x}} \pm i\hat{\mathbf{y}}\right)\cdot\hat{\mathbf{r}} = E_{10}^2\left|\nabla n_0\right|\left(\cos\theta \pm i\sin\theta\right)E_{10}^2\left|\nabla n_0\right|e^{\pm i\theta}$$