An introduction to femtosecond laser techniques: Part 1



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Introduction



Outline

- propagation of pulses
- nonlinear optics
- nanoscale optics
- nonlinear optics at the nanoscale

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Governed by wave equation

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In dispersive media $n = n(\omega)$.

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In vacuum:
$$f\lambda = \frac{\omega}{k} = c \implies \omega = c k$$



In medium: $v = \frac{c}{\sqrt{\epsilon(\omega)}} = \frac{c}{n} \implies \omega = \frac{c}{\sqrt{\epsilon}} k$



Which charges participate?















Electron on a string: $F_{binding} = -m_e \omega_o^2 x$

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Polarization

$$P(t) = \left(\frac{Ne^2}{m}\right) \sum_{j} \frac{f_j}{(\omega_j^2 - \omega^2) - i\gamma_j \omega} E(t) \equiv \epsilon_o \chi_e E(t)$$

$$\epsilon(\omega) \equiv 1 + \chi_e = 1 + \frac{Ne^2}{\epsilon_o m} \sum_{j} \frac{f_j}{(\omega_j^2 - \omega^2) - i\gamma_j \omega}$$
Dielectric function

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Q: For a single resonance, is the value of $\epsilon(\omega)$ at high frequency

- 1. larger than,
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- 3. smaller than the value at low frequency?

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Amplitude of bound charge oscillation



Below resonance: bound charges keep up with driving field \Rightarrow field attenuated, wave propagates more slowly



At resonance: energy transfer from wave to bound charges ⇒ wave attenuates (absorption)



Above resonance: bound charges cannot keep up with driving field \Rightarrow dielectric like a vacuum



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Low frequency ($\omega \ll \gamma$) \Rightarrow current generated

$$J = -Ne \frac{dx}{dt} = \frac{Ne^2}{m} \frac{1}{\gamma - i\omega} E \approx \frac{Ne^2}{m\gamma} E \equiv \sigma E$$

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Dielectric function:

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Add damping:

$$\gamma \lesssim \omega_p$$





Plasma acts like a high-pass filter







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log <i>N</i> (cm⁻³)	ω_p (rad s ⁻¹)	$oldsymbol{\lambda}_p$
22	6 x 10 ¹⁵	330 nm
18	6 x 10 ¹³	33 µm
14	6 x 10 ¹¹	3.3 mm
10	6 x 10 ⁹	0.33 m



 \bigvee \bigvee $\omega < \omega_p$











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$$y_1 = A \sin(k_1 x - \omega_1 t)$$
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Superposition:

$$y = A[\sin (k_1 x - \omega_1 t) + \sin (k_2 x - \omega_2 t)]$$

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$$y = A[\sin (k_1 x - \omega_1 t) + \sin (k_2 x - \omega_2 t)]$$

$$\sin \alpha + \sin \beta = 2 \cos \left(\frac{\alpha - \beta}{2} \right) \sin \left(\frac{\alpha + \beta}{2} \right)$$

$$y = 2A \cos \frac{1}{2} [(k_1 - k_2)x - (\omega_1 - \omega_2)t] \sin \left[\frac{k_1 + k_2}{2}x - \frac{\omega_1 + \omega_2}{2}t\right]$$

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Let: $k_1 - k_2 \equiv \Delta k$ and $\omega_1 - \omega_2 \equiv \Delta \omega$

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 and $\frac{\omega_1+\omega_2}{2} \equiv \omega$

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traveling sine wave, with amplitude modulation.
$$y = 2A \cos \frac{1}{2}(x\Delta k - t\Delta \omega) \sin (kx - \omega t)$$

At
$$t = 0$$
: $y = 2A \cos \frac{1}{2}(x\Delta k) \sin (kx)$



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$$t = 0$$
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 $y = 2A \cos \frac{1}{2}(x\Delta k) \sin (kx)$
carrier



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At
$$t = 0$$
:
 $y = 2A \cos \frac{1}{2}(x\Delta k) \sin (kx)$
envelope carrier



$$y = 2A \cos \frac{1}{2}(x\Delta k - t\Delta \omega) \sin (kx - \omega t)$$

$$y = 2A \cos \frac{1}{2} (x \Delta k - t \Delta \omega) \sin (kx - \omega t)$$

speed of carrier

$$v_p = \frac{\omega}{k} = f\lambda$$

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$$v_p = \frac{\omega}{k} = f\lambda$$

speed of envelope

$$v_g = \frac{\Delta \omega}{\Delta k} = \frac{d\omega}{dk}$$

$$y = 2A \cos \frac{1}{2} (x \Delta k - t \Delta \omega) \sin (kx - \omega t)$$

speed of carrier ('phase velocity'):

$$v_p = \frac{\omega}{k} = f\lambda$$

speed of envelope ('group velocity'):

$$v_g = rac{\Delta \omega}{\Delta k} = rac{d\omega}{dk}$$

let's practice a bit!

(please complete worksheet)

For each wave, determine the wavevector k, the frequency ω , and the propagation speed v:

$$k_{1} = \frac{8.0}{k_{1}} \qquad \text{and} \qquad k_{2} = \frac{72}{0.45} = 7.6 < k_{1}$$
(B)
$$\omega_{1} = \frac{8.0}{k_{1}} \qquad \text{and} \qquad \omega_{2} = 7.2$$

$$v_{1} = \frac{\omega_{1}}{k_{1}} = 1. \qquad \text{and} \qquad v_{2} = \frac{\omega_{2}}{k_{2}} = \frac{7.2}{2.77} = 0.95$$

Does the red get ahead of the blue or the other way around? Why?

Is the dispersion in the medium through which these waves propagate normal or anomalous? Why?

What is the phase velocity of the superposition of y_1 and y_2 ?

$$V_{p} = \frac{(w_{1} + w_{2})/2}{(k_{1} + k_{2})/2} = \frac{7.6}{7.8} = 0.98$$

What is the group velocity of the superposition of y_1 and y_2 ?

$$V_g = \frac{W_1 - W_2}{k_1 - k_2} = \frac{0.8}{0.4} = 2$$

Do the crests of the carrier wave travel forward or backward through the envelope? Does your prediction agree with your observation?

$$y = 2A \cos \frac{1}{2}(x\Delta k - t\Delta \omega) \sin (kx - \omega t)$$

If no dispersion

$$v_p = \frac{\omega_1}{k_1} = \frac{\omega_2}{k_2}$$

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group velocity:

$$v_g = \frac{\Delta \omega}{\Delta k} = \frac{\omega_1 - \omega_2}{k_1 - k_2} = \frac{\omega_1 / k_1 - \omega_2 / k_1}{1 - k_2 / k_1} = \frac{v_p - \omega_2 / k_1}{1 - k_2 / k_1}$$

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$$v_g = \frac{v_p - \omega_2/k_2(k_2/k_1)}{1 - k_2/k_1} = v_p \frac{1 - k_2/k_1}{1 - k_2/k_1} = v_p$$

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If no dispersion

$$v_p = \frac{\omega_1}{k_1} = \frac{\omega_2}{k_2}$$

group and phase velocities are the same:

$$v_g = v_p$$

and so the envelope and carrier travel together

$$y = 2A \cos \frac{1}{2} (x \Delta k - t \Delta \omega) \sin (kx - \omega t)$$

Types of dispersion:

$$\frac{dn}{d\omega} > 0$$
 normal dispersion

 $\frac{dn}{d\omega} = 0$ no dispersion

 $\frac{dn}{d\omega} < 0$ anomalous dispersion



$$y = 2A \cos \frac{1}{2} (x \Delta k - t \Delta \omega) \sin (kx - \omega t)$$

Types of dispersion:





medium causes pulse to stretch





medium causes pulse to stretch



compensate by rearranging spectral components!









How do these arrangements work?

How do these arrangements work? (please complete worksheet)







Does path length difference compensate?



grating gives low frequency longer path length!









Does path length difference compensate?



...so prism gives low frequency shorter path length!

consider traveling Gaussian pulse again:

$$y(t) = \exp\left[-\frac{(x-v_g t)^2}{2\sigma_t^2}\right] \sin(kx - \omega t)$$
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Q: Can you tell if the medium is dispersive or not?

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- 1. Yes, it is dispersive
- 2. No, it is not dispersive (pulse shape is constant)
- 3. Cannot tell

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A: Cannot tell (the medium is dispersive if $v_g \neq \frac{\omega}{k}$)

...but Gaussian shape of pulse is constant!













only nonlinear dispersion changes pulse shape!



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	$rac{dl_{eff}}{d\omega}$	$\frac{d^2\phi}{d\omega^2}$
dispersion	+	+
gratings	-	-
prisms	+	_

So not path length but
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 matters!



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and so:

$$P = P^{(1)} + P^{(2)} + P^{(3)} + \dots$$
$$P^{(2)} \approx P^{(1)} \text{ when } E = E_{at} \approx \frac{e}{a} \text{, and so } \chi^{(n)} \approx \frac{\chi^{(1)}}{E_{at}^{n-1}}$$

Nonlinear polarization can drive new field:

$$\nabla^2 \vec{E} + \frac{n^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 \vec{P}}{\partial t^2}$$

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But even terms disappear in media with inversion symmetry!

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Invert all vectors:

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and so $\chi^{(2)} = -\chi^{(2)} = 0$.

Consider oscillating electric field:

 $E(t) = E e^{i\omega t} + \text{c.c.}$

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Second-order polarization:

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$$n = \sqrt{\epsilon} = \sqrt{1 + \chi_{eff}} \approx \sqrt{1 + \chi^{(1)}} + \frac{1}{2} \frac{\chi^{(3)}I}{\sqrt{1 + \chi^{(1)}}} = n_o + n_2 I$$

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- **Q:** Sketch the time dependence of the frequency shift for a Gaussian pulse and determine which is true (assume $n_2 > 0$):
 - 1. Leading edge is blue shifted, trailing edge red shifted
 - 2. Leading and trailing edge blue shifted, center red shifted
 - 3. Leading edge is red shifted, trailing edge blue shifted
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Phase:

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$$b = \frac{2\pi}{\lambda} L(n_o + n_2 I)$$

Frequency change:

$$\Delta \omega = -\frac{d\phi}{dt} = \frac{-2\pi}{\lambda} Ln_2 \frac{dI}{dt}$$




Intensity-dependent index of refraction:

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self-focusing



but susceptibility is complex!

susceptibility	real part	imaginary part
linear	refraction	absorption
nonlinear	SHG, SFG, DFG, THG,	multiphoton absorption

$$\alpha = \alpha_o + \beta I + \gamma I^2 + \dots$$



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