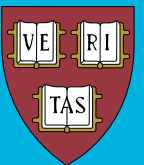
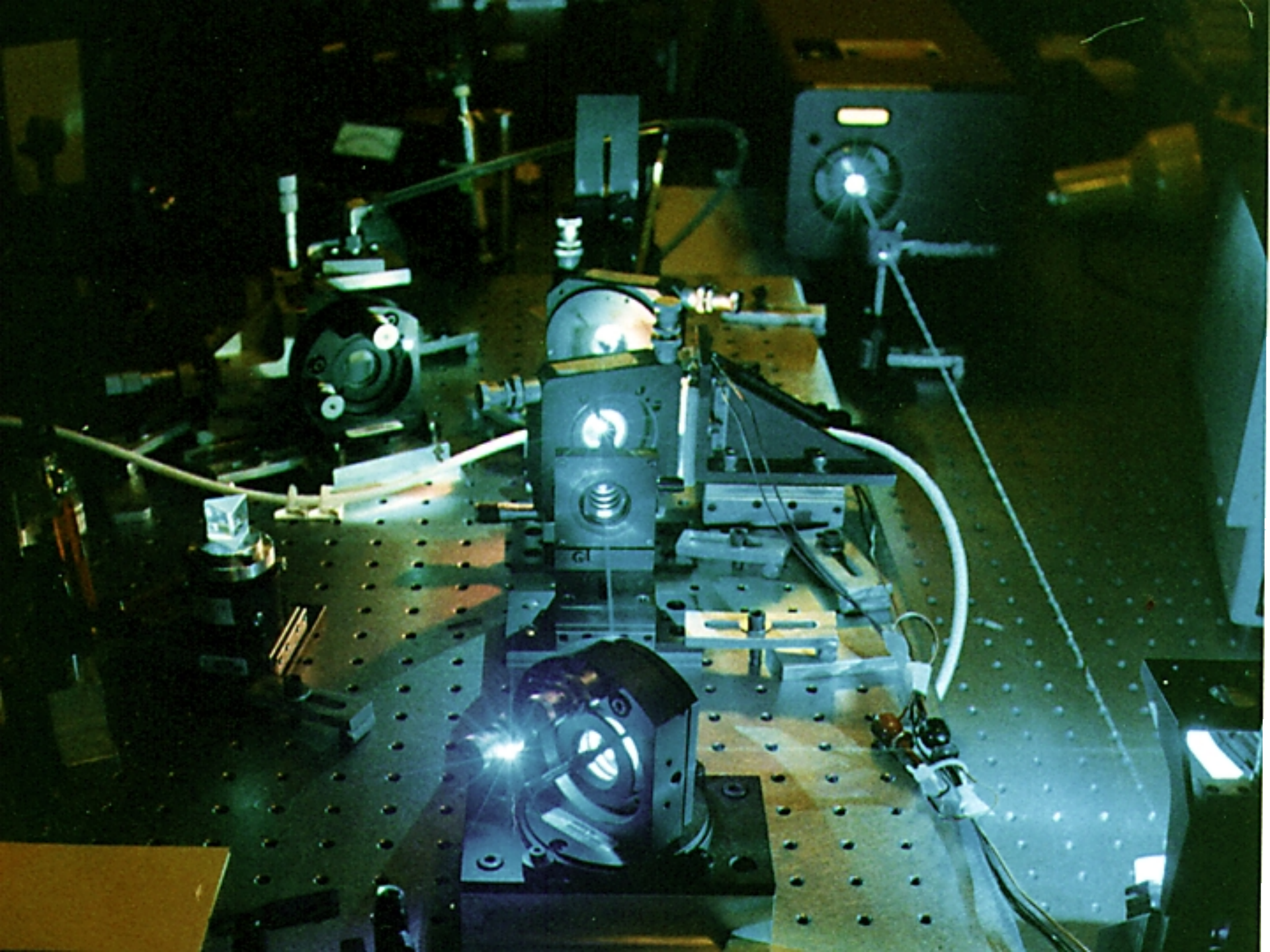


AN INTRODUCTION TO FEMTOSECOND LASER SCIENCE

**Eric Mazur
Harvard University**

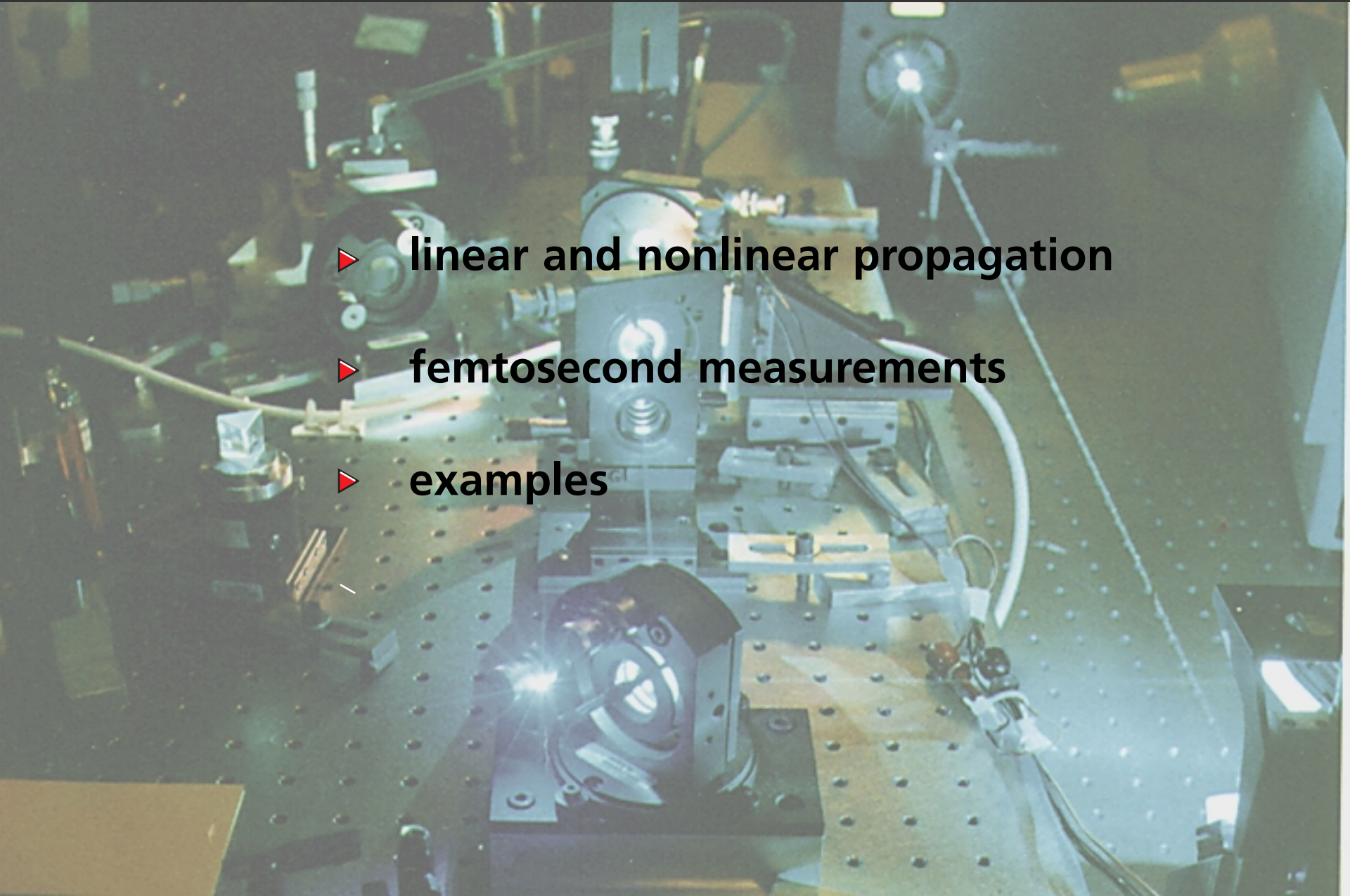
**Short Course SC101
OSA Annual Meeting 2003, Tucson AZ**





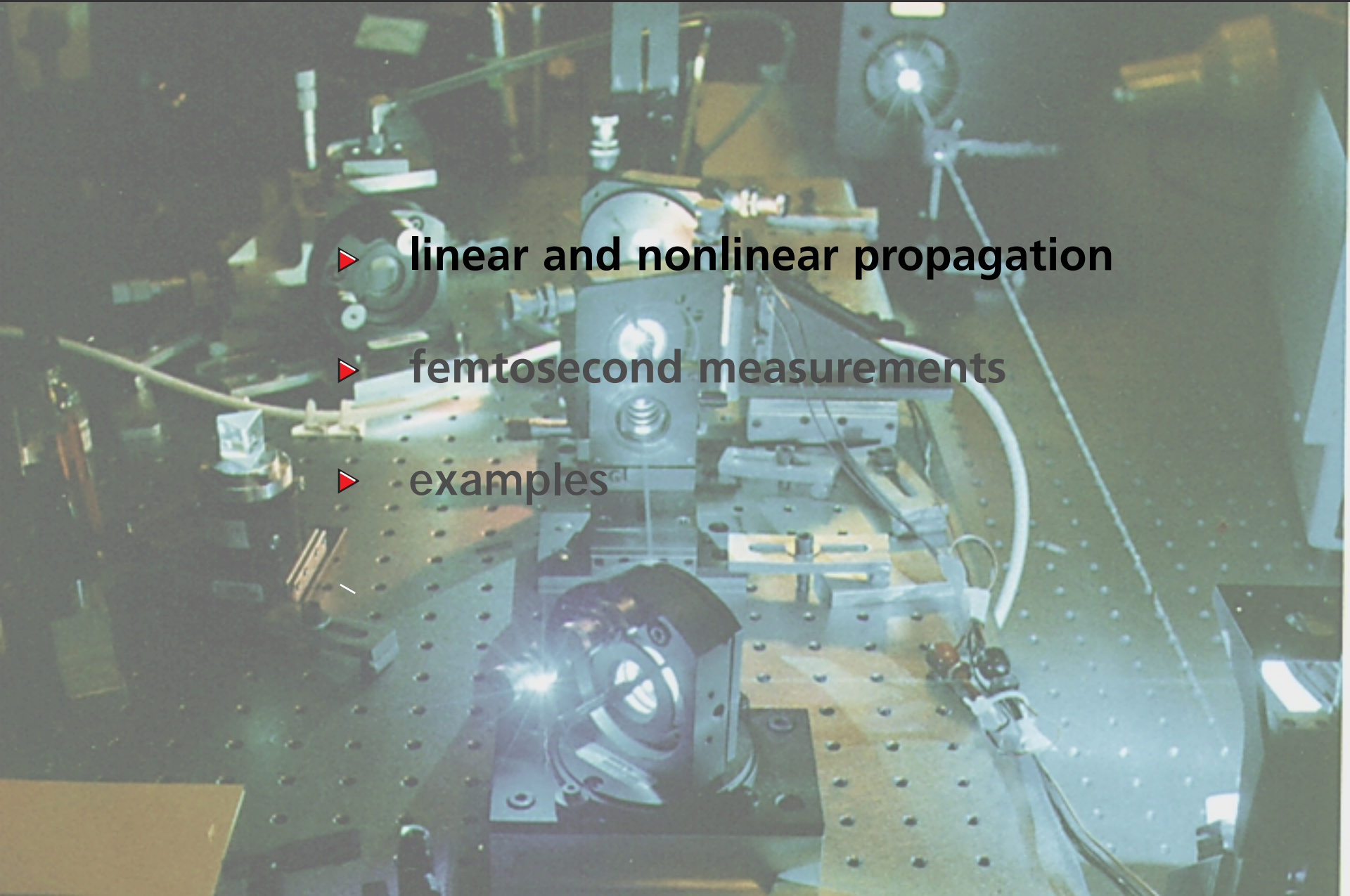
Outline

- ▶ **linear and nonlinear propagation**
- ▶ **femtosecond measurements**
- ▶ **examples**

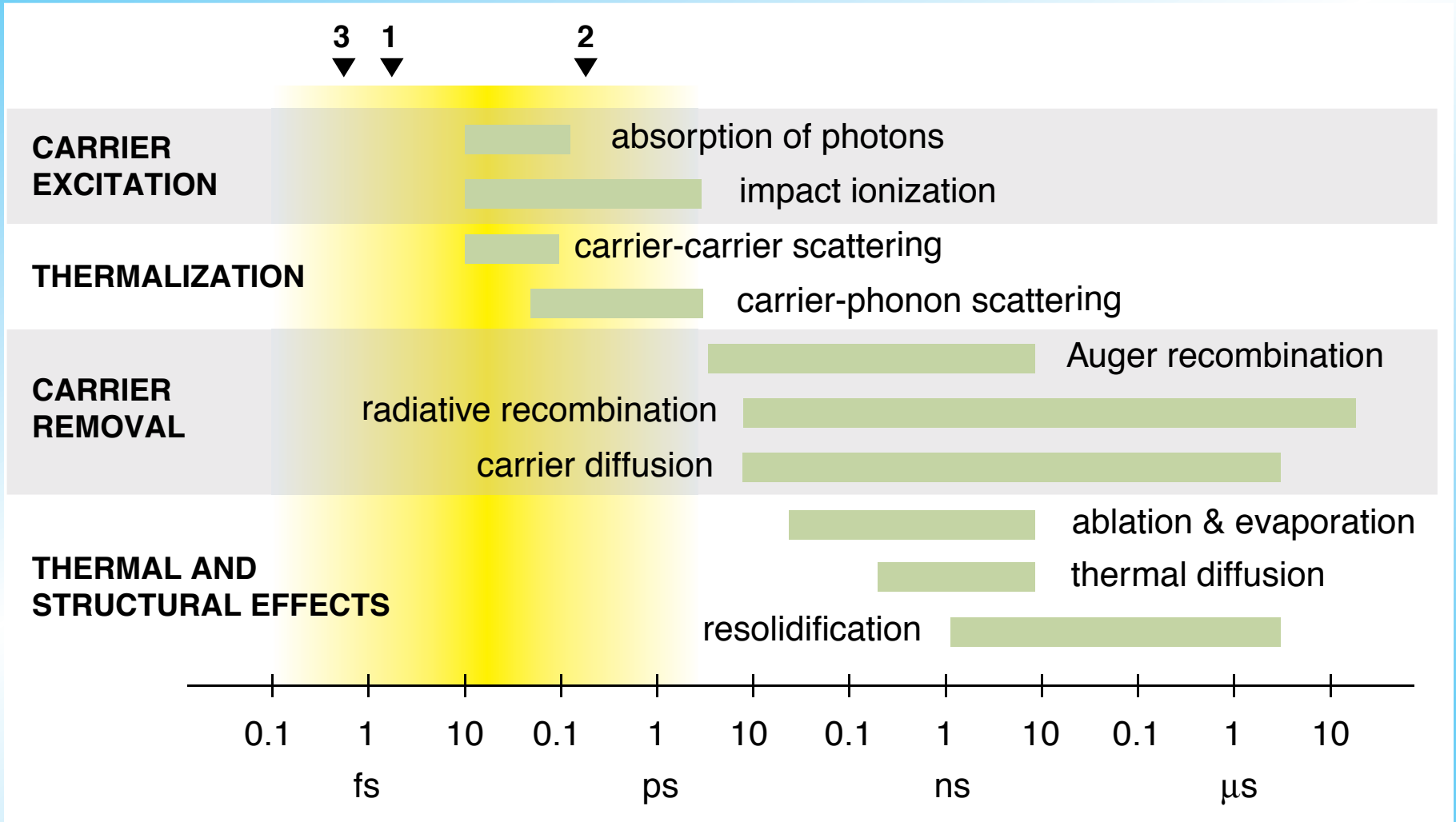


Outline

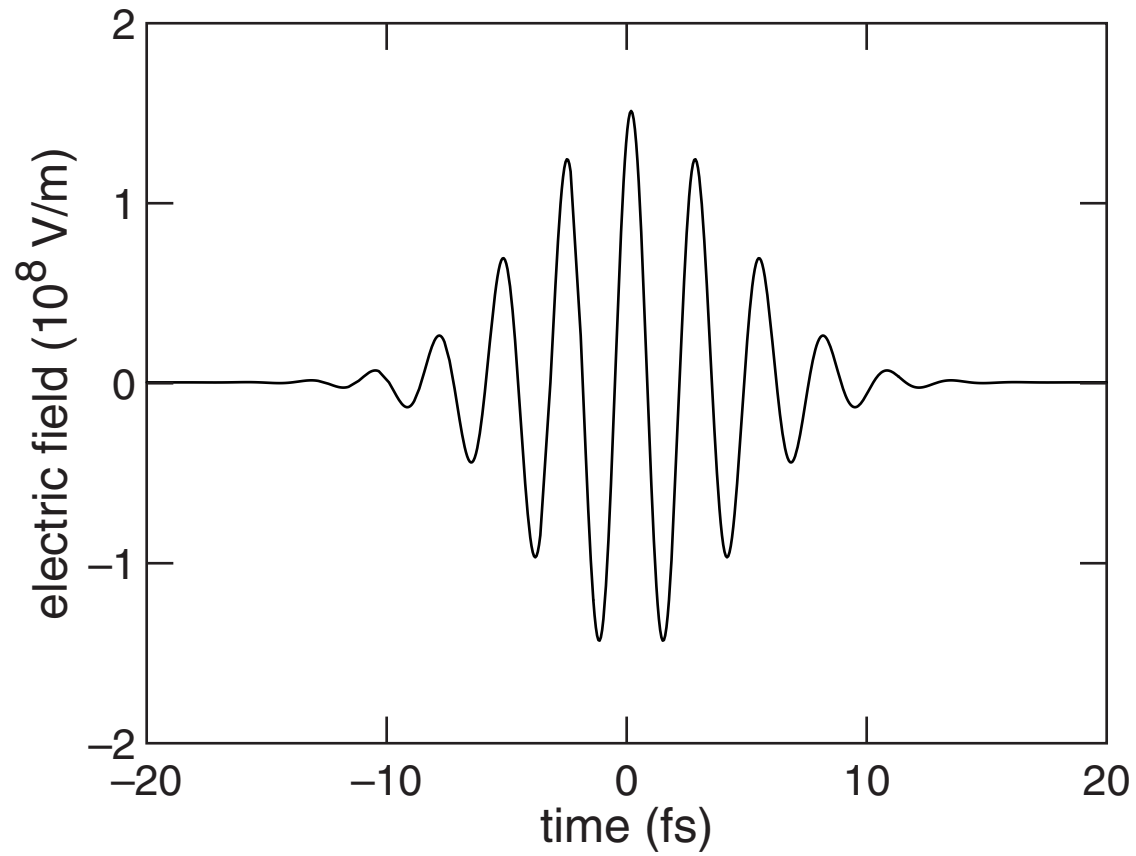
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- ▶ **femtosecond measurements**
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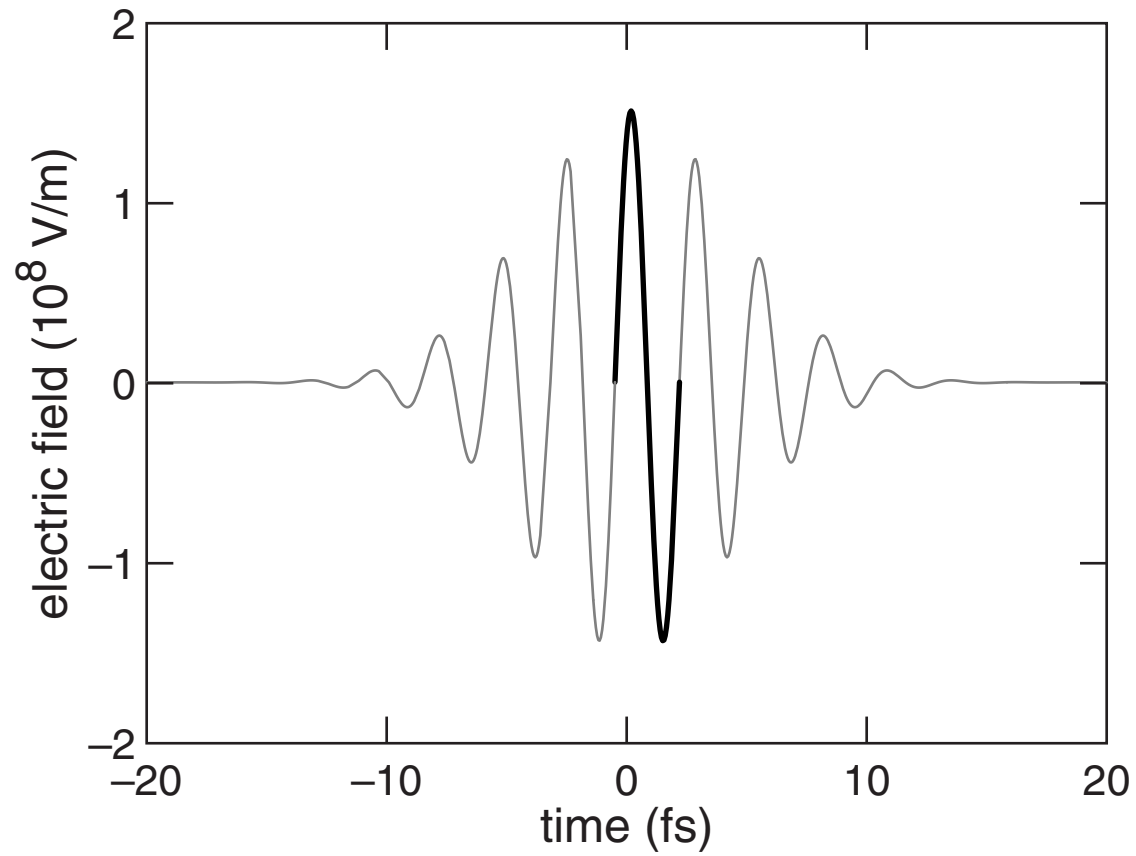
Introduction



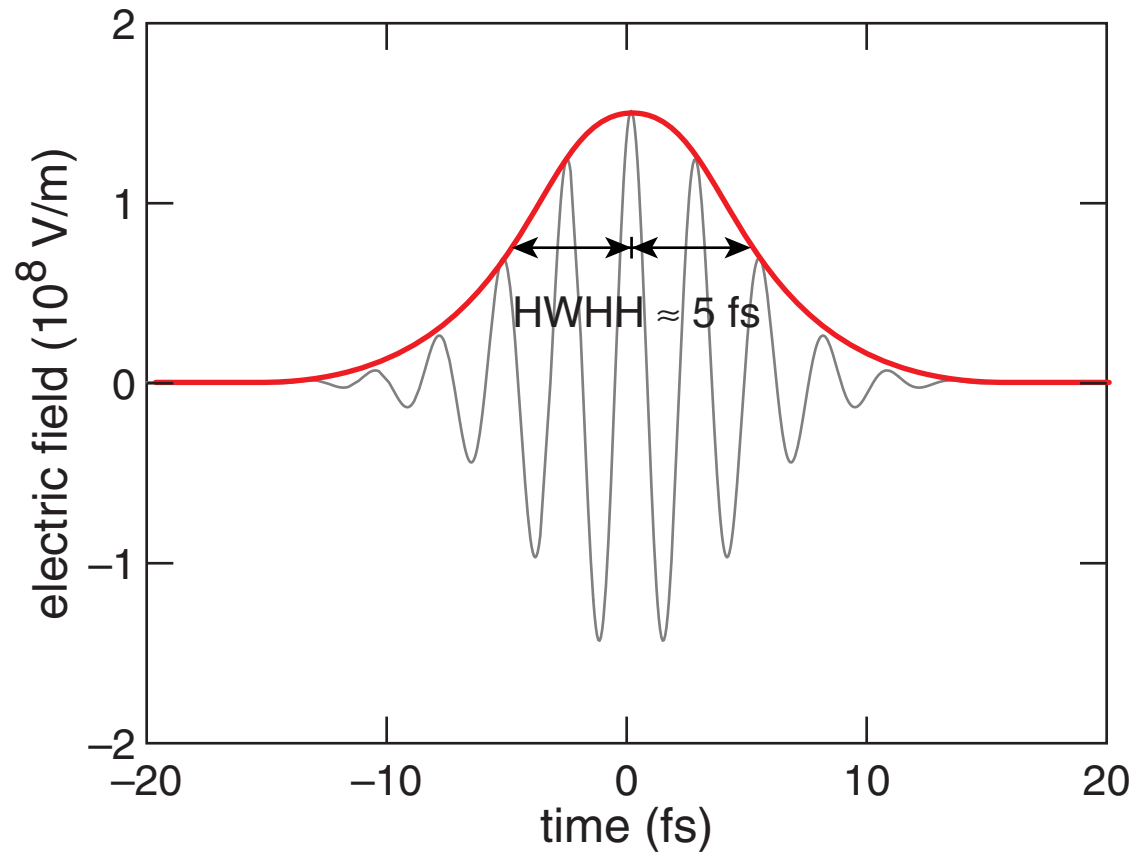
Introduction



Introduction



Introduction



Introduction

- ▶ **time resolution**
- ▶ **high intensity**
- ▶ **nonlinear optics**
- ▶ **new physics**

Propagation of EM waves through medium

Governed by wave equation

$$\nabla^2 \vec{E} - \frac{\mu\epsilon}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = 0$$

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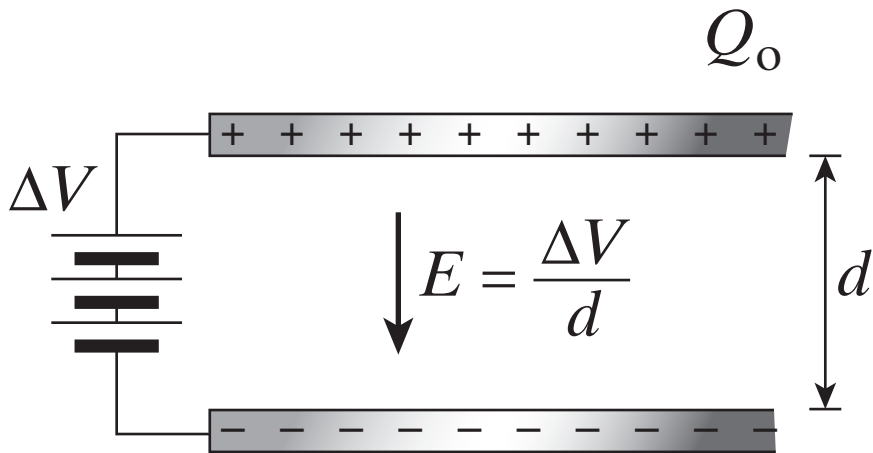
In non-ferromagnetic media $\mu \approx 1$, and so $n \approx \sqrt{\epsilon}$.

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

Propagation of EM waves through medium

Dielectric constant measures increase in capacitance

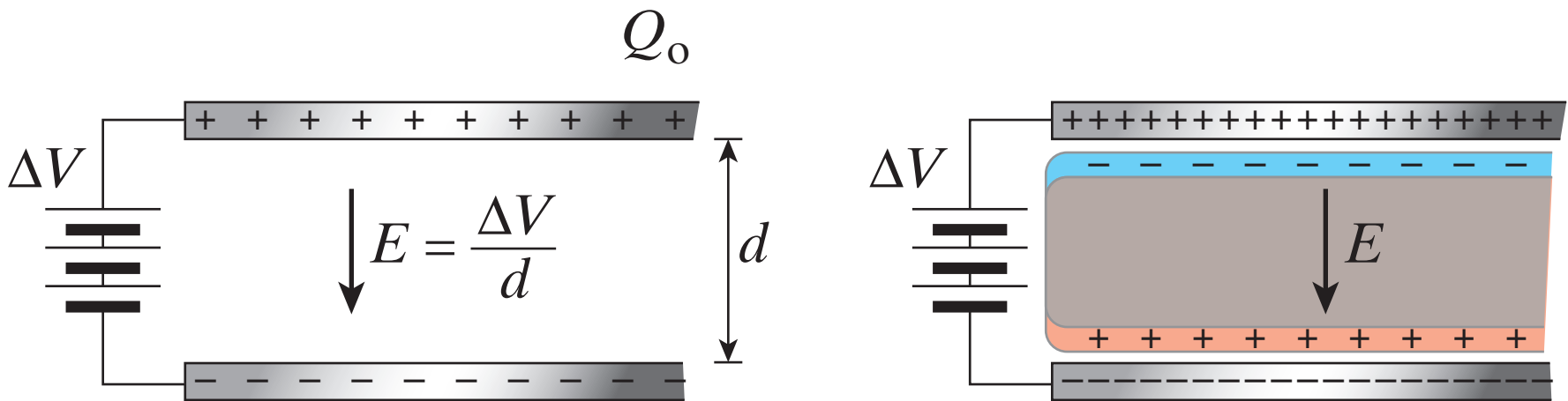
$$\epsilon = \frac{C_d}{C_o}$$



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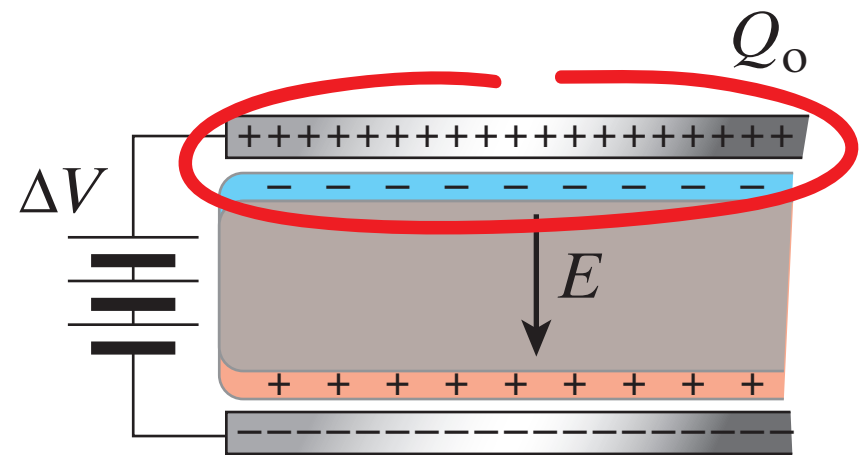
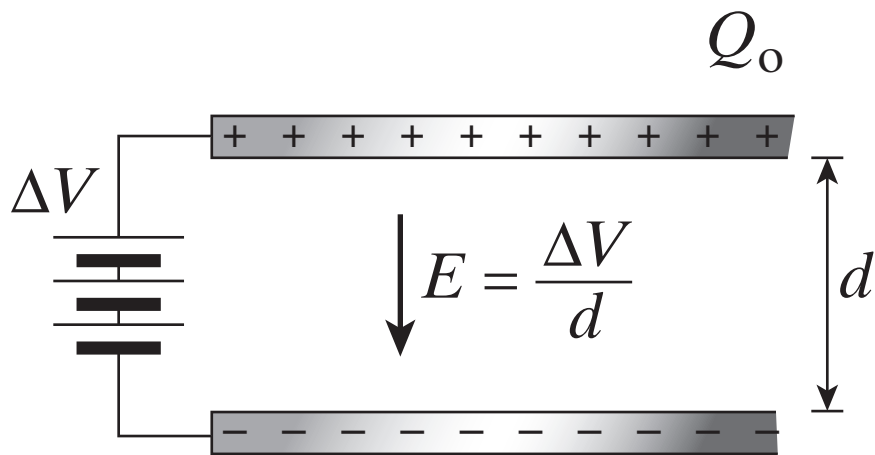
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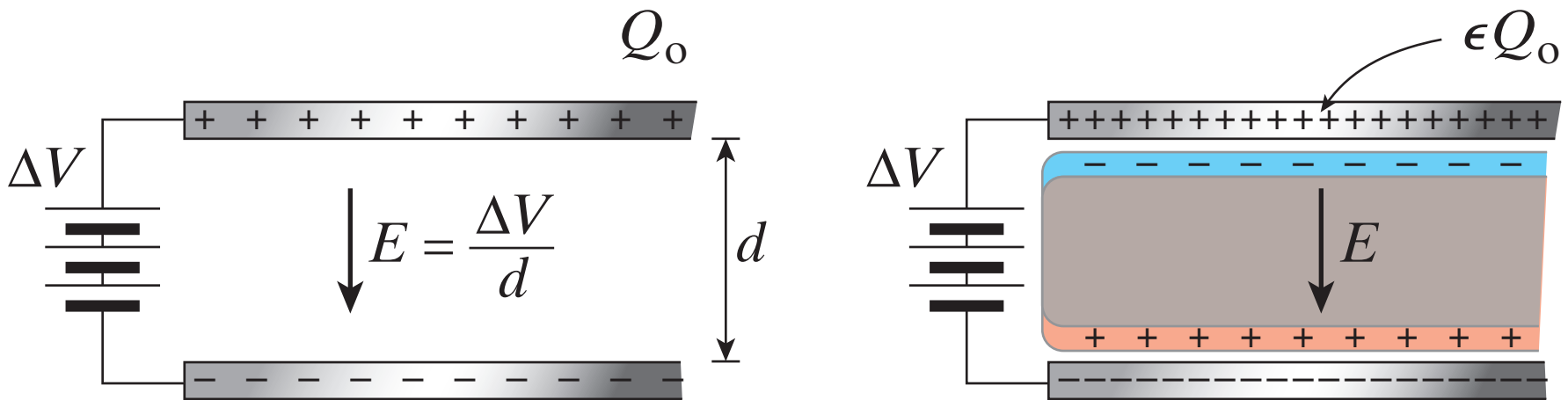
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Propagation of EM waves through medium

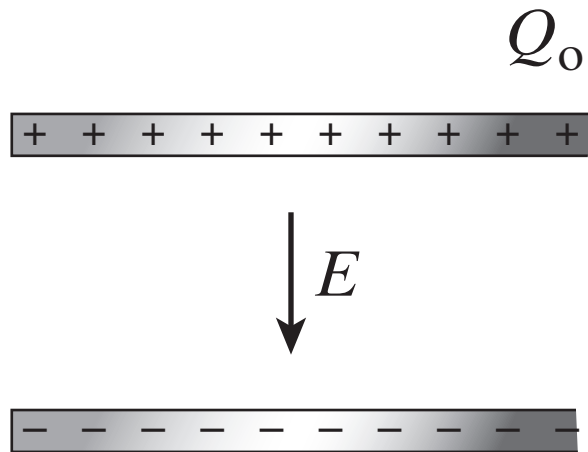
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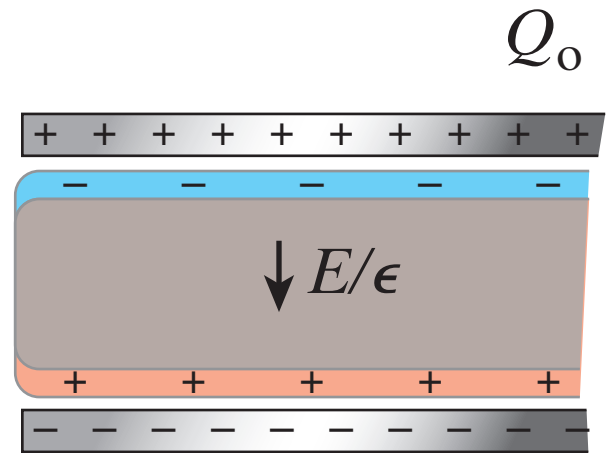
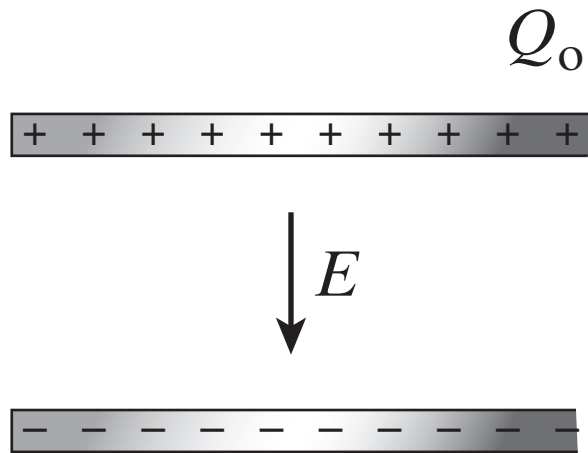
Propagation of EM waves through medium

Alternatively, ϵ is measure of the attenuation of the field



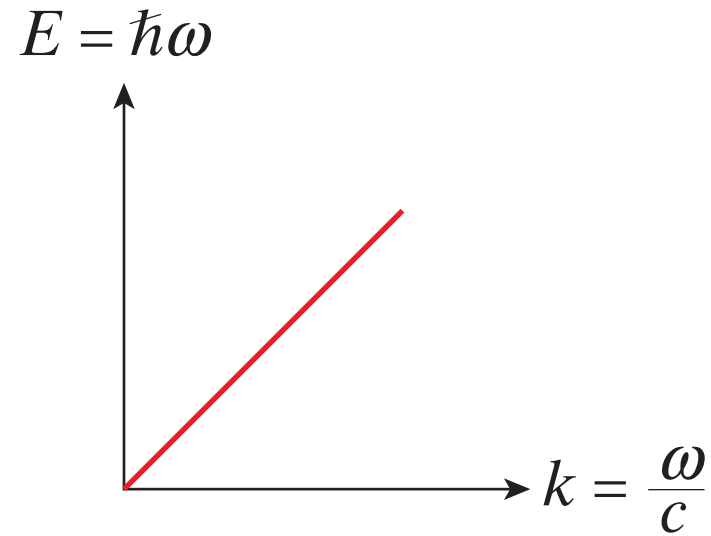
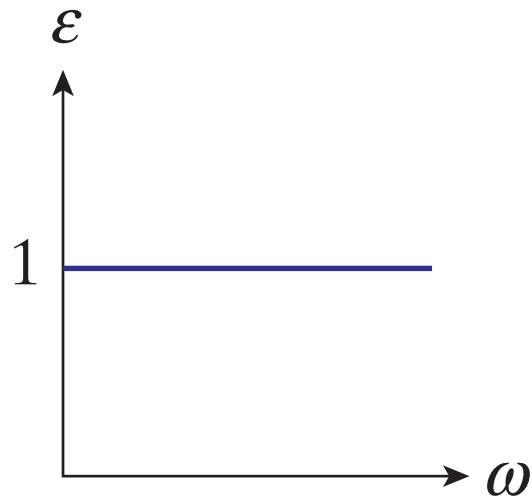
Propagation of EM waves through medium

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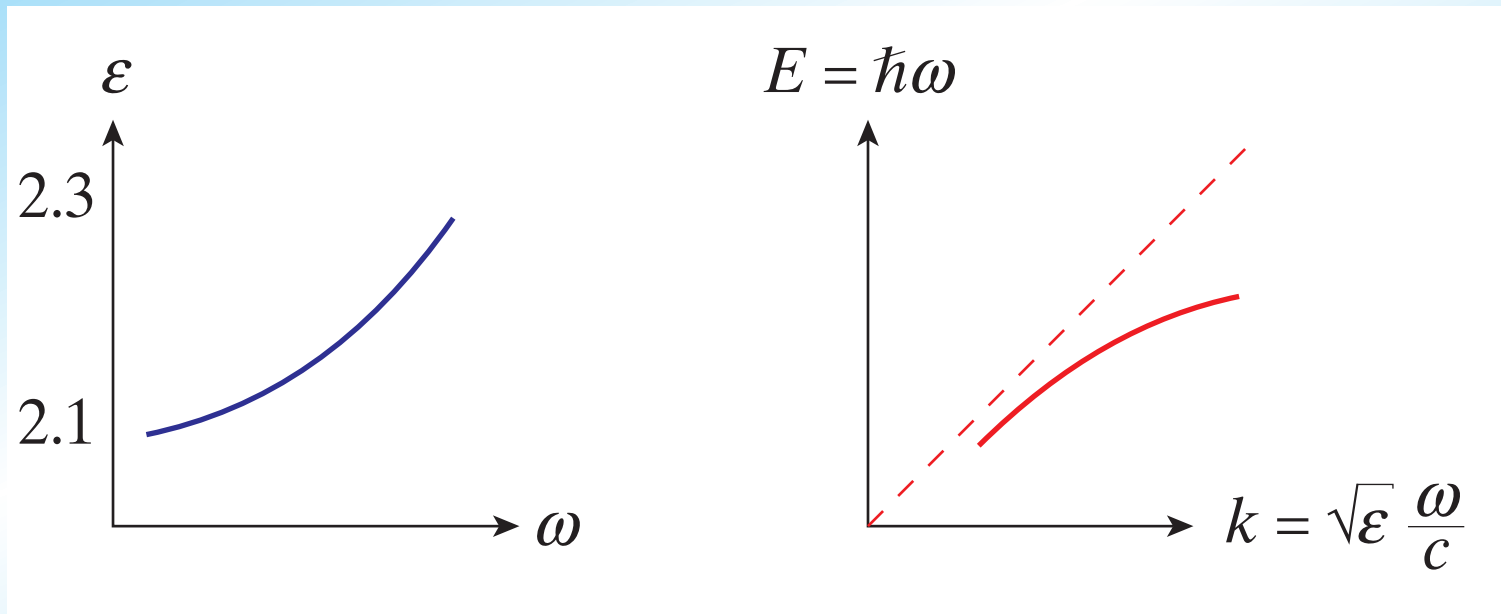
Propagation of EM waves through medium

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

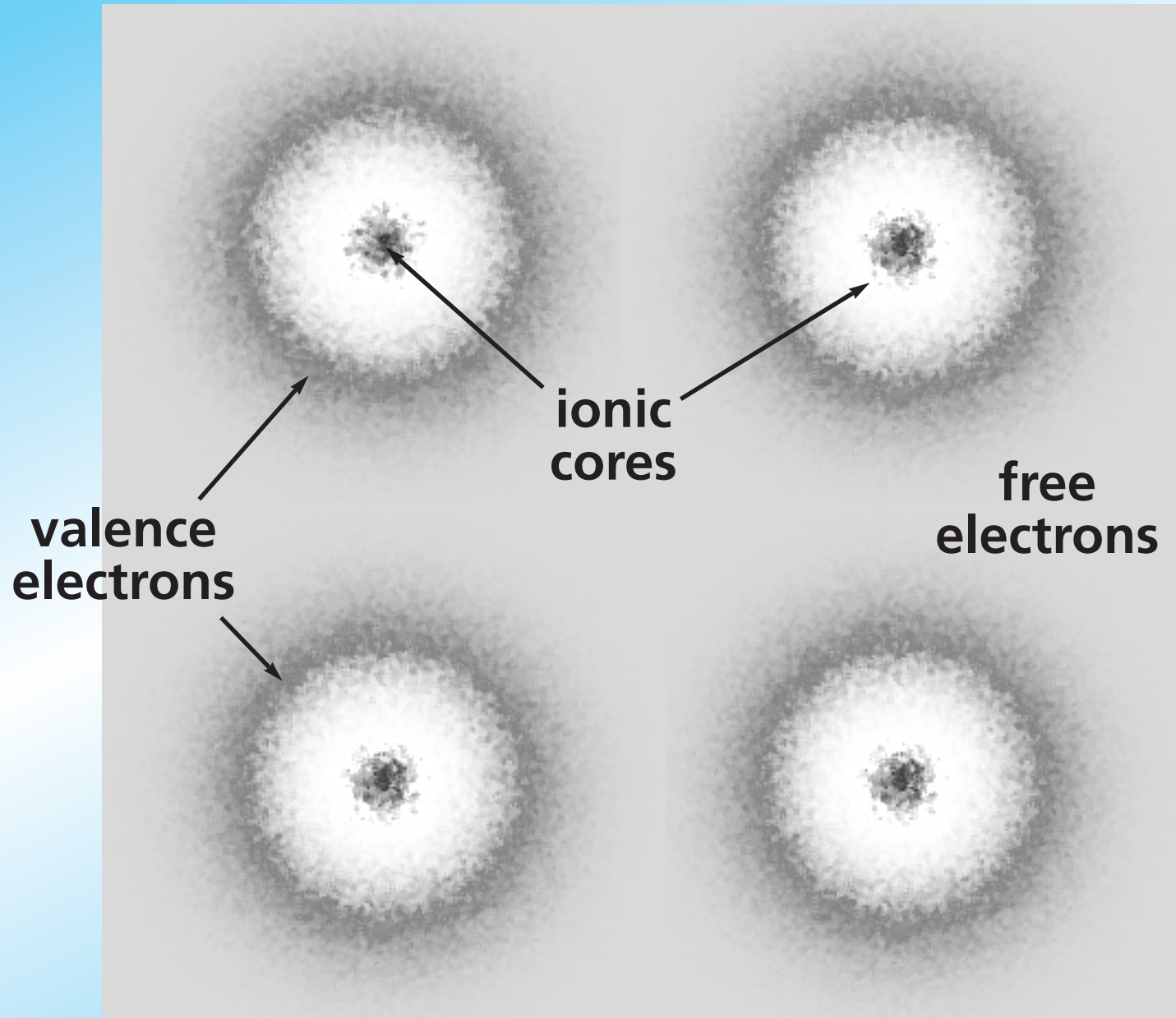


Propagation of EM waves through medium

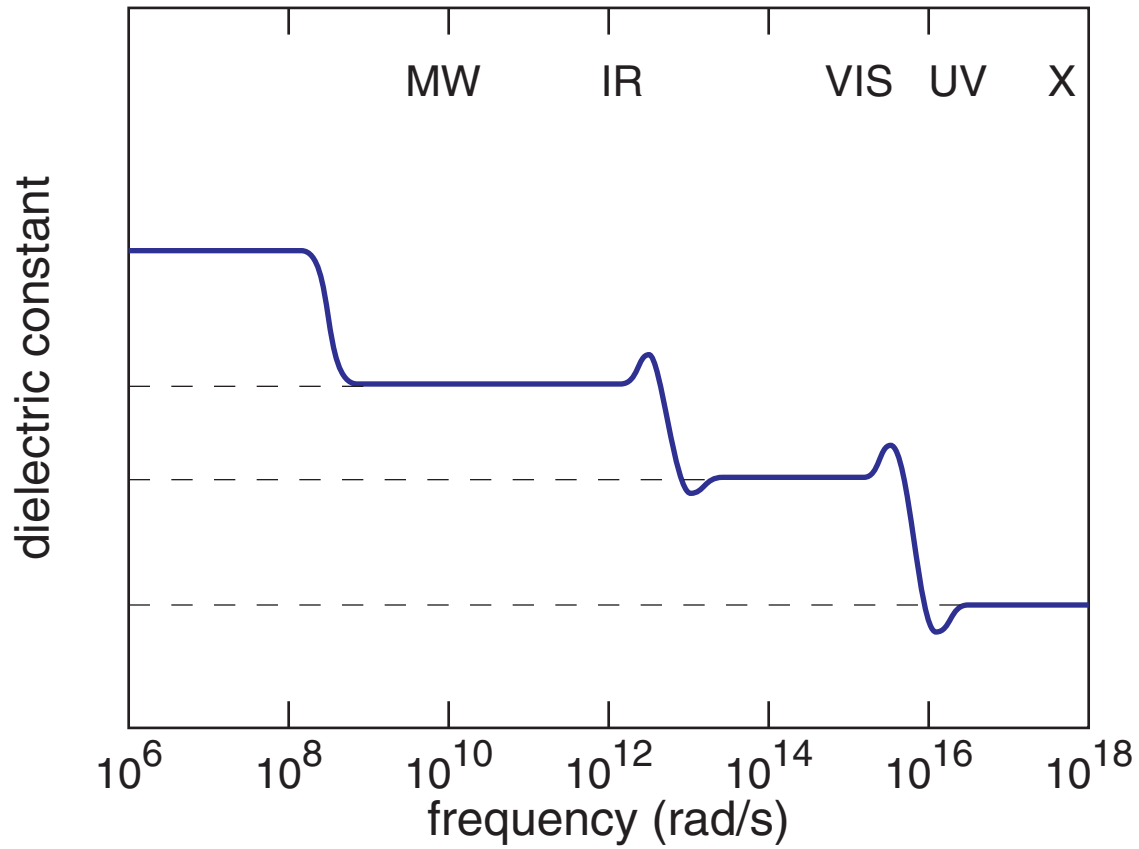
In medium: $v = \frac{c}{\sqrt{\epsilon(\omega)}} = \frac{c}{n} \quad \Rightarrow \quad \omega = \frac{c}{\sqrt{\epsilon}} k$



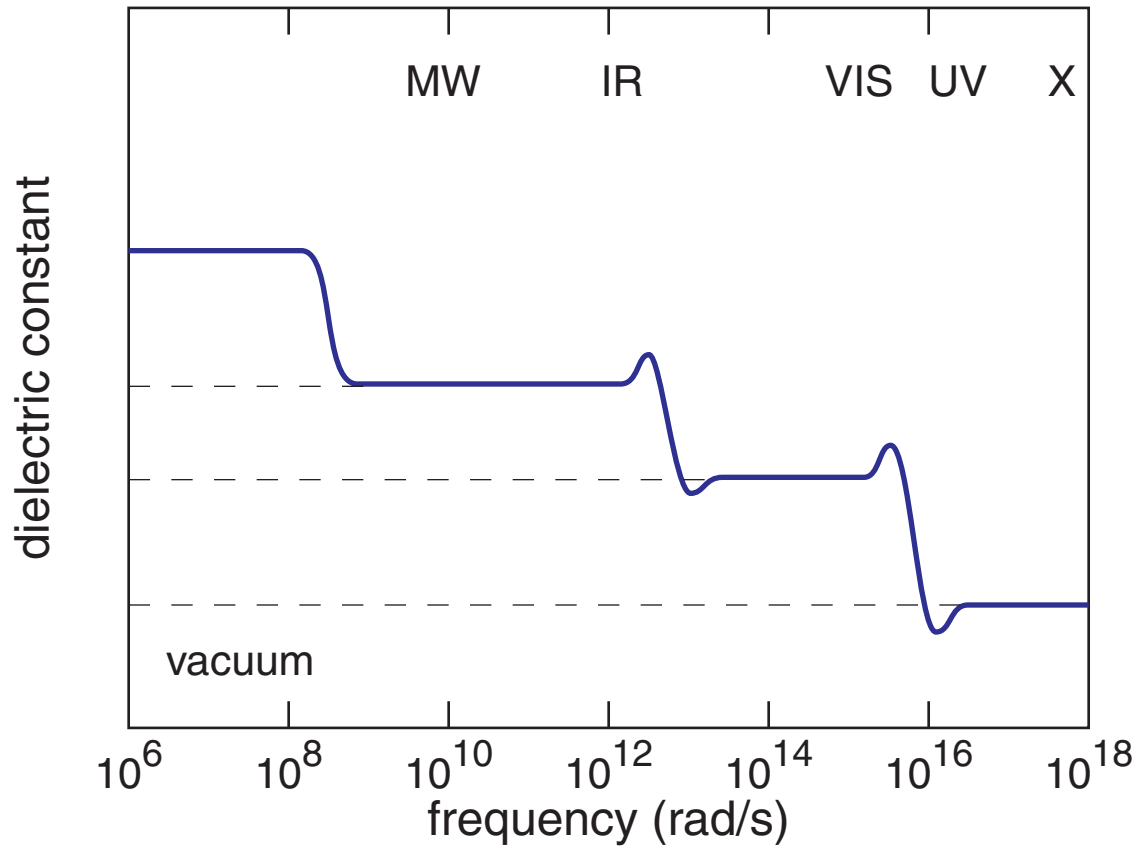
Which charges participate?



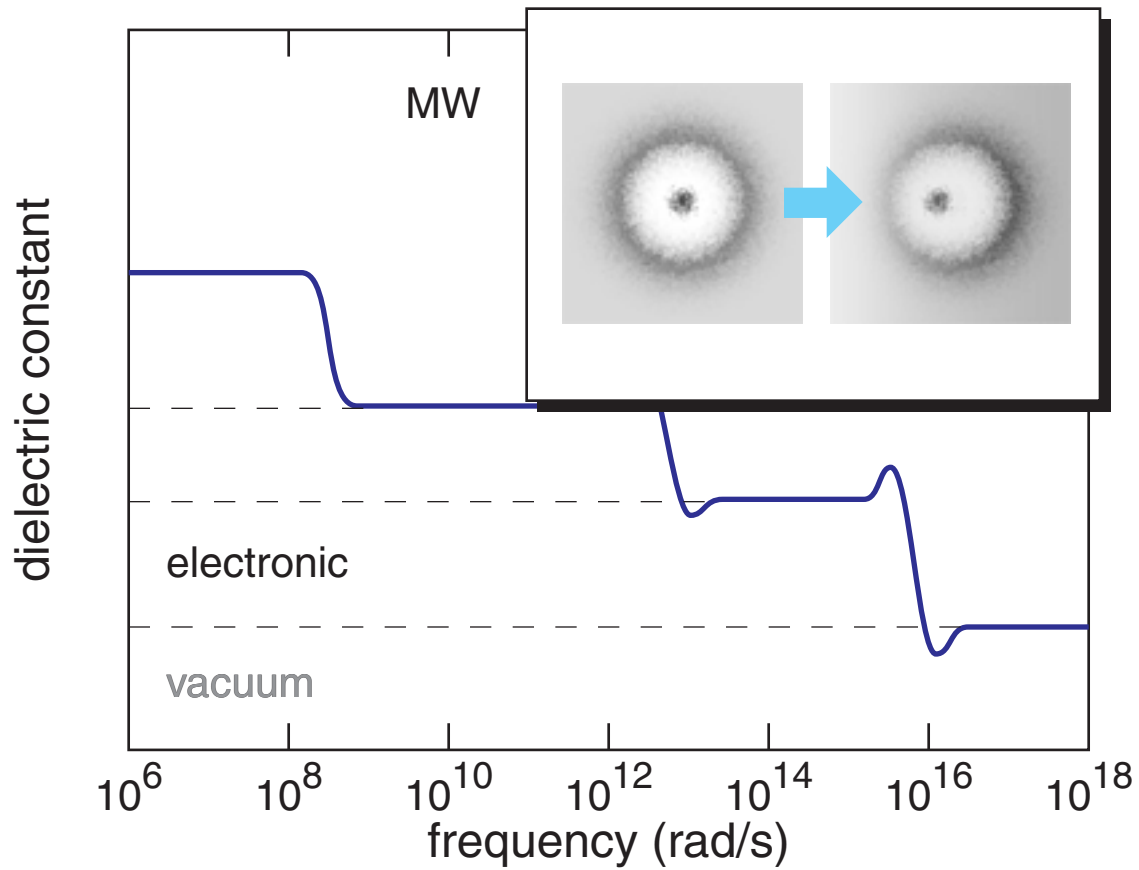
Dielectric function



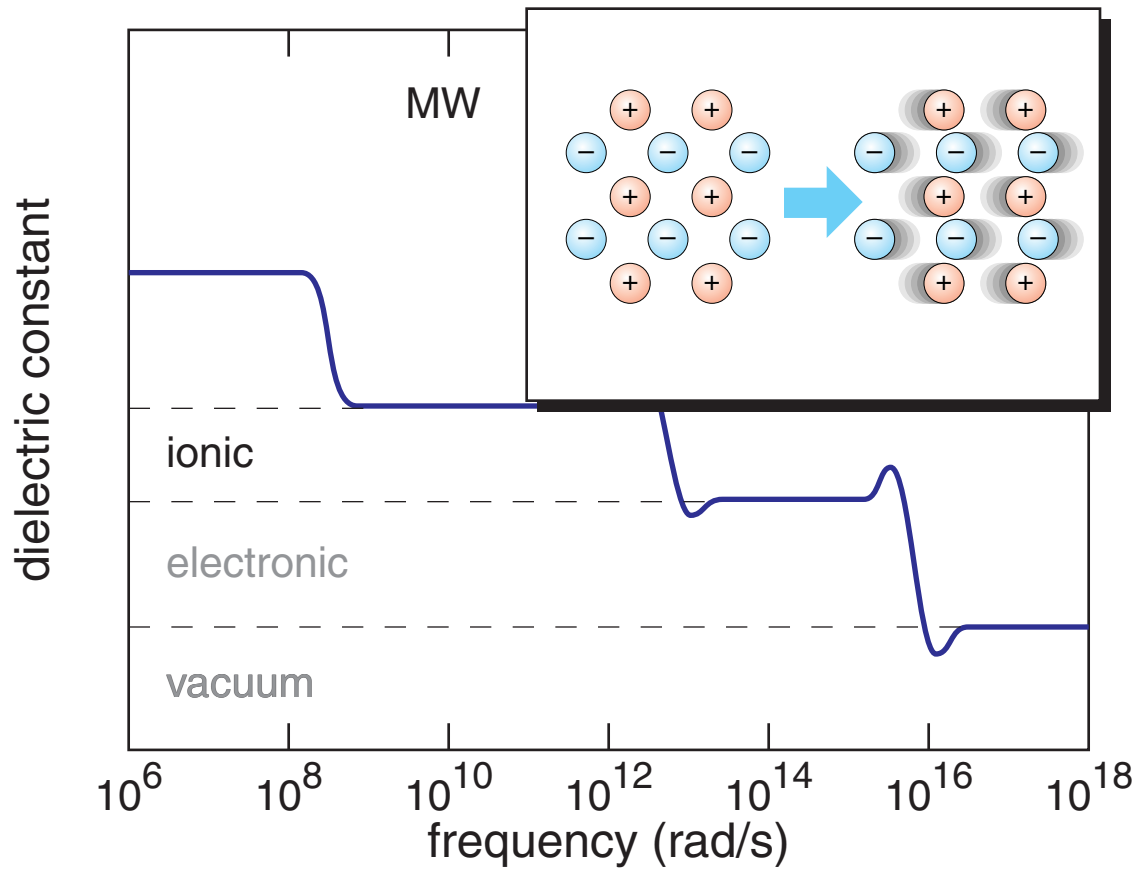
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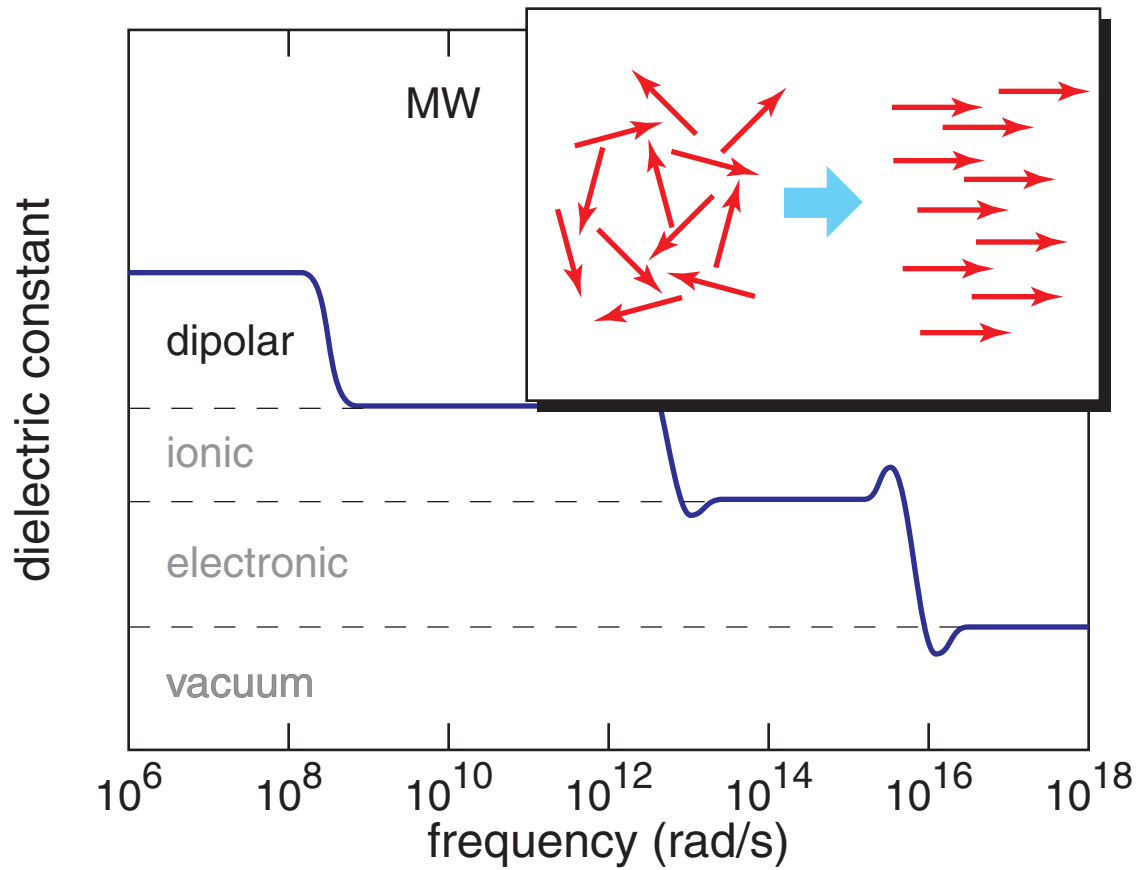
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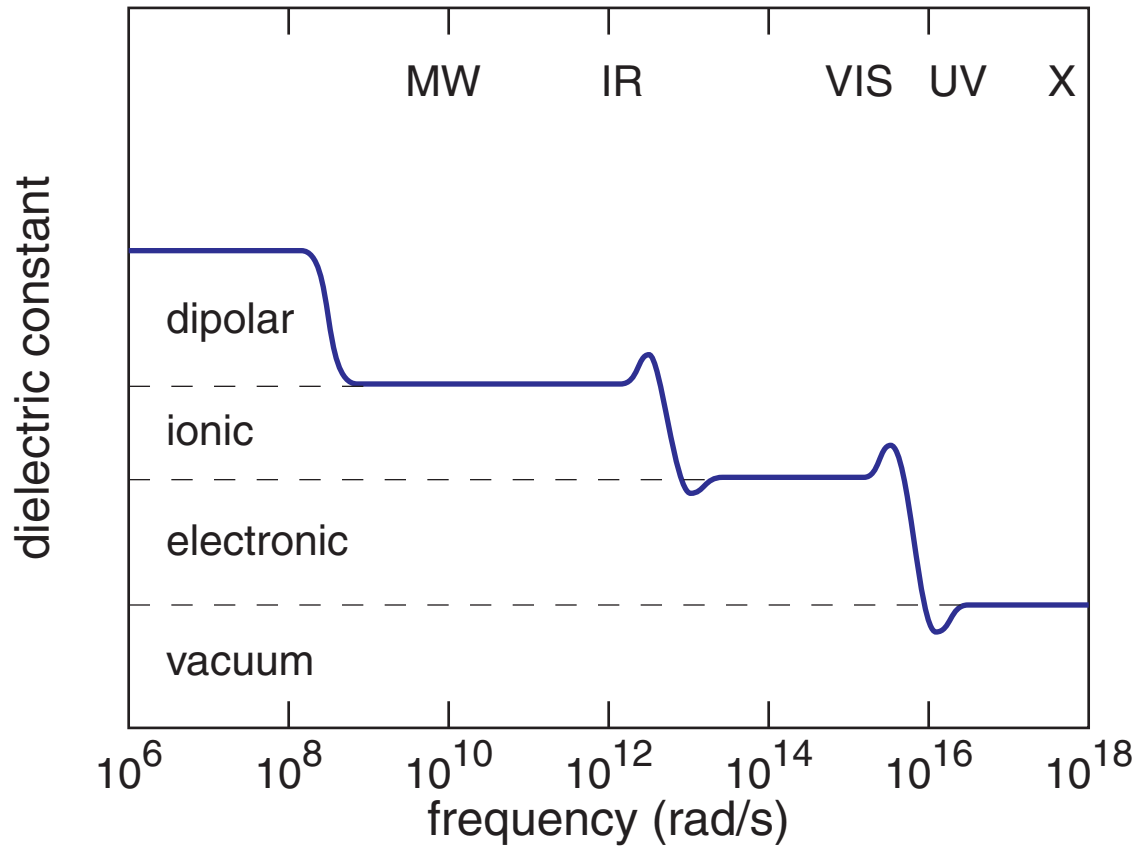
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Bound electrons

Electron on a string:

$$F_{binding} = - m_e \omega_o^2 x$$

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$$m \frac{d^2 x}{dt^2} = \sum F$$

$$m \frac{d^2 x}{dt^2} + m \gamma \frac{dx}{dt} + m \omega_o^2 x = -eE$$

Bound electrons

Steady state: electron oscillates at driving frequency

$$x(t) = x_o e^{-i\omega t} \quad x_o = -\frac{e}{m} \frac{1}{(\omega_o^2 - \omega^2) - i\gamma\omega} E_o$$

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

Bound electrons

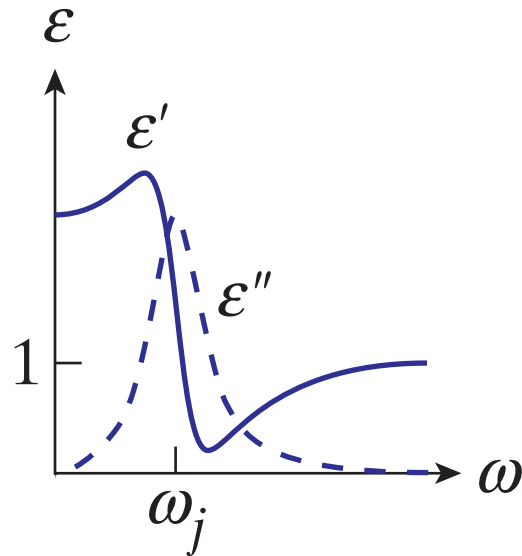
Dielectric function

$$\epsilon(\omega) \equiv 1 + \chi_e = 1 + \frac{Ne^2}{\epsilon_0 m} \sum_j \frac{f_j}{(\omega_j^2 - \omega^2) - i\gamma_j \omega}$$

Bound electrons

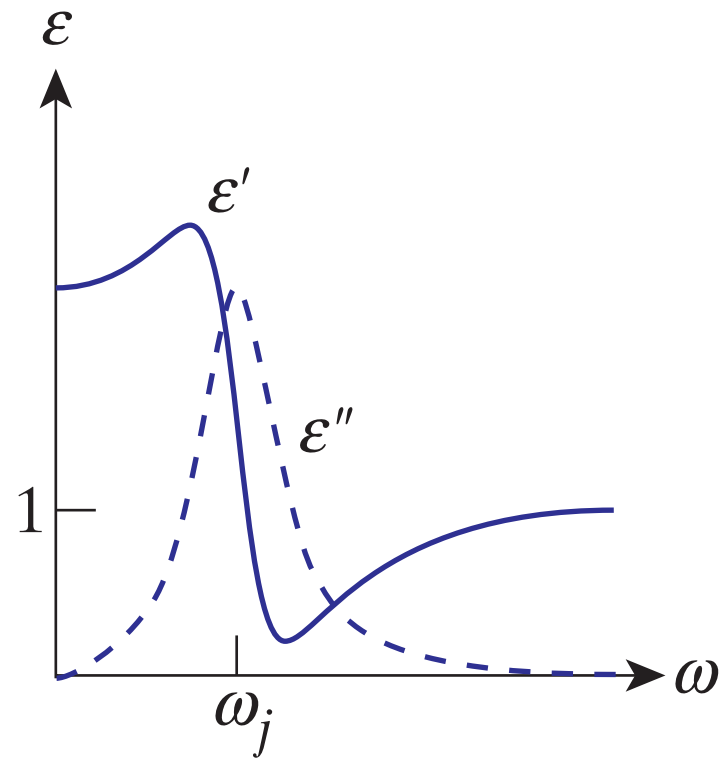
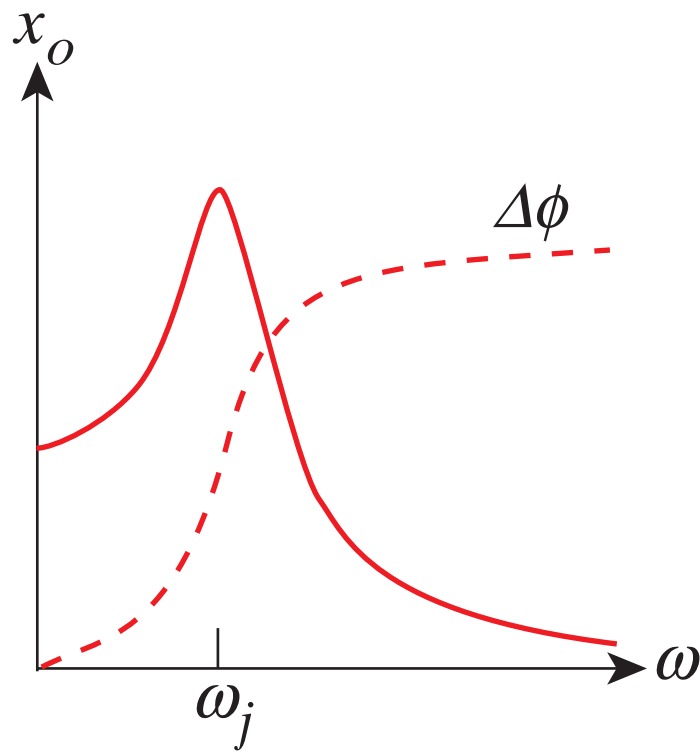
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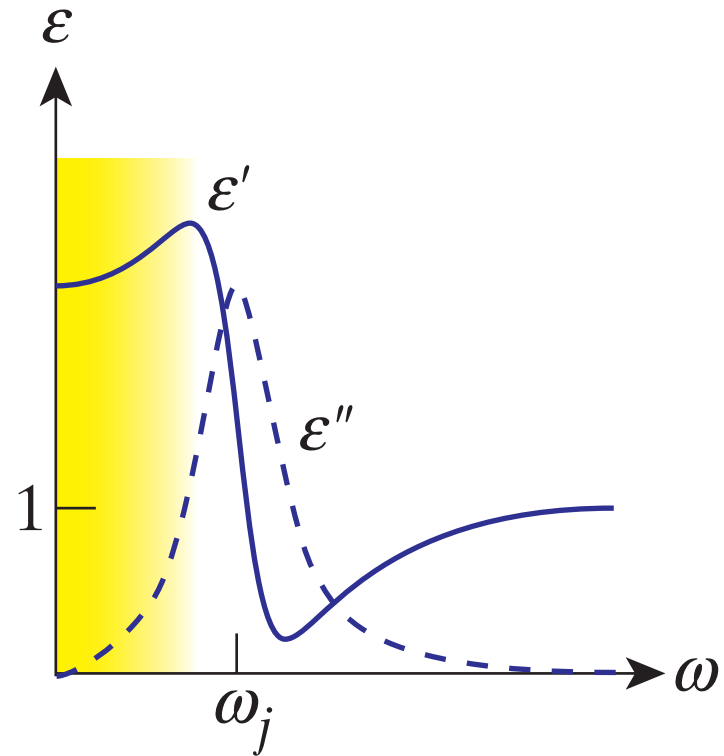
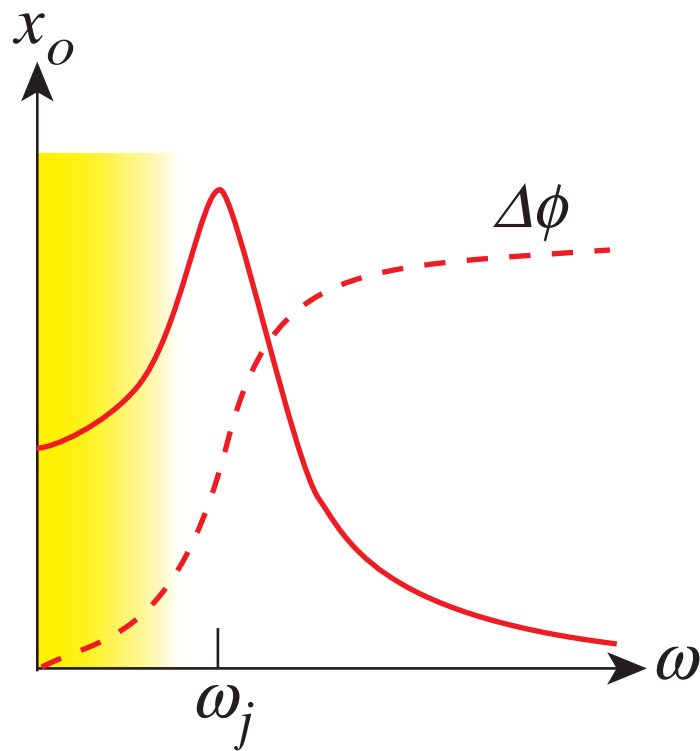
Bound electrons

amplitude of bound charge oscillation



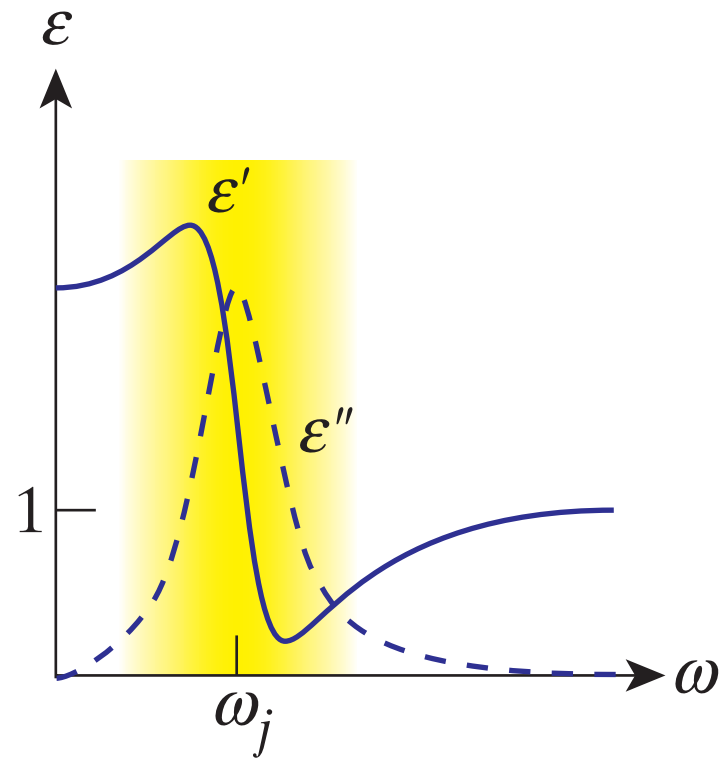
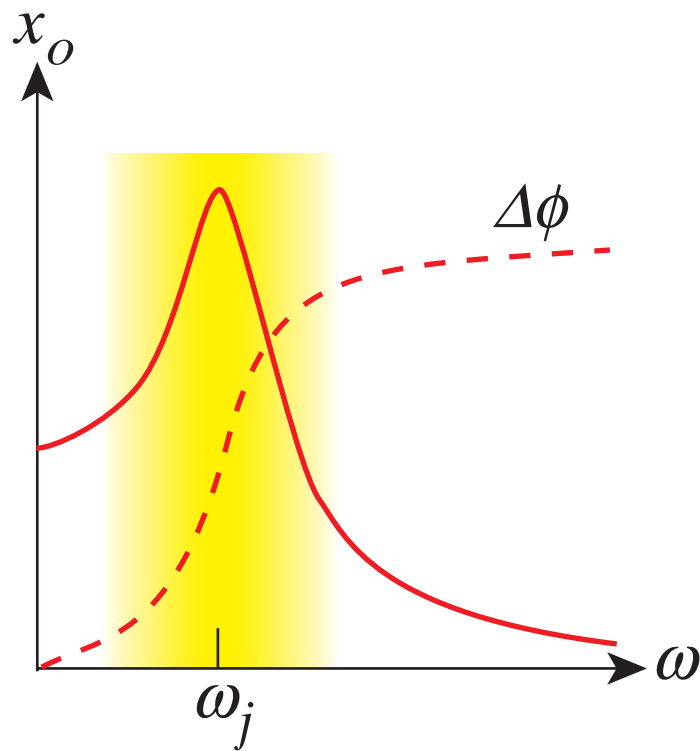
Bound electrons

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



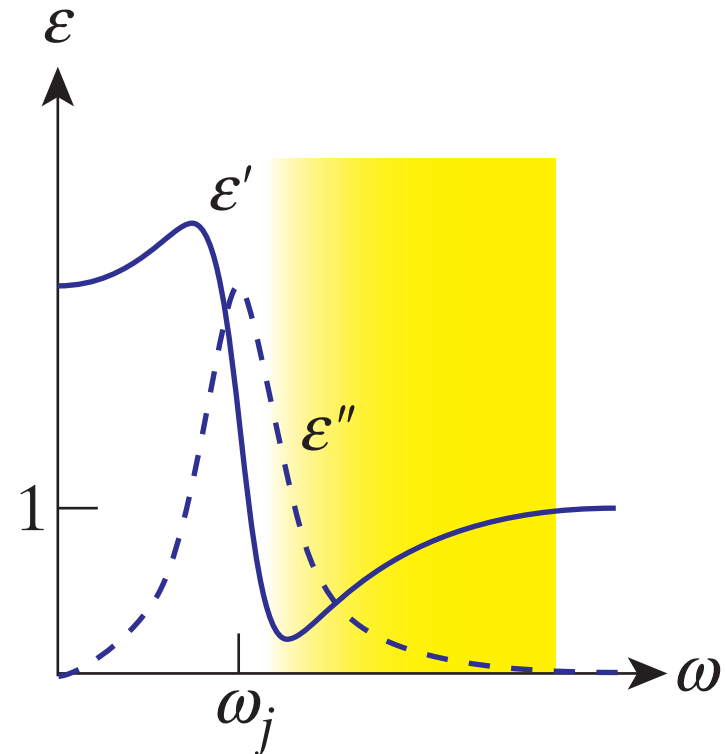
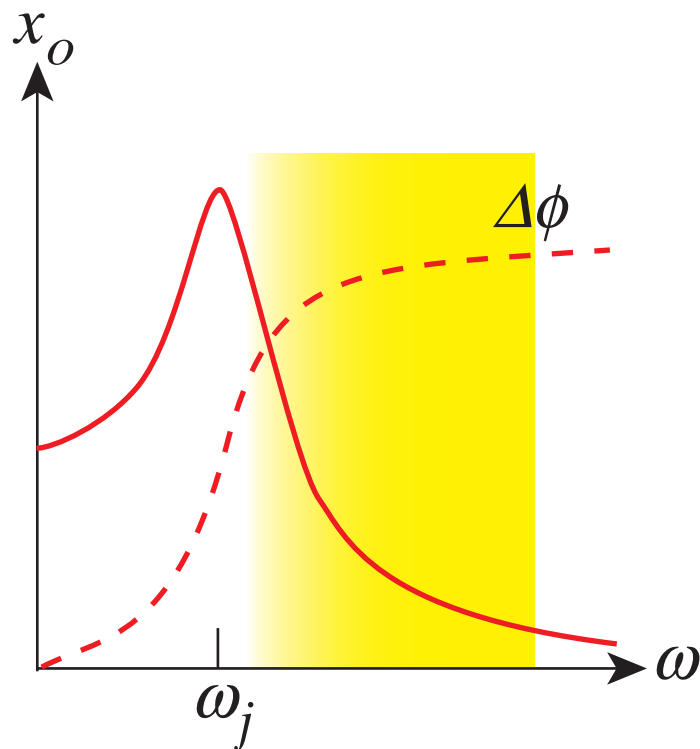
Bound electrons

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



Bound electrons

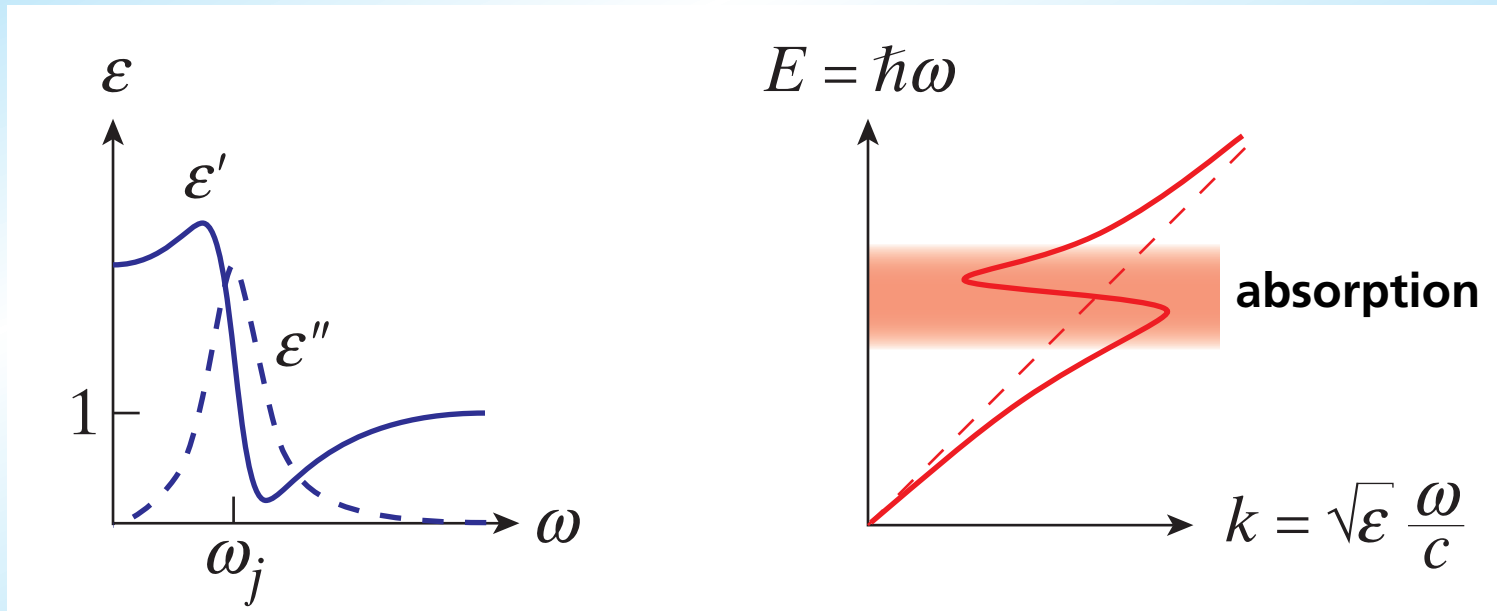
Above resonance: bound charges cannot keep up with driving field \Rightarrow dielectric like a vacuum



Bound electrons

Dielectric function

$$\epsilon(\omega) \equiv 1 + \chi_e = 1 + \frac{Ne^2}{\epsilon_0 m} \sum_j \frac{f_j}{(\omega_j^2 - \omega^2) - i\gamma_j \omega}$$



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Low frequency ($\omega \ll 1$) \Rightarrow current generated

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

Free electrons

$\omega \gg \gamma$: σ complex $\Rightarrow J$ out of phase with E

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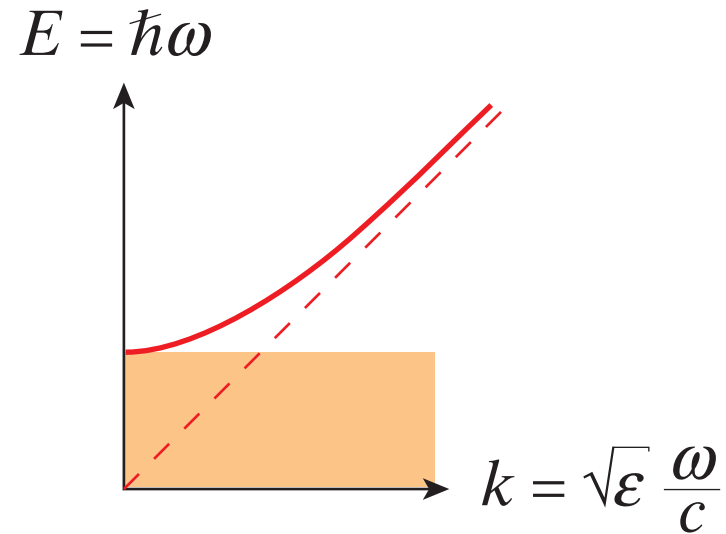
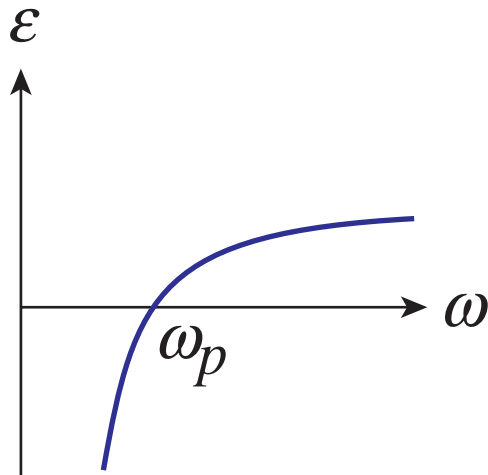
Dielectric function

$$\epsilon(\omega) \equiv 1 + \chi_e = 1 - \frac{Ne^2}{m\epsilon_0} \frac{1}{\omega^2 + i\gamma\omega} = \epsilon'(\omega) + i\epsilon''(\omega)$$

Plasma

$$\gamma \approx 0 \quad \Rightarrow \quad \epsilon'' = 0$$

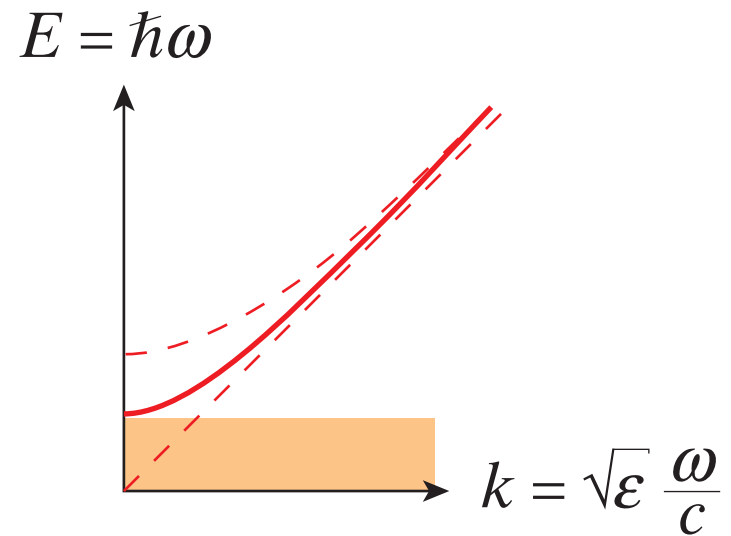
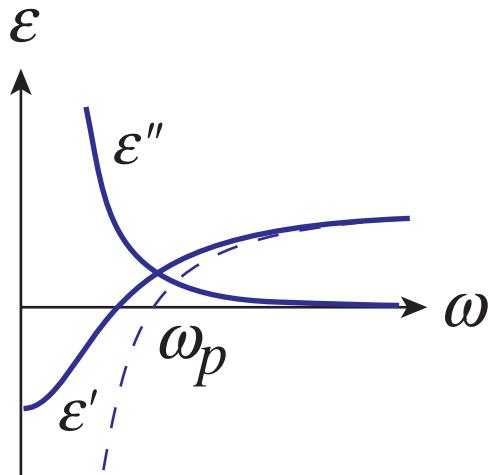
$$\epsilon'(\omega) = 1 - \frac{Ne^2}{m\epsilon_0} \frac{1}{\omega^2} \equiv 1 - \frac{\omega_p^2}{\omega^2}$$



Plasma

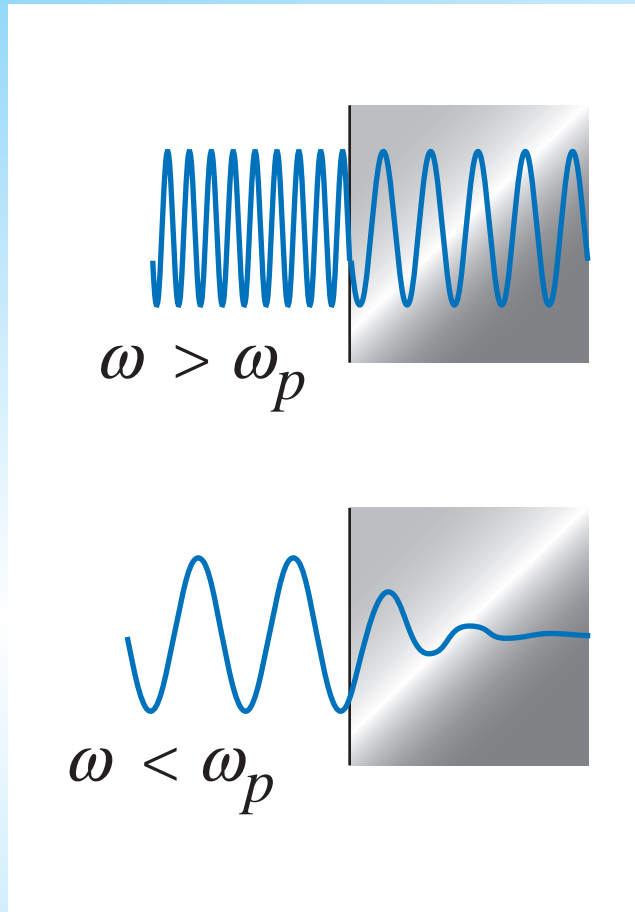
Add damping

$$\gamma \lesssim \omega_p$$



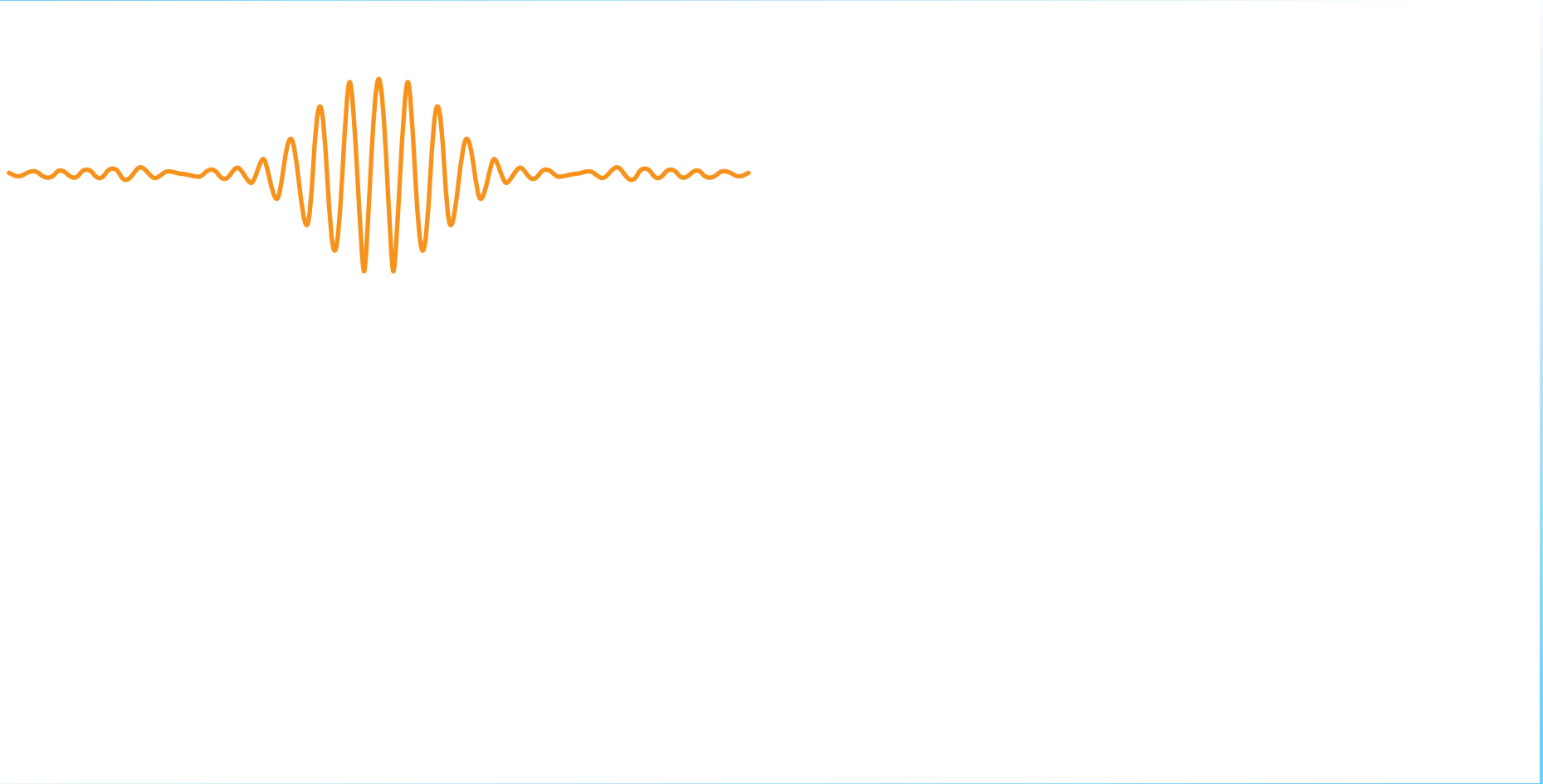
Plasma

Plasma acts like a high-pass filter:

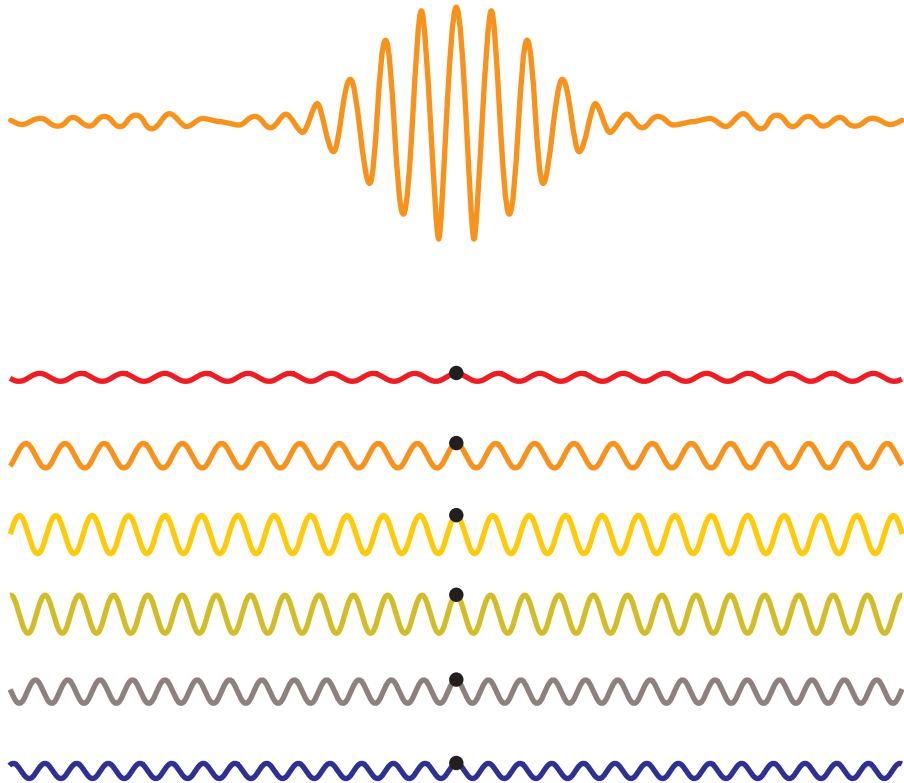


$\log N$ (cm^{-3})	ω_p (rad s^{-1})	λ_p
22	6×10^{15}	330 nm
18	6×10^{13}	33 μm
14	6×10^{11}	3.3 mm
10	6×10^9	0.33 m

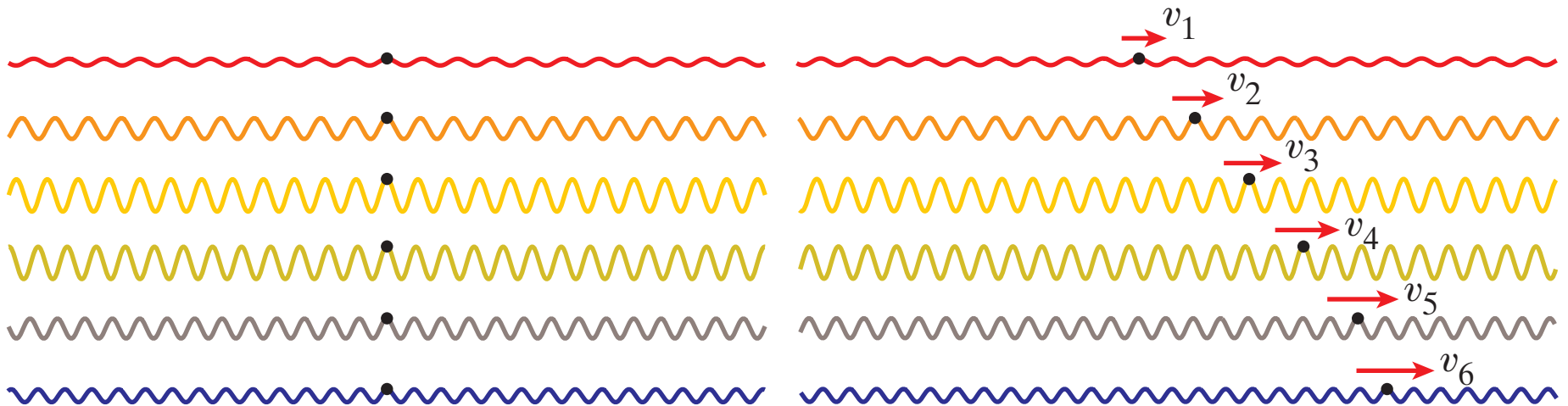
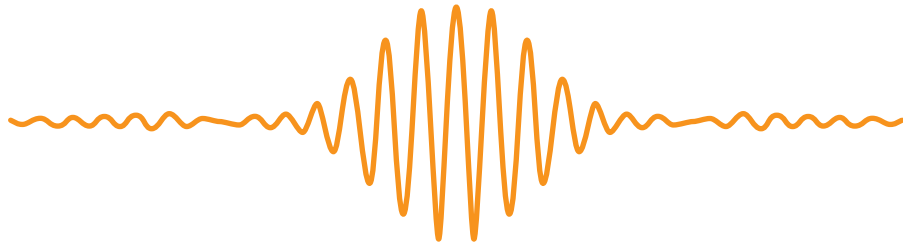
Pulse dispersion



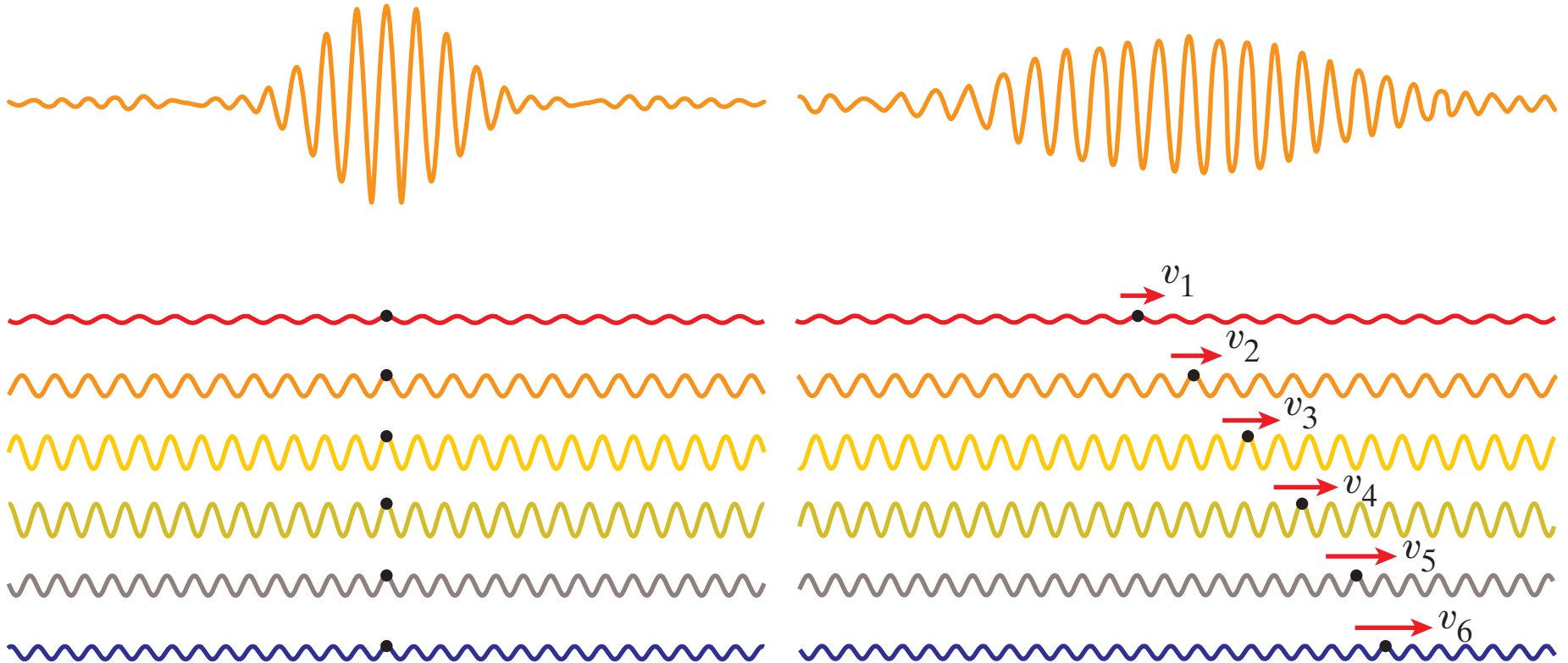
Pulse dispersion



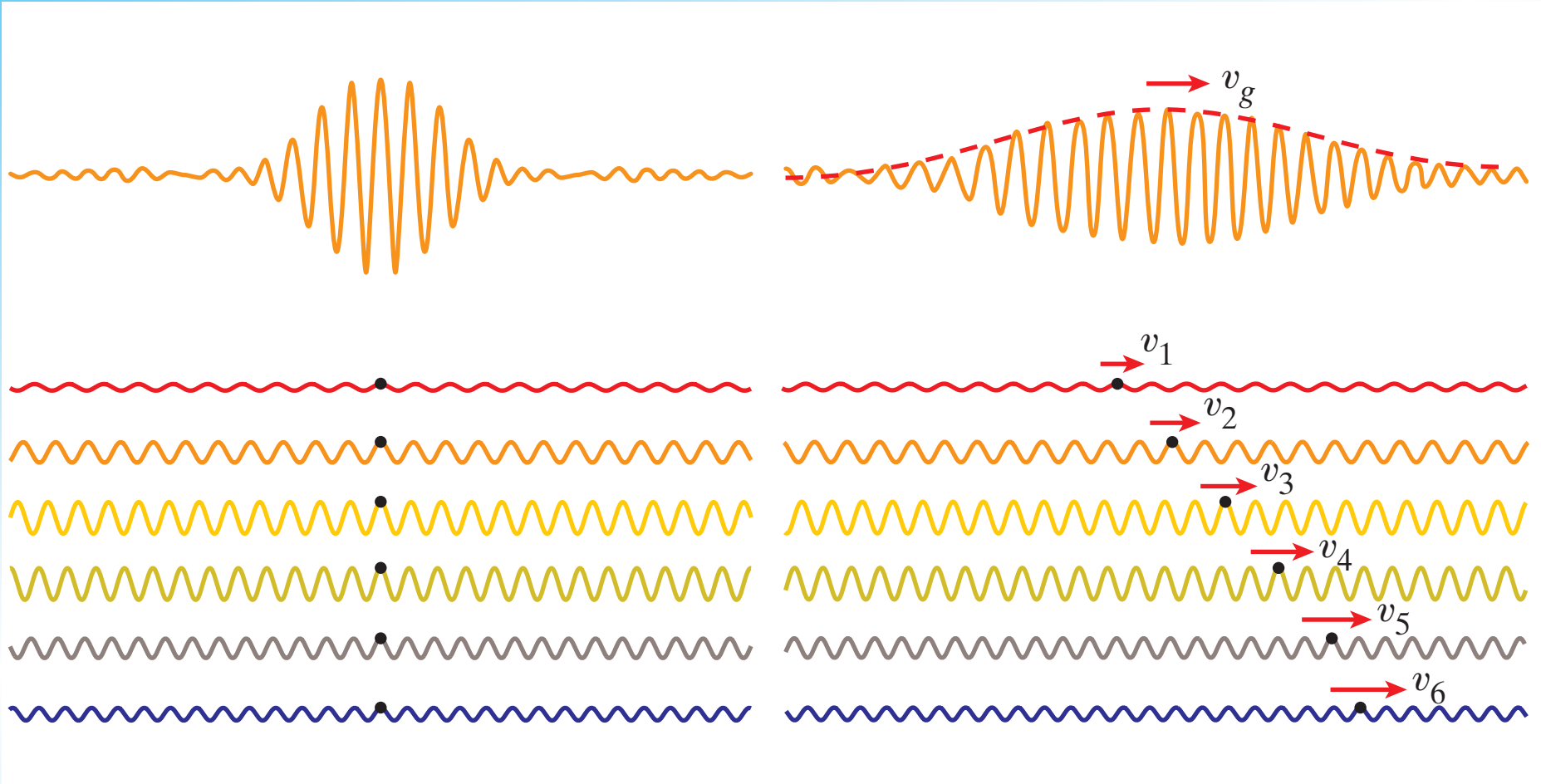
Pulse dispersion



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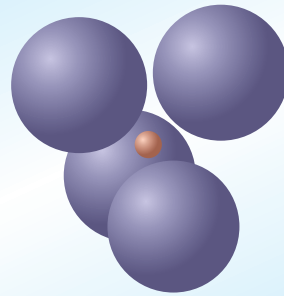
Pulse dispersion



Nonlinear optics

Linear response

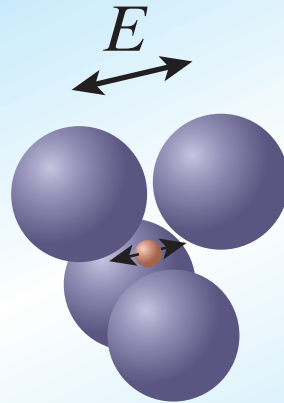
$$P(t) = \epsilon_0 \chi_e E(t)$$



Nonlinear optics

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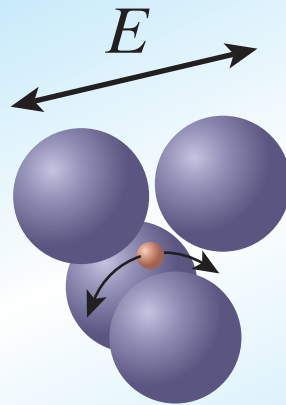
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Nonlinear optics

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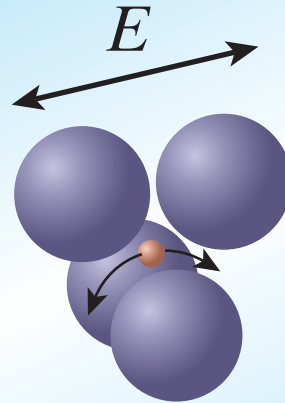
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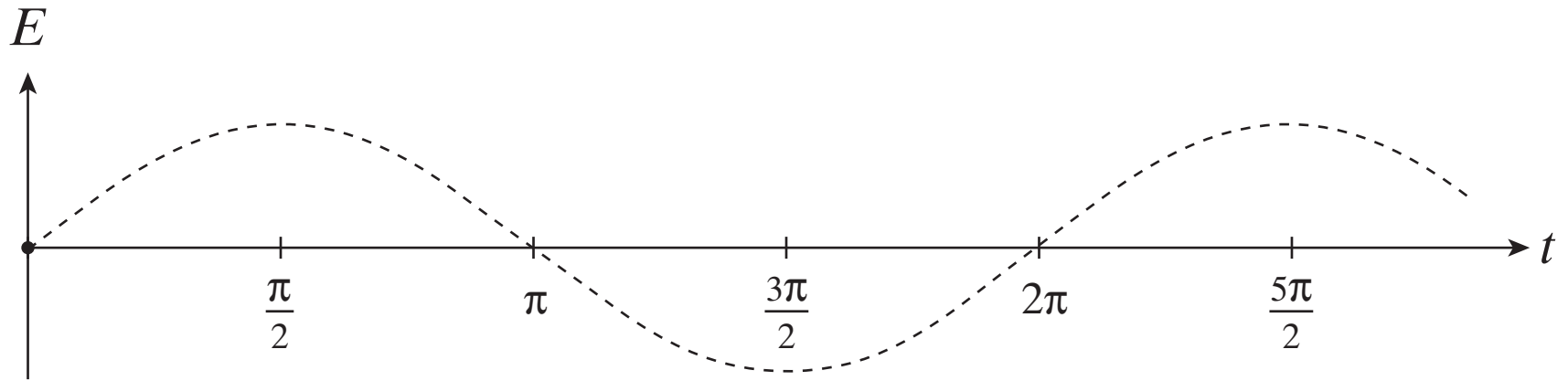


Nonlinear polarization:

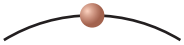
$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots$$

$$P = P^{(1)} + P^{(2)} + P^{(3)} + \dots$$

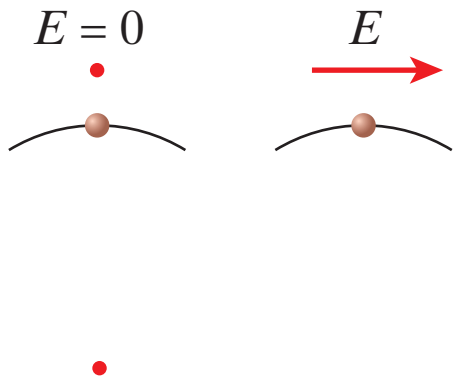
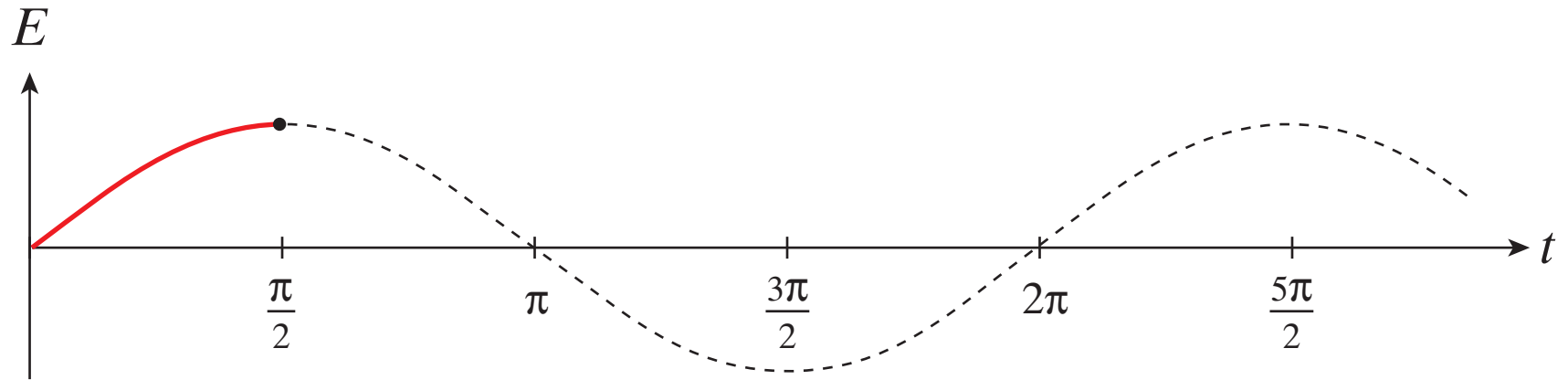
Nonlinear optics



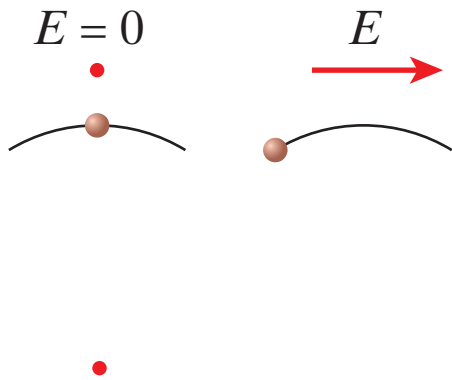
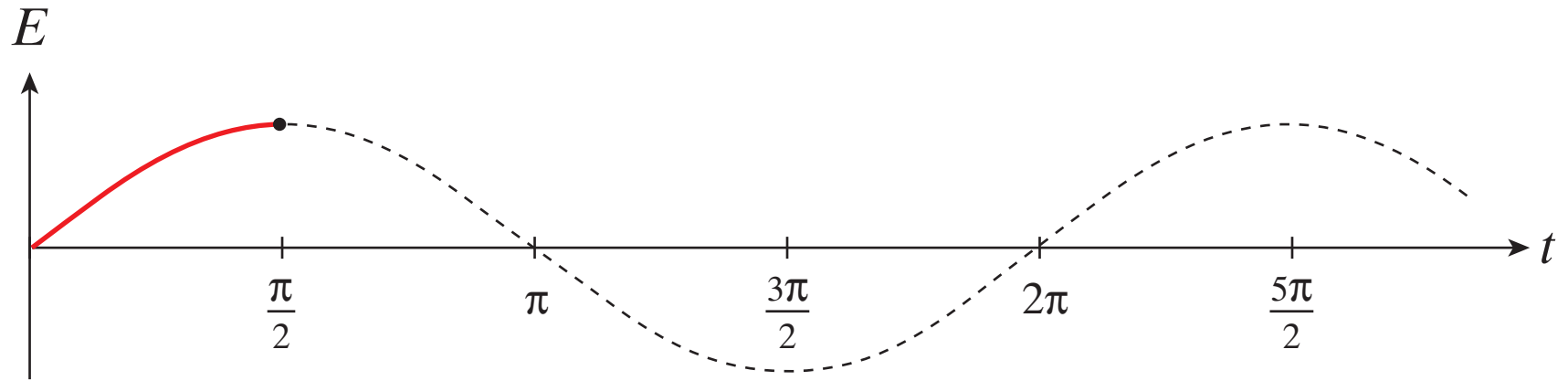
$E = 0$



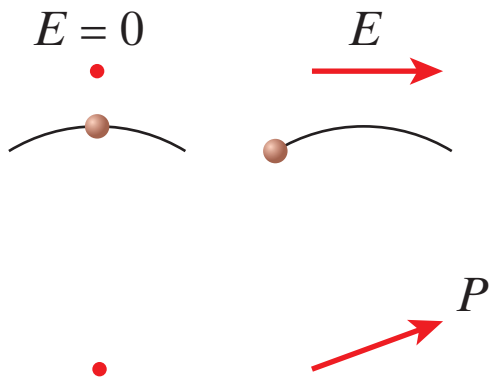
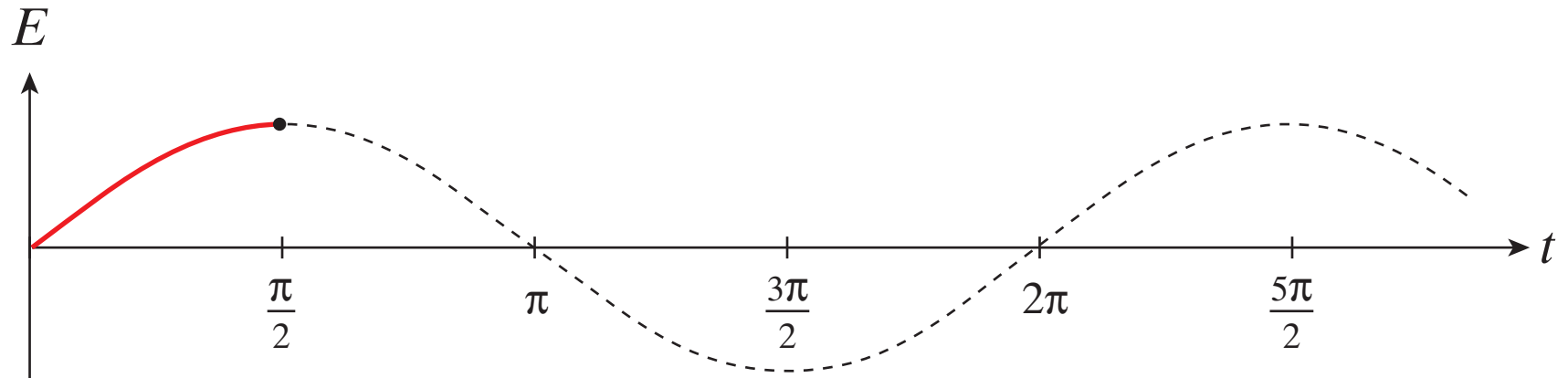
Nonlinear optics



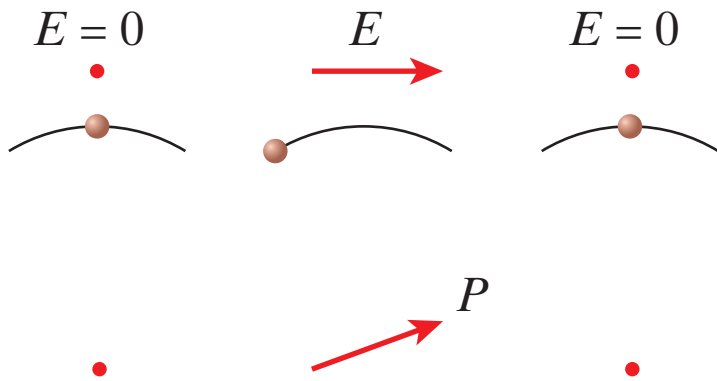
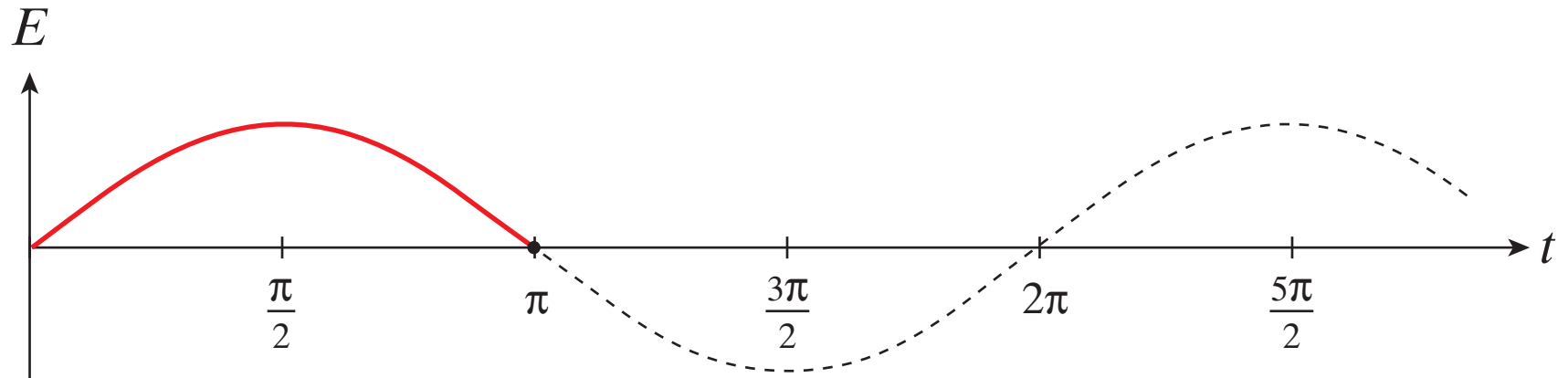
Nonlinear optics



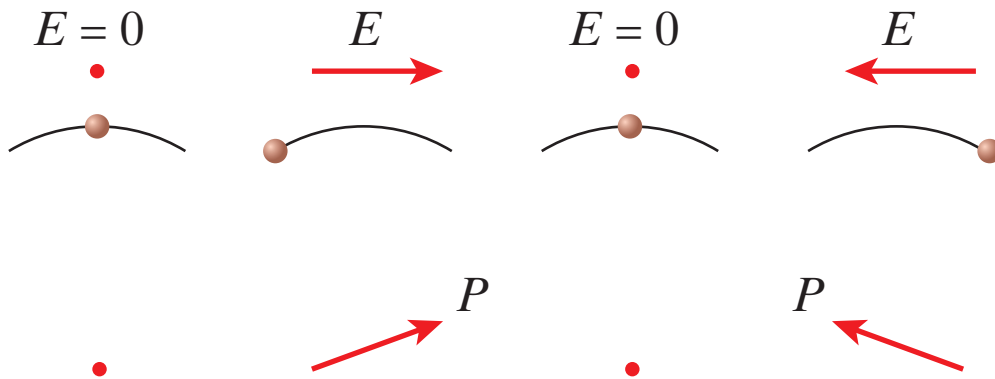
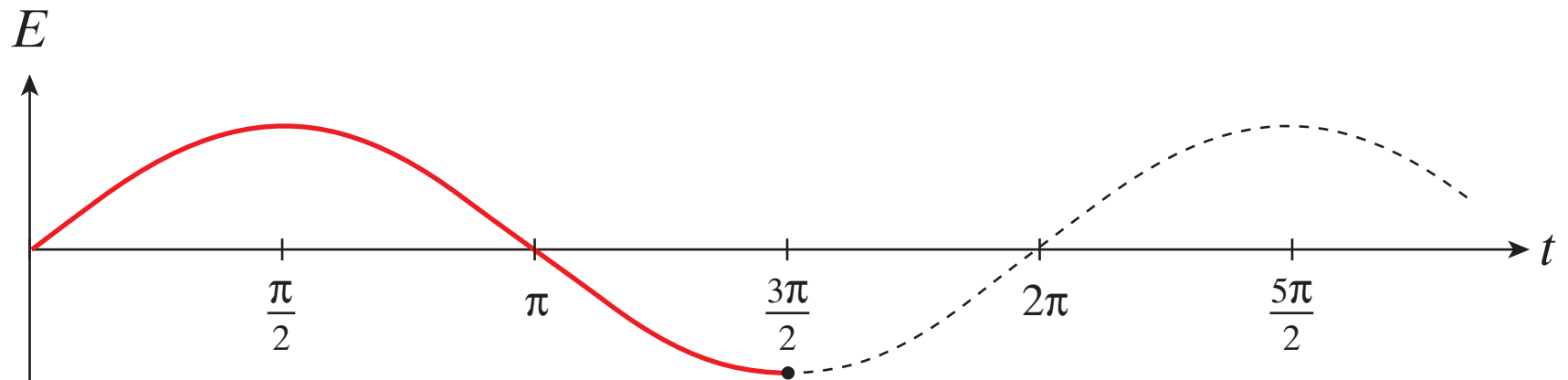
Nonlinear optics



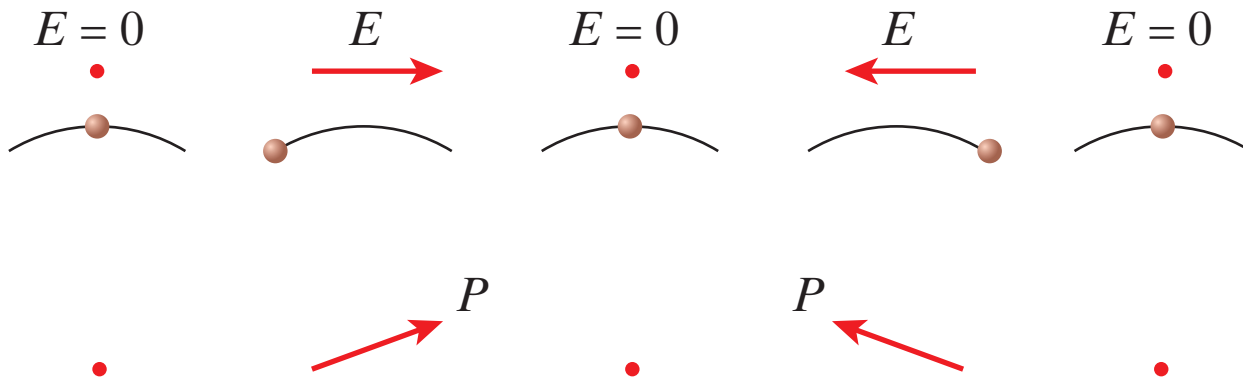
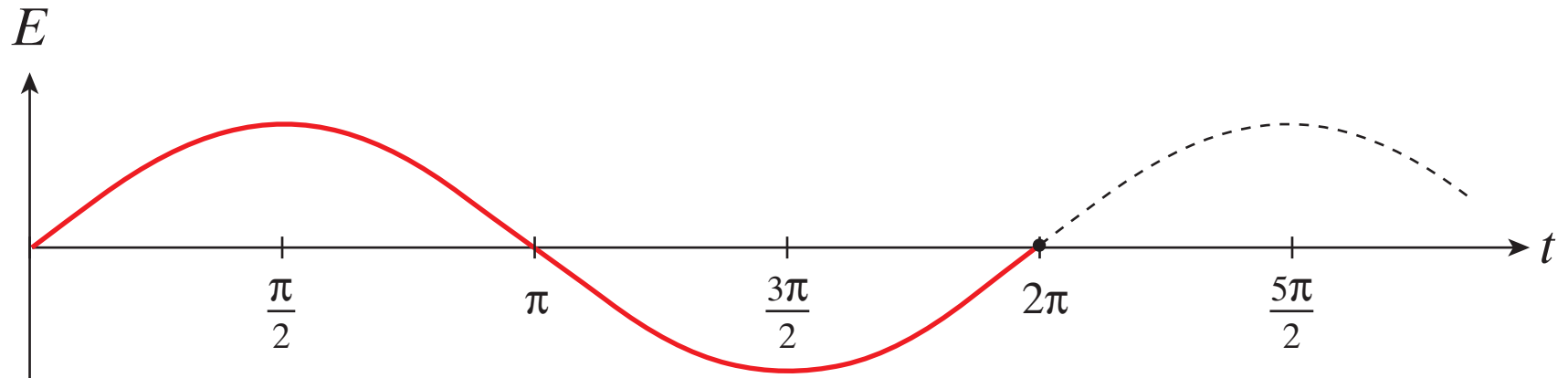
Nonlinear optics



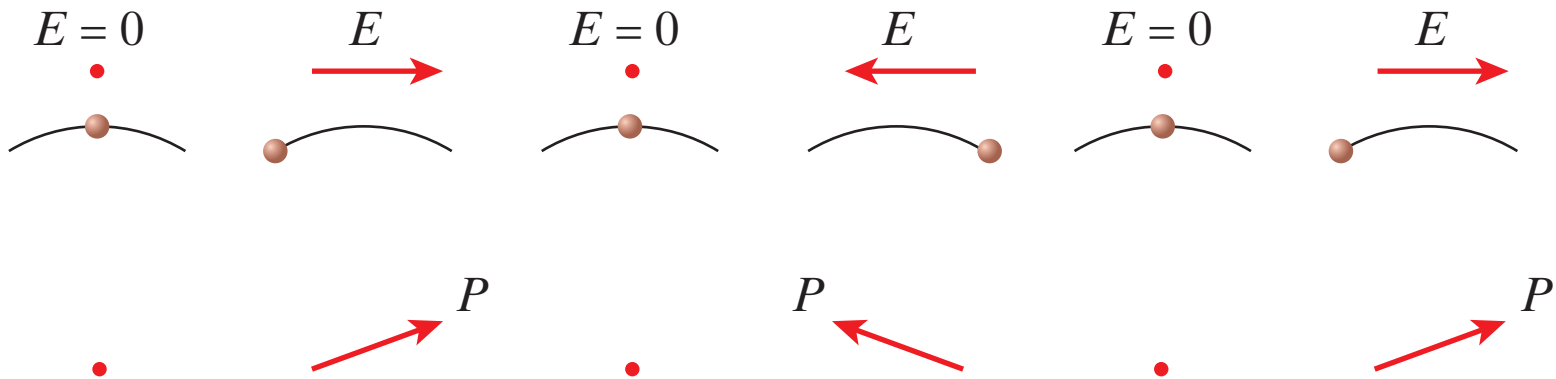
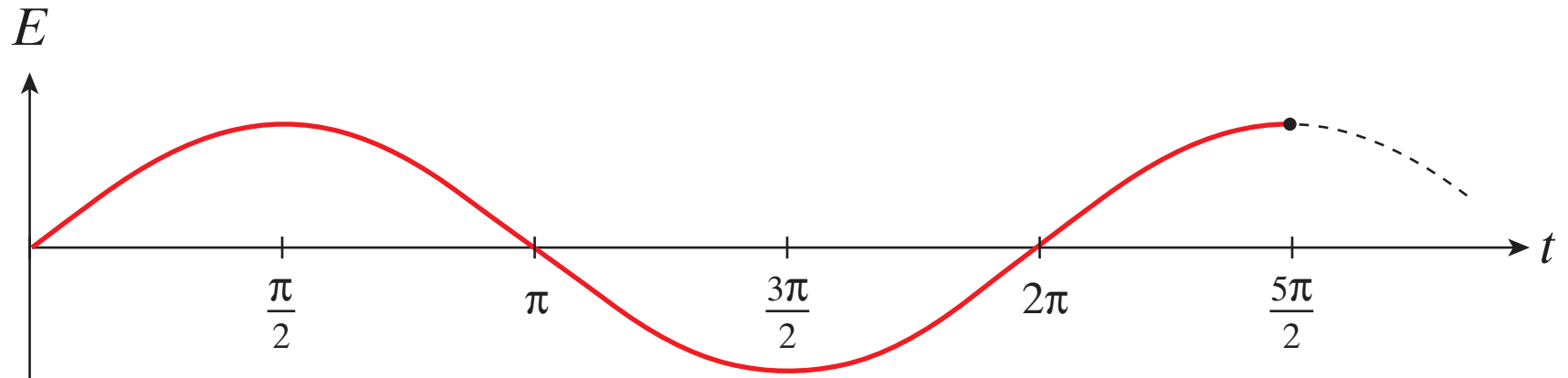
Nonlinear optics



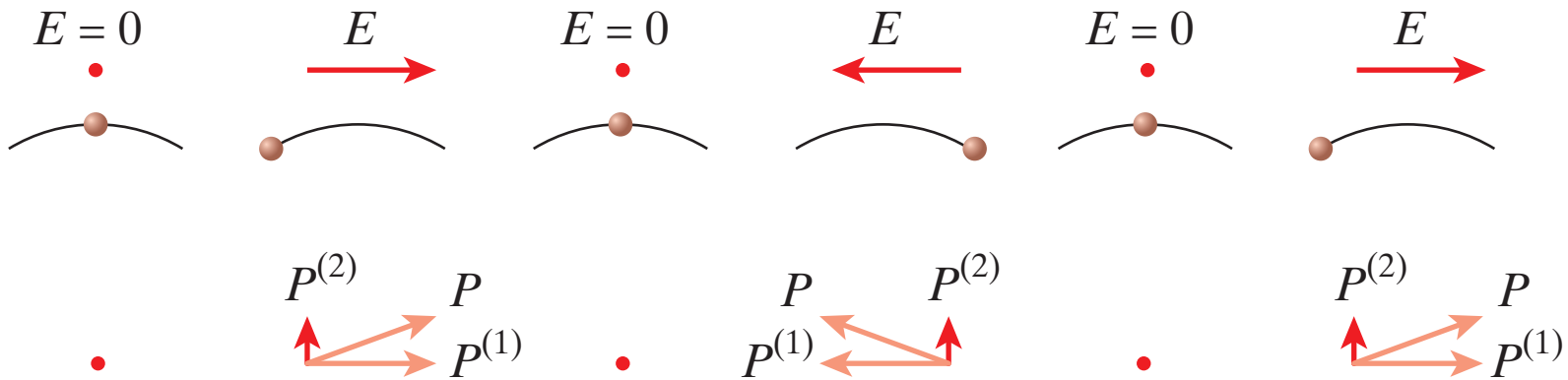
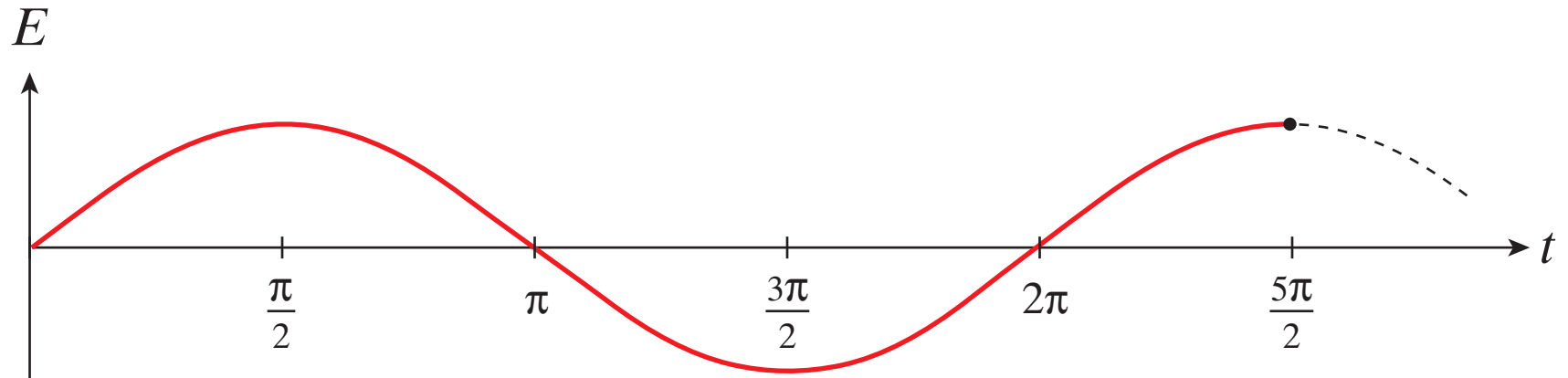
Nonlinear optics



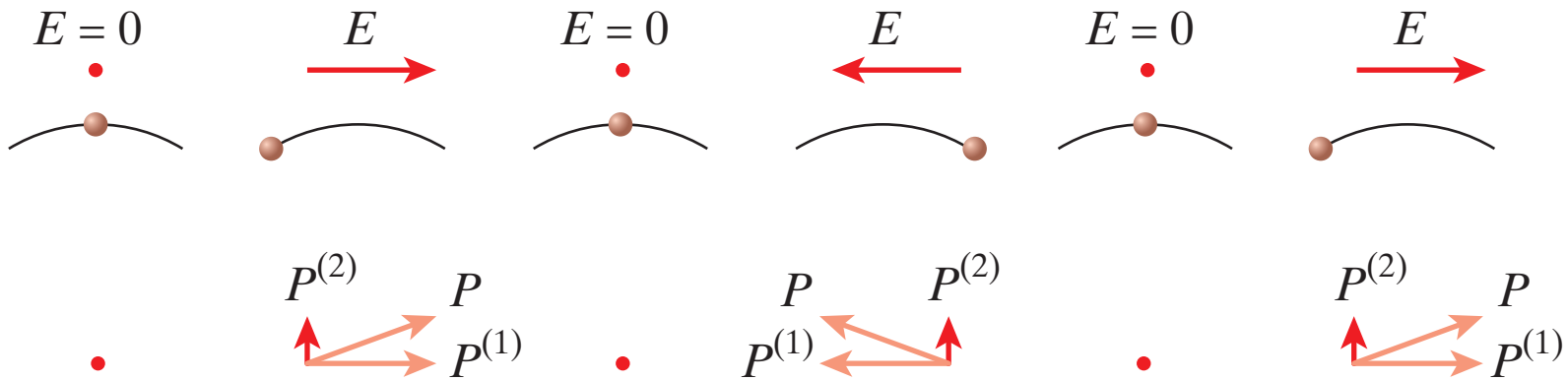
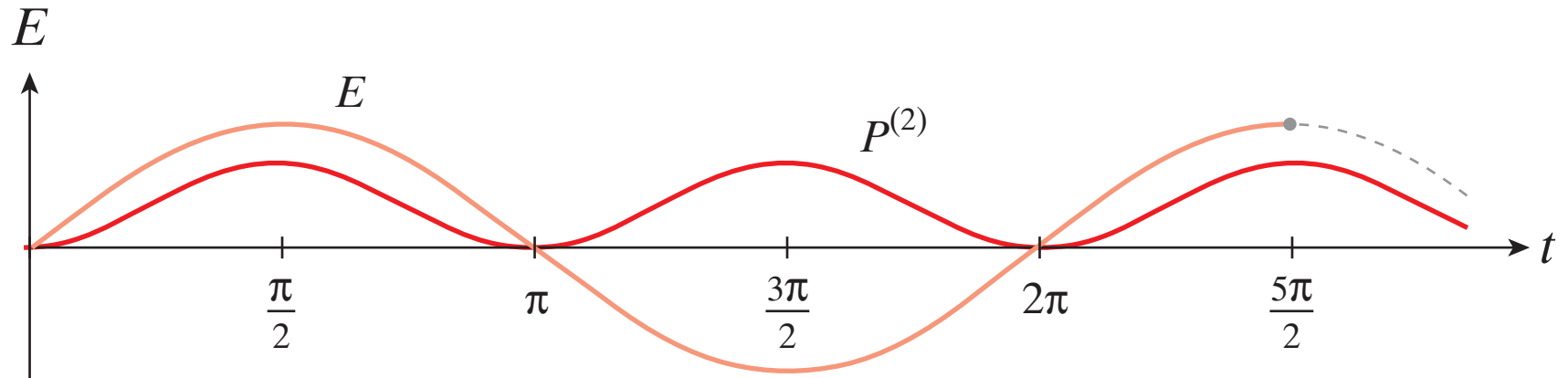
Nonlinear optics



Nonlinear optics



Nonlinear optics



Nonlinear optics

In medium with inversion symmetry

$$\vec{P}^{(2)} = \vec{\chi}^{(2)} : \vec{E} \vec{E} \quad \Rightarrow \quad -\vec{P}^{(2)} = \vec{\chi}^{(2)} : (-\vec{E})(-\vec{E})$$

Nonlinear optics

In medium with inversion symmetry

$$\vec{P}^{(2)} = \vec{\chi}^{(2)} : \vec{E} \vec{E} \quad \Rightarrow \quad -\vec{P}^{(2)} = \vec{\chi}^{(2)} : (-\vec{E})(-\vec{E})$$

and so

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

Nonlinear optics

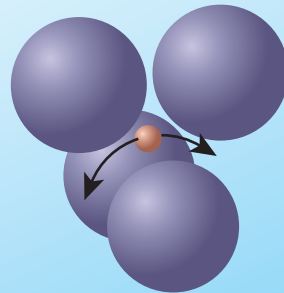
In medium with inversion symmetry

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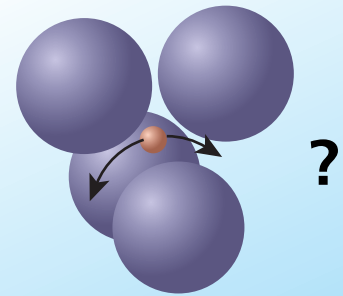
$$\chi^{(2)} = -\chi^{(2)} = 0$$

... but ...

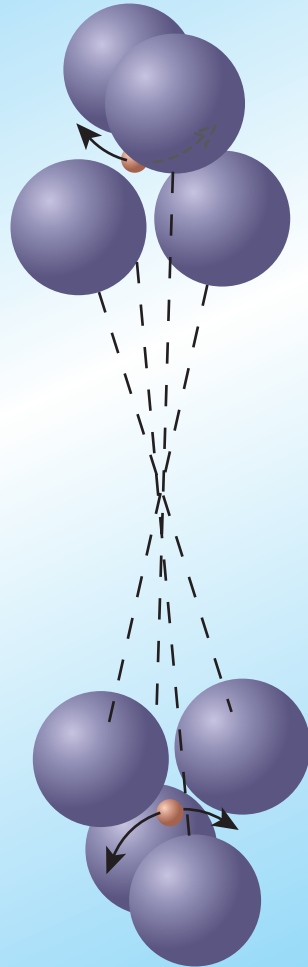


Nonlinear optics

How to reconcile $\chi^{(2)} = -\chi^{(2)} = 0$ with



Nonlinear optics



Nonlinear optics

Nonlinear polarization:

$$P = \chi^{(1)}E + \chi^{(3)}E^3 + \dots$$

Nonlinear optics

Nonlinear polarization:

$$P = \chi^{(1)}E + \chi^{(3)}E^3 + \dots$$

Third order polarization

$$P^{(3)}(t) = \chi^{(3)}E(t)E^*(t)E(t) = \chi^{(3)}I(t)E(t)$$

Nonlinear optics

Nonlinear polarization:

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Nonlinear optics

Nonlinear polarization:

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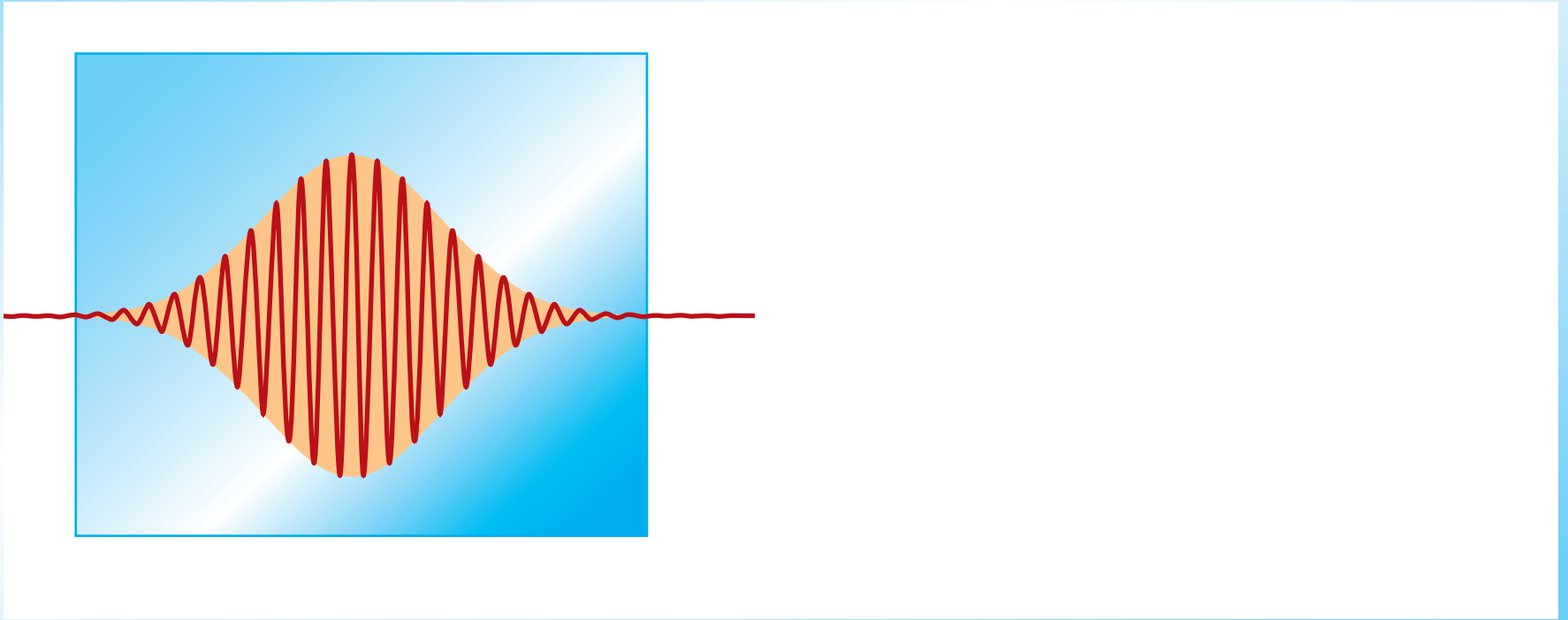
and so $P = P^{(1)} + P^{(3)} = (\chi^{(1)} + \chi^{(3)}I)E \equiv \chi_{eff}E$

$$n = \sqrt{\epsilon} = \sqrt{1 + \chi_{eff}} \approx \sqrt{1 + \chi^{(1)}} + \frac{1}{2} \frac{\chi^{(3)}I}{\sqrt{1 + \chi^{(1)}}} = n_o + n_2I$$

Nonlinear optics

Intensity dependent index of refraction:

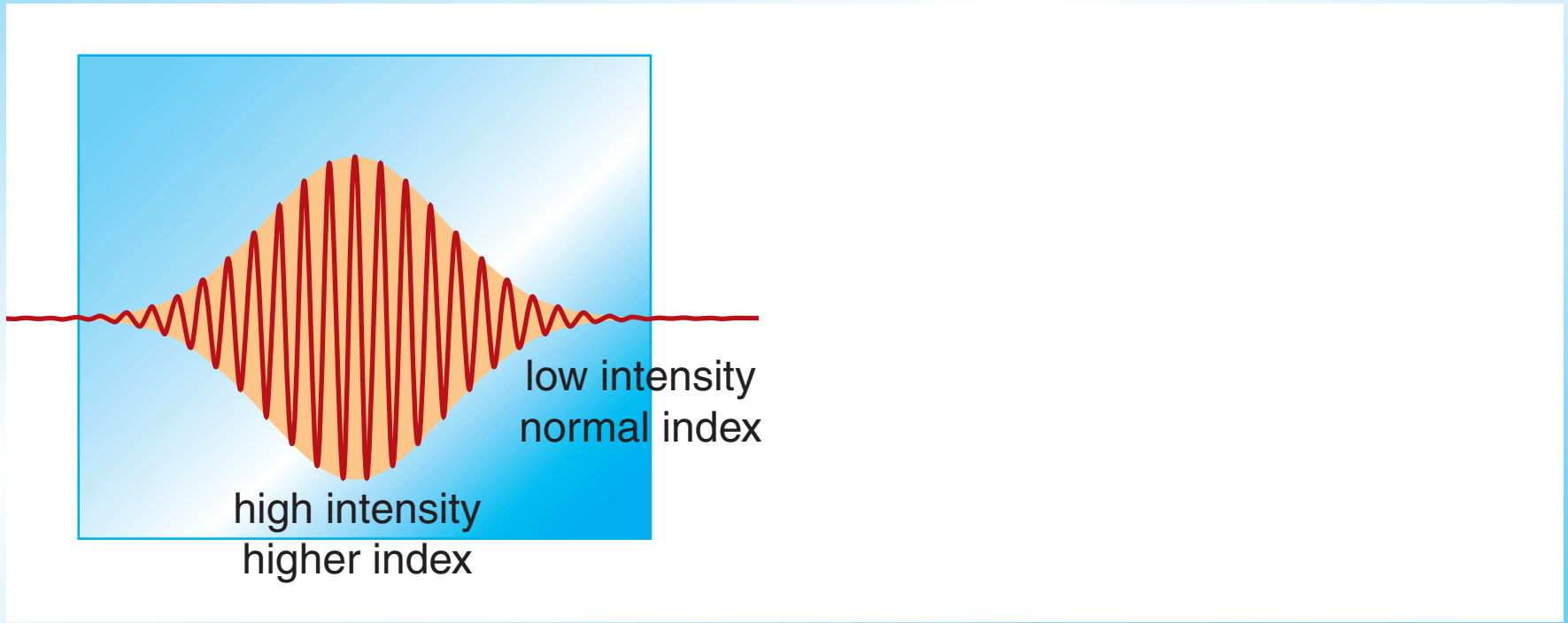
$$n = n_o + n_2 I$$



Nonlinear optics

Intensity dependent index of refraction:

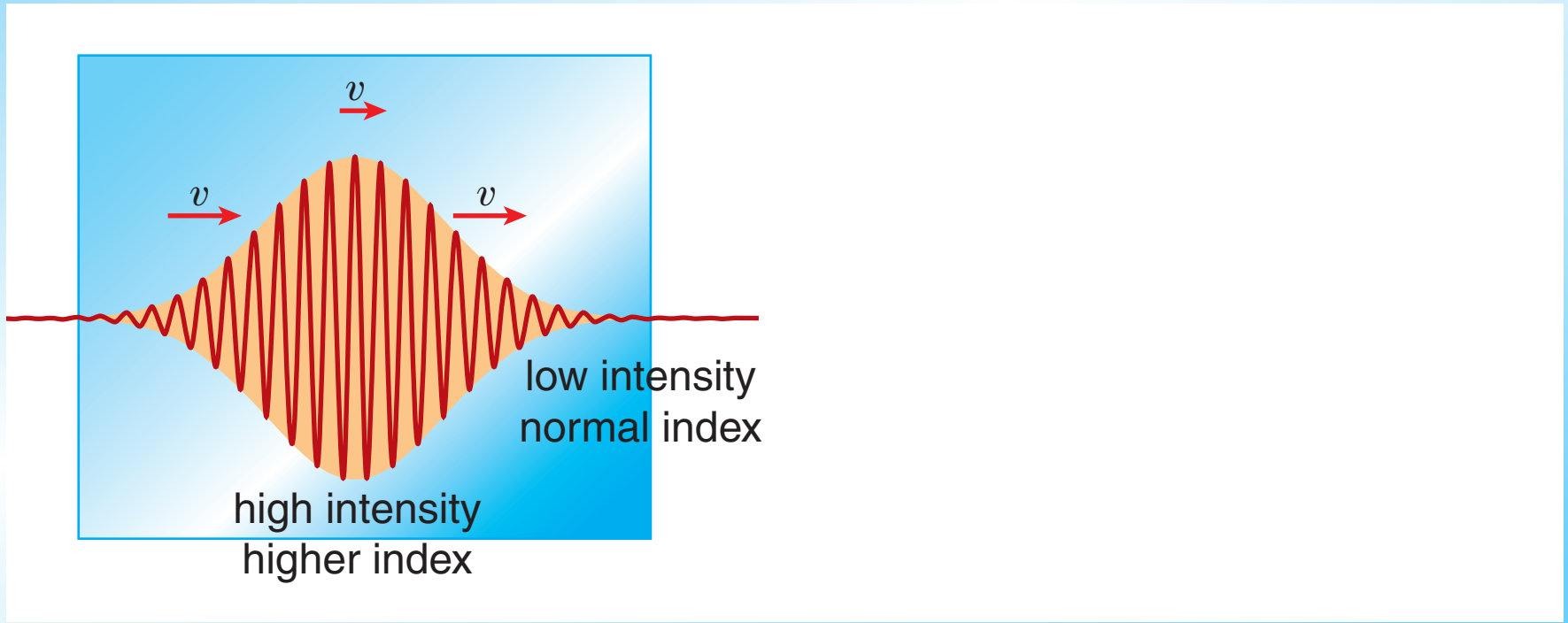
$$n = n_o + n_2 I$$



Nonlinear optics

Intensity dependent index of refraction:

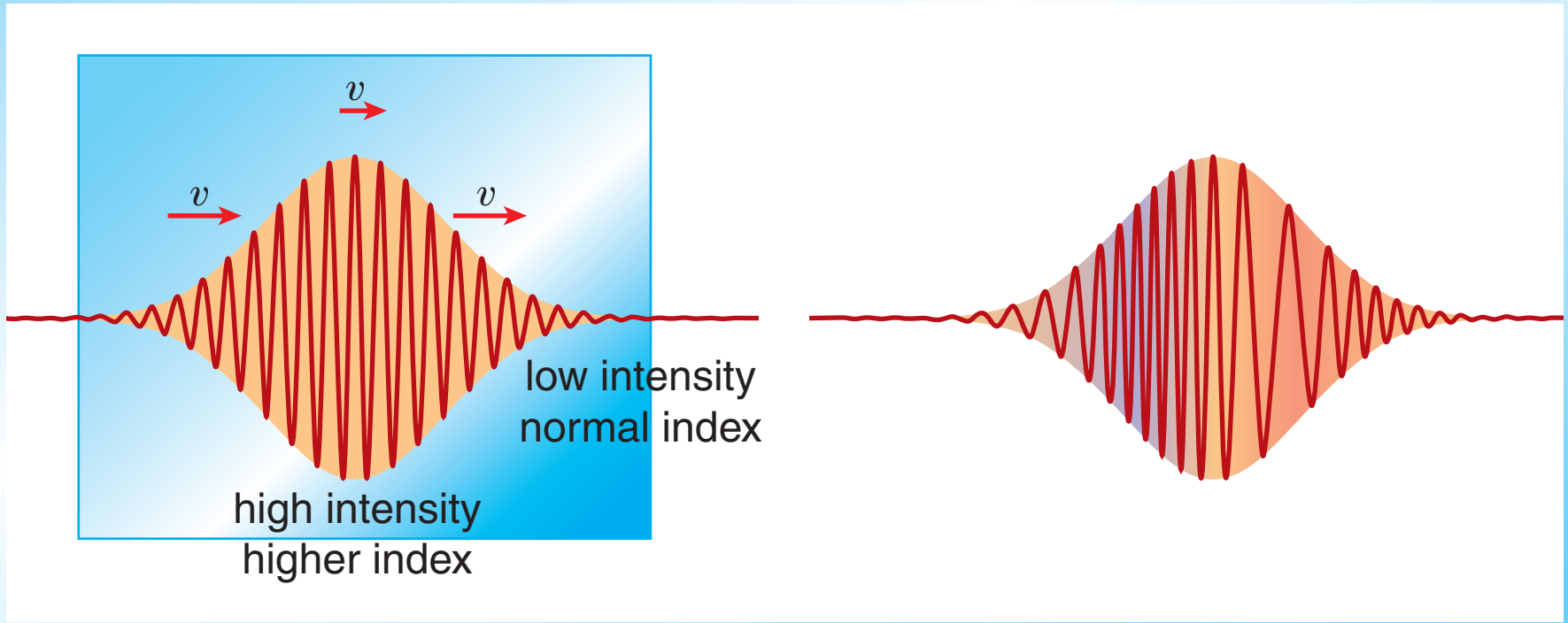
$$n = n_o + n_2 I$$



Nonlinear optics

Intensity dependent index of refraction:

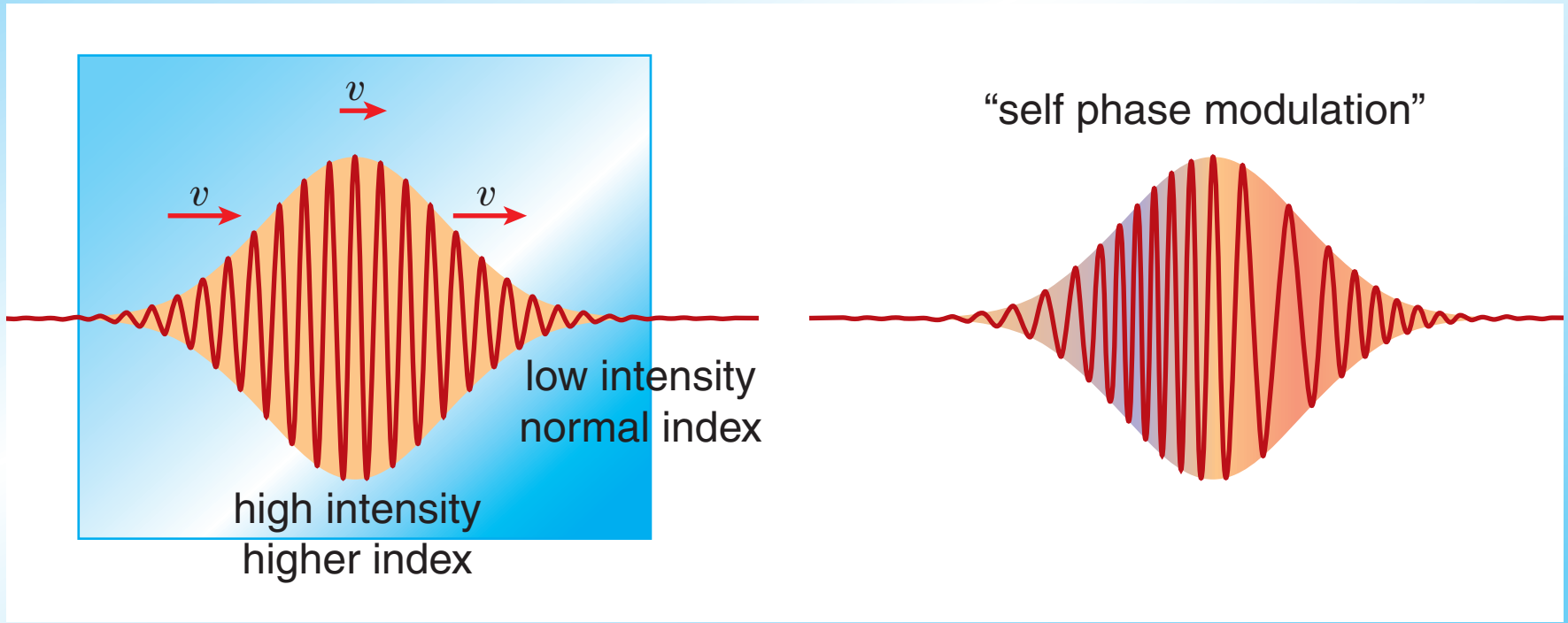
$$n = n_o + n_2 I$$



Nonlinear optics

Intensity dependent index of refraction:

$$n = n_o + n_2 I$$



Nonlinear optics

Phase:

$$\frac{\phi}{2\pi} = \frac{nL}{\lambda}$$

Nonlinear optics

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

Nonlinear optics

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Frequency change:

$$\Delta\omega = - \frac{d\phi}{dt} = \frac{-2\pi}{\lambda} L n_2 \frac{dI}{dt}$$

Nonlinear optics

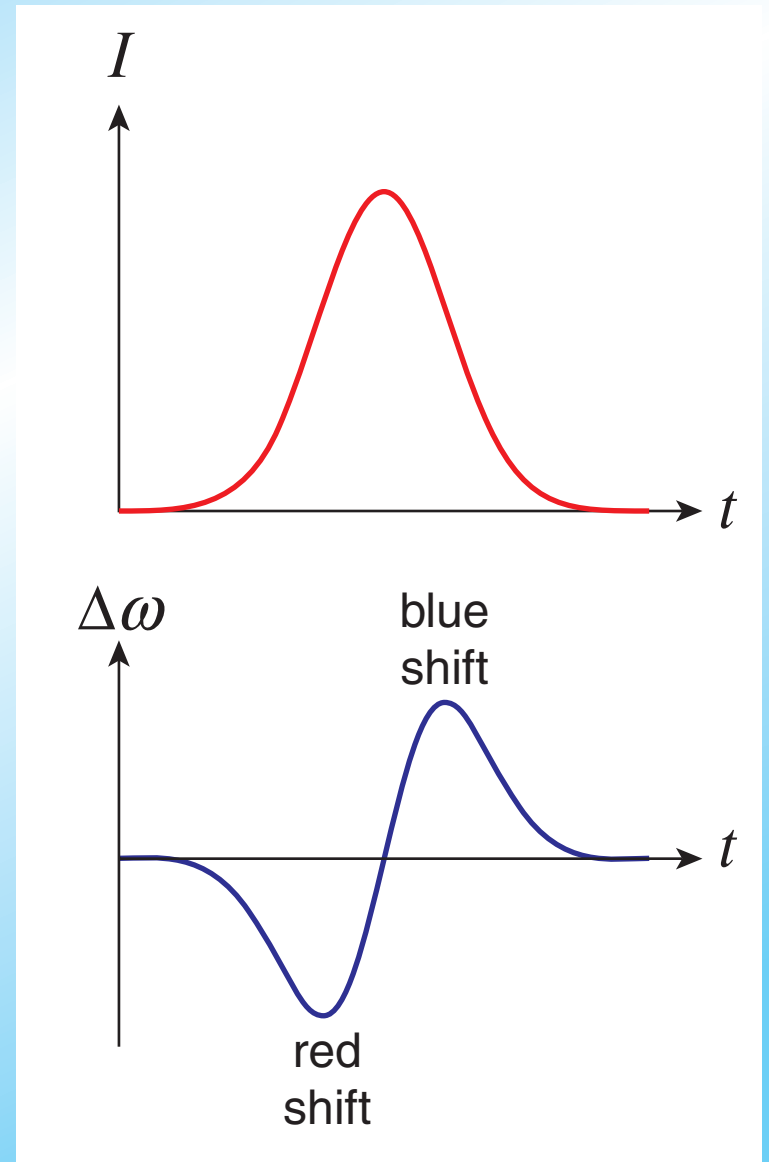
Phase:

$$\frac{\phi}{2\pi} = \frac{nL}{\lambda}$$

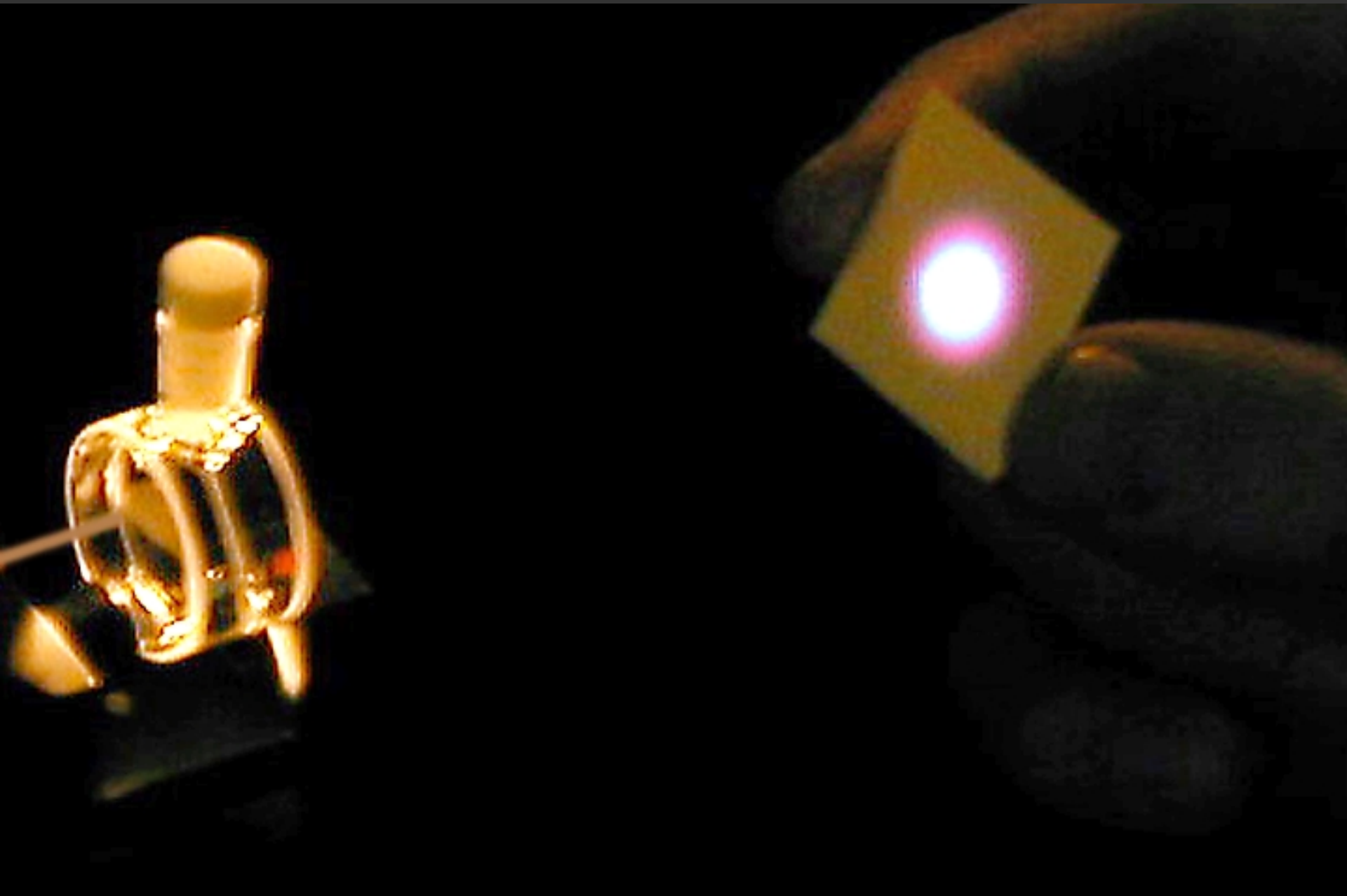
$$\phi = \frac{2\pi}{\lambda} L(n_o + n_2 I)$$

Frequency change:

$$\Delta\omega = -\frac{d\phi}{dt} = \frac{-2\pi}{\lambda} L n_2 \frac{dI}{dt}$$

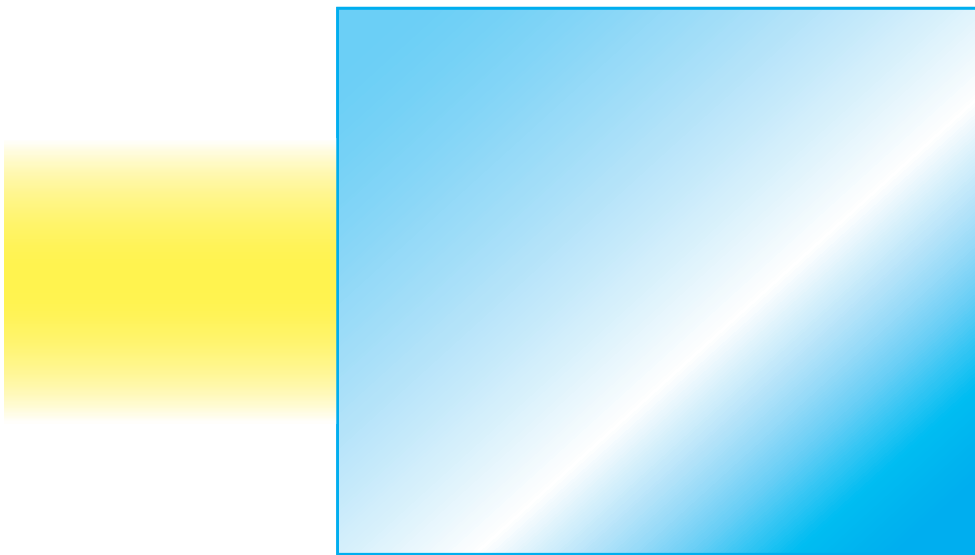


Nonlinear optics



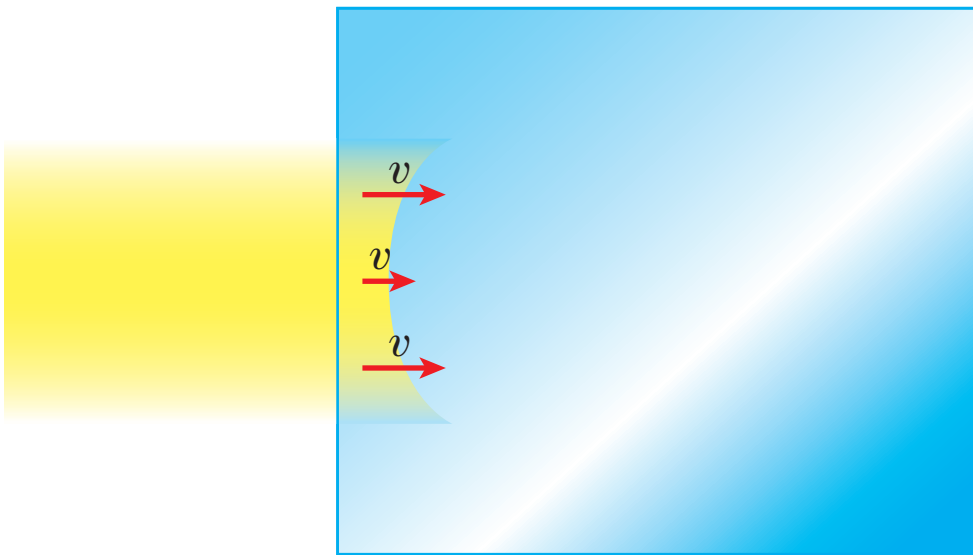
Nonlinear optics

Spatial intensity profile...



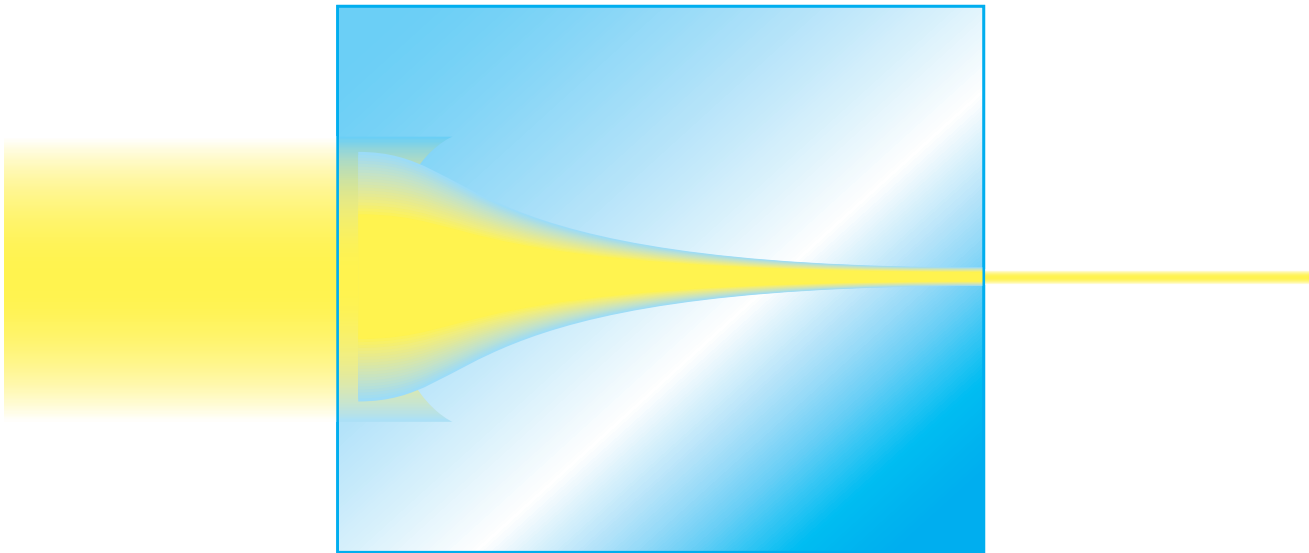
Nonlinear optics

Spatial intensity profile...



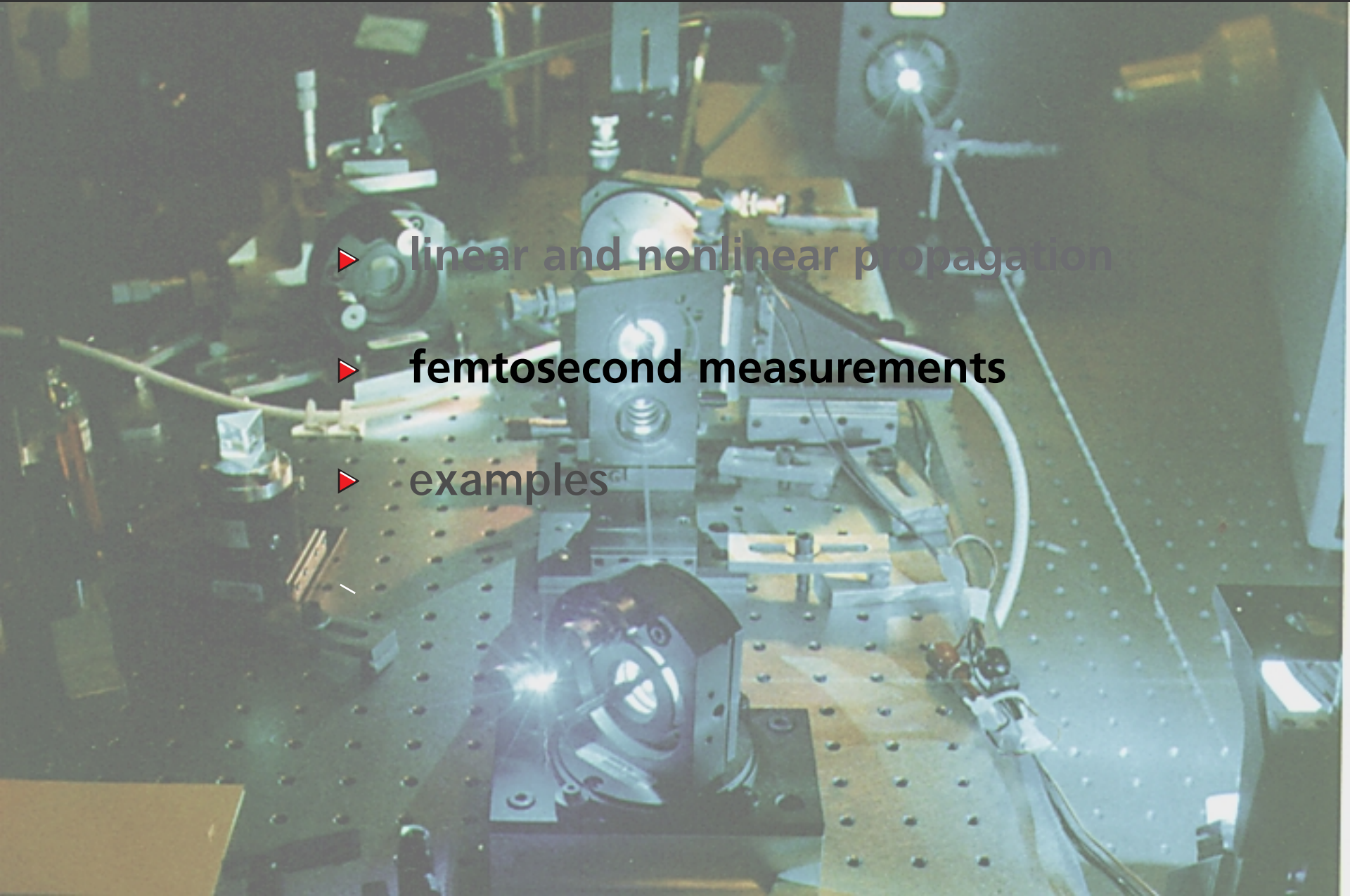
Nonlinear optics

...causes self-focusing



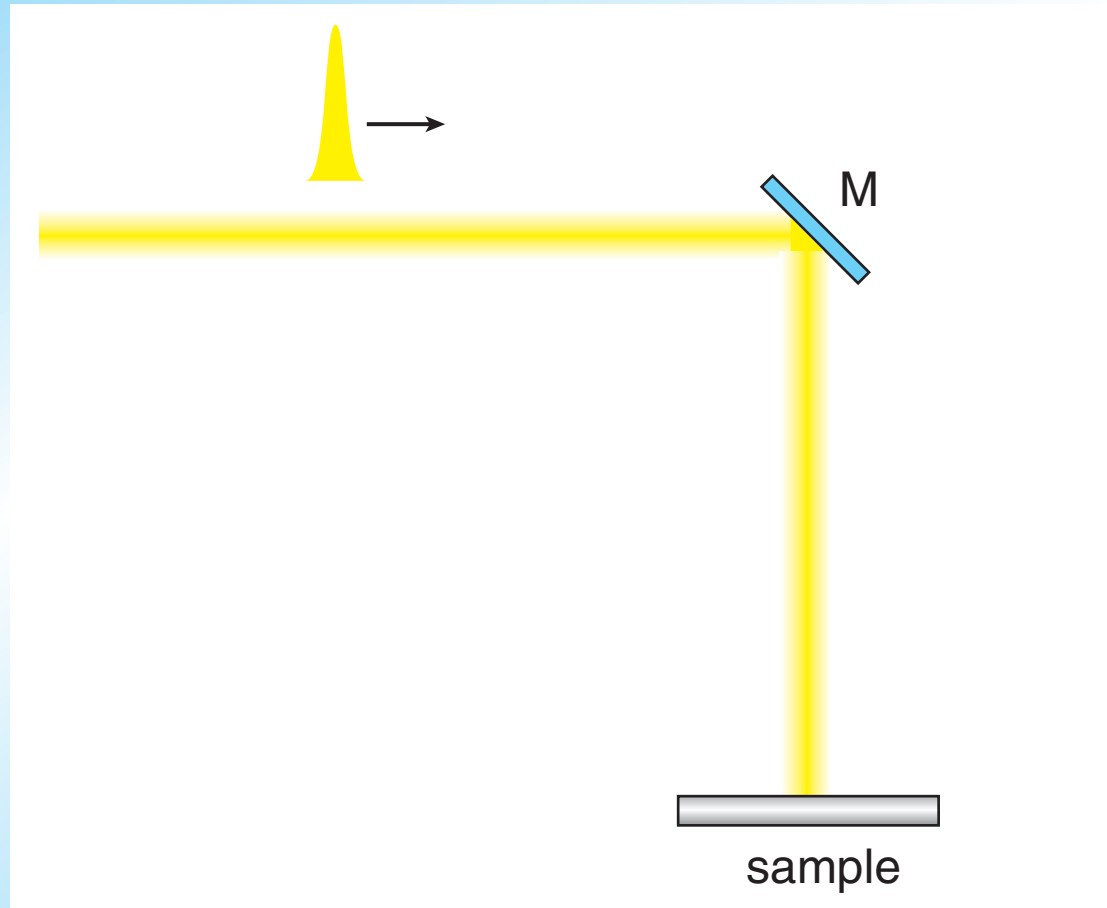
Outline

- ▶ linear and nonlinear propagation
- ▶ femtosecond measurements
- ▶ examples



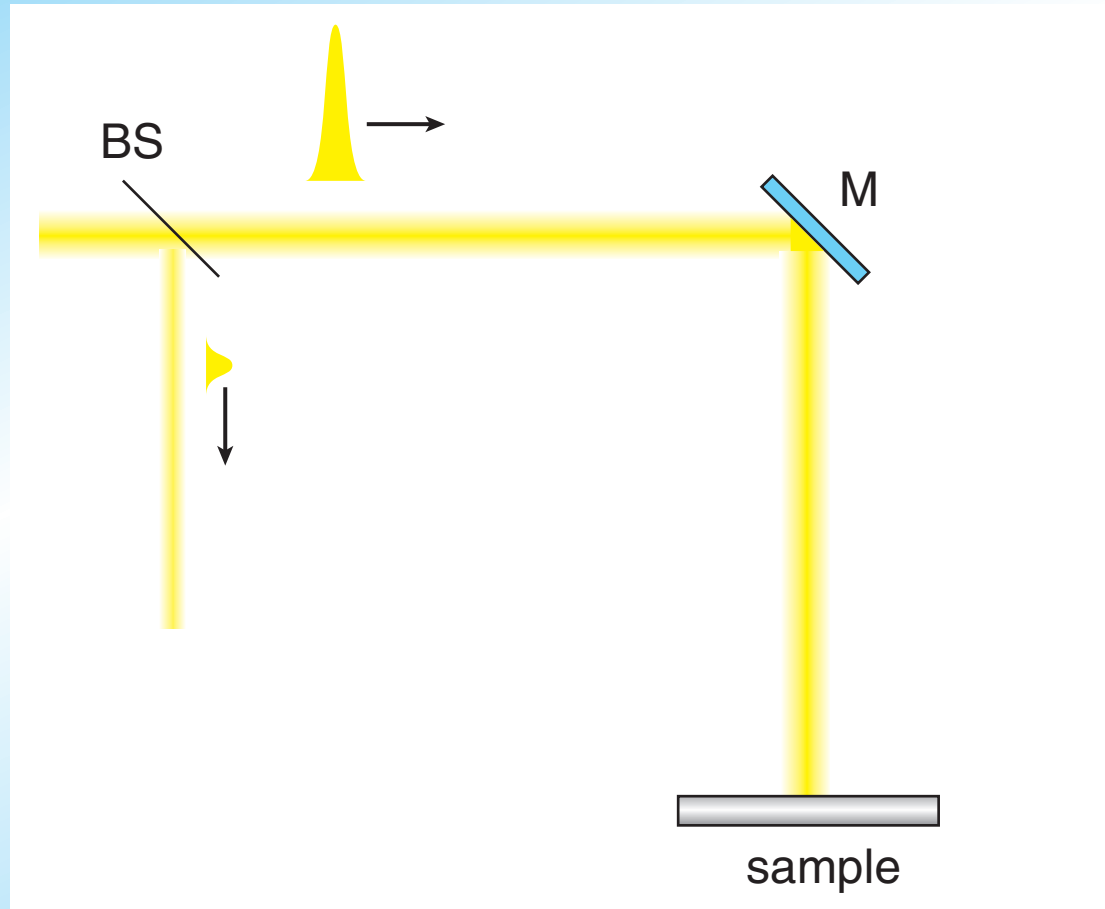
Introduction

How to measure on the femtosecond time scale?



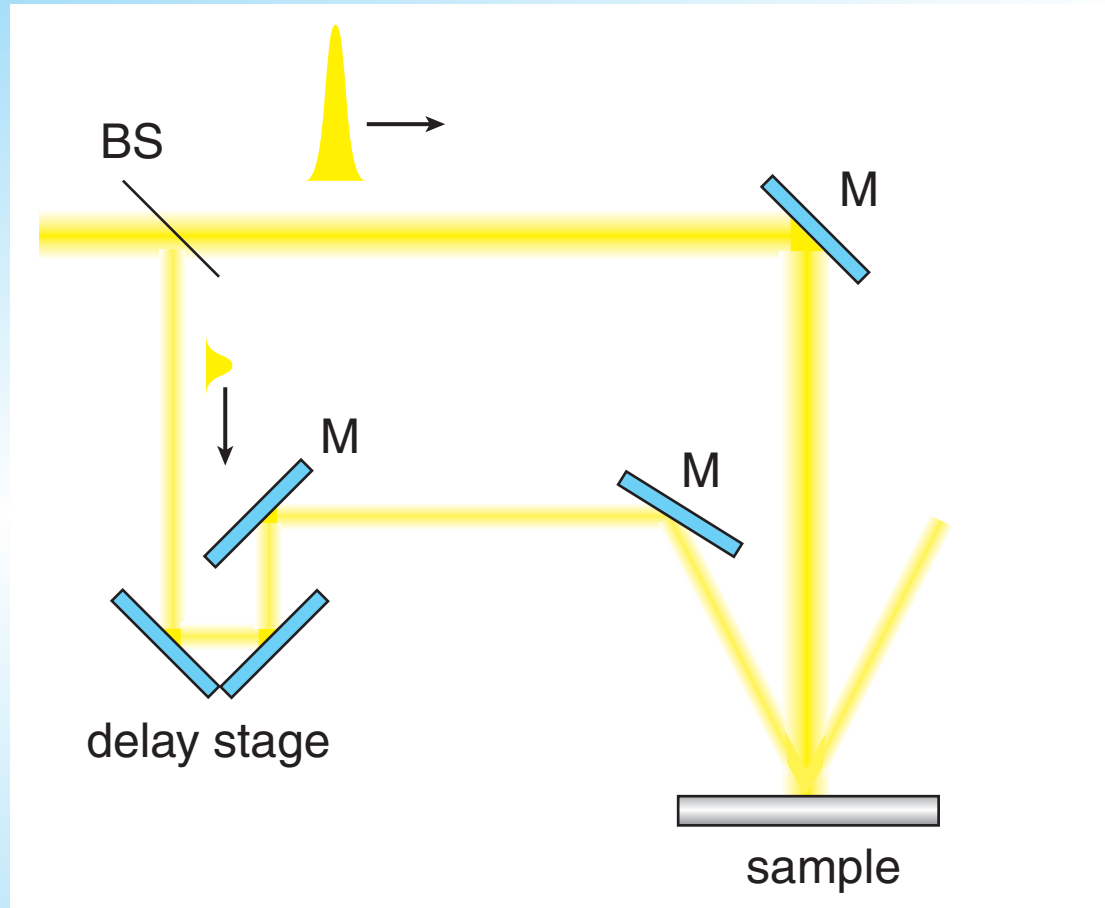
Introduction

Use pump-probe technique



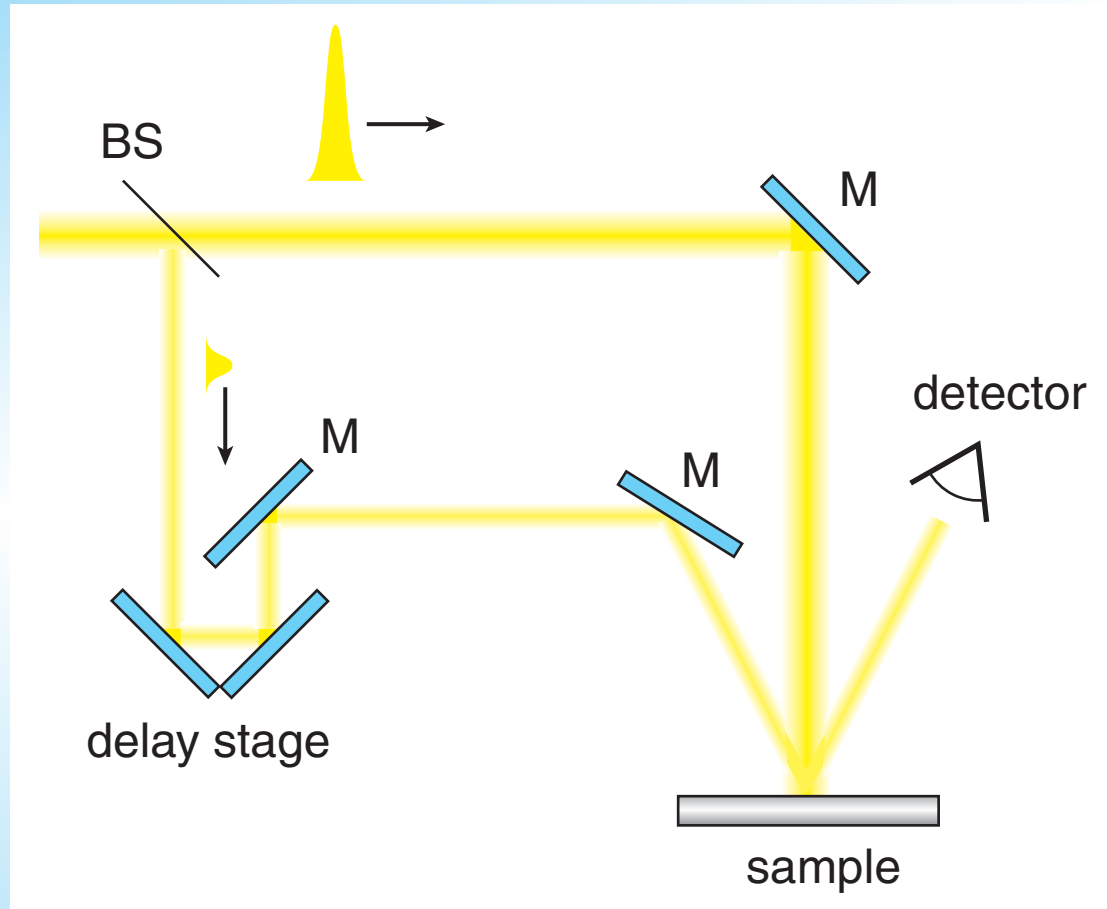
Introduction

Use pump-probe technique



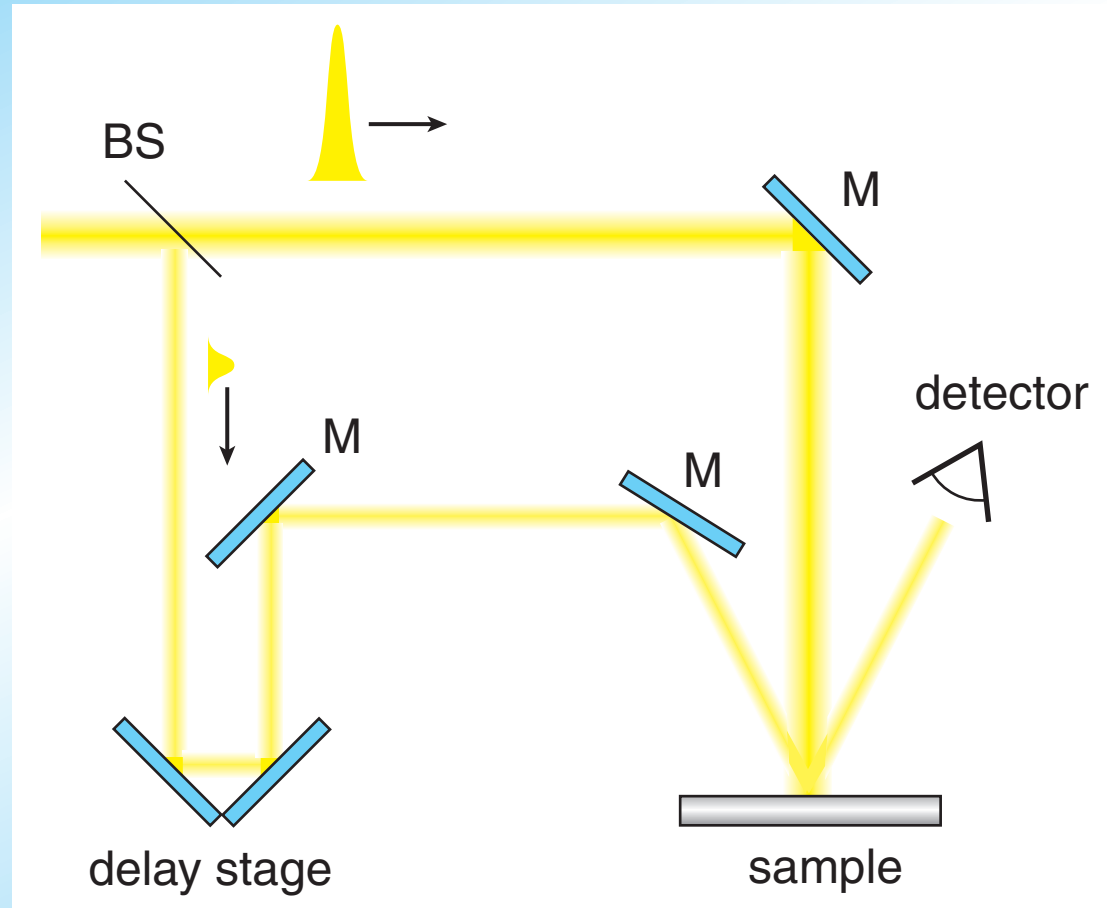
Introduction

Use pump-probe technique



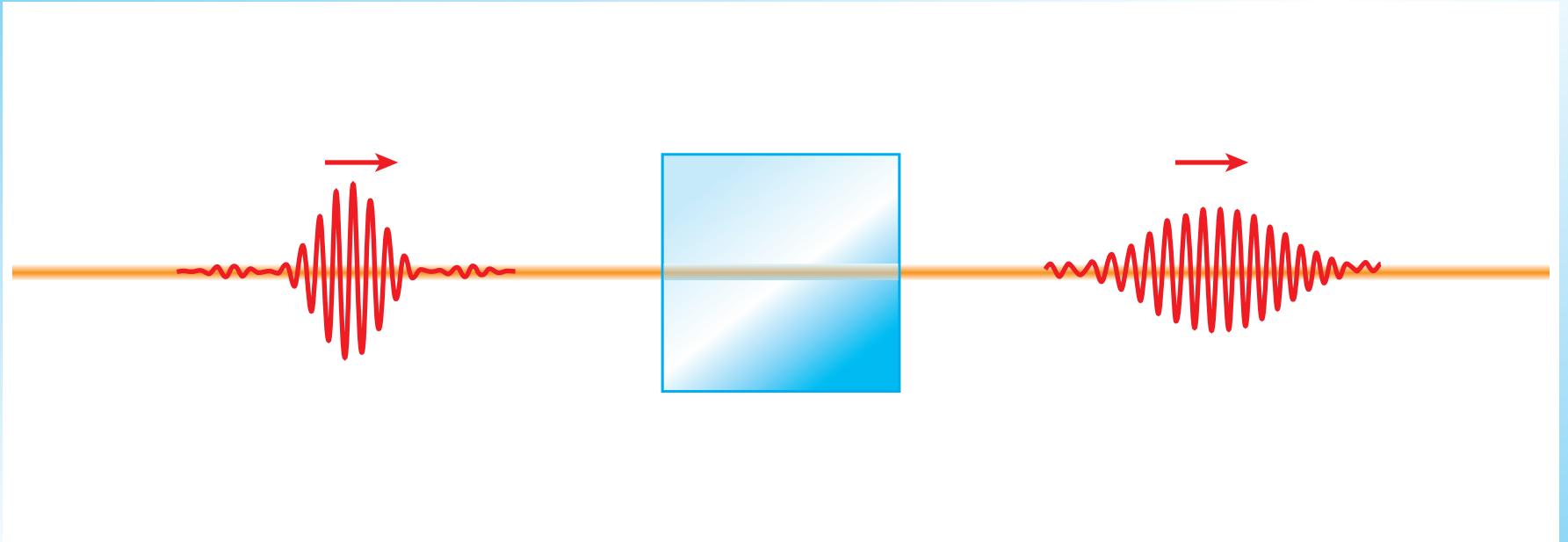
Introduction

Vary delay to get time resolution



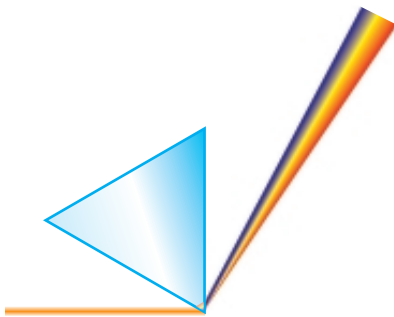
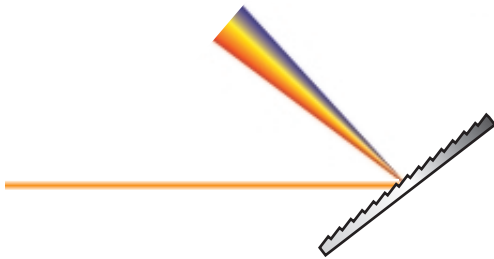
Dispersion compensation

Dispersion stretches the pulse

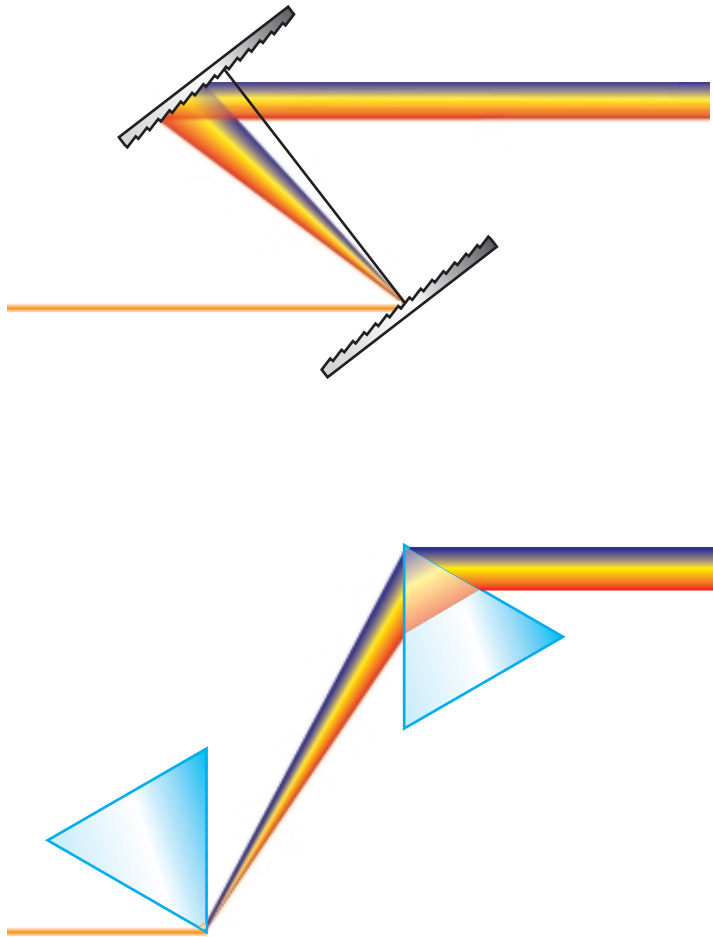


Compensate by rearranging spectral components!

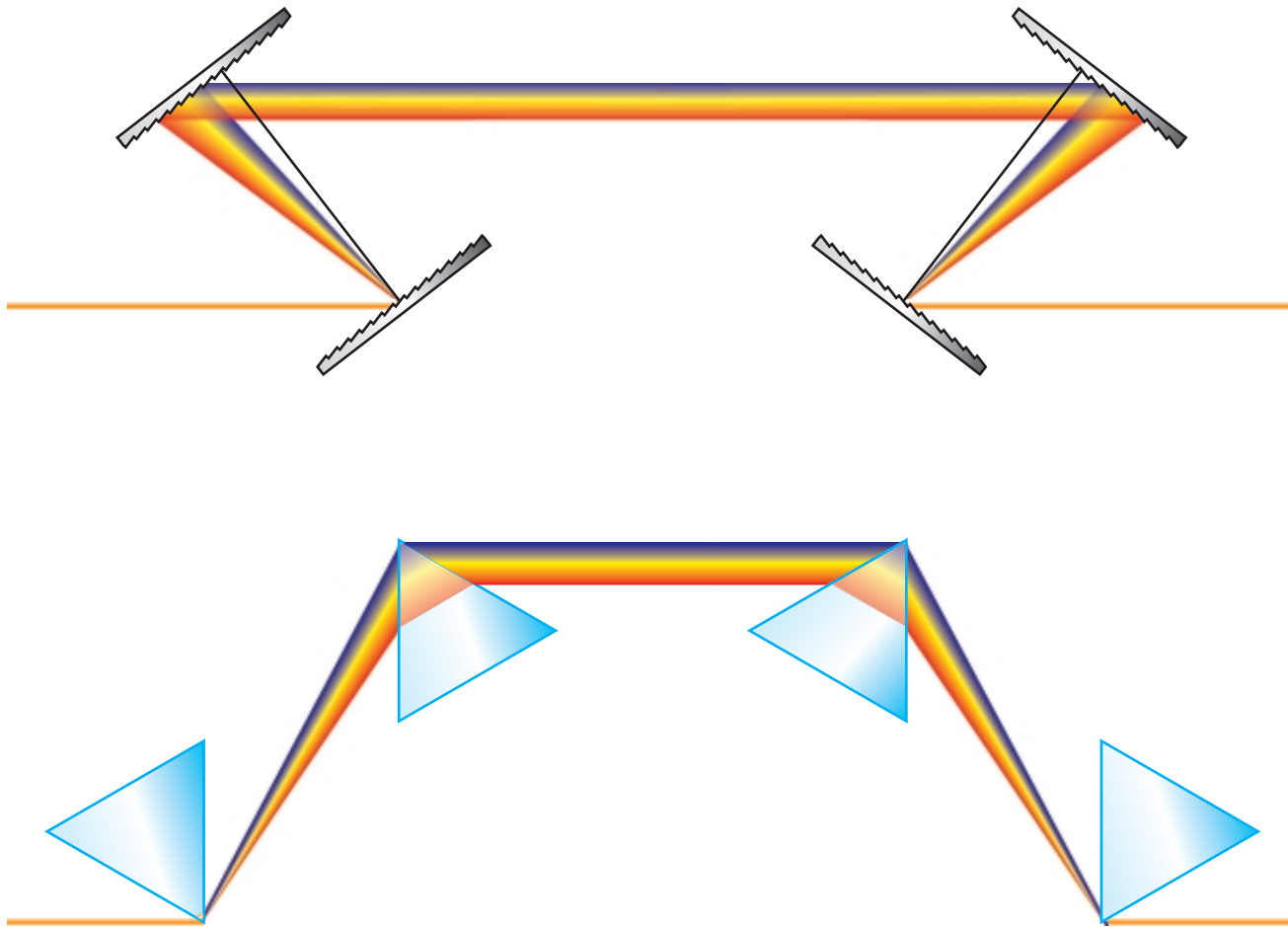
Dispersion compensation



Dispersion compensation



Dispersion compensation

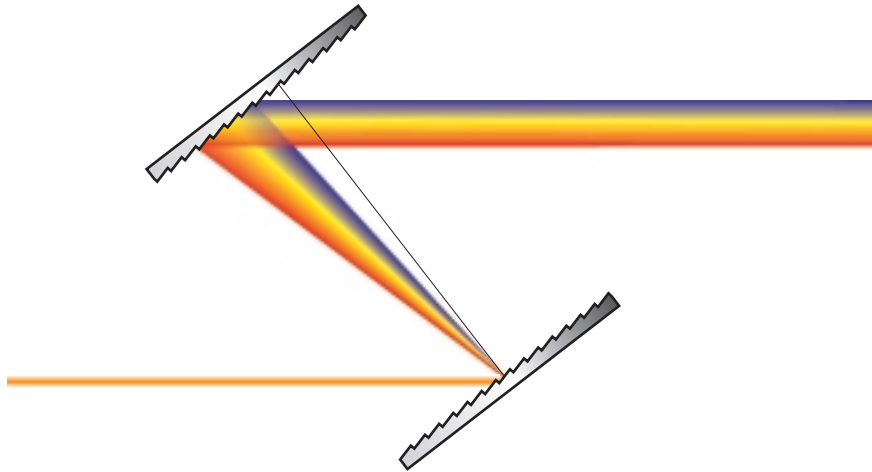


Dispersion compensation

How do these arrangements work?

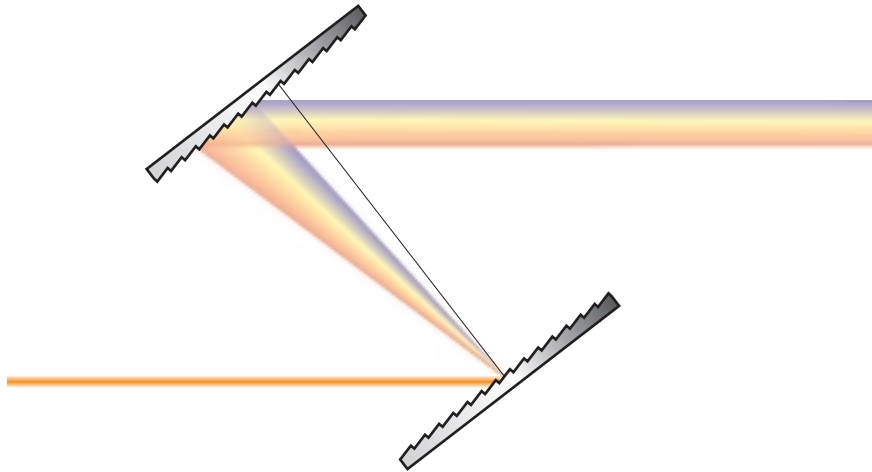
Dispersion compensation

Does path length difference compensate?



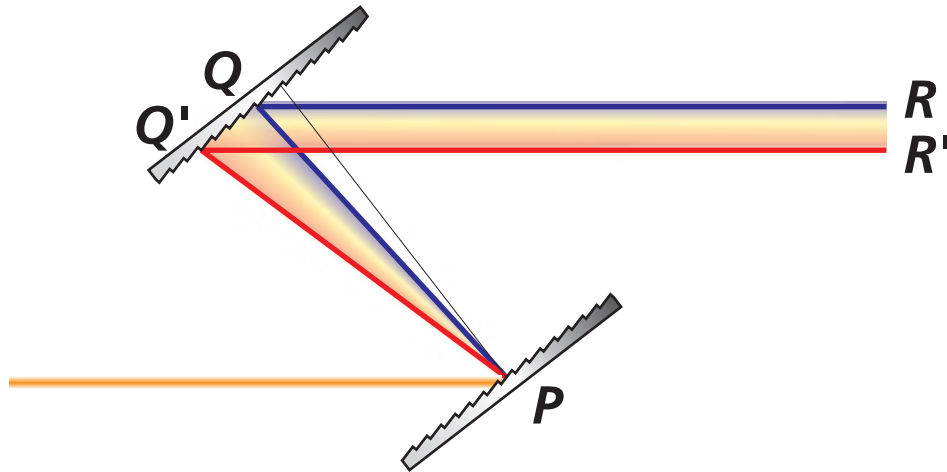
Dispersion compensation

Does path length difference compensate?



Dispersion compensation

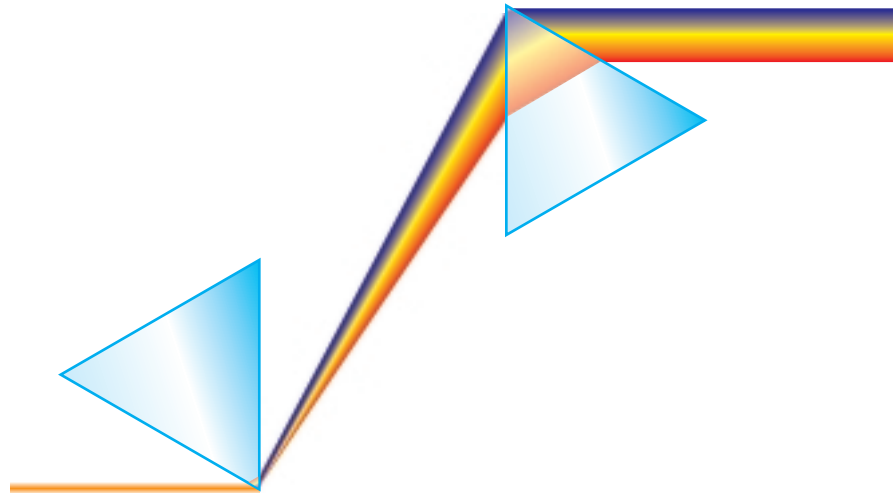
Does path length difference compensate?



Grating gives low frequency longer path length...

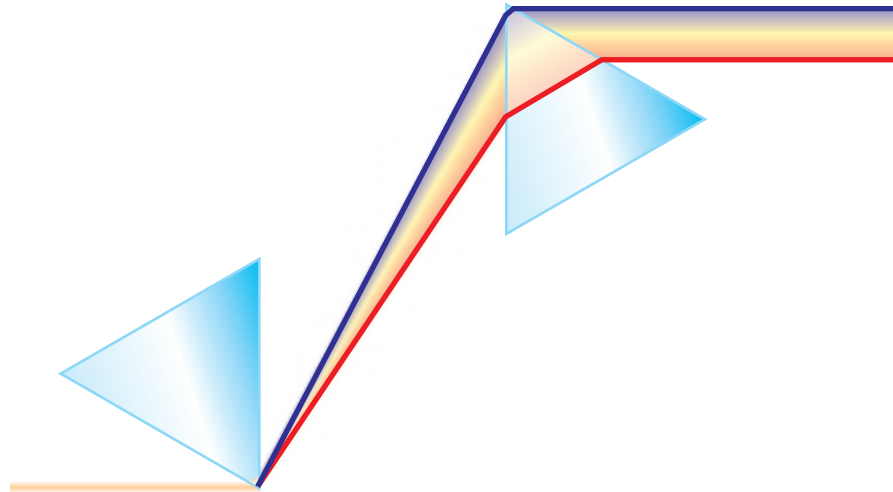
Dispersion compensation

Does path length difference compensate?



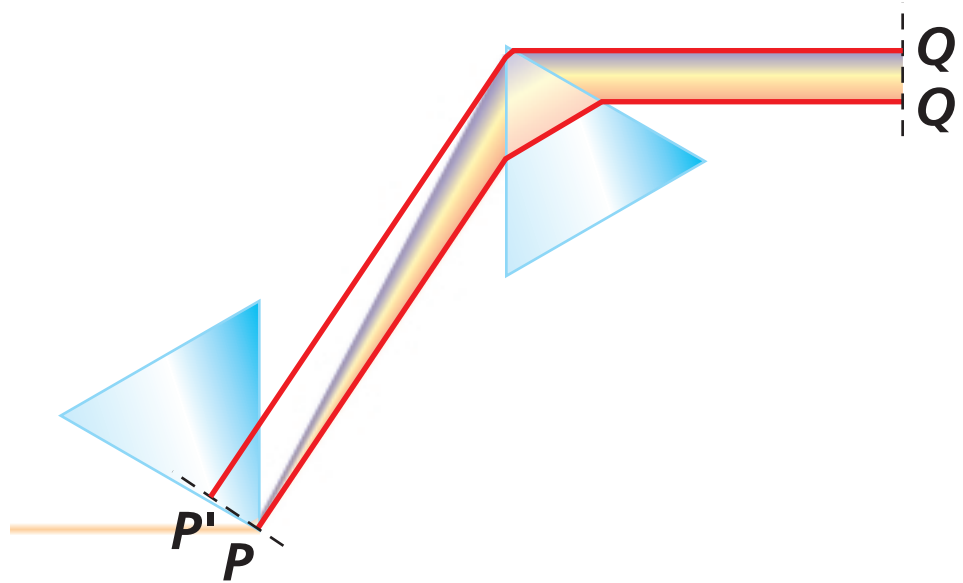
Dispersion compensation

Does path length difference compensate?



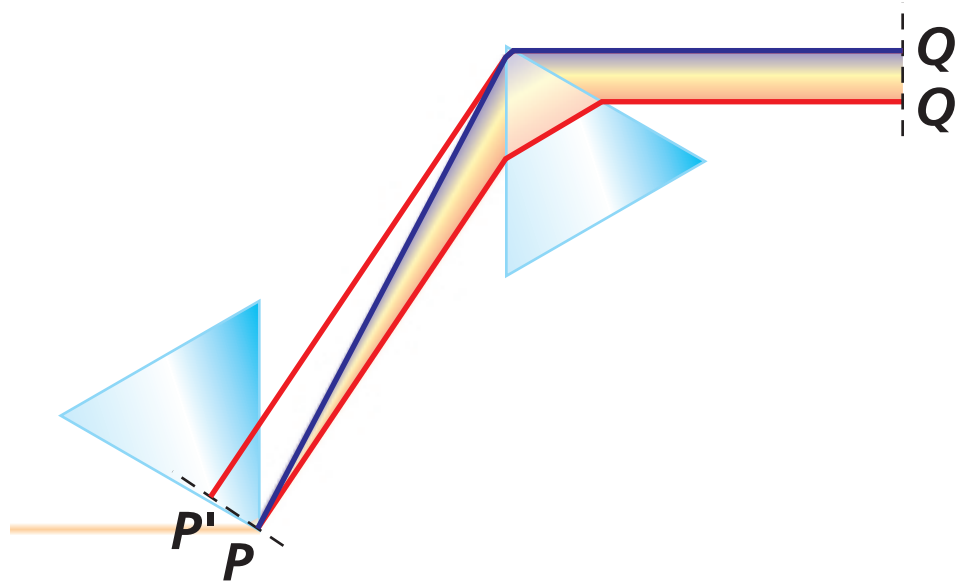
Dispersion compensation

Does path length difference compensate?



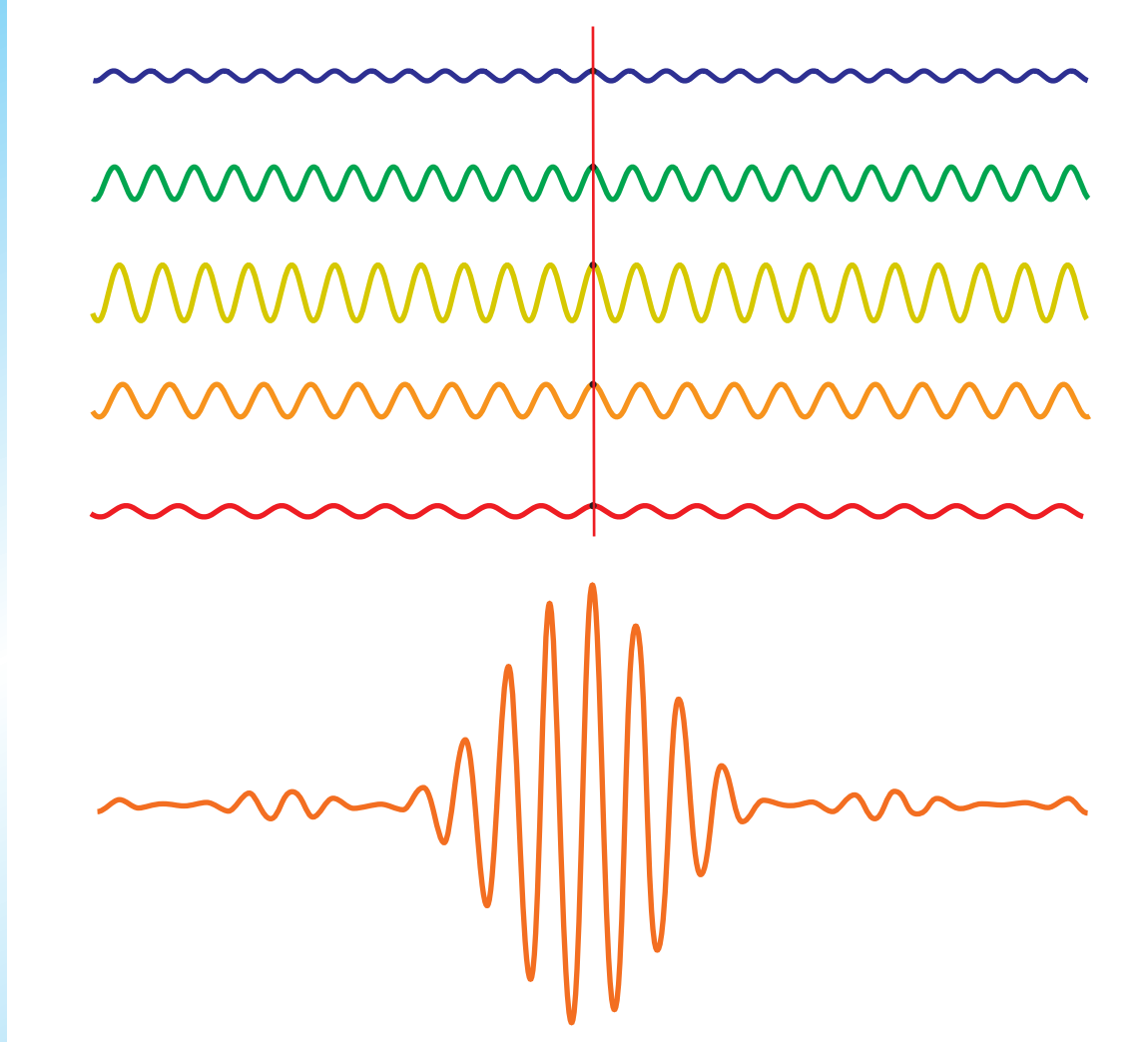
Dispersion compensation

Does path length difference compensate?

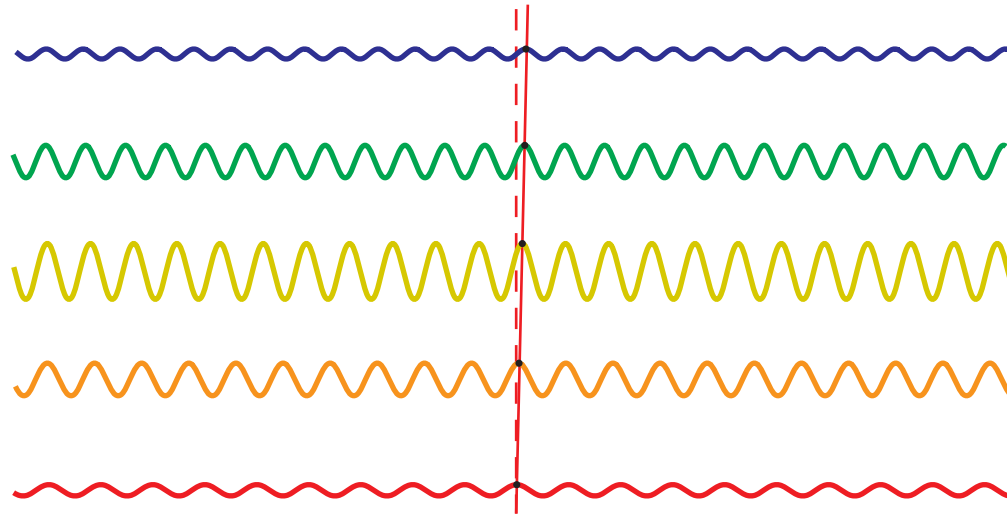


...so prism gives low frequency *shorter* path length...

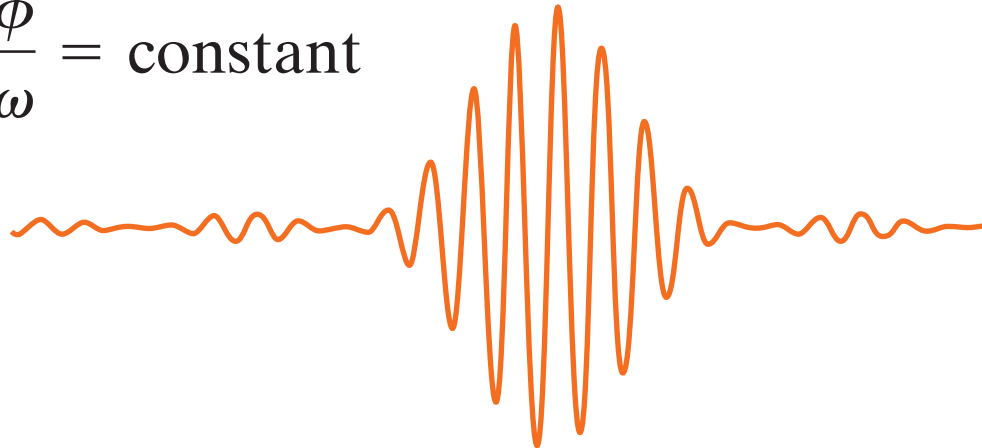
Dispersion compensation



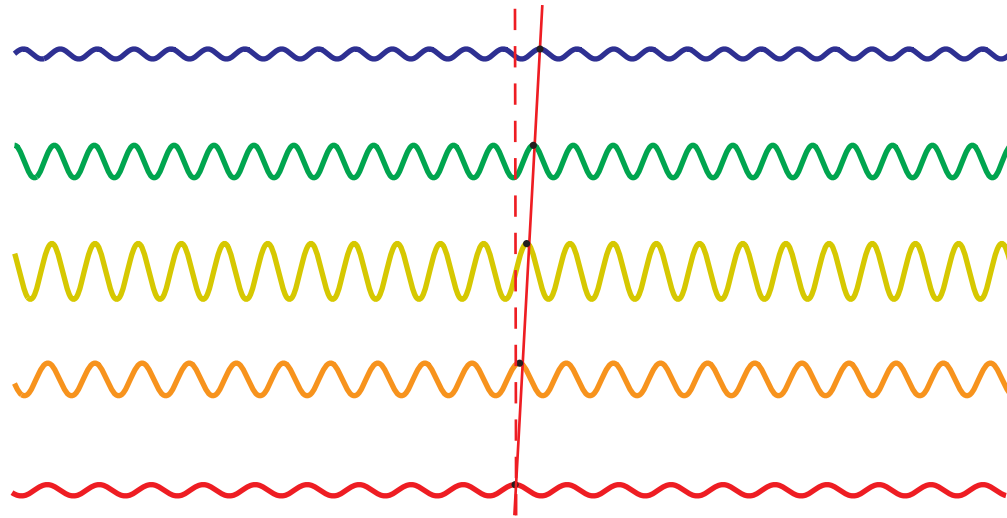
Dispersion compensation



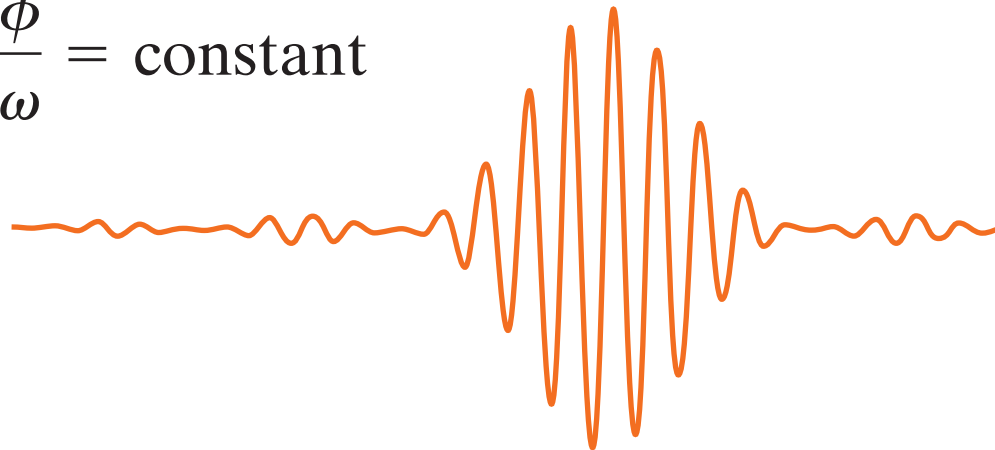
$$\frac{d\phi}{d\omega} = \text{constant}$$



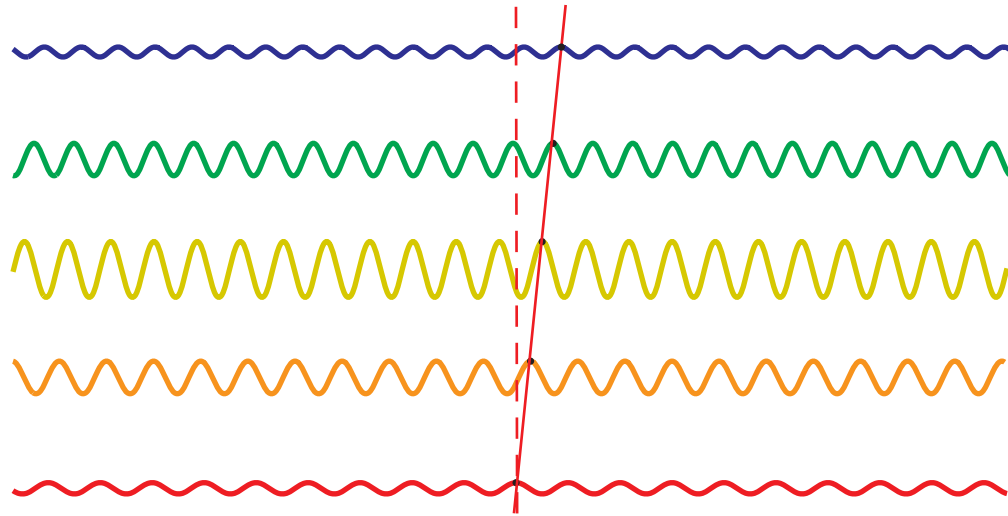
Dispersion compensation



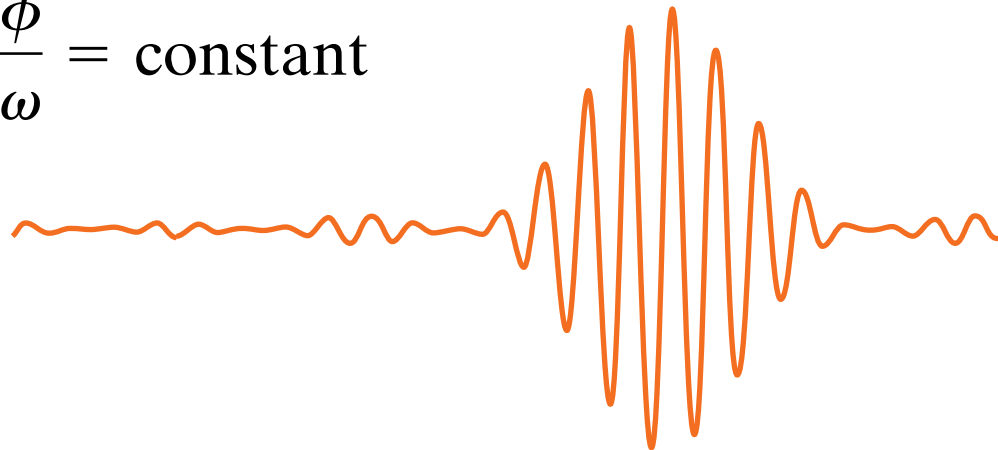
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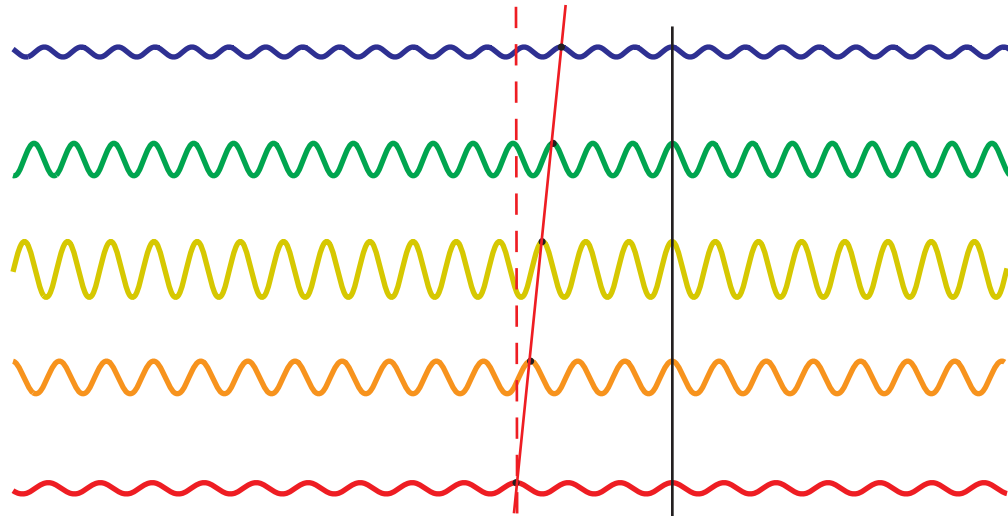
Dispersion compensation



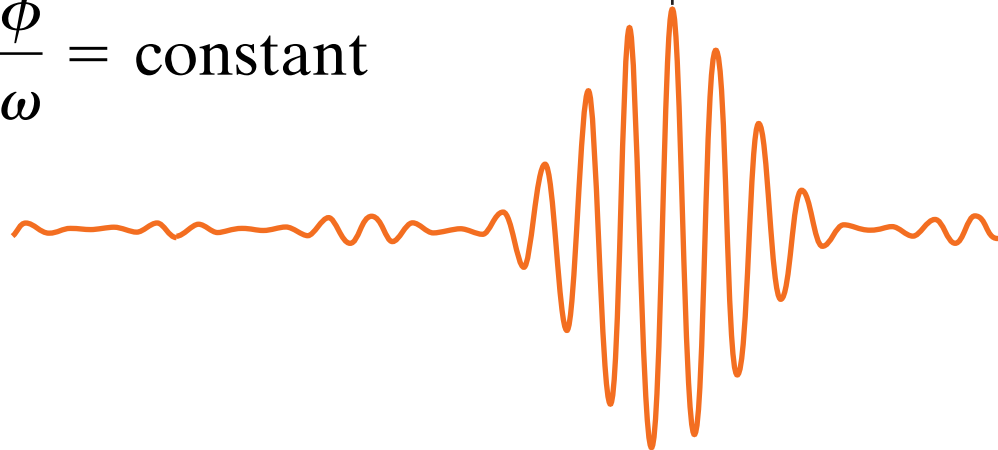
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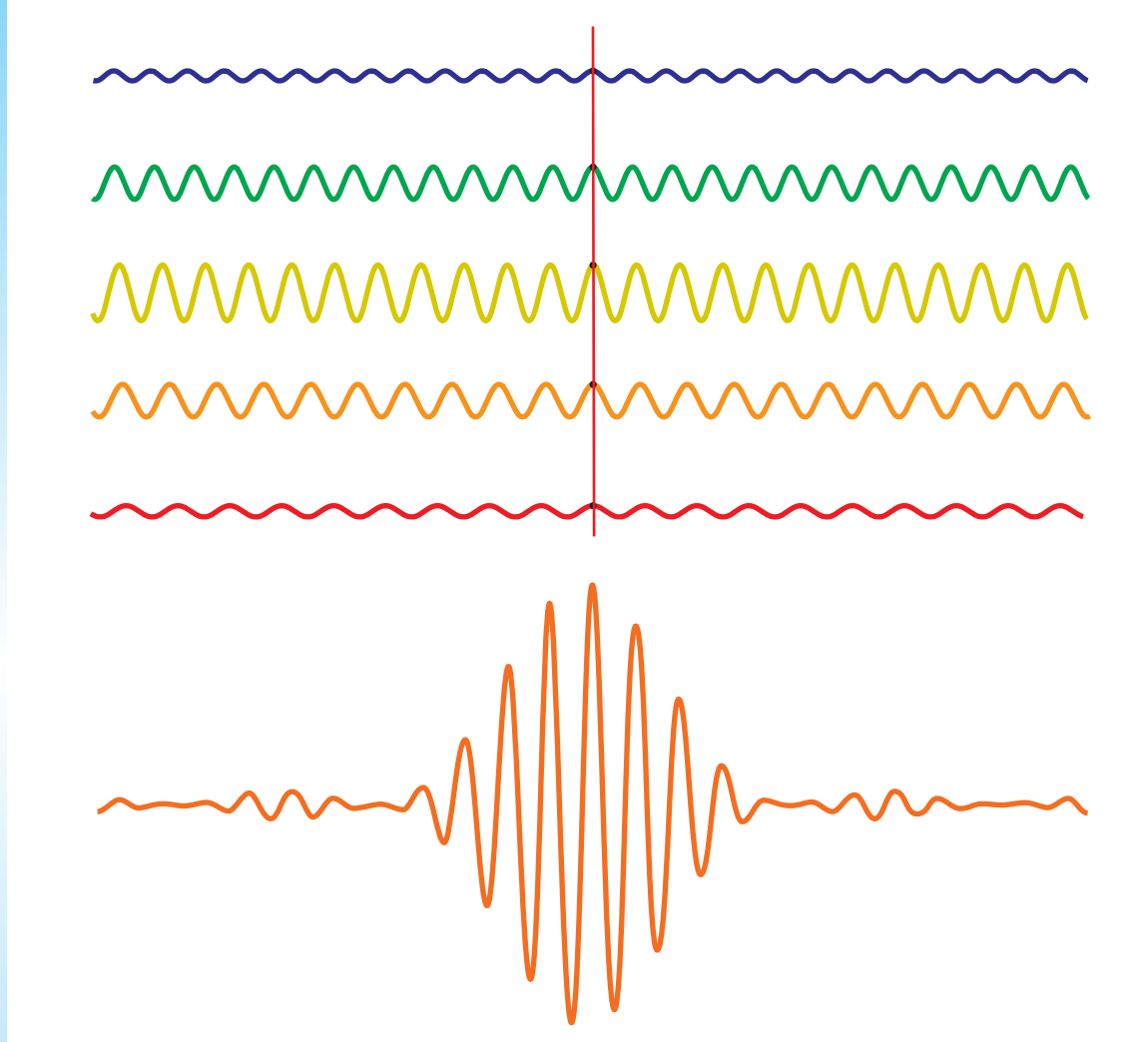
Dispersion compensation



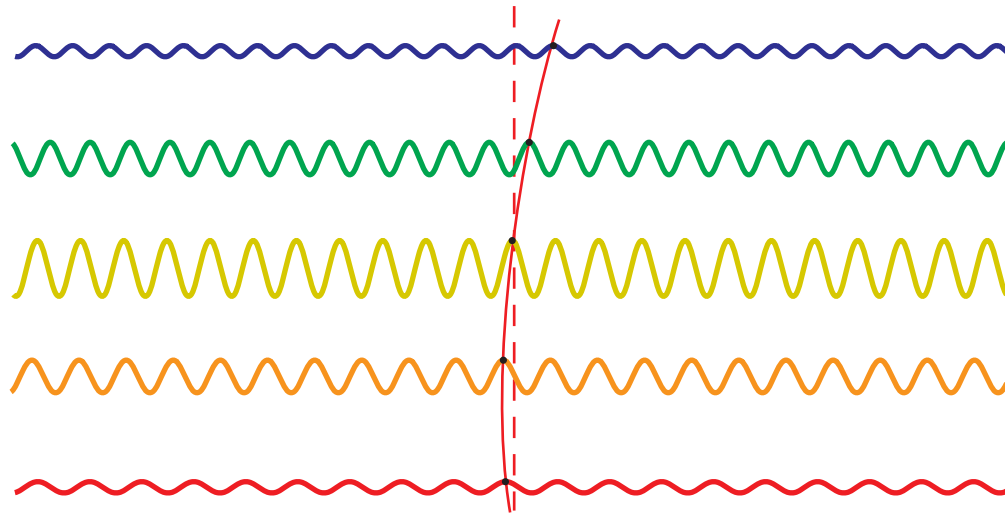
$$\frac{d\phi}{d\omega} = \text{constant}$$



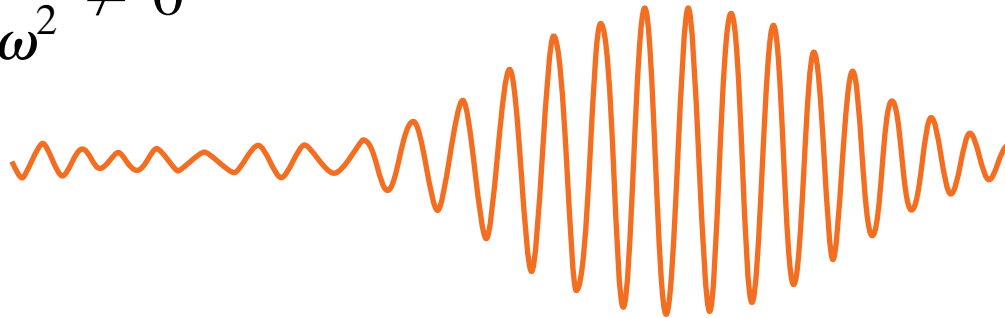
Dispersion compensation



Dispersion compensation



$$\frac{d^2\phi}{d\omega^2} \neq 0$$

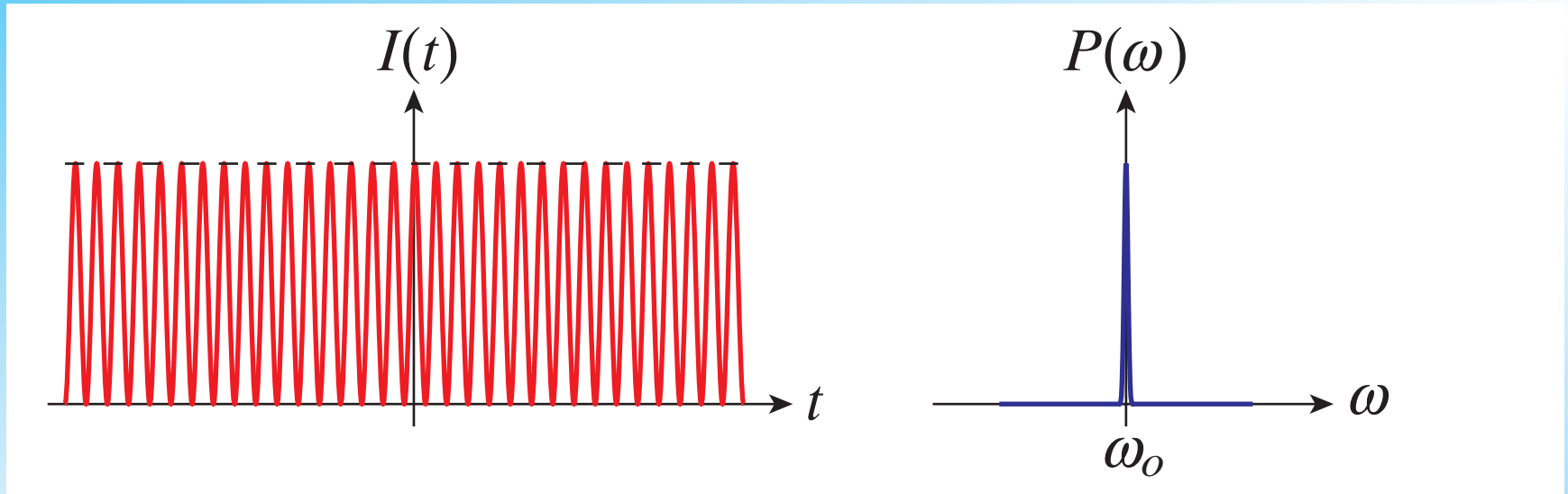


Dispersion compensation

So not path length but $\frac{d^2\phi}{d\omega^2}$ matters!

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

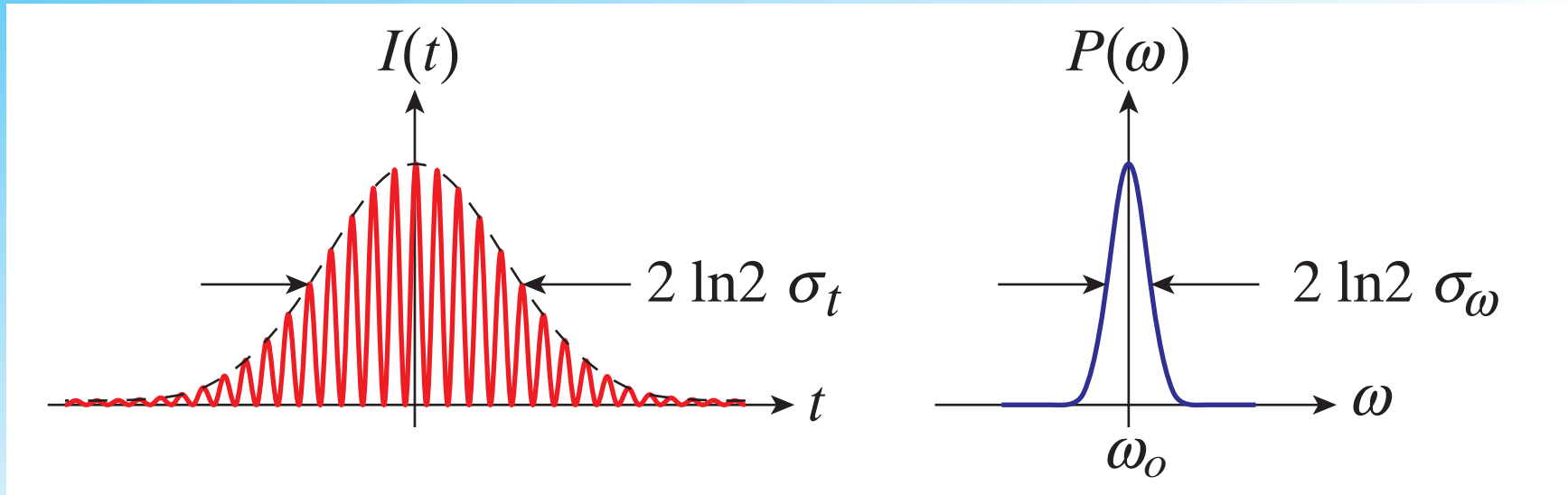
Representation of pulses



Spectrum of sinusoidal intensity is a delta function

$$I(t) = \cos^2(\omega_0 t) \quad \Rightarrow \quad P(\omega) = \delta(\omega - \omega_0)$$

Representation of pulses



Modulate amplitude

$$I(t) = \exp\left[-\frac{t^2}{\sigma_t^2}\right] \cos^2(\omega_0 t)$$

Representation of pulses

Fourier relations:

$$E(t) = \exp\left[-\frac{t^2}{2\sigma_t^2} - i\omega_0 t\right]$$

Representation of pulses

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$$E(\omega) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \exp\left[-\frac{t^2}{2\sigma_t^2} + i(\omega - \omega_o)t\right] dt =$$

Representation of pulses

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$$= \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{\sigma_t^2(\omega - \omega_o)^2}{2}\right] \int_{-\infty}^{\infty} \exp\left[-\left[\frac{t}{\sqrt{2}\sigma_t} - i\frac{(\omega - \omega_o)\sigma_t}{\sqrt{2}}\right]^2\right] dt =$$

Representation of pulses

Fourier relations:

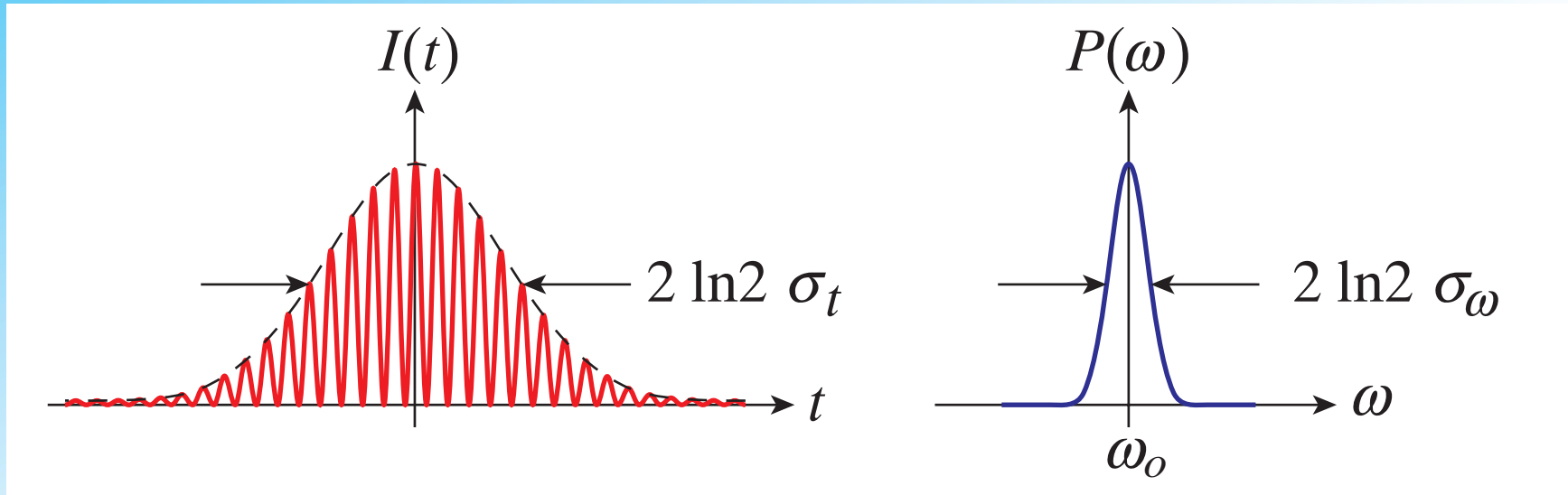
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$$= \frac{1}{\sqrt{2\pi}} \exp\left[-\frac{\sigma_t^2(\omega - \omega_0)^2}{2}\right] \int_{-\infty}^{\infty} \exp\left[-\left[\frac{t}{\sqrt{2}\sigma_t} - i\frac{(\omega - \omega_0)\sigma_t}{\sqrt{2}}\right]^2\right] dt =$$

$$= \sigma_t \exp\left[-\frac{\sigma_t^2(\omega - \omega_0)^2}{2}\right] \equiv \sigma_t \exp\left[-\frac{(\omega - \omega_0)^2}{2\sigma_\omega^2}\right]$$

Representation of pulses

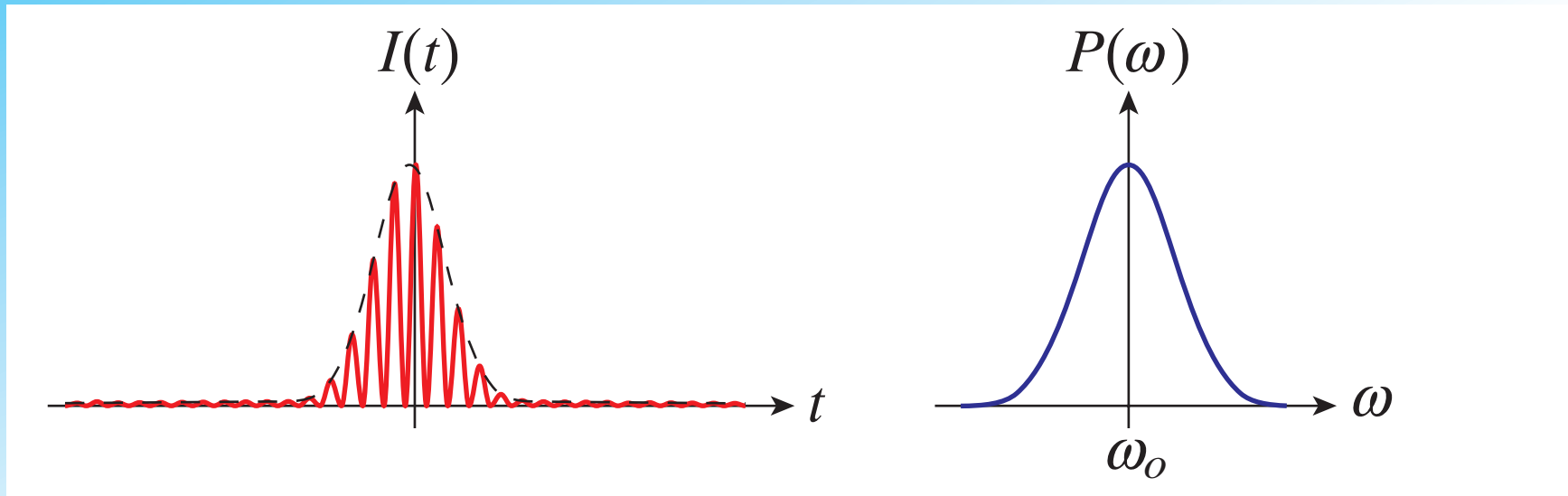


Pulse duration-bandwidth product: $\sigma_t \sigma_\omega = 1$

$$I(t) = [\text{Re } E(t)]^2 \propto \exp\left[-\frac{t^2}{\sigma_t^2}\right] \cos^2(\omega_0 t)$$

$$P(\omega) = E(\omega)E^*(\omega) = \exp\left[-\frac{(\omega - \omega_0)^2}{\sigma_\omega^2}\right]$$

Representation of pulses



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Joint time-frequency representation

Wigner representation:

$$\begin{aligned} W(t, \omega) &= \frac{1}{2\pi} \int_{-\infty}^{\infty} E\left(\omega + \frac{\omega'}{2}\right) E^*\left(\omega - \frac{\omega'}{2}\right) e^{-i\omega't} d\omega' = \\ &= \int_{-\infty}^{\infty} E\left(t + \frac{t'}{2}\right) E^*\left(t - \frac{t'}{2}\right) e^{-i\omega t'} dt' \end{aligned}$$

Joint time-frequency representation

Wigner representation:

$$W(t, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E\left(\omega + \frac{\omega'}{2}\right) E^*\left(\omega - \frac{\omega'}{2}\right) e^{-i\omega't} d\omega' =$$

$$= \int_{-\infty}^{\infty} E\left(t + \frac{t'}{2}\right) E^*\left(t - \frac{t'}{2}\right) e^{-i\omega t'} dt'$$

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} W(t, \omega) d\omega = |E(t)|^2 = I(t)$$

Joint time-frequency representation

Wigner representation:

$$W(t, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} E\left(\omega + \frac{\omega'}{2}\right) E^*\left(\omega - \frac{\omega'}{2}\right) e^{-i\omega't} d\omega' =$$

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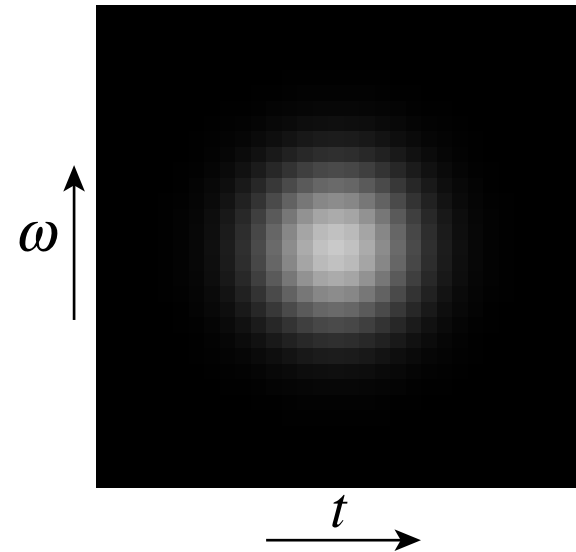
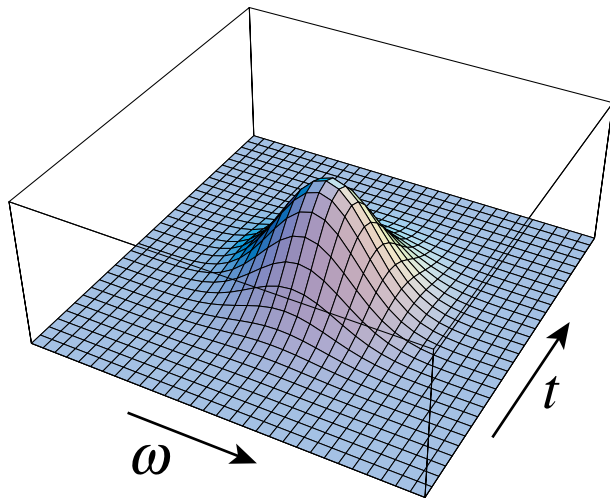
$$\frac{1}{2\pi} \int_{-\infty}^{\infty} W(t, \omega) d\omega = |E(t)|^2 = I(t)$$

$$\int_{-\infty}^{\infty} W(t, \omega) dt = |E(\omega)|^2 = I(\omega)$$

Joint time-frequency representation

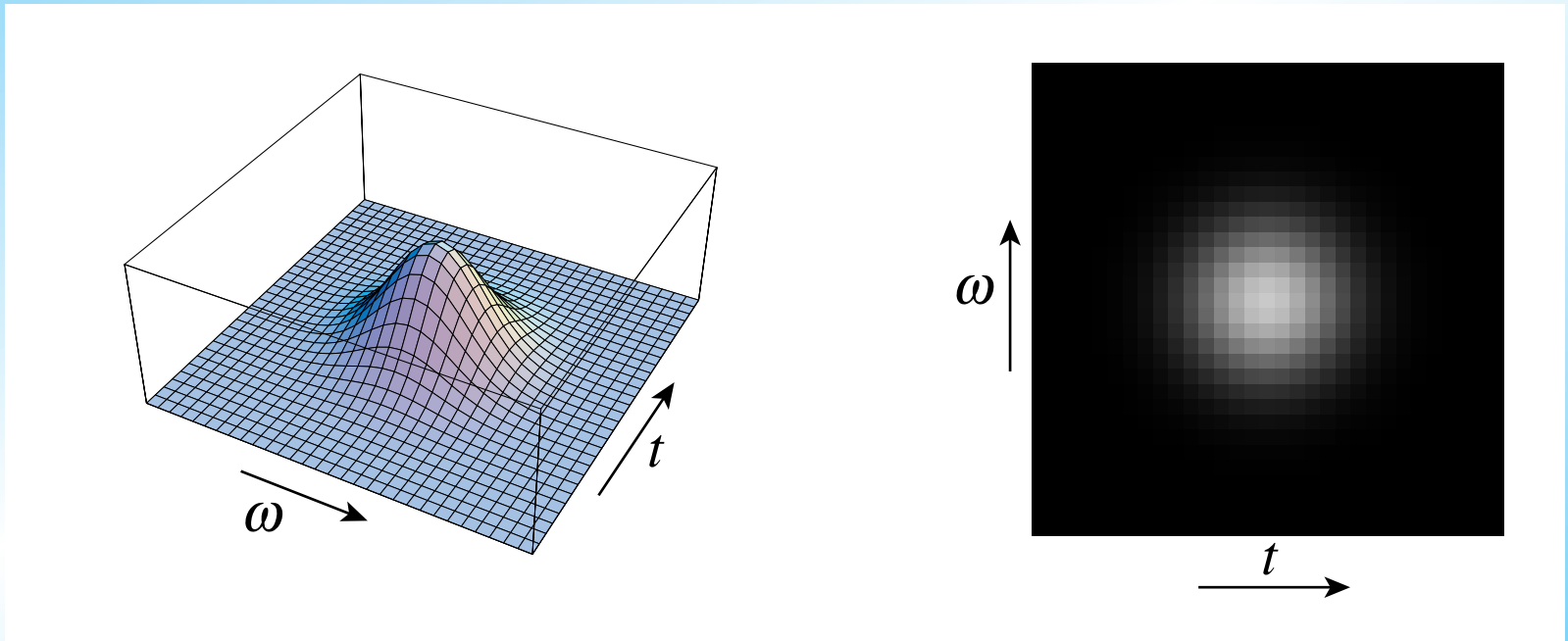
Energy:

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} W(t, \omega) dt d\omega$$



Joint time-frequency representation

Energy:
$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} W(t, \omega) dt d\omega$$

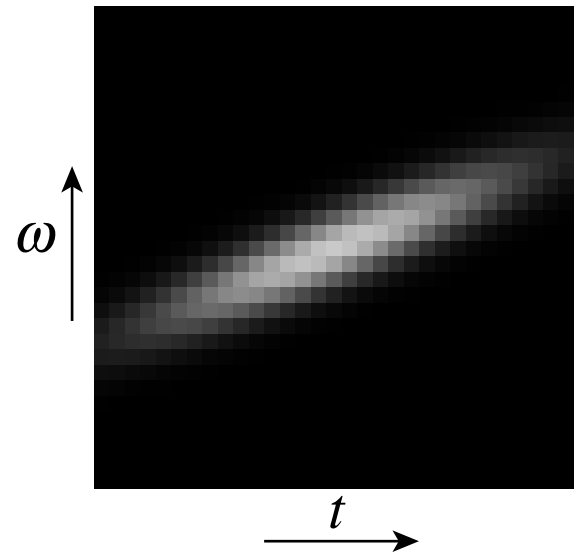


$W(t, \omega)$ must be nonzero in phase-space area larger than π

Joint time-frequency representation

Energy:
$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} W(t, \omega) dt d\omega$$

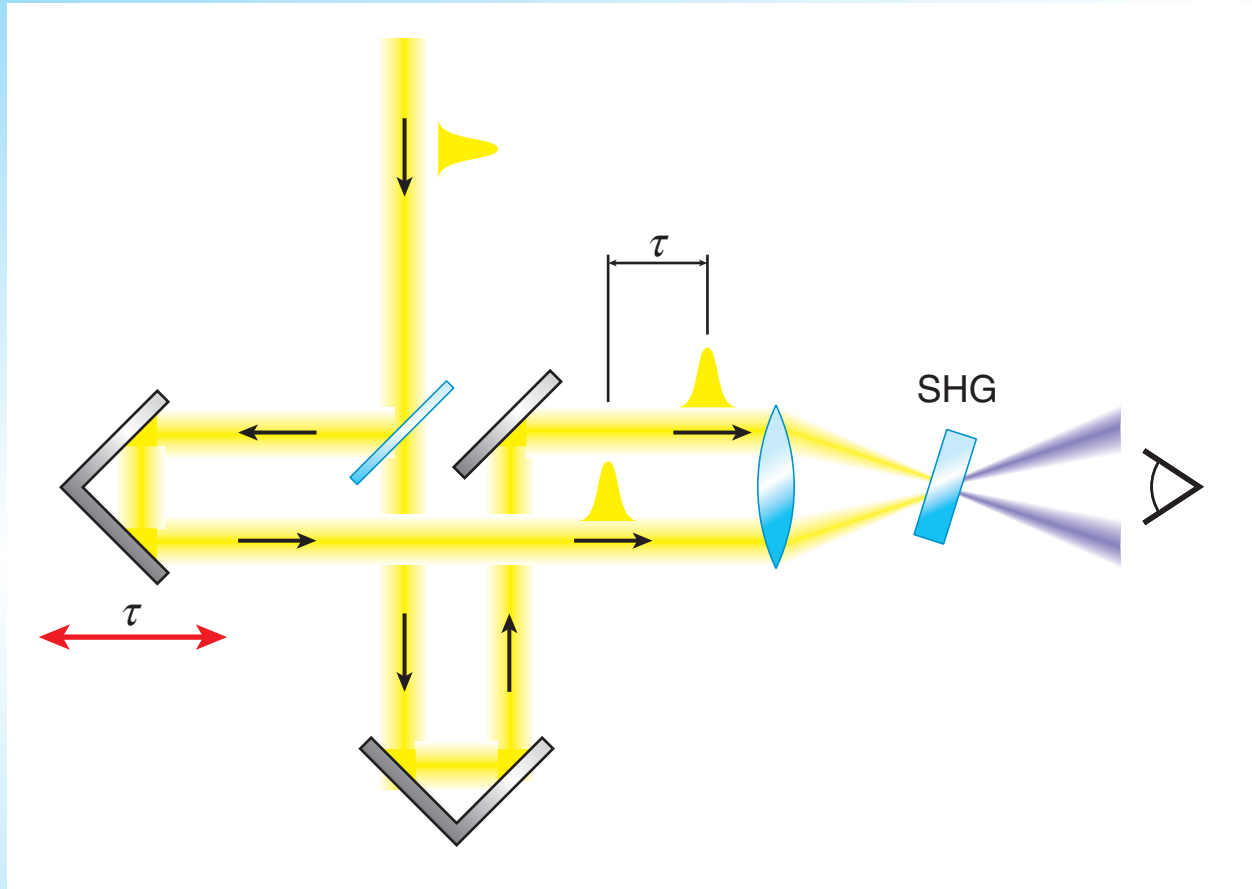
chirped pulse



$W(t, \omega)$ must be nonzero in phase-space area larger than π

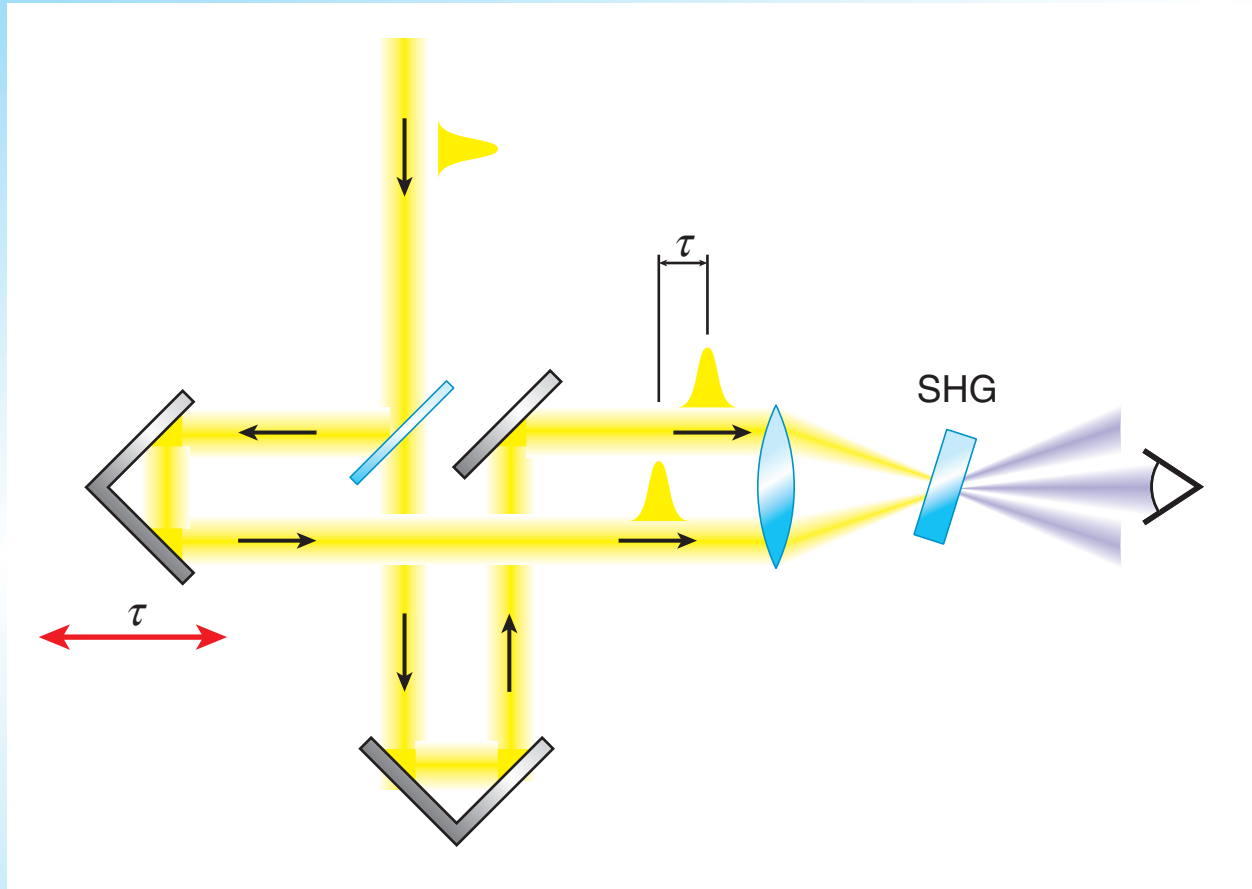
Temporal characterization

Use pulse to measure itself...



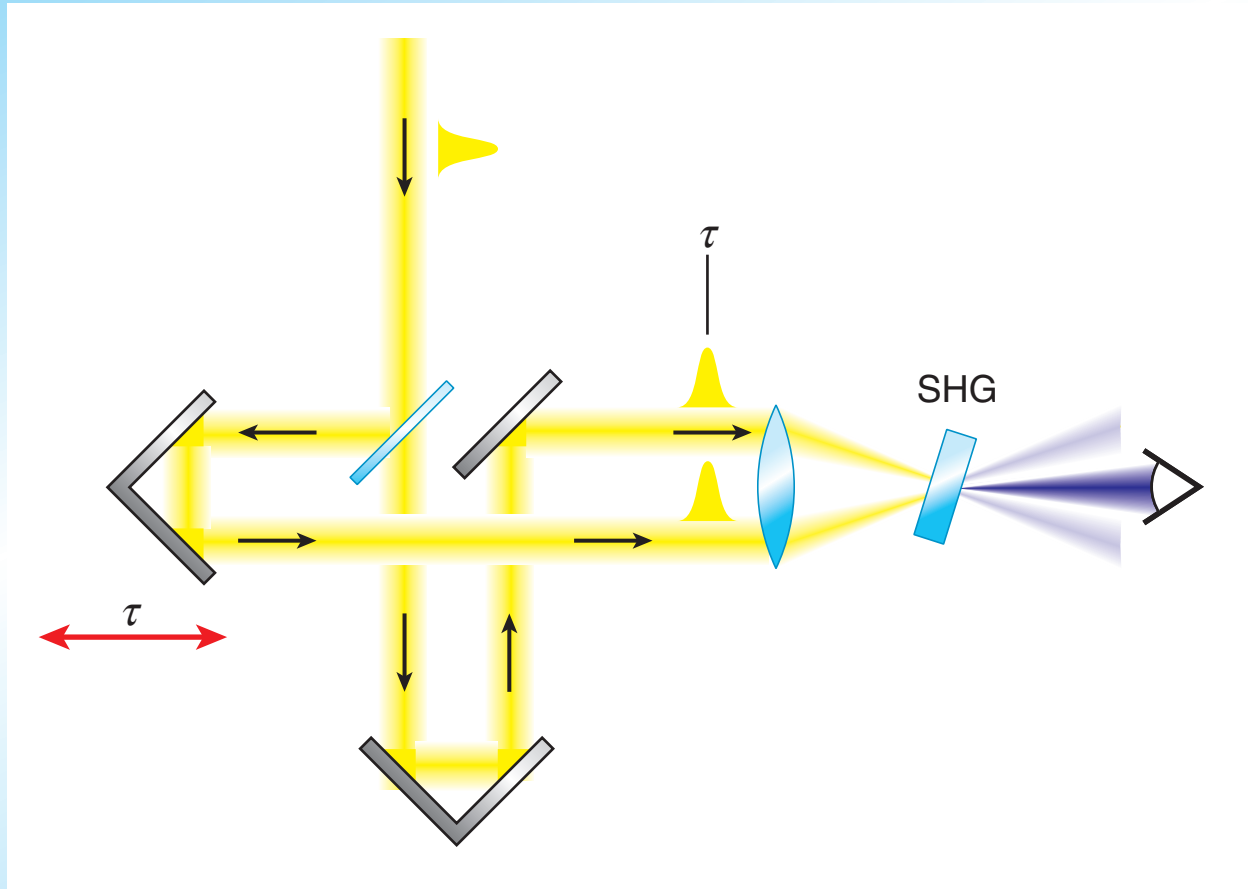
Temporal characterization

Use pulse to measure itself...



Temporal characterization

Use pulse to measure itself...



Temporal characterization

Electric field at SHG crystal

$$E_{tot}(t, \tau) = \frac{1}{\sqrt{2}}E_1(t) + \frac{1}{\sqrt{2}}E_2(t + \tau)$$

Temporal characterization

Electric field at SHG crystal

$$E_{tot}(t, \tau) = \frac{1}{\sqrt{2}}E_1(t) + \frac{1}{\sqrt{2}}E_2(t + \tau)$$

Second harmonic field

$$E_{2\omega} \propto \chi^{(2)} E_{tot}^2$$

Temporal characterization

Electric field at SHG crystal

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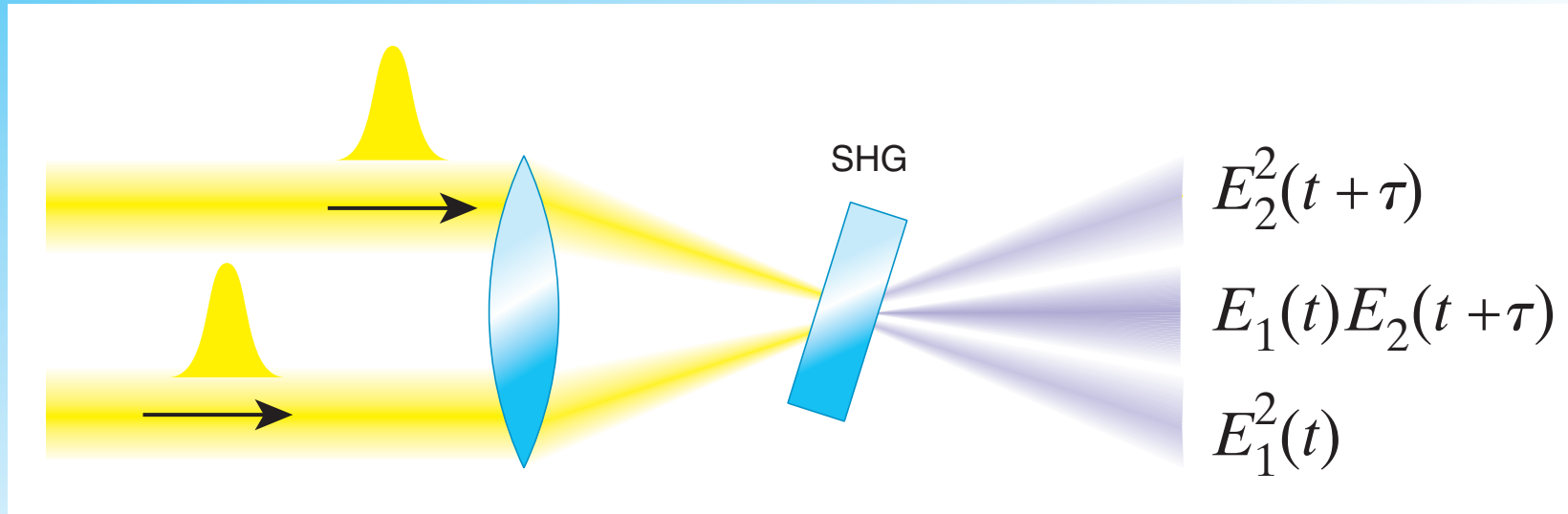
Second harmonic field

$$E_{2\omega} \propto \chi^{(2)} E_{tot}^2$$

Second harmonic intensity

$$I_{2\omega}(t, \tau) \propto |\chi^{(2)}|^2 |E_{tot}^2|^2 = |\chi^{(2)}|^2 |E_1^2(t) + 2E_1(t)E_2(t + \tau) + E_2^2(t + \tau)|^2$$

Temporal characterization



Second harmonic intensity

$$I_{2\omega}(t, \tau) \propto |\chi^{(2)}|^2 |E_{tot}^2|^2 = |\chi^{(2)}|^2 |E_1^2(t) + 2E_1(t)E_2(t + \tau) + E_2^2(t + \tau)|^2$$

detector selects middle term

Temporal characterization

Integrated detector signal yields intensity autocorrelation

$$A(\tau) = \int I_{2\omega}(t, \tau) dt \propto \int |\chi^{(2)}|^2 4|E_1(t)|^2 |E_2(t + \tau)|^2 dt$$

Temporal characterization

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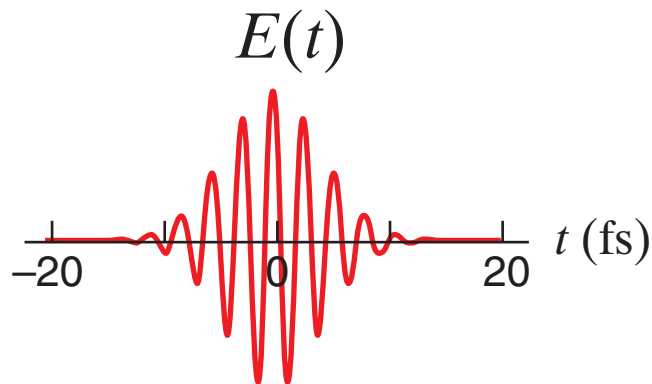
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Temporal characterization

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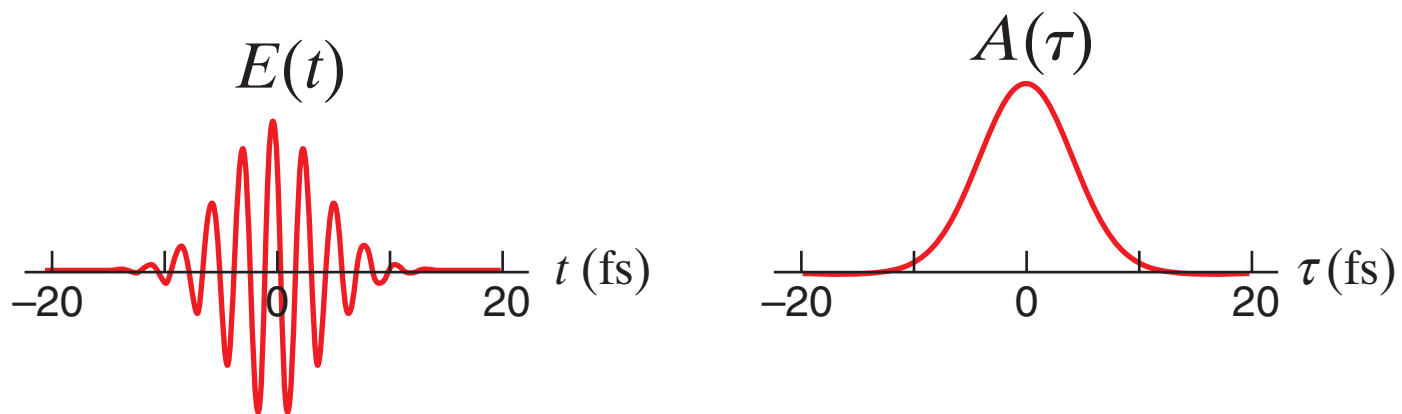


Temporal characterization

Integrated detector signal yields intensity autocorrelation

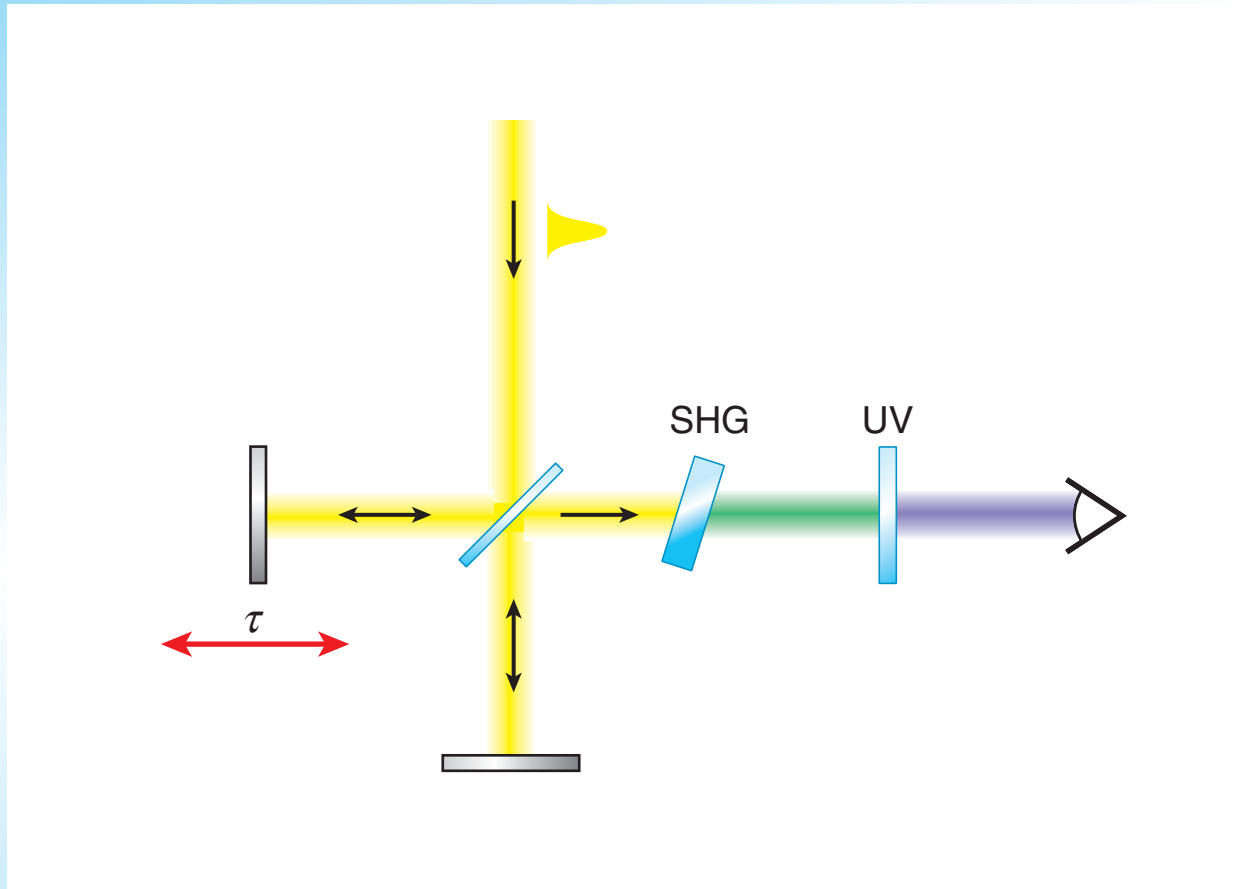
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Temporal characterization

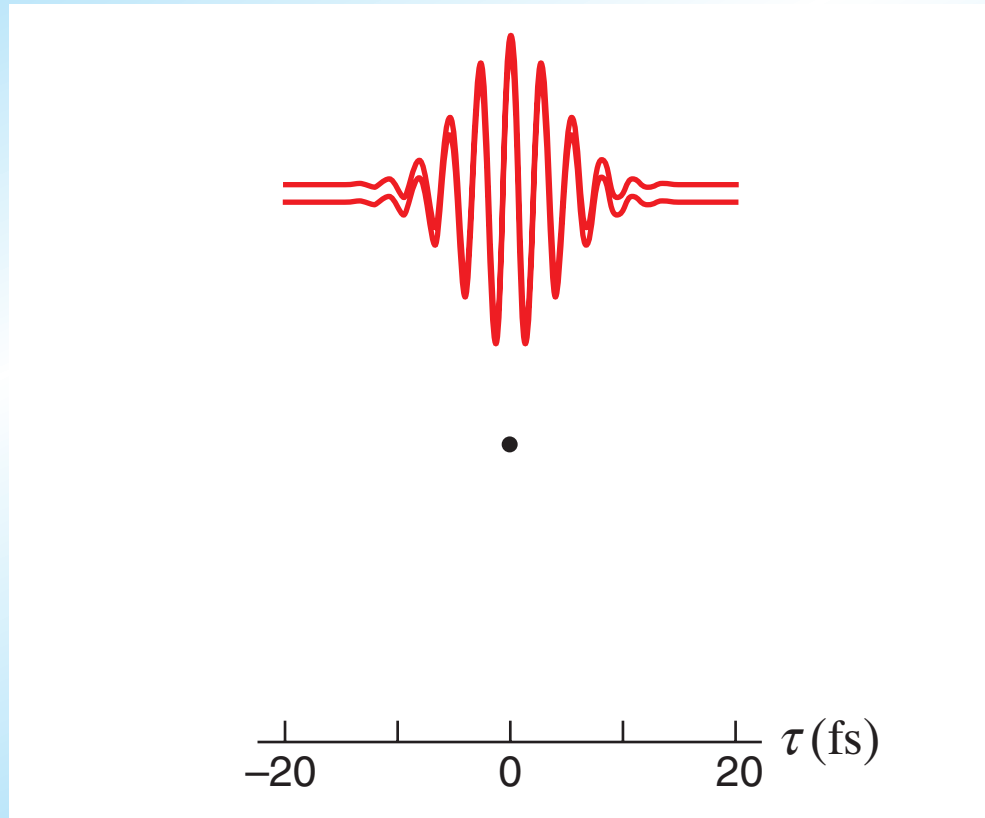
Alternative colinear geometry



Temporal characterization

All terms now contribute:

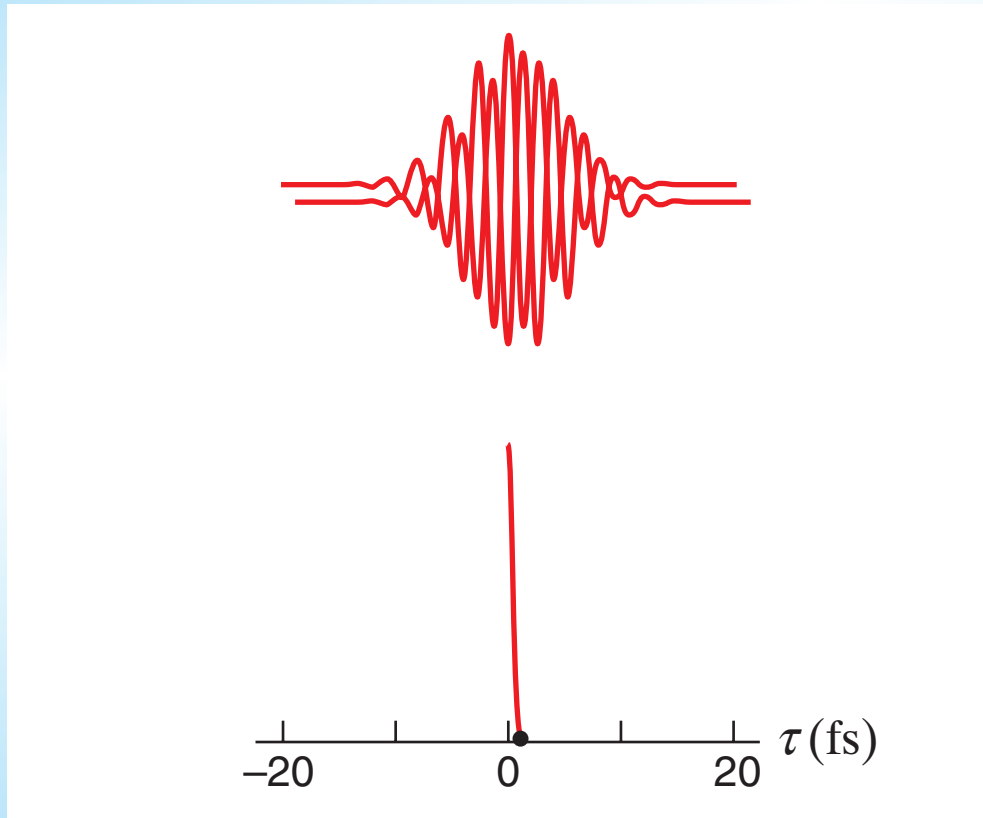
$$I_{2\omega}(t, \tau) \propto |\chi^{(2)}|^2 |E_{tot}^2|^2 = |\chi^{(2)}|^2 |E_1^2(t) + 2E_1(t)E_2(t+\tau) + E_2^2(t+\tau)|^2$$



Temporal characterization

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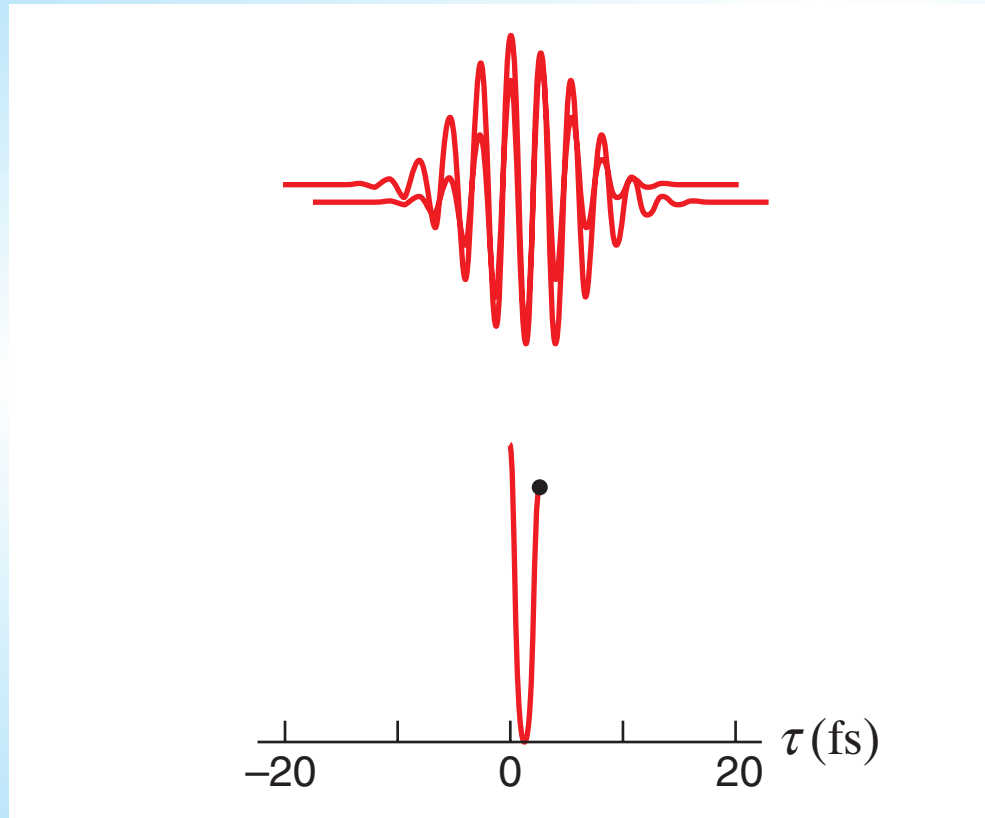
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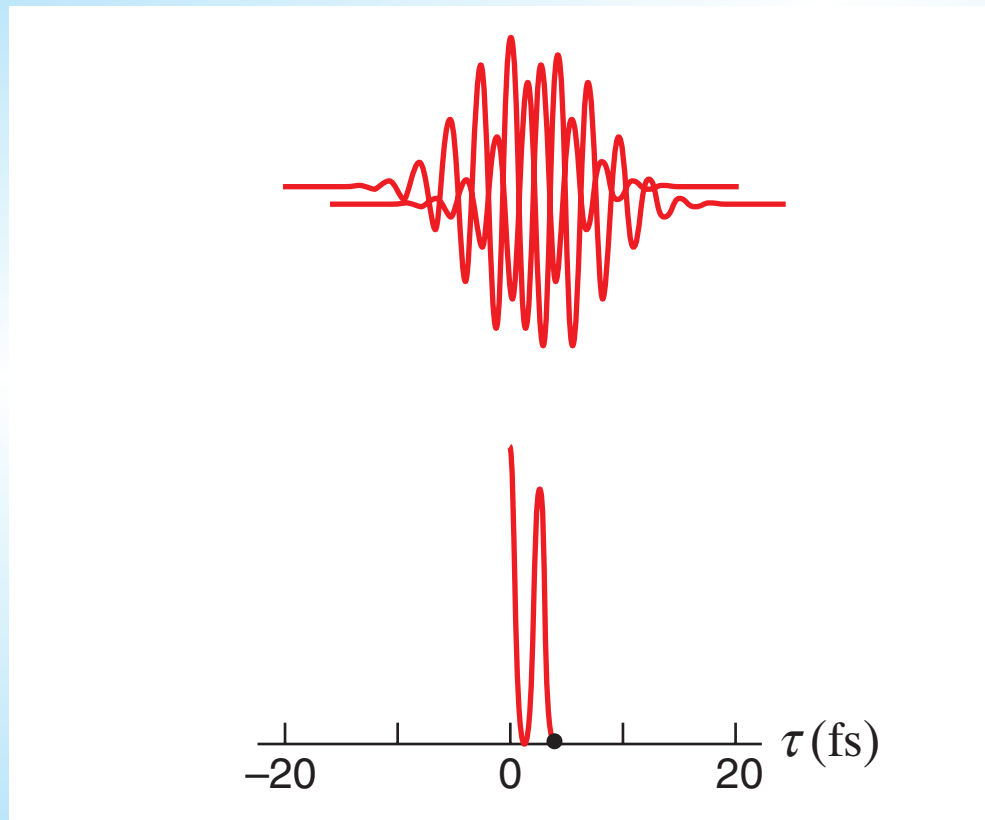
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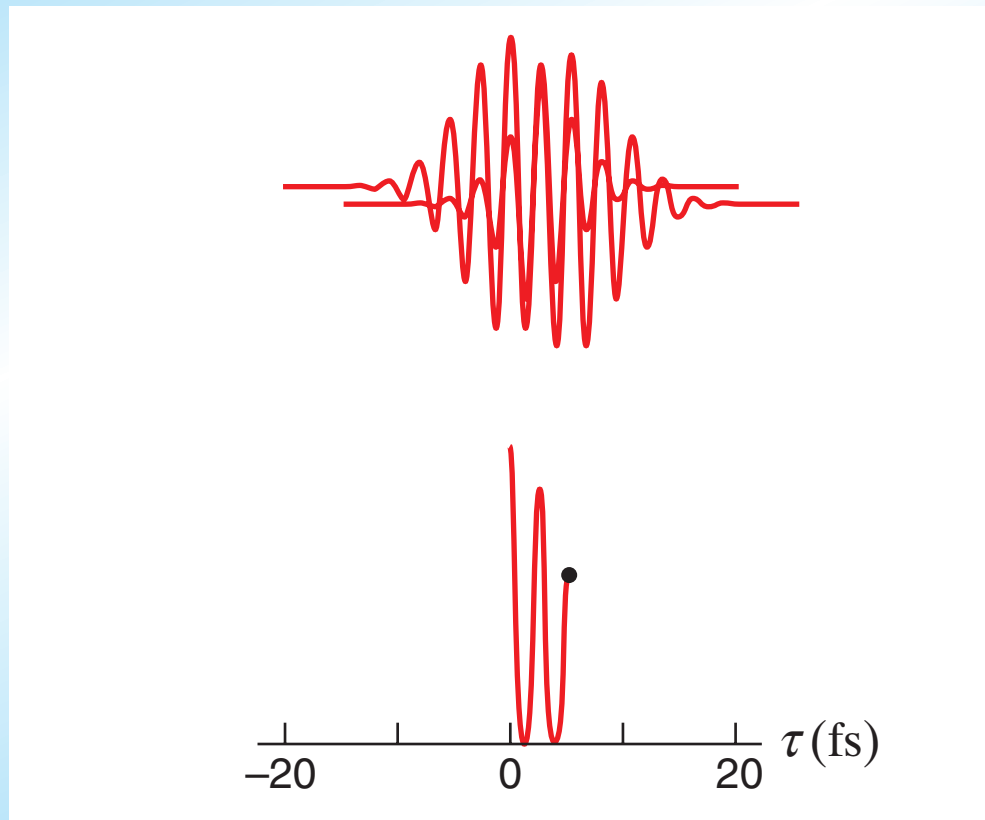
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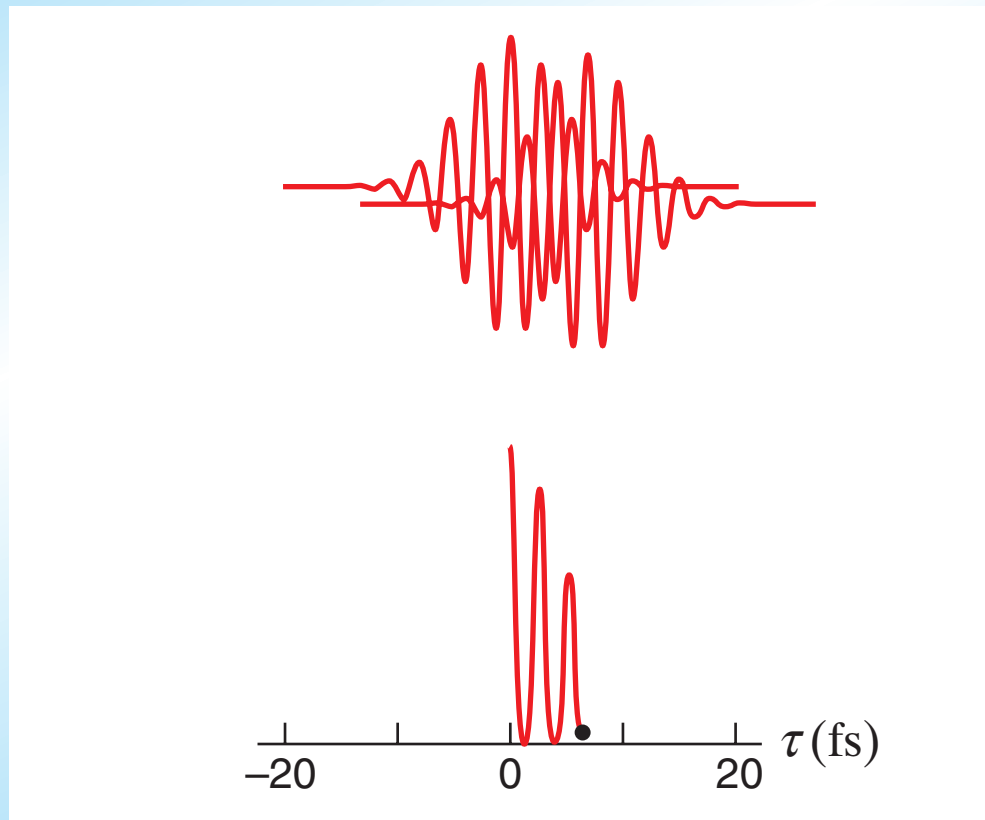
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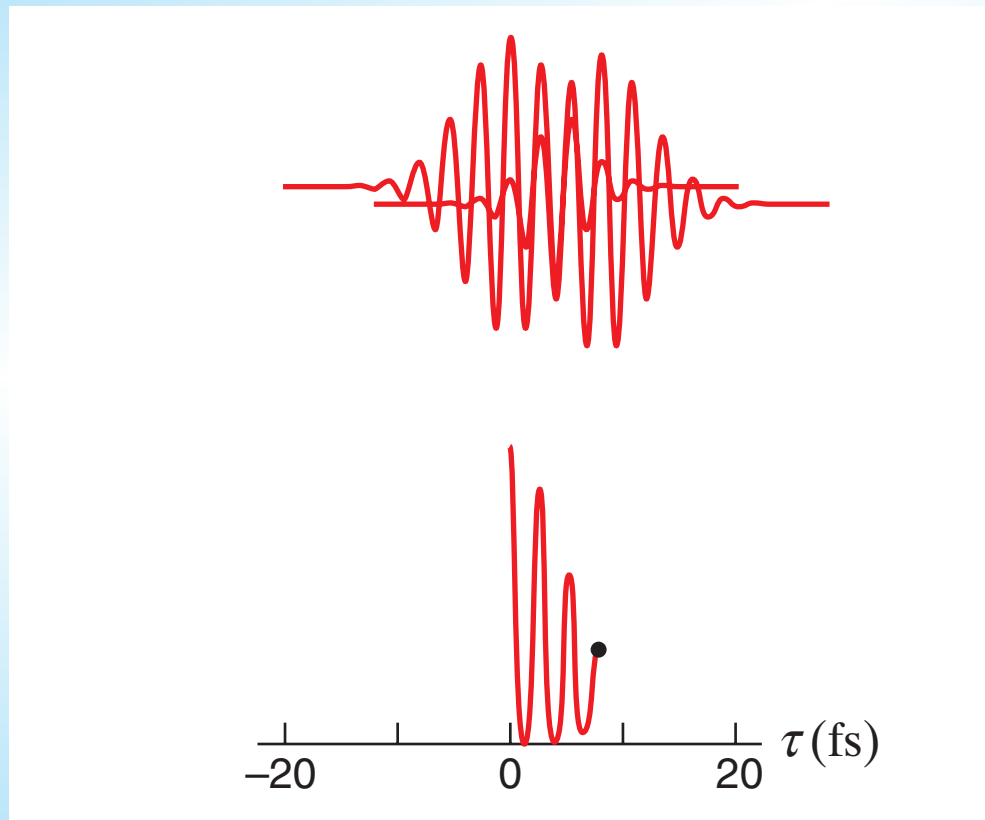
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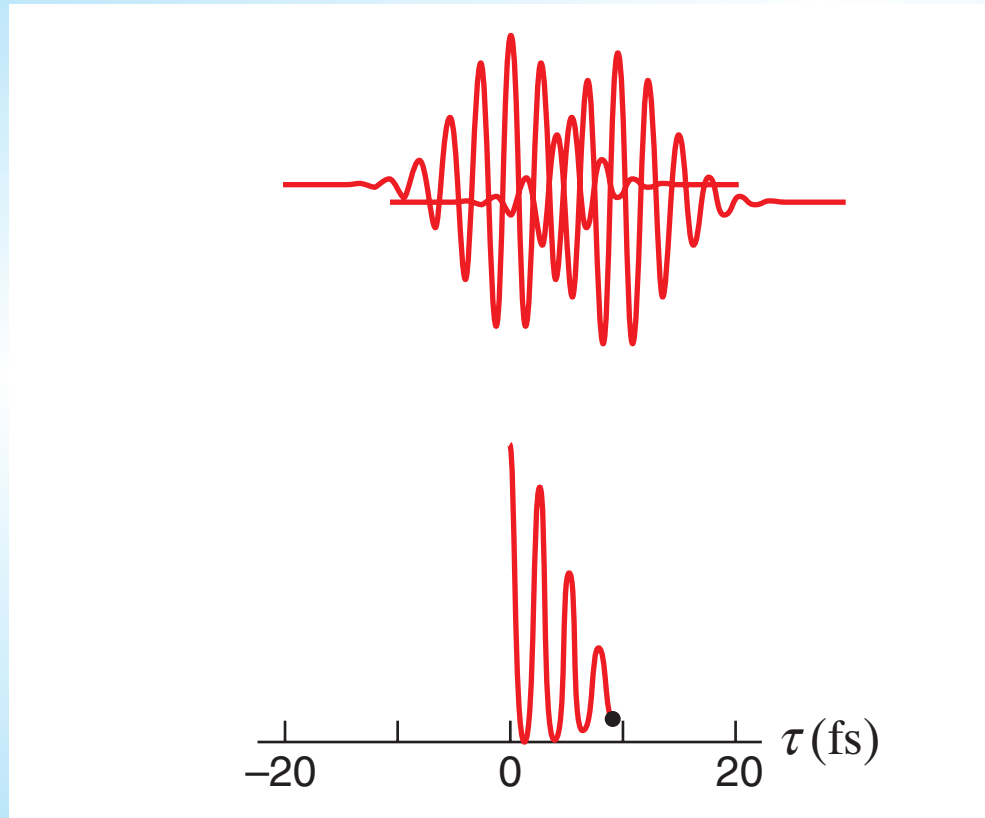
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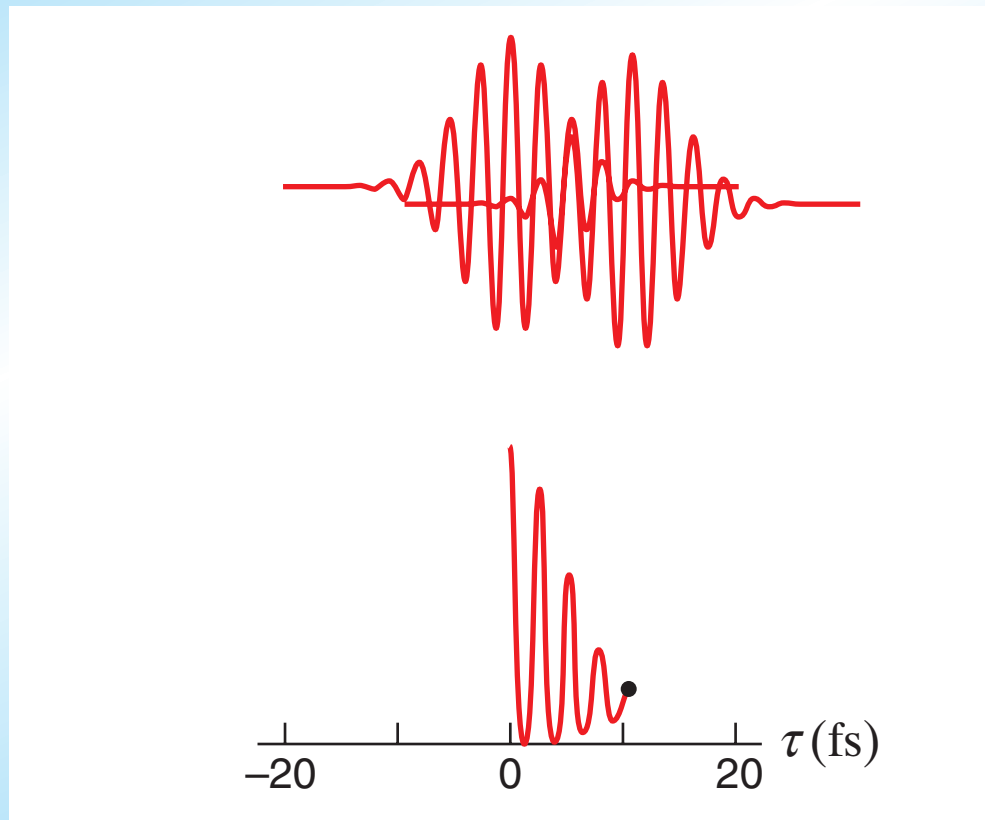
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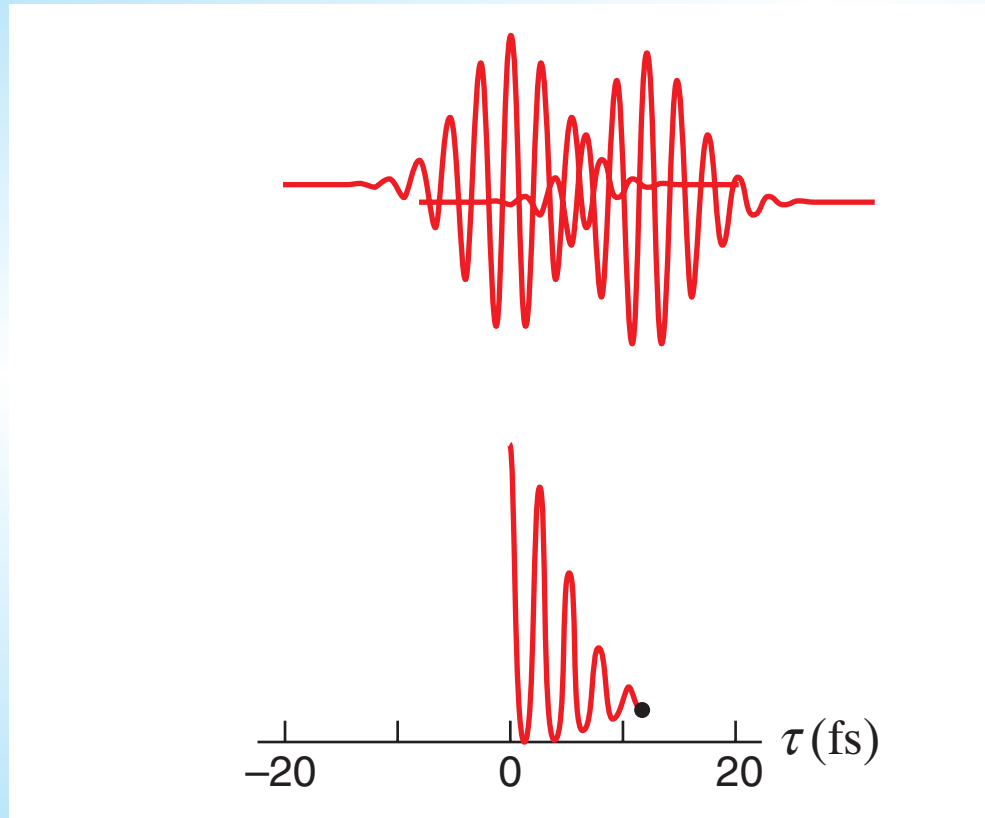
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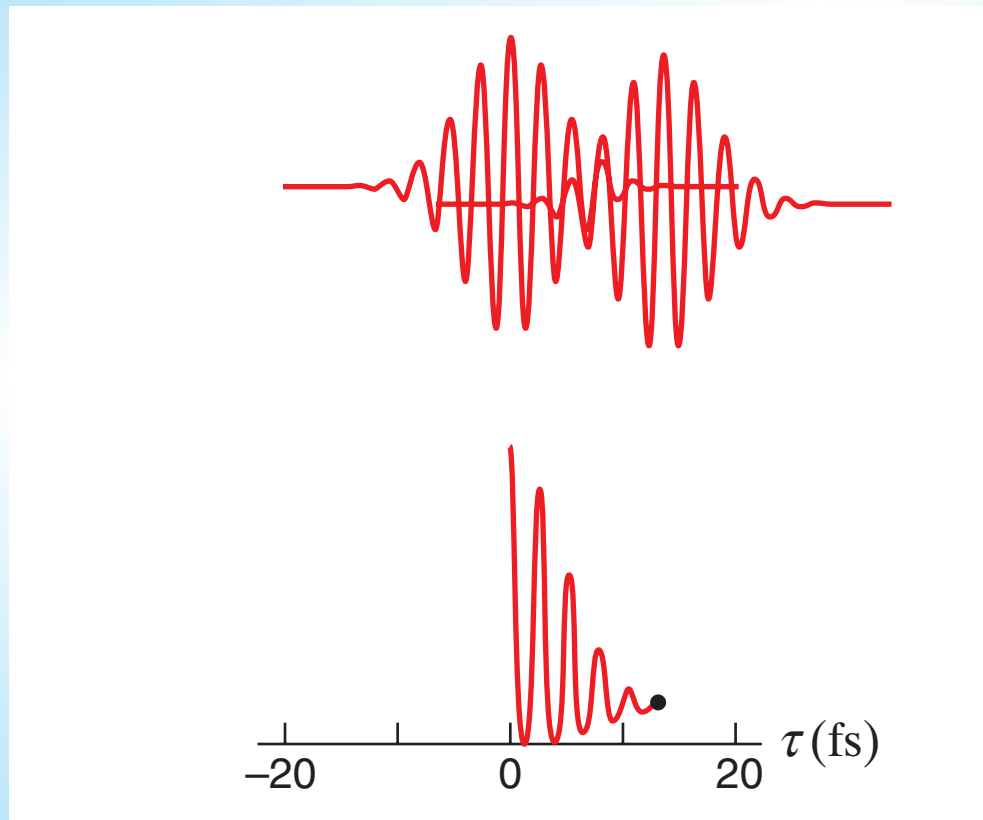
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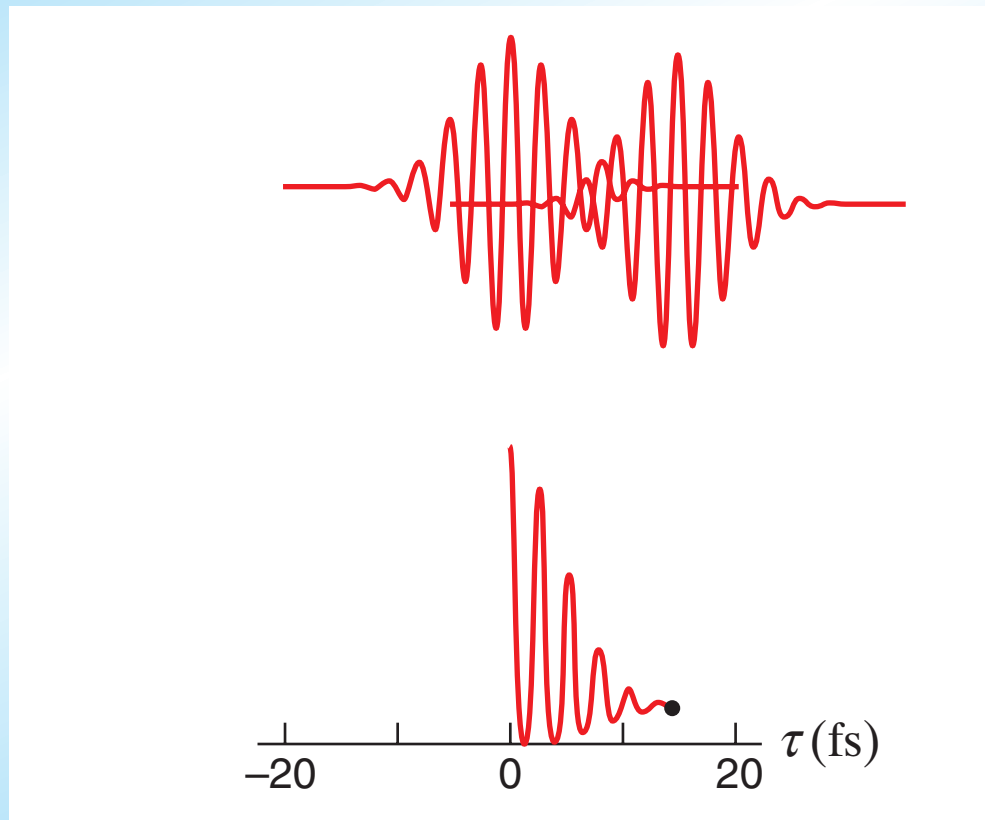
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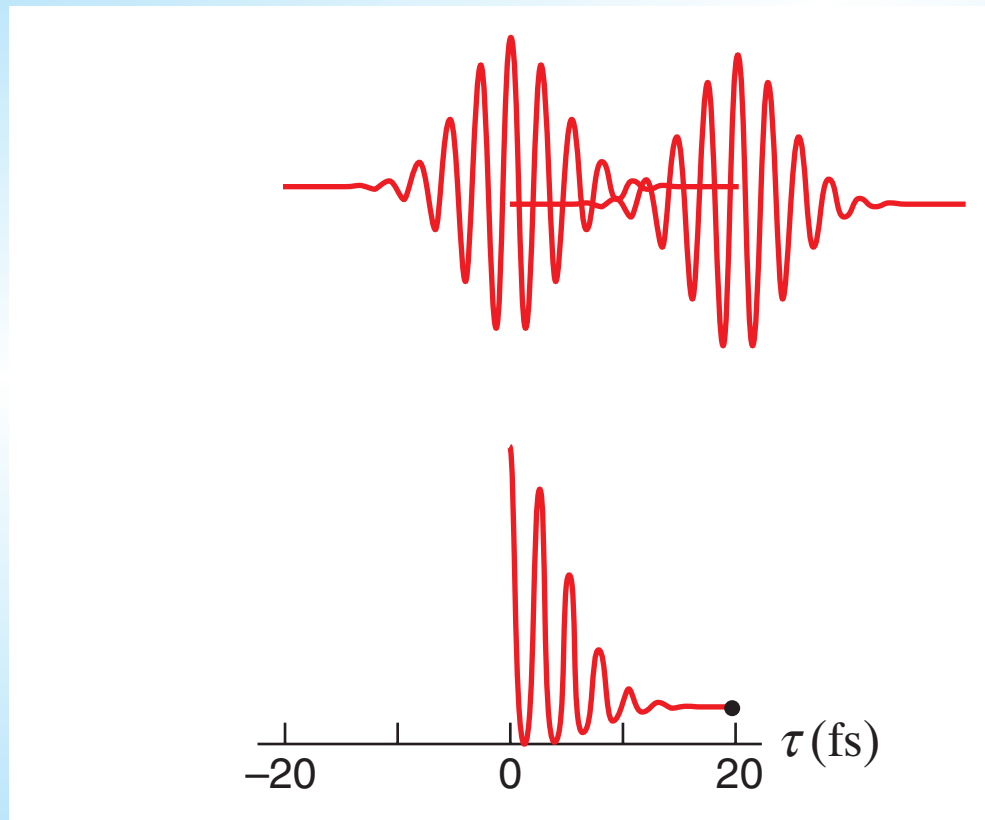
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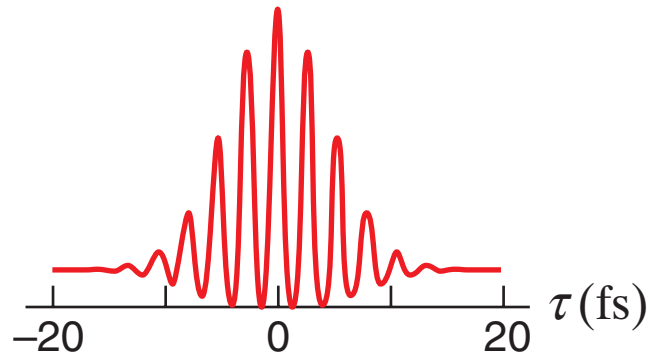
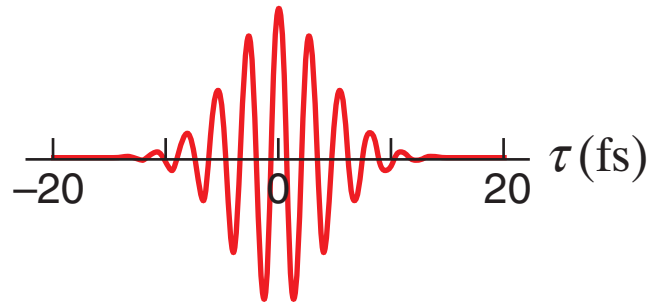
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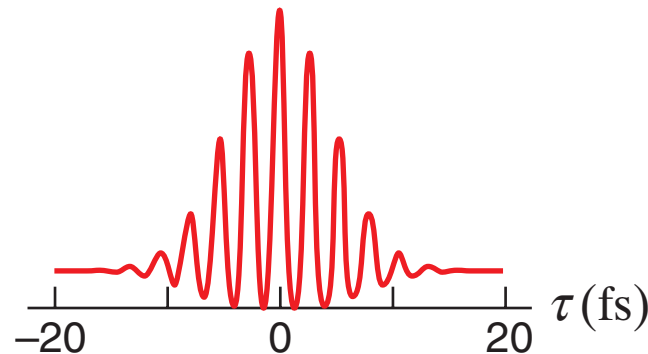
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at $\tau = 0$:

$$I_{2\omega}(t, \tau) \propto 16E^4(t)$$



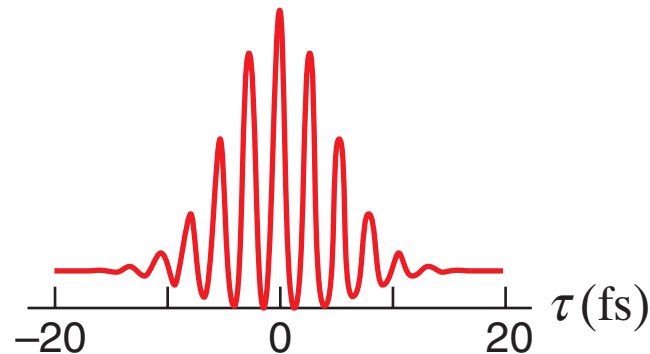
Temporal characterization

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at $\tau = 0$: $I_{2\omega}(t, \tau) \propto 16E^4(t)$

as $\tau \rightarrow \pm \infty$: $I_{2\omega}(t, \tau) \propto 2E^4(t)$



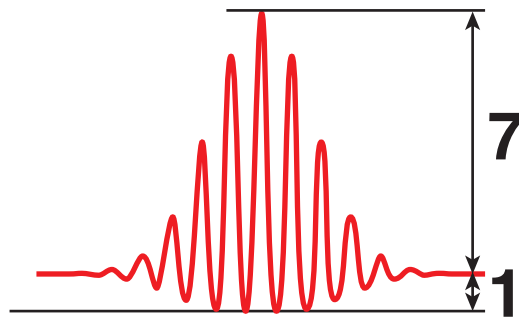
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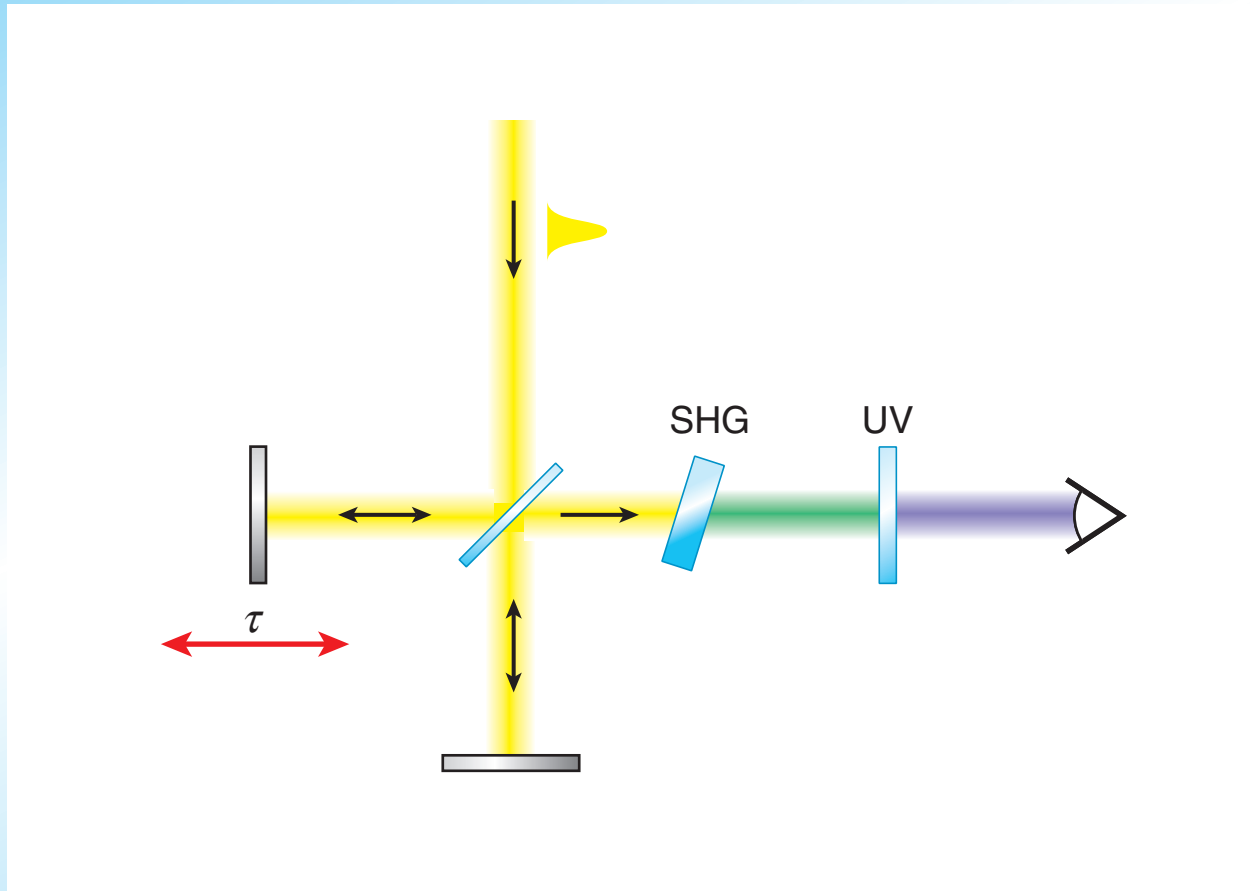
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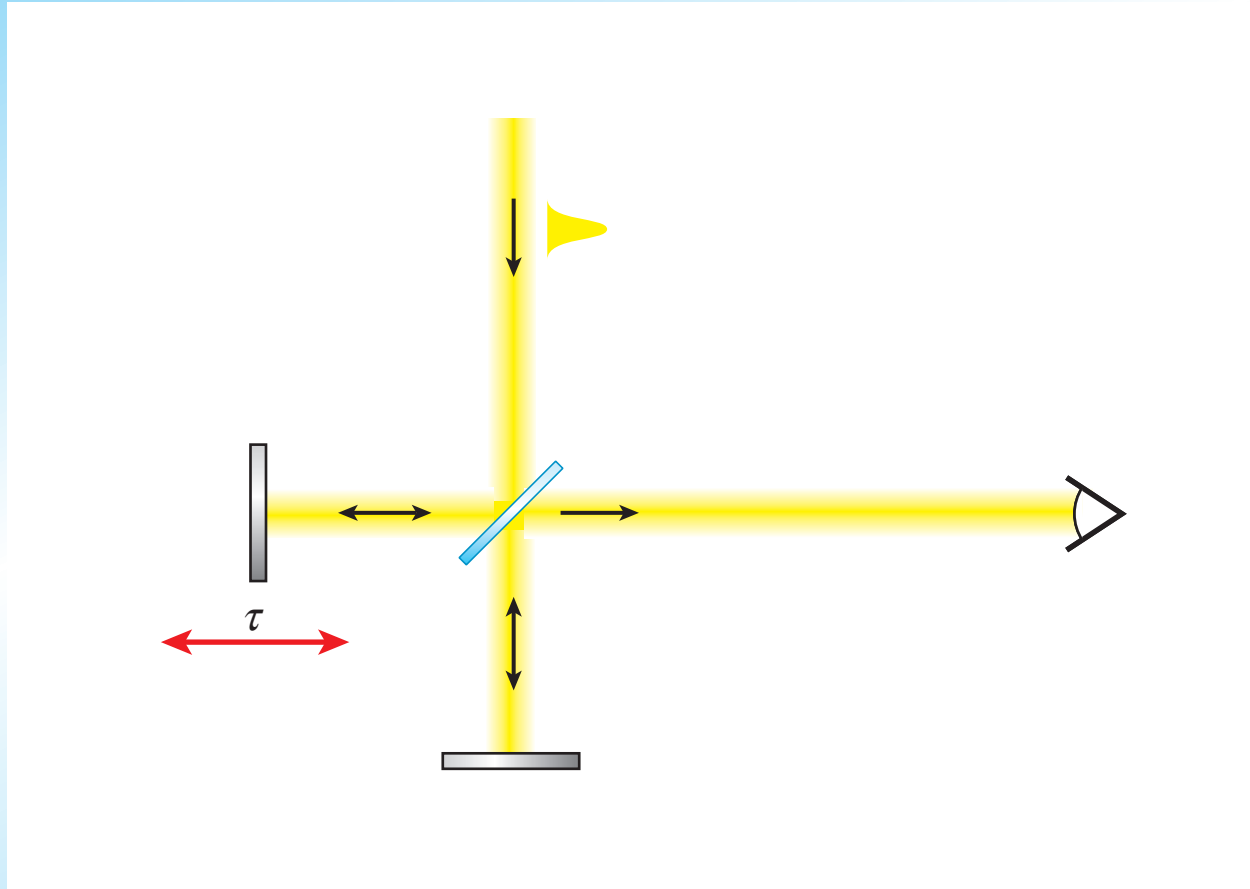
Temporal characterization

Do we really need the second-harmonic crystal...?



Temporal characterization

Would this work?



Temporal characterization

Intensity at detector

$$I_{\omega}(t, \tau) \propto |E_1(t) + E_2(t + \tau)|^2$$

Temporal characterization

Intensity at detector

$$I_{\omega}(t, \tau) \propto |E_1(t) + E_2(t + \tau)|^2$$

Detected signal

$$S_{\omega}(\tau) = \int I_{\omega}(t, \tau) dt$$

Temporal characterization

Intensity at detector

$$I_{\omega}(t, \tau) \propto |E_1(t) + E_2(t + \tau)|^2$$

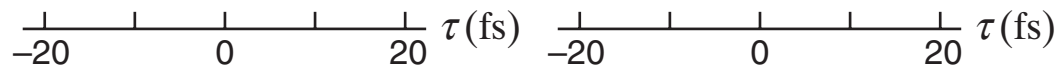
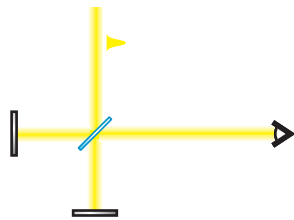
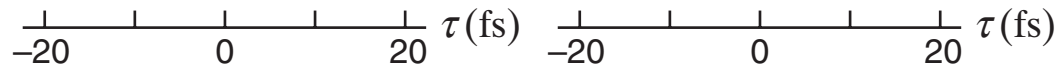
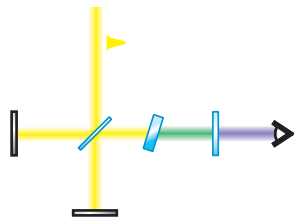
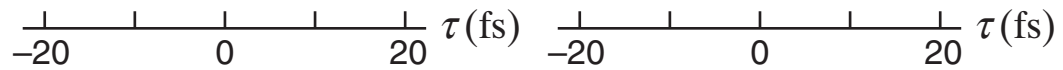
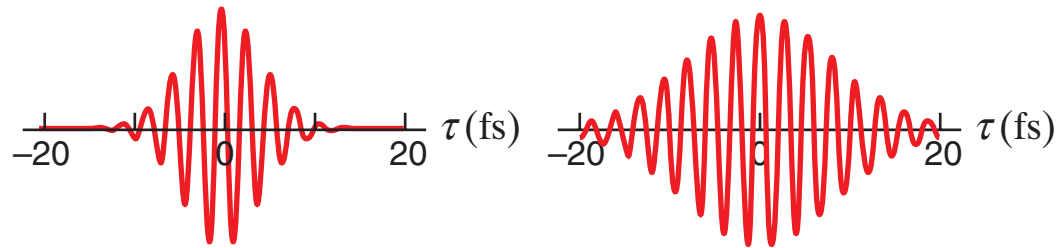
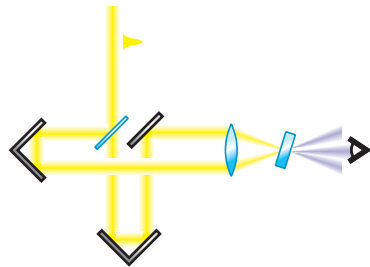
Detected signal

$$S_{\omega}(\tau) = \int I_{\omega}(t, \tau) dt$$

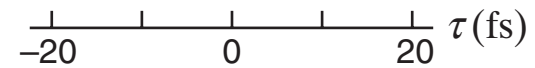
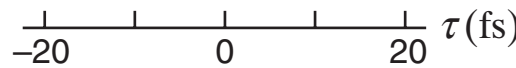
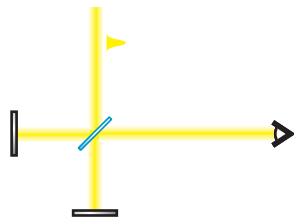
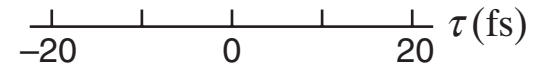
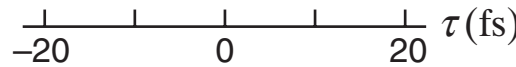
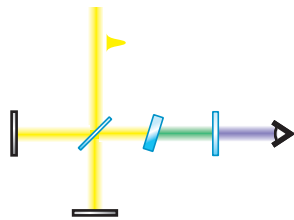
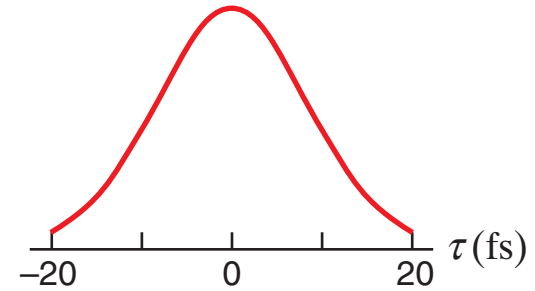
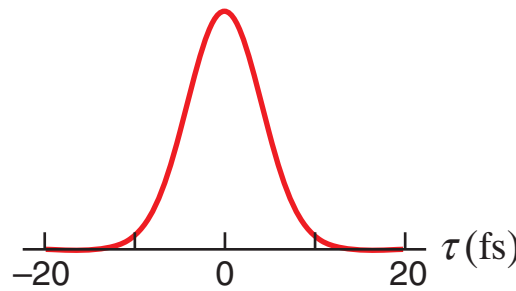
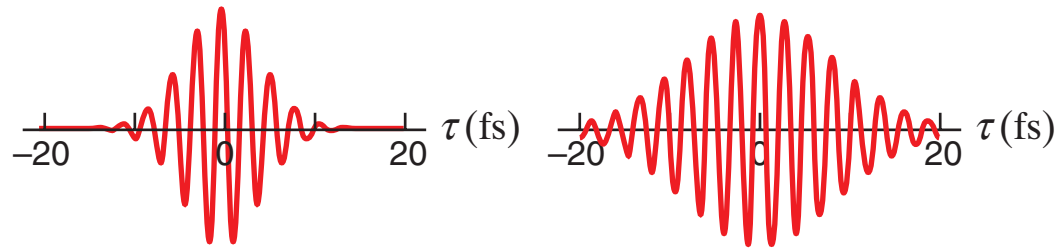
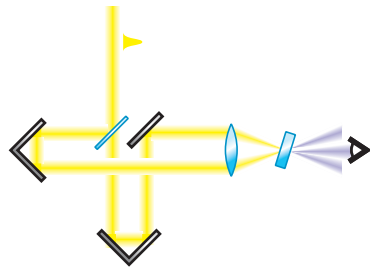
so

$$S_{\omega}(\tau) \propto \int \{|E_1(t)|^2 + |E_2(t + \tau)|^2 + E_1(t)E_2^*(t + \tau) + E_1^*(t)E_2(t + \tau)\} dt$$

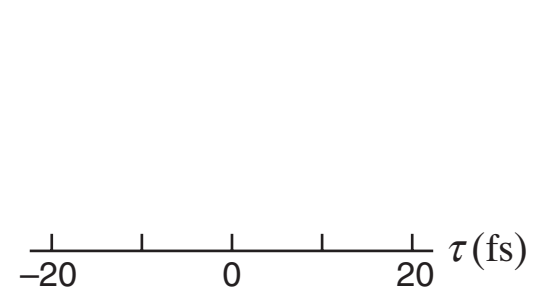
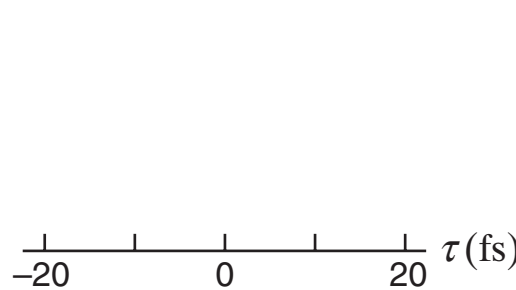
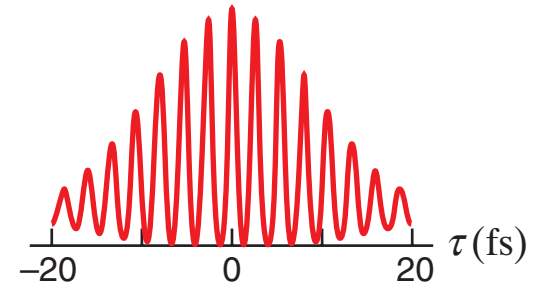
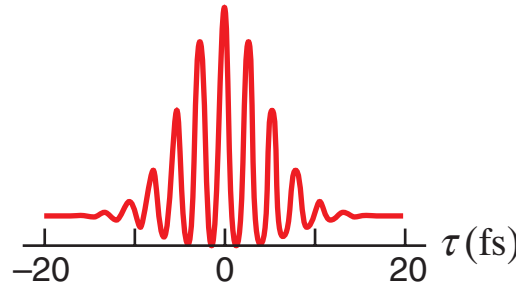
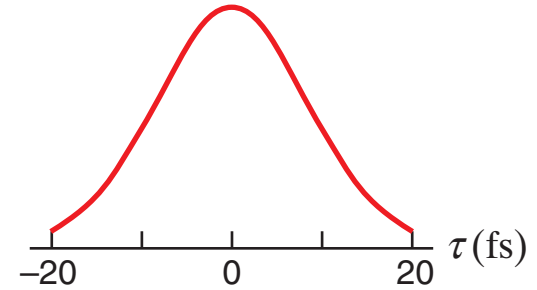
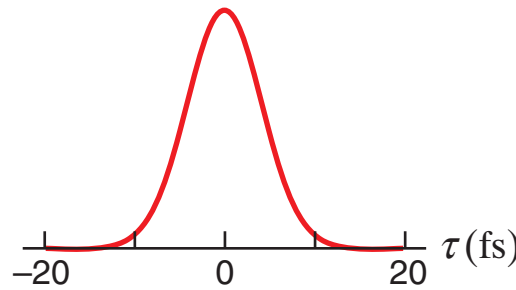
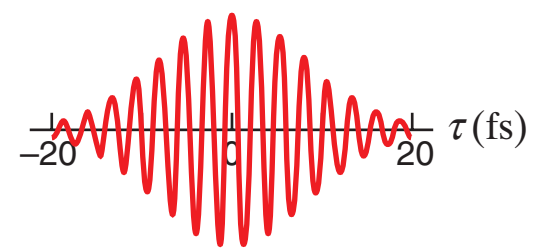
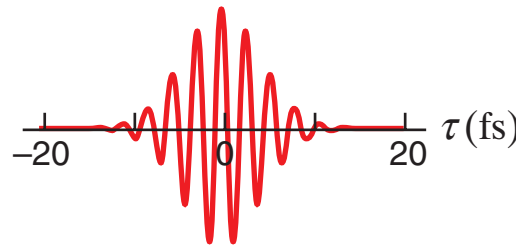
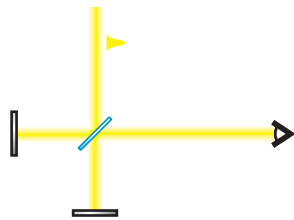
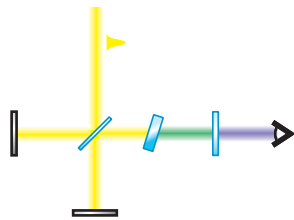
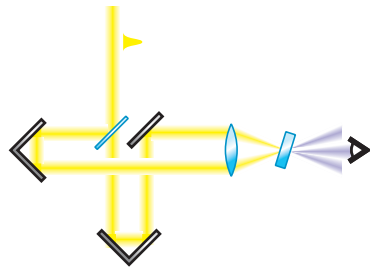
Temporal characterization



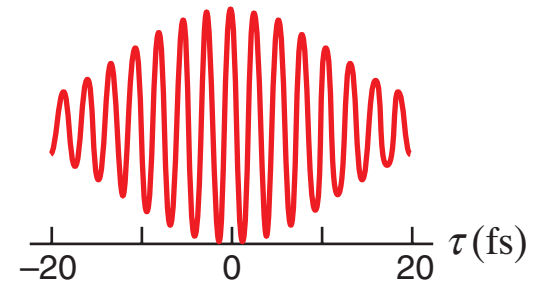
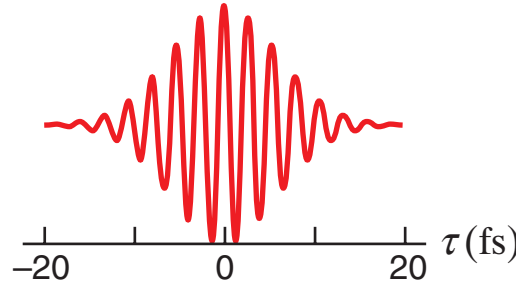
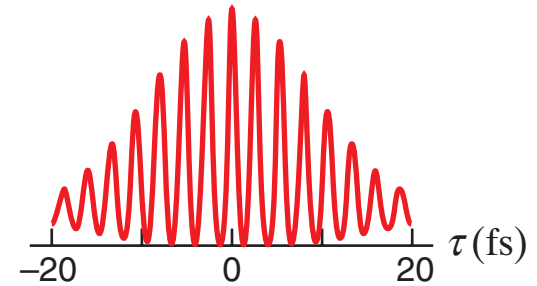
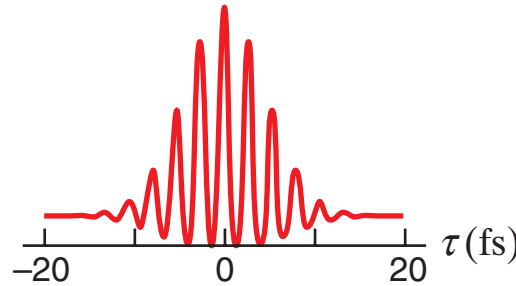
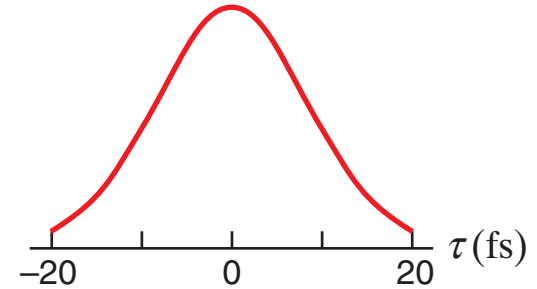
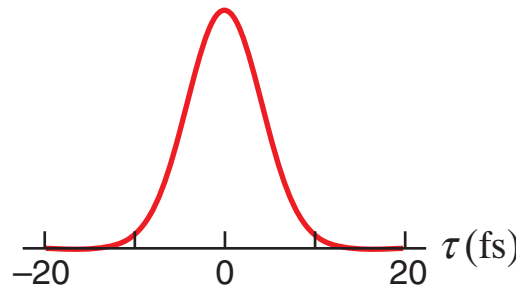
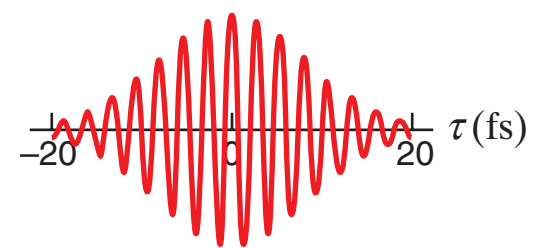
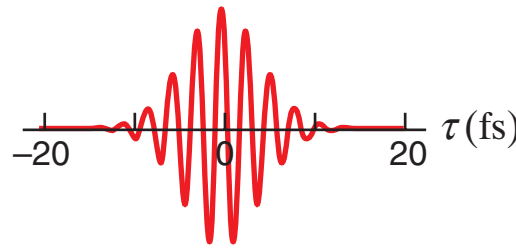
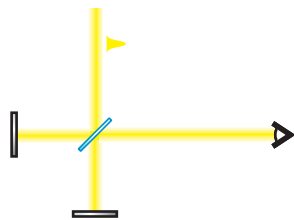
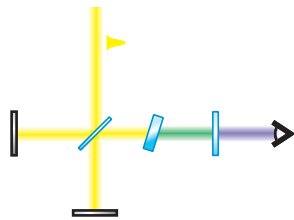
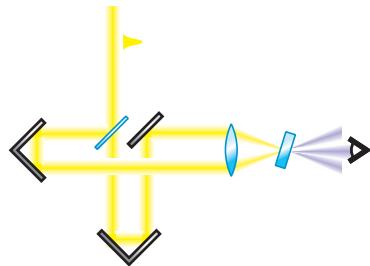
Temporal characterization



Temporal characterization



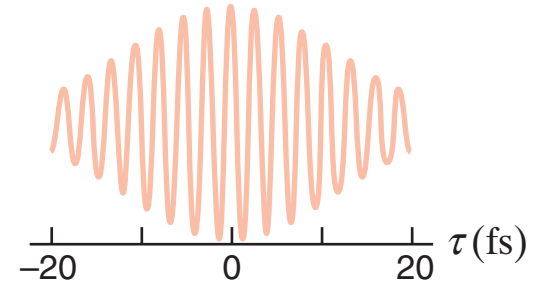
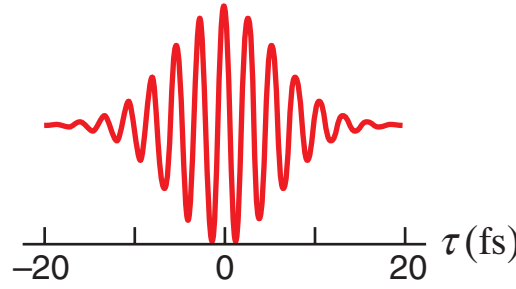
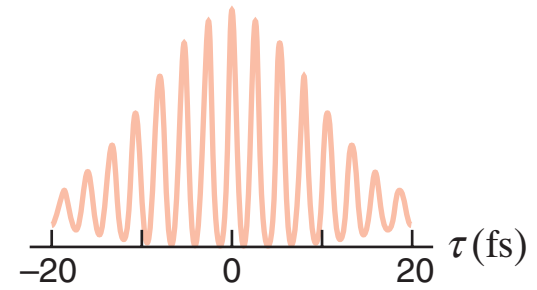
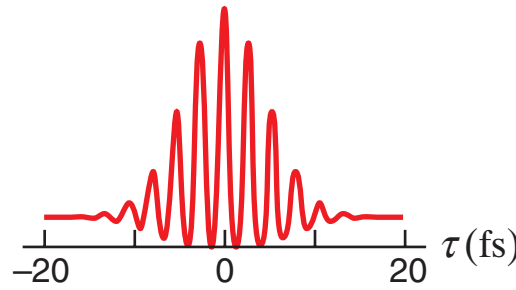
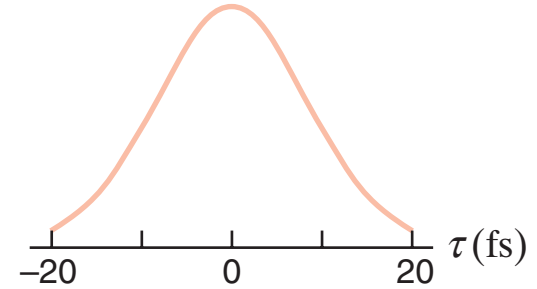
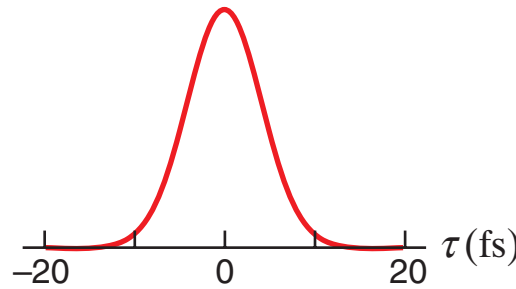
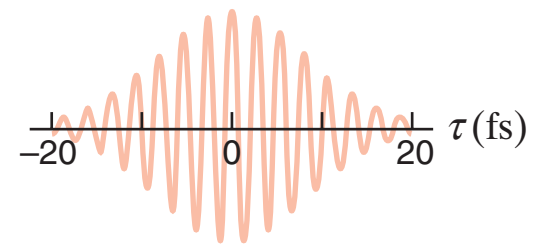
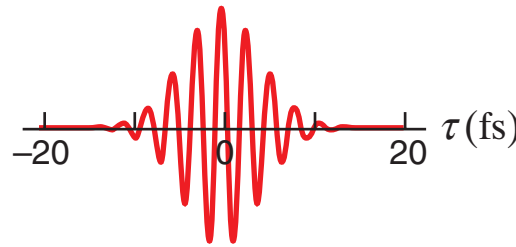
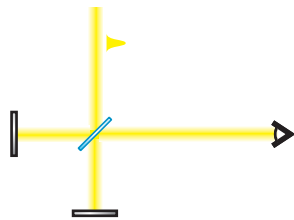
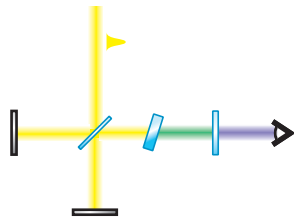
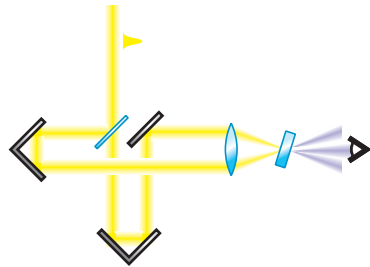
Temporal characterization



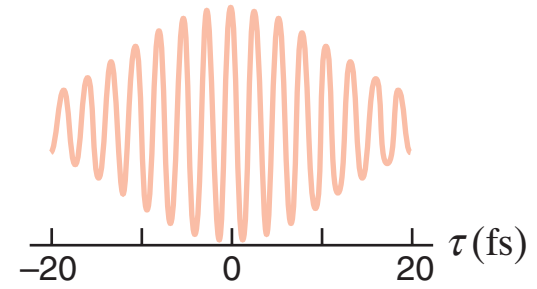
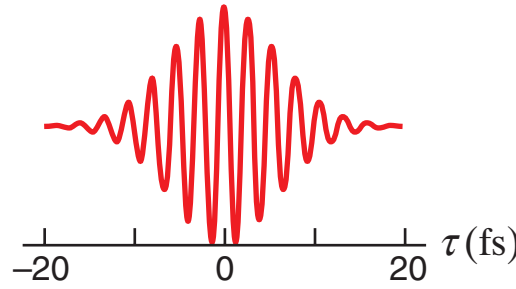
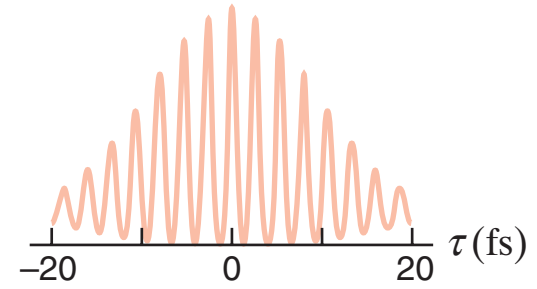
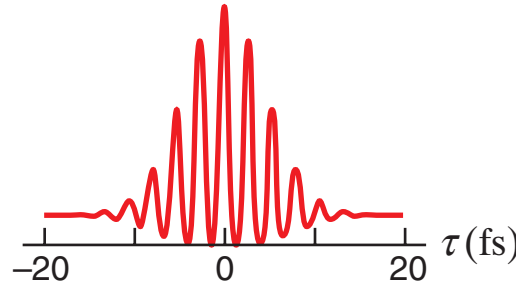
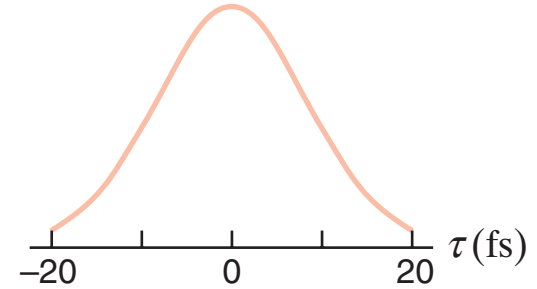
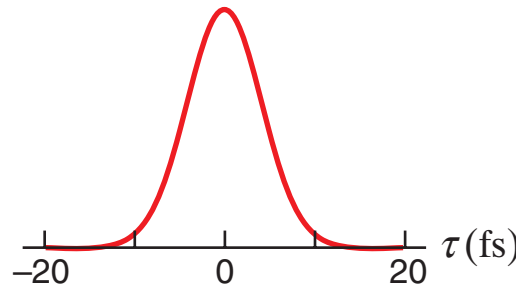
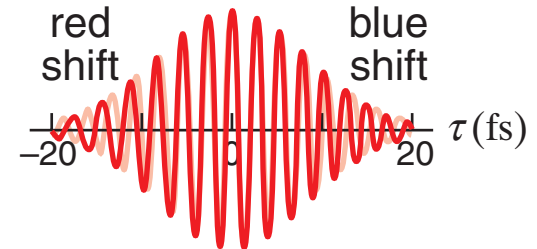
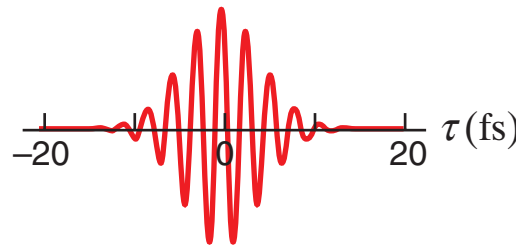
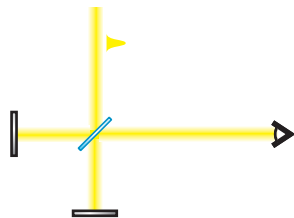
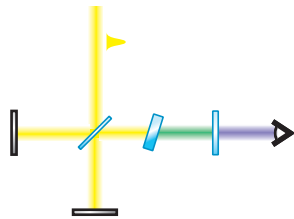
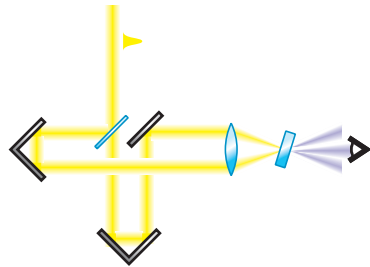
Temporal characterization

But what about dispersion?

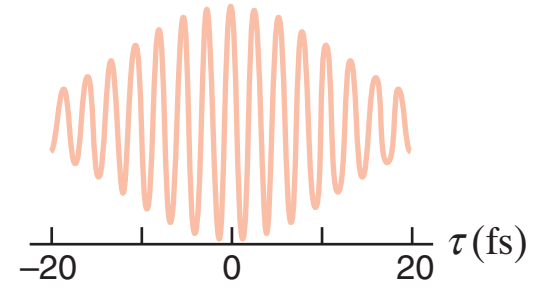
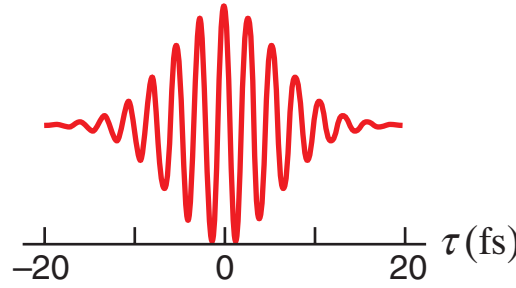
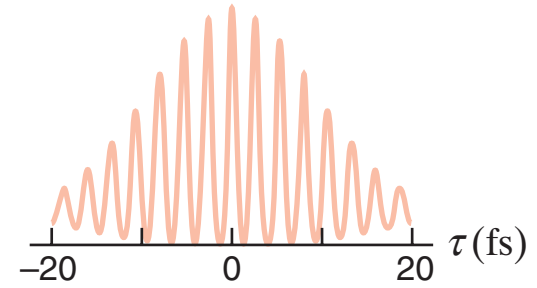
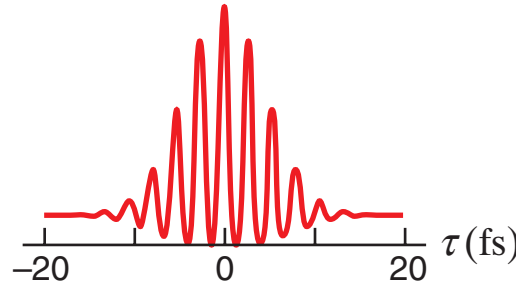
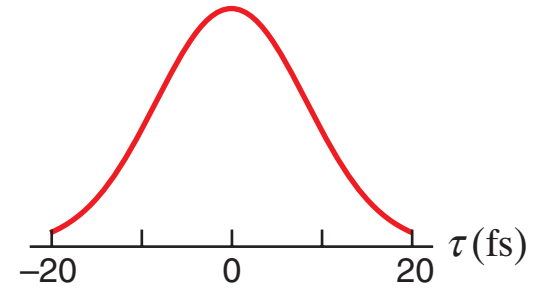
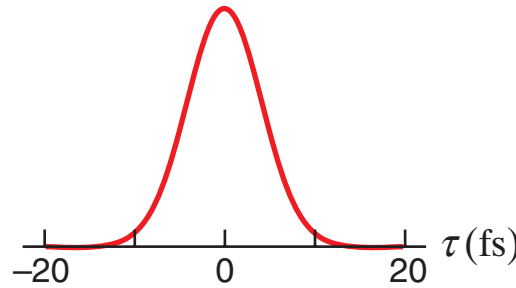
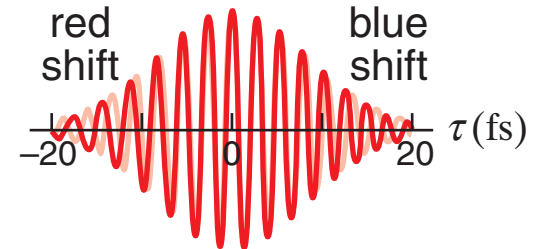
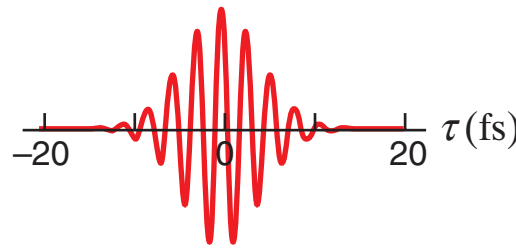
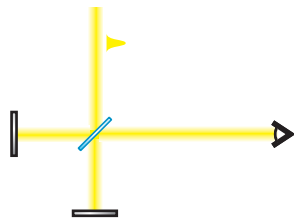
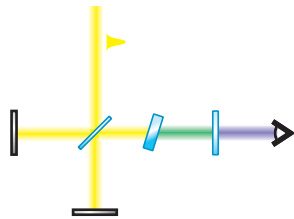
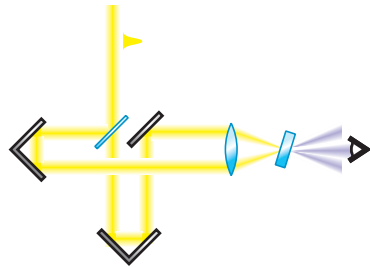
Temporal characterization



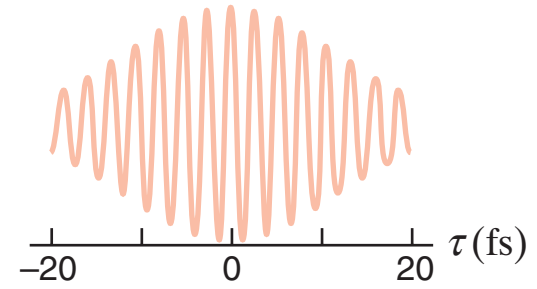
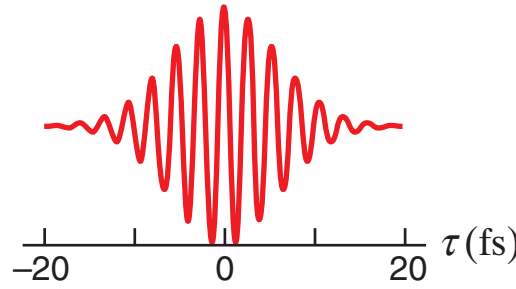
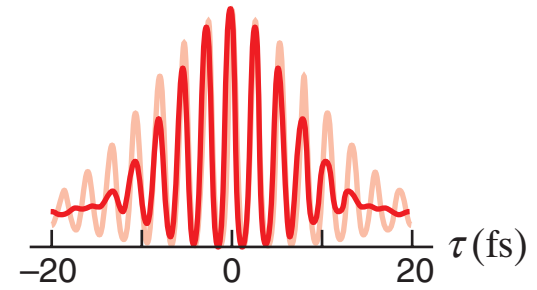
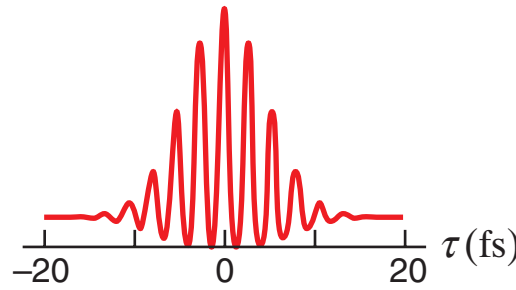
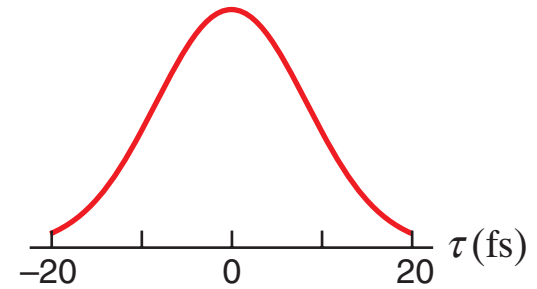
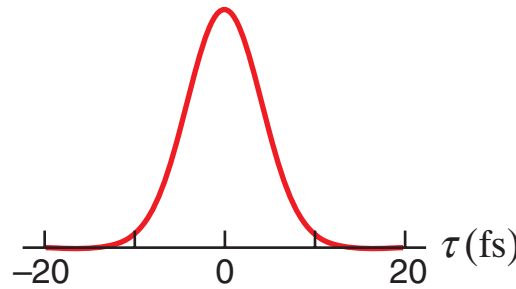
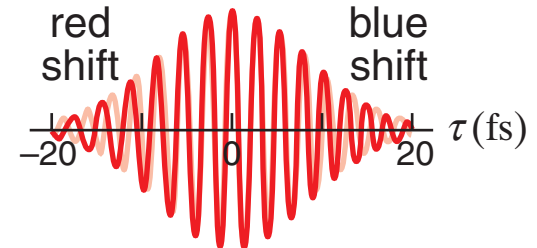
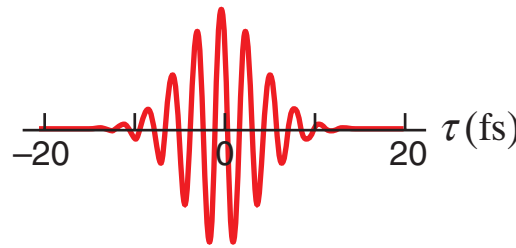
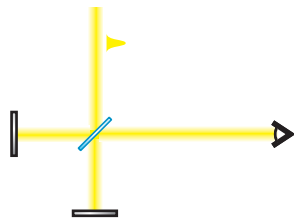
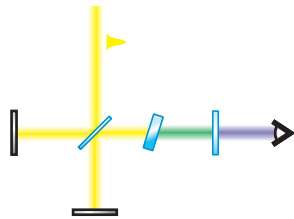
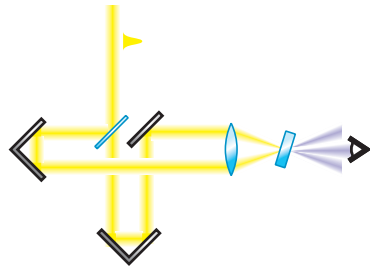
Temporal characterization



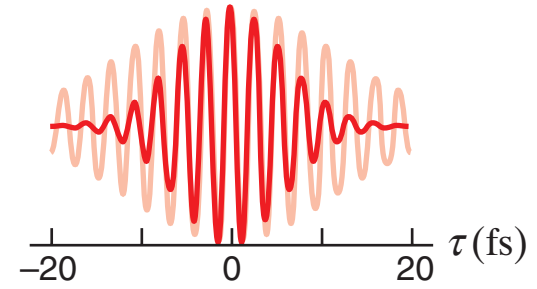
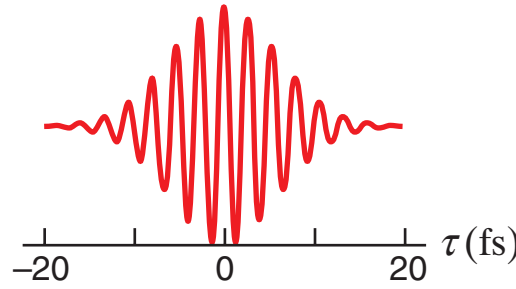
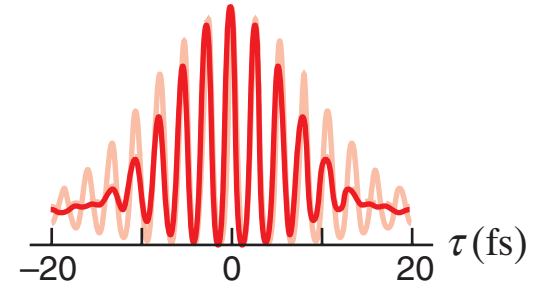
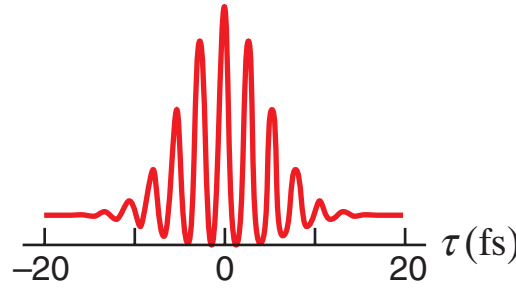
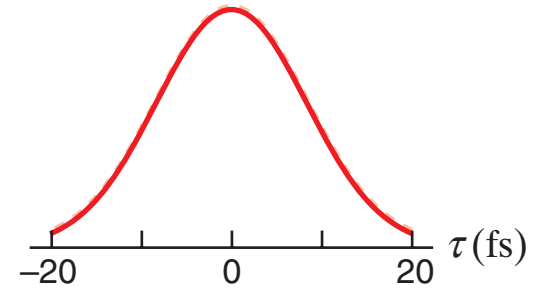
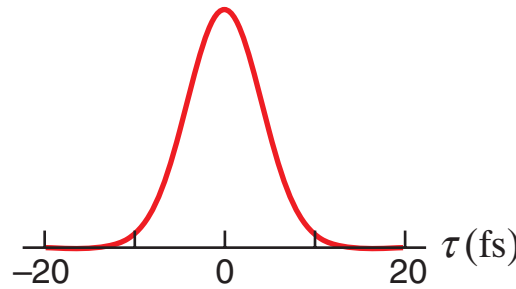
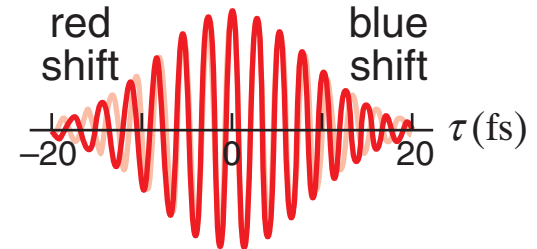
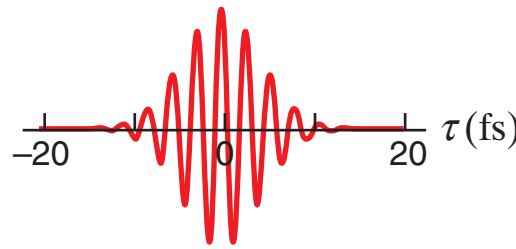
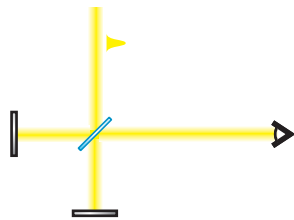
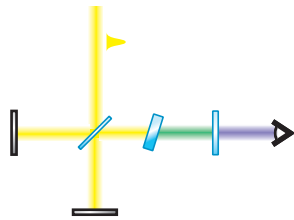
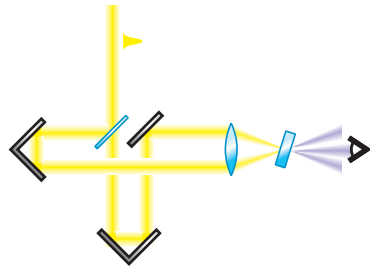
Temporal characterization



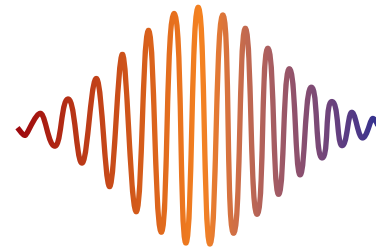
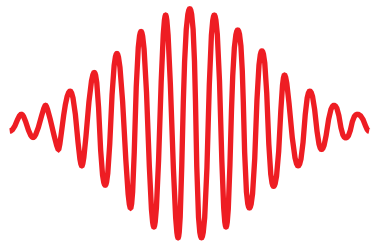
Temporal characterization



Temporal characterization

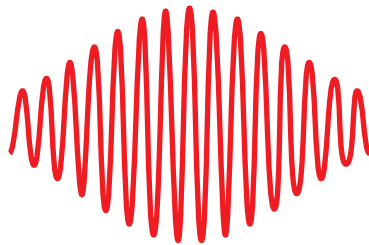
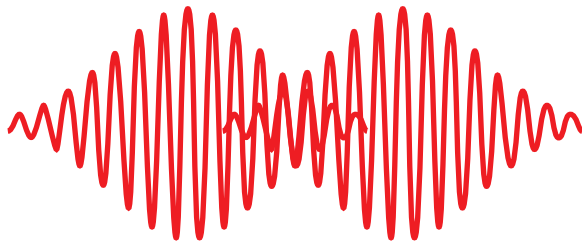


Temporal characterization

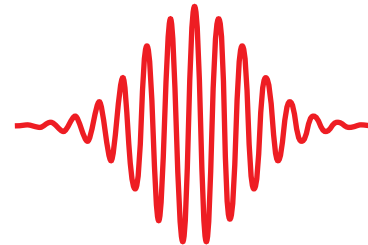
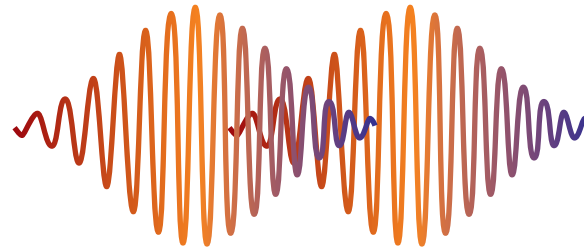


Temporal characterization

good
interference



poor
interference



Temporal characterization

Let $E_{disp}(\omega) = E_{orig}(\omega)e^{-i\phi(\omega)}$.

Temporal characterization

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Interference term in linear autocorrelation:

$$\int E_{disp}(t+\tau)E_{disp}^*(t) dt = \mathcal{F}^{-1}\{E_{disp}(\omega)E_{disp}^*(\omega)\} =$$

Temporal characterization

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Temporal characterization

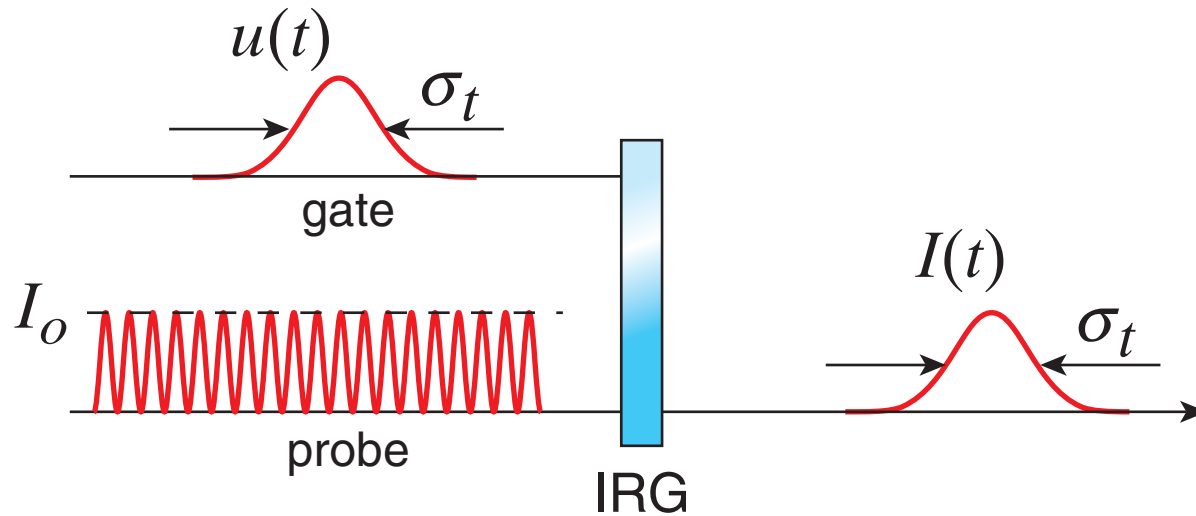
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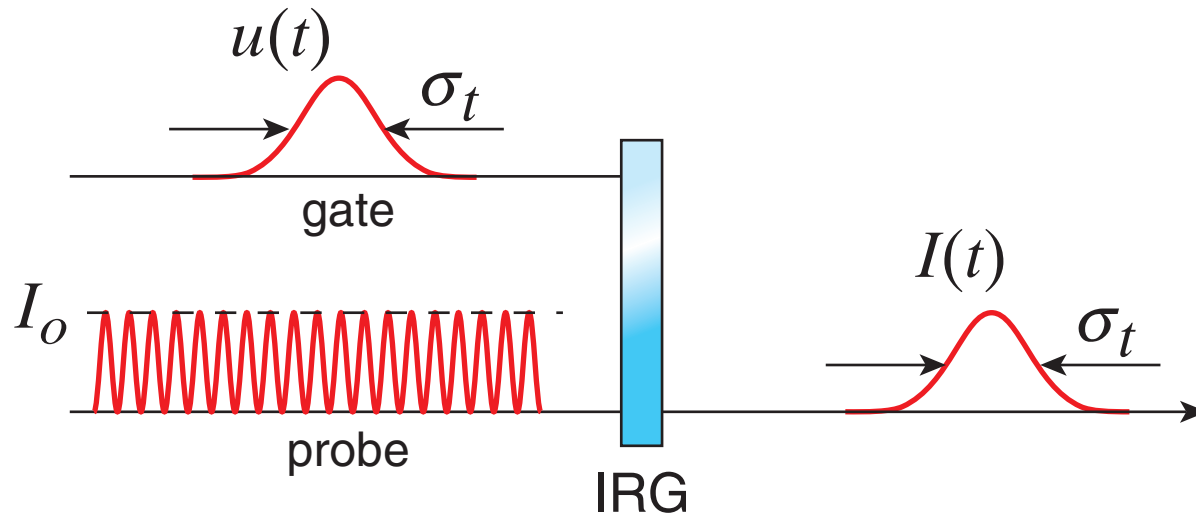
Joint time-frequency measurements



IRG ("instantaneous response gate"): device whose transmittance of a weak probe pulse is proportional to the intensity envelope of the pump ("gate")

$$T(t) = u(t)$$

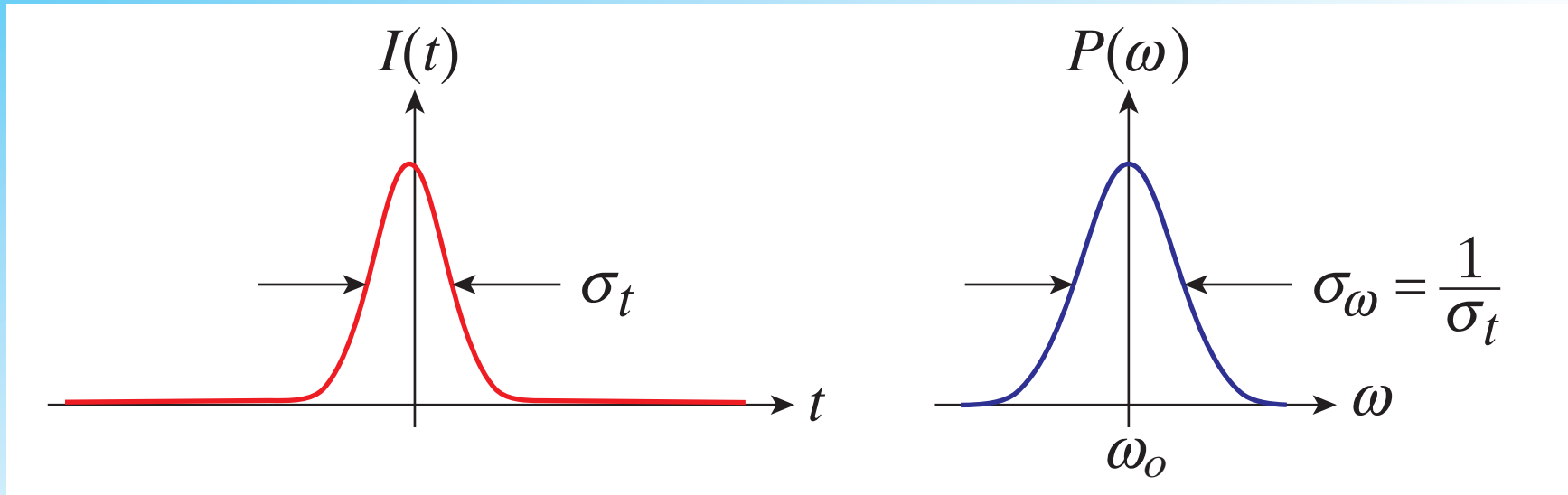
Joint time-frequency measurements



Transmitted intensity

$$I(t) = I_o T(t) = I_o u(t) = I_o \exp\left[-\frac{t^2}{\sigma_t^2}\right]$$

Joint time-frequency measurements

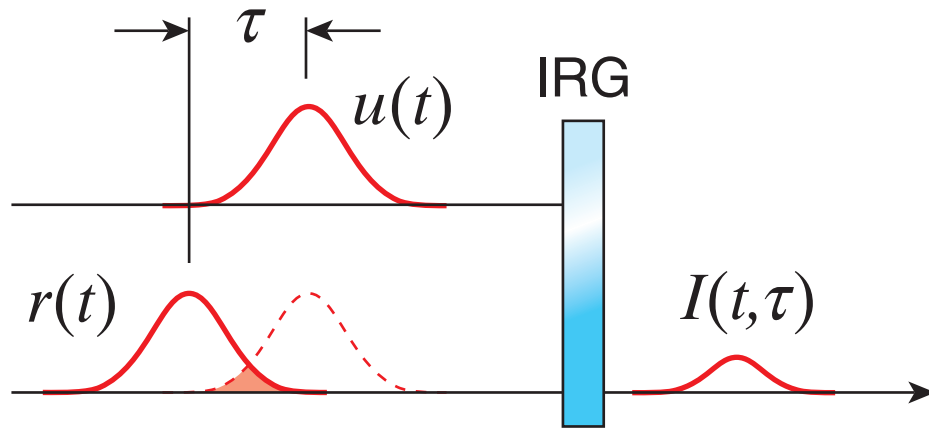


Transmitted intensity

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$$\sigma_t \sigma_\omega = 1$$

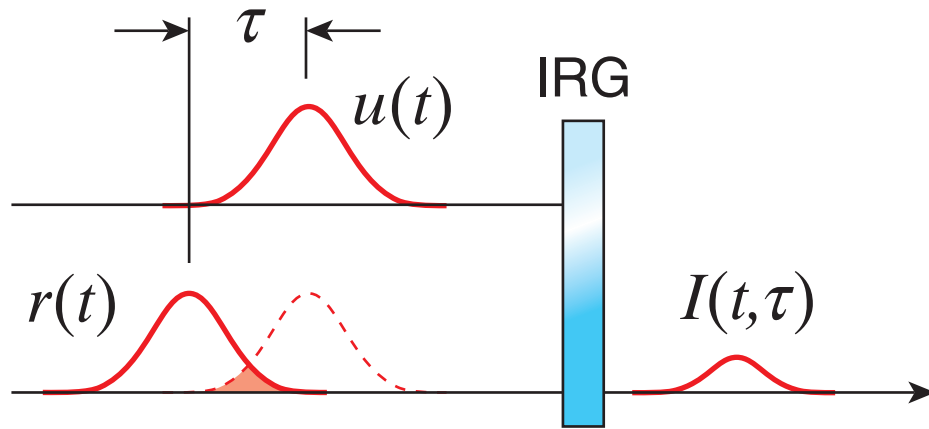
Joint time-frequency measurements



Transmitted intensity

$$\begin{aligned} I(t, \tau) &= u(t)r(t + \tau) = \exp\left[-\frac{t^2}{\sigma^2}\right] \exp\left[-\frac{(t + \tau)^2}{\sigma^2}\right] = \\ &= \exp\left[-\frac{2t^2 + 2t\tau + \tau^2}{\sigma^2}\right] = \exp\left[-\frac{2t^2 + 2t\tau + \tau^2/2}{\sigma^2}\right] \exp\left[-\frac{\tau^2}{2\sigma^2}\right] = \end{aligned}$$

Joint time-frequency measurements

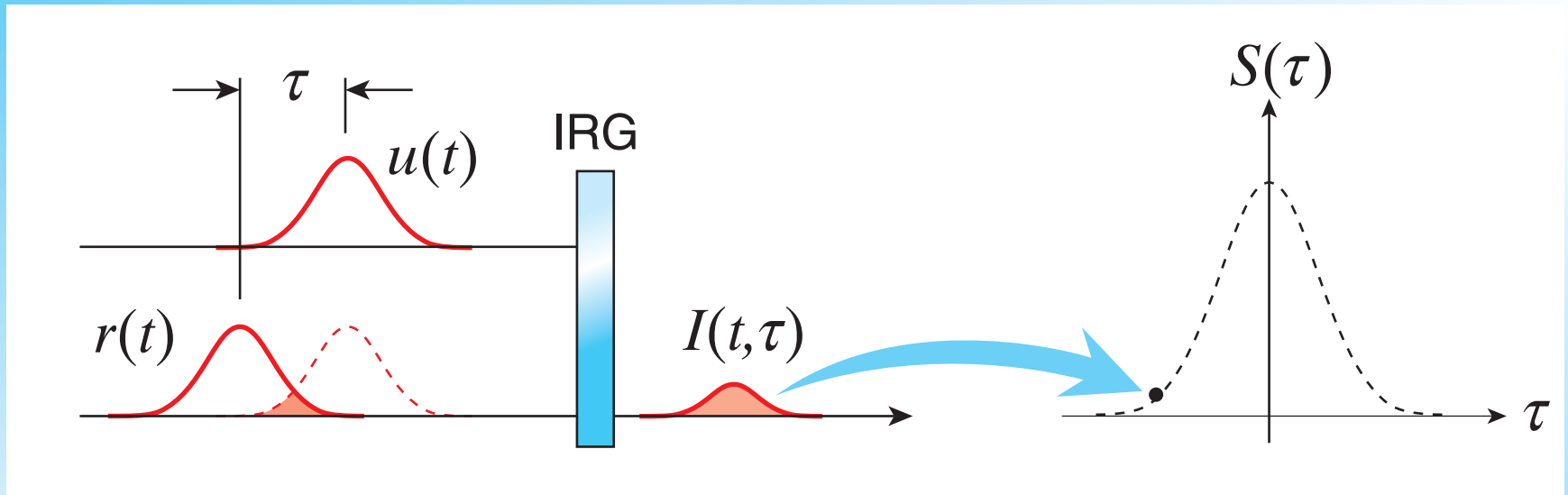


Transmitted intensity

$$I(t, \tau) = \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \exp\left[-\left(\frac{t + \tau/2}{\sigma/\sqrt{2}}\right)^2\right]$$

so $I(t, \tau)$ narrowed by $\sqrt{2}$

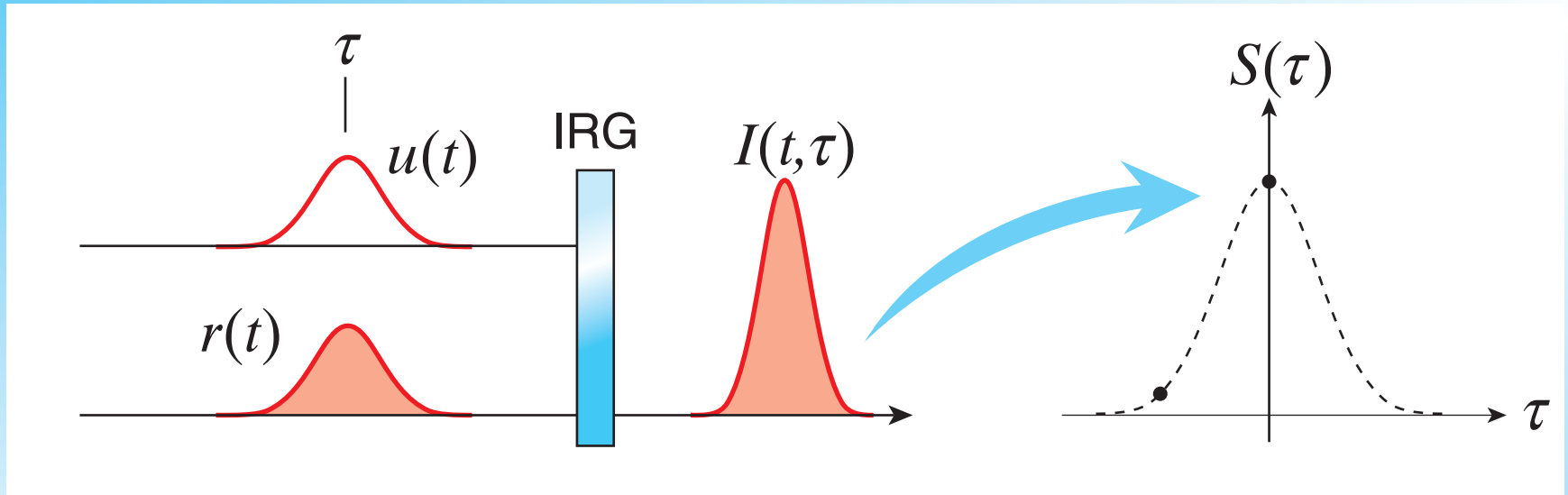
Joint time-frequency measurements



...but detector integrates $I(t, \tau)$:

$$S(\tau) = \int_{-\infty}^{\infty} I(t, \tau) dt = \int_{-\infty}^{\infty} \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \exp\left[-2\left(\frac{t + \tau/2}{\sigma}\right)^2\right] dt$$

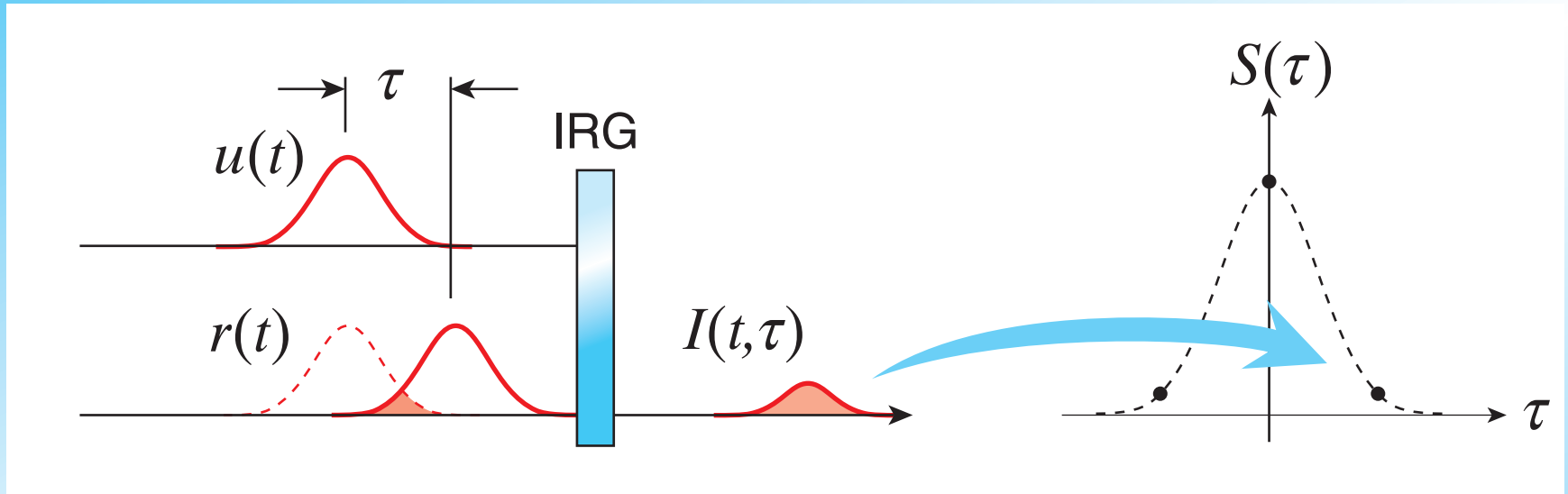
Joint time-frequency measurements



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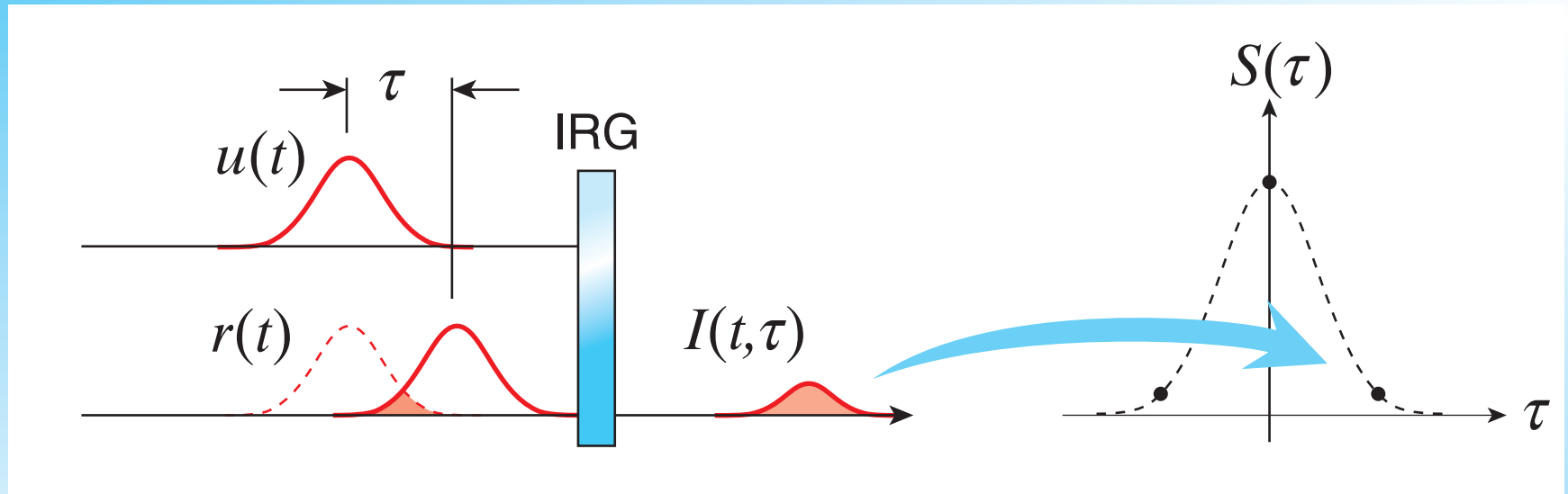
Joint time-frequency measurements



...but detector integrates $I(t, \tau)$:

$$S(\tau) = \int_{-\infty}^{\infty} I(t, \tau) dt = \int_{-\infty}^{\infty} \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \exp\left[-2\left(\frac{t + \tau/2}{\sigma}\right)^2\right] dt$$

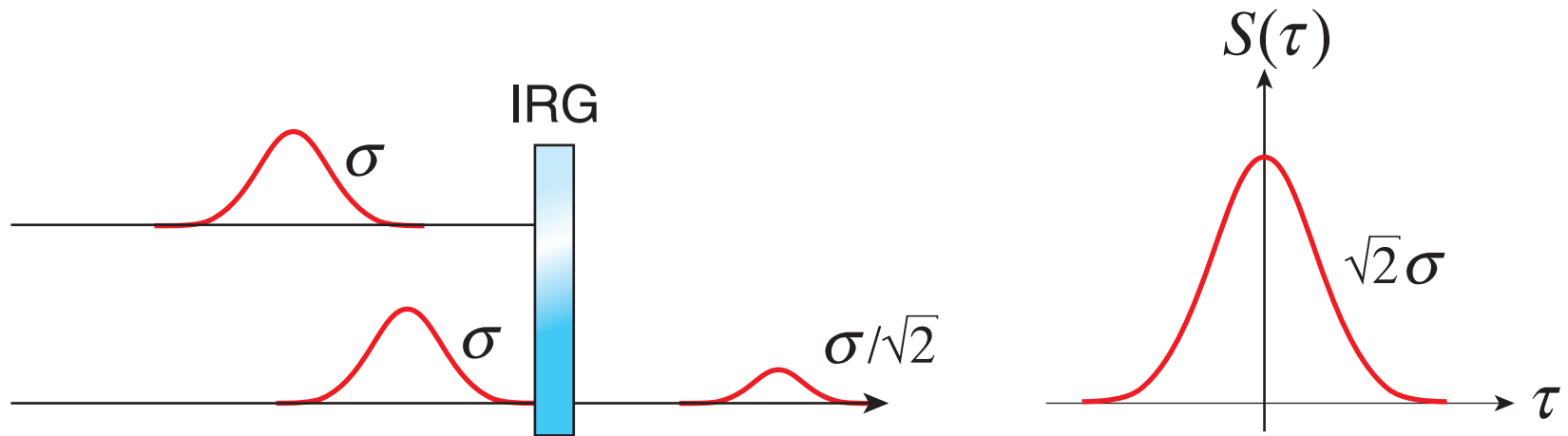
Joint time-frequency measurements



...but detector integrates $I(t, \tau)$:

$$S(\tau) = \int_{-\infty}^{\infty} I(t, \tau) dt = \int_{-\infty}^{\infty} \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \exp\left[-2\left(\frac{t + \tau/2}{\sigma}\right)^2\right] dt$$
$$= \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \int_{-\infty}^{\infty} \exp\left[-\frac{2u^2}{\sigma^2}\right] du = \sqrt{\frac{\pi}{2}} \sigma \exp\left[-\frac{\tau^2}{(\sigma\sqrt{2})^2}\right]$$

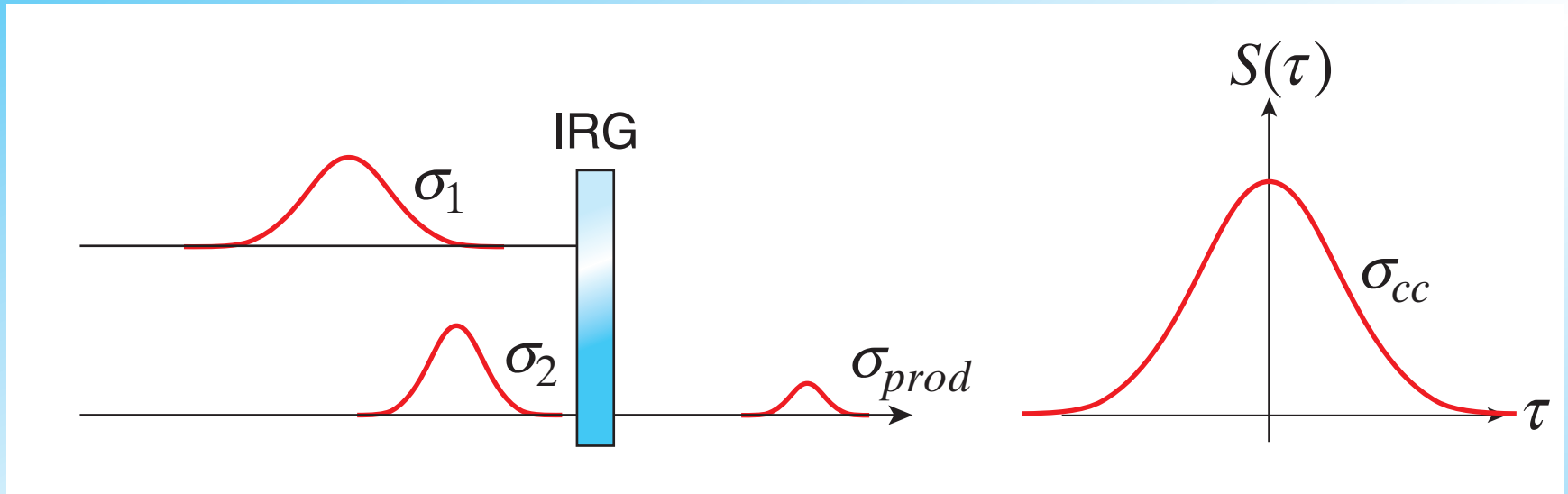
Joint time-frequency measurements



...but detector integrates $I(t, \tau)$:

$$\begin{aligned} S(\tau) &= \int_{-\infty}^{\infty} I(t, \tau) dt = \int_{-\infty}^{\infty} \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \exp\left[-2\left(\frac{t + \tau/2}{\sigma}\right)^2\right] dt \\ &= \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \int_{-\infty}^{\infty} \exp\left[-\frac{2u^2}{\sigma^2}\right] du = \sqrt{\frac{\pi}{2}} \sigma \exp\left[-\frac{\tau^2}{(\sigma\sqrt{2})^2}\right] \end{aligned}$$

Joint time-frequency measurements

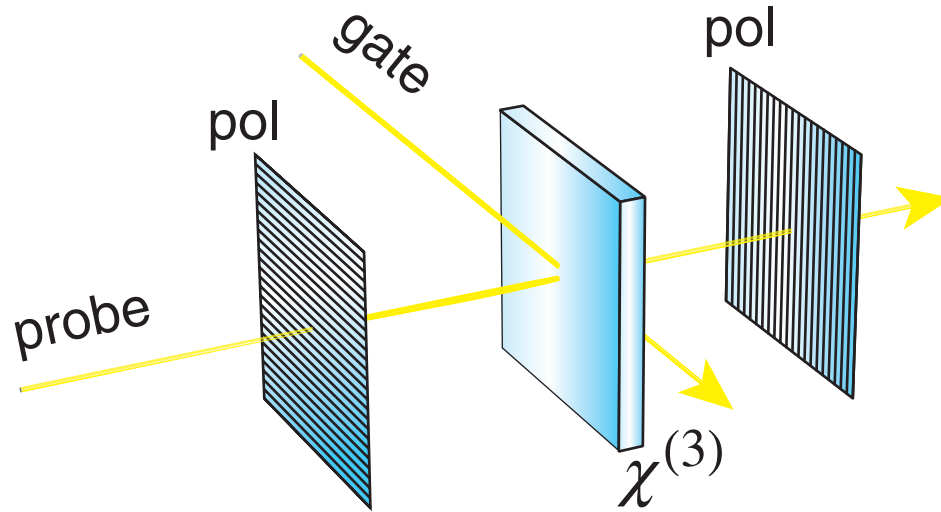


If gate and probe unequal:

$$\sigma_{prod}^2 = \frac{\sigma_1^2 \sigma_2^2}{\sigma_1^2 + \sigma_2^2} \quad \text{(narrower than both)}$$

$$\sigma_{cc}^2 = \sigma_1^2 + \sigma_2^2 \quad \text{(wider than both)}$$

Joint time-frequency measurements

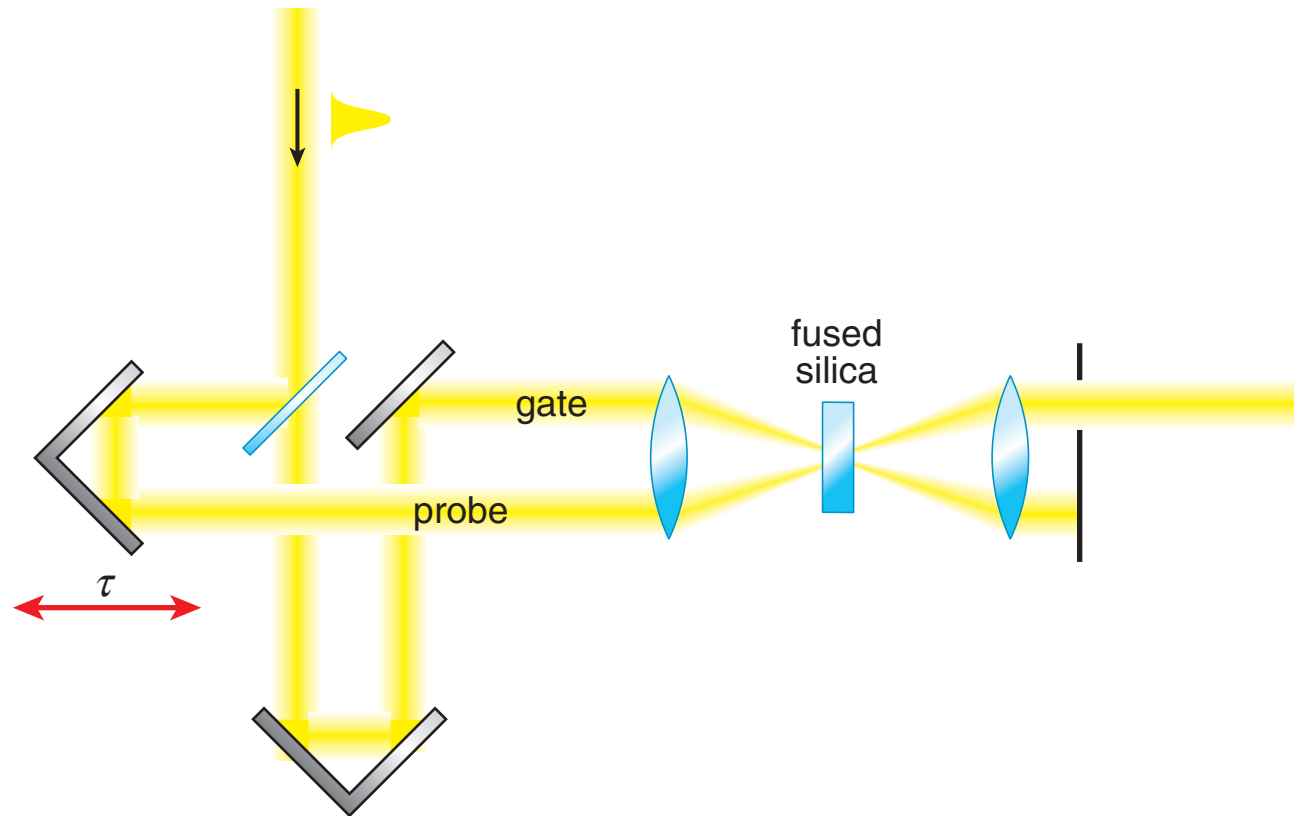


Transmitted field:

$$E_{trans}(t, \tau) \propto \chi^{(3)} E_{probe}(t) |E_{gate}(t + \tau)|^2$$

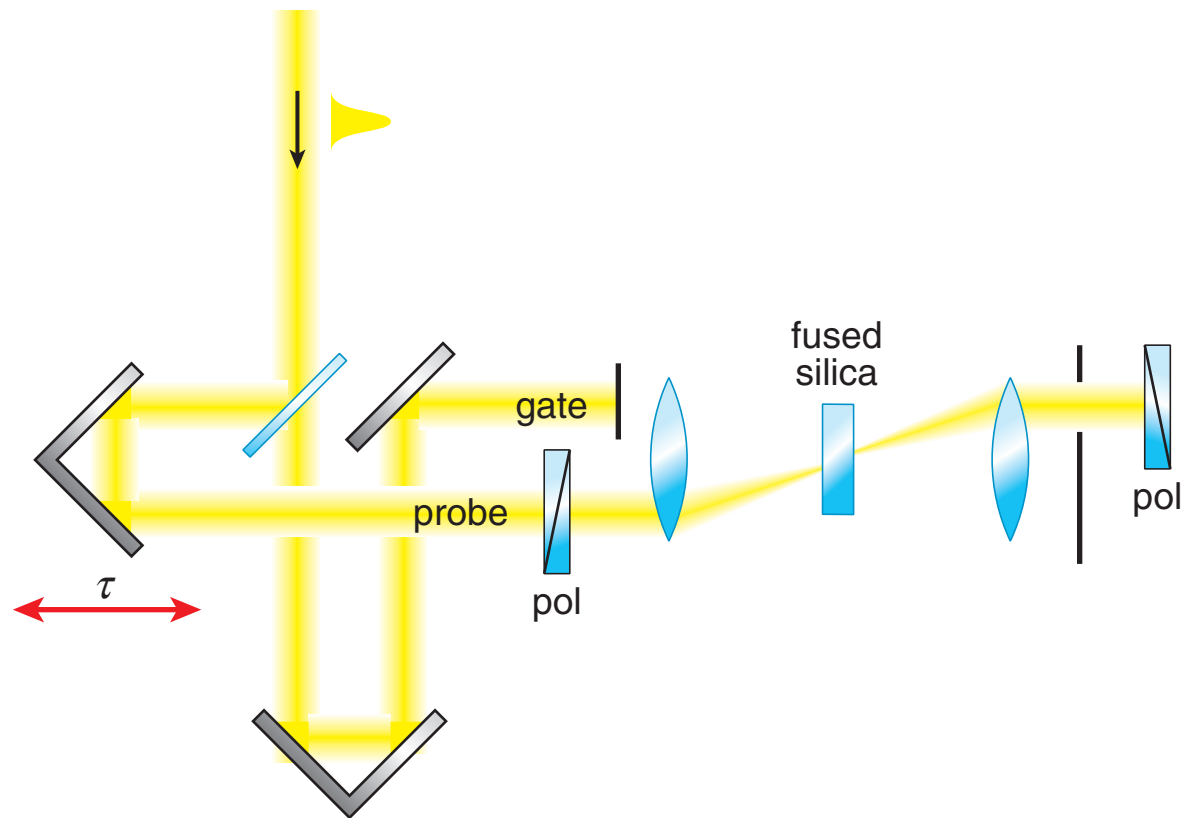
Joint time-frequency measurements

FROG: frequency-resolved optical gating



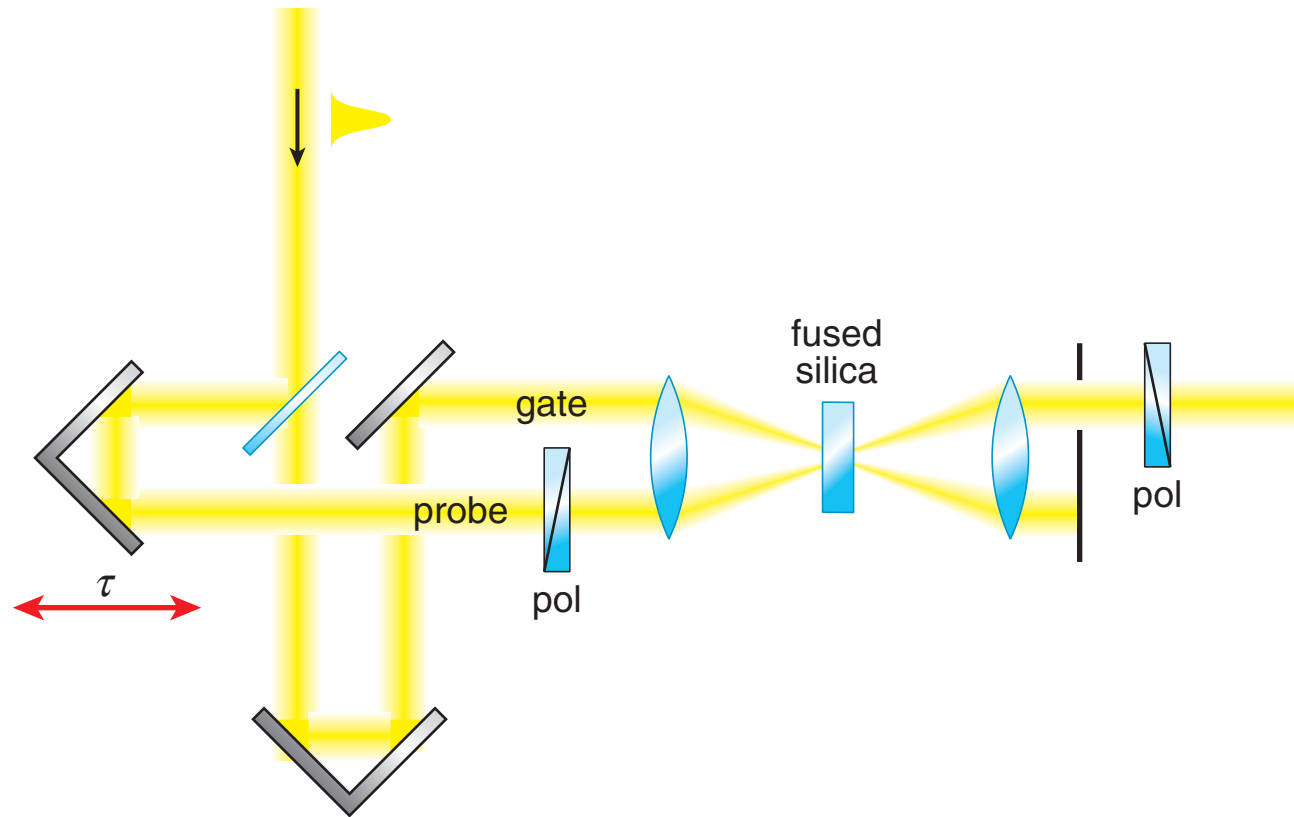
Joint time-frequency measurements

FROG: frequency-resolved optical gating



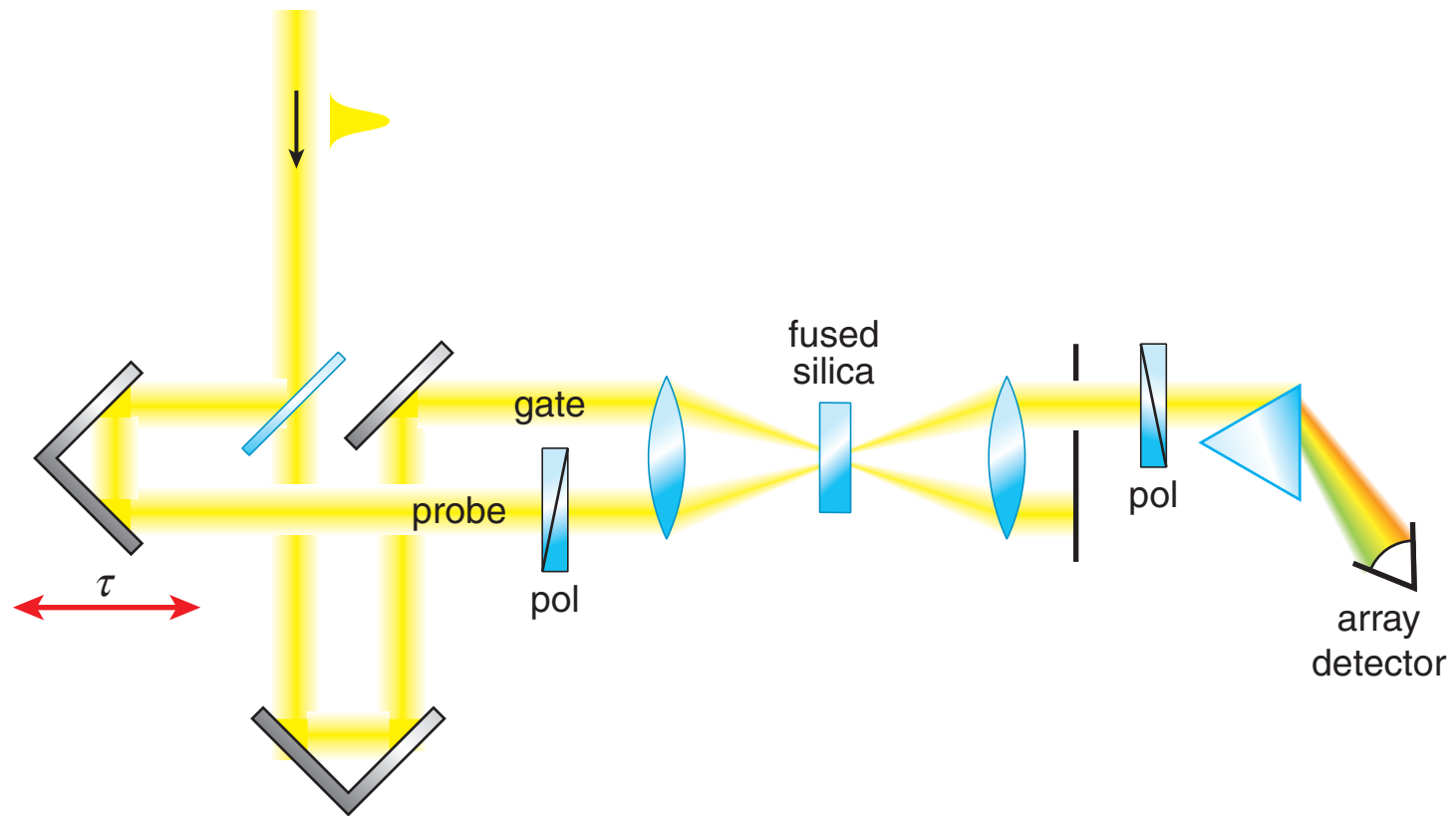
Joint time-frequency measurements

FROG: frequency-resolved optical gating

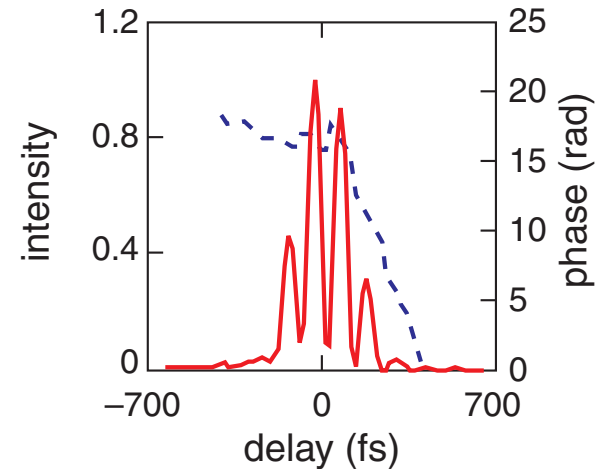
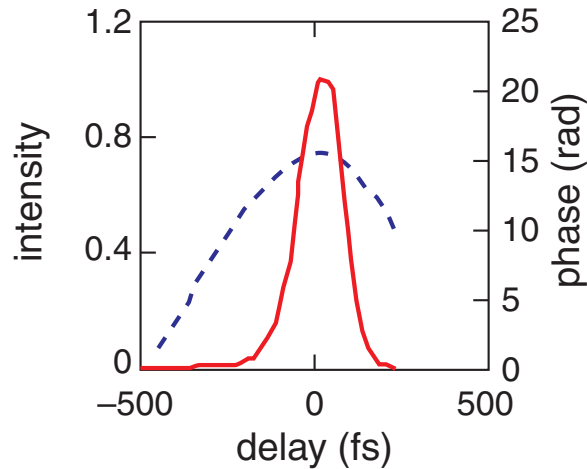
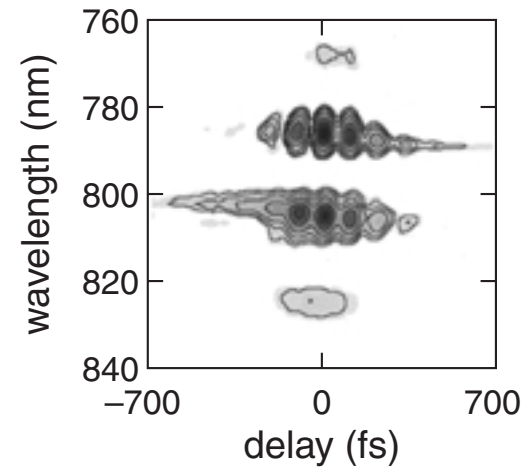
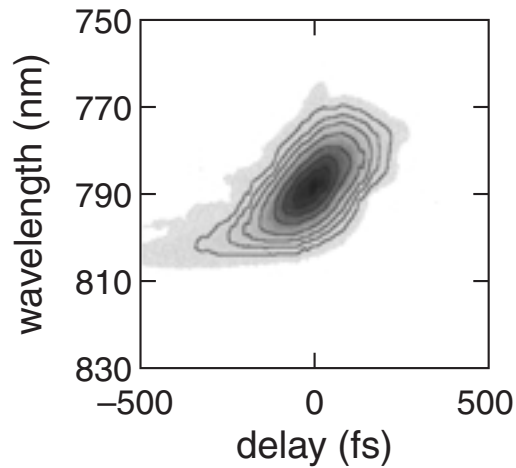


Joint time-frequency measurements

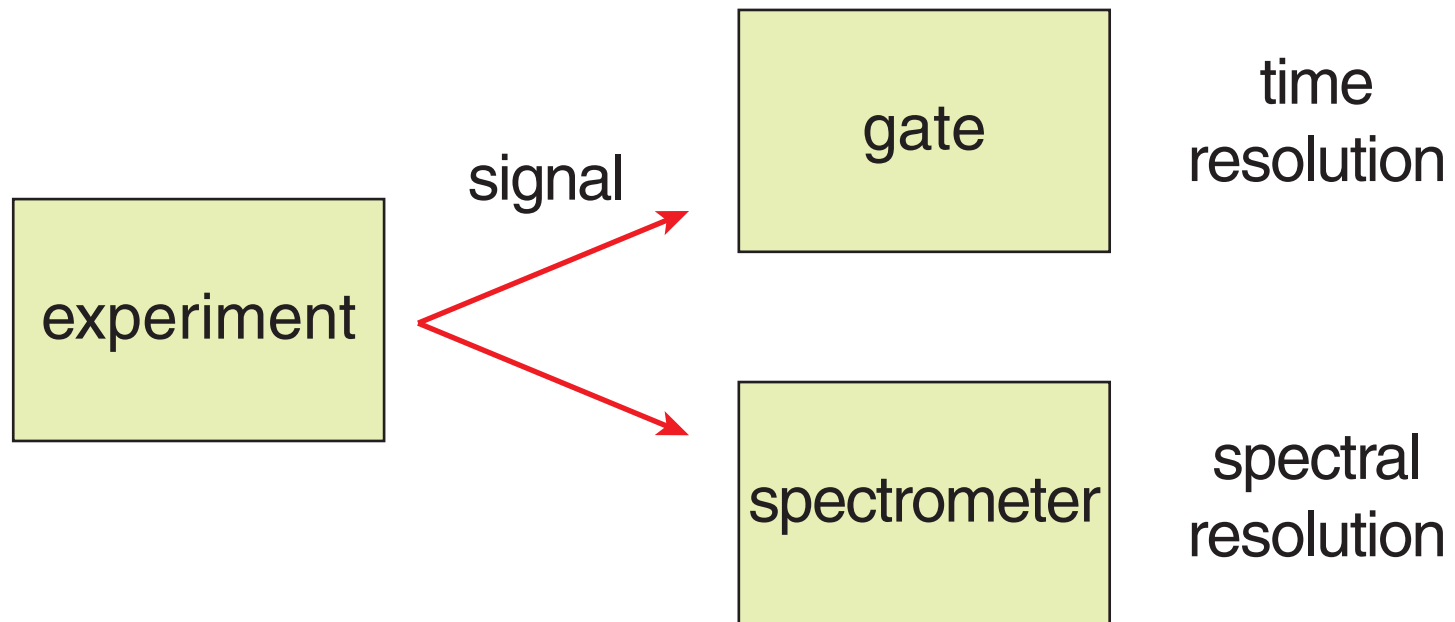
FROG: frequency-resolved optical gating



Joint time-frequency measurements



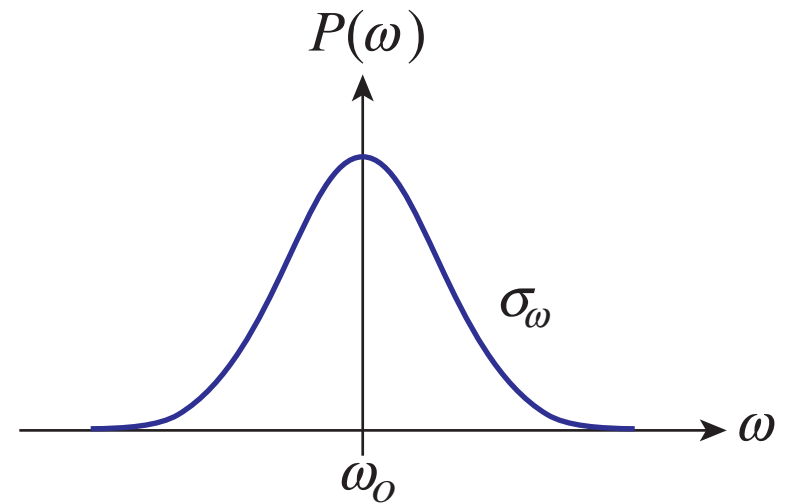
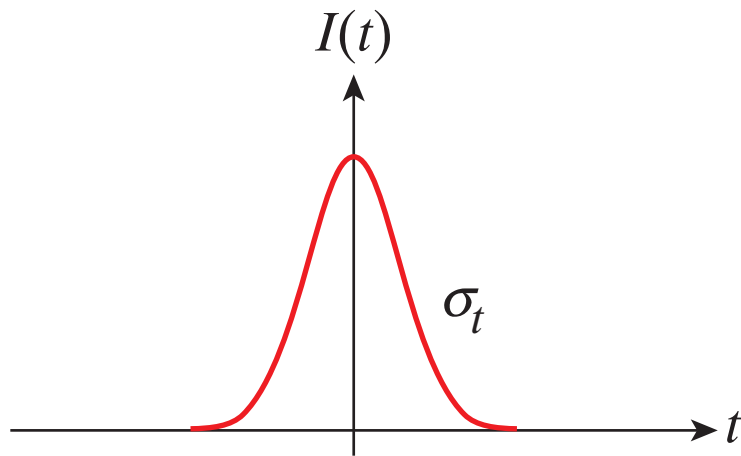
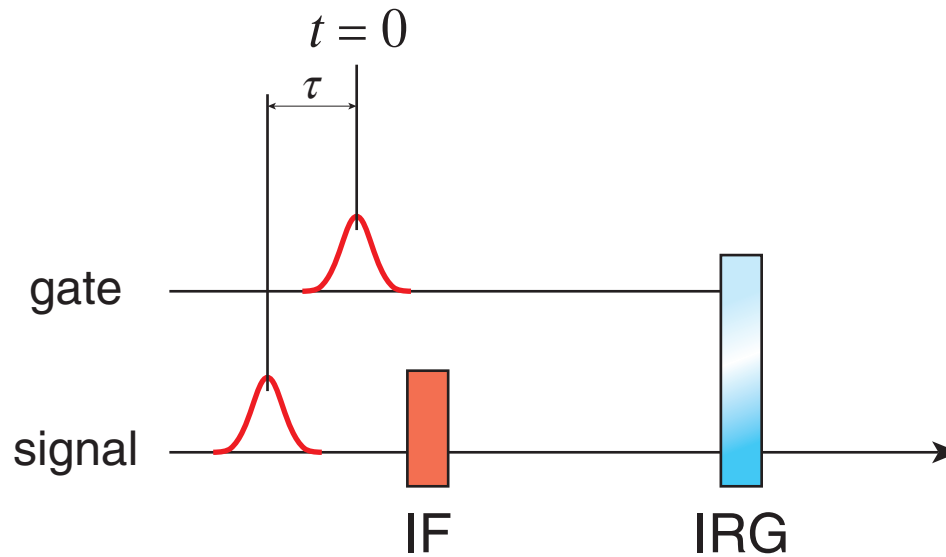
Joint time-frequency measurements



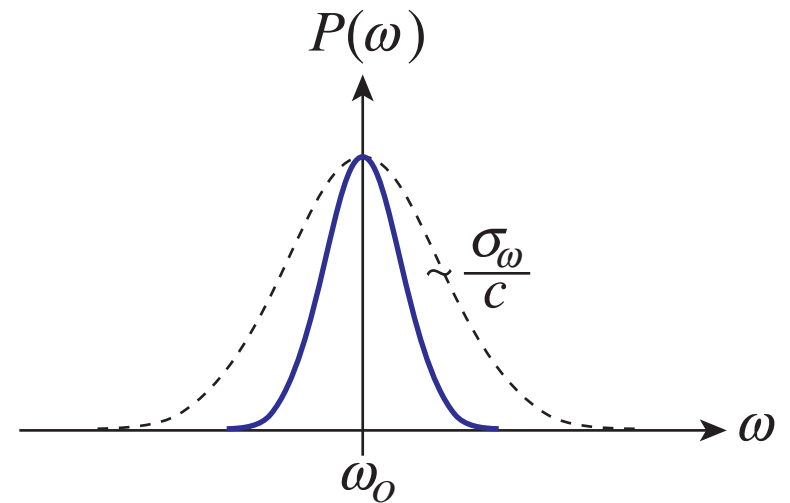
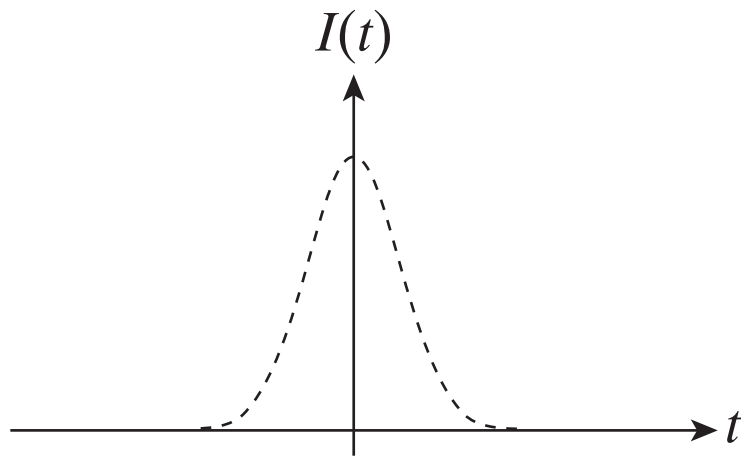
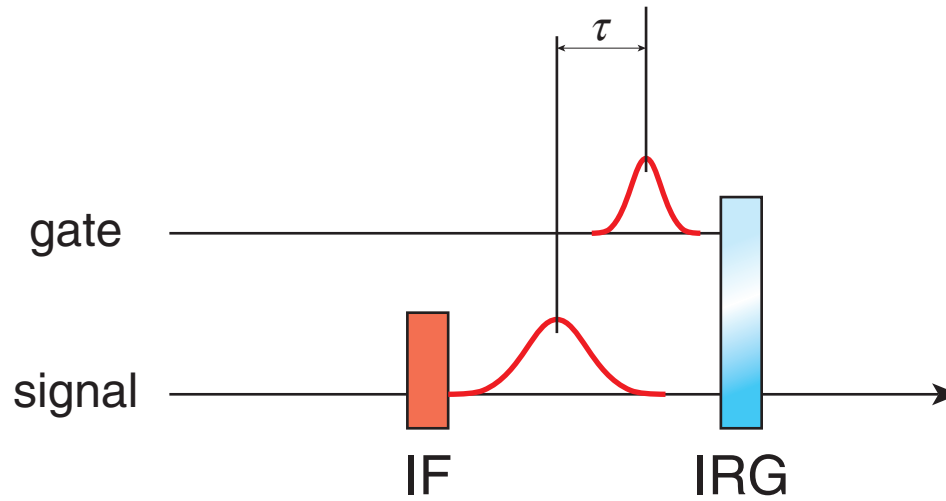
Joint time-frequency measurements

What are the resolution limits?

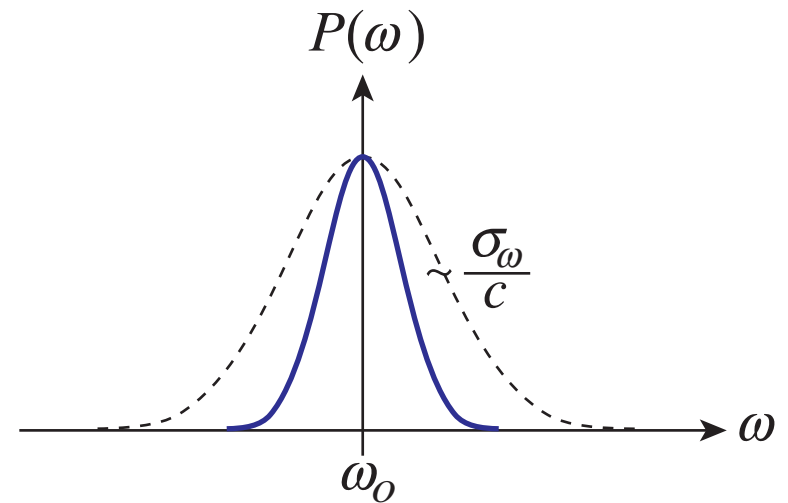
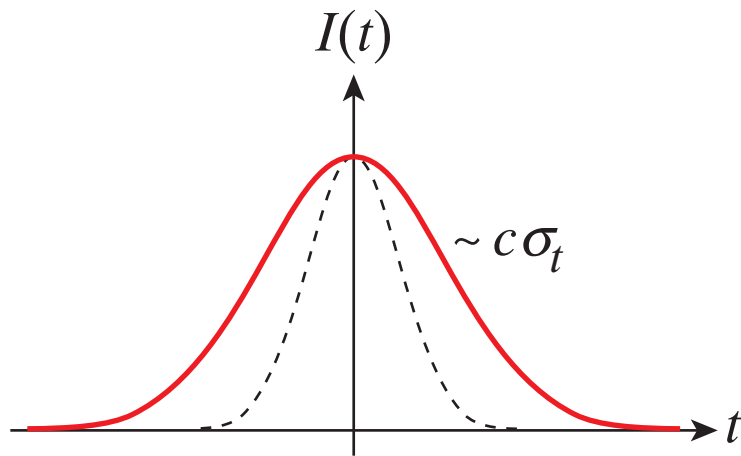
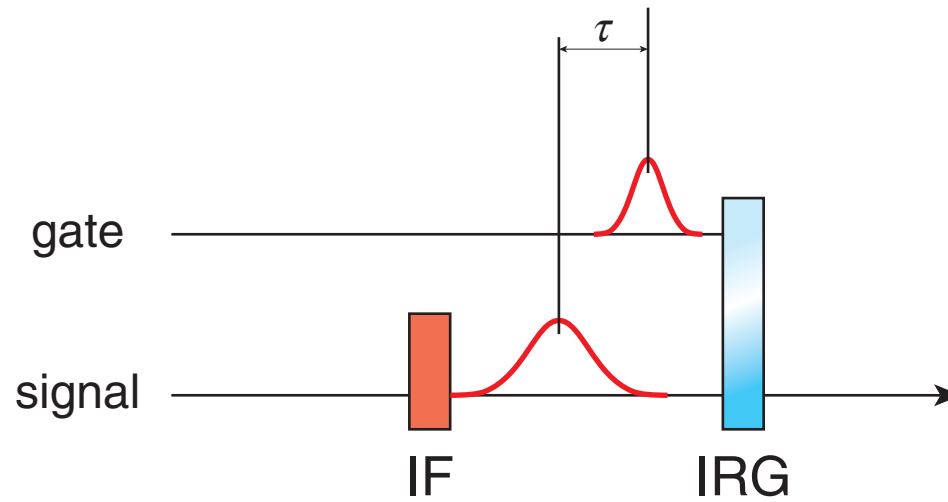
Experiment 1



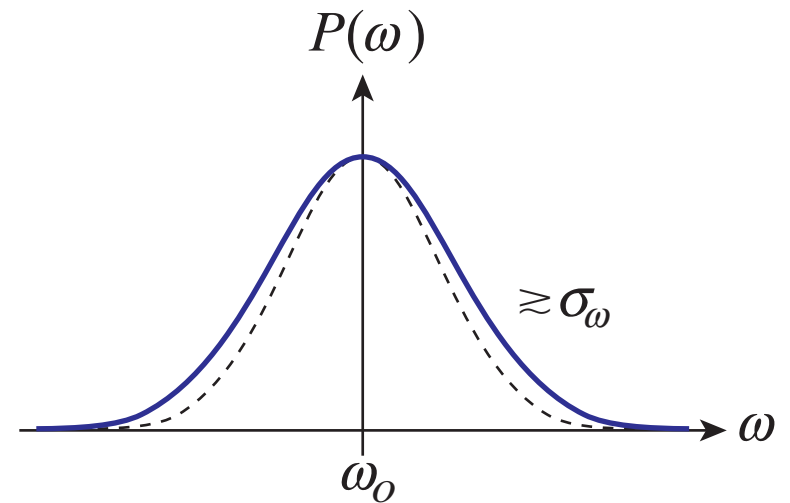
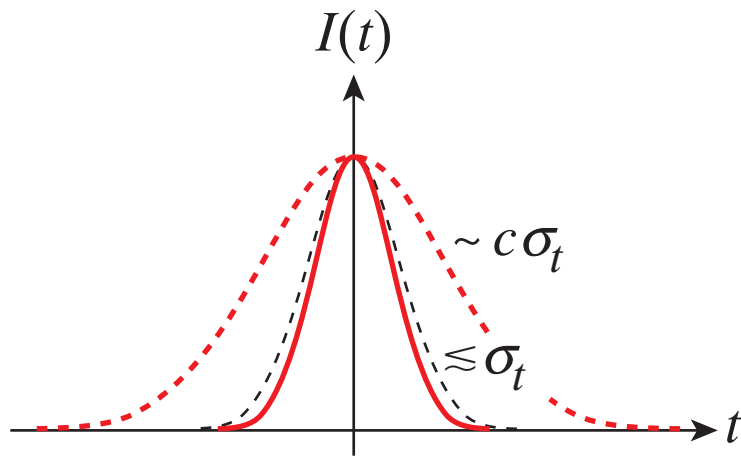
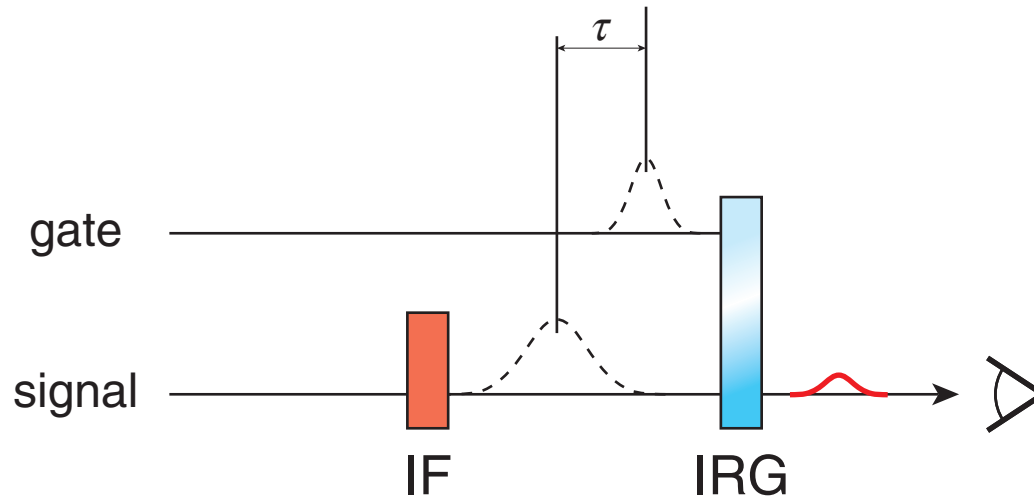
Experiment 1



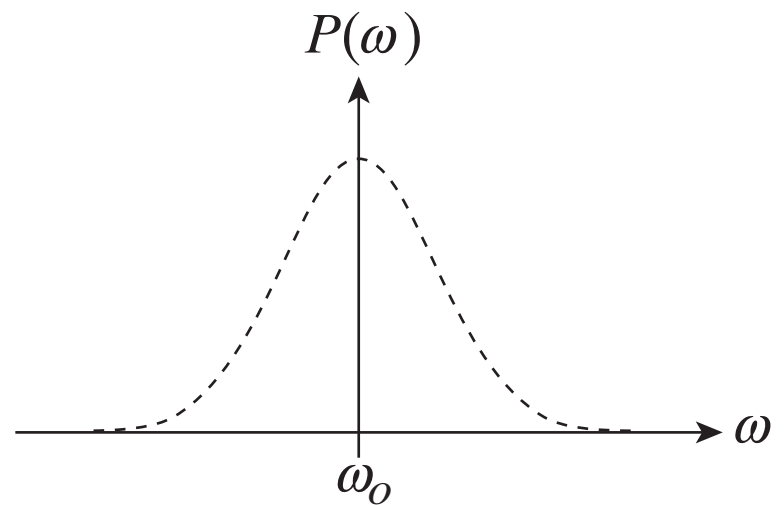
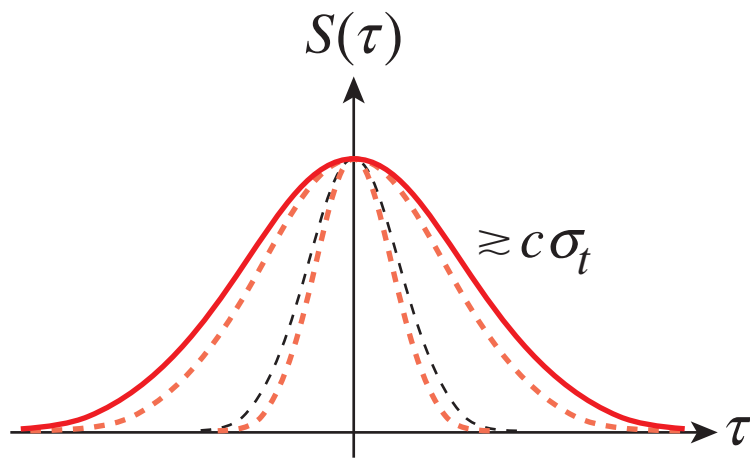
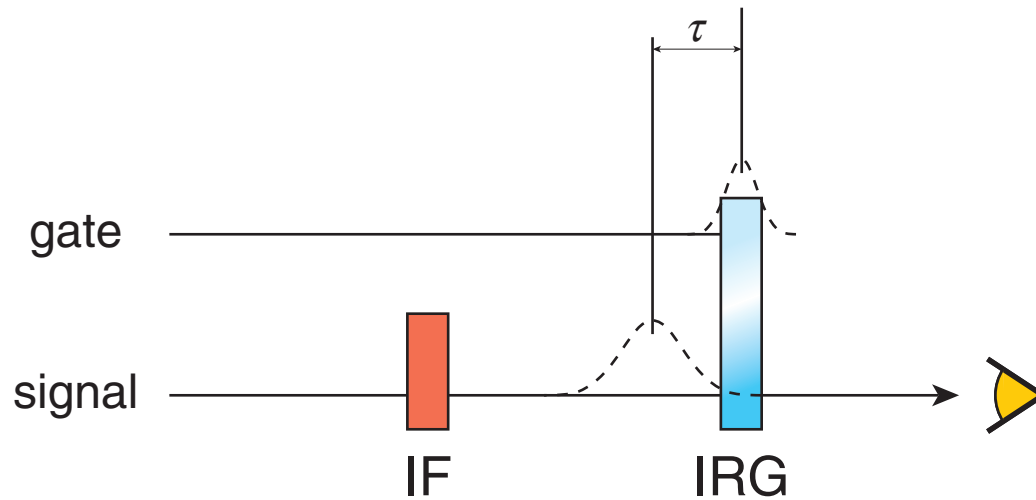
Experiment 1



Experiment 1

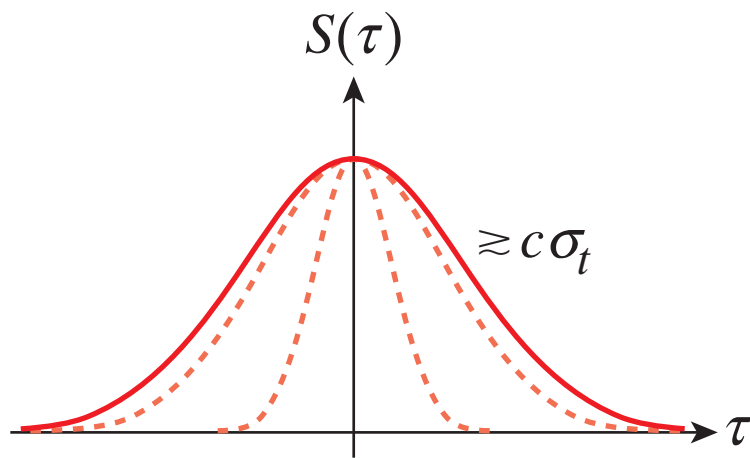
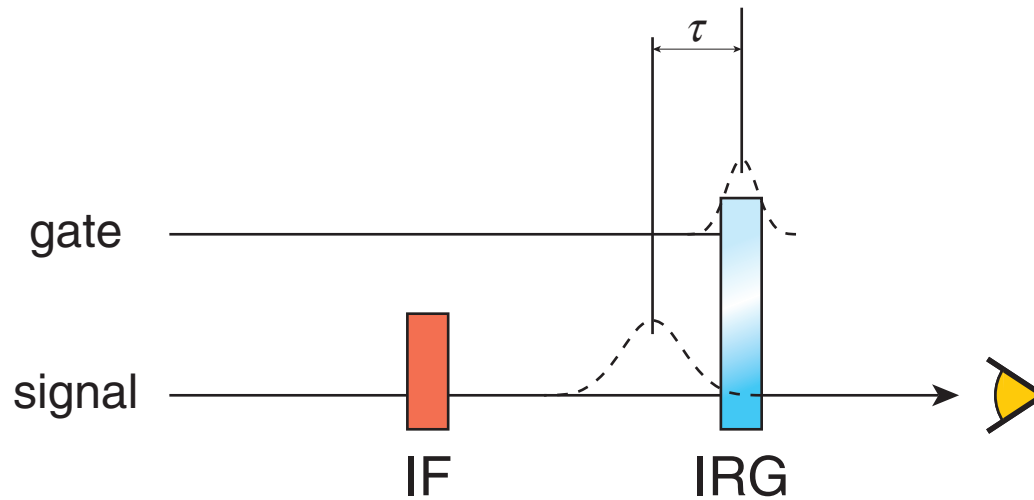


Experiment 1

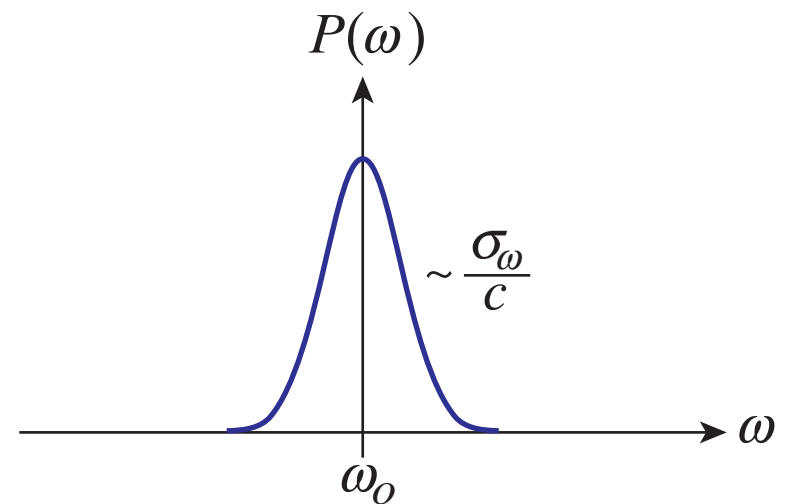


time resolution

Experiment 1

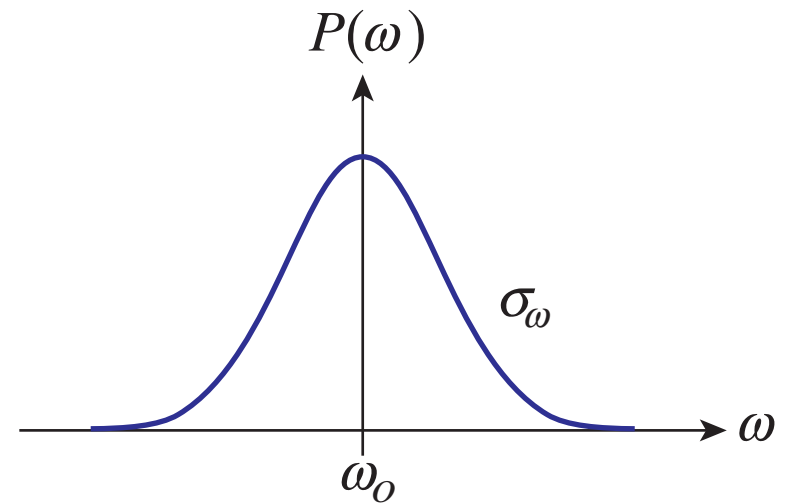
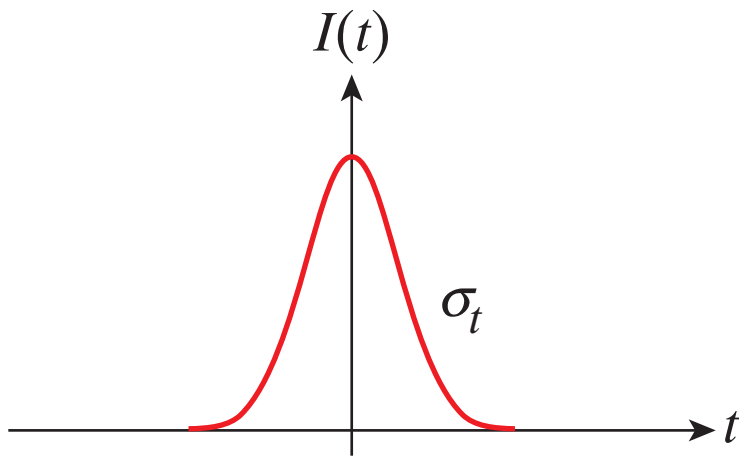
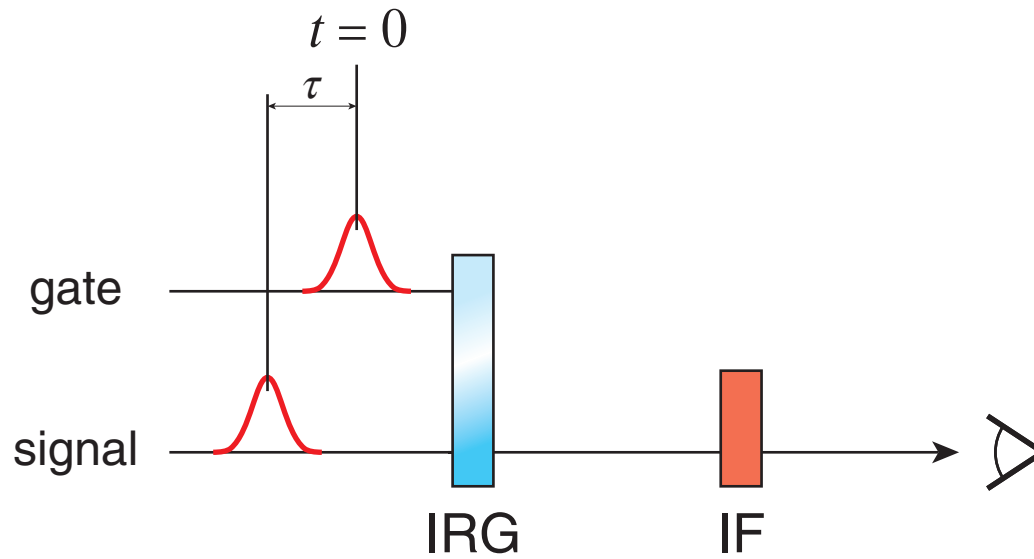


time resolution

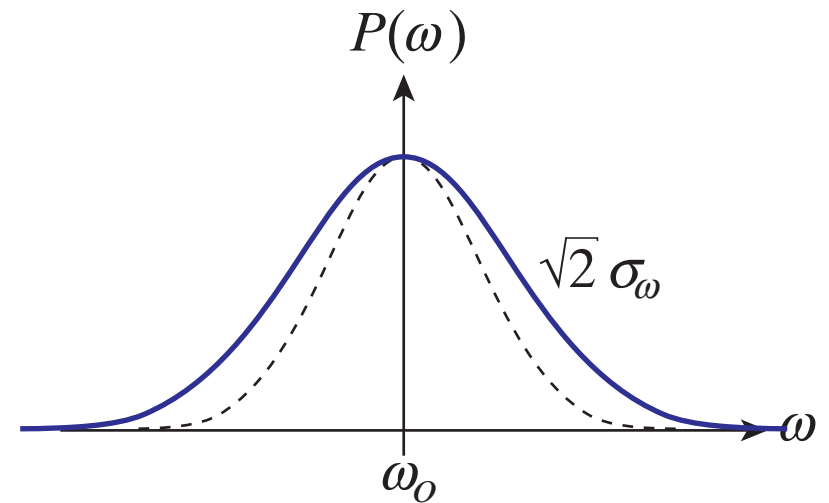
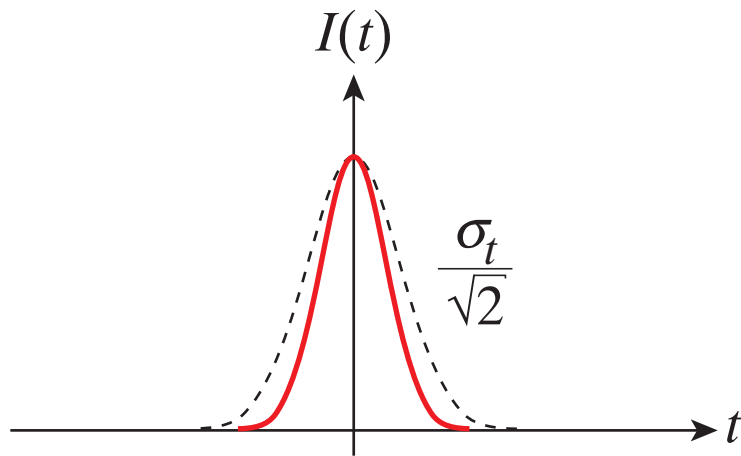
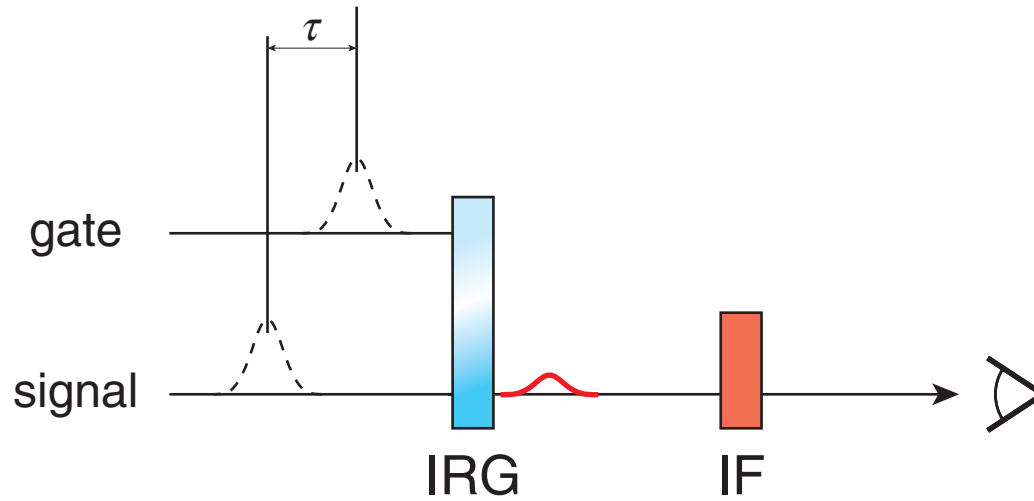


frequency resolution

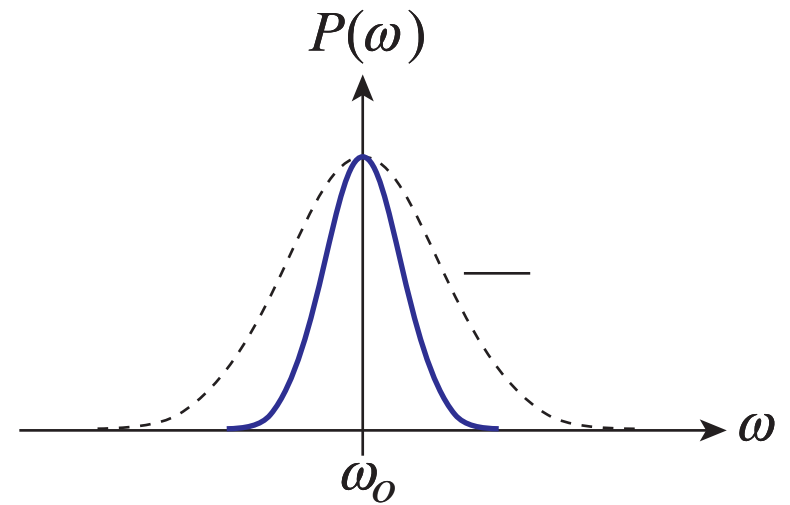
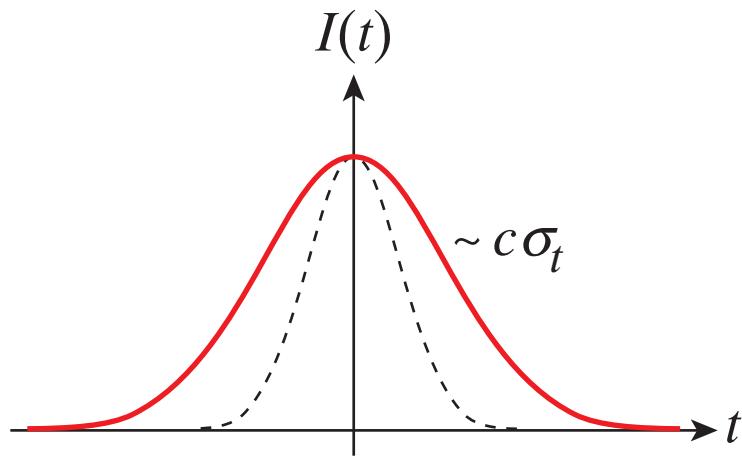
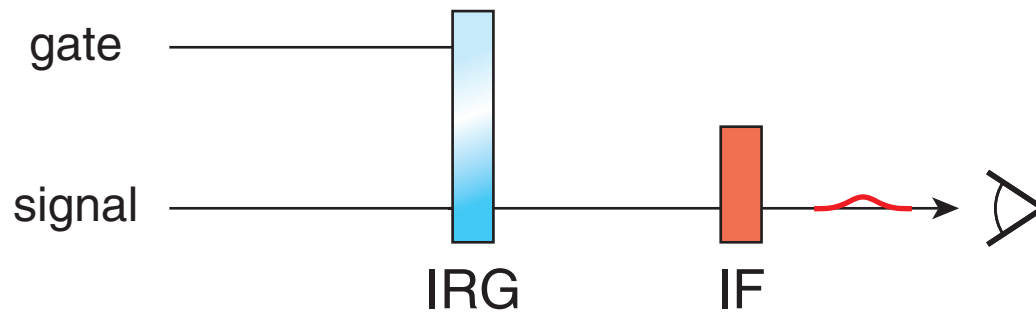
Experiment 2



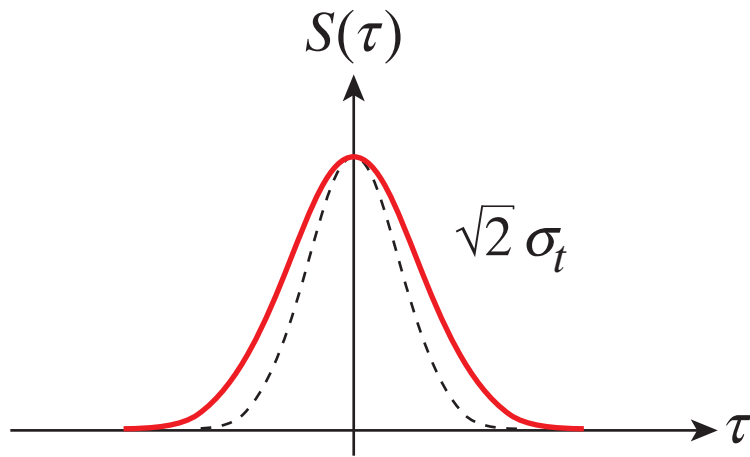
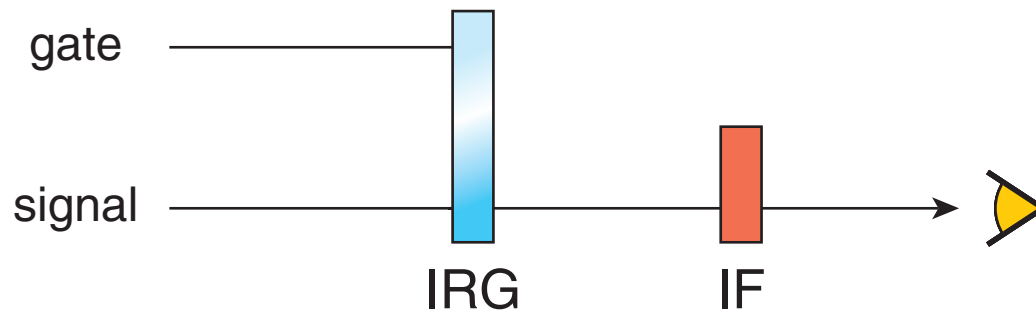
Experiment 2



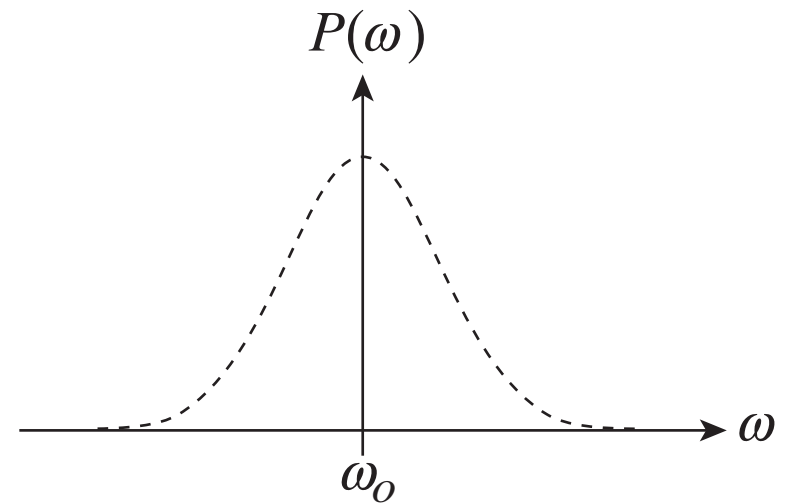
Experiment 2



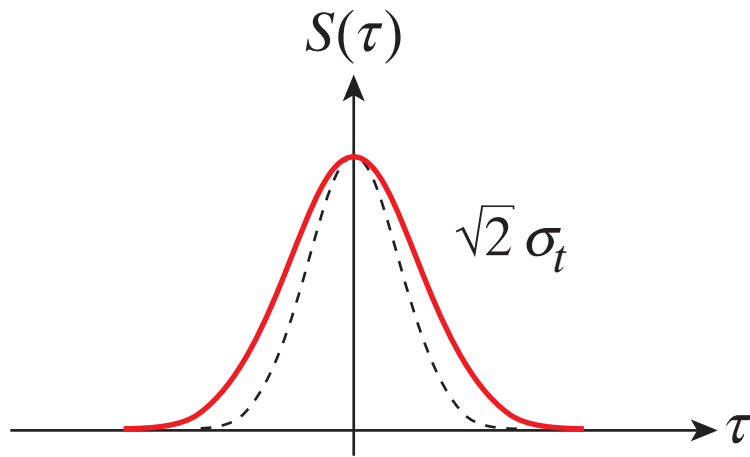
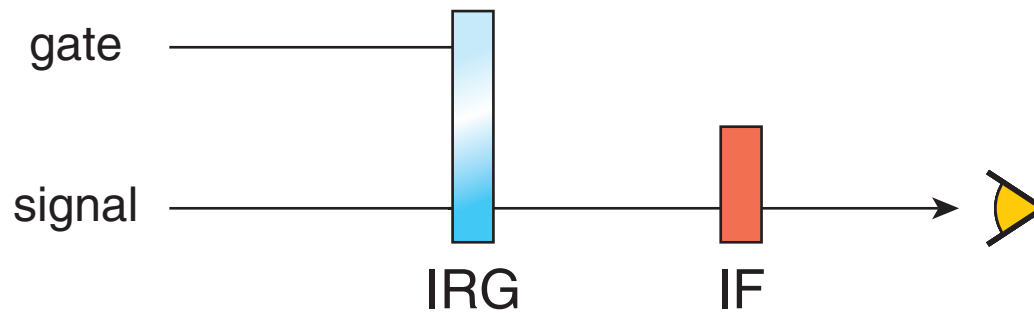
Experiment 2



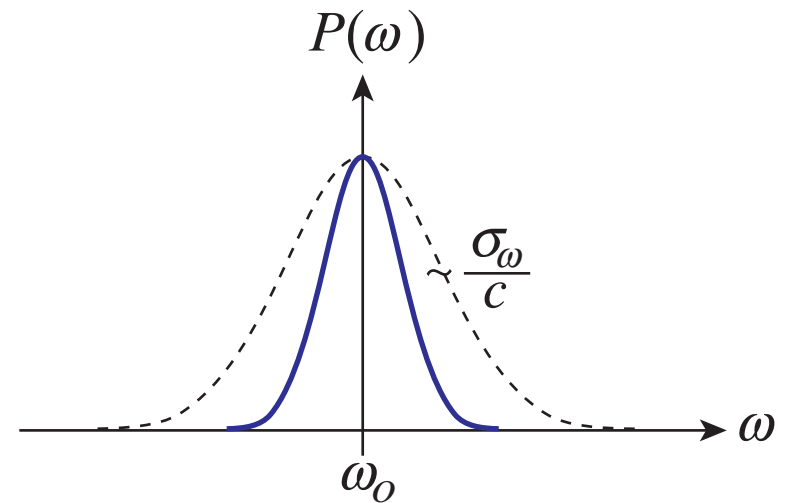
time resolution



Experiment 2



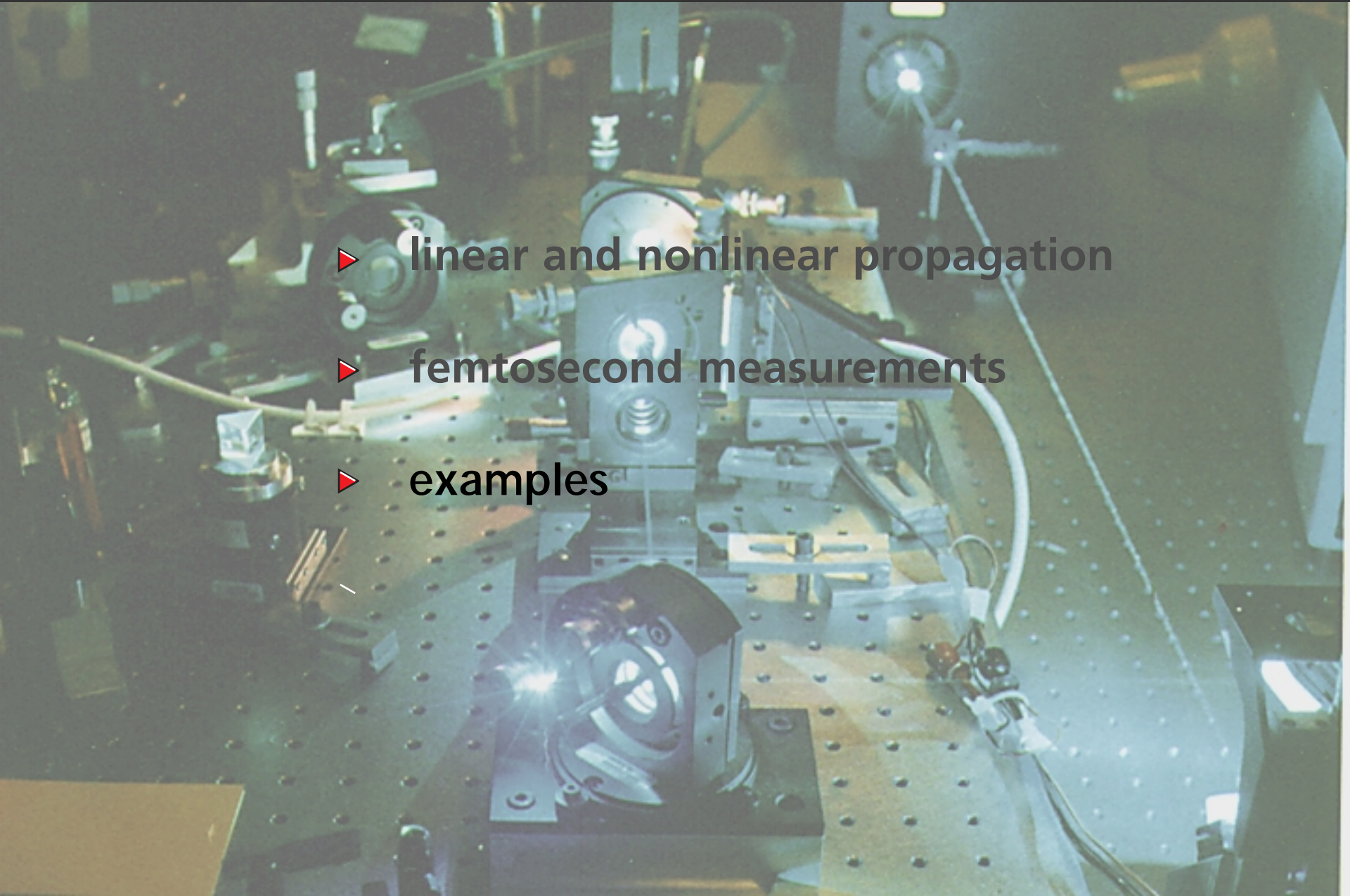
time resolution



spectral resolution

Outline

- ▶ linear and nonlinear propagation
- ▶ femtosecond measurements
- ▶ examples

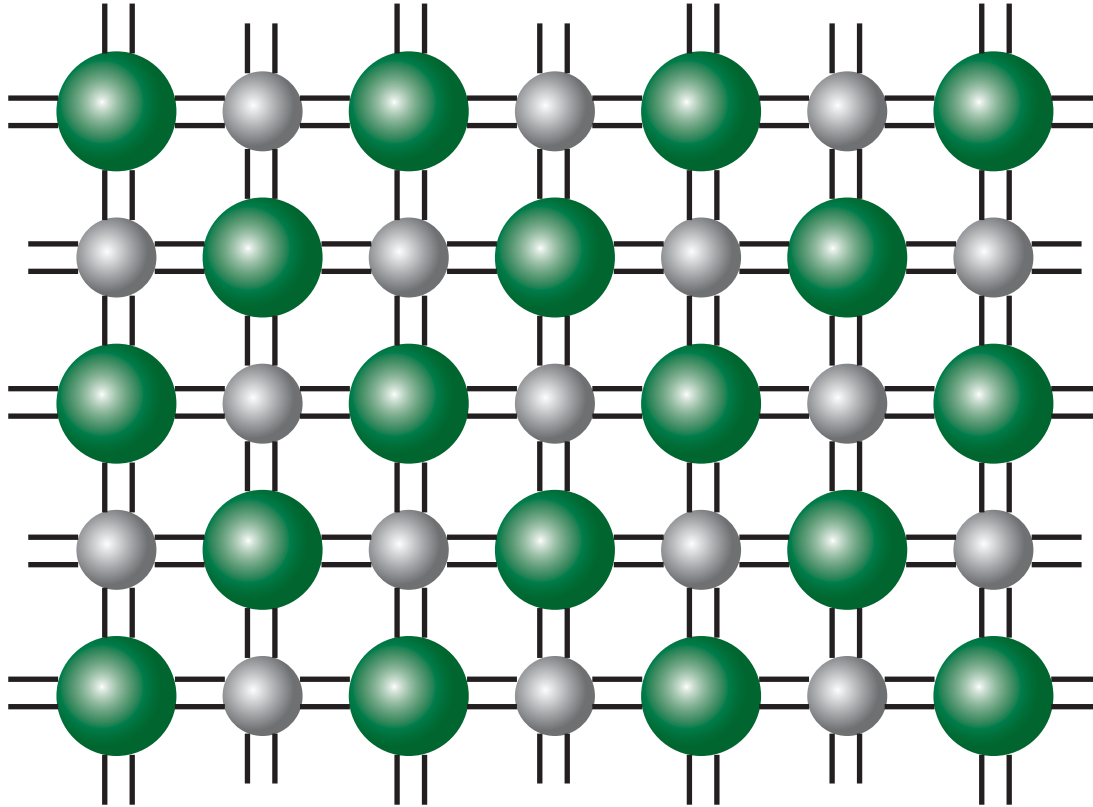


Introduction

short laser pulses can drive structural transitions

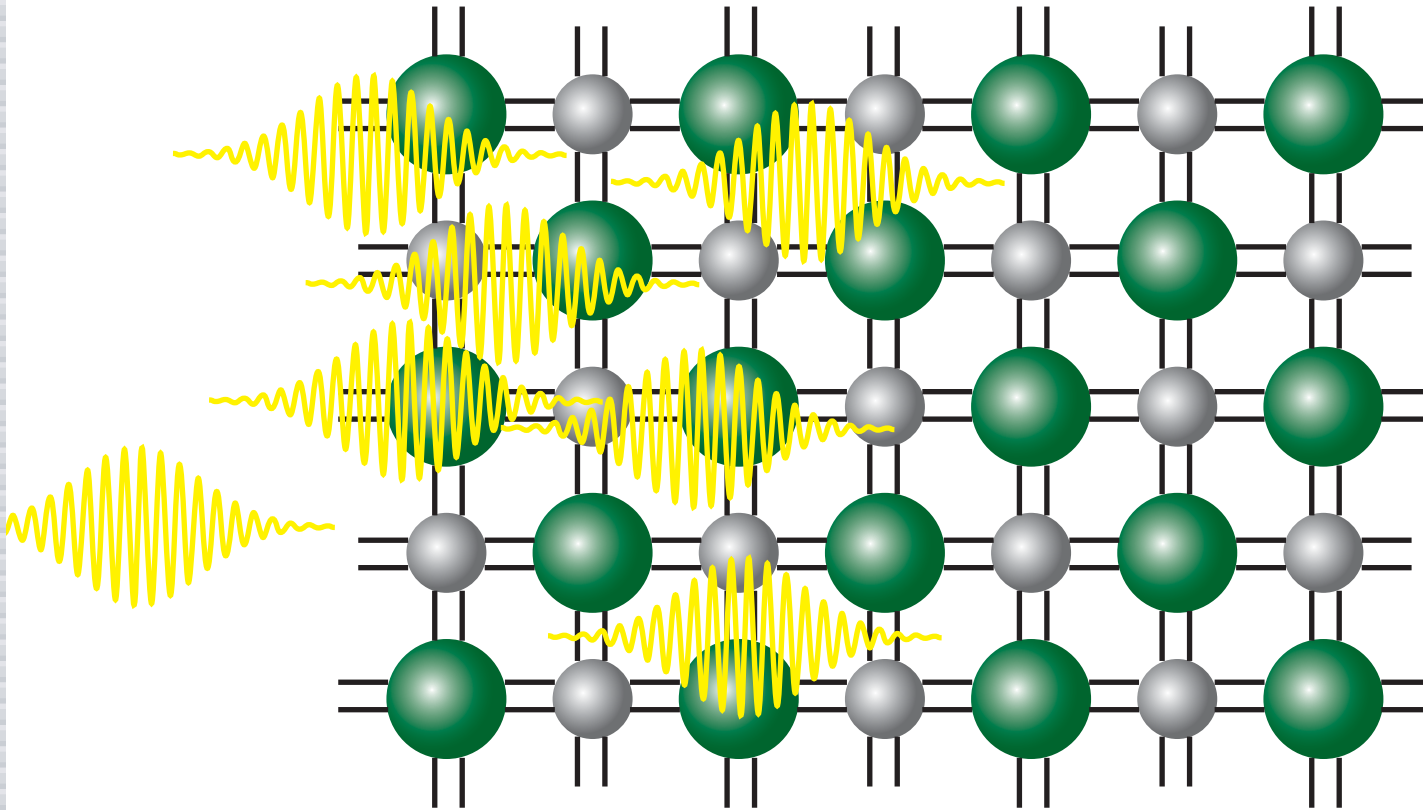
Introduction

how do femtosecond laser pulses alter a solid?



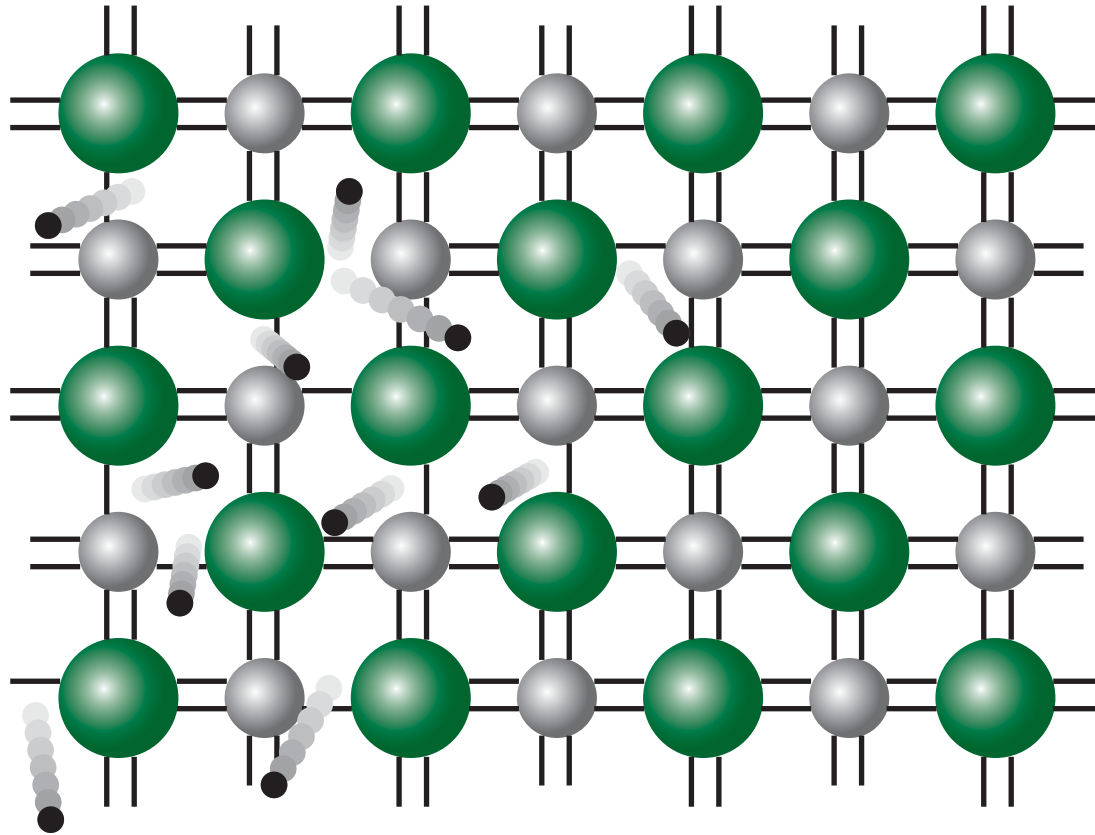
Introduction

photons excite valence electrons...



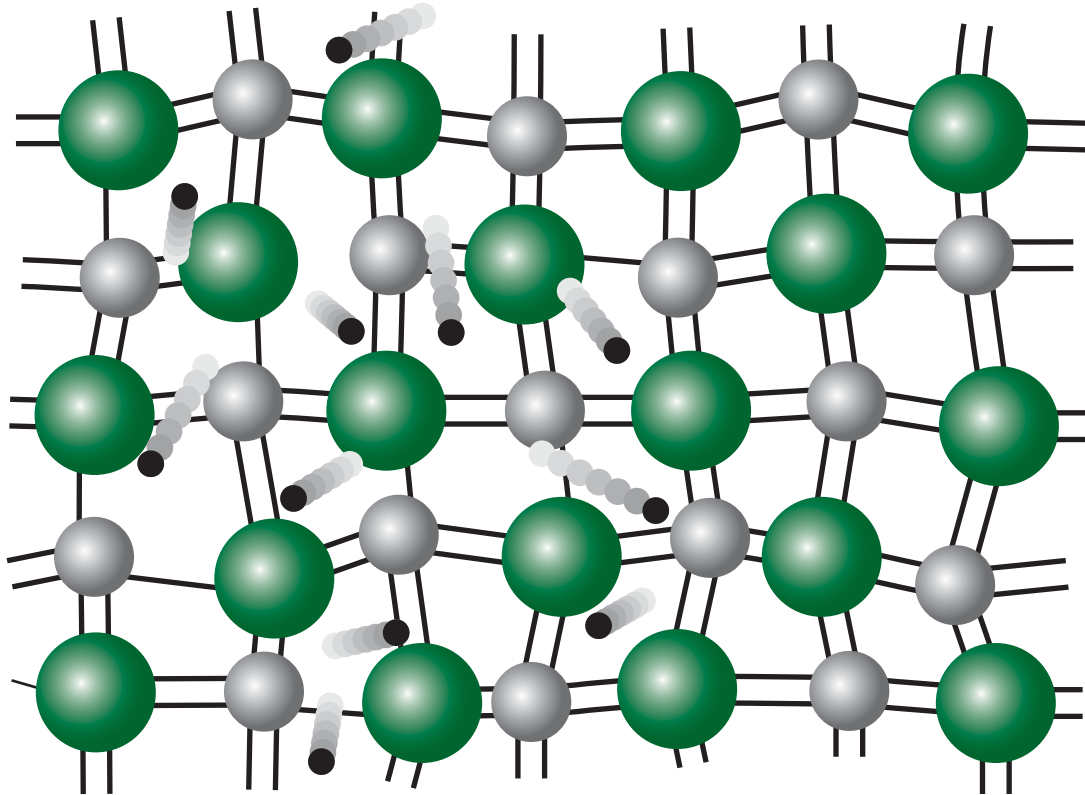
Introduction

... and create free electrons...



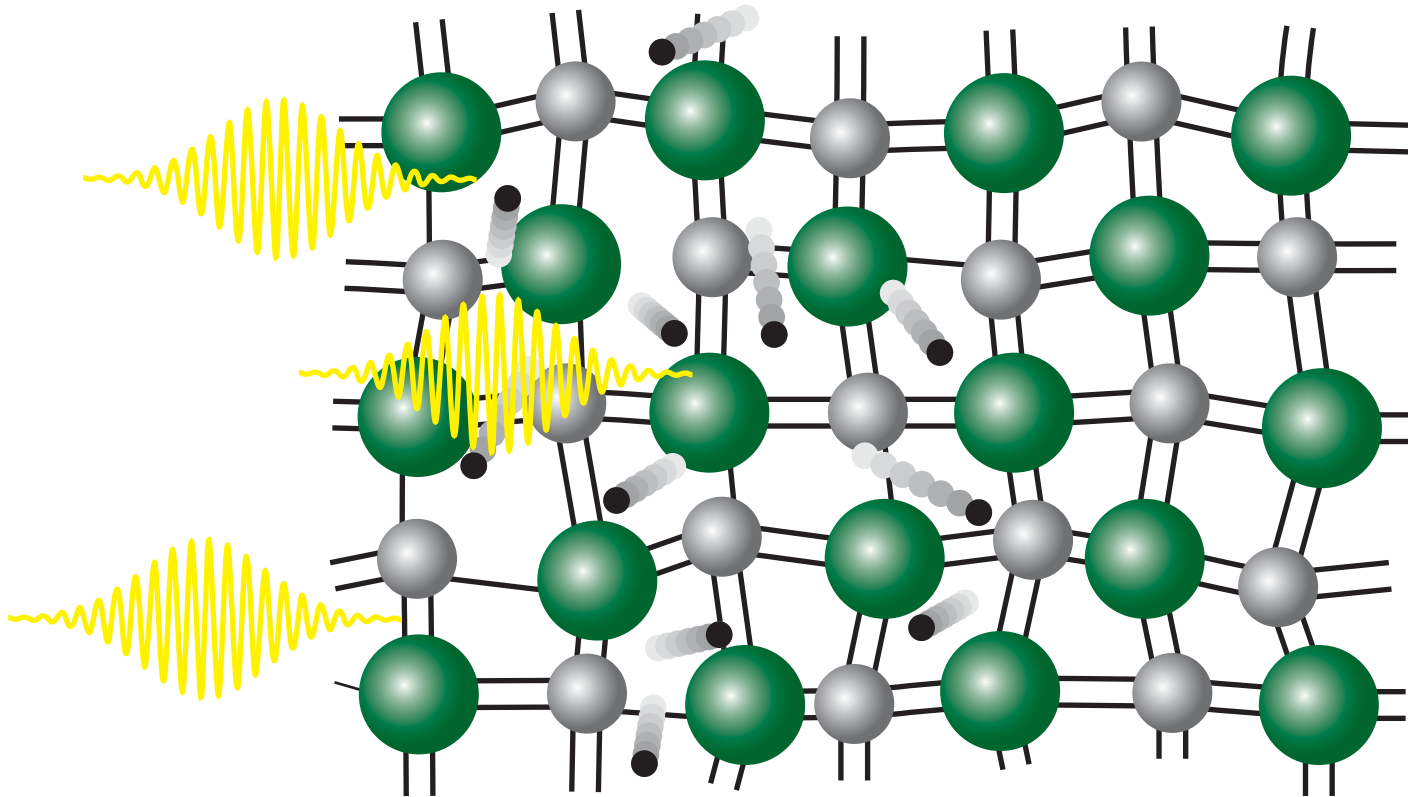
Introduction

... causing electronic and structural changes...



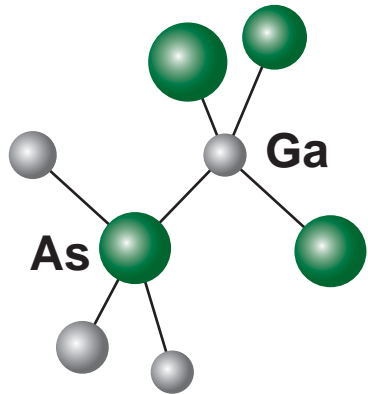
Introduction

... which we detect with a second laser pulse



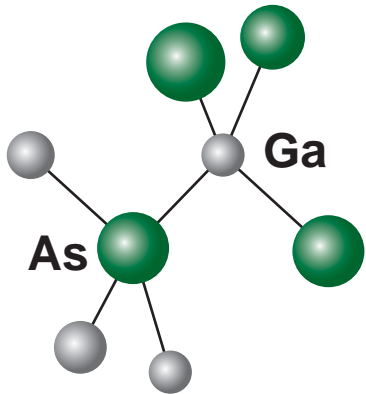
Introduction

structure

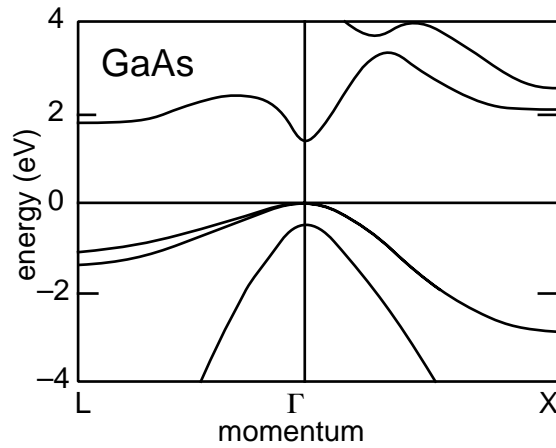


Introduction

structure

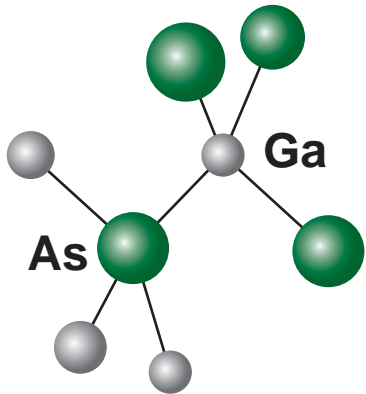


band structure

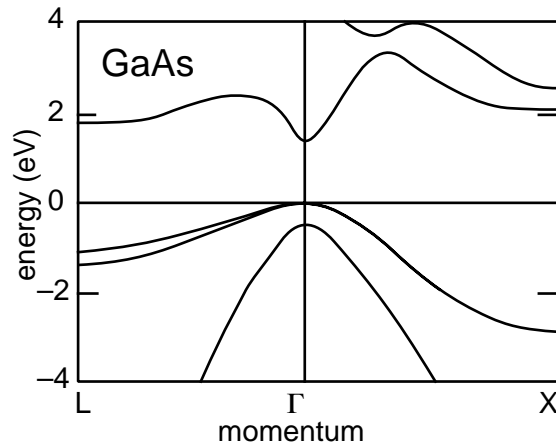


Introduction

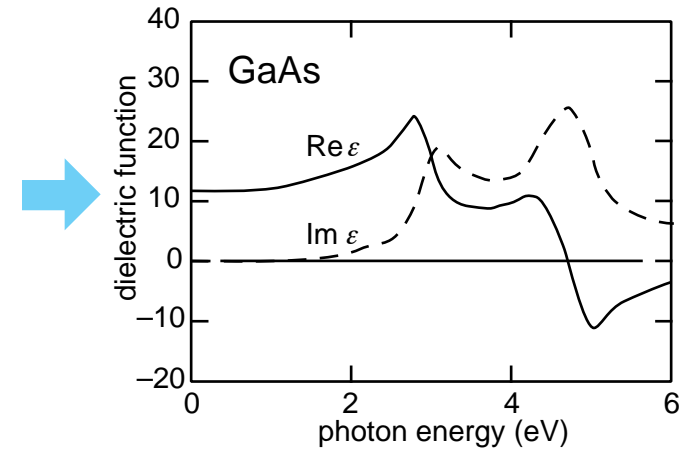
structure



band structure

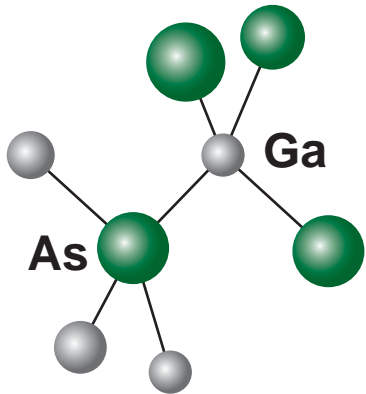


dielectric function

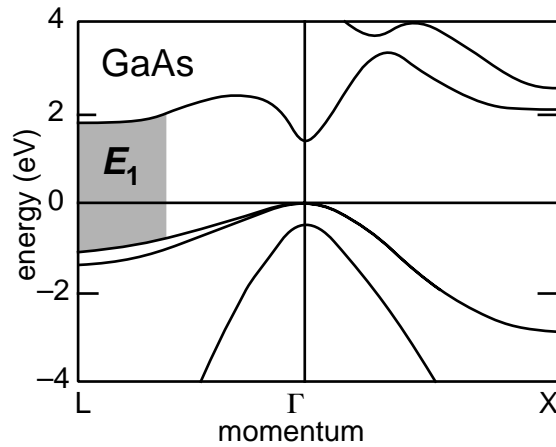


Introduction

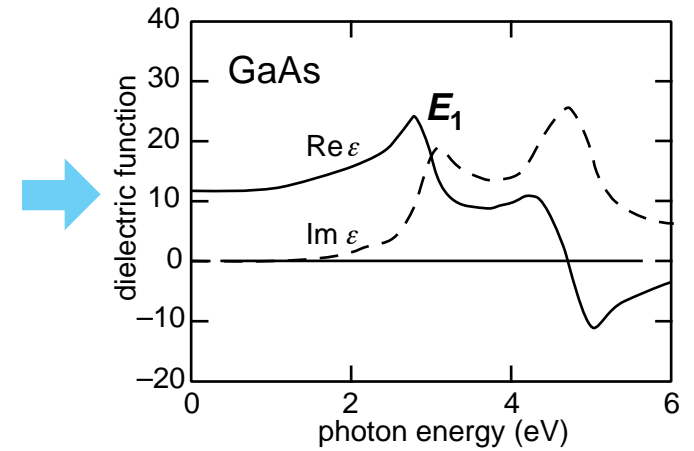
structure



band structure

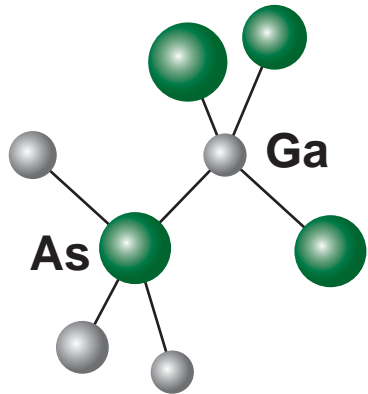


dielectric function

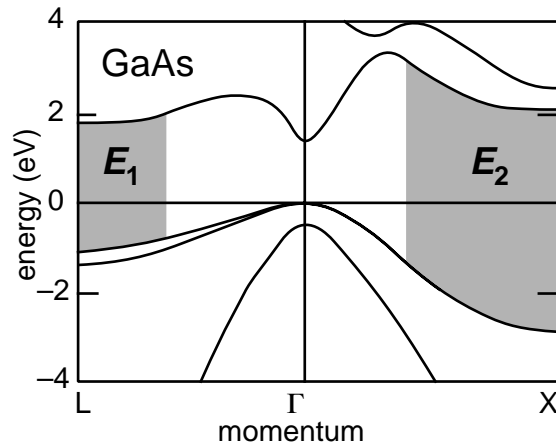


Introduction

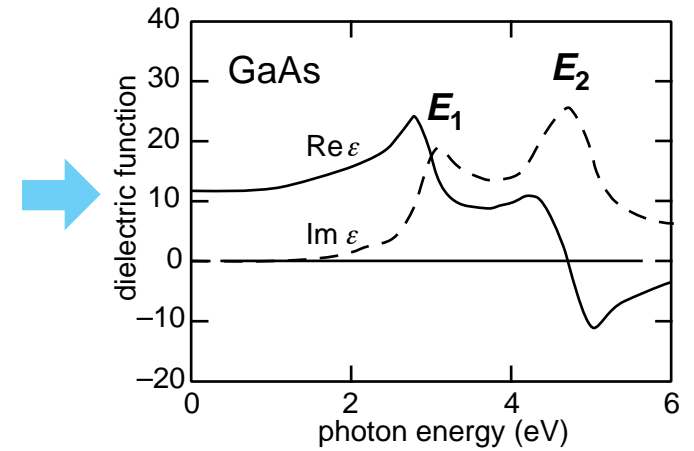
structure



band structure

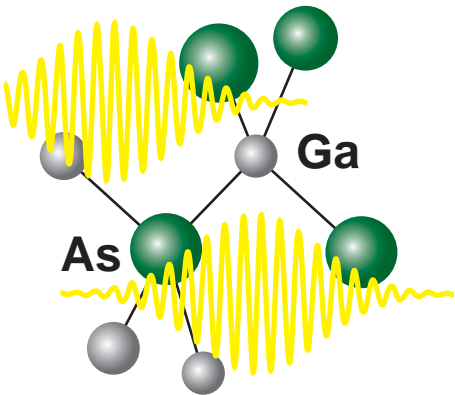


dielectric function

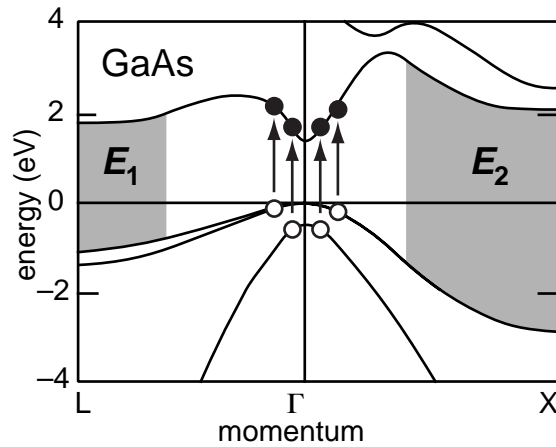


Introduction

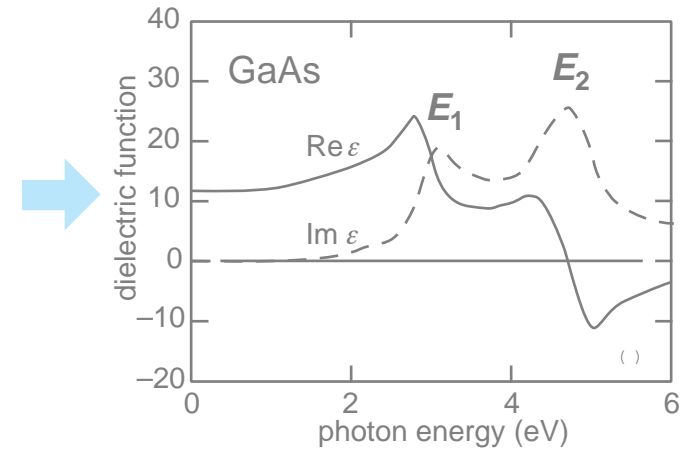
structure



band structure

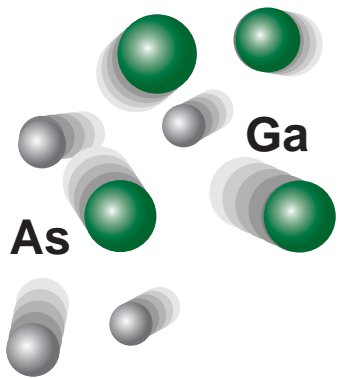


dielectric function

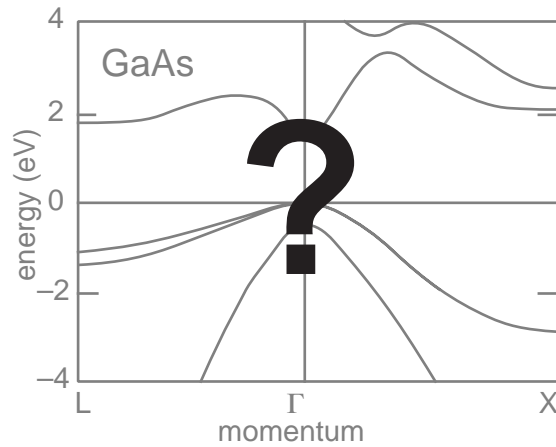


Introduction

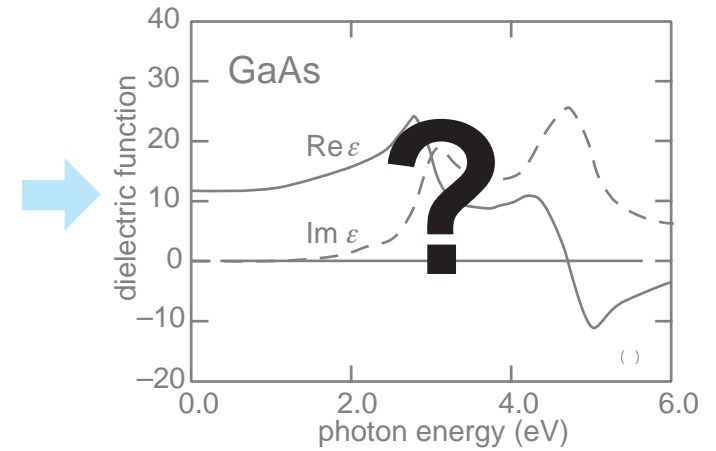
structure



band structure

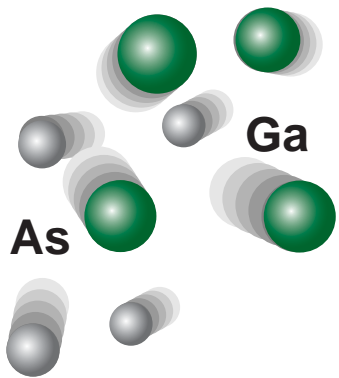


dielectric function

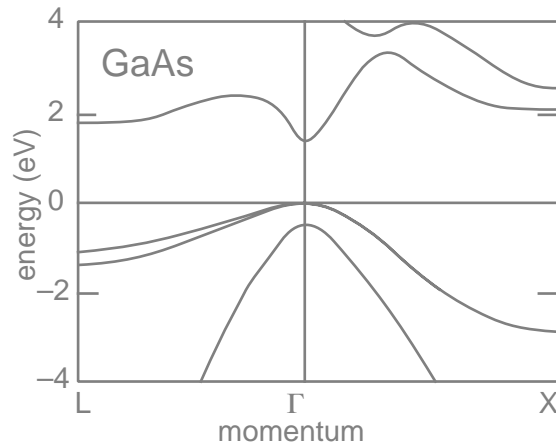


Introduction

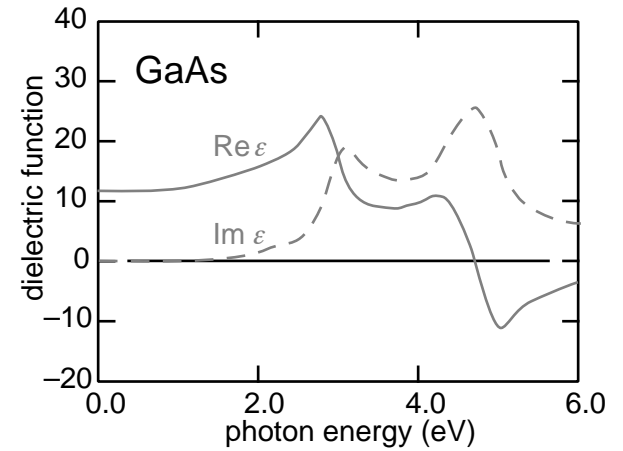
structure



band structure

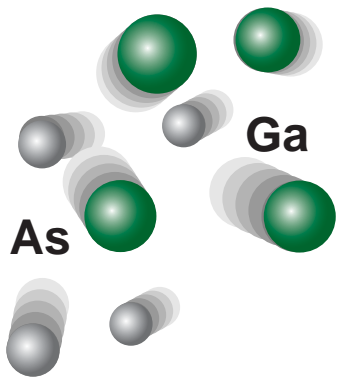


dielectric function

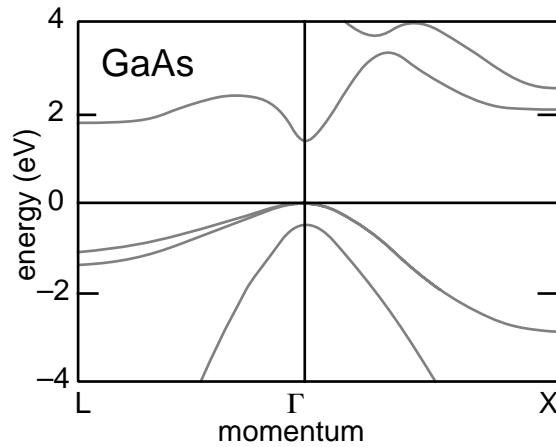


Introduction

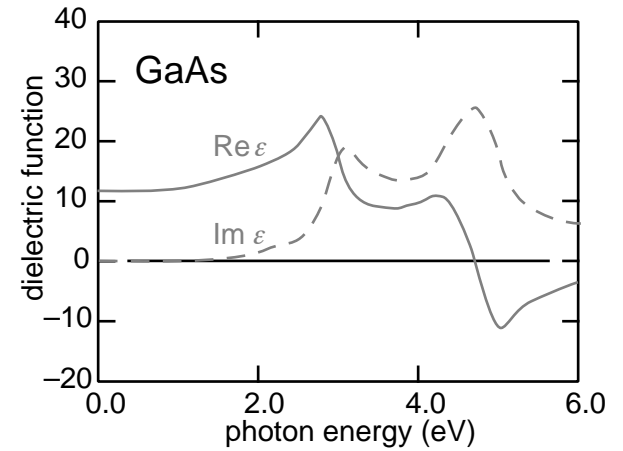
structure



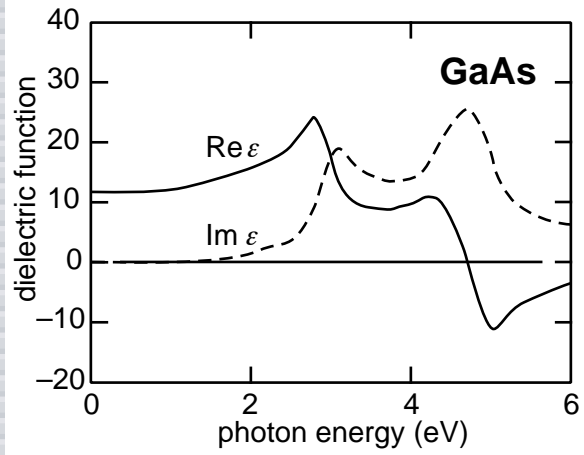
band structure



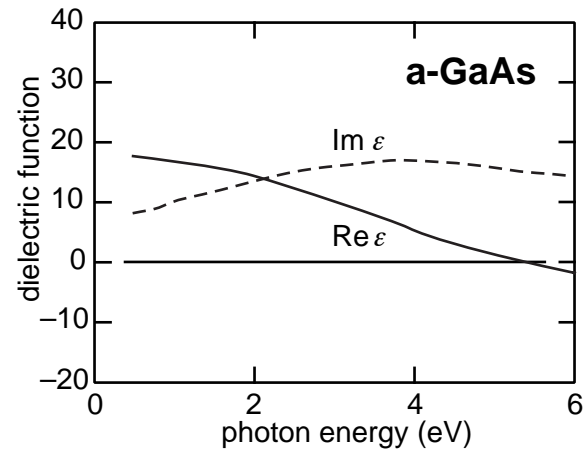
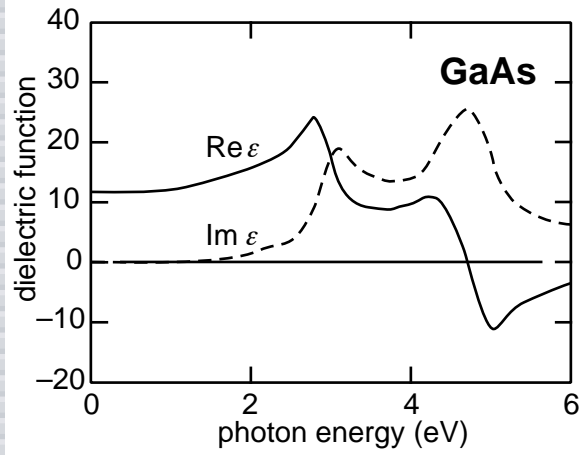
dielectric function



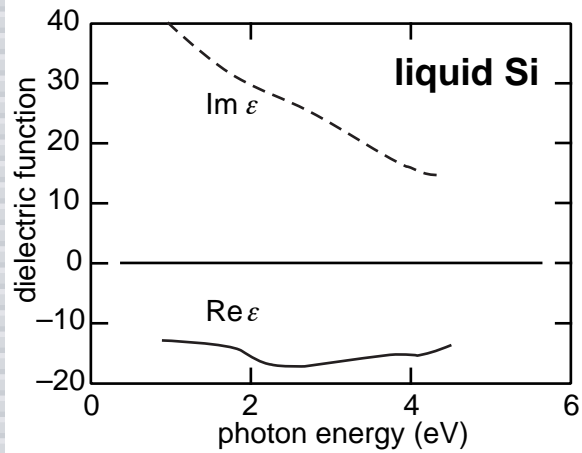
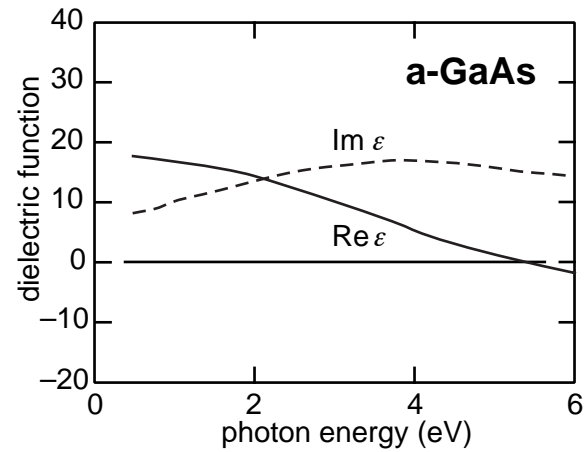
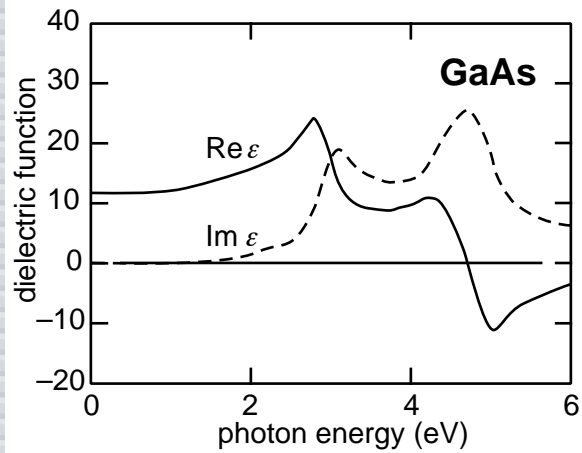
Introduction



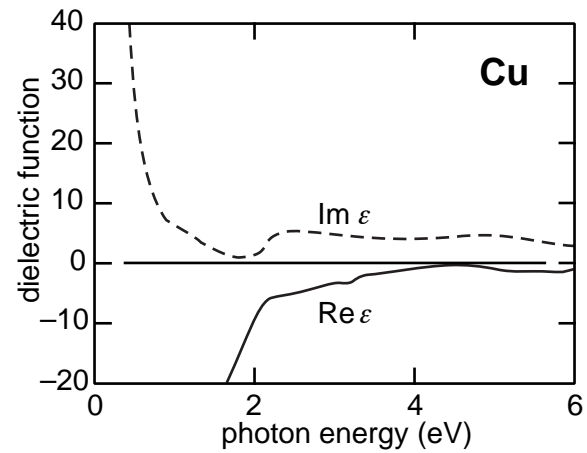
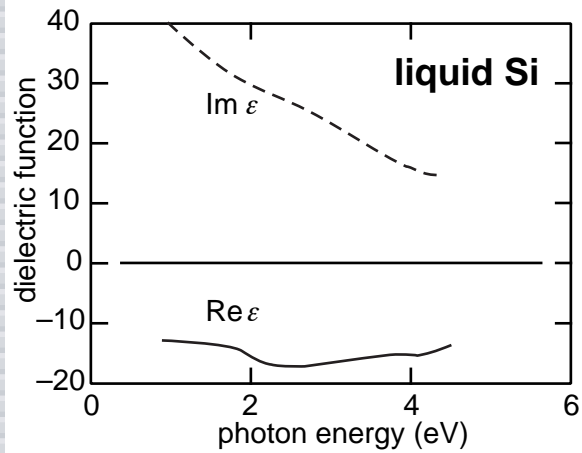
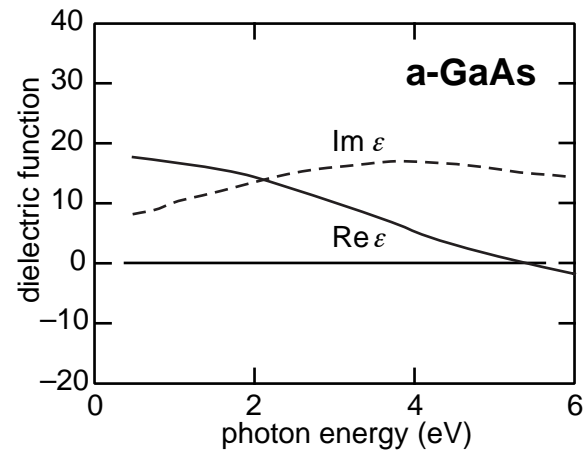
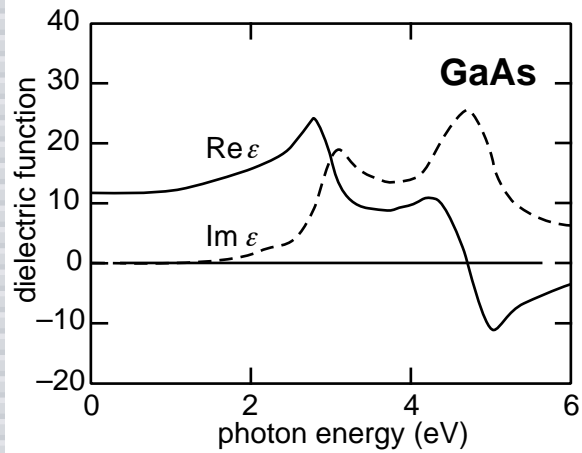
Introduction



Introduction

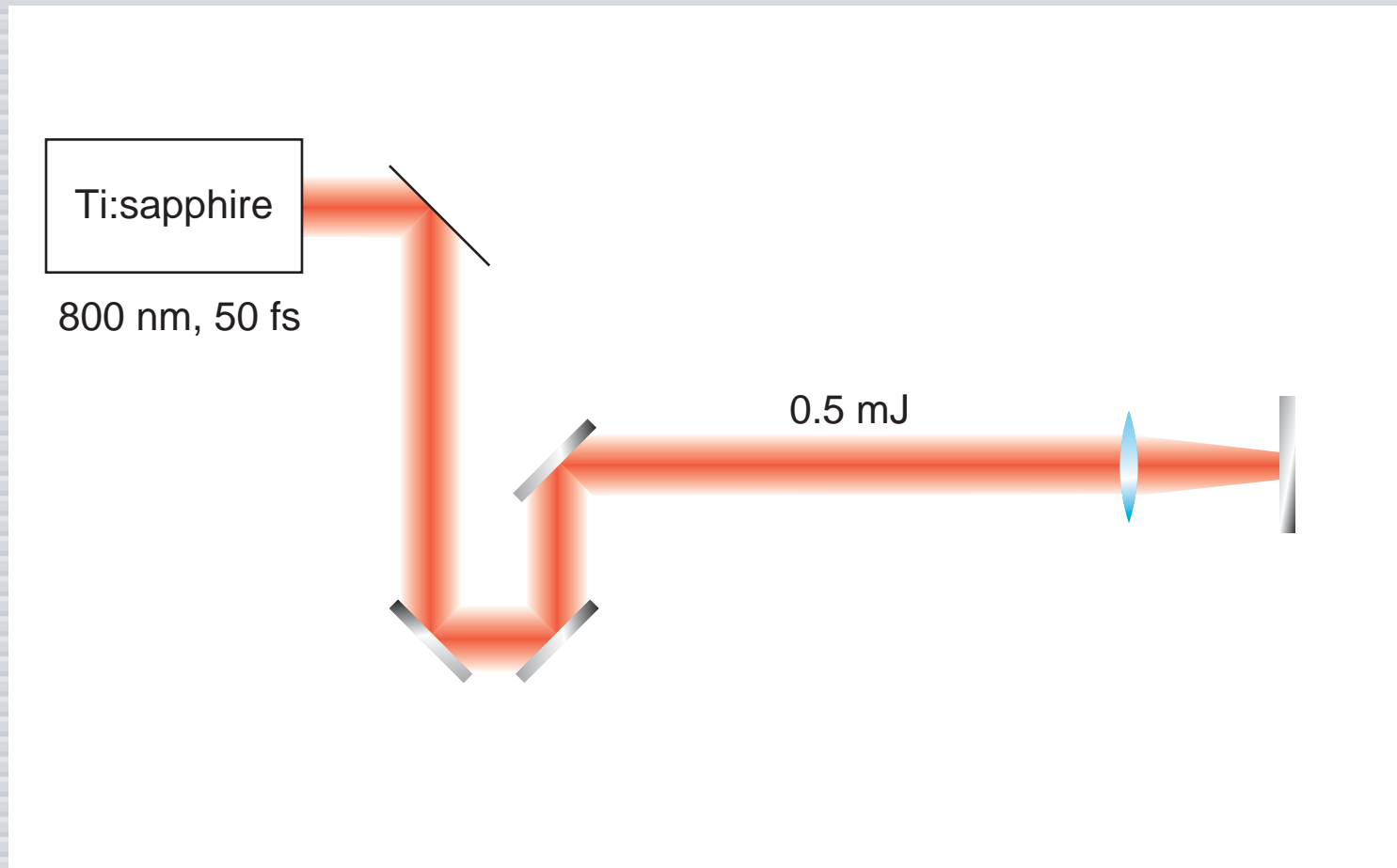


Introduction



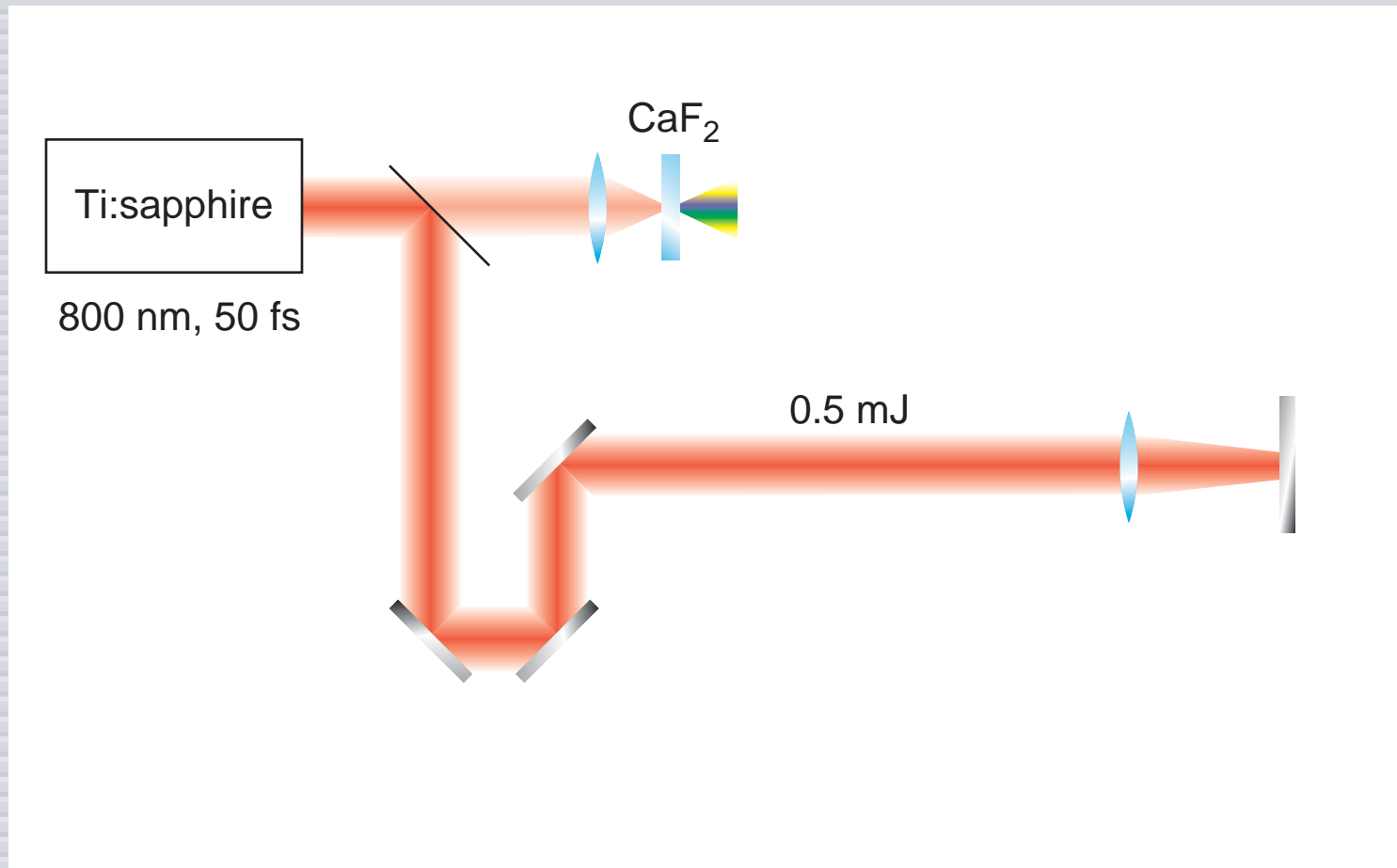
Technique

broadband time-resolved ellipsometry



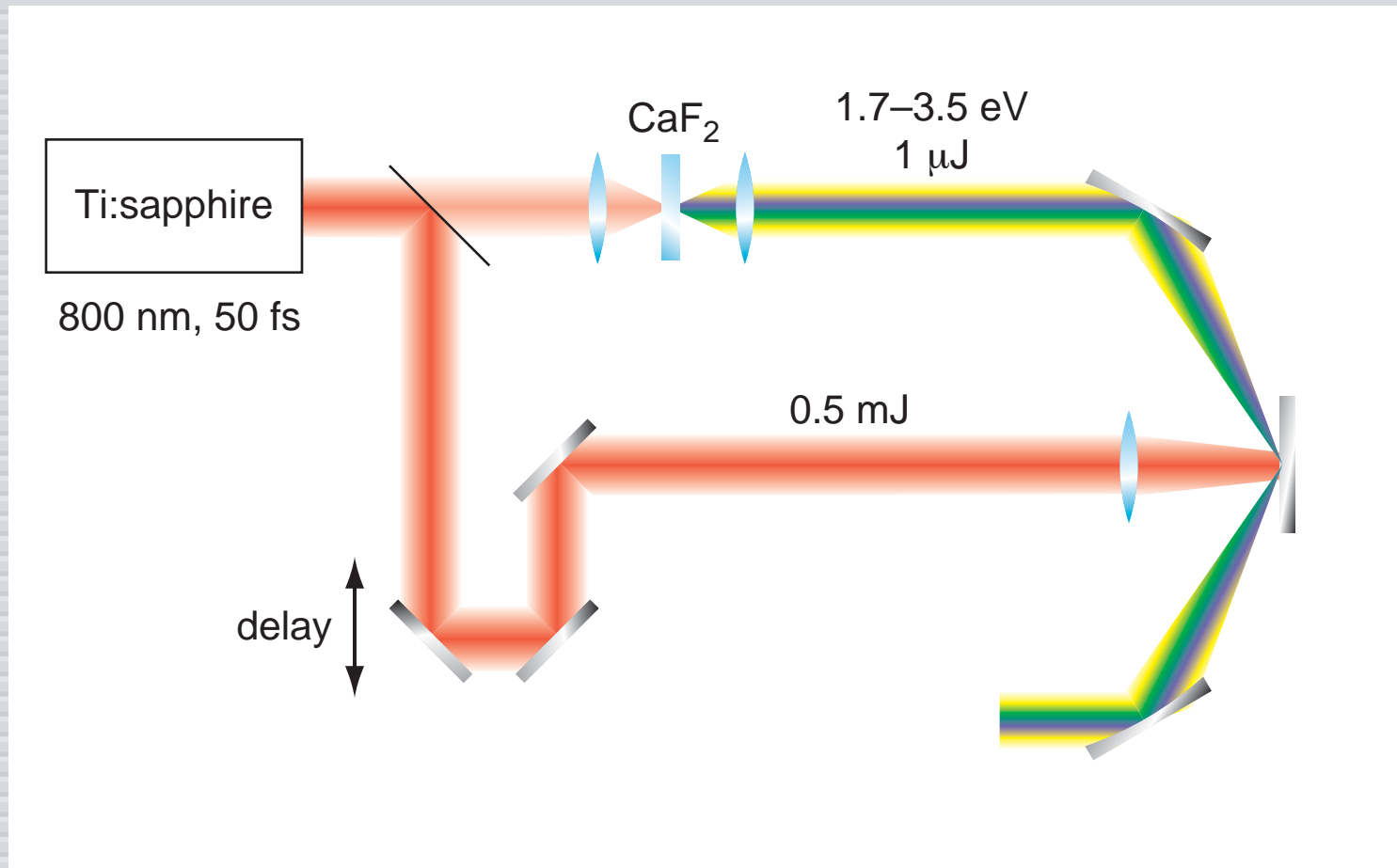
Technique

broadband time-resolved ellipsometry



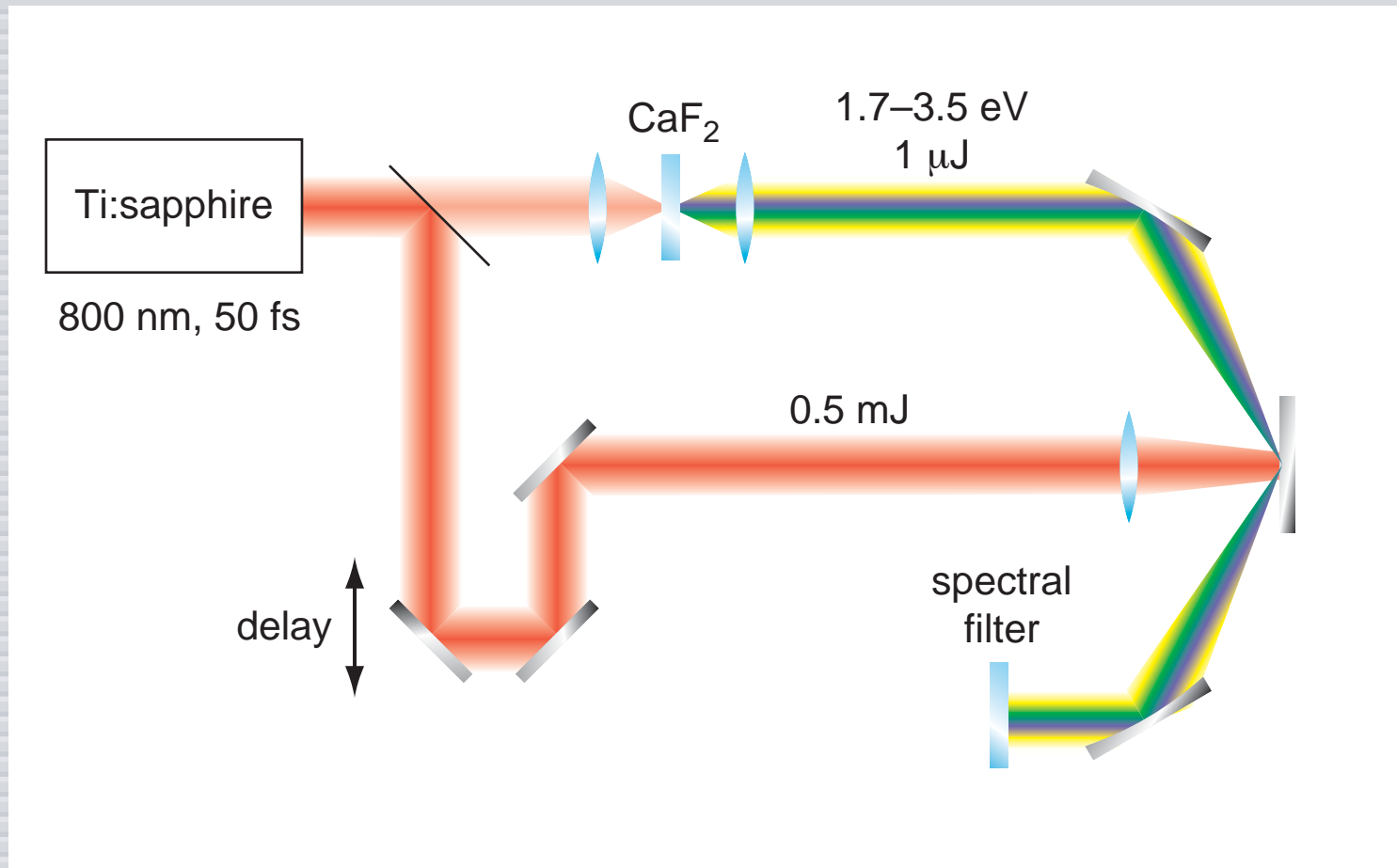
Technique

broadband time-resolved ellipsometry



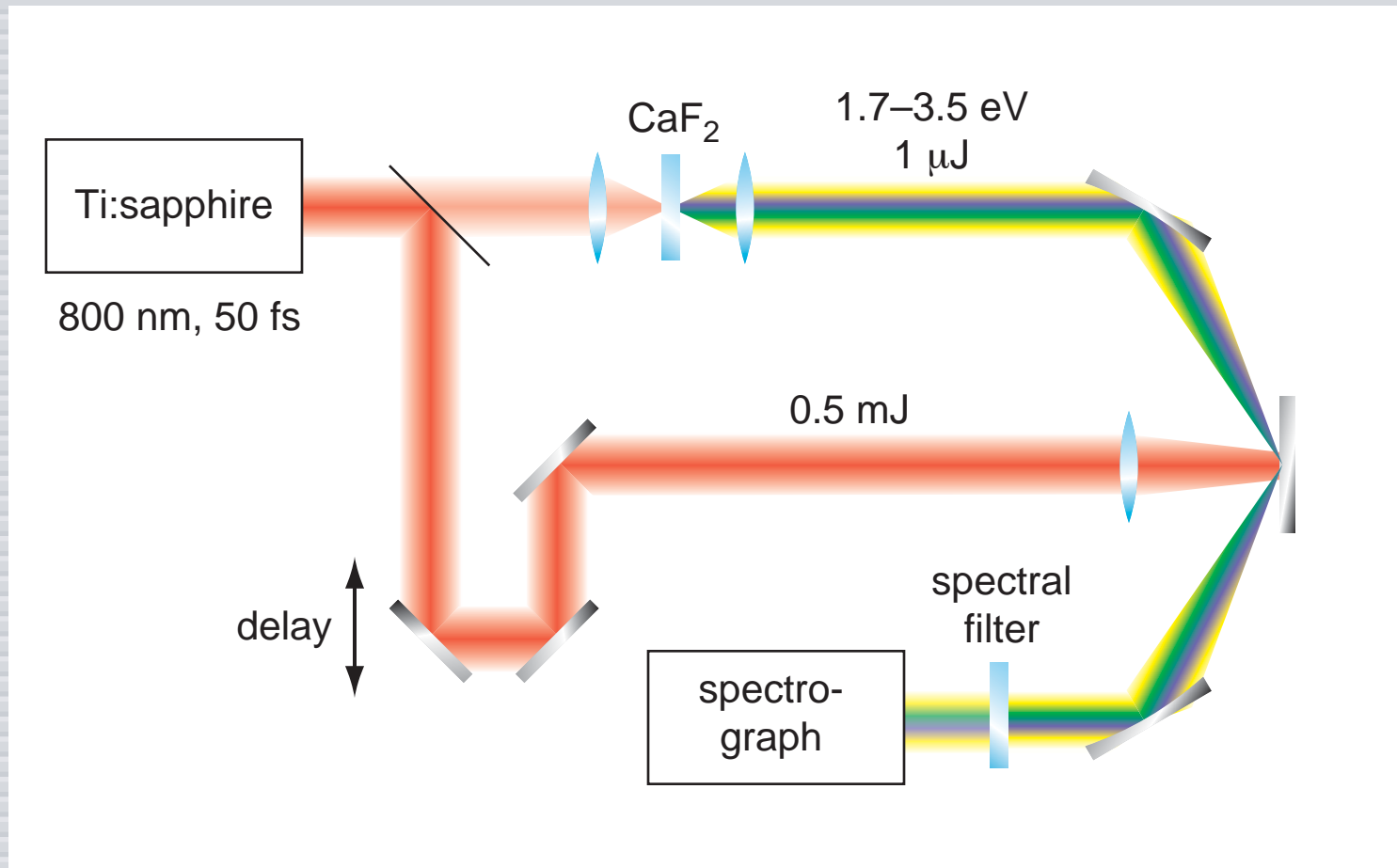
Technique

broadband time-resolved ellipsometry



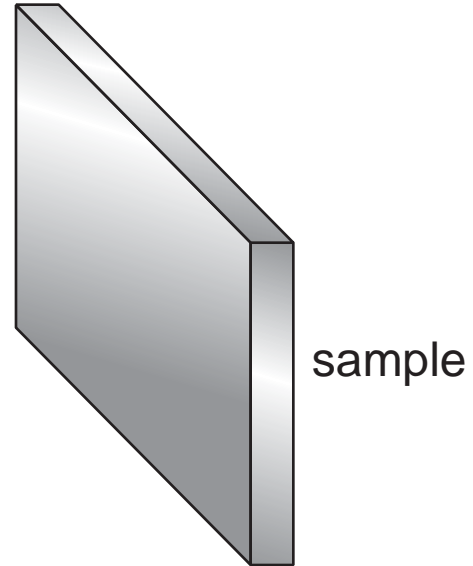
Technique

broadband time-resolved ellipsometry



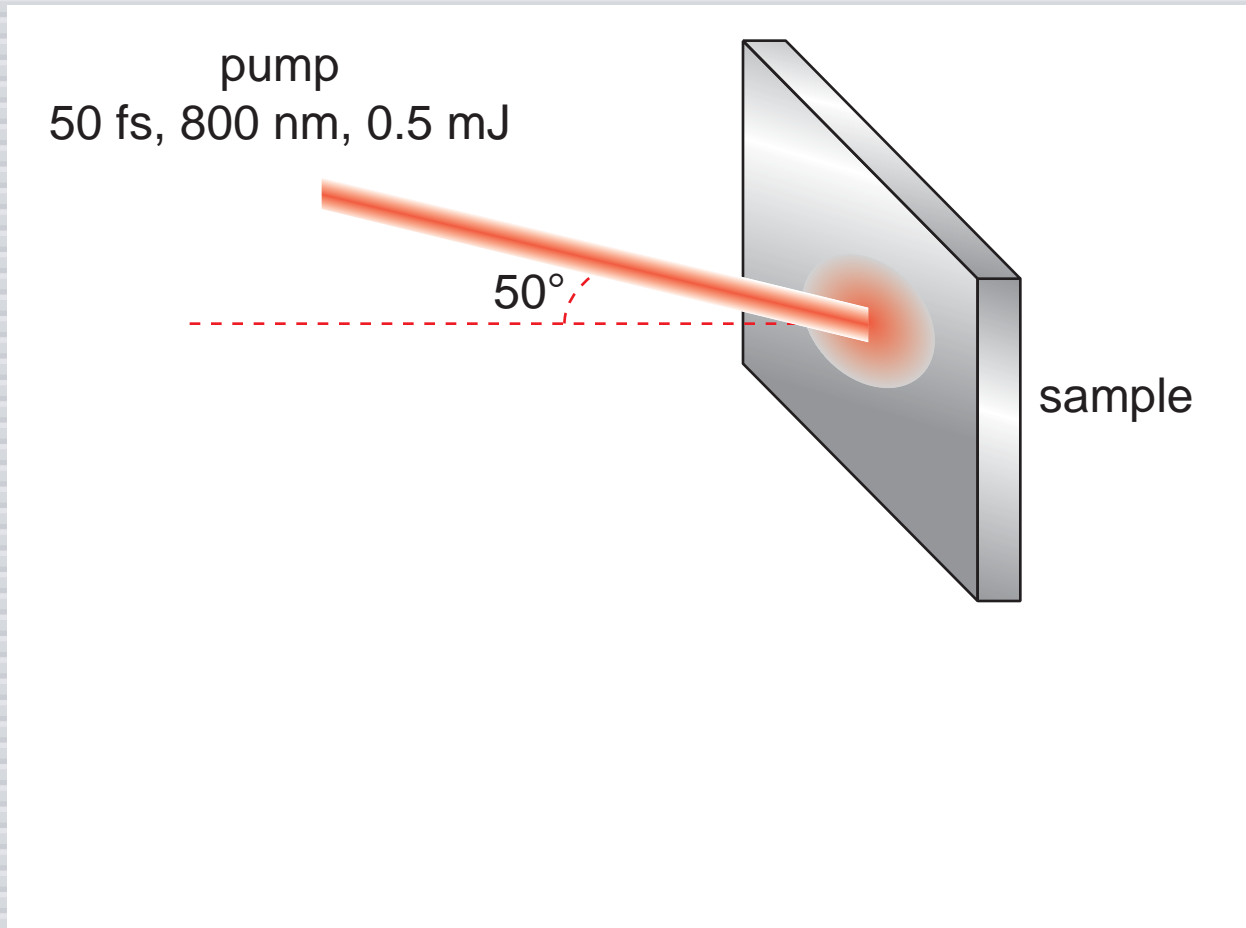
Technique

broadband time-resolved ellipsometry



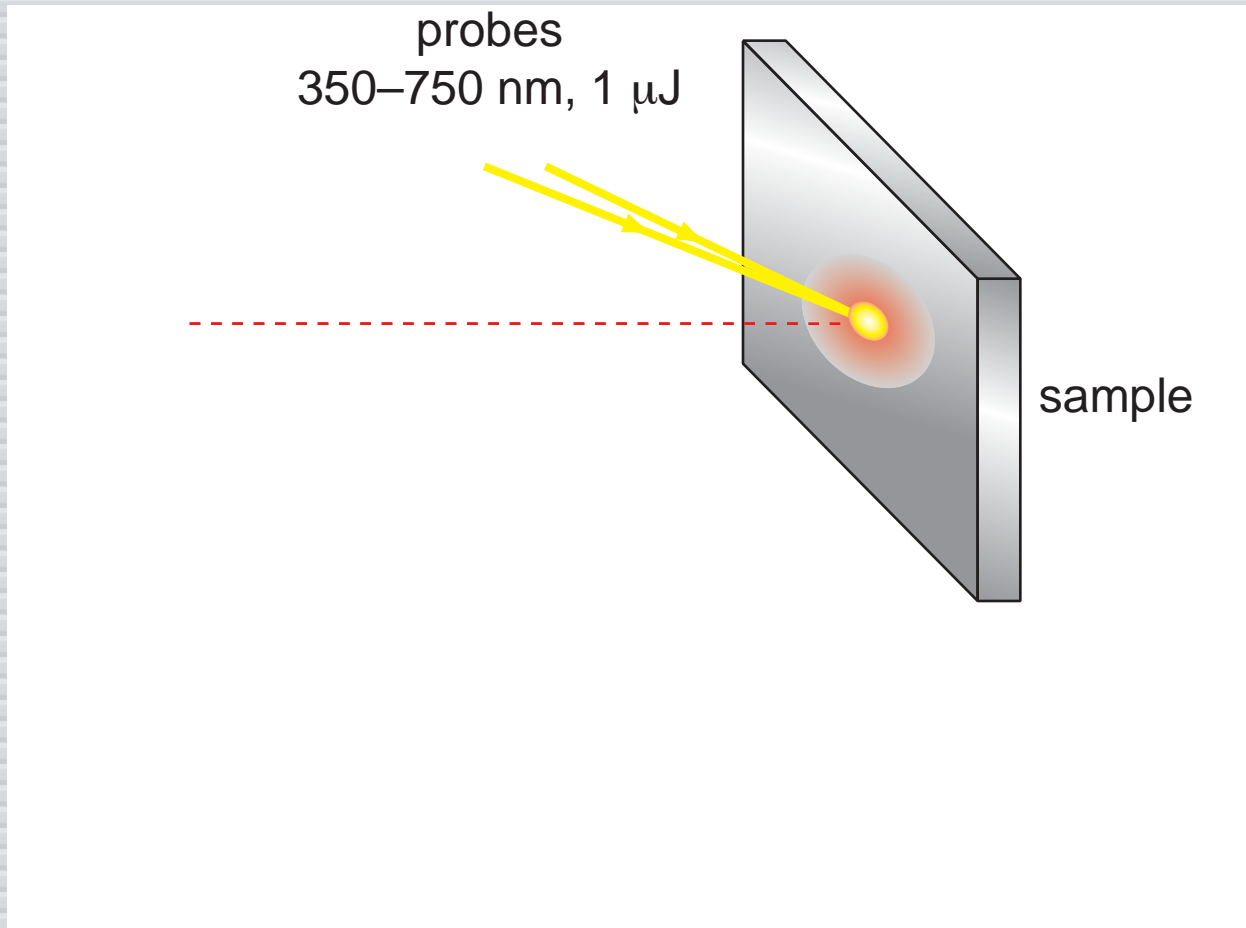
Technique

broadband time-resolved ellipsometry



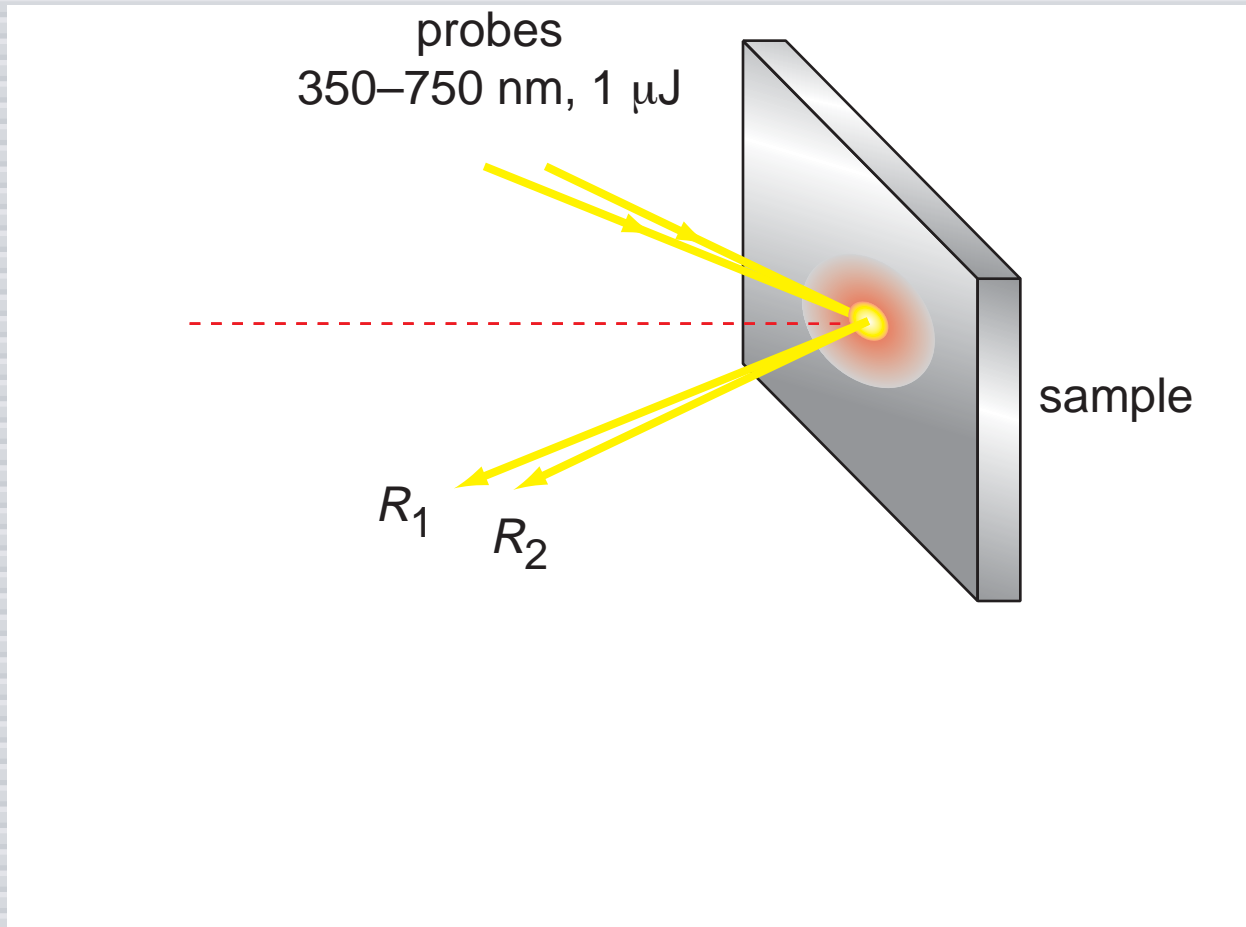
Technique

broadband time-resolved ellipsometry



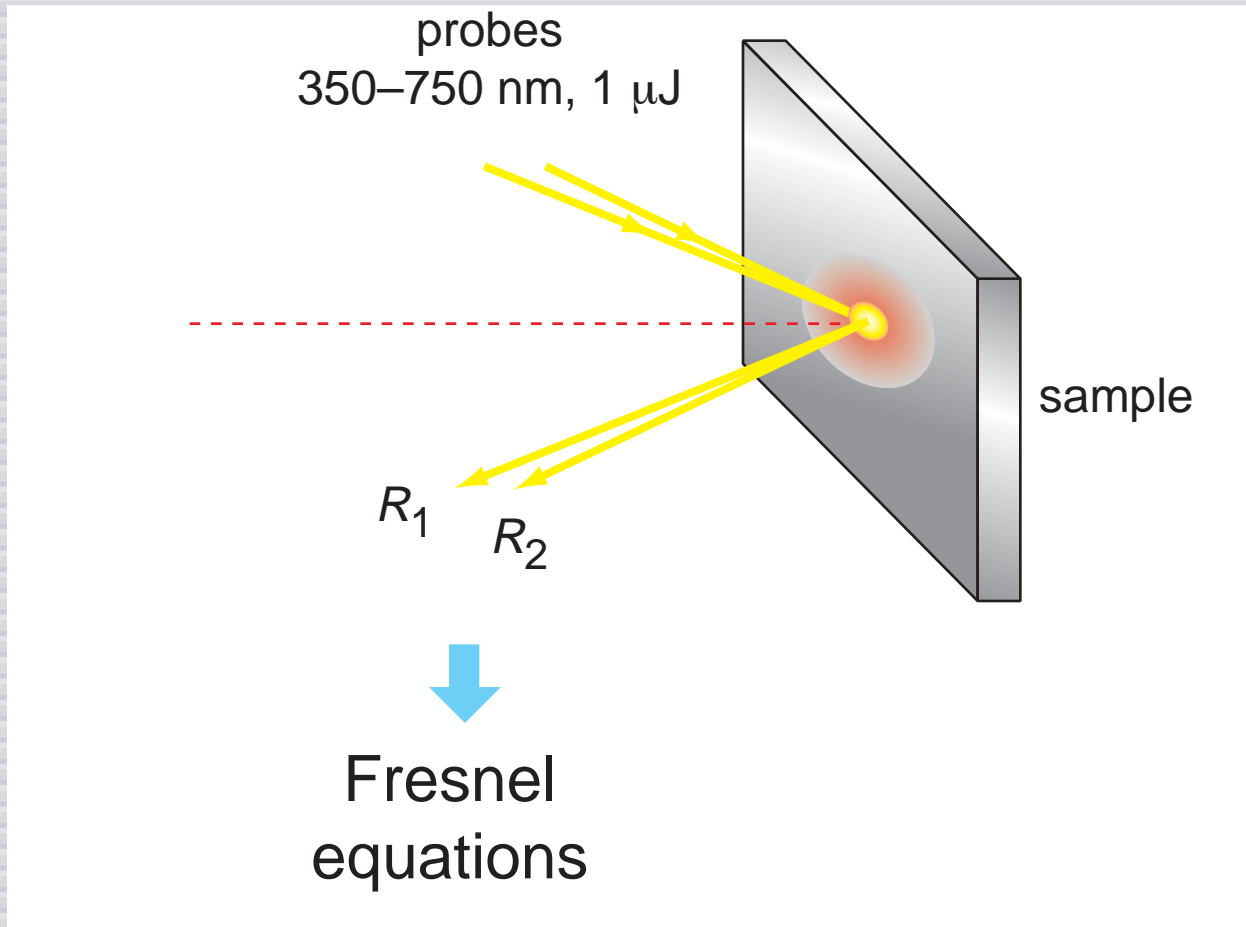
Technique

broadband time-resolved ellipsometry



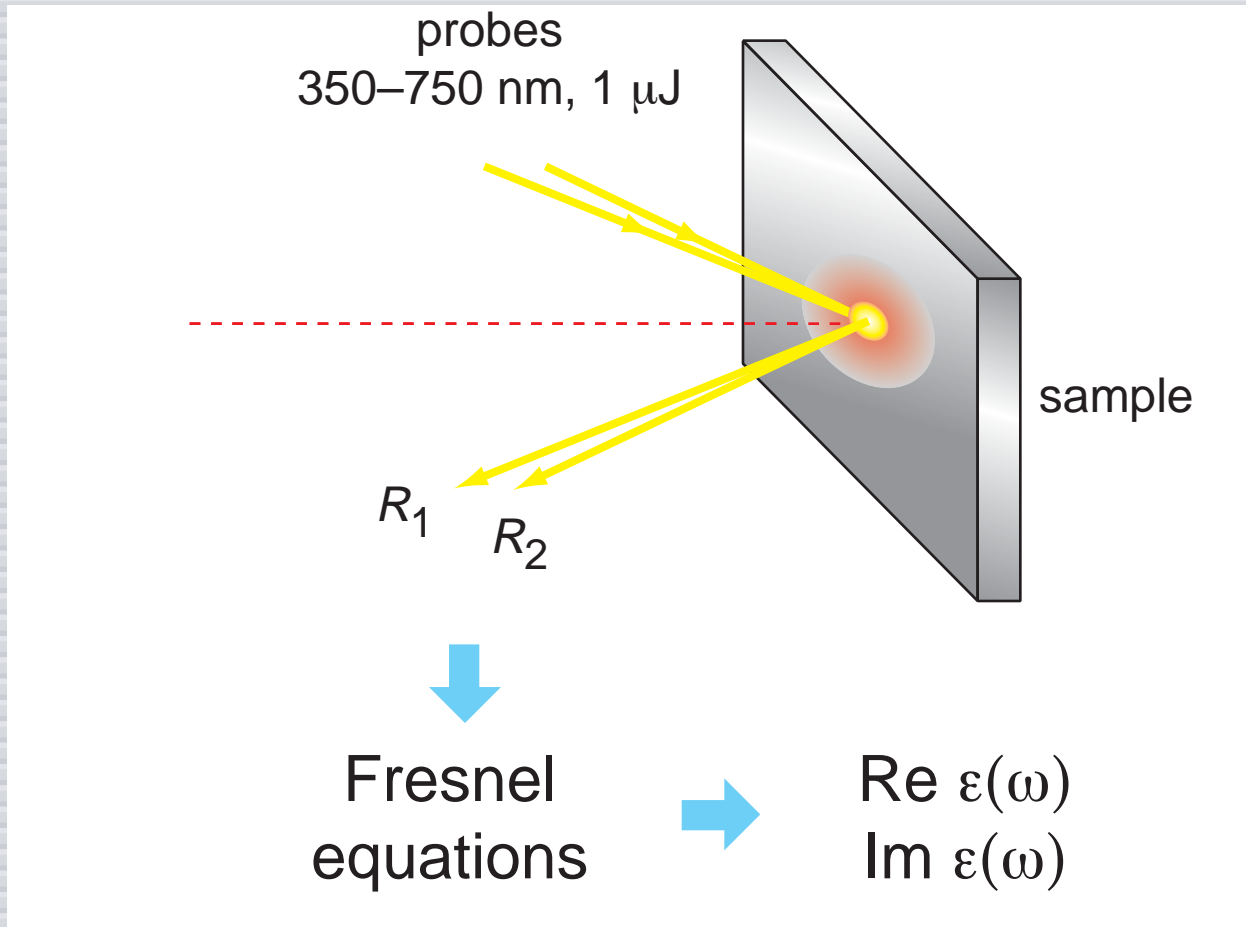
Technique

broadband time-resolved ellipsometry



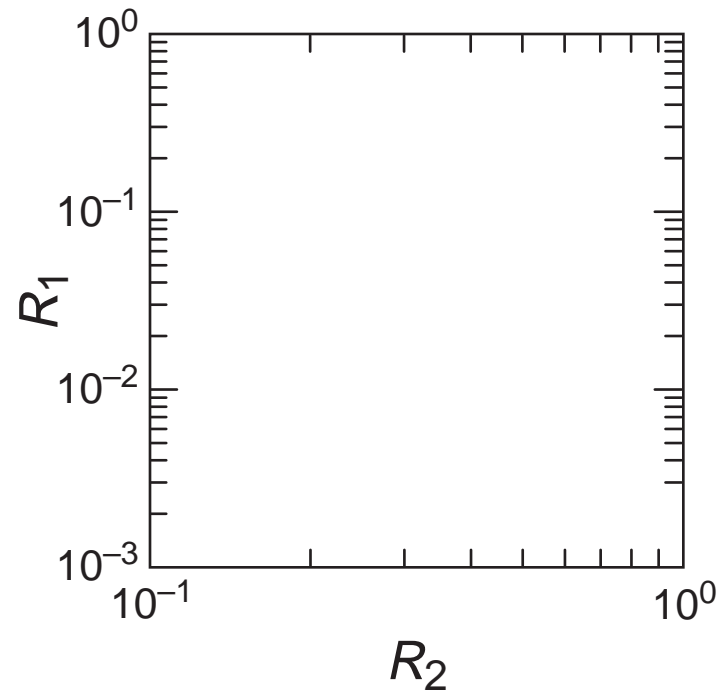
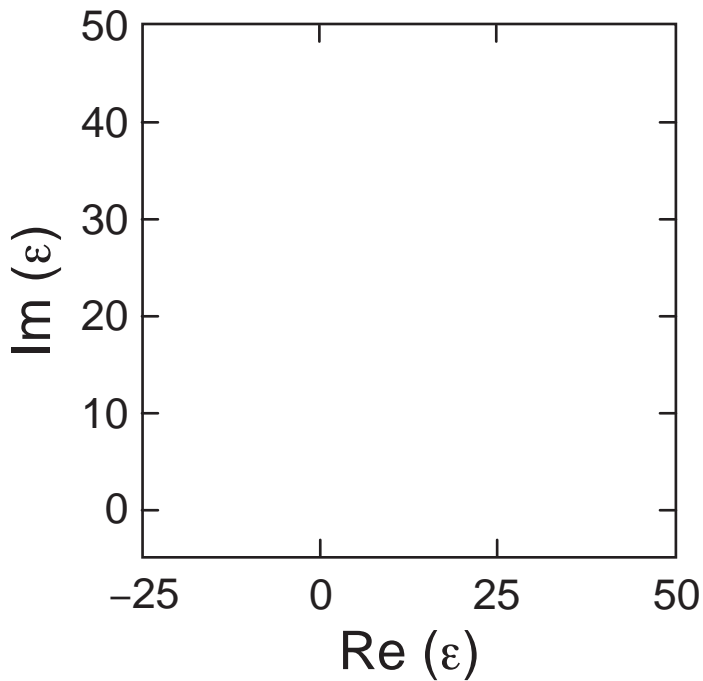
Technique

broadband time-resolved ellipsometry



Technique

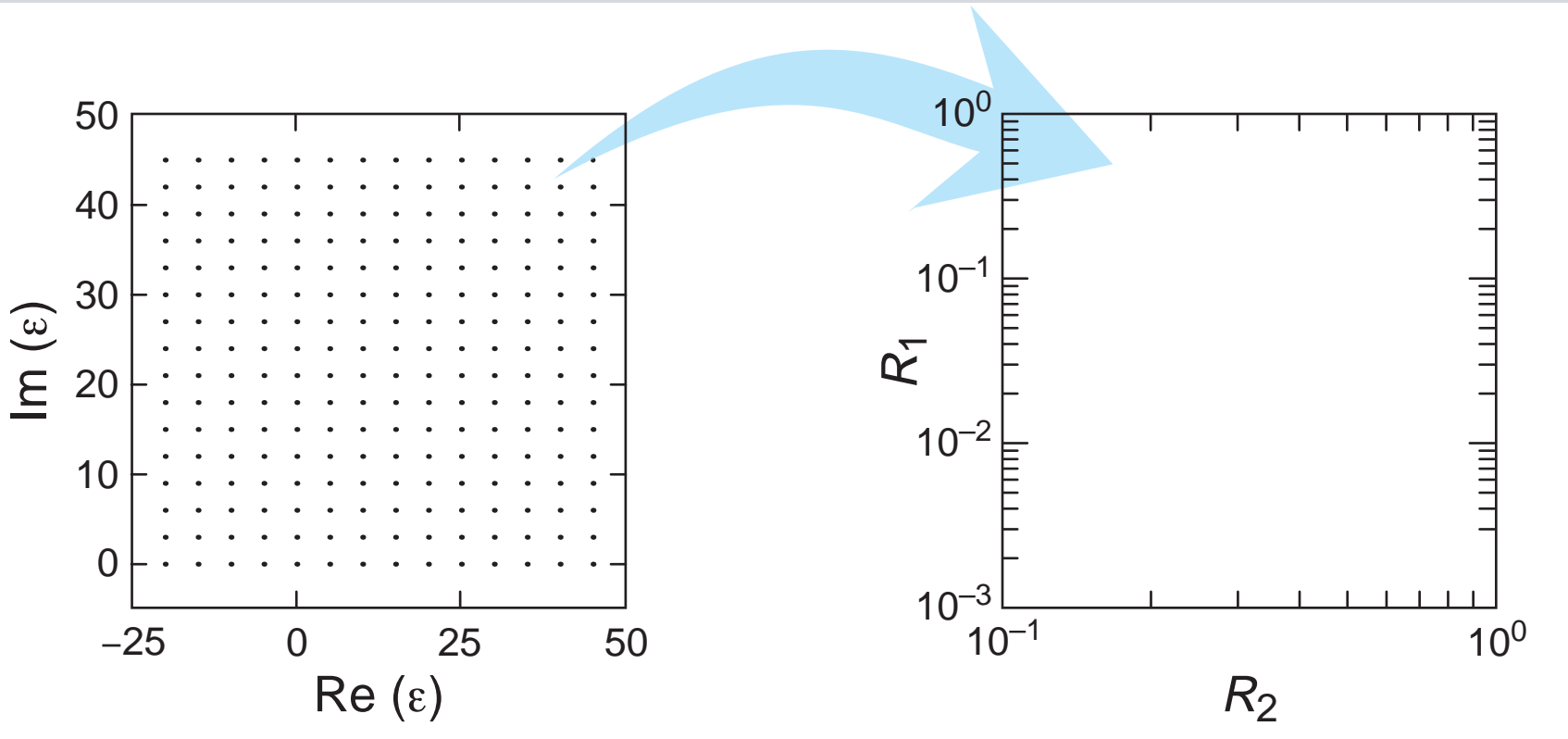
choice of angles



Fresnel equations cannot be inverted analytically

Technique

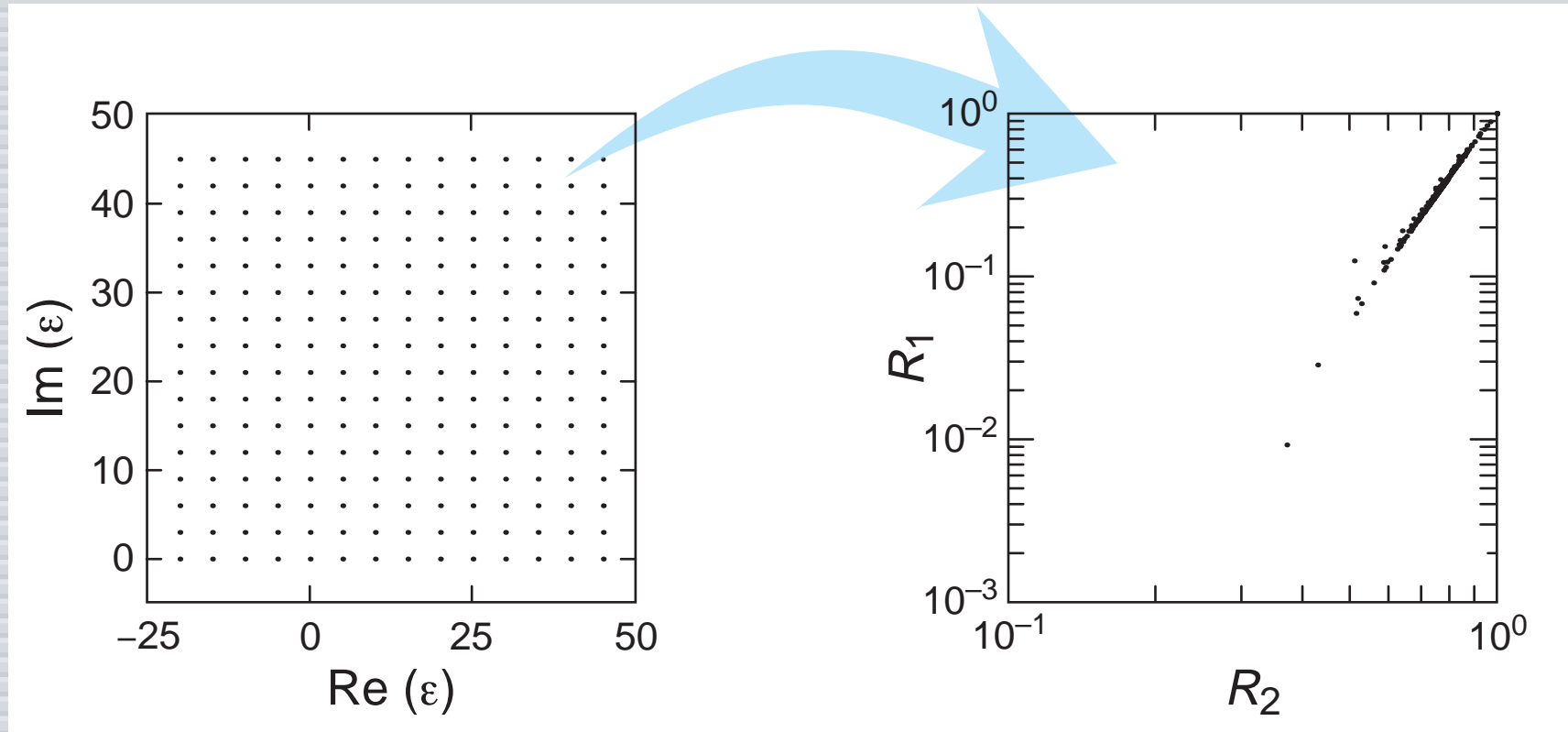
choice of angles



need numerical inversion

Technique

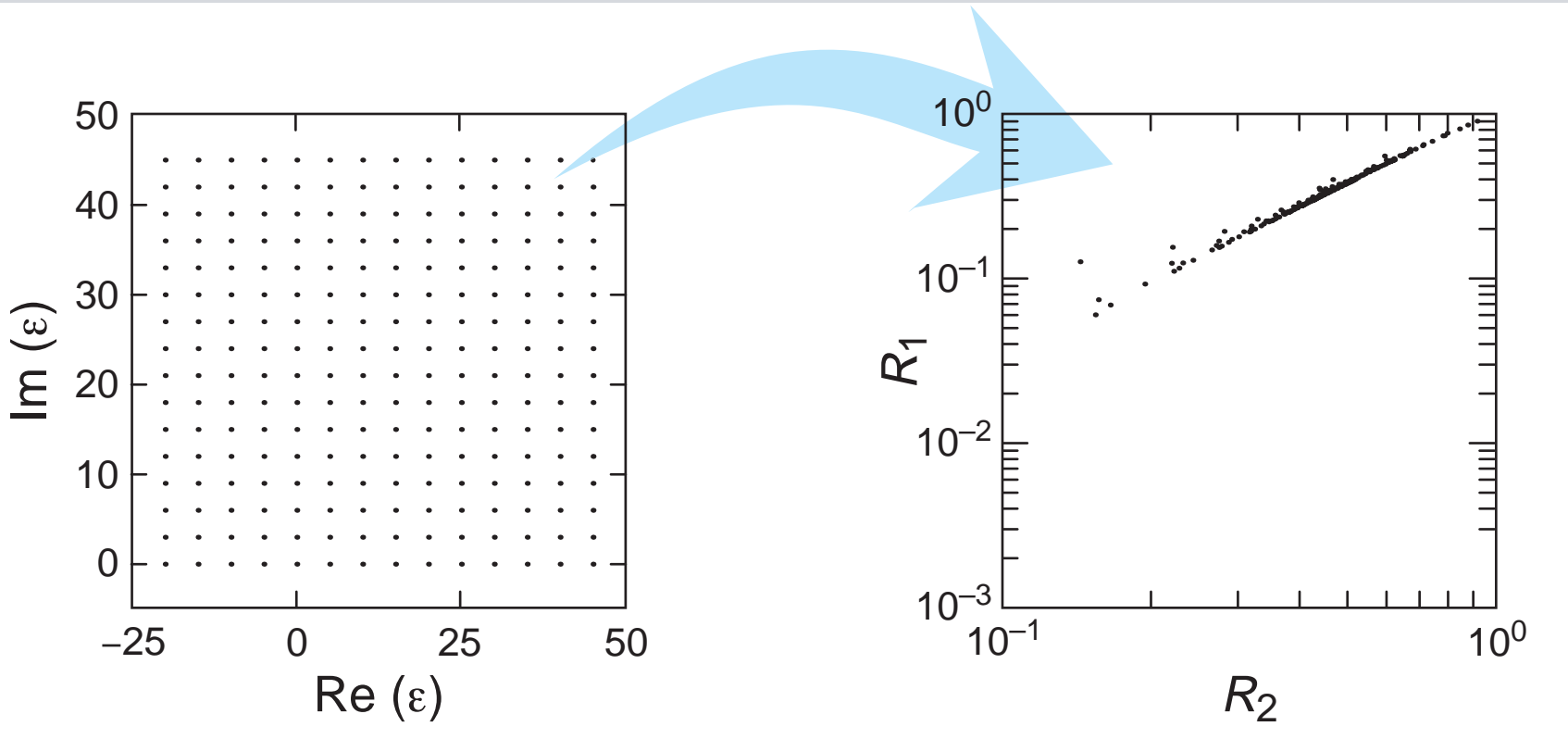
choice of angles



$$R_1 = 45^\circ \text{ p-pol}, R_2 = 45^\circ \text{ s-pol}$$

Technique

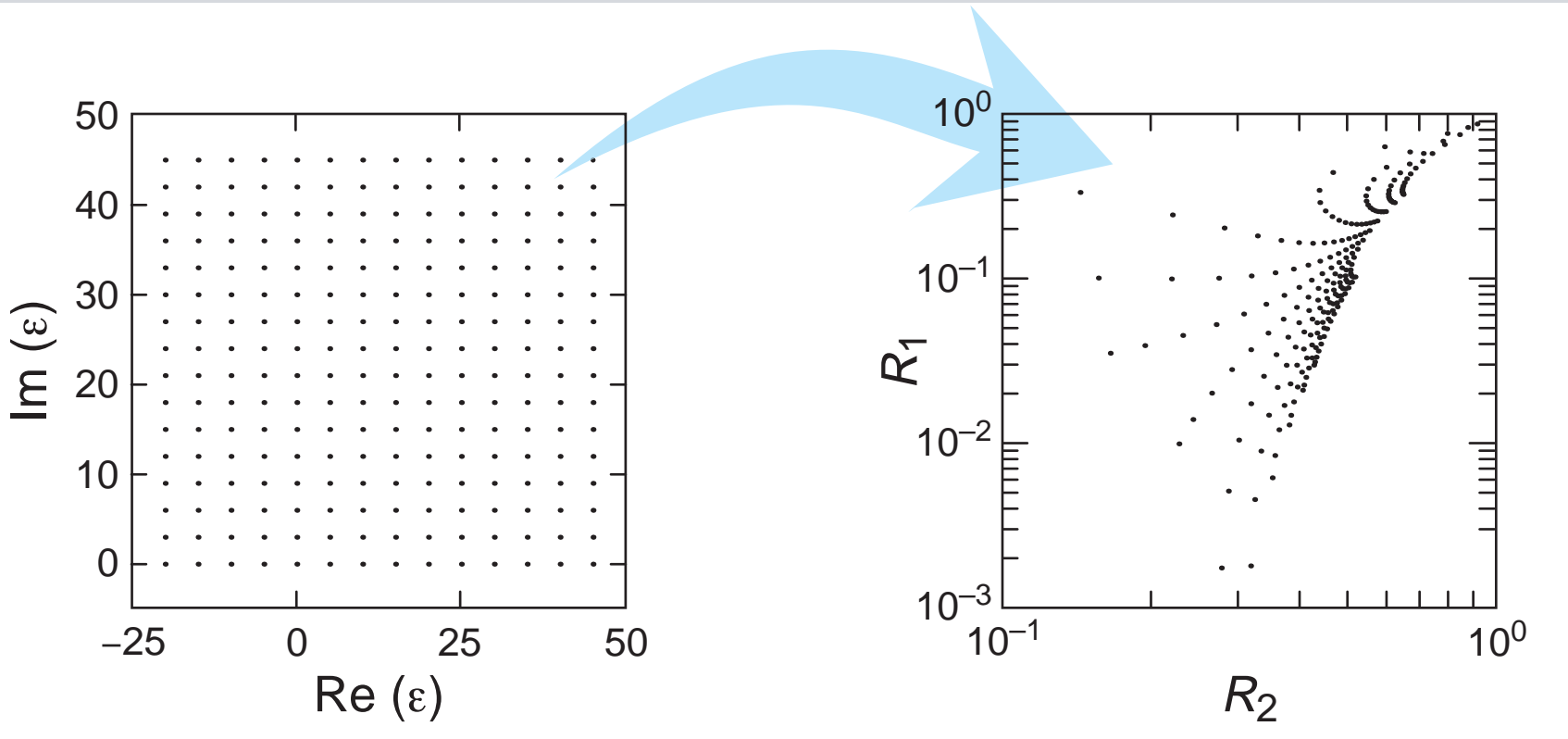
choice of angles



$$R_1 = 60^\circ \text{ p-pol}, R_2 = 45^\circ \text{ p-pol}$$

Technique

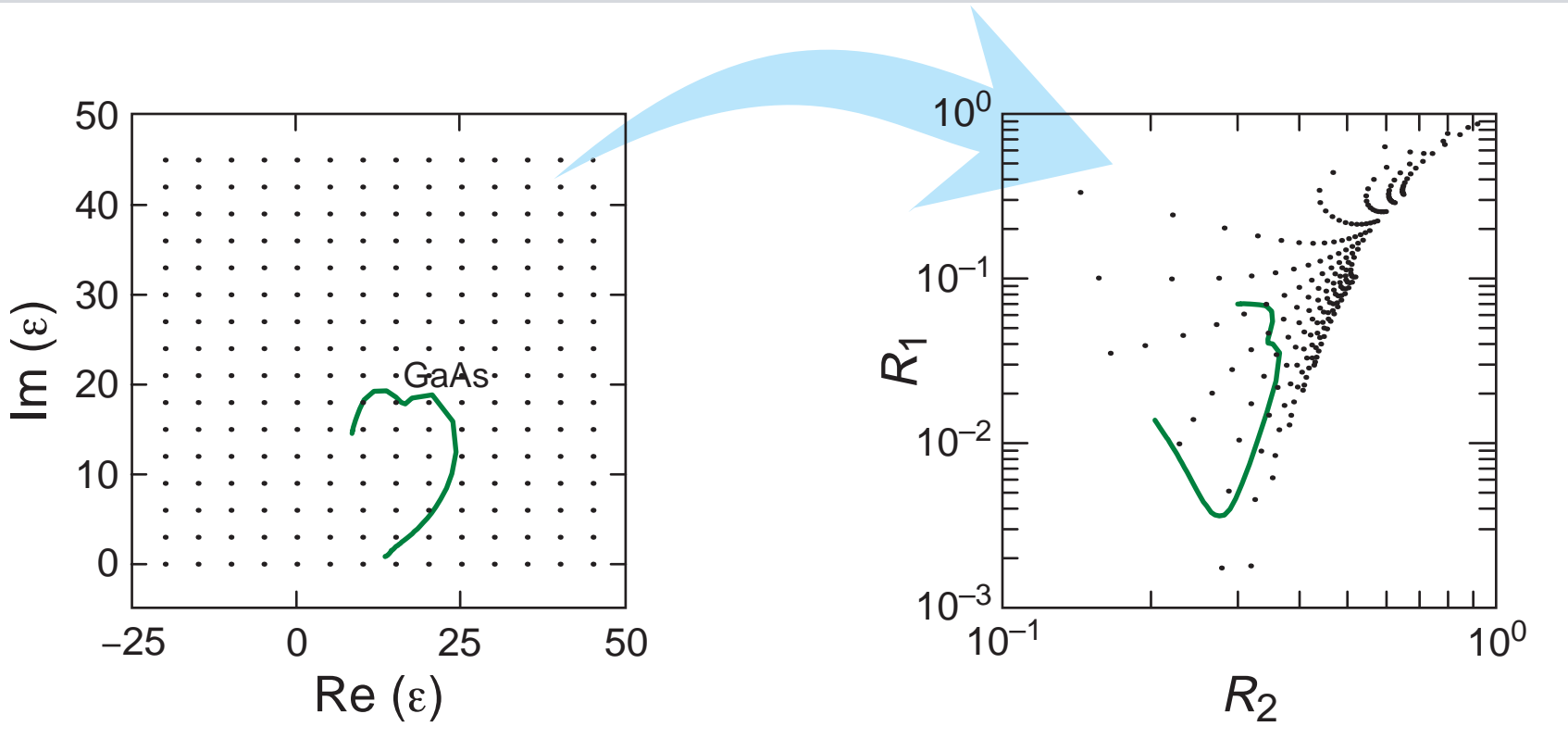
choice of angles



$$R_1 = 78^\circ \text{ p-pol}, R_2 = 45^\circ \text{ p-pol}$$

Technique

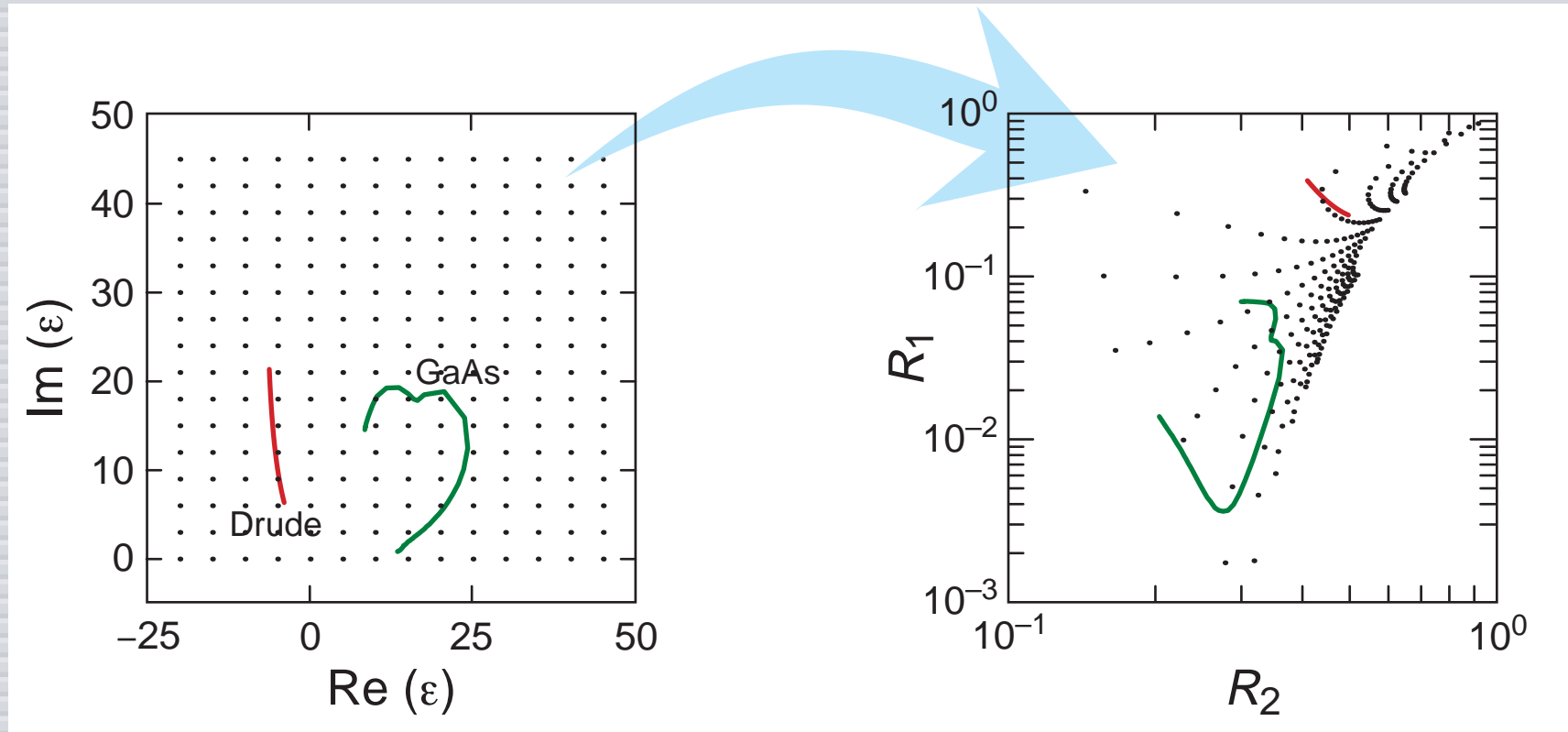
choice of angles



$$R_1 = 78^\circ \text{ p-pol}, R_2 = 45^\circ \text{ p-pol}$$

Technique

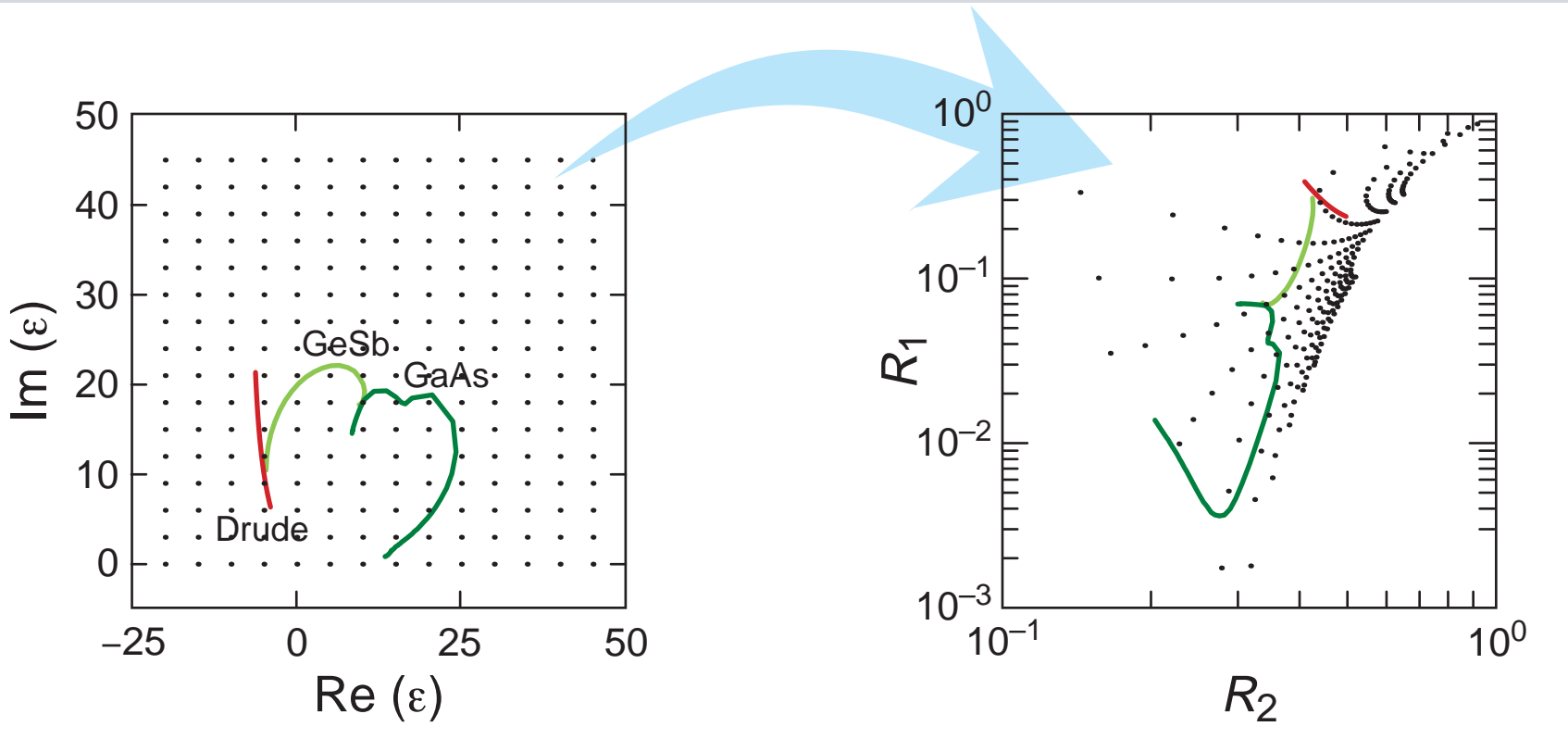
choice of angles



$$R_1 = 78^\circ \text{ p-pol}, R_2 = 45^\circ \text{ p-pol}$$

Technique

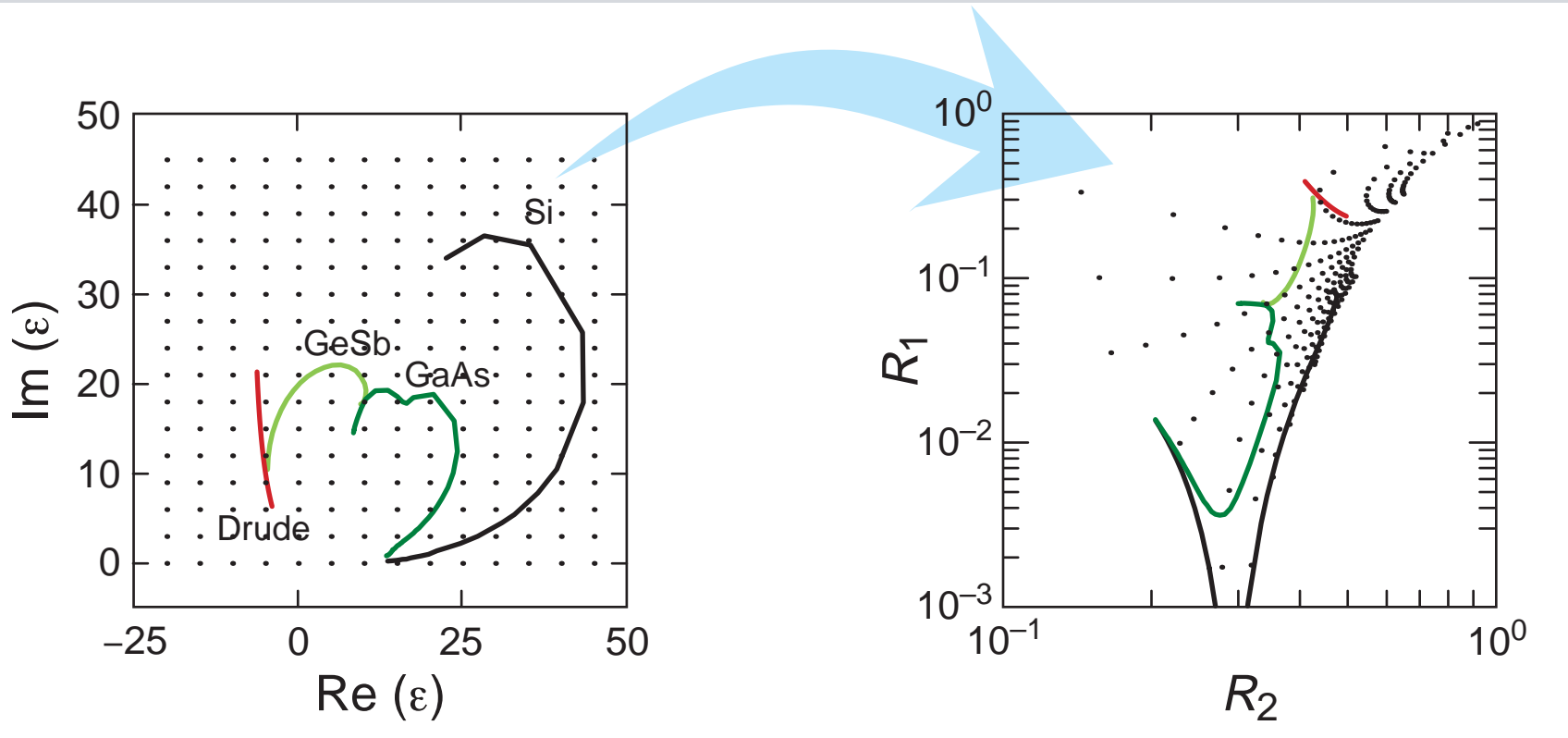
choice of angles



$$R_1 = 78^\circ \text{ p-pol}, R_2 = 45^\circ \text{ p-pol}$$

Technique

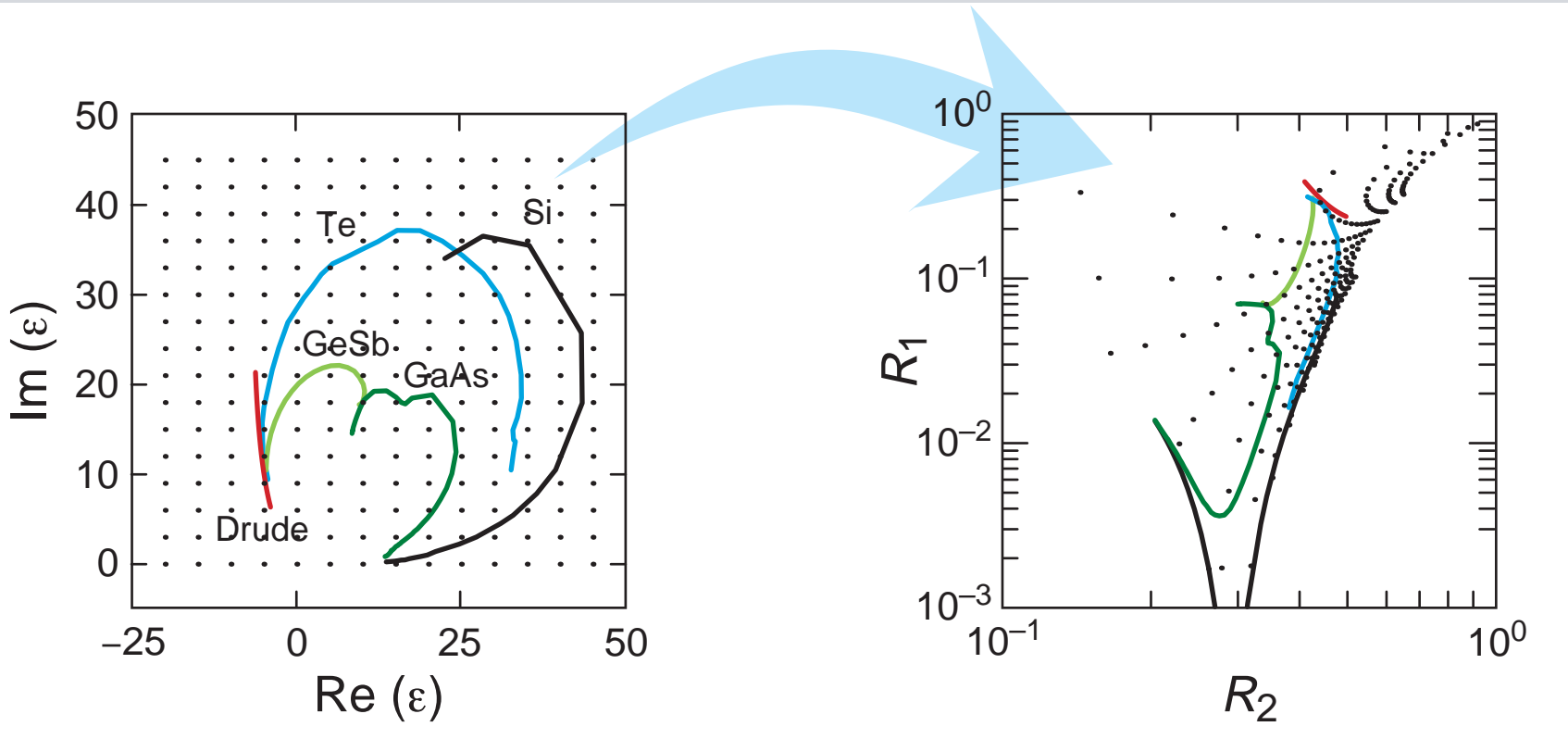
choice of angles



$$R_1 = 78^\circ \text{ p-pol}, R_2 = 45^\circ \text{ p-pol}$$

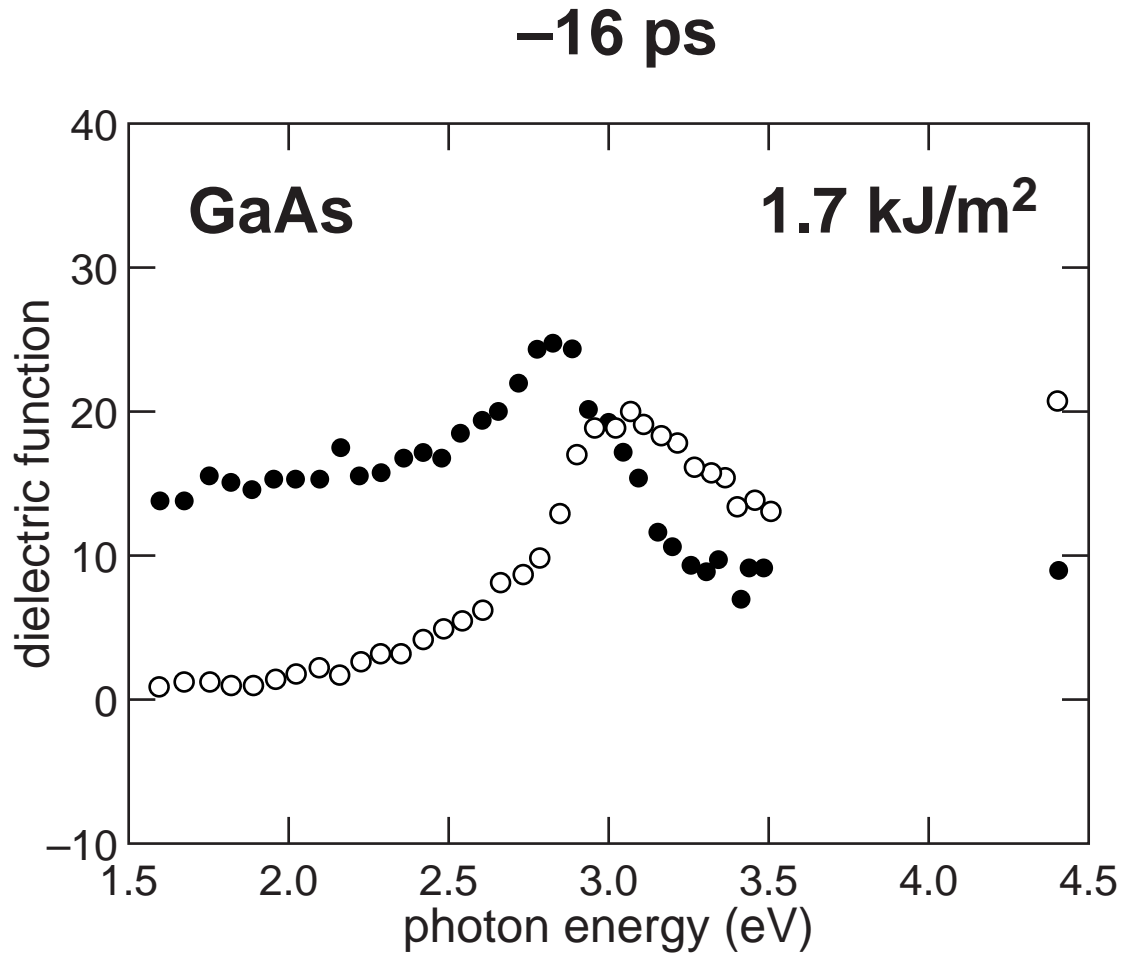
Technique

choice of angles

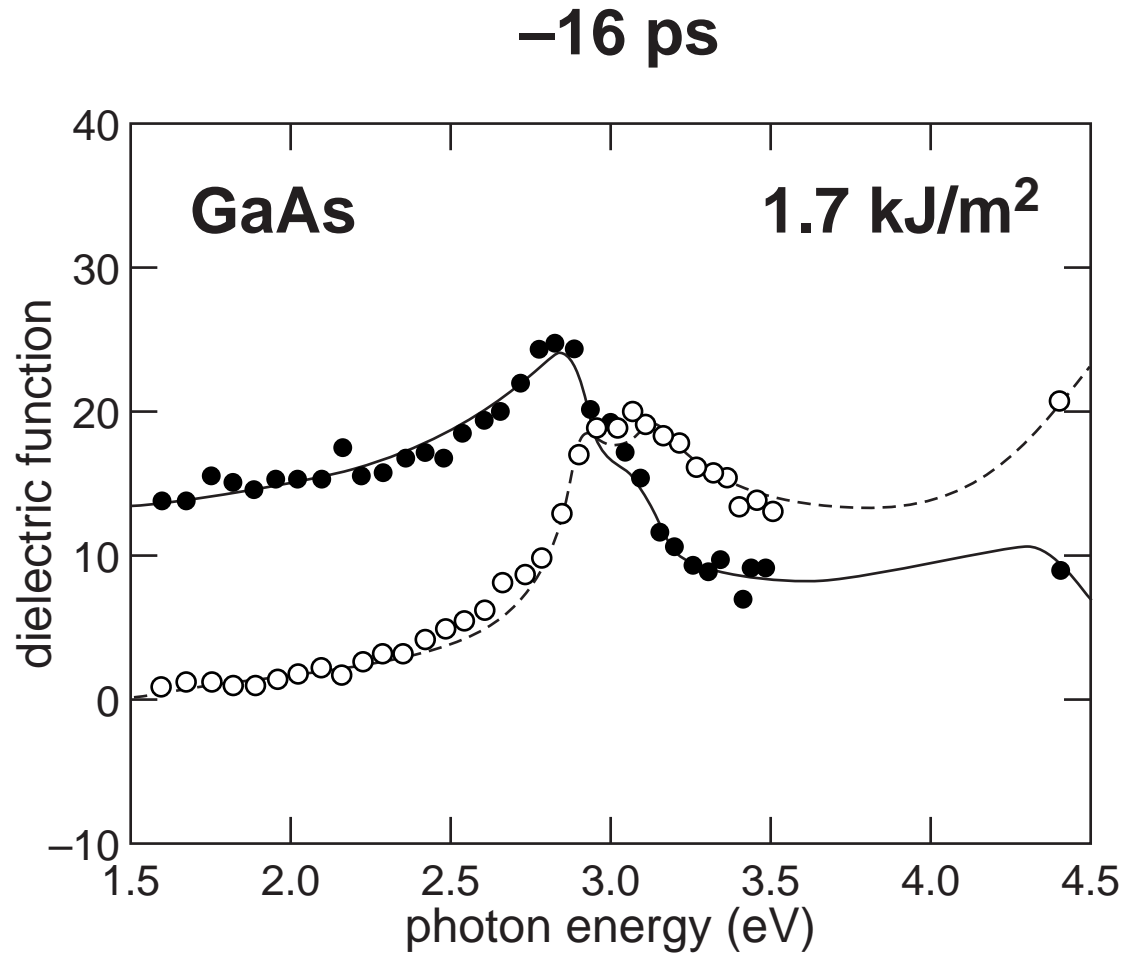


$$R_1 = 78^\circ \text{ p-pol}, R_2 = 45^\circ \text{ p-pol}$$

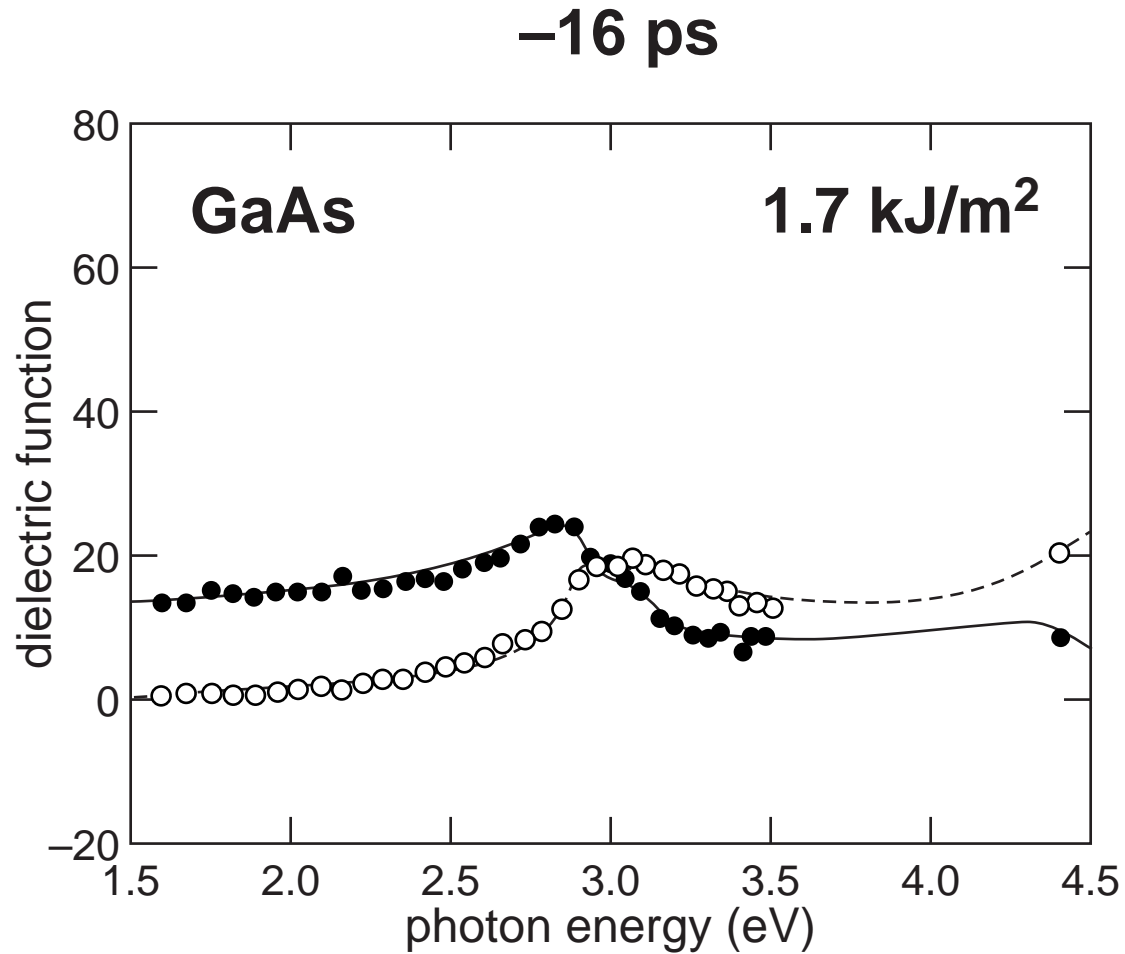
Technique



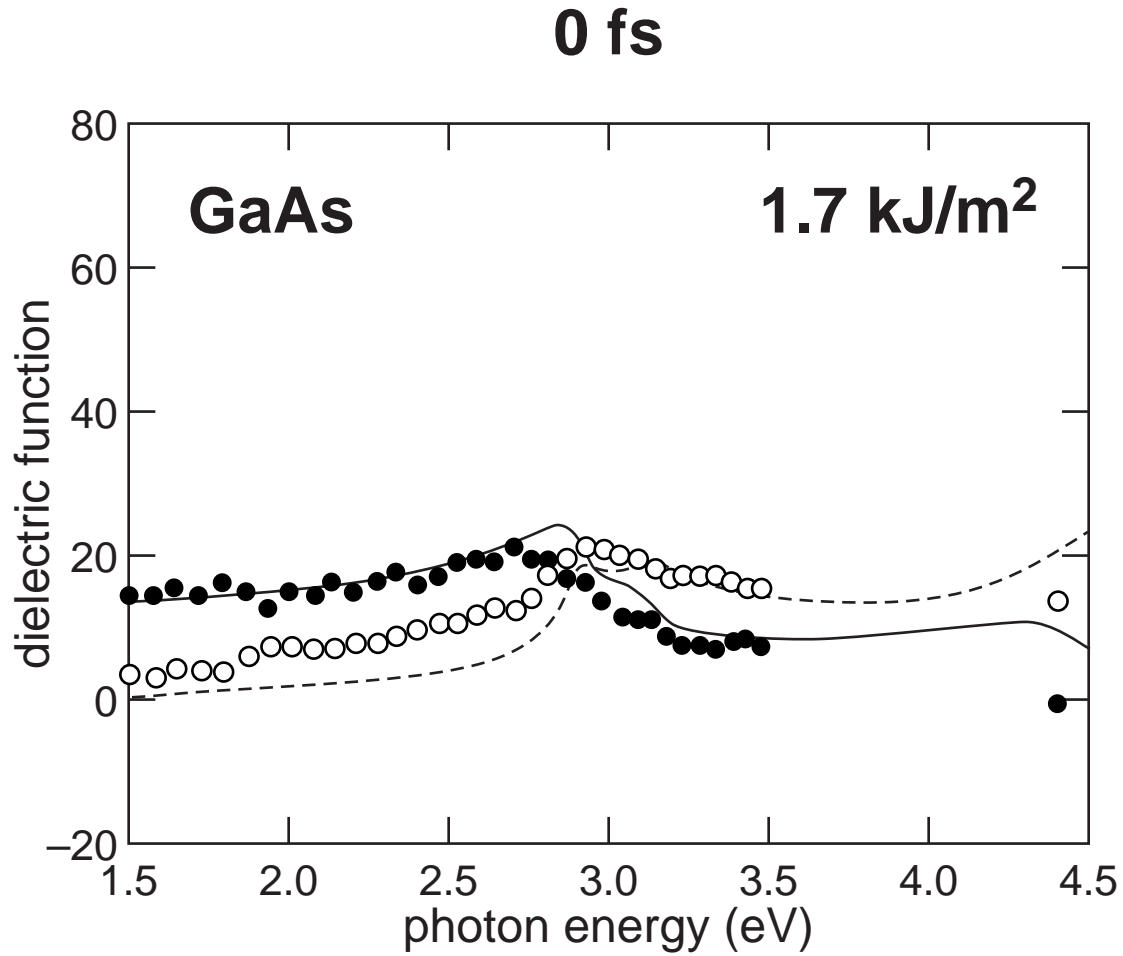
Technique



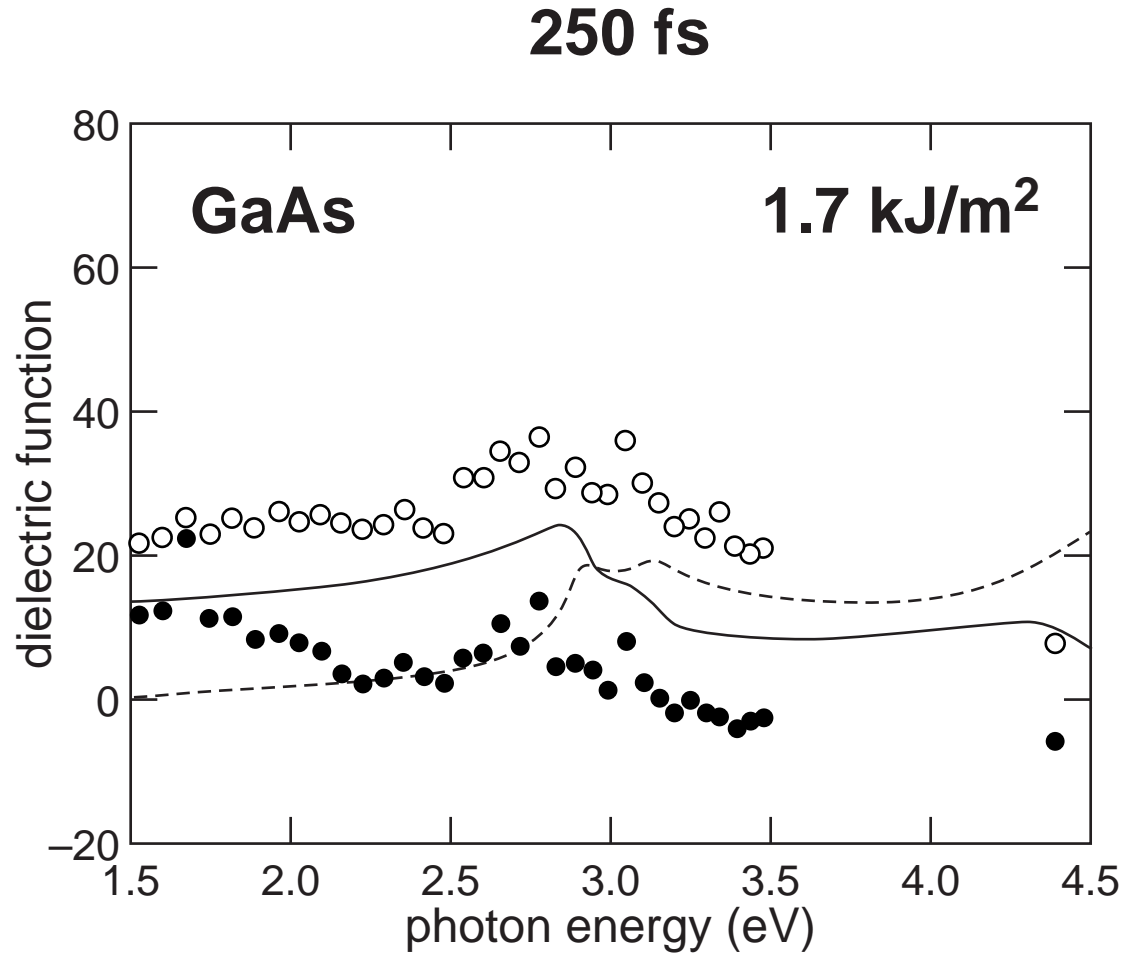
Technique



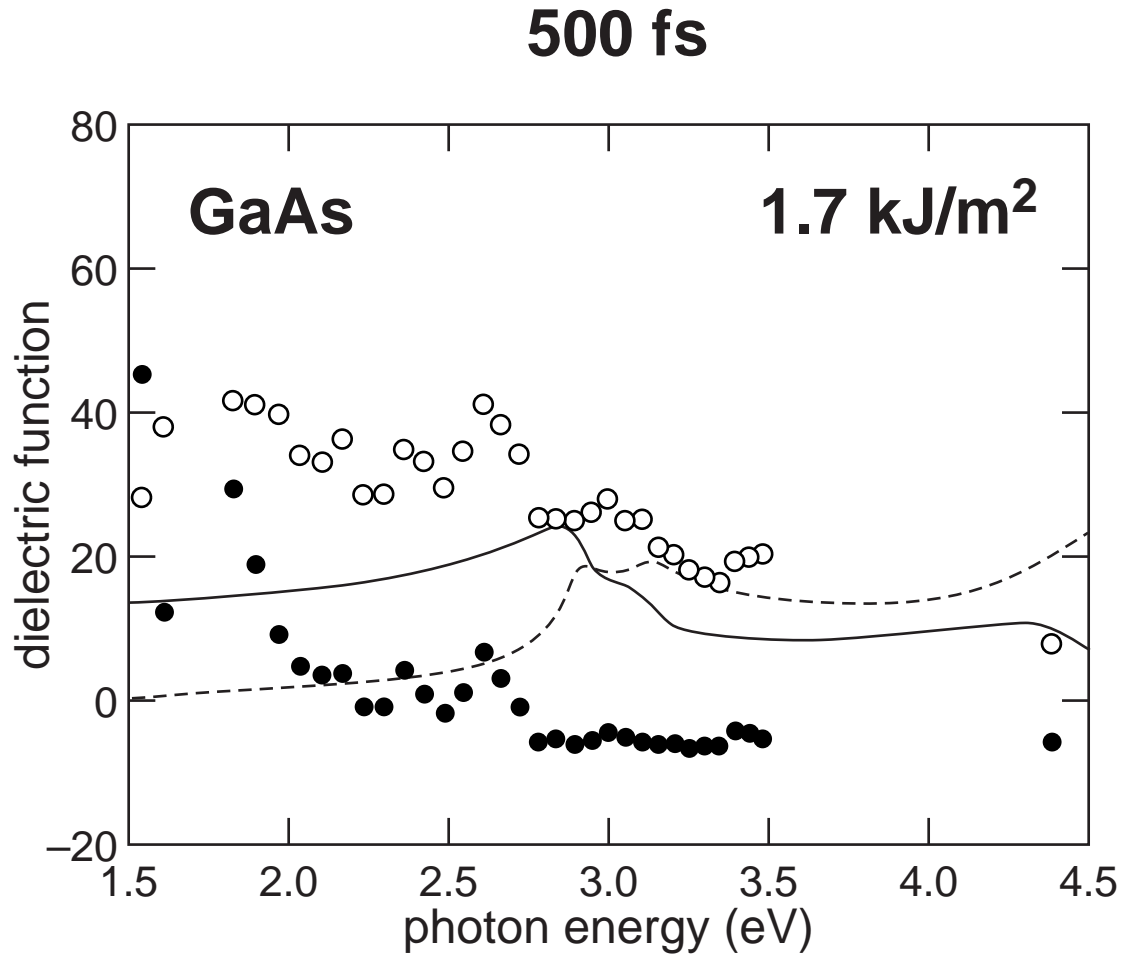
Technique



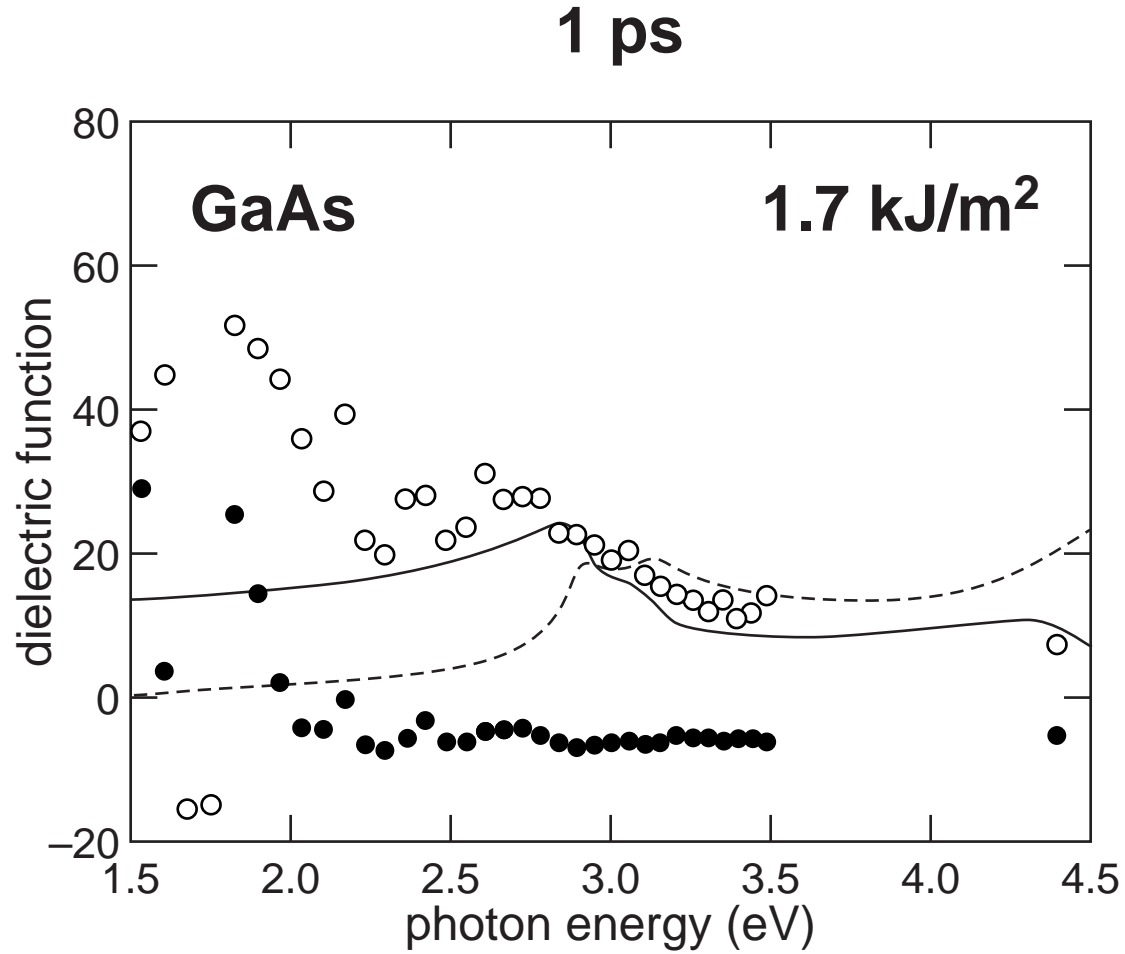
Technique



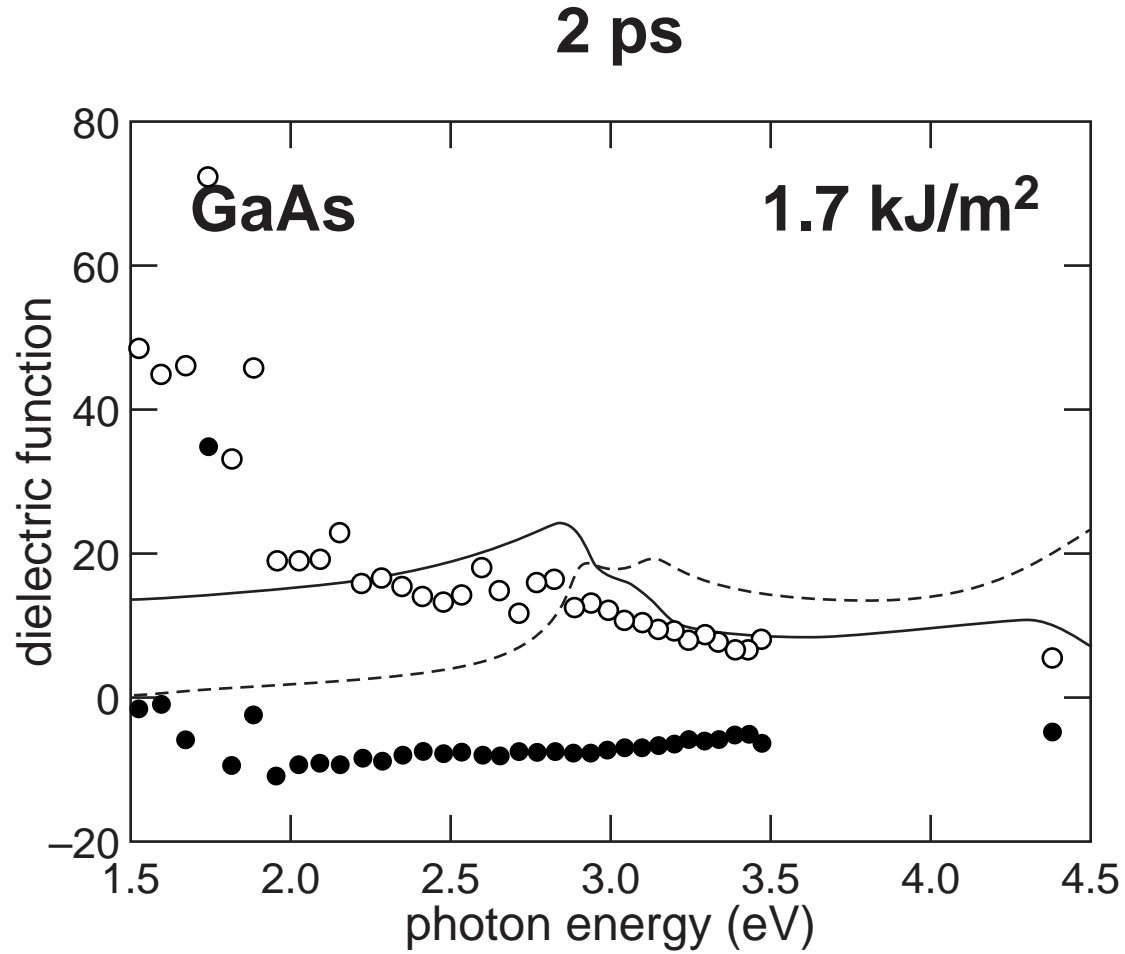
Technique



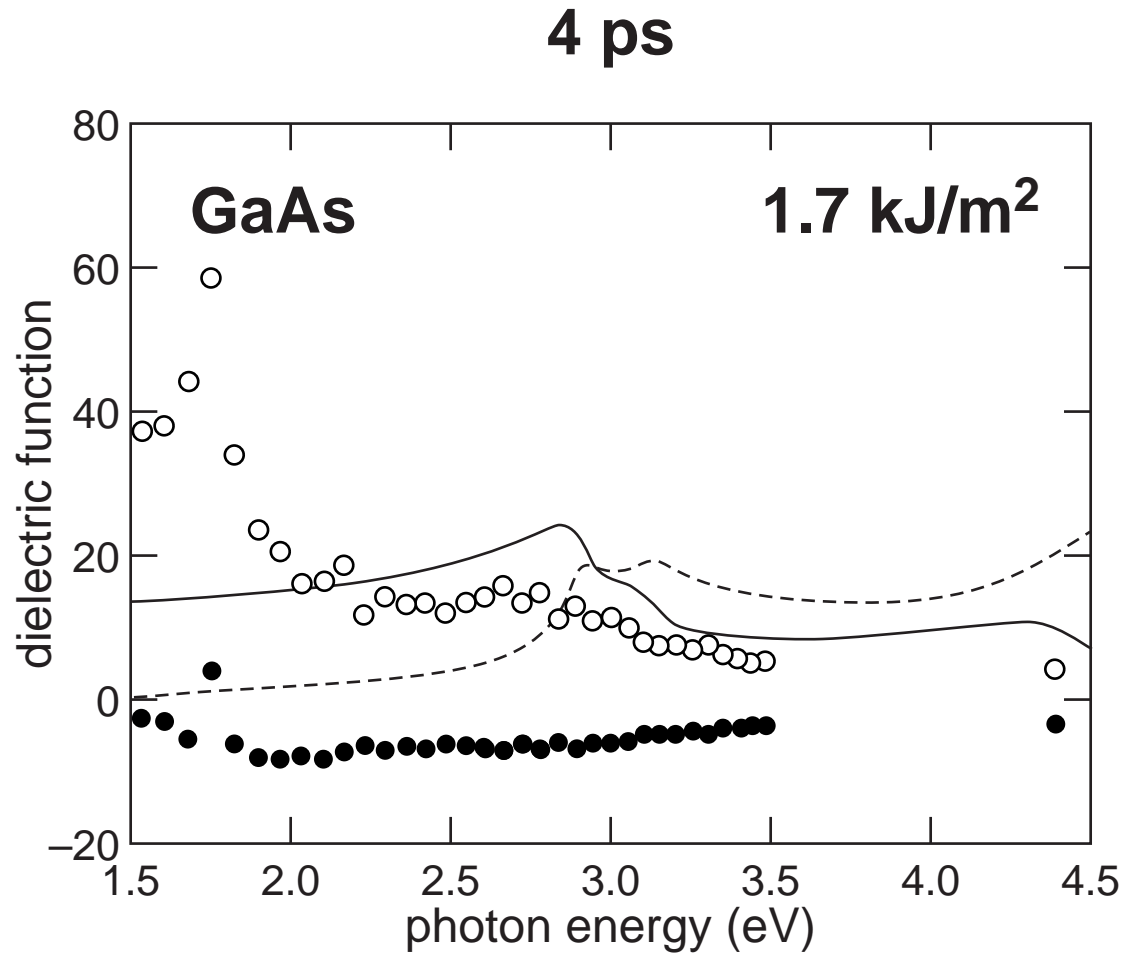
Technique



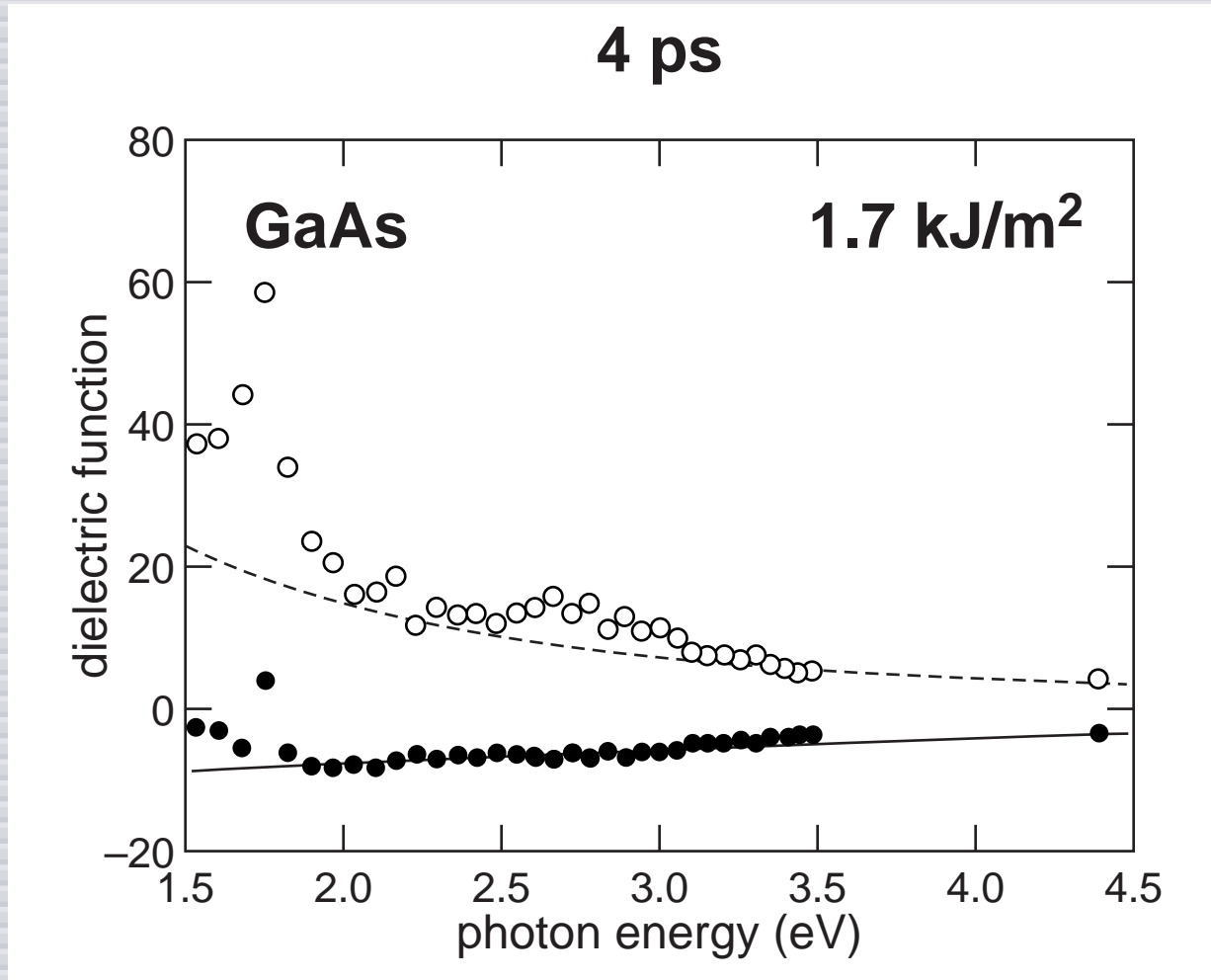
Technique



Technique

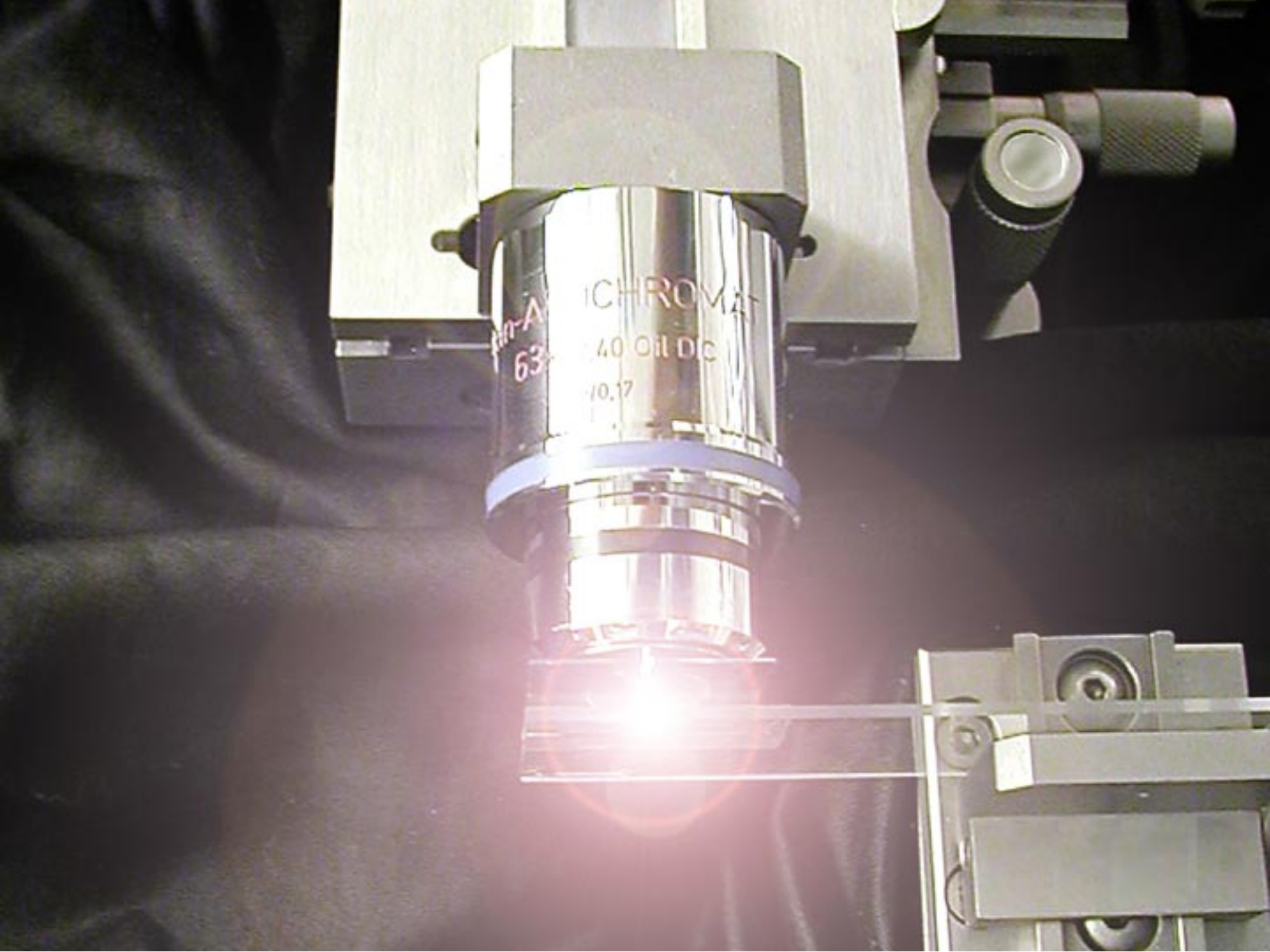


Technique



Technique

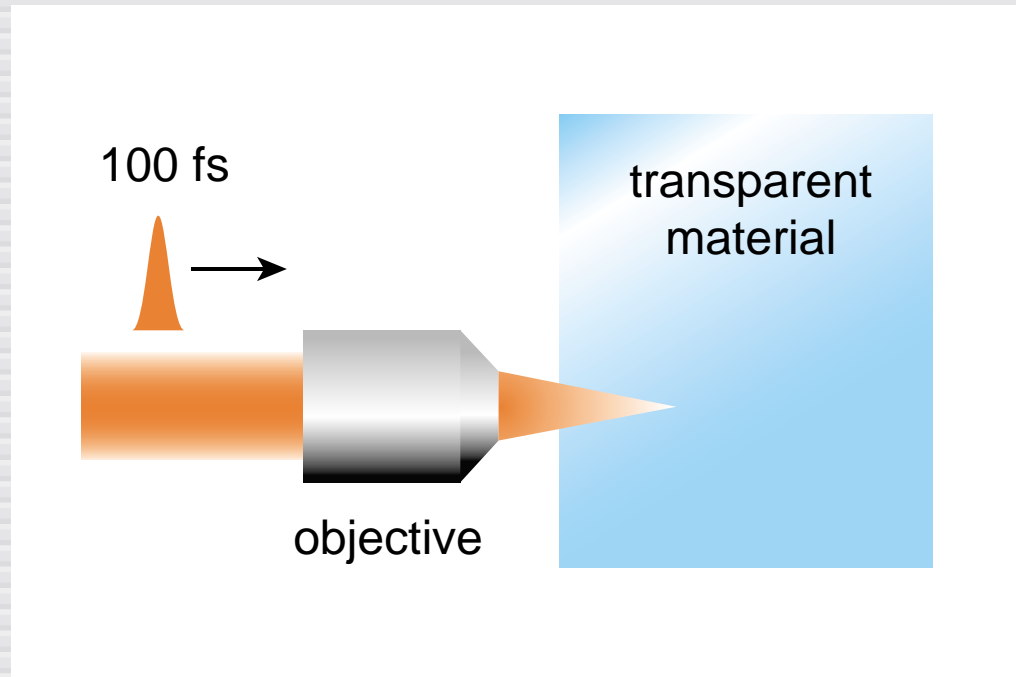
- ▶ **direct observation of semiconductor-to-metal transition**
- ▶ **order-disorder transition**
- ▶ **transition structural, not electronic**



10x/0.25

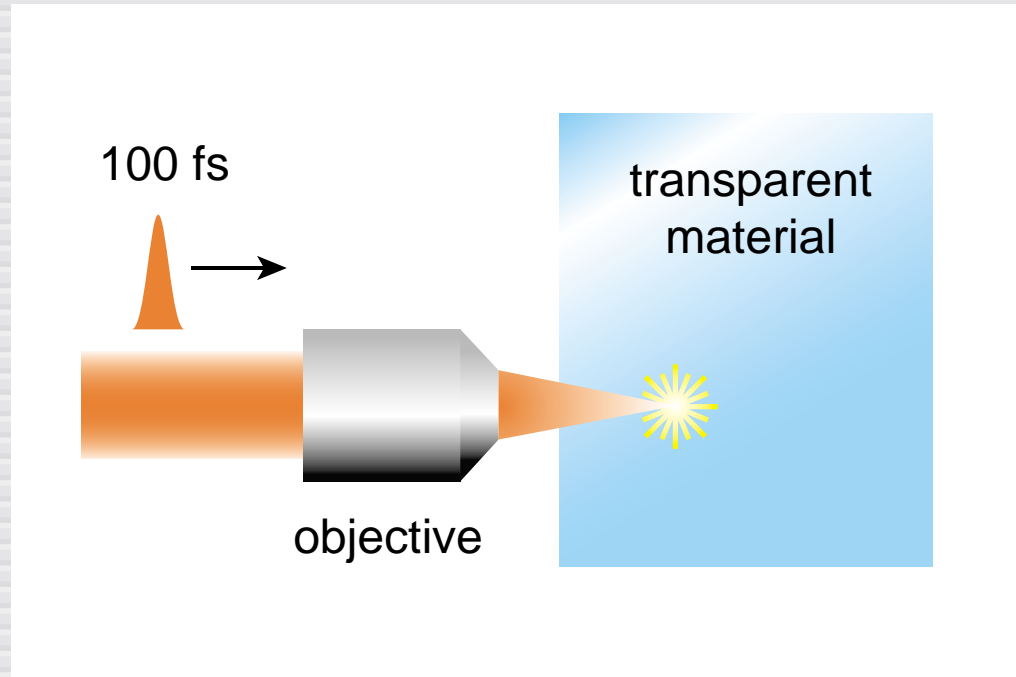
ACHROMAT
40 Oil DC
10.17

Processing with fs pulses



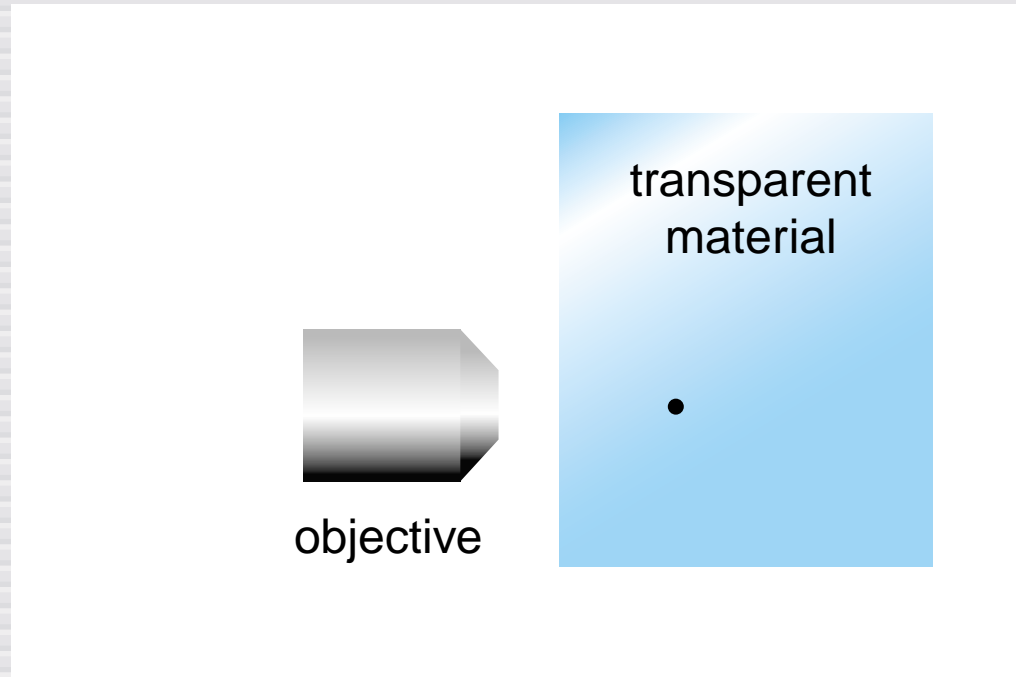
high intensity at focus...

Processing with fs pulses



... causes nonlinear ionization...

Processing with fs pulses



and 'microexplosion' causes microscopic damage

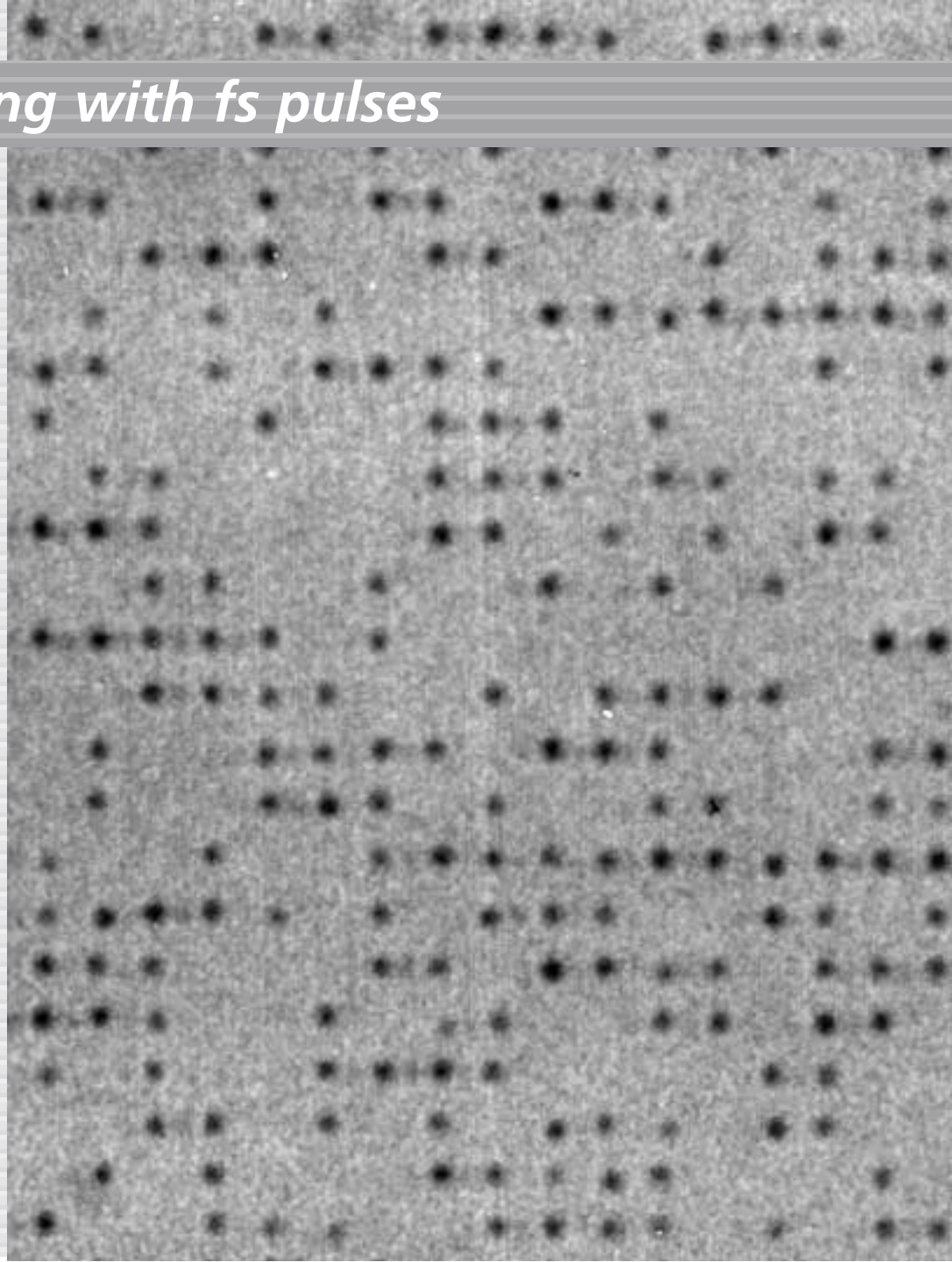
Processing with fs pulses

2 x 2 μm array

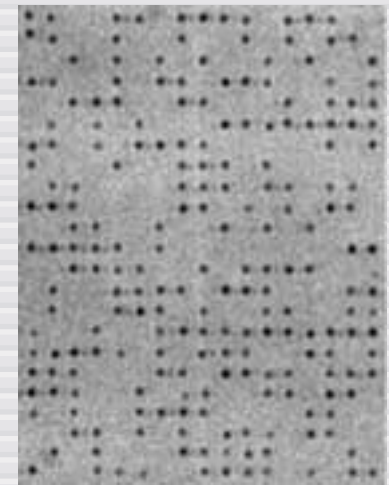
fused silica, 0.65 NA

0.5 μJ , 100 fs, 800 nm

***Opt. Lett.* 21, 2023 (1996)**



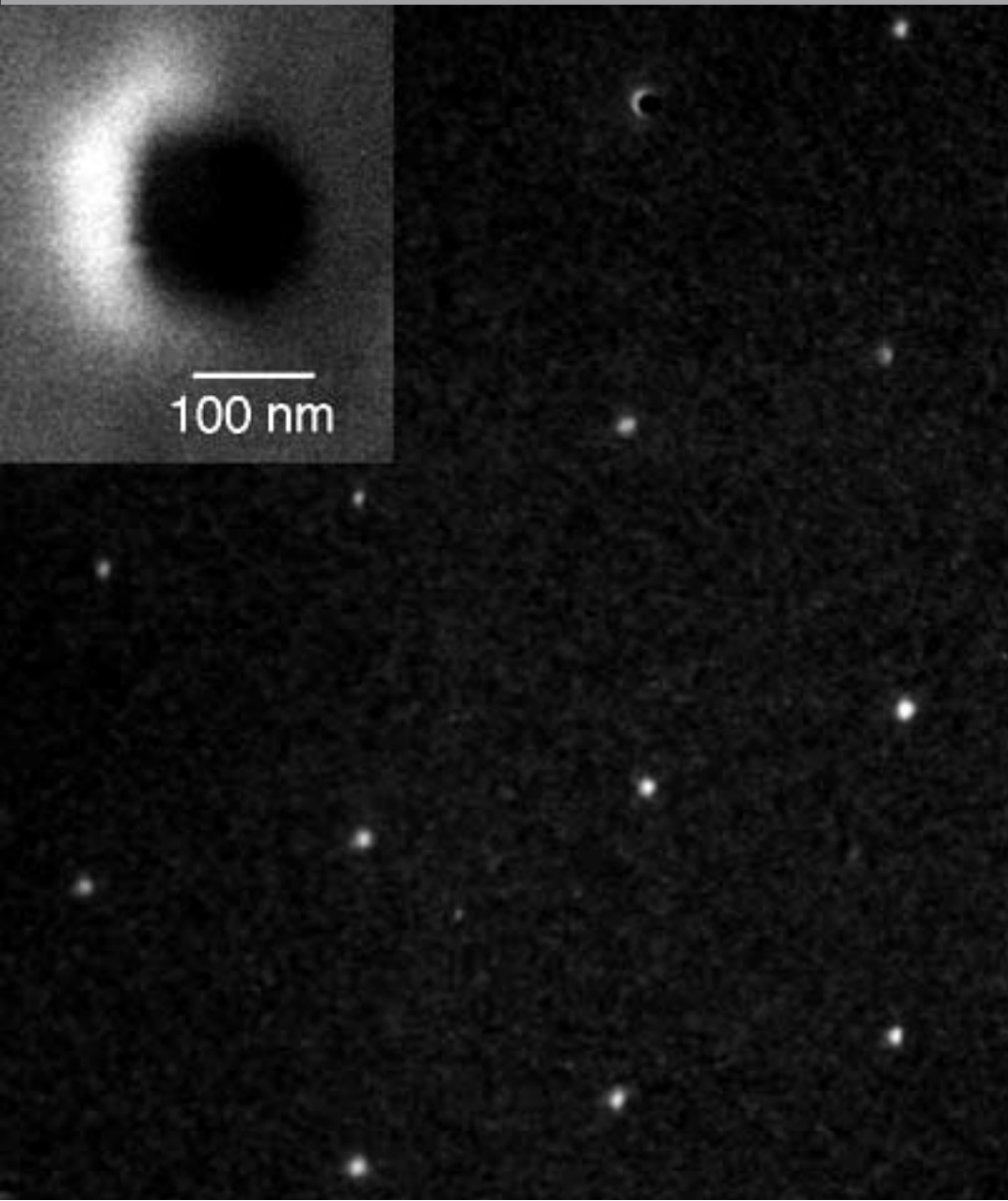
Processing with fs pulses



100 fs
0.5 μ J

200 ps
9 μ J

Processing with fs pulses



5 x 5 μm array

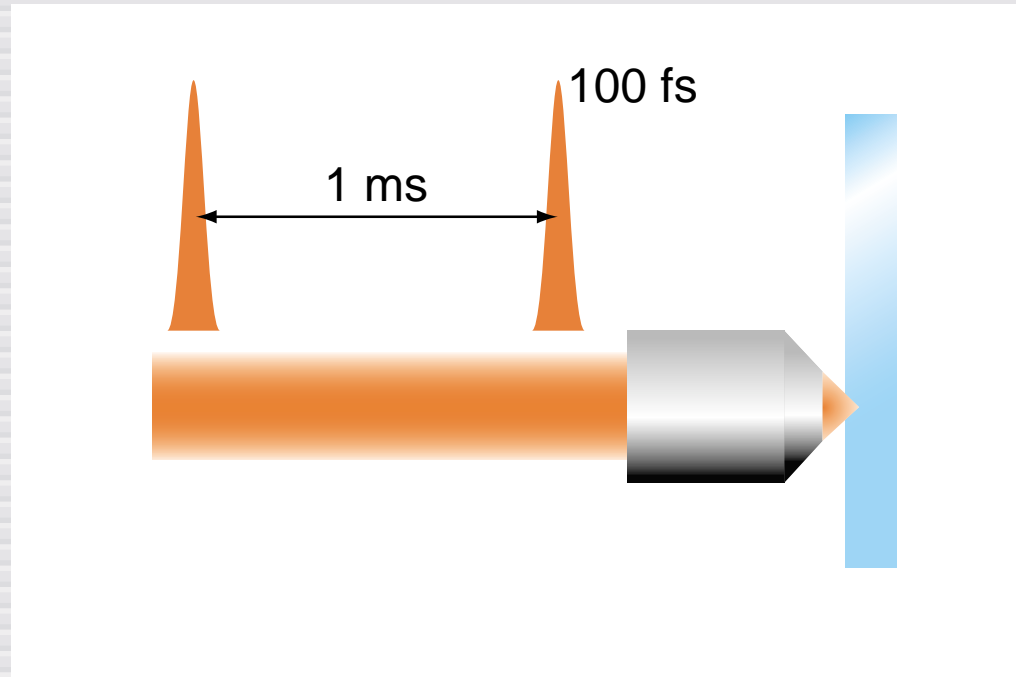
fused silica, 0.65 NA

0.5 μJ , 100 fs, 800 nm

Opt. Lett. 21, 2023 (1996)

Low-energy processing

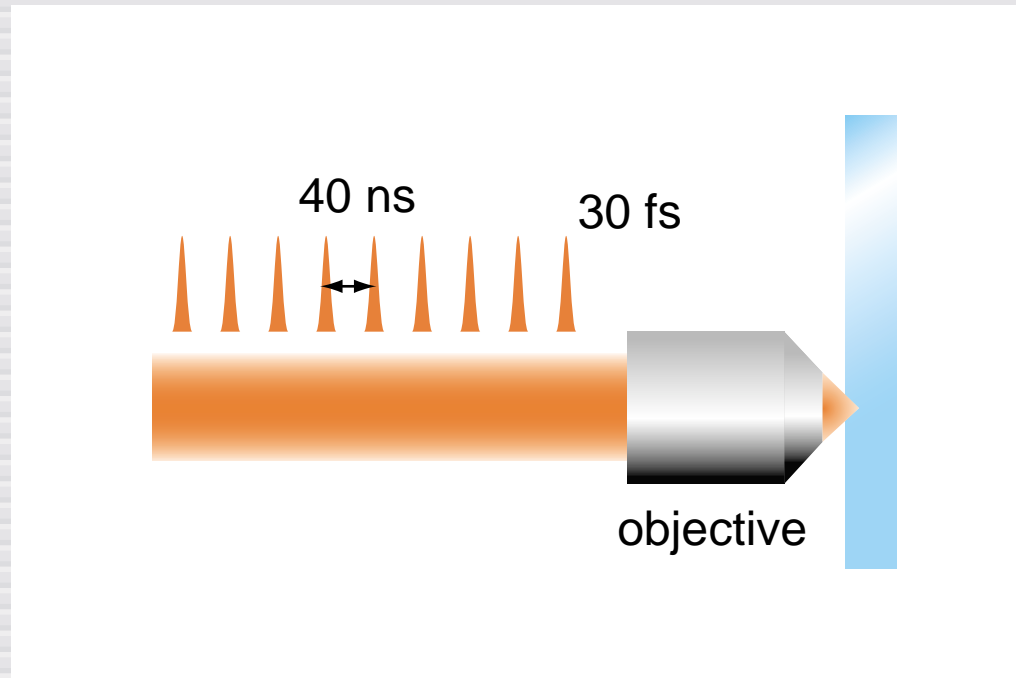
amplified laser



heat-diffusion time: $\tau_{diff} \approx 1 \mu s$

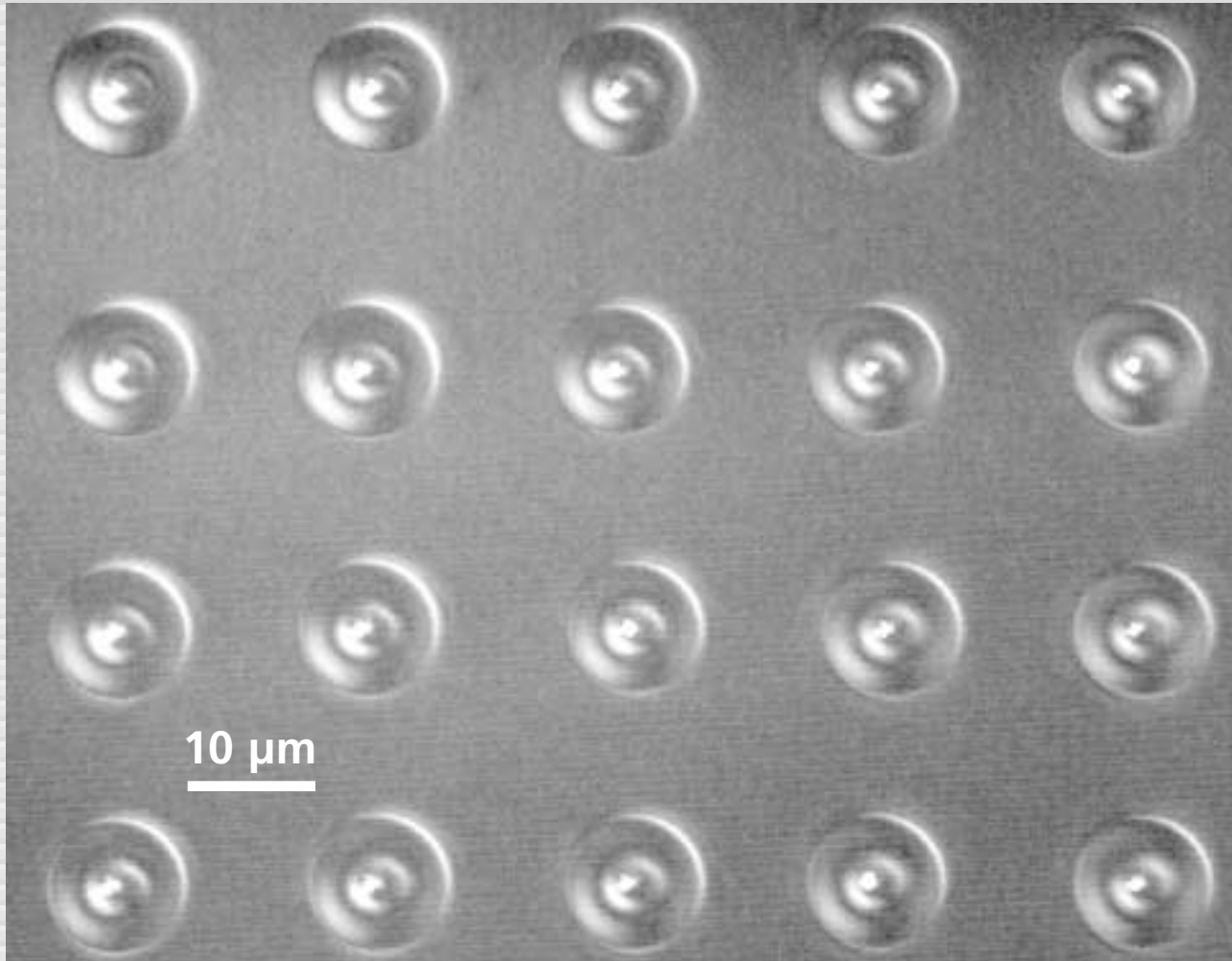
Low-energy processing

long-cavity Ti:sapphire oscillator

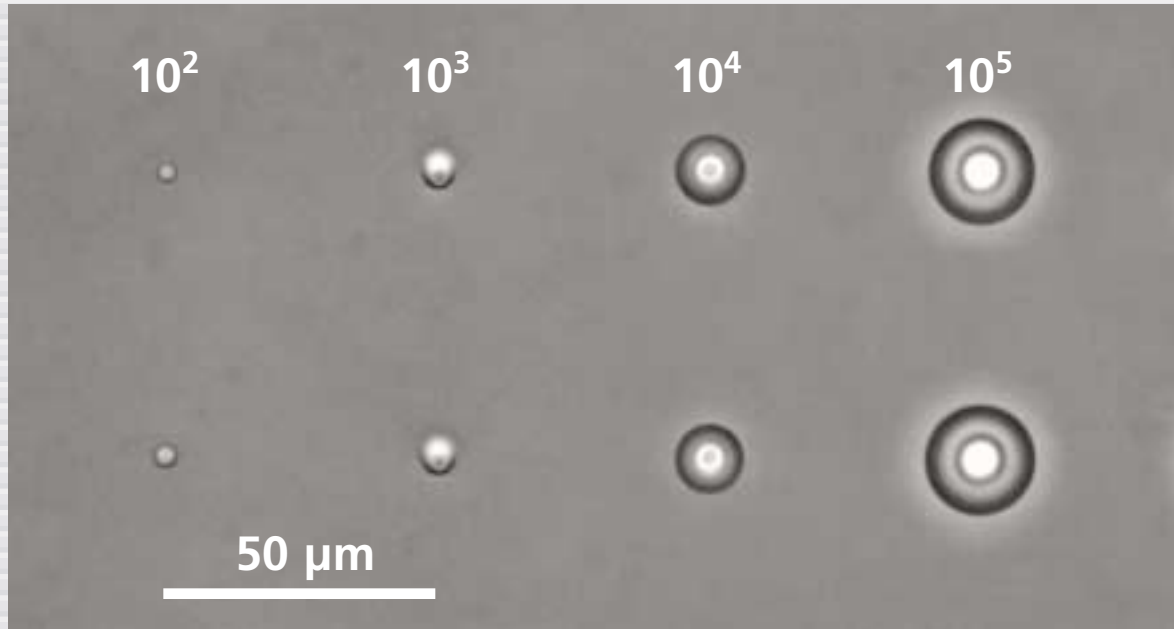


heat-diffusion time: $\tau_{diff} \approx 1 \mu\text{s}$

Low-energy processing

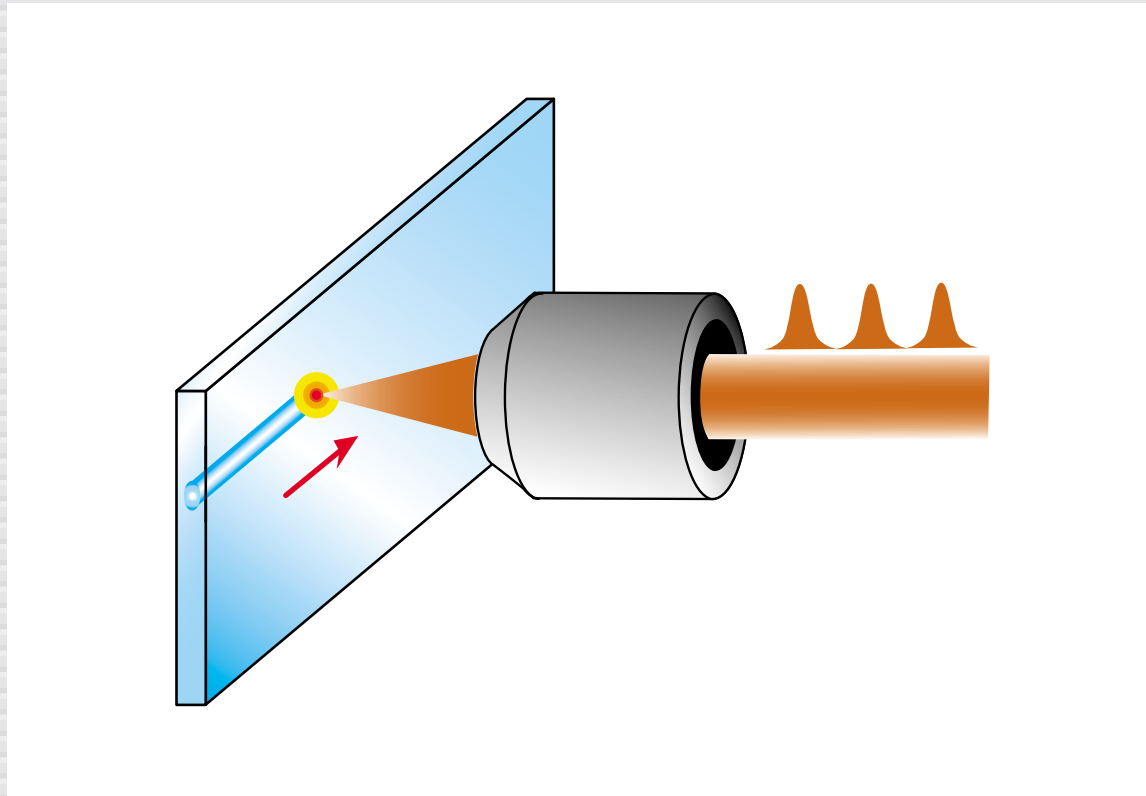


Low-energy processing



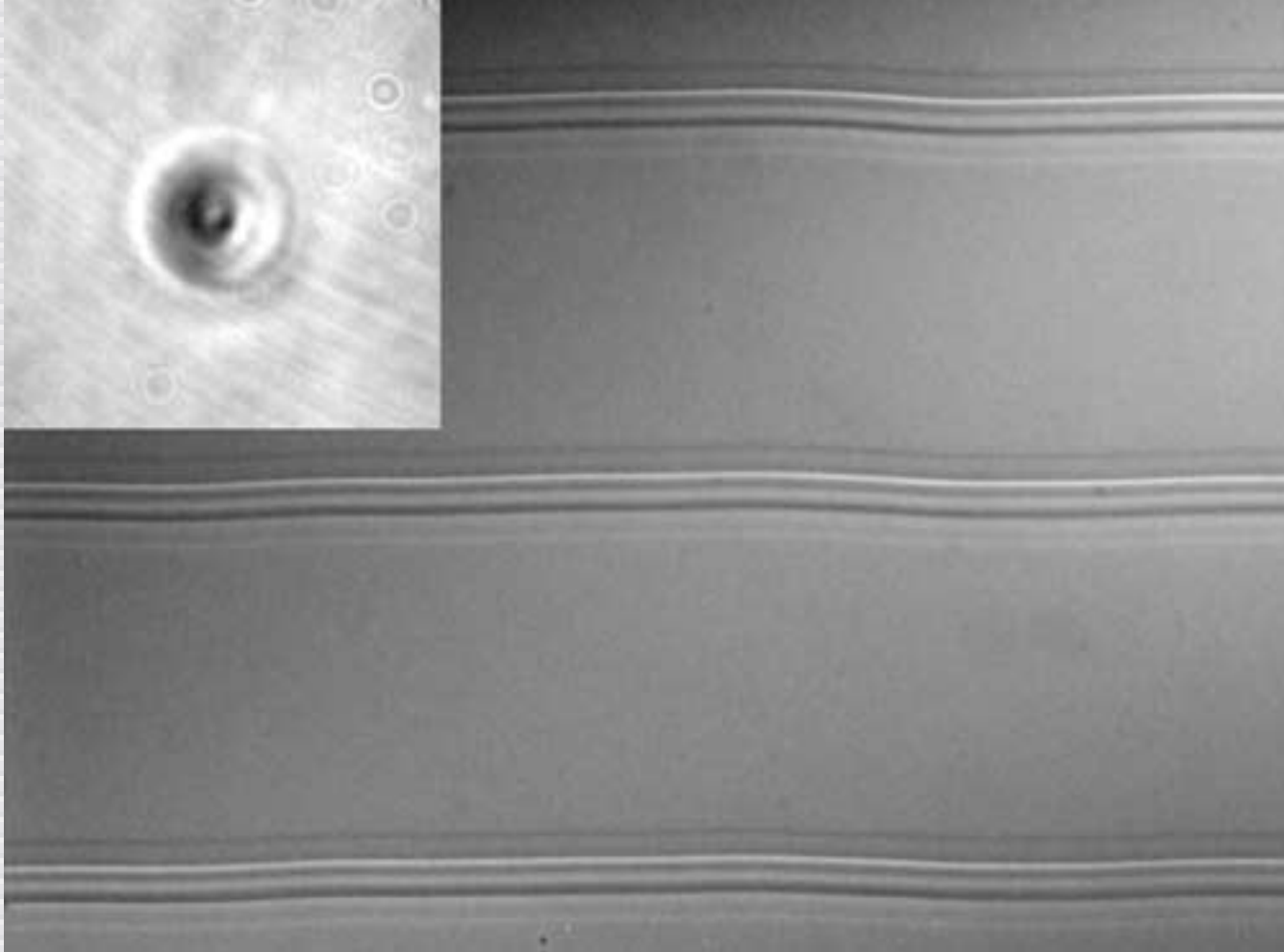
Low-energy processing

waveguide machining



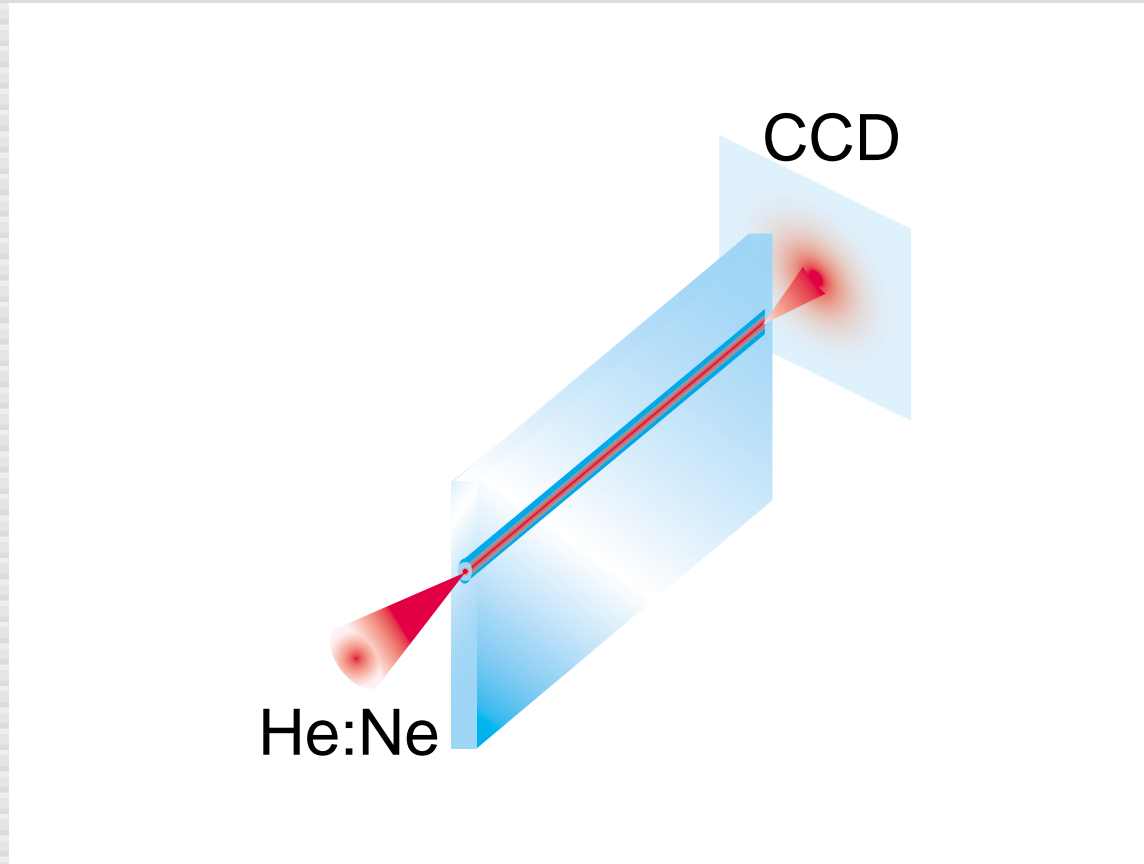
Low-energy processing

waveguide machining



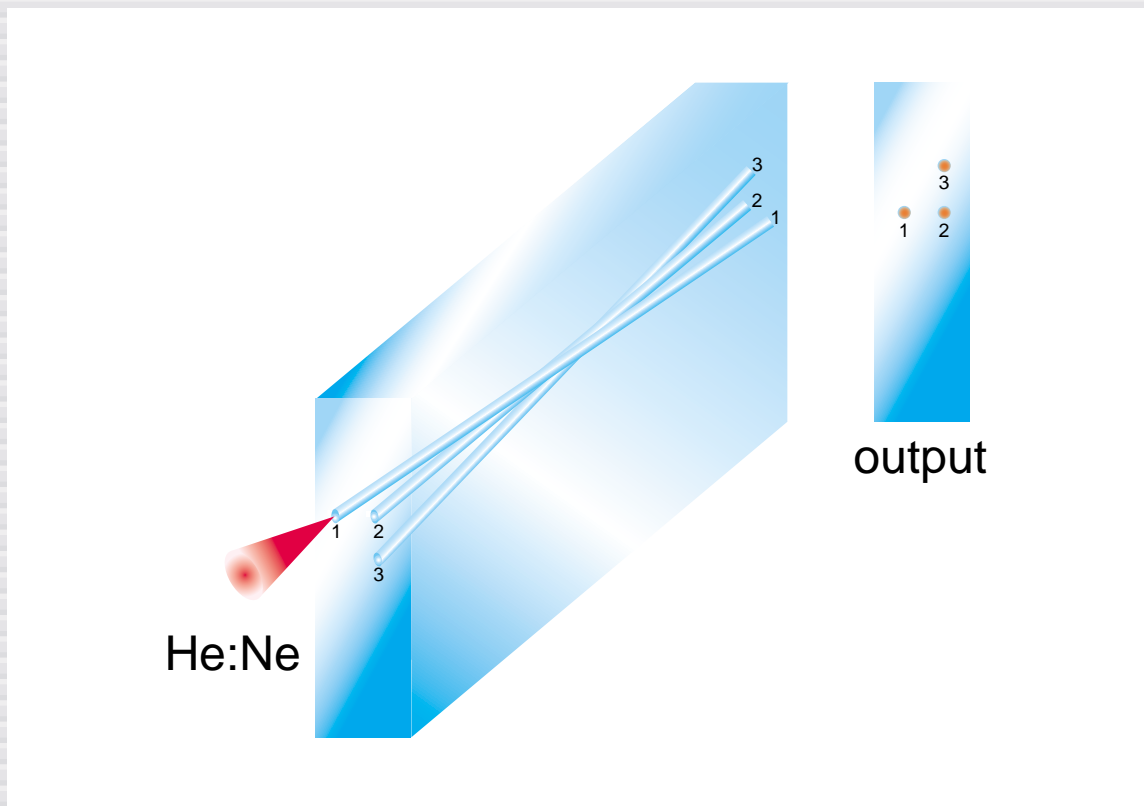
Low-energy processing

waveguide mode analysis



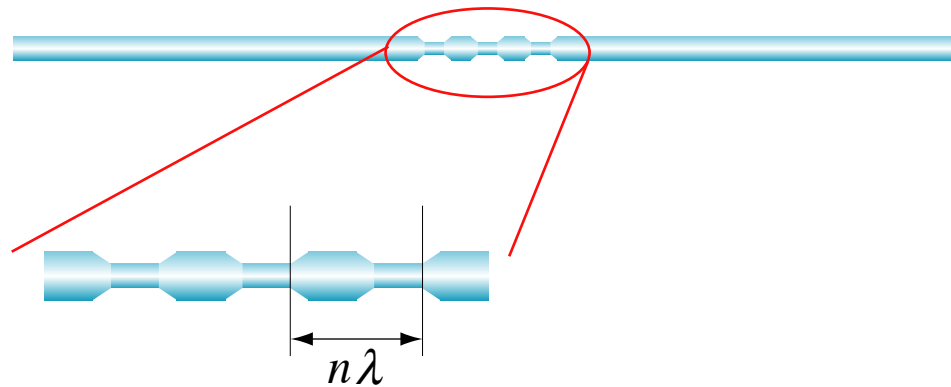
Low-energy processing

3D wave splitter



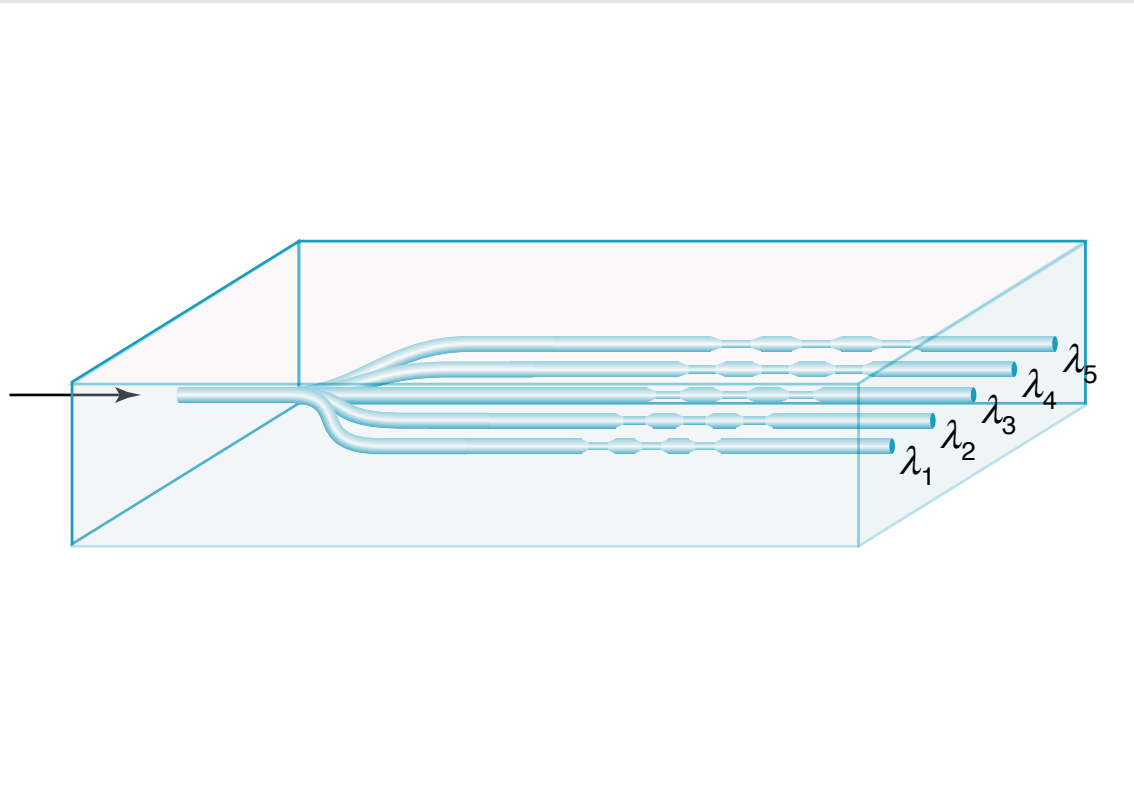
Low-energy processing

Bragg grating



Low-energy processing

Bragg grating



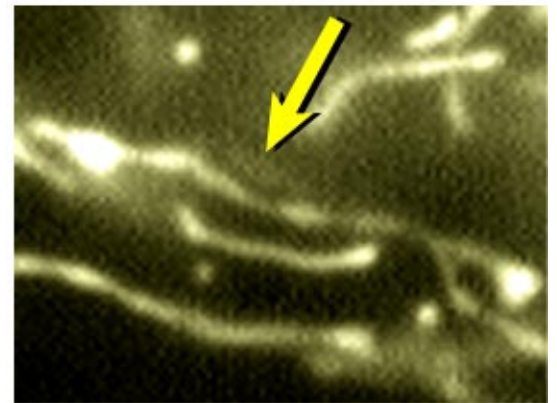
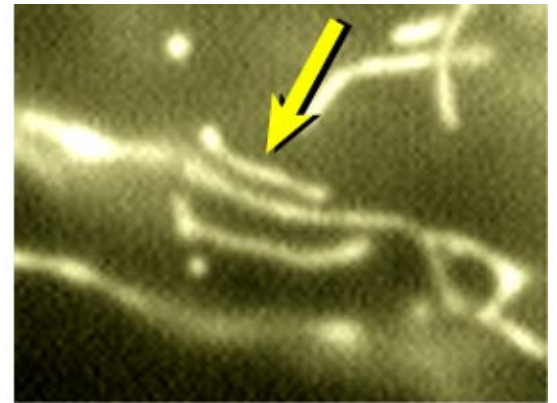
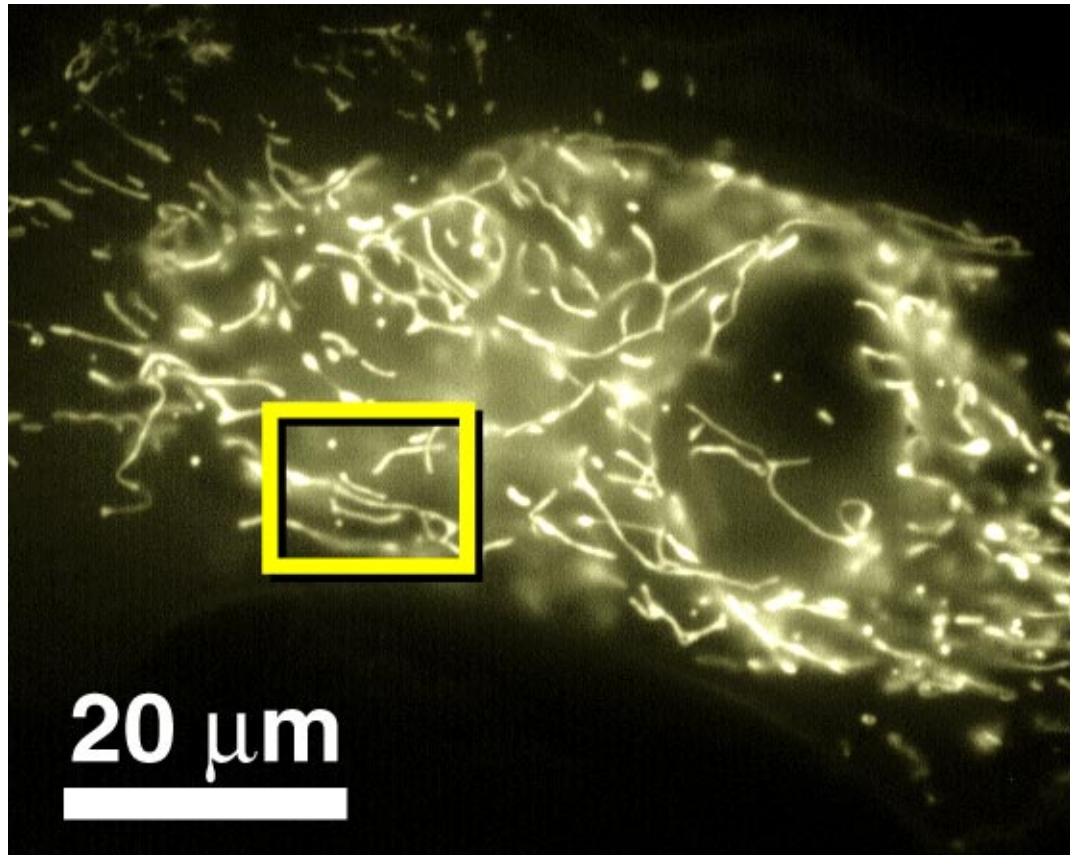
Low-energy processing

monolithic amplifier

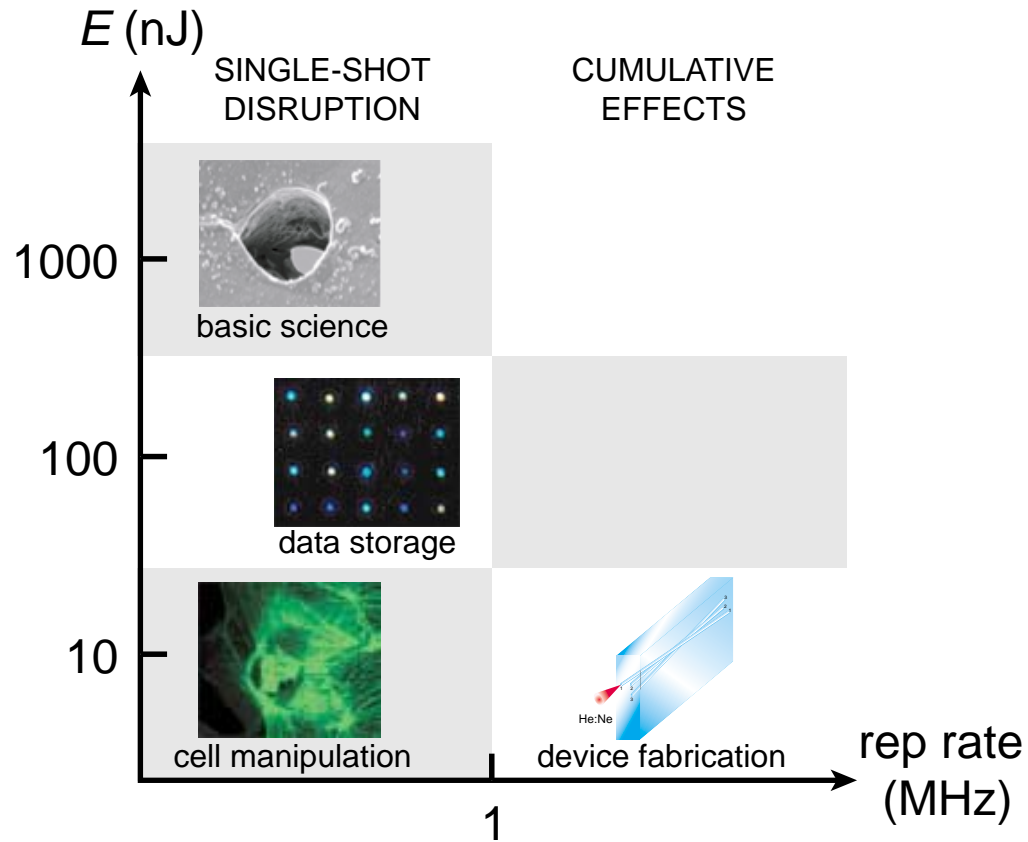


laser active glass

Low-energy processing



Summary



Conclusion

- ▶ **wiring optoelectronics circuits of the future**
- ▶ **manipulating the machinery of life**

Summary

Femtosecond laser pulses offer:

- ▶ **Unprecedented view into dynamics**
- ▶ **Extreme conditions with little energy**
- ▶ **New opportunities for research and processing**



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Prof. N. Bloembergen
Prof. H. Ehrenreich
Prof. T. Kaxiras**

**For a copy of this talk and
additional information, see:**

<http://mazur-www.harvard.edu>