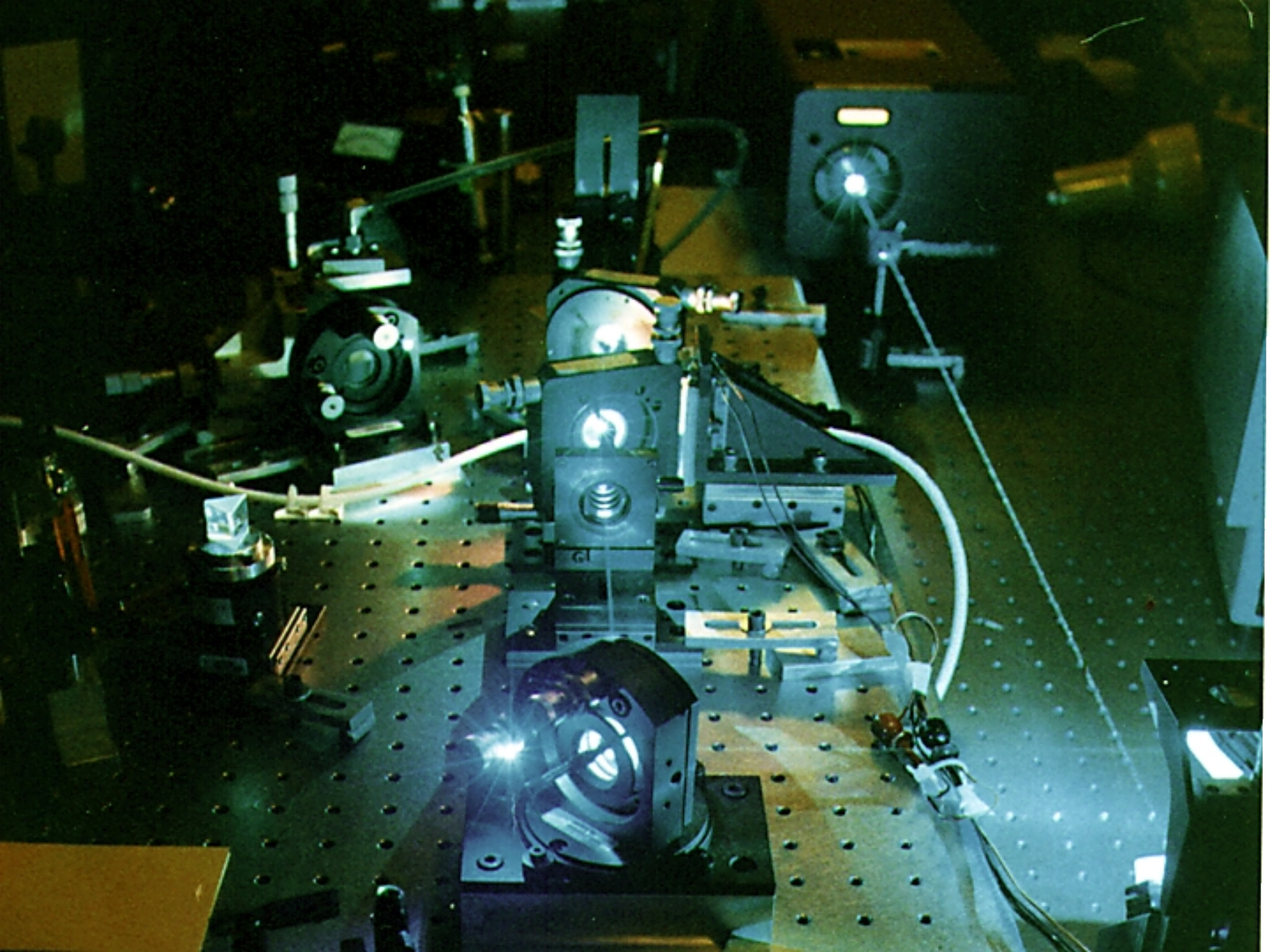
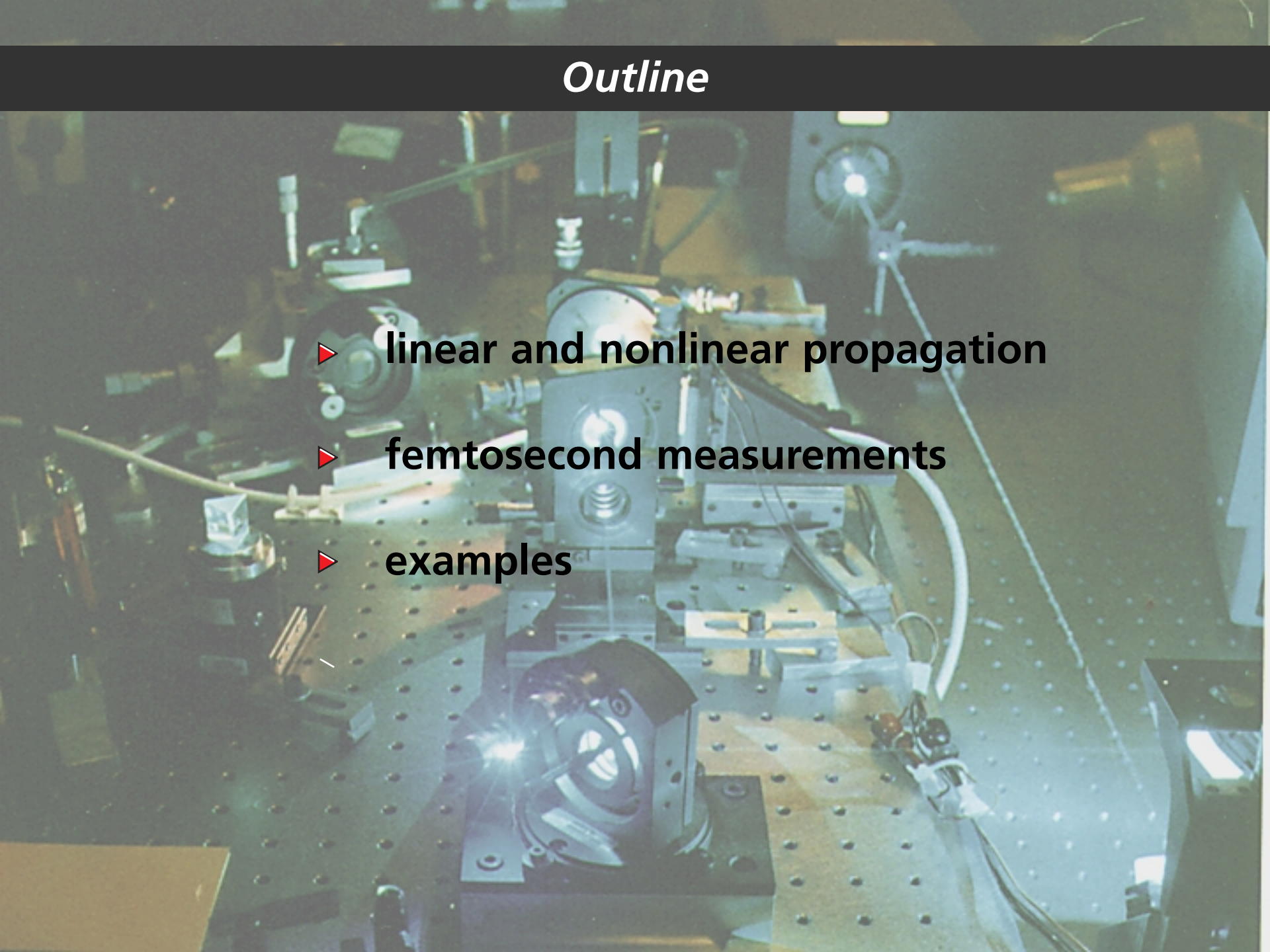
AN INTRODUCTION TO FEMTOSECOND LASER SCIENCE

Eric Mazur
Harvard University

Short Course SC541
Photonics West 2004, San Jose, CA



Outline



- linear and nonlinear propagation
- femtosecond measurements
- examples

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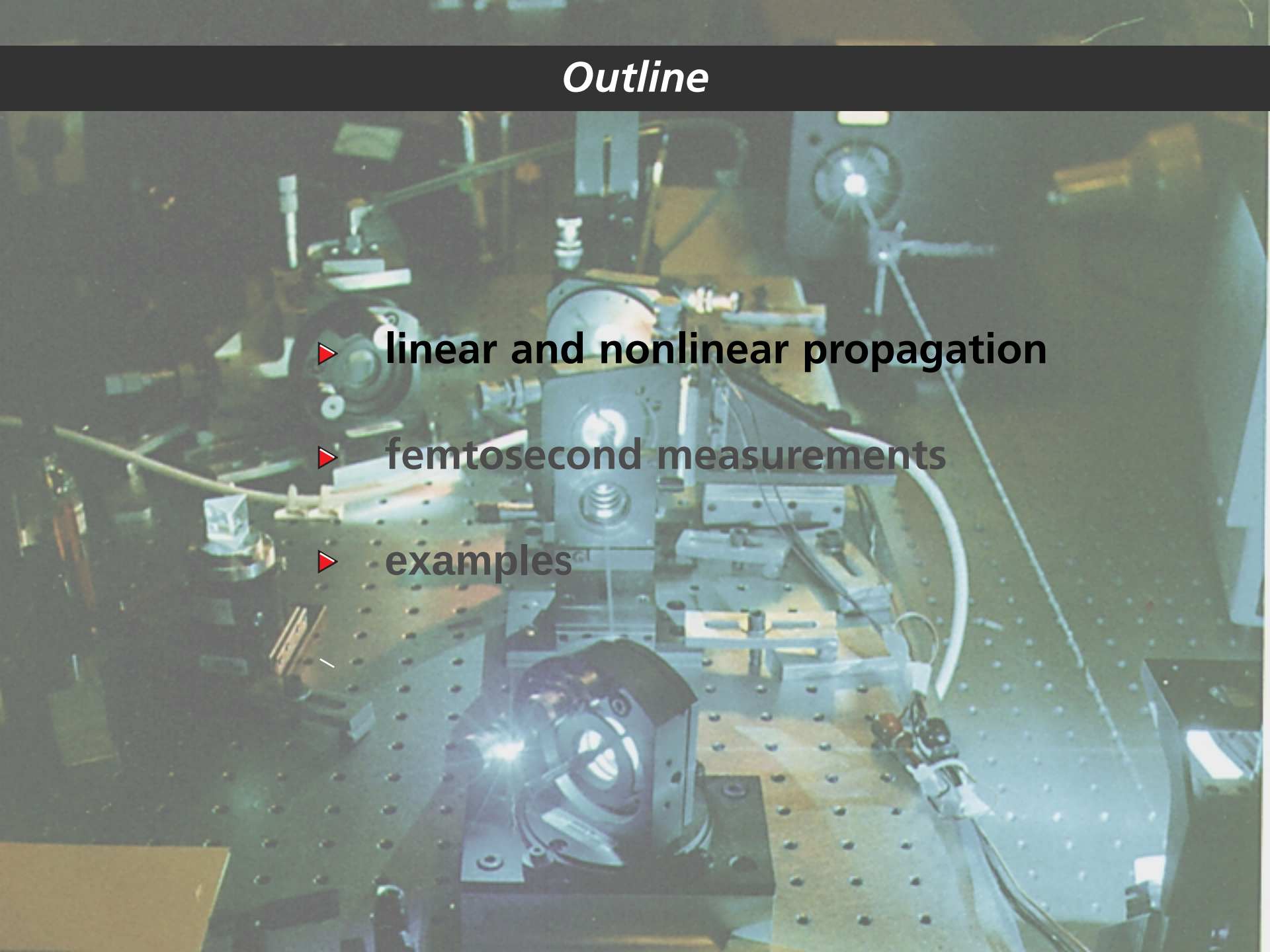

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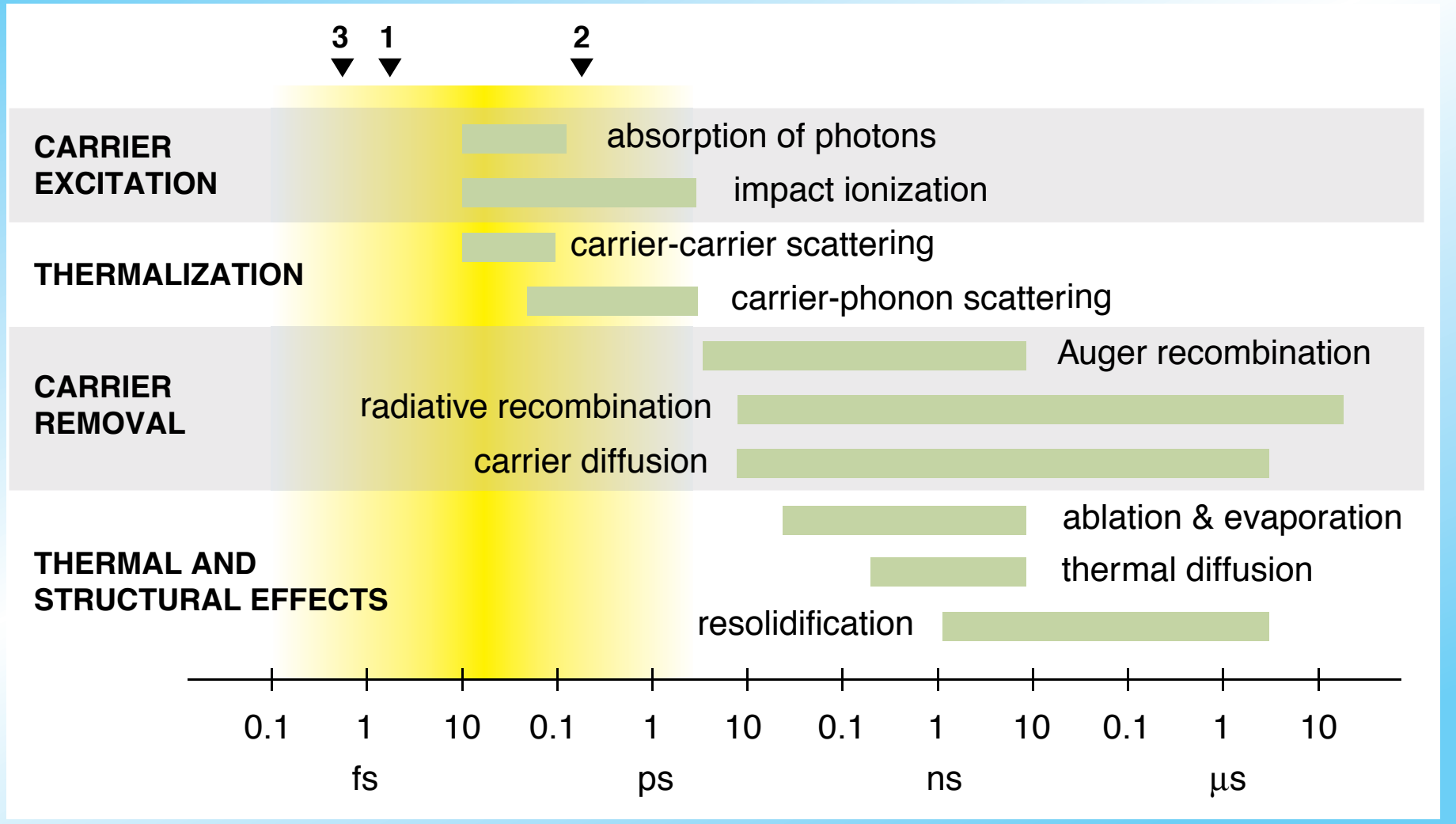

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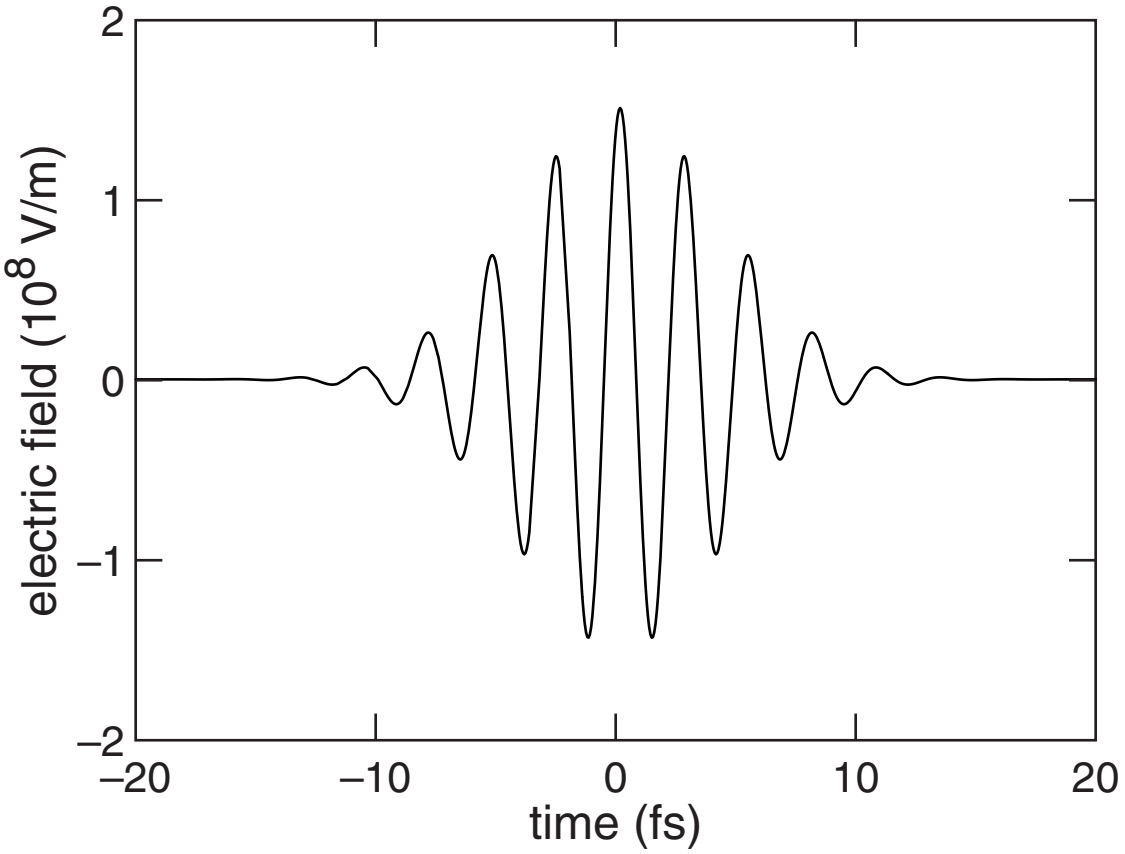

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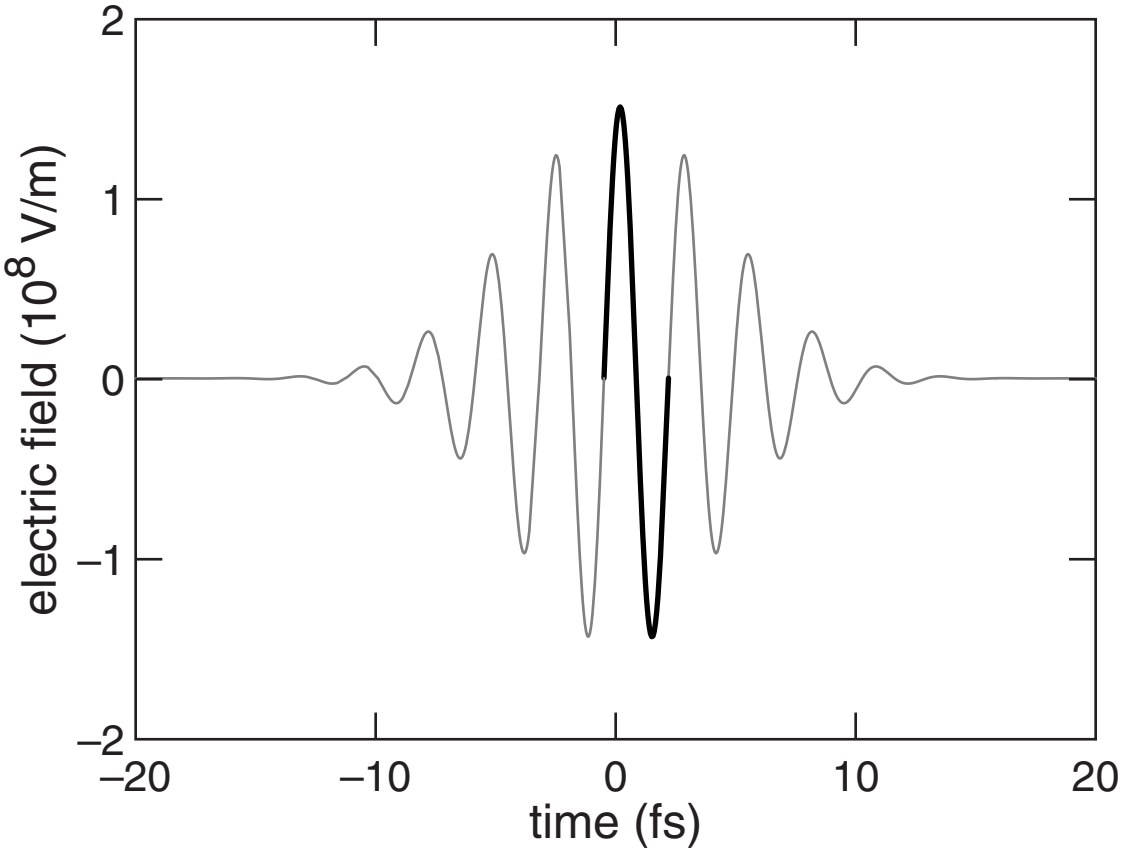

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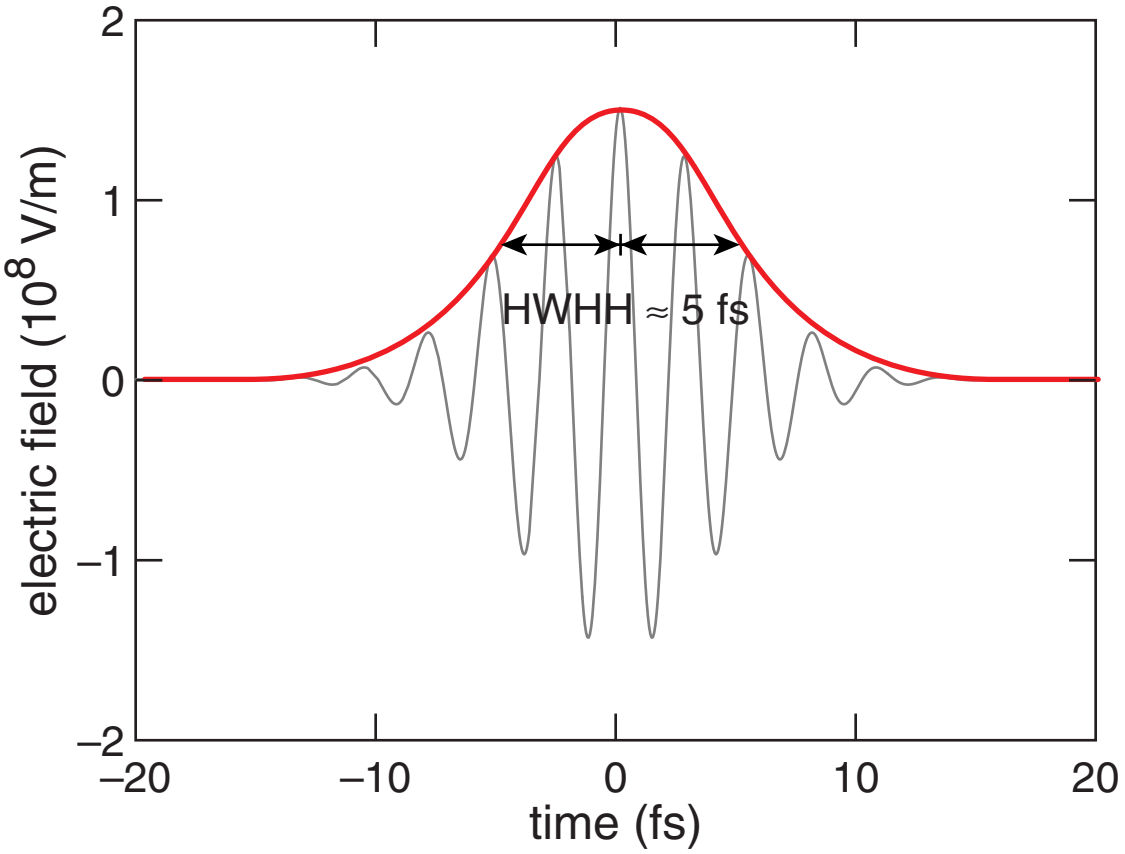

Introduction

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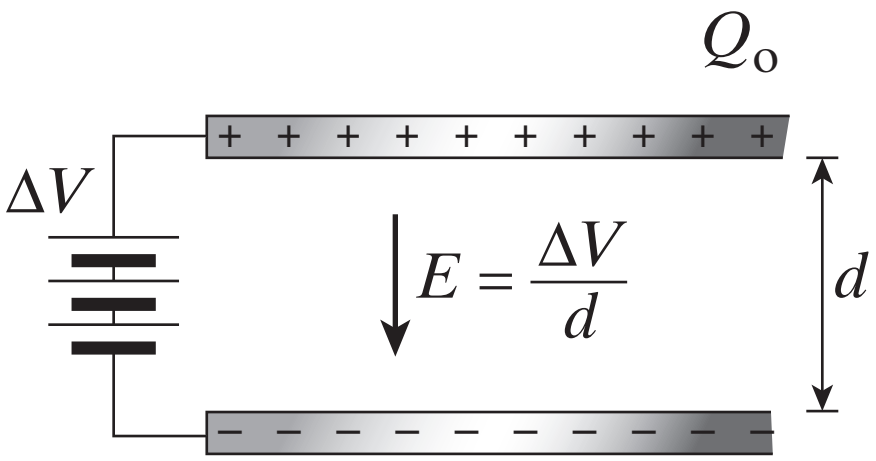
In non-ferromagnetic media $\mu \approx 1$**, and so** $n \approx \sqrt{\epsilon}$**.**

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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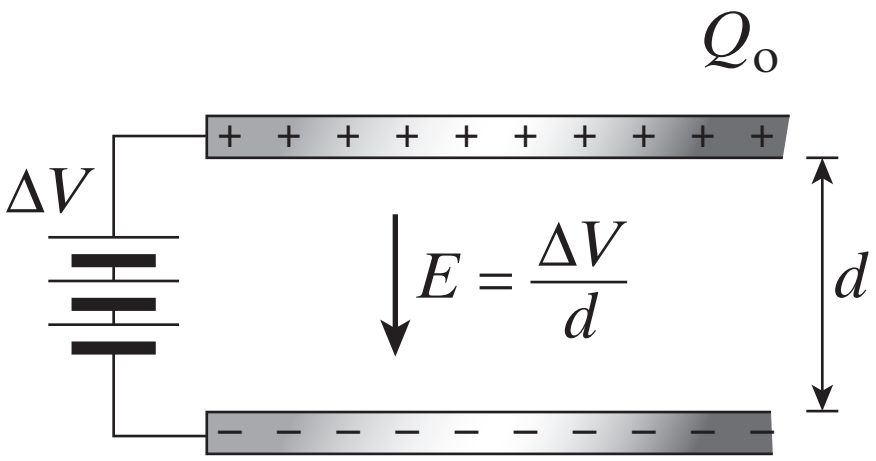

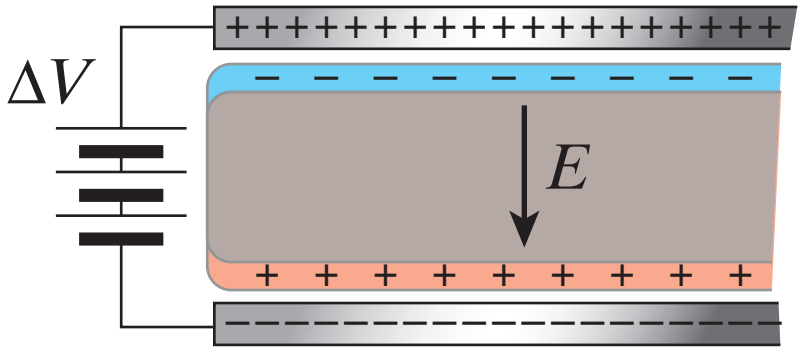

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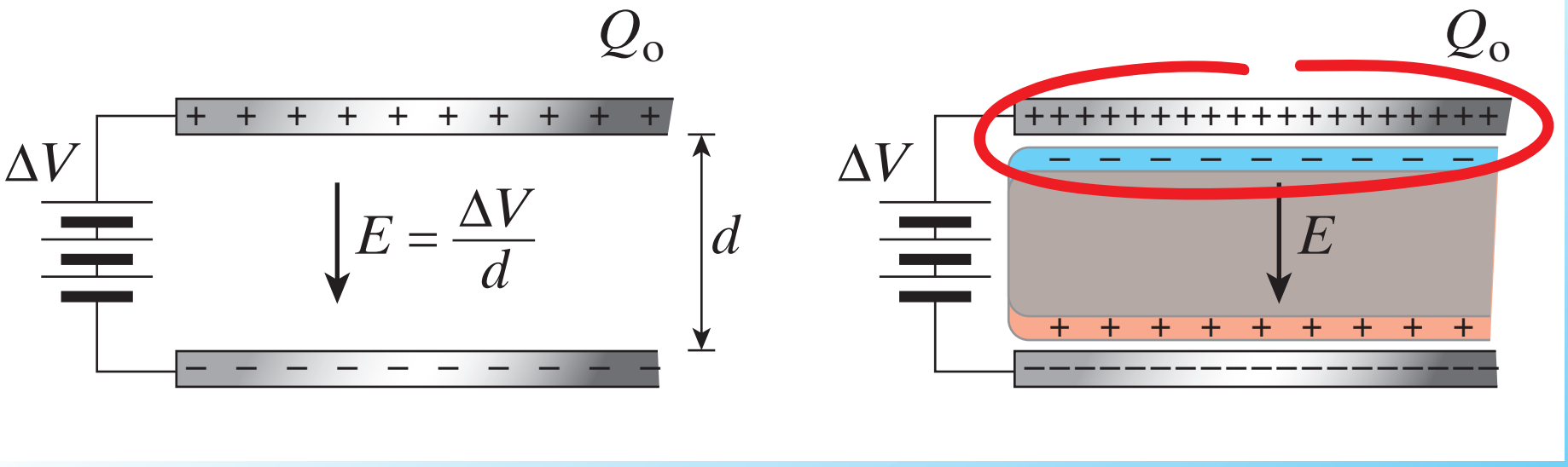

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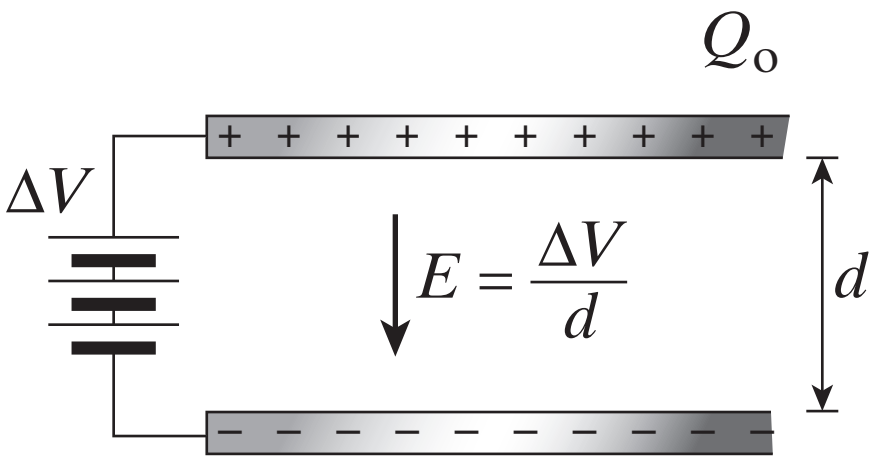
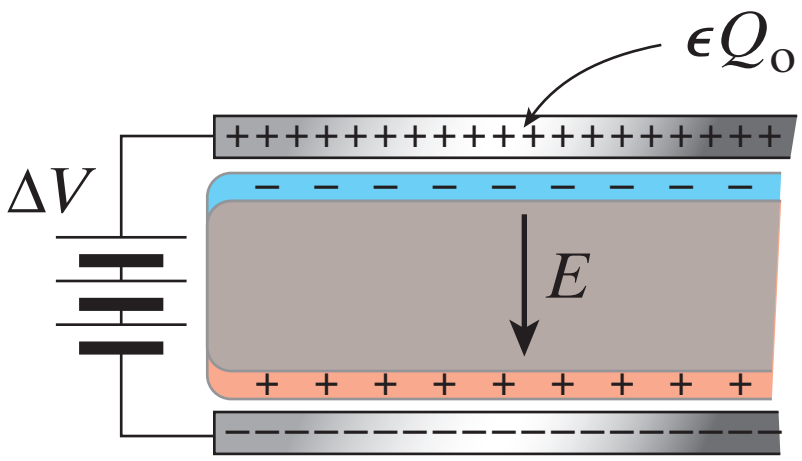
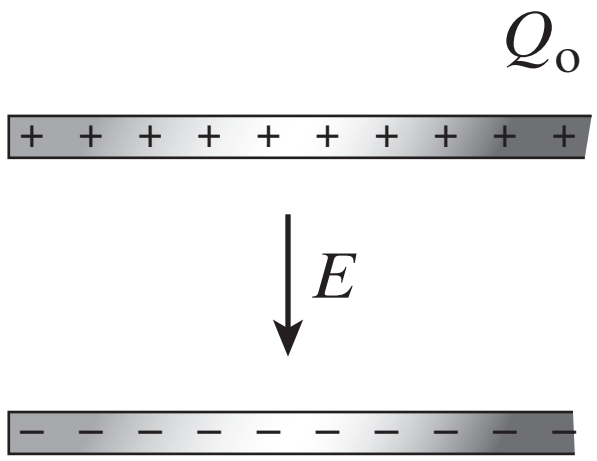




Propagation of EM waves through medium

Alternatively, ϵ is measure of the attenuation of the field



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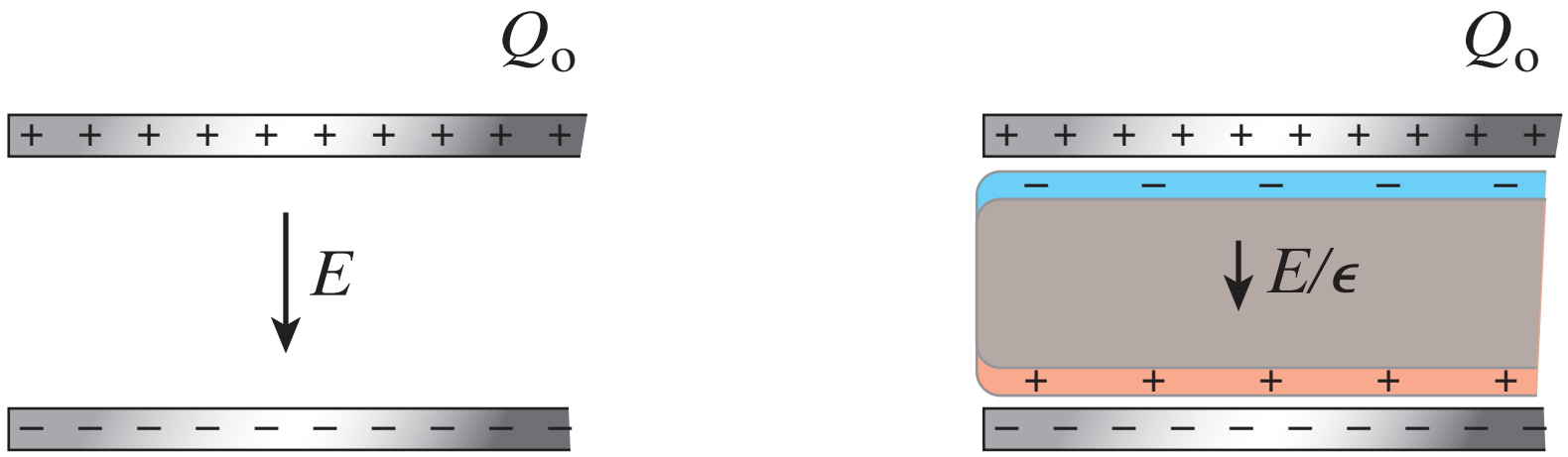
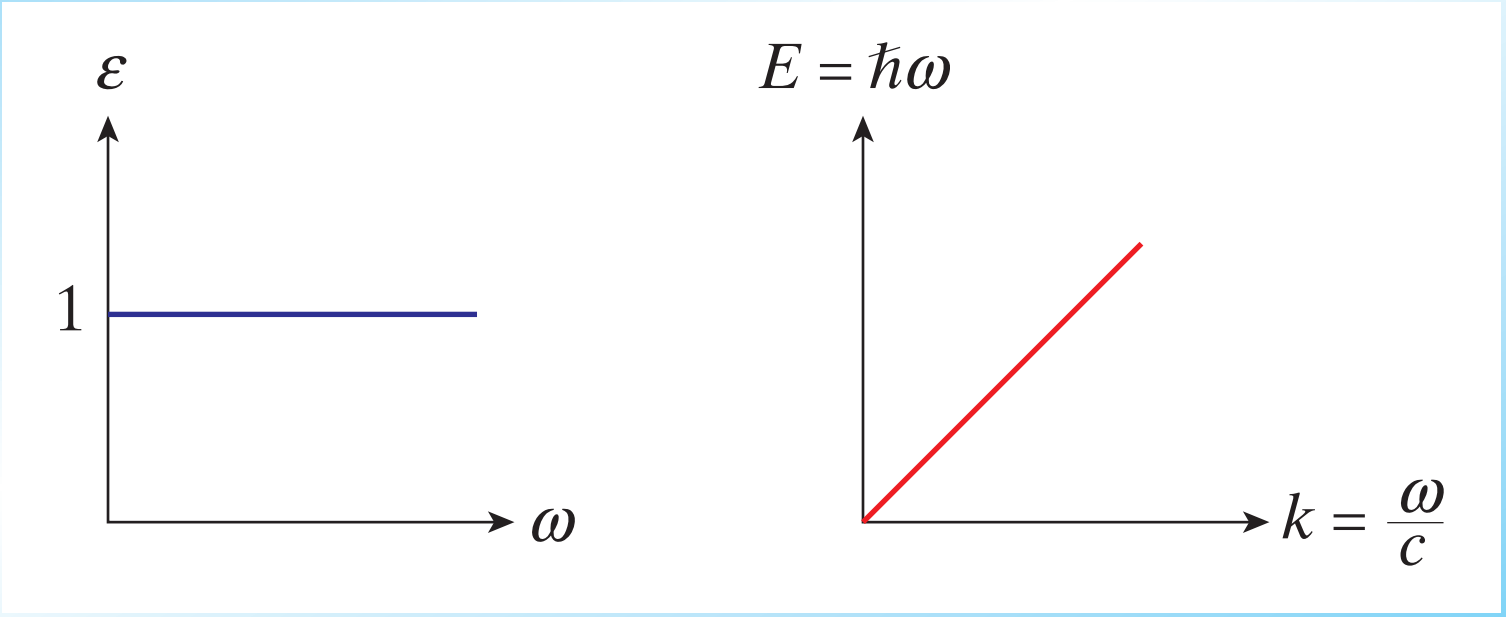


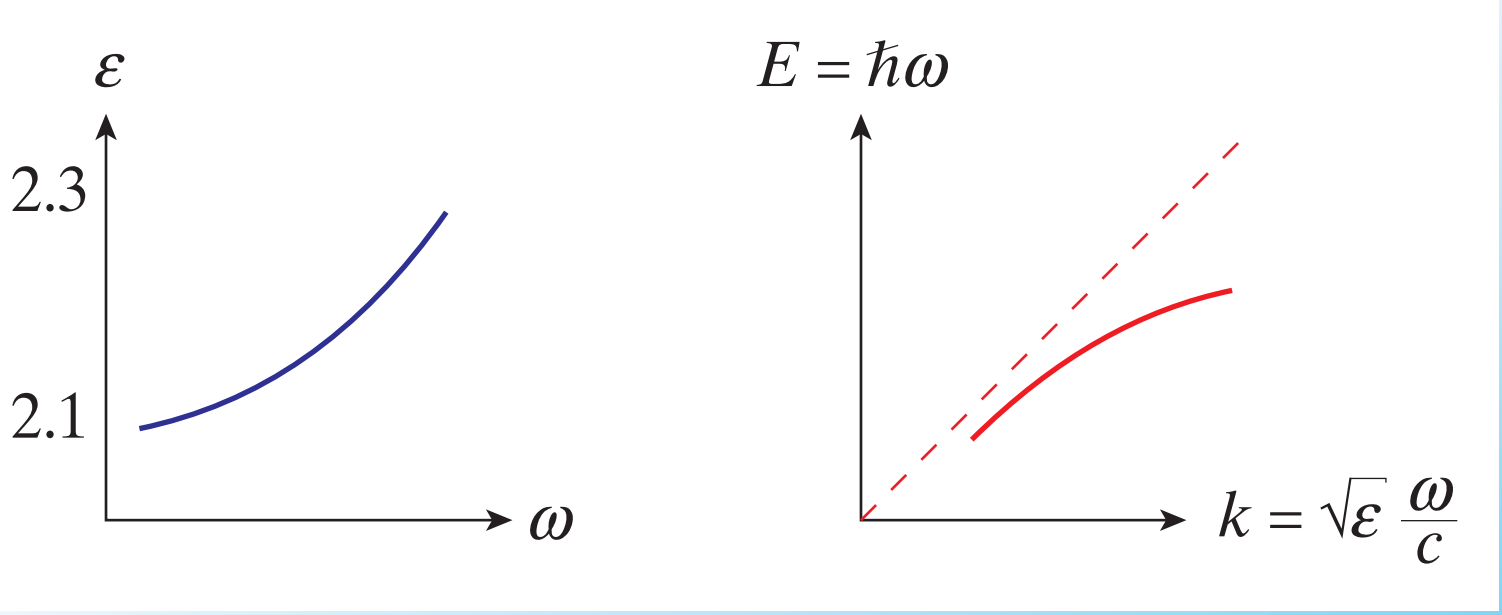
Propagation of EM waves through medium

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

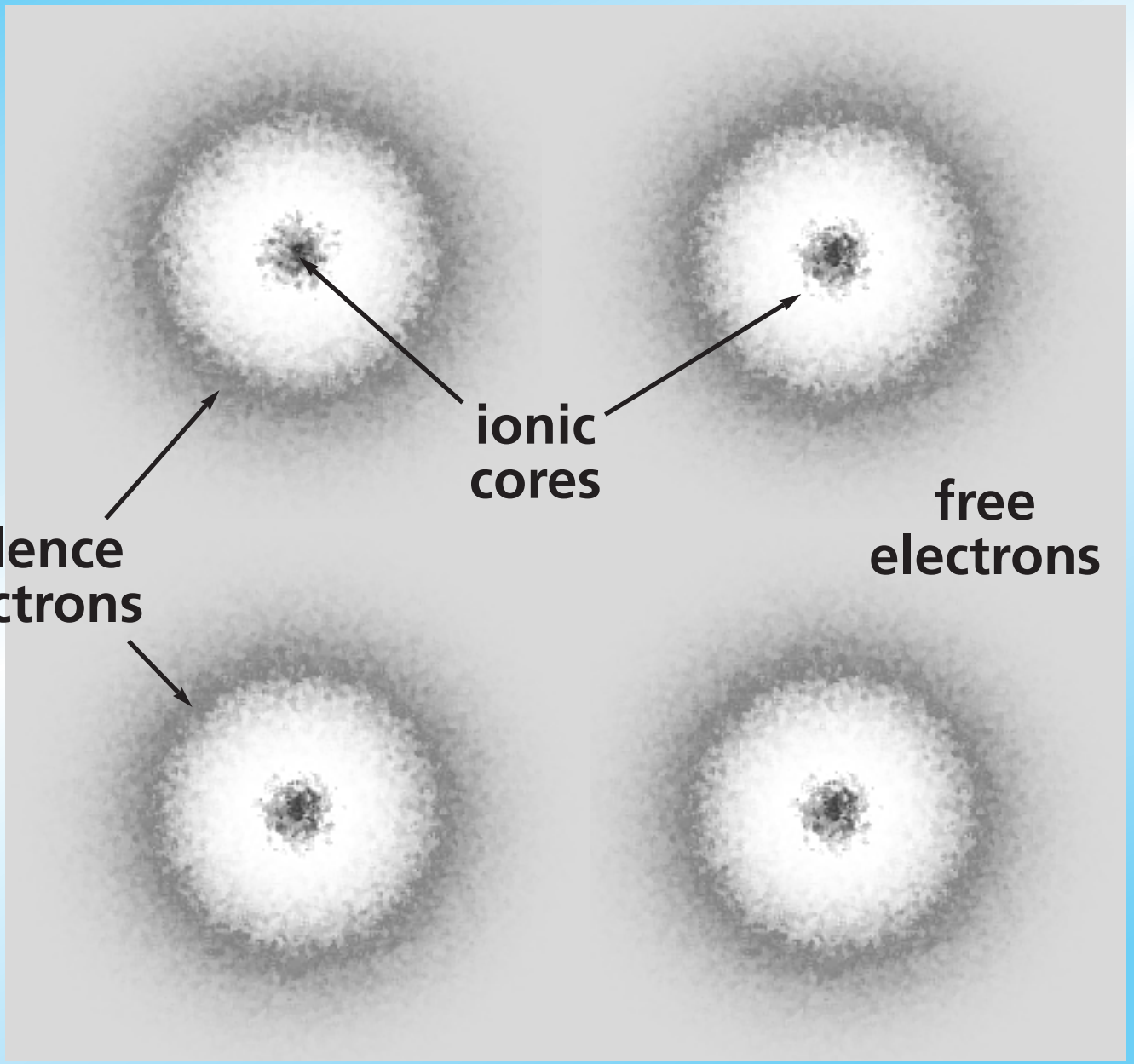


Propagation of EM waves through medium

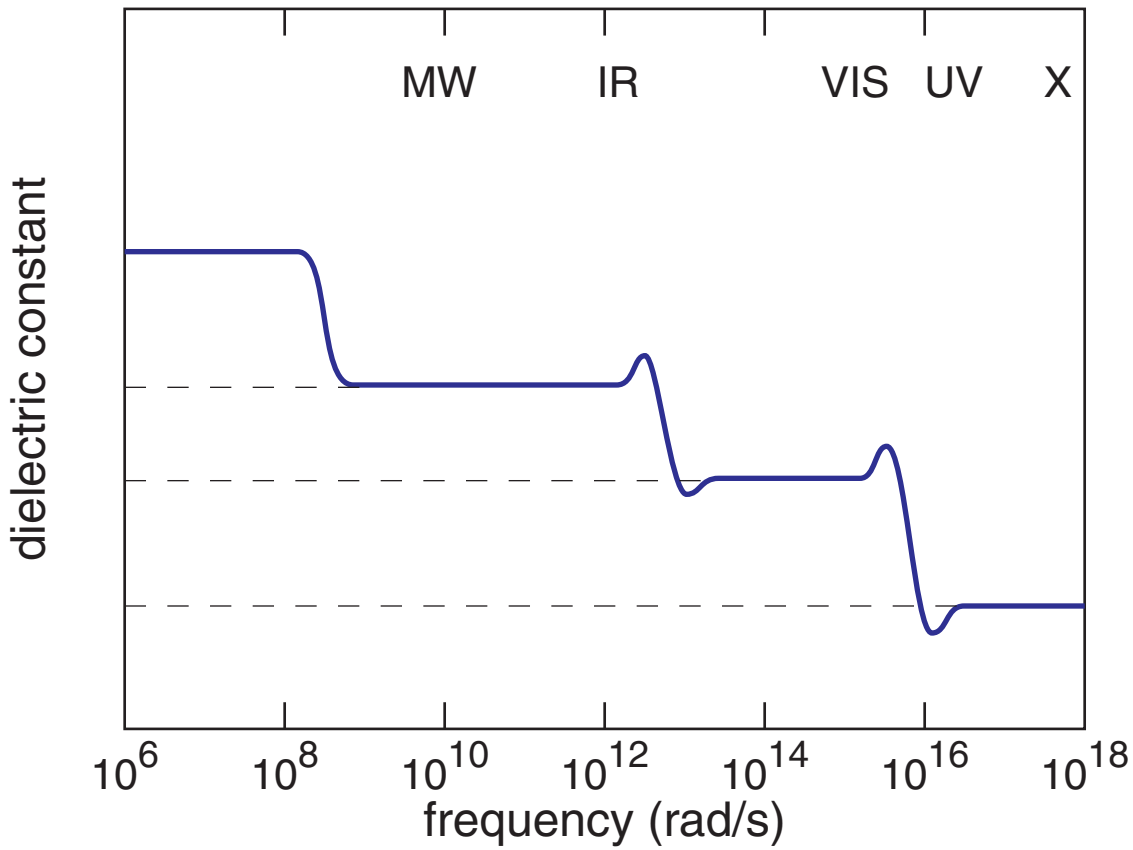
In medium: $v = \dfrac{c}{\sqrt{\epsilon(\omega)}} = \dfrac{c}{n} \quad \Rightarrow \quad \omega = \dfrac{c}{\sqrt{\epsilon}} k$



Which charges participate?



Dielectric function



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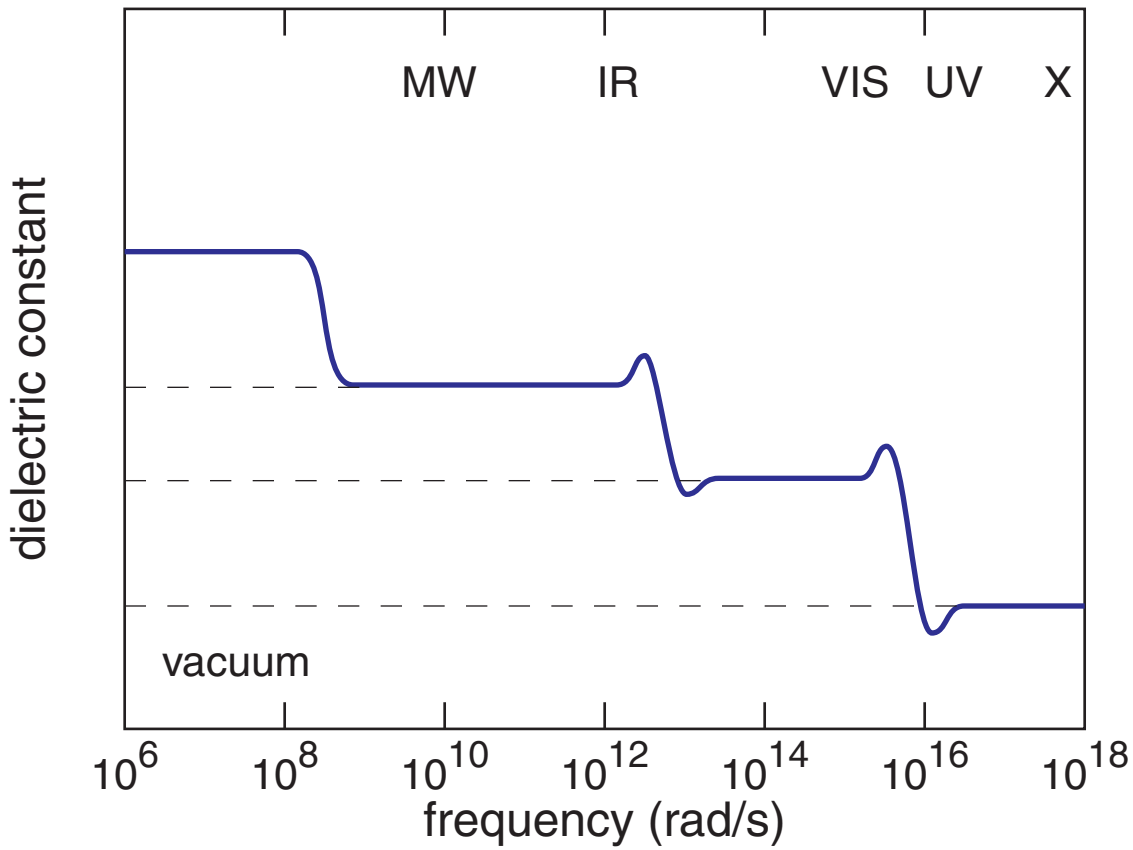

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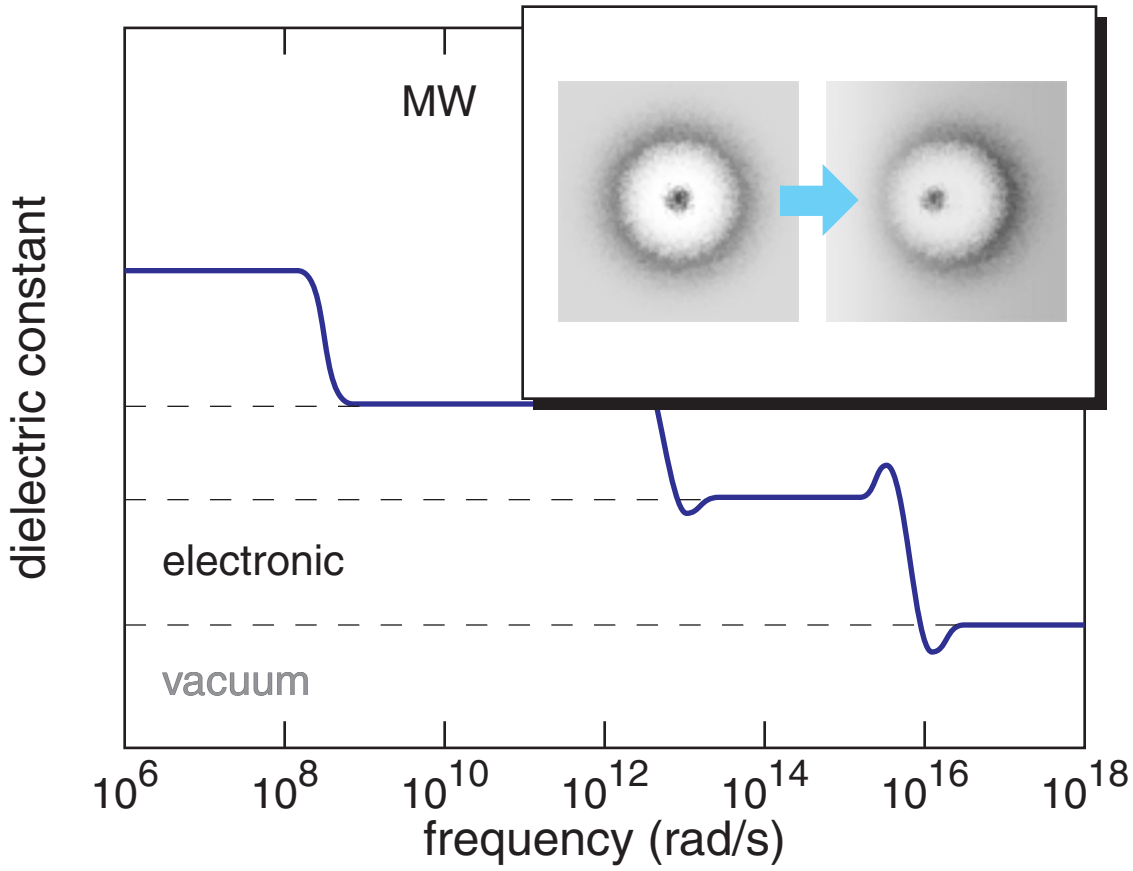

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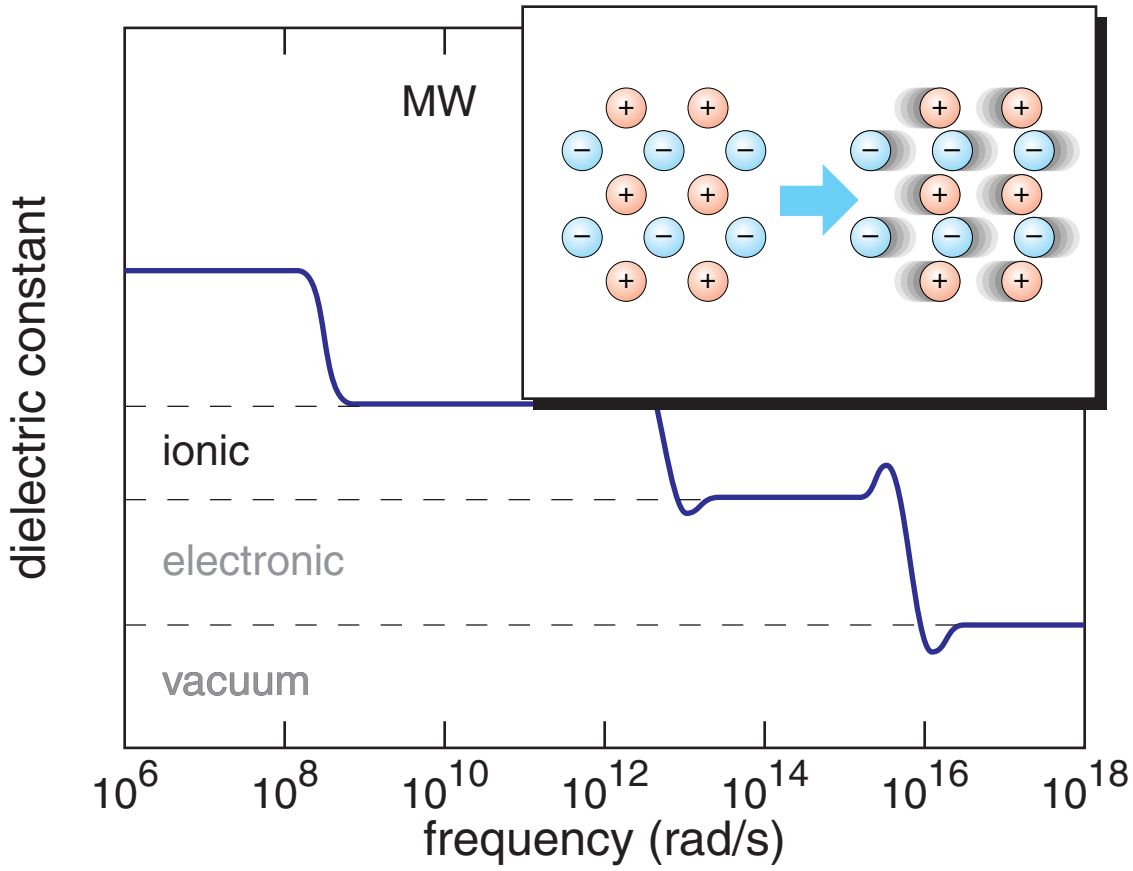

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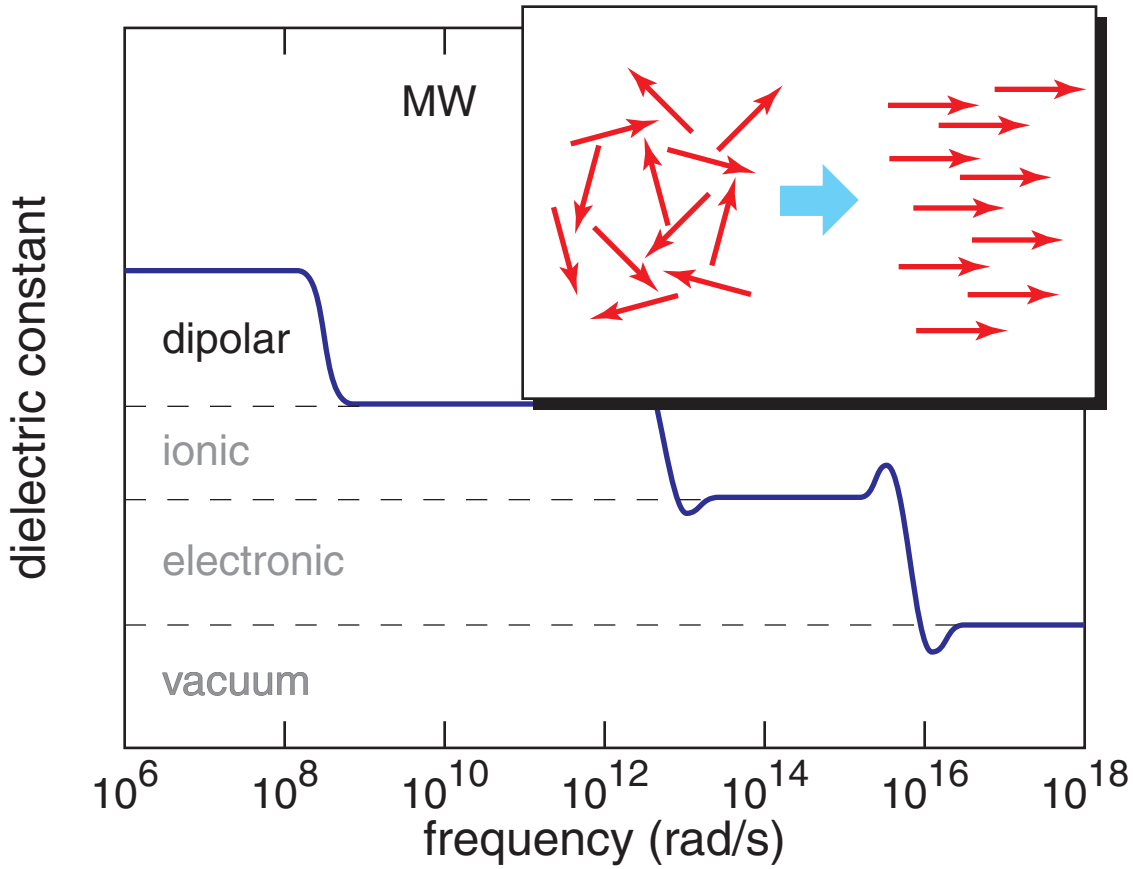

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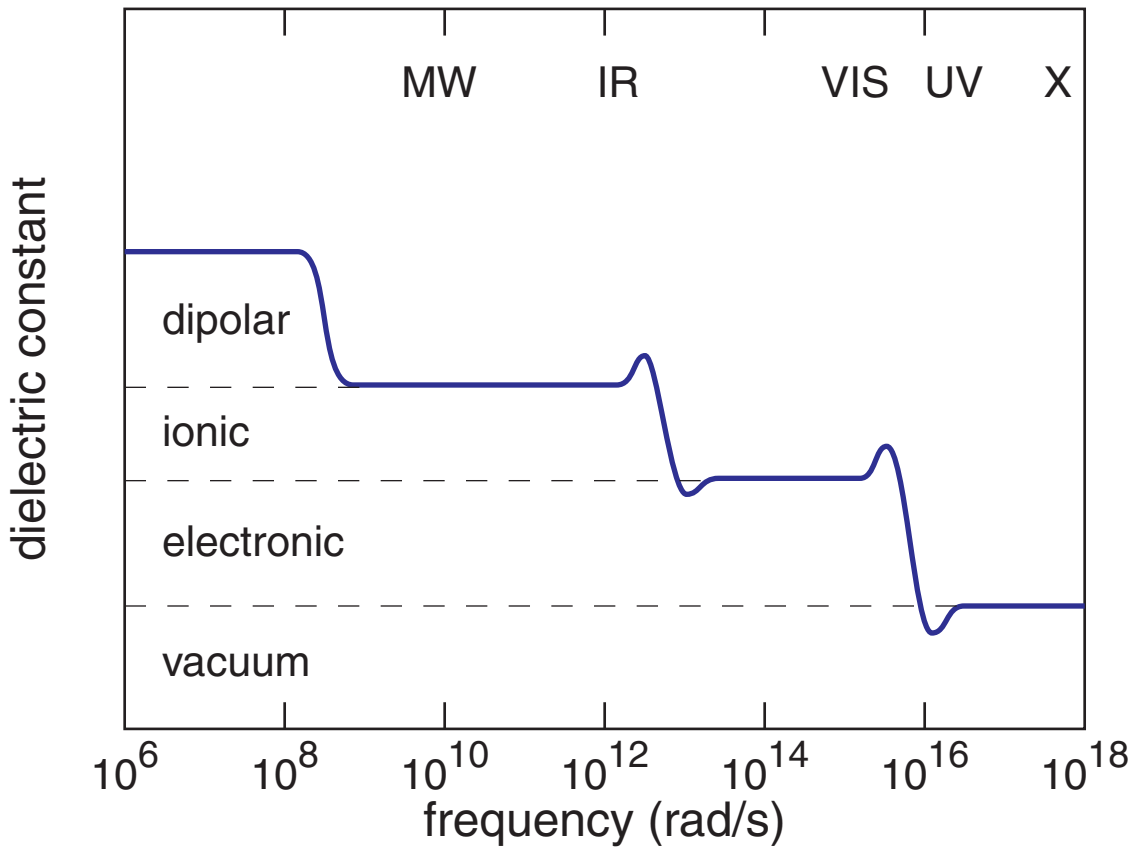

Bound electrons

Electron on a string:

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

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

Bound electrons

Steady state: electron oscillates at driving frequency

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

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Polarization

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

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Dielectric function

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

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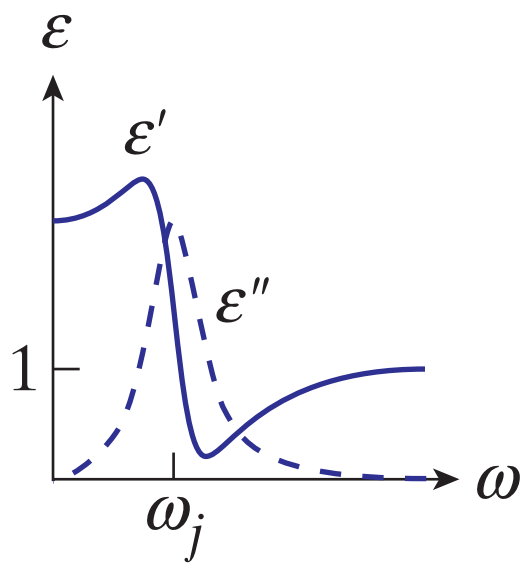
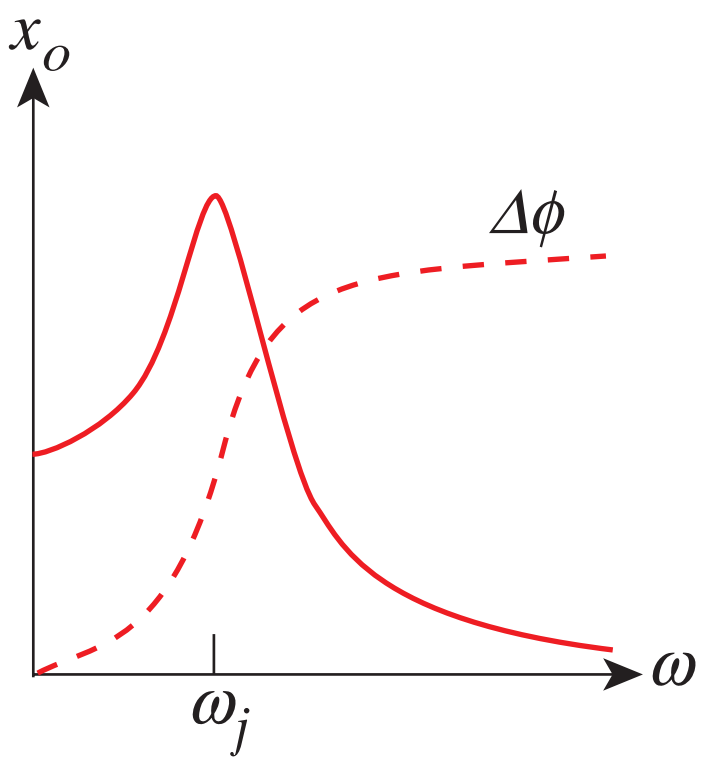
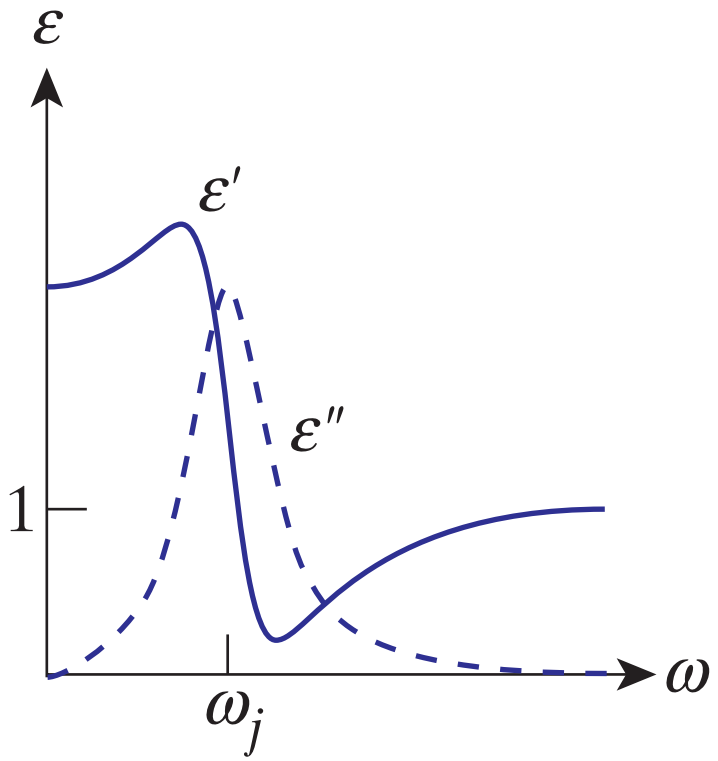


Bound electrons

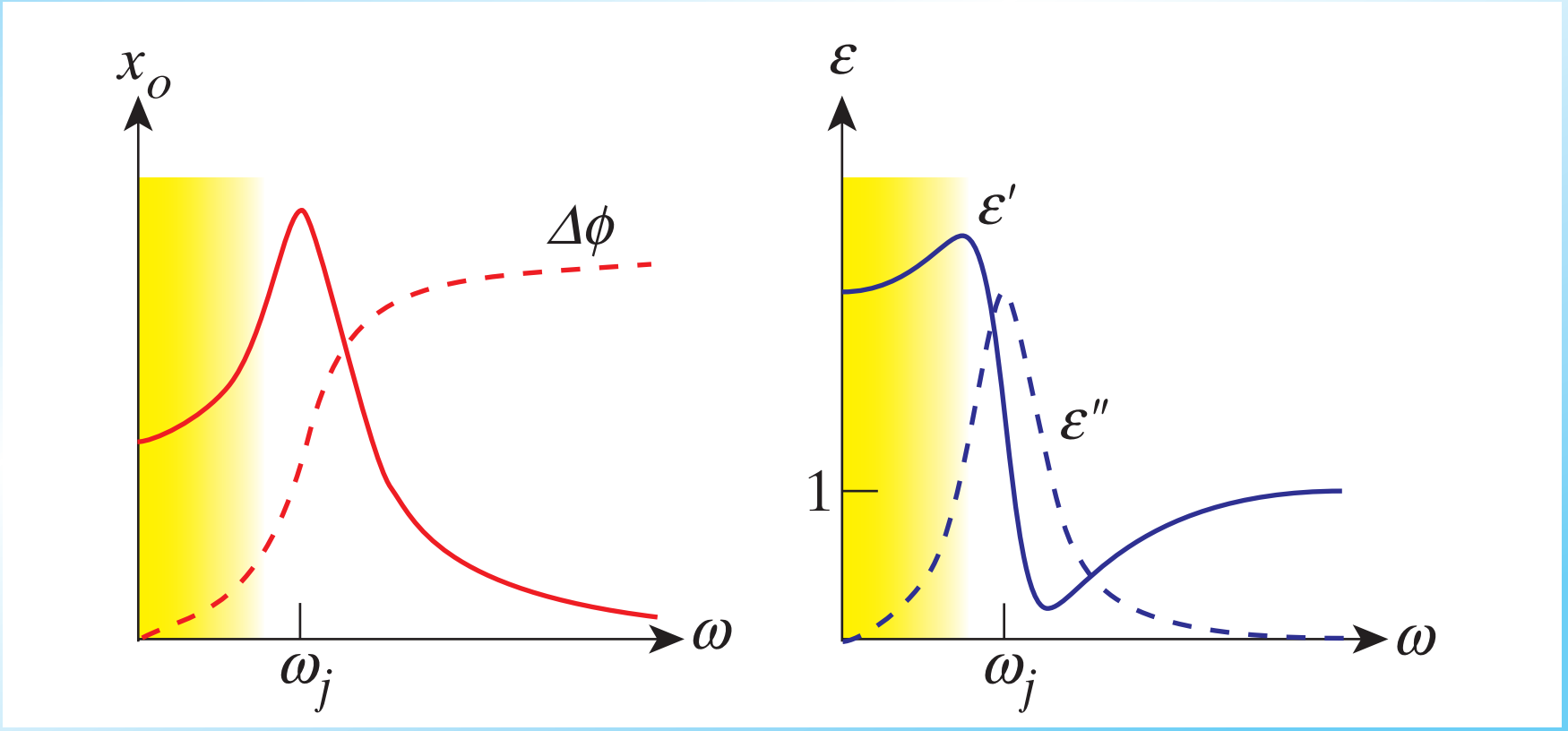
amplitude of bound charge oscillation





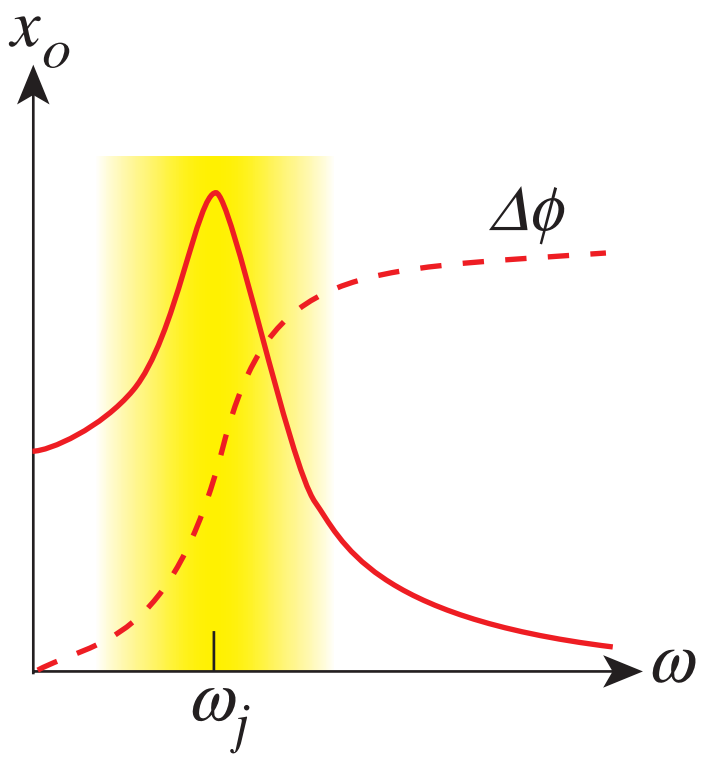
Bound electrons

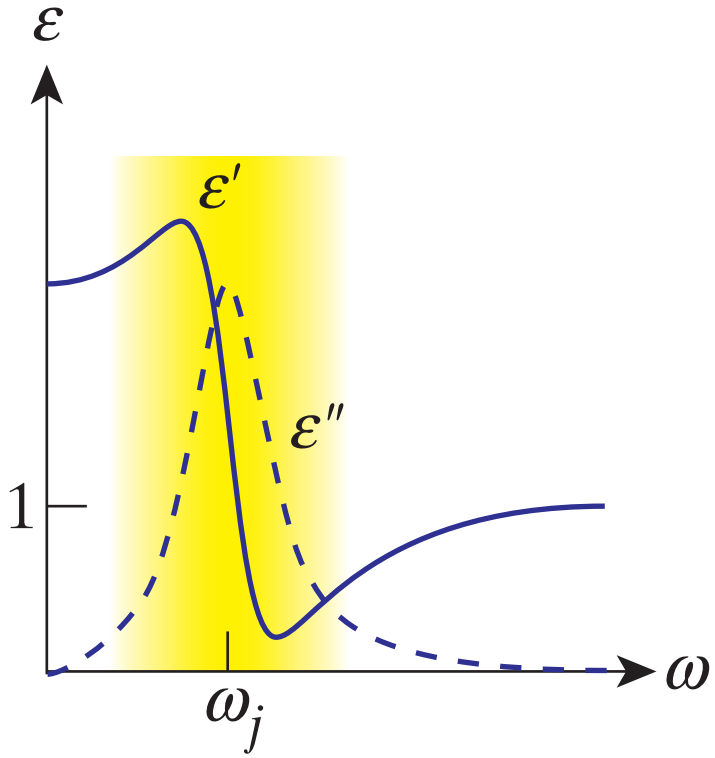
Below resonance: bound charges keep up with driving field ⇒ field attenuated, wave propagates more slowly



Bound electrons

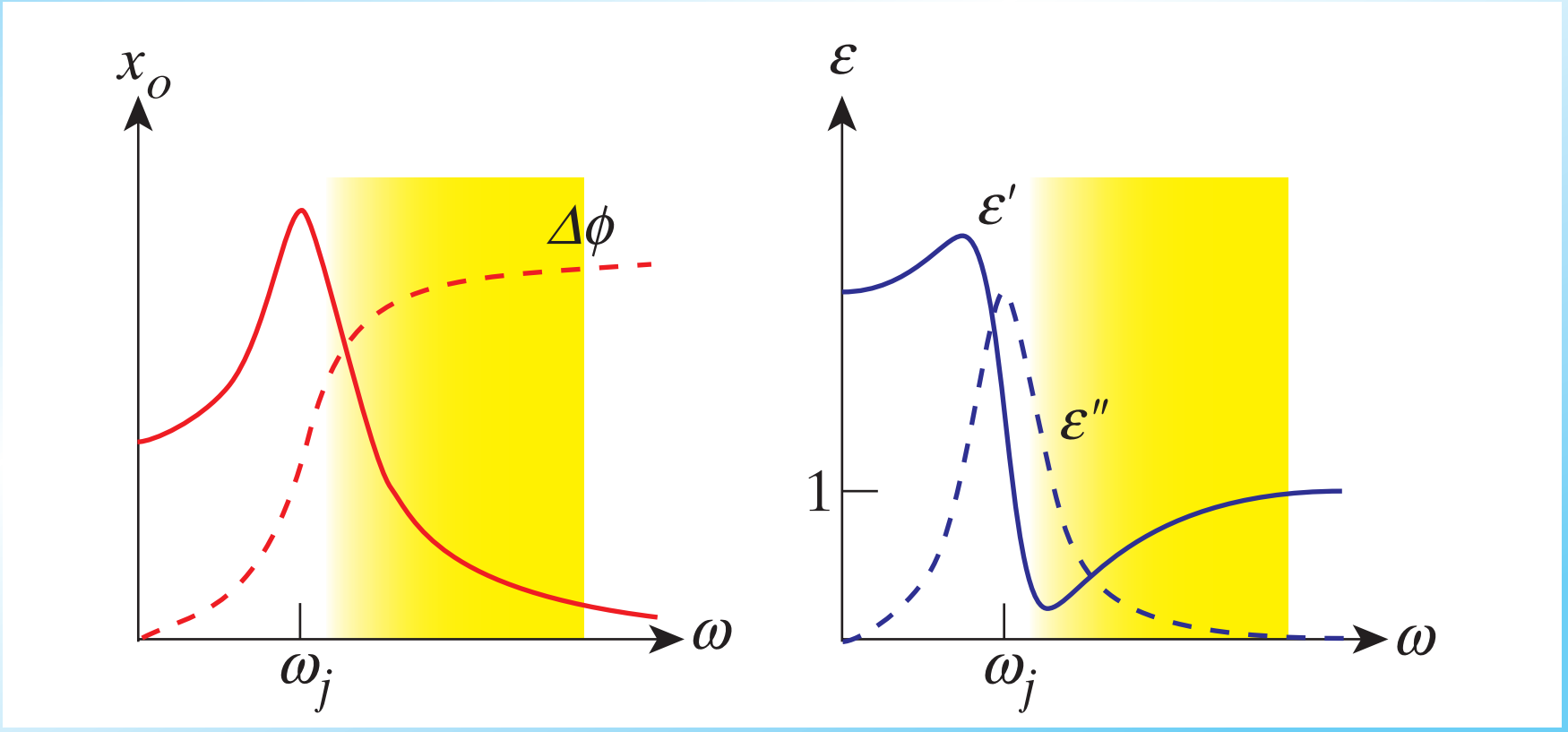
At resonance: energy transfer from wave to bound charges ⇒ wave attenuates (absorption)





Bound electrons

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



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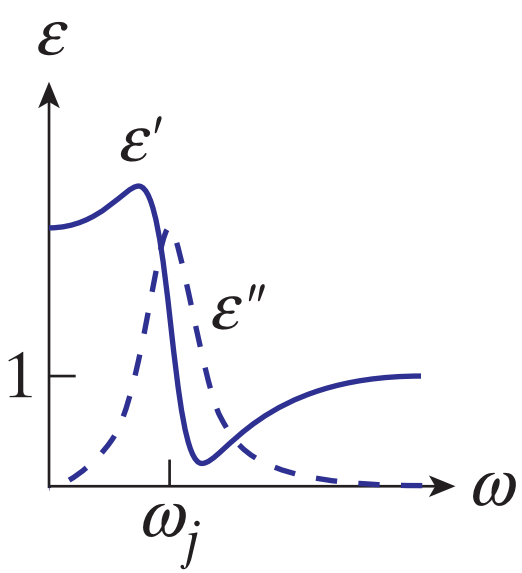

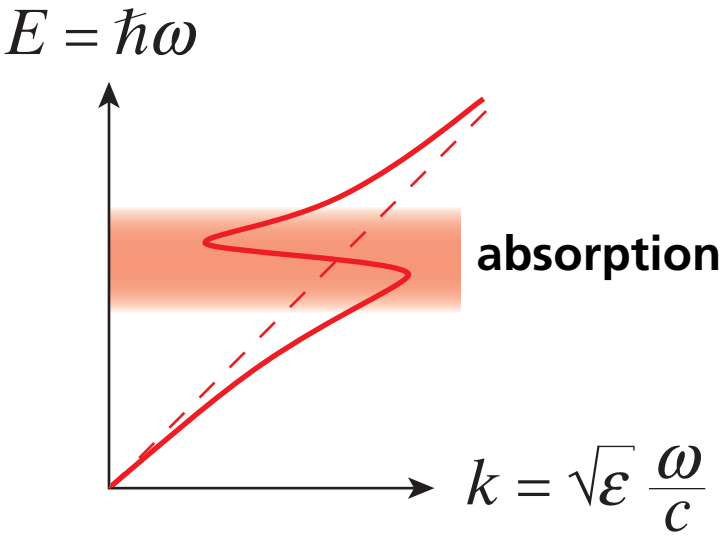

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Low frequency ($\omega \ll 1$) $\Rightarrow$ current generated

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

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

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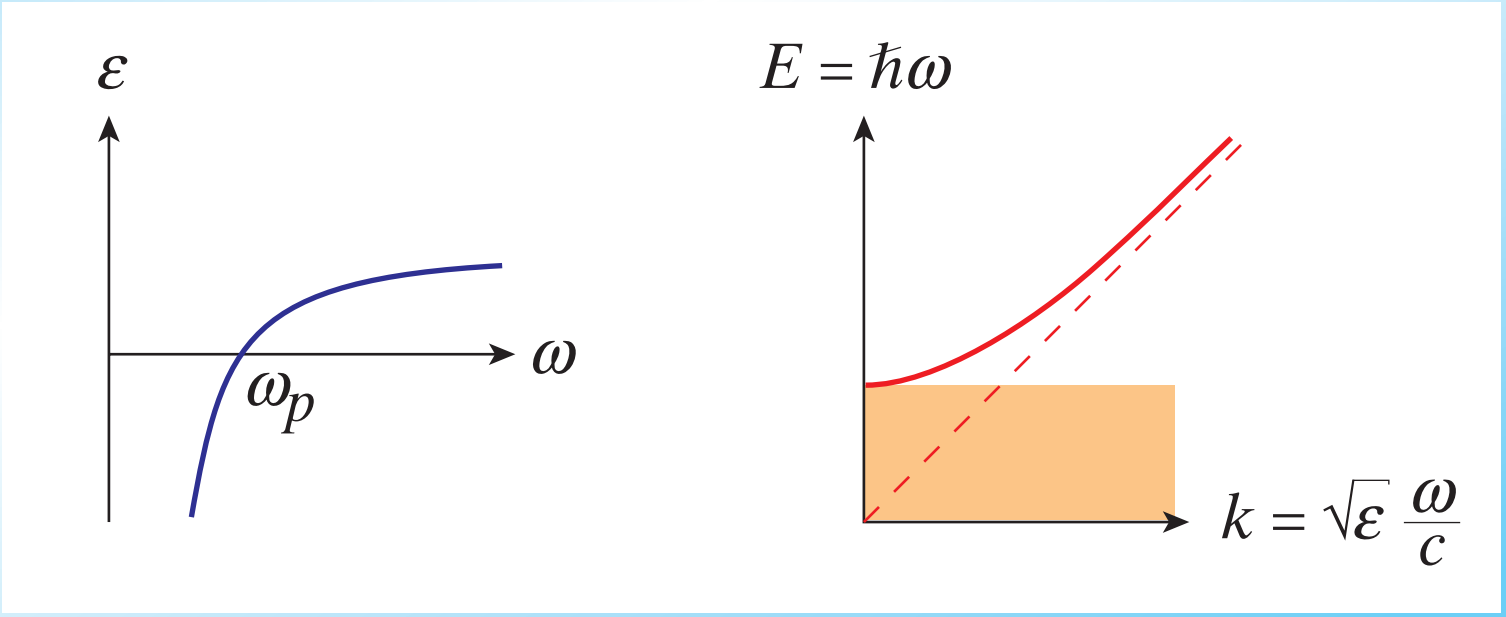
Dielectric function

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

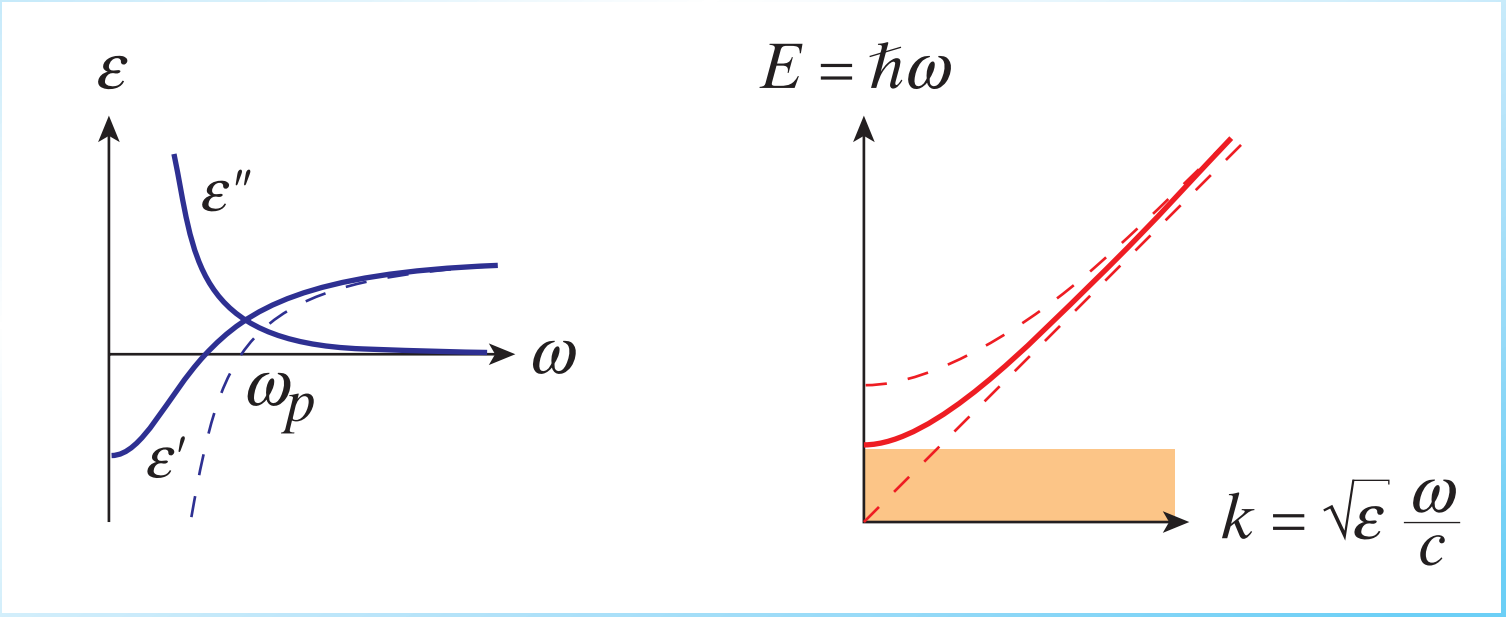
Plasma

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

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

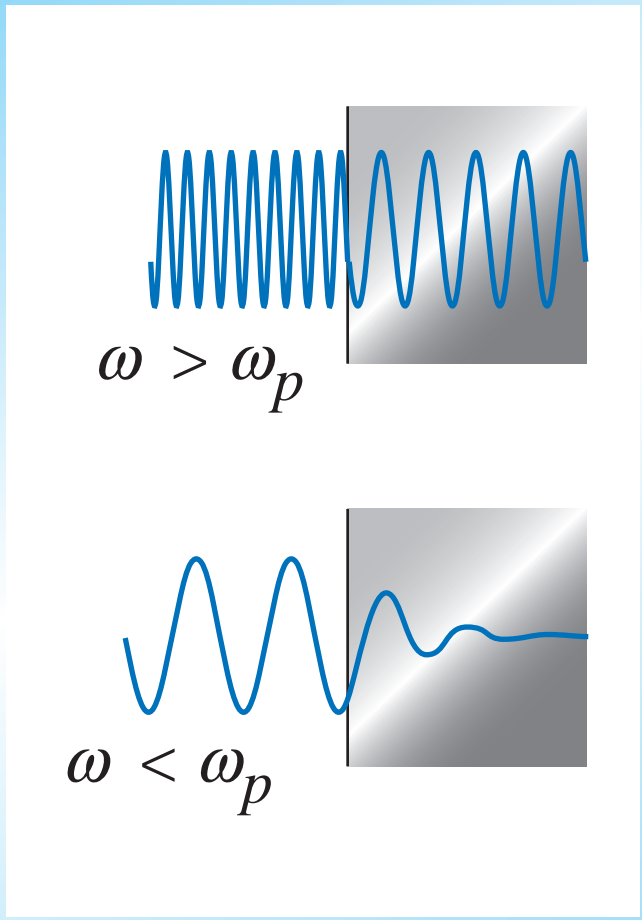


Add damping $\gamma \lesssim \omega_p$



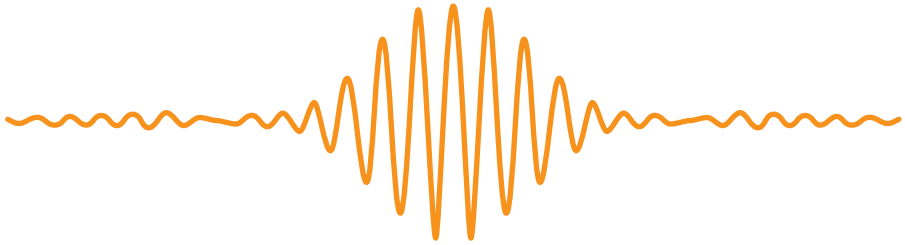
Plasma

Plasma acts like a high-pass filter:

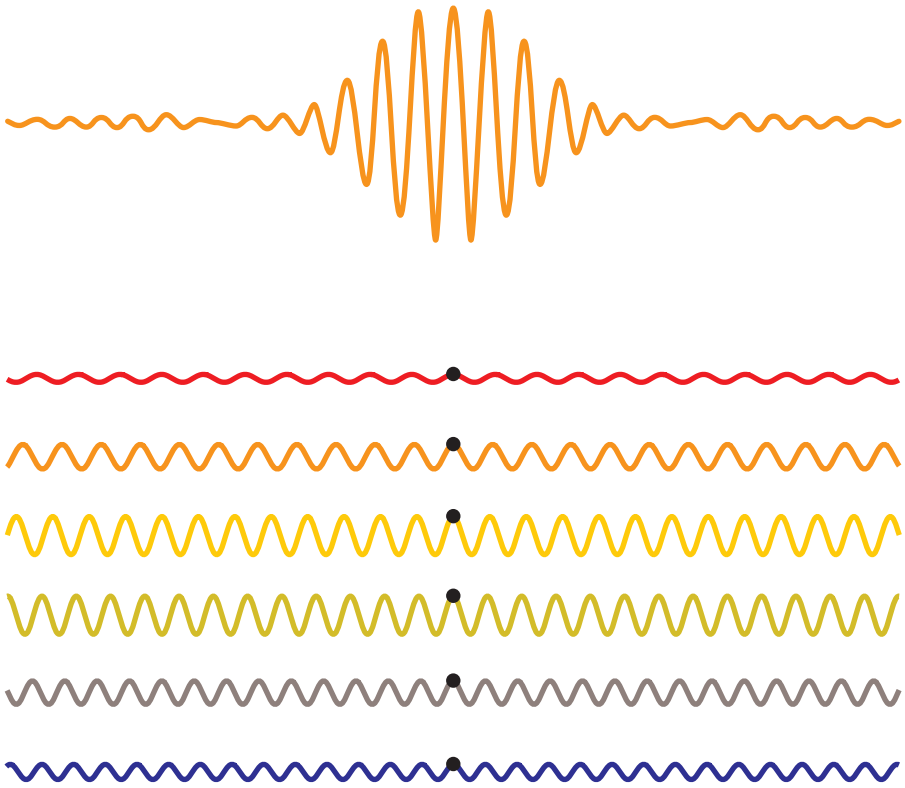


log N (cm^{-3})	ω_p (rad s^{-1})	λ_p
22	6×10^{15}	330 nm
18	6×10^{13}	33 μm
14	6×10^{11}	3.3 mm
10	6×10^{9}	0.33 m

Pulse dispersion



Pulse dispersion



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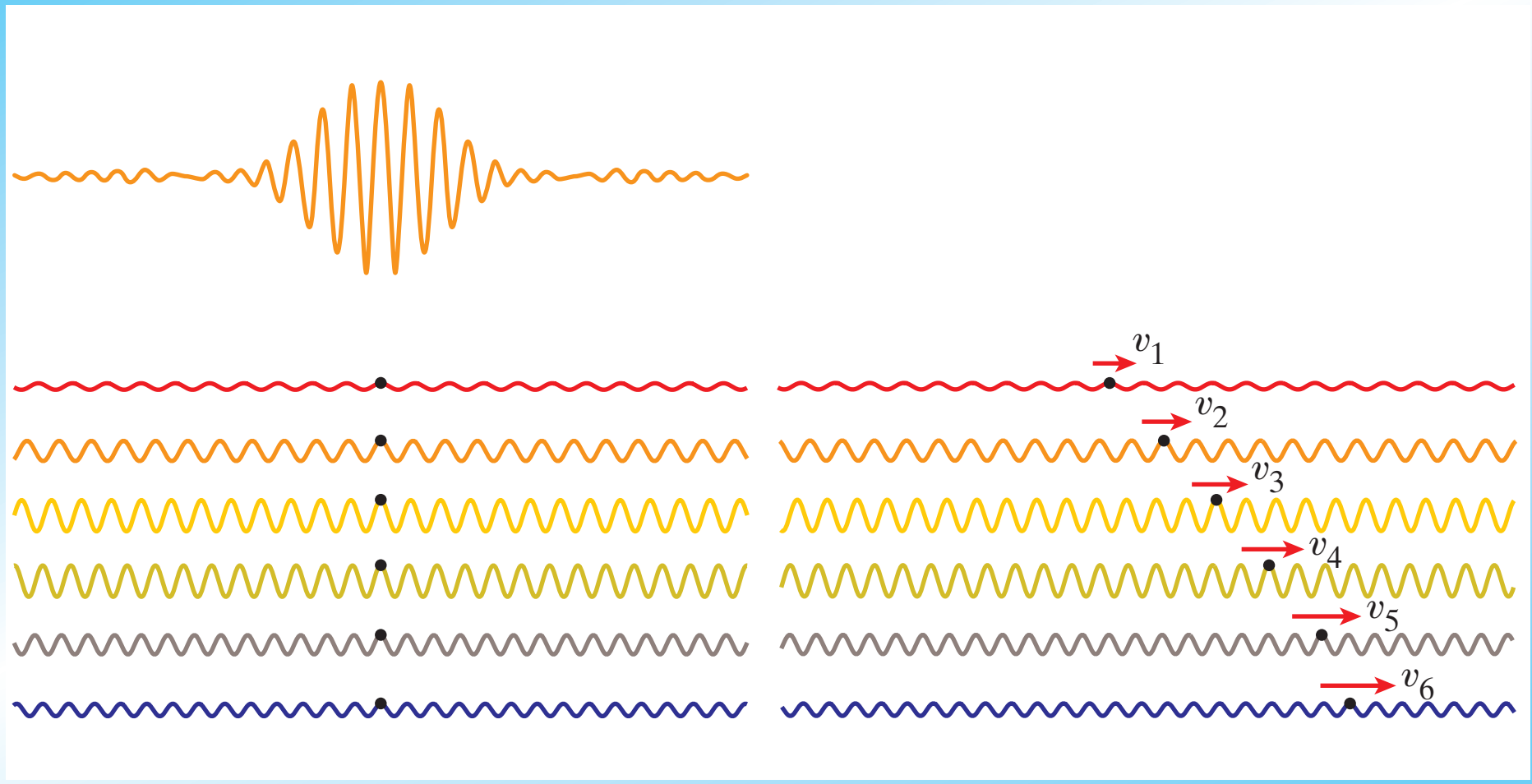

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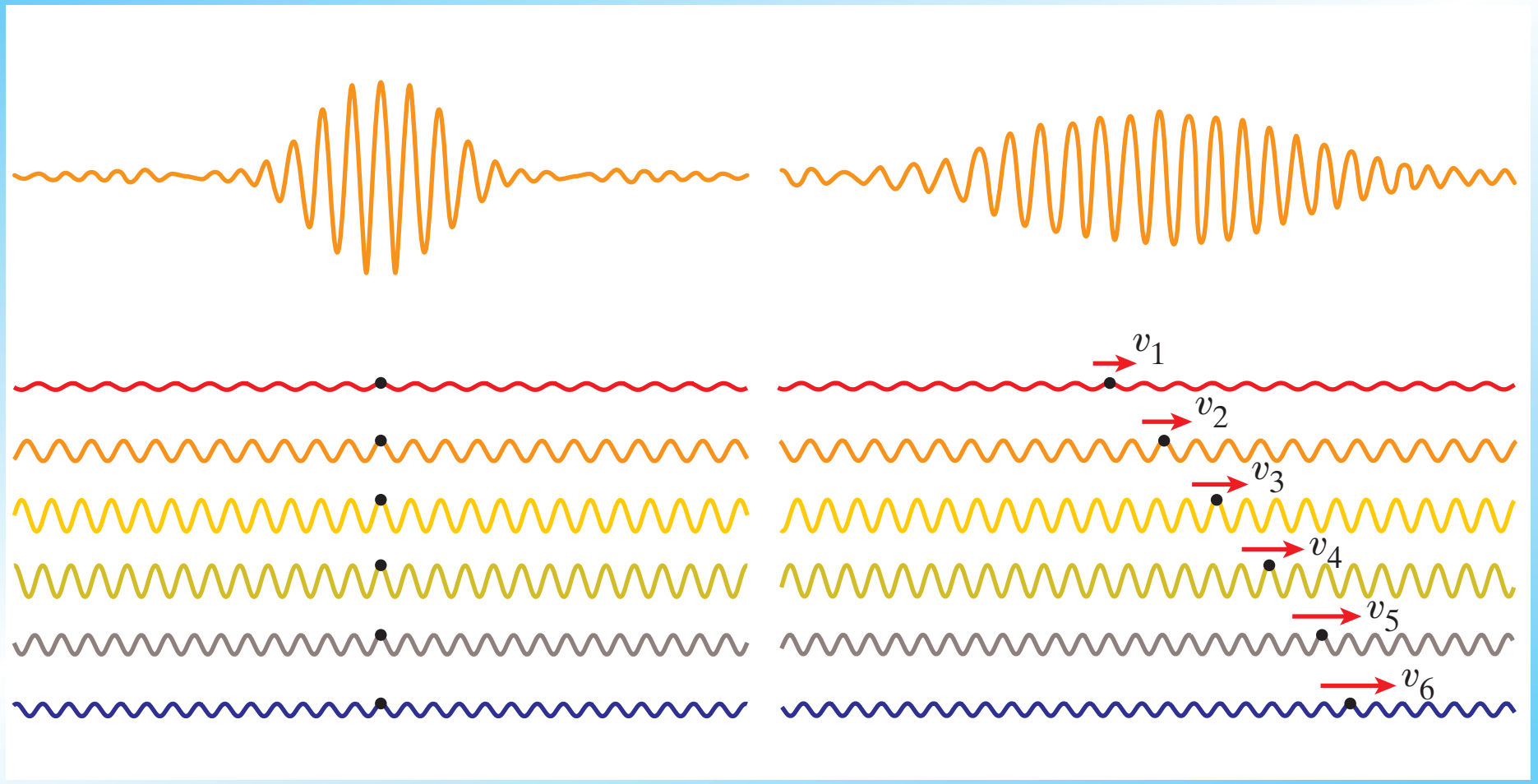

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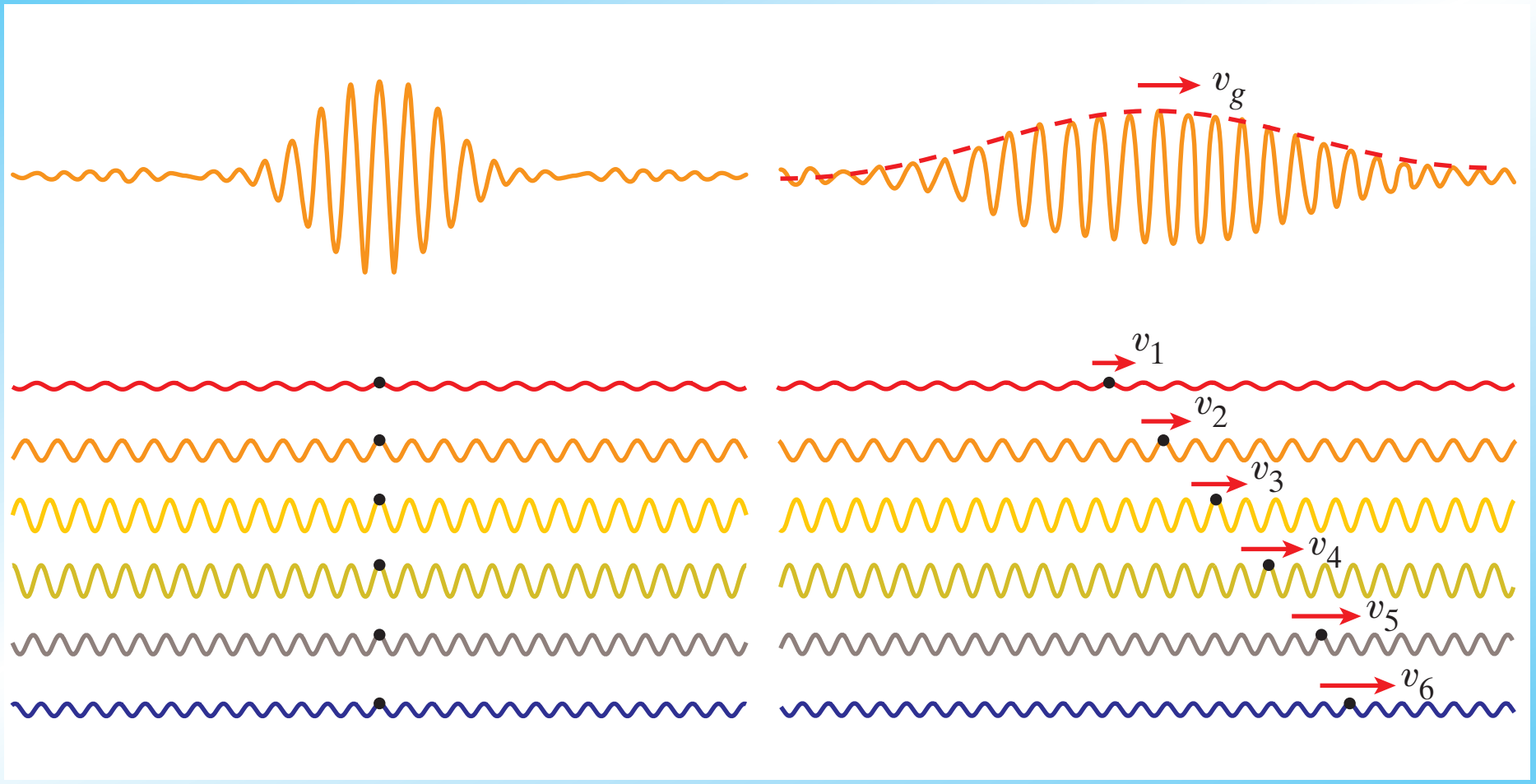
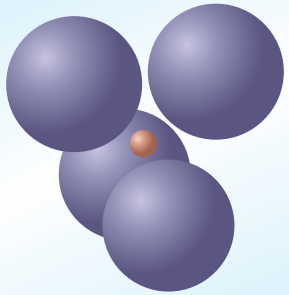


Nonlinear optics

Linear response $P(t) = \epsilon_o \chi_e E(t)$



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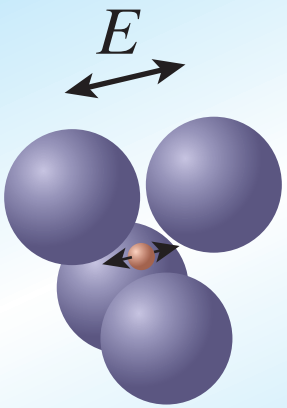

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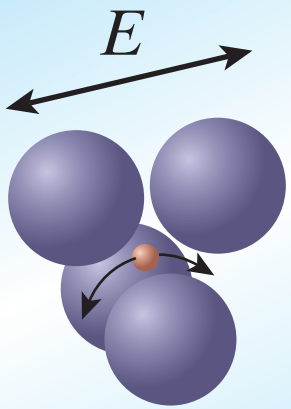

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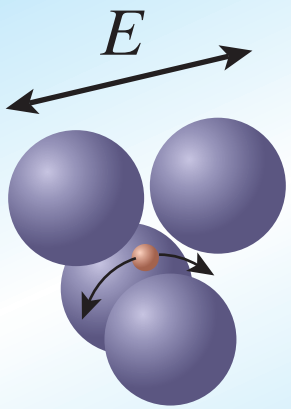

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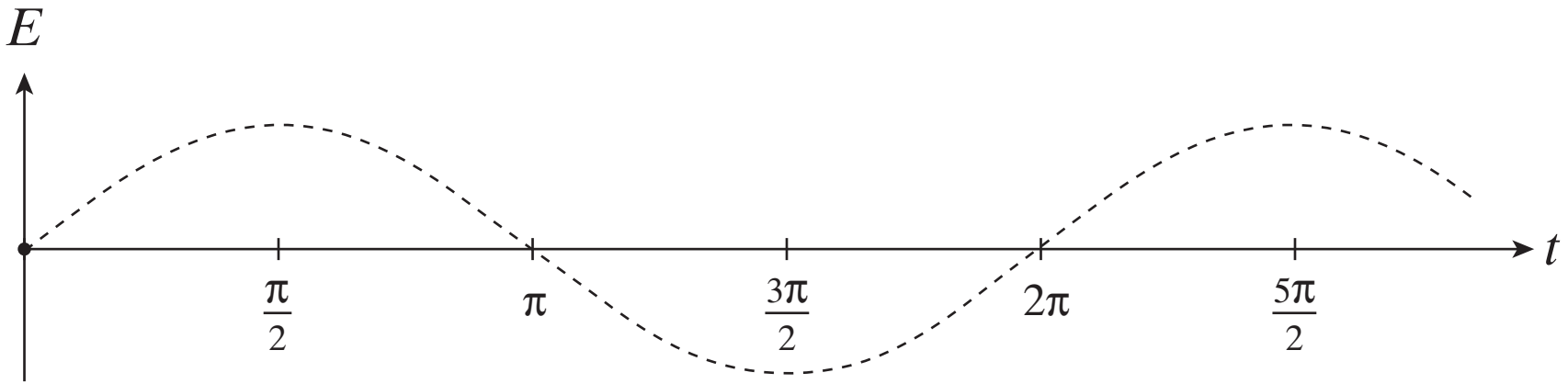
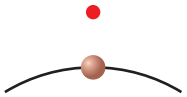
Nonlinear optics



$E = 0$



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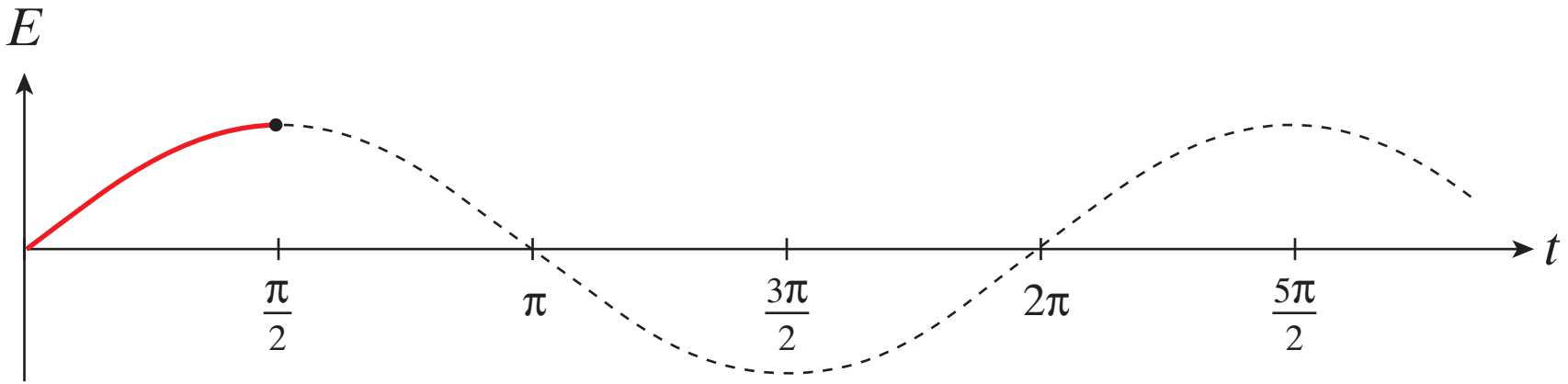

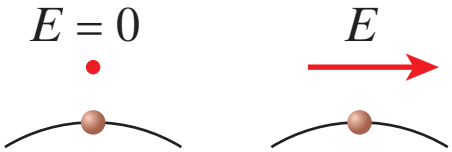

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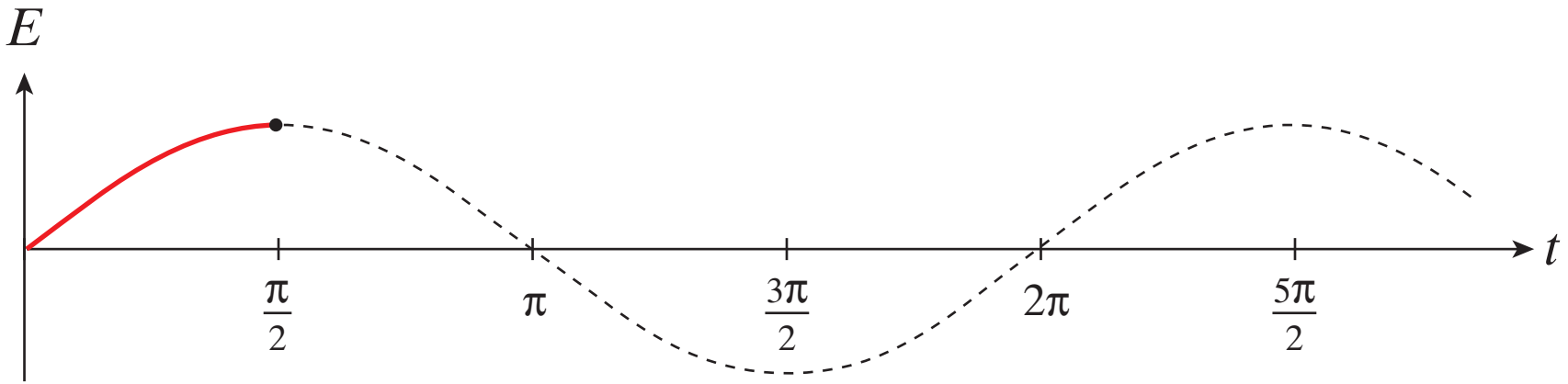

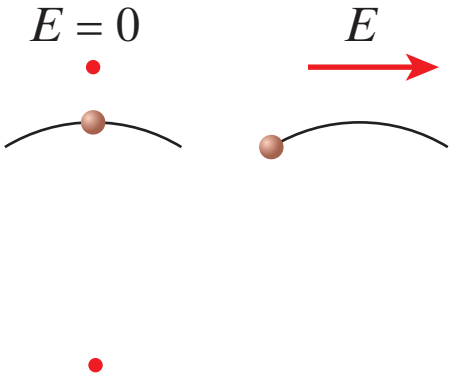

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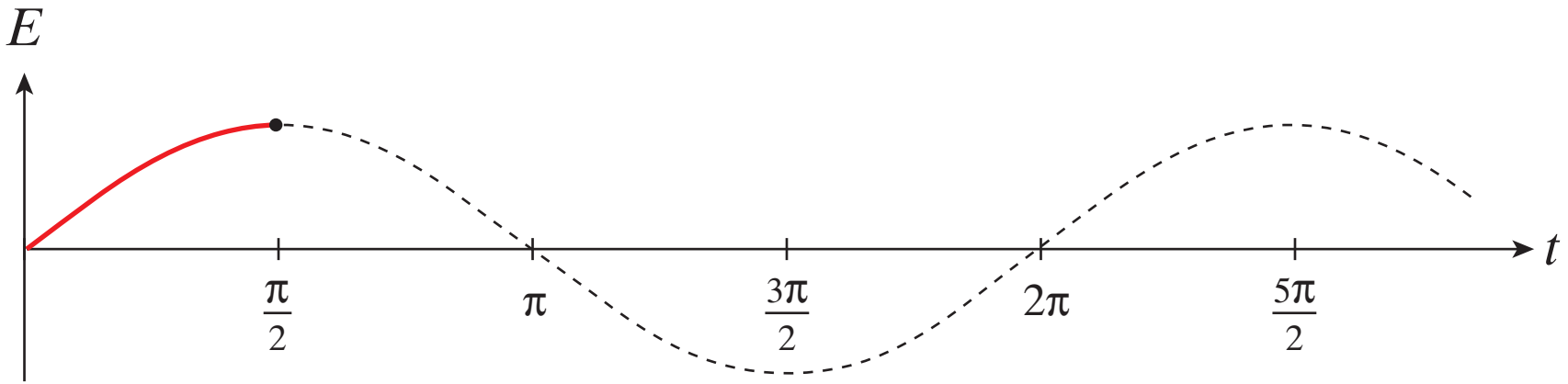

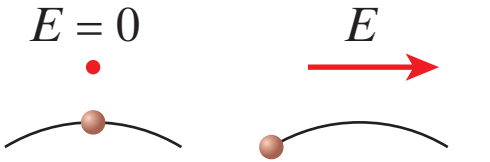

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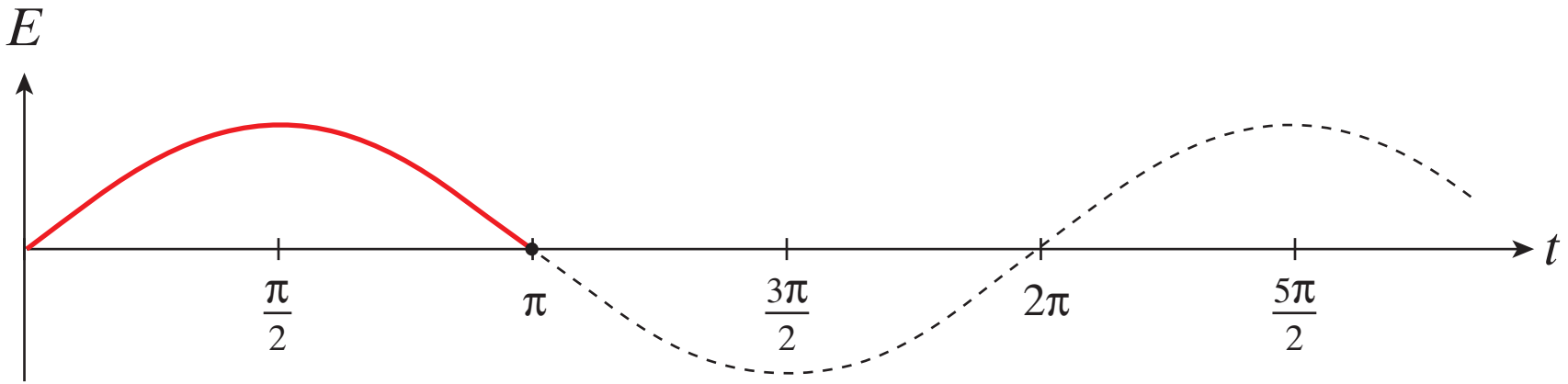

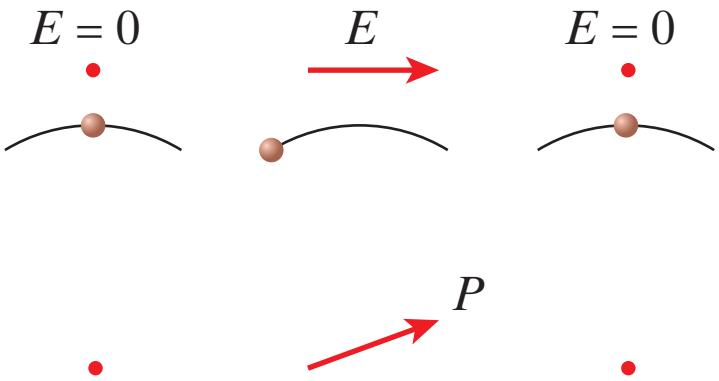

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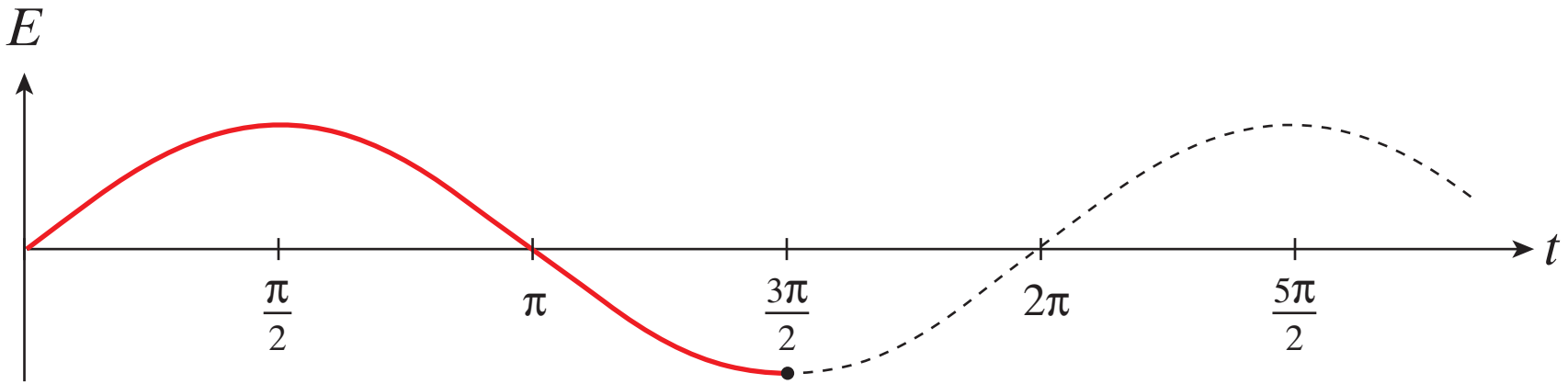

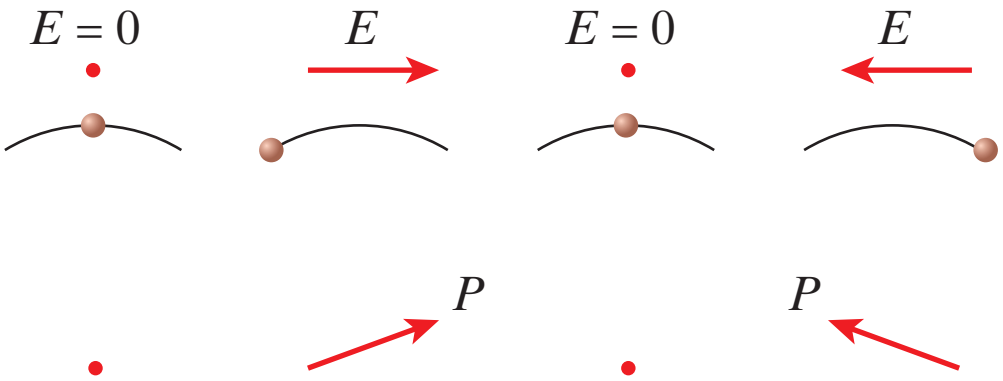

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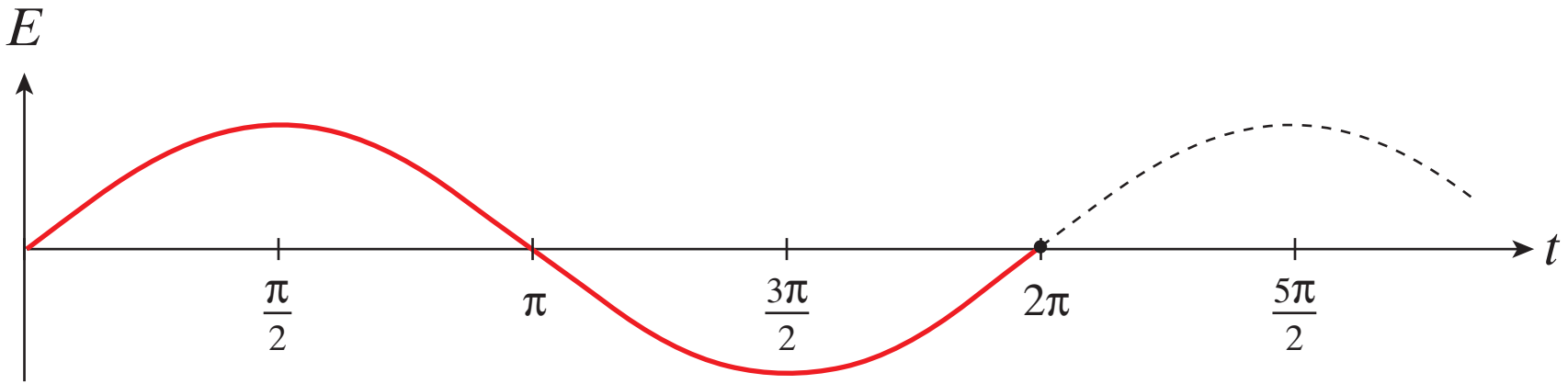

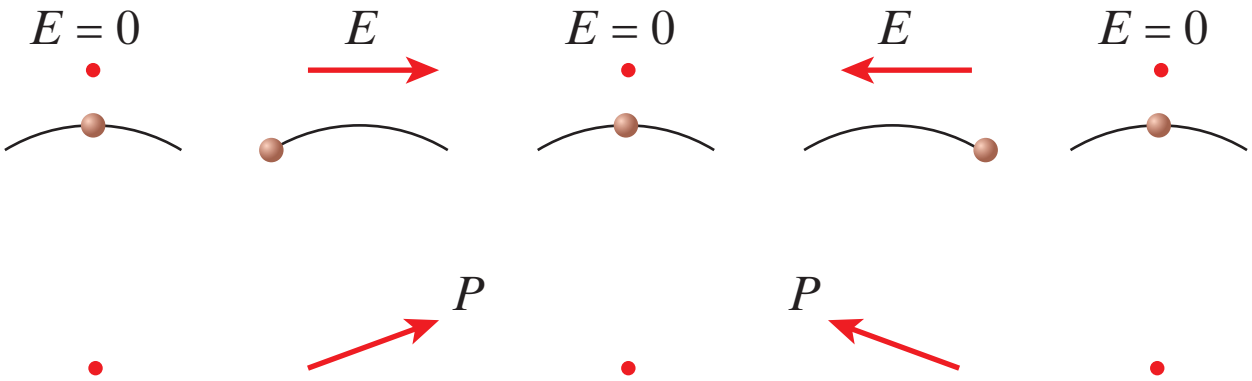

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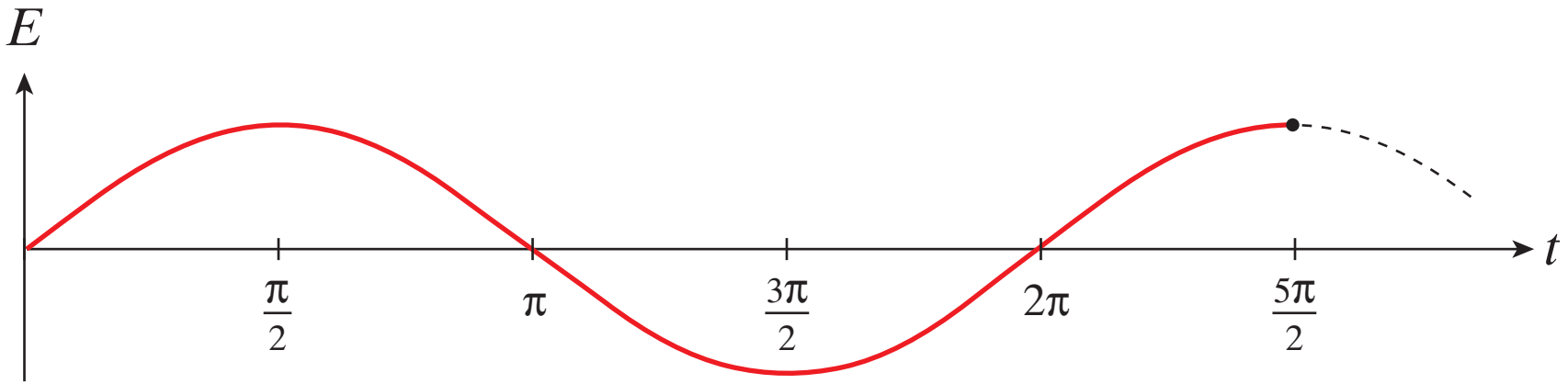

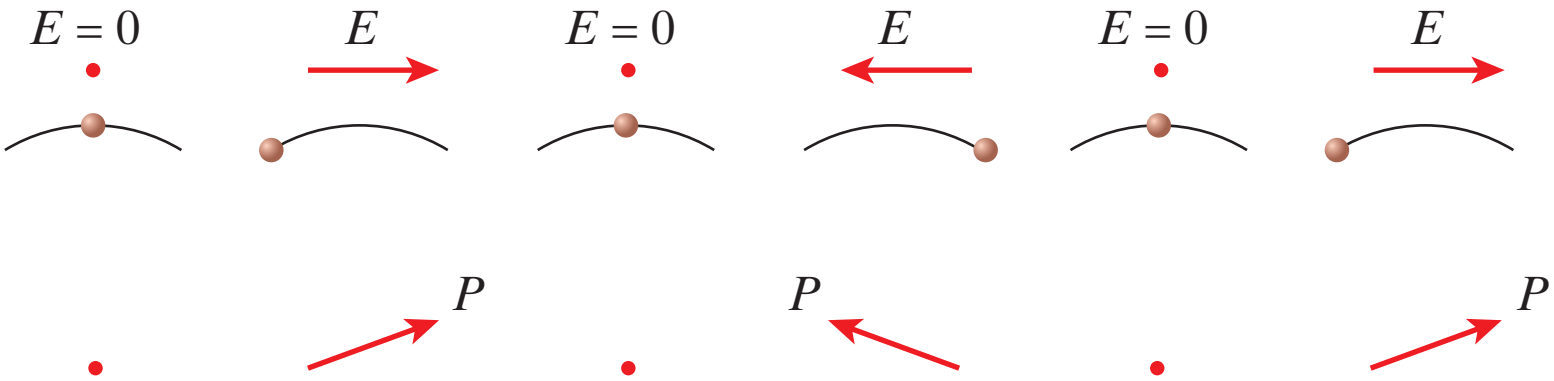

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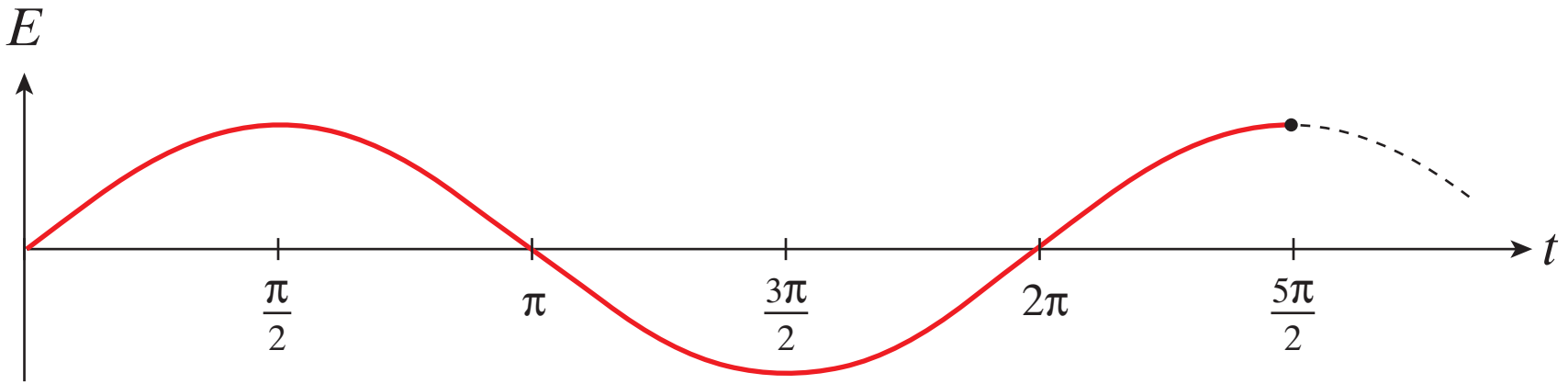

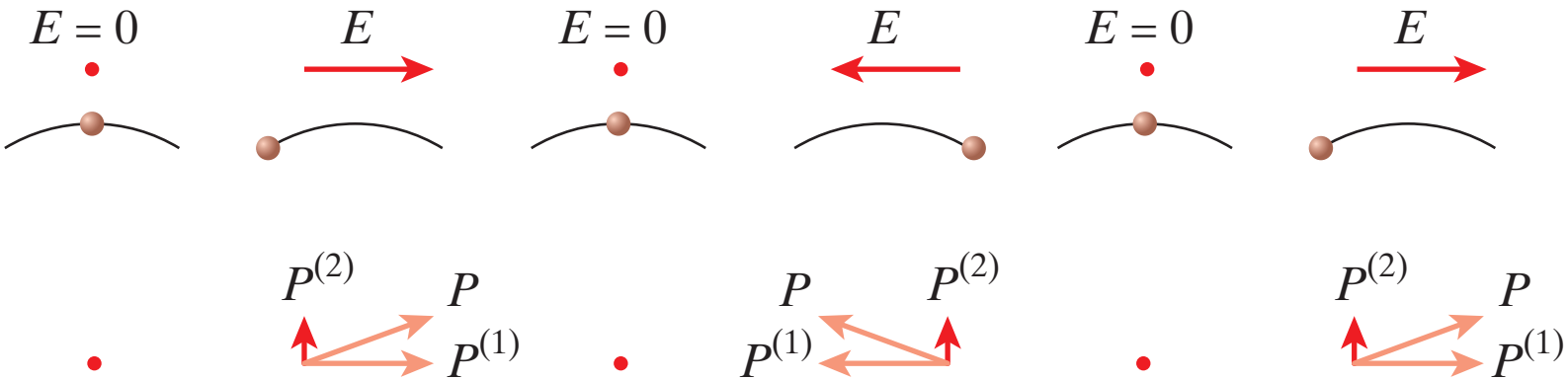

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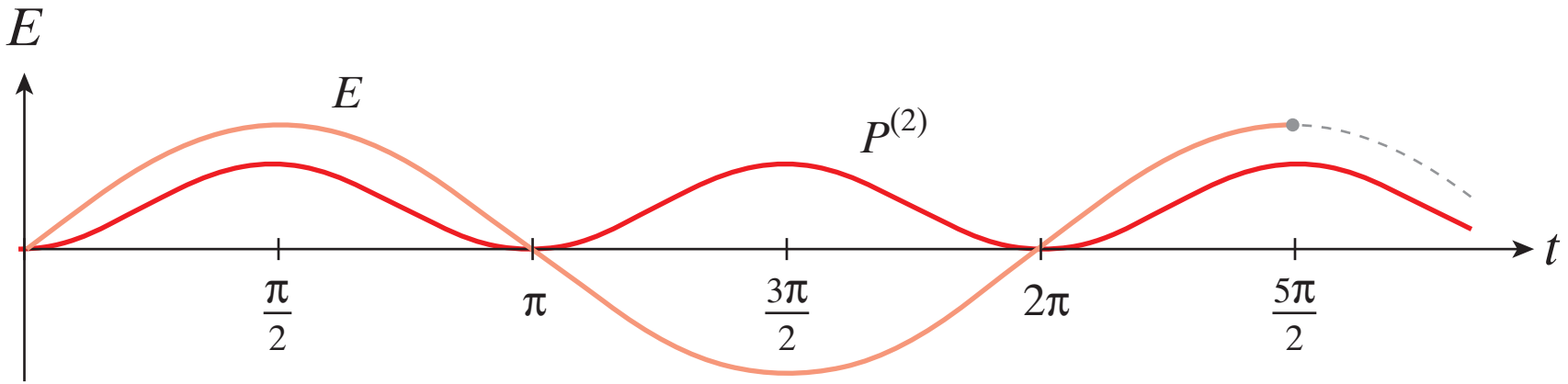

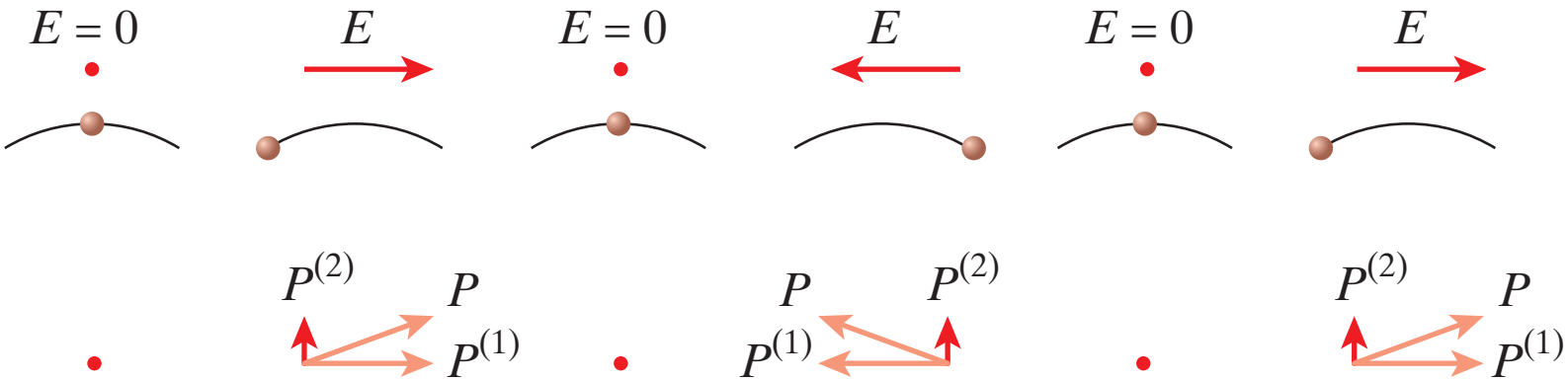

In medium with inversion symmetry

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

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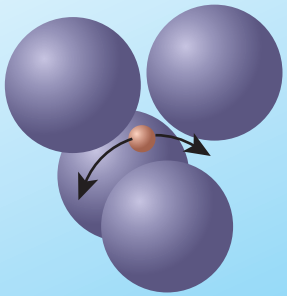
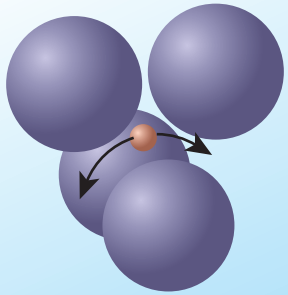
… but …



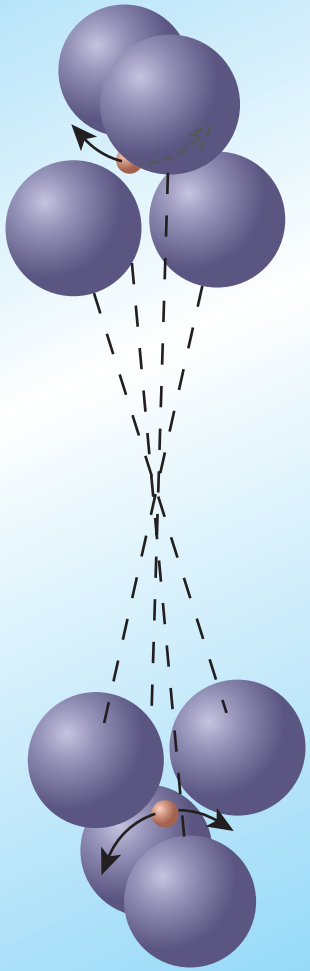
Nonlinear optics

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



?

Nonlinear optics



Nonlinear optics

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Nonlinear optics

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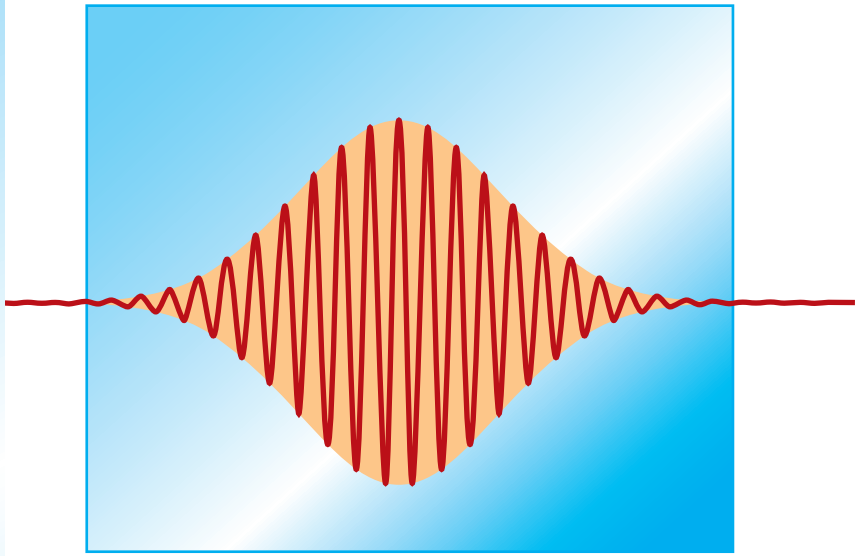
$$n = \sqrt{\epsilon} = \sqrt{1 + \chi_{eff}} \approx \sqrt{1 + \chi^{(1)}} + \tfrac{1}{2}\frac{\chi^{(3)}I}{\sqrt{1 + \chi^{(1)}}} = n_o + n_2 I$$

Nonlinear optics

Intensity dependent index of refraction:

$$n = n_o + n_2 I$$



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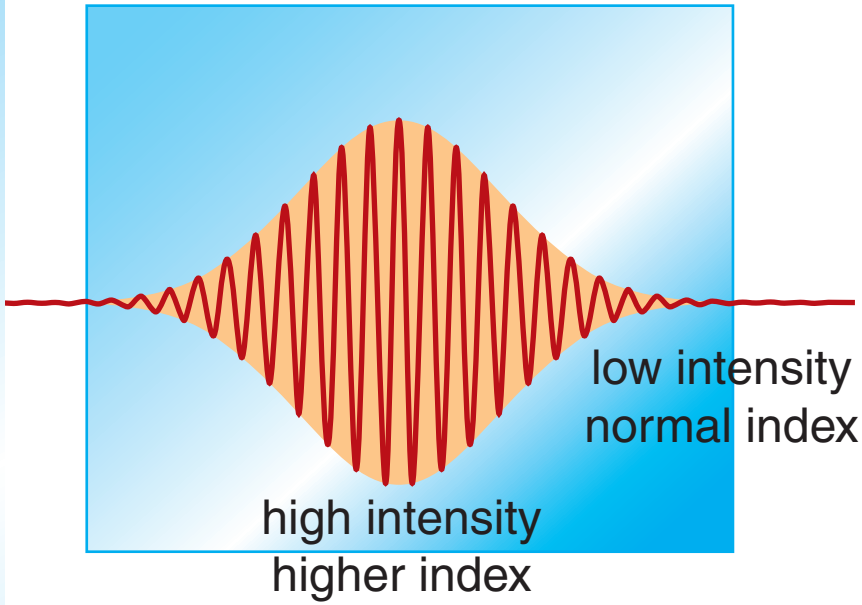

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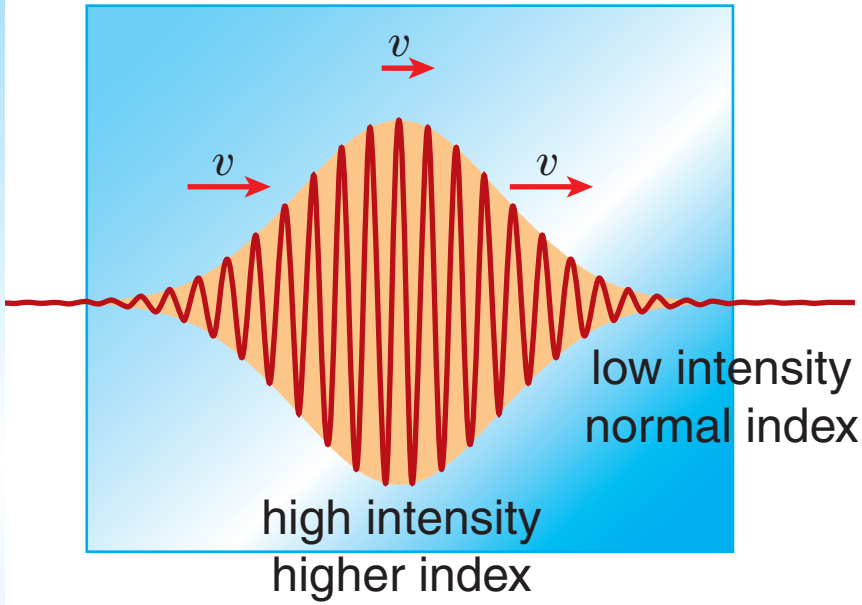

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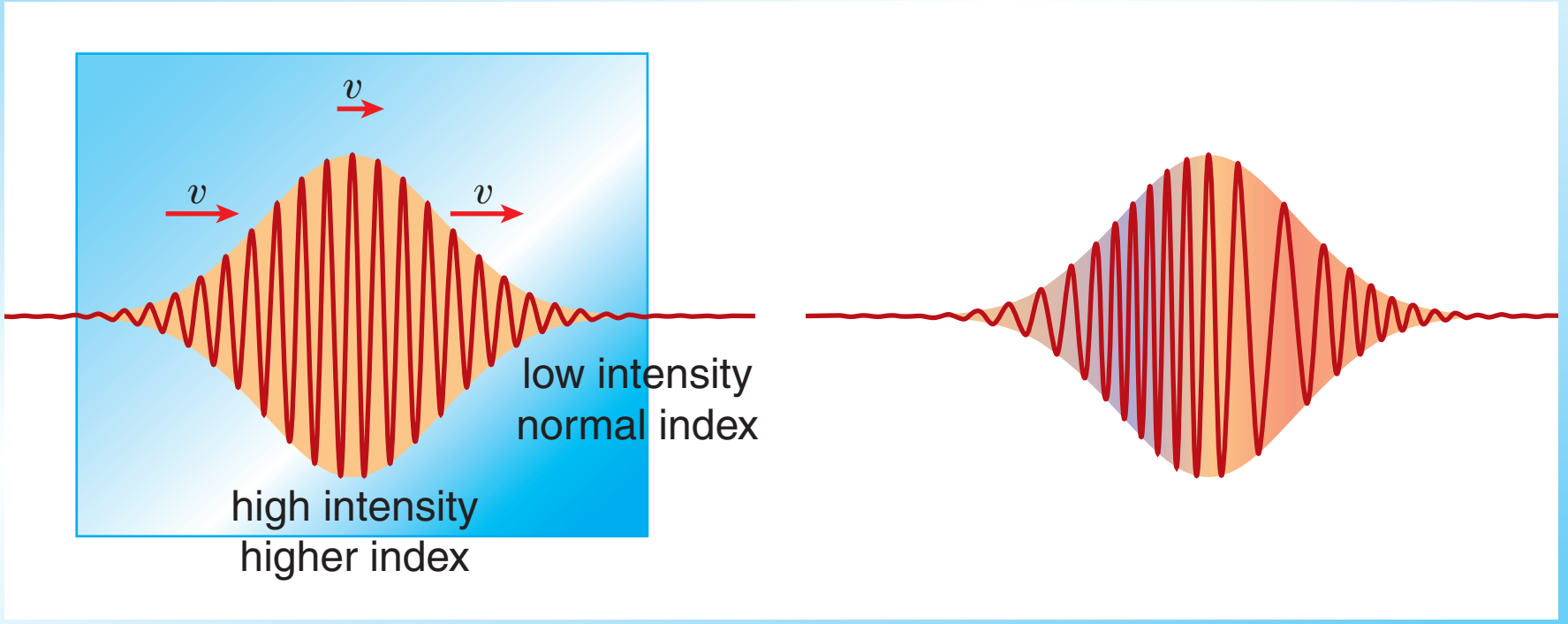

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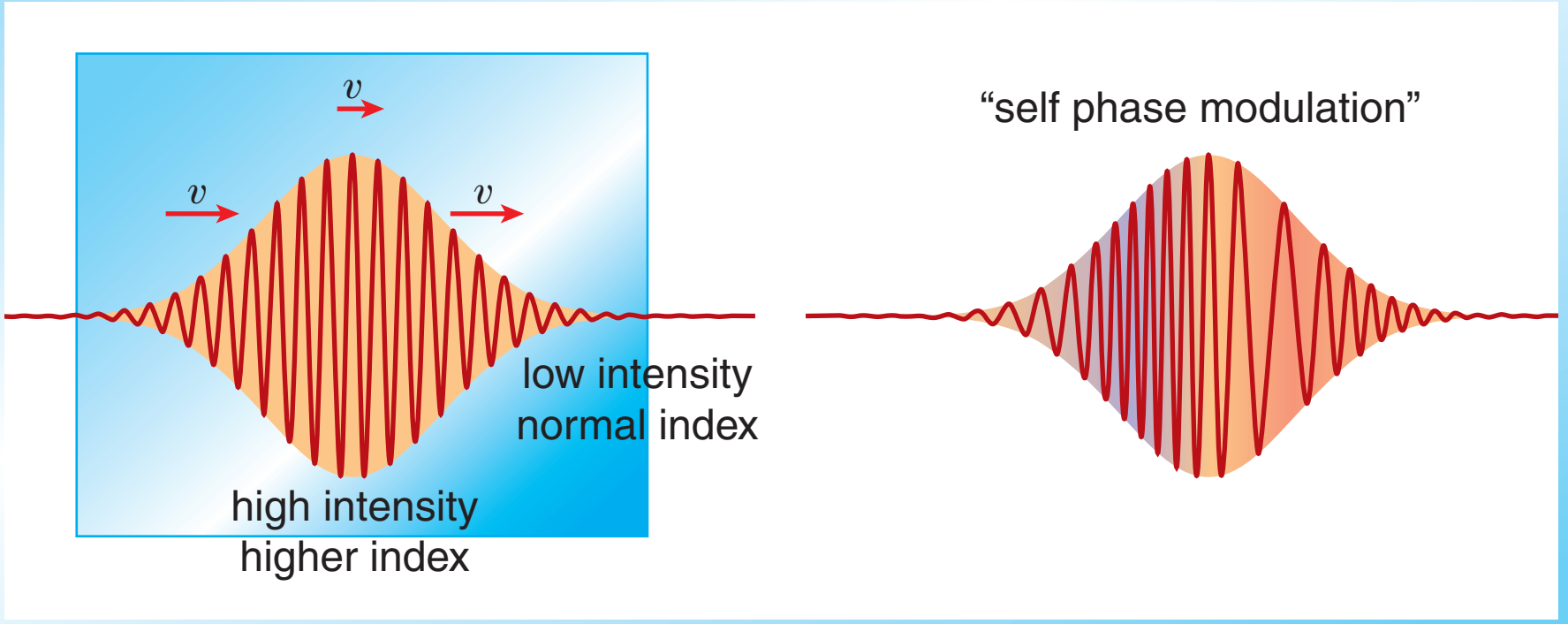

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Nonlinear optics

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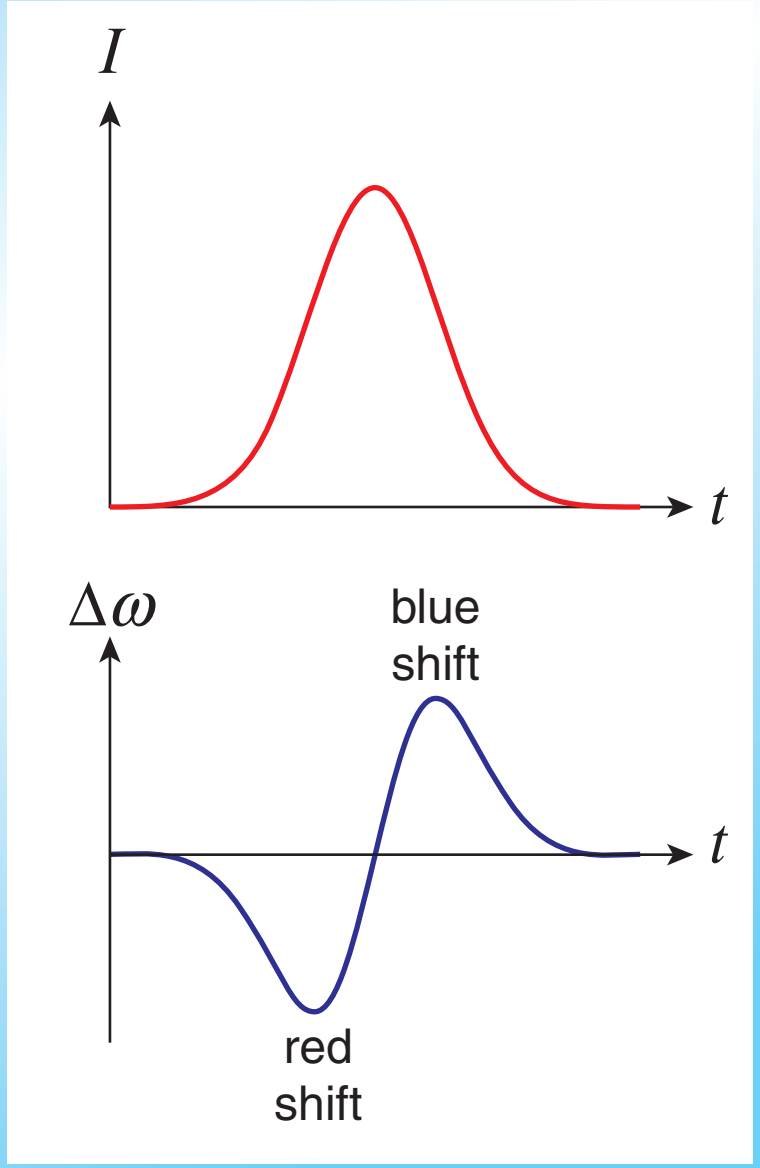
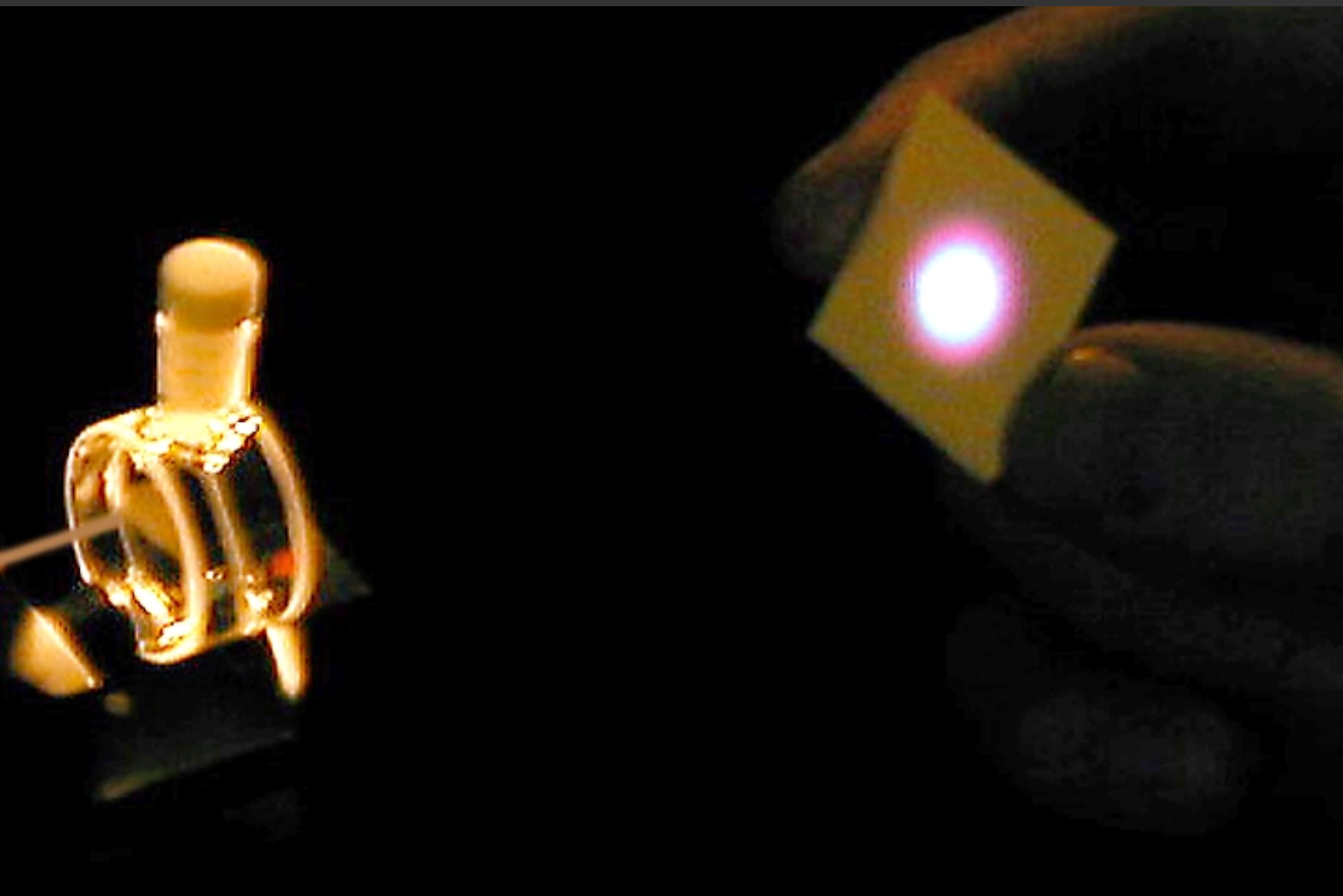
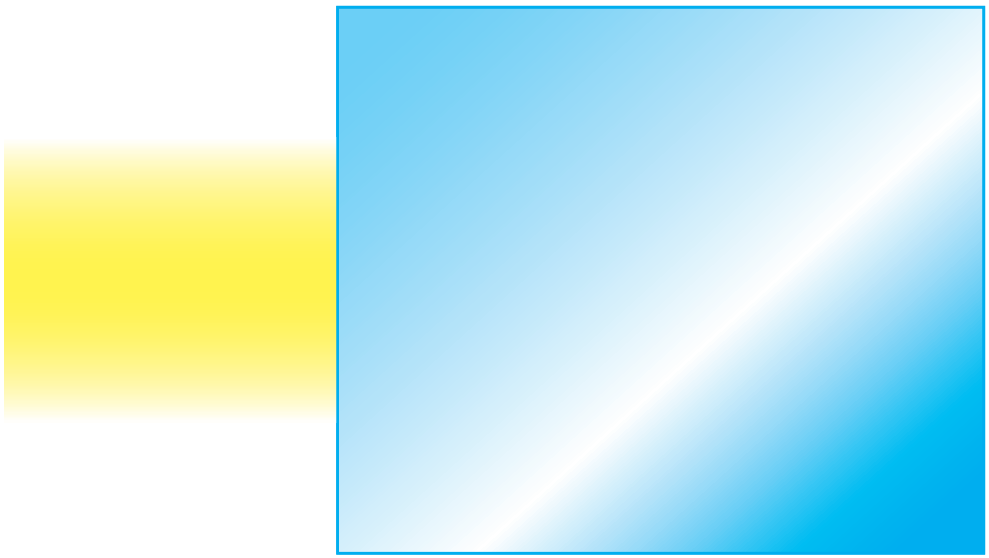


Nonlinear optics



Spatial intensity profile…



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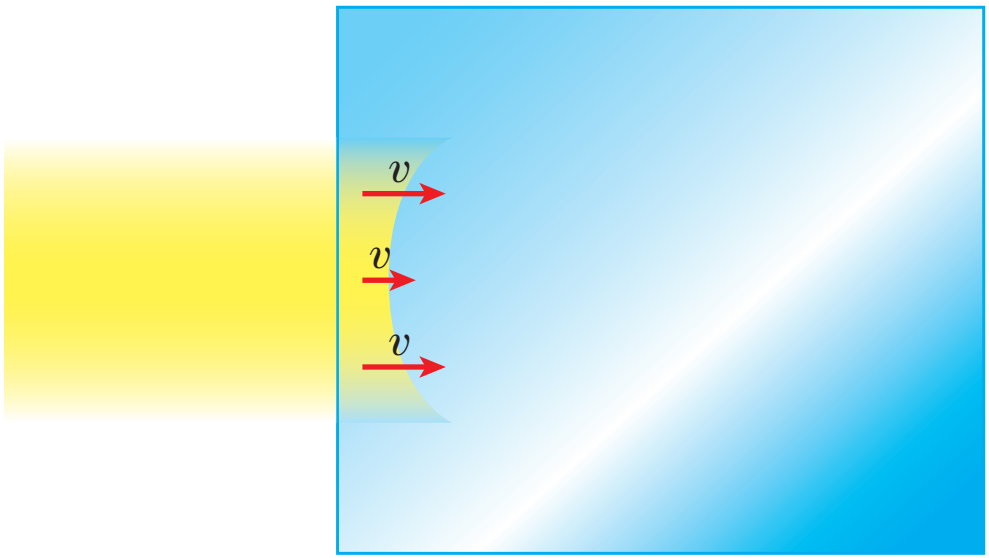
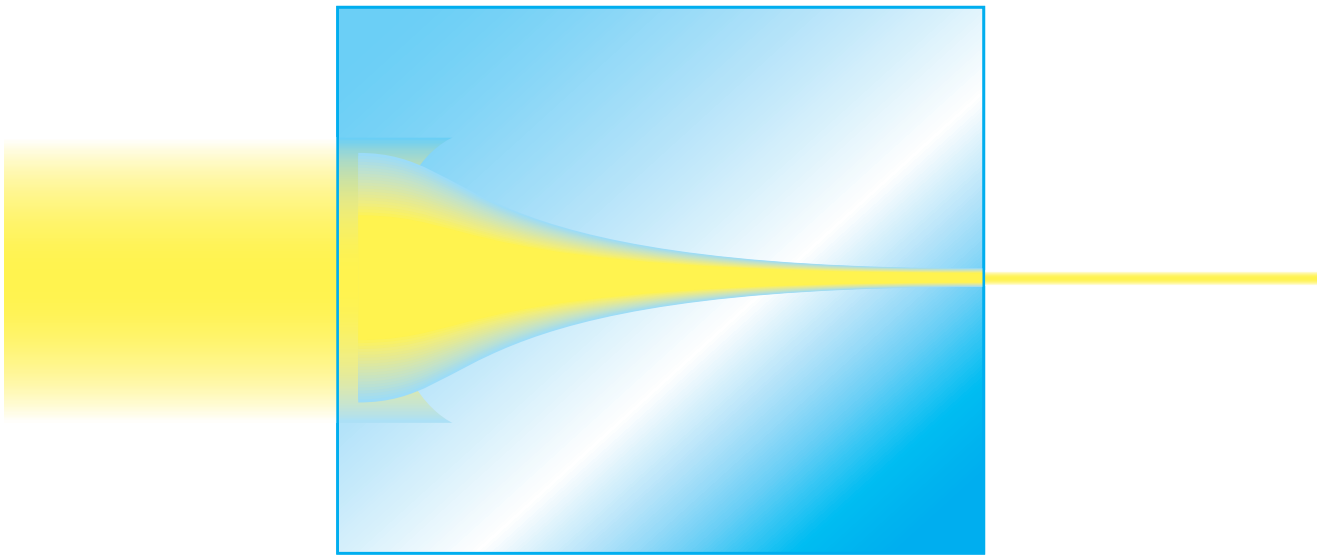


…causes self-focusing

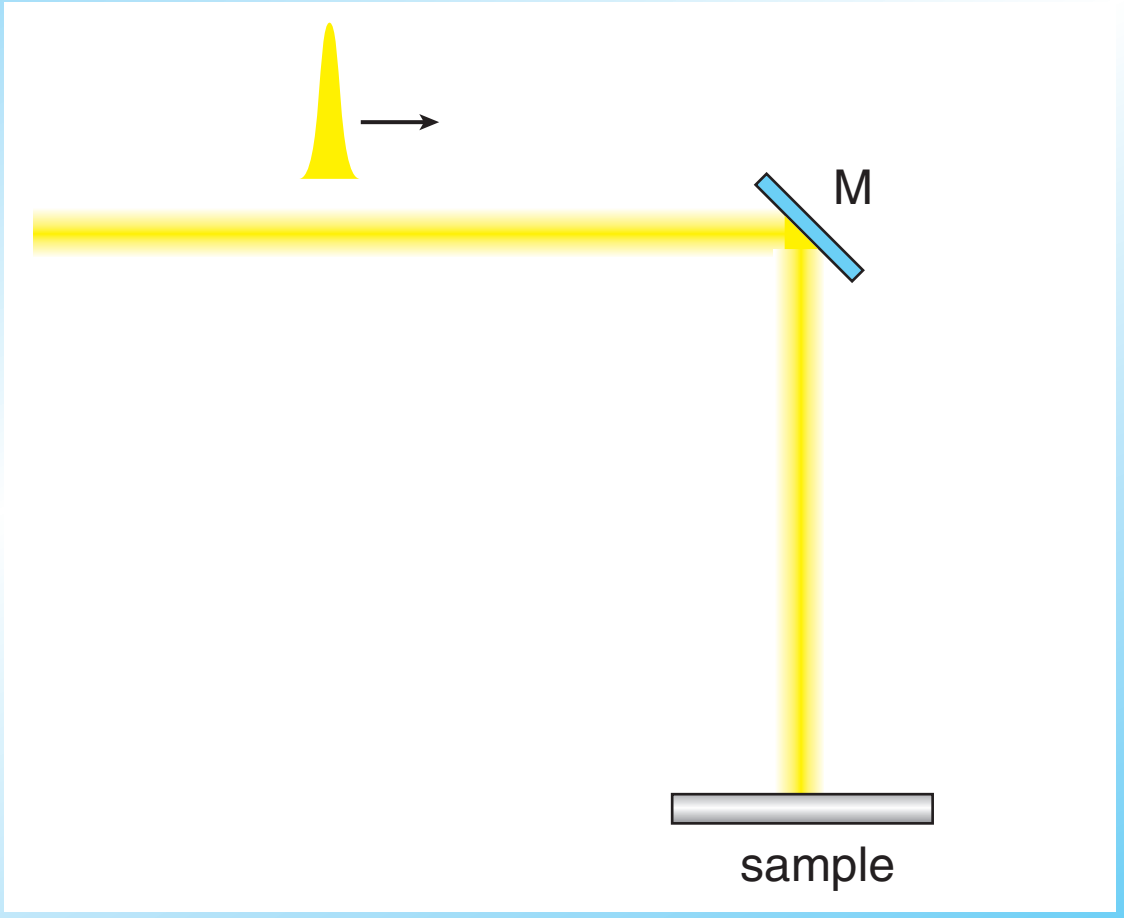


Outline

- linear and nonlinear propagation
- femtosecond measurements
- examples

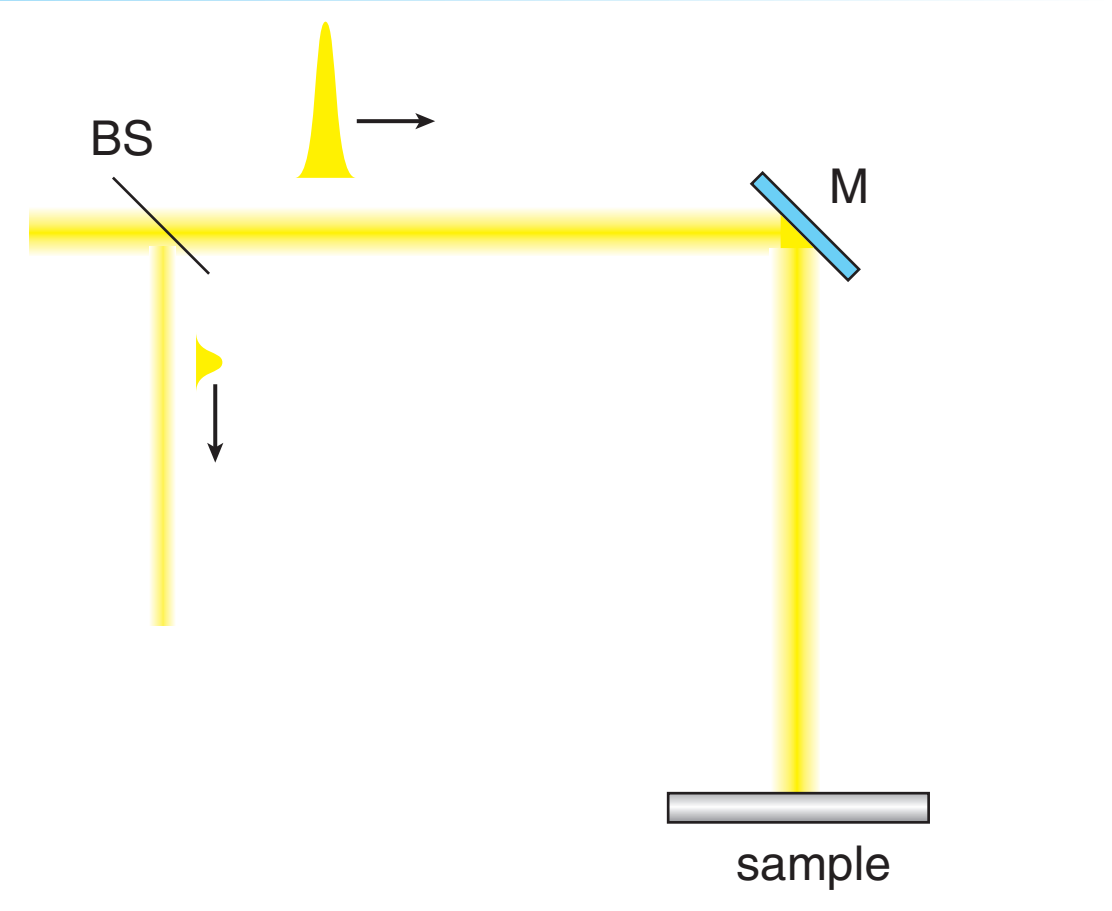
Introduction

How to measure on the femtosecond time scale?



Introduction

Use pump-probe technique



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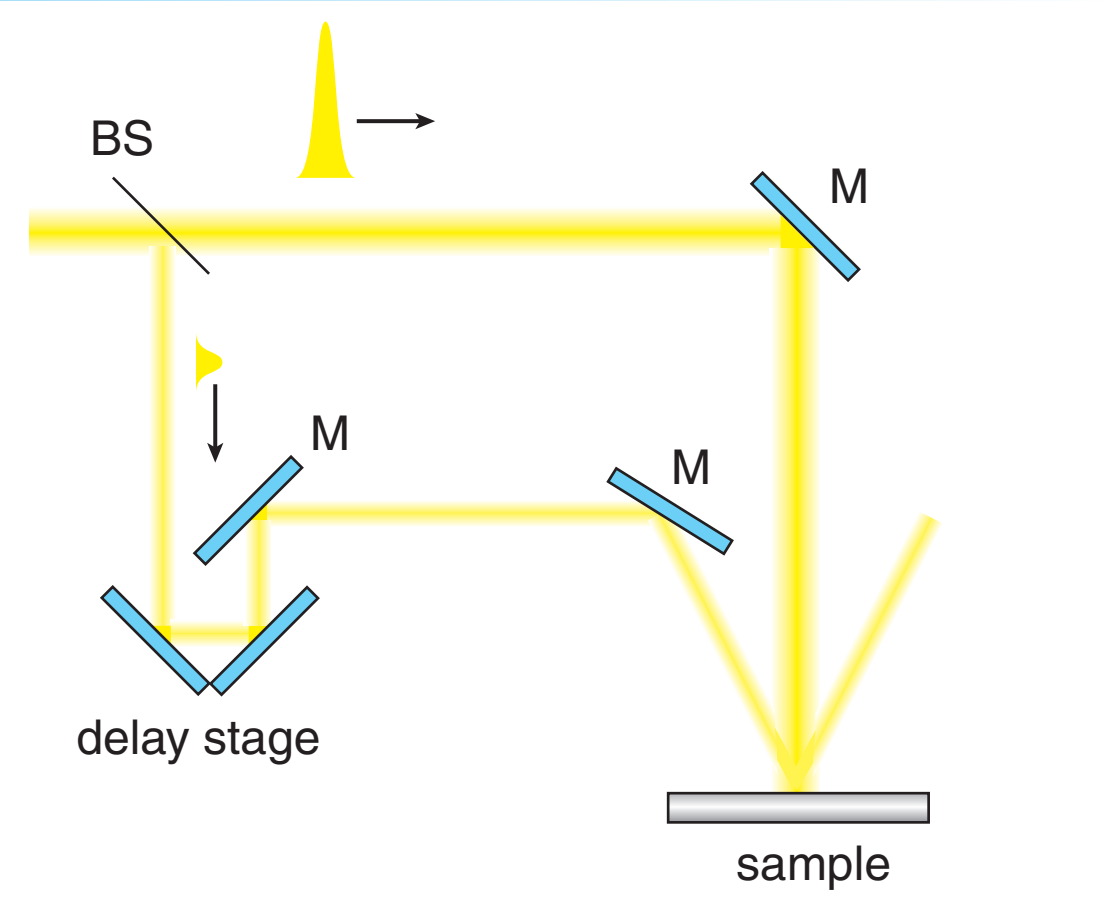

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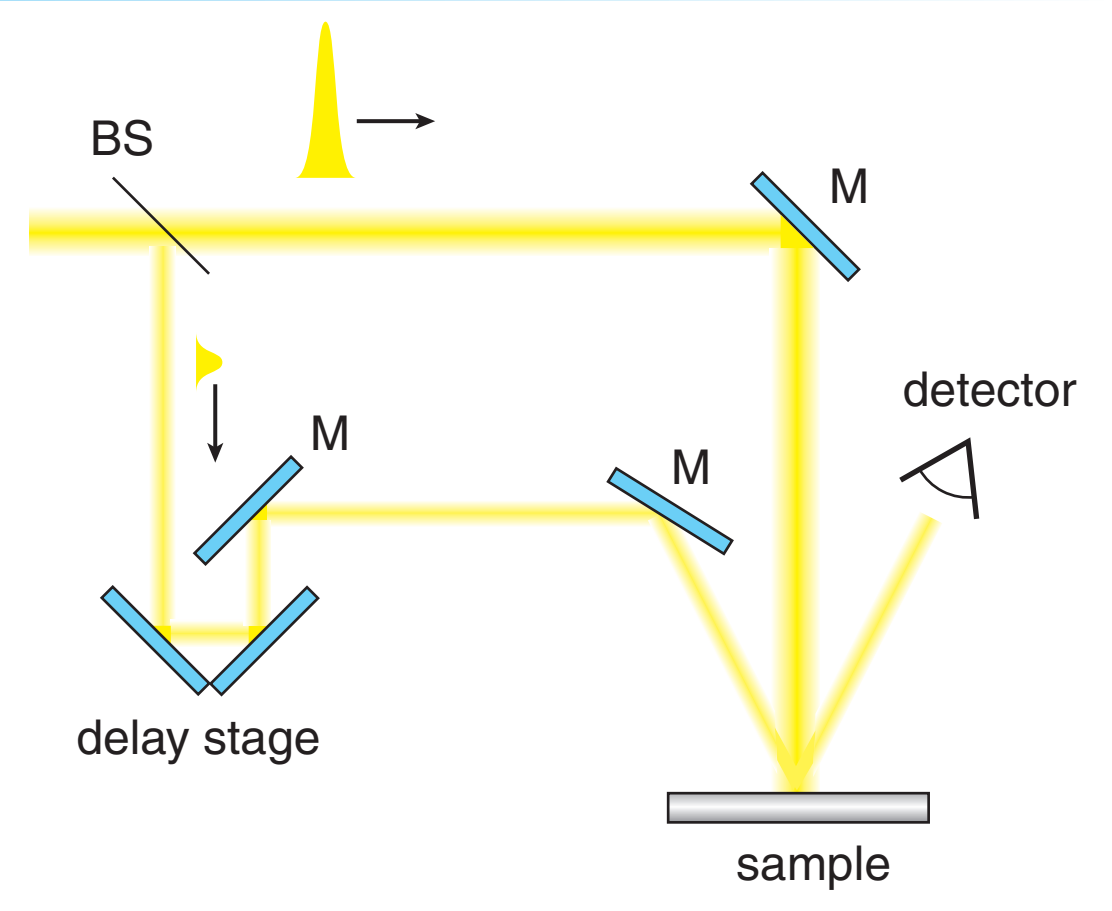
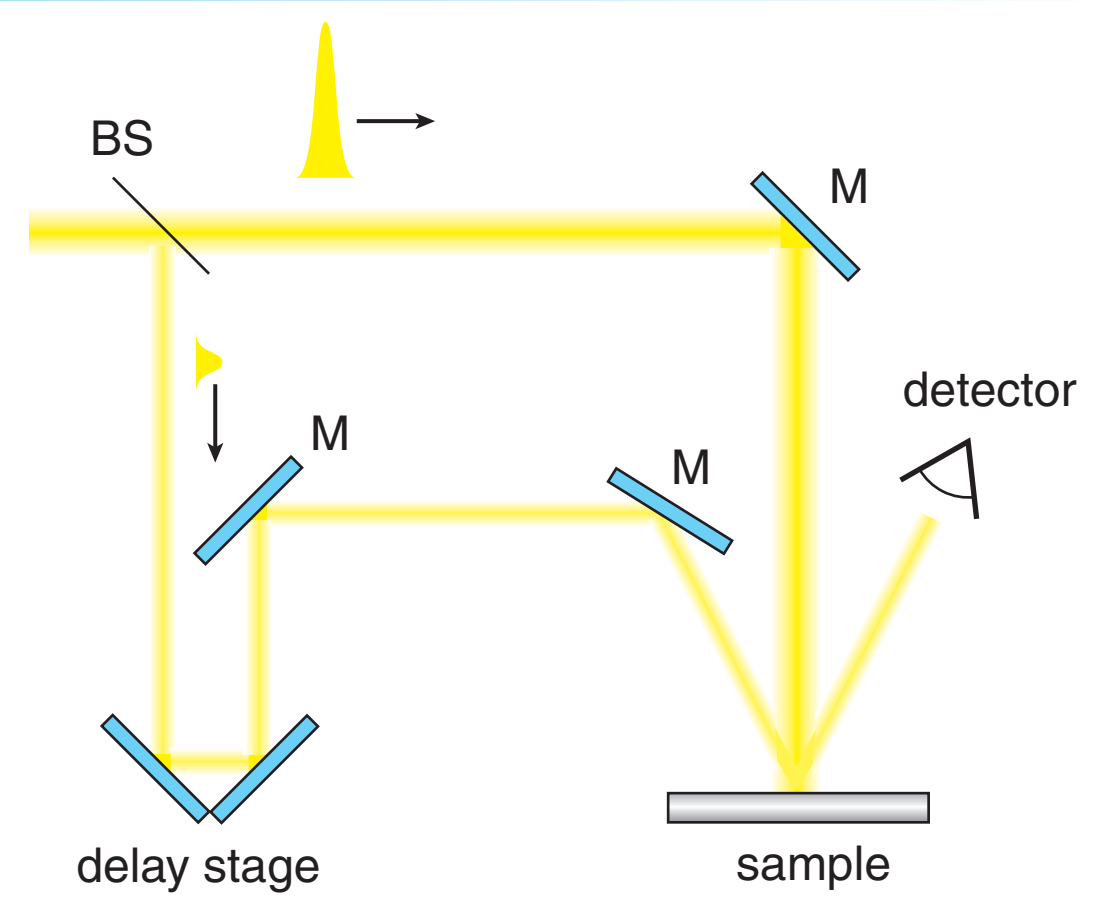


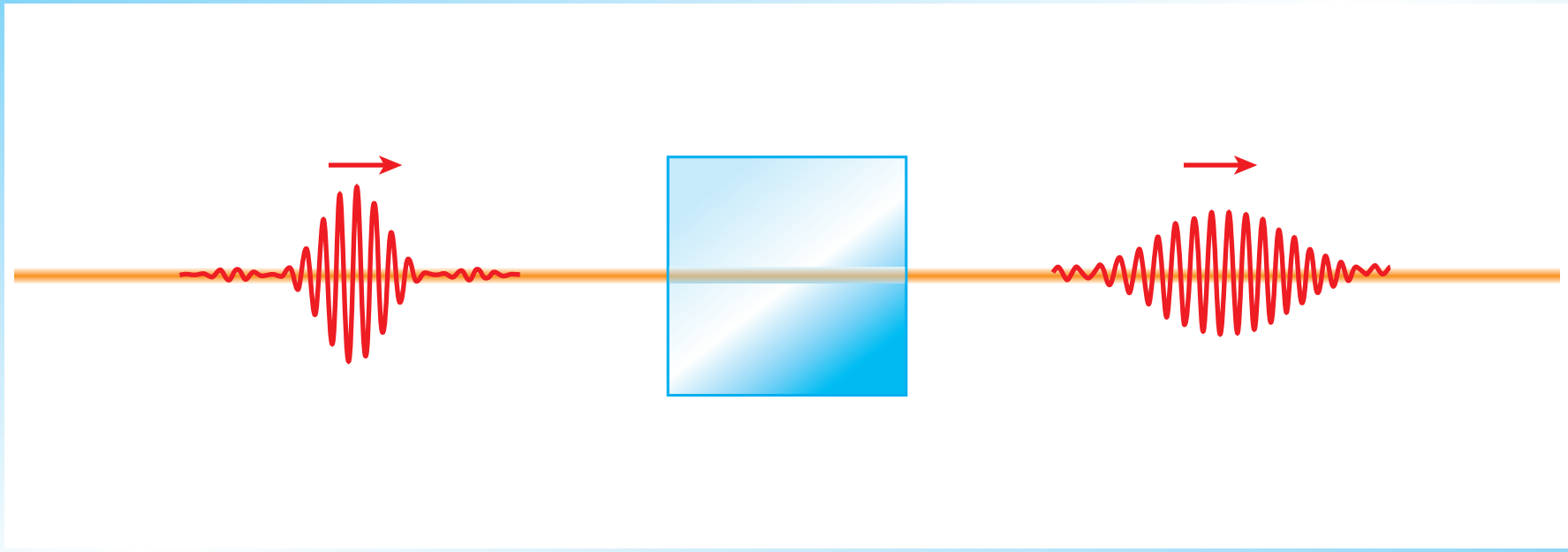
Introduction

Vary delay to get time resolution



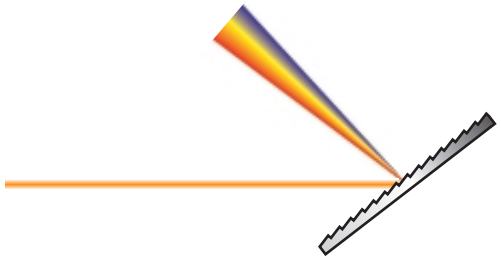
Dispersion compensation

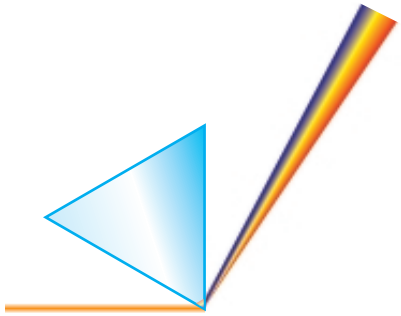
Dispersion stretches the pulse



Compensate by rearranging spectral components!

Dispersion compensation





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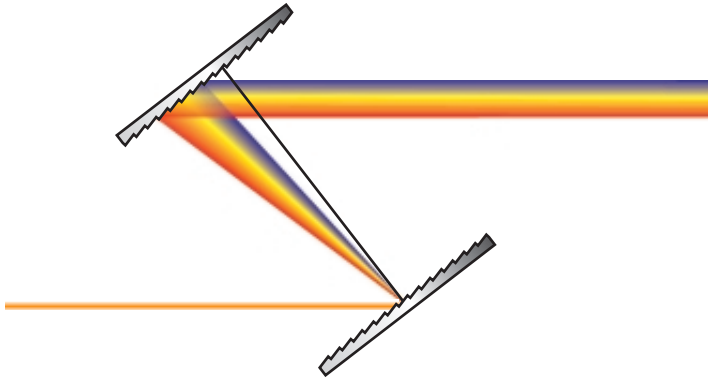

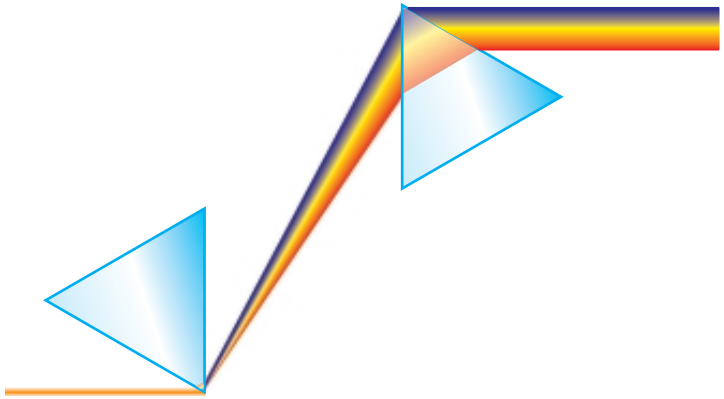

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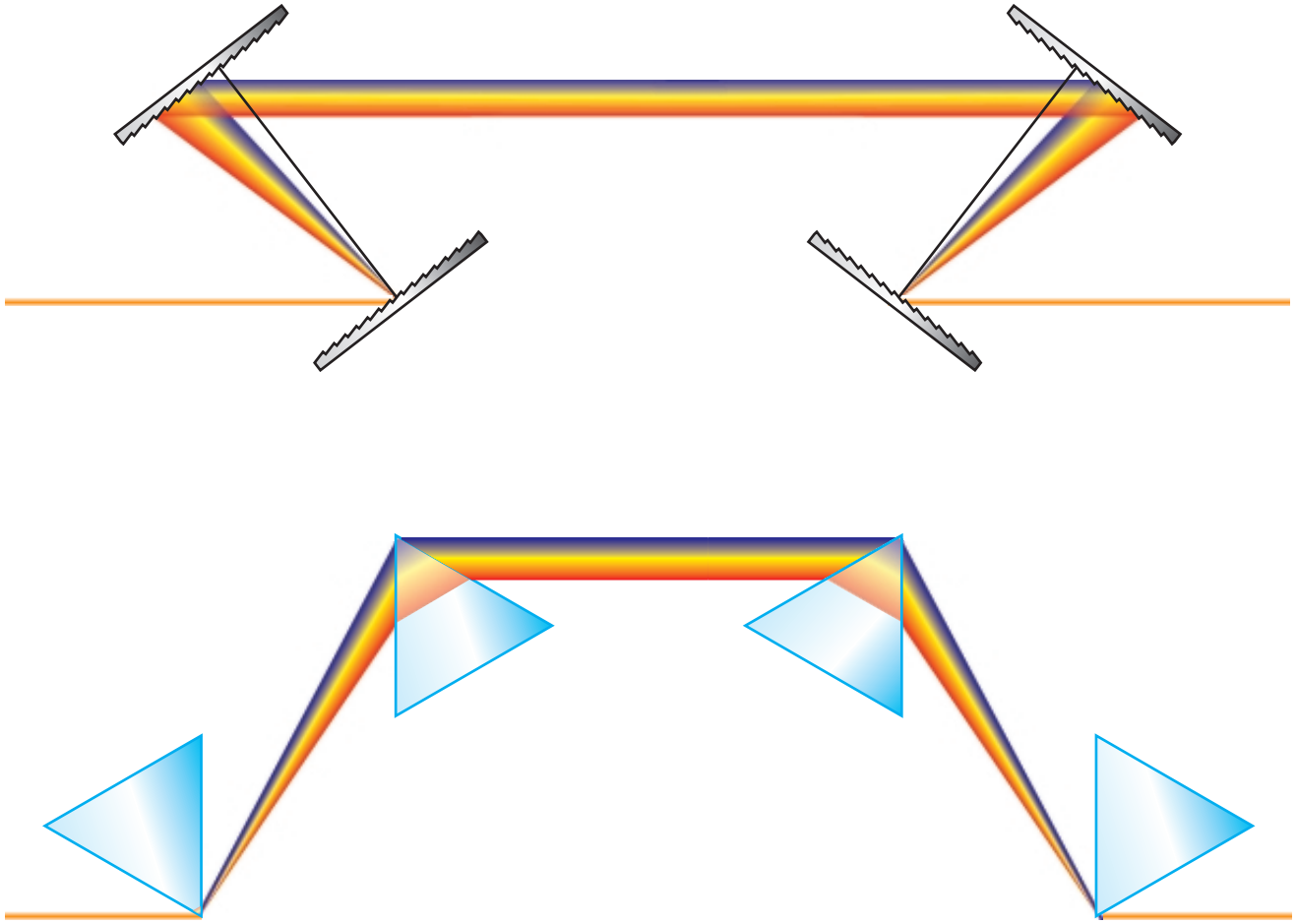
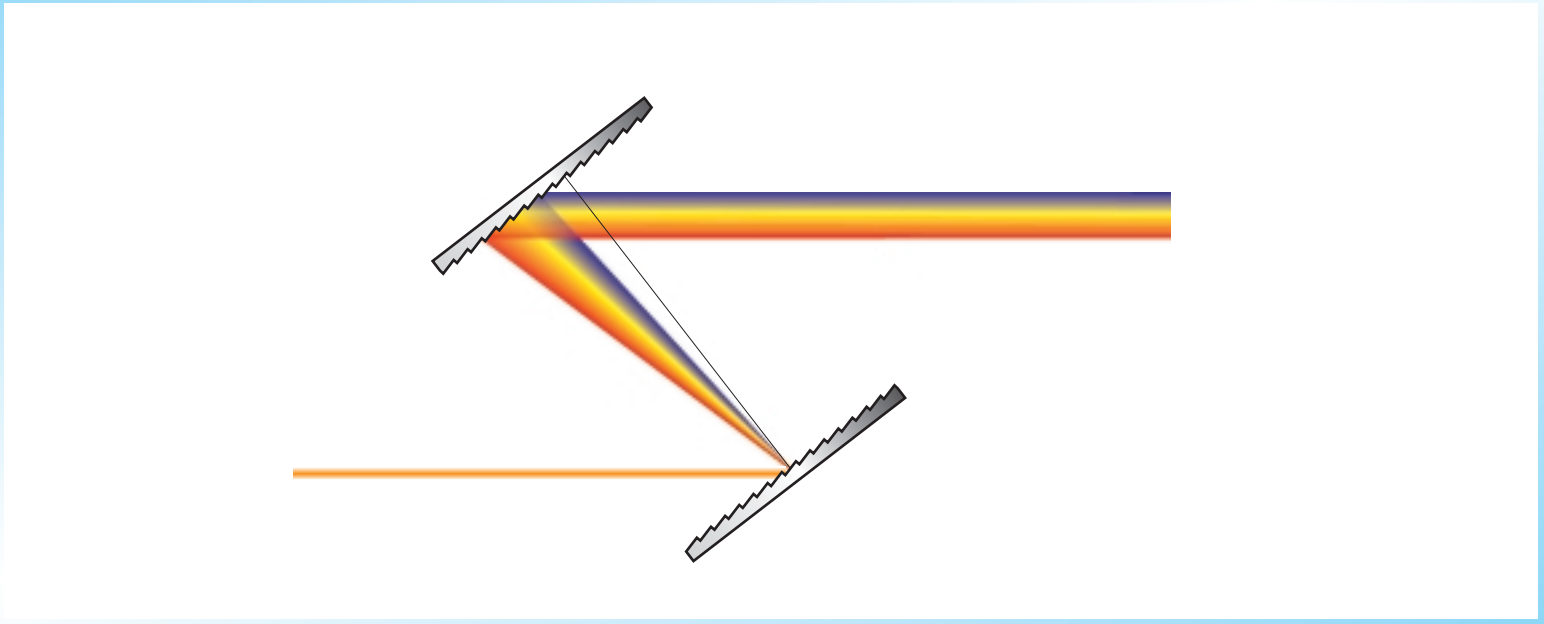


Dispersion compensation

How do these arrangements work?

Dispersion compensation

Does path length difference compensate?



Dispersion compensation

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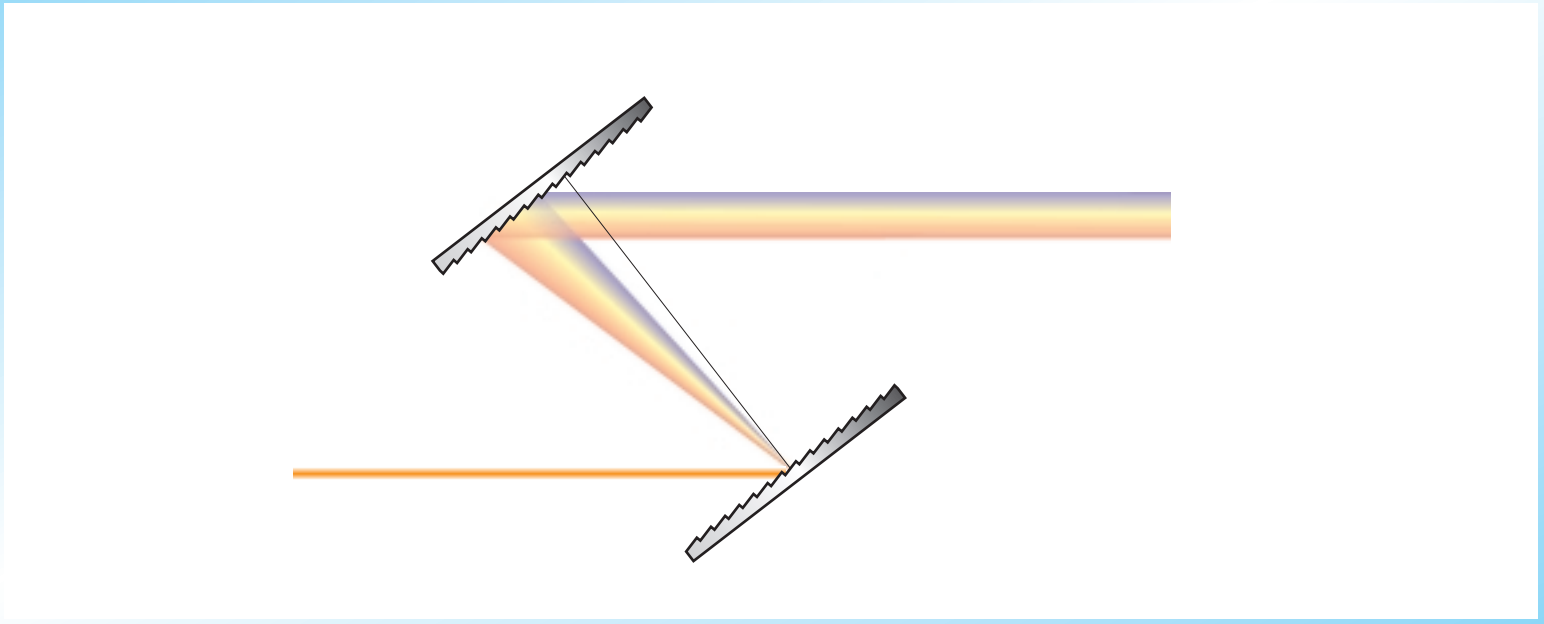

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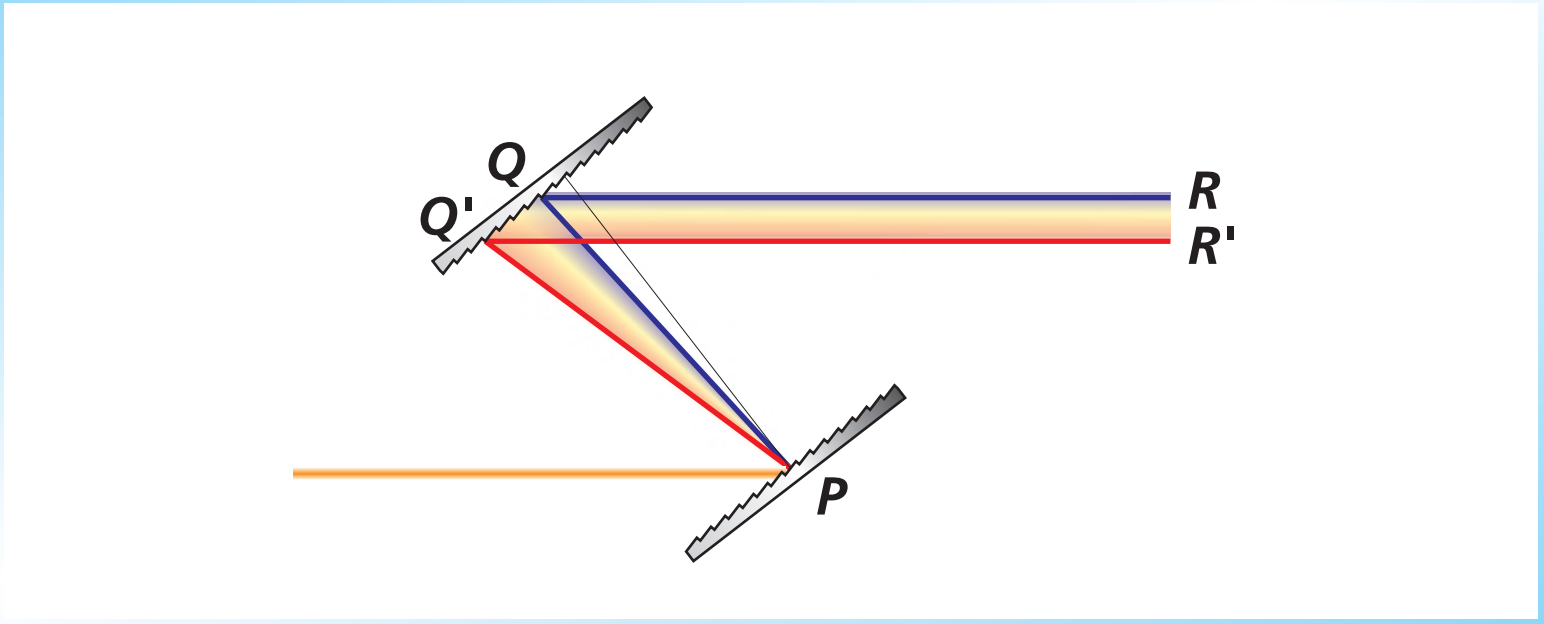
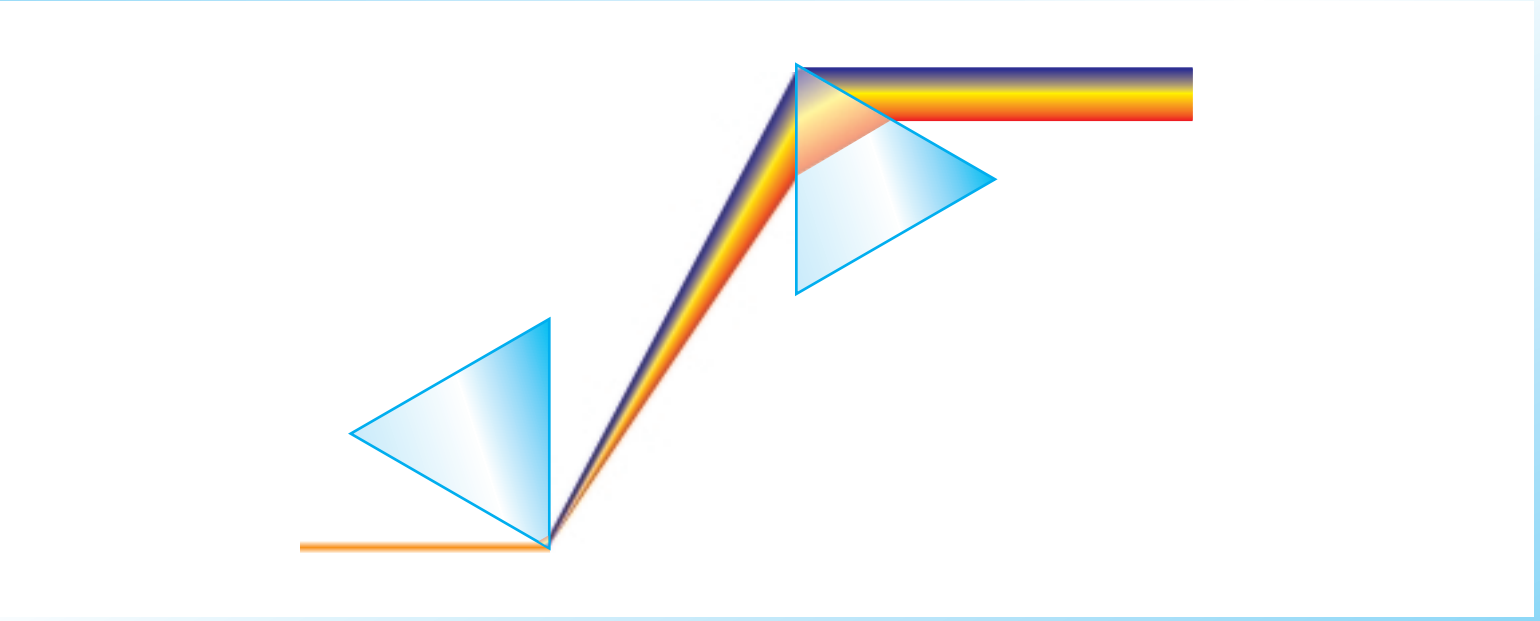


Grating gives low frequency longer path length…

Dispersion compensation

Does path length difference compensate?



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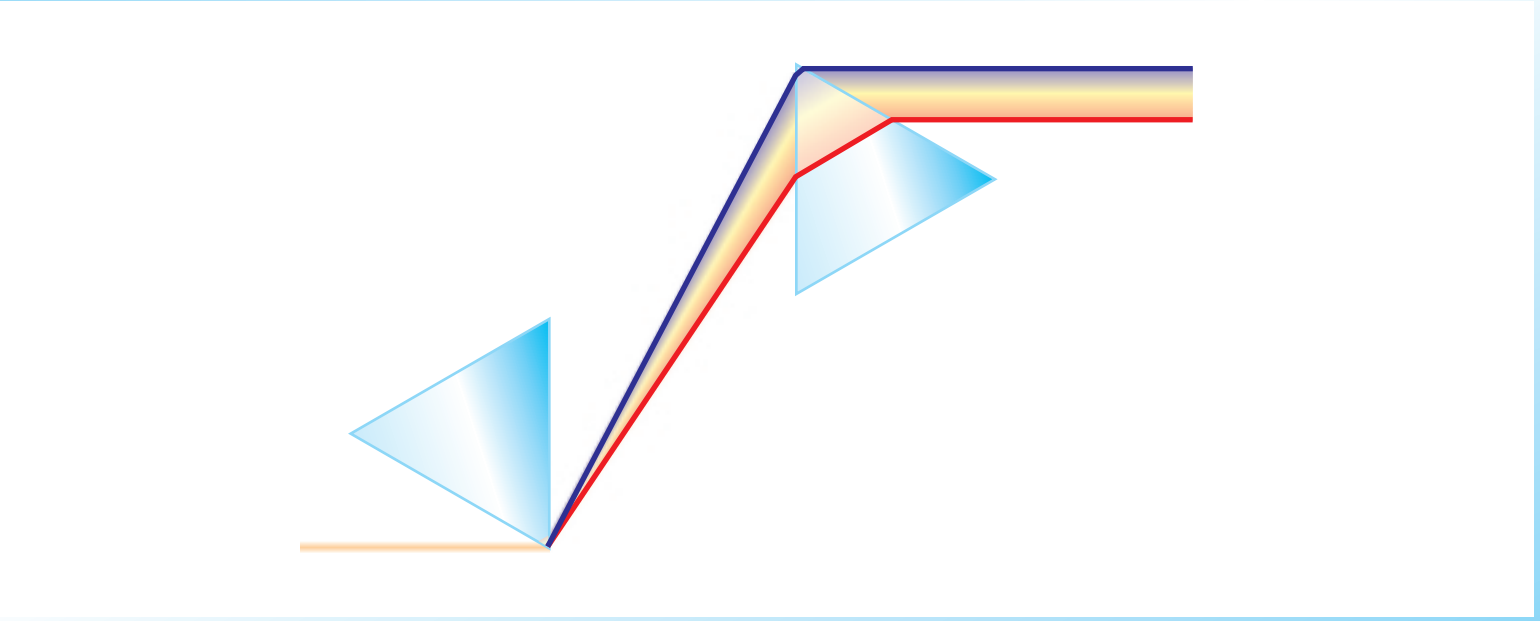

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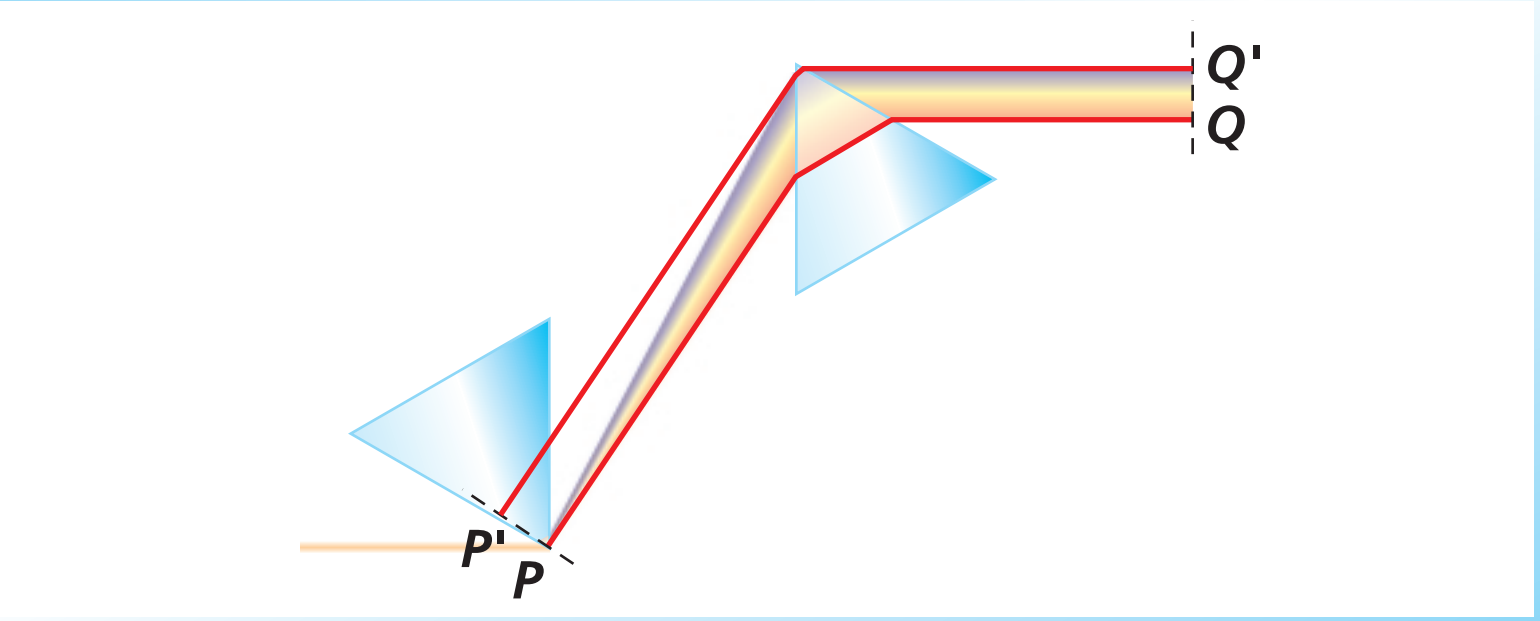
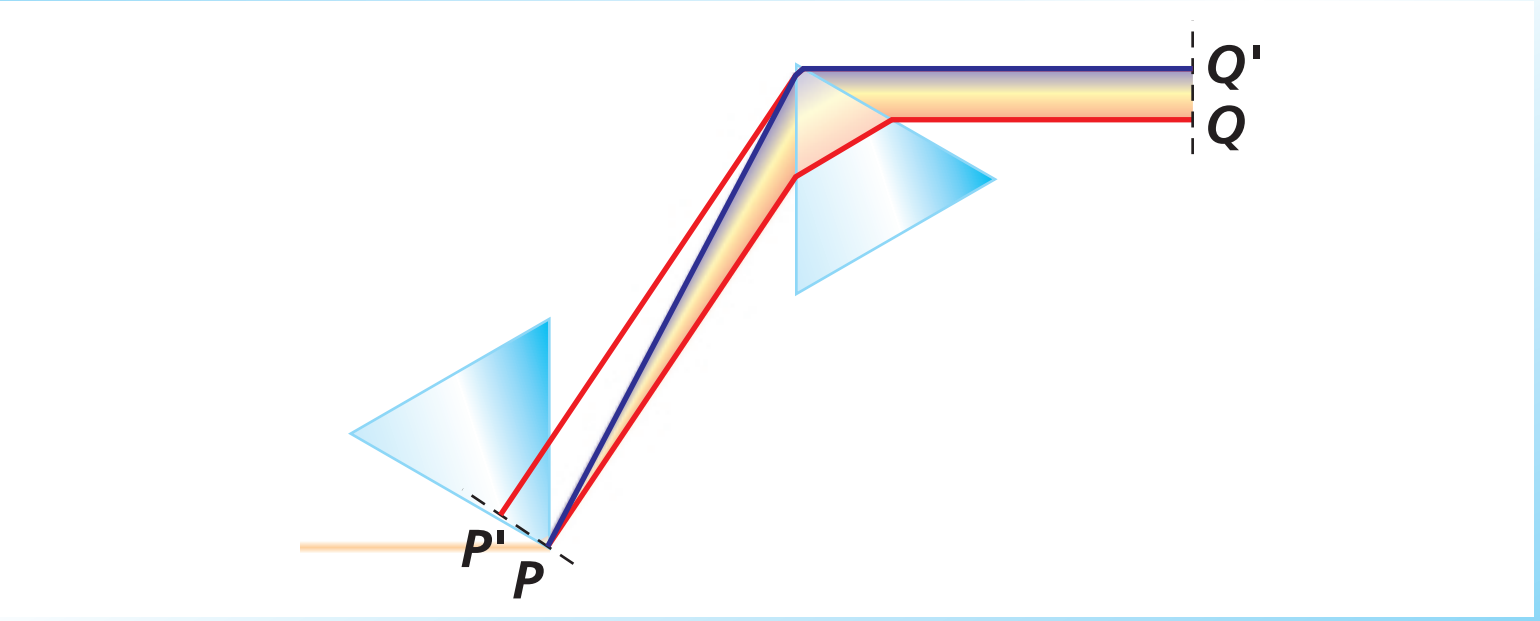


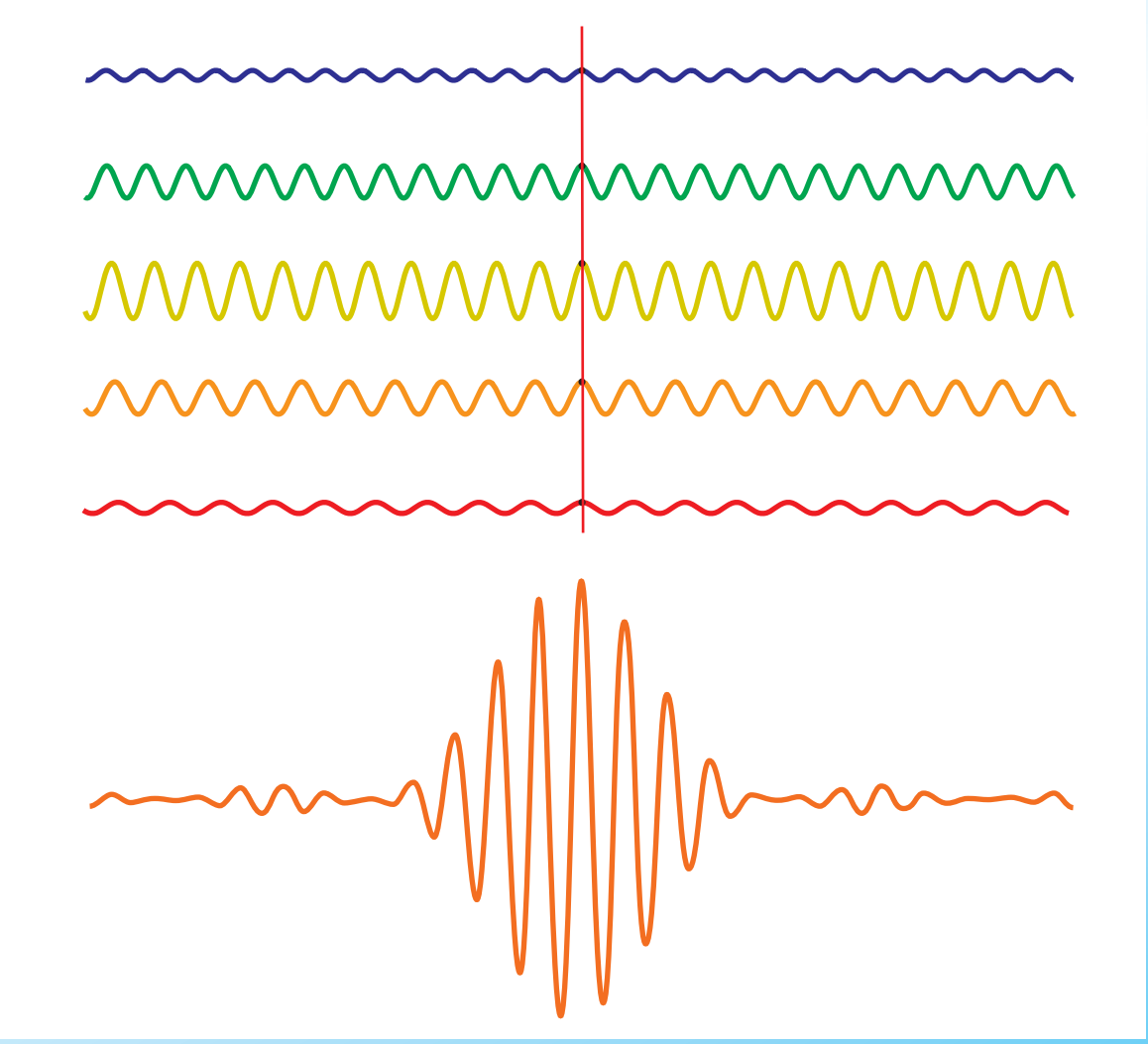
Dispersion compensation

Does path length difference compensate?



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

Dispersion compensation



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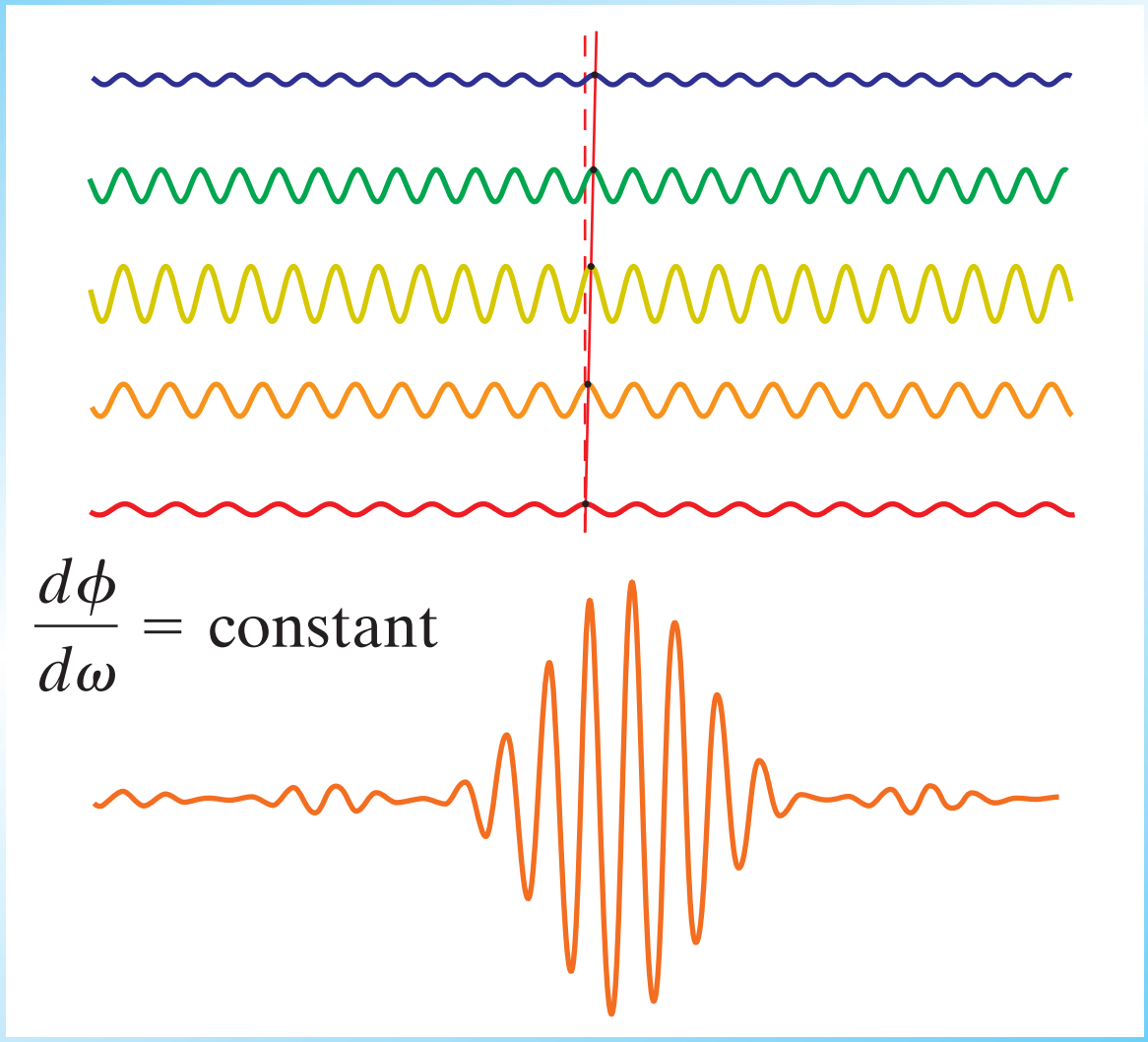

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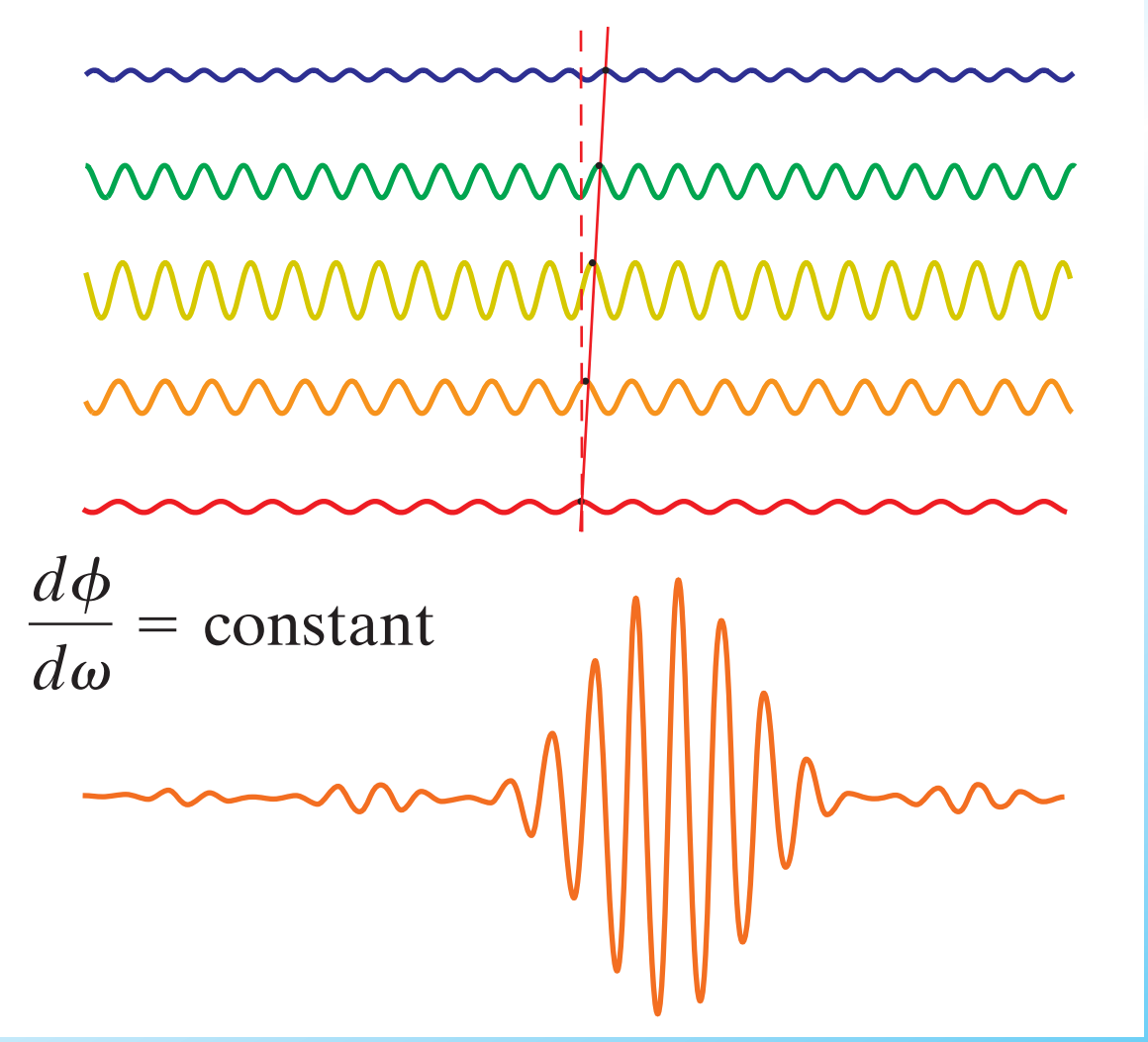

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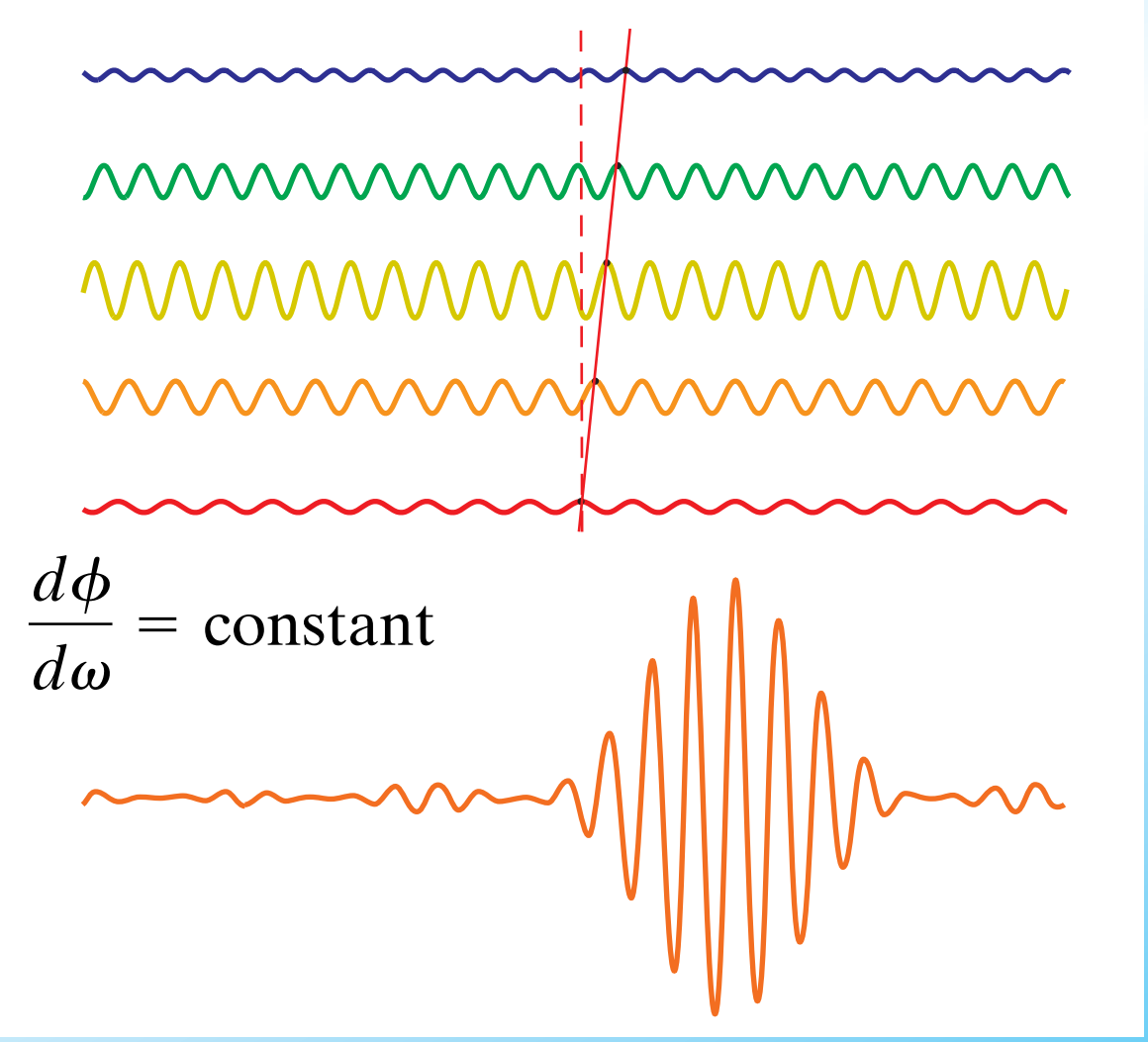

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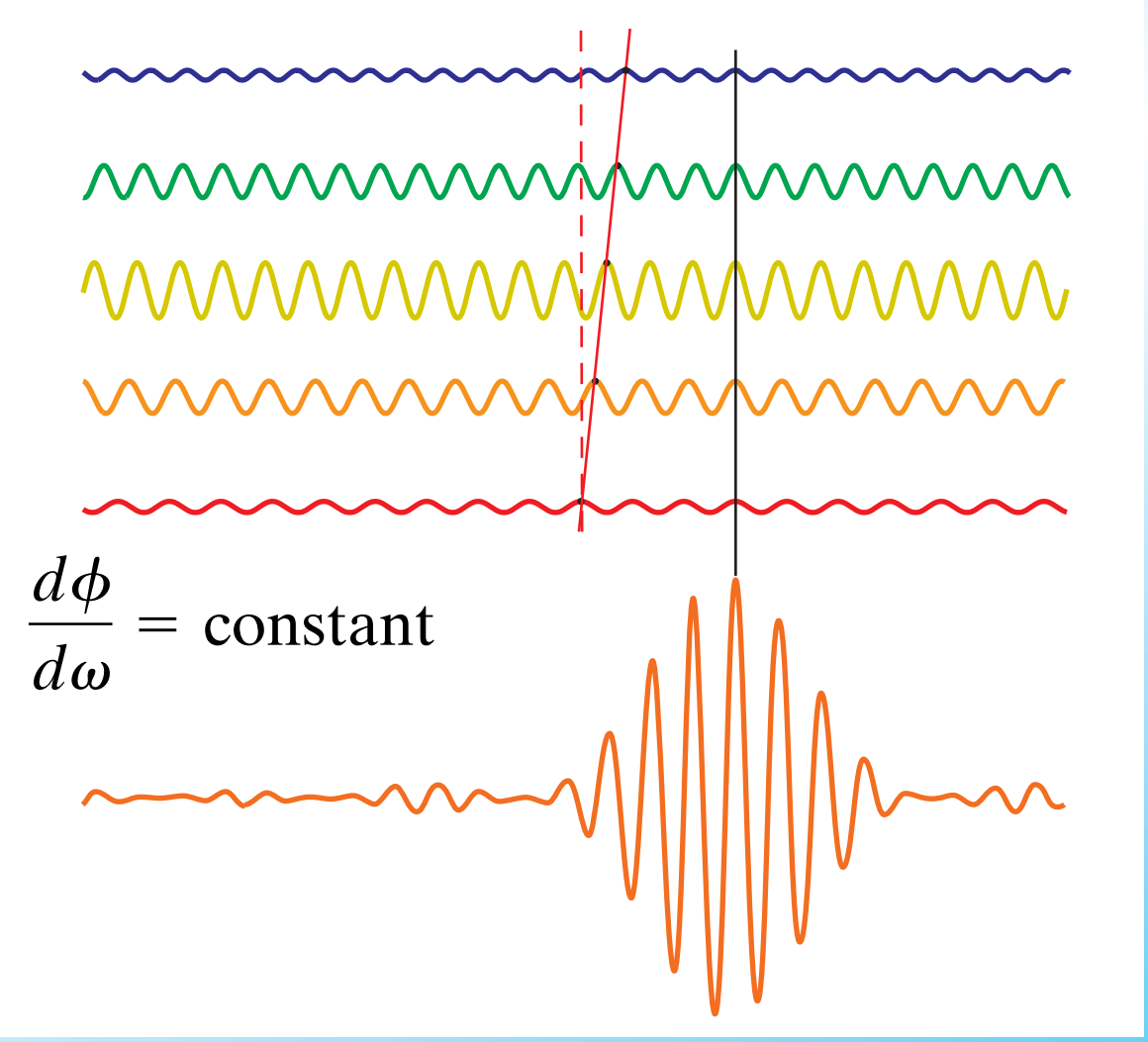

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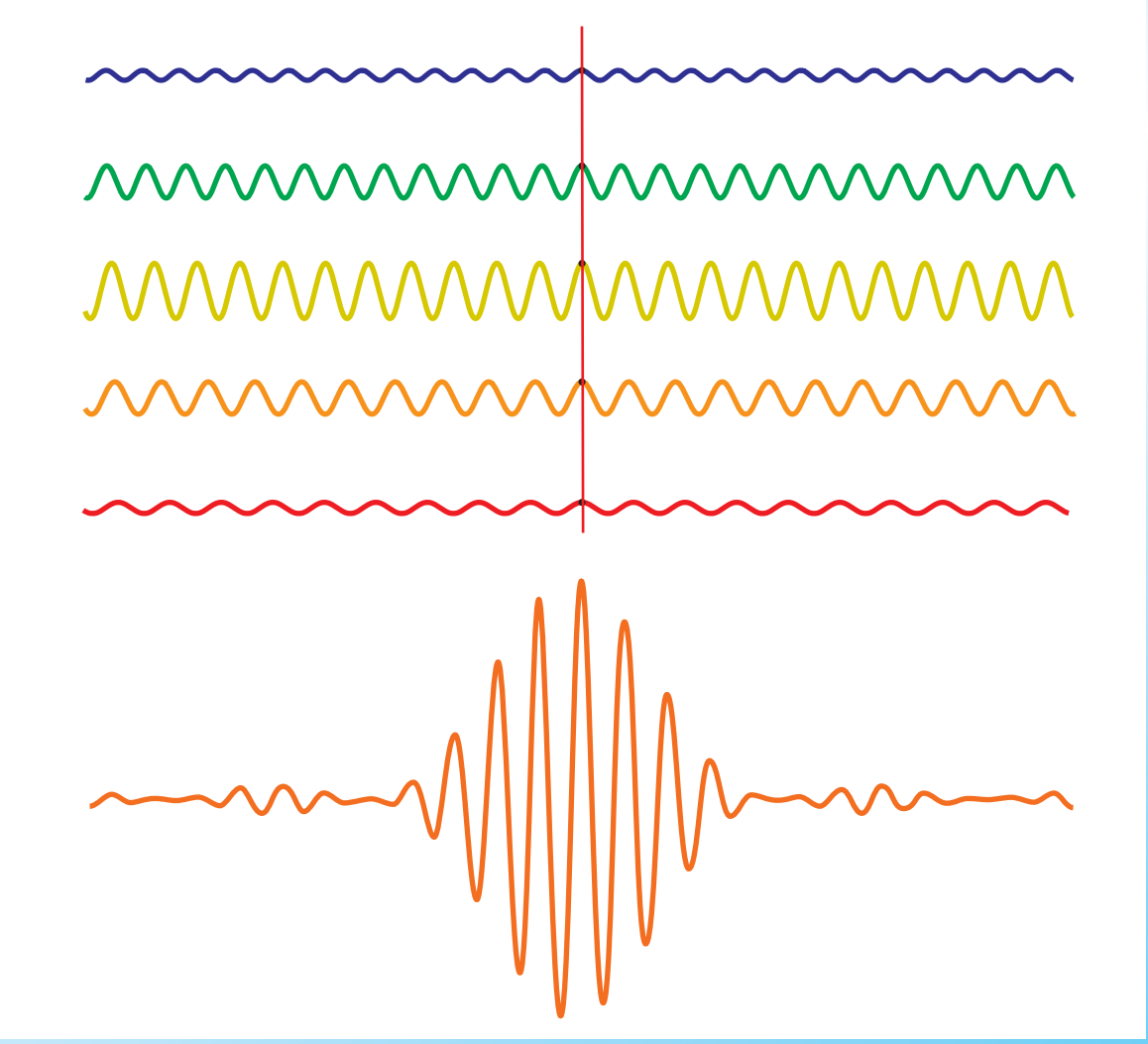

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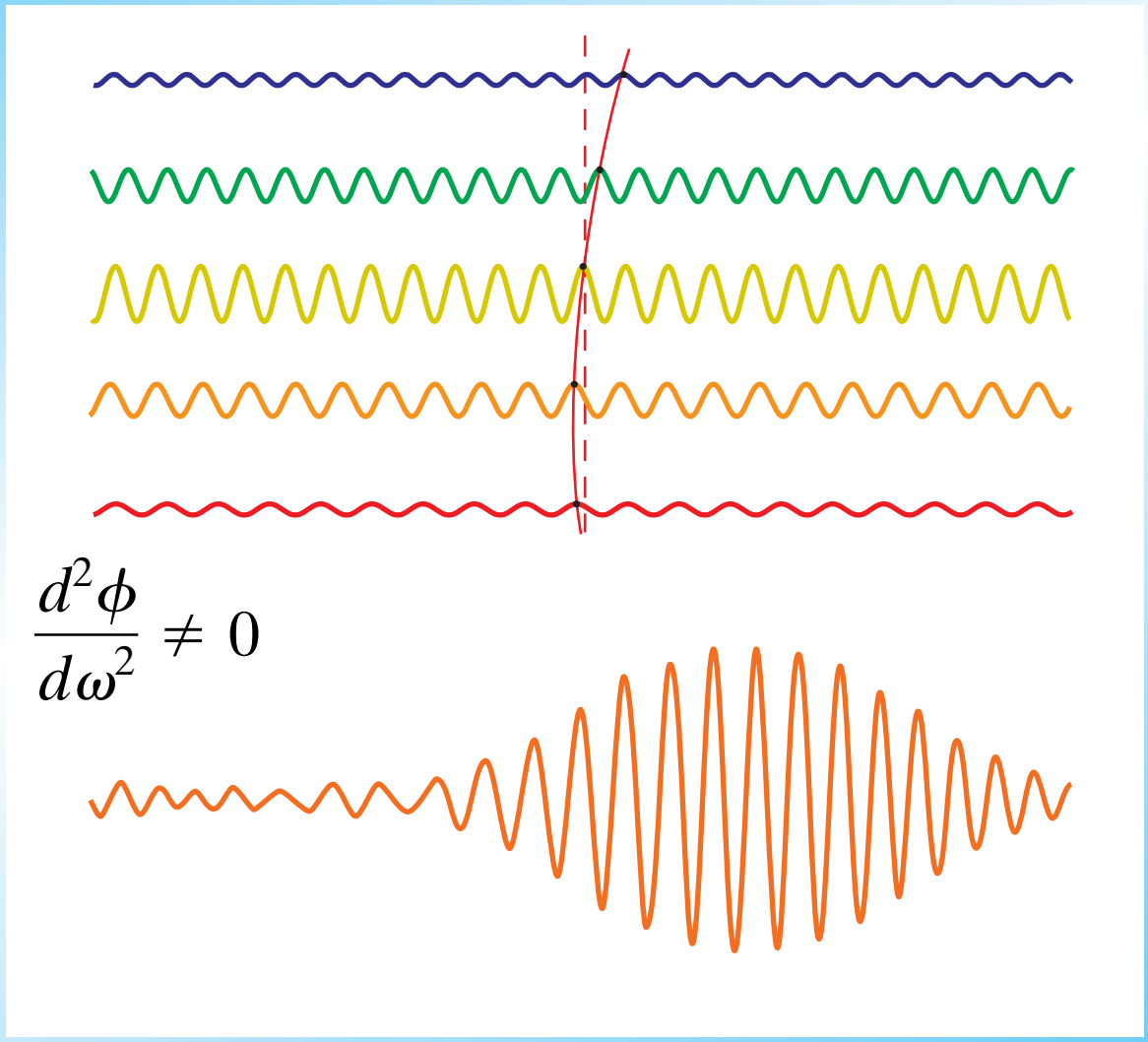
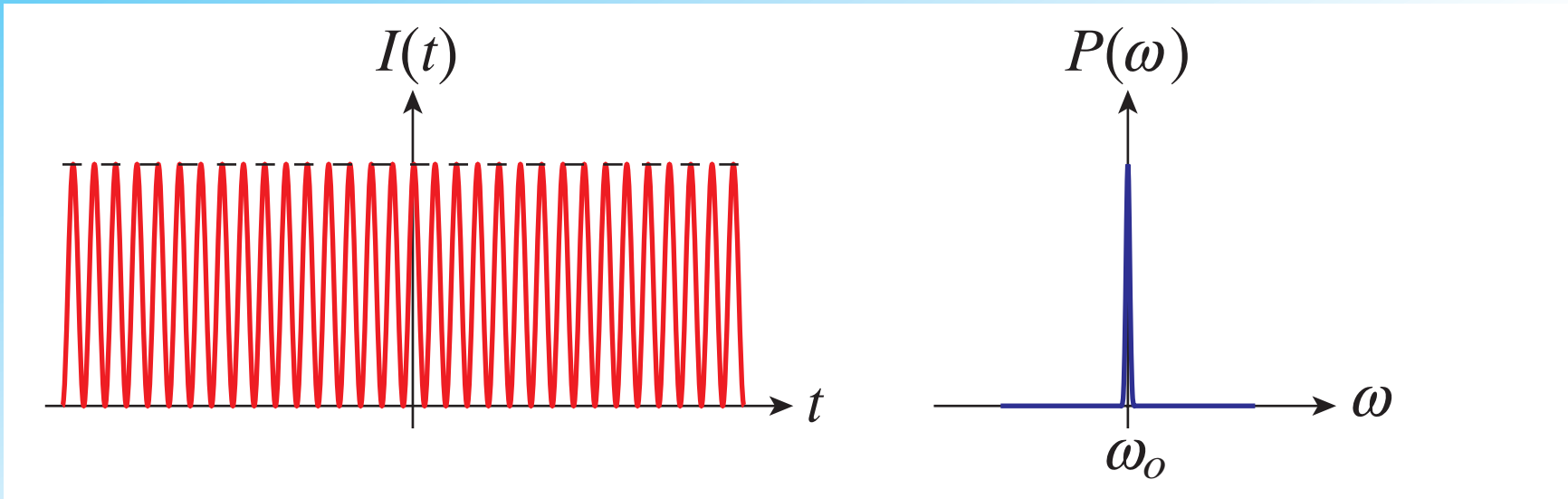


Dispersion compensation

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	+	–

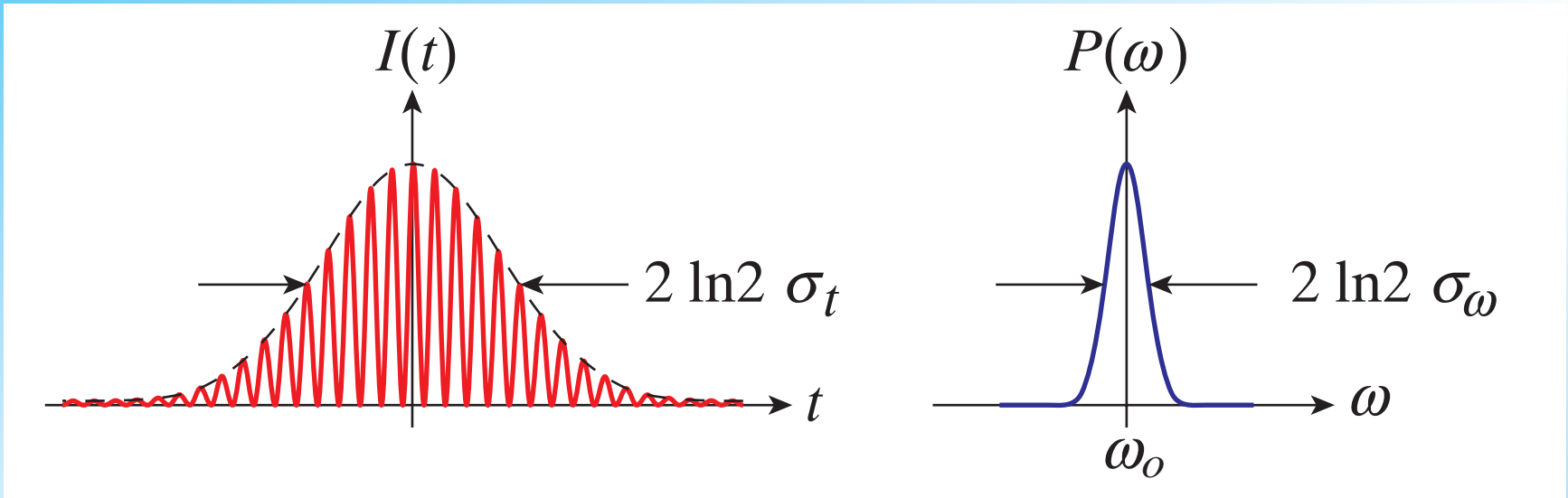
Representation of pulses



Spectrum of sinusoidal intensity is a delta function

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

Representation of pulses



Modulate amplitude

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

Representation of pulses

Fourier relations:

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

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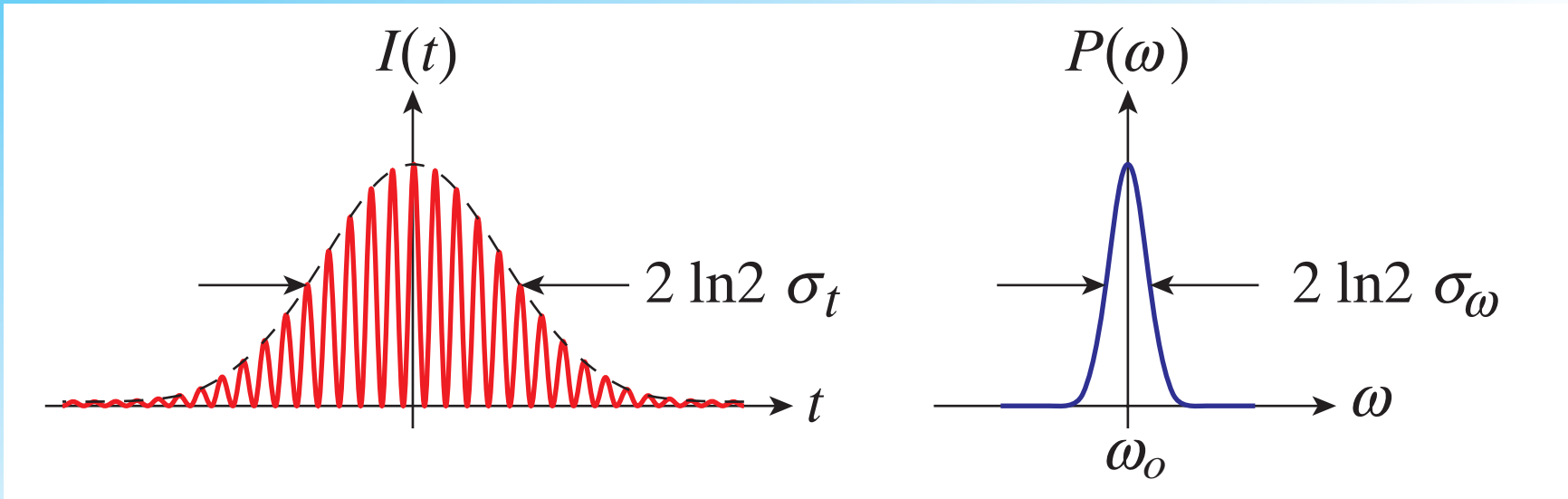
$$= \sigma_t \exp\left[-\frac{\sigma_t^2(\omega - \omega_o)^2}{2} \right] \equiv \sigma_t \exp\left[-\frac{(\omega - \omega_o)^2}{2\sigma_\omega^2} \right]$$

Representation of pulses



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

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

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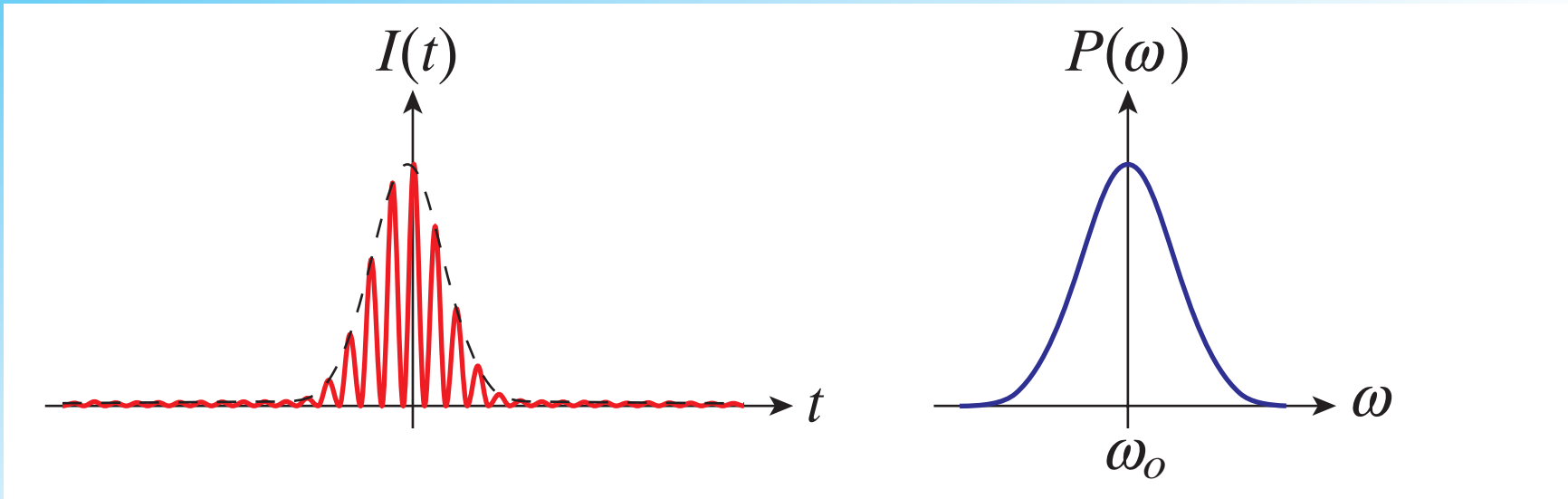

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Joint time-frequency representation

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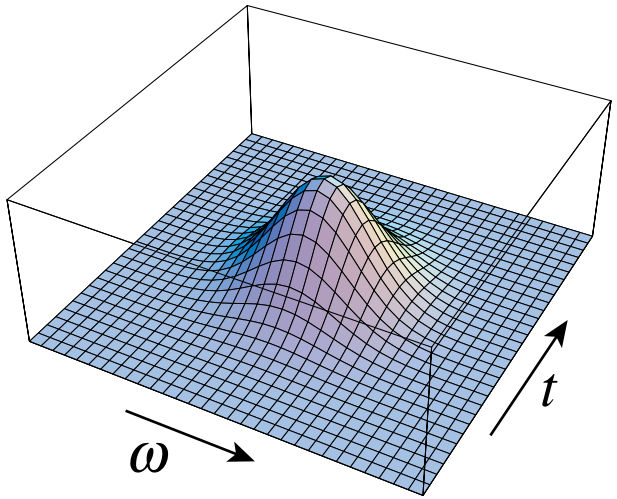
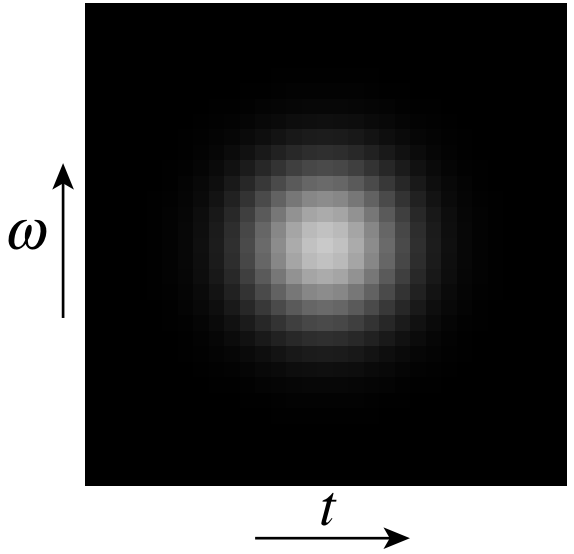
$$\int_{-\infty}^{\infty} W(t,\omega)\,dt = \left| E(\omega) \right|^2 = I(\omega)$$

Joint time-frequency representation

Energy:

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





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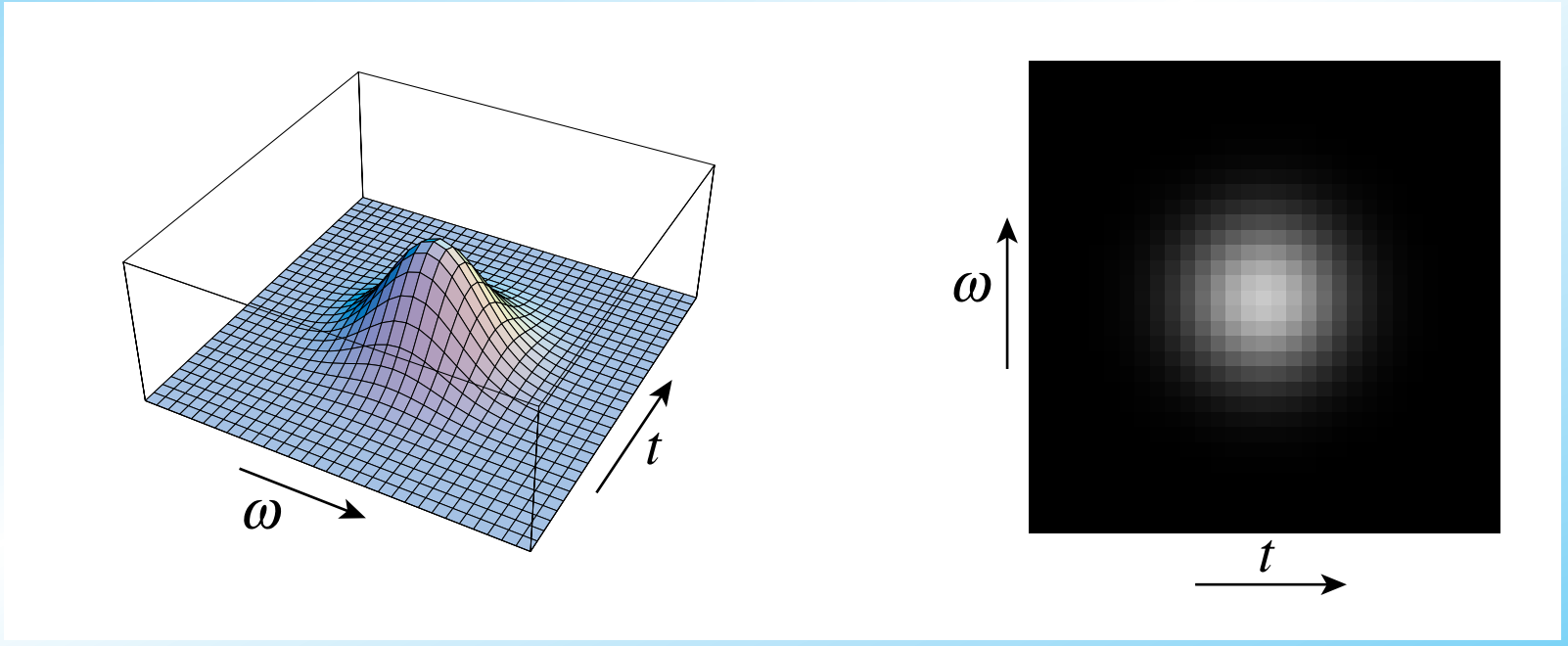

$W(t,\omega)$ **must be nonzero in phase-space area larger than** π

Joint time-frequency representation

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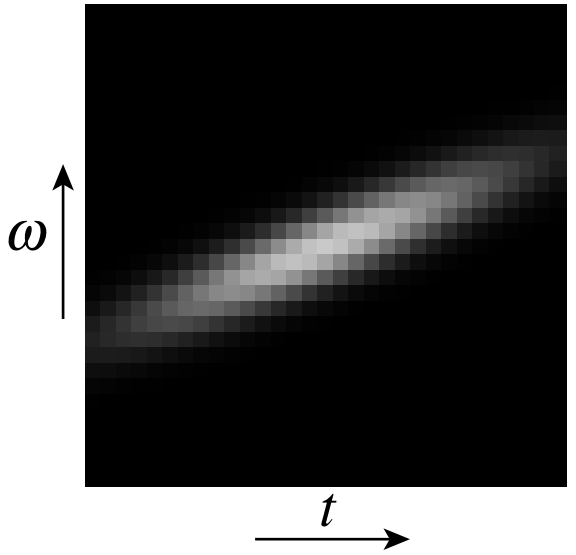
chirped pulse



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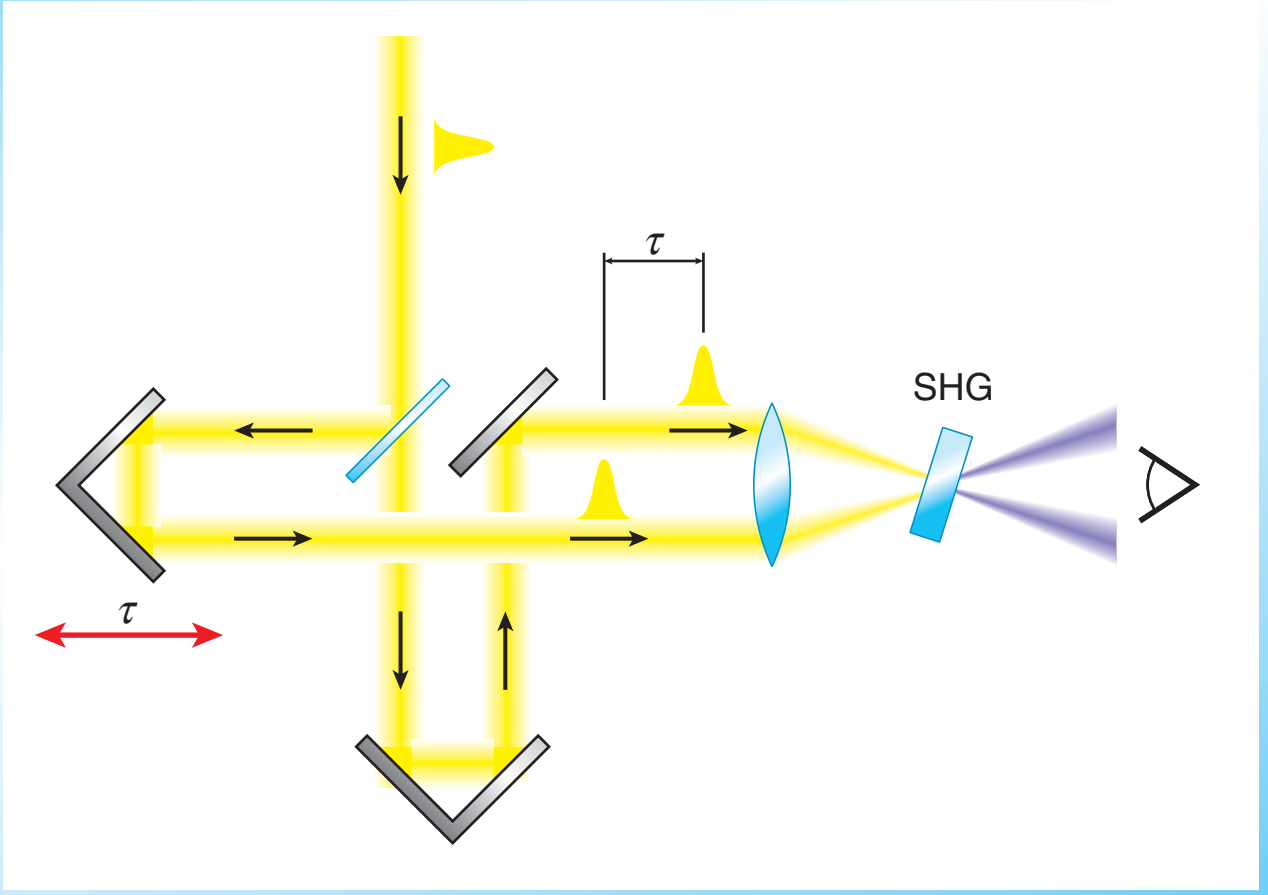
Temporal characterization

Use pulse to measure itself…



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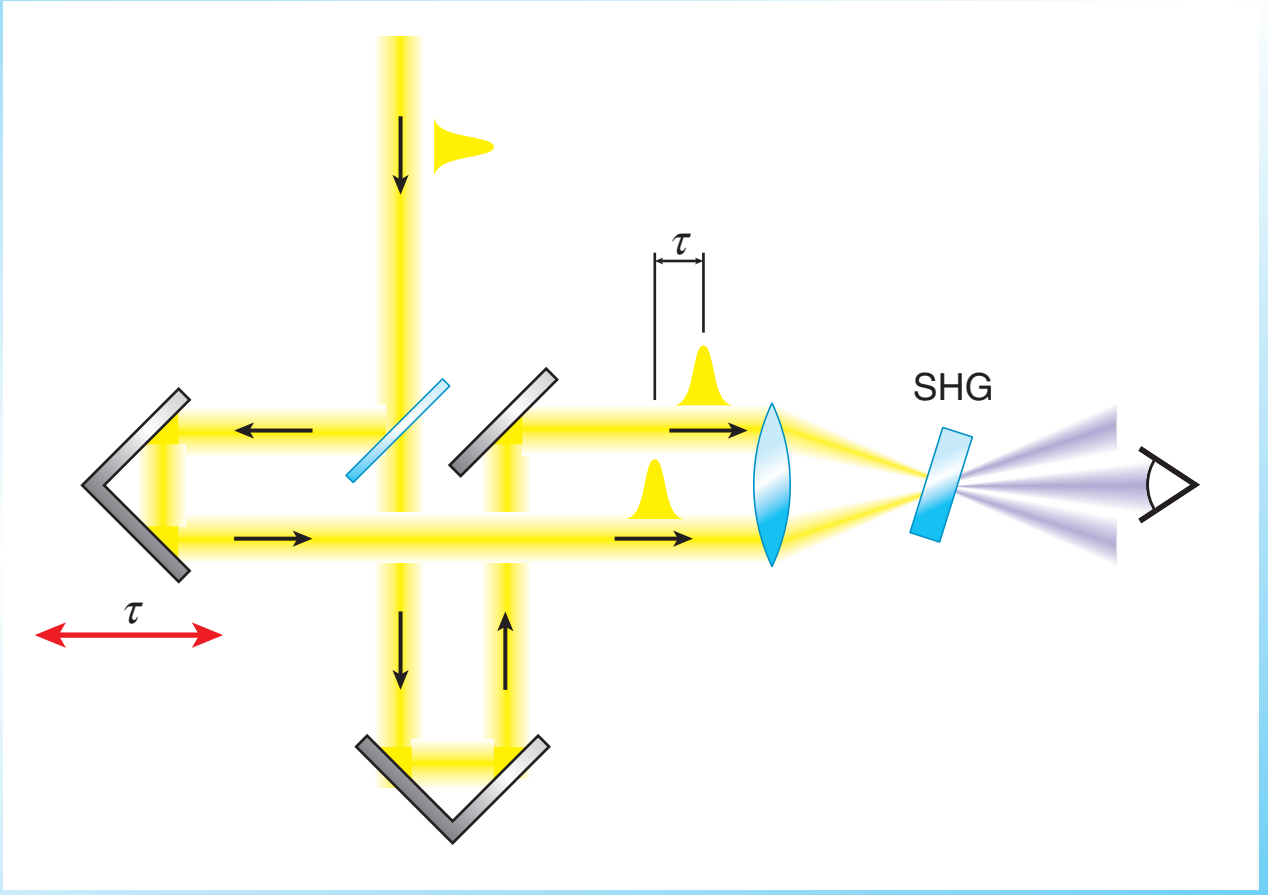

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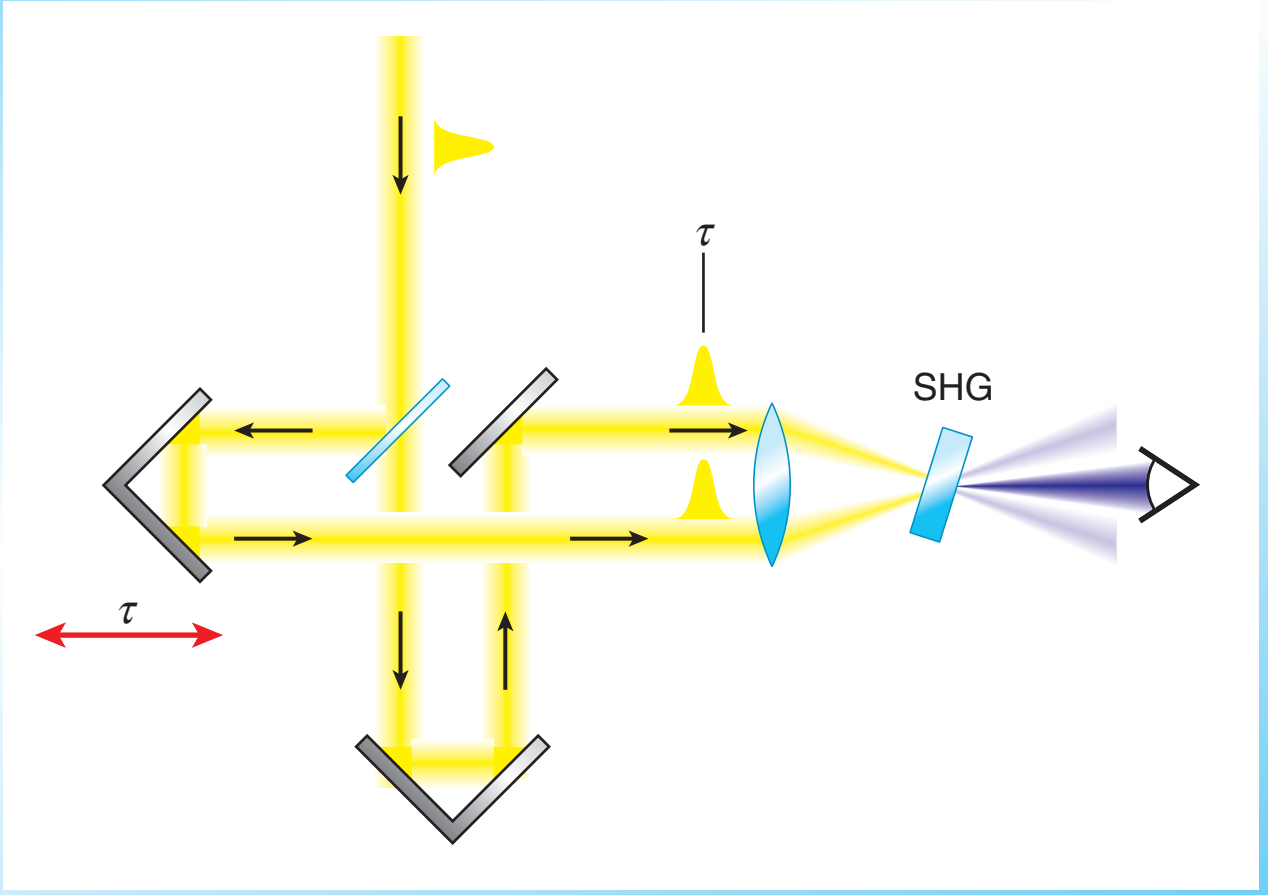

Temporal characterization

Electric field at SHG crystal

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

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Second harmonic field

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

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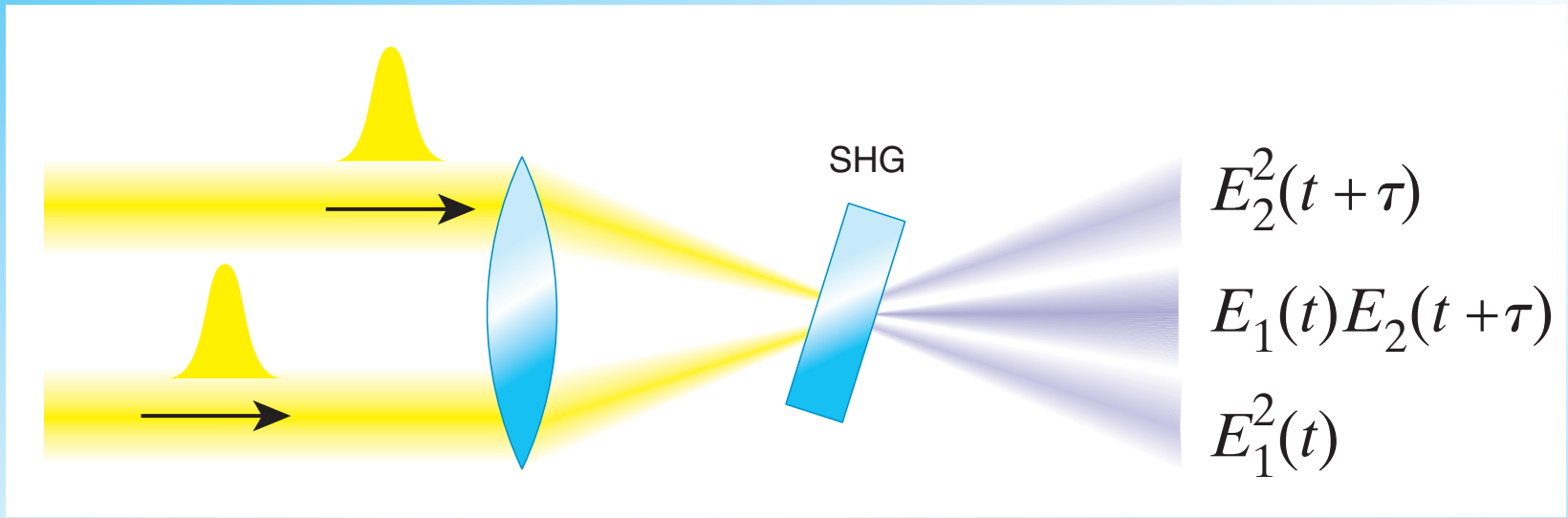
Second harmonic intensity

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

Temporal characterization



Second harmonic intensity

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detector selects middle term

Temporal characterization

Integrated detector signal yields intensity autocorrelation

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

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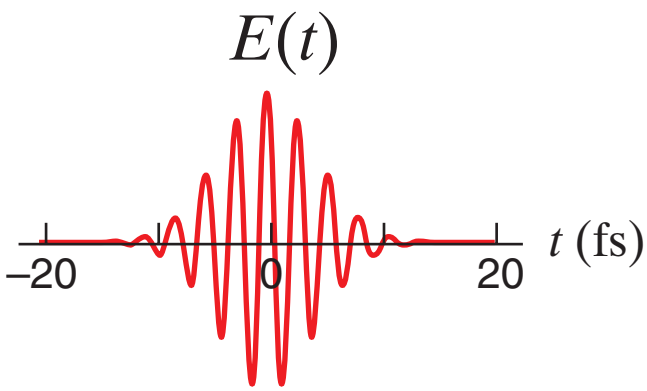

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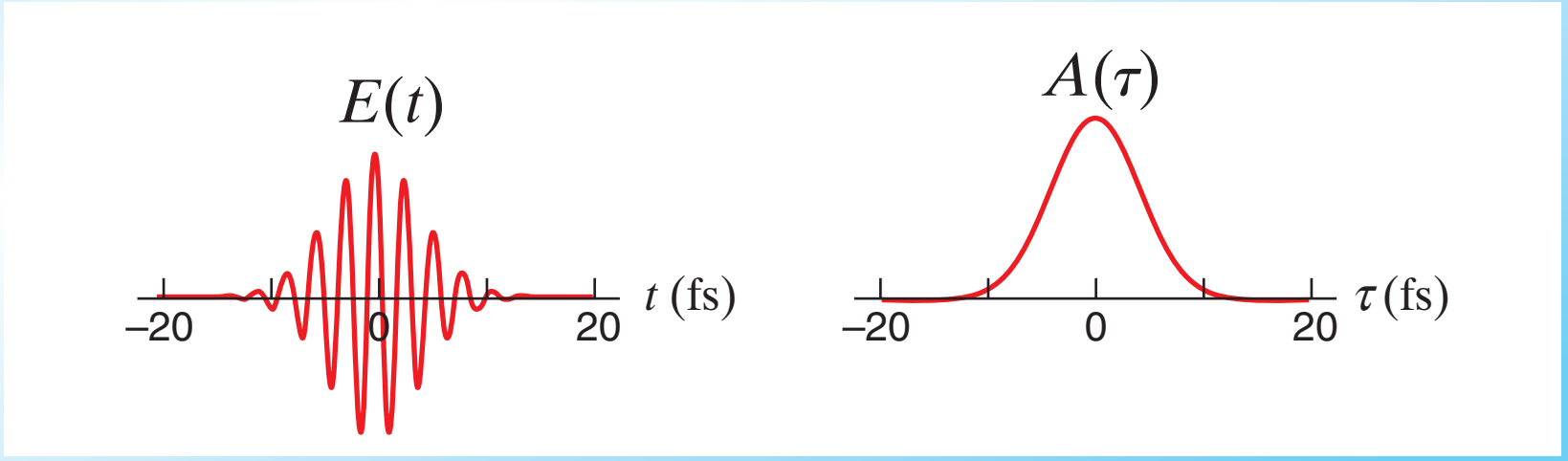
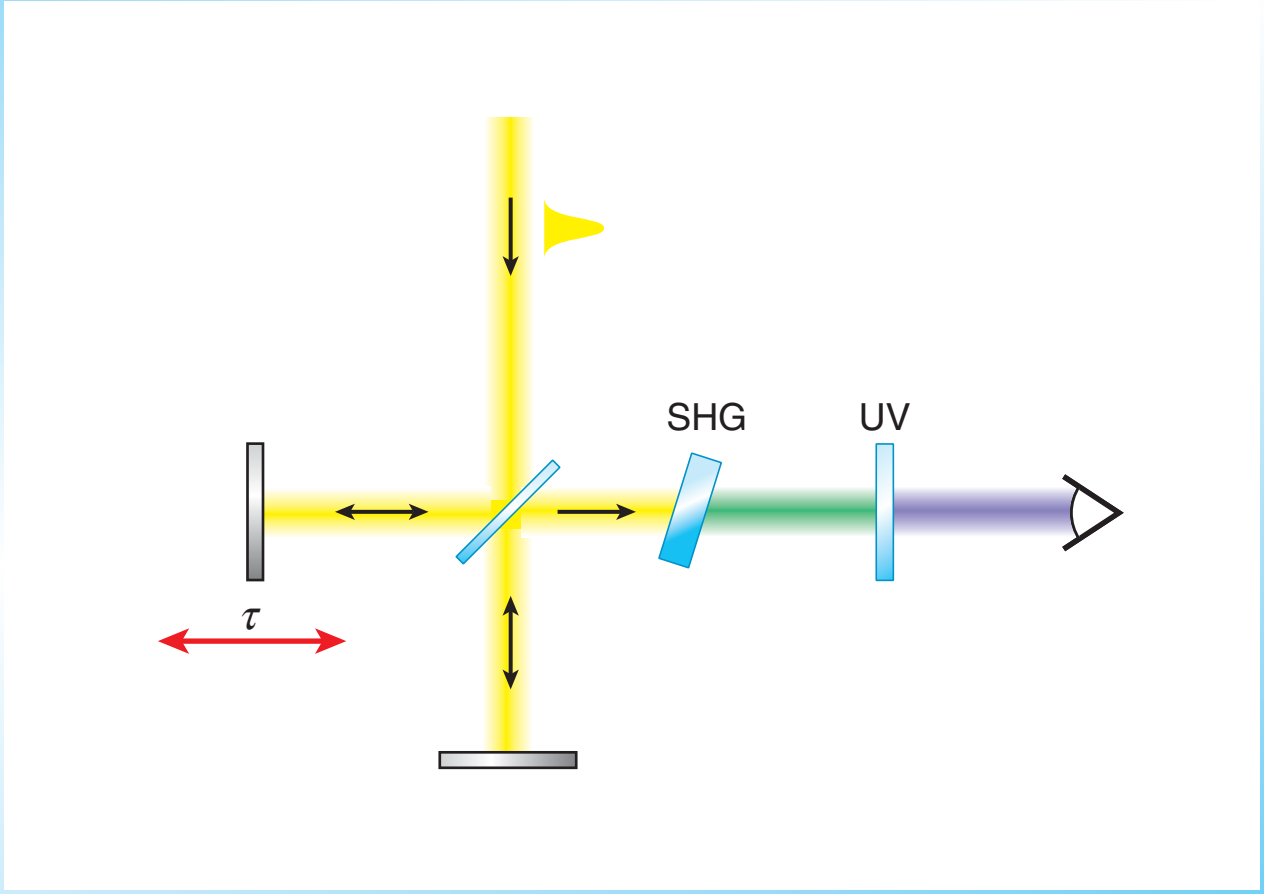


Temporal characterization

Alternative colinear geometry



Temporal characterization

All terms now contribute:

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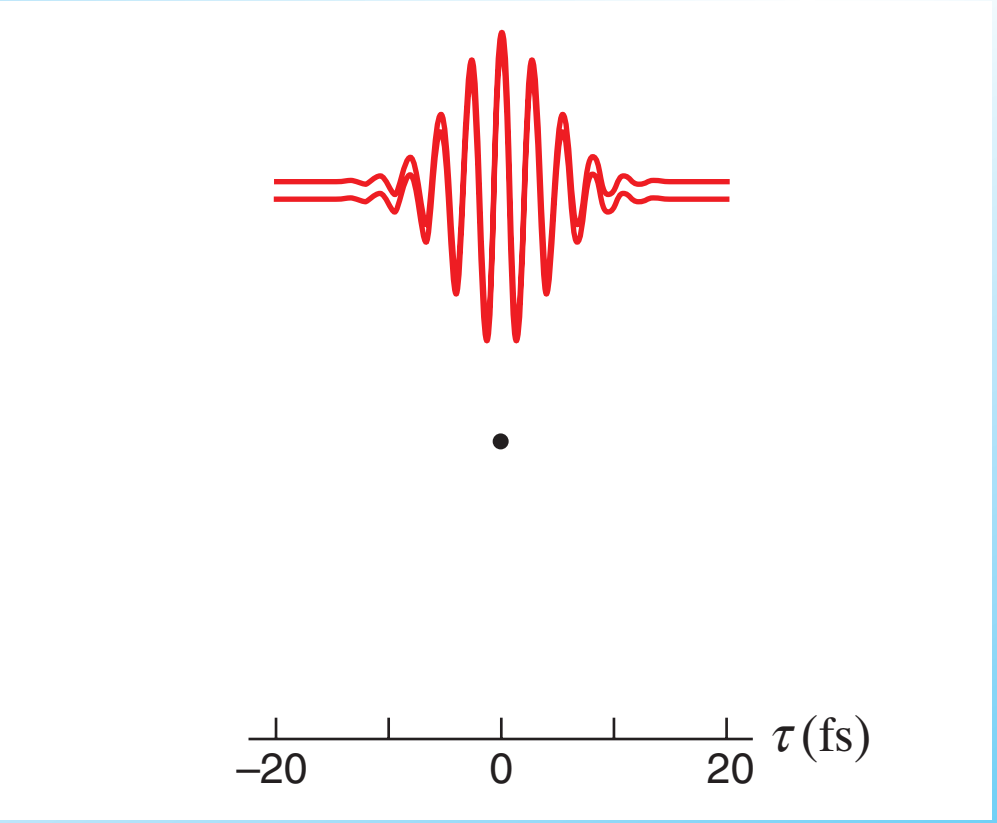

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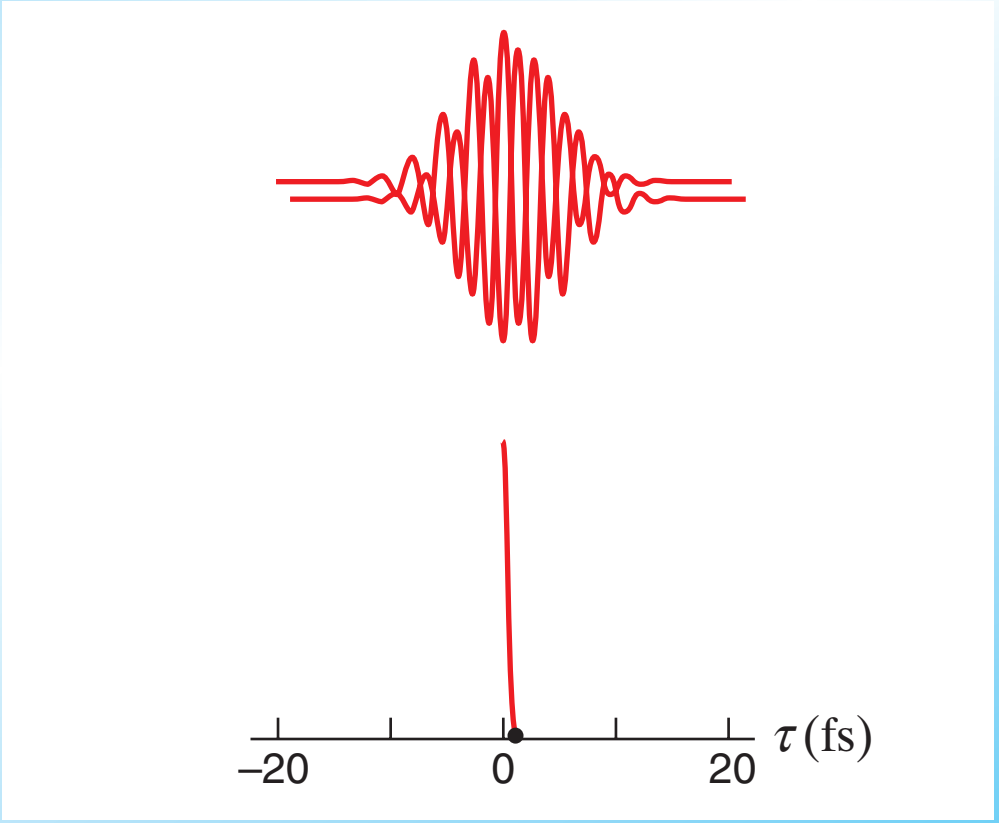

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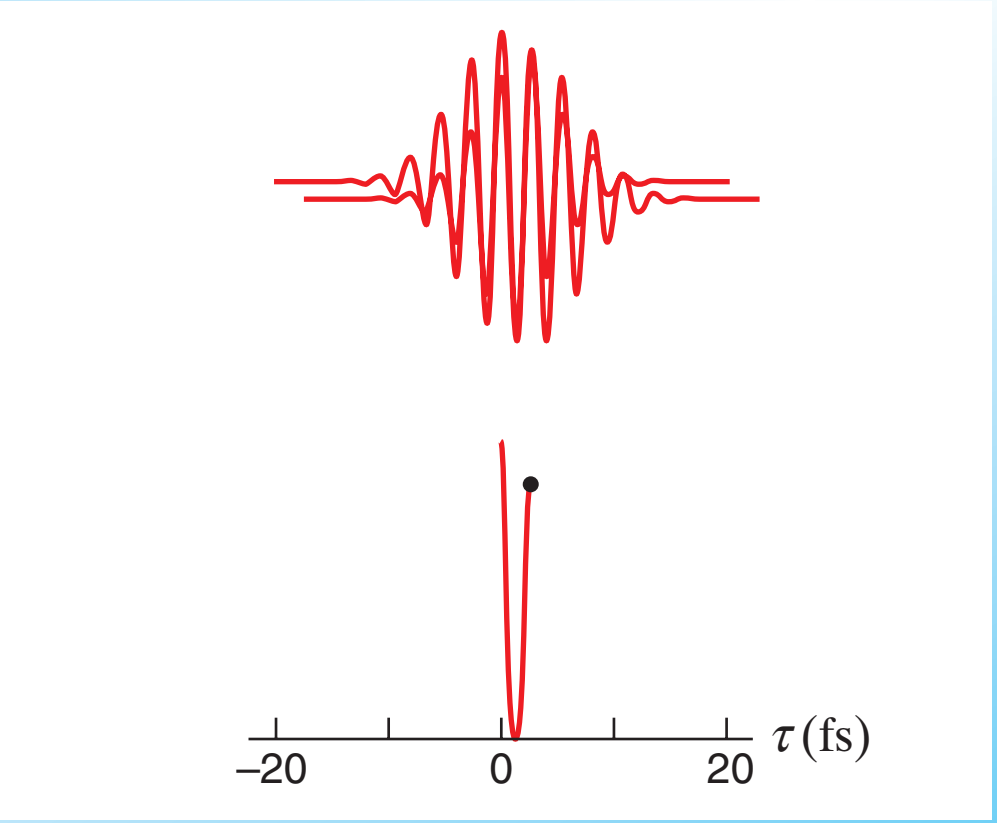

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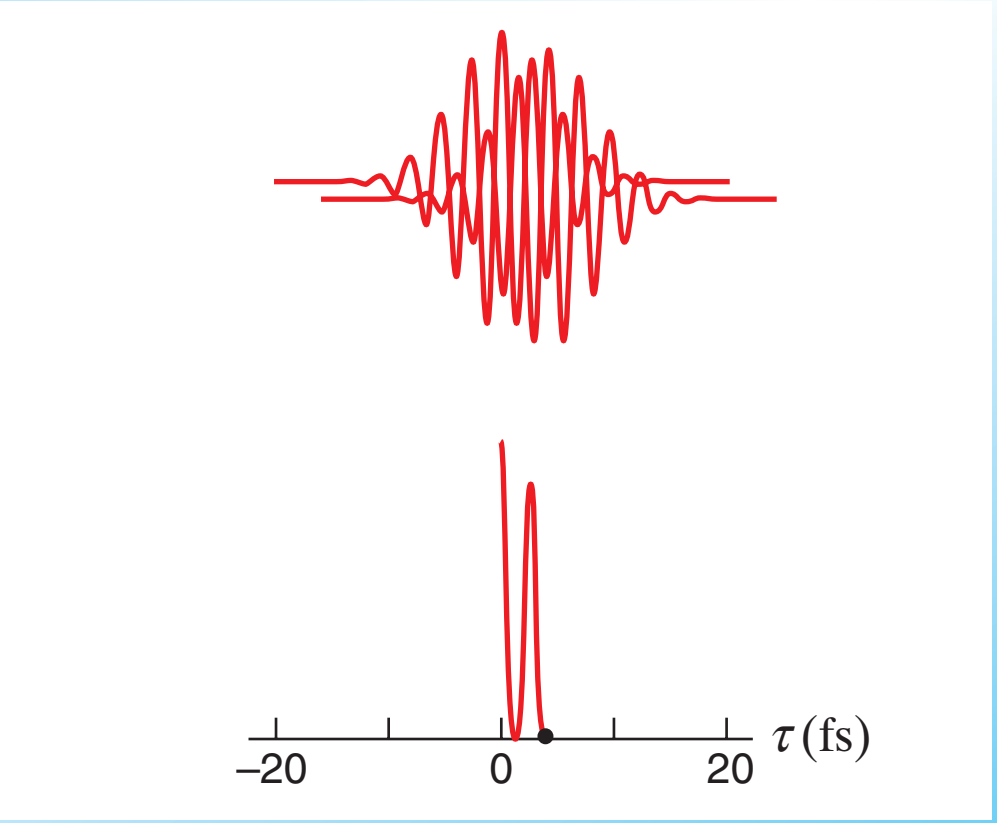

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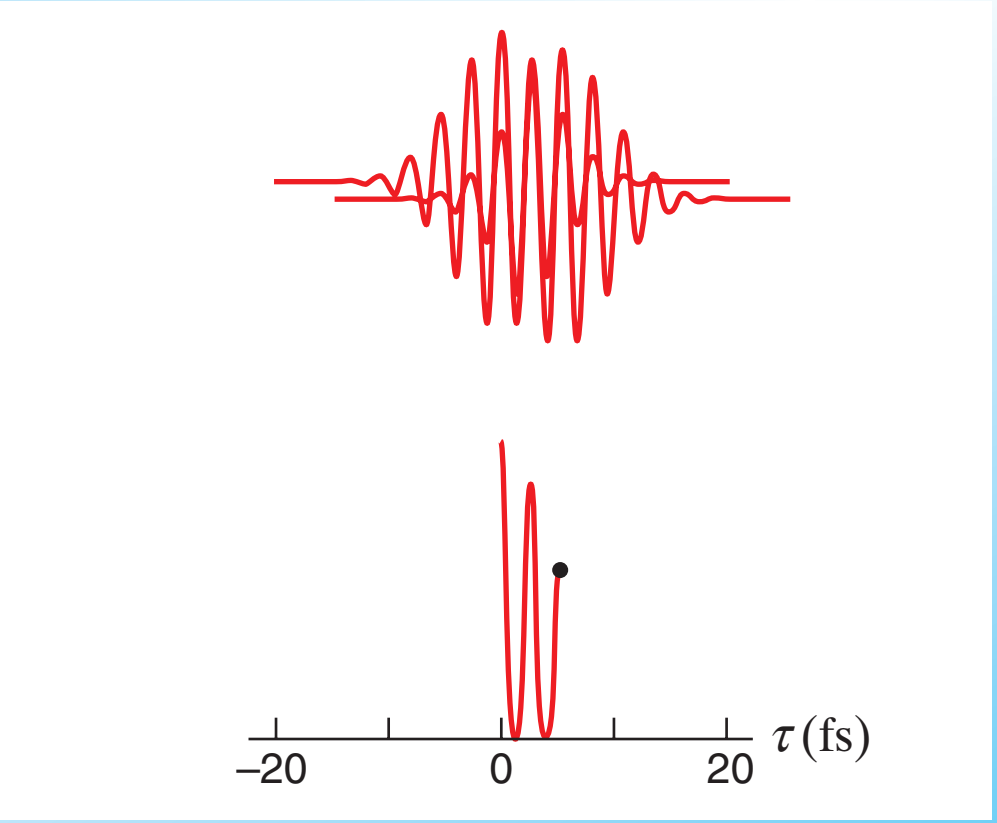

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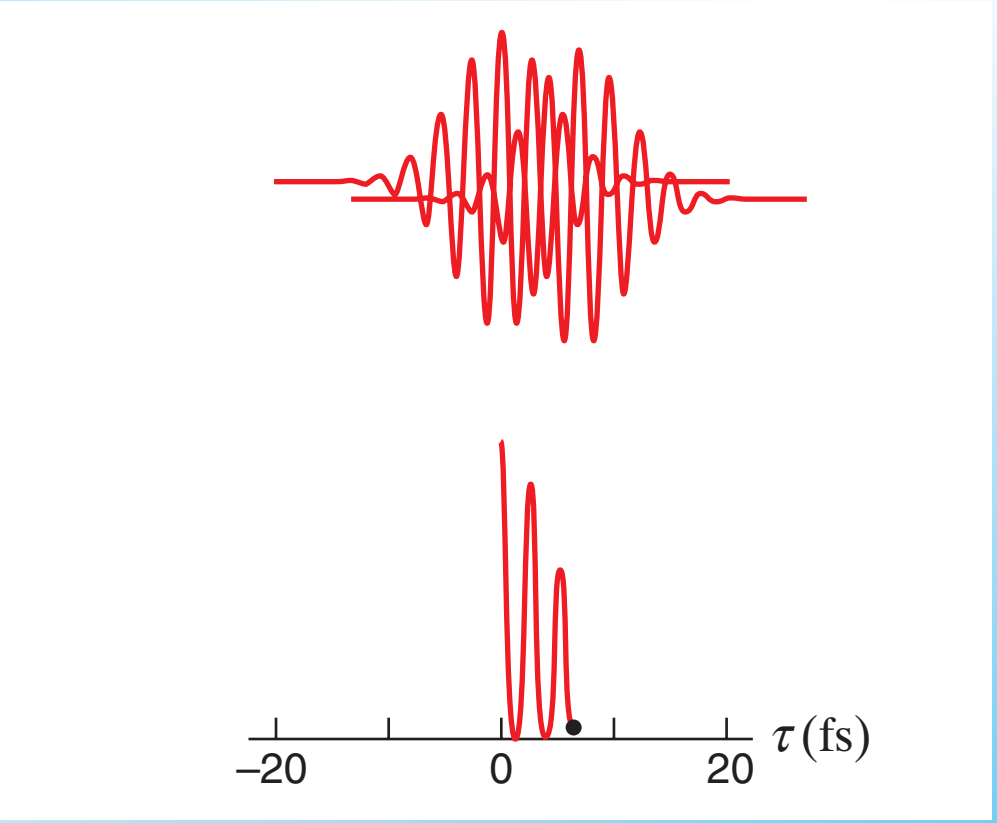

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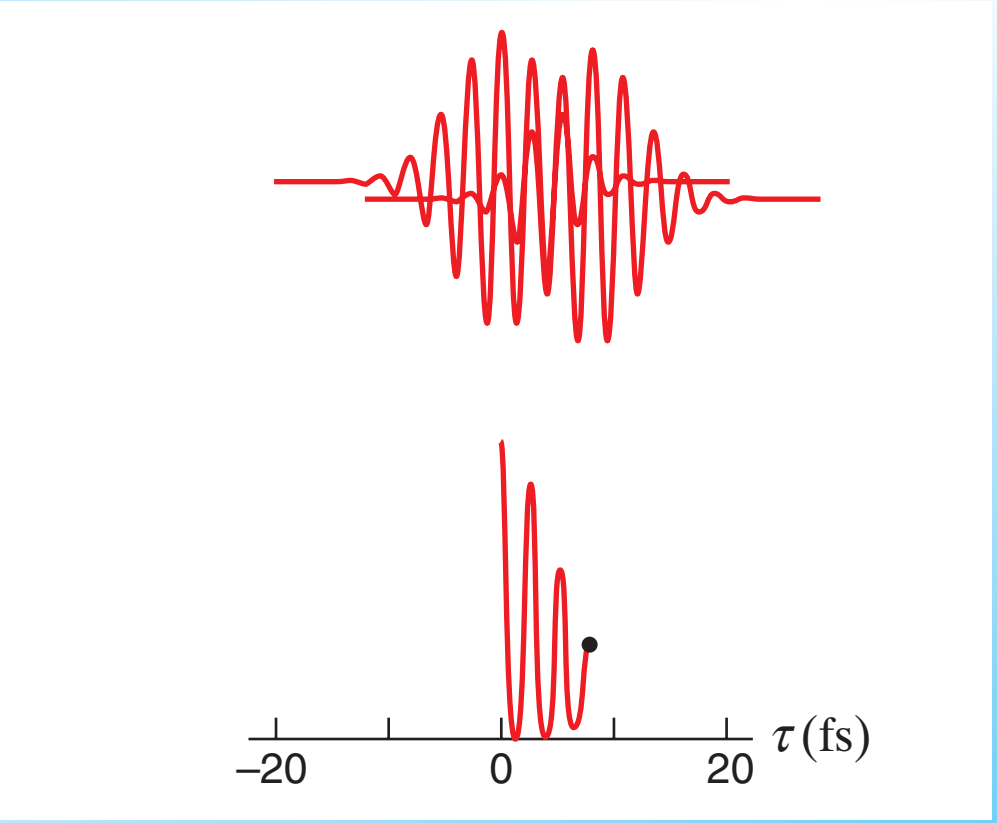

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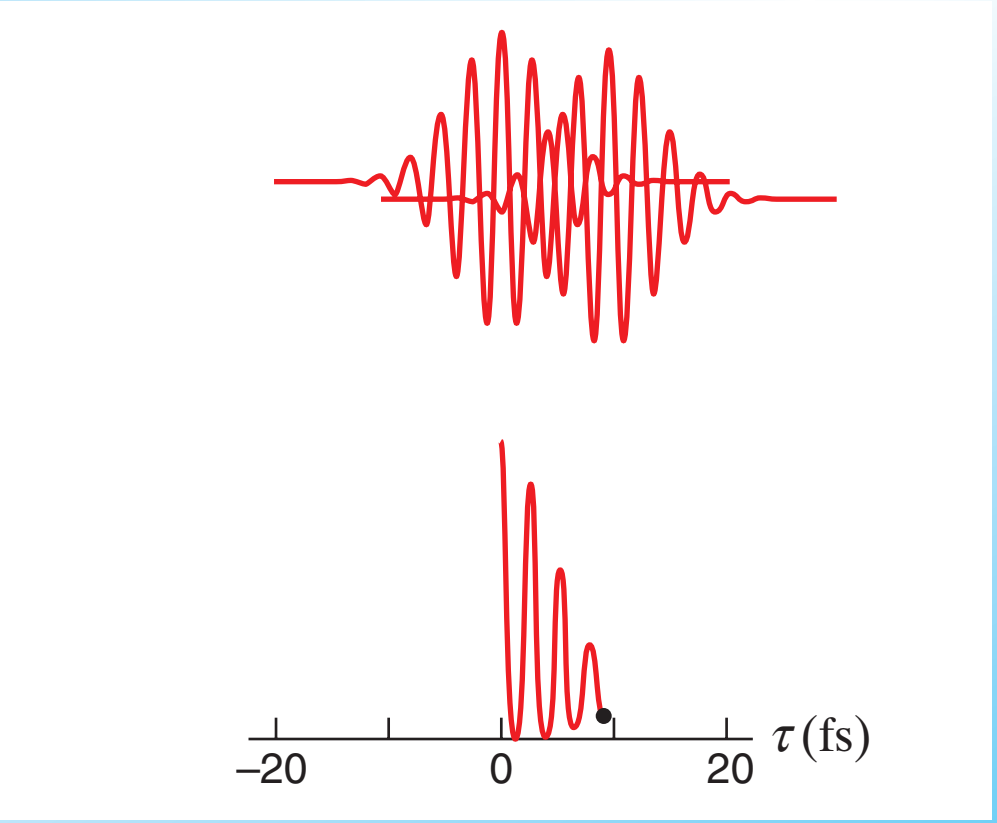

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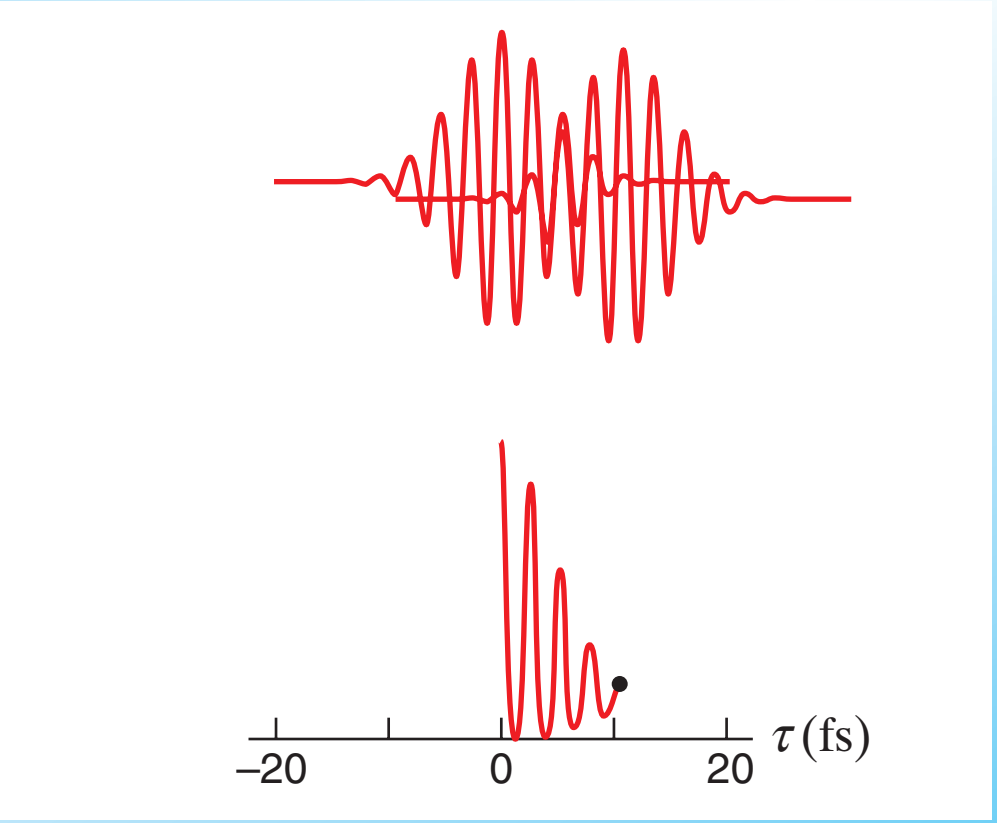

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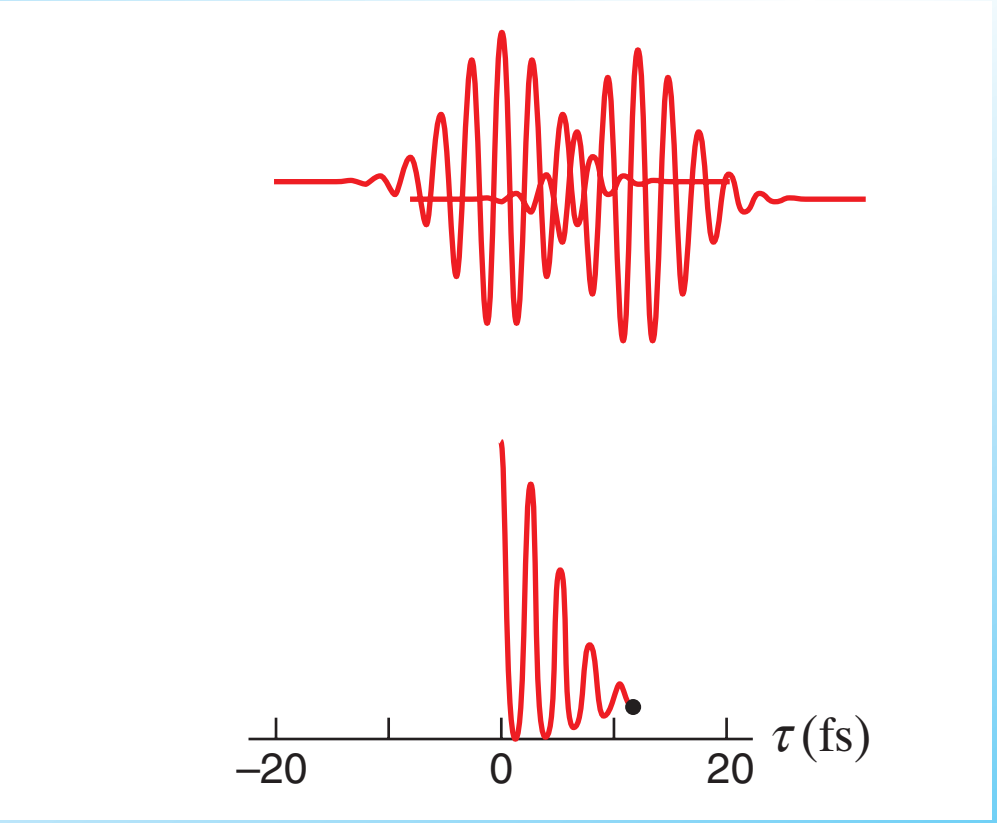

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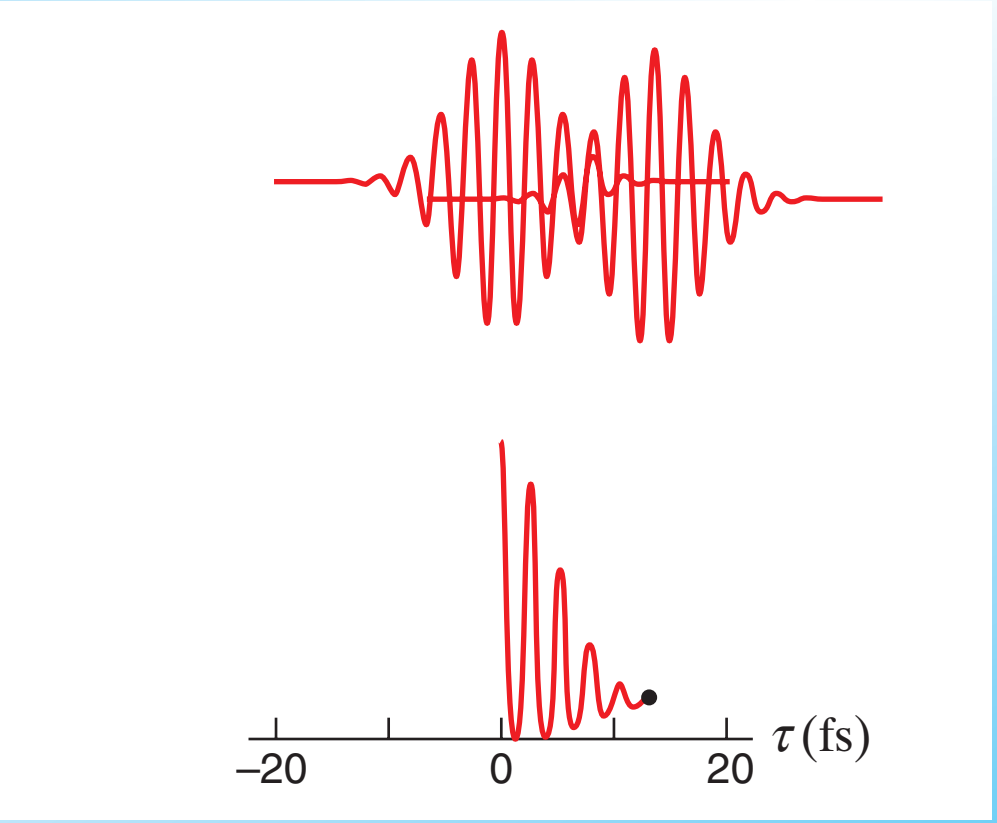

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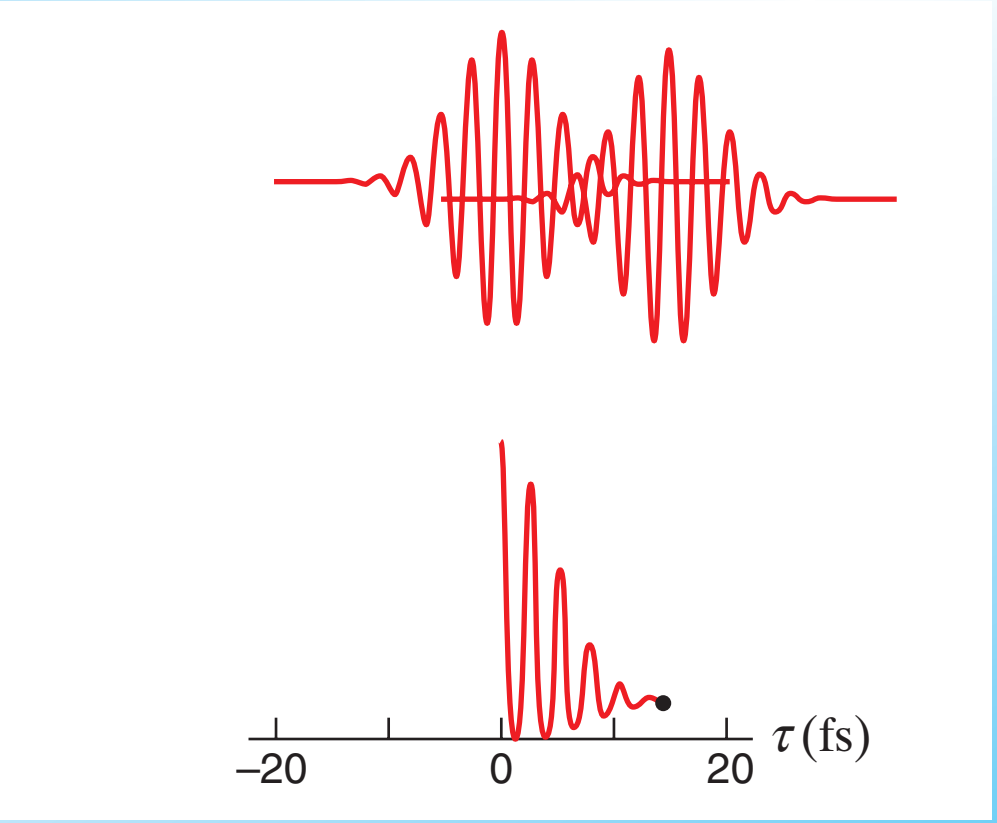

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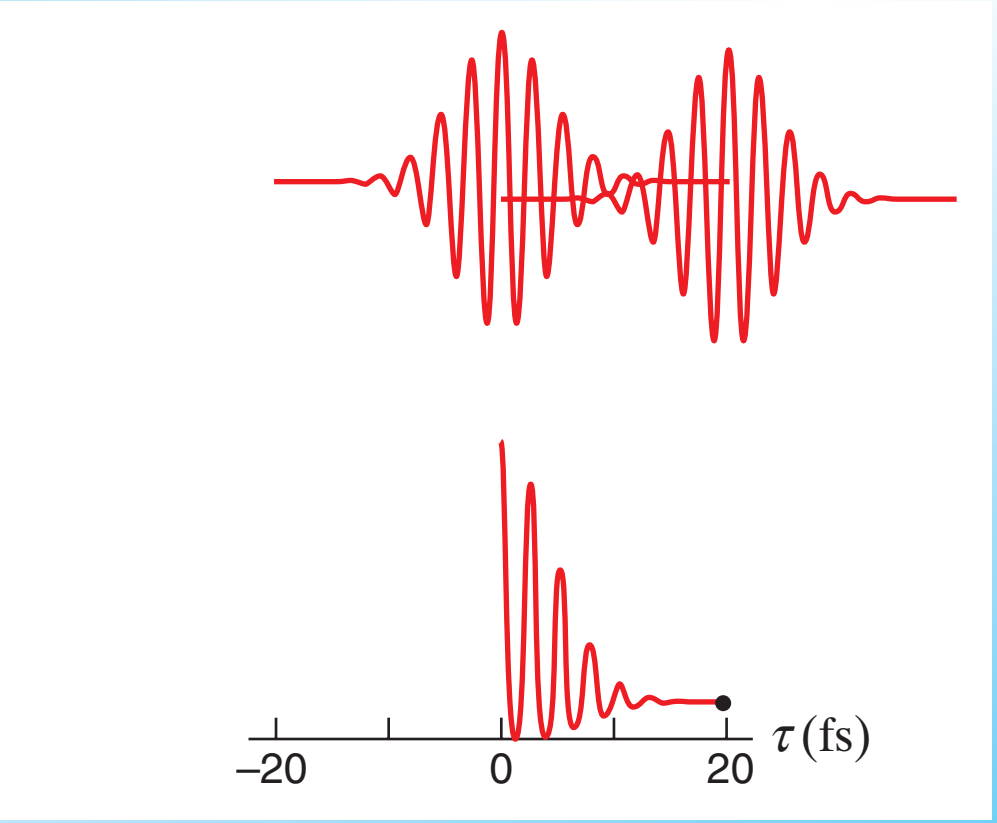

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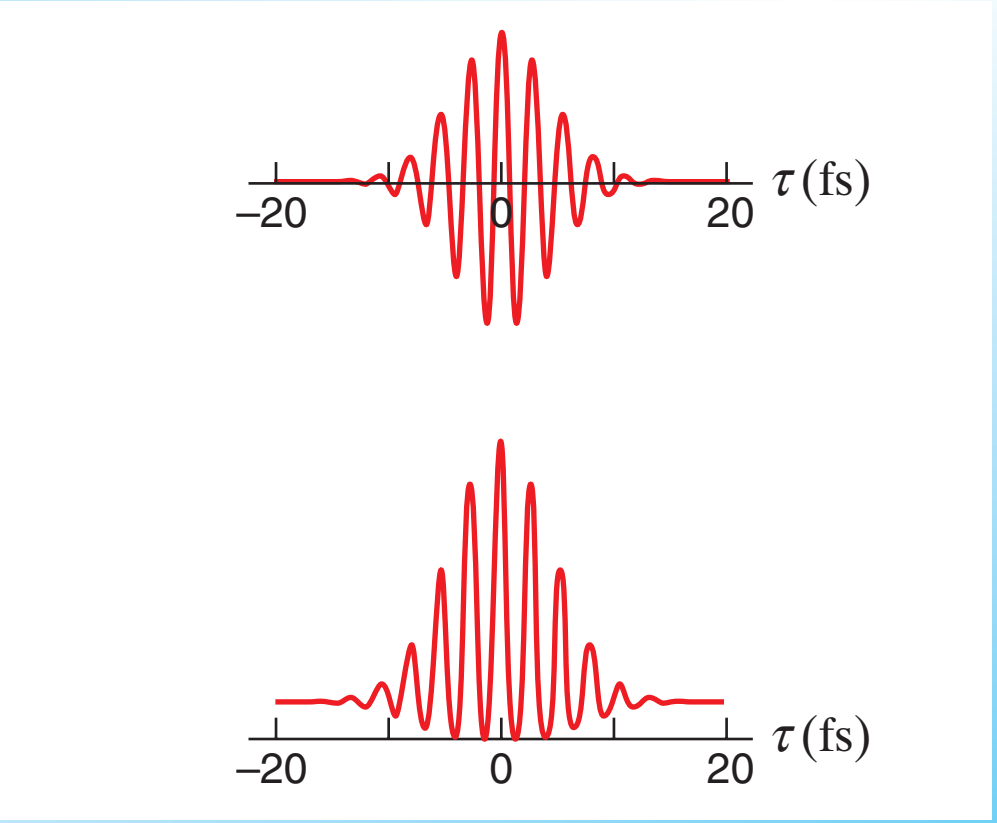

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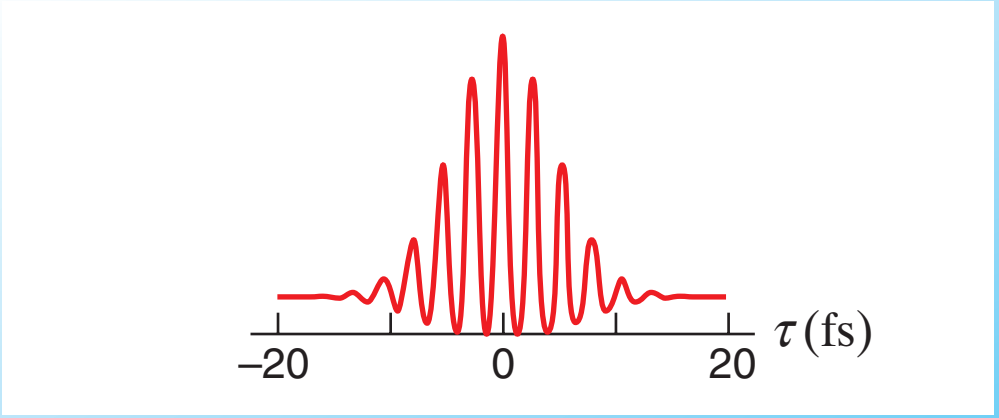
at $\tau = 0$**:**

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



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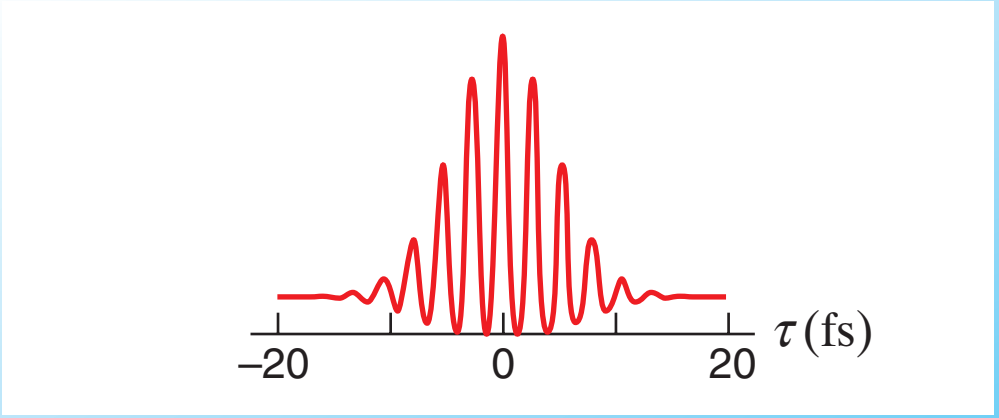

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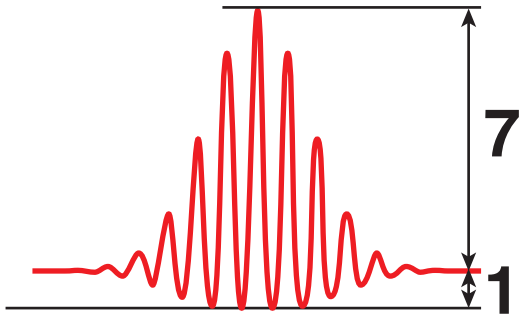
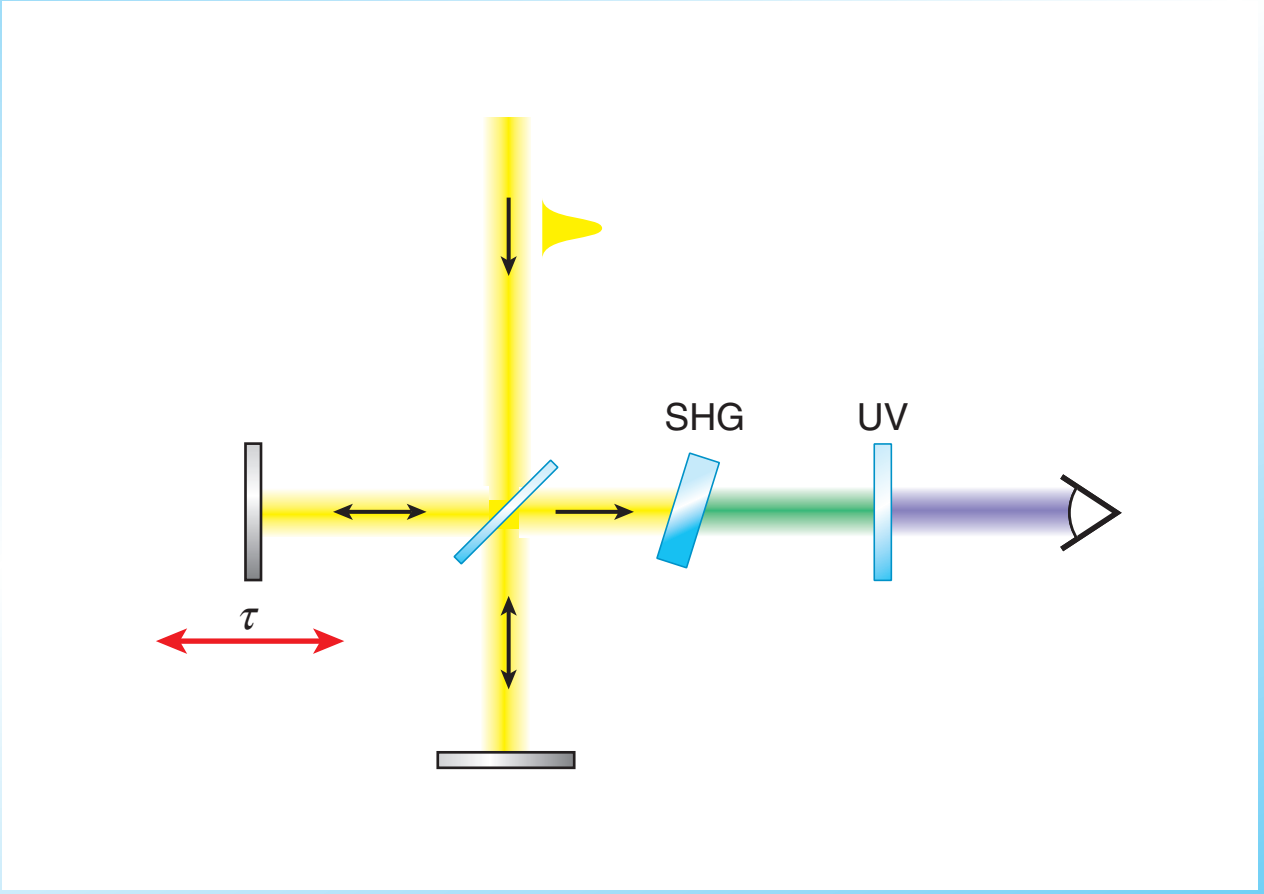


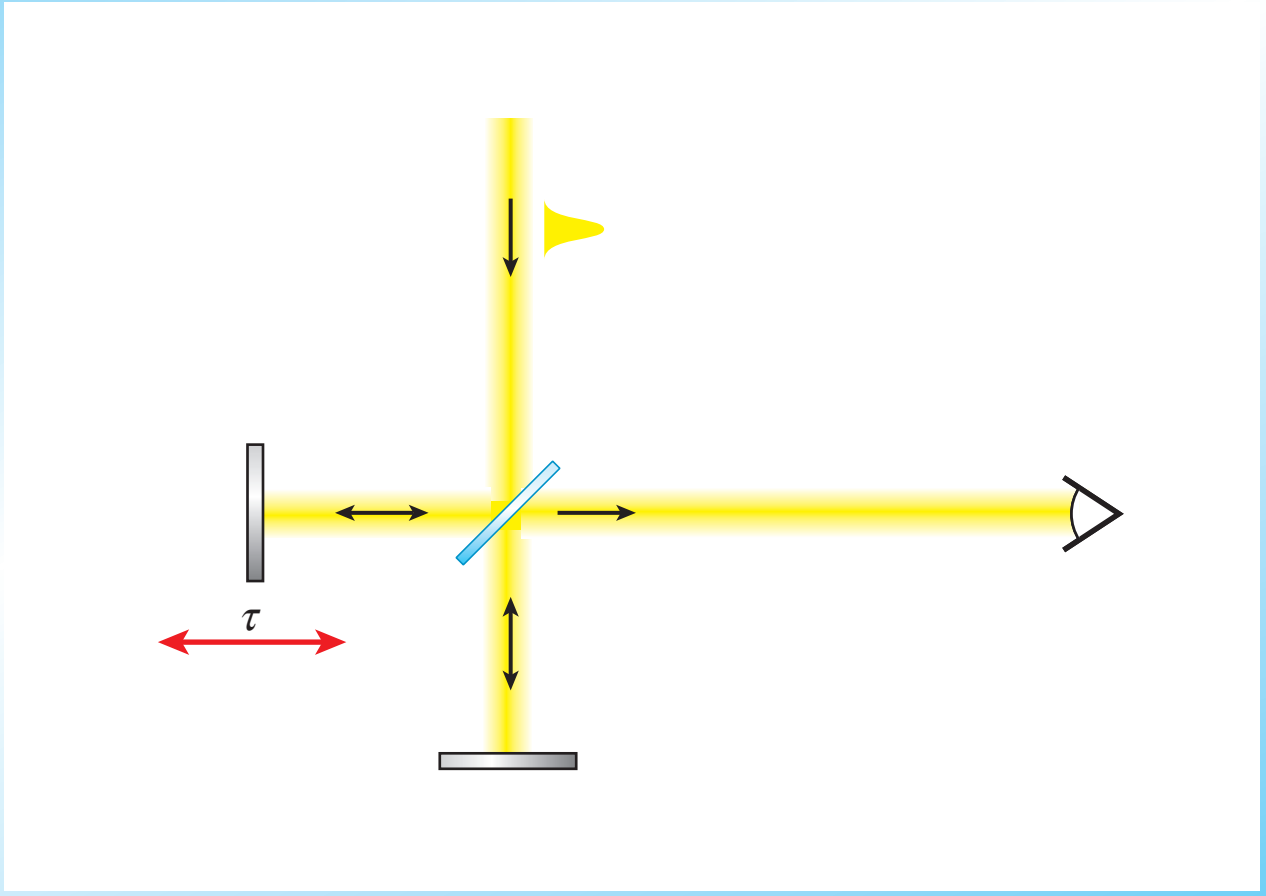
Temporal characterization

Do we really need the second-harmonic crystal…?



Temporal characterization

Would this work?



Temporal characterization

Intensity at detector

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

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Temporal characterization

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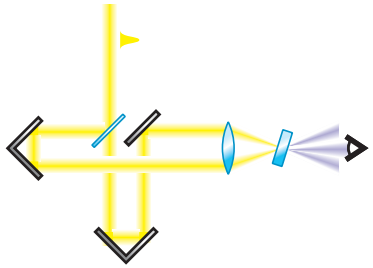
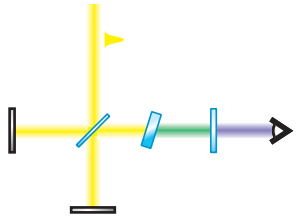
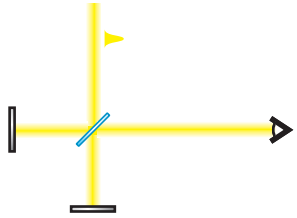
so

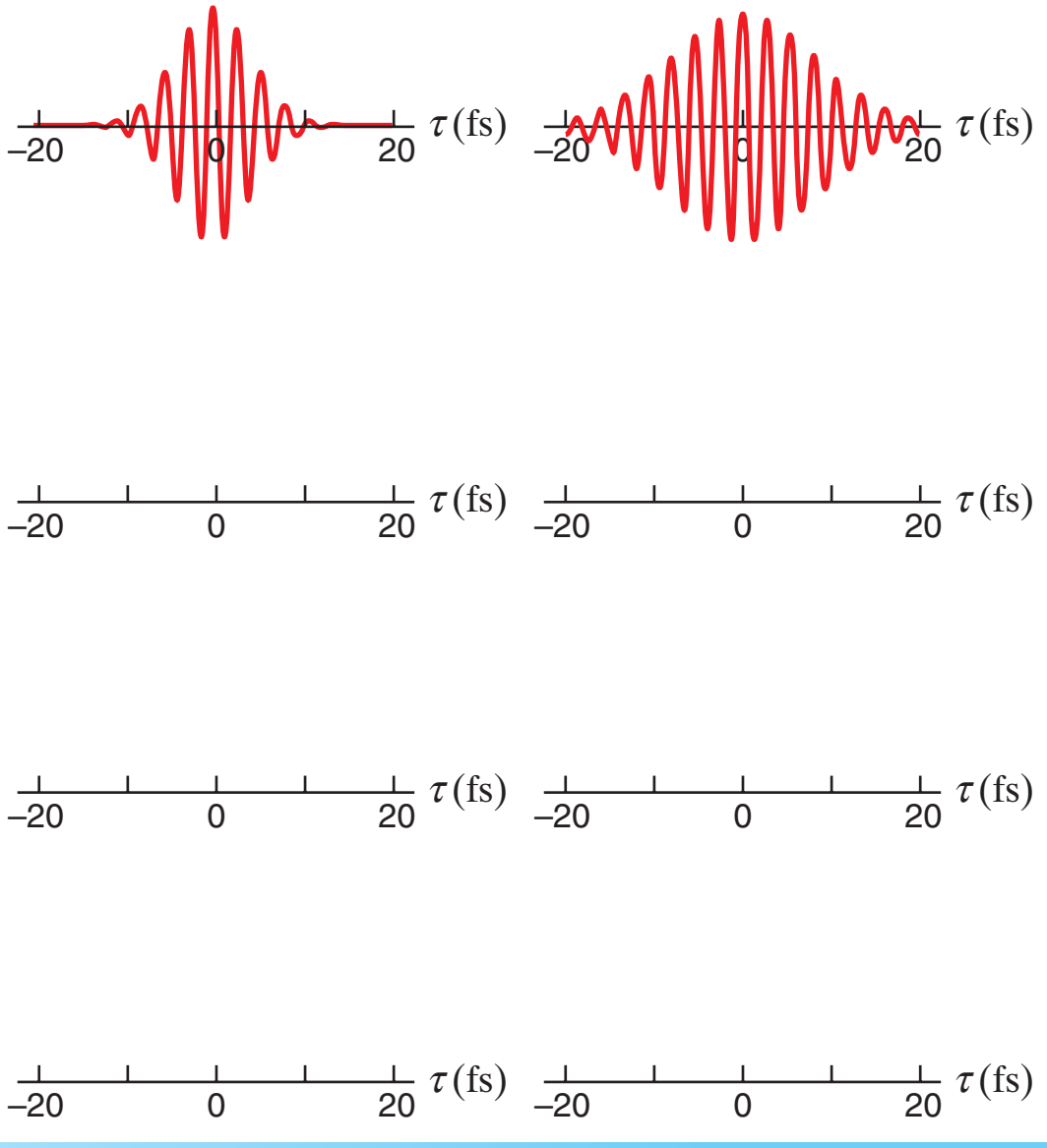
$$S_\omega(\tau) \propto \int \{ \left|E_1(t)\right|^2 + \left|E_2(t+\tau)\right|^2 + E_1(t)E_2{}^*(t+\tau) + E_1{}^*(t)E_2(t+\tau) \}\, dt$$

Temporal characterization









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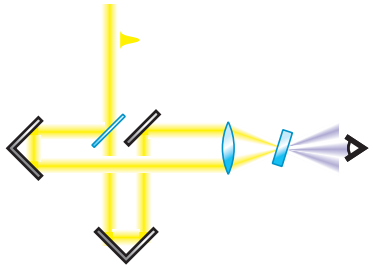

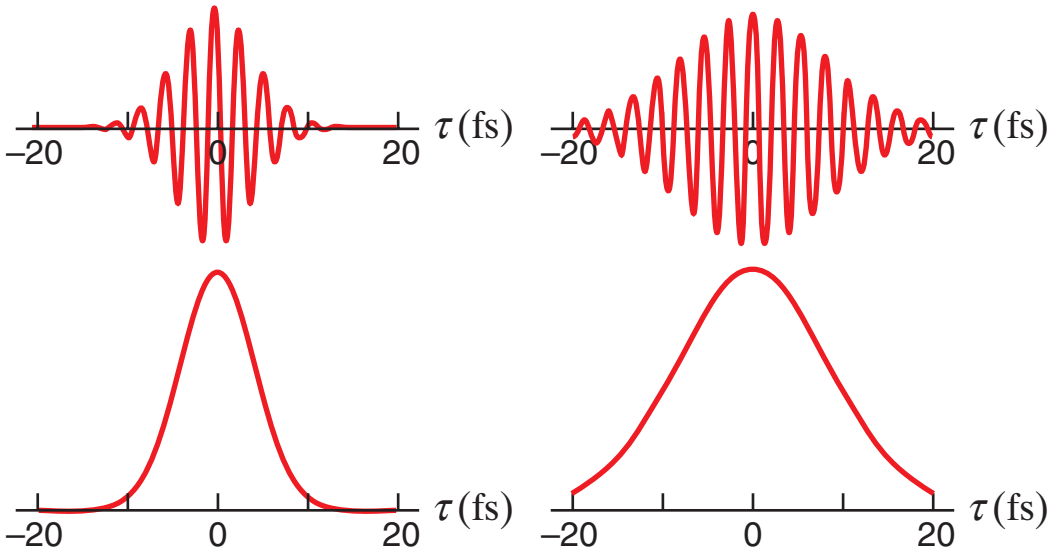

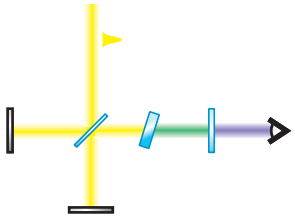

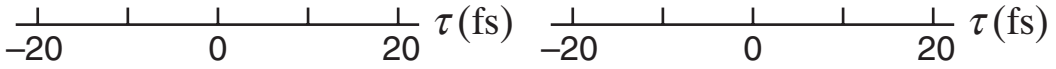

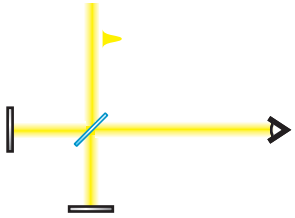

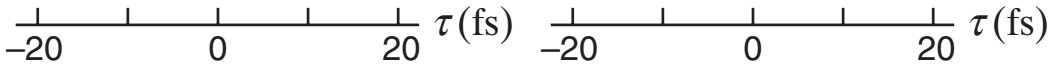

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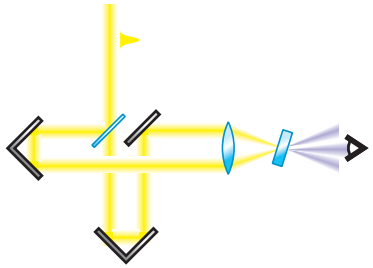

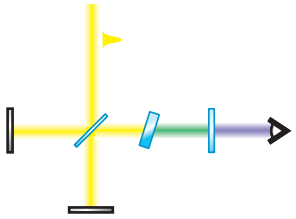

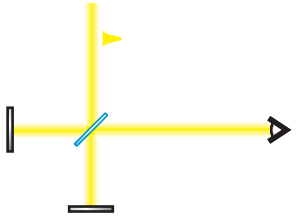

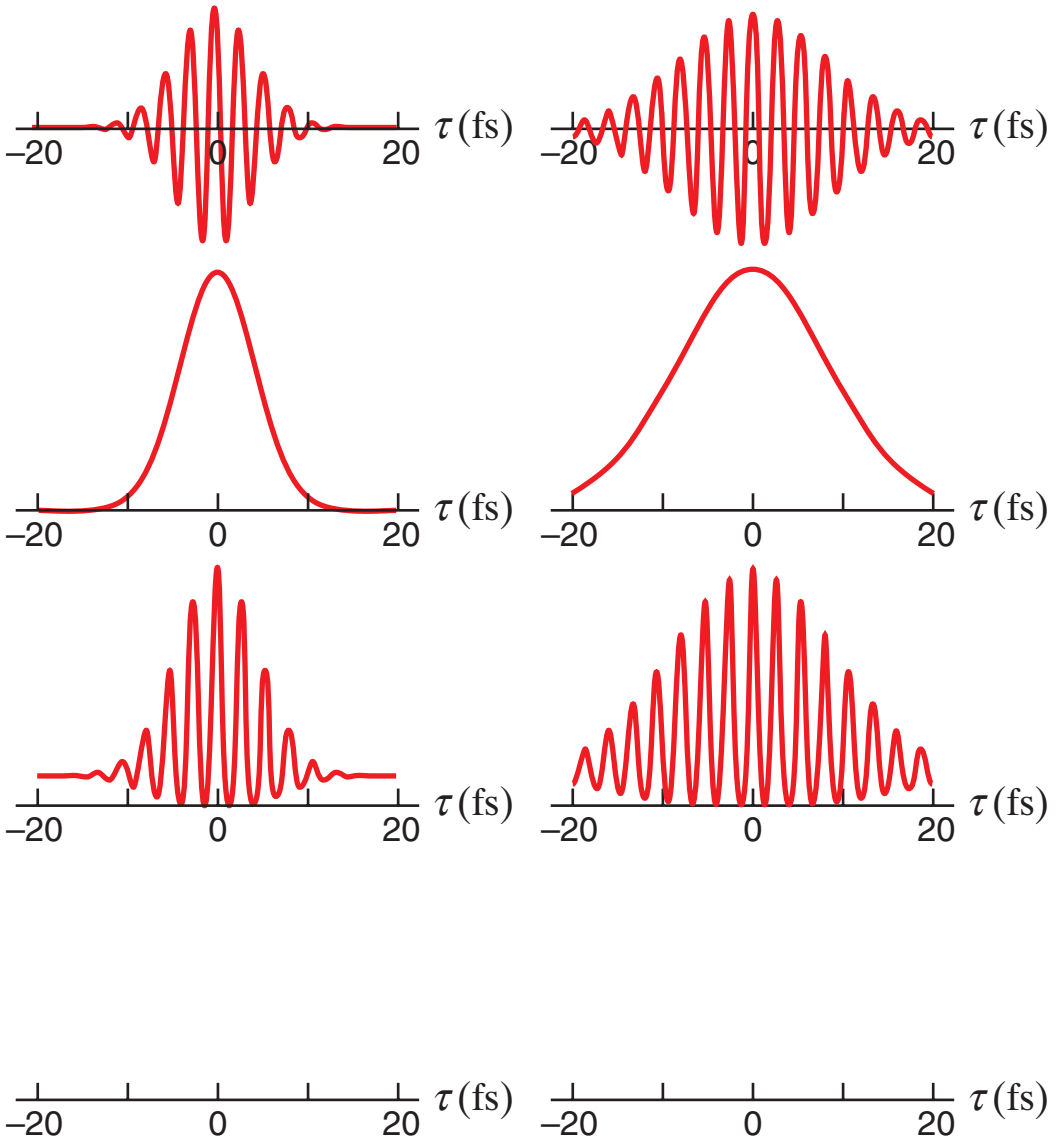

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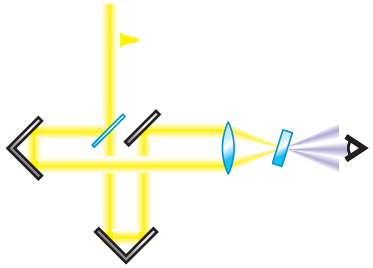
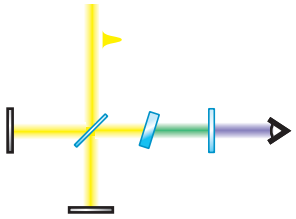
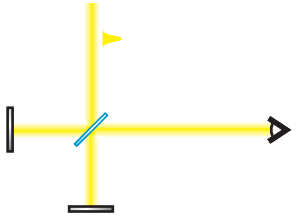
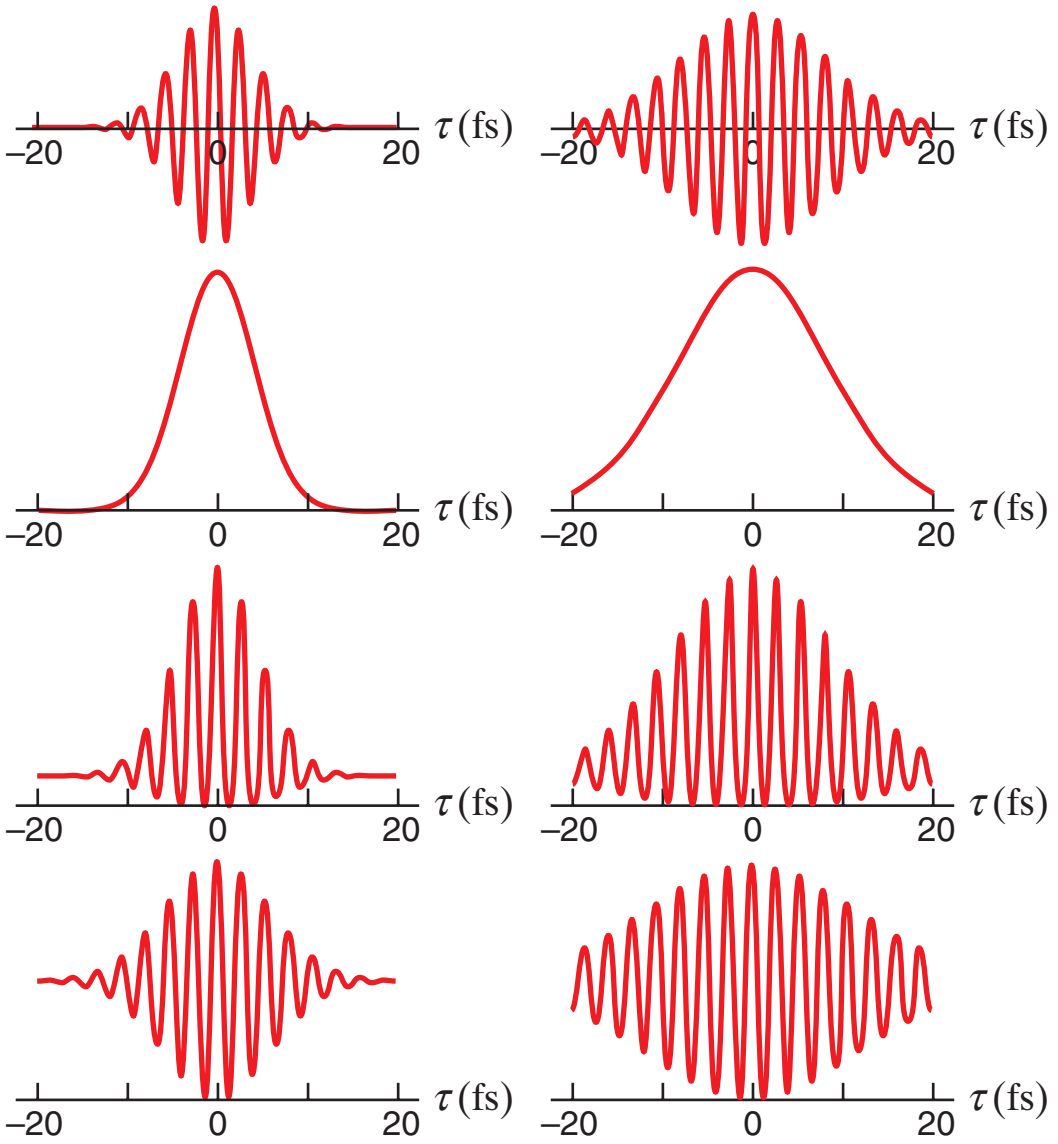




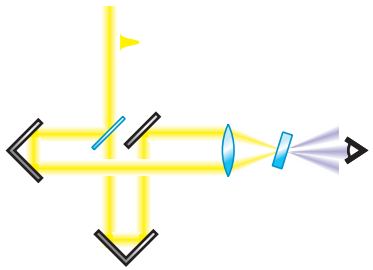


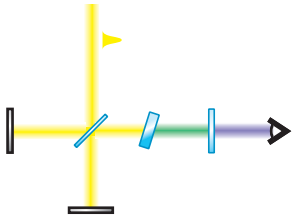


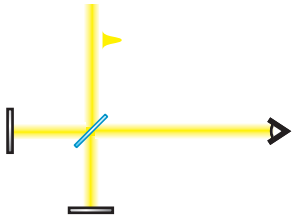
Temporal characterization

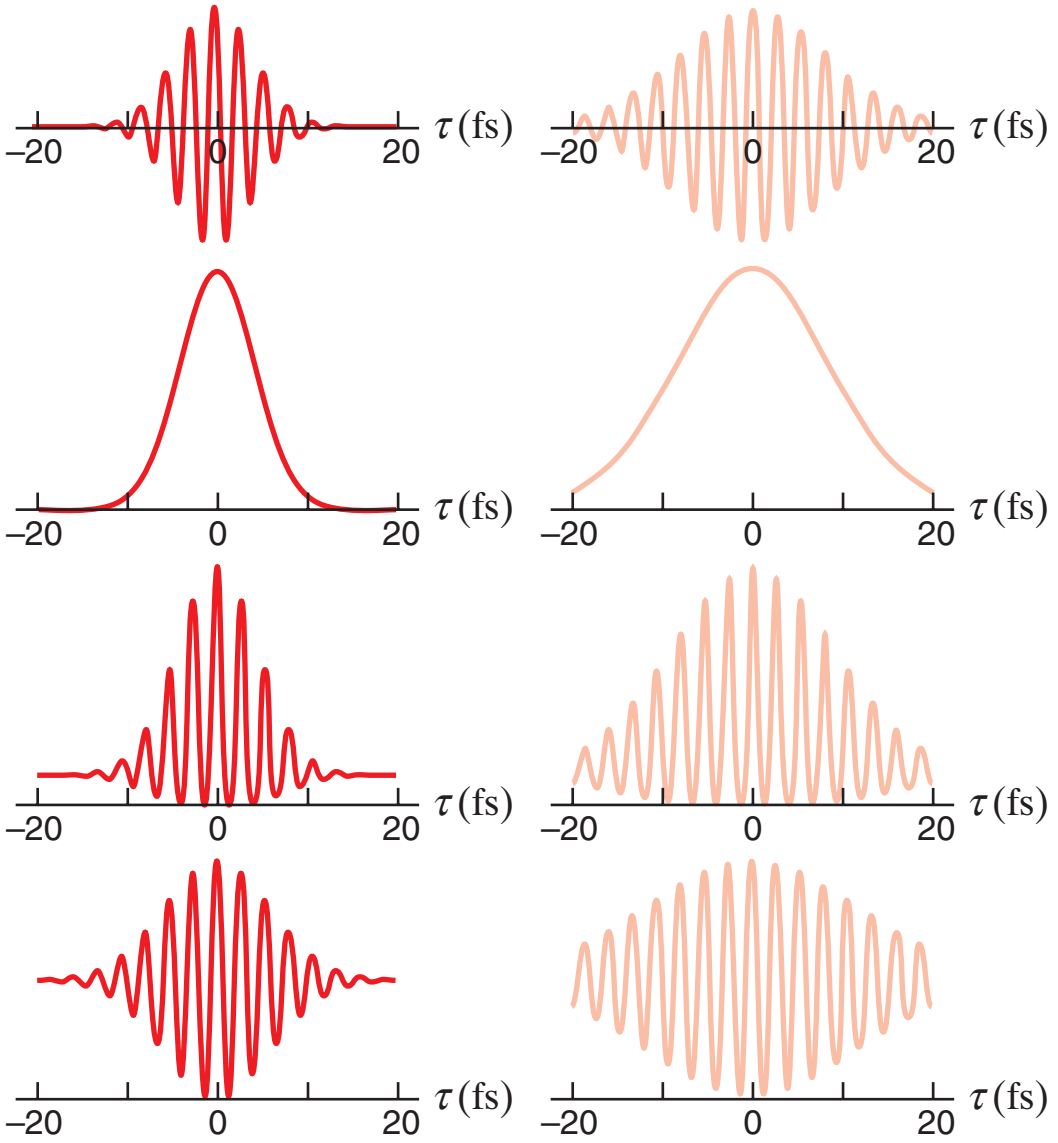
But what about dispersion?

Temporal characterization

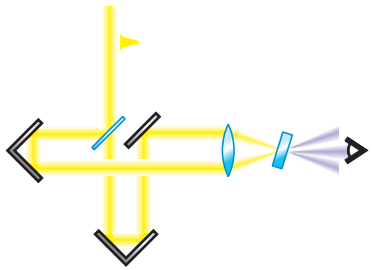


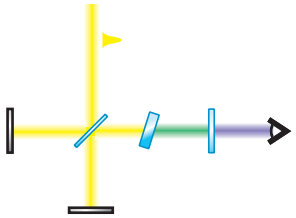


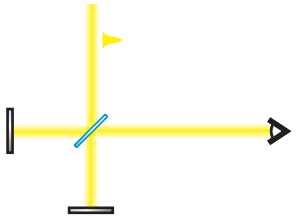


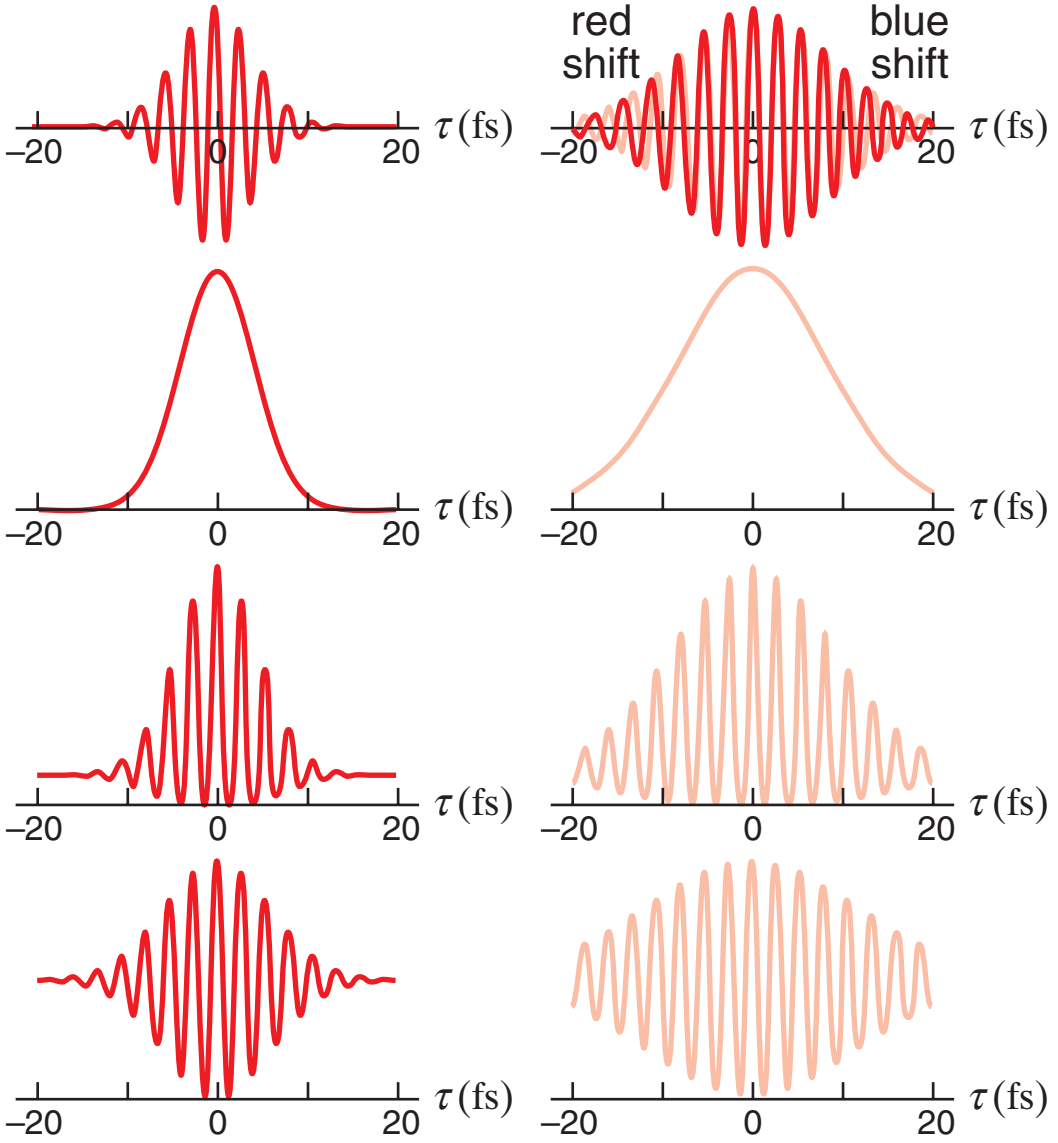


Temporal characterization

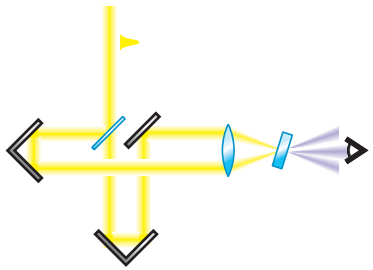


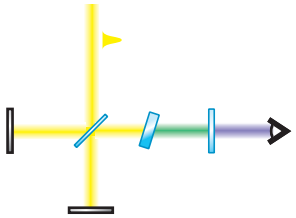


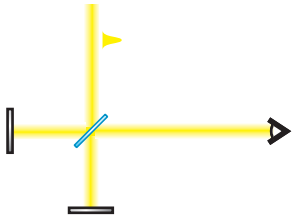


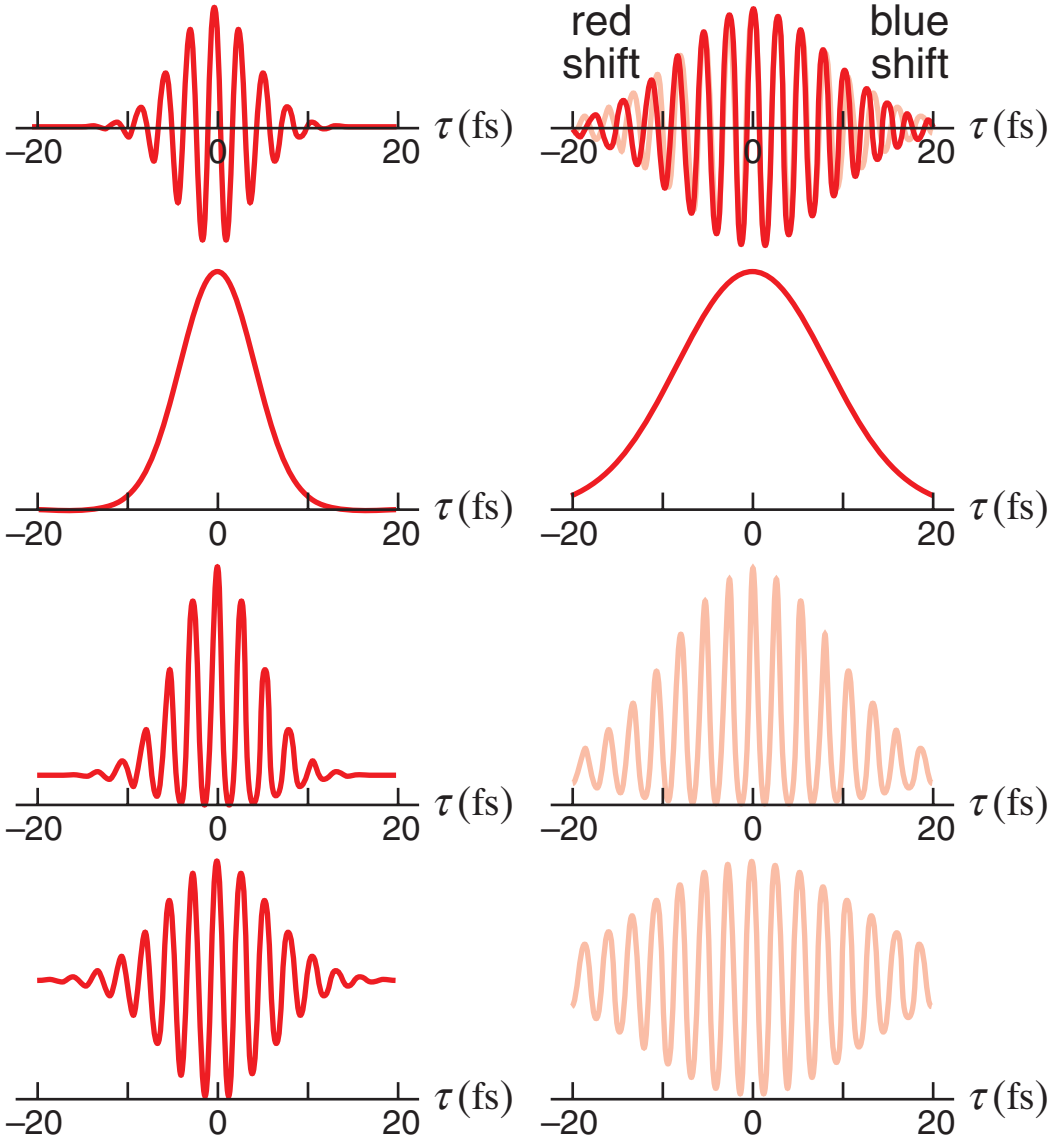


Temporal characterization









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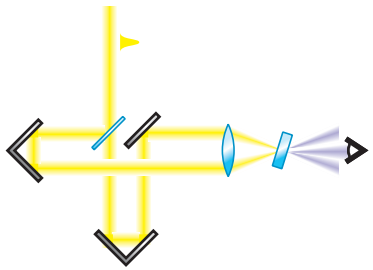

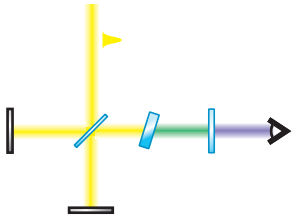

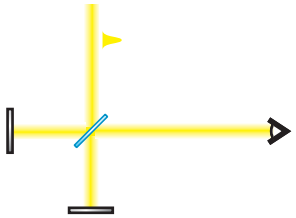

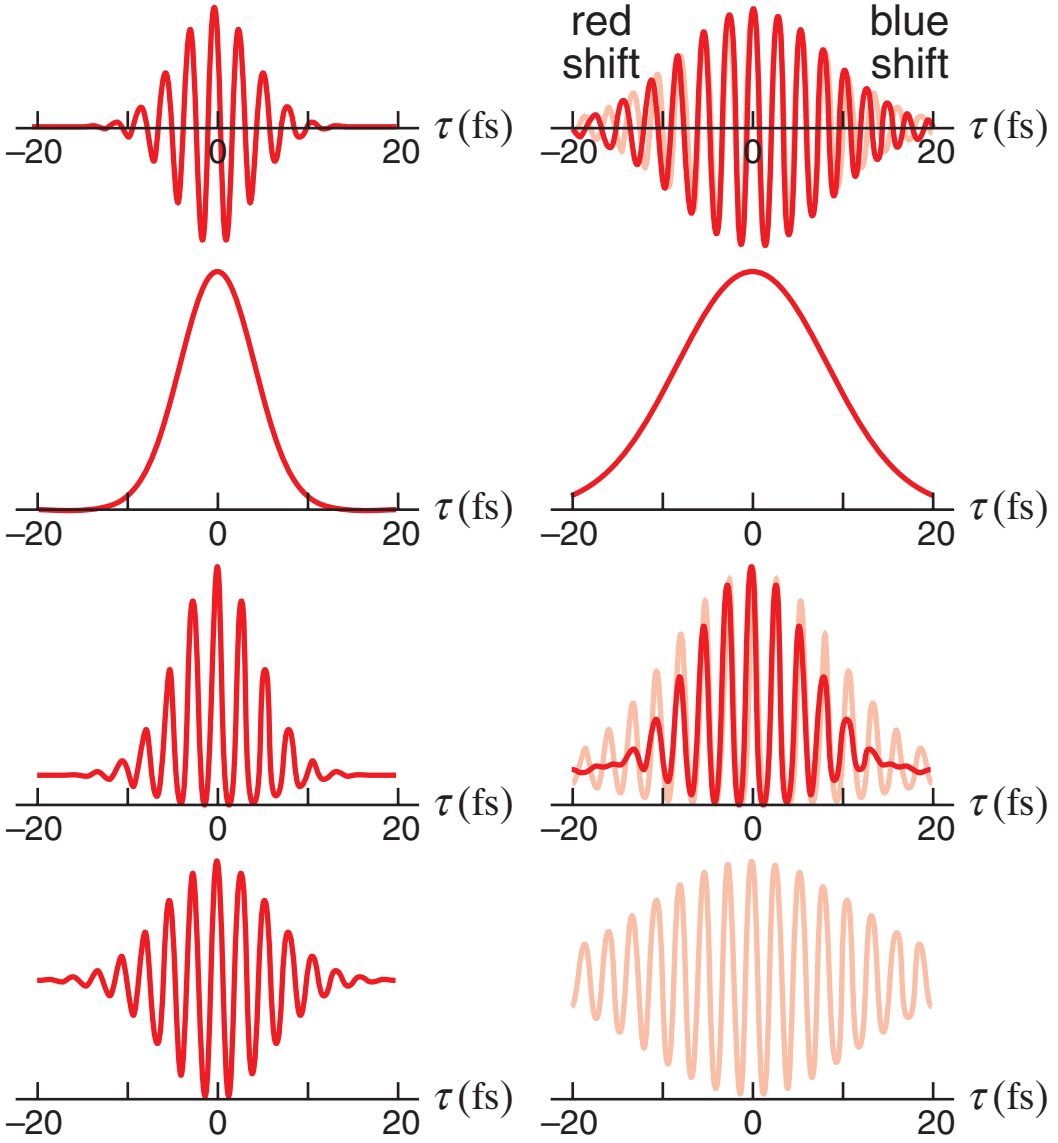

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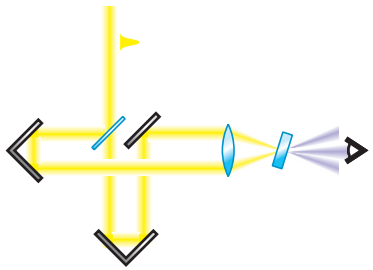

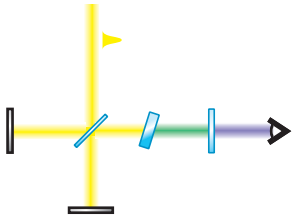

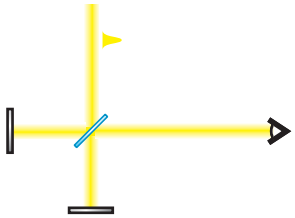

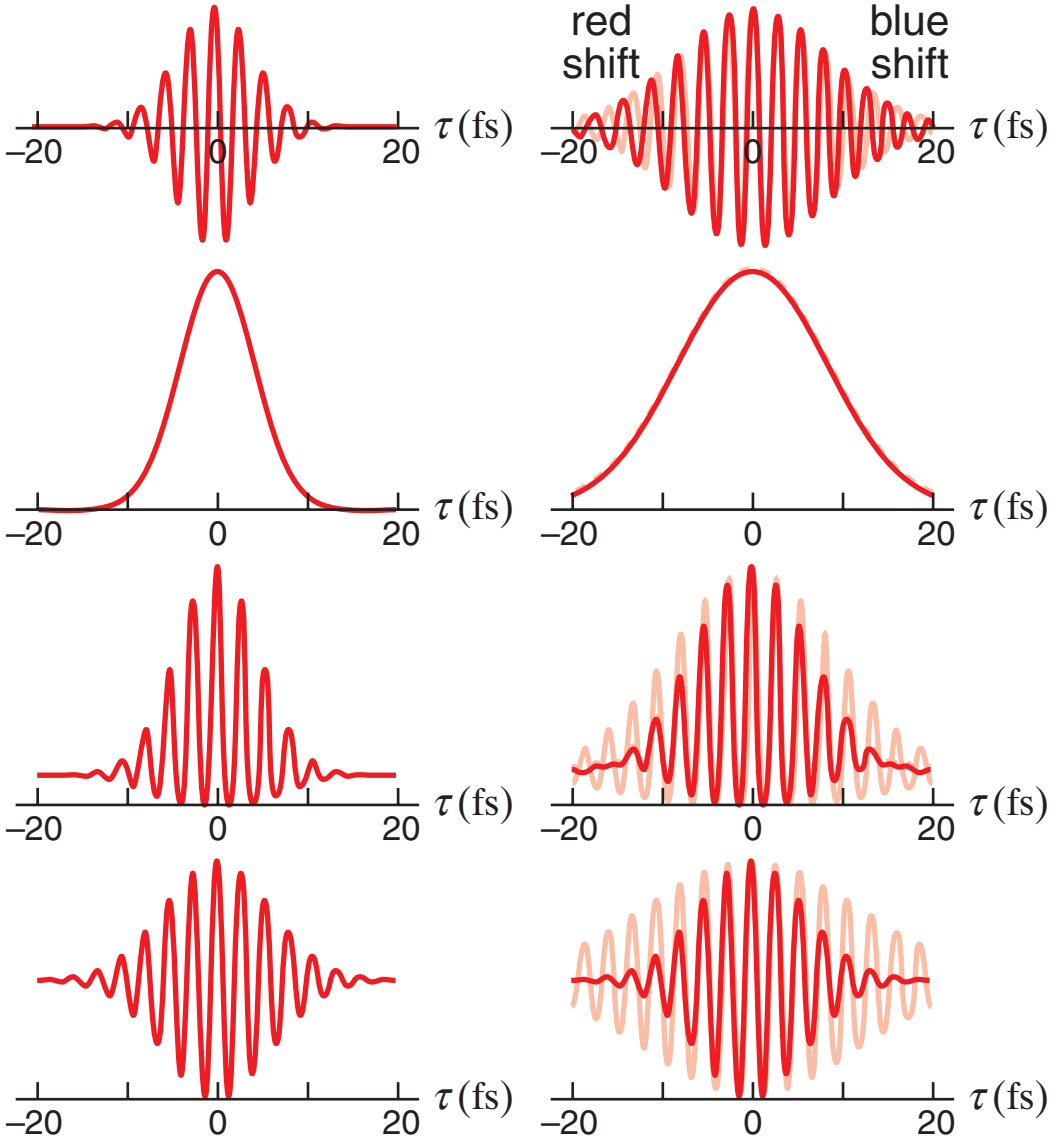

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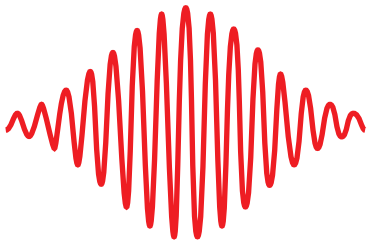

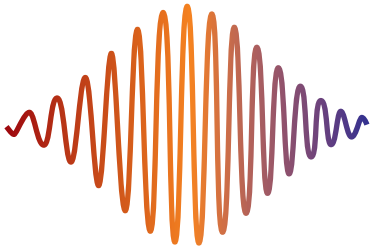

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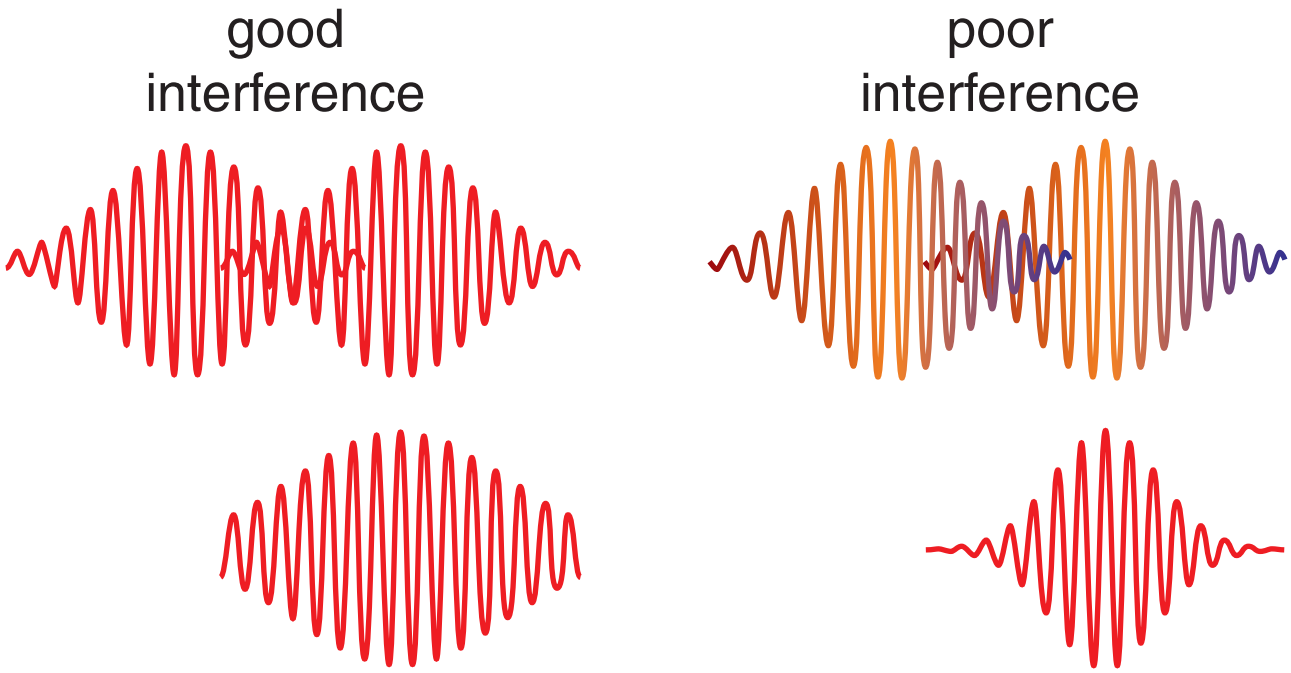

Temporal characterization

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

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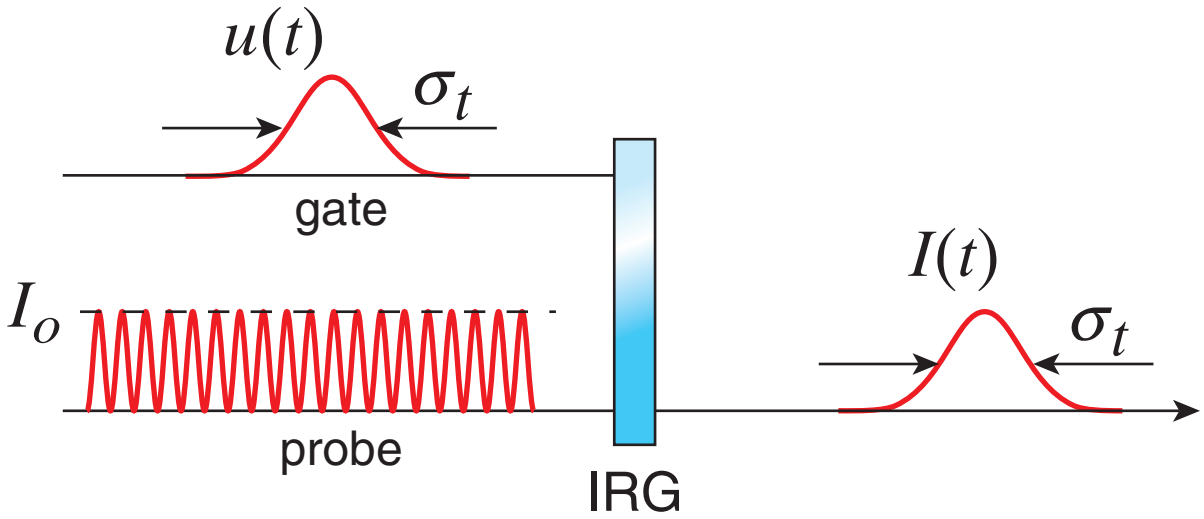
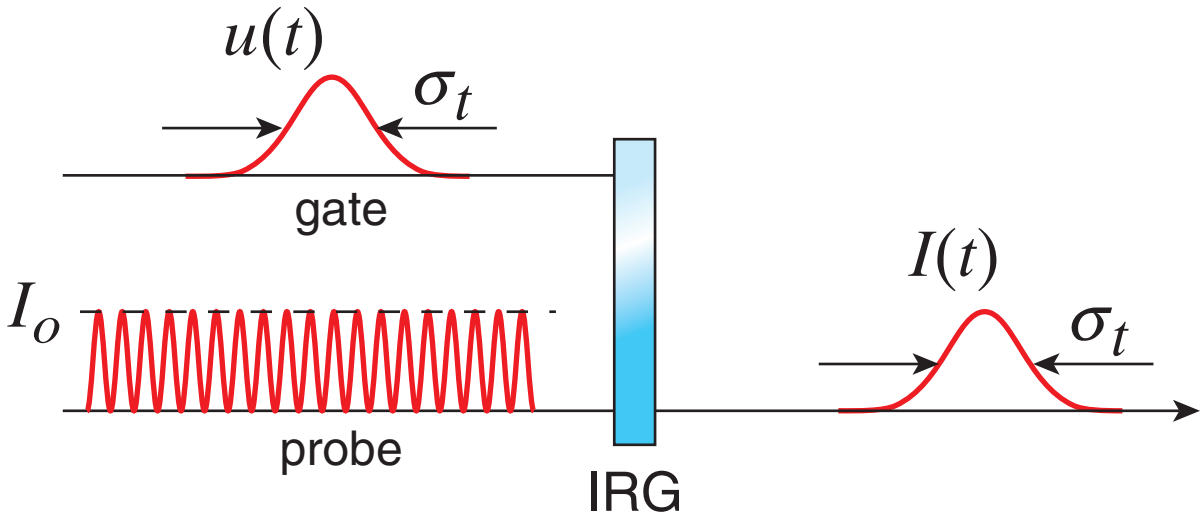
Joint time-frequency measurements



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

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

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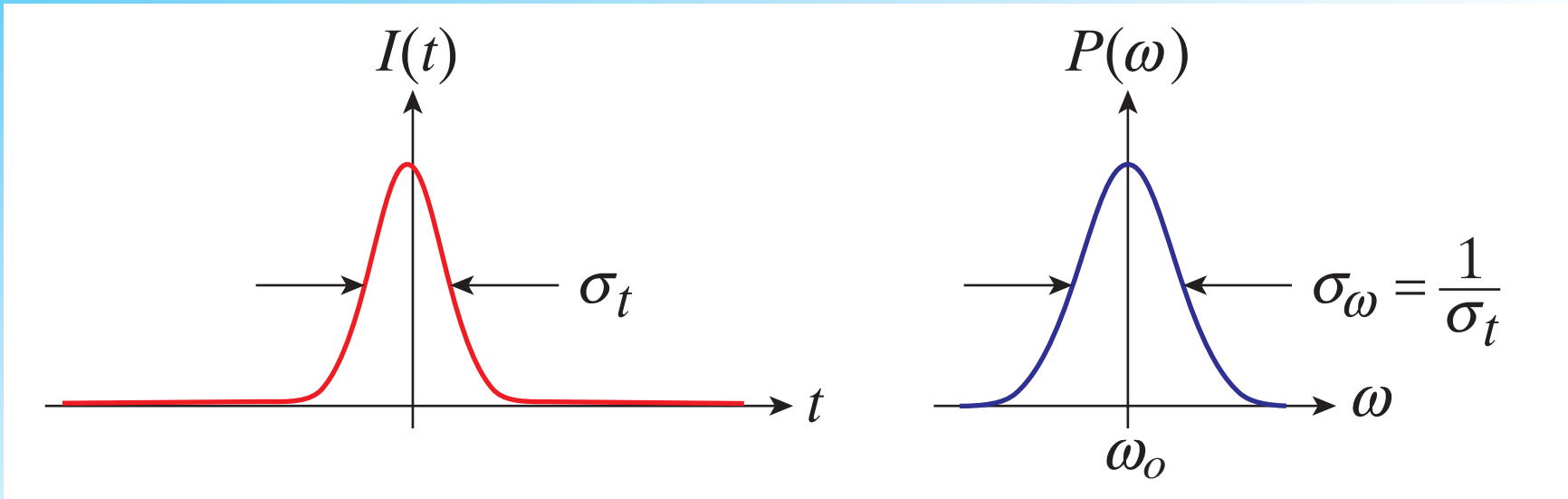

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$$\sigma_t \sigma_\omega = 1$$

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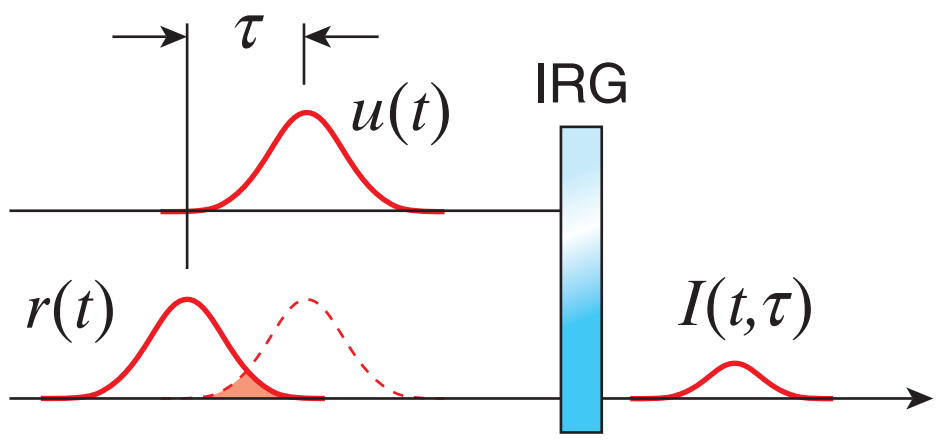
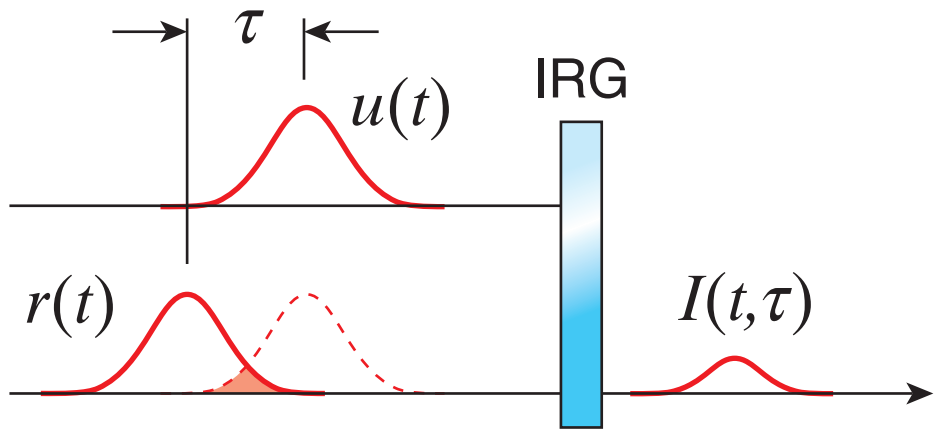


Transmitted intensity

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

$$= \exp\left[-\frac{2t^2+2t\tau+\tau^2}{\sigma^2}\right] = \exp\left[-\frac{2t^2+2t\tau+\tau^2/2}{\sigma^2}\right]\exp\left[-\frac{\tau^2}{2\sigma^2}\right] =$$

Joint time-frequency measurements

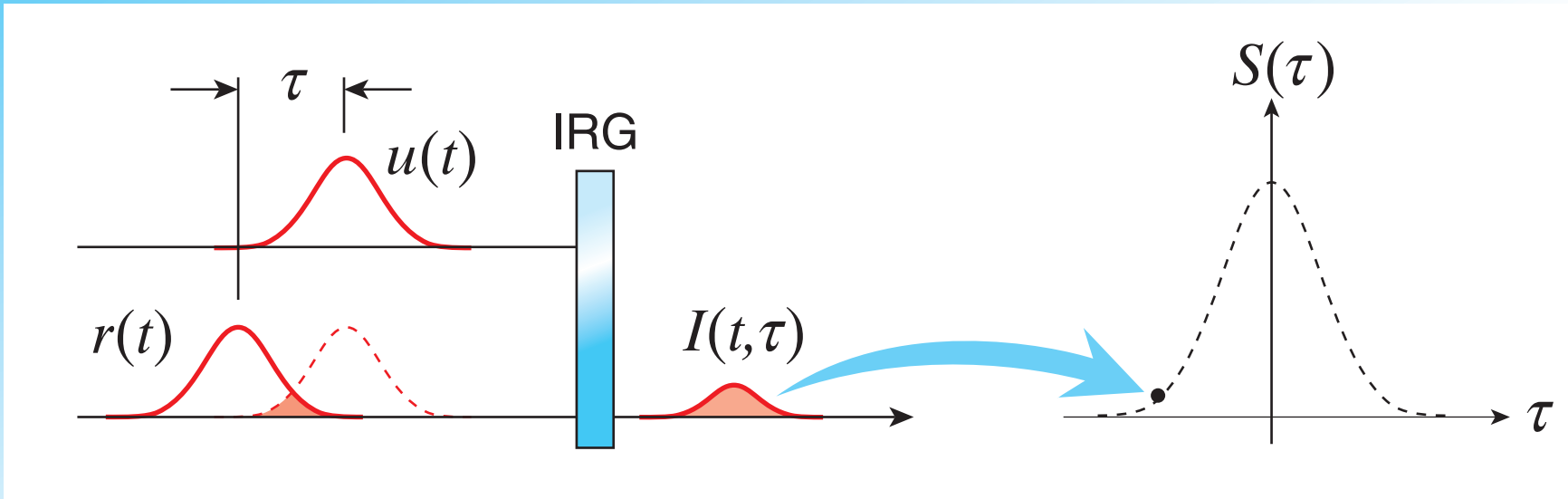


Transmitted intensity

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

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

Joint time-frequency measurements



…but detector integrates $I(t,\tau)$:

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

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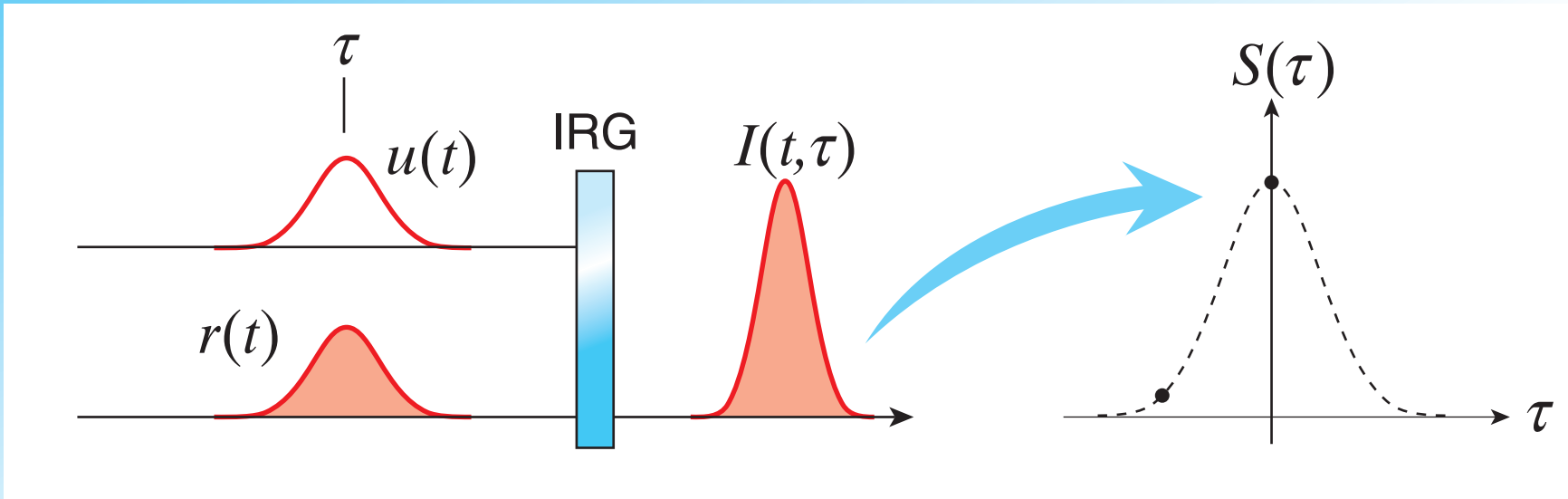

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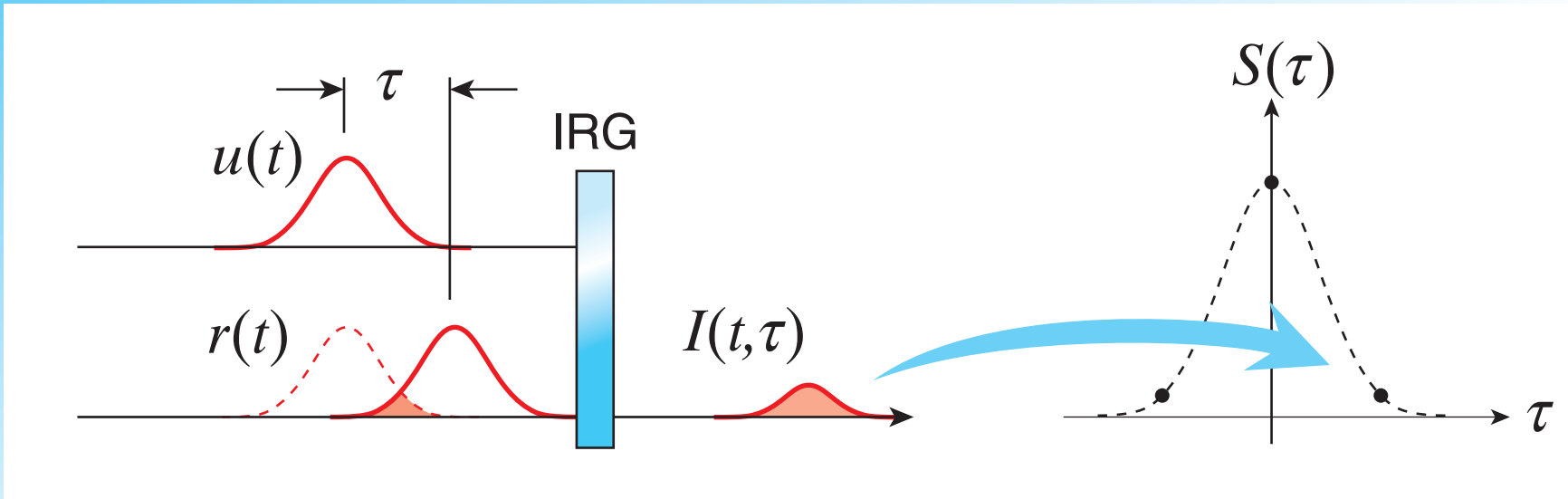

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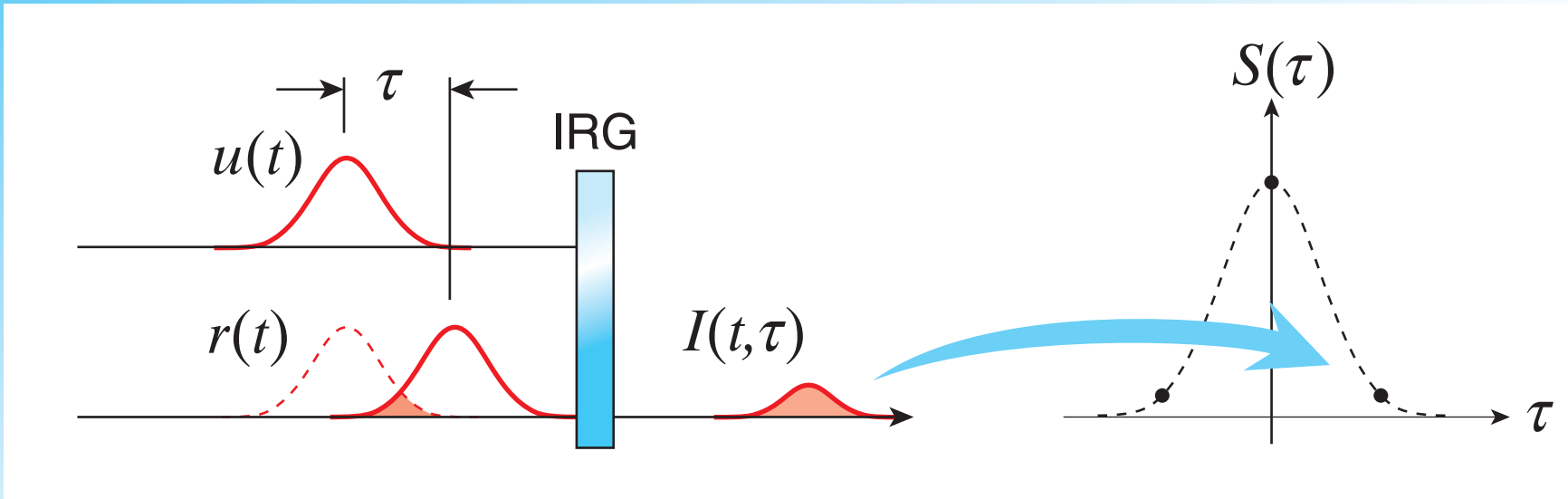

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$$= \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \int_{-\infty}^{\infty} \exp\left[-\frac{2u^2}{\sigma^2}\right] du = \sqrt{\frac{\pi}{2}}\,\sigma \exp\left[-\frac{\tau^2}{(\sigma\sqrt{2})^2}\right]$$

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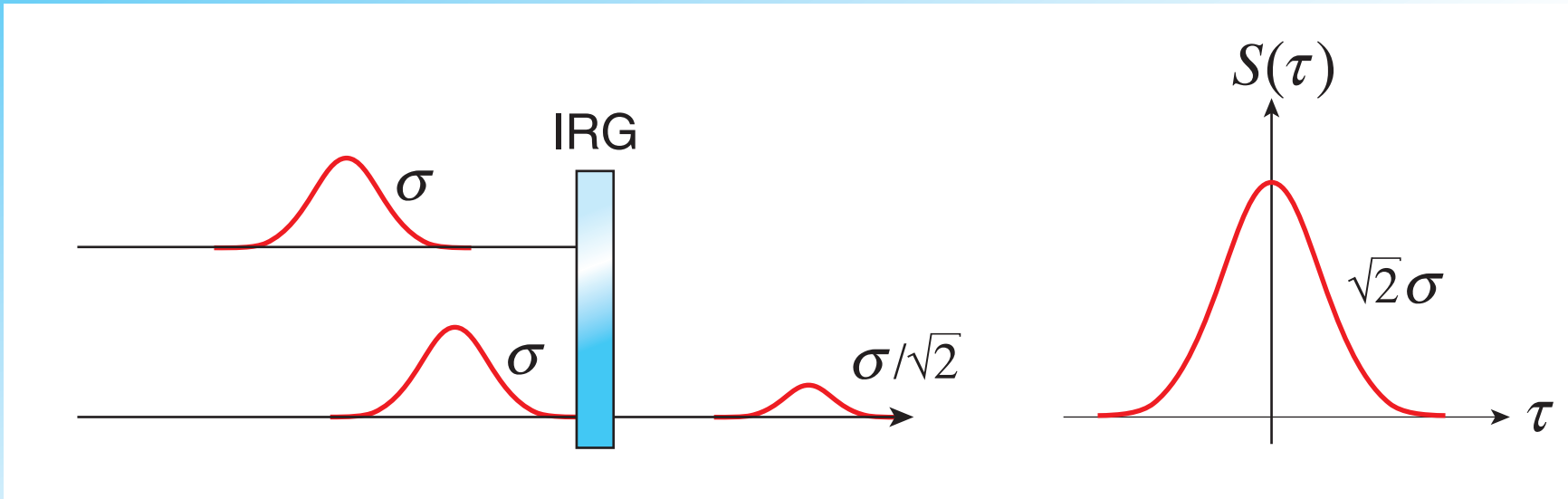

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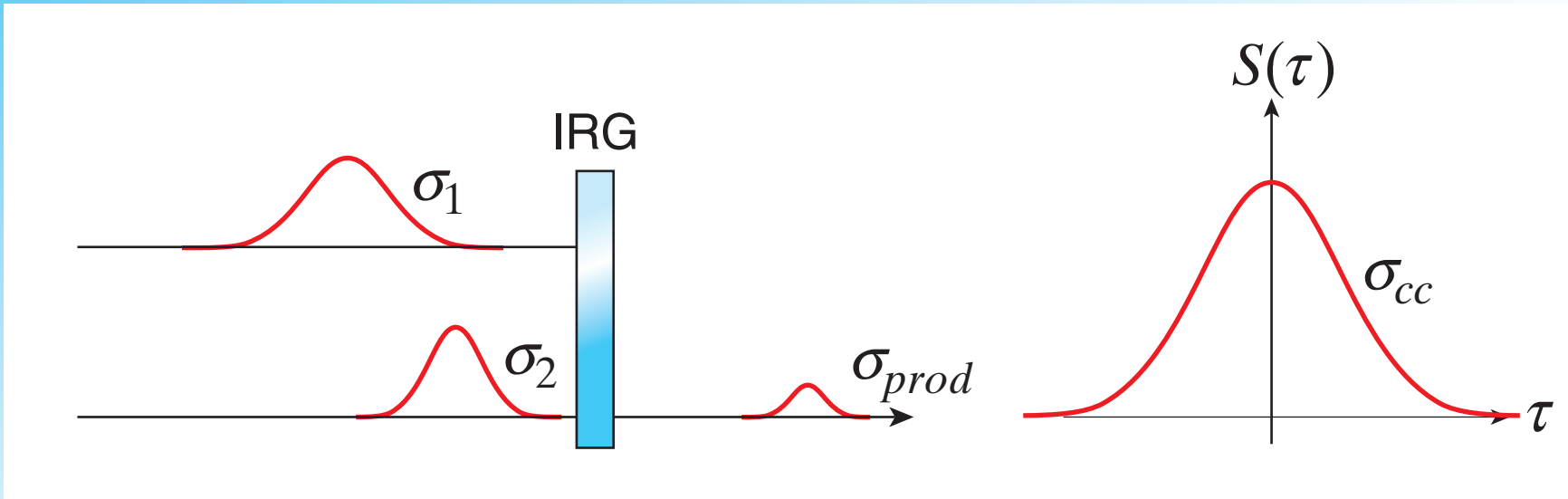
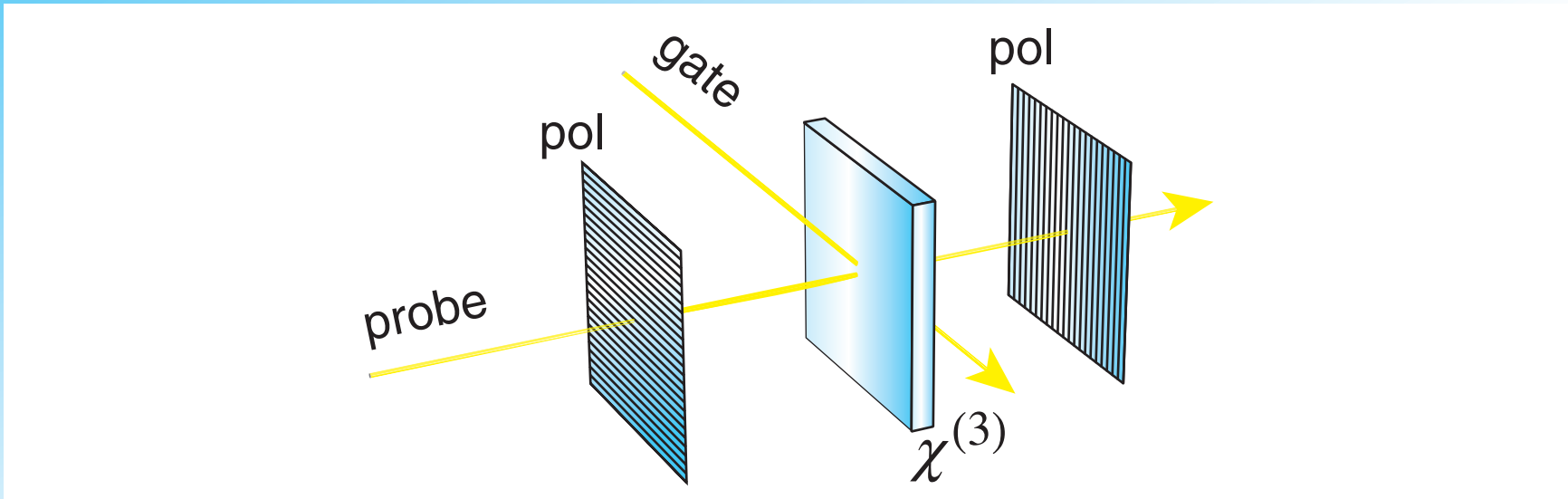


If gate and probe unequal:

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

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

Joint time-frequency measurements

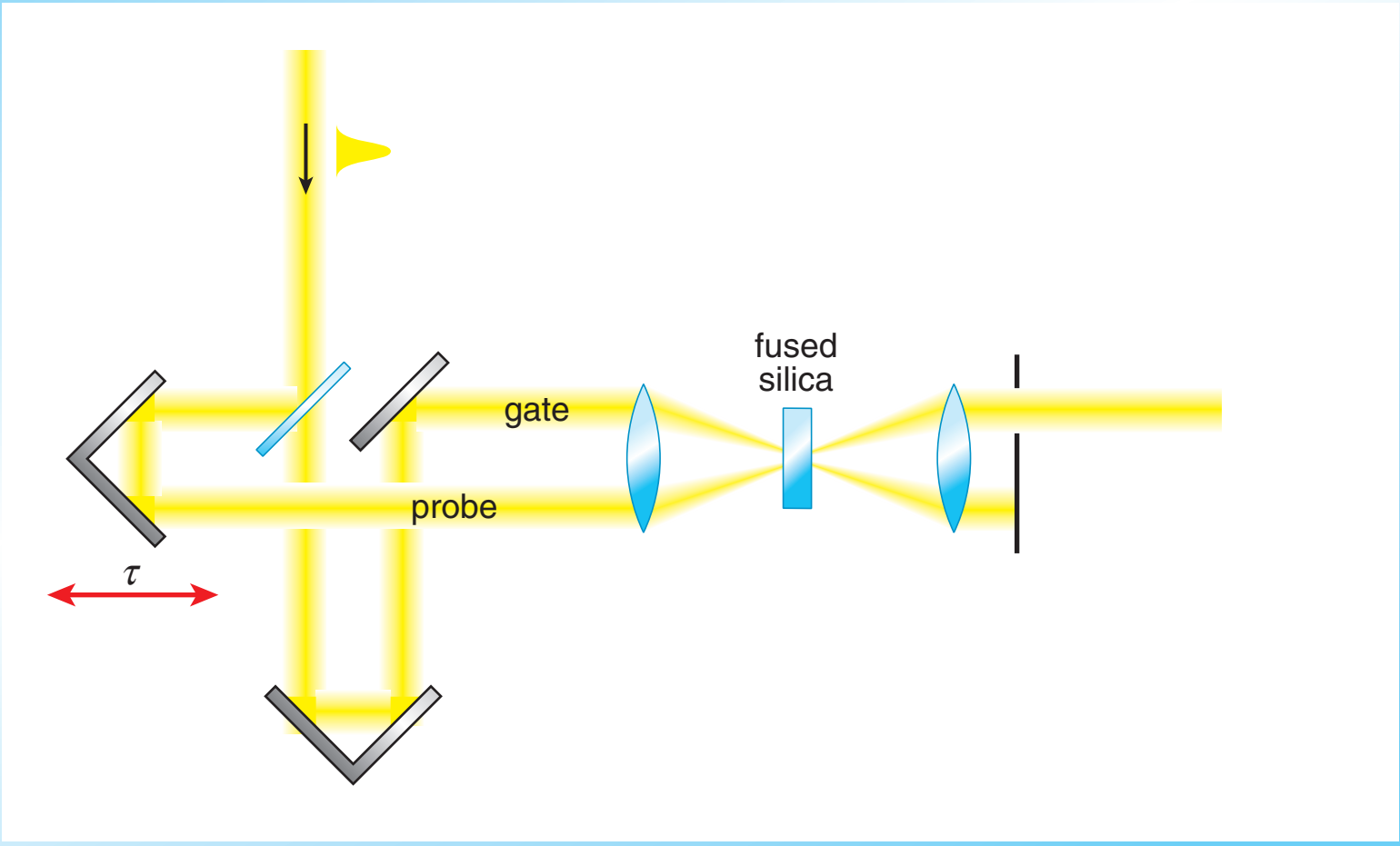


Transmitted field:

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

Joint time-frequency measurements

FROG: frequency-resolved optical gating



Joint time-frequency measurements

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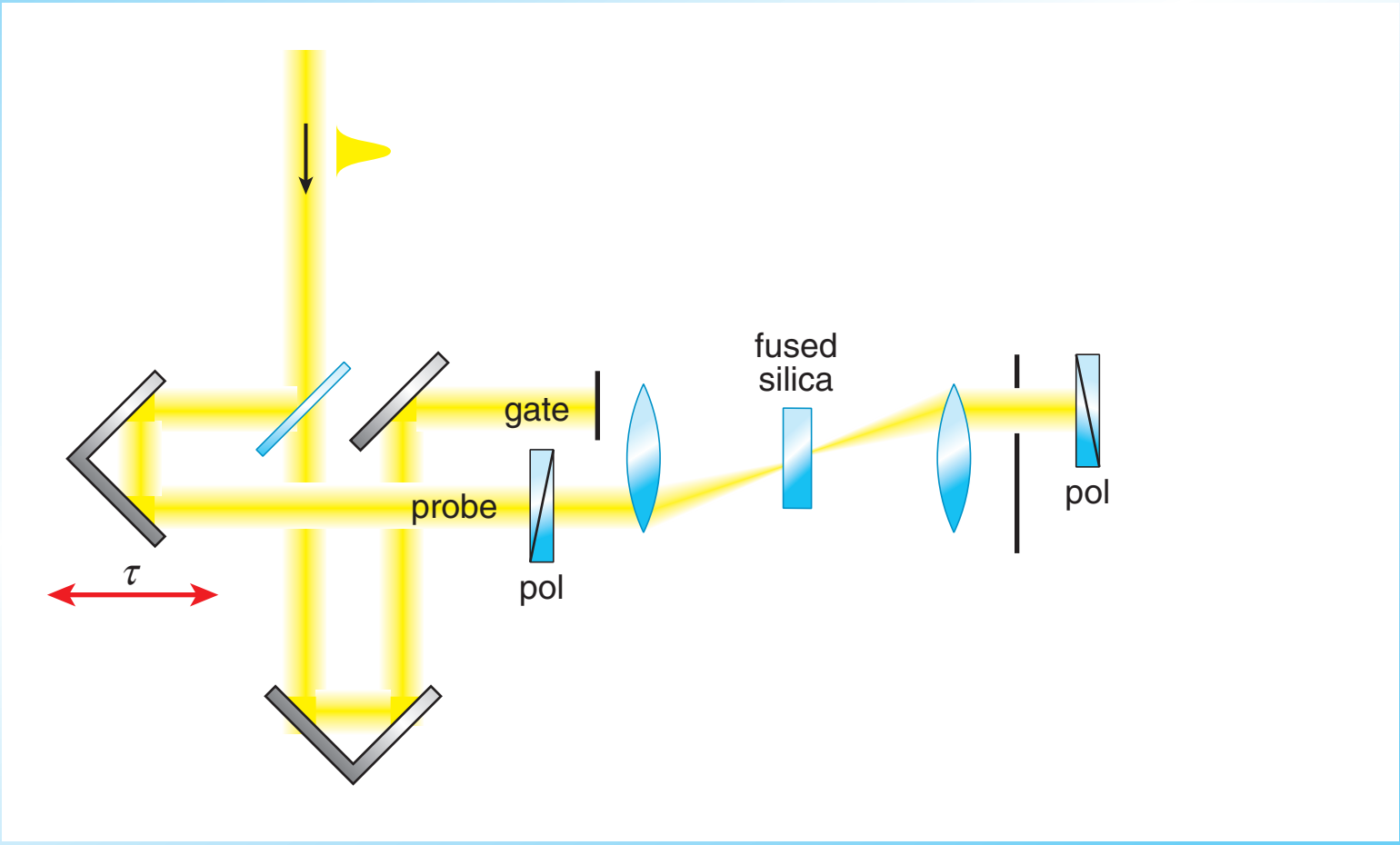

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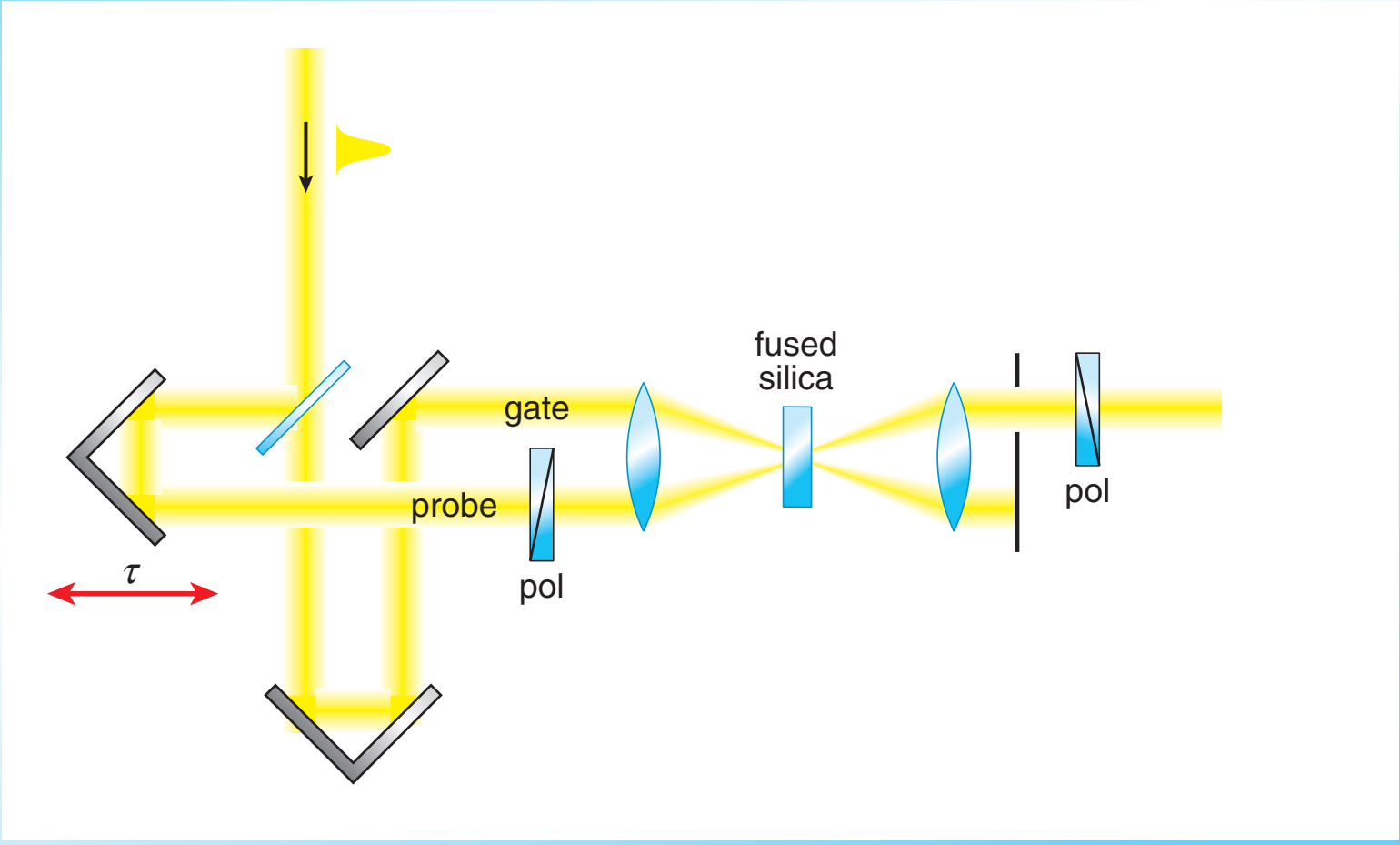

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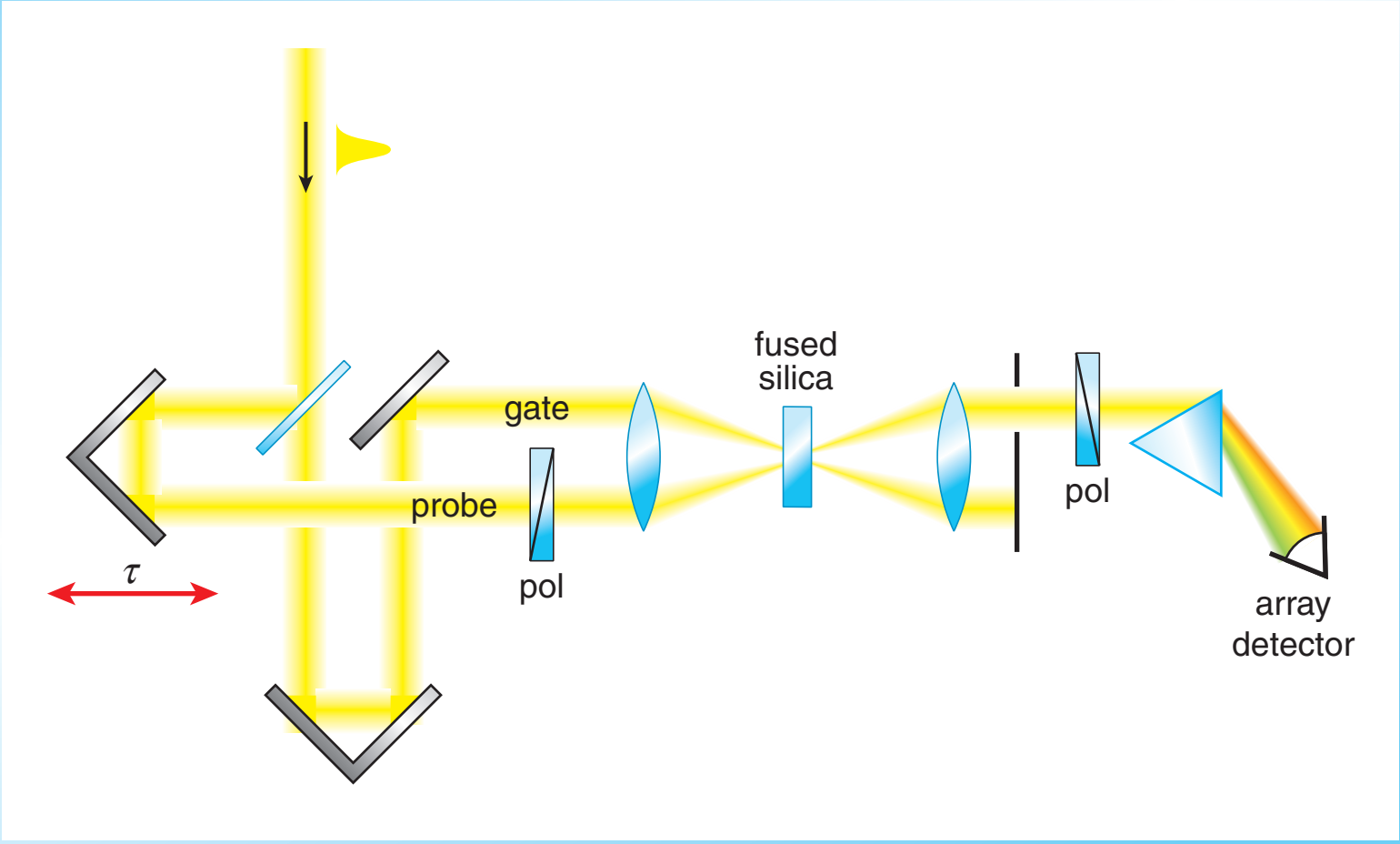

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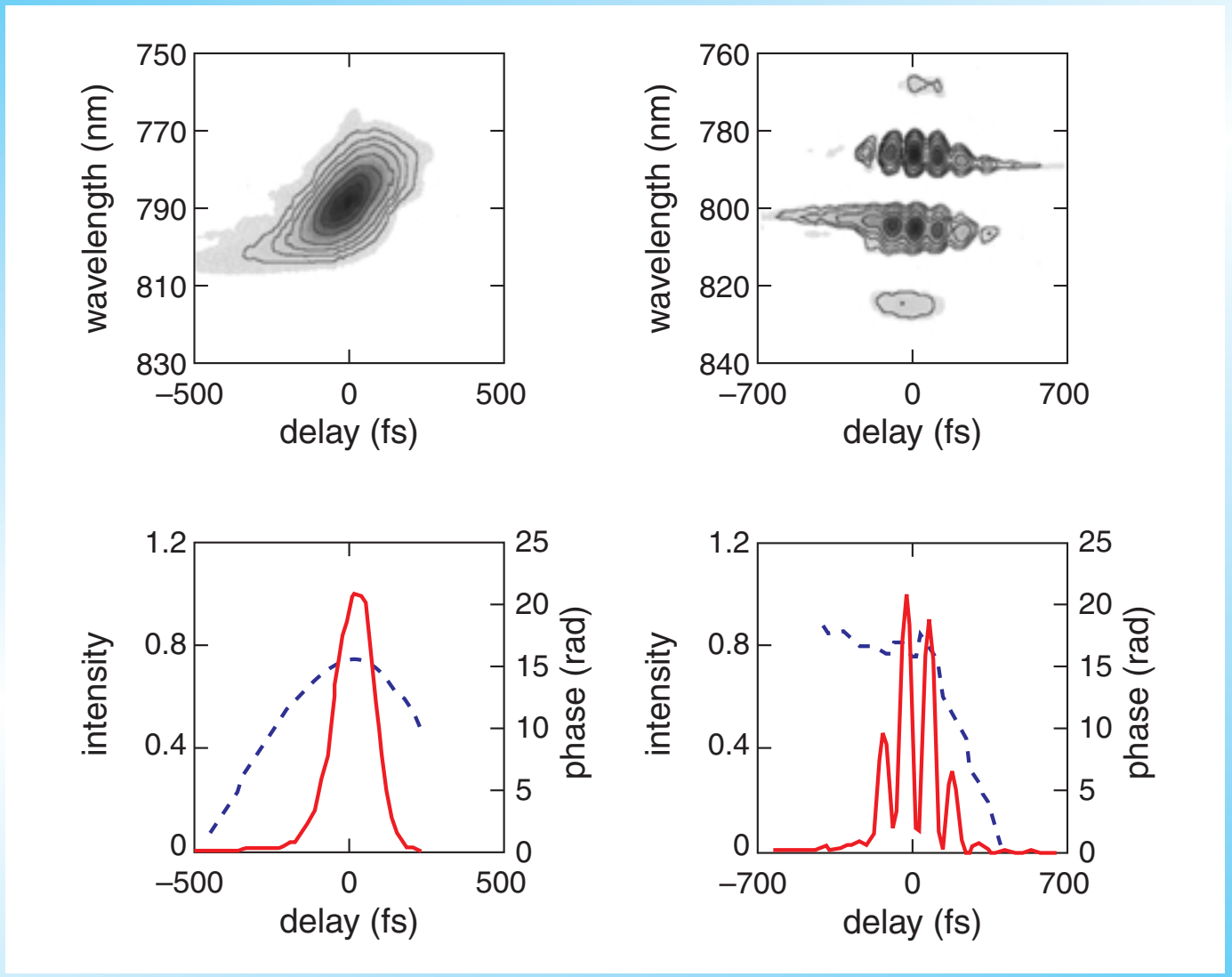
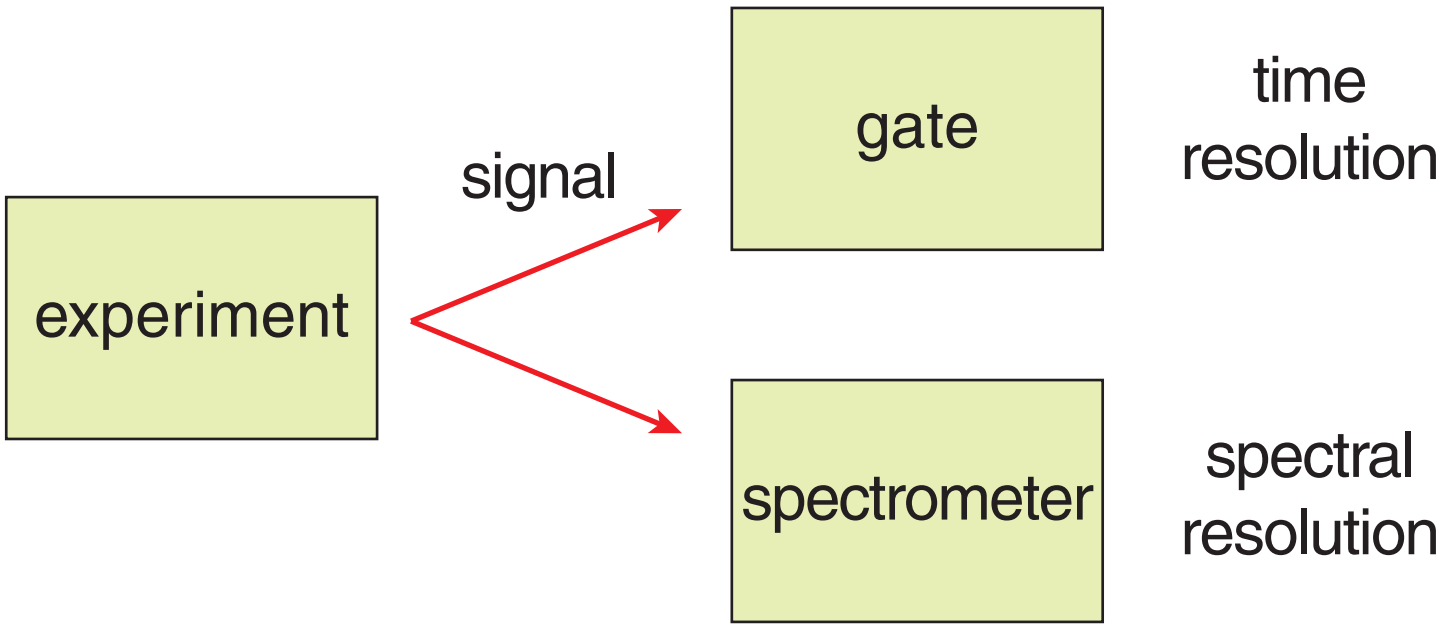


R. Trebino, *et al.*, Rev. Sci. Instrum. 68, 3277 (1997)

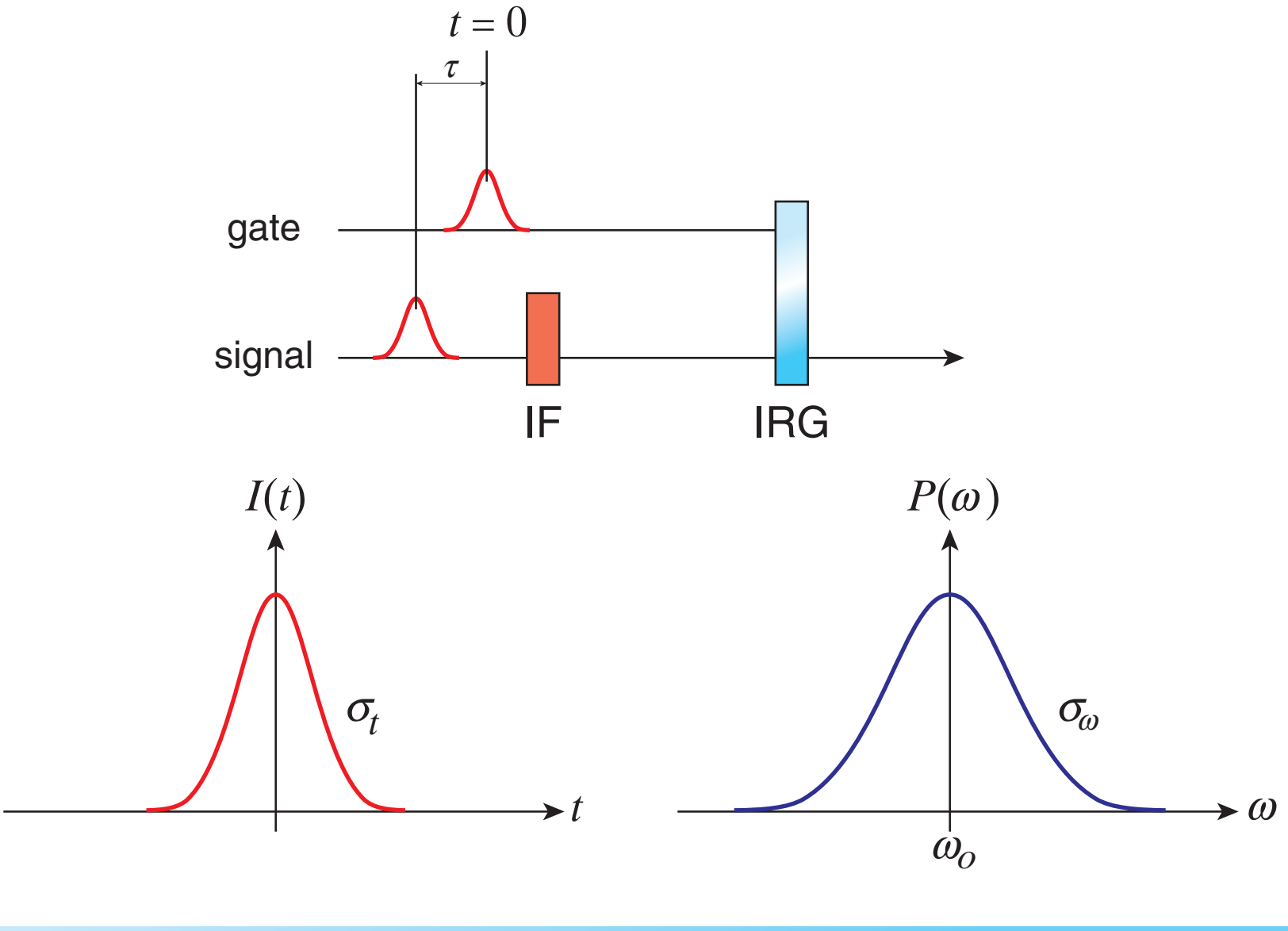
Joint time-frequency measurements



Joint time-frequency measurements

What are the resolution limits?

Experiment 1



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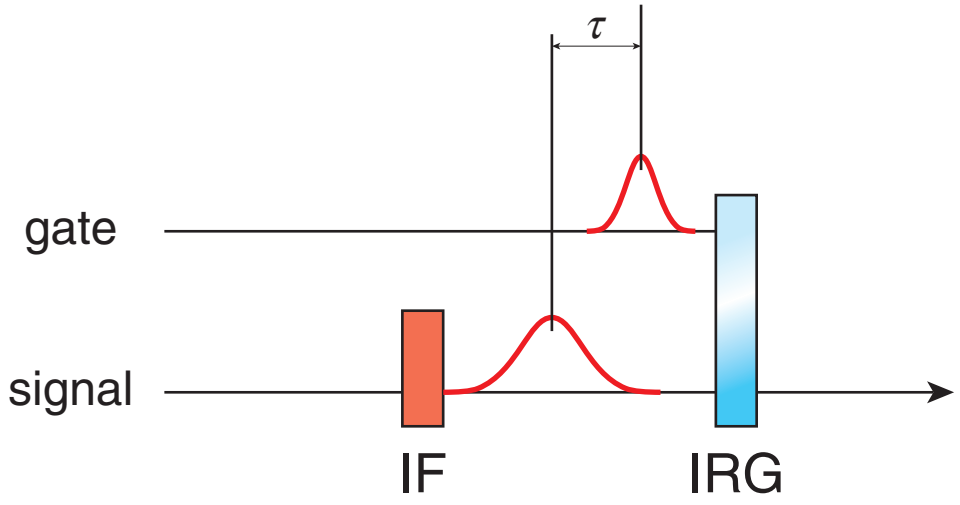

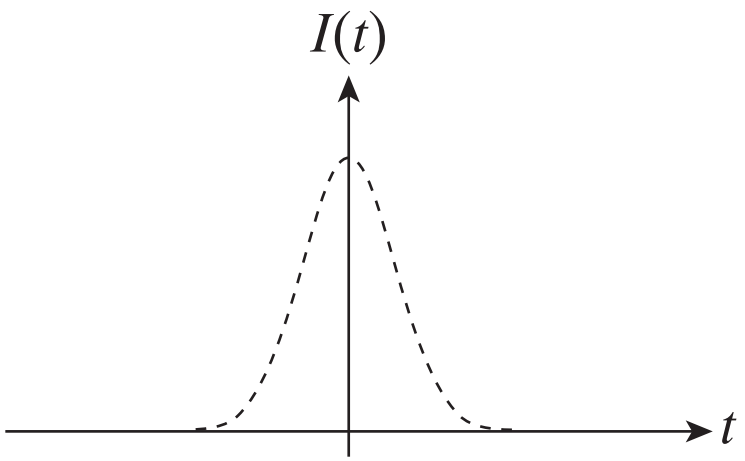

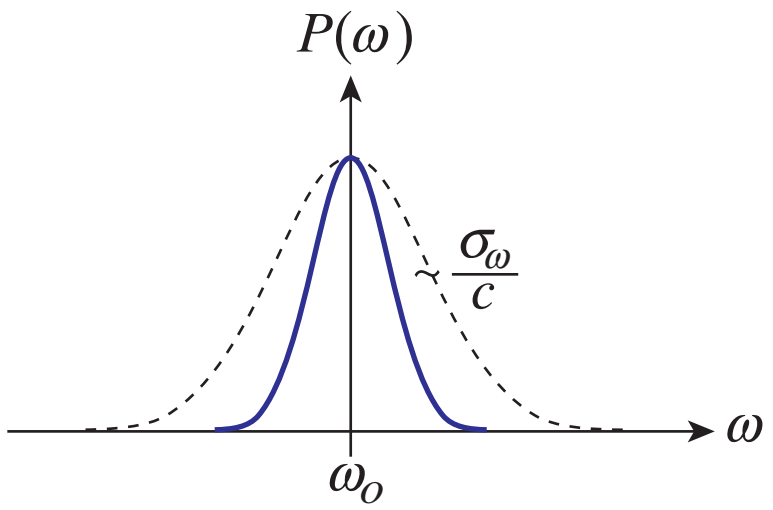

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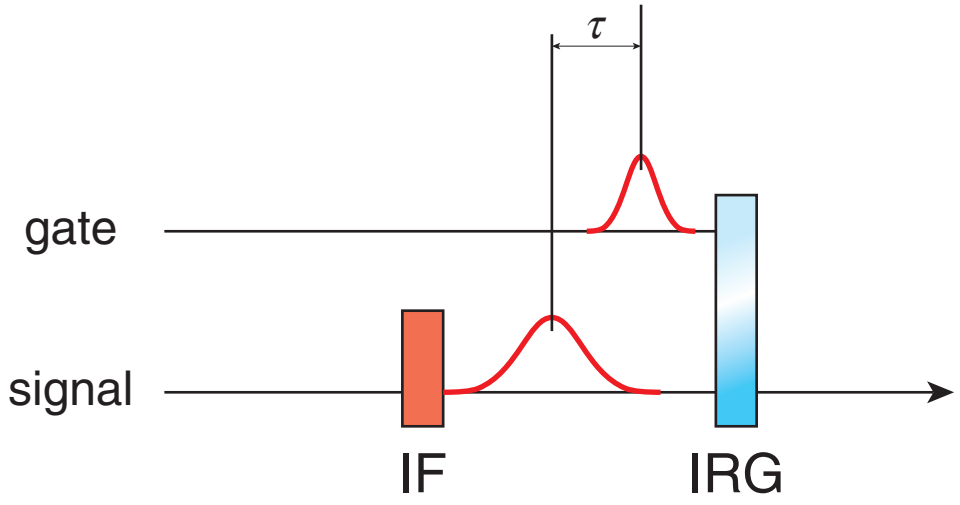

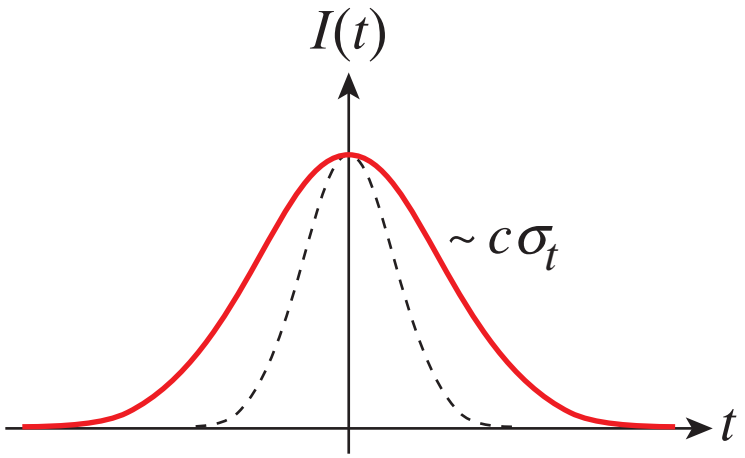

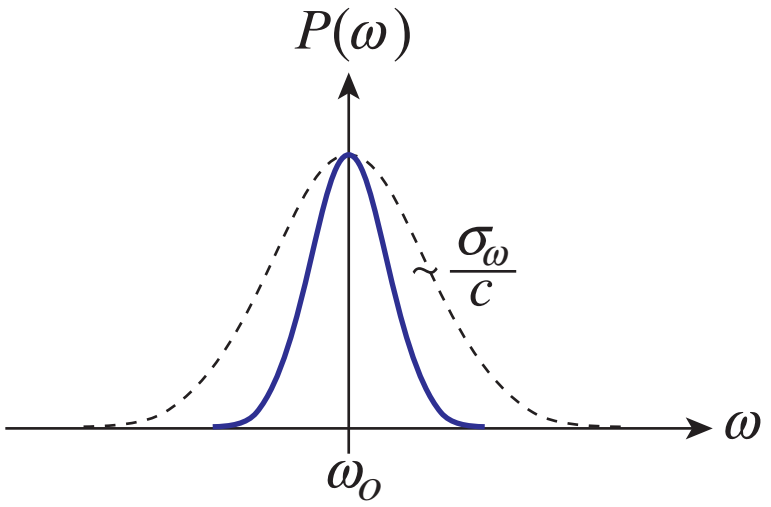

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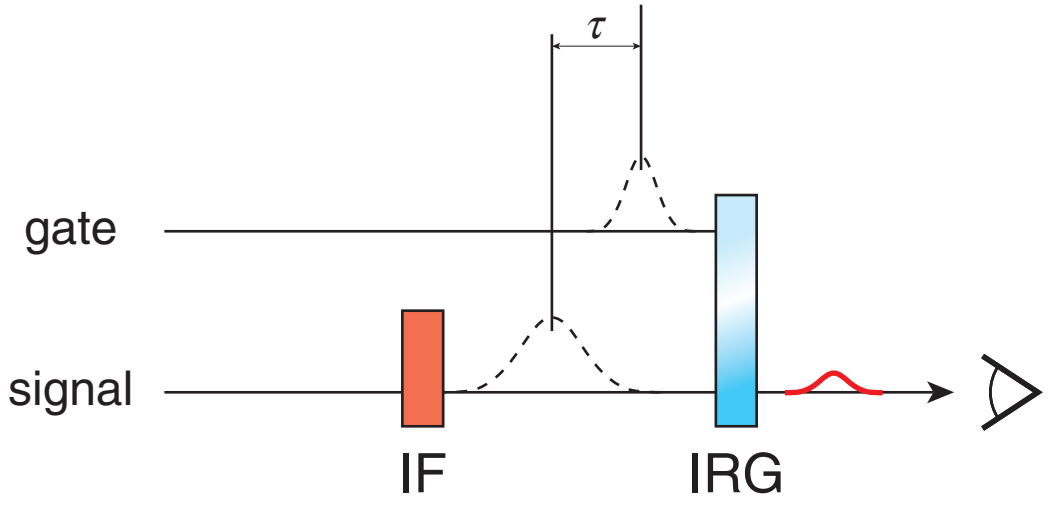
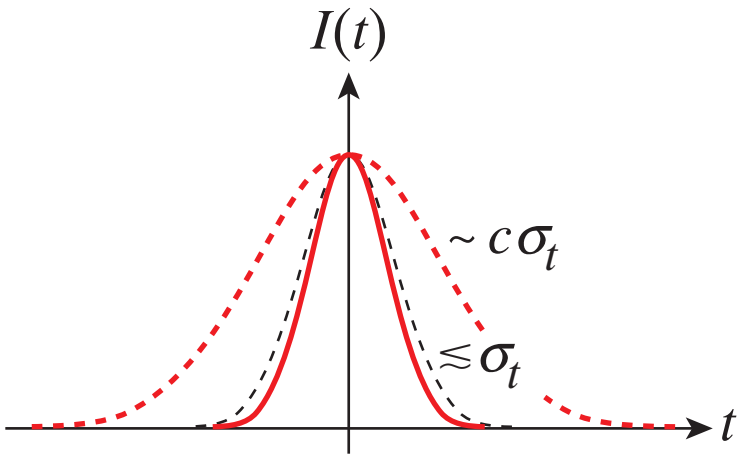
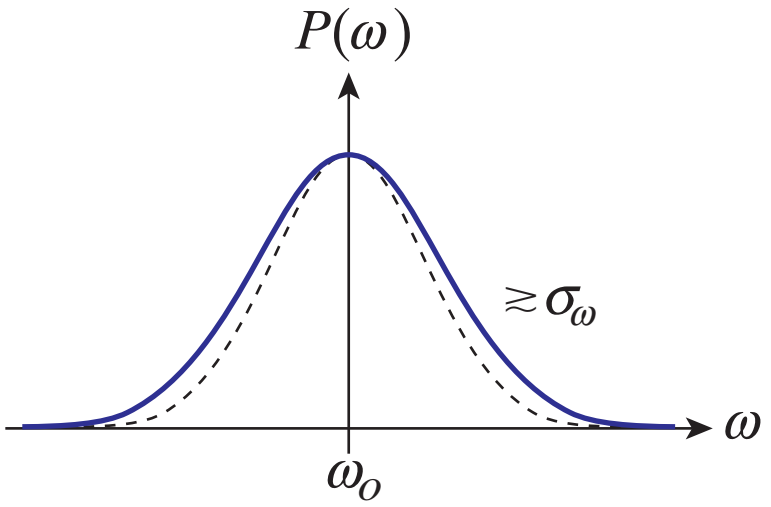
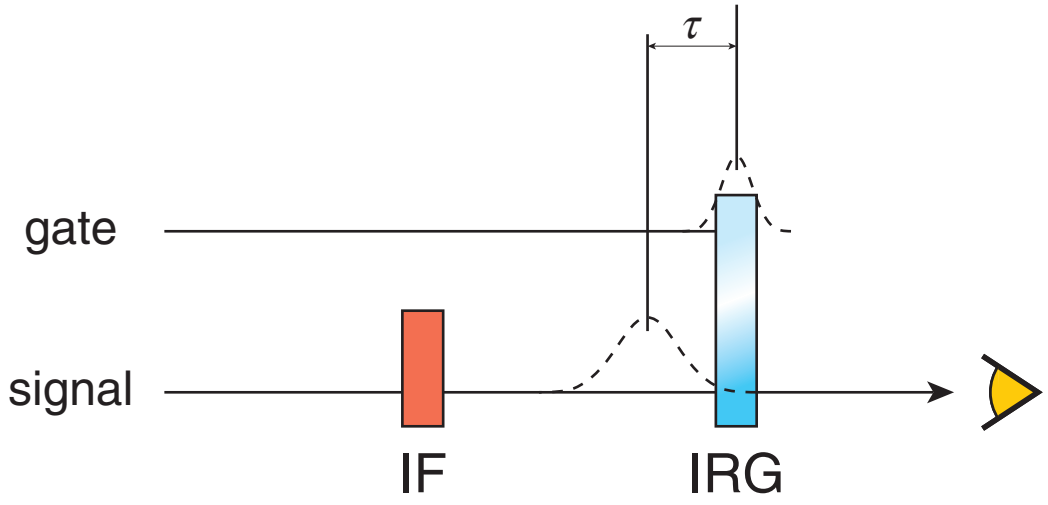
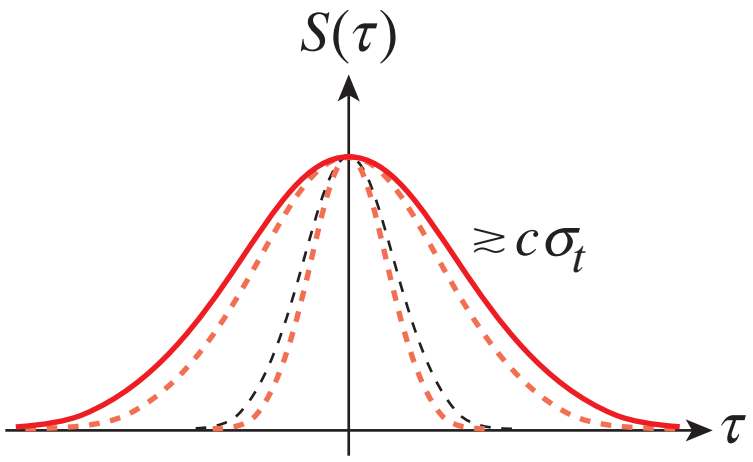




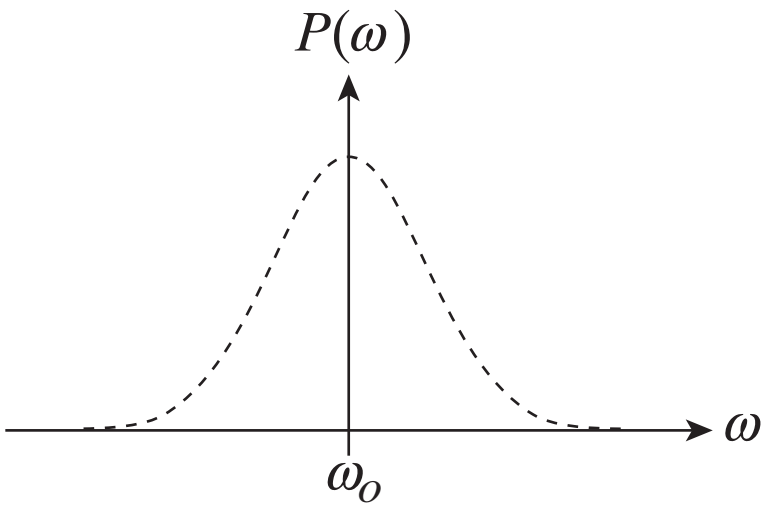


Experiment 1

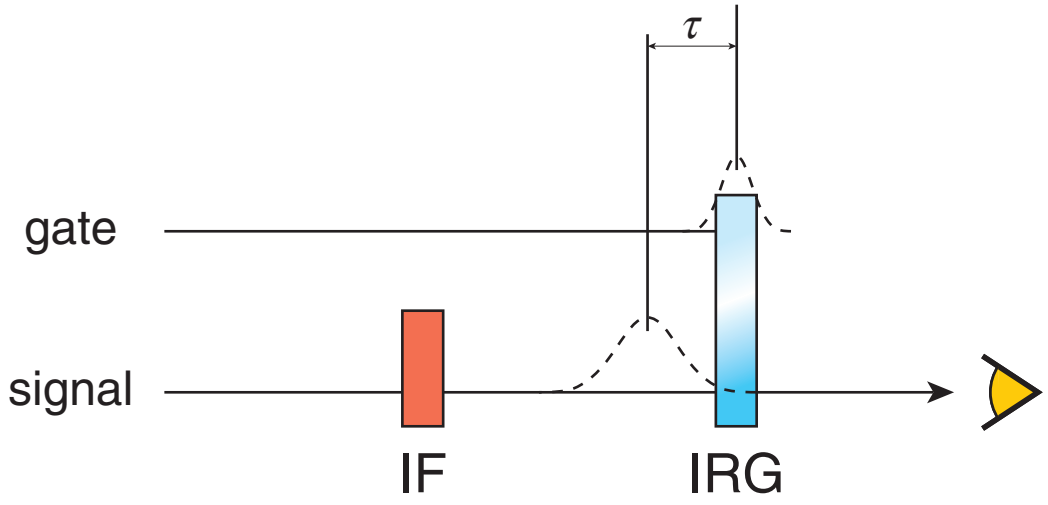


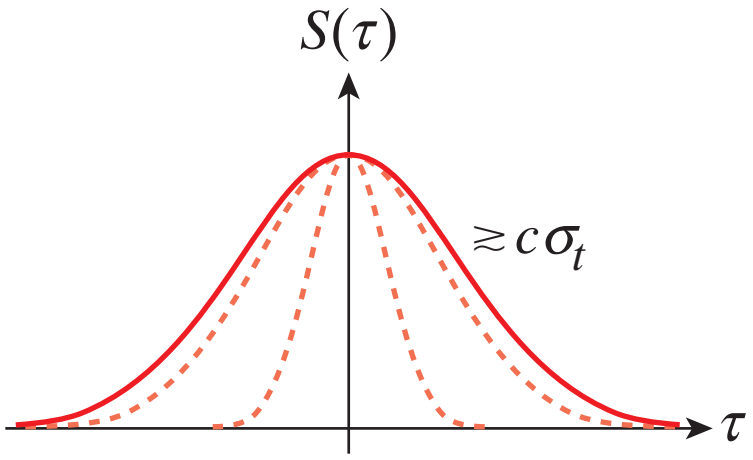


time resolution



Experiment 1





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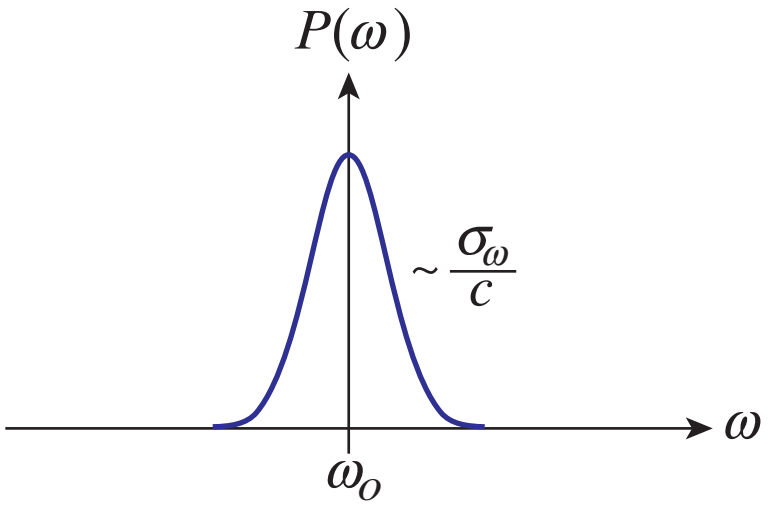
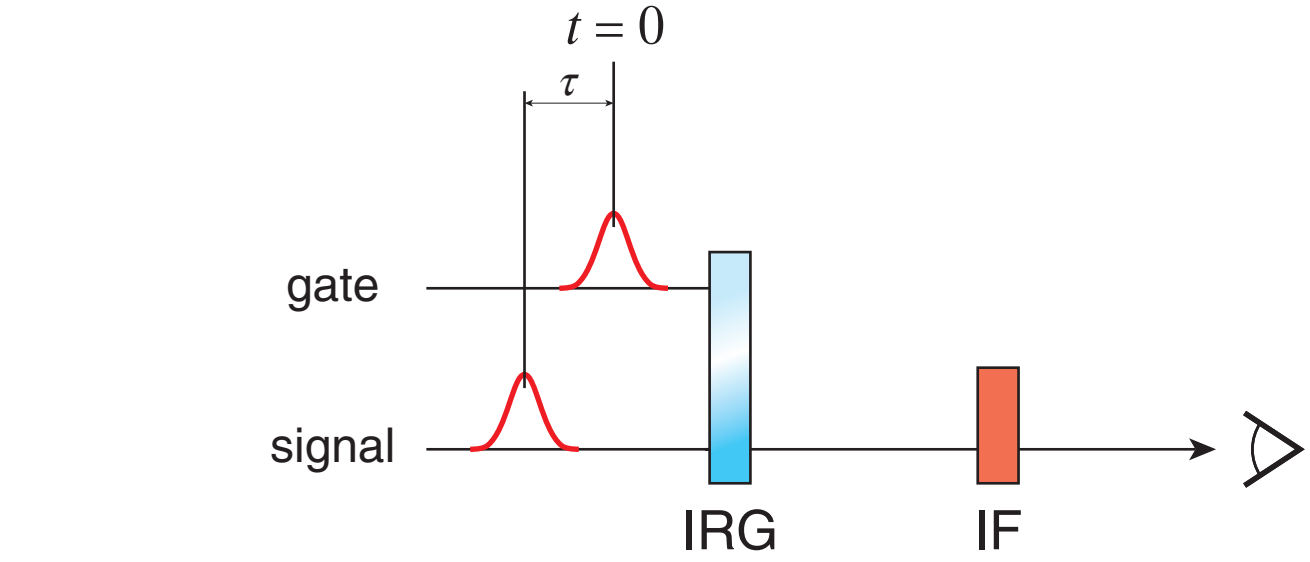
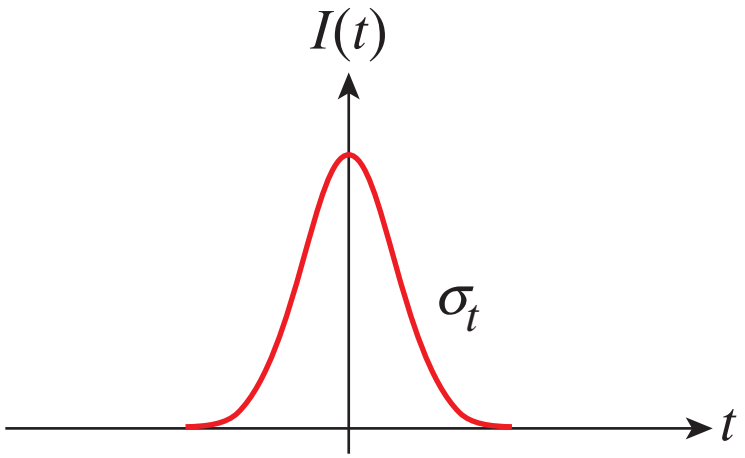
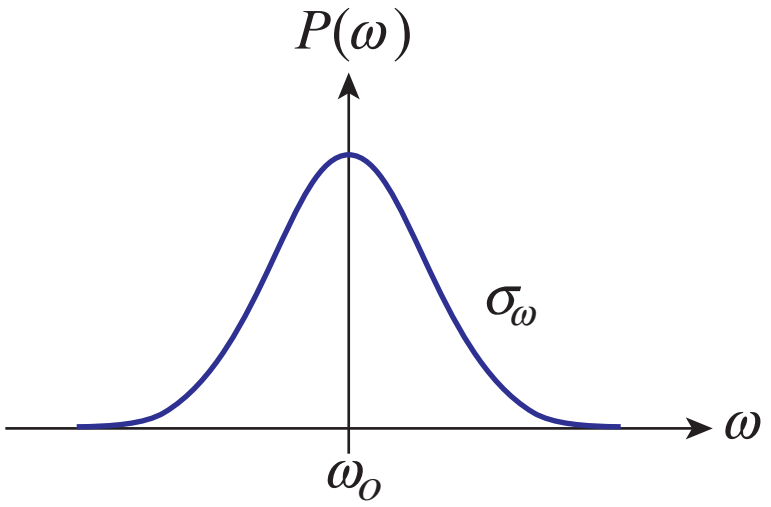


frequency resolution

Experiment 2







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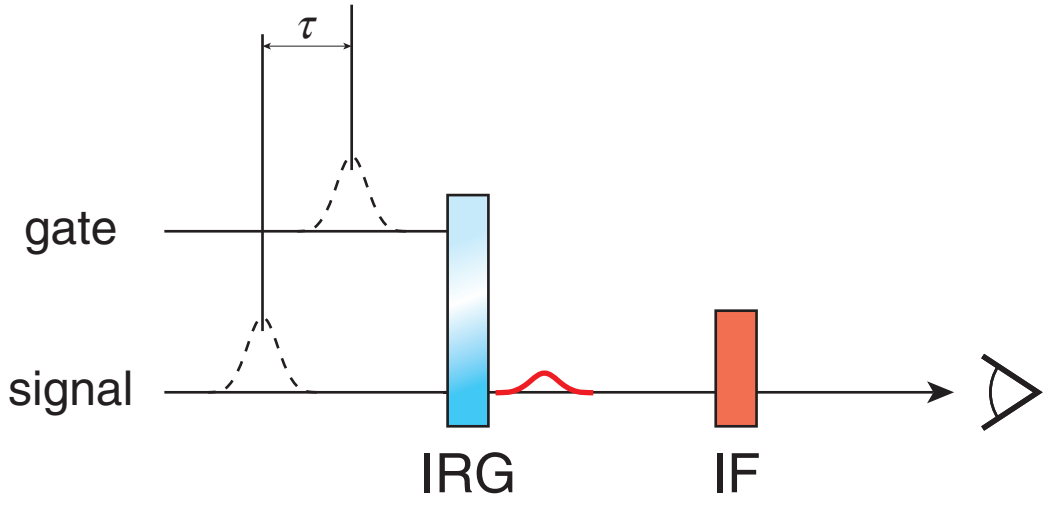

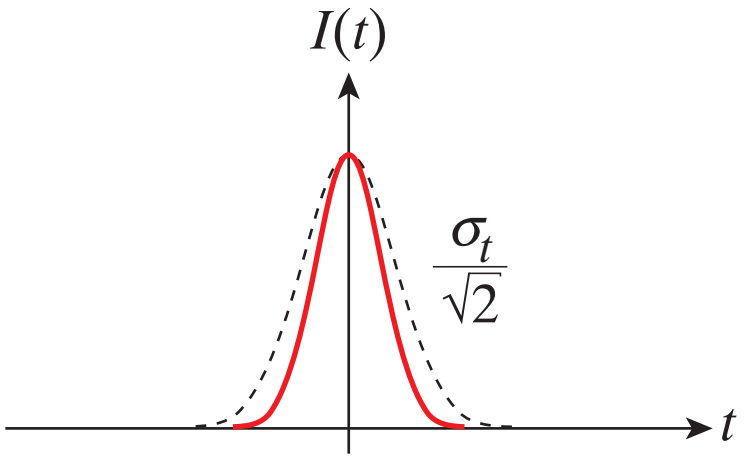

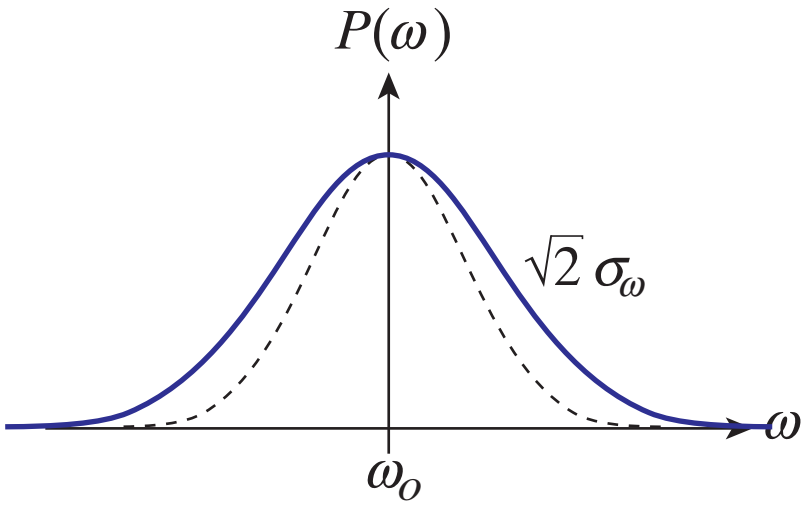

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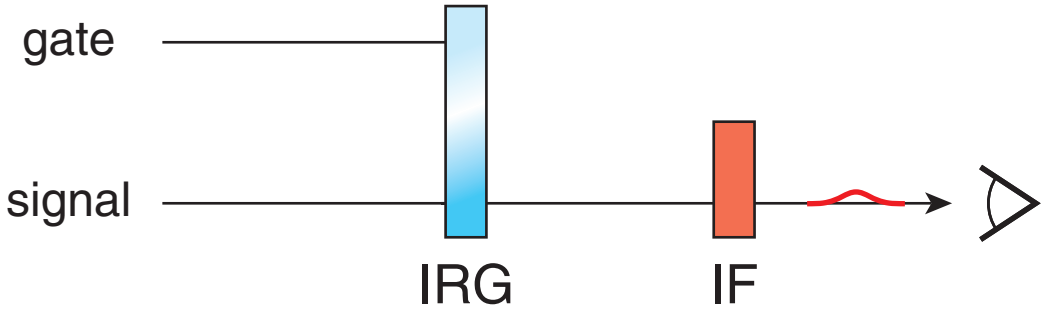

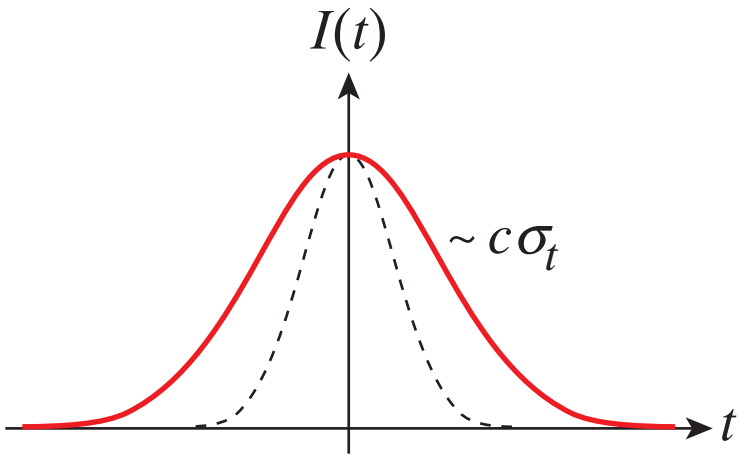

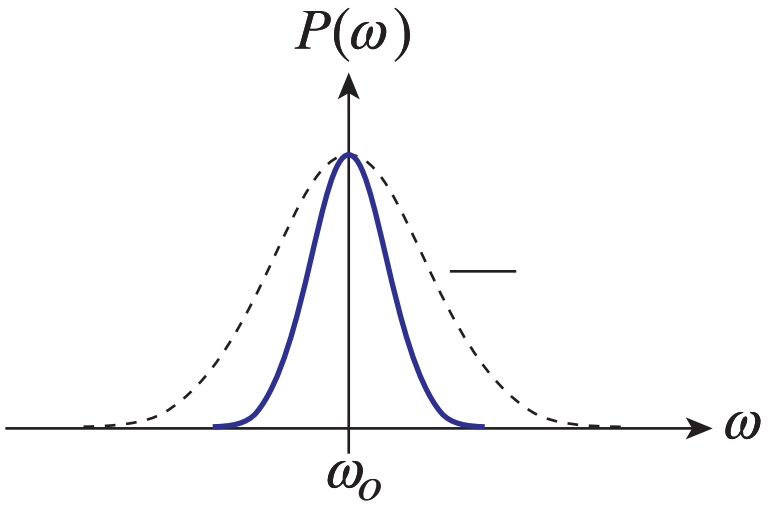

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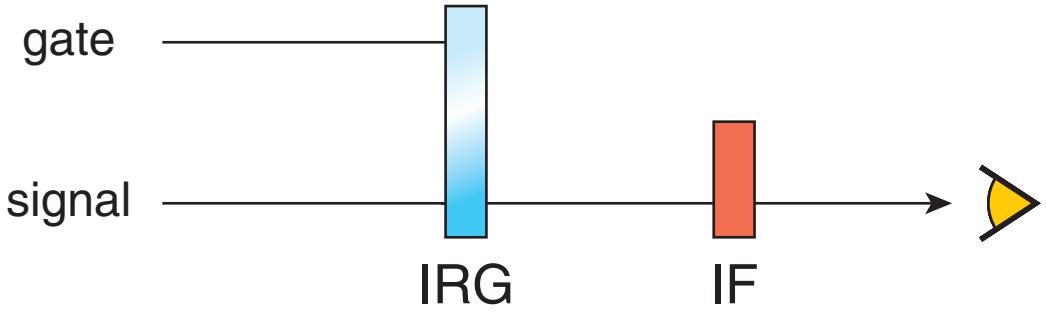
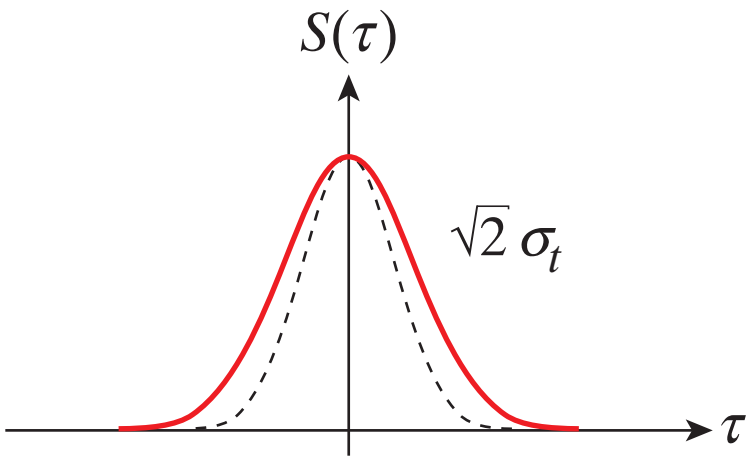
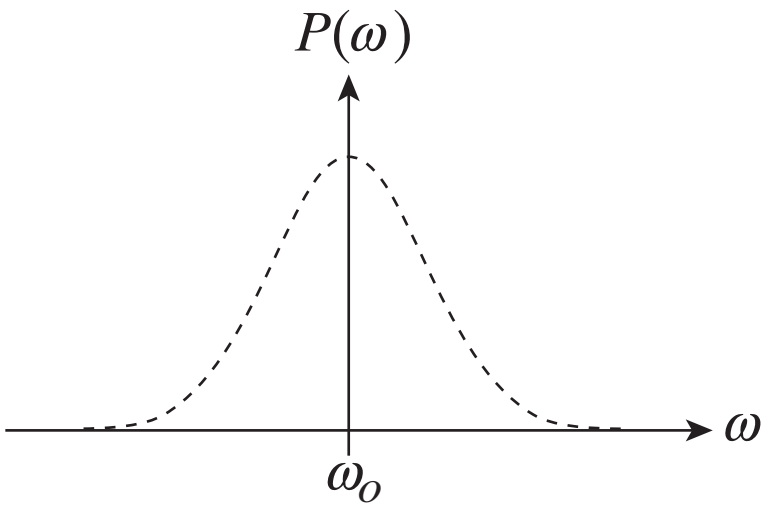




time resolution



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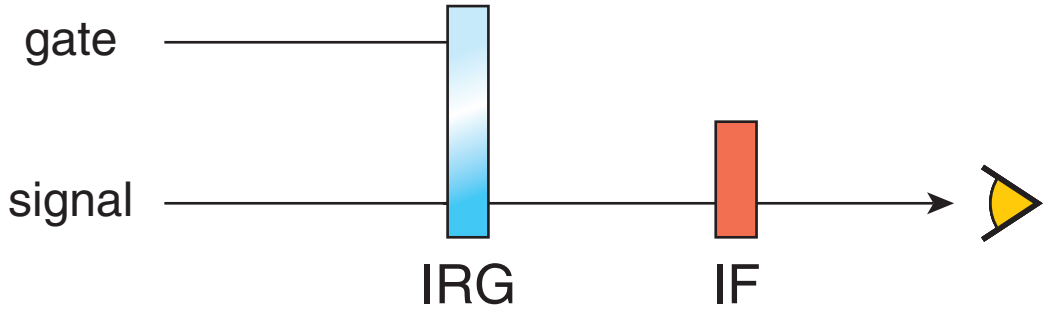

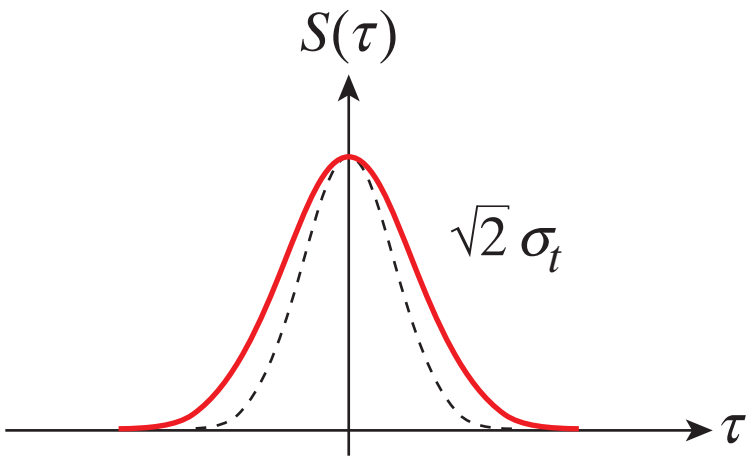

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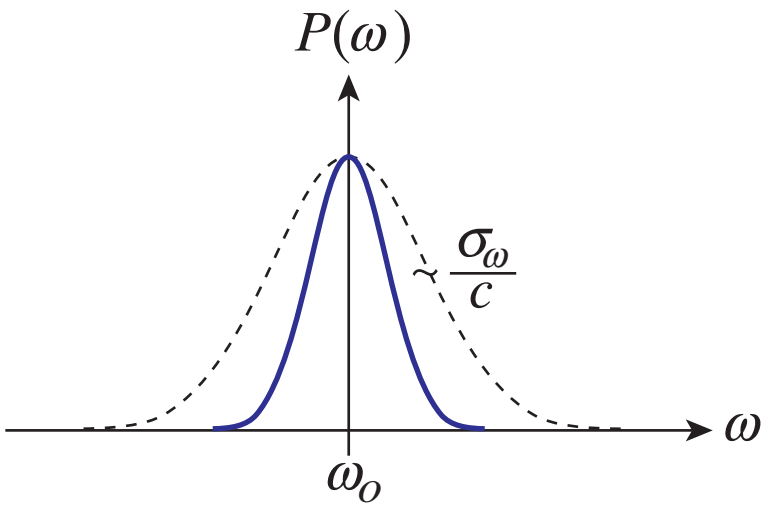
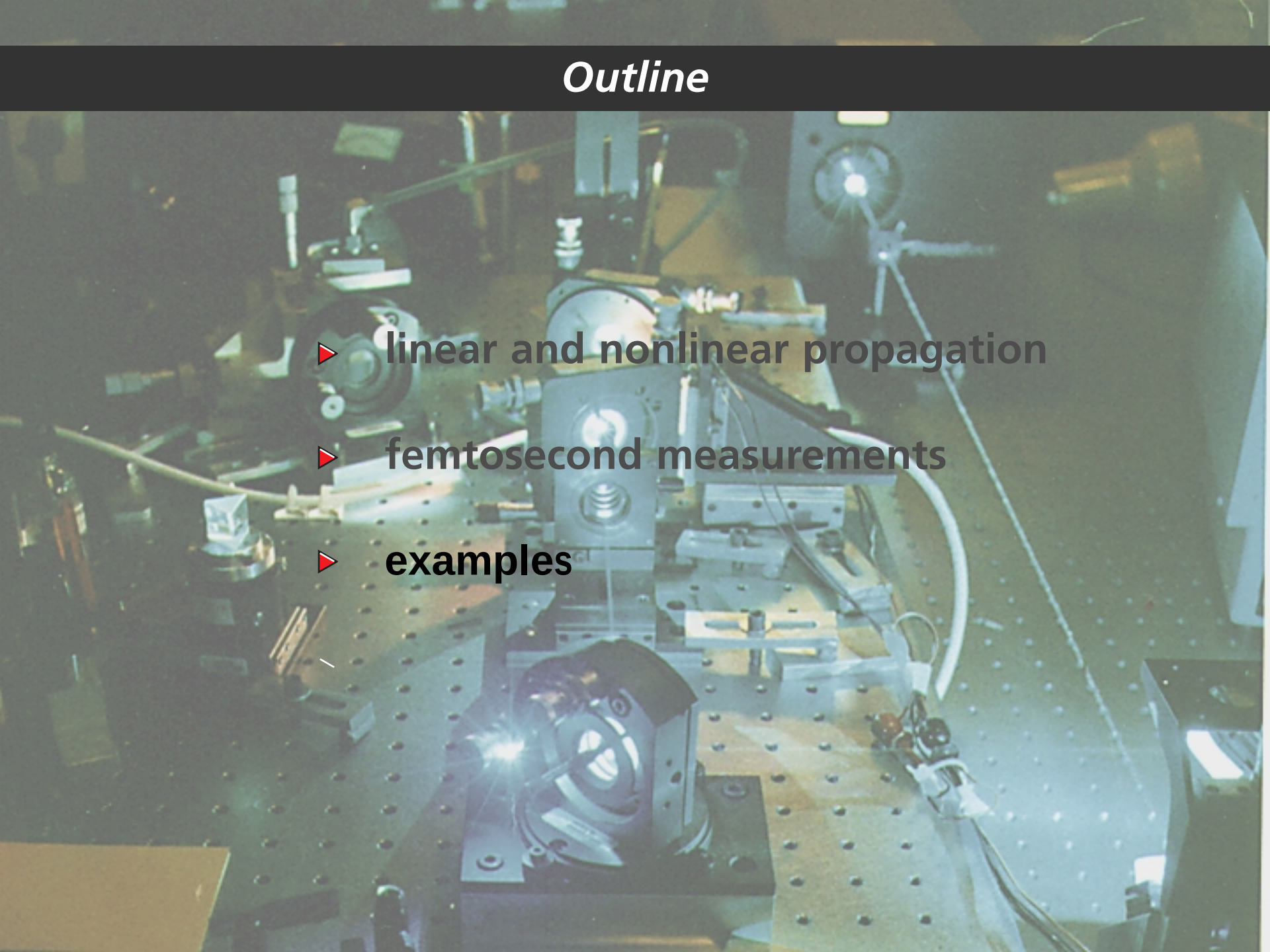


spectral resolution

Outline



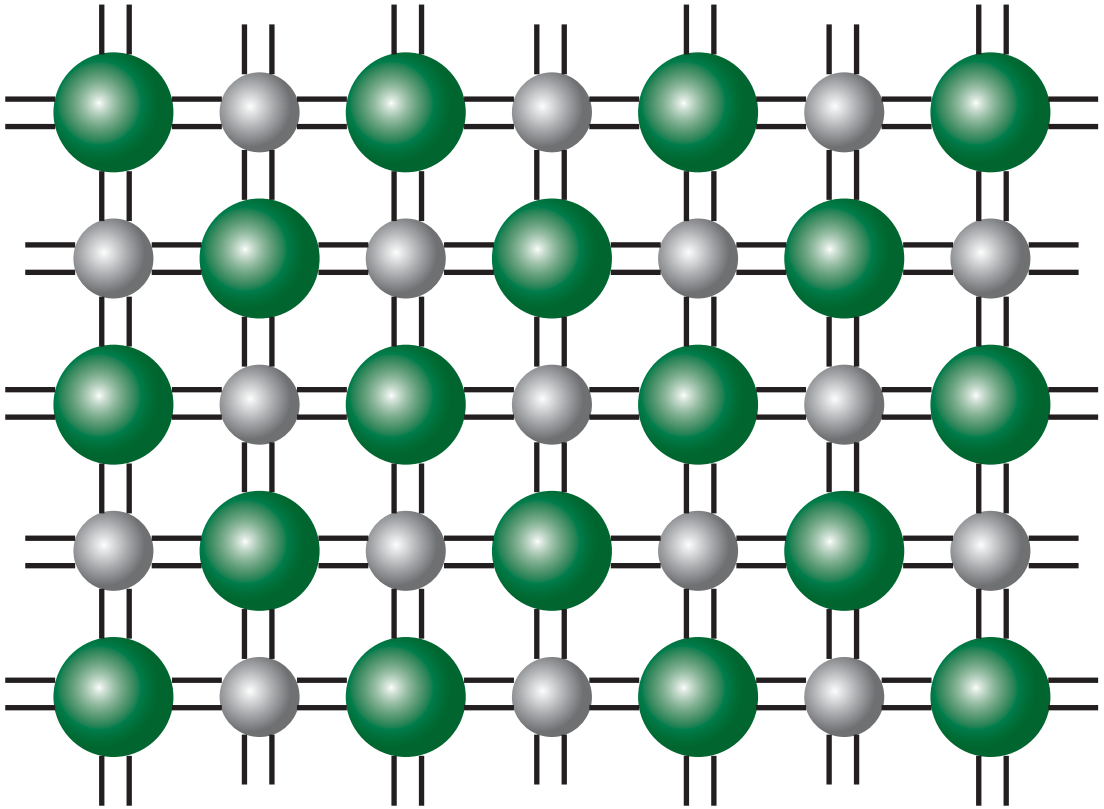
- linear and nonlinear propagation
- femtosecond measurements
- **examples**

Introduction

short laser pulses can drive structural transitions

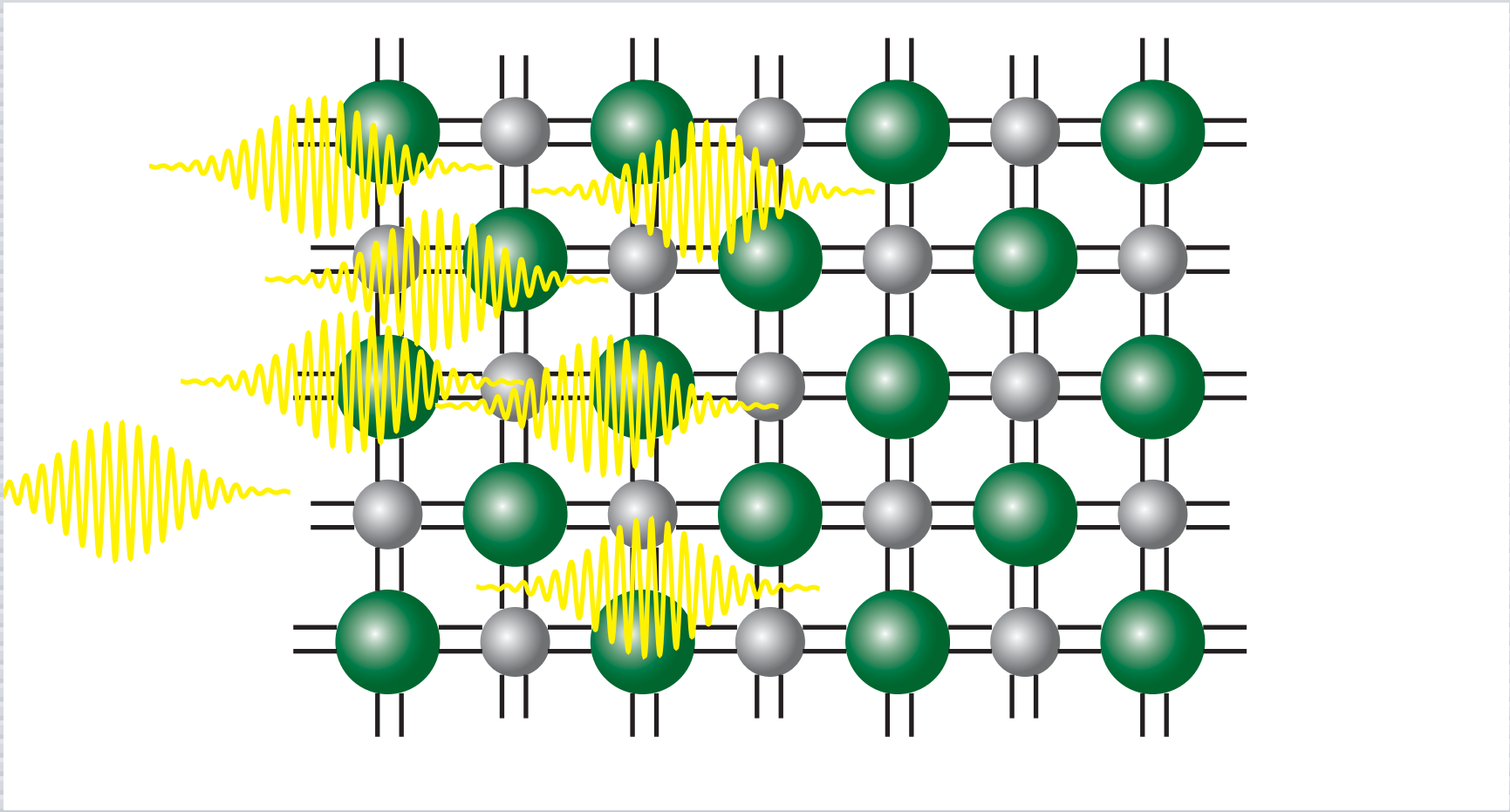
Introduction

how do femtosecond laser pulses alter a solid?

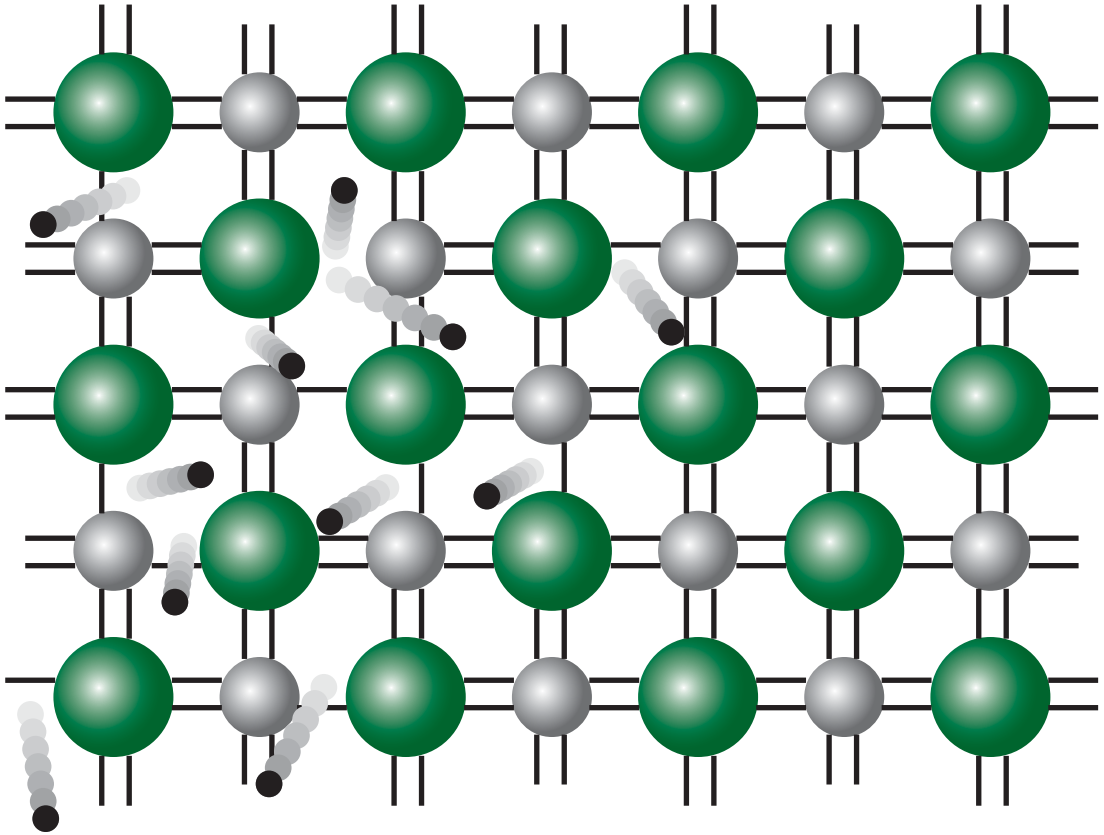


Introduction

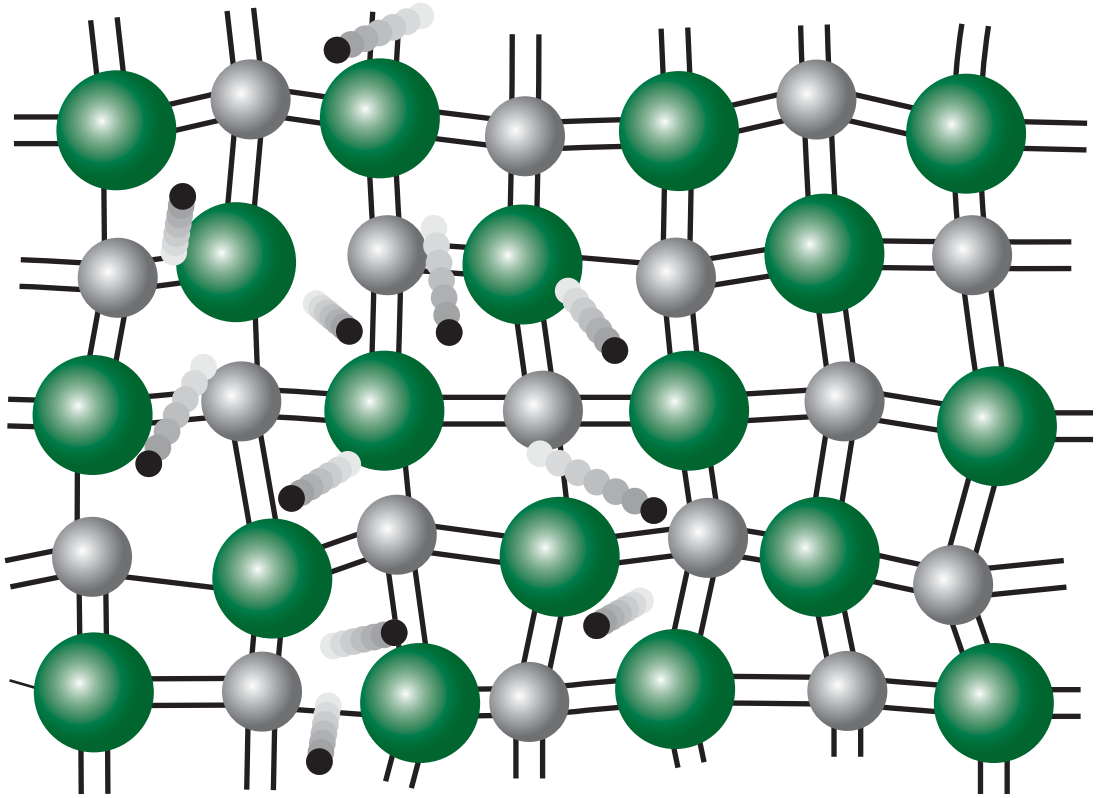
photons excite valence electrons…



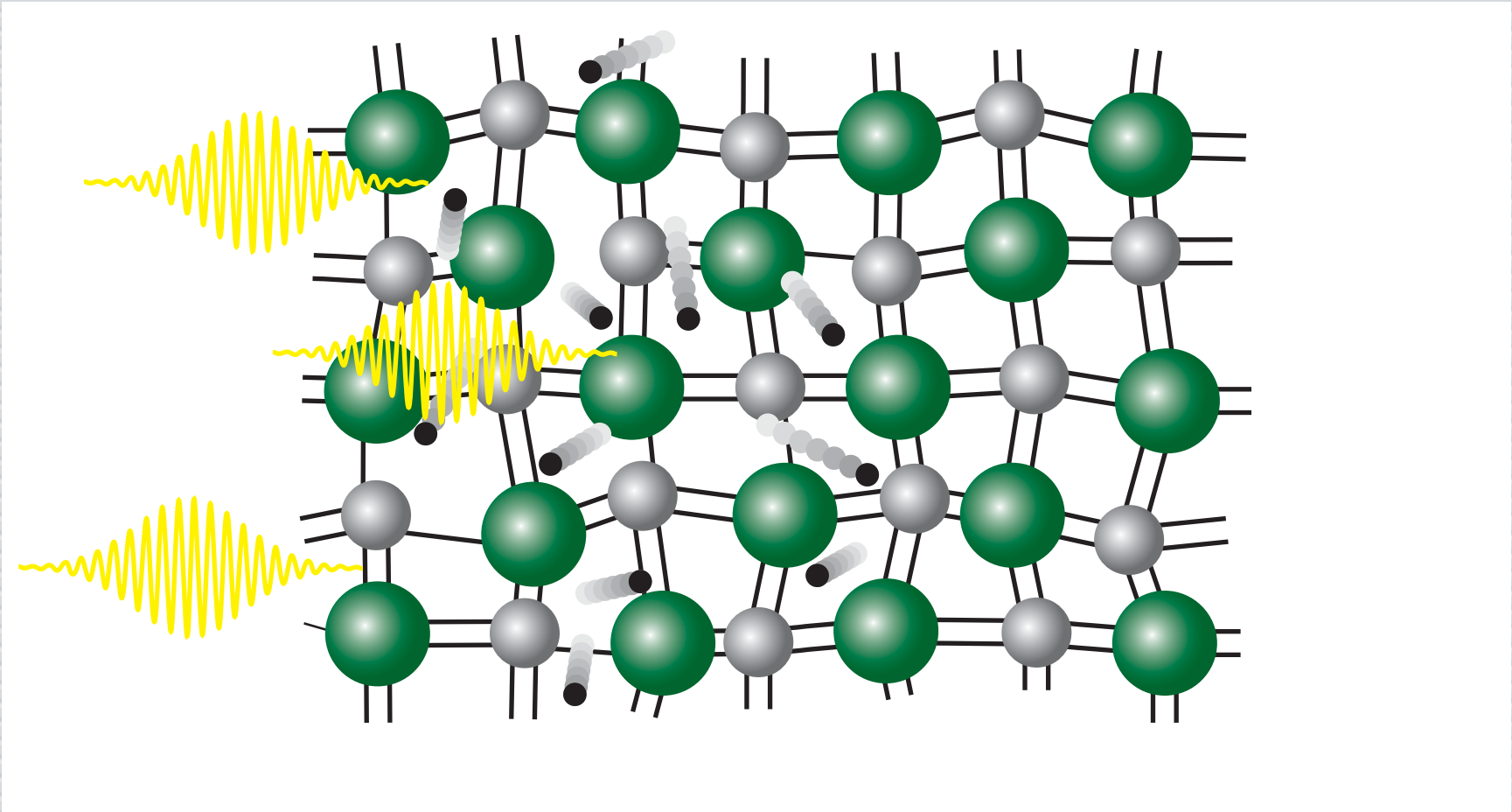
… and create free electrons…



… causing electronic and structural changes…

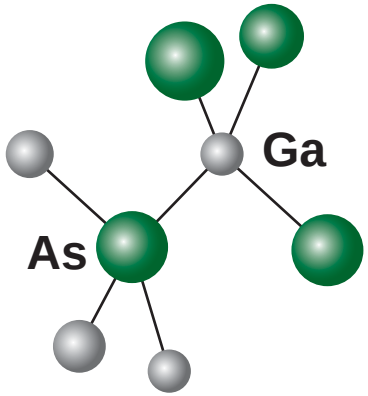


… which we detect with a second laser pulse



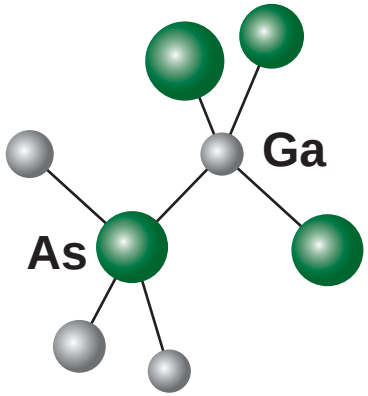
Introduction

structure

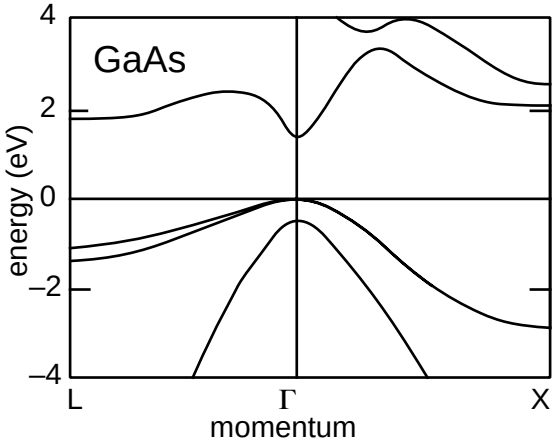


Introduction

structure

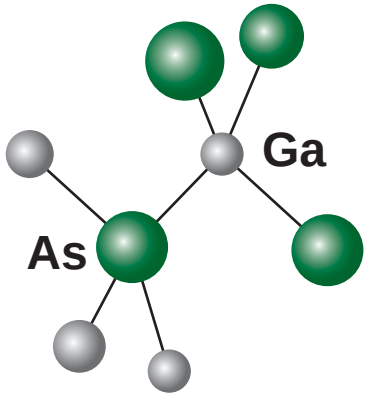


band structure

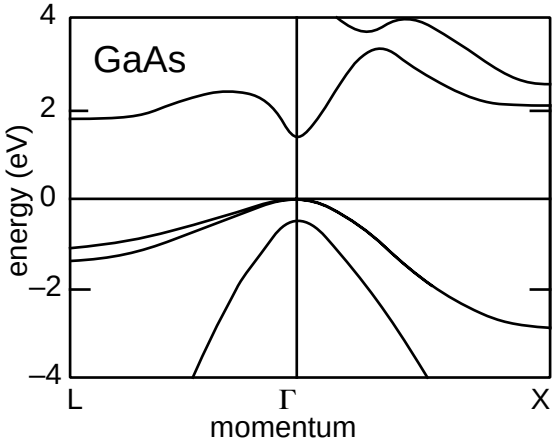


Introduction

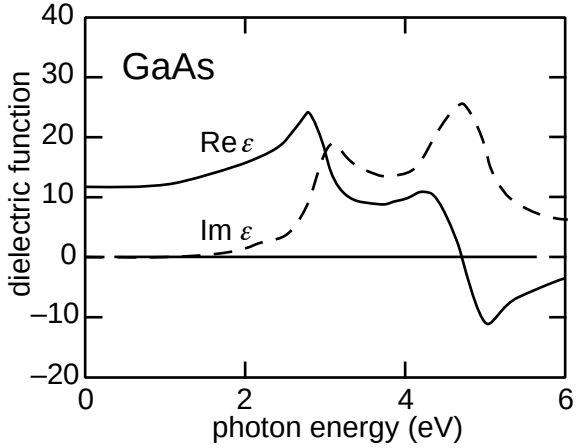
structure



band structure

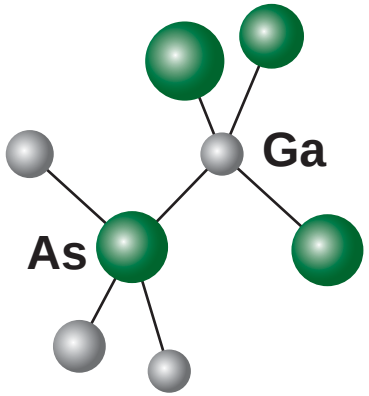


dielectric function

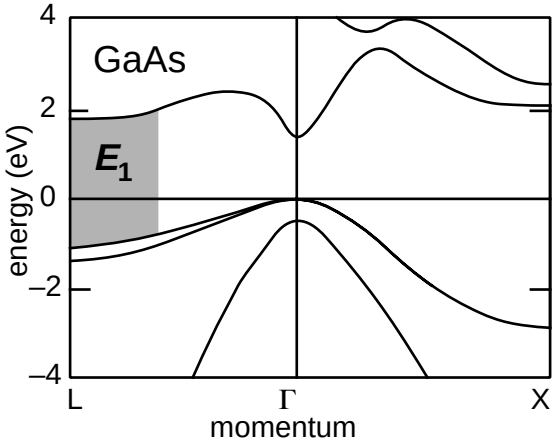


Introduction

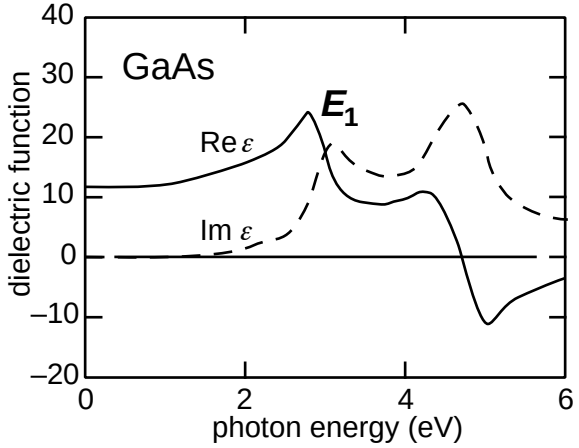
structure



band structure

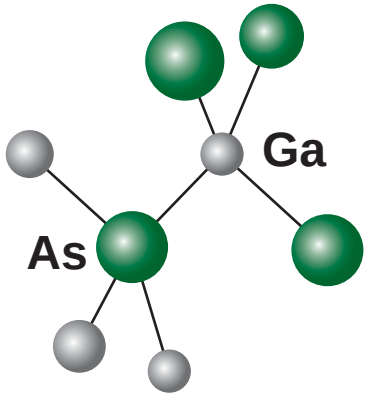


dielectric function



Introduction

structure



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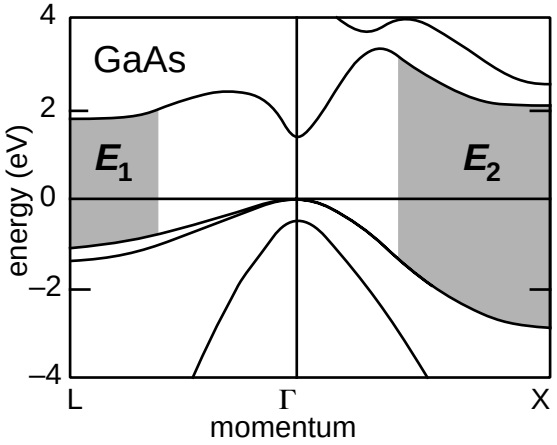

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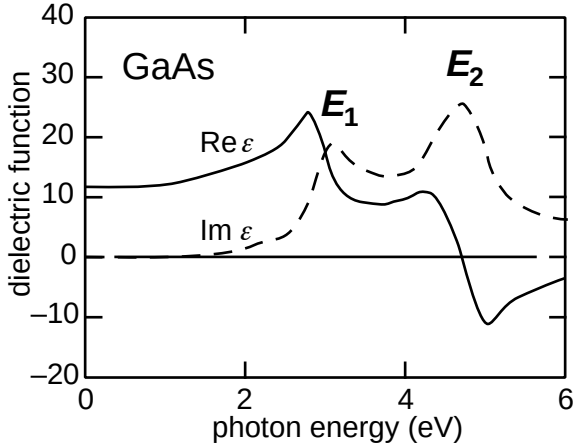
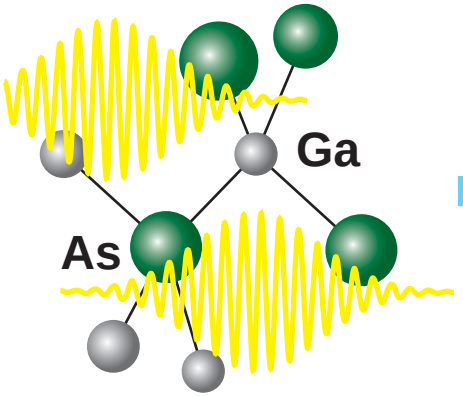
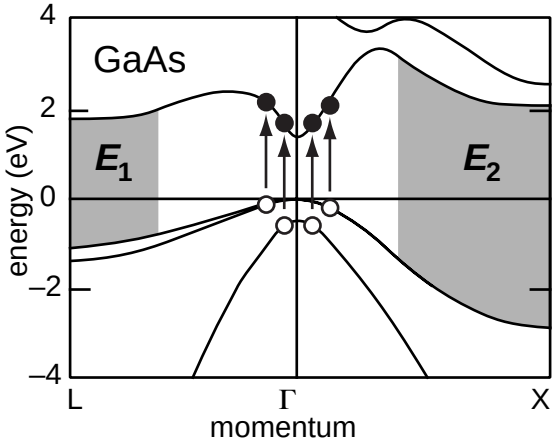


Introduction

structure



band structure



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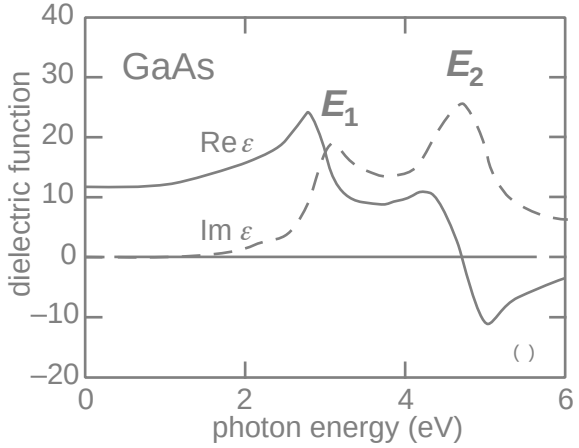
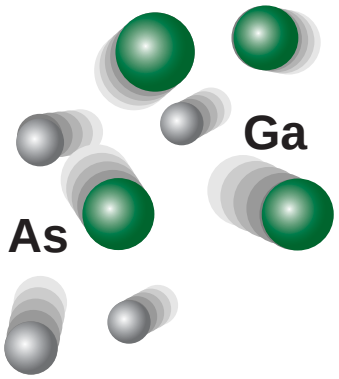


Introduction

structure



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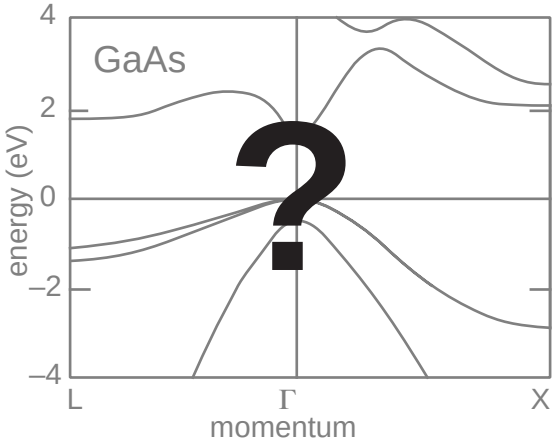

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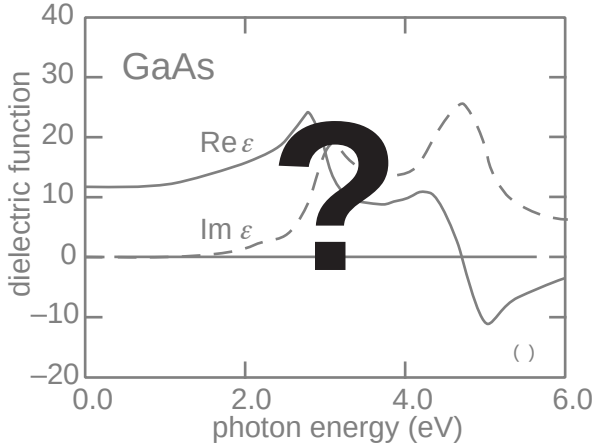

Introduction

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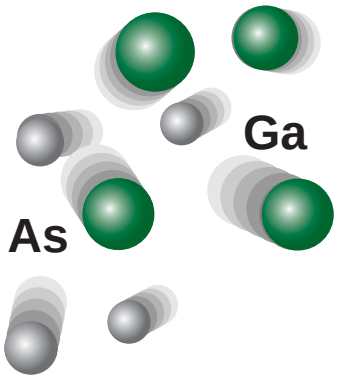

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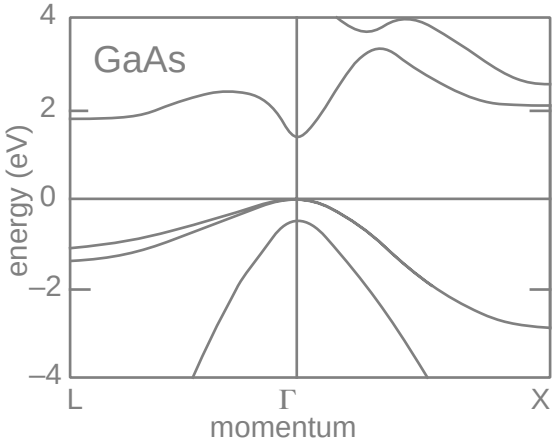

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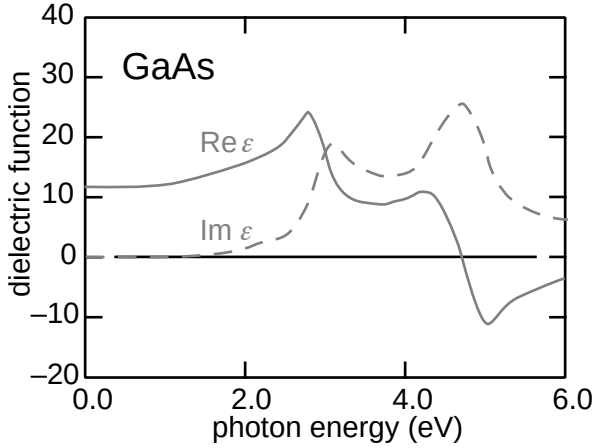
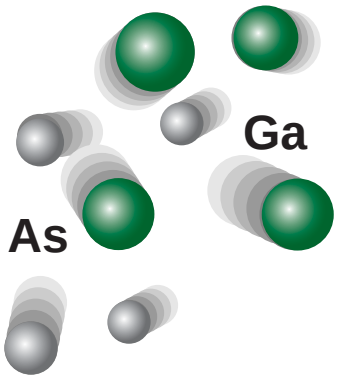


Introduction

structure



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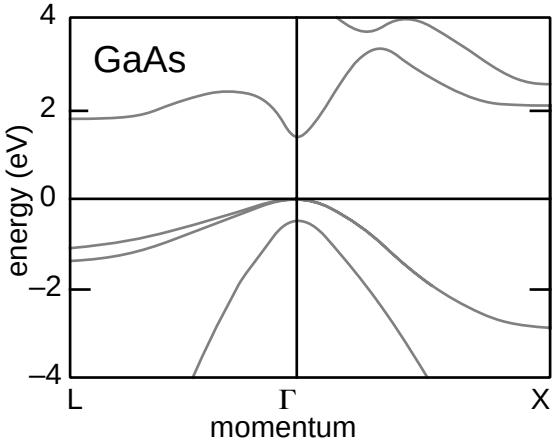

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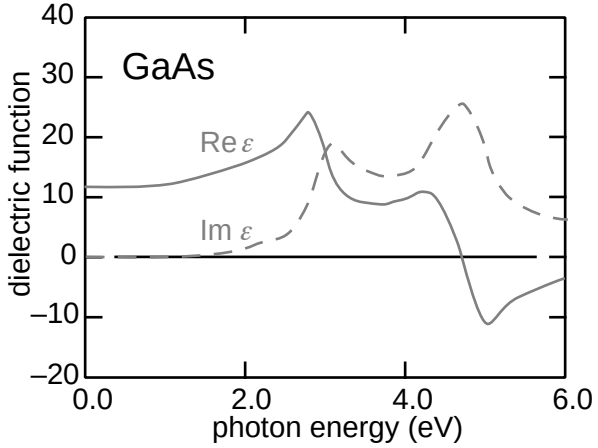

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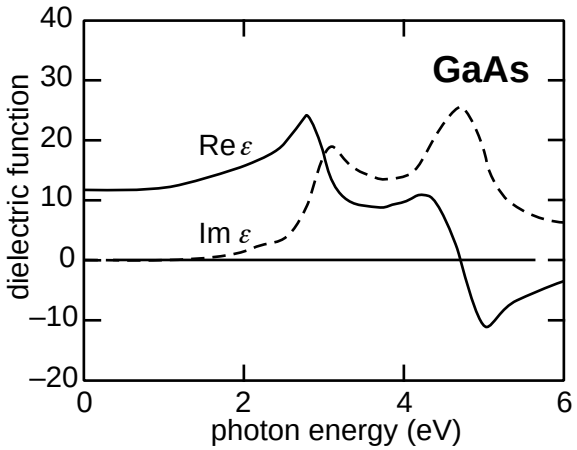

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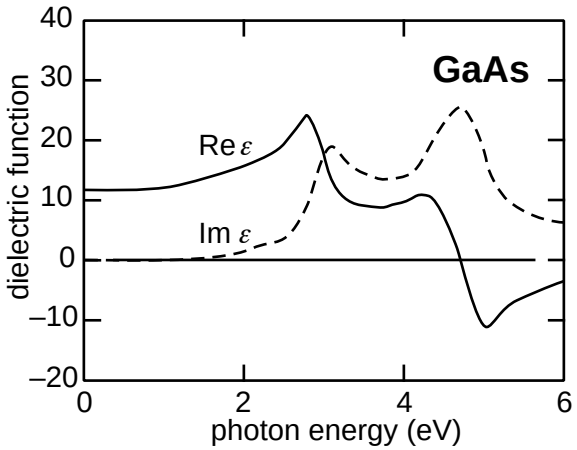
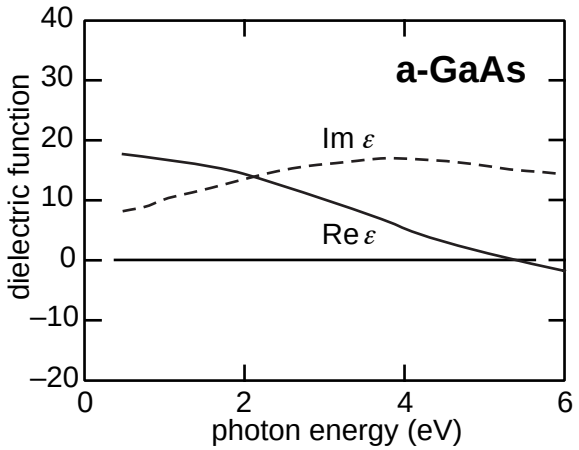
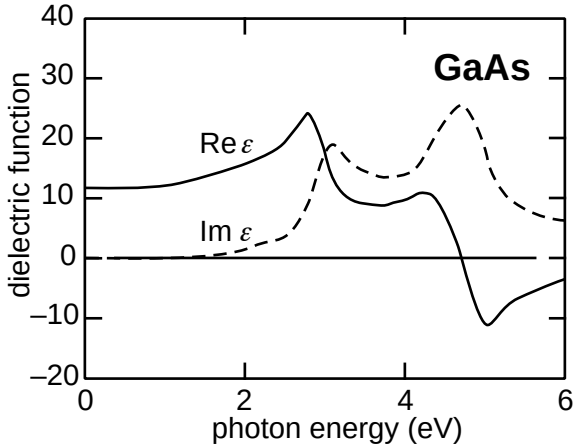
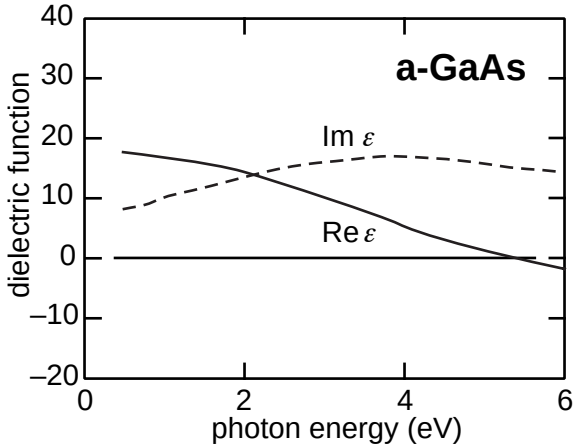
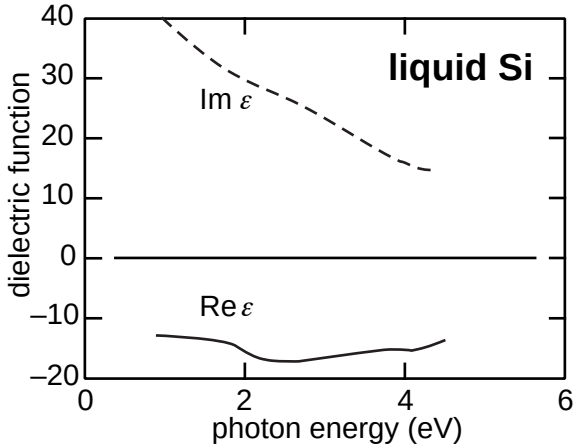




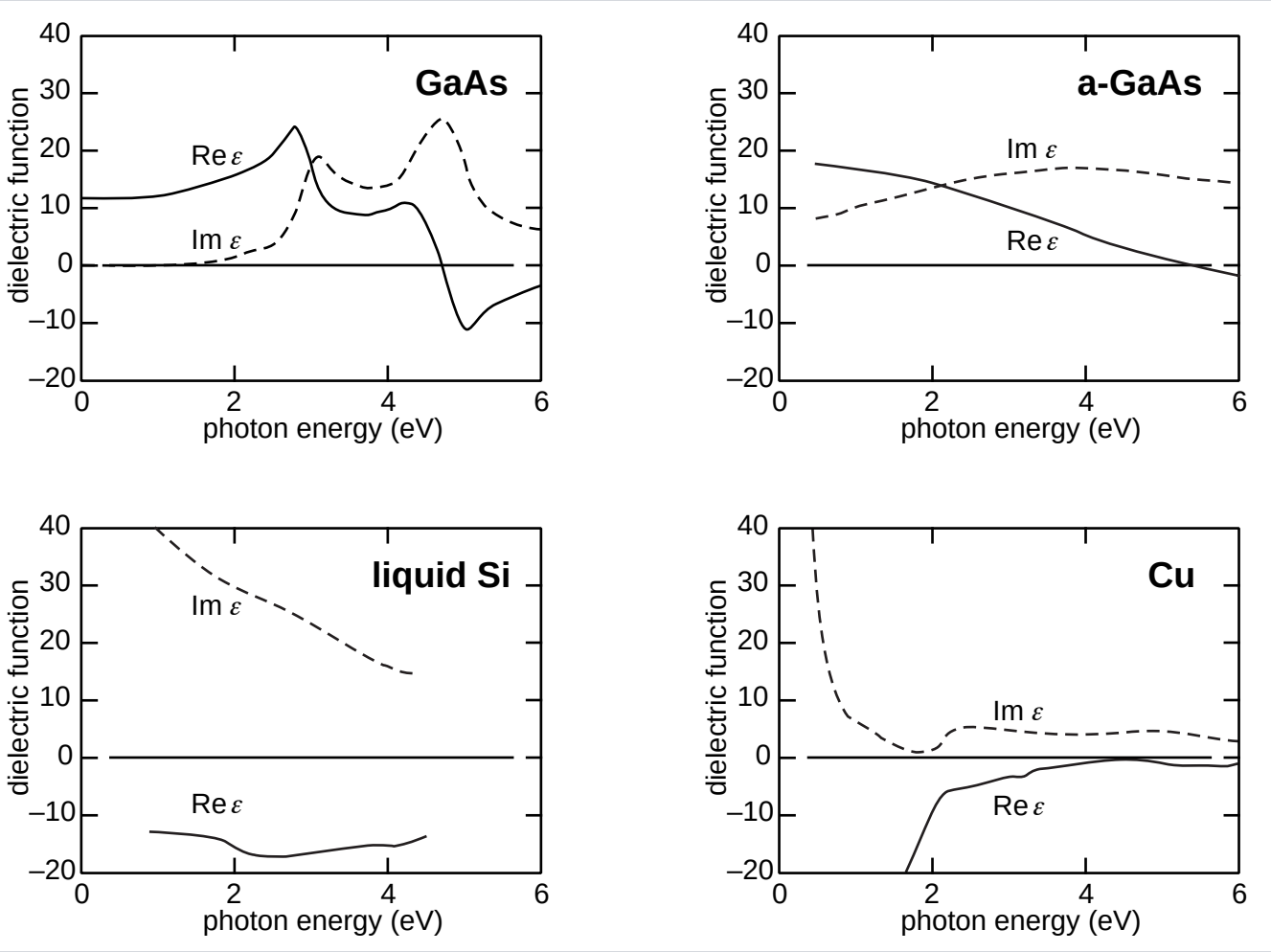
Introduction





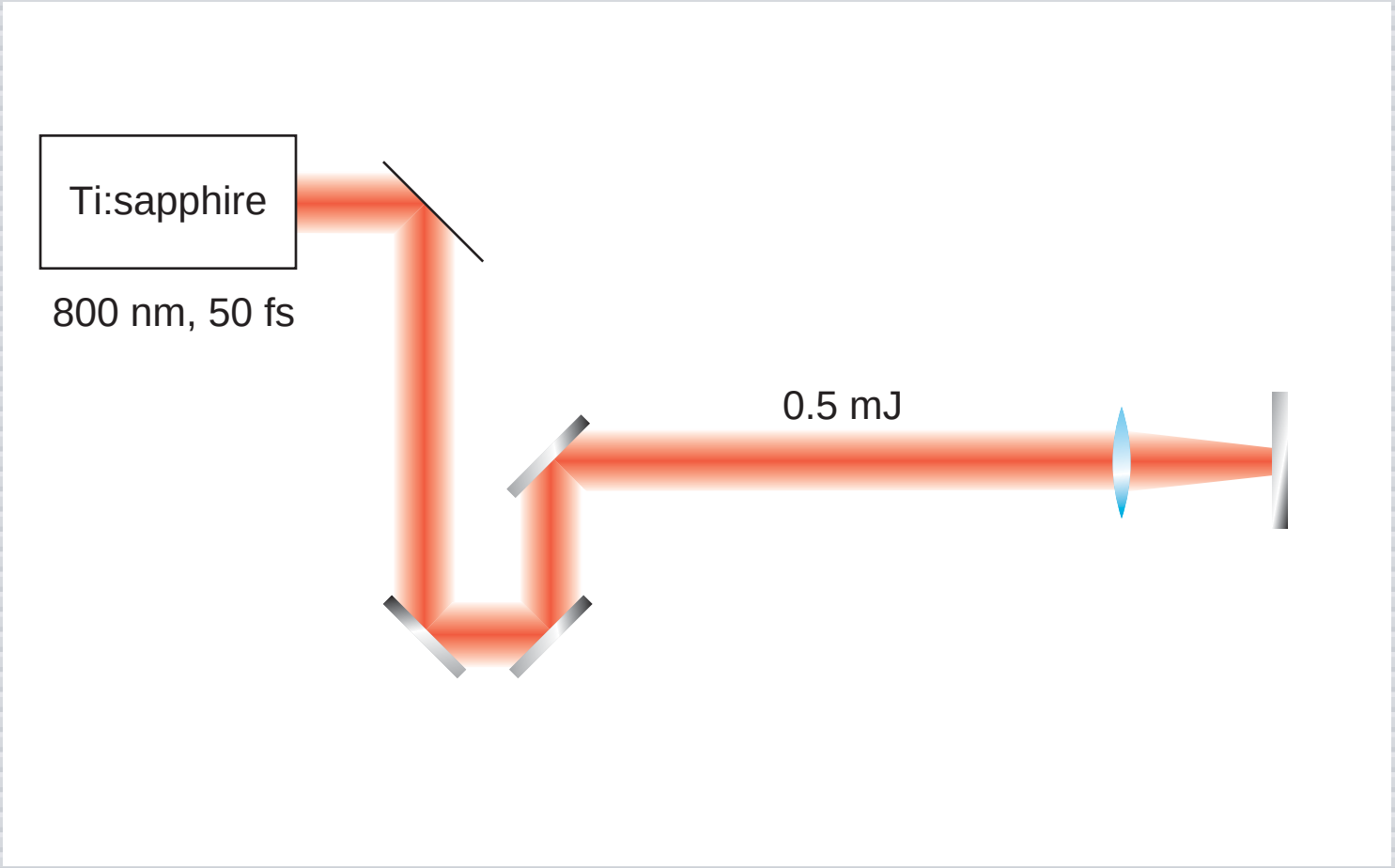


Introduction



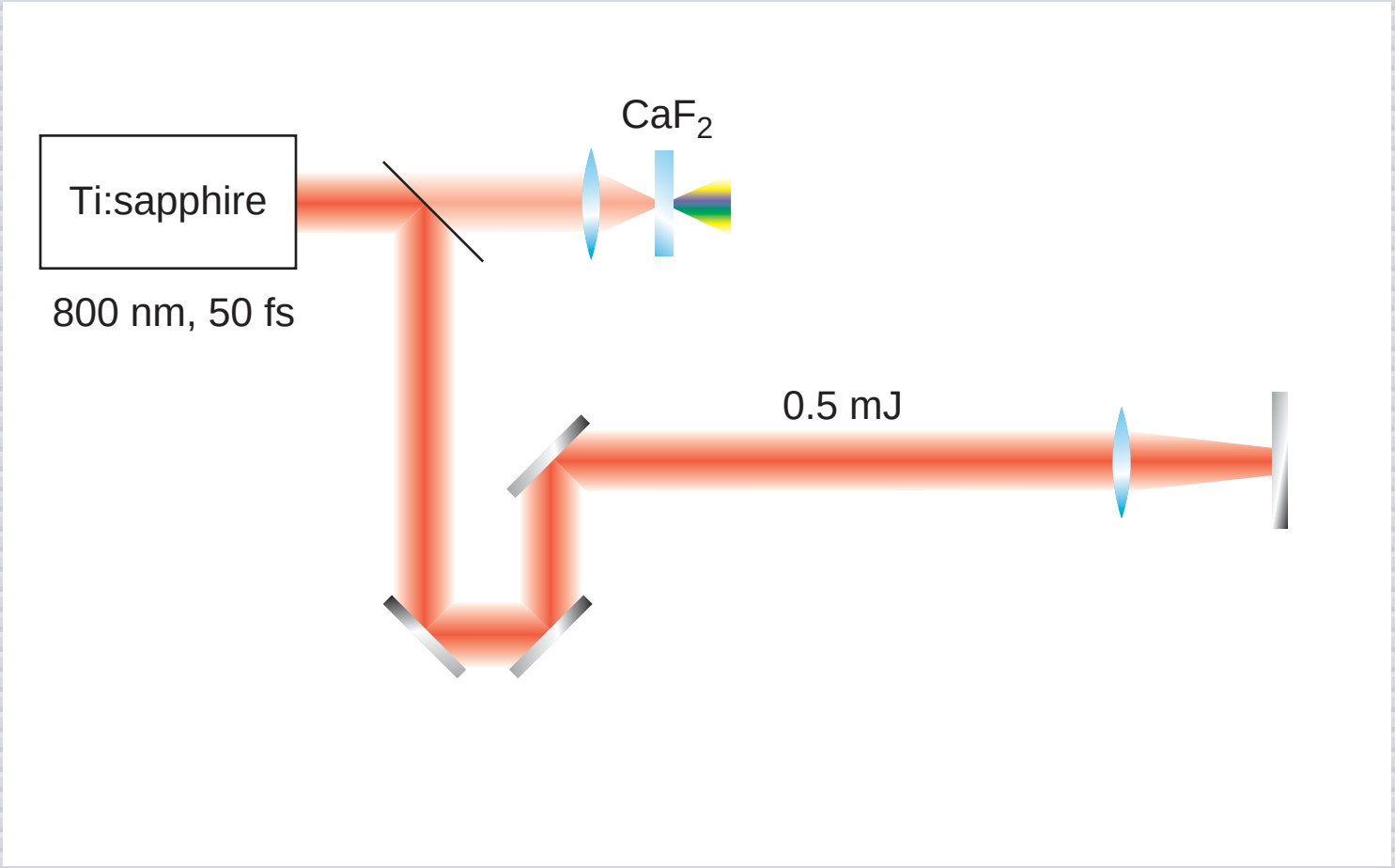
Technique

broadband time-resolved ellipsometry

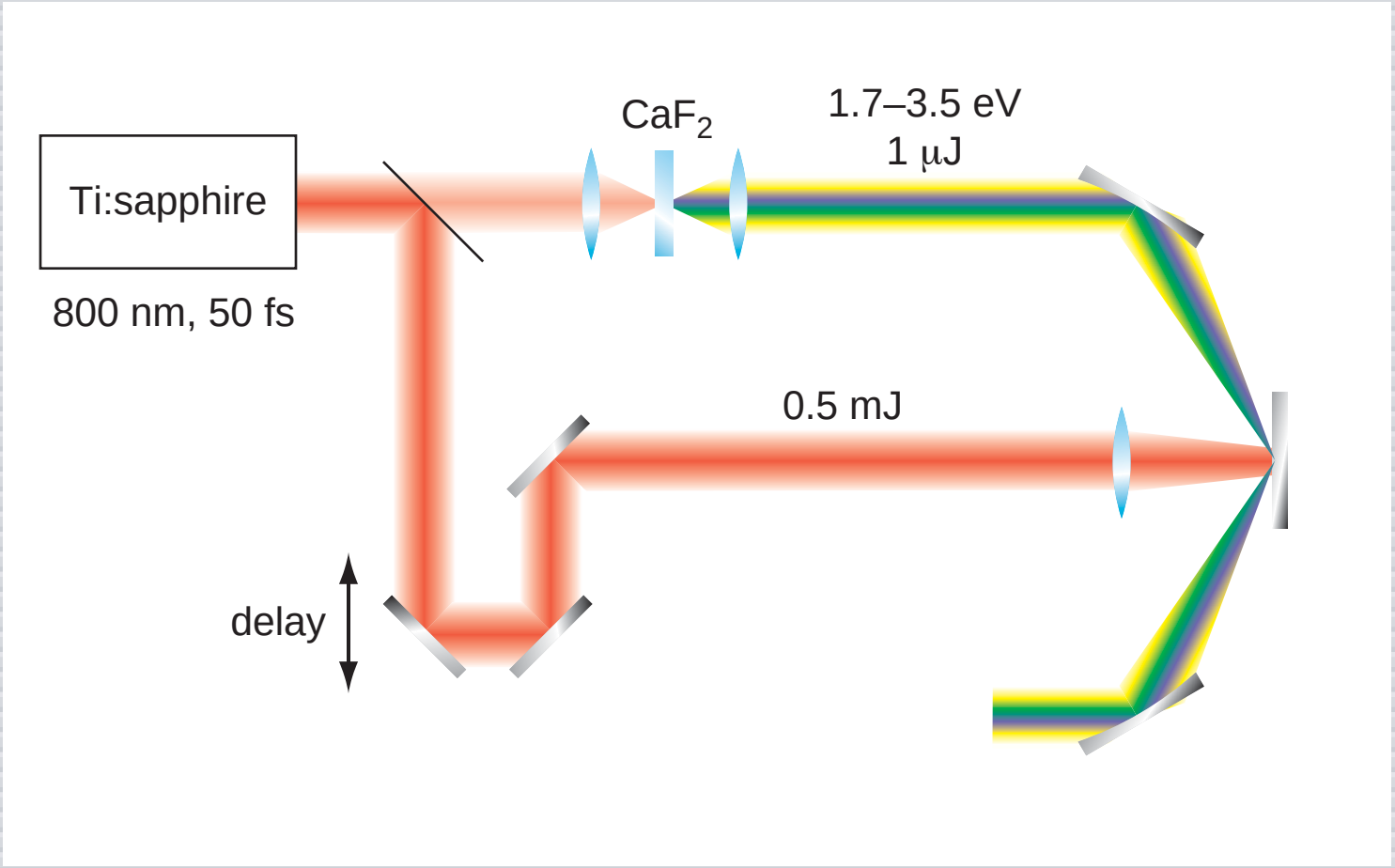


Technique

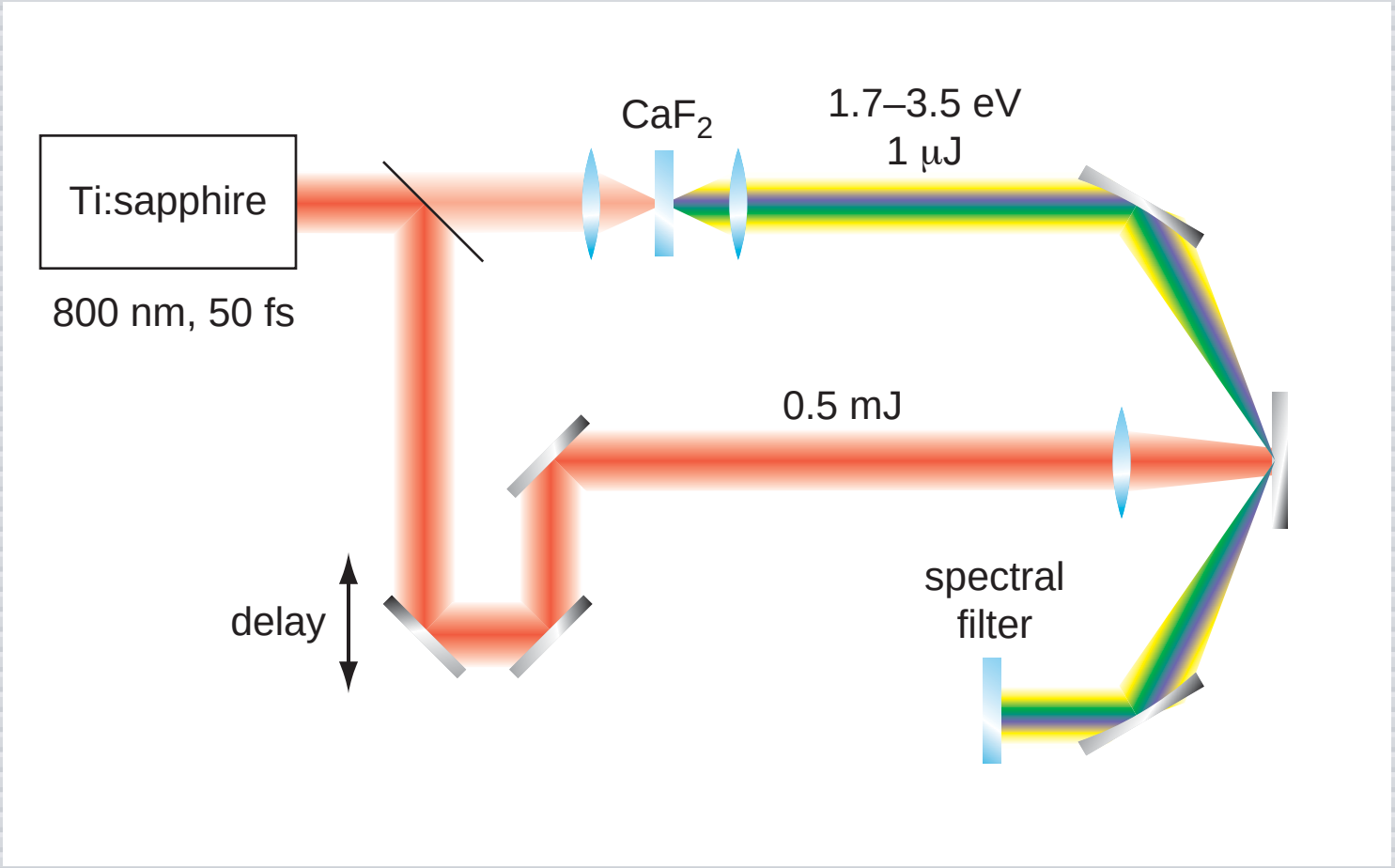
broadband time-resolved ellipsometry



broadband time-resolved ellipsometry

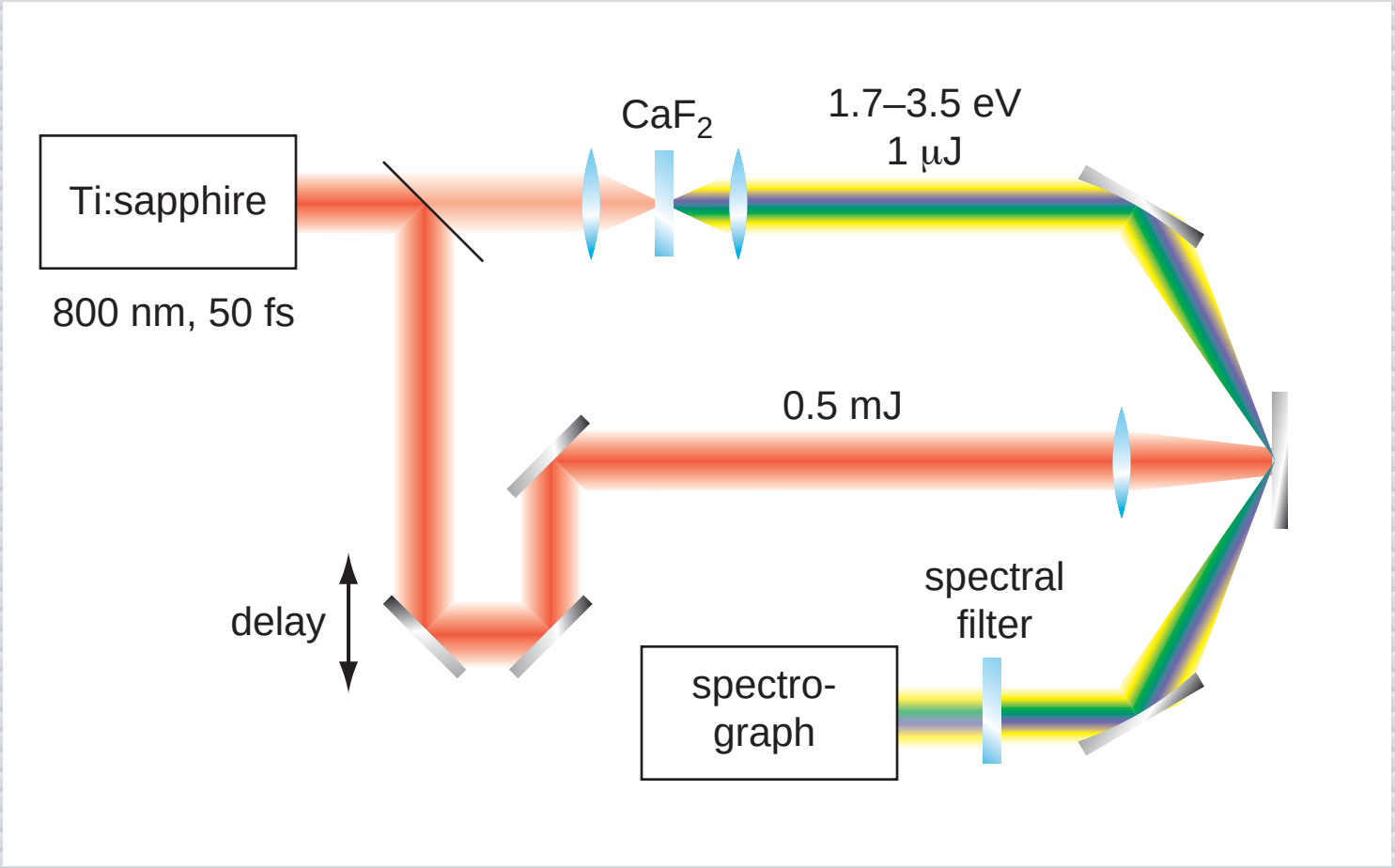


broadband time-resolved ellipsometry

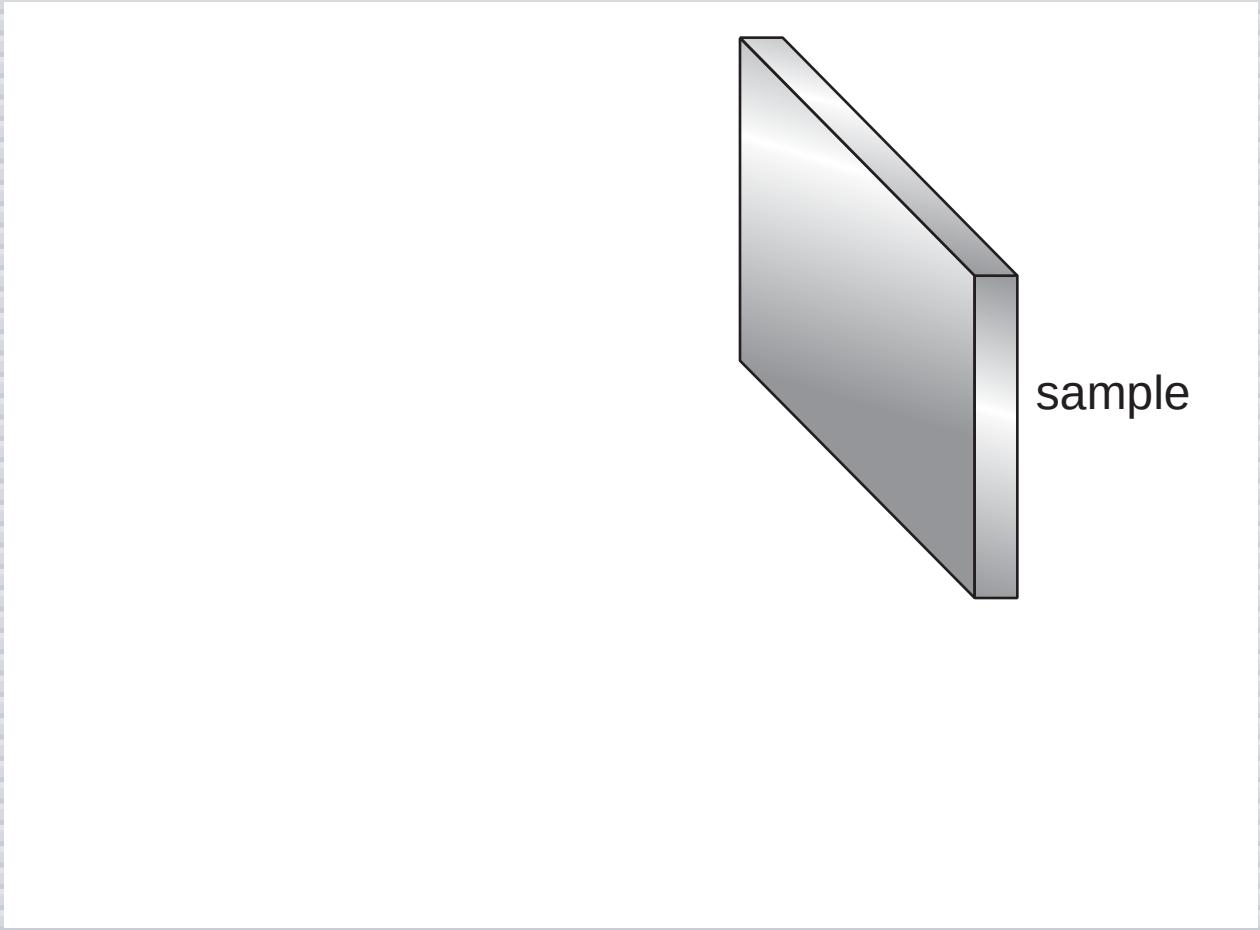


Technique

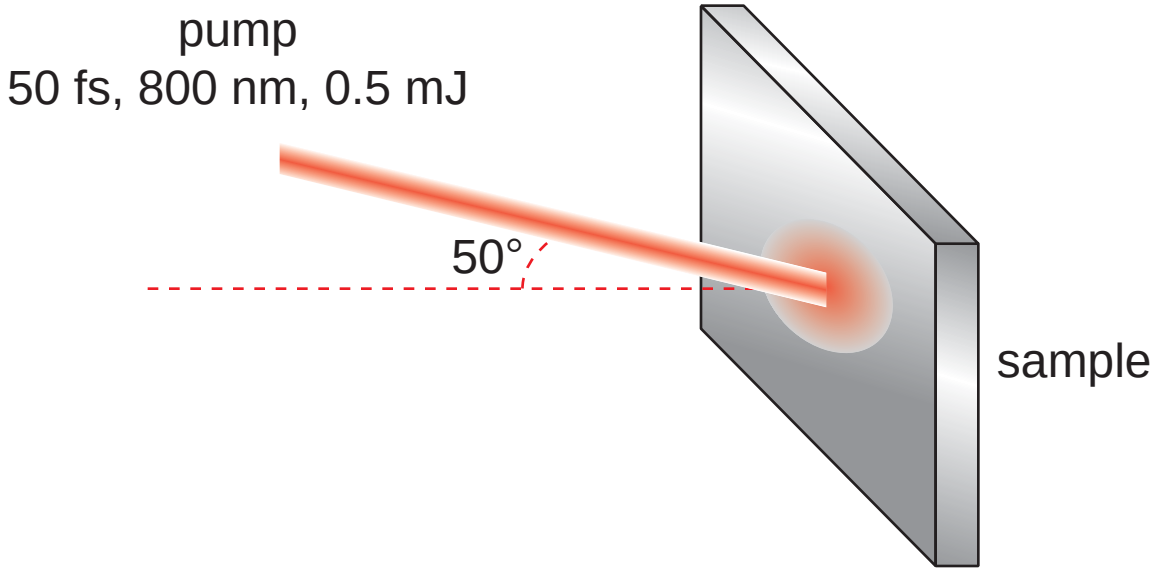
broadband time-resolved ellipsometry



broadband time-resolved ellipsometry

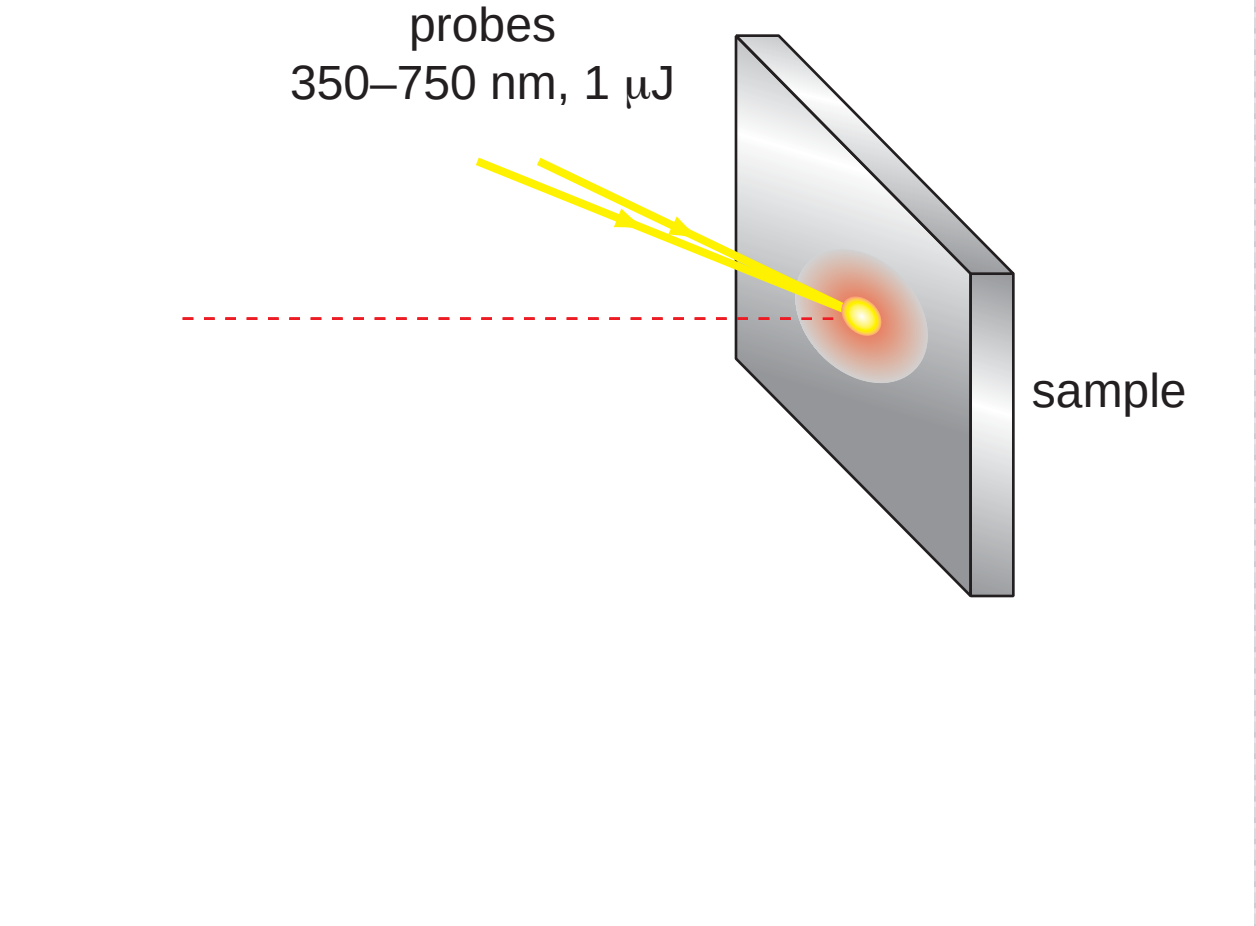


broadband time-resolved ellipsometry



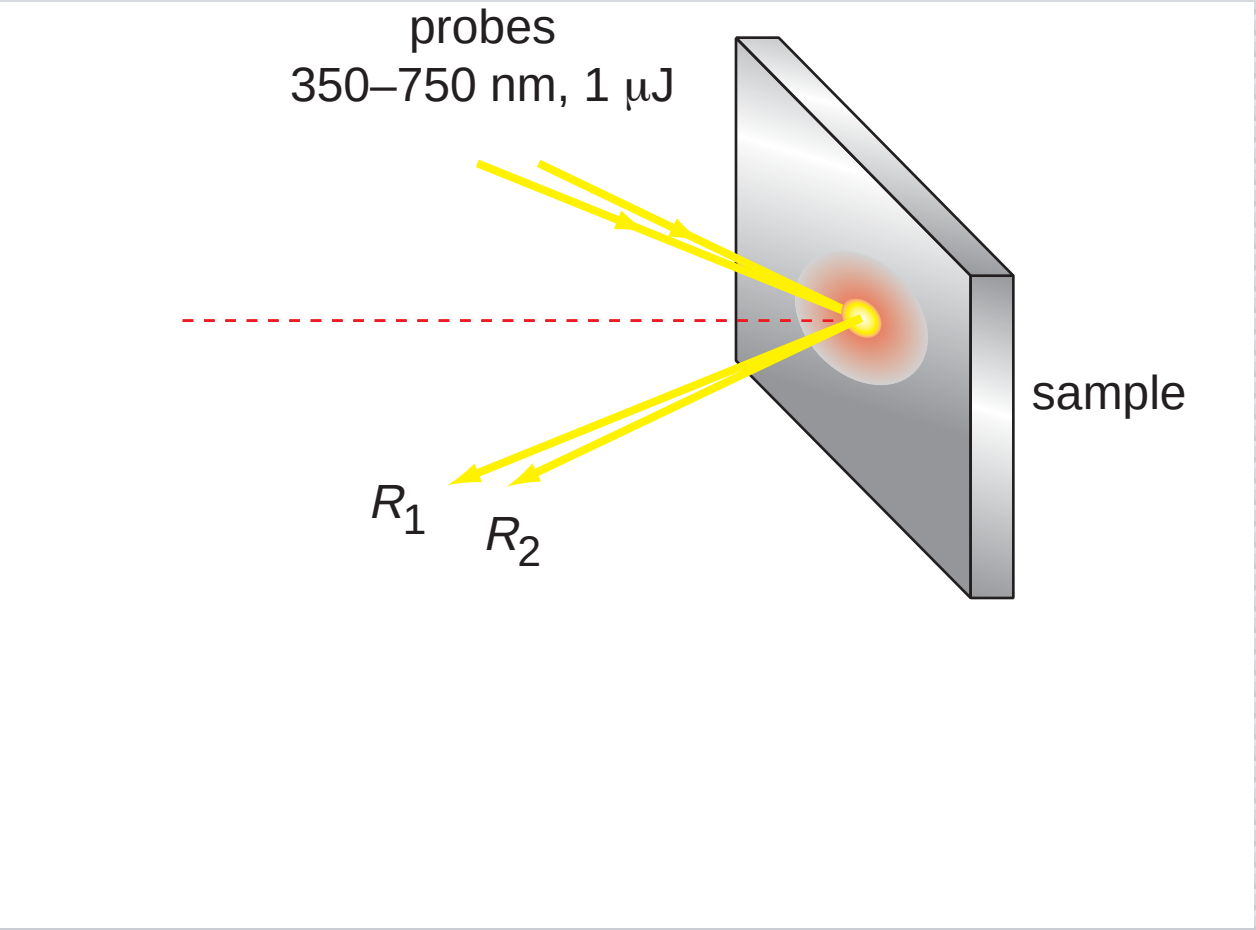
Technique

broadband time-resolved ellipsometry



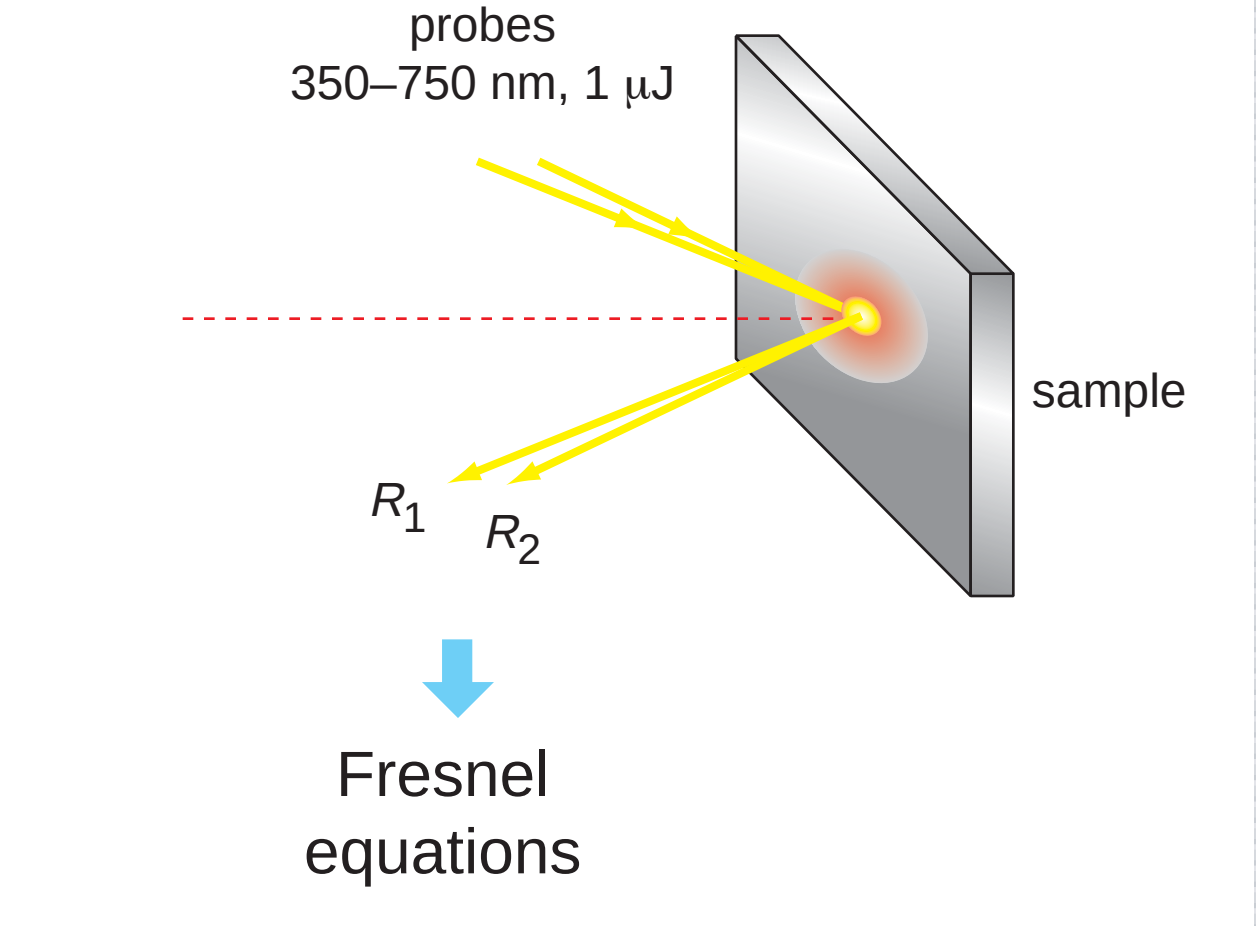
Technique

broadband time-resolved ellipsometry



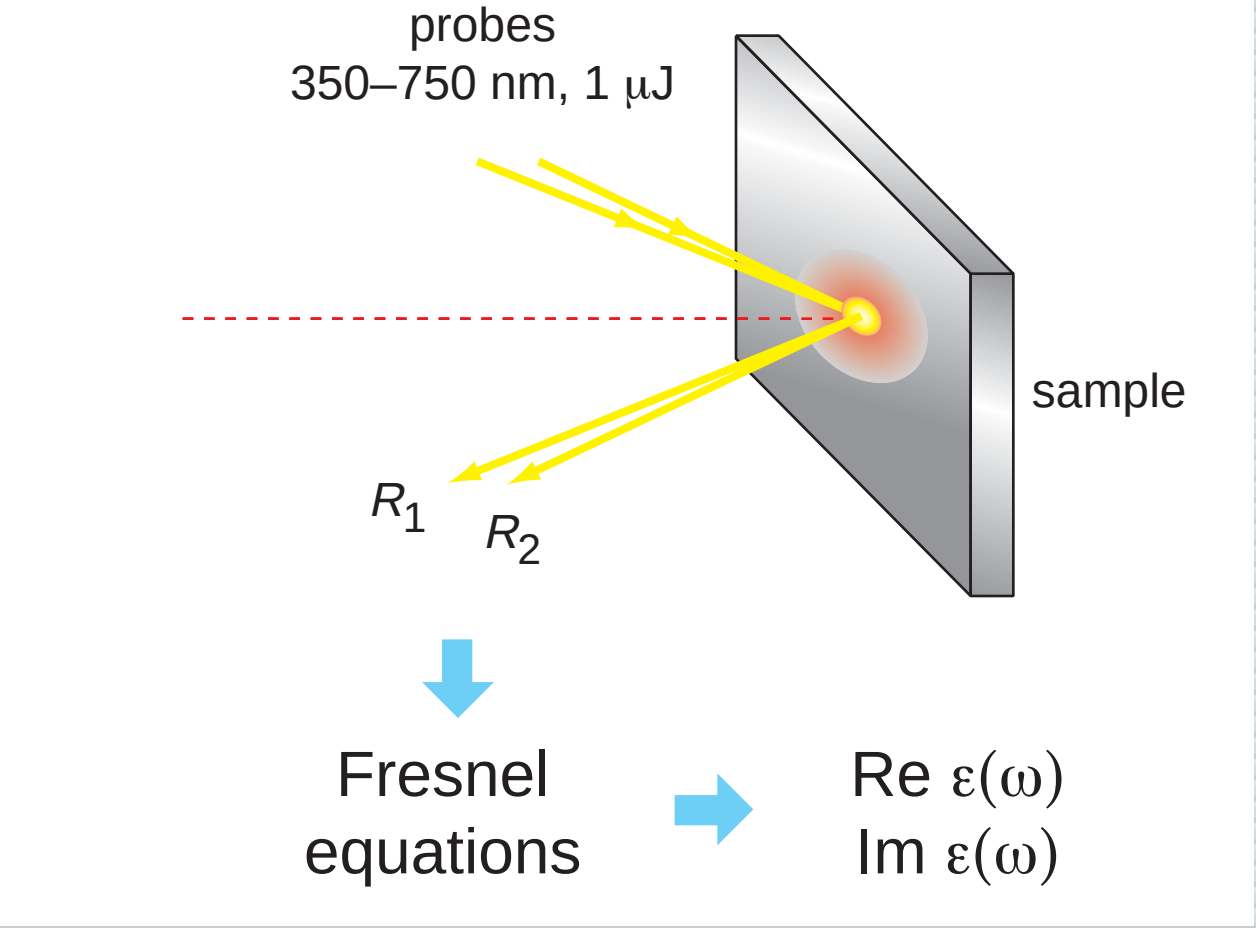
Technique

broadband time-resolved ellipsometry

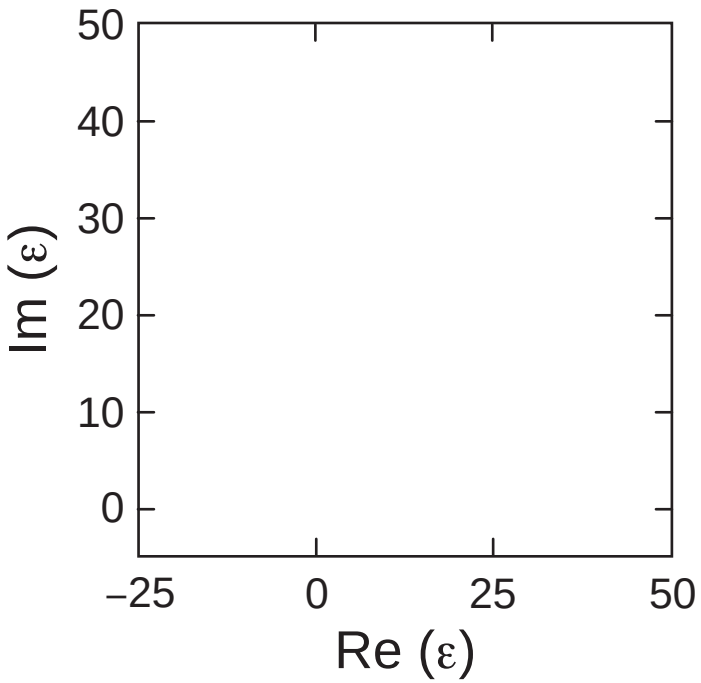


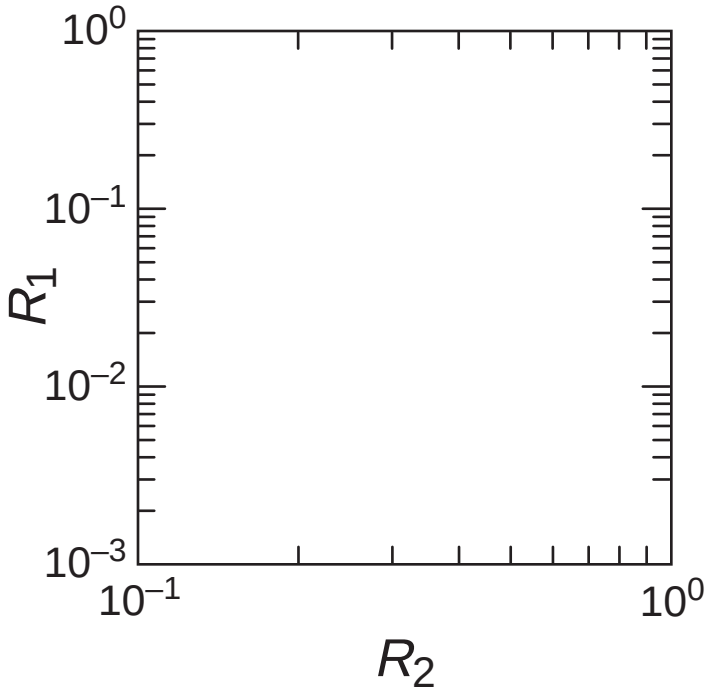
Technique

broadband time-resolved ellipsometry



choice of angles

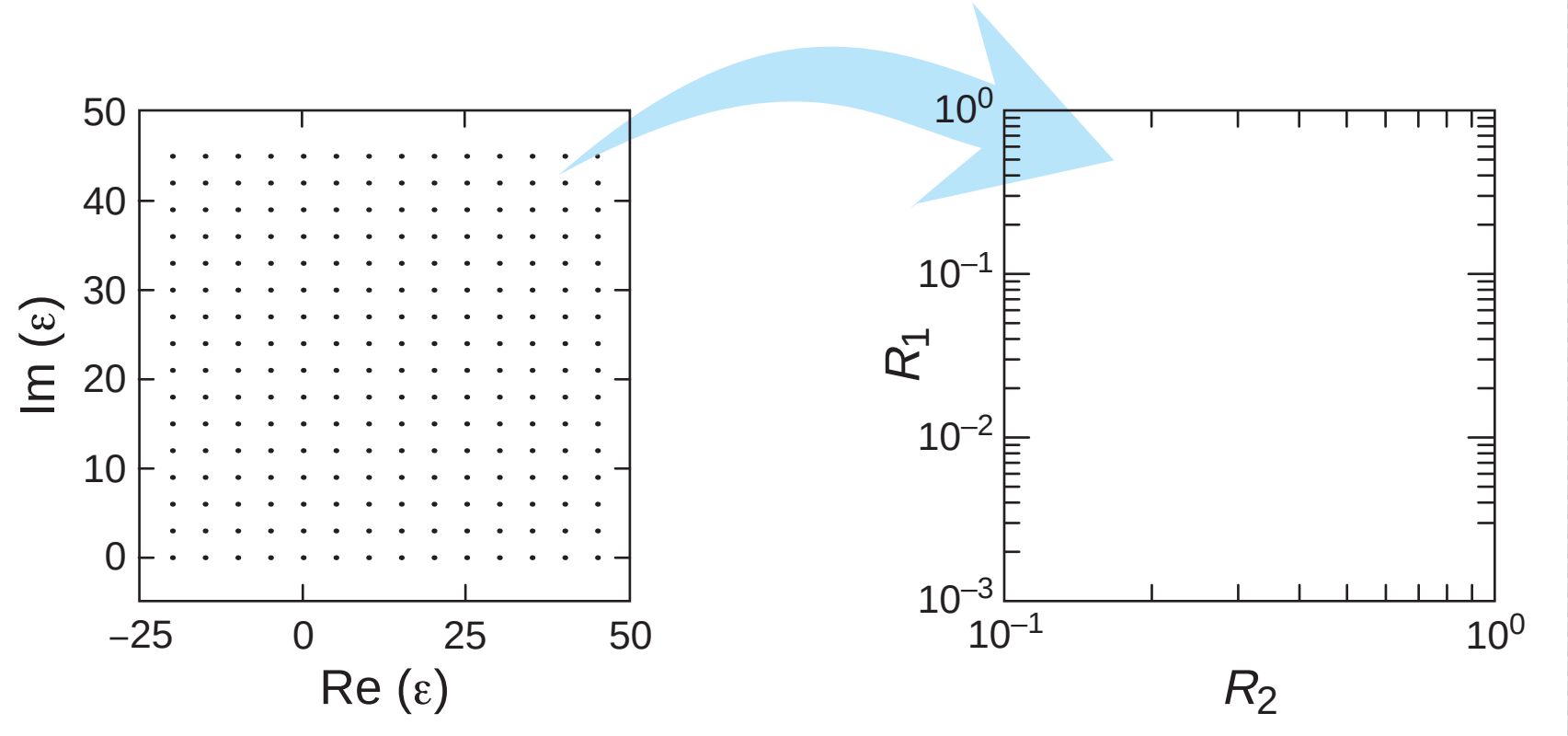




Fresnel equations cannot be inverted analytically

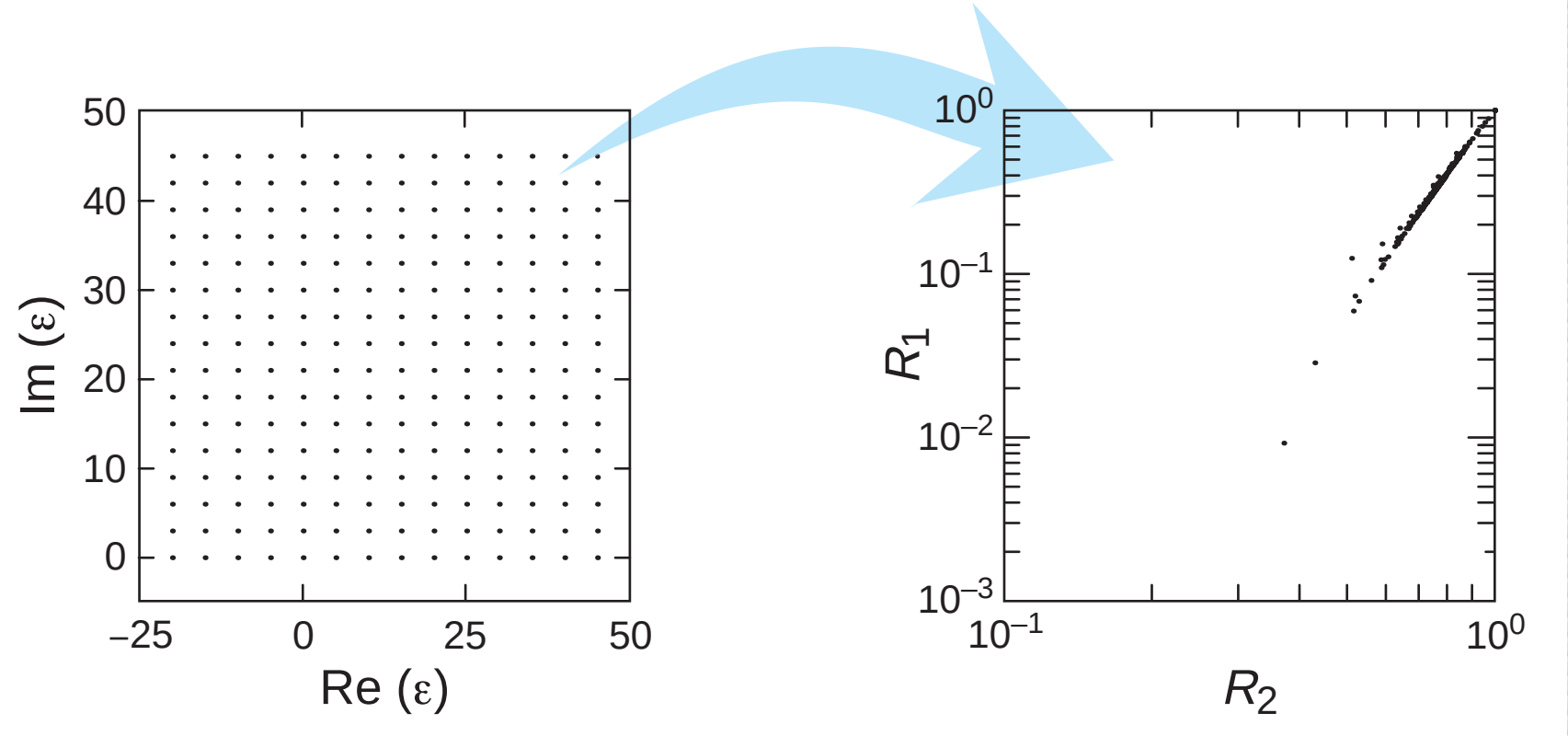
Technique

choice of angles



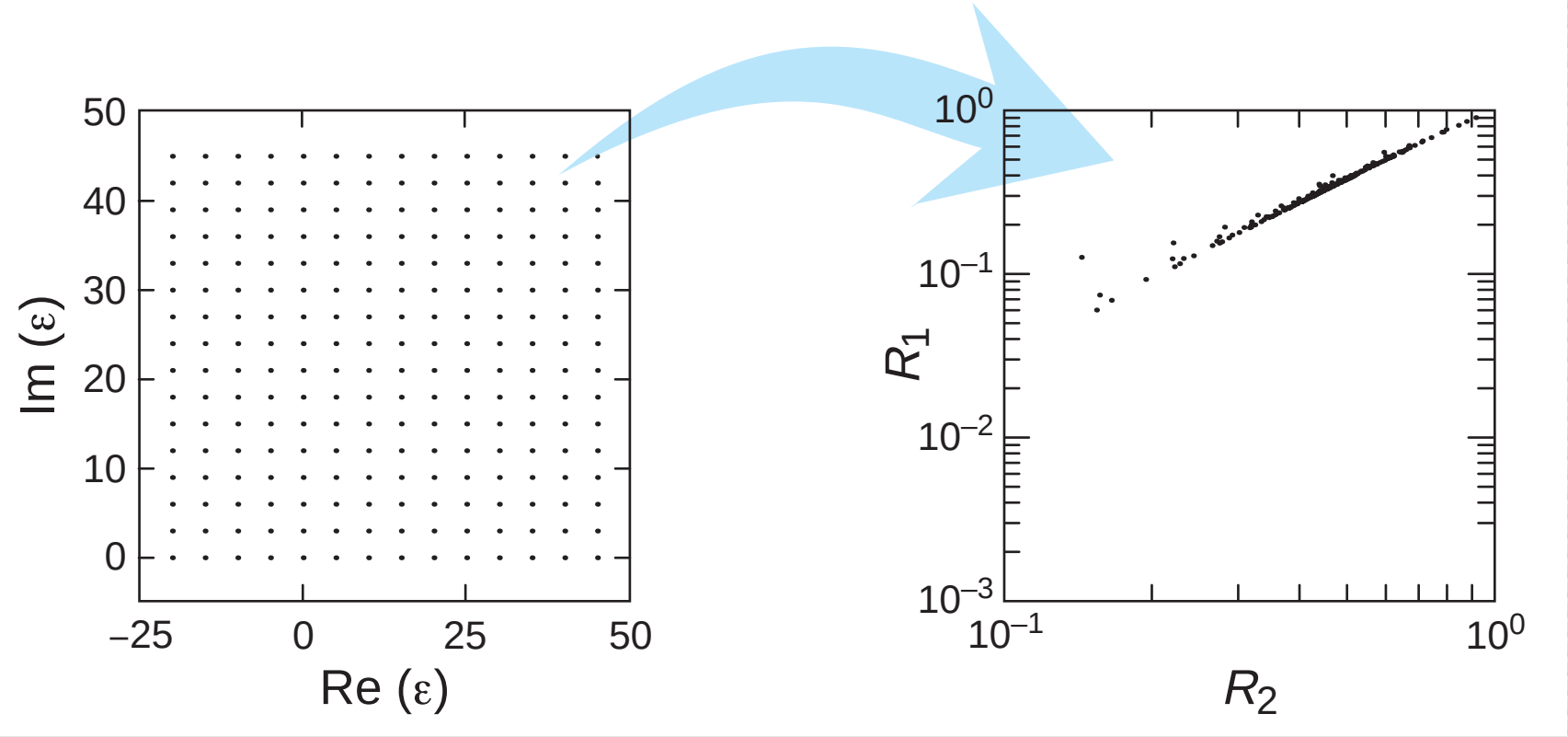
need numerical inversion

choice of angles



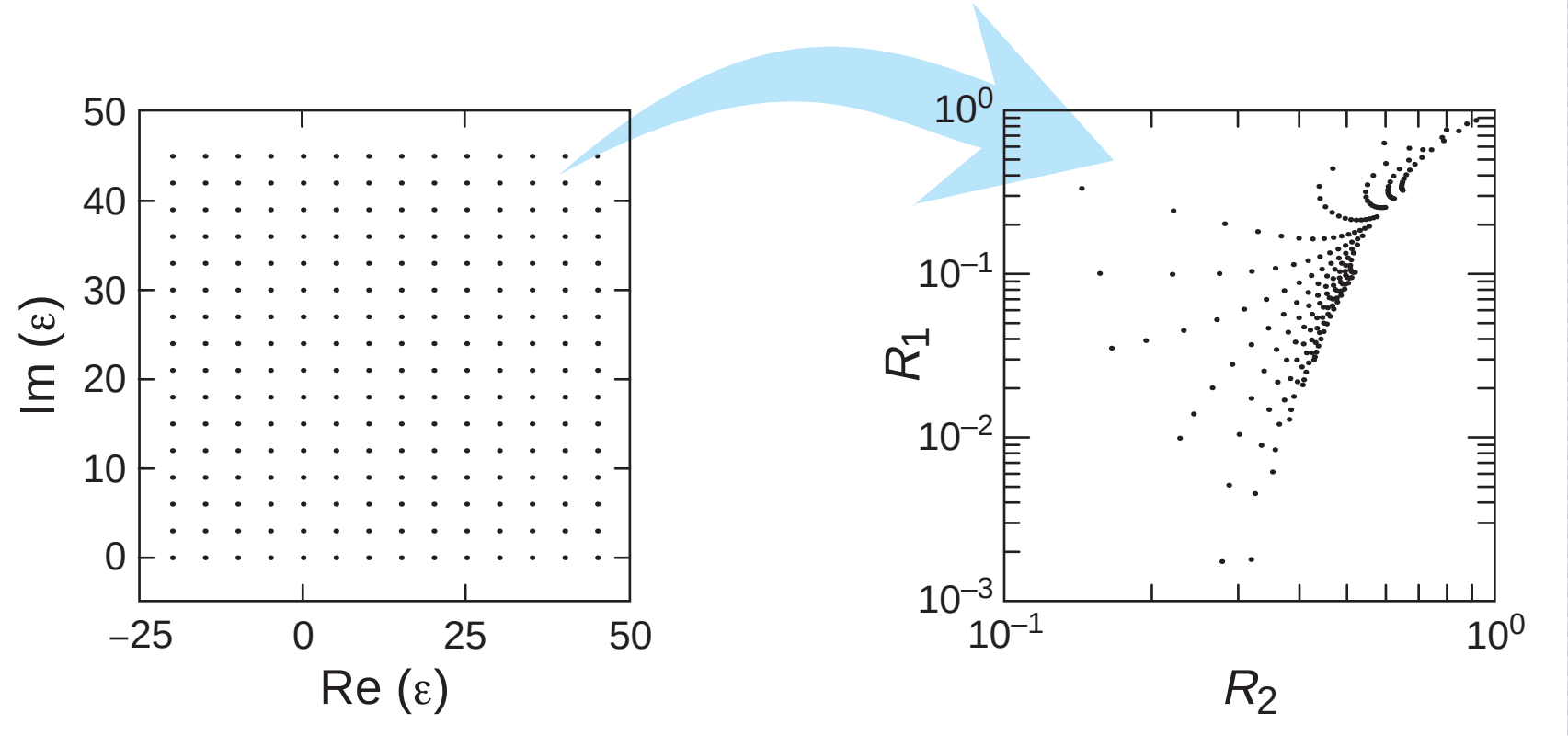
R_1 = 45° *p*-pol, R_2 = 45° *s*-pol

choice of angles



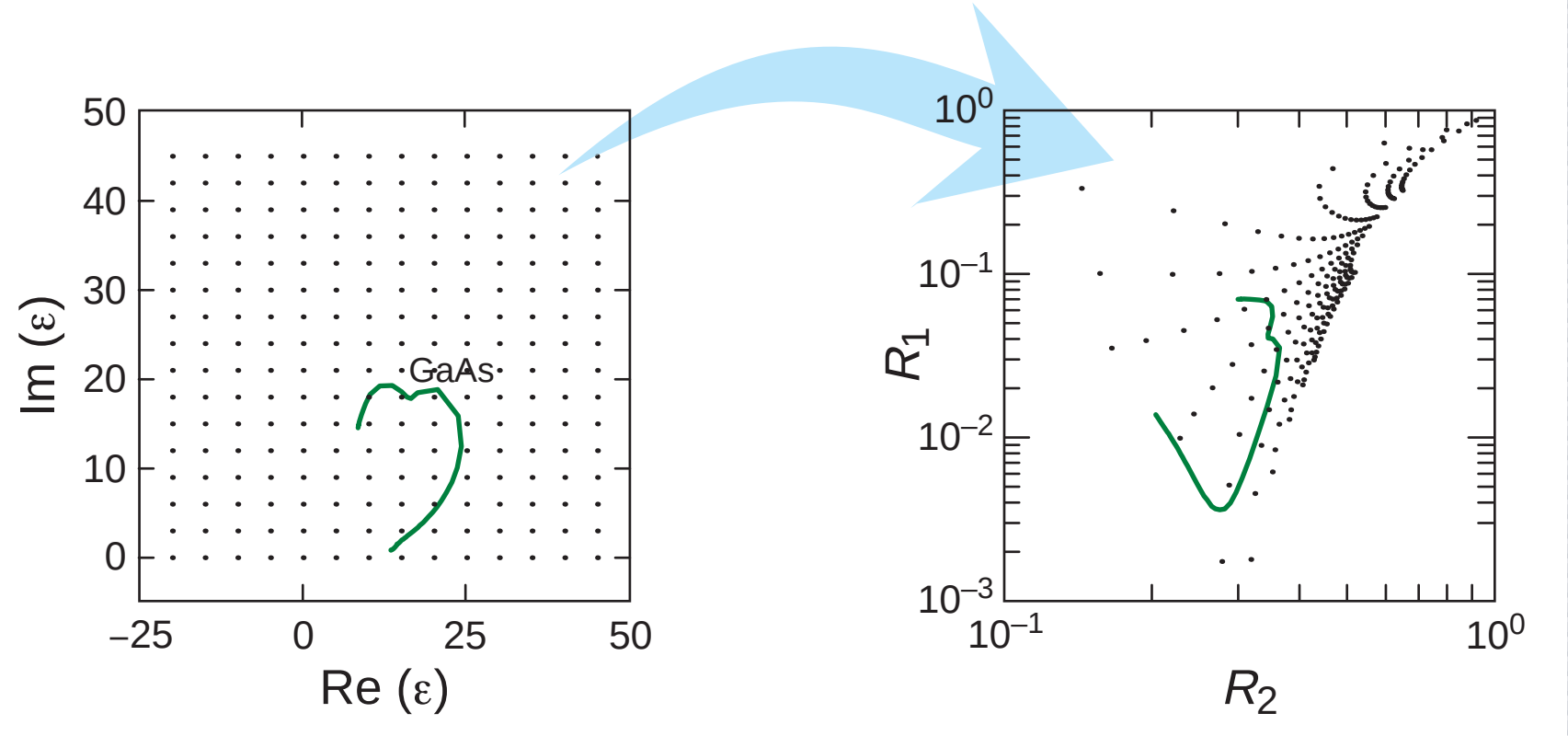
R_1 = 60° *p*-pol, R_2 = 45° *p*-pol

choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

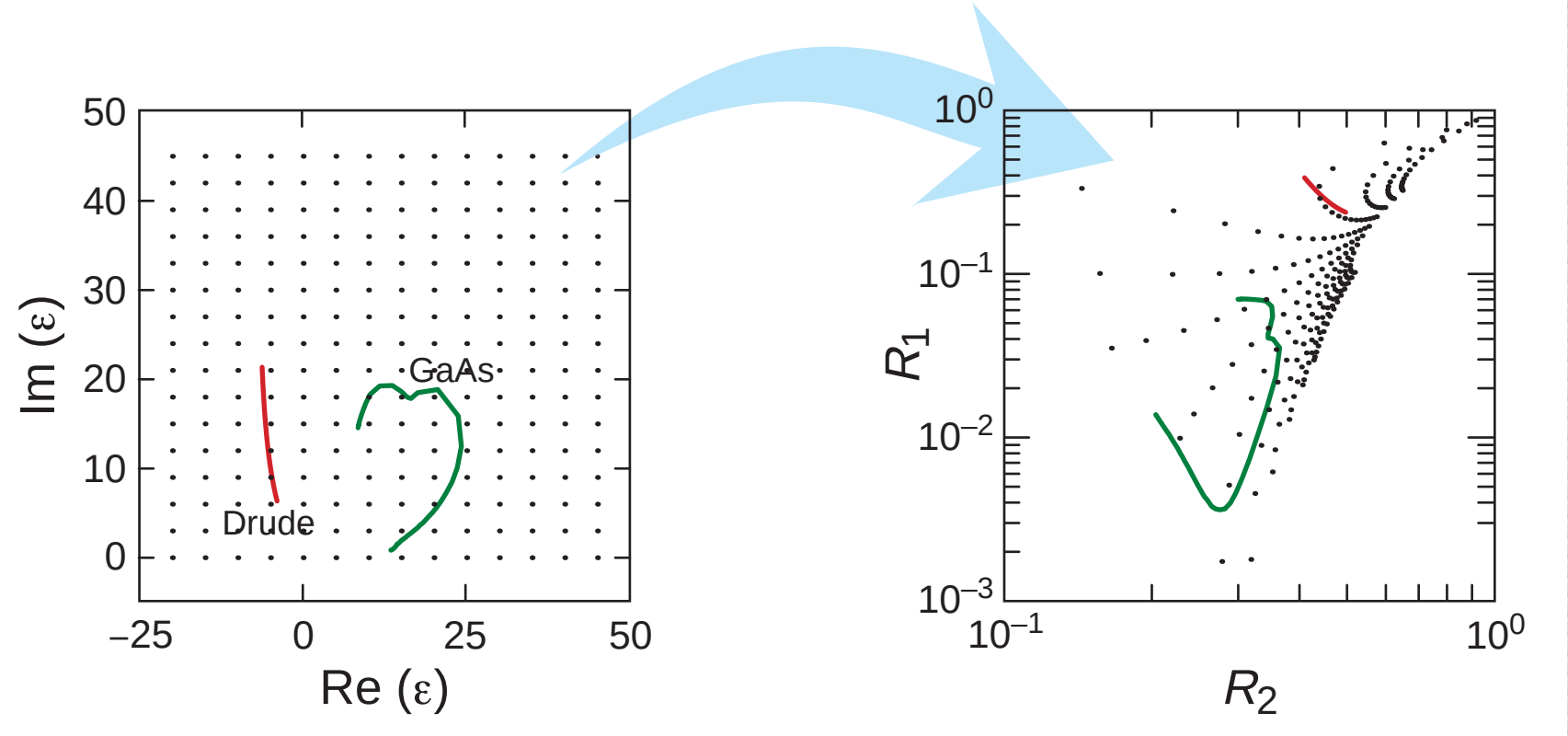
choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

Technique

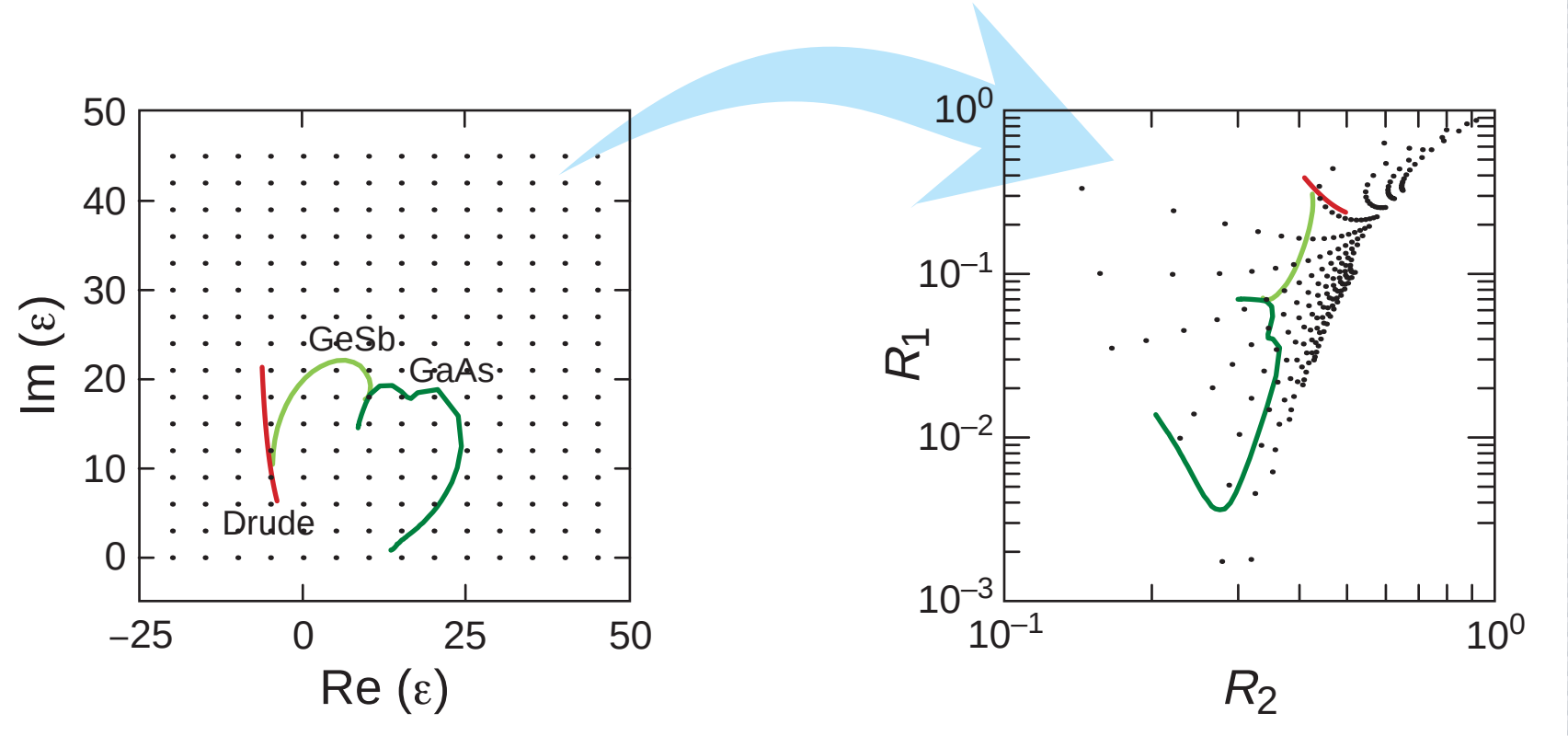
choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

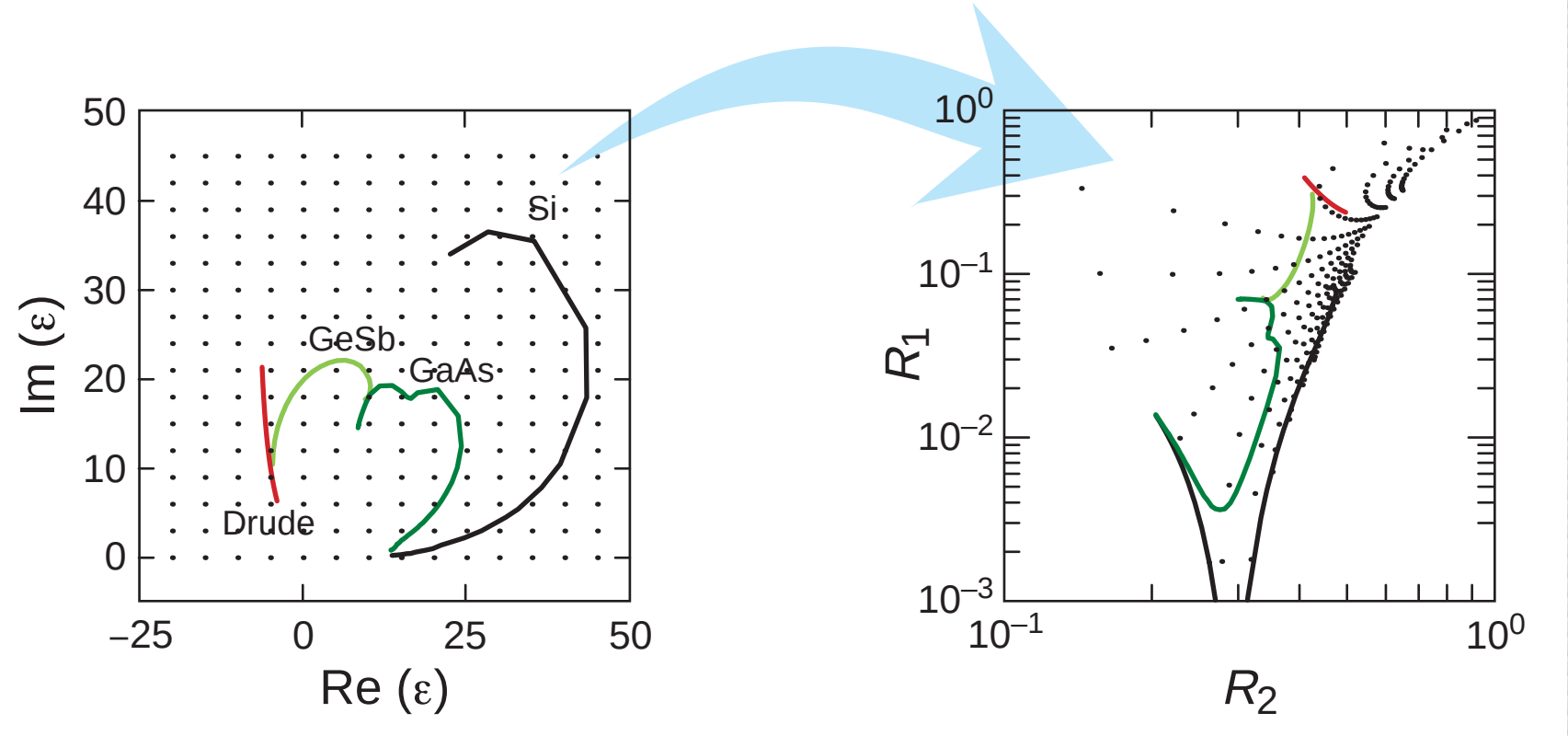
Technique

choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

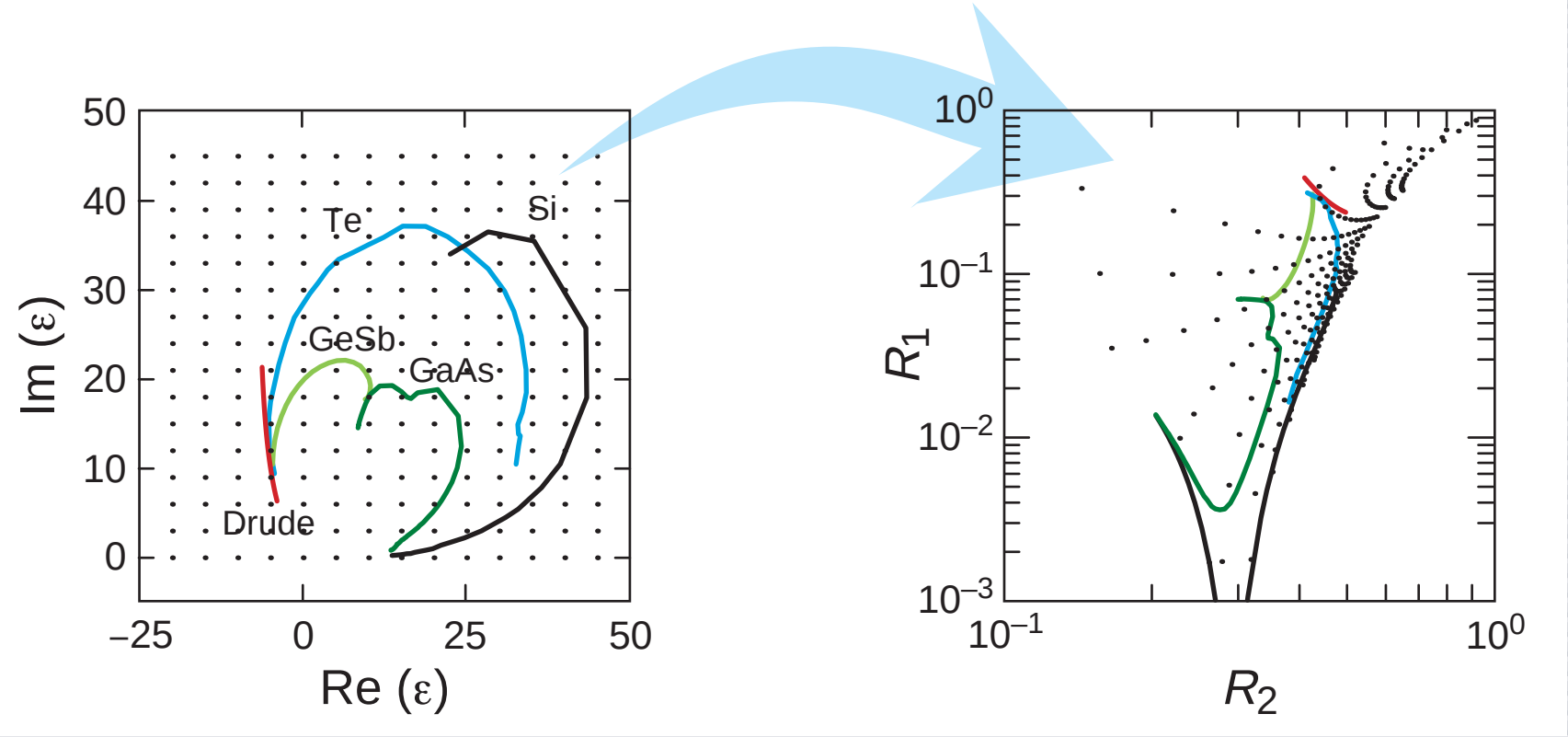
choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

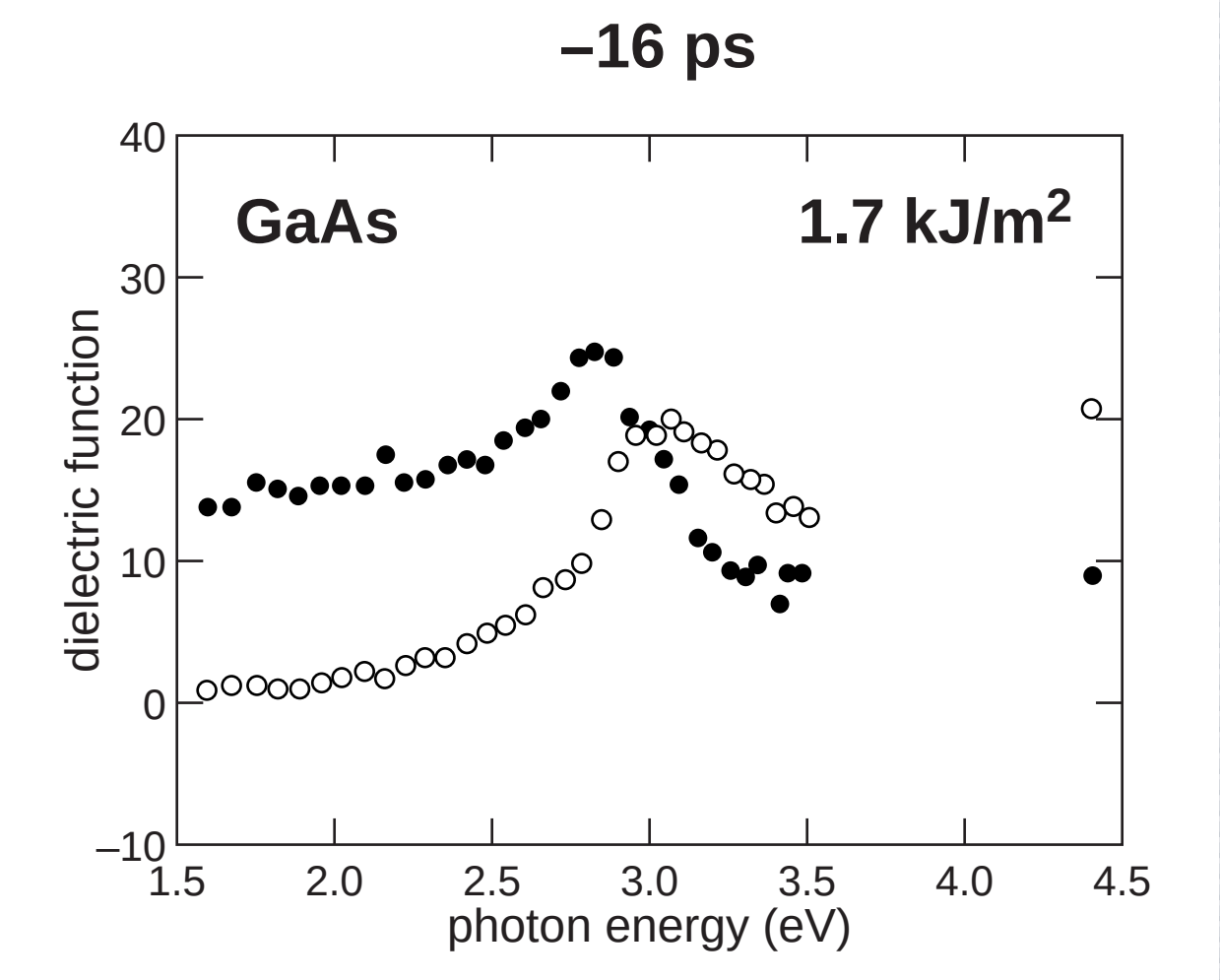
Technique

choice of angles



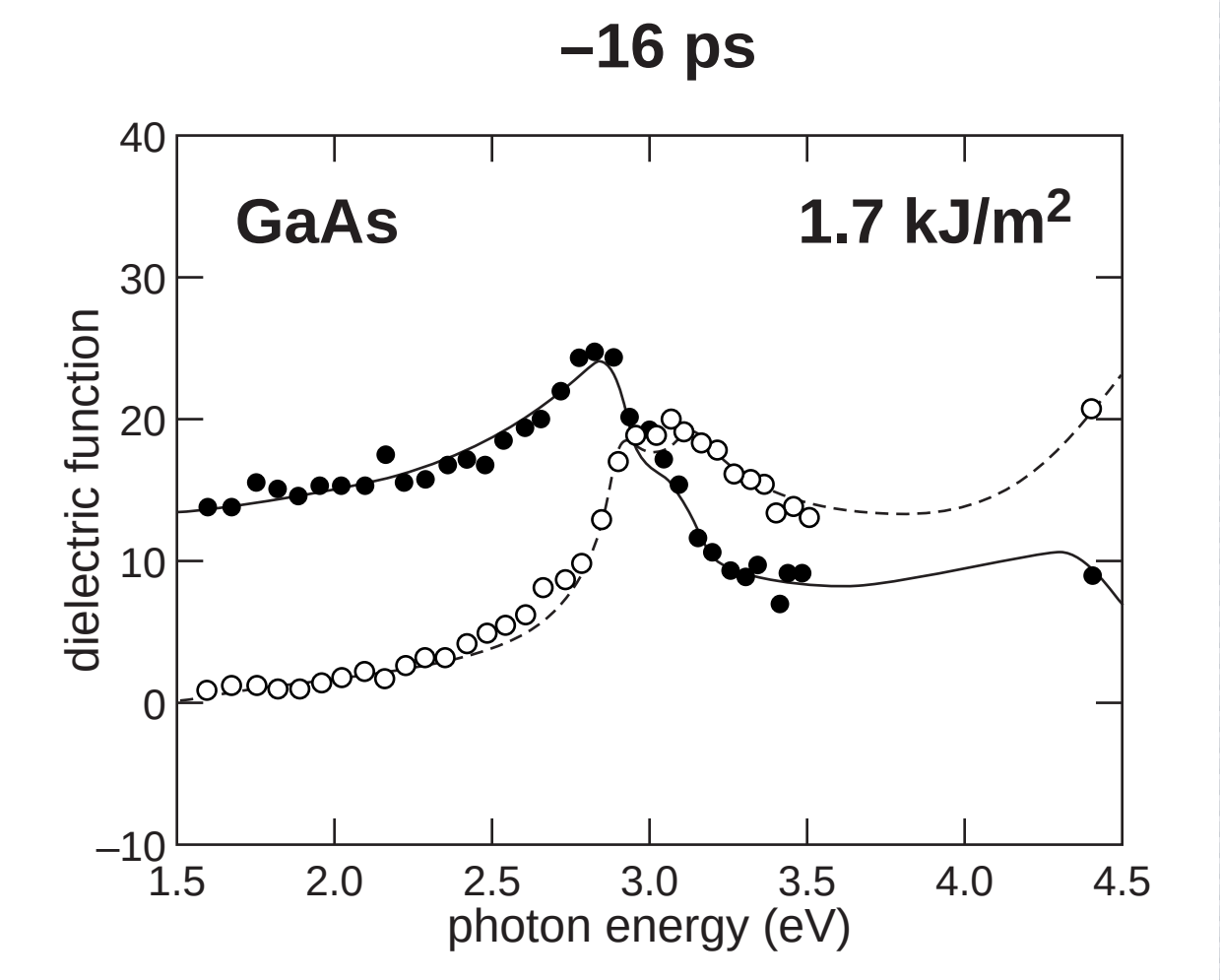
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

Technique



***Phys. Rev. Lett.* 80, 185 (1998)**

Technique



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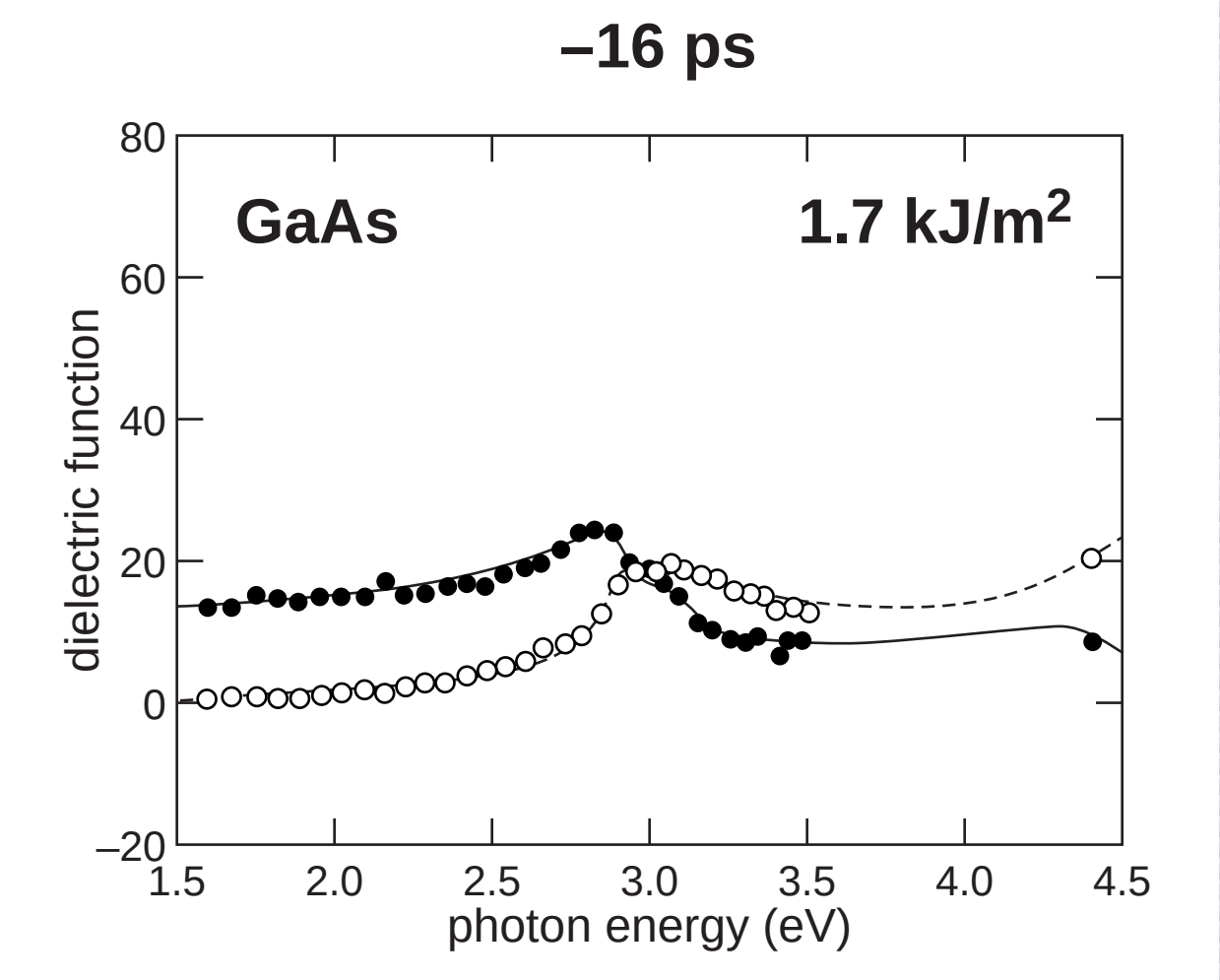

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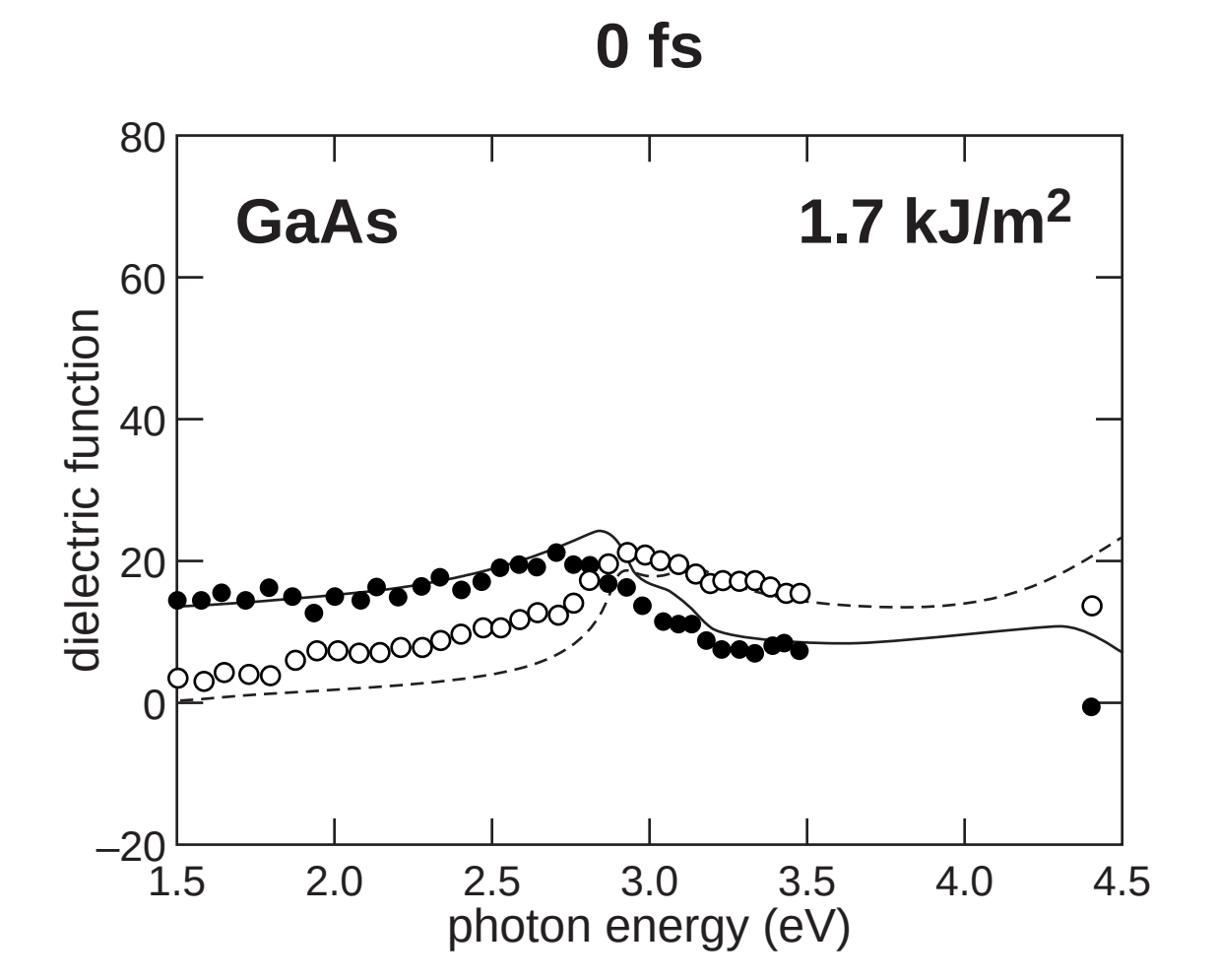

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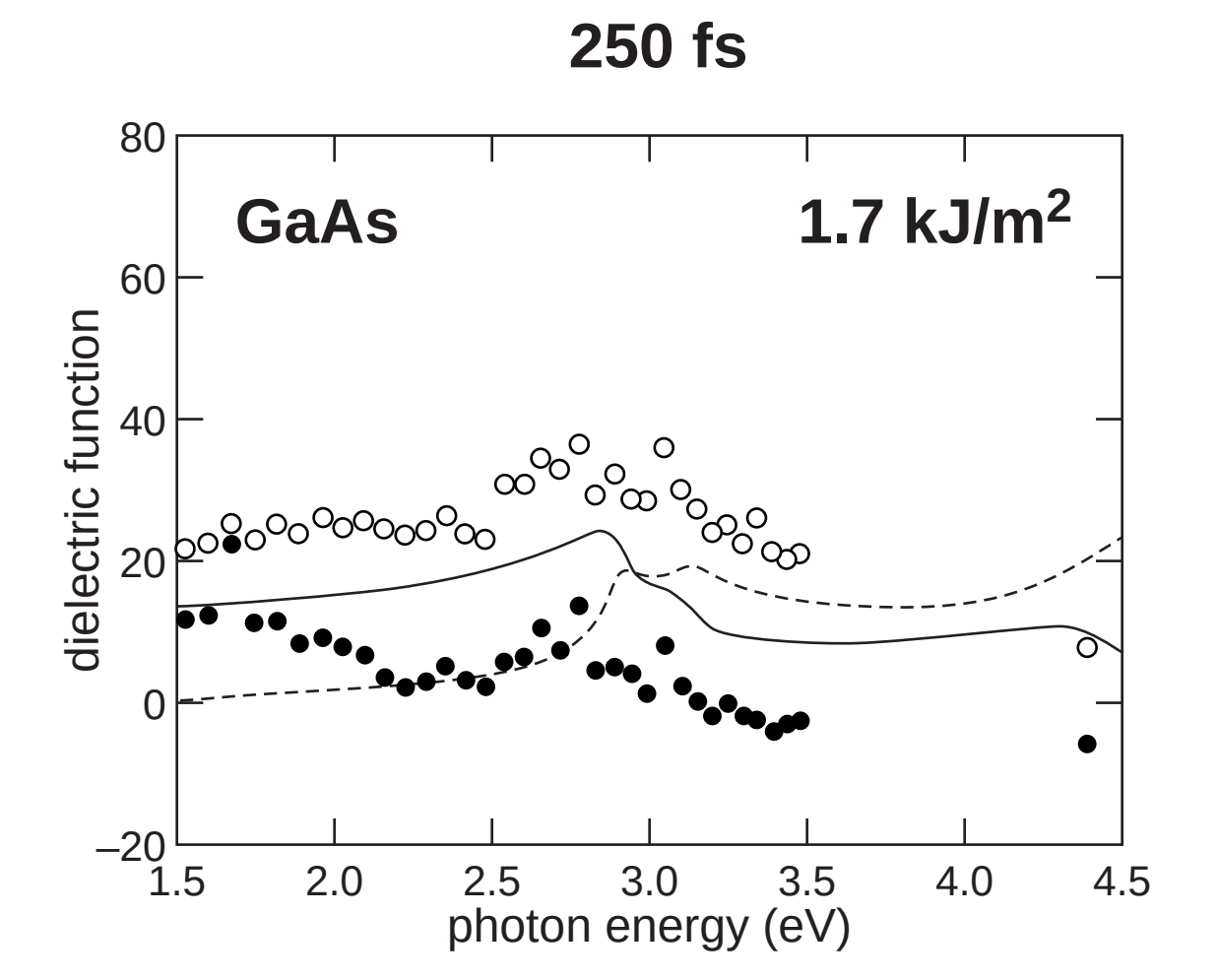

Phys. Rev. Lett. **80, 185 (1998)**

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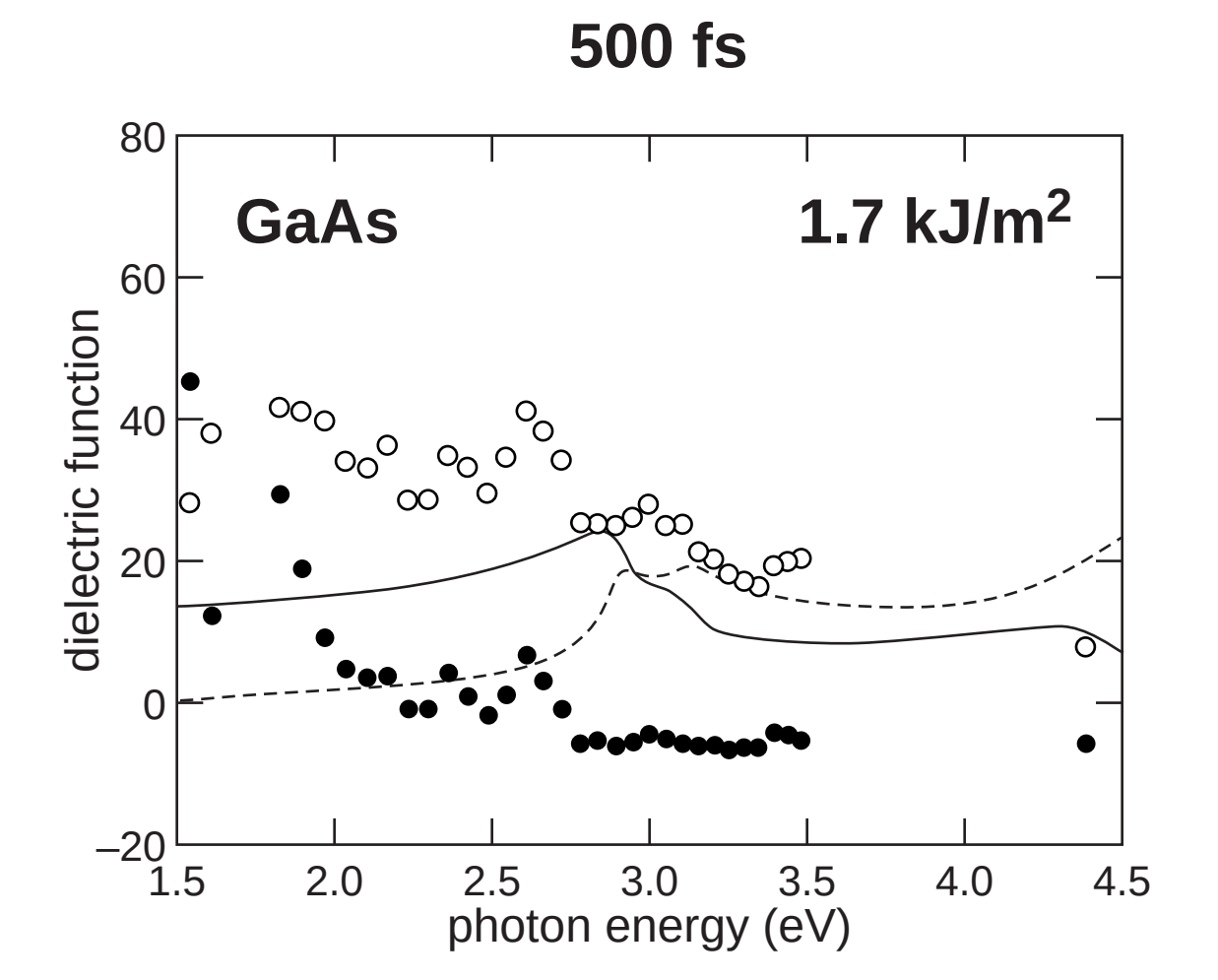
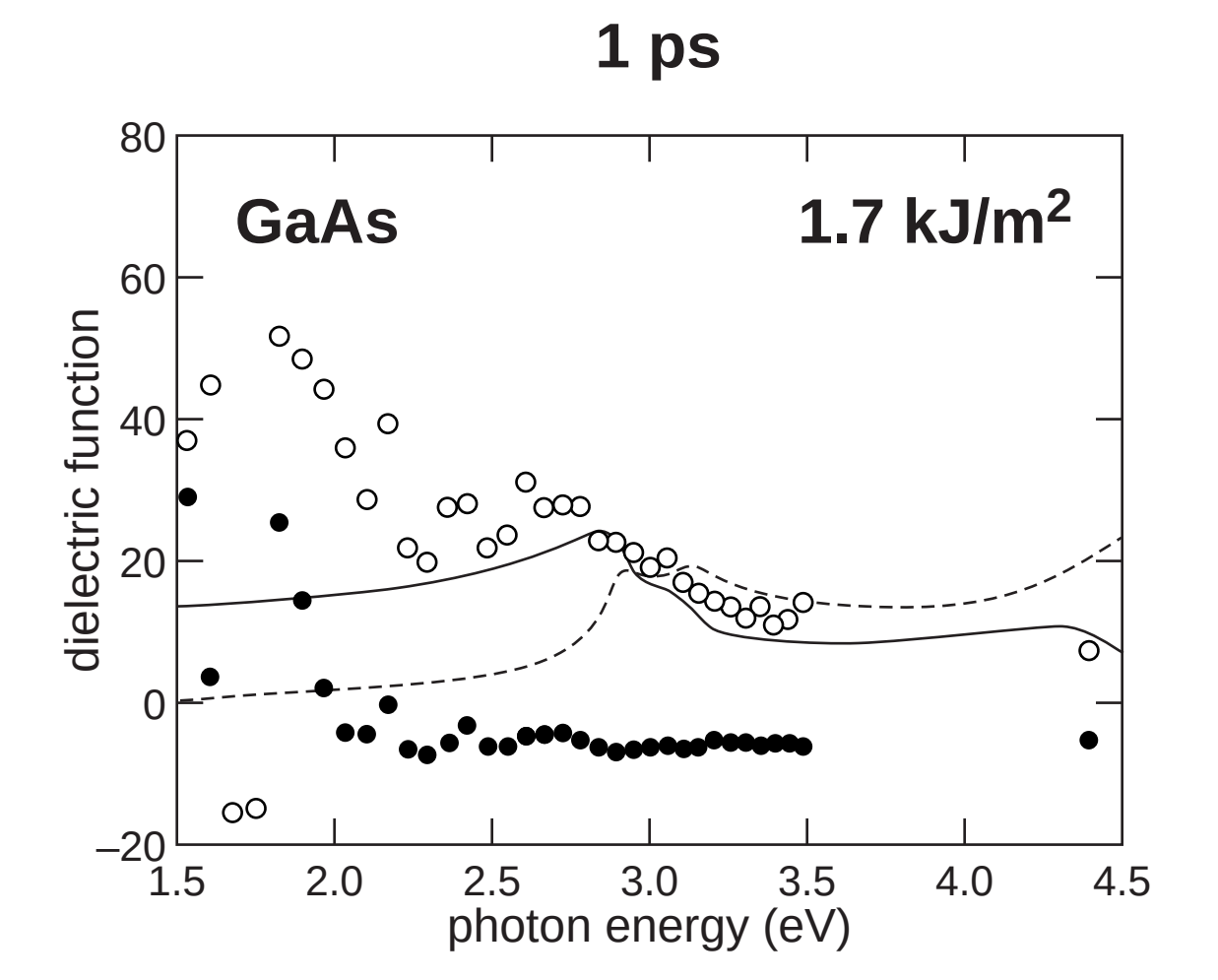


Phys. Rev. Lett. **80, 185 (1998)**

Technique



***Phys. Rev. Lett.* 80, 185 (1998)**

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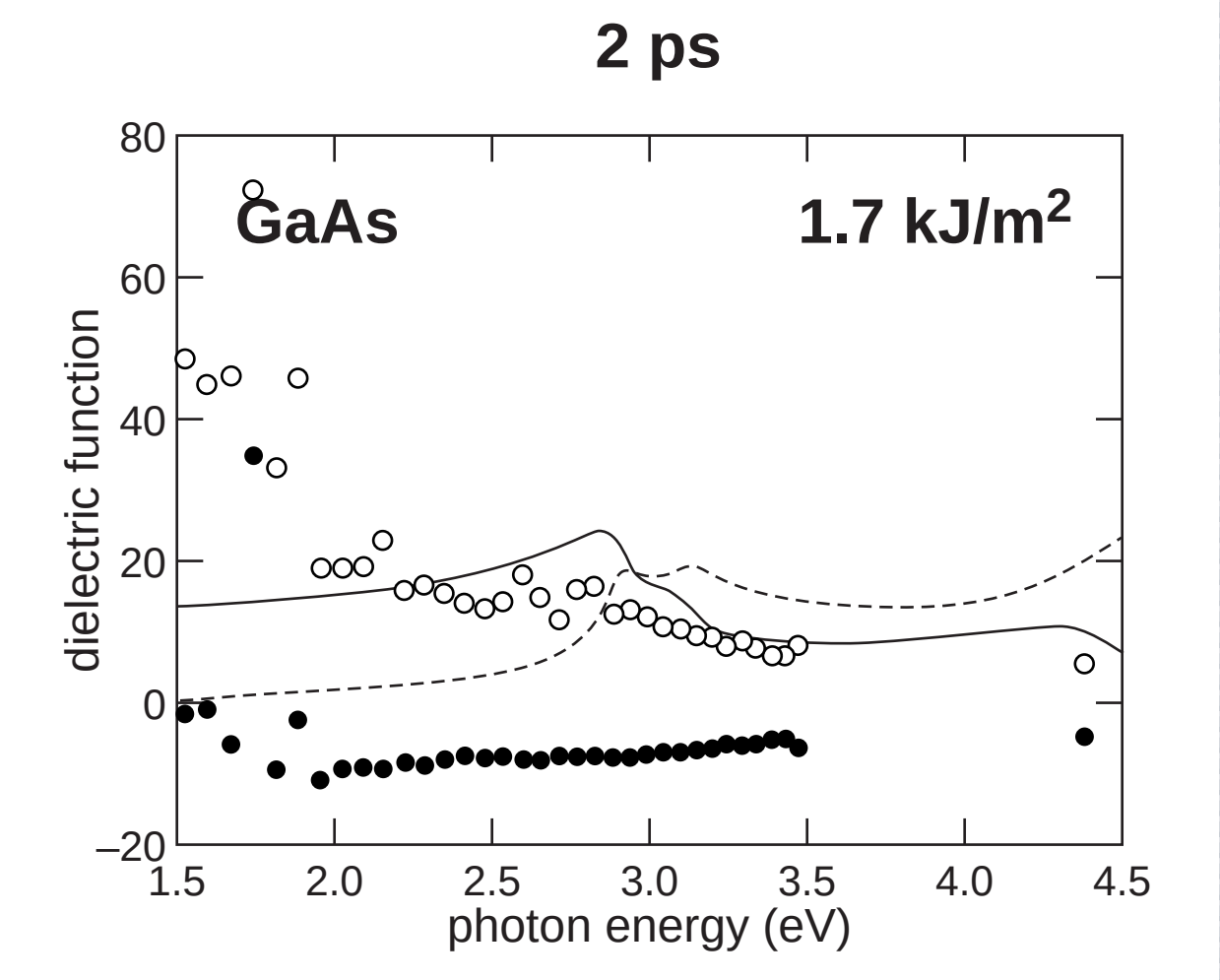

***Phys. Rev. Lett.* 80, 185 (1998)**

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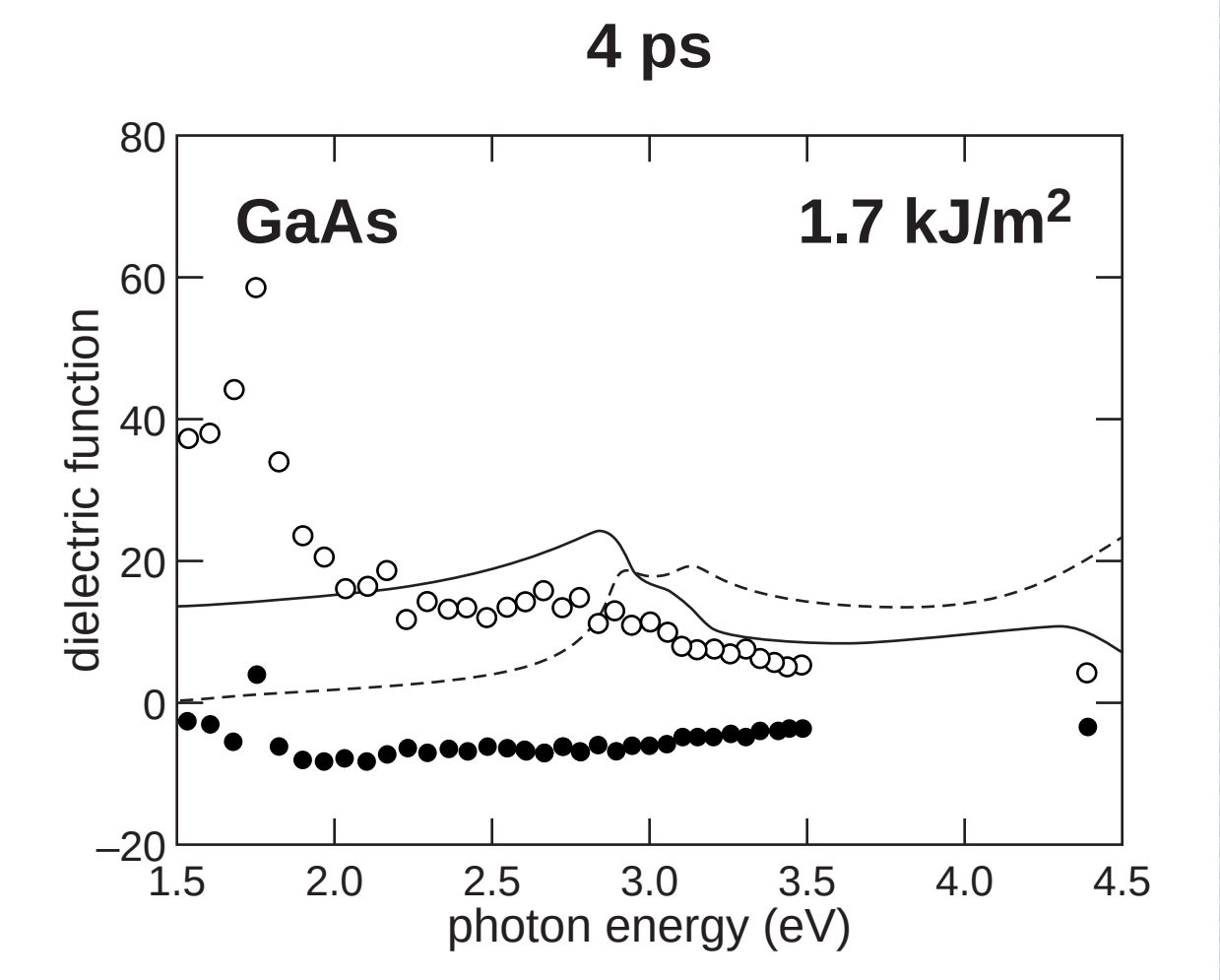
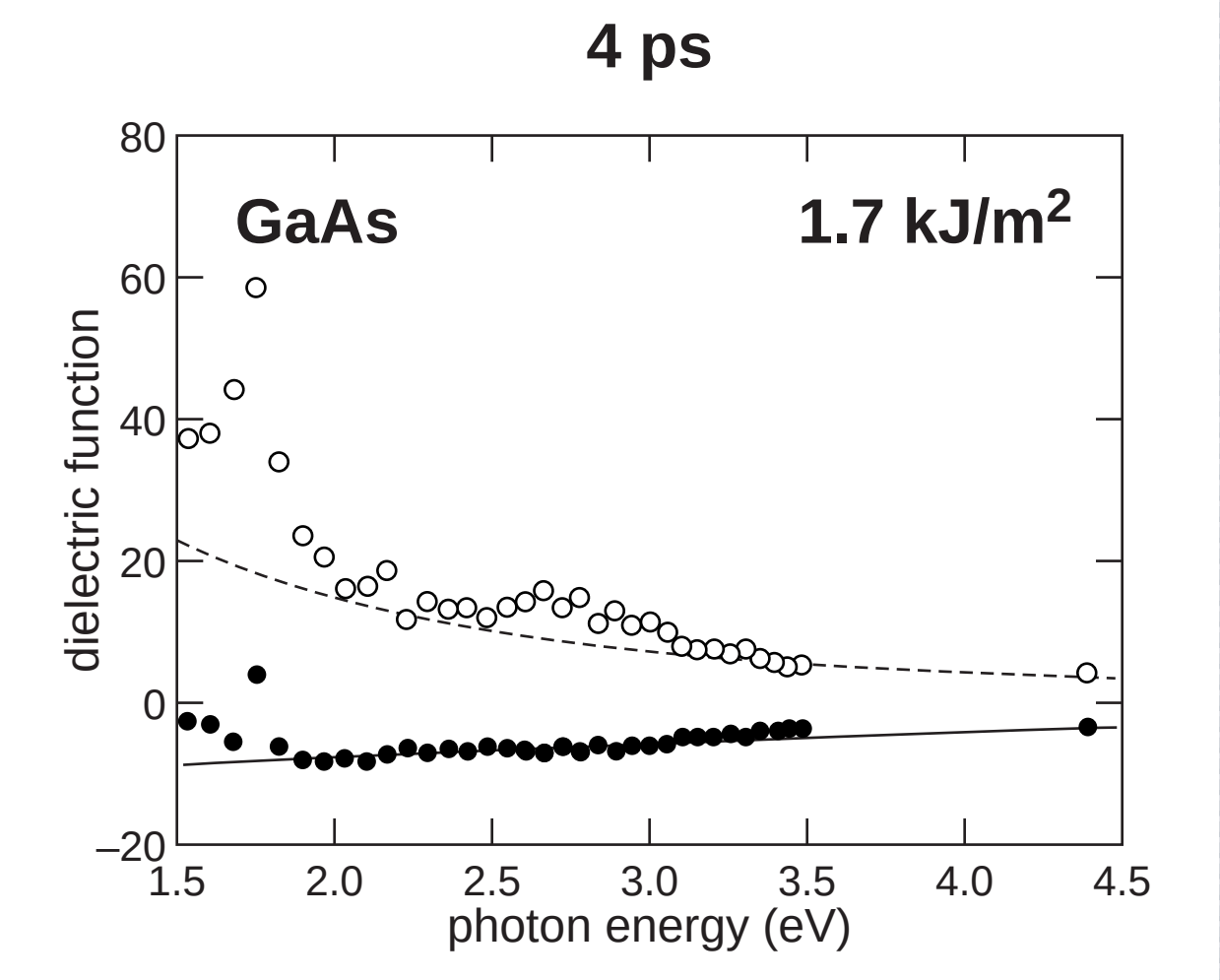


Phys. Rev. Lett. **80, 185 (1998)**

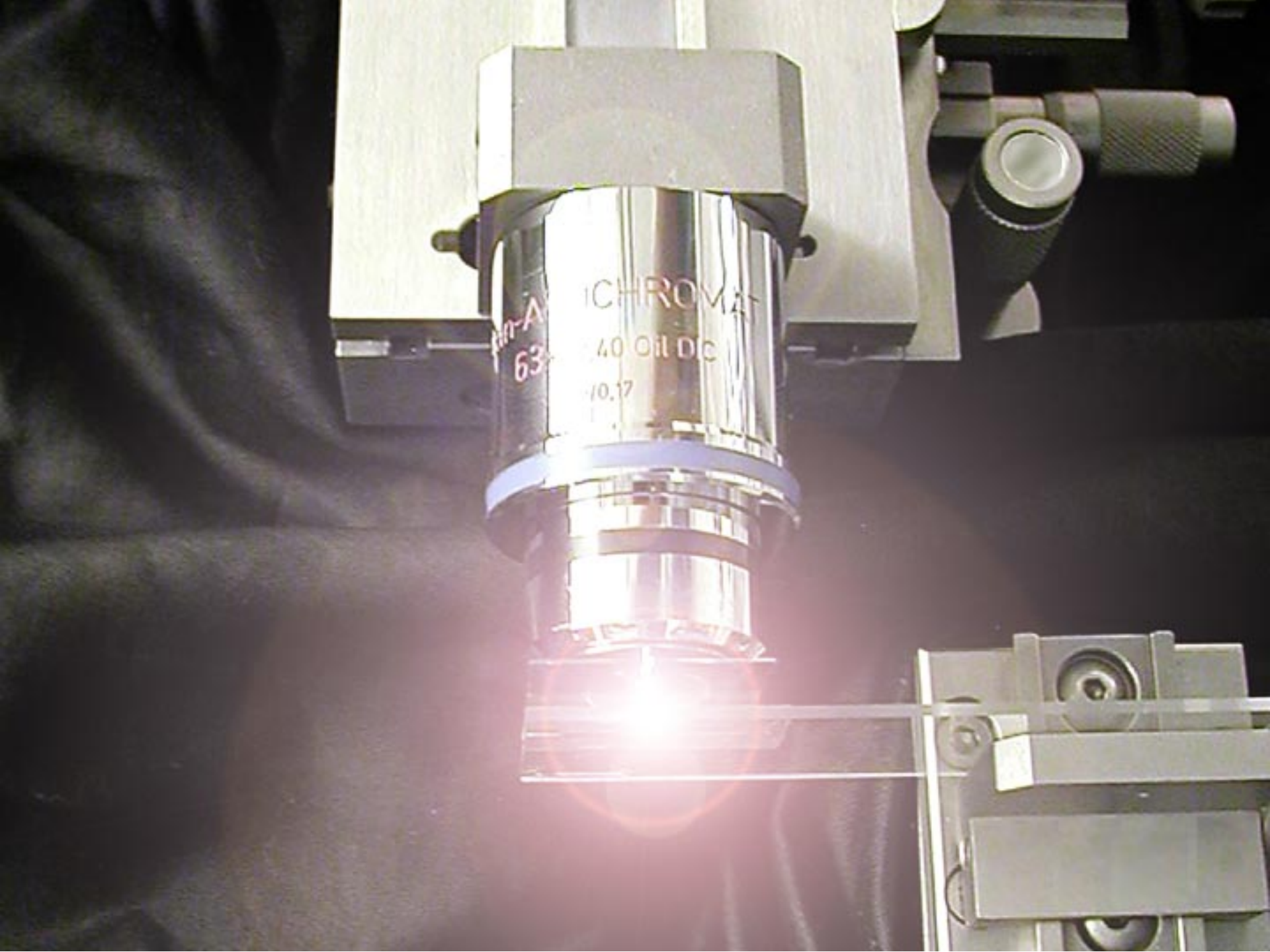
Technique



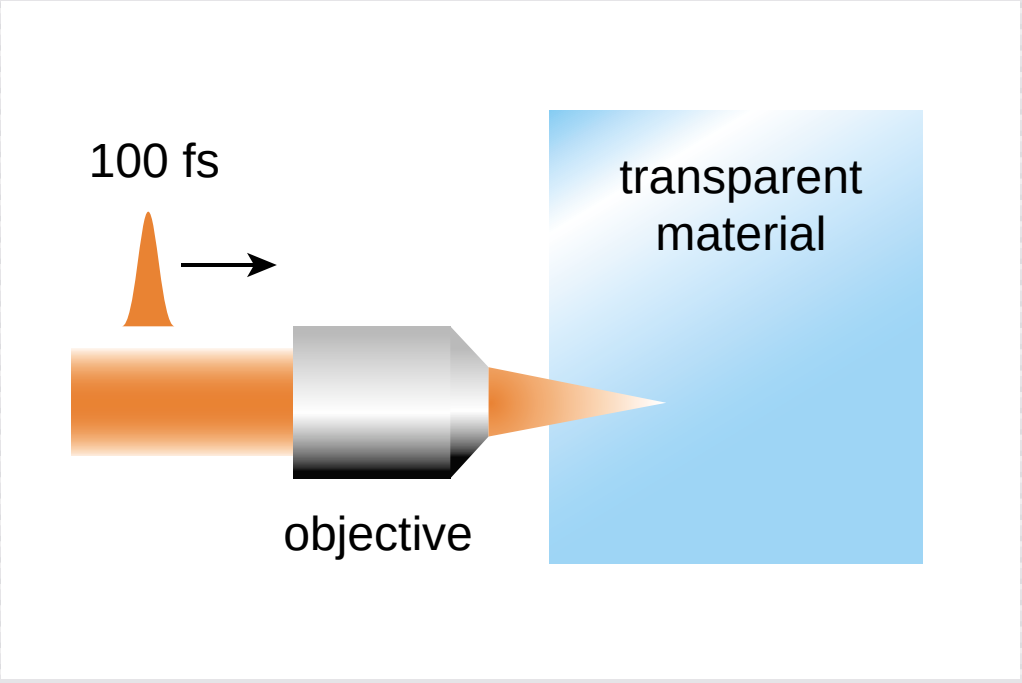
***Phys. Rev. Lett.* 80, 185 (1998)**

Technique

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

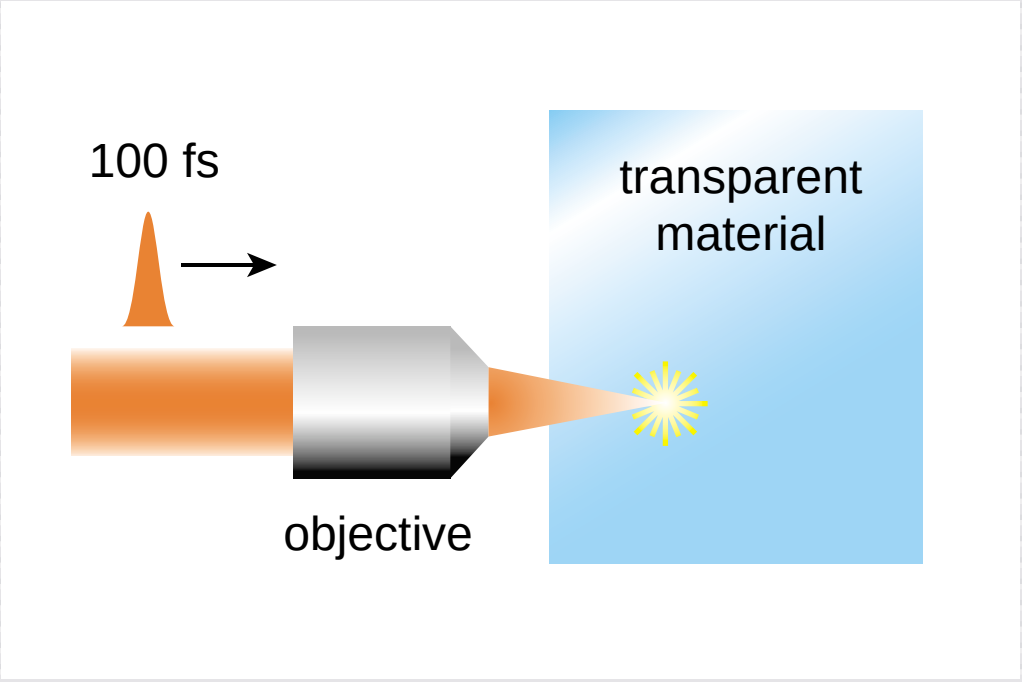

CHROMAT
40 Oil DC
0.17

Processing with fs pulses



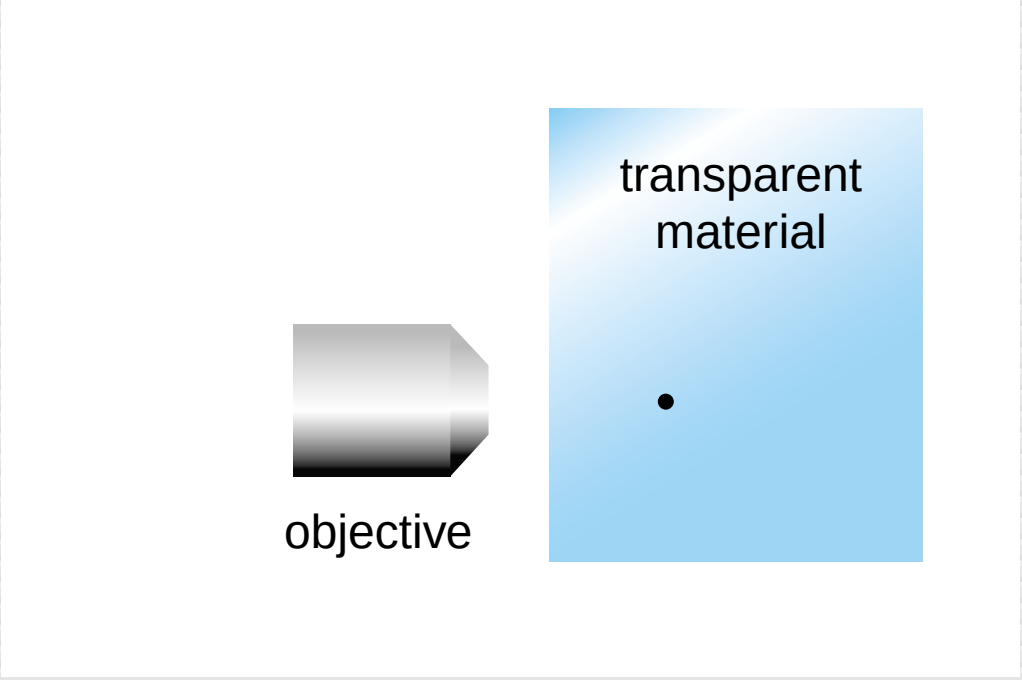
high intensity at focus…

Processing with fs pulses



… causes nonlinear ionization…

Processing with fs pulses



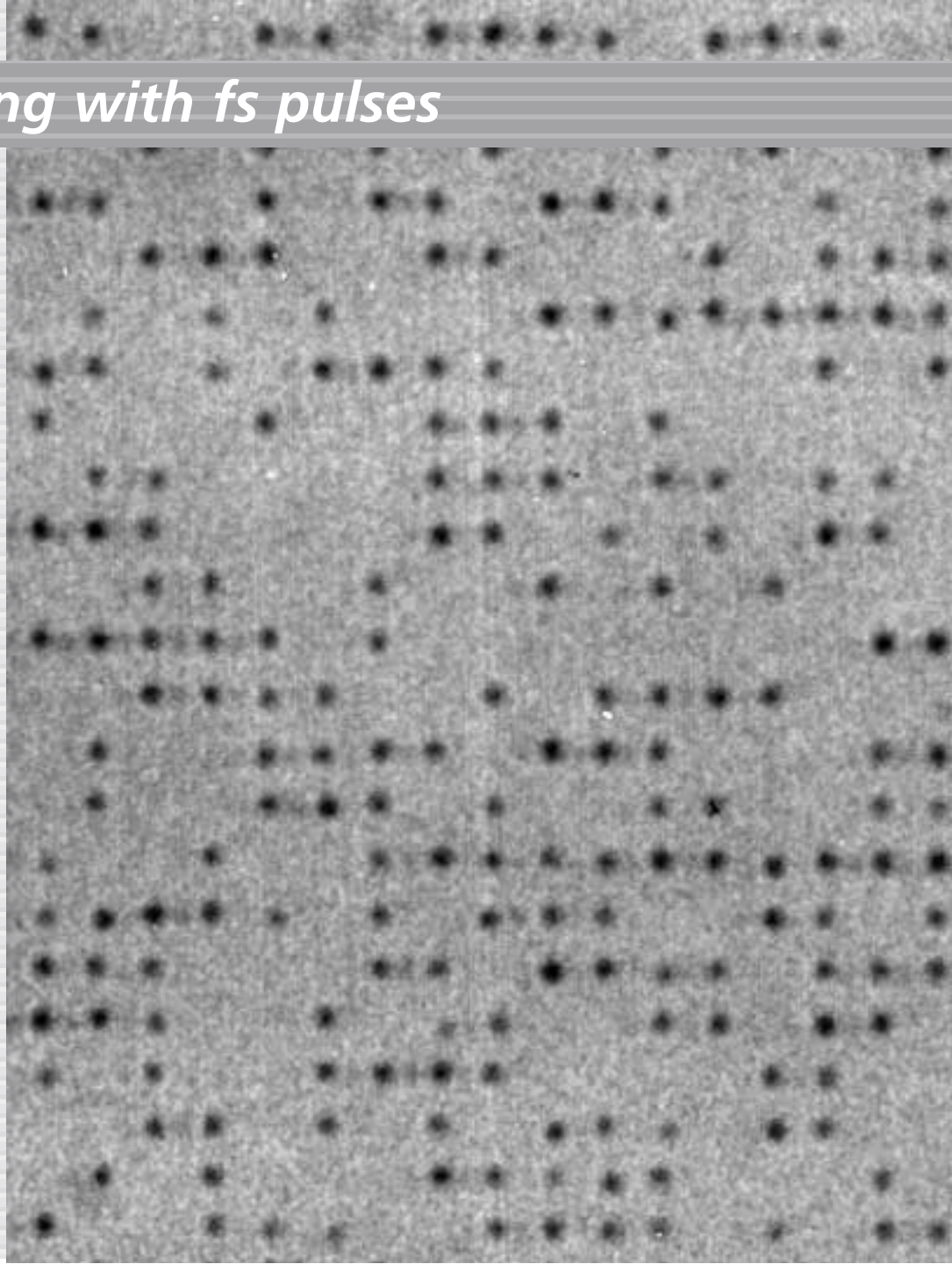
and ‘microexplosion’ causes microscopic damage

Processing with fs pulses

2 x 2 μm array

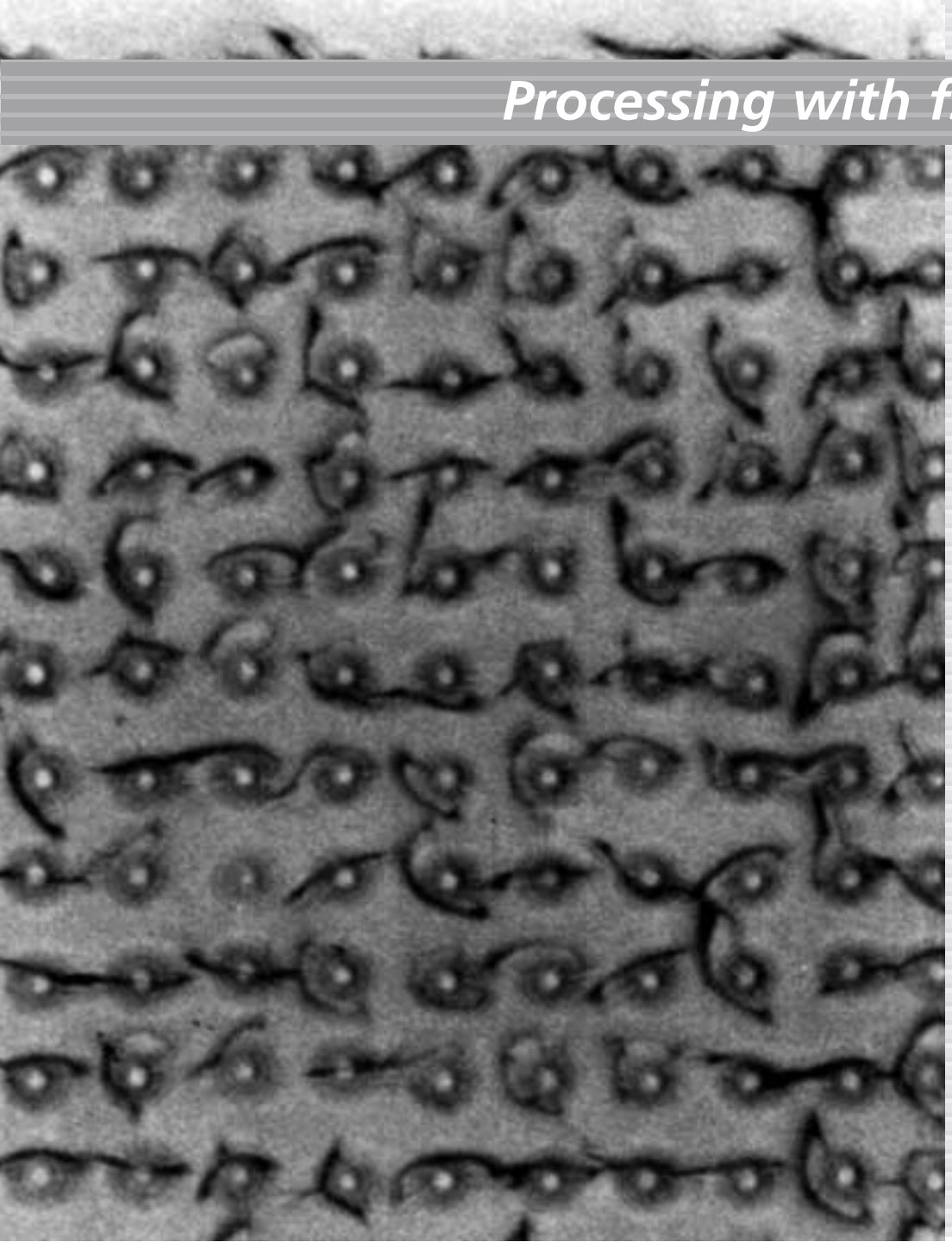
fused silica, 0.65 NA

0.5 μJ, 100 fs, 800 nm

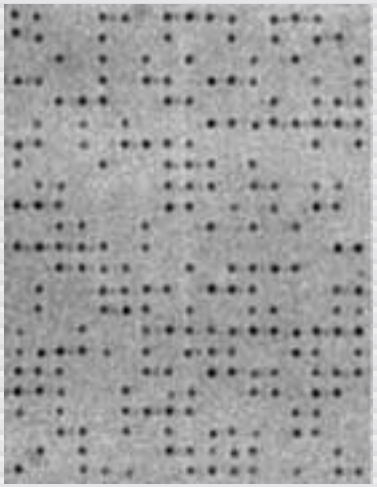


***Opt. Lett.* 21, 2023 (1996)**

Processing with fs pulses

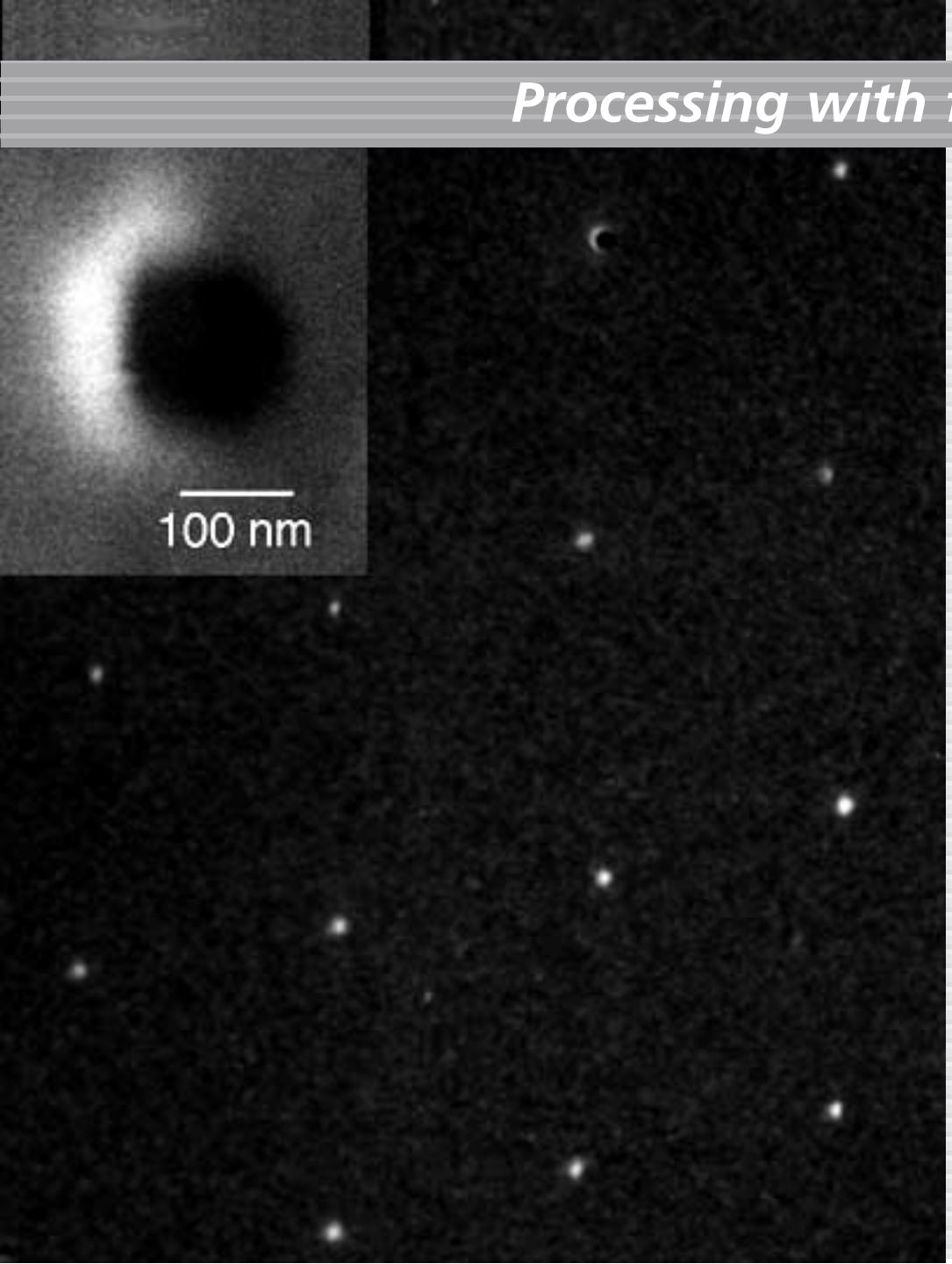


200 ps
9 µJ



100 fs
0.5 µJ

Processing with fs pulses



5 x 5 μm array

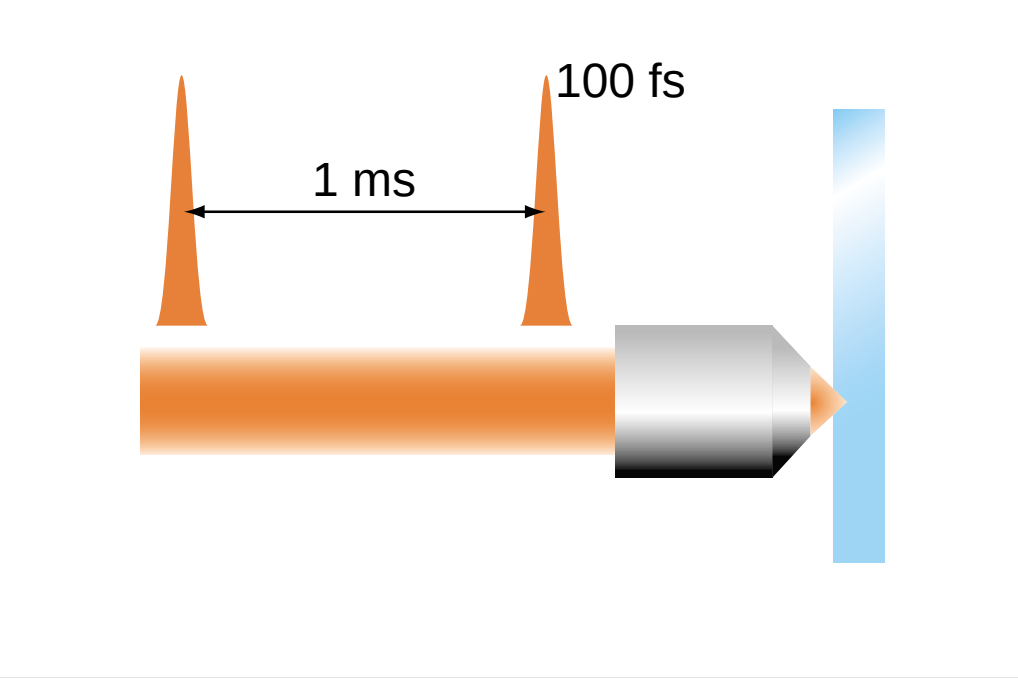
fused silica, 0.65 NA

0.5 μJ, 100 fs, 800 nm

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

Low-energy processing

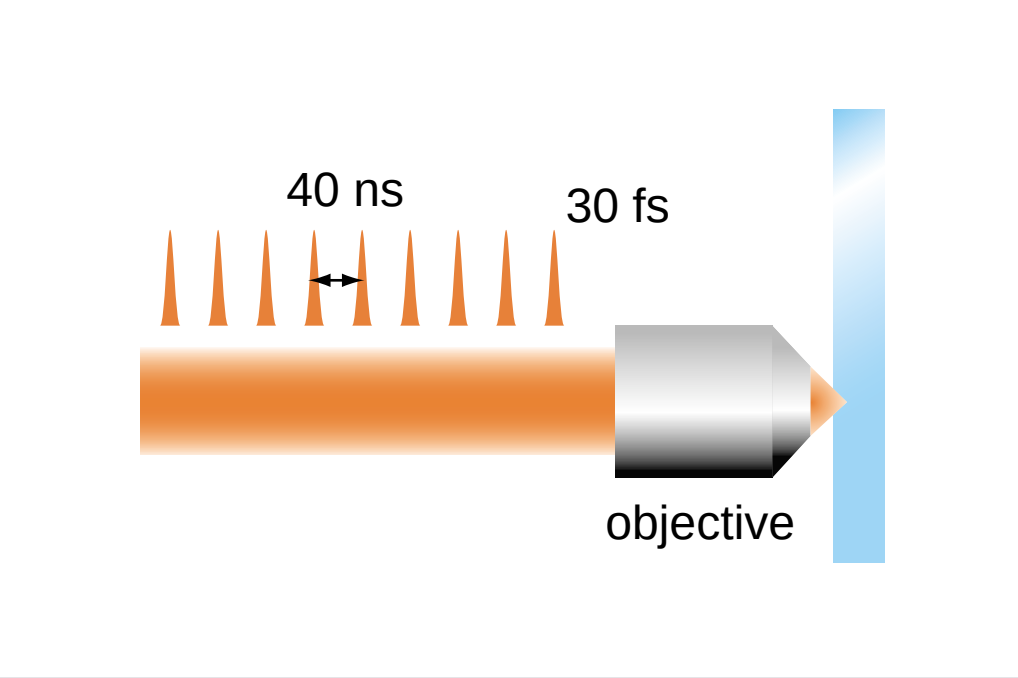
amplified laser



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

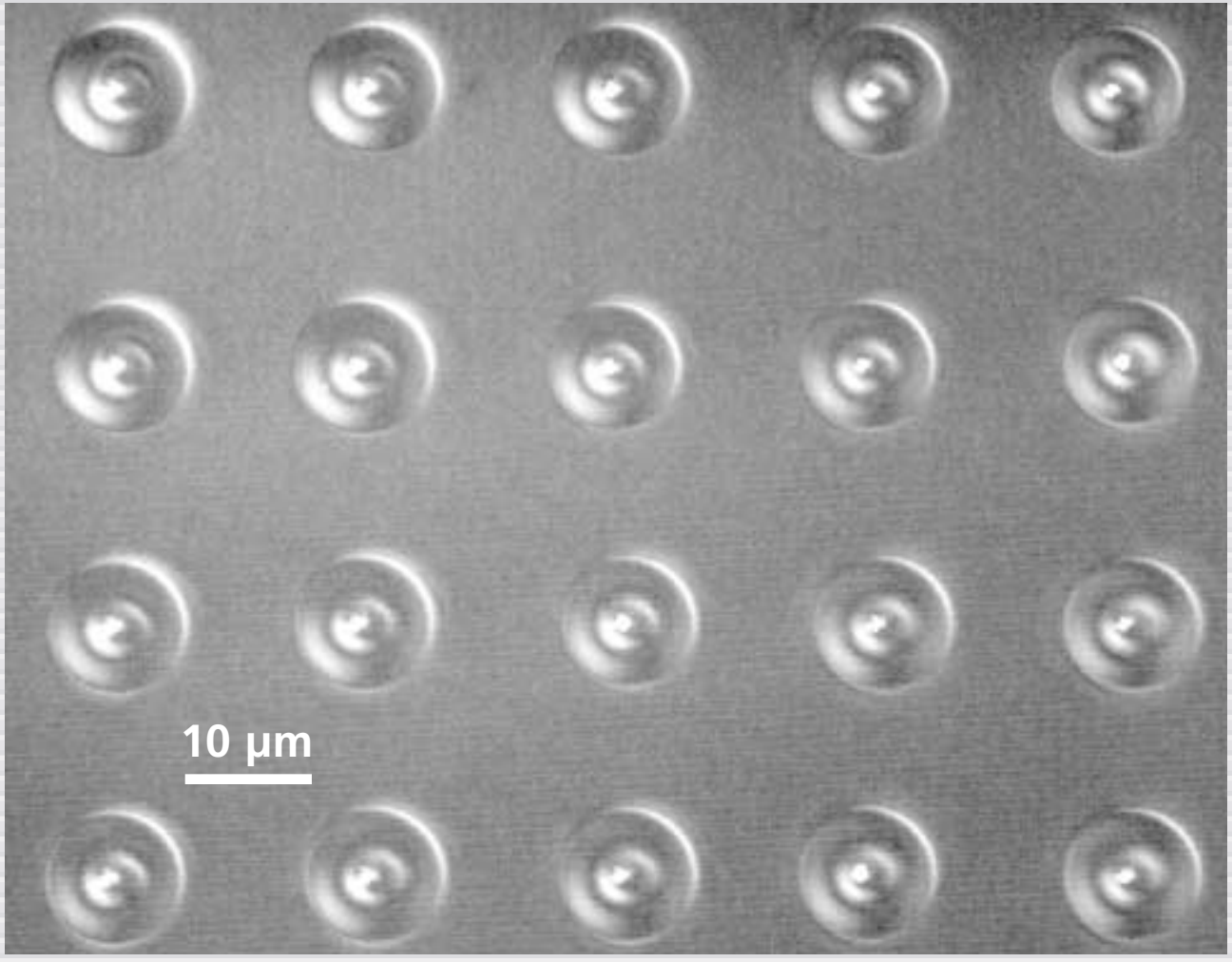
Low-energy processing

long-cavity Ti:sapphire oscillator

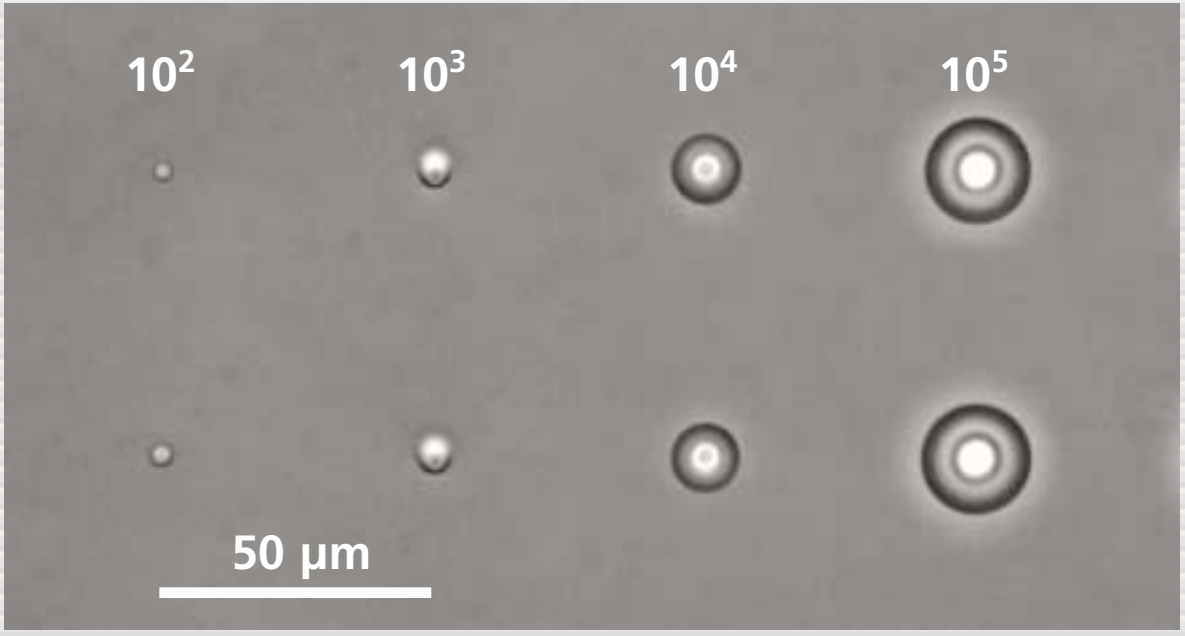


heat-diffusion time: $\tau_{diff} \approx 1\ \mu s$

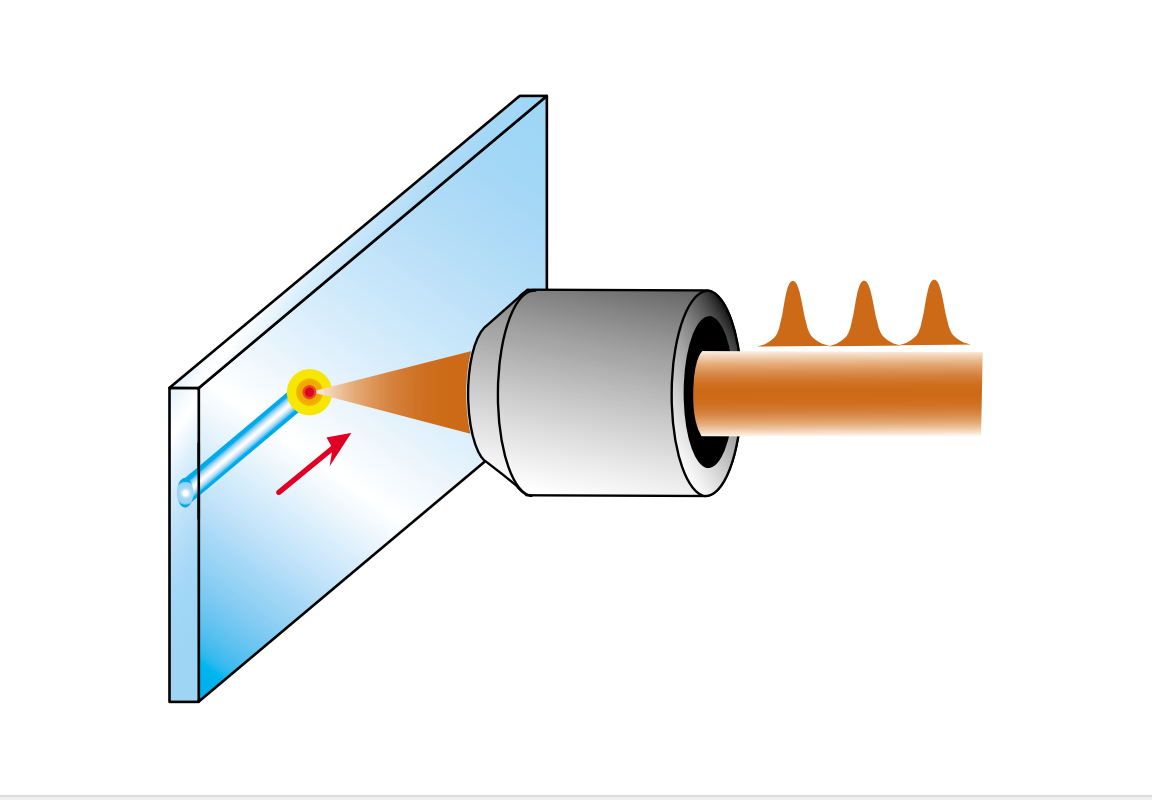
Low-energy processing



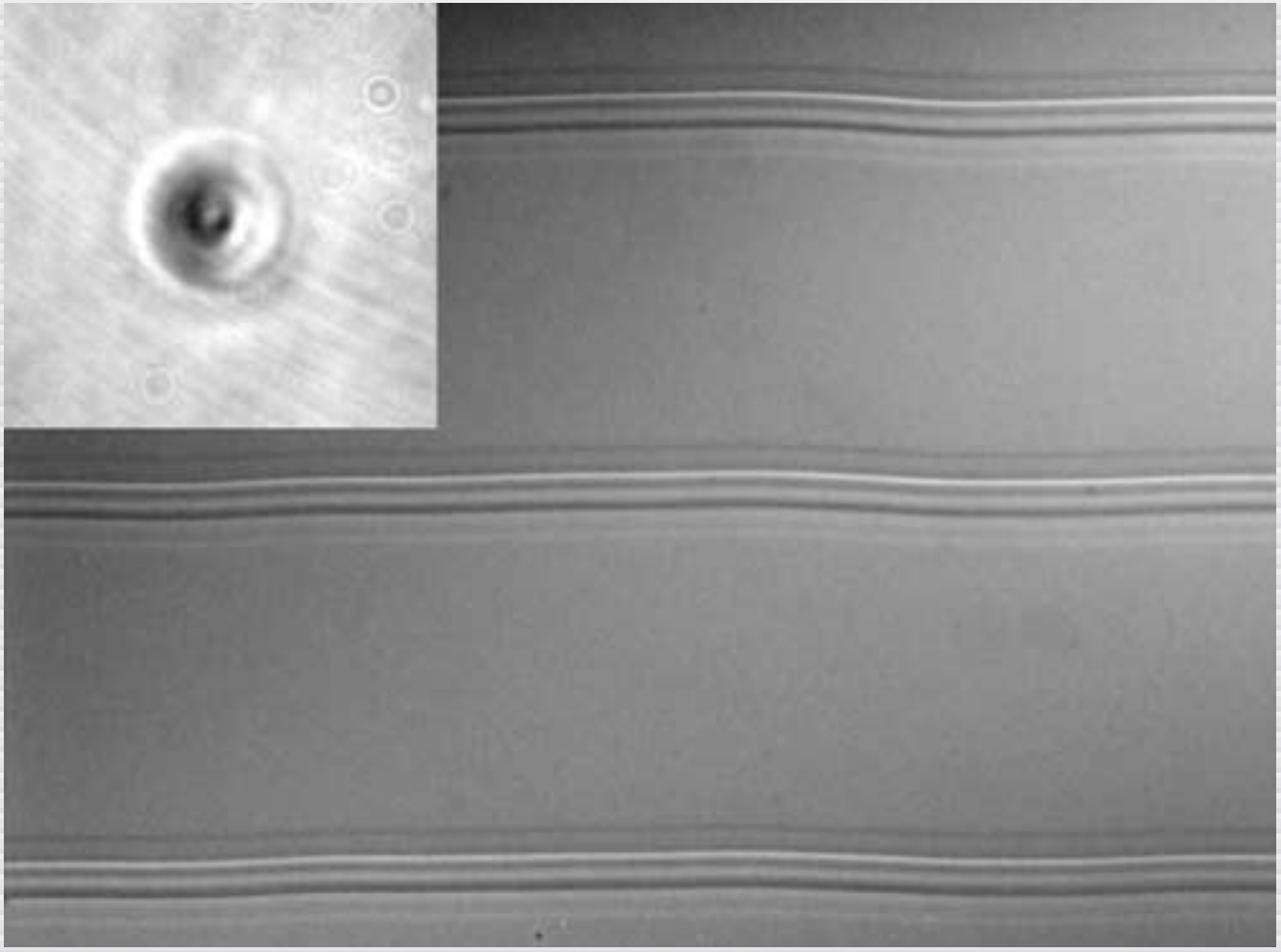
Low-energy processing



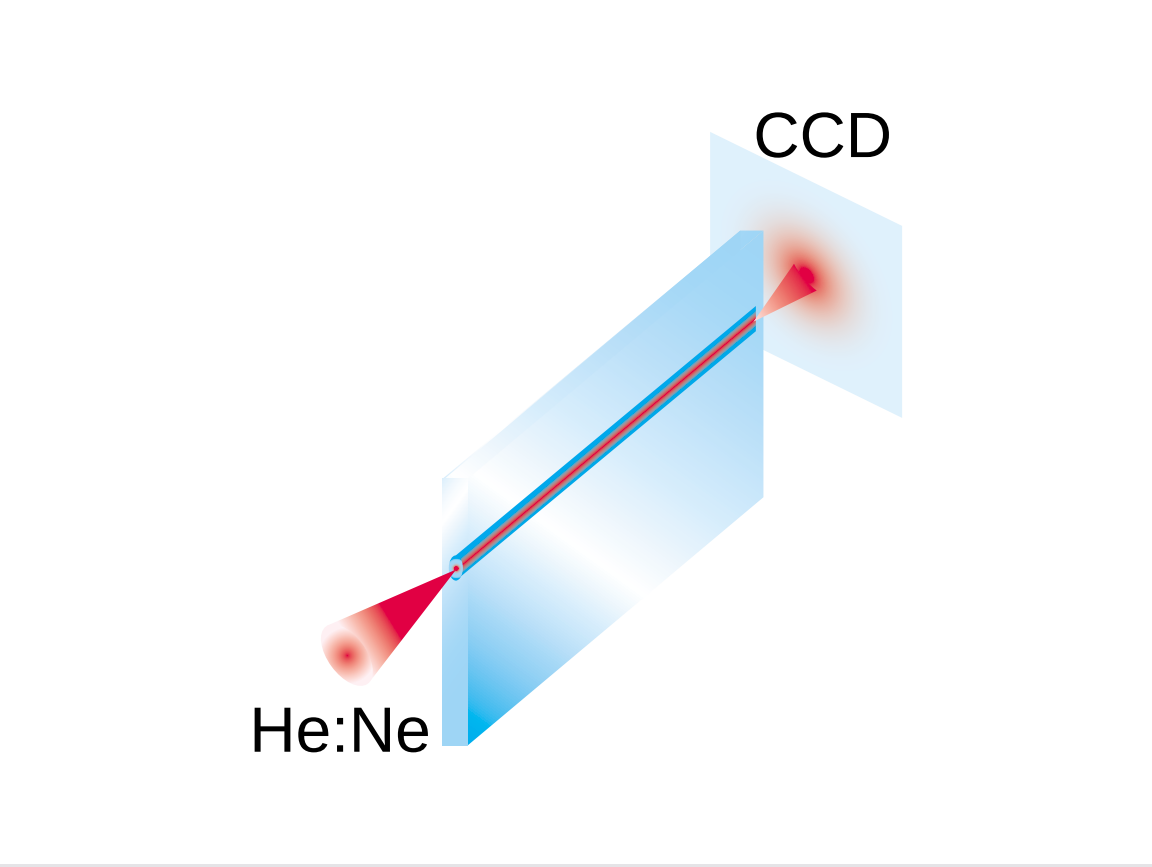
waveguide machining



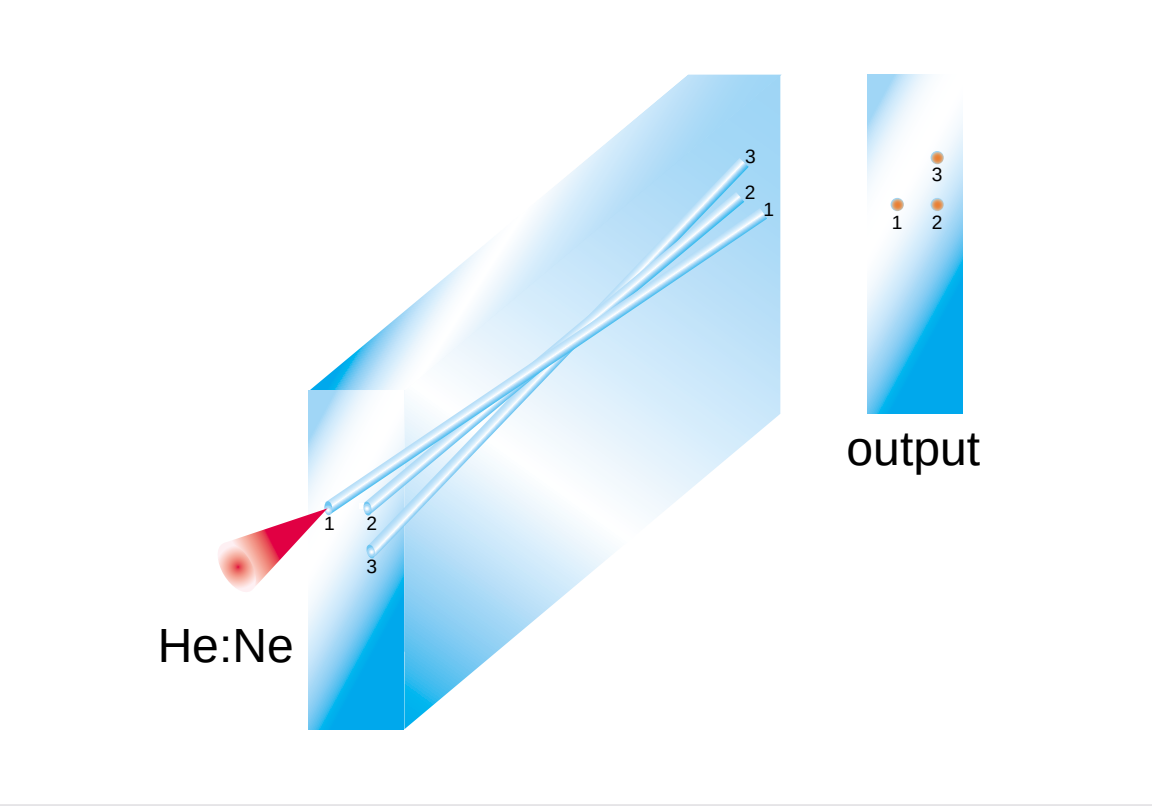
waveguide machining



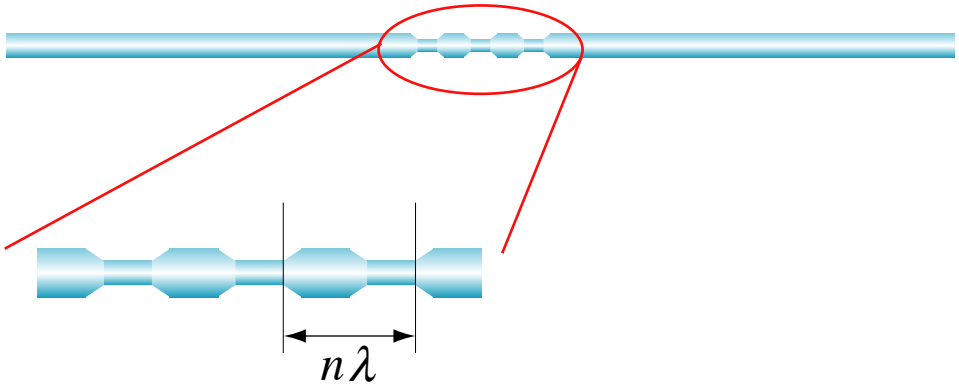
waveguide mode analysis



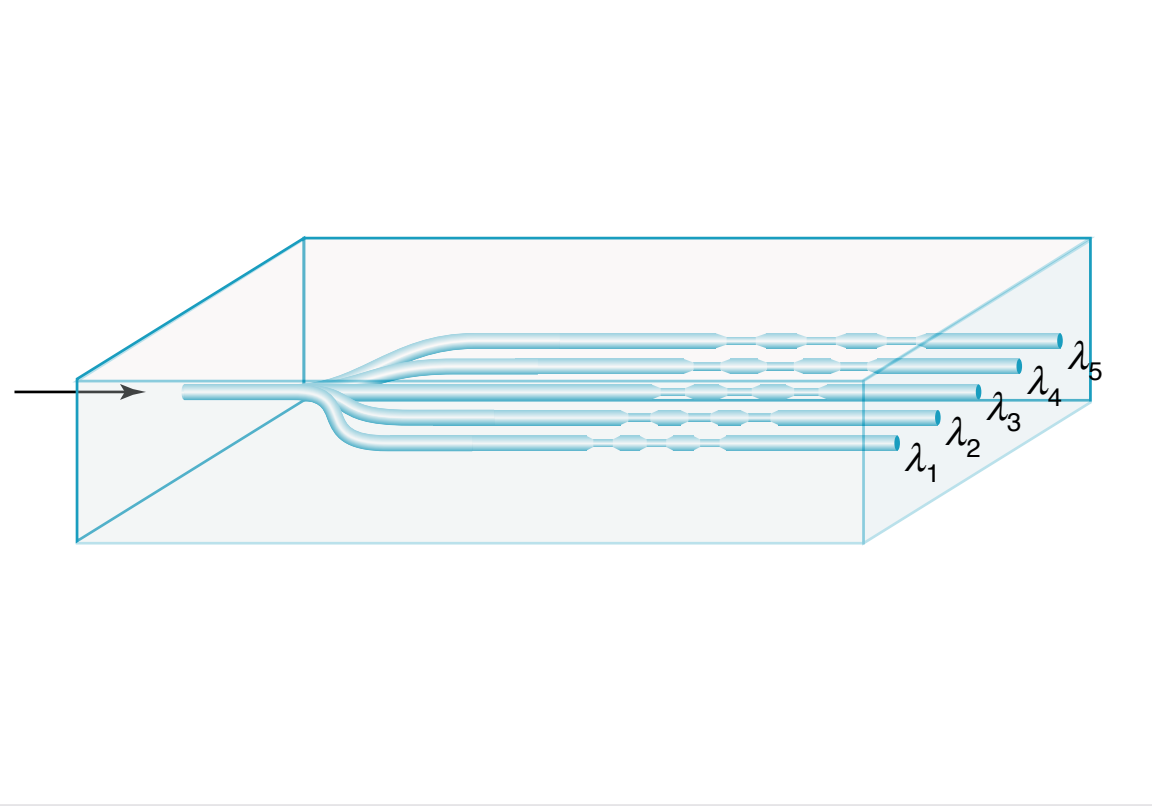
3D wave splitter



Bragg grating



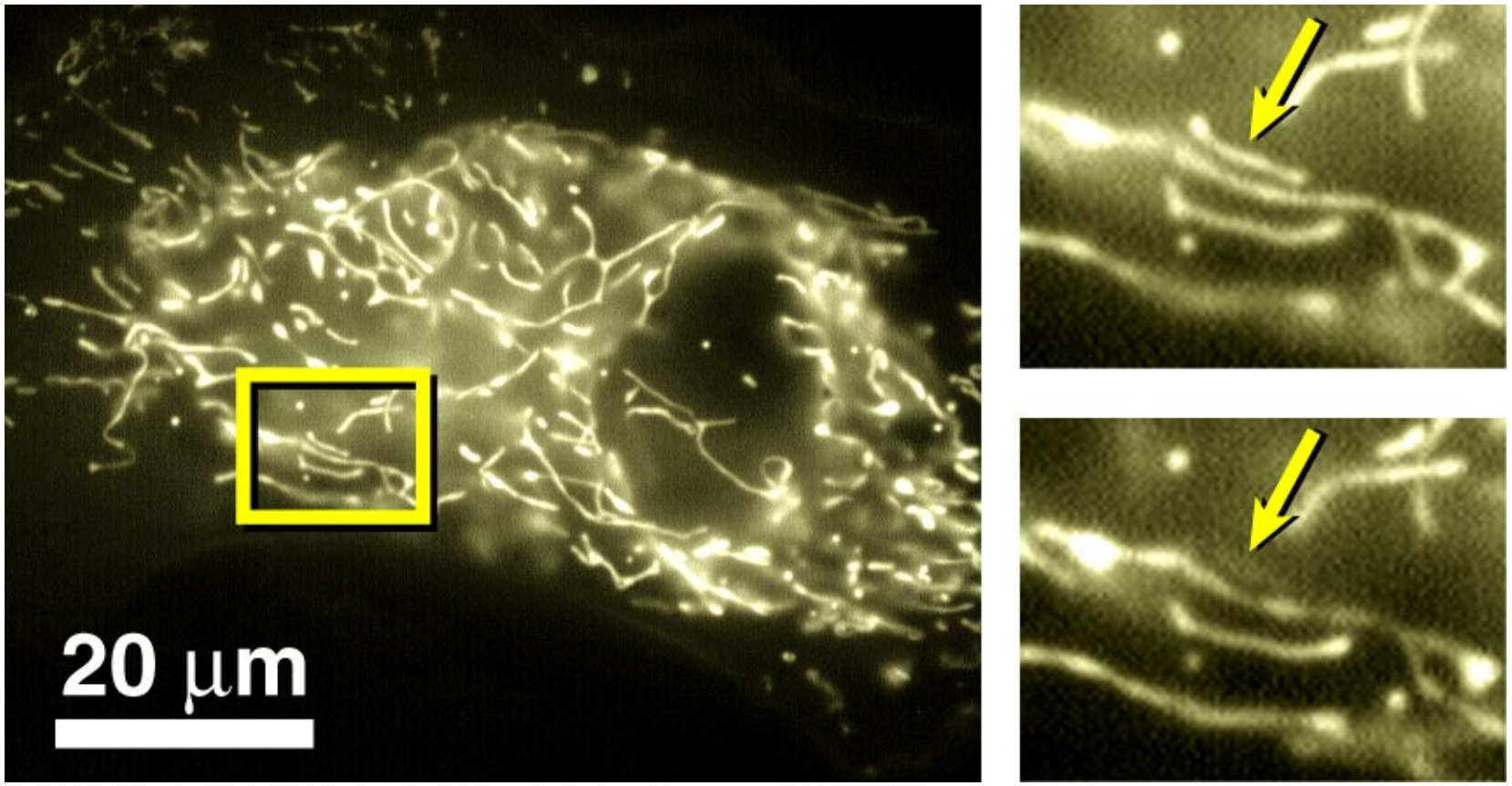
Bragg grating



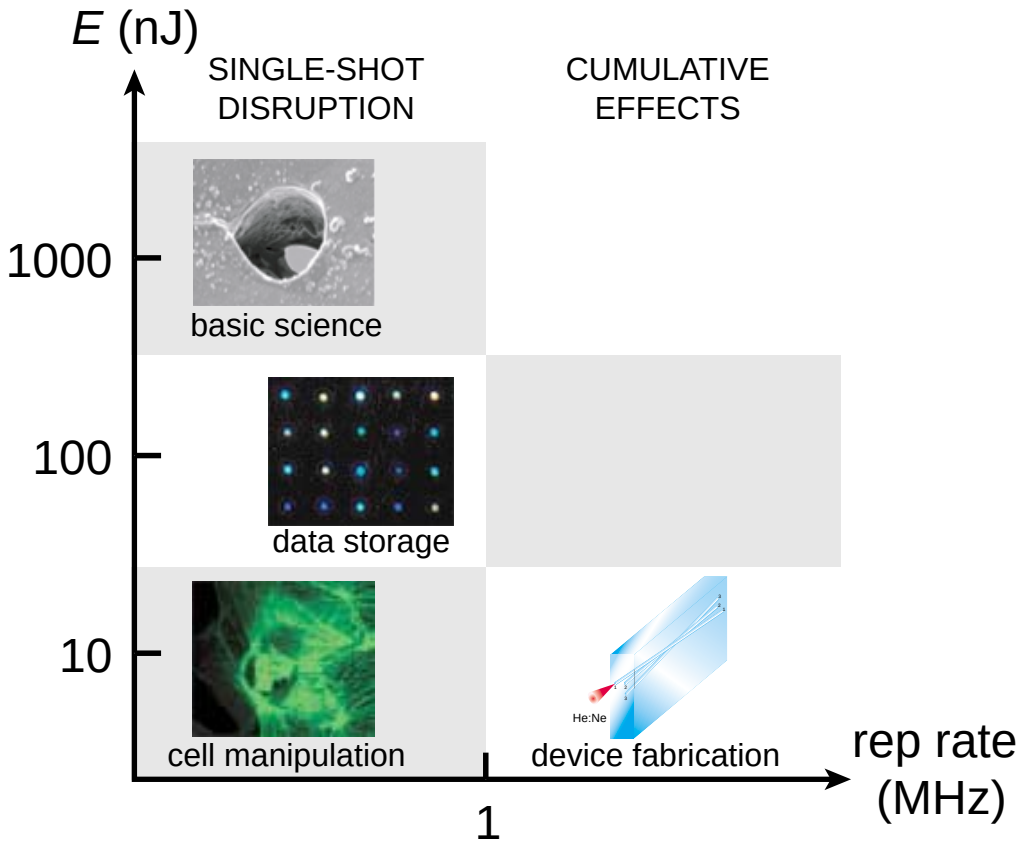
monolithic amplifier

laser active glass

Low-energy processing



Summary


E (nJ)
SINGLE-SHOT DISRUPTION
CUMULATIVE EFFECTS
1000
100
10
basic science
data storage
cell manipulation
He:Ne
device fabrication
1
rep rate (MHz)

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

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