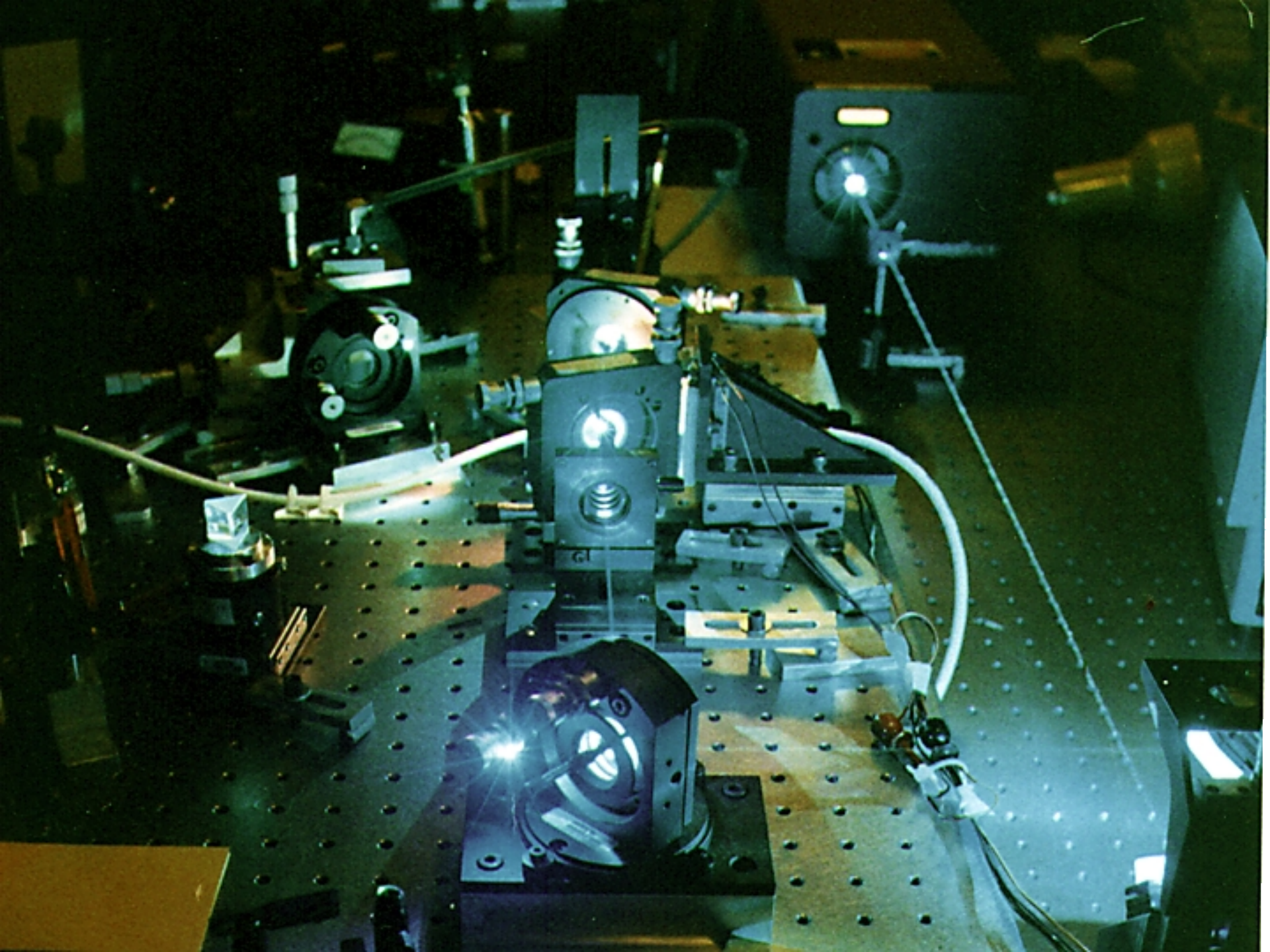
AN INTRODUCTION TO FEMTOSECOND LASER SCIENCE

Eric Mazur
Harvard University

Short Course SC541
Photonics West 2005, San Jose, CA



Outline

- linear and nonlinear propagation
- femtosecond measurements
- examples

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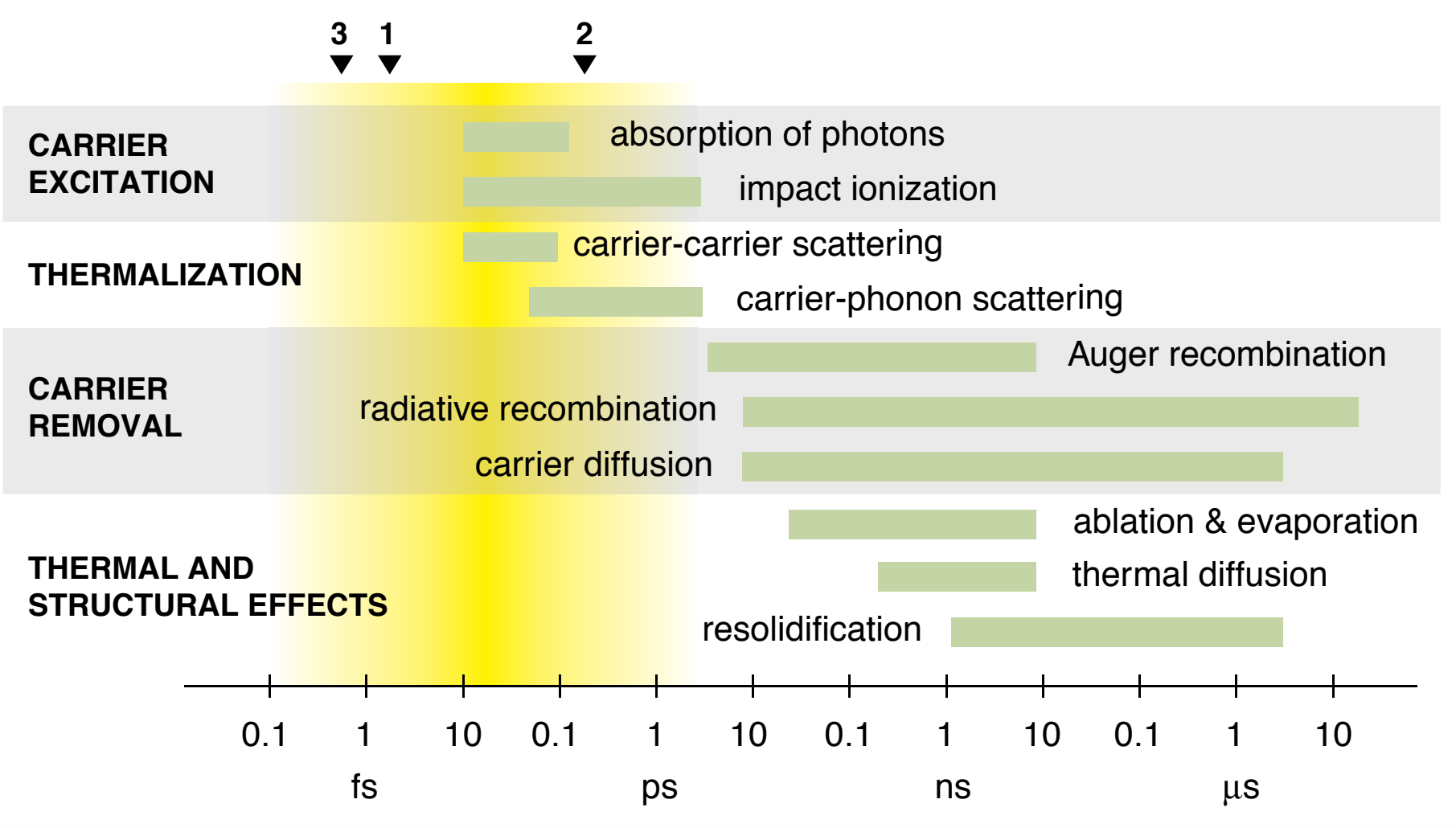

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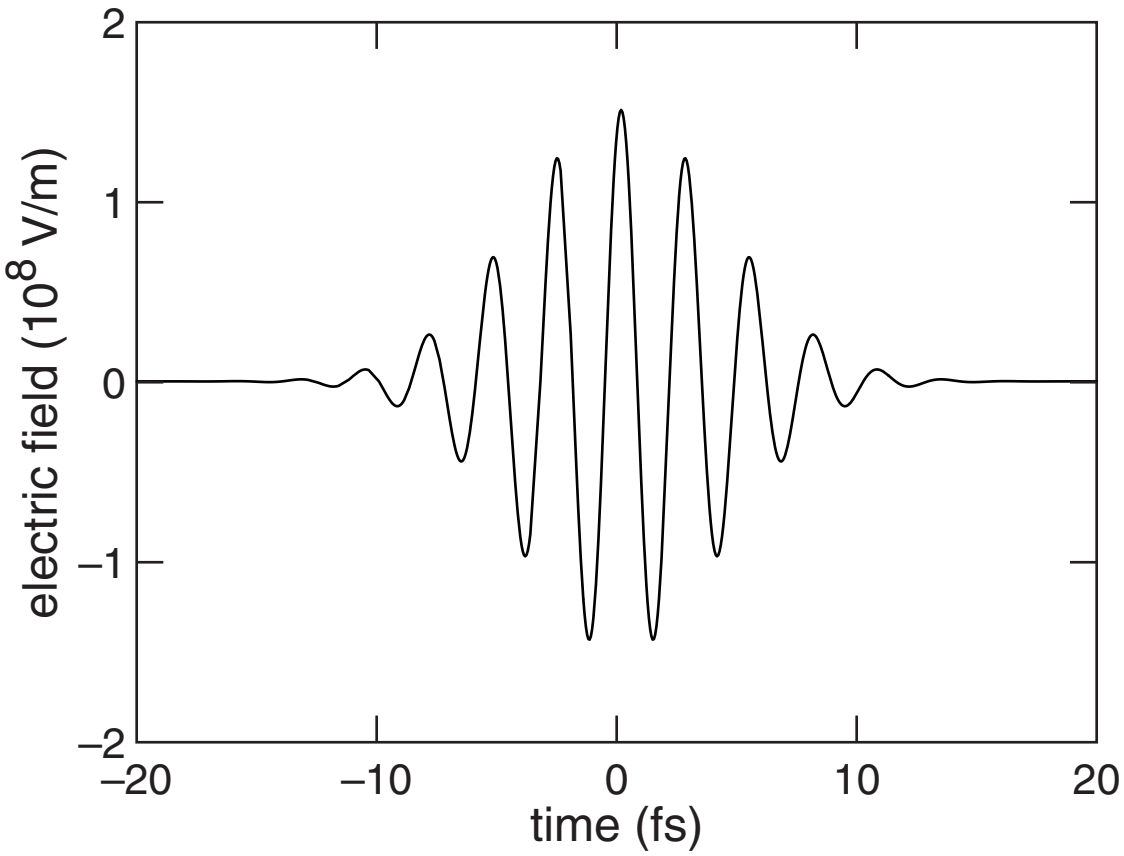

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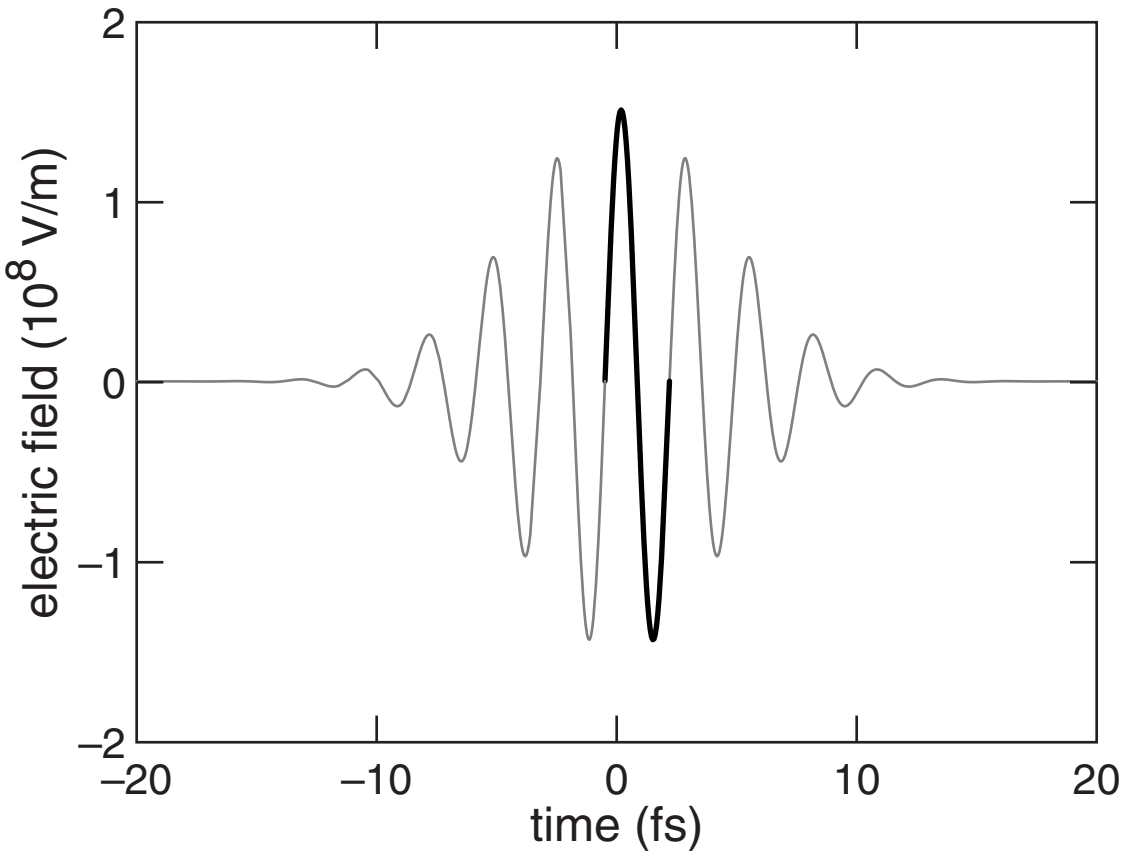

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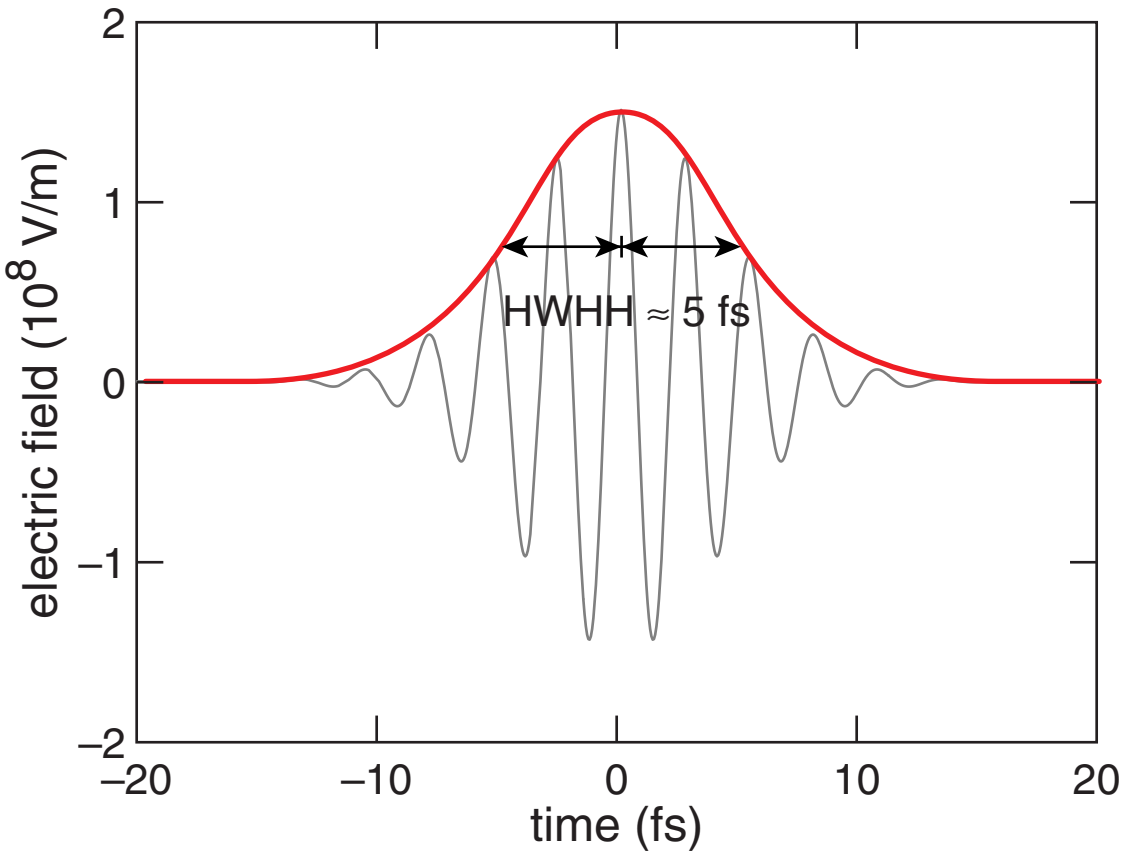

Introduction

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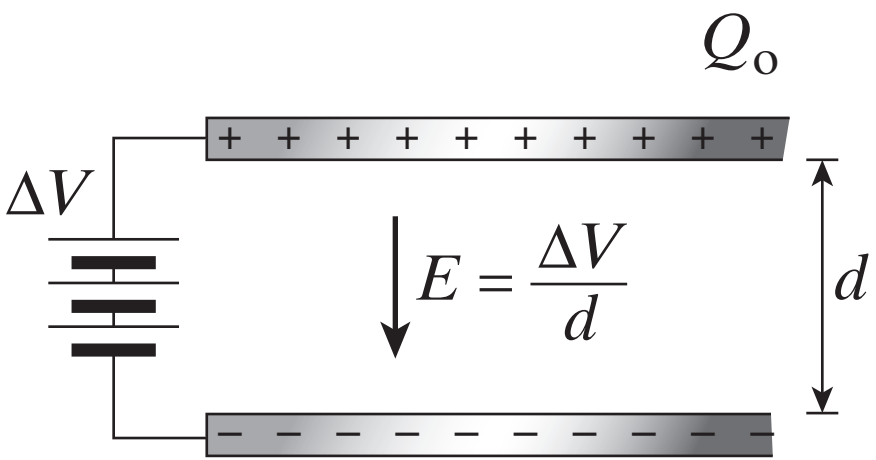
In non-ferromagnetic media $\mu \approx 1$**, and so** $n \approx \sqrt{\epsilon}$**.**

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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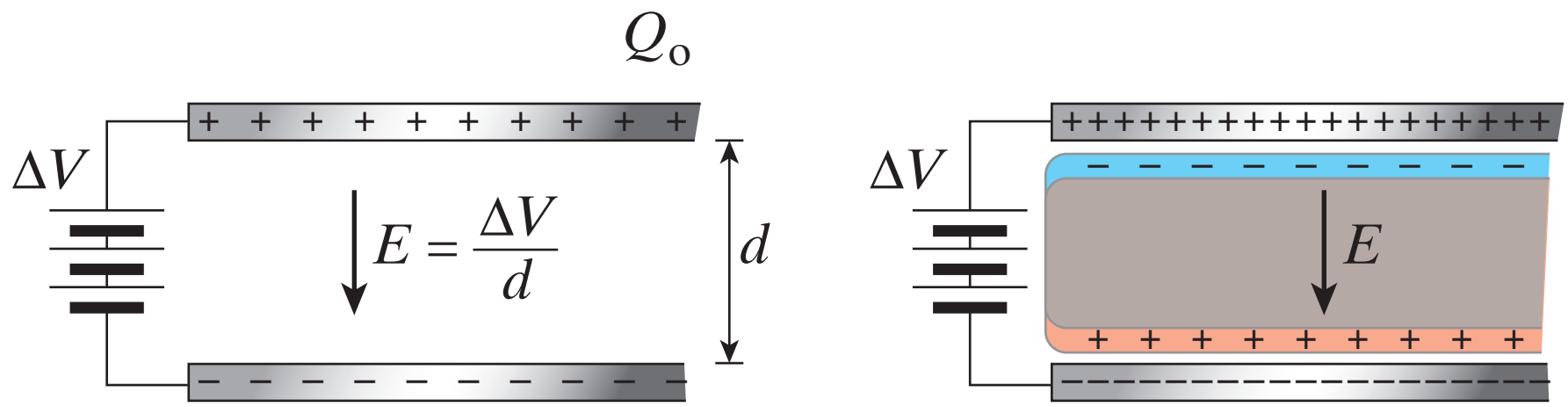

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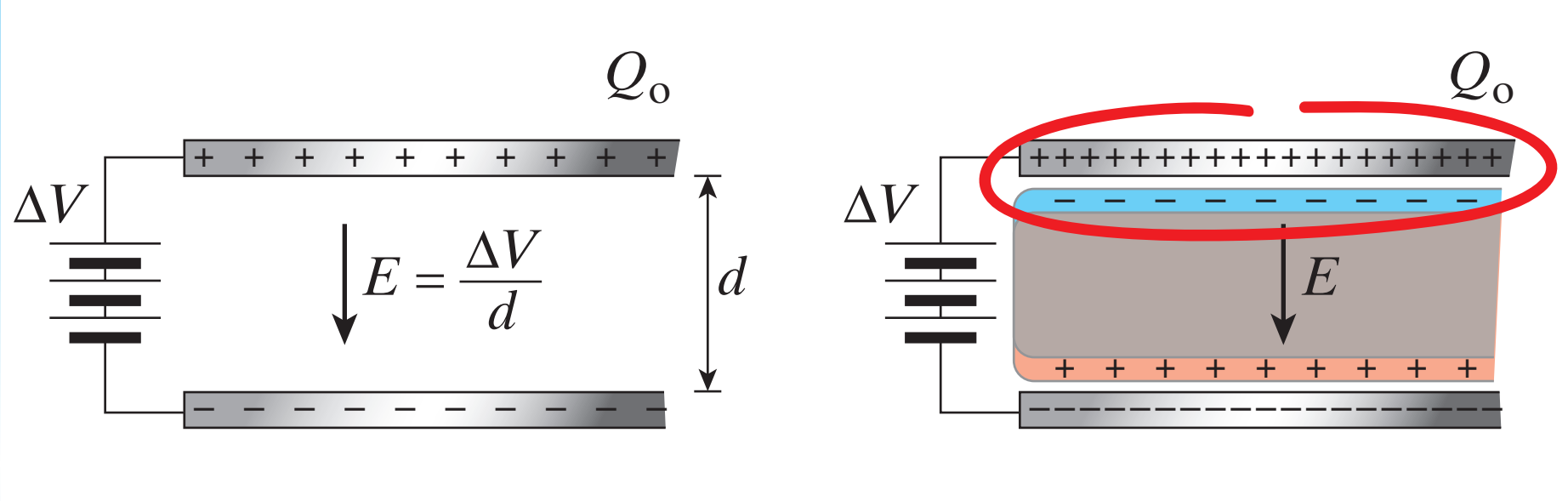

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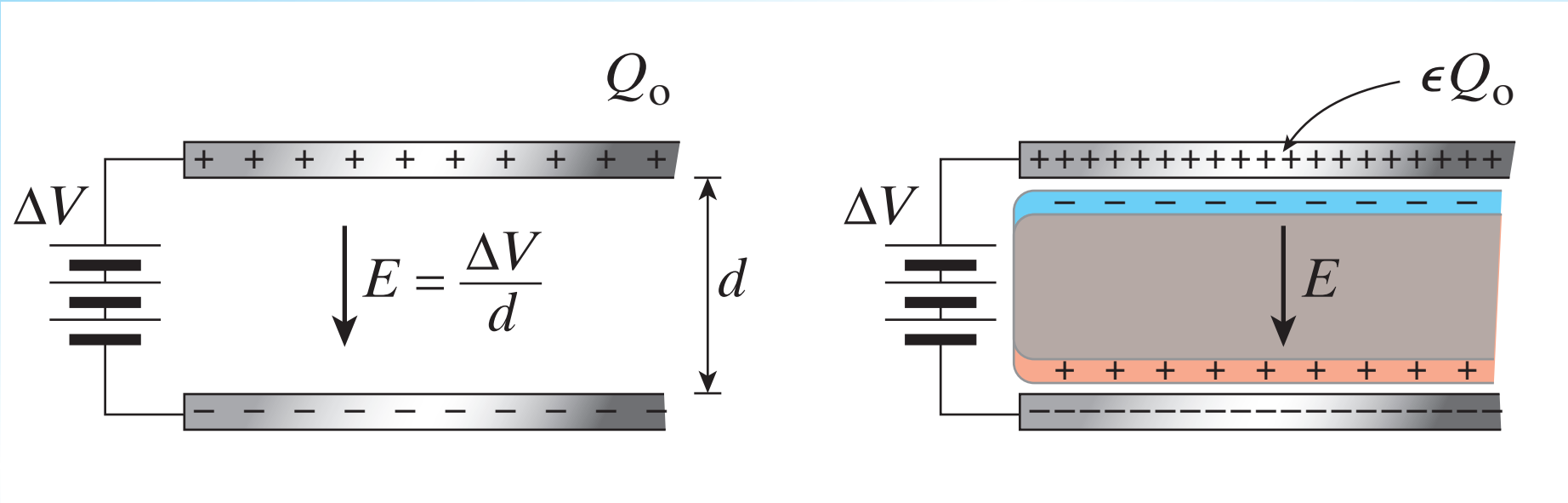
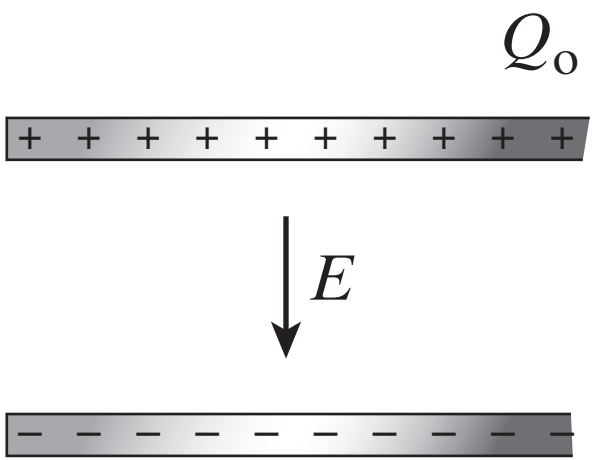


Propagation of EM waves through medium

Alternatively, ϵ is measure of the attenuation of the field



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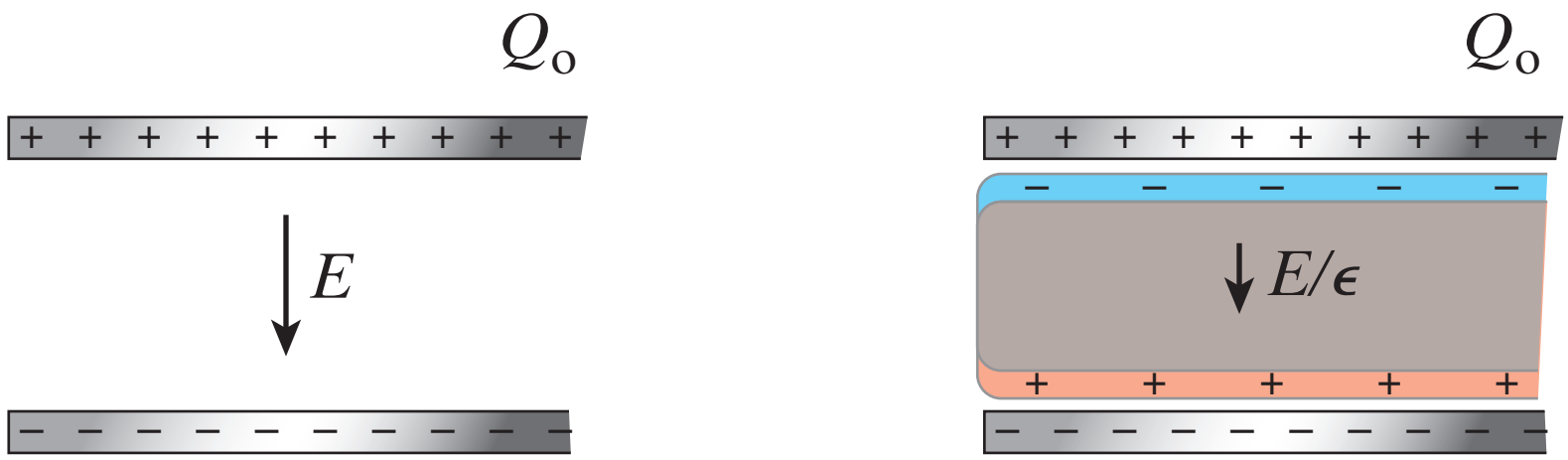
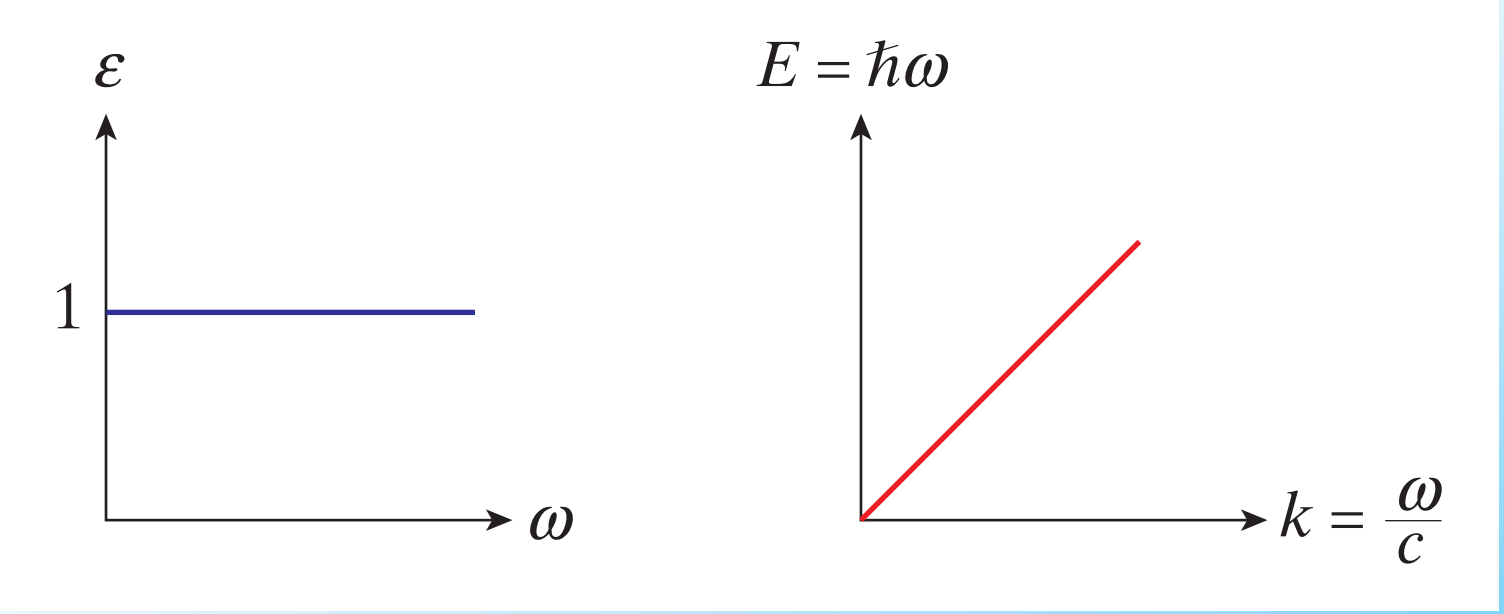


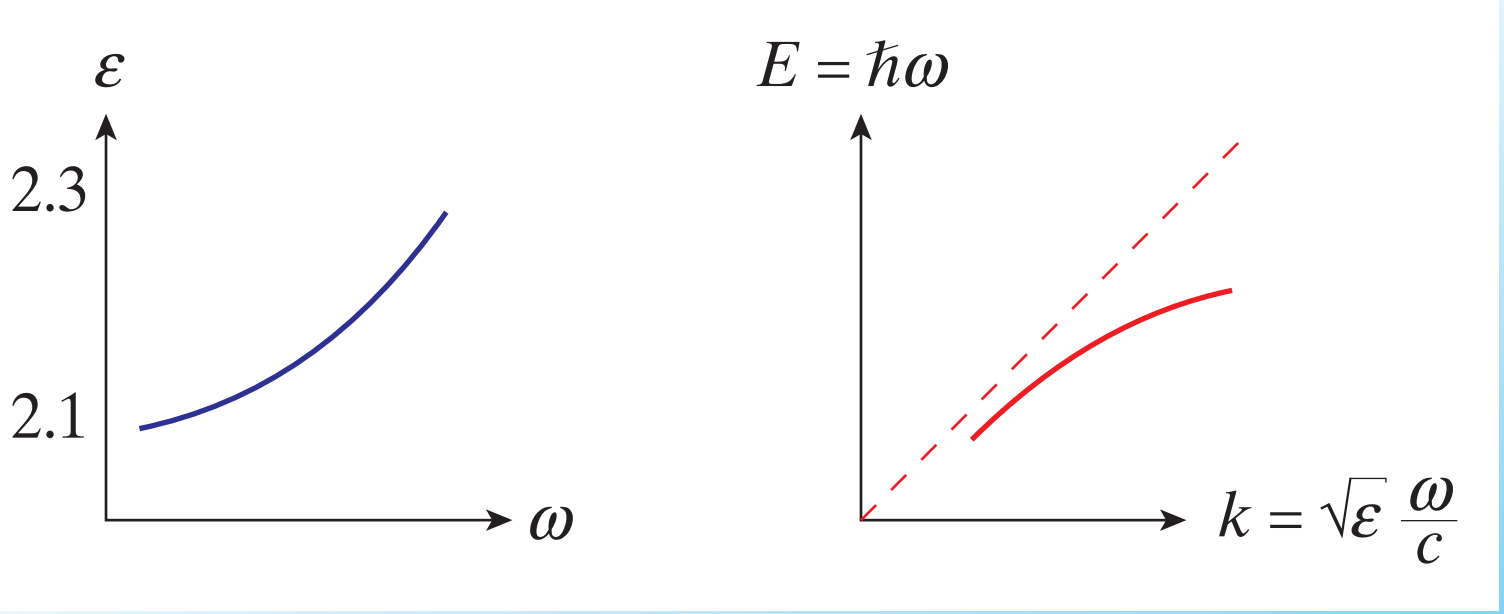
Propagation of EM waves through medium

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

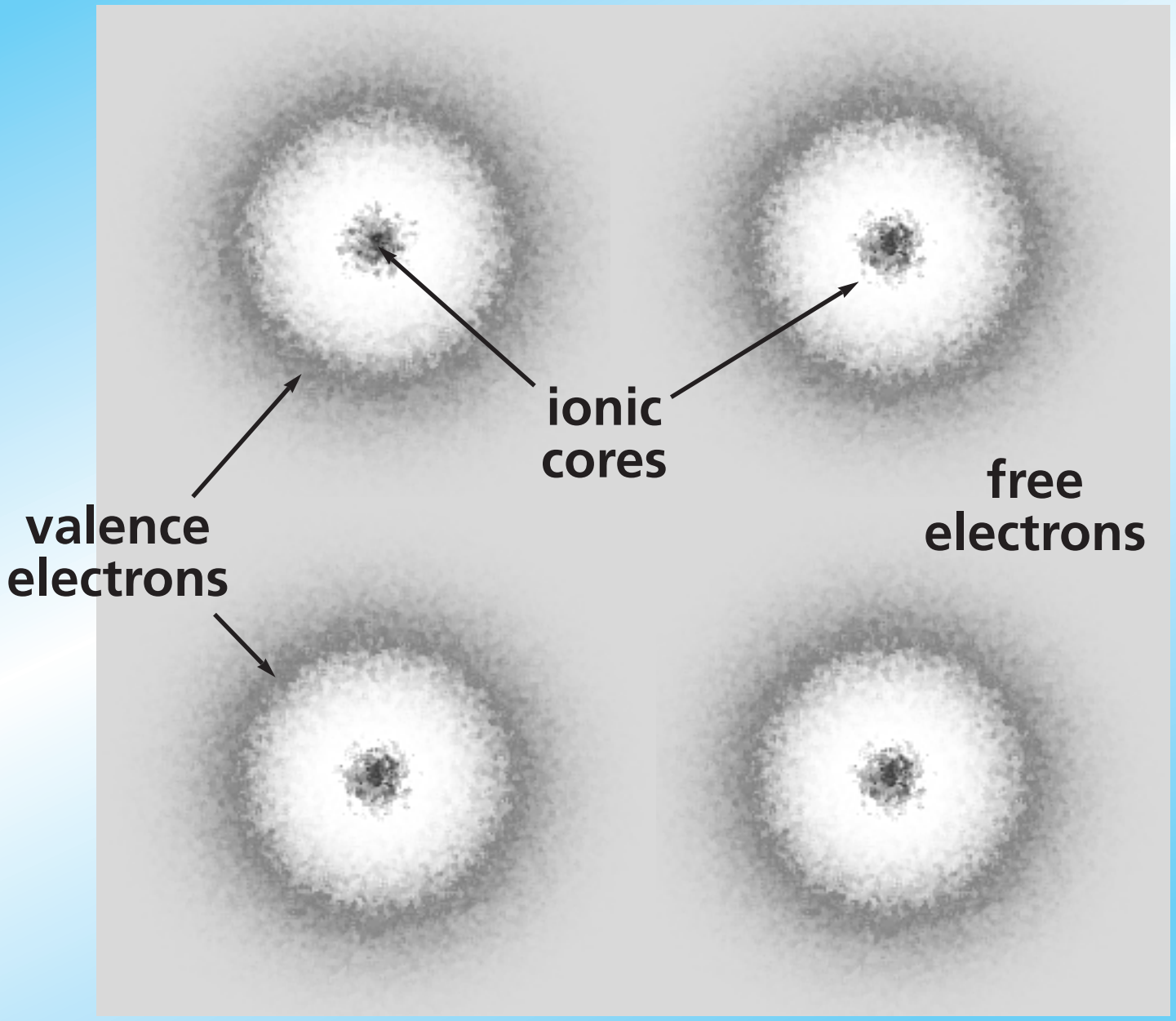


Propagation of EM waves through medium

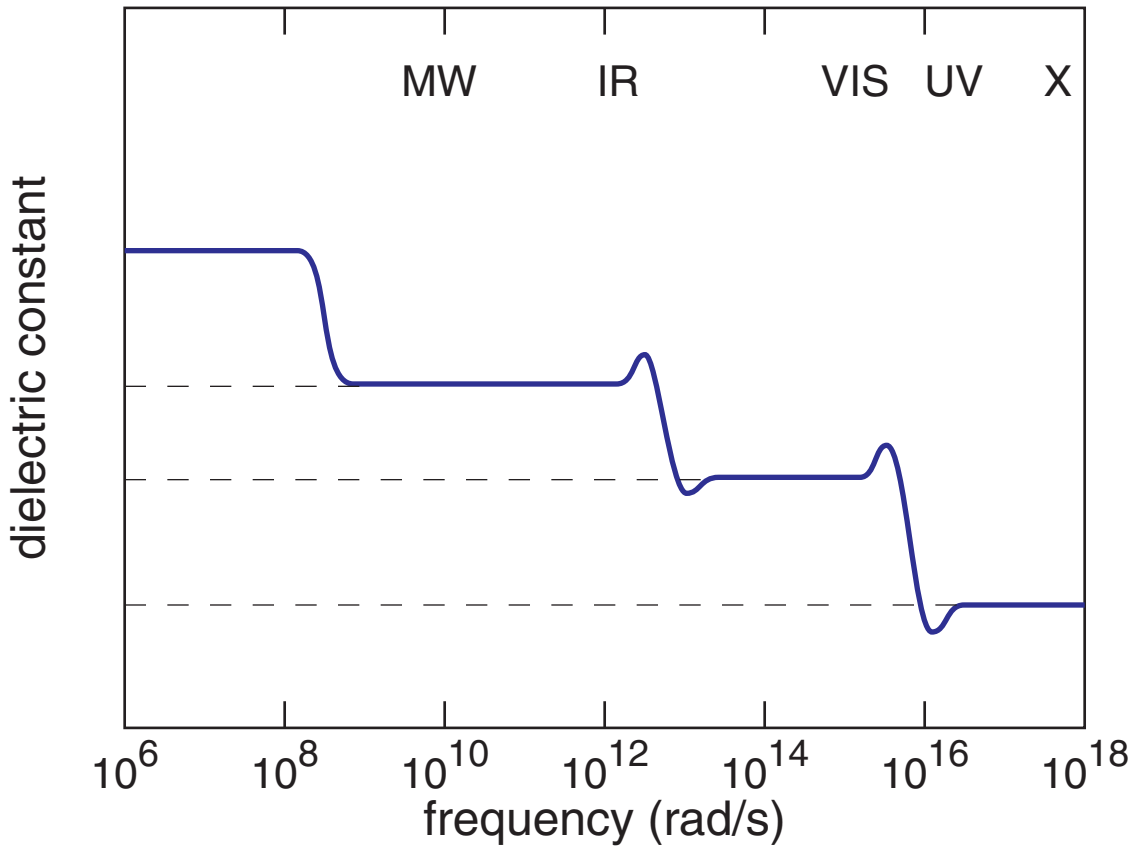
In medium: $$v = \frac{c}{\sqrt{\epsilon(\omega)}} = \frac{c}{n} \quad \Rightarrow \quad \omega = \frac{c}{\sqrt{\epsilon}} k$$



Which charges participate?



Dielectric function



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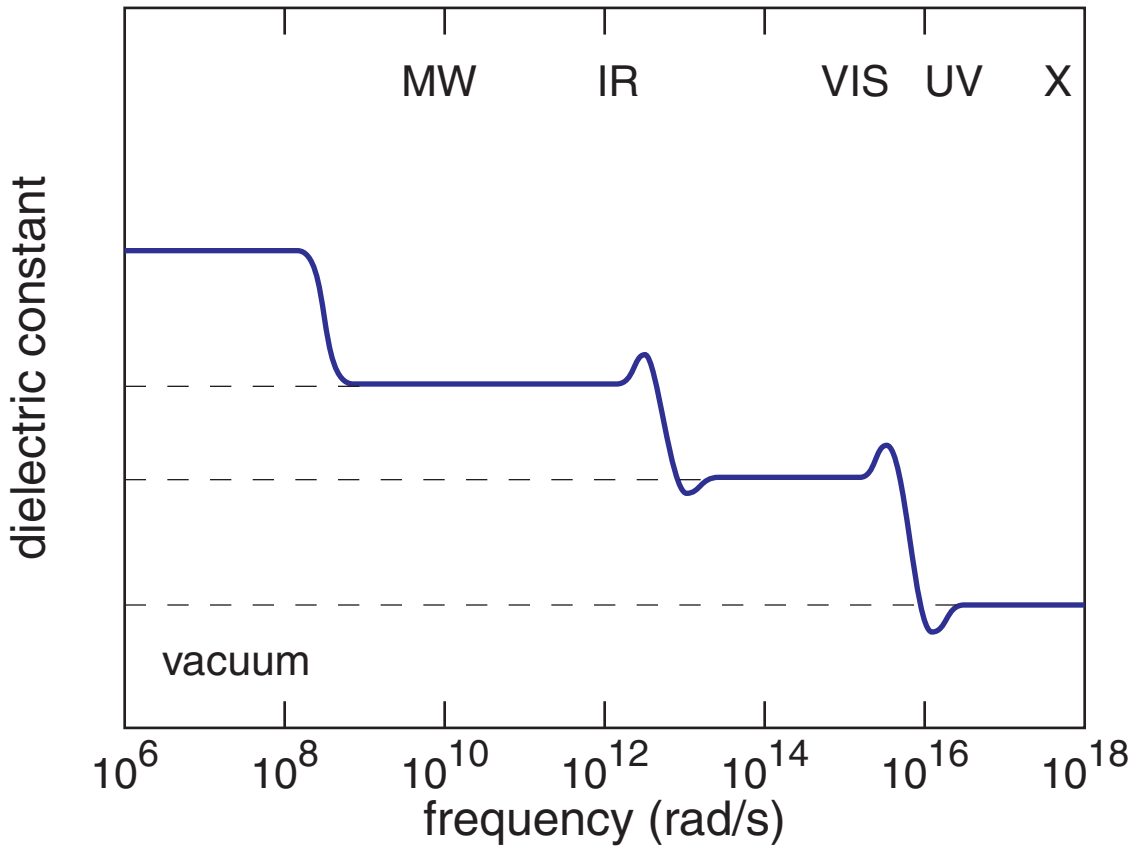

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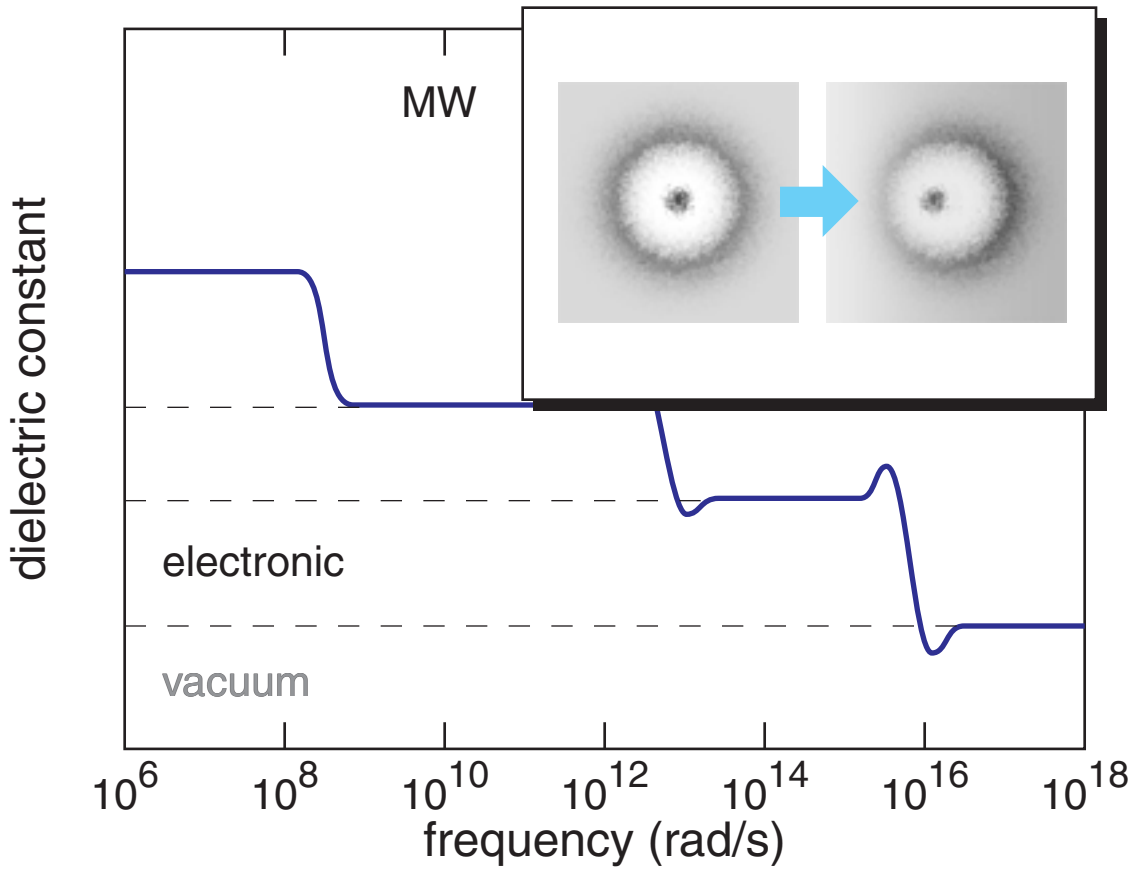

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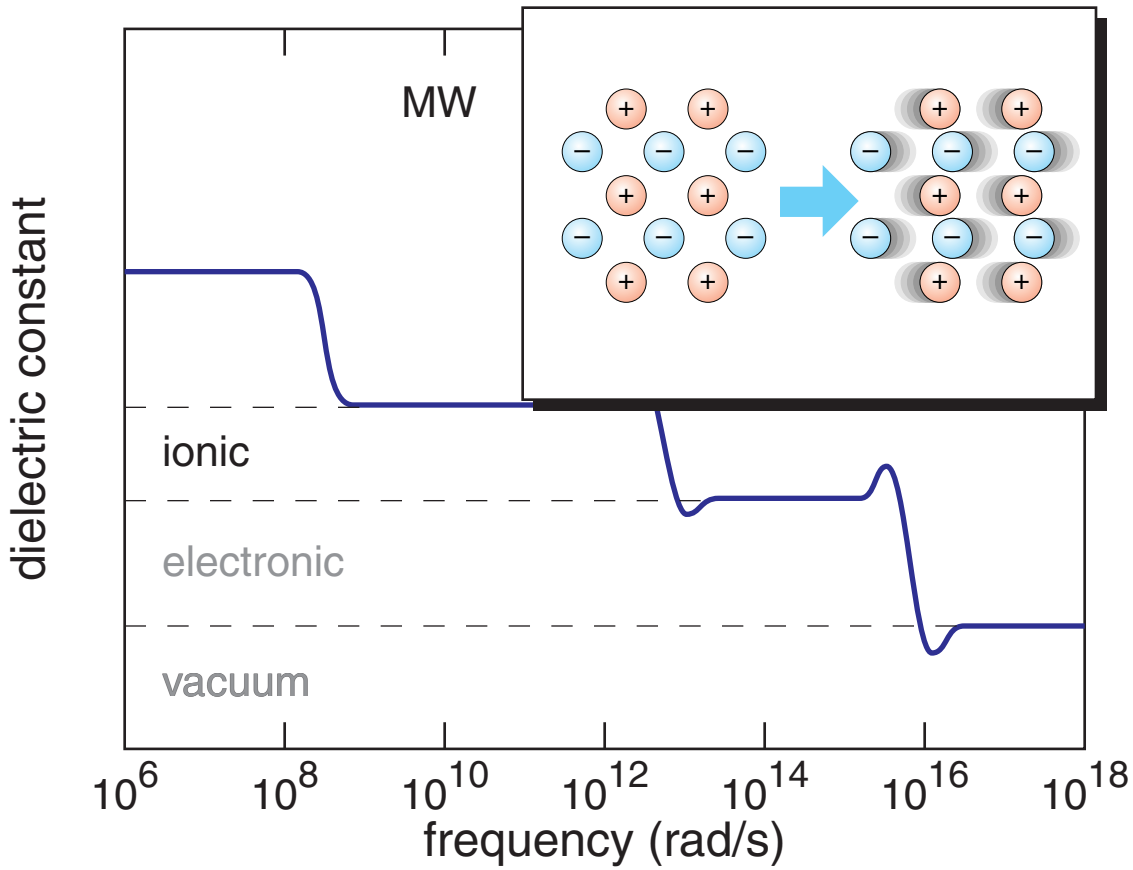

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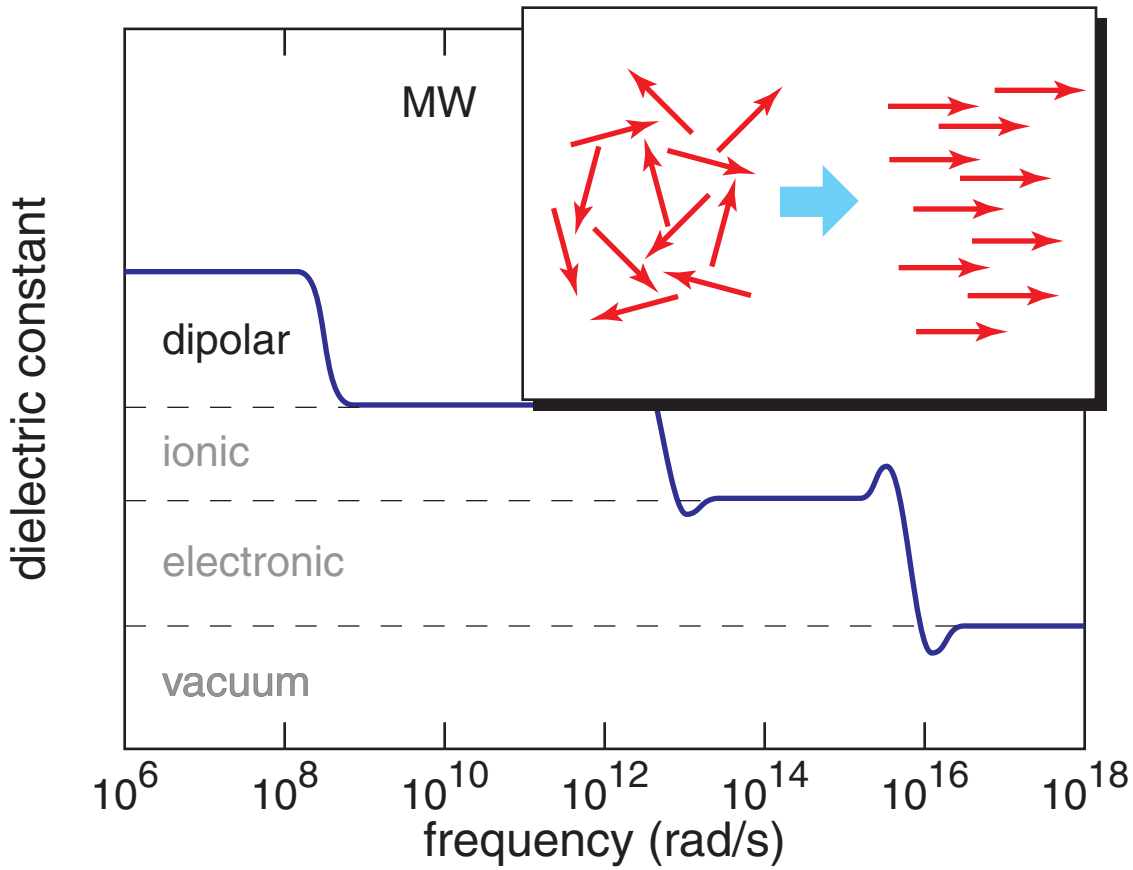

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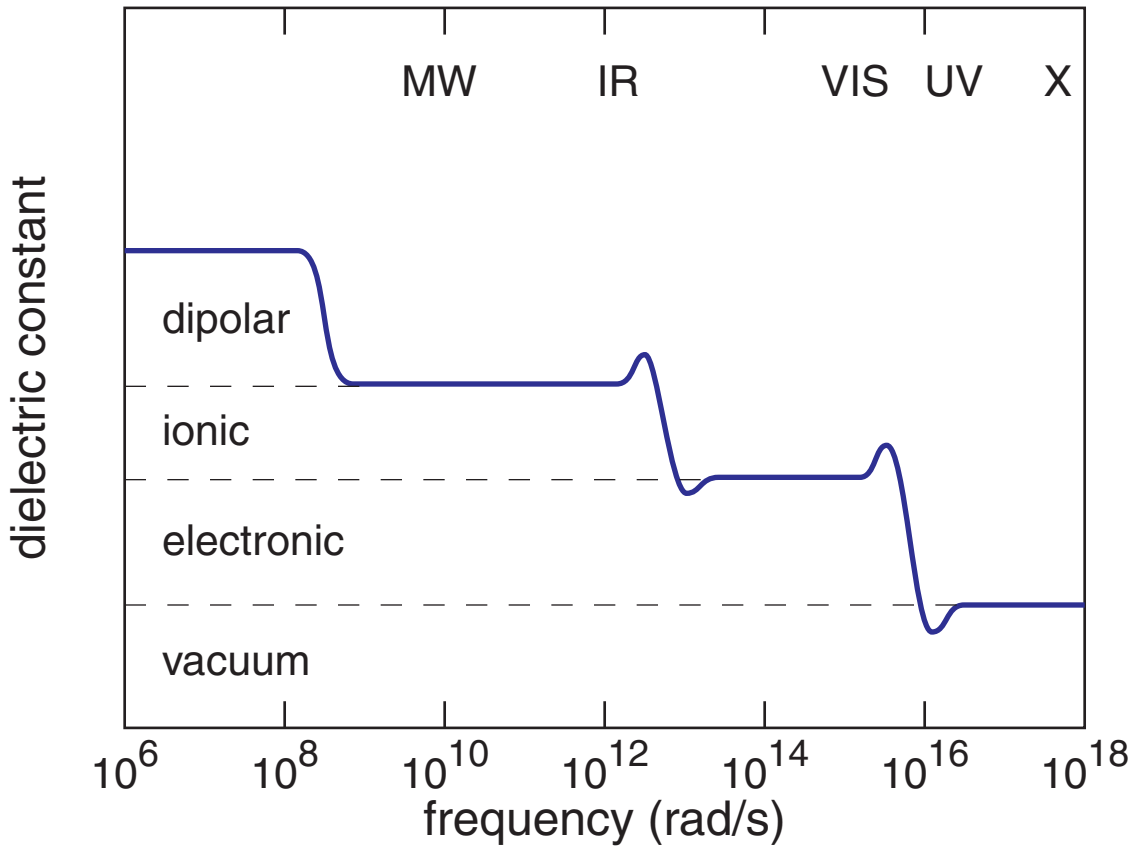

Bound electrons

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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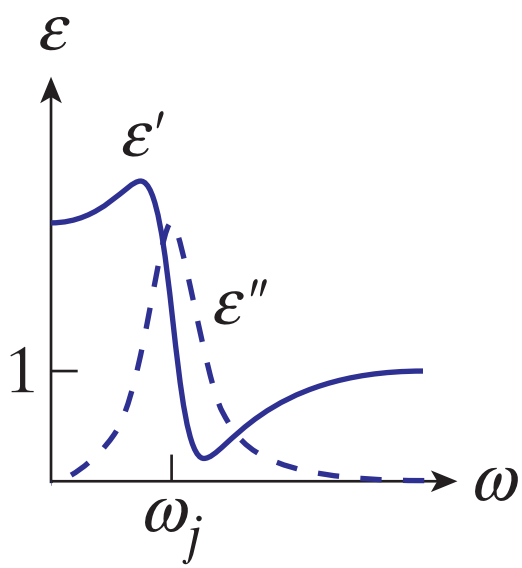
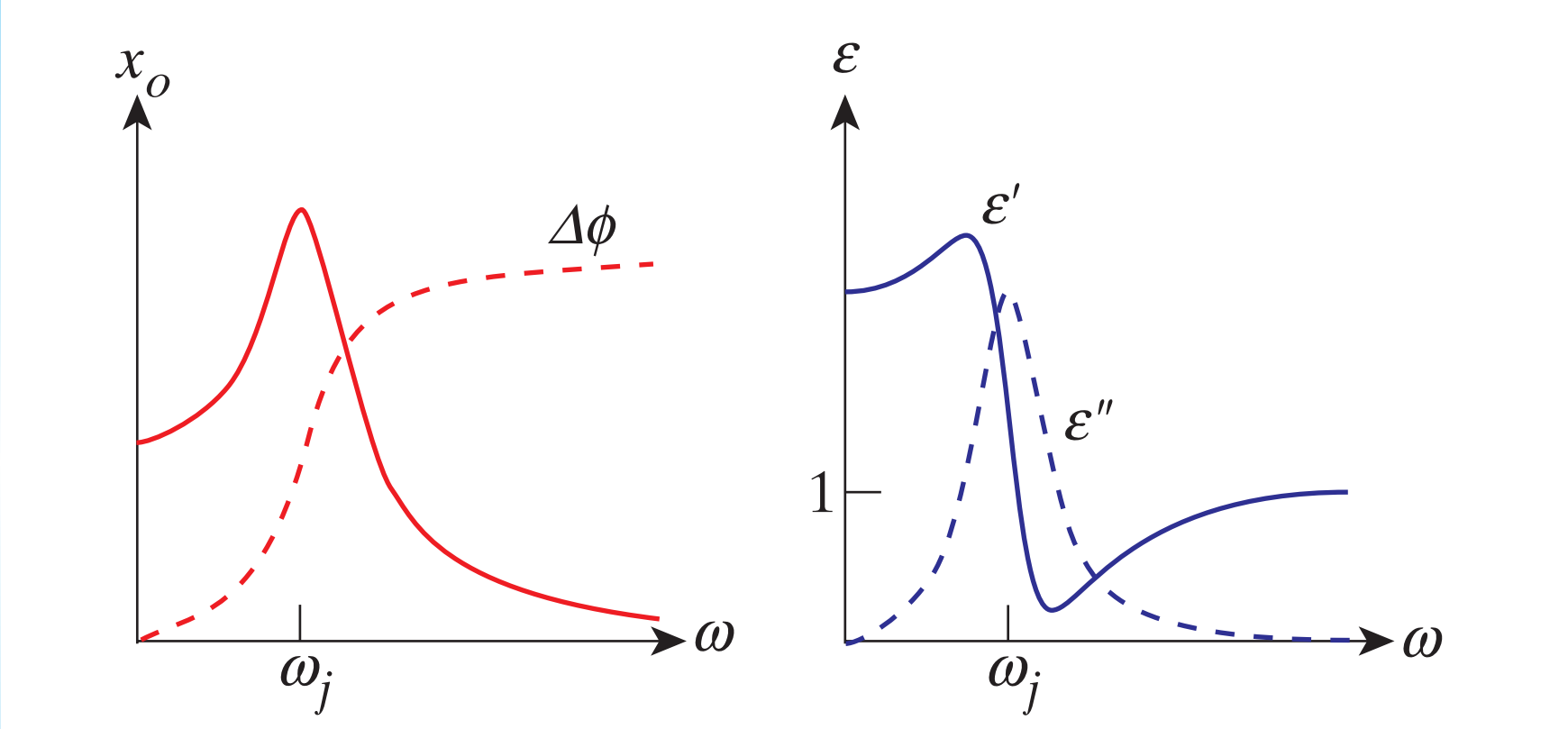


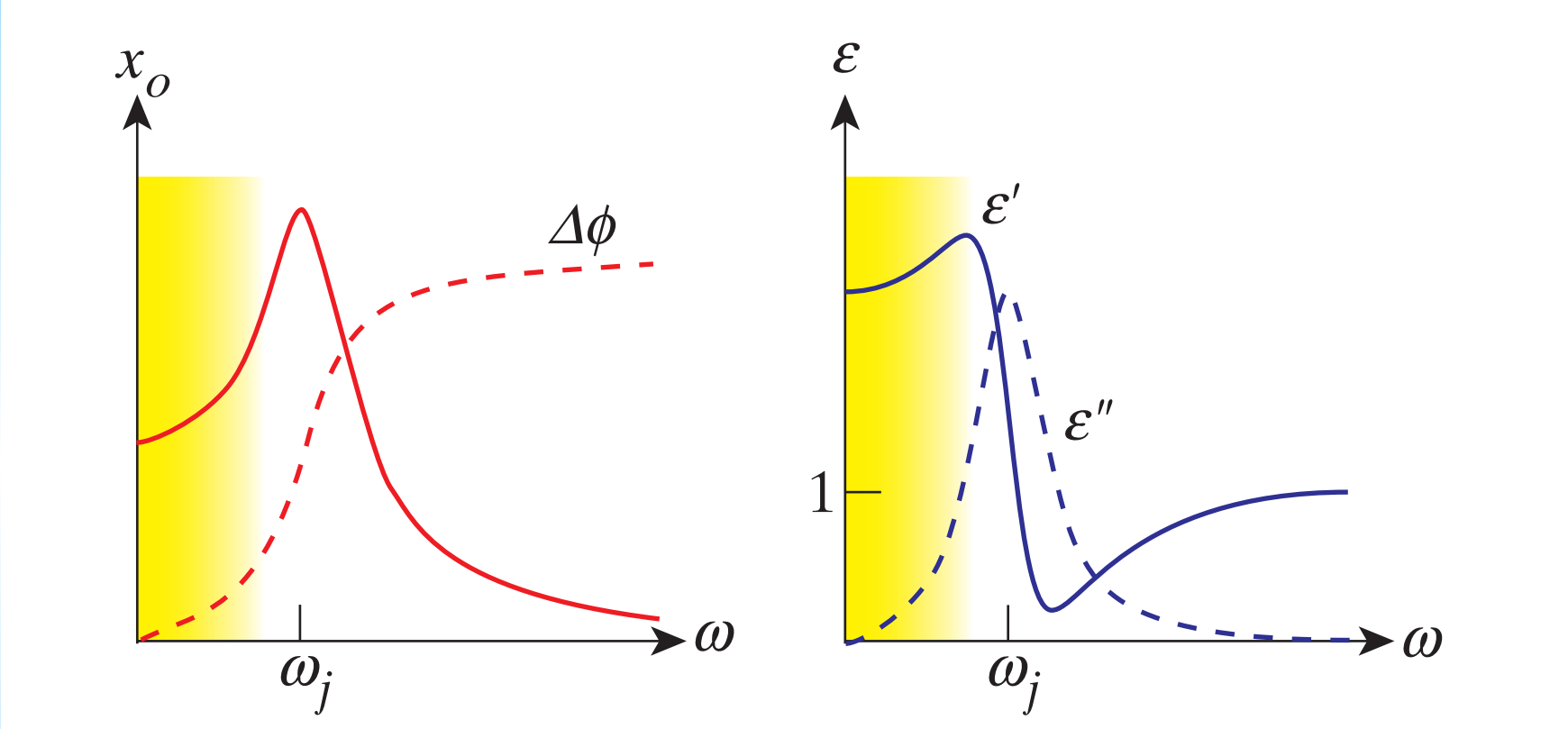
Bound electrons

amplitude of bound charge oscillation



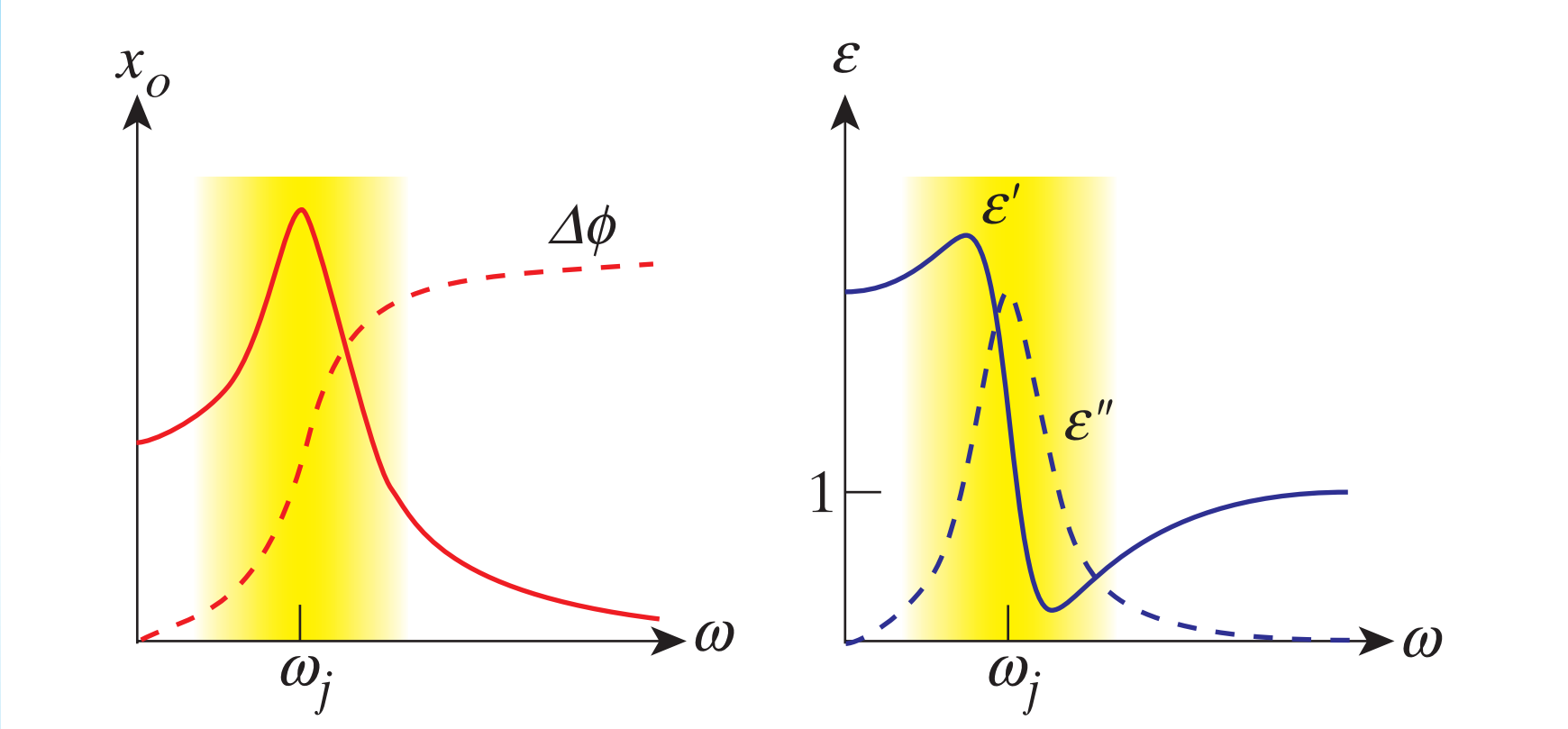
Bound electrons

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



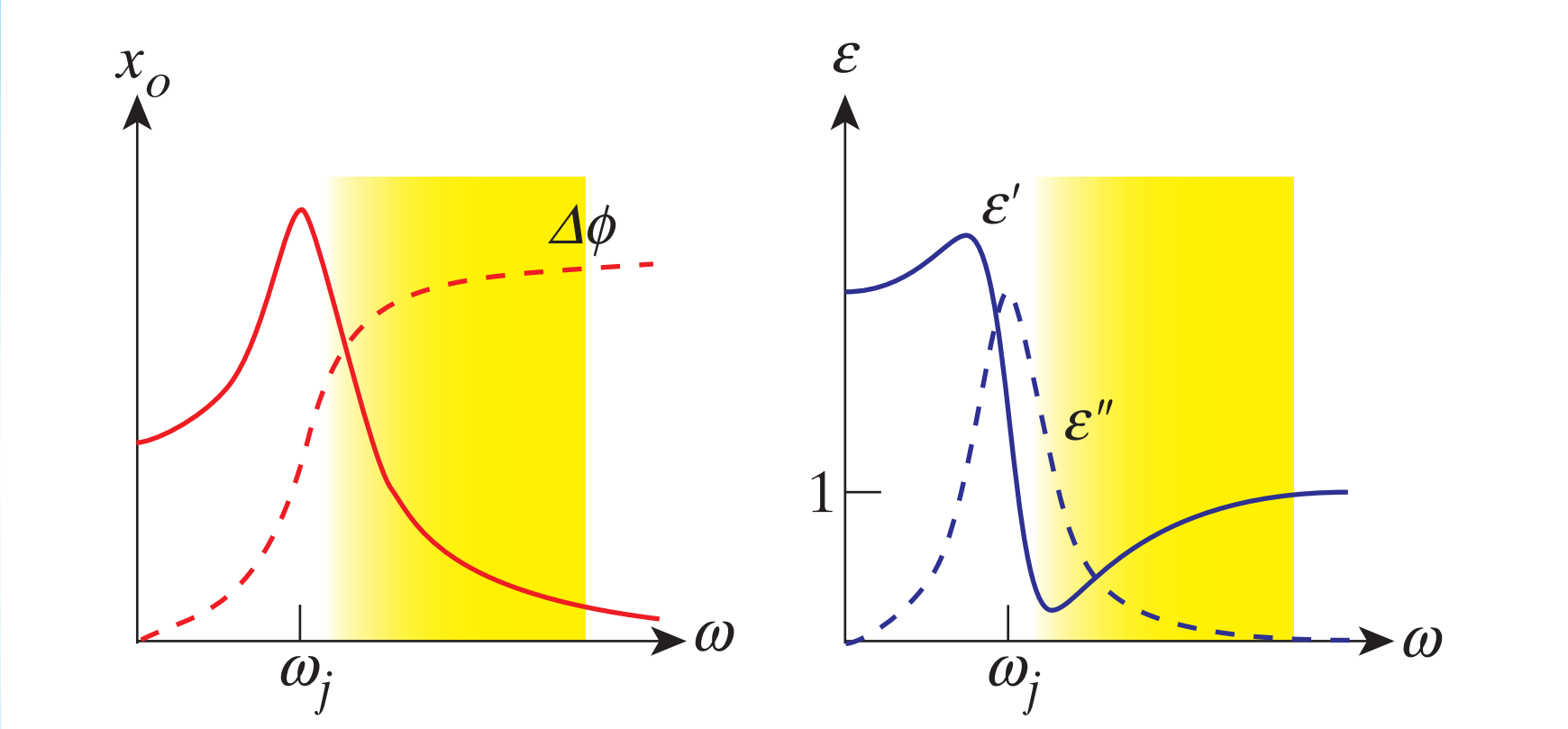
Bound electrons

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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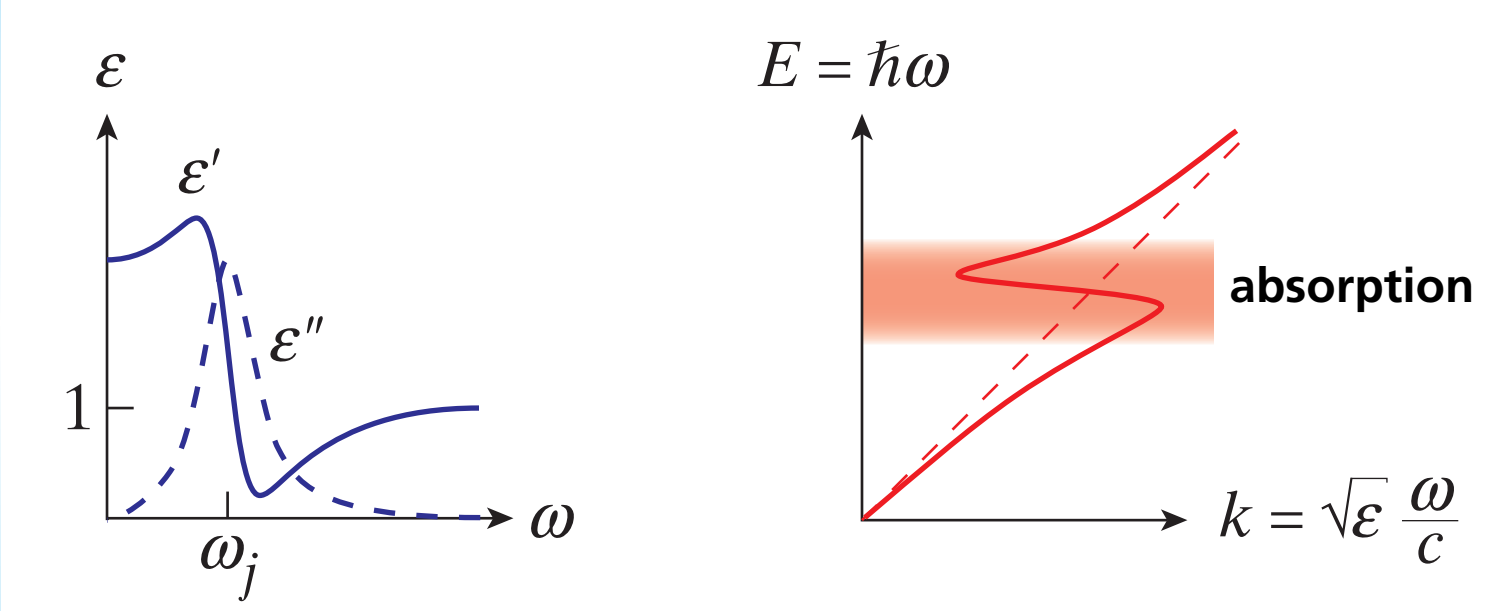

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Low frequency ($\omega \ll 1$) $\Rightarrow$ current generated

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

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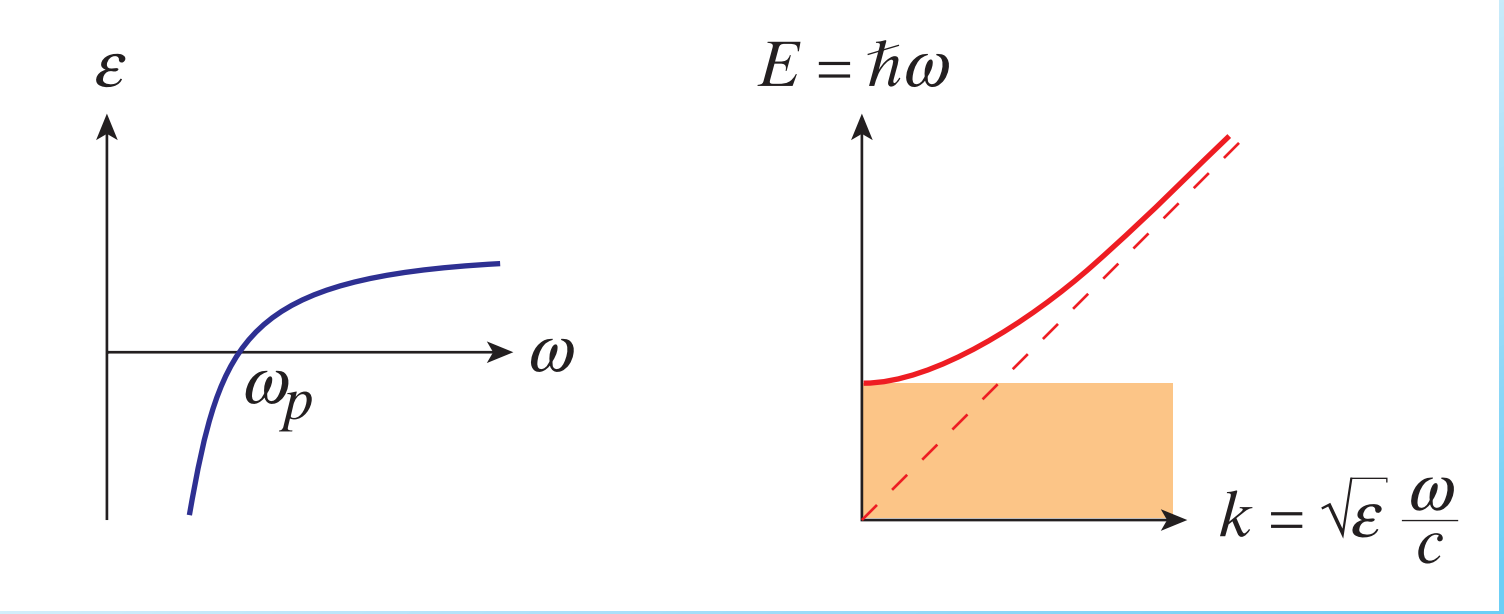
Dielectric function

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

Plasma

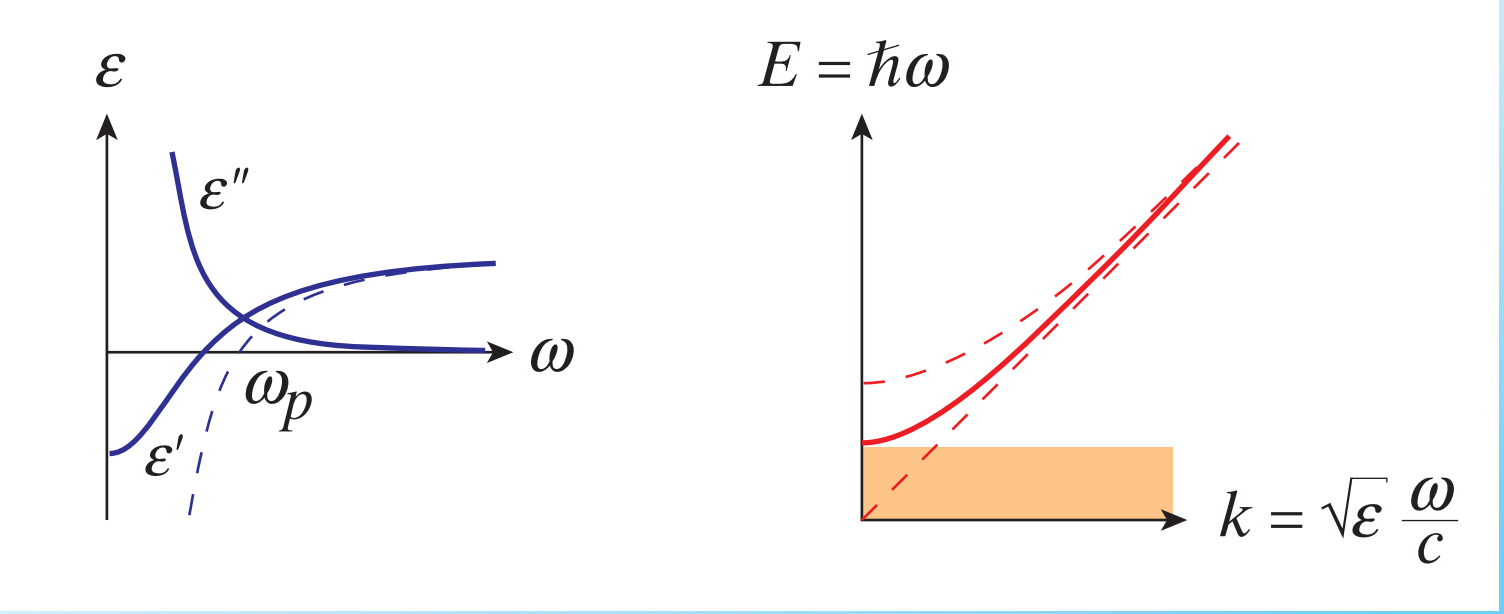
$$\gamma \approx 0 \qquad \Rightarrow \quad \epsilon'' = 0$$

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



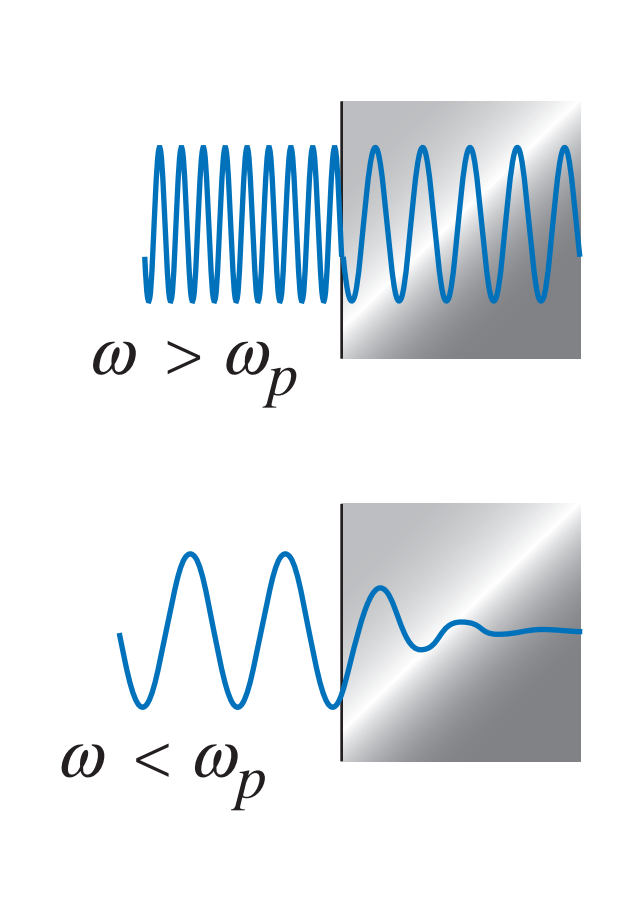
Plasma

Add damping $\gamma \lesssim \omega_p$



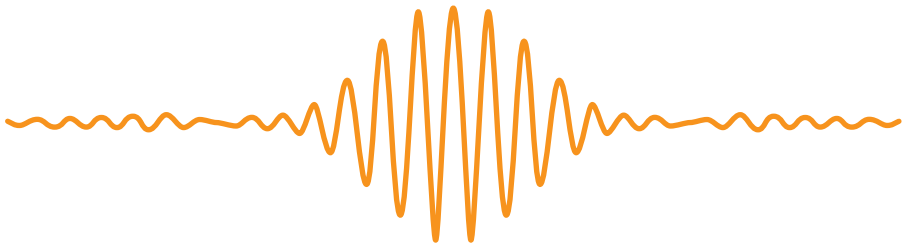
Plasma

Plasma acts like a high-pass filter:



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

Pulse dispersion



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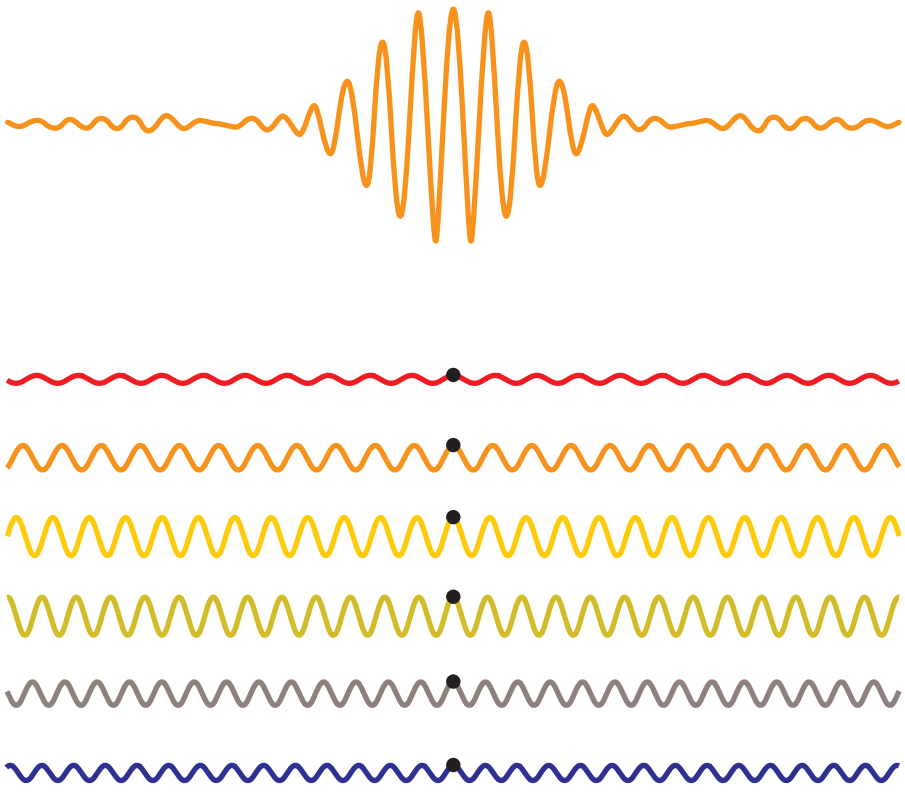

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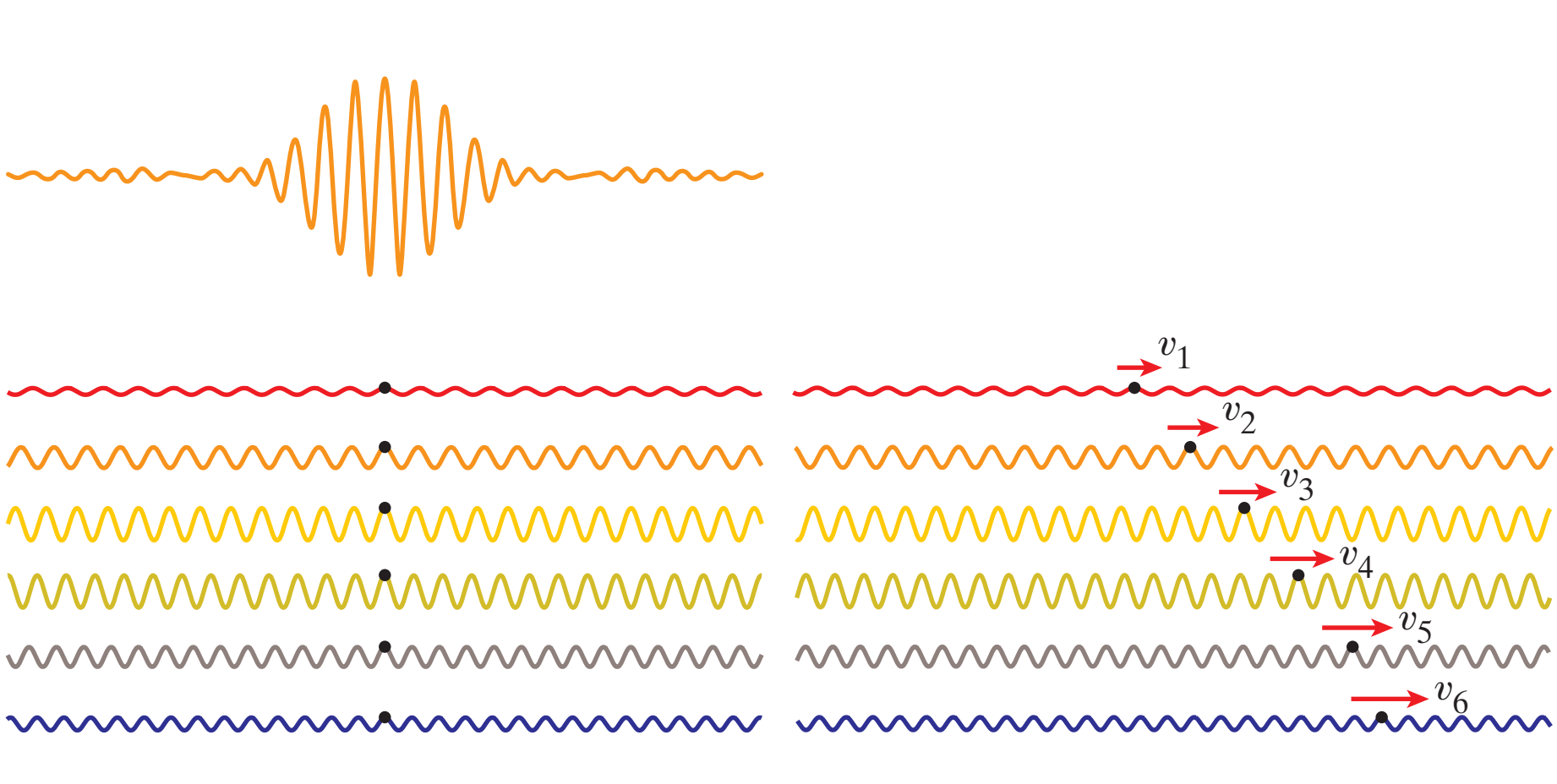

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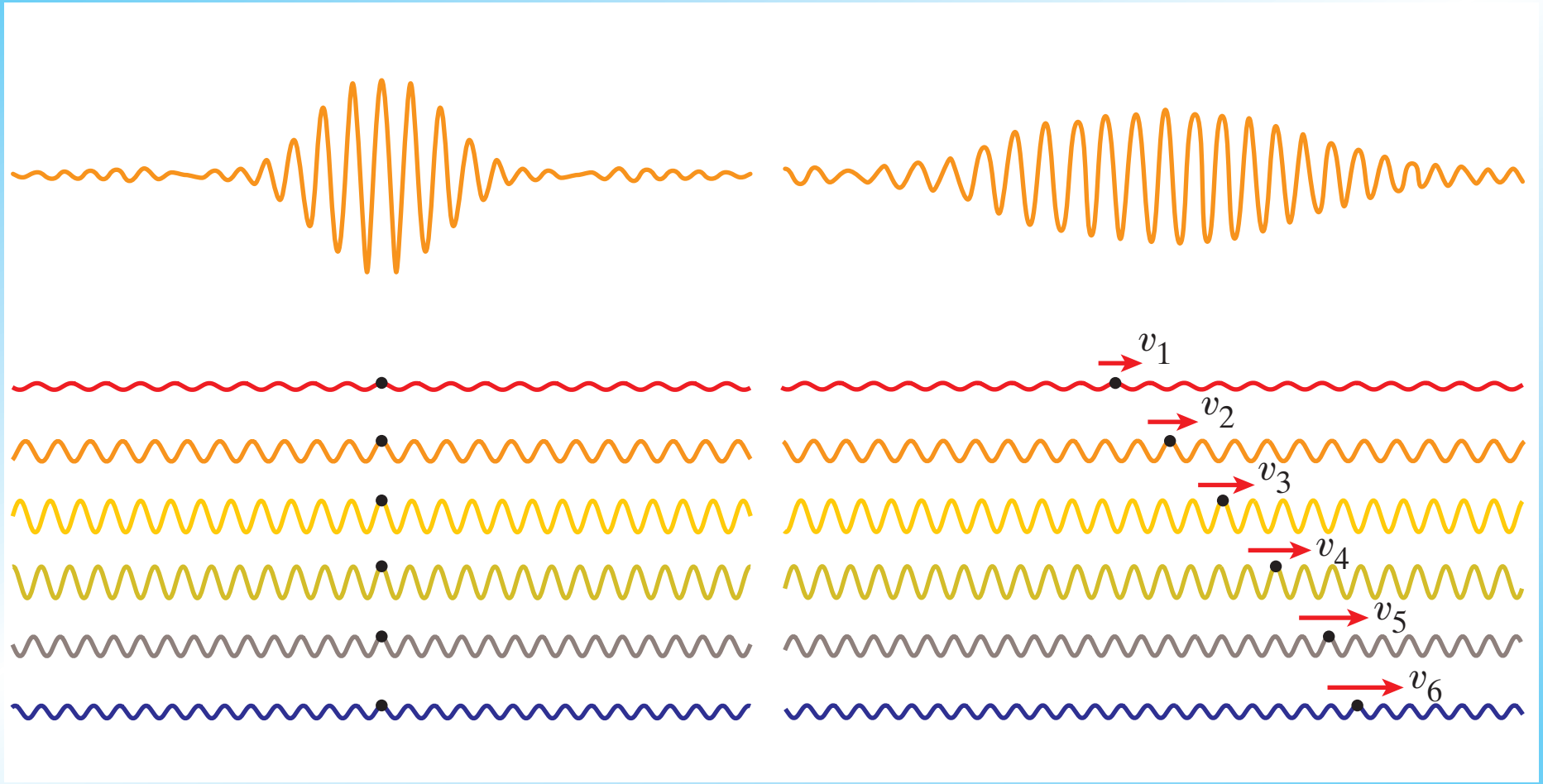

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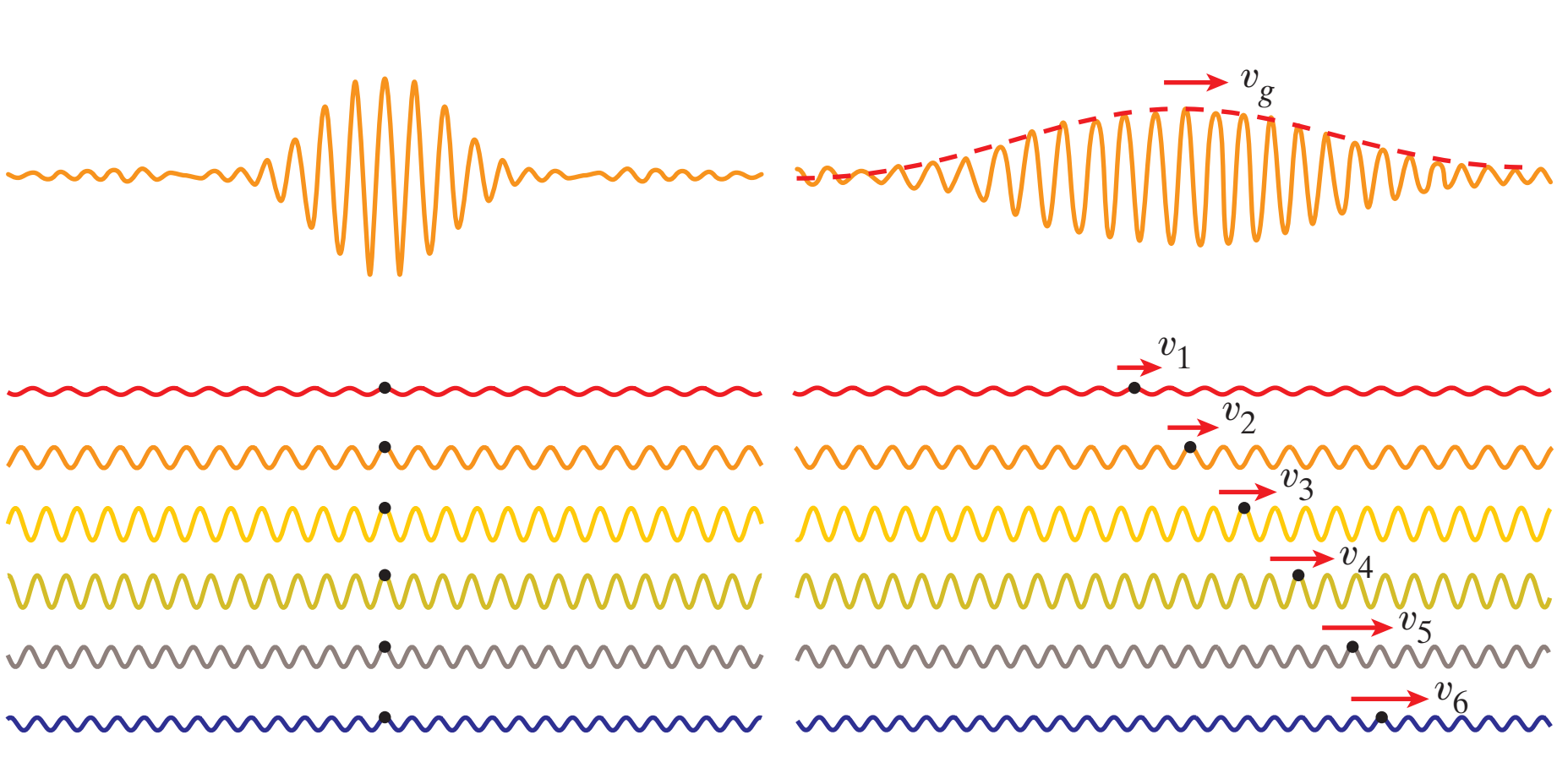
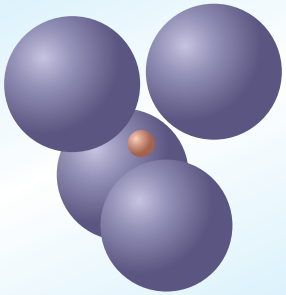


Nonlinear optics

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



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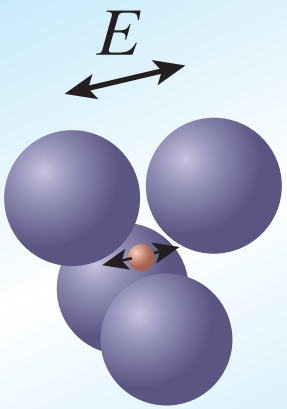

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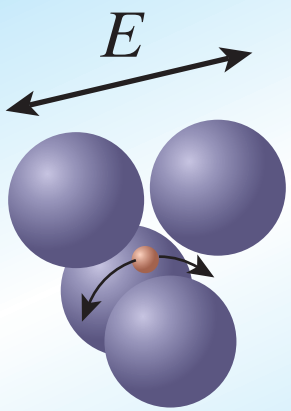

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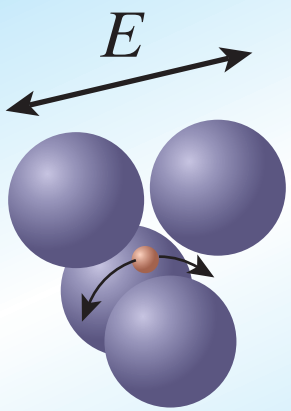
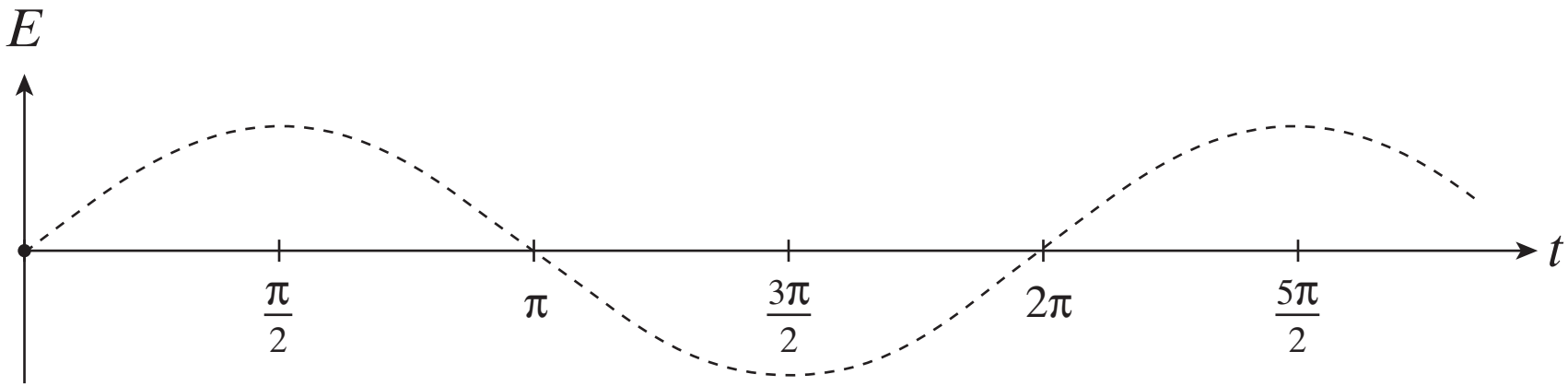


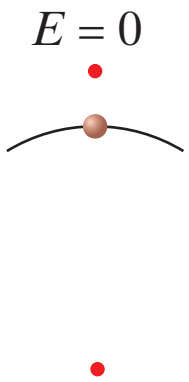
Nonlinear polarization:

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

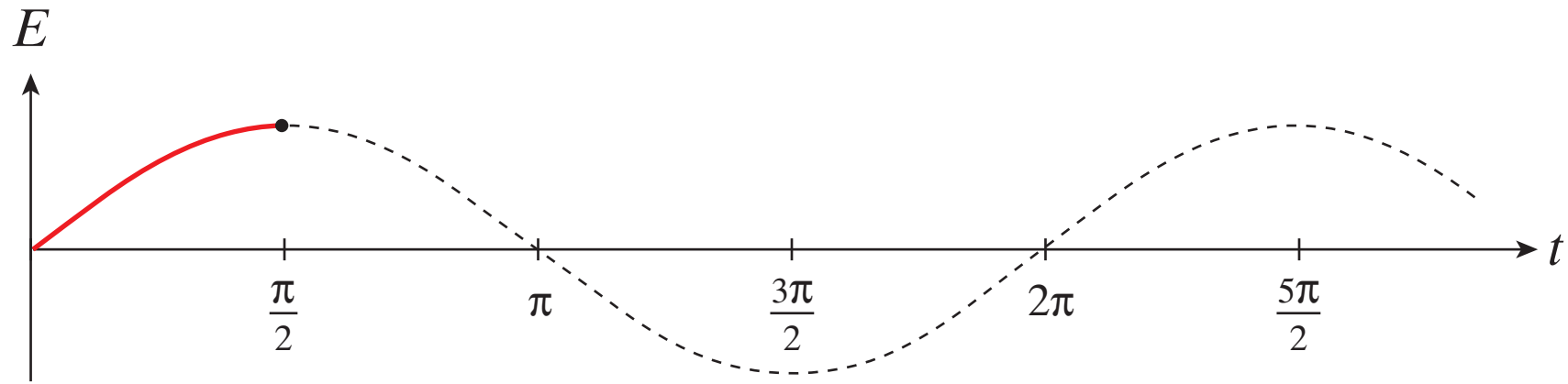
$$P = P^{(1)} + P^{(2)} + P^{(3)} + \ldots$$

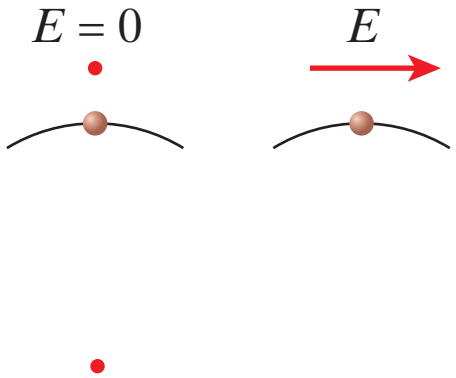
Nonlinear optics



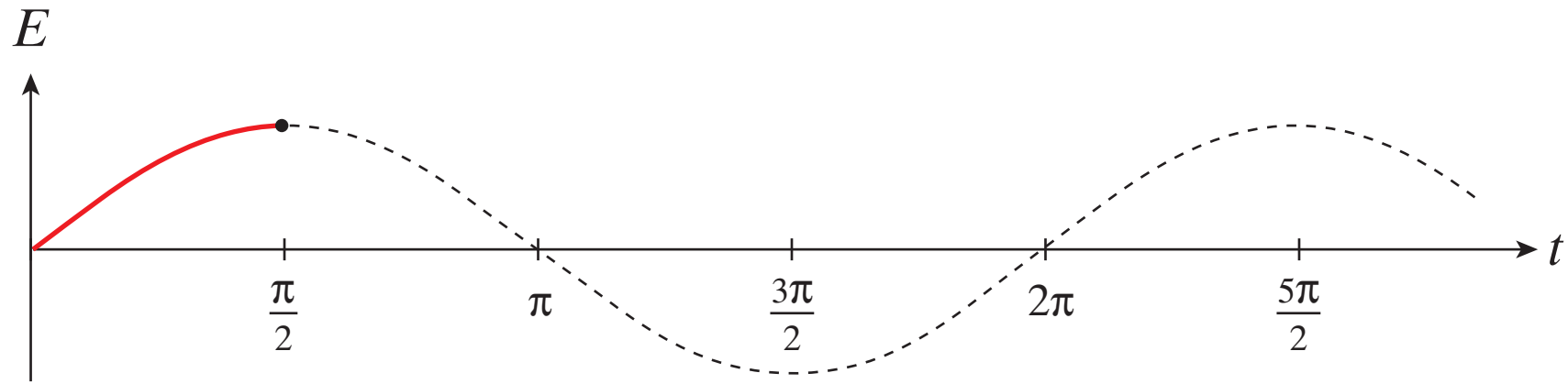


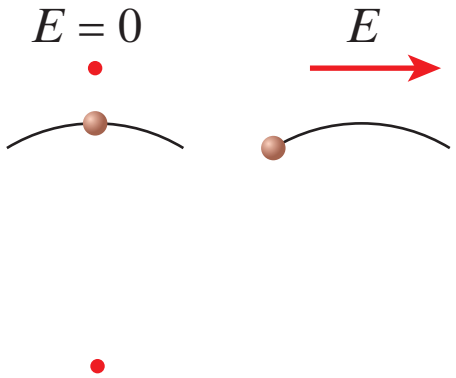
Nonlinear optics



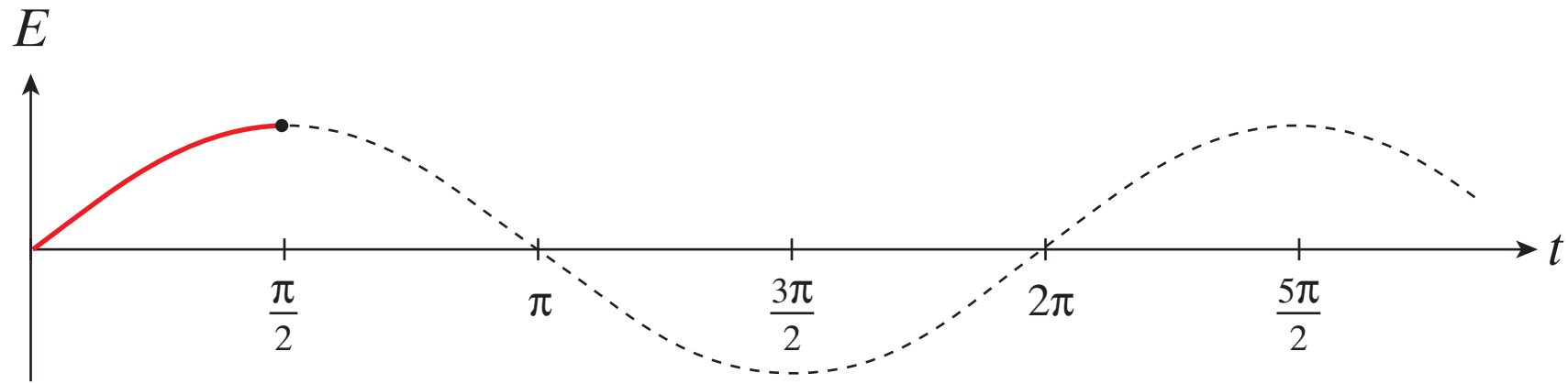


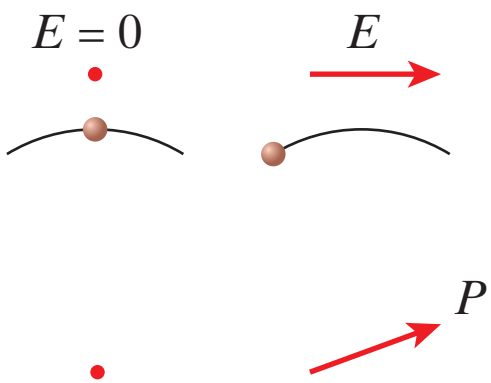
Nonlinear optics



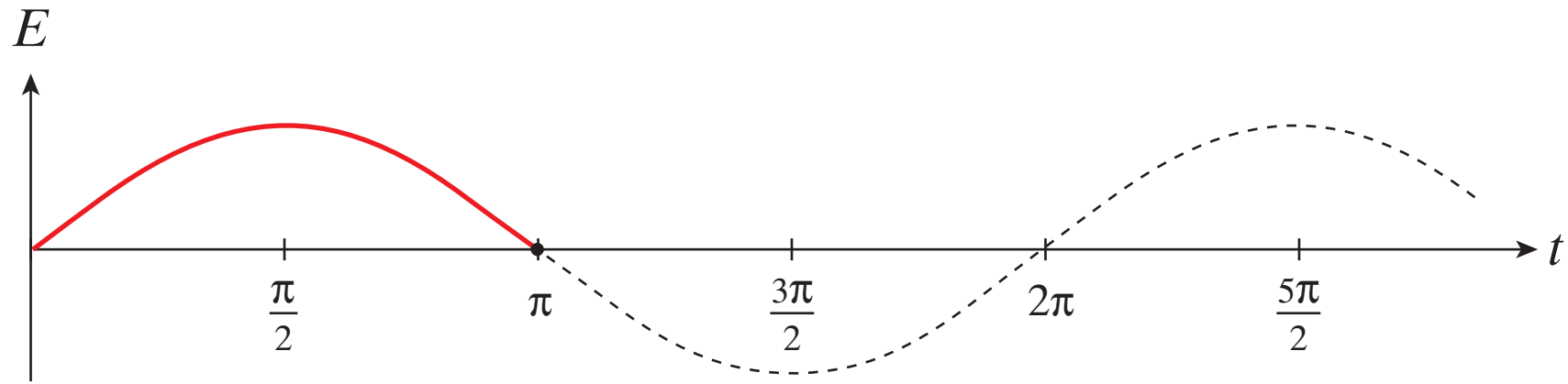


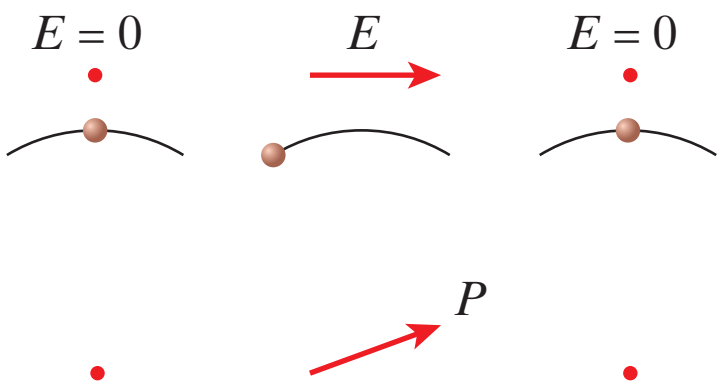
Nonlinear optics



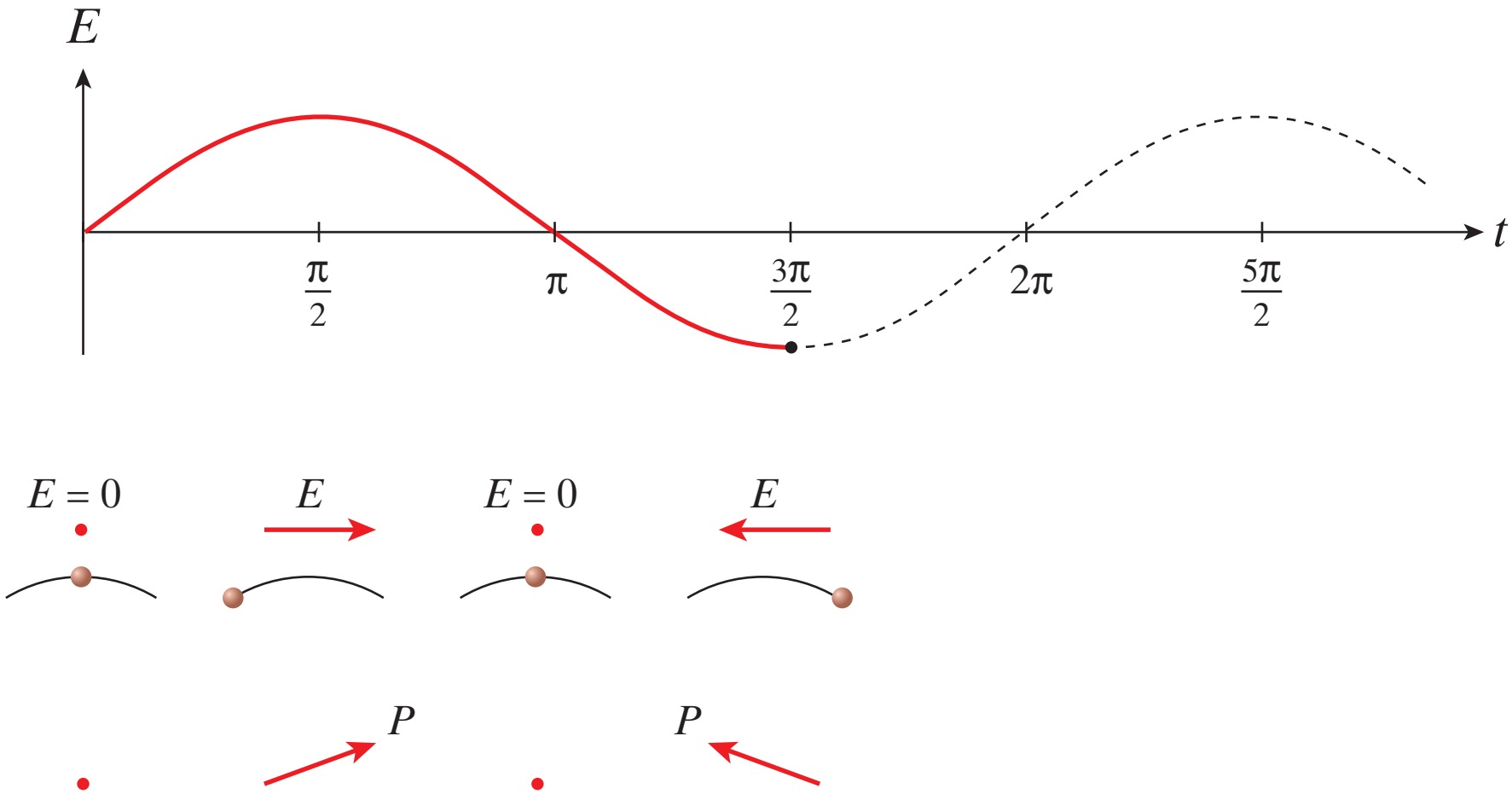


Nonlinear optics

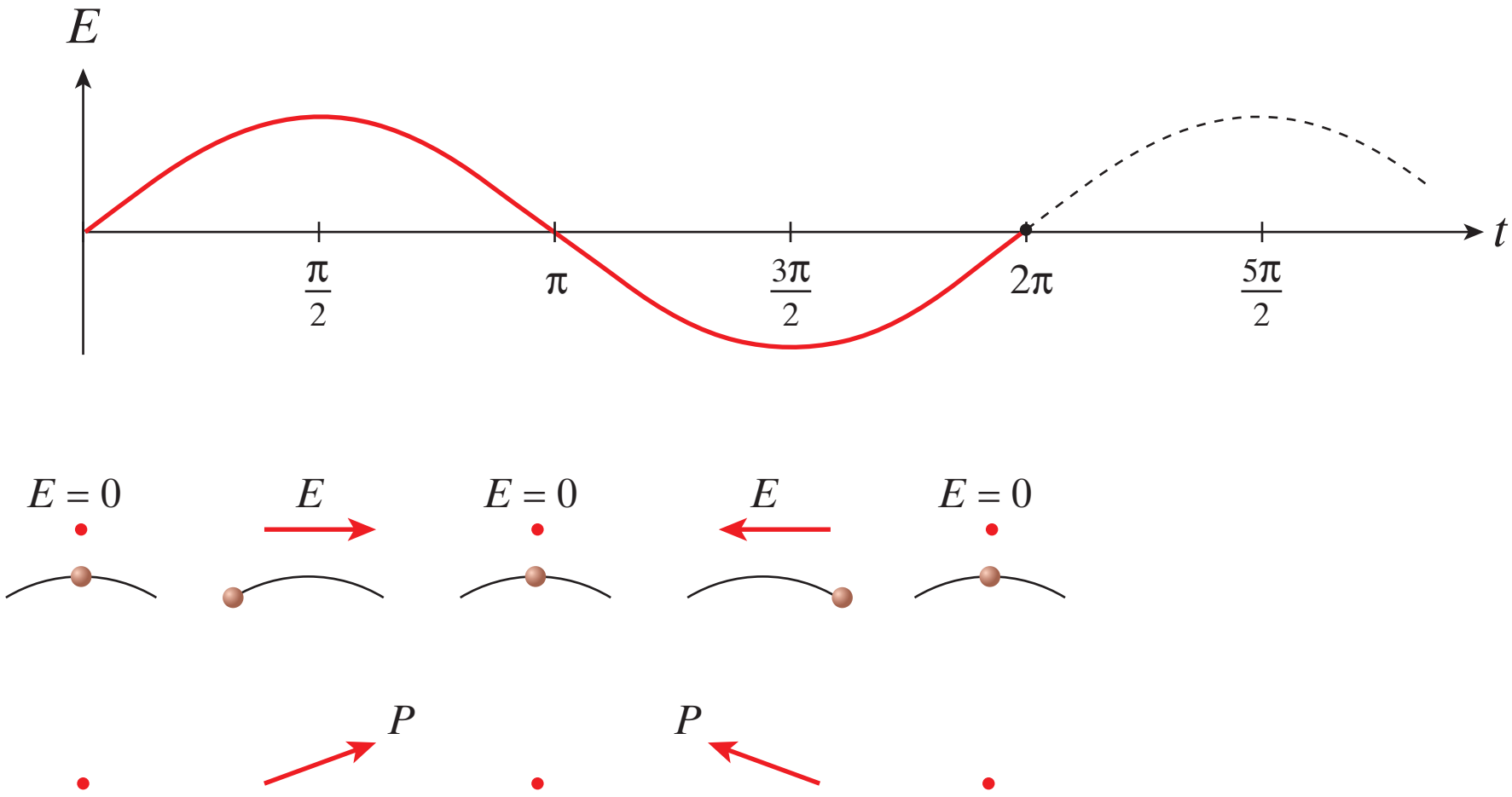




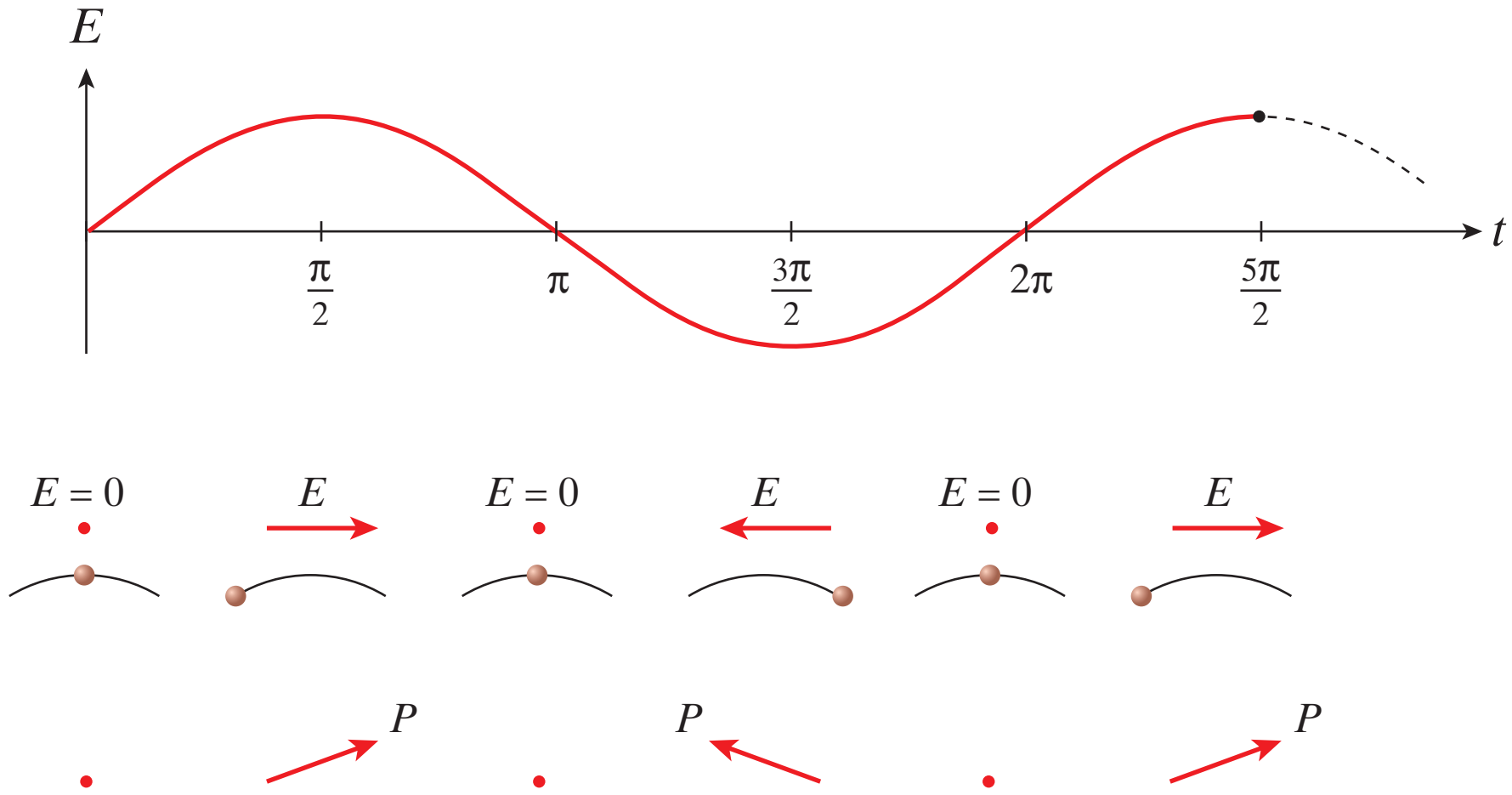
Nonlinear optics



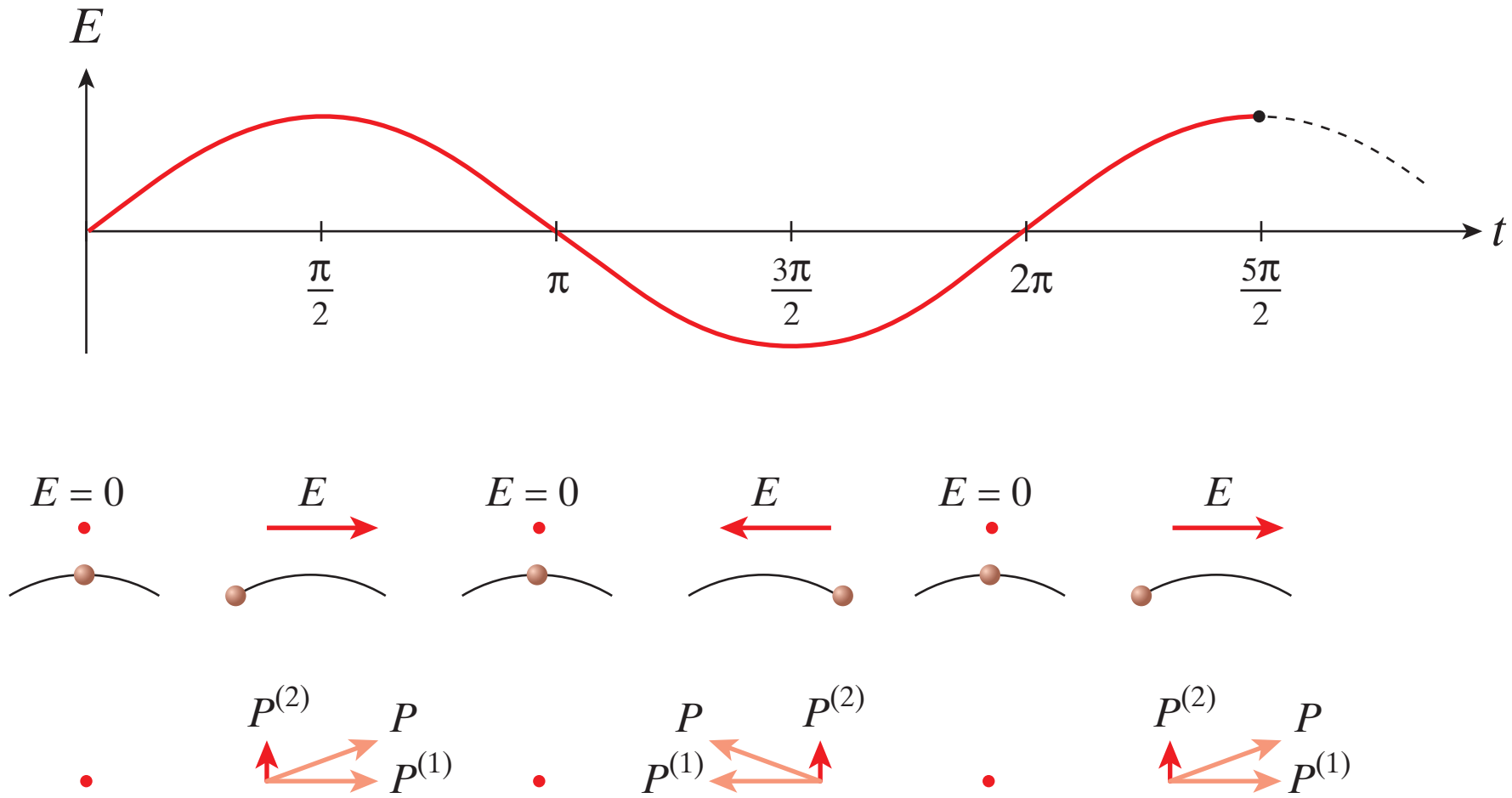
Nonlinear optics



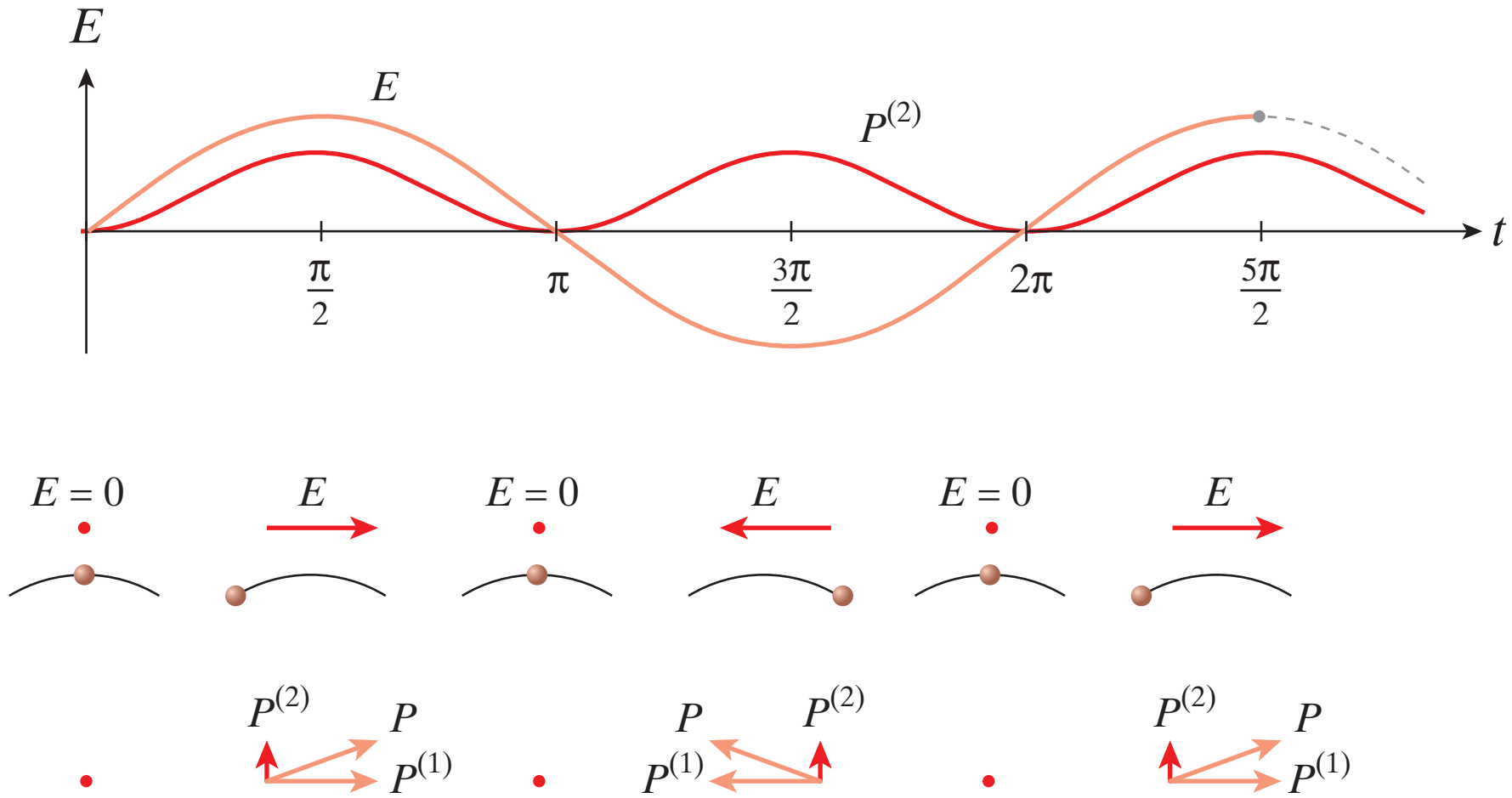
Nonlinear optics



Nonlinear optics



Nonlinear optics



In medium with inversion symmetry

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

Nonlinear optics

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

and so

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

Nonlinear optics

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and so

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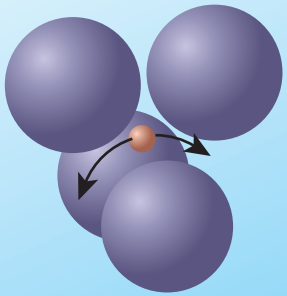
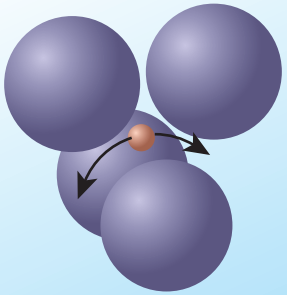
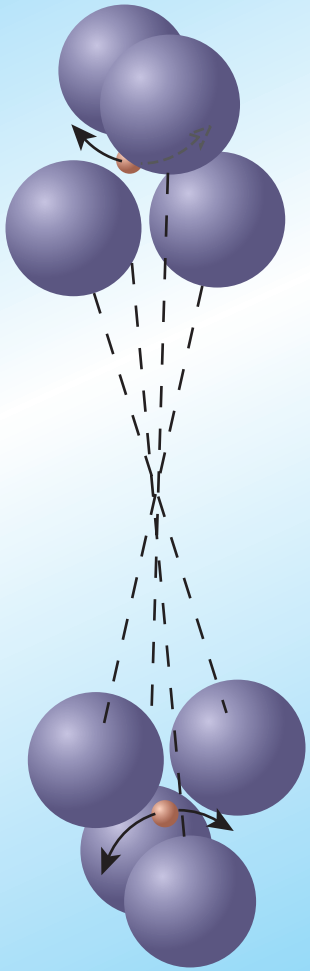
… but …



Nonlinear optics

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

Nonlinear optics



Nonlinear optics

Nonlinear polarization:

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

Nonlinear optics

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Third order polarization

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

Nonlinear optics

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Nonlinear optics

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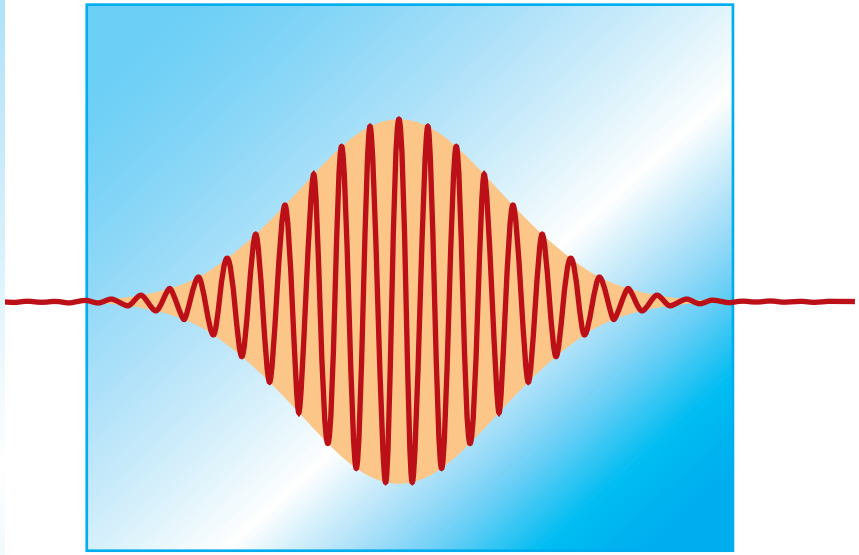
and so $$P = P^{(1)} + P^{(3)} = (\chi^{(1)} + \chi^{(3)}I)E \equiv \chi_{eff}E$$

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

Nonlinear optics

Intensity dependent index of refraction:

$$n = n_o + n_2 I$$



Nonlinear optics

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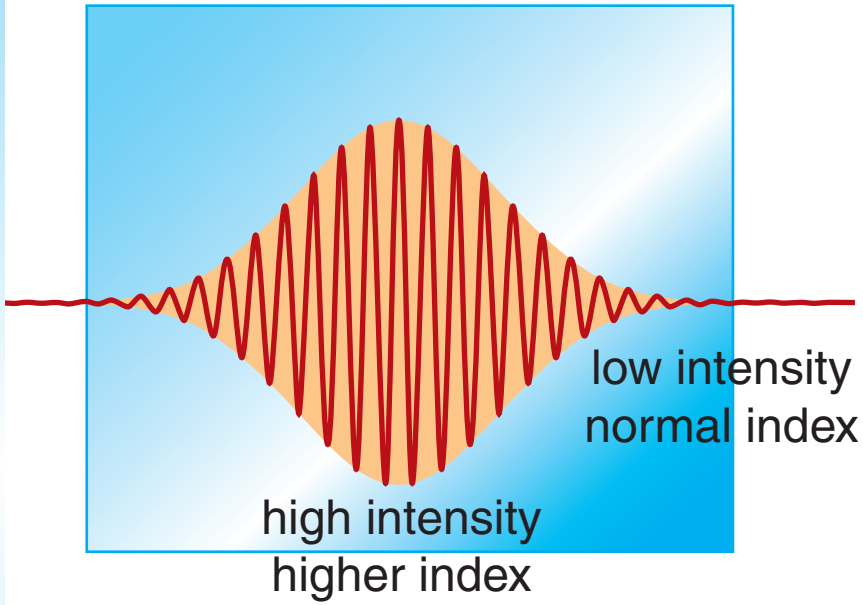

Nonlinear optics

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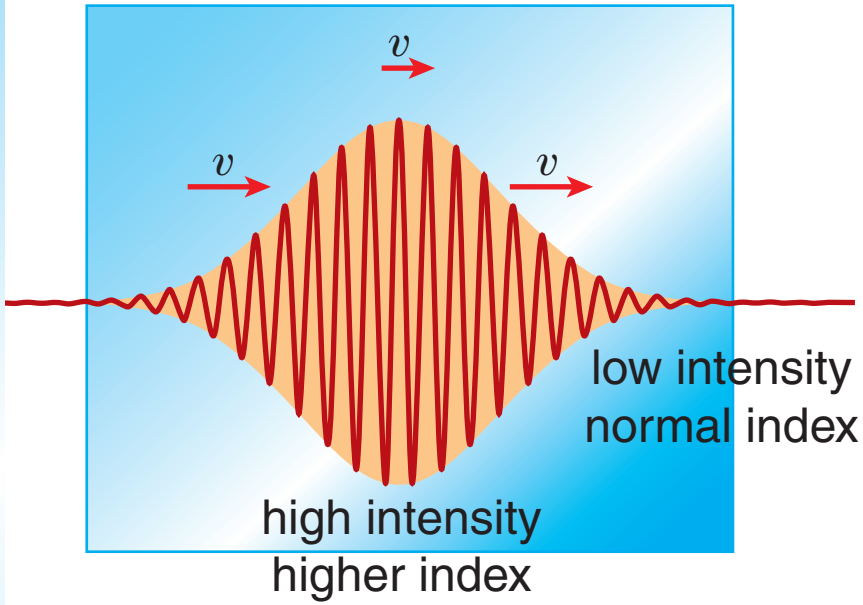

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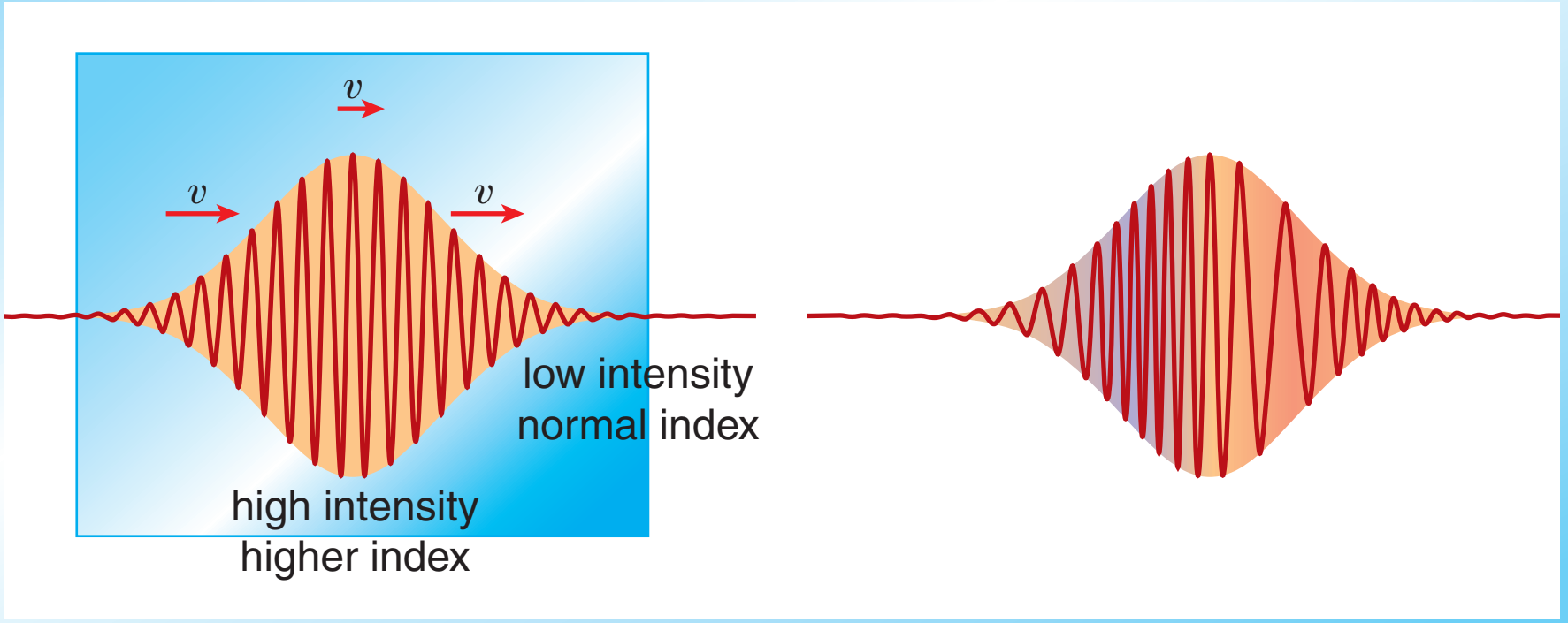

Nonlinear optics

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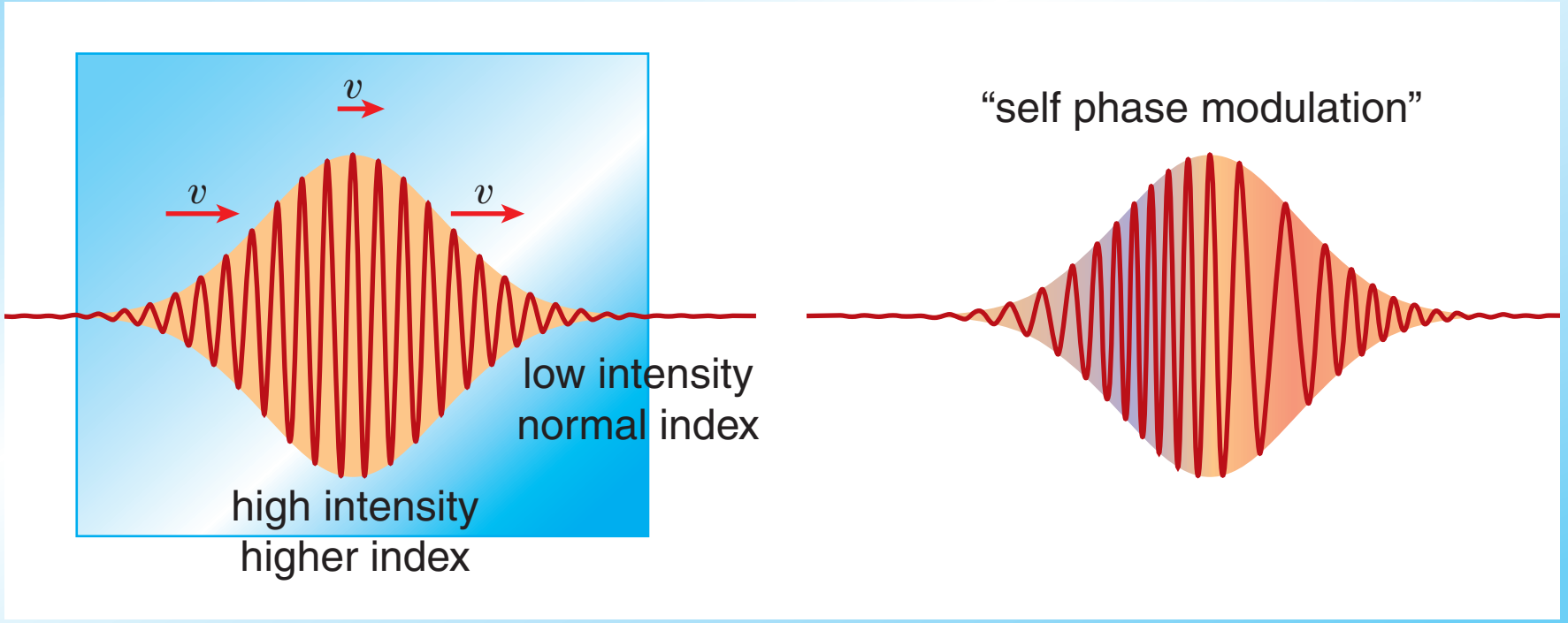

Phase:

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

Nonlinear optics

Phase:

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

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Frequency change:

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Nonlinear optics

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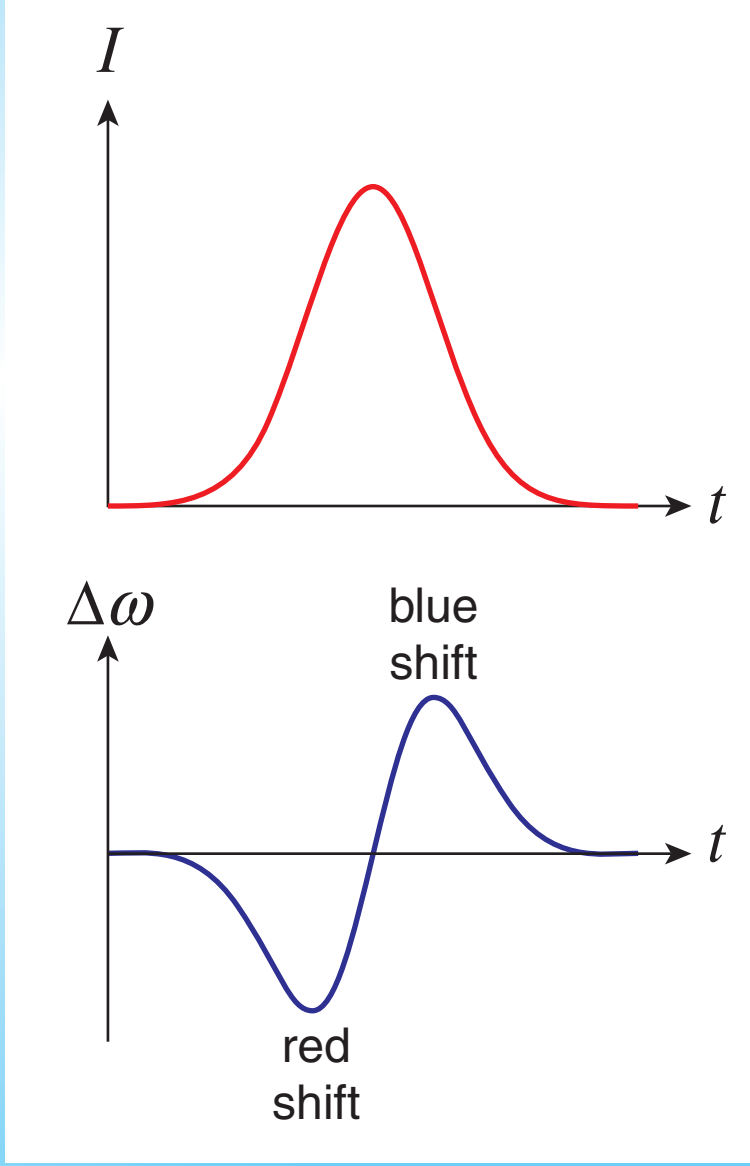
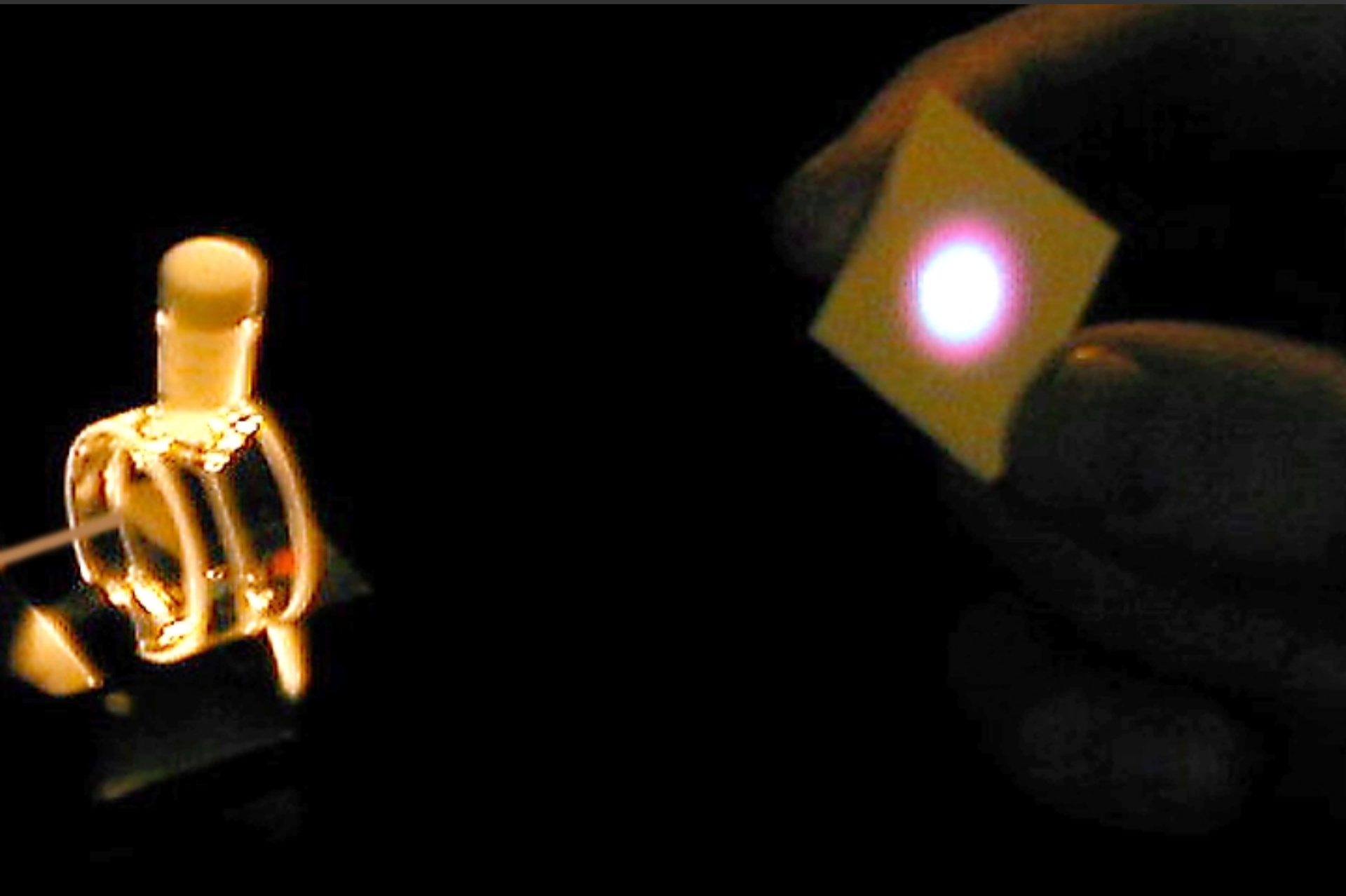
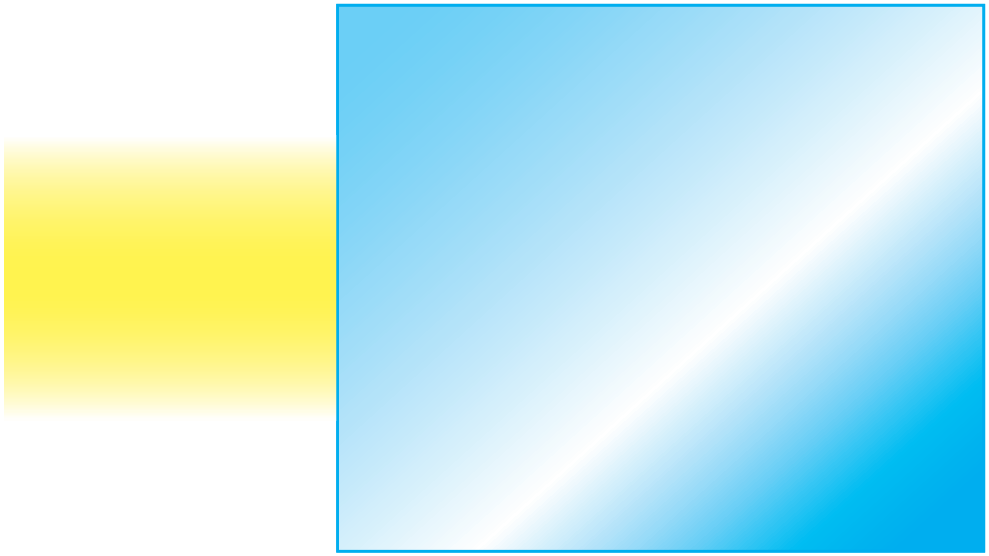


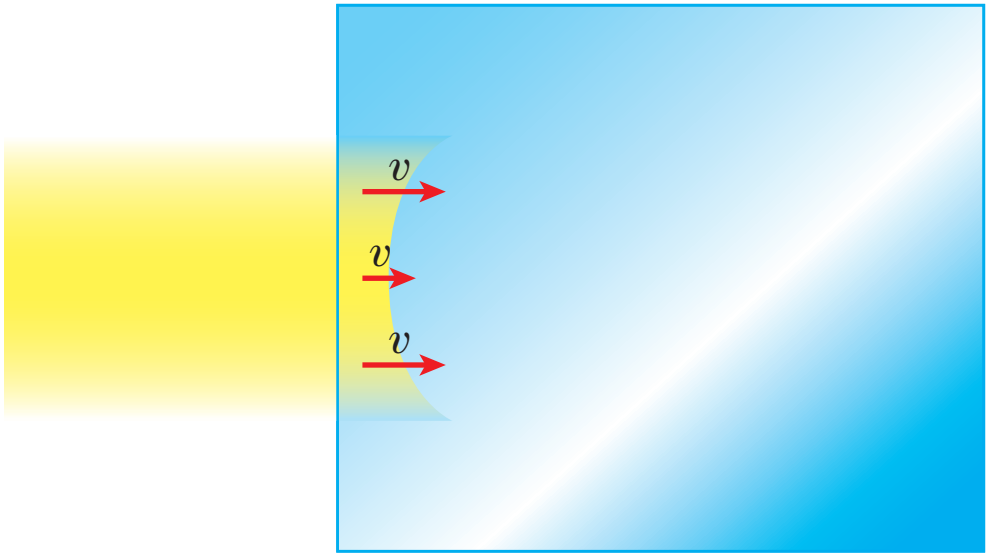
Nonlinear optics



Spatial intensity profile…

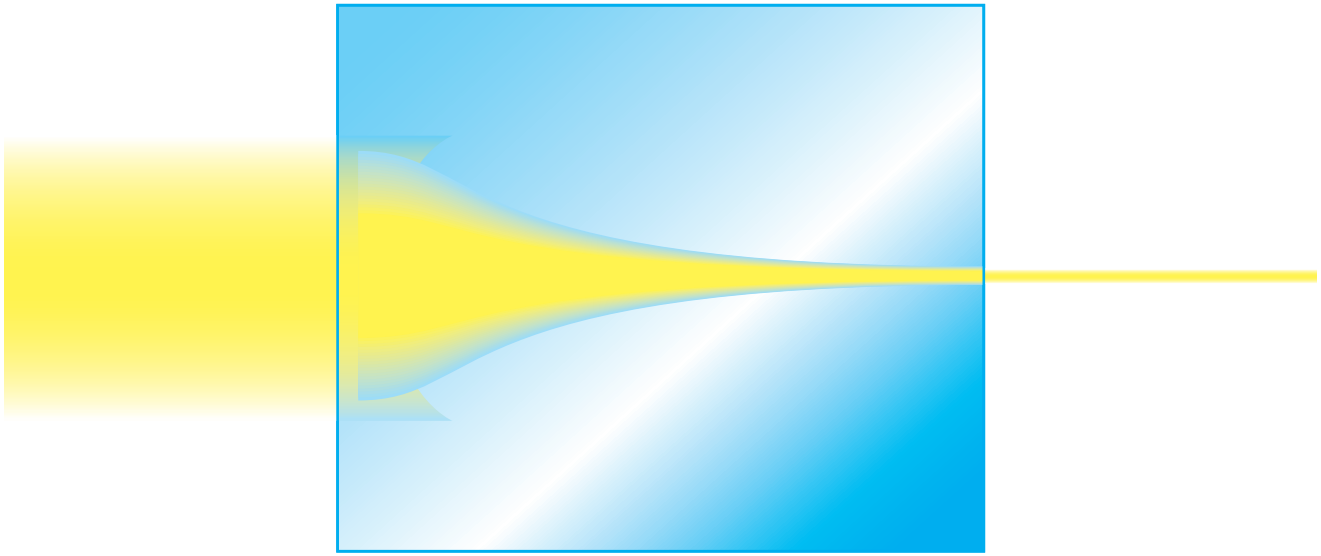


Spatial intensity profile…



Nonlinear optics

…causes self-focusing

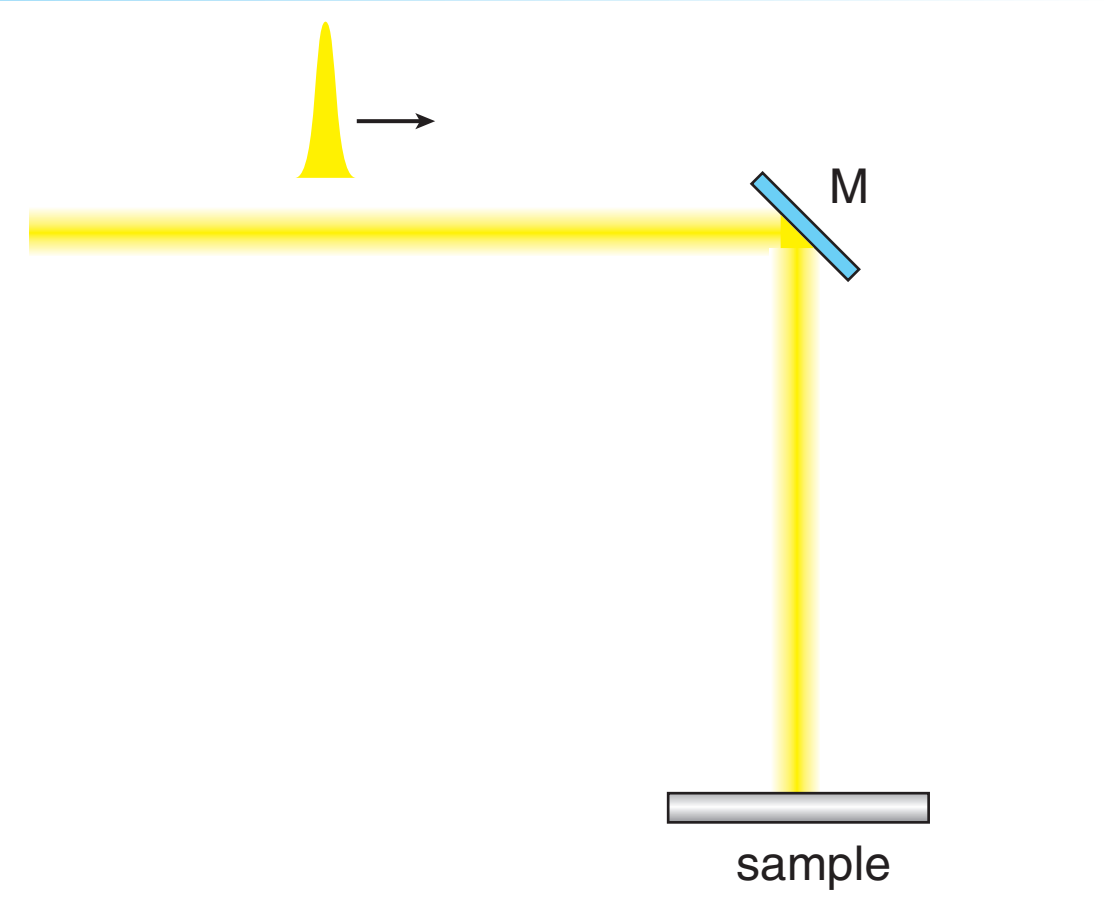


Outline

- linear and nonlinear propagation
- femtosecond measurements
- examples

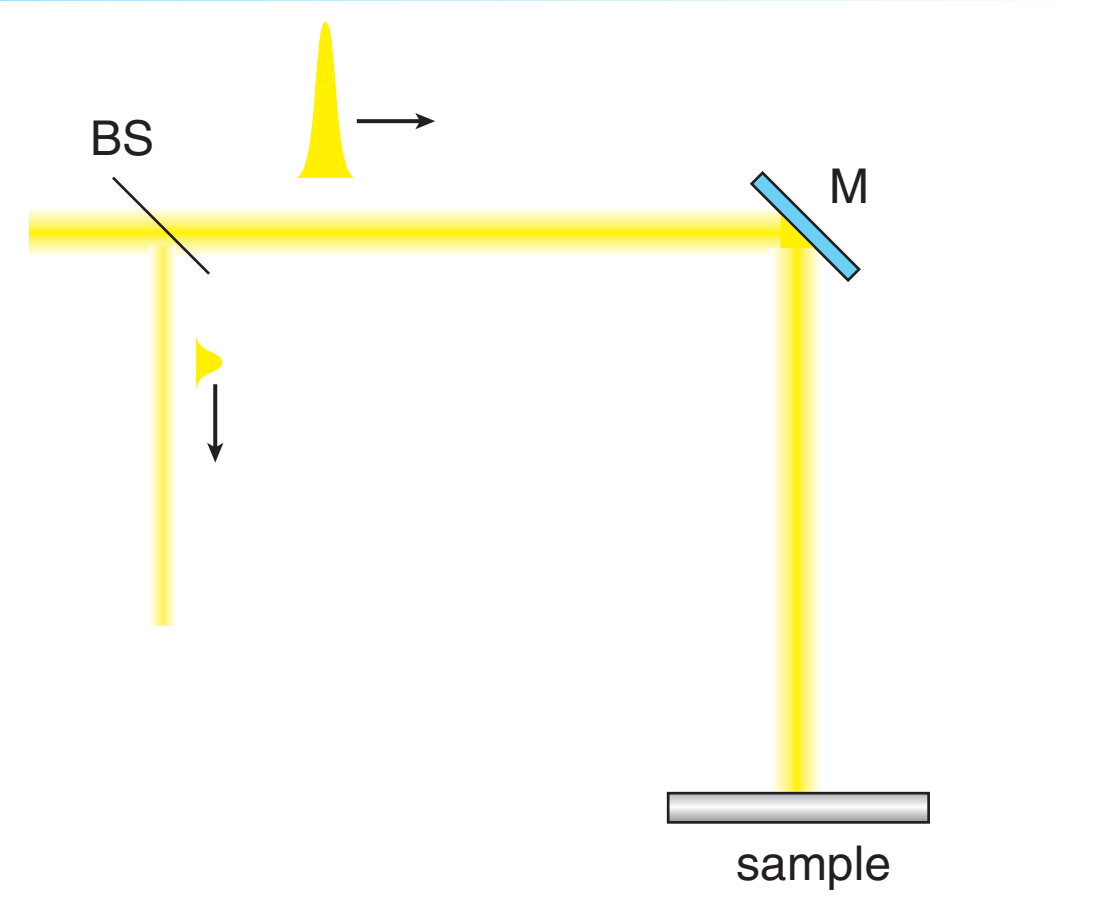
Introduction

How to measure on the femtosecond time scale?



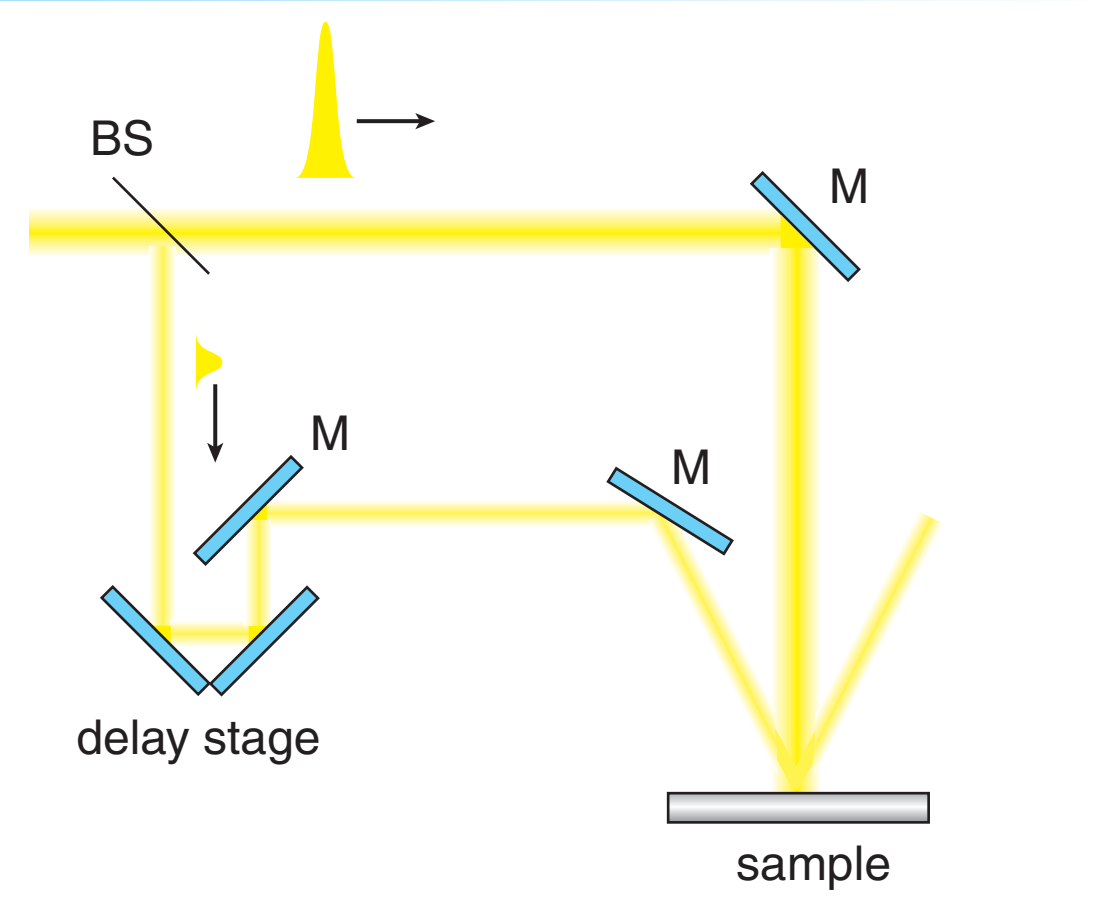
Introduction

Use pump-probe technique



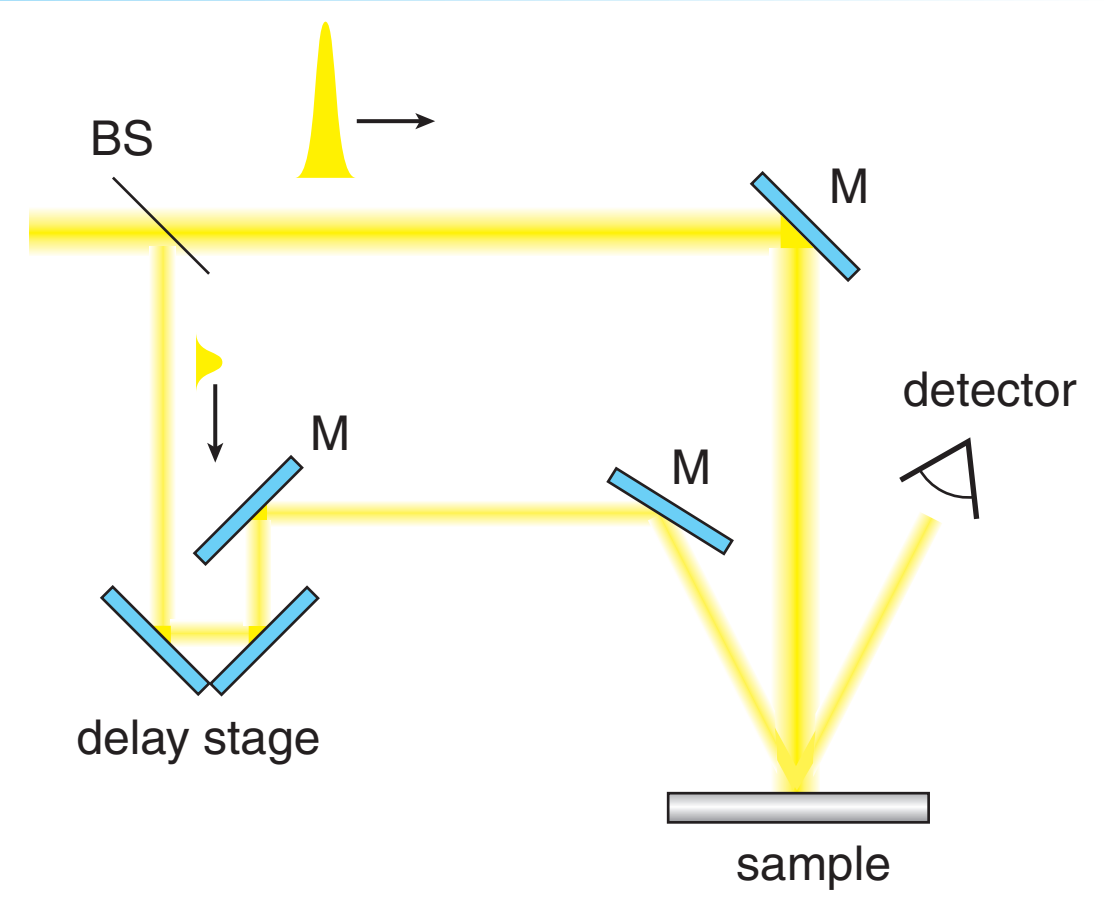
Introduction

Use pump-probe technique



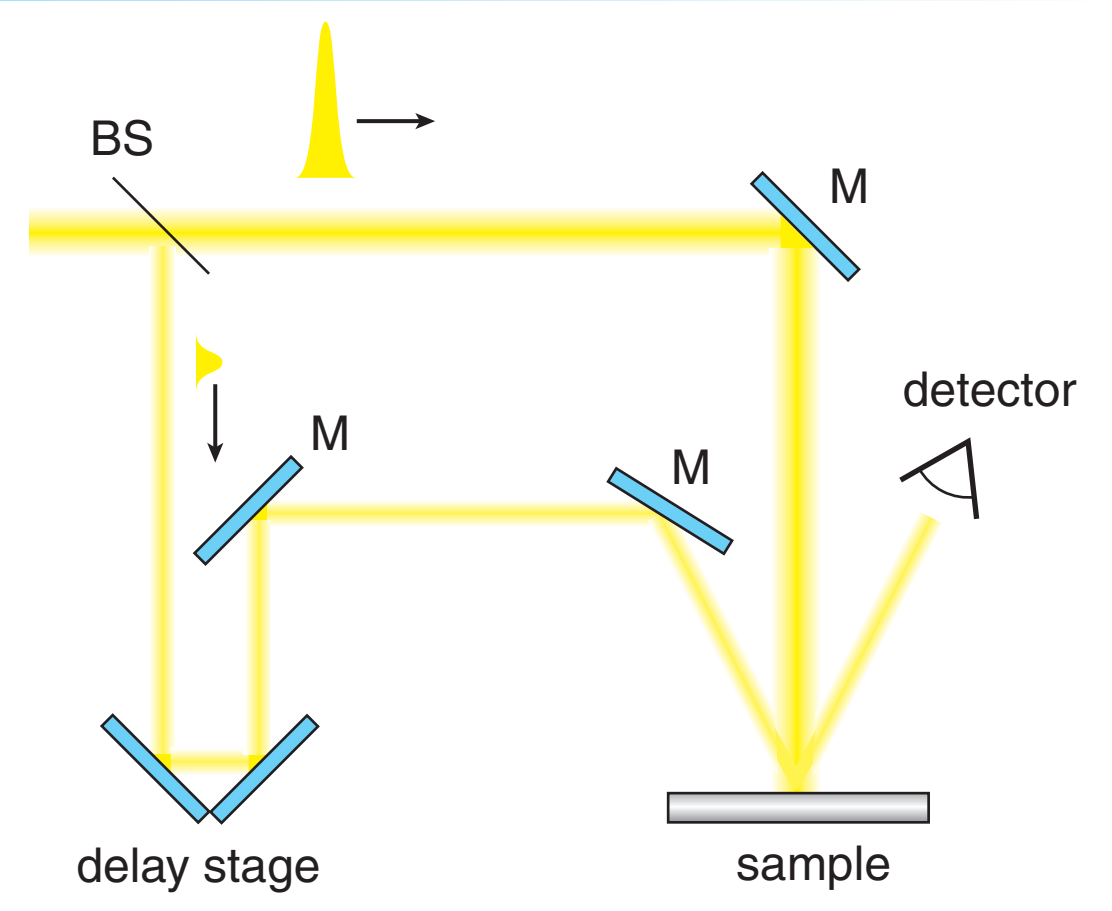
Introduction

Use pump-probe technique



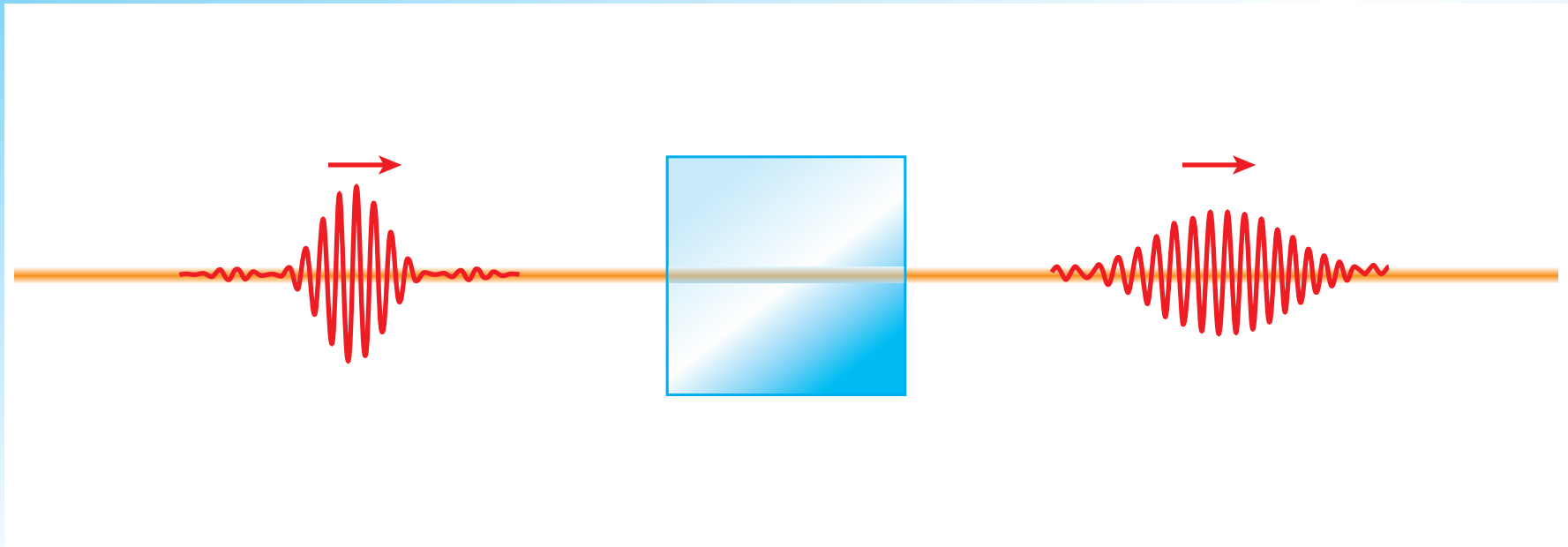
Introduction

Vary delay to get time resolution



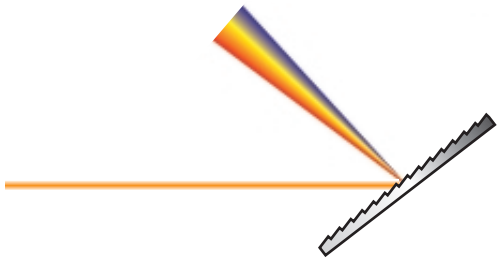
Dispersion compensation

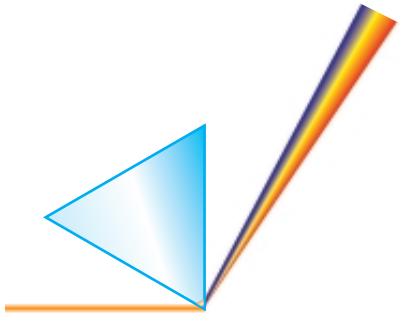
Dispersion stretches the pulse



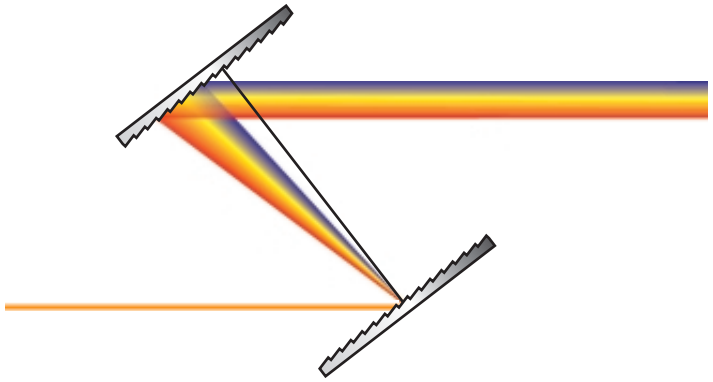
Compensate by rearranging spectral components!

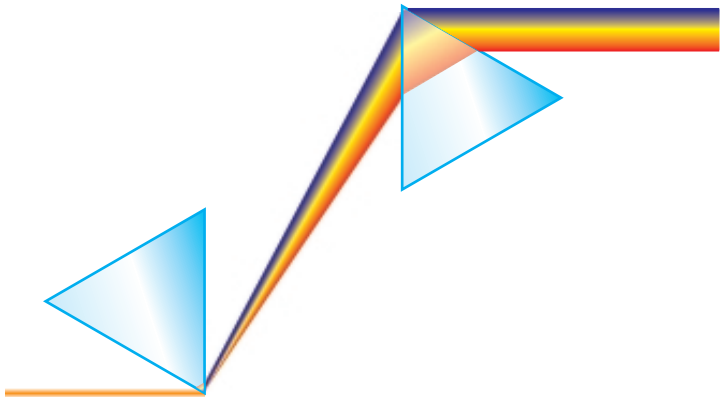
Dispersion compensation



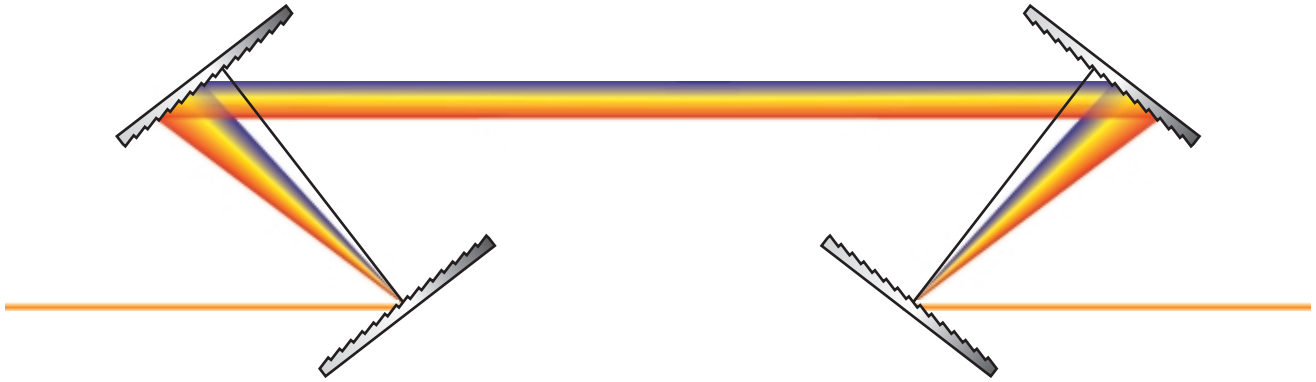


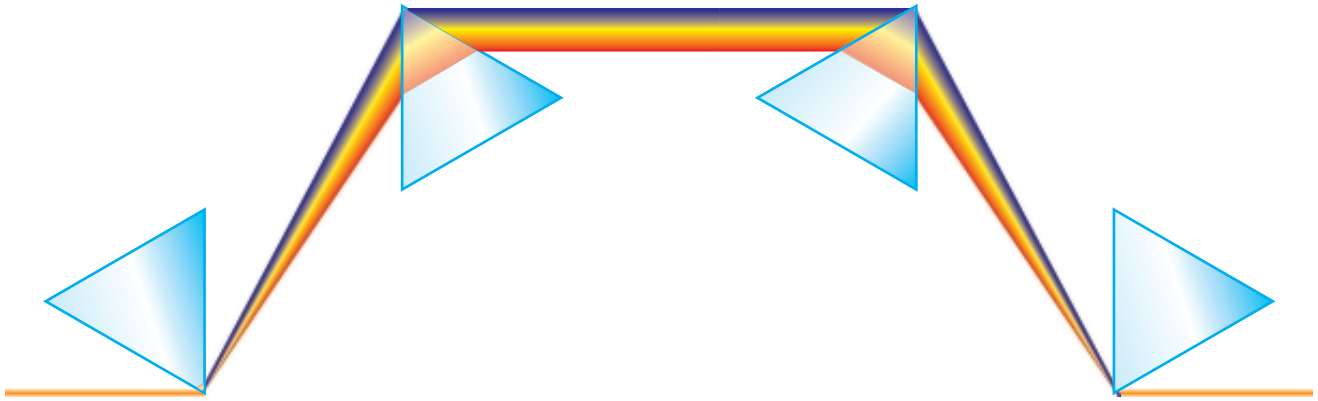
Dispersion compensation





Dispersion compensation



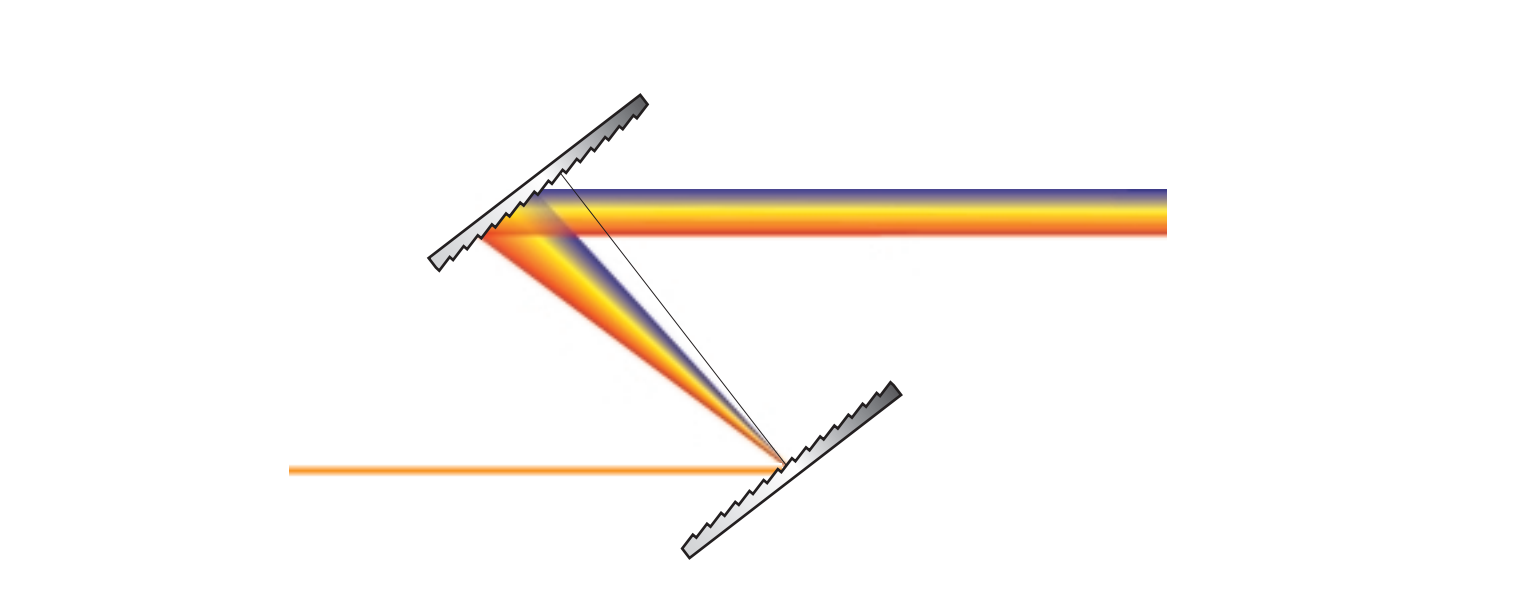


Dispersion compensation

How do these arrangements work?

Dispersion compensation

Does path length difference compensate?



Dispersion compensation

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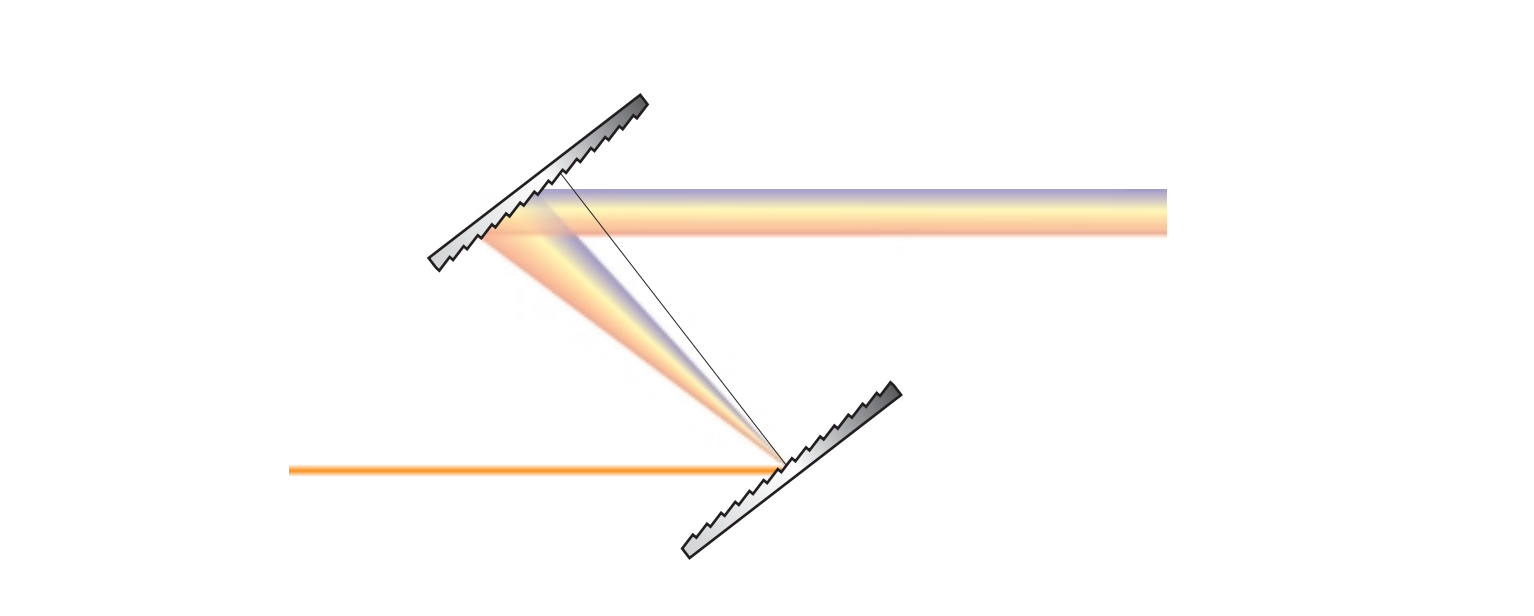

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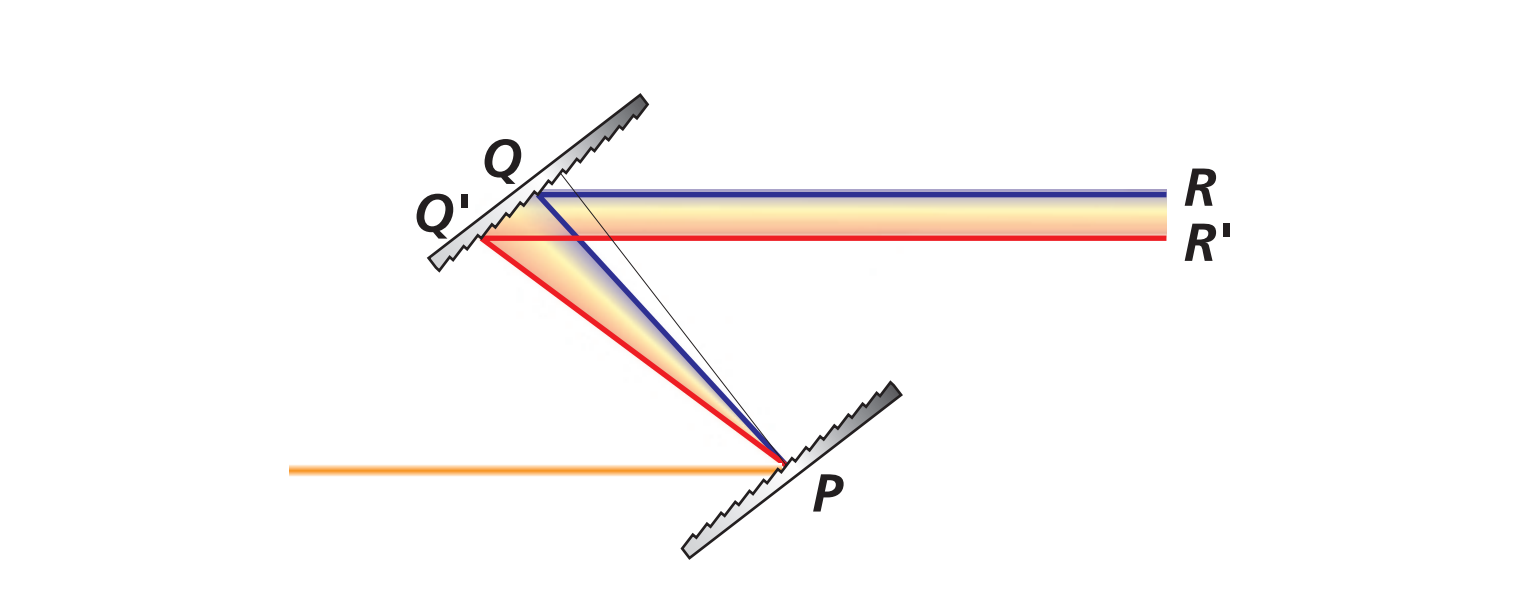
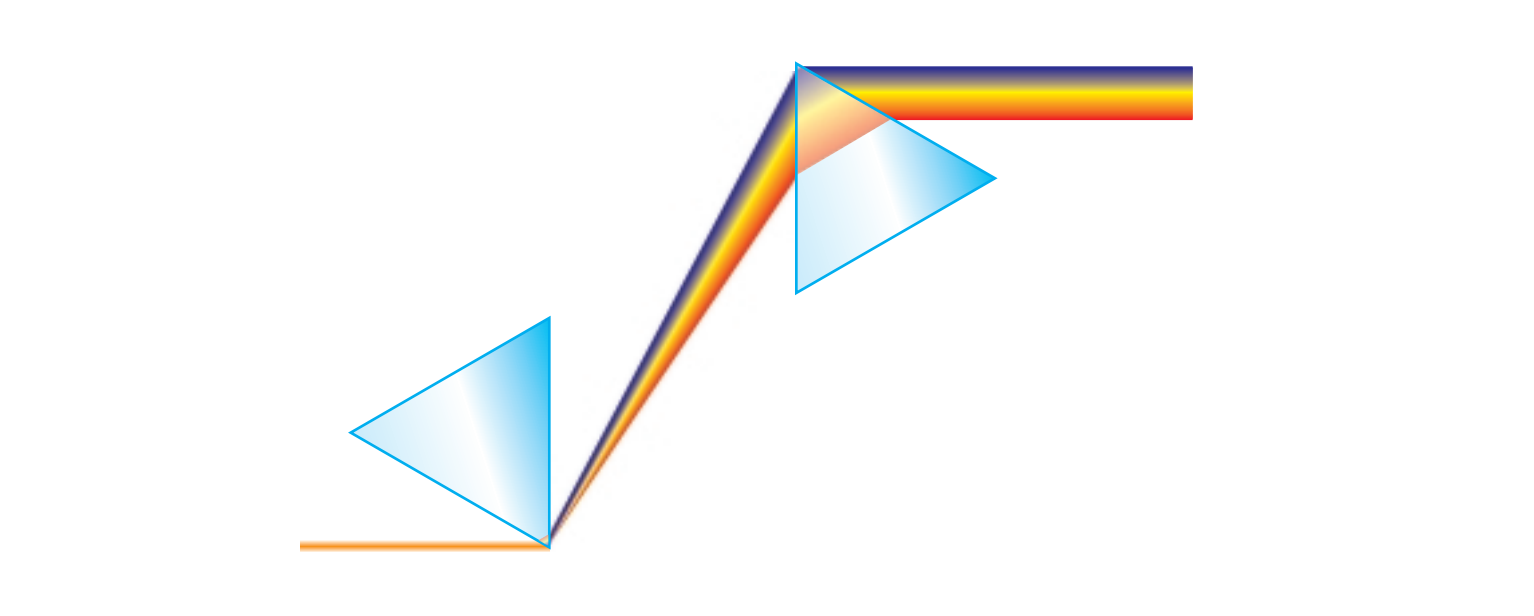


Grating gives low frequency longer path length…

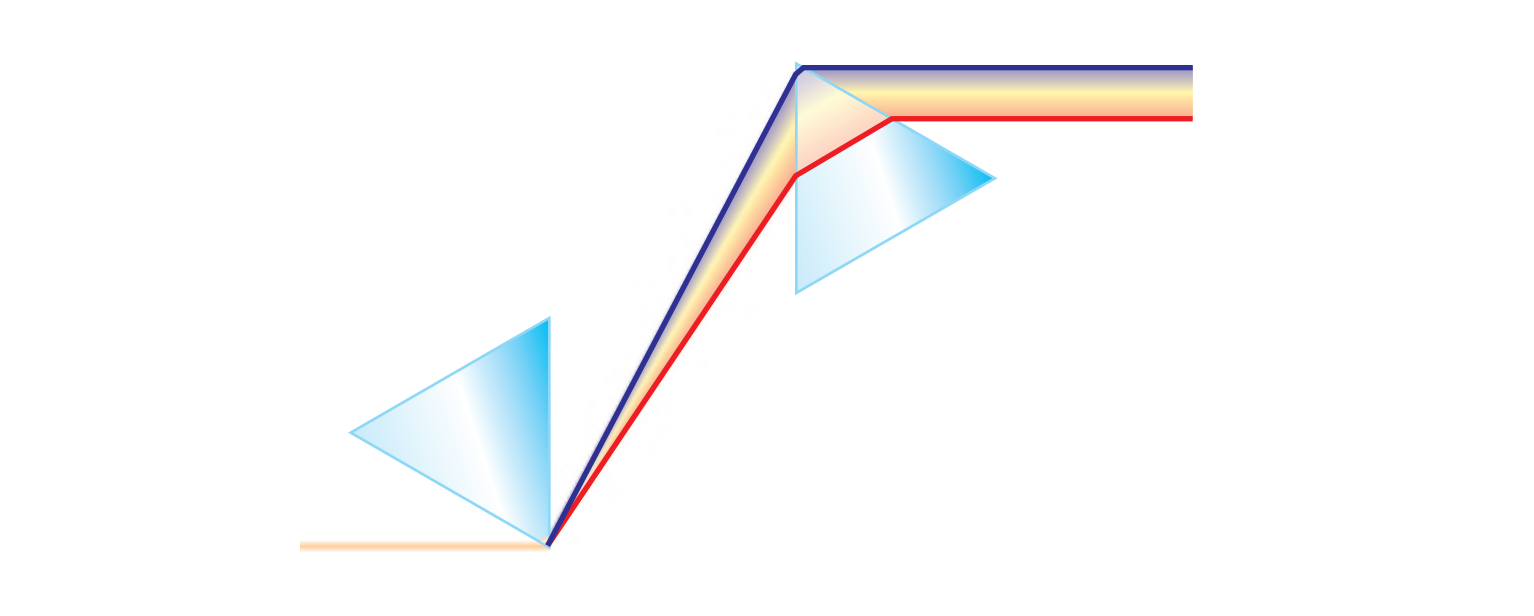
Dispersion compensation

Does path length difference compensate?



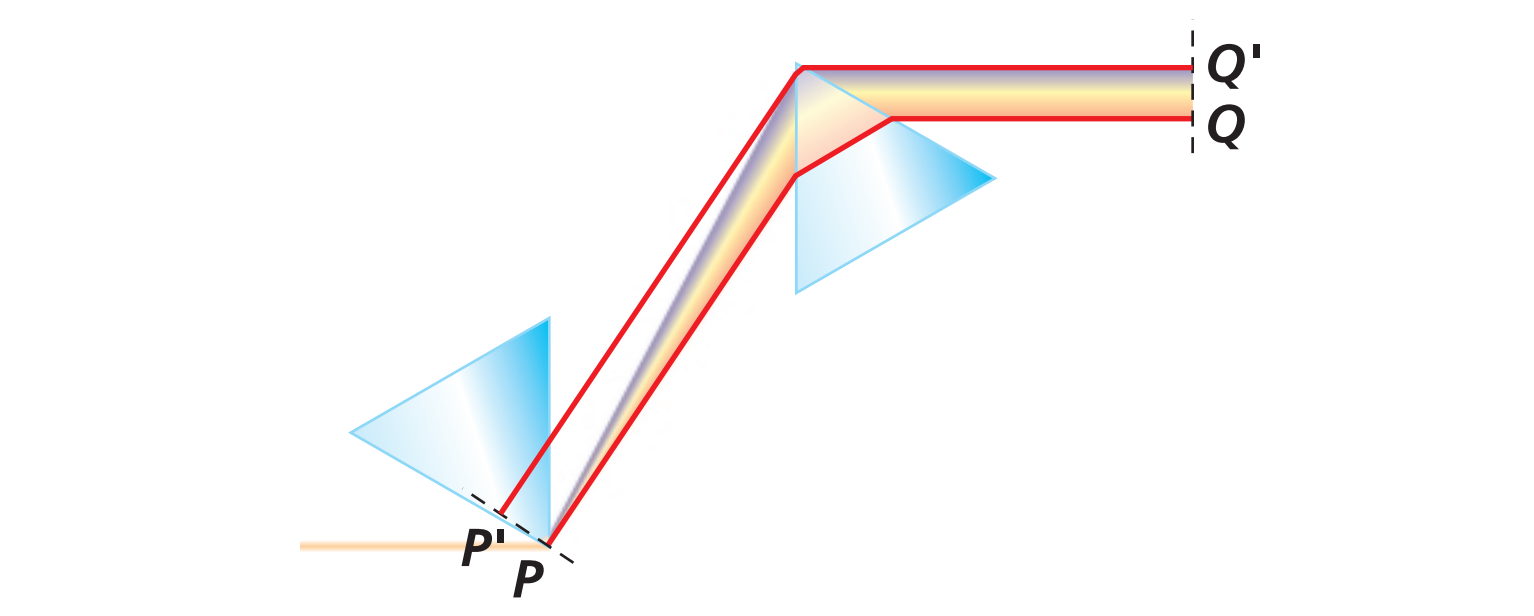
Dispersion compensation

Does path length difference compensate?



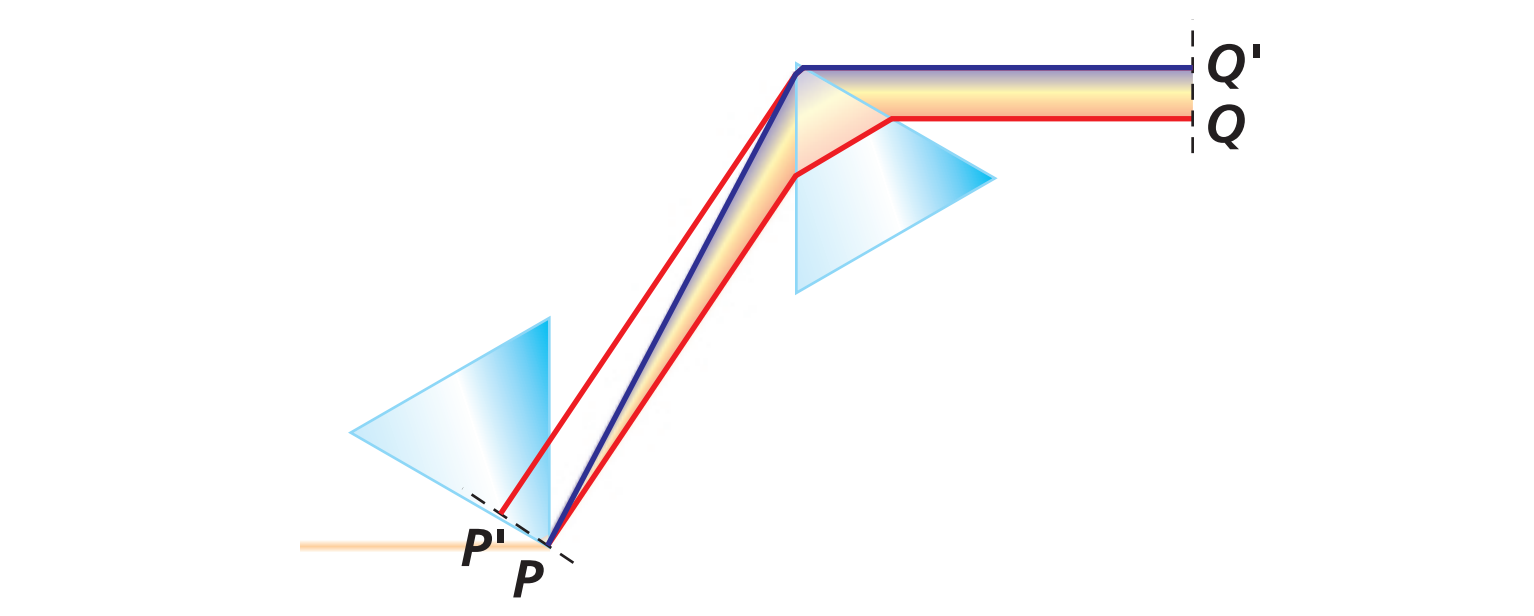
Dispersion compensation

Does path length difference compensate?



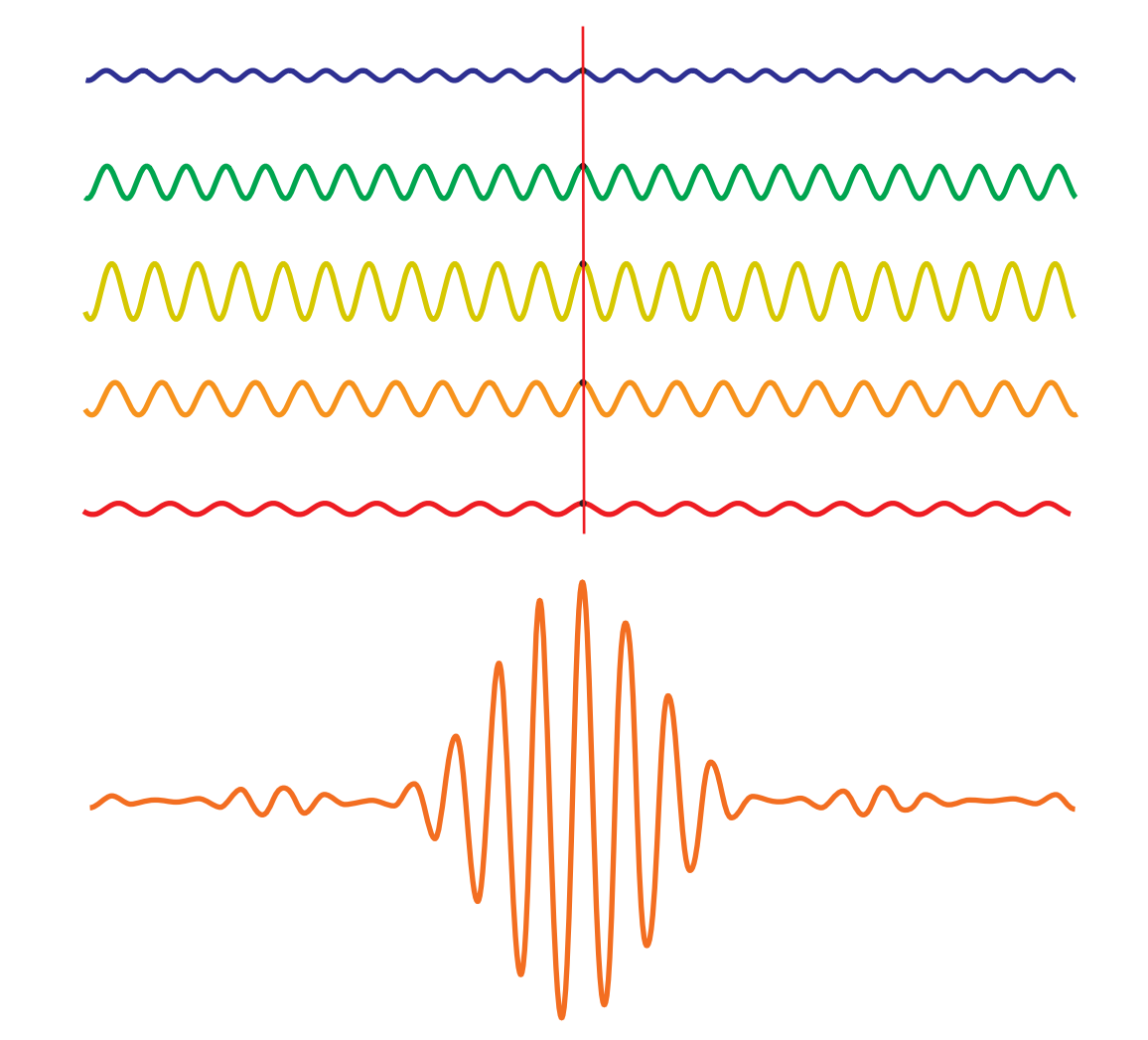
Dispersion compensation

Does path length difference compensate?

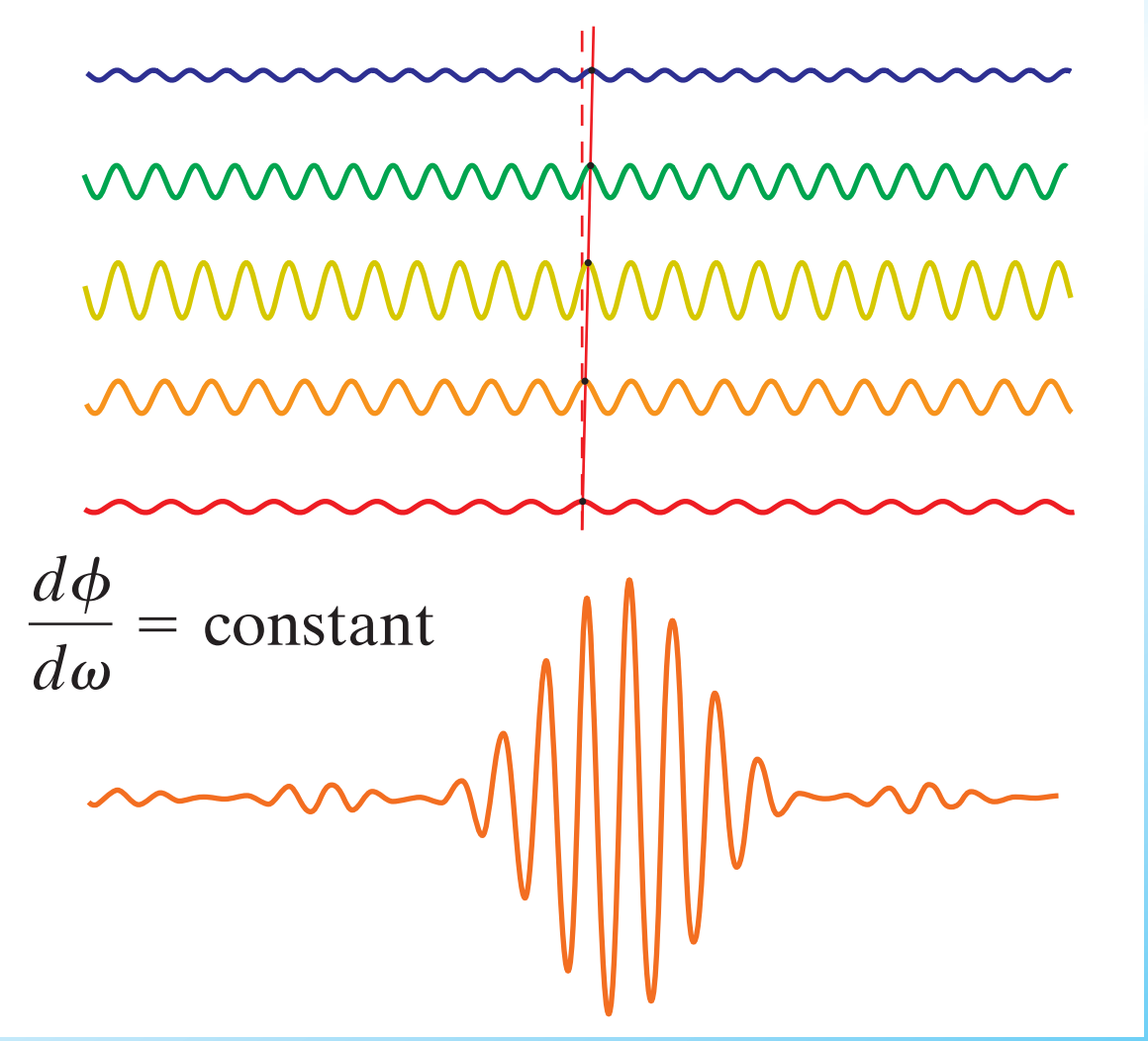


…so prism gives low frequency *shorter* path length…

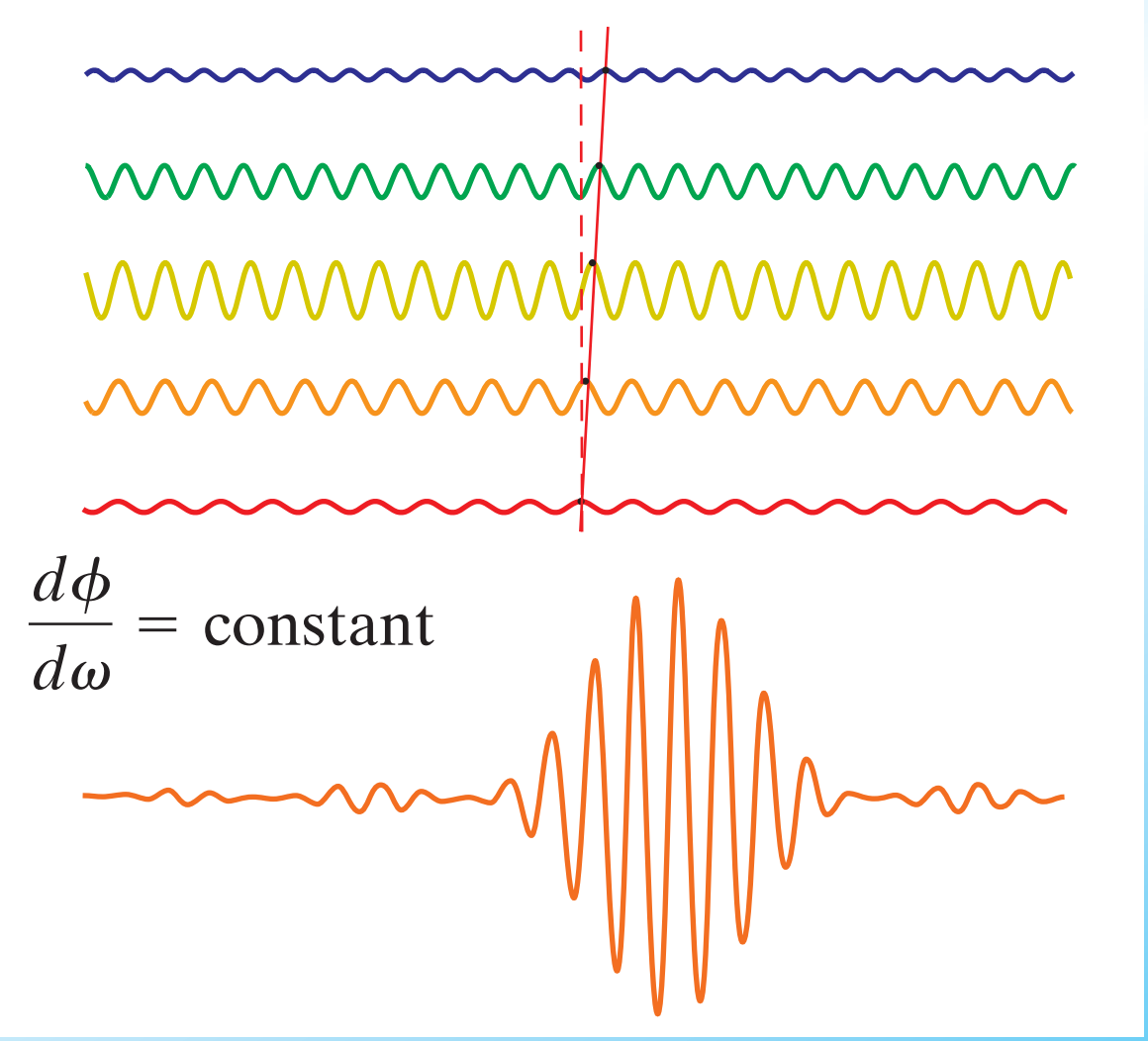
Dispersion compensation



Dispersion compensation



Dispersion compensation



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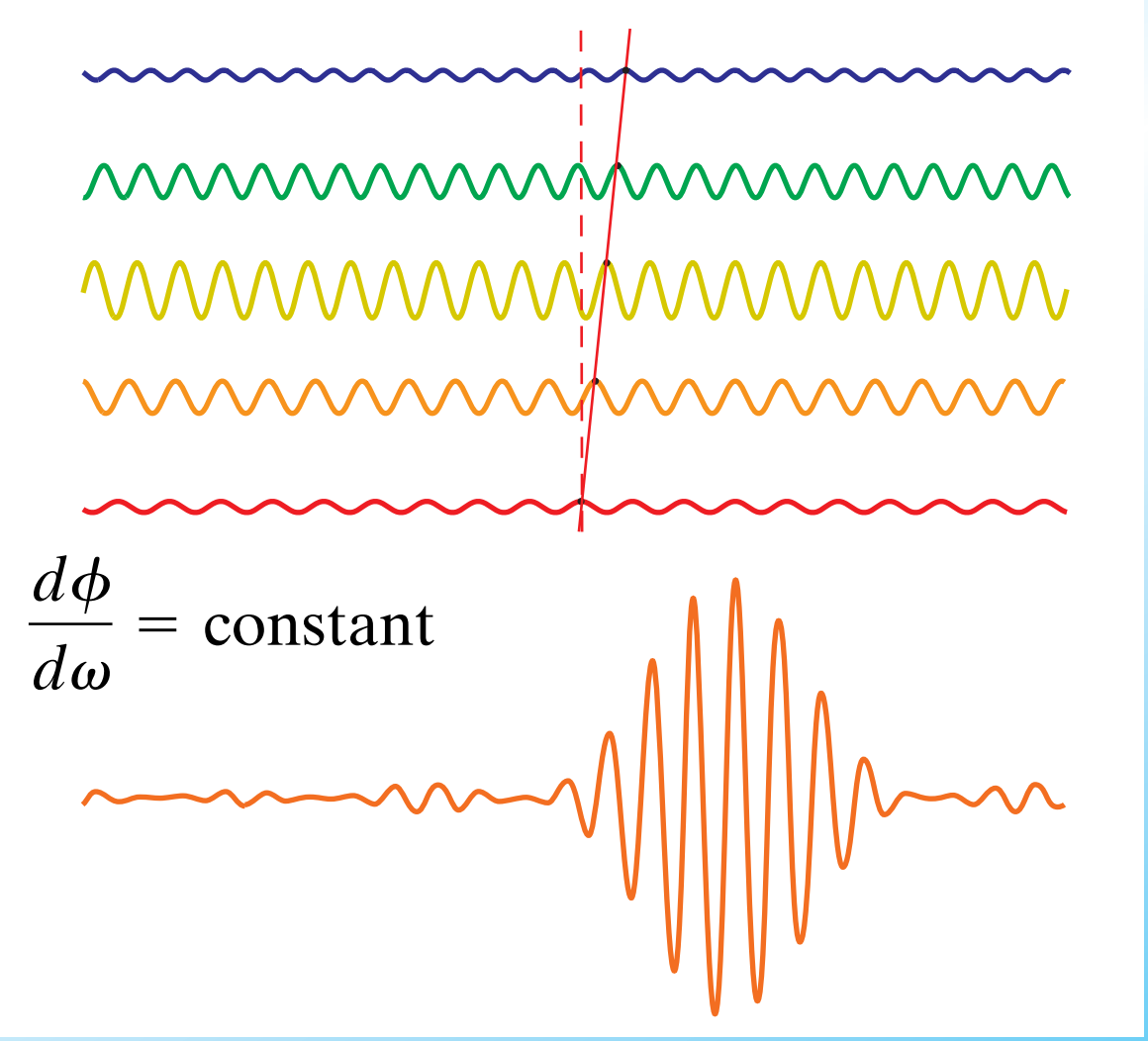
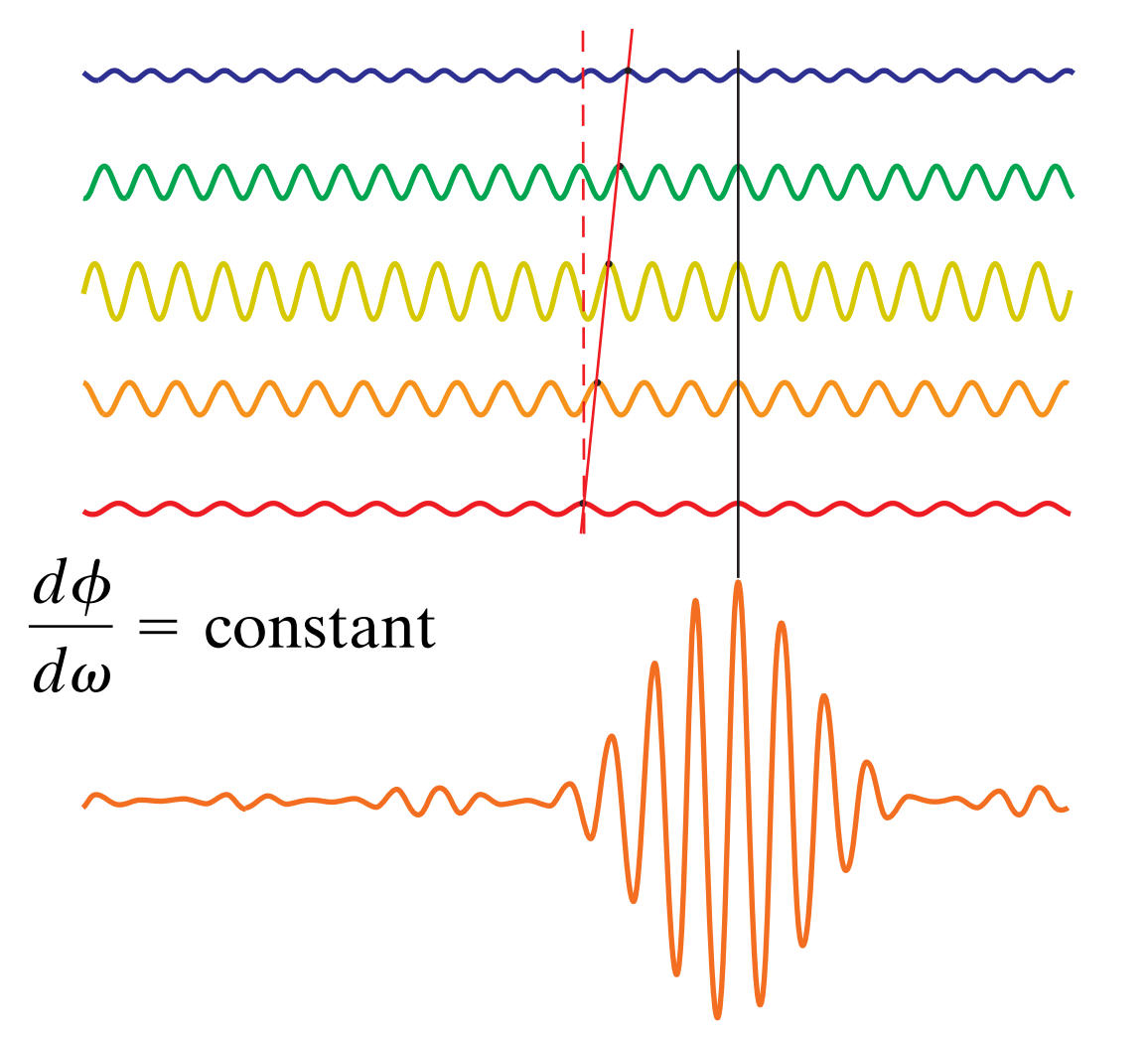
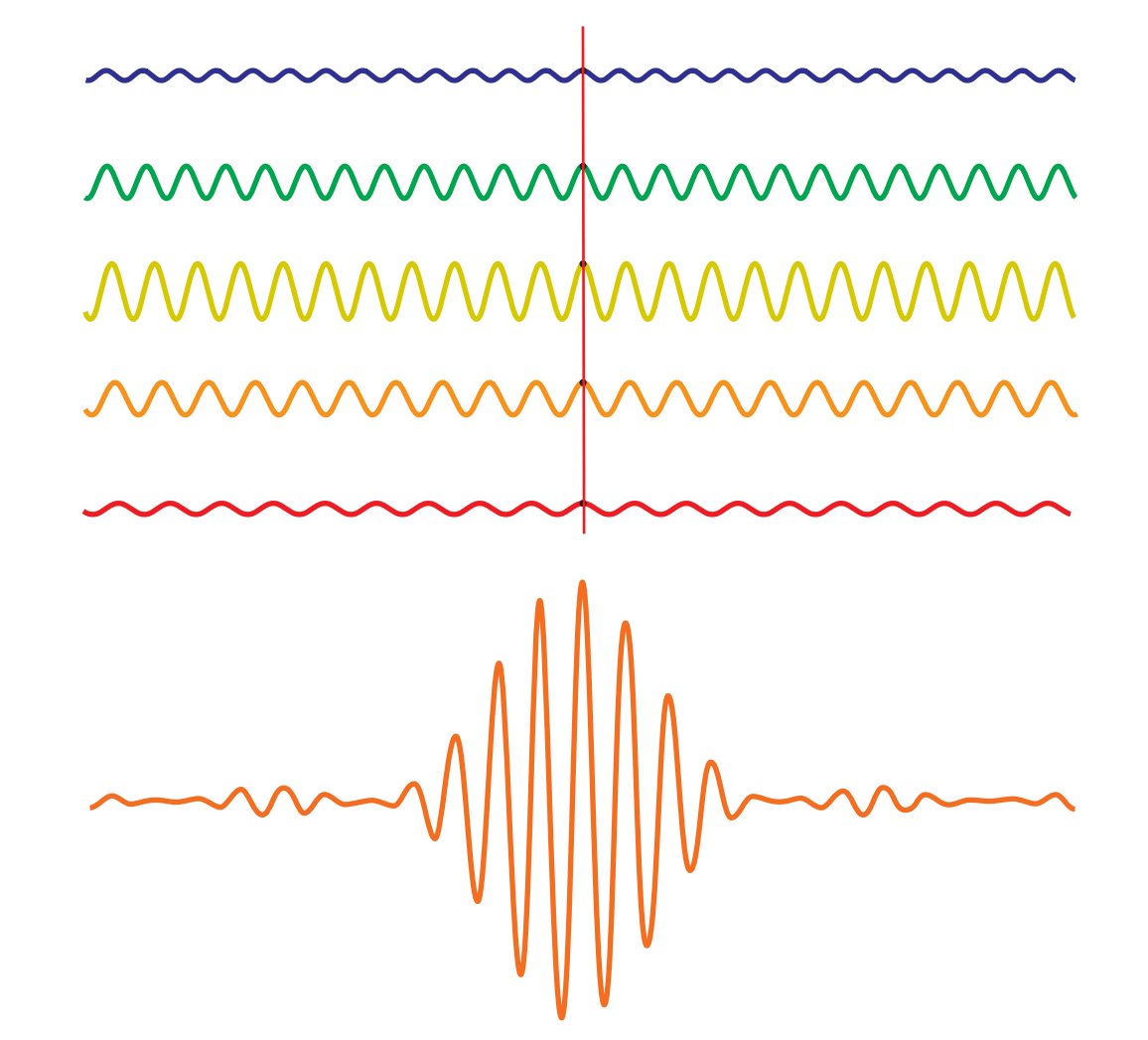


Dispersion compensation



Dispersion compensation



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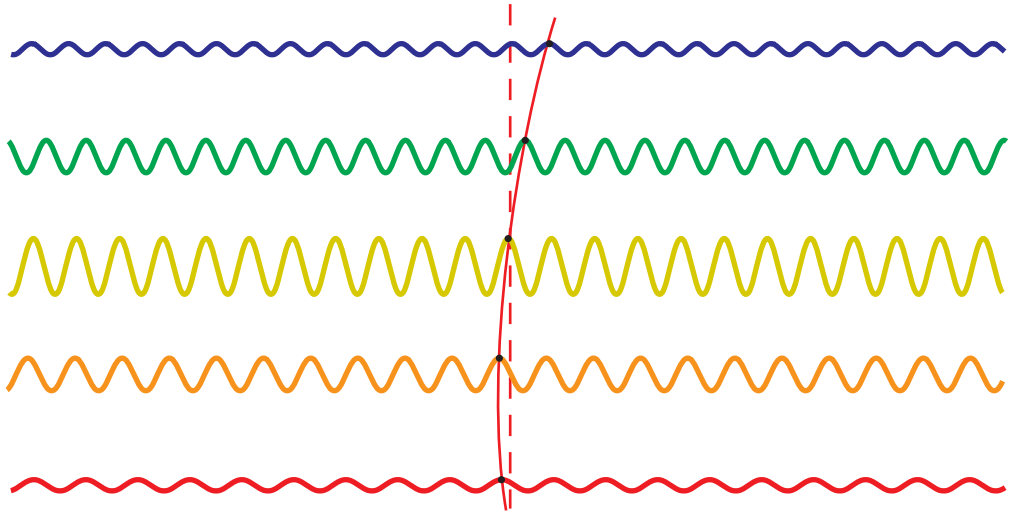
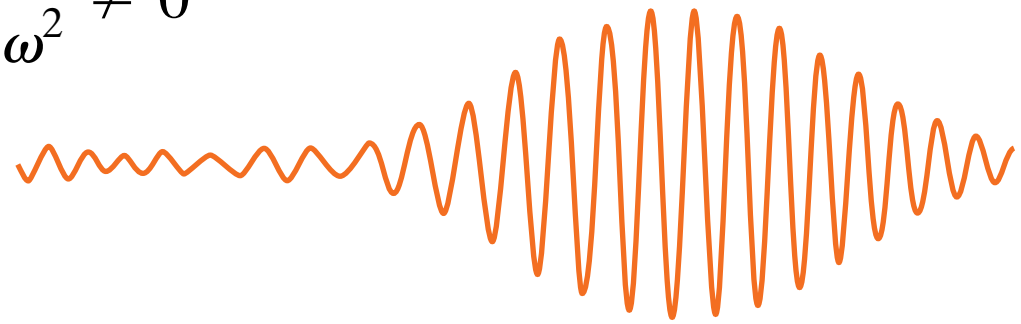


$$\frac{d^2\phi}{d\omega^2} \neq 0$$

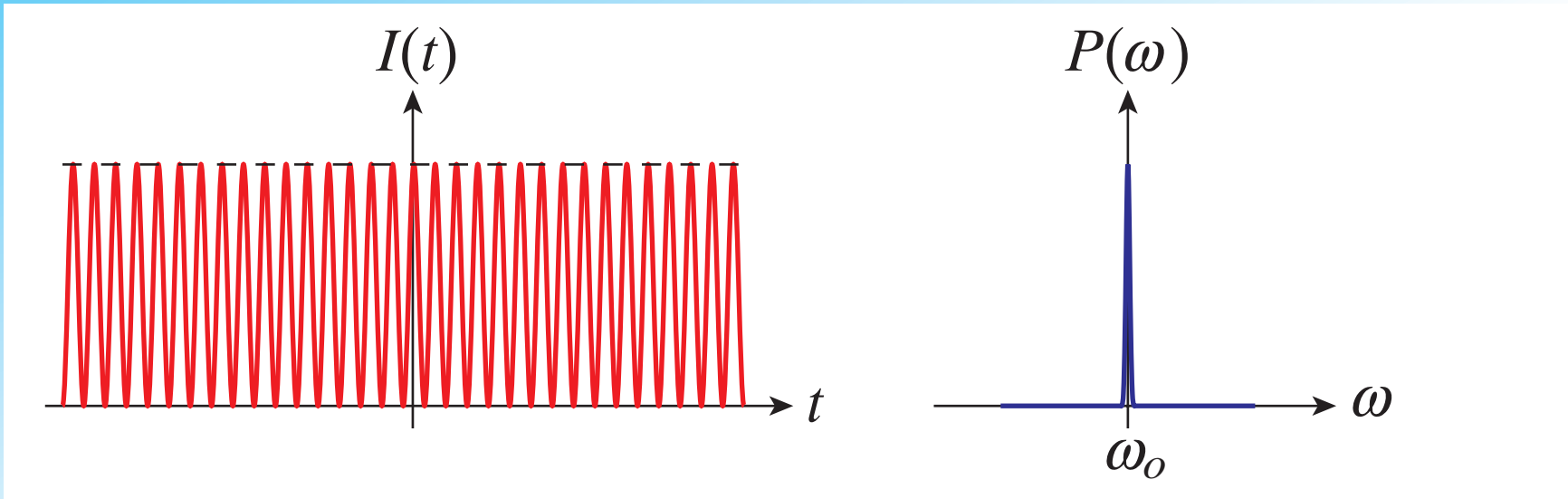


Dispersion compensation

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

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

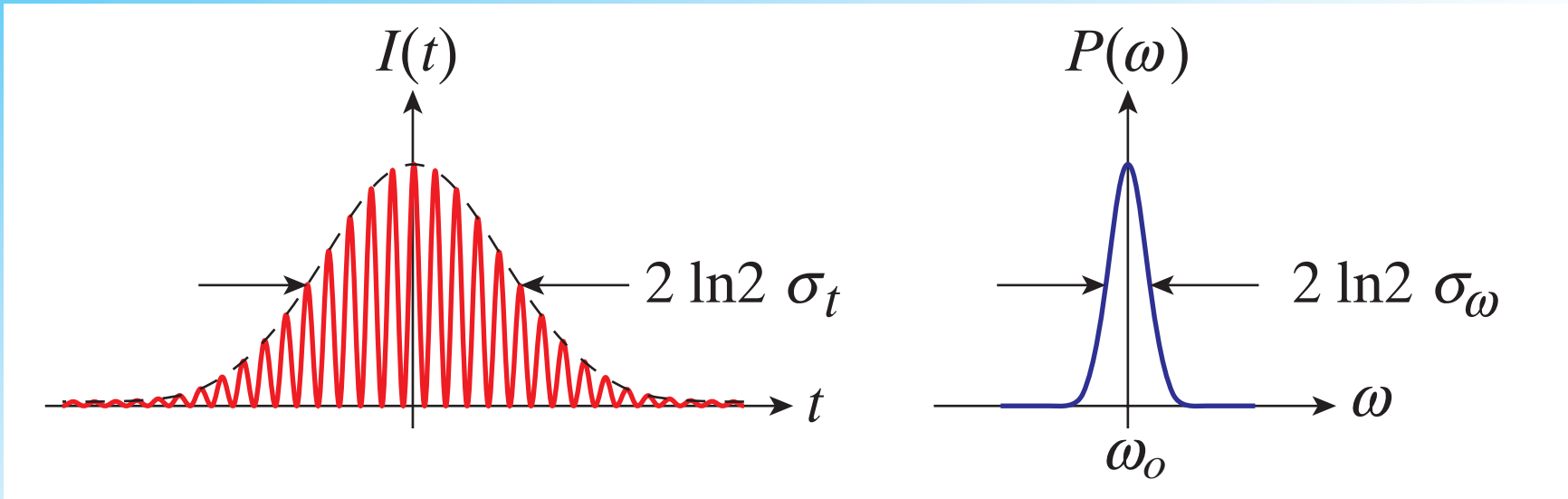
Representation of pulses



Spectrum of sinusoidal intensity is a delta function

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

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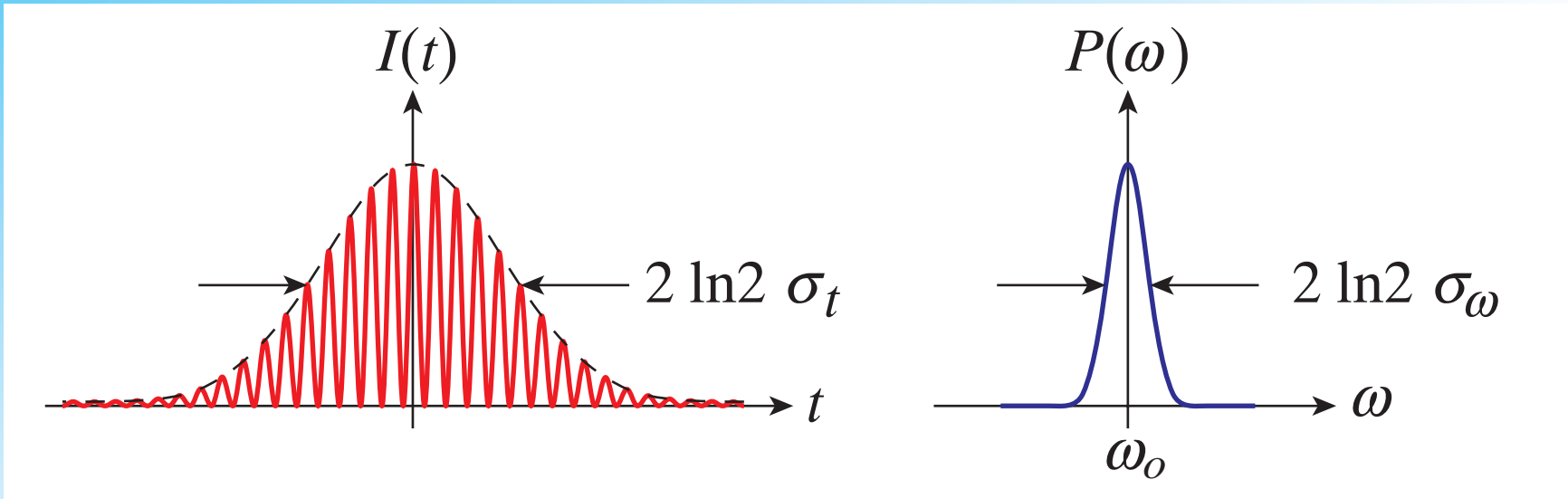
$$= \sigma_t\exp\left[-\frac{\sigma_t^2(\omega-\omega_o)^2}{2}\right] \equiv \sigma_t\exp\left[-\frac{(\omega-\omega_o)^2}{2\sigma_\omega^2}\right]$$

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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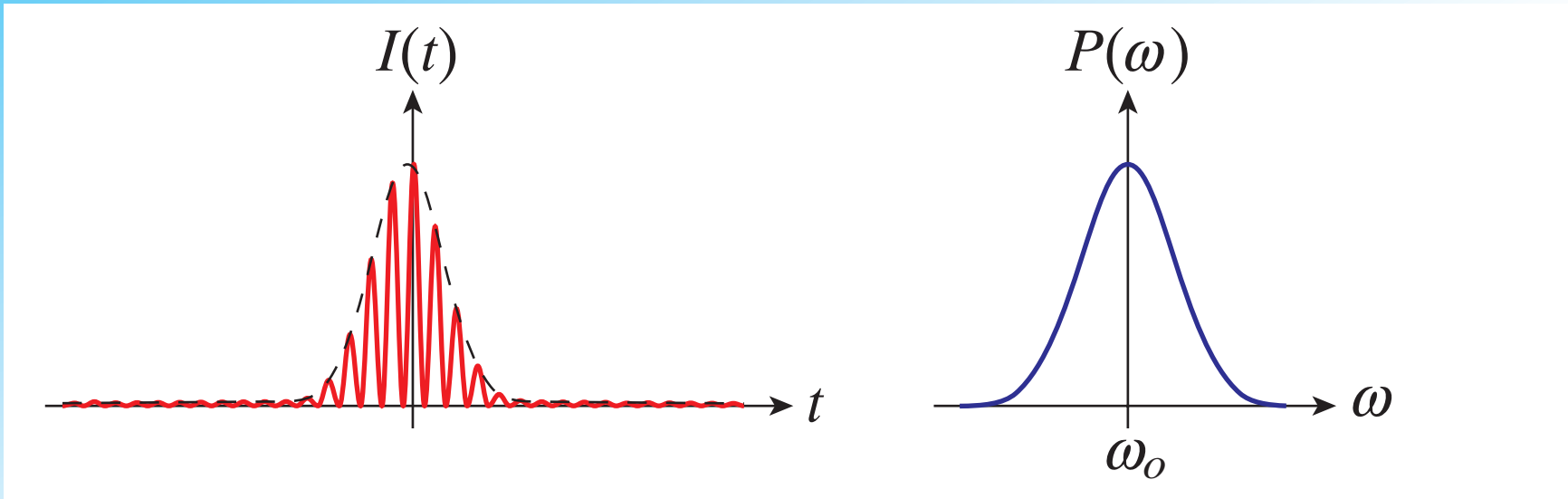

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Joint time-frequency representation

Wigner representation:

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

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

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Joint time-frequency representation

Energy:

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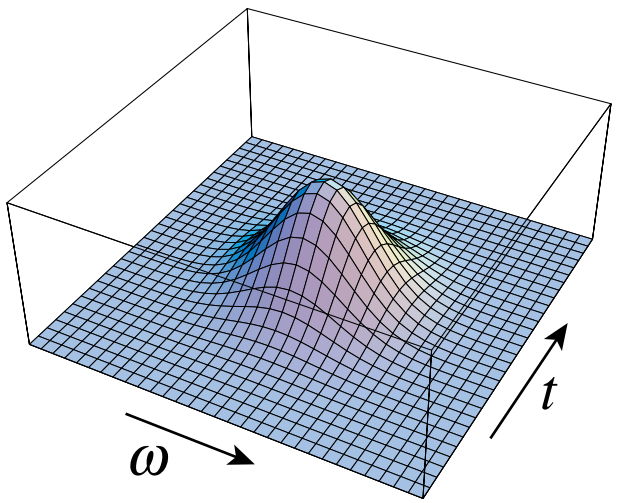

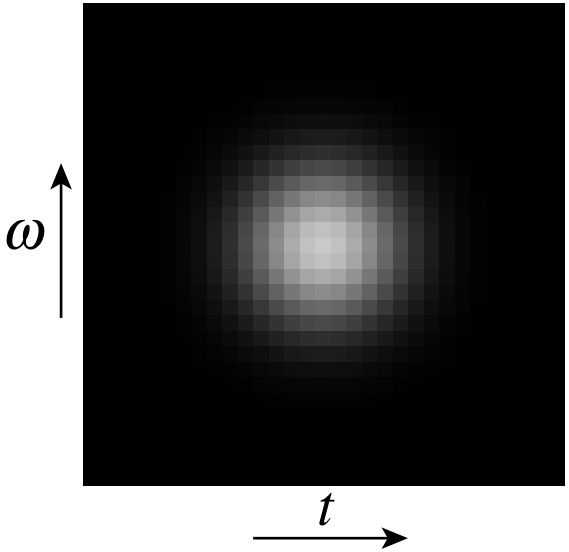

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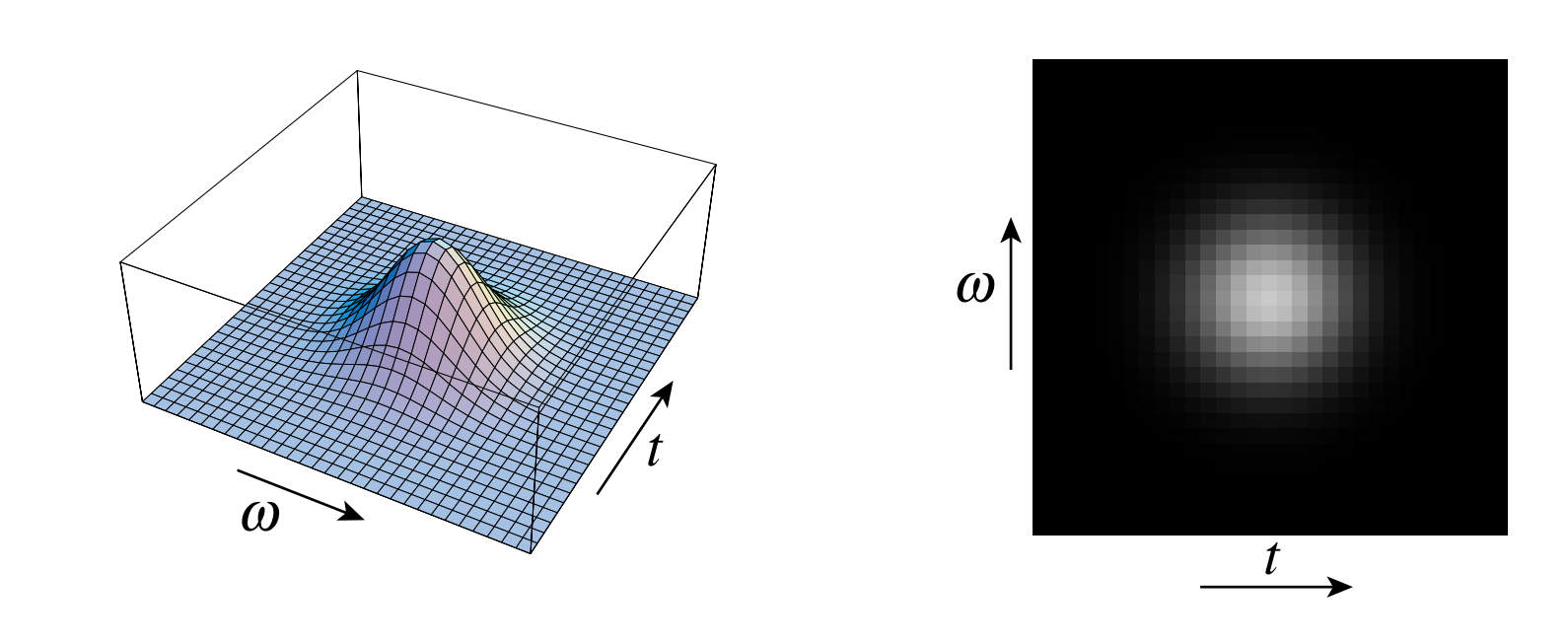

$W(t,\omega)$ **must be nonzero in phase-space area larger than** π

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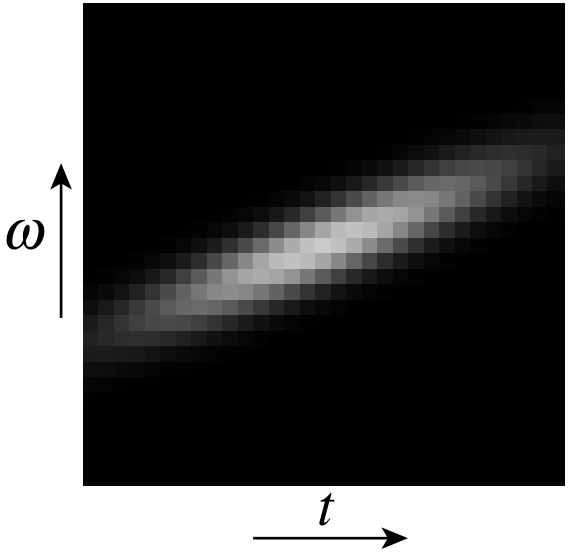
chirped pulse



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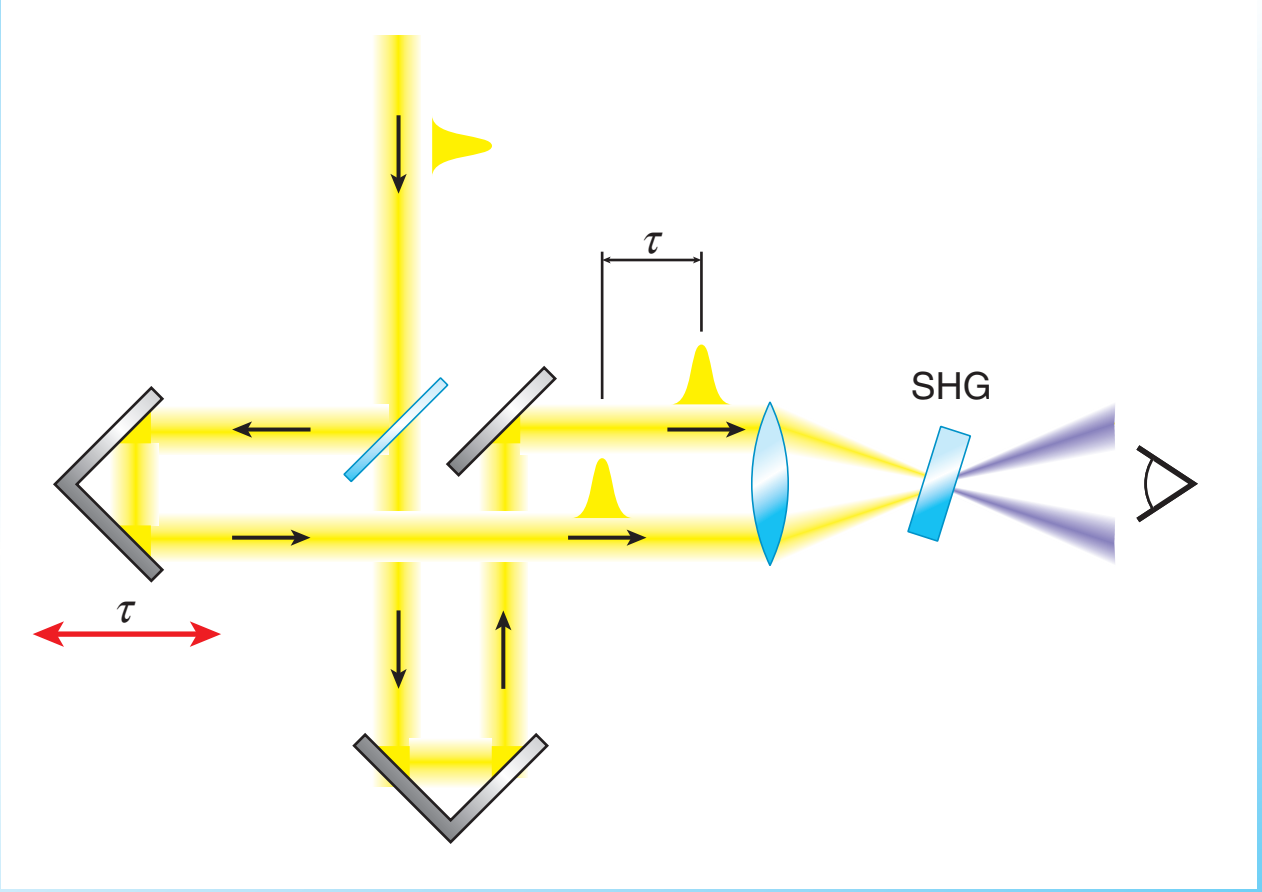
Temporal characterization

Use pulse to measure itself…



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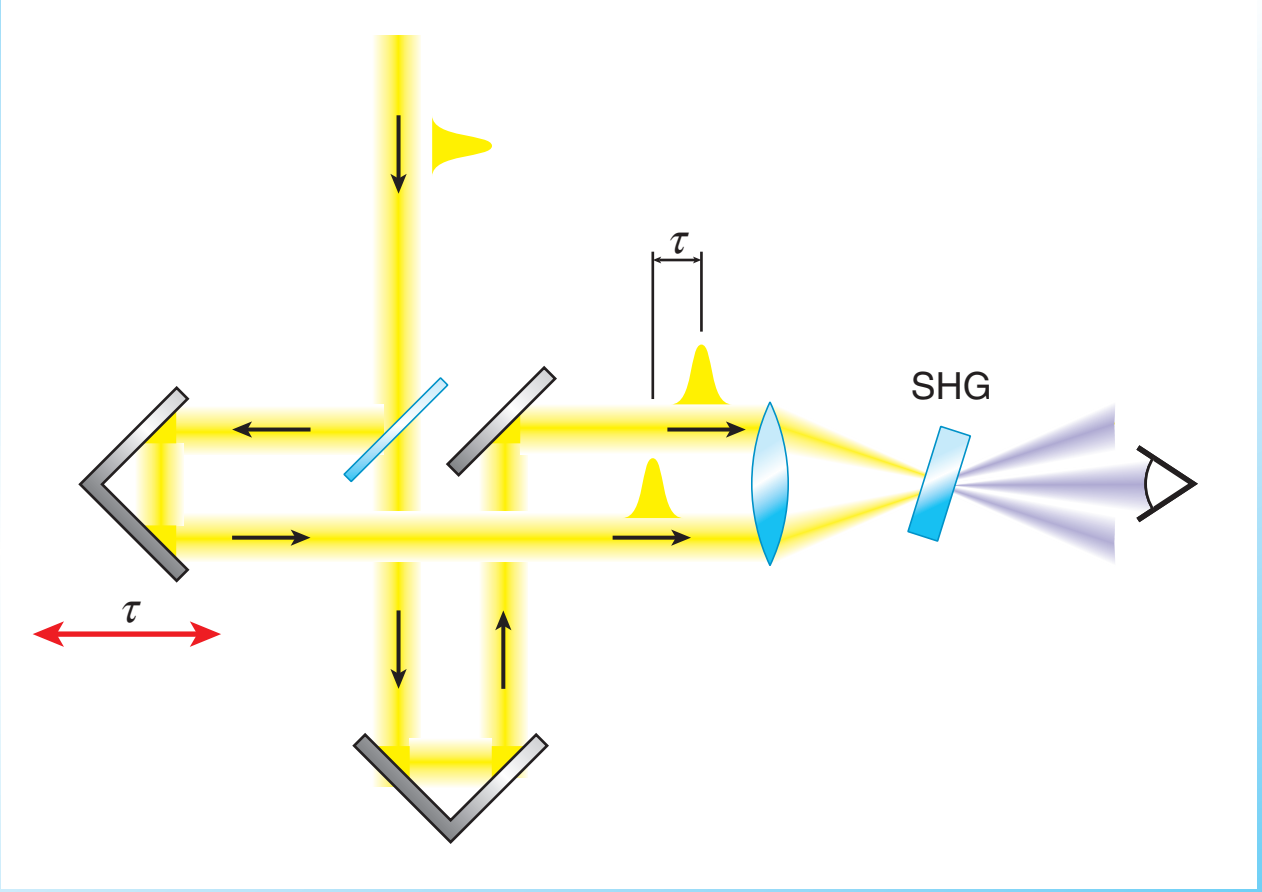

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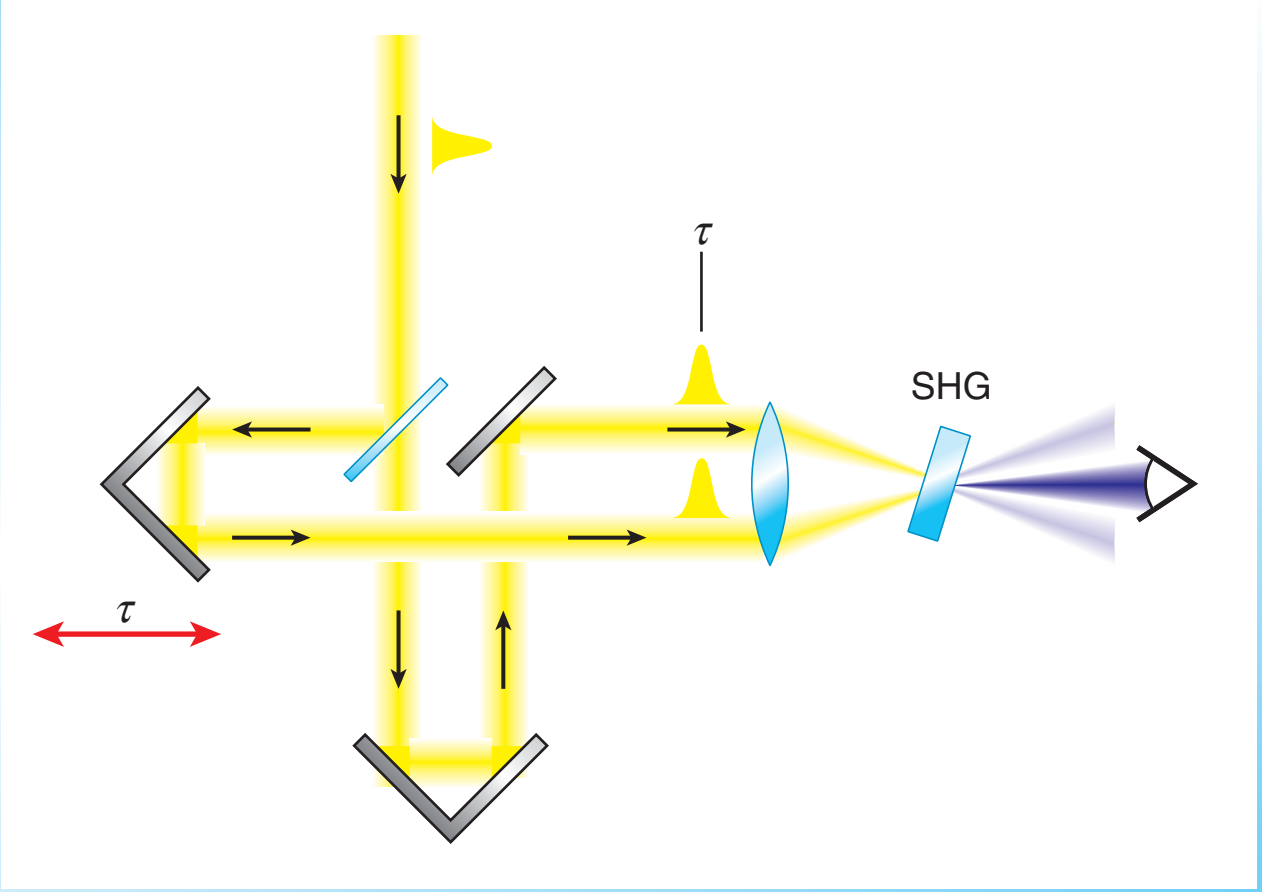

Temporal characterization

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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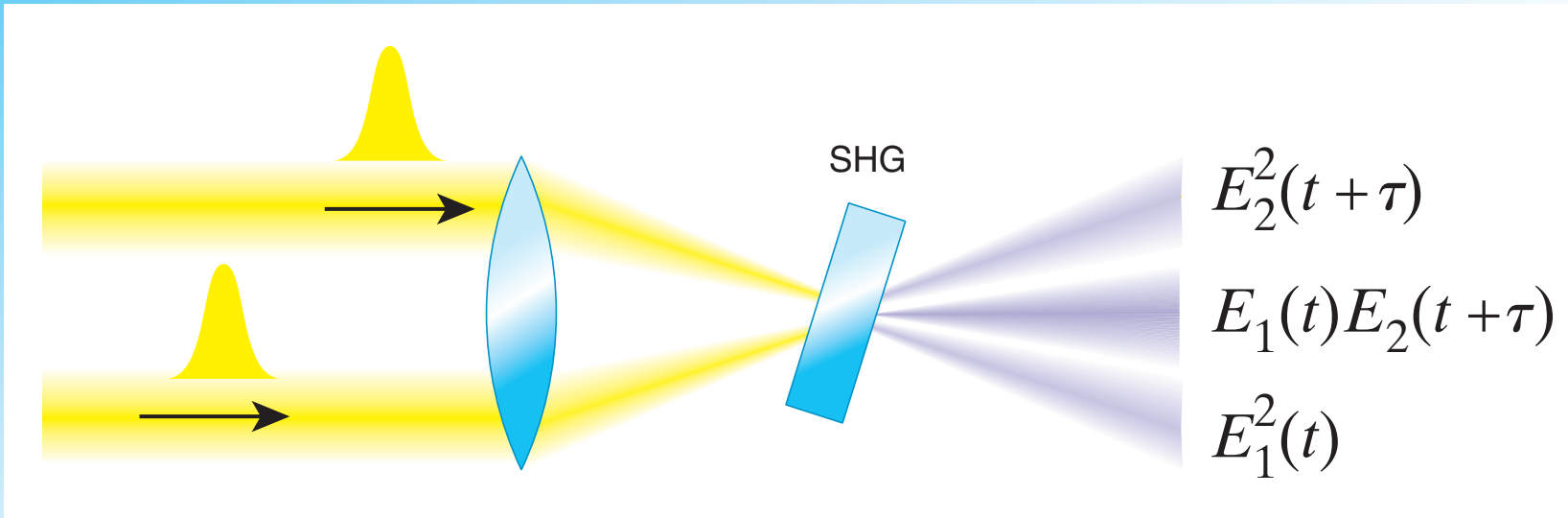
Temporal characterization



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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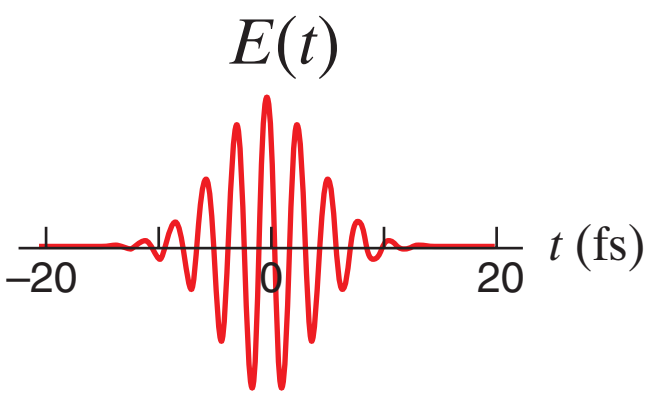

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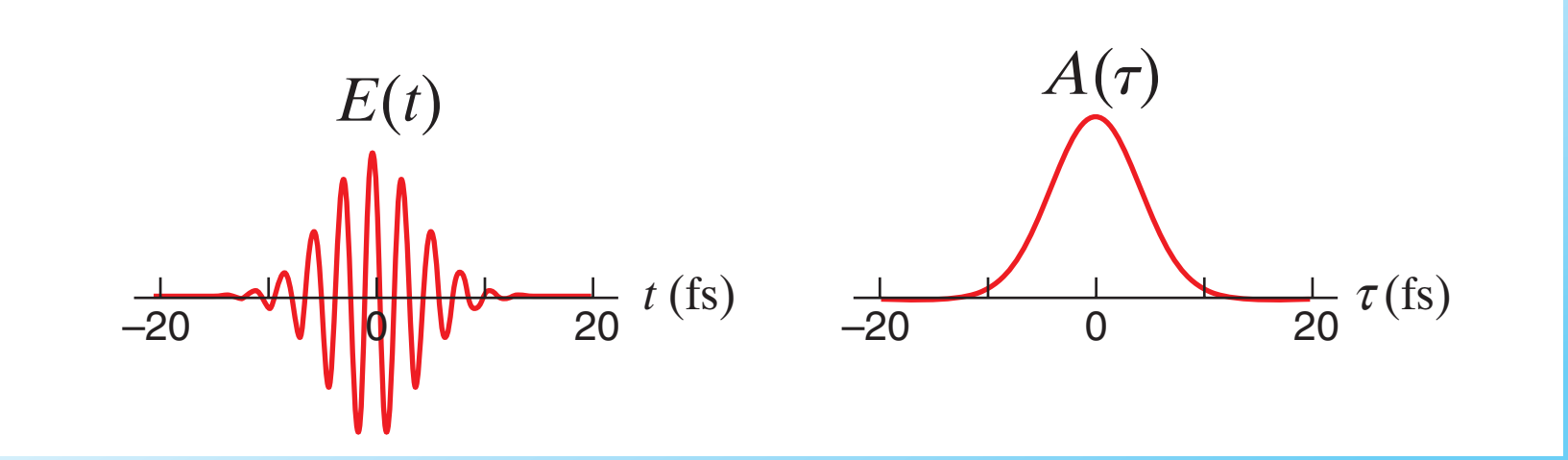
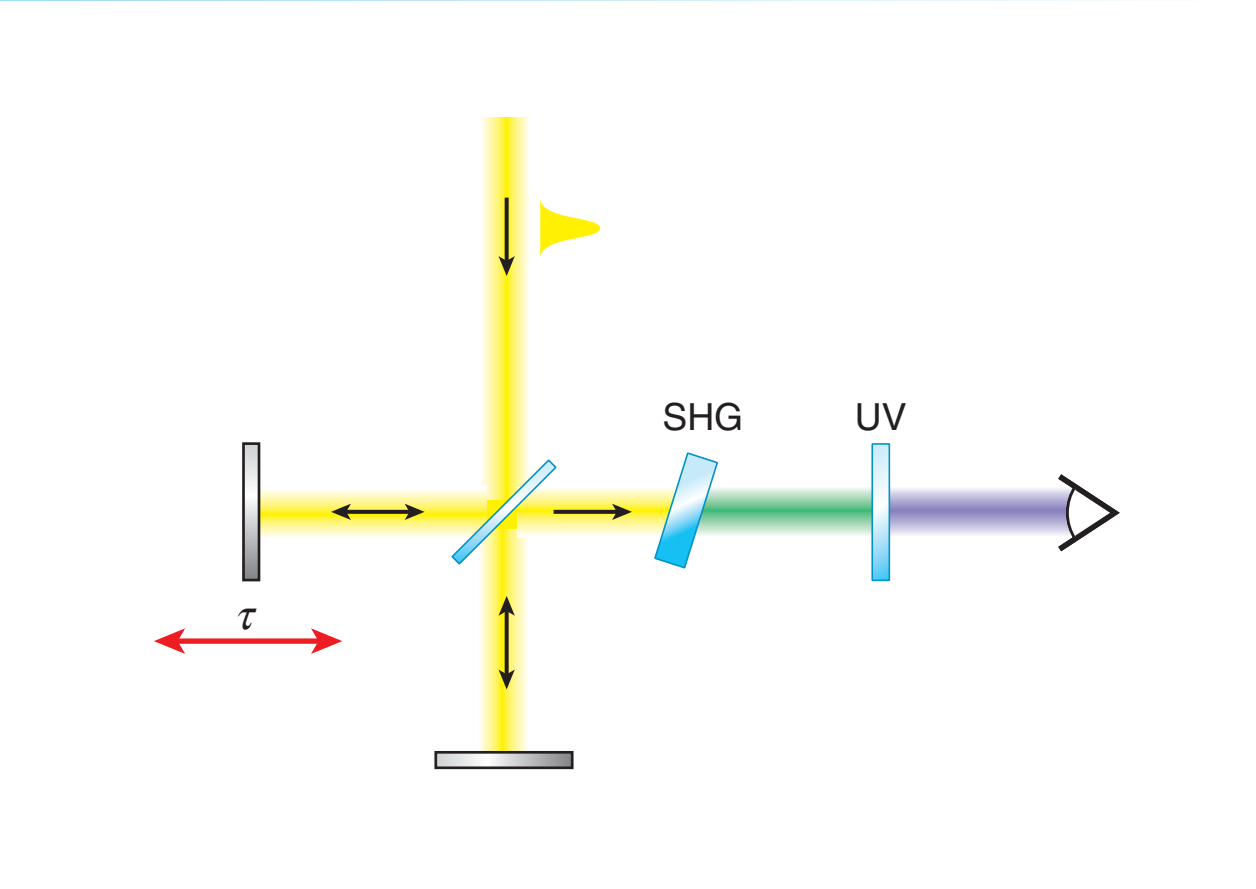


Temporal characterization

Alternative colinear geometry



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All terms now contribute:

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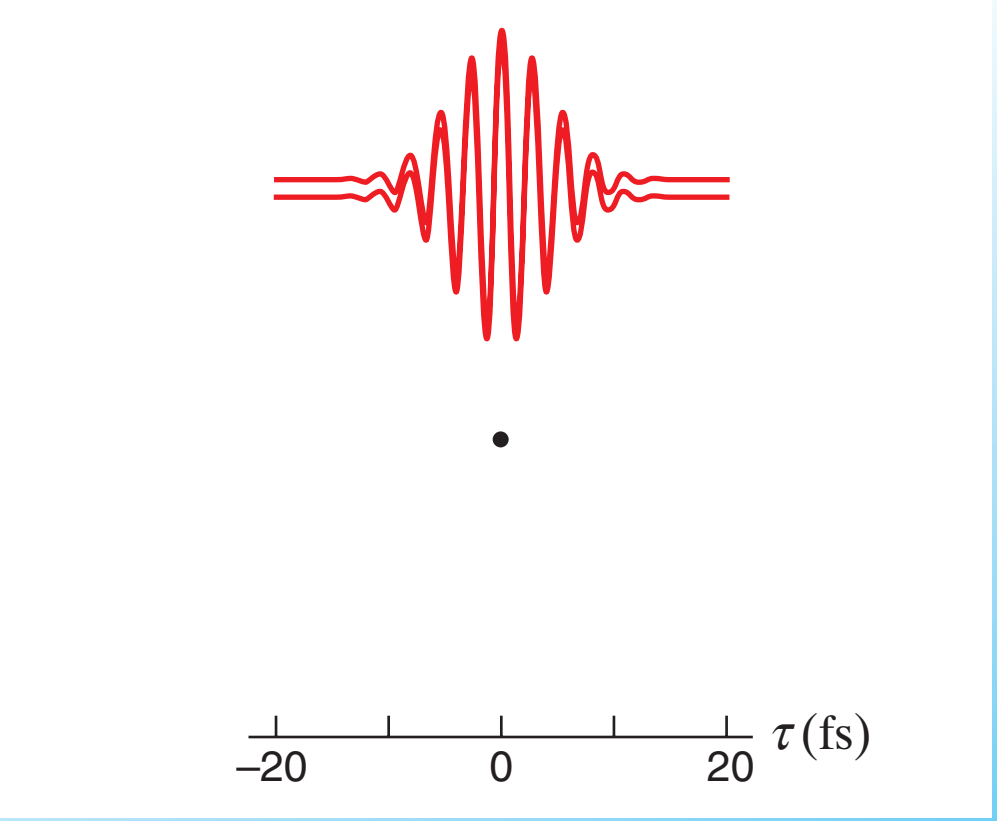

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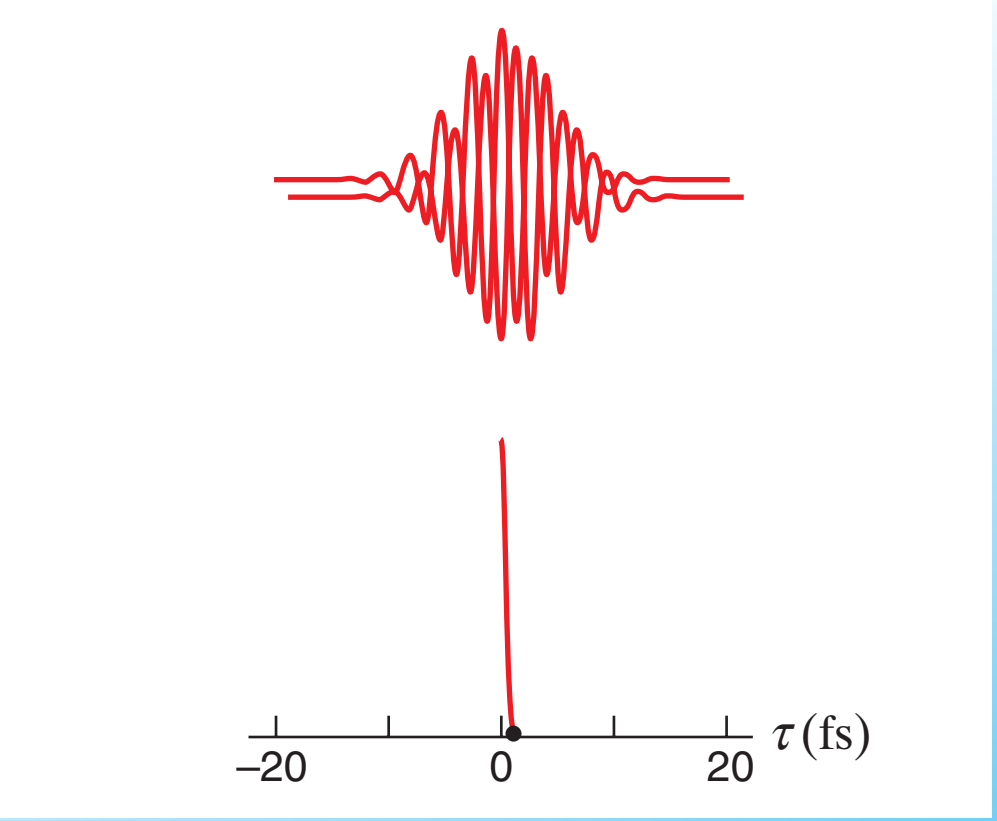

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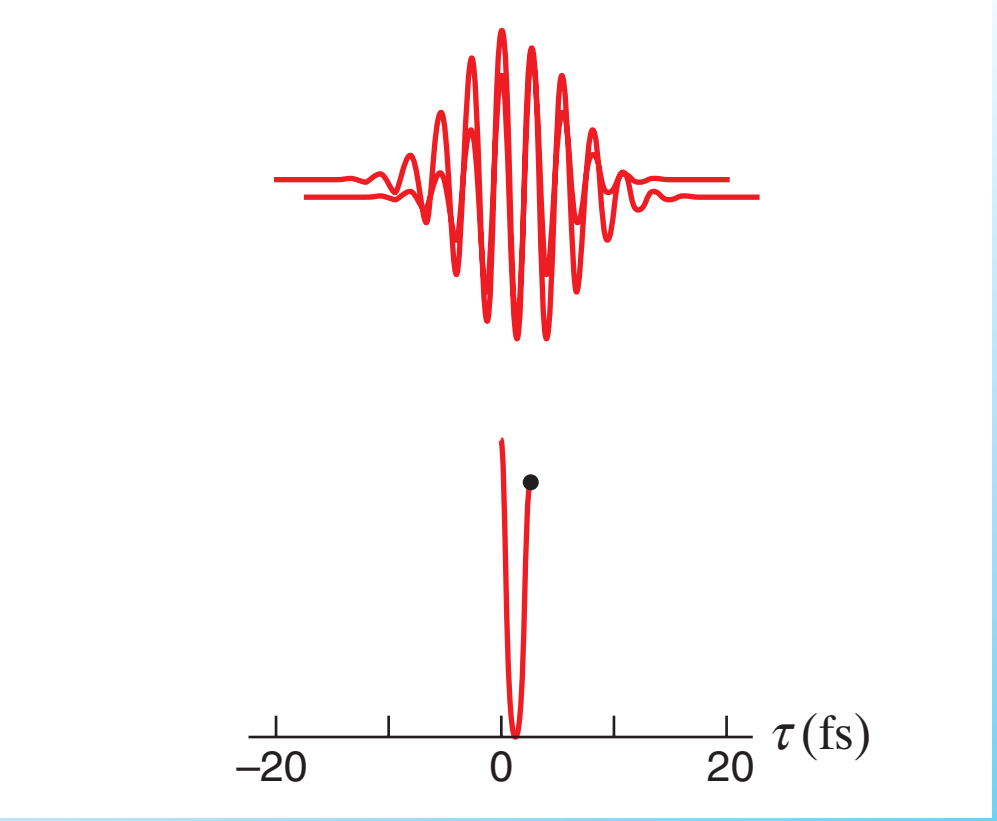

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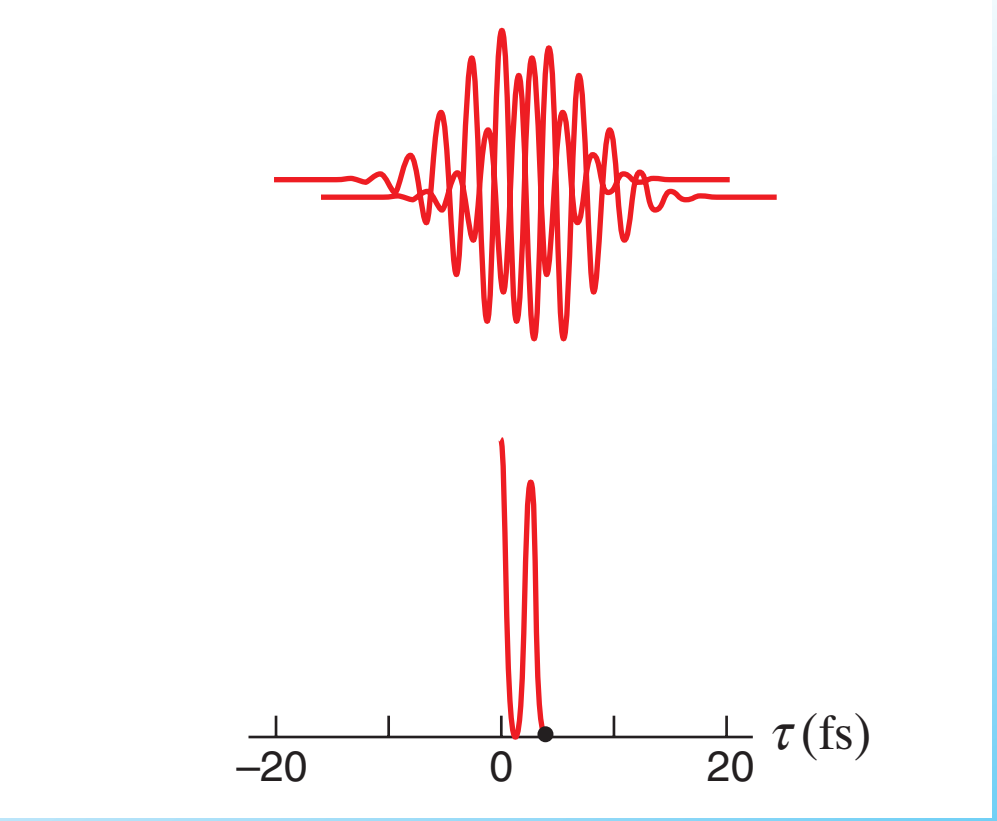

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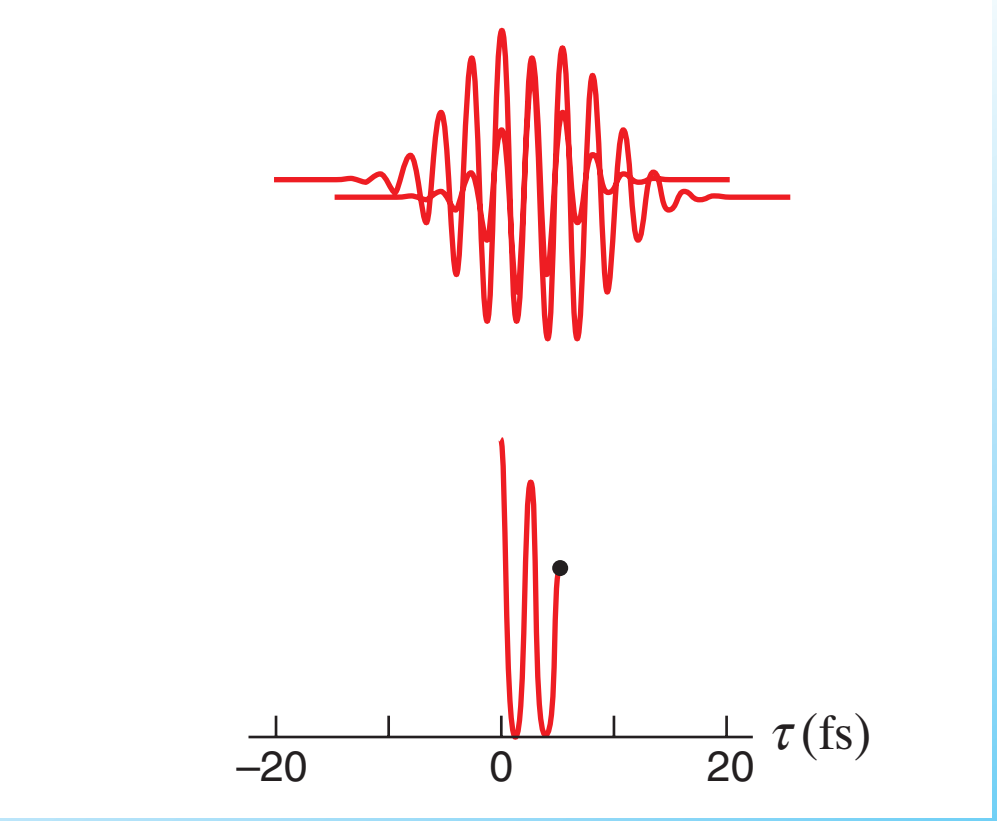

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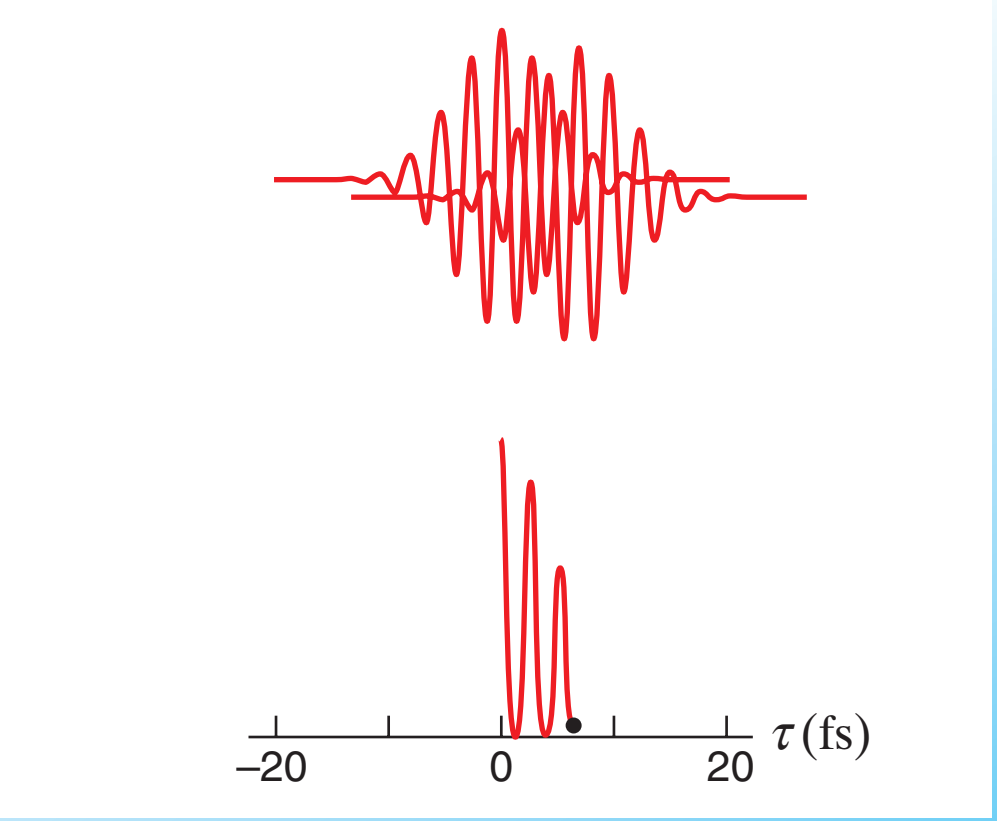

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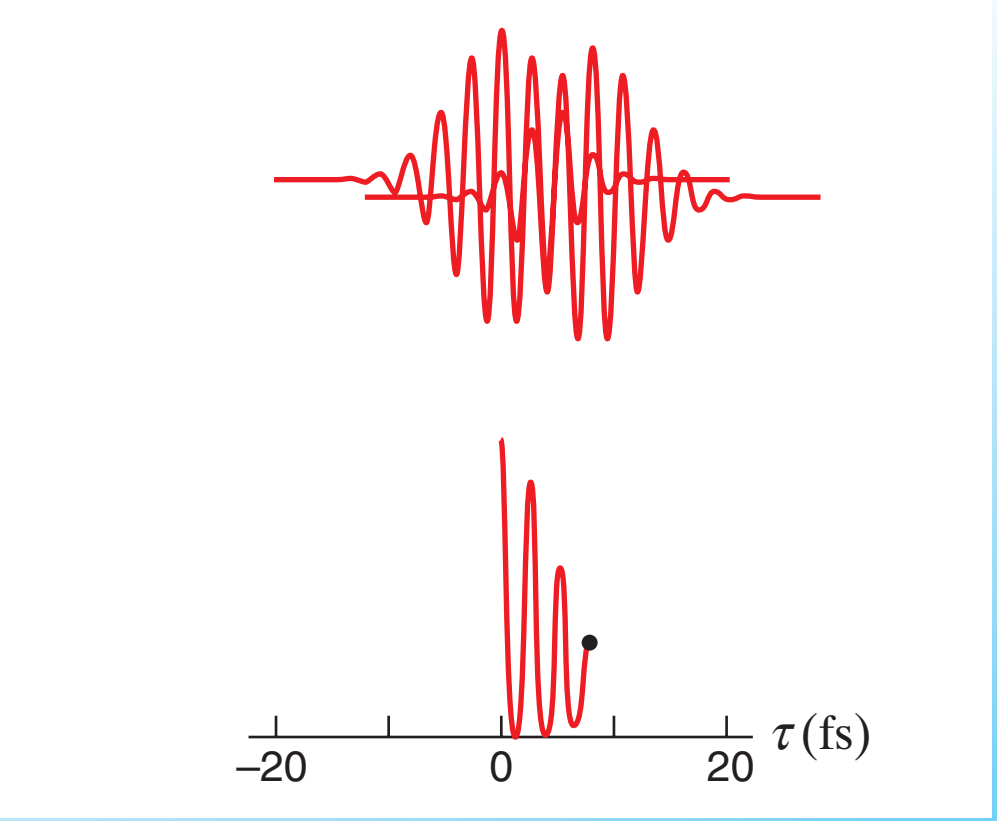

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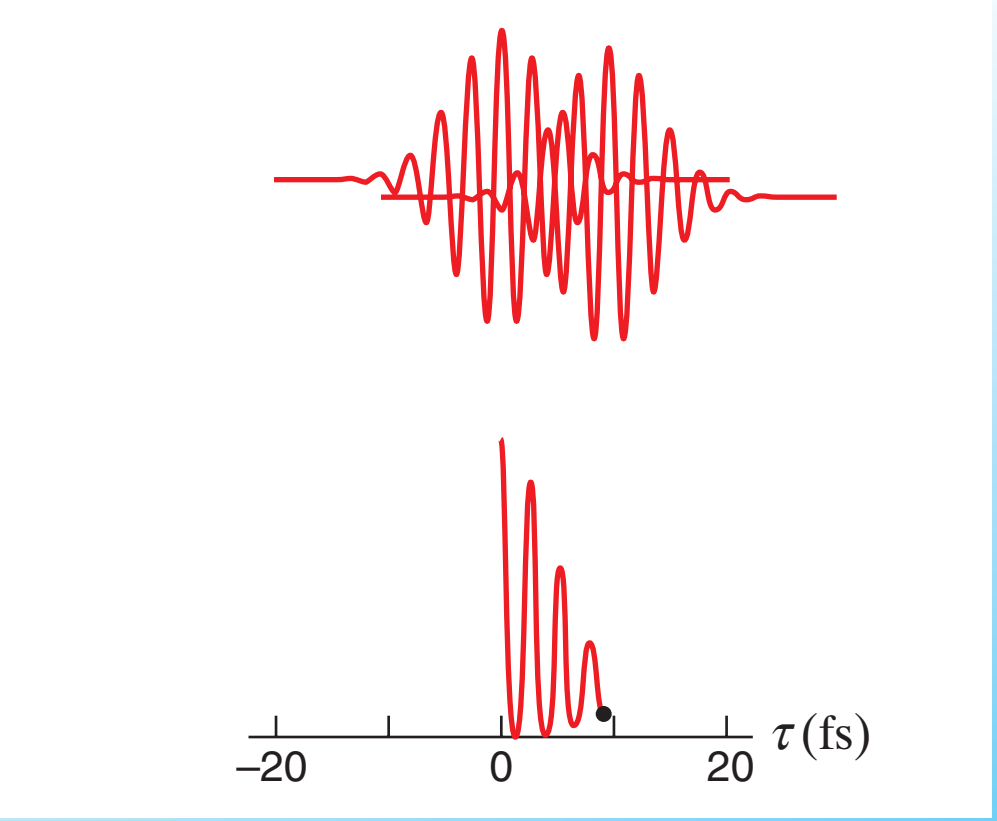

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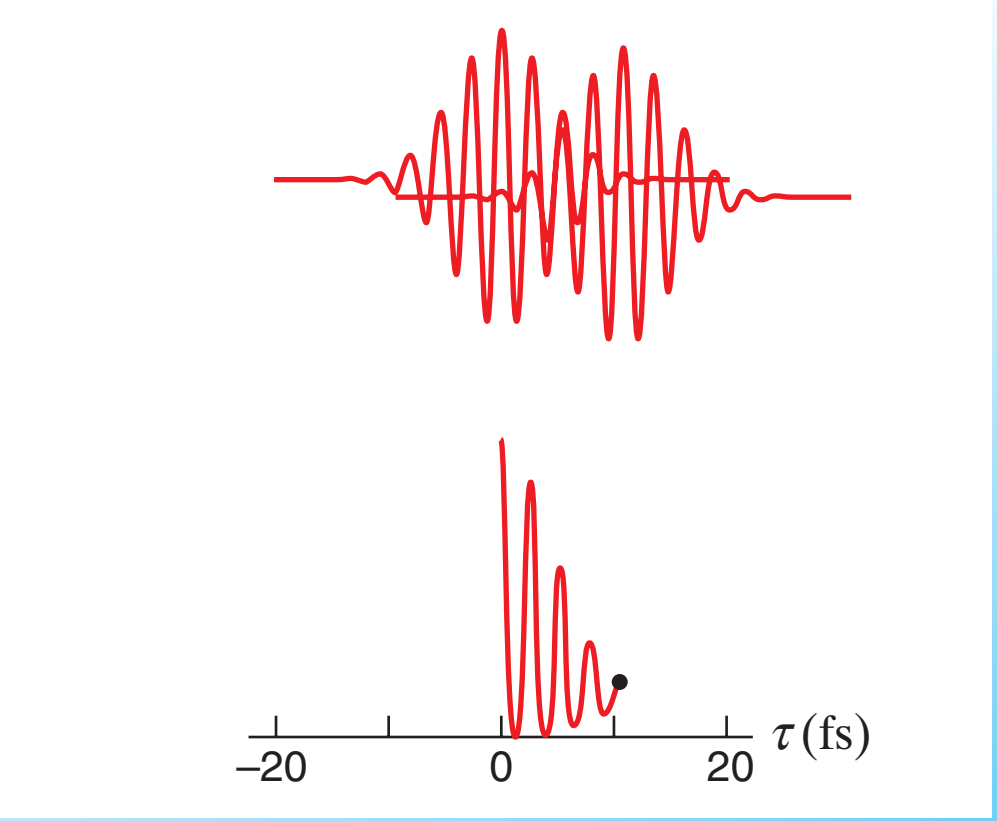

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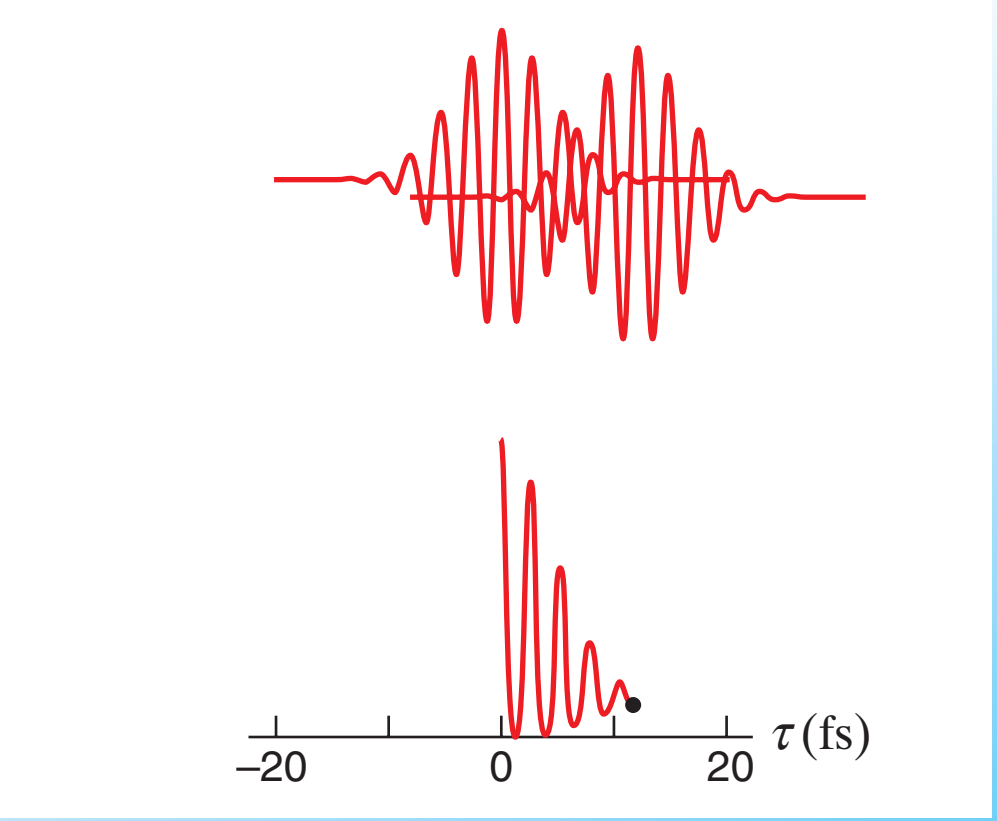

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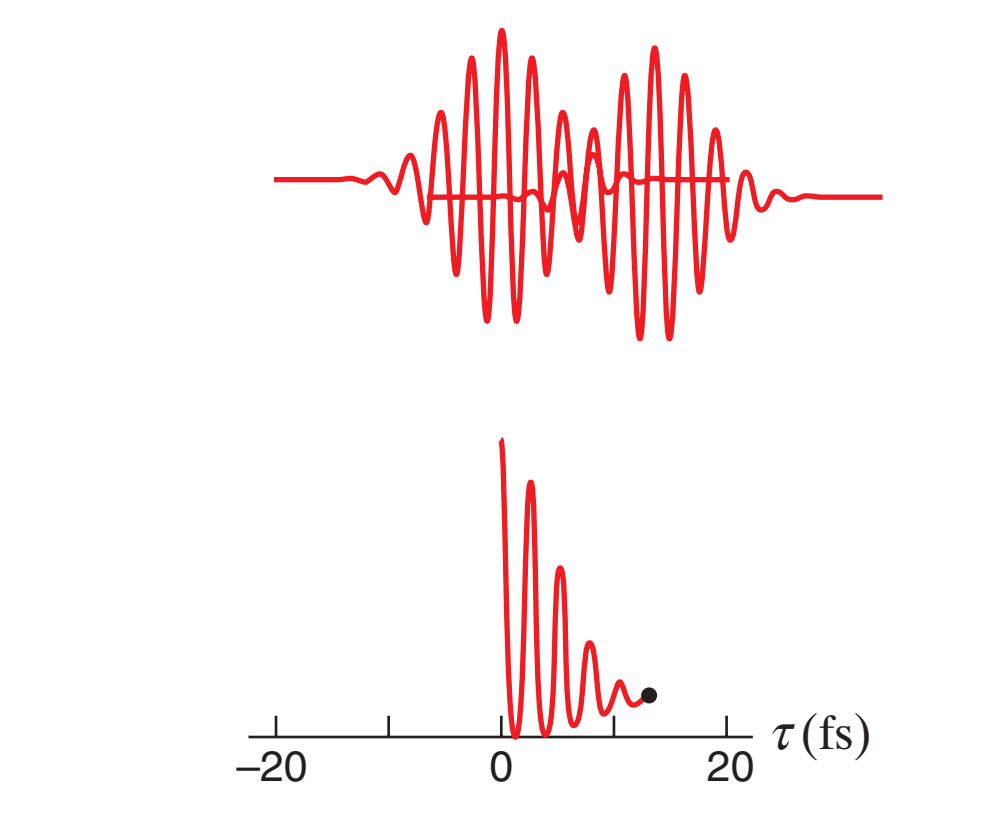

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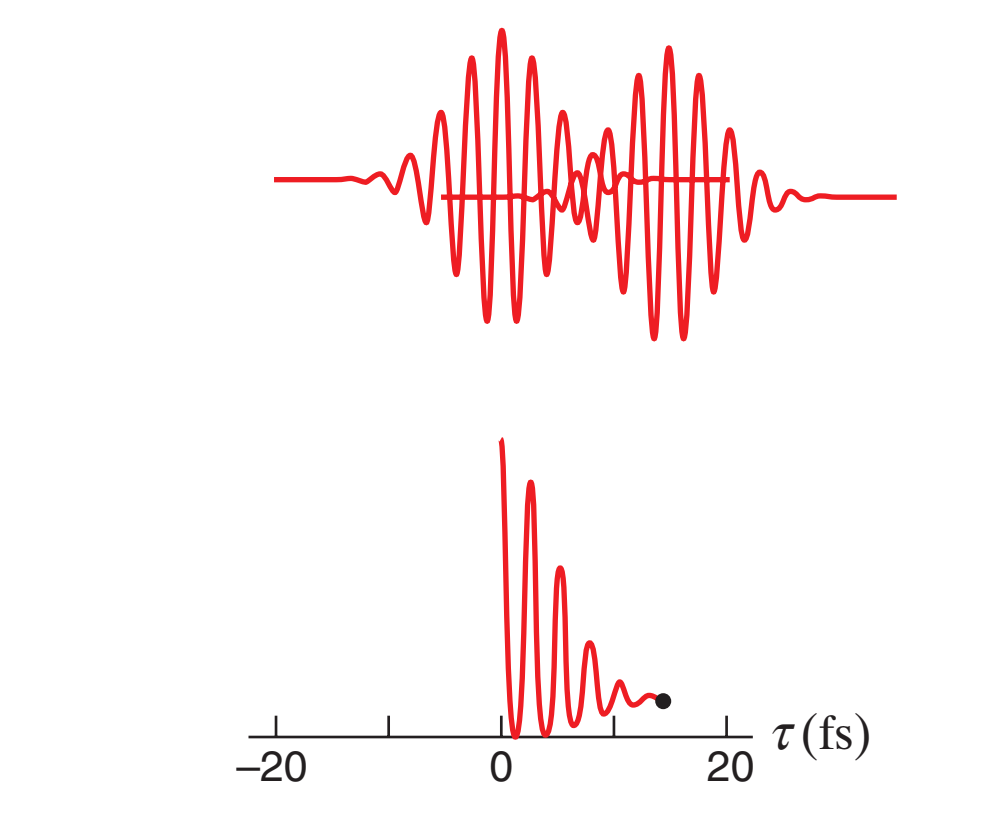

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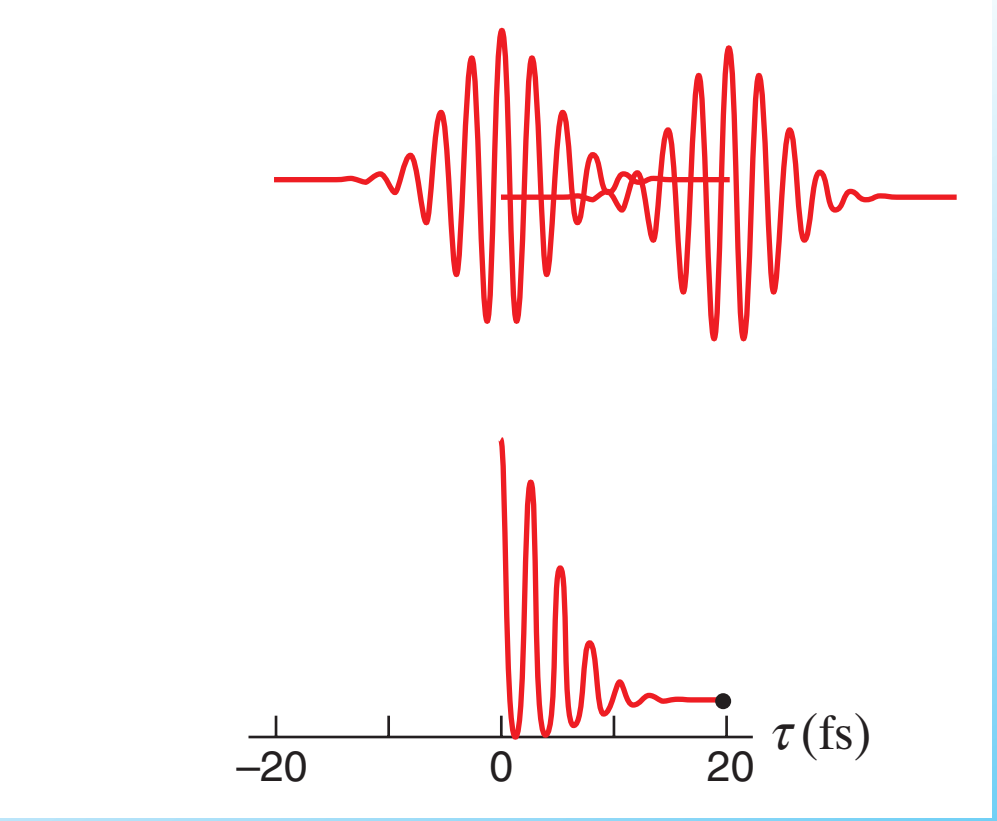

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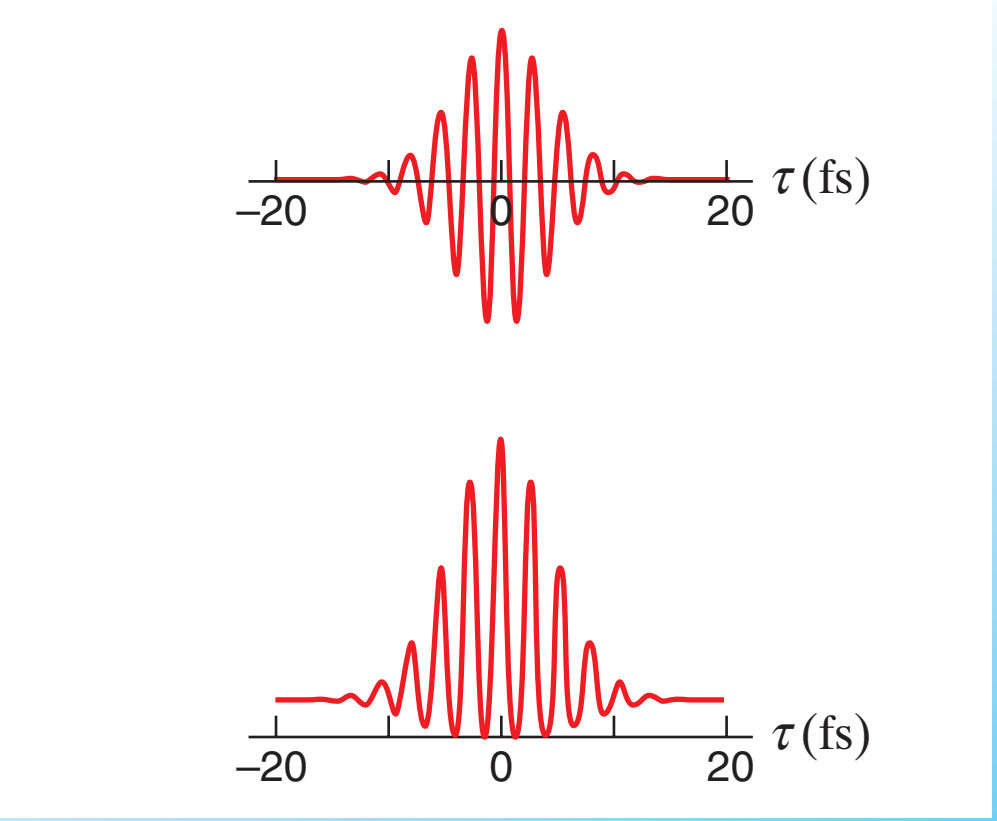
$$I_{2\omega}(t,\tau) \propto \left|\chi^{(2)}\right|^2 \left|E_{tot}^2\right|^2 = \left|\chi^{(2)}\right|^2 \left|E_1^2(t) + 2E_1(t)E_2(t+\tau) + E_2^2(t+\tau)\right|^2$$



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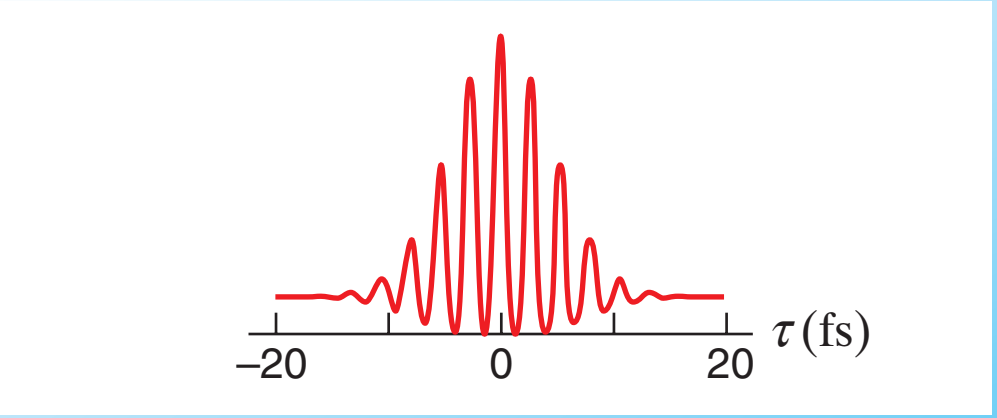
at $\tau = 0$**:**

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



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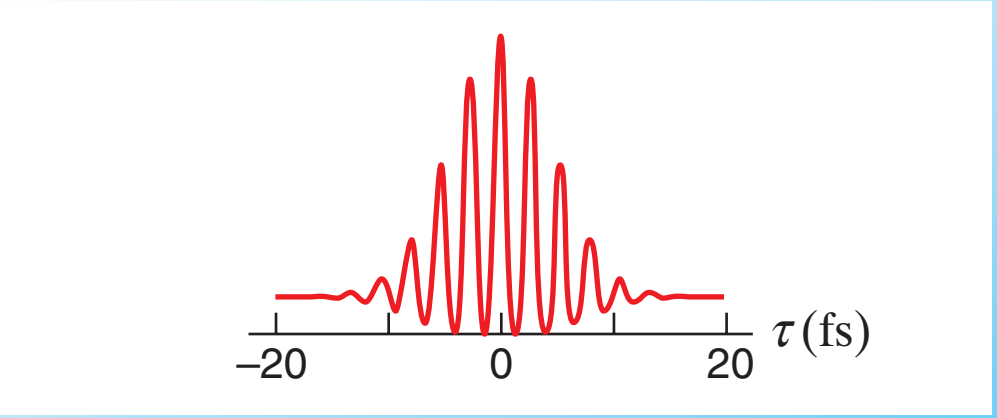
at $\tau = 0$**:** $$I_{2\omega}(t,\tau) \propto 16E^4(t)$$

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



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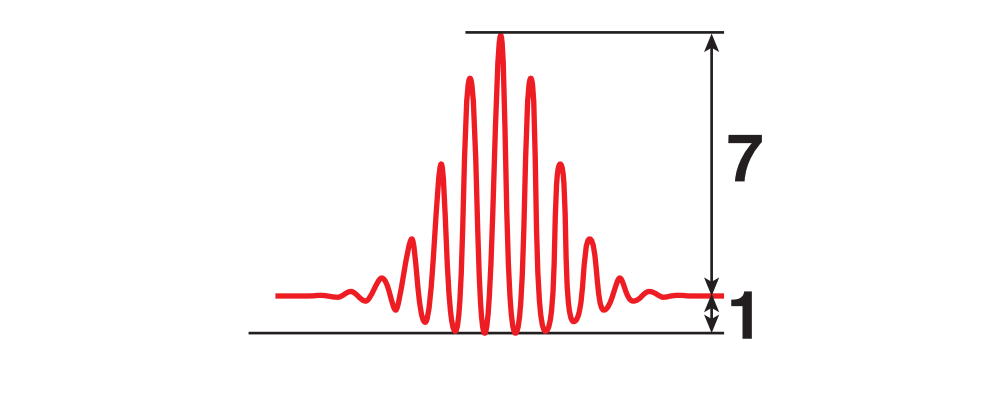
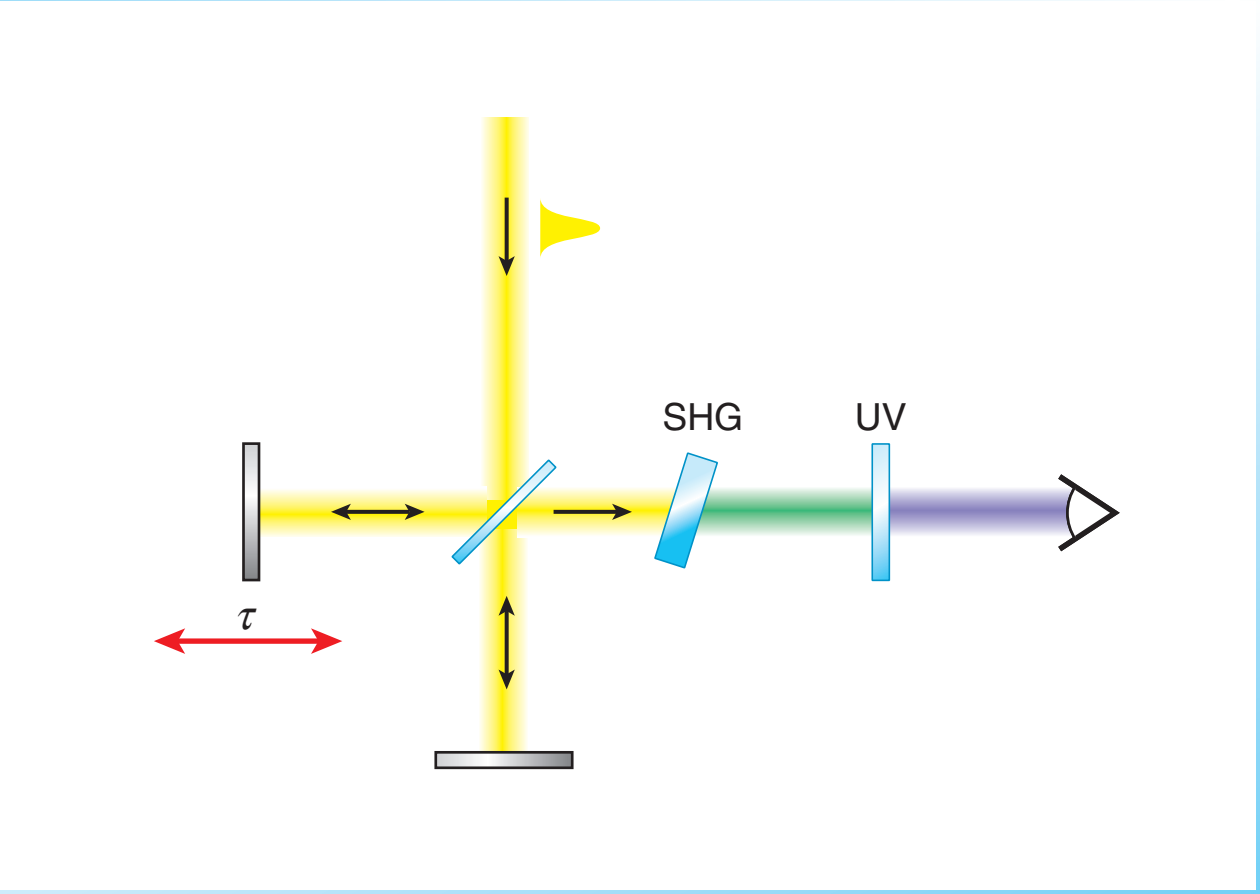


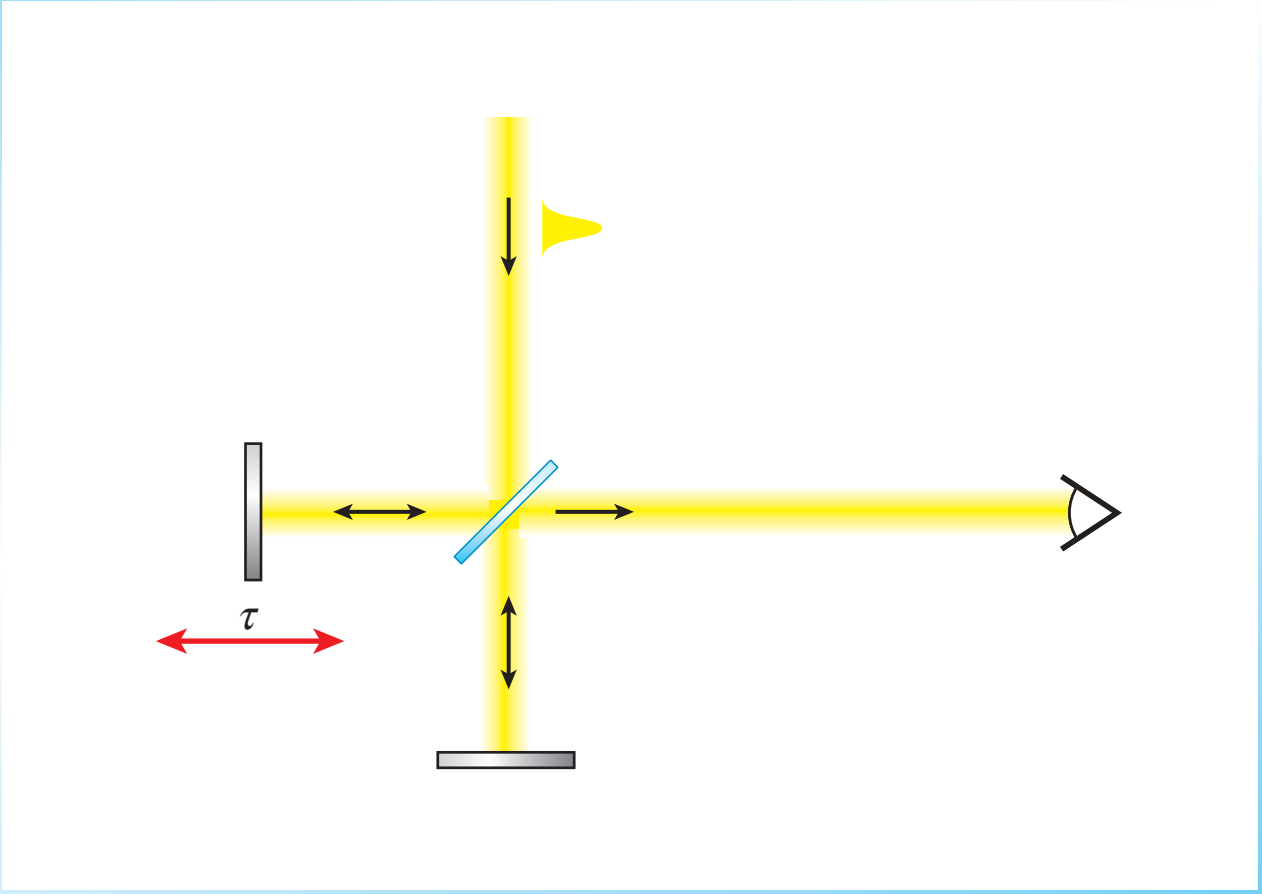
Temporal characterization

Do we really need the second-harmonic crystal…?



Temporal characterization

Would this work?



Temporal characterization

Intensity at detector

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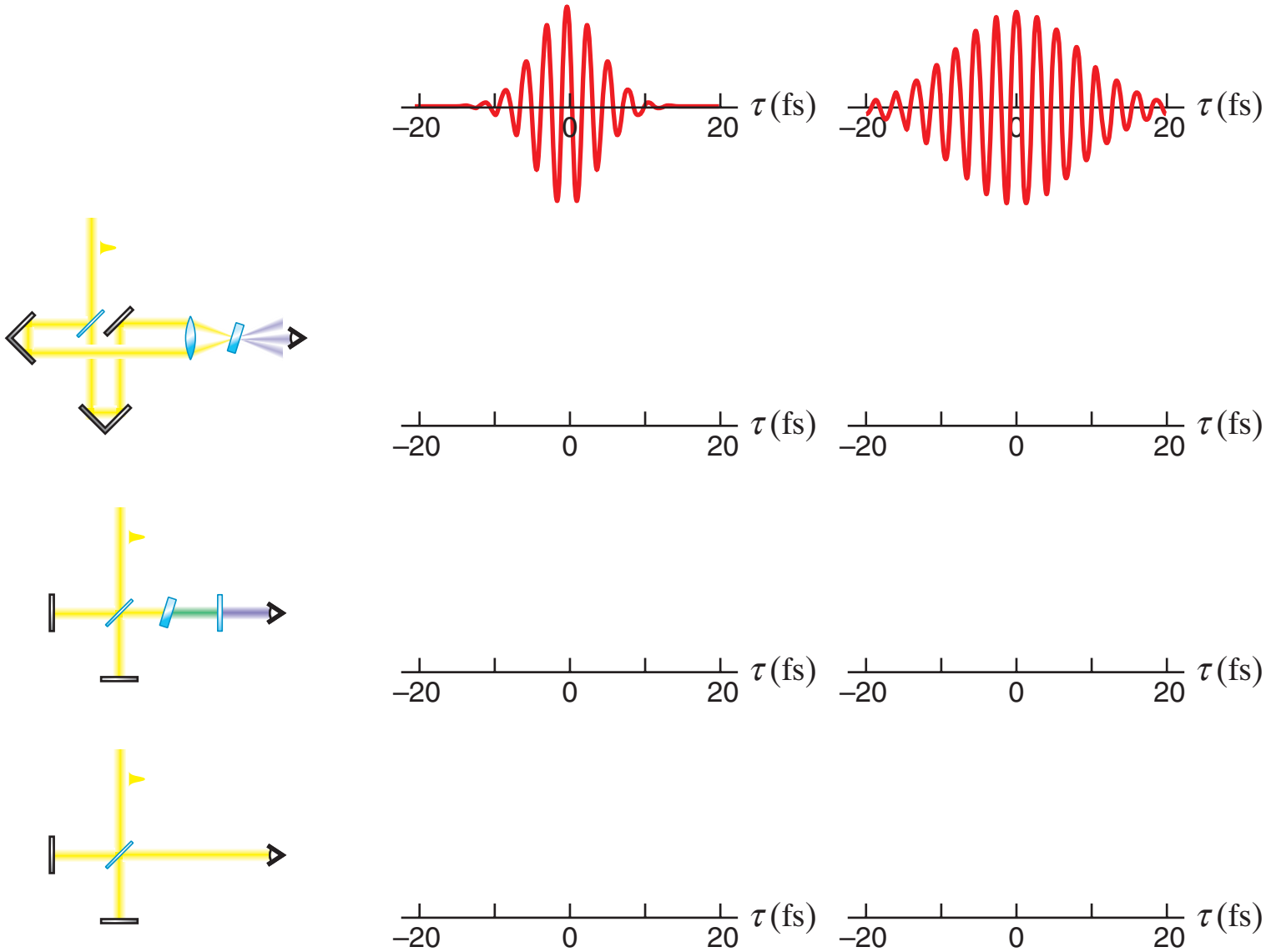
so

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

Temporal characterization



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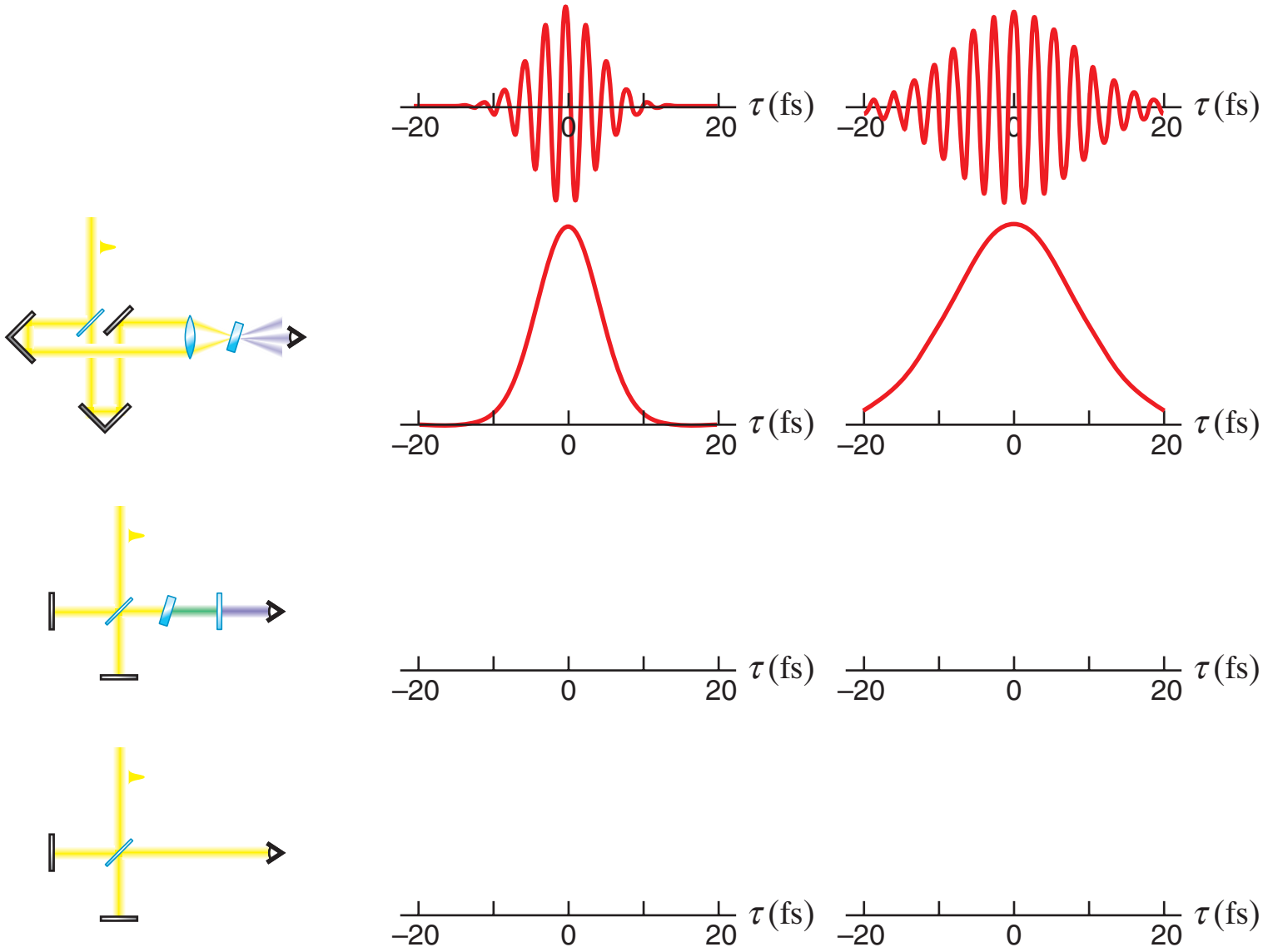

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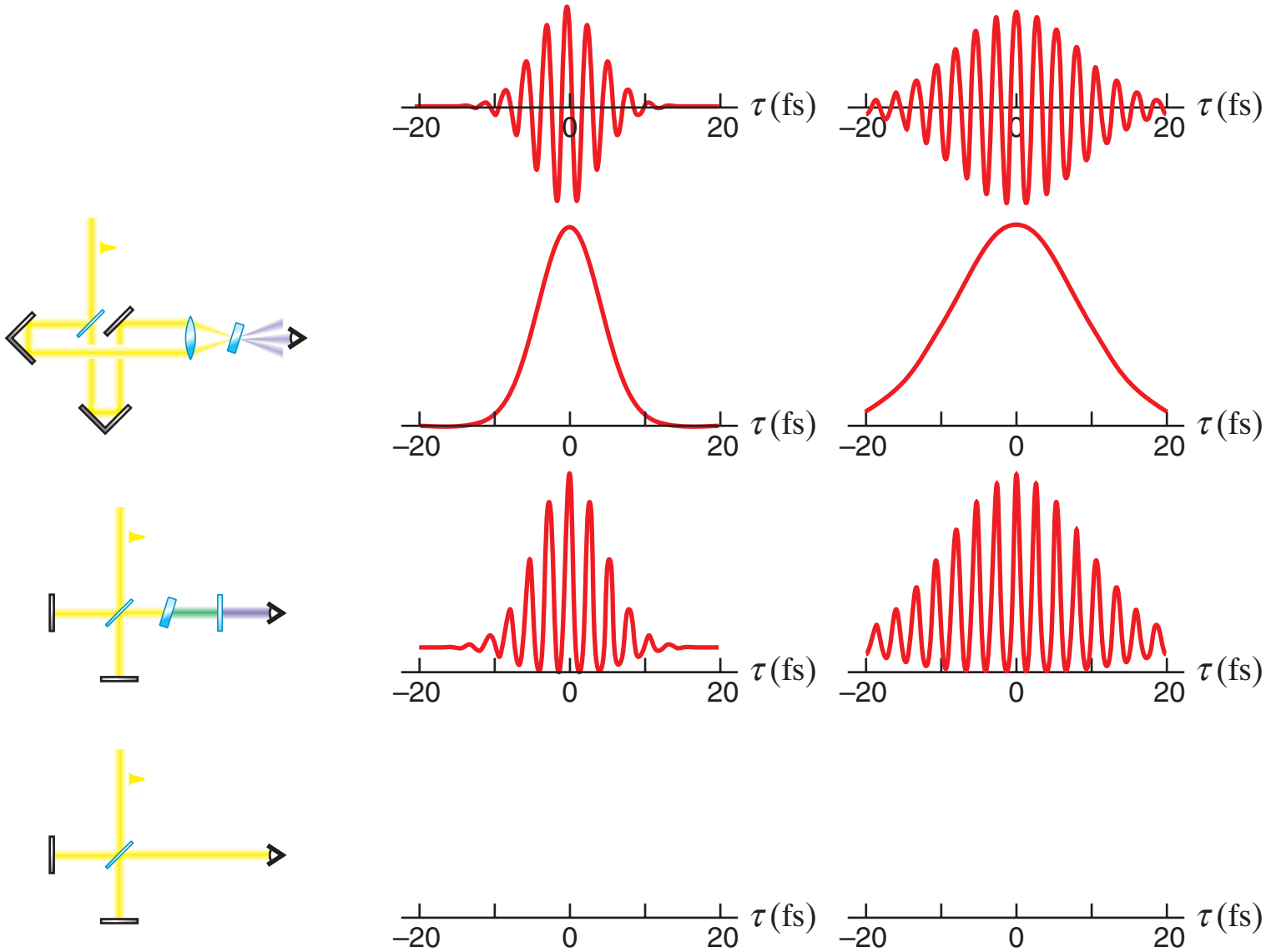

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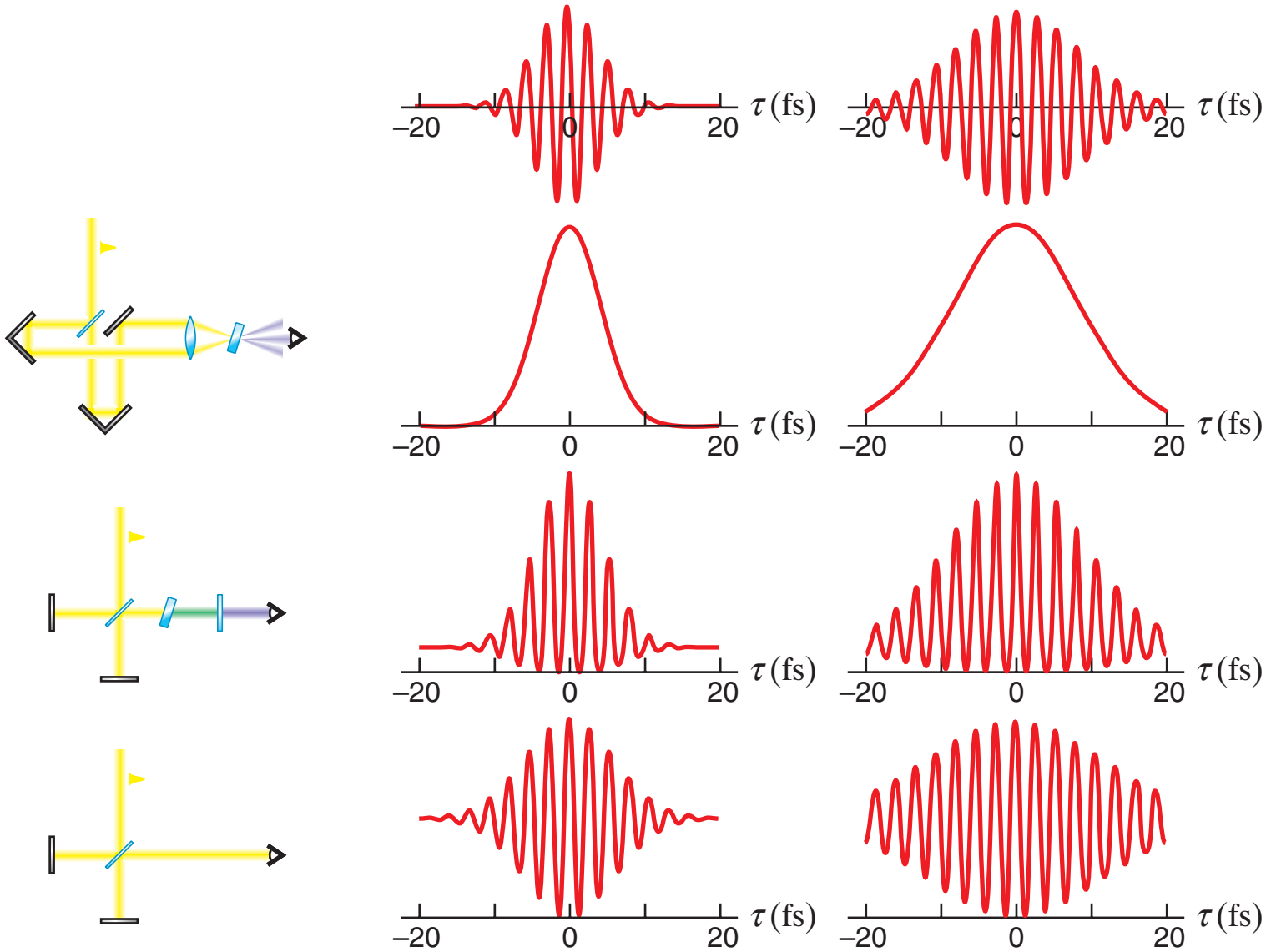
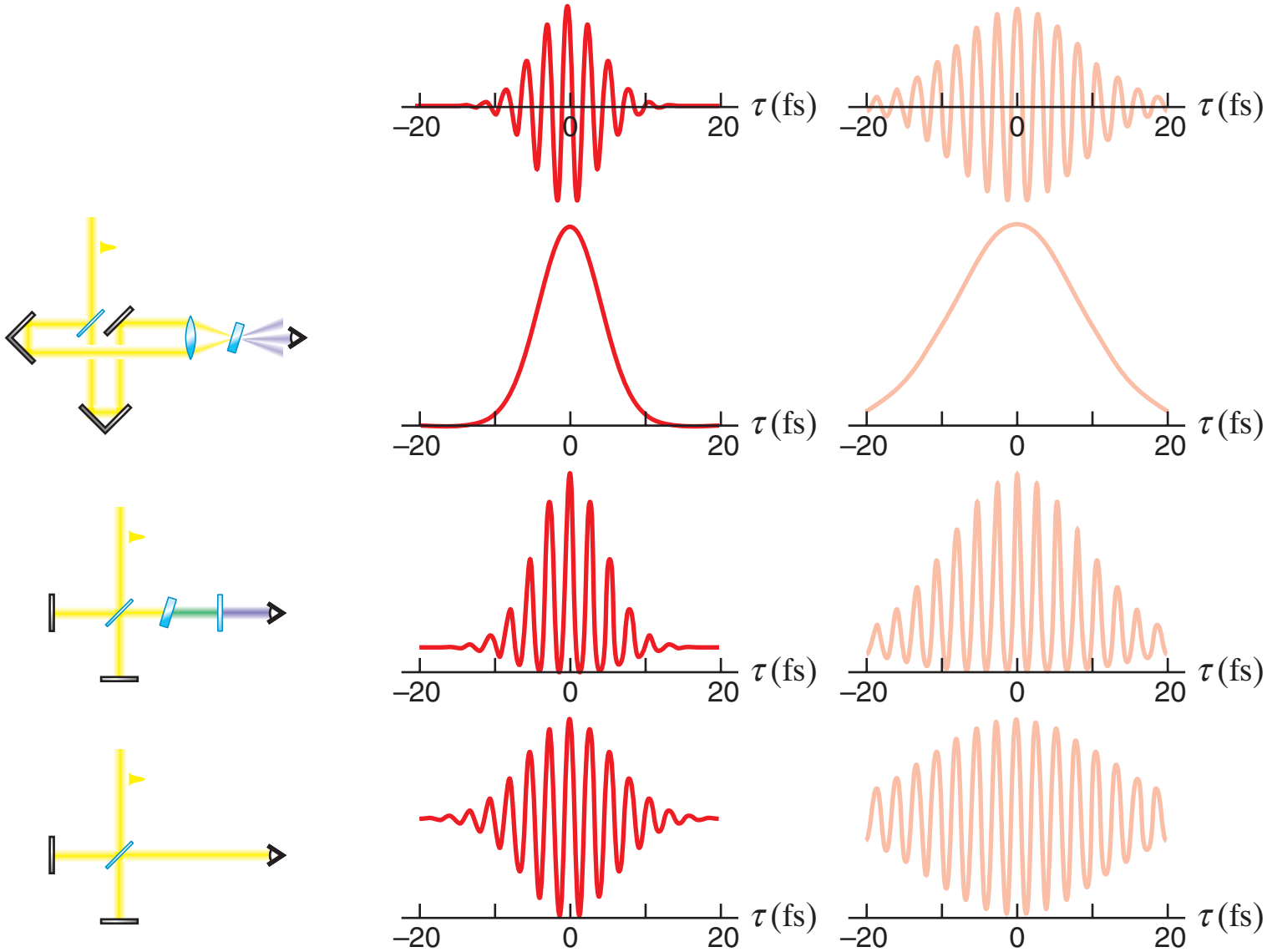


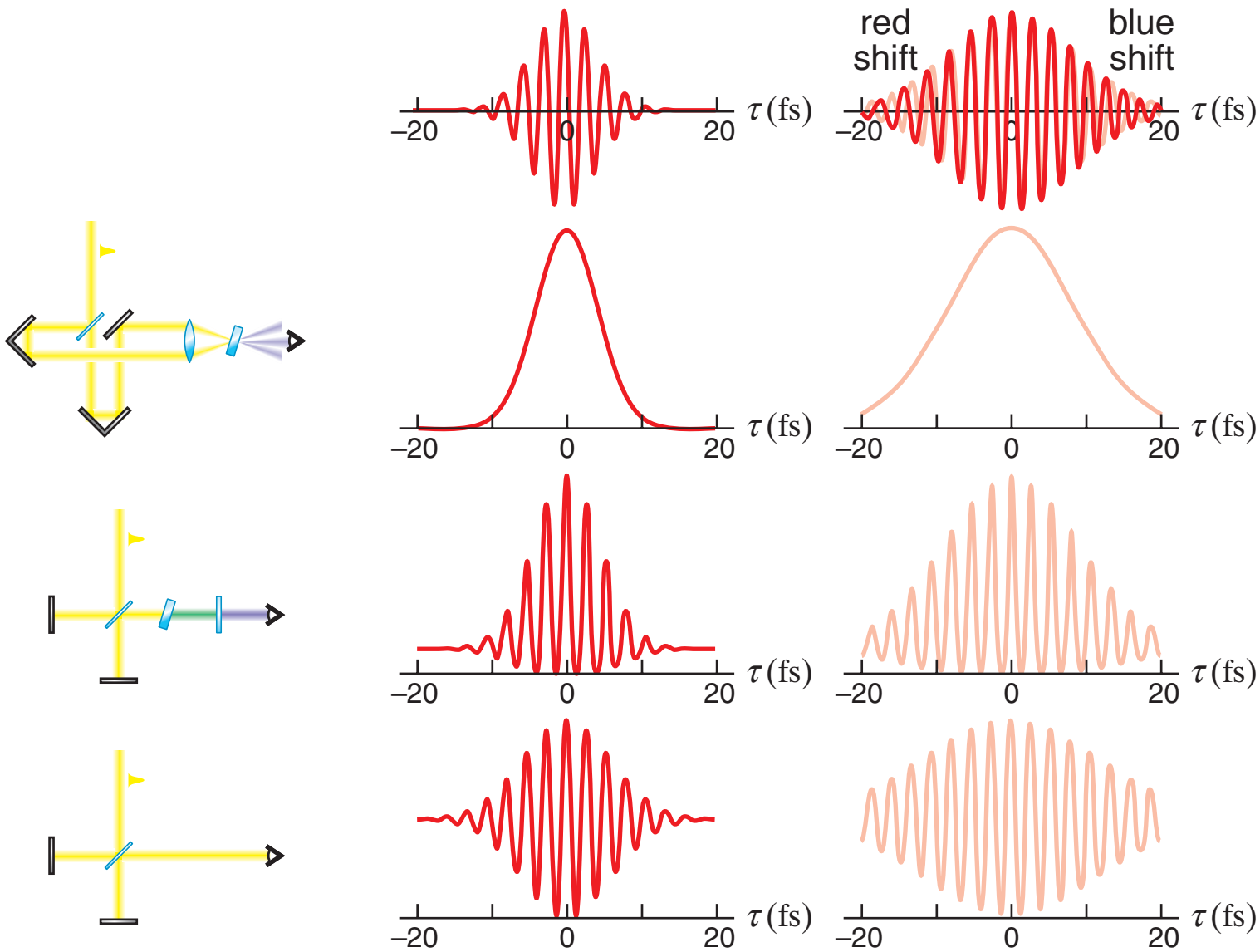
Temporal characterization

But what about dispersion?

Temporal characterization



Temporal characterization



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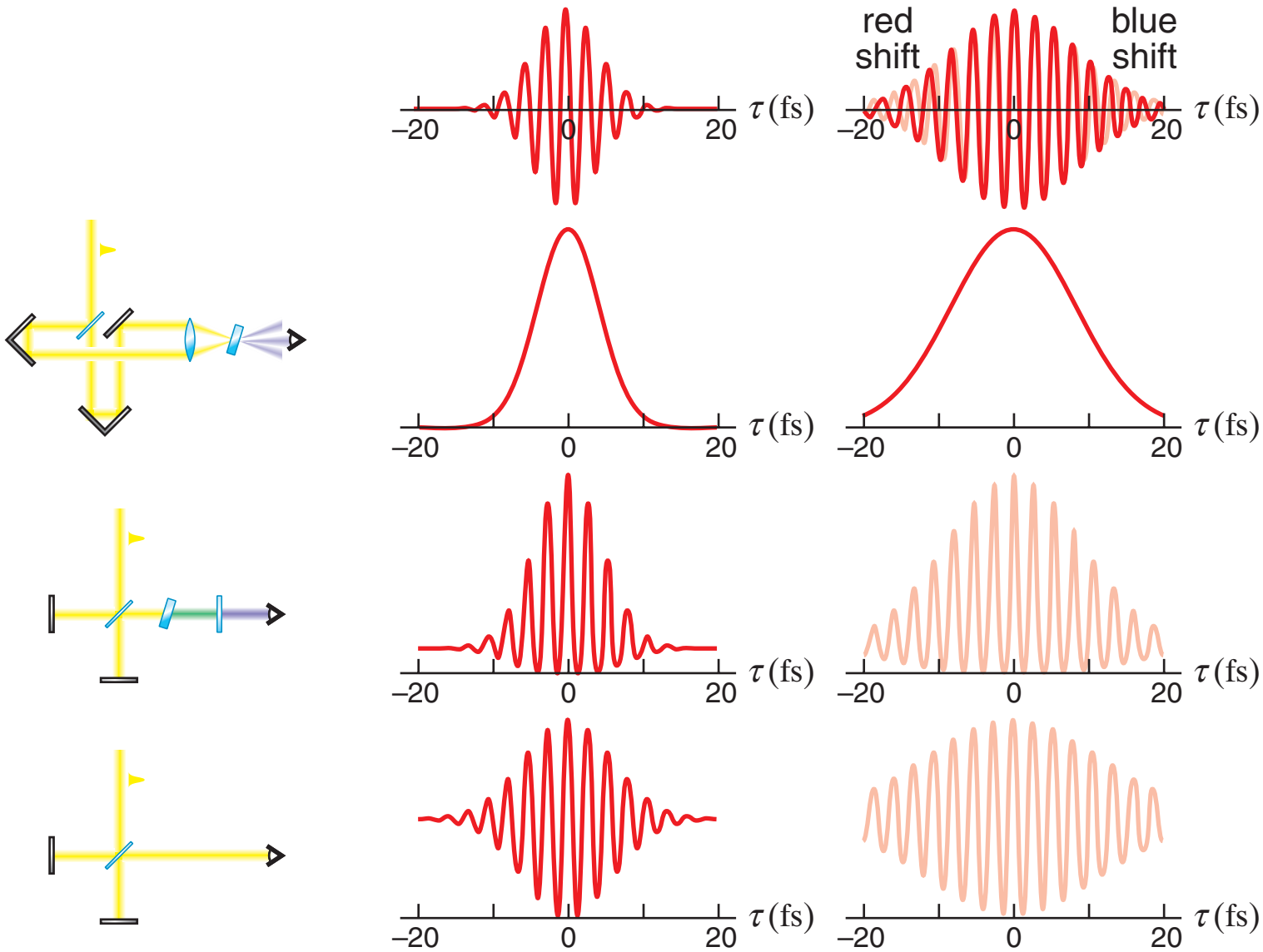

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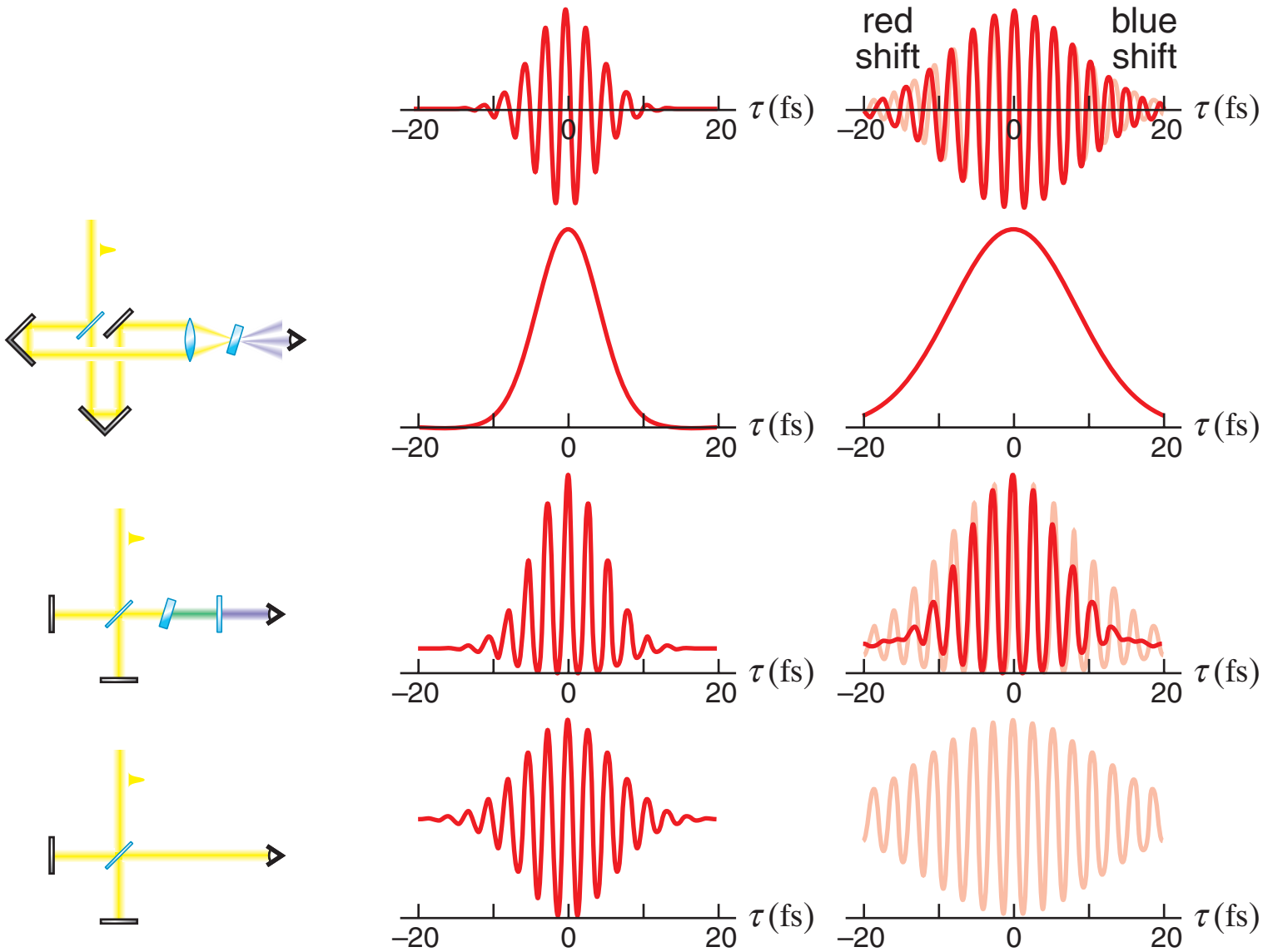

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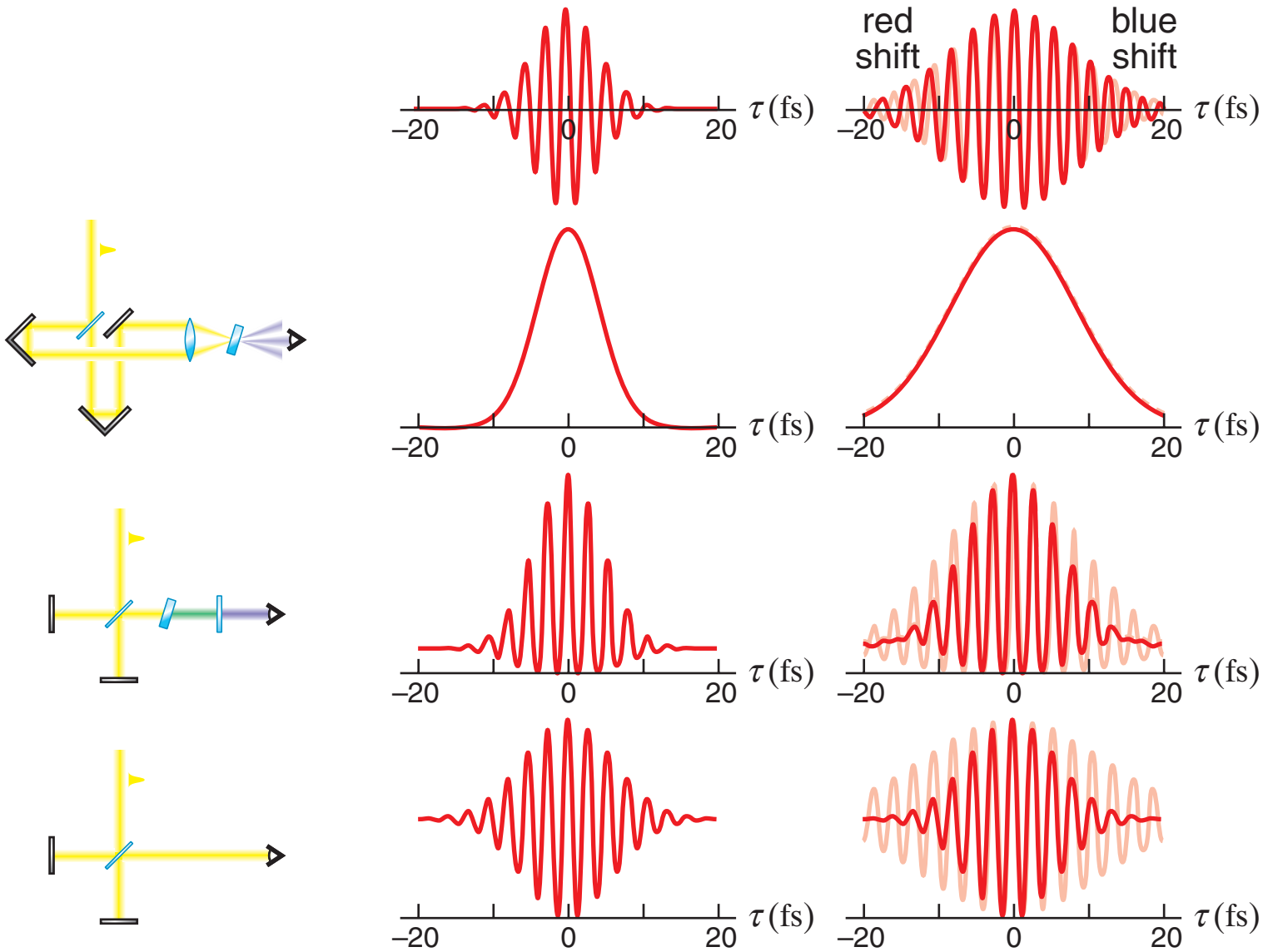

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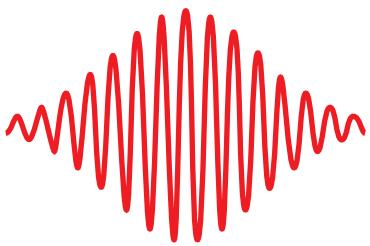

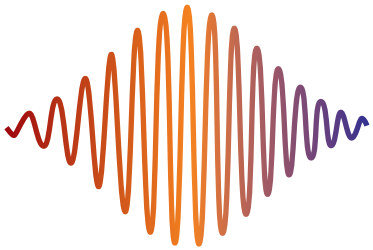

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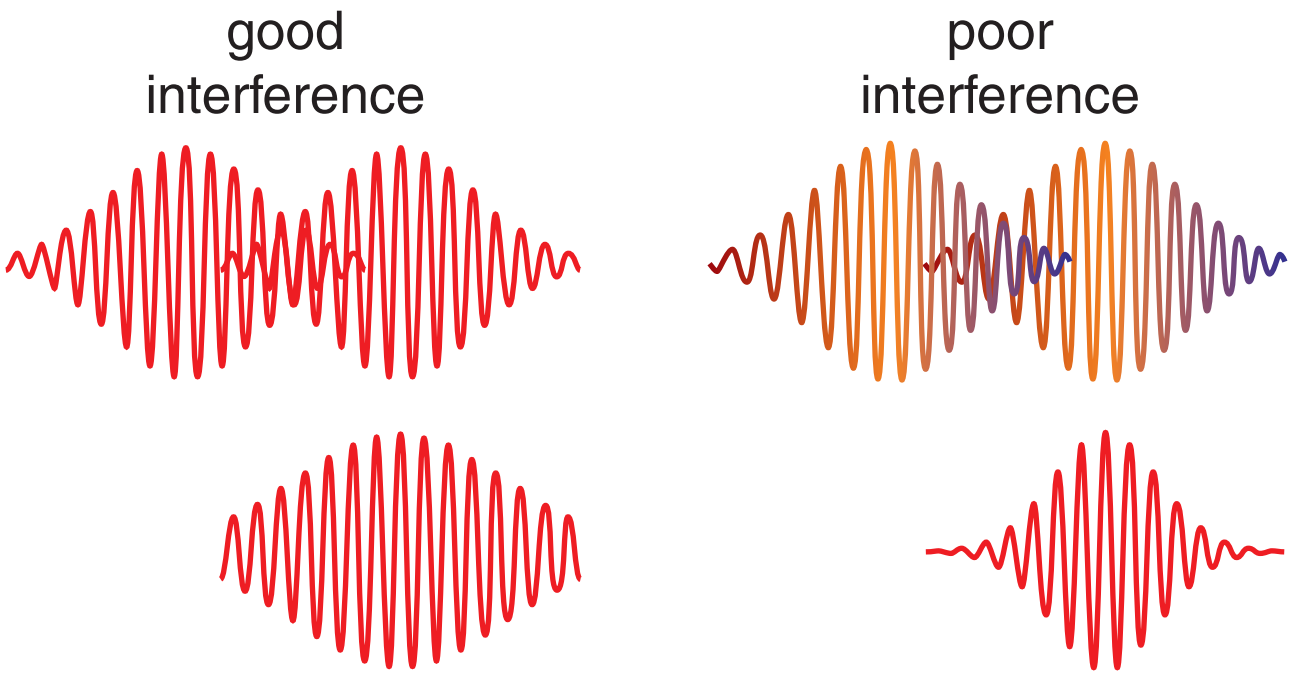

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Let $E_{disp}(\omega) = E_{orig}(\omega)e^{-i\phi(\omega)}$.

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Interference term in linear autocorrelation:

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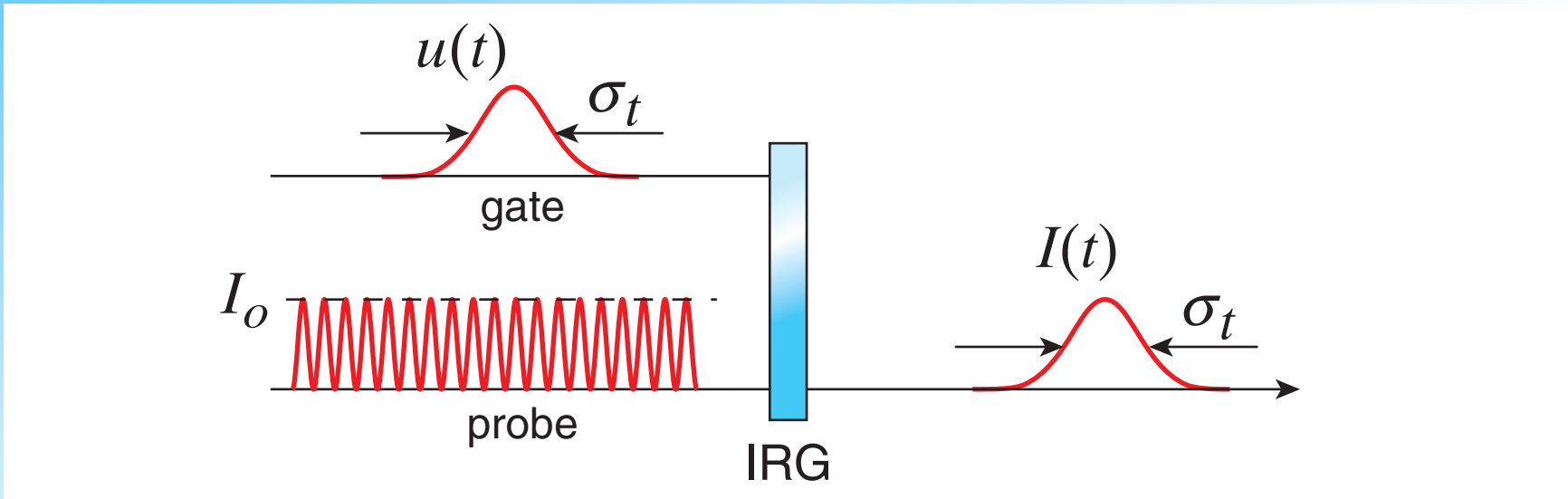
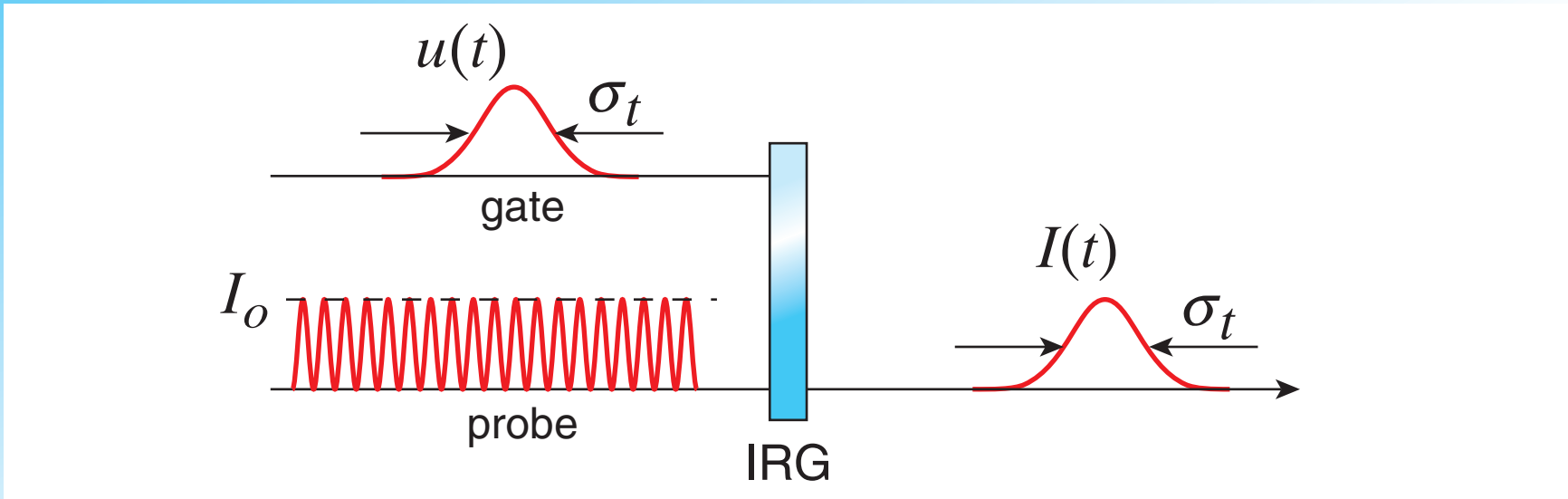
Joint time-frequency measurements



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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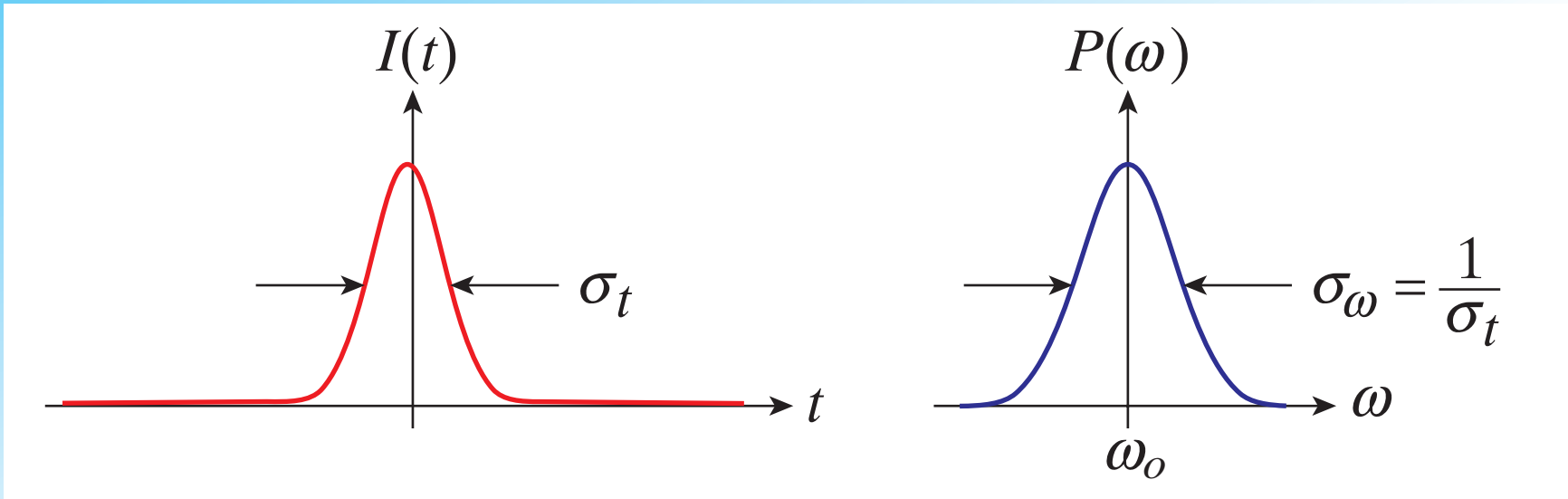

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$$\sigma_t \sigma_\omega = 1$$

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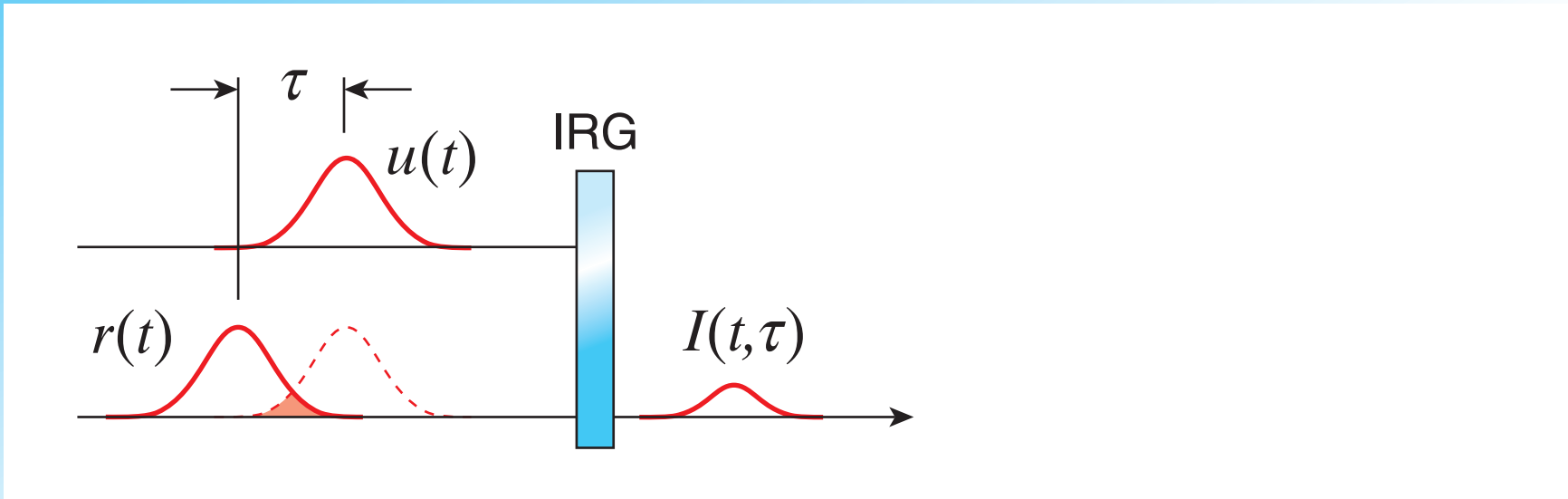
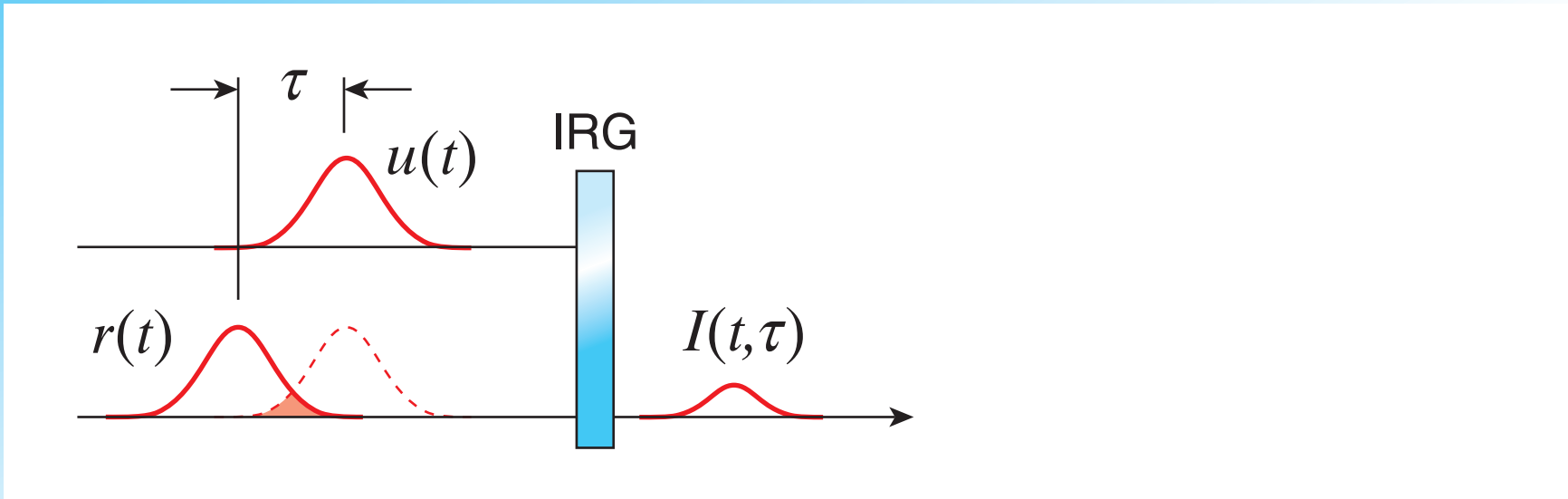


Transmitted intensity

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

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

Joint time-frequency measurements

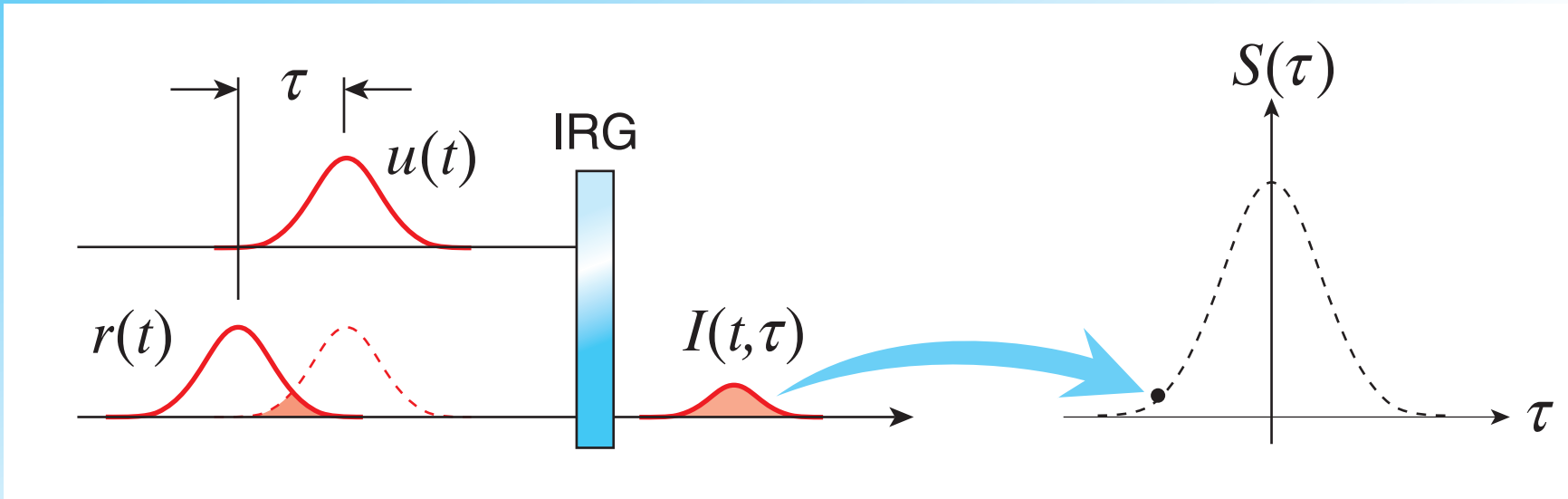


Transmitted intensity

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

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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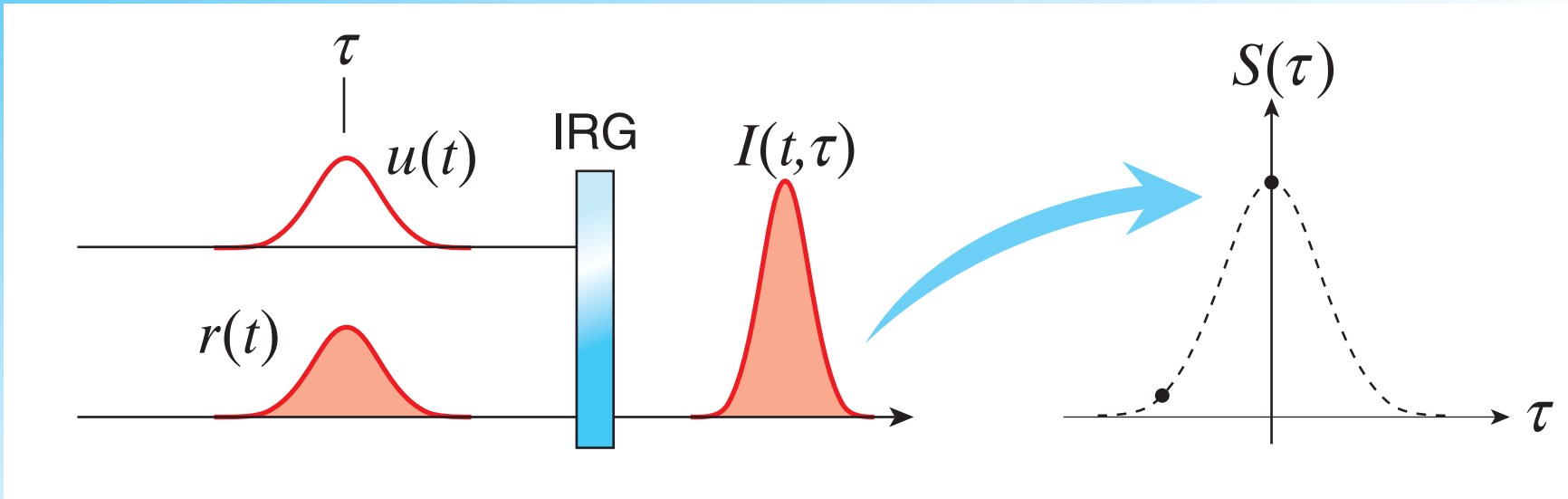

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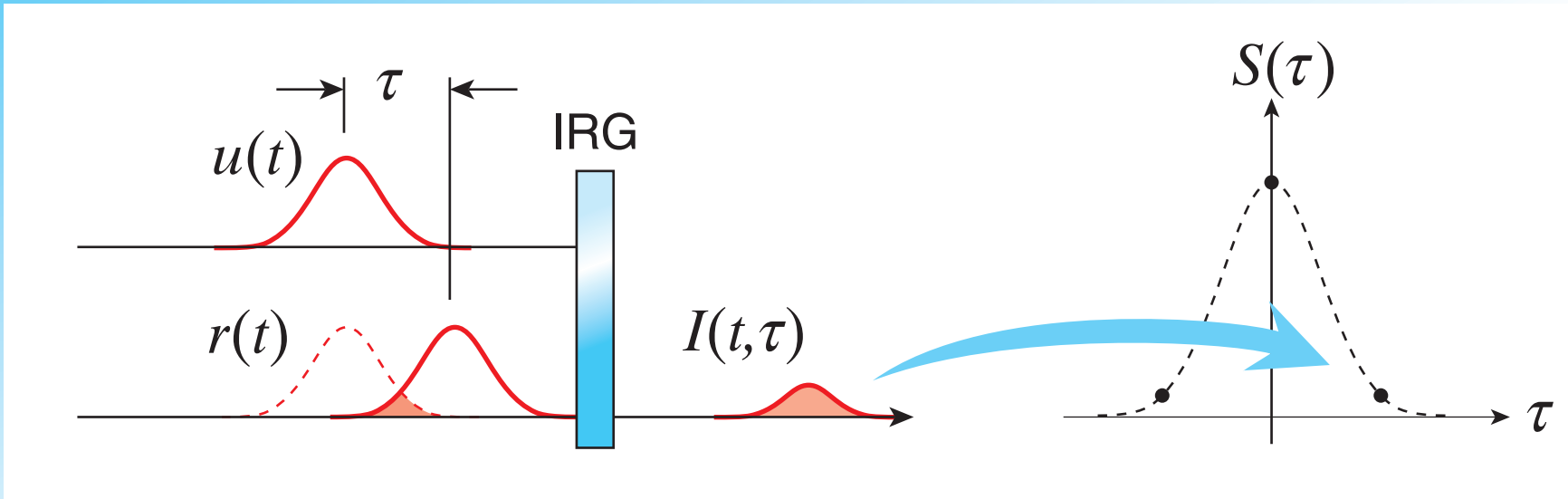

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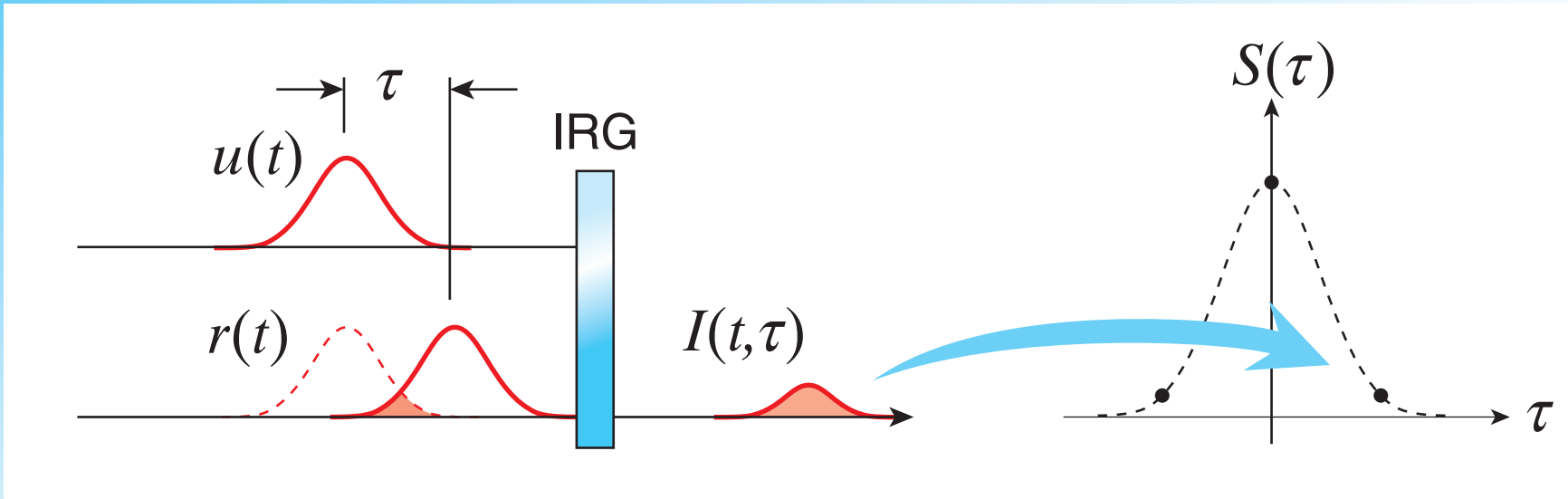

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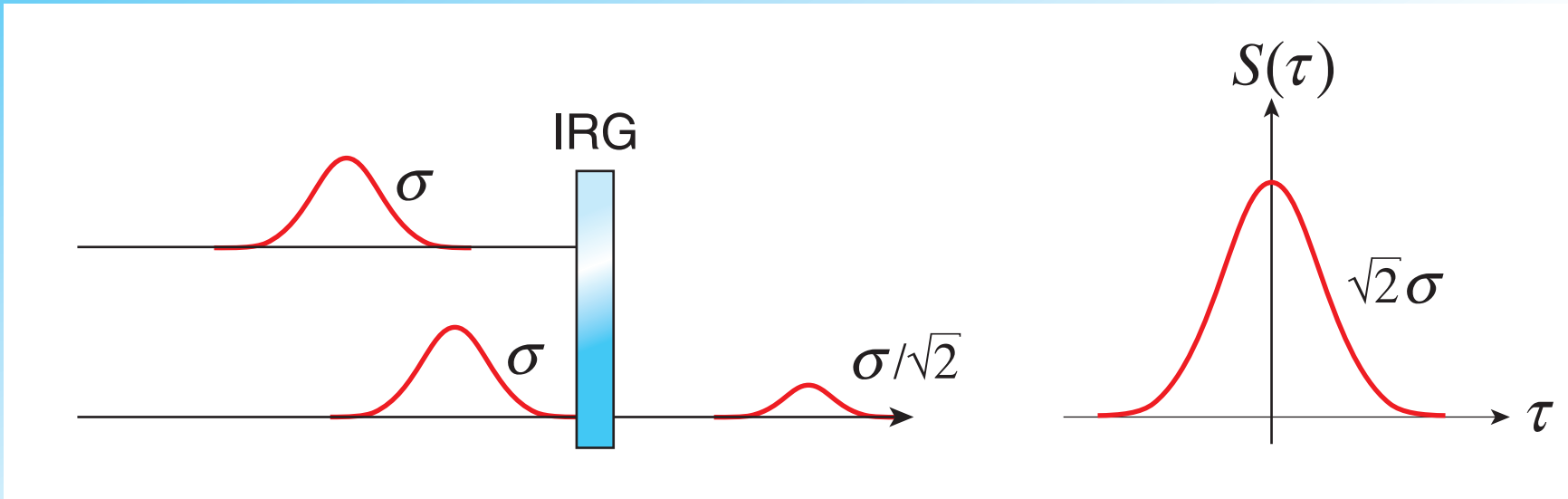
$$= \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \int_{-\infty}^{\infty} \exp\left[-\frac{2u^2}{\sigma^2}\right] du = \sqrt{\frac{\pi}{2}}\,\sigma \exp\left[-\frac{\tau^2}{(\sigma\sqrt{2})^2}\right]$$

Joint time-frequency measurements



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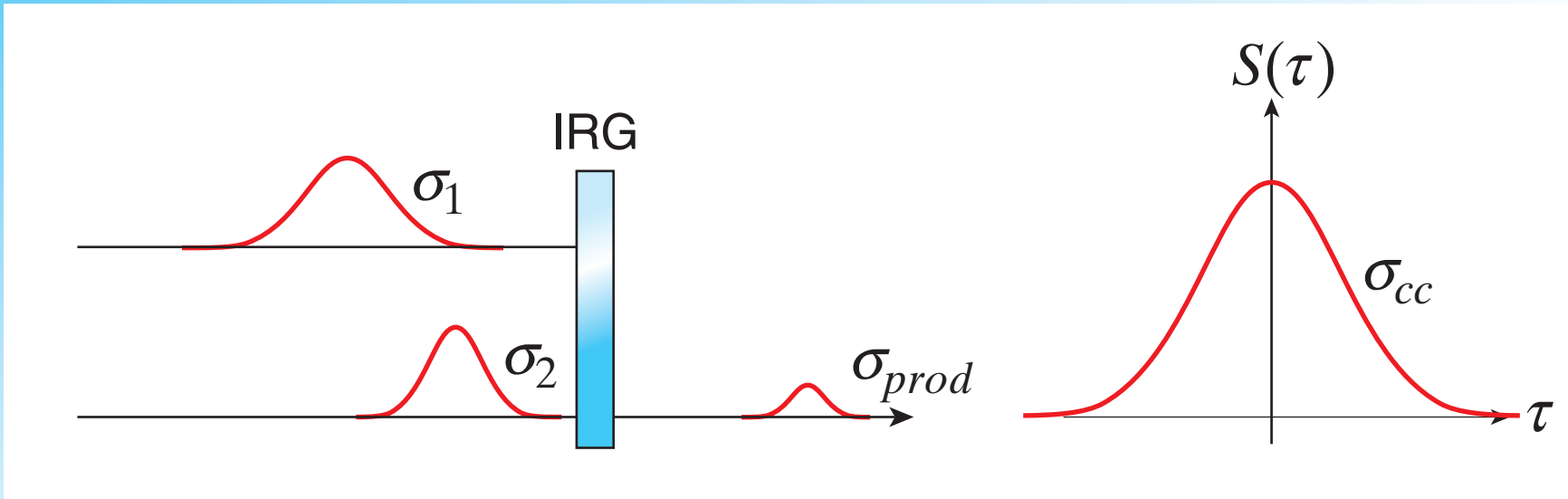
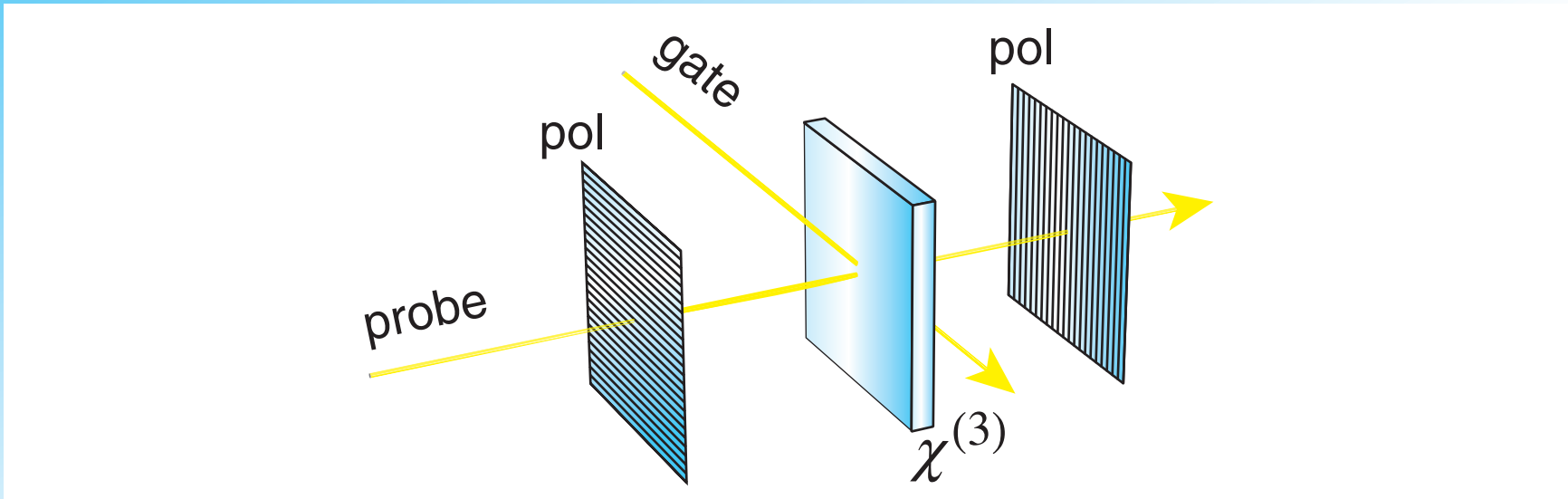


If gate and probe unequal:

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

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

Joint time-frequency measurements

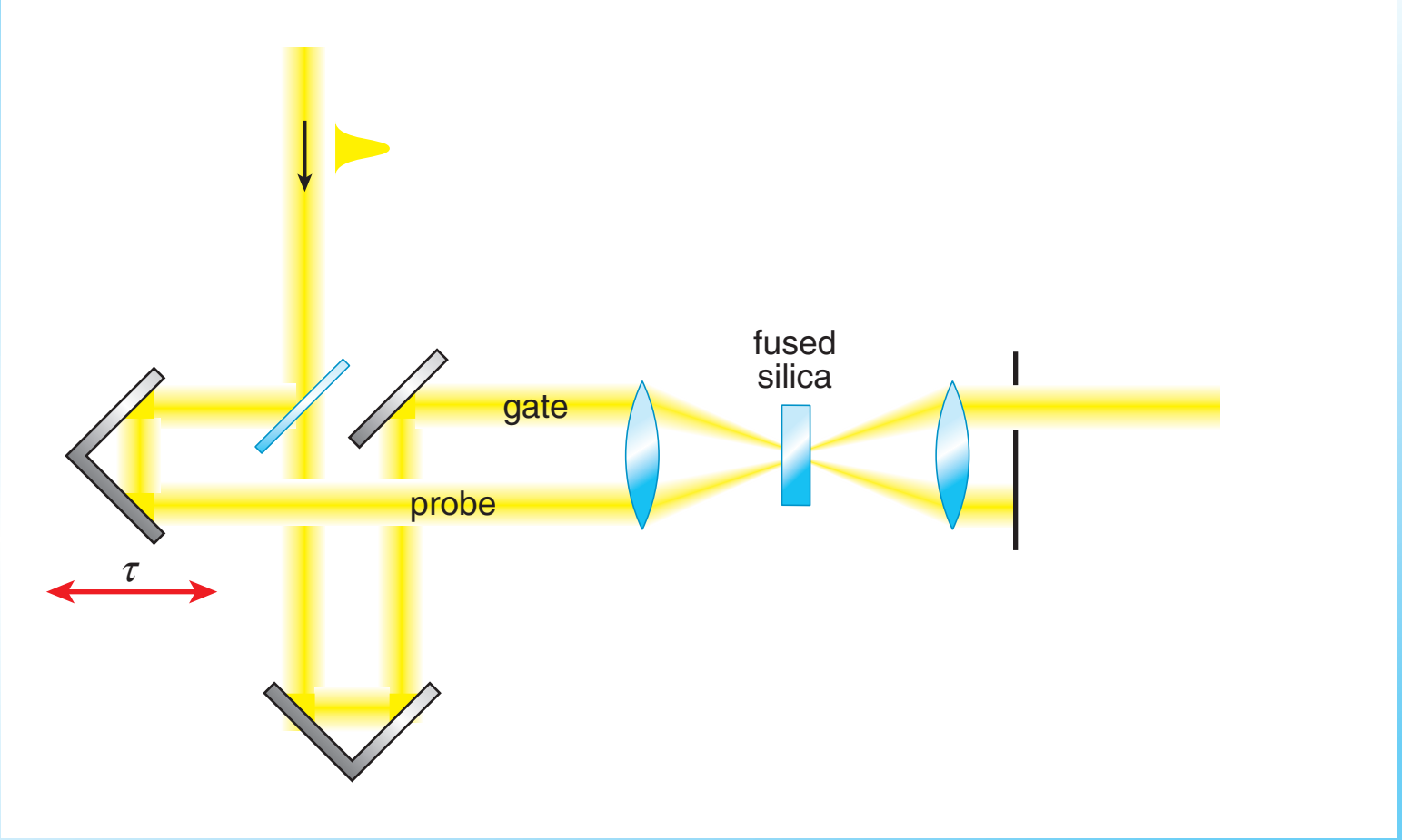


Transmitted field:

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

Joint time-frequency measurements

FROG: frequency-resolved optical gating



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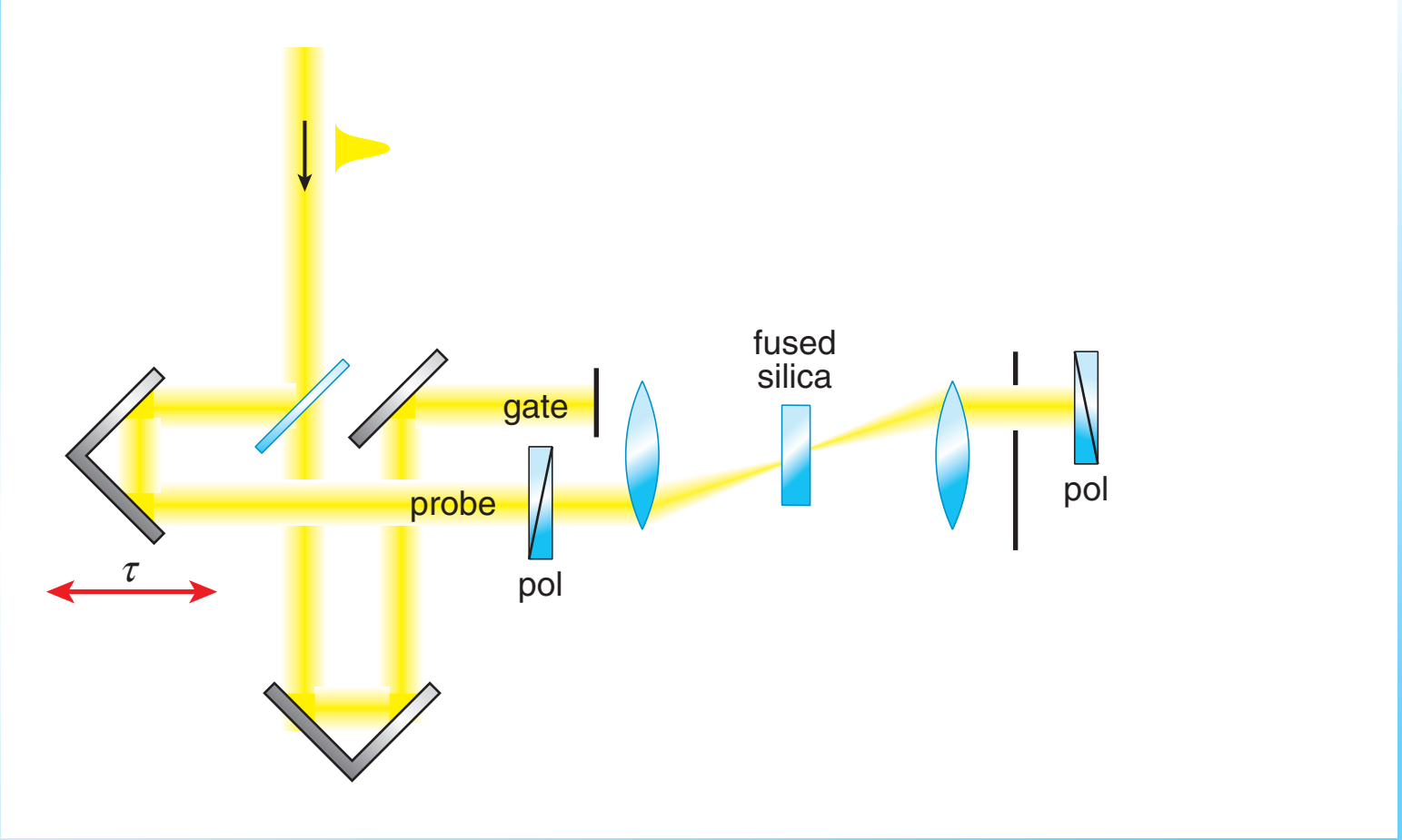

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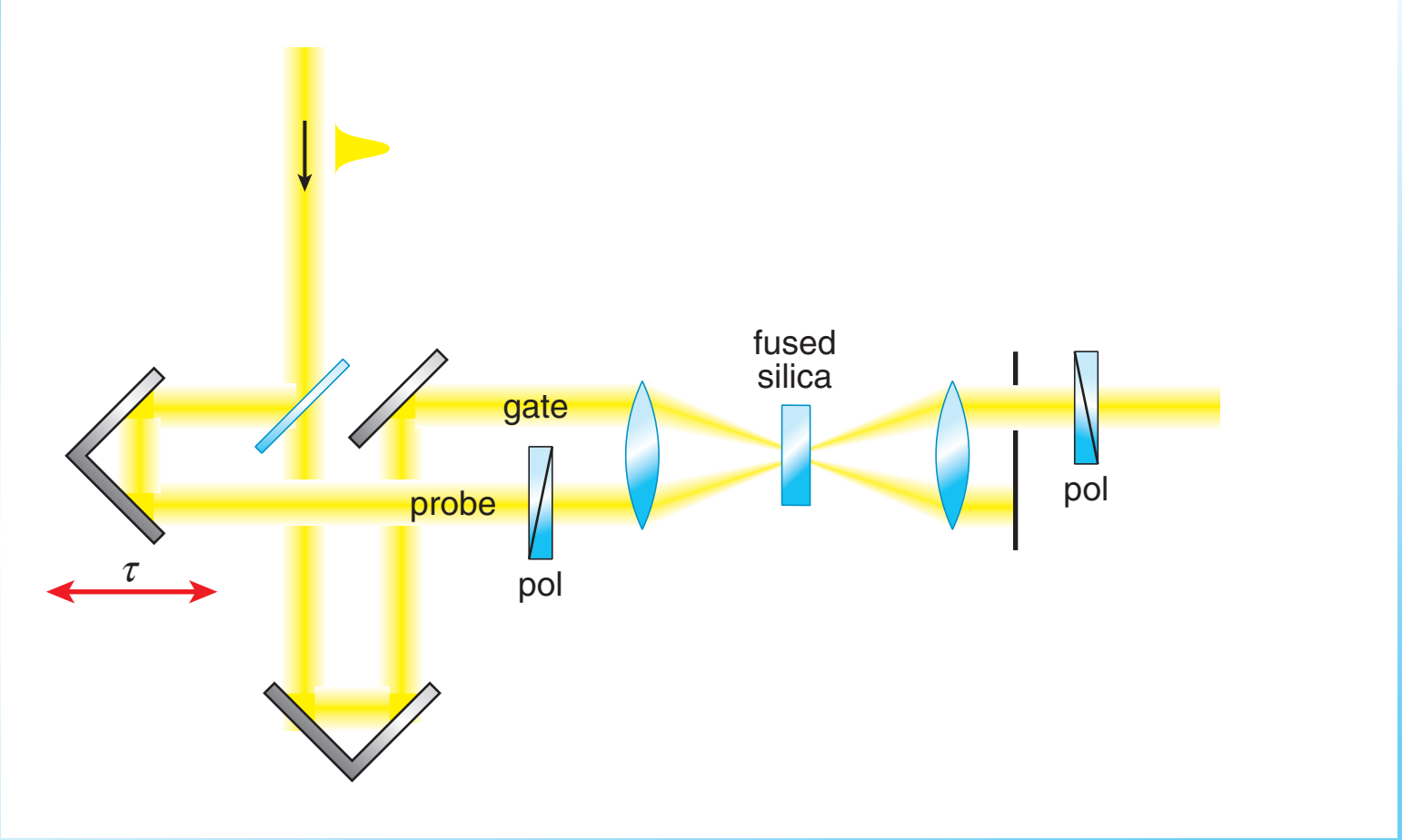

Joint time-frequency measurements

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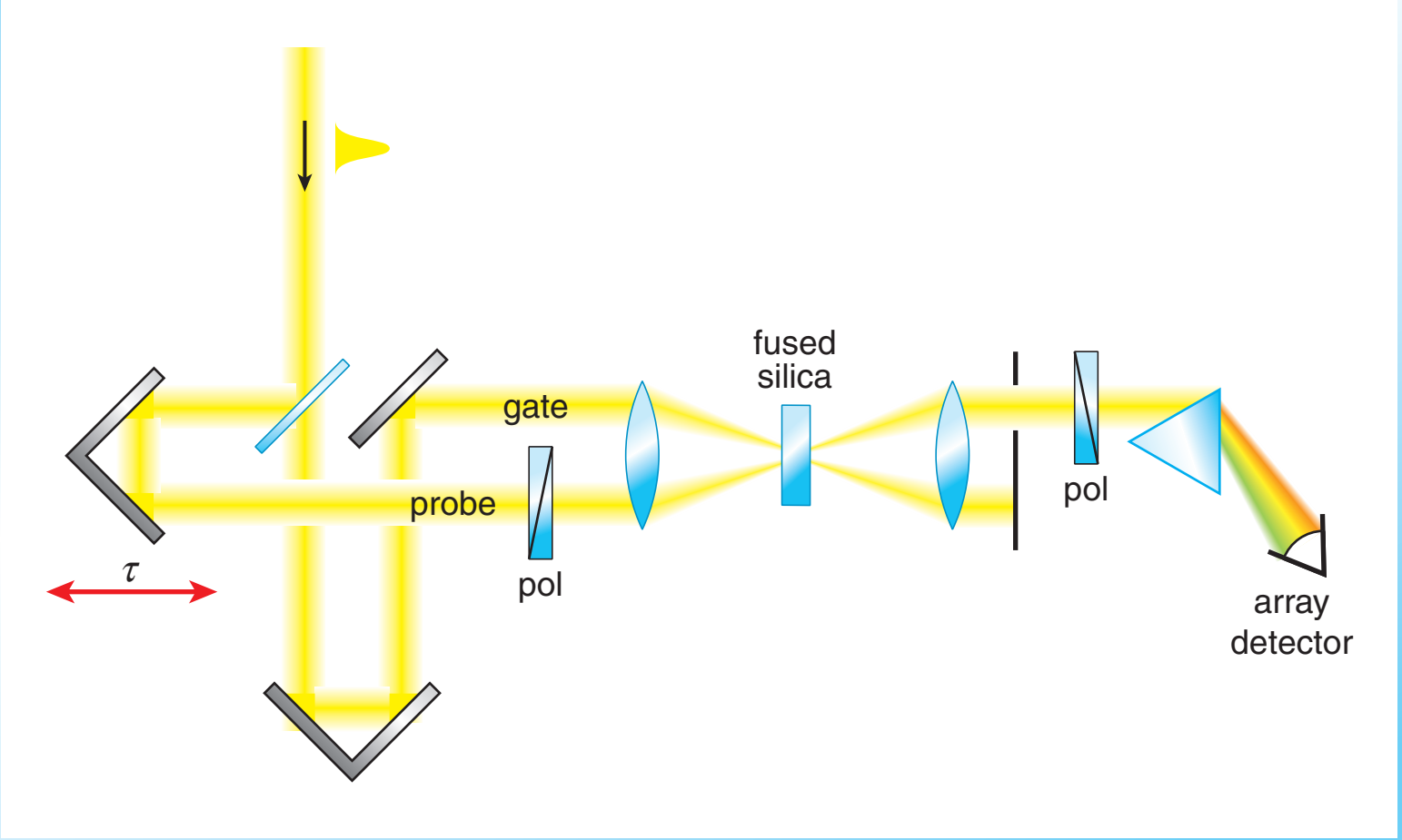
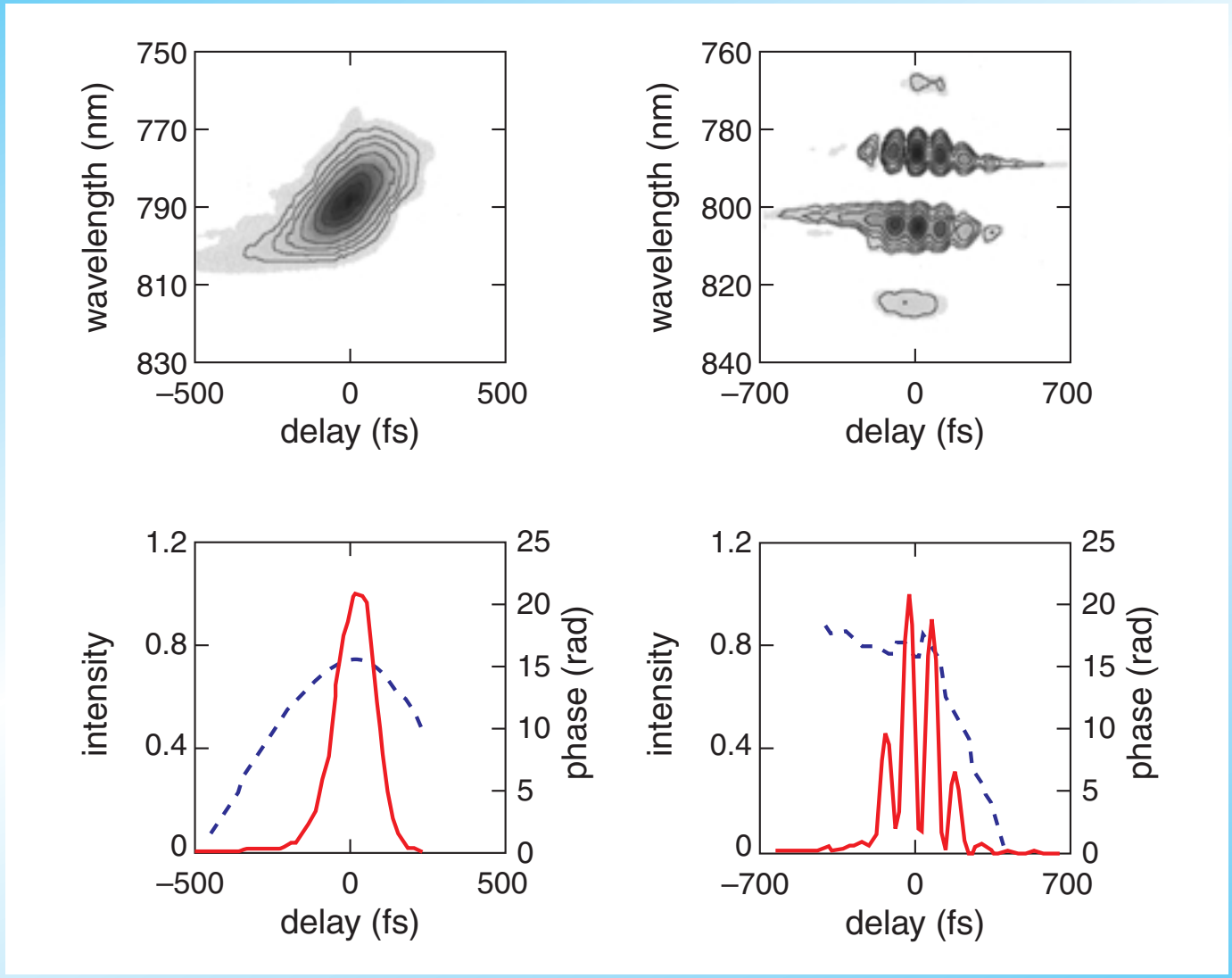
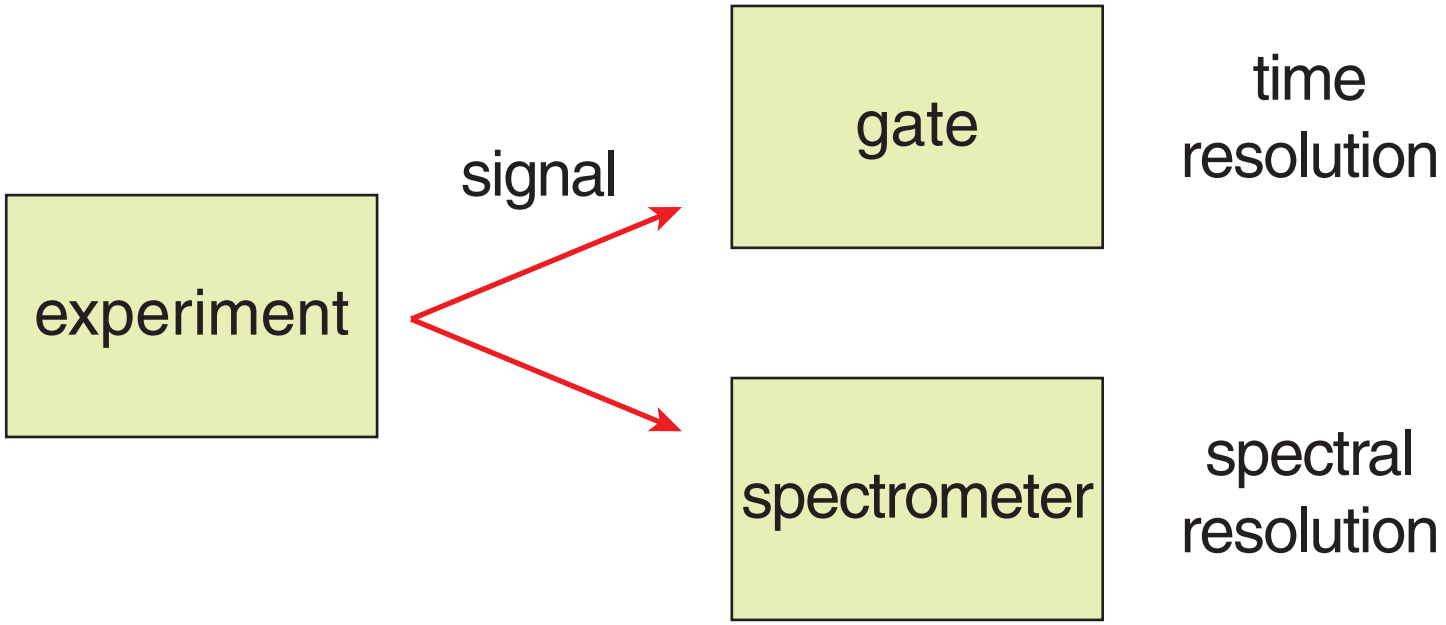


Joint time-frequency measurements



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

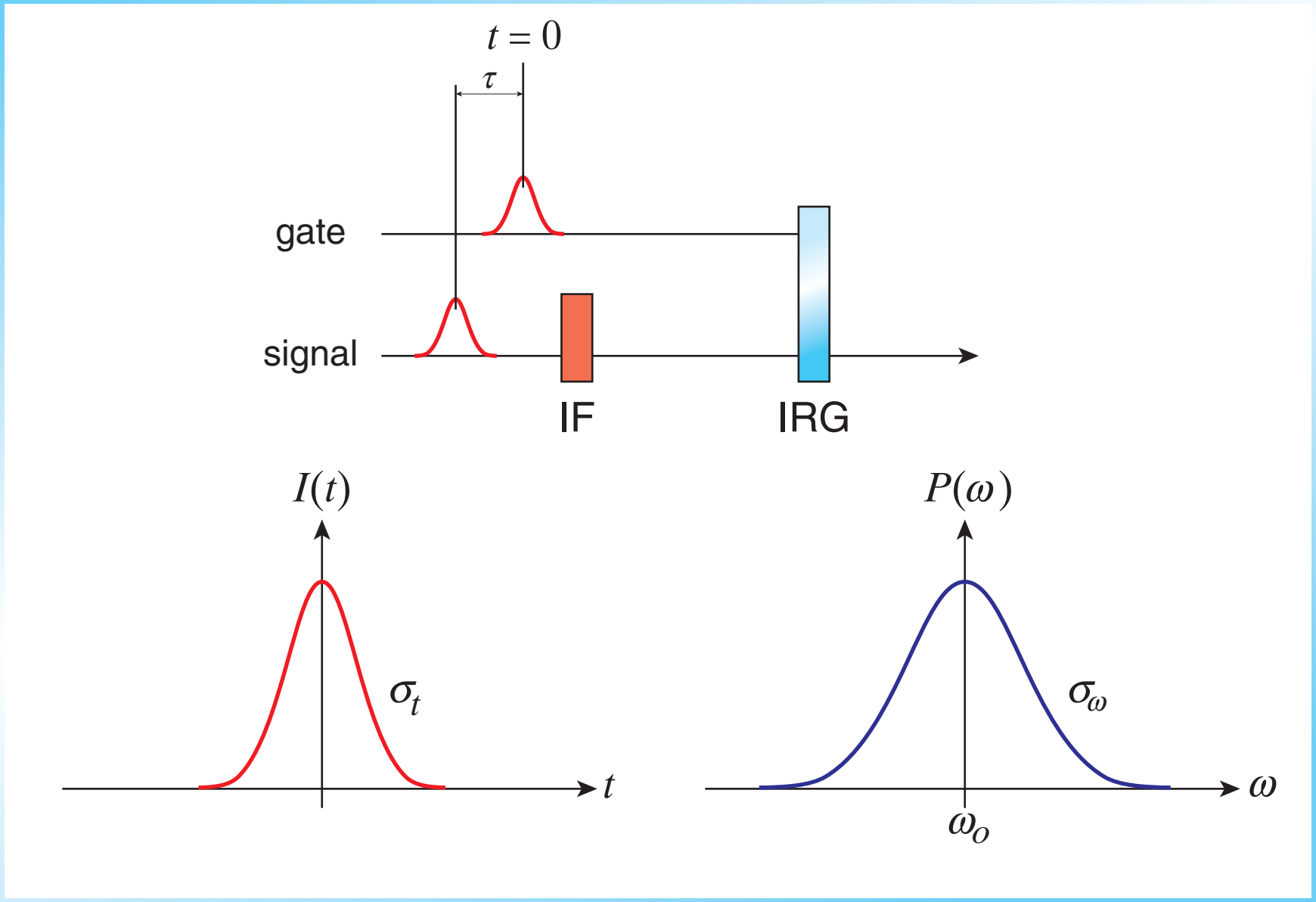
Joint time-frequency measurements



Joint time-frequency measurements

What are the resolution limits?

Experiment 1



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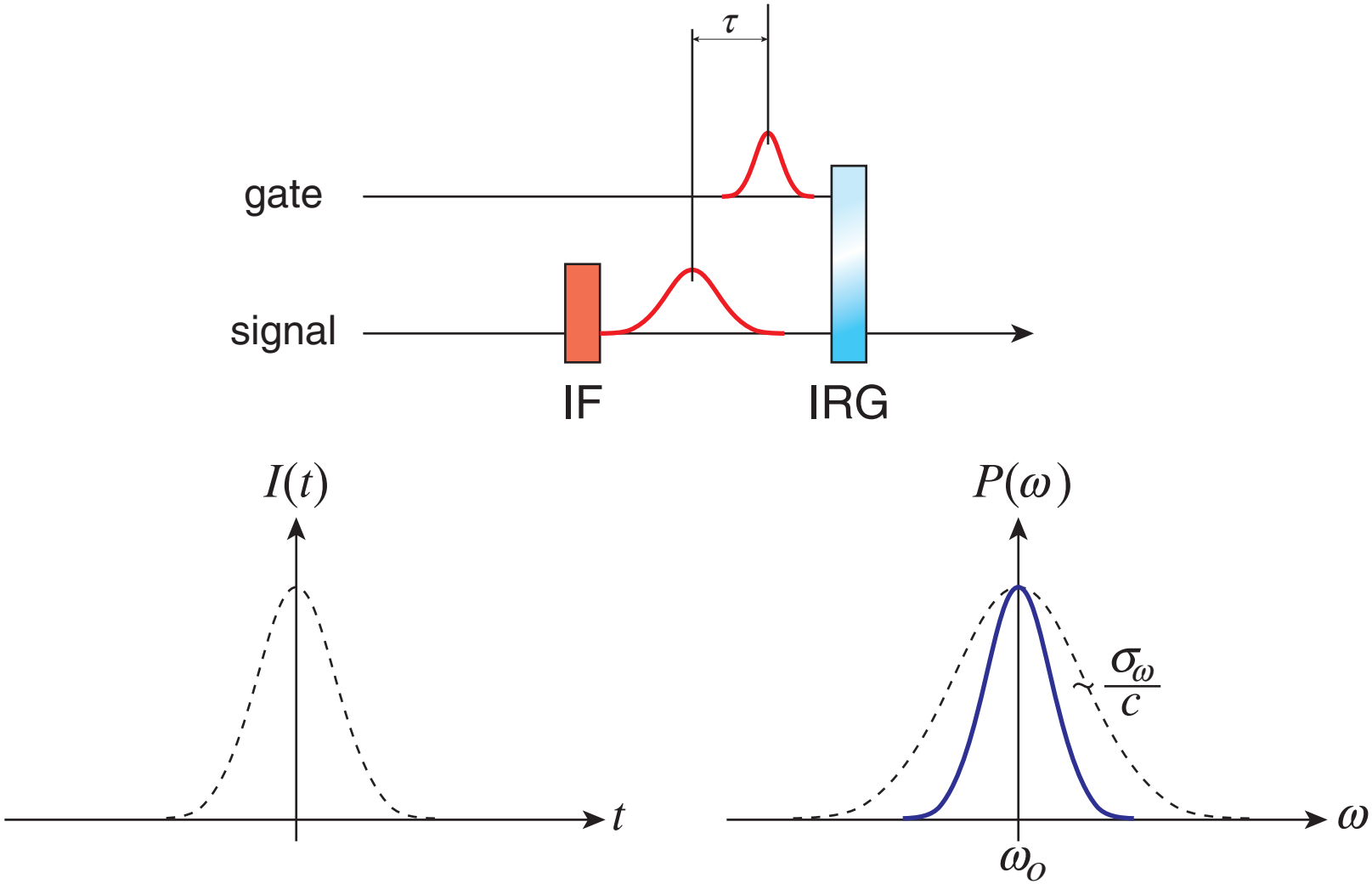

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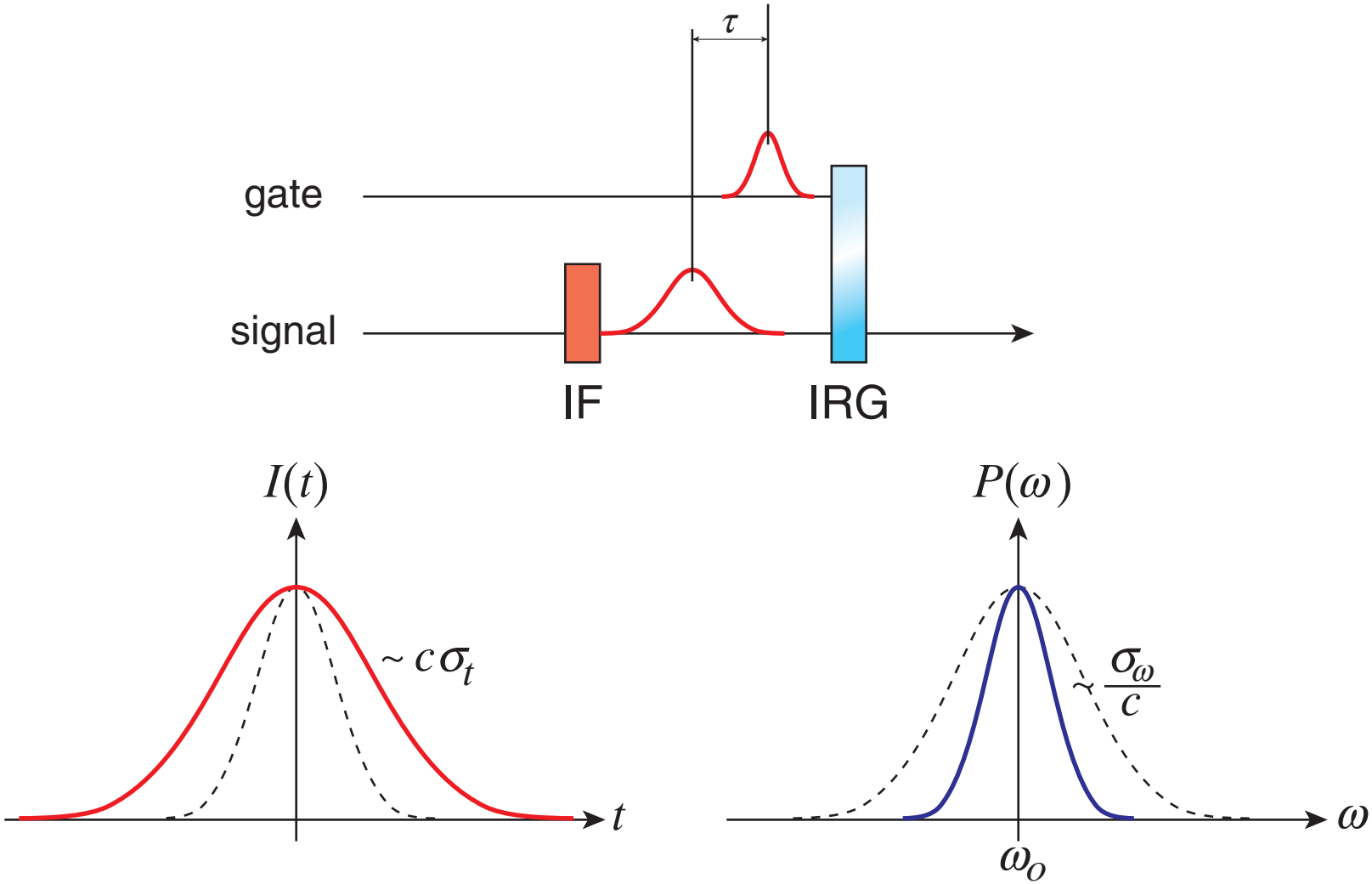

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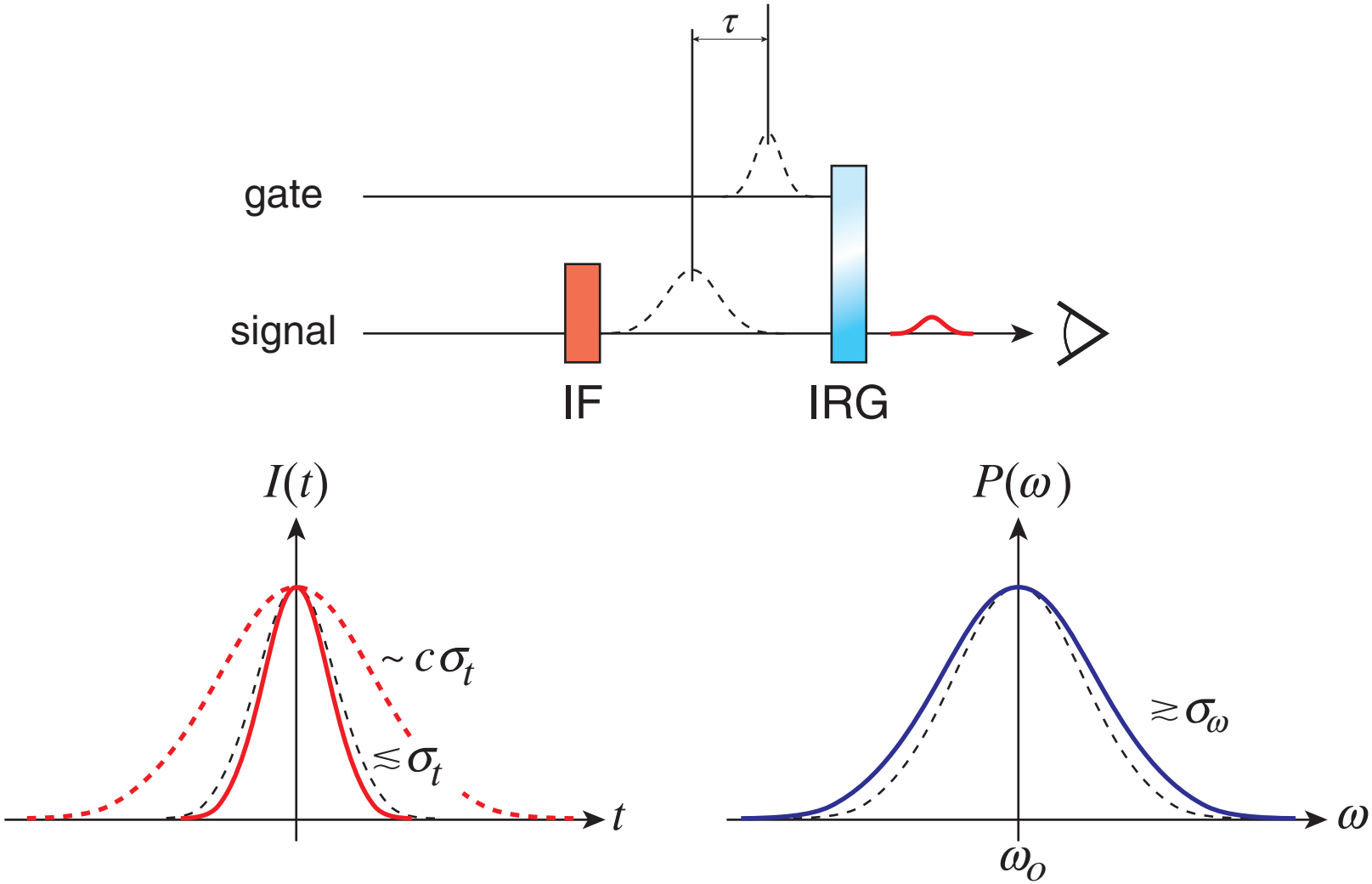
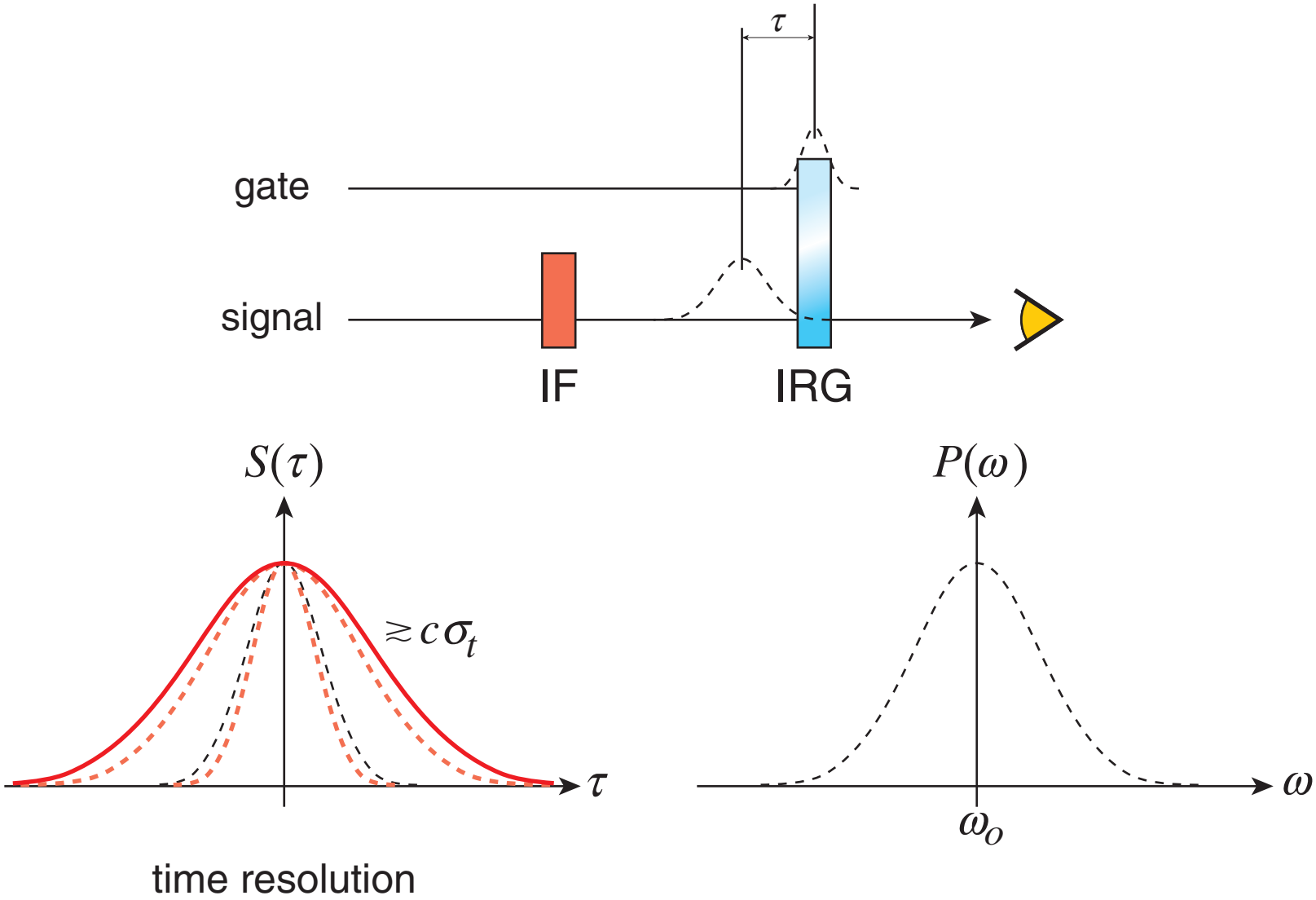
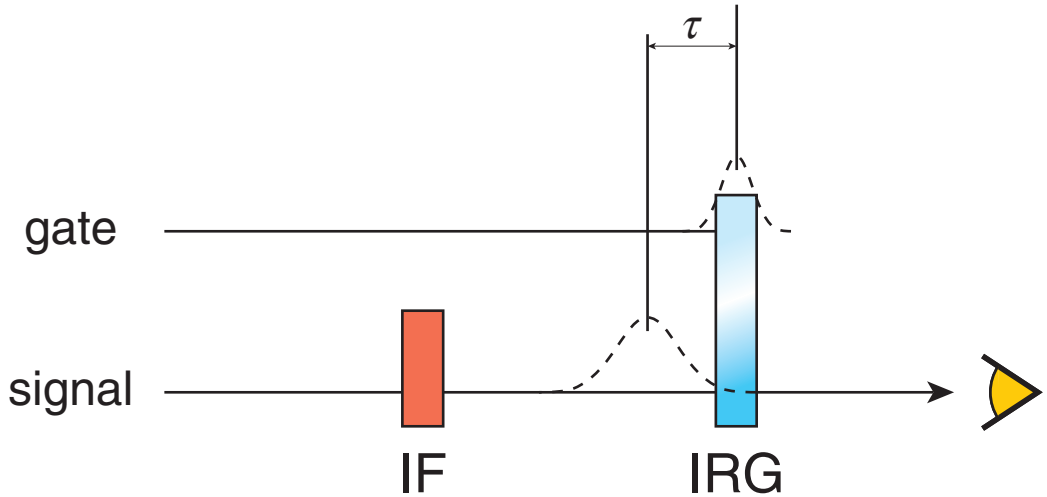
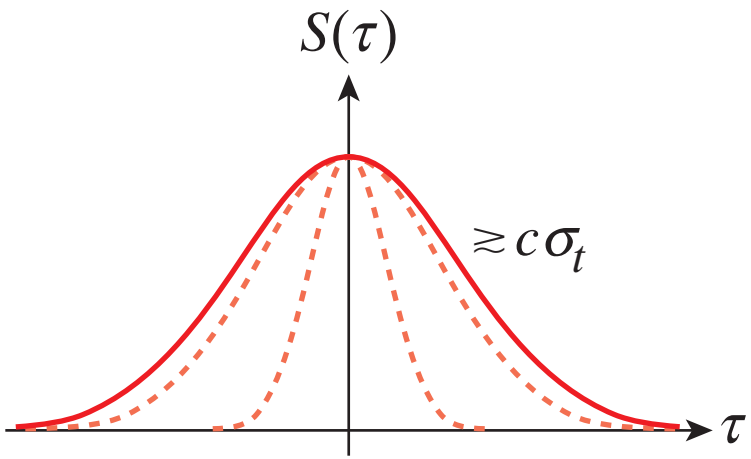


Experiment 1

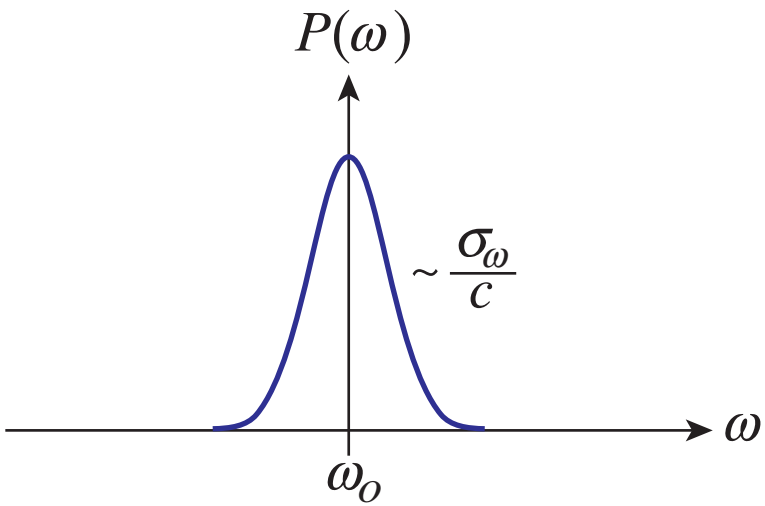


Experiment 1



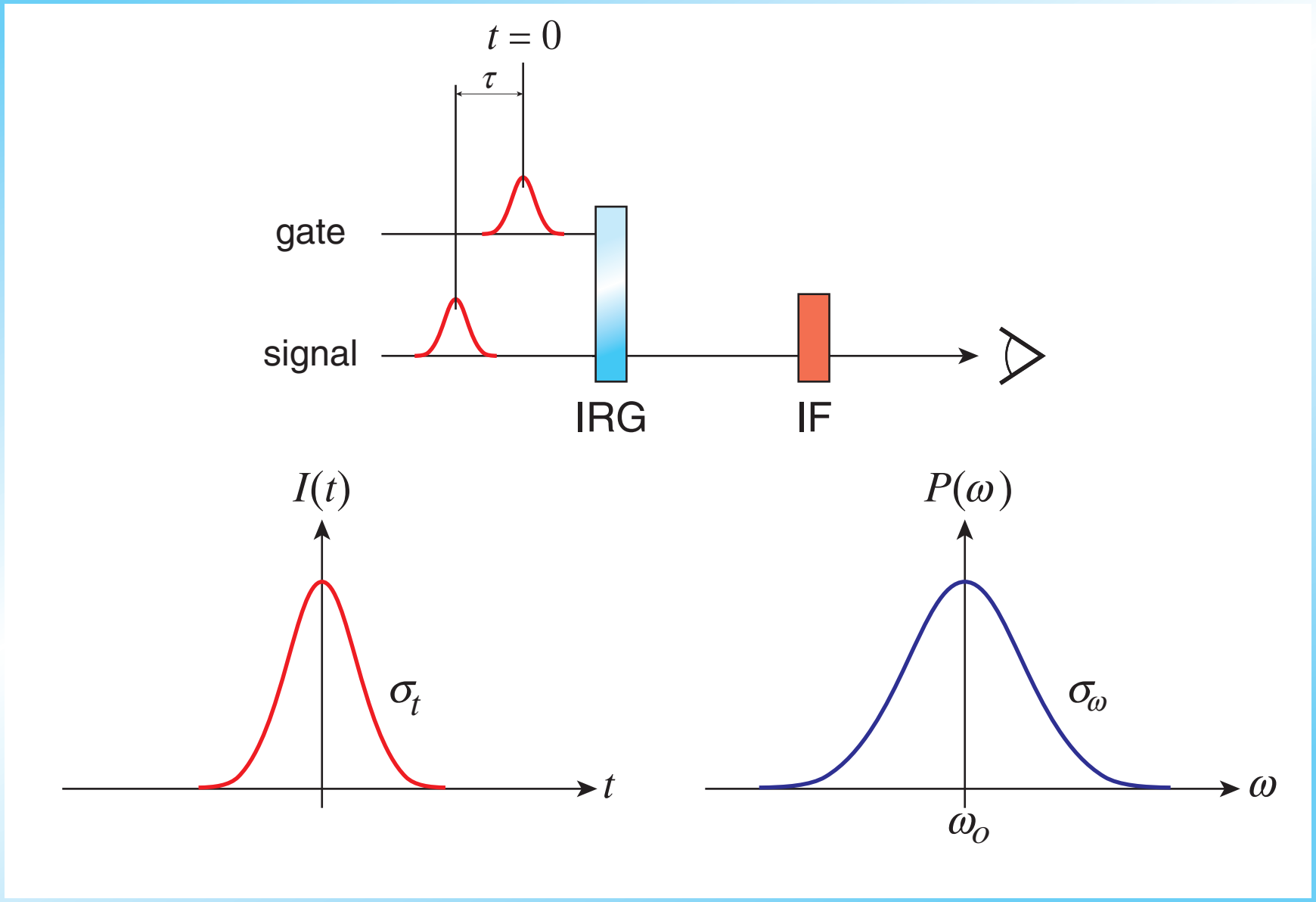


time resolution

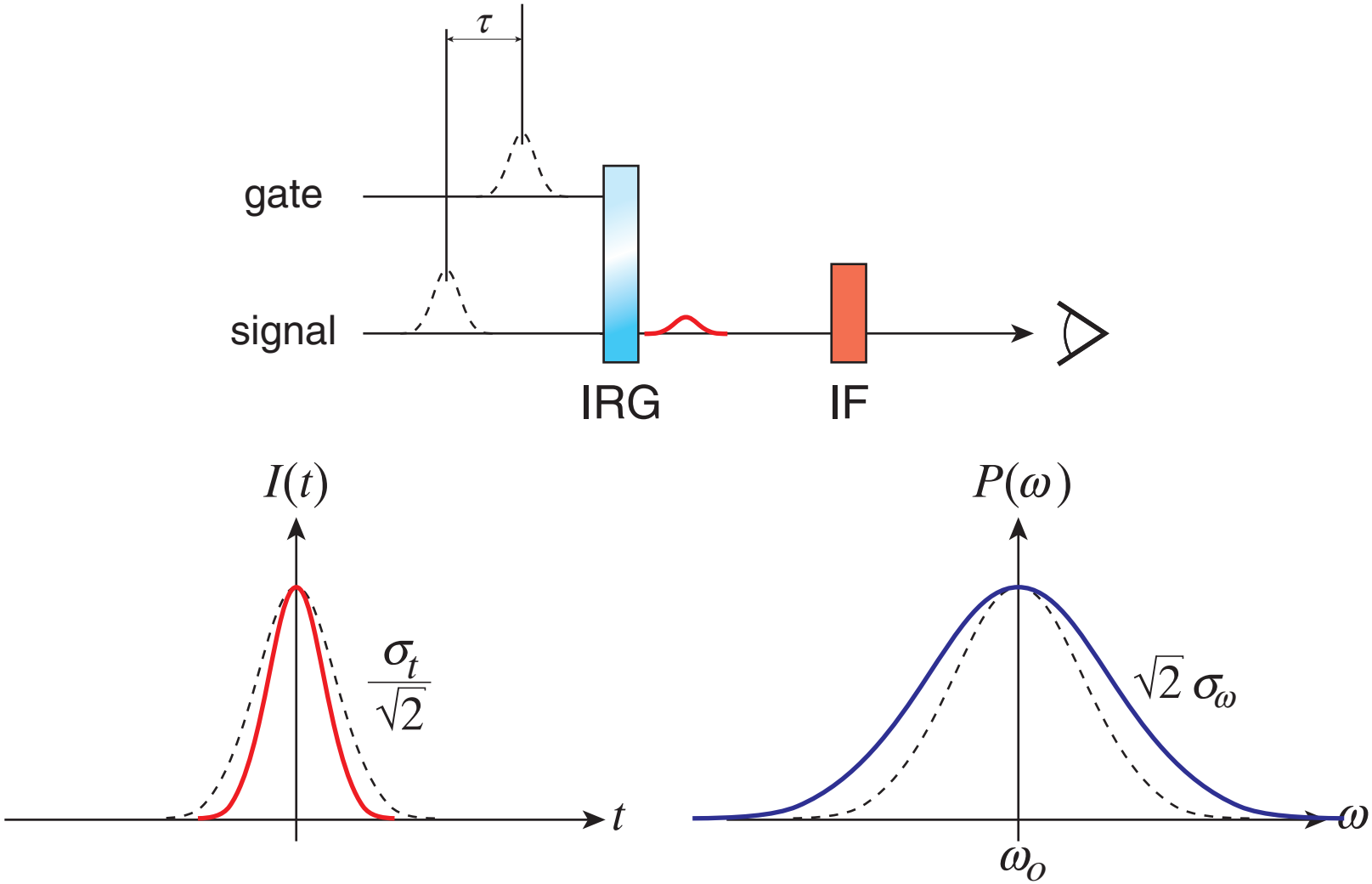


frequency resolution

Experiment 2



Experiment 2



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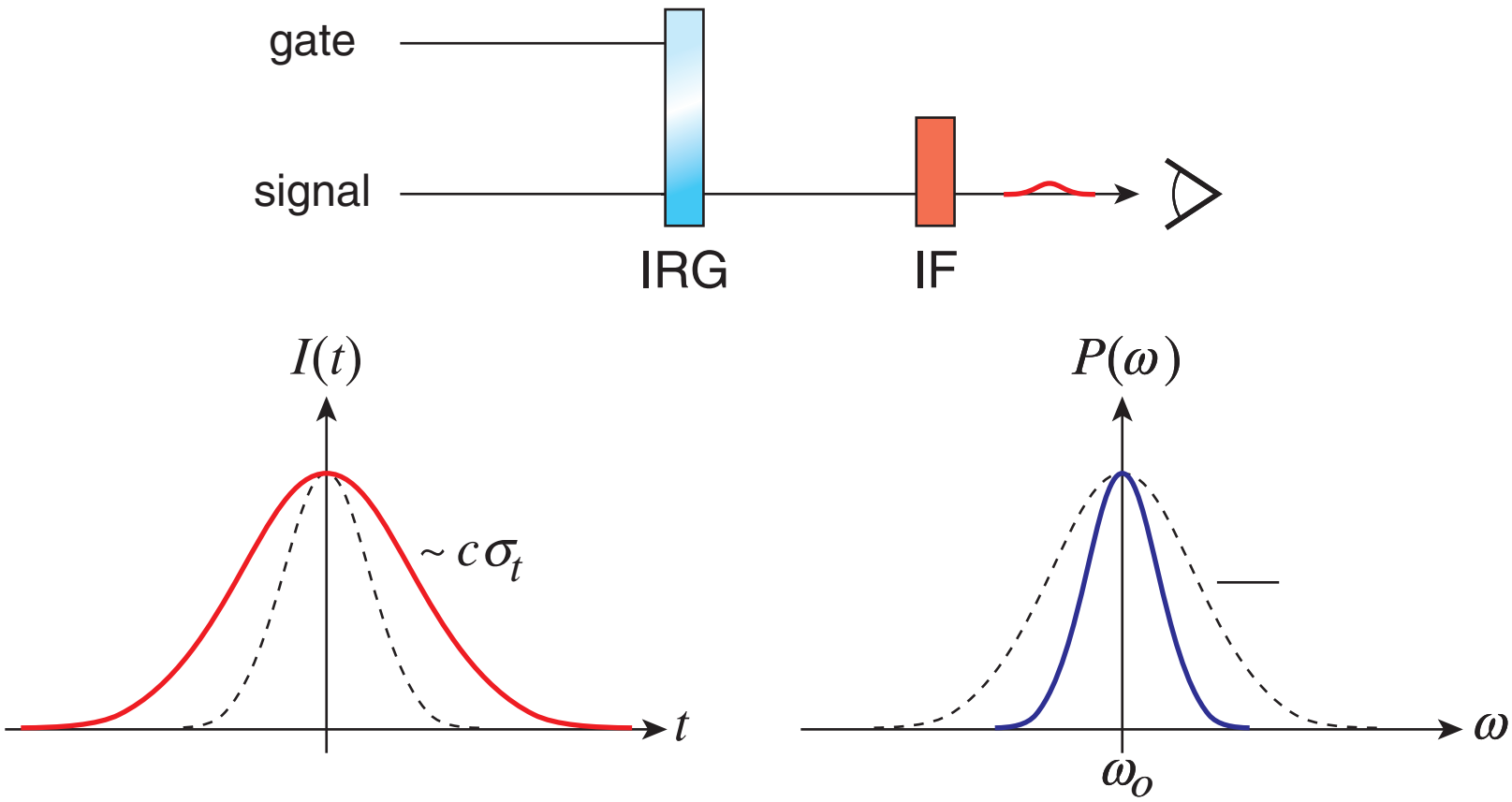
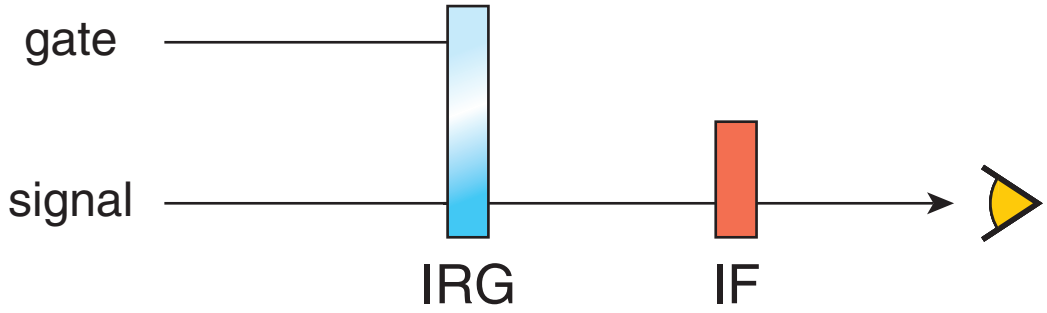
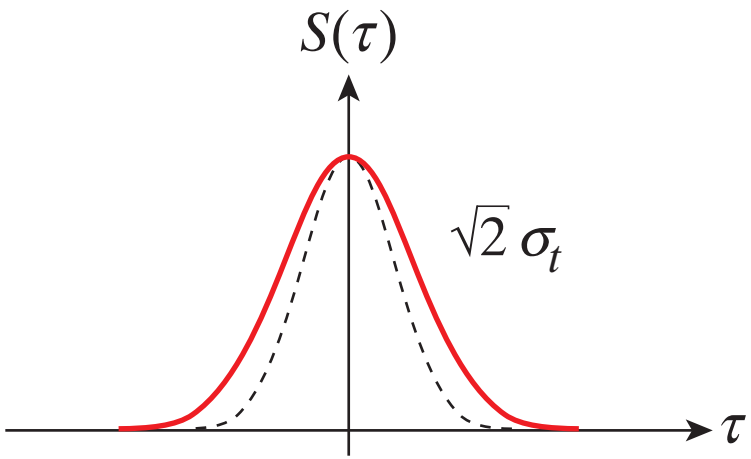
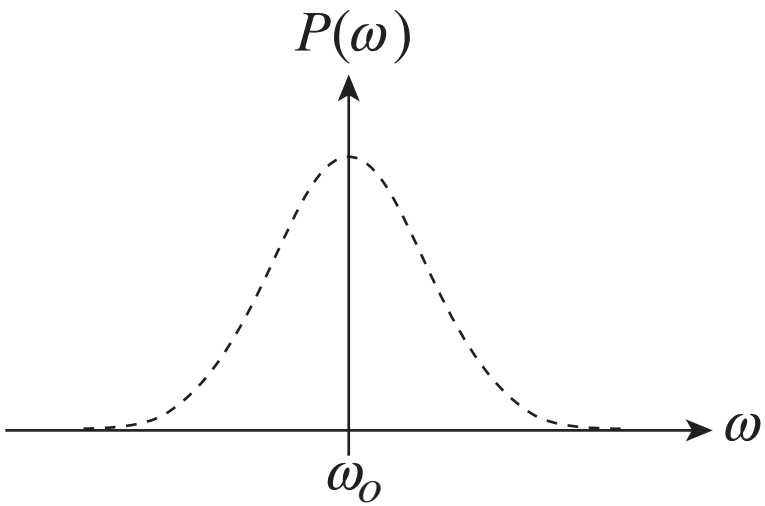


Experiment 2

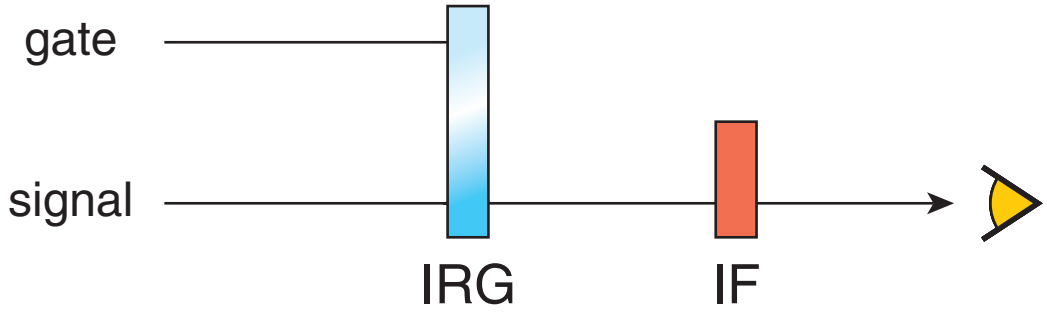


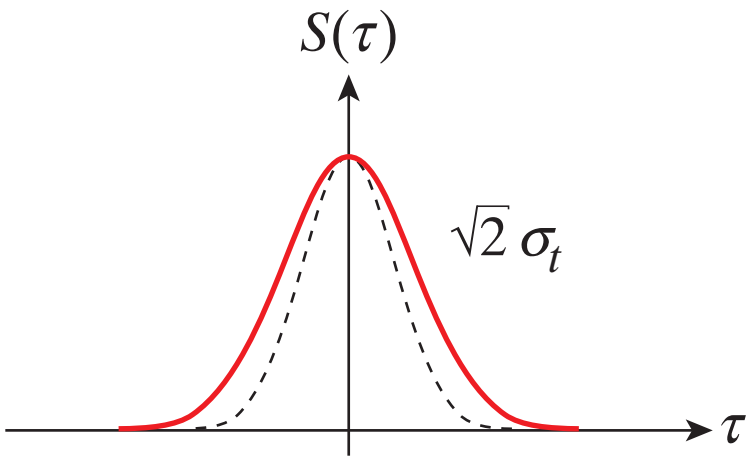


time resolution

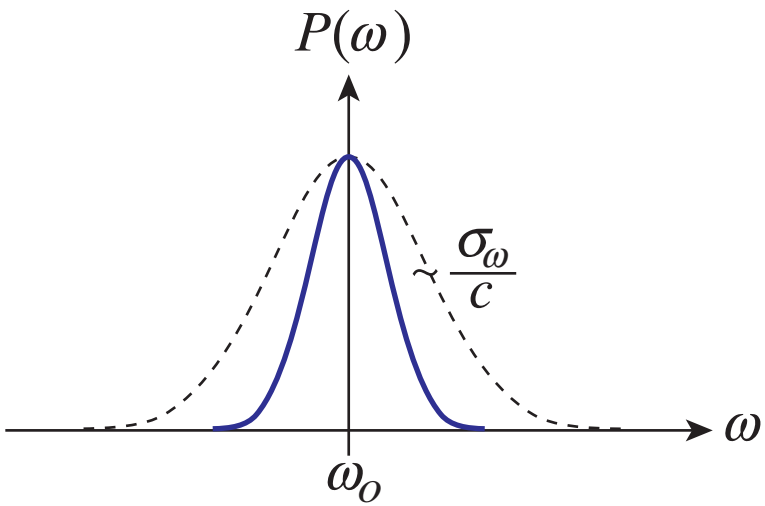


Experiment 2





time resolution



spectral resolution

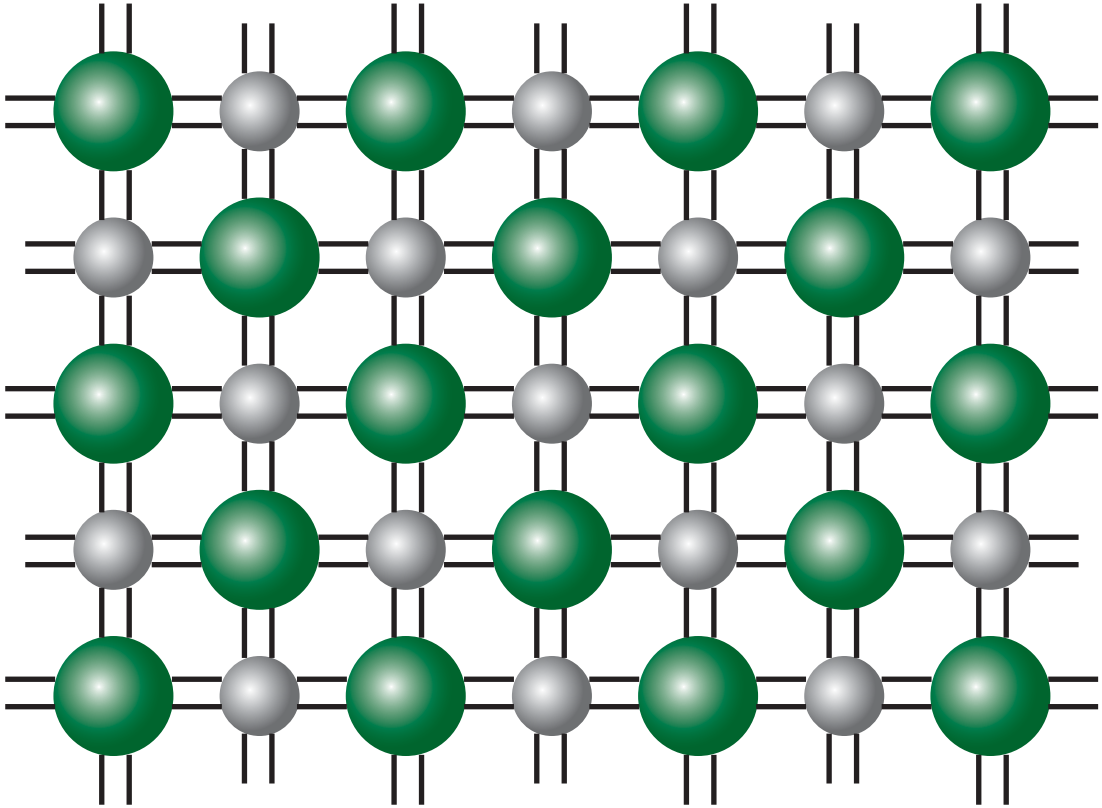
Outline

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

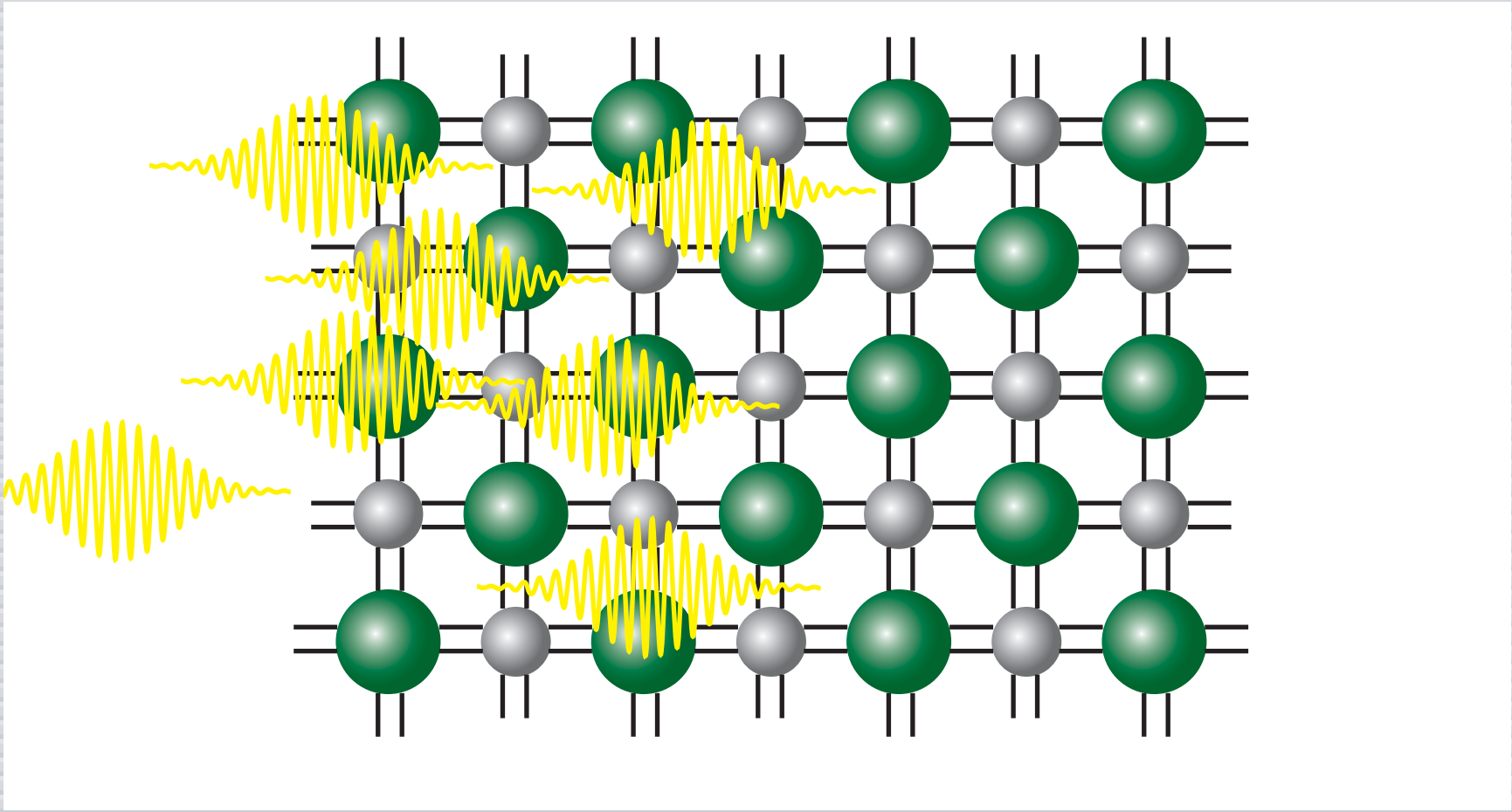
Introduction

short laser pulses can drive structural transitions

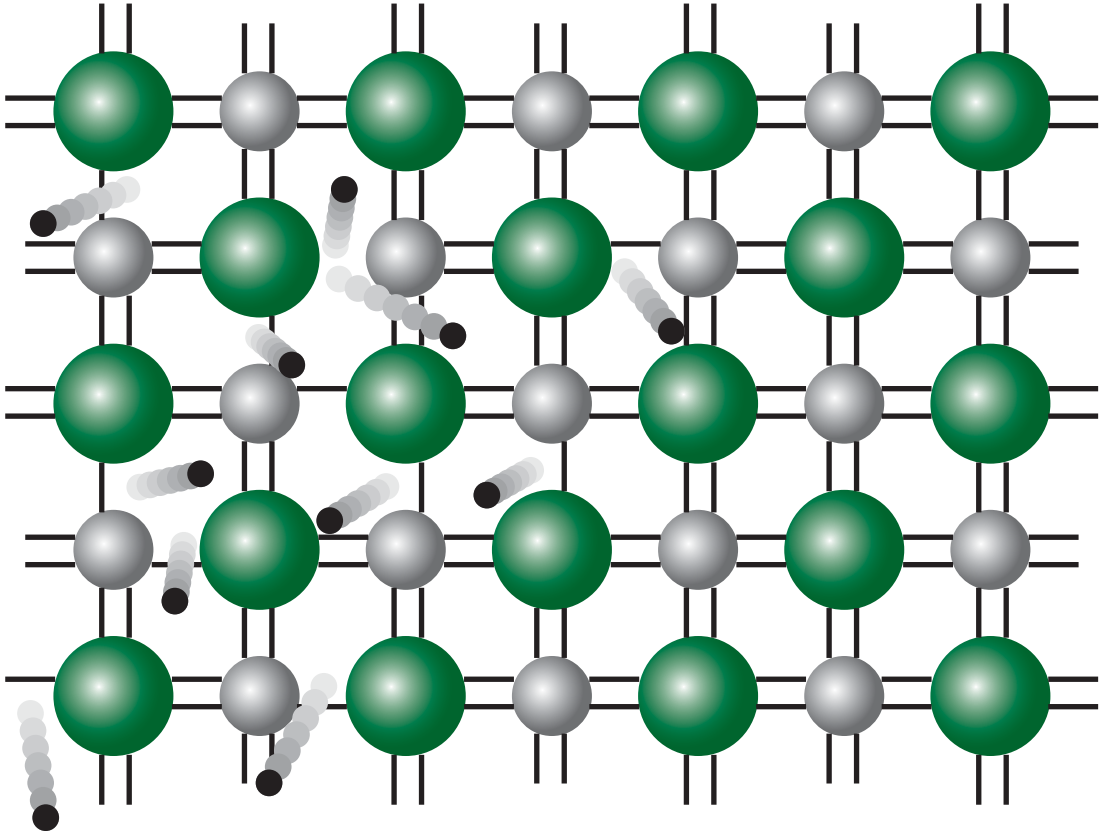
how do femtosecond laser pulses alter a solid?



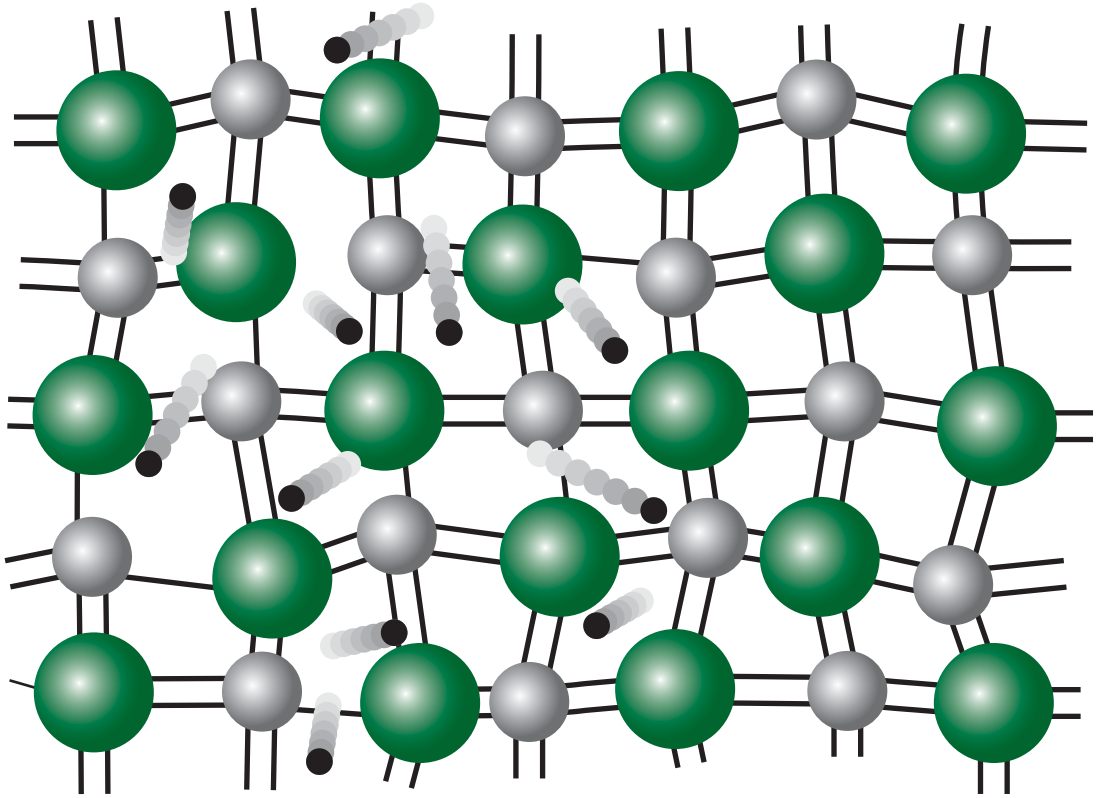
photons excite valence electrons…



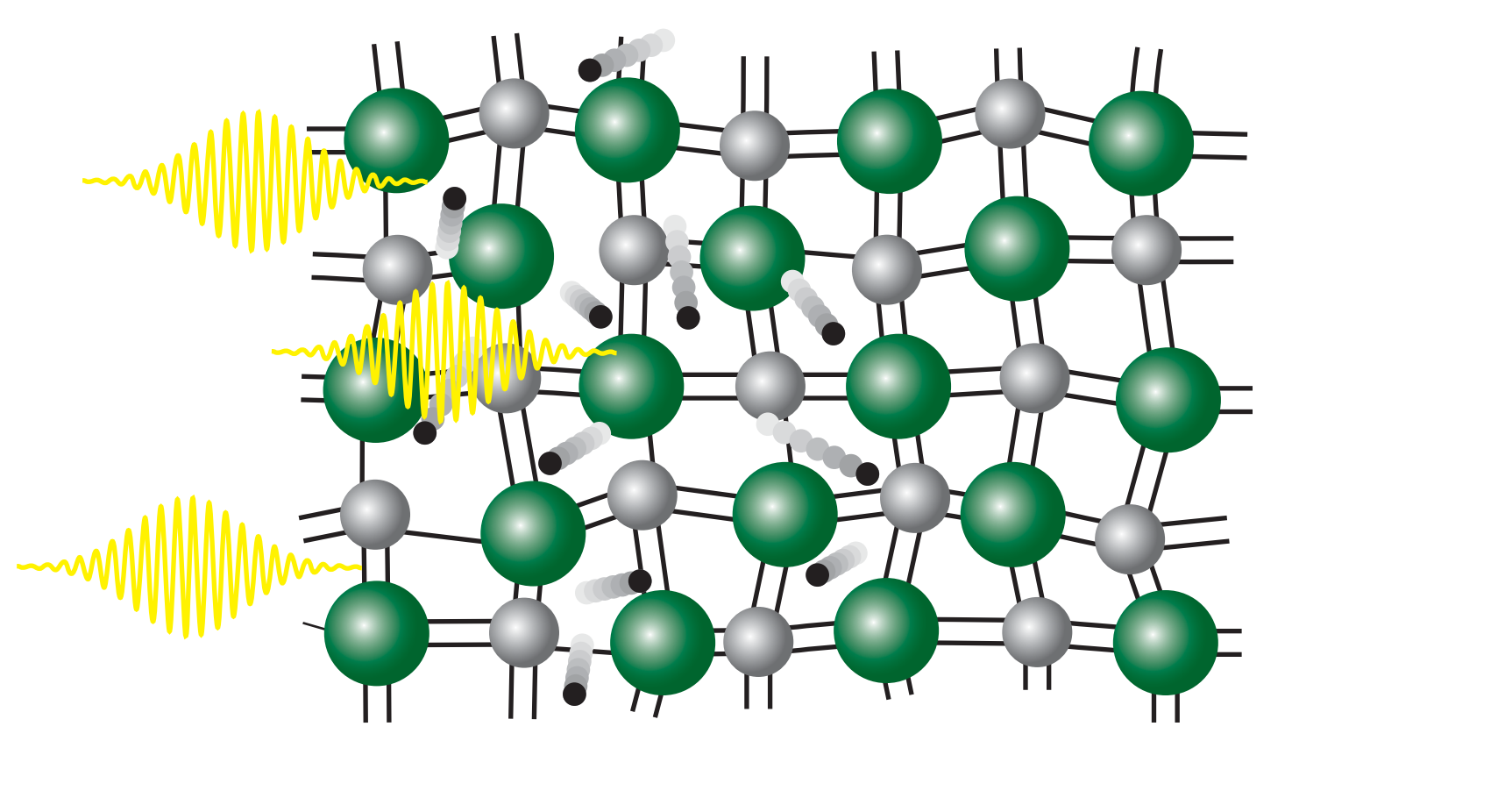
… and create free electrons…



… causing electronic and structural changes…



… which we detect with a second laser pulse



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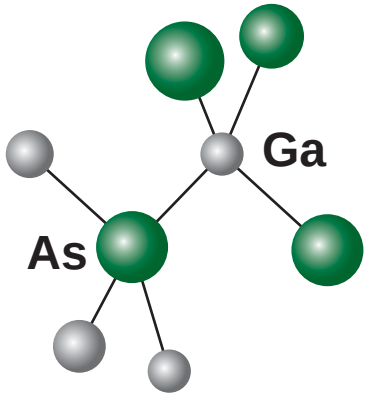
structure



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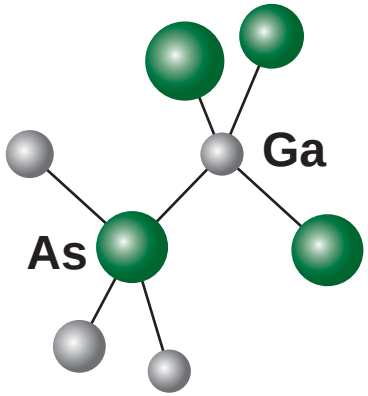
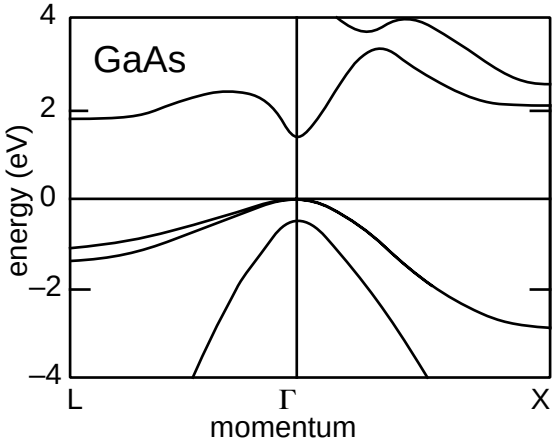
structure



band structure



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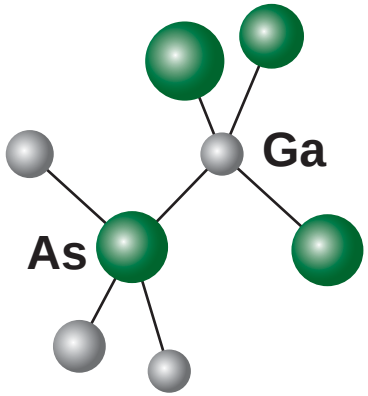
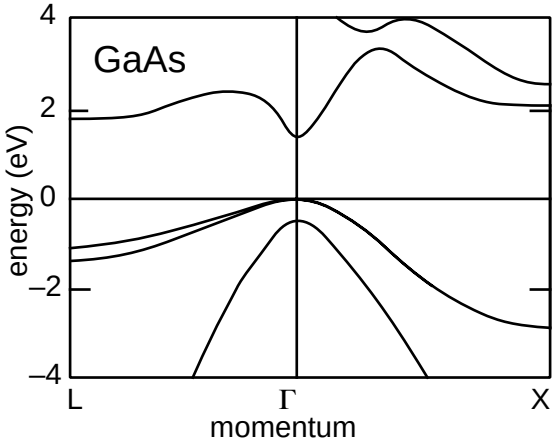
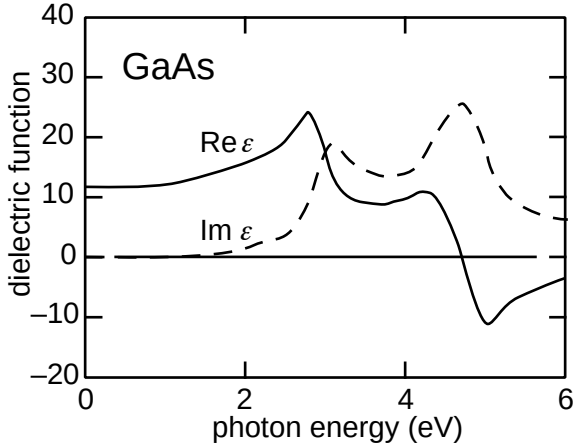
structure



band structure



dielectric function



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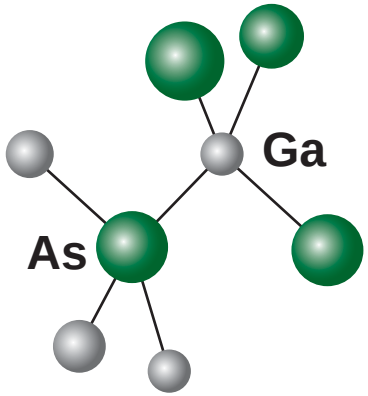
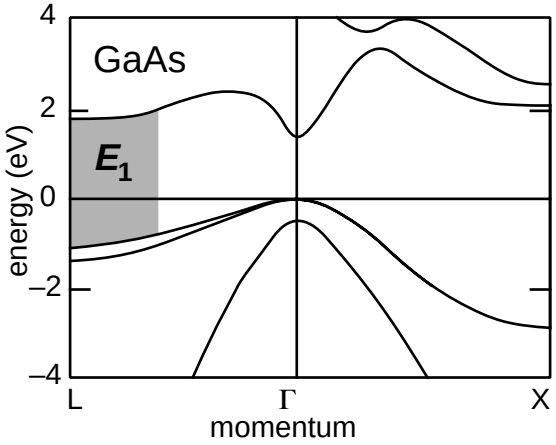
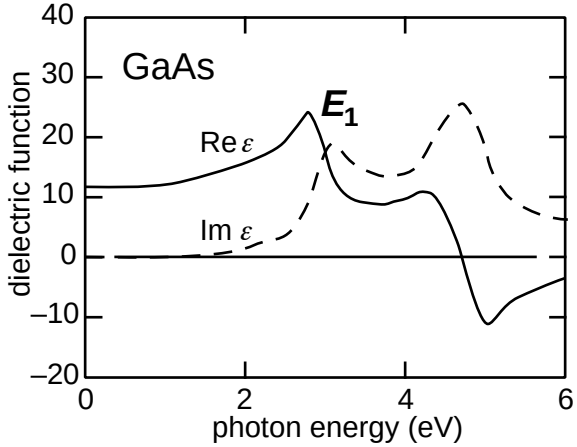
structure



band structure

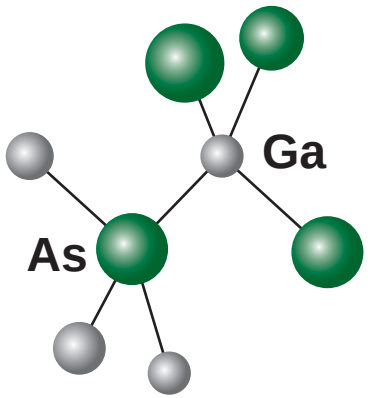


dielectric function

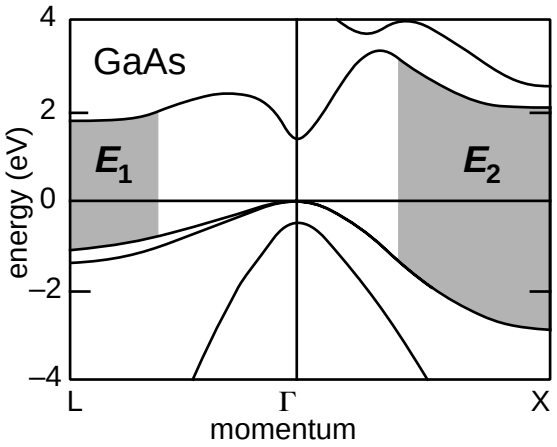


Introduction

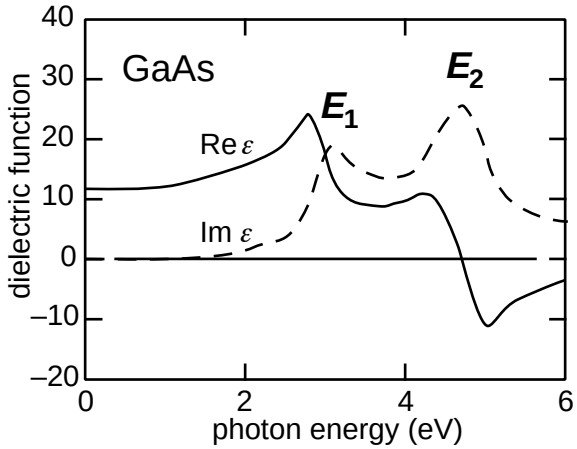
structure



band structure



dielectric function



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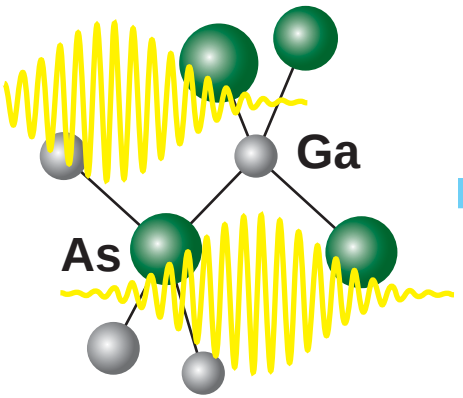
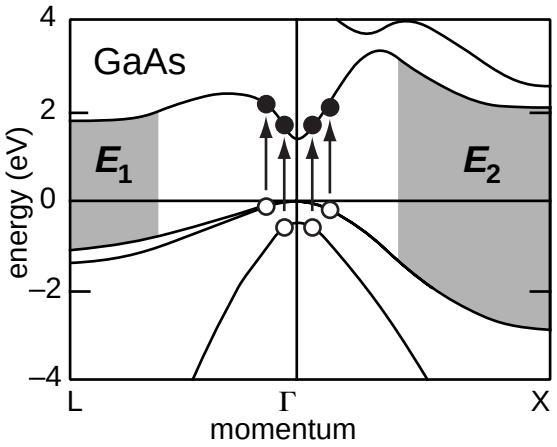
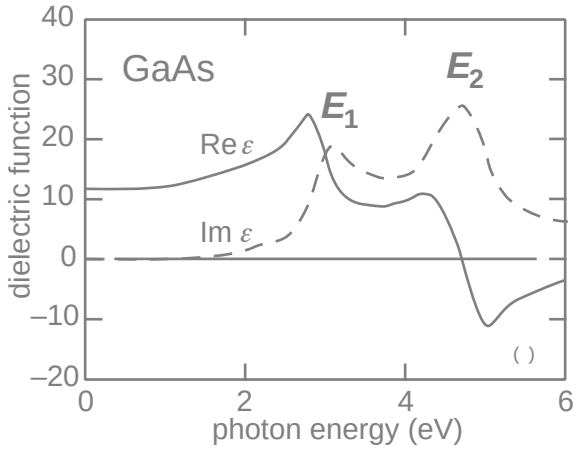
structure



band structure



dielectric function



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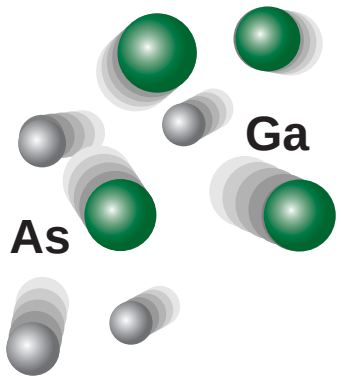
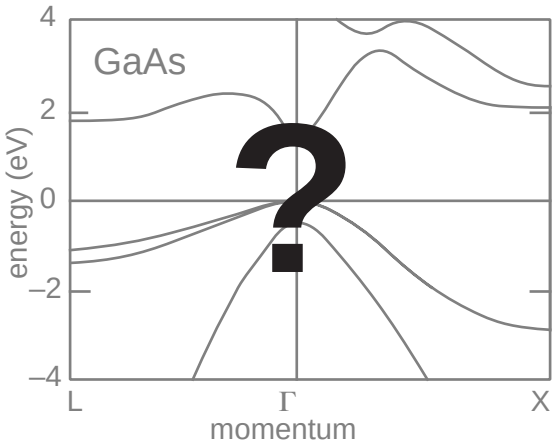
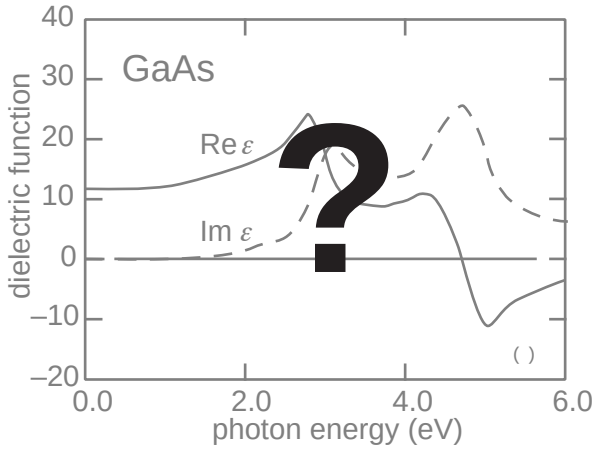
structure



band structure



dielectric function



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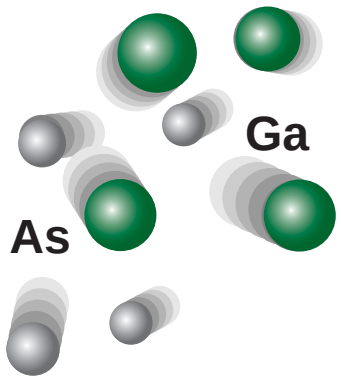
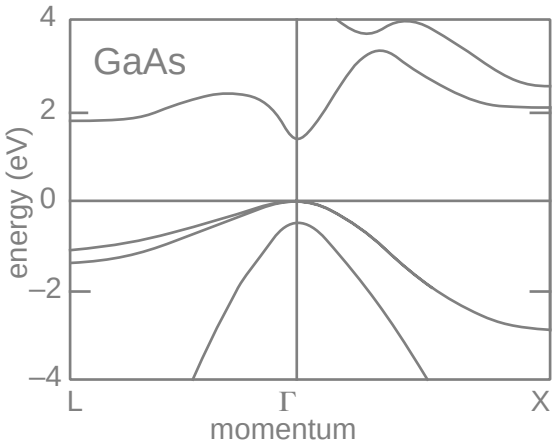
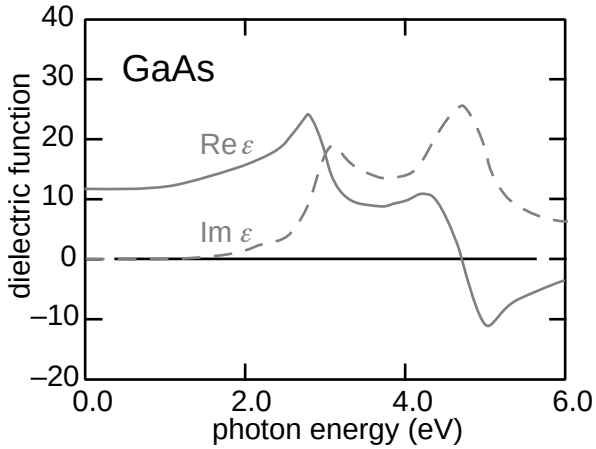
structure



band structure



dielectric function



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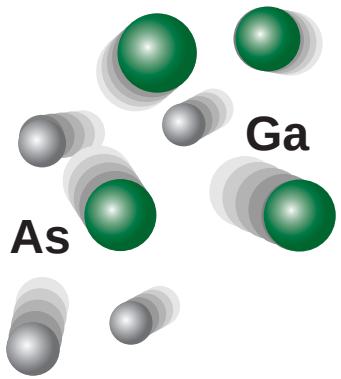
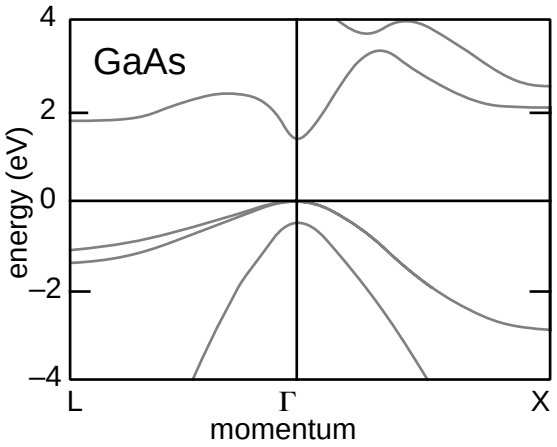
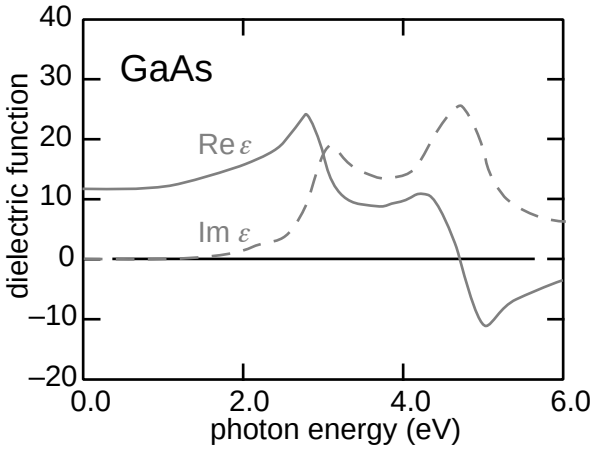
structure



band structure



dielectric function



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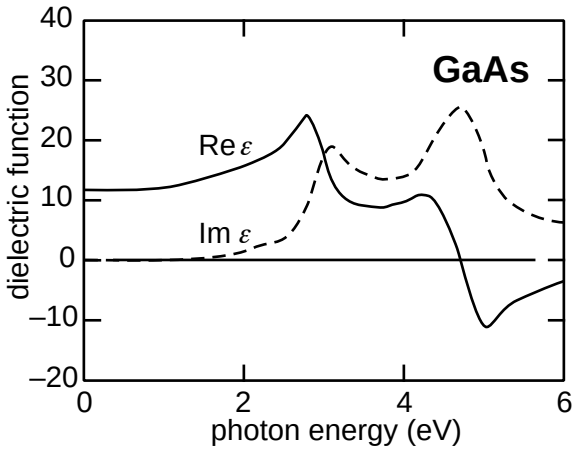

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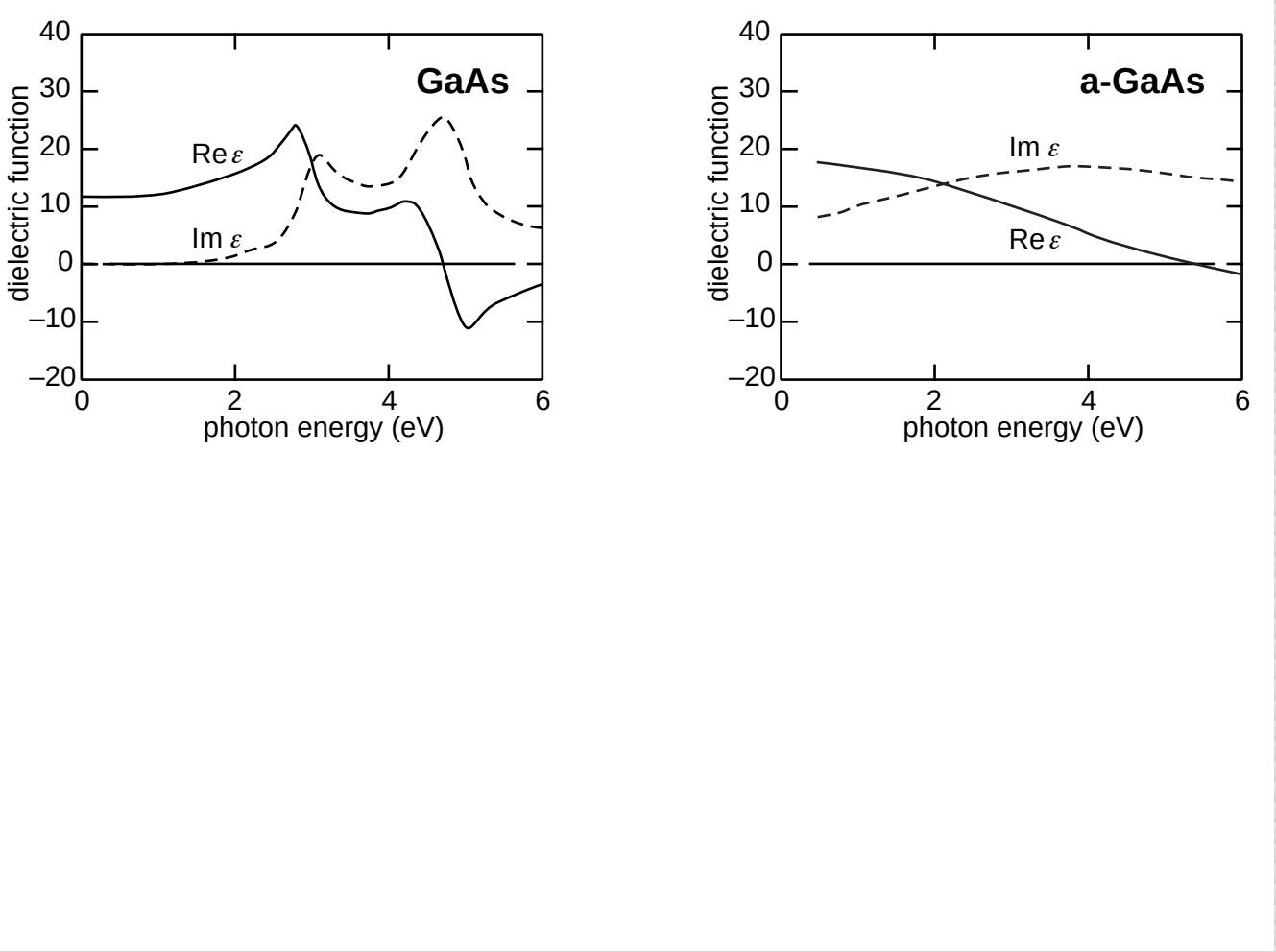

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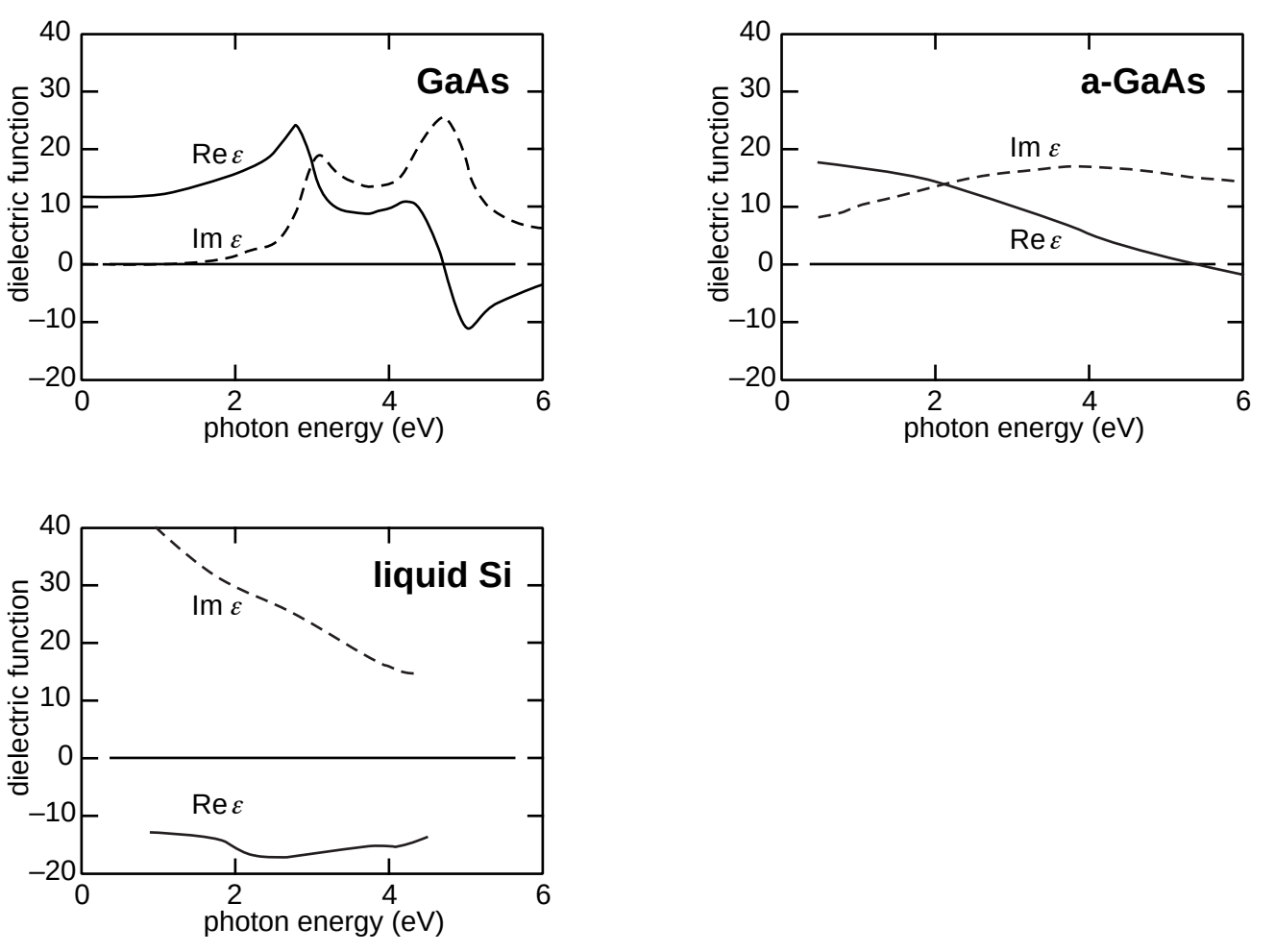

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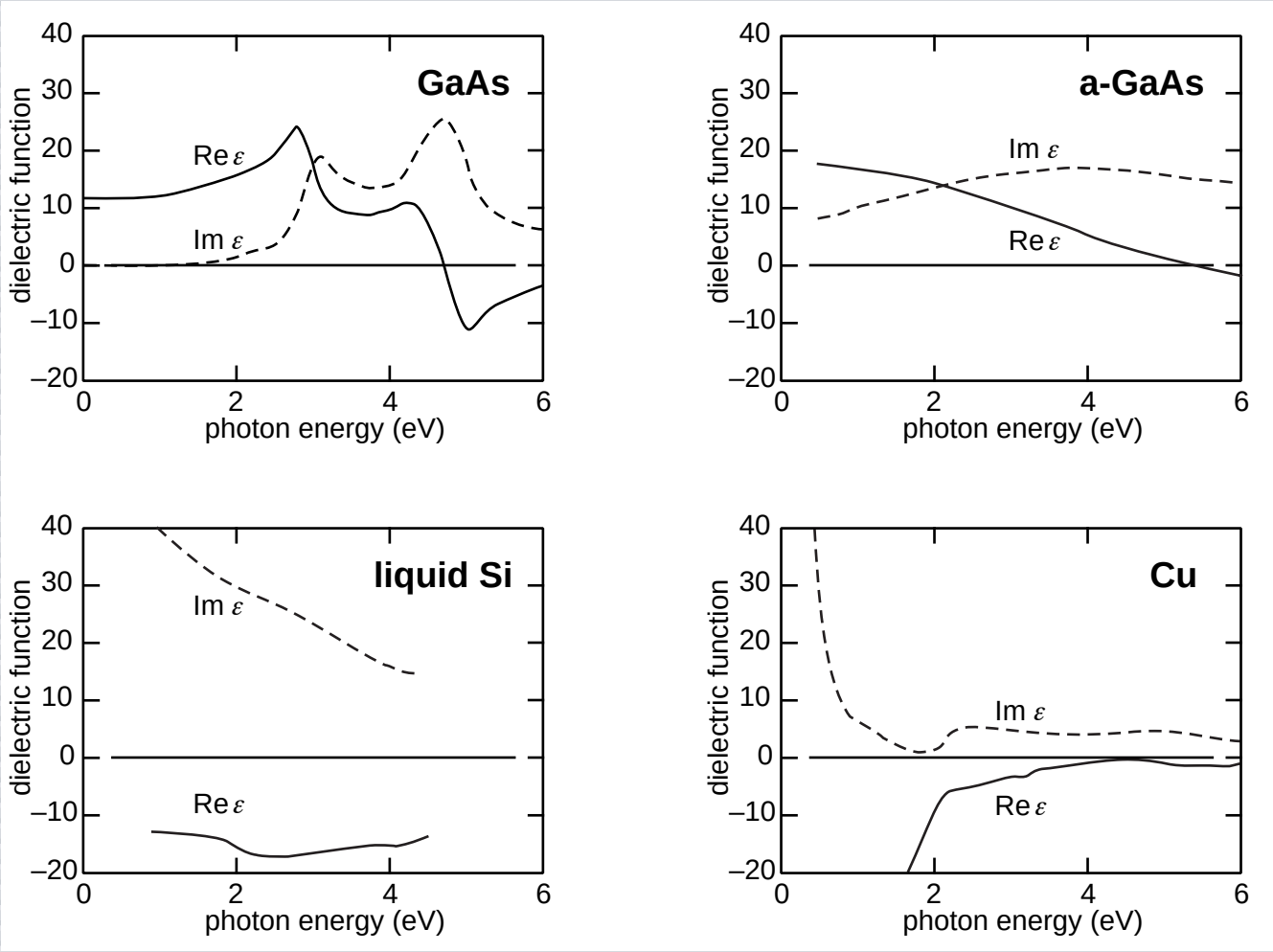
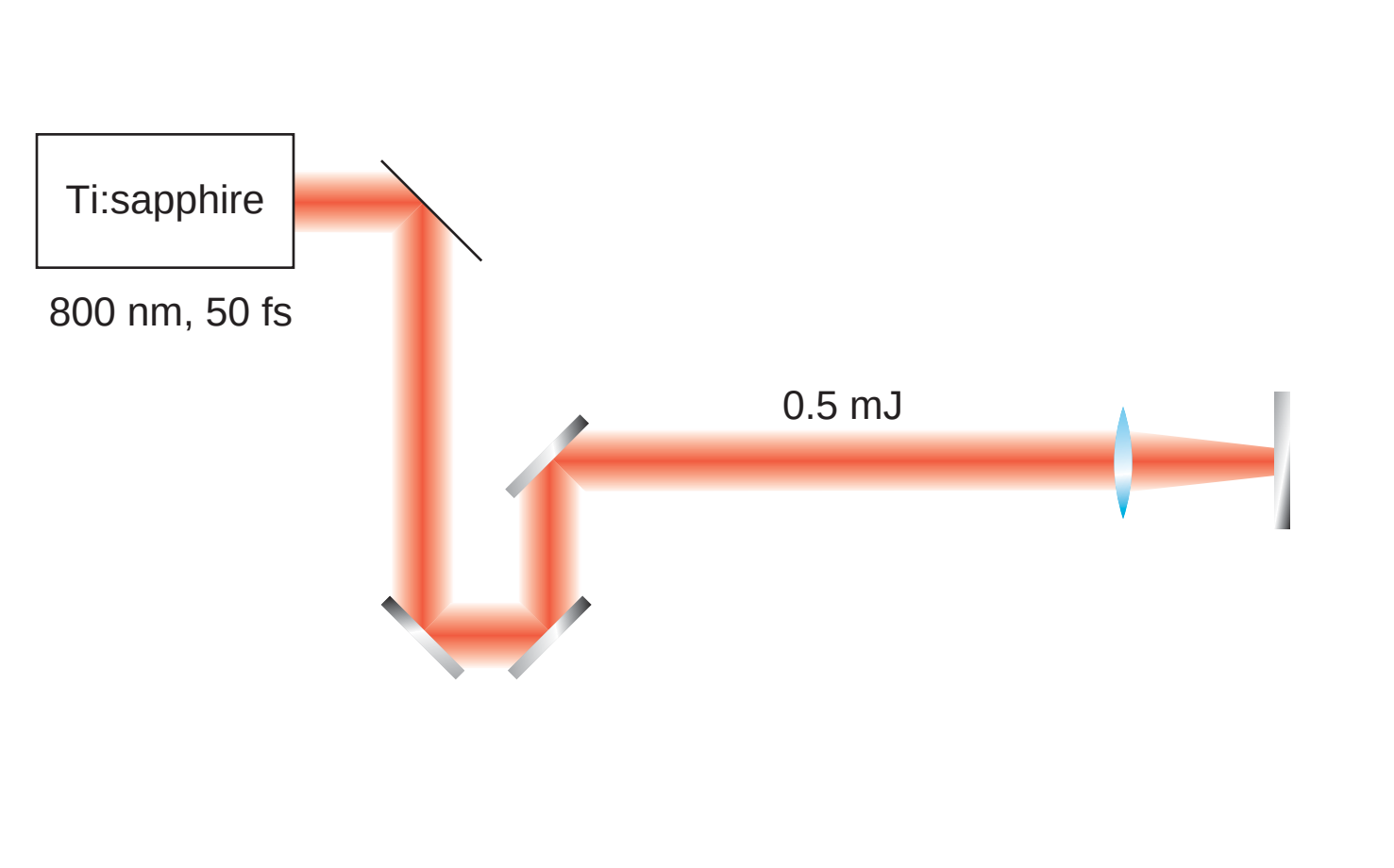
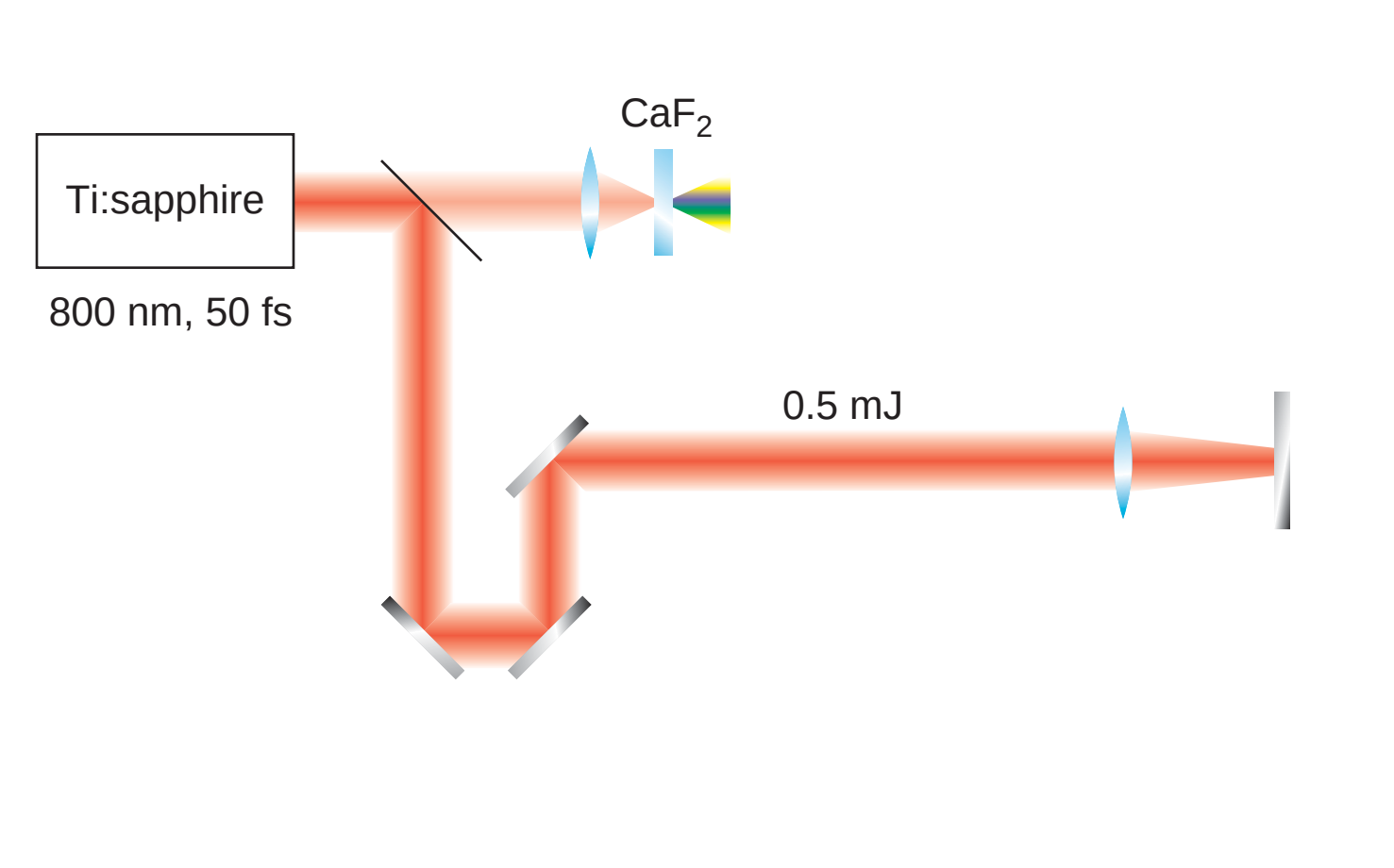


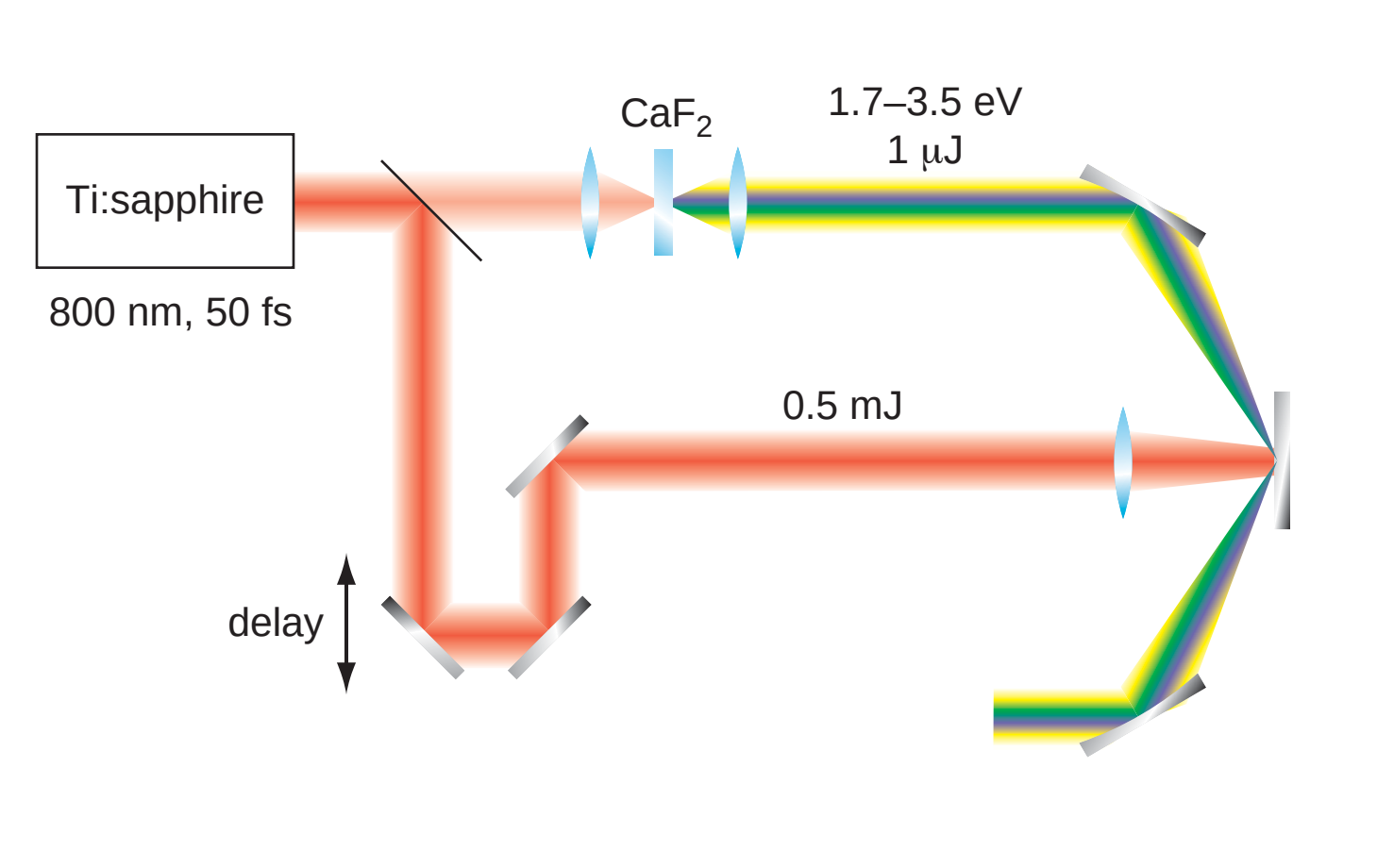
broadband time-resolved ellipsometry



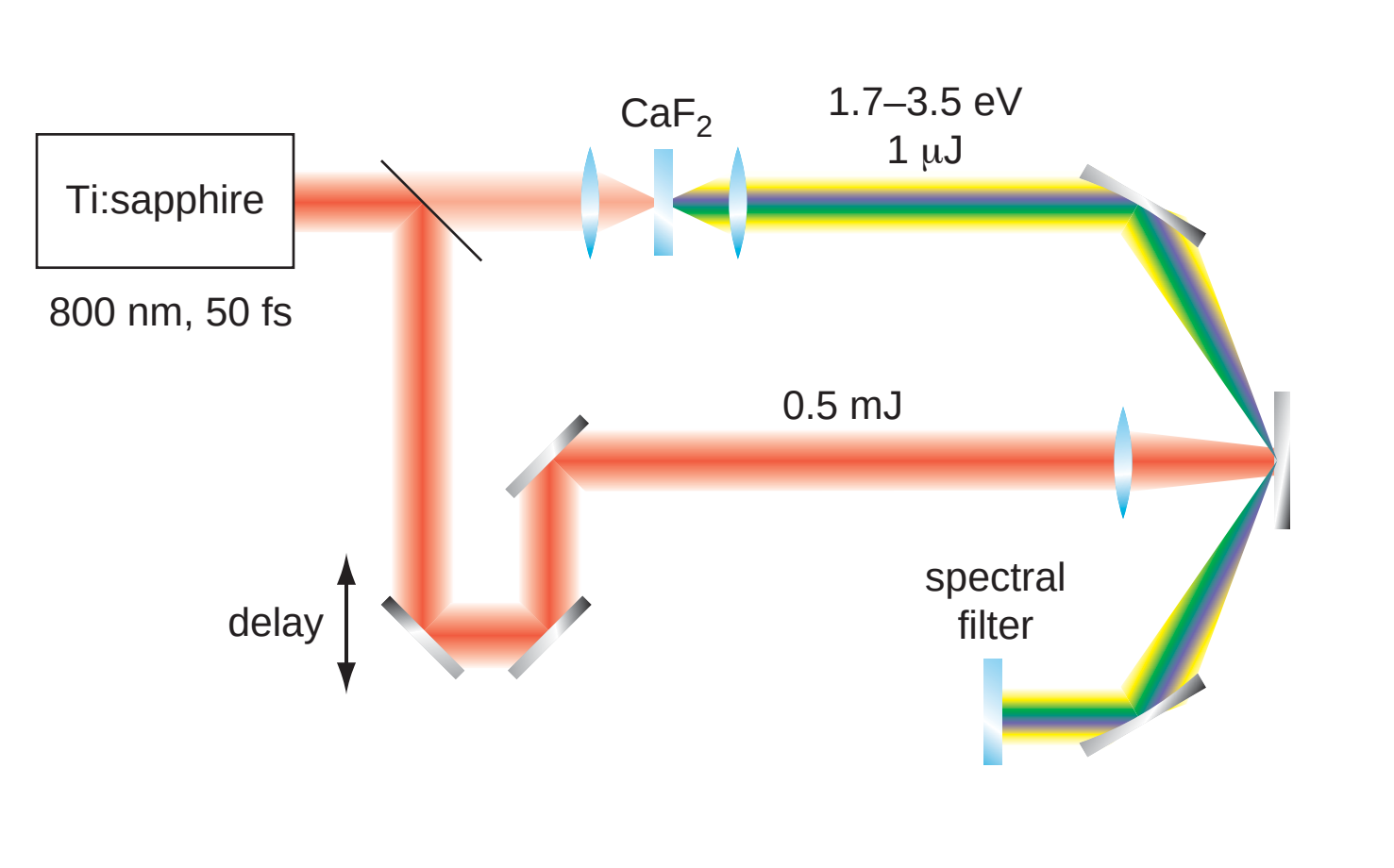
broadband time-resolved ellipsometry



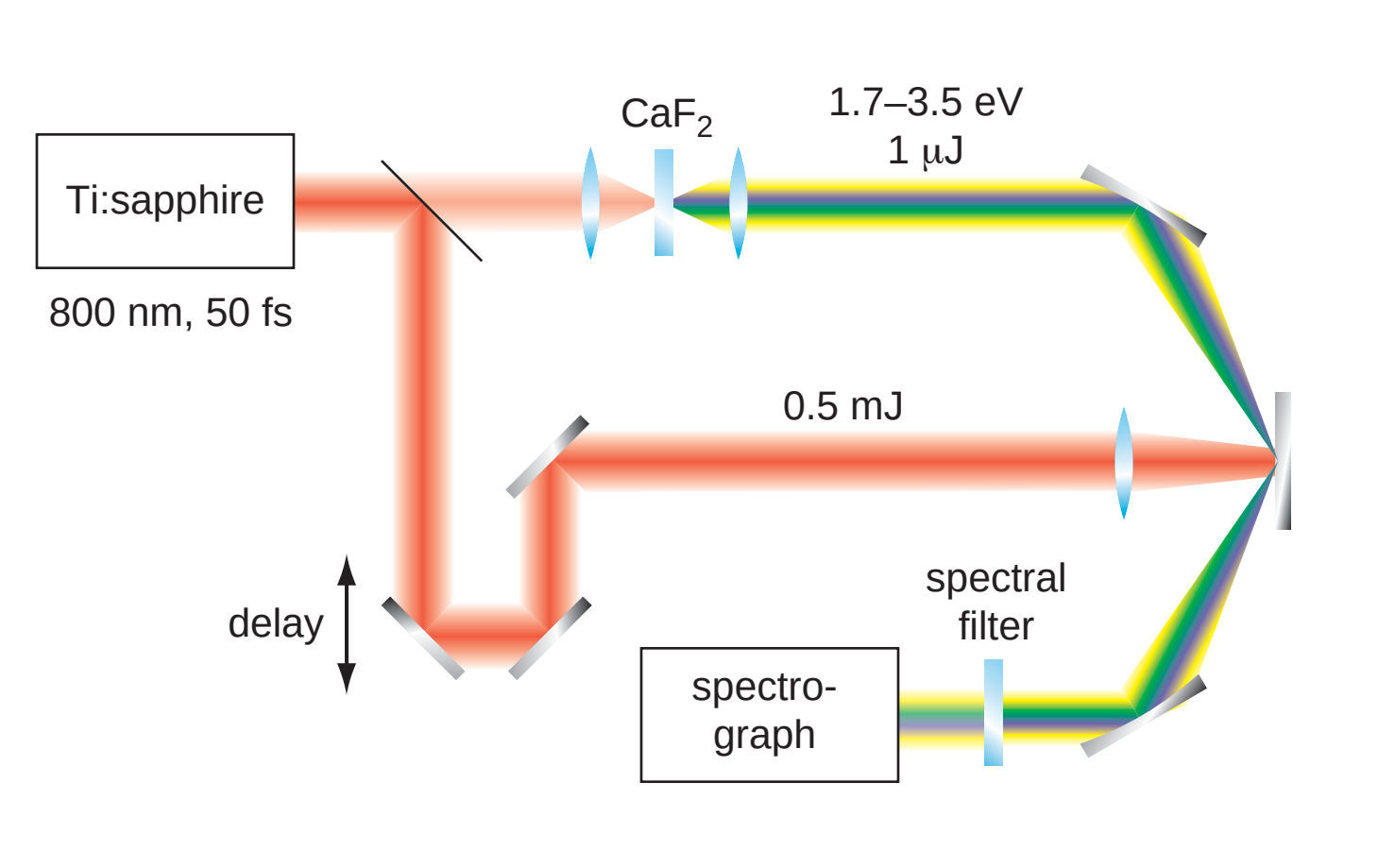
broadband time-resolved ellipsometry



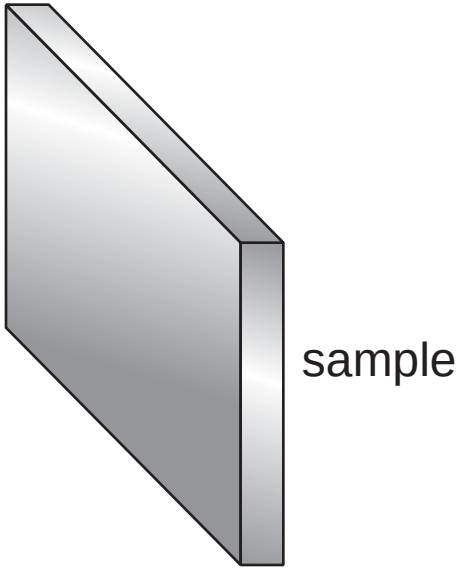
broadband time-resolved ellipsometry



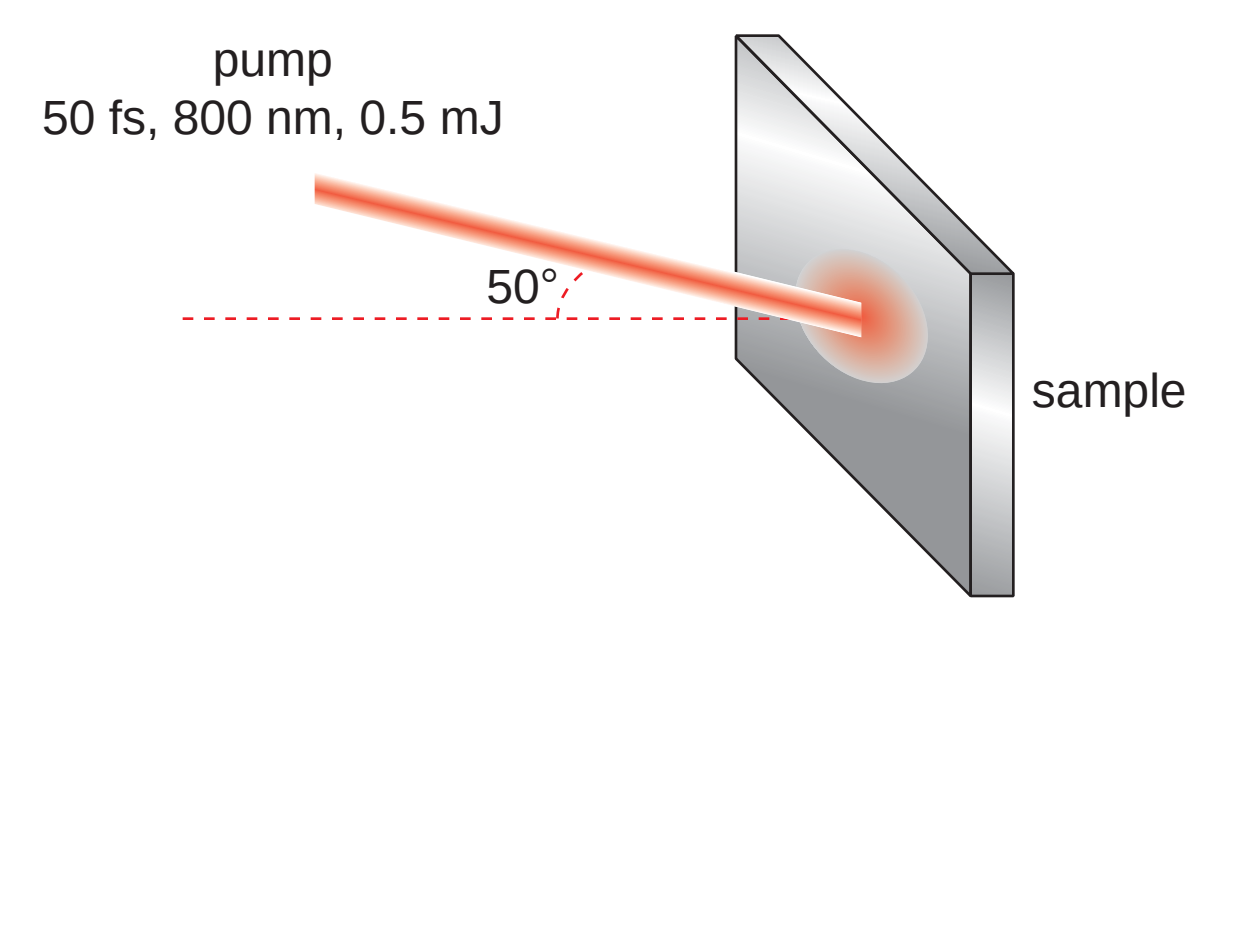
broadband time-resolved ellipsometry



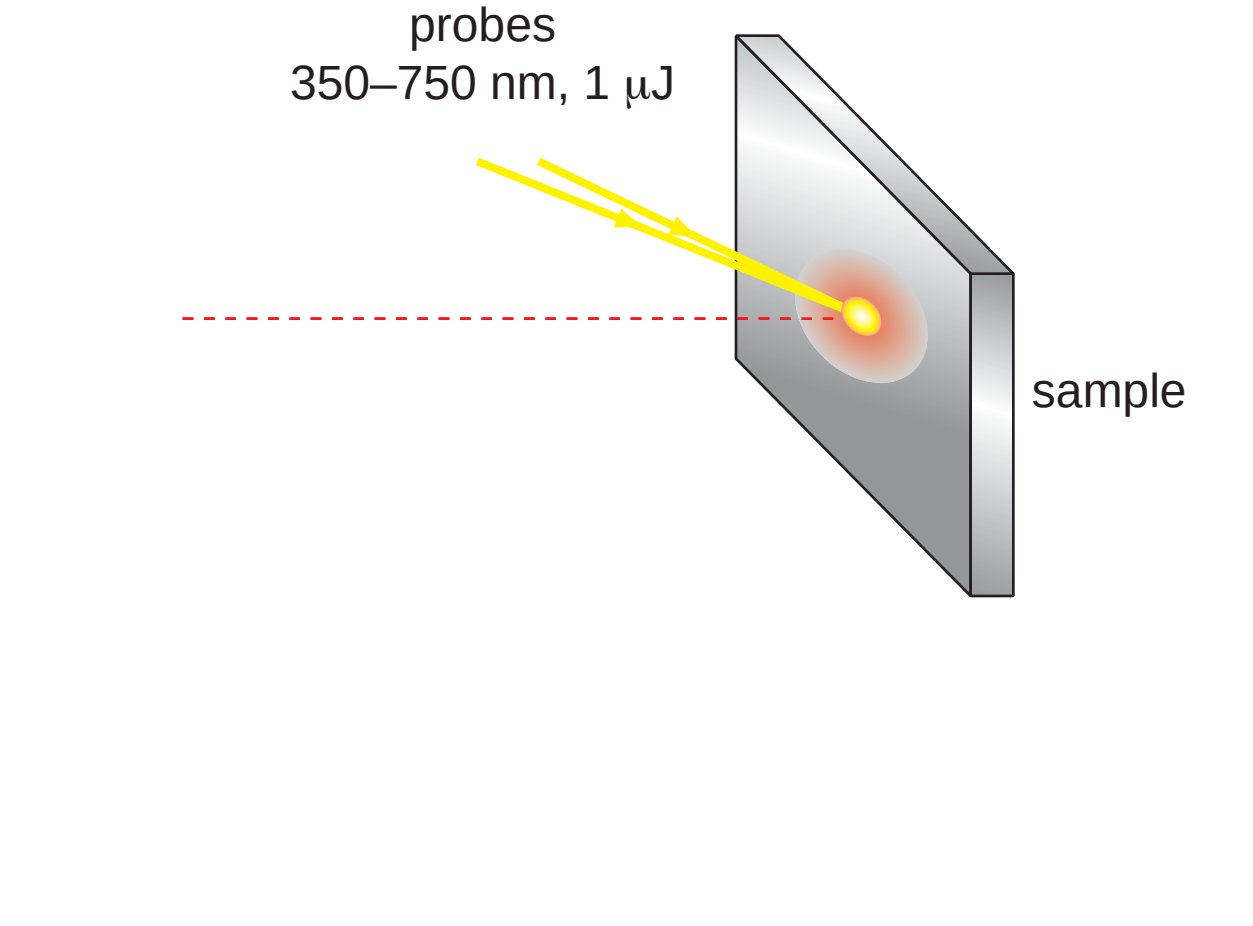
broadband time-resolved ellipsometry



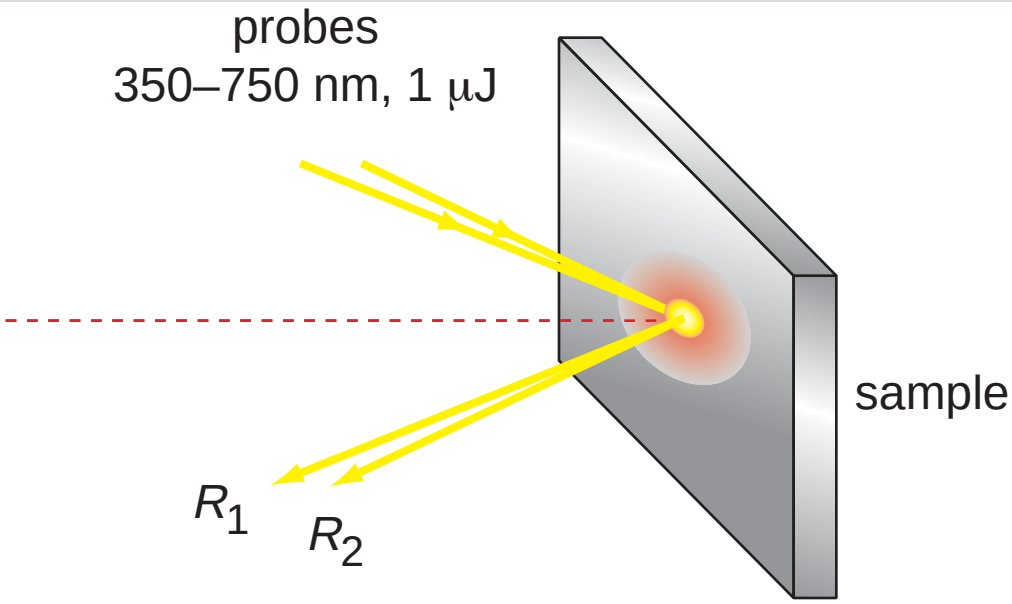
broadband time-resolved ellipsometry



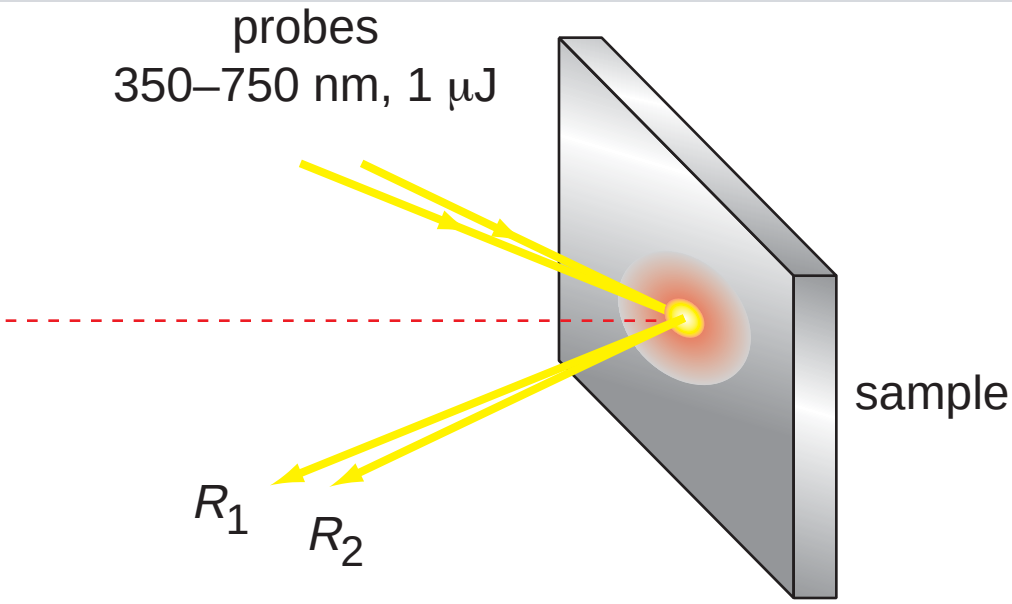
broadband time-resolved ellipsometry



broadband time-resolved ellipsometry

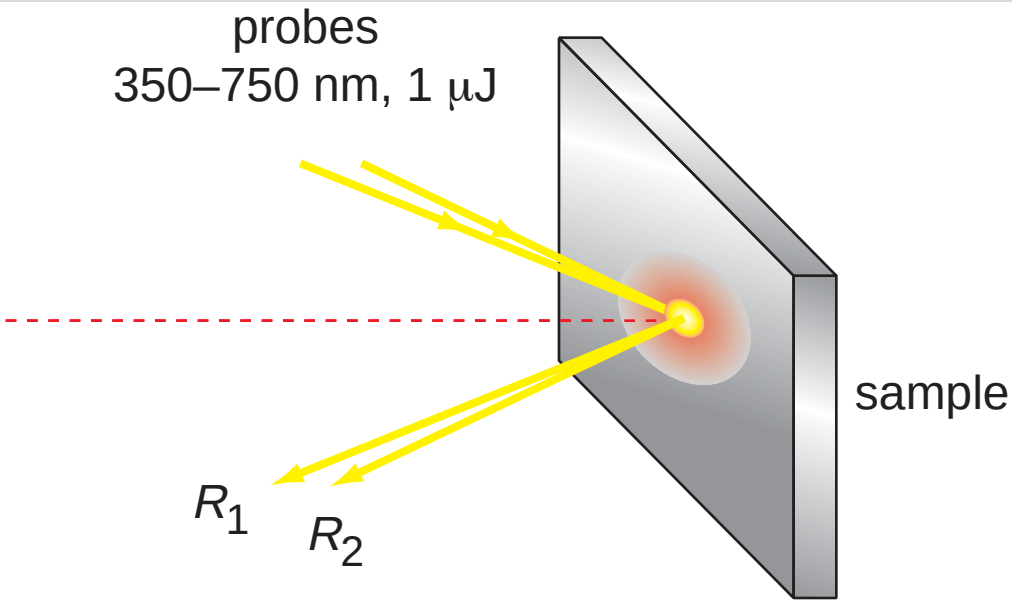


broadband time-resolved ellipsometry



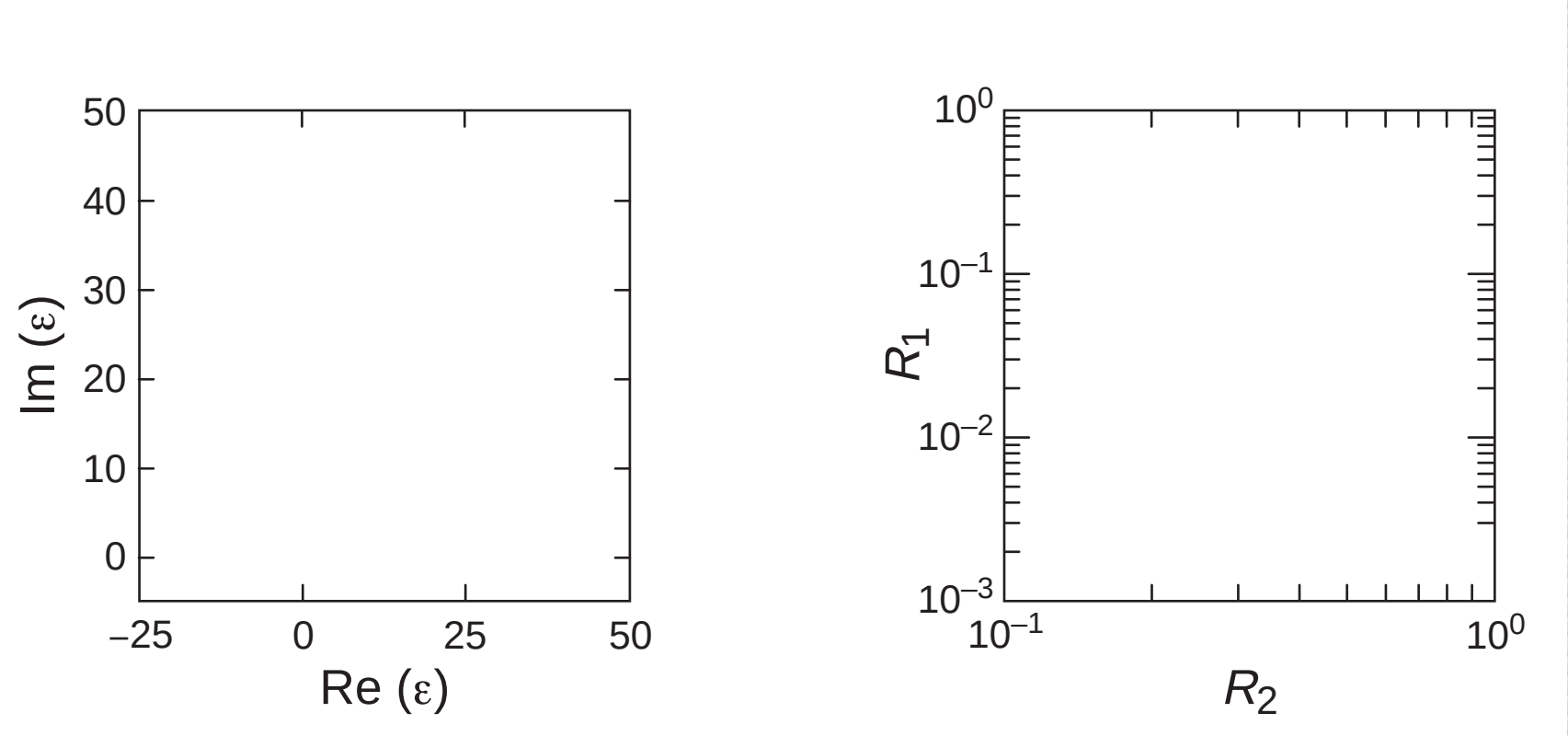
Fresnel equations

broadband time-resolved ellipsometry



Fresnel equations → Re $\varepsilon(\omega)$, Im $\varepsilon(\omega)$

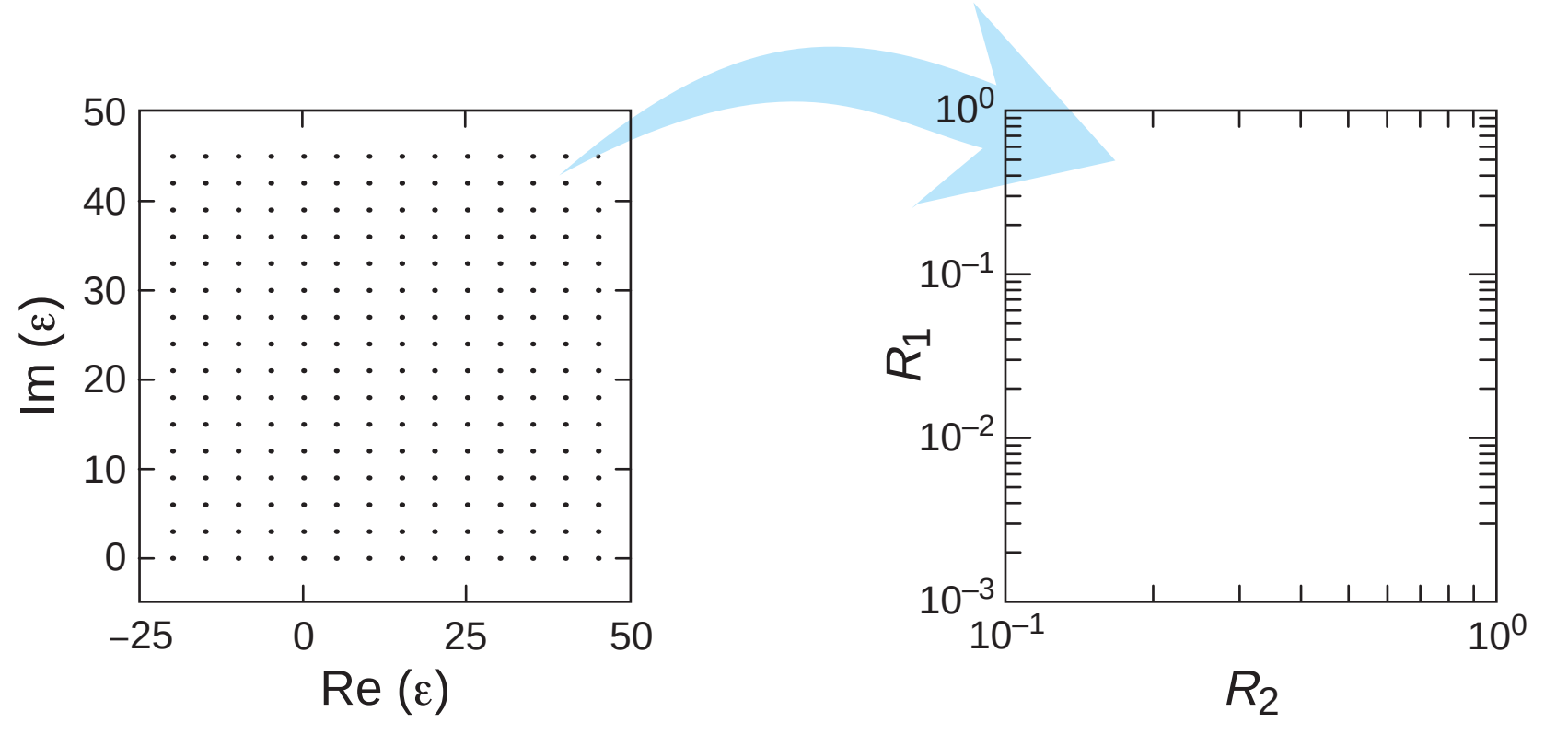
choice of angles



Fresnel equations cannot be inverted analytically

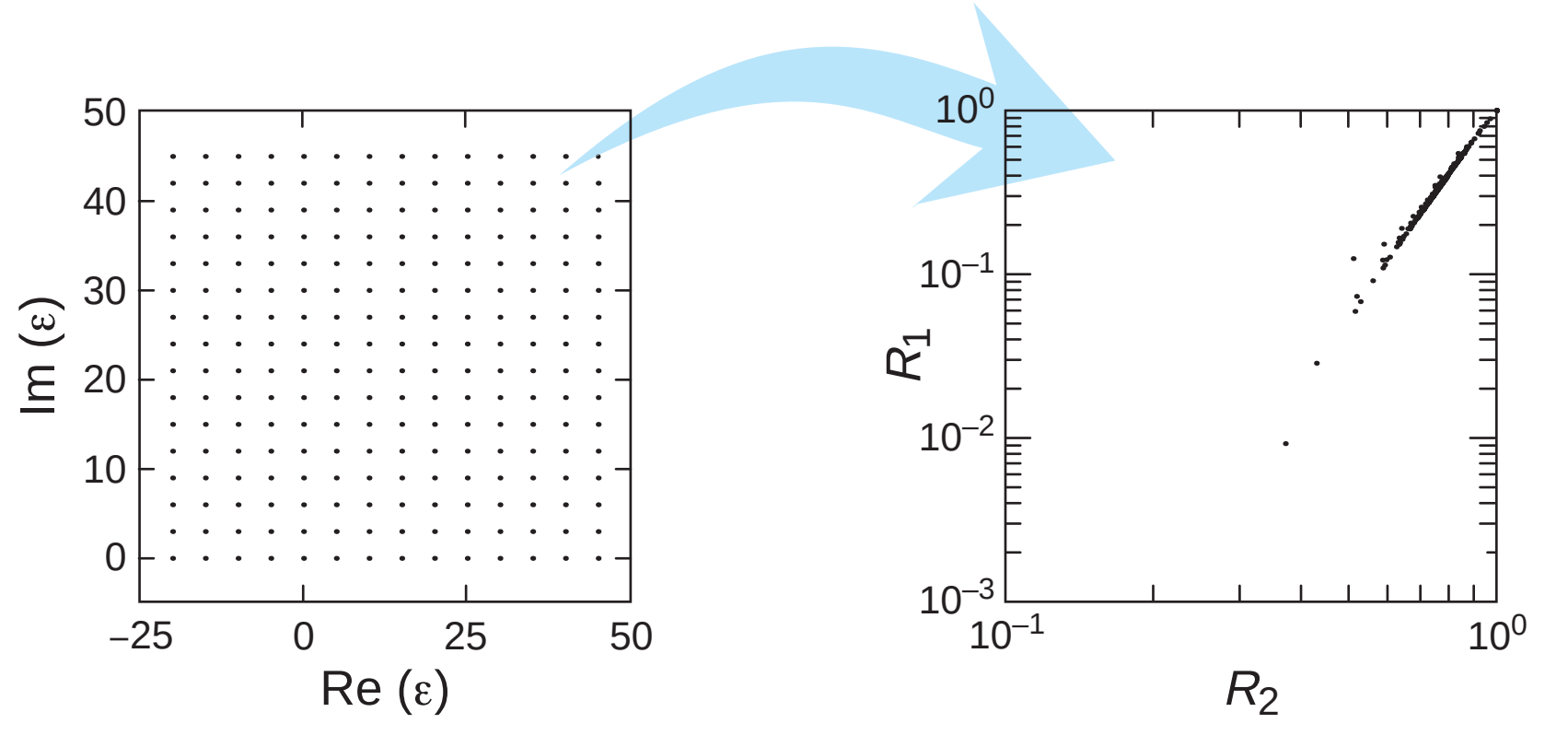
Technique

choice of angles



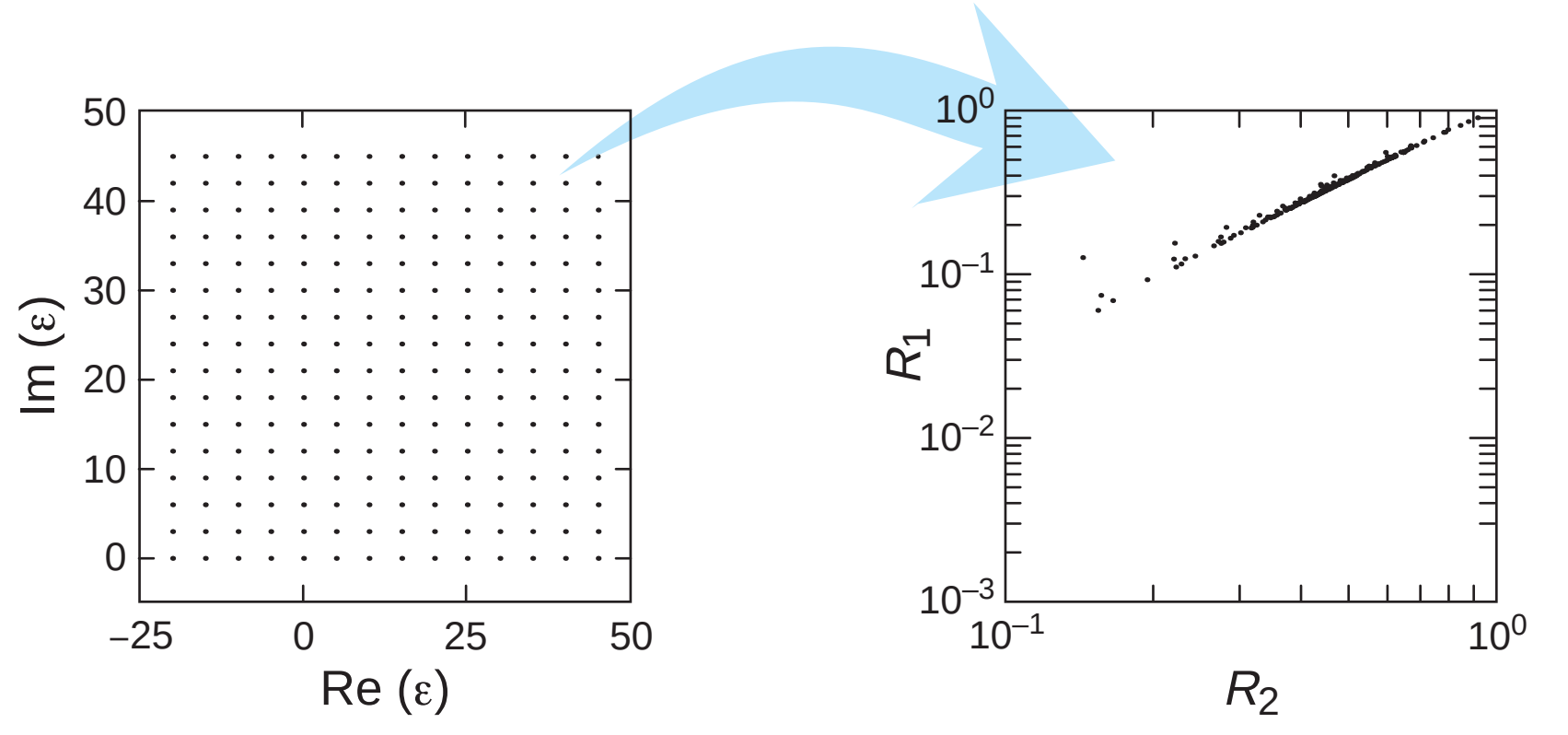
need numerical inversion

choice of angles



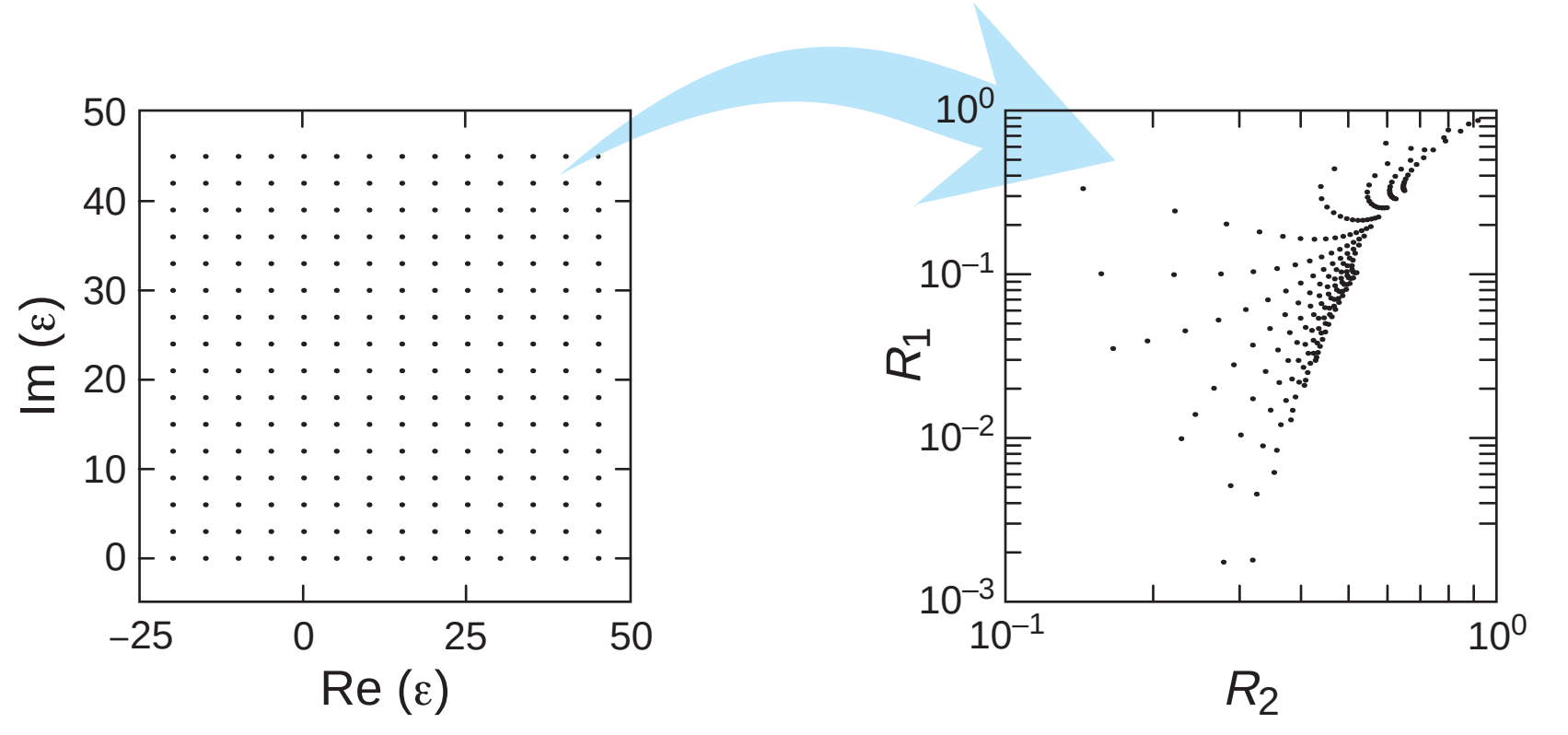
R_1 = 45° *p*-pol, R_2 = 45° *s*-pol

choice of angles



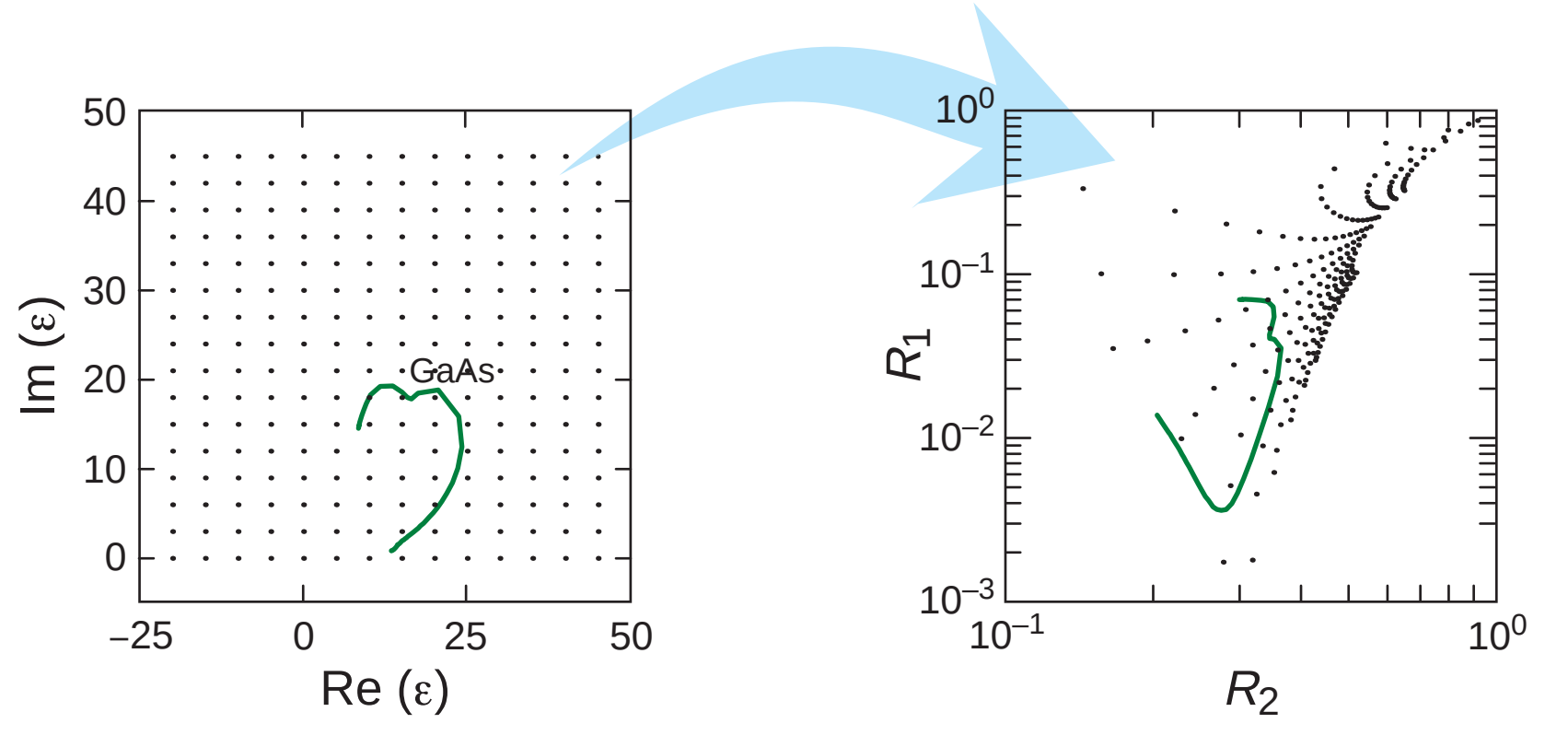
R_1 = 60° *p*-pol, R_2 = 45° *p*-pol

choice of angles



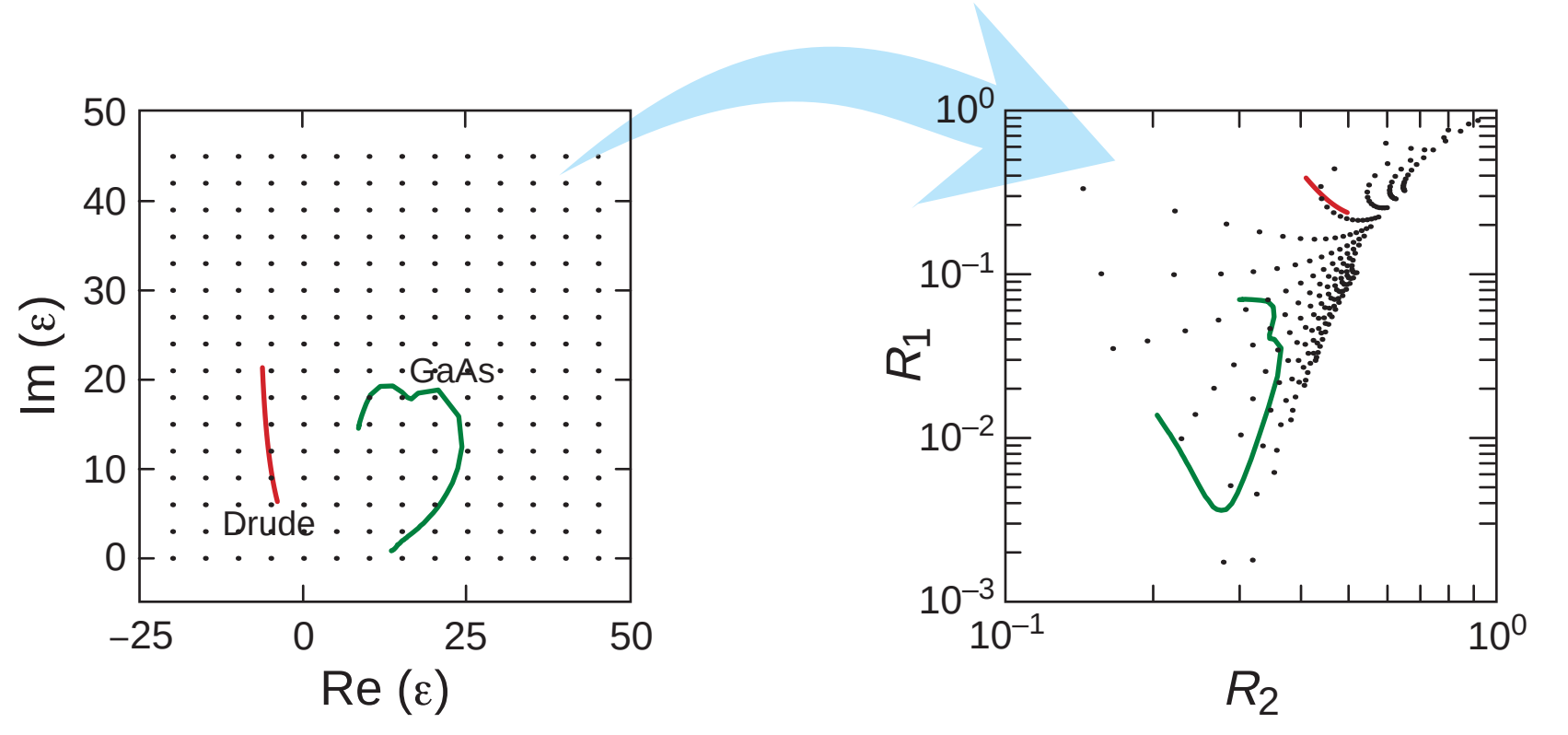
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

choice of angles



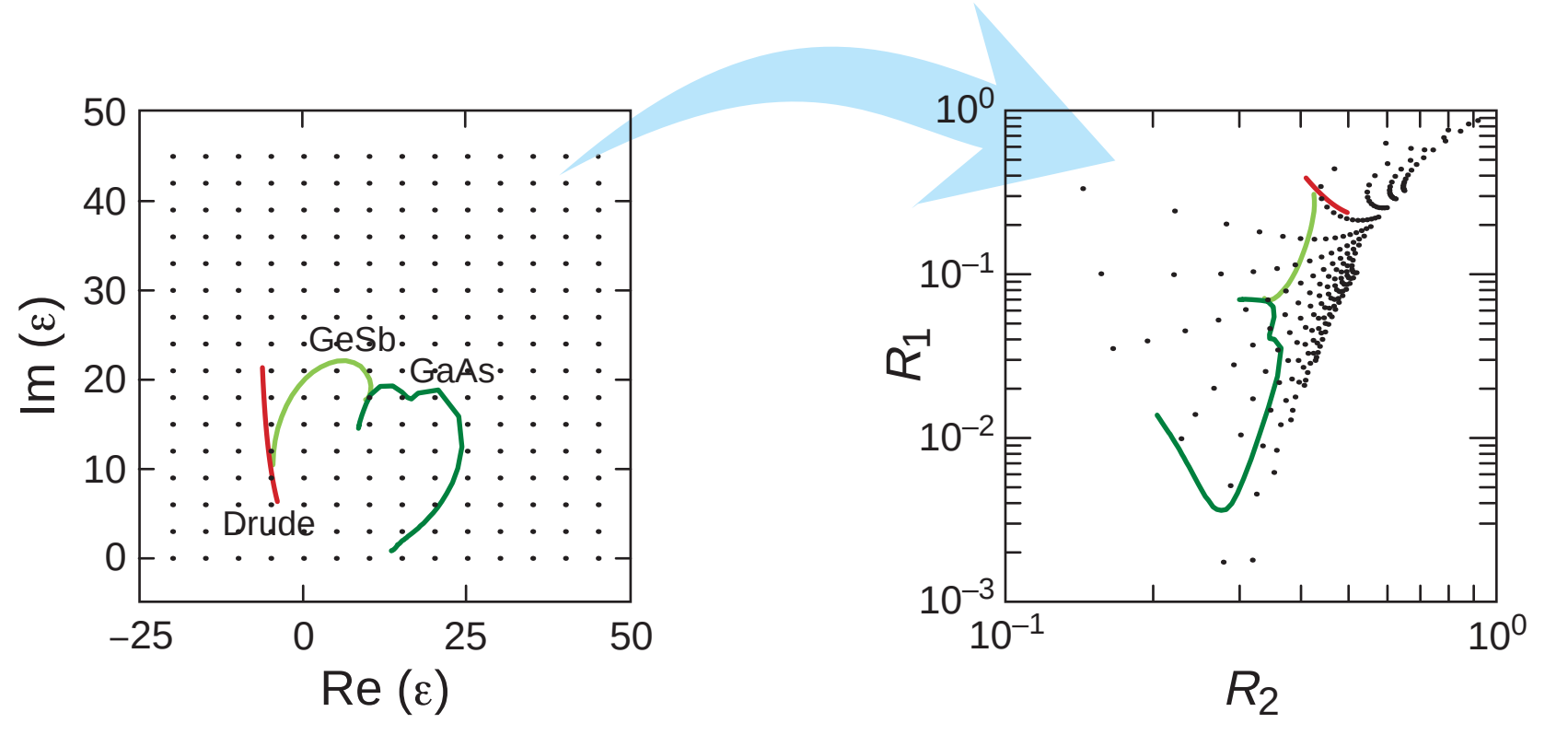
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

choice of angles



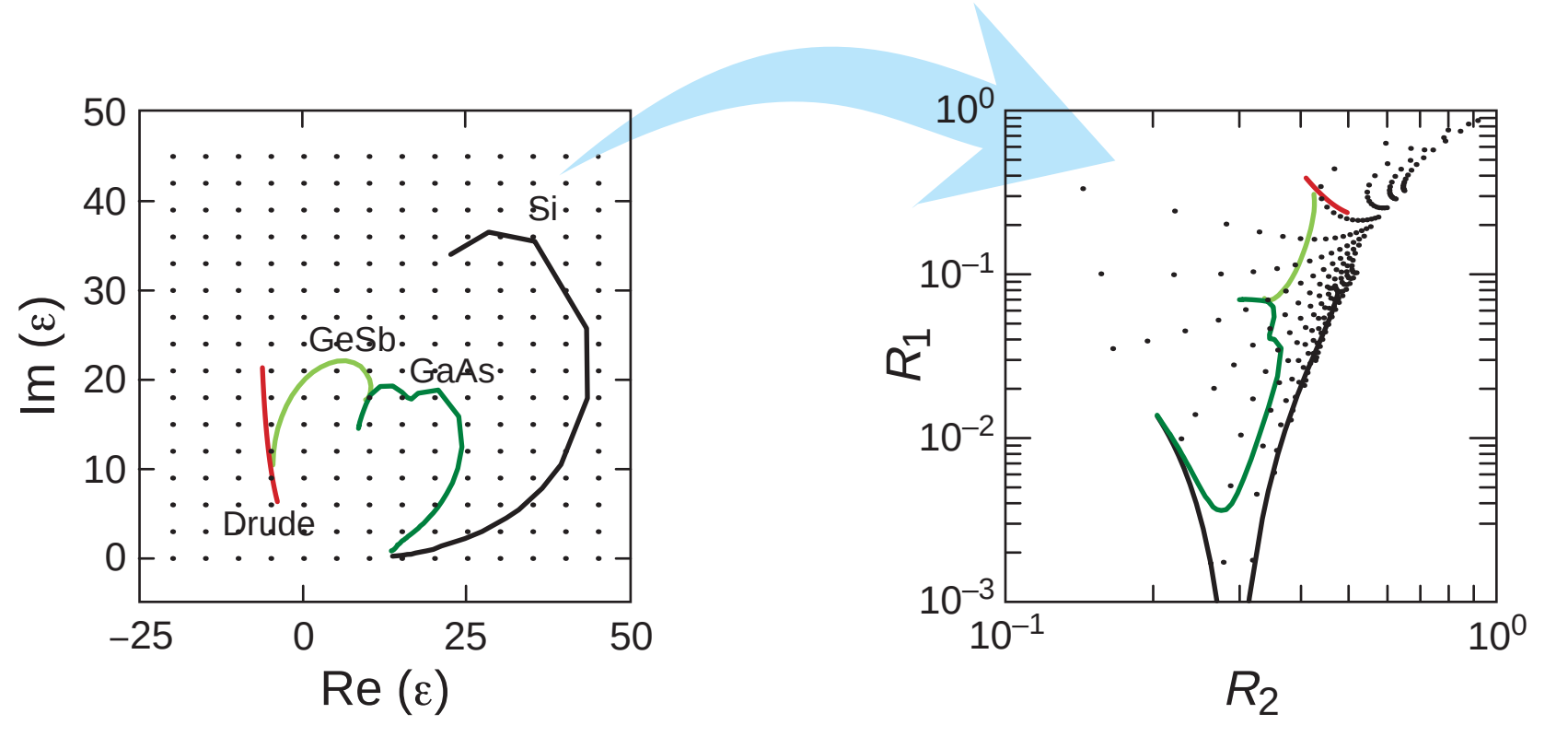
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

choice of angles



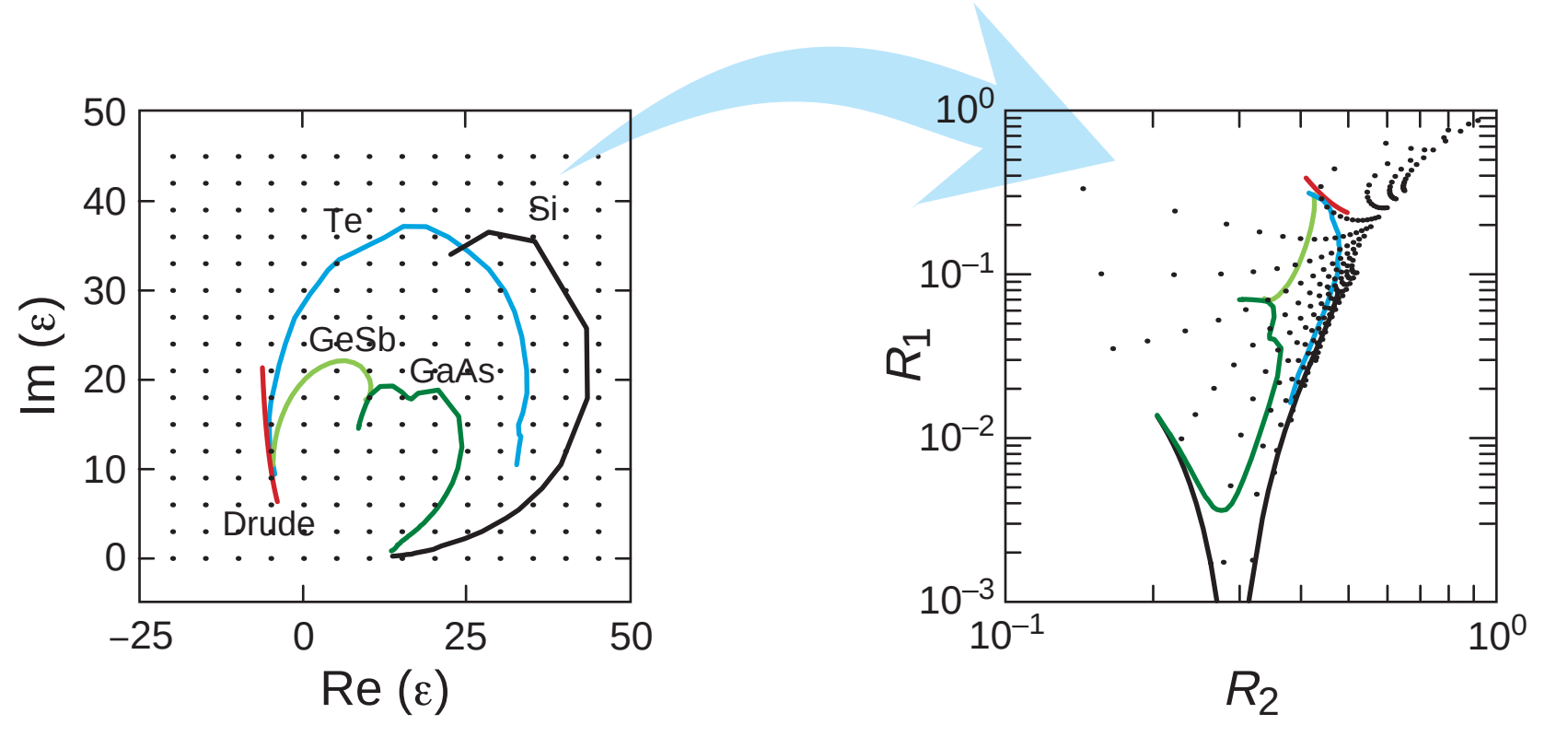
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

choice of angles



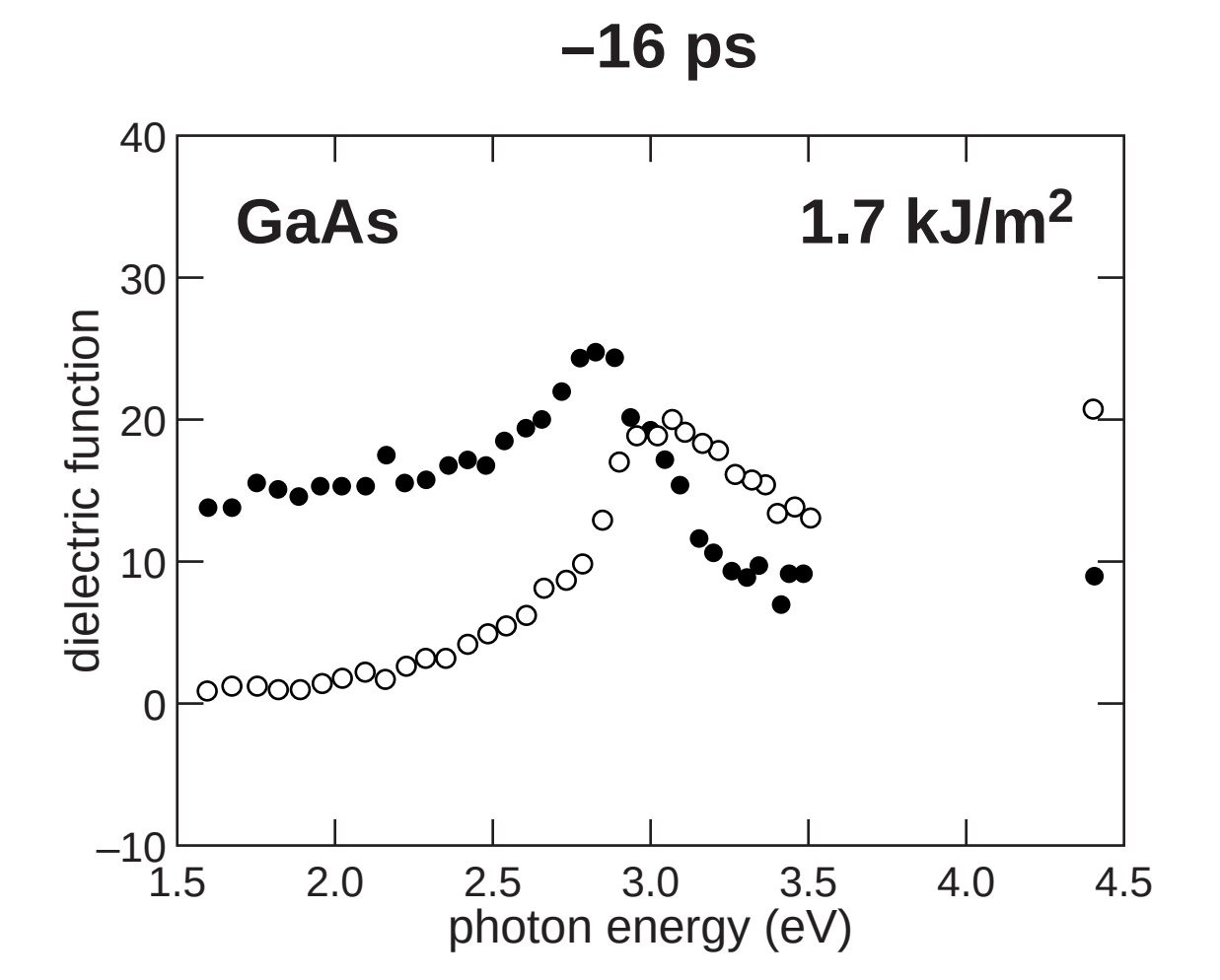
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

choice of angles



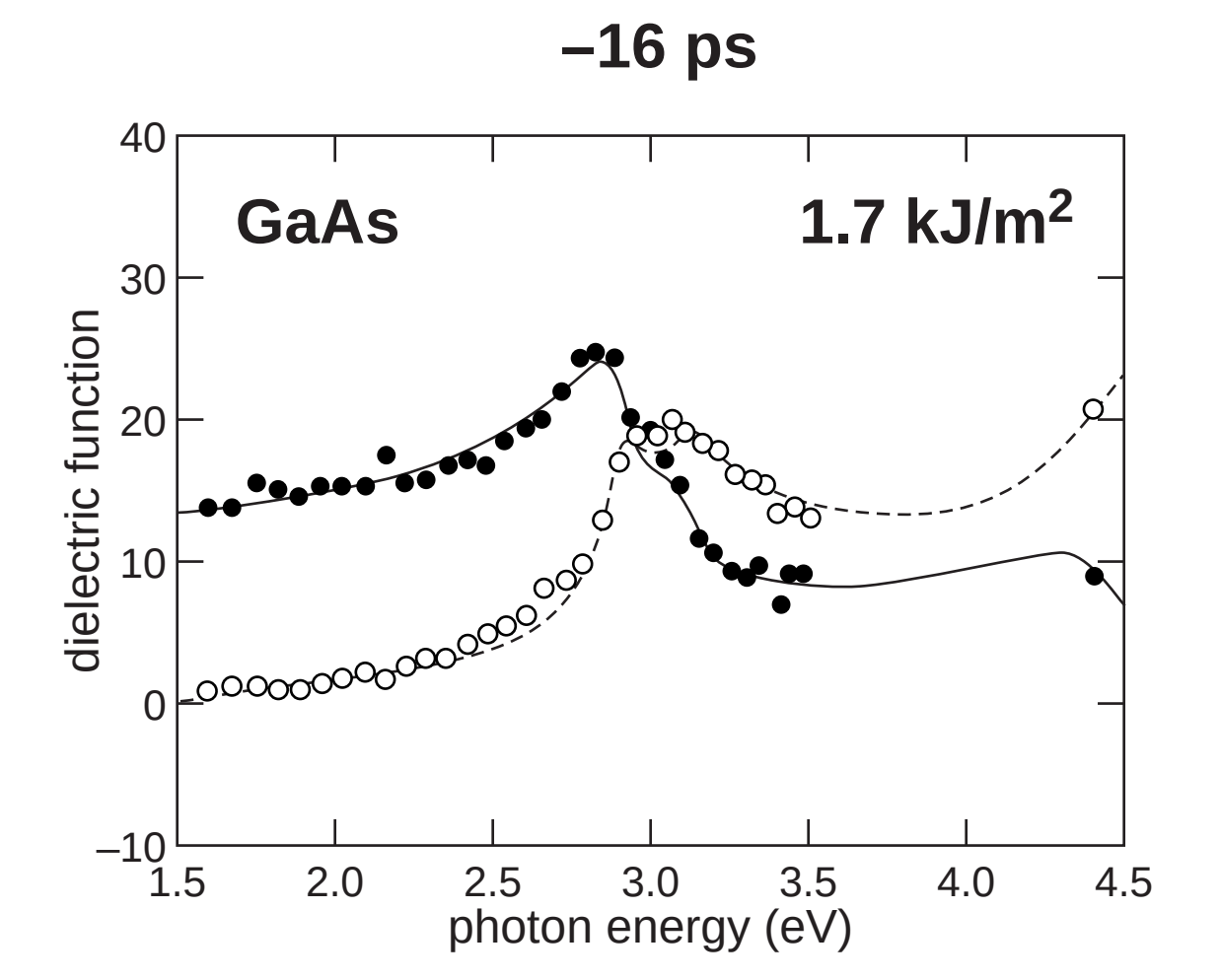
R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

Technique



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

Technique



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

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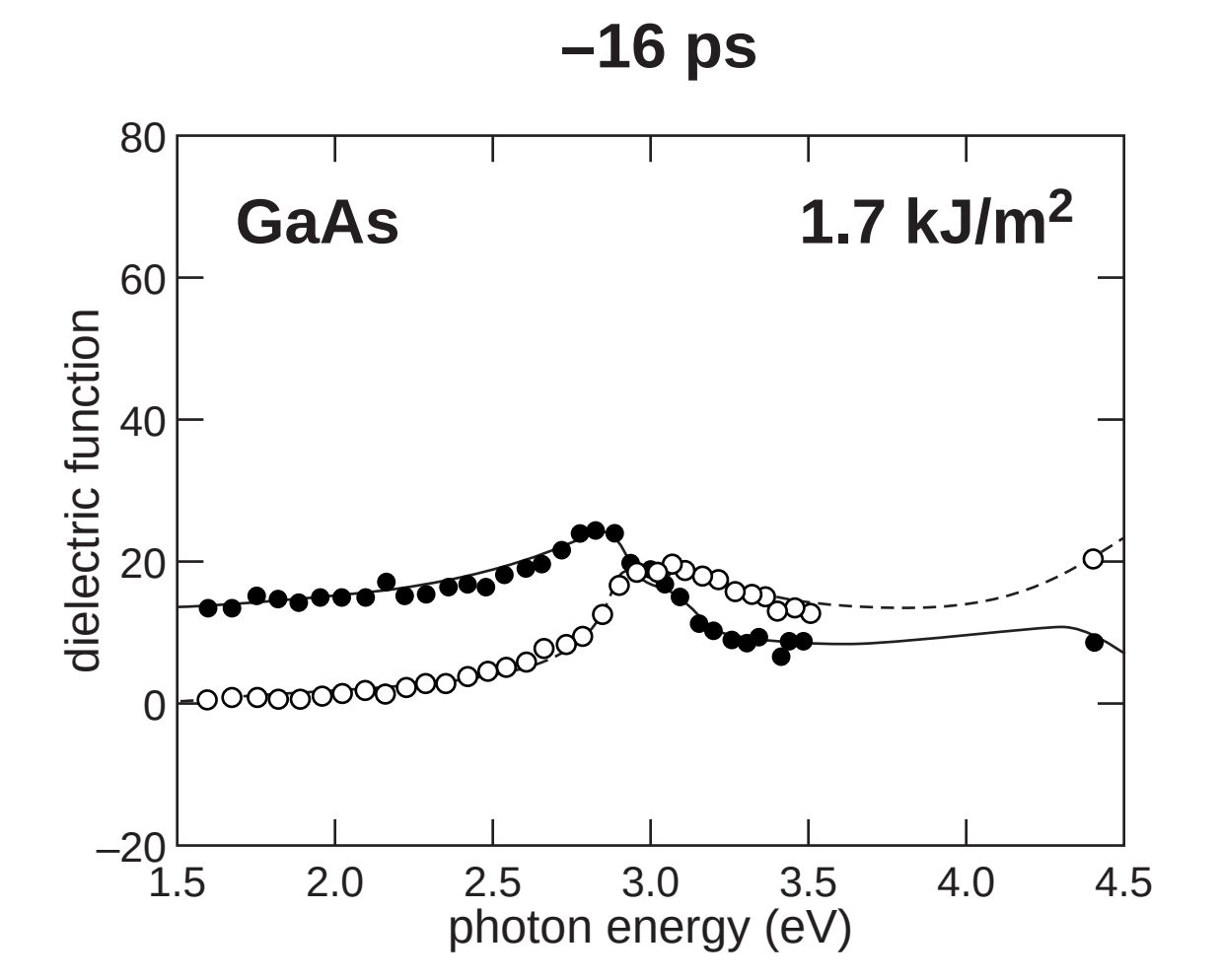

Phys. Rev. Lett. **80, 185 (1998)**

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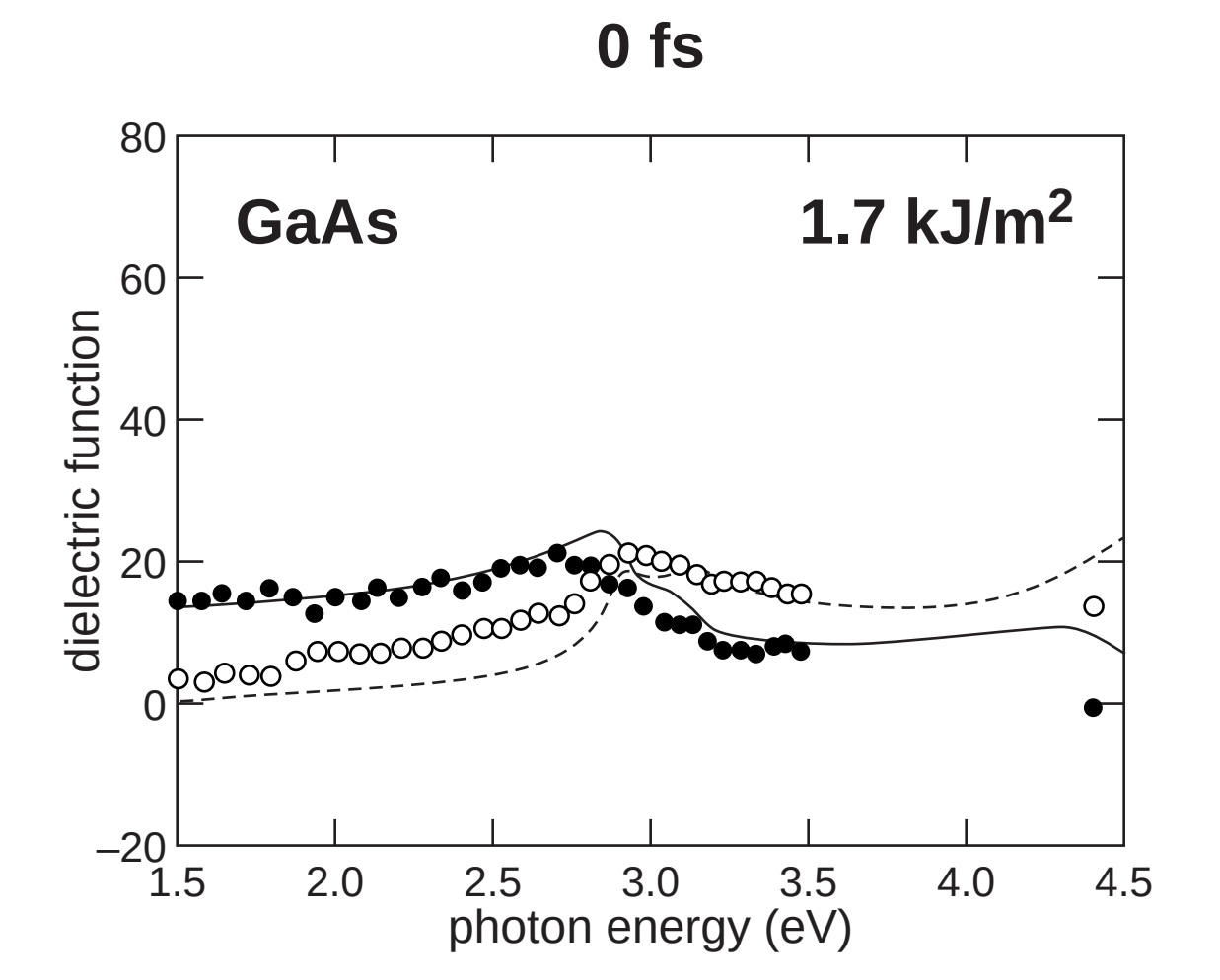

Phys. Rev. Lett. **80, 185 (1998)**

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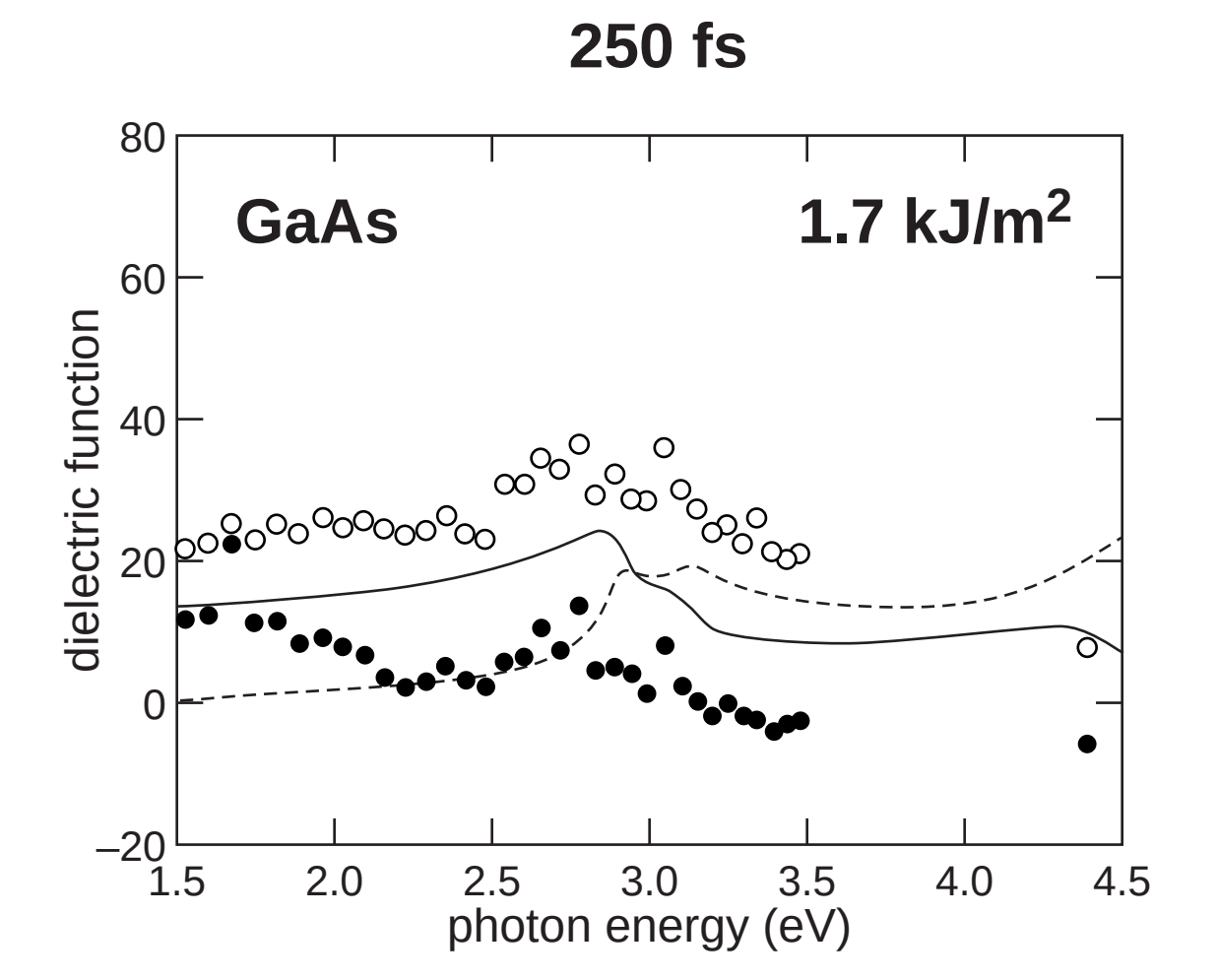

Phys. Rev. Lett. **80, 185 (1998)**

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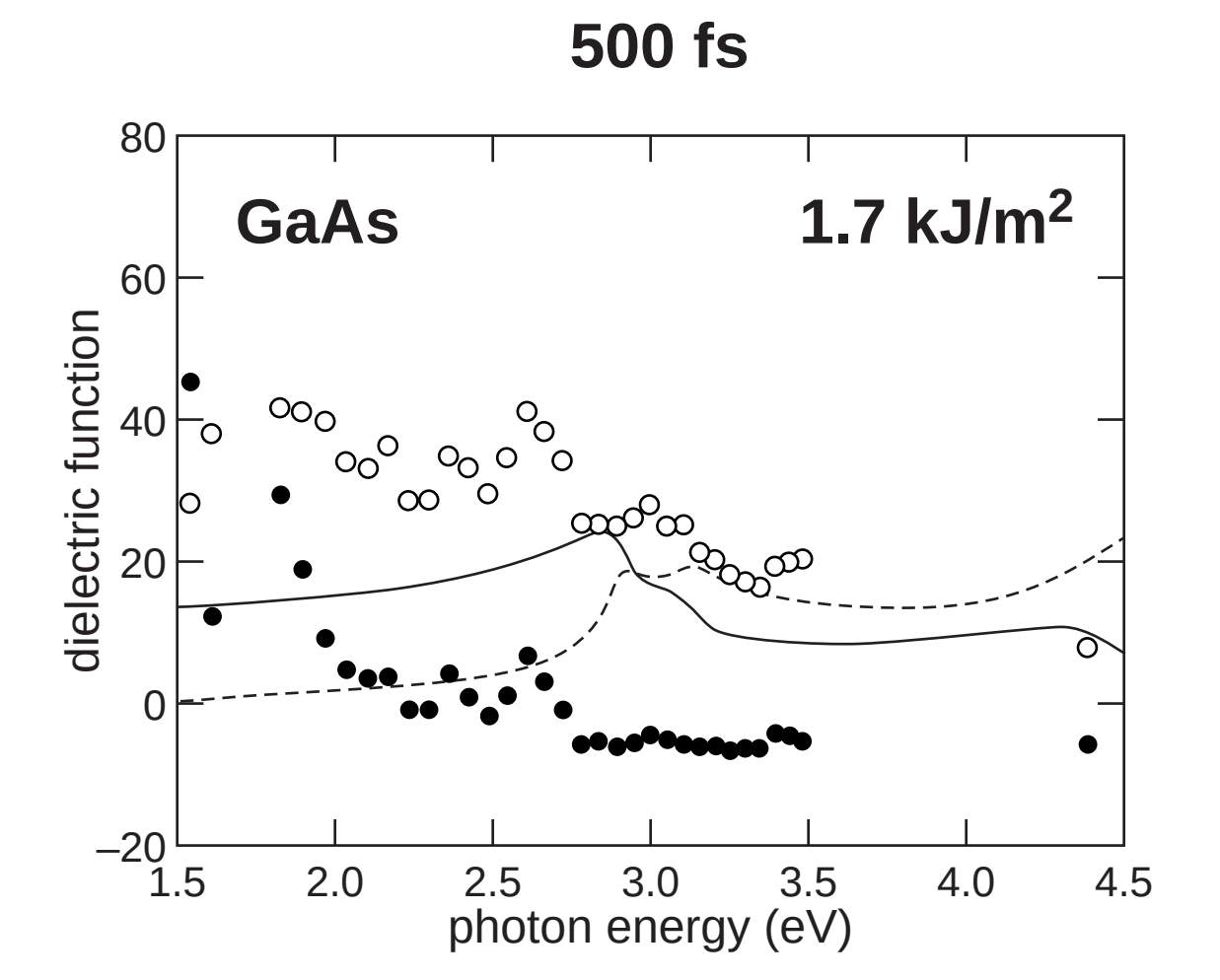

Phys. Rev. Lett. **80, 185 (1998)**

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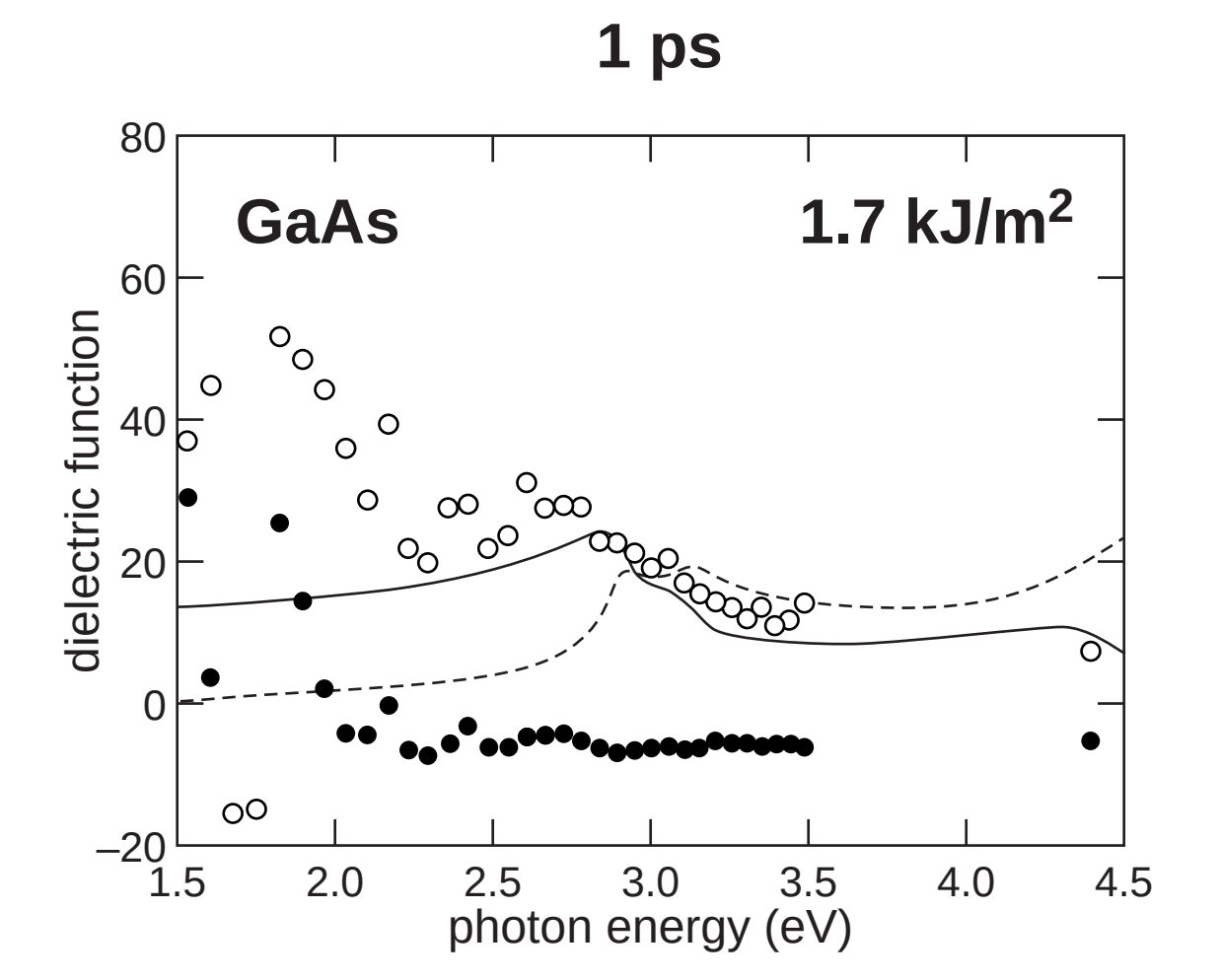
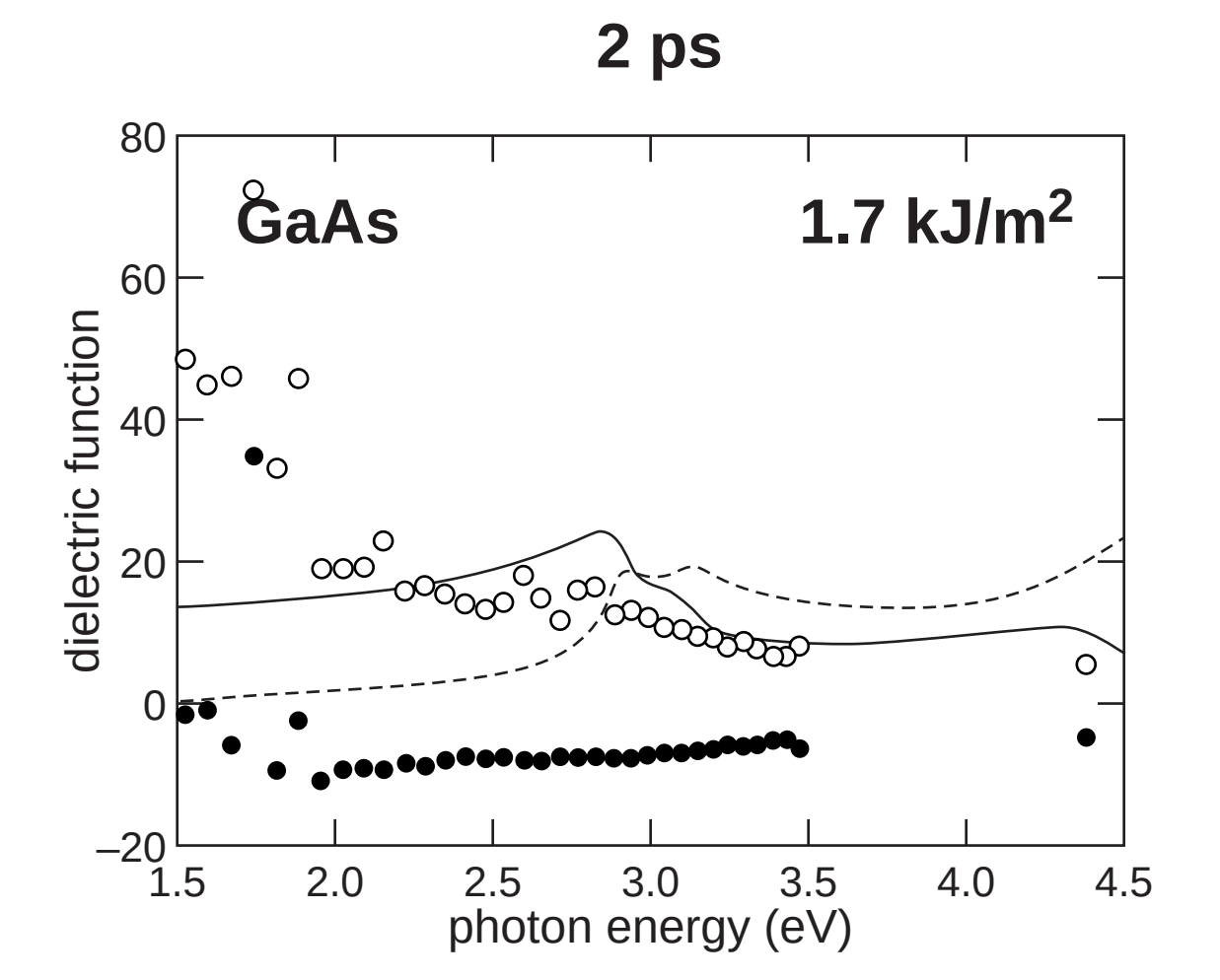


Phys. Rev. Lett. **80, 185 (1998)**

Technique



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

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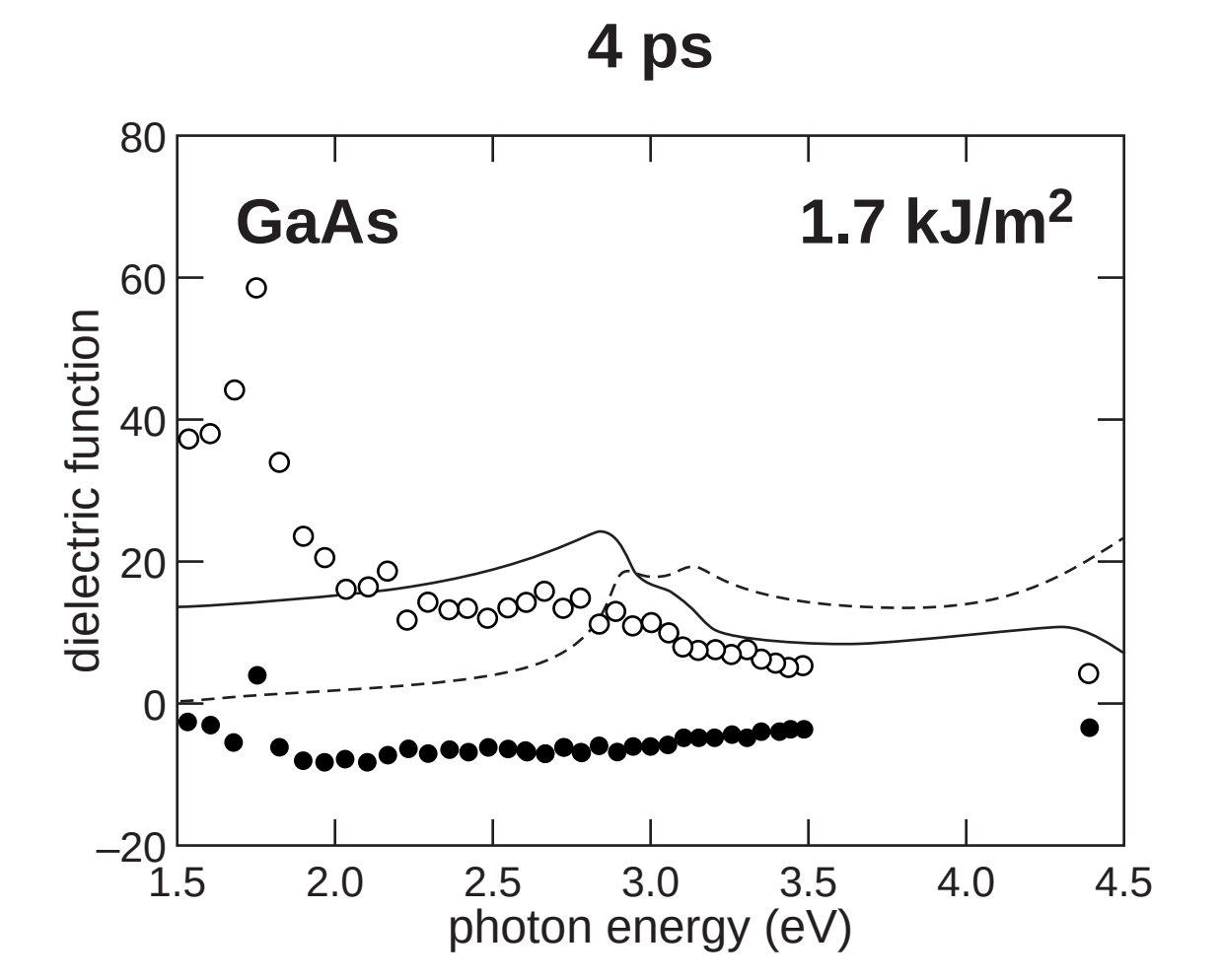
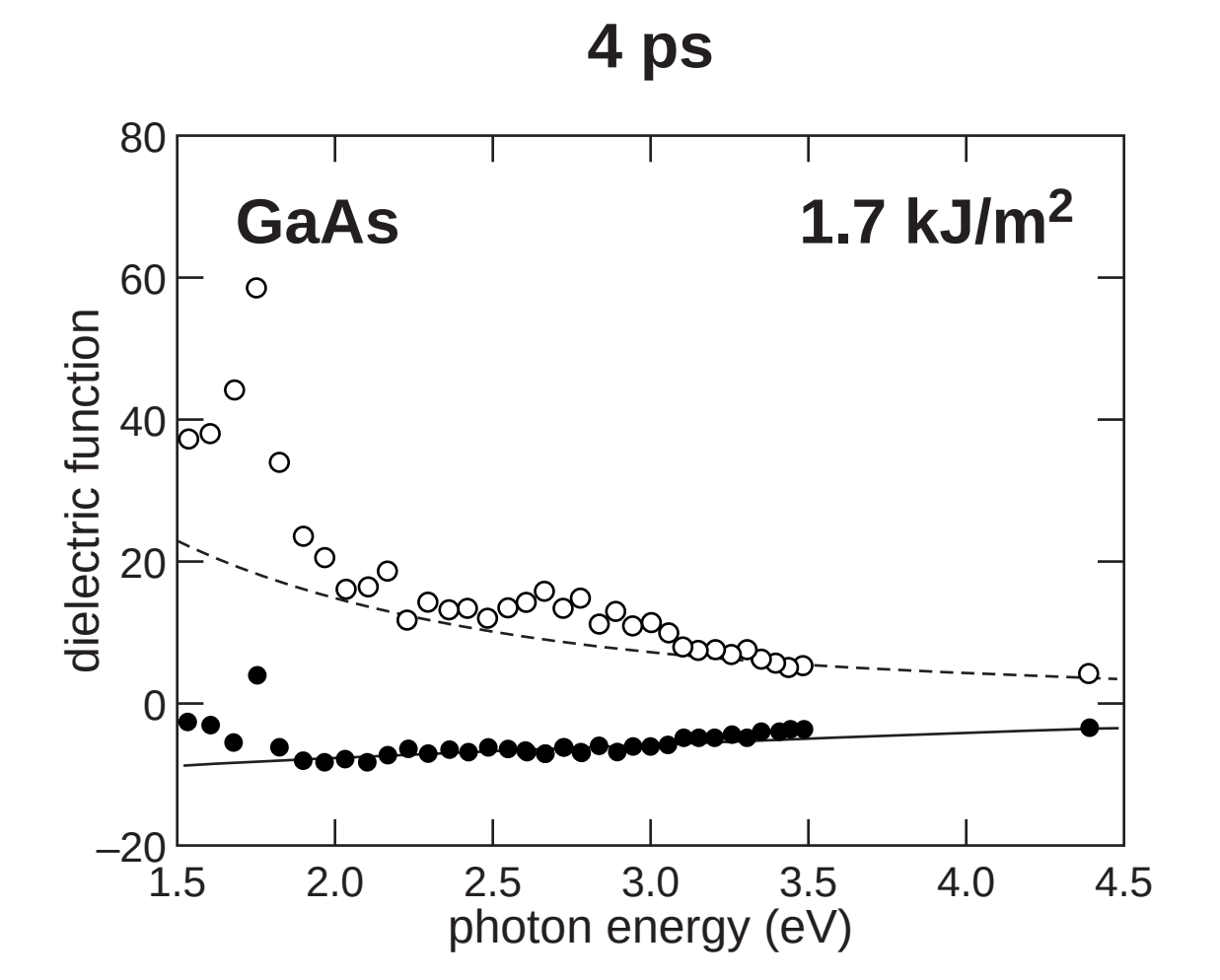


Phys. Rev. Lett. **80, 185 (1998)**

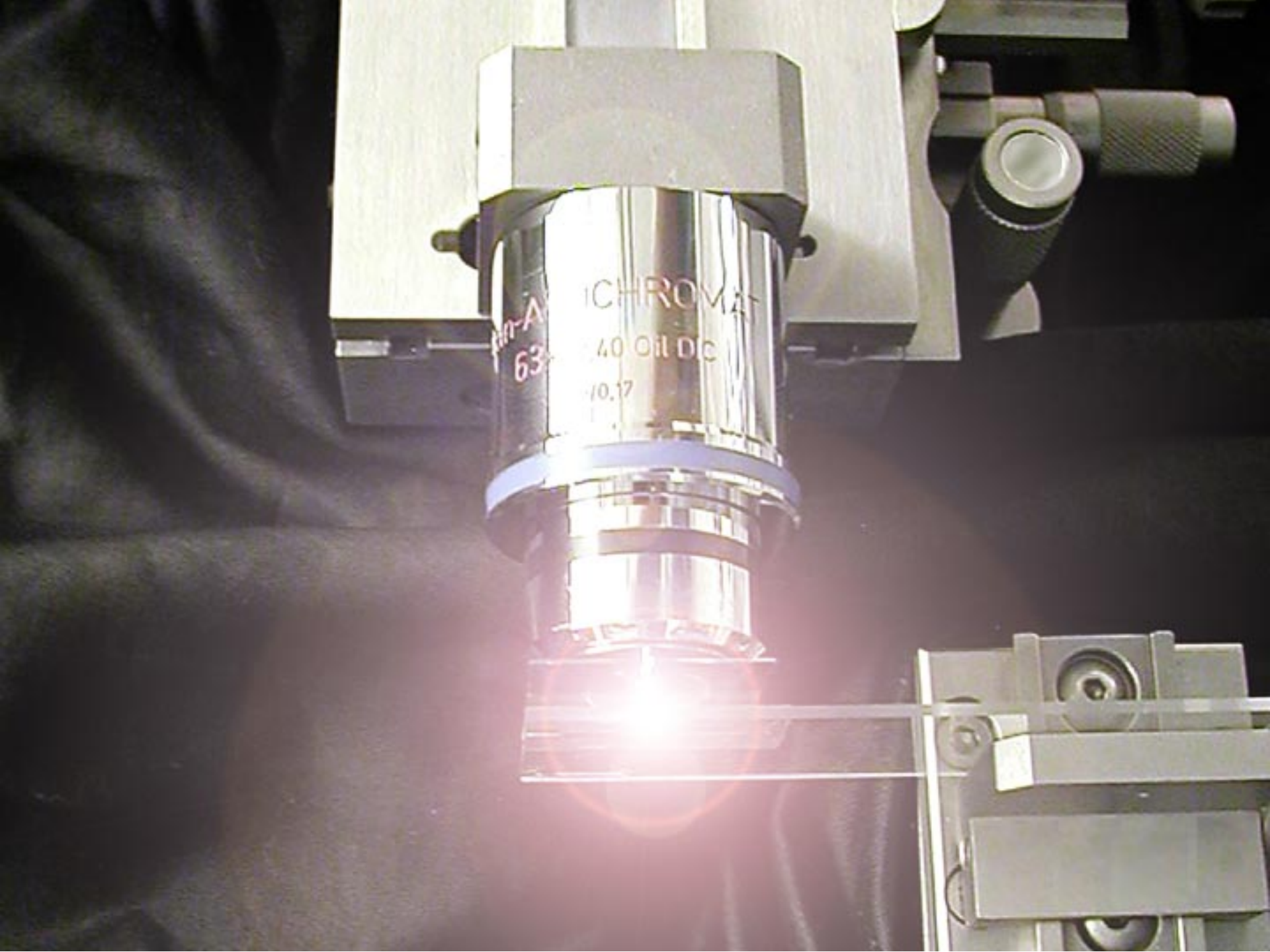
Technique



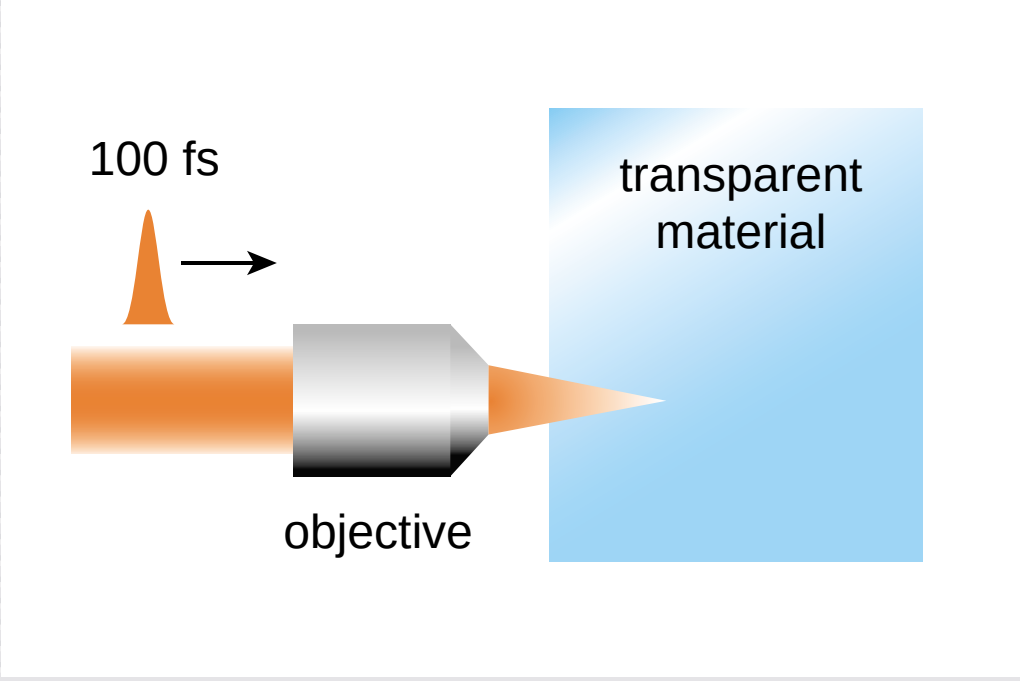
Phys. Rev. Lett. **80, 185 (1998)**

Technique

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

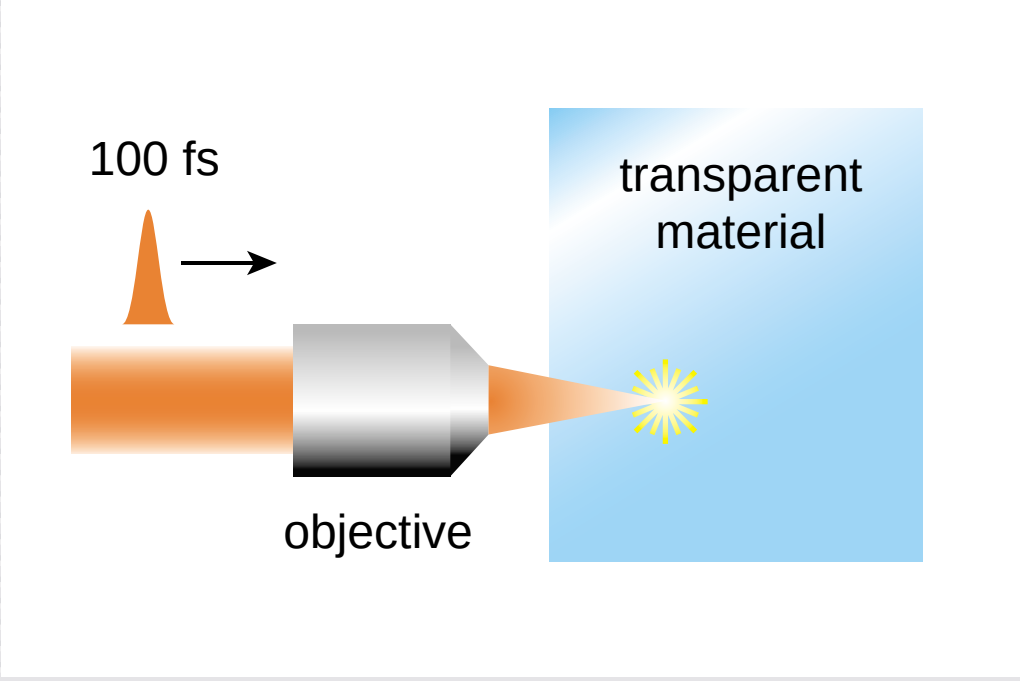

CHROMAT
40 Oil DC
0.17

Processing with fs pulses



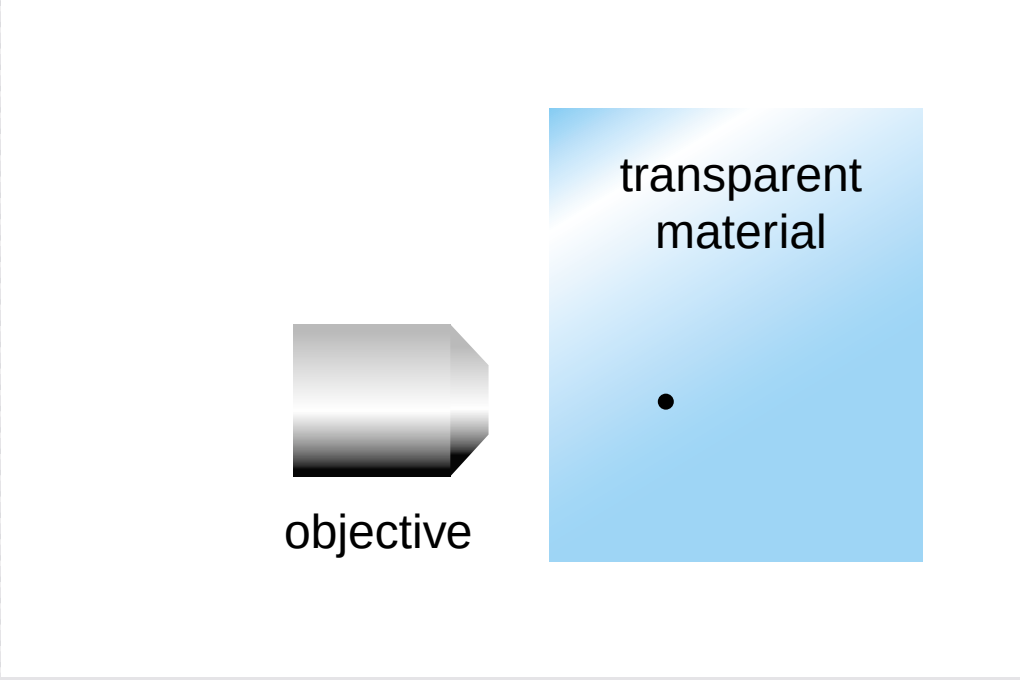
high intensity at focus…

Processing with fs pulses



… causes nonlinear ionization…

Processing with fs pulses



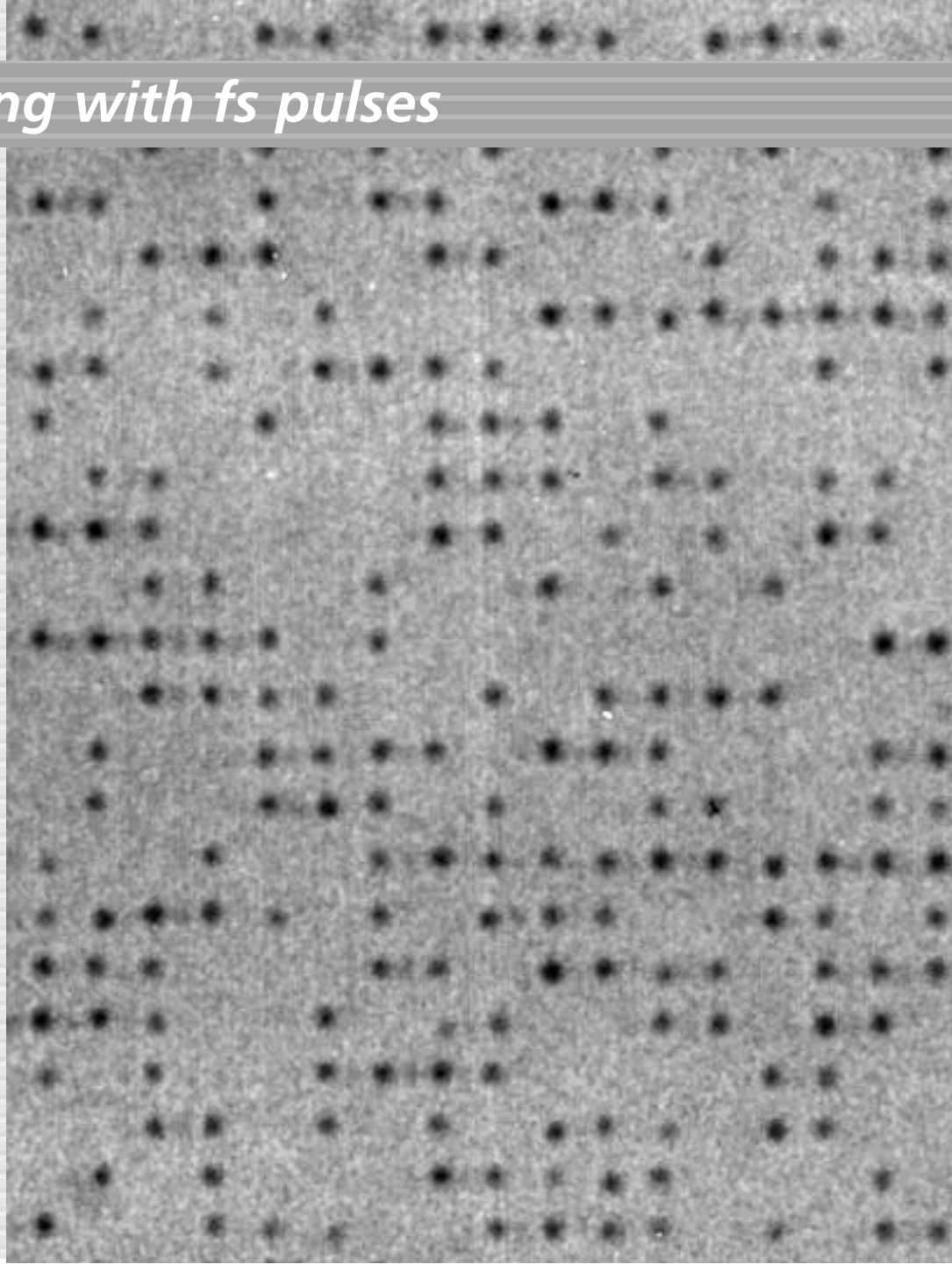
and ‘microexplosion’ causes microscopic damage

Processing with fs pulses

2 x 2 μm array

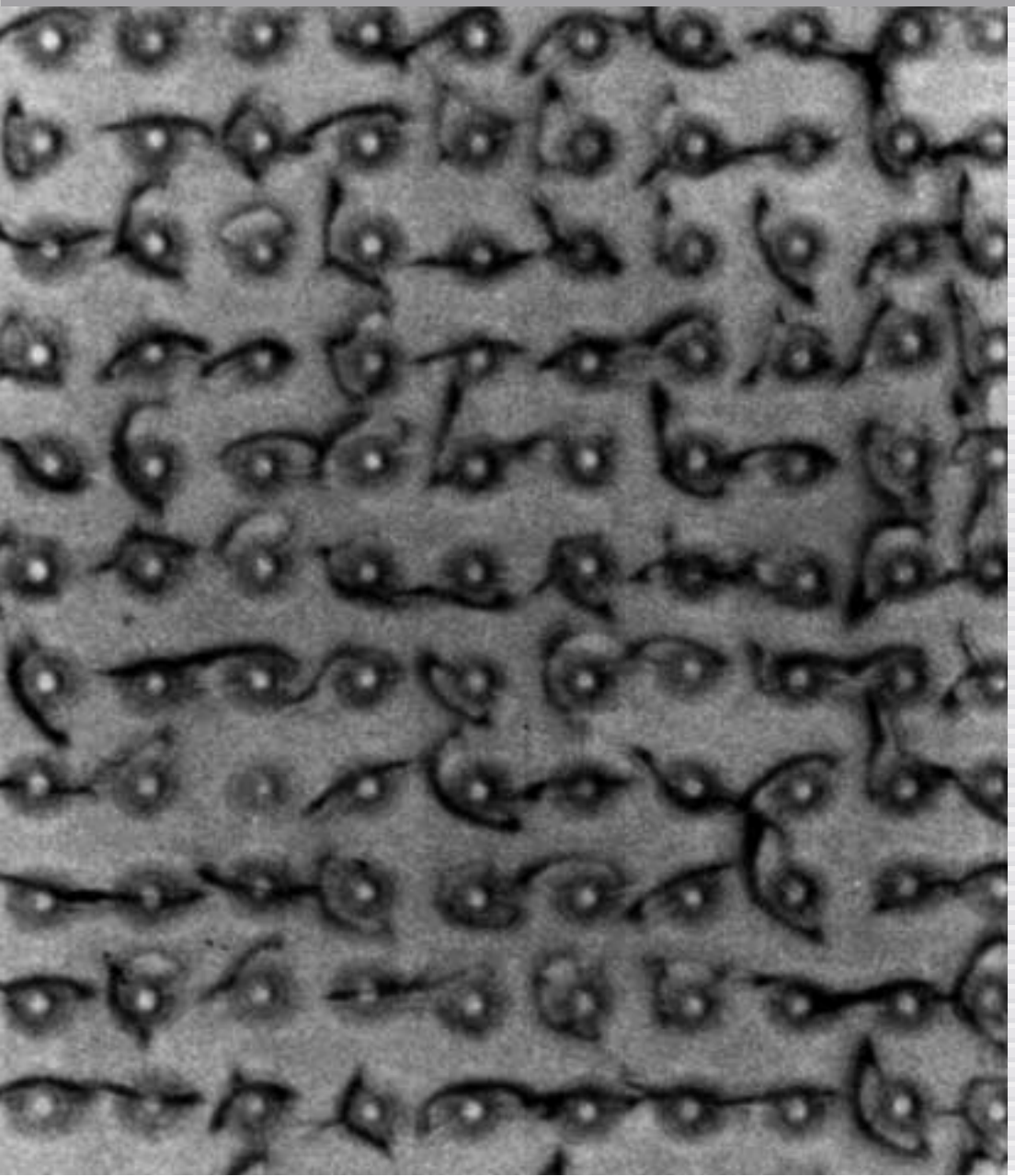
fused silica, 0.65 NA

0.5 μJ, 100 fs, 800 nm

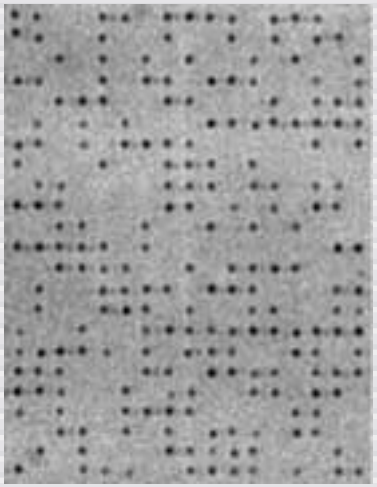


***Opt. Lett.* 21, 2023 (1996)**

Processing with fs pulses

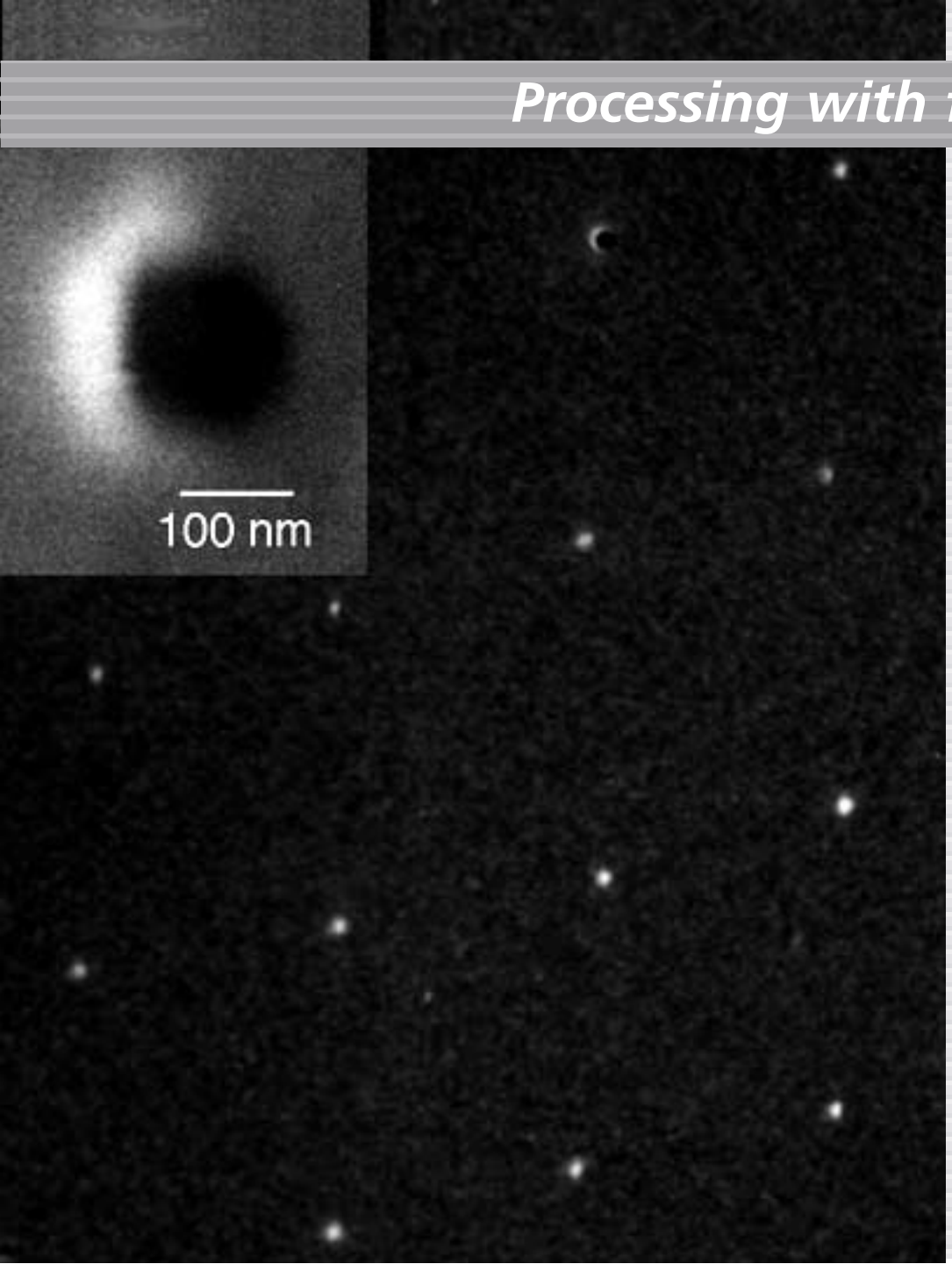


200 ps
9 µJ



100 fs
0.5 µJ

Processing with fs pulses



5 x 5 μm array

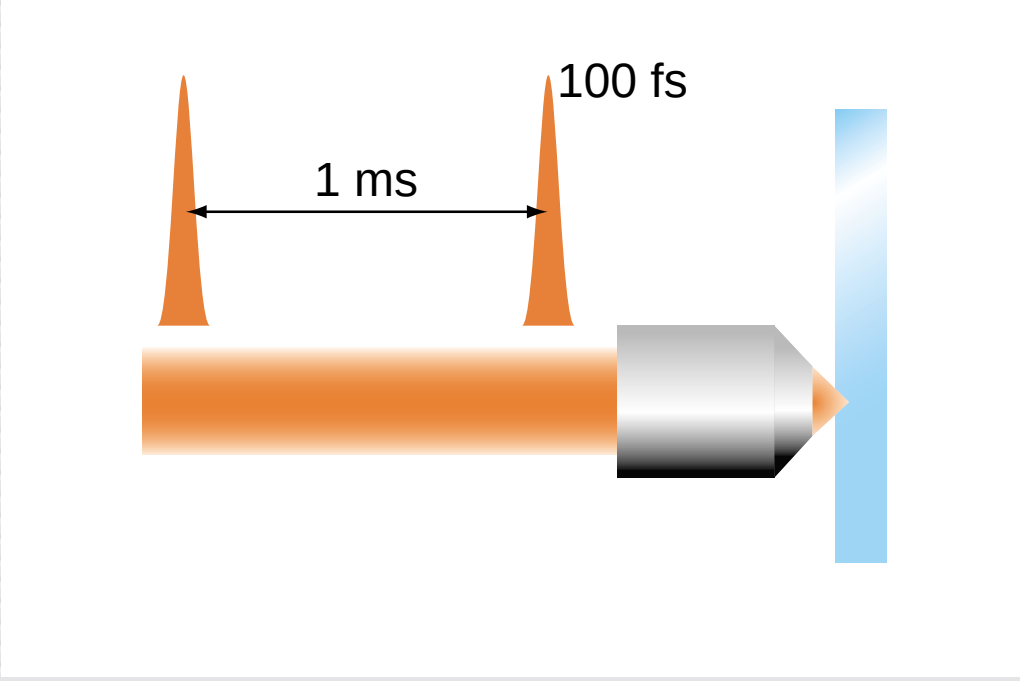
fused silica, 0.65 NA

0.5 μJ, 100 fs, 800 nm

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

Low-energy processing

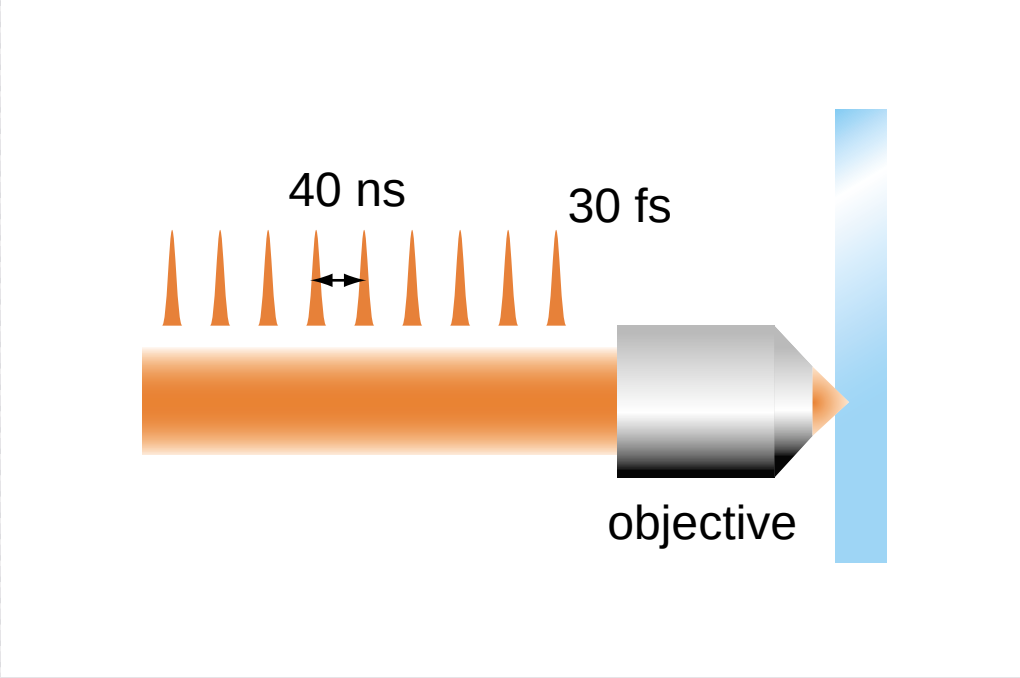
amplified laser



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

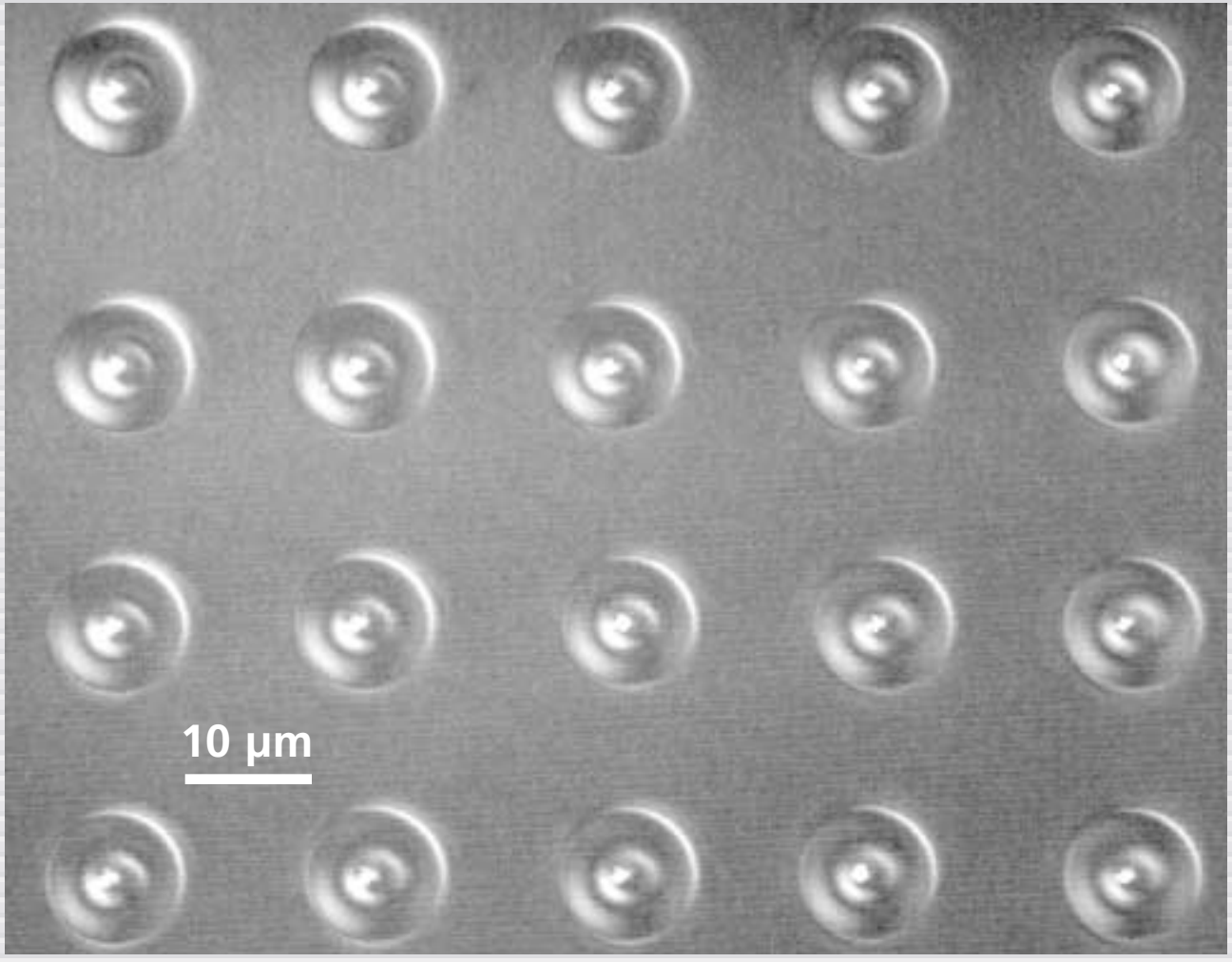
Low-energy processing

long-cavity Ti:sapphire oscillator

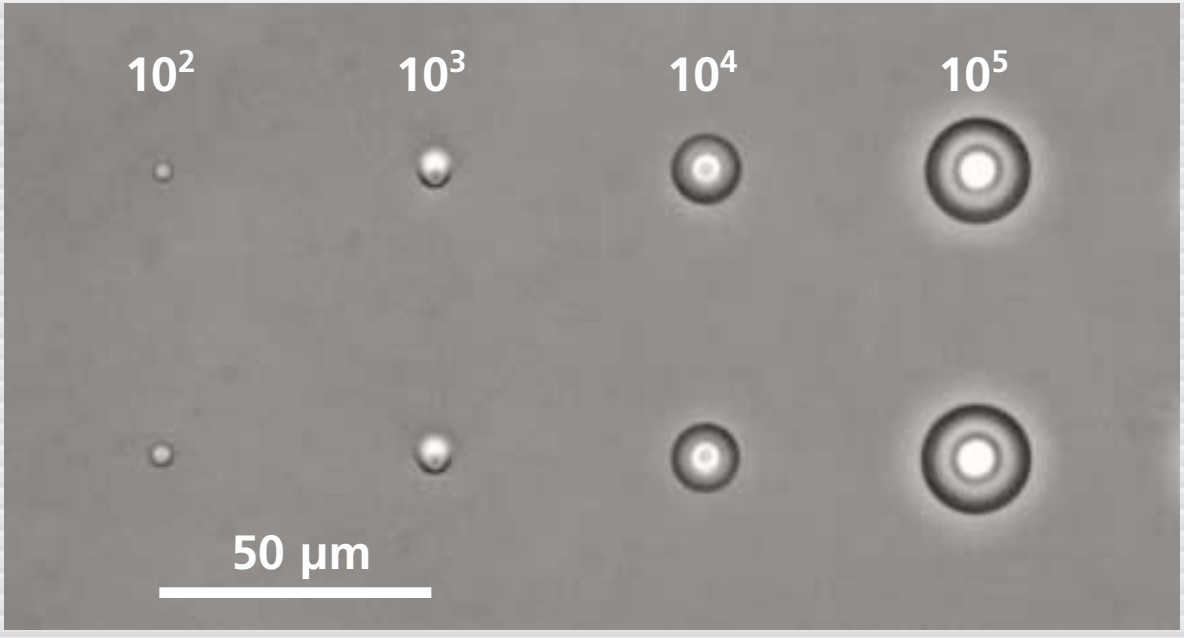


heat-diffusion time: $\tau_{diff} \approx 1\ \mu s$

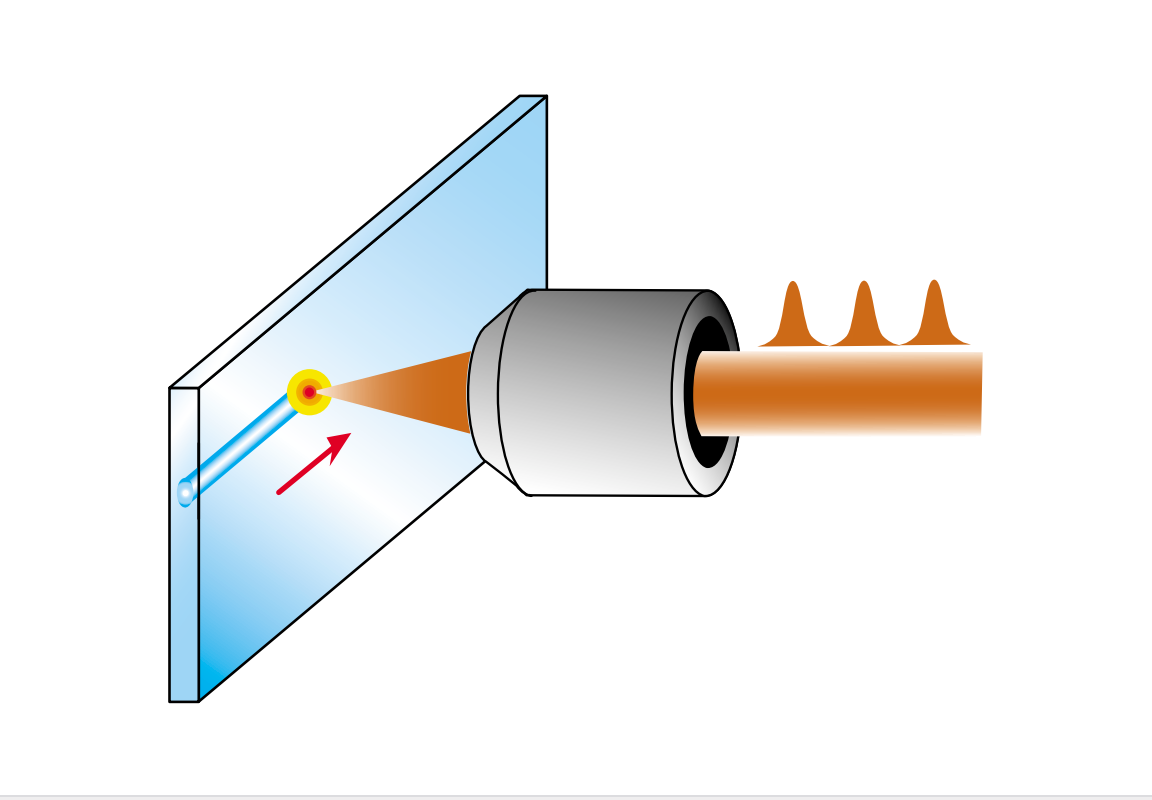
Low-energy processing



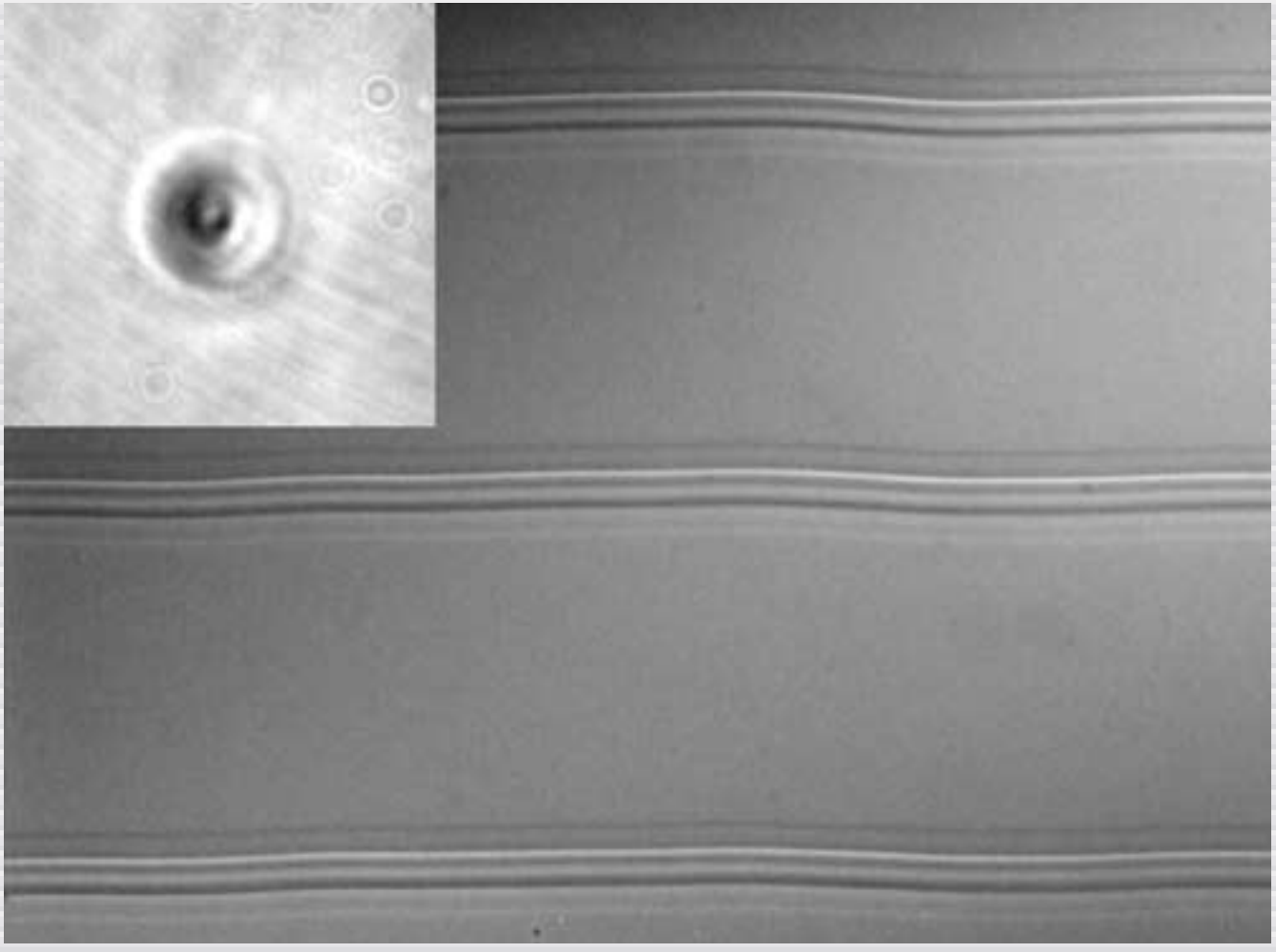
Low-energy processing



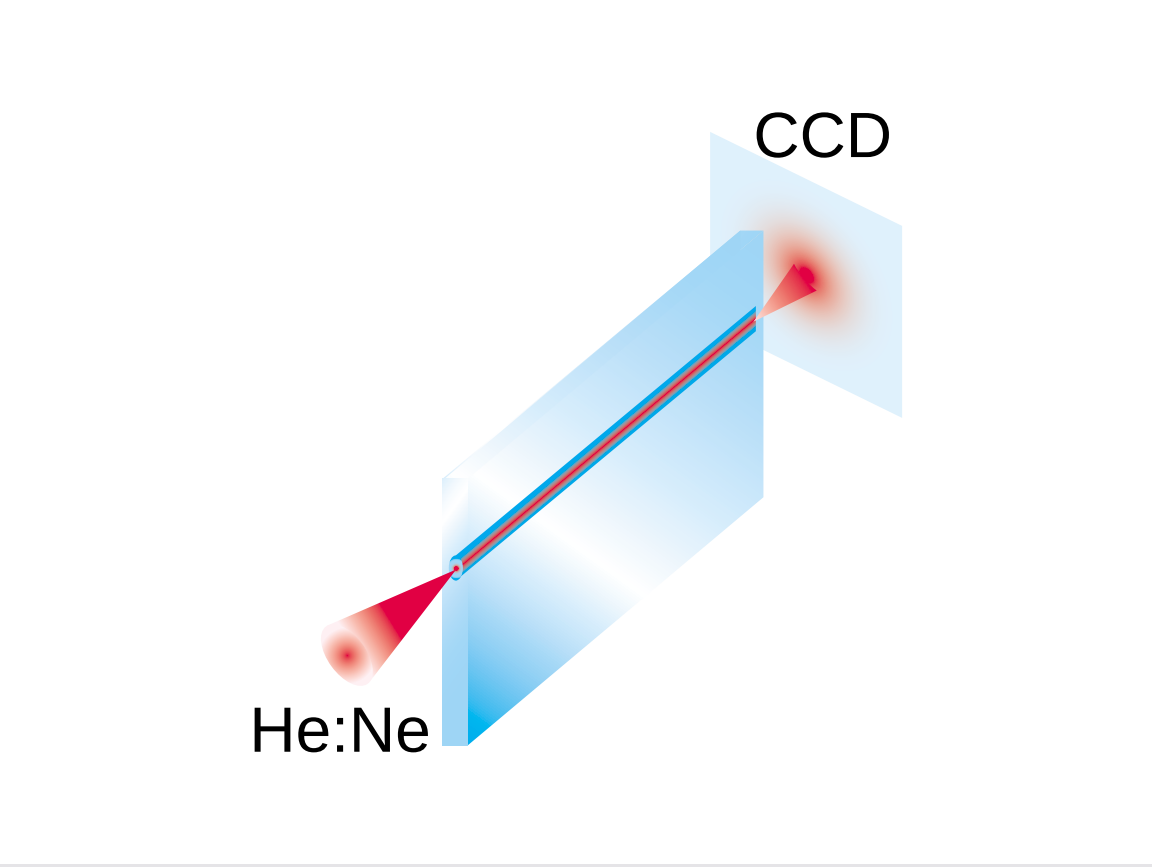
waveguide machining



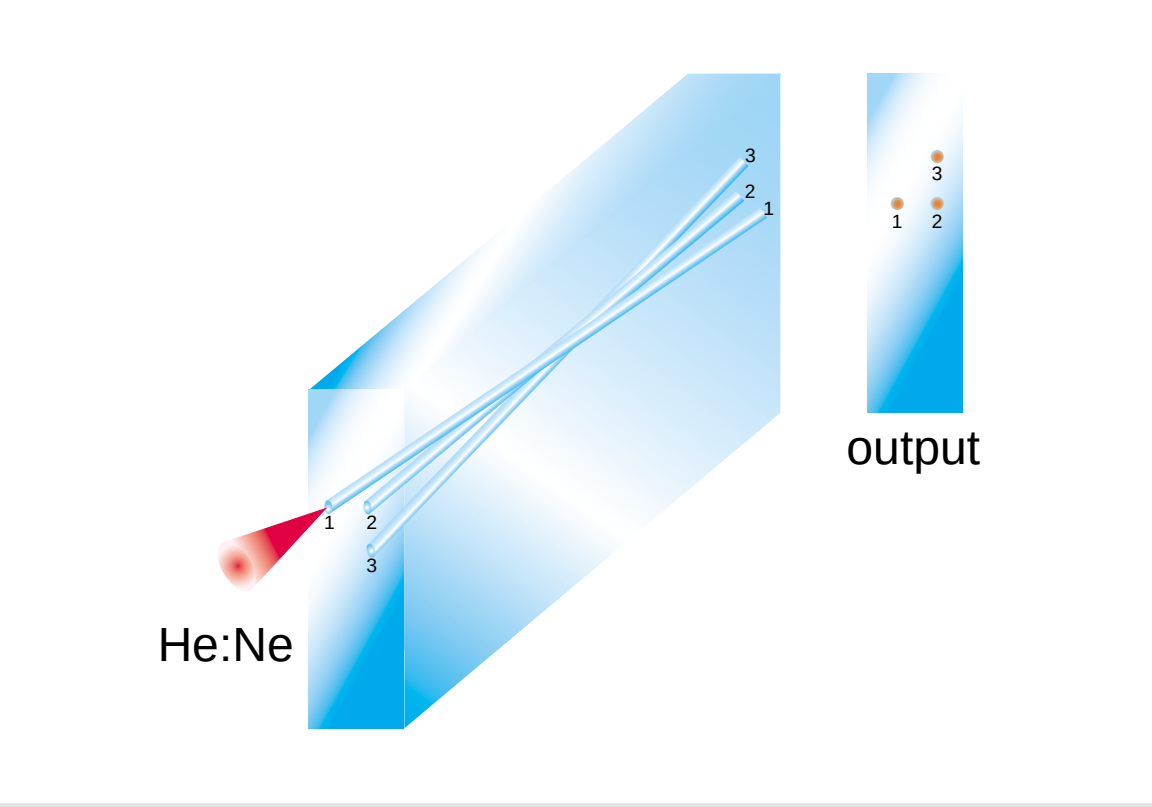
waveguide machining



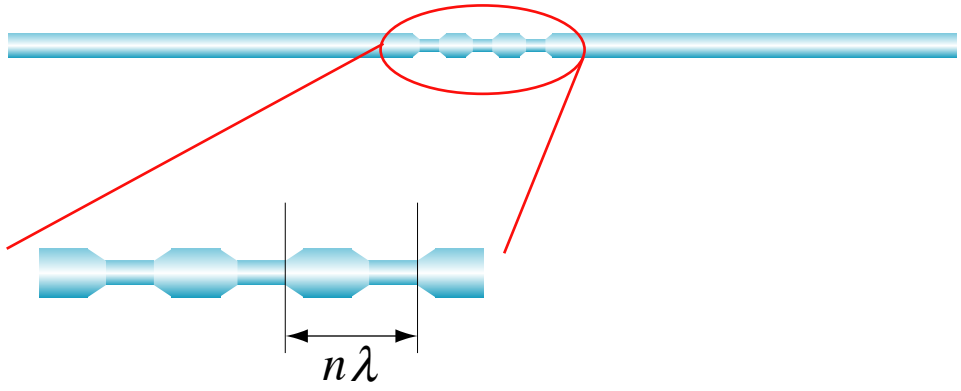
waveguide mode analysis



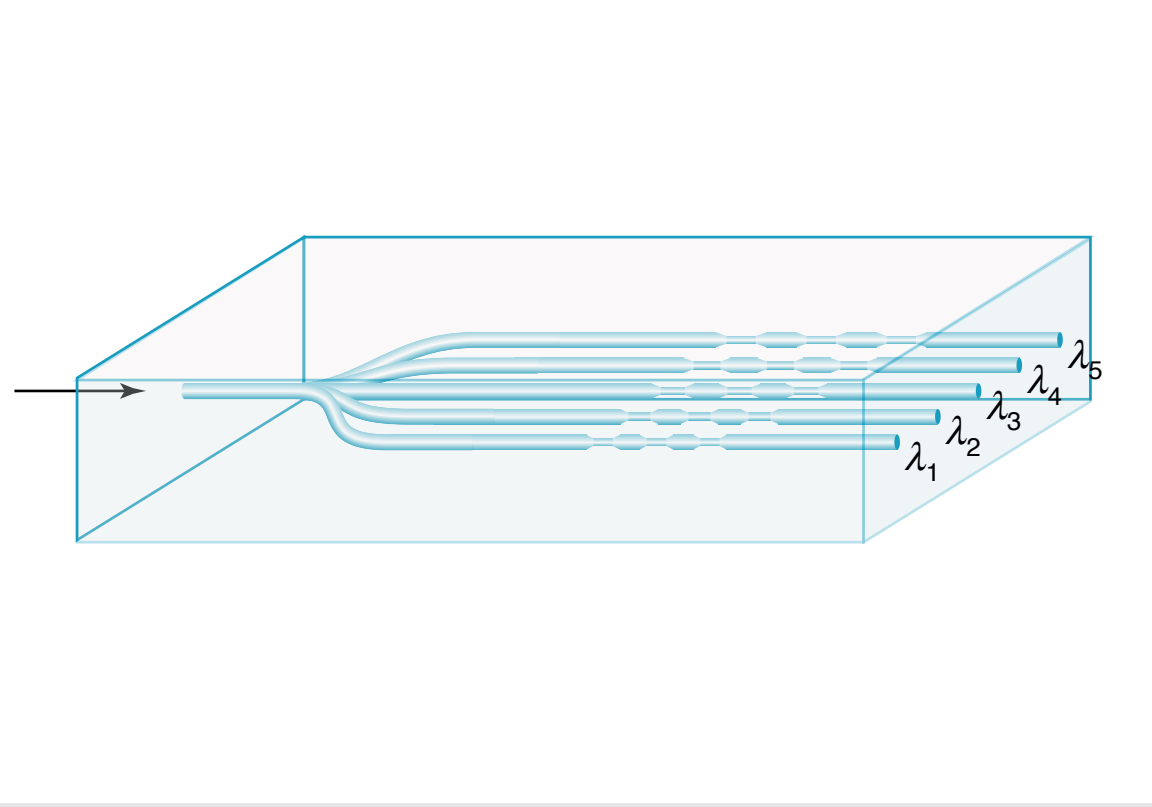
3D wave splitter



Bragg grating



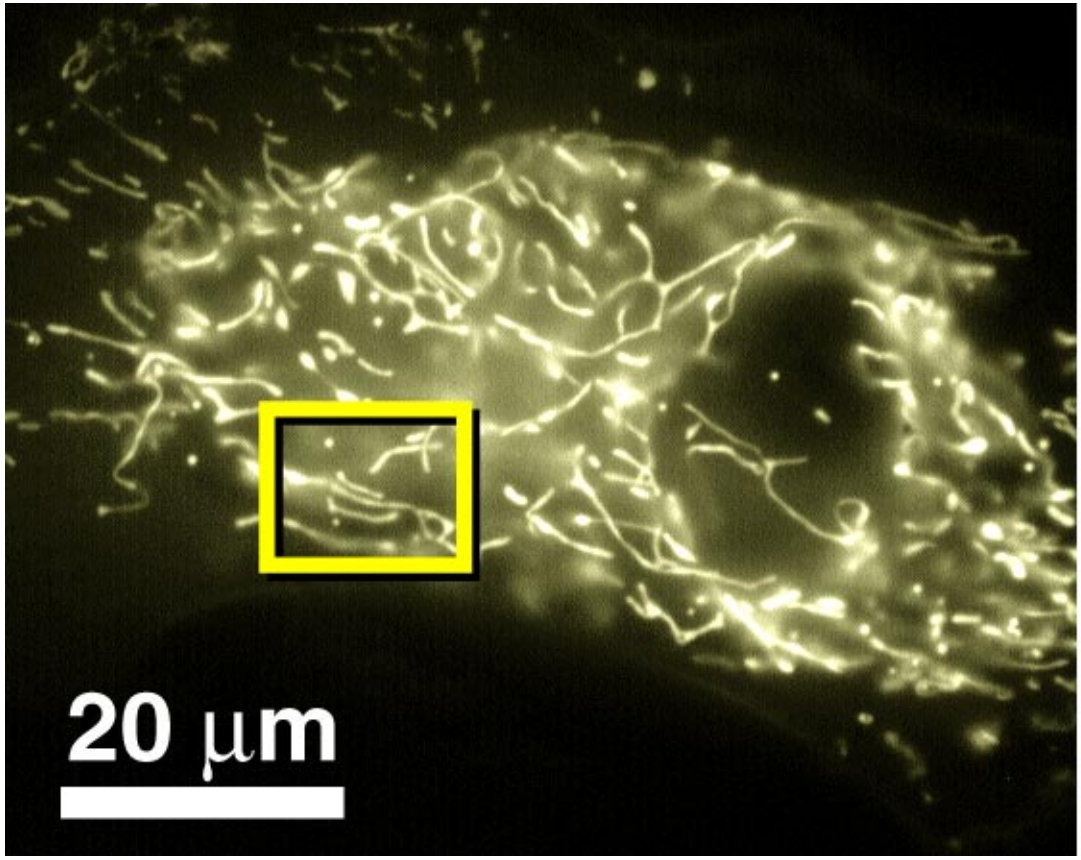
Bragg grating

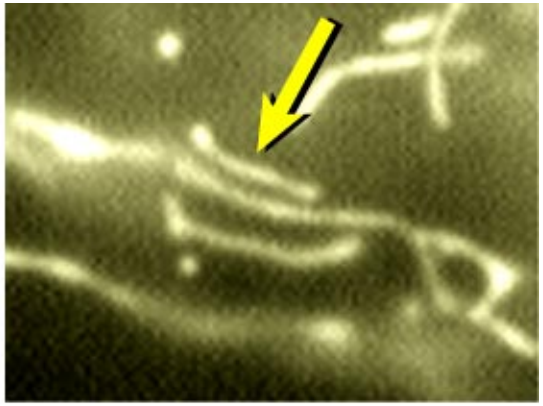


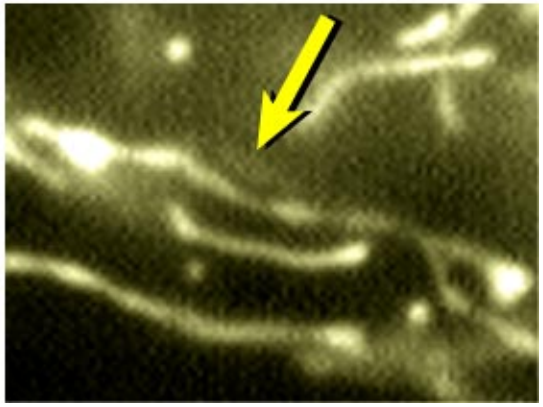
monolithic amplifier

laser active glass

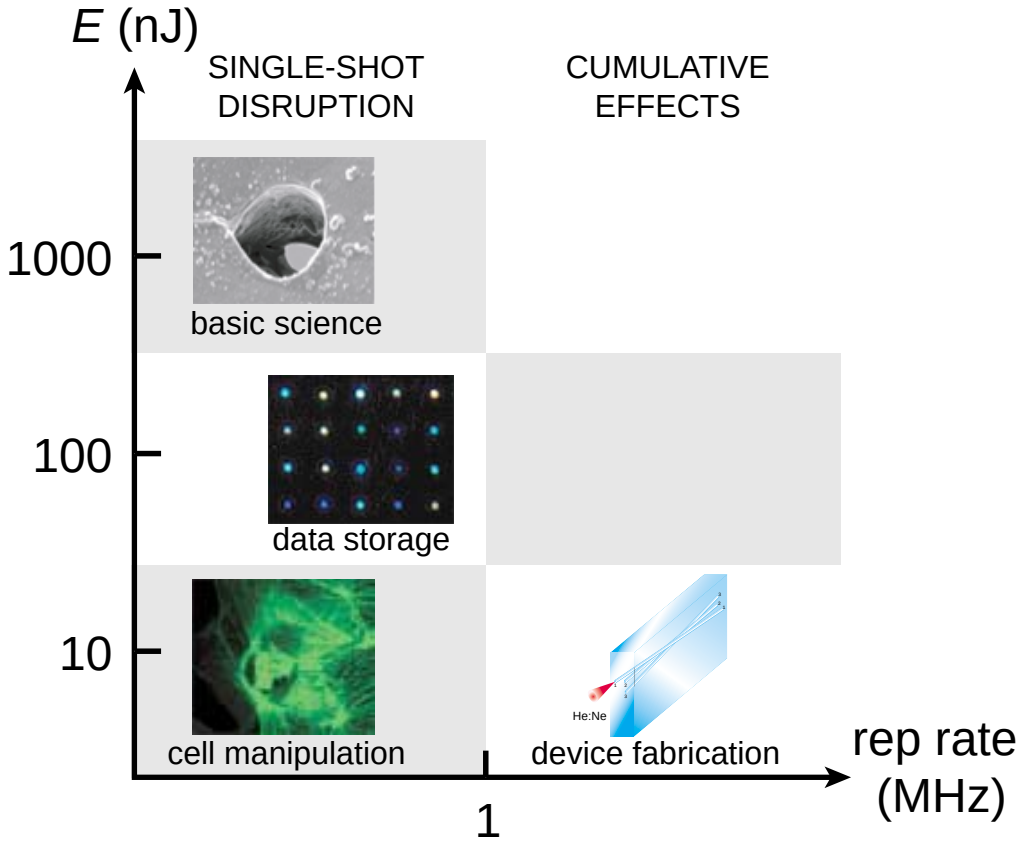
Low-energy processing







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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Prof. H. Ehrenreich
Prof. T. Kaxiras

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

http://mazur-www.harvard.edu