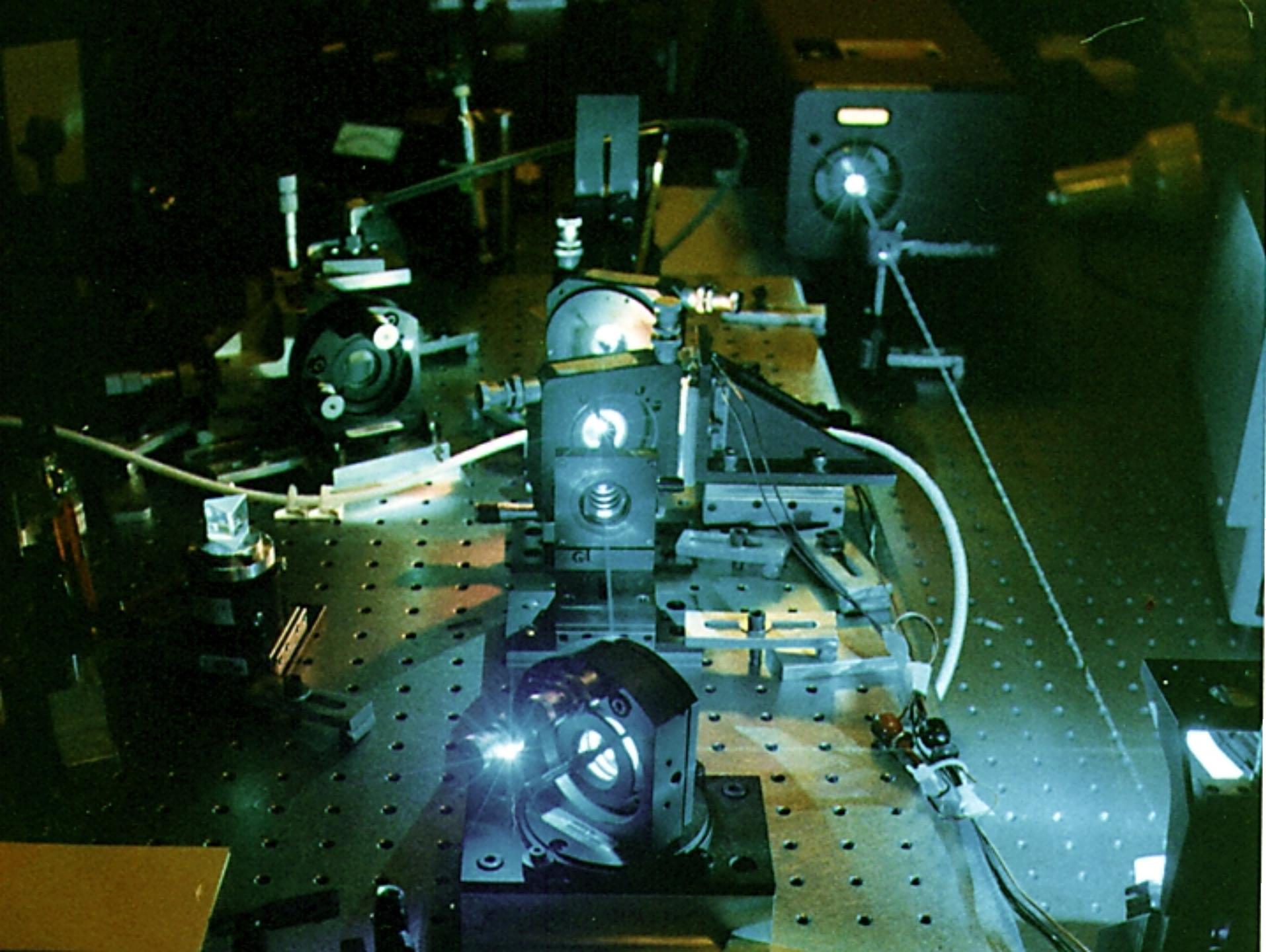


The behavior of solids under excitation with intense femtosecond laser pulses

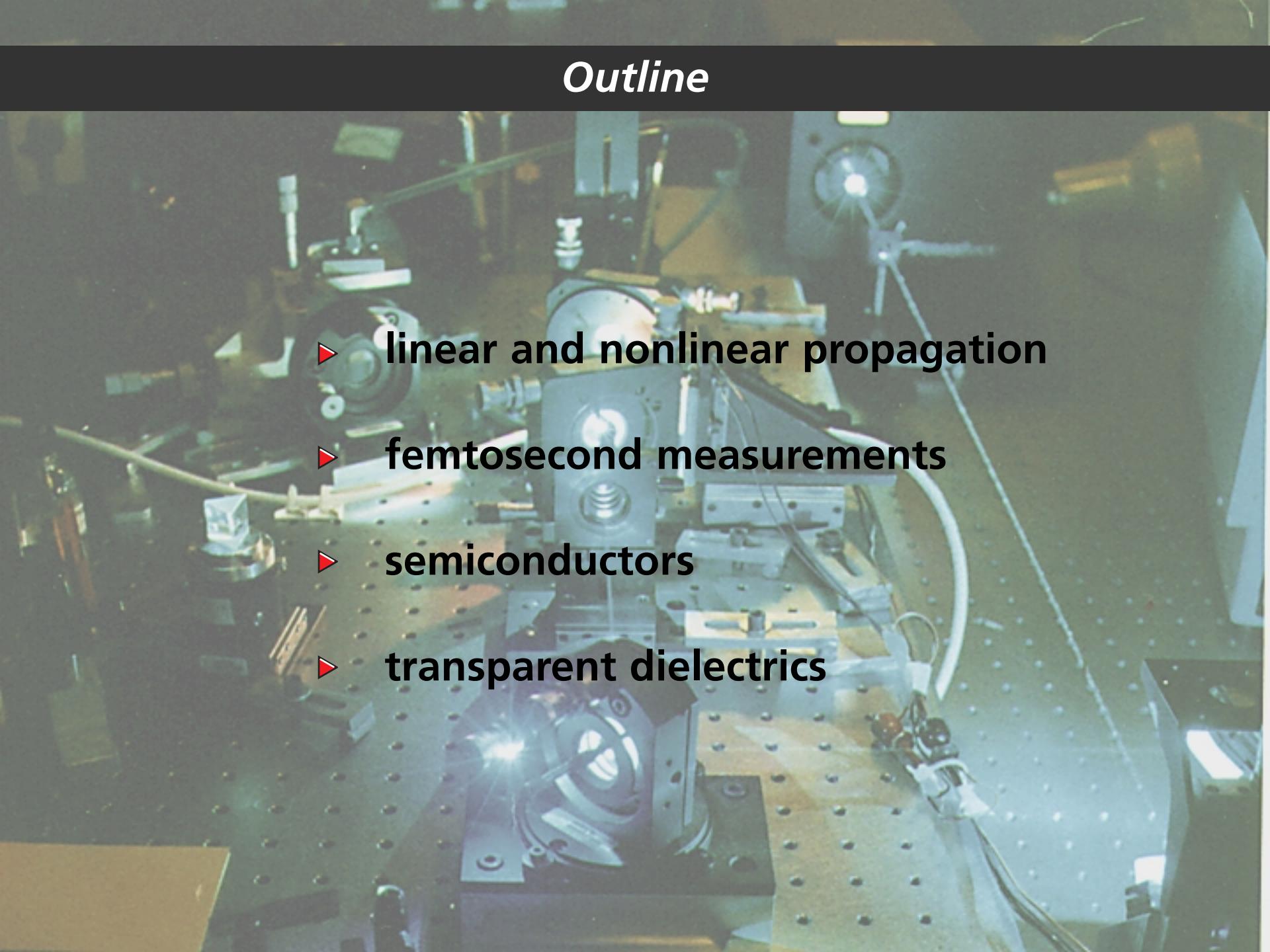
**Eric Mazur
Harvard University**

**Advances in Energy Transfer Processes
Erice, Italy
26 June 1999**

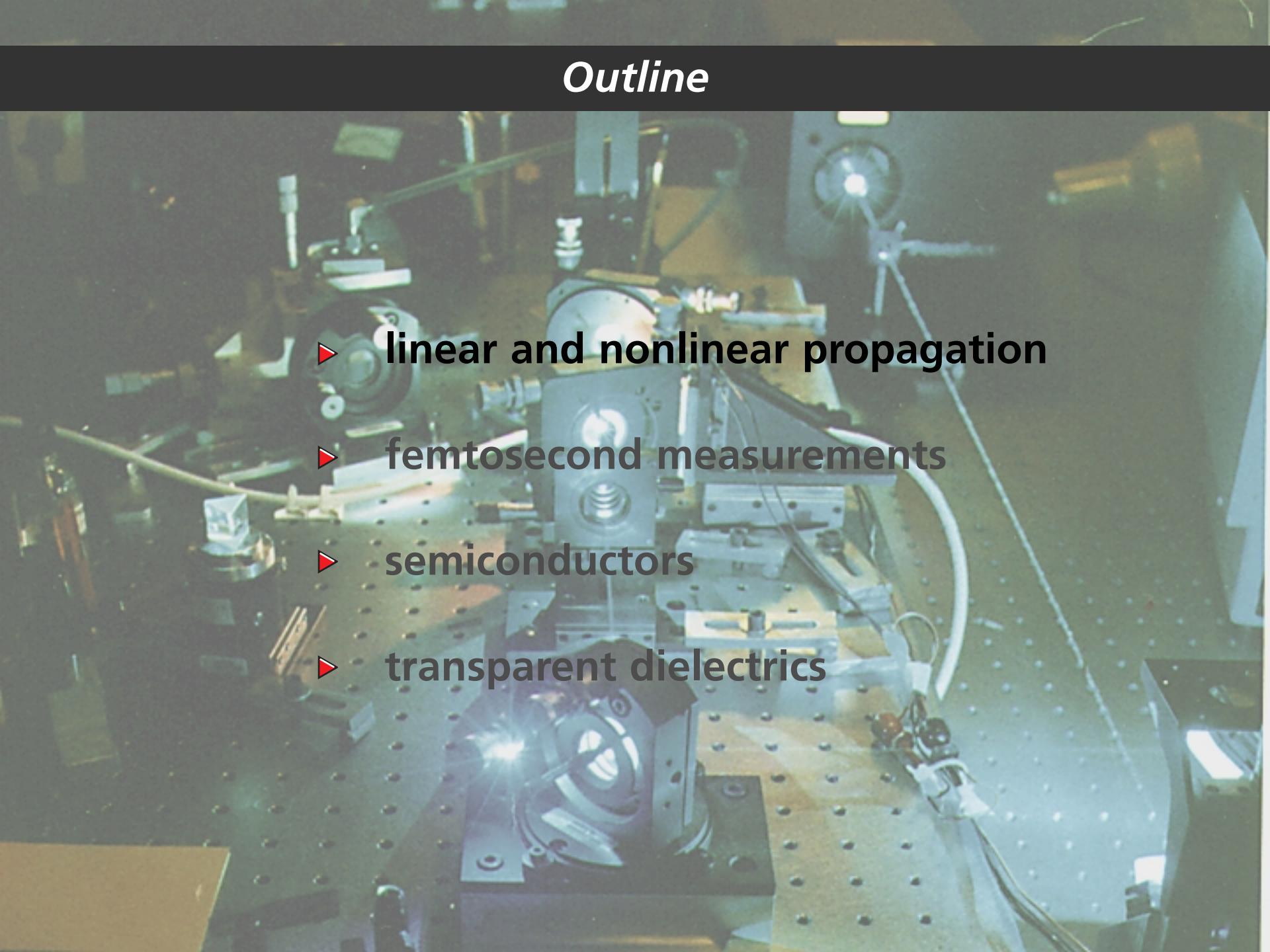




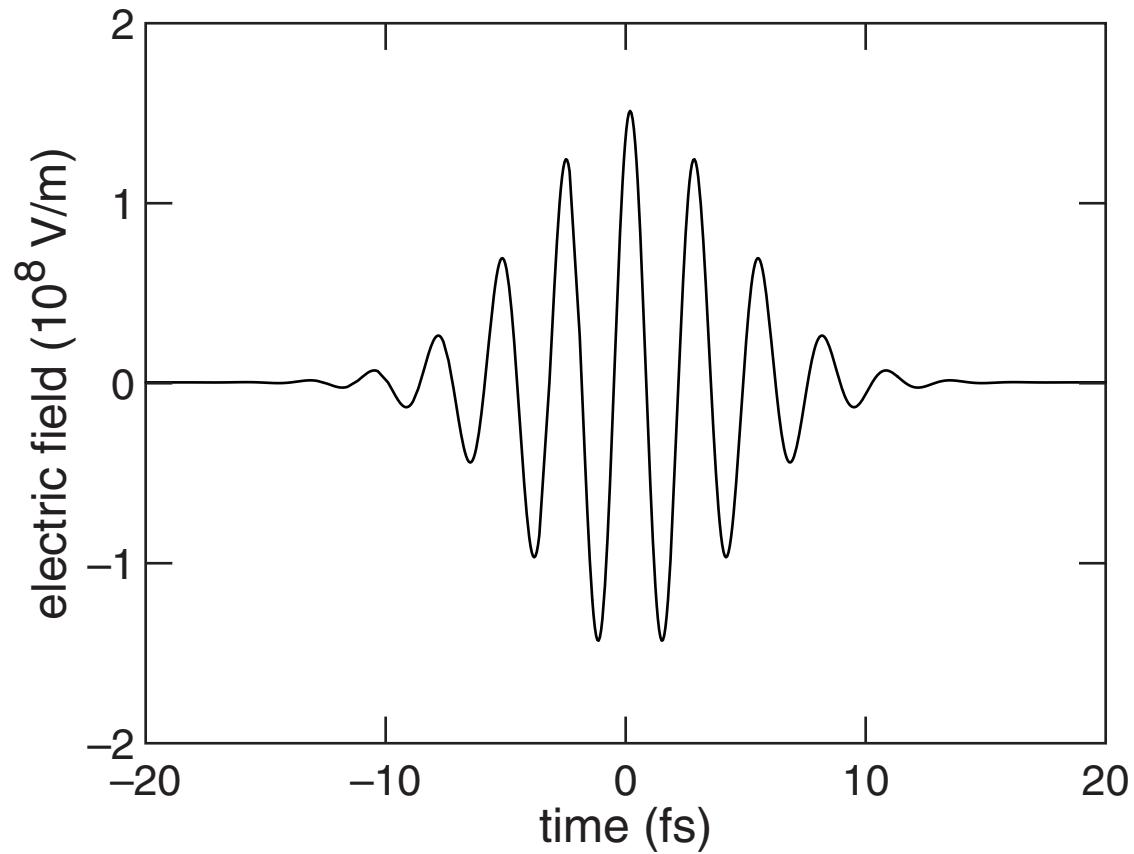
Outline

- 
- ▶ linear and nonlinear propagation
 - ▶ femtosecond measurements
 - ▶ semiconductors
 - ▶ transparent dielectrics

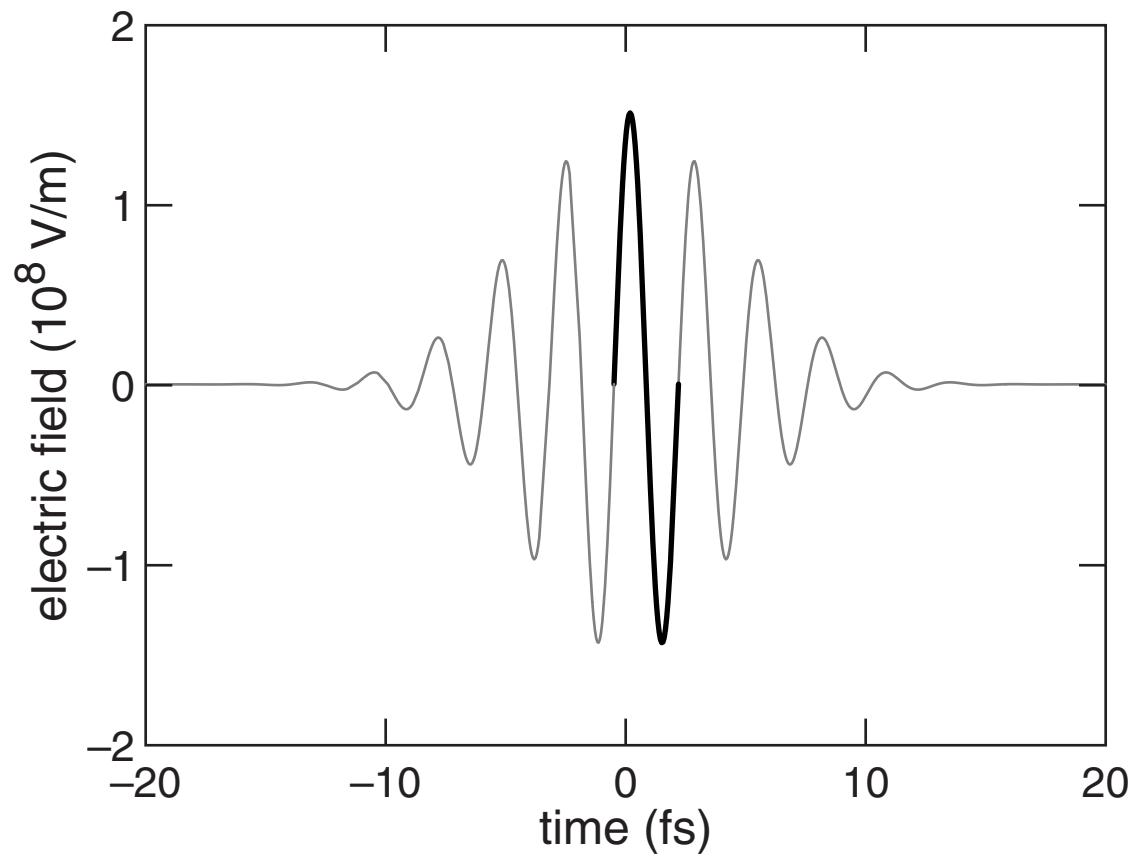
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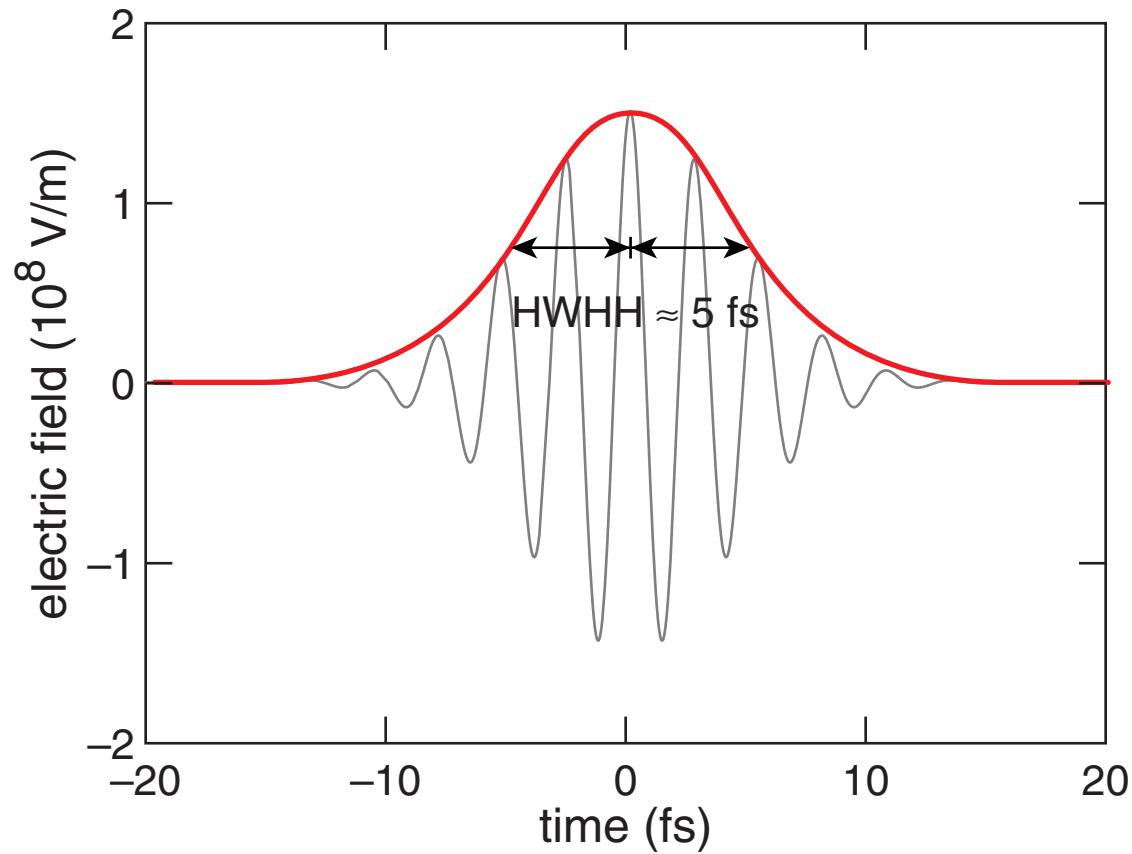
Introduction



Introduction



Introduction



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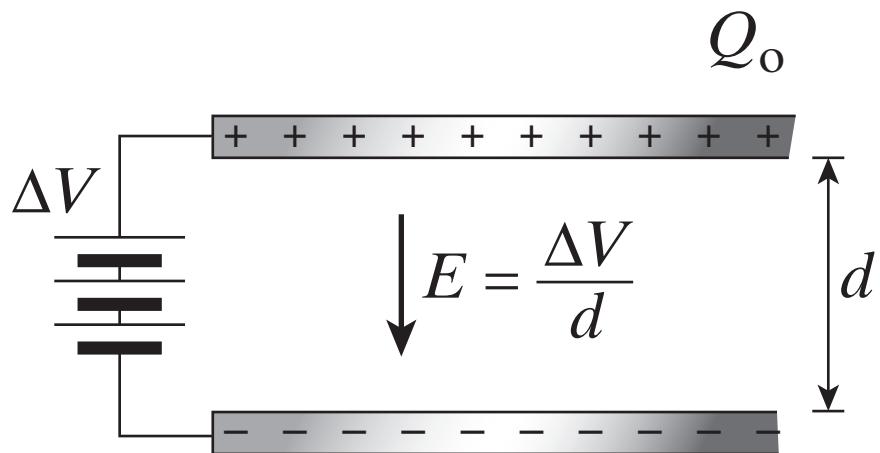
In non-ferromagnetic media $\mu \approx 1$, and so $n \approx \sqrt{\epsilon}$.

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

Propagation of EM waves through medium

Dielectric constant measures increase in capacitance

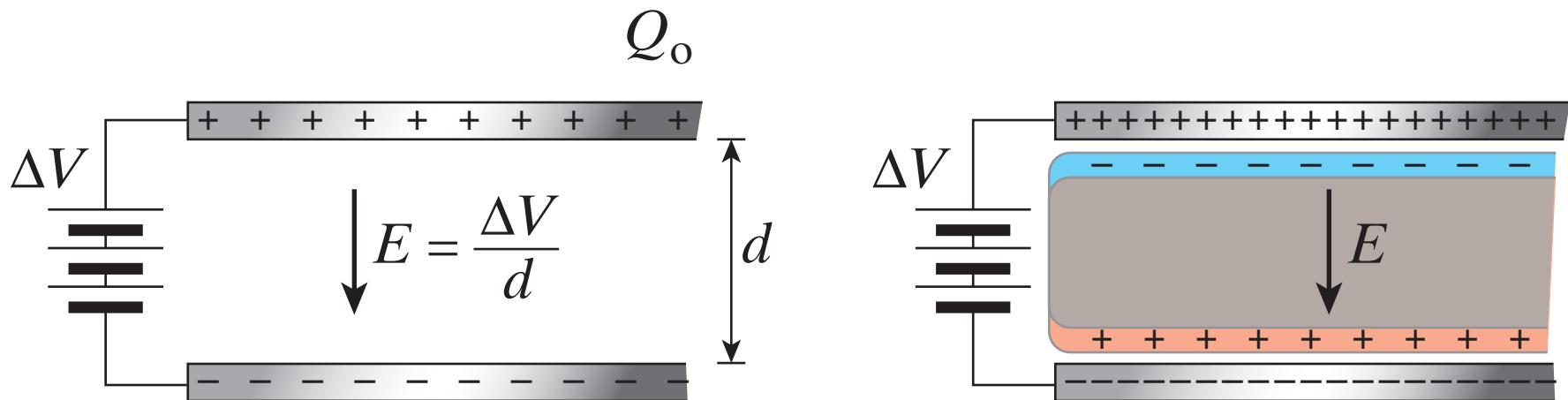
$$\epsilon = \frac{C_d}{C_o}$$



Propagation of EM waves through medium

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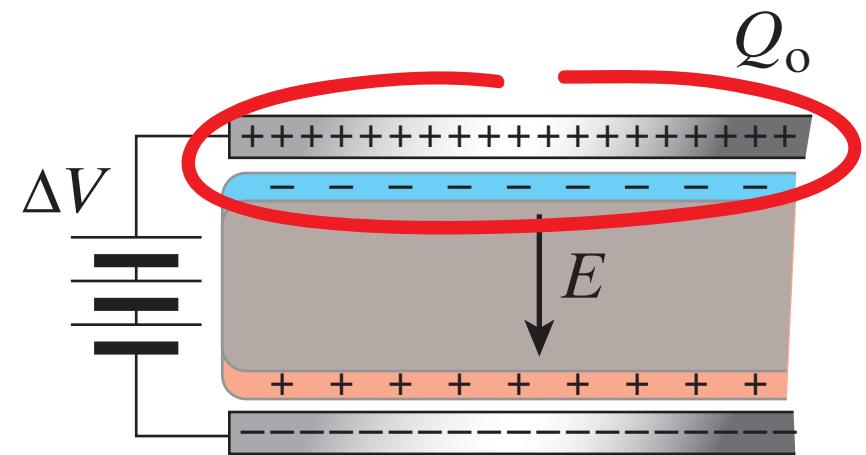
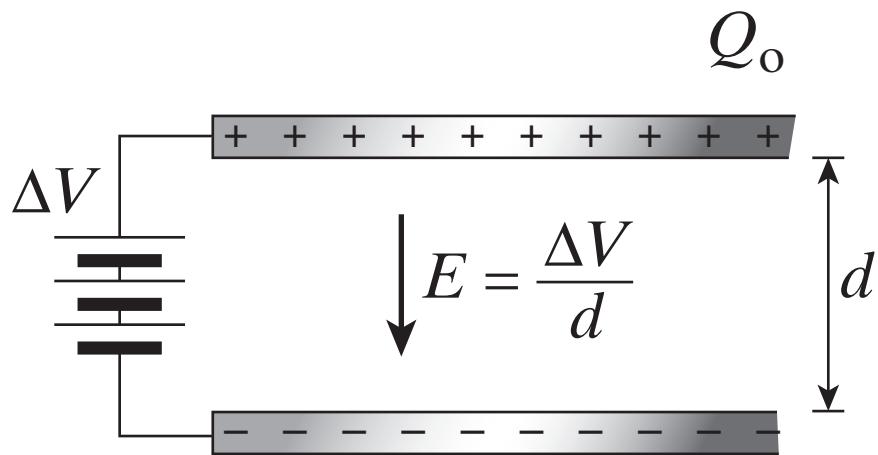
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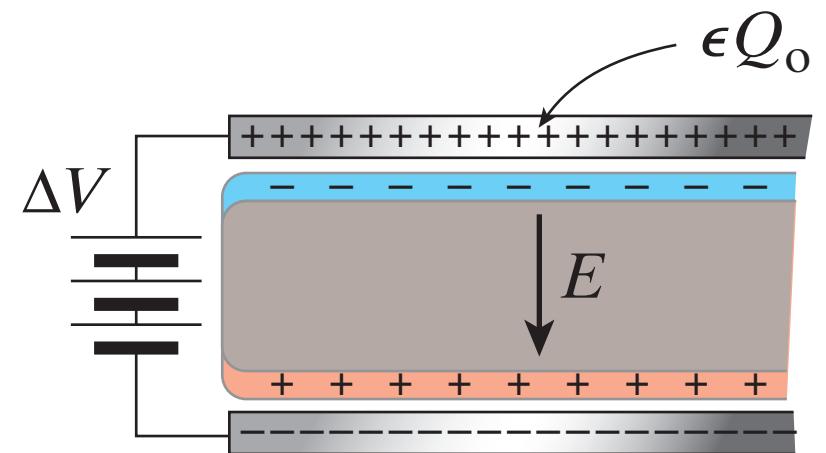
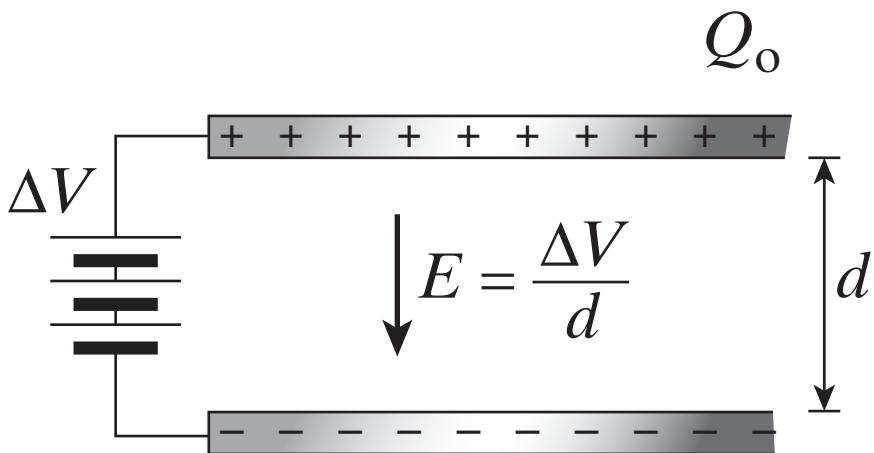
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Propagation of EM waves through medium

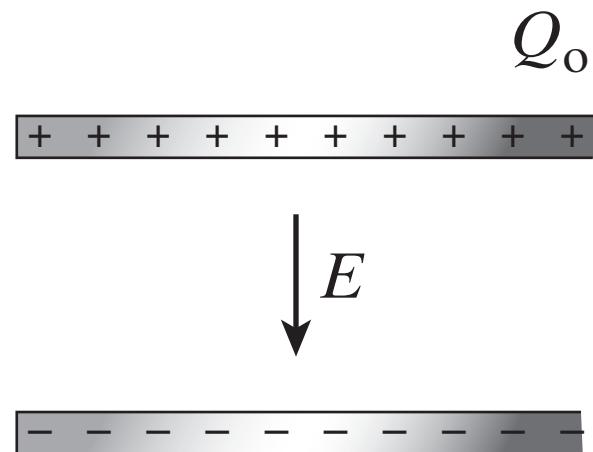
Dielectric constant measures increase in capacitance

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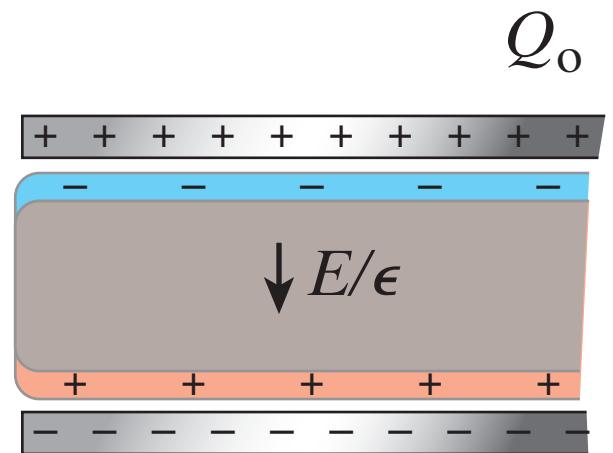
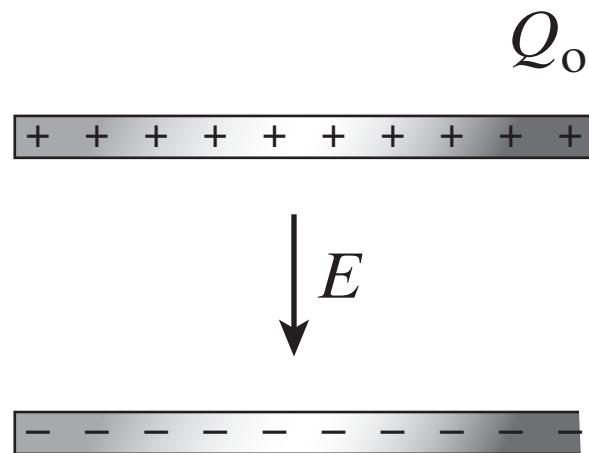
Propagation of EM waves through medium

Alternatively, ϵ is measure of the attenuation of the field



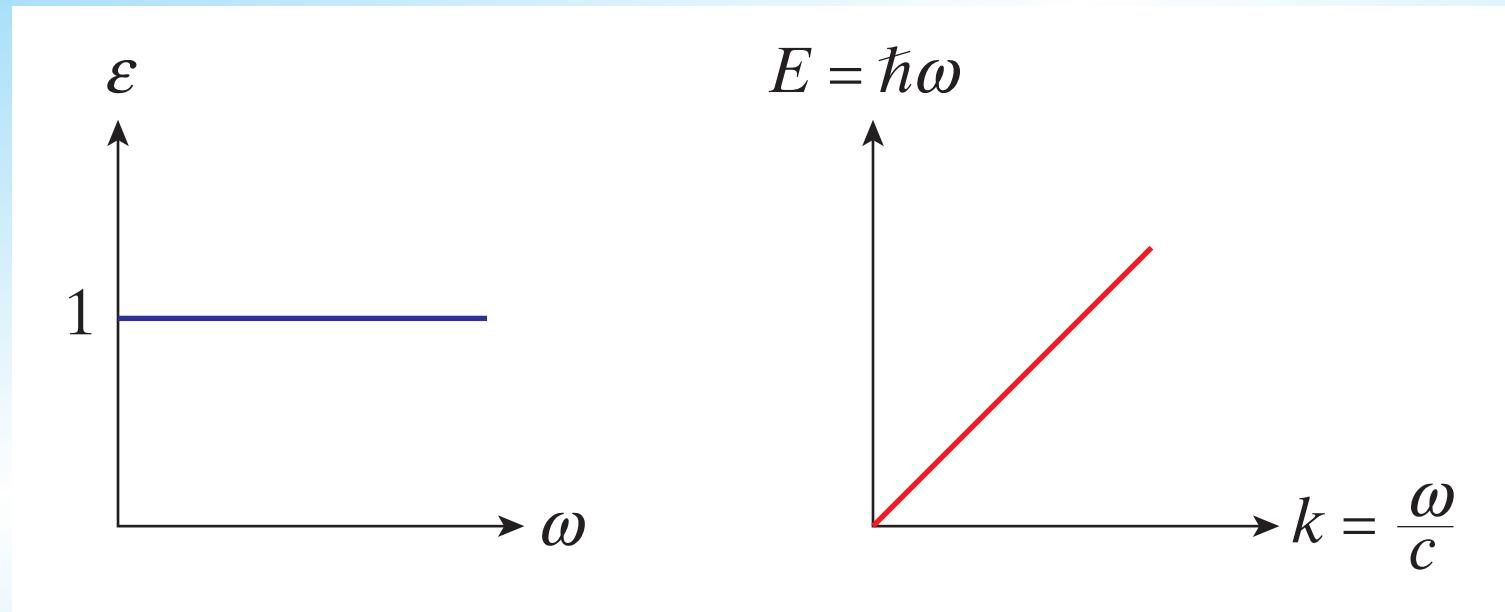
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Propagation of EM waves through medium

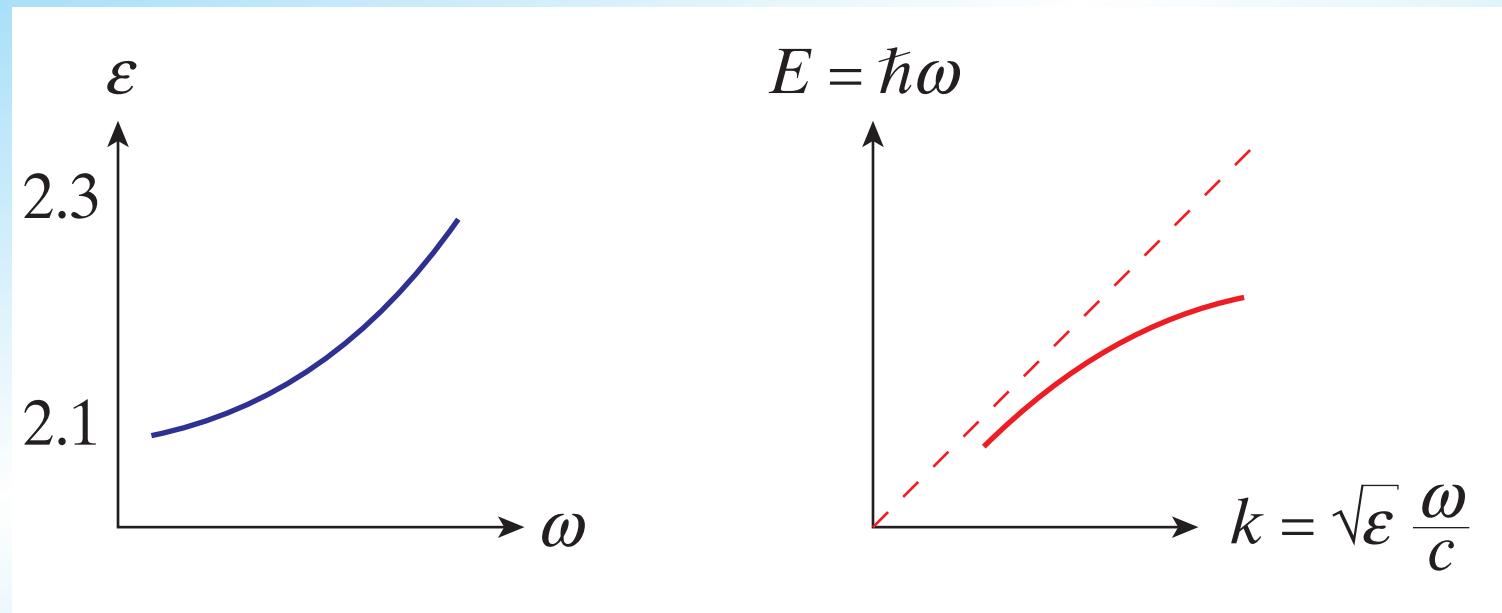
In vacuum: $f\lambda = \frac{\omega}{k} = c \Rightarrow \omega = c k$



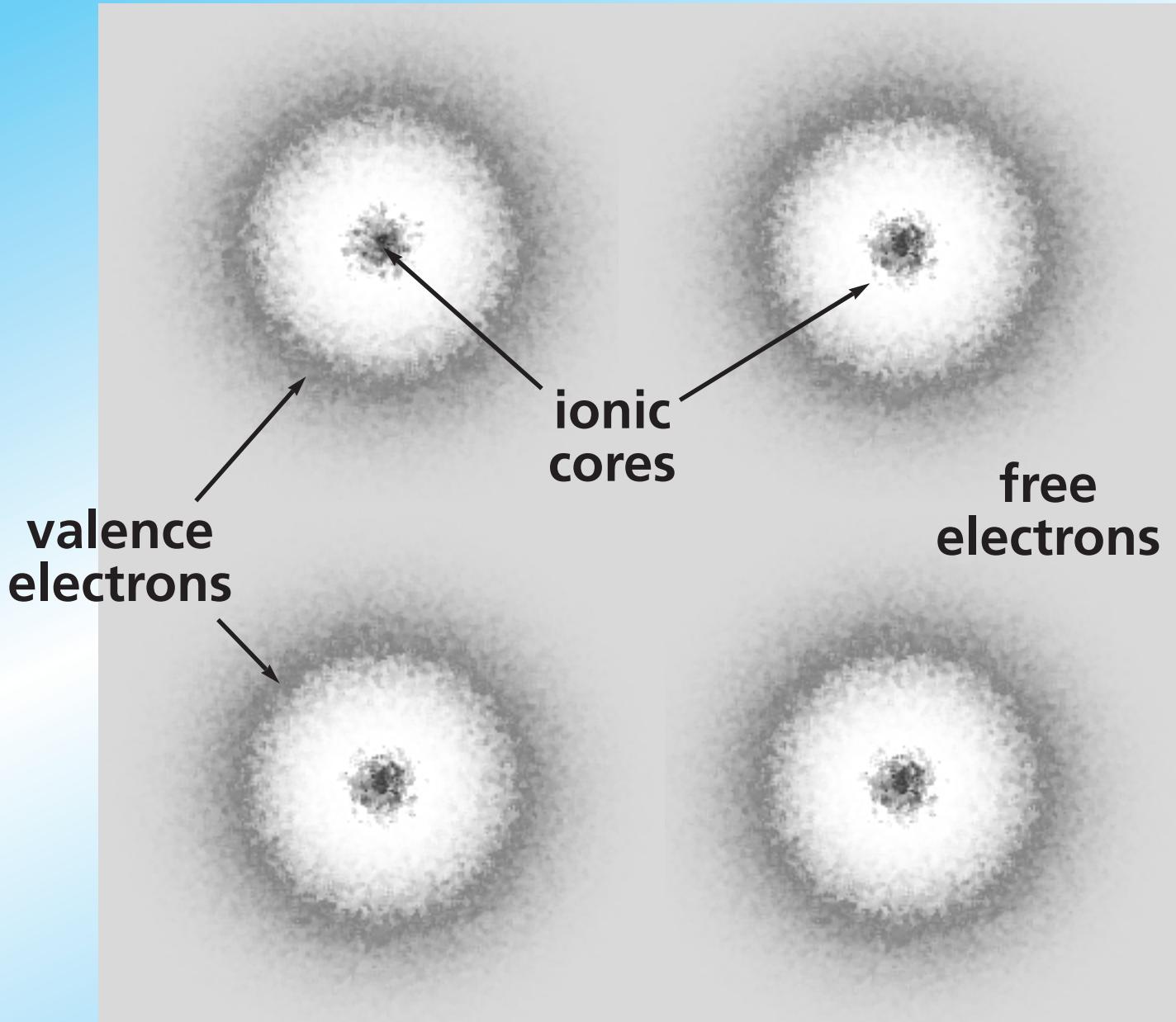
Propagation of EM waves through medium

In medium:

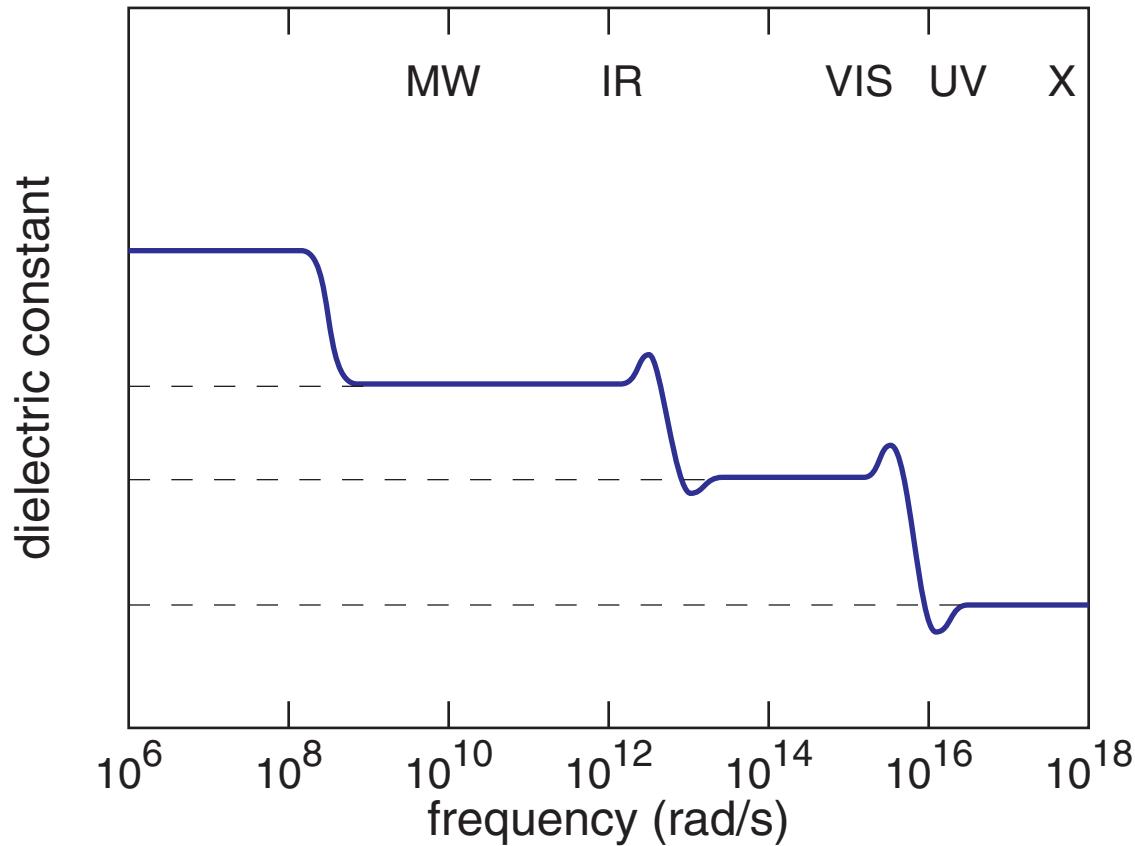
$$v = \frac{c}{\sqrt{\epsilon(\omega)}} = \frac{c}{n} \quad \Rightarrow \quad \omega = \frac{c}{\sqrt{\epsilon}} k$$



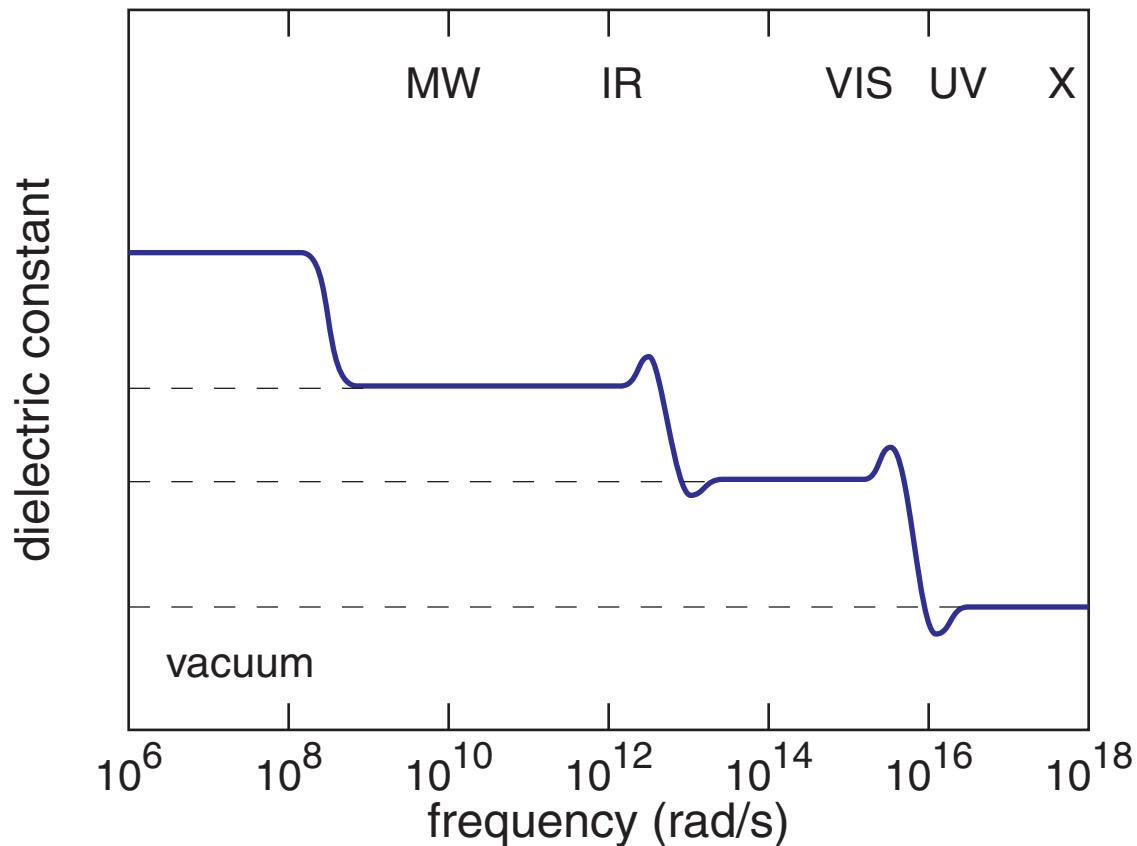
Which charges participate?



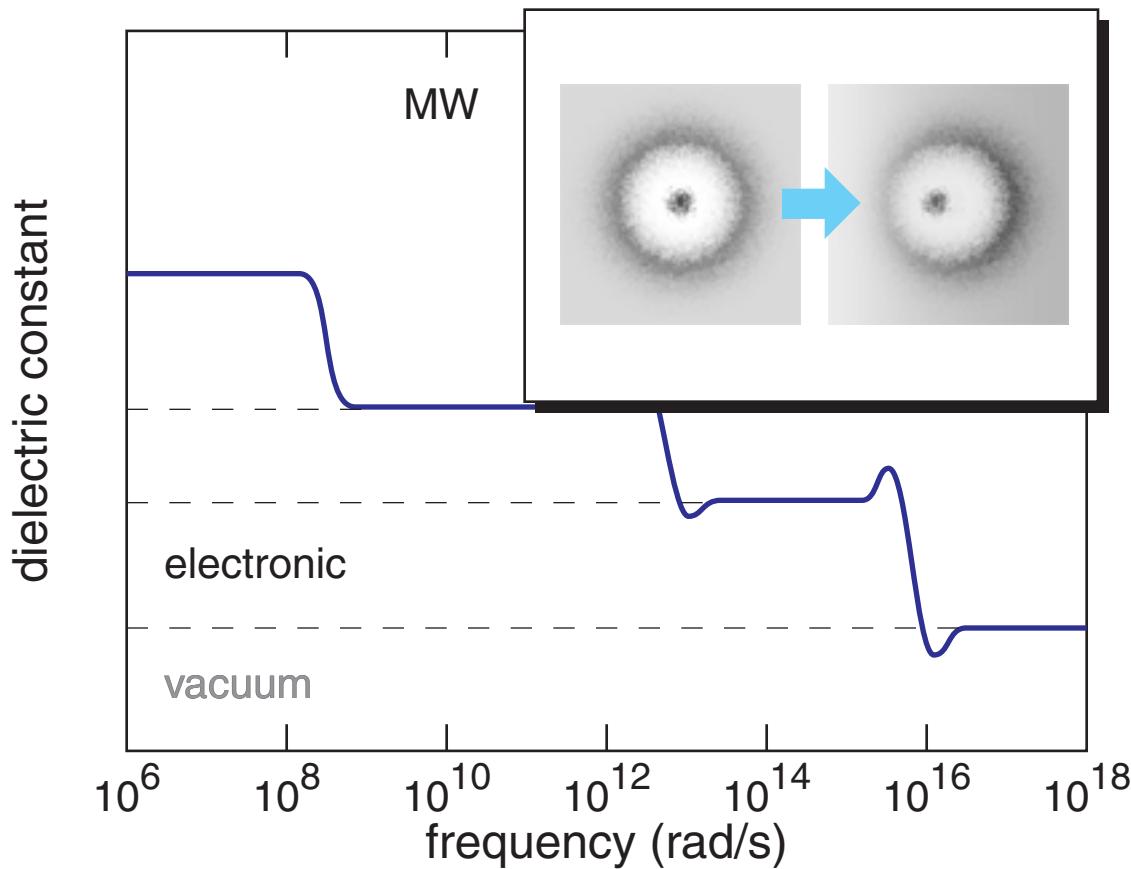
Dielectric function



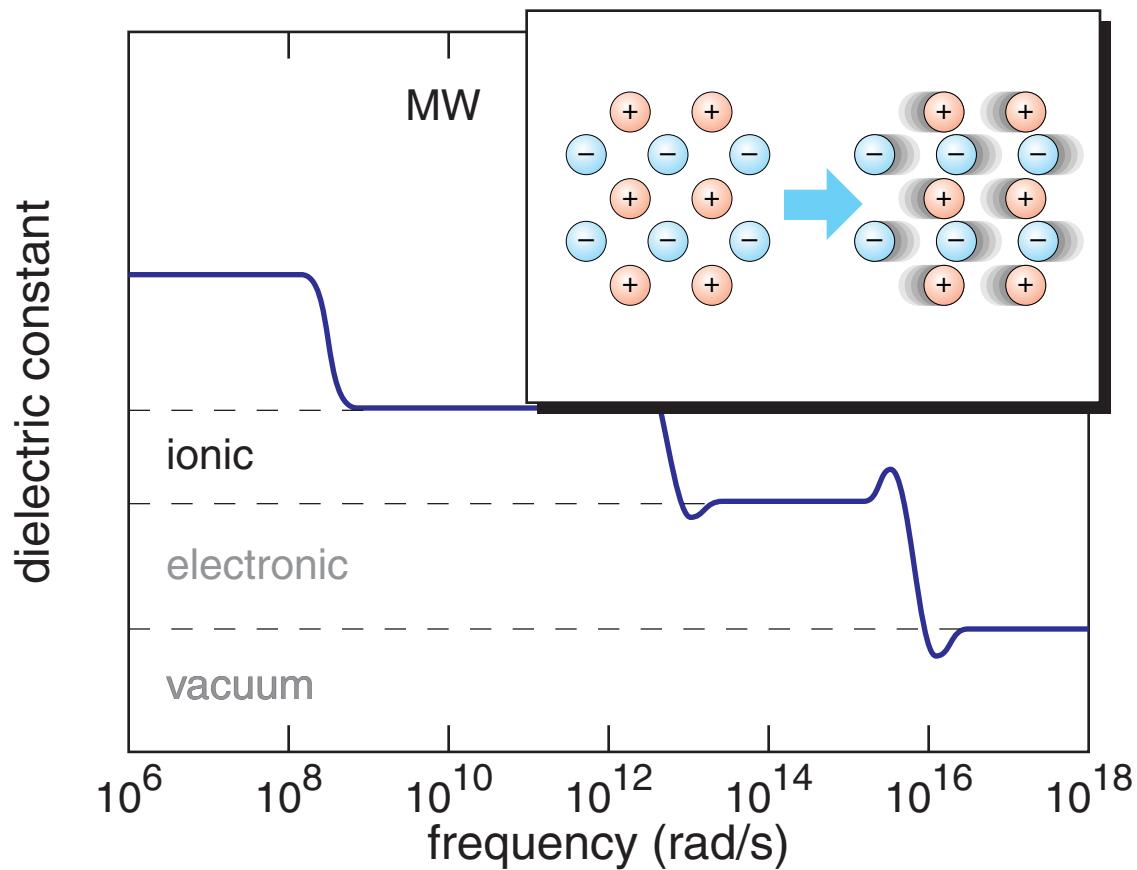
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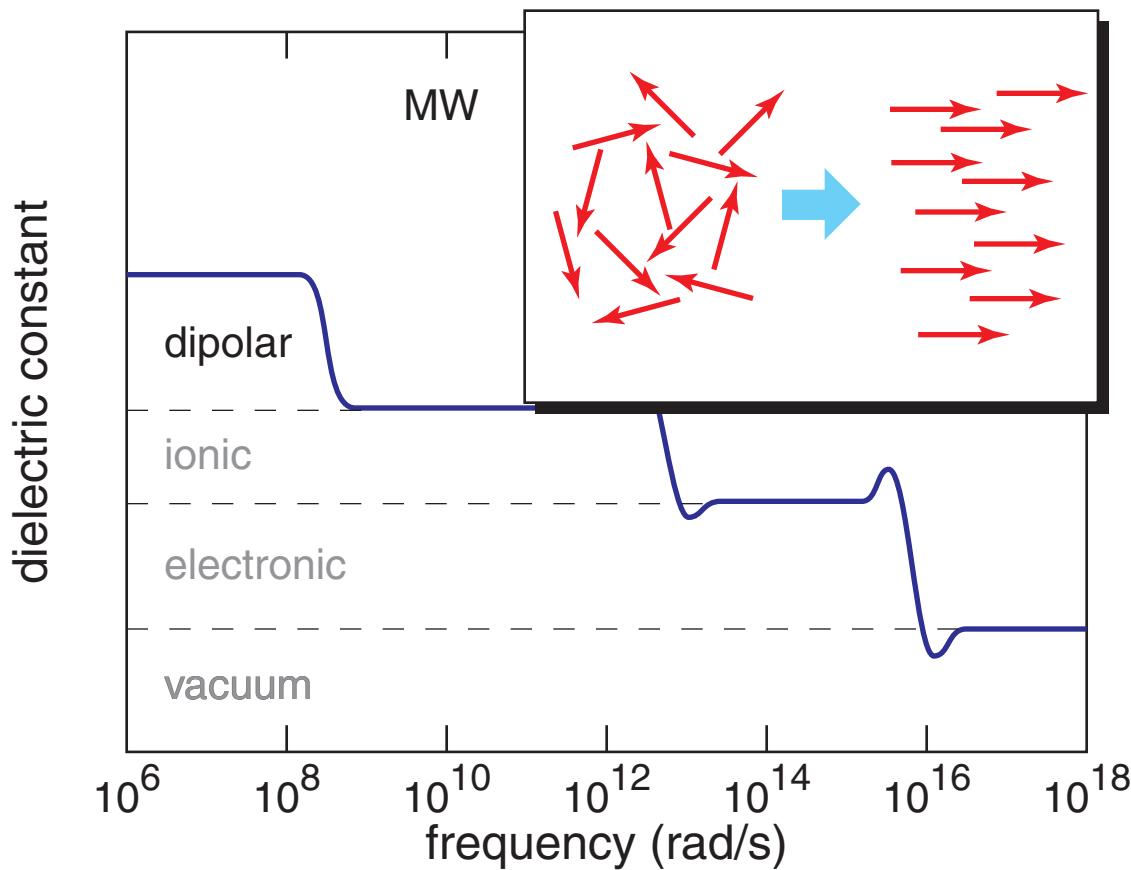
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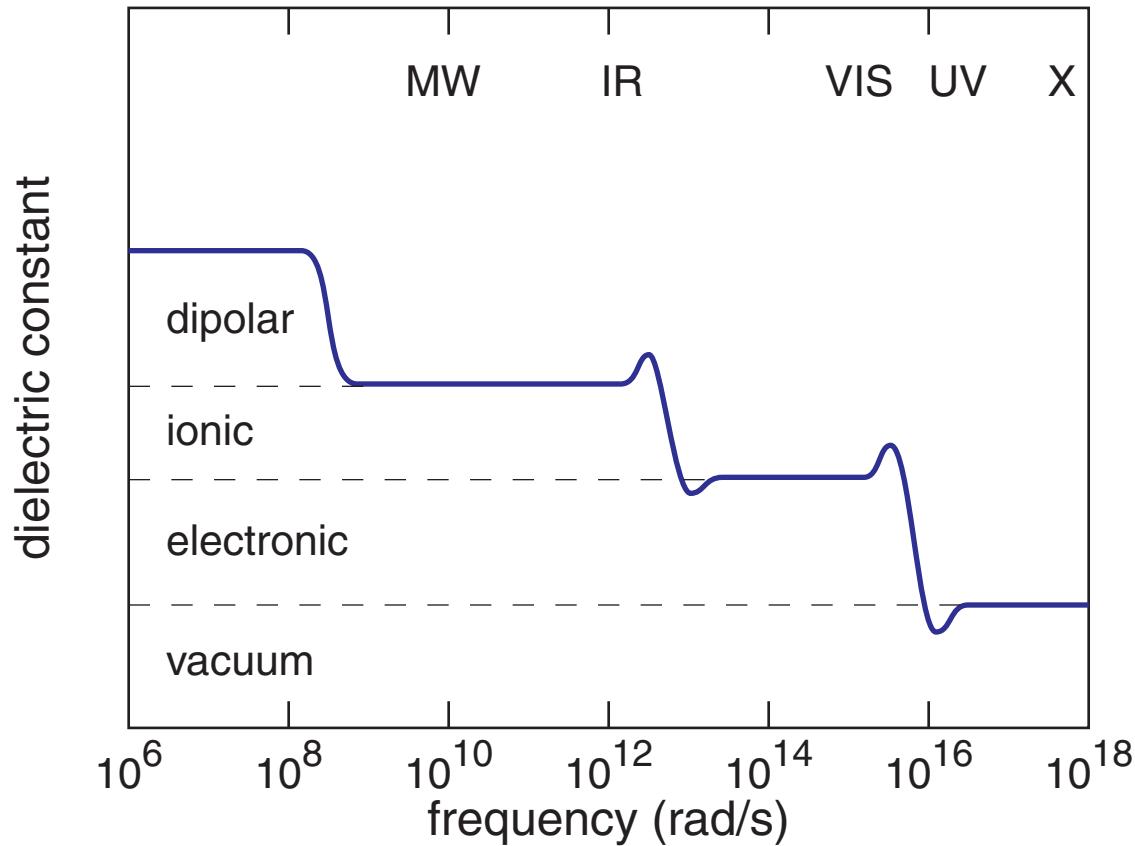
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Bound electrons

Electron on a string:

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

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

Bound electrons

Steady state: electron oscillates at driving frequency

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

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Polarization

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

Bound electrons

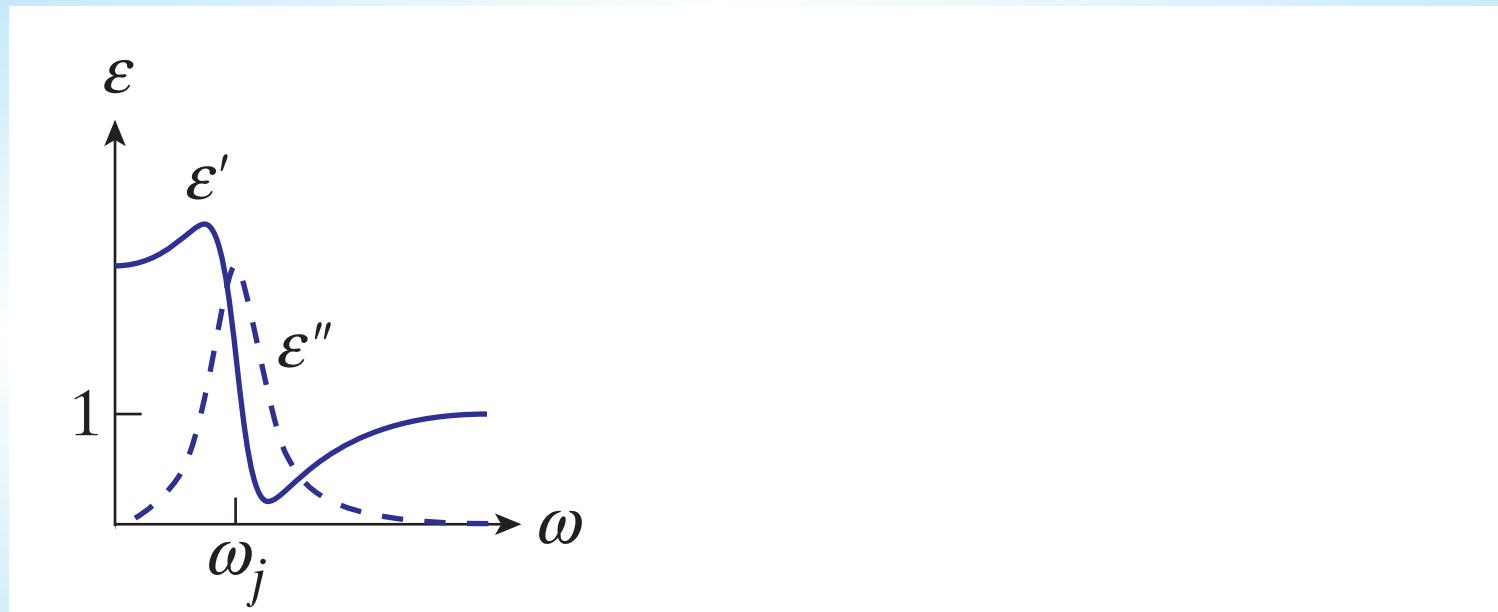
Dielectric function

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

Bound electrons

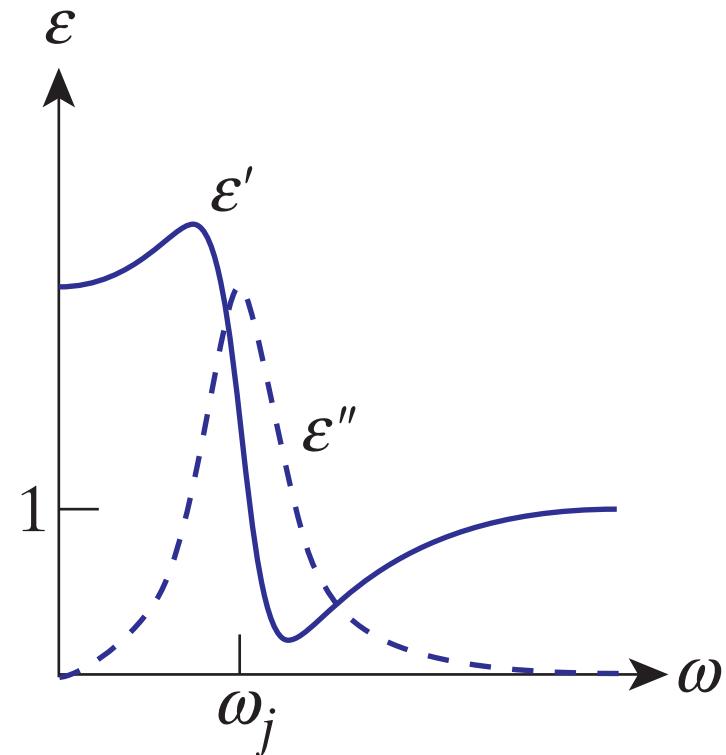
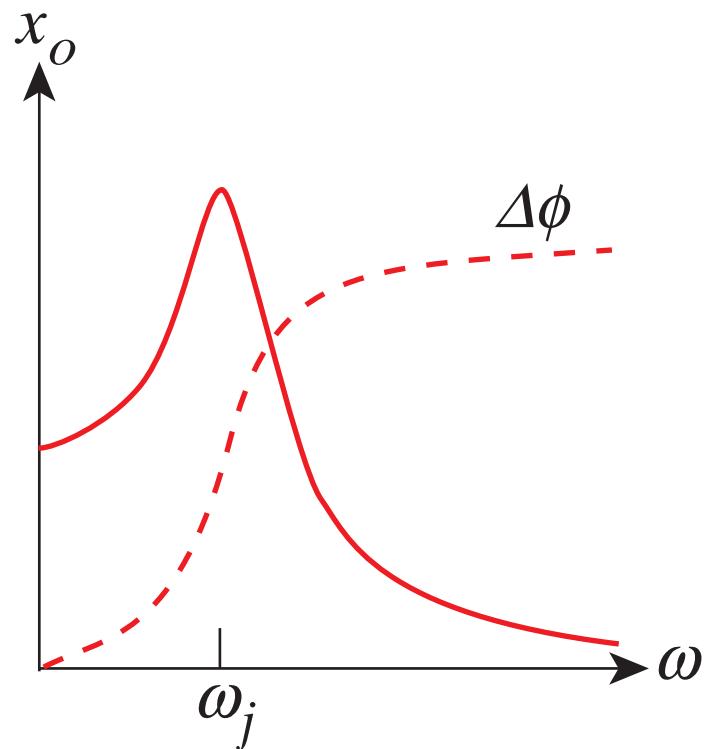
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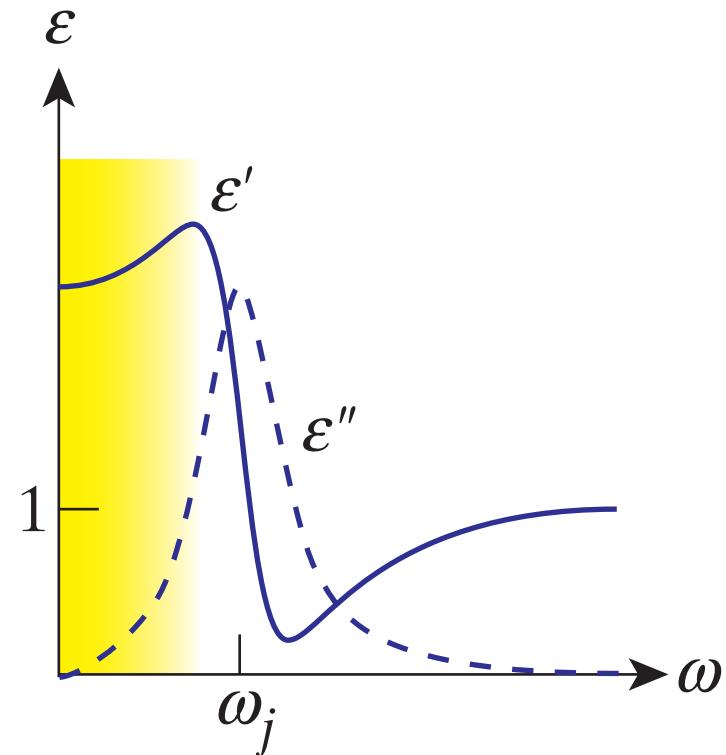
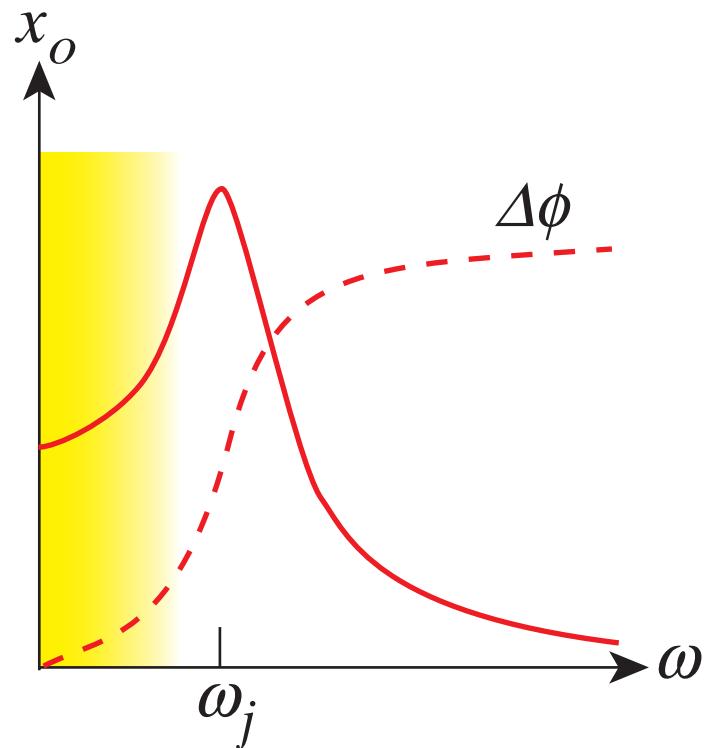
Bound electrons

amplitude of bound charge oscillation



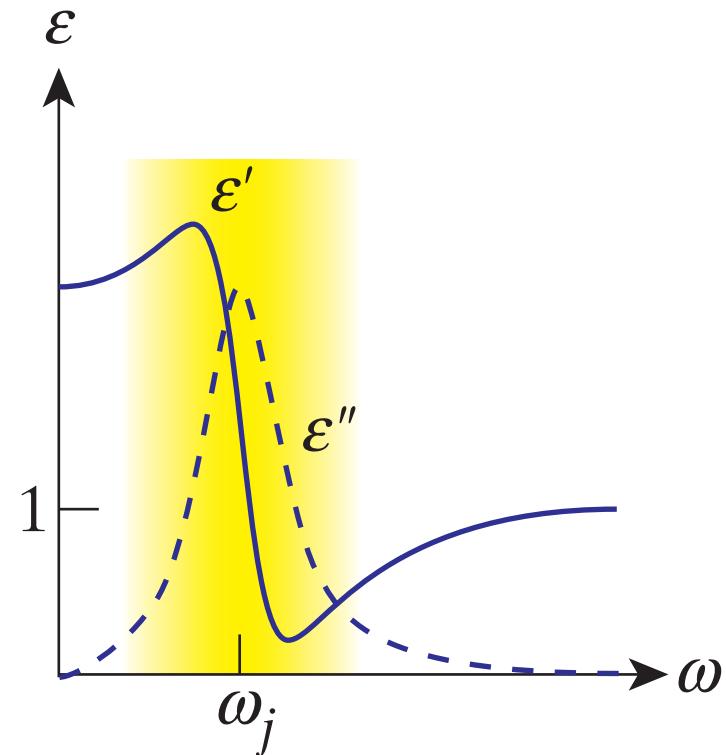
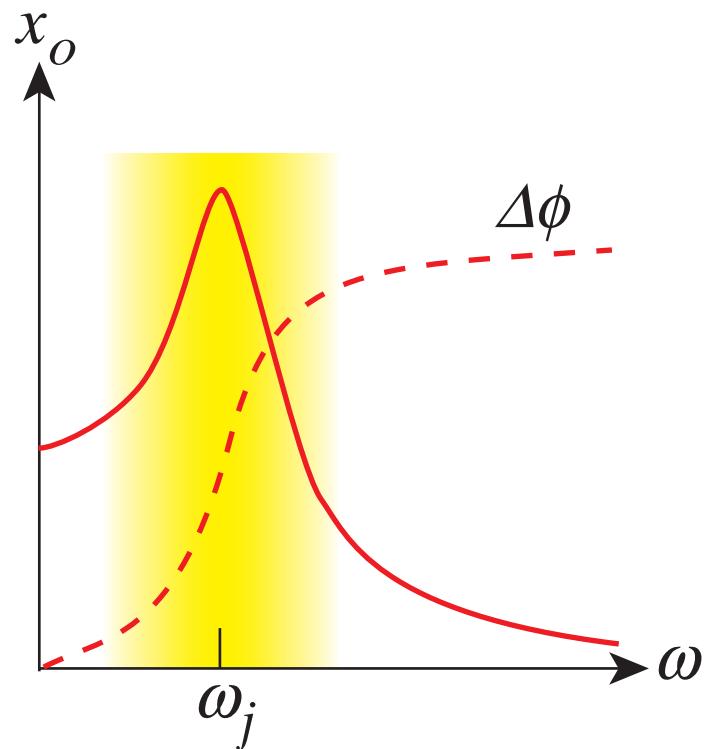
Bound electrons

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



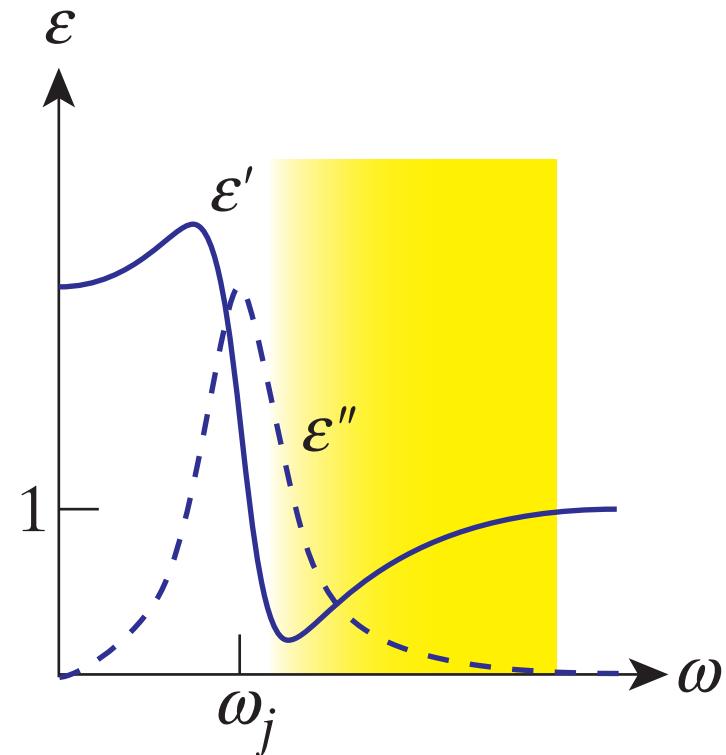
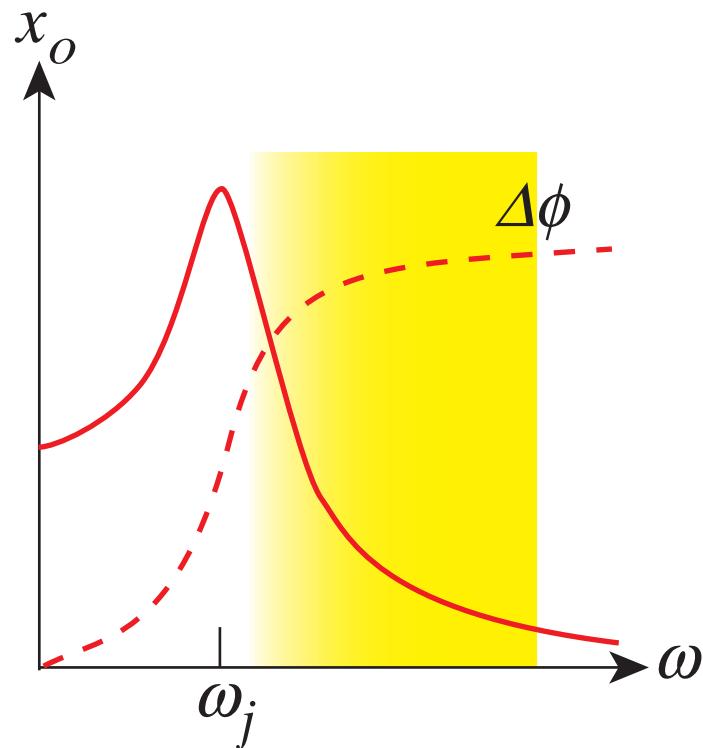
Bound electrons

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



Bound electrons

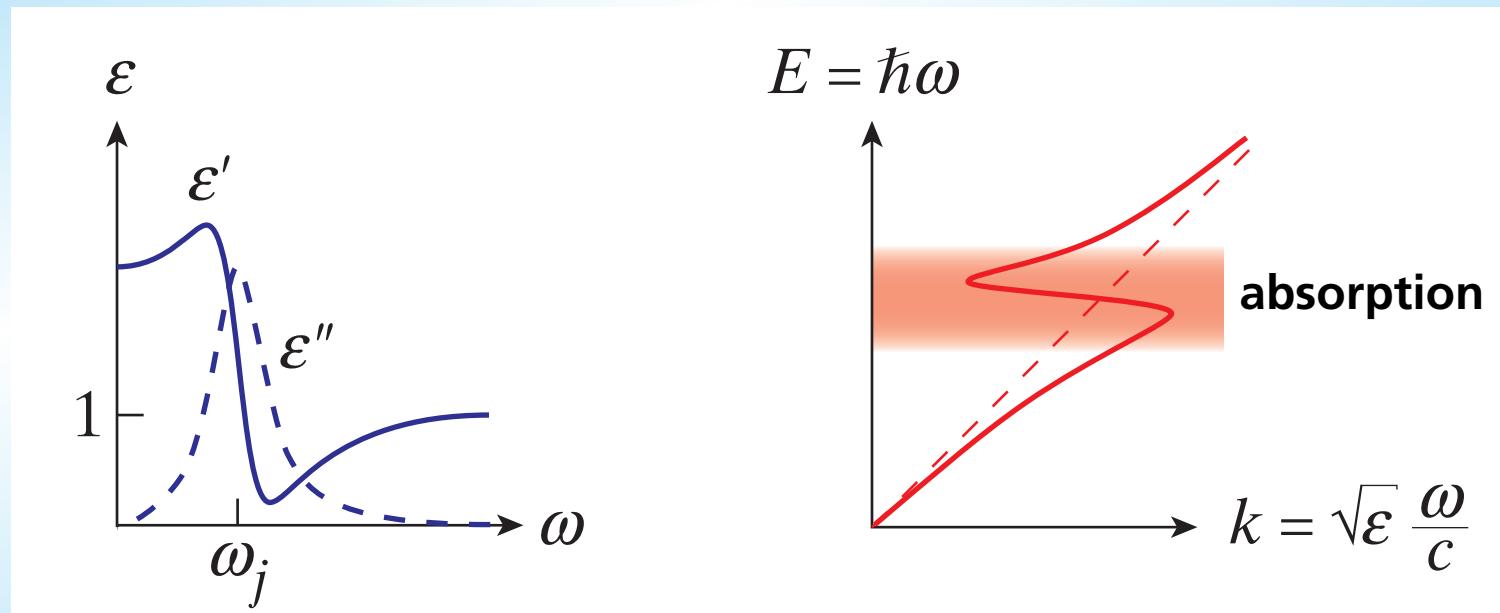
Above resonance: bound charges cannot keep up with driving field \Rightarrow dielectric like a vacuum



Bound electrons

Dielectric function

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



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

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

Free electrons

$\omega \gg 1$: **σ complex** \Rightarrow J out of phase with E

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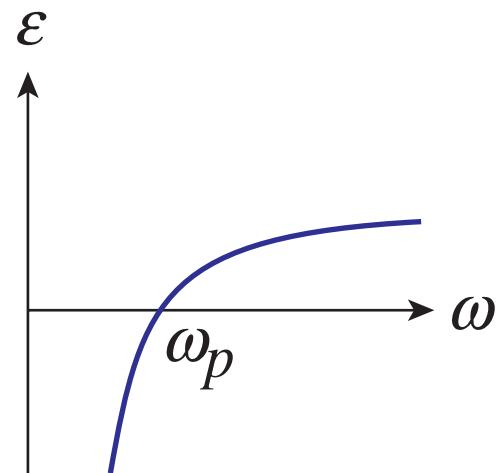
Dielectric function

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

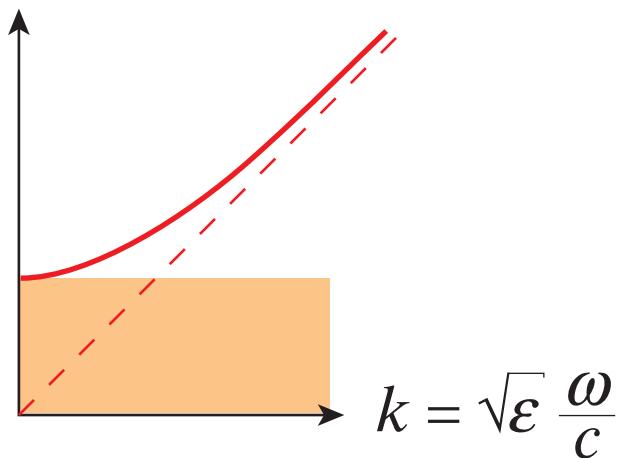
Plasma

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

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



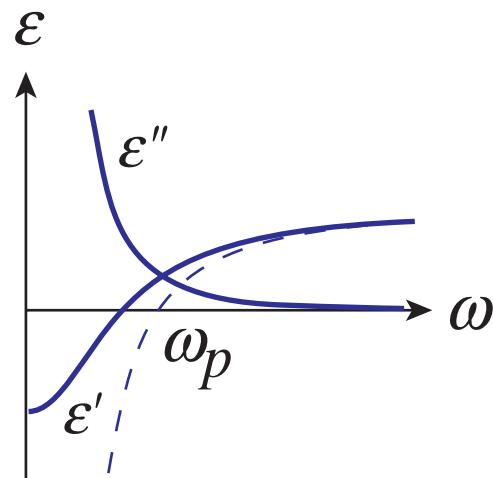
$$E = \hbar\omega$$



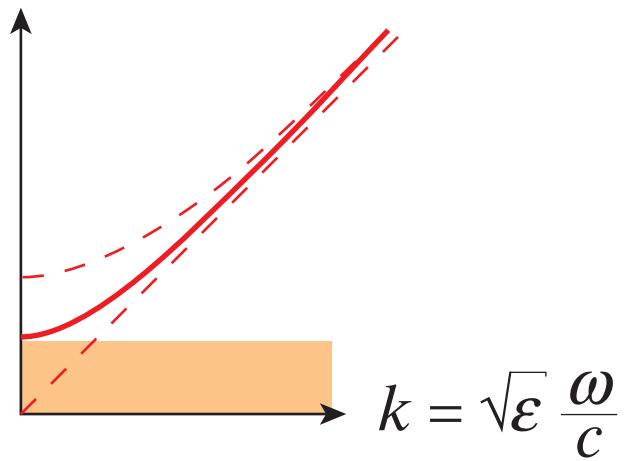
Plasma

Add damping

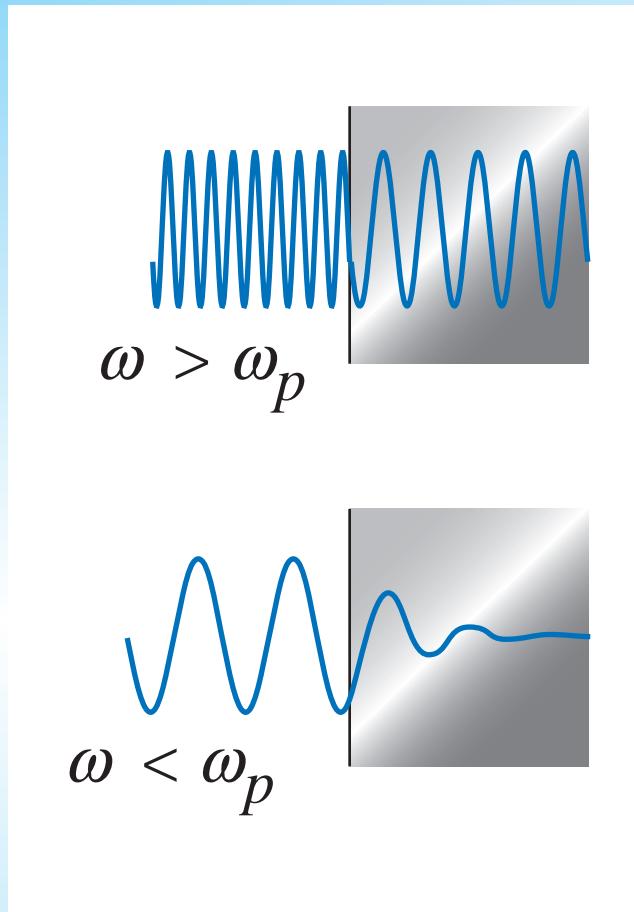
$$\gamma \lesssim \omega_p$$



$$E = \hbar\omega$$

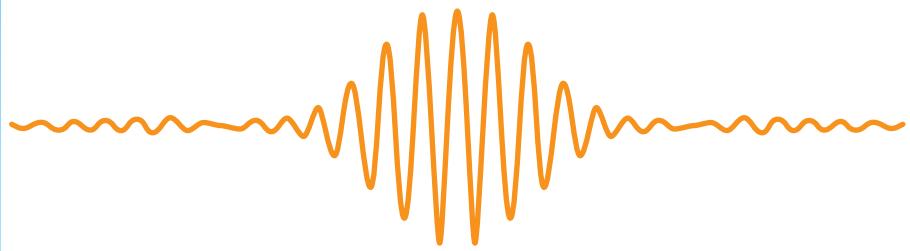


Plasma acts like a high-pass filter:

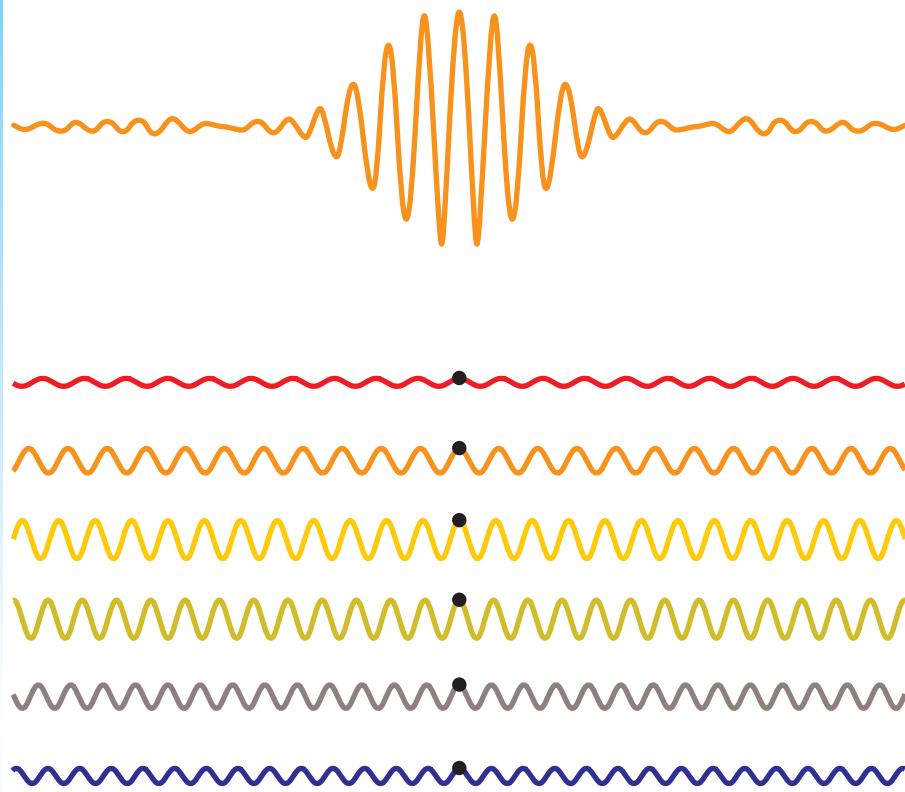


$\log N$ (cm $^{-3}$)	ω_p (rad s $^{-1}$)	λ_p
22	6×10^{15}	330 nm
18	6×10^{13}	33 μm
14	6×10^{11}	3.3 mm
10	6×10^9	0.33 m

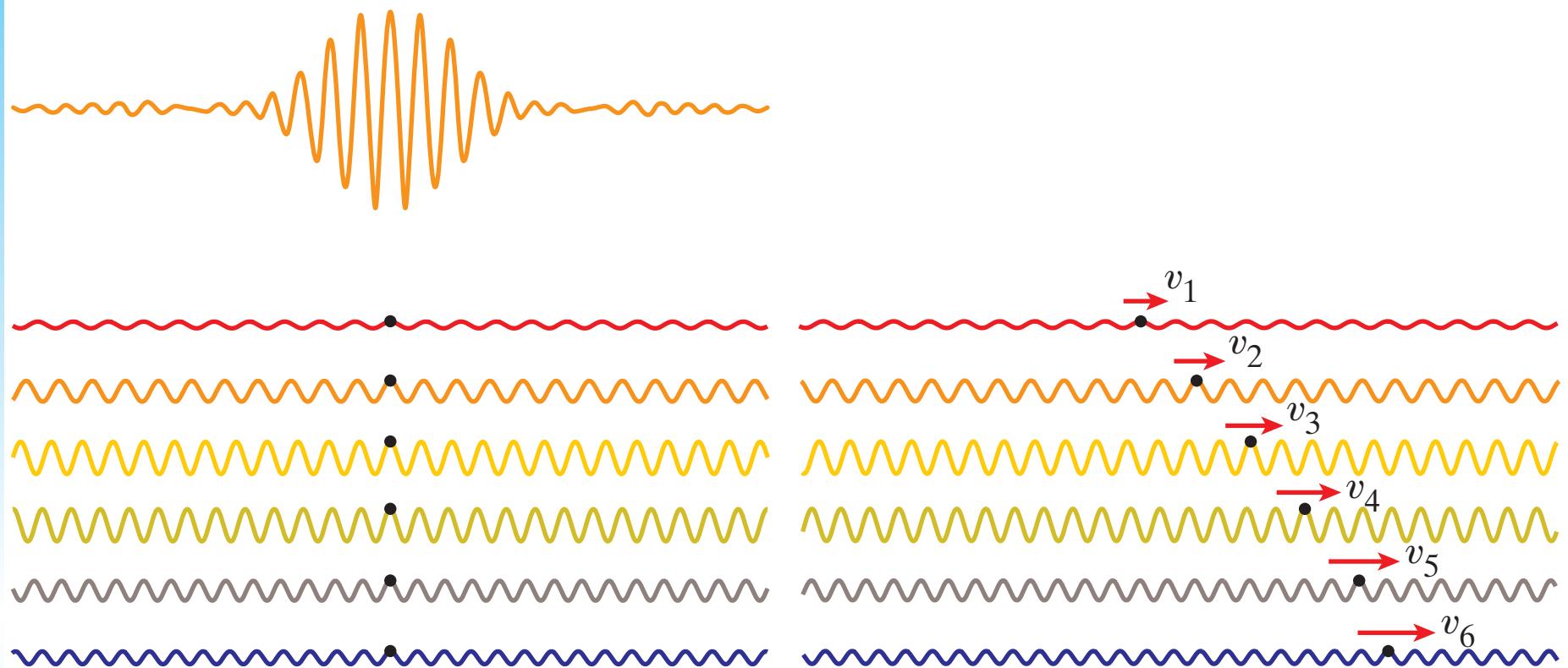
Pulse dispersion



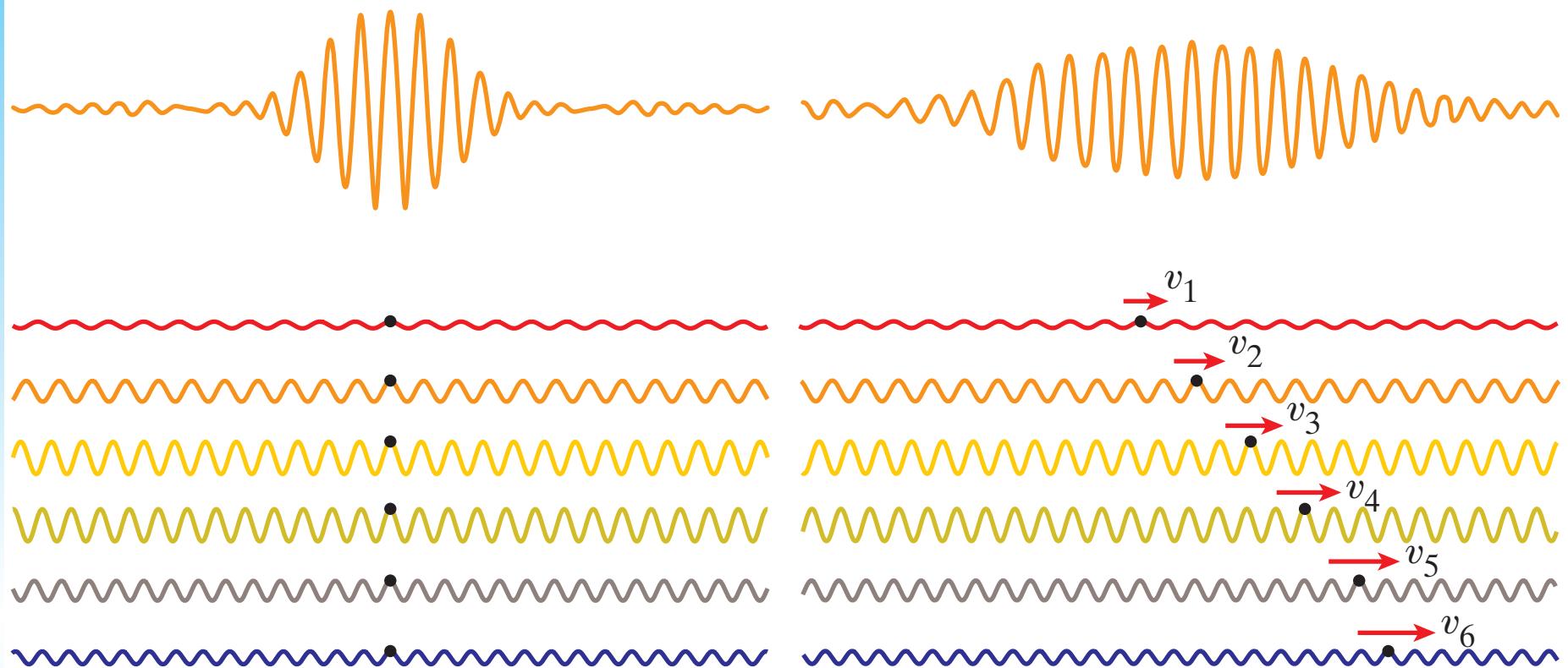
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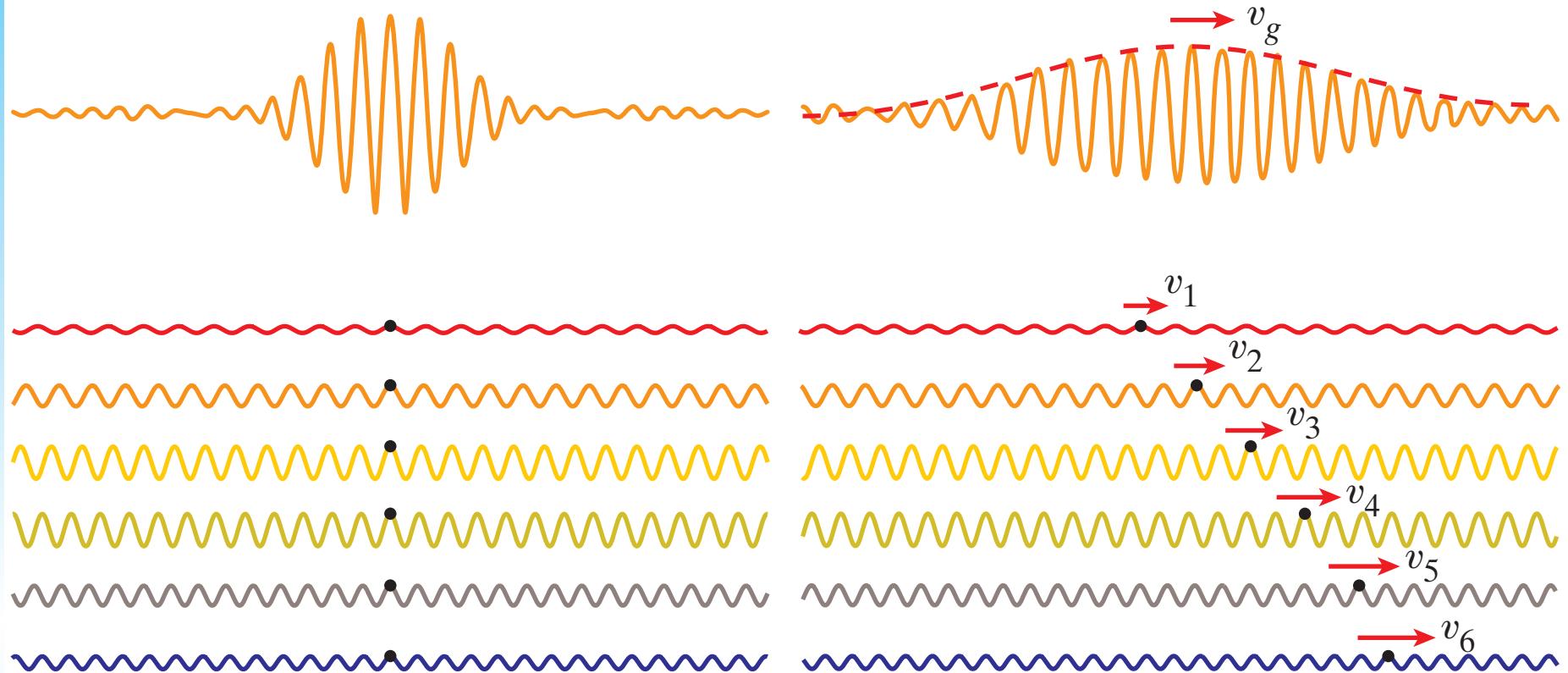
Pulse dispersion



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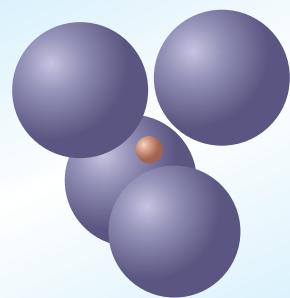
Pulse dispersion



Nonlinear optics

Linear response

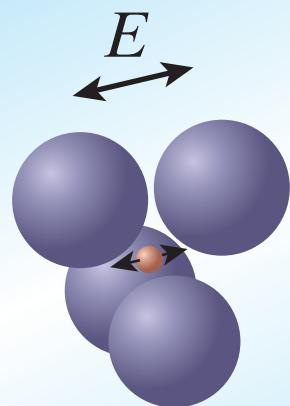
$$P(t) = \epsilon_0 \chi_e E(t)$$



Nonlinear optics

Linear response

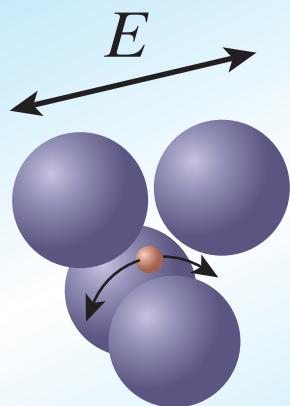
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Nonlinear optics

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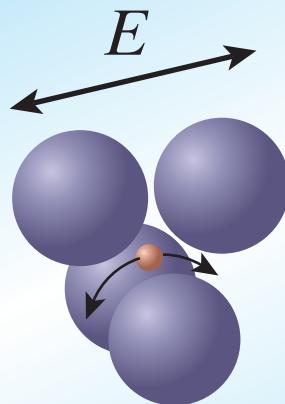
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Nonlinear optics

Linear response

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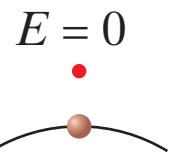
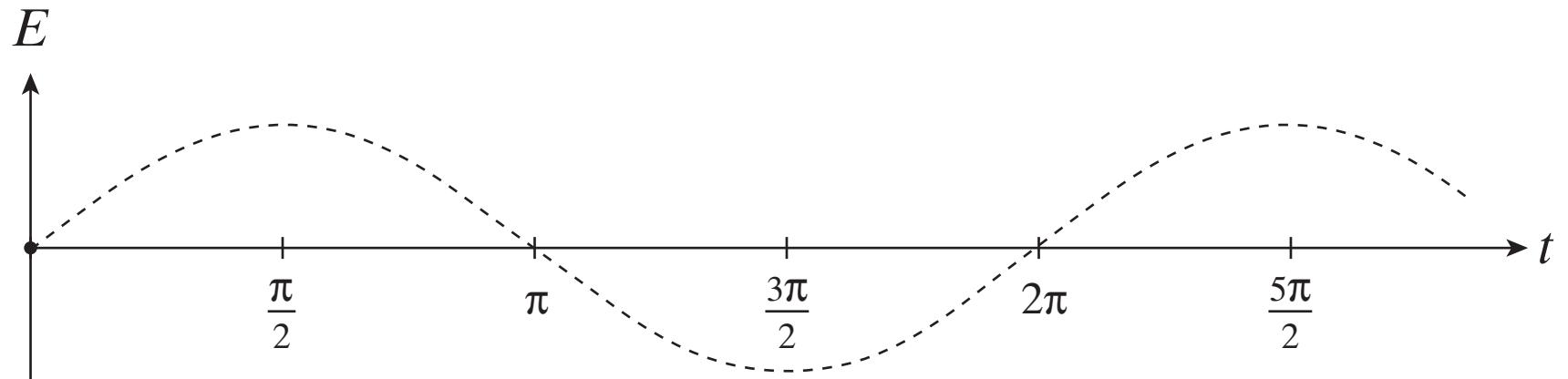


Nonlinear polarization:

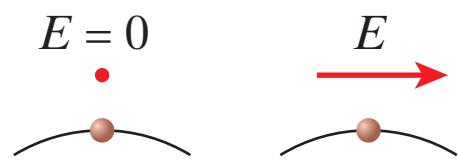
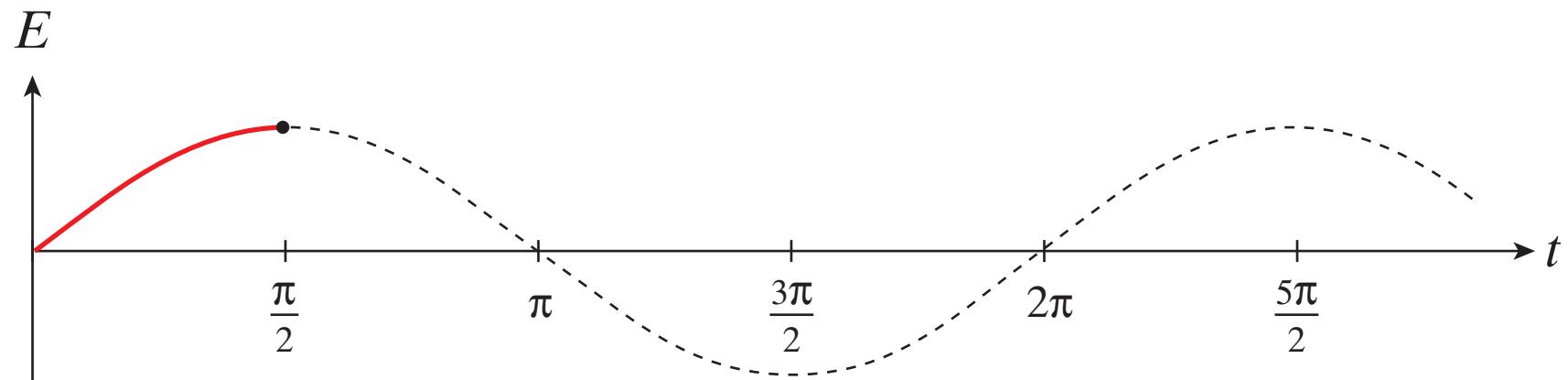
$$P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + \dots$$

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

Nonlinear optics

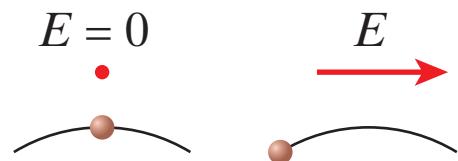
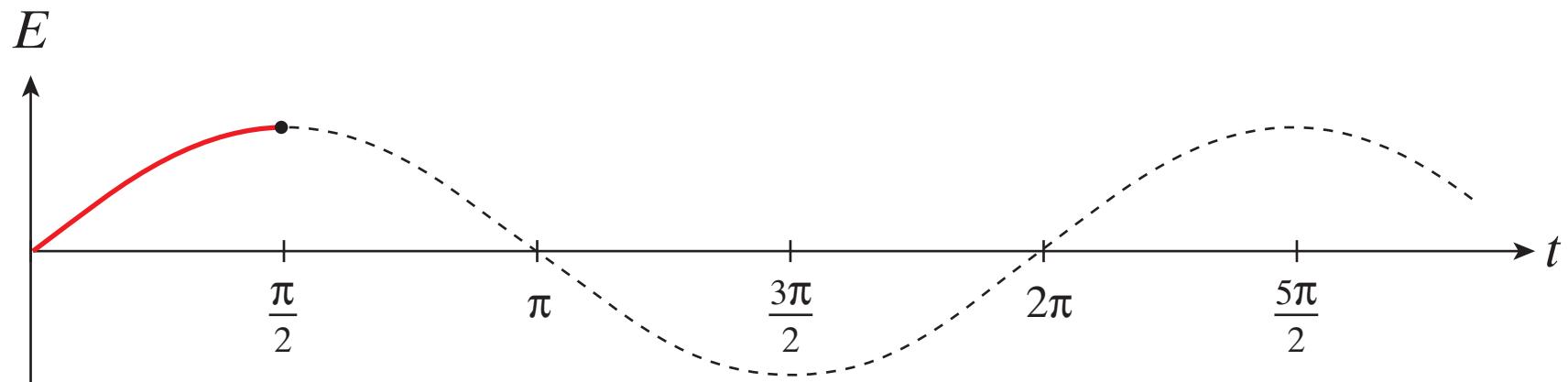


Nonlinear optics

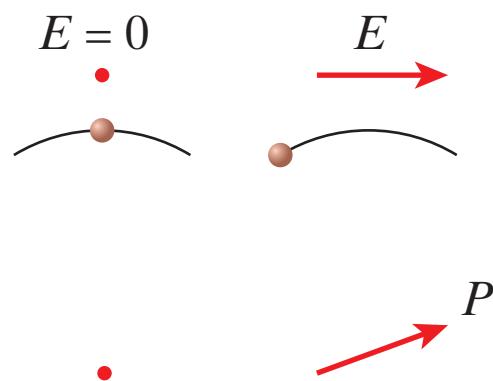
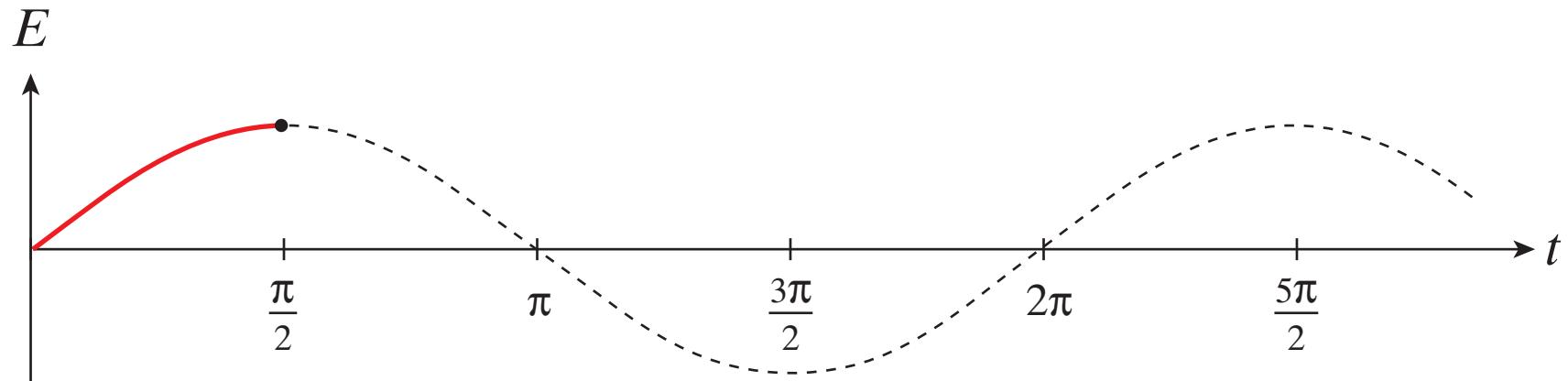


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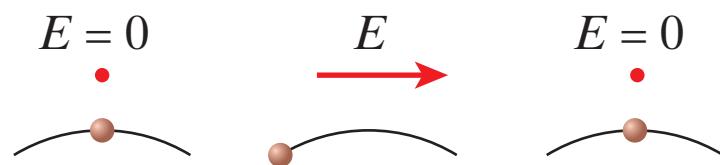
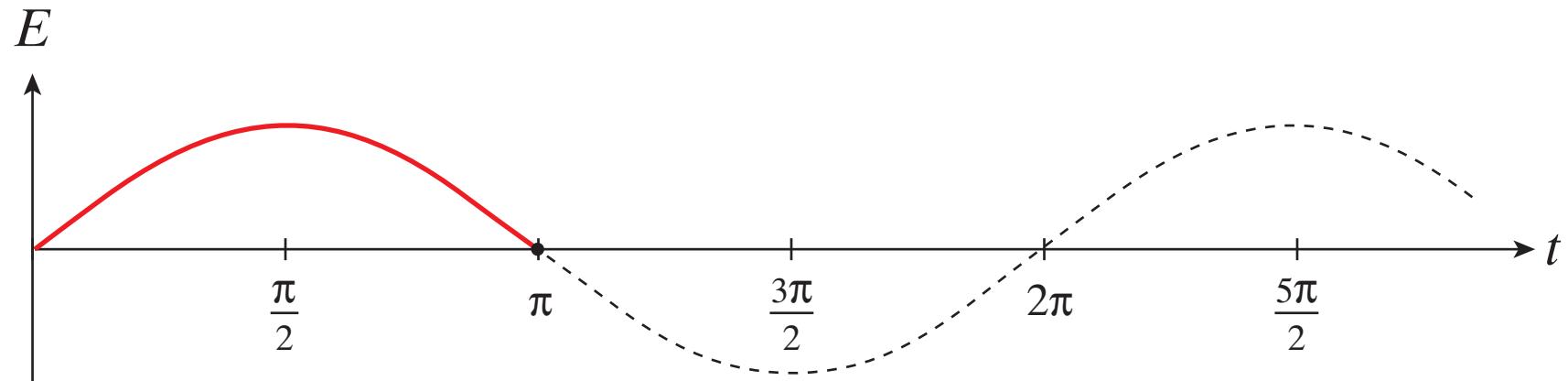
Nonlinear optics



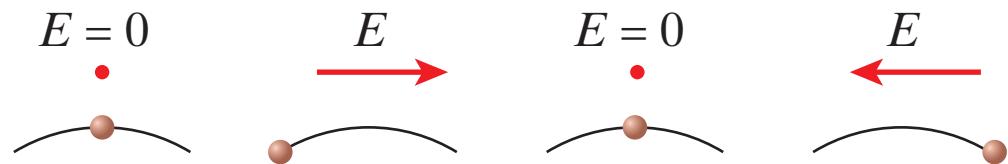
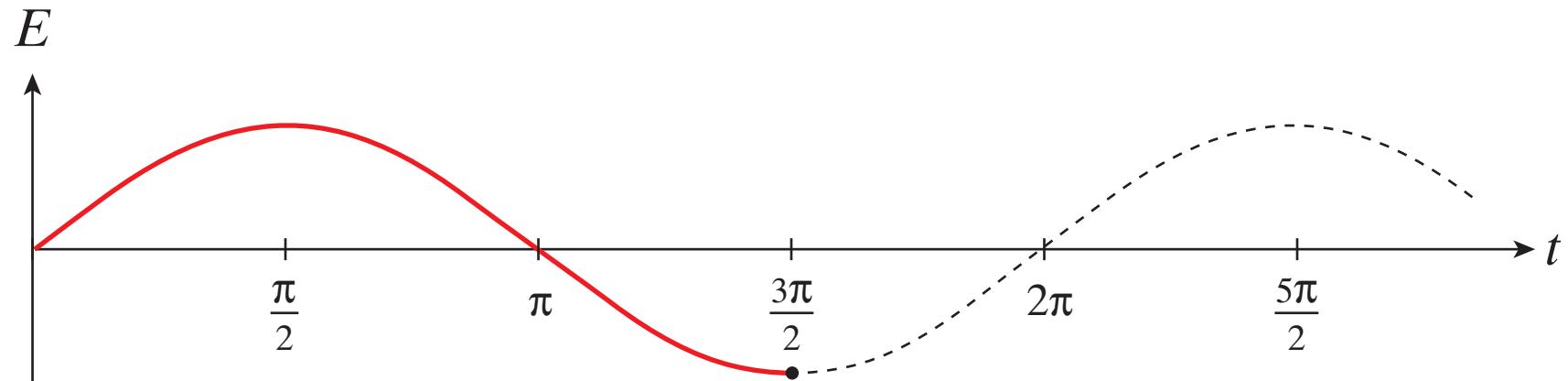
Nonlinear optics



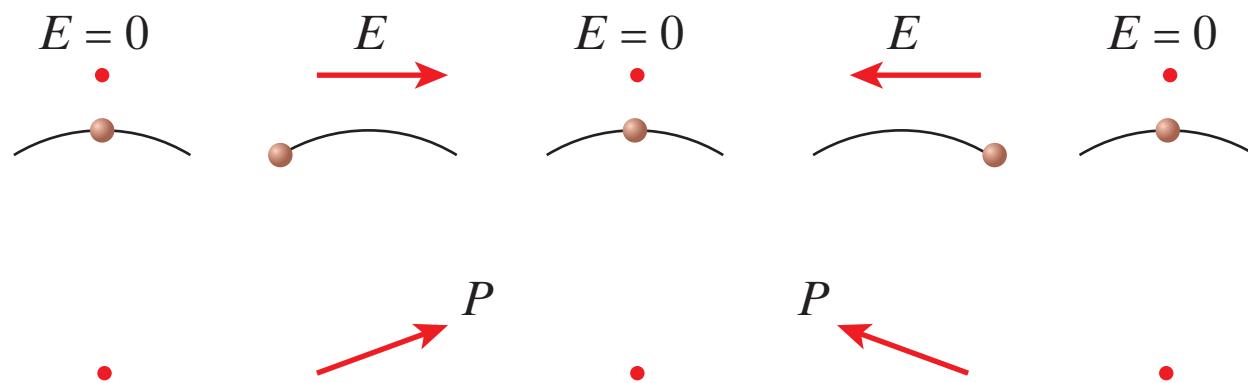
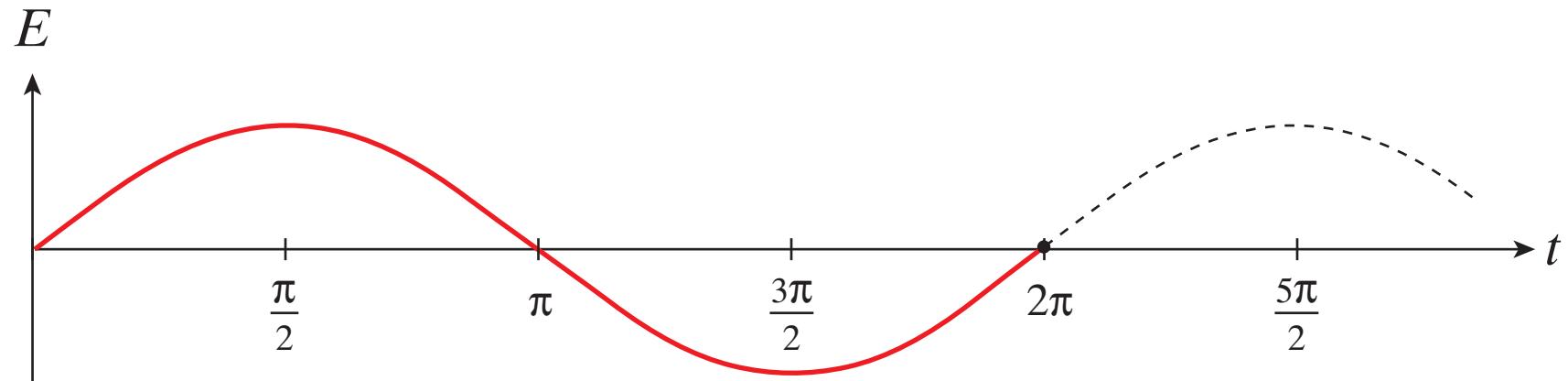
Nonlinear optics



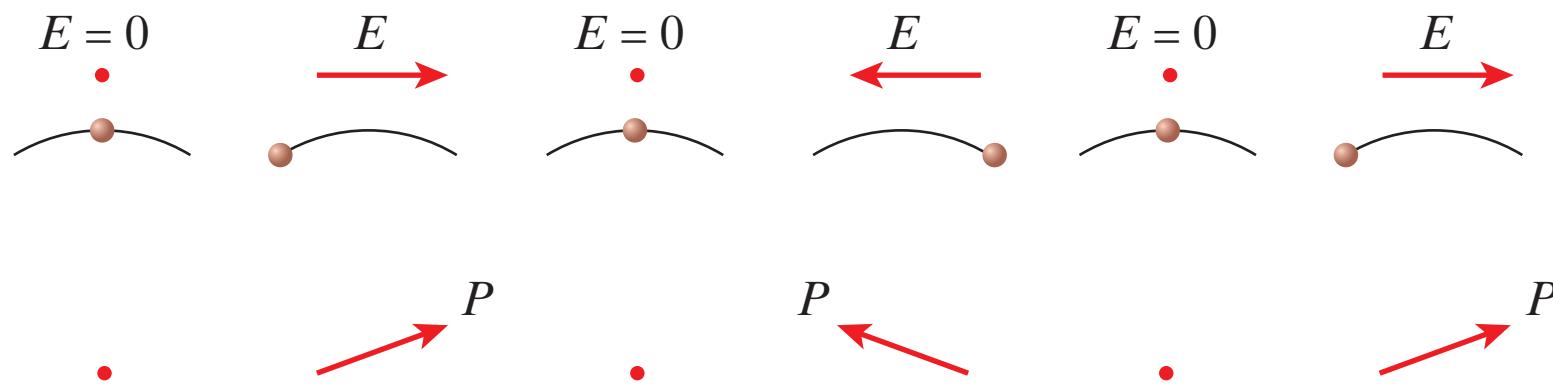
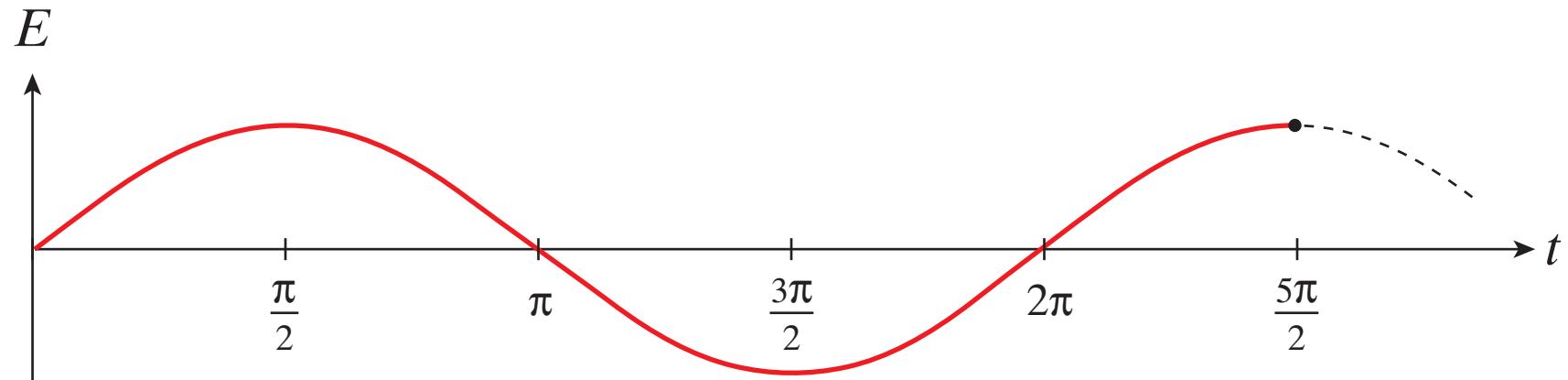
Nonlinear optics



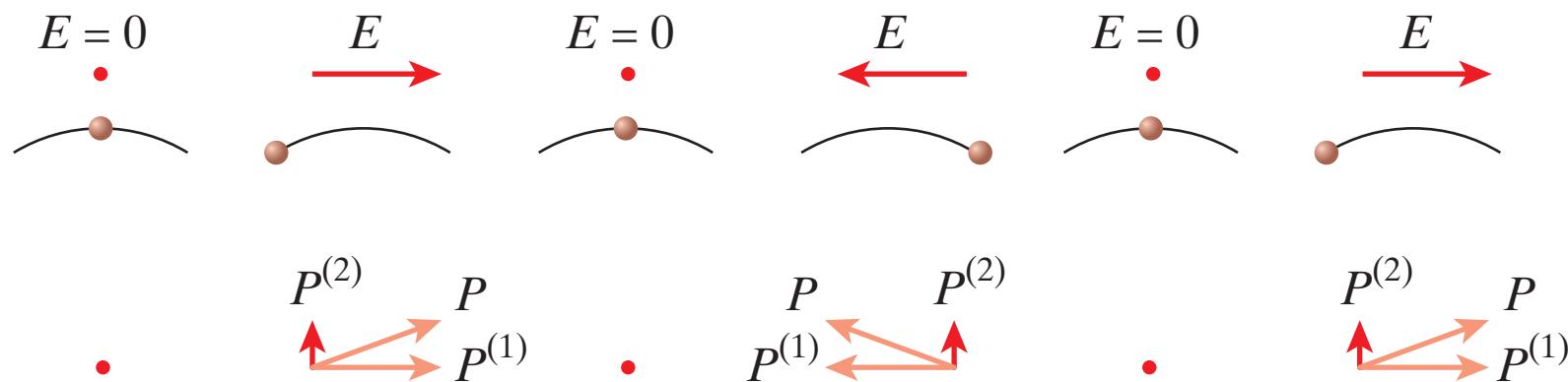
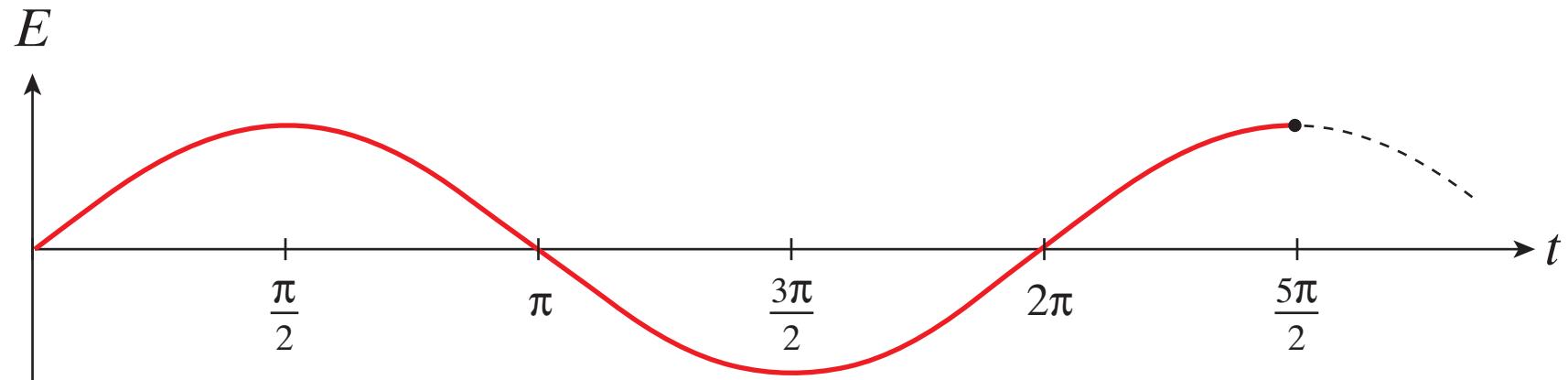
Nonlinear optics



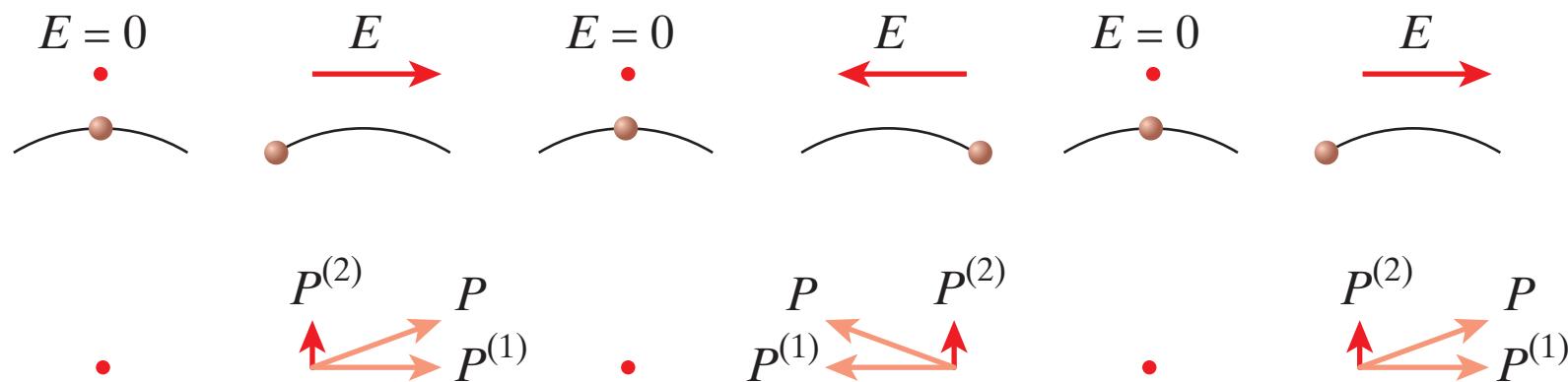
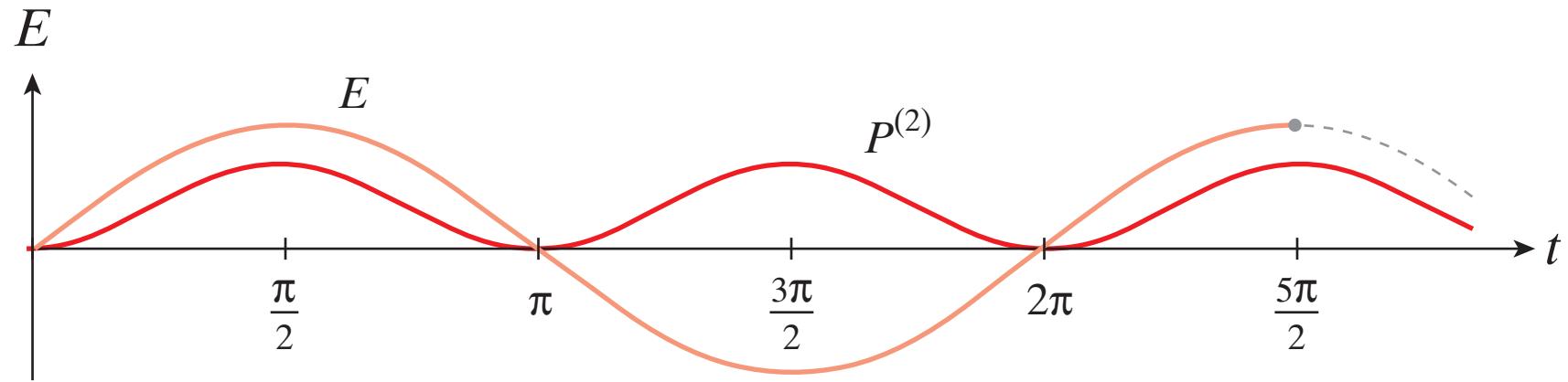
Nonlinear optics



Nonlinear optics



Nonlinear optics



In medium with inversion symmetry

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

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and so

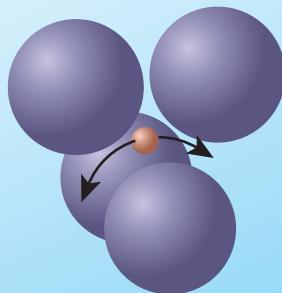
$$\chi^{(2)} = -\chi^{(2)} = 0$$

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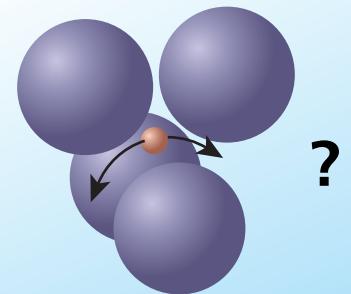
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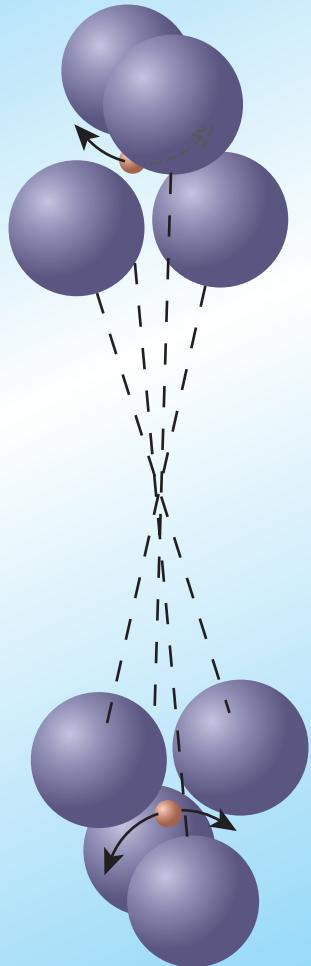
... but ...

Nonlinear optics

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



Nonlinear optics



Nonlinear optics

Nonlinear polarization:

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

Nonlinear optics

Nonlinear polarization:

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

Third order polarization

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

Nonlinear optics

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Nonlinear optics

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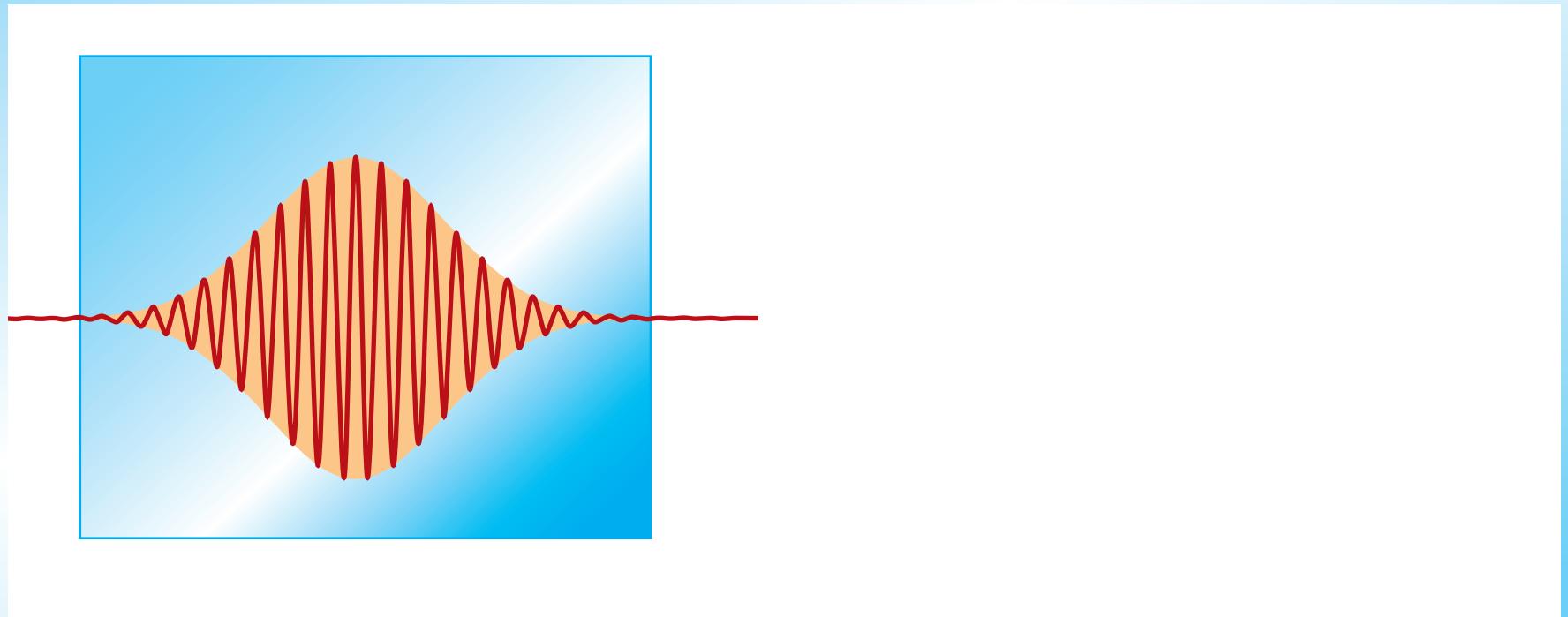
and so $P = P^{(1)} + P^{(3)} = (\chi^{(1)} + \chi^{(3)}I)E \equiv \chi_{eff}E$

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

Nonlinear optics

Intensity dependent index of refraction:

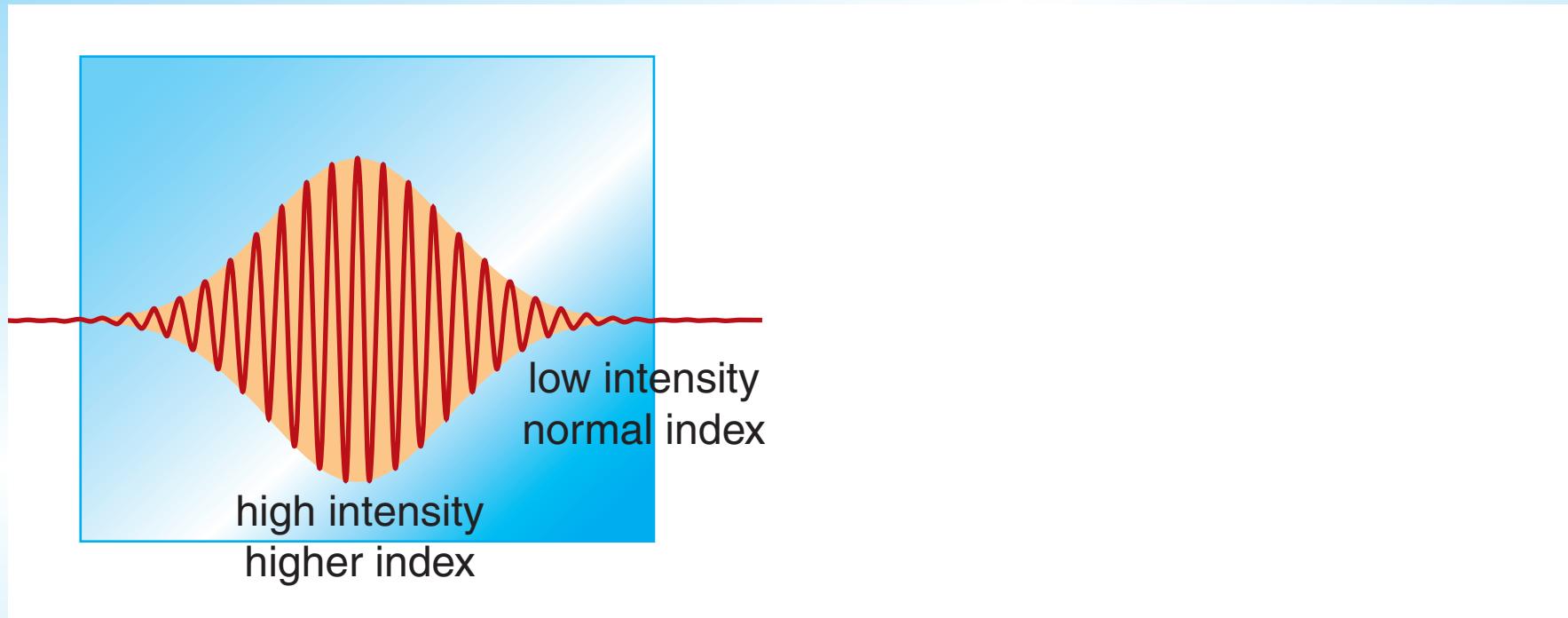
$$n = n_o + n_2 I$$



Nonlinear optics

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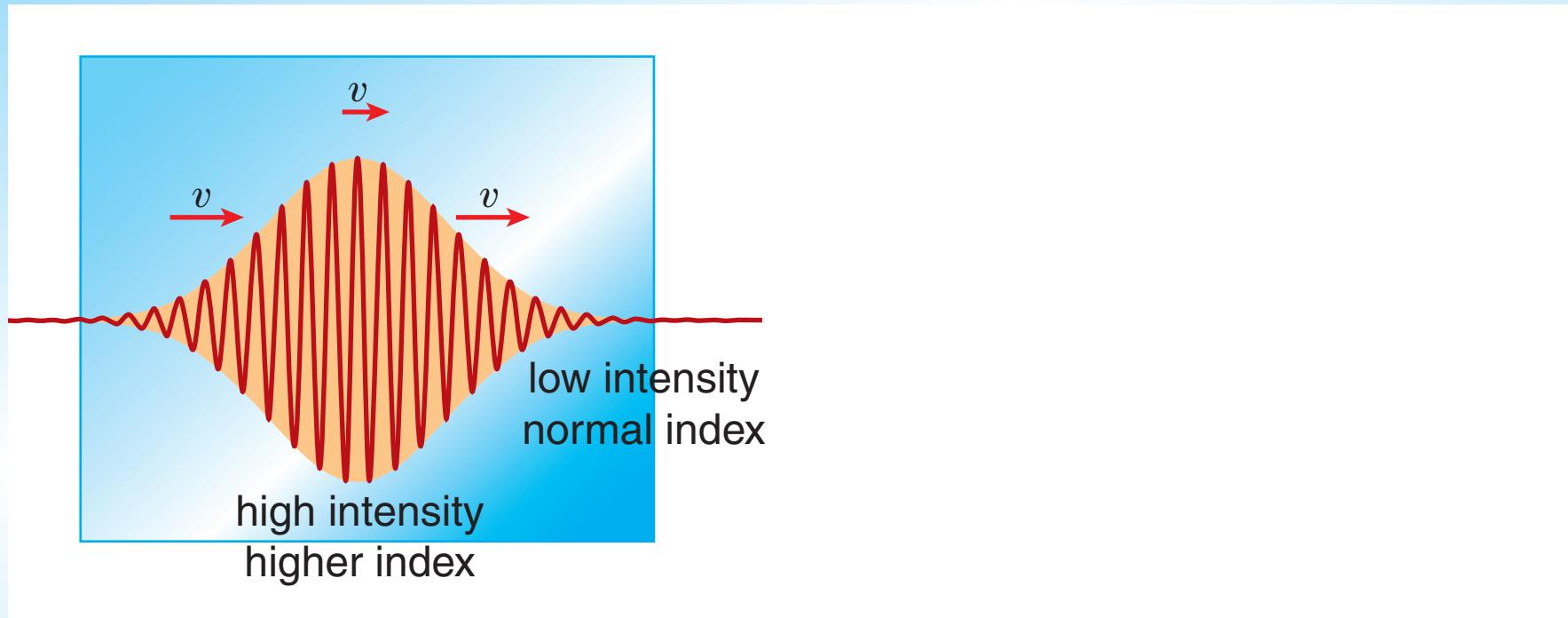
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Nonlinear optics

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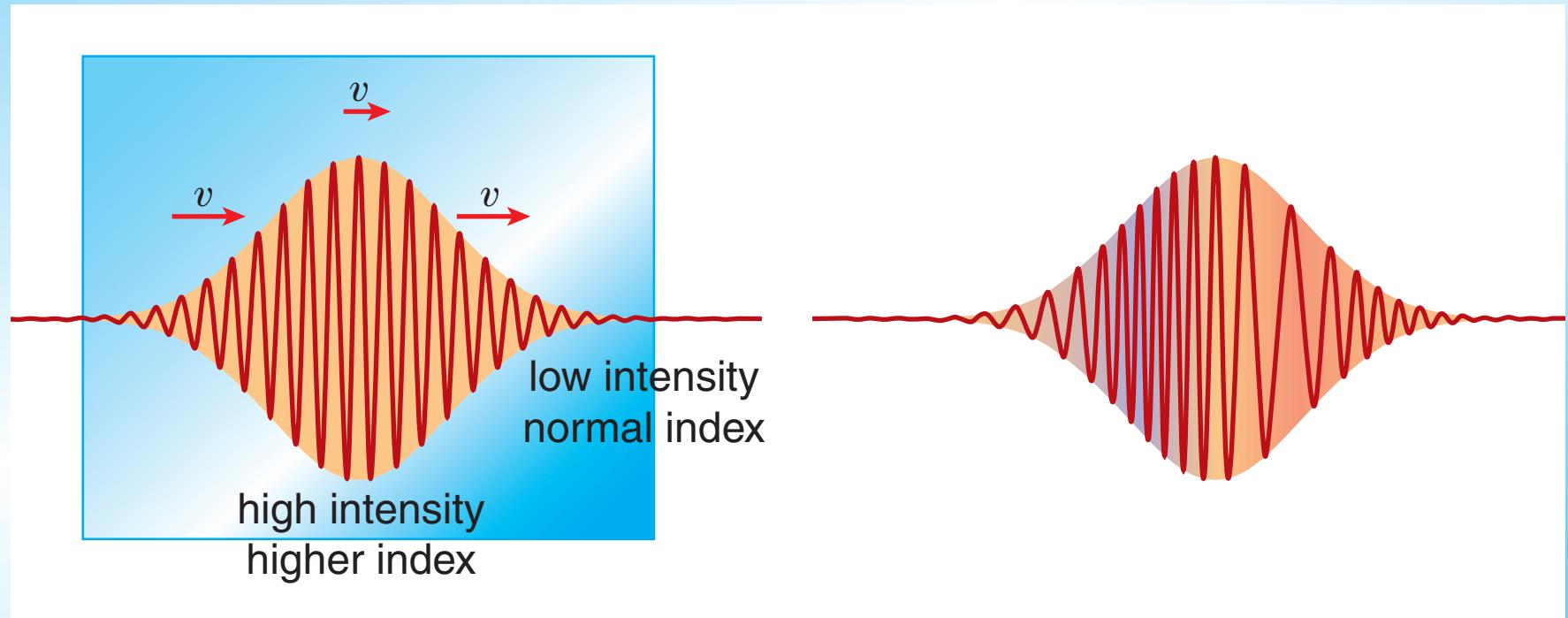
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Nonlinear optics

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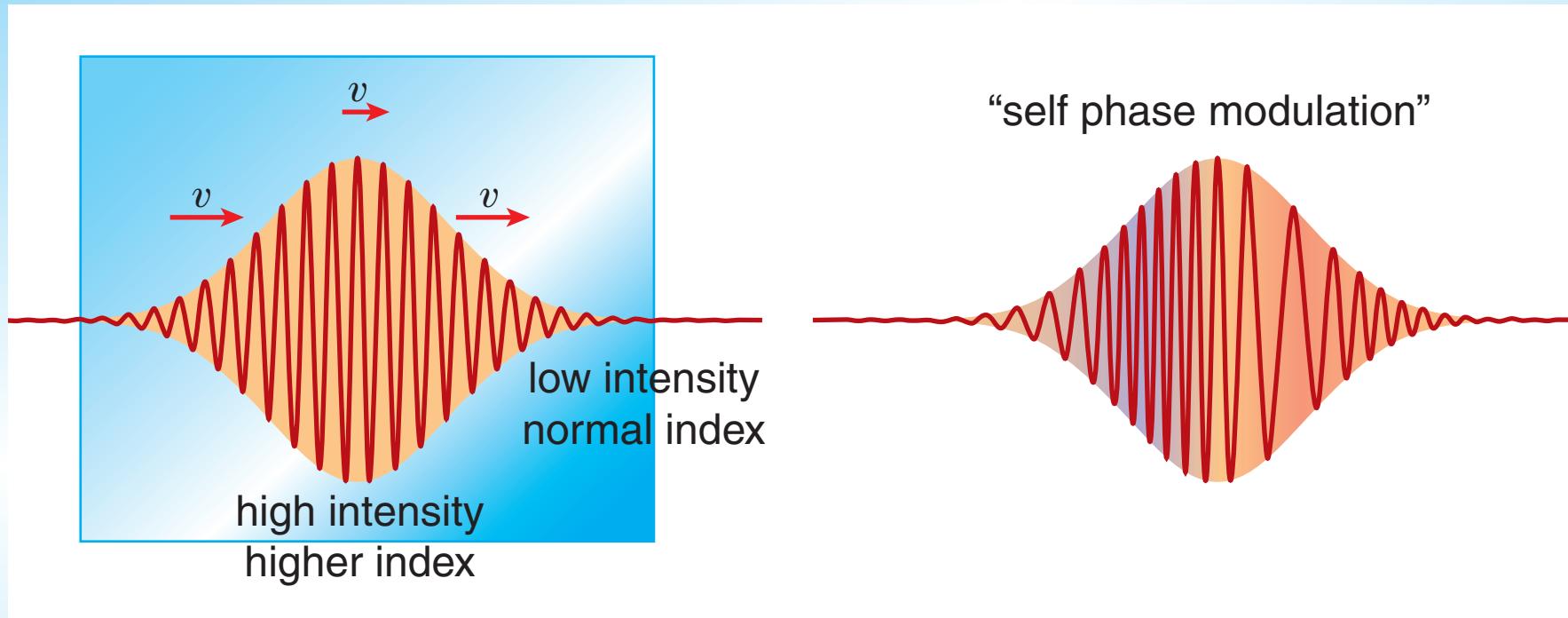
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Nonlinear optics

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Nonlinear optics

Phase:

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

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Nonlinear optics

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Frequency change:

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

Nonlinear optics

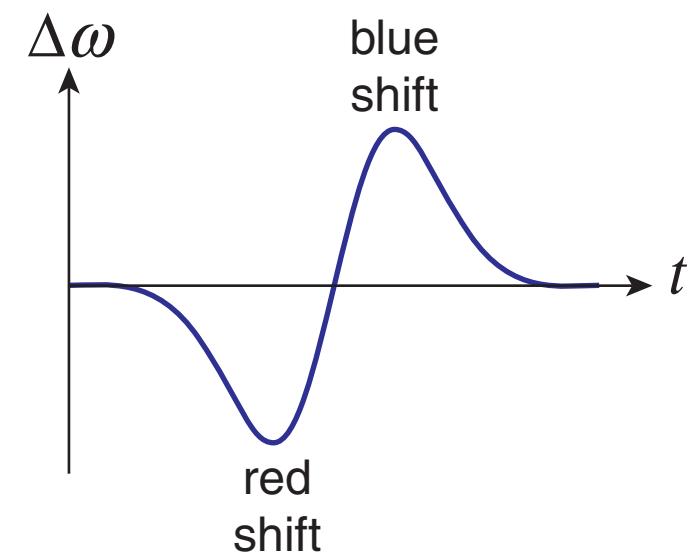
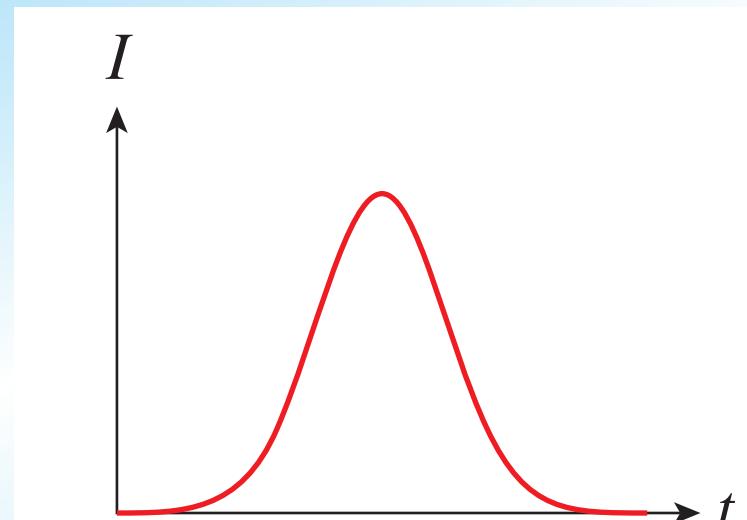
Phase:

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

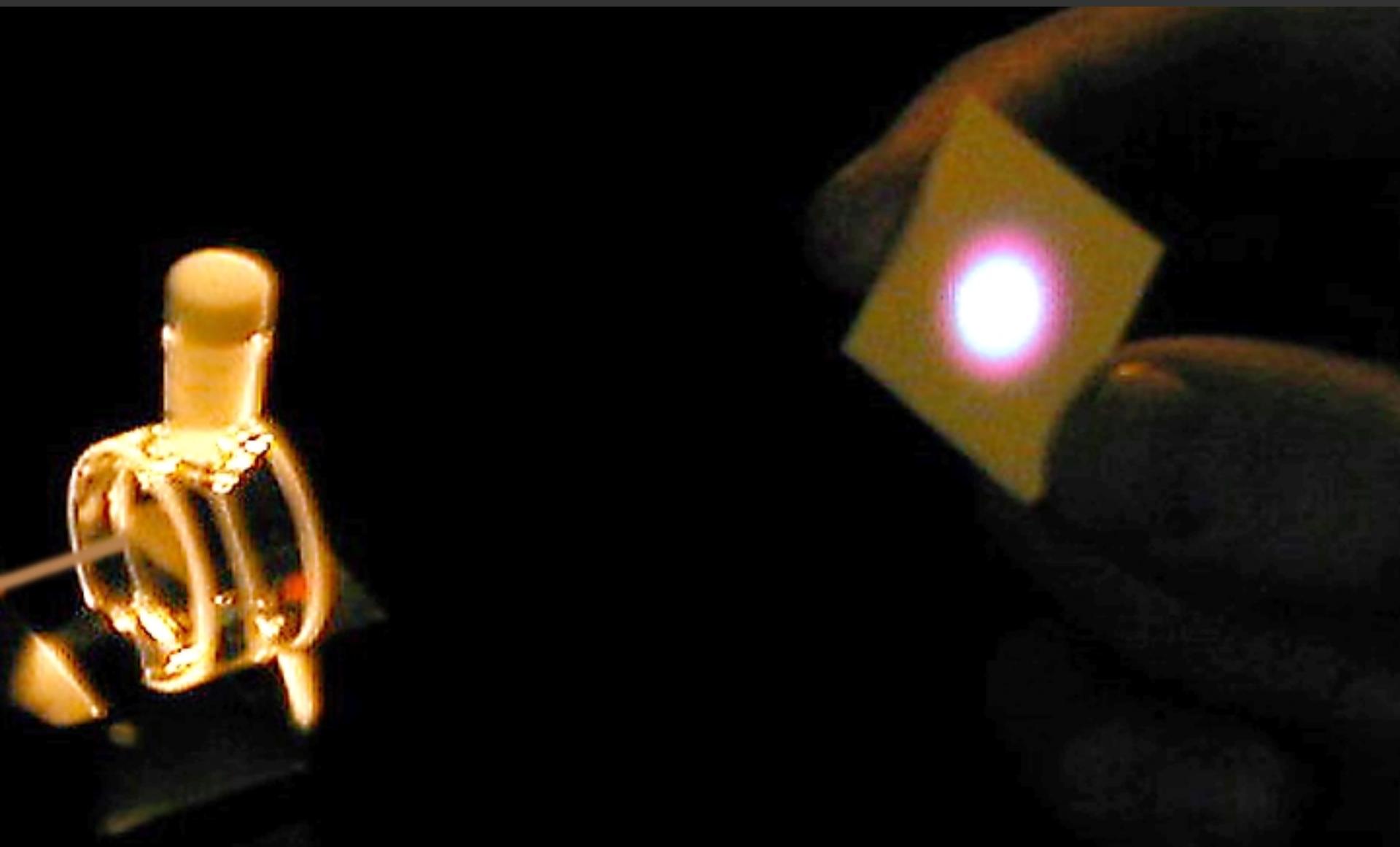
$$\phi = \frac{2\pi}{\lambda} L(n_o + n_2 I)$$

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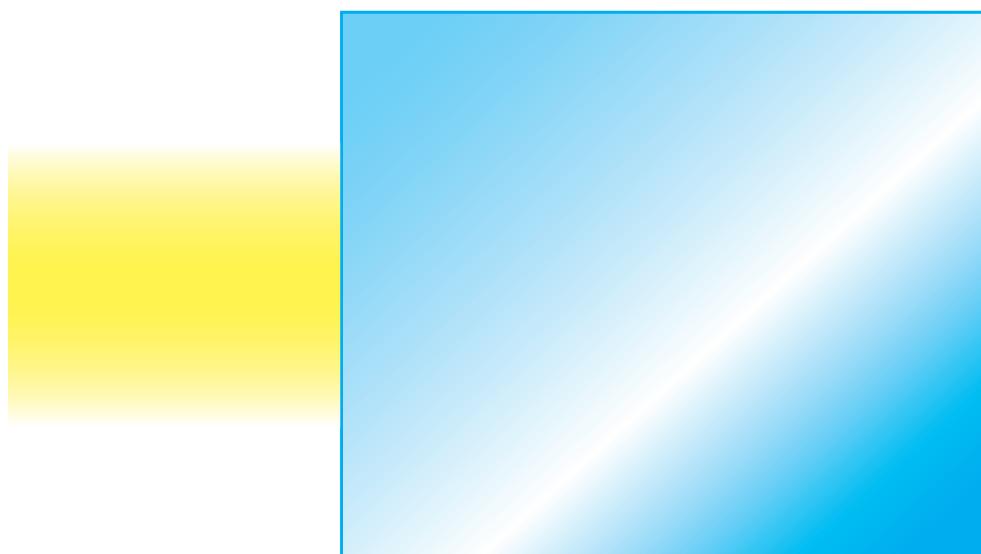


Nonlinear optics



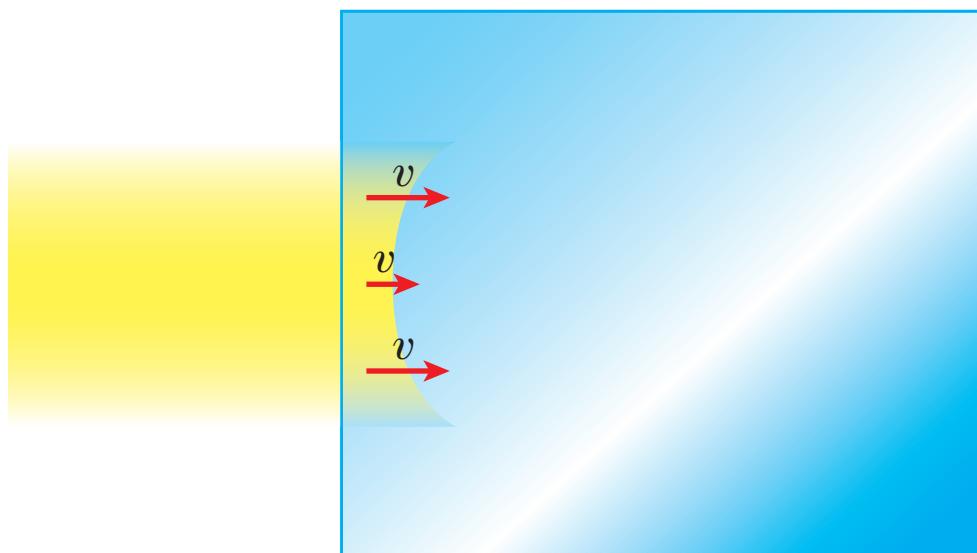
Nonlinear optics

Spatial intensity profile...



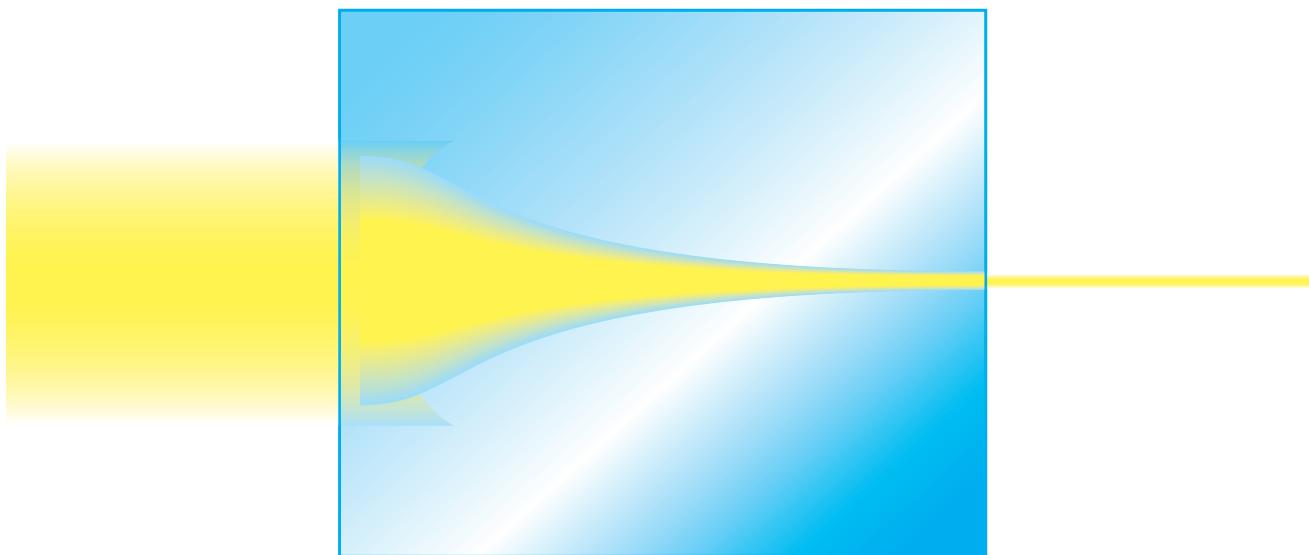
Nonlinear optics

Spatial intensity profile...

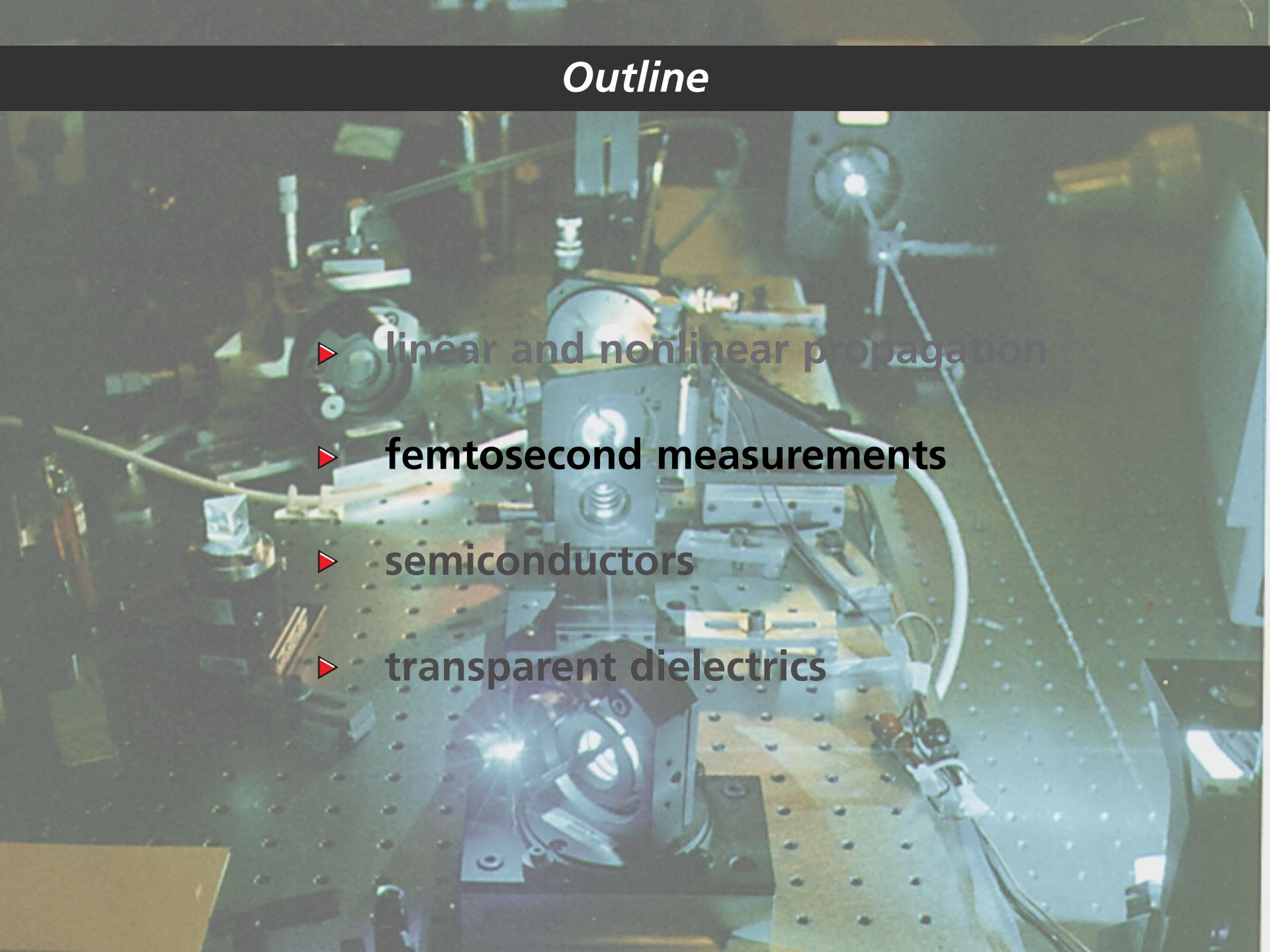


Nonlinear optics

...causes self-focusing

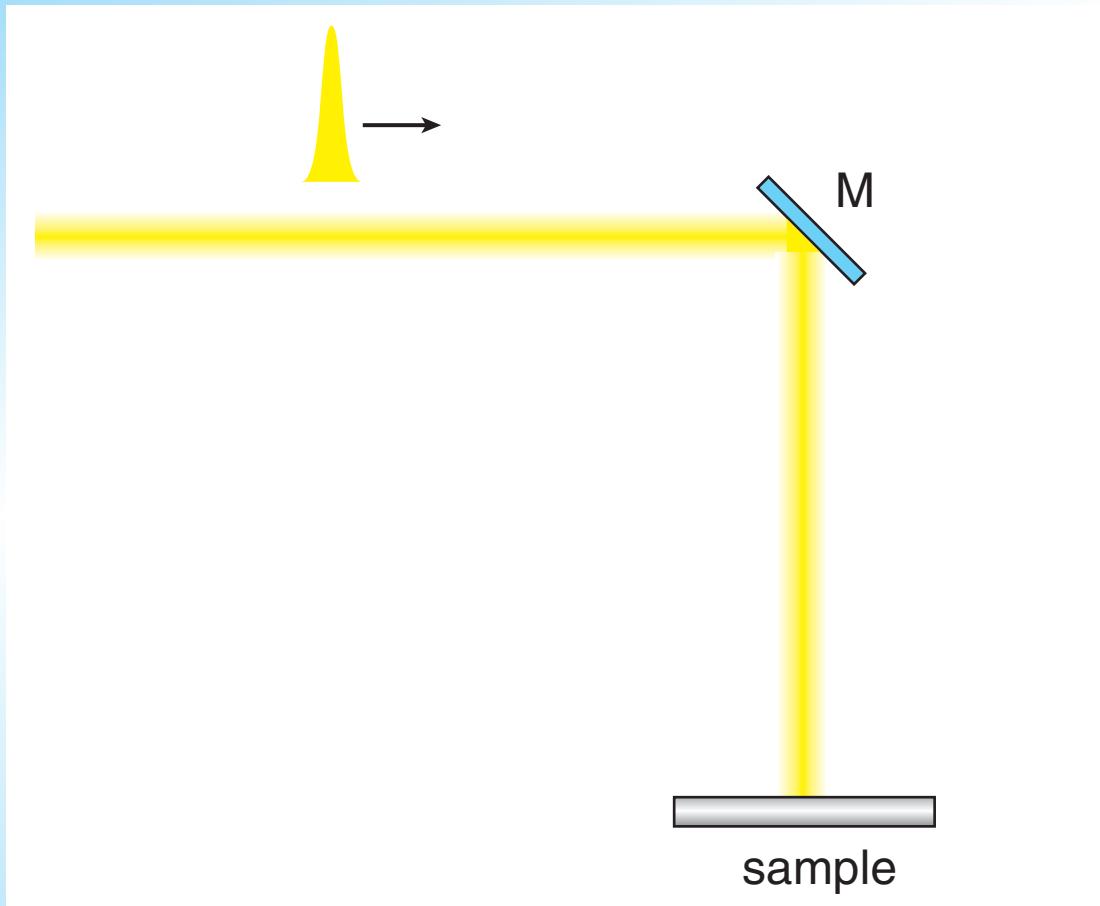


Outline

- 
- ▶ linear and nonlinear propagation
 - ▶ femtosecond measurements
 - ▶ semiconductors
 - ▶ transparent dielectrics

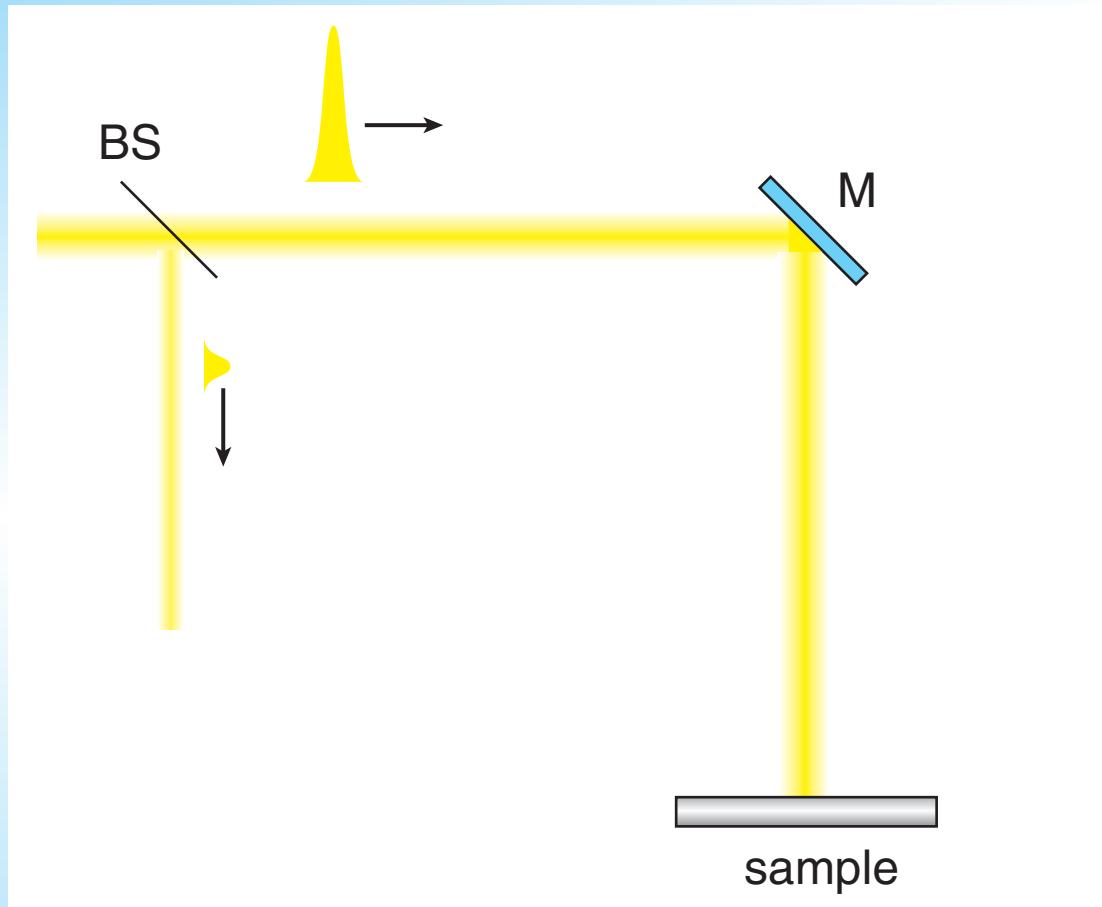
Introduction

How to measure on the femtosecond time scale?



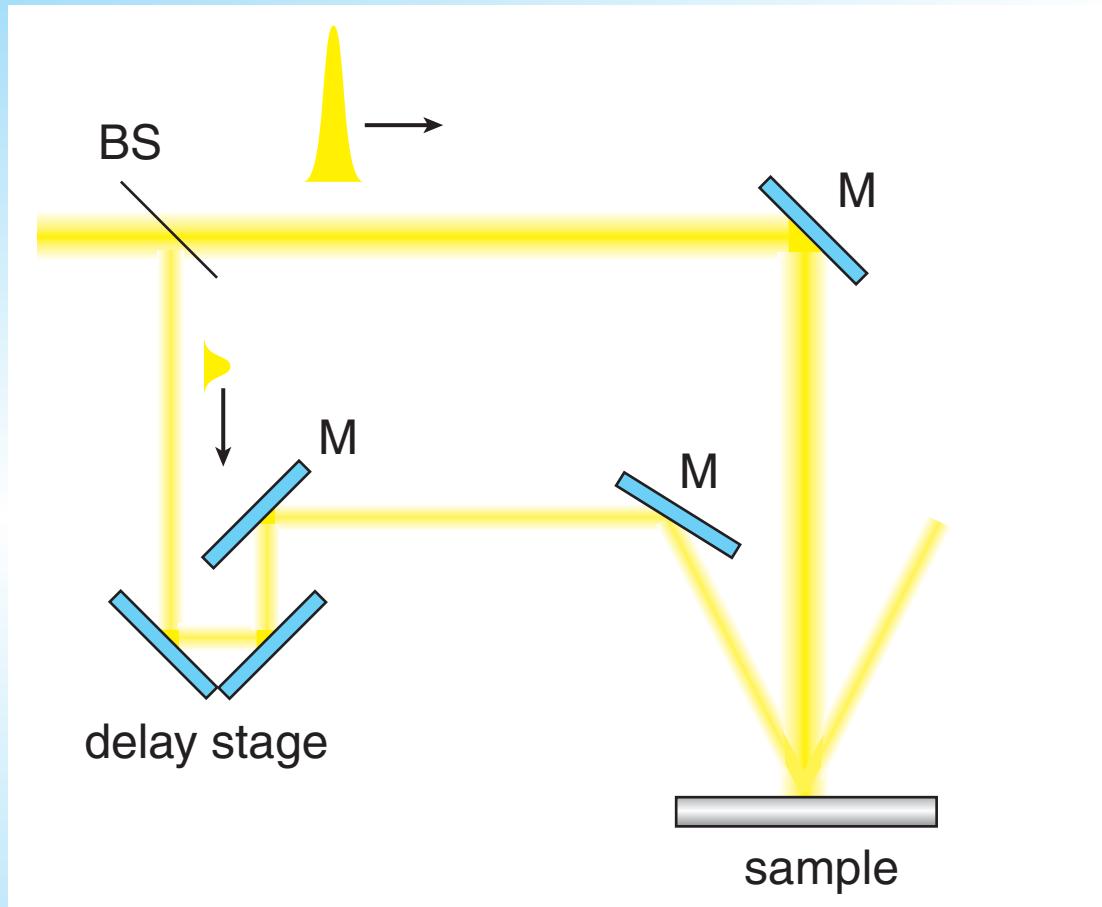
Introduction

Use pump-probe technique



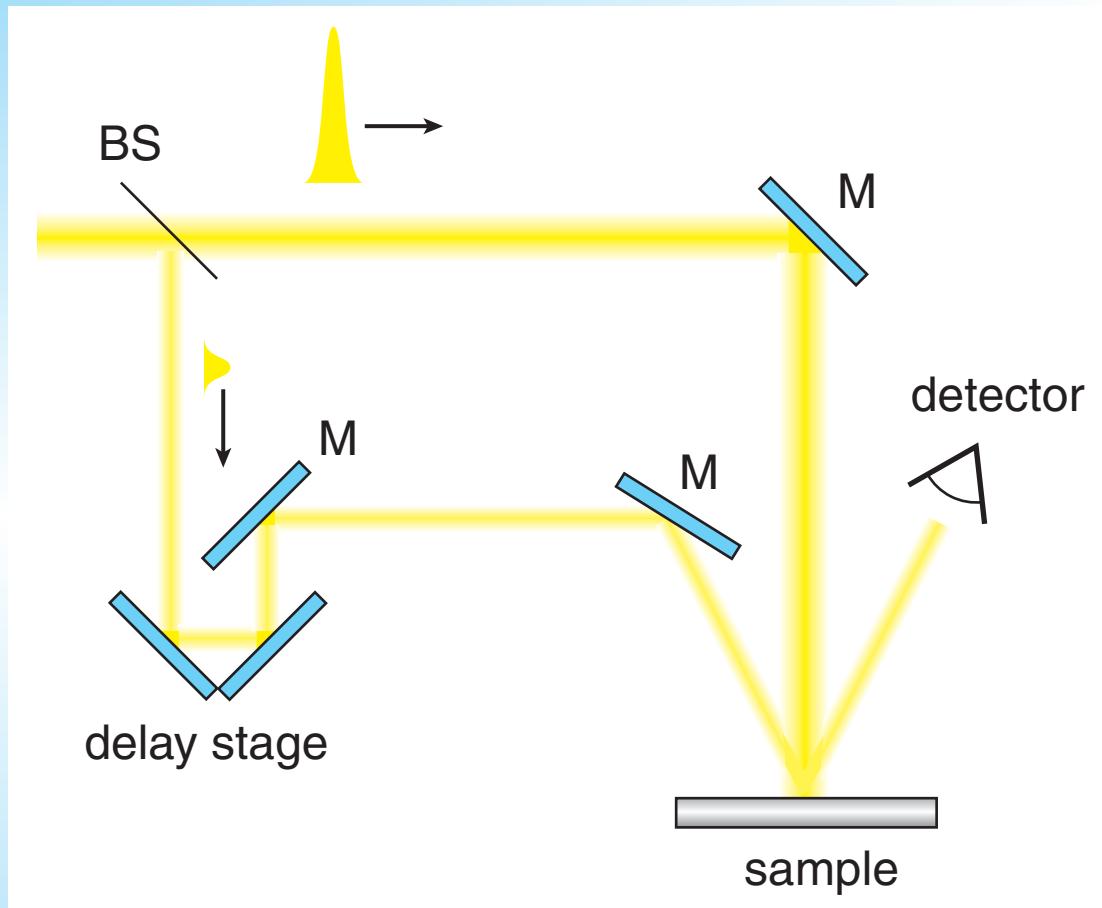
Introduction

Use pump-probe technique



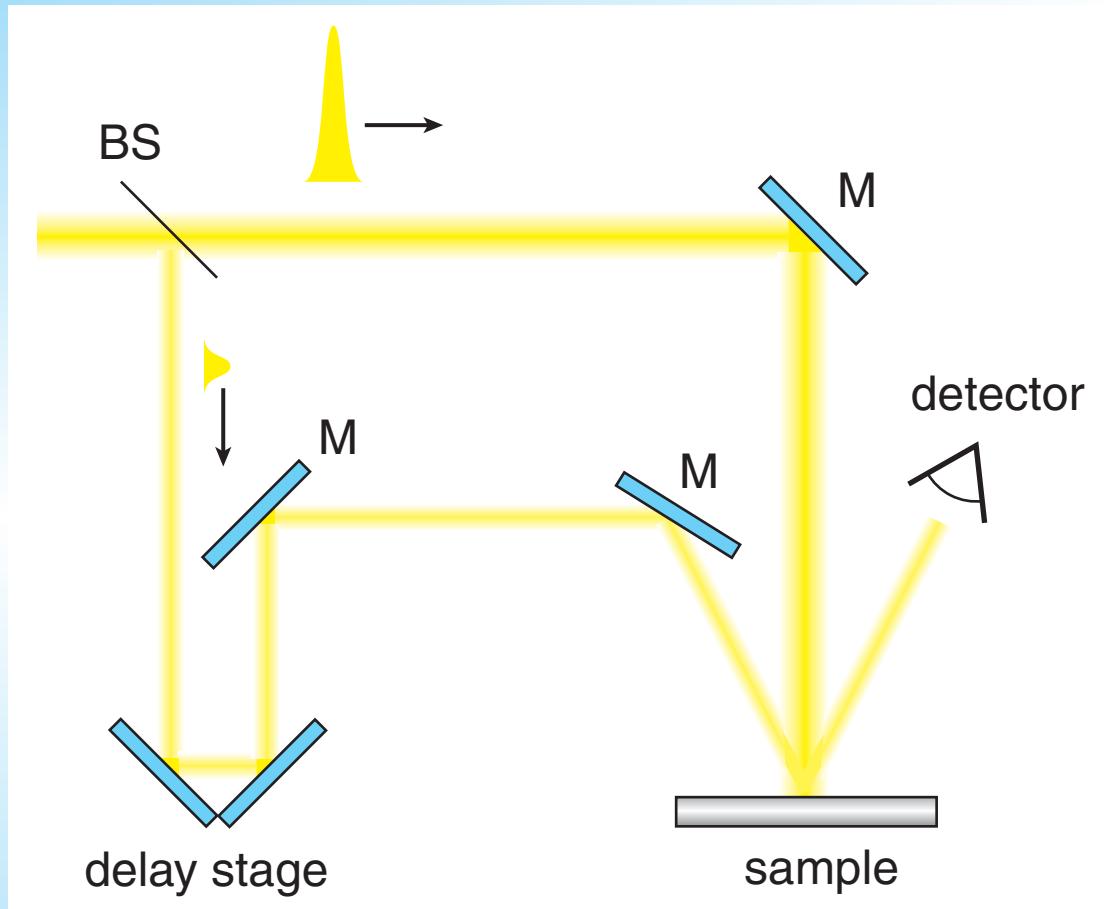
Introduction

Use pump-probe technique



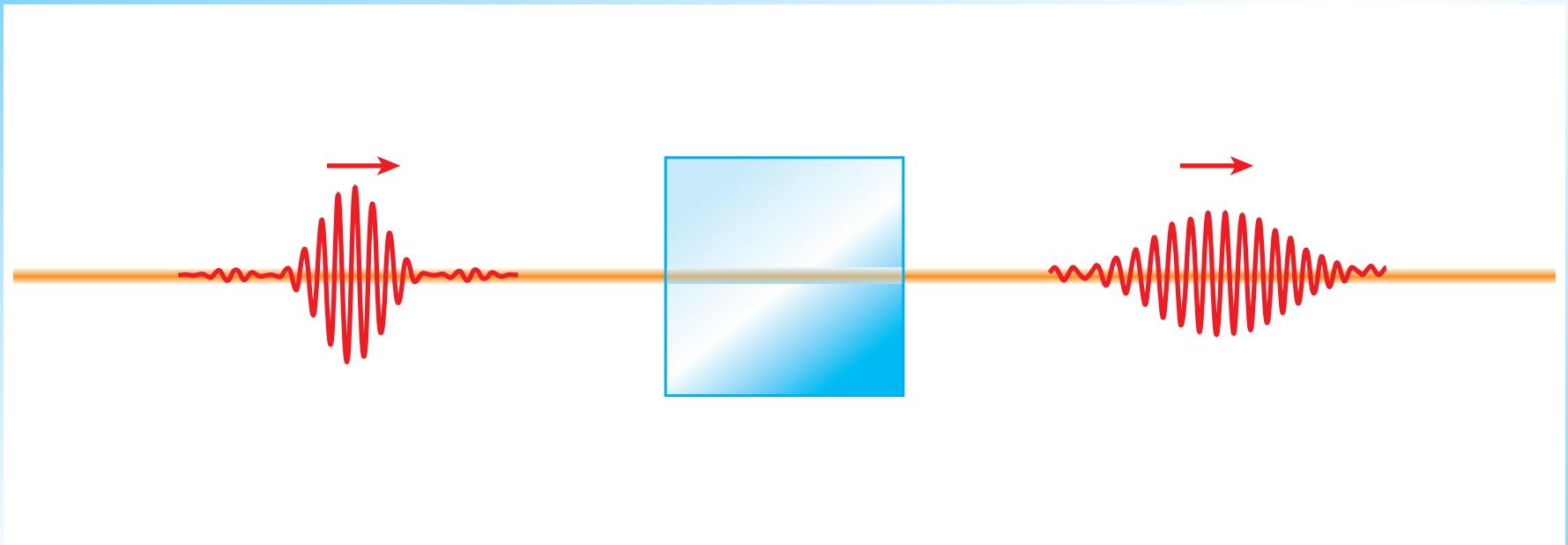
Introduction

Vary delay to get time resolution



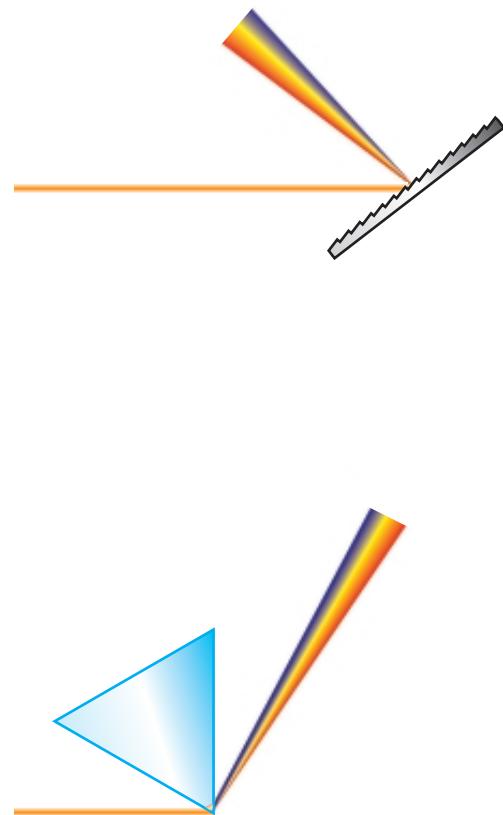
Dispersion compensation

Dispersion stretches the pulse

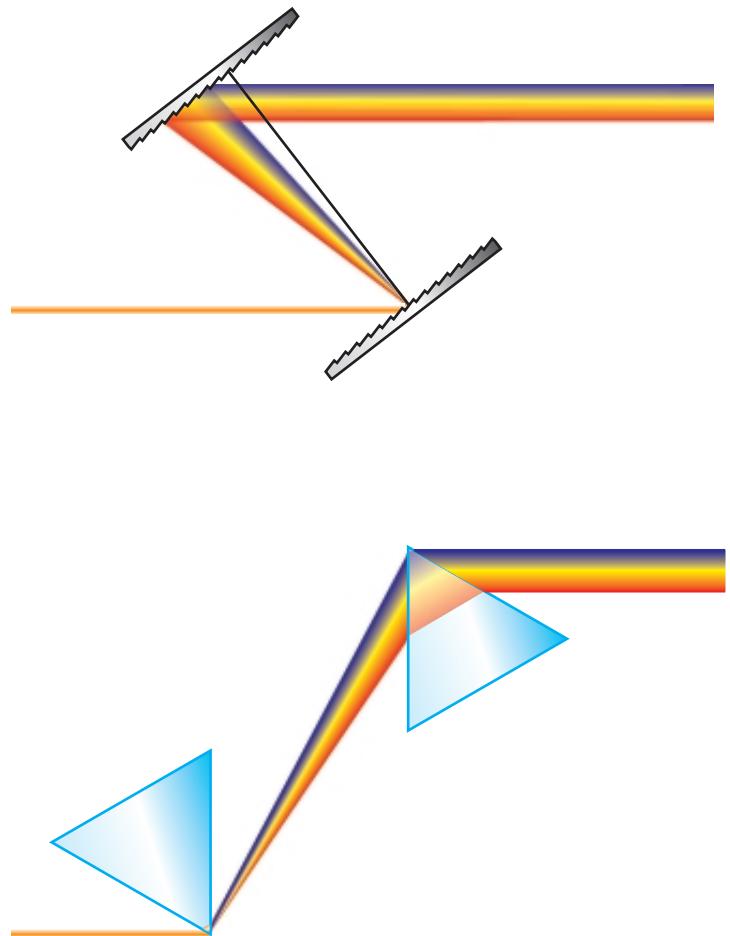


Compensate by rearranging spectral components!

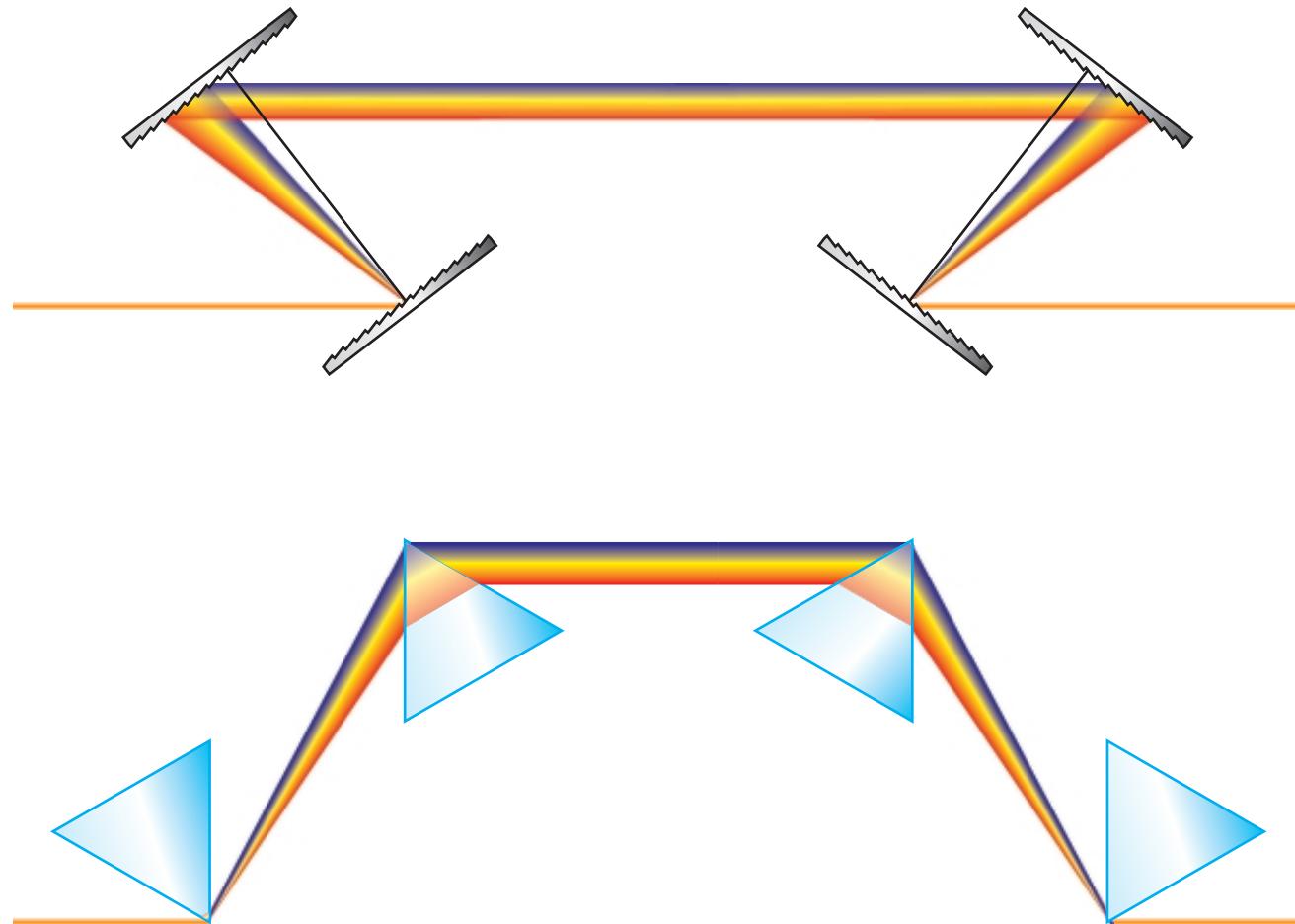
Dispersion compensation



Dispersion compensation



Dispersion compensation

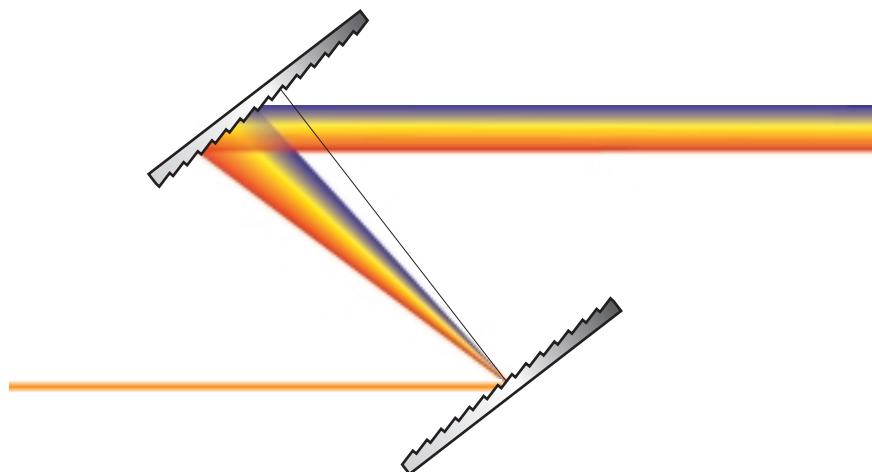


Dispersion compensation

How do these arrangements work?

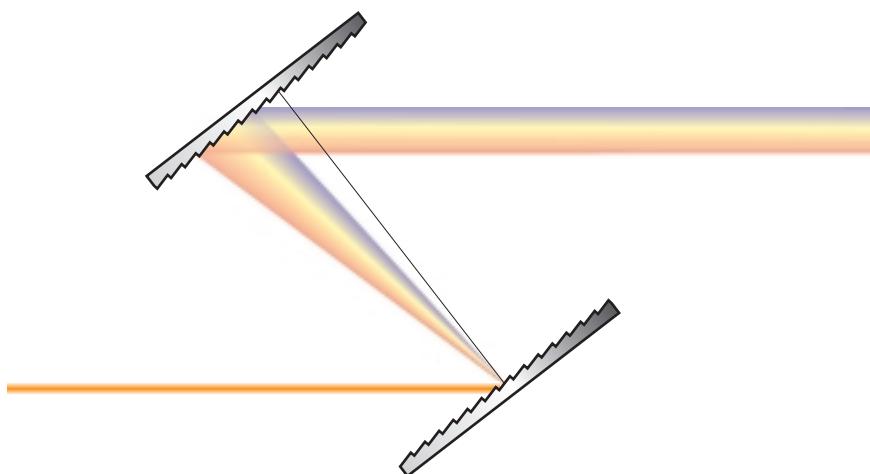
Dispersion compensation

Does path length difference compensate?



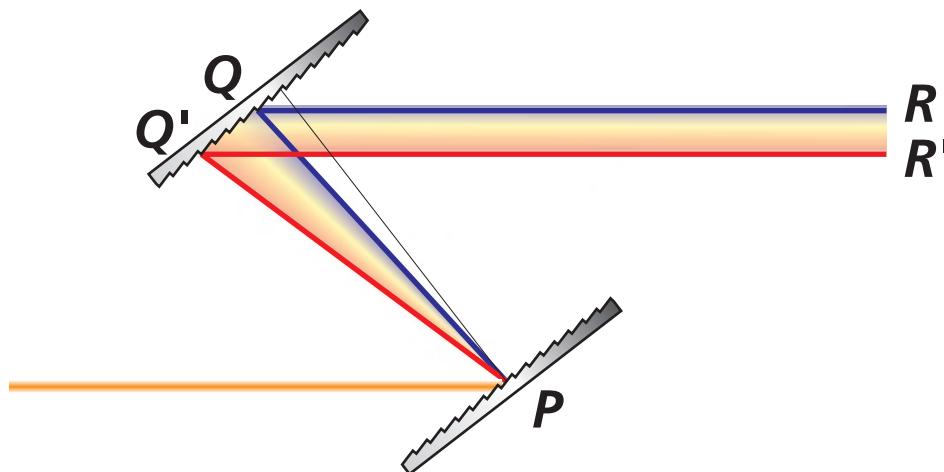
Dispersion compensation

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Dispersion compensation

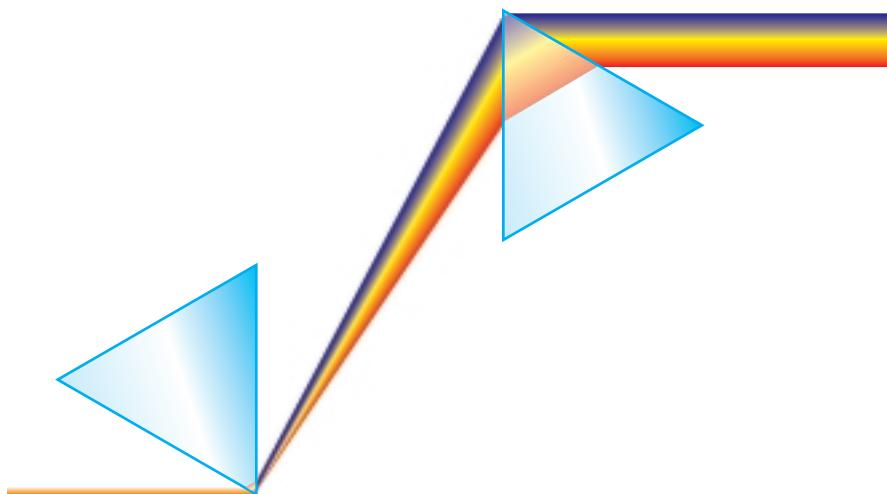
Does path length difference compensate?



Grating gives low frequency longer path length...

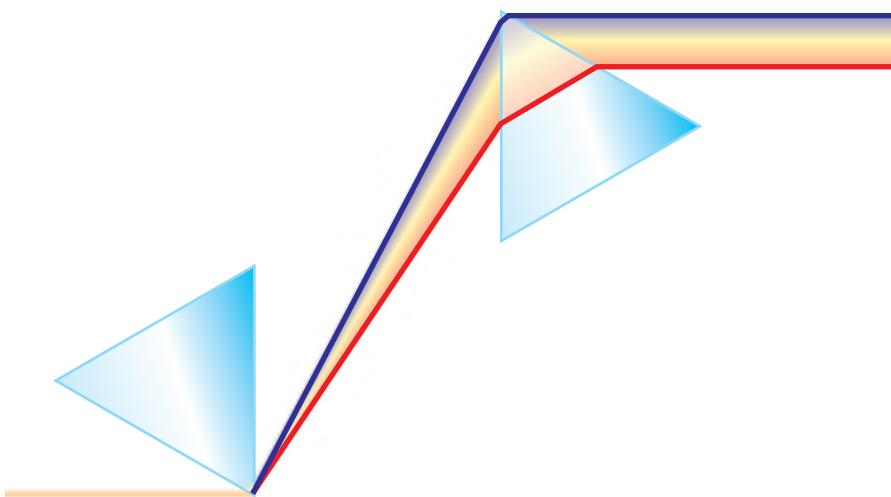
Dispersion compensation

Does path length difference compensate?



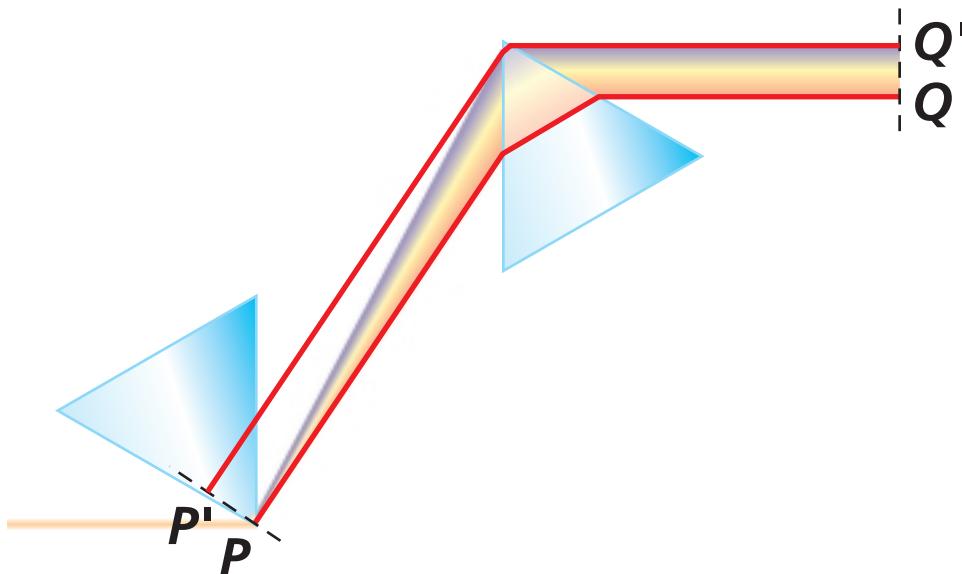
Dispersion compensation

Does path length difference compensate?



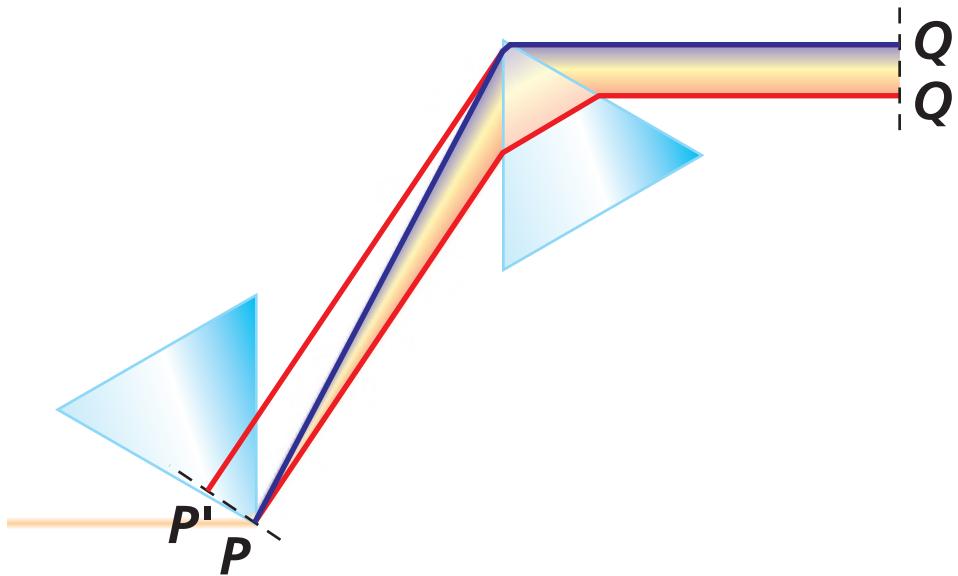
Dispersion compensation

Does path length difference compensate?



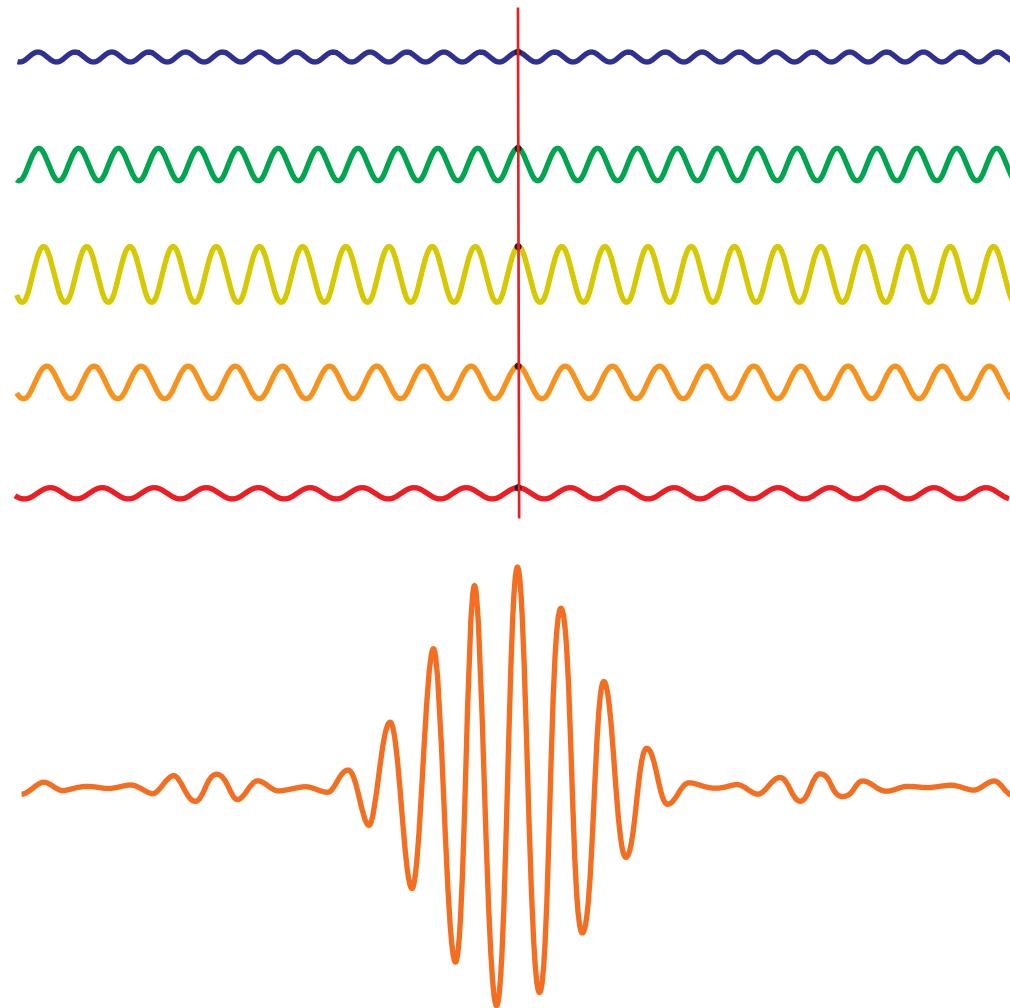
Dispersion compensation

Does path length difference compensate?

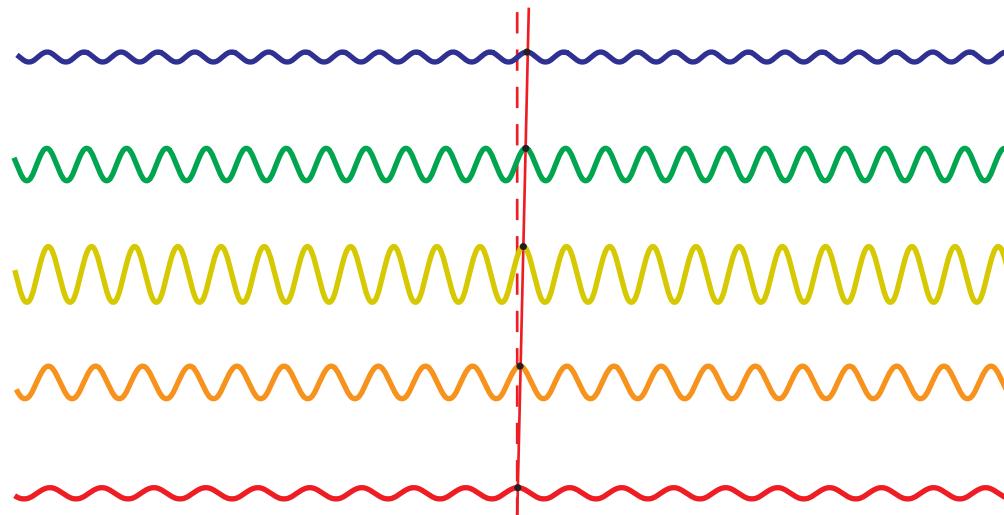


...so prism gives low frequency *shorter* path length...

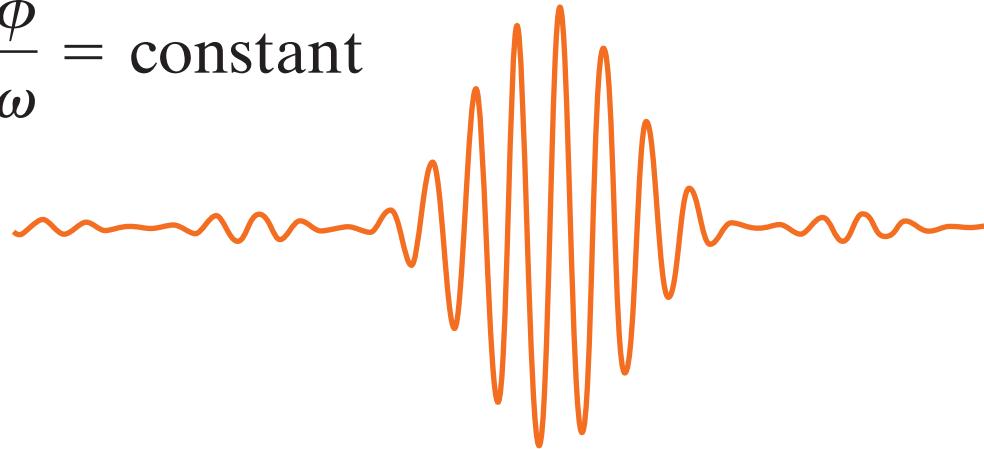
Dispersion compensation



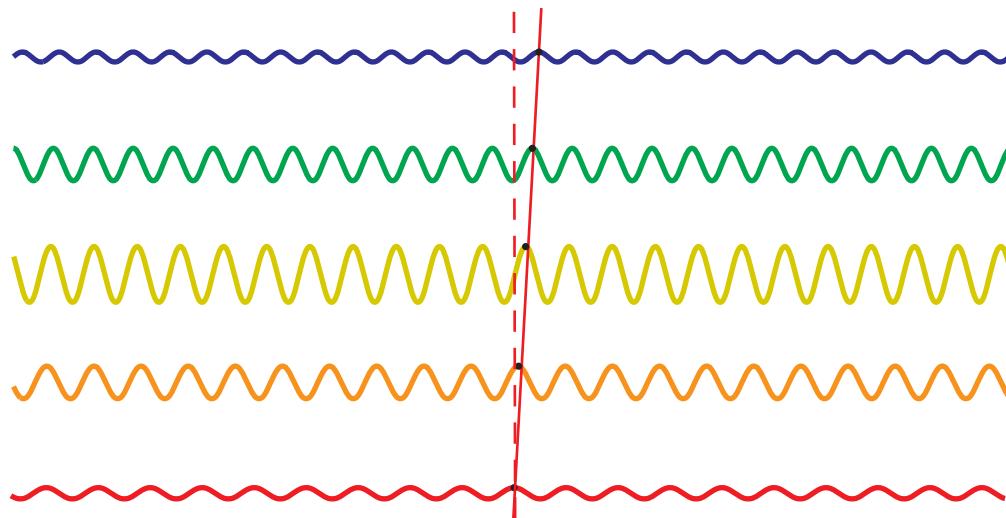
Dispersion compensation



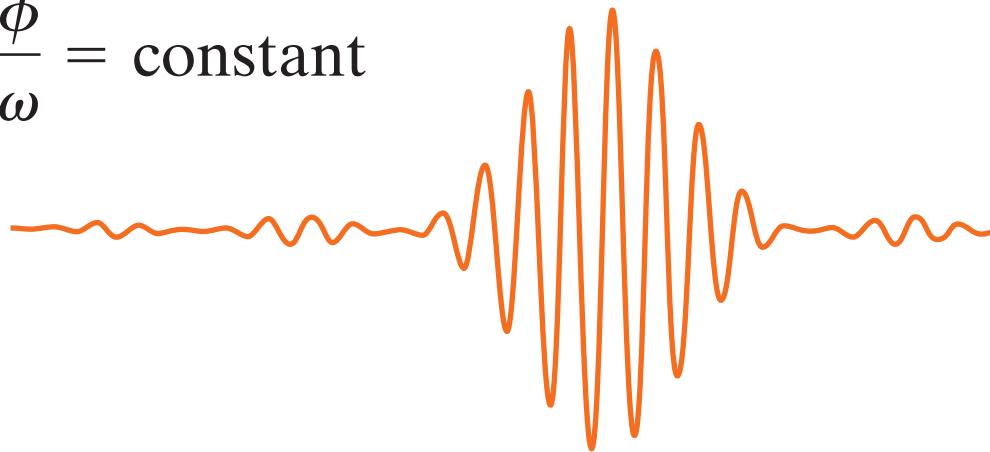
$$\frac{d\phi}{d\omega} = \text{constant}$$



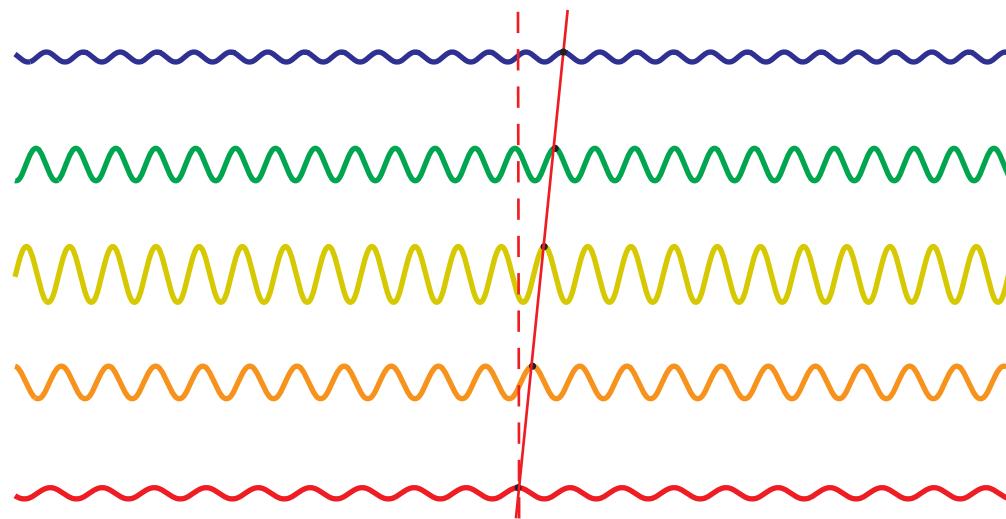
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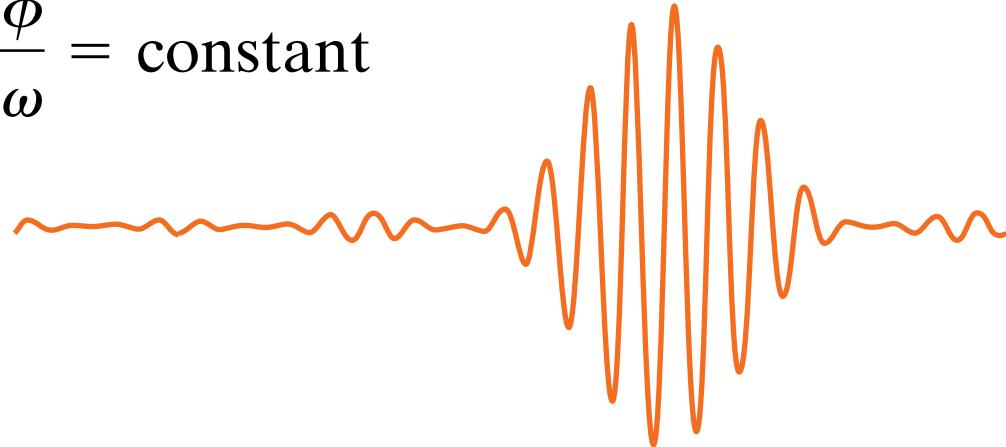
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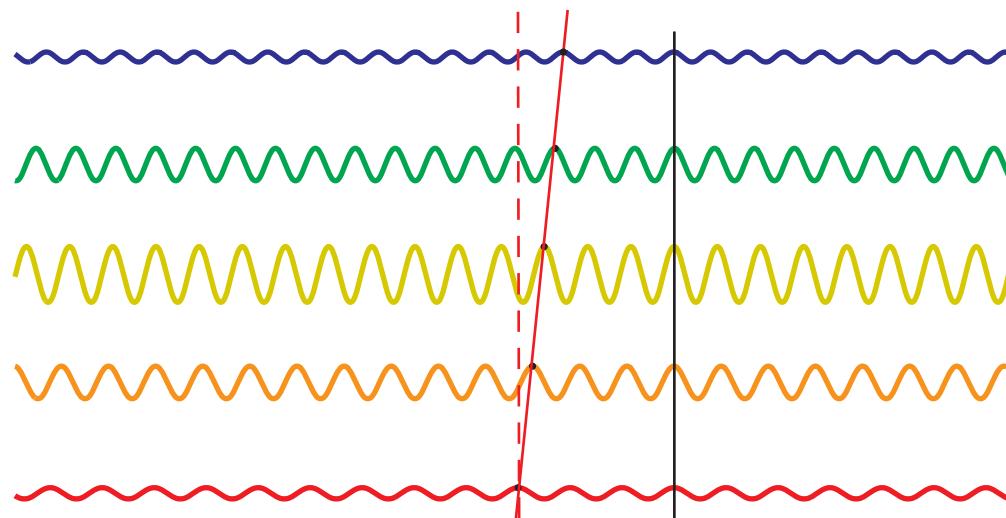
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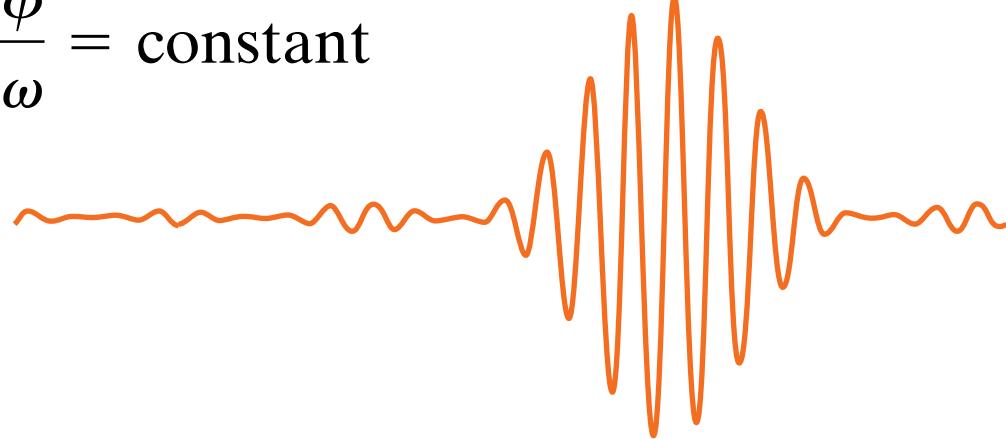
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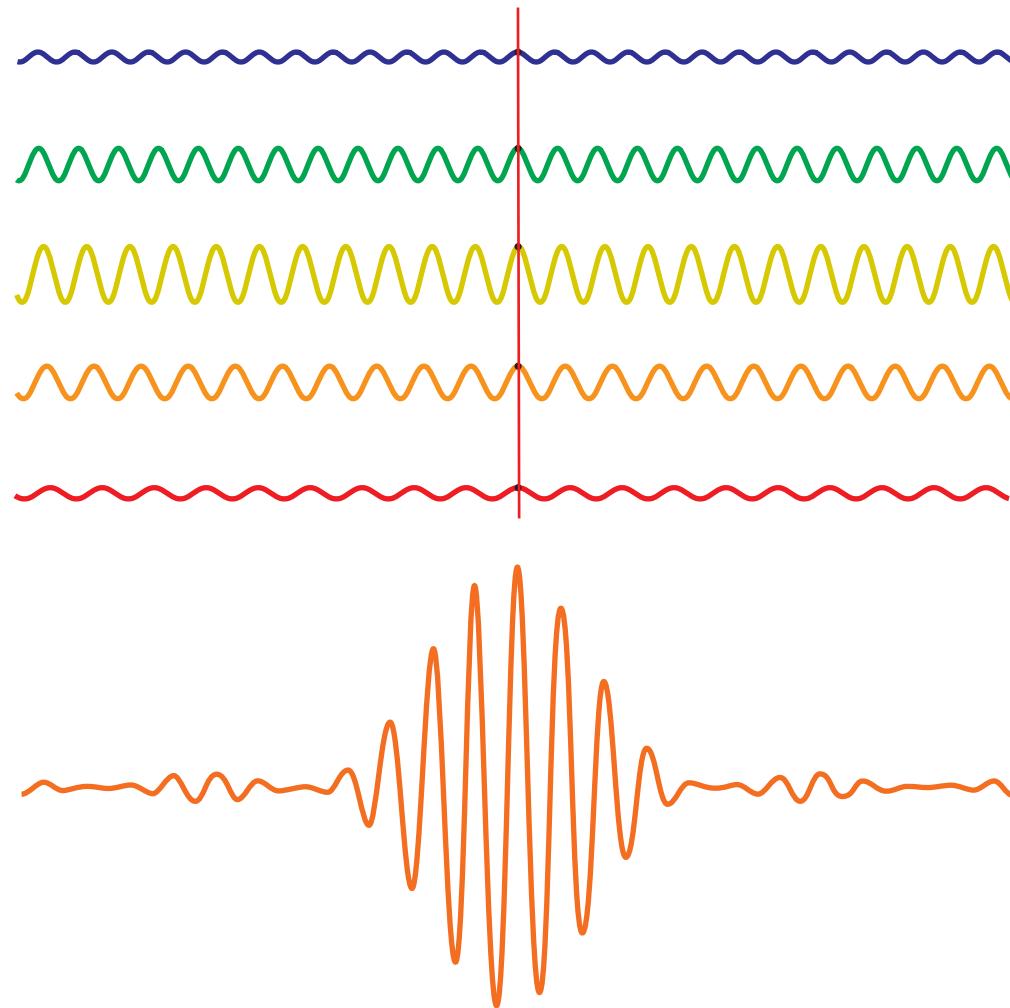
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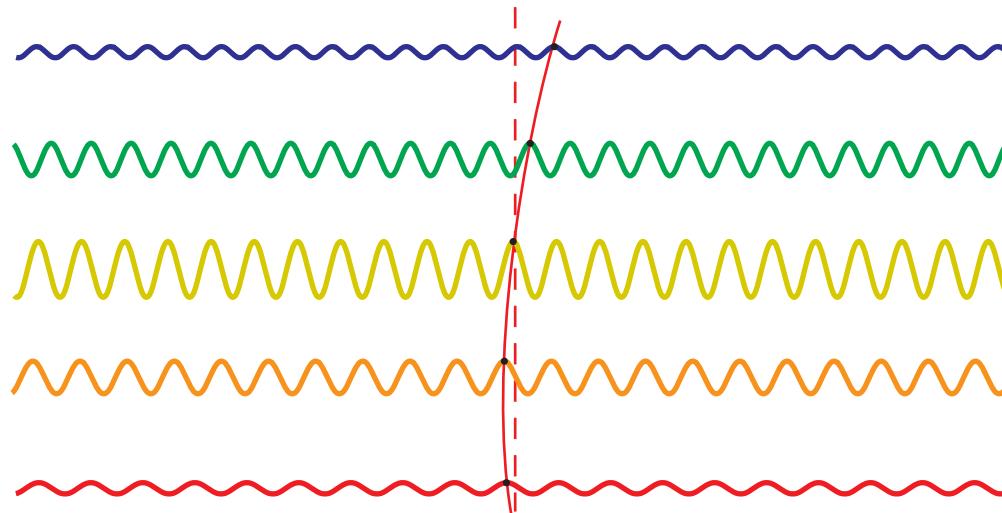
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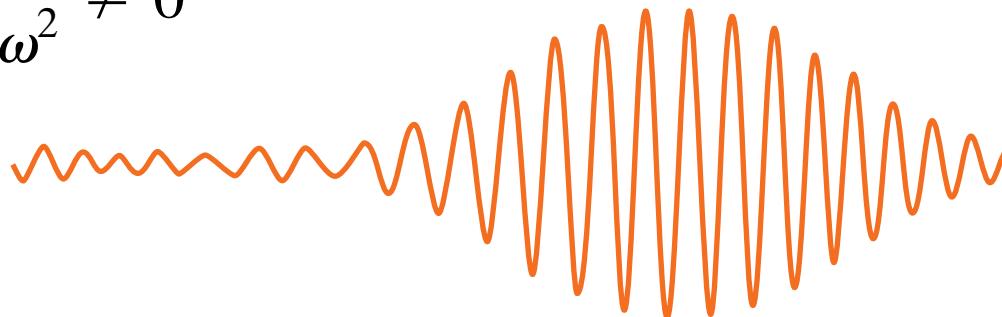
Dispersion compensation



Dispersion compensation



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

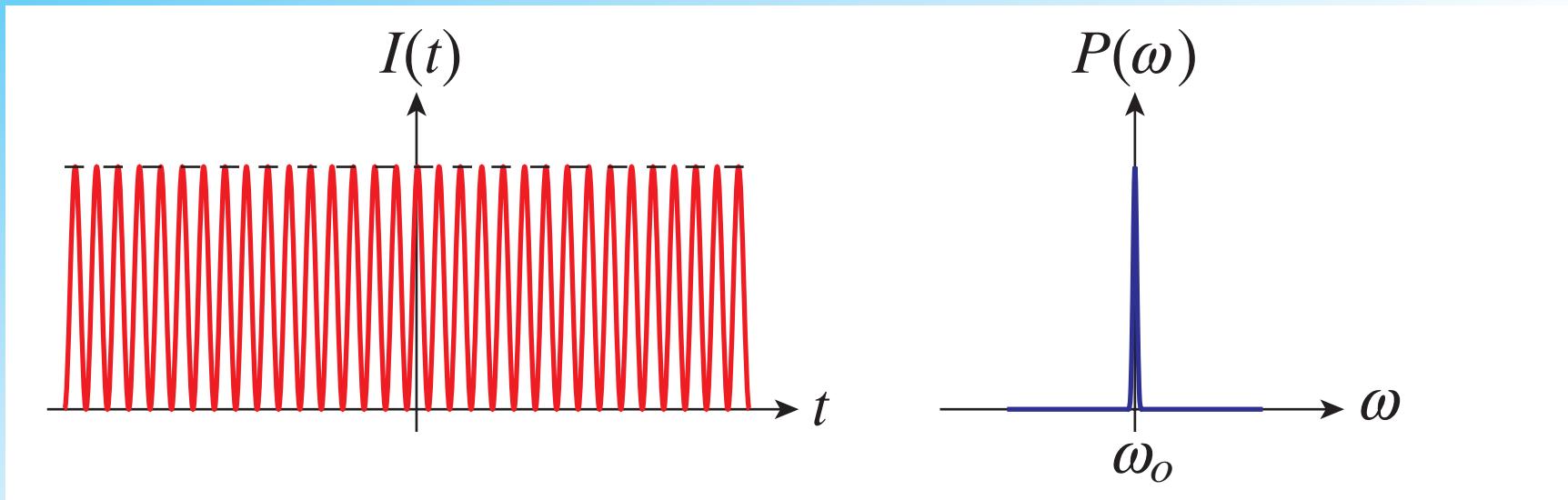


Dispersion compensation

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

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

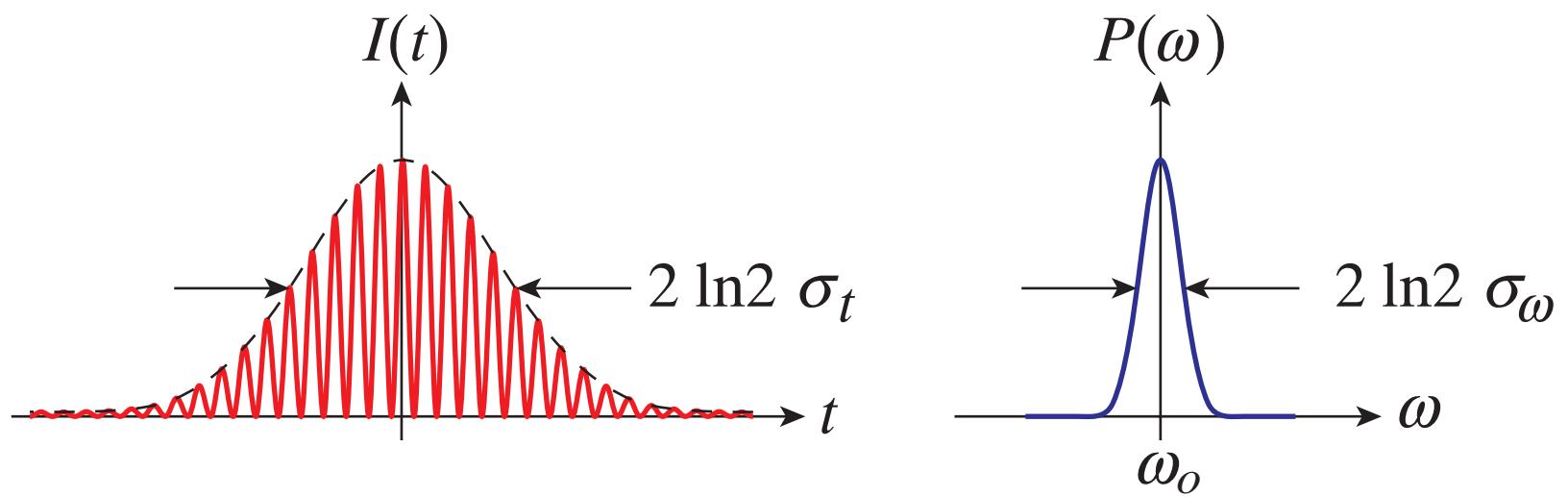
Representation of pulses



Spectrum of sinusoidal intensity is a delta function

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

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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Representation of pulses

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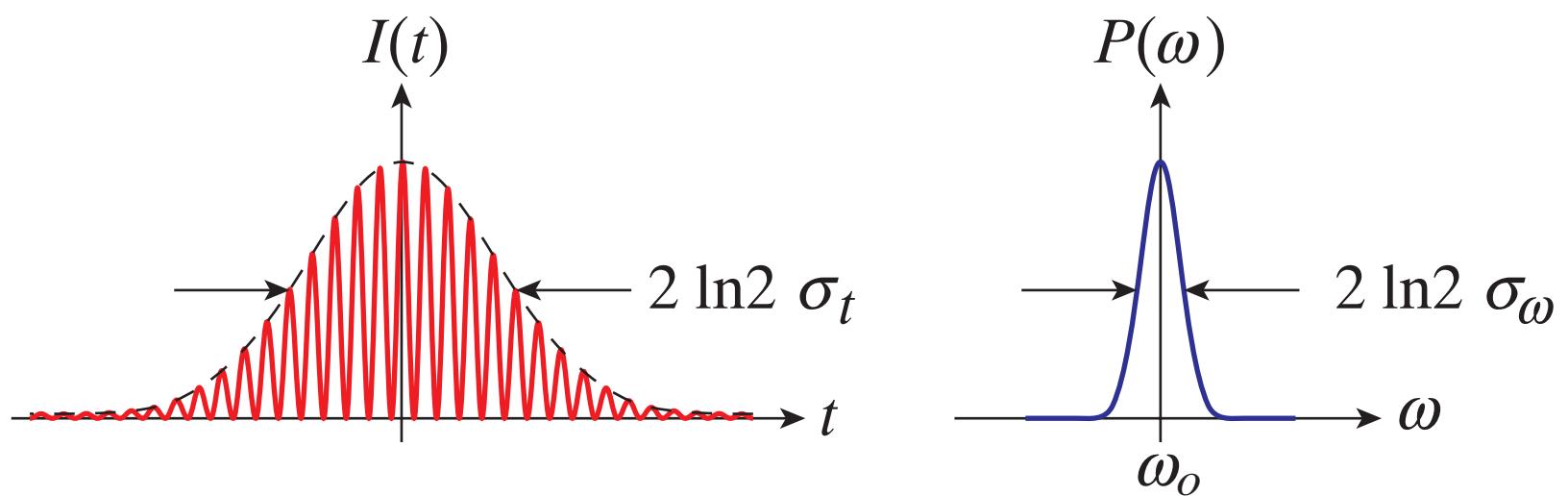
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$$= \sigma_t \exp\left[-\frac{\sigma_t^2(\omega - \omega_o)^2}{2}\right] \equiv \sigma_t \exp\left[-\frac{(\omega - \omega_o)^2}{2\sigma_\omega^2}\right]$$

Representation of pulses

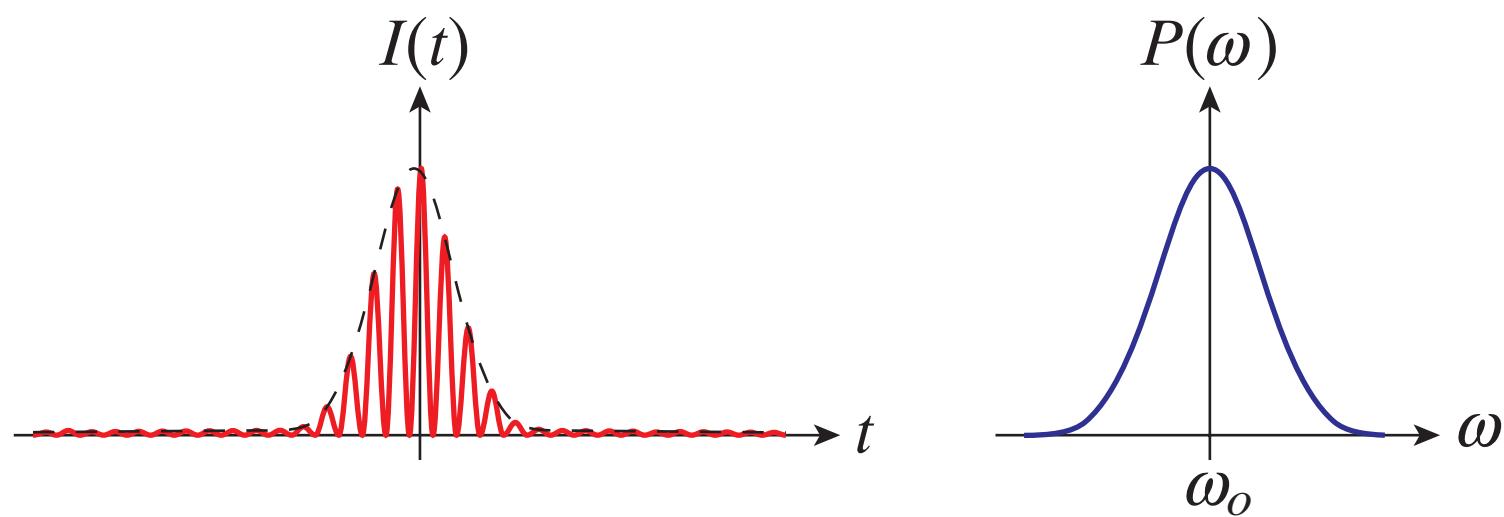


Pulse duration-bandwidth product: $\sigma_t \sigma_\omega = 1$

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

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

Representation of pulses



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Joint time-frequency representation

Wigner representation:

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

Joint time-frequency representation

Wigner representation:

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

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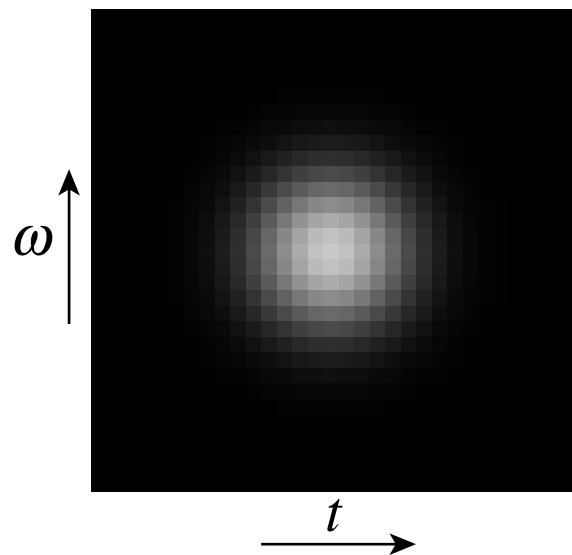
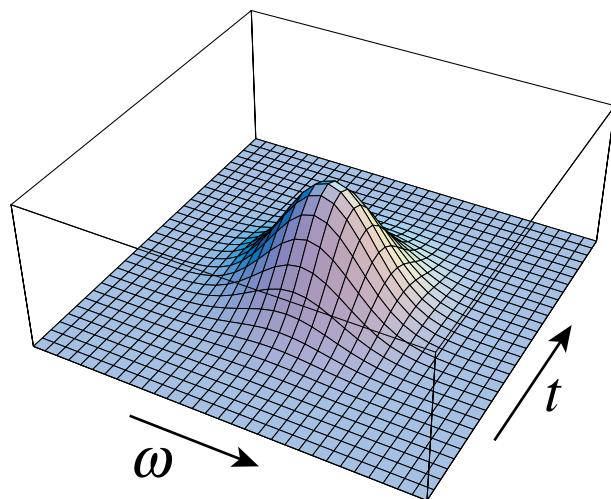
$$\frac{1}{2\pi} \int_{-\infty}^{\infty} W(t, \omega) d\omega = |E(t)|^2 = I(t)$$

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

Joint time-frequency representation

Energy:

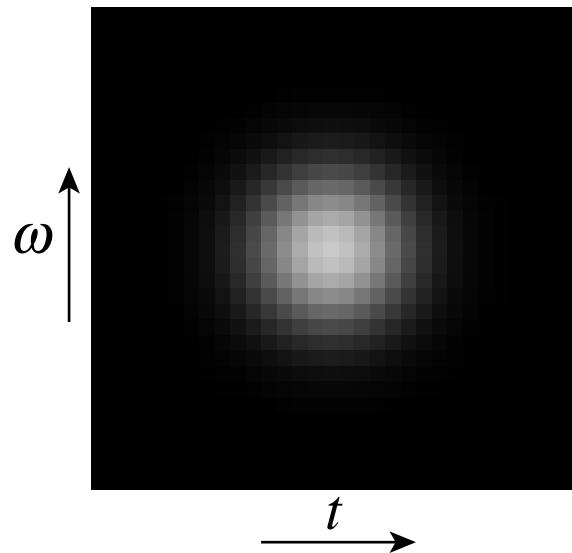
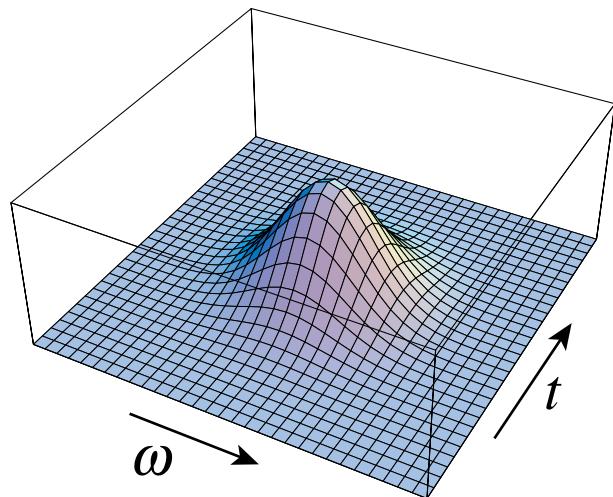
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Joint time-frequency representation

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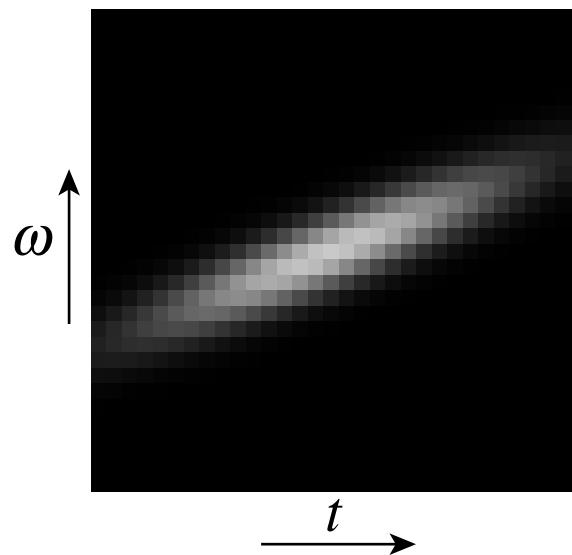
$W(t, \omega)$ must be nonzero in phase-space area larger than π

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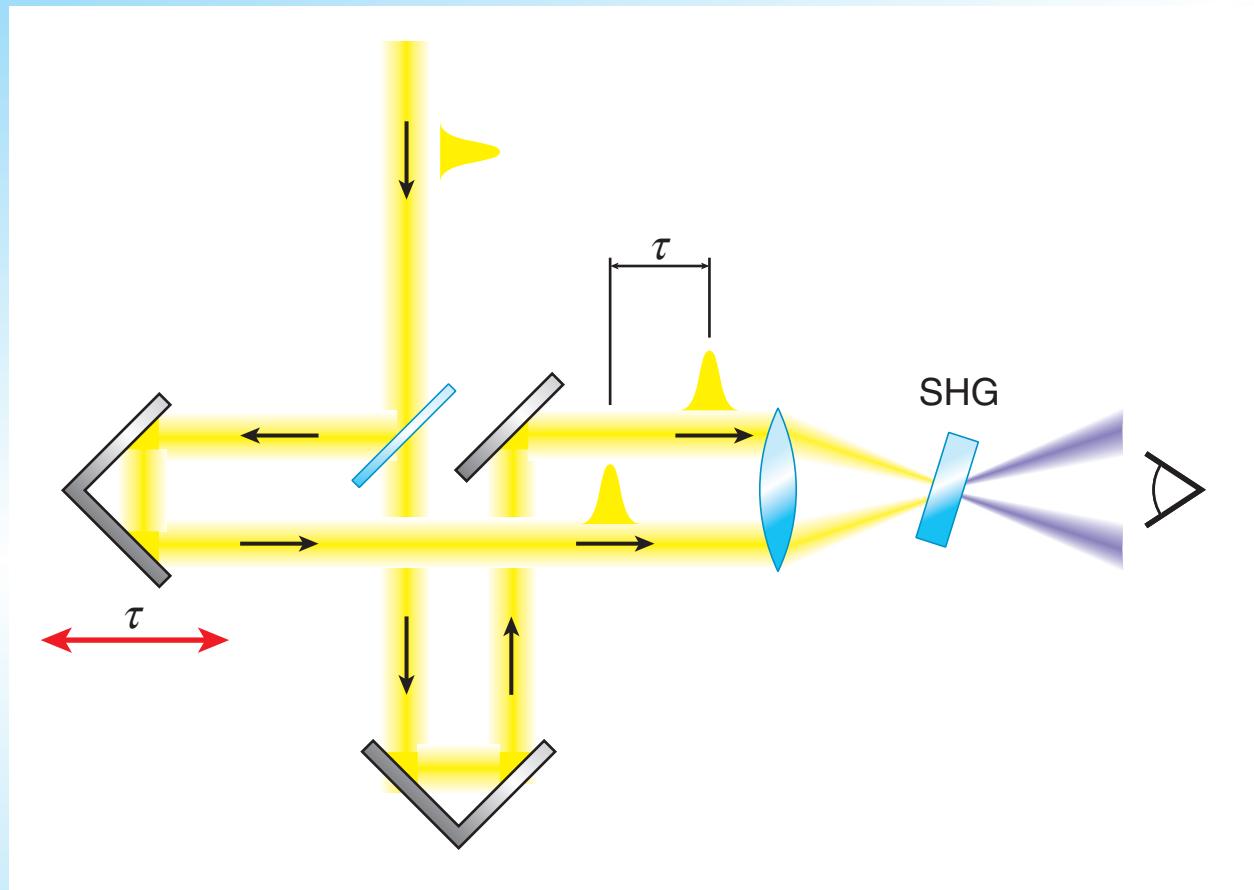
chirped pulse



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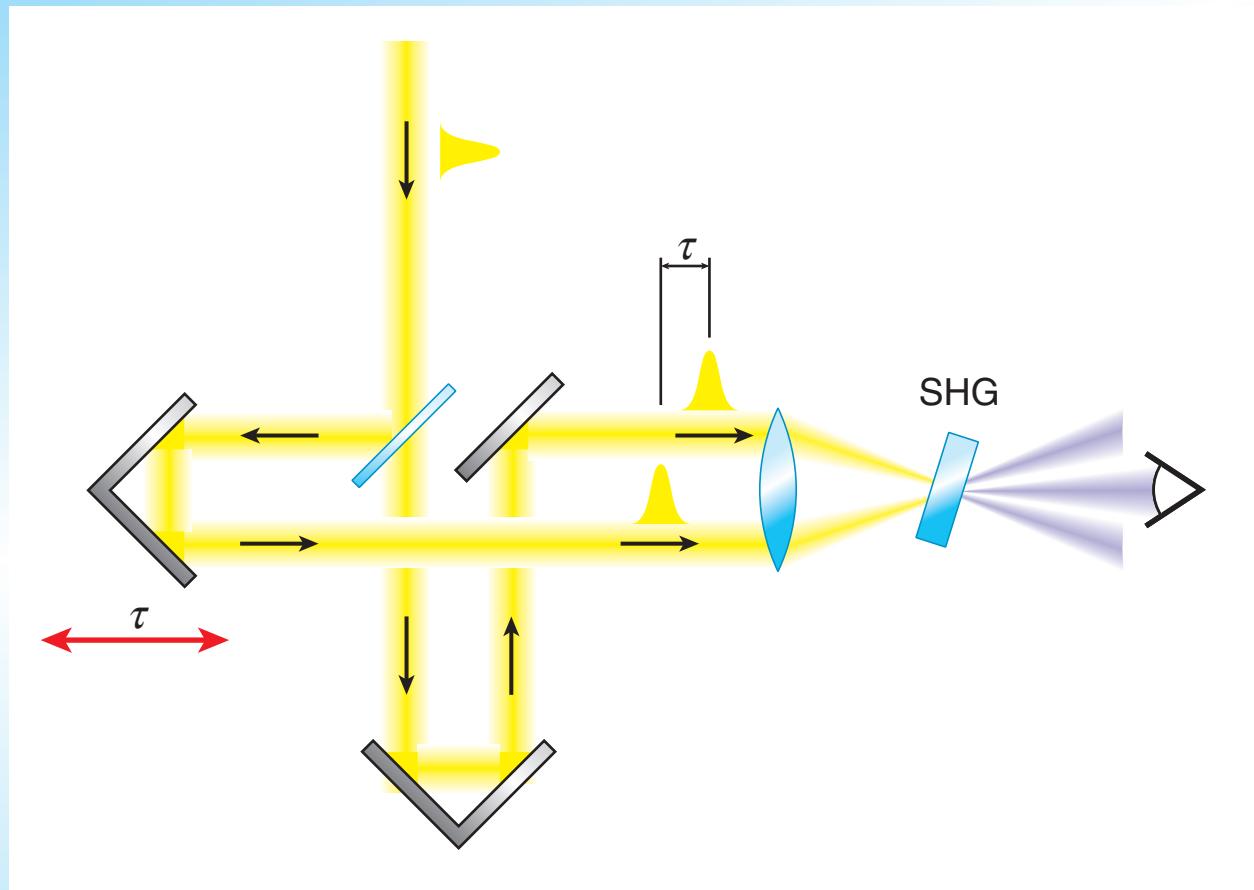
Temporal characterization

Use pulse to measure itself...



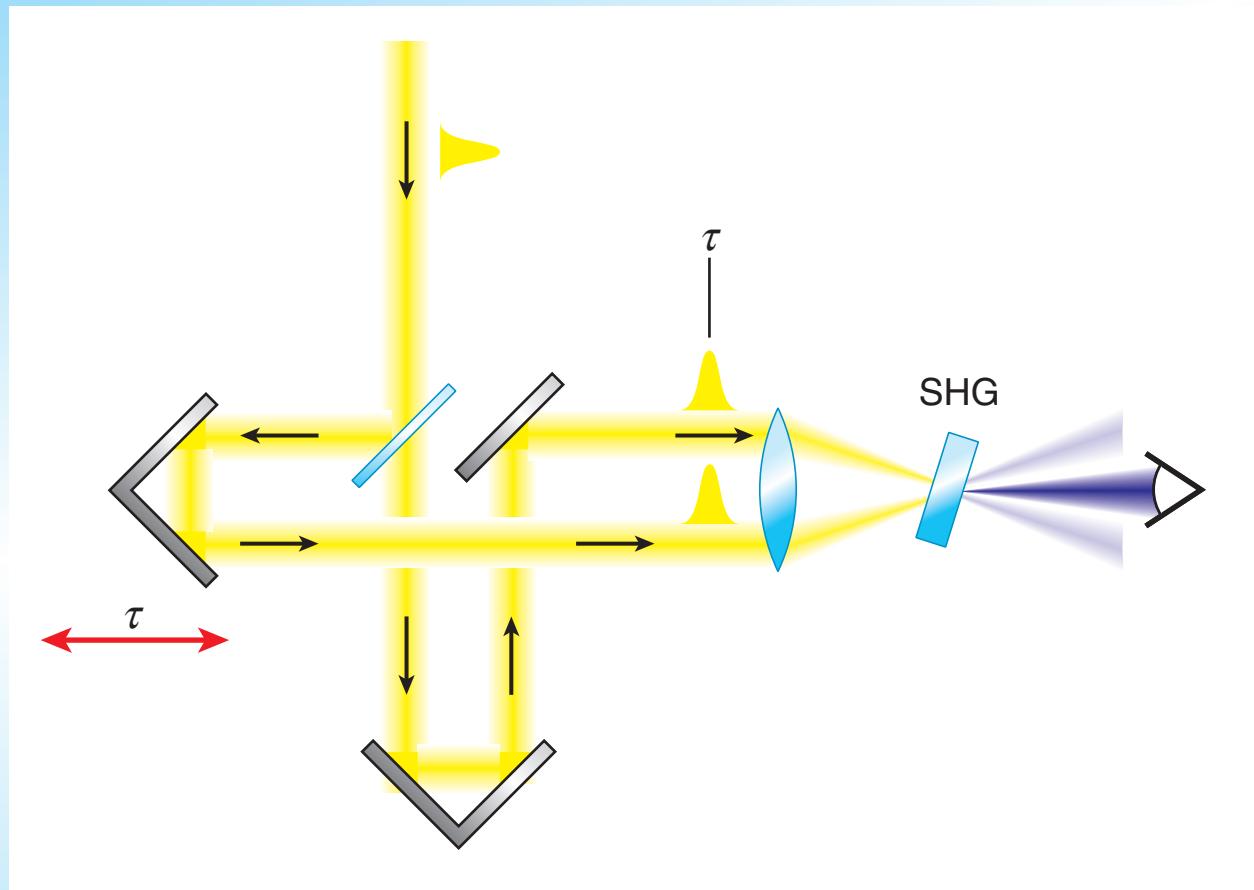
Temporal characterization

Use pulse to measure itself...



Temporal characterization

Use pulse to measure itself...



Temporal characterization

Electric field at SHG crystal

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

Temporal characterization

Electric field at SHG crystal

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Second harmonic field

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

Temporal characterization

Electric field at SHG crystal

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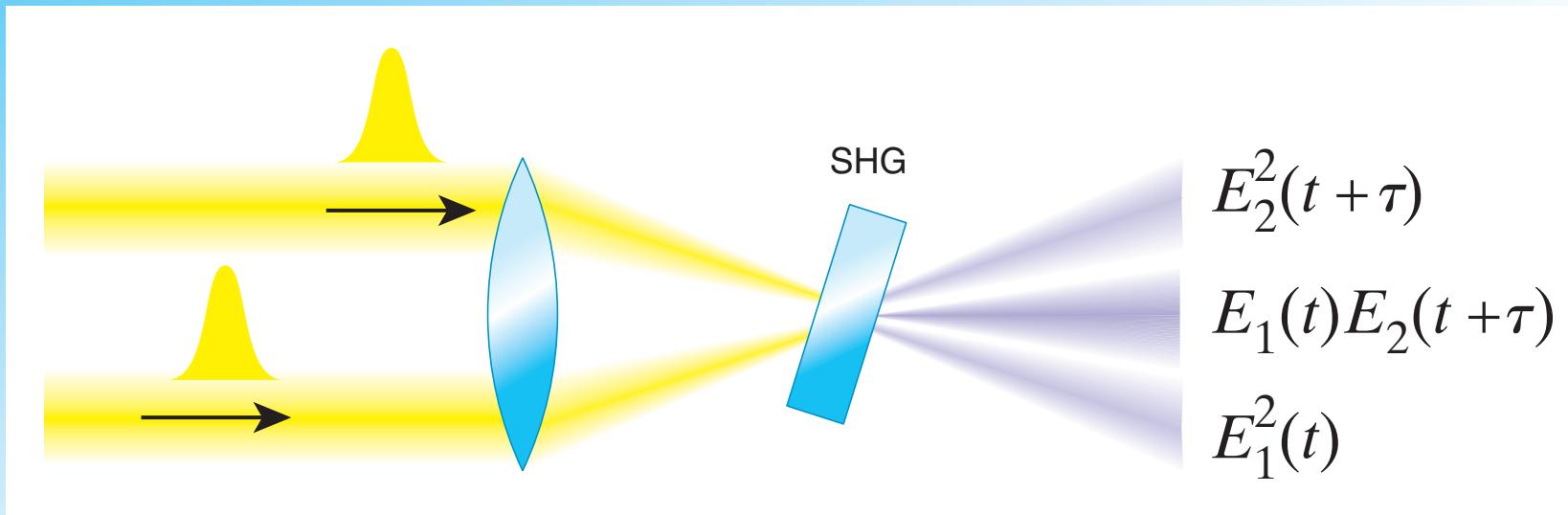
Second harmonic field

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

Second harmonic intensity

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

Temporal characterization



Second harmonic intensity

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detector selects middle term

Temporal characterization

Integrated detector signal yields intensity autocorrelation

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

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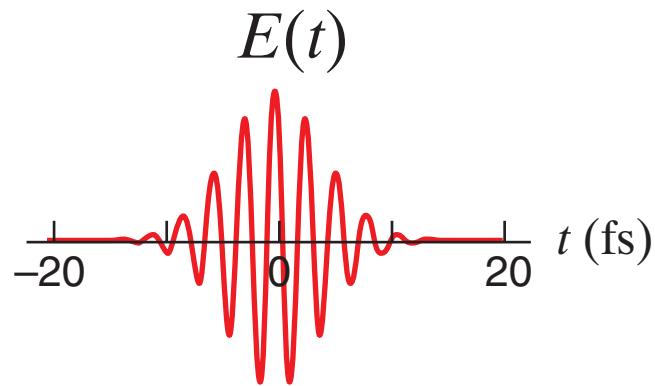
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Temporal characterization

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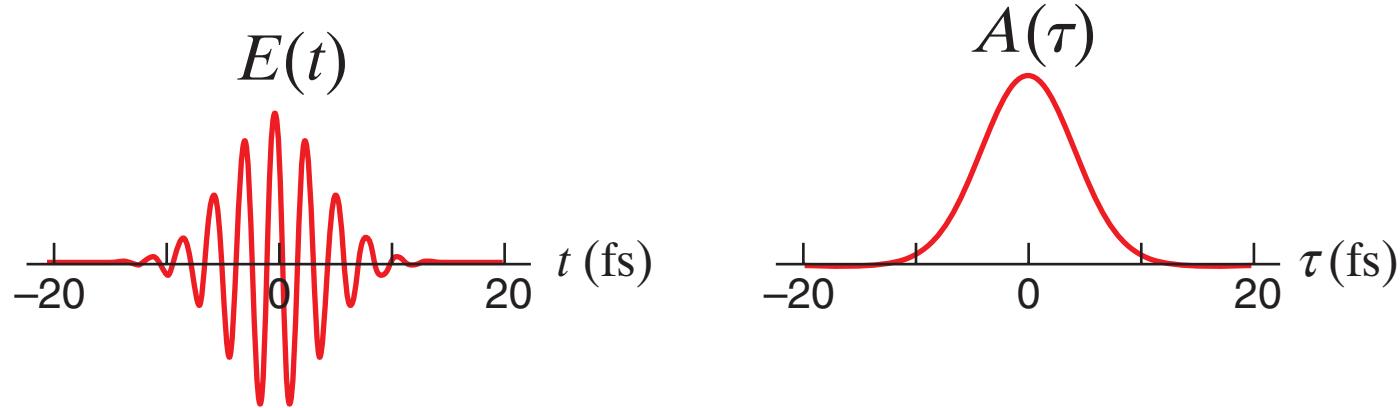


Temporal characterization

Integrated detector signal yields intensity autocorrelation

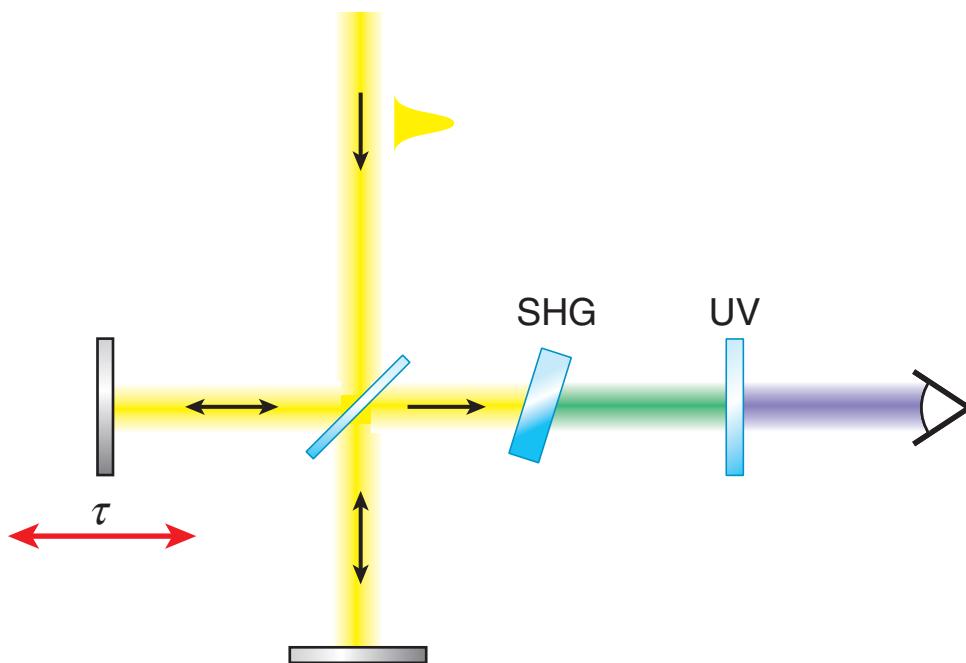
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Temporal characterization

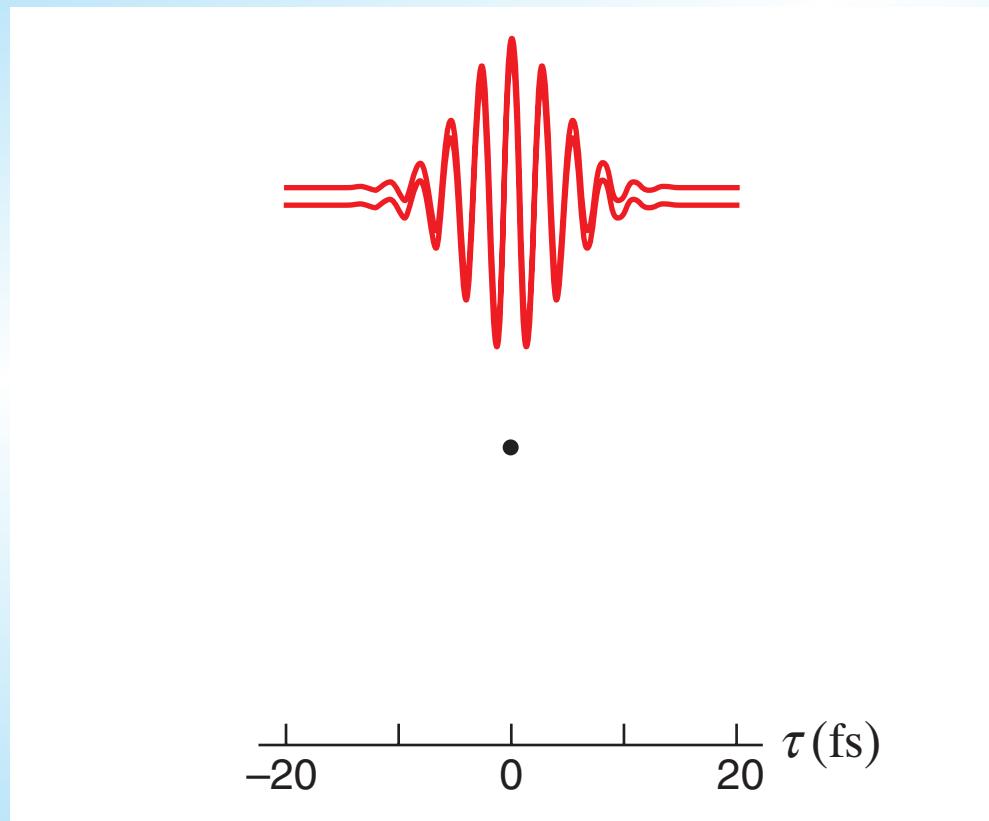
Alternative colinear geometry



Temporal characterization

All terms now contribute:

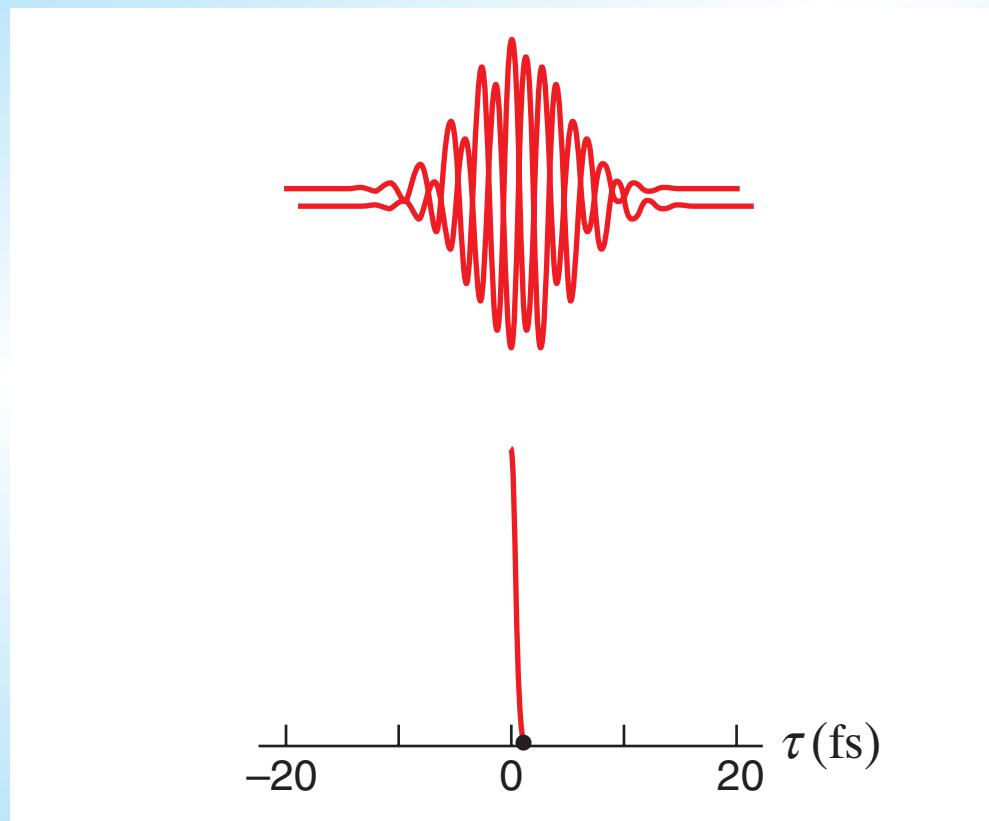
$$I_{2\omega}(t, \tau) \propto |\chi^{(2)}|^2 |E_{tot}^2|^2 = |\chi^{(2)}|^2 |E_1^2(t) + 2E_1(t)E_2(t+\tau) + E_2^2(t+\tau)|^2$$



Temporal characterization

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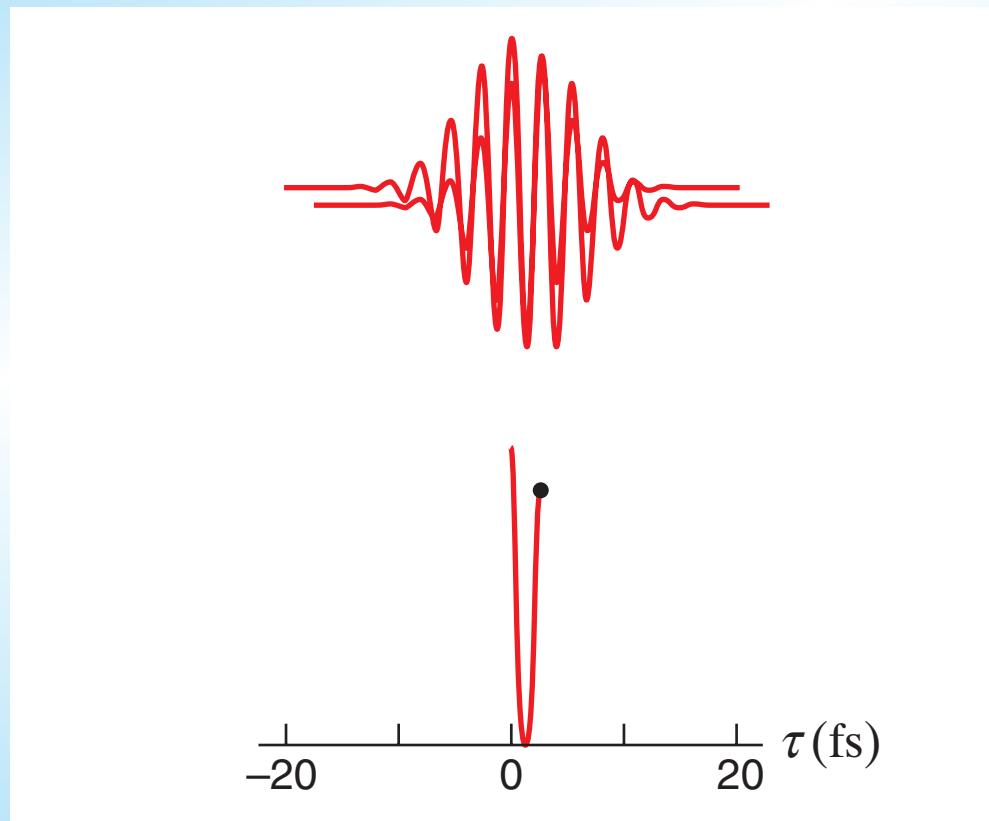
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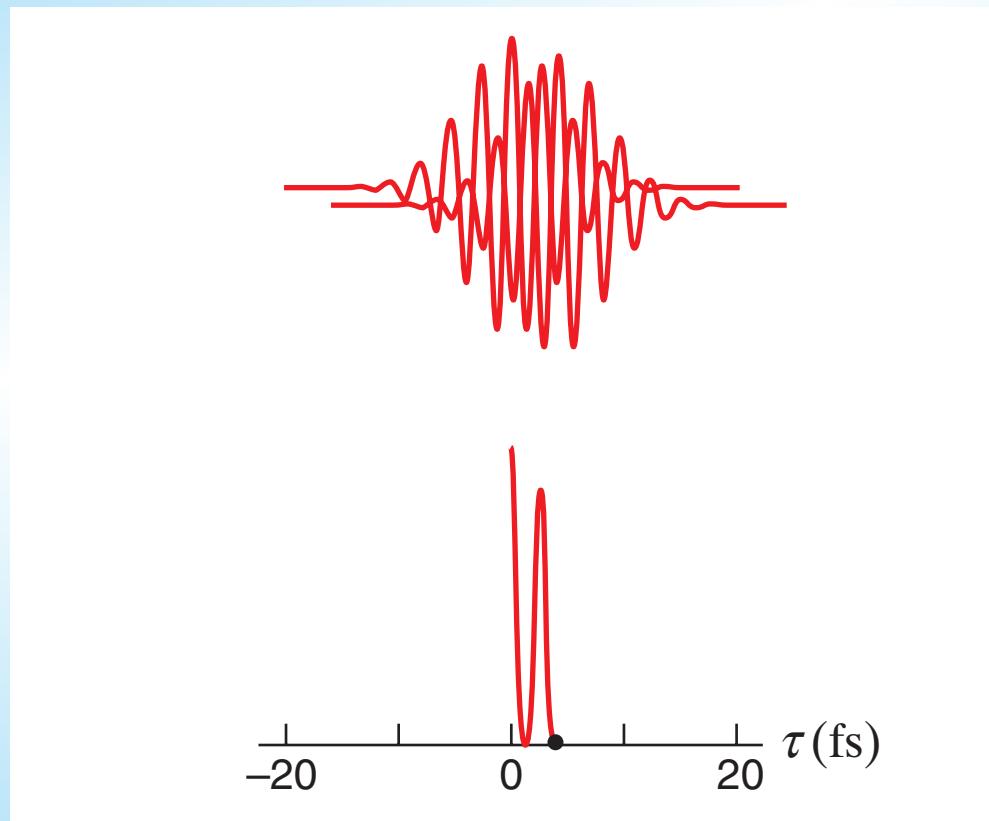
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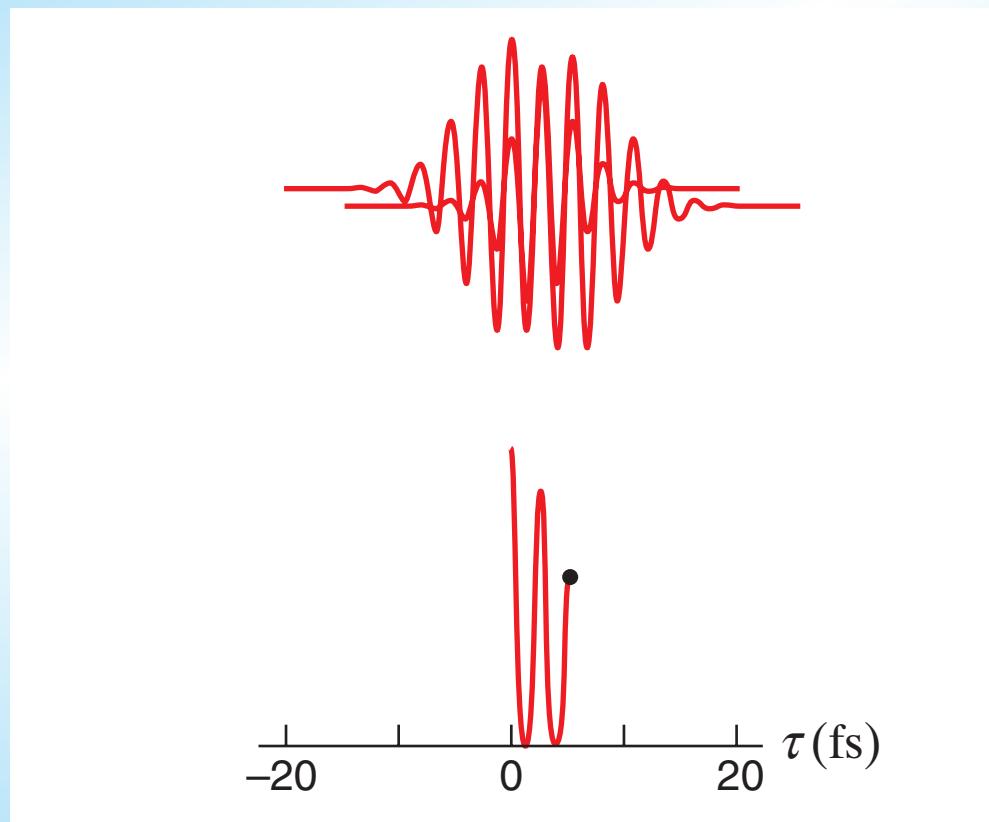
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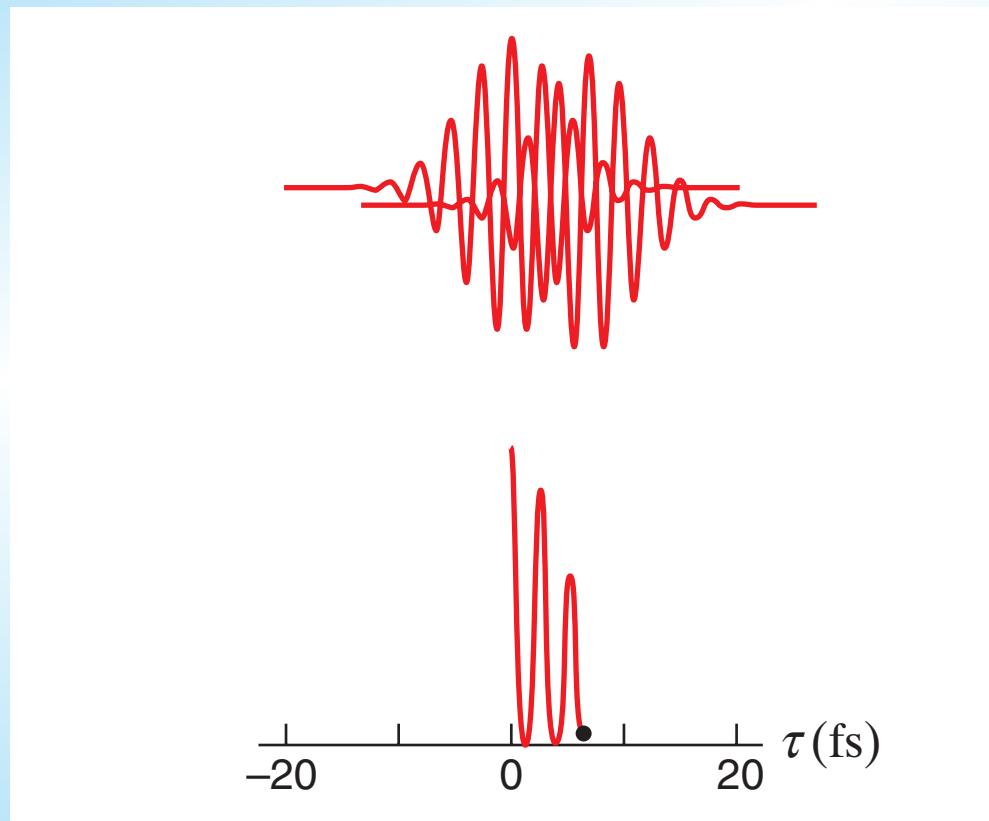
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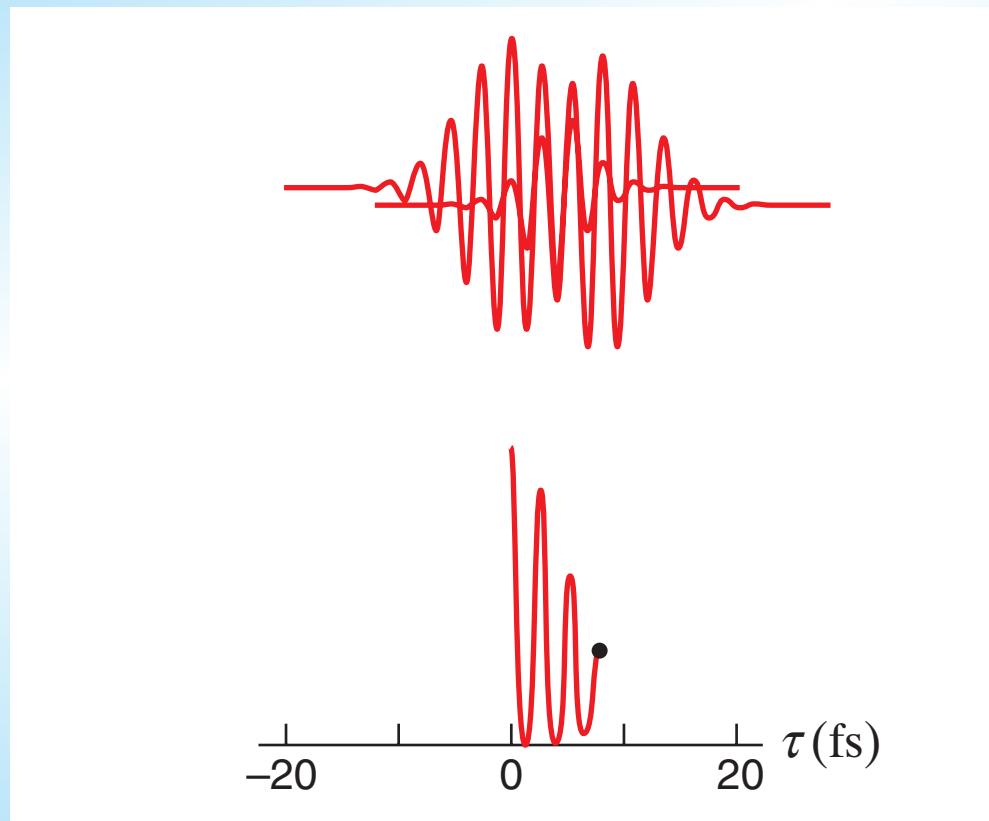
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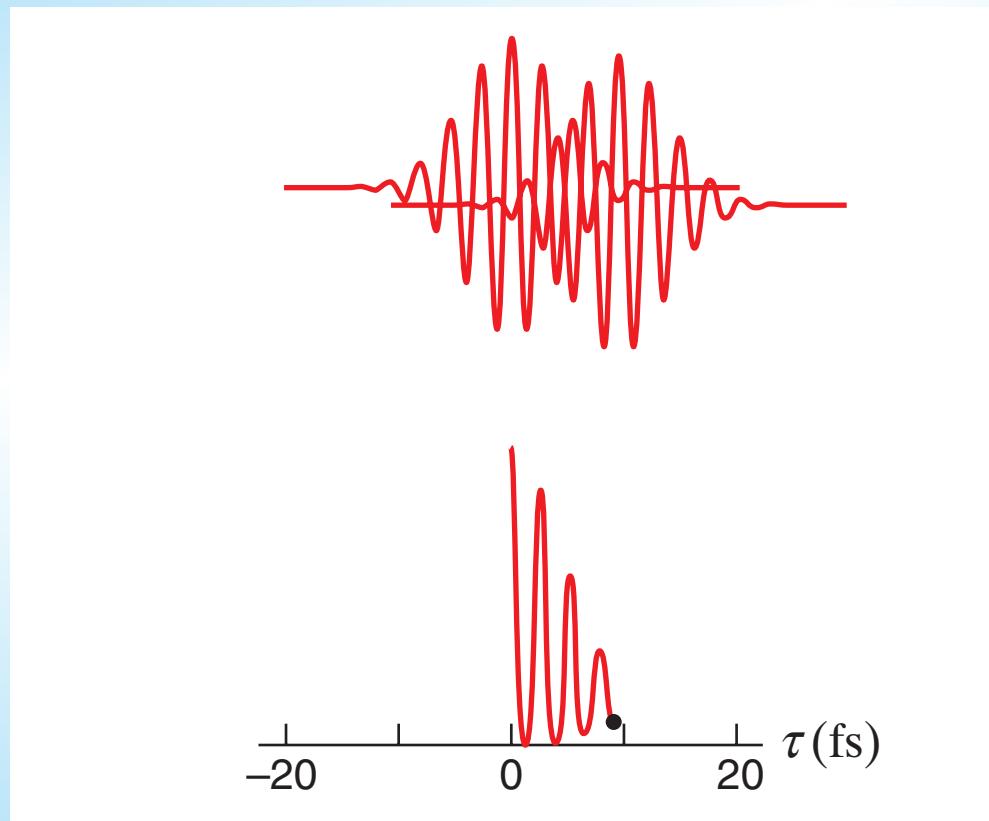
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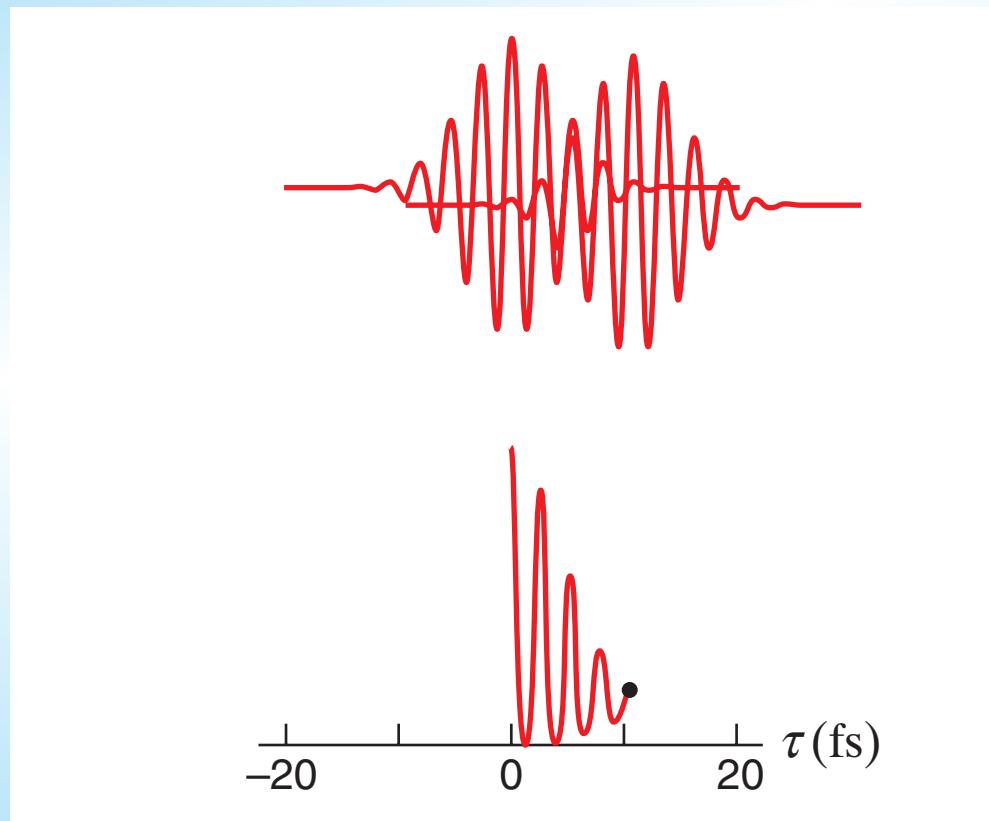
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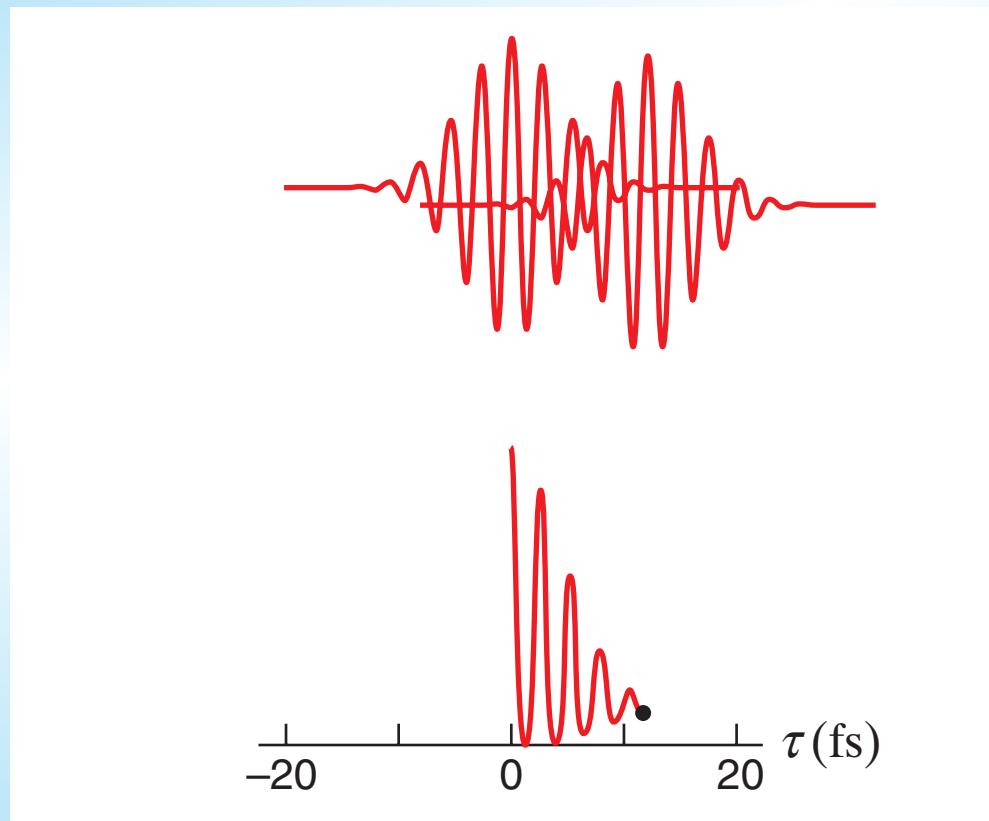
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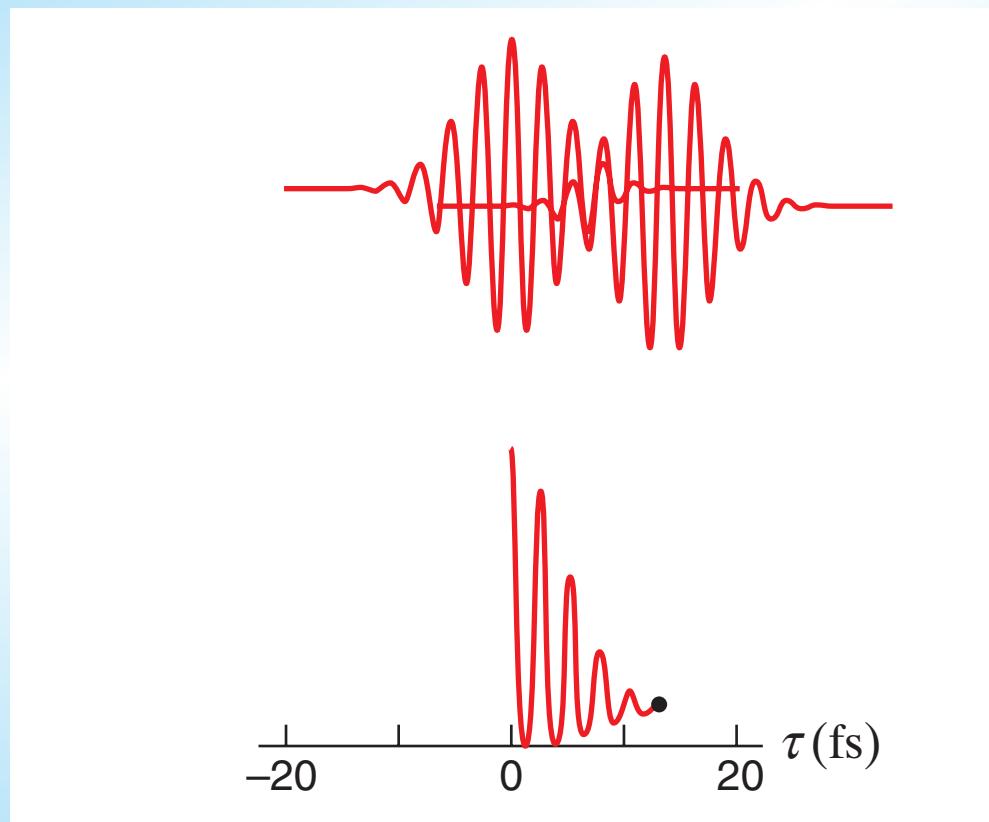
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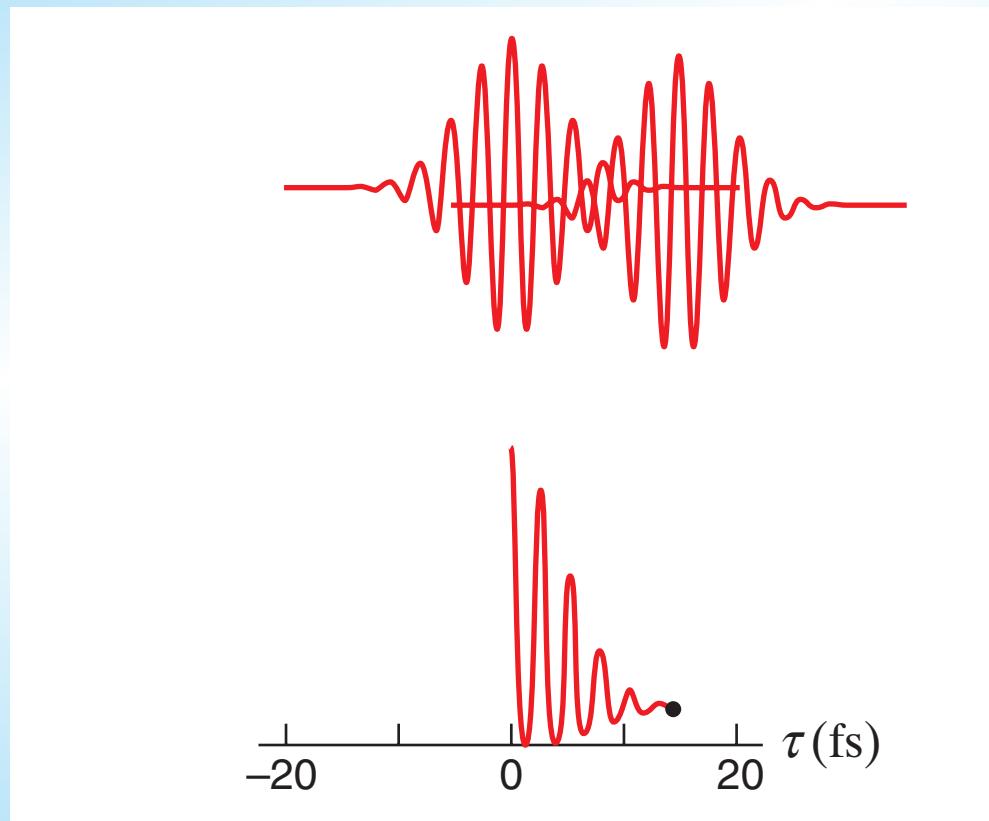
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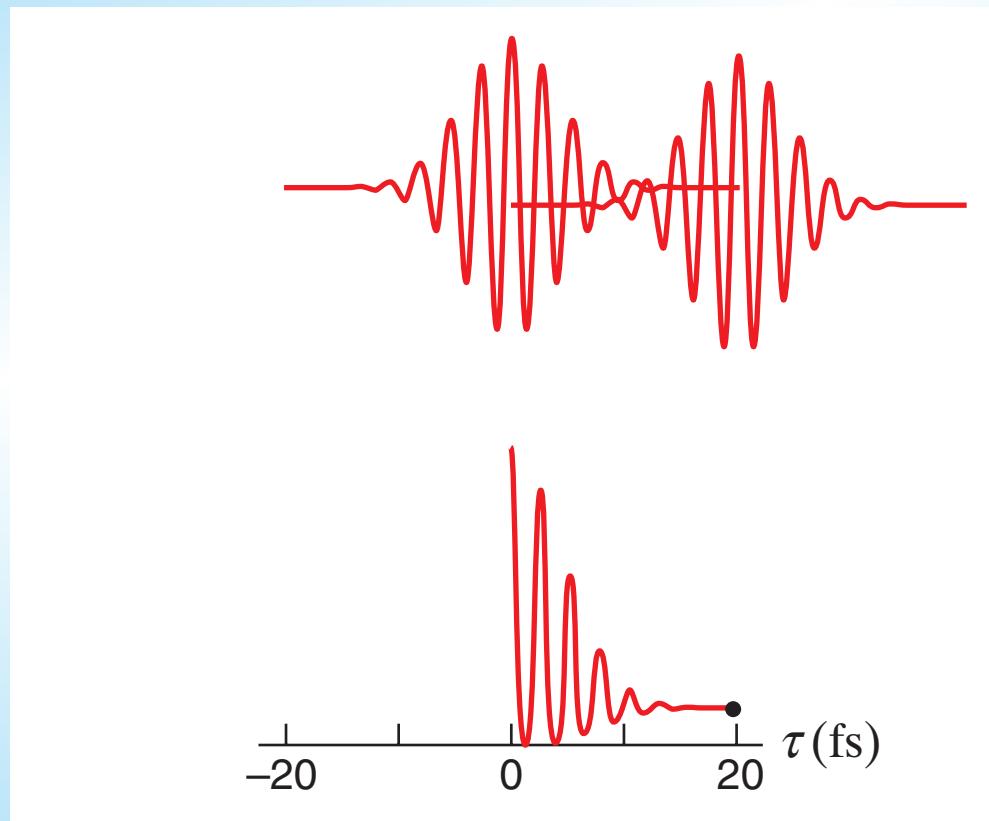
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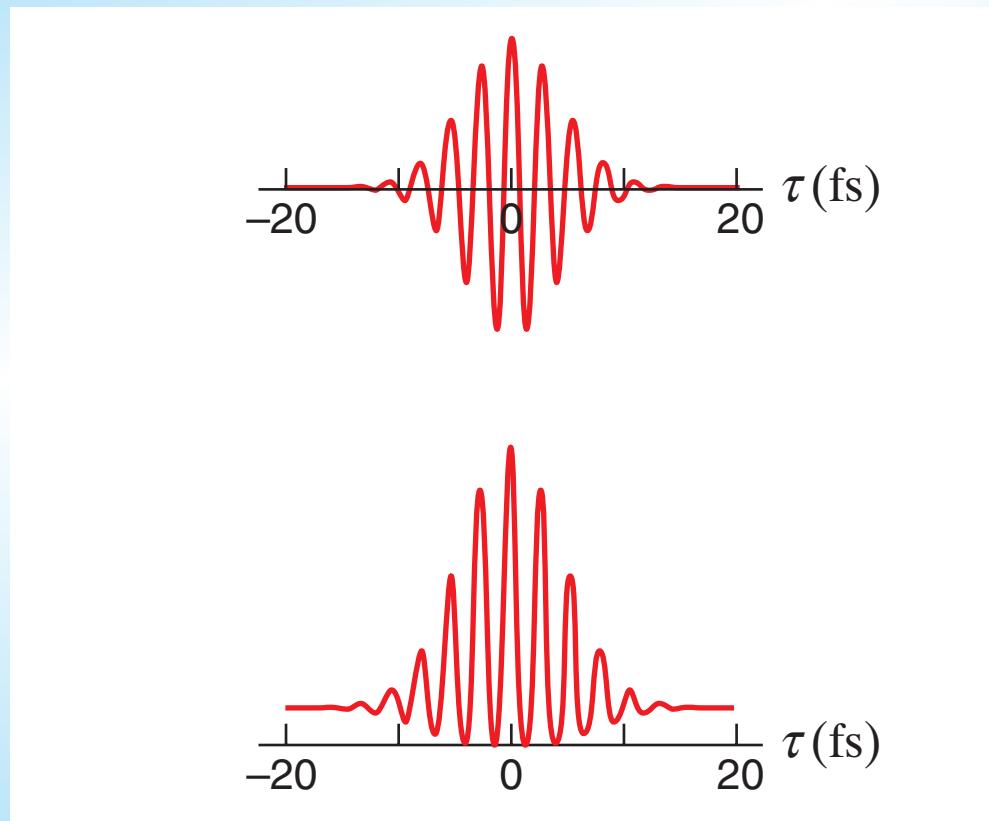
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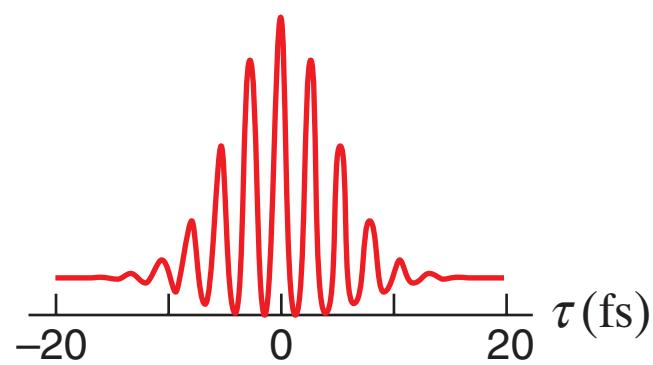
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at $\tau = 0$:

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



Temporal characterization

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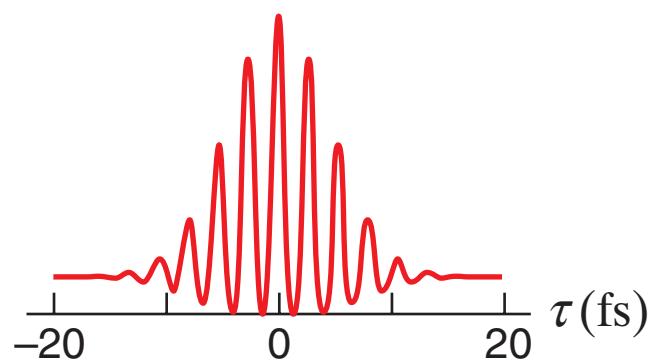
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as $\tau \rightarrow \pm\infty$:

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Temporal characterization

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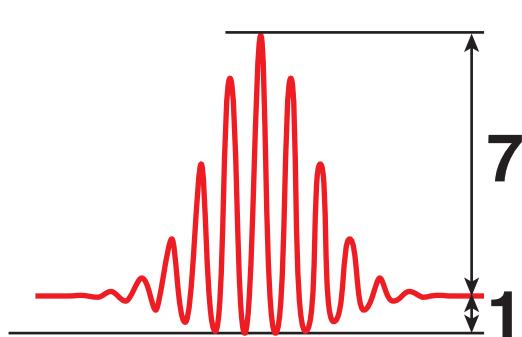
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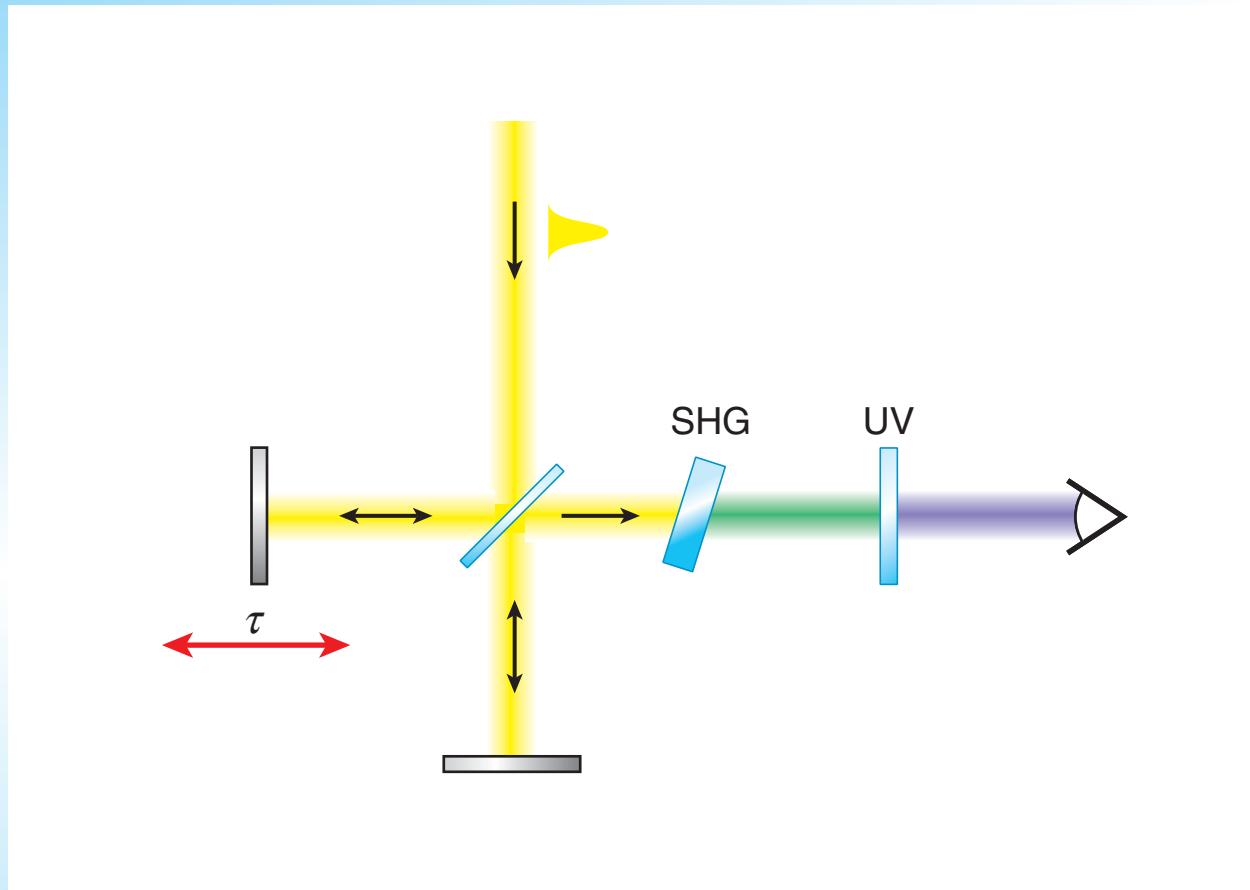
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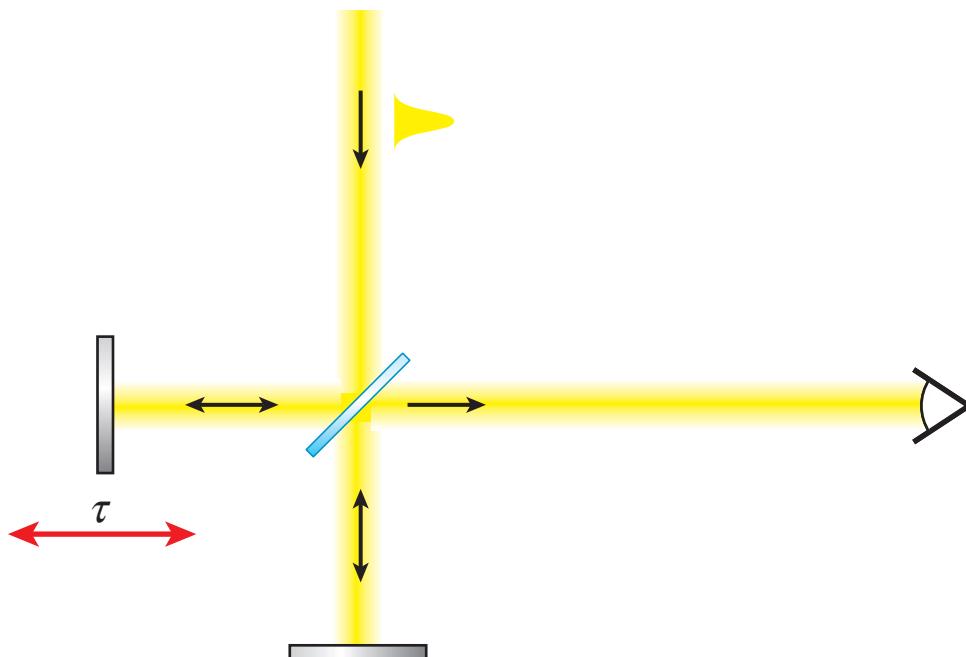
Temporal characterization

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



Temporal characterization

Would this work?



Temporal characterization

Intensity at detector

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

Temporal characterization

Intensity at detector

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

Detected signal

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

Temporal characterization

Intensity at detector

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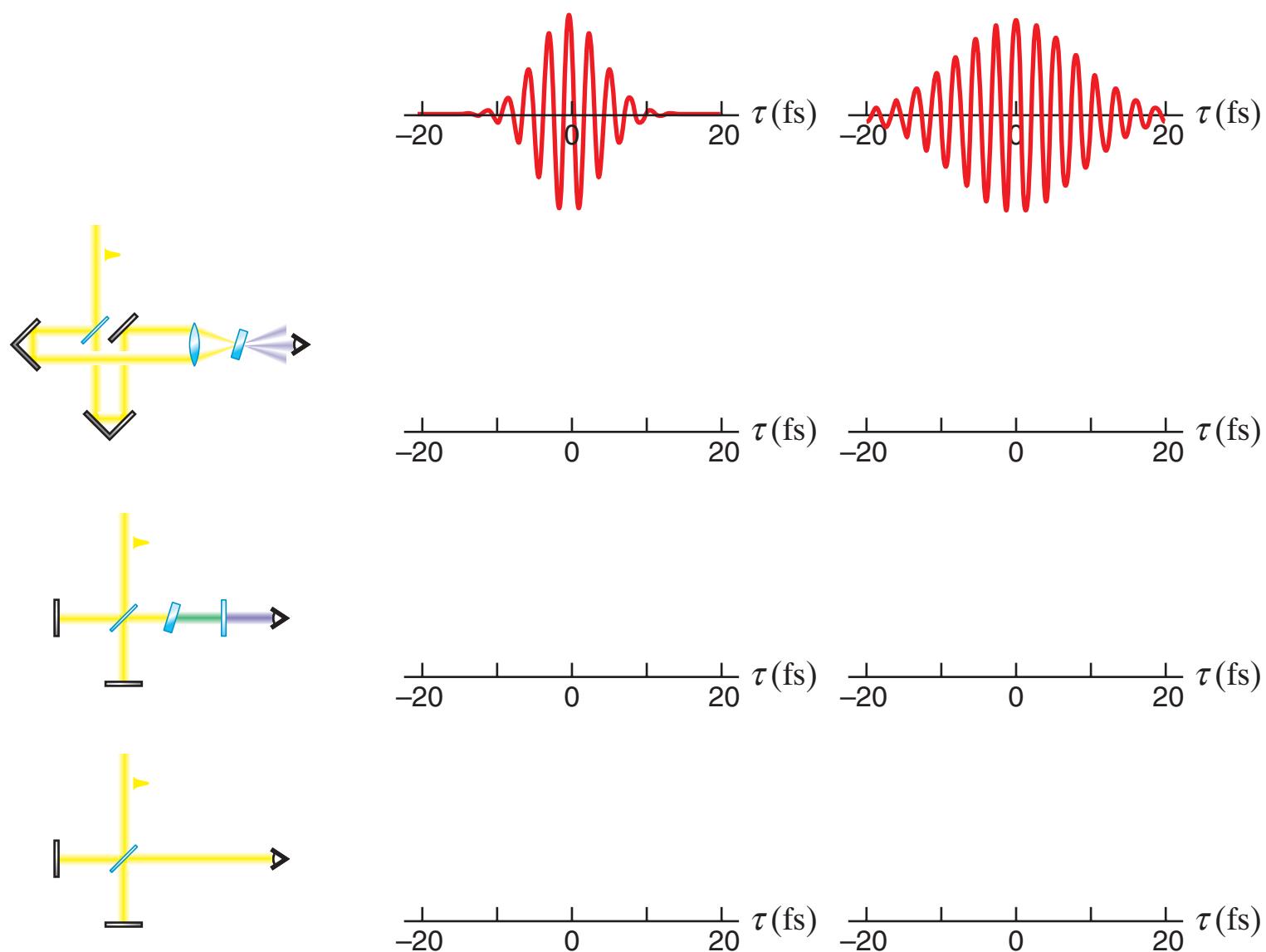
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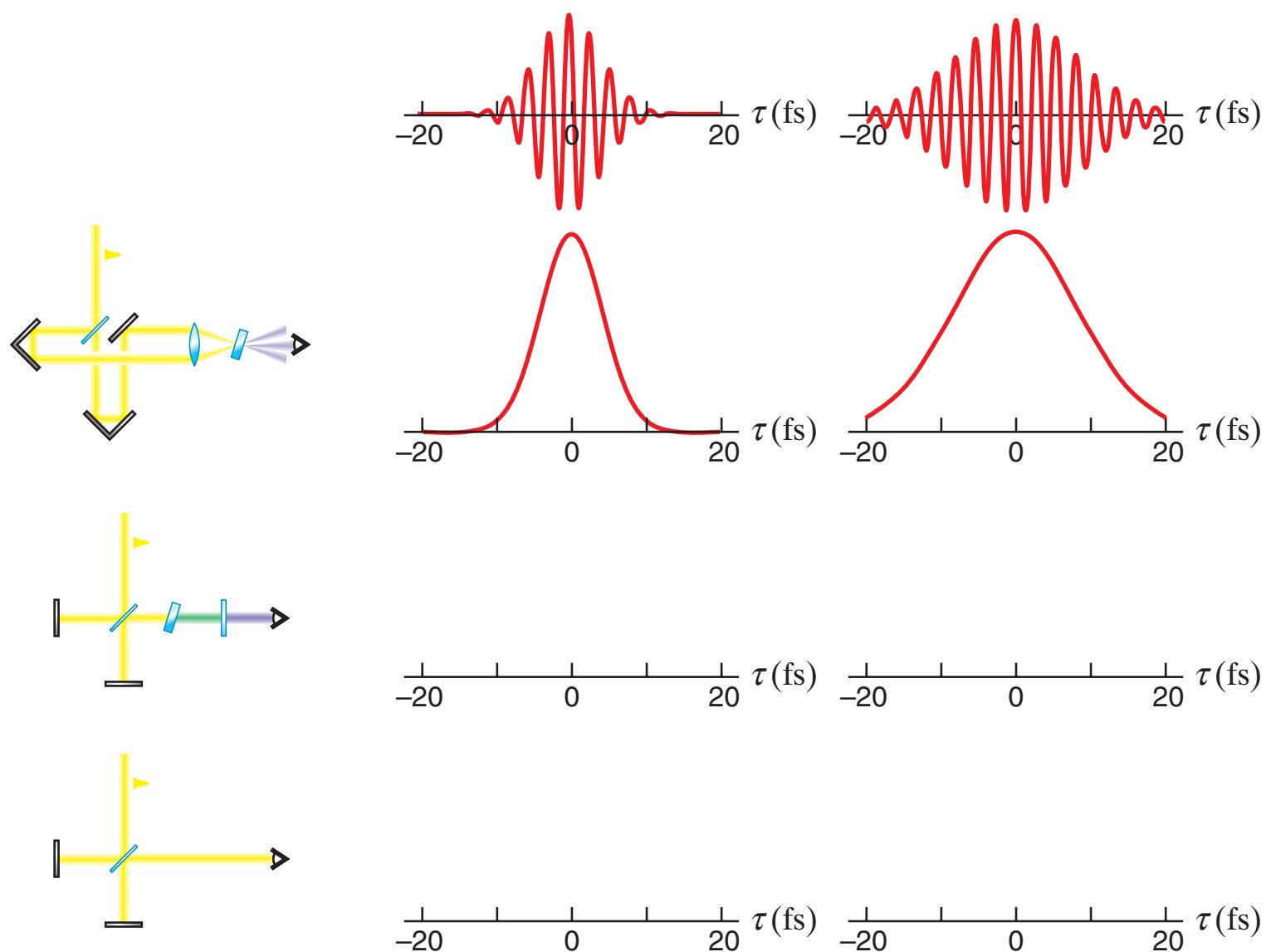
so

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

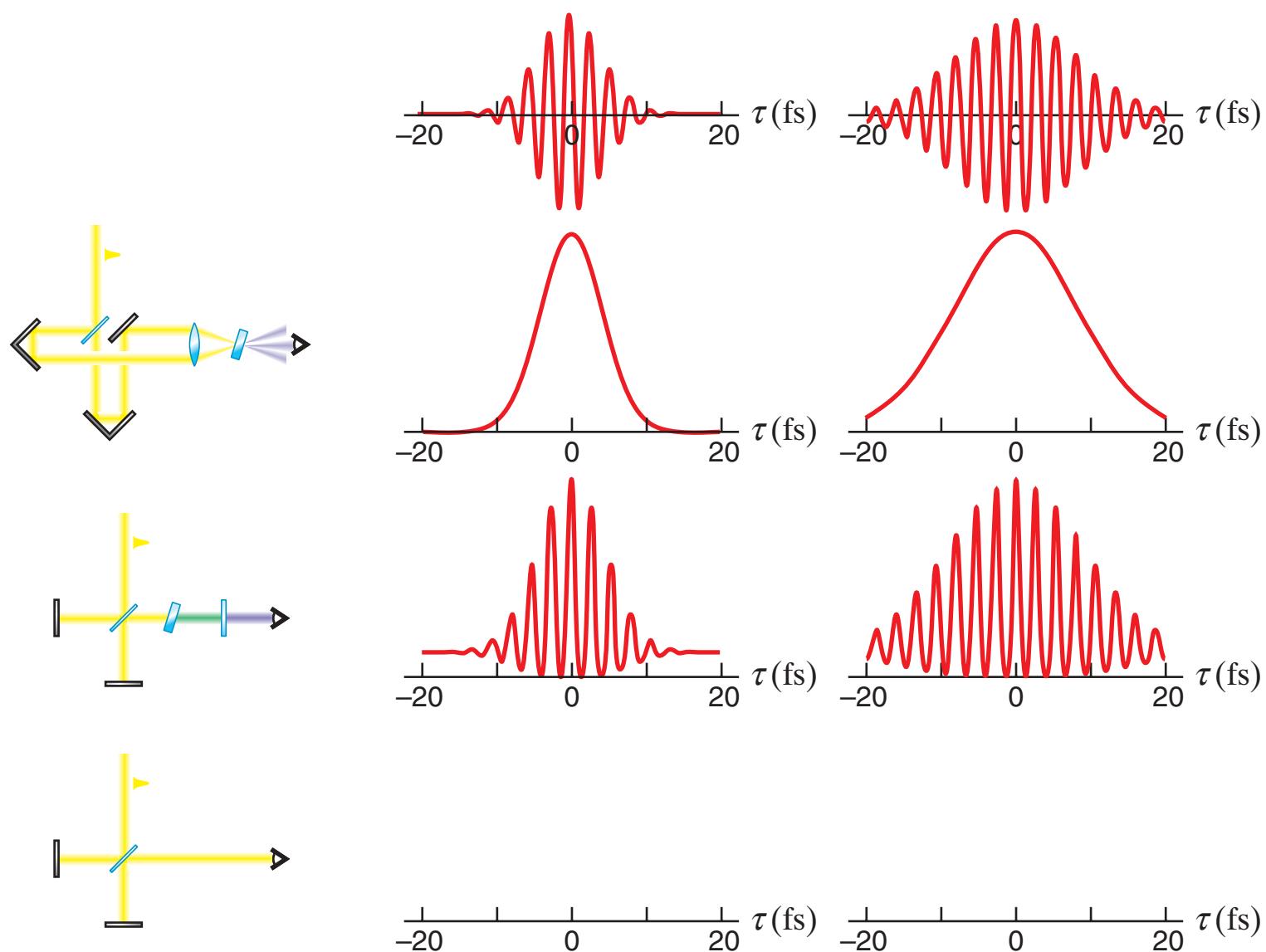
Temporal characterization



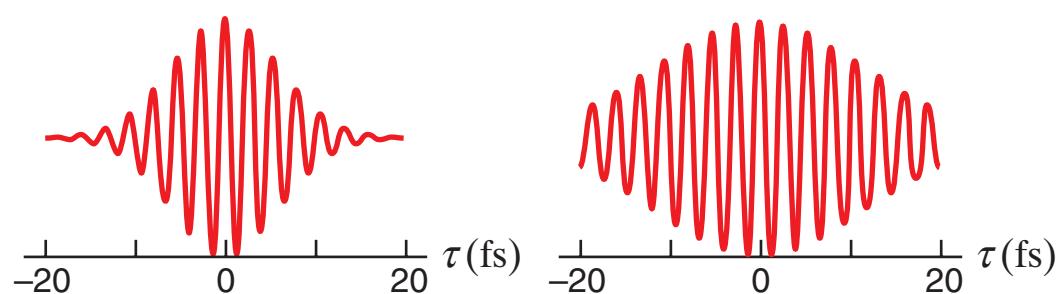
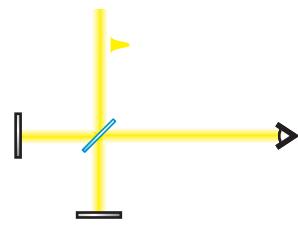
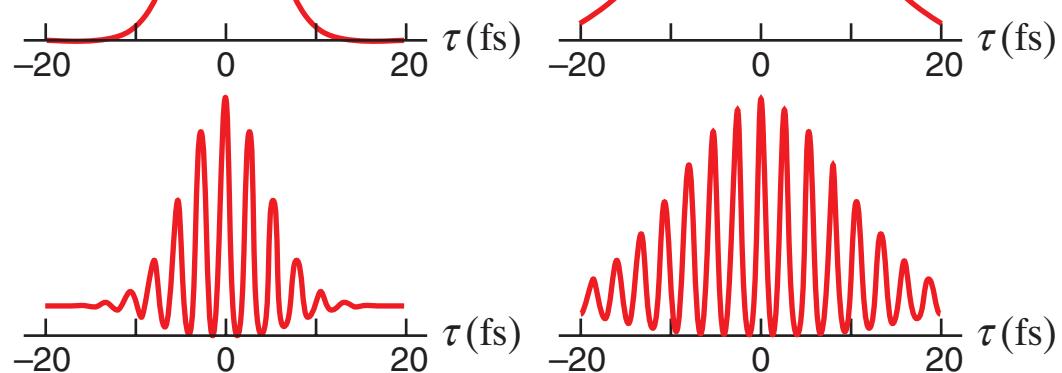
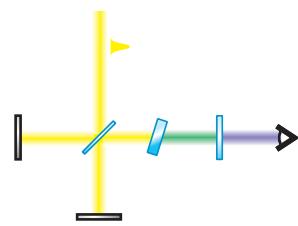
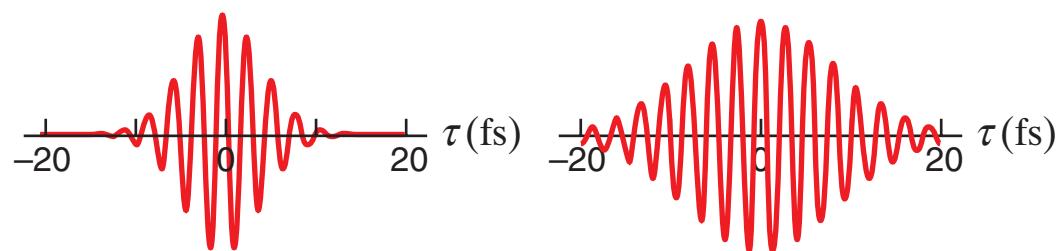
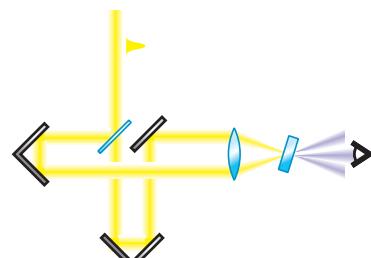
Temporal characterization



Temporal characterization



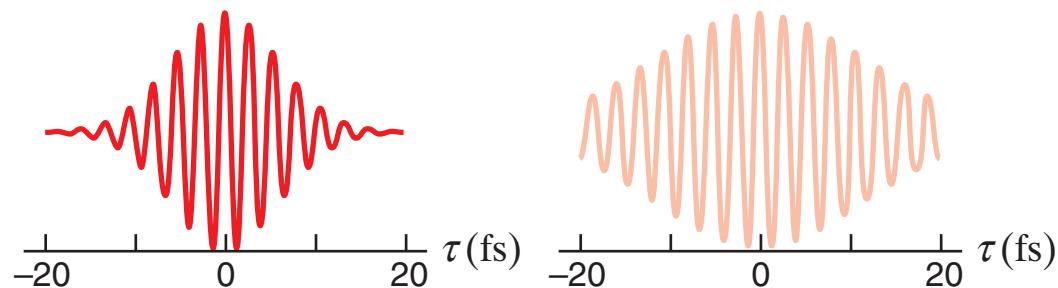
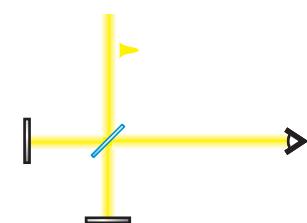
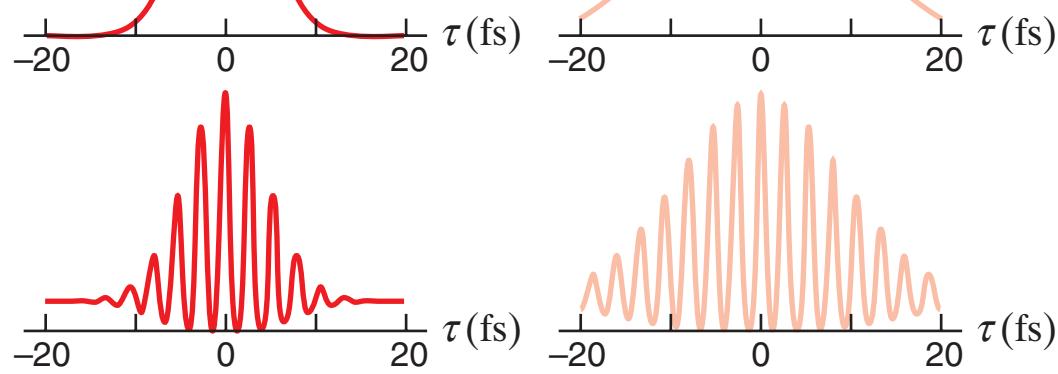
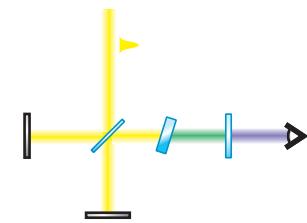
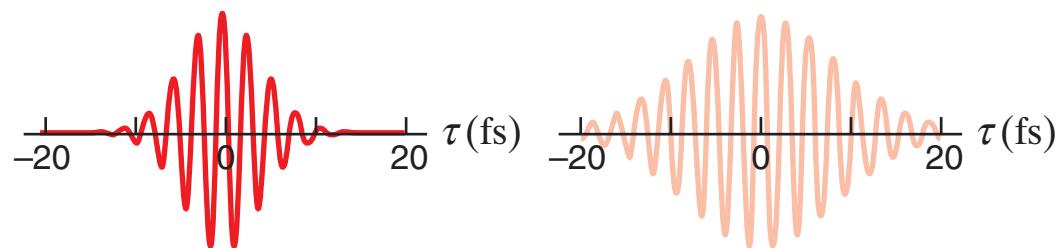
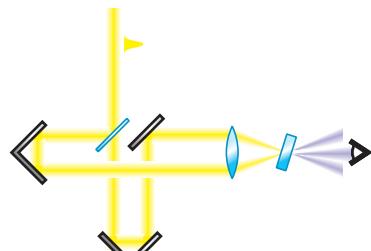
Temporal characterization



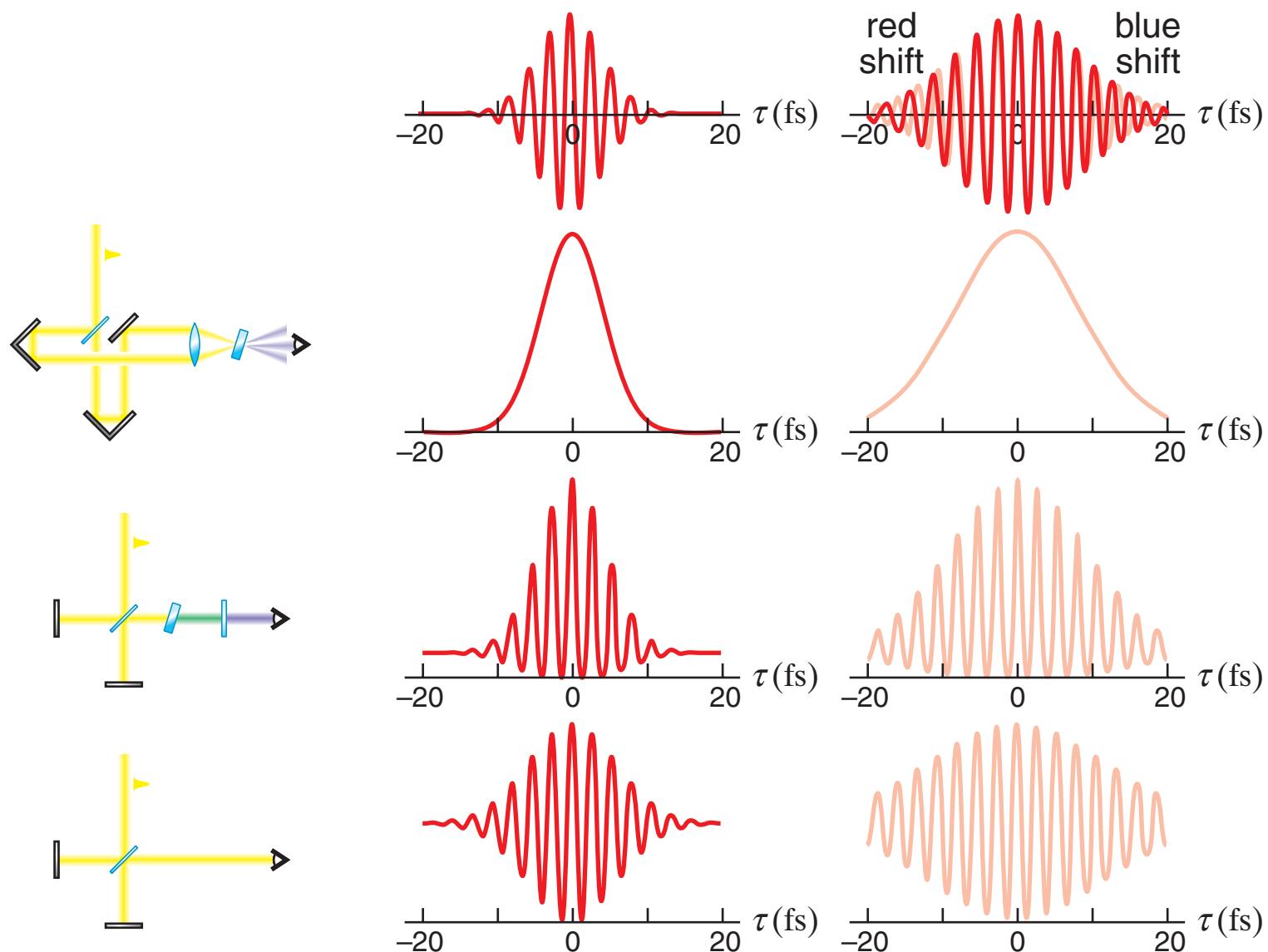
Temporal characterization

But what about dispersion?

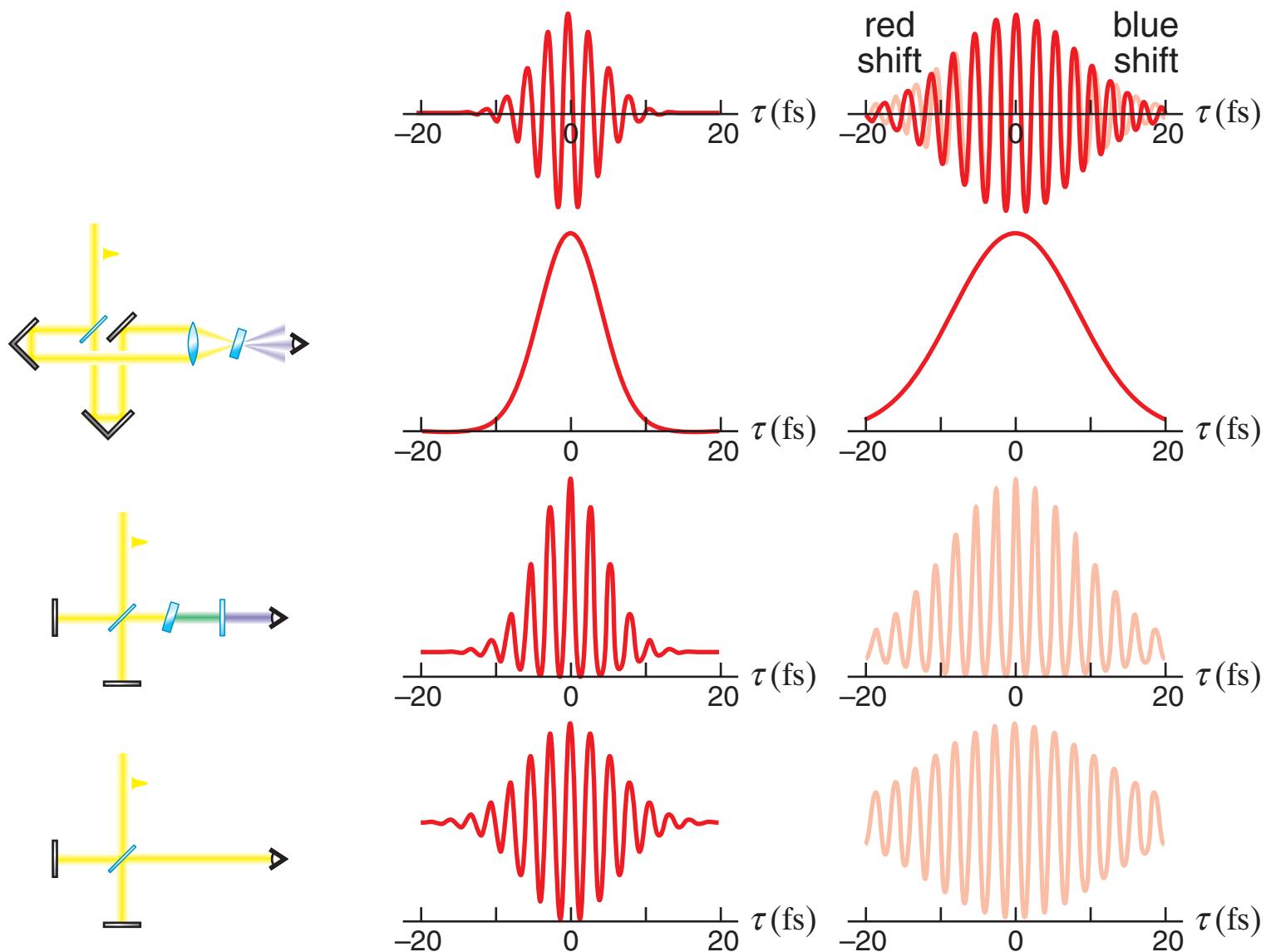
Temporal characterization



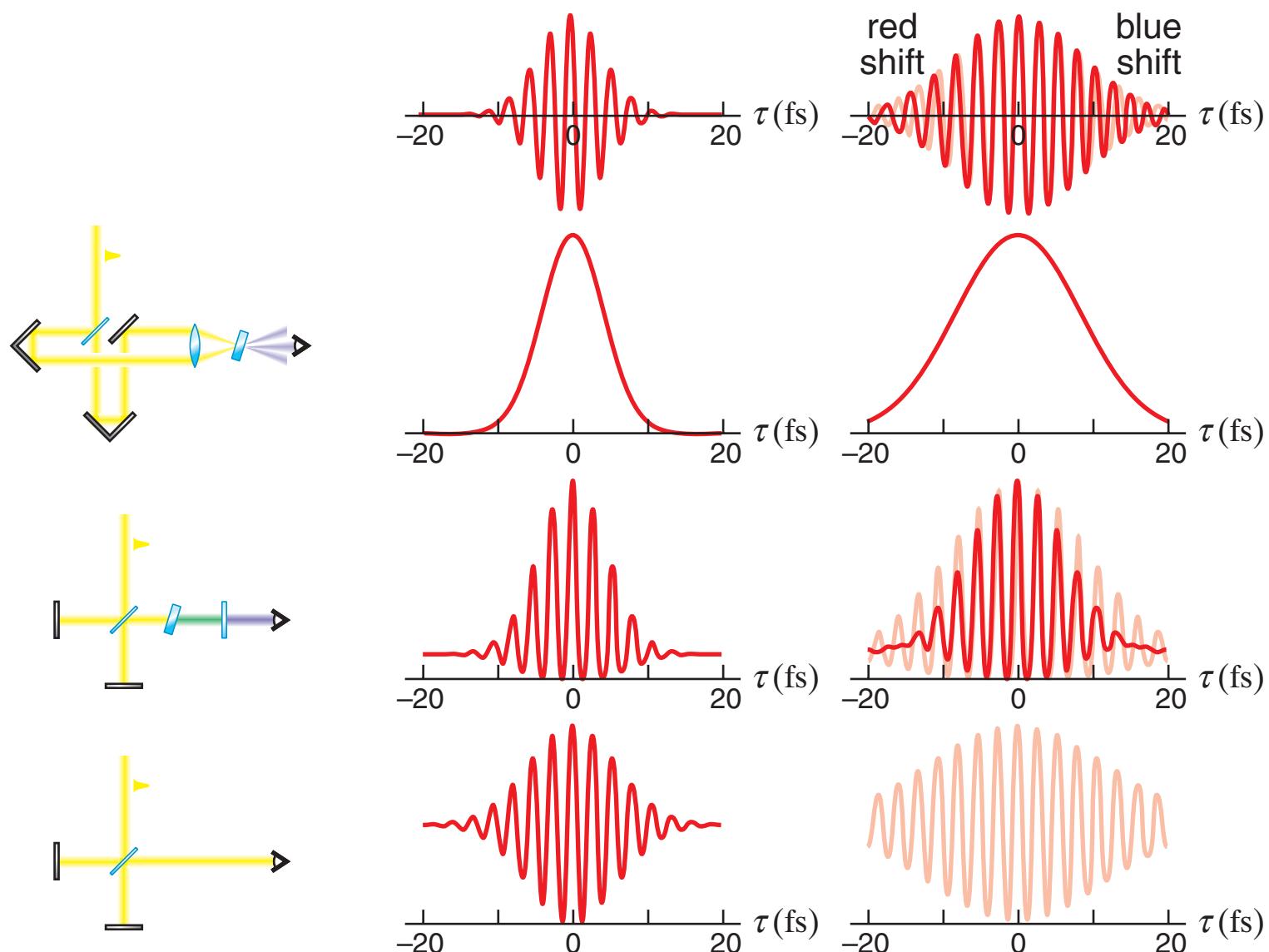
Temporal characterization



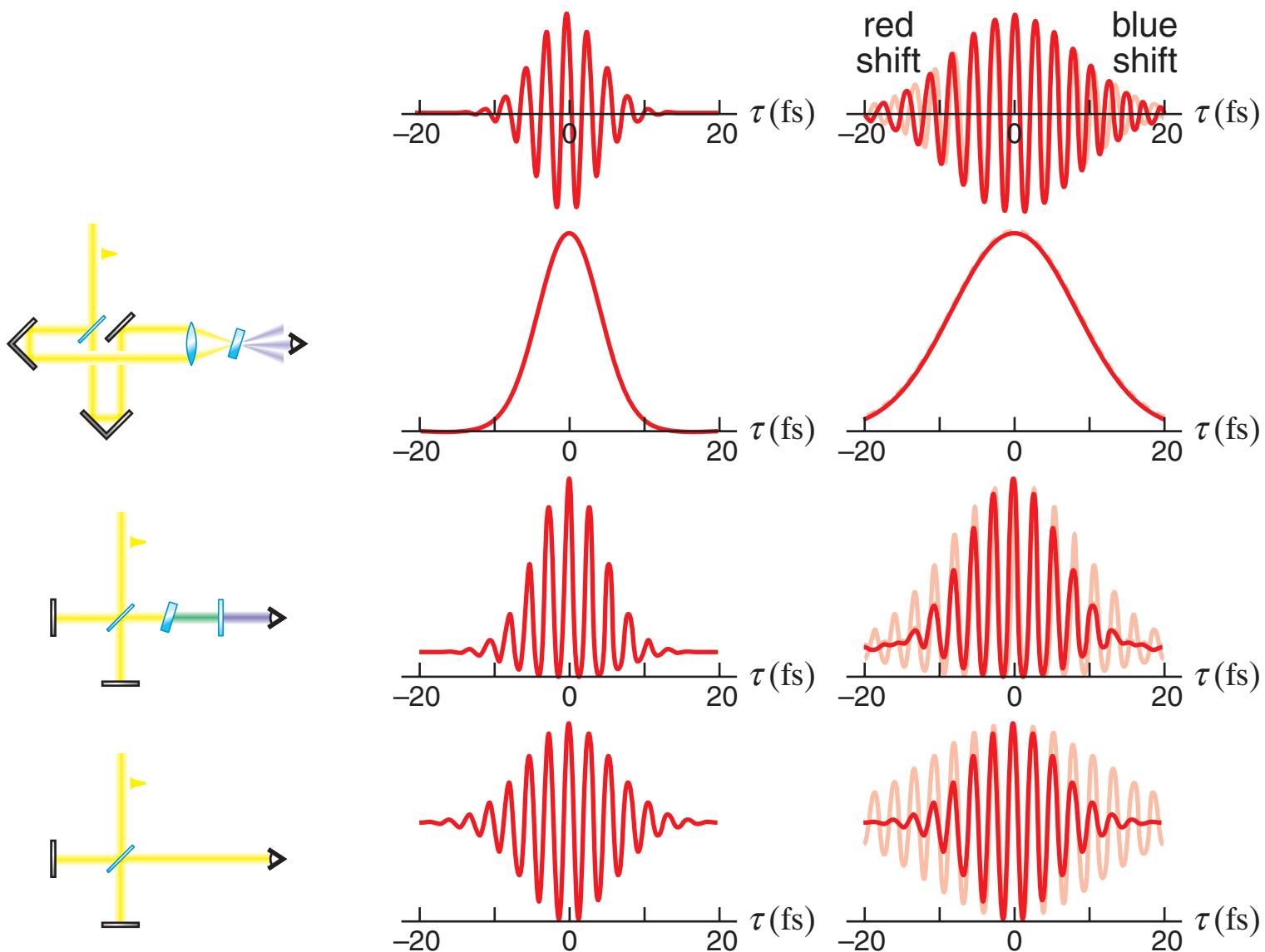
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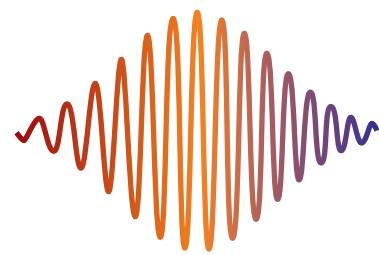
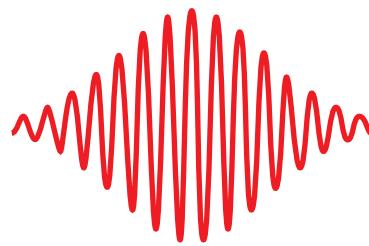
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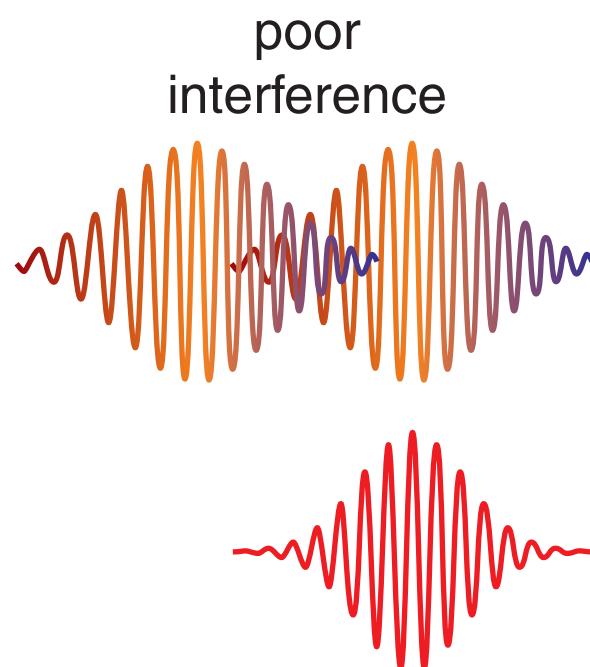
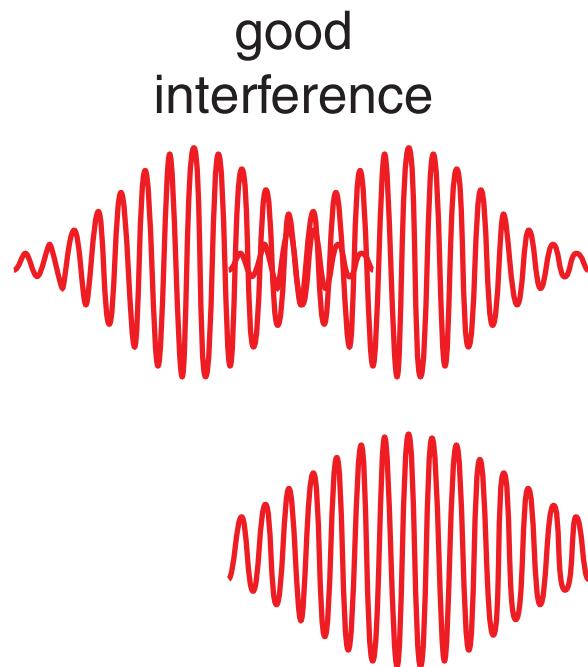
Temporal characterization



Temporal characterization



Temporal characterization



Temporal characterization

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

Temporal characterization

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$$f_1(t) \otimes f_2(t) \equiv \int f_1(t+\tau) {f_2}^*(t) dt = \mathcal{F}^{-1}\{f_1(\omega) {f_2}^*(\omega)\}$$

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Interference term in linear autocorrelation:

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

Temporal characterization

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Interference term in linear autocorrelation:

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Temporal characterization

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

$$f_1(t) \otimes f_2(t) \equiv \int f_1(t + \tau) {f_2}^*(t) dt = \mathcal{F}^{-1}\{f_1(\omega) {f_2}^*(\omega)\}$$

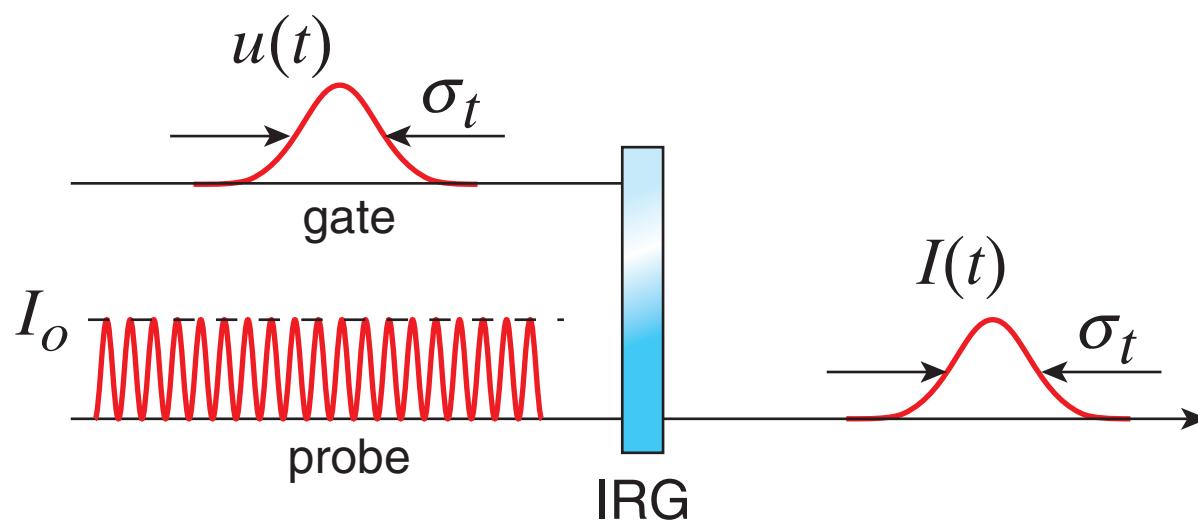
Interference term in linear autocorrelation:

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

$$= \mathcal{F}^{-1}\{E_{orig}(\omega) e^{i\phi(\omega)} {E_{orig}}^*(\omega) e^{-i\phi(\omega)}\} =$$

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

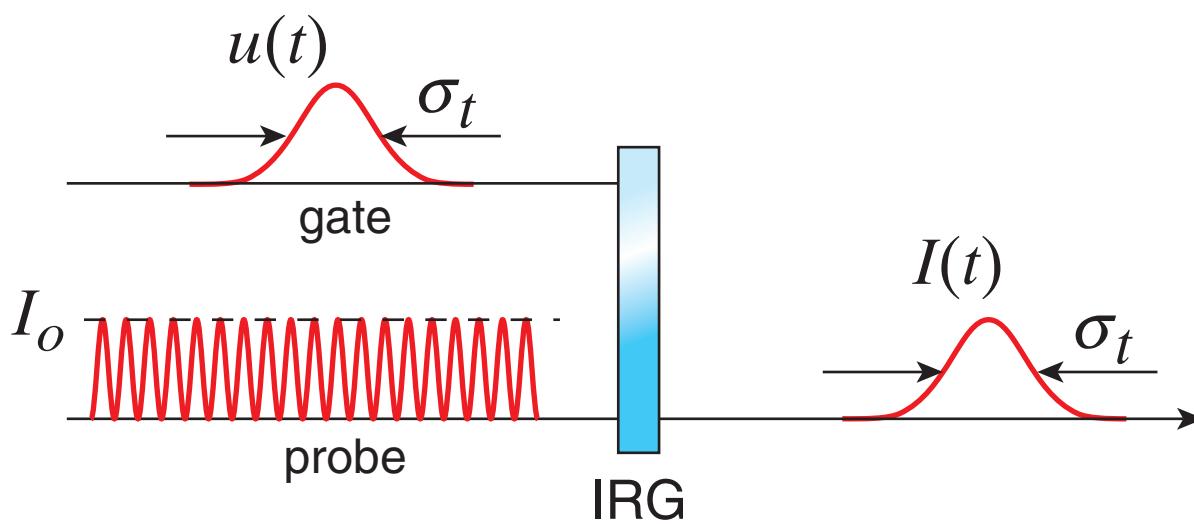
Joint time-frequency measurements



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

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

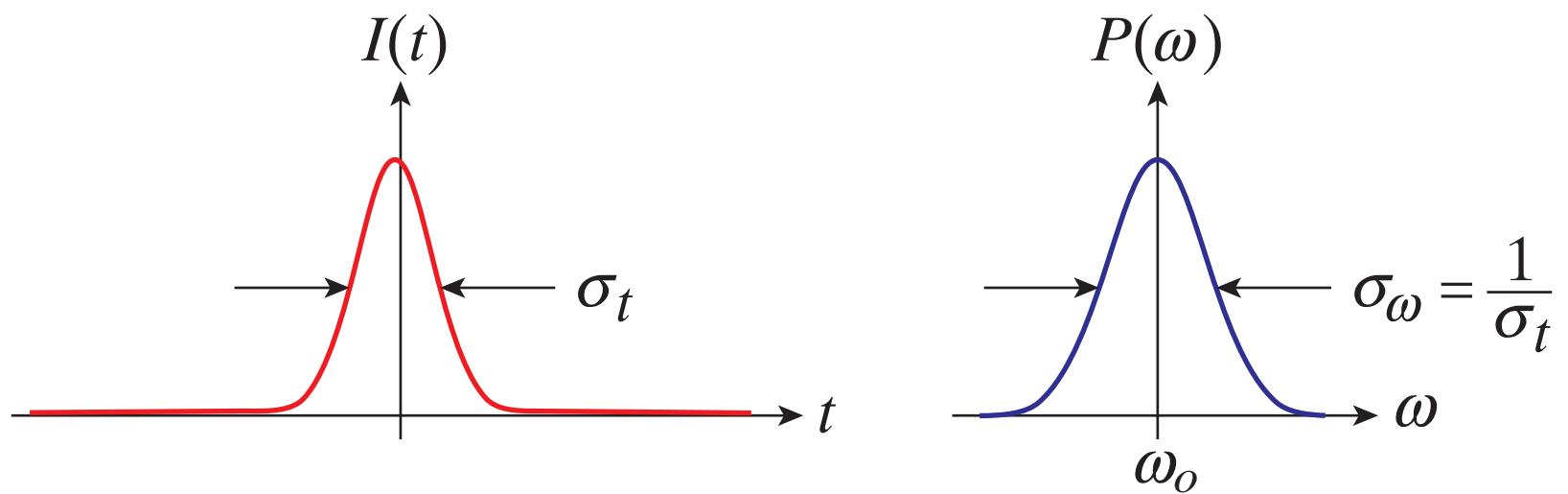
Joint time-frequency measurements



Transmitted intensity

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

Joint time-frequency measurements

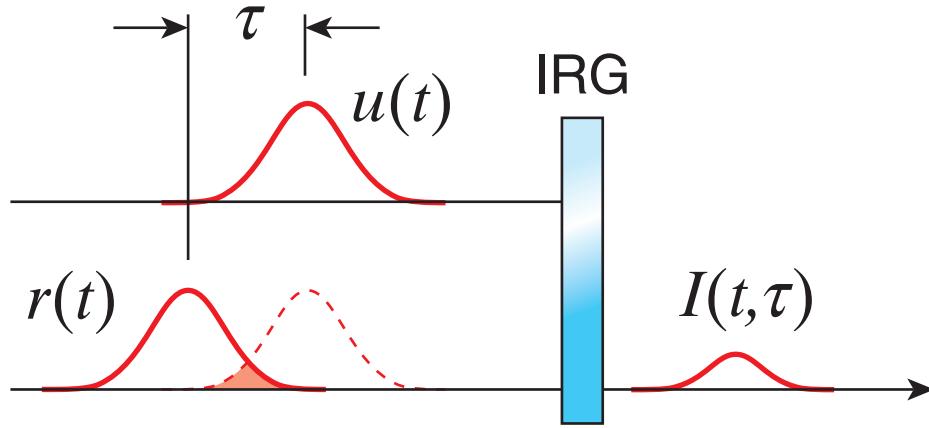


Transmitted intensity

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

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

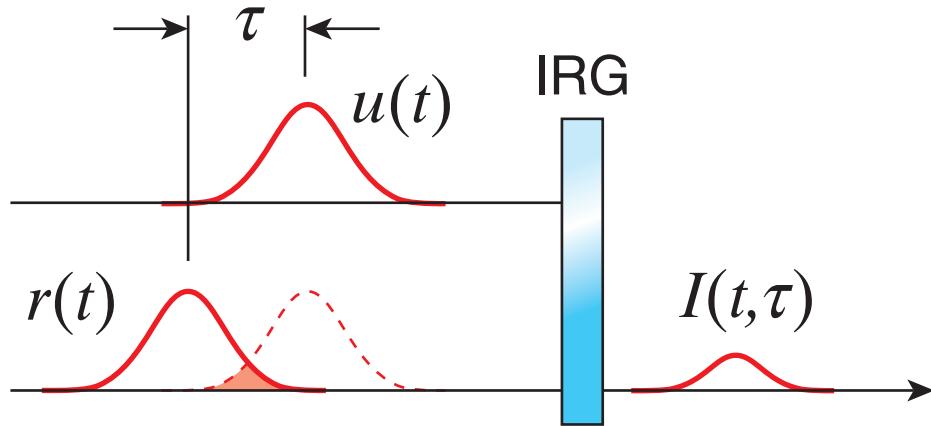
Joint time-frequency measurements



Transmitted intensity

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

Joint time-frequency measurements

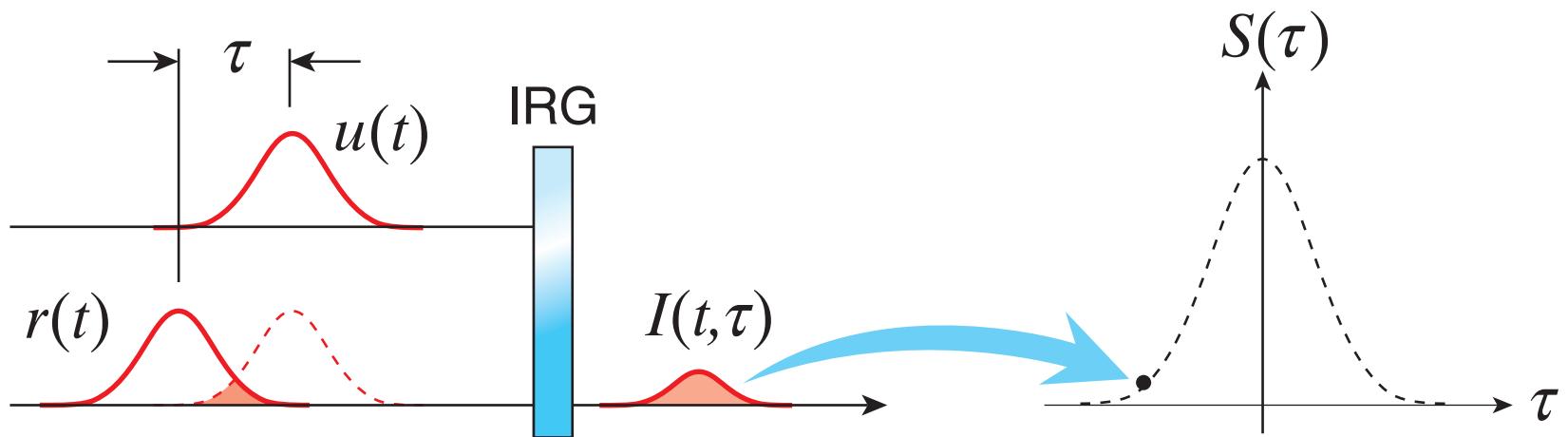


Transmitted intensity

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

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

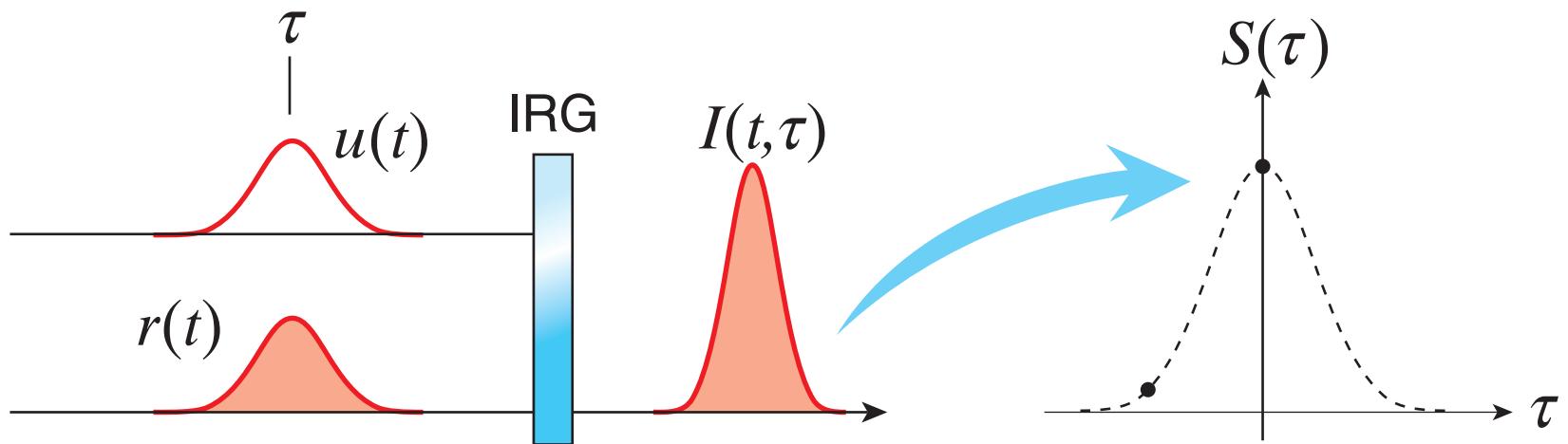
Joint time-frequency measurements



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

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

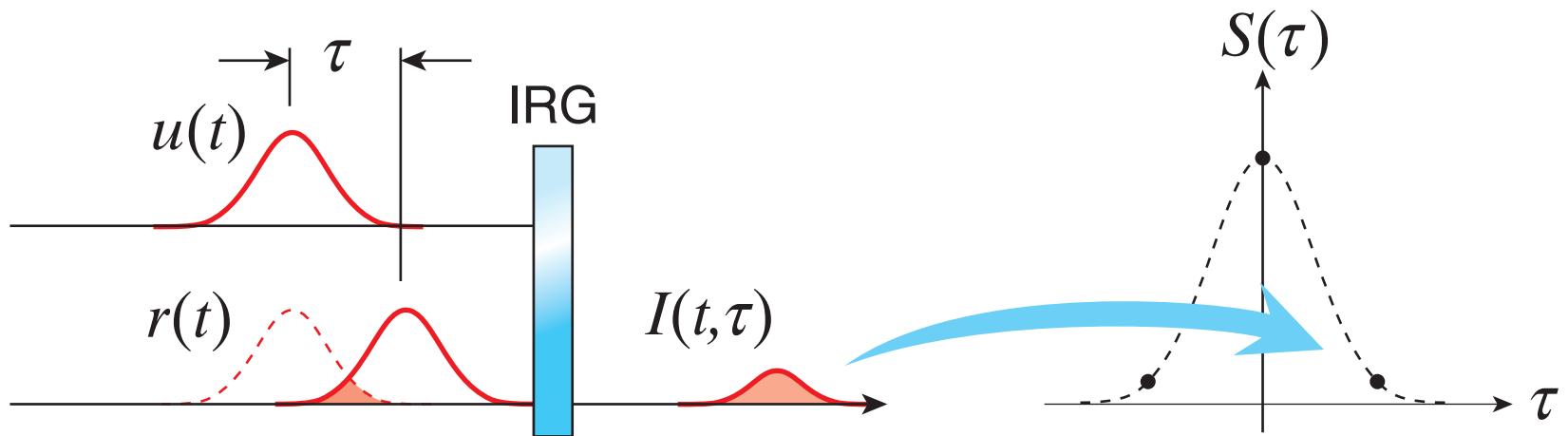
Joint time-frequency measurements



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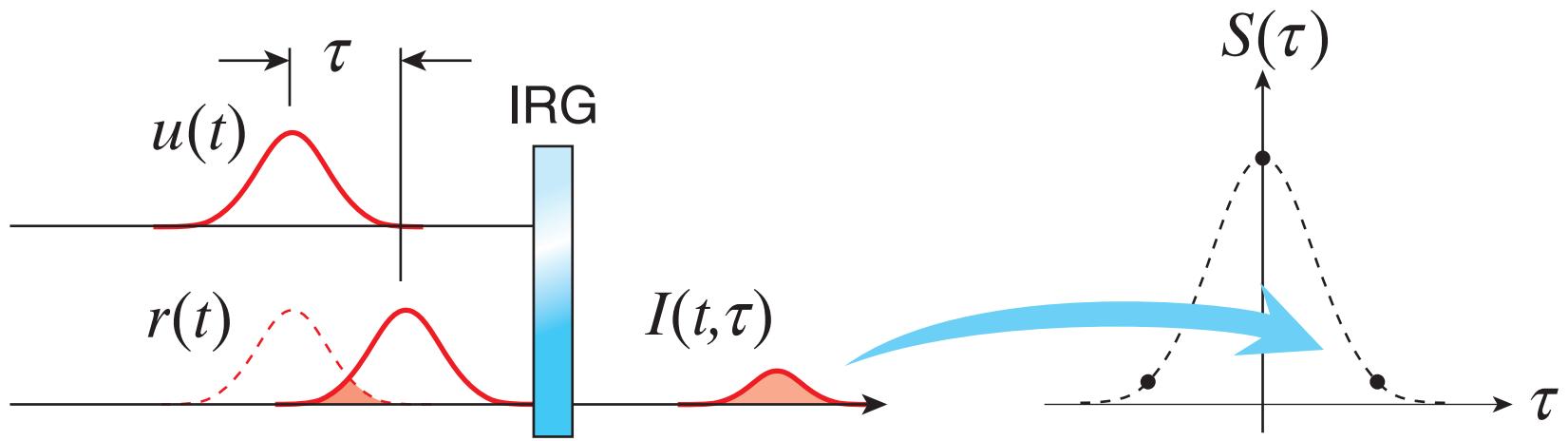
Joint time-frequency measurements



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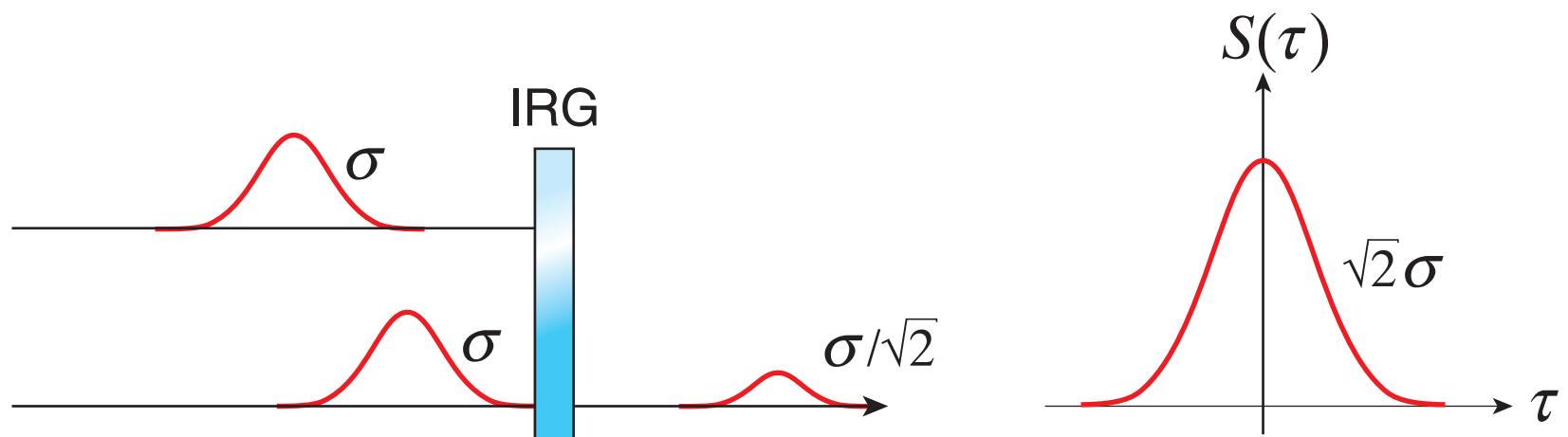
Joint time-frequency measurements



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

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

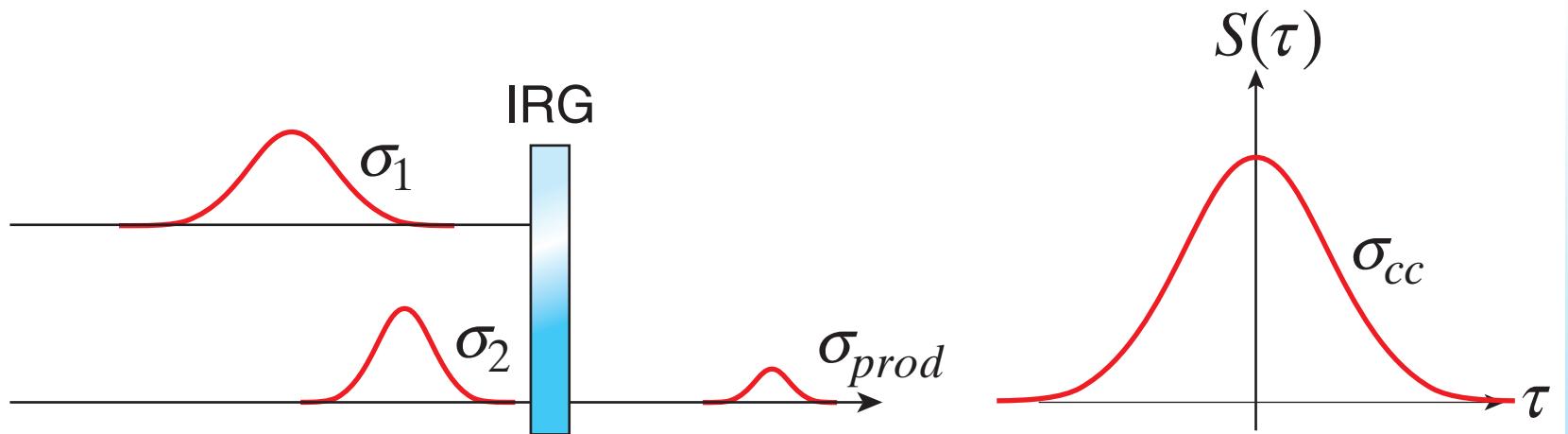
Joint time-frequency measurements



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Joint time-frequency measurements

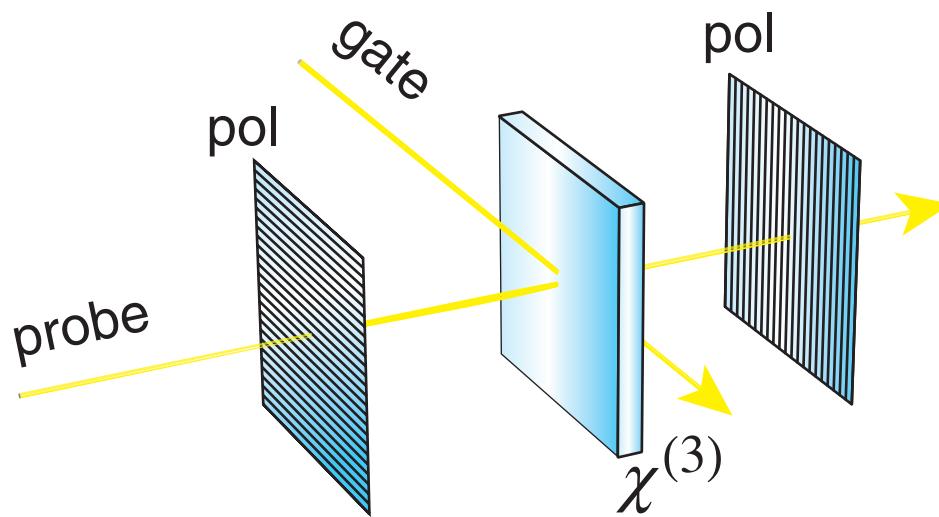


If gate and probe unequal:

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

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

Joint time-frequency measurements

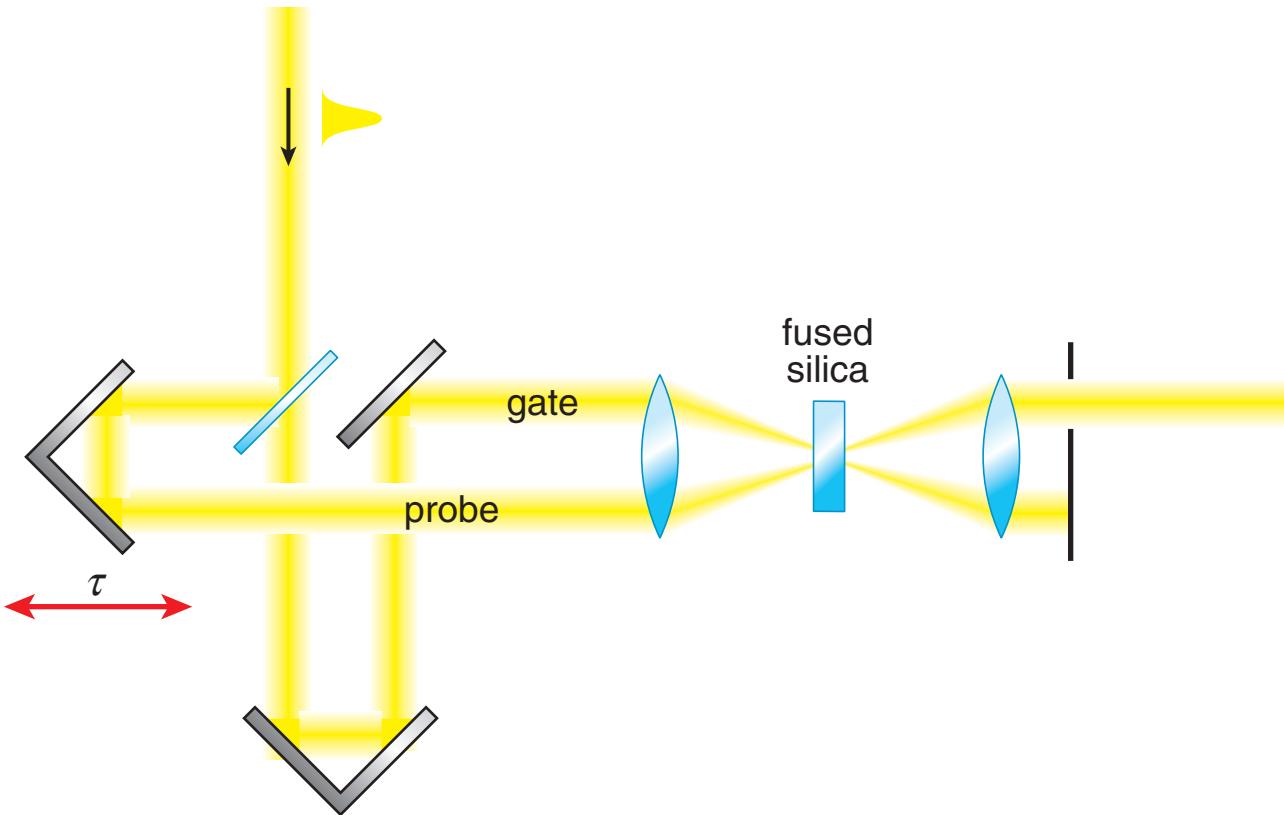


Transmitted field:

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

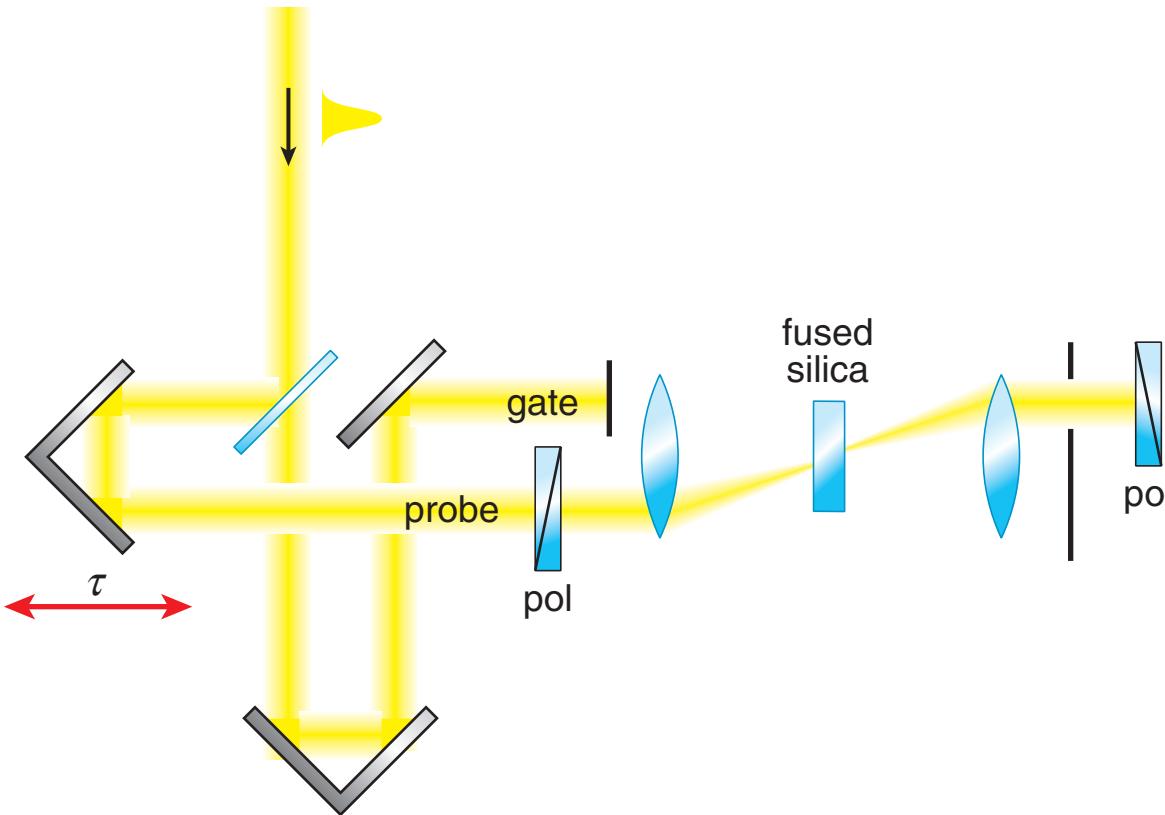
Joint time-frequency measurements

FROG: frequency-resolved optical gating



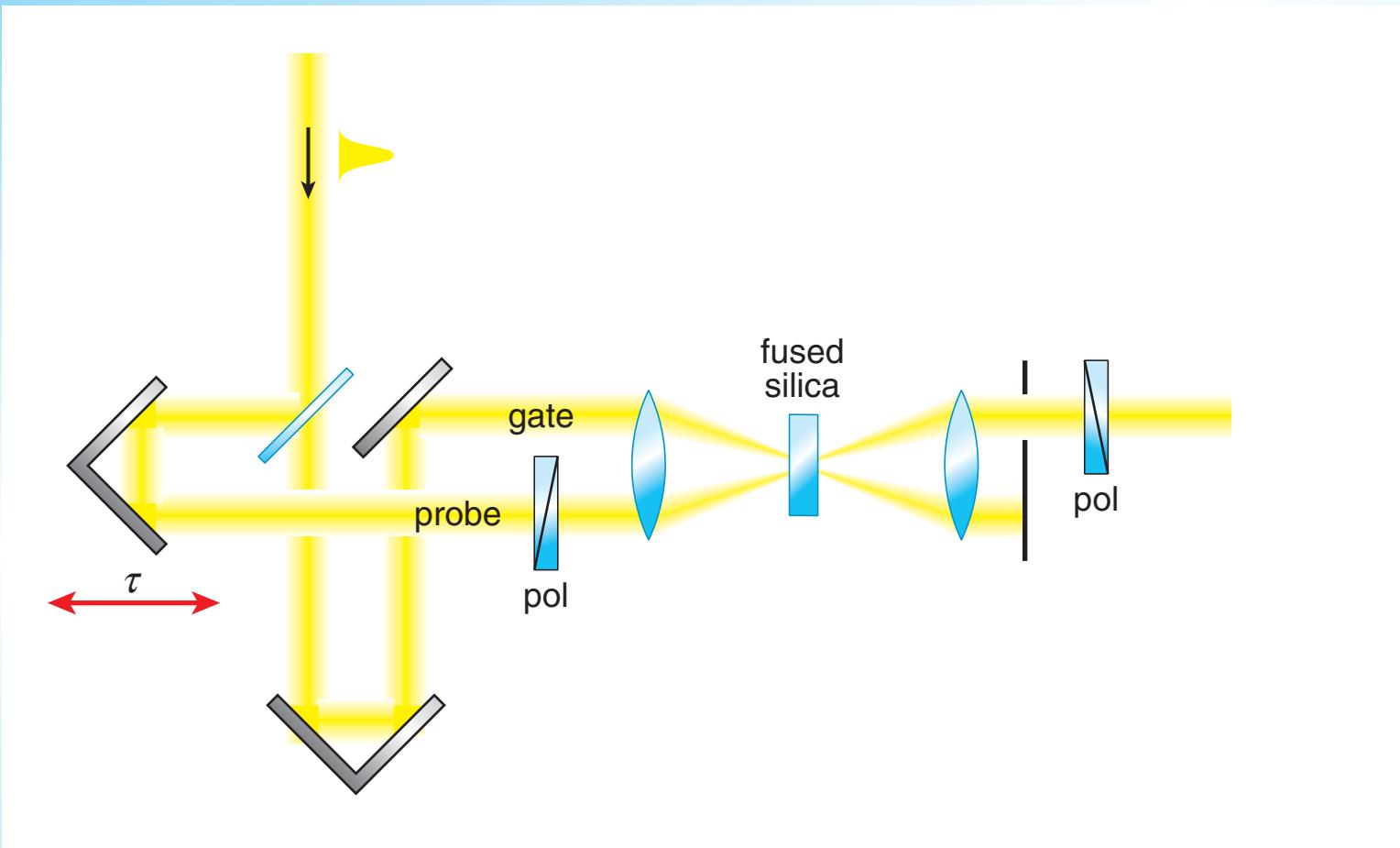
Joint time-frequency measurements

FROG: frequency-resolved optical gating



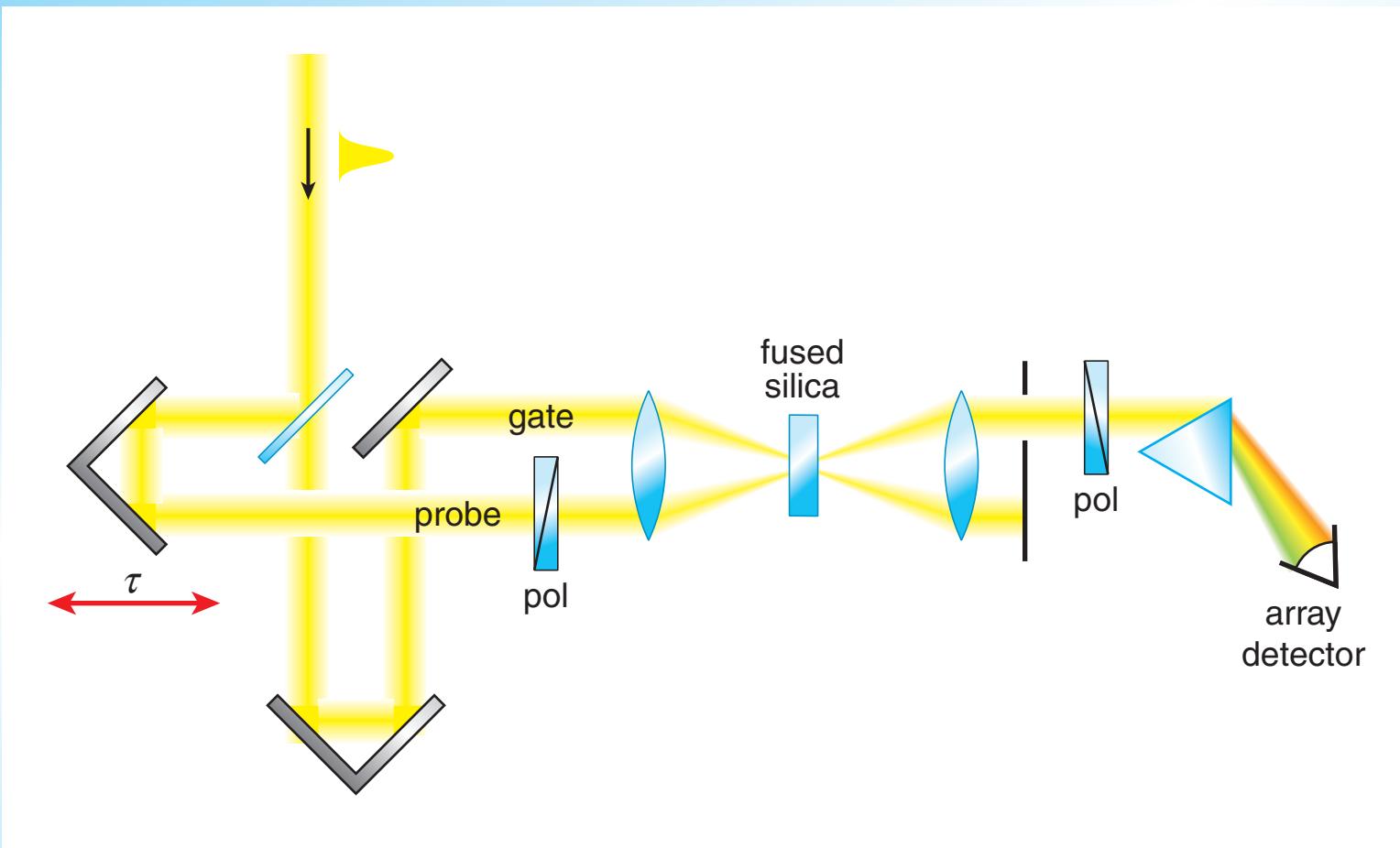
Joint time-frequency measurements

FROG: frequency-resolved optical gating

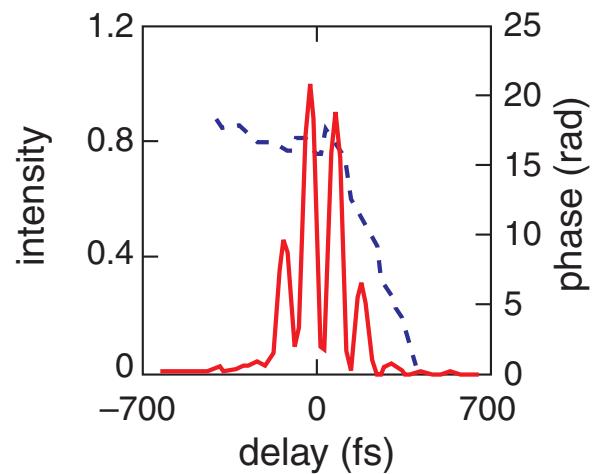
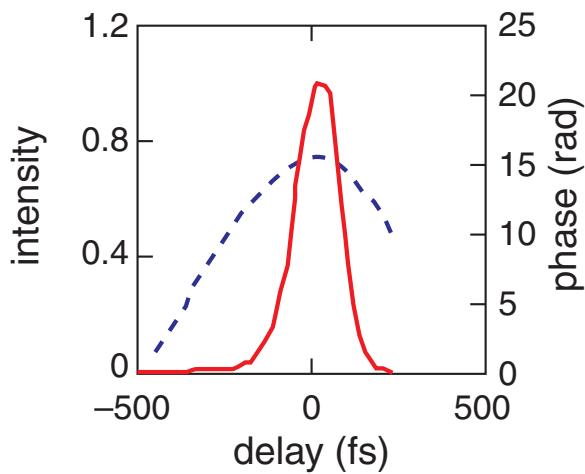
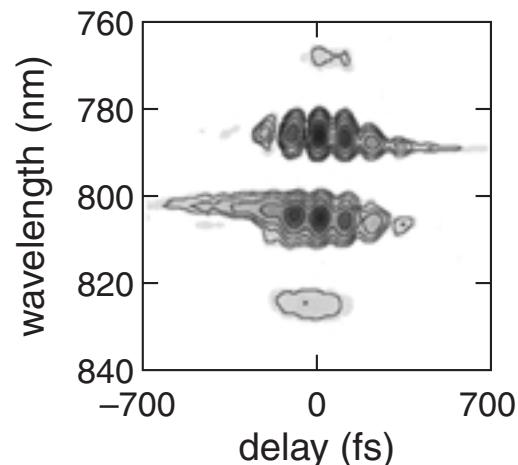
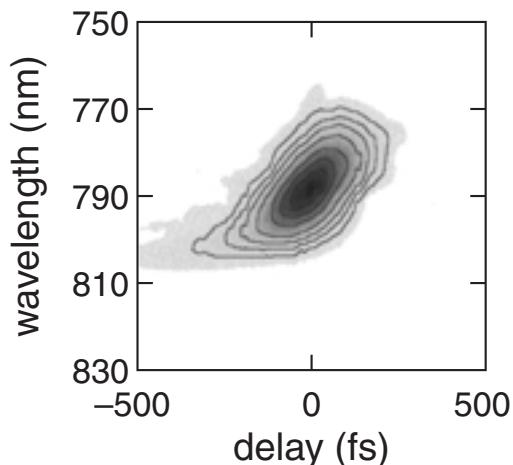


Joint time-frequency measurements

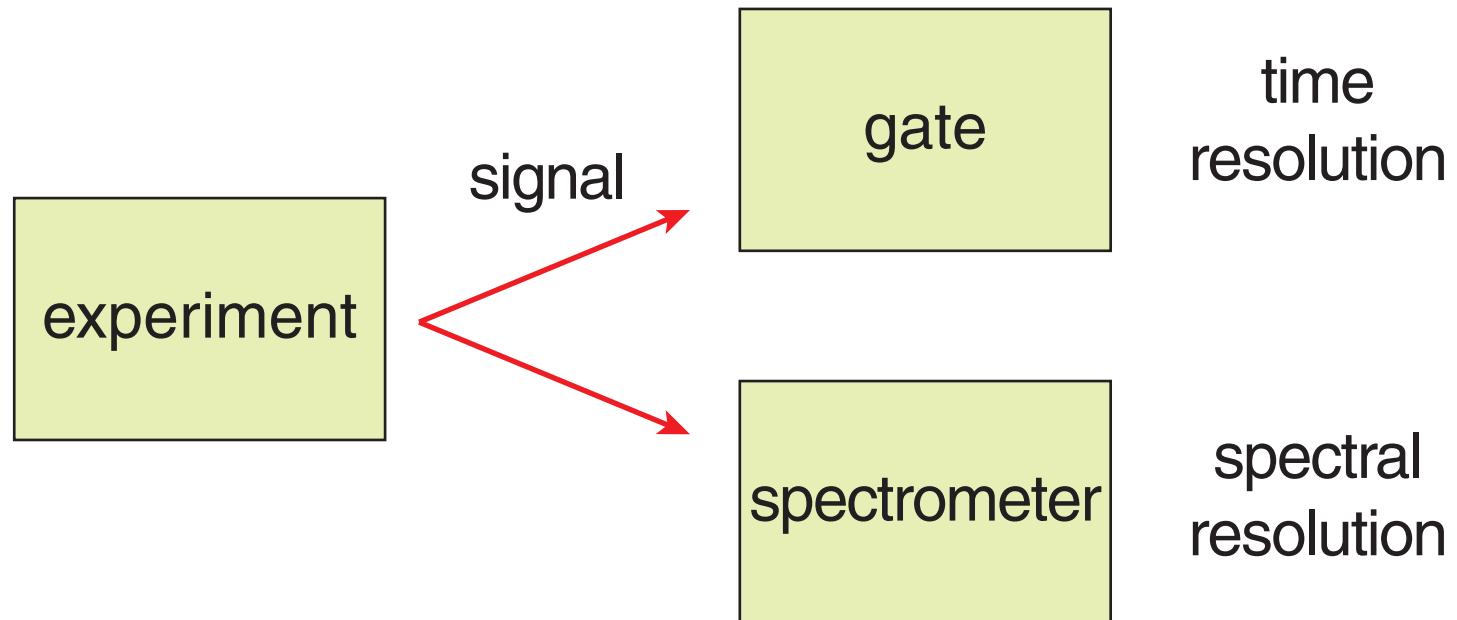
FROG: frequency-resolved optical gating



Joint time-frequency measurements



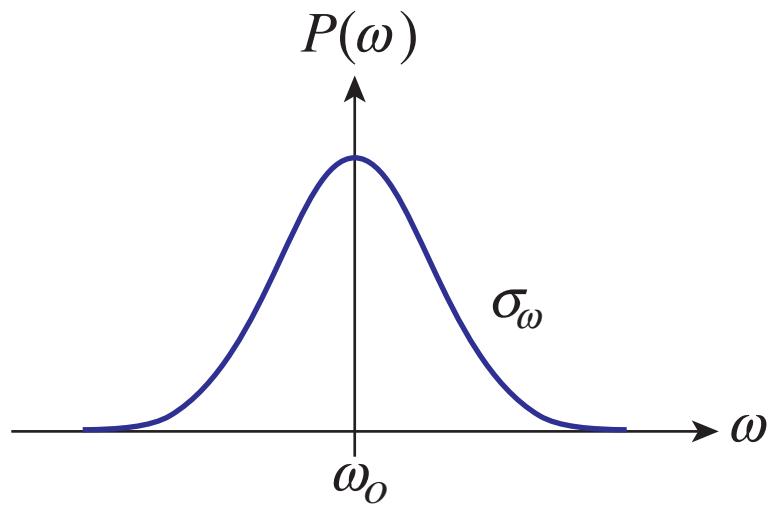
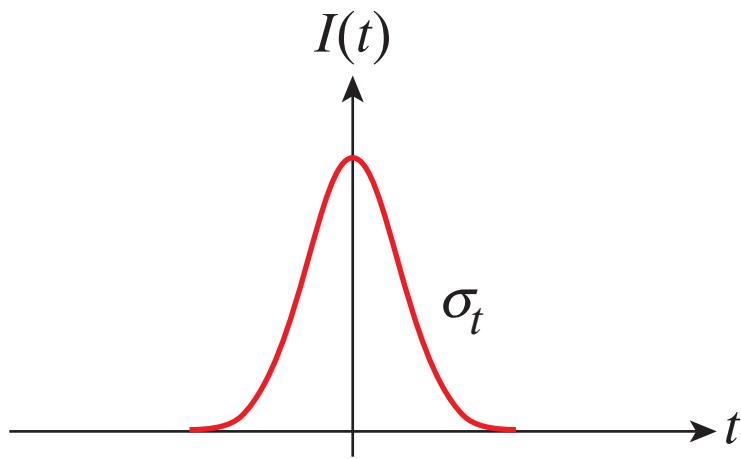
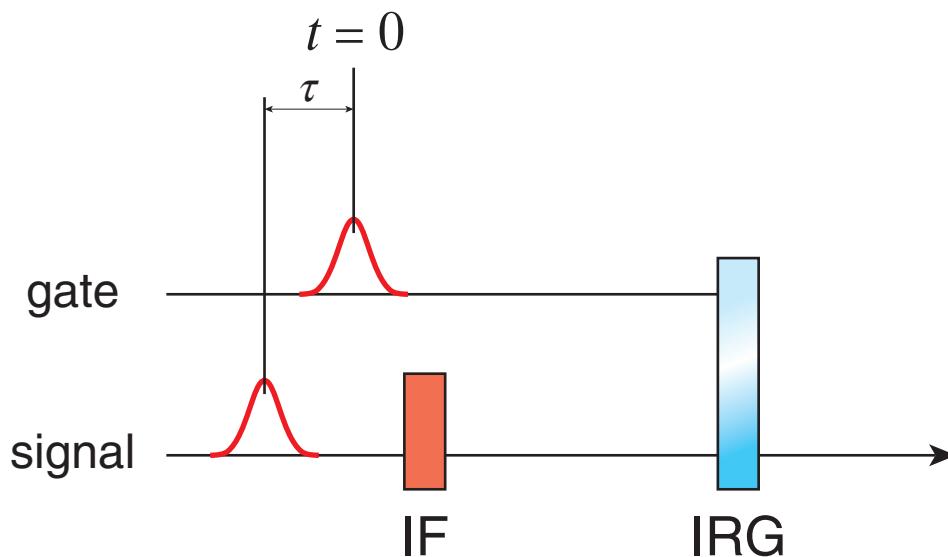
Joint time-frequency measurements



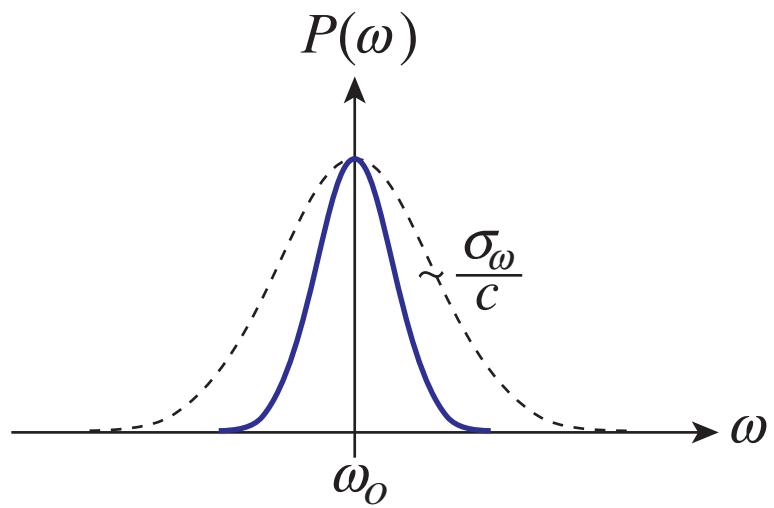
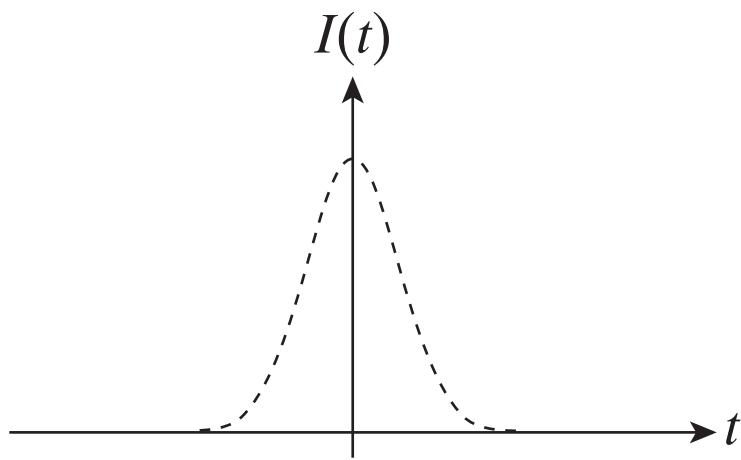
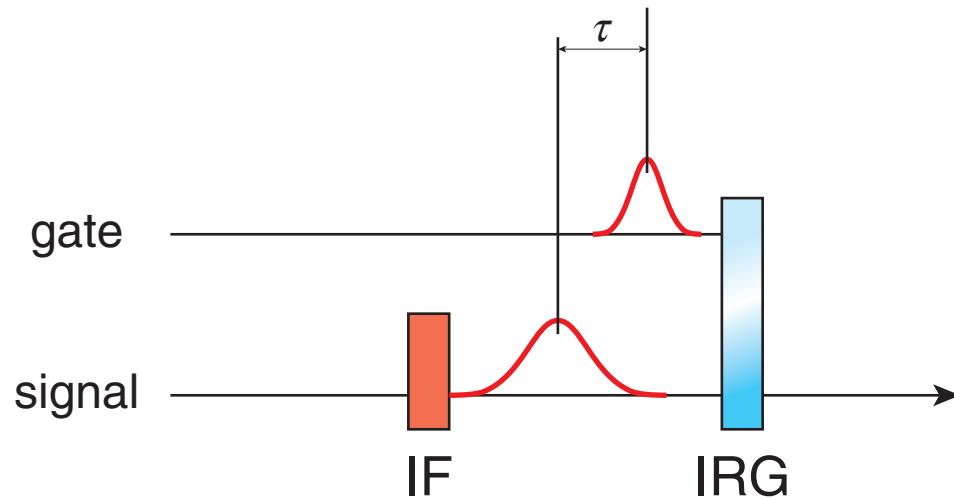
Joint time-frequency measurements

What are the resolution limits?

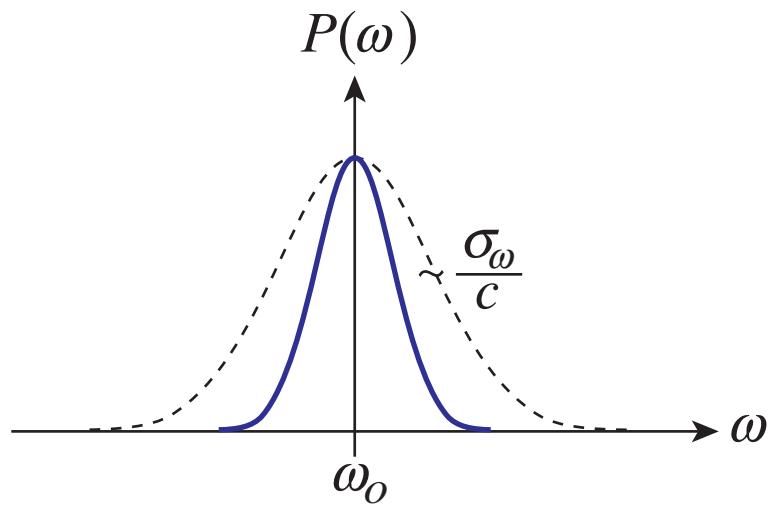
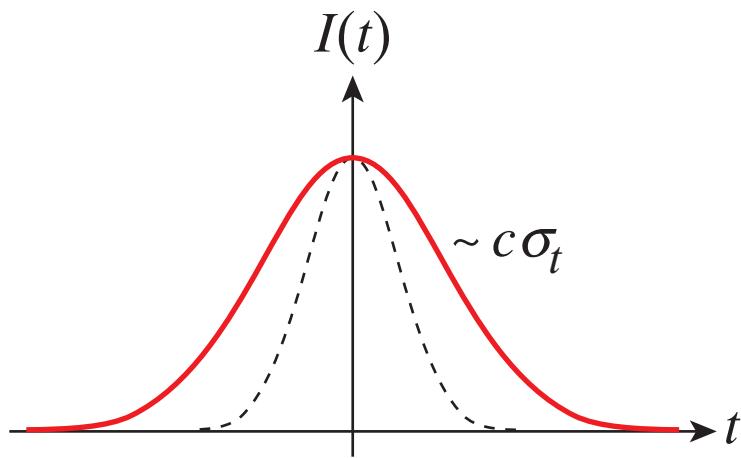
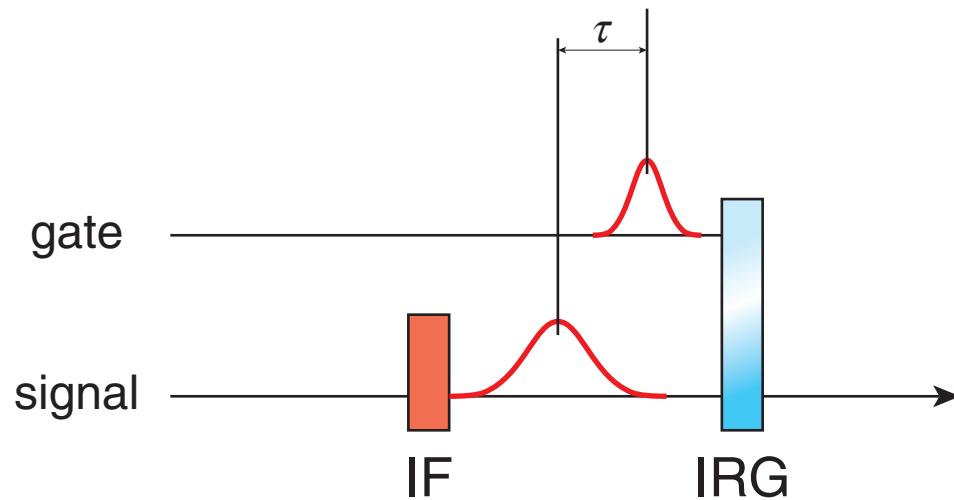
Experiment 1



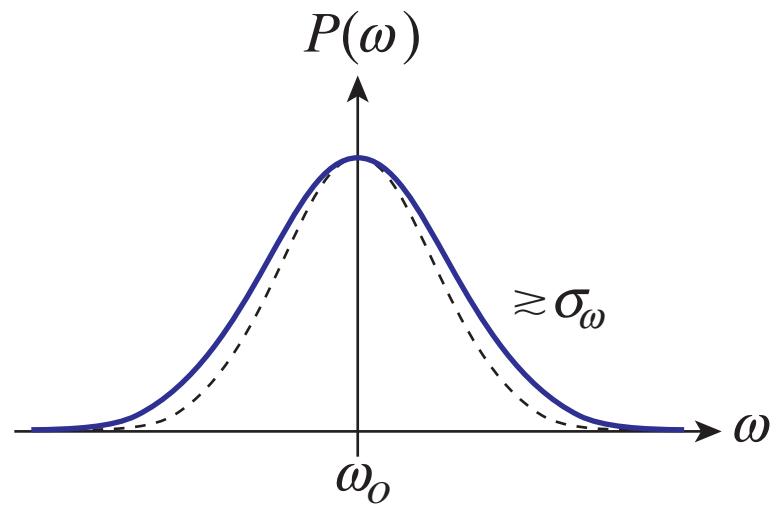
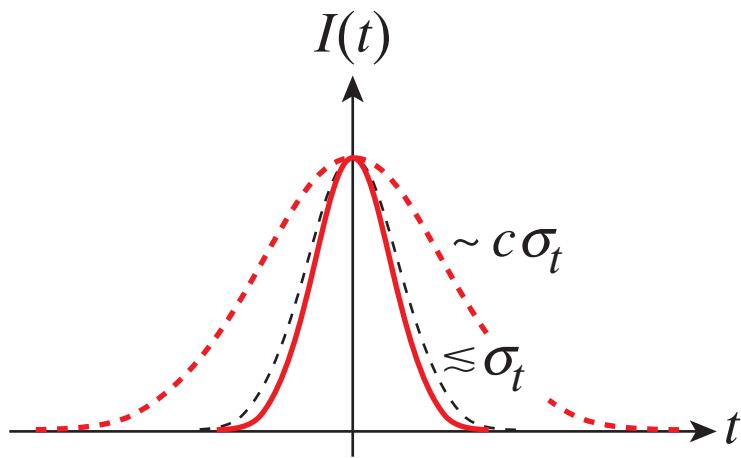
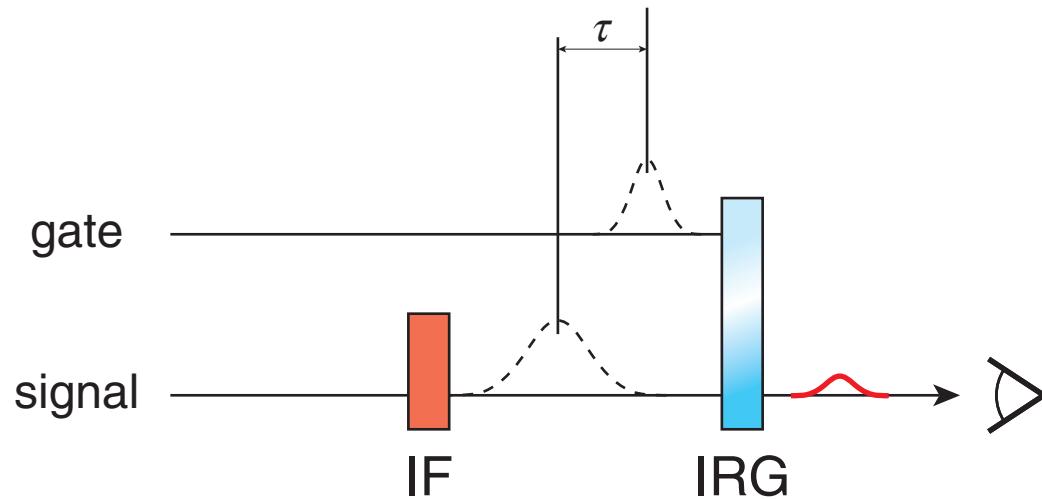
Experiment 1



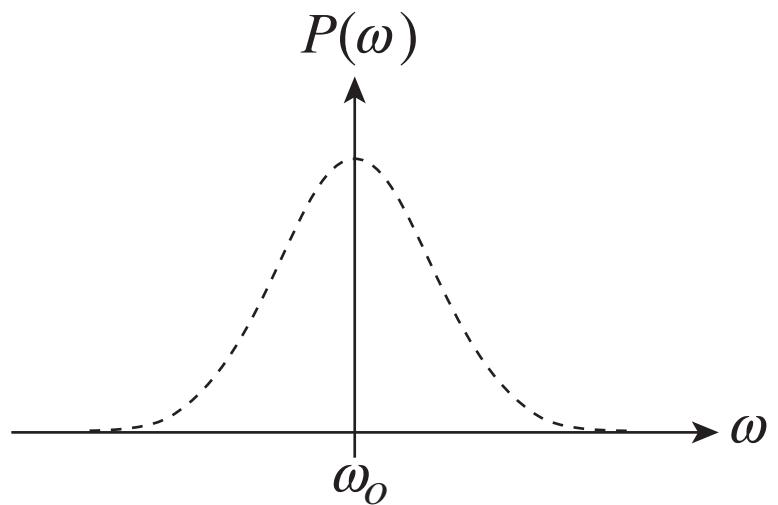
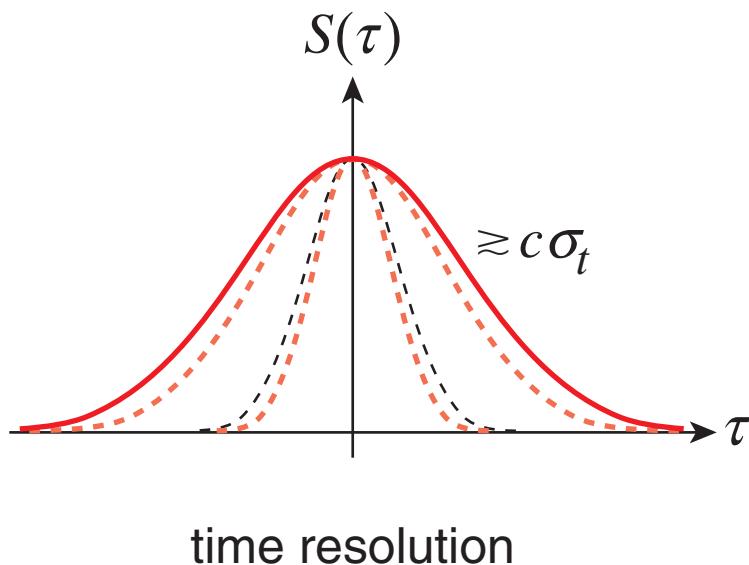
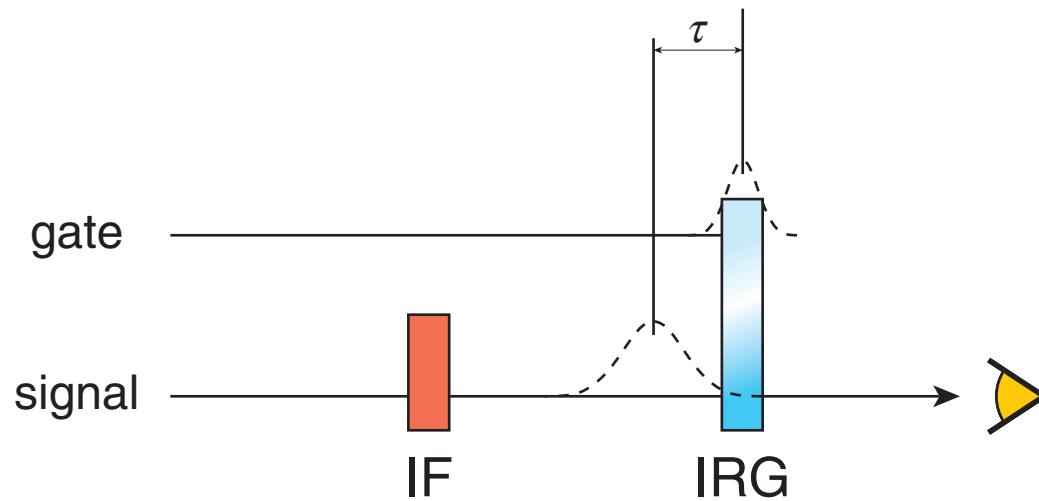
Experiment 1



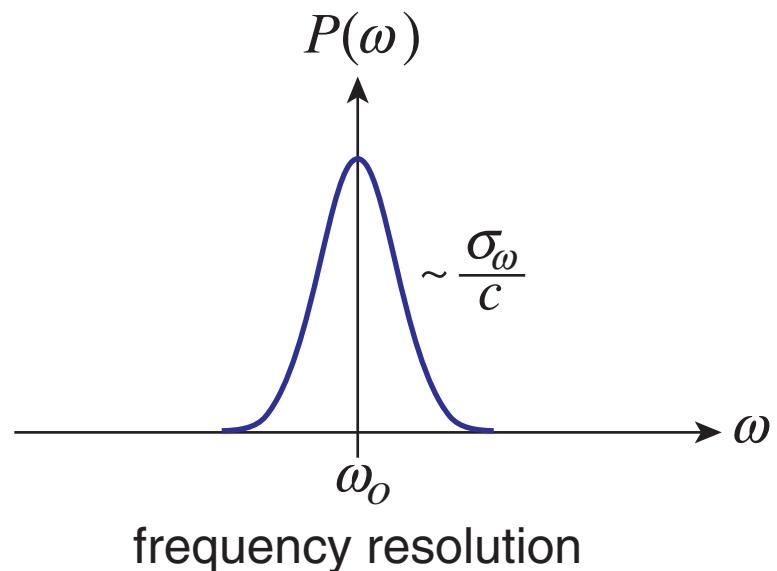
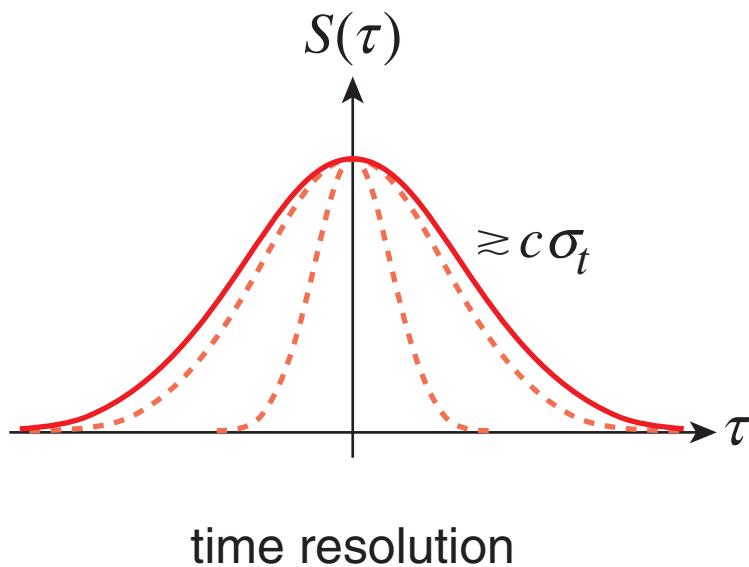
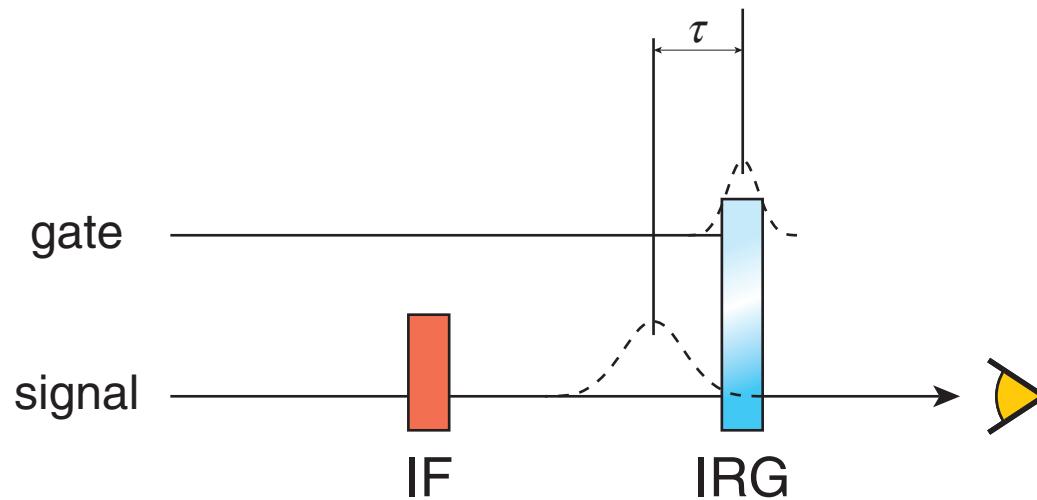
Experiment 1



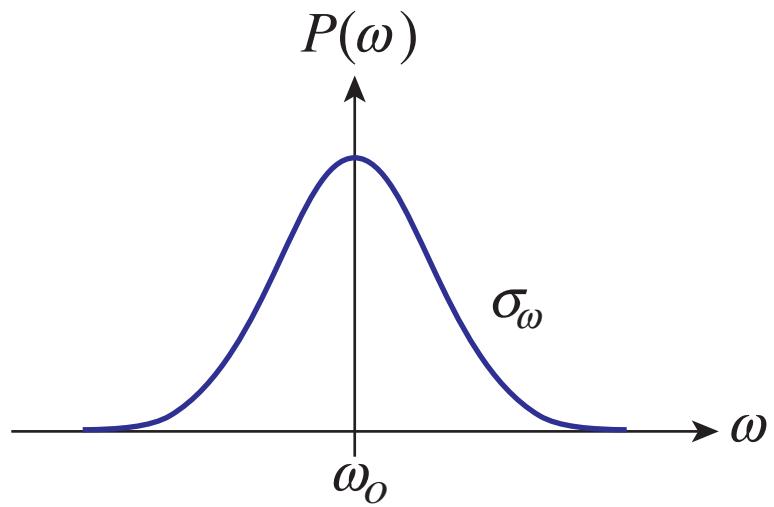
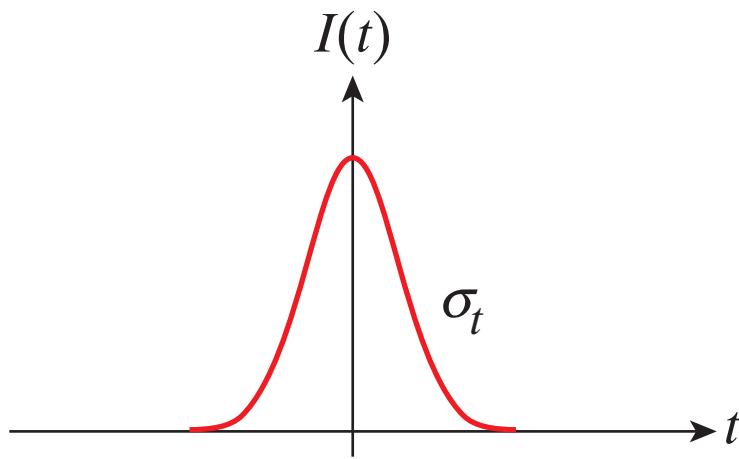
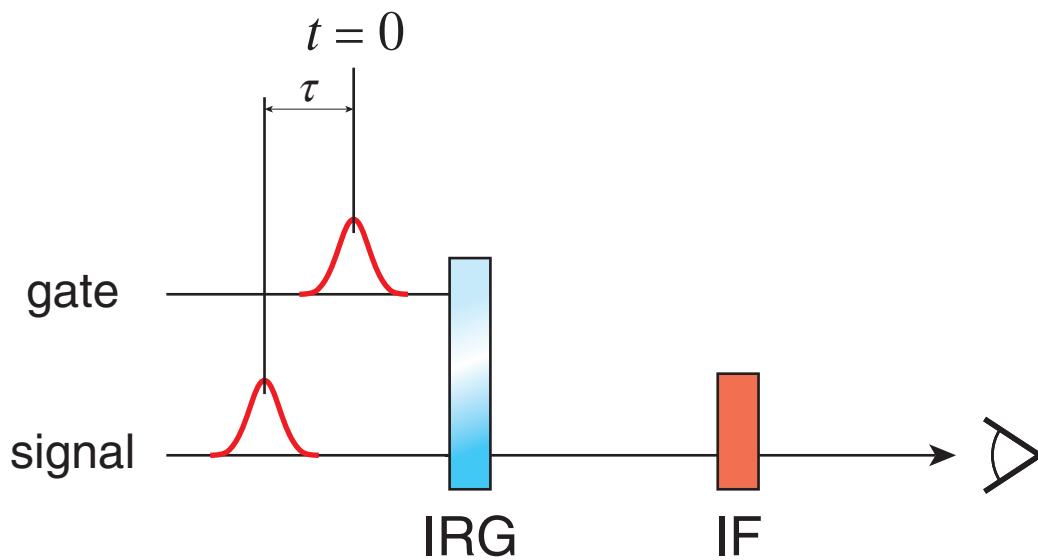
Experiment 1



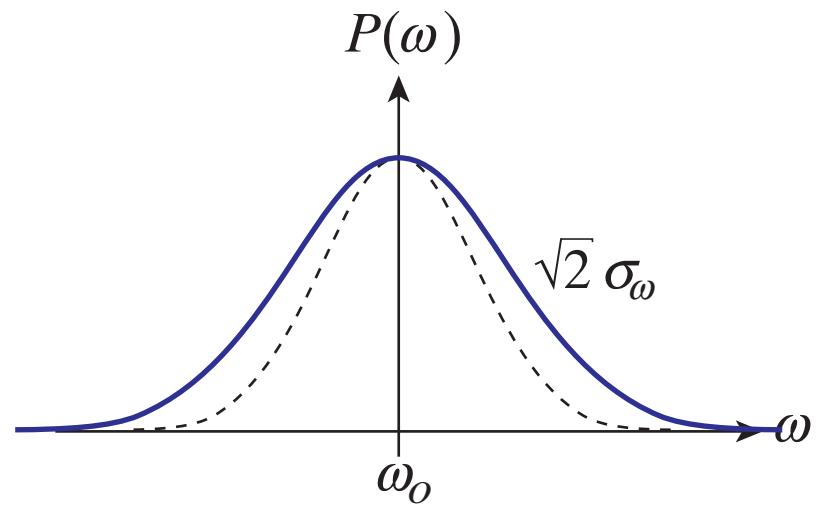
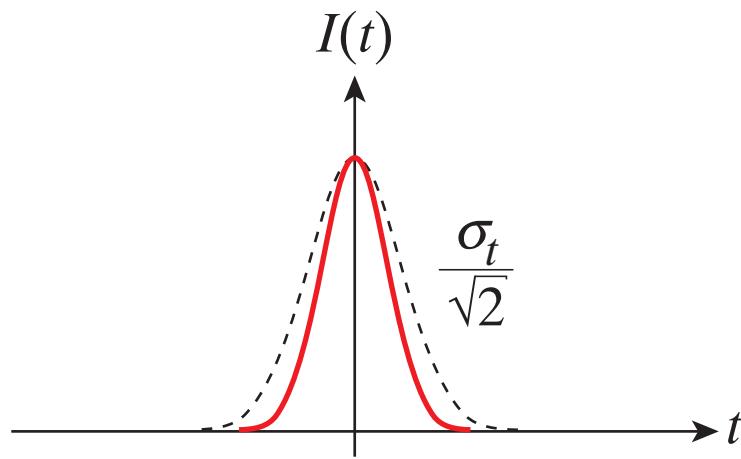
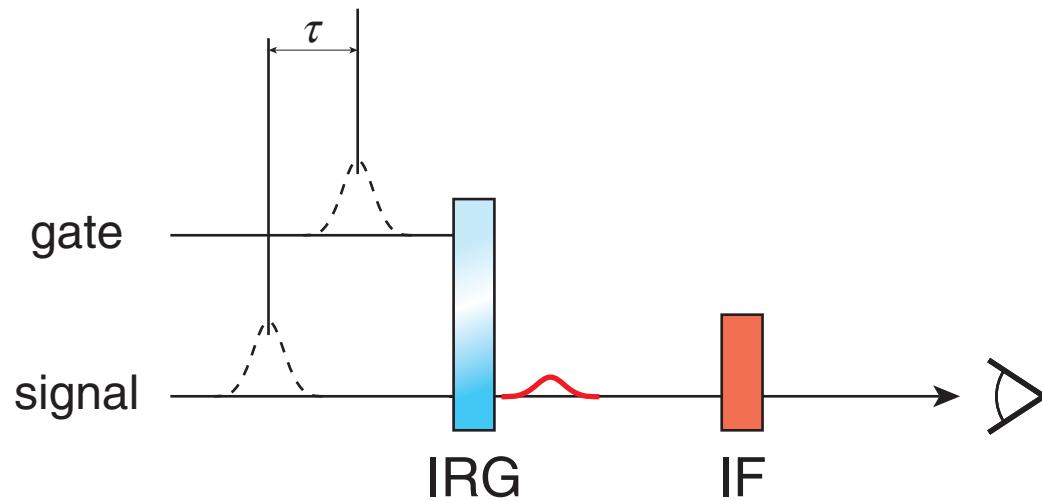
Experiment 1



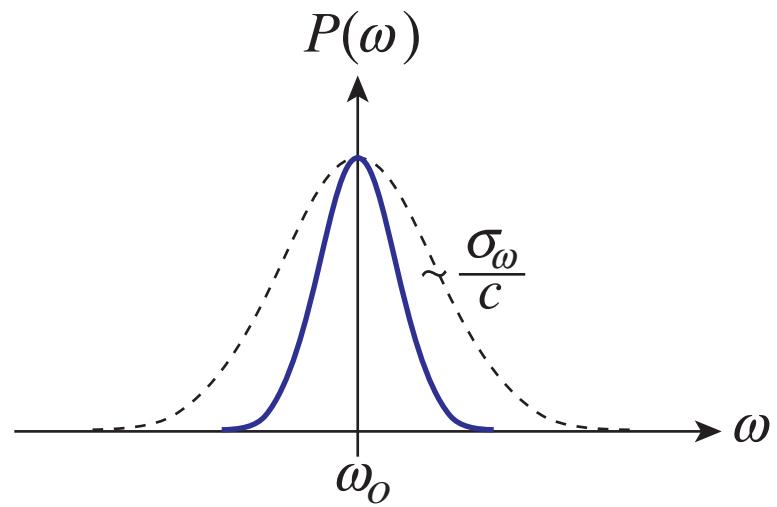
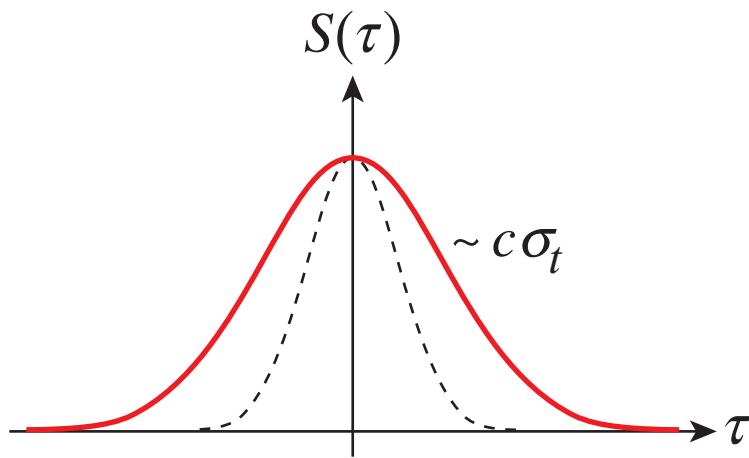
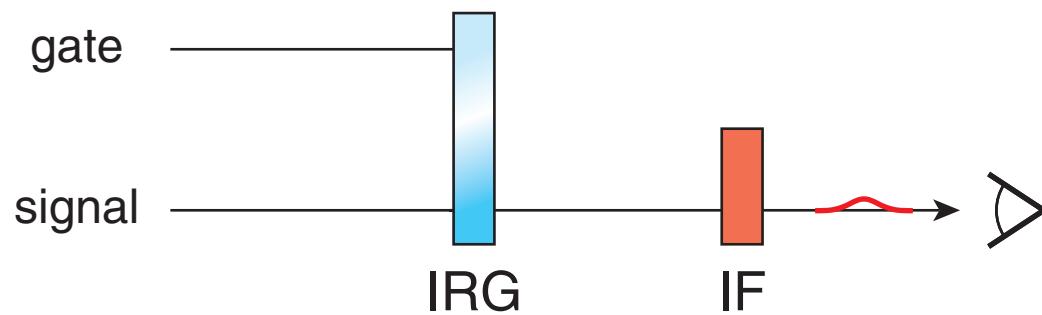
Experiment 2



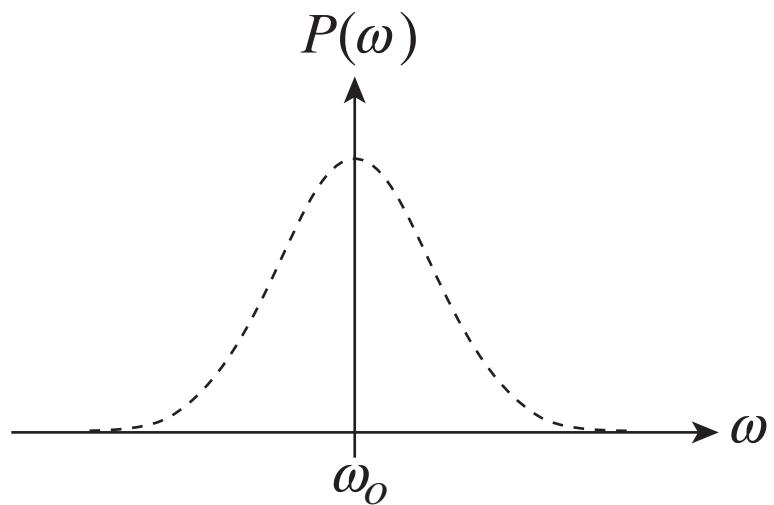
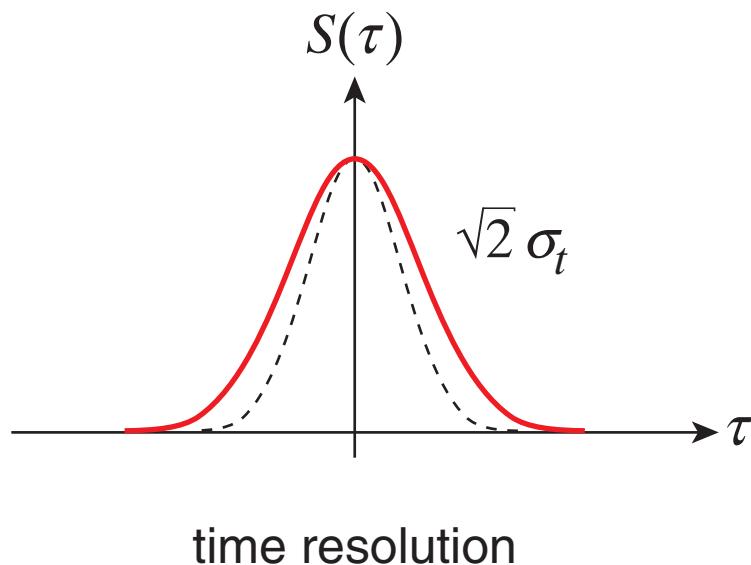
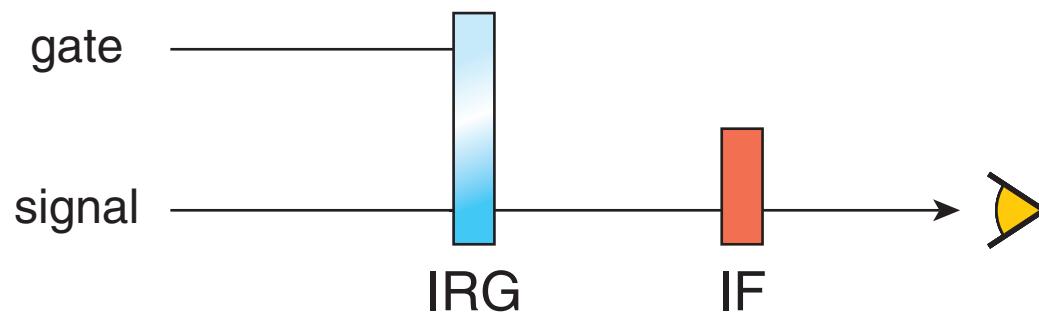
Experiment 2



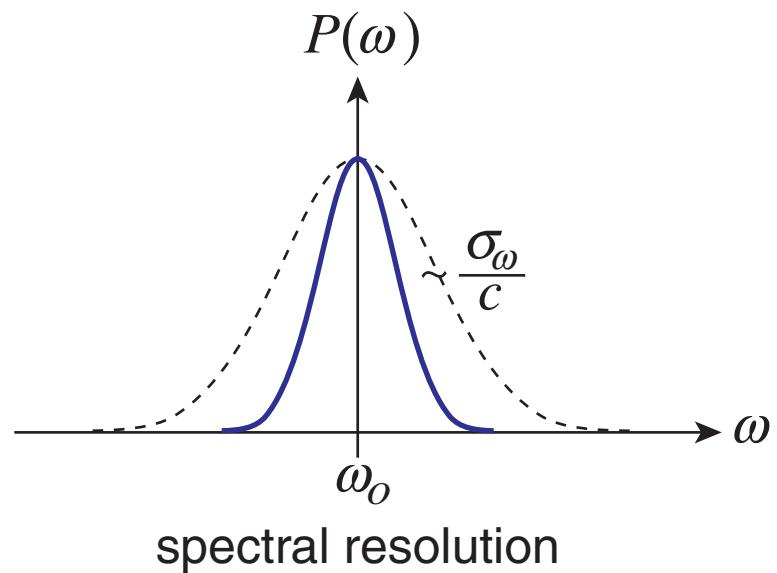
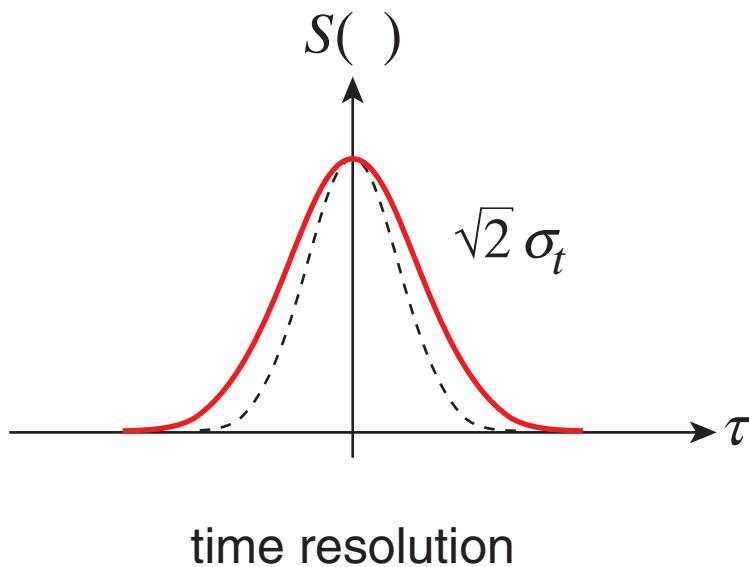
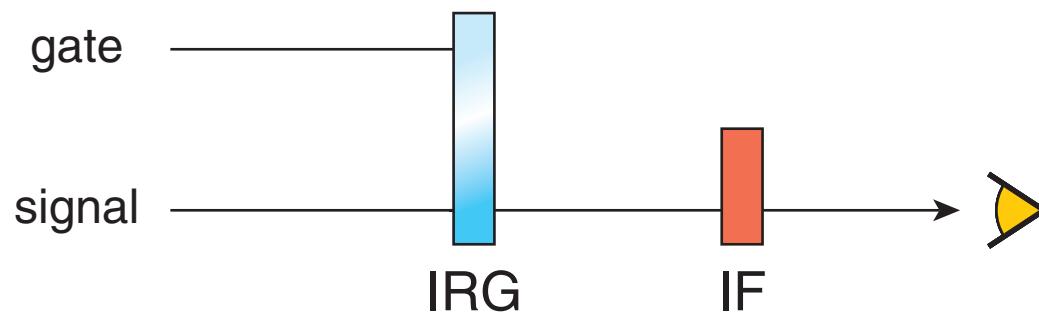
Experiment 2



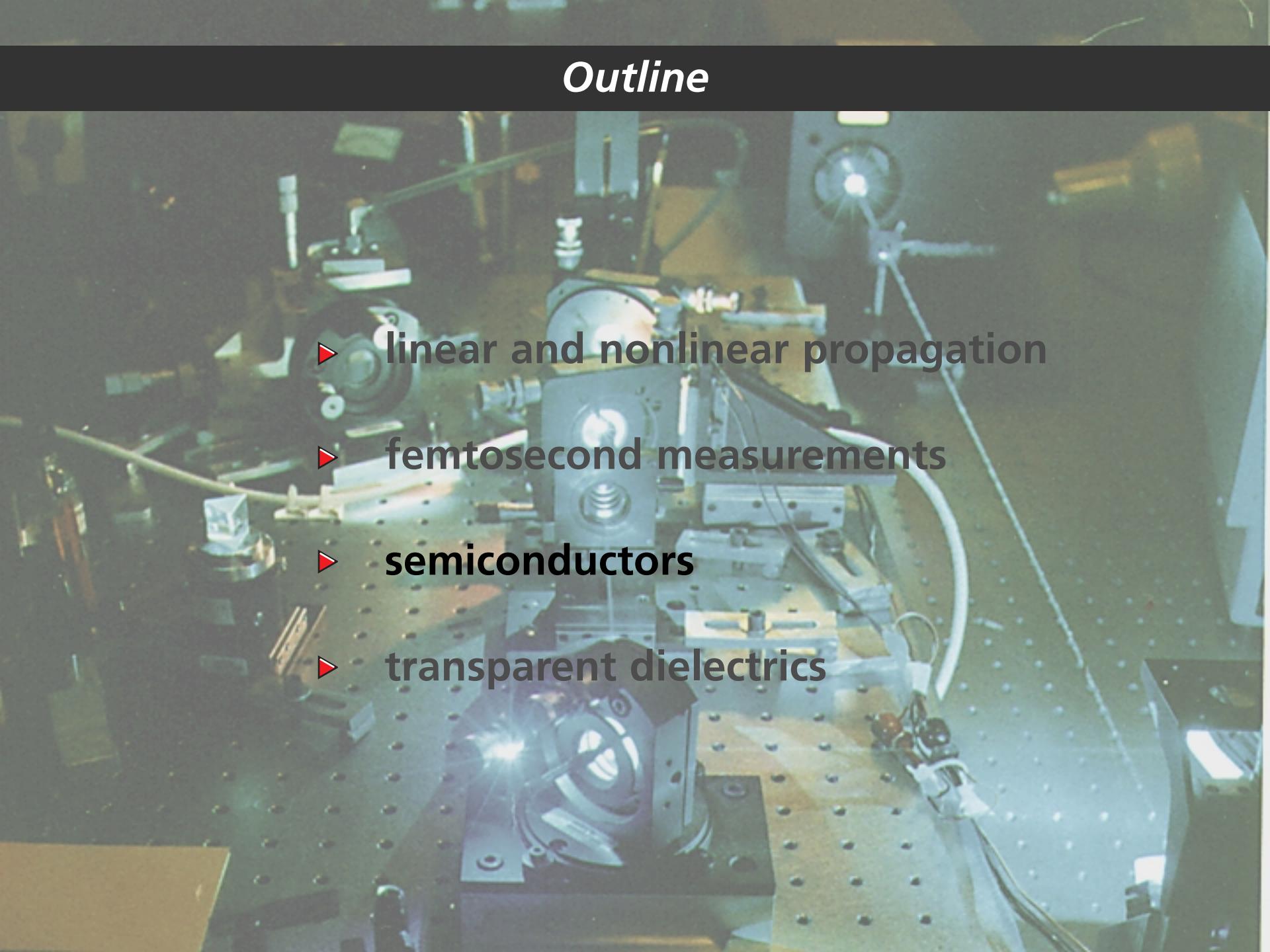
Experiment 2



Experiment 2

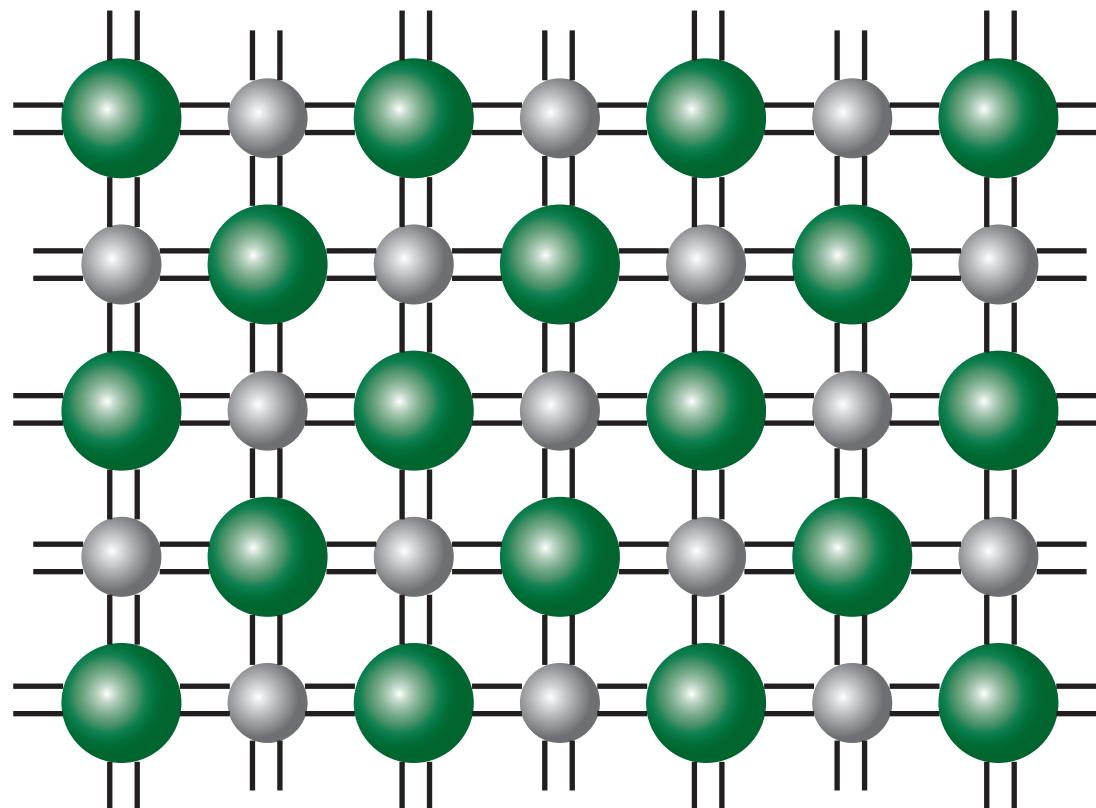


Outline

- 
- ▶ linear and nonlinear propagation
 - ▶ femtosecond measurements
 - ▶ semiconductors
 - ▶ transparent dielectrics

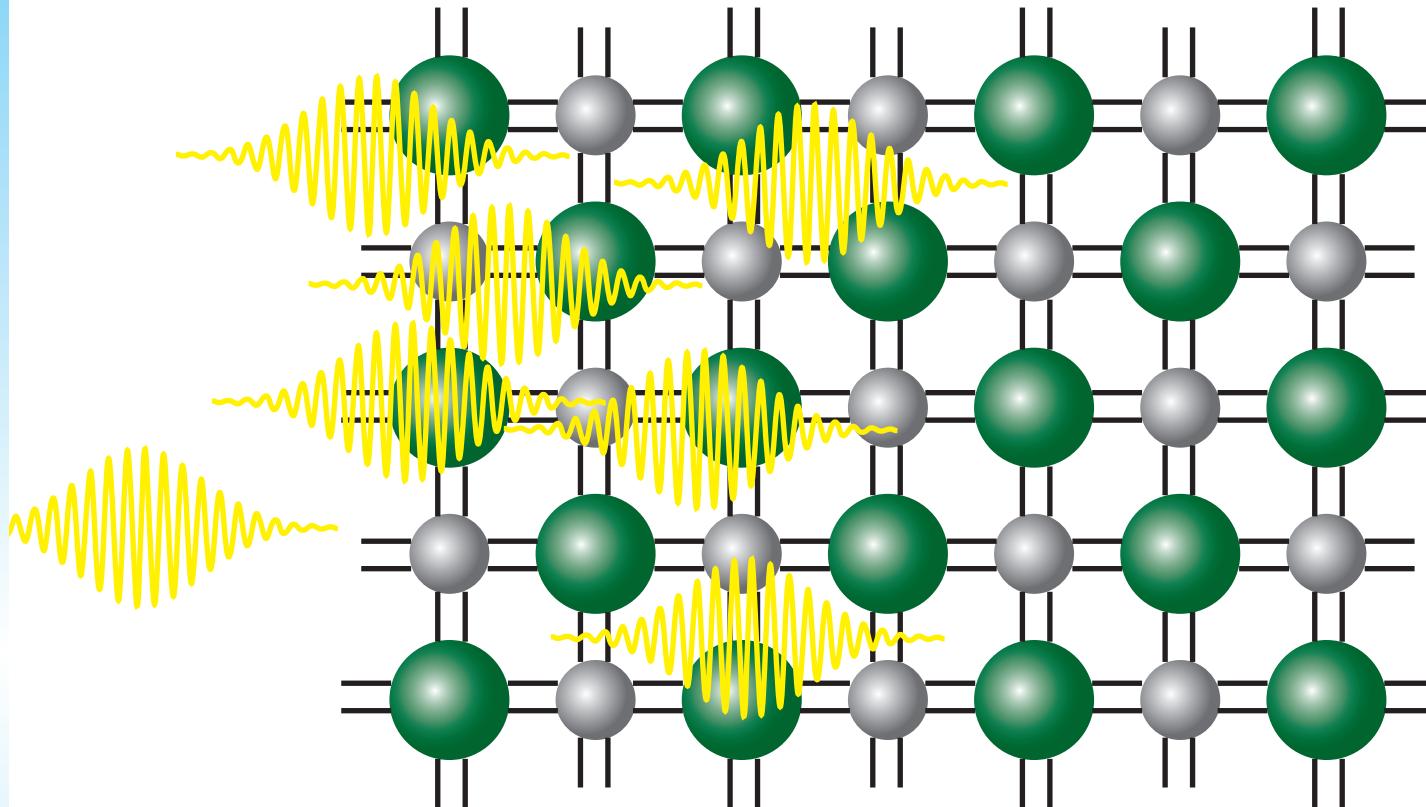
Introduction

how do femtosecond laser pulses alter a solid?



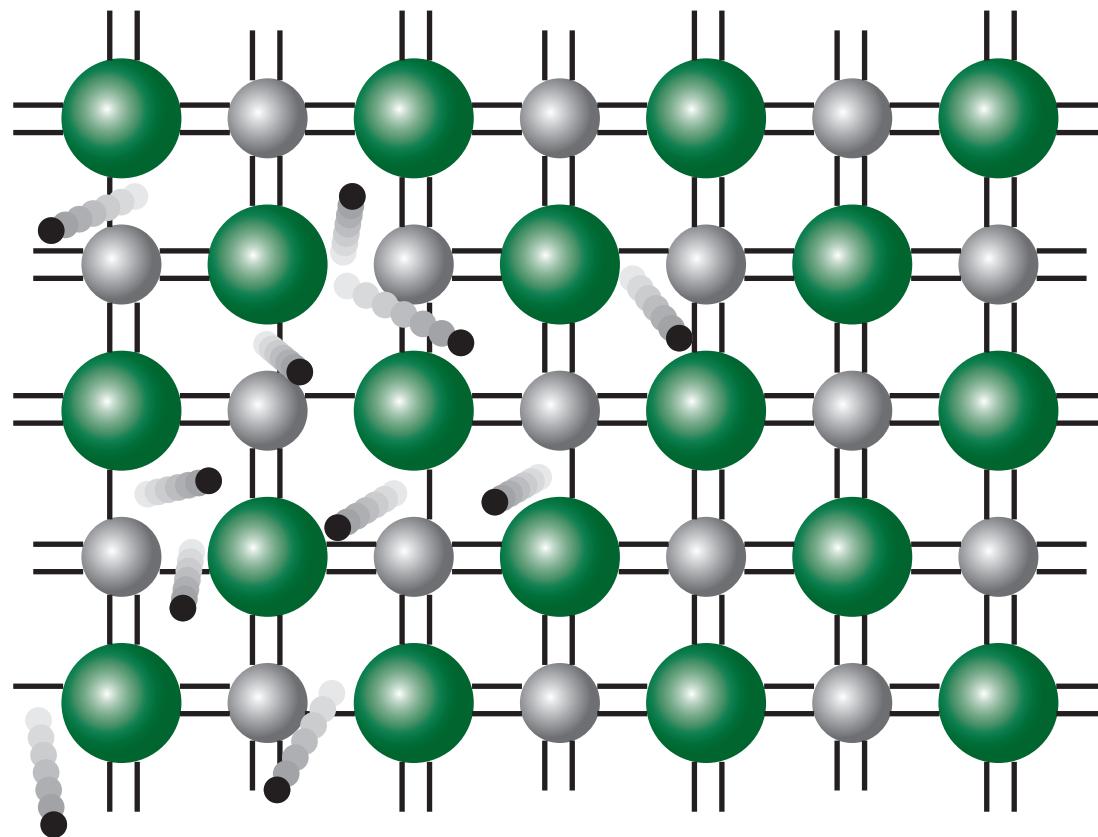
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photons excite valence electrons...



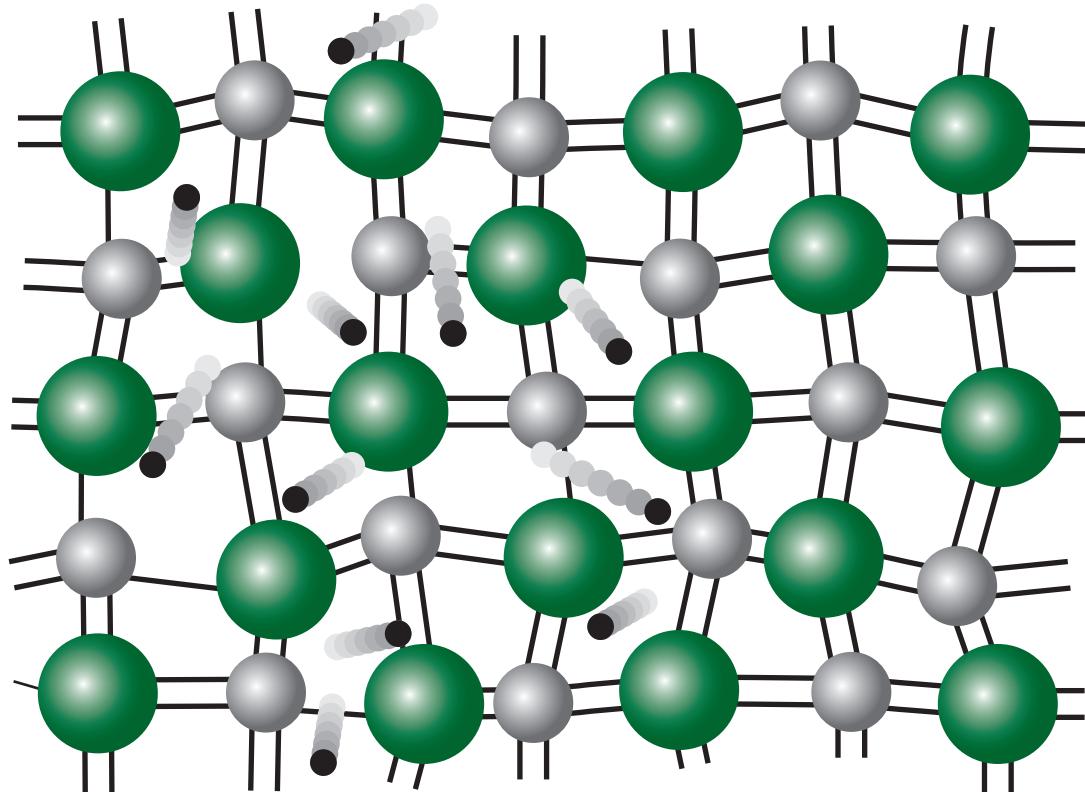
Introduction

...and create free electrons...



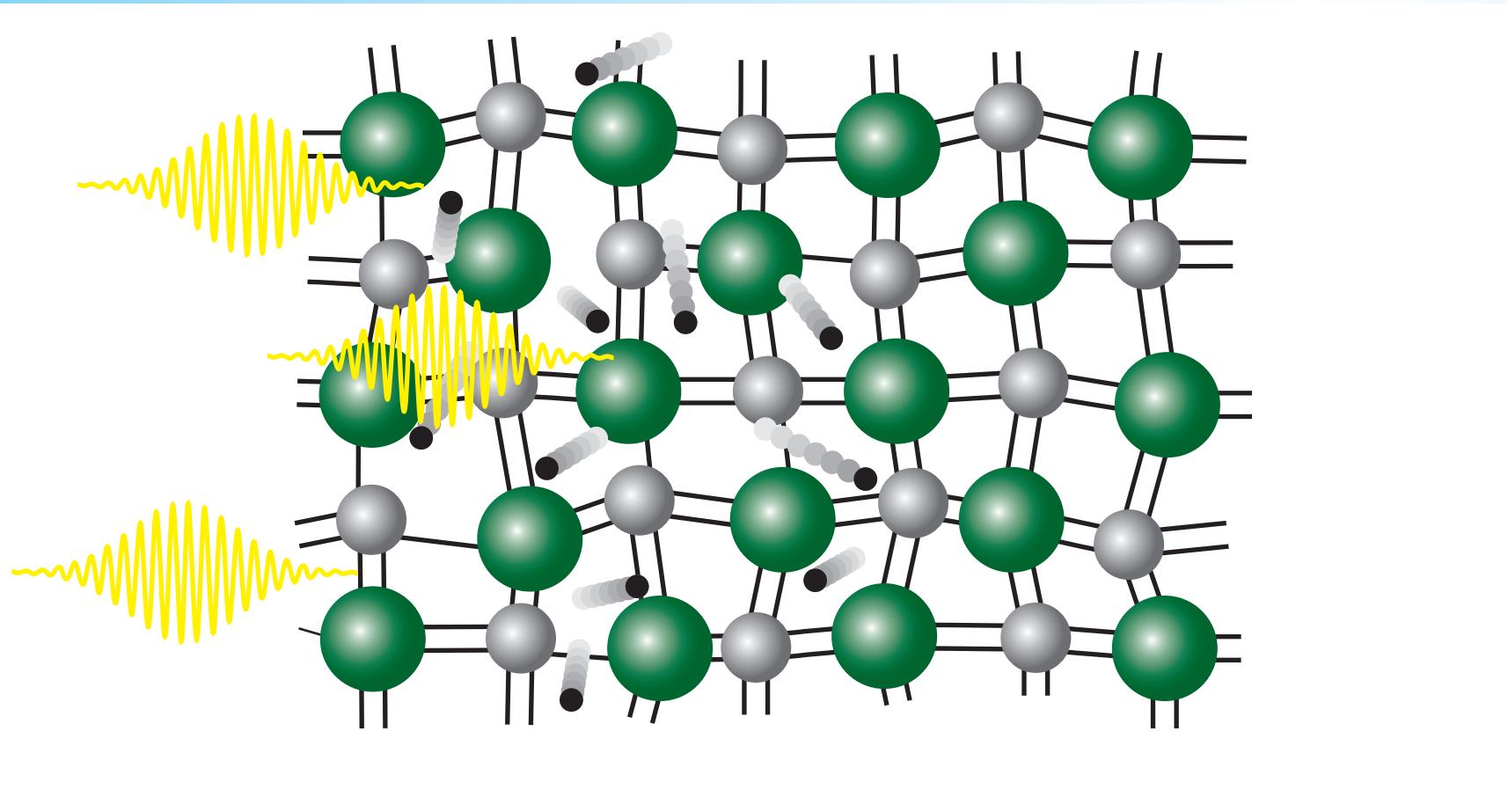
Introduction

...causing electronic and structural changes...



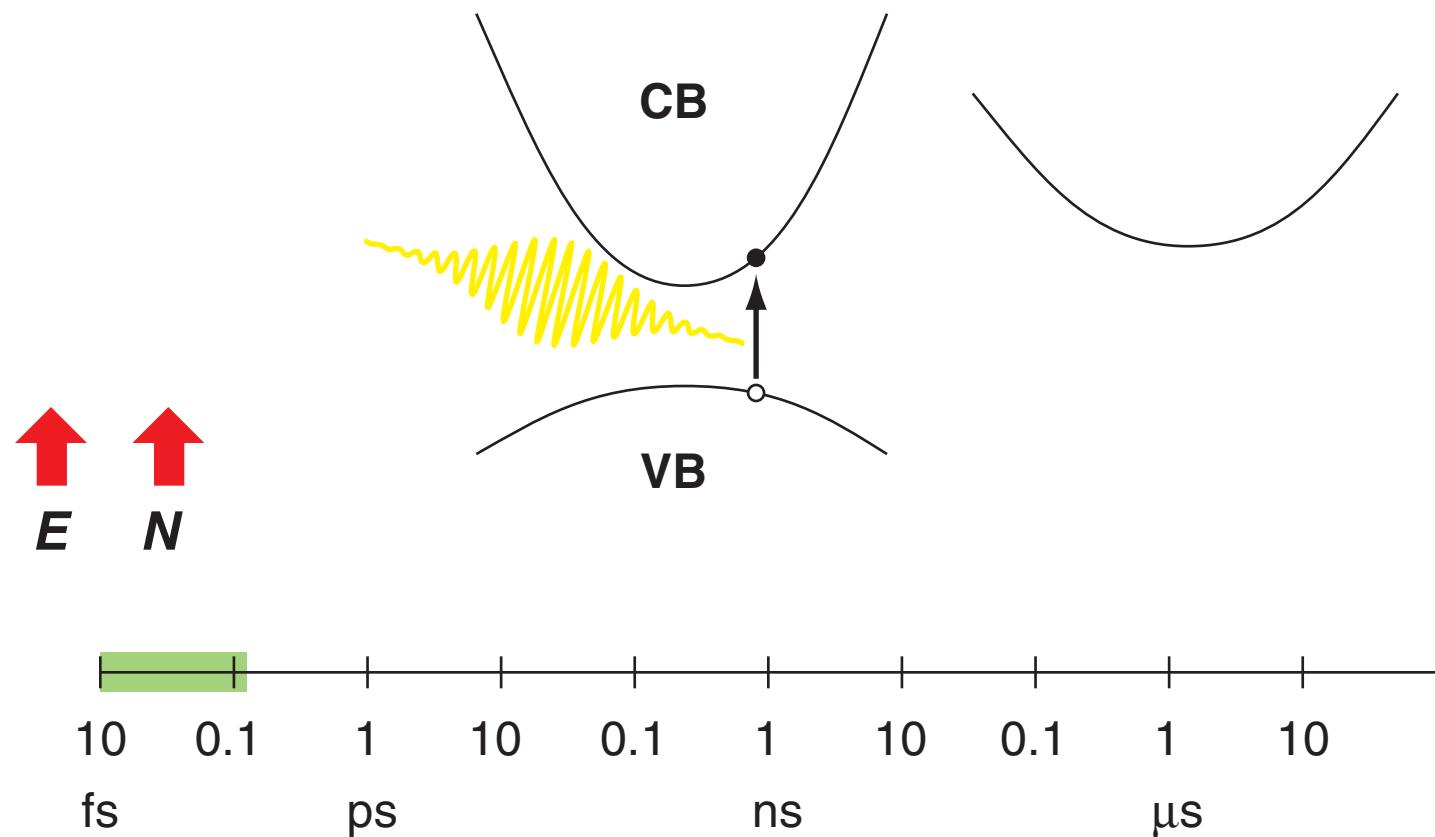
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...which we measure with another pulse



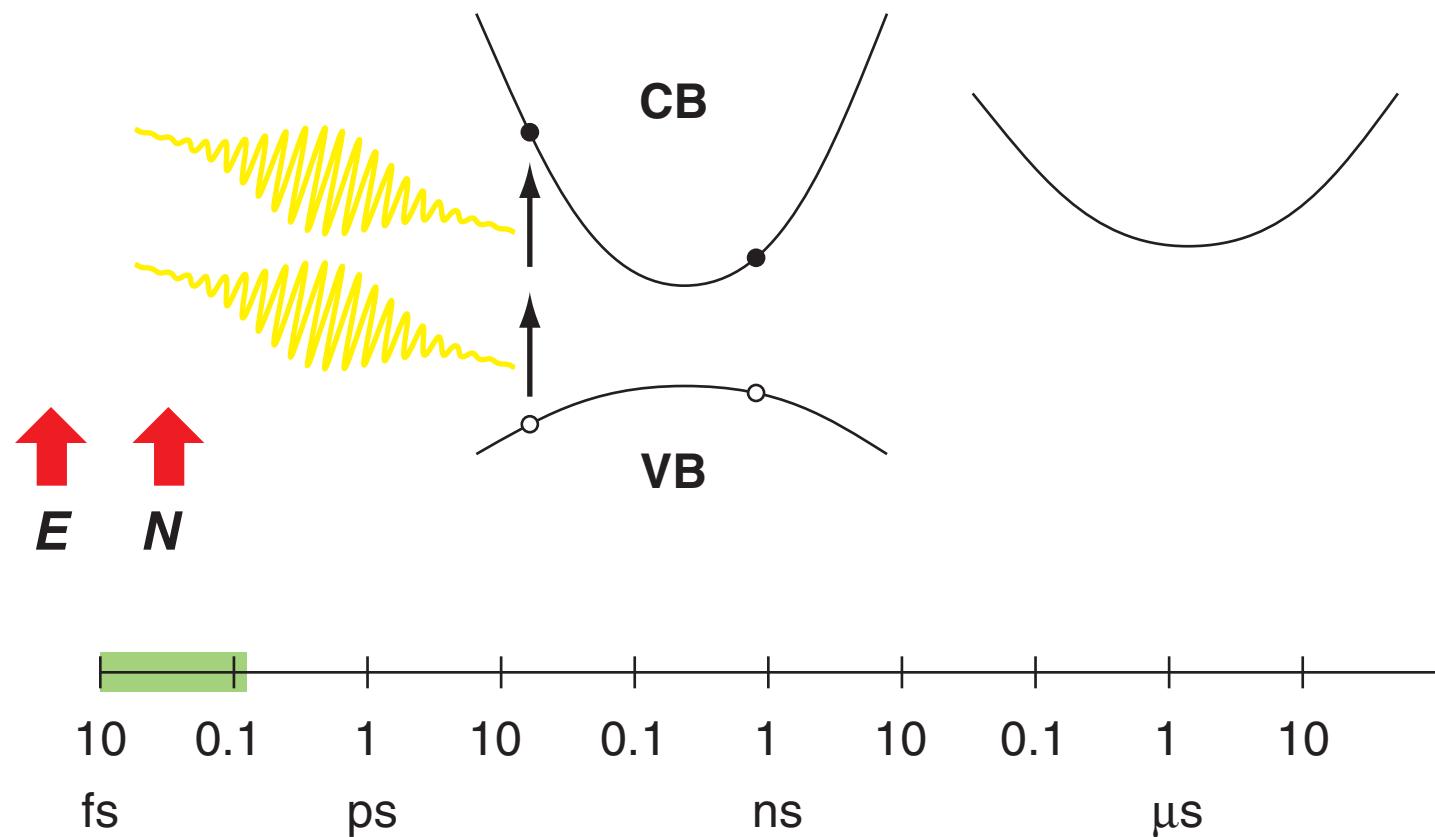
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single photon excitation

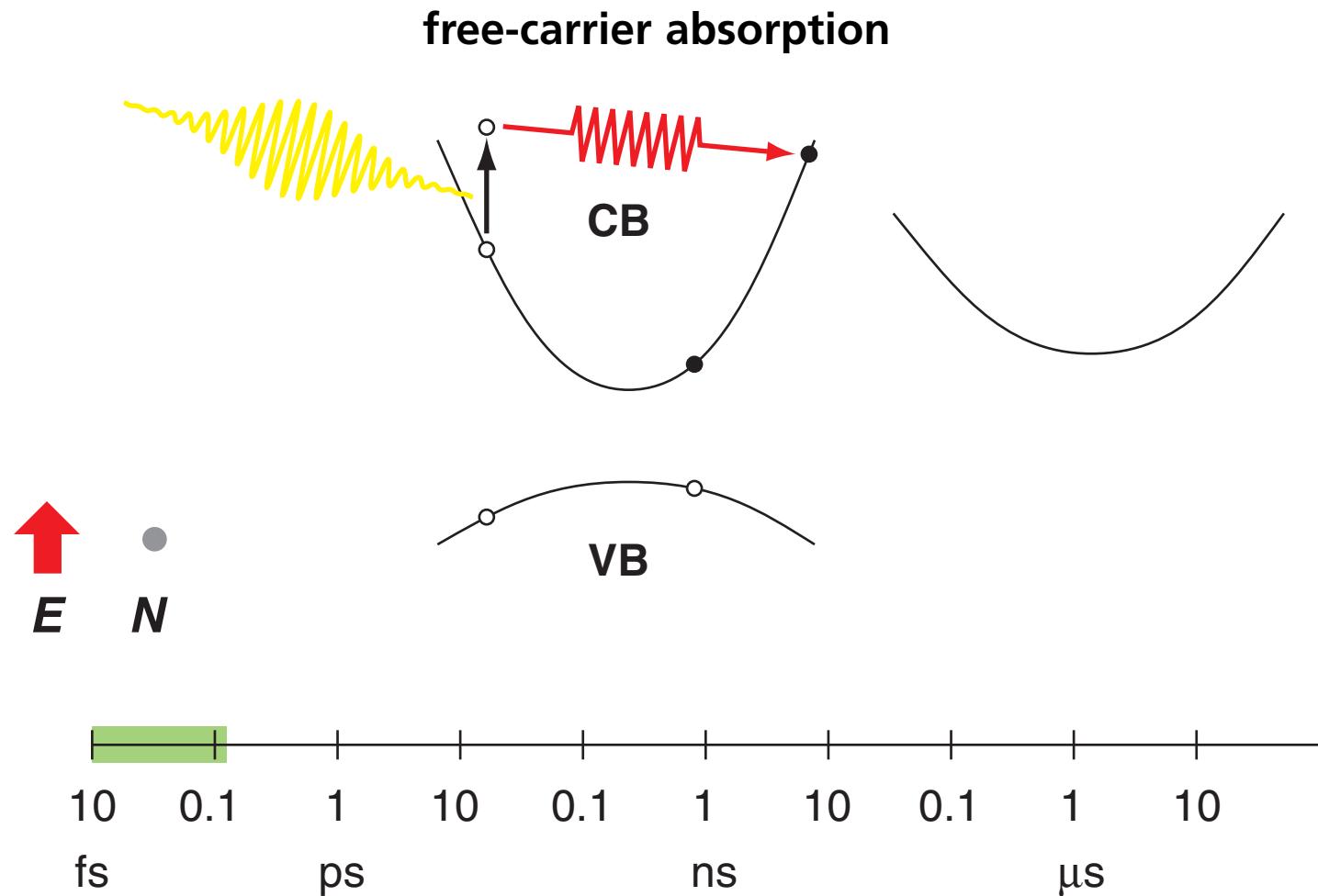


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two-photon excitation

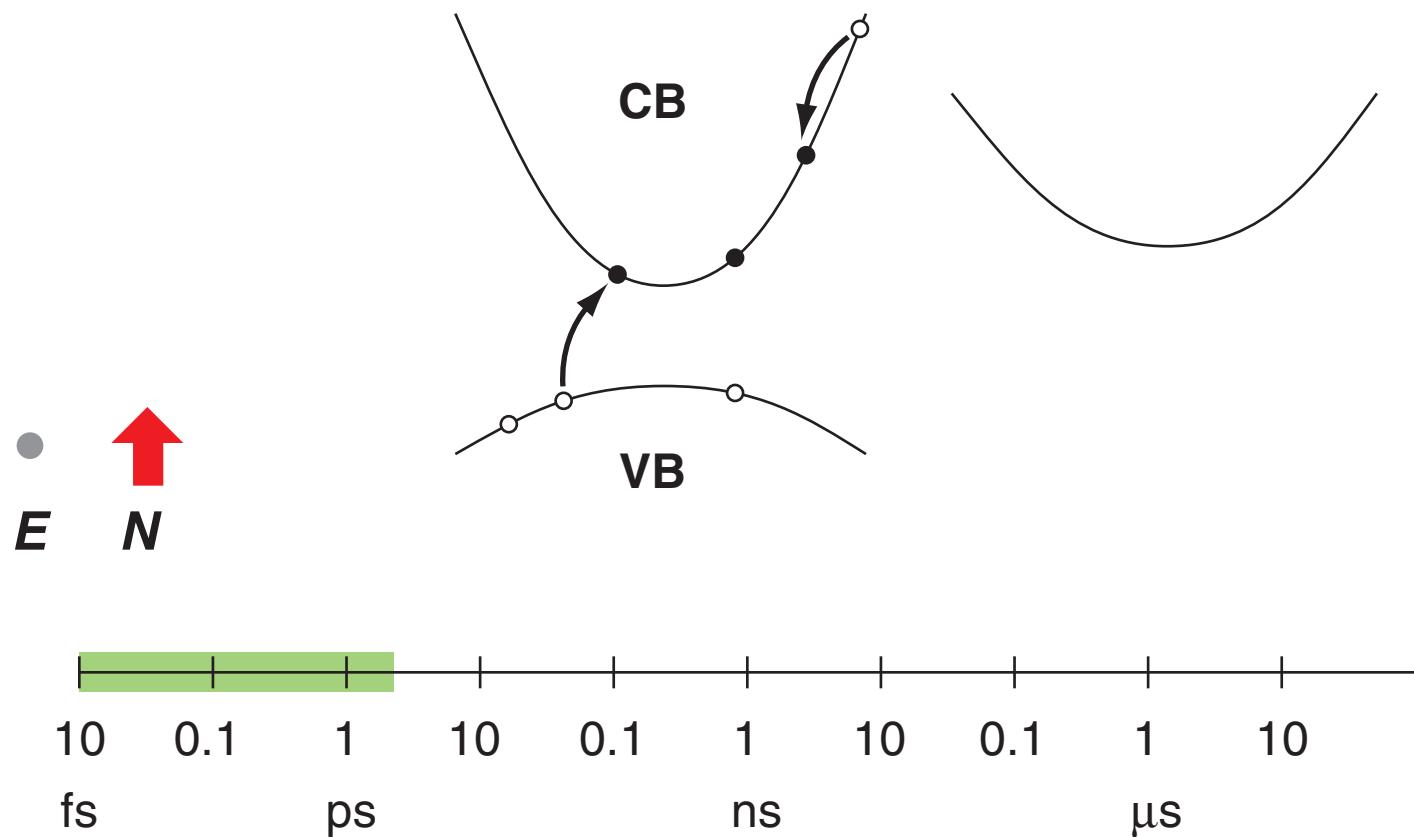


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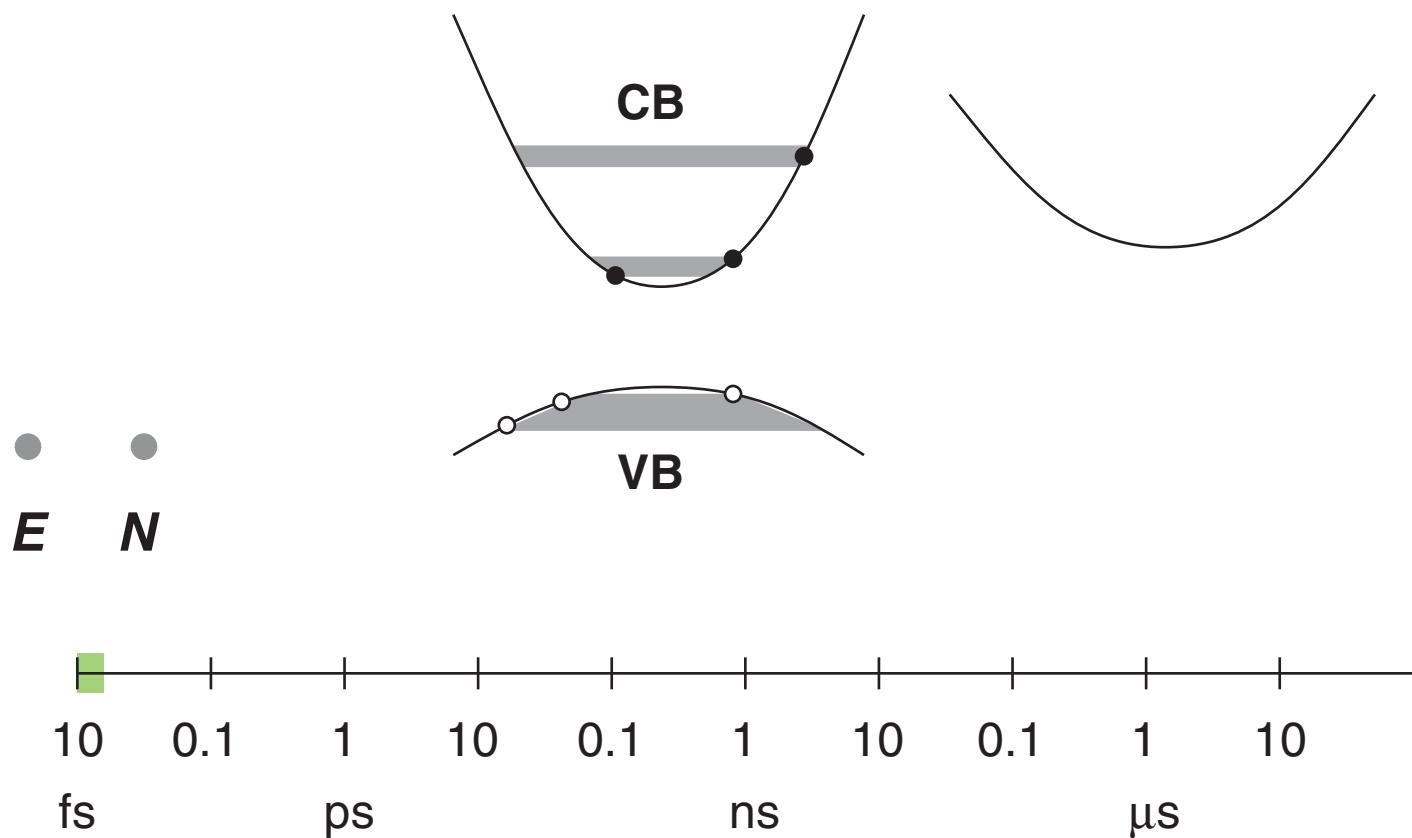
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impact ionization



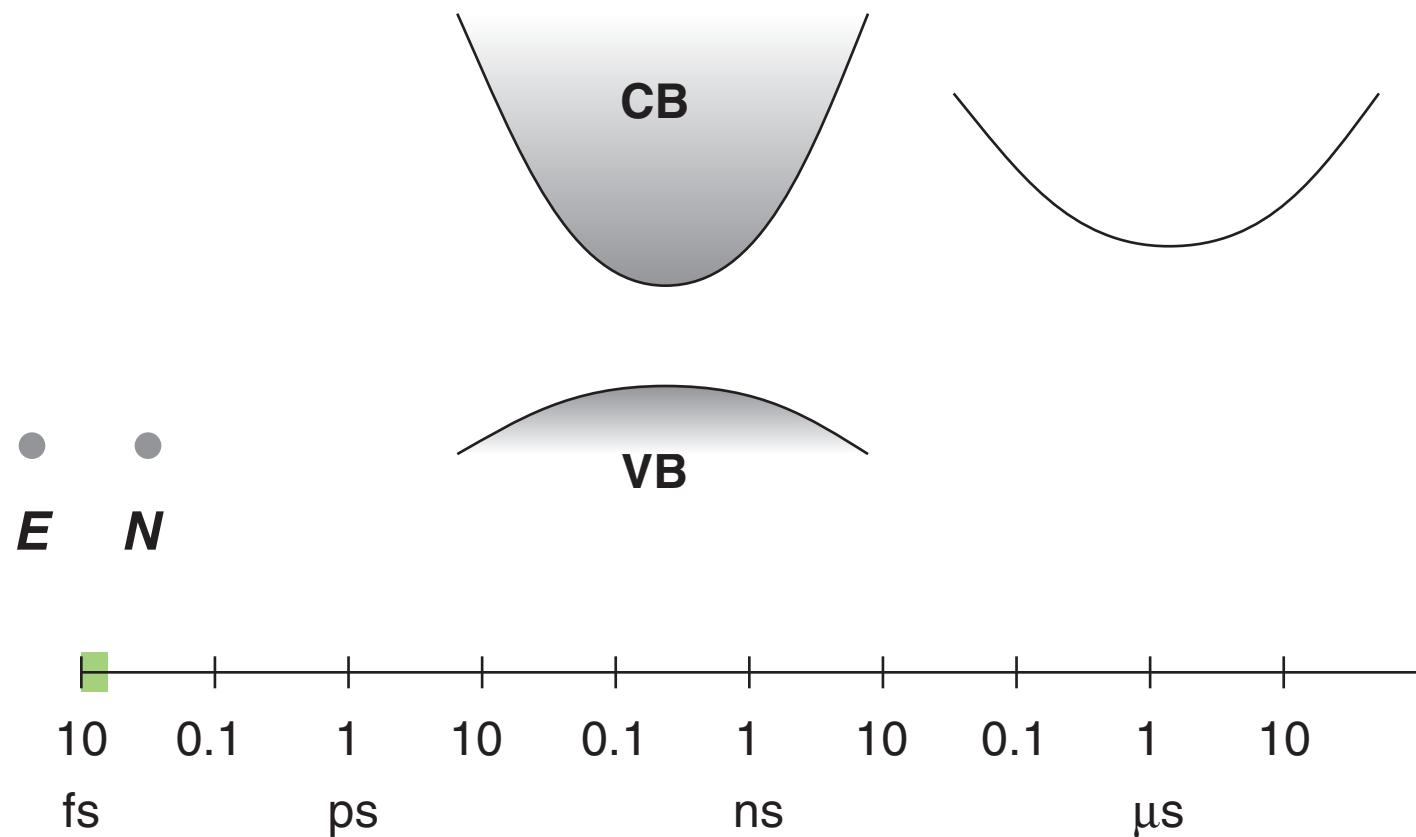
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carrier-carrier scattering



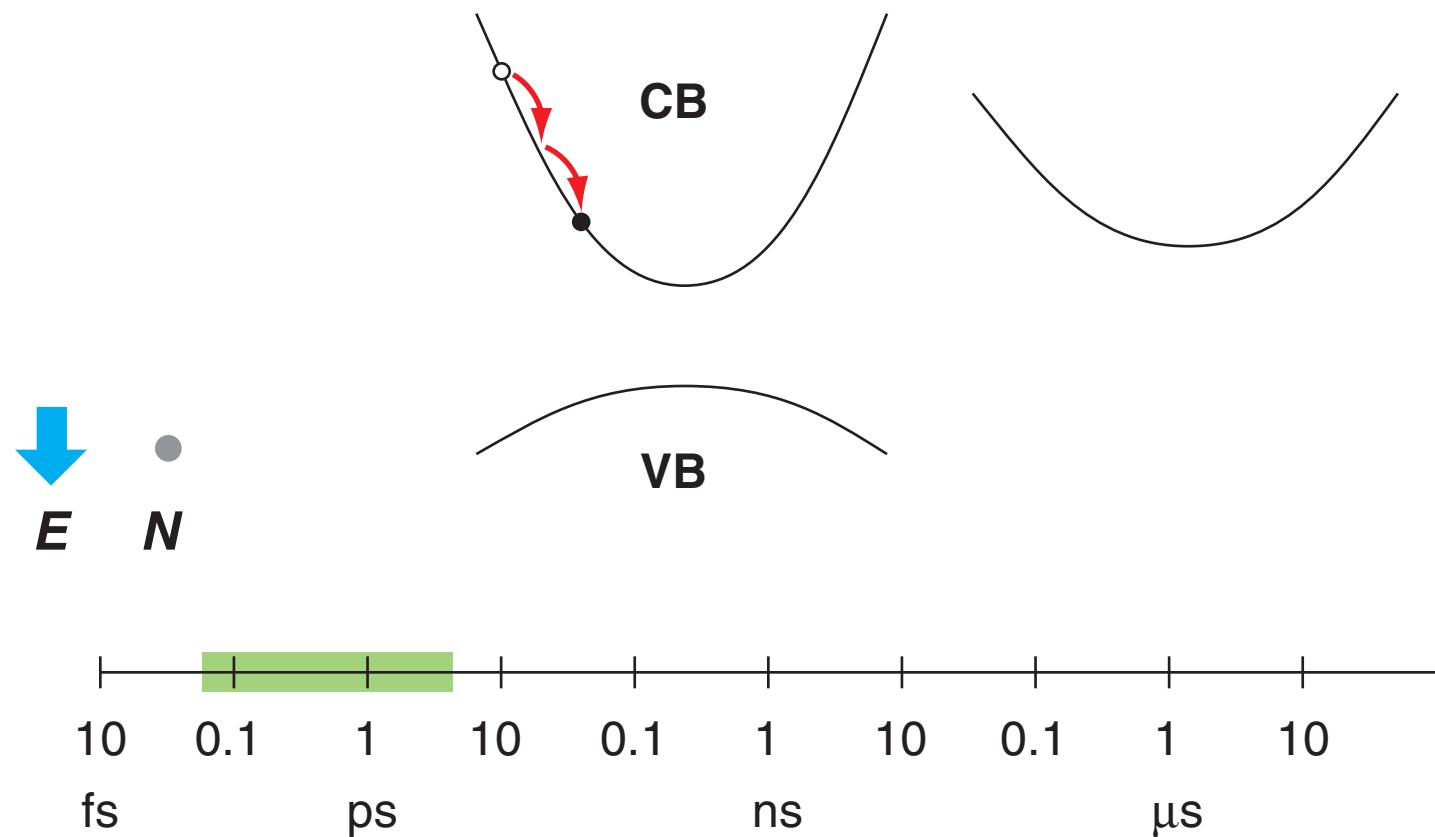
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carrier-carrier scattering



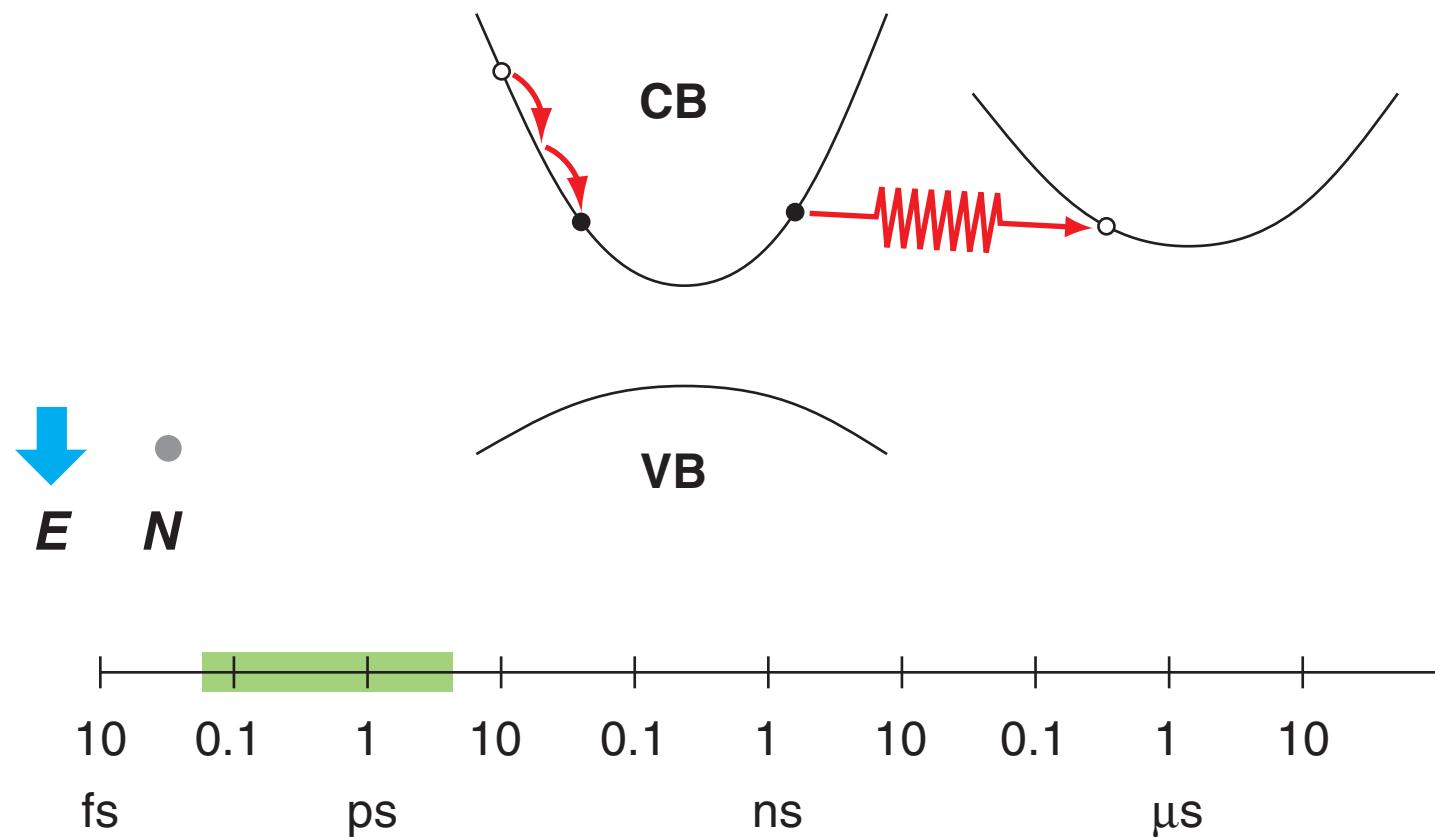
Introduction

carrier-phonon scattering



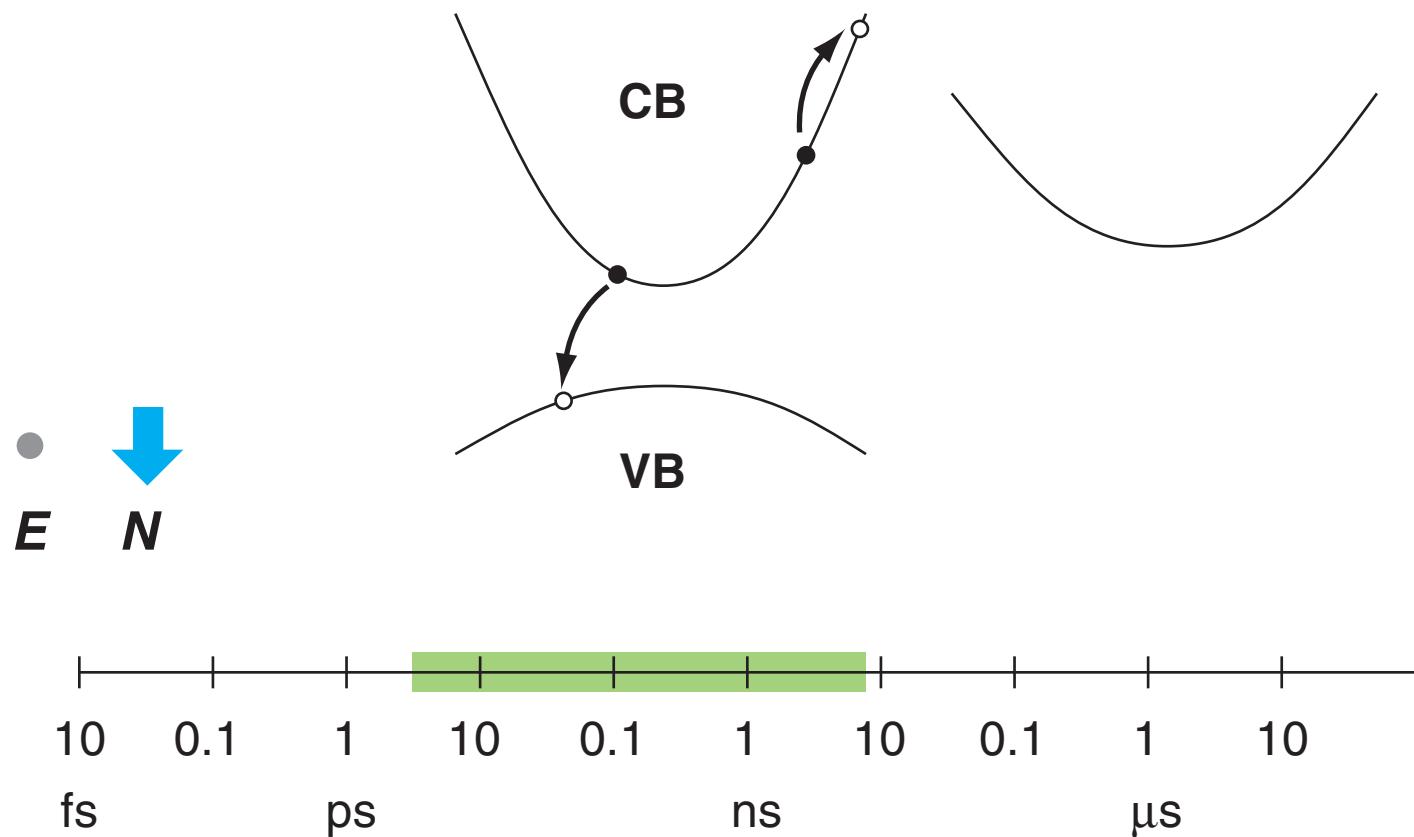
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carrier-phonon scattering



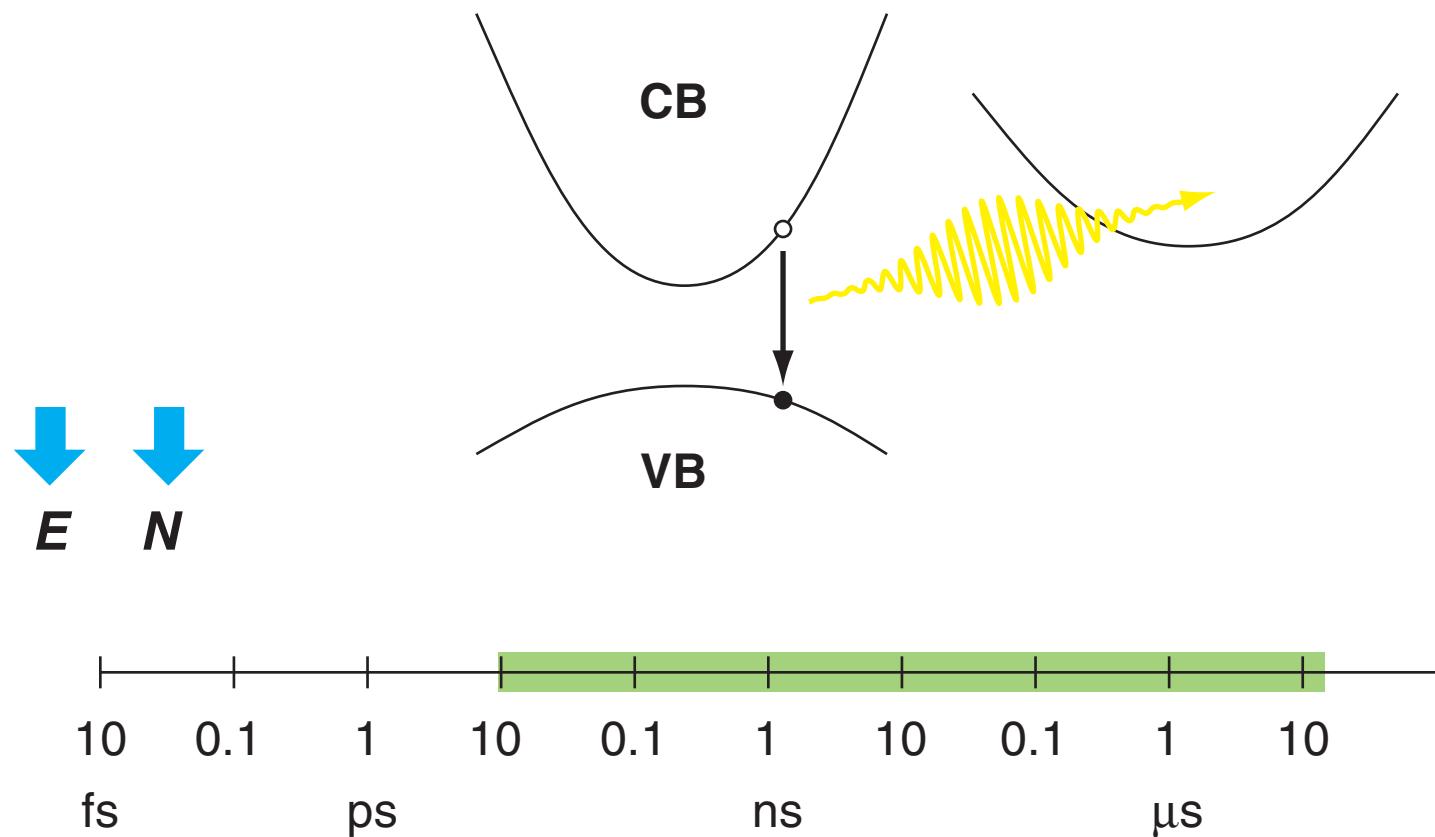
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Auger recombination



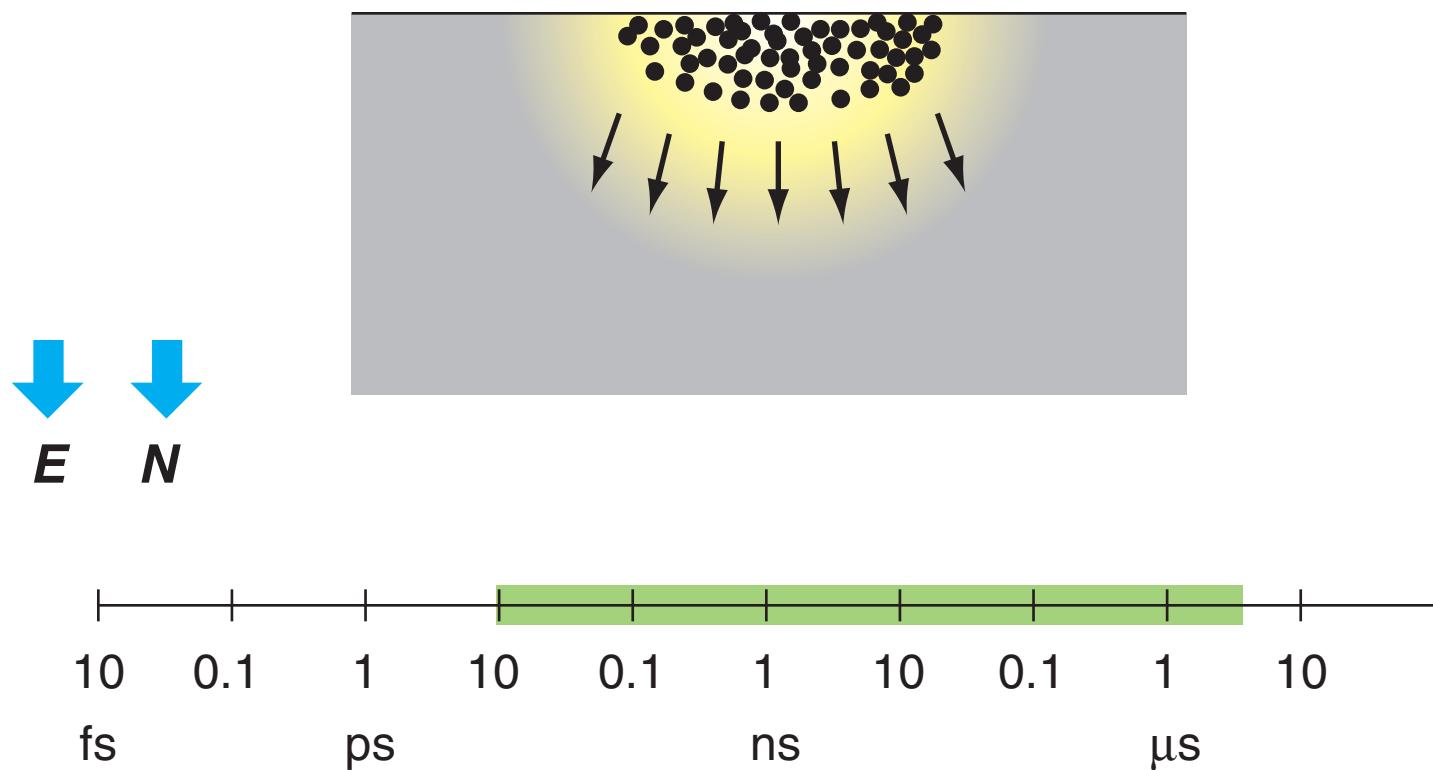
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radiative recombination



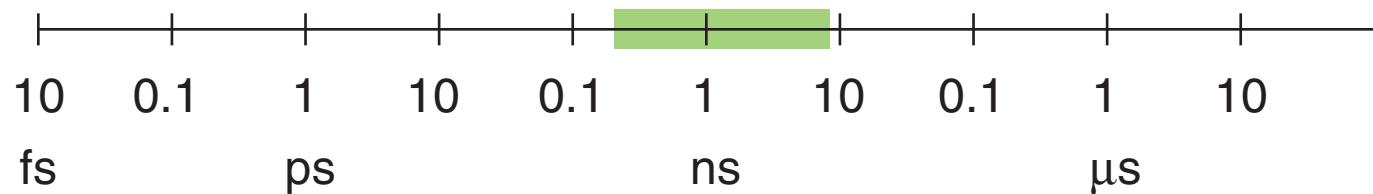
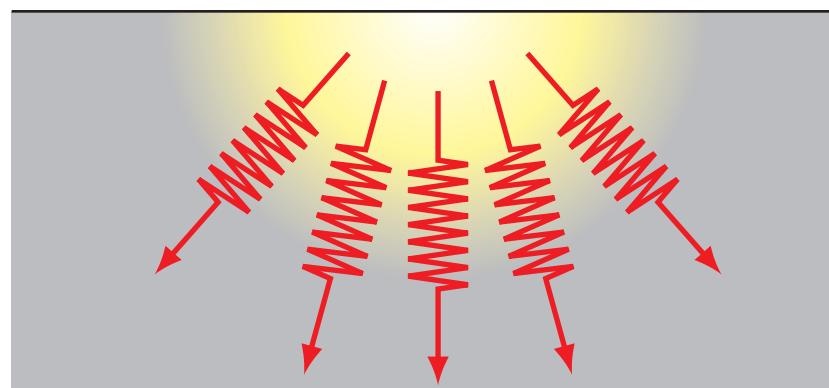
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carrier diffusion



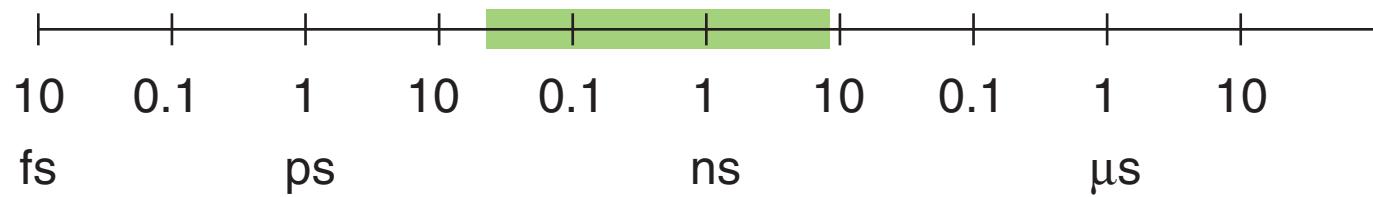
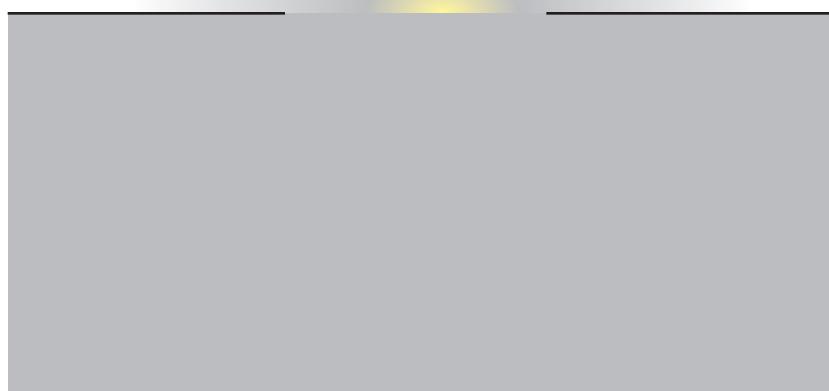
Introduction

thermal diffusion



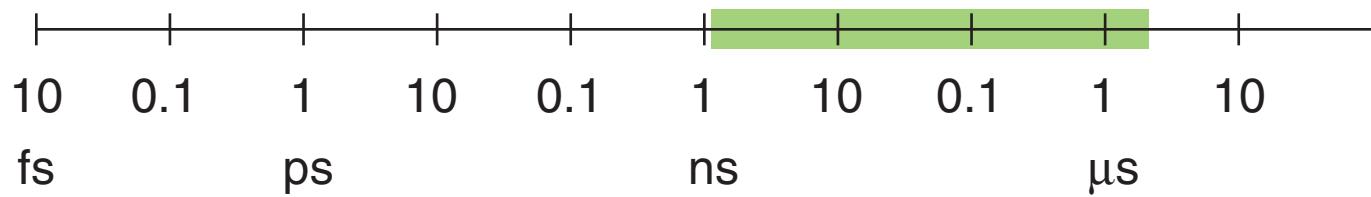
Introduction

ablation and evaporation

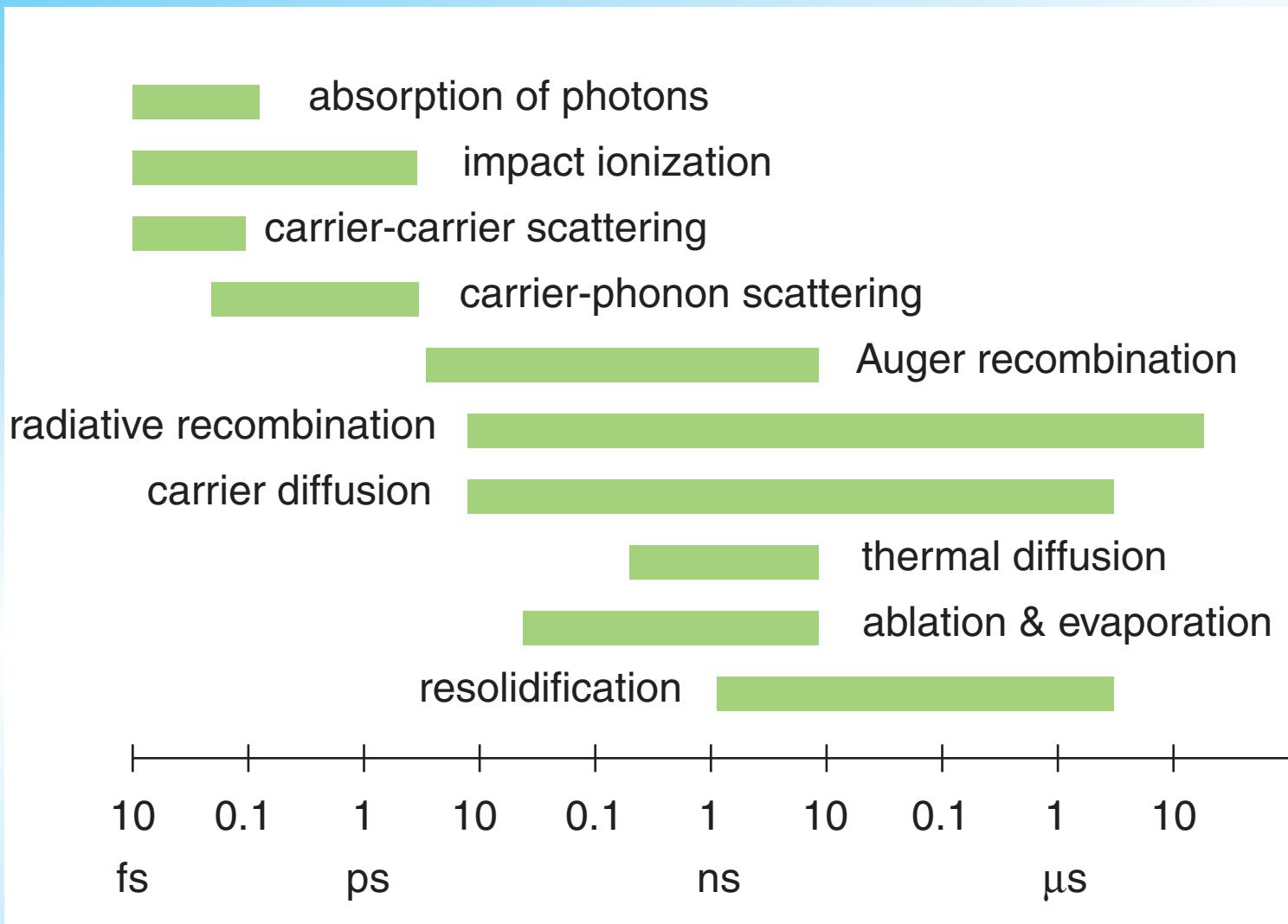


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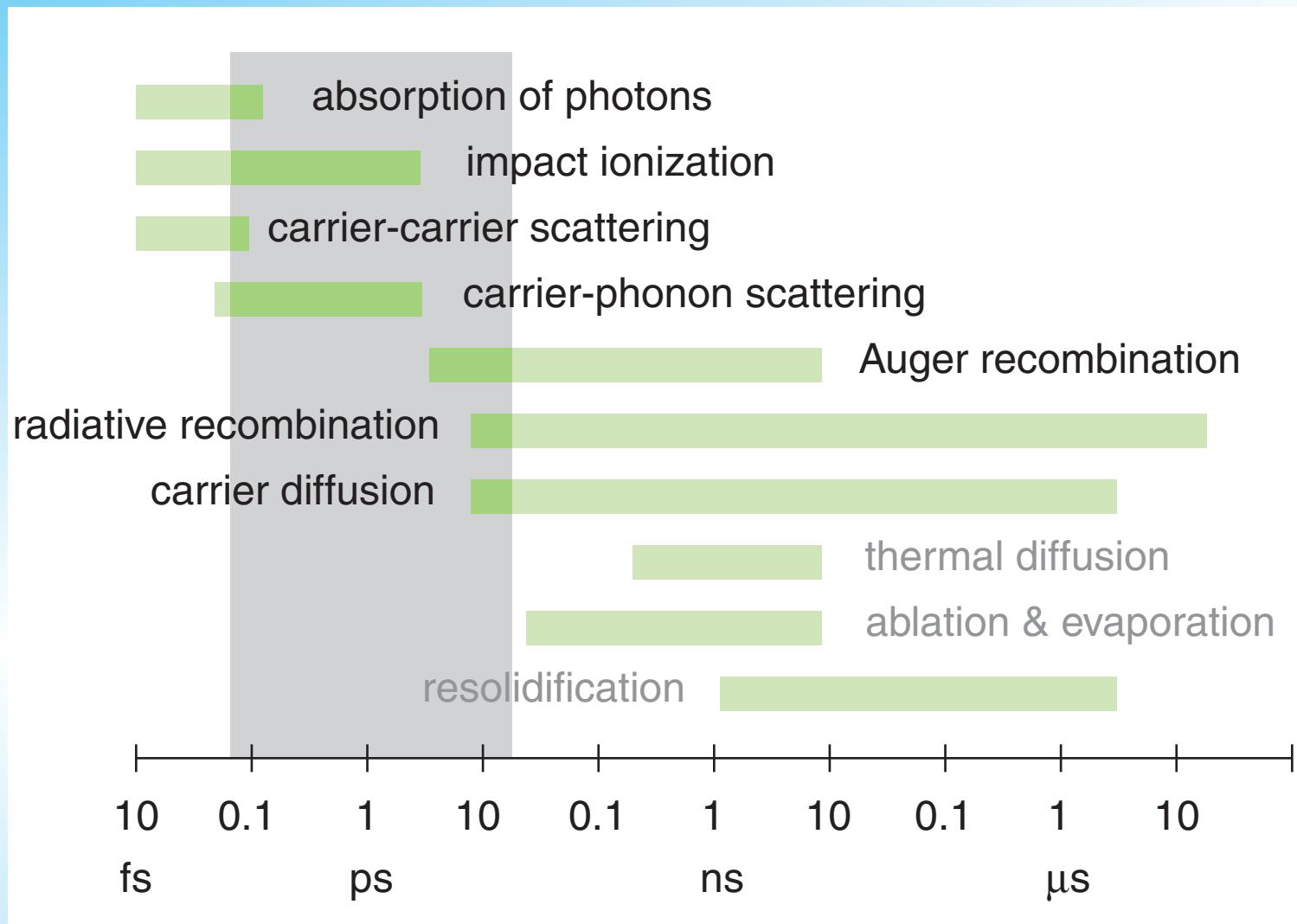
resolidification



Introduction

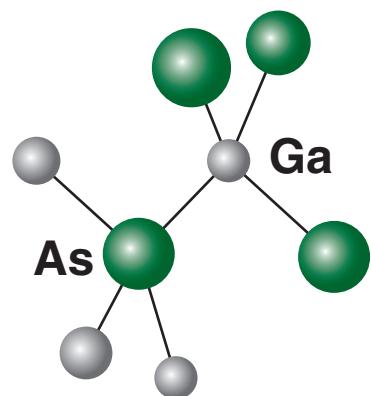


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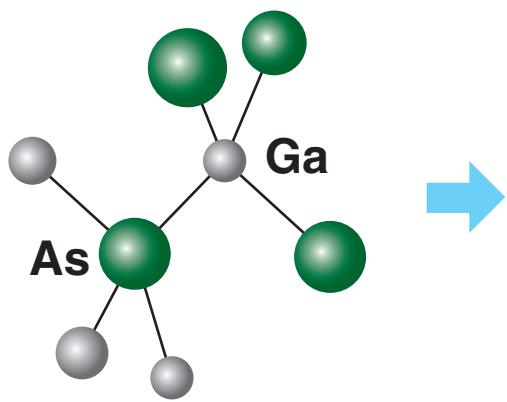
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structure

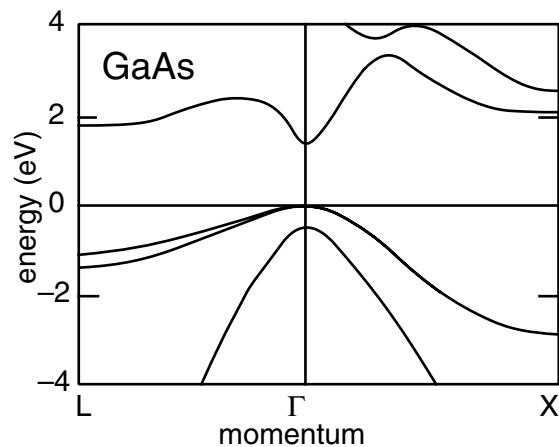


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structure

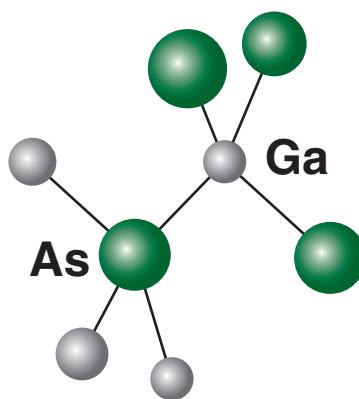


bandstructure

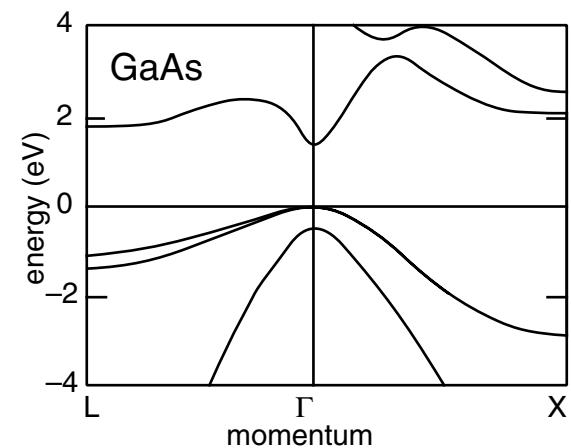


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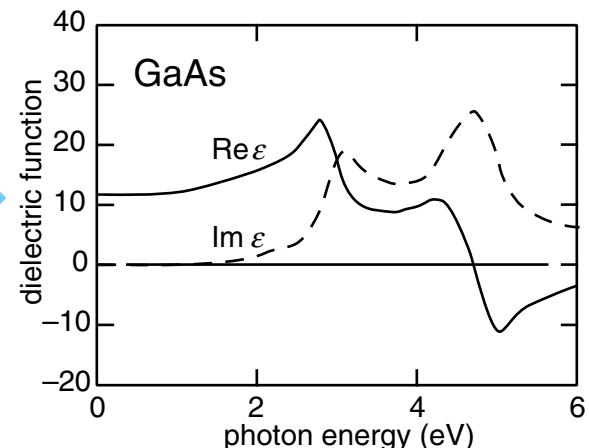
structure



bandstructure

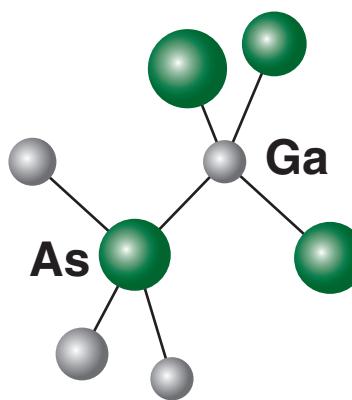


dielectric function

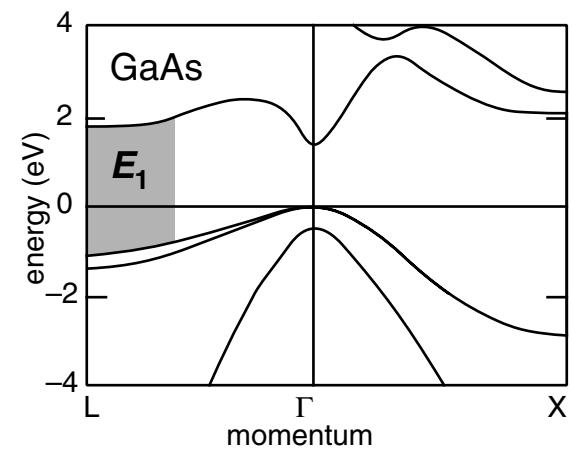


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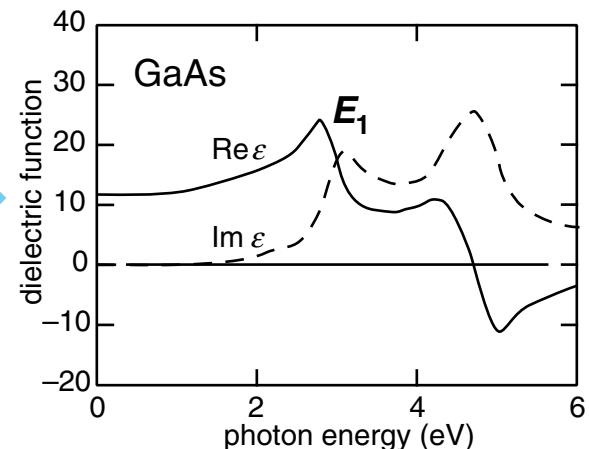
structure



bandstructure

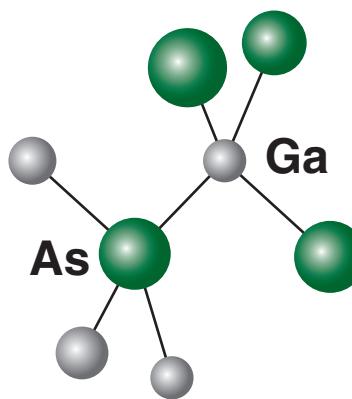


dielectric function

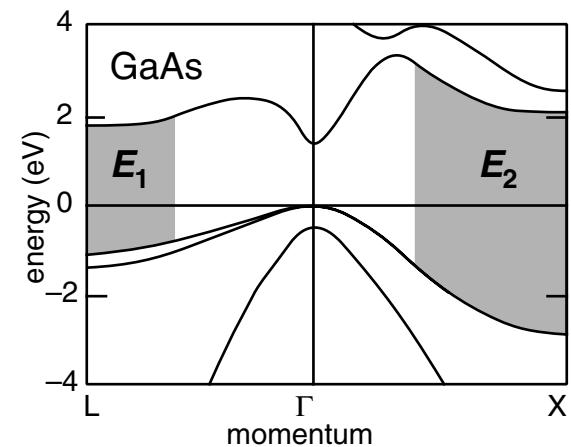


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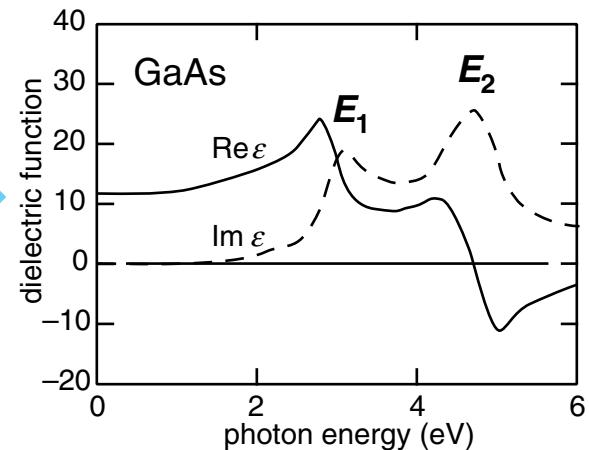
structure



bandstructure

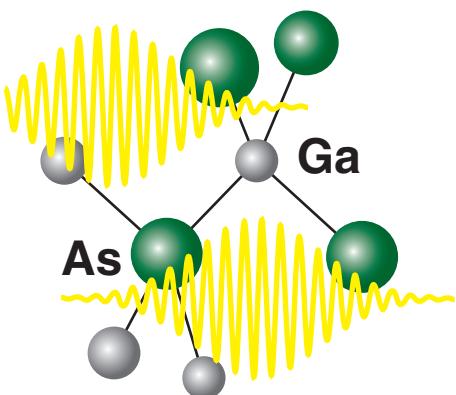


dielectric function



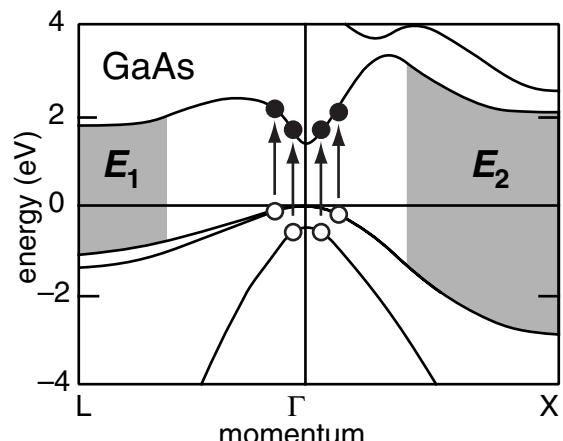
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structure



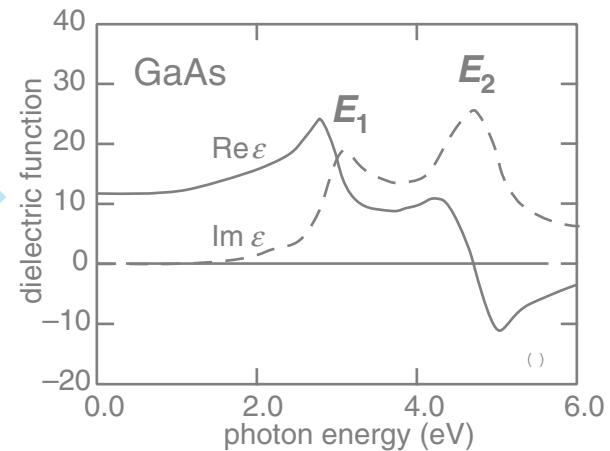
70 fs, 10^{16} W/m^2

bandstructure



$10^{20} - 10^{22} \text{ cm}^{-3}$

dielectric function



Introduction

structure

bandstructure

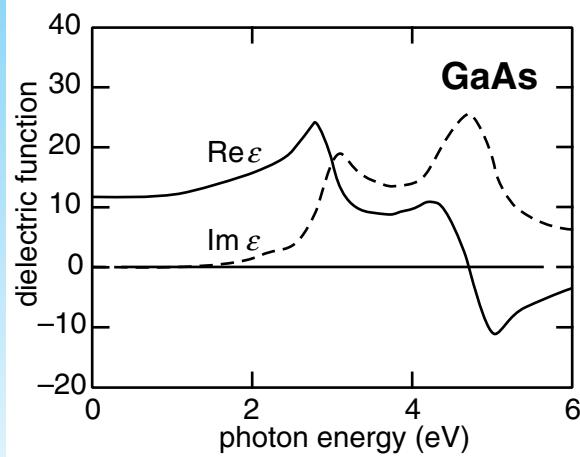
dielectric function

?

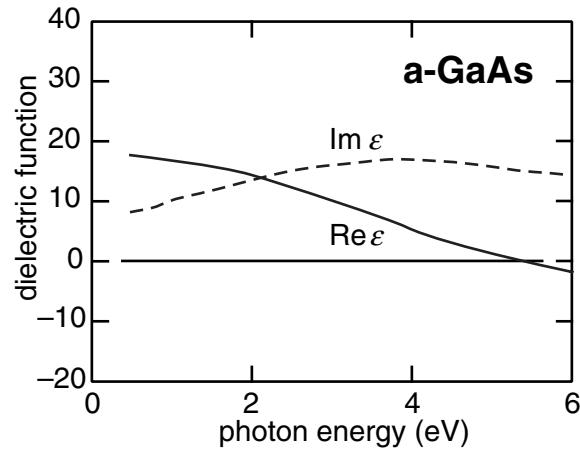
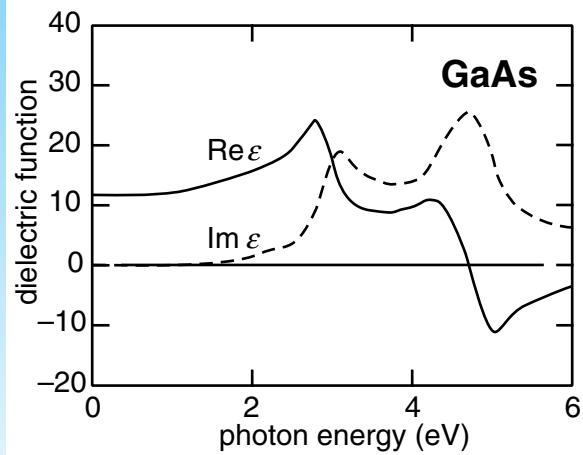
?

?

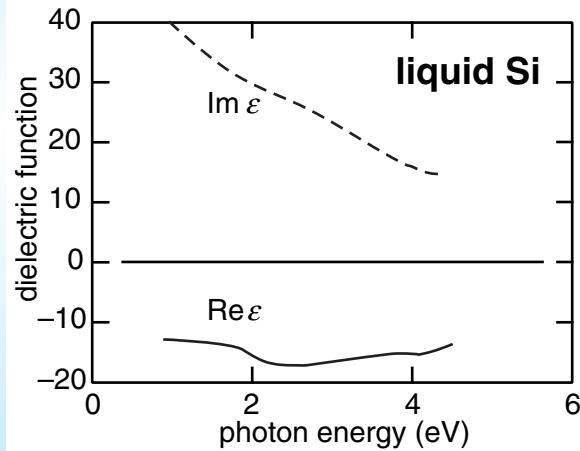
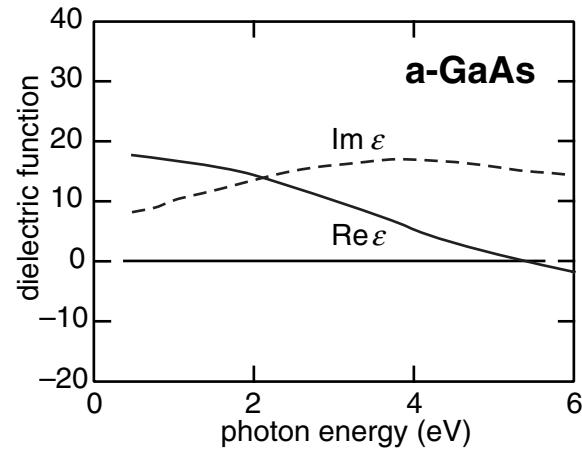
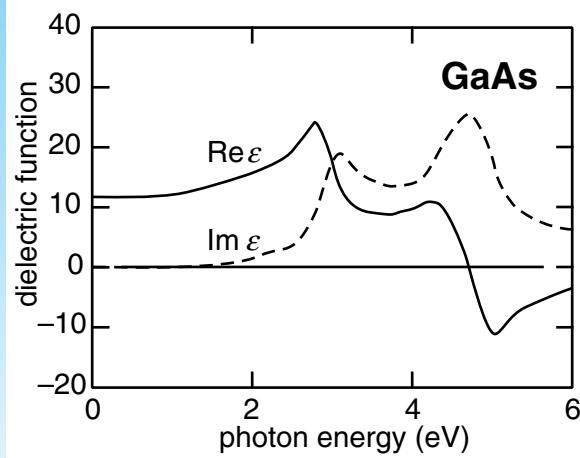
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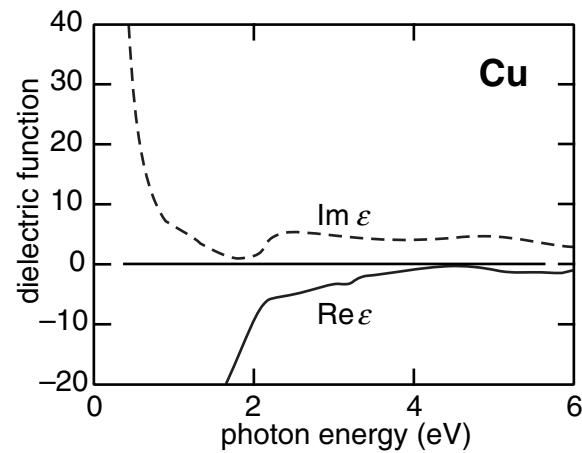
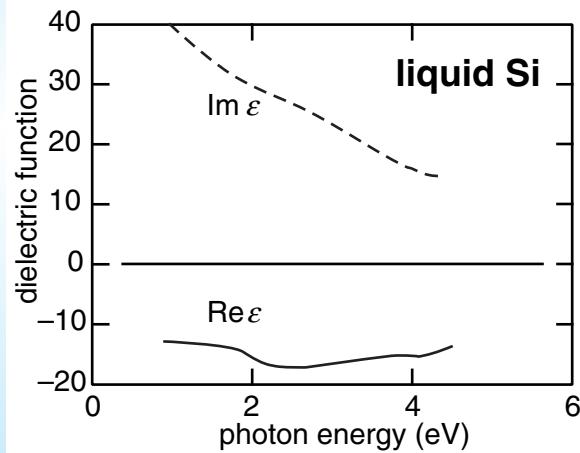
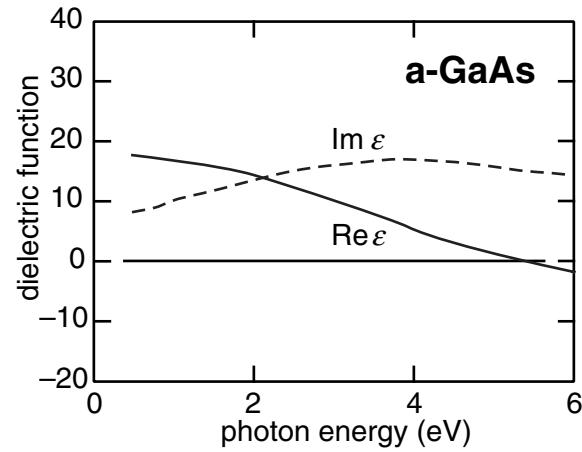
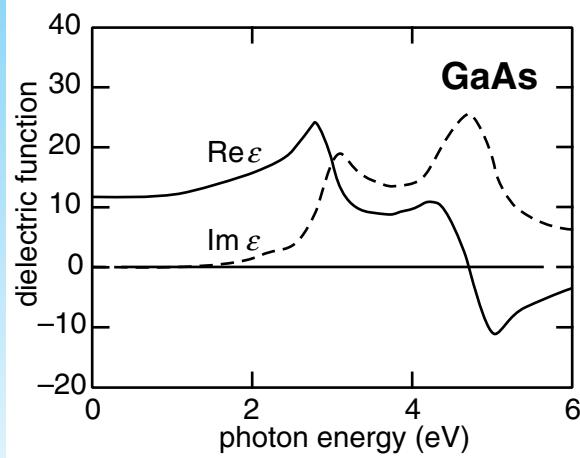
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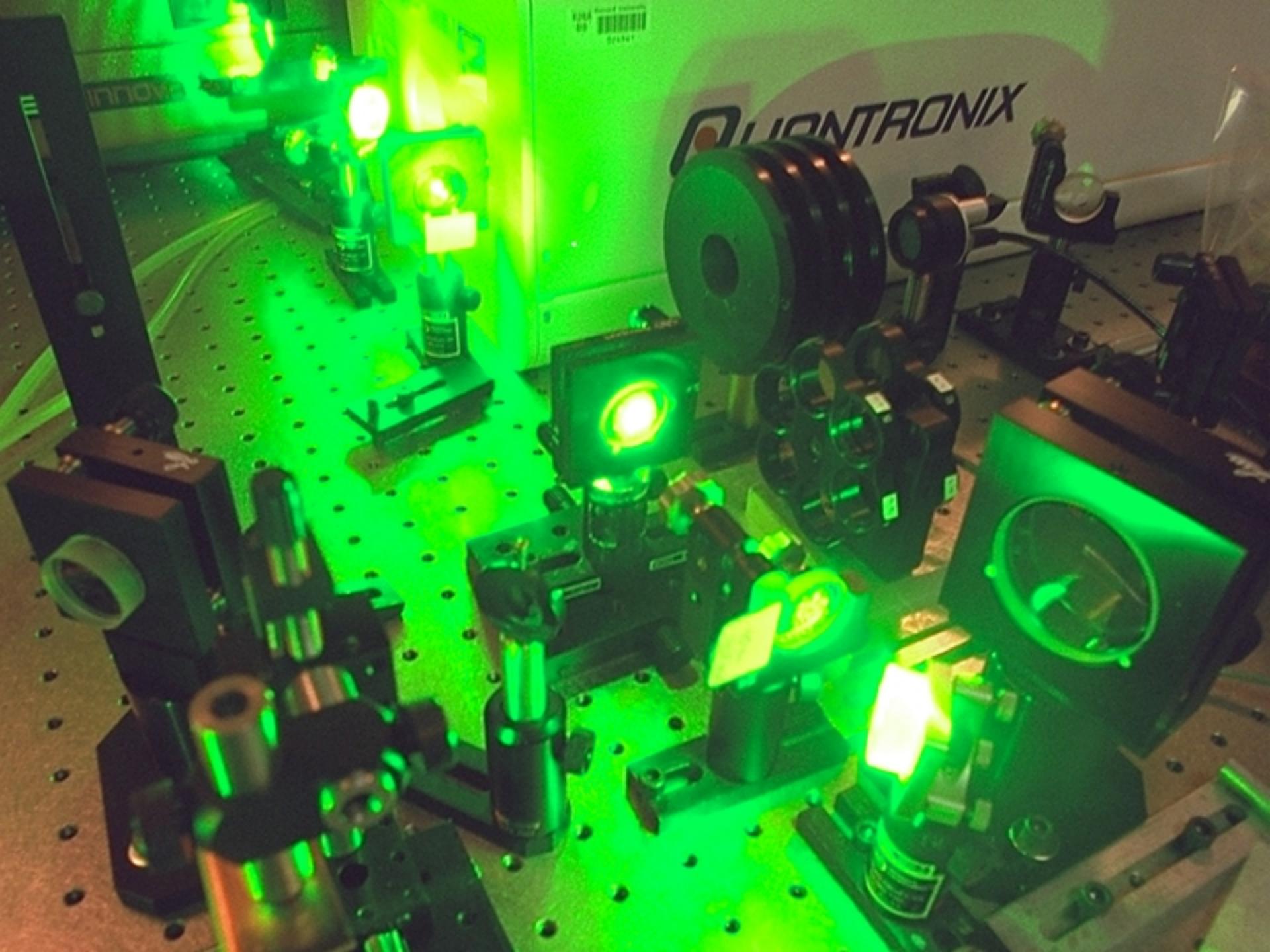


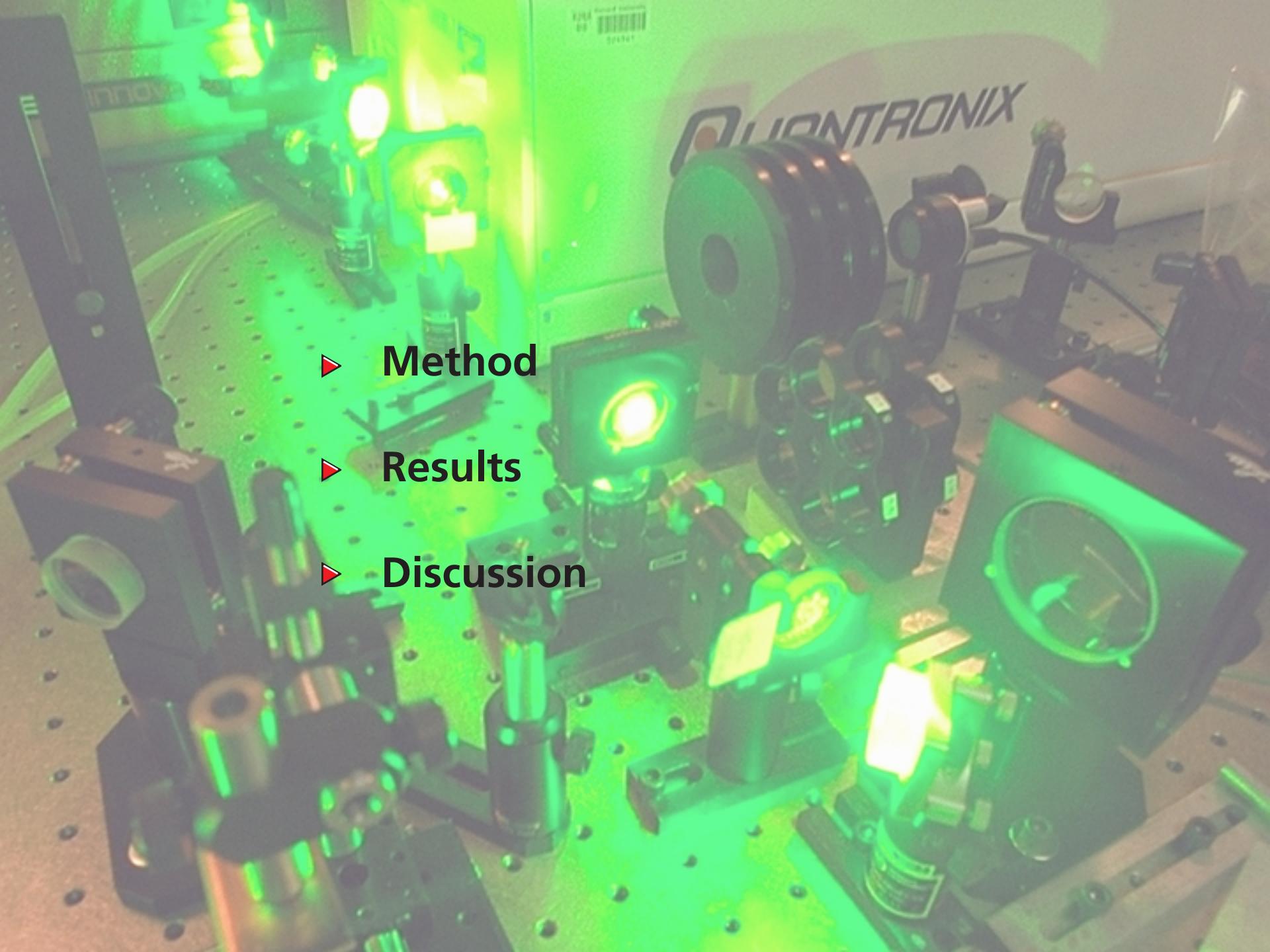
Introduction



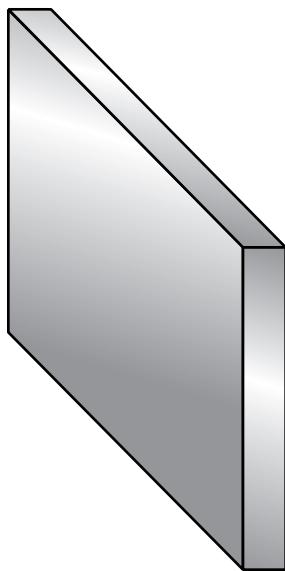
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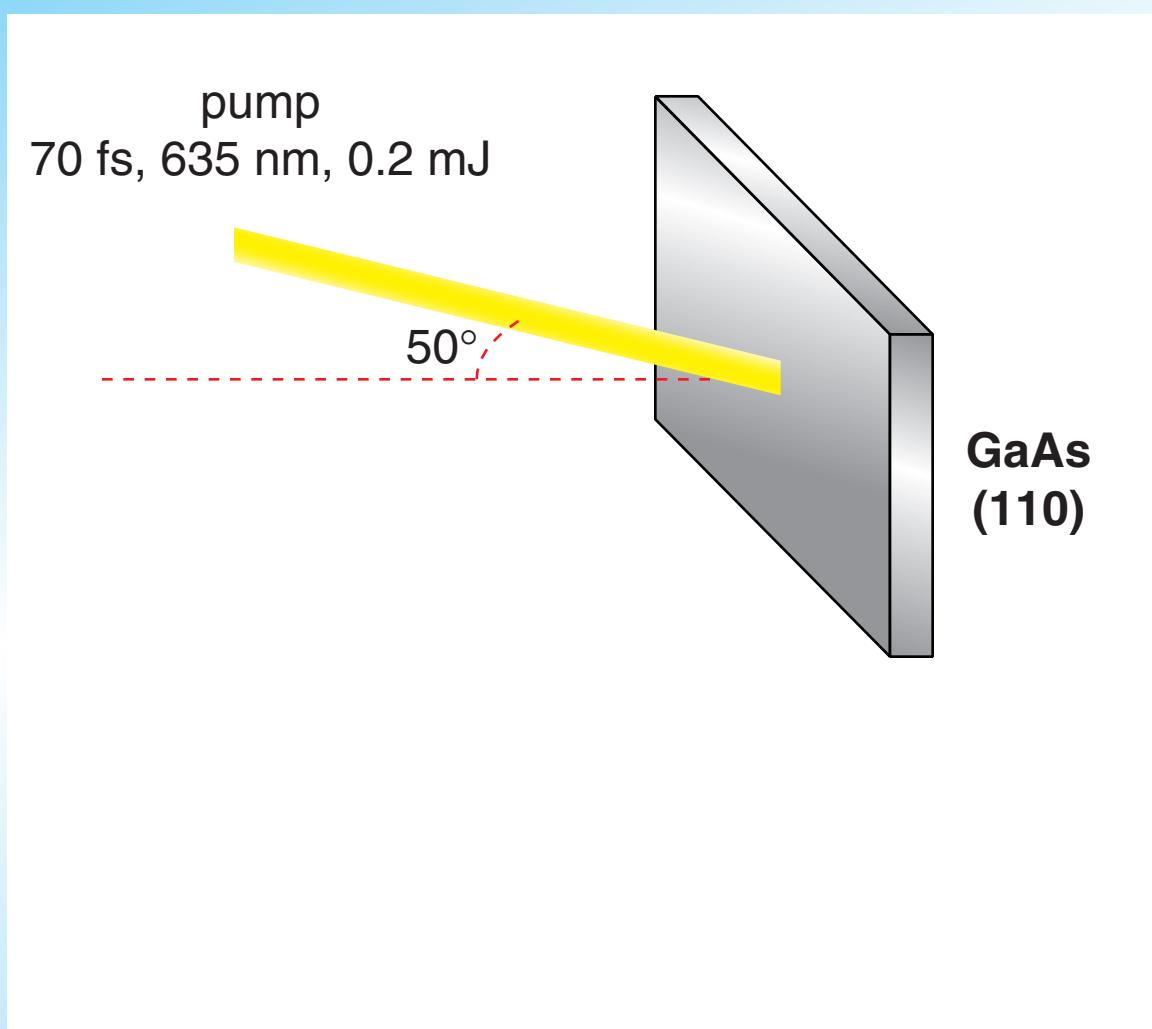
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 - ▶ Discussion

Method



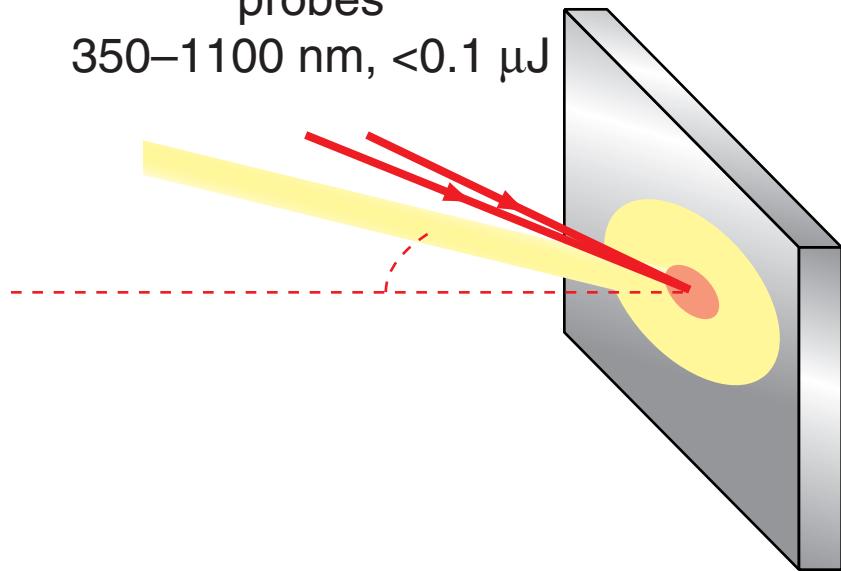
**GaAs
(110)**

Method



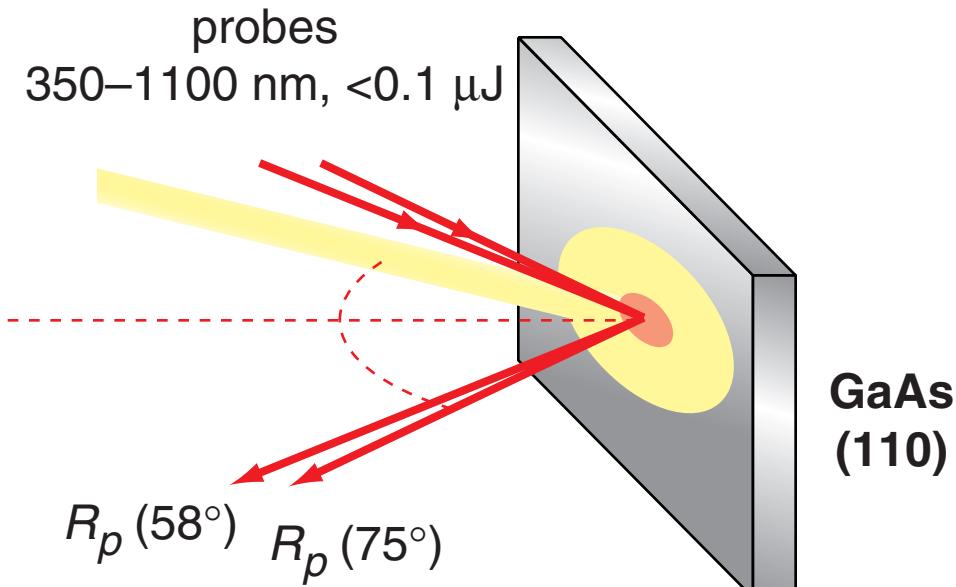
Method

probes
350–1100 nm, $<0.1 \mu\text{J}$



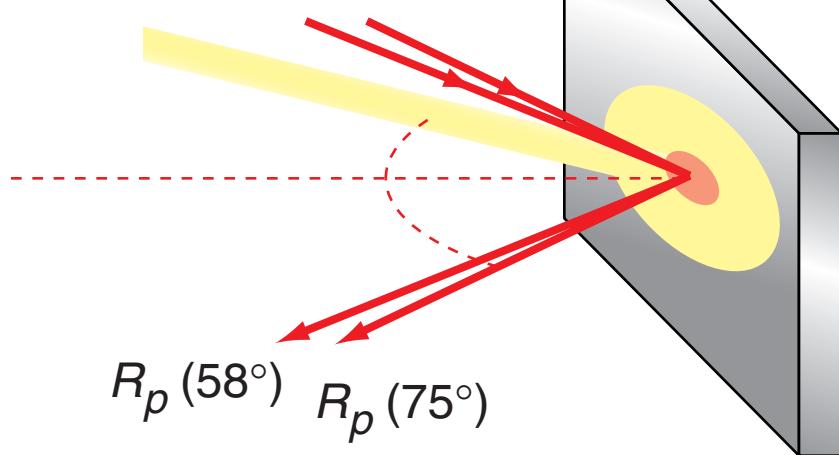
**GaAs
(110)**

Method



Method

probes
350–1100 nm, <0.1 μ J



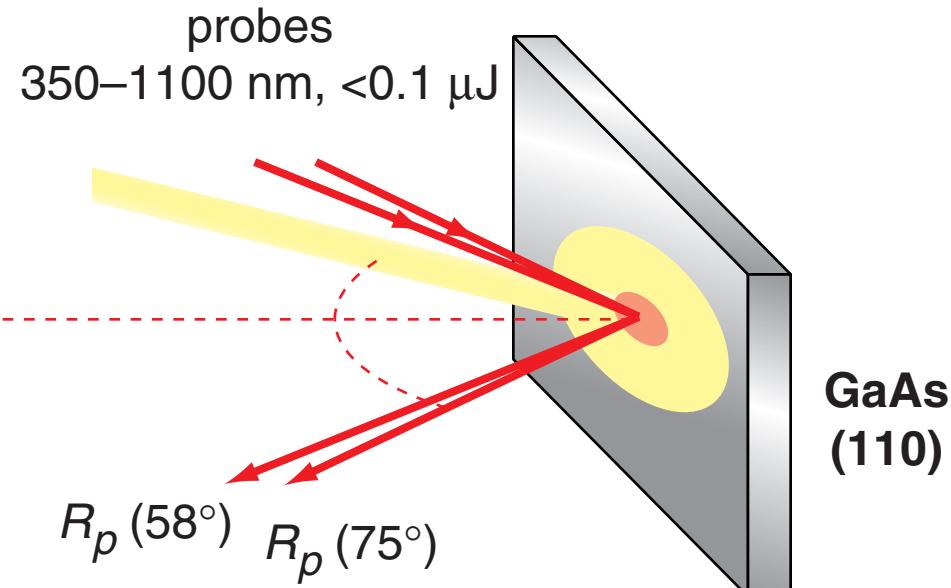
GaAs
(110)

$R_p (58^\circ)$ $R_p (75^\circ)$



Fresnel
equations

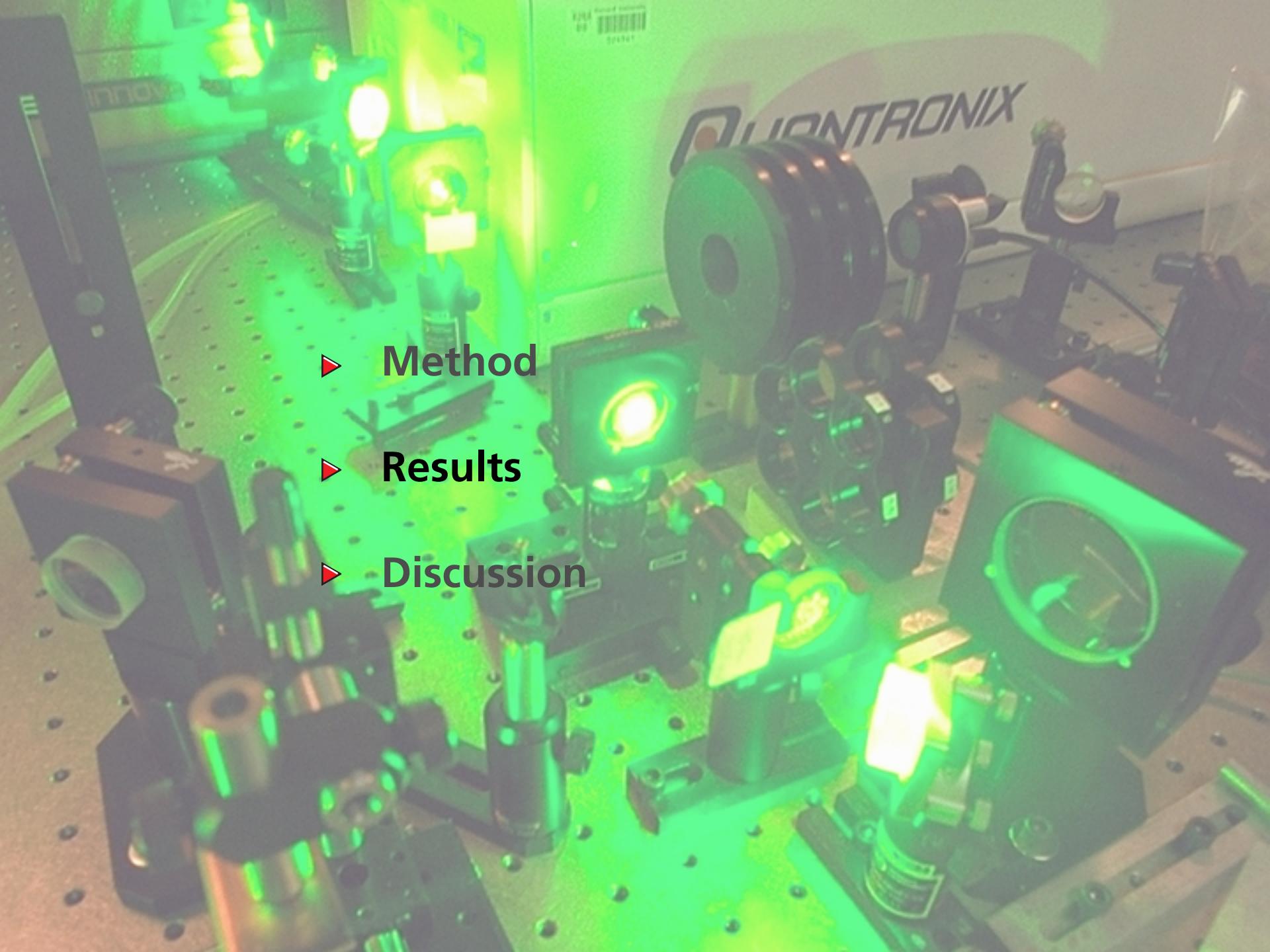
Method



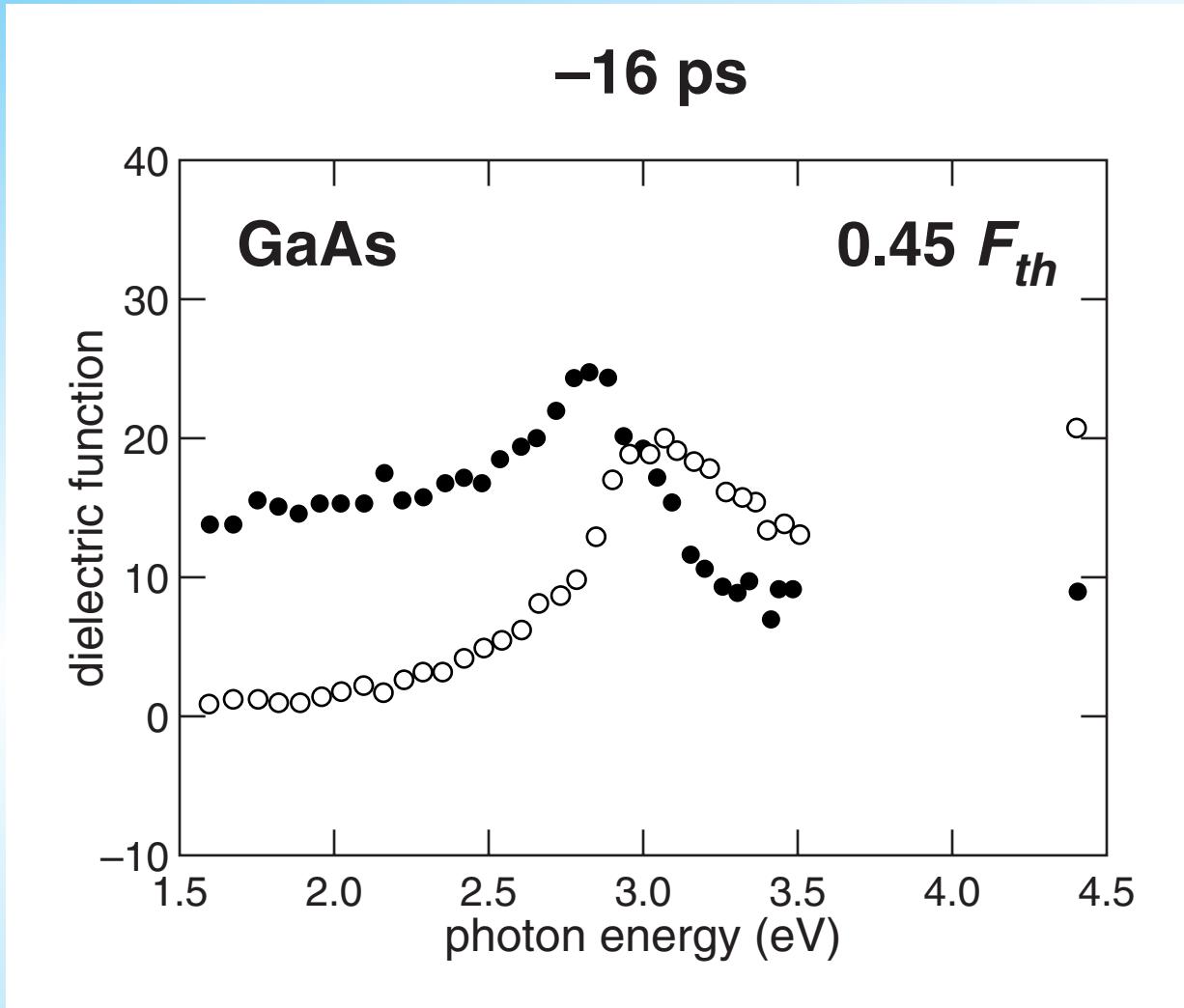
Fresnel
equations



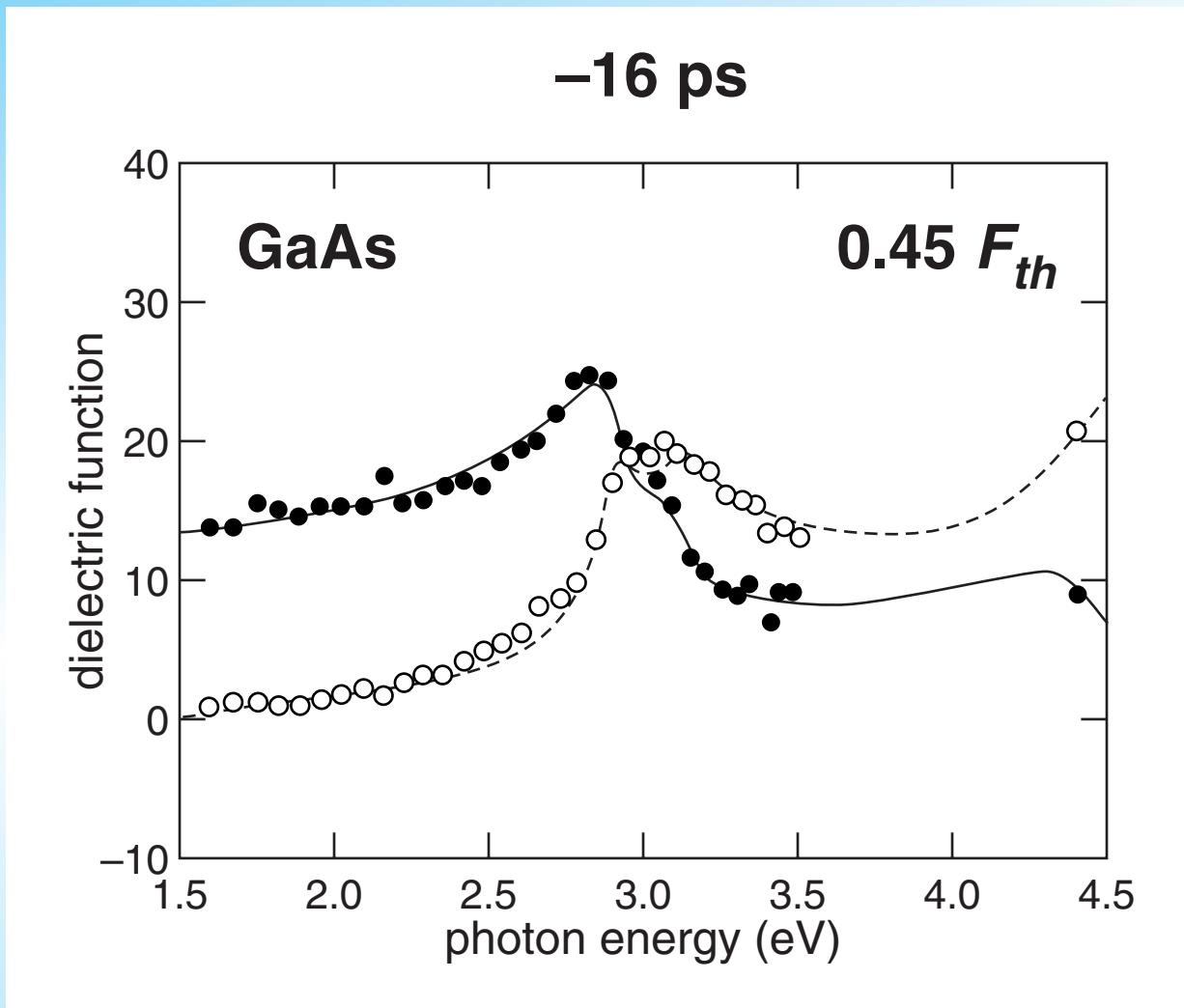
$\text{Re } \varepsilon(\omega)$
 $\text{Im } \varepsilon(\omega)$

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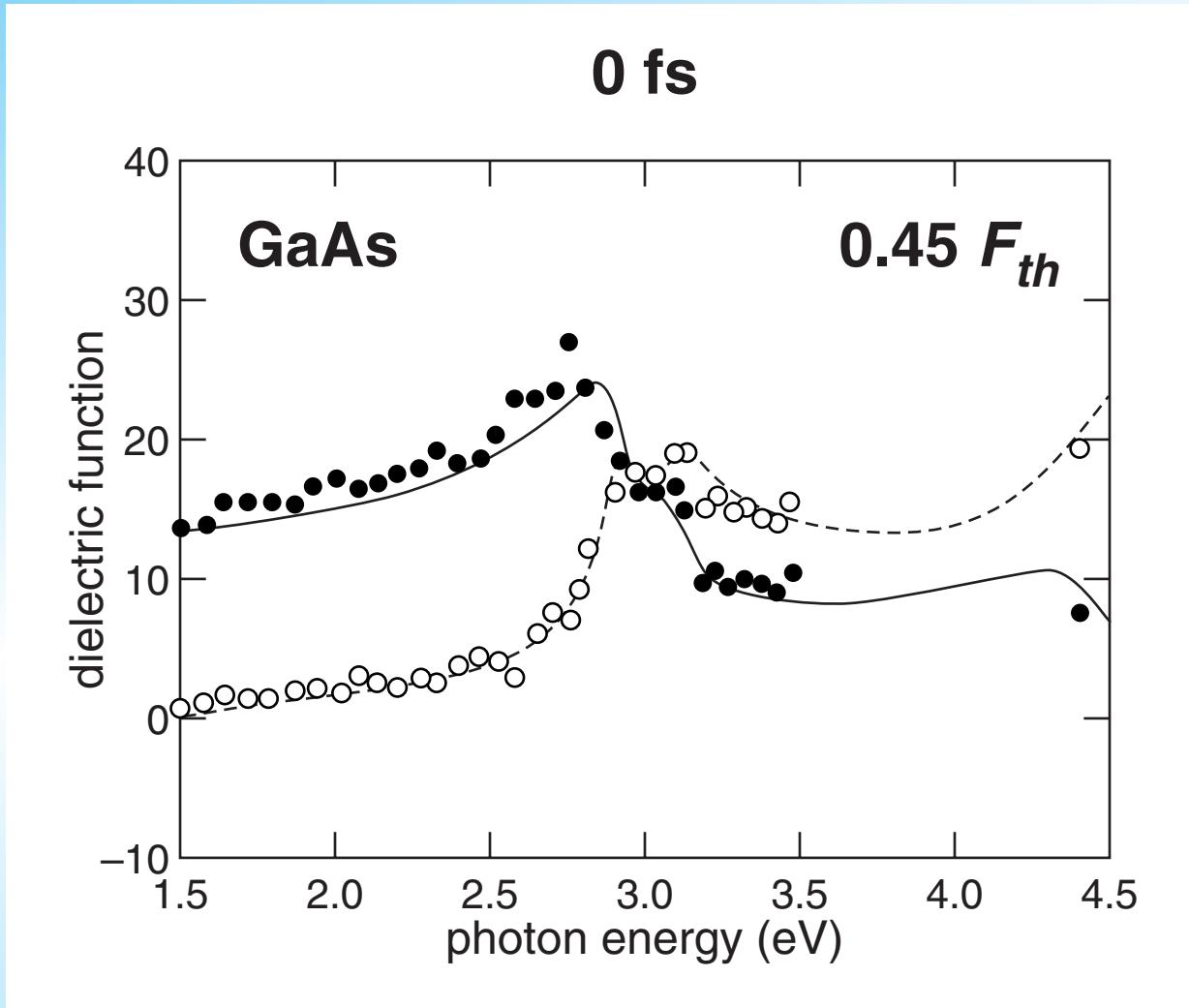
Results



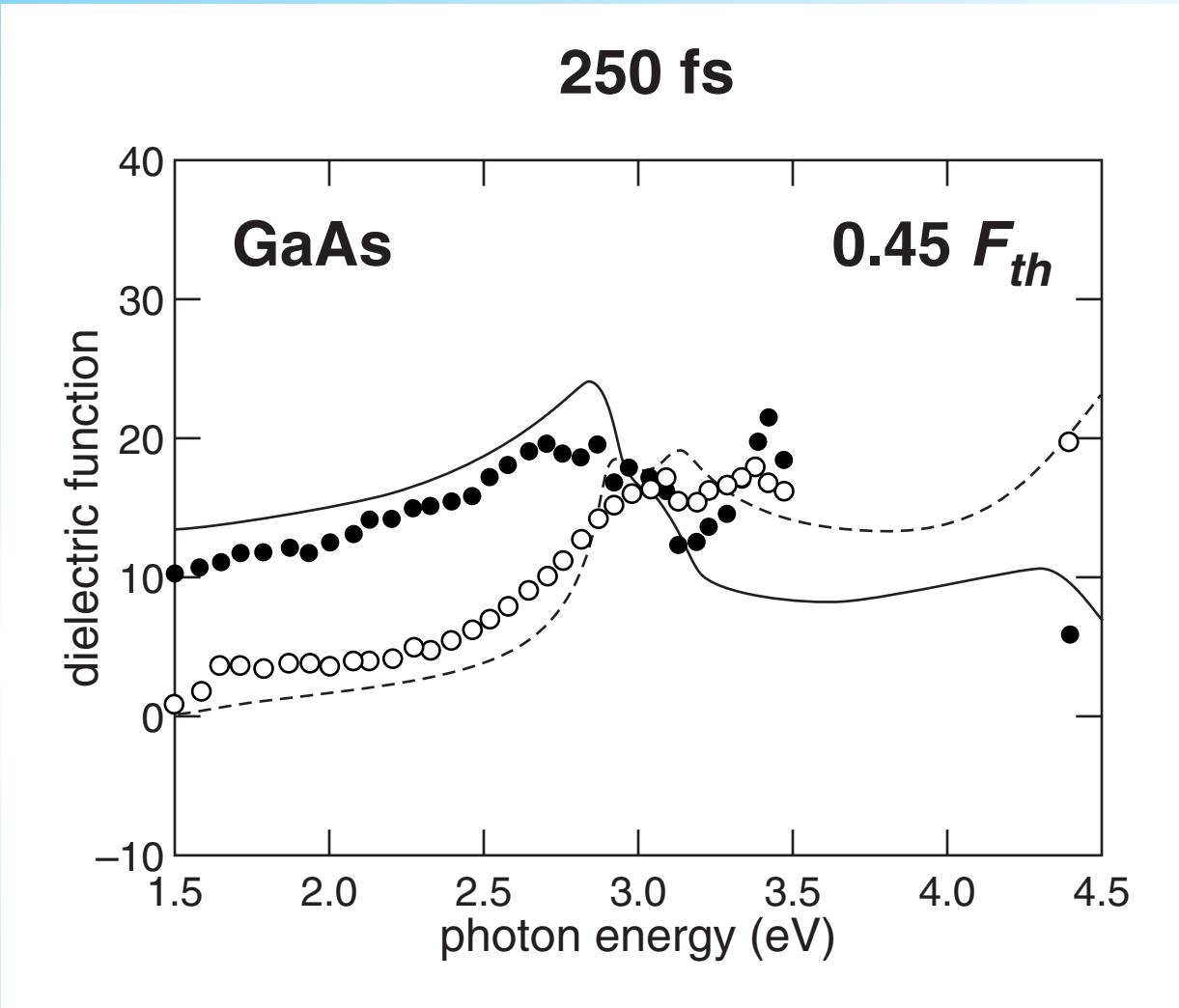
Results



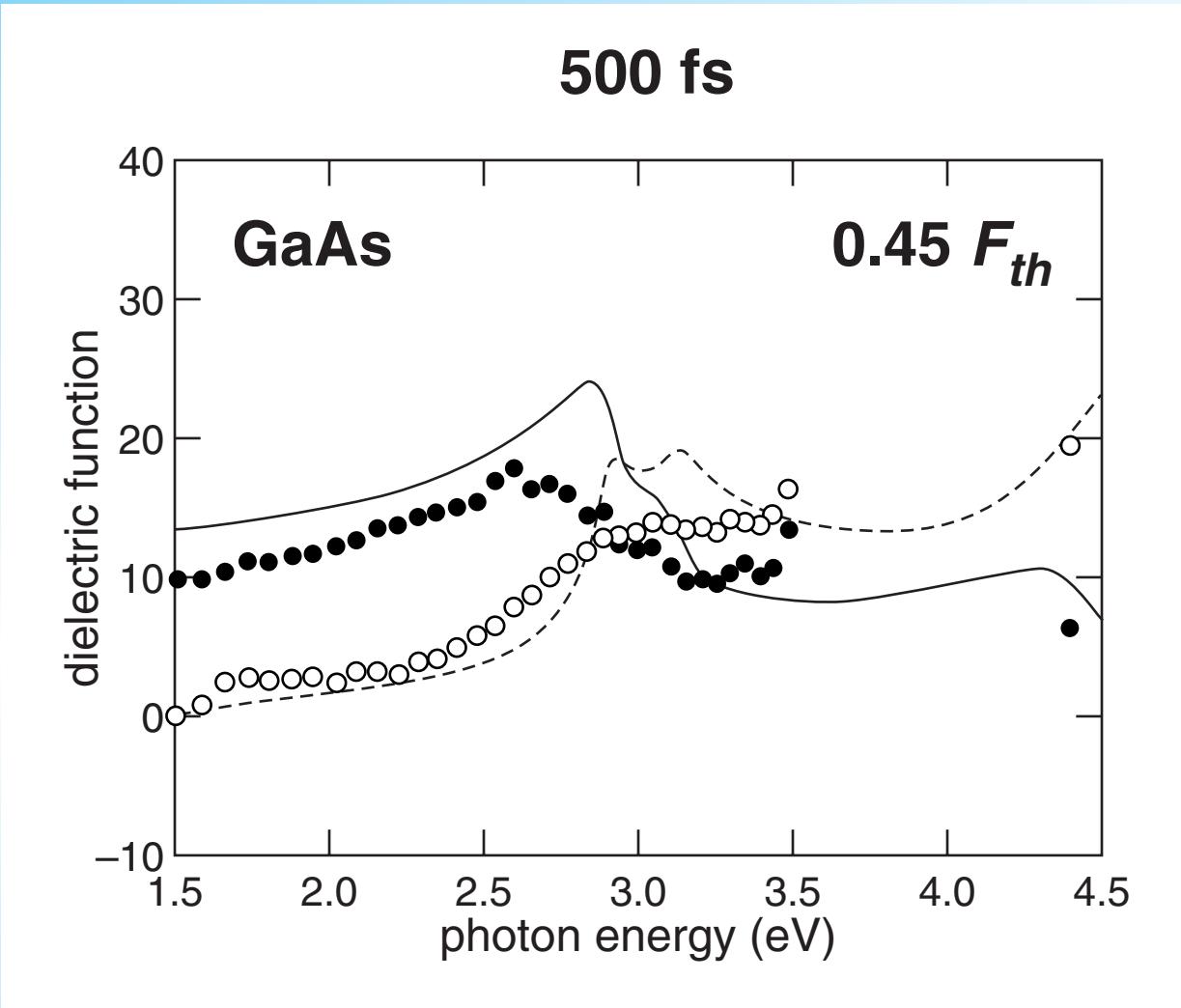
Results



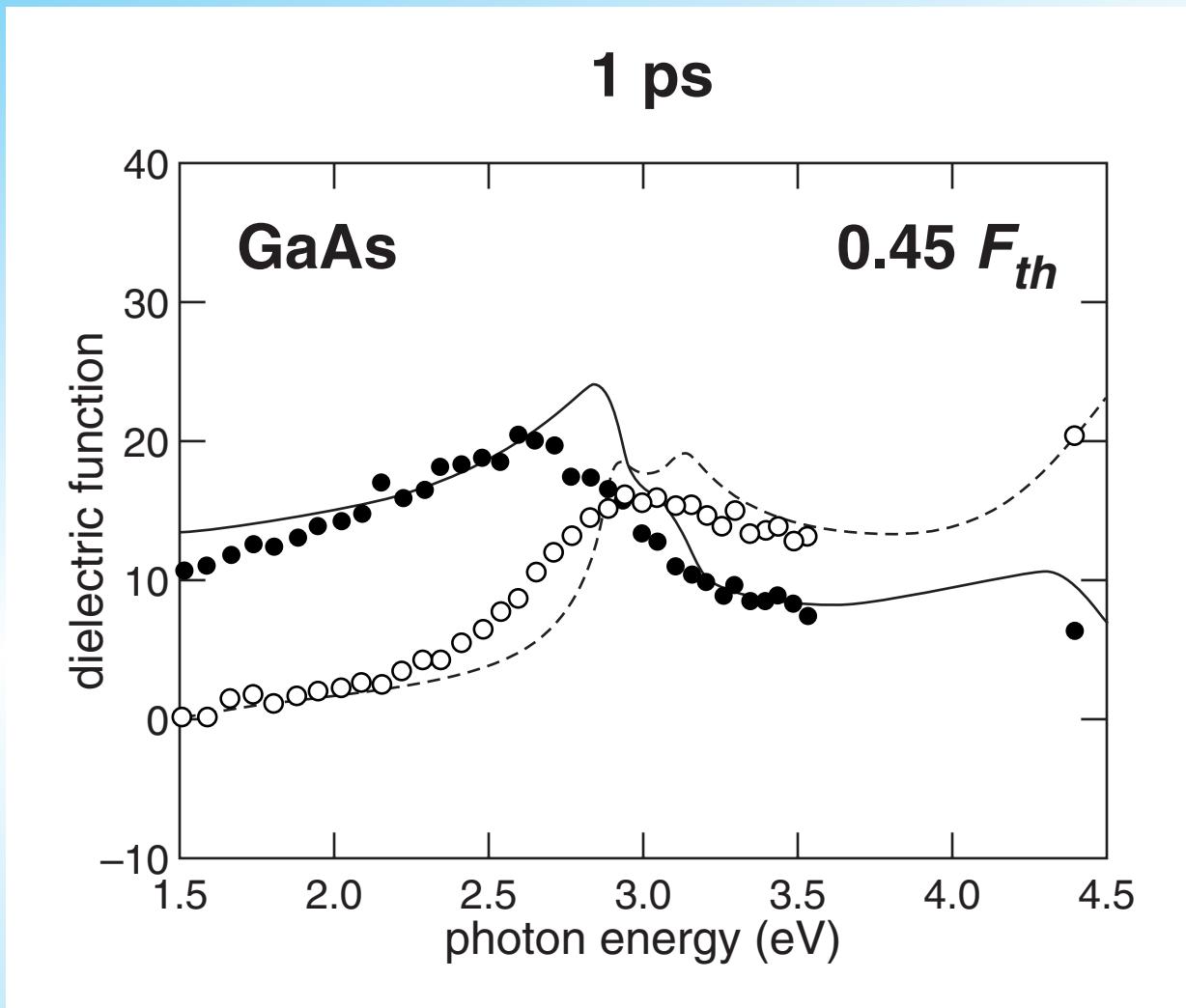
Results



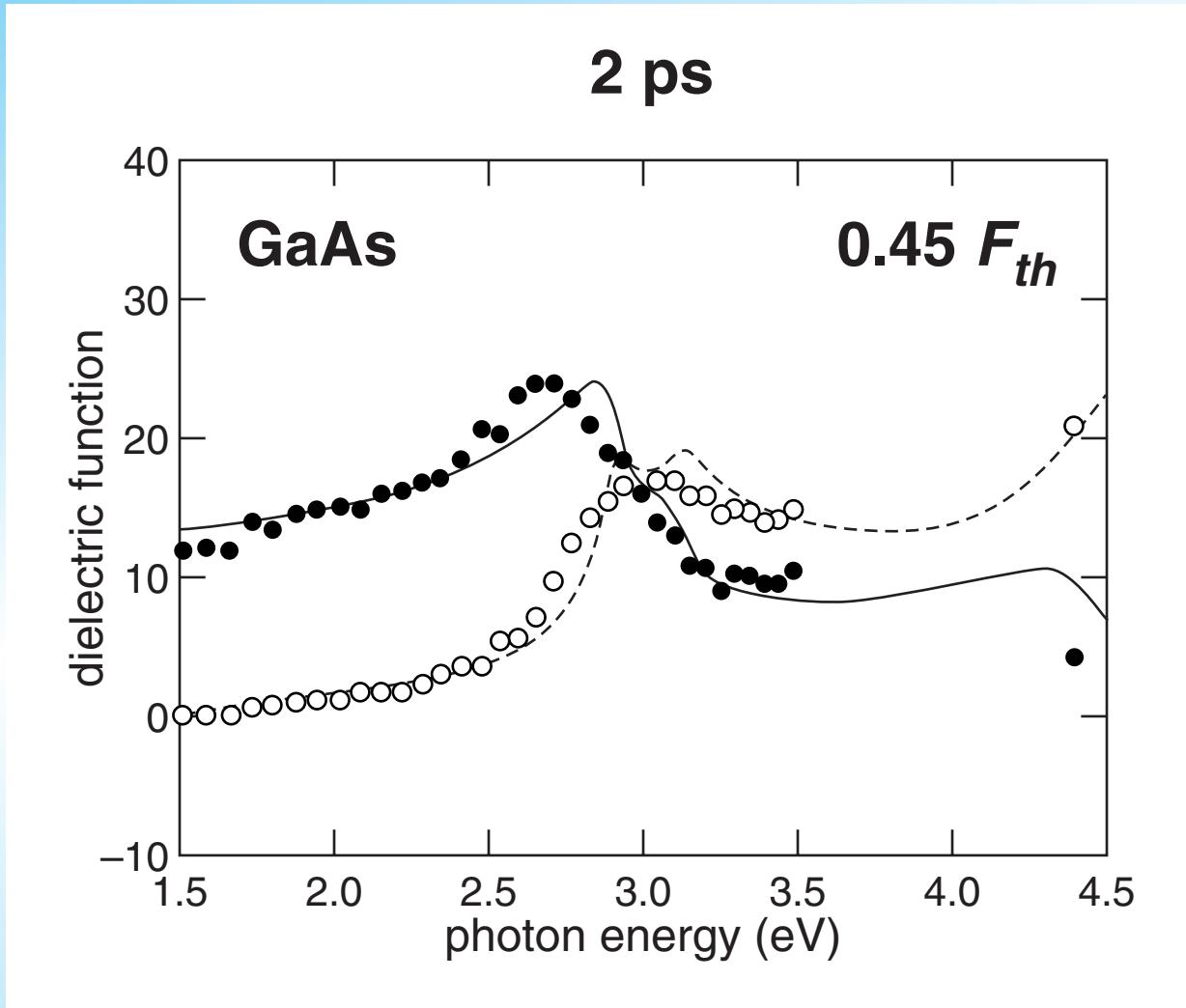
Results



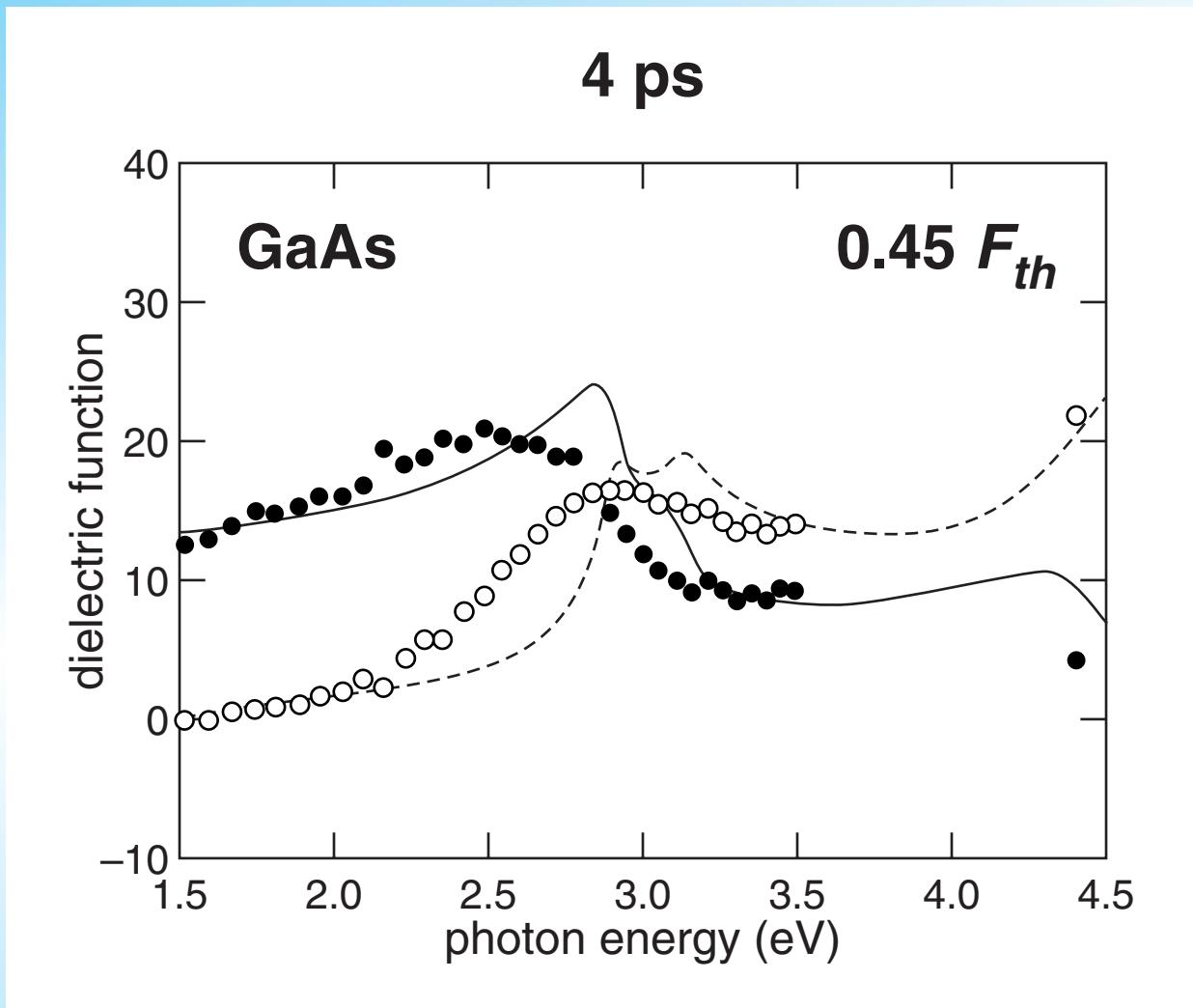
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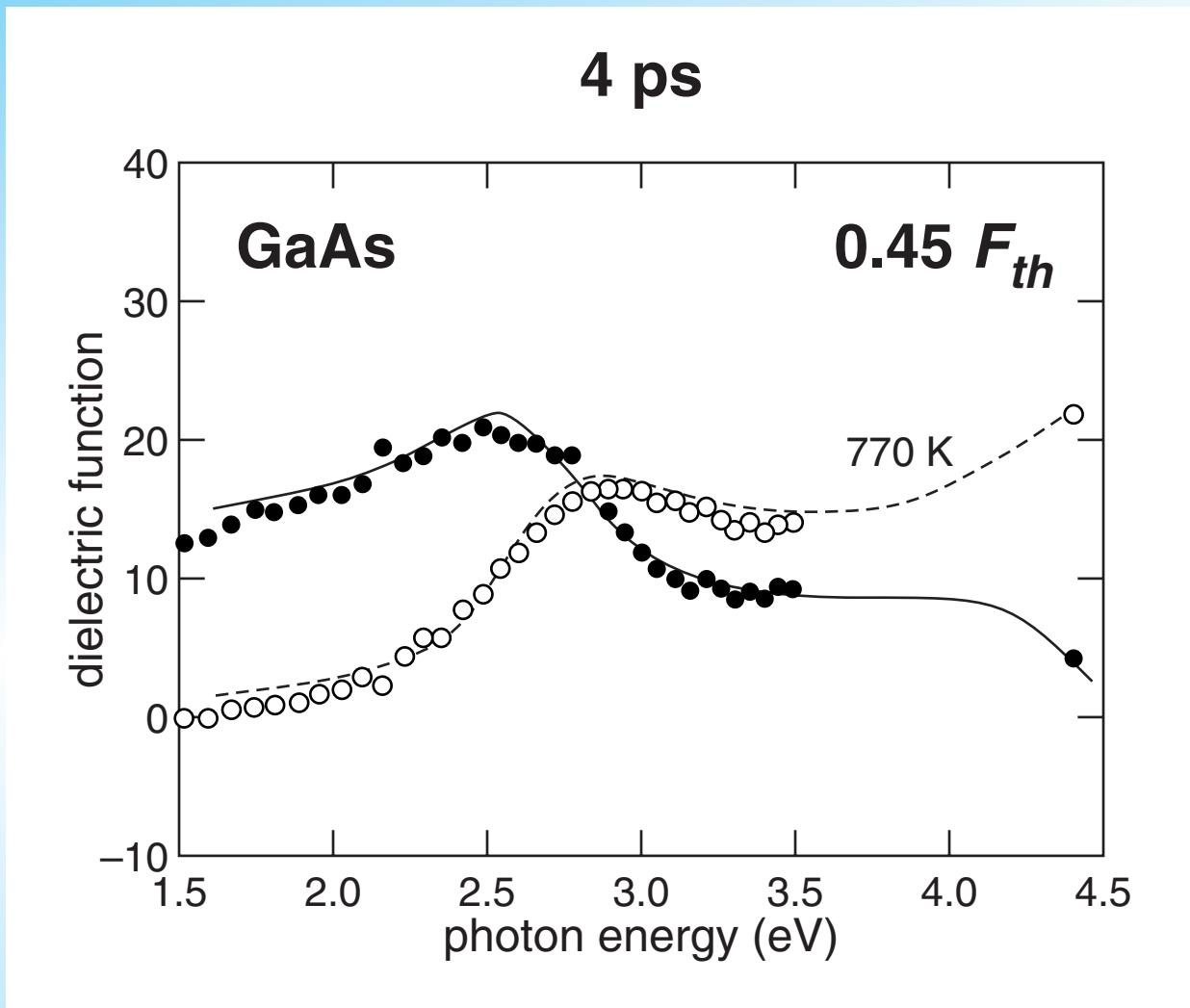
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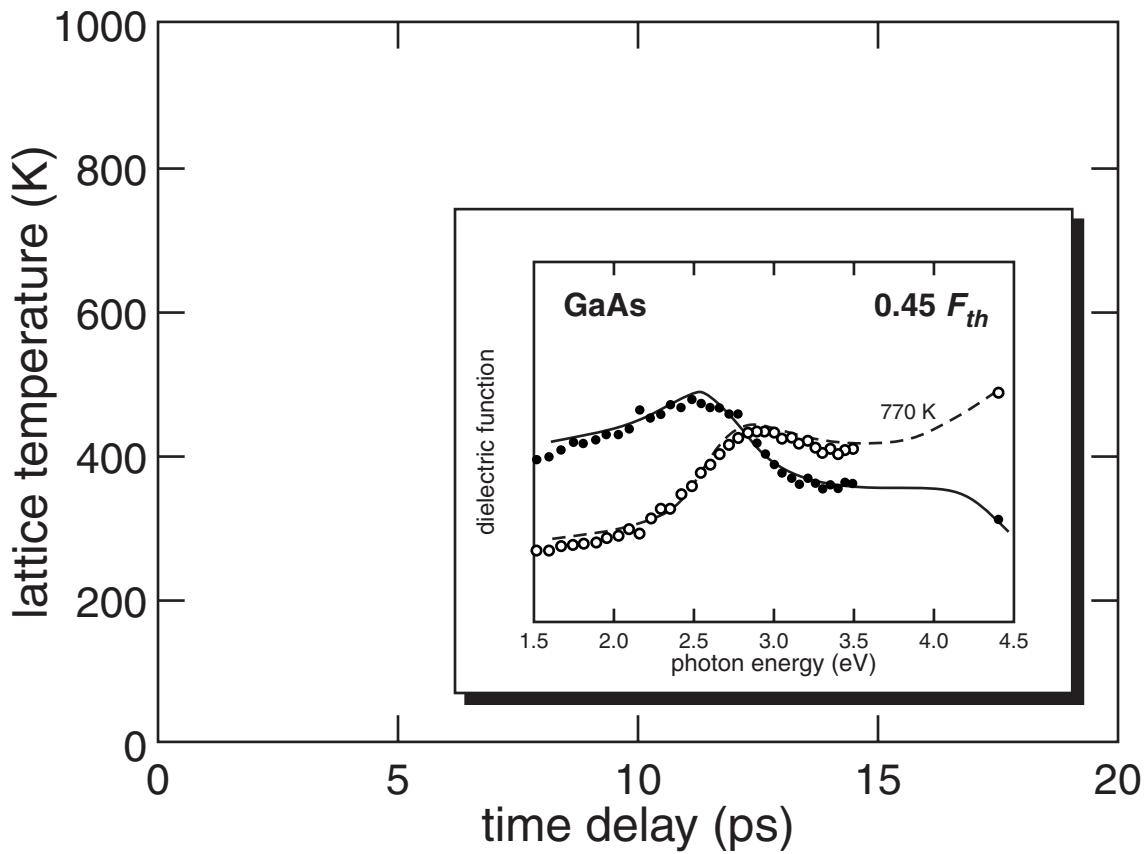
Results



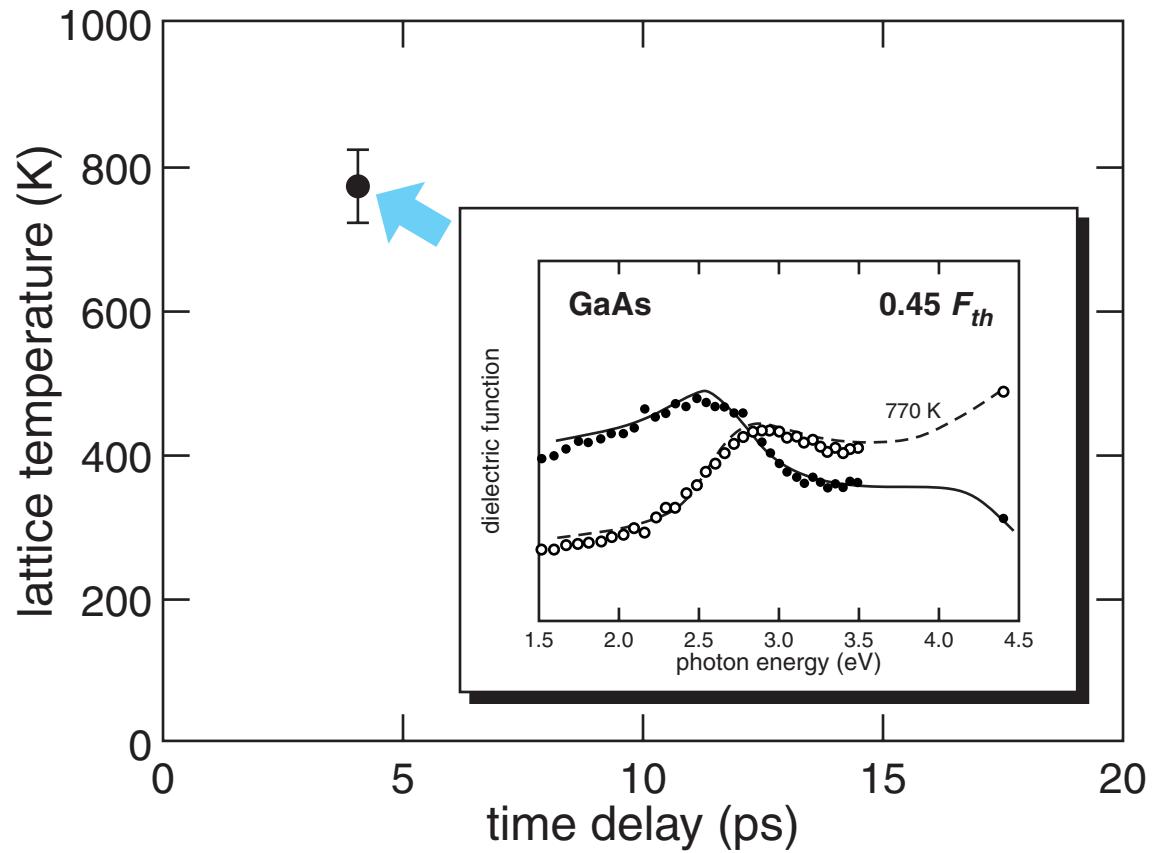
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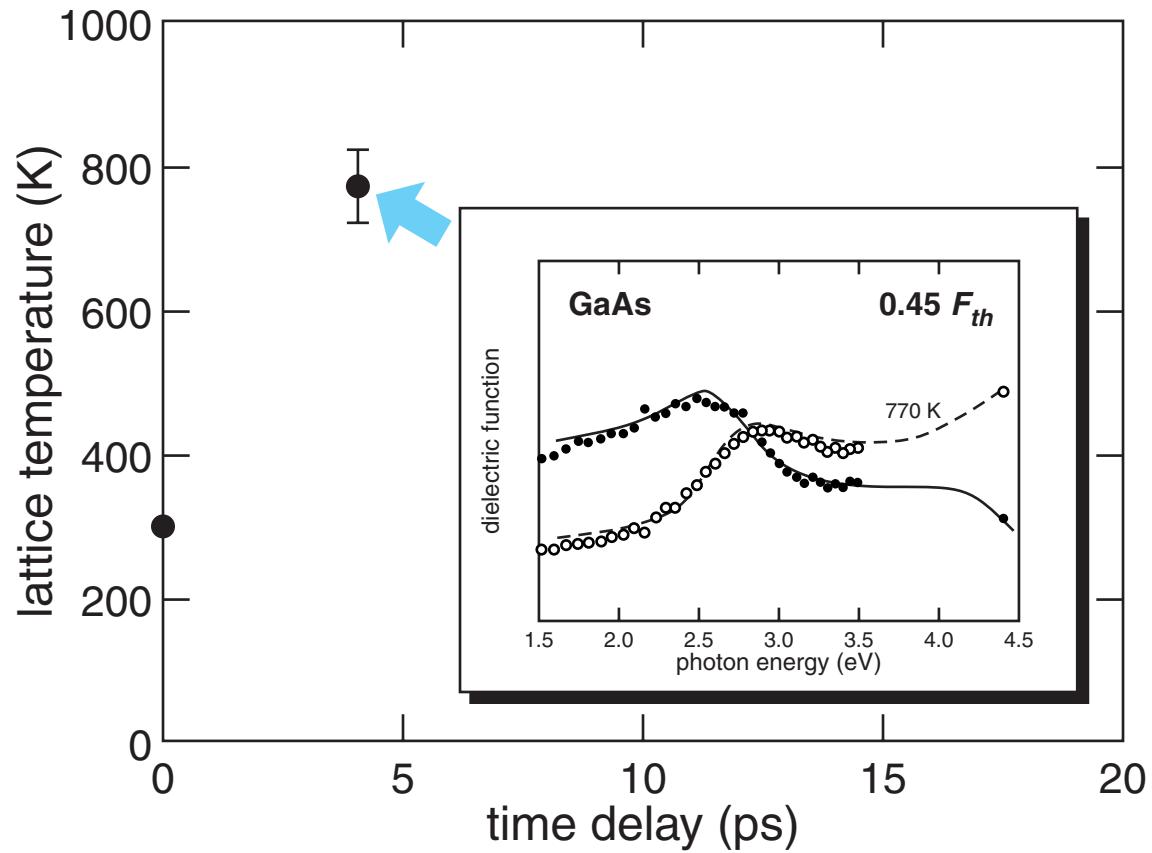
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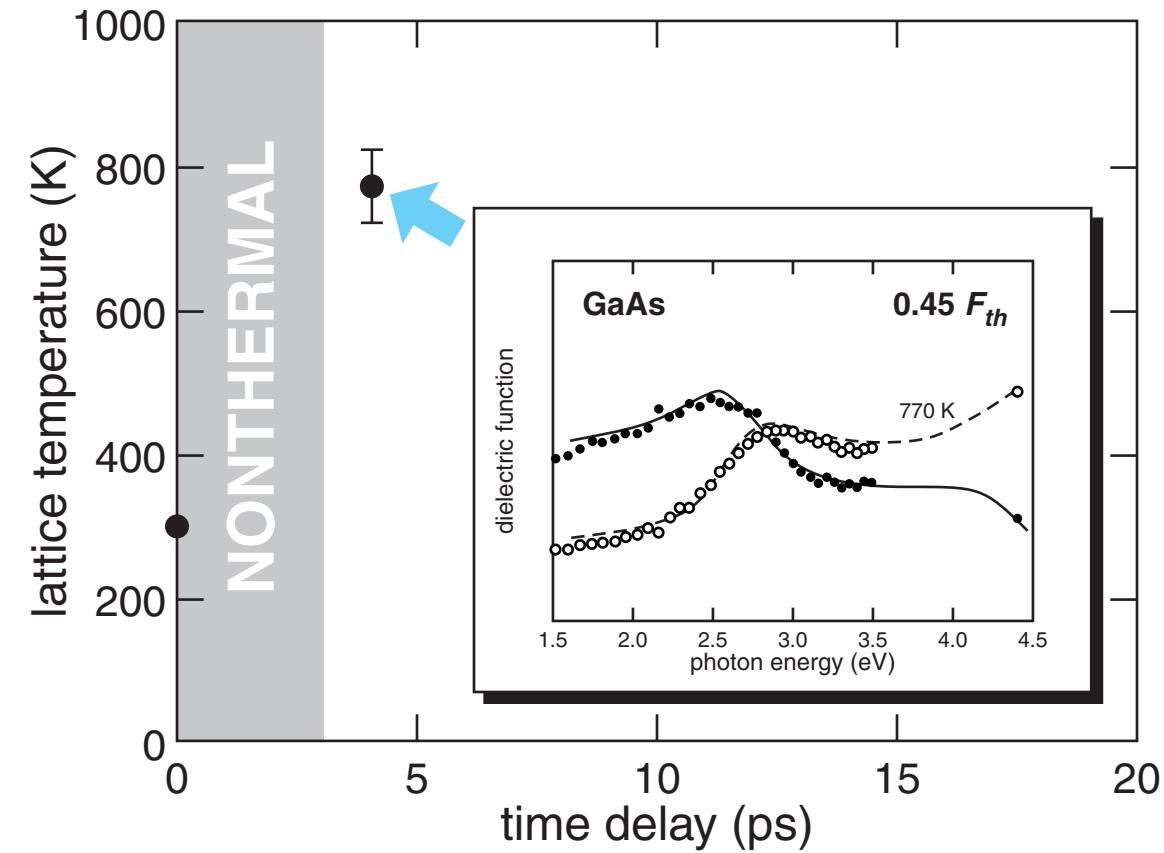
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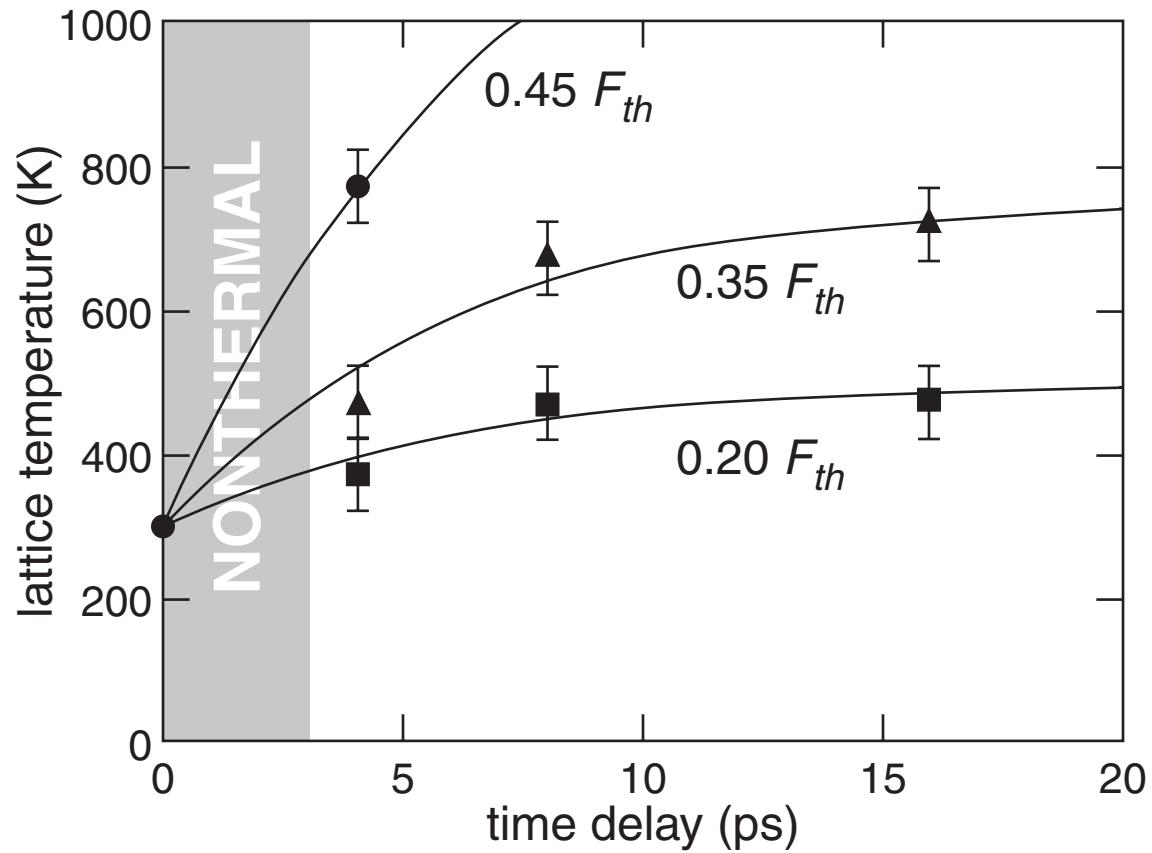
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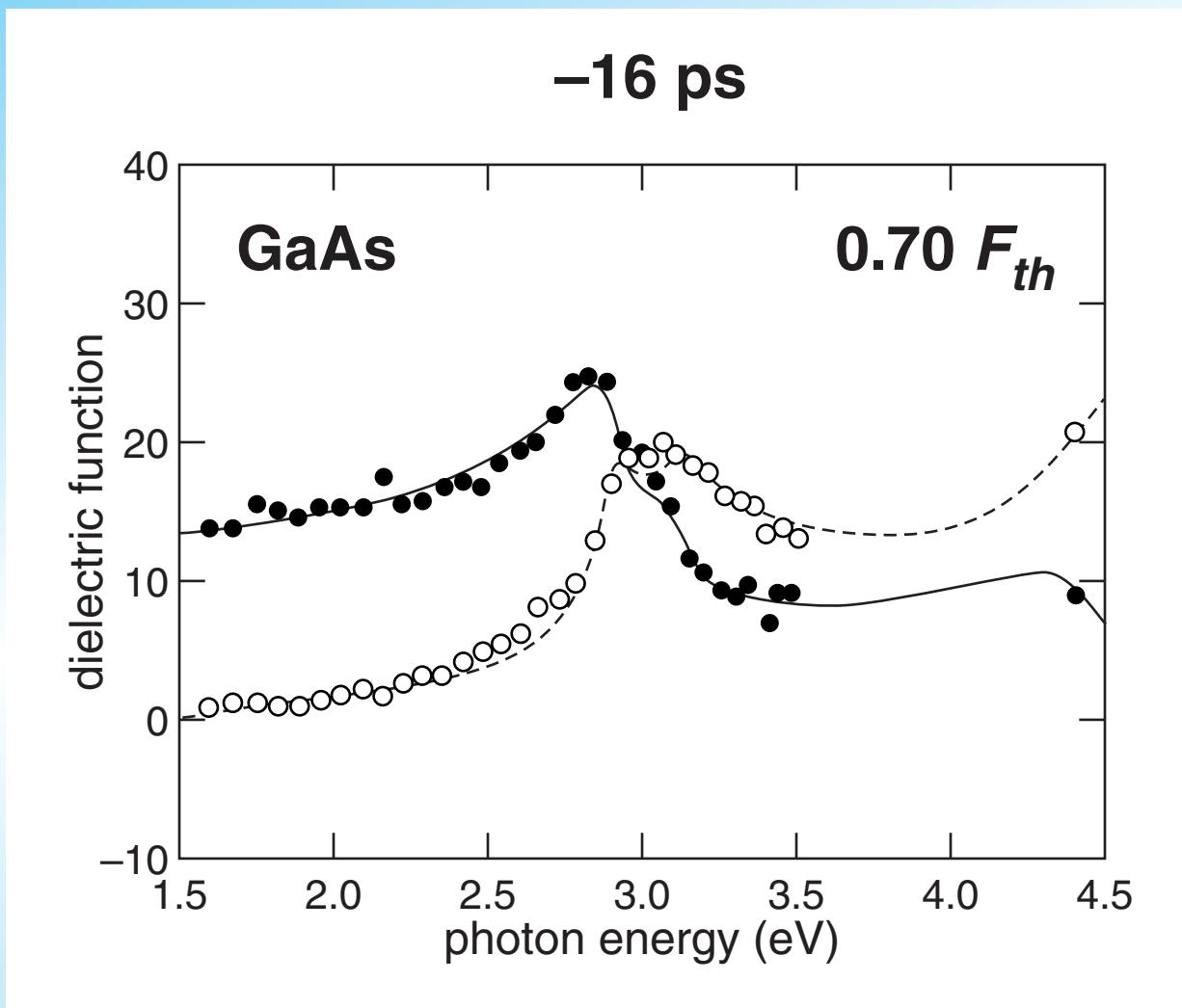
Results



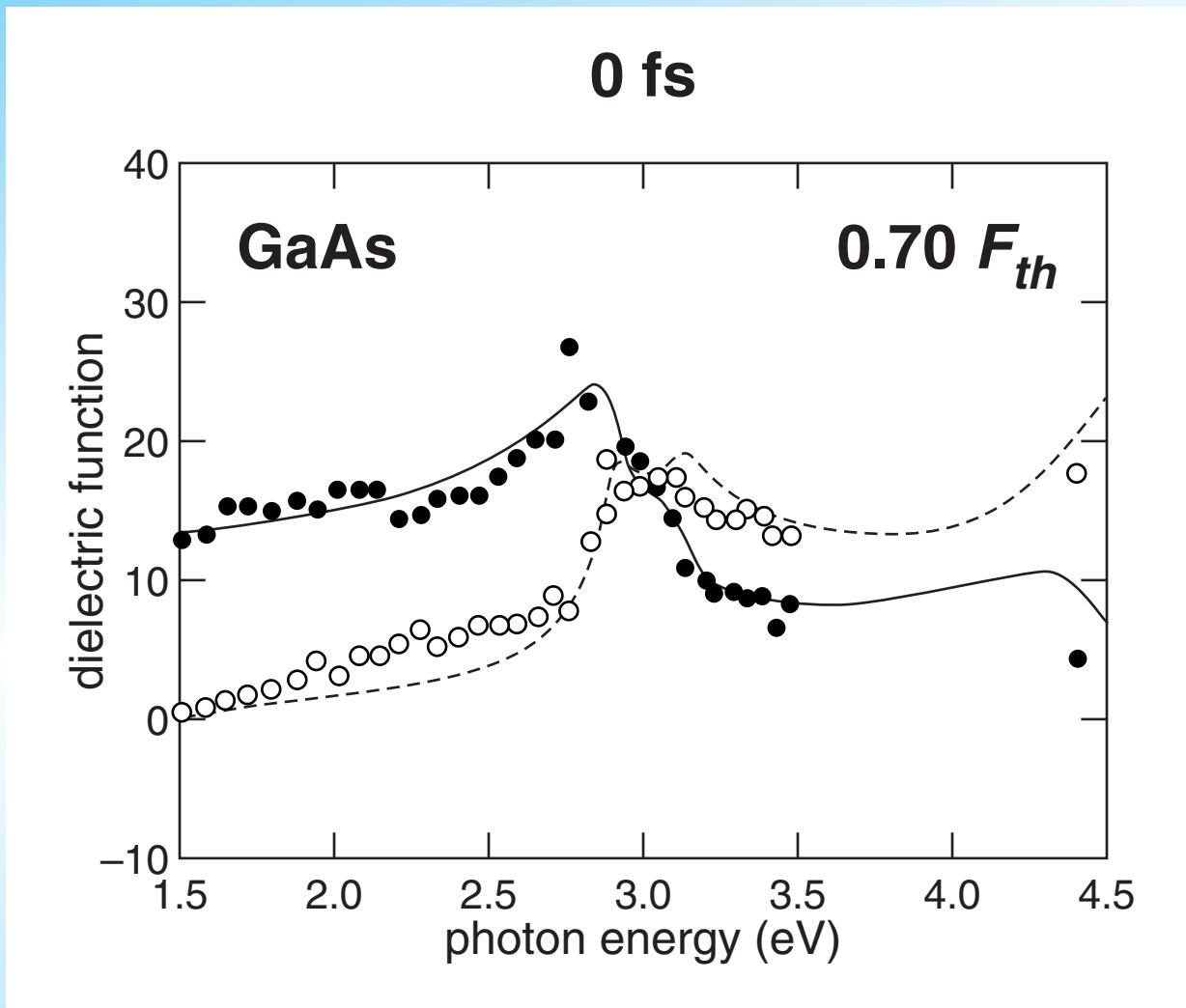
Results



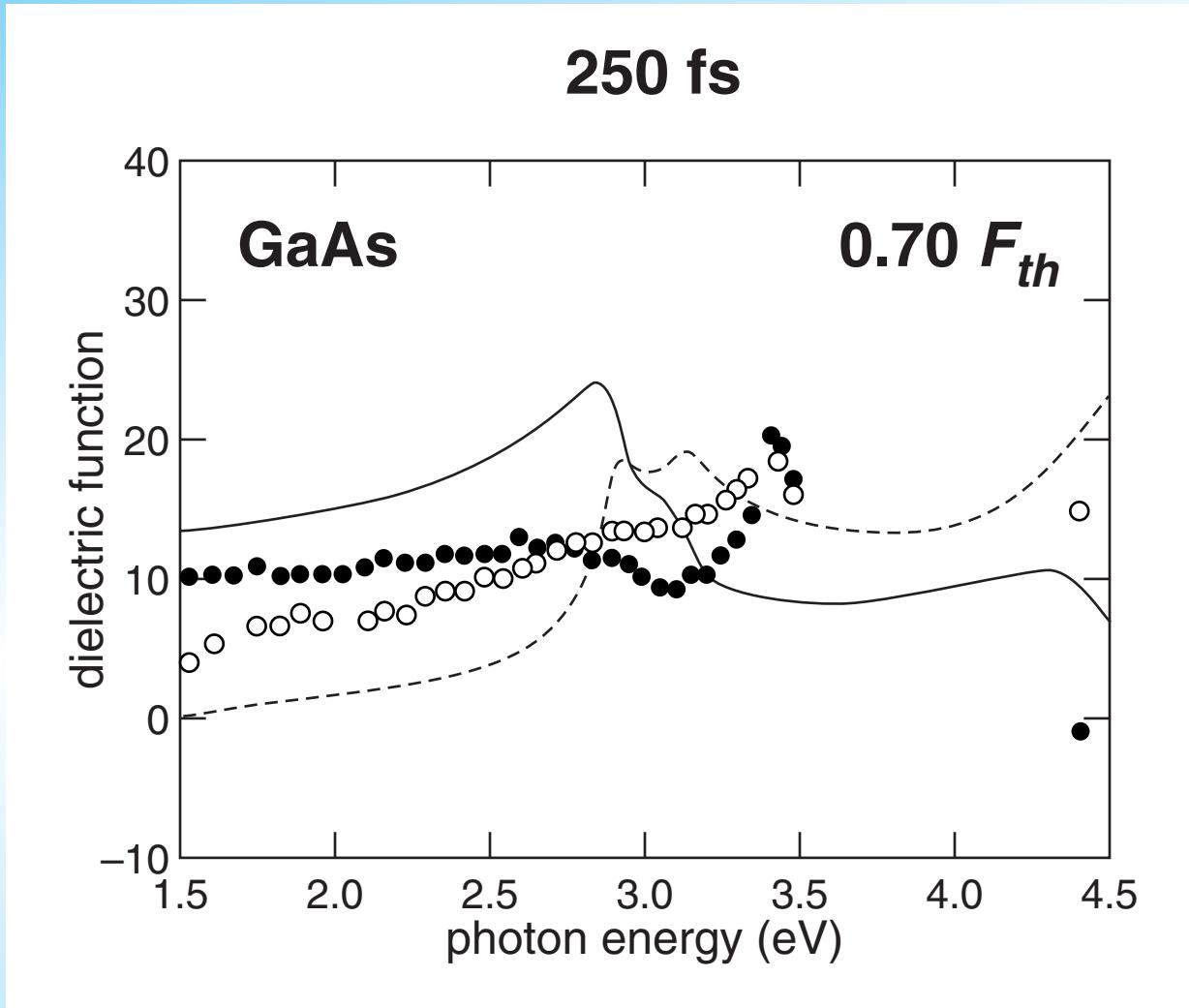
Results



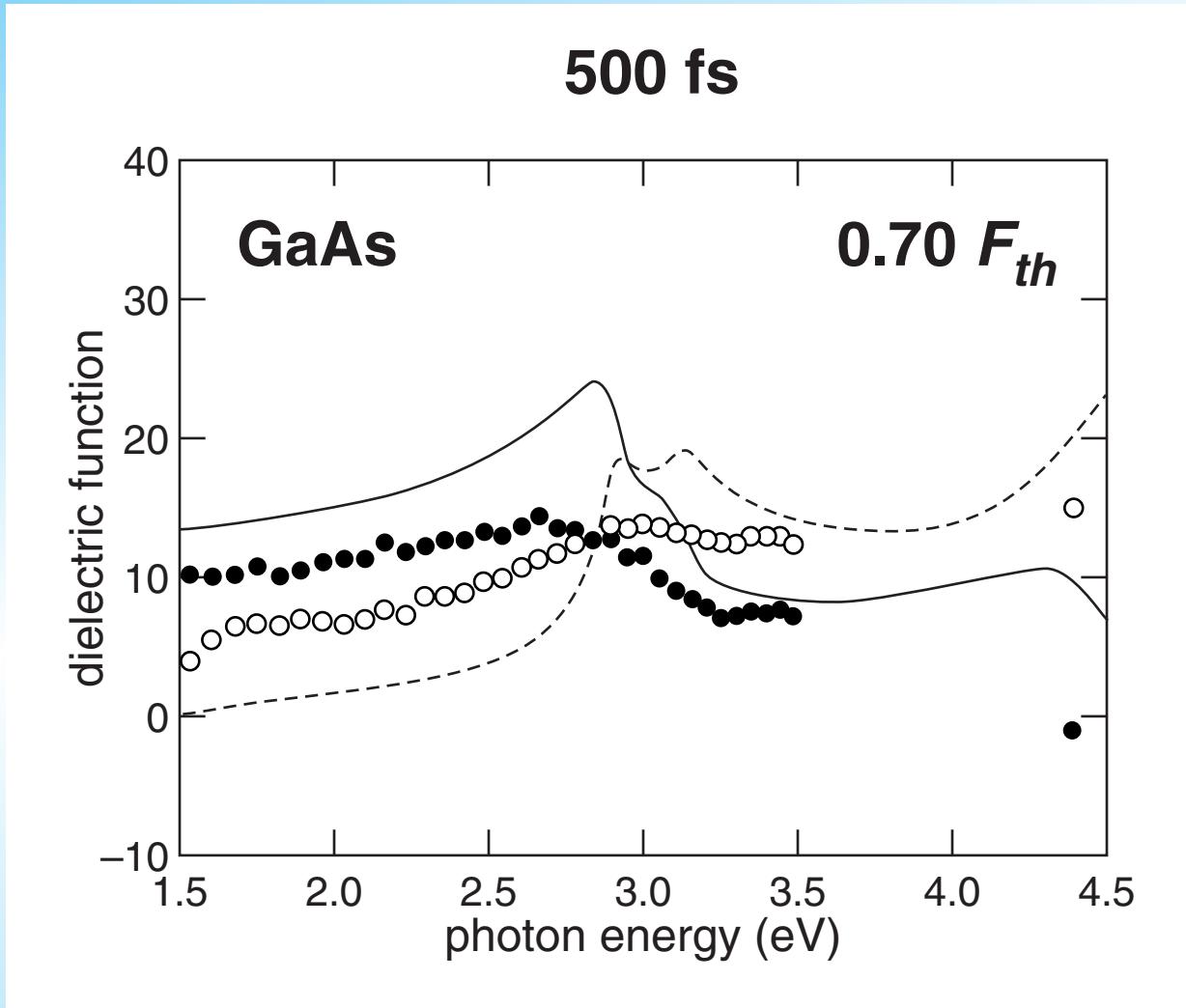
Results



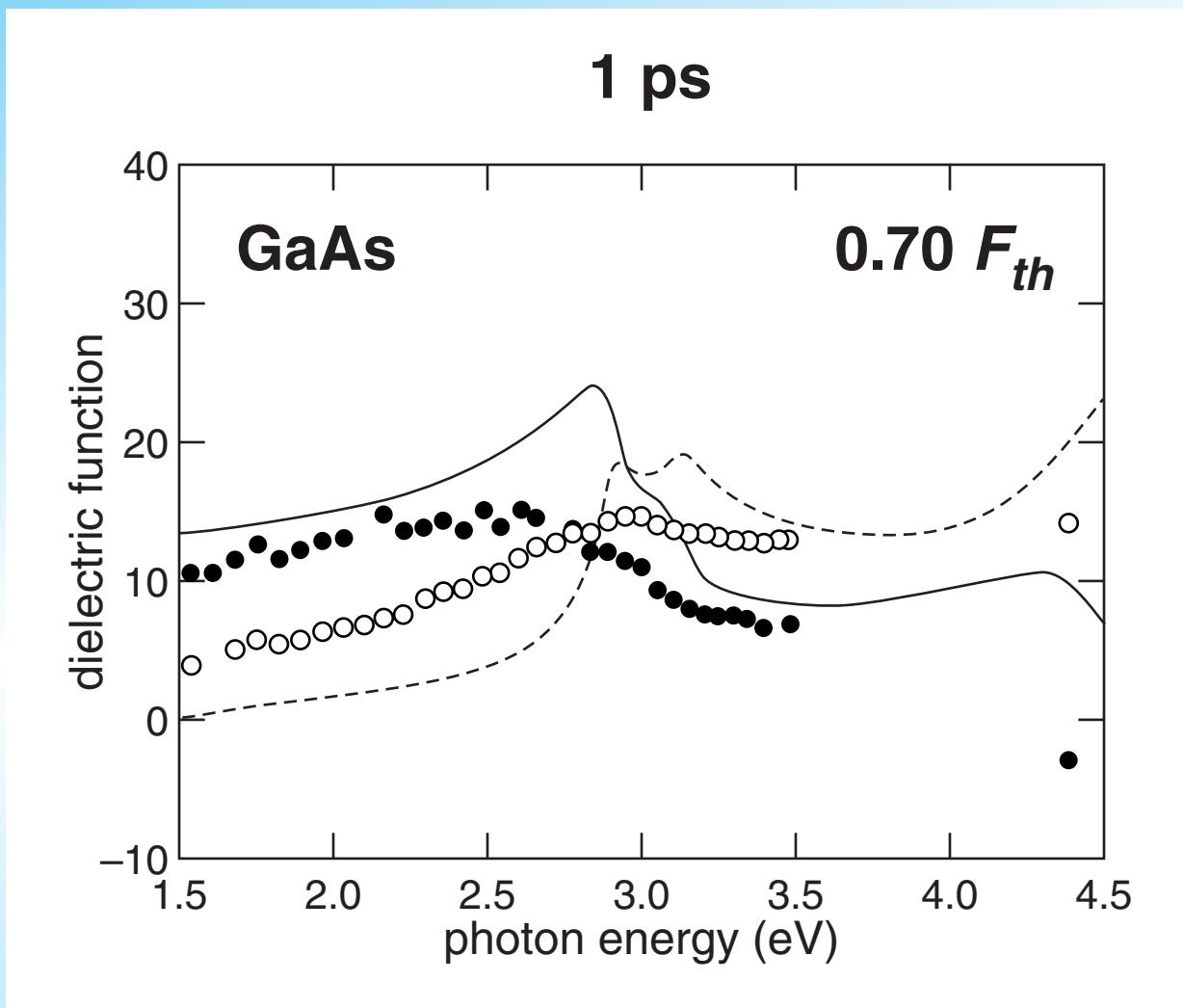
Results



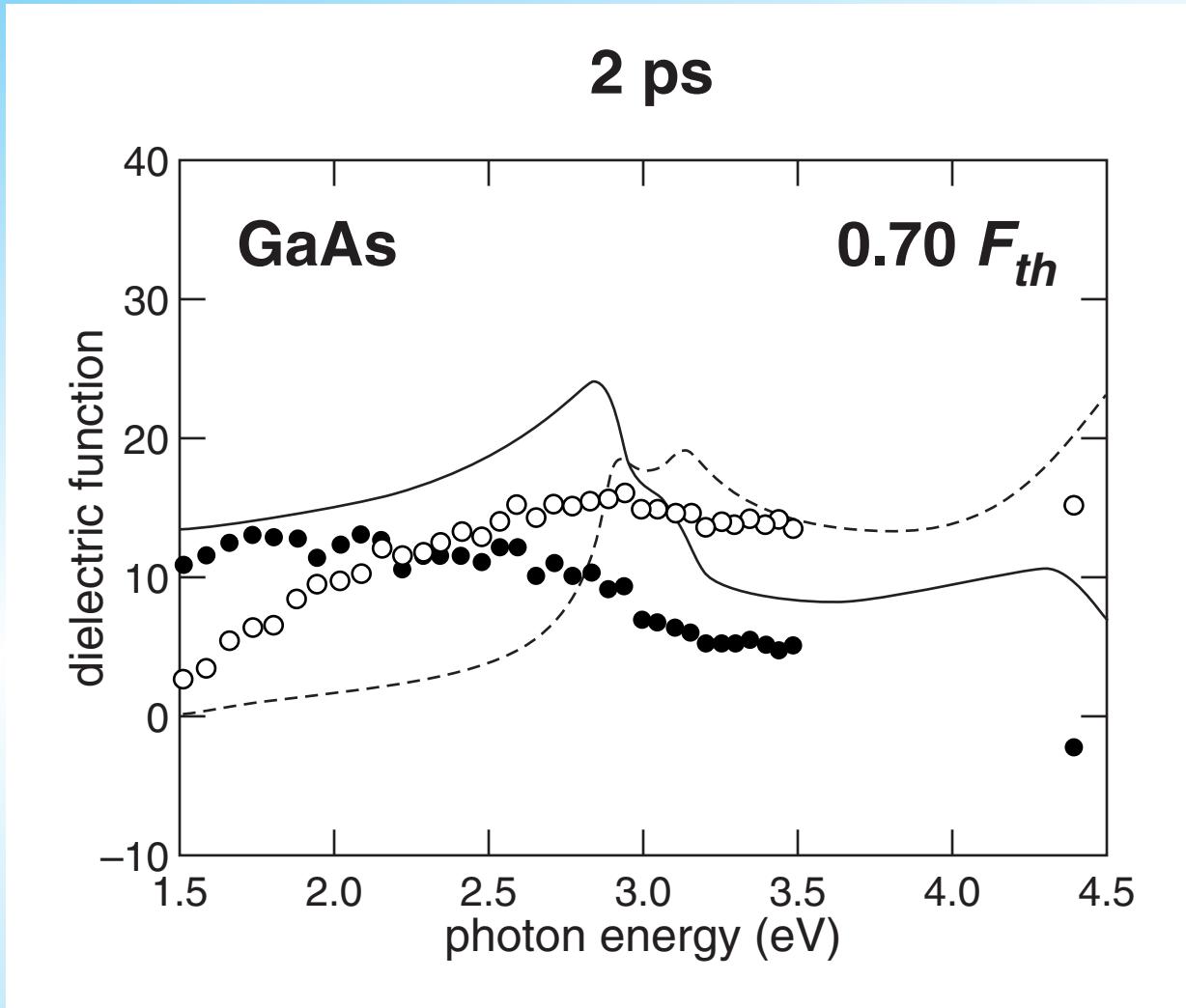
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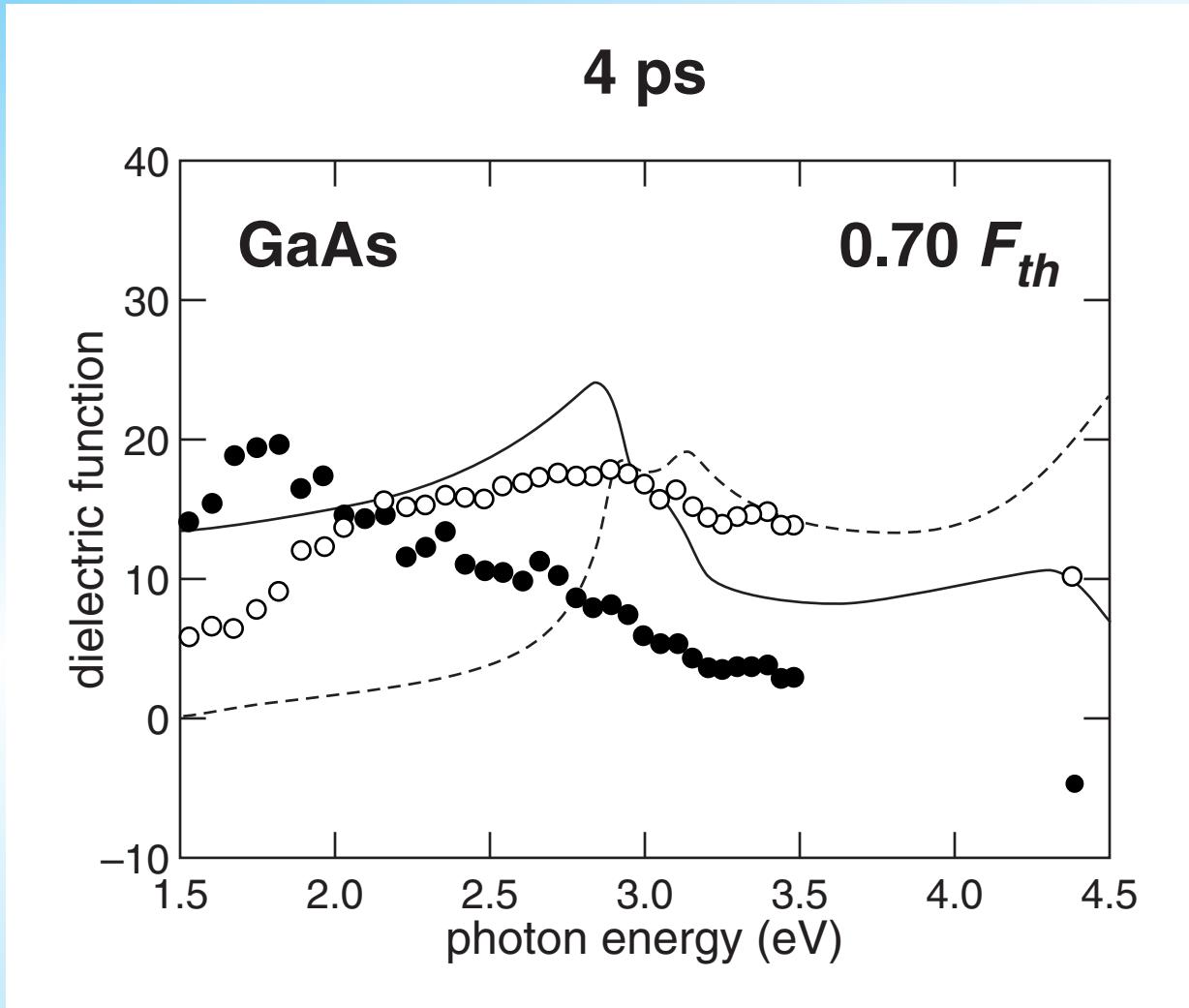
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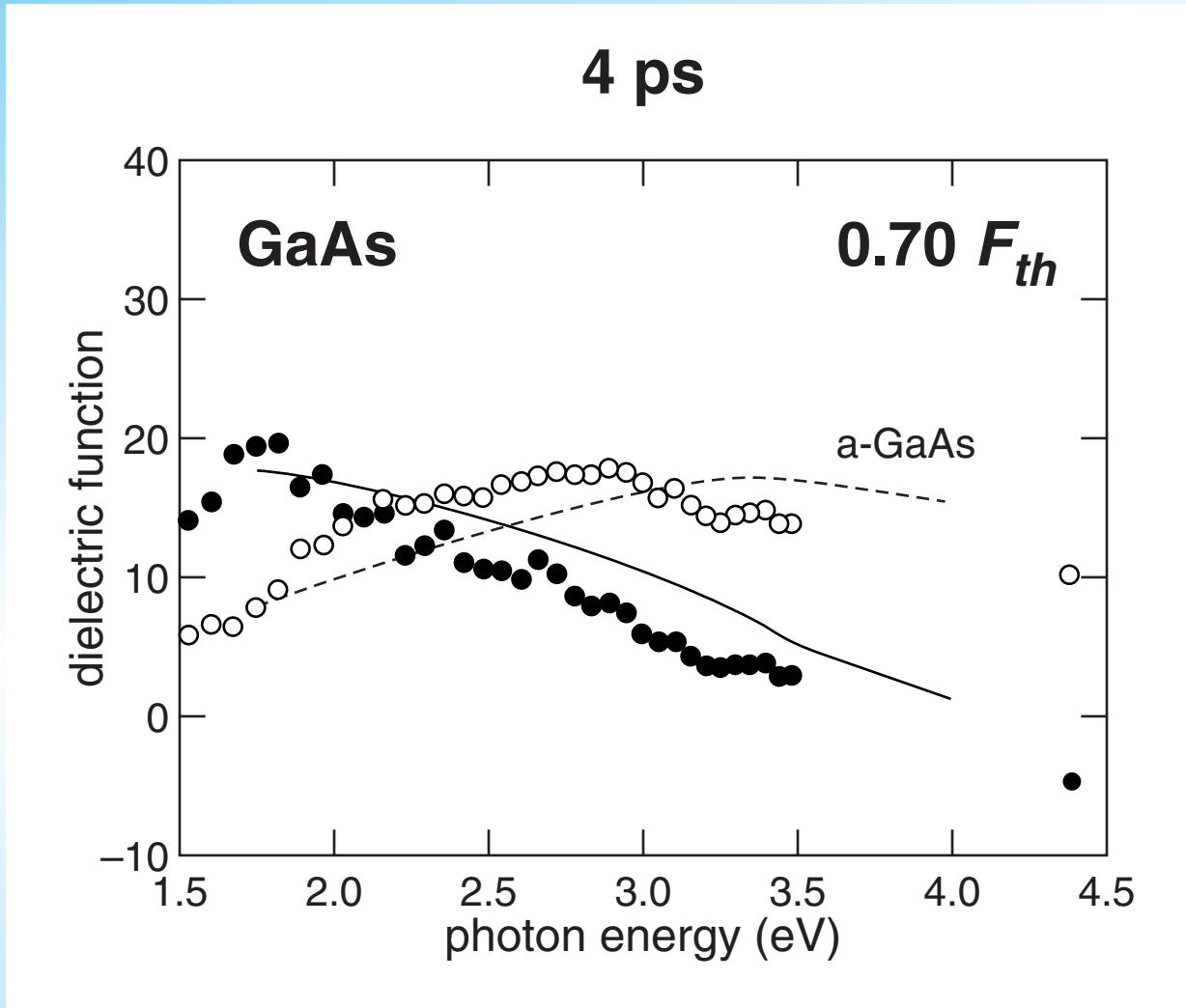
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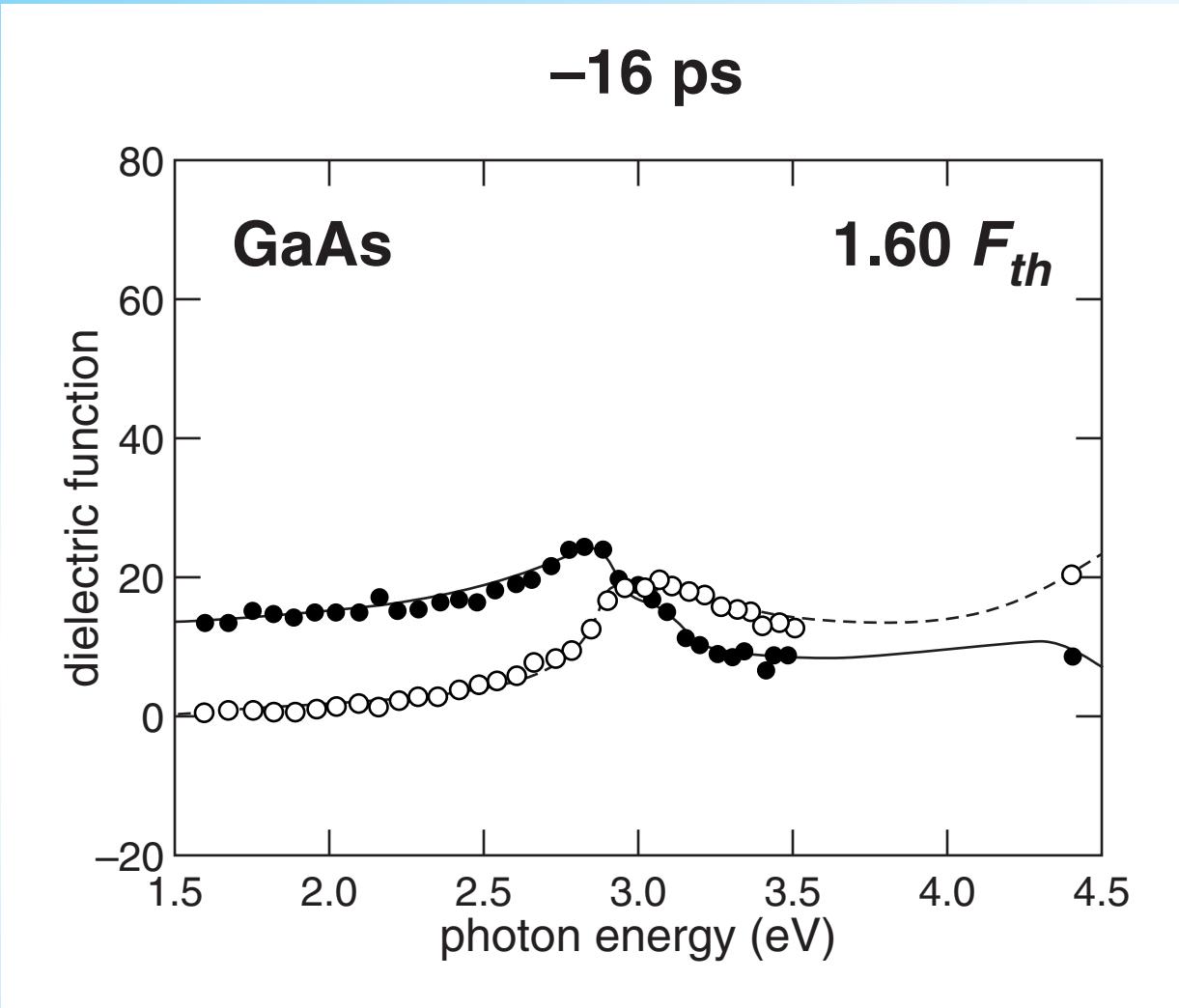
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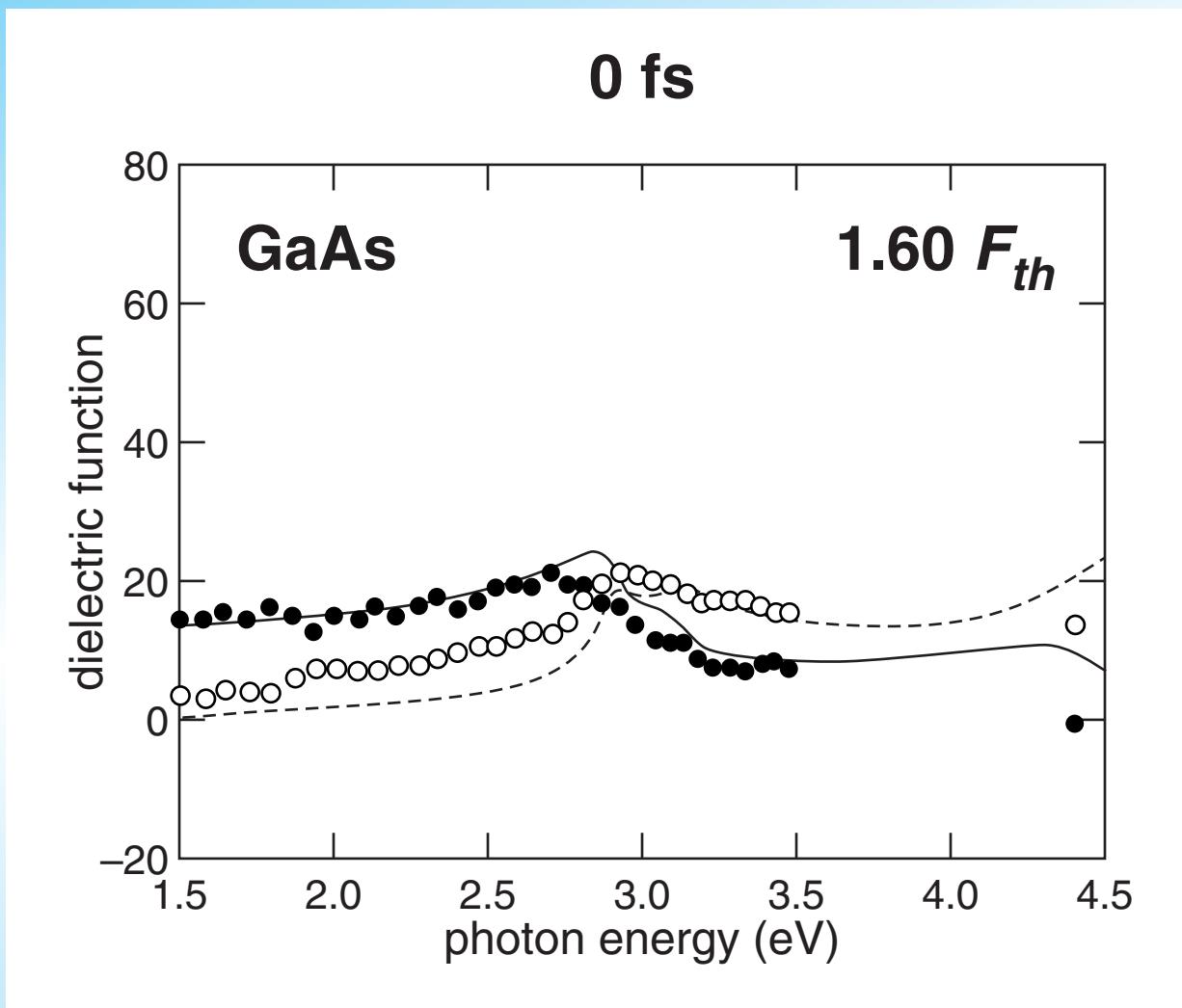
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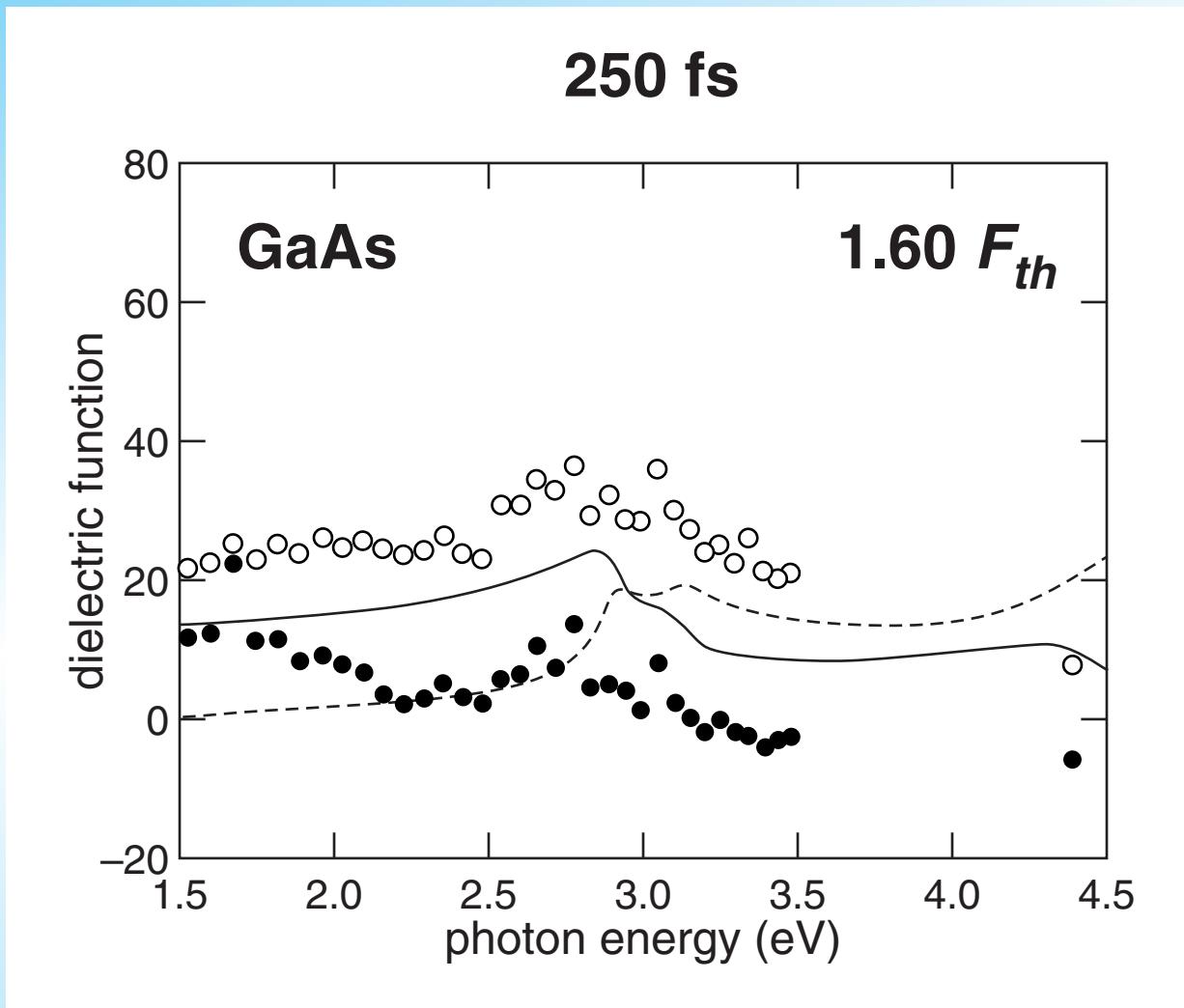
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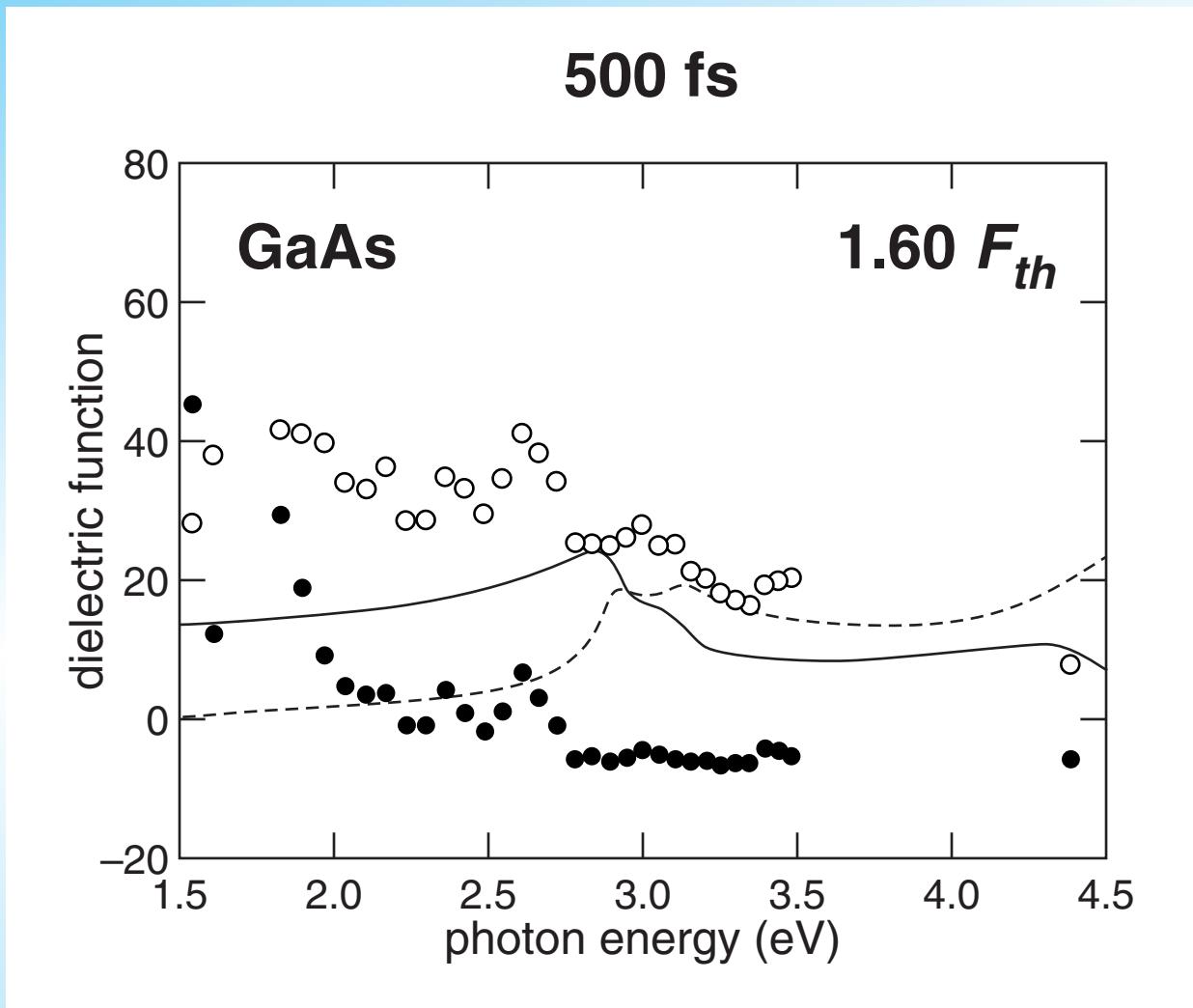
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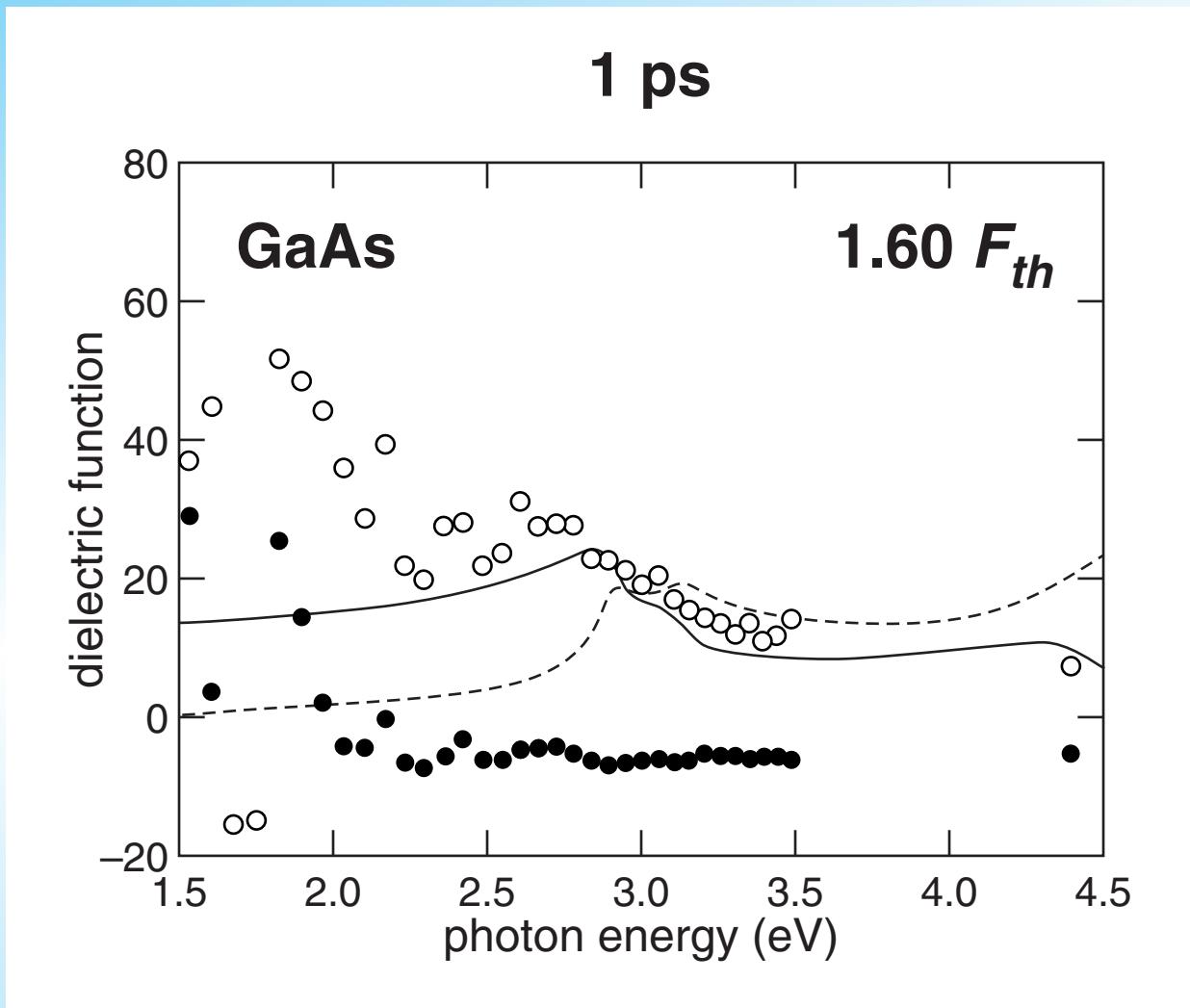
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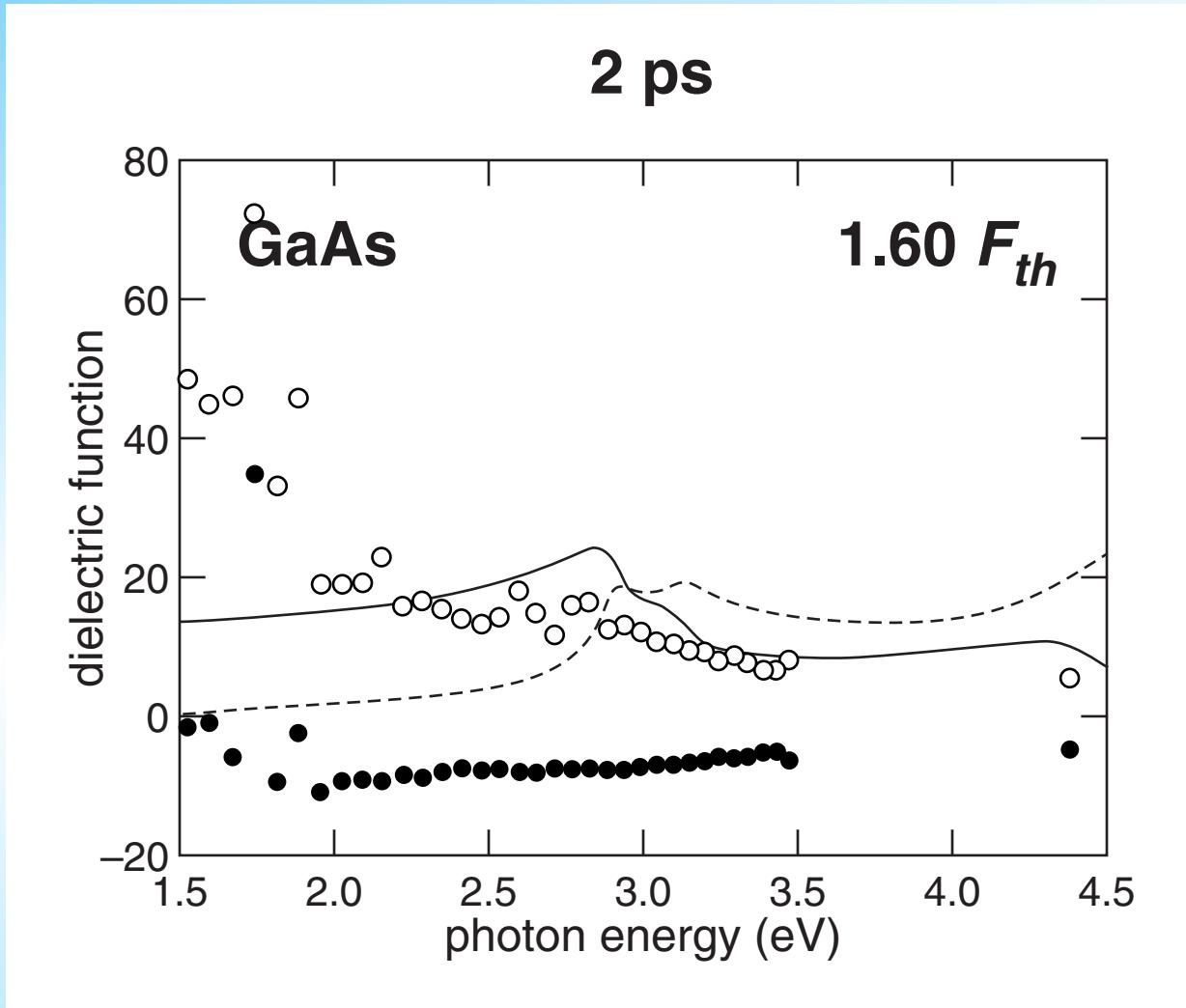
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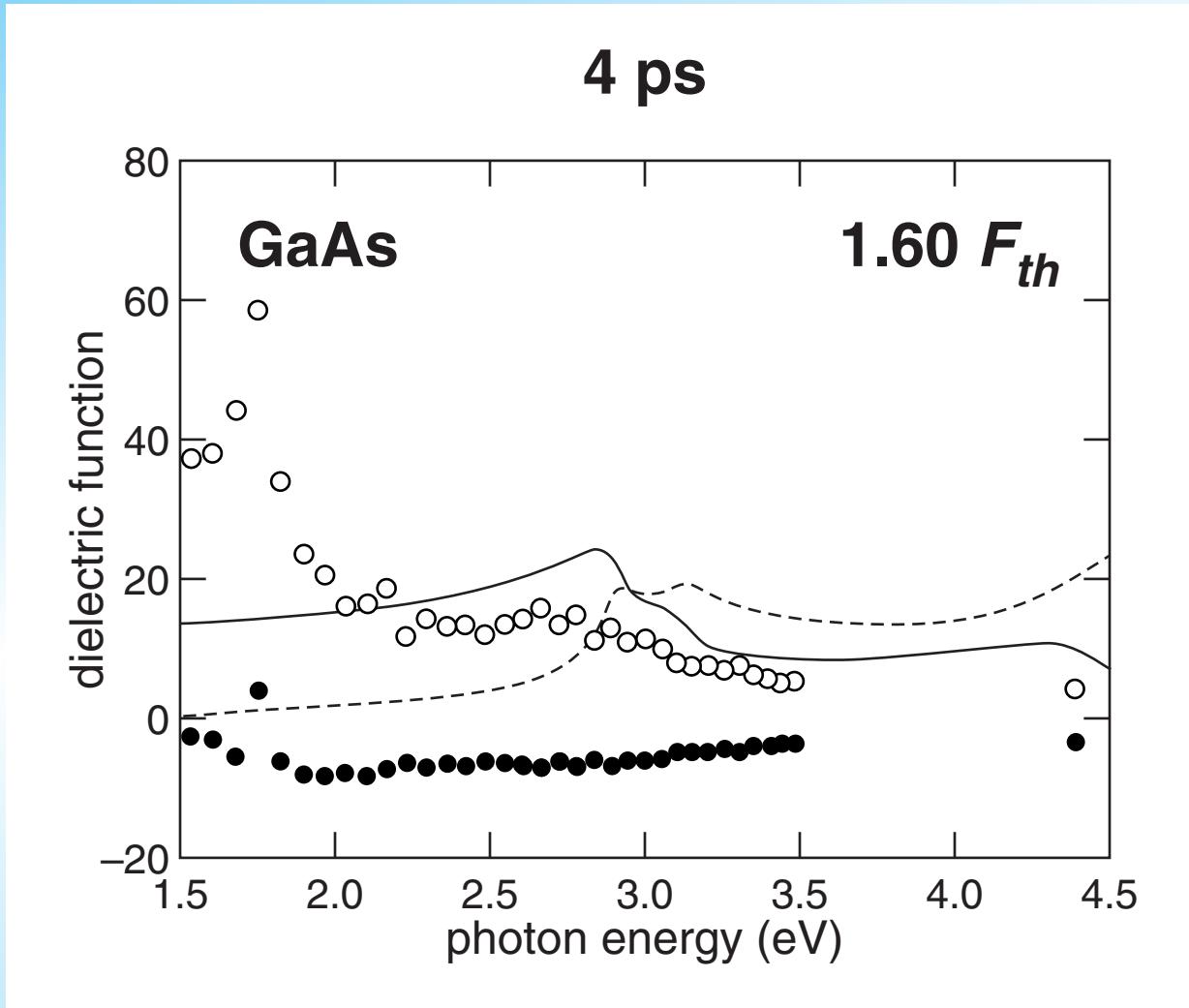
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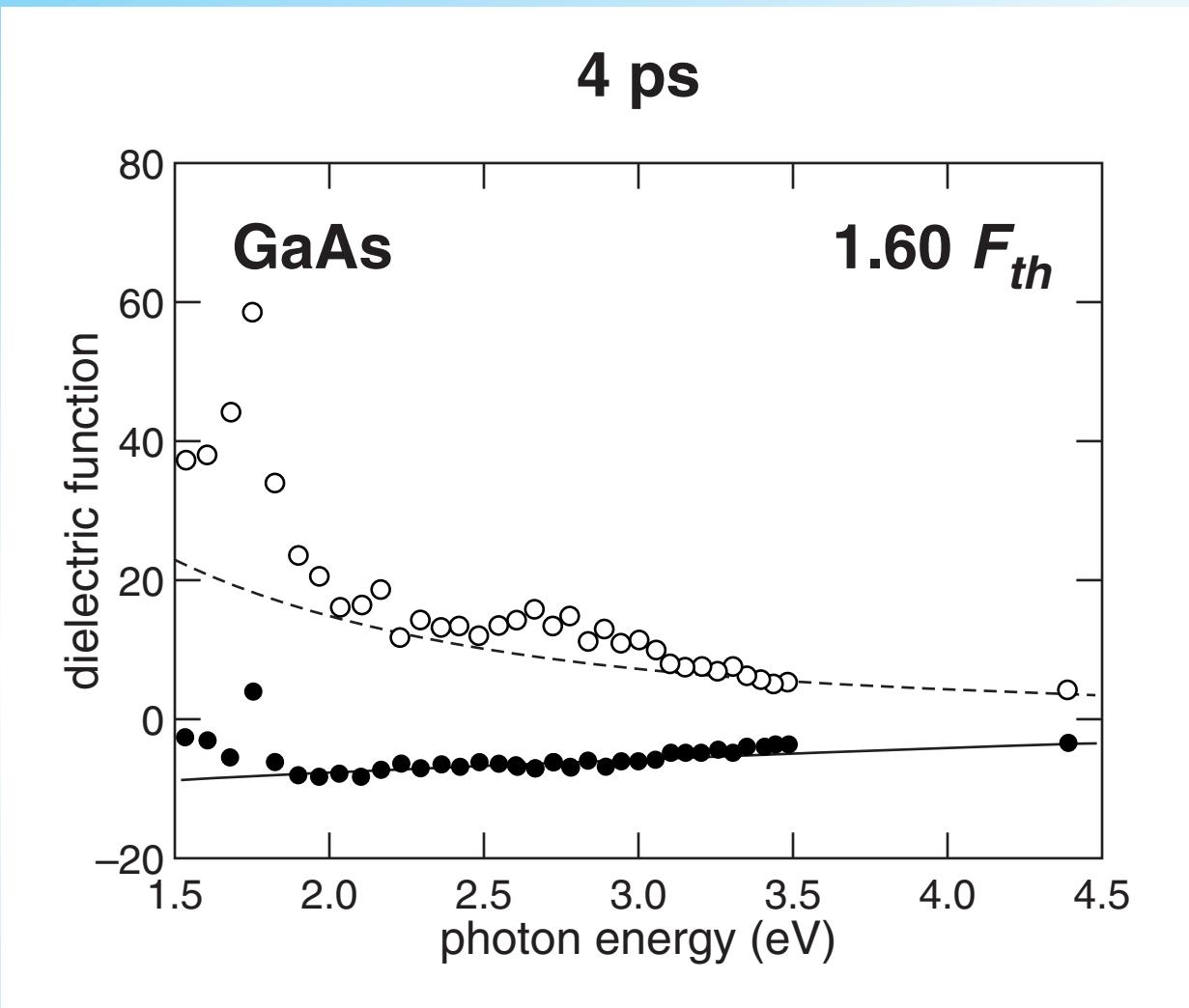
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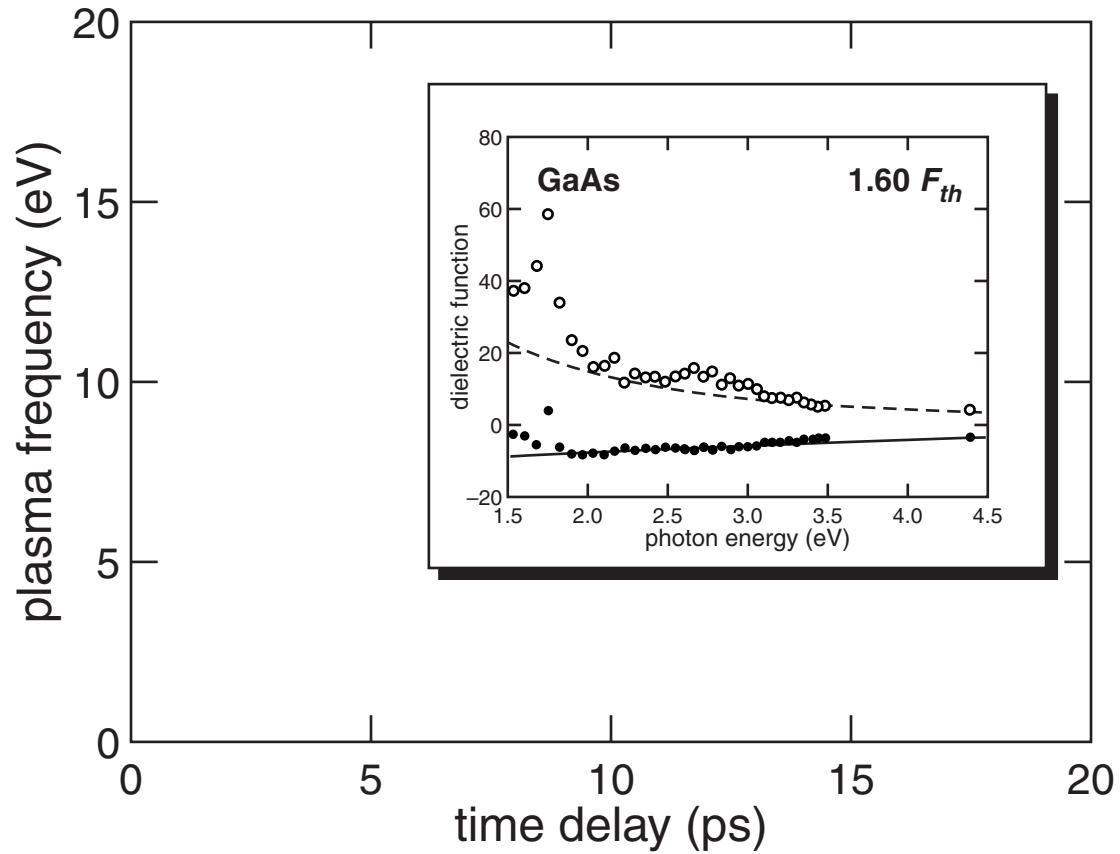
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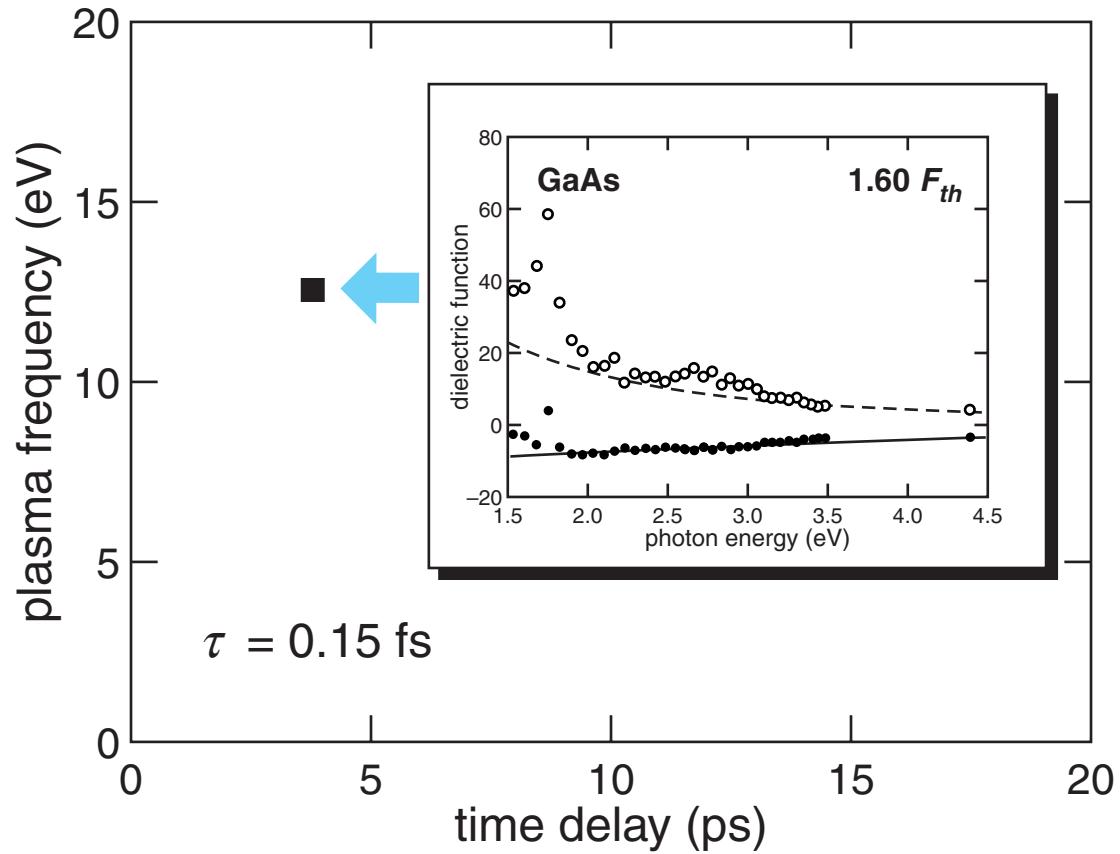
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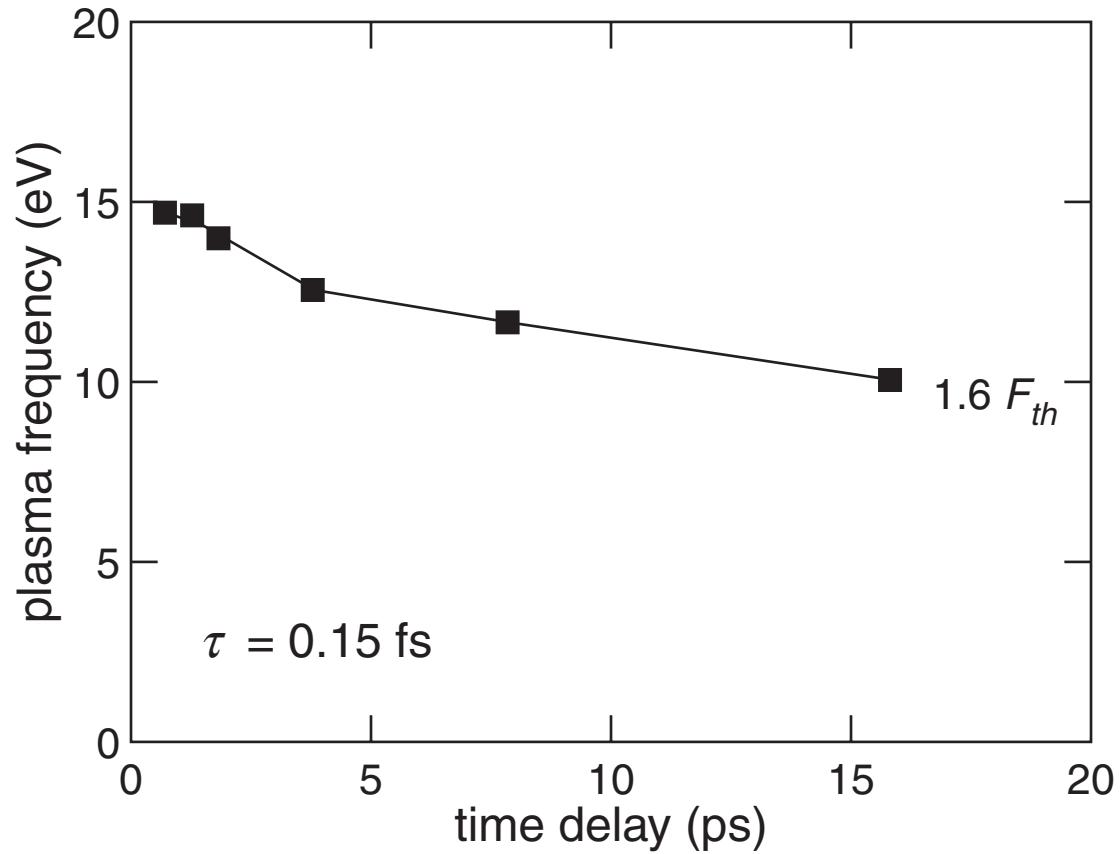
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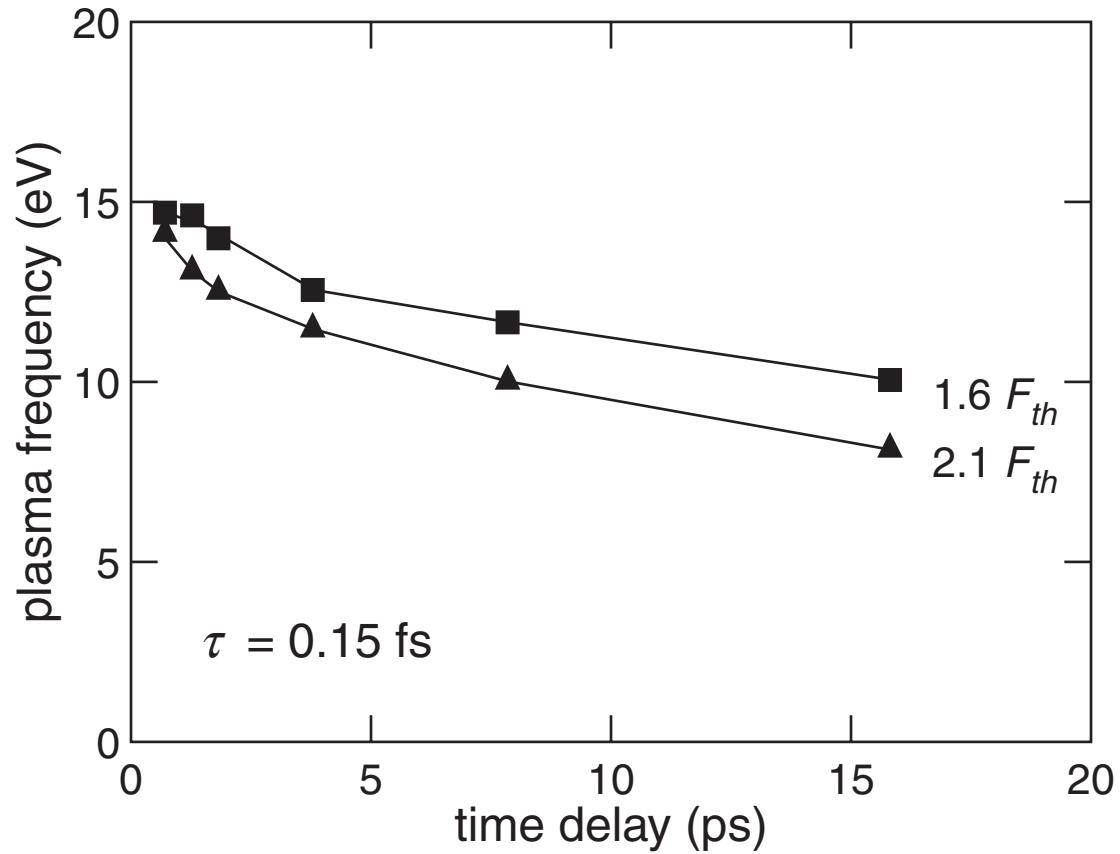
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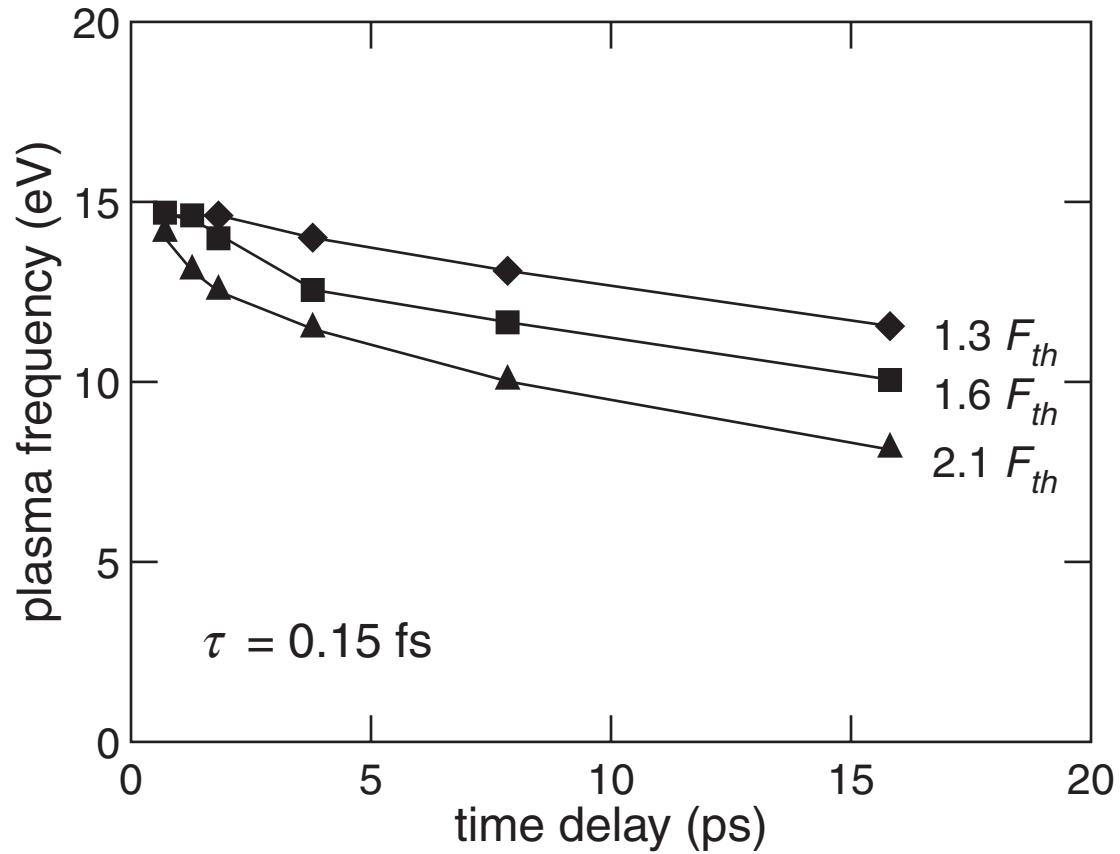
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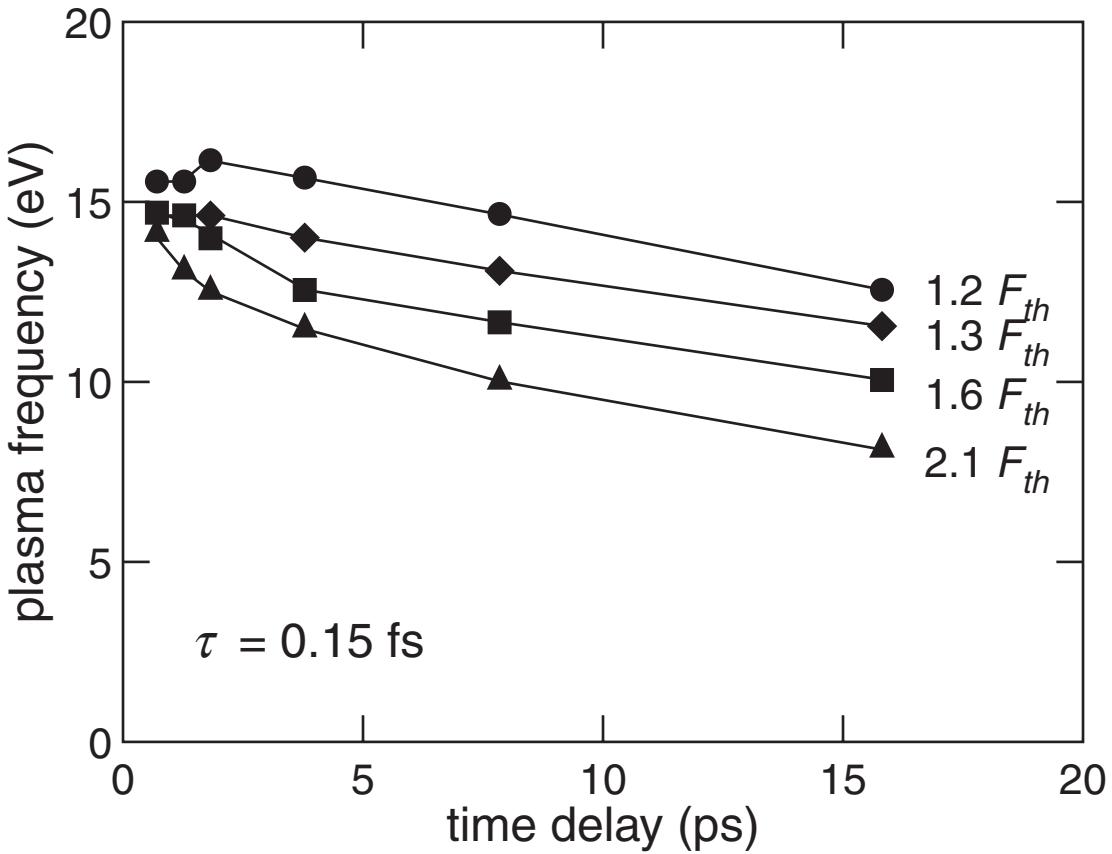
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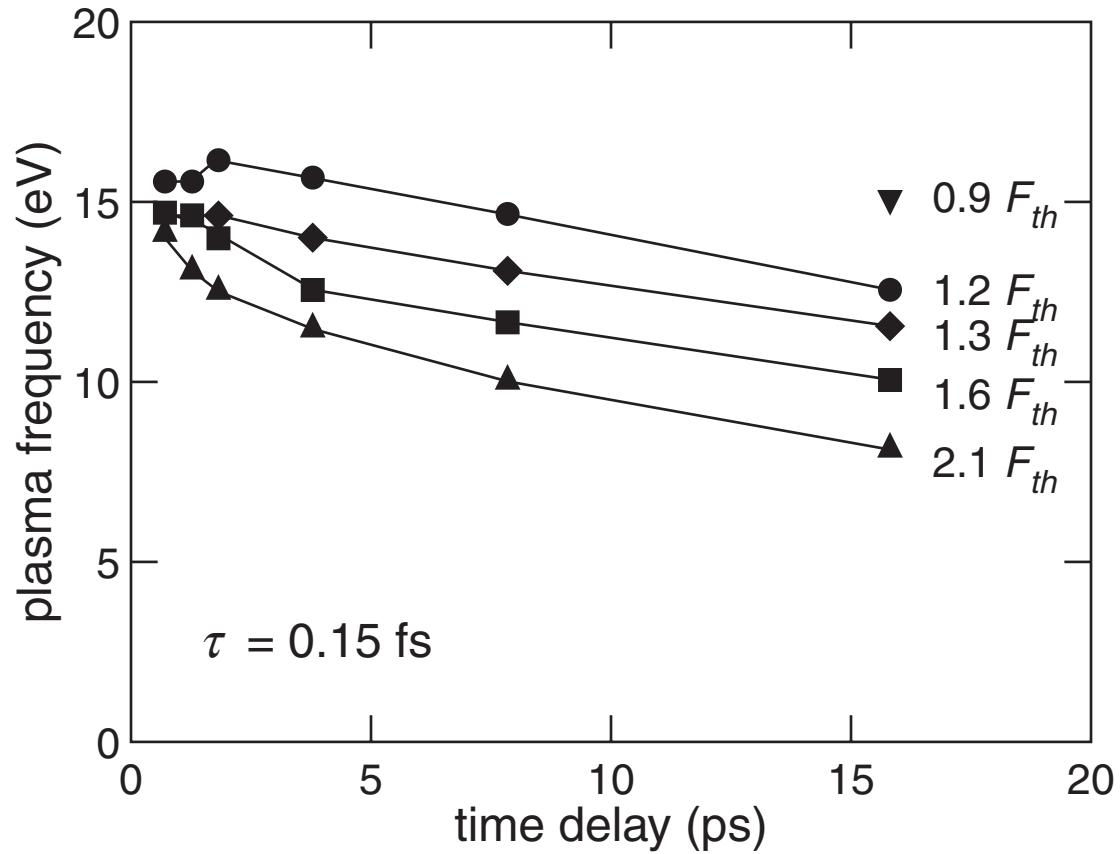
Results

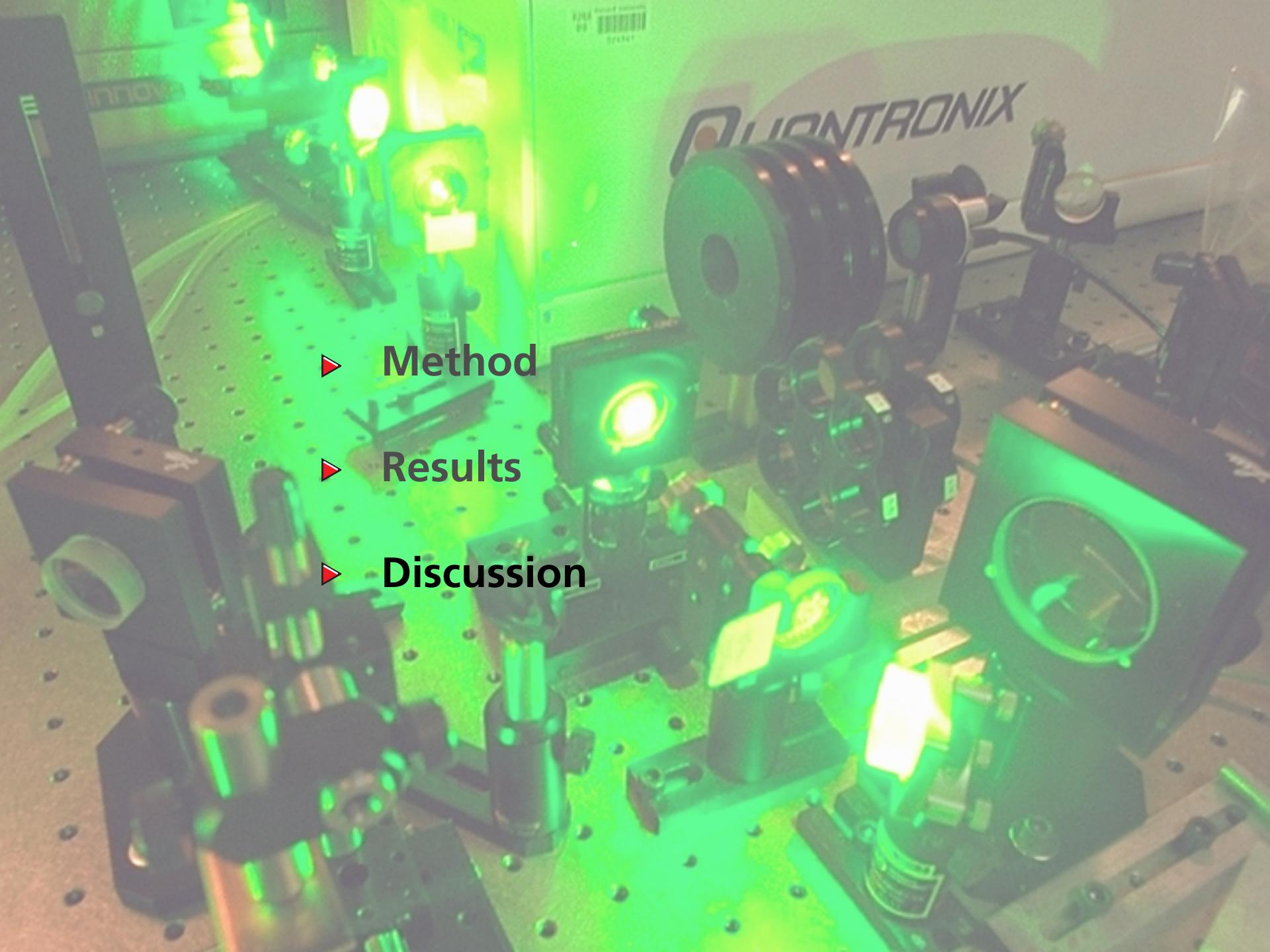


Results



Results

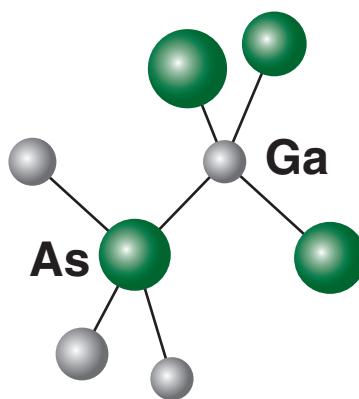


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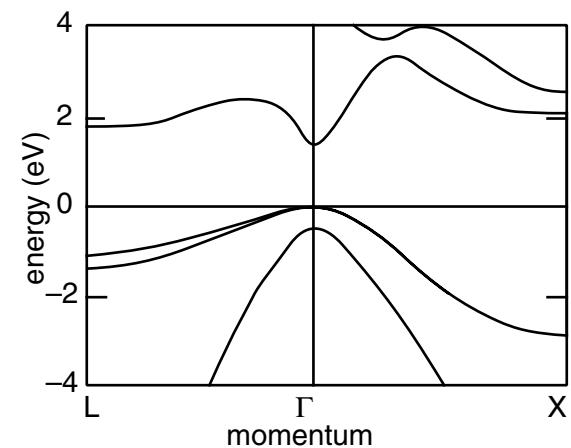
Discussion

short time scale

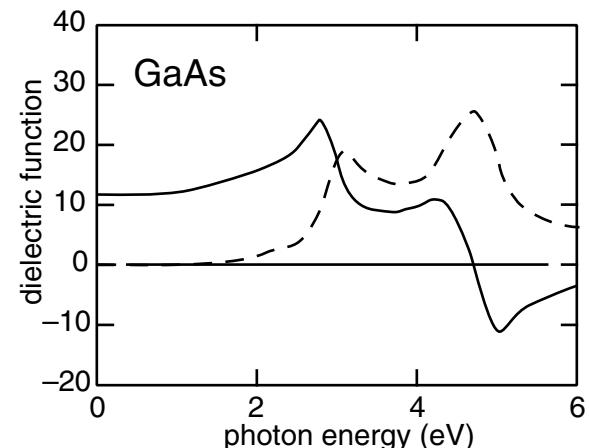
structure



bandstructure



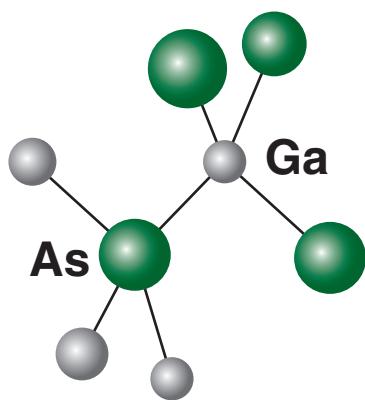
dielectric function



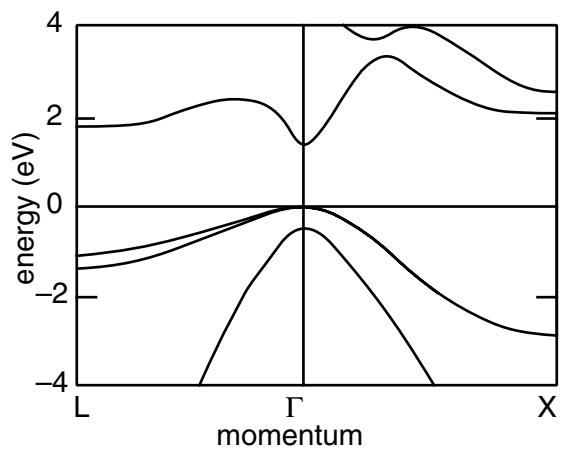
Discussion

short time scale

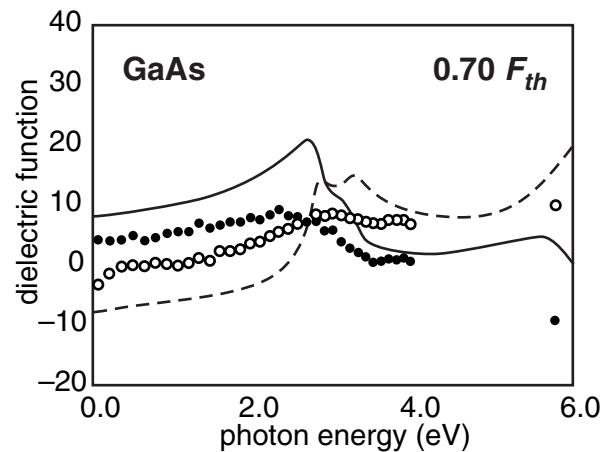
structure



bandstructure



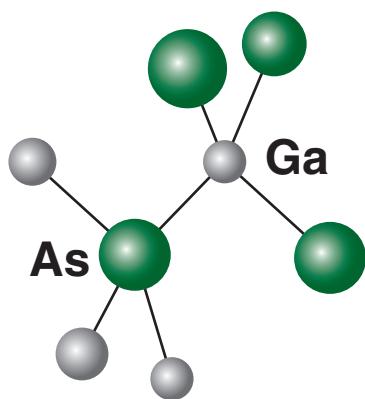
dielectric function



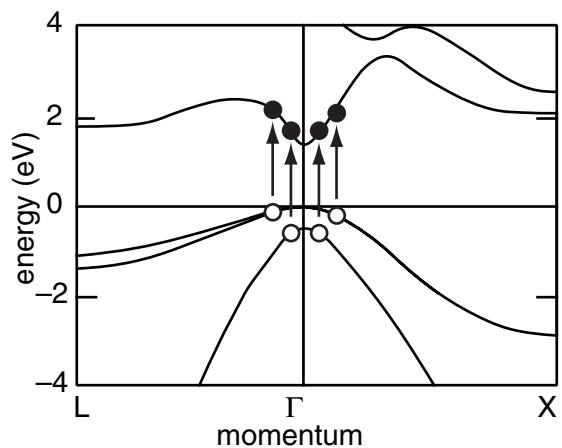
Discussion

short time scale

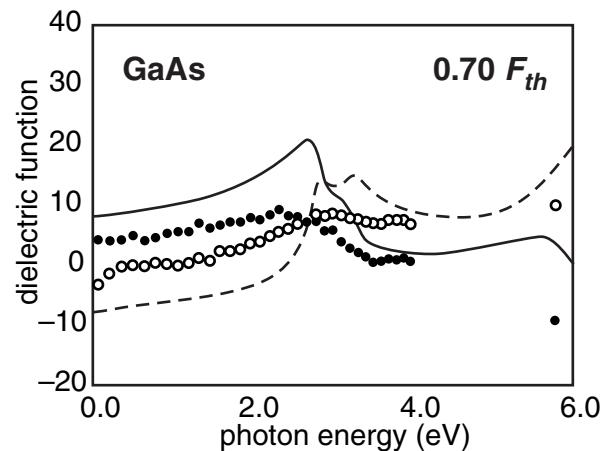
structure



bandstructure



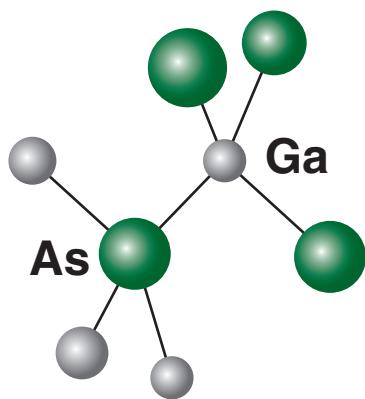
dielectric function



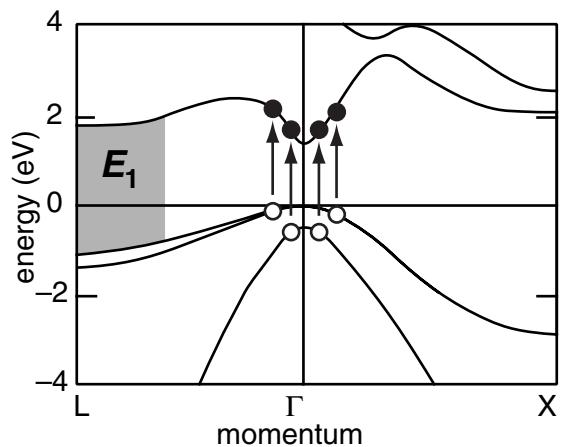
Discussion

short time scale

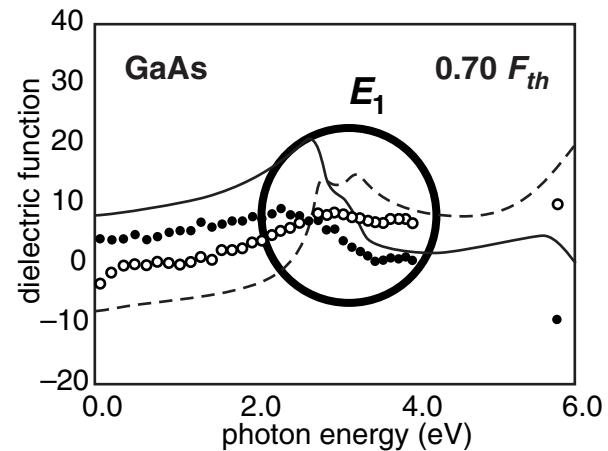
structure



bandstructure



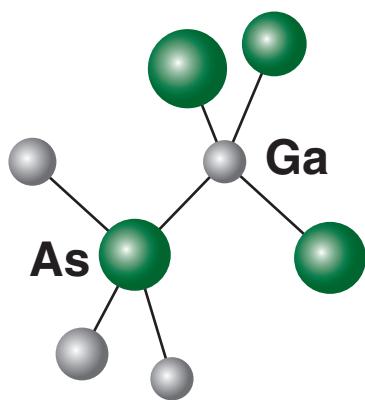
dielectric function



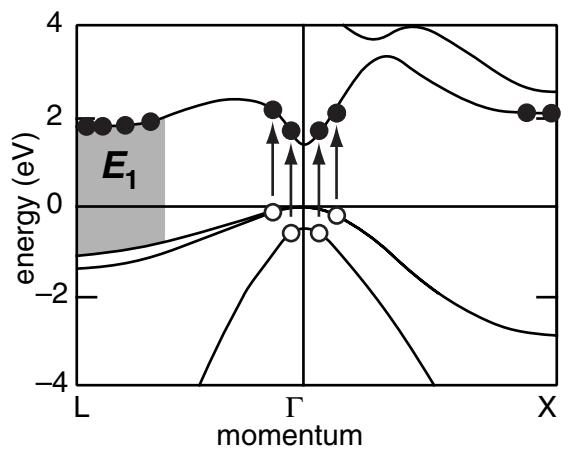
Discussion

short time scale

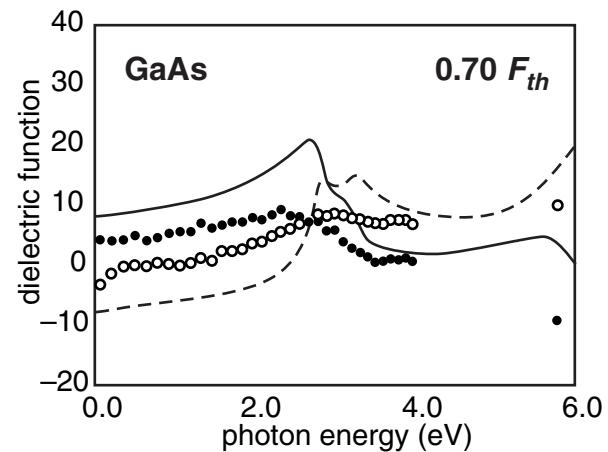
structure



bandstructure



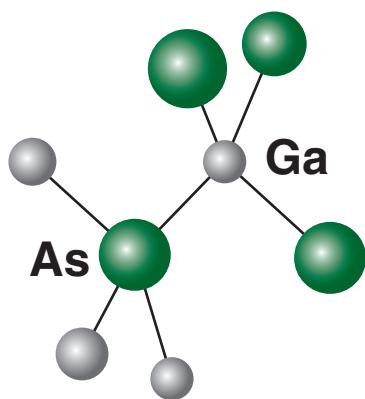
dielectric function



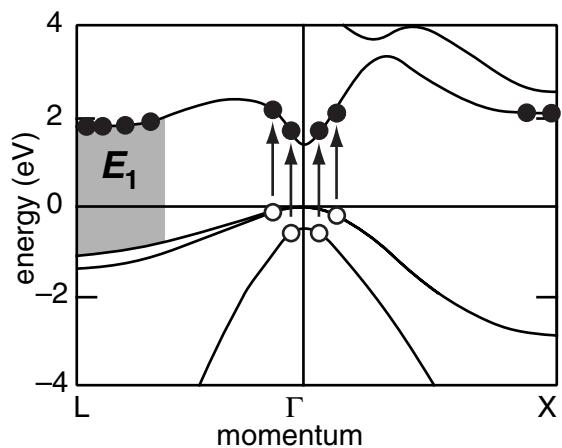
Discussion

short time scale

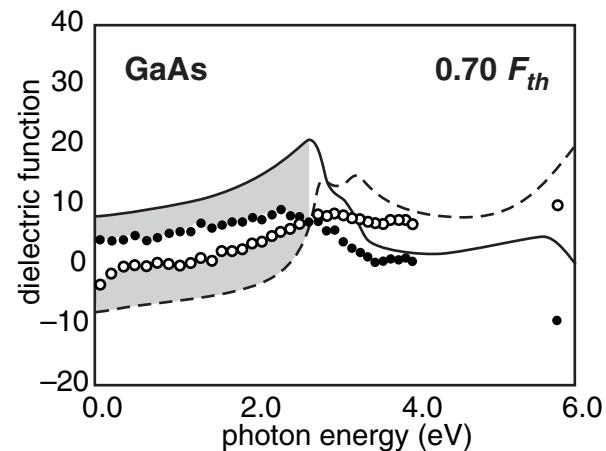
structure



bandstructure



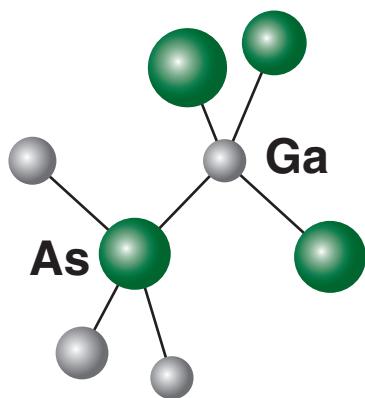
dielectric function



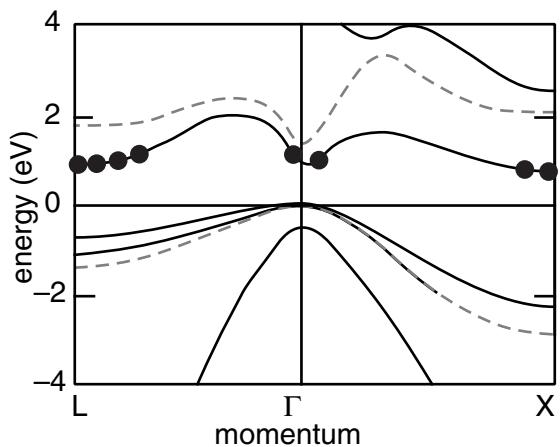
Discussion

short time scale

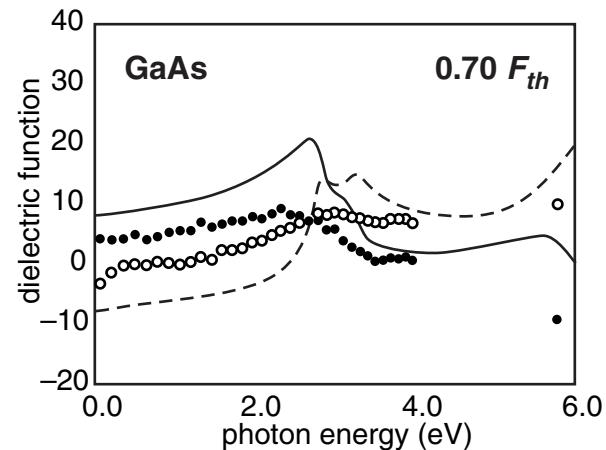
structure



bandstructure



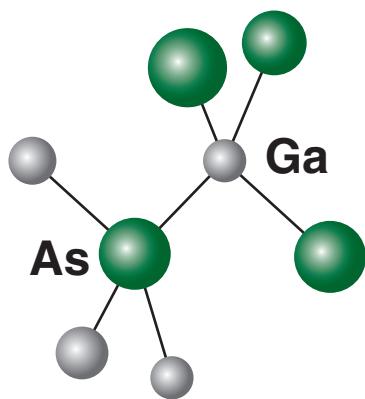
dielectric function



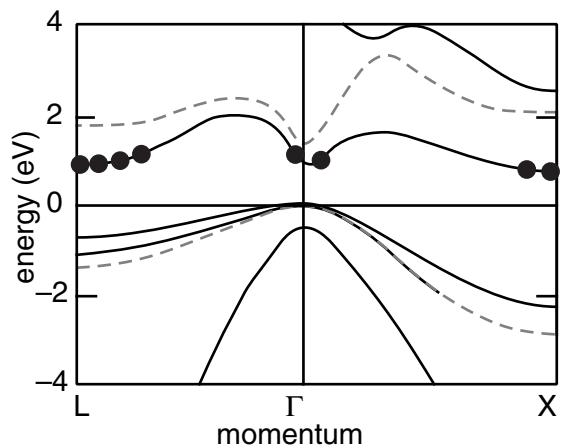
Discussion

short time scale

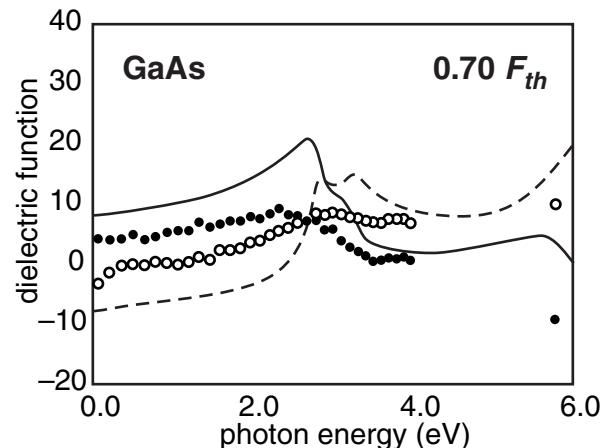
structure



bandstructure



dielectric function

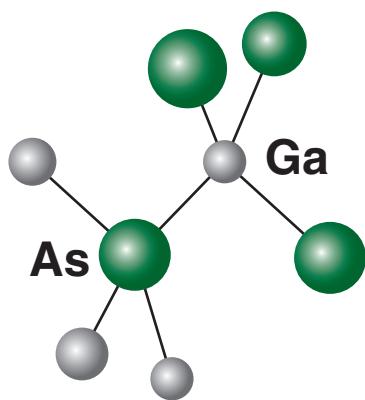


D.H. Kim, et al., Sol. State Comm. 89, 119 (1994)

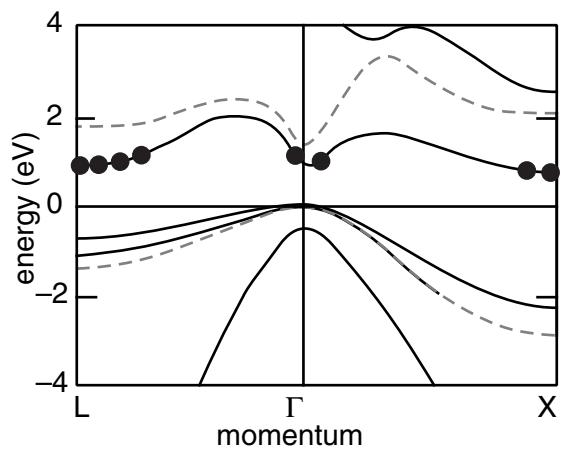
Discussion

short time scale

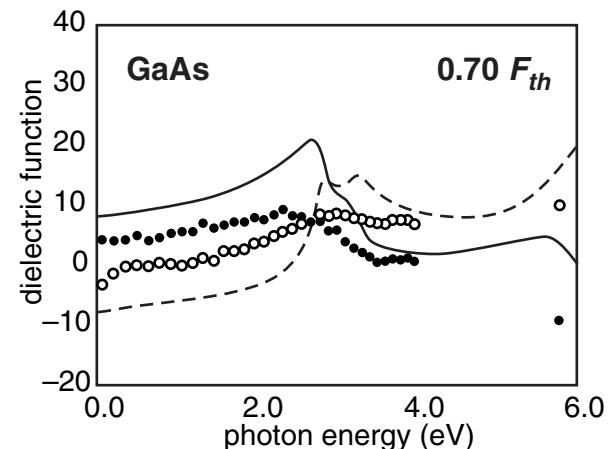
structure



bandstructure



dielectric function

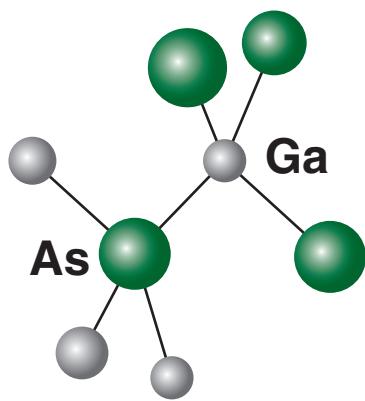


electronic effects dominate at short time scales...

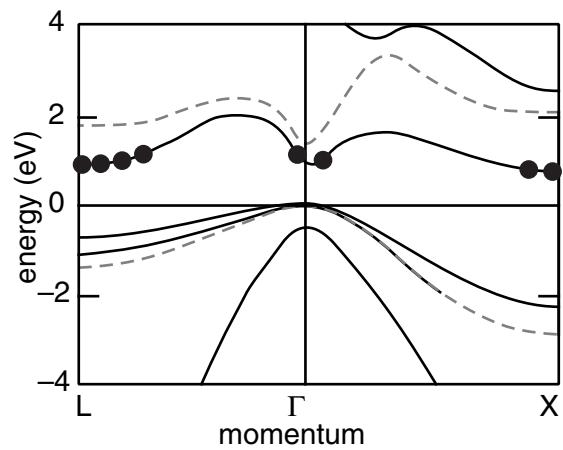
Discussion

short time scale

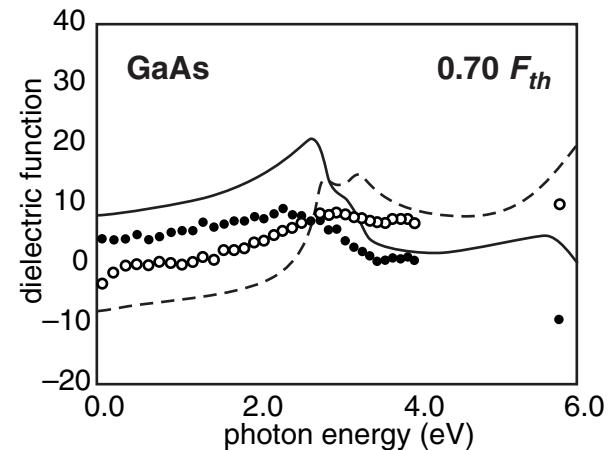
structure



bandstructure



dielectric function

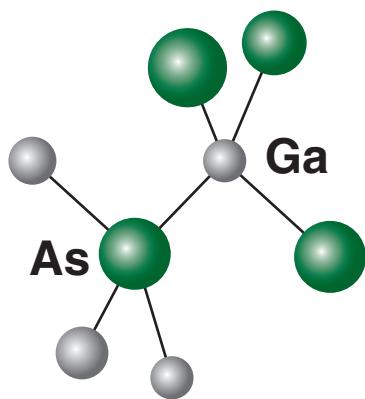


...but they are not as simple as we used to think

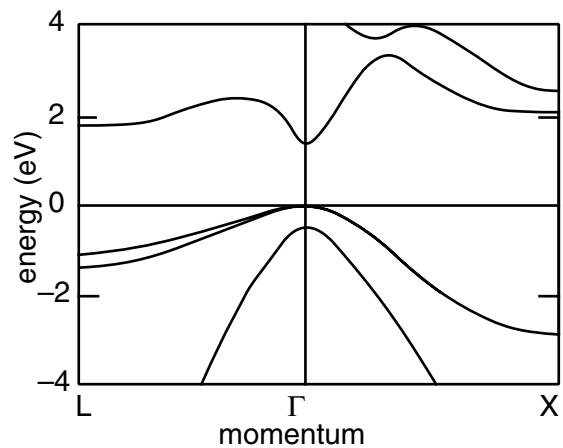
Discussion

long time scale, low fluence

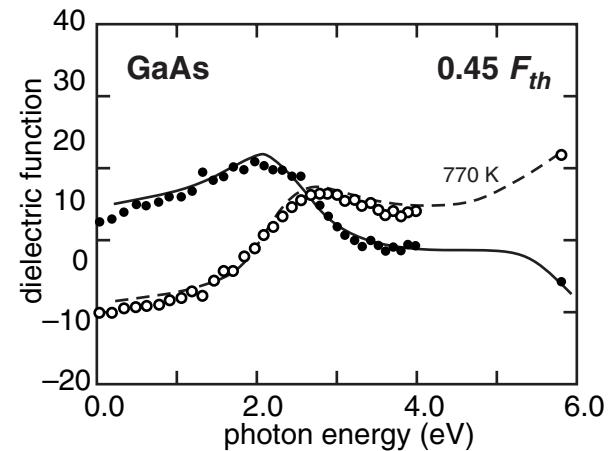
structure



bandstructure



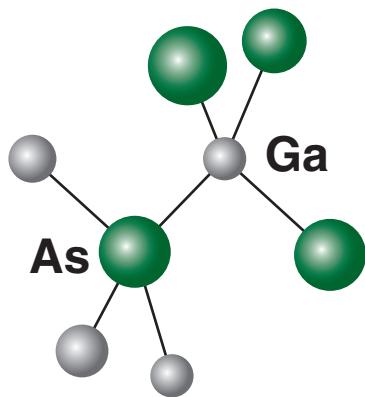
dielectric function



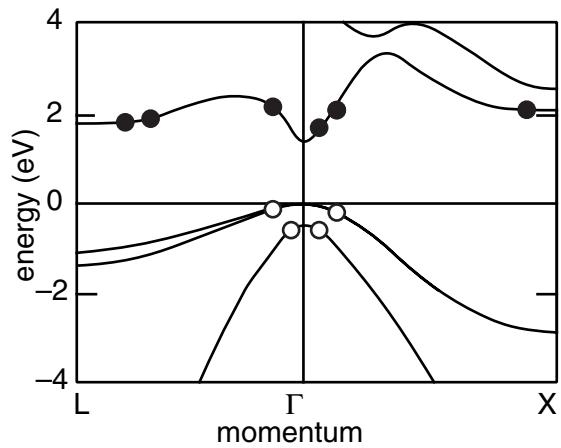
Discussion

long time scale, low fluence

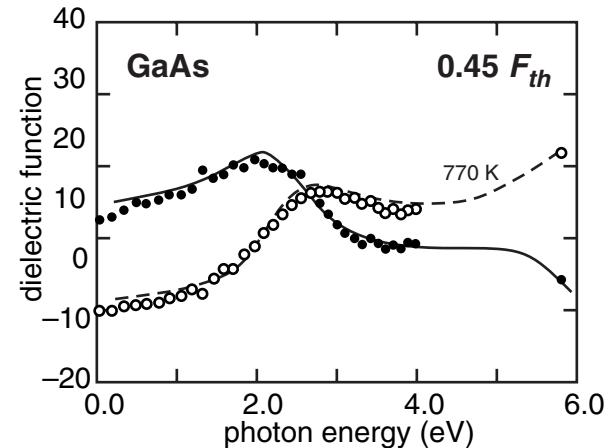
structure



bandstructure



dielectric function

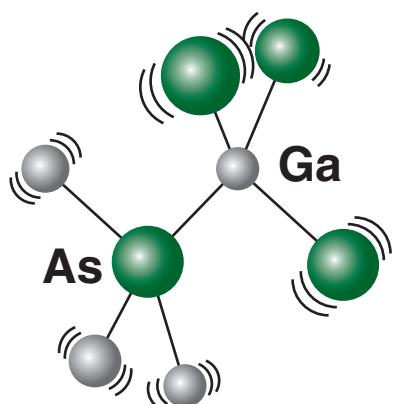


carrier density down, electronic effects subsided...

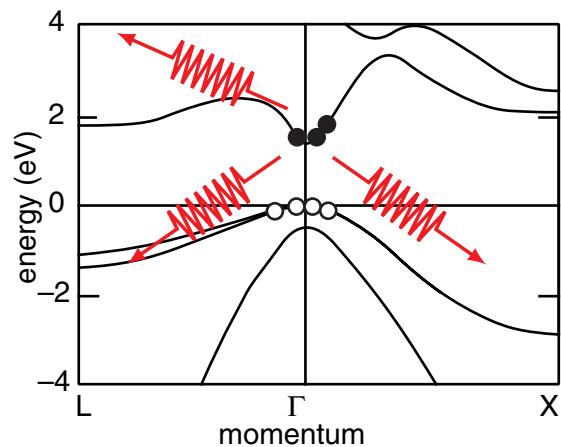
Discussion

long time scale, low fluence

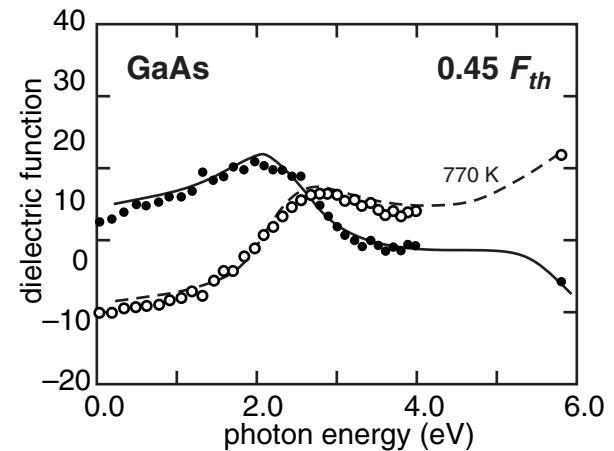
structure



bandstructure



dielectric function

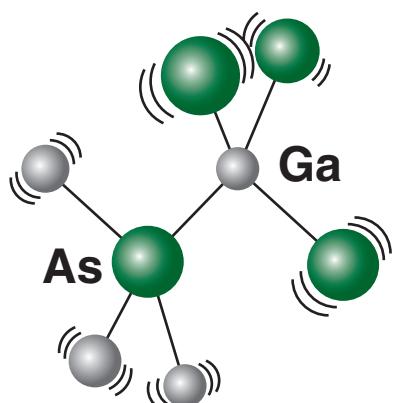


...and lattice heats due to carrier relaxation

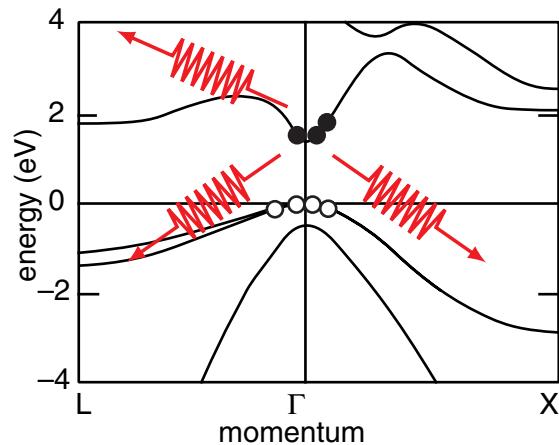
Discussion

long time scale, low fluence

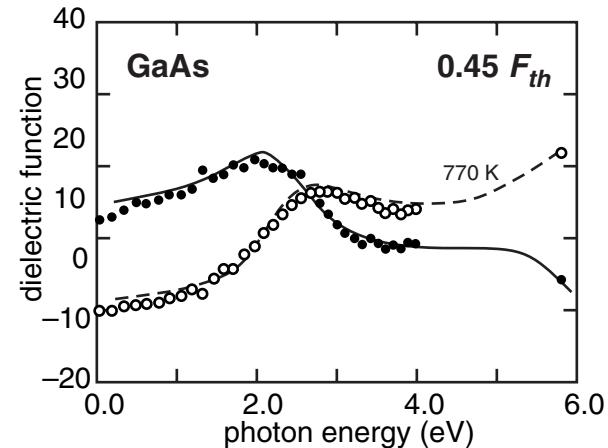
structure



bandstructure



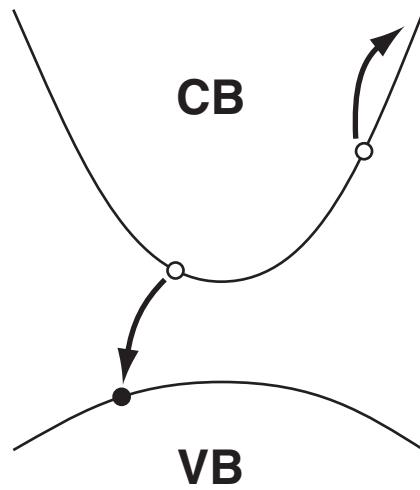
dielectric function



But... why do electronic effects disappear after 2 ps
while lattice heats in 7 ps?

Discussion

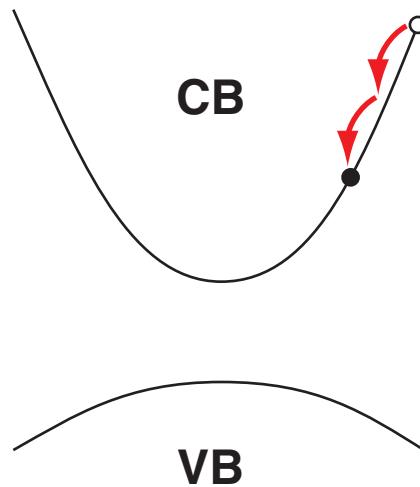
Auger recombination



Auger lowers N without changing E in 2–4 ps...

Discussion

carrier-phonon scattering

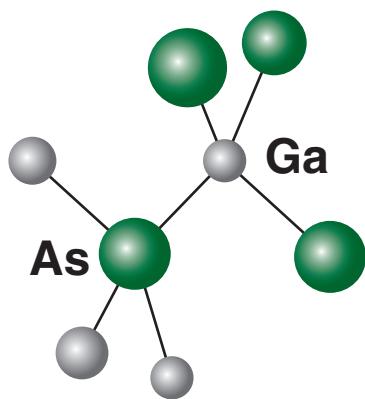


...and highly excited electrons cool to lattice in 7 ps

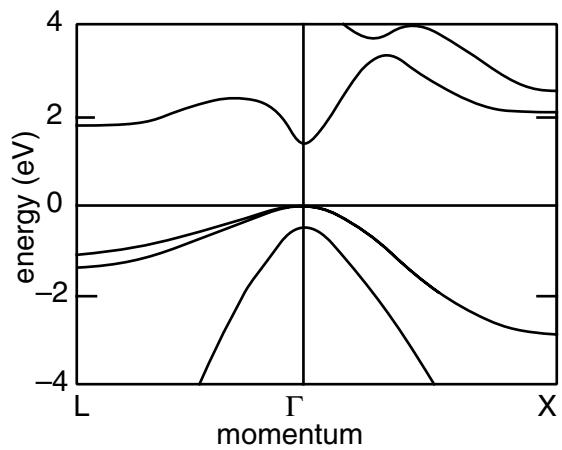
Discussion

long time scale, high fluence

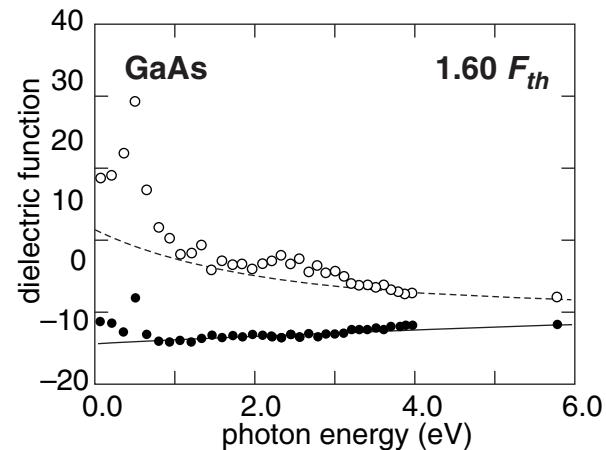
structure



bandstructure



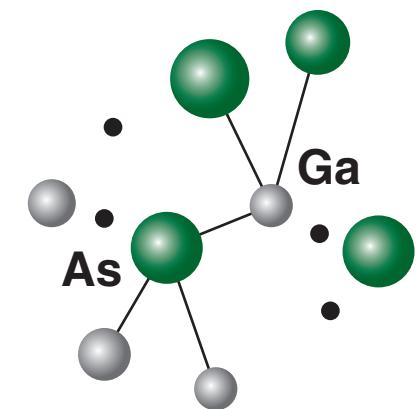
dielectric function



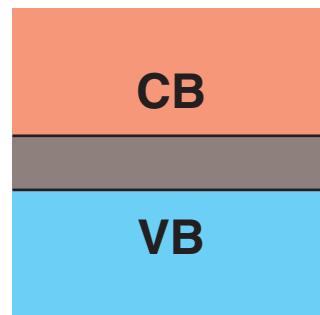
Discussion

long time scale, high fluence

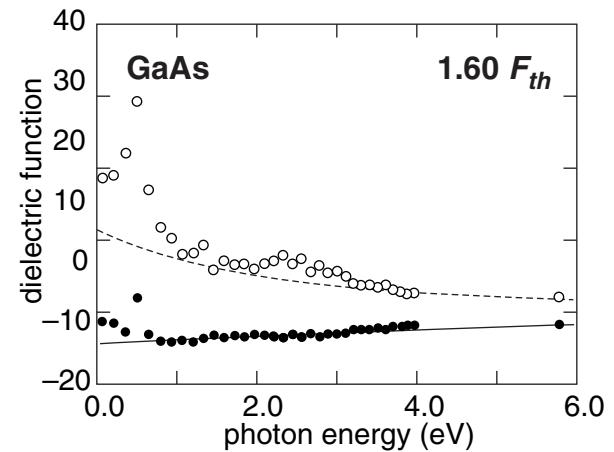
structure



bandstructure

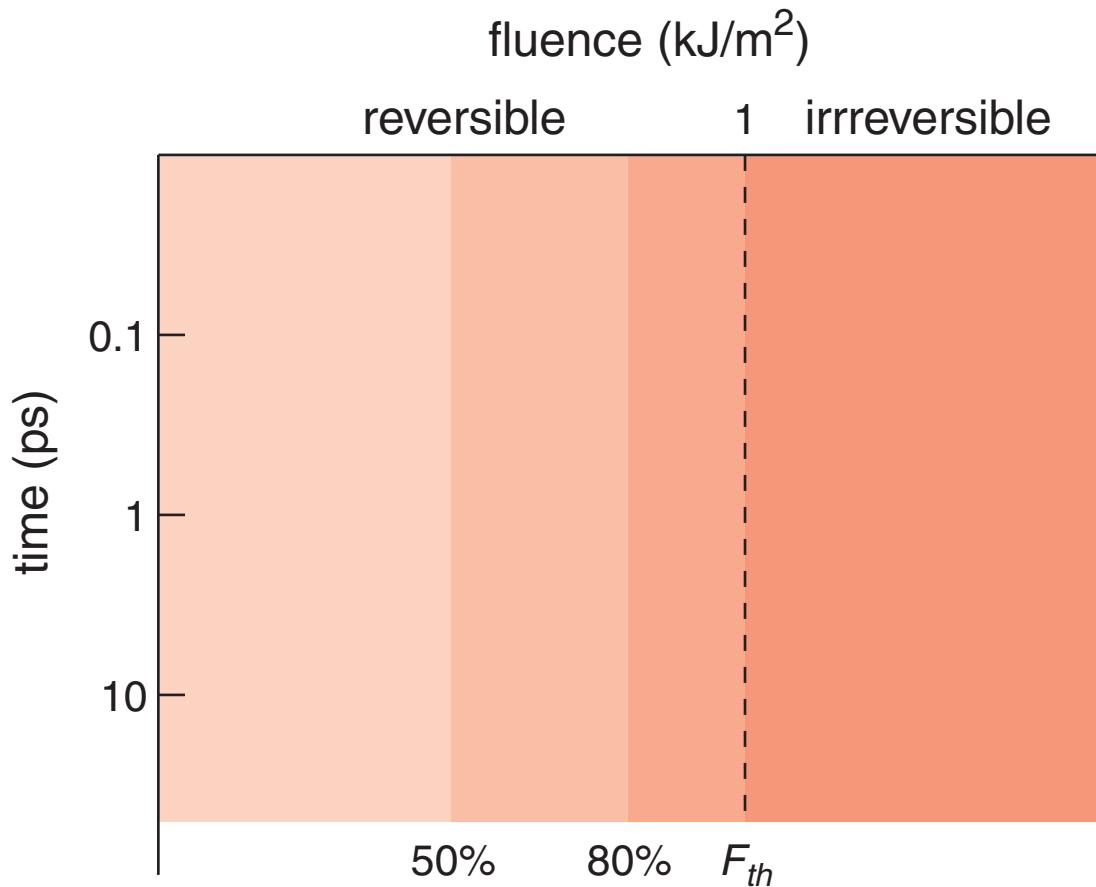


dielectric function

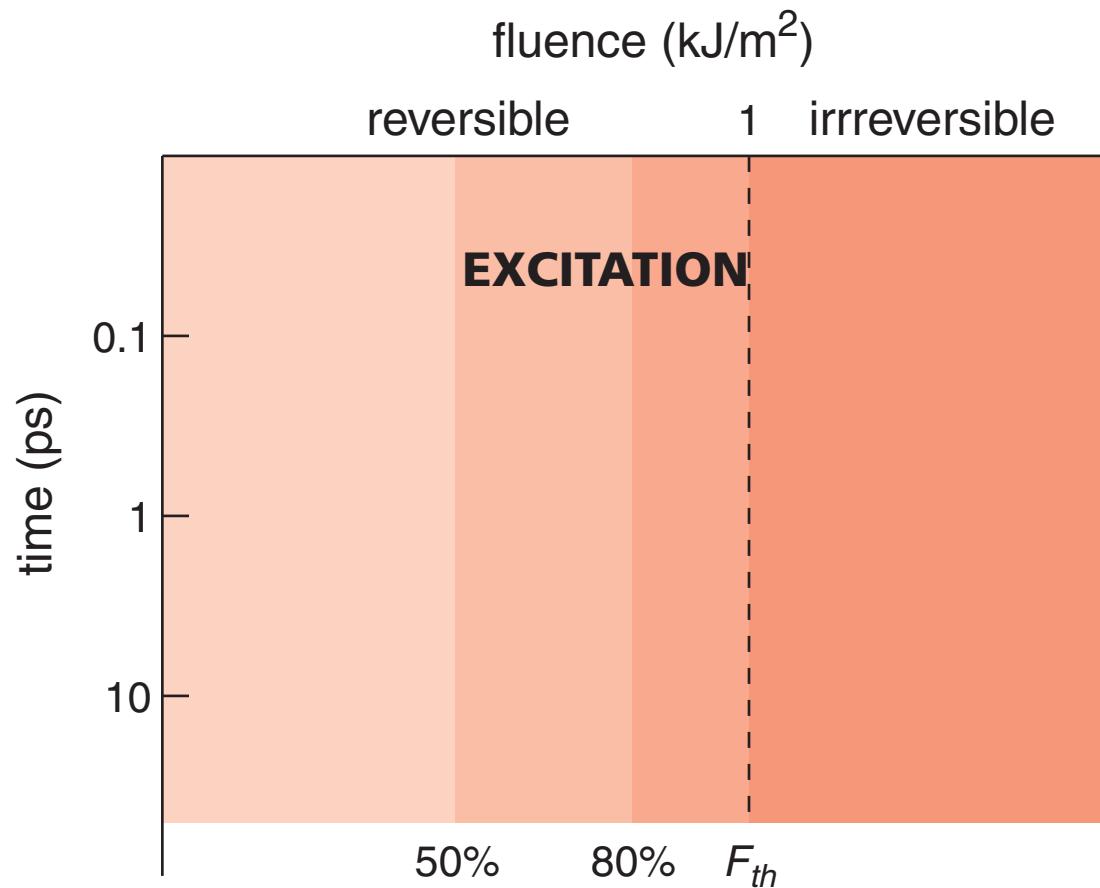


gradual drop in gap → not electronic effect

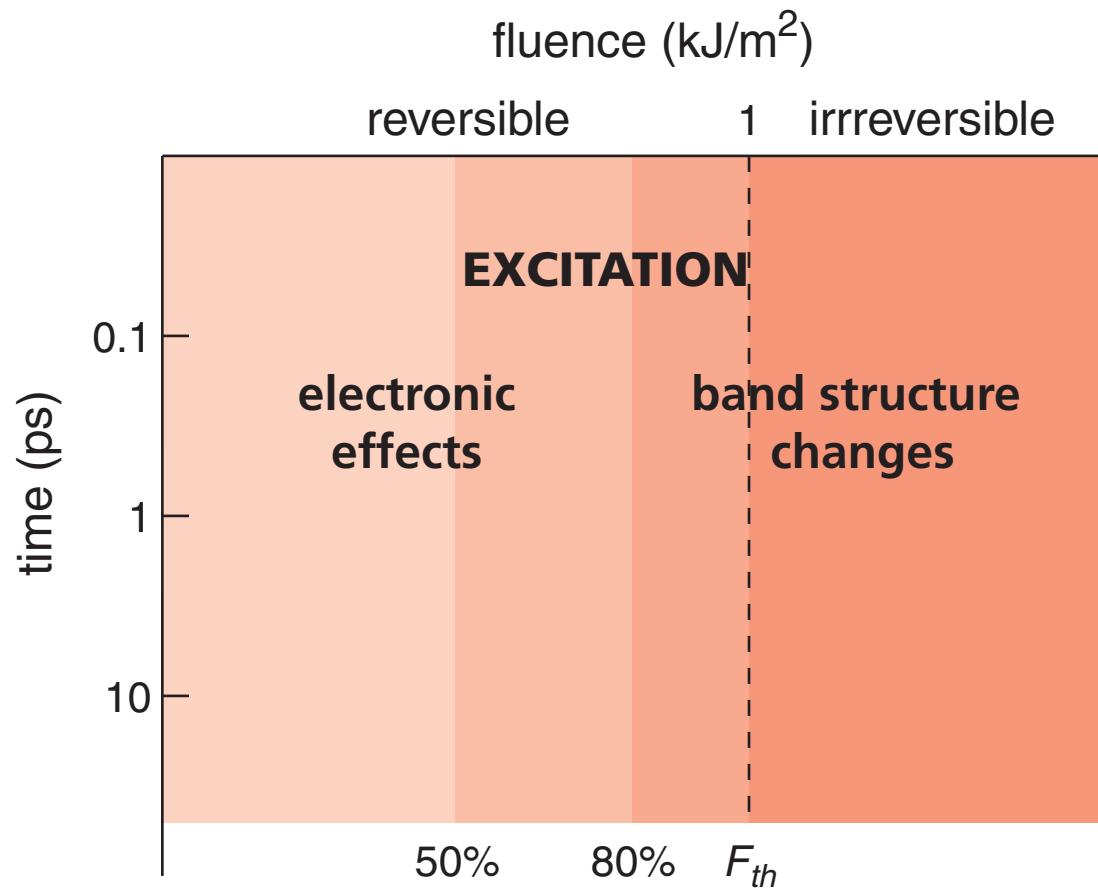
Discussion



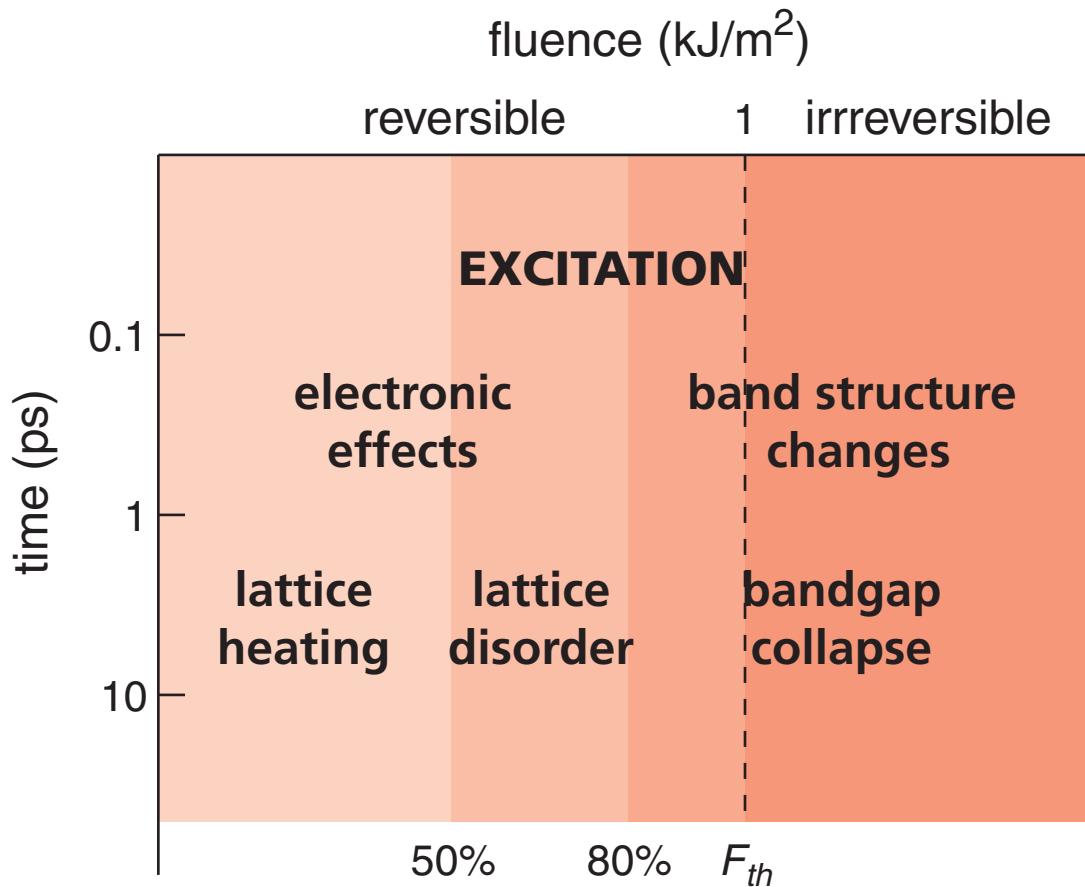
Discussion



Discussion



Discussion



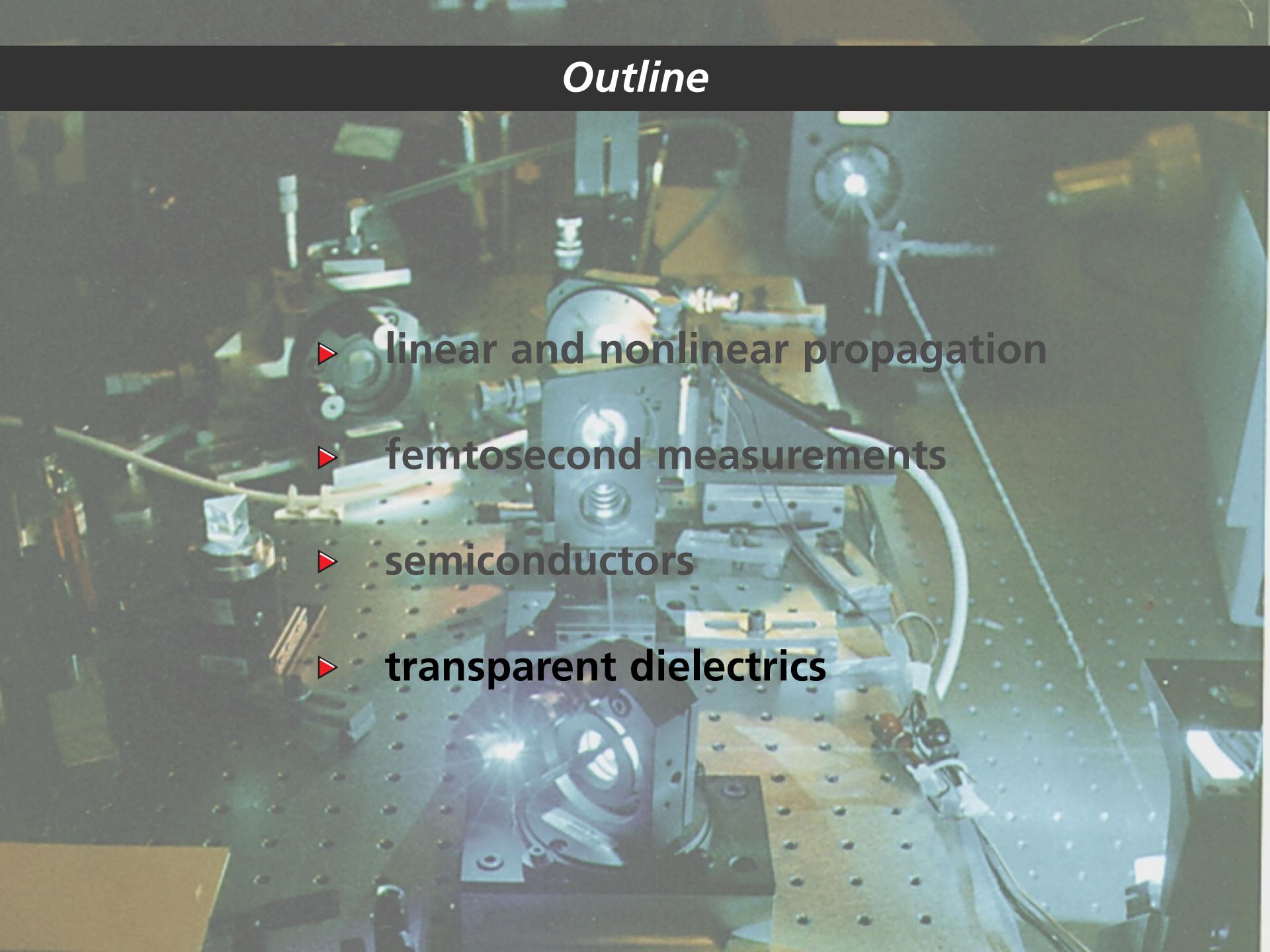
Summary

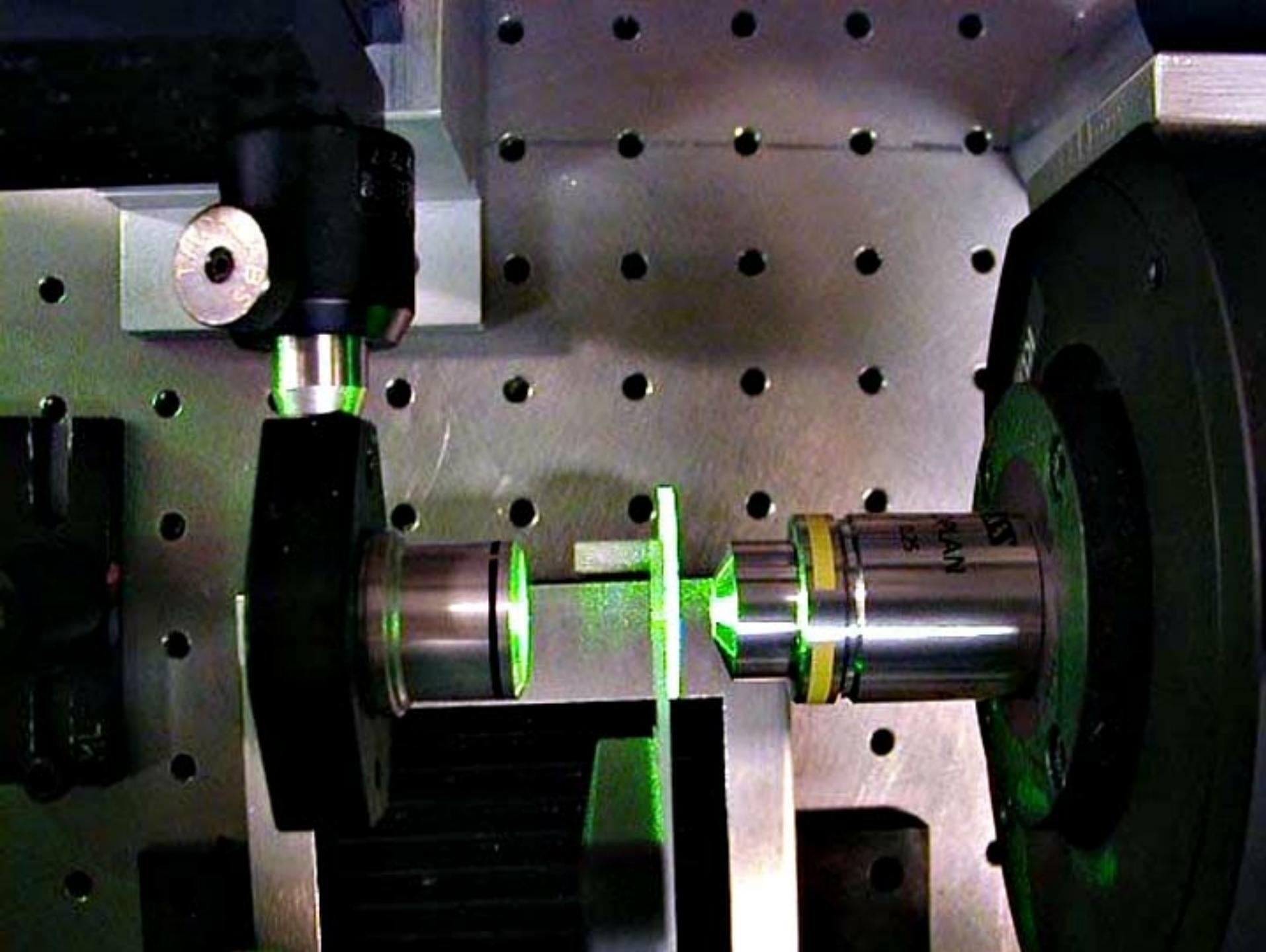
- ▶ **measurement of $\epsilon(\omega)$ identifies ultrafast phase changes**
- ▶ **initial response is electronic, via band structure and electron occupation changes**
- ▶ **structural effects dominate after a few ps**
- ▶ **interesting reversible regime**

Conclusions

- ▶ **strong electronic excitation can drive a structural transition**
- ▶ **femtosecond lasers allow us to see the dynamics of the transition**

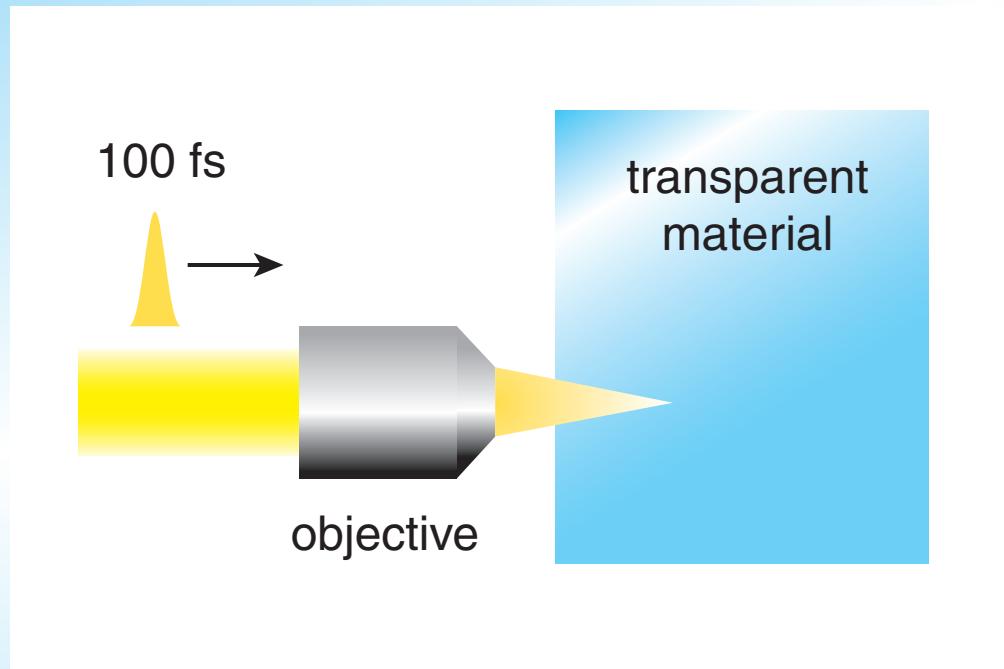
Outline

- 
- ▶ linear and nonlinear propagation
 - ▶ femtosecond measurements
 - ▶ semiconductors
 - ▶ transparent dielectrics



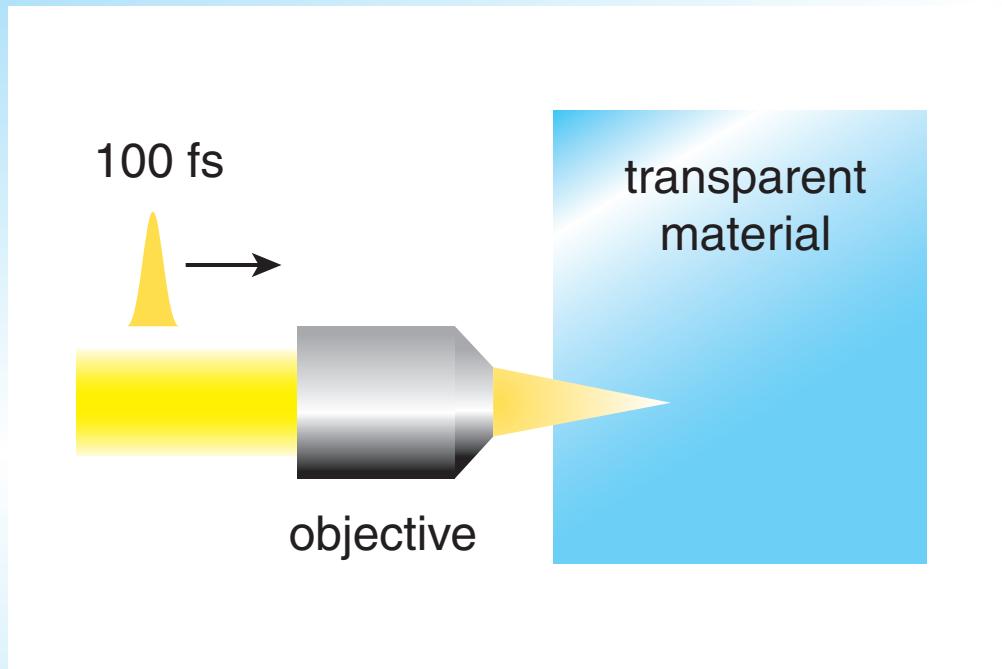
Introduction

focus laser beam inside material...



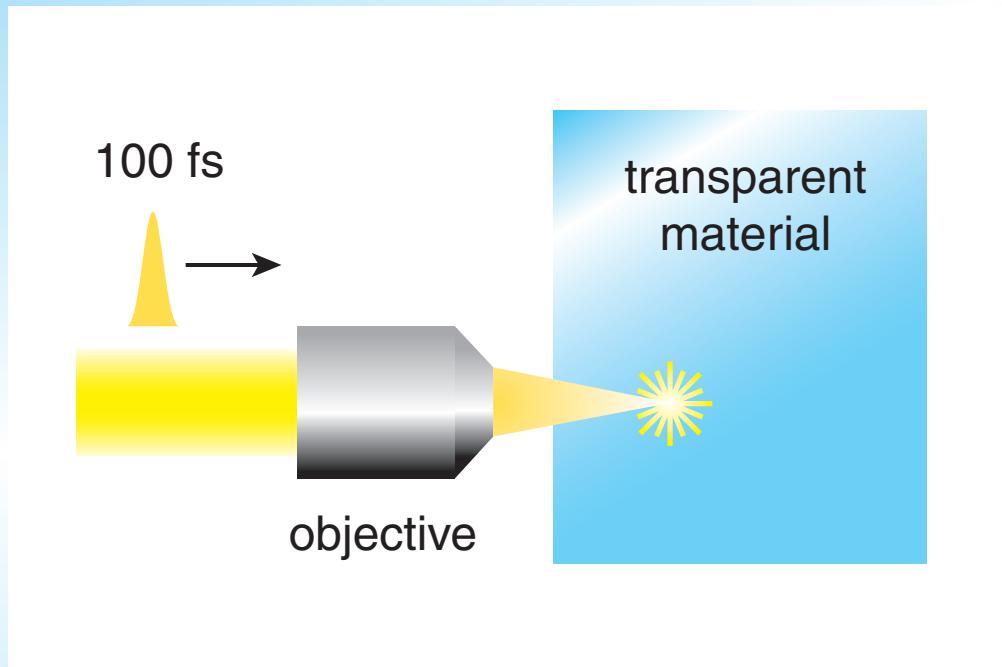
Introduction

high intensity at focus...



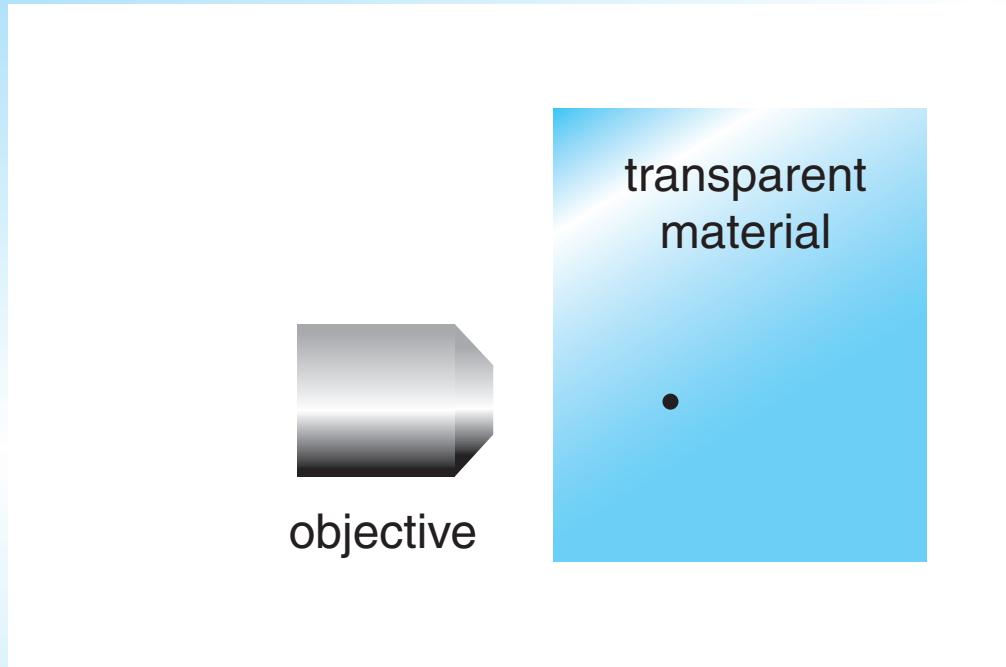
Introduction

... causes nonlinear ionization...



Introduction

and microscopic bulk damage



Glezer, et al., *Opt. Lett.* 21, 2023 (1996)

Introduction

What are the conditions at focus?



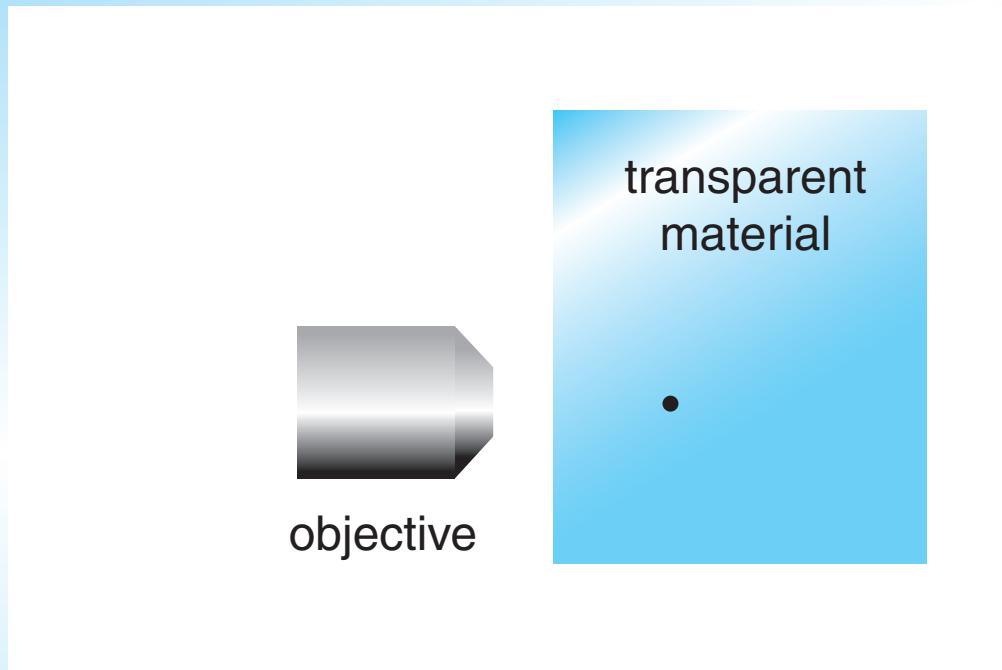
objective

transparent
material

•

Introduction

What are the conditions at focus?



laser deposits energy in $\sim 1 \mu\text{m}^3$

Introduction

What temperature?

Introduction

What temperature?

$$\Delta E = C_V \rho V \Delta T$$

Introduction

What temperature?

$$\Delta E = C_V \rho V \Delta T$$

$$C_V = 0.75 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$$

$$\rho = 2.2 \times 10^3 \text{ kg/m}^3$$

Introduction

What temperature?

$$\Delta E = C_V \rho V \Delta T$$

$$C_V = 0.75 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$$

$$\rho = 2.2 \times 10^3 \text{ kg/m}^3$$

So, 1 μJ in 1 μm^3 gives

$\sim 1,000,000 \text{ K!}$

Introduction

What pressure?

Introduction

What pressure?

Treat ionized material as an ideal gas:

$$pV = nRT$$

Introduction

What pressure?

Treat ionized material as an ideal gas:

$$pV = nRT$$

Gives

$$p = 10 \text{ MBar!}$$

Introduction

So:

microexplosion

$$T \approx 1 \text{ MK}$$

$$p \approx 10 \text{ MBar}$$

$$\rho \quad 2.2 \times 10^3 \text{ kg/m}^3$$

Introduction

So:

	microexplosion	sun
T	$\approx 1 \text{ MK}$	$2\text{--}15 \text{ MK}$
p	$\approx 10 \text{ MBar}$	
ρ	$2.2 \times 10^3 \text{ kg/m}^3$	$0.15\text{--}150 \times 10^3 \text{ kg/m}^3$

Introduction

So:

	microexplosion	sun
T	$\approx 1 \text{ MK}$	$2\text{--}15 \text{ MK}$
p	$\approx 10 \text{ MBar}$	
ρ	$2.2 \times 10^3 \text{ kg/m}^3$	$0.15\text{--}150 \times 10^3 \text{ kg/m}^3$

creating stellar conditions in lab!

Outline

- ▶ Post-mortem analysis
- ▶ Energy deposition
- ▶ Microexplosion dynamics

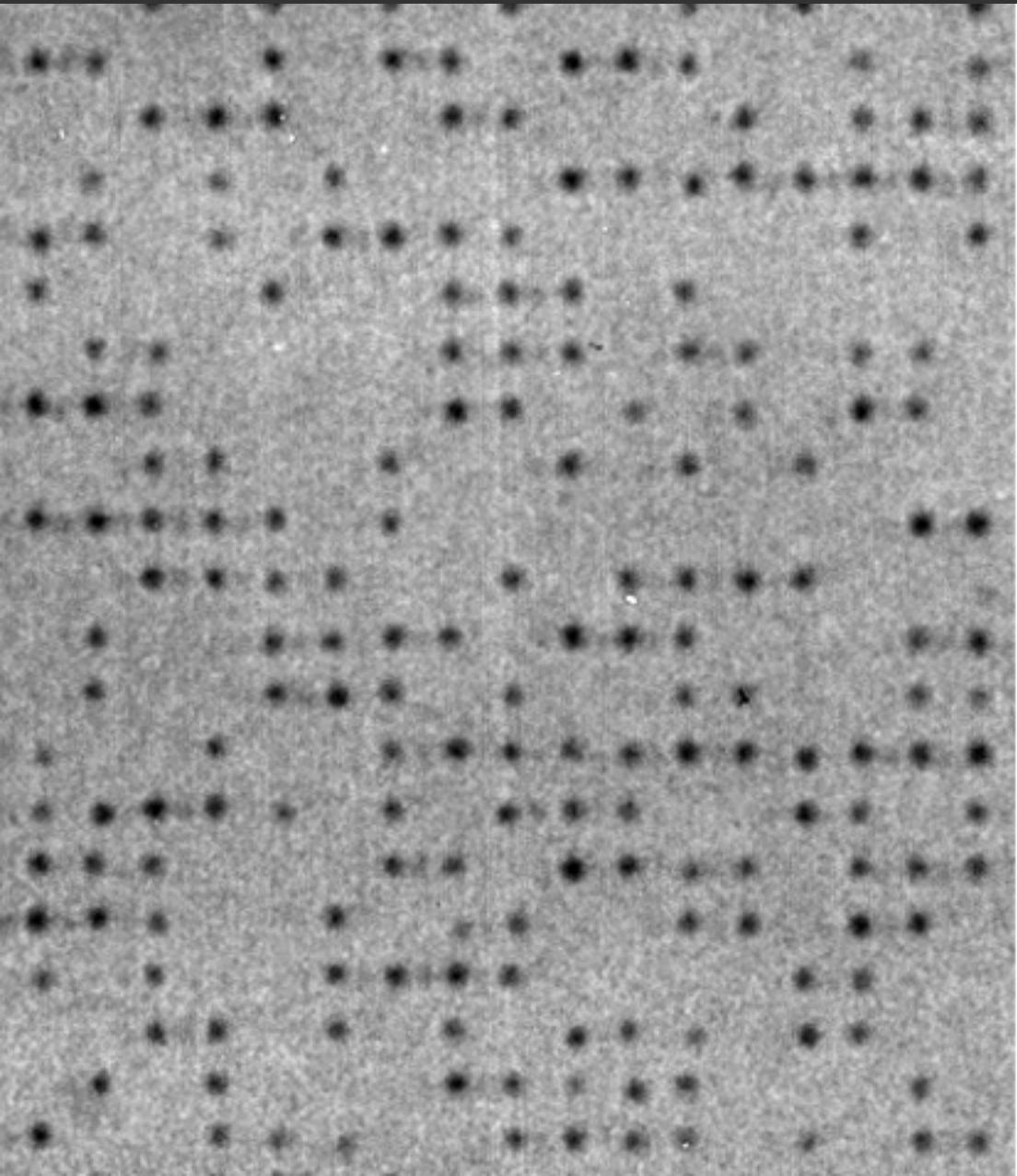
Post-mortem analysis

optical microscopy

2 x 2 μm array

fused silica

0.5 μJ , 100 fs, 800 nm



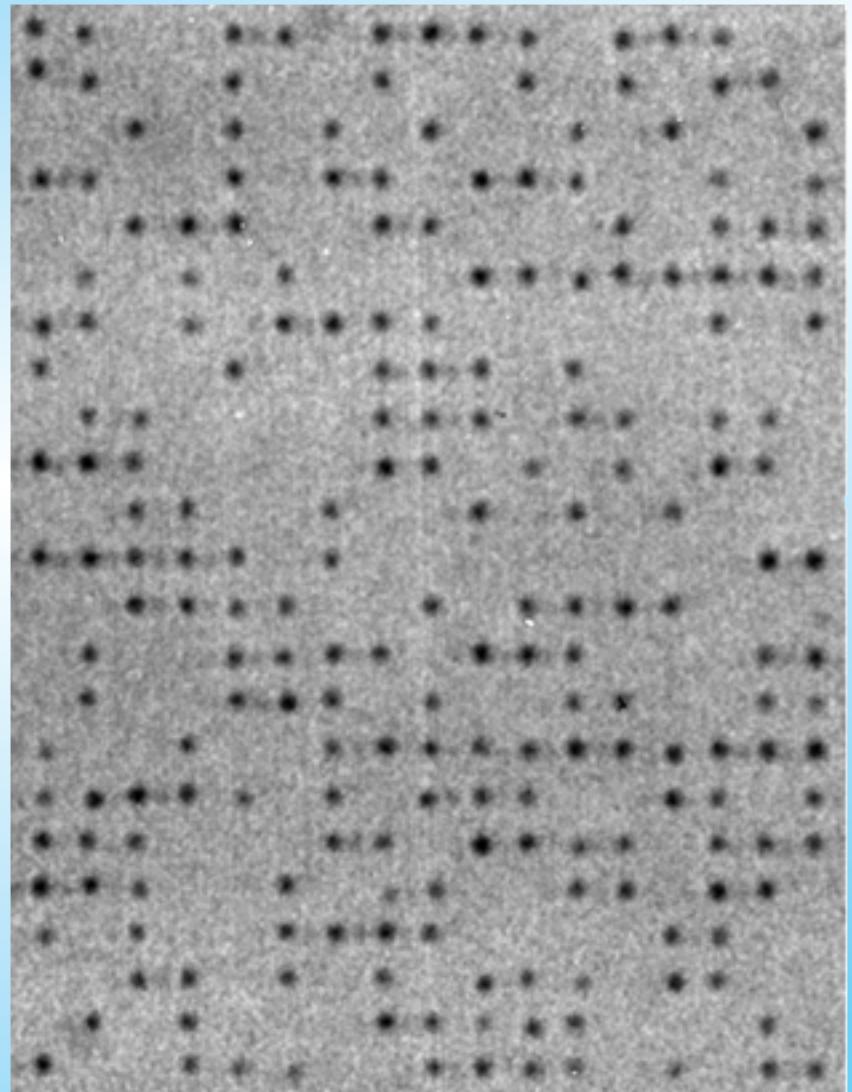
Post-mortem analysis

optical microscopy

2 x 2 μm array

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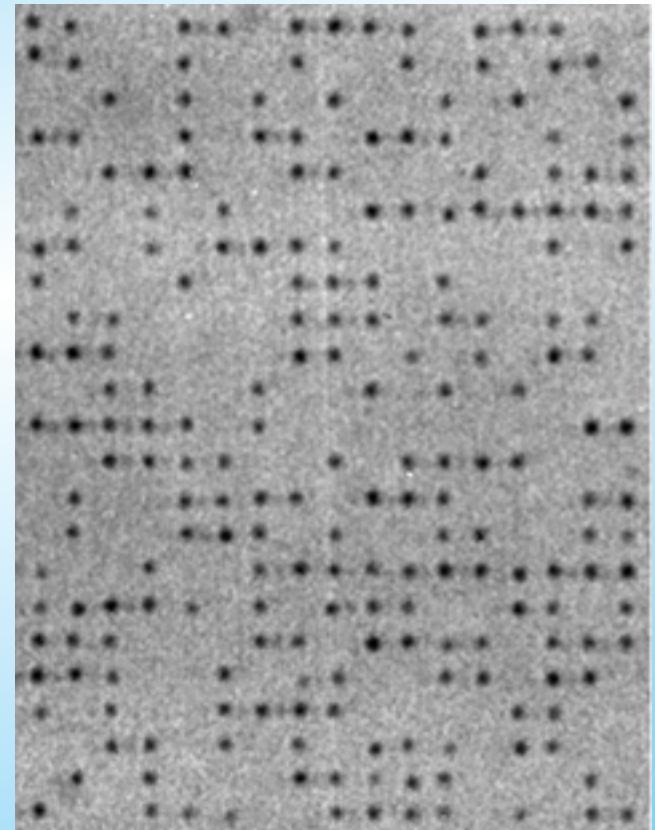
Post-mortem analysis

optical microscopy

2 x 2 μm array

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0.5 μJ , 100 fs, 800 nm



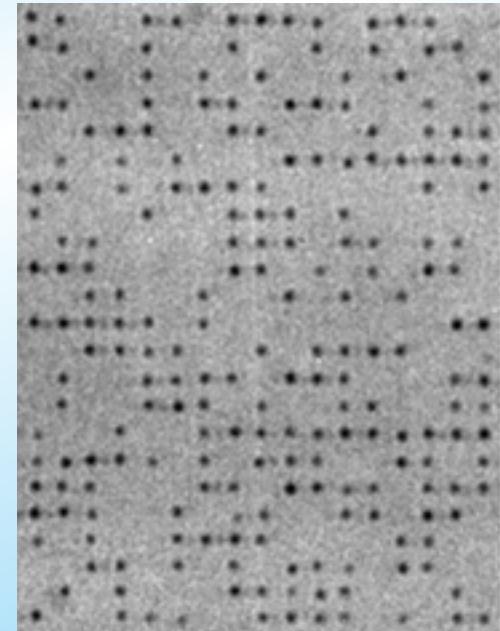
Post-mortem analysis

optical microscopy

2 x 2 μm array

fused silica

0.5 μJ , 100 fs, 800 nm



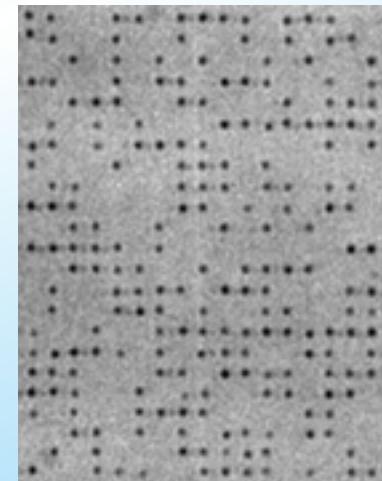
Post-mortem analysis

optical microscopy

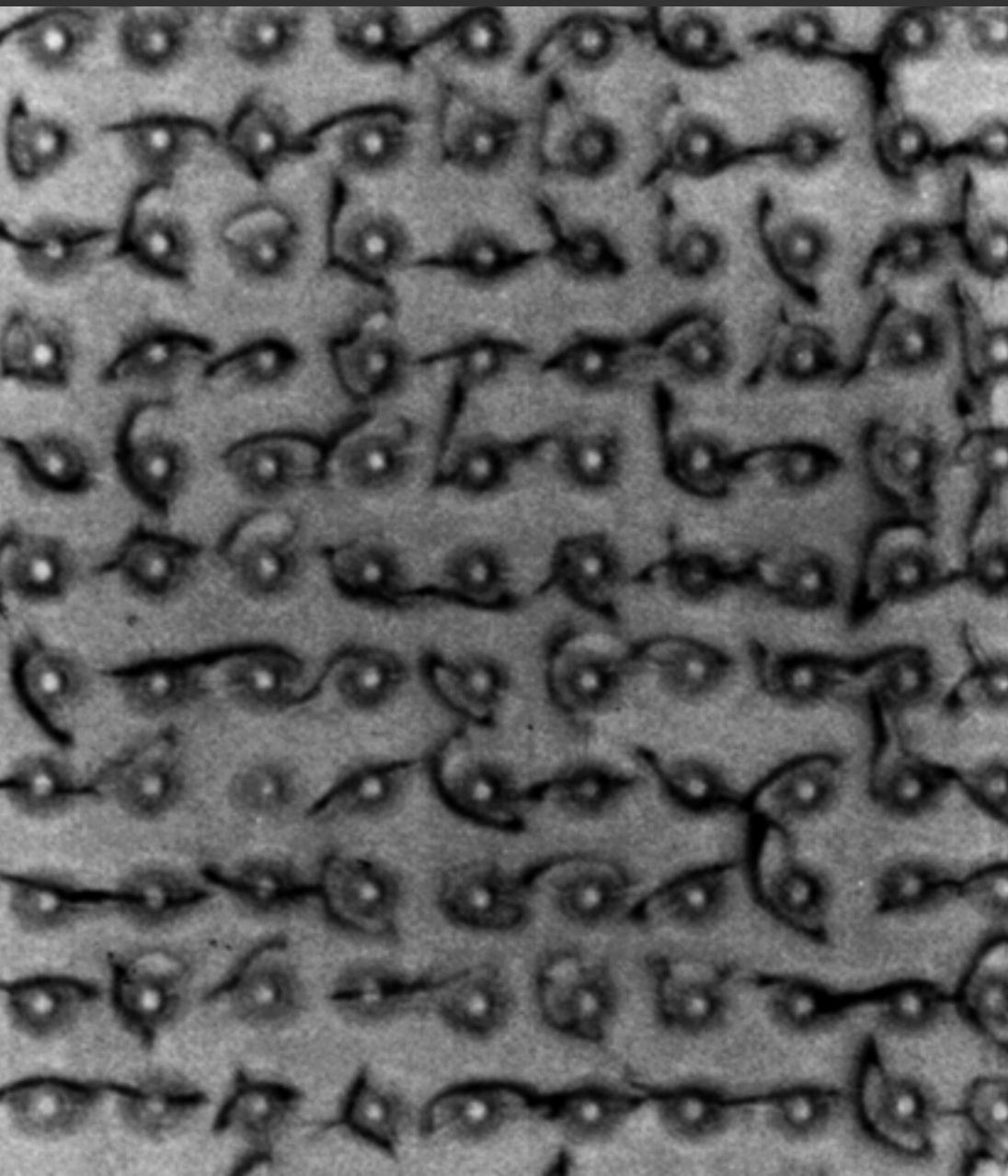
2 x 2 μm array

fused silica

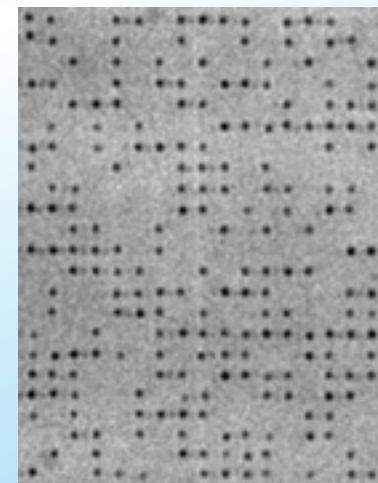
0.5 μJ , 100 fs, 800 nm



Post-mortem analysis

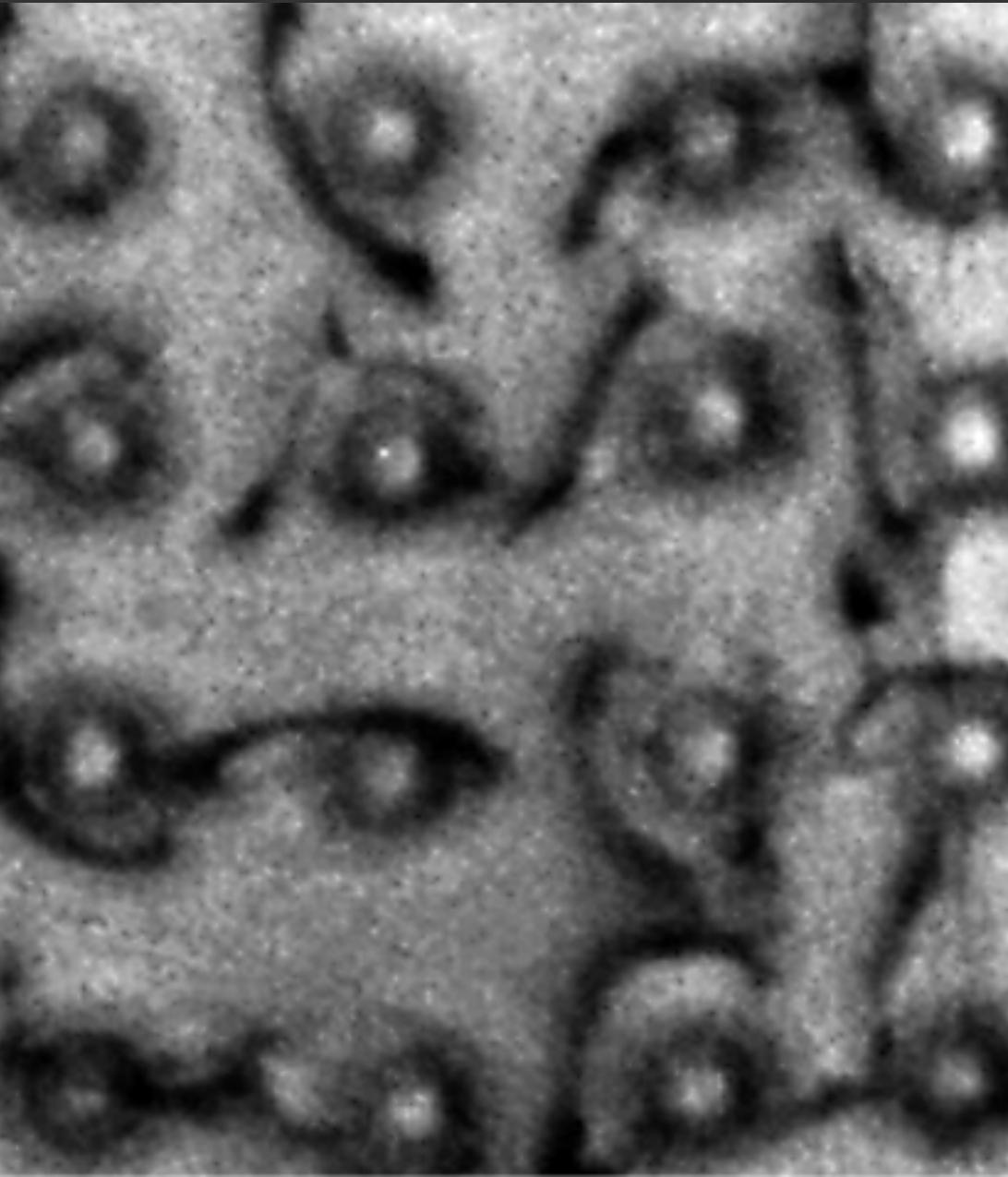


200 ps



100 fs

Post-mortem analysis



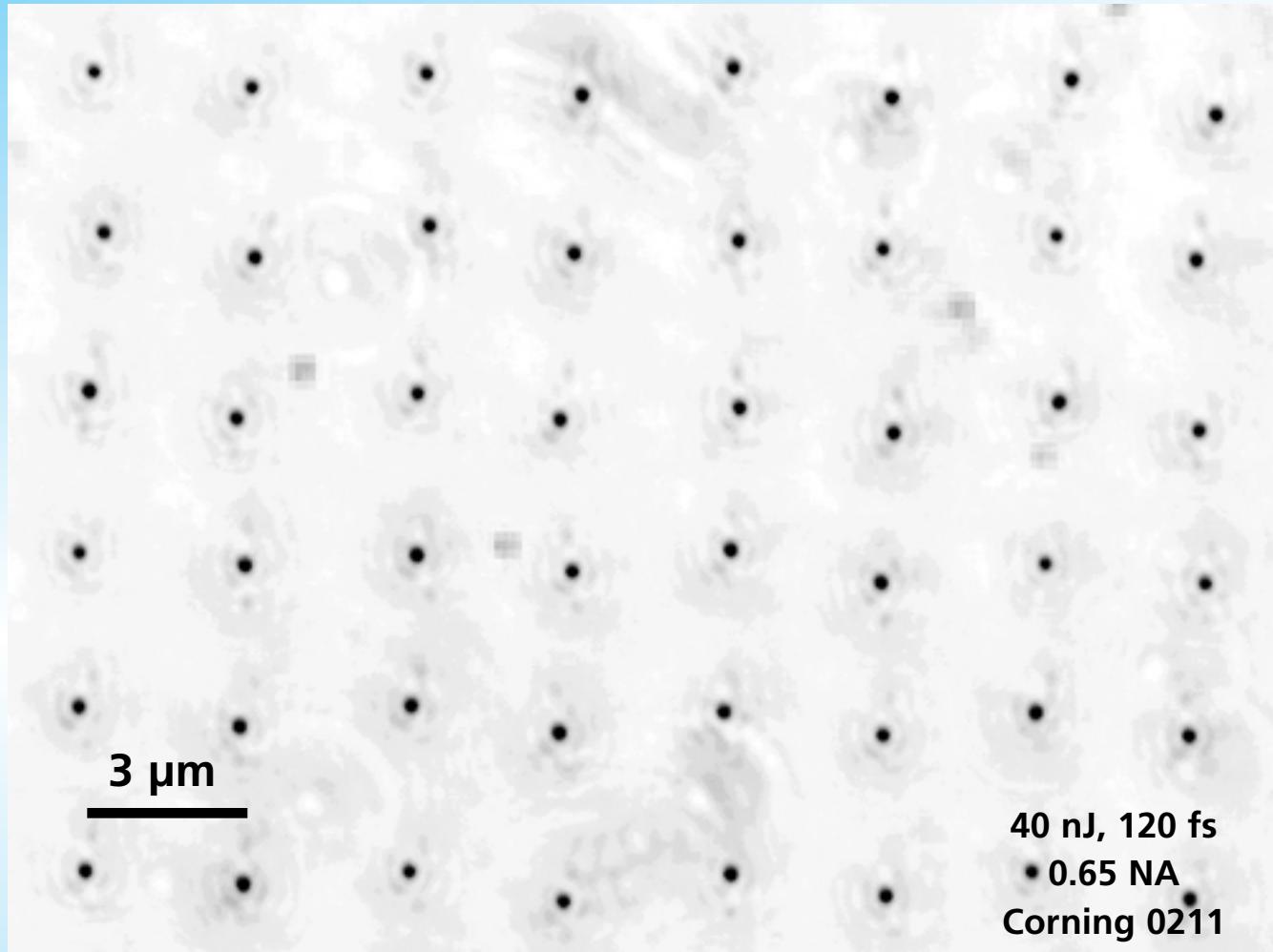
optical microscopy

10 x 10 μm array

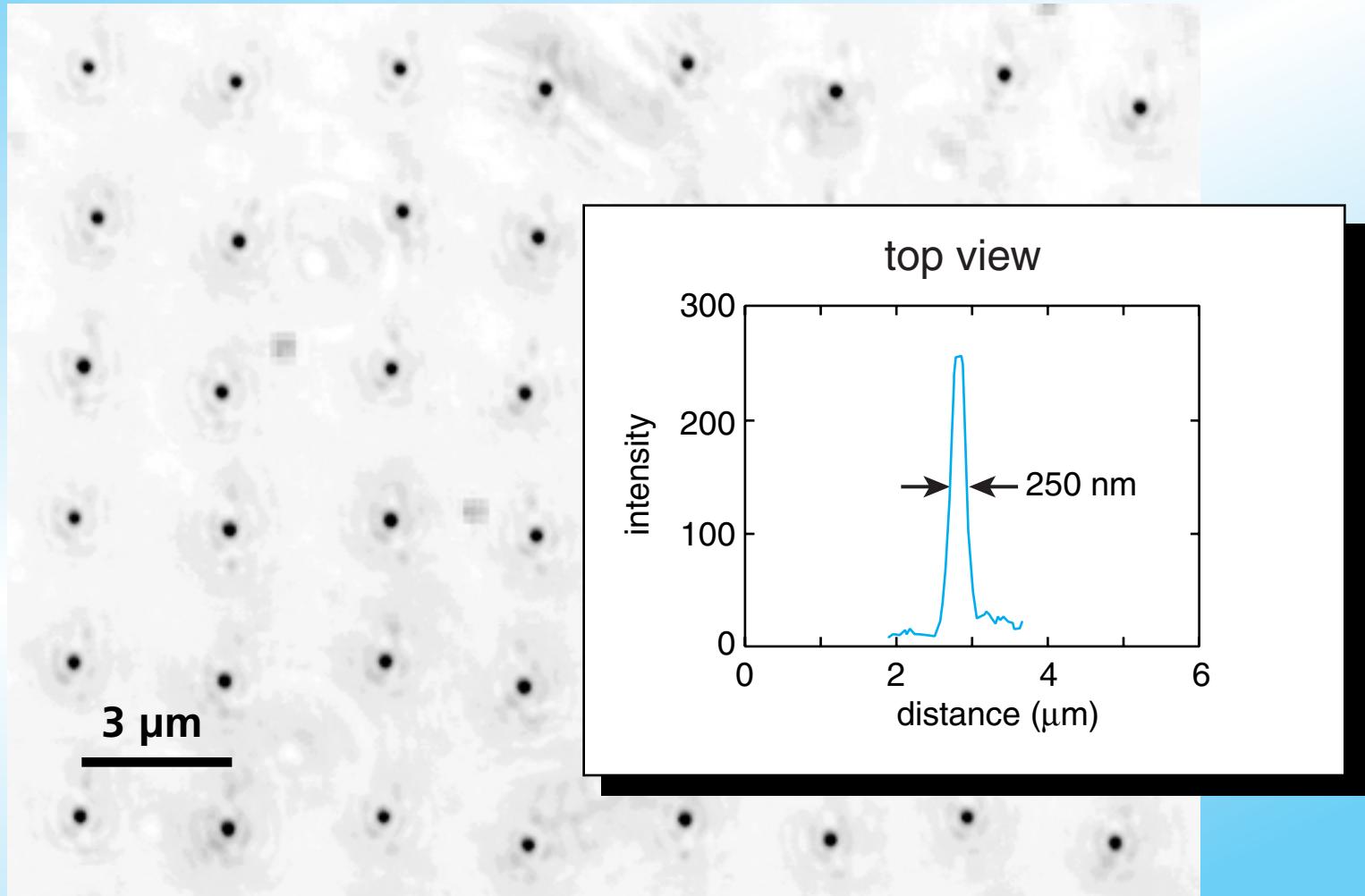
fused silica

9 μJ , 200 ps, 800 nm

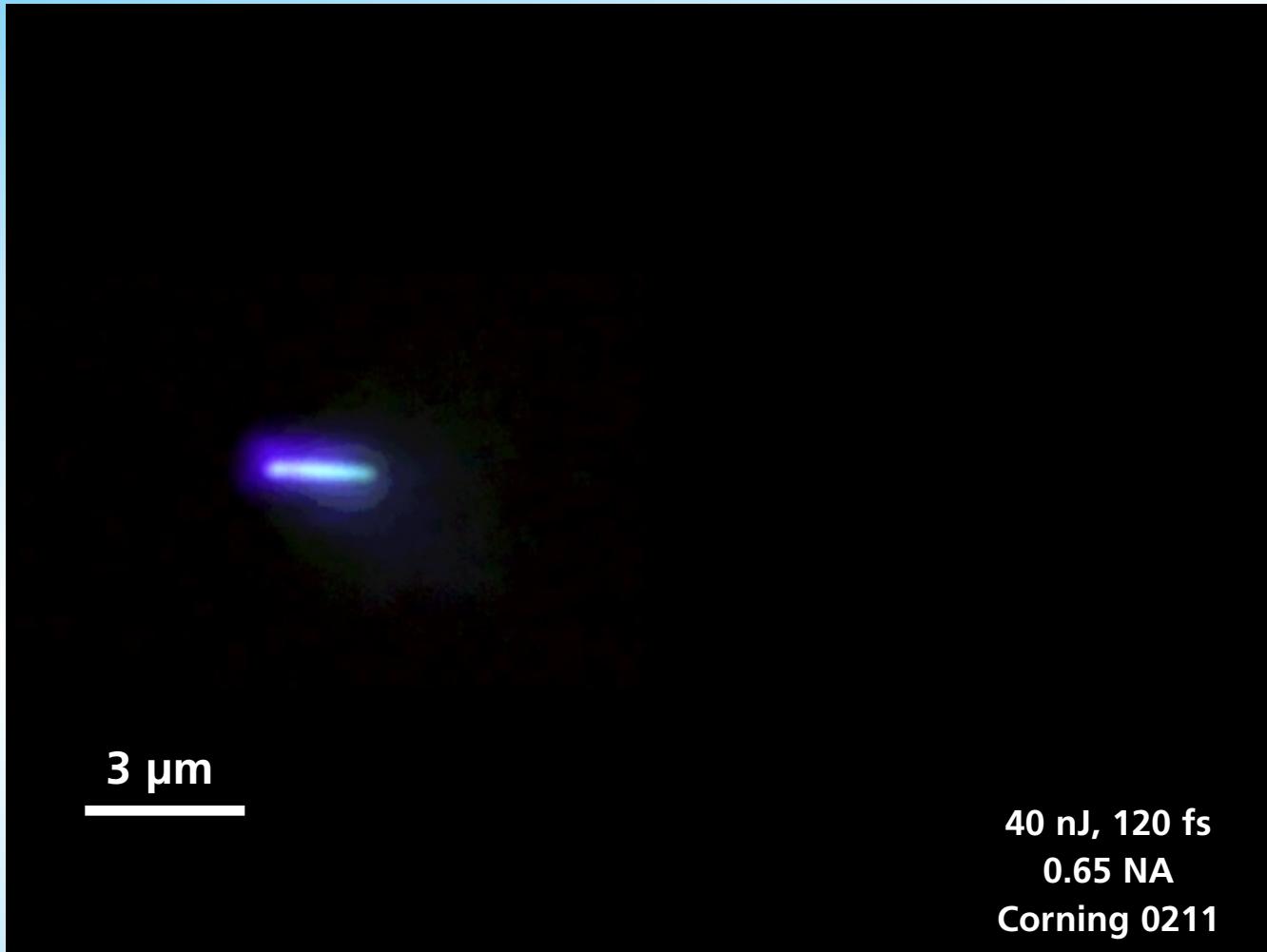
Post-mortem analysis



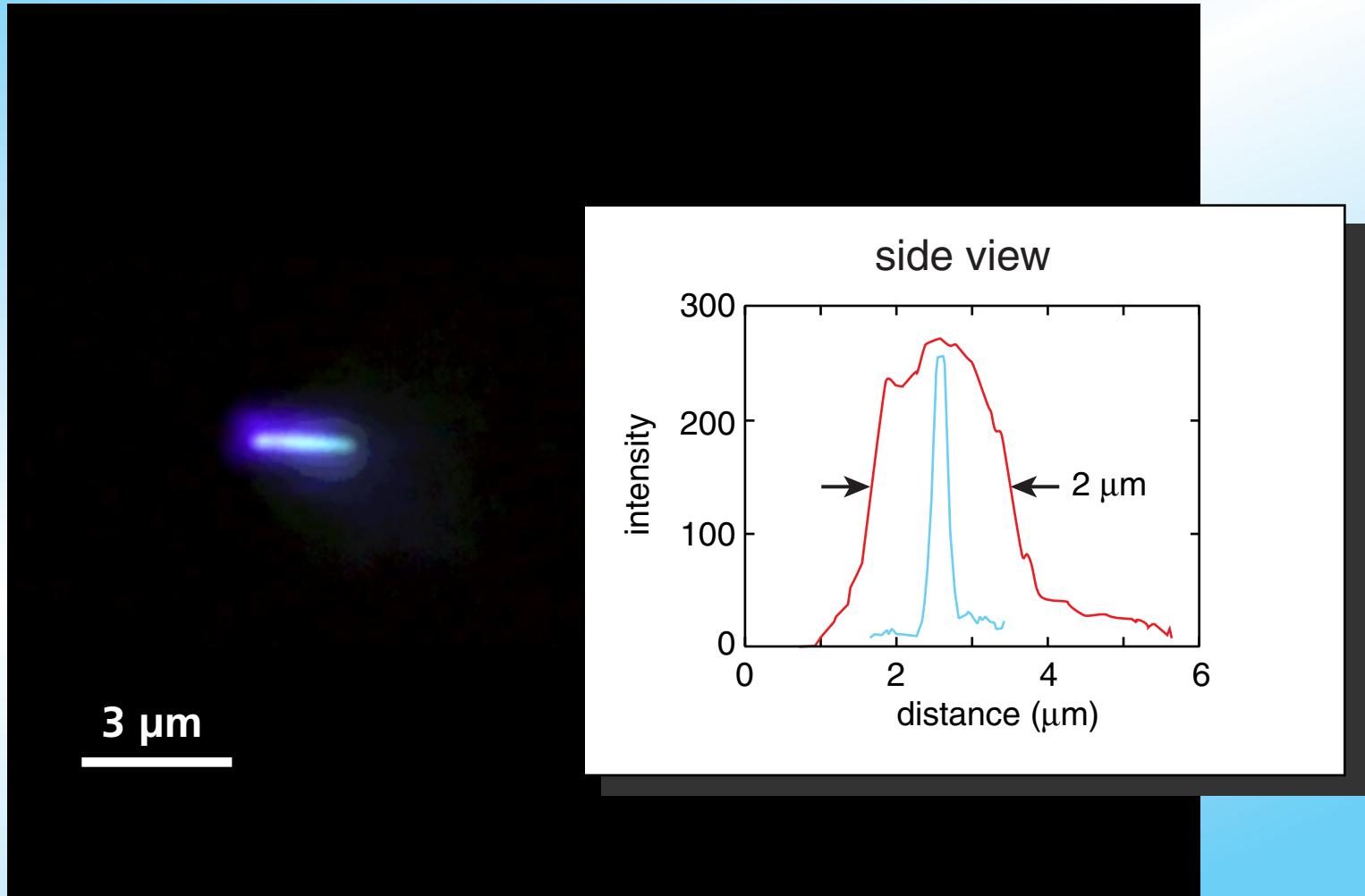
Post-mortem analysis



Post-mortem analysis



Post-mortem analysis



Post-mortem analysis

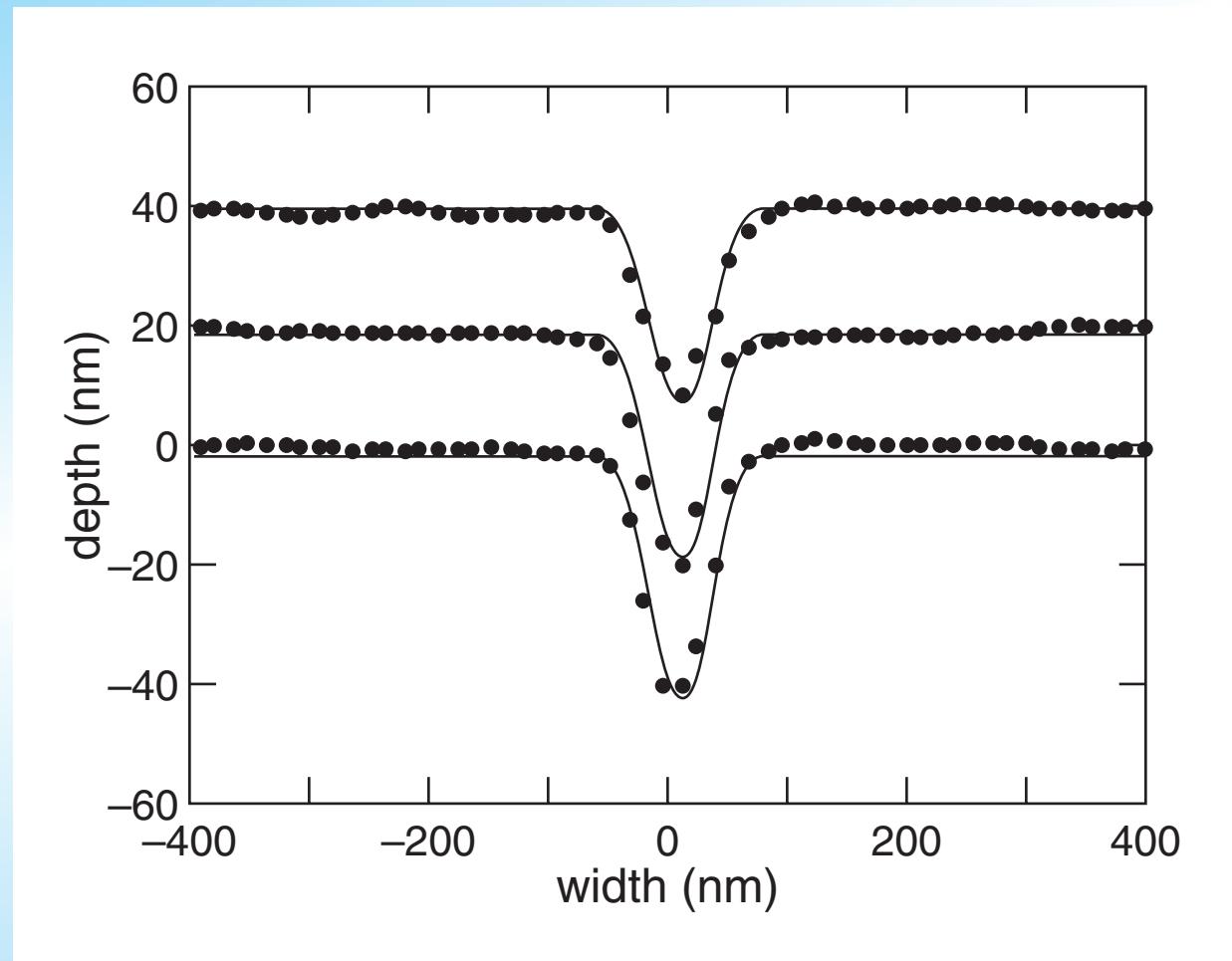
100 nm

SEM:

bumps & pits!

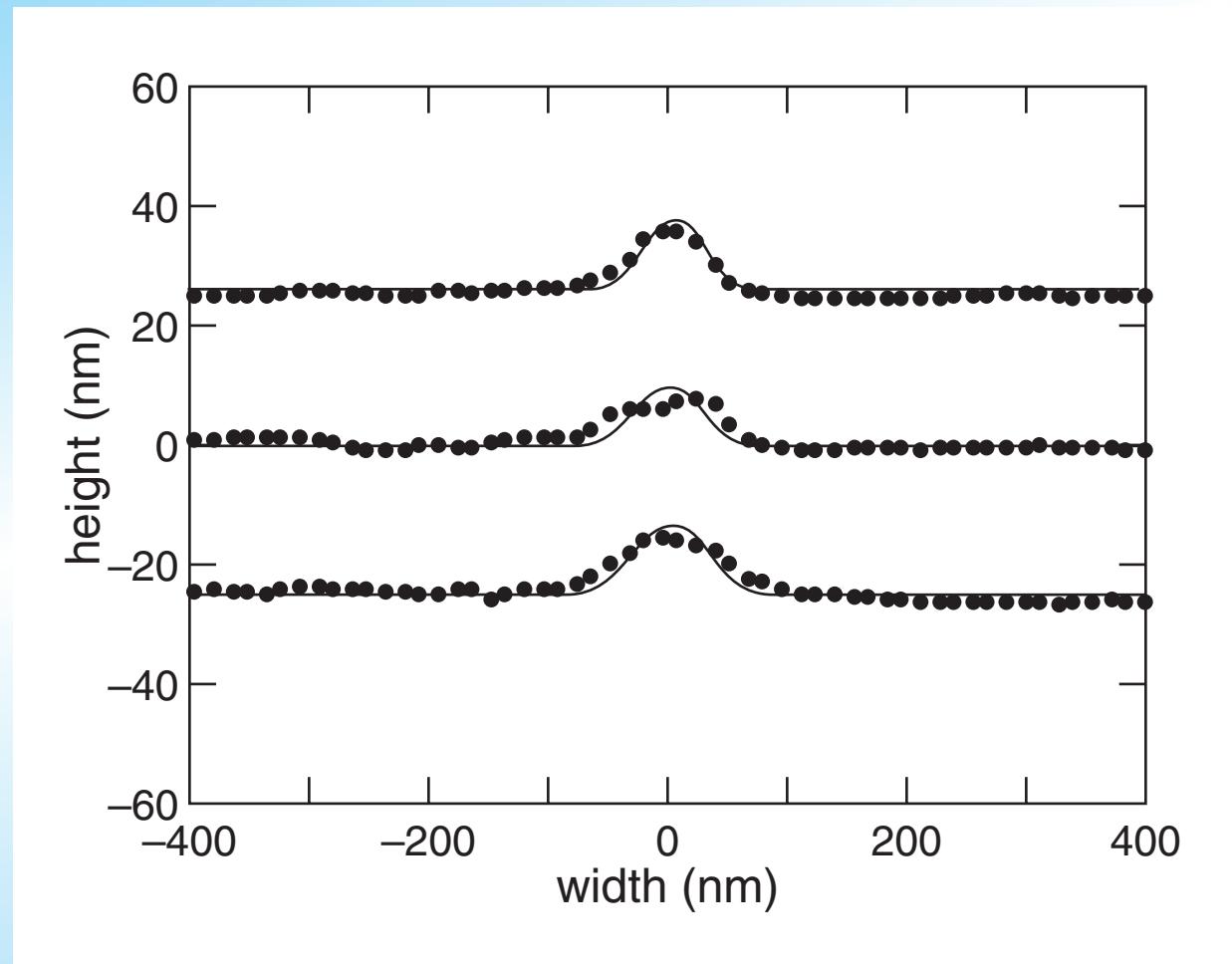
Post-mortem analysis

AFM scans of pits



Post-mortem analysis

AFM scans of bump



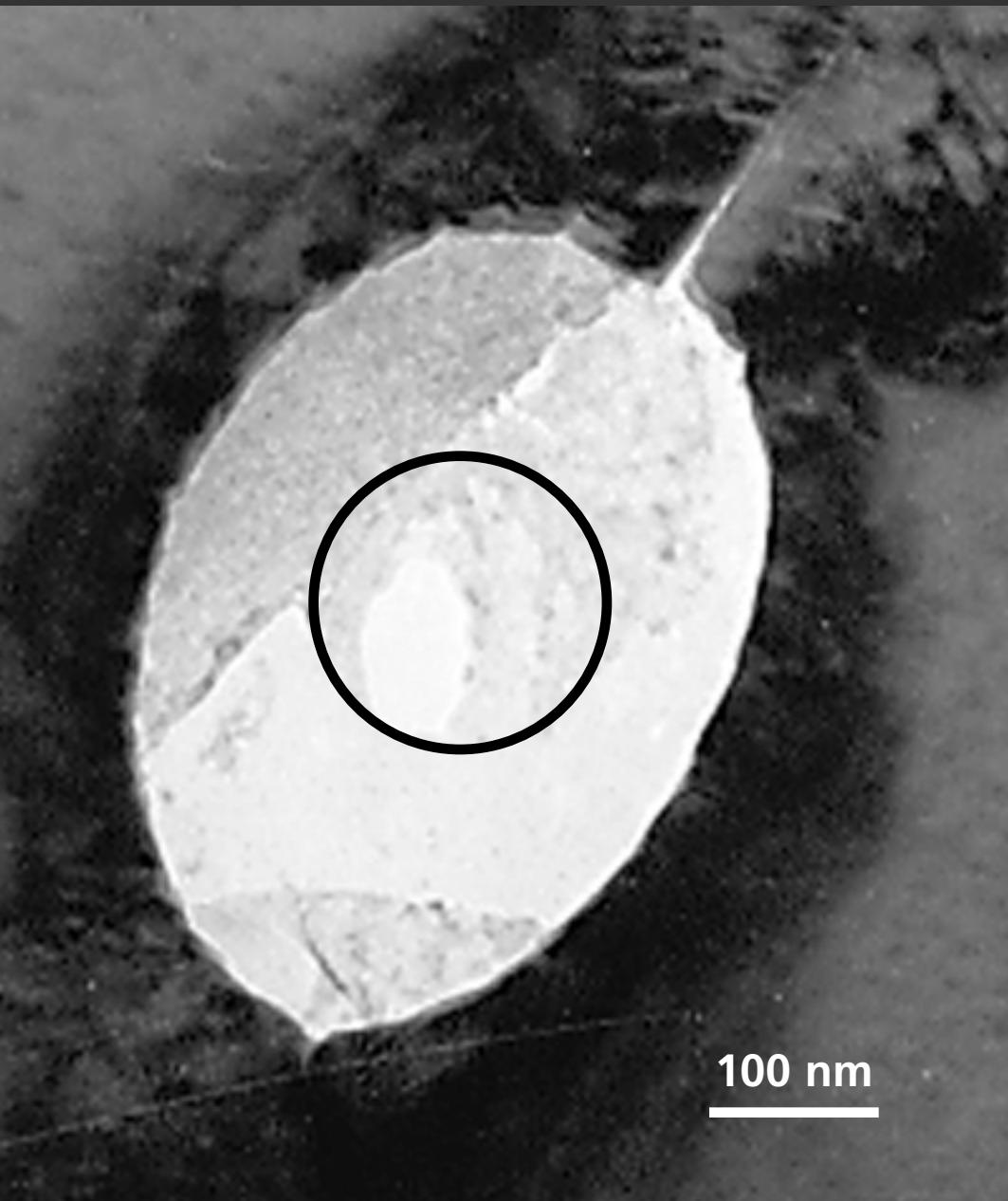
Post-mortem analysis



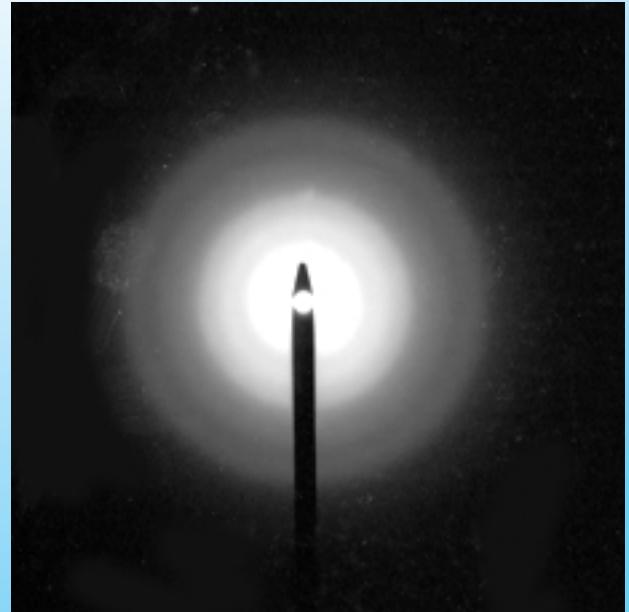
TEM picture

sapphire

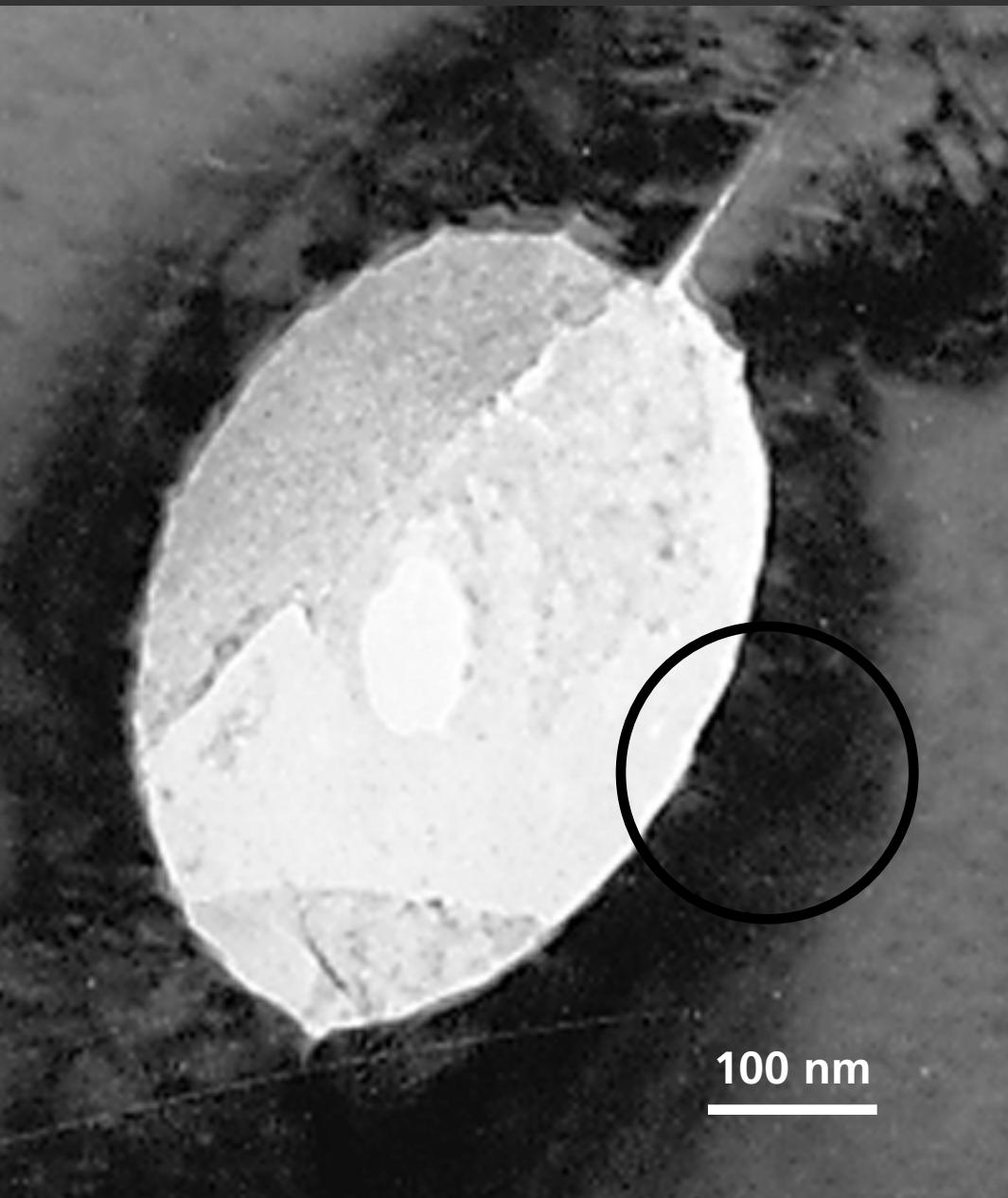
Post-mortem analysis



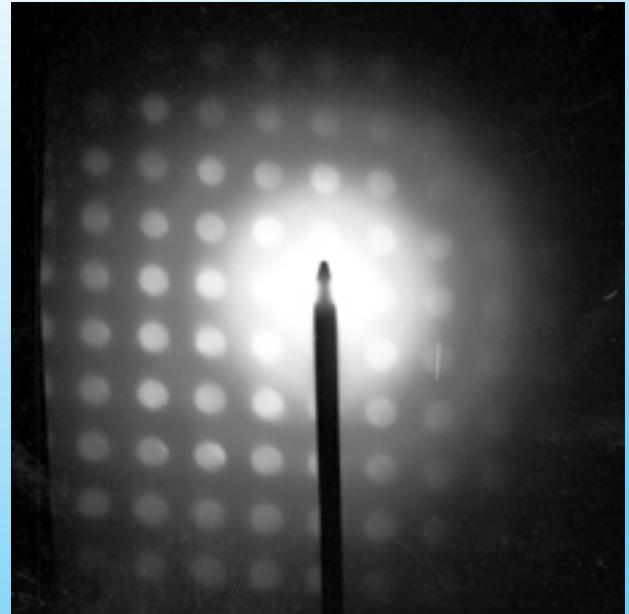
**electron diffraction:
amorphous?**



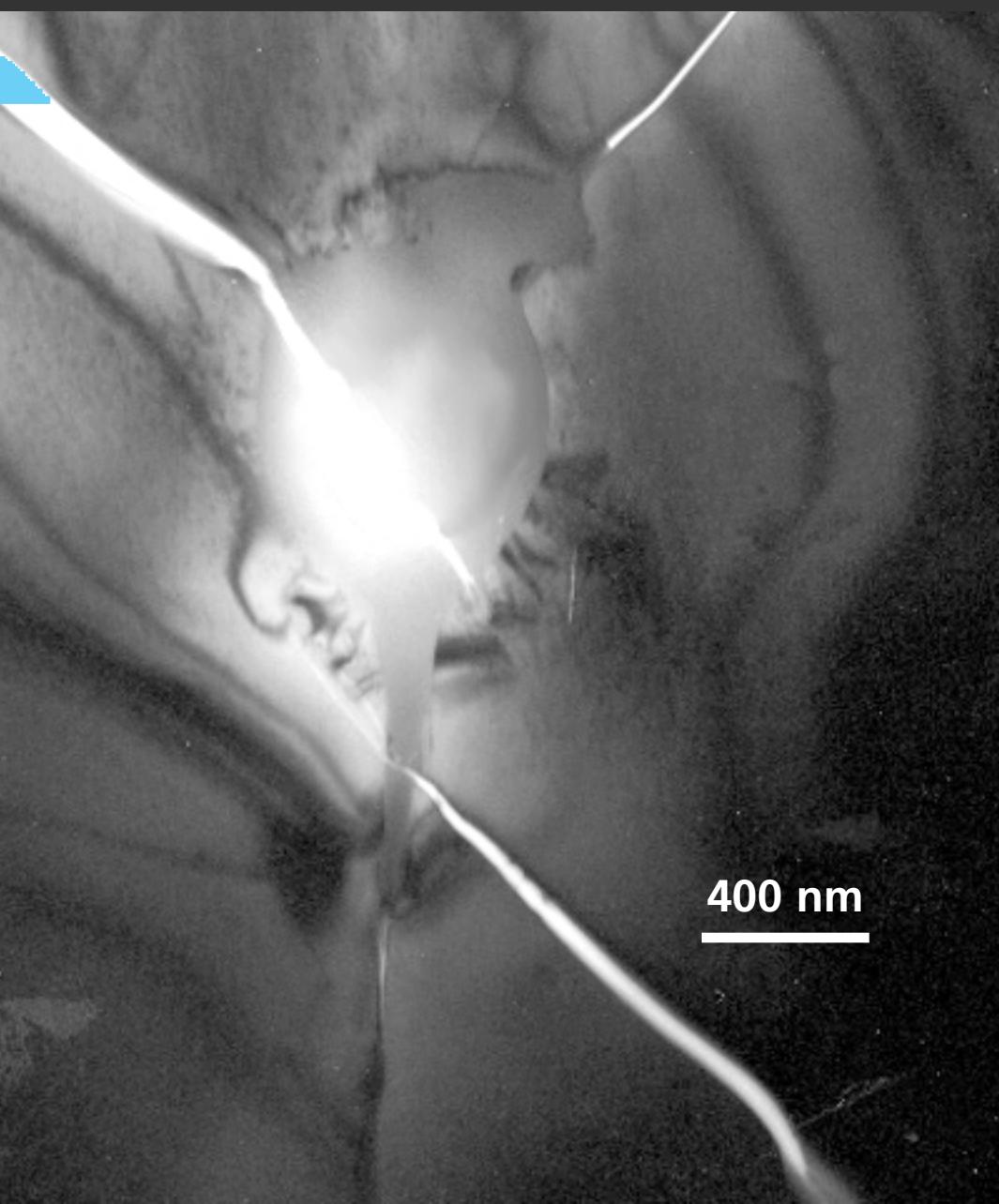
Post-mortem analysis



**electron diffraction:
crystalline**



Post-mortem analysis

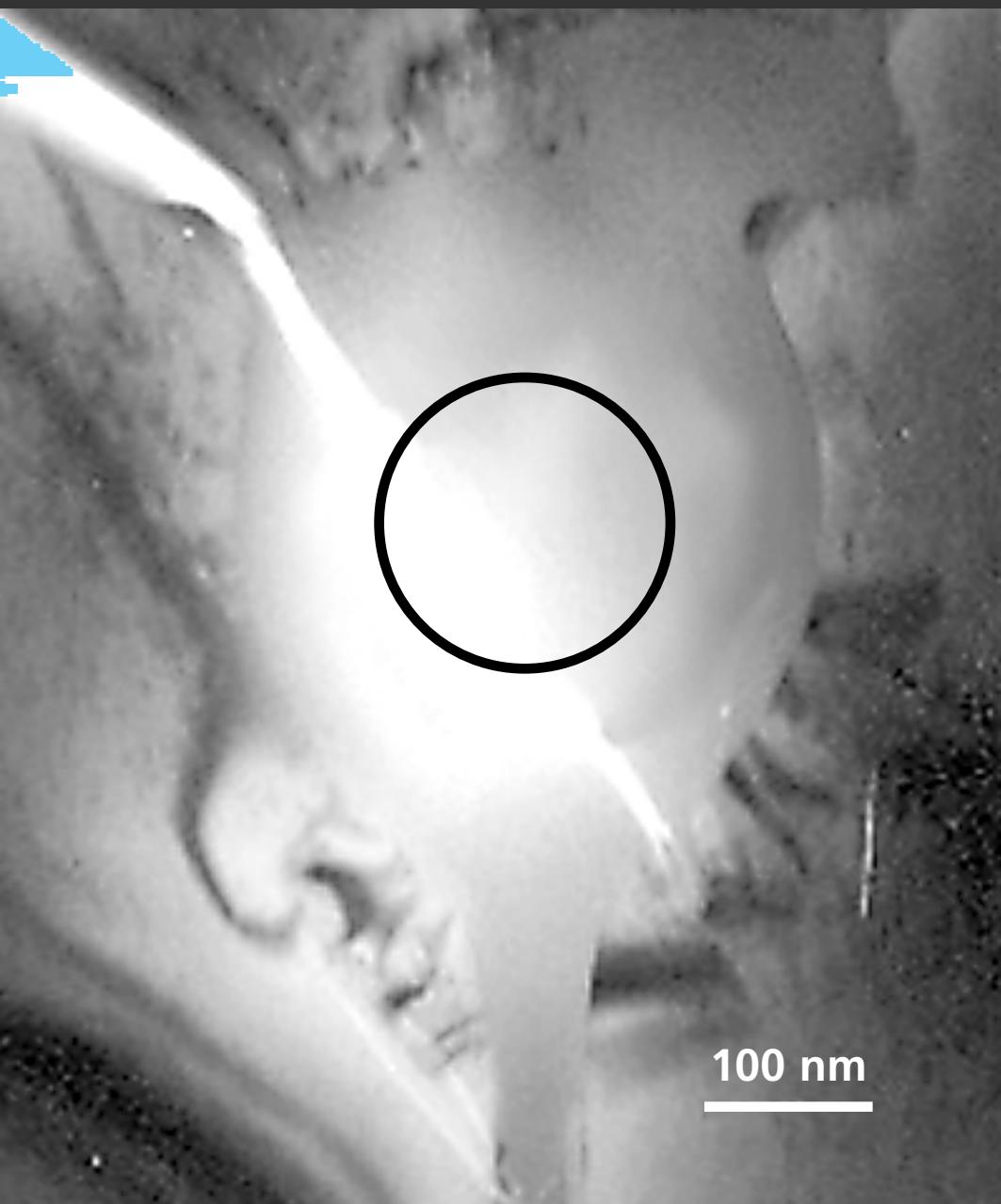


400 nm

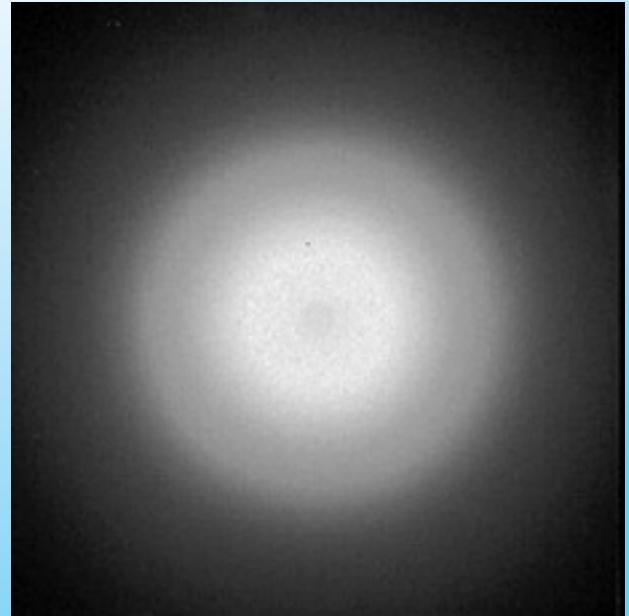
TEM picture

quartz

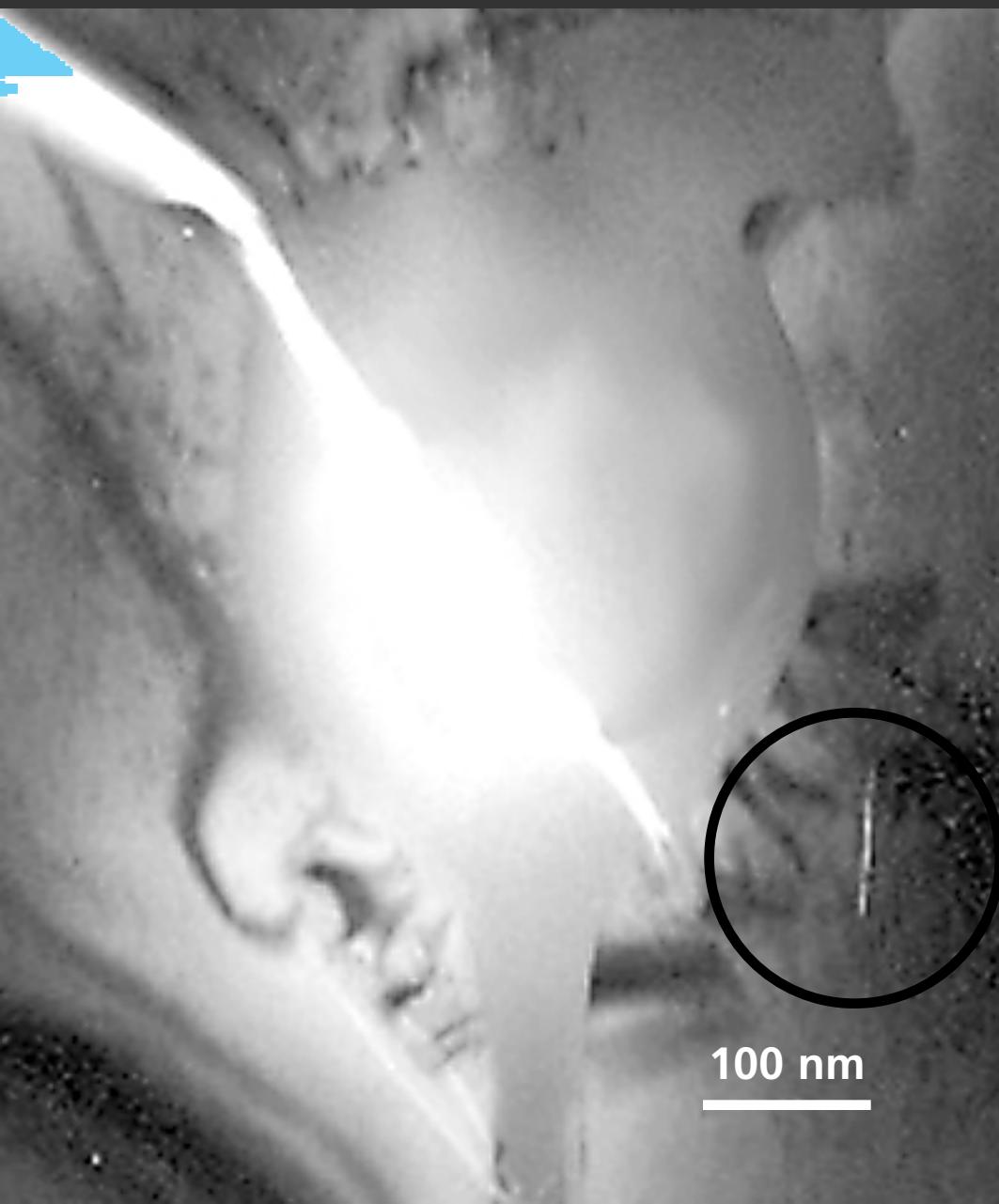
Post-mortem analysis



**electron diffraction:
amorphous**

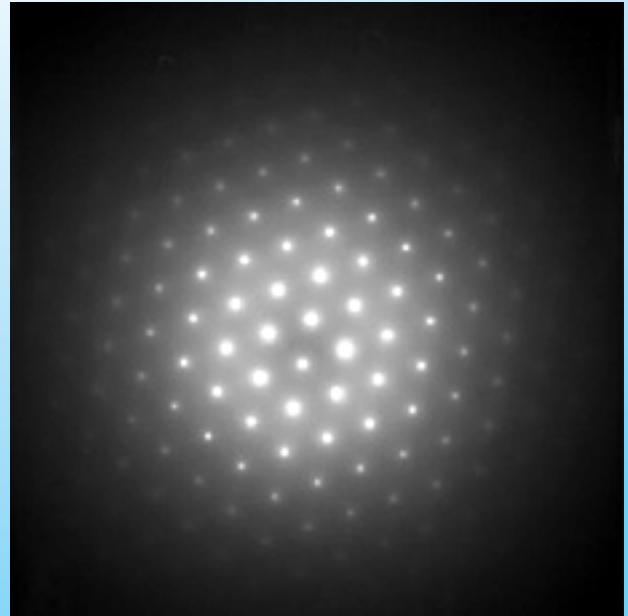


Post-mortem analysis



100 nm

**electron diffraction:
crystalline**



Post-mortem analysis

SEM
microscopy

200 nm

Outline

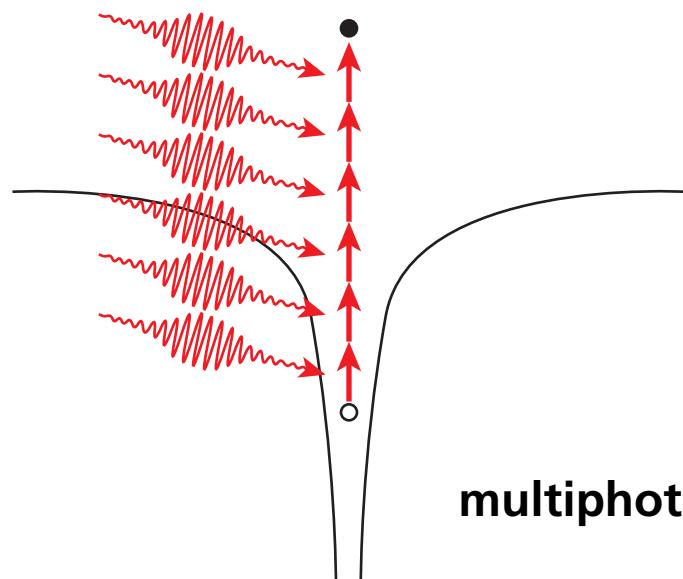
- ▶ Post-mortem analysis
- ▶ Energy deposition
- ▶ Microexplosion dynamics

Energy deposition

how little energy produces permanent changes?

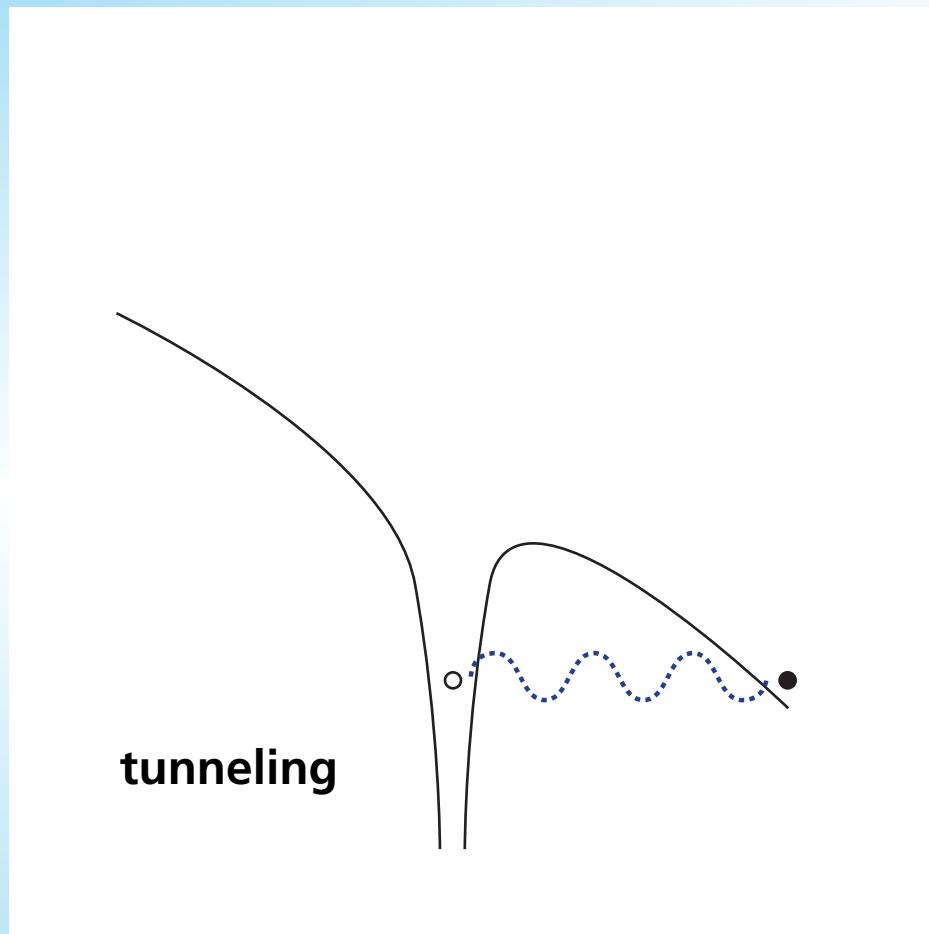
Energy deposition

laser field ionization



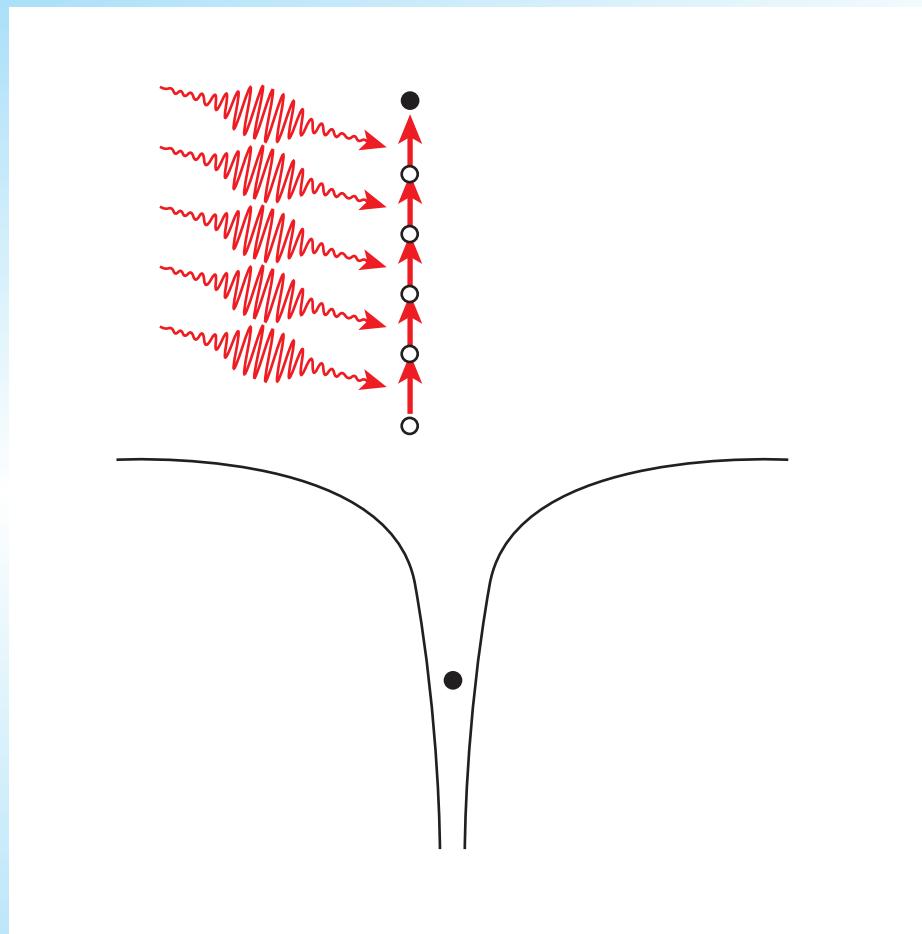
Energy deposition

laser field ionization



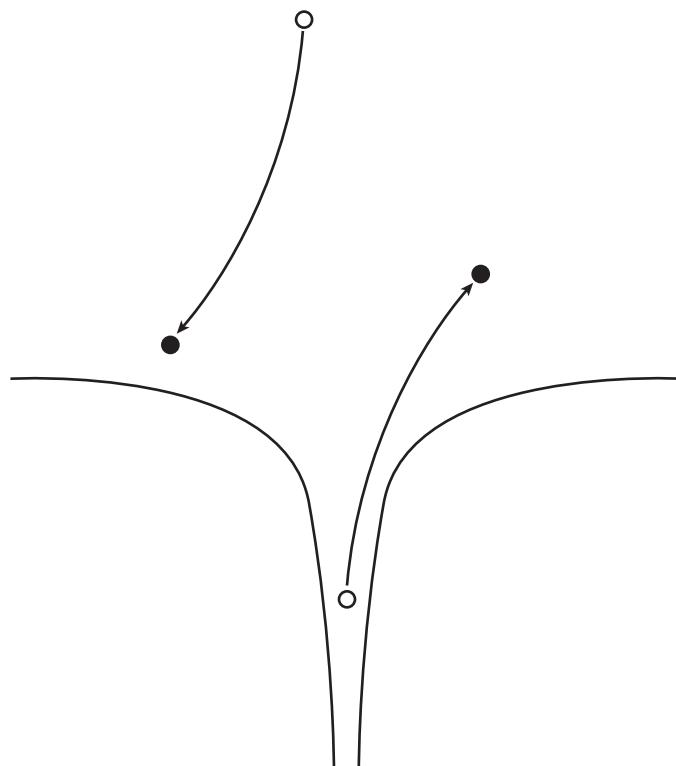
Energy deposition

impact ionization



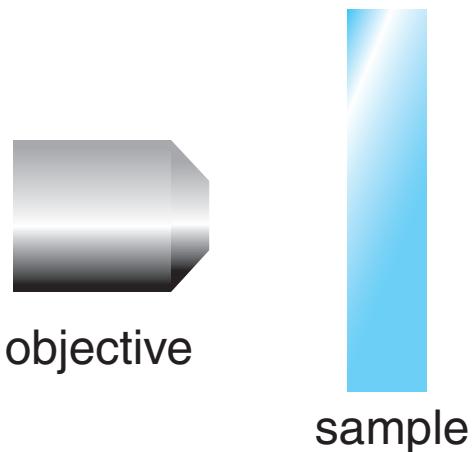
Energy deposition

impact ionization



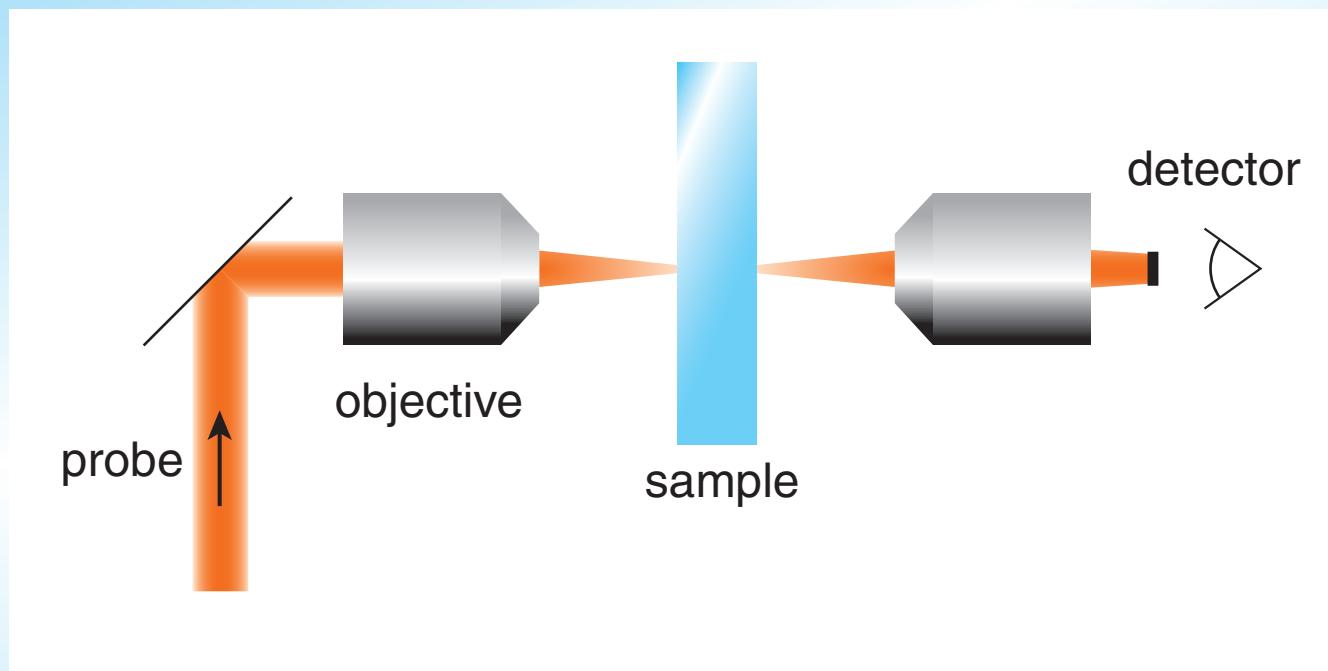
Energy deposition

Dark-field scattering



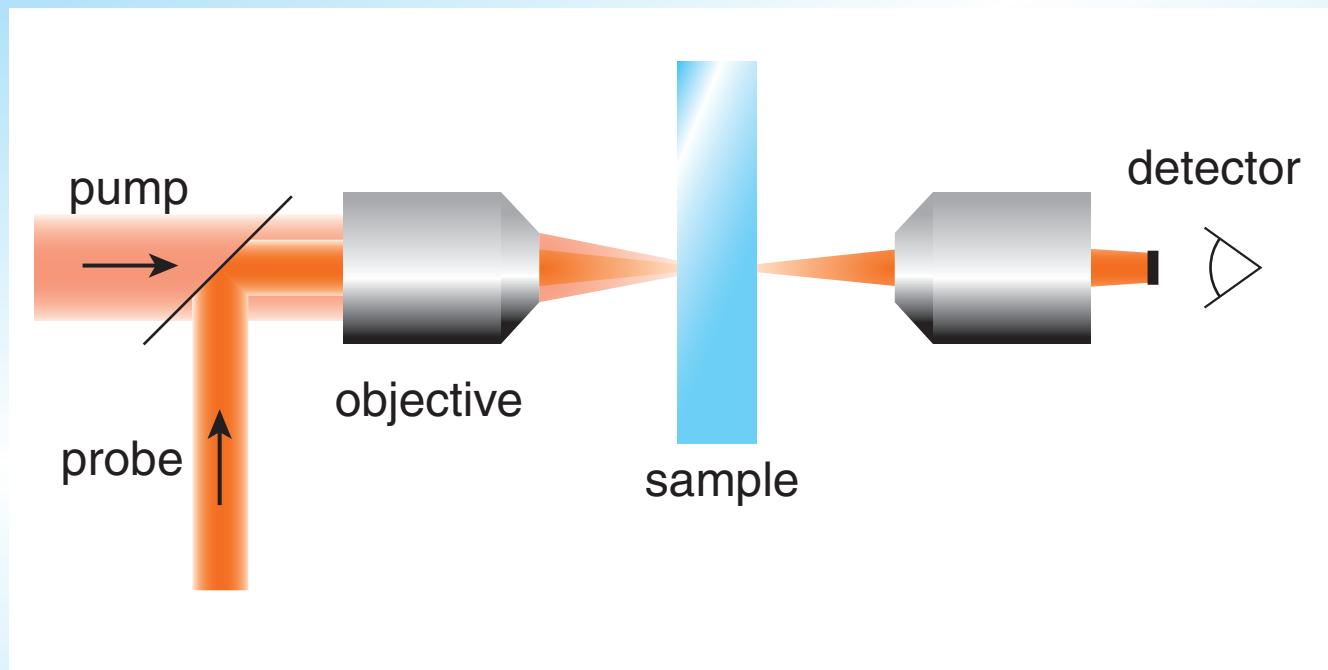
Energy deposition

block probe beam...



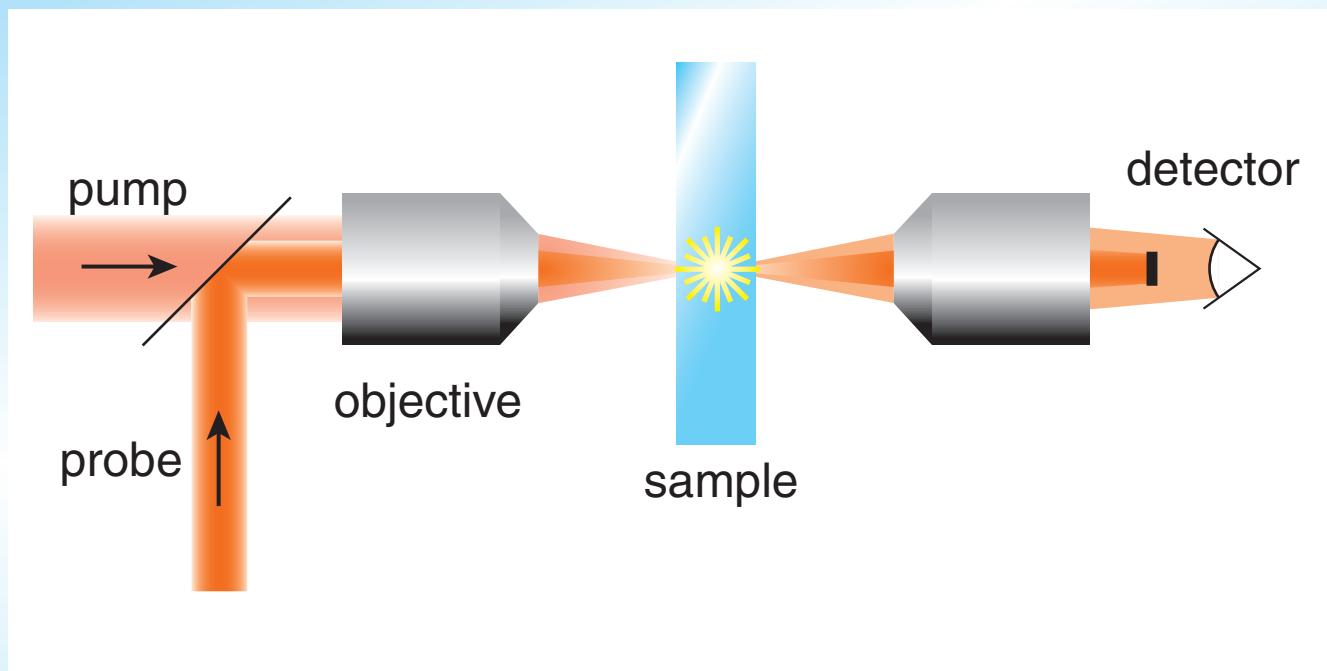
Energy deposition

...bring in pump beam...

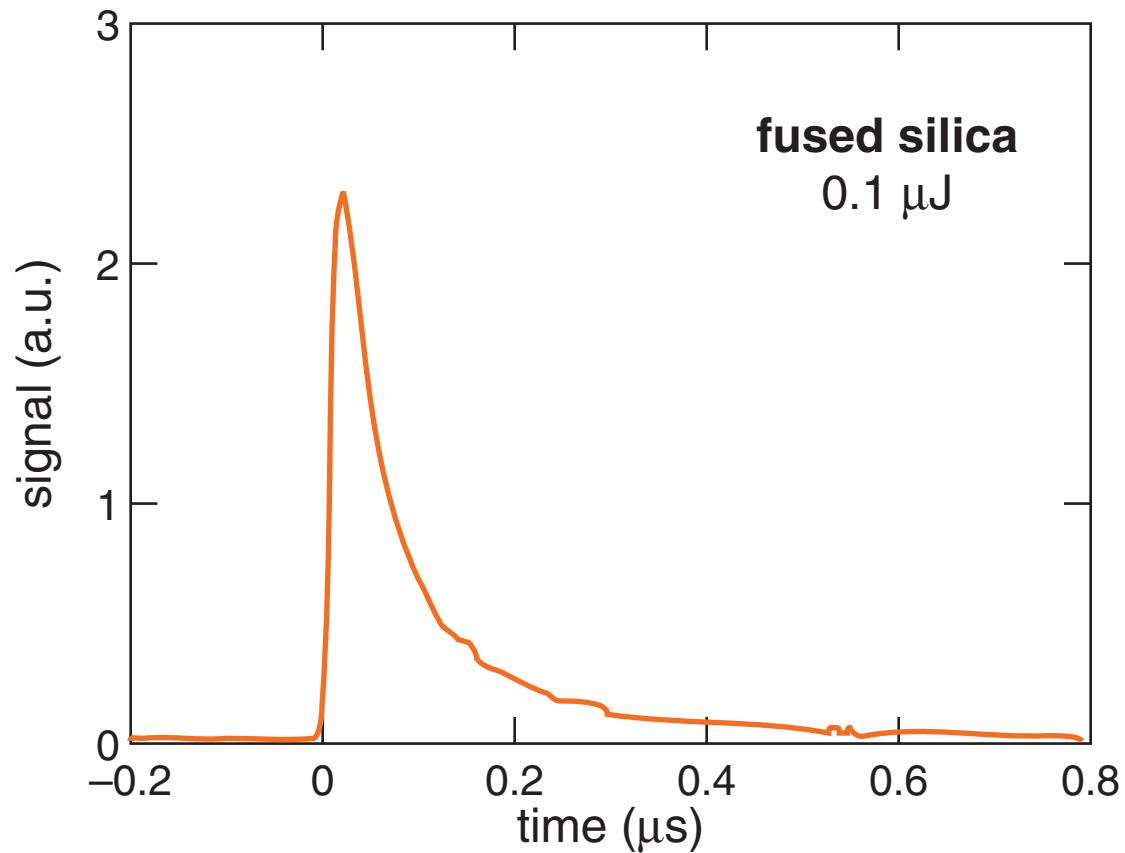


Energy deposition

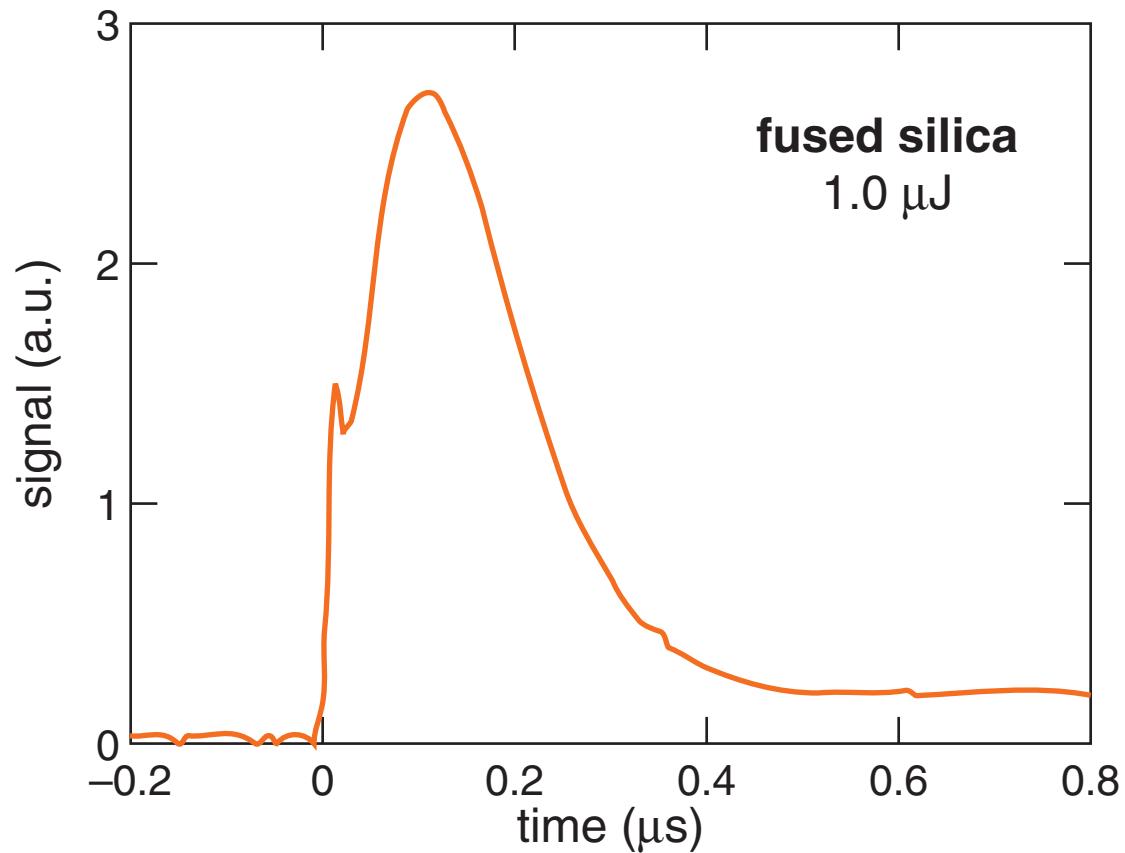
...damage scatters probe beam



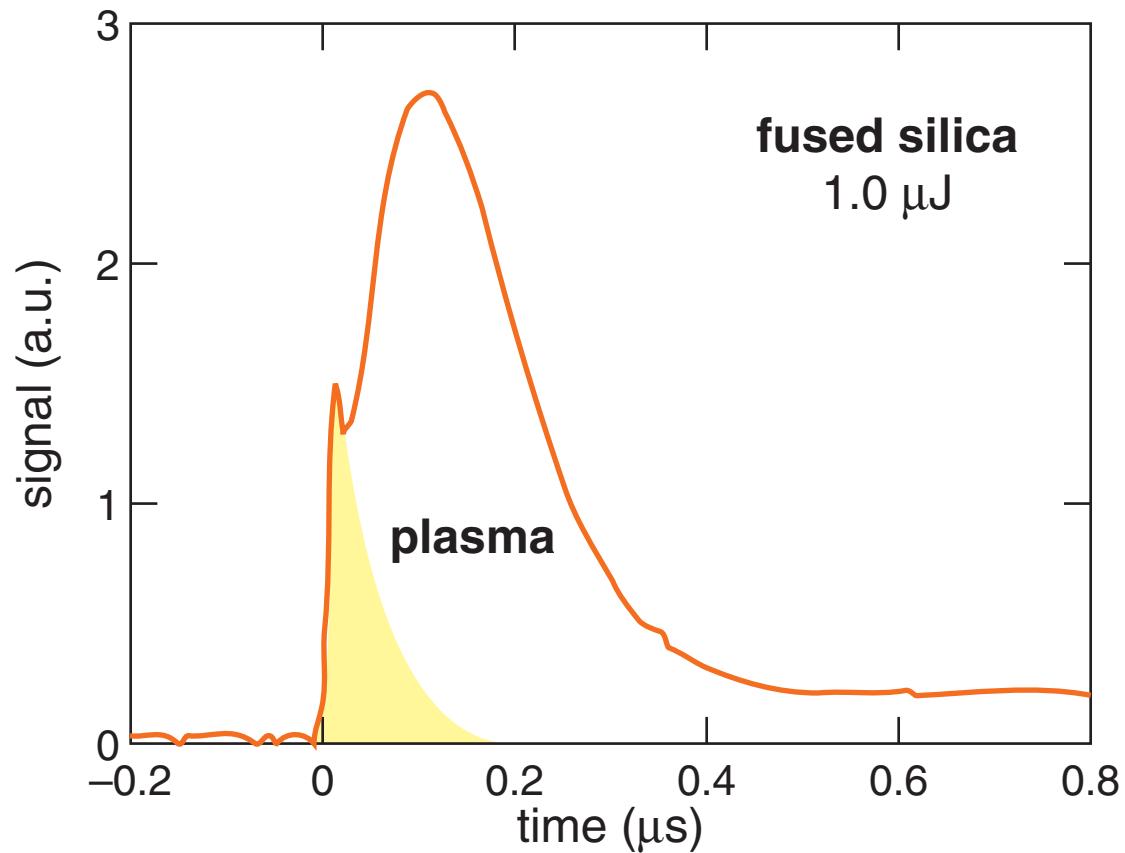
Energy deposition



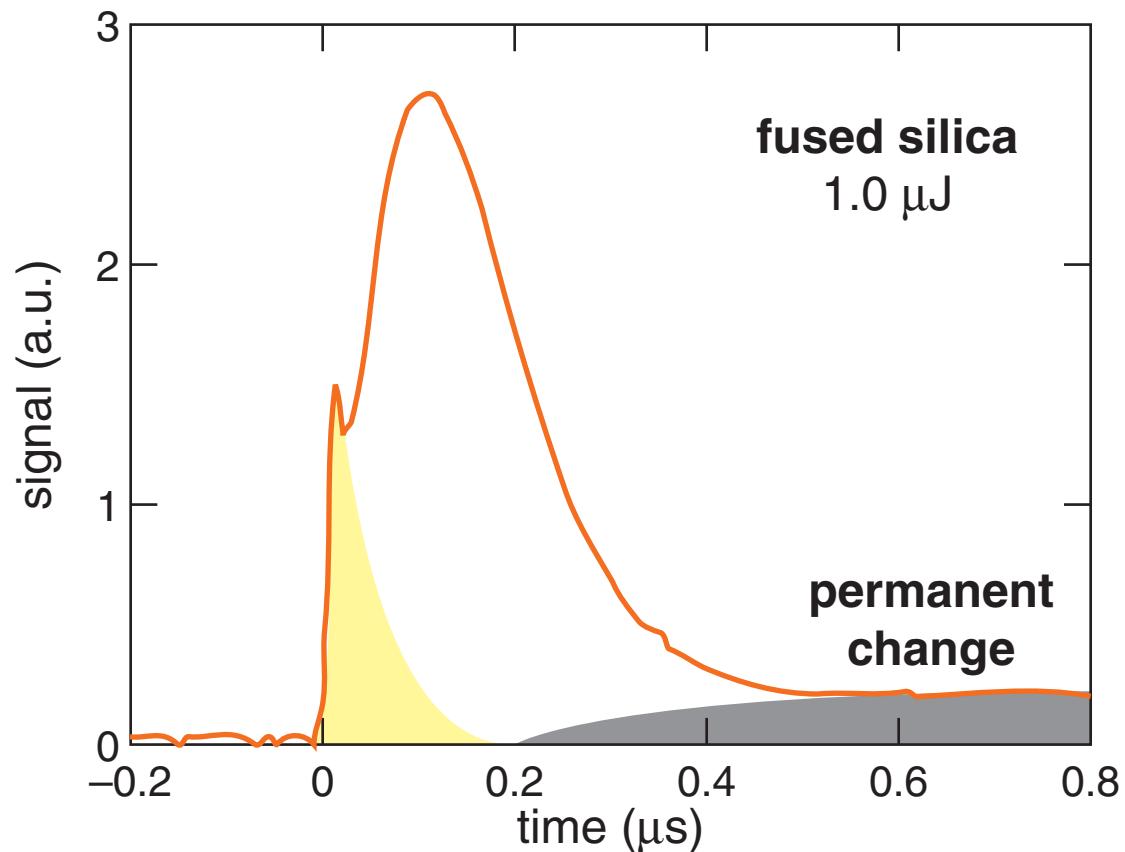
Energy deposition



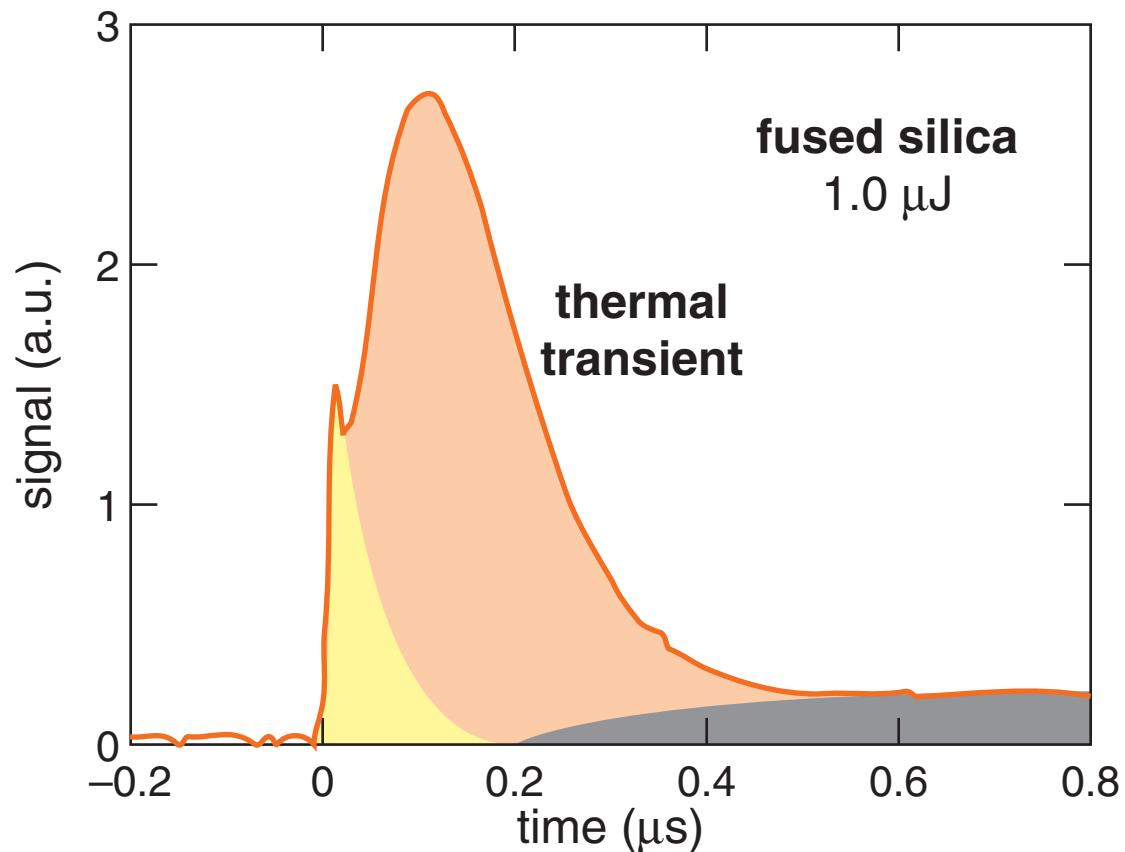
Energy deposition



Energy deposition

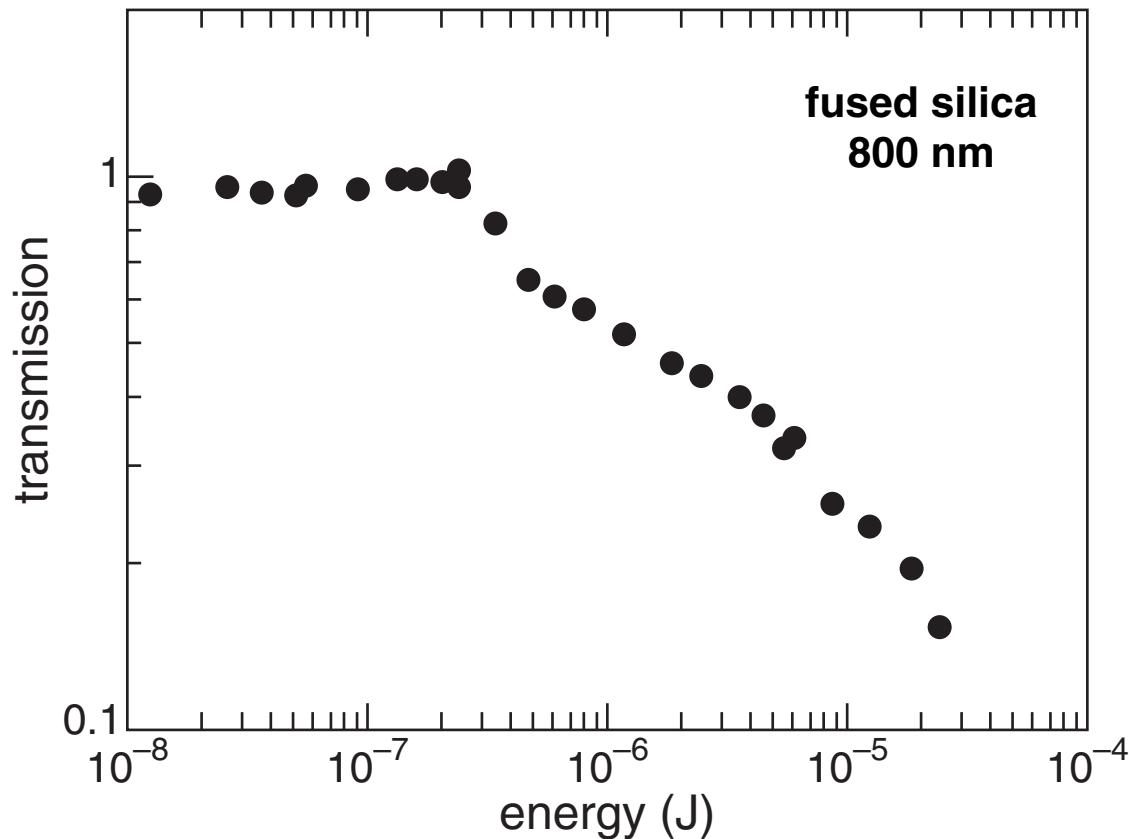


Energy deposition



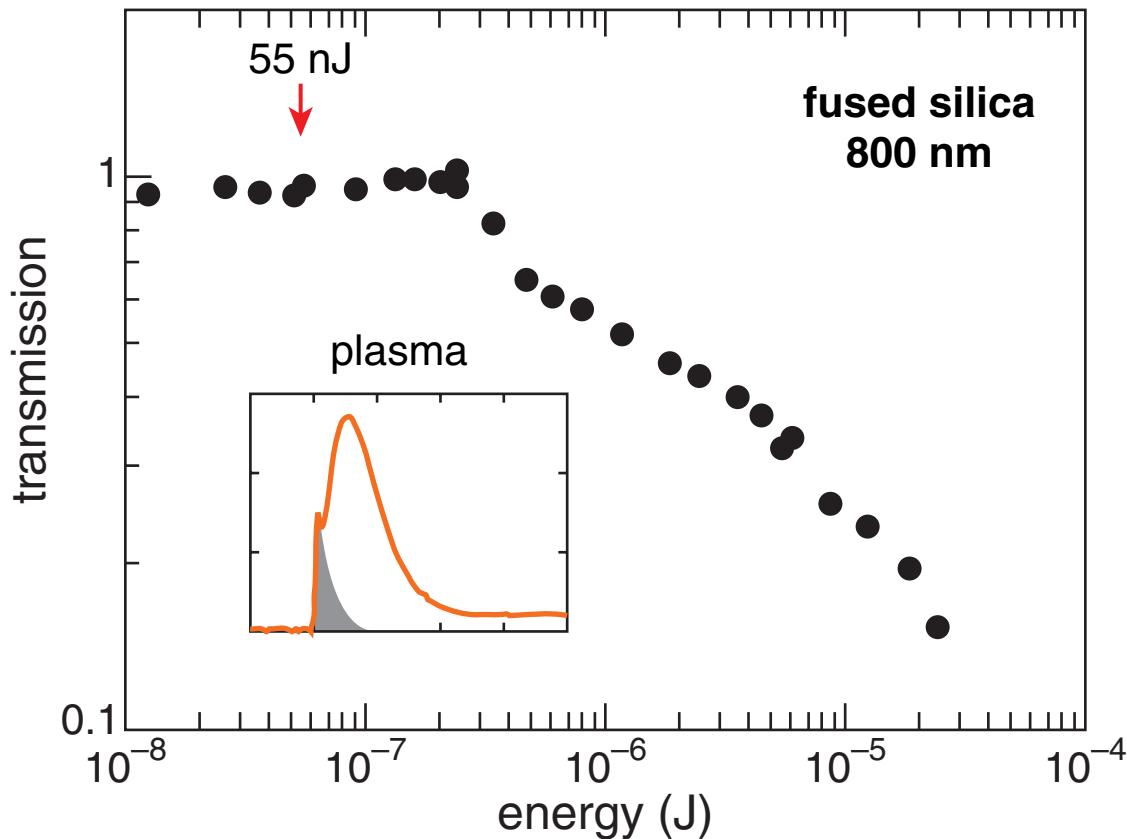
Energy deposition

transmission of pump beam in fused silica



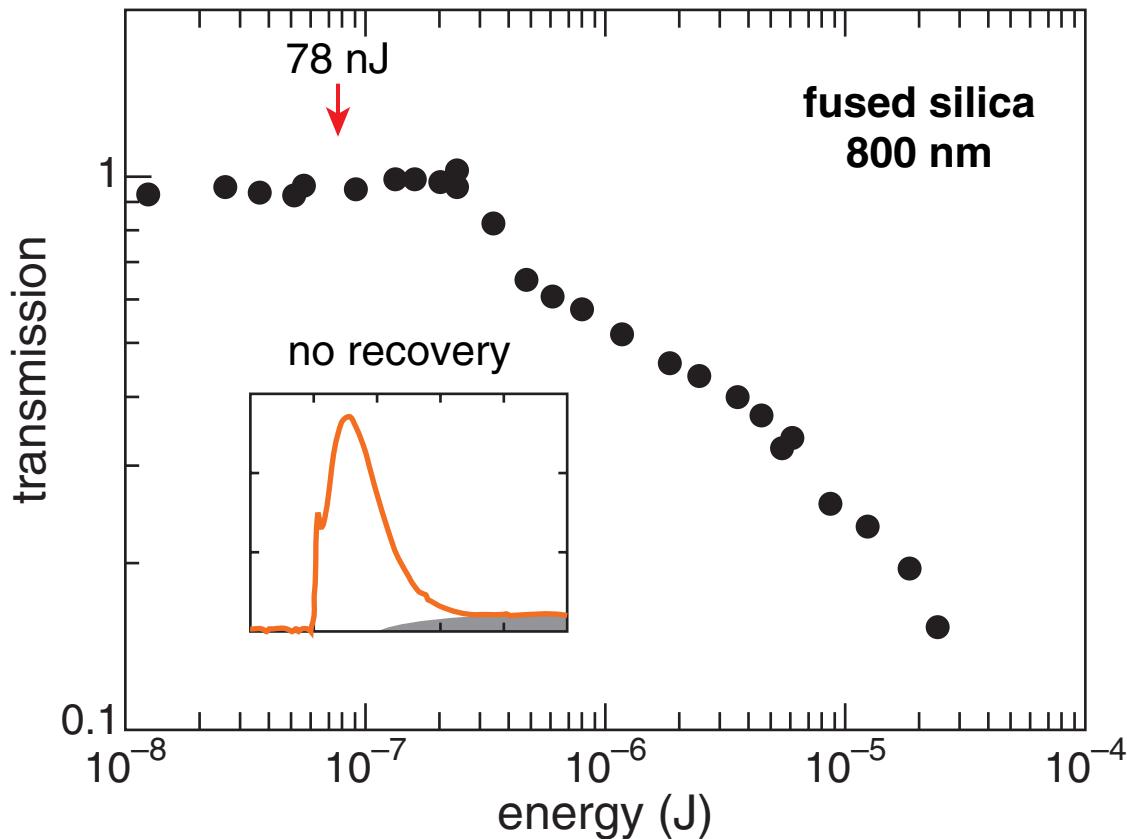
Energy deposition

transmission of pump beam in fused silica



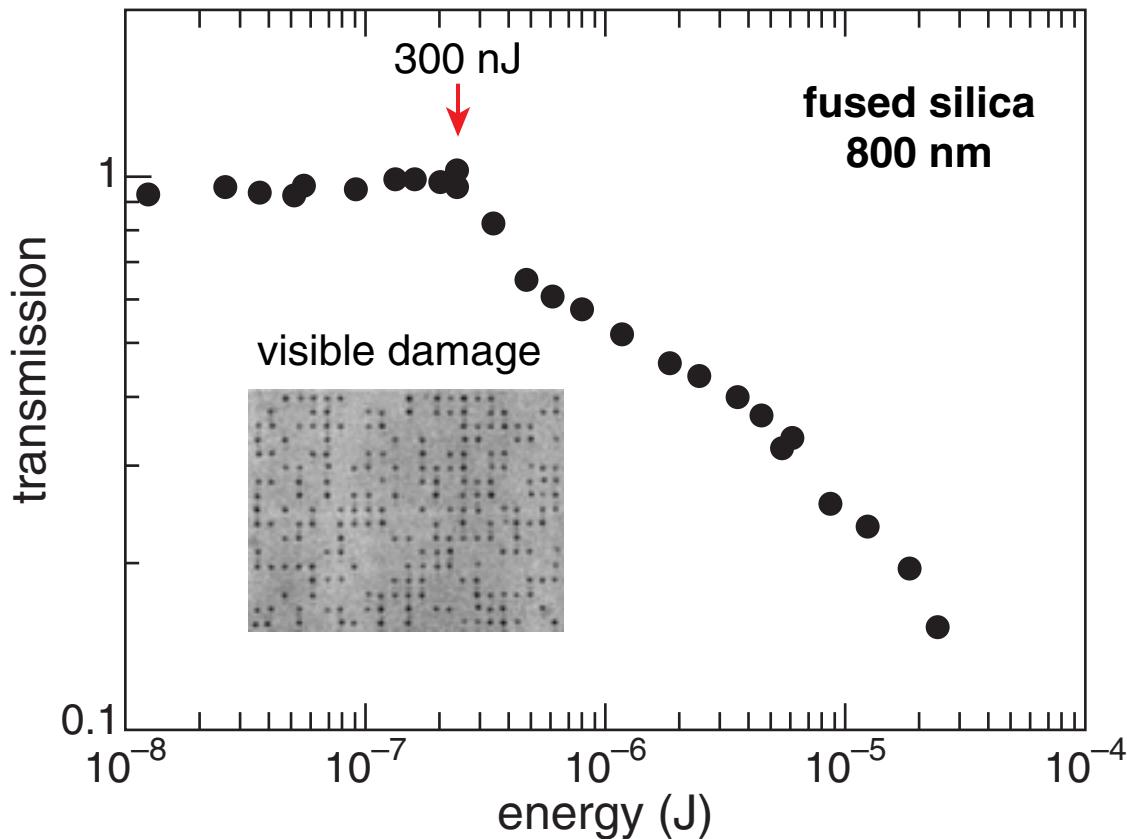
Energy deposition

transmission of pump beam in fused silica



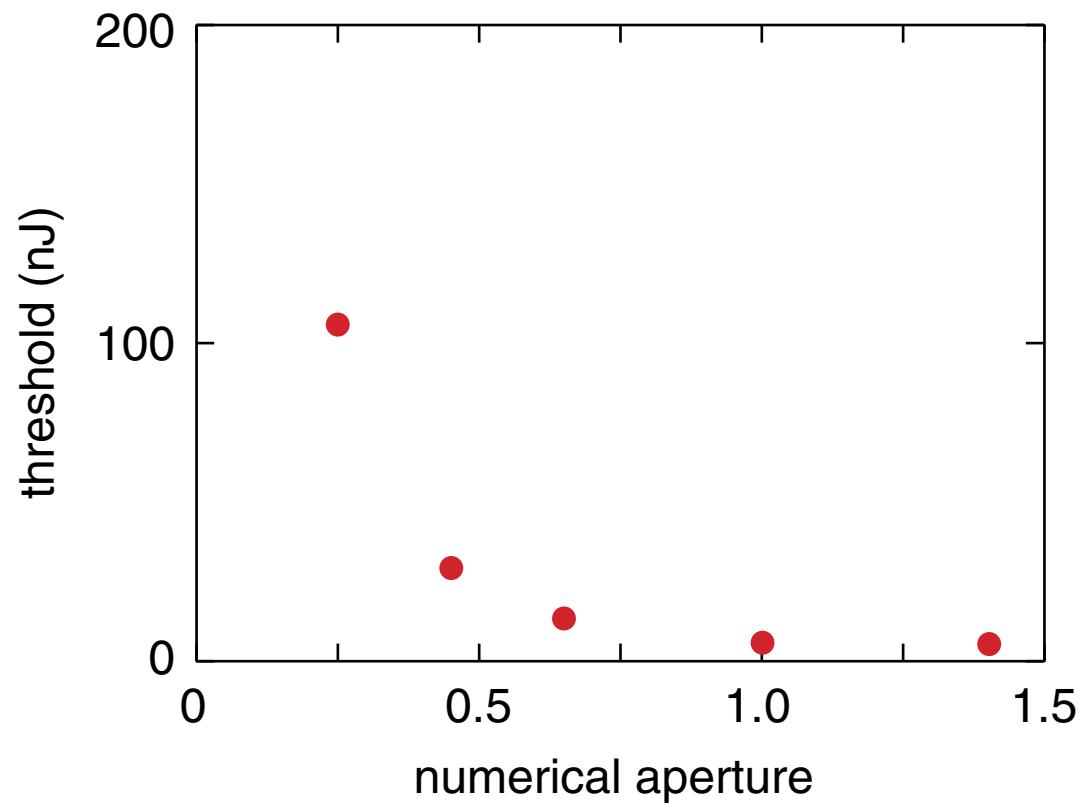
Energy deposition

transmission of pump beam in fused silica



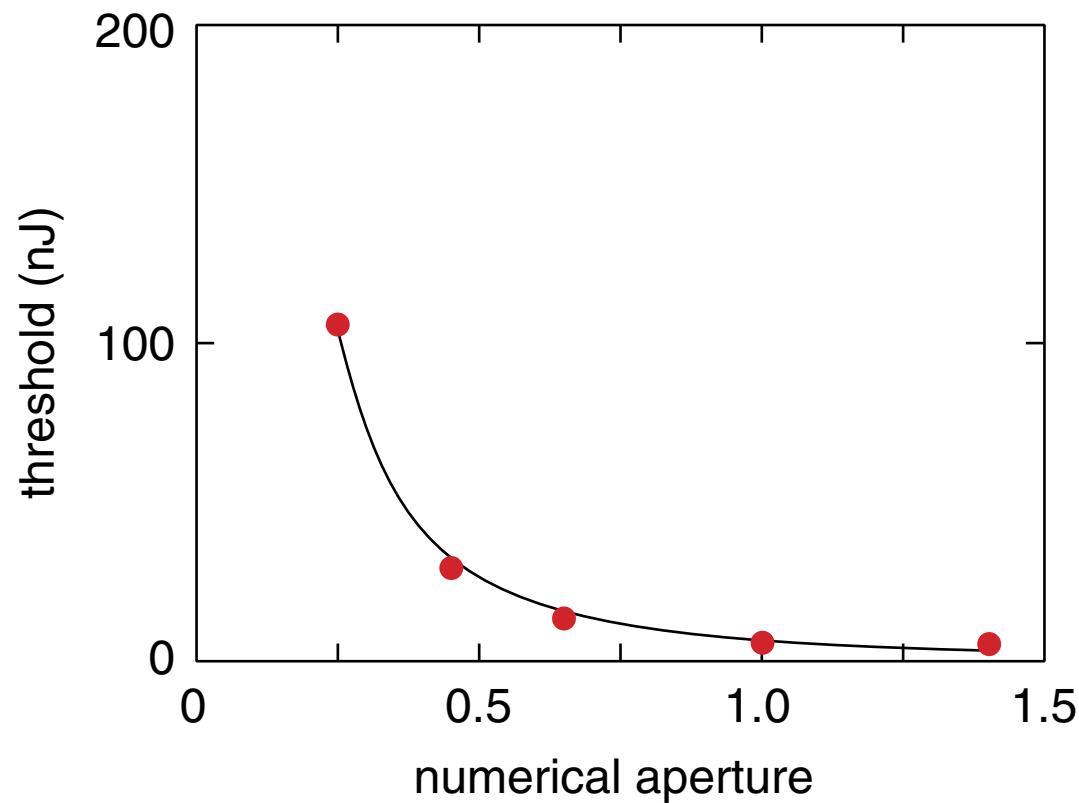
Energy deposition

vary numerical aperture in Corning 0211



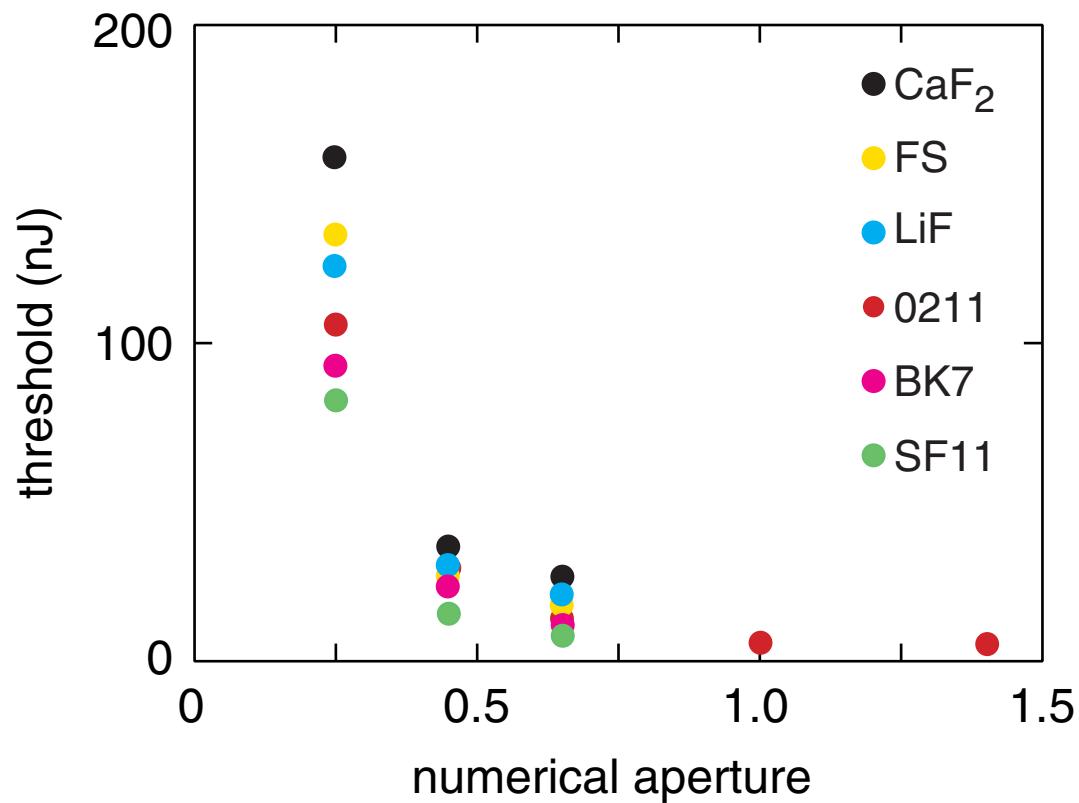
Energy deposition

fit gives threshold intensity: $I_o = 2.7 \times 10^{17} \text{ W/m}^2$



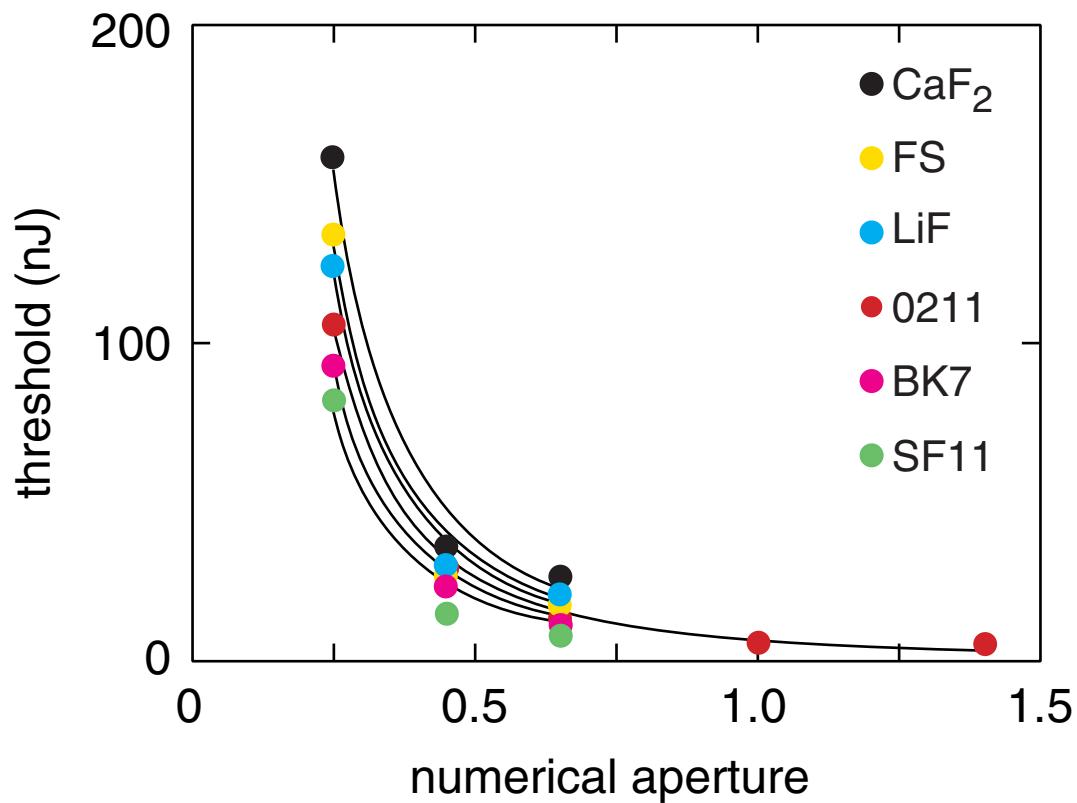
Energy deposition

other materials...



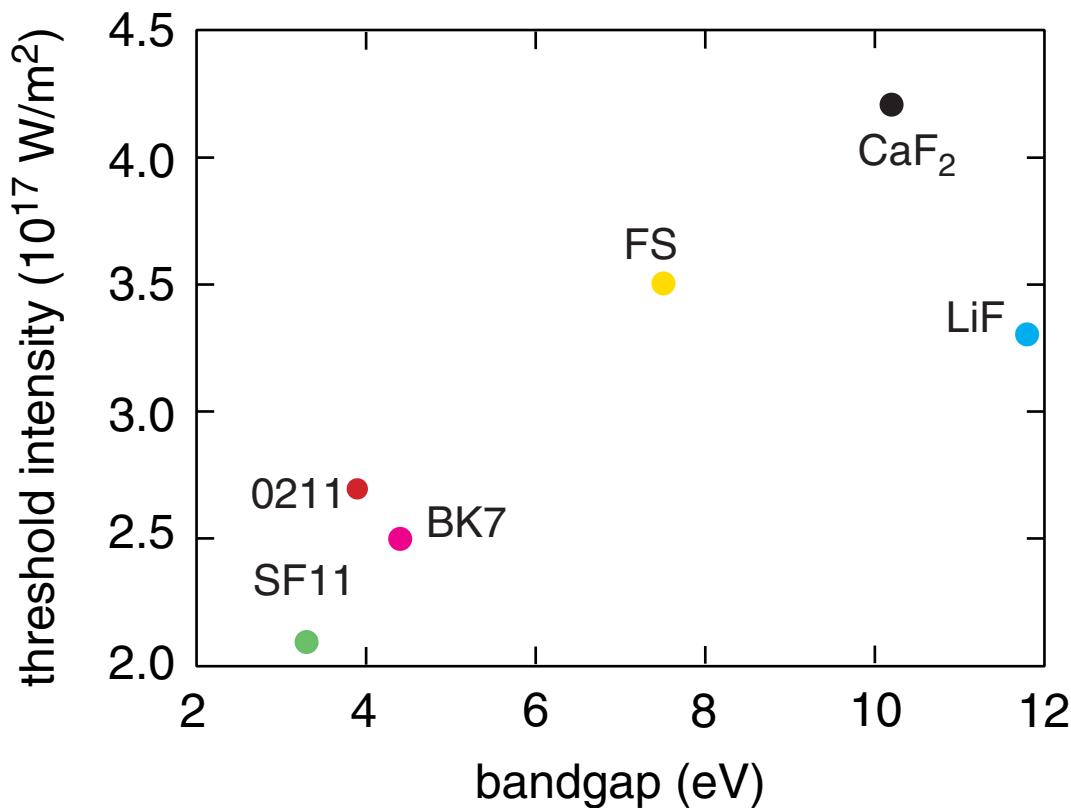
Energy deposition

... give other thresholds



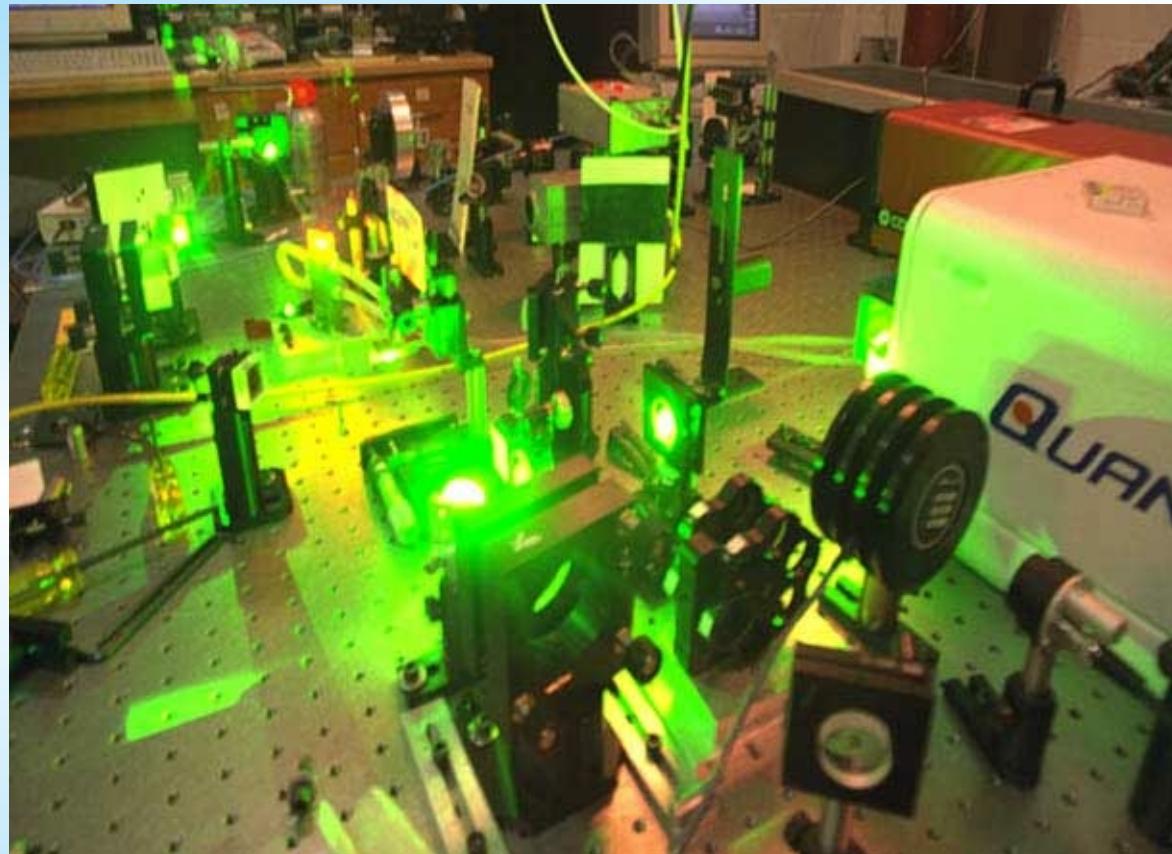
Energy deposition

threshold increases with bandgap



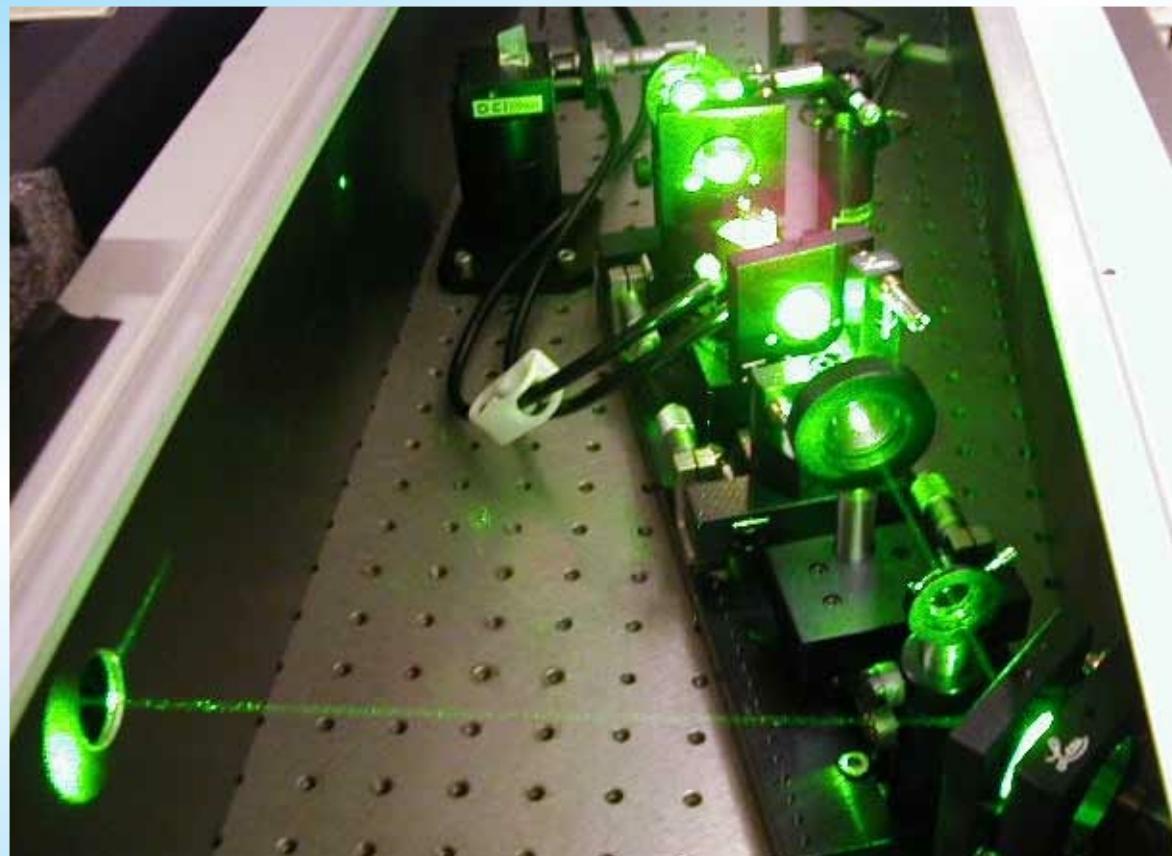
Energy deposition

10-nJ threshold: unamplified micromachining



Energy deposition

10-nJ threshold: unamplified micromachining



Energy deposition

- ▶ **plasma below damage threshold**
- ▶ **damage with only tens of nanojoules**
- ▶ **weak dependence on bandgap**
- ▶ **no shot-to-shot variation**

Outline

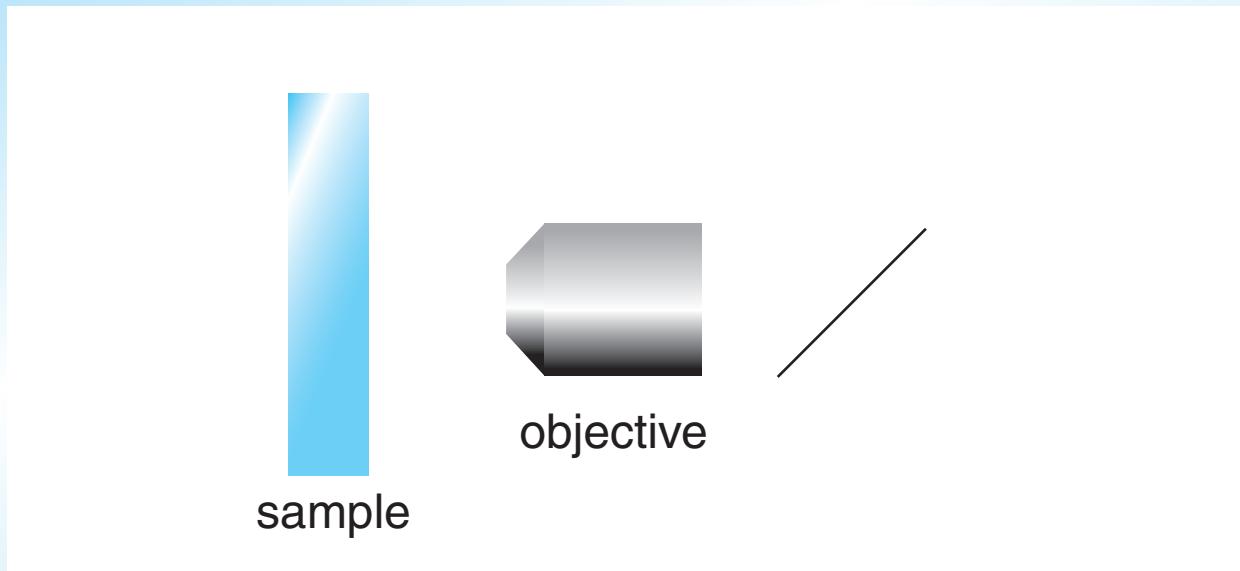
- ▶ Post-mortem analysis
- ▶ Energy deposition
- ▶ Microexplosion dynamics

Microexplosion dynamics

what happens after the energy is deposited?

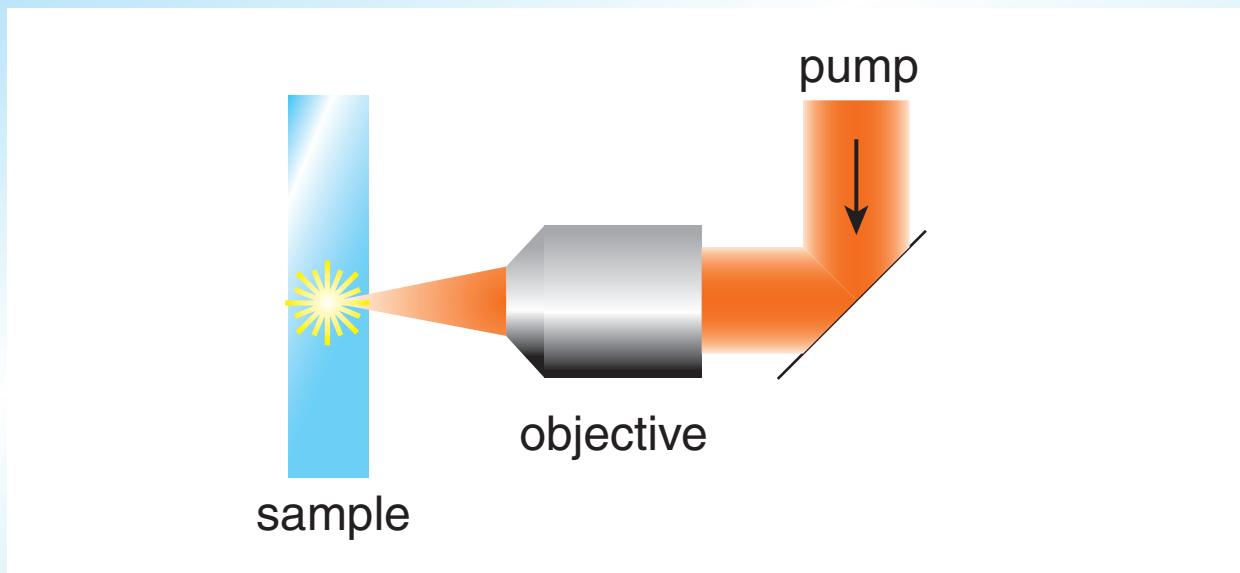
Microexplosion dynamics

imaging setup



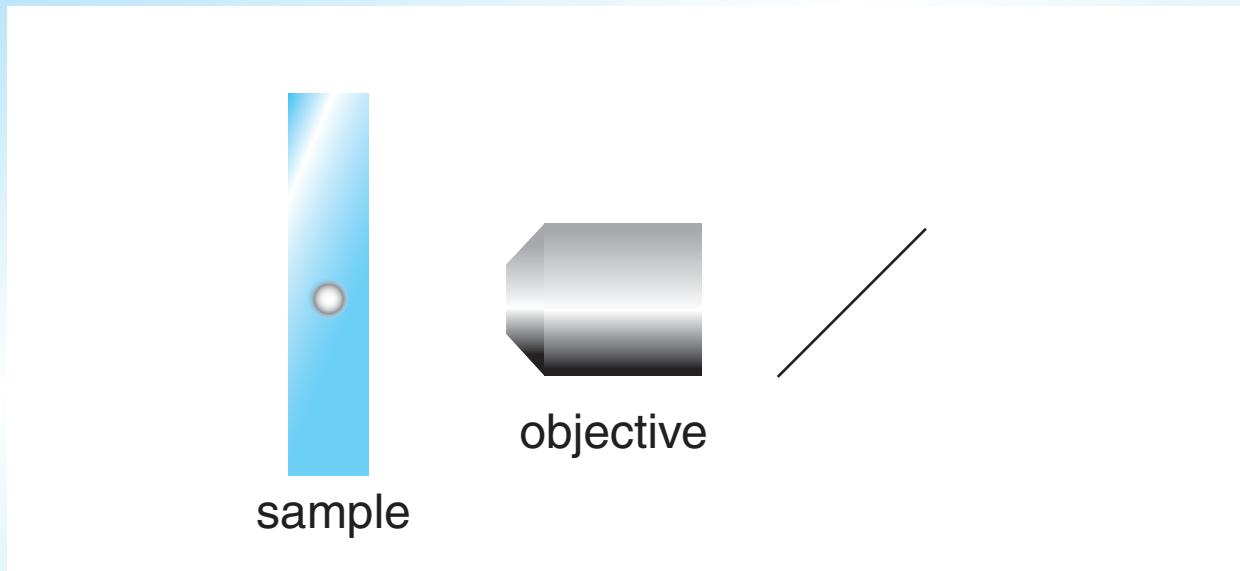
Microexplosion dynamics

imaging setup



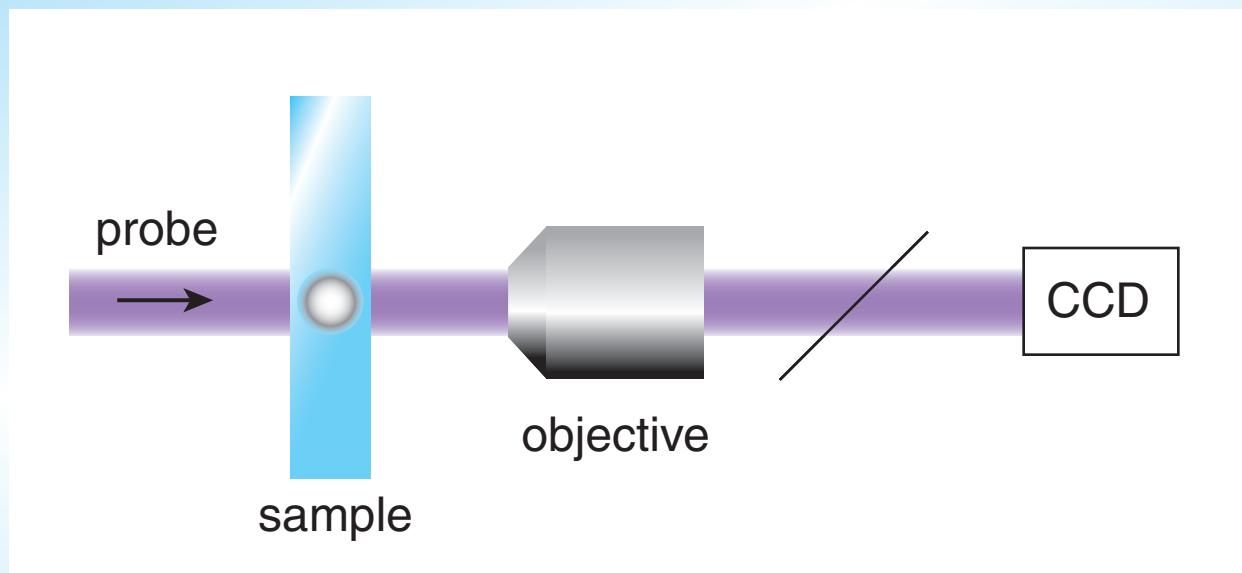
Microexplosion dynamics

imaging setup



Microexplosion dynamics

imaging setup



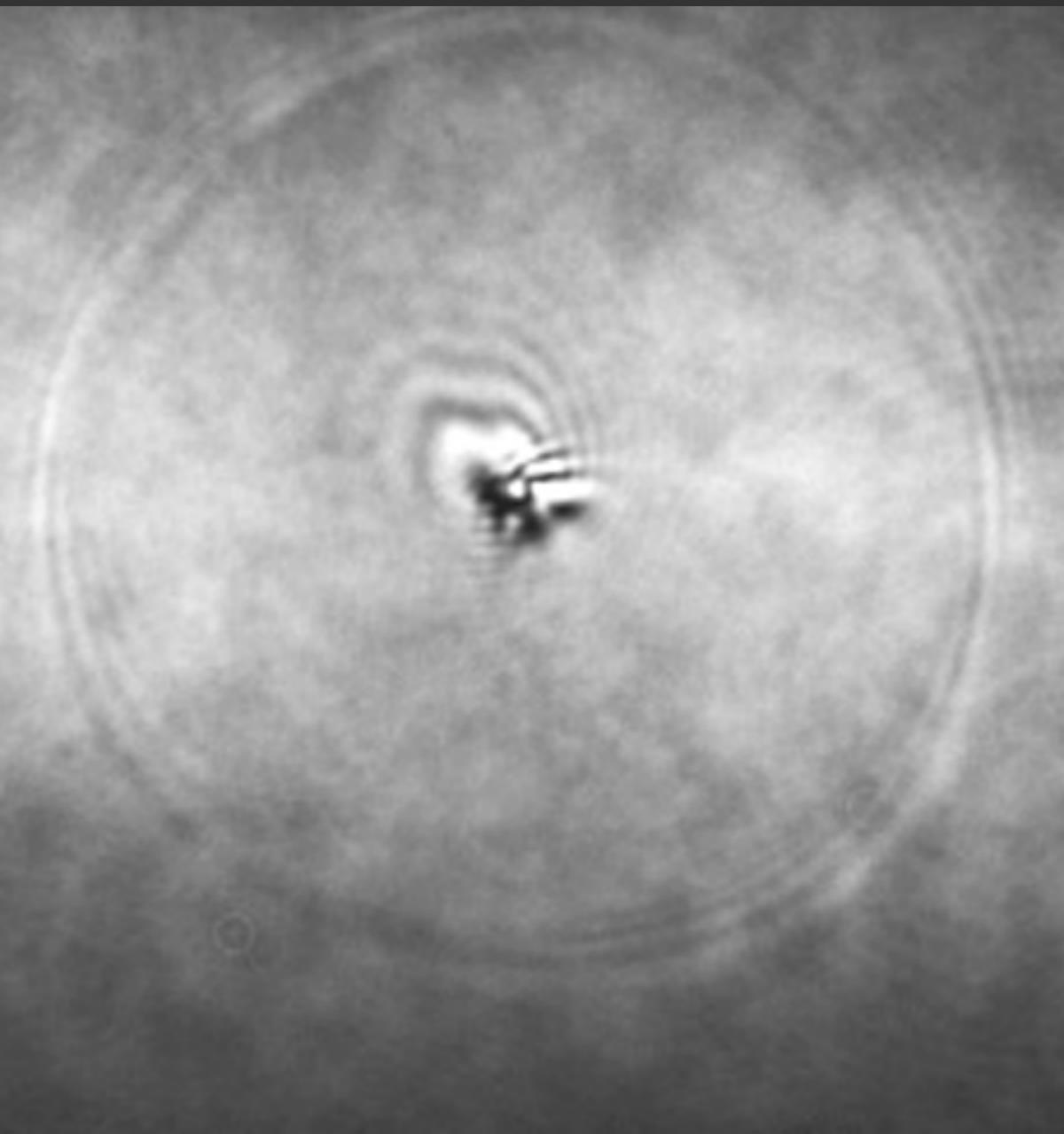
Microexplosion dynamics

sapphire

3 μJ pulse

3.8 ns delay

40 μm radius



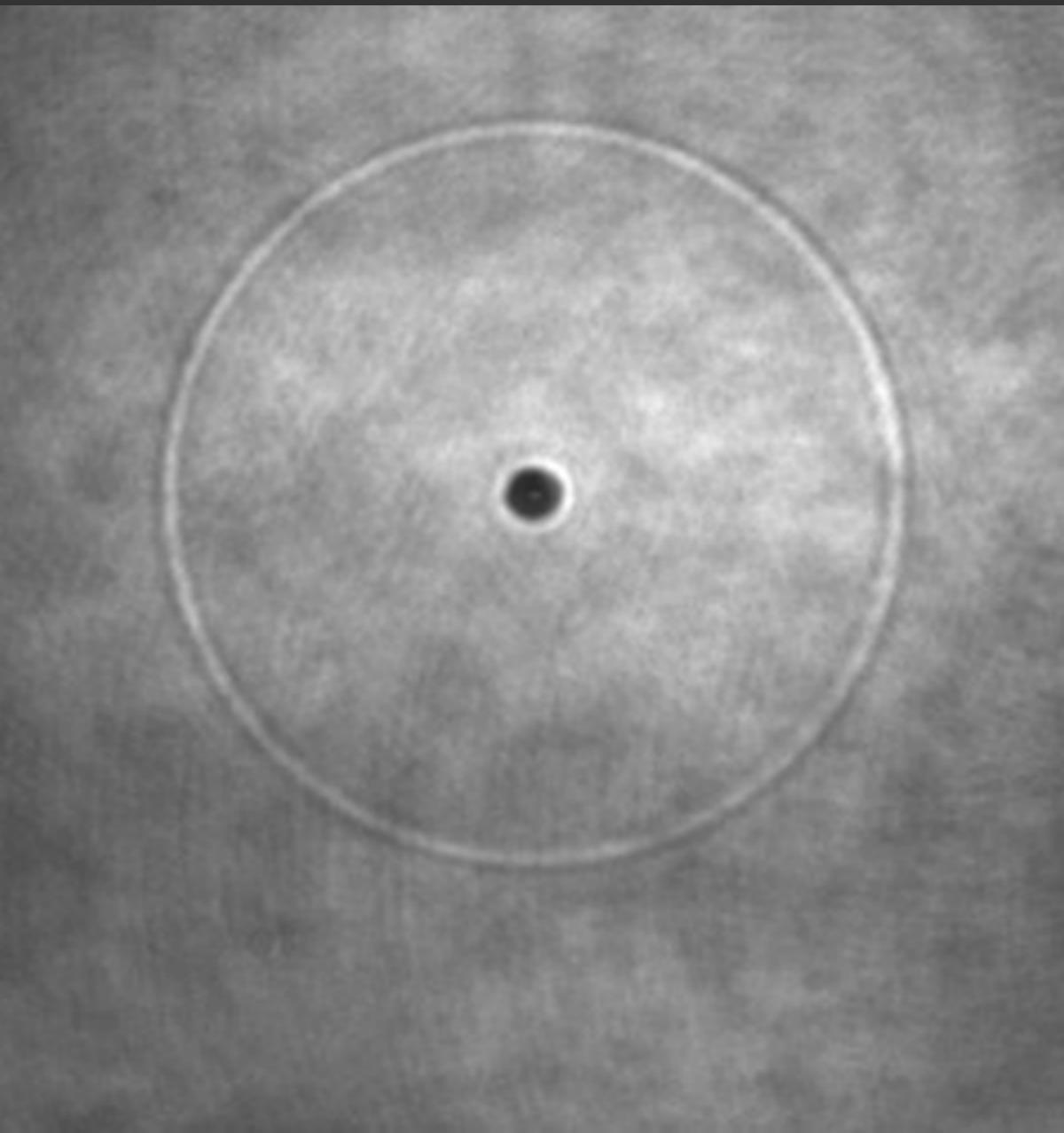
Microexplosion dynamics

water

1.0 μJ pulse

35 ns delay

58 μm radius



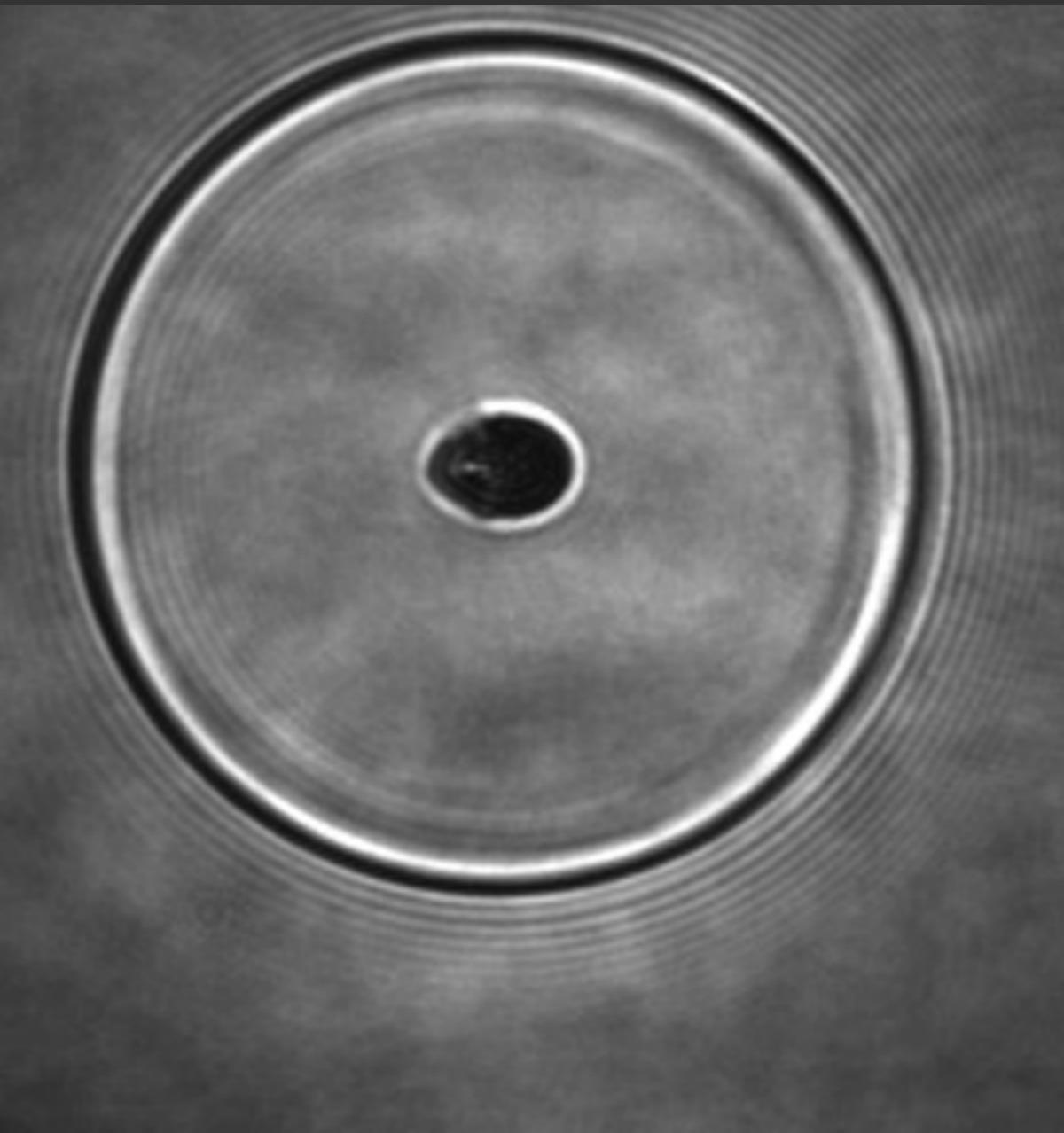
Microexplosion dynamics

water

14 μJ pulse

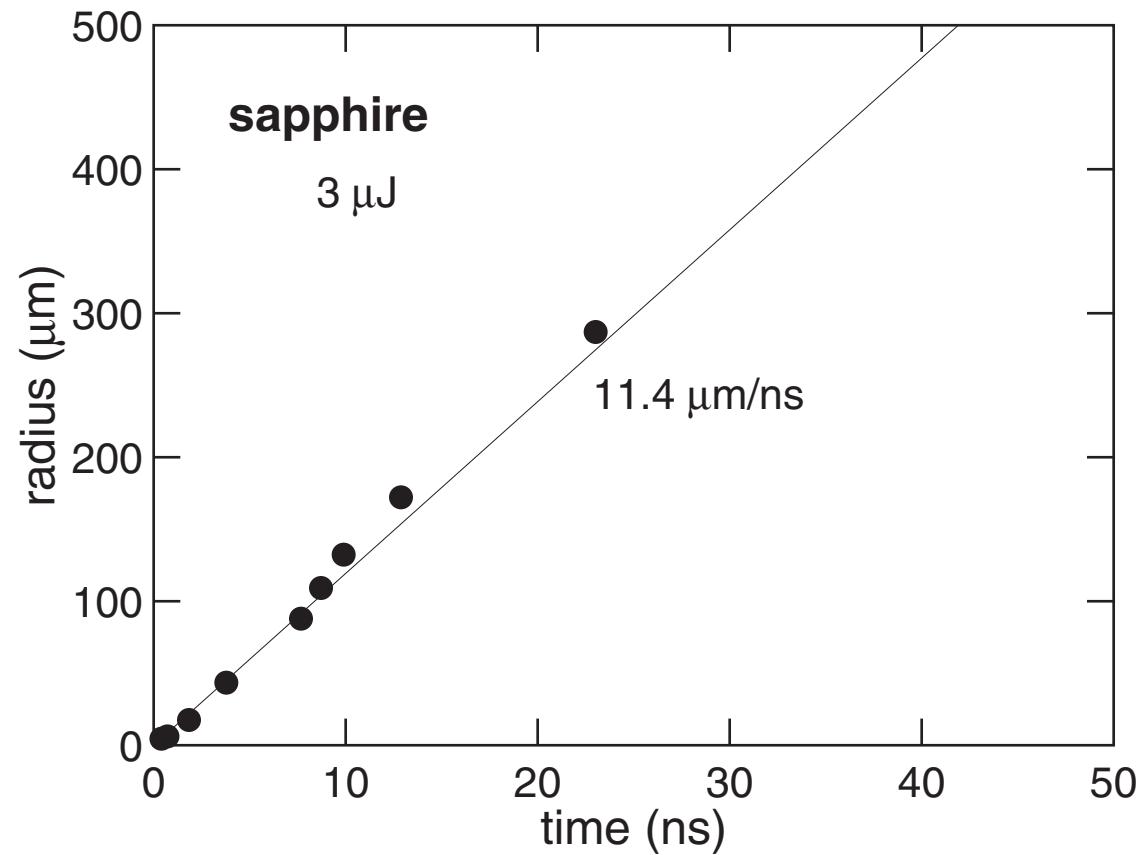
35 ns delay

64 μm radius

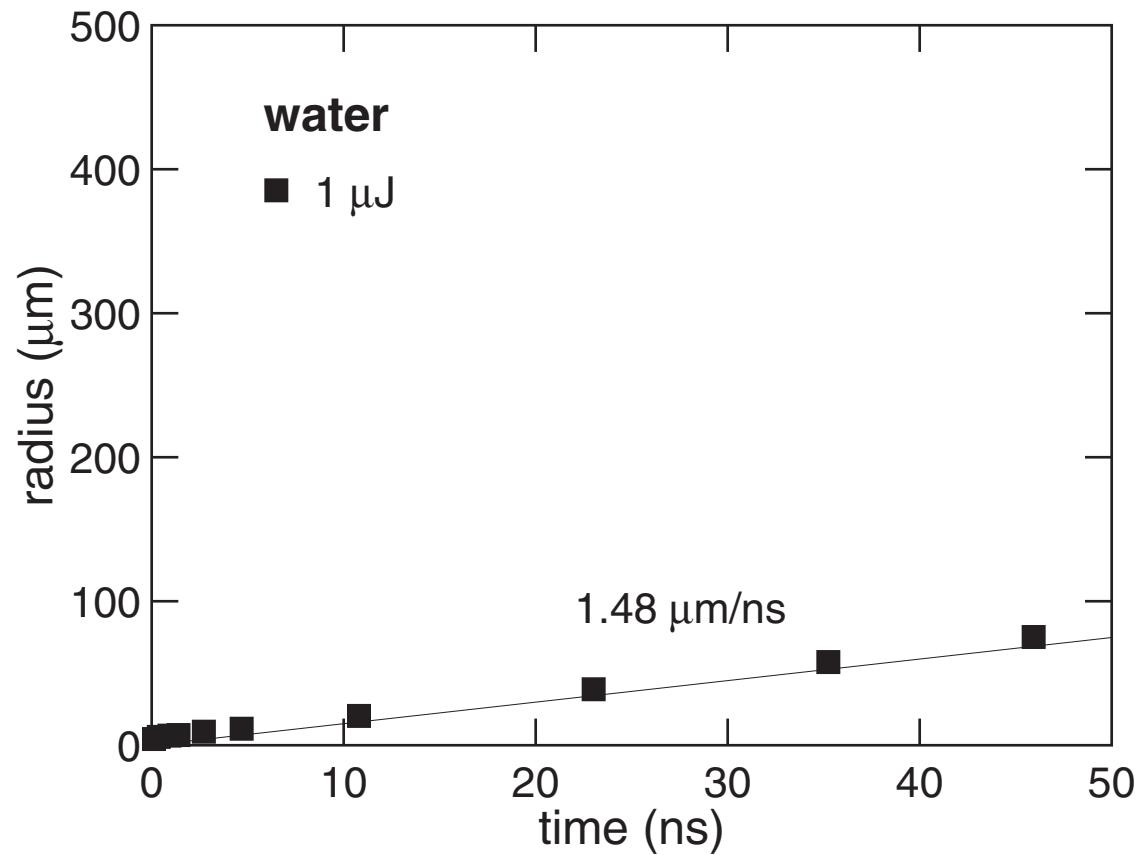


Microexplosion dynamics

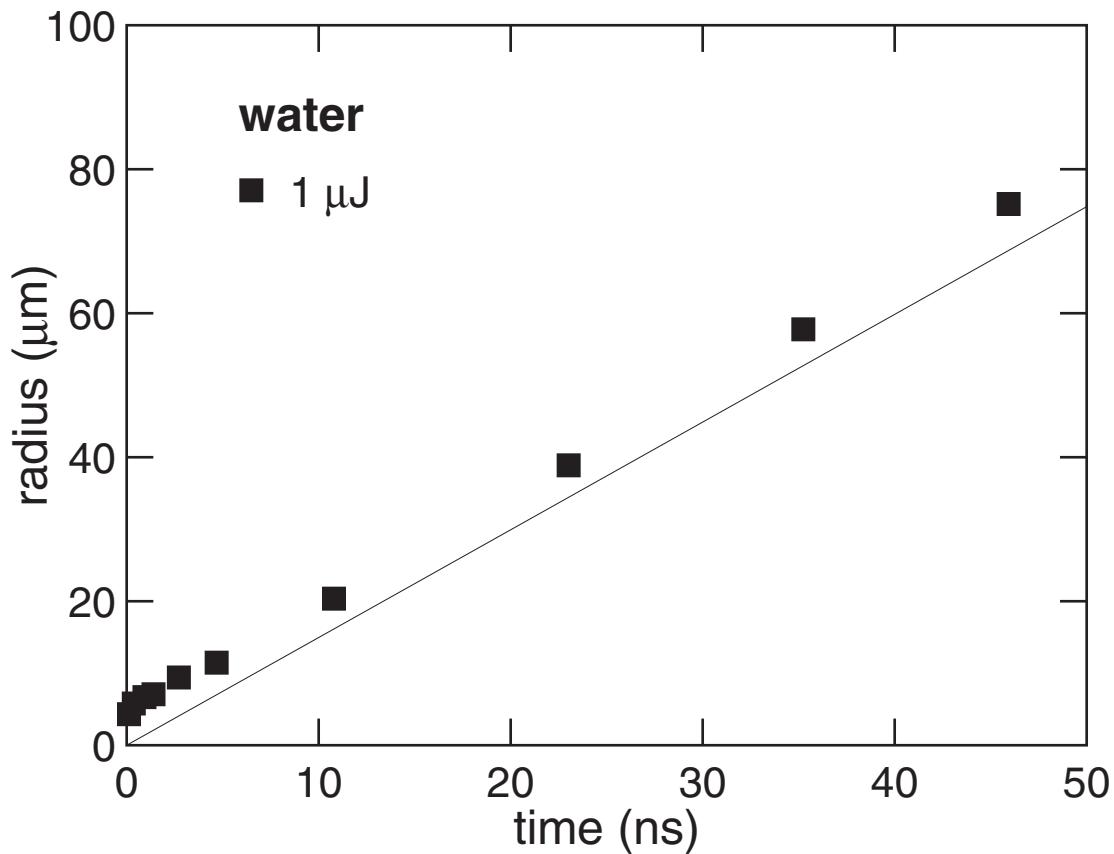
Microexplosion dynamics



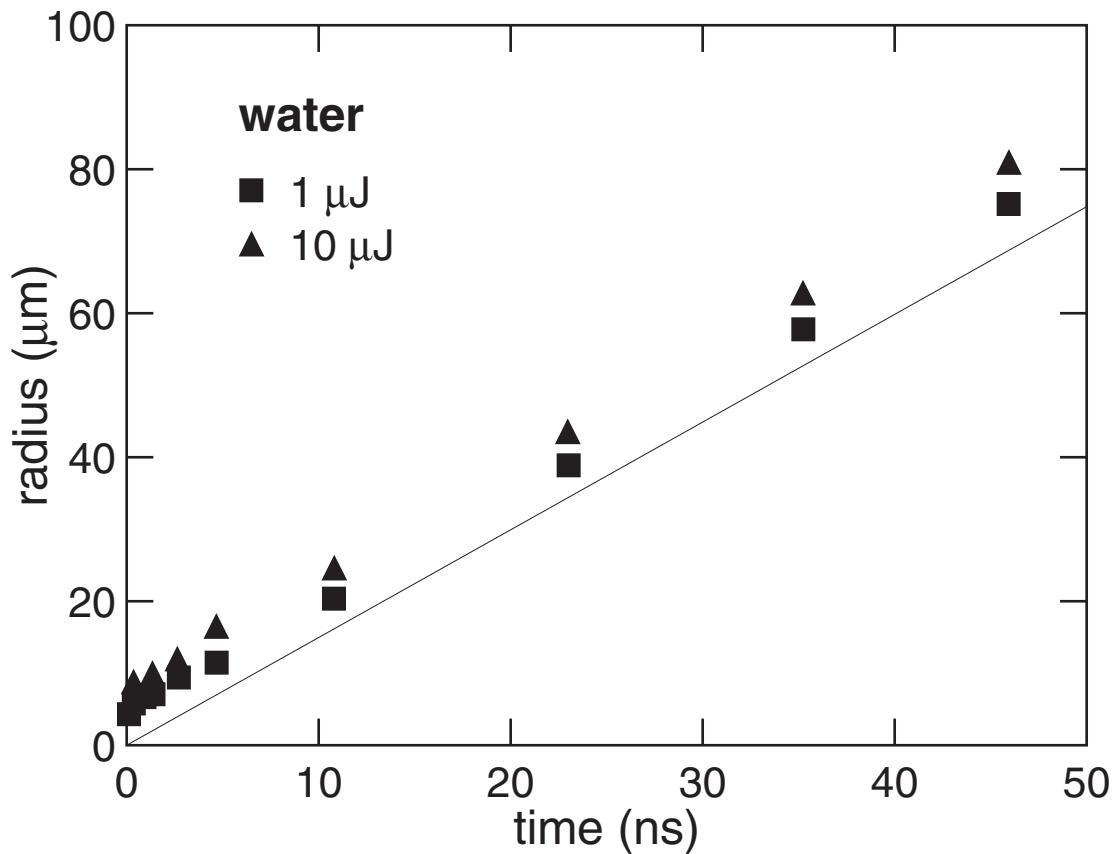
Microexplosion dynamics



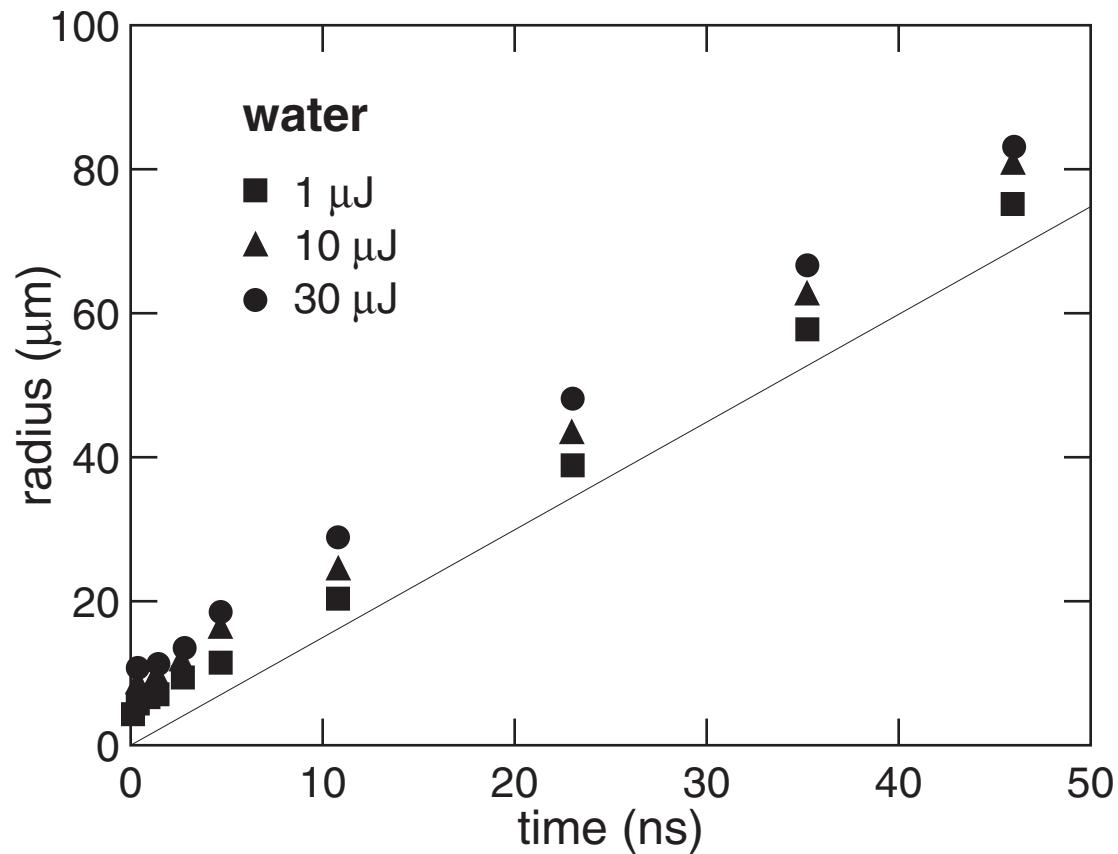
Microexplosion dynamics



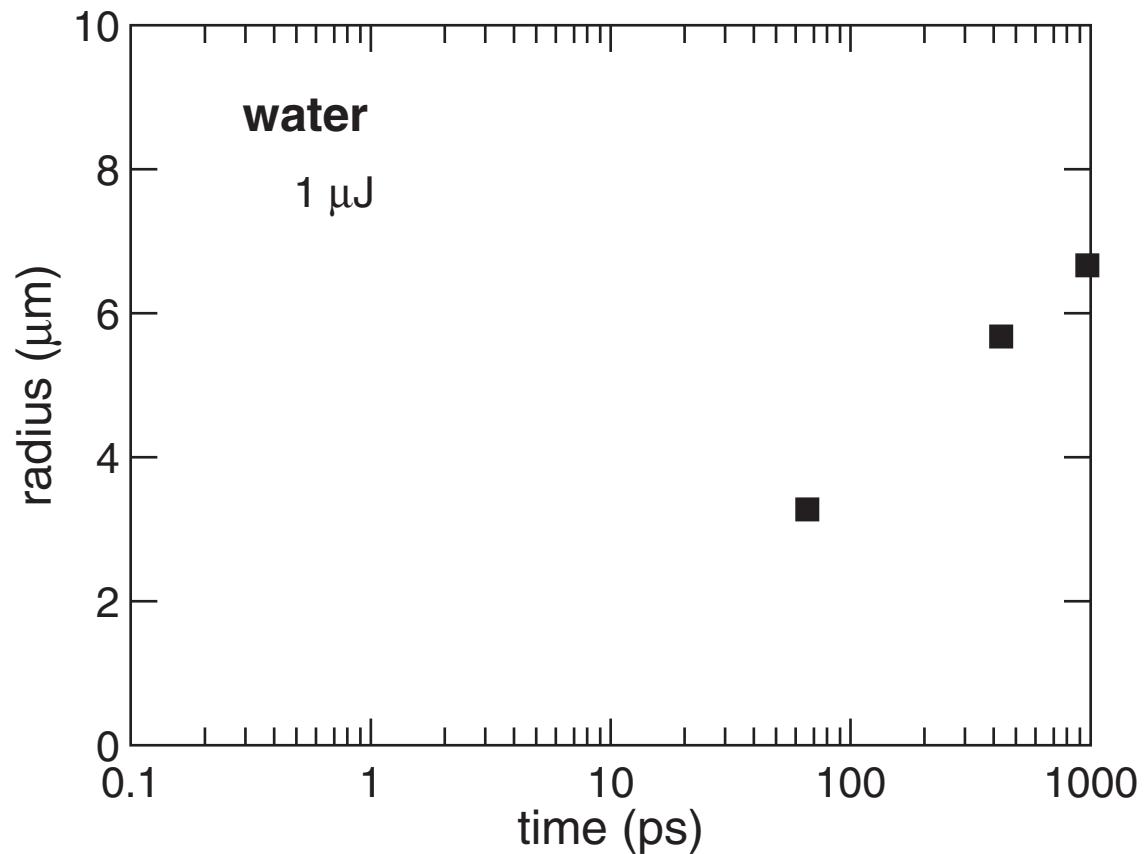
Microexplosion dynamics



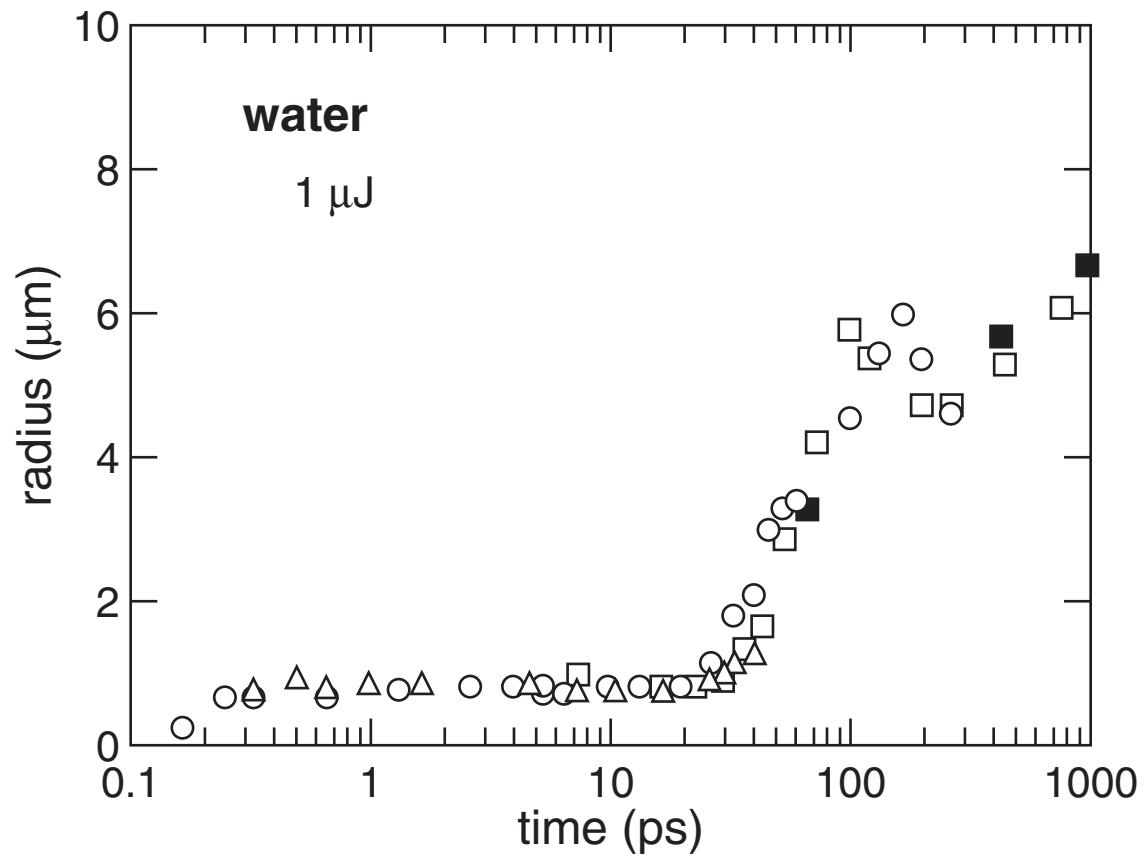
Microexplosion dynamics



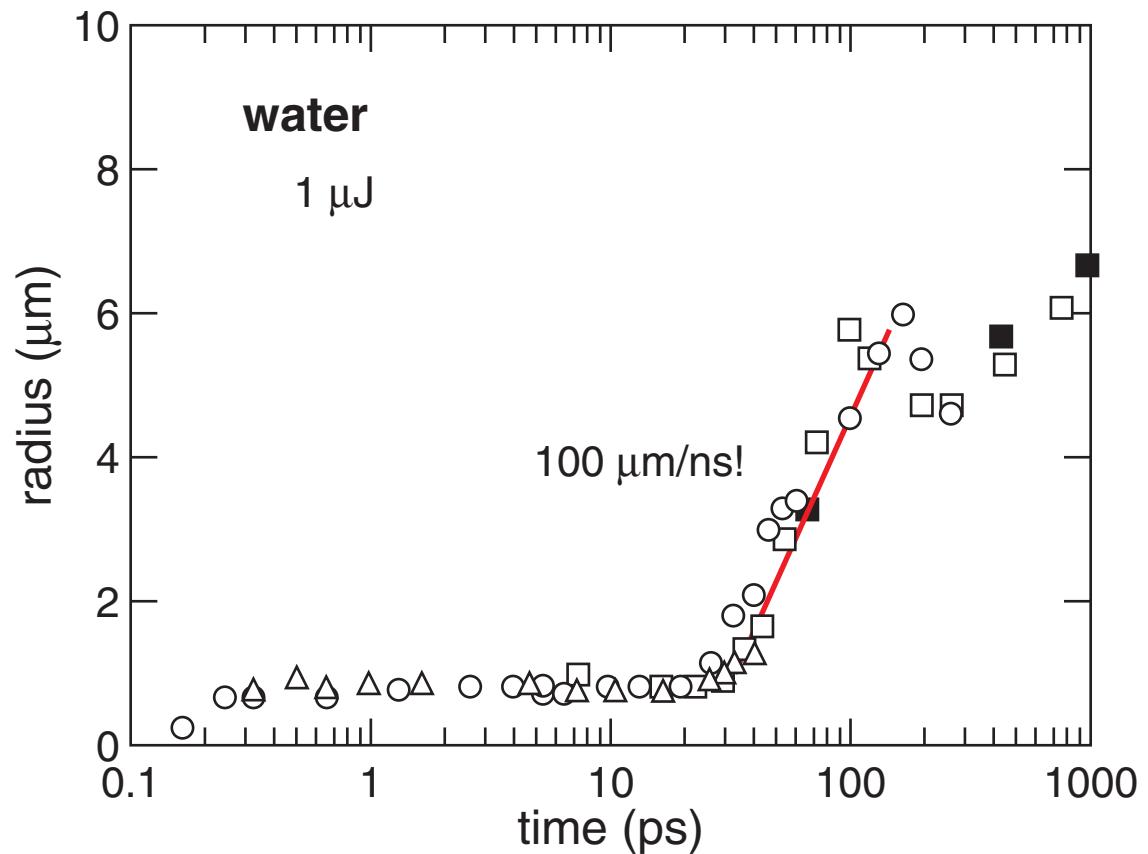
Microexplosion dynamics



Microexplosion dynamics



Microexplosion dynamics



Summary

- ▶ **extreme conditions with only nanojoules**
- ▶ **microstructuring without amplifiers**
- ▶ **view into dynamics**

Applications

- ▶ **data storage (17 GBits/cm³)**
- ▶ **internal microstructuring**
- ▶ **microsurgery**

Questions

- ▶ **stellar conditions?**
- ▶ **material dependence?**
- ▶ **models?**

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LABORATORY OF
APPLIED SCIENCE



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