Creation, Characterization and Applications of Ultrashort Laser Pulses

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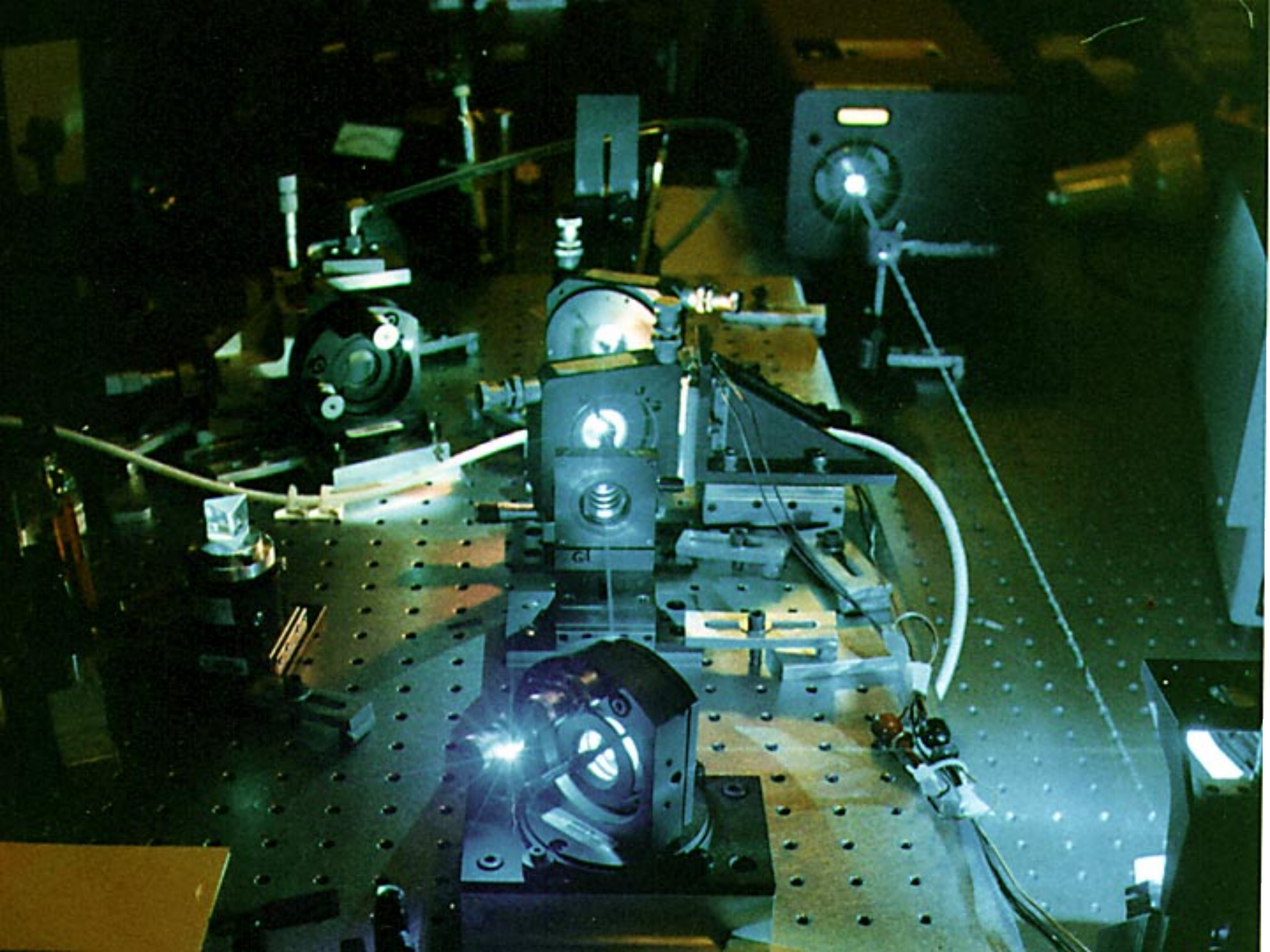
Spectroscopy of systems with confined spatial structures
20 June 2001



Outline

- linear and nonlinear propagation
- femtosecond measurements
- dynamics of phase transitions
- laser-assisted surface etching

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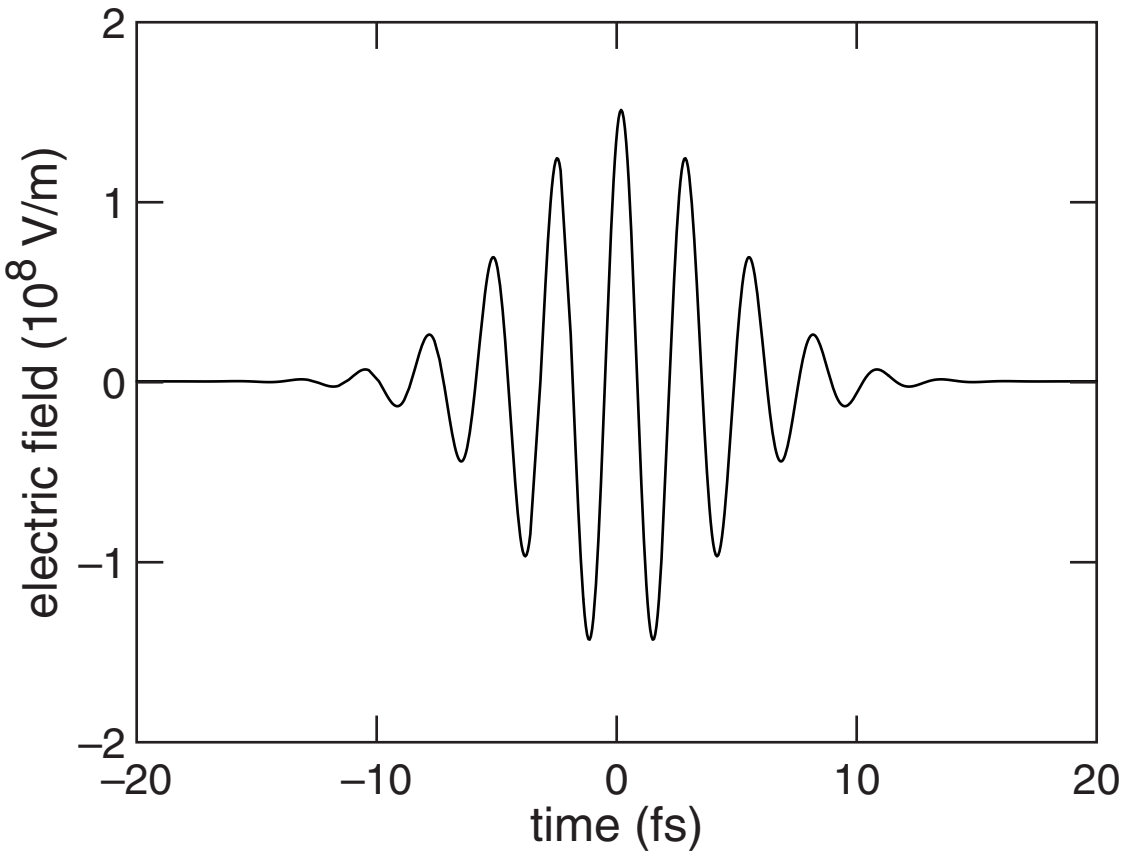
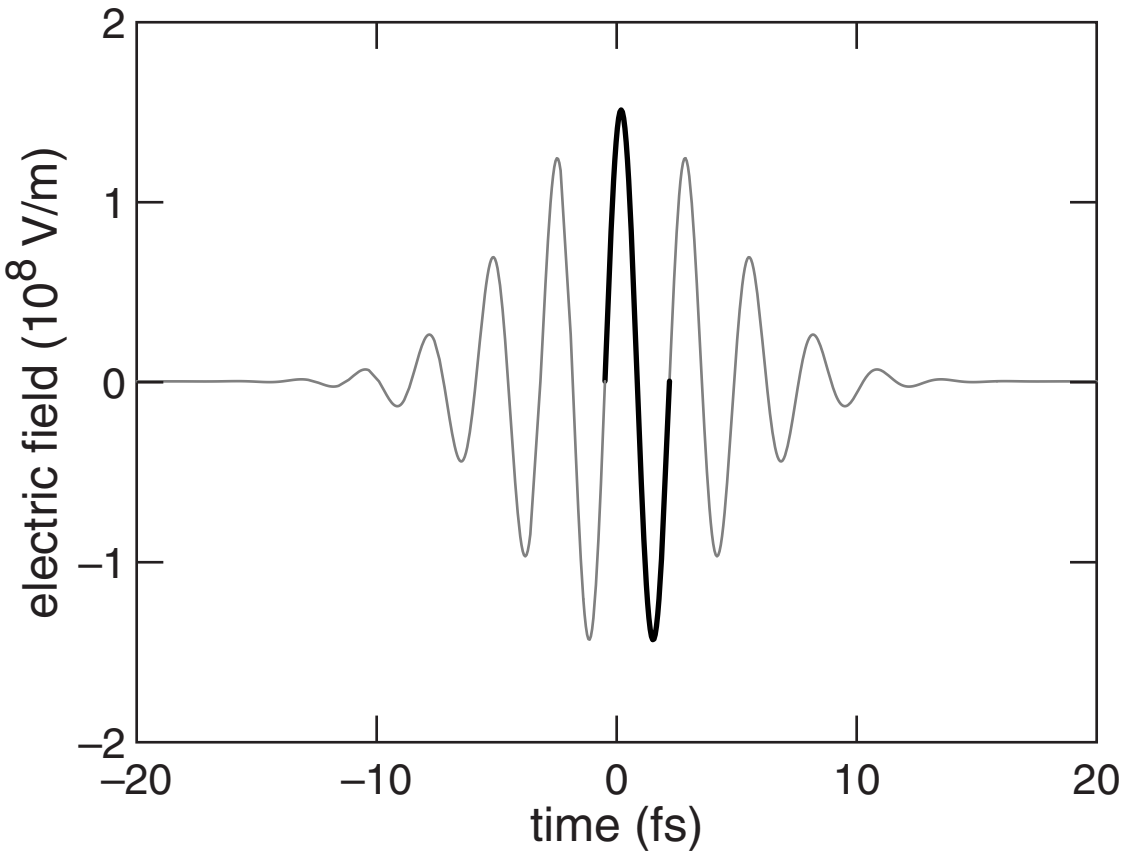
- **linear and nonlinear propagation**
- **femtosecond measurements**
- **dynamics of phase transitions**
- **laser-assisted surface etching**

Introduction



Introduction



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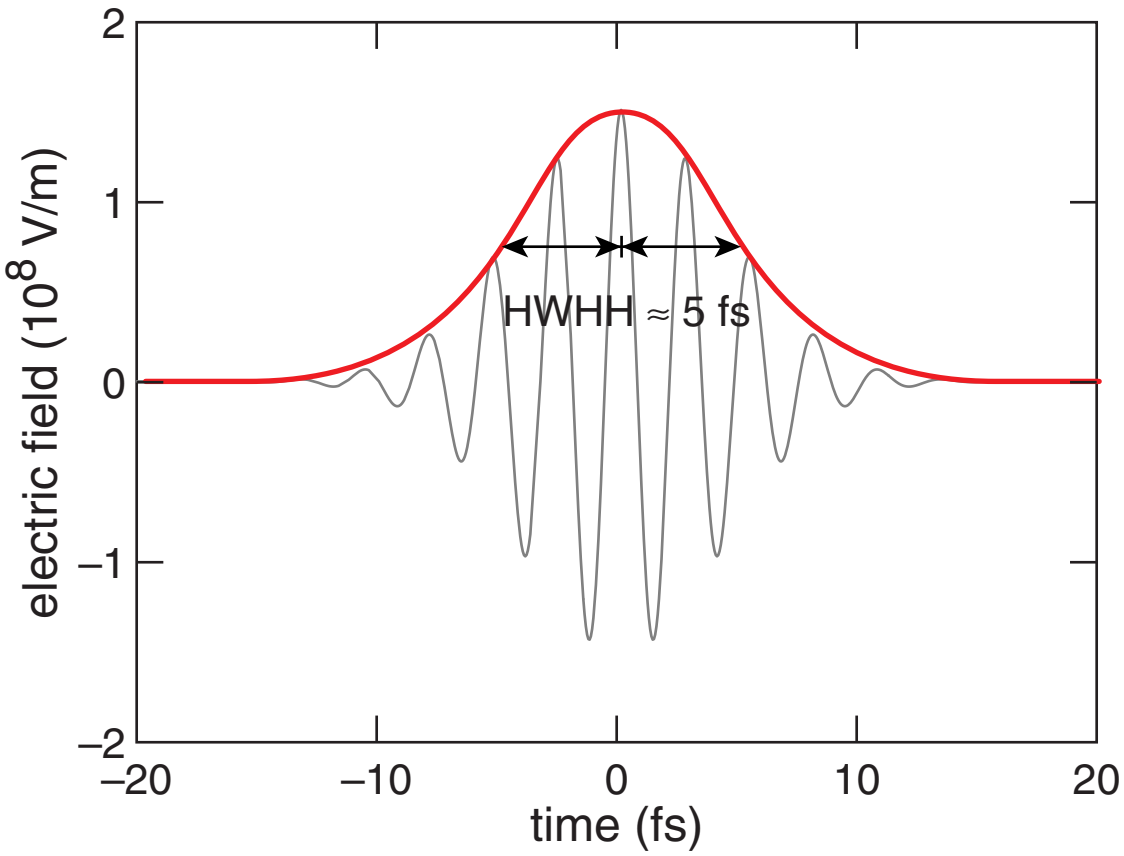

Introduction

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

Propagation of EM waves through medium

Governed by wave equation

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

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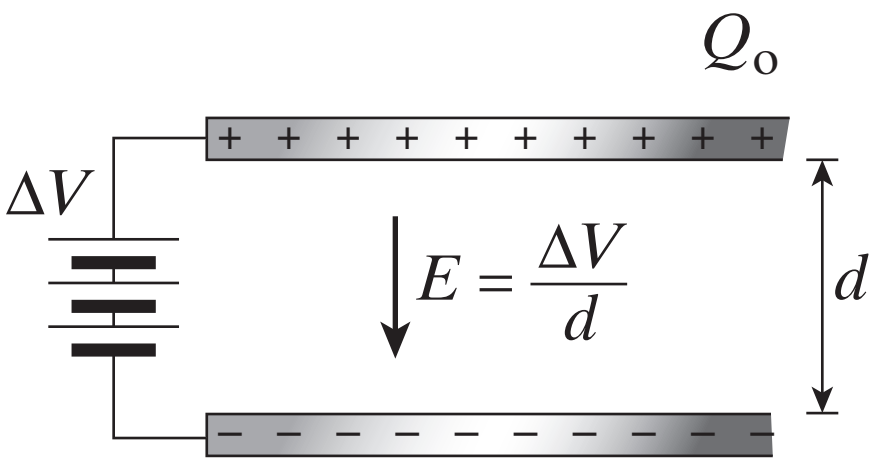
In non-ferromagnetic media $\mu \approx 1$**, and so** $n \approx \sqrt{\epsilon}$**.**

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

Propagation of EM waves through medium

Dielectric constant measures increase in capacitance

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



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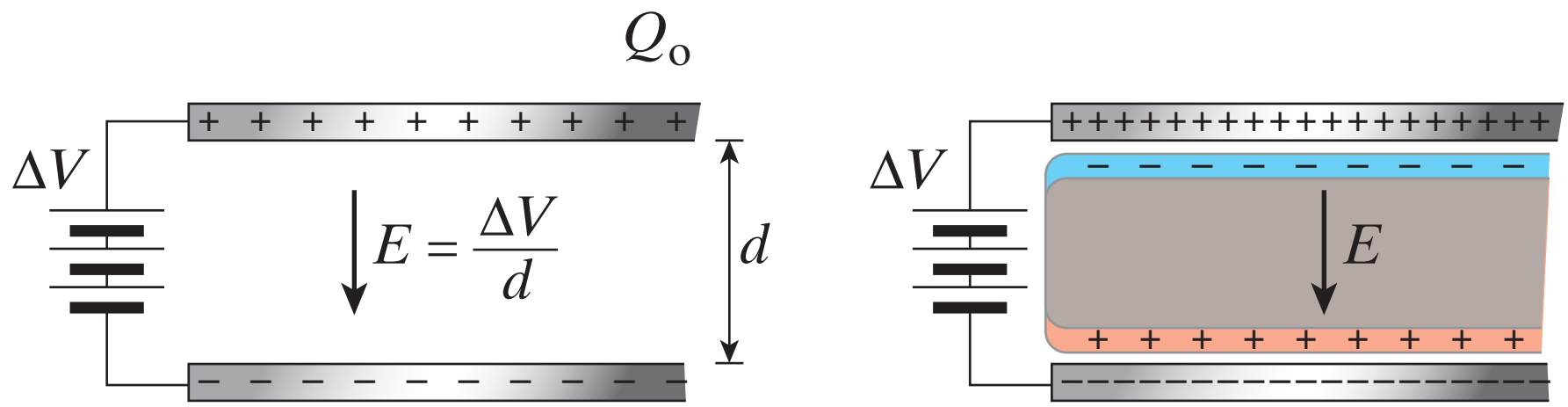

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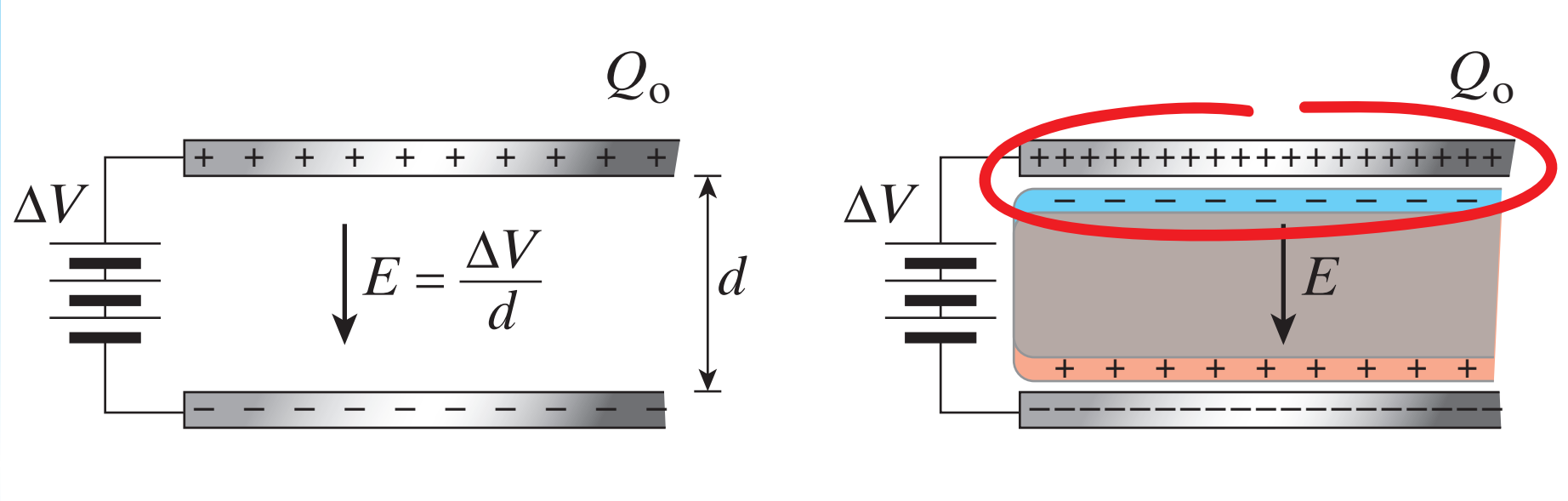

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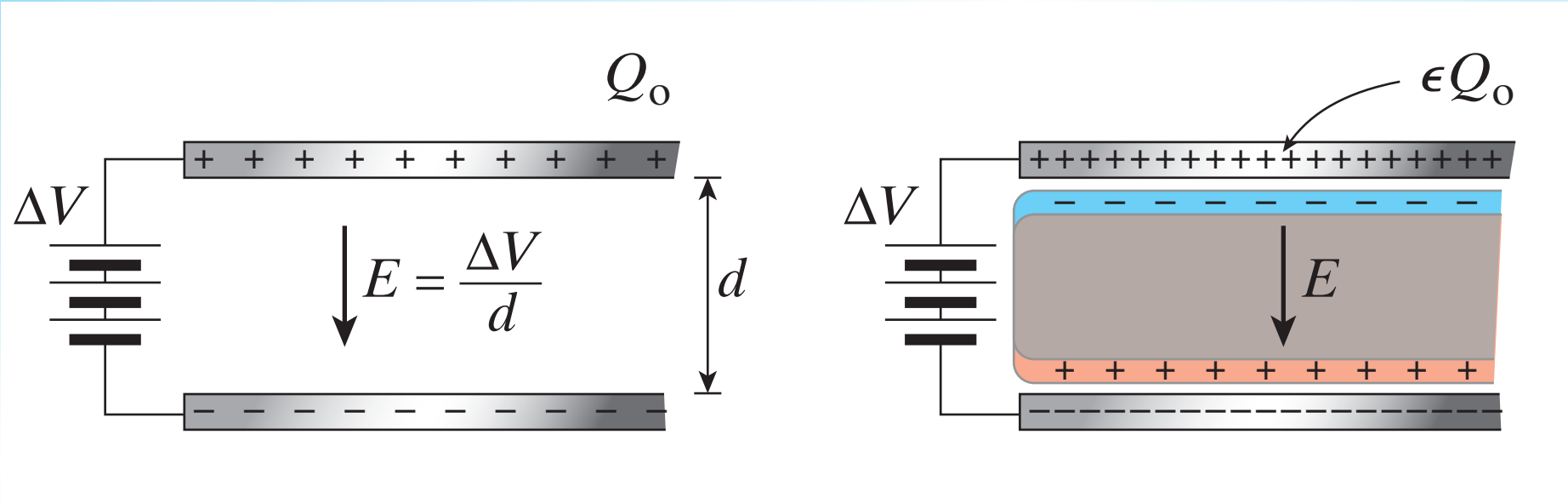
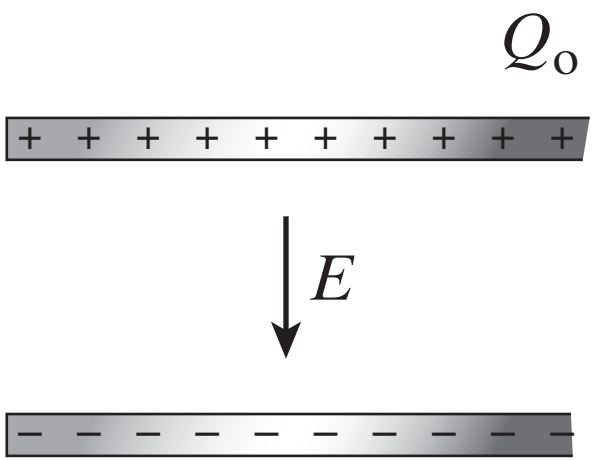


Propagation of EM waves through medium

Alternatively, ϵ is measure of the attenuation of the field



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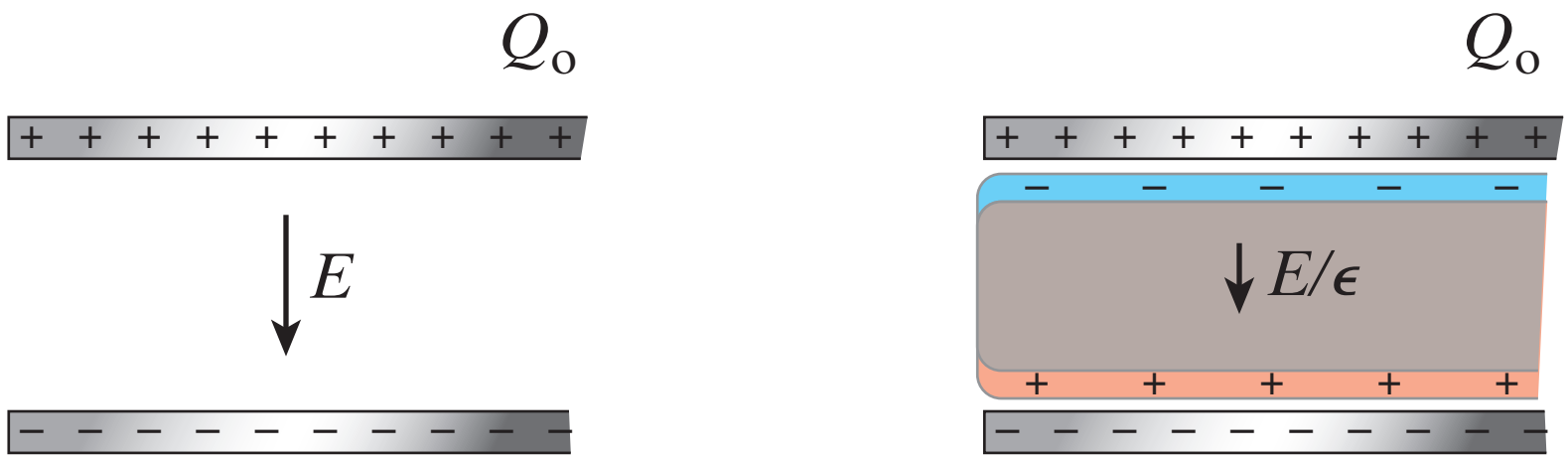
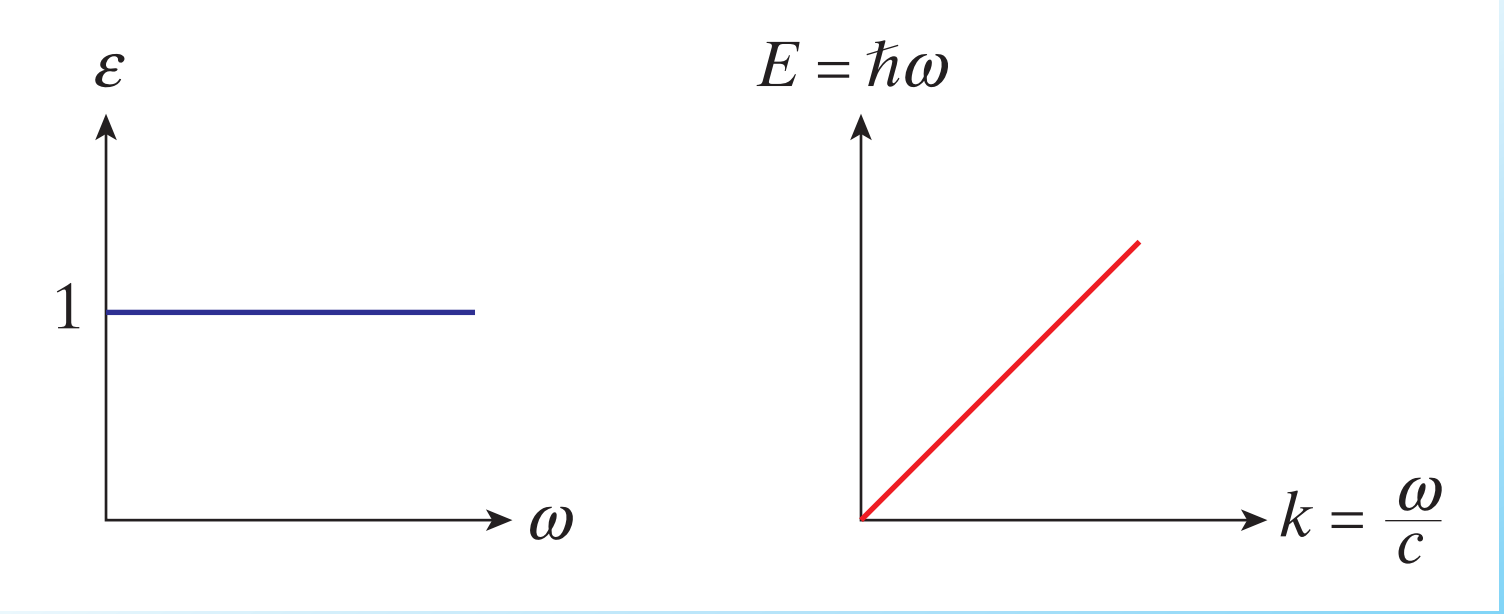


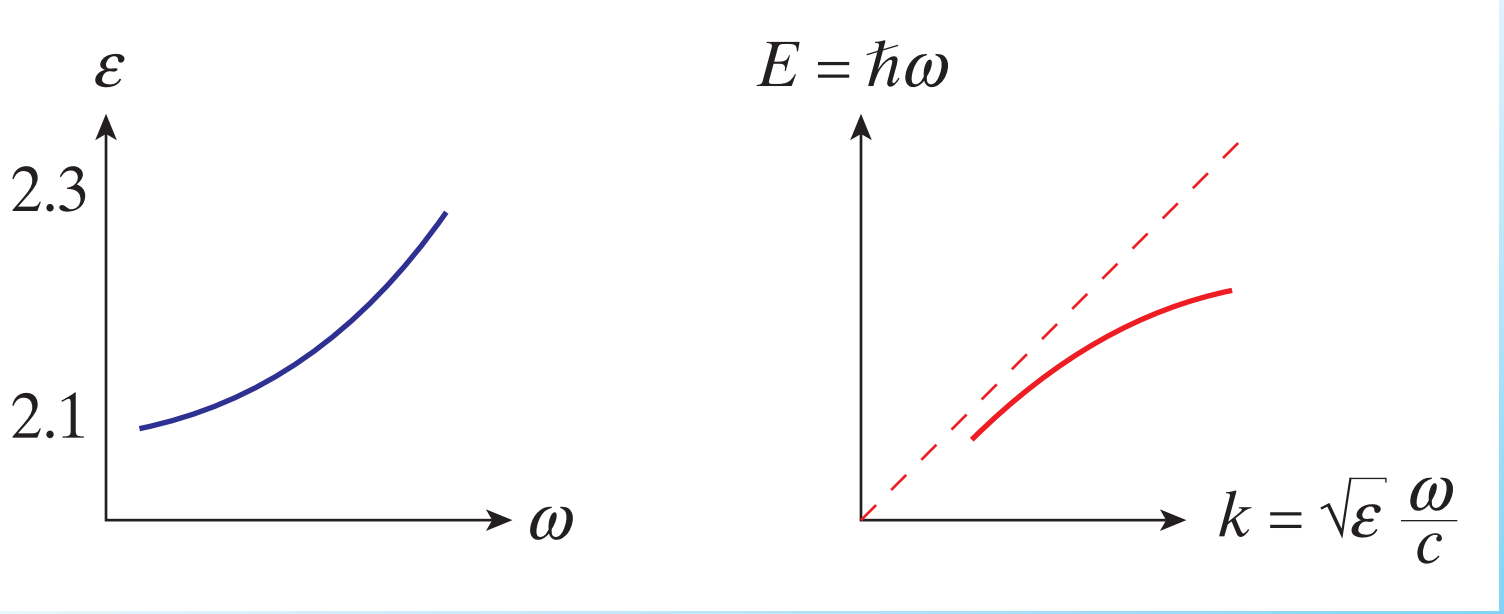
Propagation of EM waves through medium

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

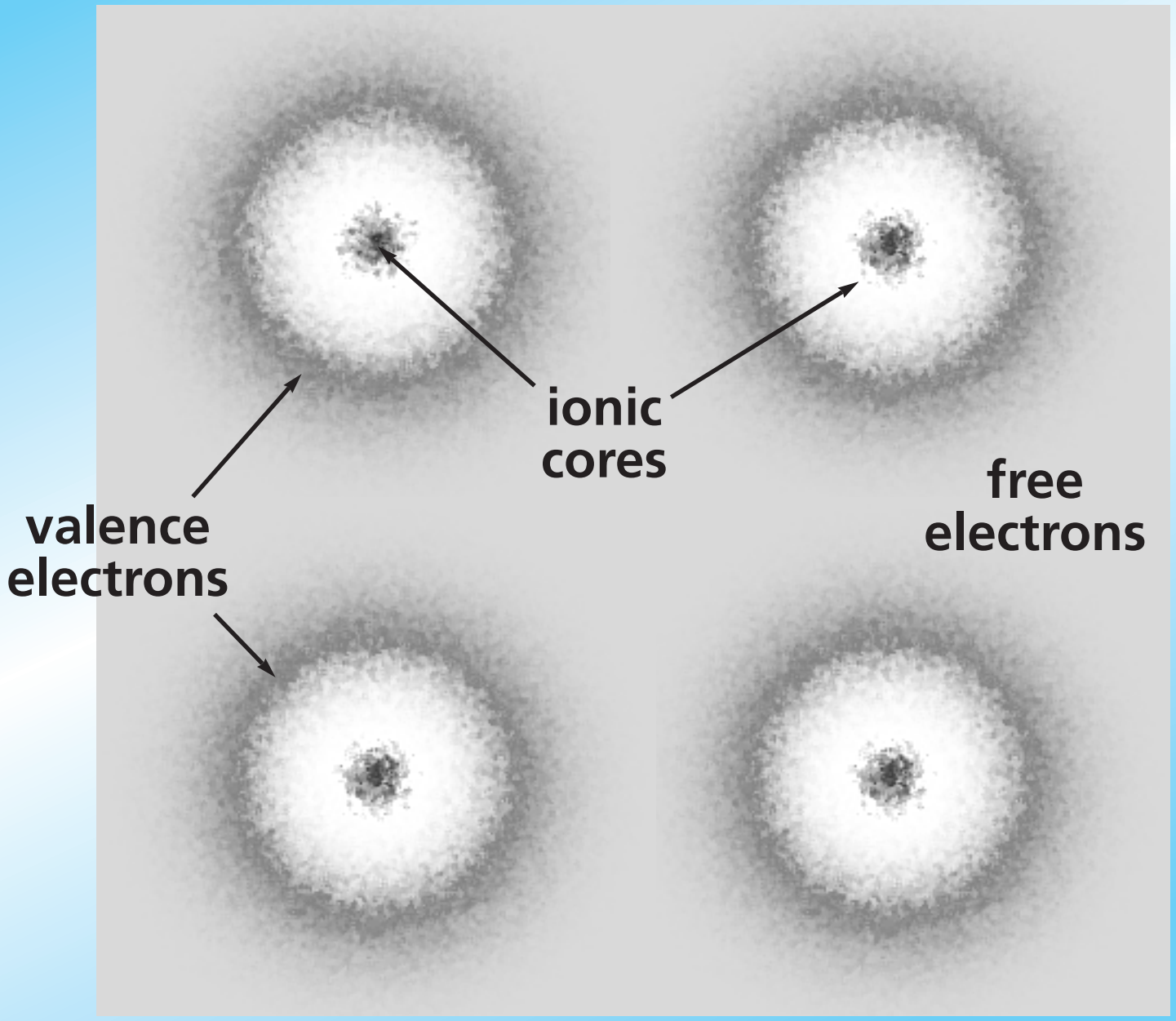


Propagation of EM waves through medium

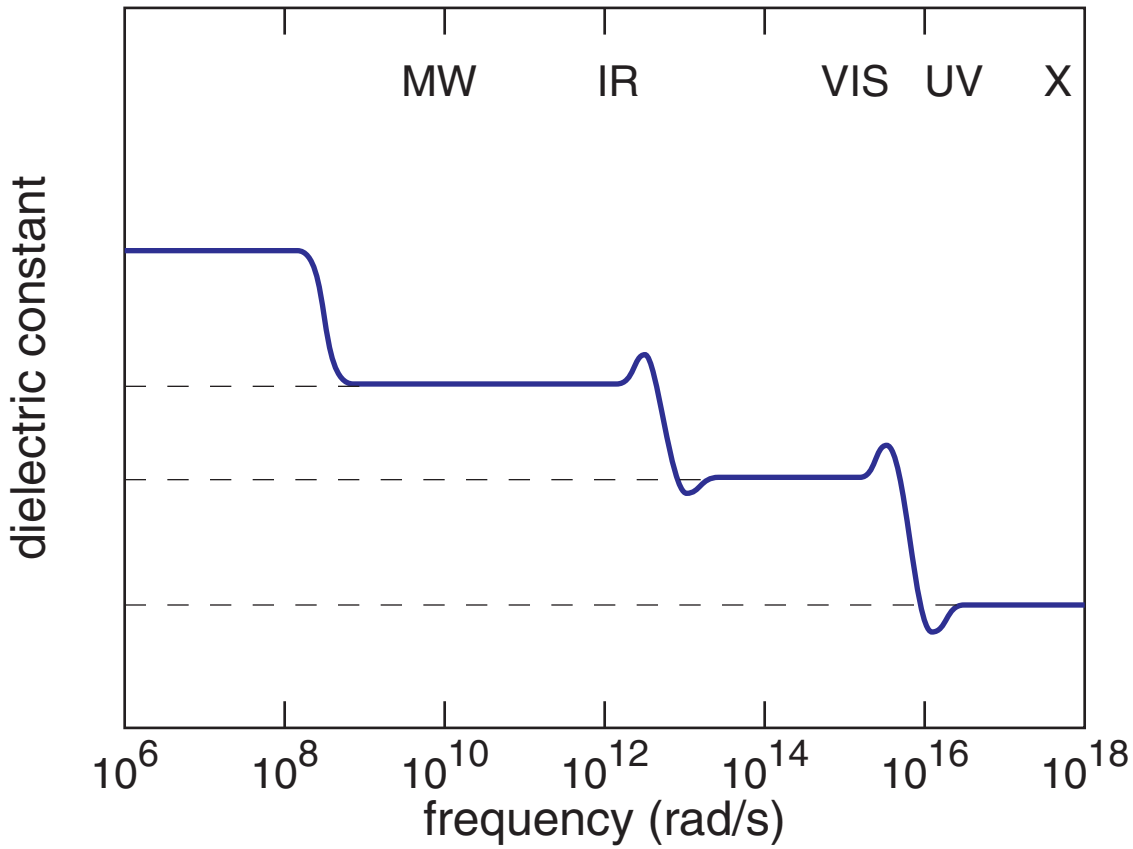
In medium: $$v = \frac{c}{\sqrt{\epsilon(\omega)}} = \frac{c}{n} \quad \Rightarrow \quad \omega = \frac{c}{\sqrt{\epsilon}} k$$



Which charges participate?



Dielectric function



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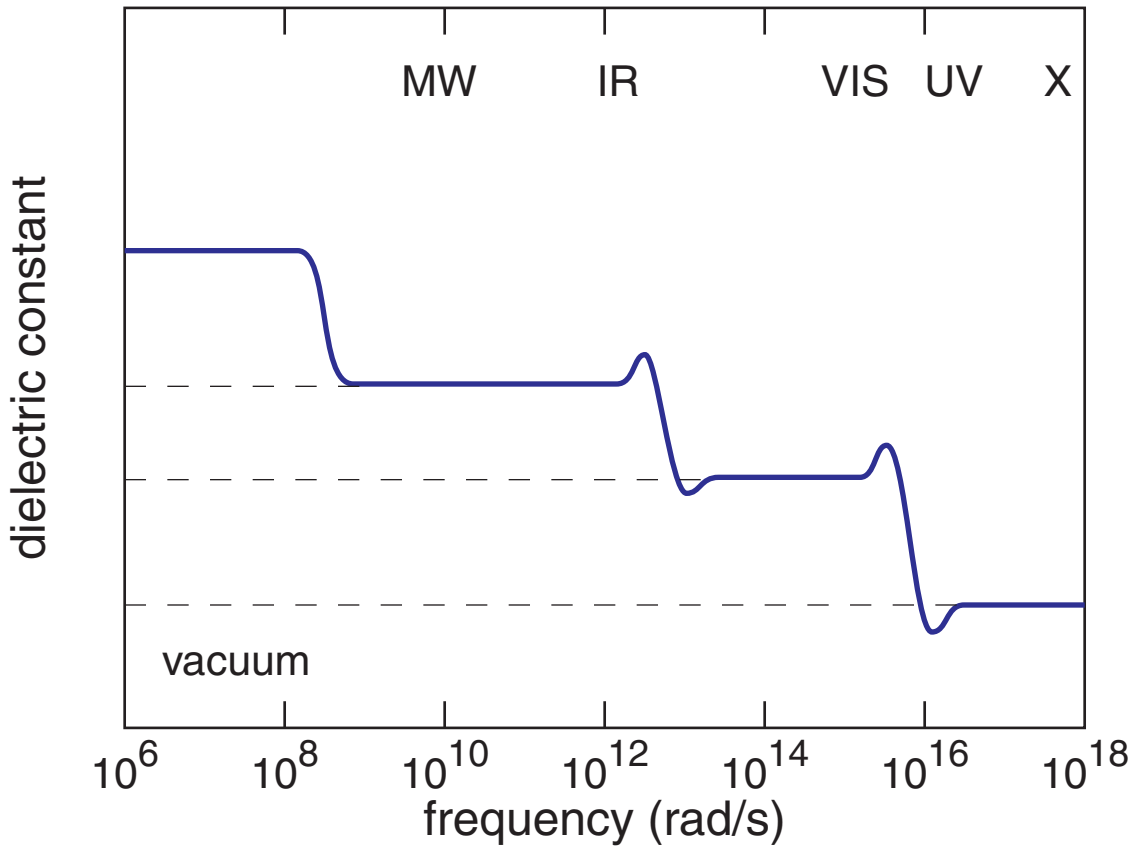

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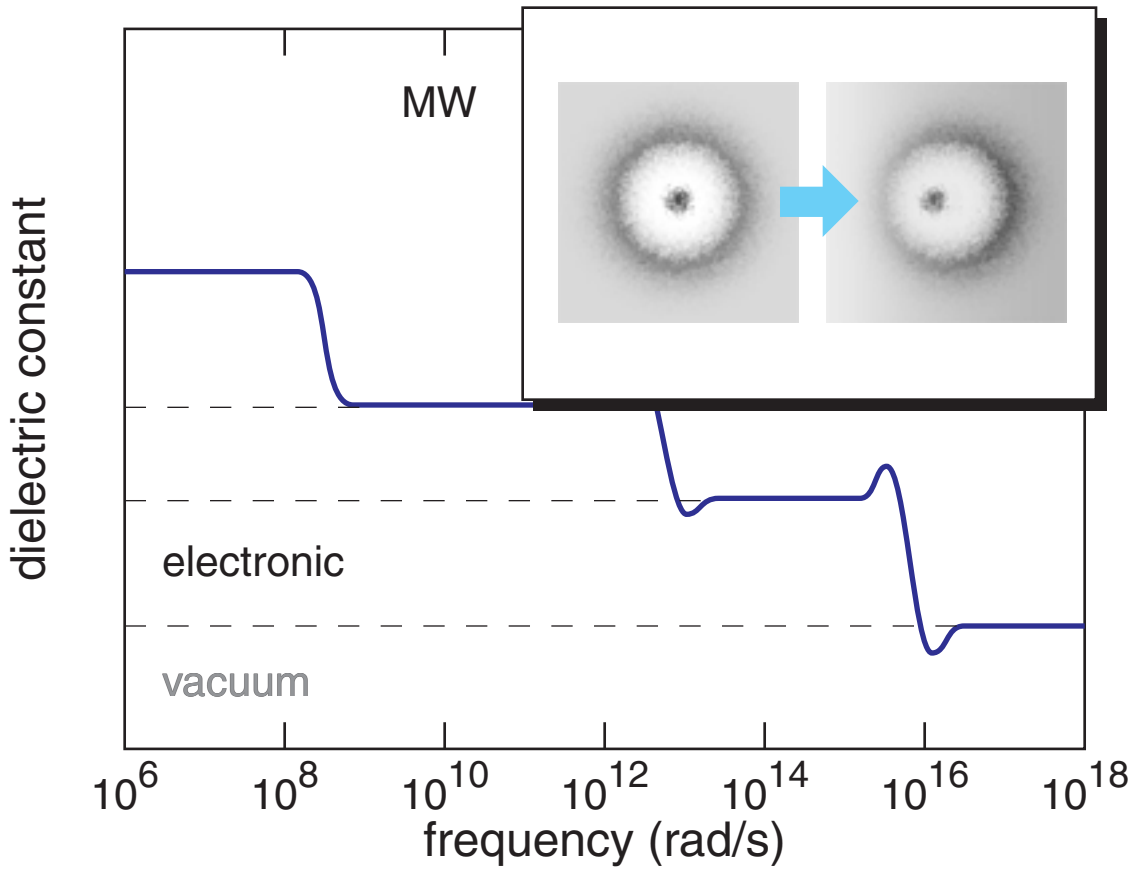

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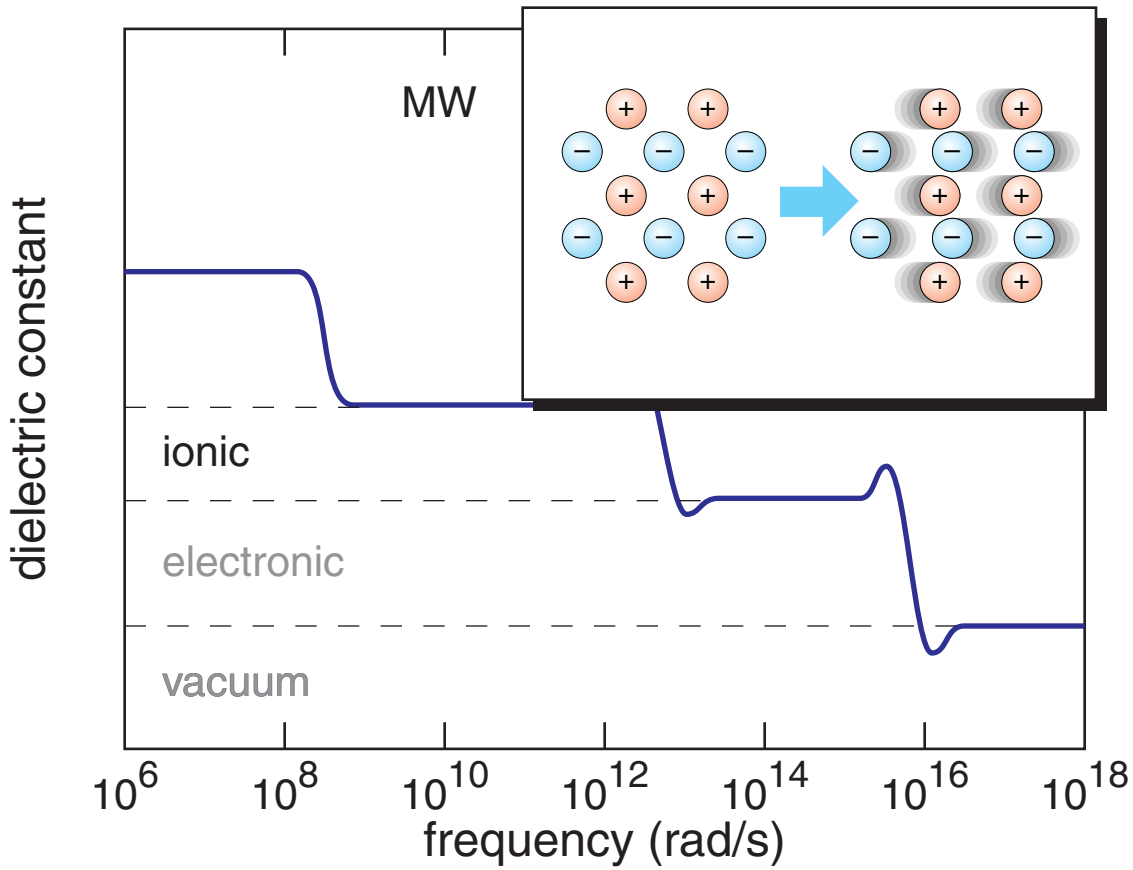

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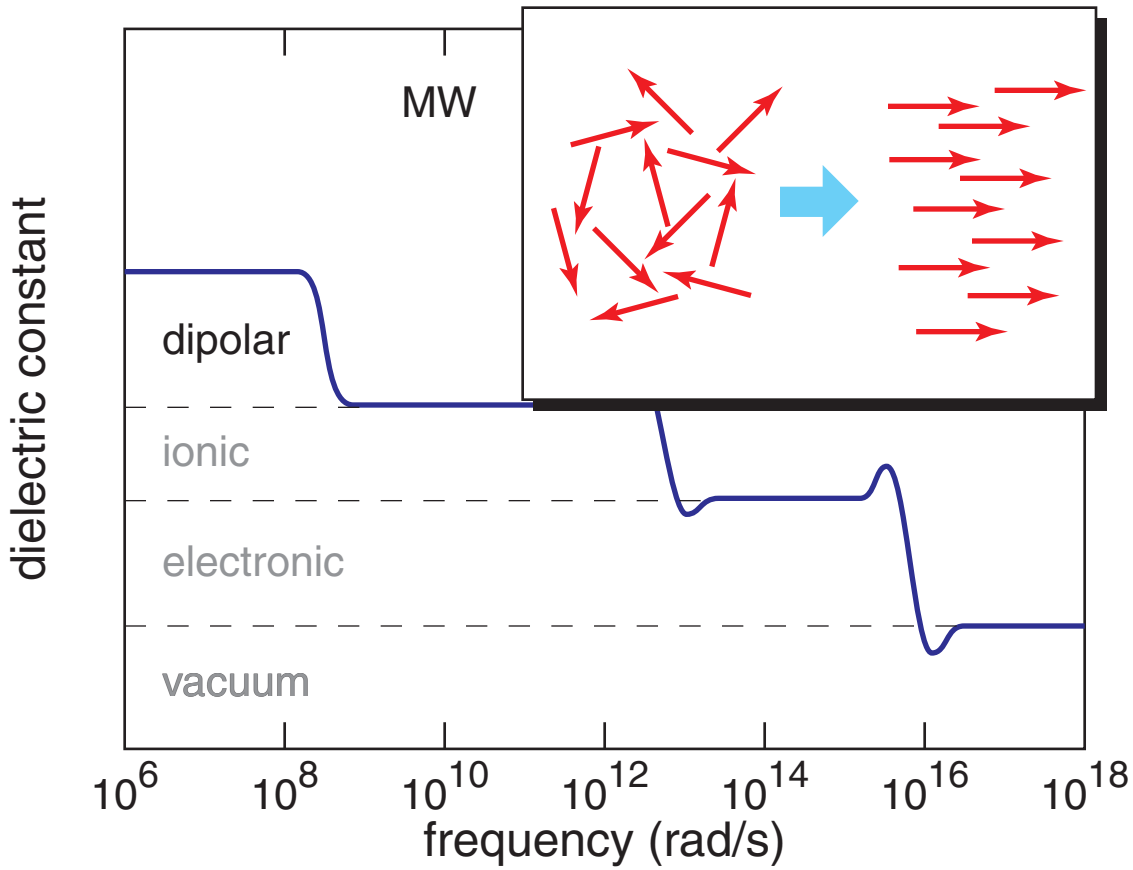

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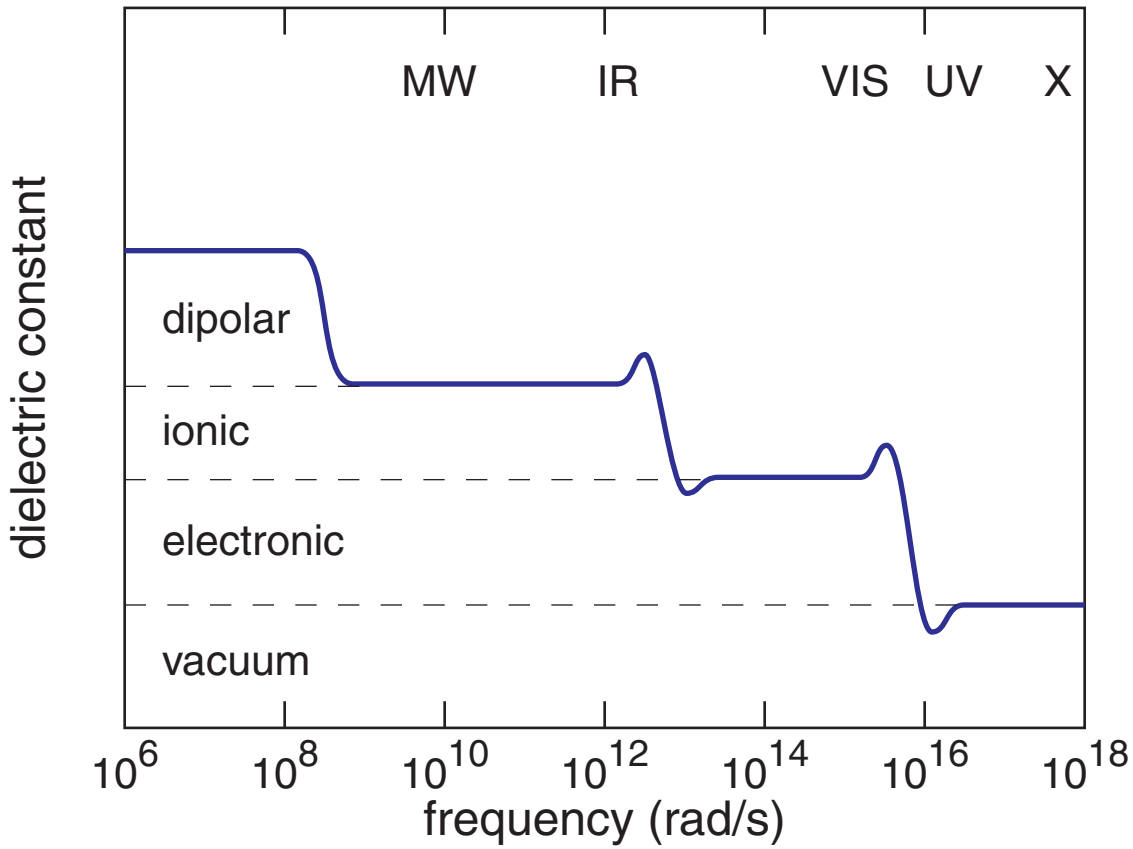

Bound electrons

Electron on a string:

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

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Bound electrons

Steady state: electron oscillates at driving frequency

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

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Polarization

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

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Dielectric function

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

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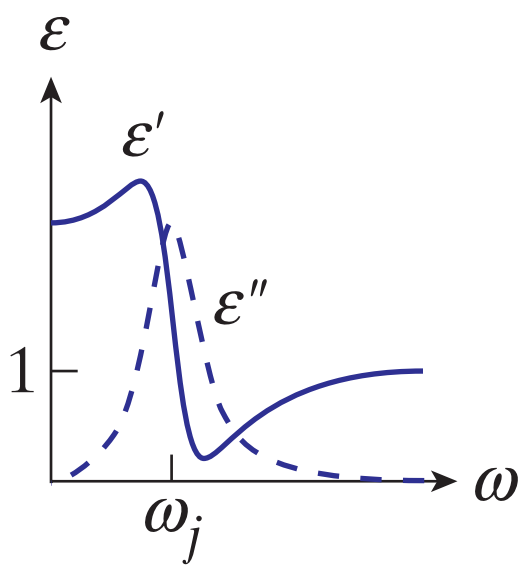
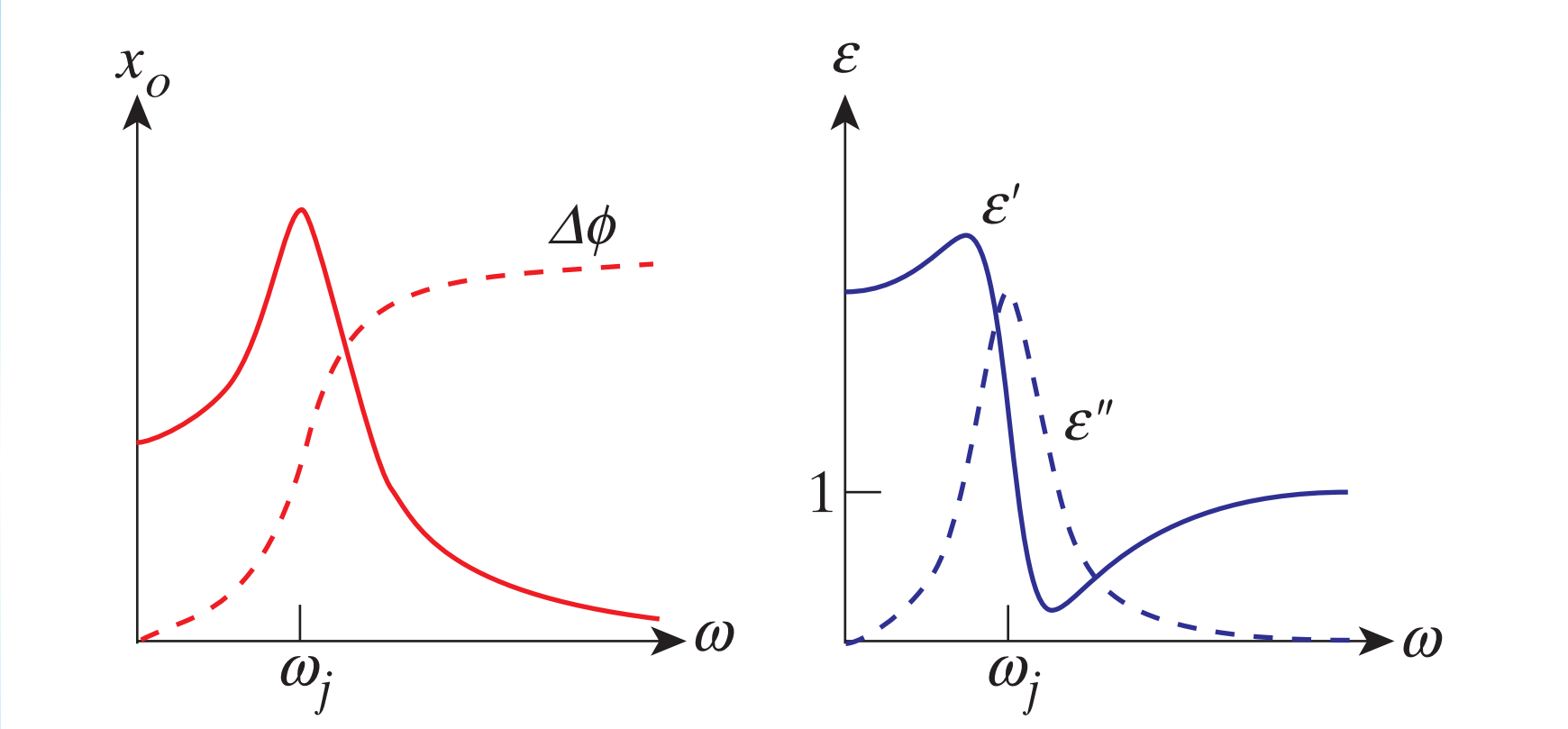


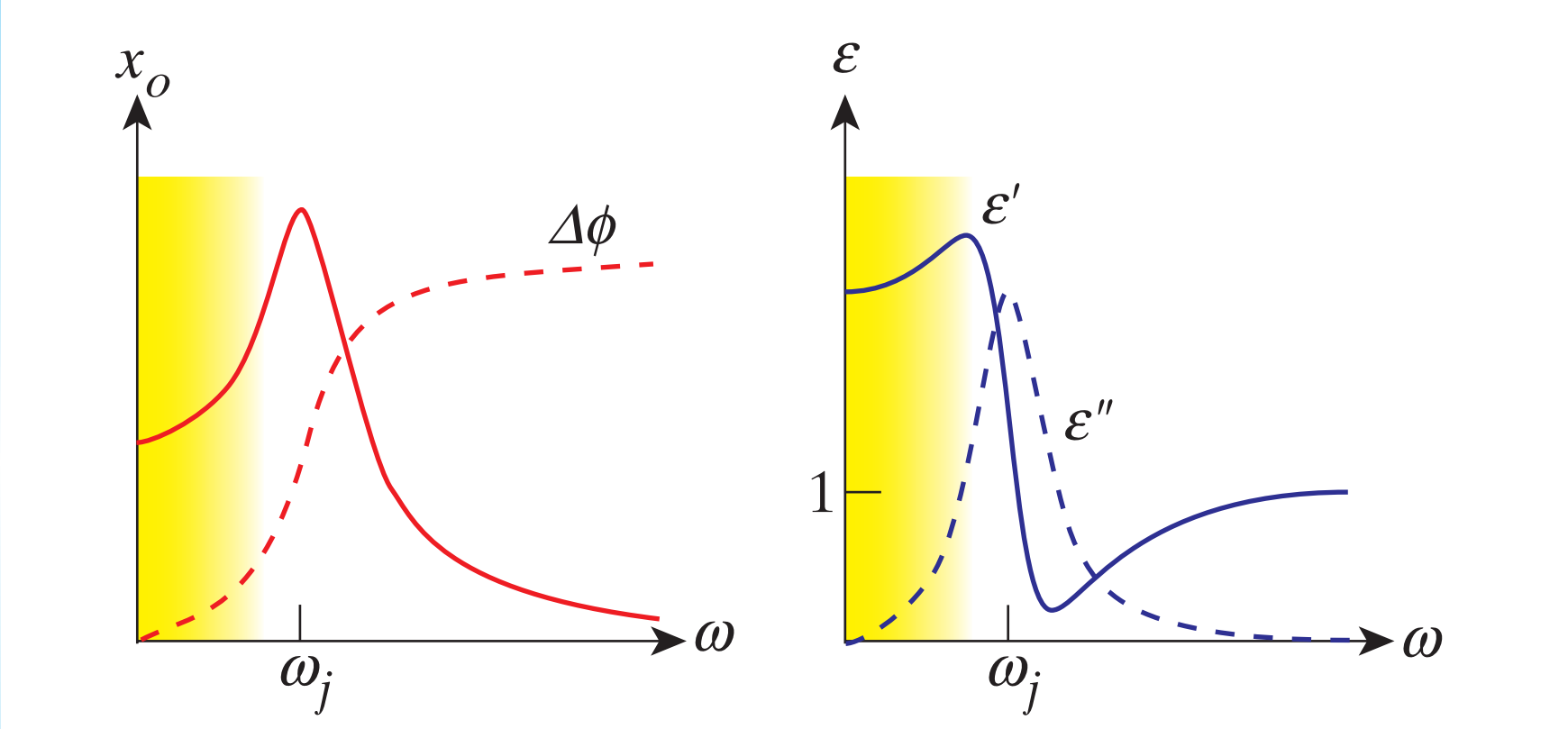
Bound electrons

amplitude of bound charge oscillation



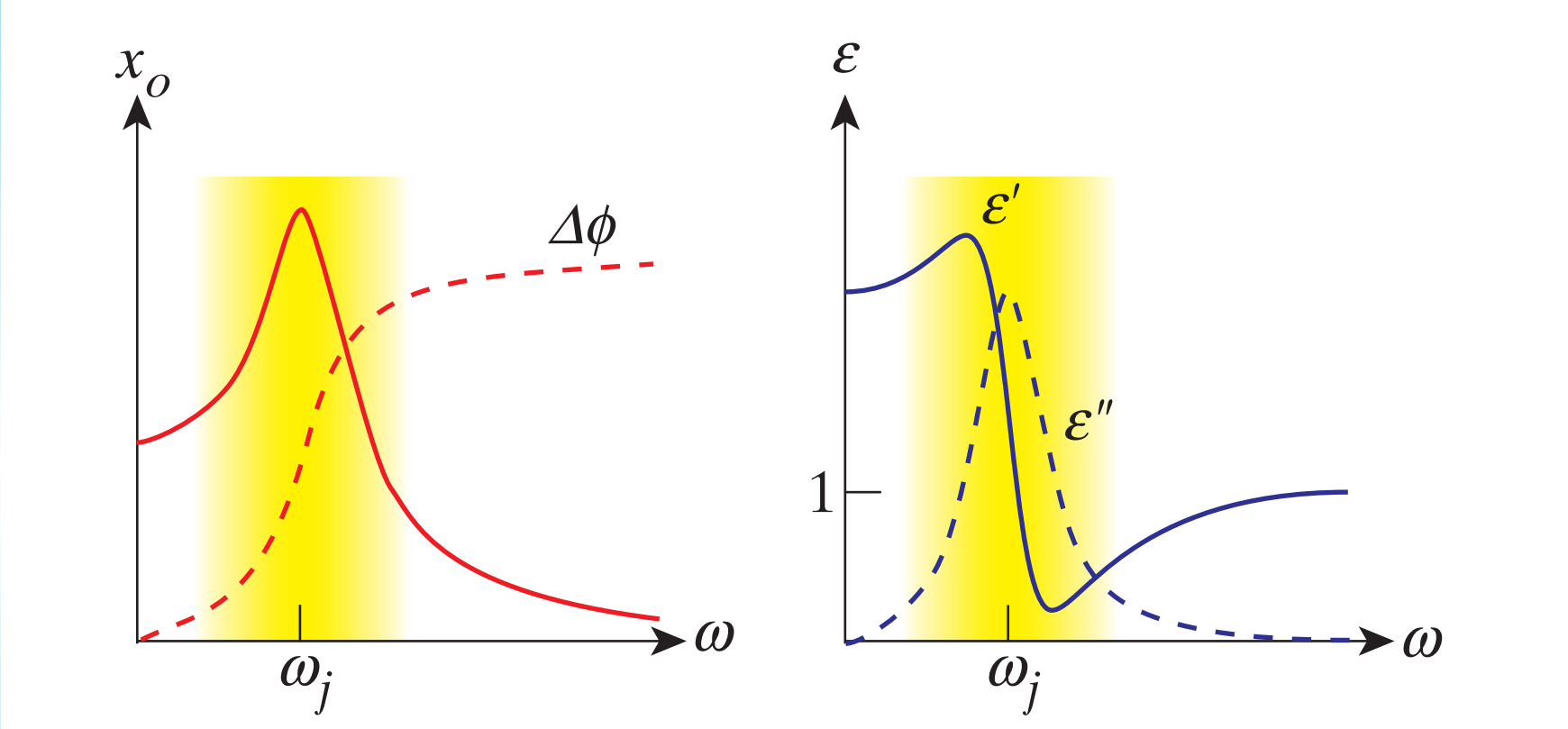
Bound electrons

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



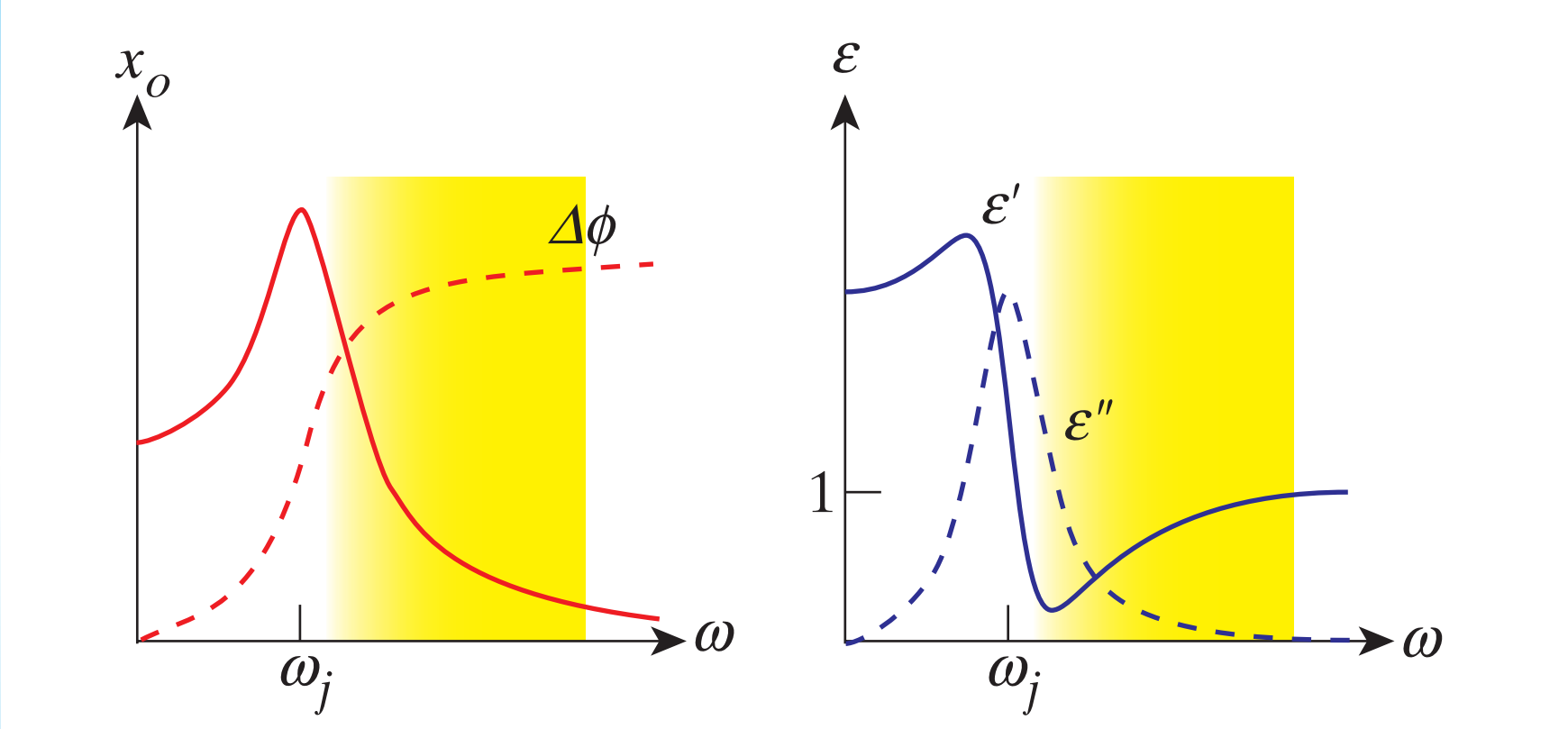
Bound electrons

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



Bound electrons

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



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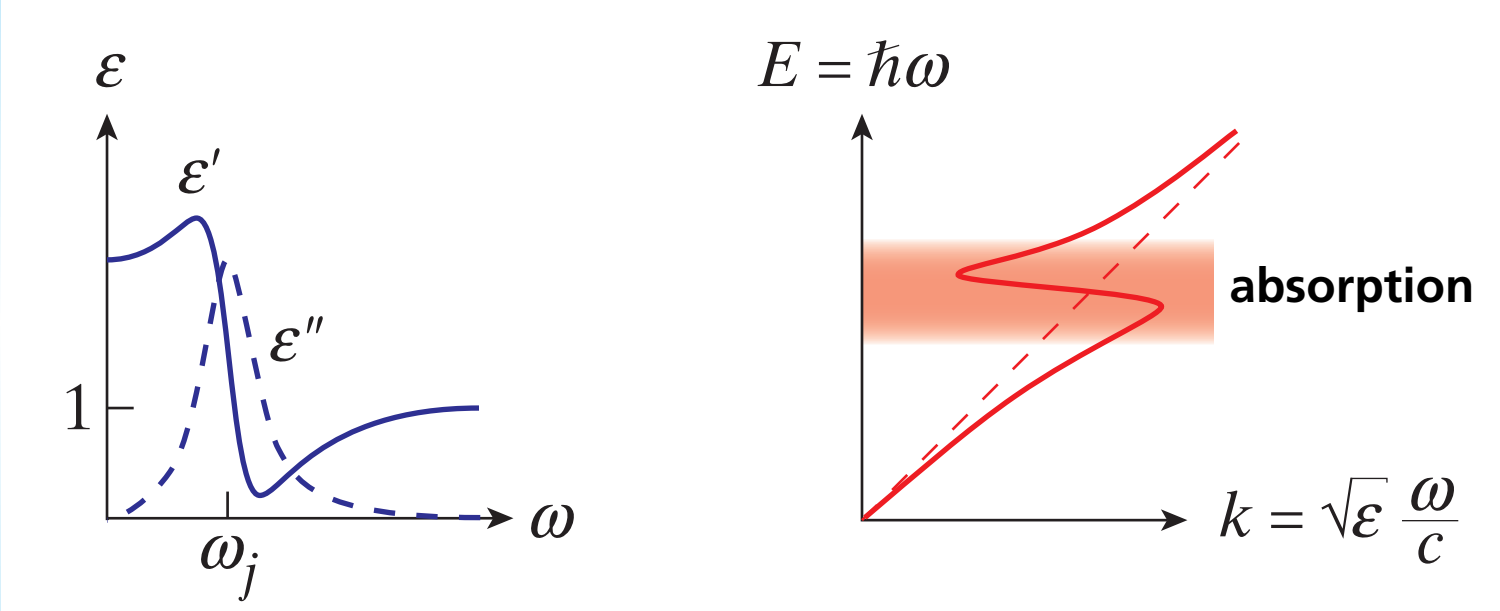
$$\epsilon(\omega) \equiv 1 + \chi_e = 1 + \frac{Ne^2}{\epsilon_o m} \sum_j \frac{f_j}{(\omega_j^2 - \omega^2) - i\gamma_j\omega}$$



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

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

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

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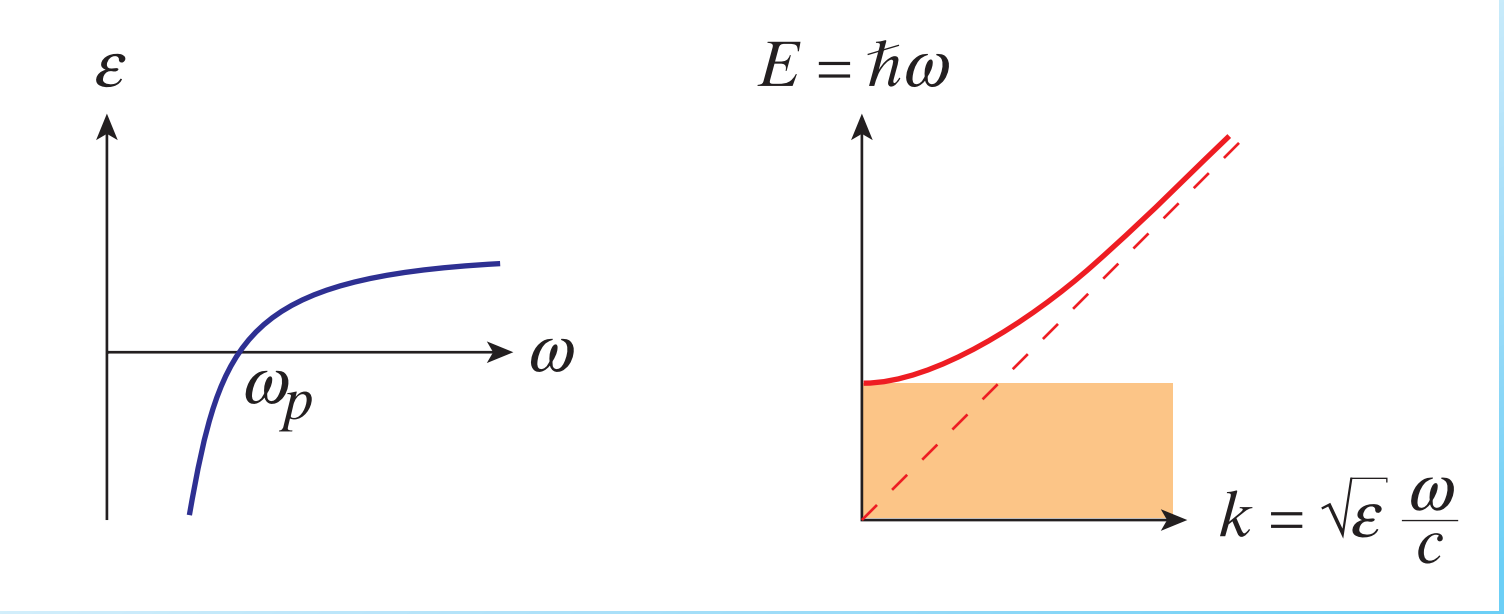
Dielectric function

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

Plasma

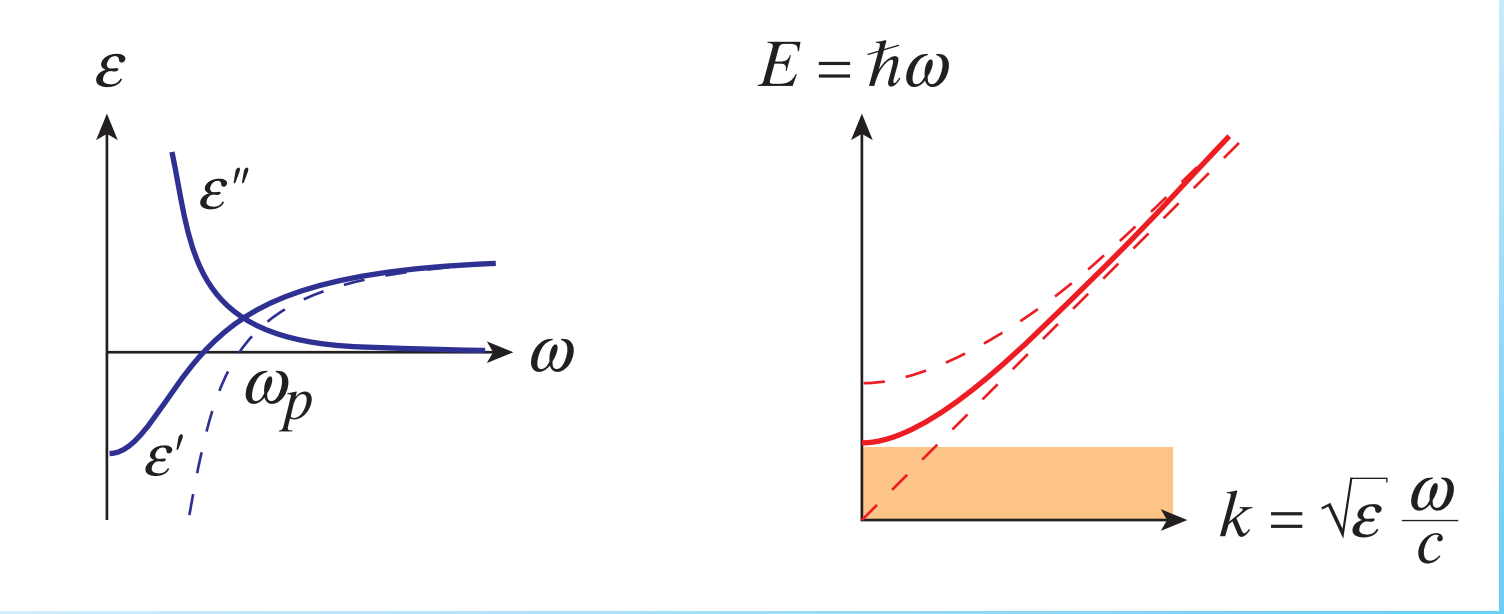
$$\gamma \approx 0 \qquad \Rightarrow \quad \epsilon'' = 0$$

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



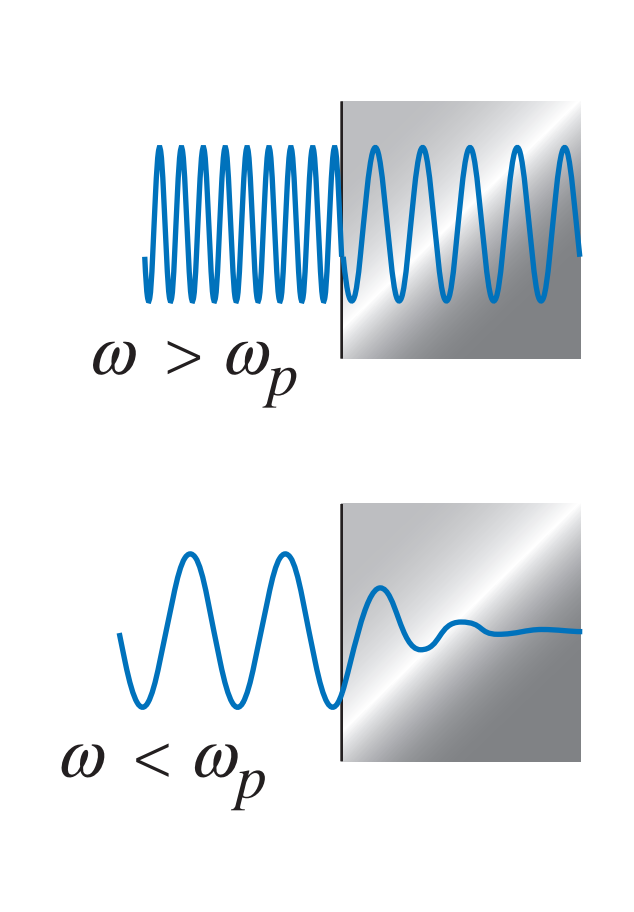
Plasma

Add damping $\gamma \lesssim \omega_p$



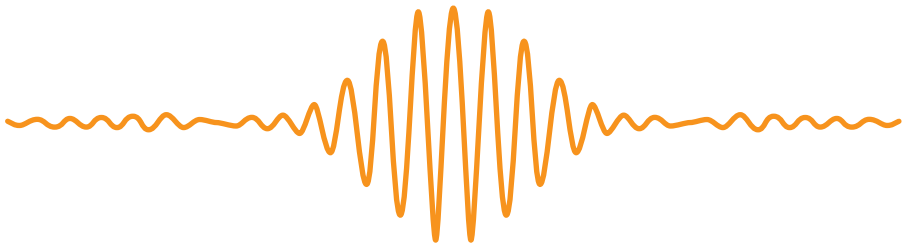
Plasma

Plasma acts like a high-pass filter:



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

Pulse dispersion



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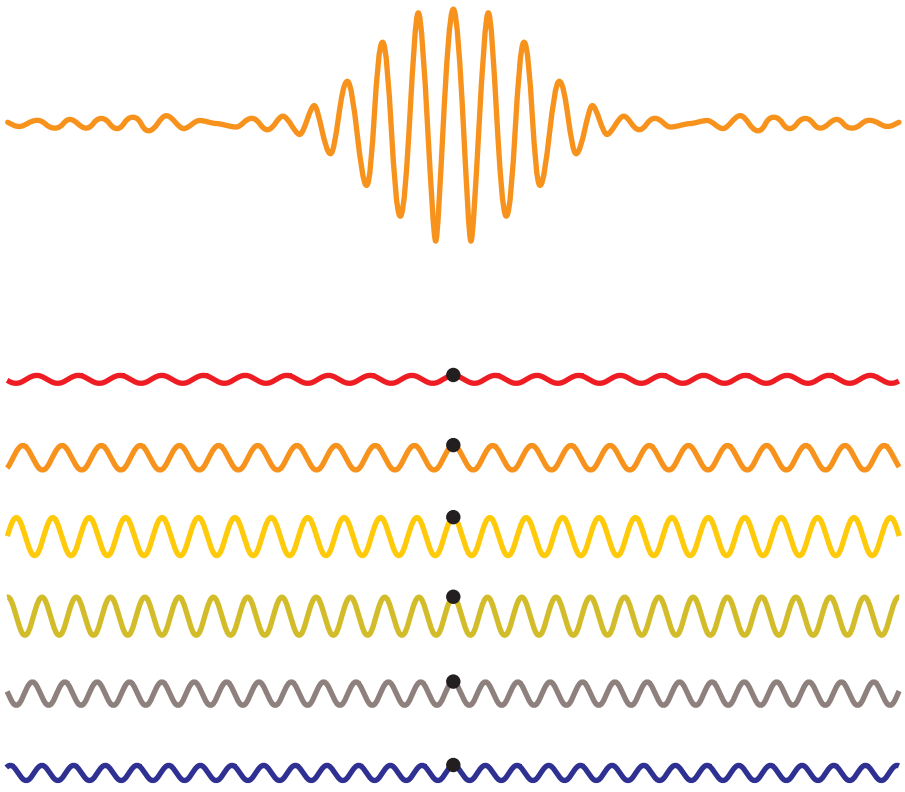

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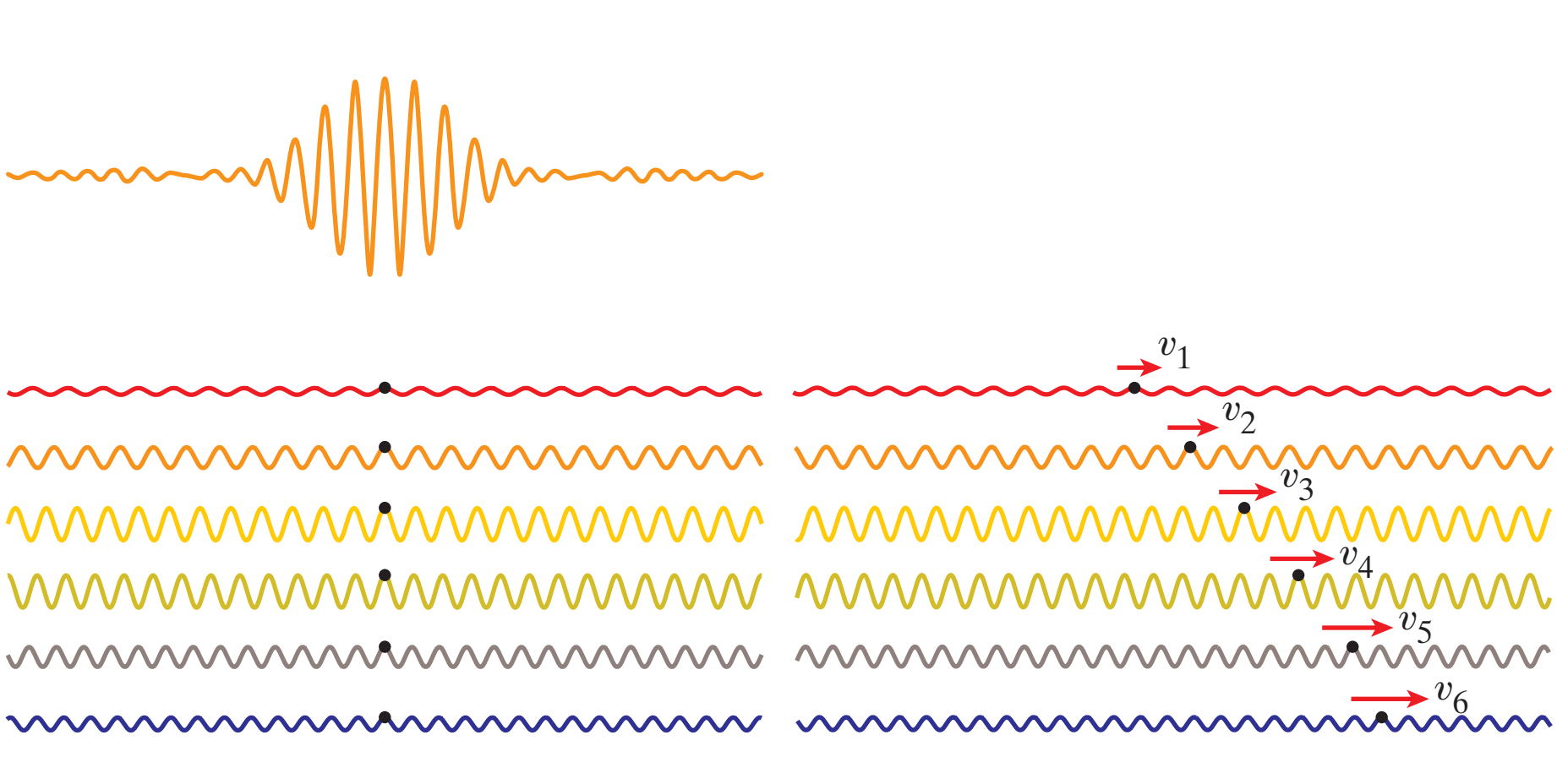

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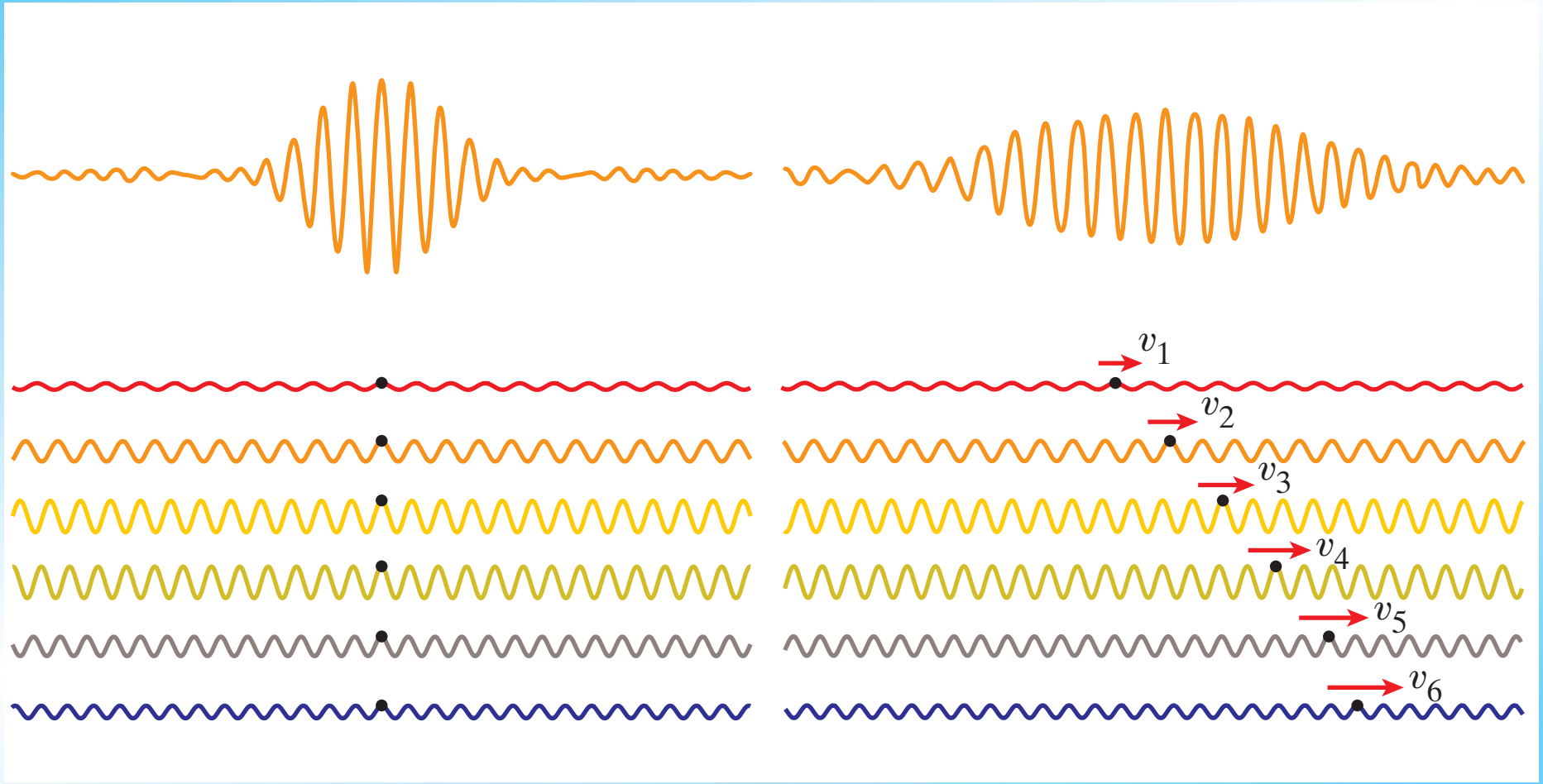

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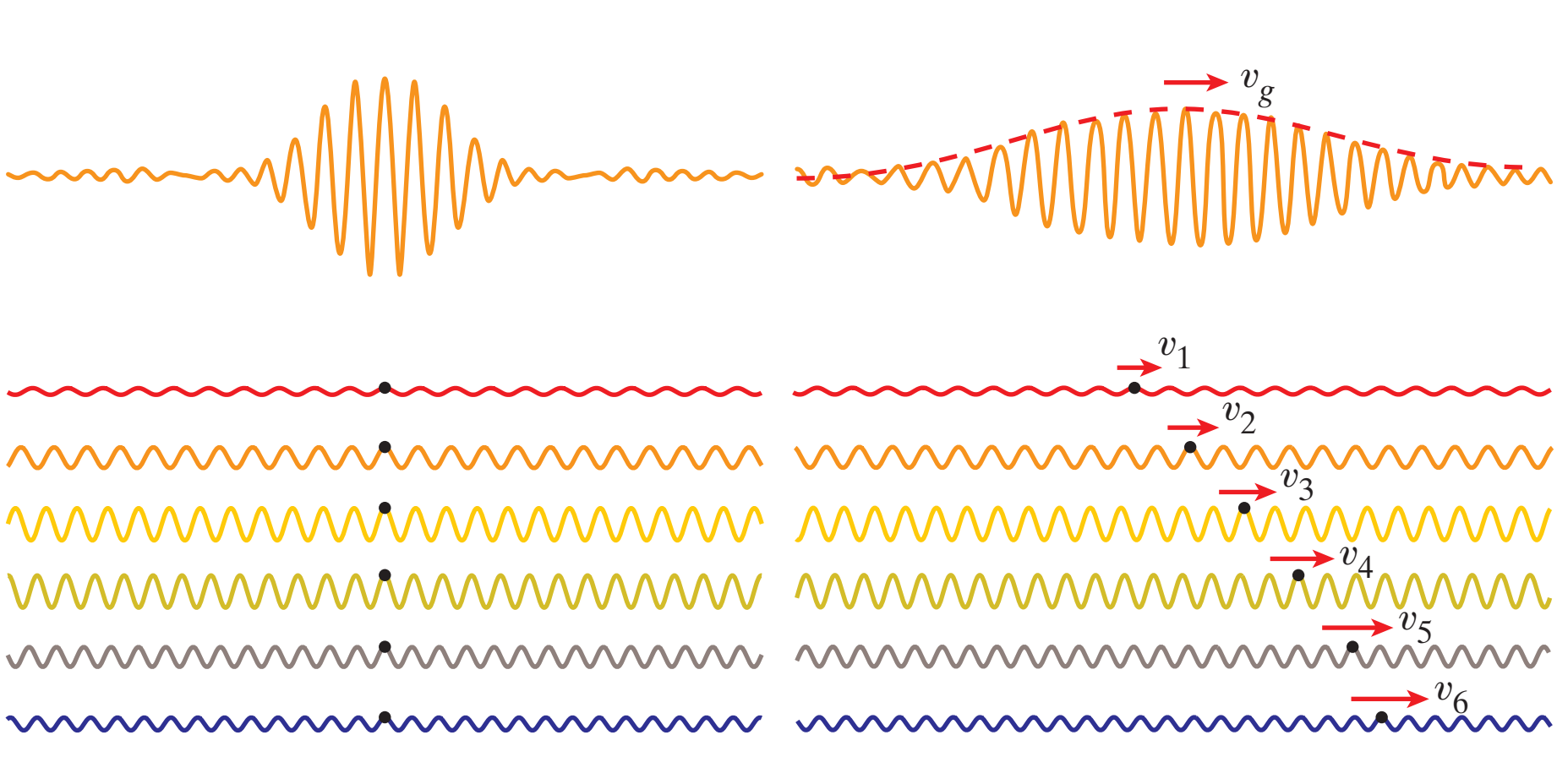
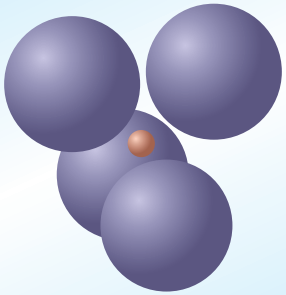


Nonlinear optics

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



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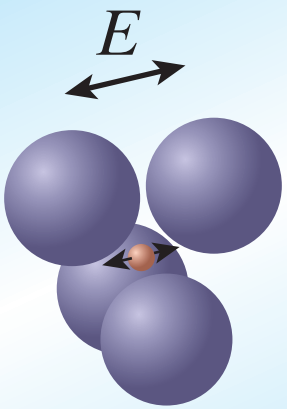

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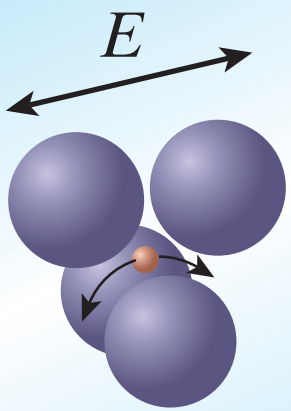

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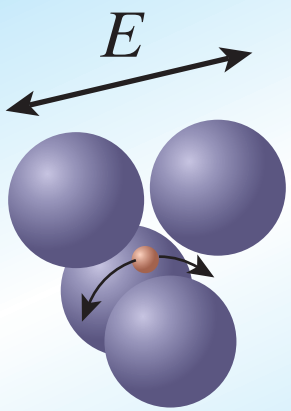
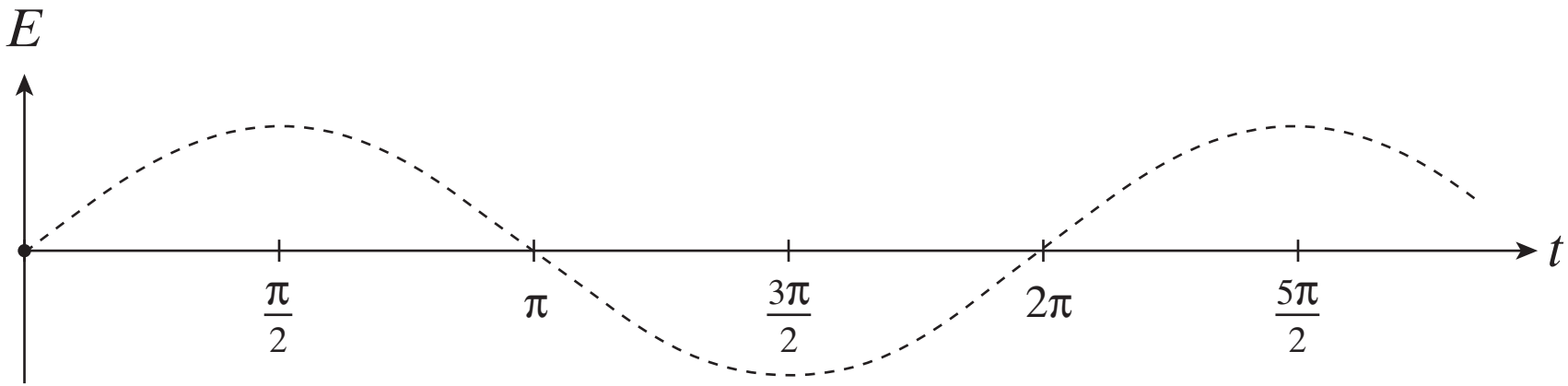


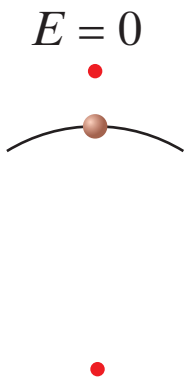
Nonlinear polarization:

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

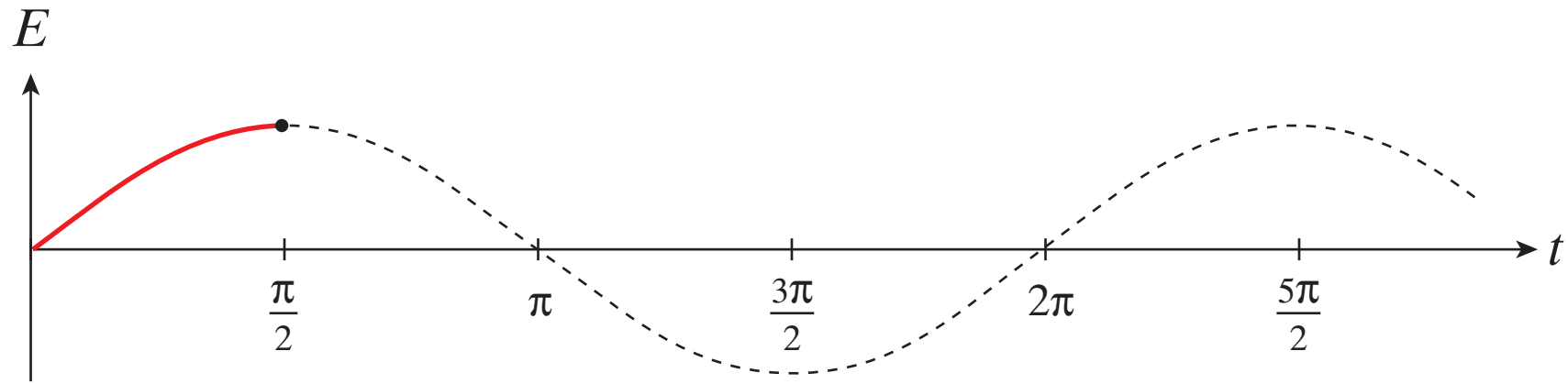
$$P = P^{(1)} + P^{(2)} + P^{(3)} + \ldots$$

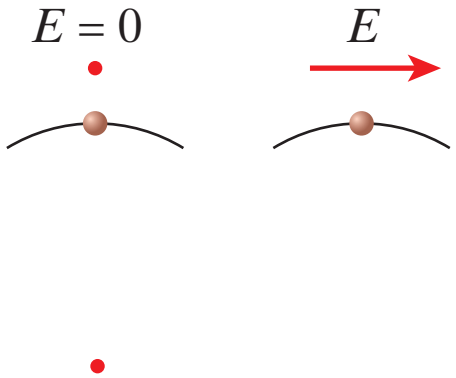
Nonlinear optics



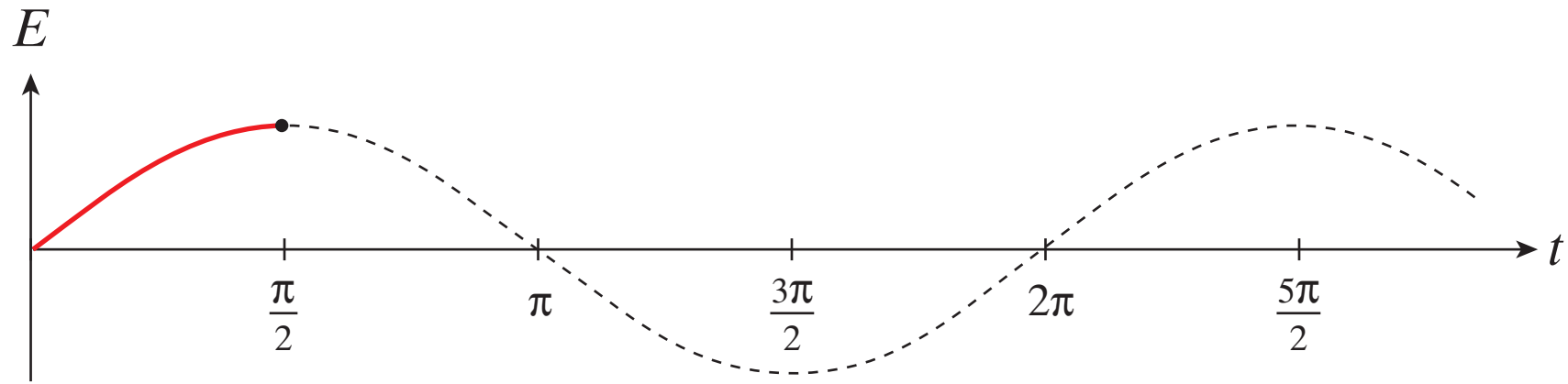


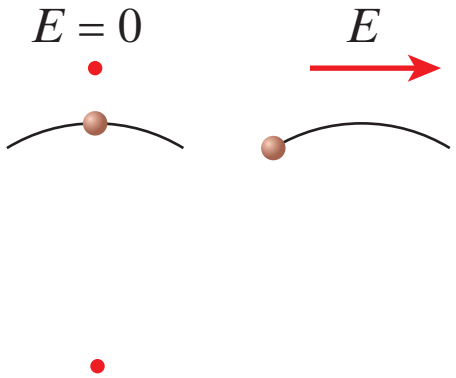
Nonlinear optics



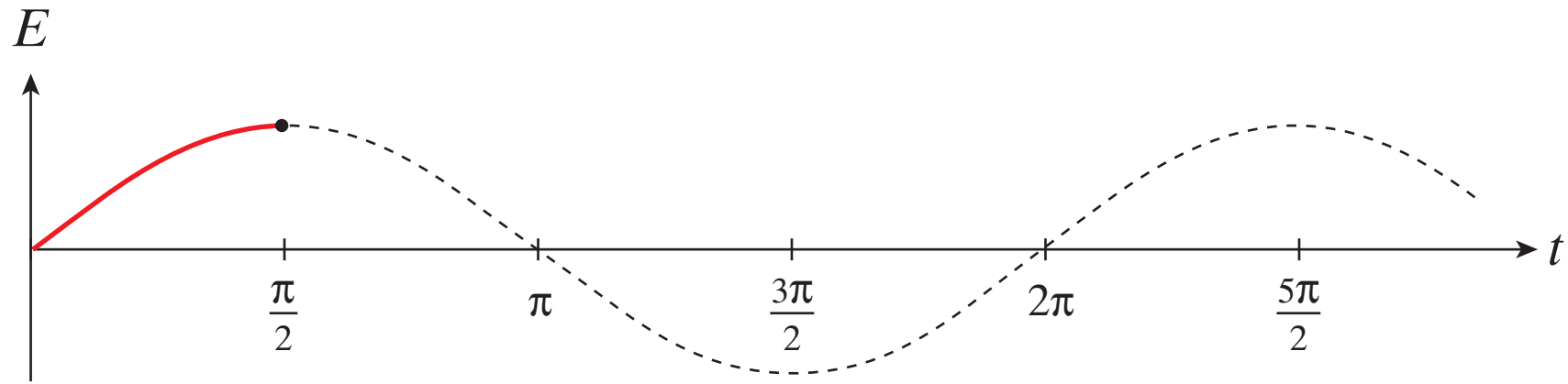


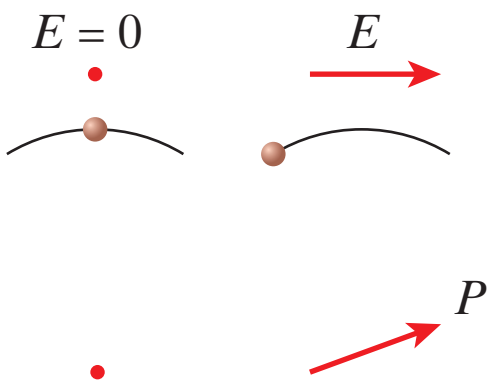
Nonlinear optics



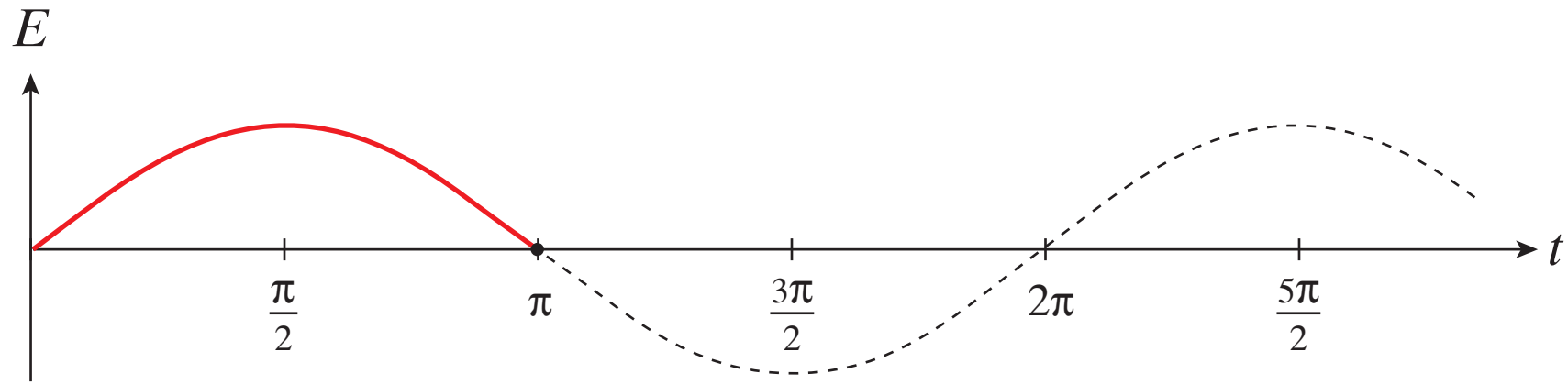


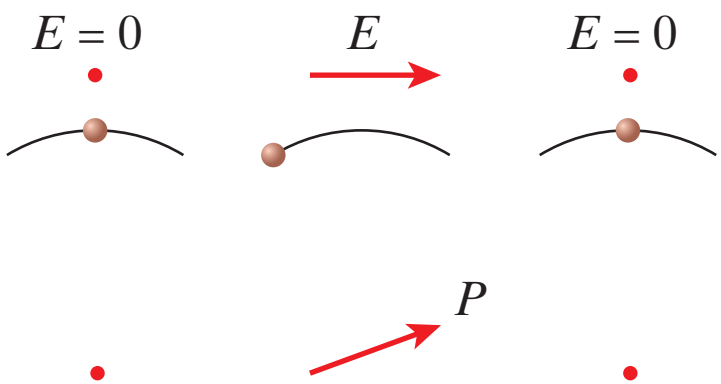
Nonlinear optics



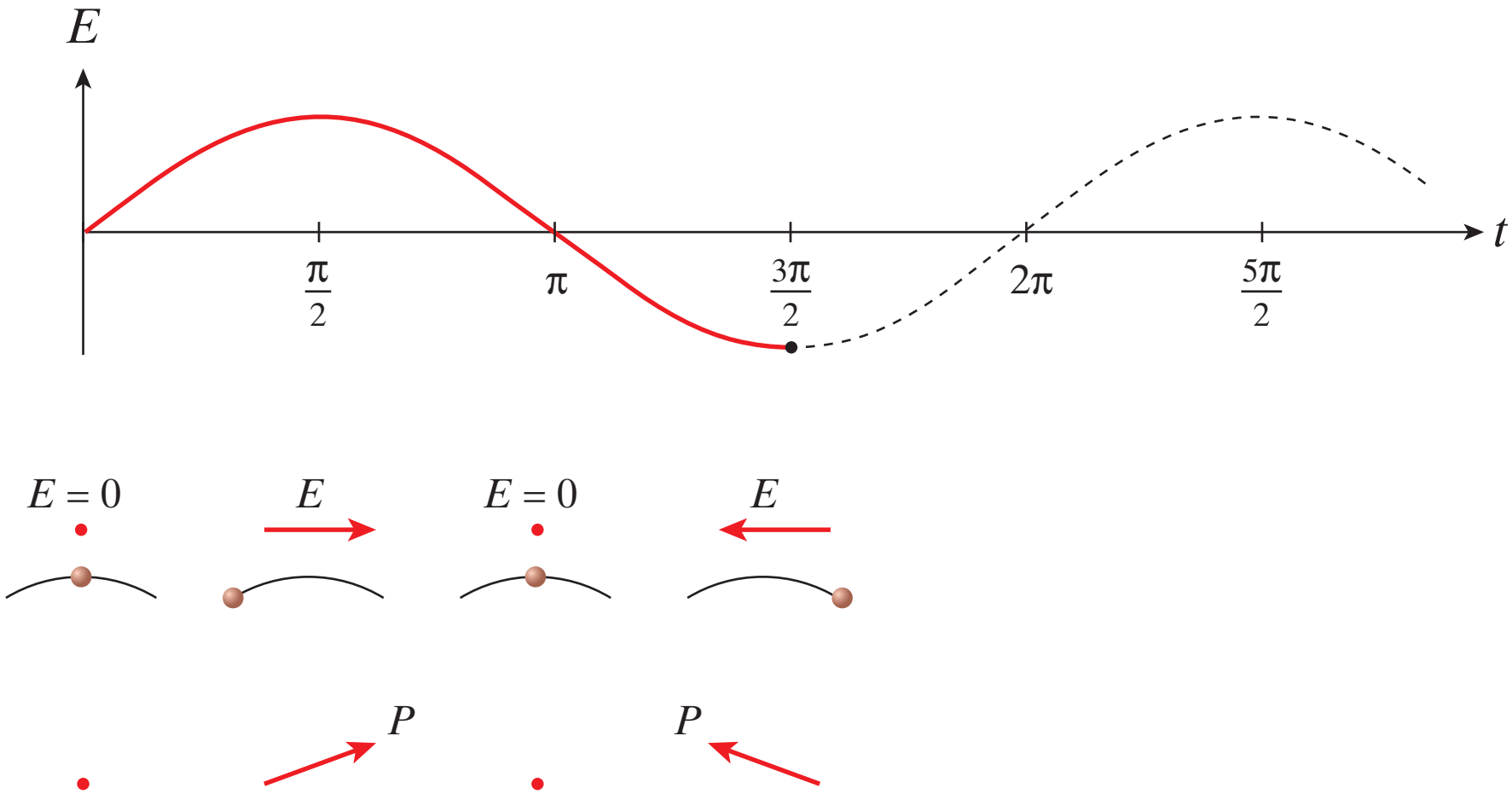


Nonlinear optics

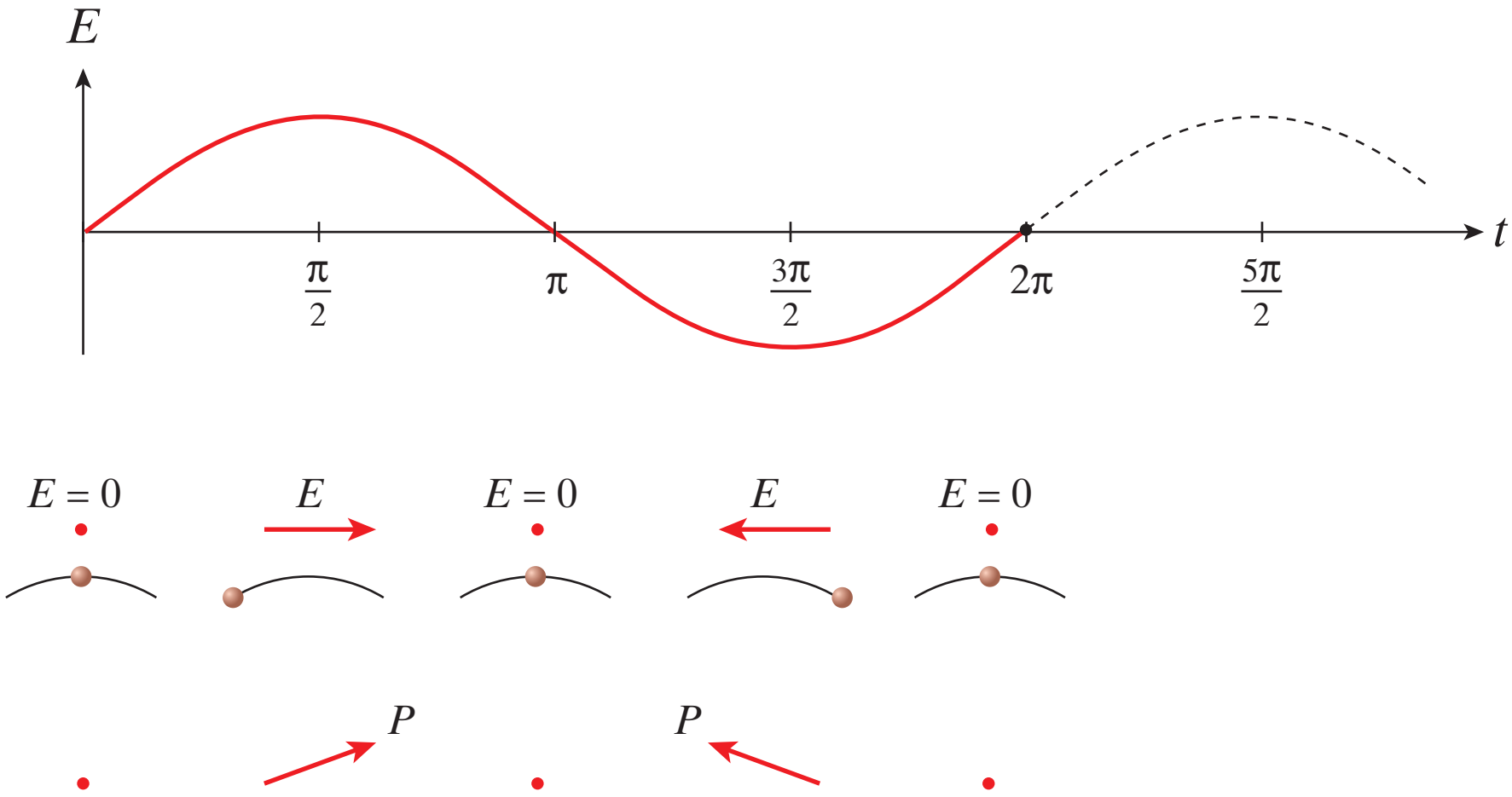




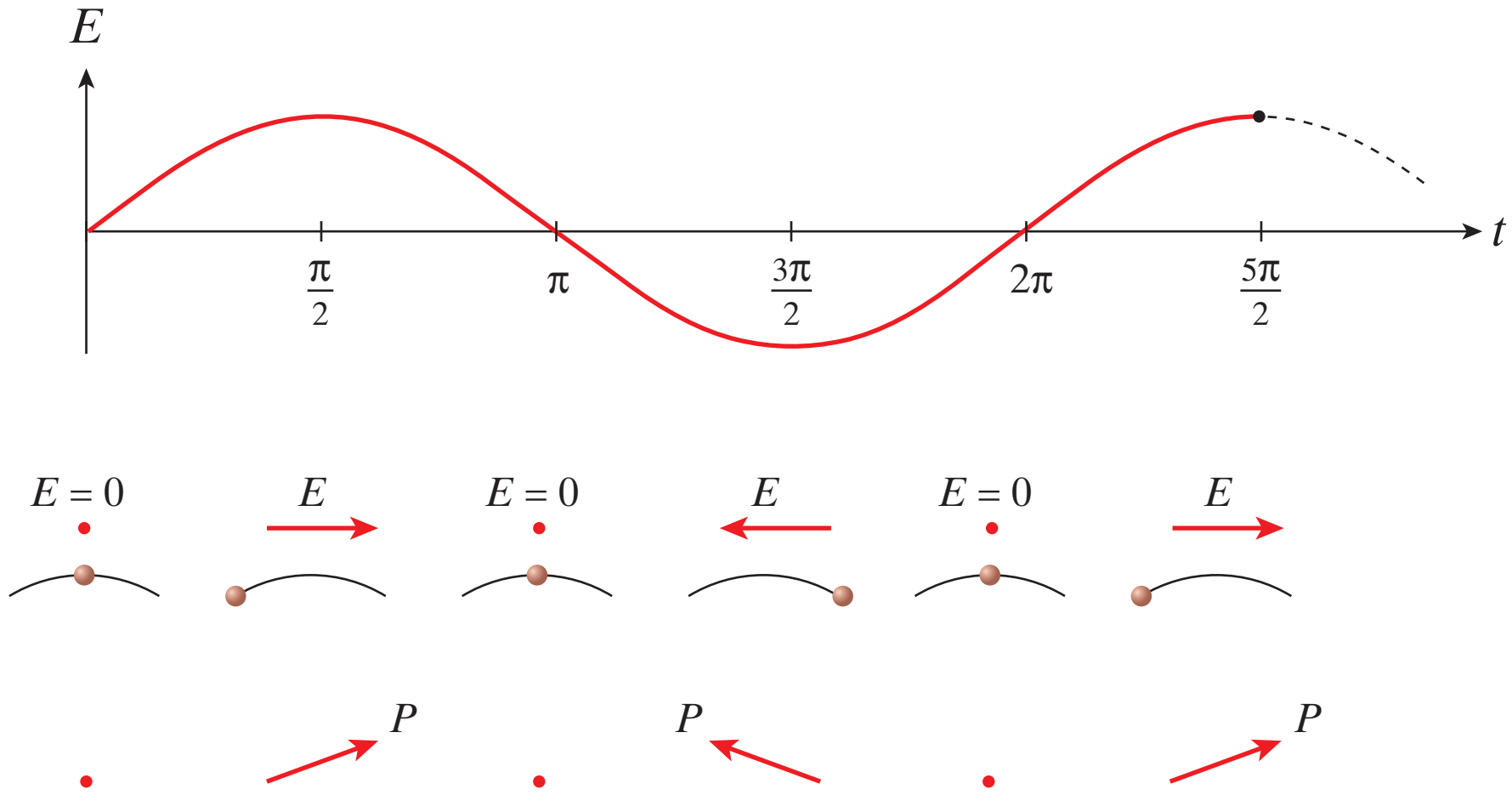
Nonlinear optics



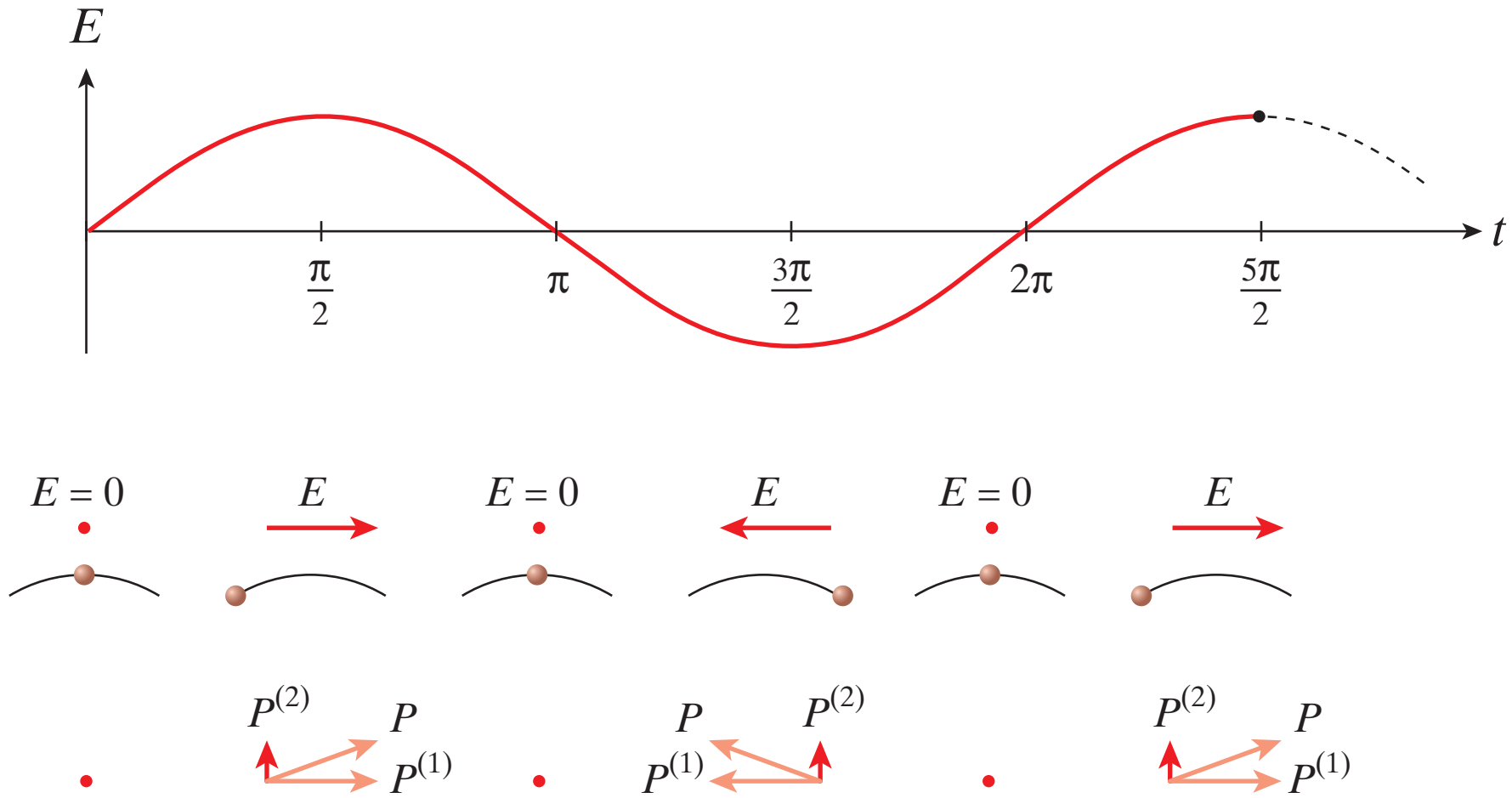
Nonlinear optics



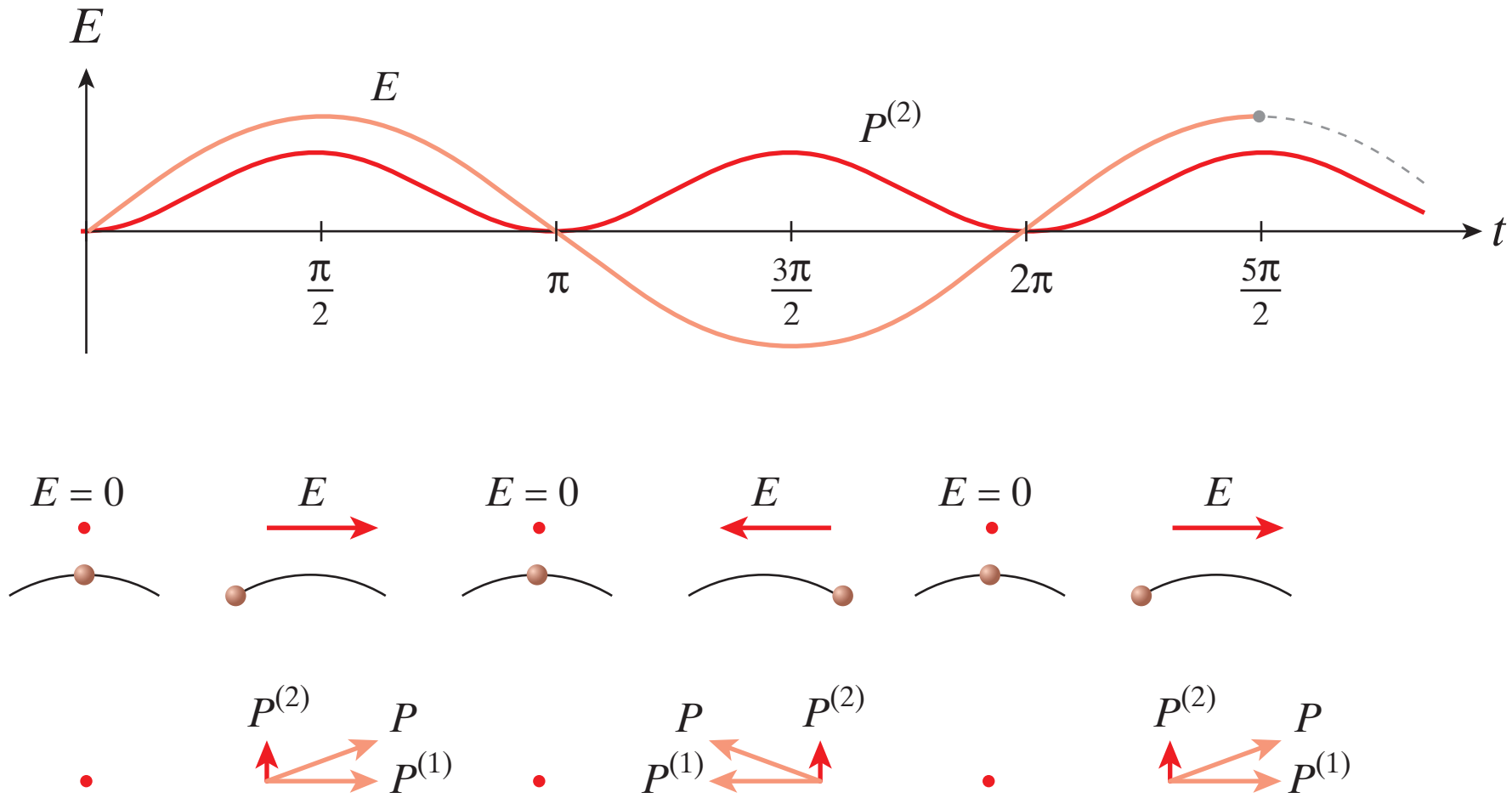
Nonlinear optics



Nonlinear optics



Nonlinear optics



In medium with inversion symmetry

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

Nonlinear optics

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

and so

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

Nonlinear optics

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and so

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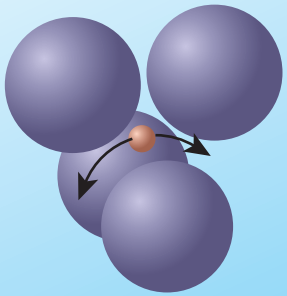
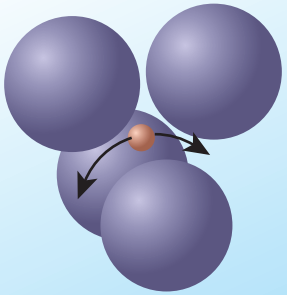
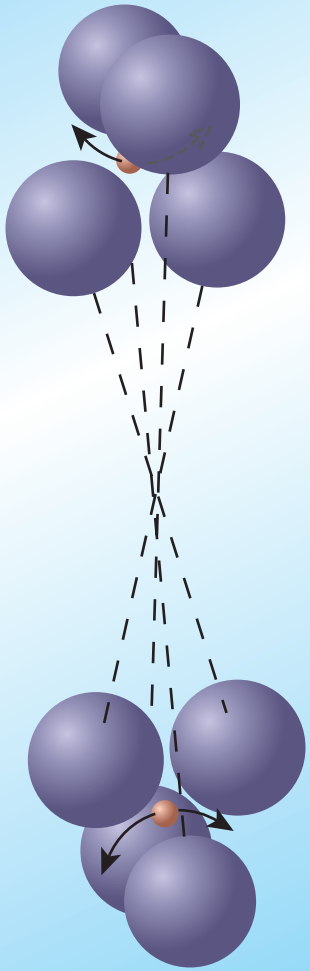
… but …



Nonlinear optics

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

Nonlinear optics



Nonlinear optics

Nonlinear polarization:

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

Nonlinear optics

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Third order polarization

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

Nonlinear optics

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Nonlinear optics

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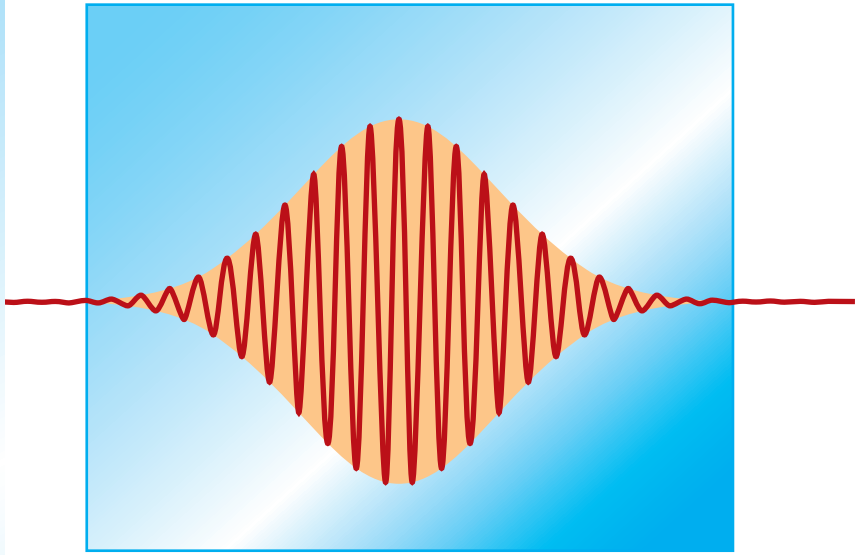
$$n = \sqrt{\epsilon} = \sqrt{1 + \chi_{eff}} \approx \sqrt{1 + \chi^{(1)}} + \frac{1}{2}\frac{\chi^{(3)}I}{\sqrt{1 + \chi^{(1)}}} = n_o + n_2 I$$

Nonlinear optics

Intensity dependent index of refraction:

$$n = n_o + n_2 I$$



Nonlinear optics

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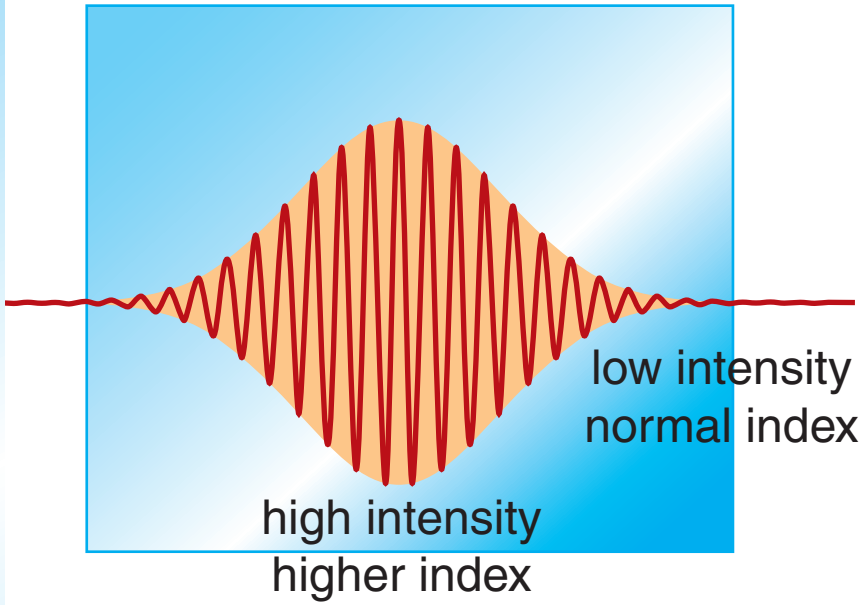

Nonlinear optics

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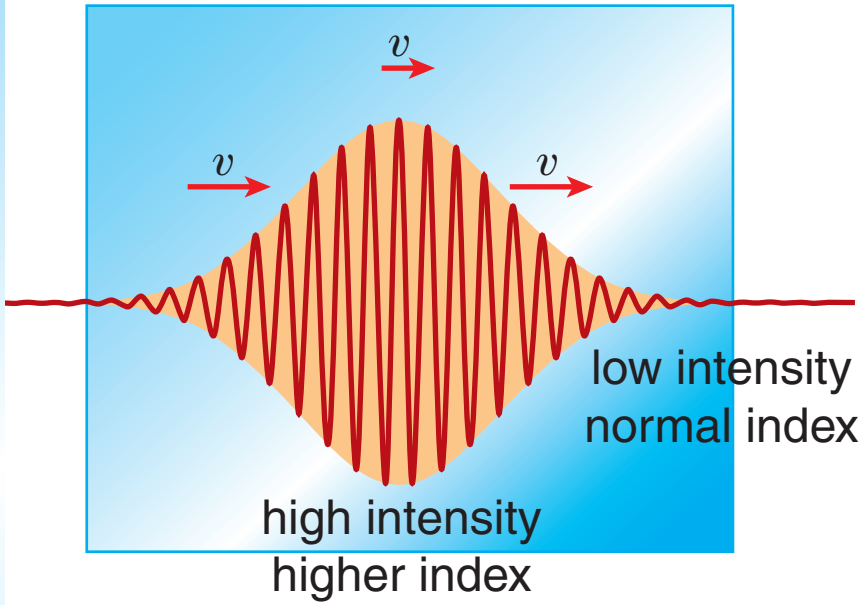

Nonlinear optics

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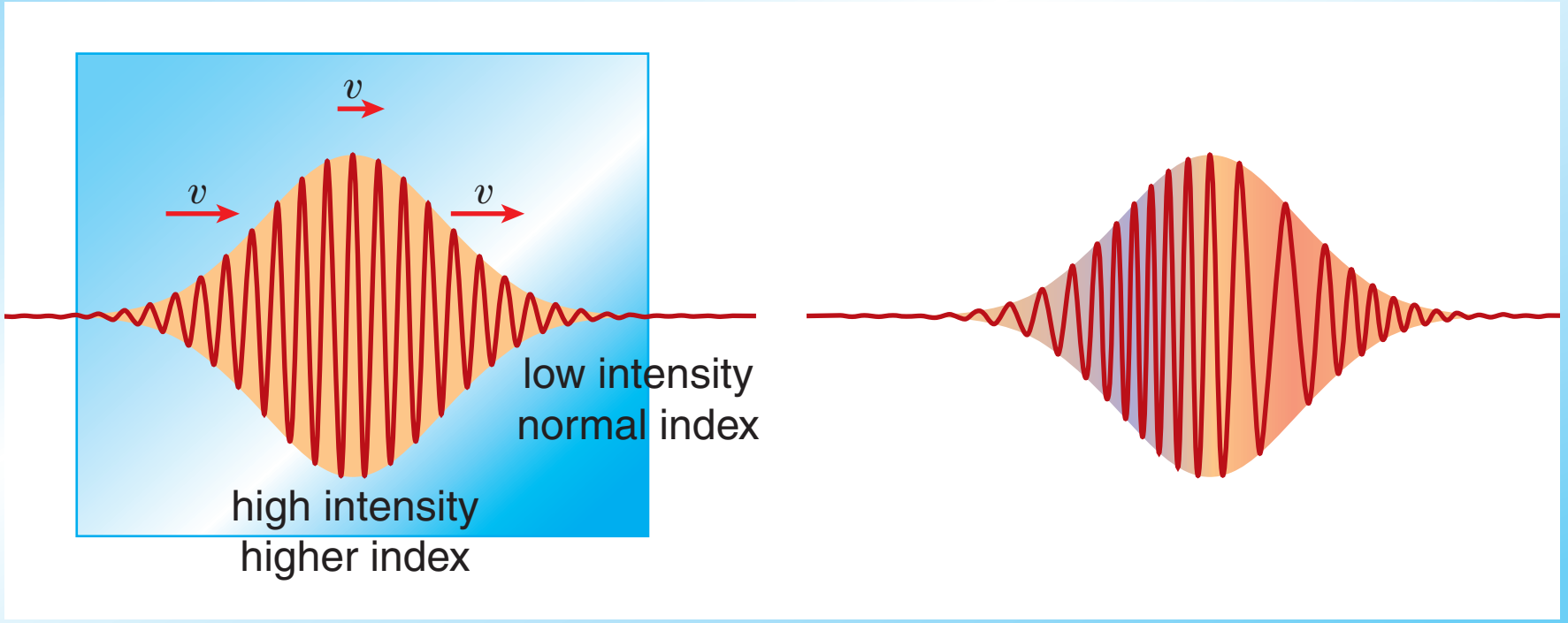

Nonlinear optics

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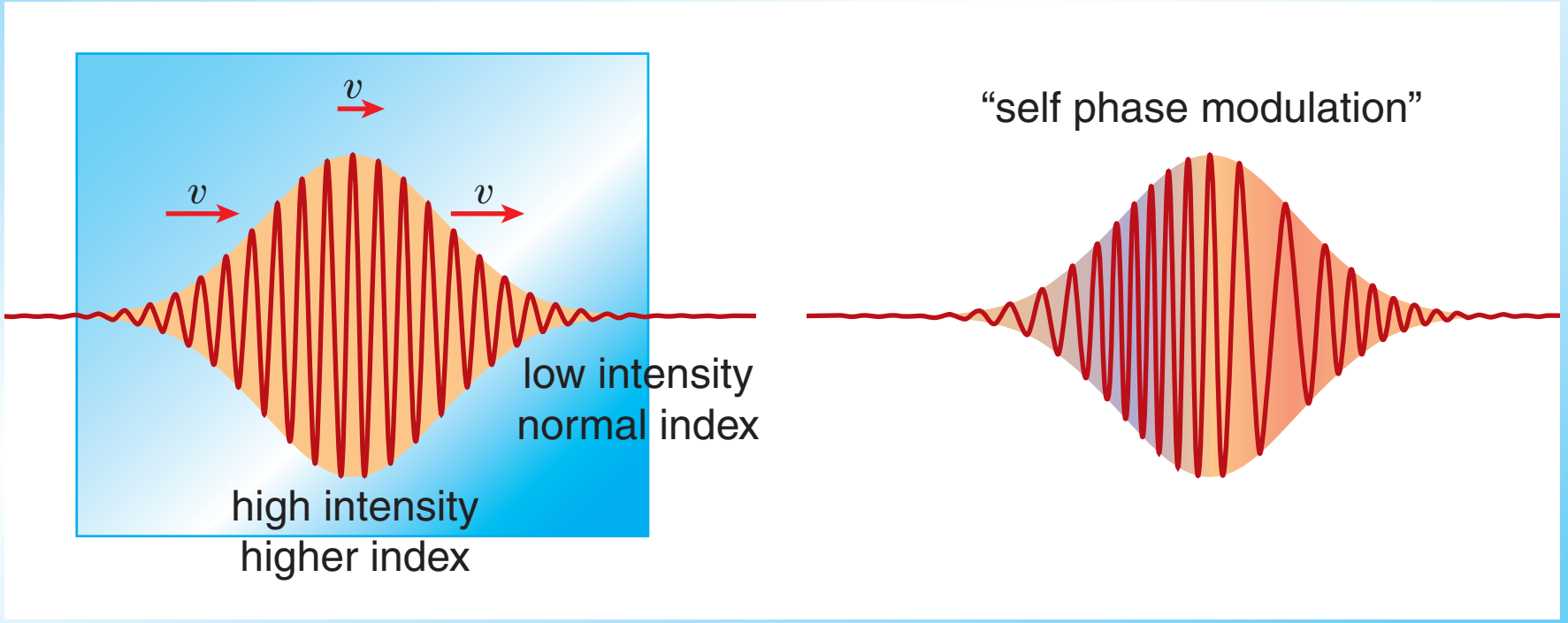
$$n = n_o + n_2 I$$



Phase:

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

Nonlinear optics

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

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Frequency change:

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

Nonlinear optics

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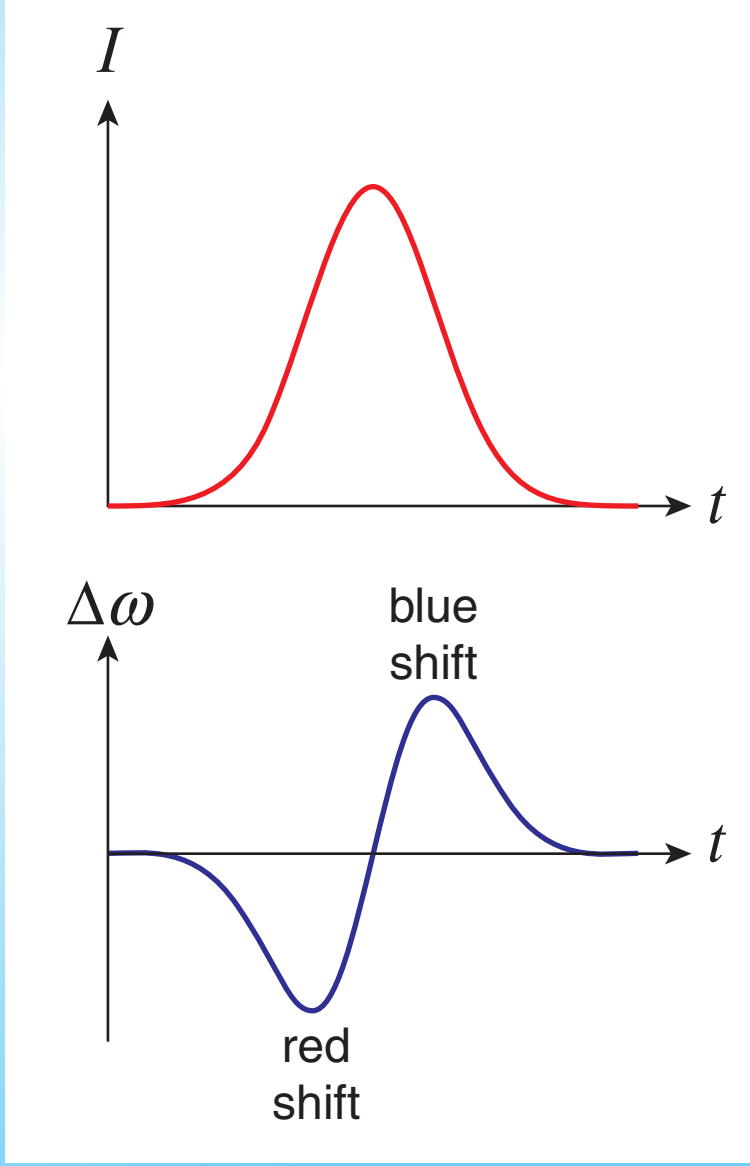
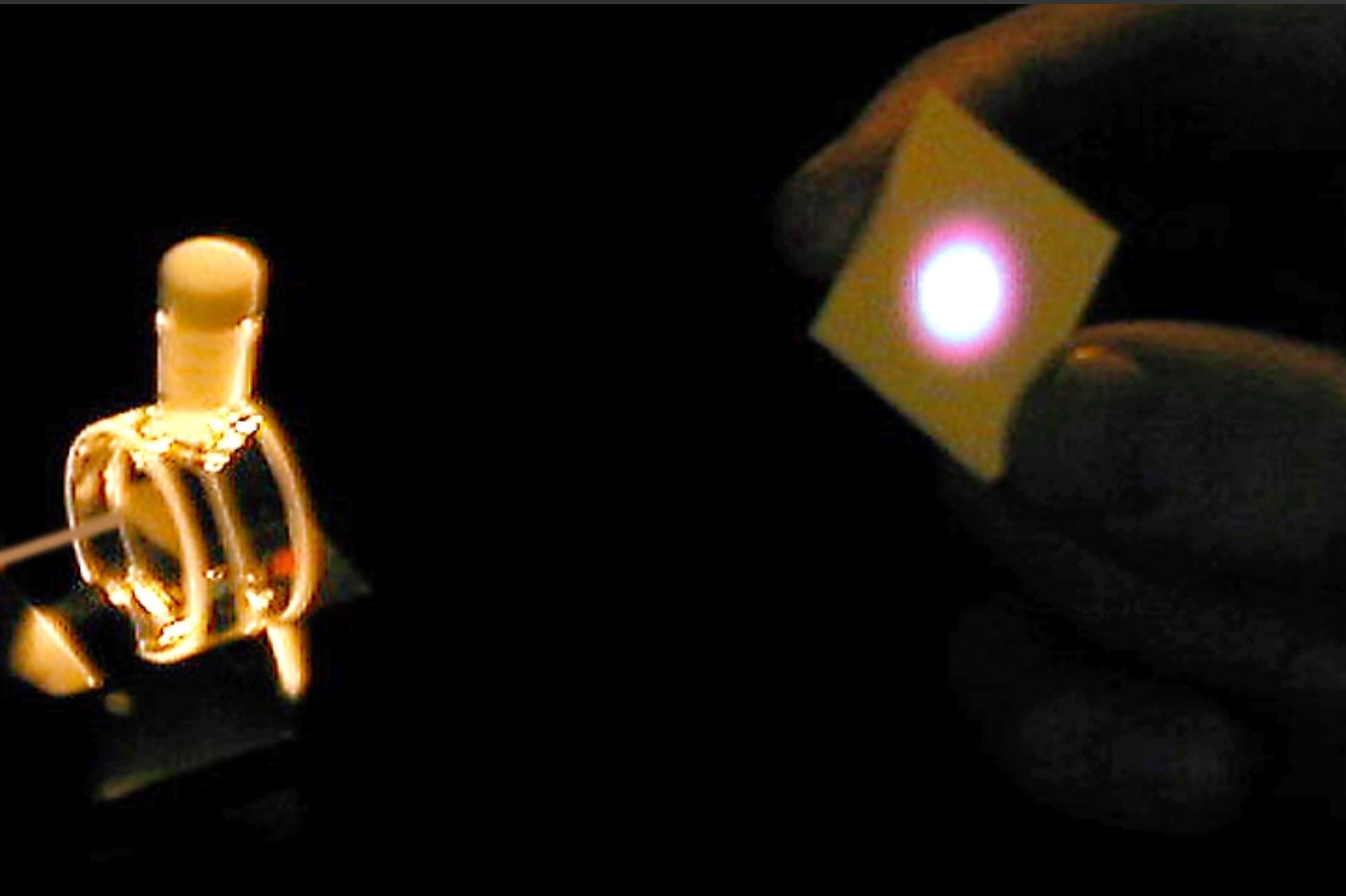
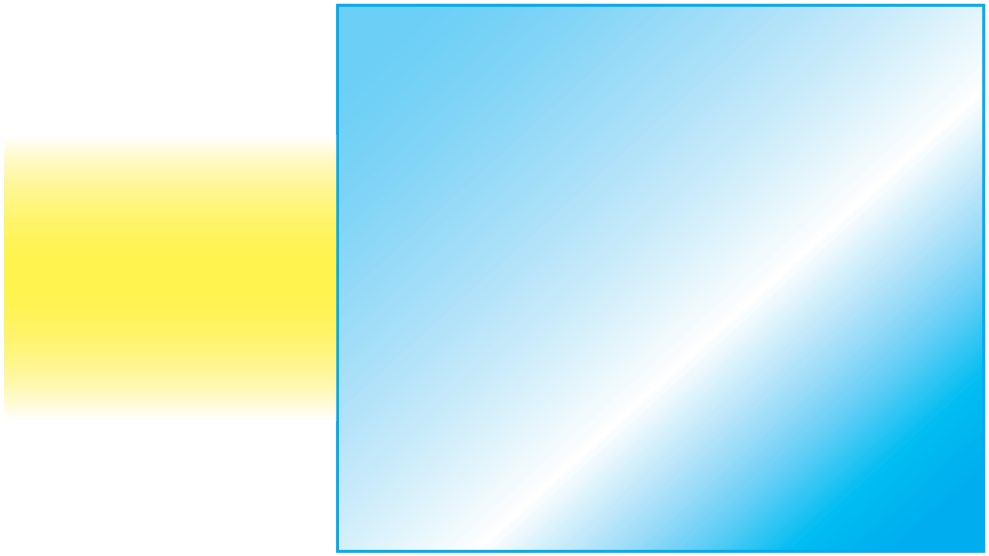


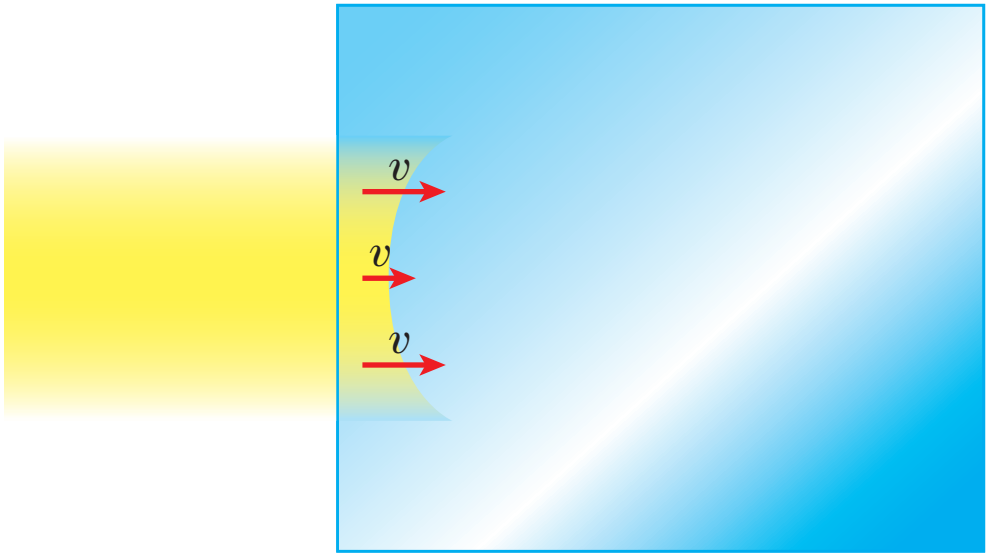
Nonlinear optics



Spatial intensity profile…

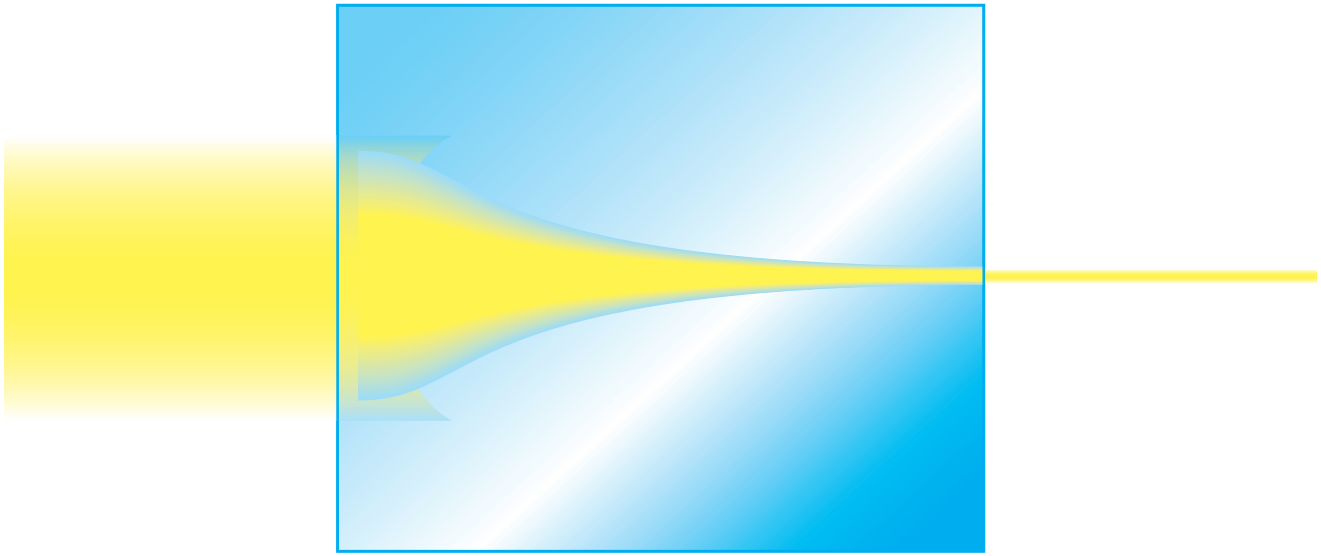


Spatial intensity profile…



Nonlinear optics

…causes self-focusing

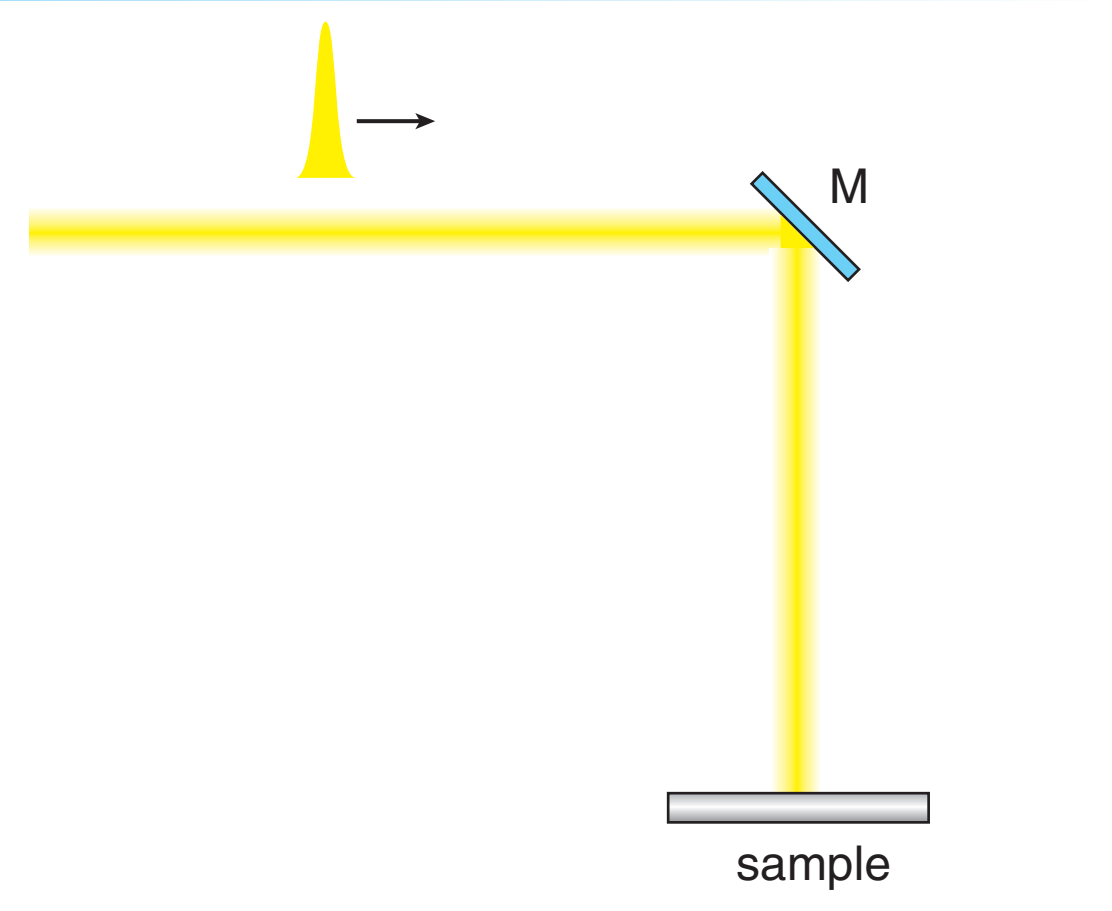


Outline

- linear and nonlinear propagation
- **femtosecond measurements**
- dynamics of phase transitions
- laser-assisted surface etching

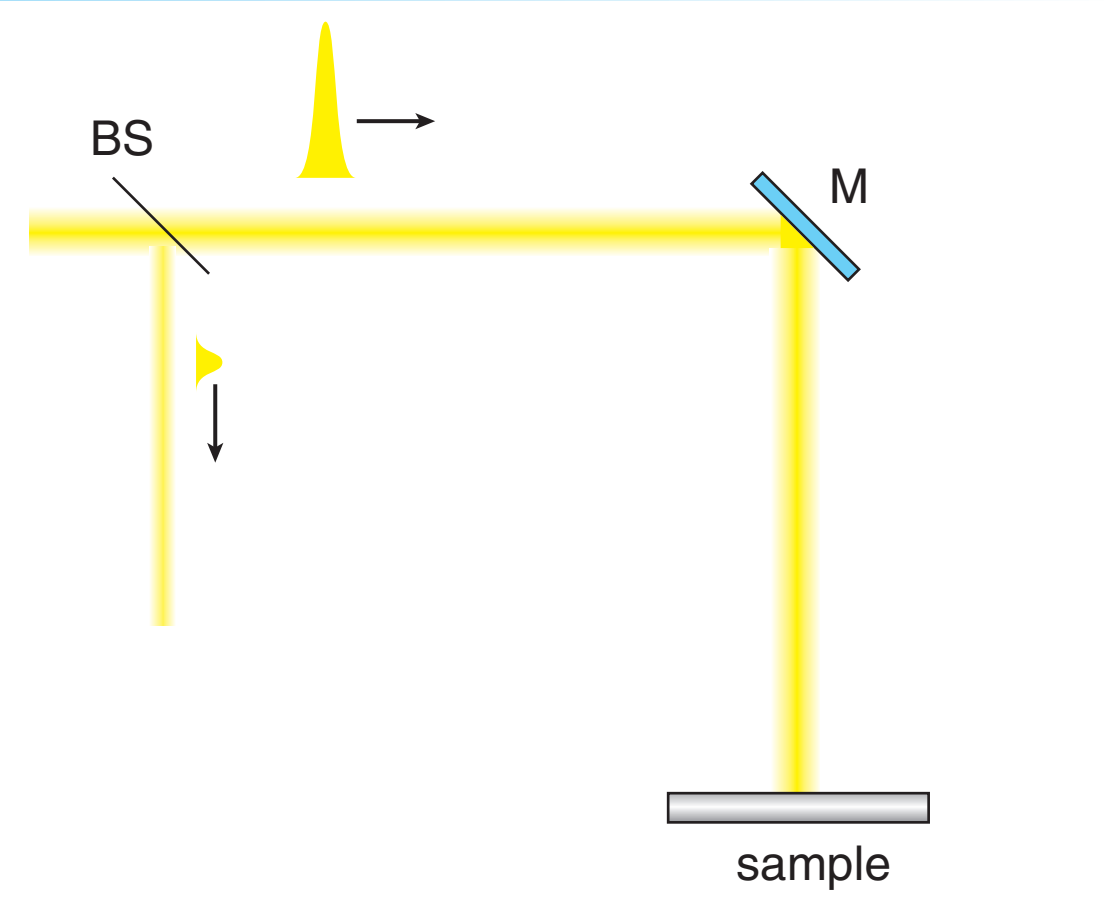
Introduction

How to measure on the femtosecond time scale?



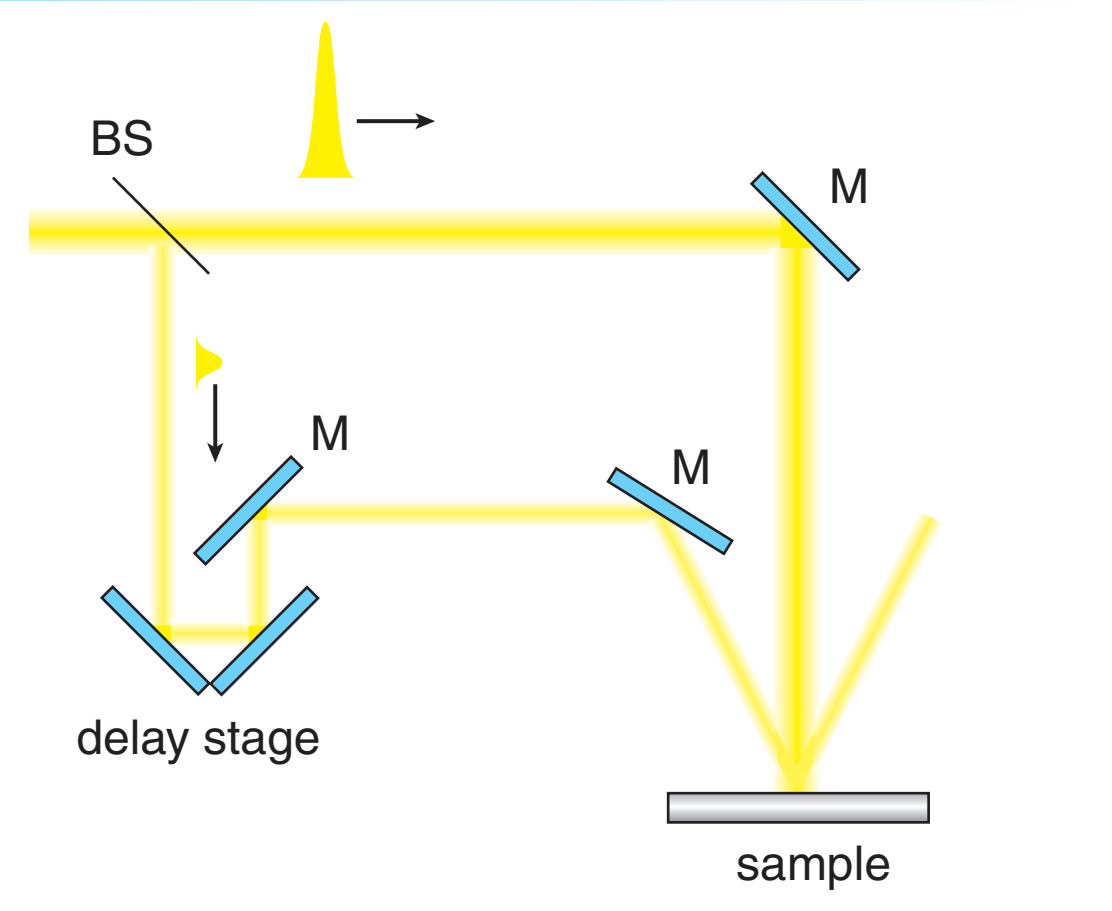
Introduction

Use pump-probe technique



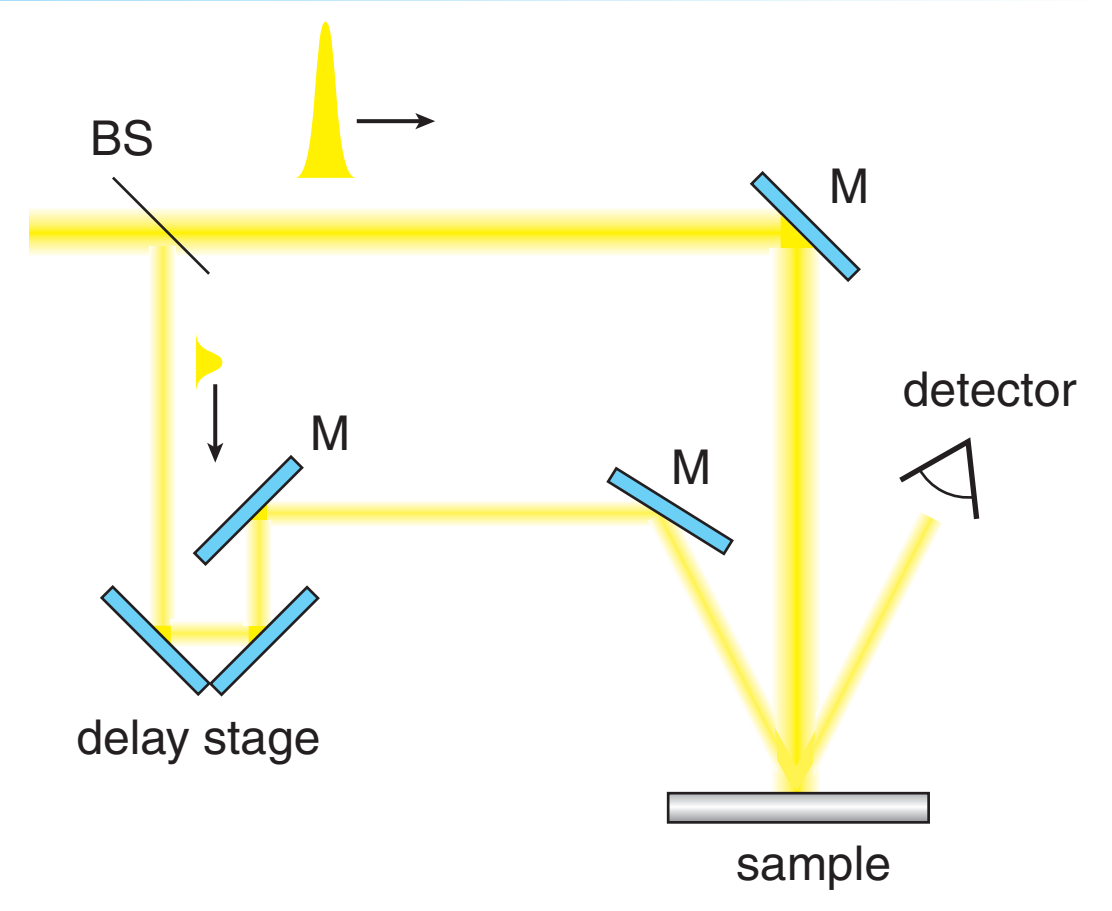
Introduction

Use pump-probe technique



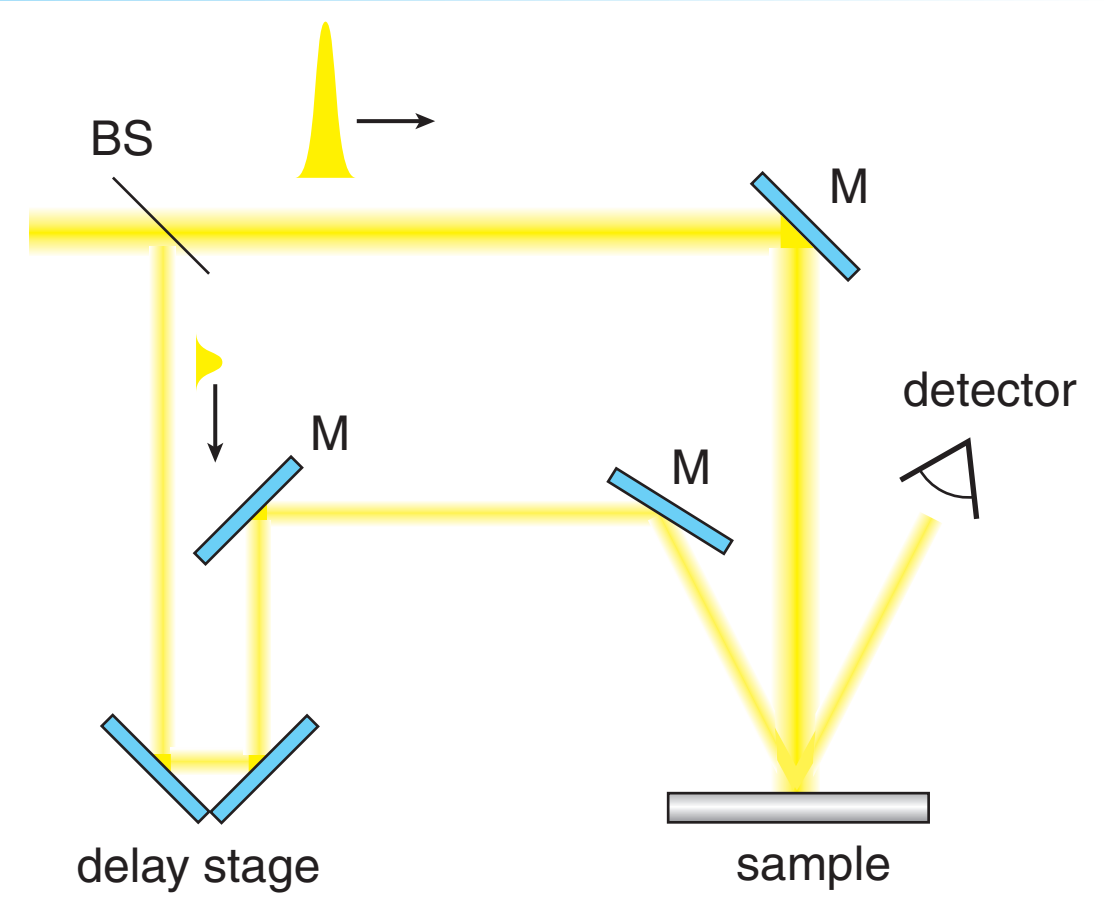
Introduction

Use pump-probe technique



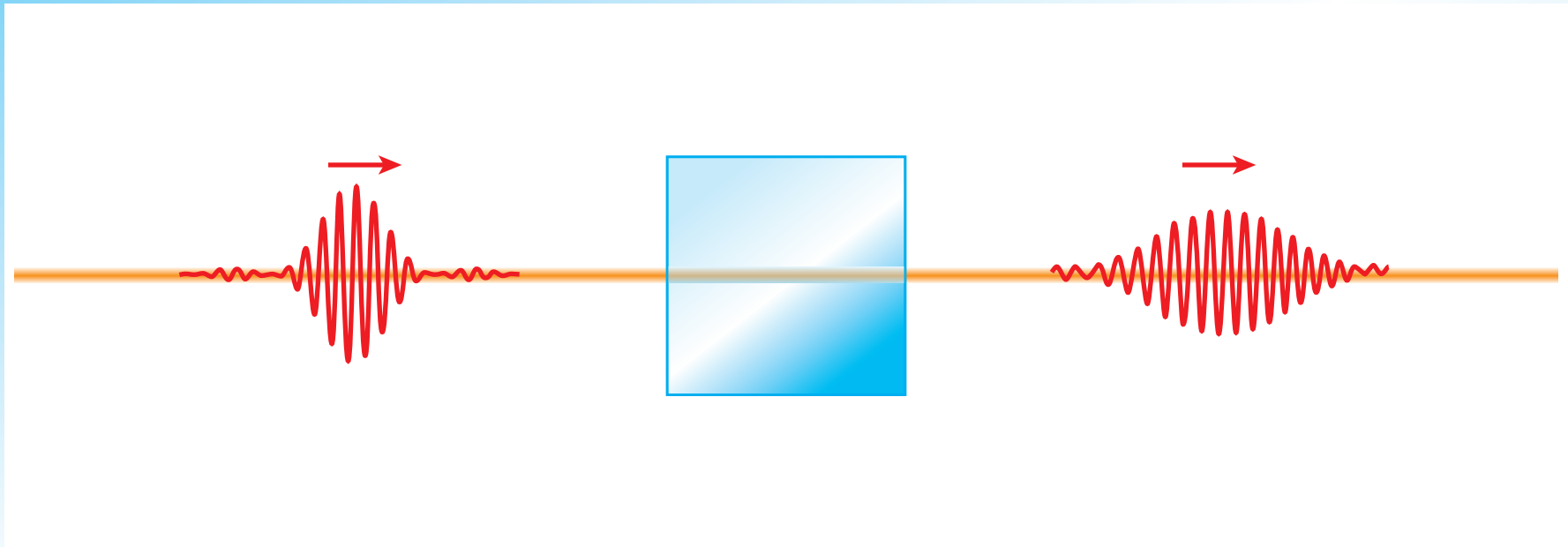
Introduction

Vary delay to get time resolution



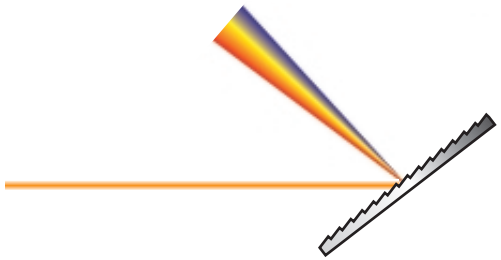
Dispersion compensation

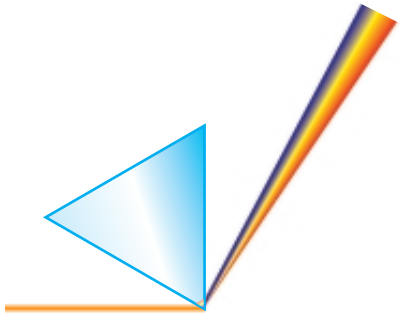
Dispersion stretches the pulse



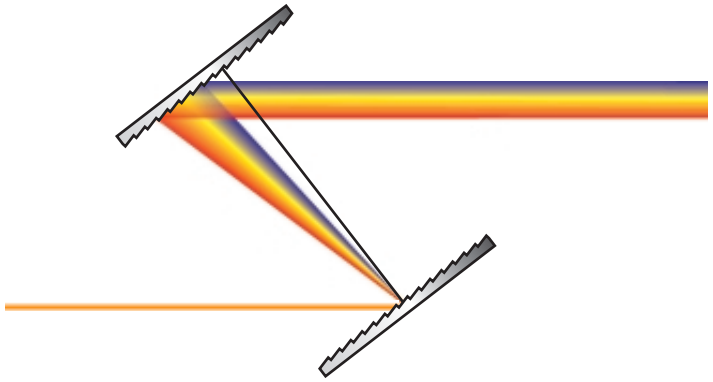
Compensate by rearranging spectral components!

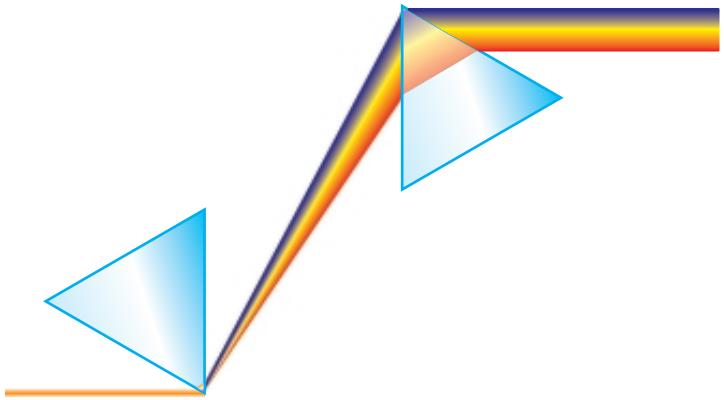
Dispersion compensation



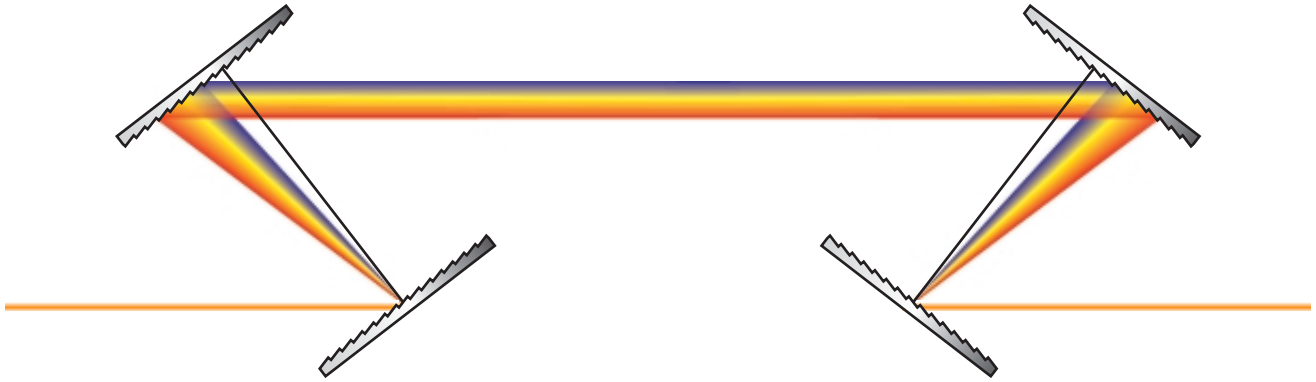


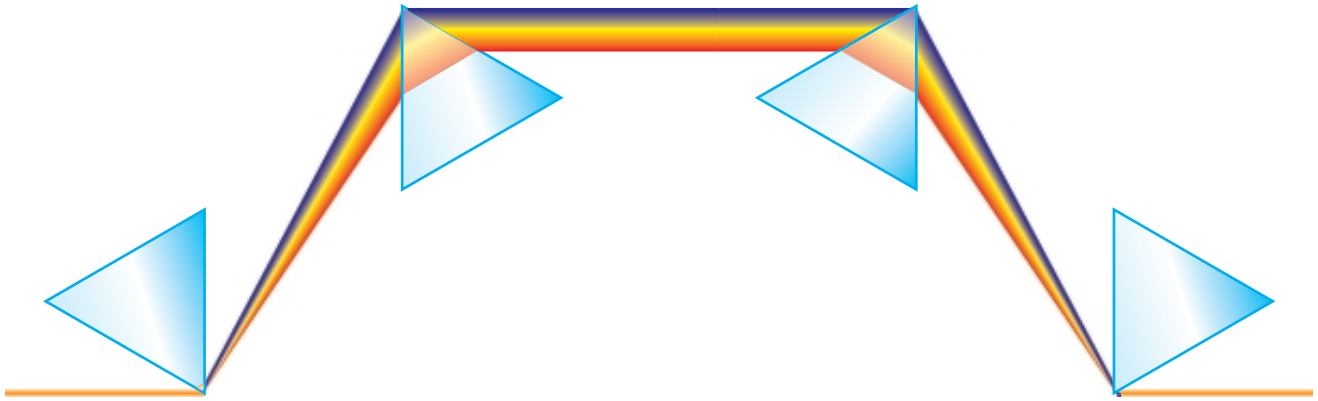
Dispersion compensation





Dispersion compensation



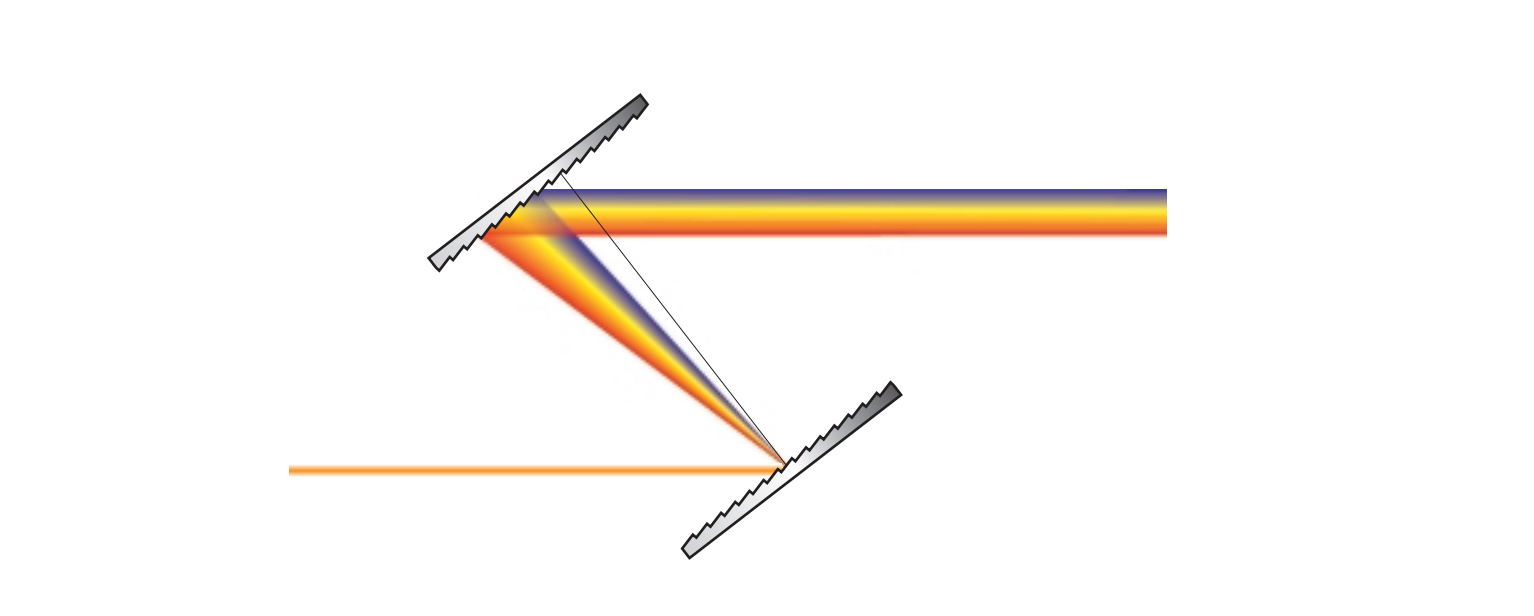


Dispersion compensation

How do these arrangements work?

Dispersion compensation

Does path length difference compensate?



Dispersion compensation

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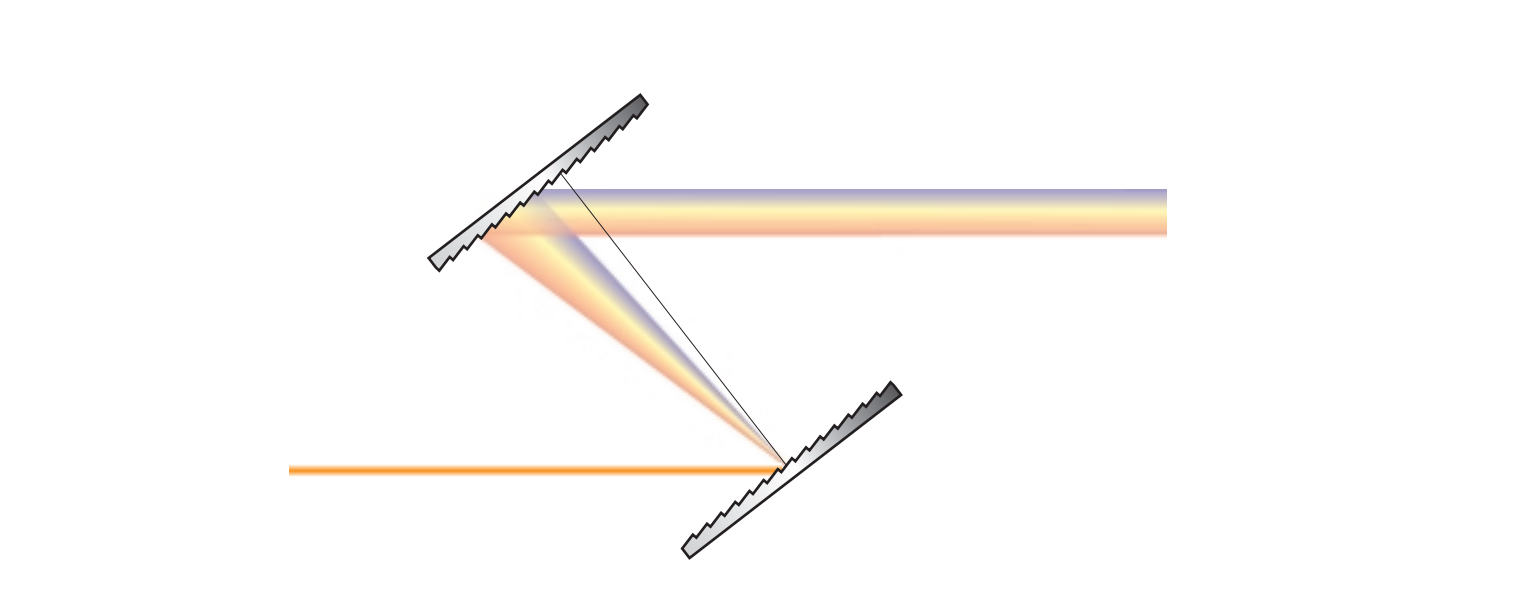

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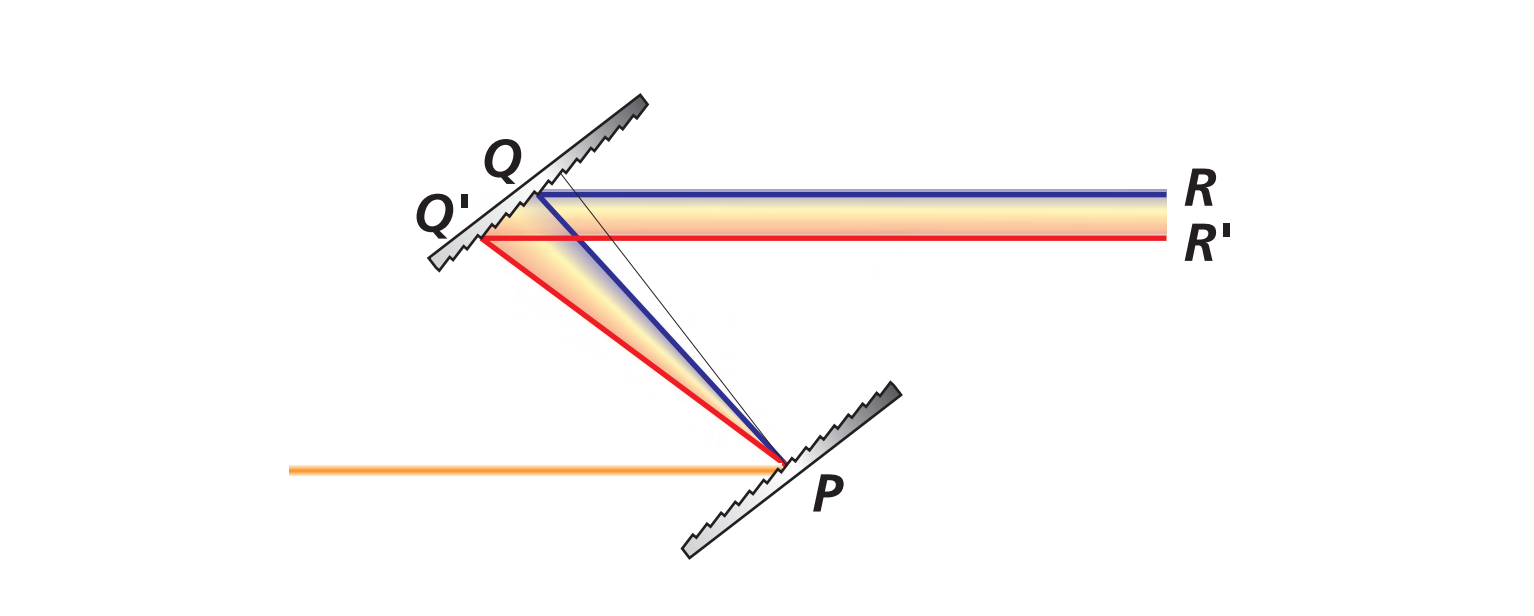
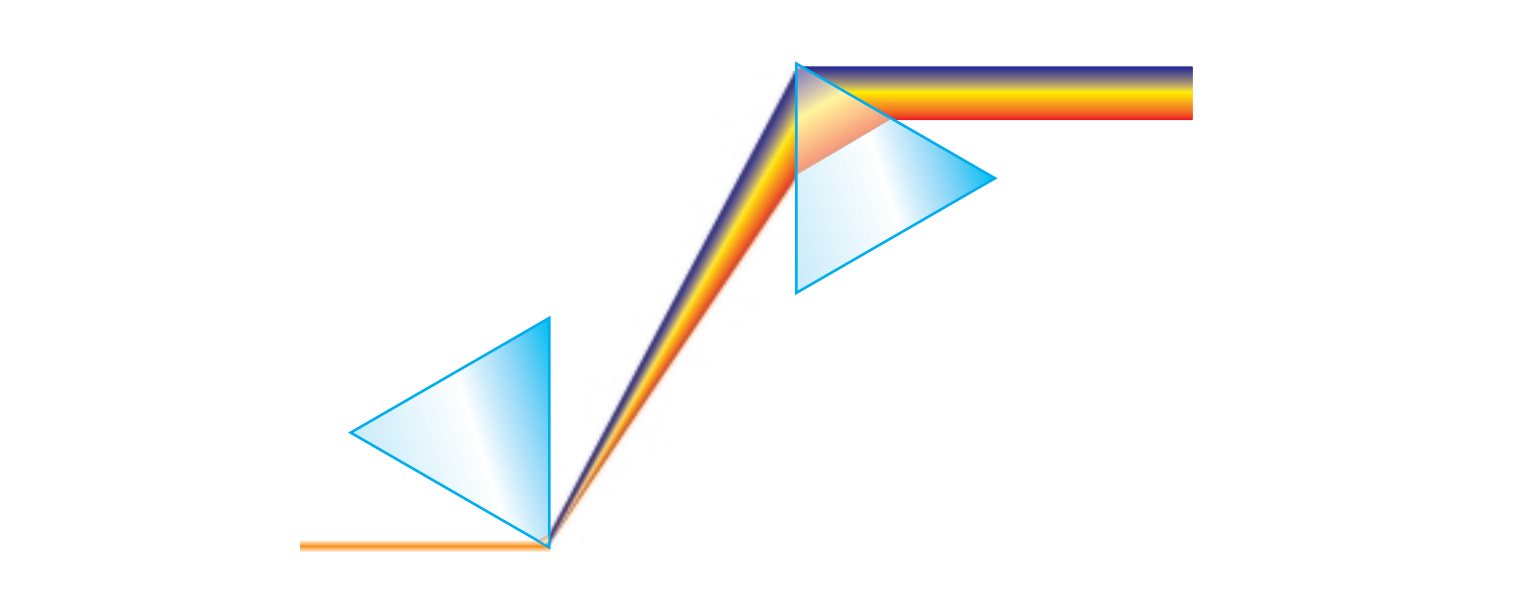


Grating gives low frequency longer path length…

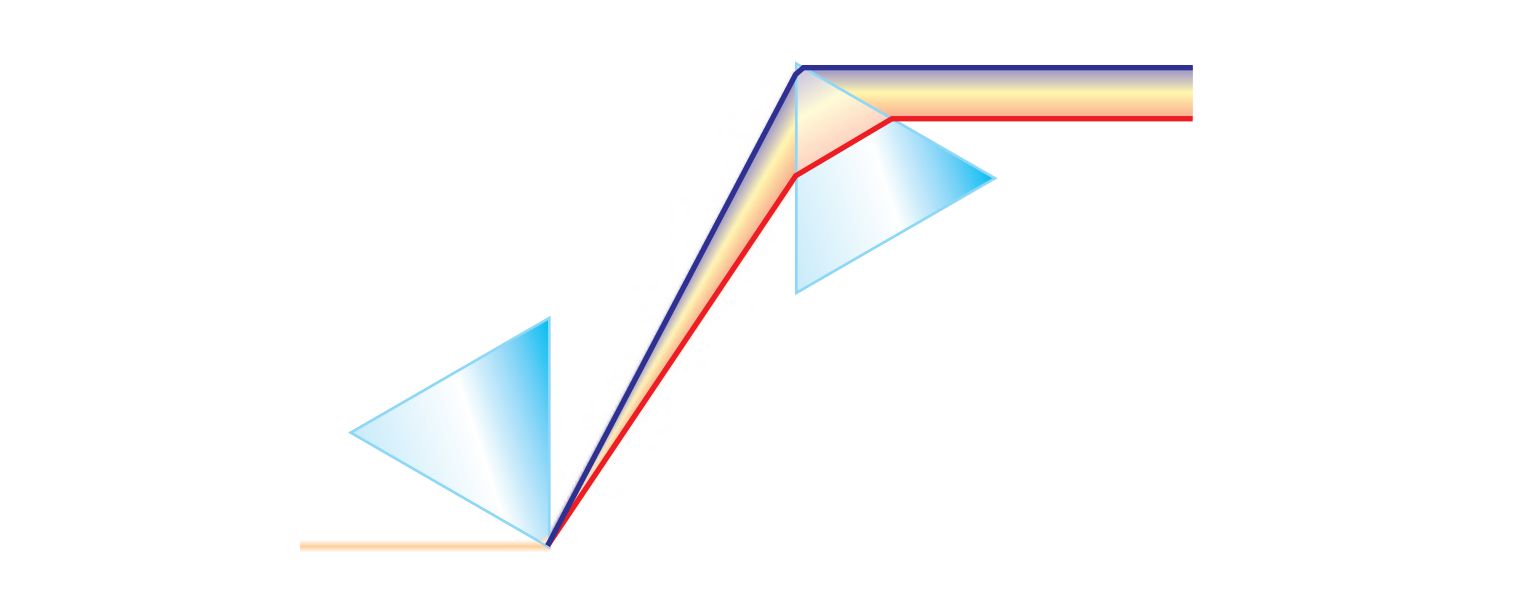
Dispersion compensation

Does path length difference compensate?



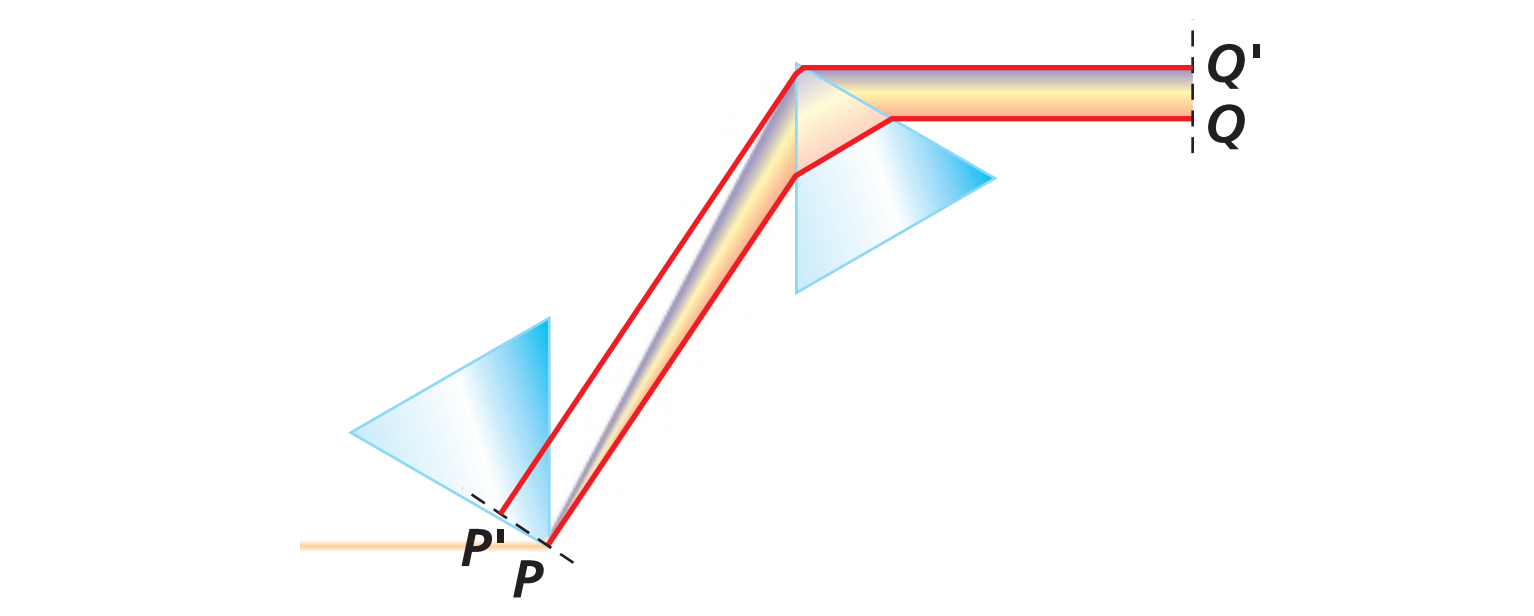
Dispersion compensation

Does path length difference compensate?



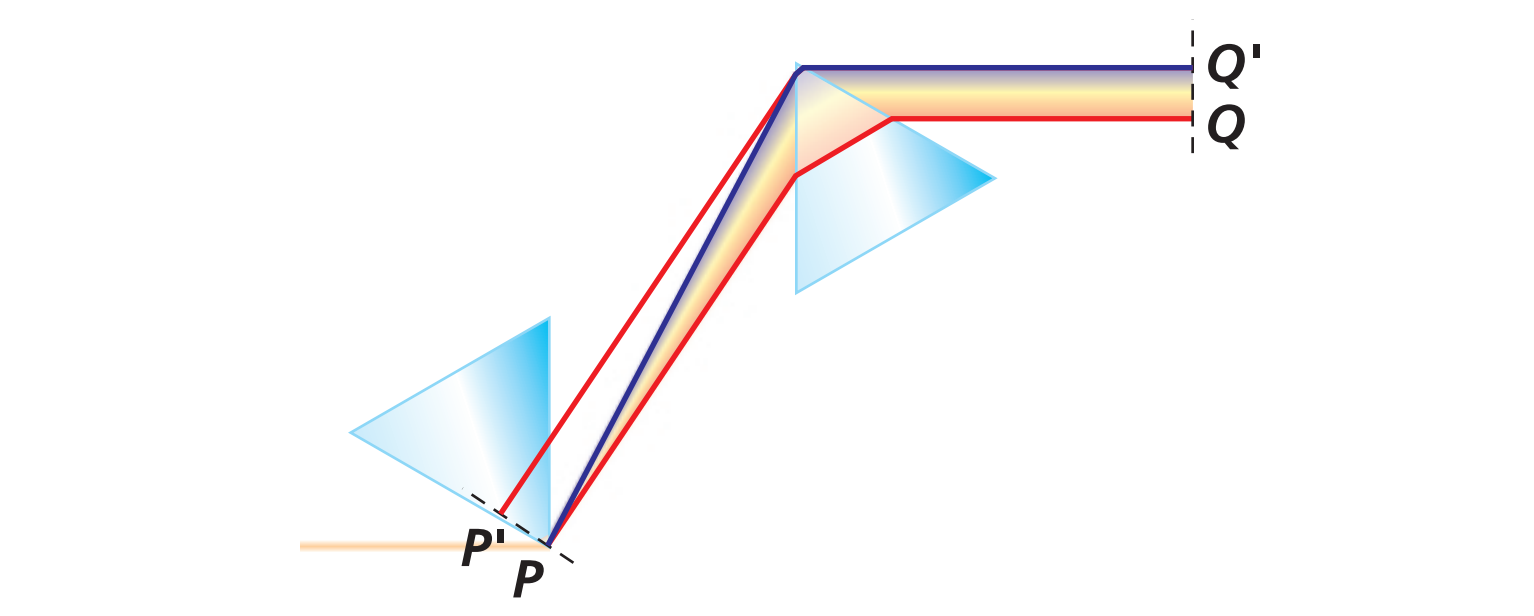
Dispersion compensation

Does path length difference compensate?



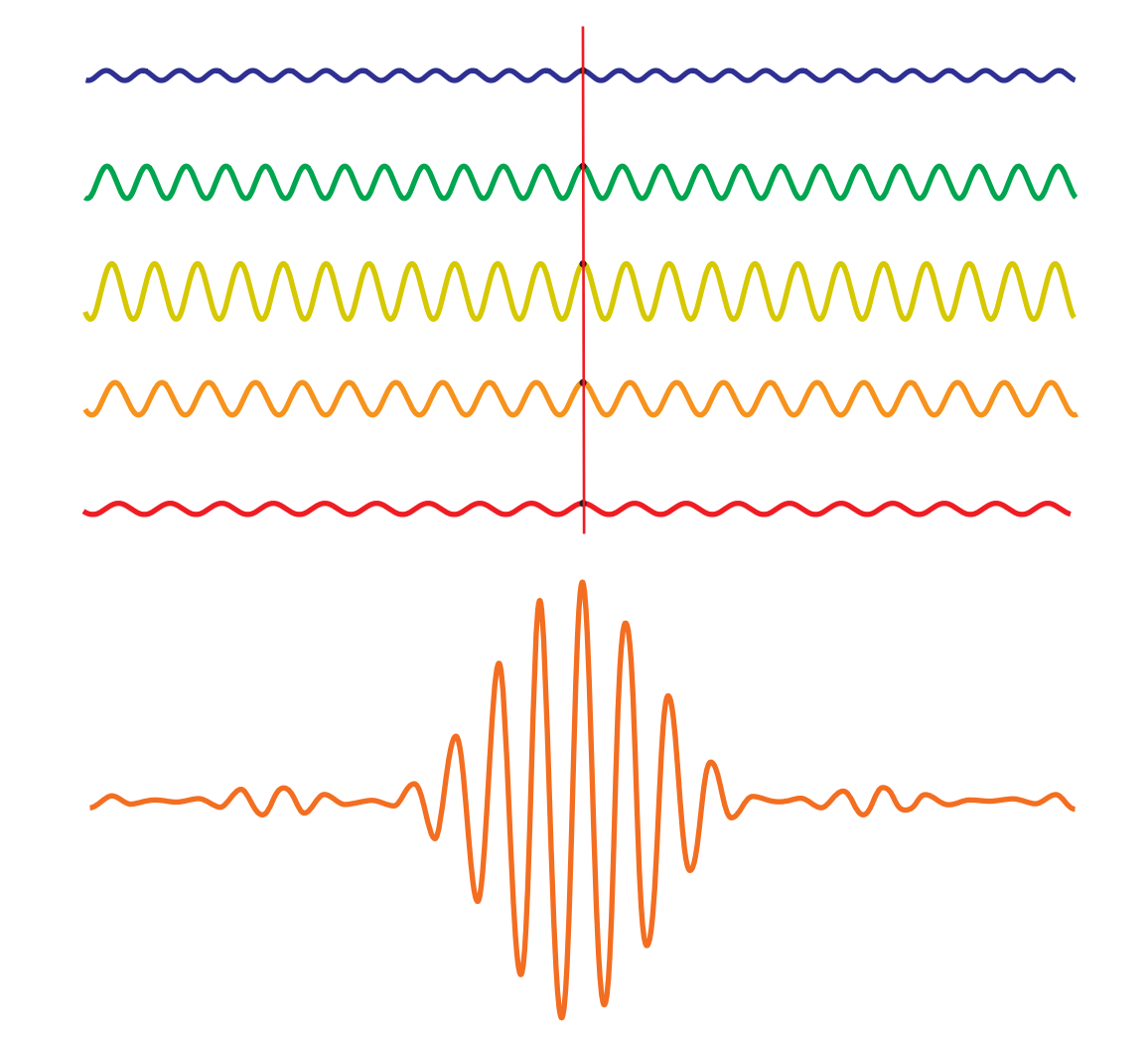
Dispersion compensation

Does path length difference compensate?

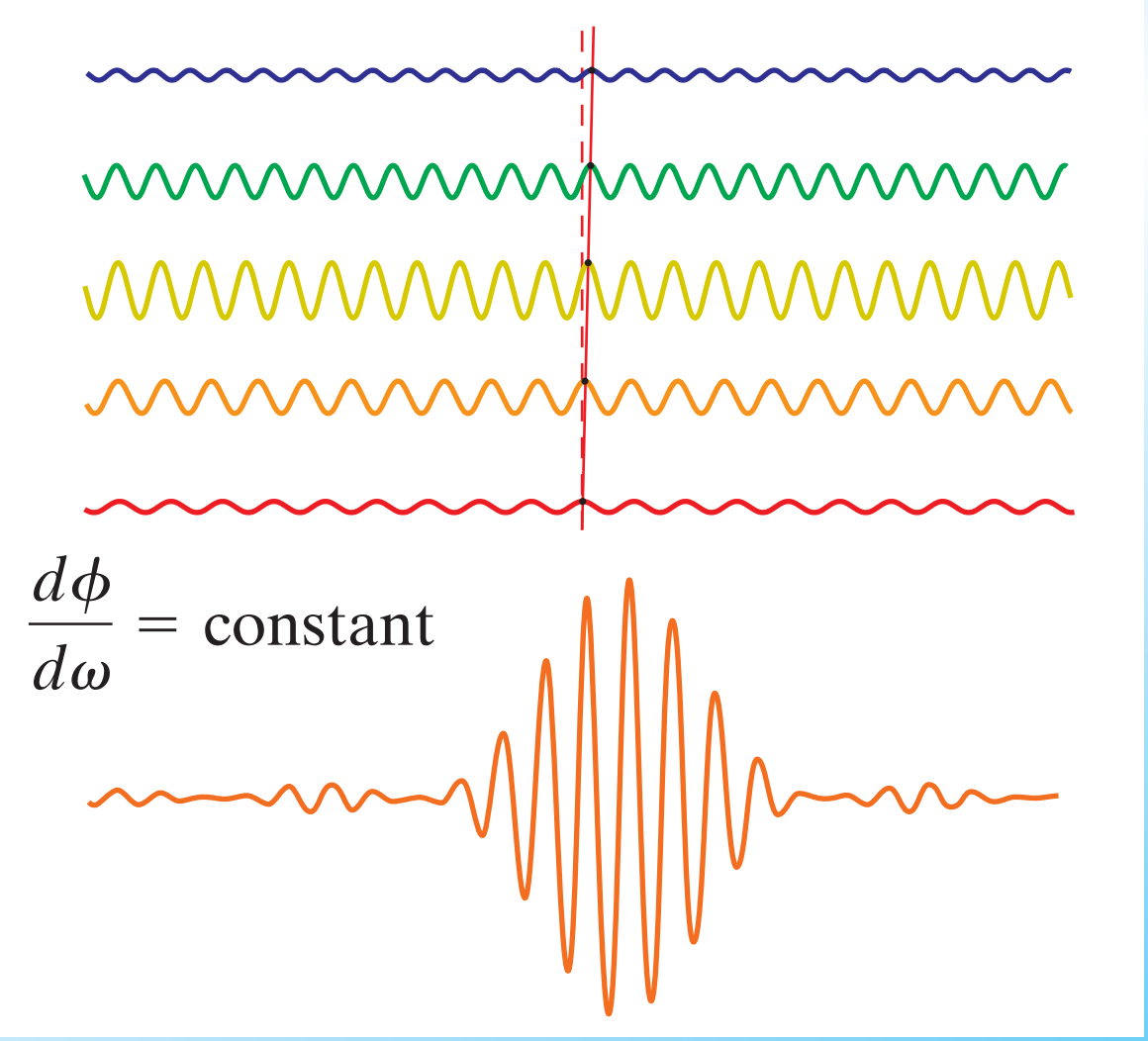


…so prism gives low frequency *shorter* path length…

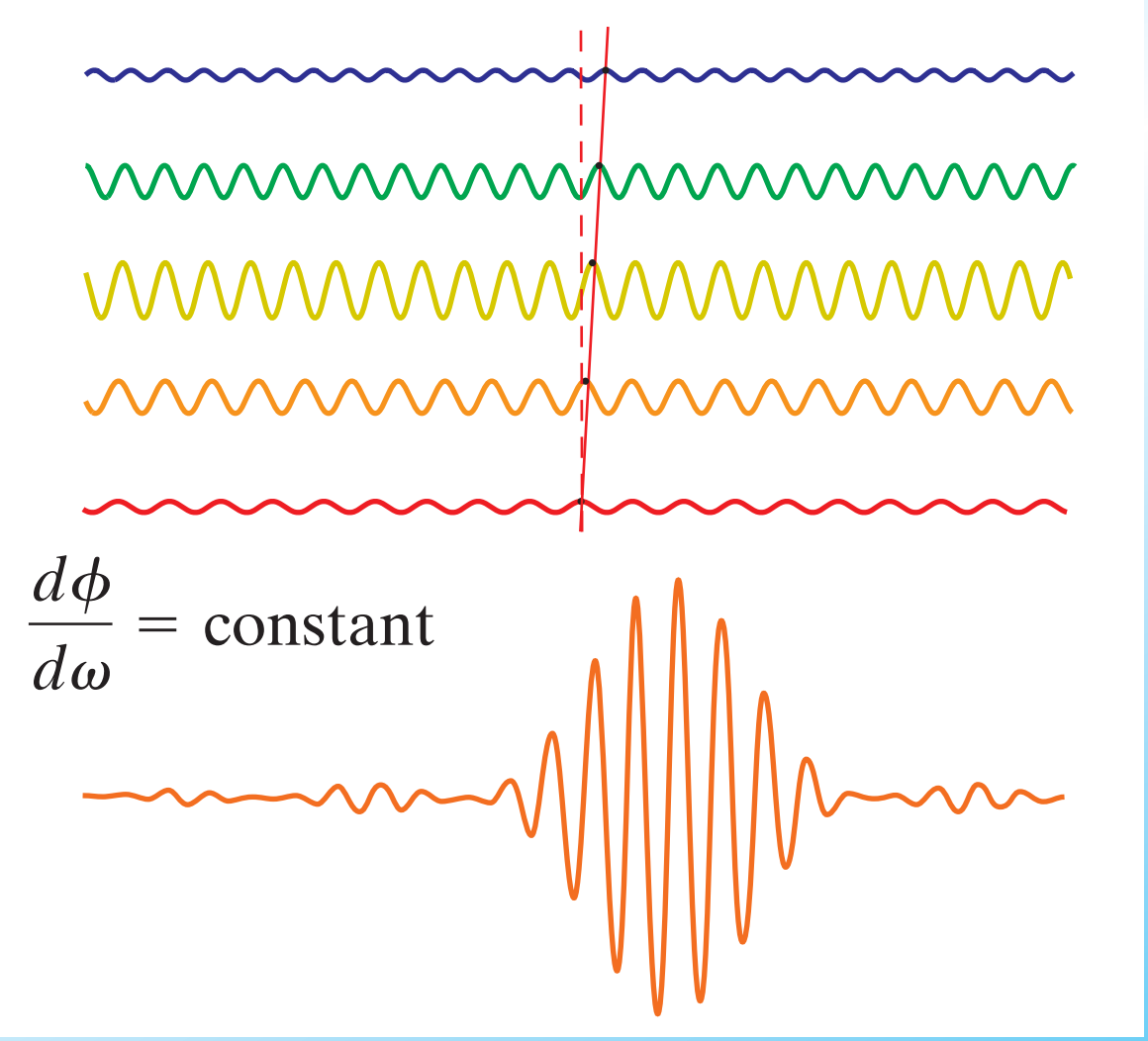
Dispersion compensation



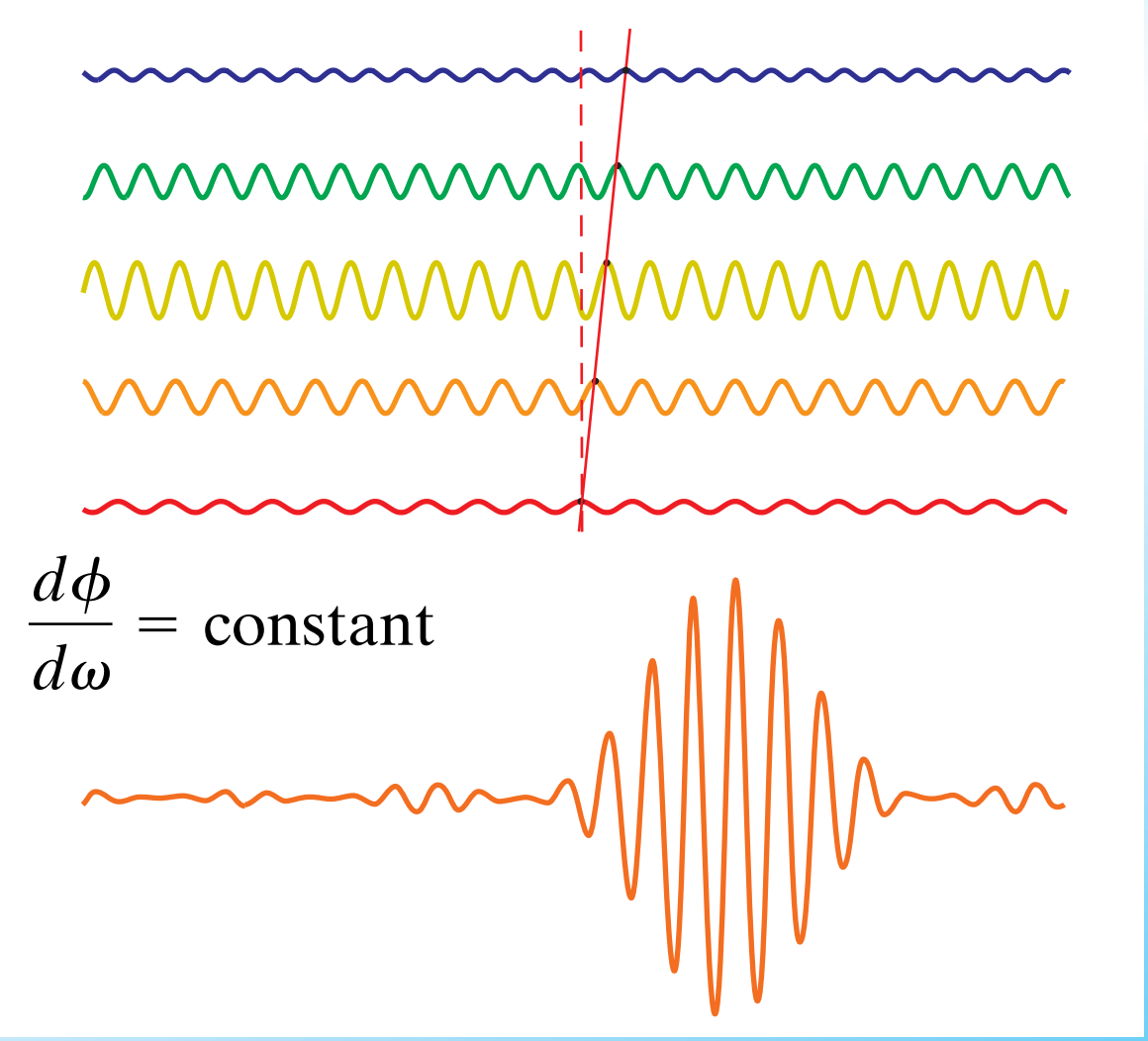
Dispersion compensation



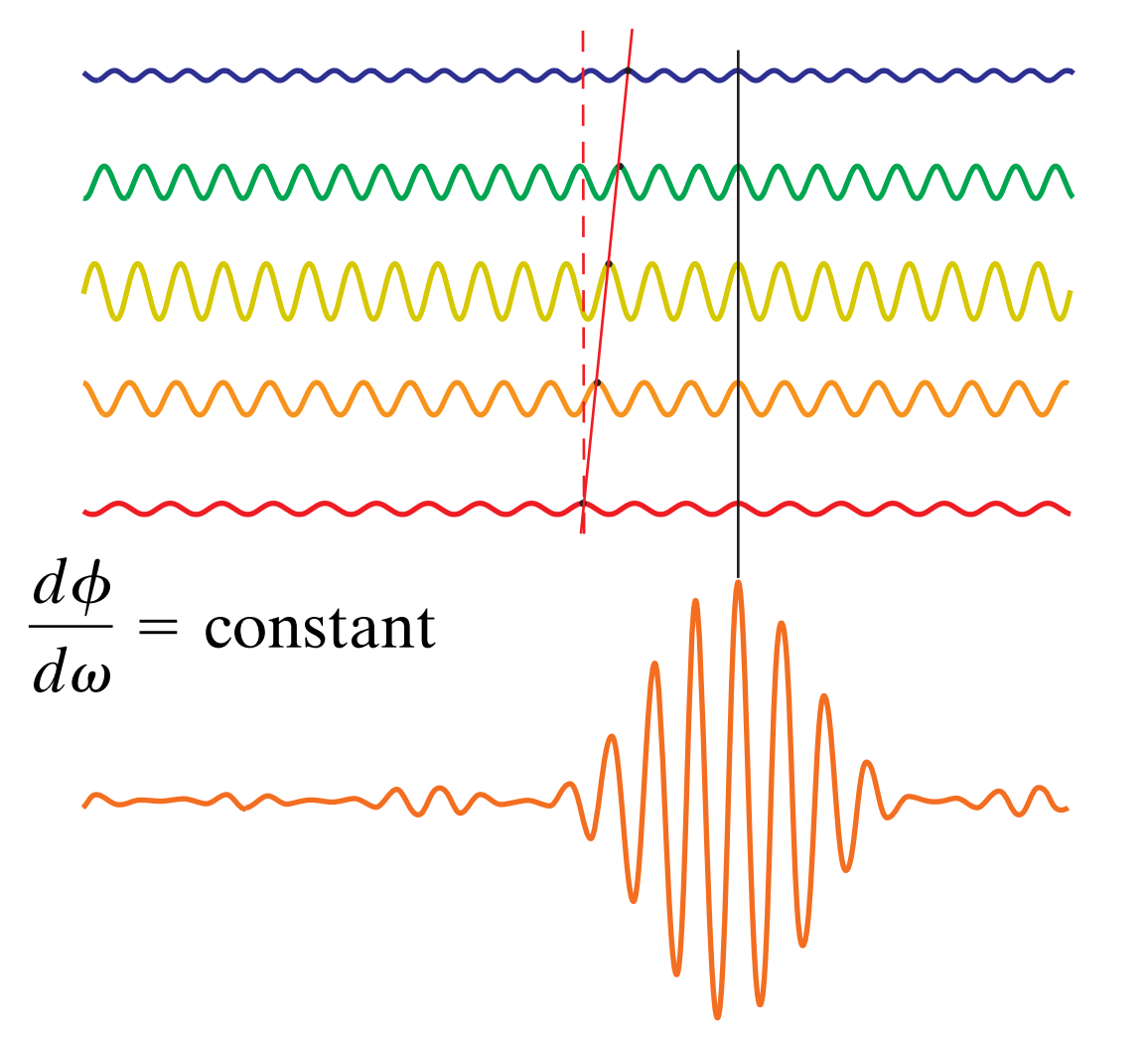
Dispersion compensation



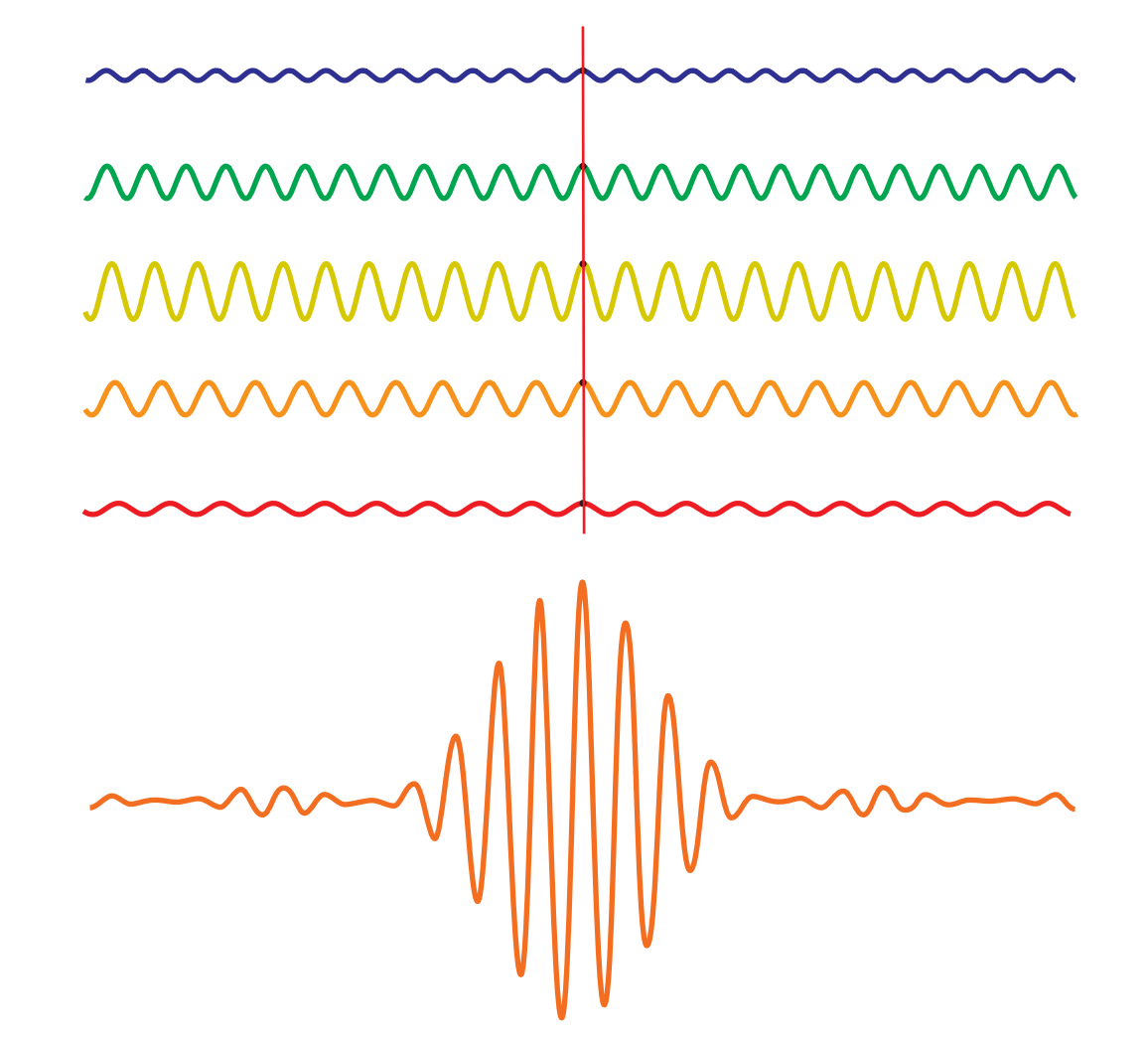
Dispersion compensation



Dispersion compensation



Dispersion compensation



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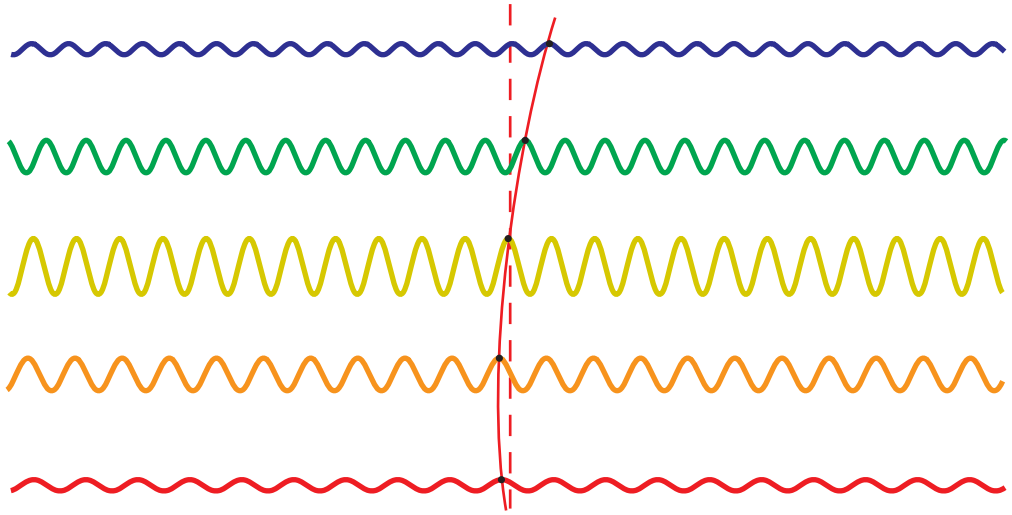
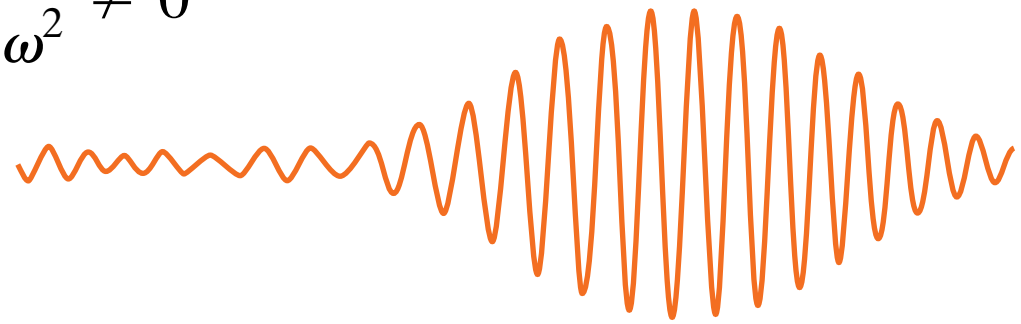


$$\frac{d^2\phi}{d\omega^2} \neq 0$$

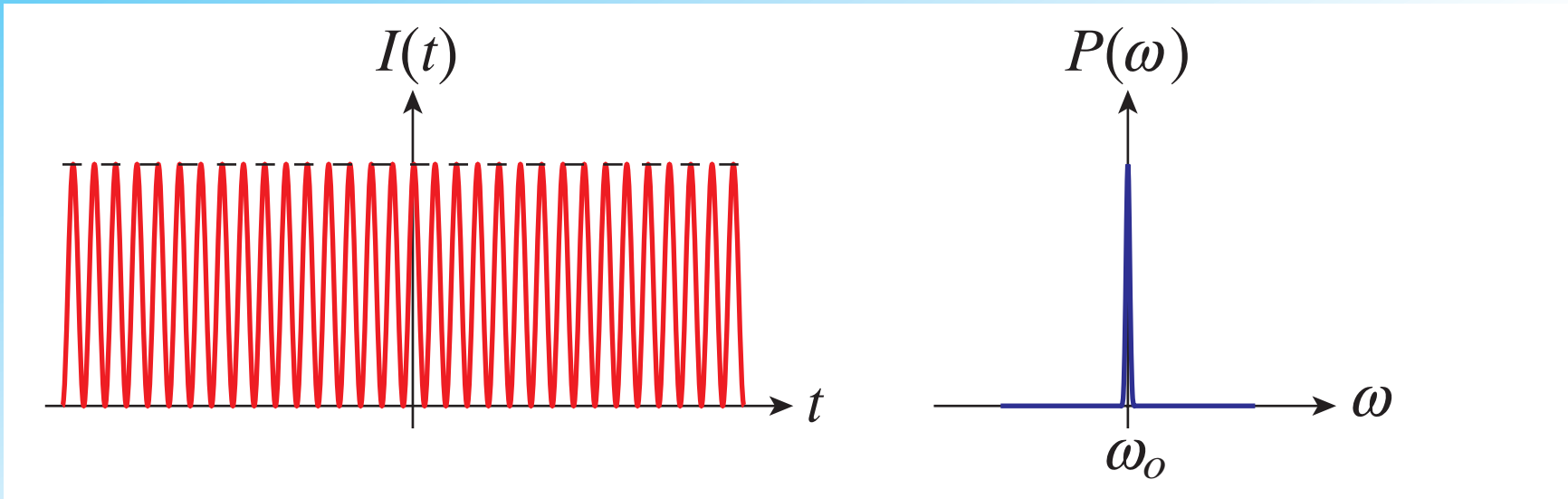


Dispersion compensation

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

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

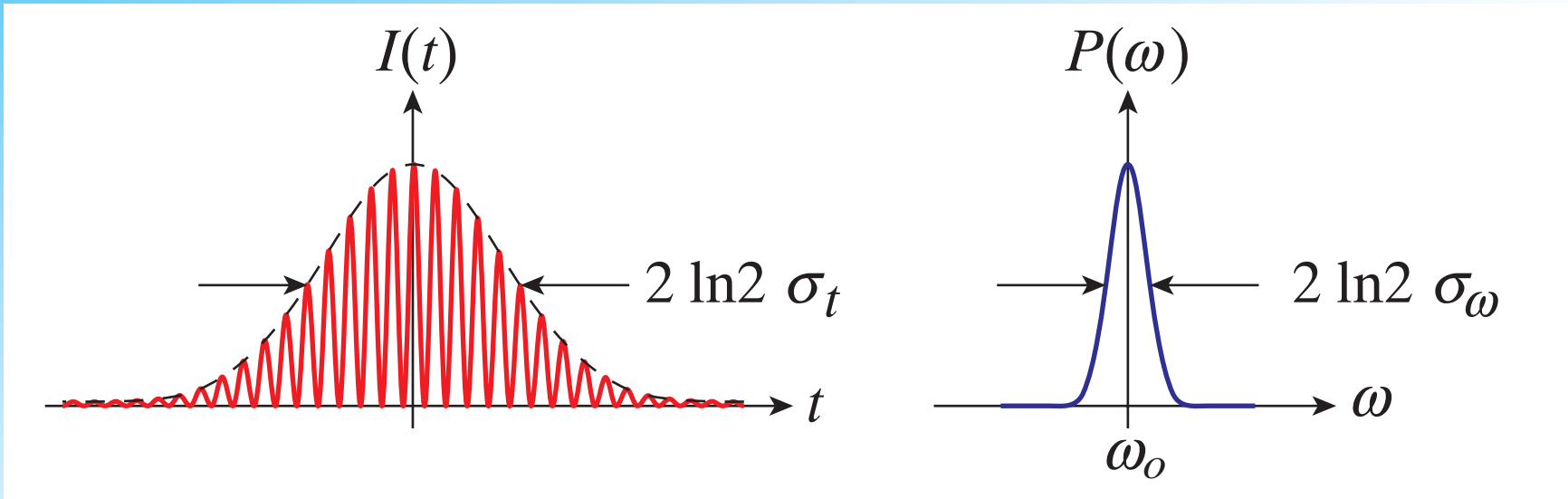
Representation of pulses



Spectrum of sinusoidal intensity is a delta function

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

Representation of pulses



Modulate amplitude

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

Representation of pulses

Fourier relations:

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

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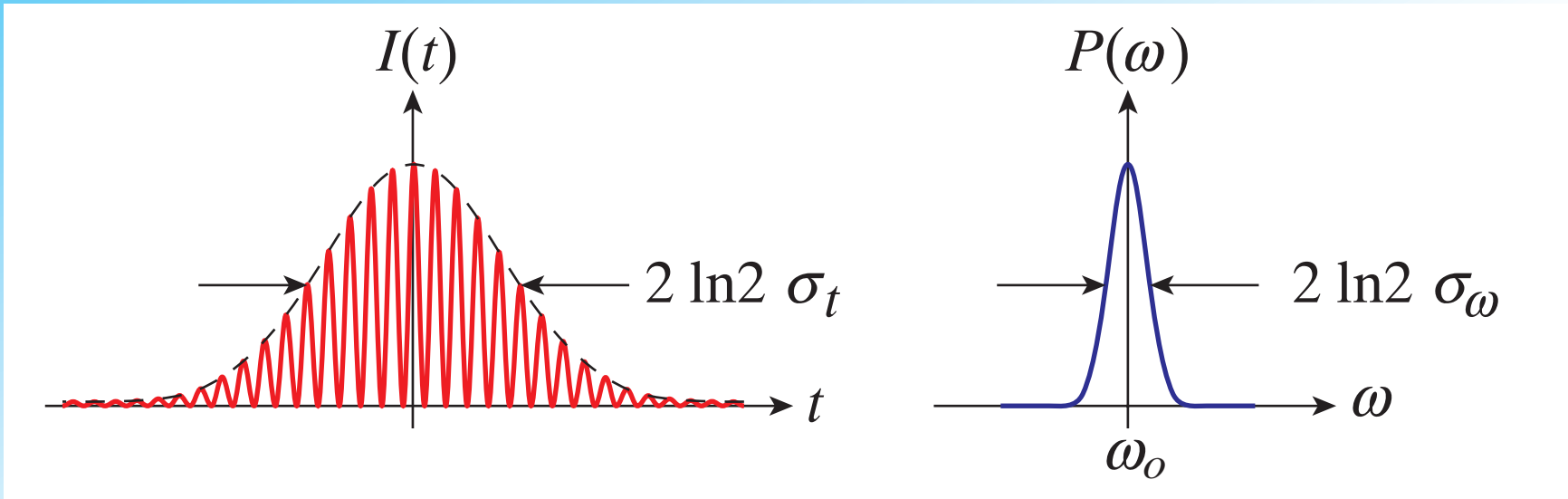
$$= \sigma_t\exp\left[-\frac{\sigma_t^2(\omega-\omega_o)^2}{2}\right] \equiv \sigma_t\exp\left[-\frac{(\omega-\omega_o)^2}{2\sigma_\omega^2}\right]$$

Representation of pulses



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

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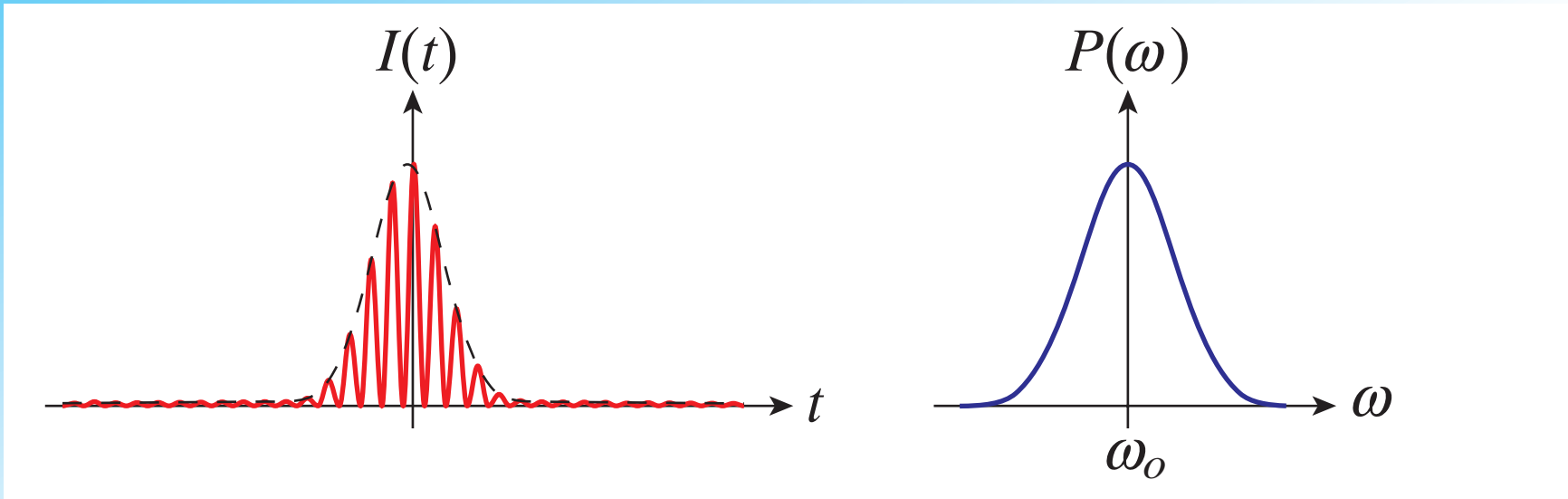

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Joint time-frequency representation

Wigner representation:

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

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

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Joint time-frequency representation

Energy:

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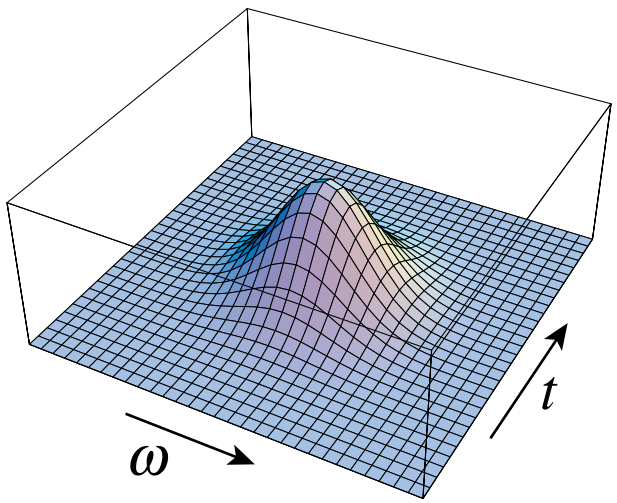

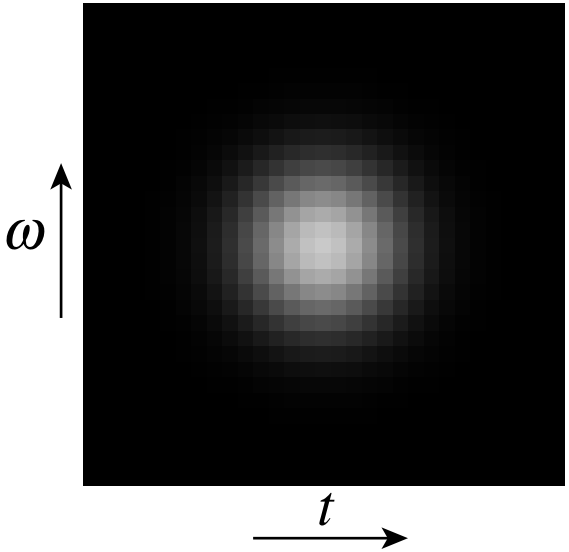

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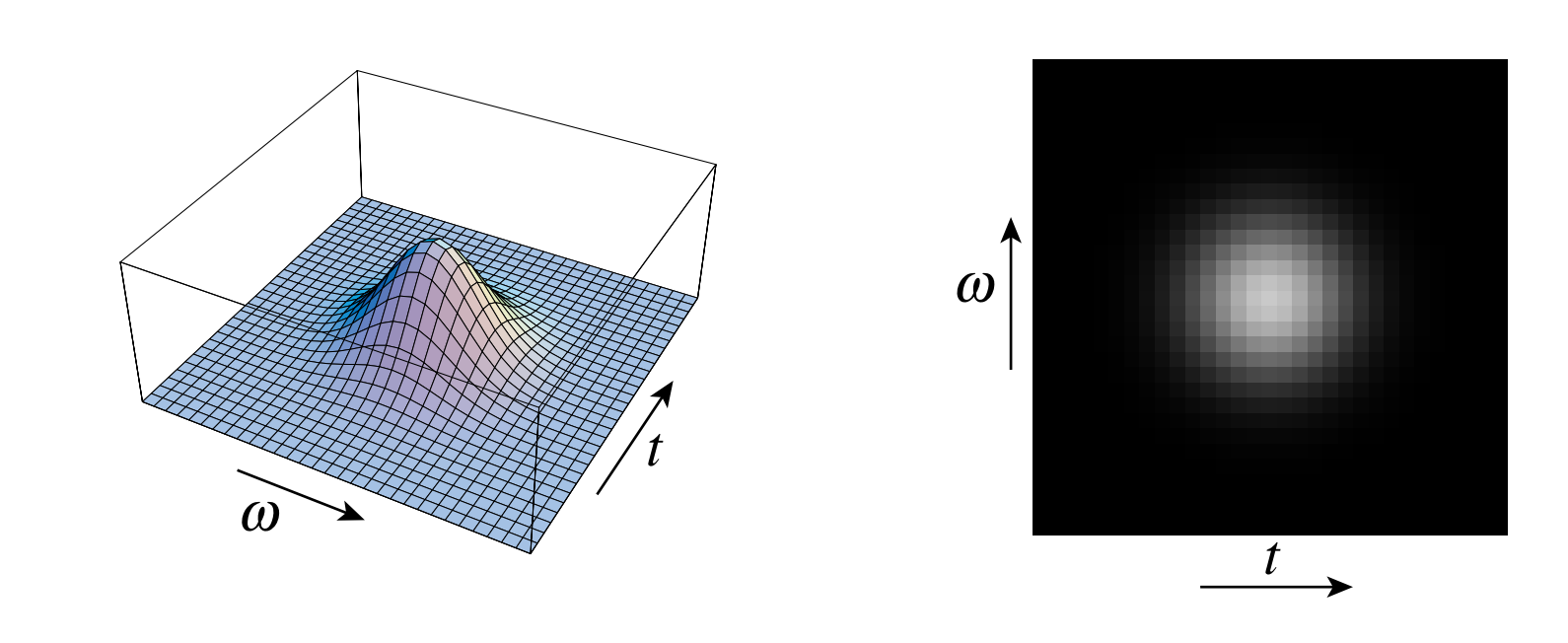

$W(t,\omega)$ **must be nonzero in phase-space area larger than** π

Joint time-frequency representation

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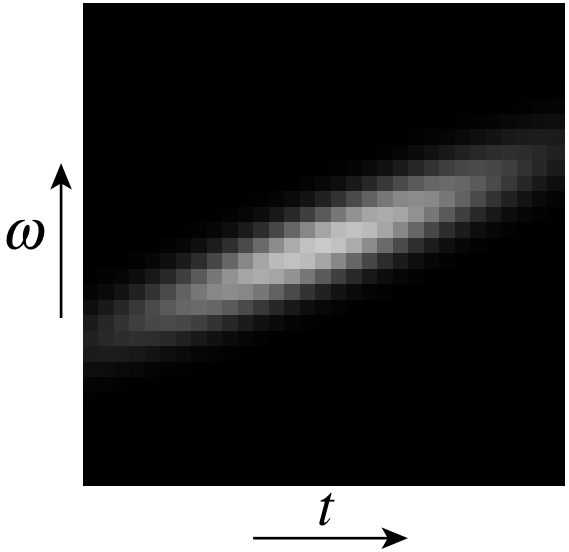
chirped pulse



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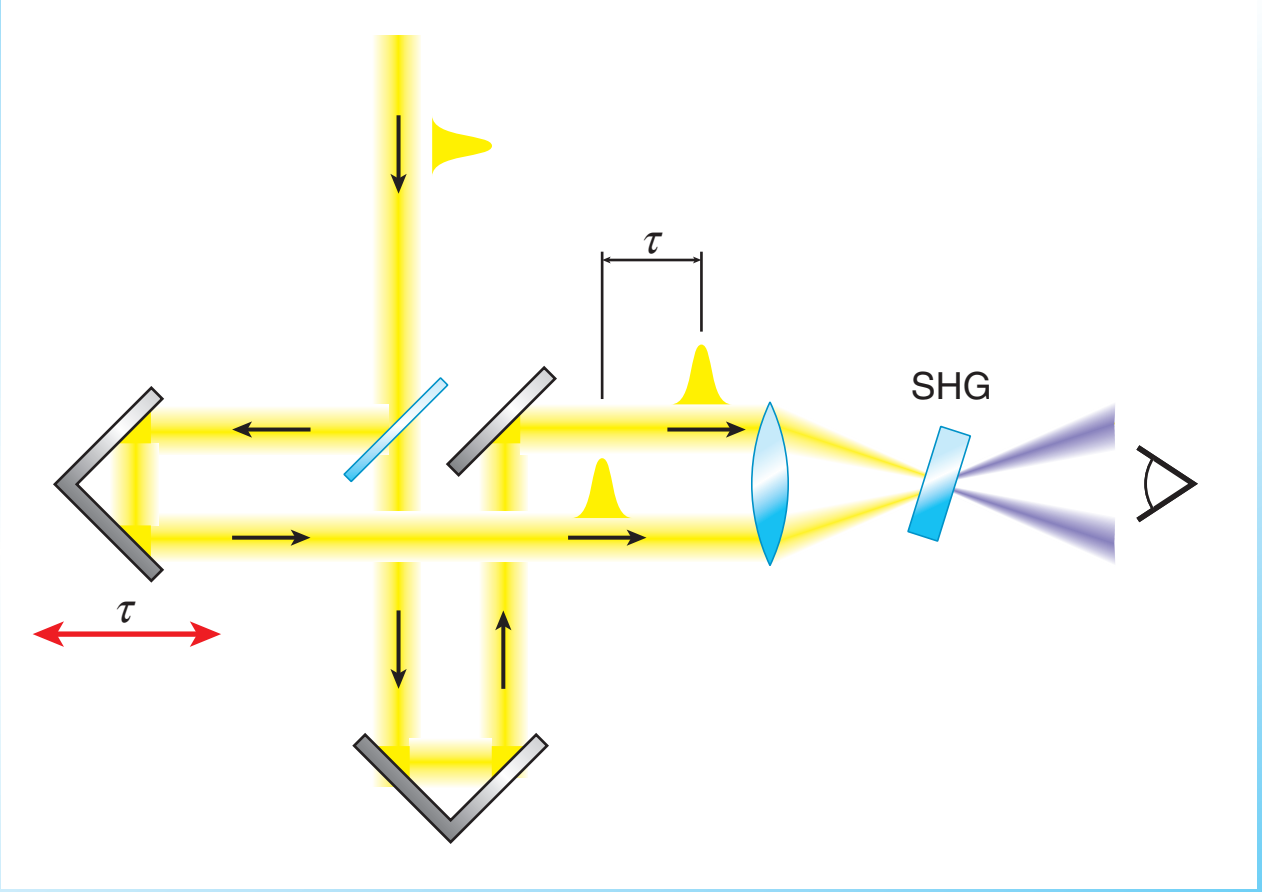
Temporal characterization

Use pulse to measure itself…



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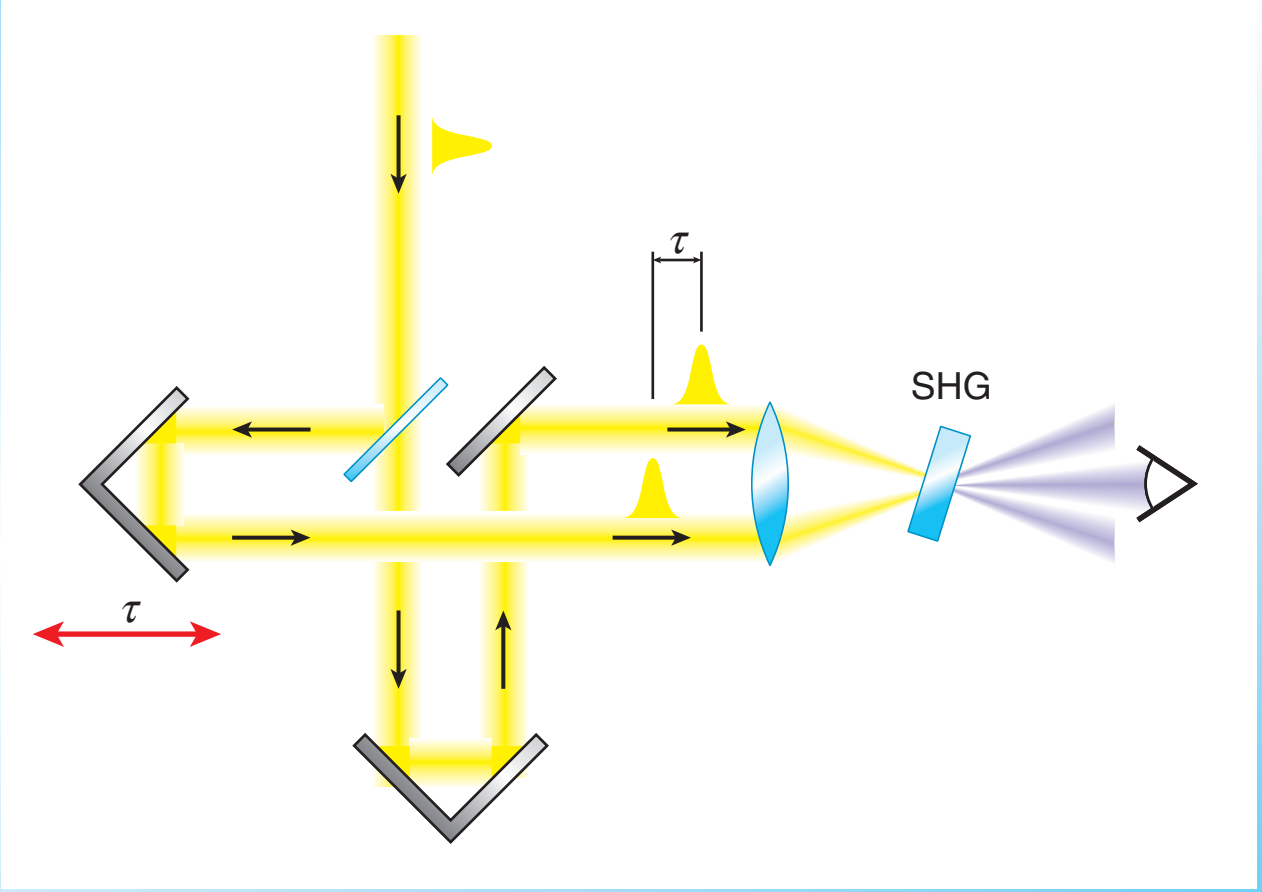

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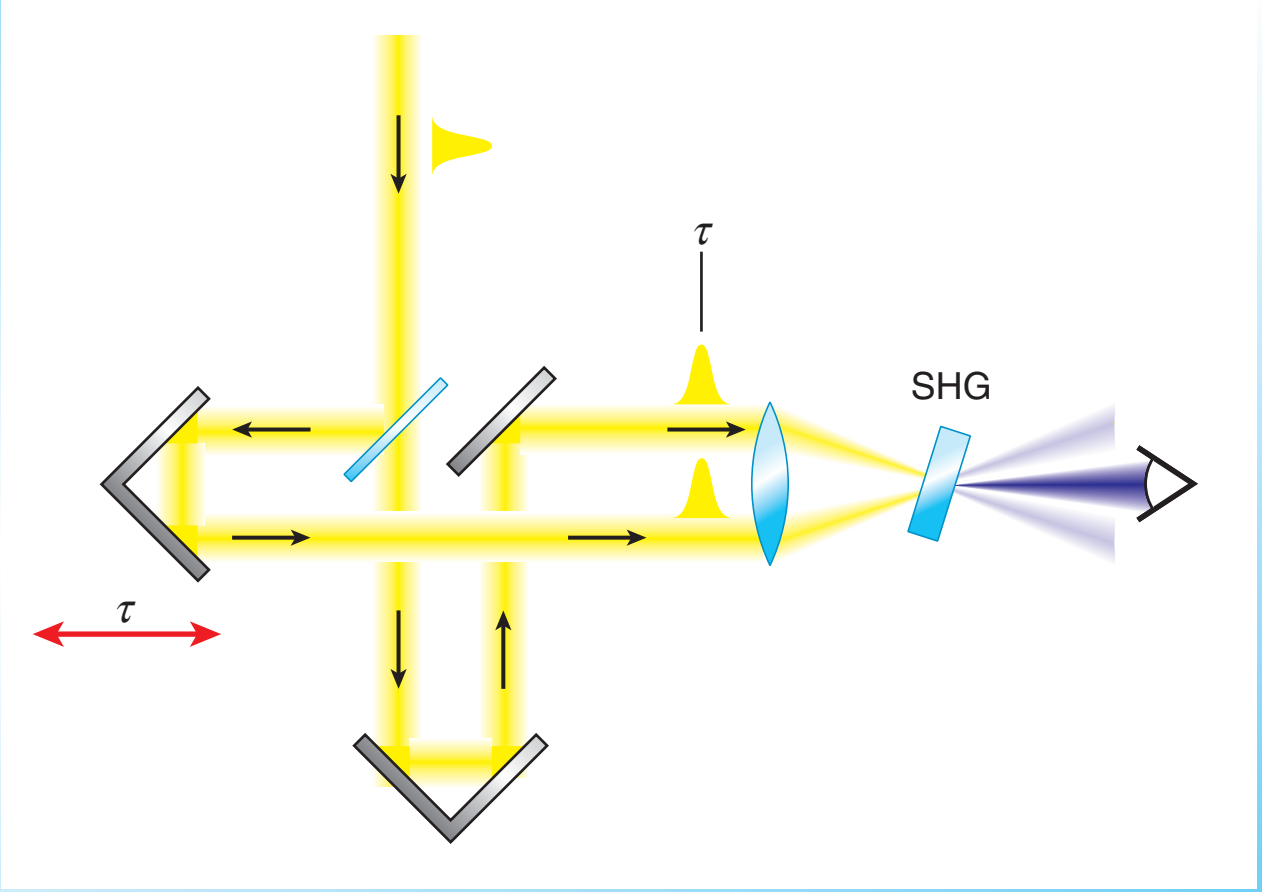

Temporal characterization

Electric field at SHG crystal

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

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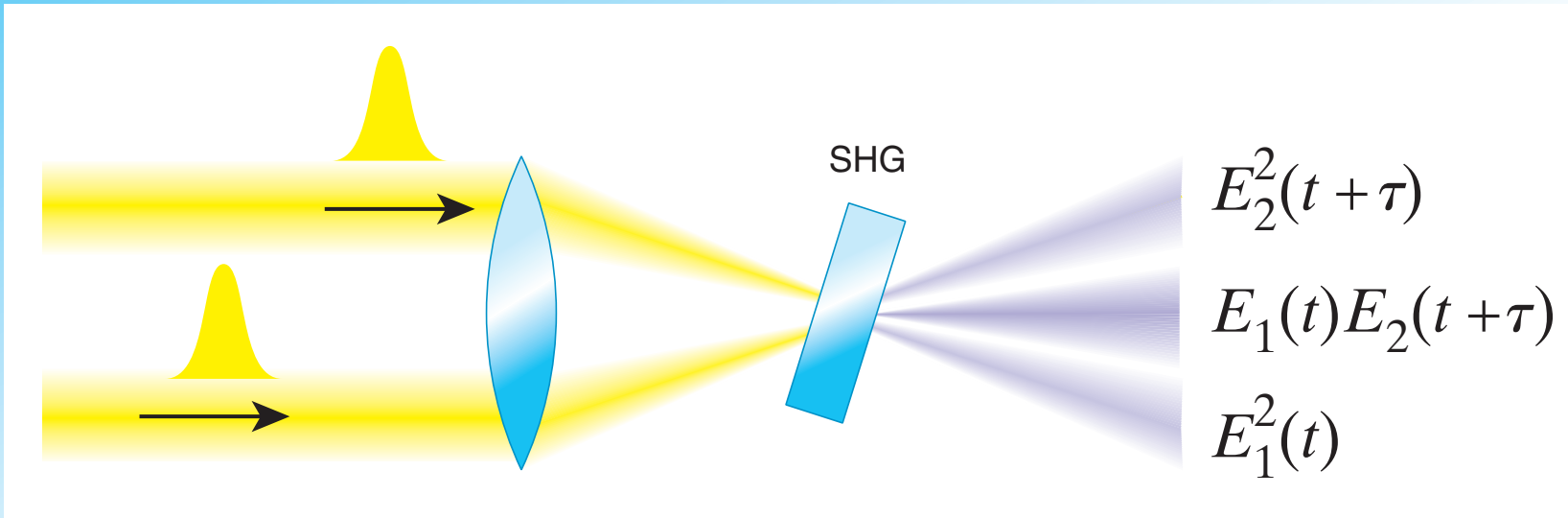
Temporal characterization



Second harmonic intensity

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detector selects middle term

Temporal characterization

Integrated detector signal yields intensity autocorrelation

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

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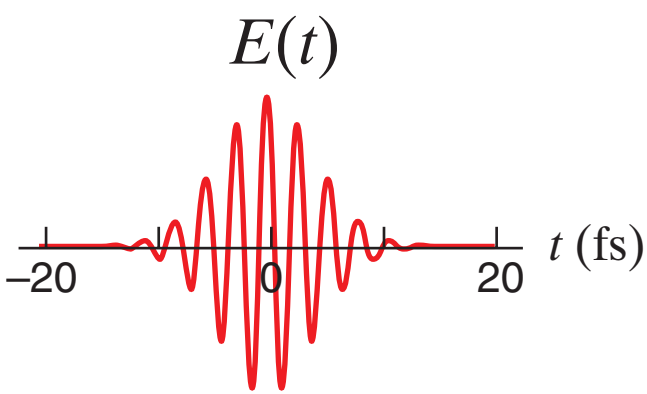

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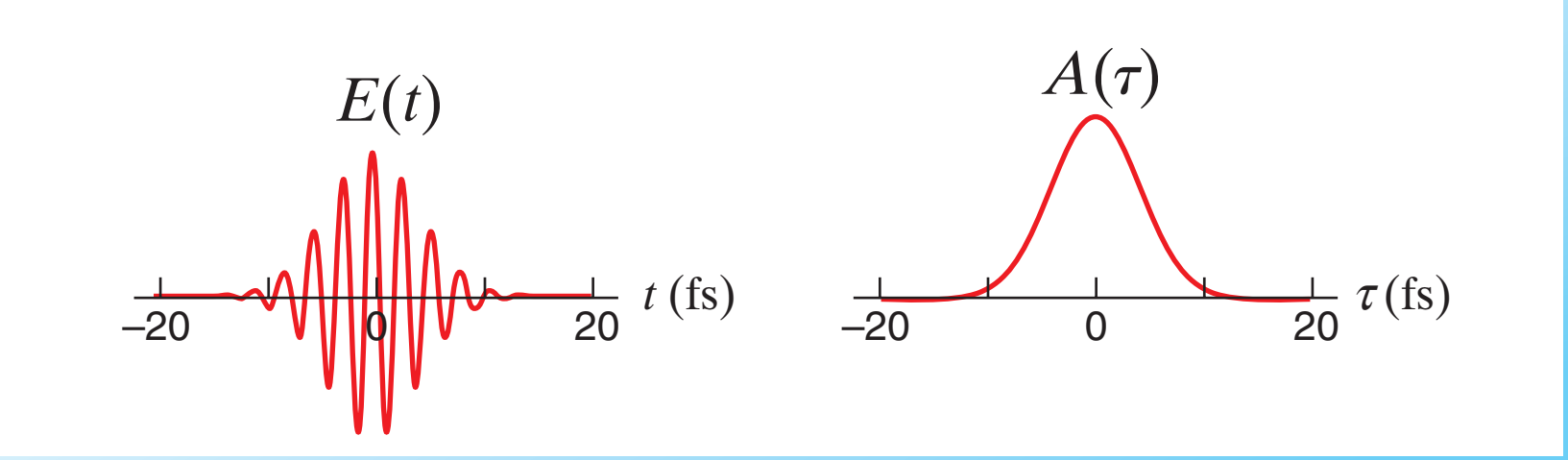
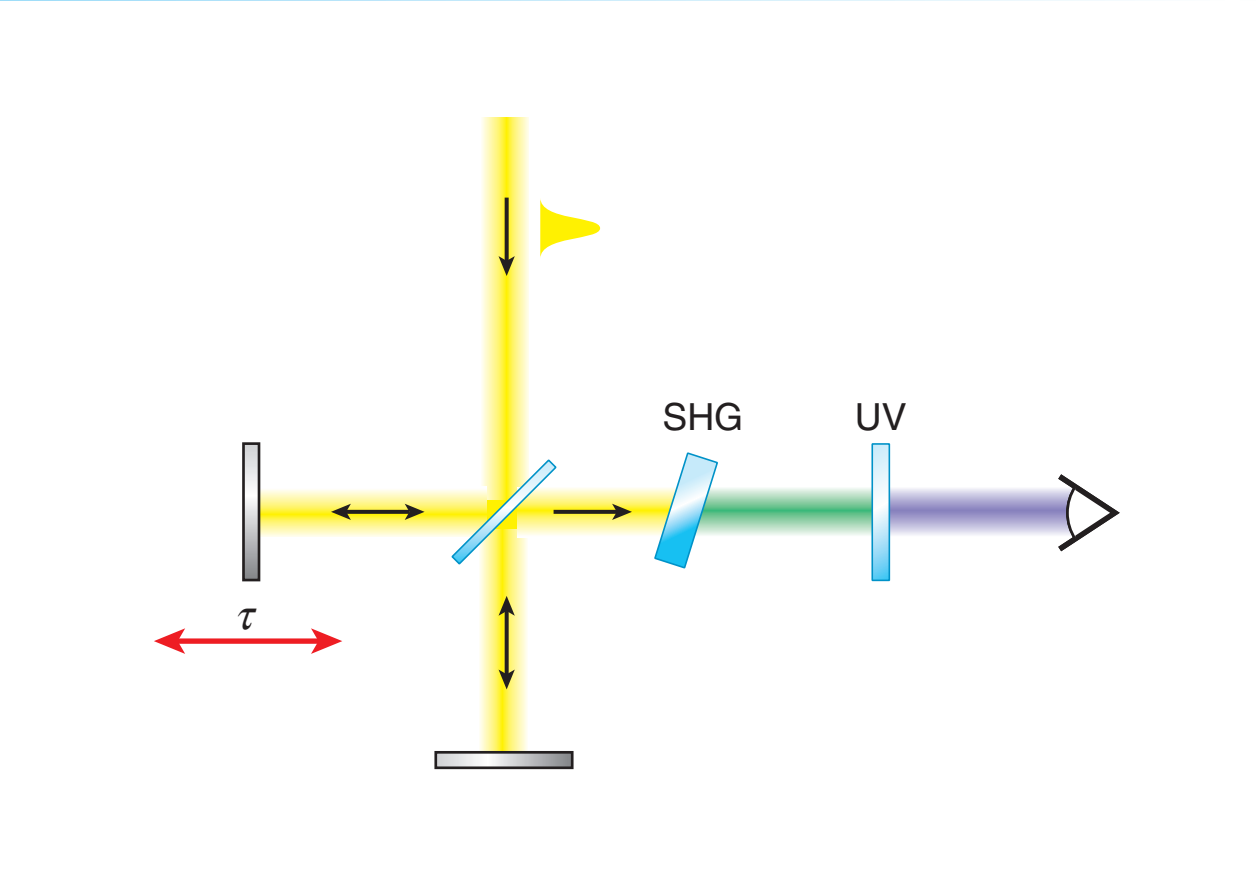


Temporal characterization

Alternative colinear geometry



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All terms now contribute:

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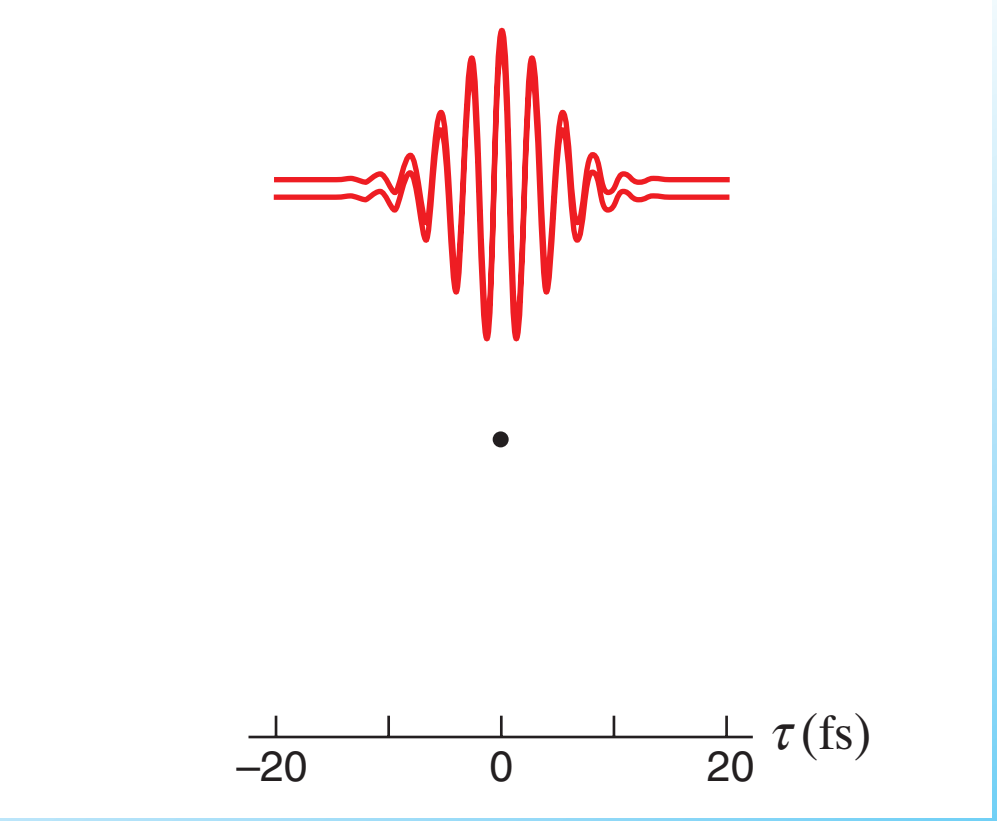

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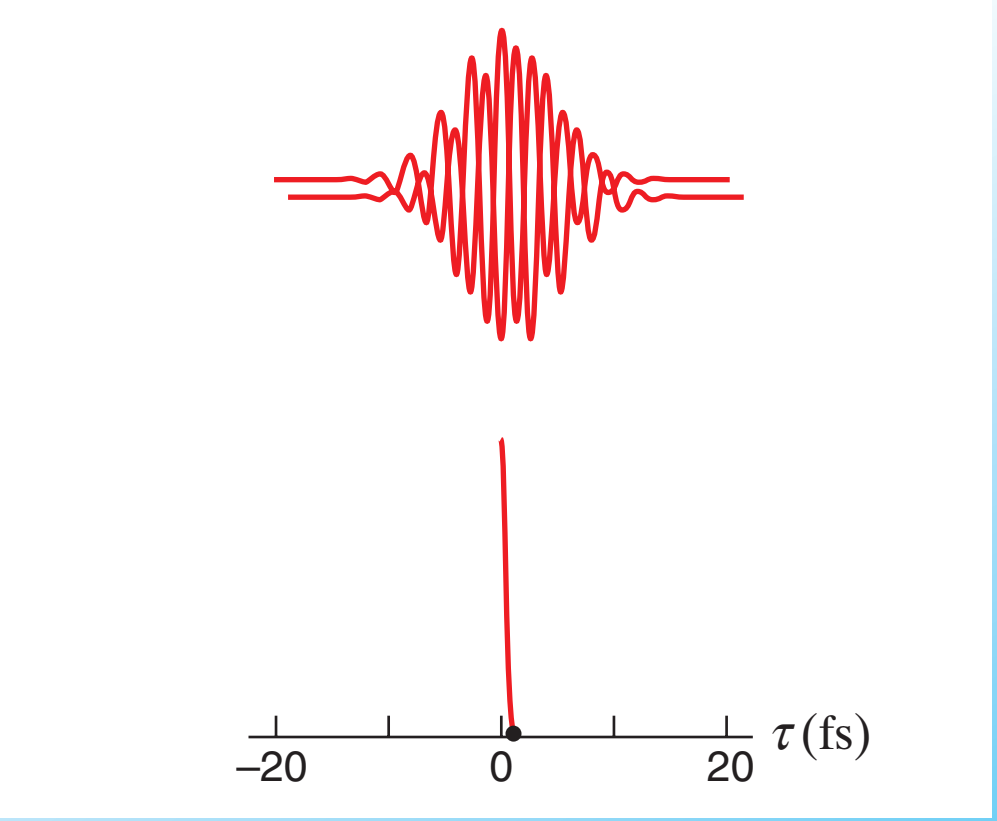

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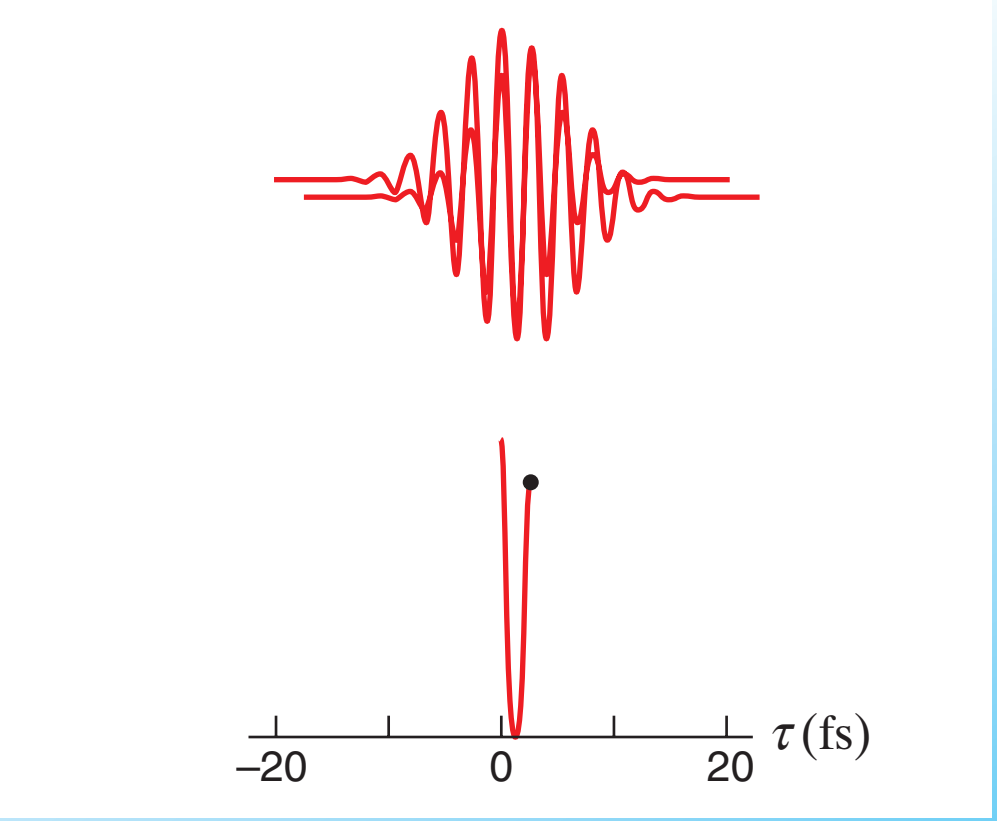

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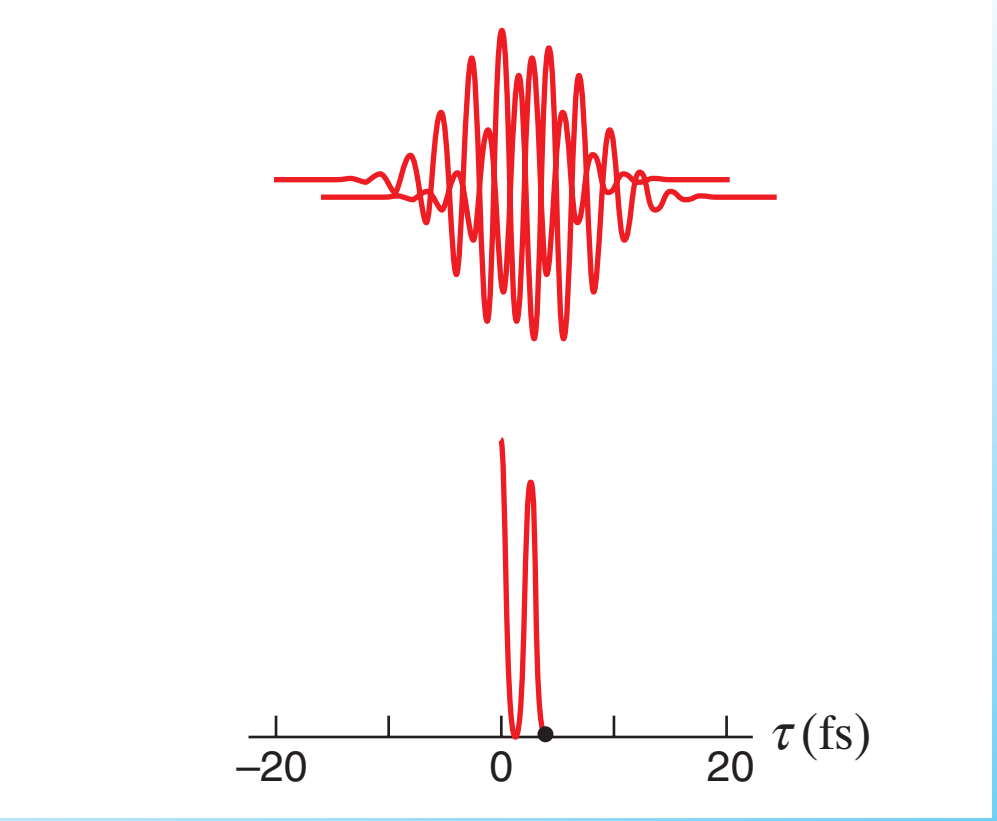

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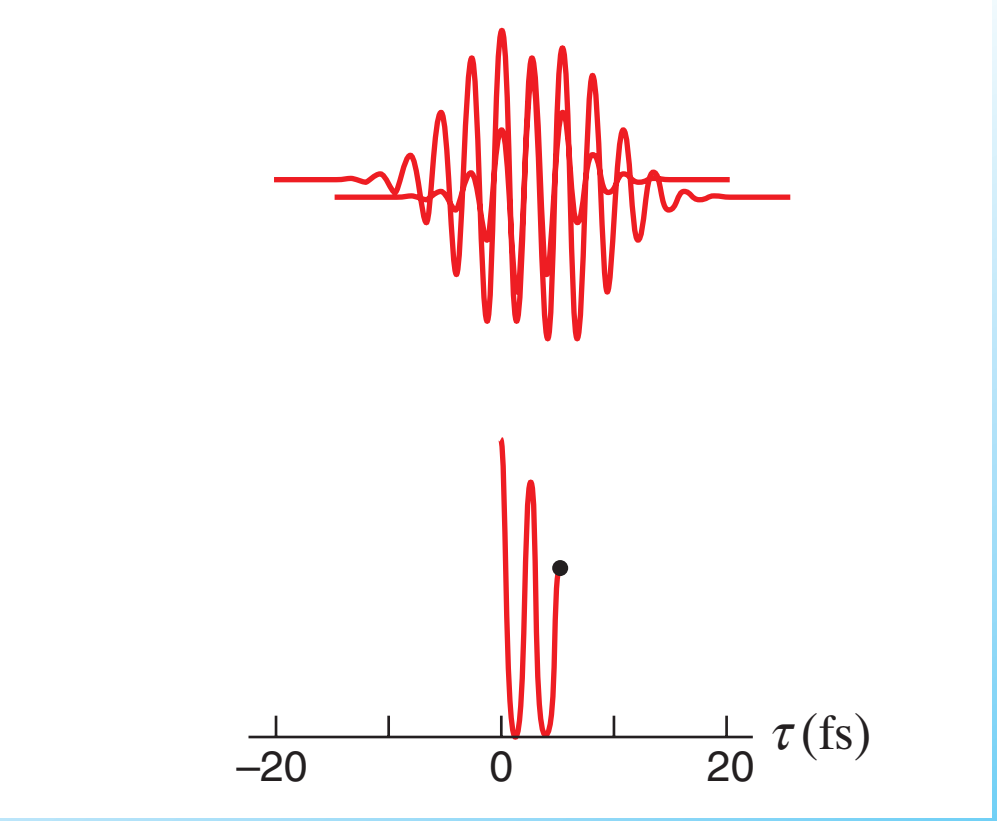

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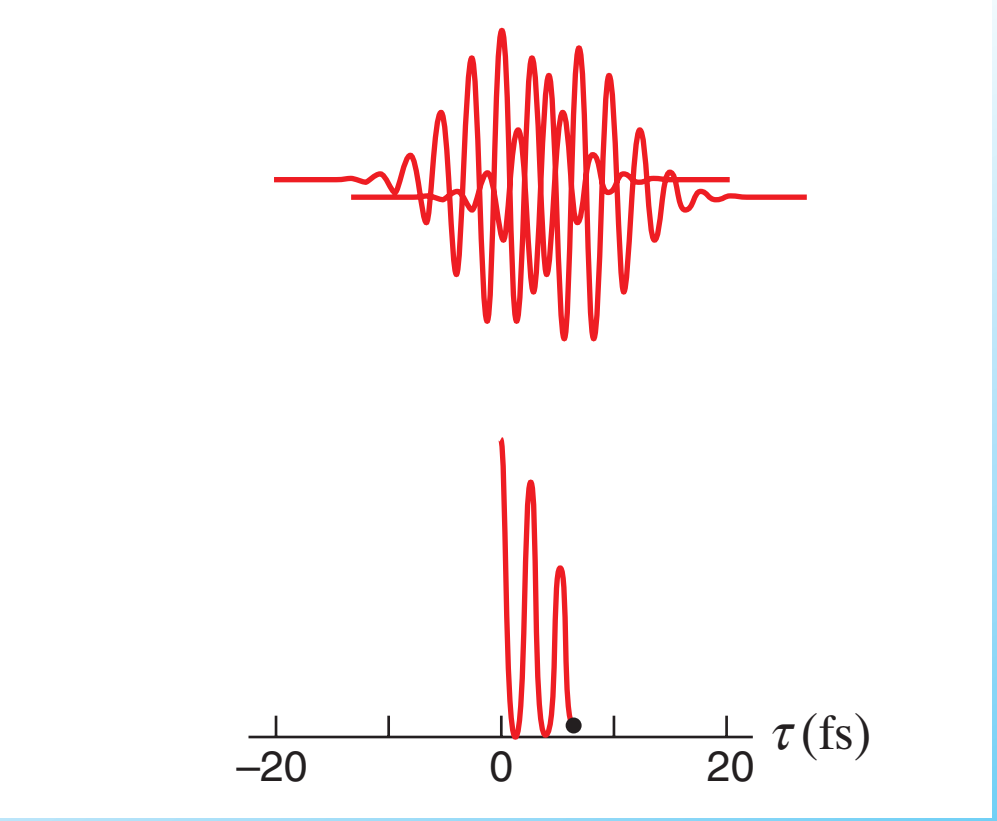

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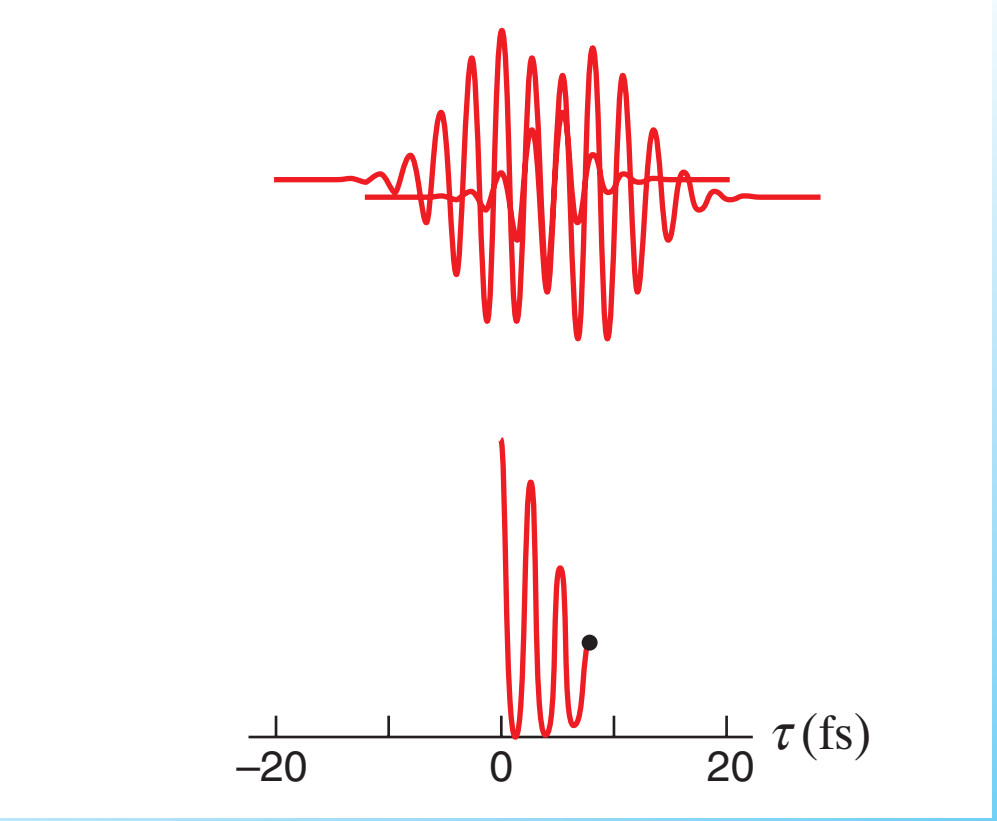

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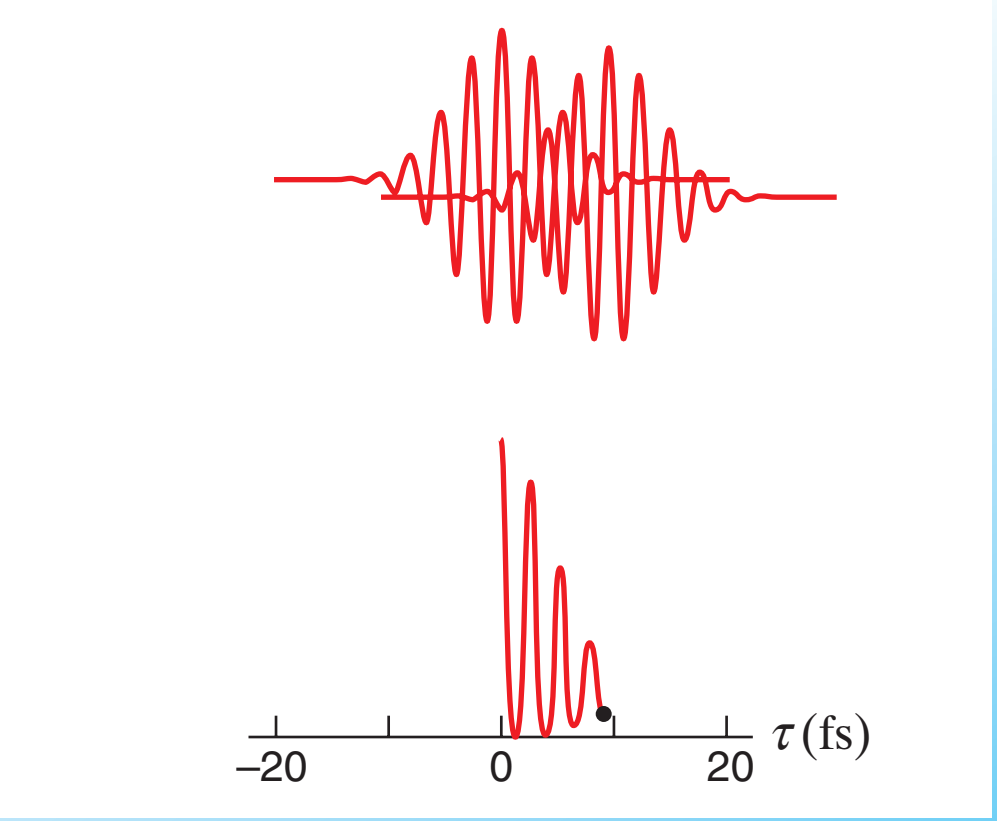

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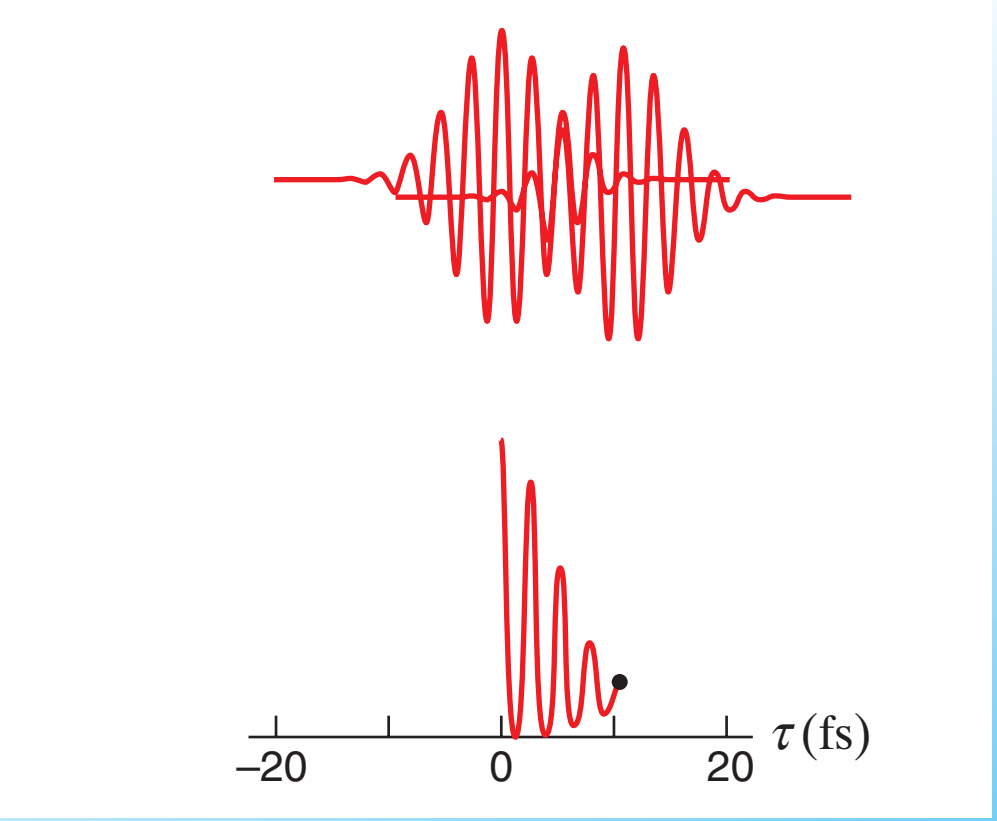

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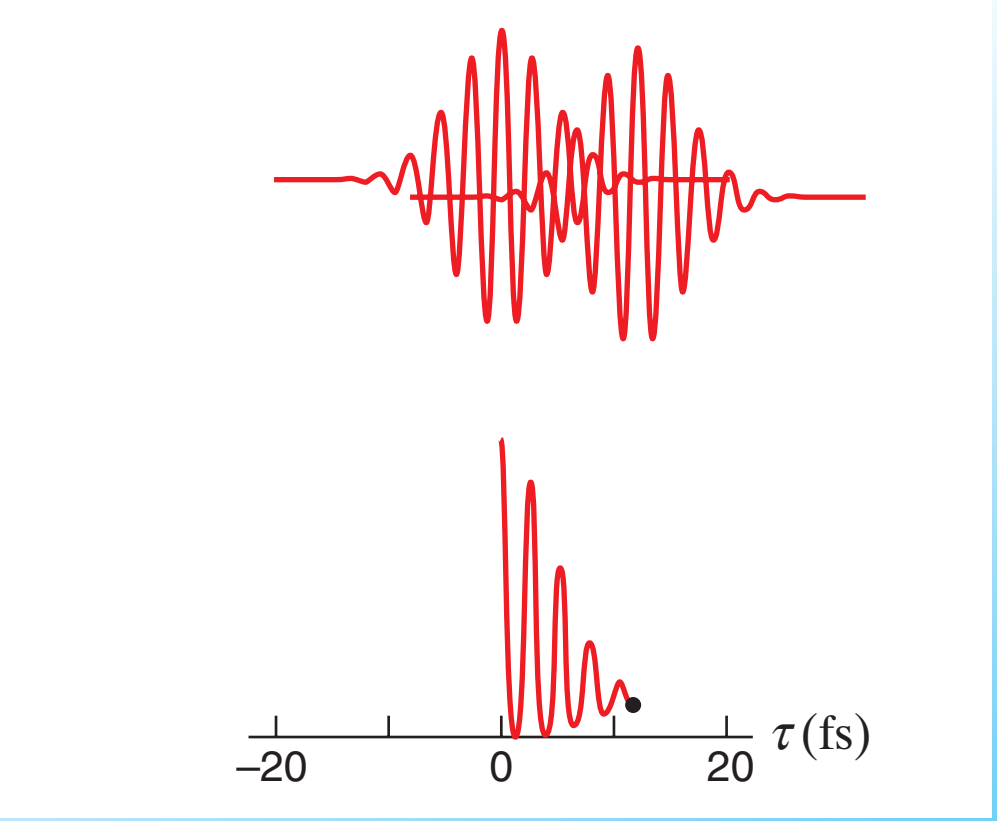

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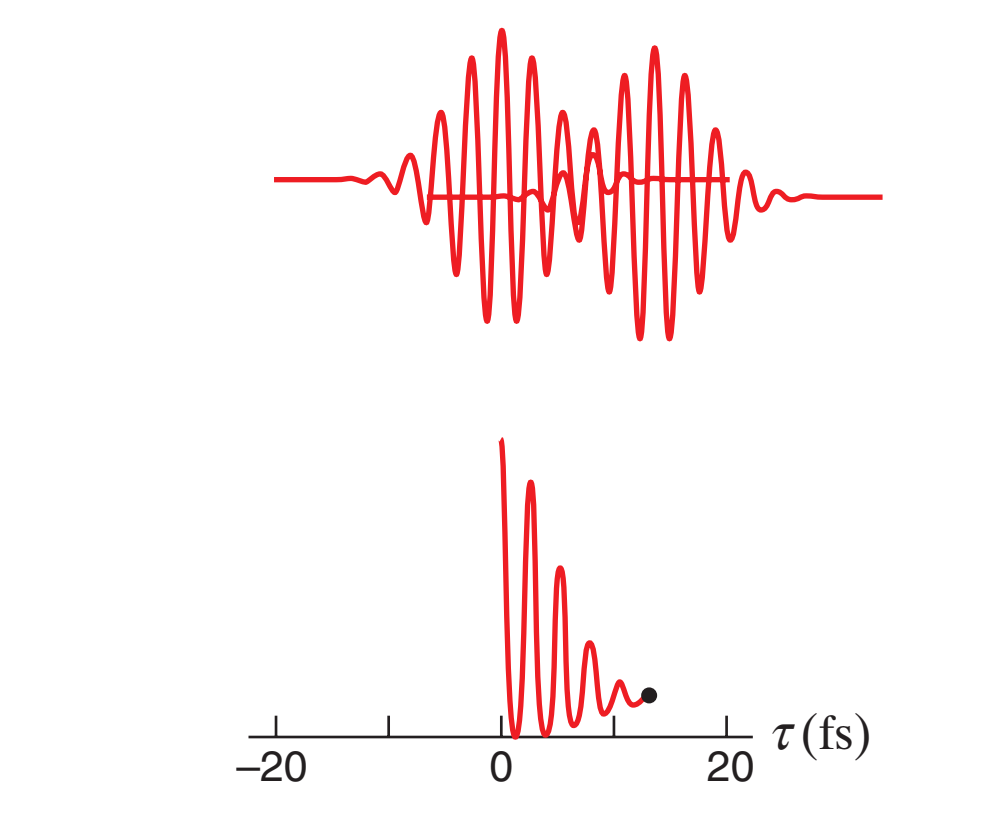

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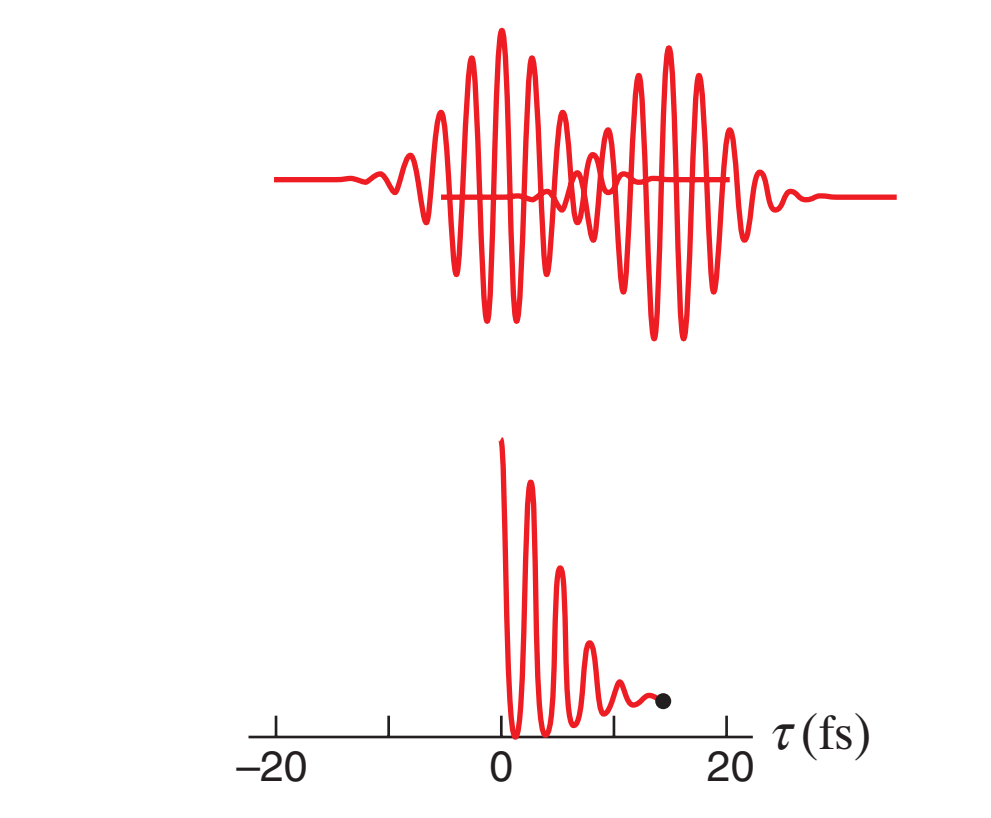

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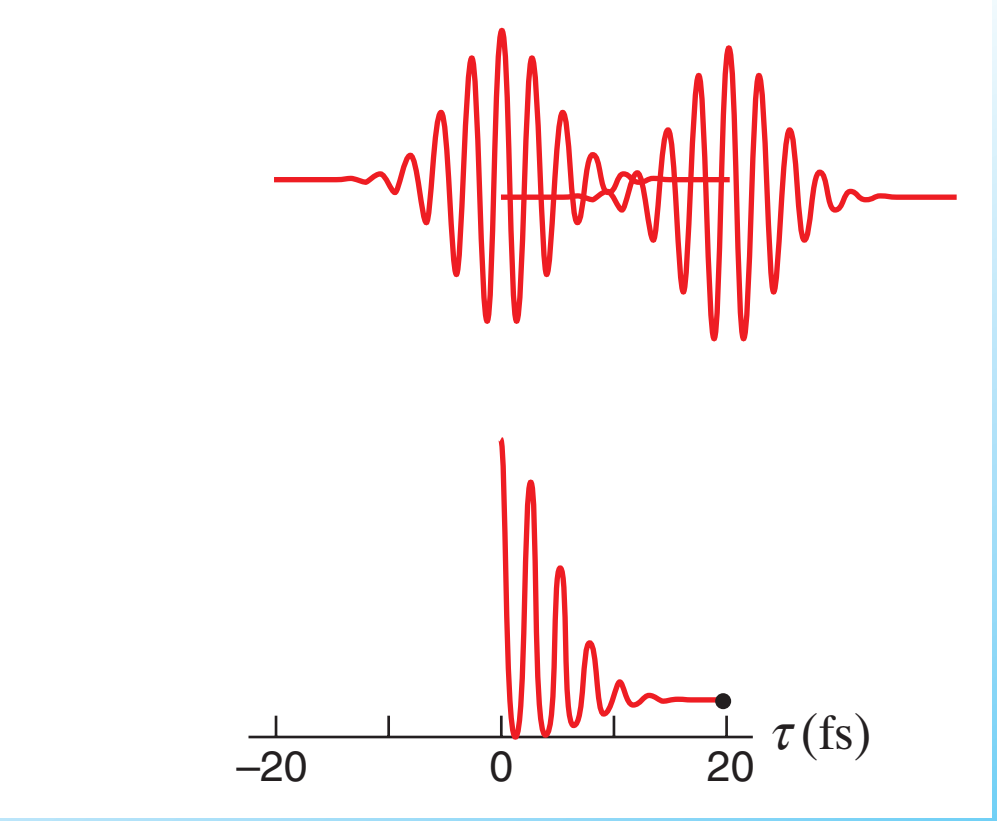

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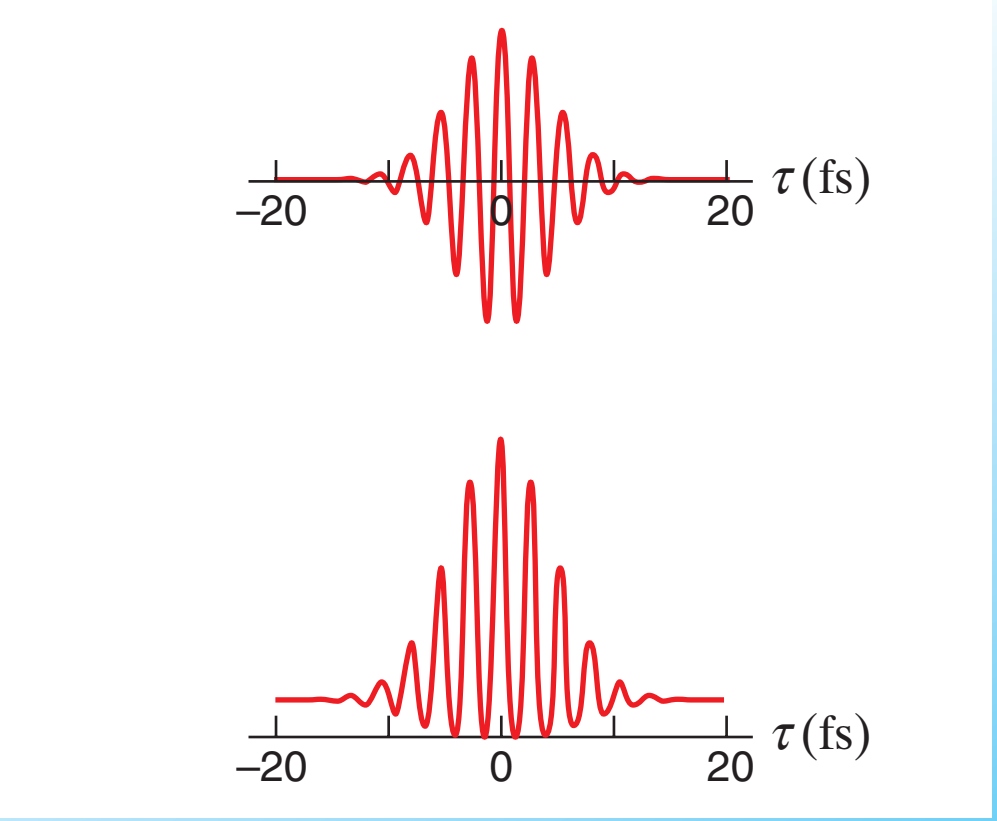
$$I_{2\omega}(t,\tau) \propto \left|\chi^{(2)}\right|^2 \left|E_{tot}^2\right|^2 = \left|\chi^{(2)}\right|^2 \left|E_1^2(t) + 2E_1(t)E_2(t+\tau) + E_2^2(t+\tau)\right|^2$$



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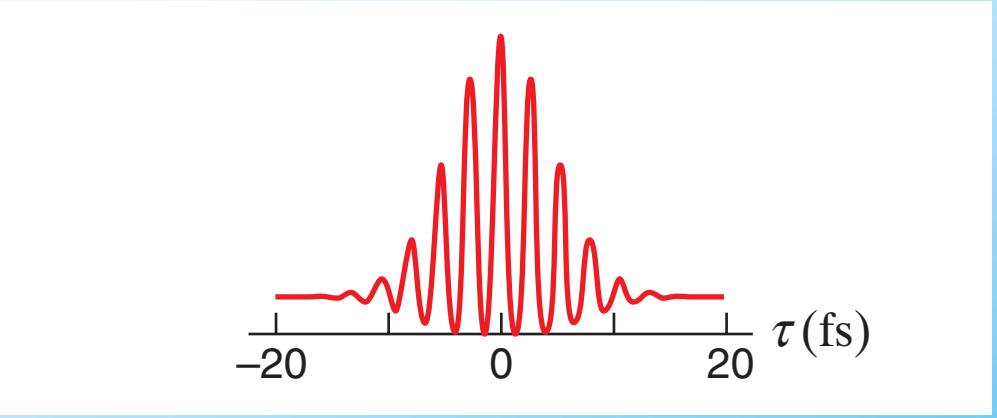
at $\tau = 0$**:**

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



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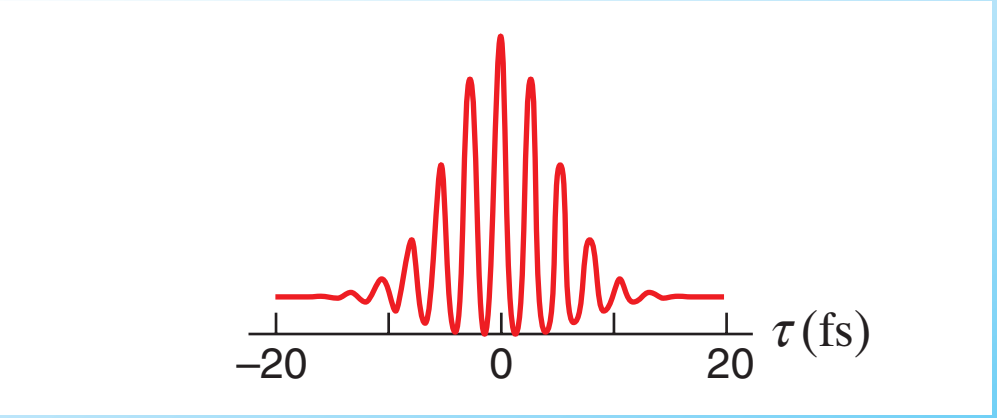
as $\tau \to \pm\infty$**:** $$I_{2\omega}(t,\tau) \propto 2E^4(t)$$



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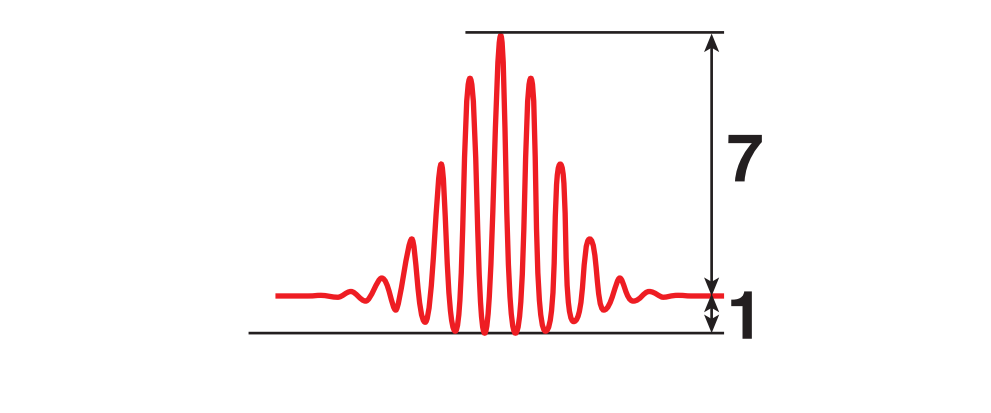
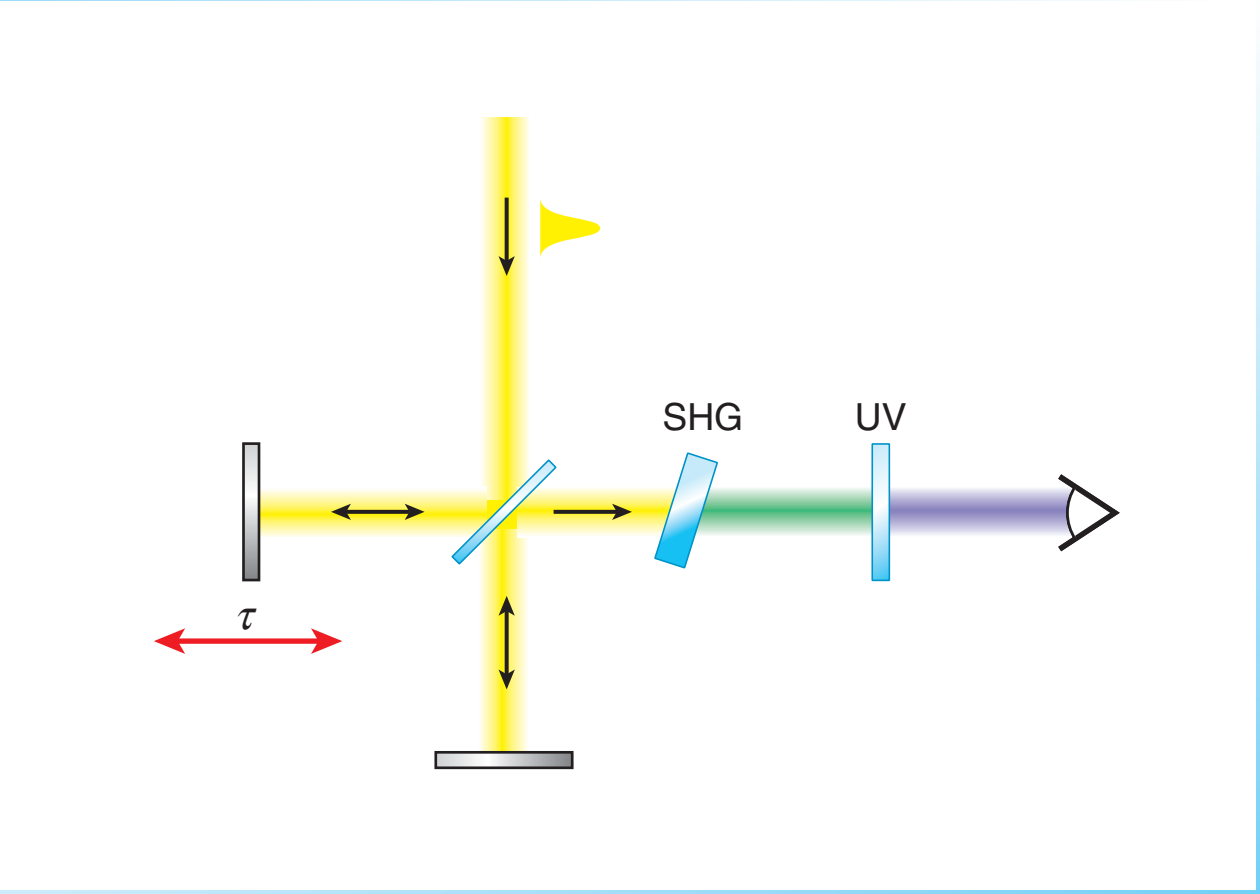


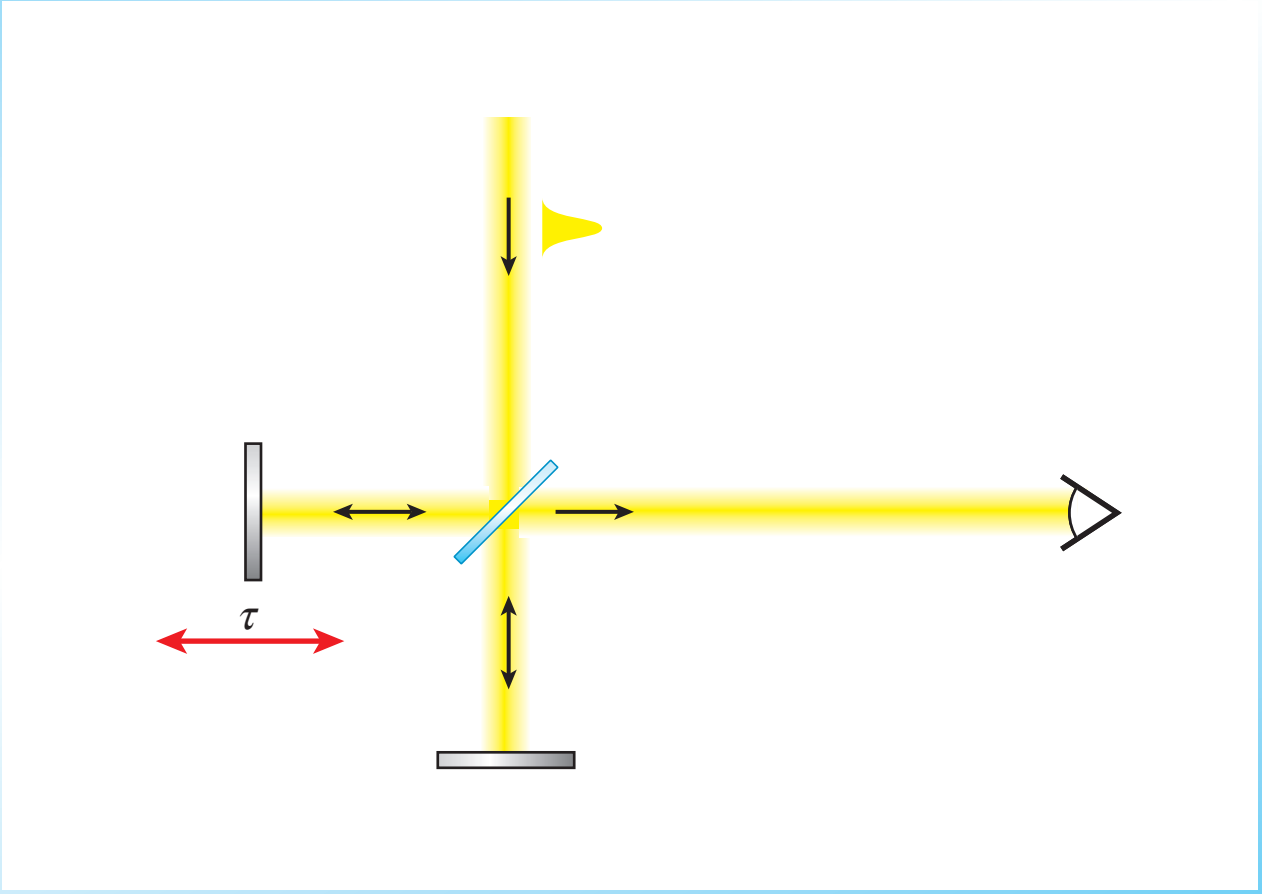
Temporal characterization

Do we really need the second-harmonic crystal…?



Temporal characterization

Would this work?



Temporal characterization

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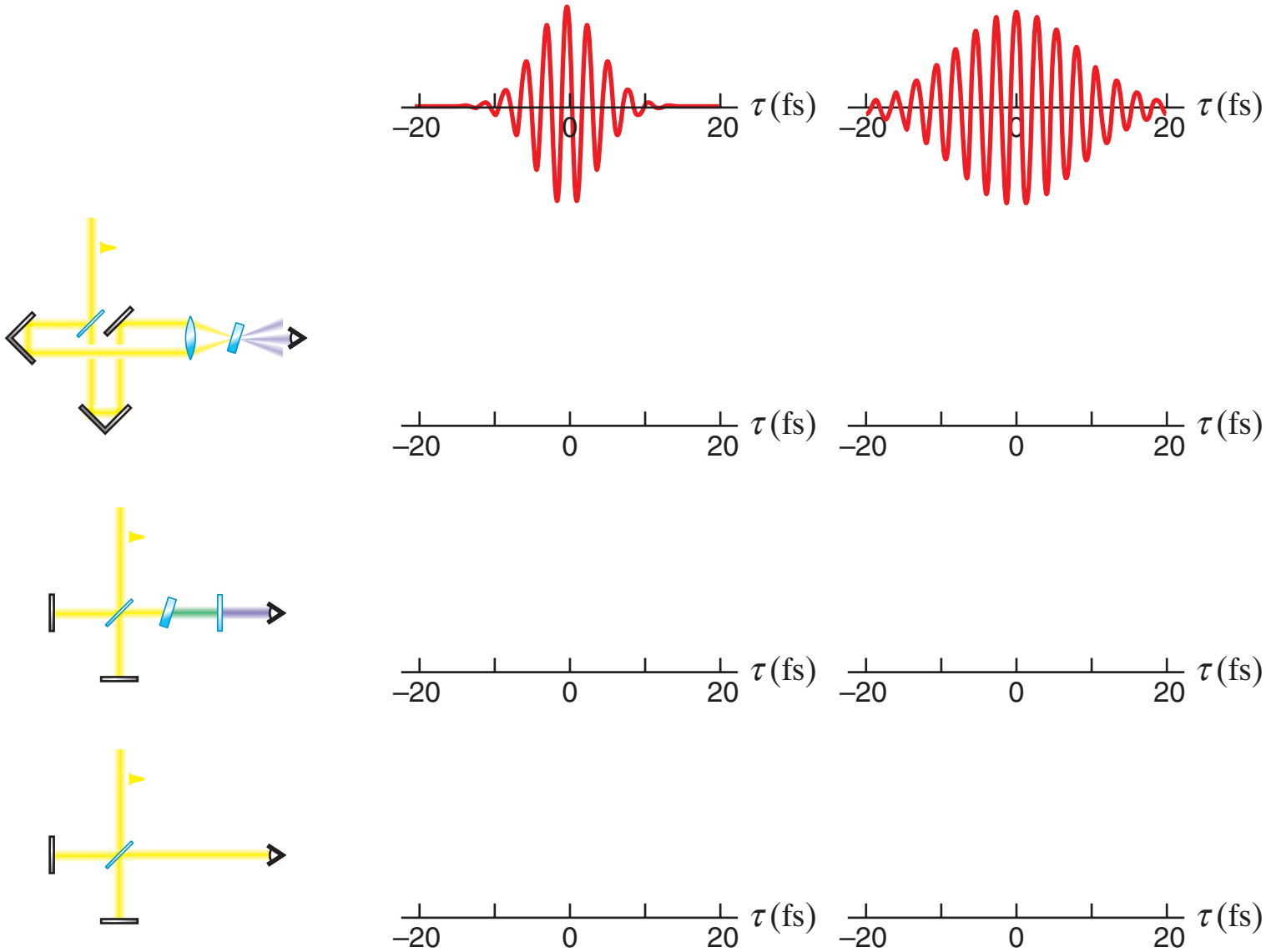
so

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

Temporal characterization



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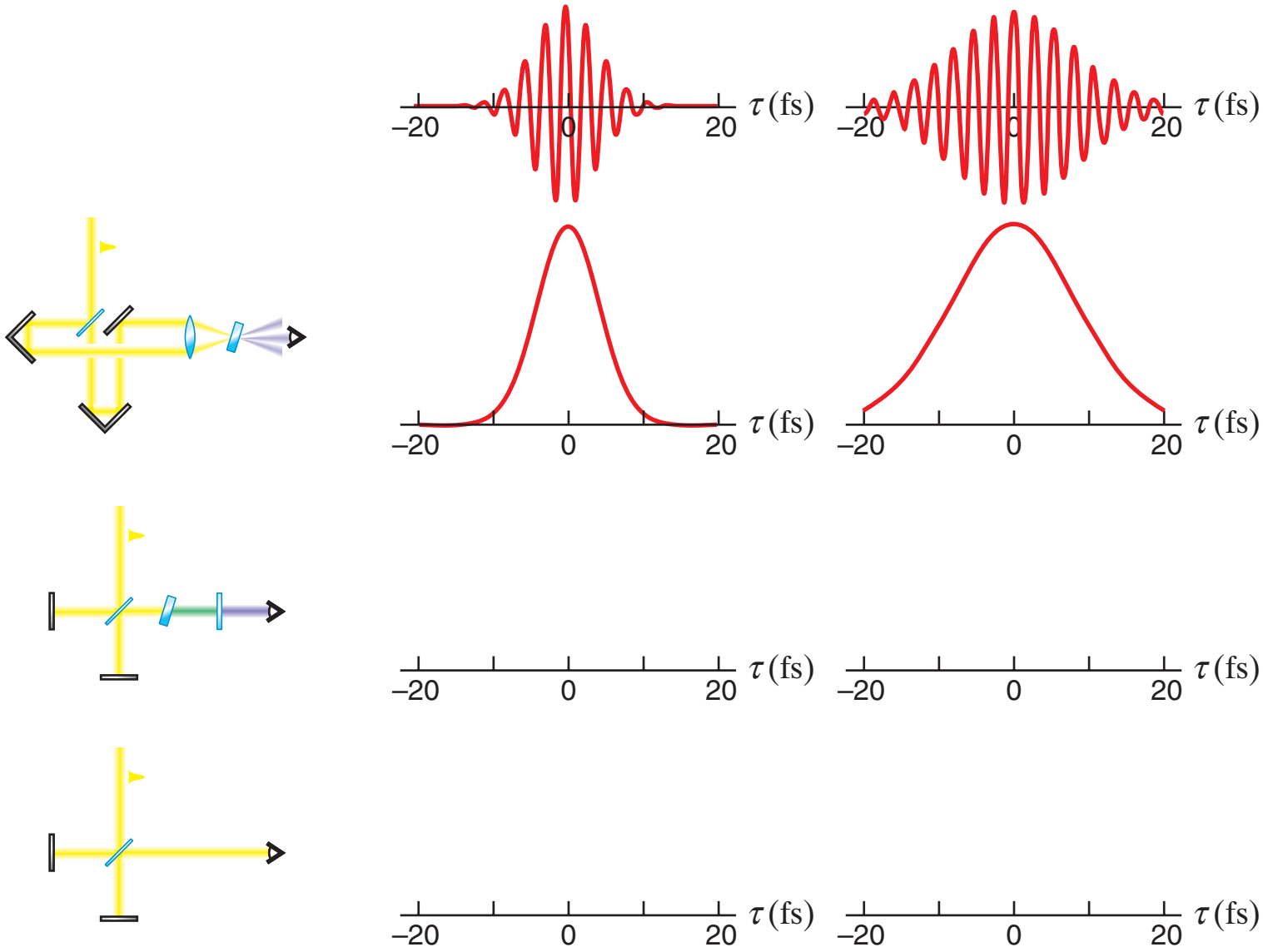

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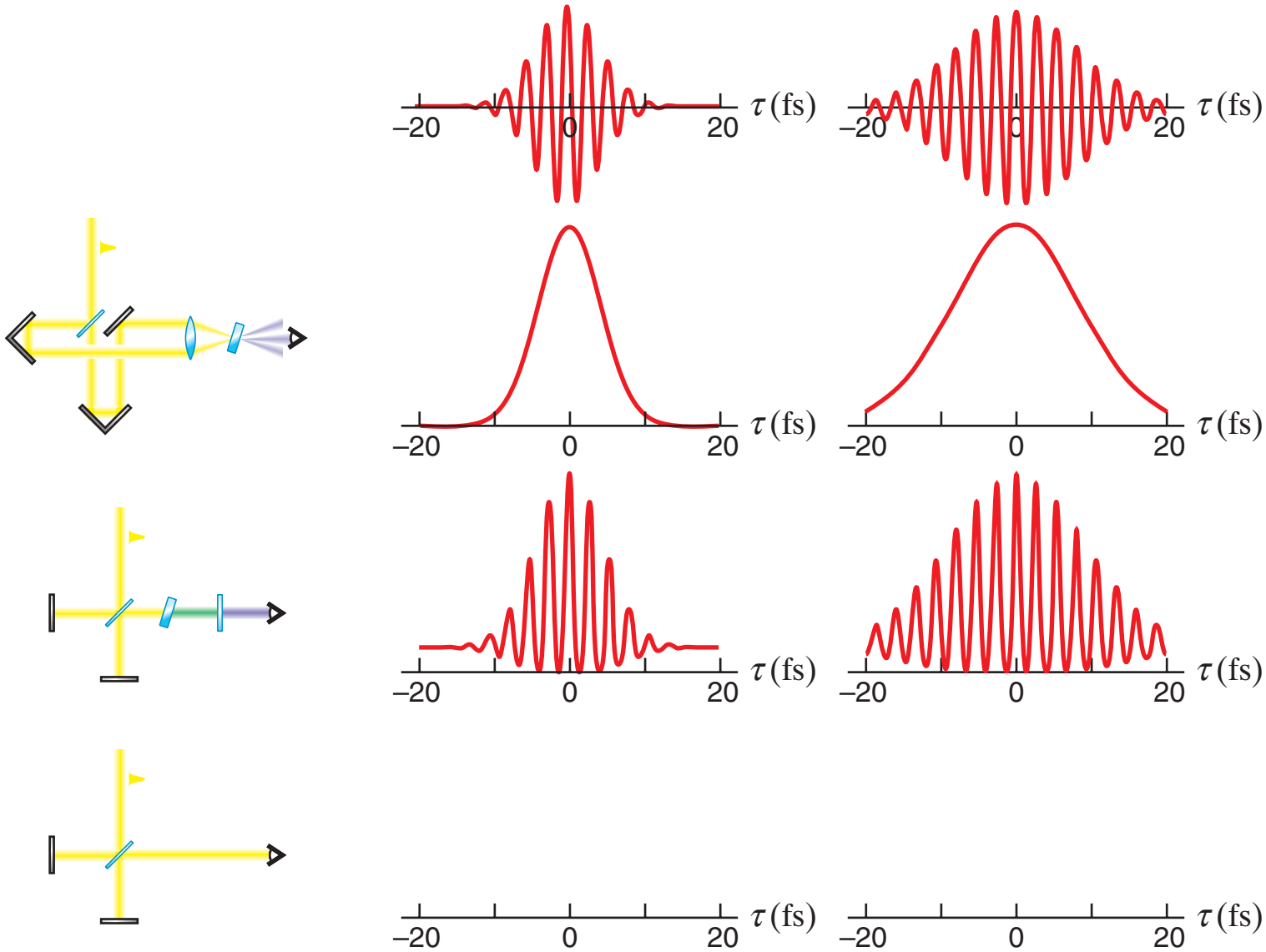

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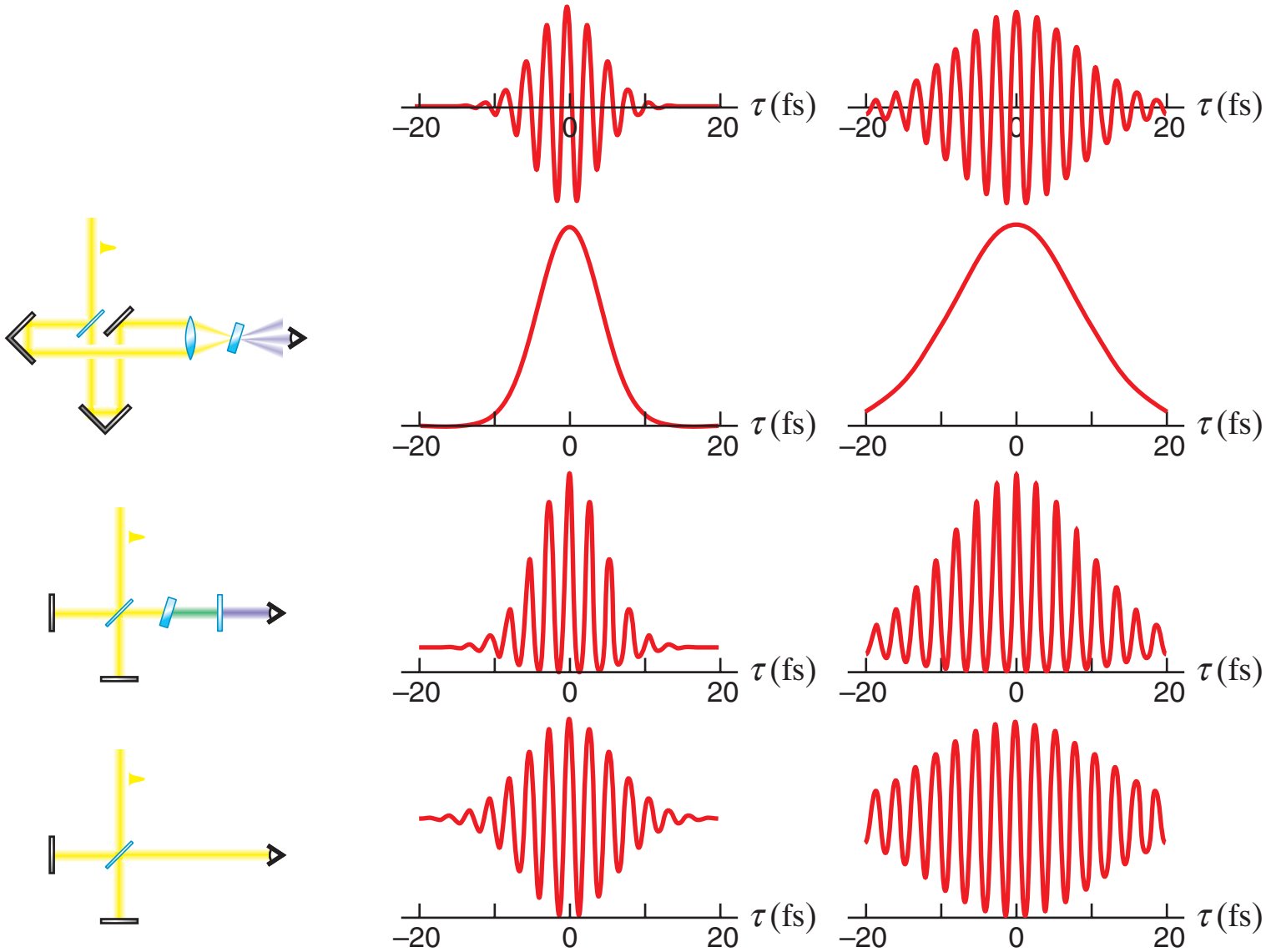
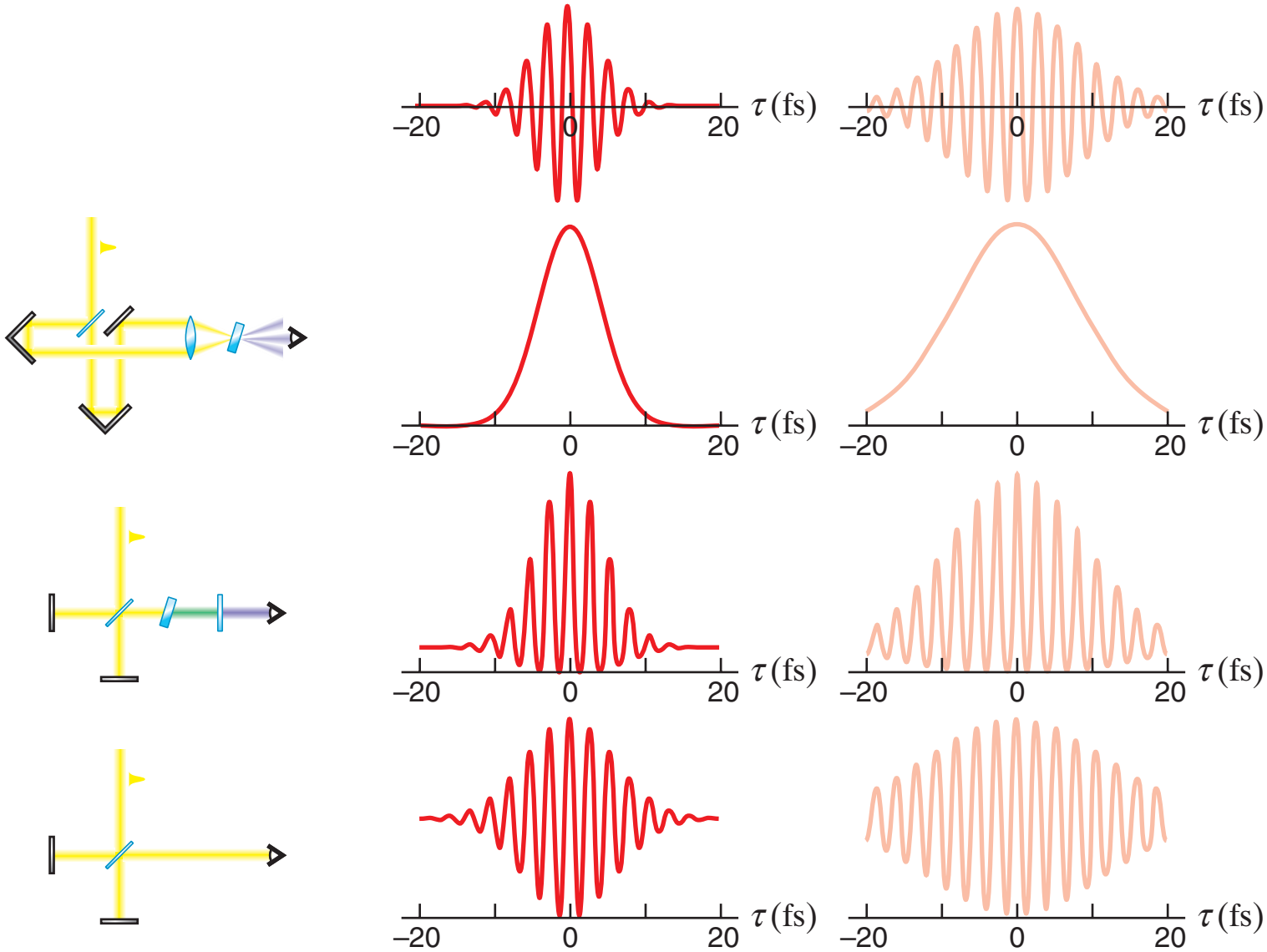


Temporal characterization

But what about dispersion?

Temporal characterization



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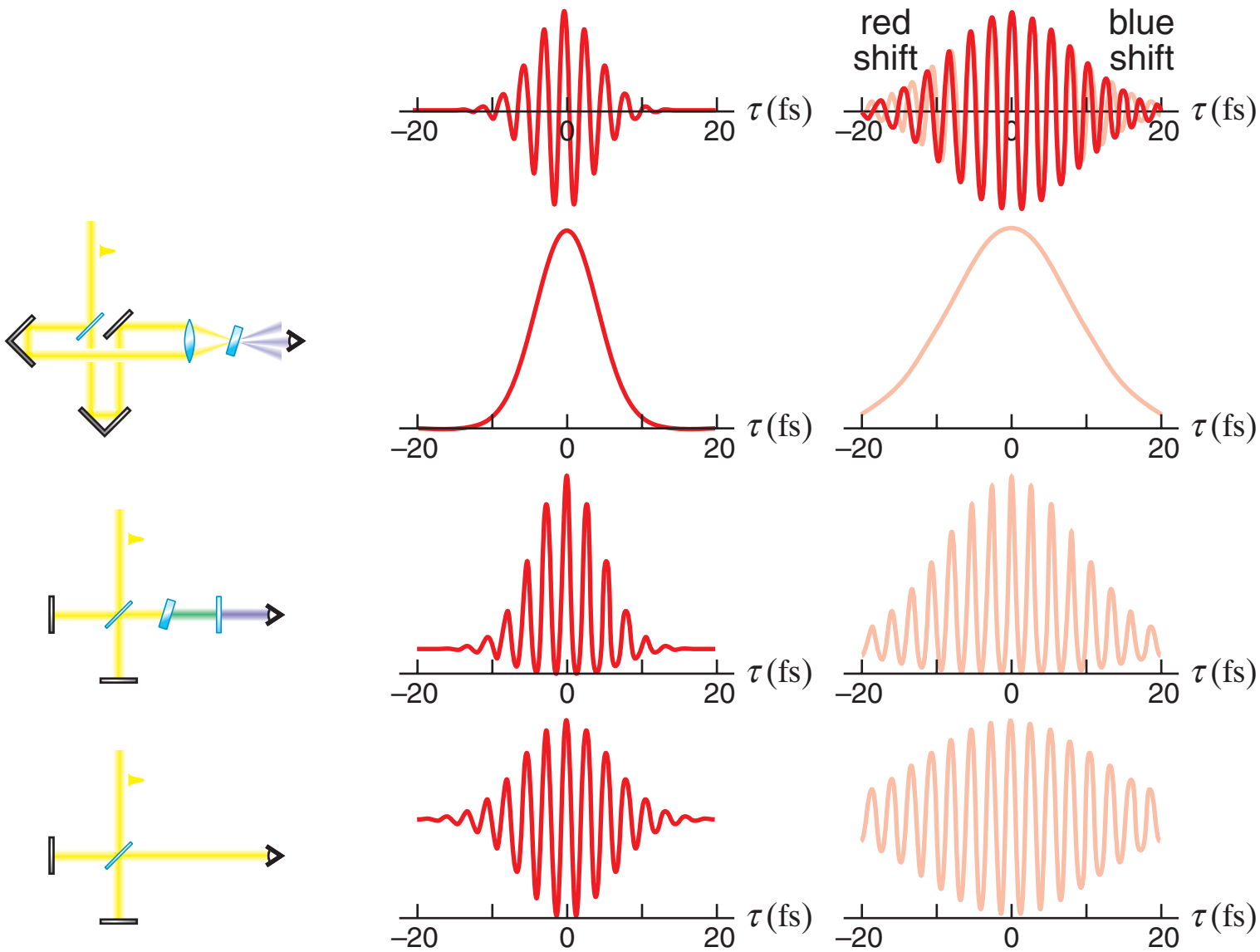

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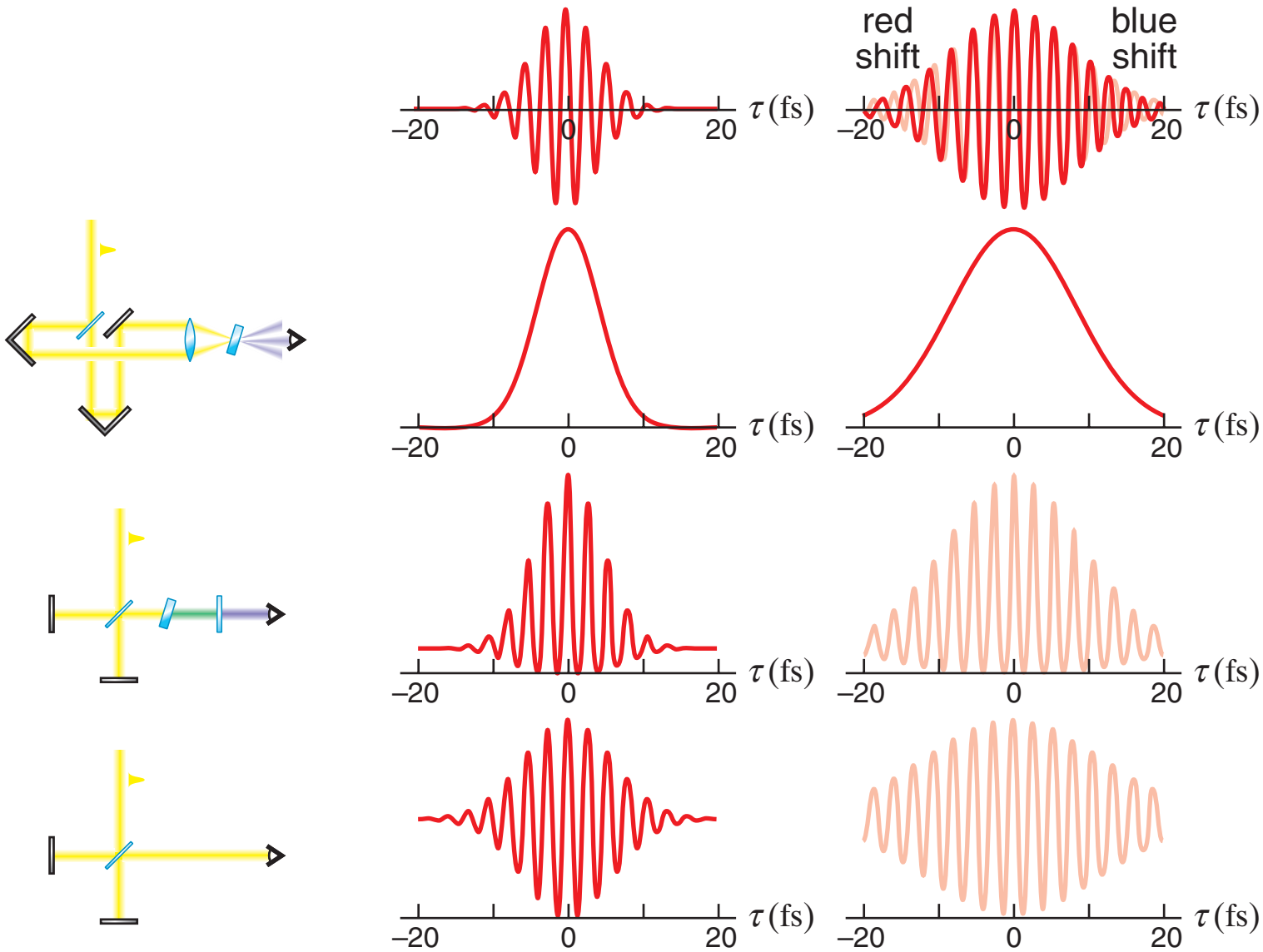

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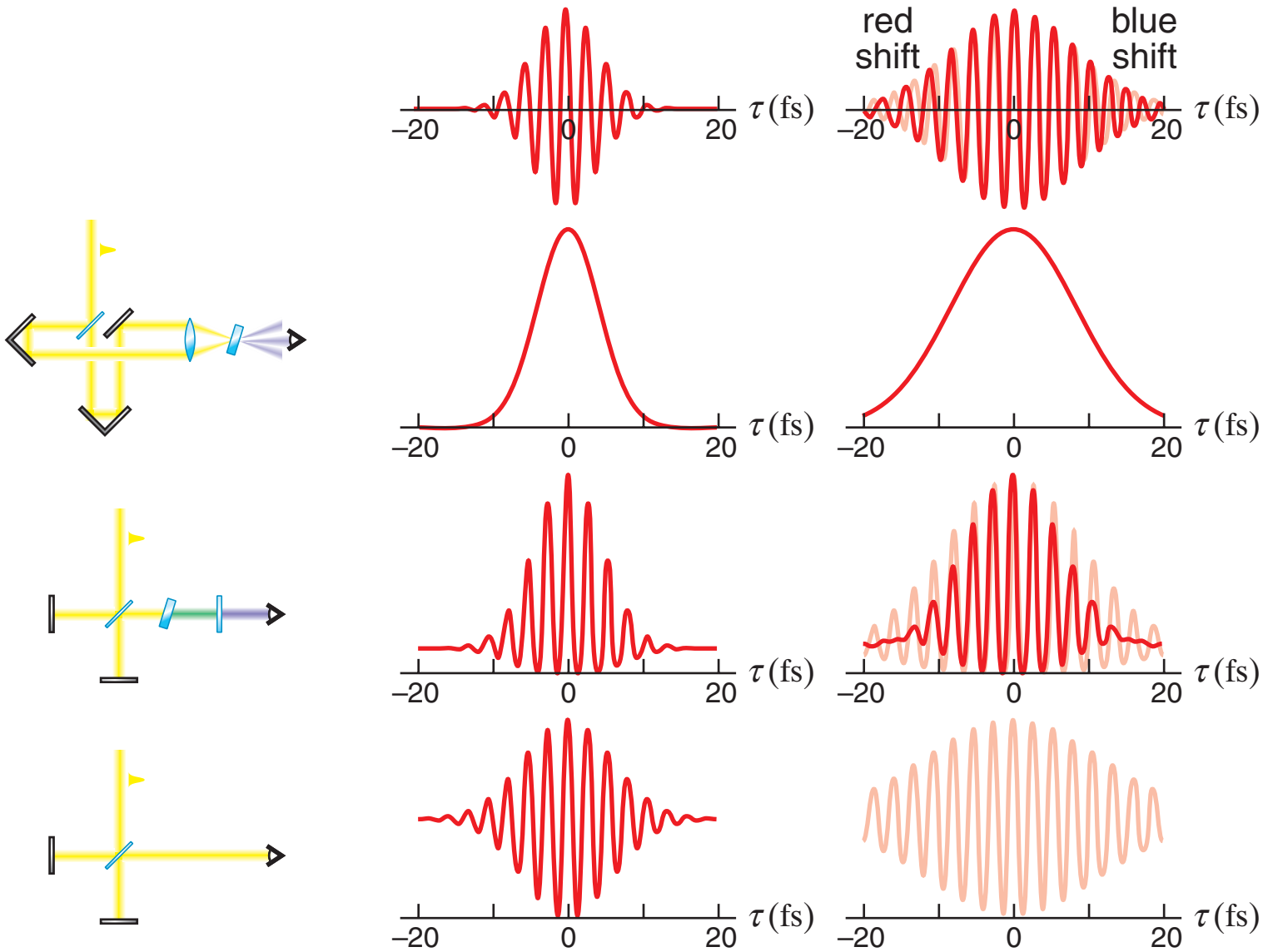

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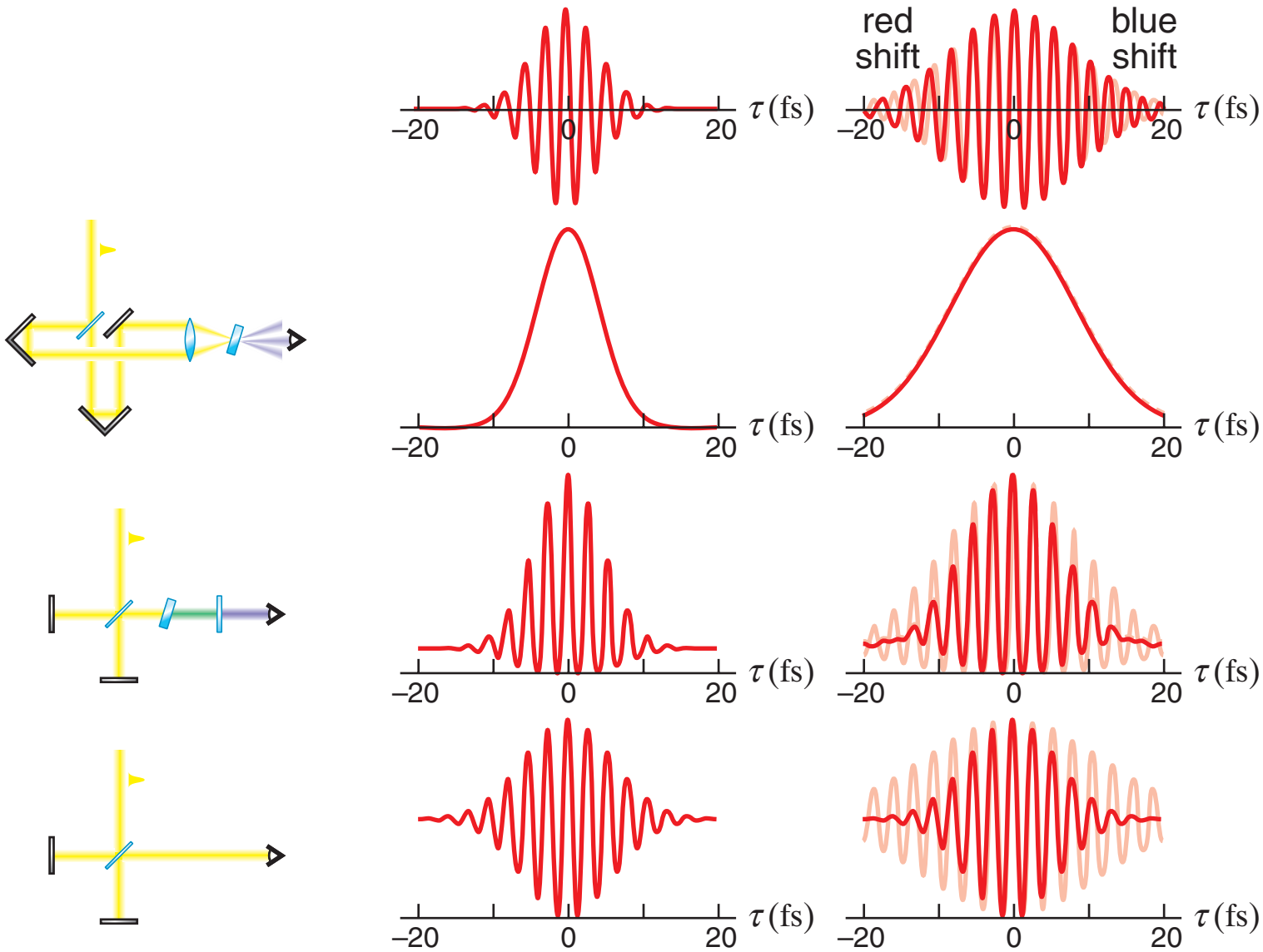

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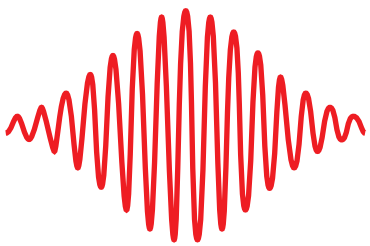

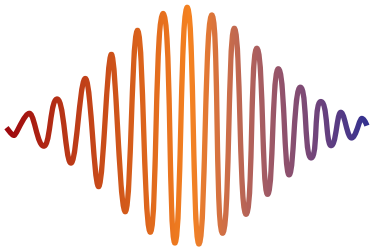

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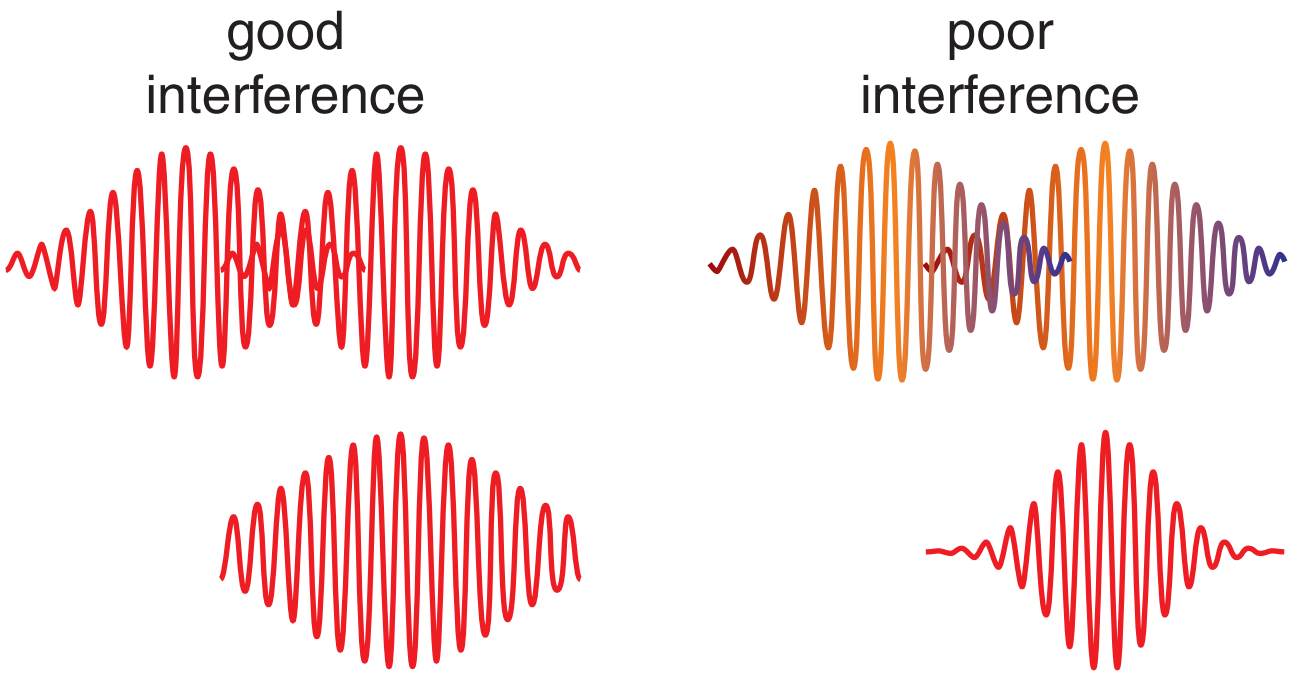

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Let $E_{disp}(\omega) = E_{orig}(\omega)e^{-i\phi(\omega)}$.

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Interference term in linear autocorrelation:

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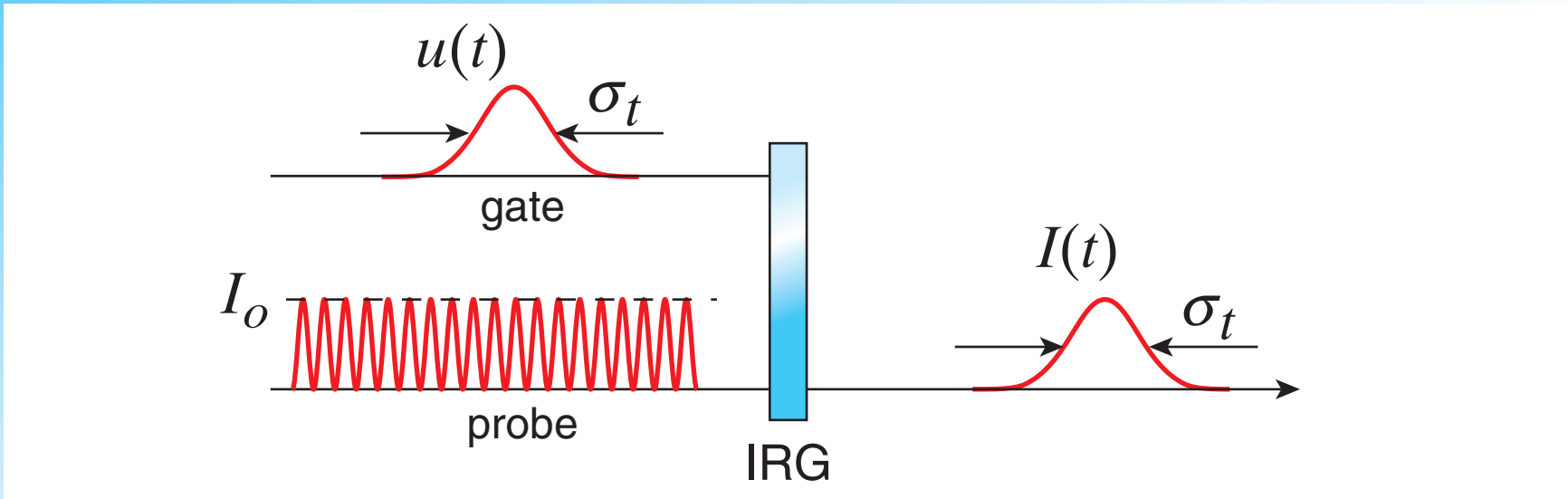
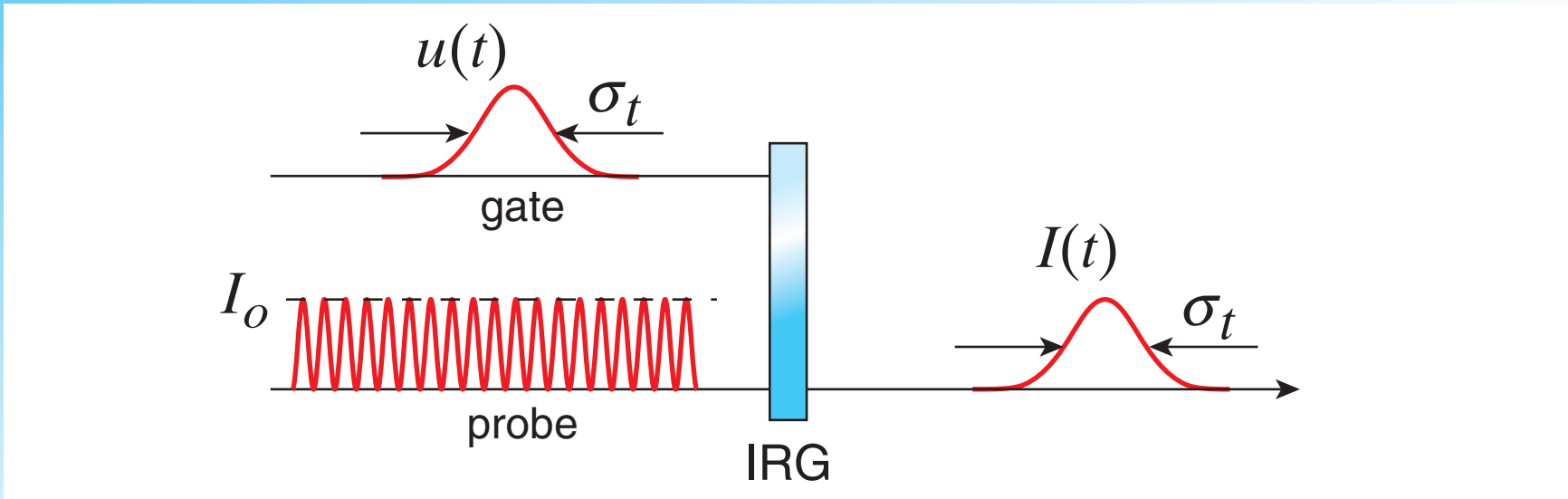
Joint time-frequency measurements



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

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

Joint time-frequency measurements



Transmitted intensity

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

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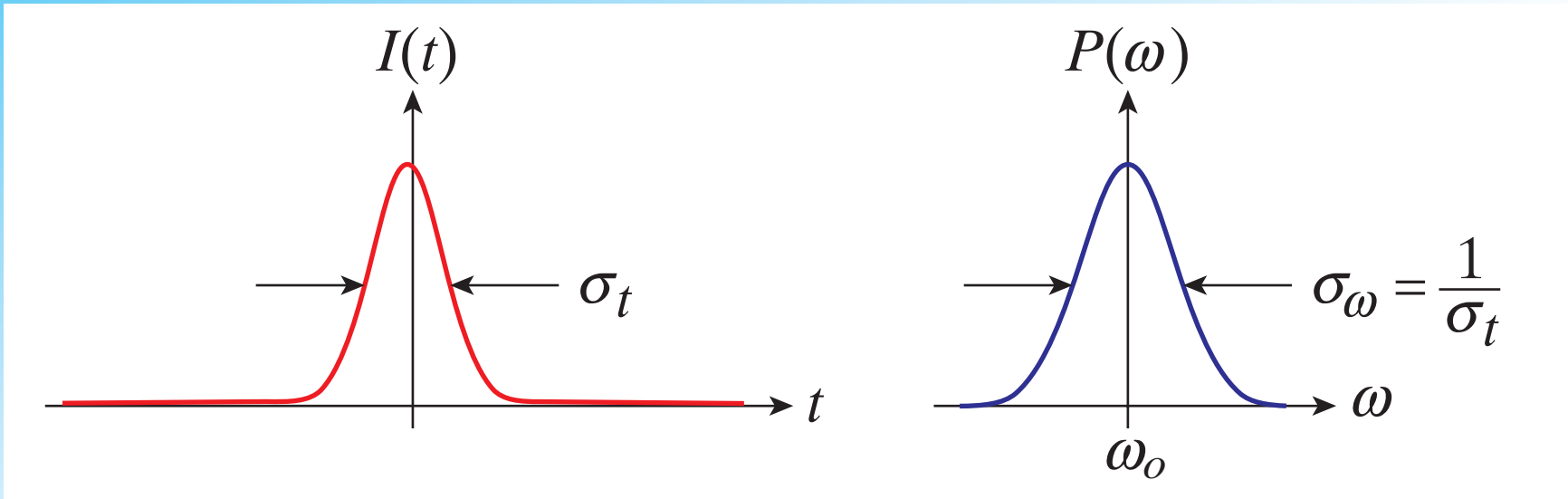

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$$\sigma_t \sigma_\omega = 1$$

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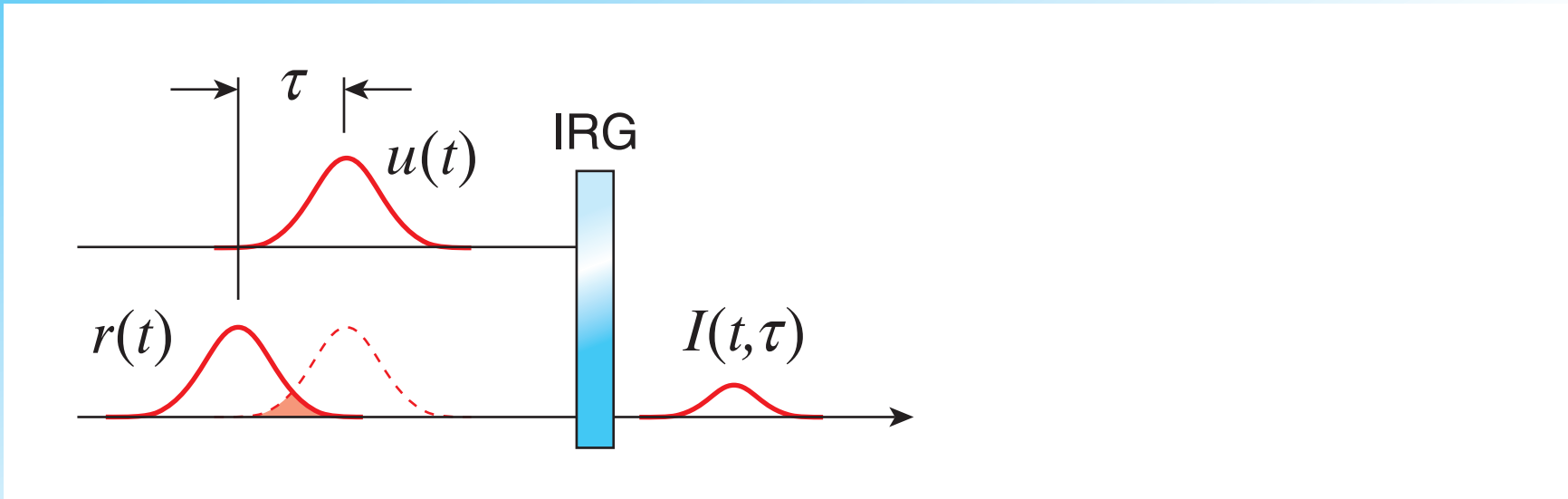
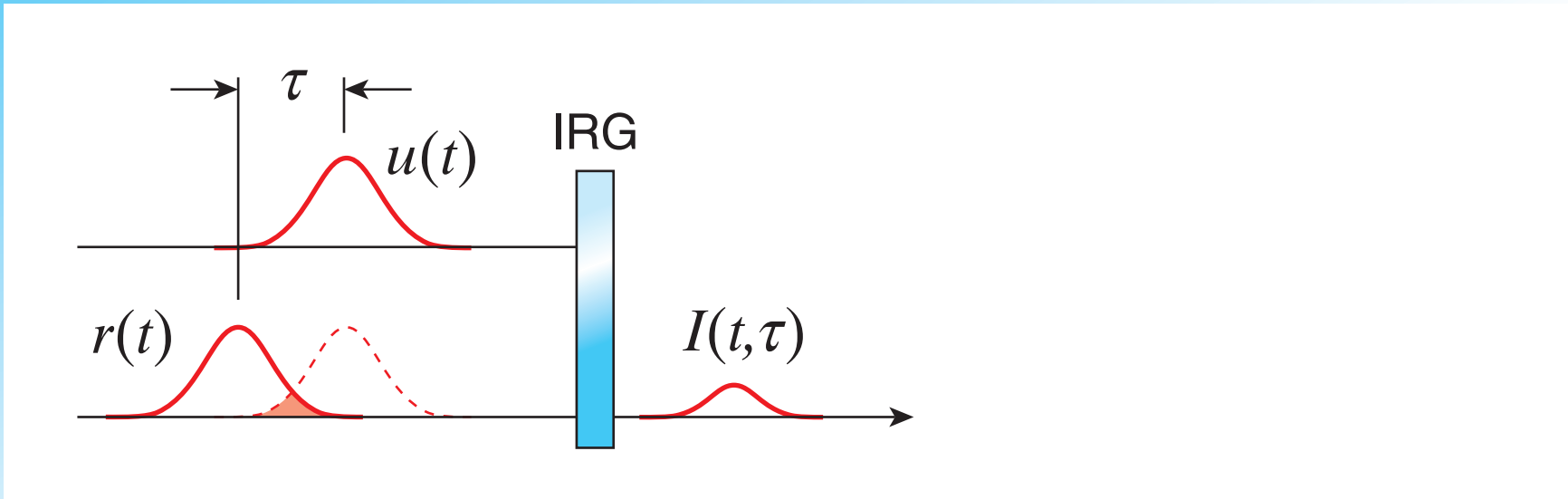


Transmitted intensity

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

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

Joint time-frequency measurements

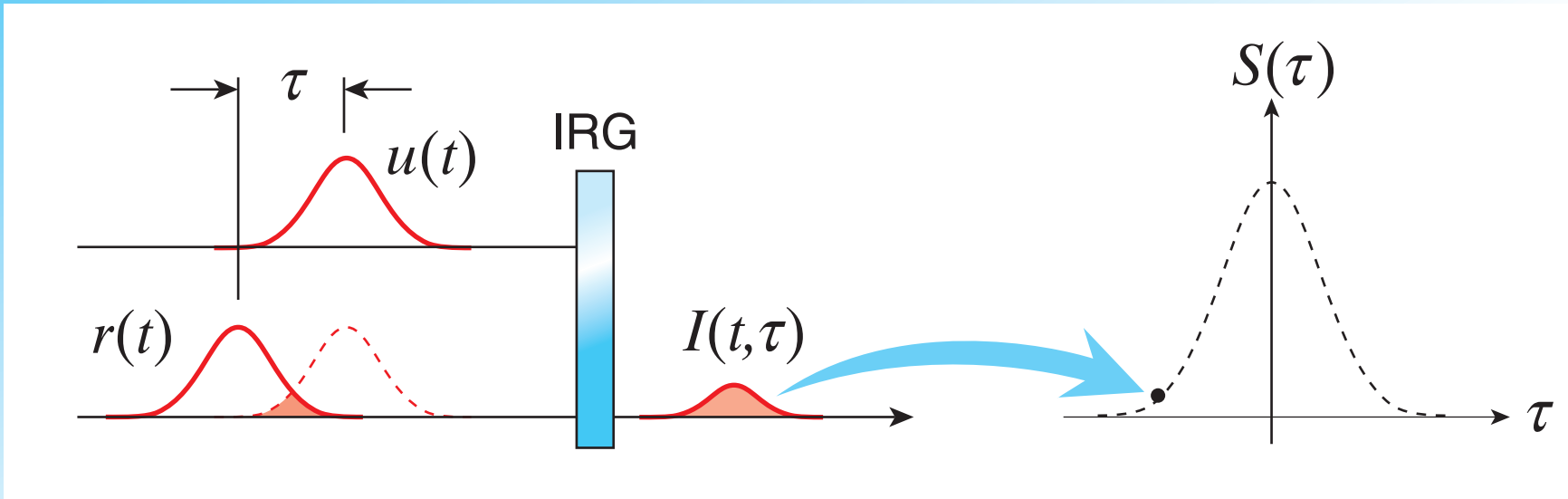


Transmitted intensity

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

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

Joint time-frequency measurements



…but detector integrates $I(t,\tau)$:

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

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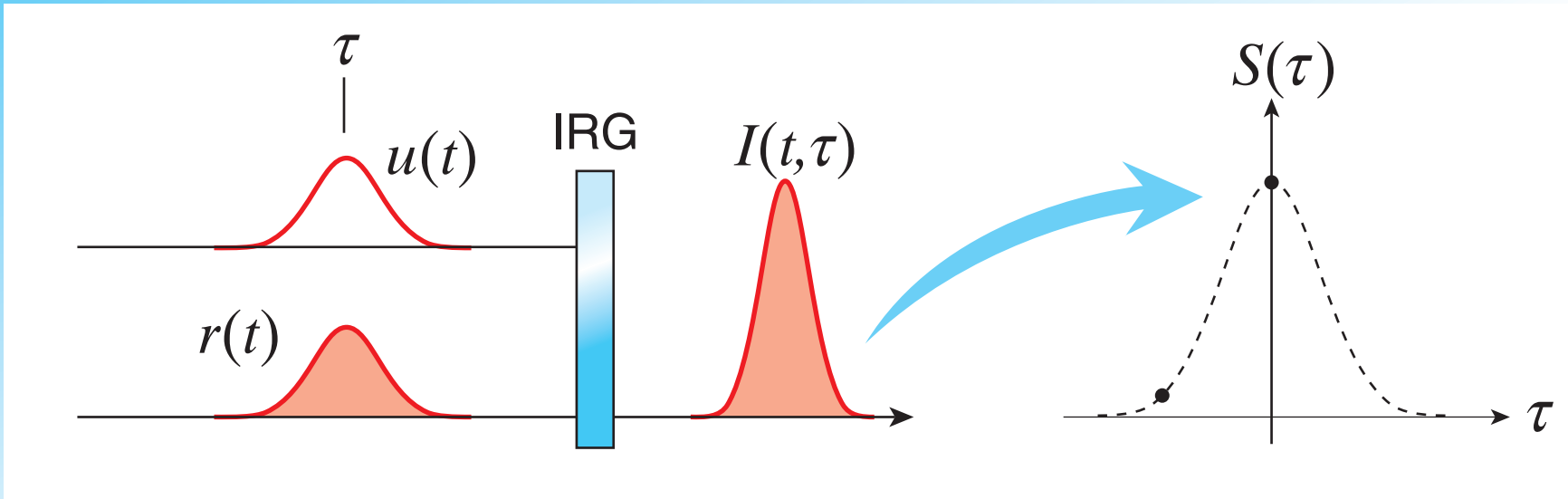

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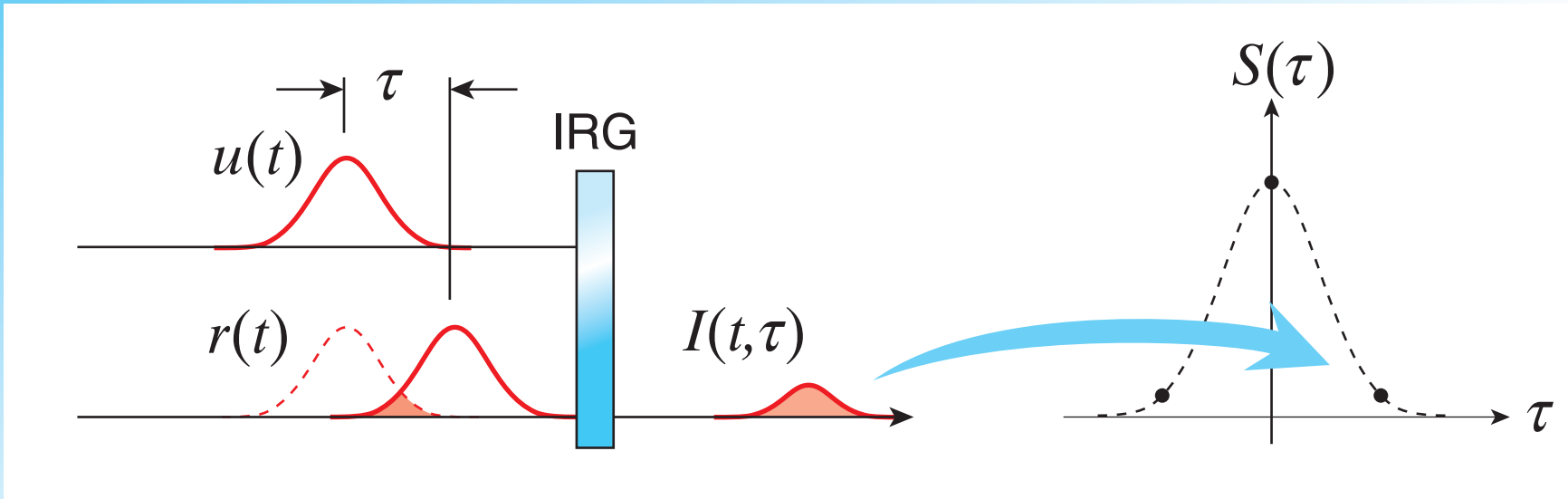

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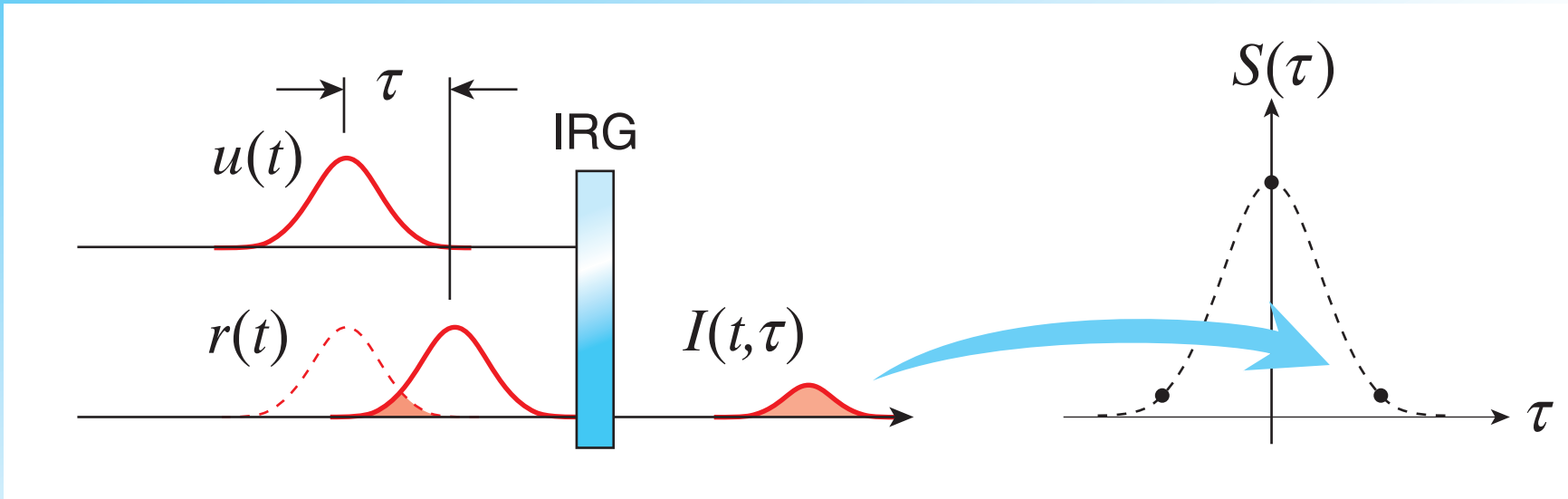

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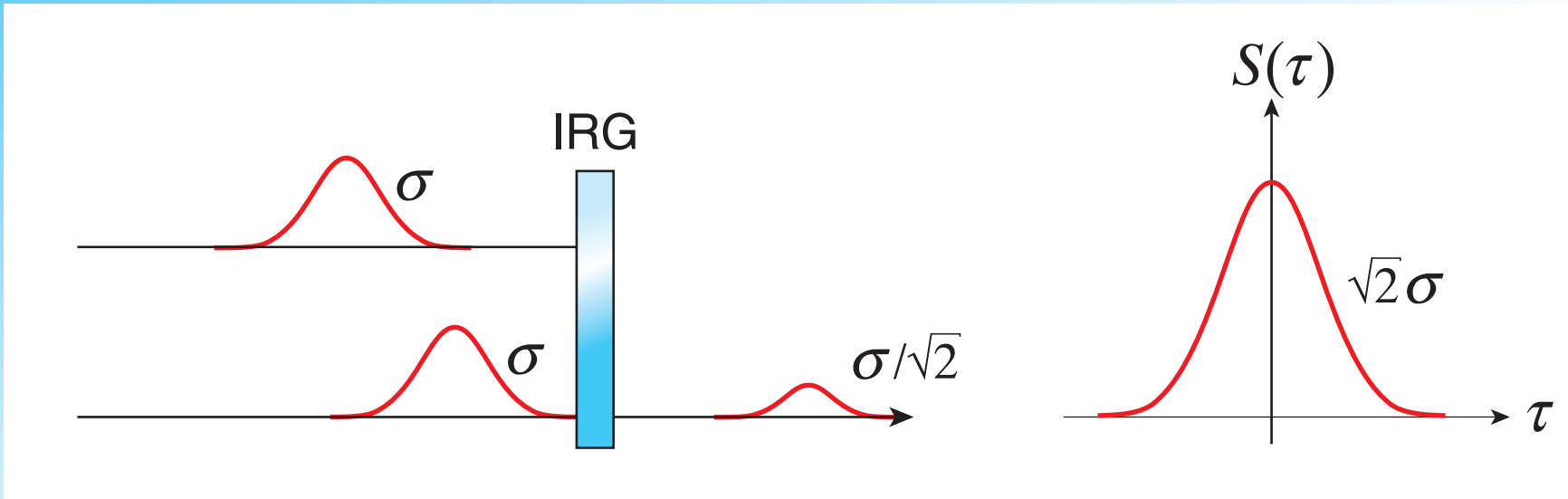
$$= \exp\left[-\frac{\tau^2}{2\sigma^2}\right] \int_{-\infty}^{\infty} \exp\left[-\frac{2u^2}{\sigma^2}\right] du = \sqrt{\frac{\pi}{2}}\,\sigma \exp\left[-\frac{\tau^2}{(\sigma\sqrt{2})^2}\right]$$

Joint time-frequency measurements



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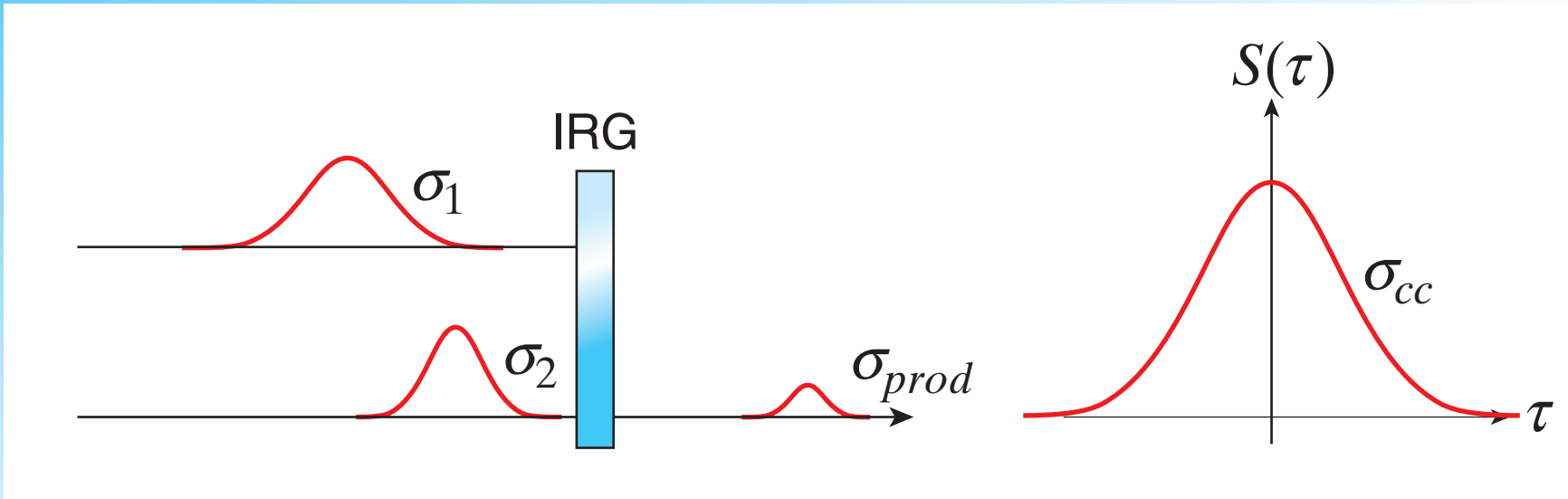
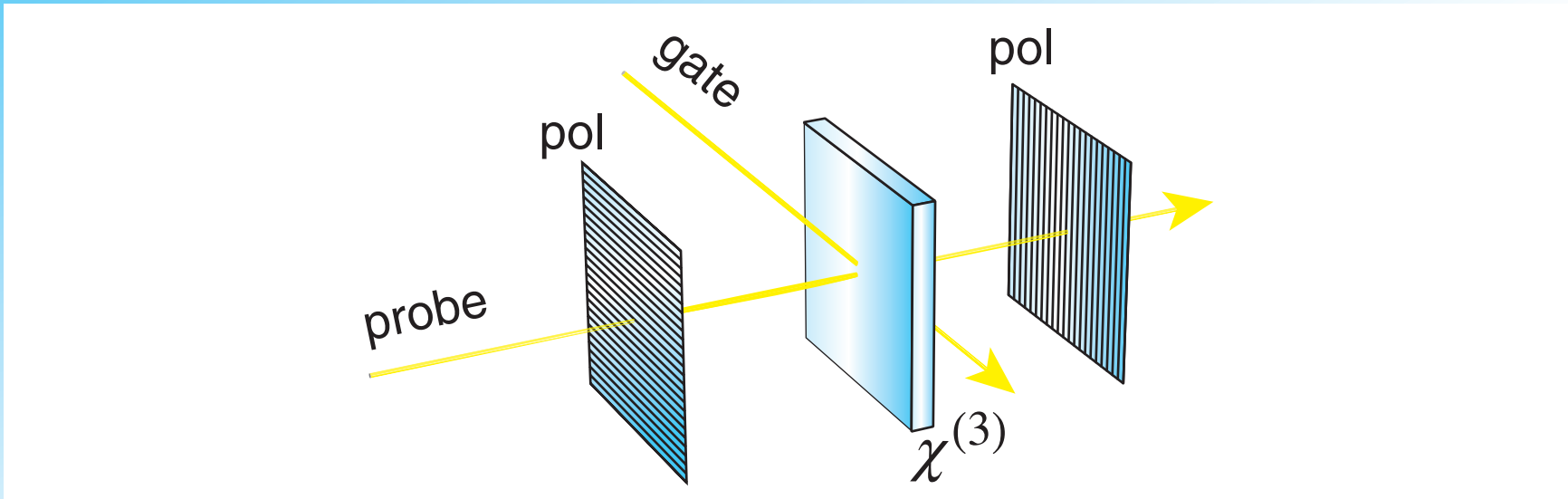


If gate and probe unequal:

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

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

Joint time-frequency measurements

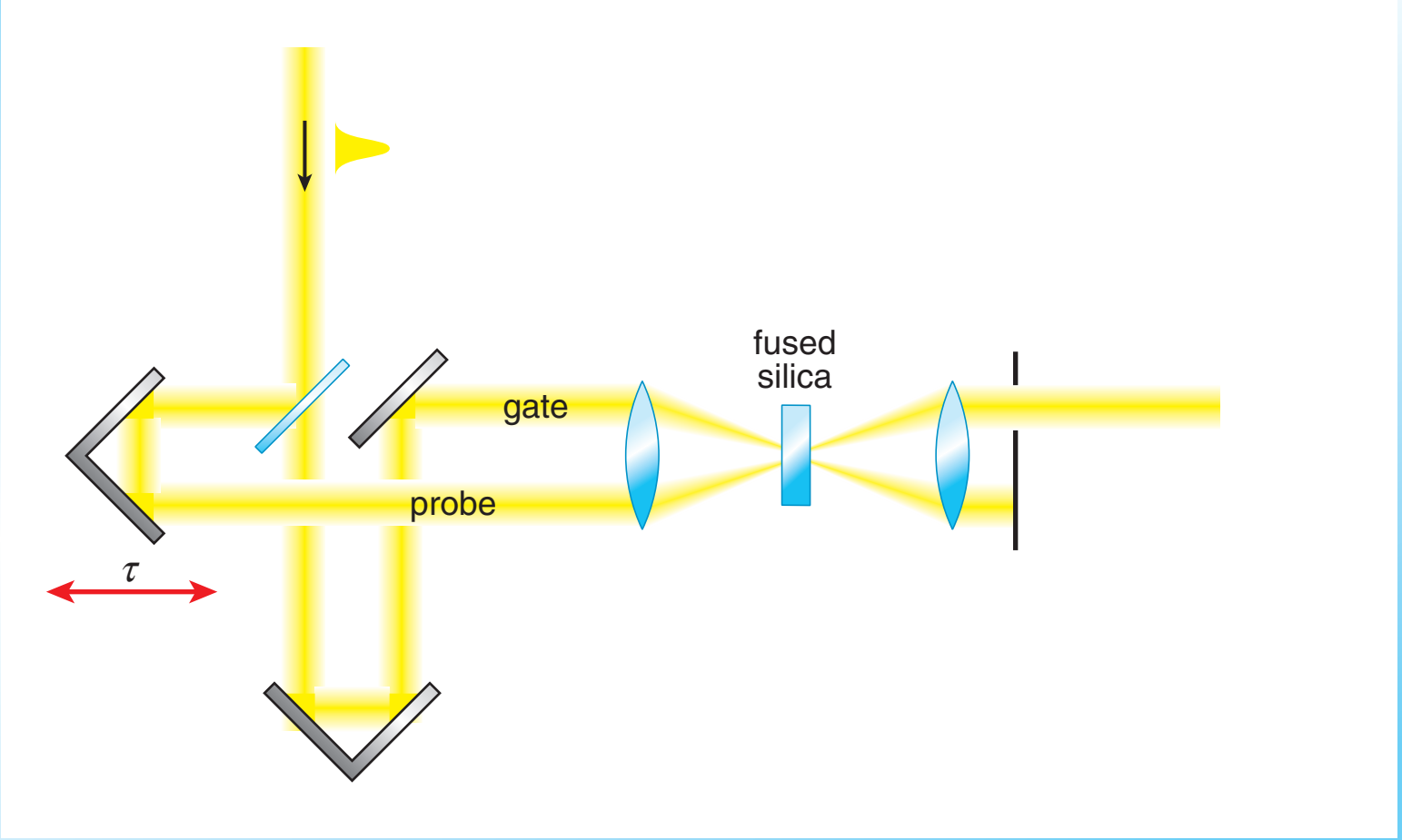


Transmitted field:

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

Joint time-frequency measurements

FROG: frequency-resolved optical gating



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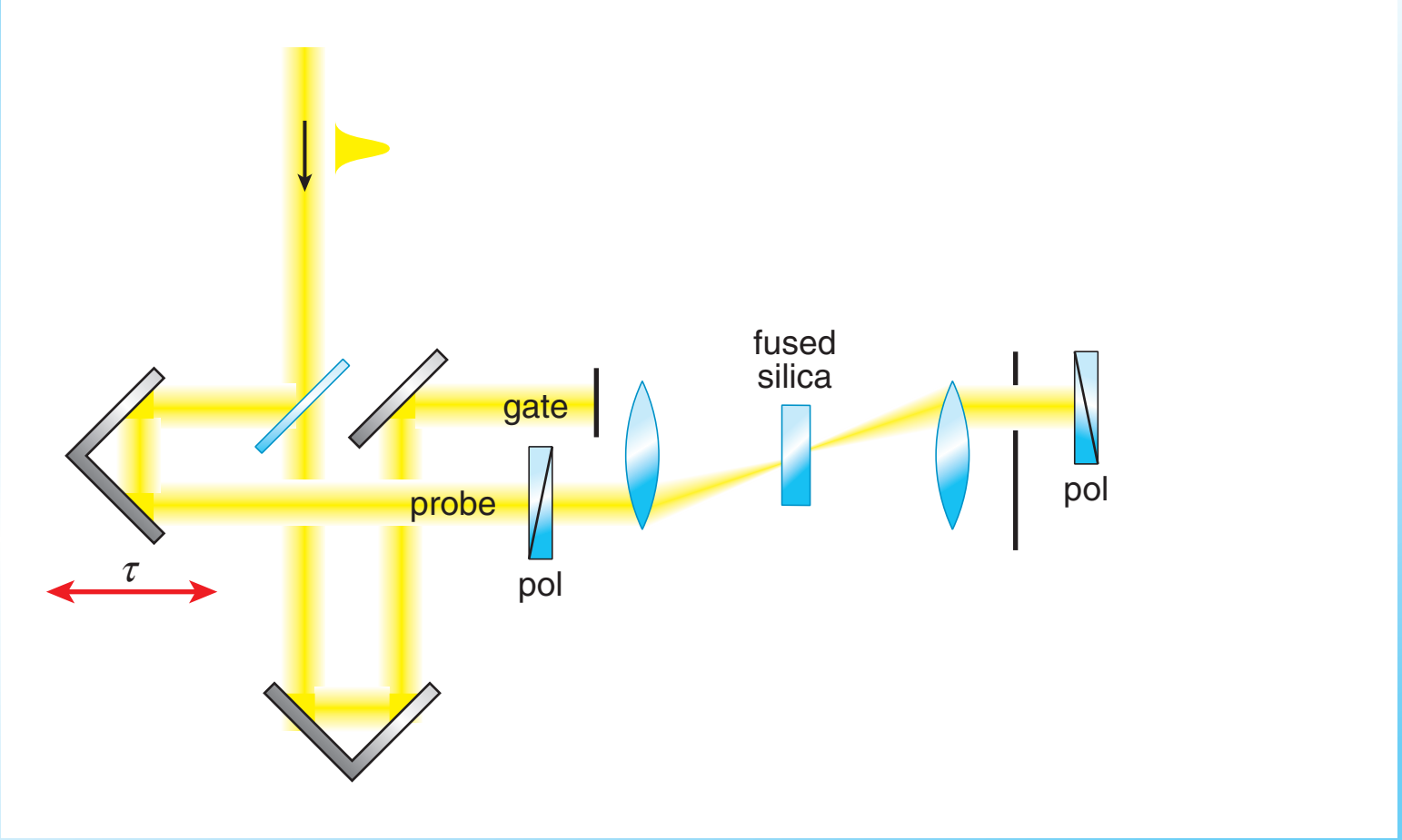

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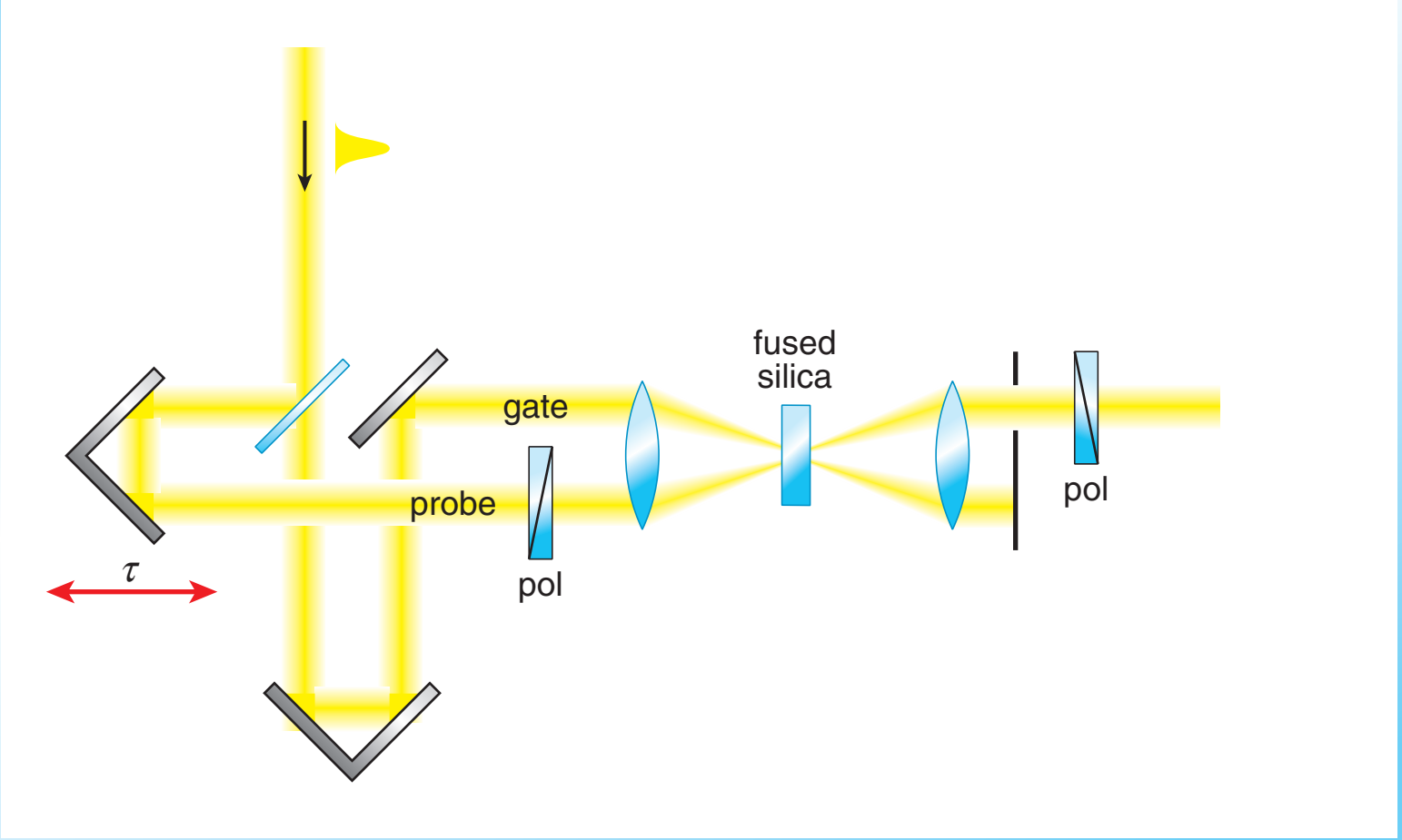

Joint time-frequency measurements

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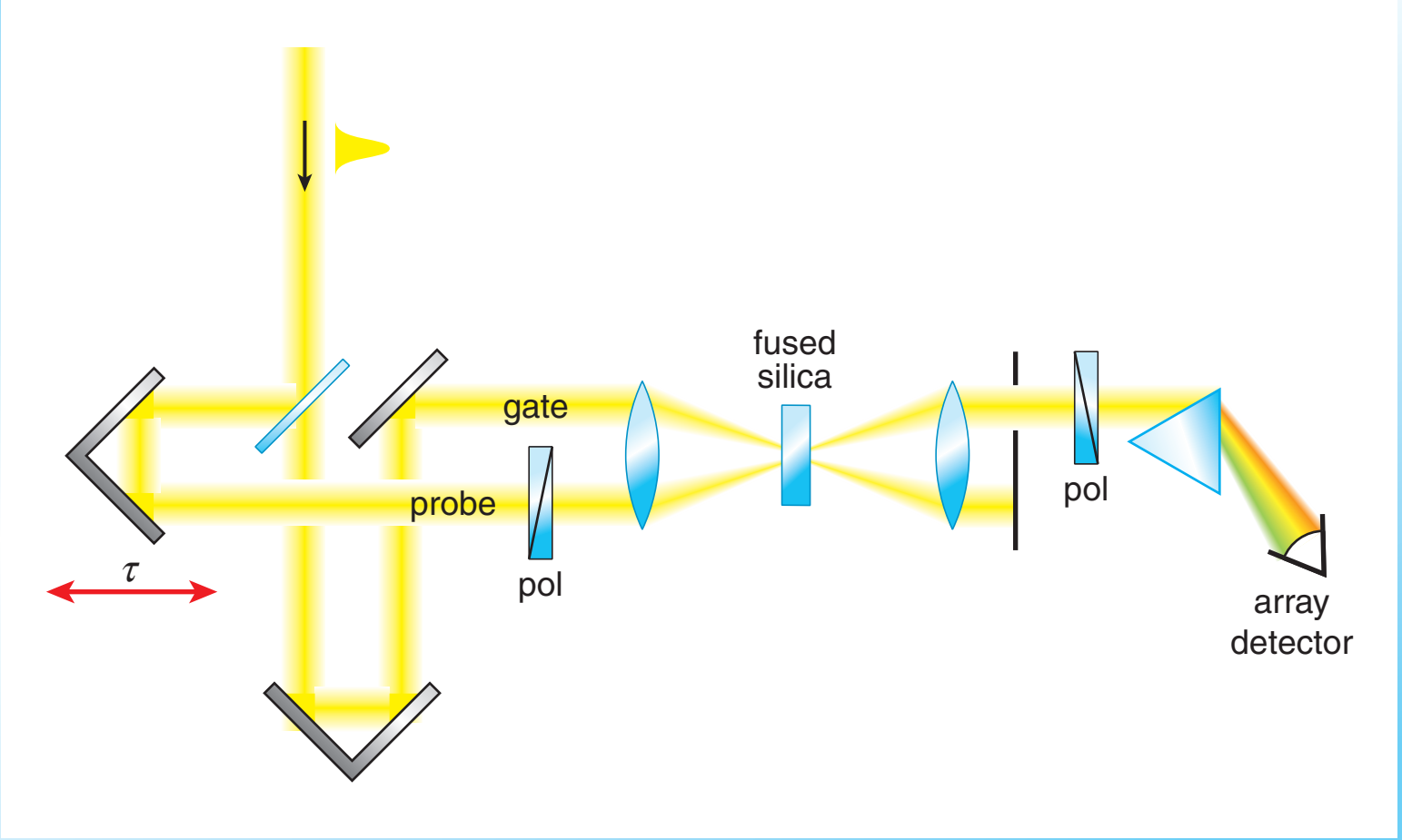
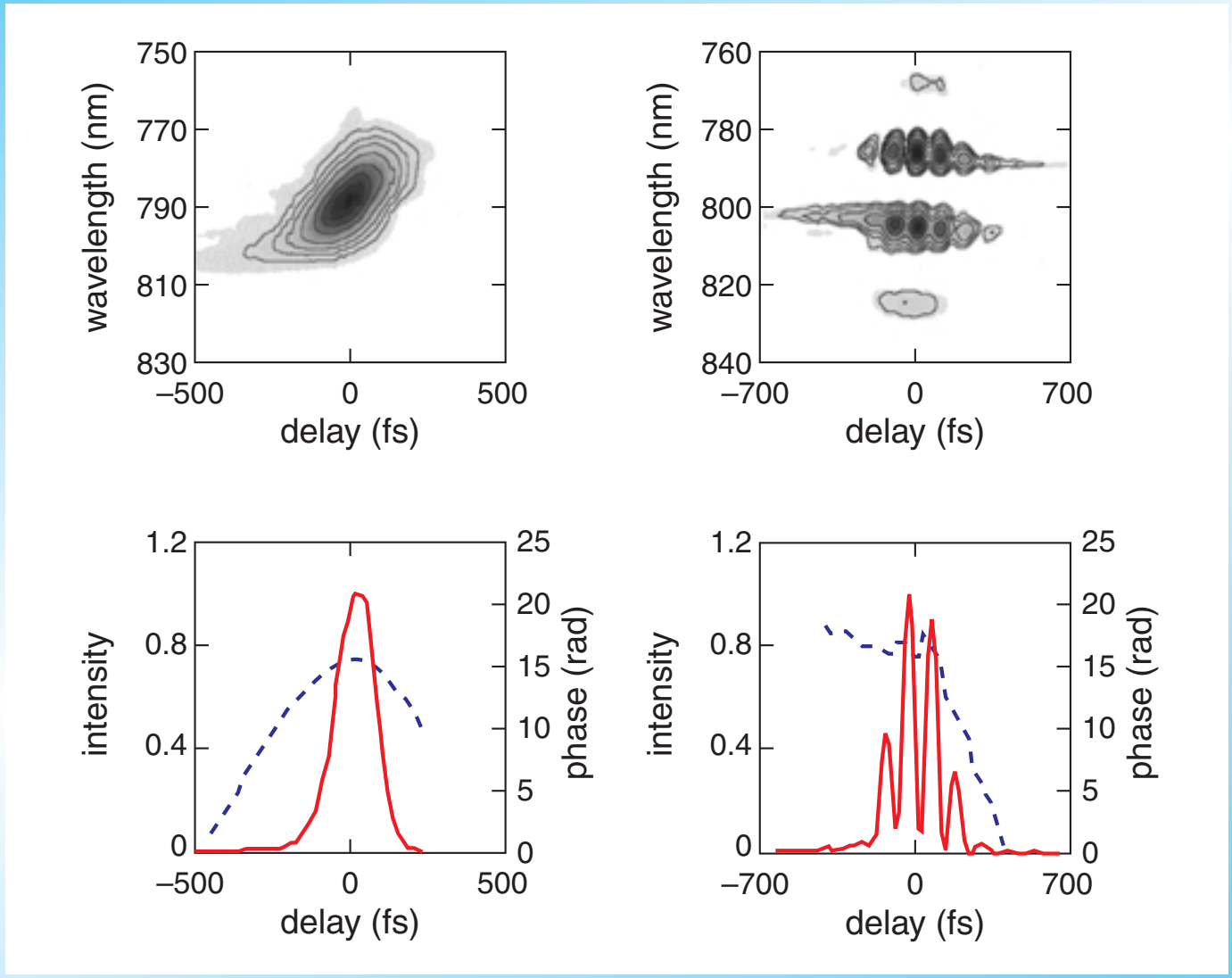
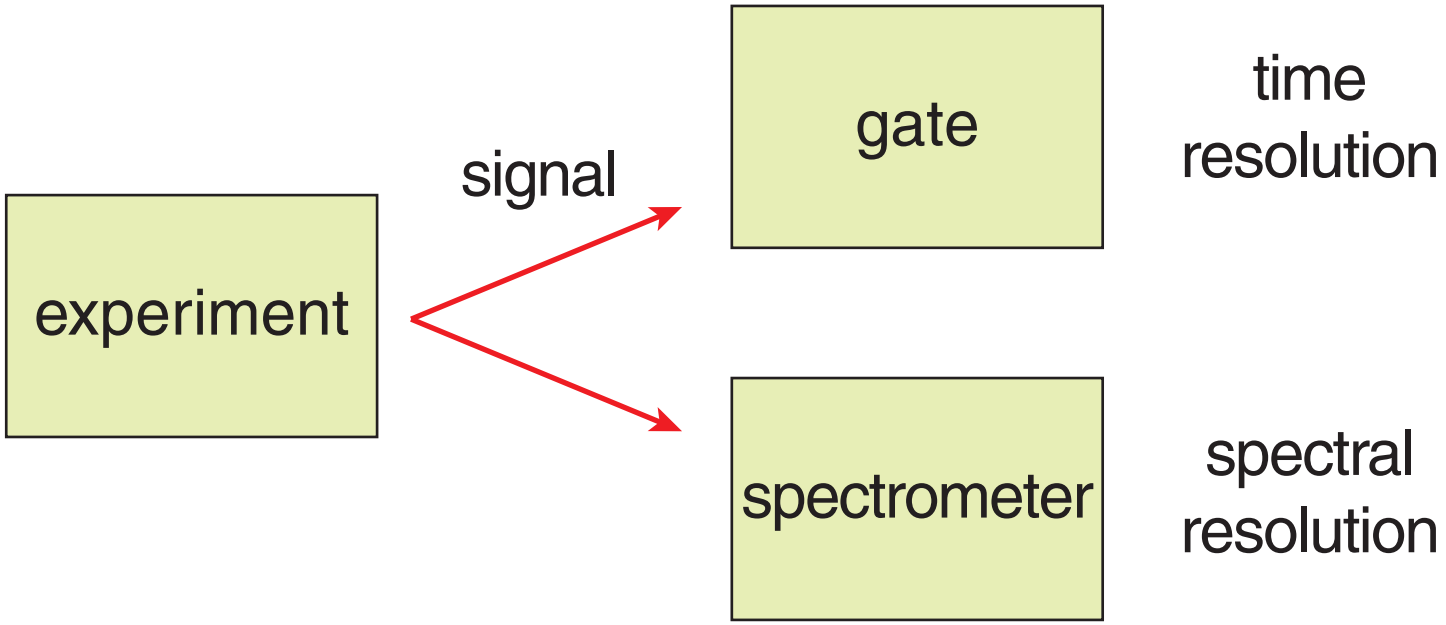


Joint time-frequency measurements



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

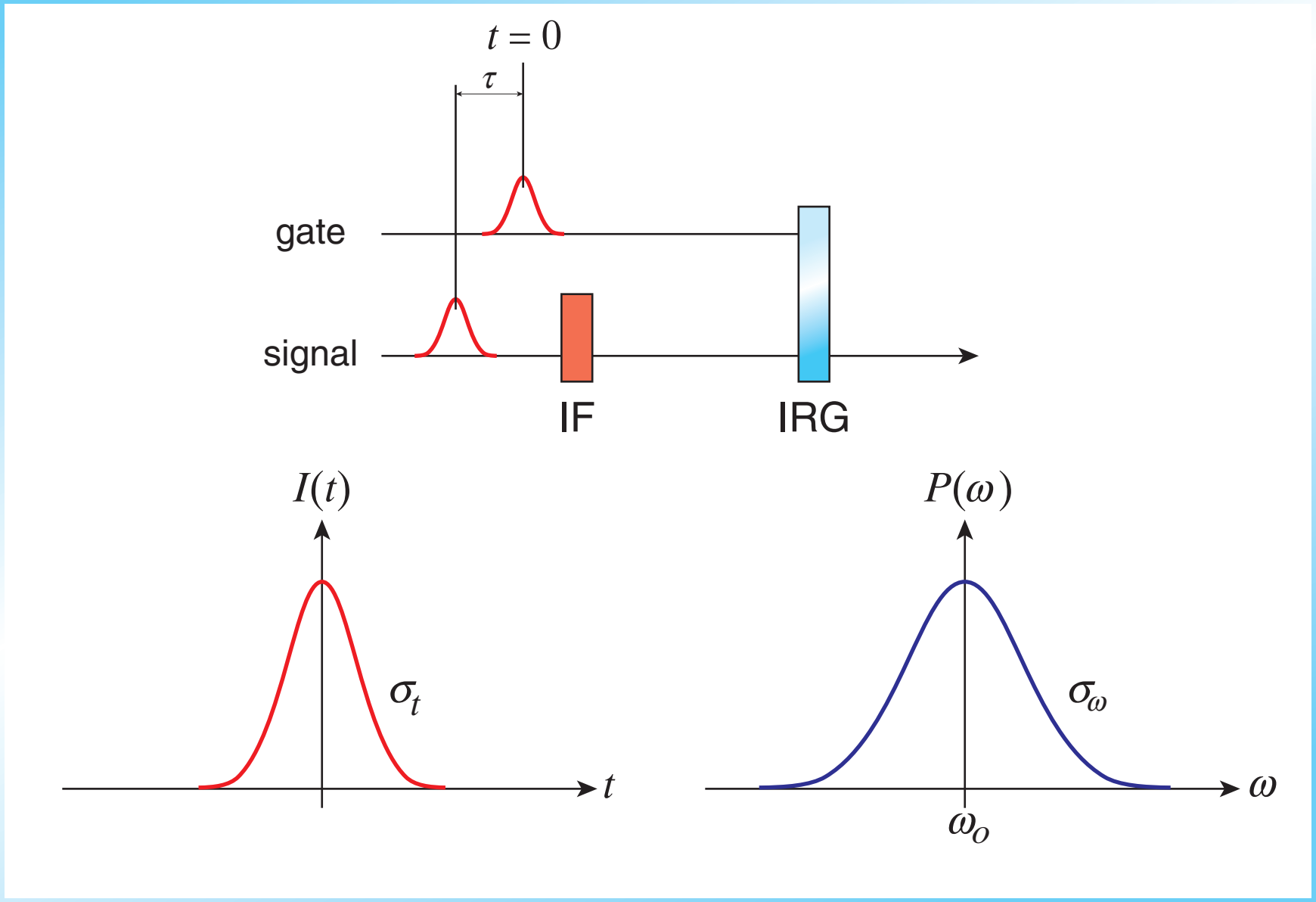
Joint time-frequency measurements



Joint time-frequency measurements

What are the resolution limits?

Experiment 1



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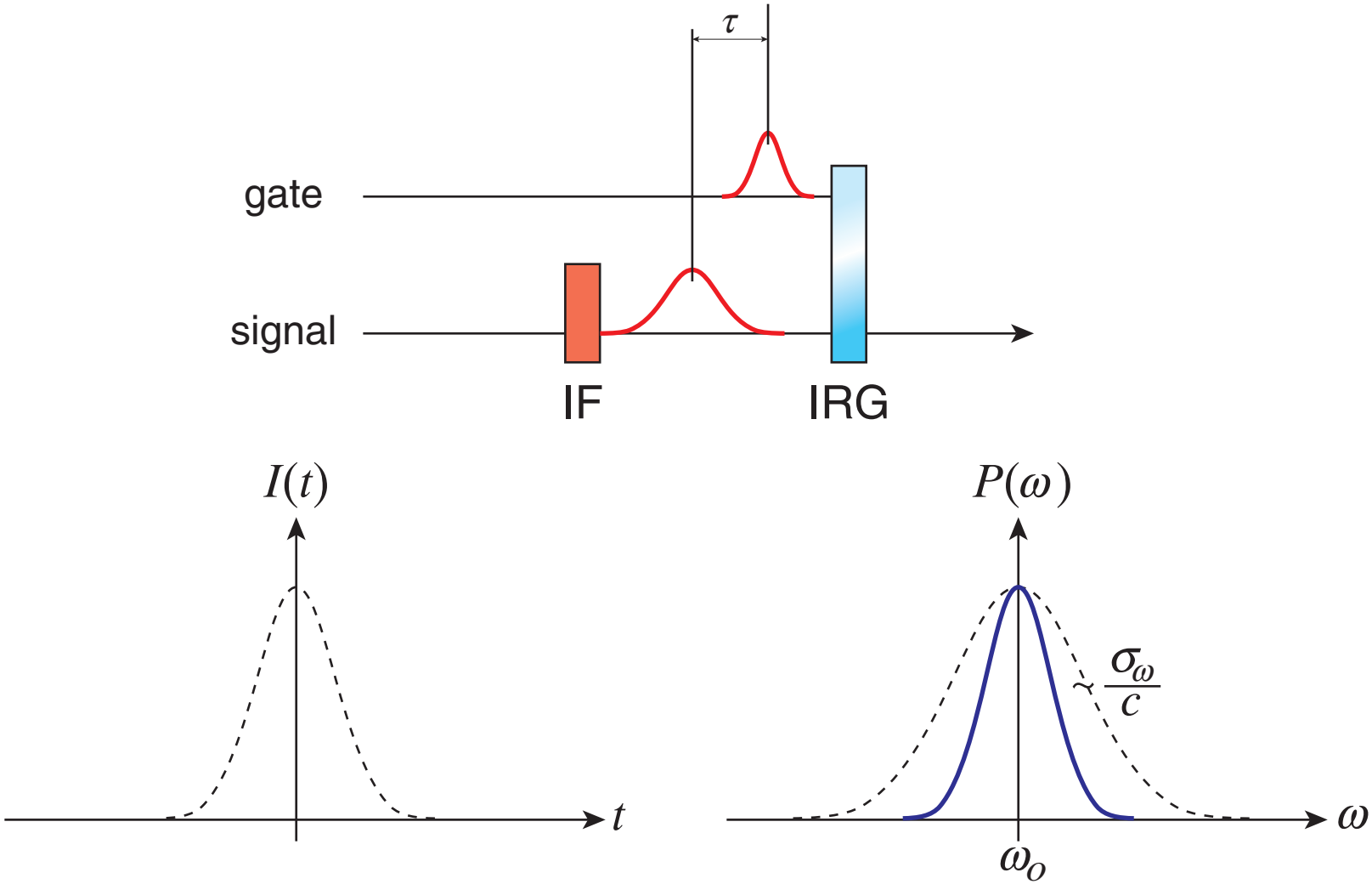

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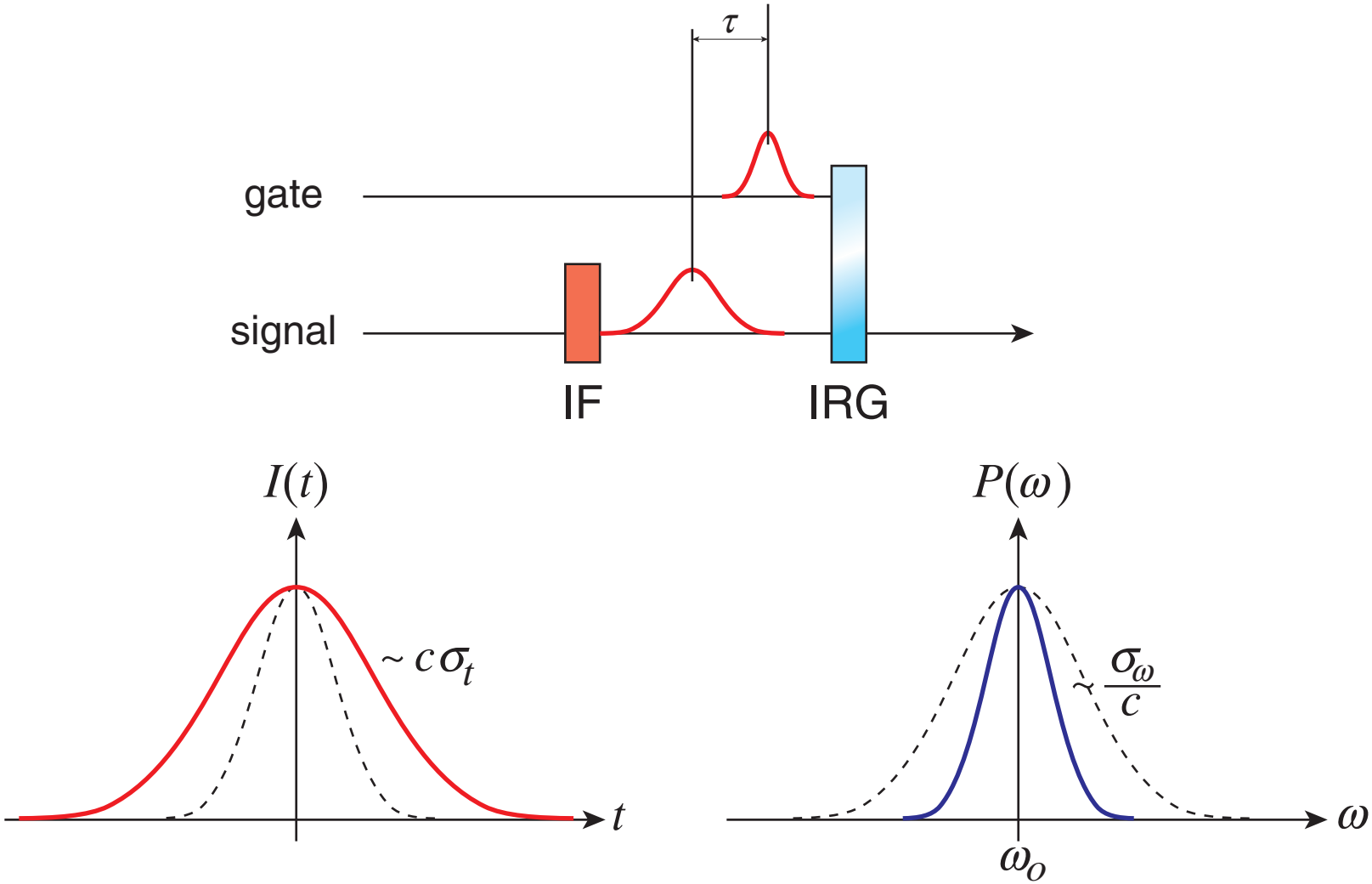

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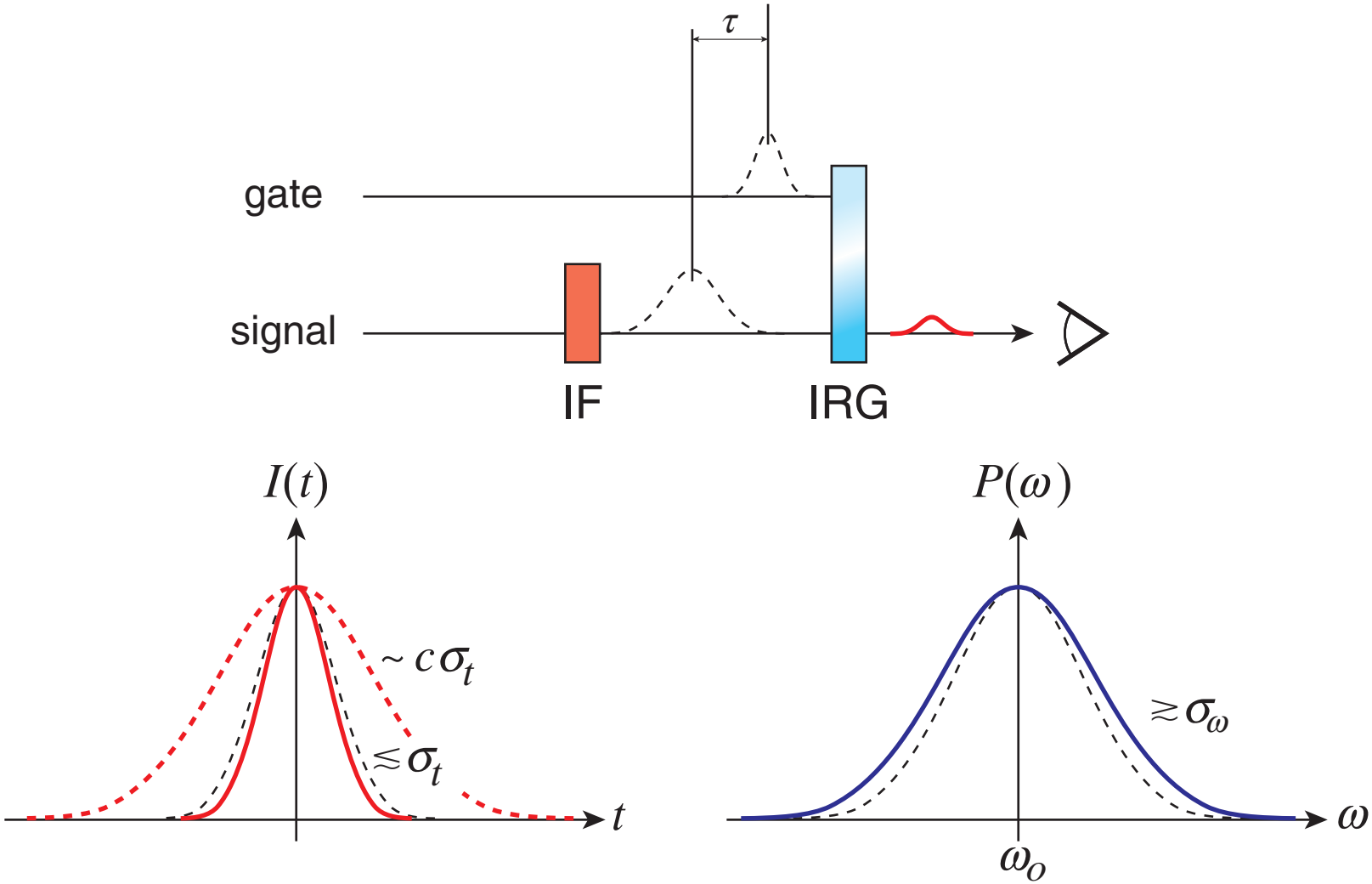
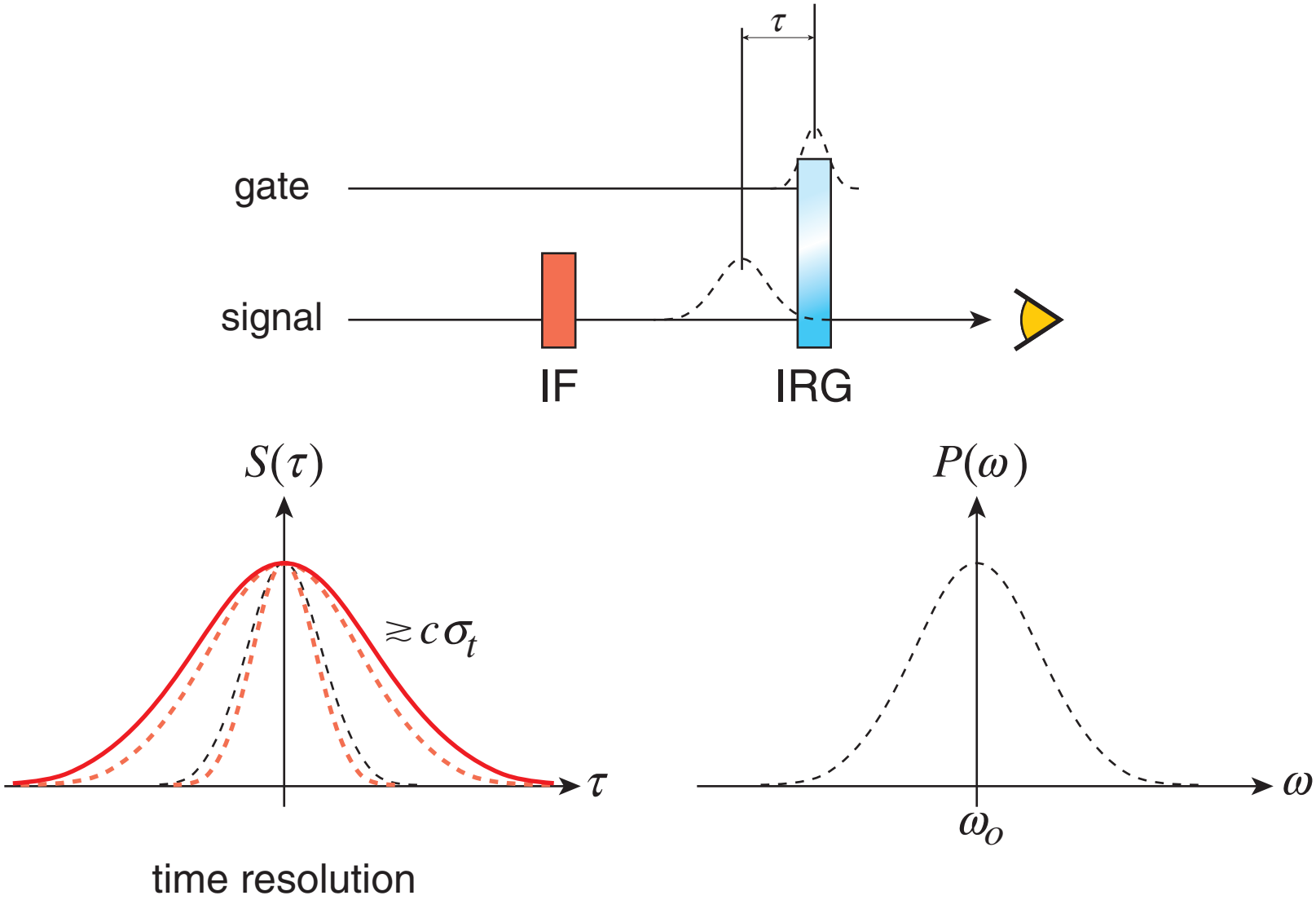
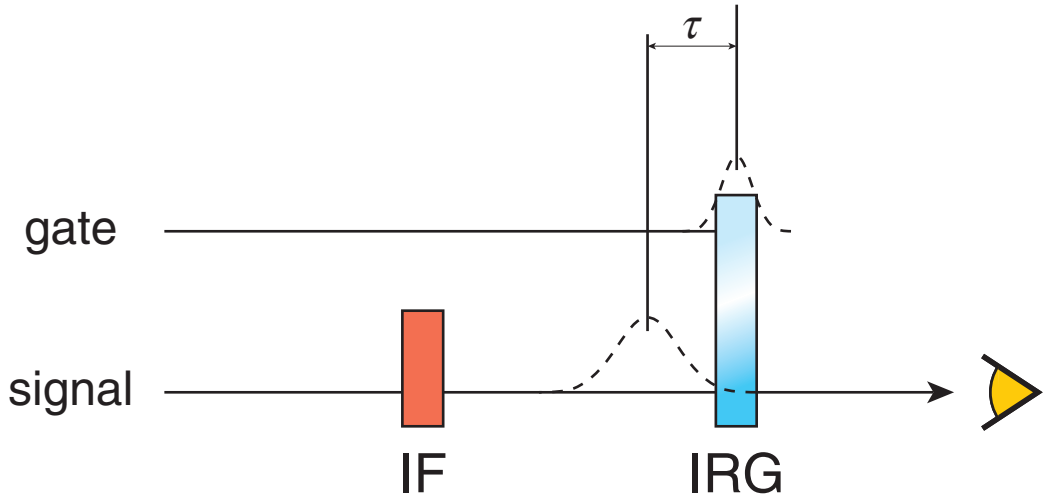
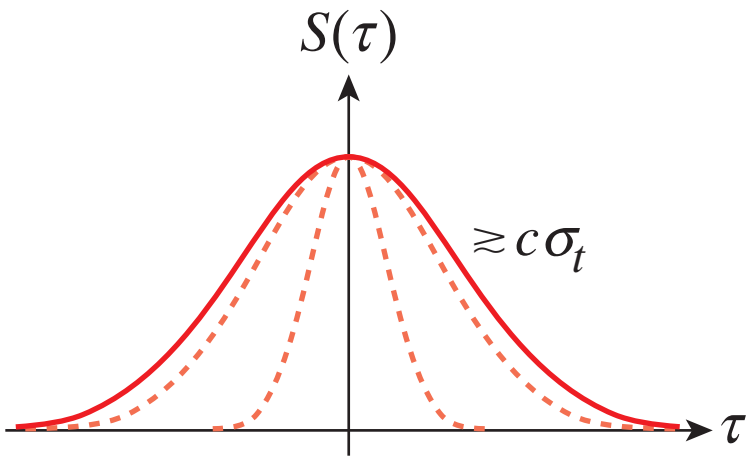


Experiment 1

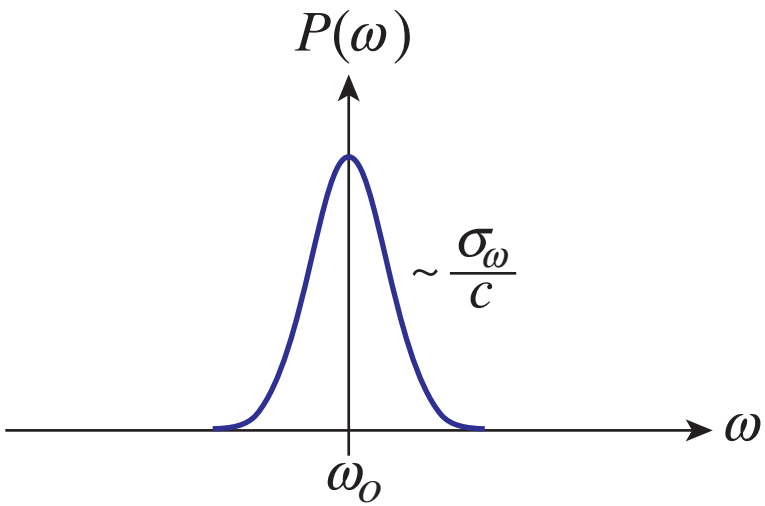


Experiment 1



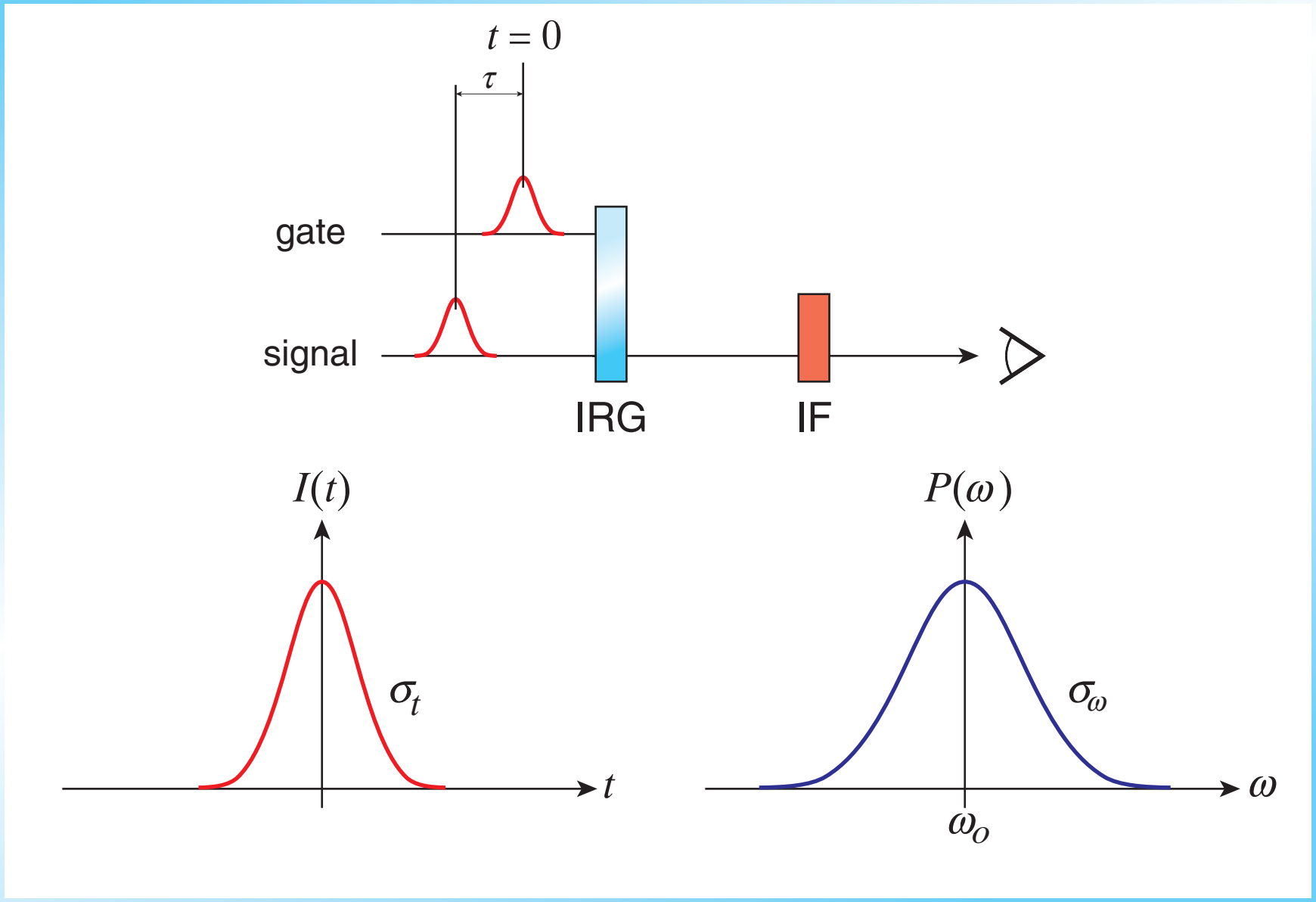


time resolution

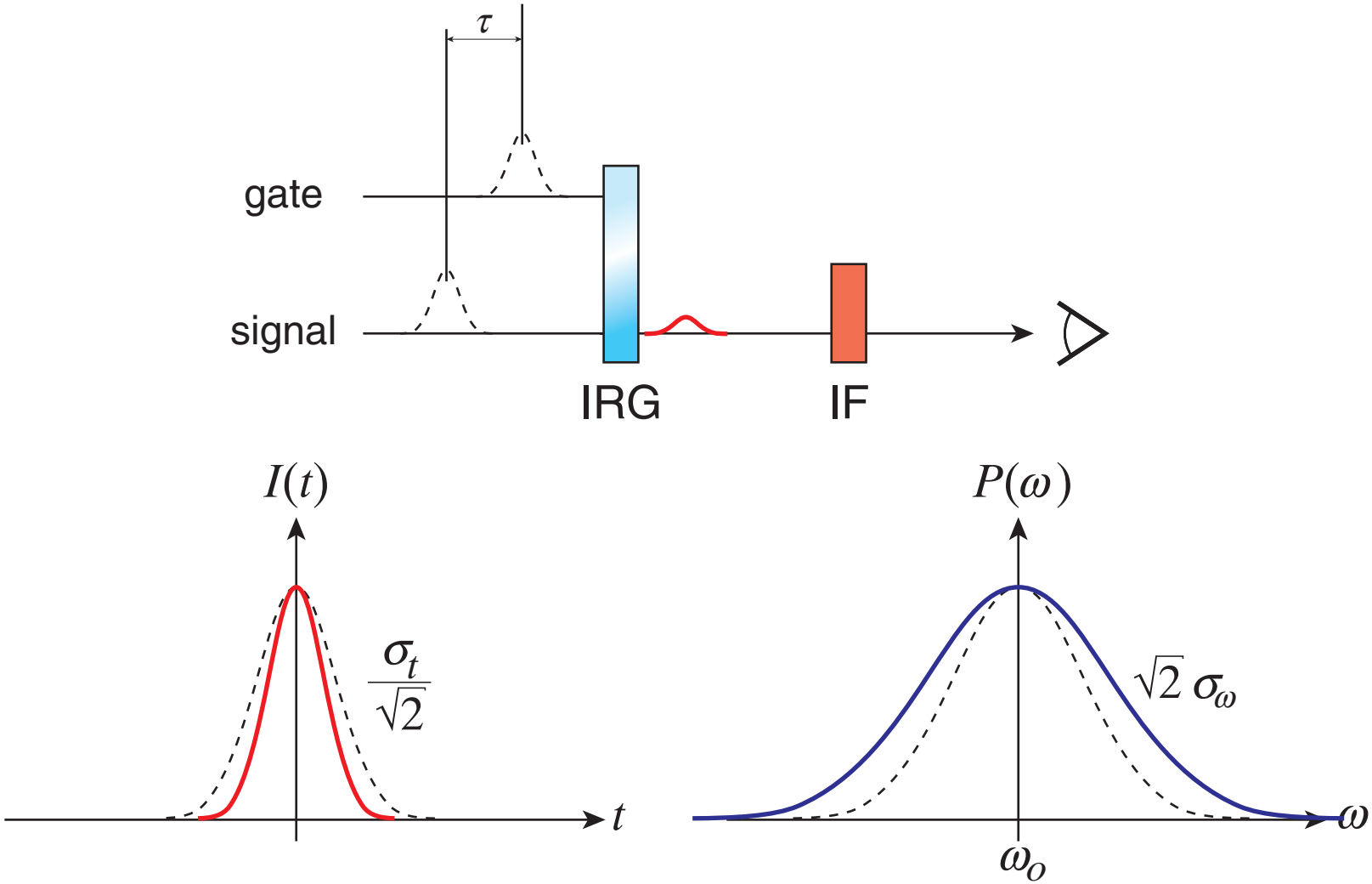


frequency resolution

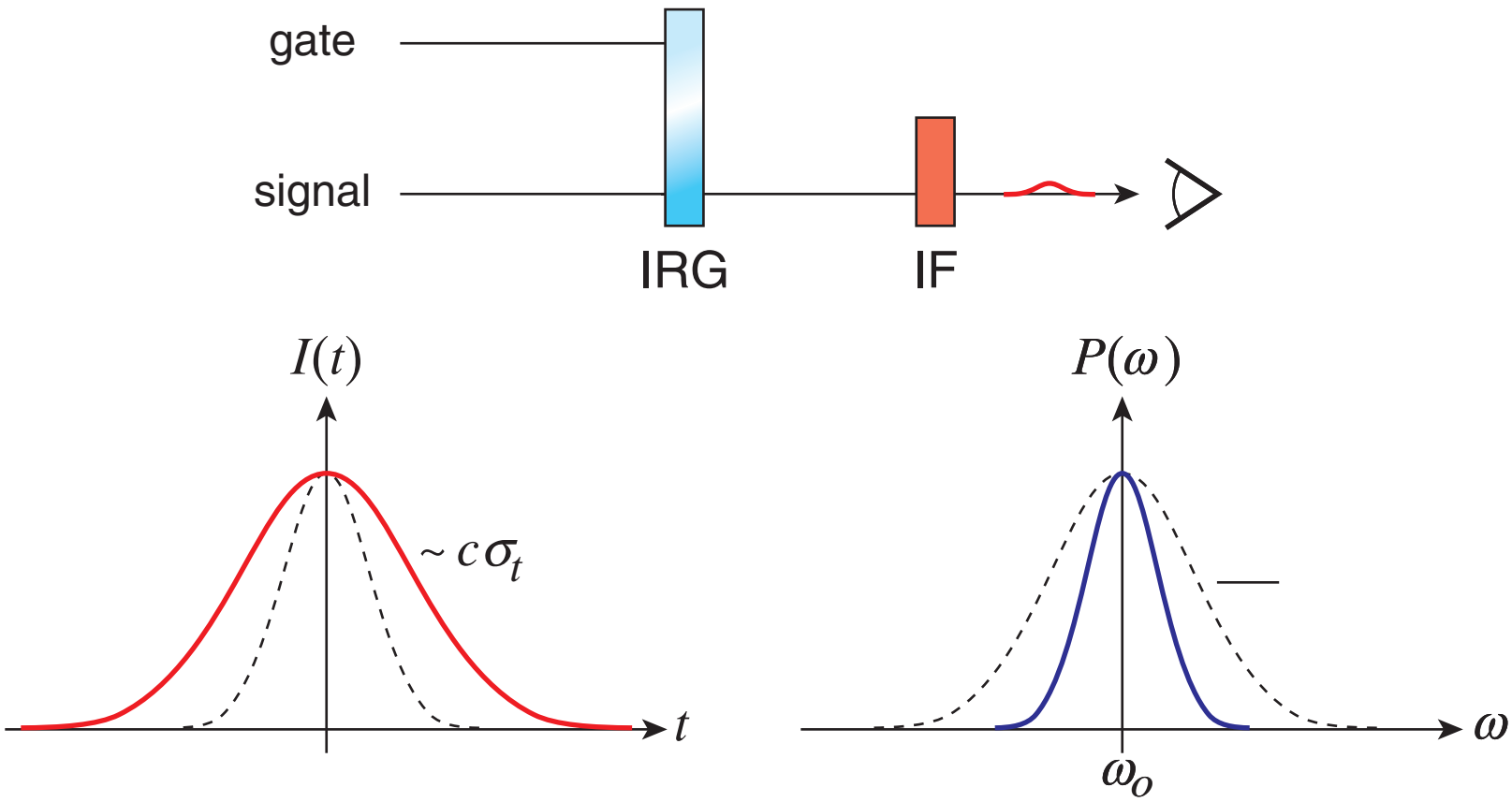
Experiment 2



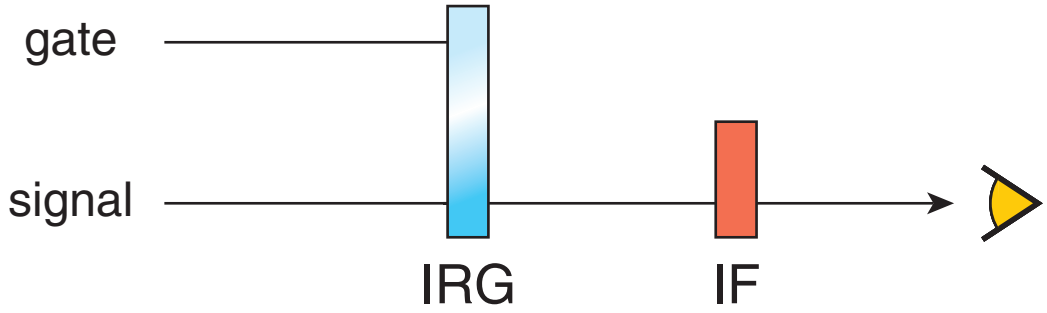
Experiment 2

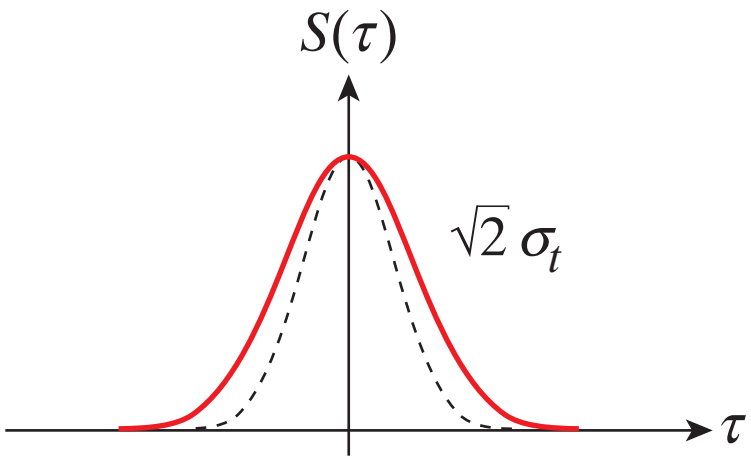


Experiment 2

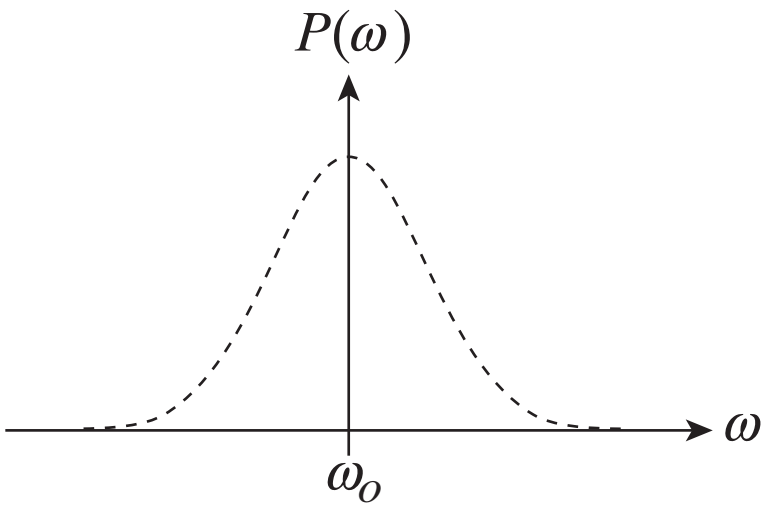


Experiment 2

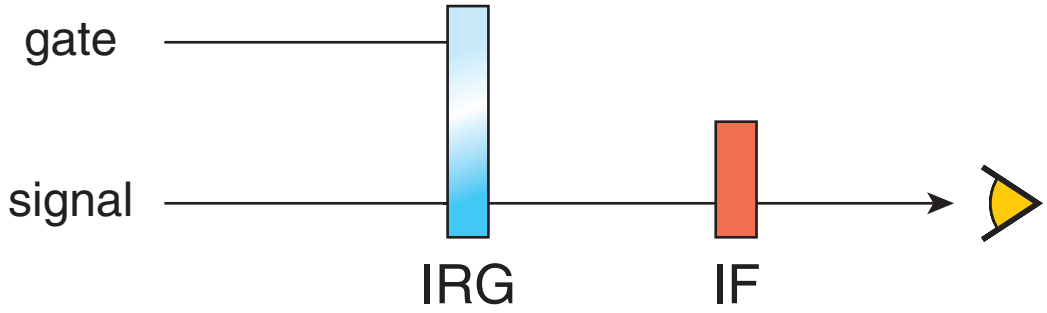


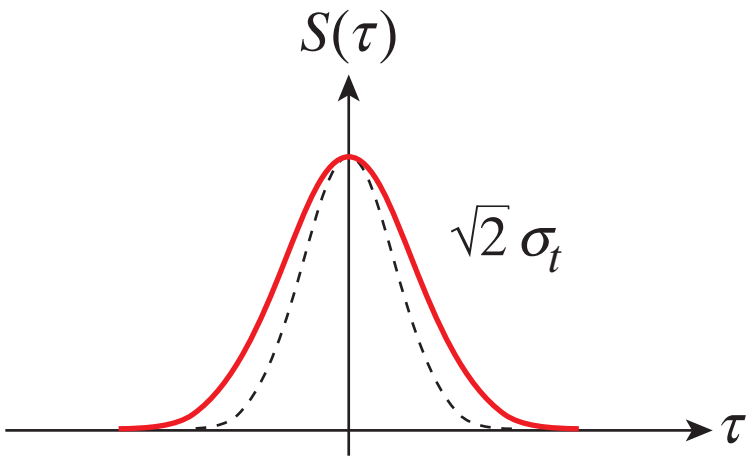


time resolution

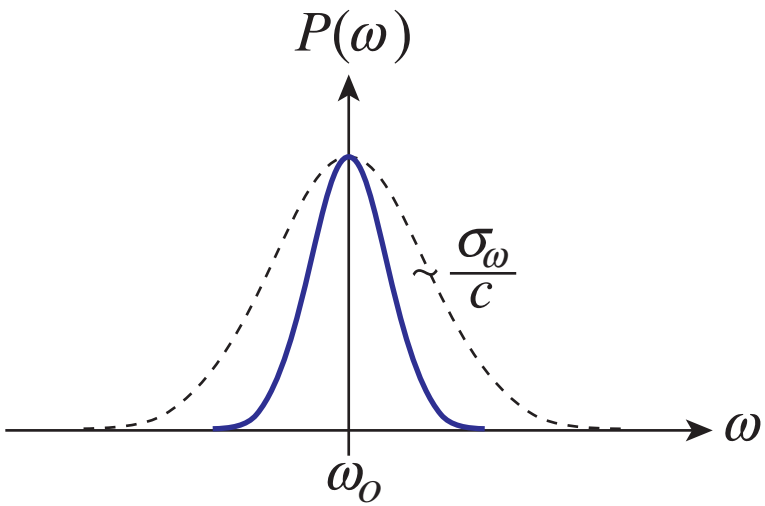


Experiment 2





time resolution



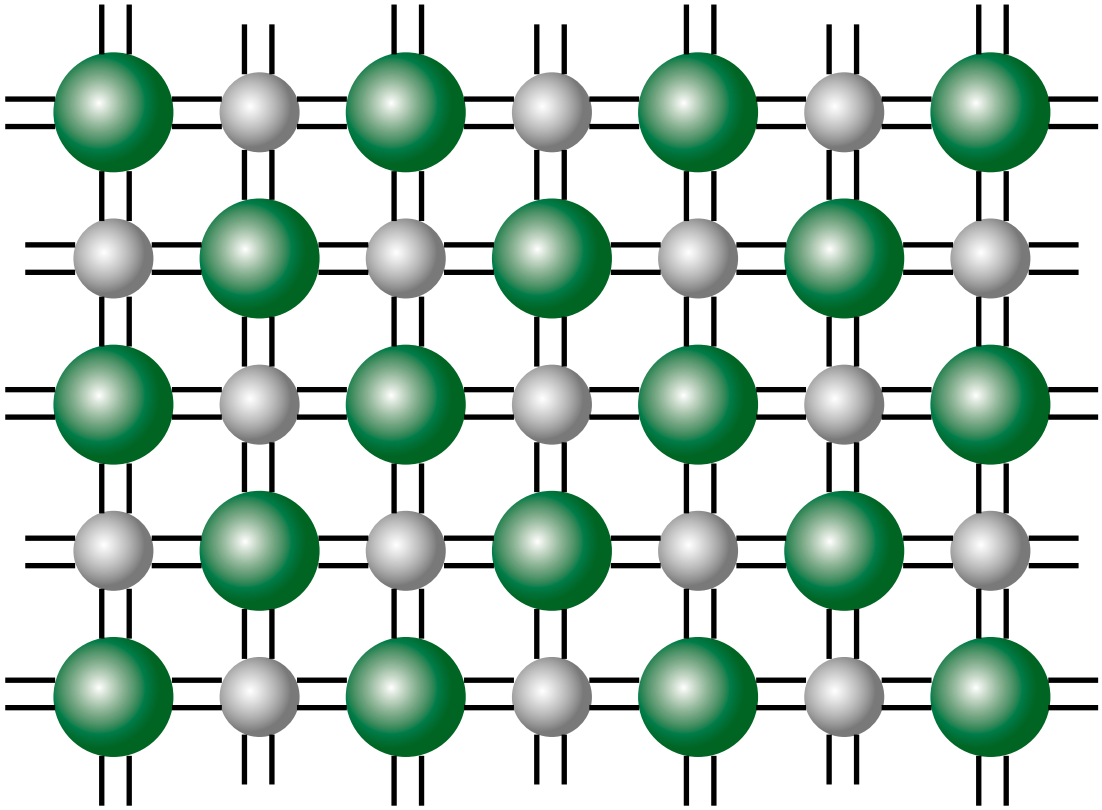
spectral resolution

Outline

- linear and nonlinear propagation
- femtosecond measurements
- **dynamics of phase transitions**
- laser-assisted surface etching

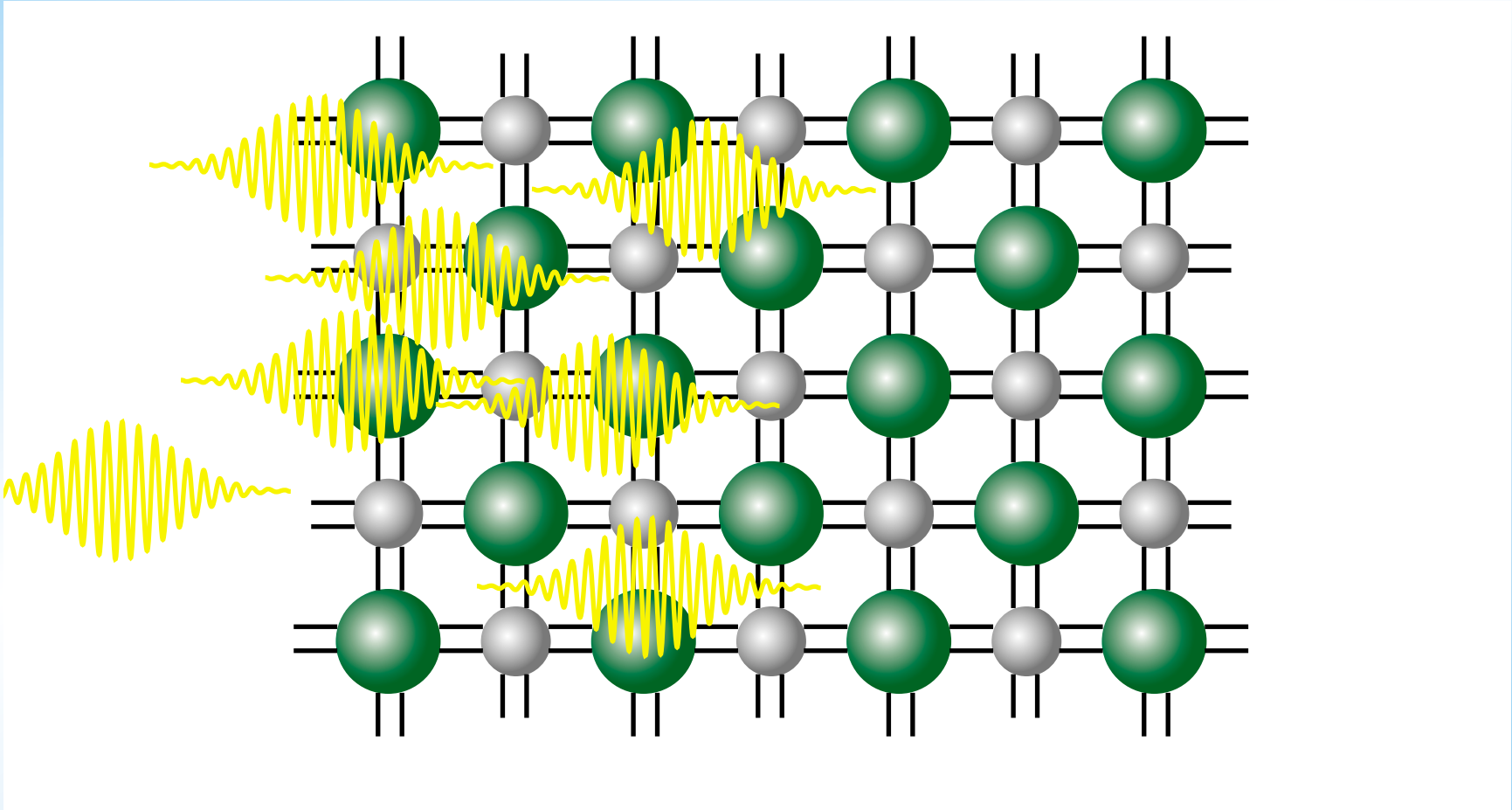
Introduction

how do femtosecond laser pulses alter a solid?



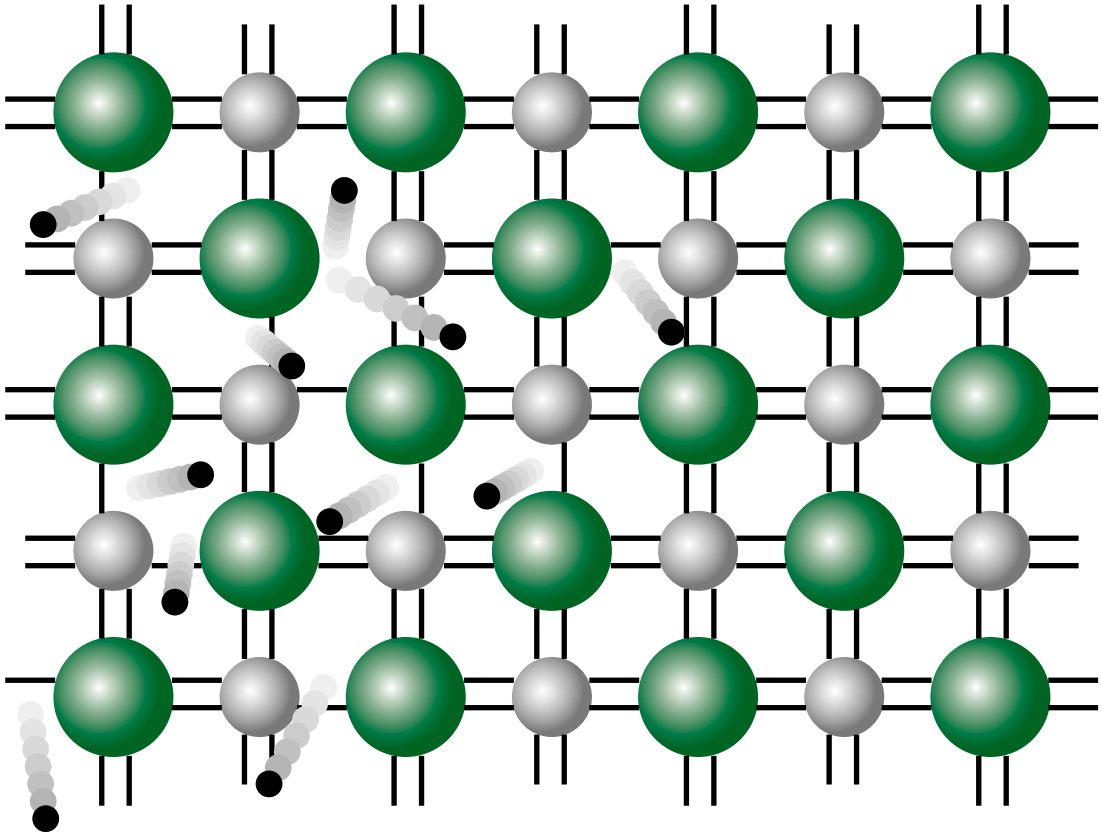
Introduction

photons excite valence electrons…



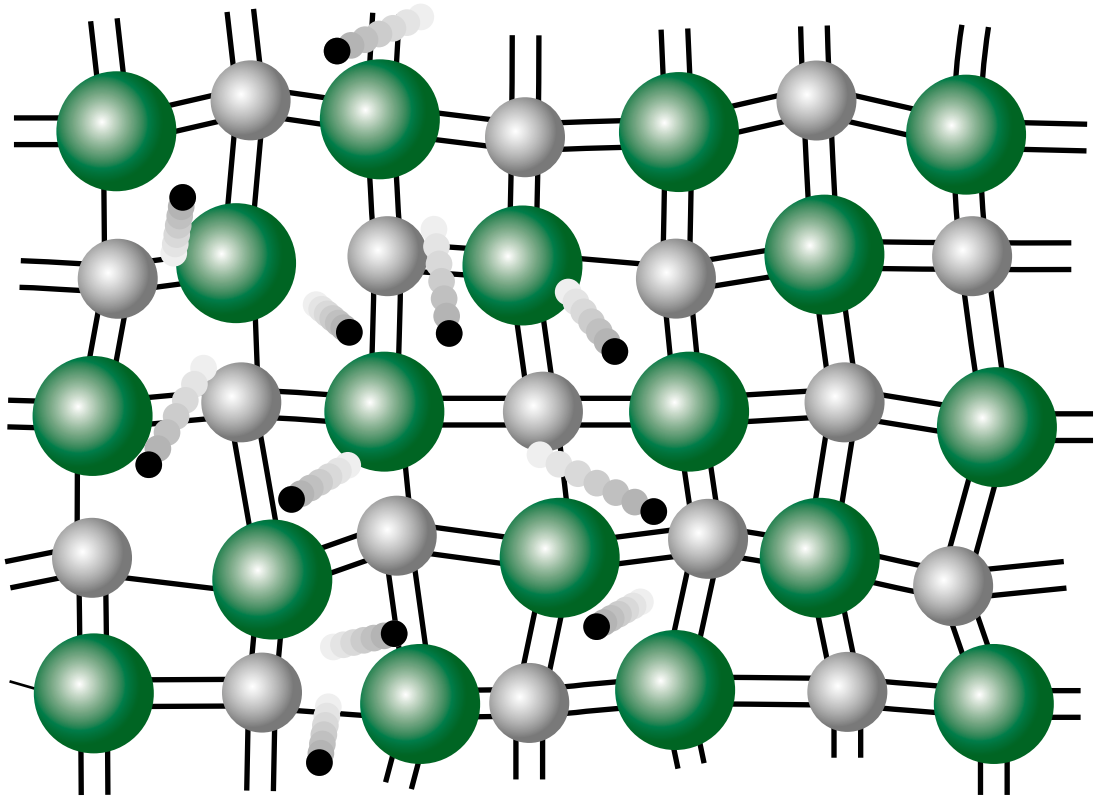
Introduction

...and create free electrons...



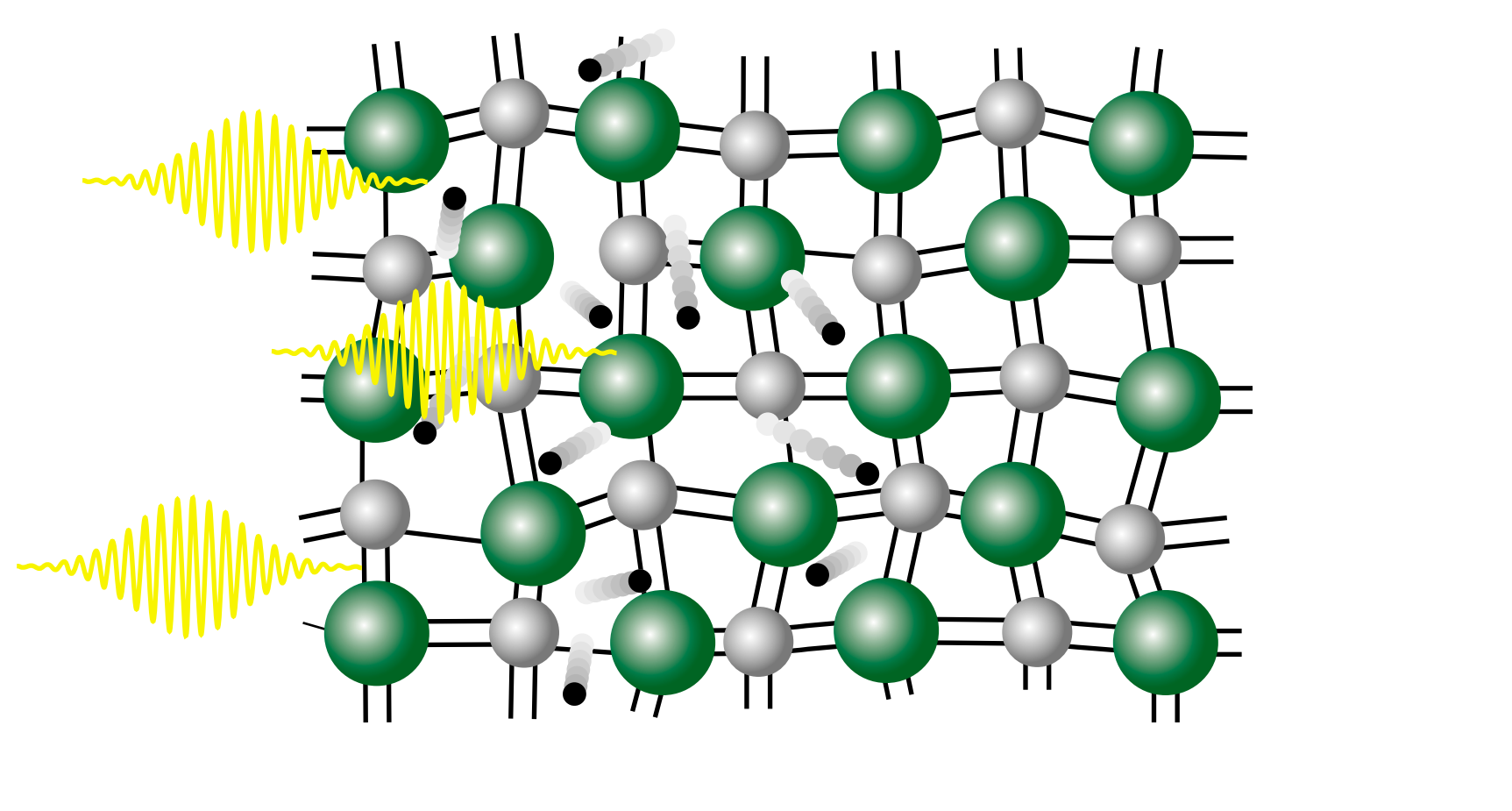
Introduction

…causing electronic and structural changes…



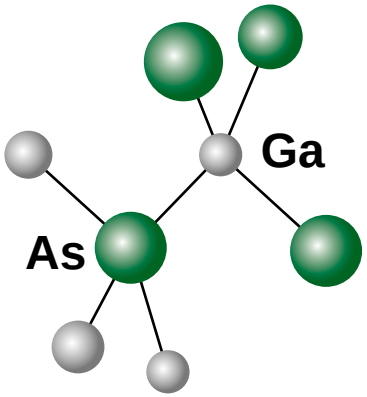
Introduction

…which we measure with another pulse



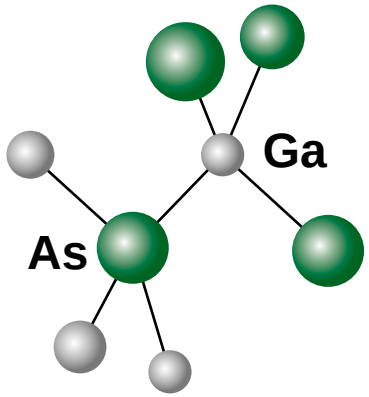
Introduction

structure

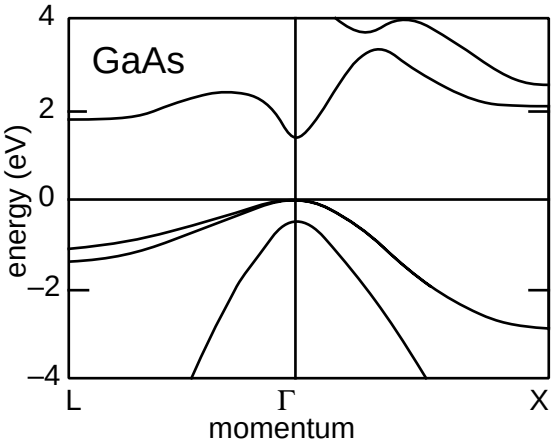


Introduction

structure

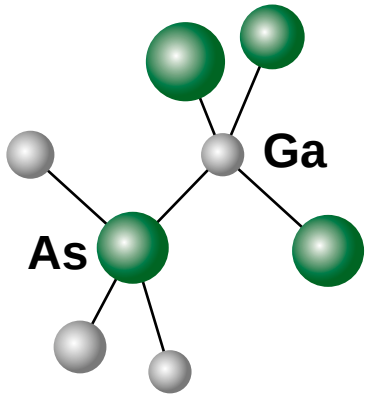


bandstructure

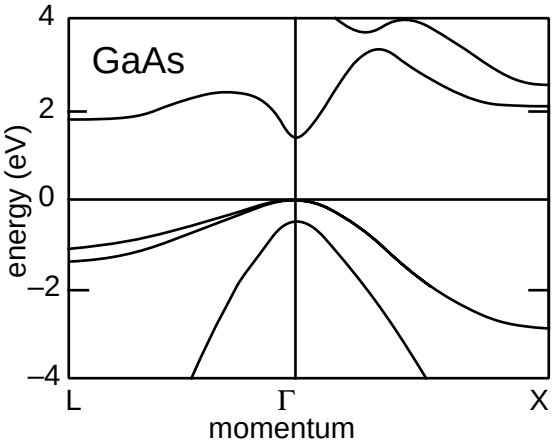


Introduction

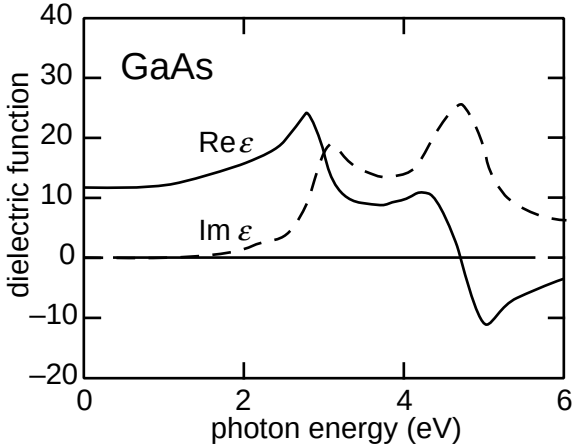
structure



bandstructure

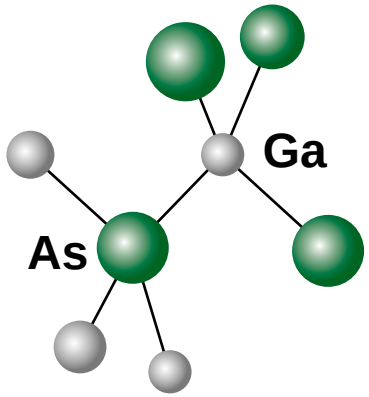


dielectric function

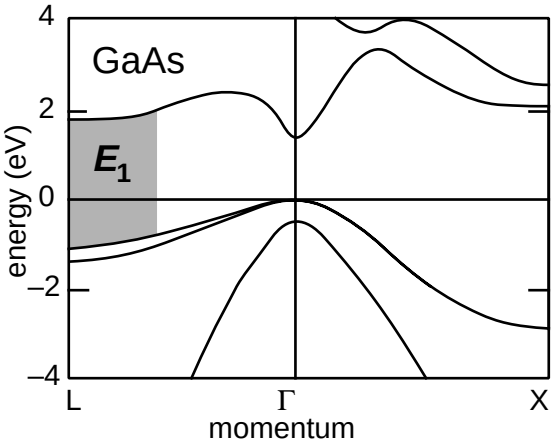


Introduction

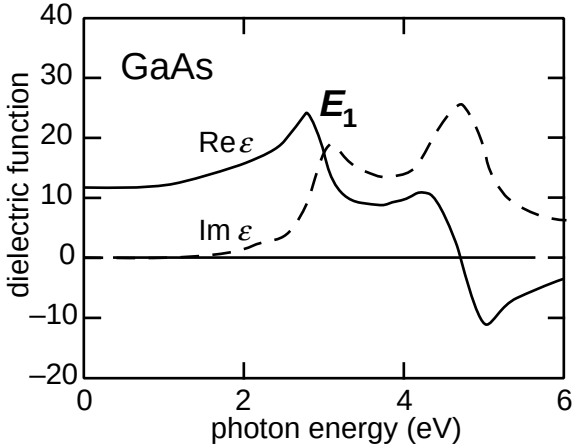
structure



bandstructure

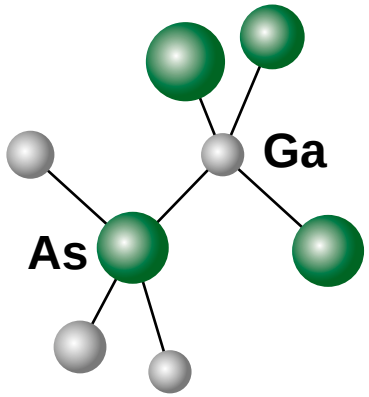


dielectric function

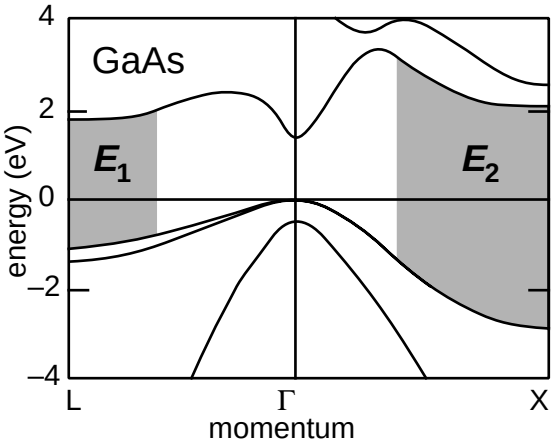


Introduction

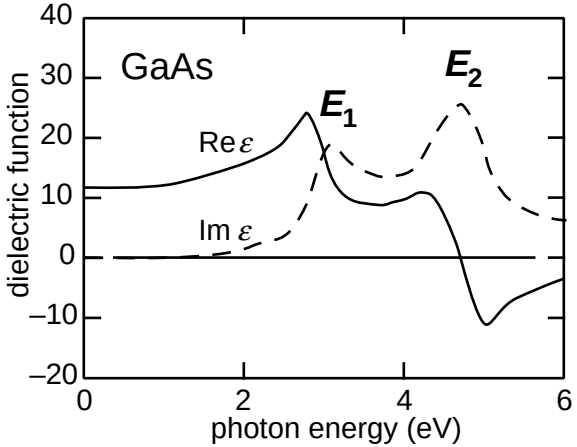
structure



bandstructure

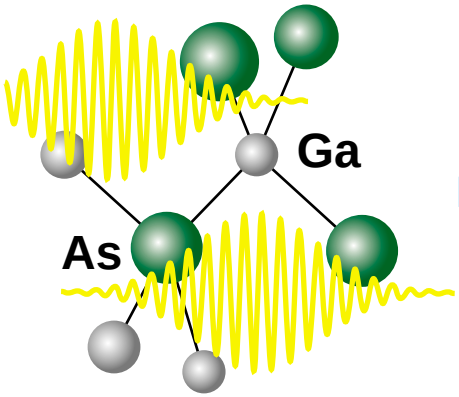


dielectric function



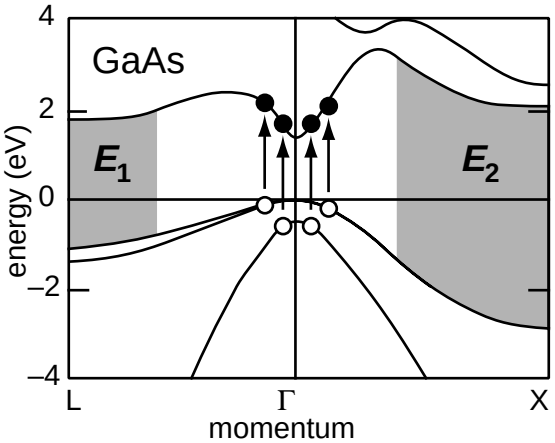
Introduction

structure



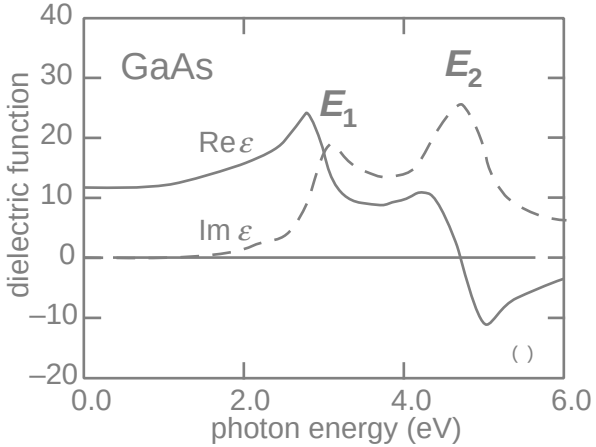
70 fs, 10^{16} W/m^2

bandstructure



10^{20} – 10^{22} cm^{-3}

dielectric function



Introduction

structure

?

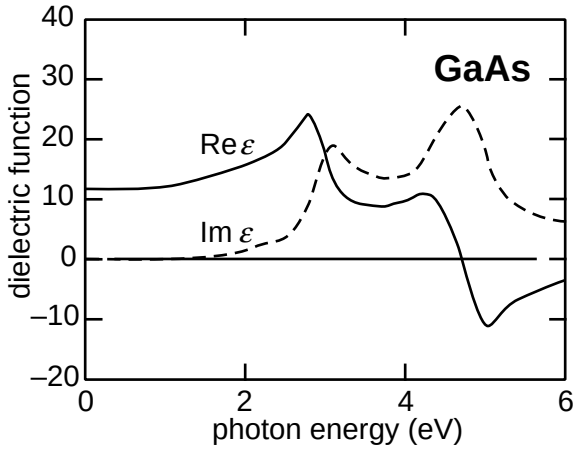
bandstructure

?

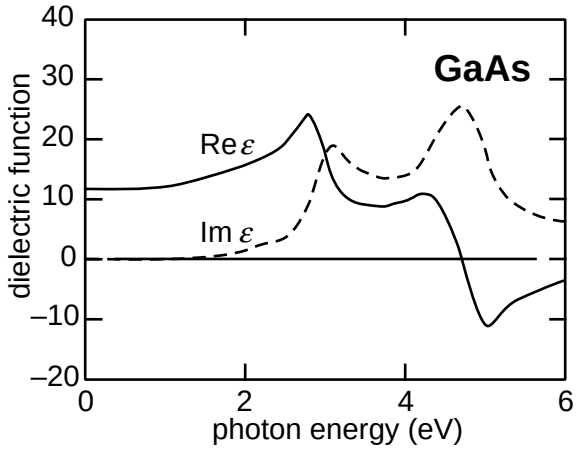
dielectric function

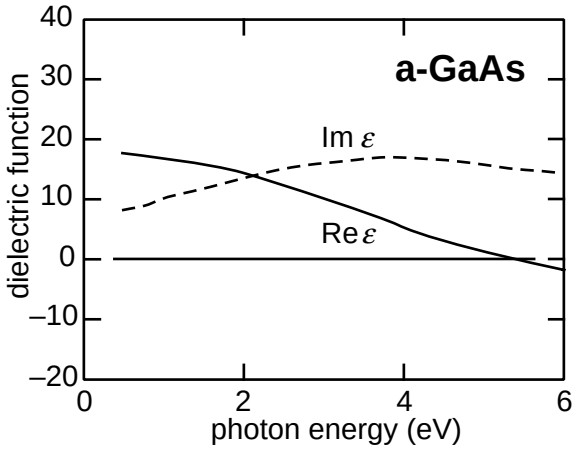
?

Introduction

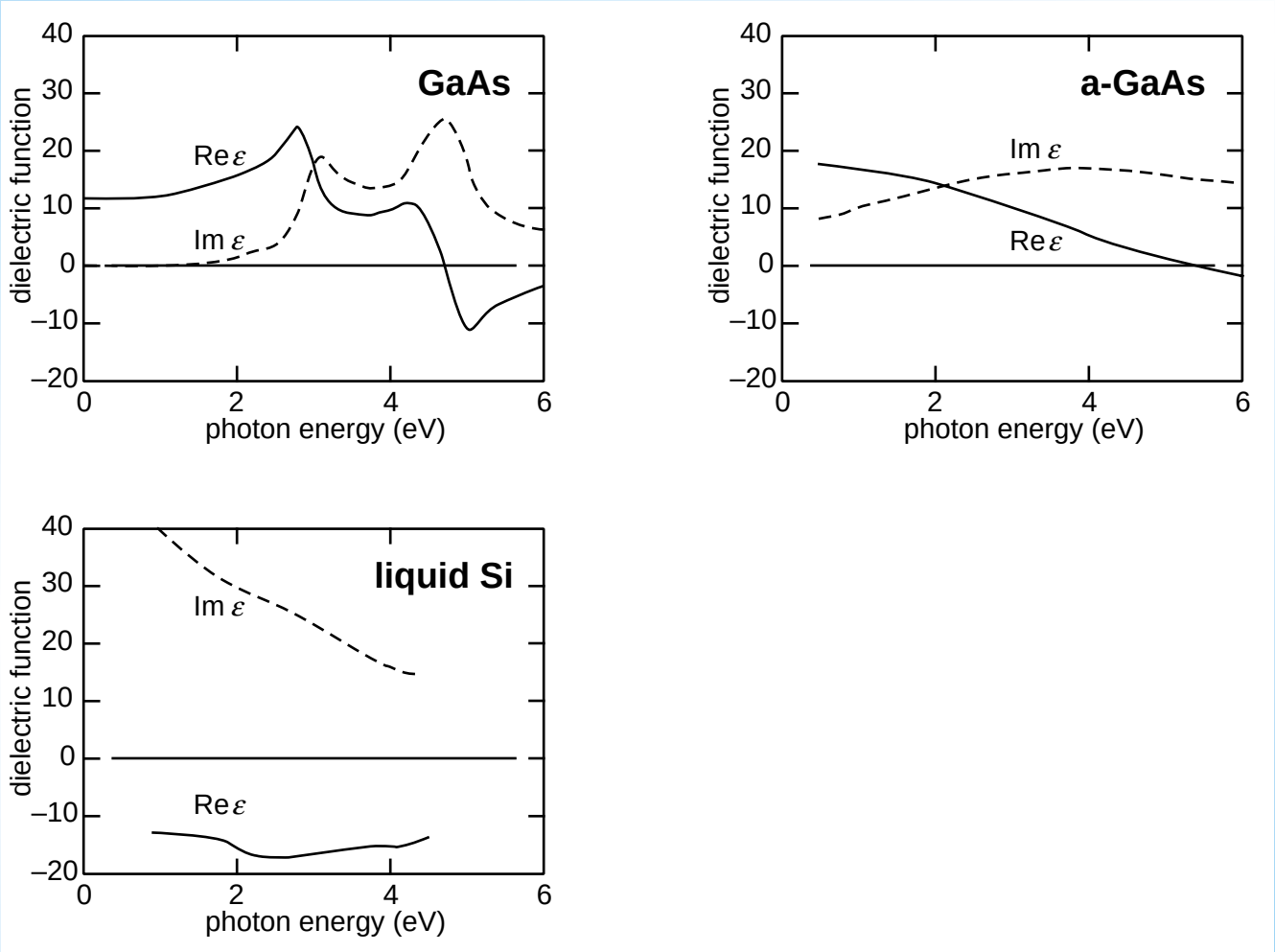


Introduction

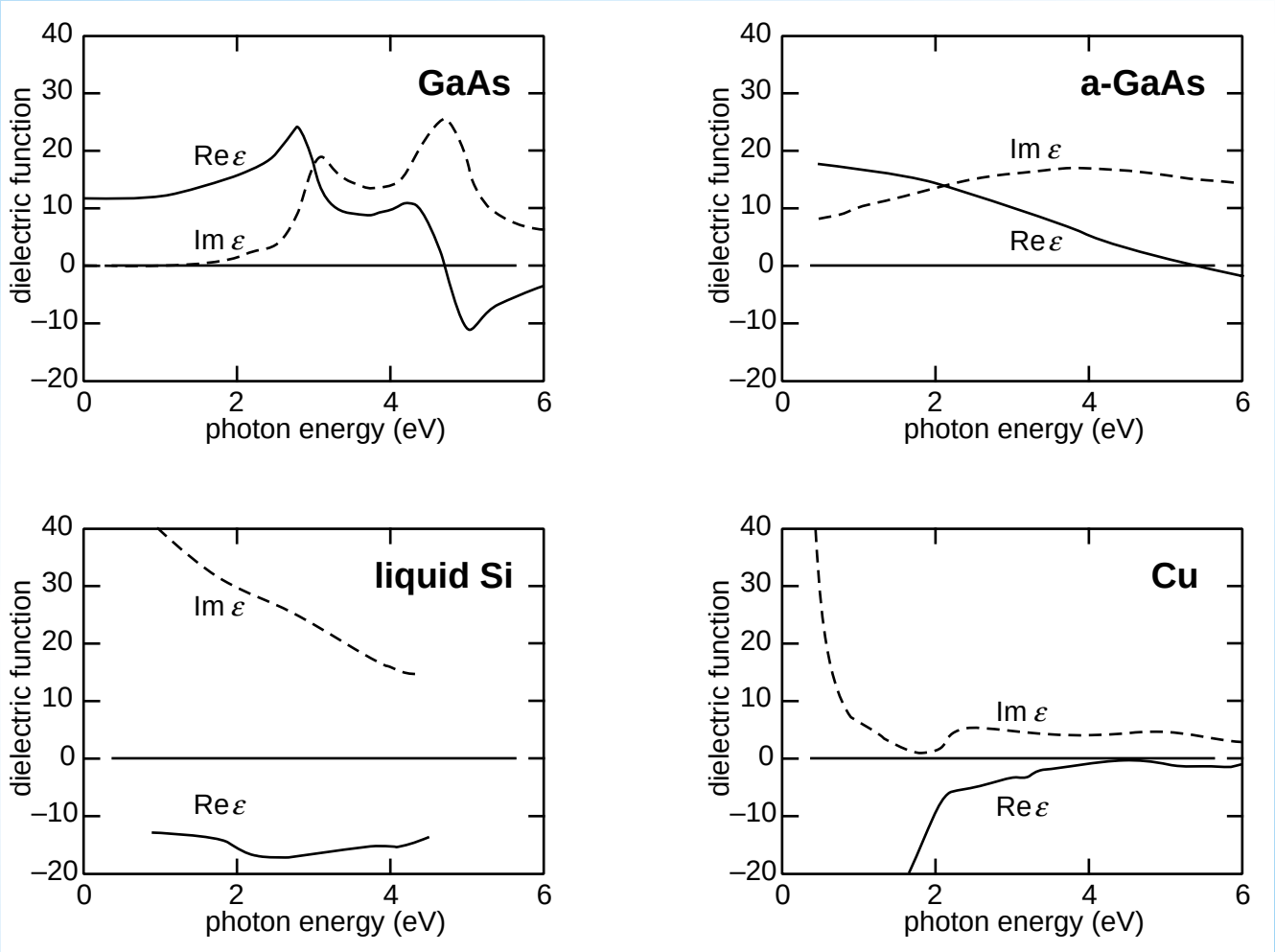


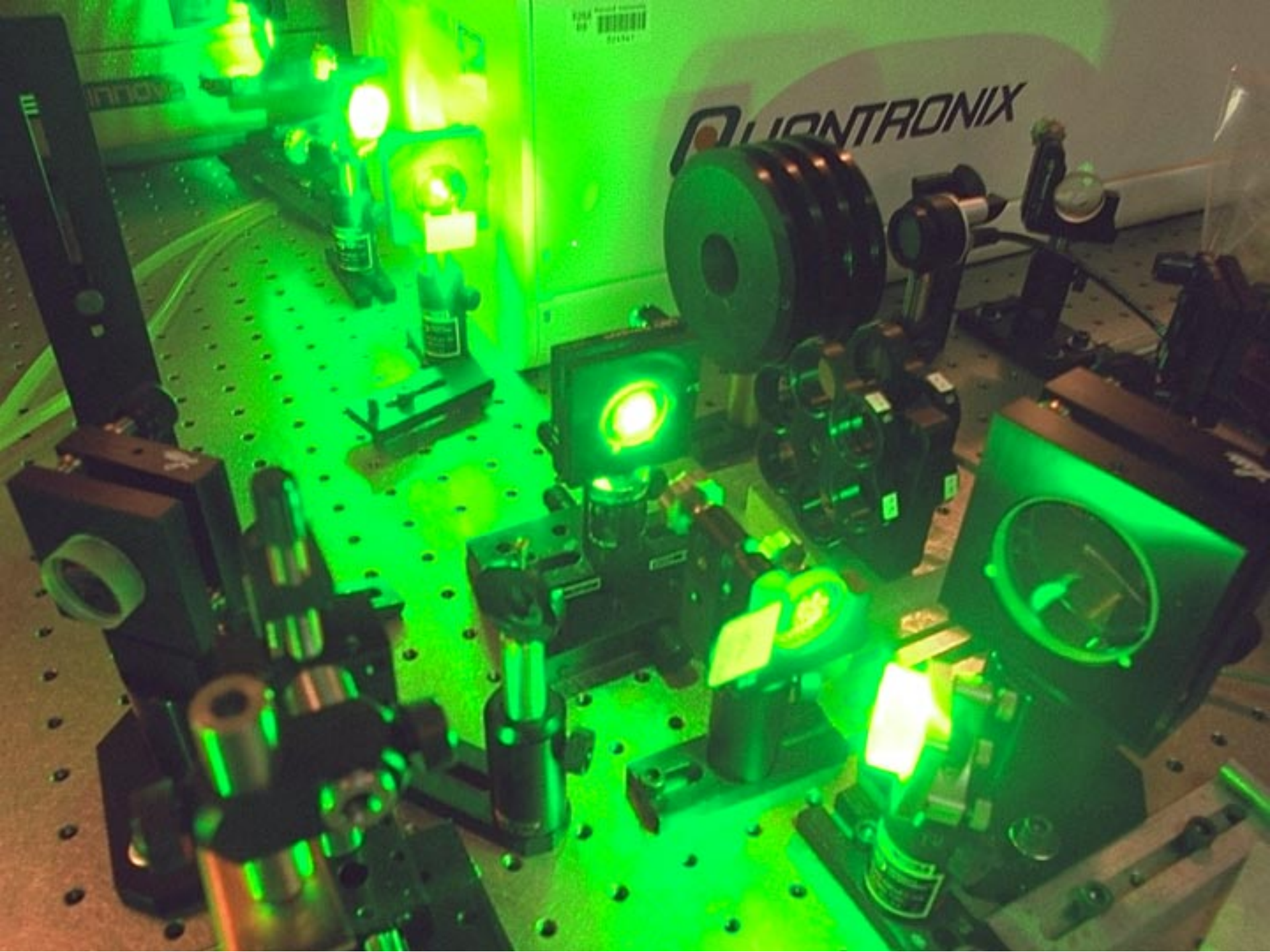


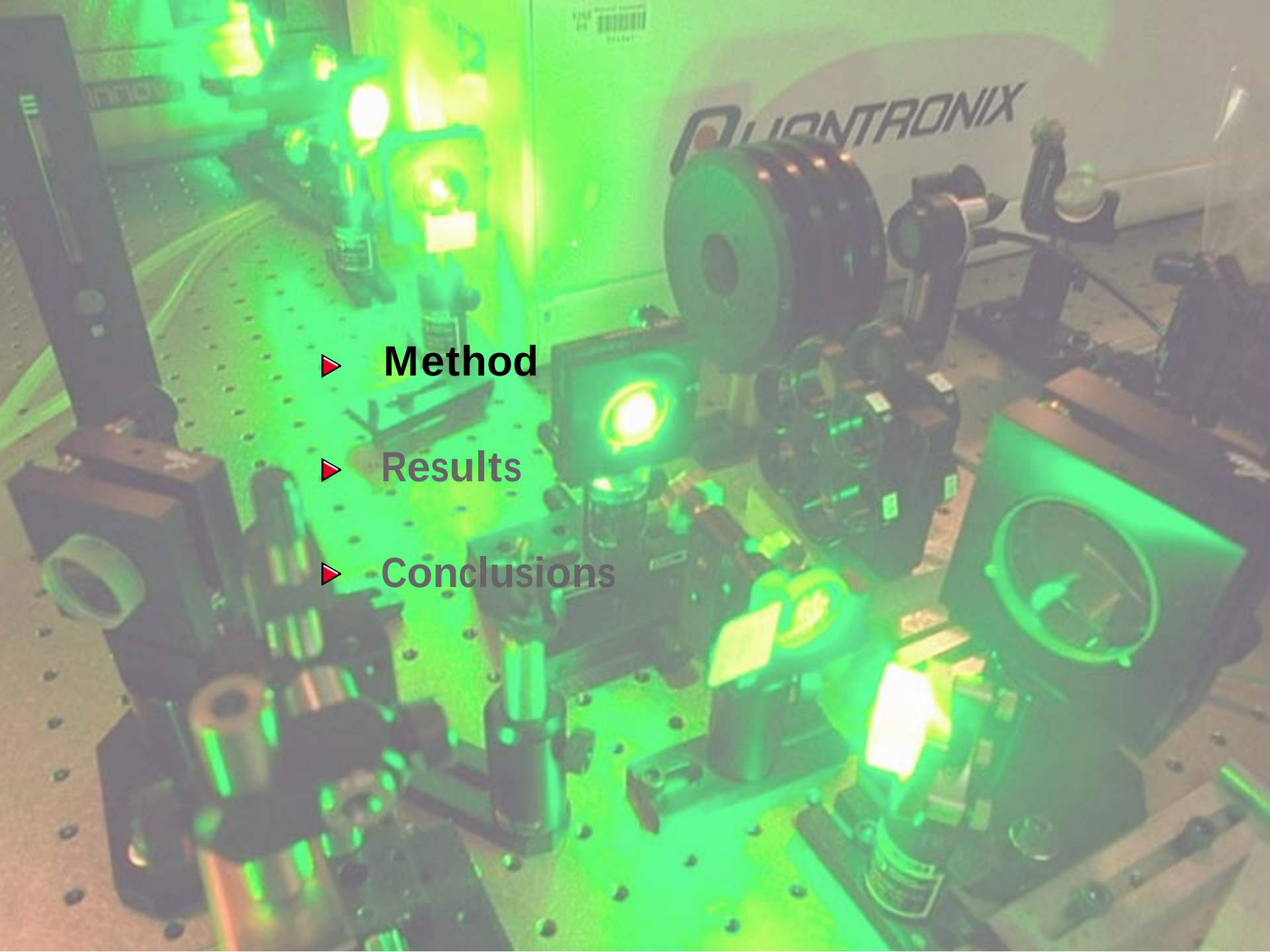
Introduction



Introduction

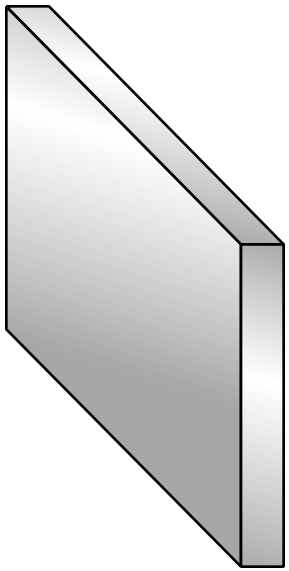



QUANTRONIX



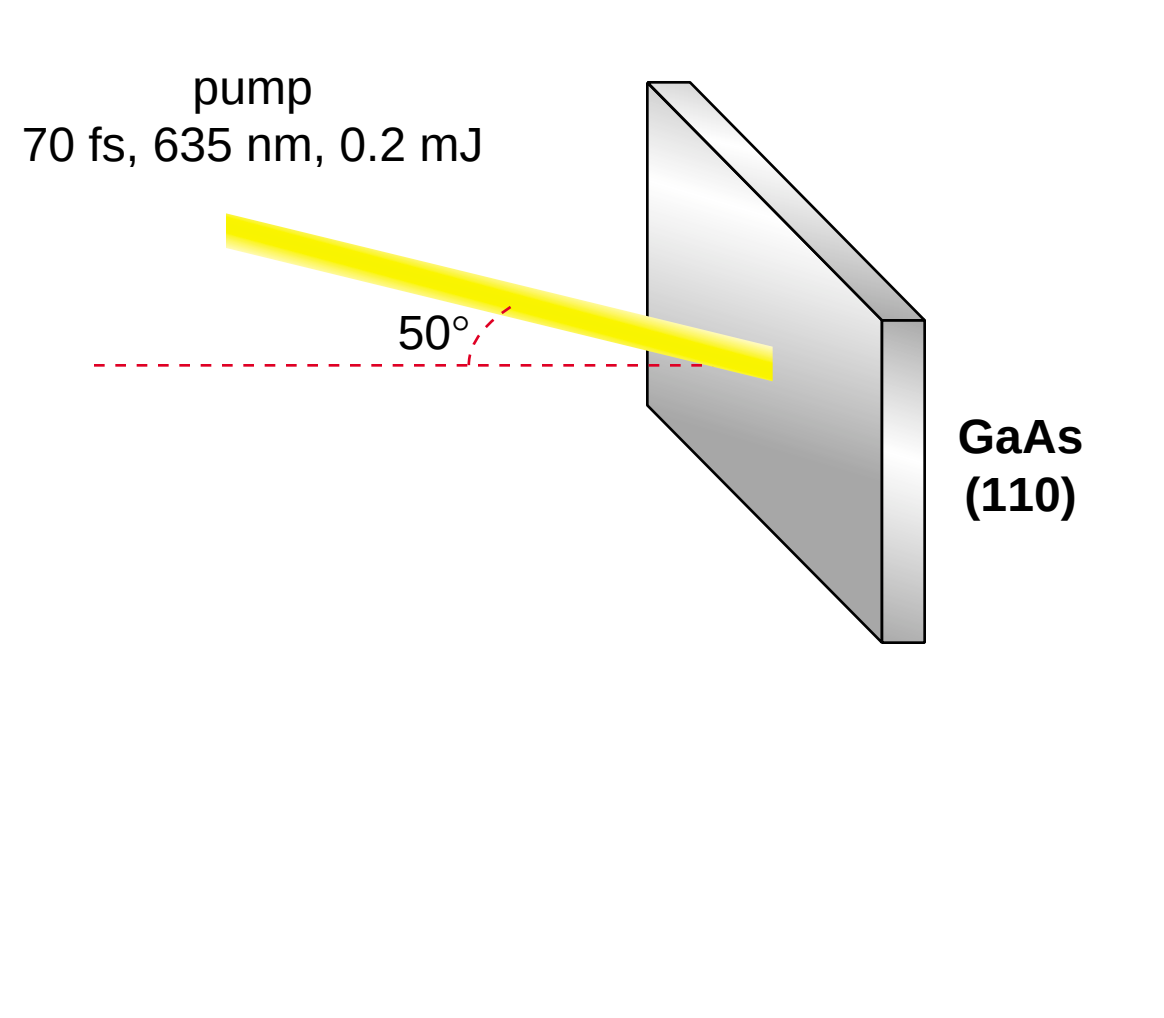
- **Method**
- Results
- Conclusions

Method

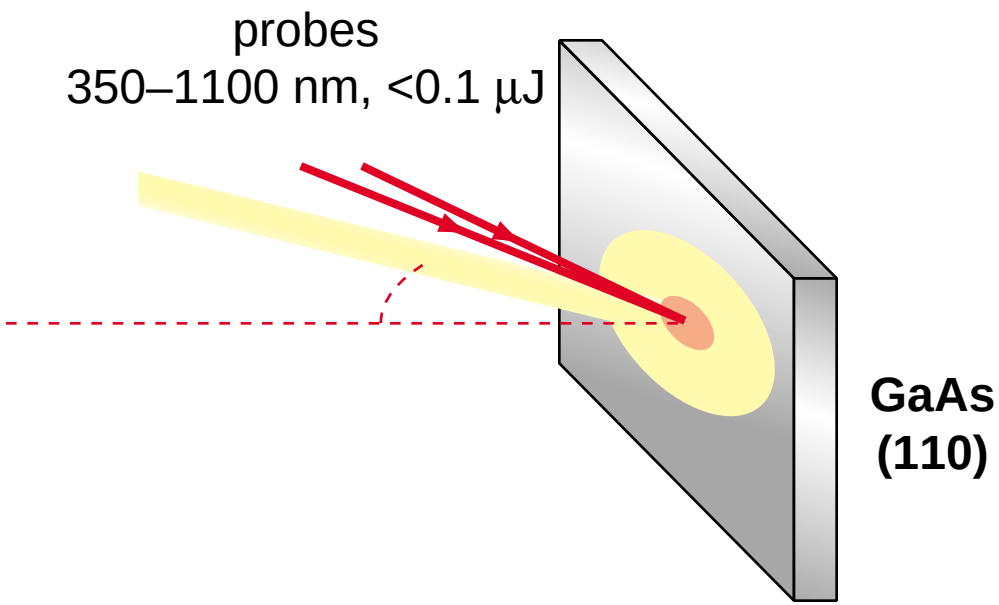


GaAs
(110)

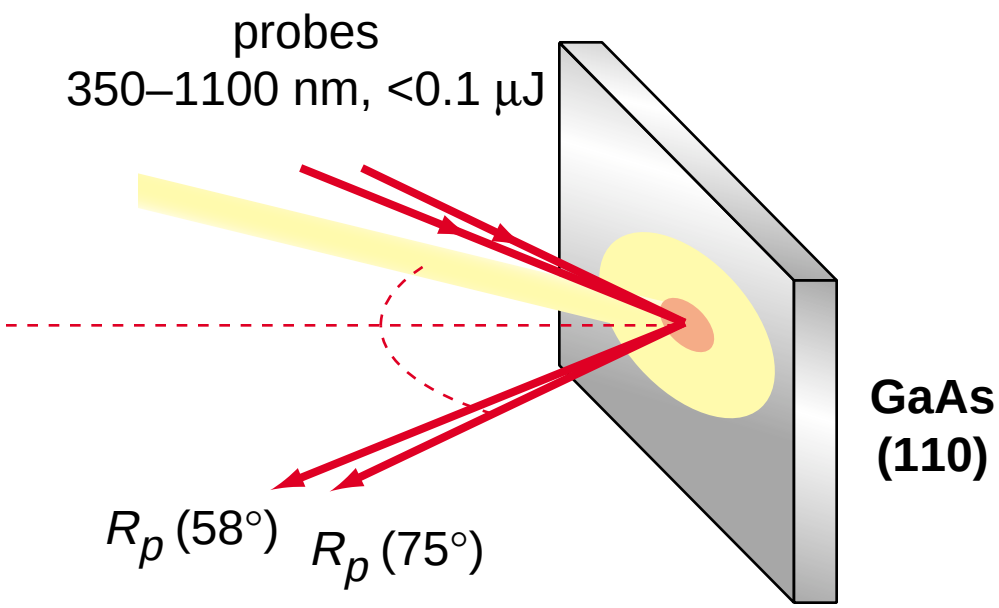
Method



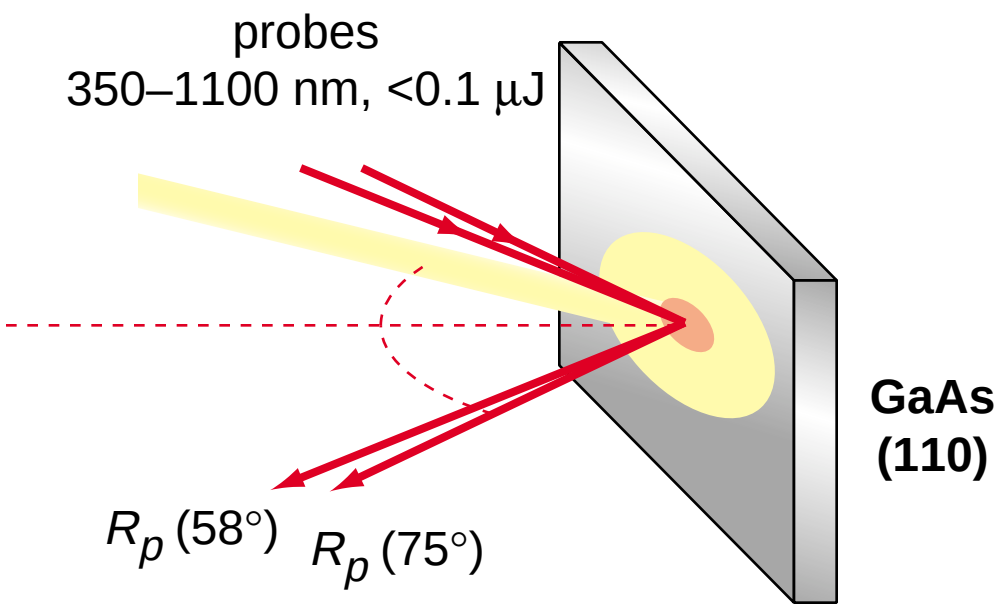
Method



Method

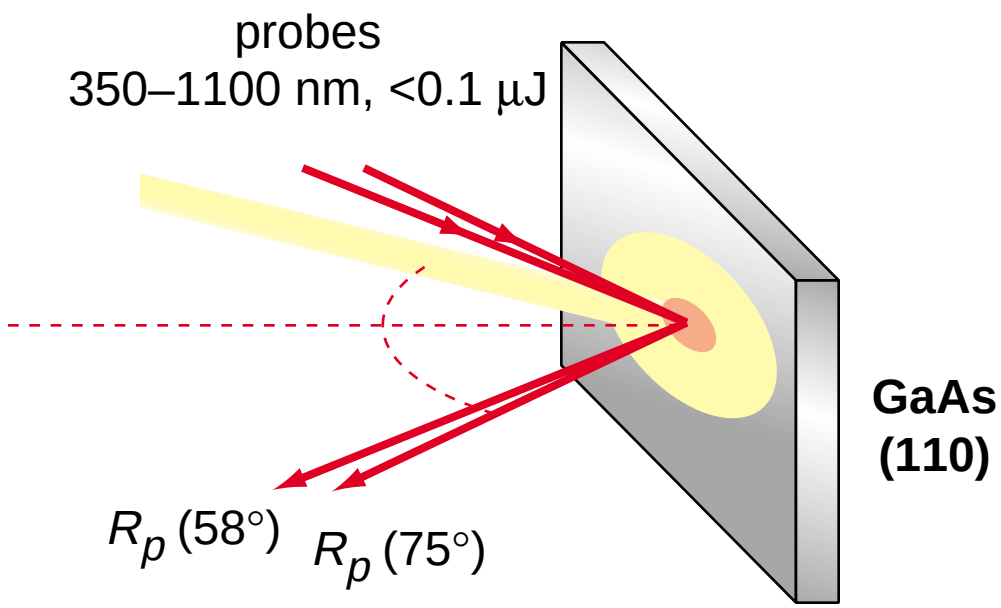


Method

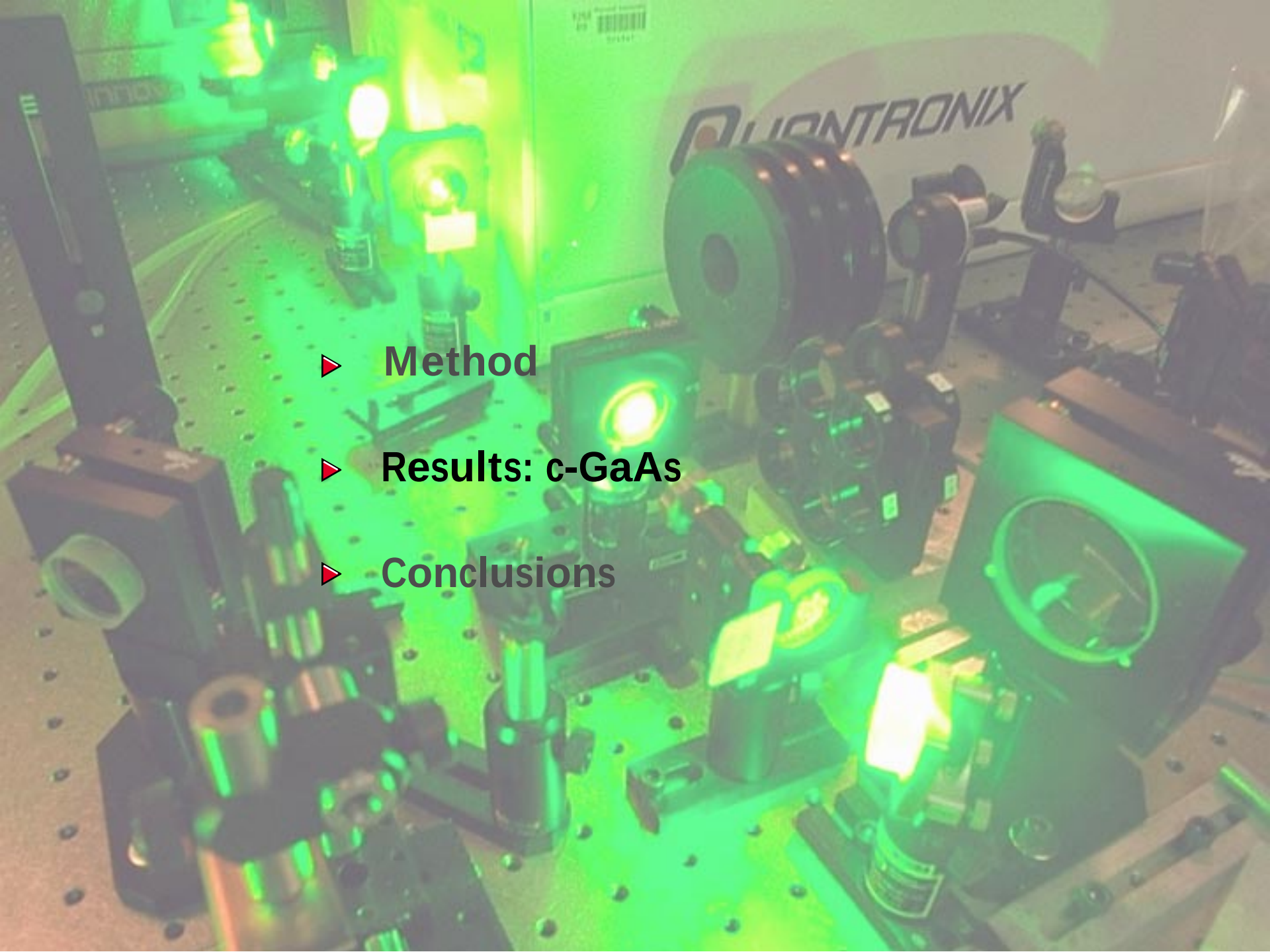


Fresnel
equations

Method

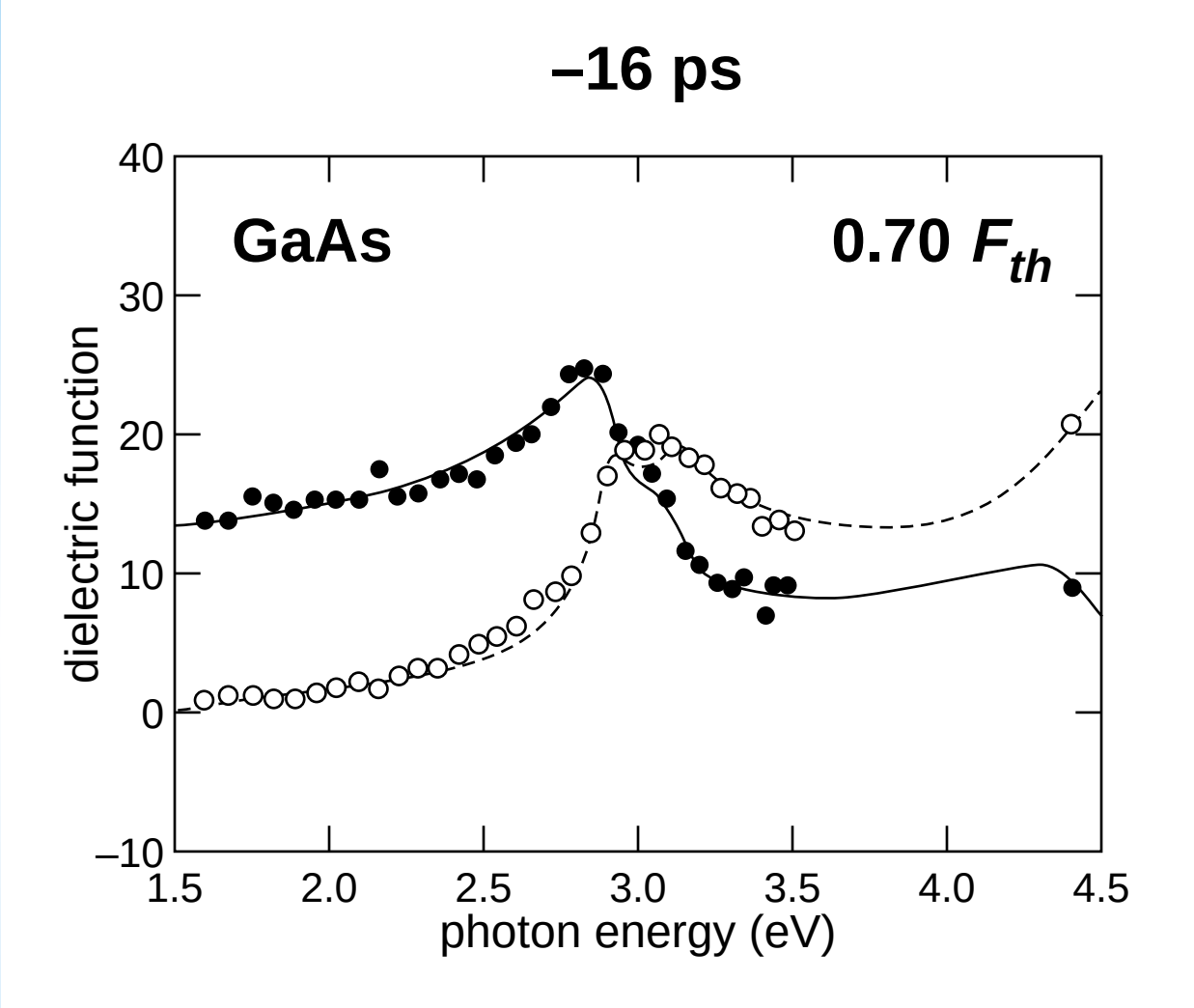


Fresnel equations → Re ε(ω), Im ε(ω)

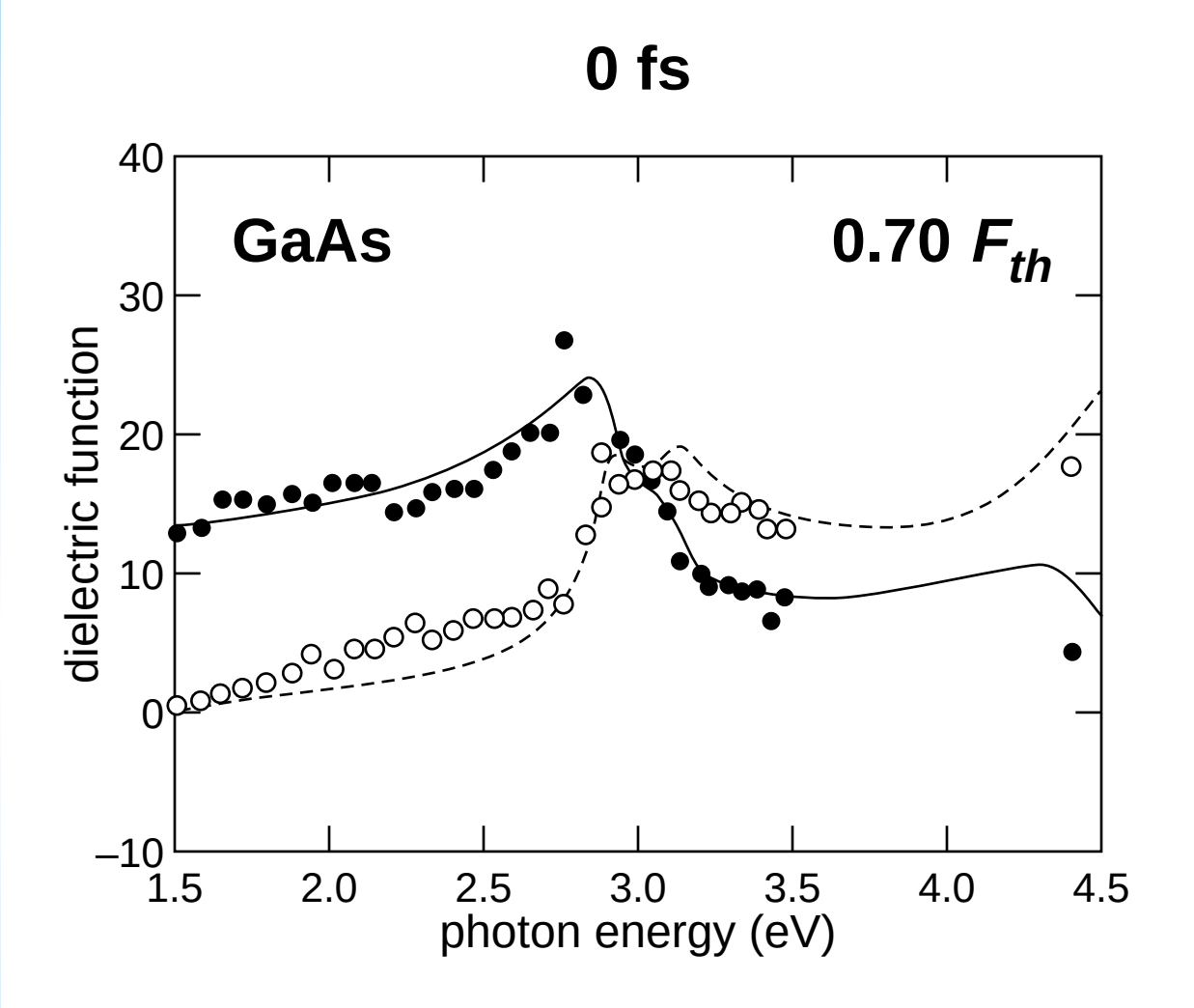


- Method
- **Results: c-GaAs**
- Conclusions

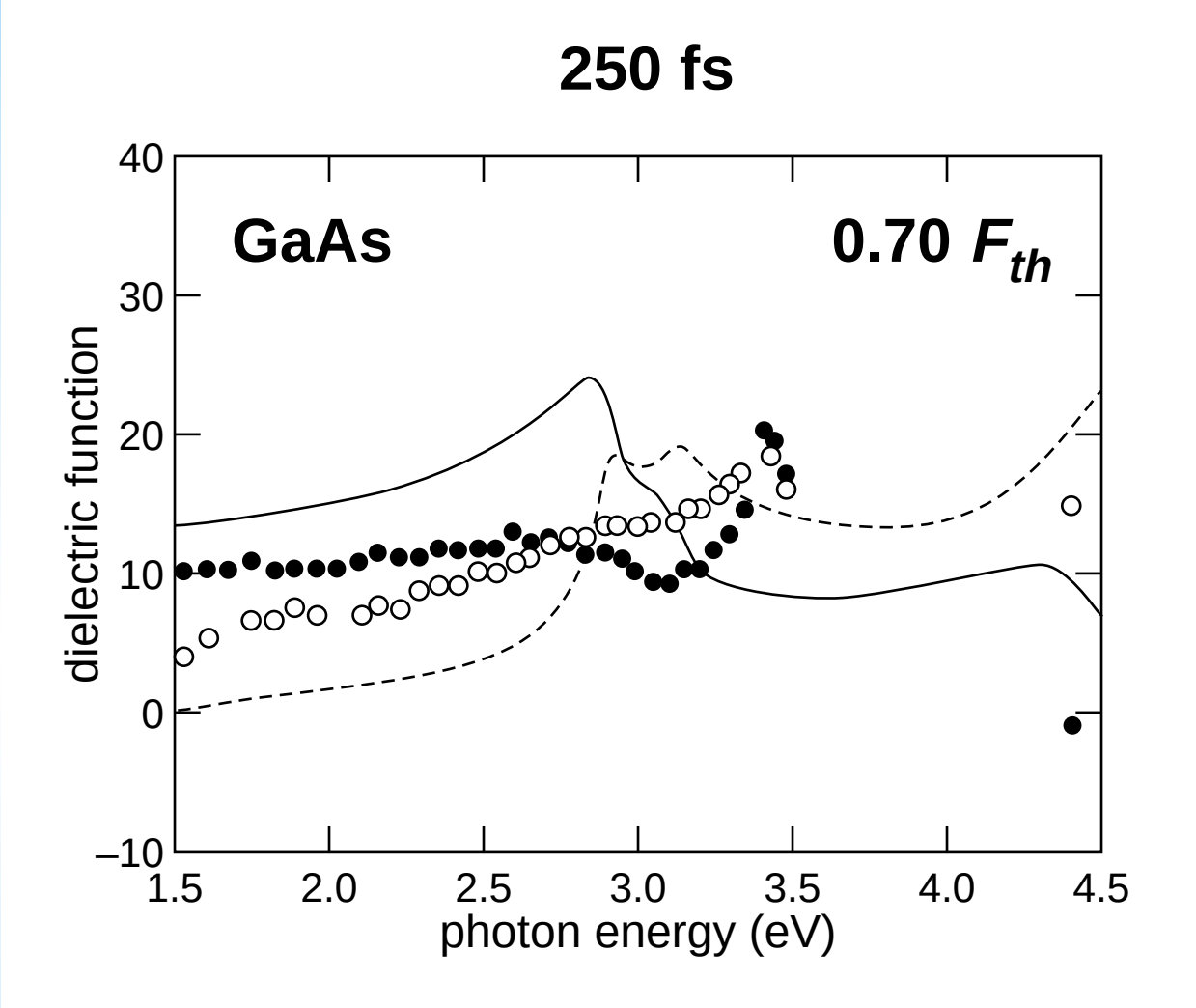
Results


–16 ps
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

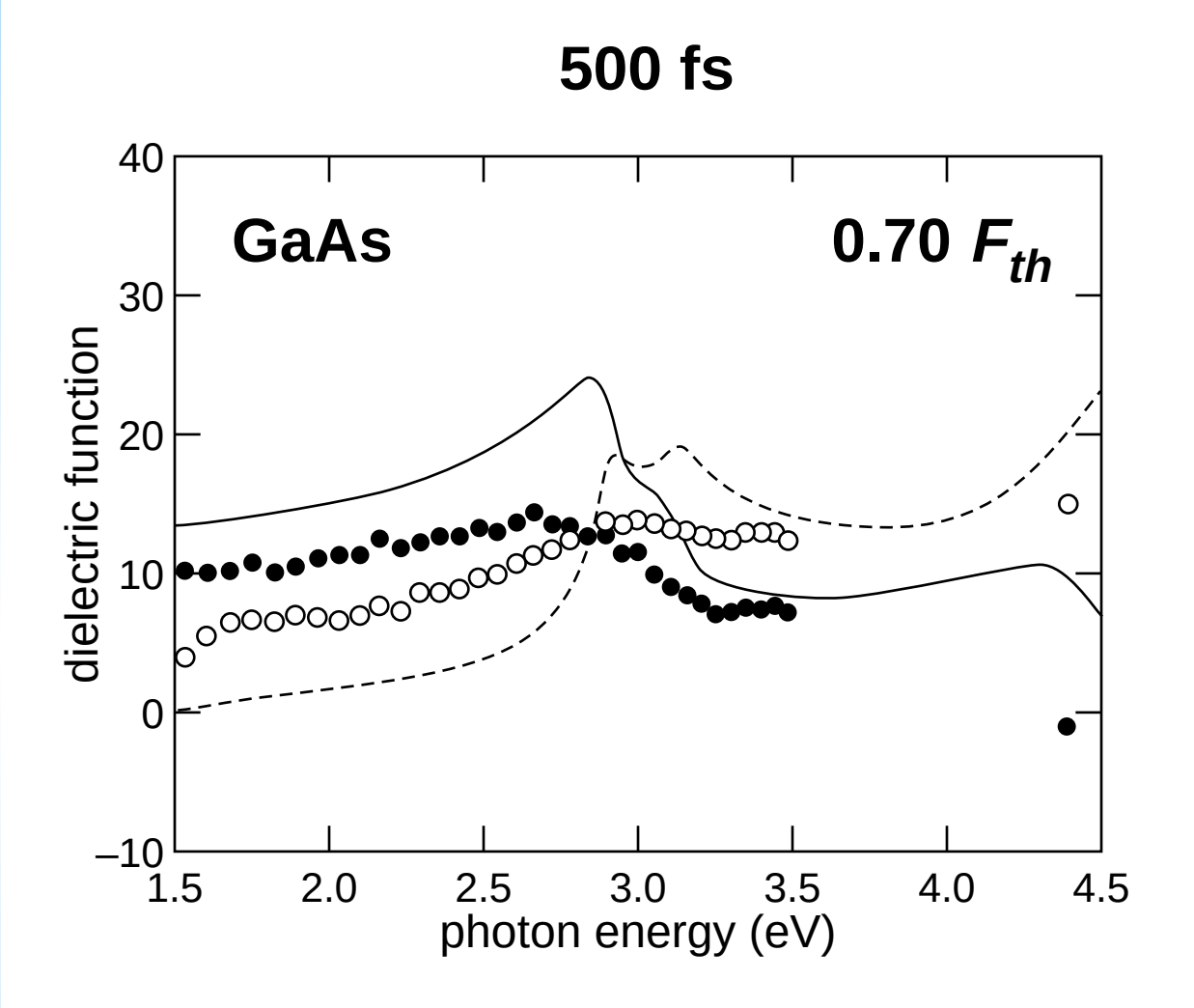
Results


0 fs
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

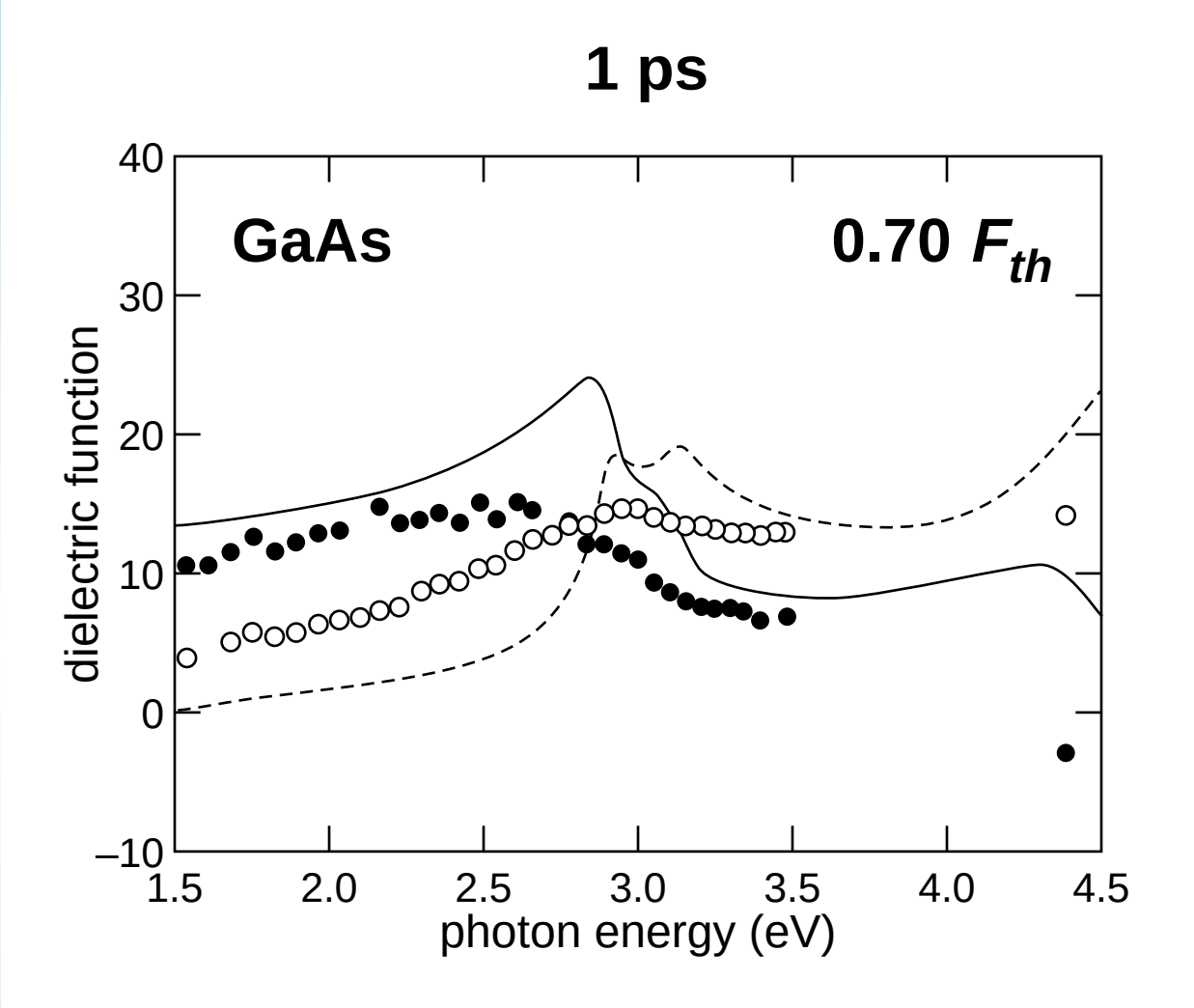
Results


250 fs
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

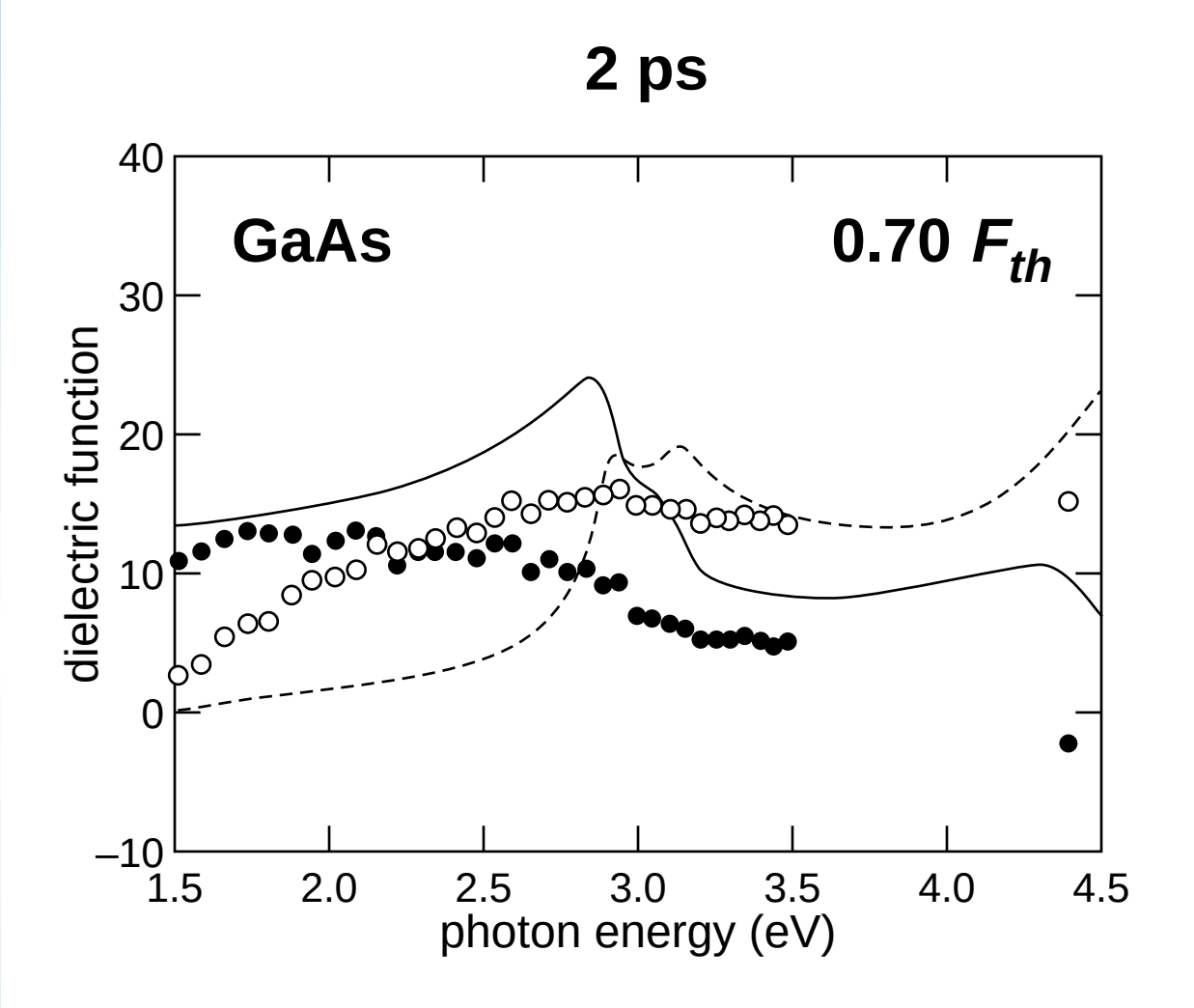
Results


500 fs
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

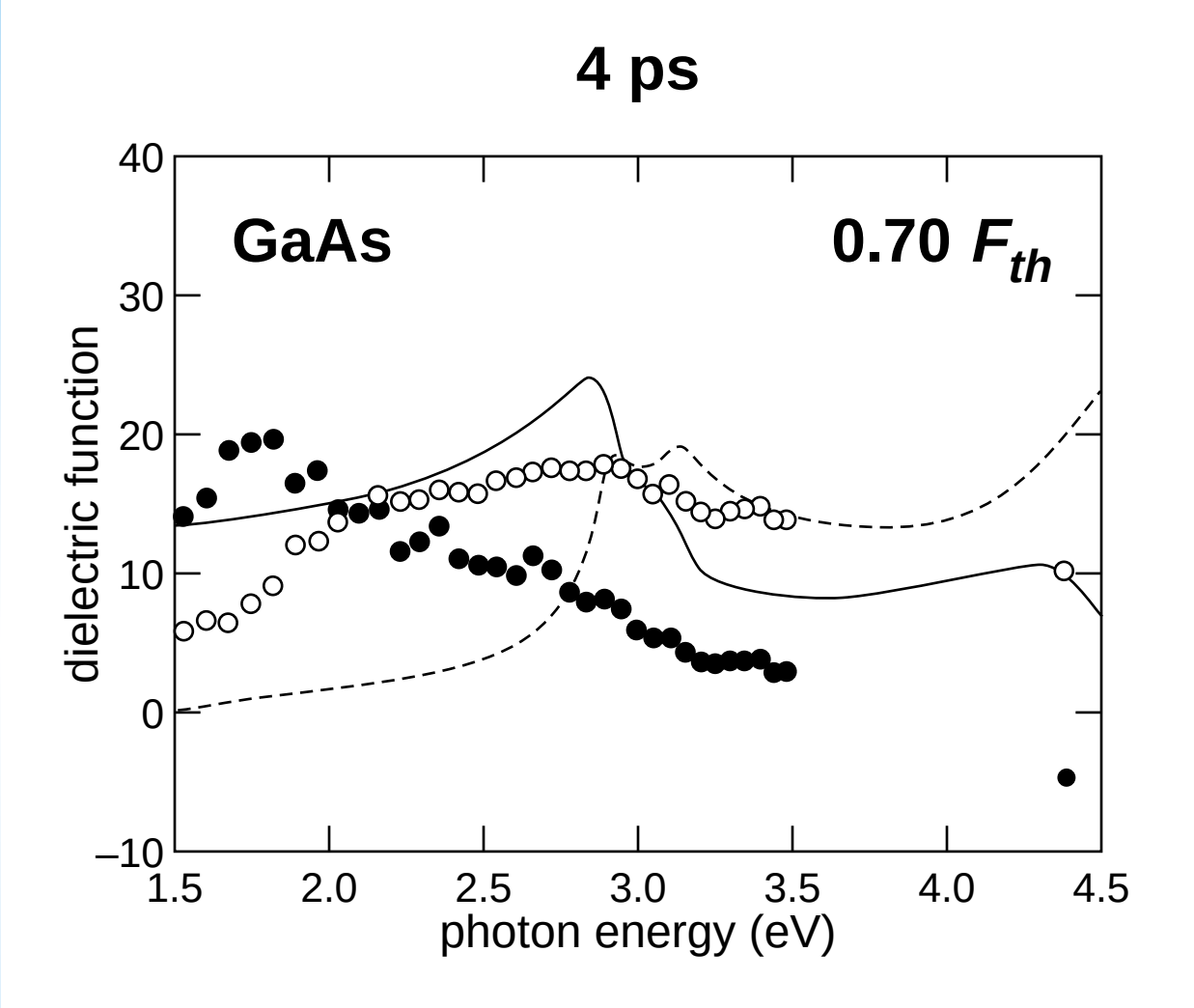
Results


1 ps
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

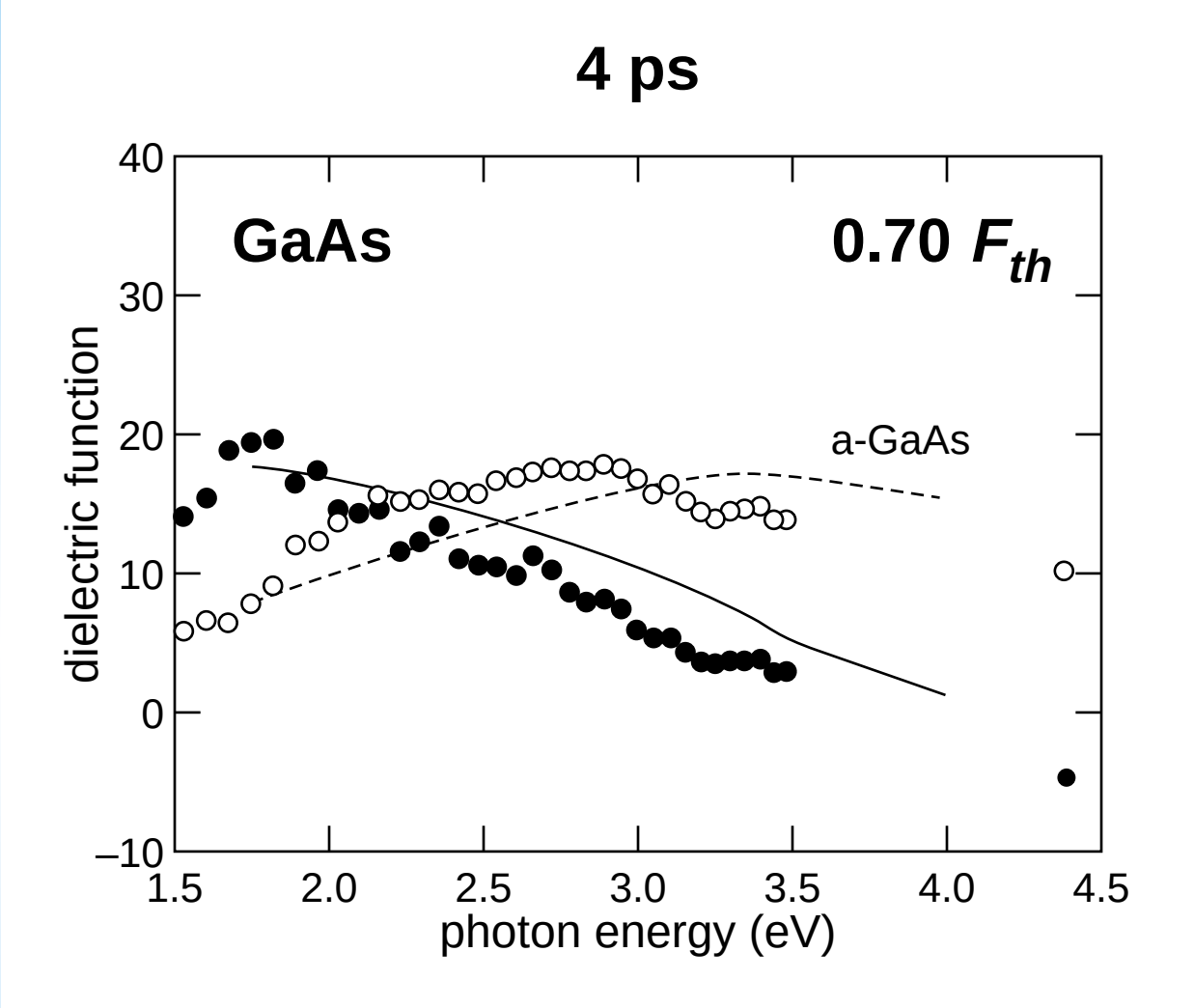
Results


2 ps
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

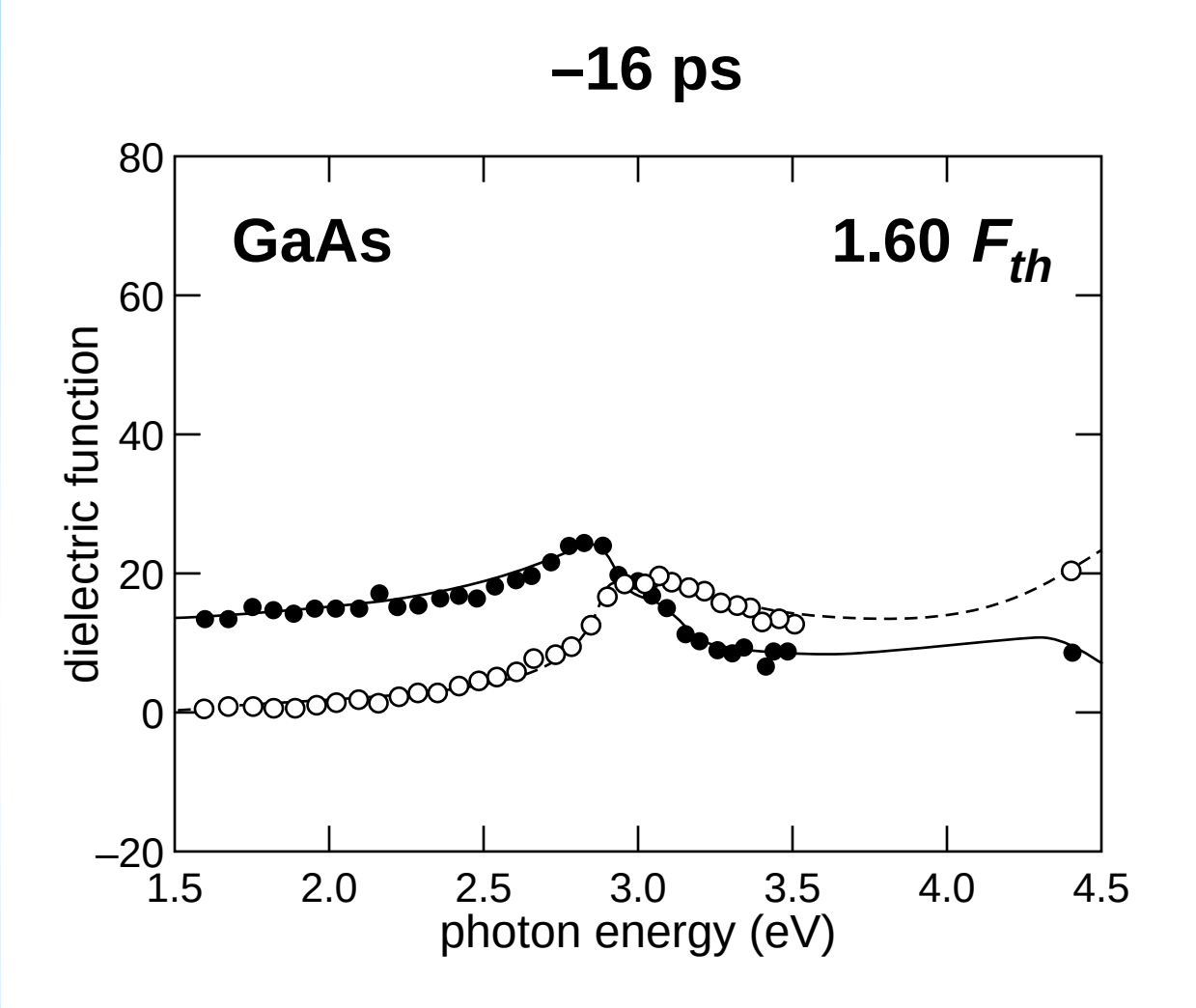
Results


4 ps
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

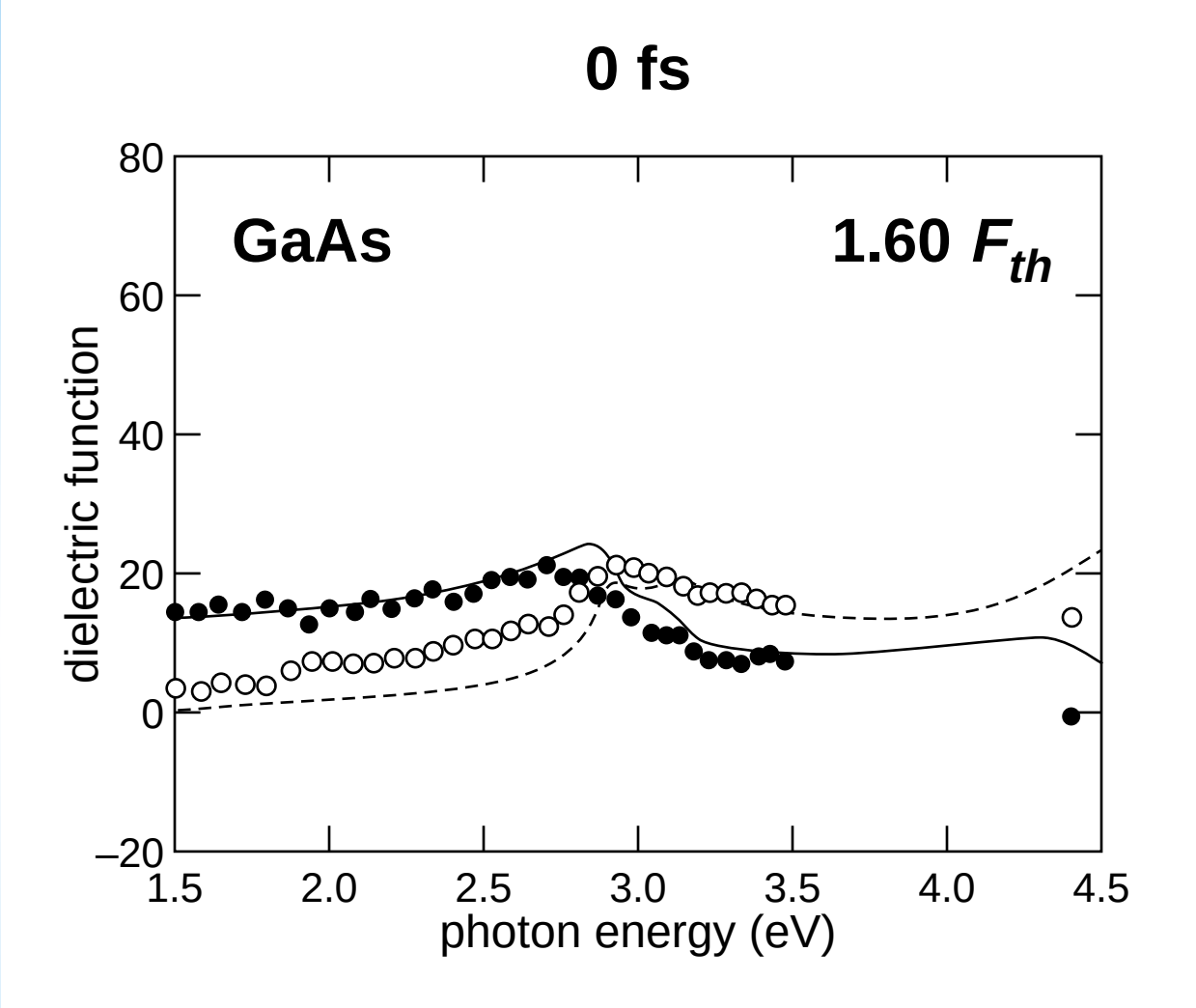
Results


4 ps
GaAs
0.70 F_{th}
a-GaAs
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

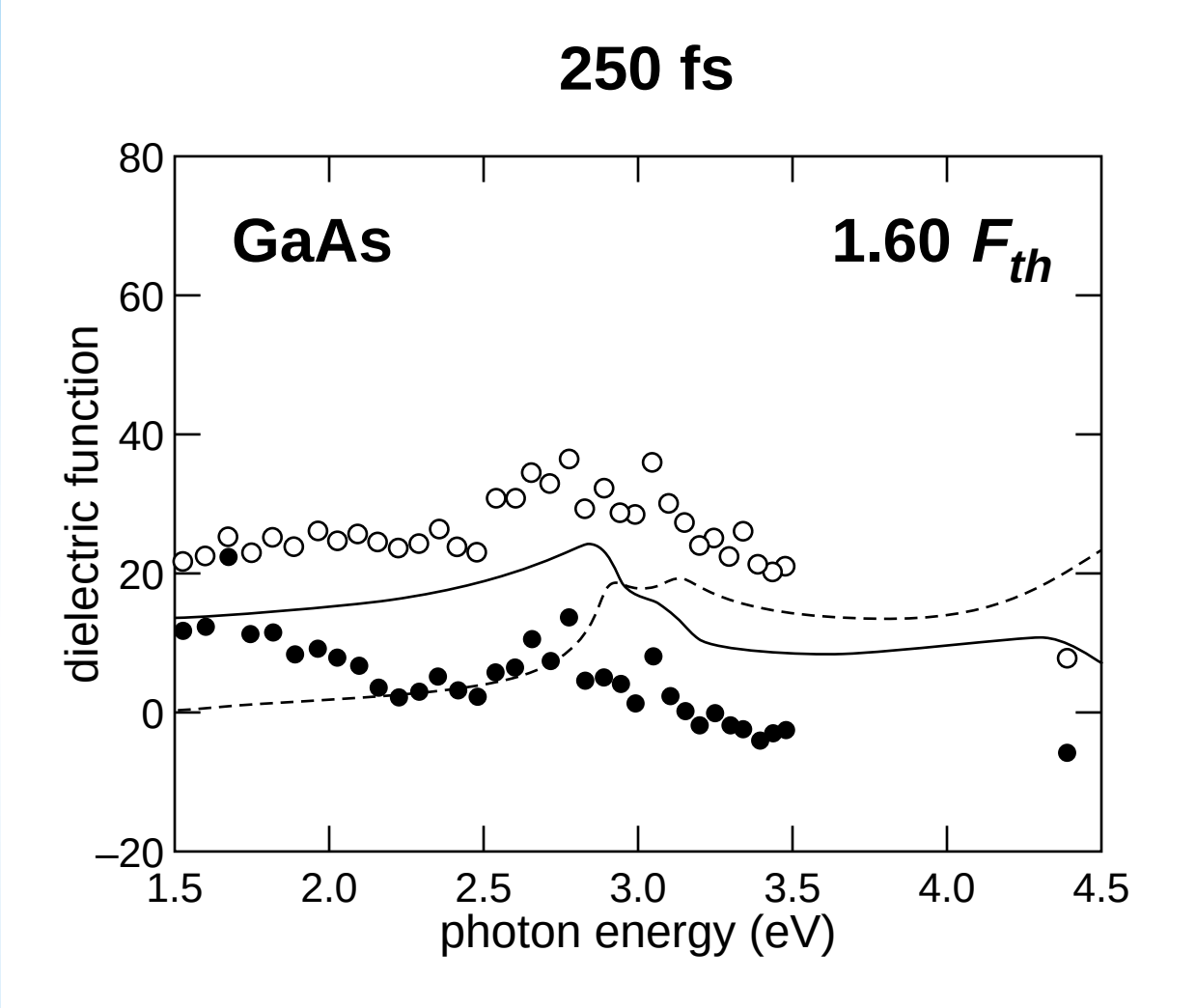
Results


–16 ps
GaAs
1.60 F_{th}
dielectric function
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

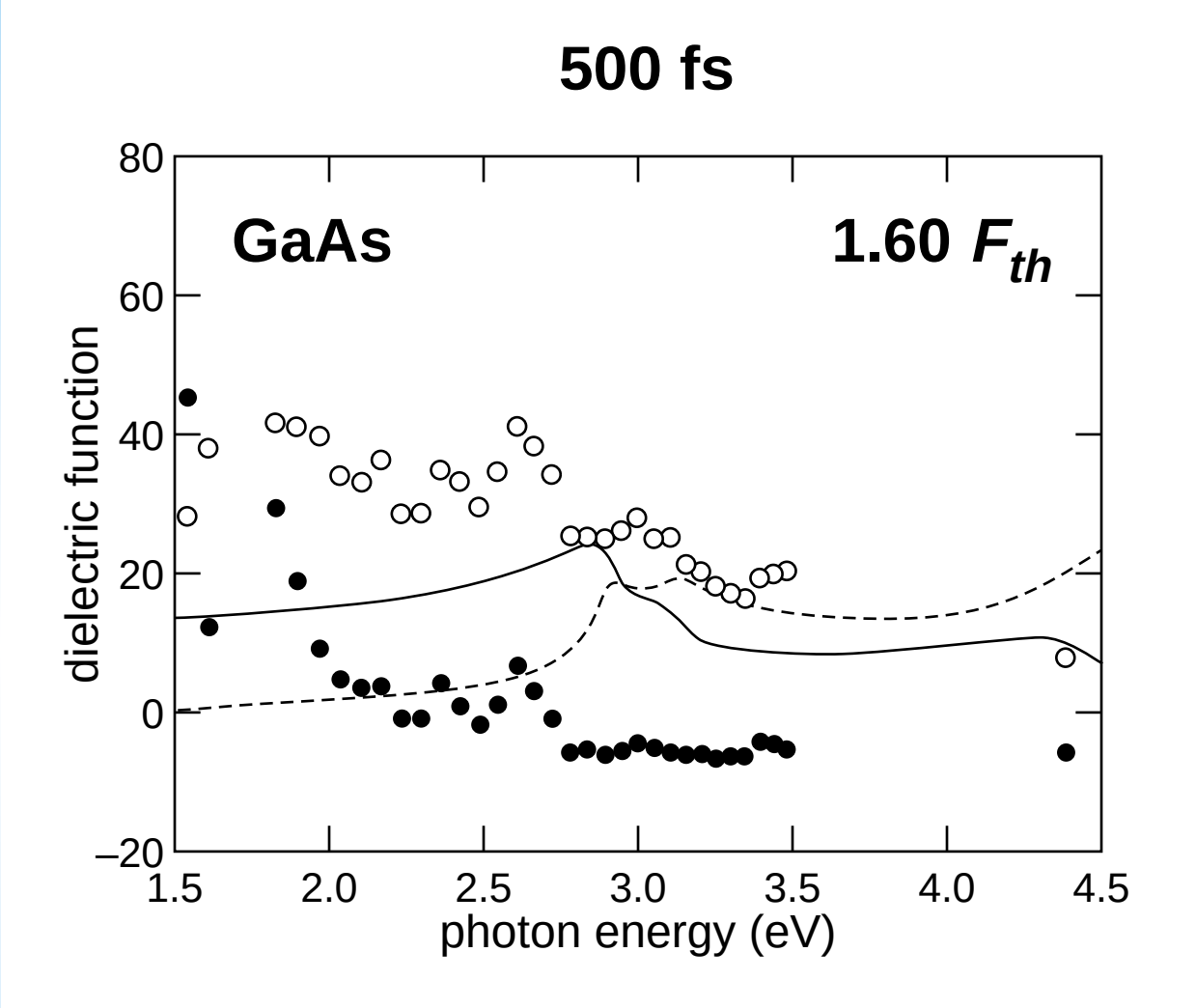
Results


0 fs
GaAs
1.60 F_{th}
dielectric function
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

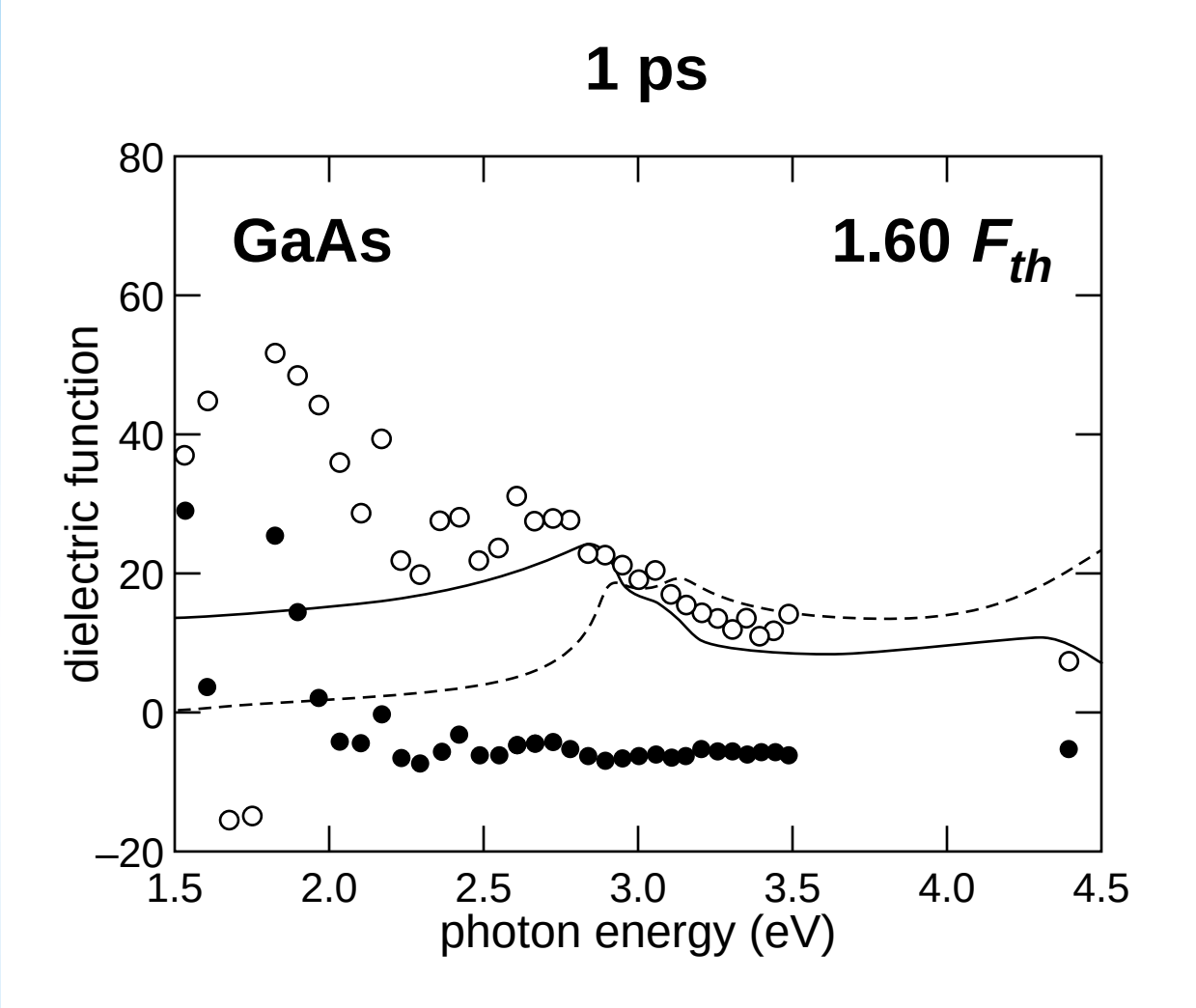
Results


250 fs
GaAs
1.60 F_{th}
dielectric function
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

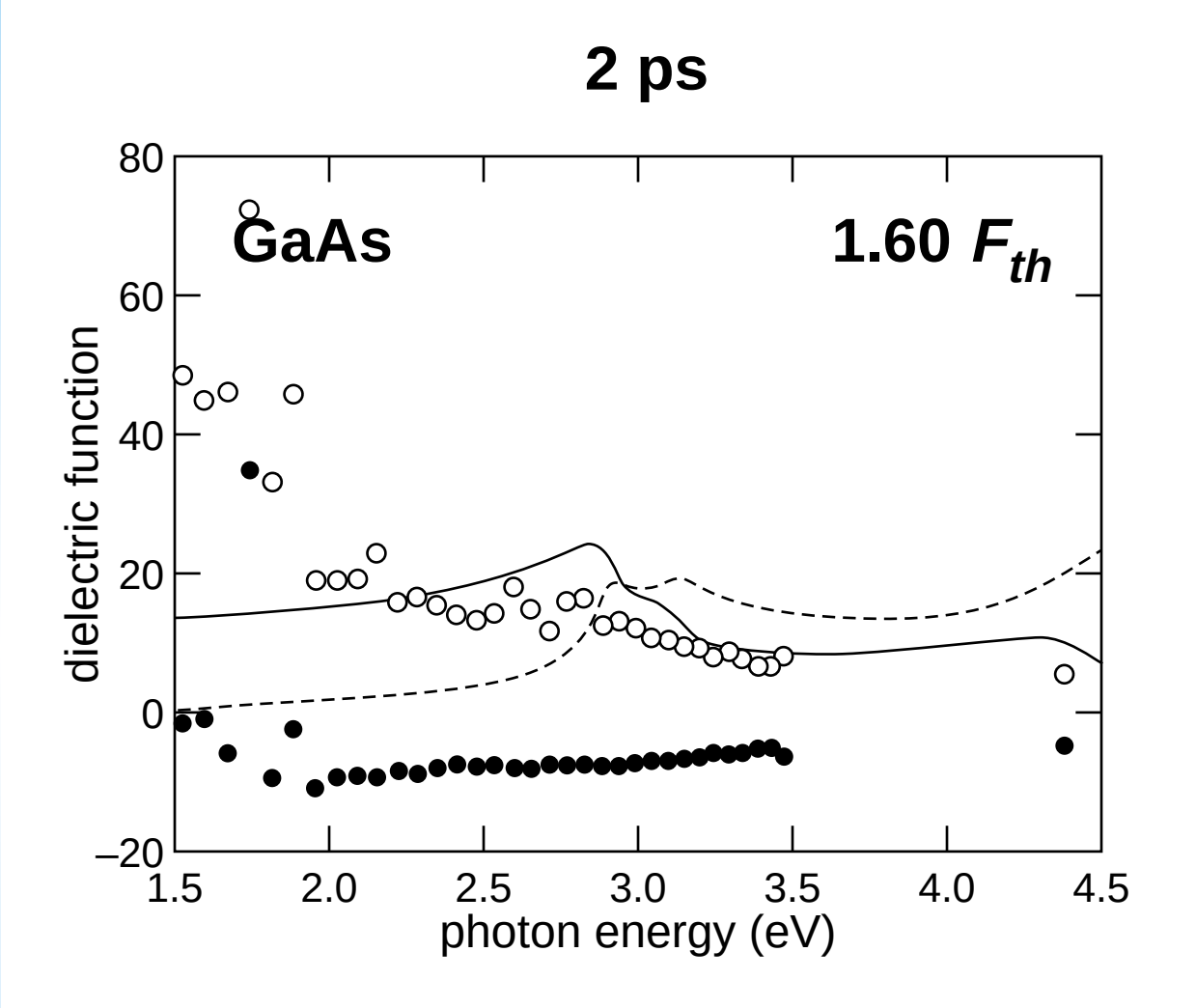
Results


500 fs
GaAs
1.60 F_{th}
dielectric function
photon energy (eV)
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

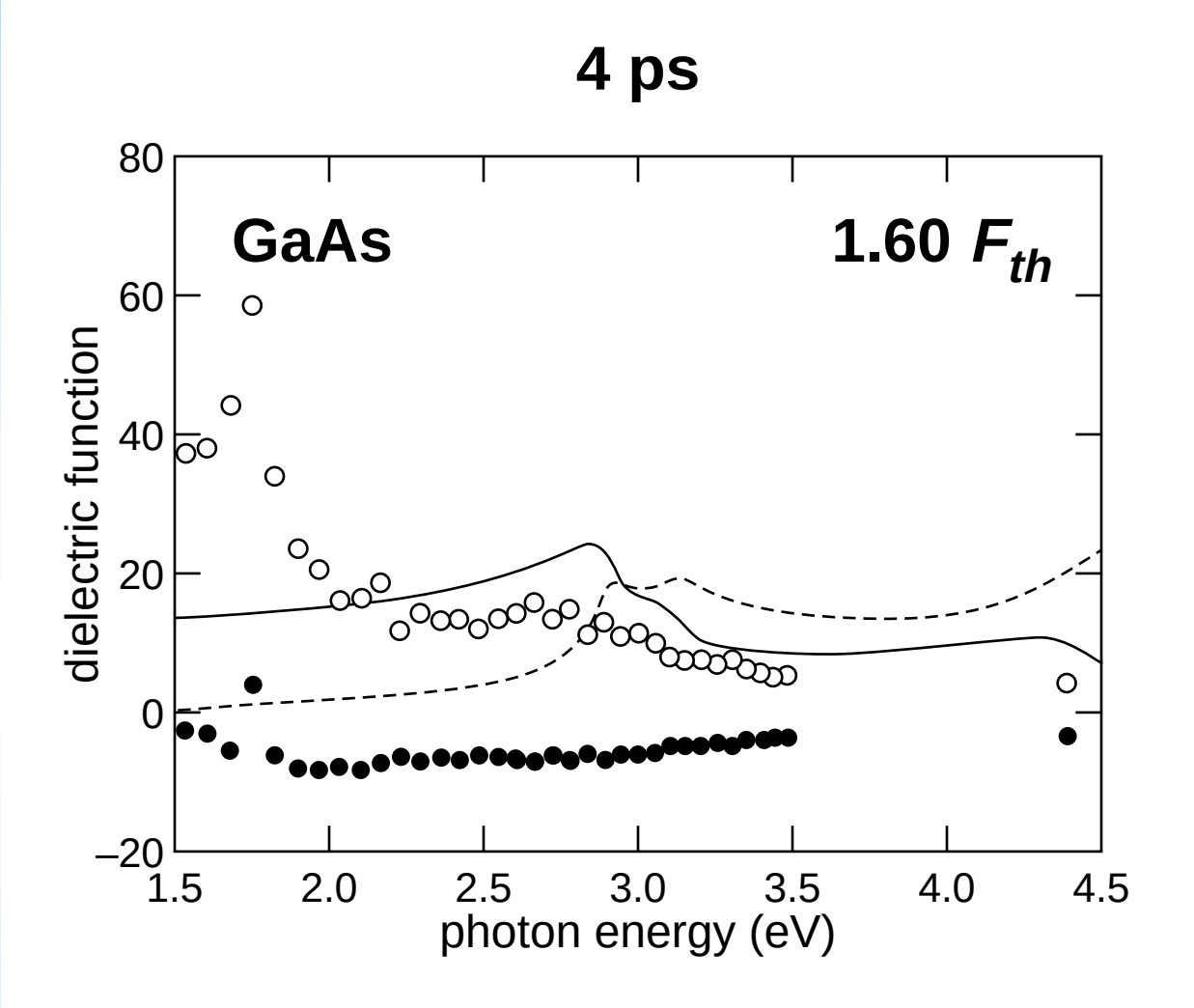
Results


1 ps
GaAs
1.60 F_{th}
dielectric function
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

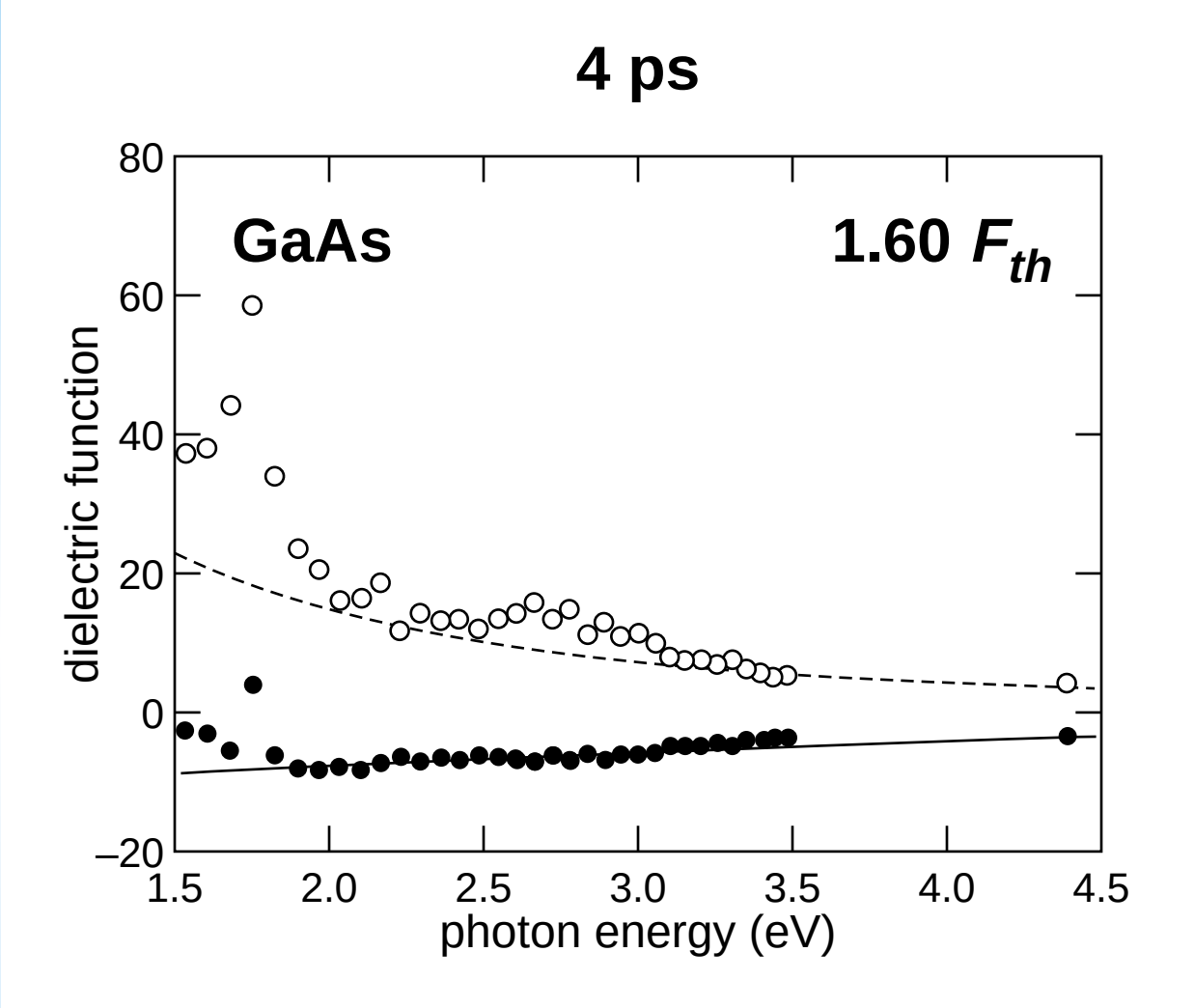
Results


2 ps
GaAs
1.60 F_{th}
dielectric function
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

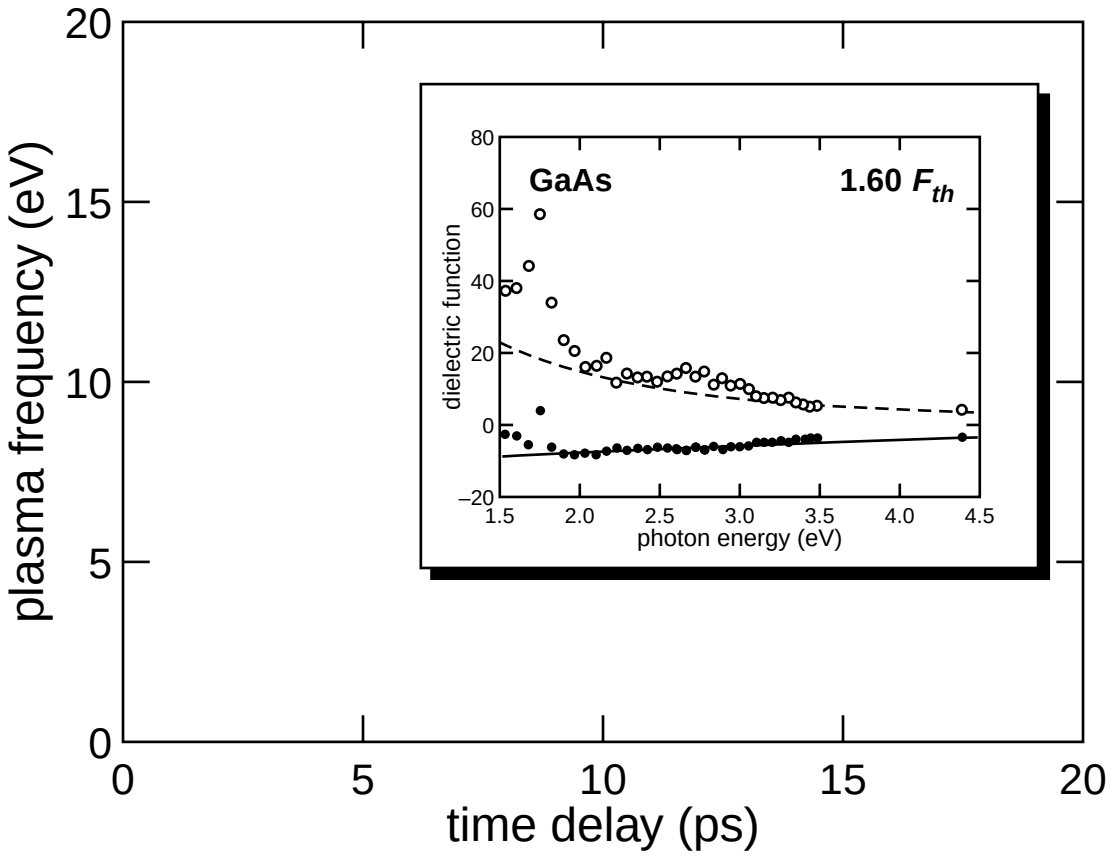
Results


4 ps
GaAs
1.60 F_{th}
dielectric function
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

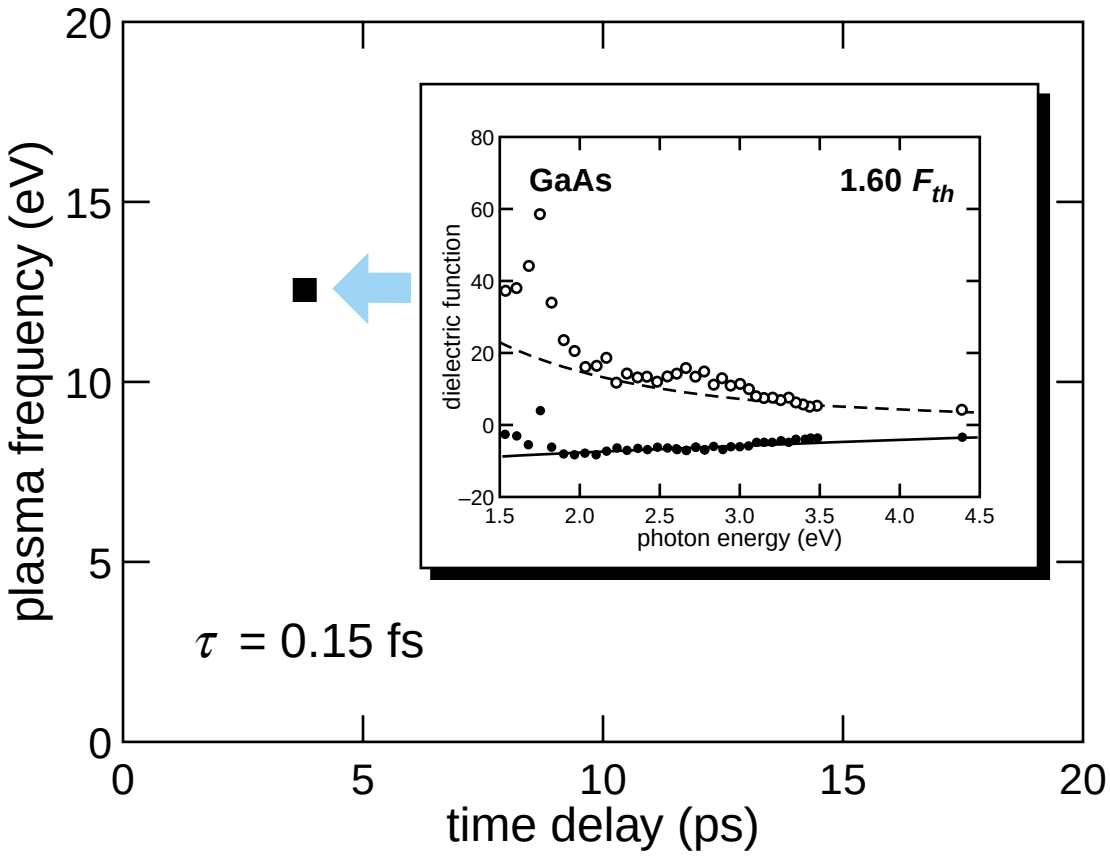
Results


4 ps
GaAs
1.60 F_{th}
dielectric function
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

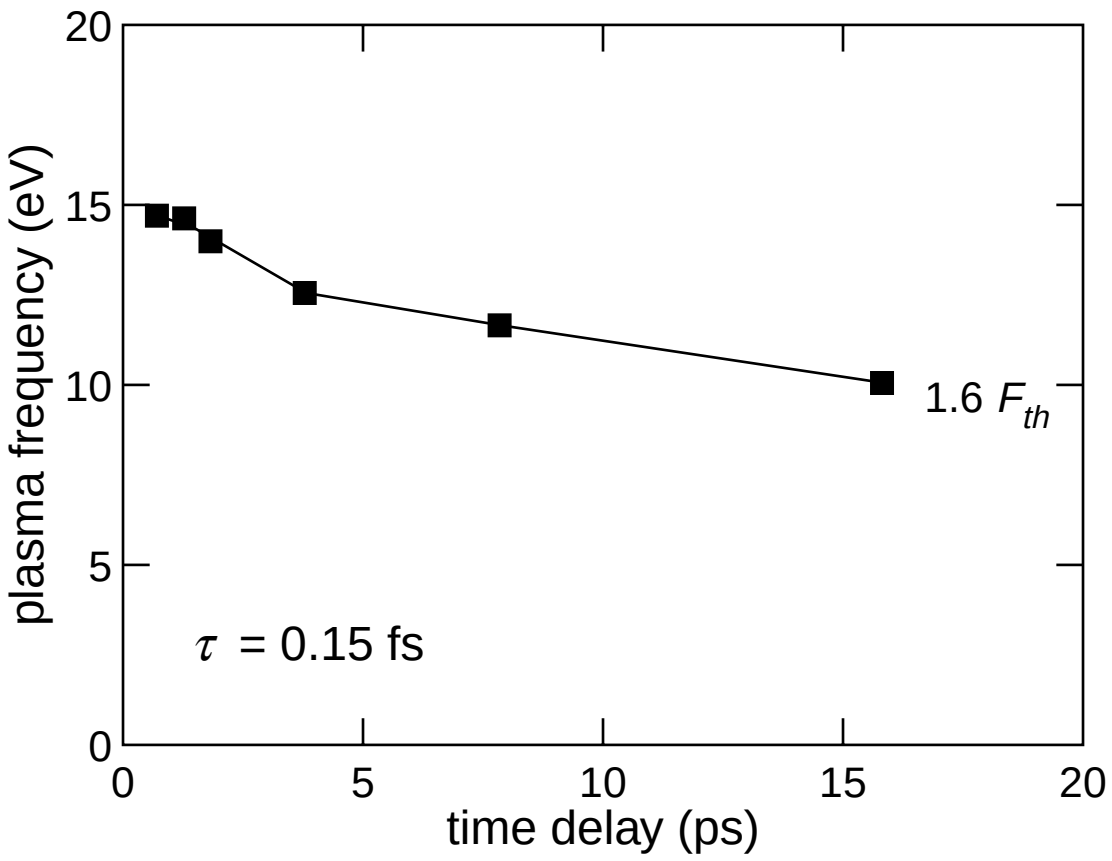
Results


plasma frequency (eV)
time delay (ps)
0
5
10
15
20
GaAs
1.60 F_{th}
dielectric function
photon energy (eV)
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

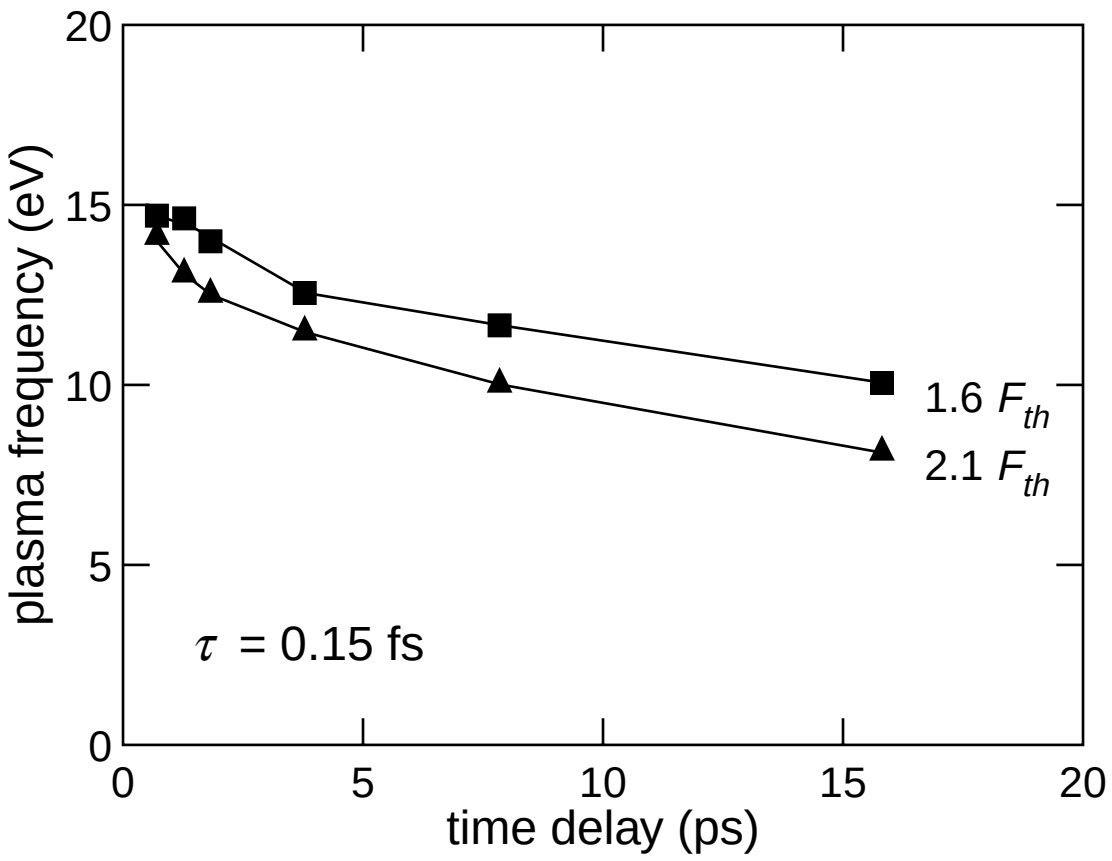
Results


plasma frequency (eV)
time delay (ps)
0
5
10
15
20
τ = 0.15 fs
GaAs
1.60 F_{th}
dielectric function
photon energy (eV)
80
60
40
20
0
−20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

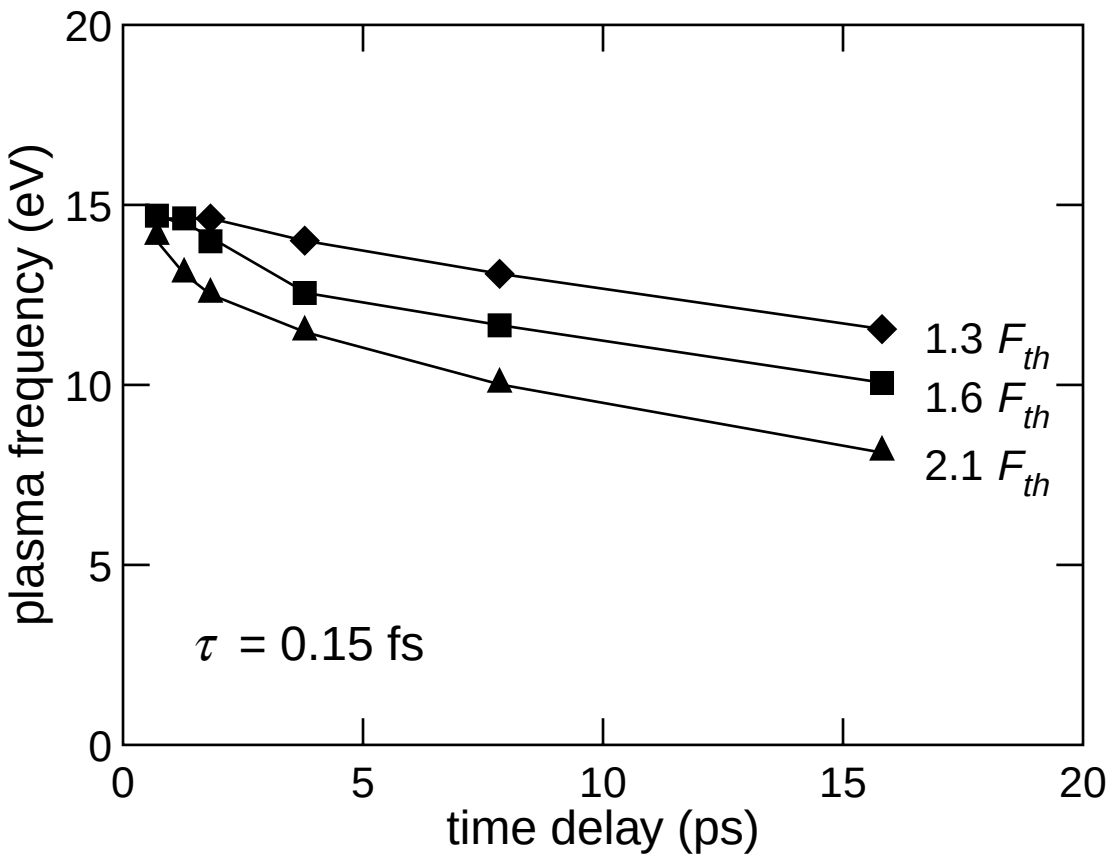
Results


20
15
10
5
0
plasma frequency (eV)
0
5
10
15
20
time delay (ps)
1.6 F_{th}
τ = 0.15 fs

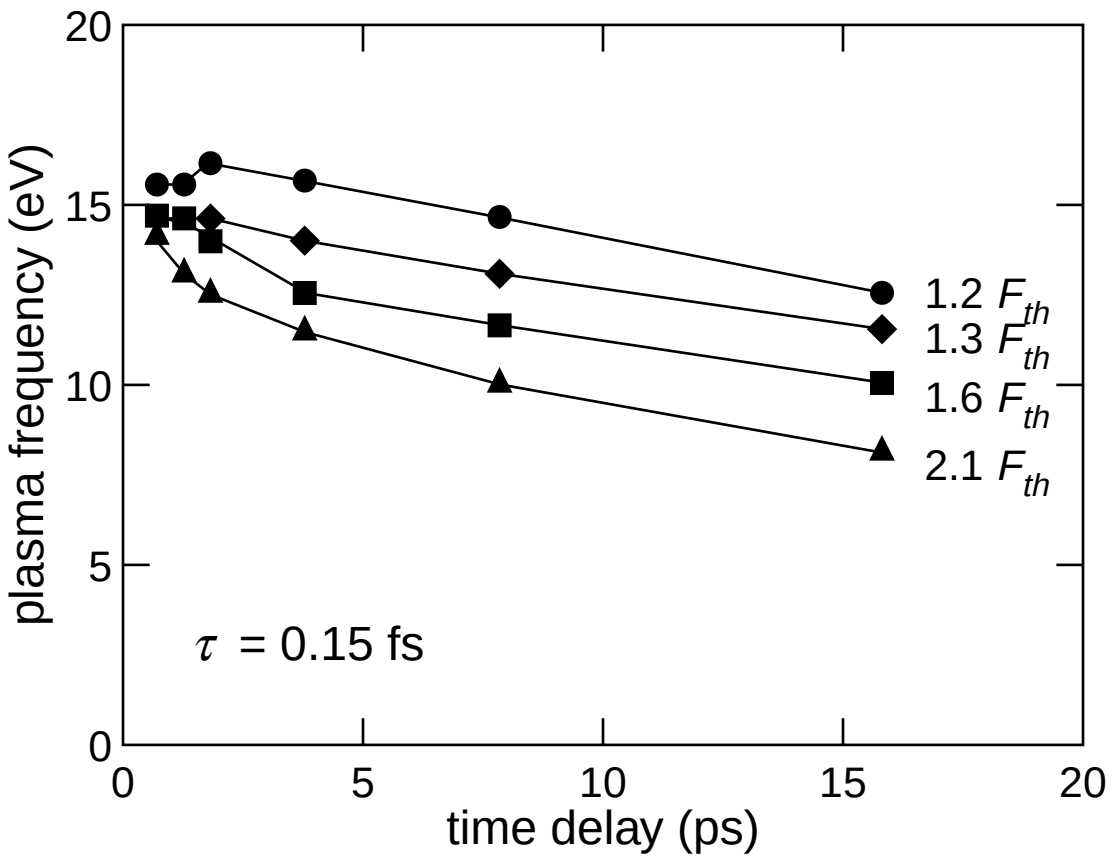
Results


20
15
10
5
0
plasma frequency (eV)
0
5
10
15
20
time delay (ps)
1.6 F_{th}
2.1 F_{th}
τ = 0.15 fs

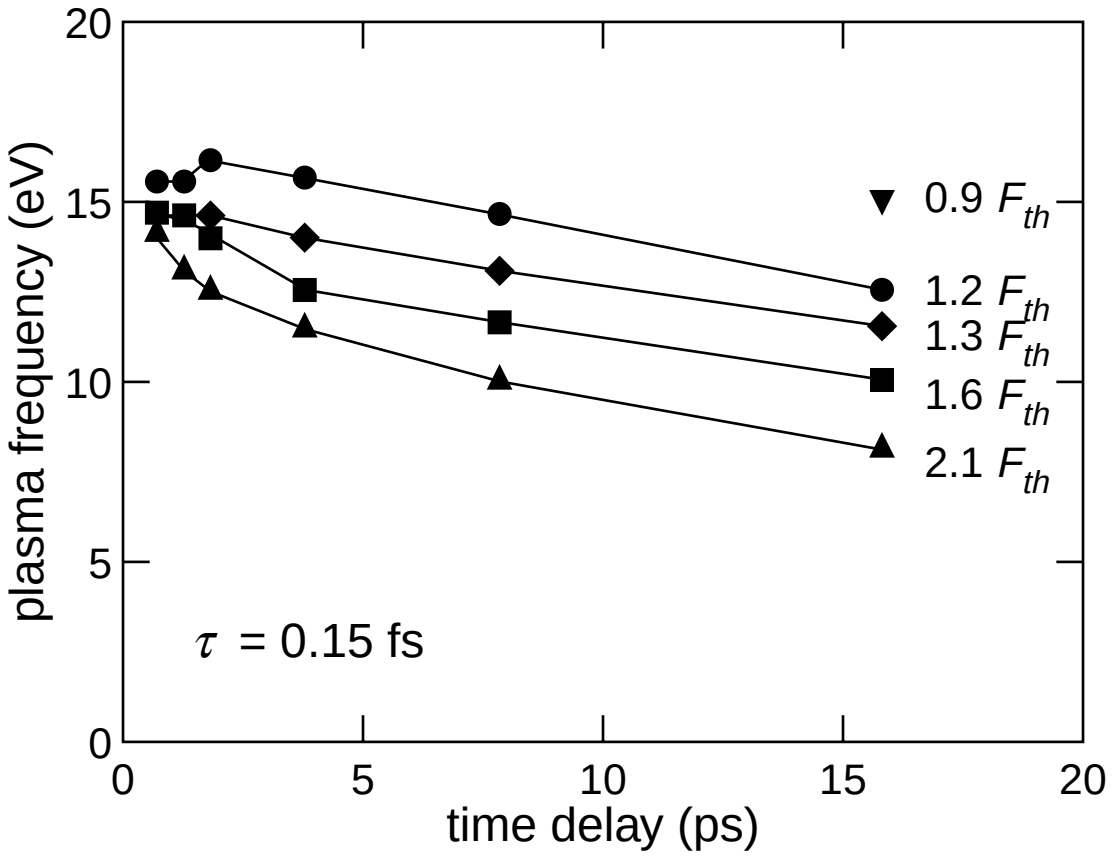
Results


20
15
10
5
0
plasma frequency (eV)
0
5
10
15
20
time delay (ps)
1.3 F_{th}
1.6 F_{th}
2.1 F_{th}
τ = 0.15 fs

Results


20
15
10
5
0
0
5
10
15
20
plasma frequency (eV)
time delay (ps)
1.2 F_{th}
1.3 F_{th}
1.6 F_{th}
2.1 F_{th}
τ = 0.15 fs

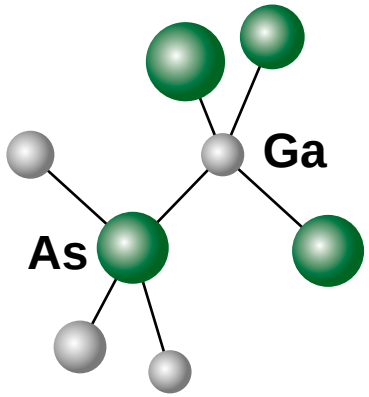
Results


20
15
10
5
0
0
5
10
15
20
plasma frequency (eV)
time delay (ps)
0.9 F_{th}
1.2 F_{th}
1.3 F_{th}
1.6 F_{th}
2.1 F_{th}
τ = 0.15 fs

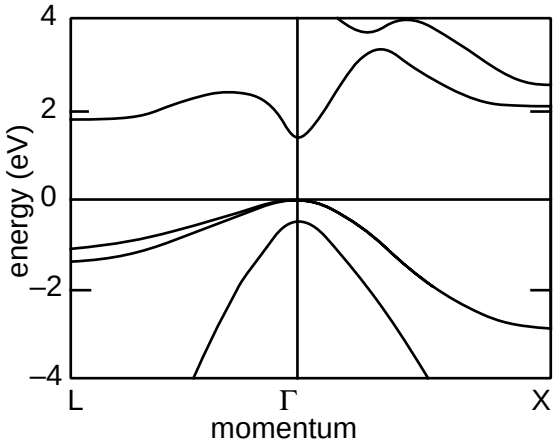
Discussion

short time scale

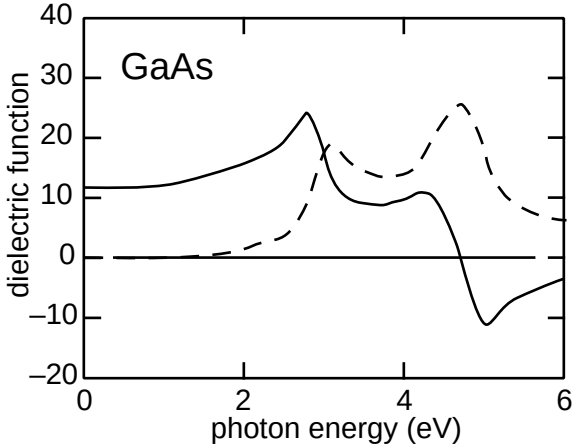
structure



bandstructure



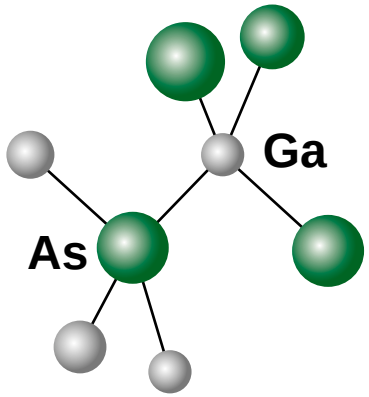
dielectric function



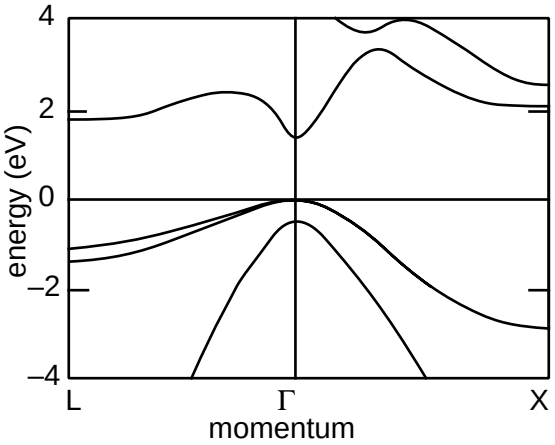
Discussion

short time scale

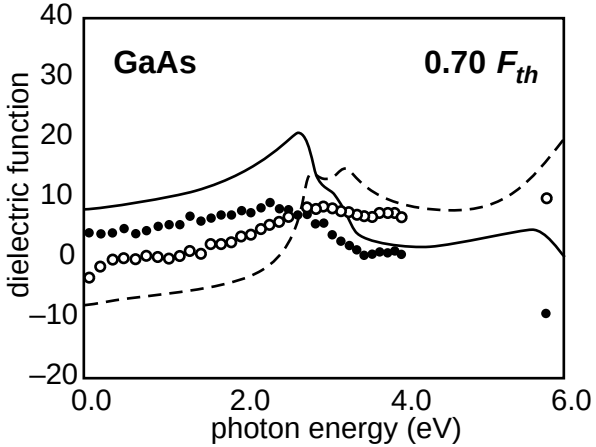
structure



bandstructure



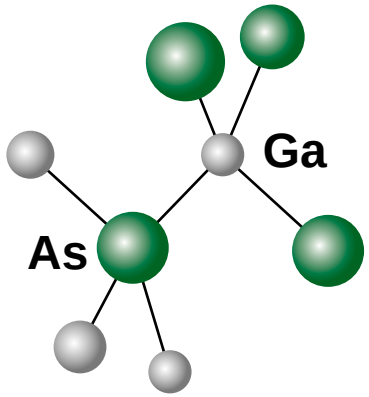
dielectric function



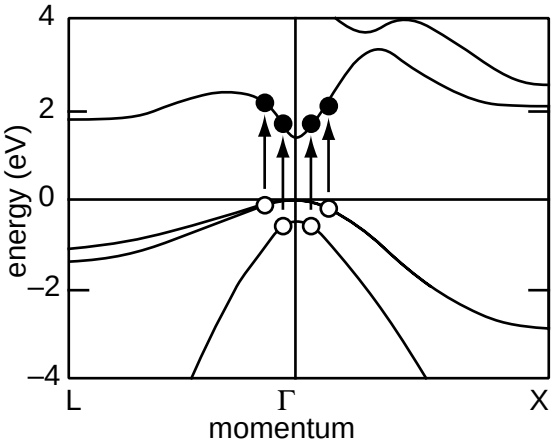
Discussion

short time scale

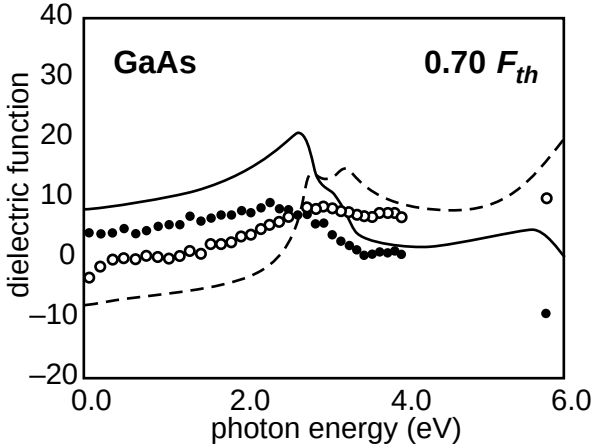
structure



bandstructure



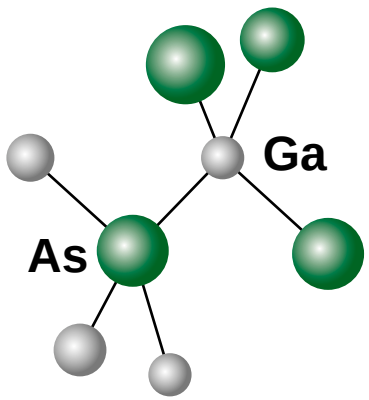
dielectric function



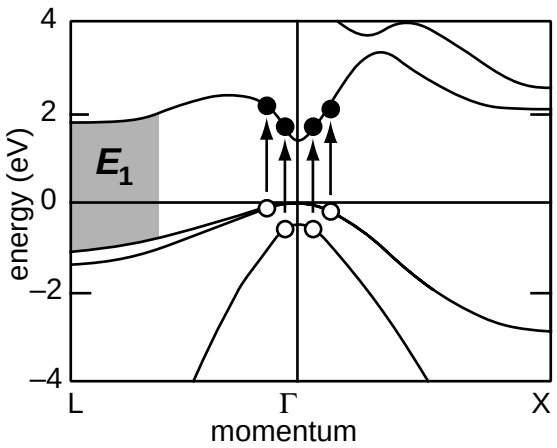
Discussion

short time scale

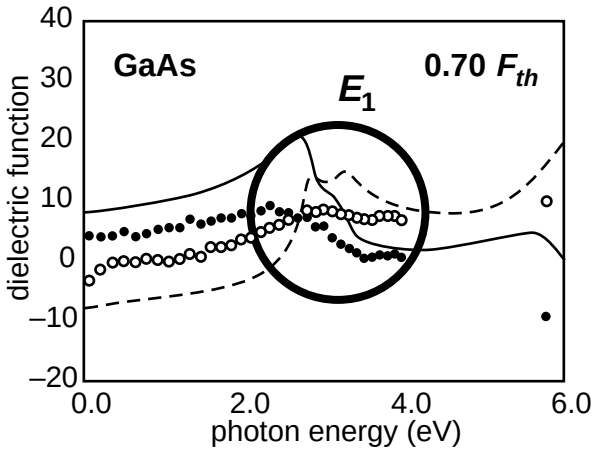
structure



bandstructure



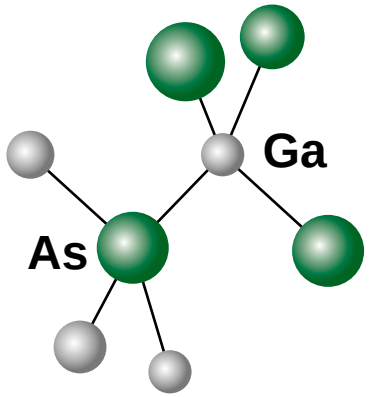
dielectric function



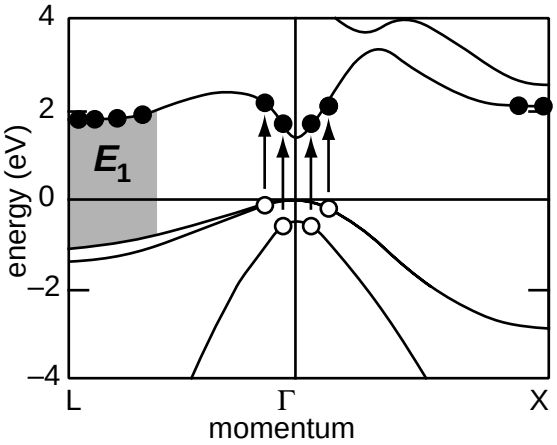
Discussion

short time scale

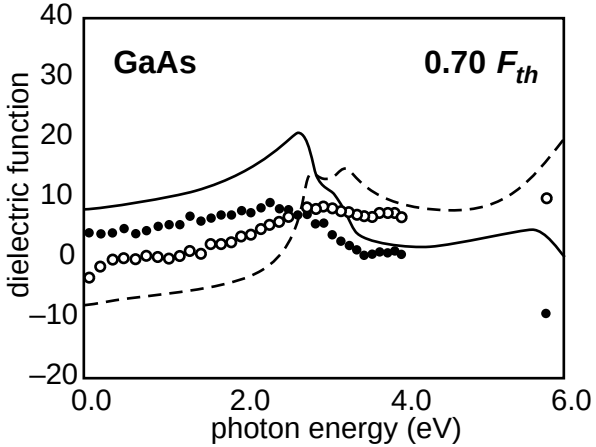
structure



bandstructure



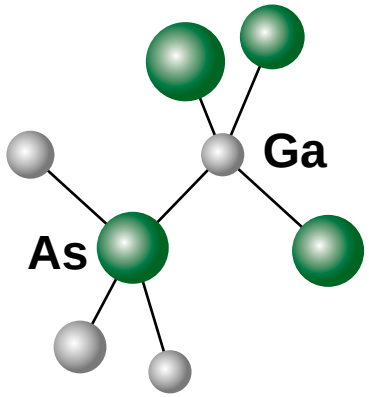
dielectric function



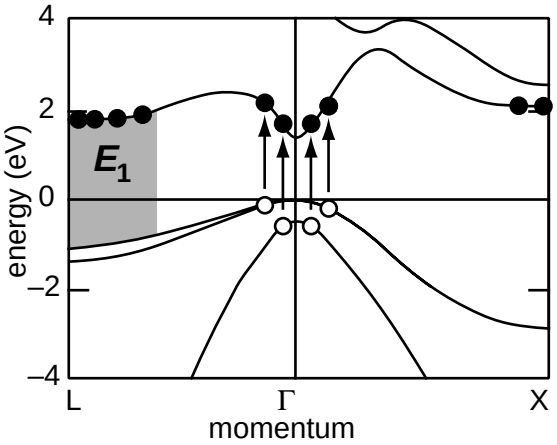
Discussion

short time scale

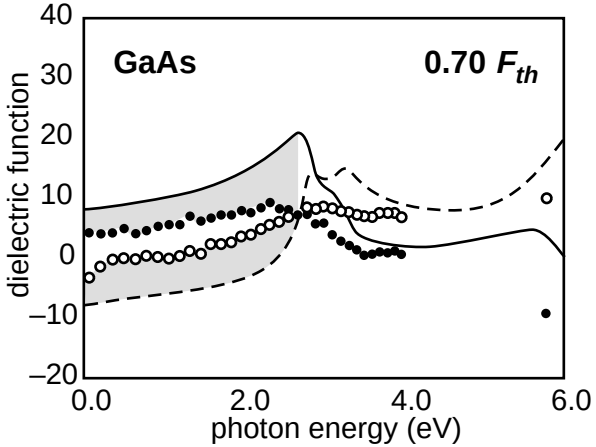
structure



bandstructure



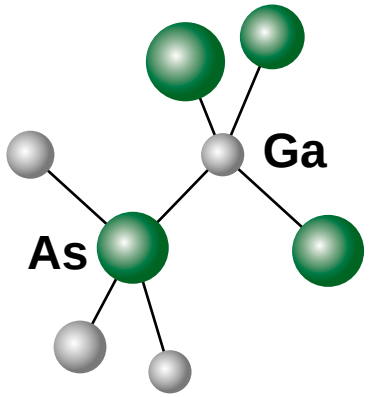
dielectric function



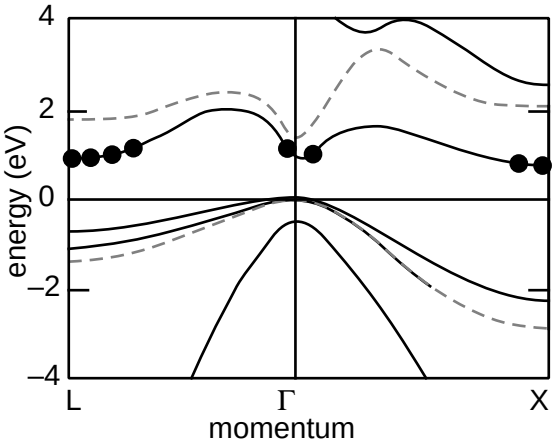
Discussion

short time scale

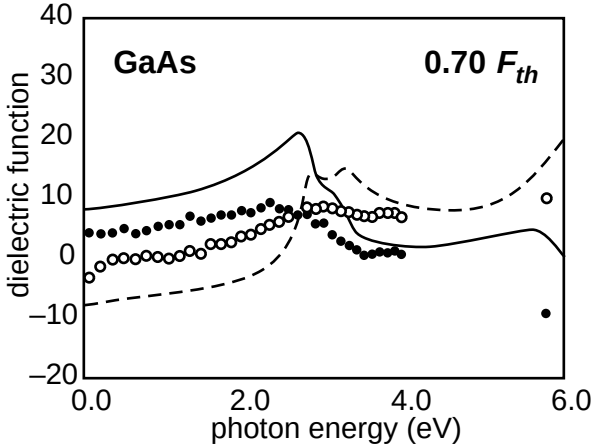
structure



bandstructure



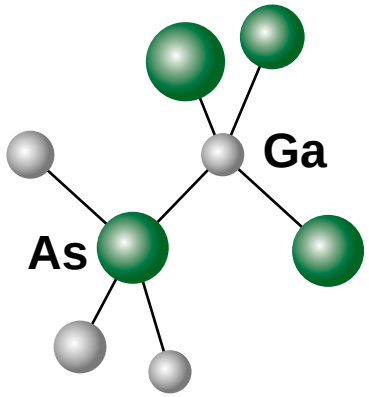
dielectric function



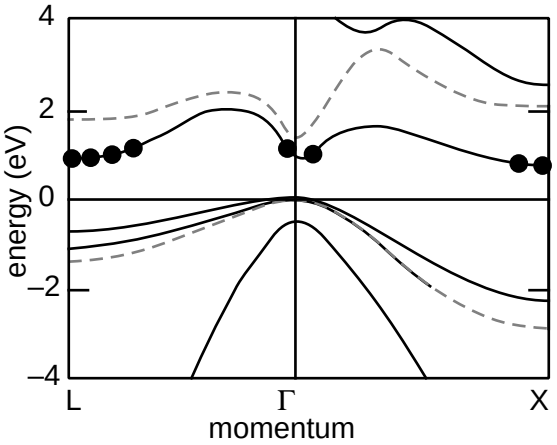
Discussion

short time scale

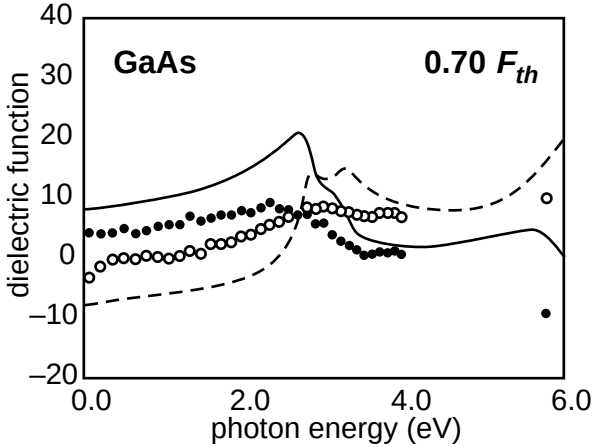
structure



bandstructure



dielectric function

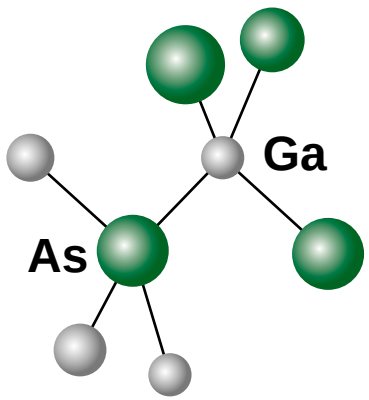


D.H. Kim, *et al.*, *Sol. State Comm.* 89, 119 (1994)

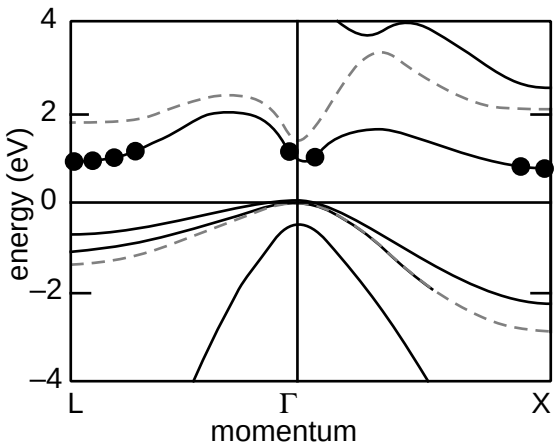
Discussion

short time scale

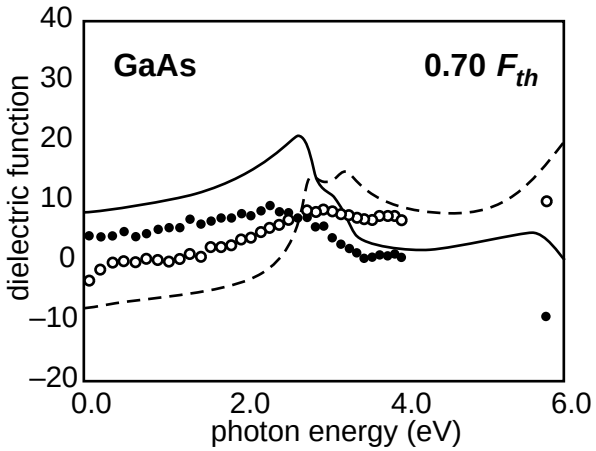
structure



bandstructure



dielectric function

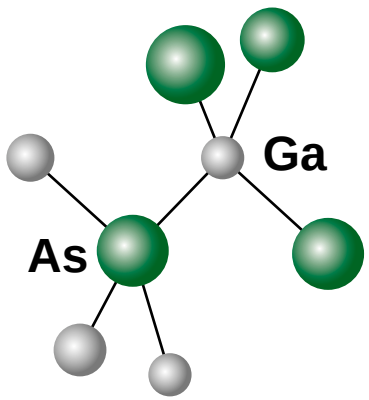


electronic effects dominate at short time scales…

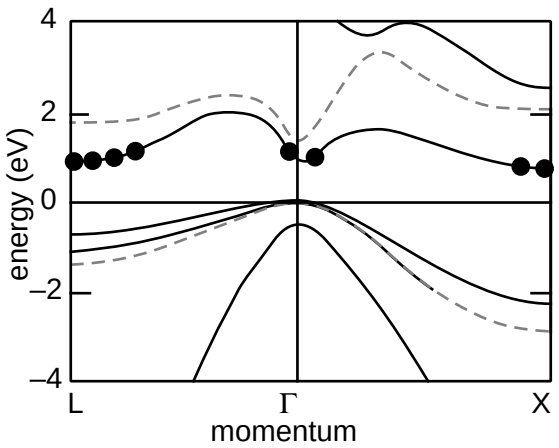
Discussion

short time scale

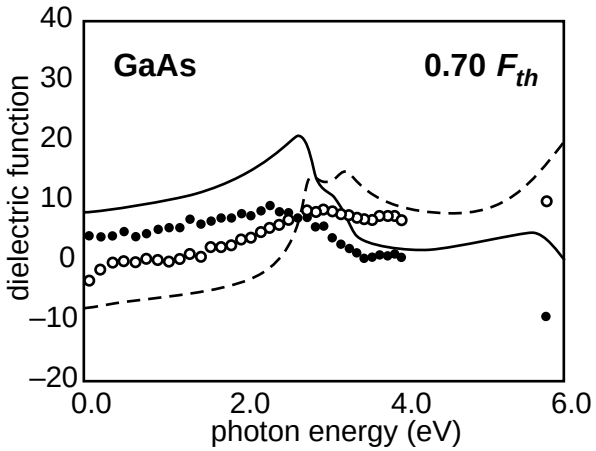
structure



bandstructure



dielectric function

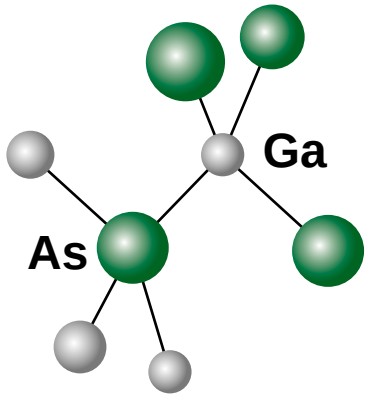


…but they are not as simple as we used to think

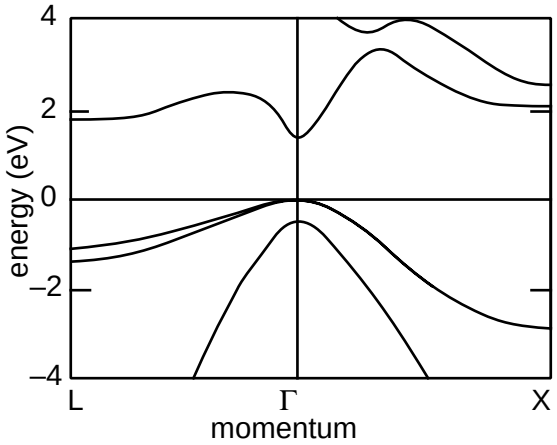
Discussion

long time scale, high fluence

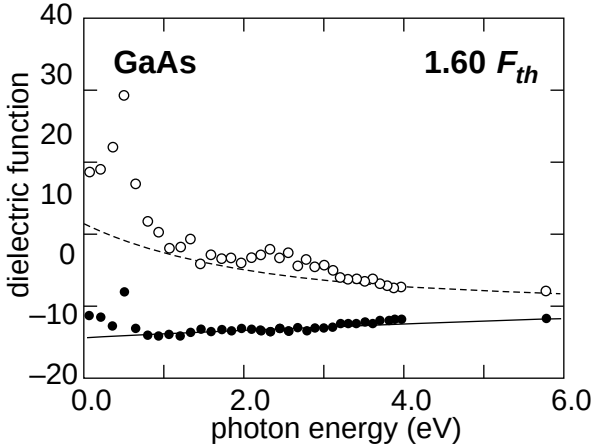
structure



bandstructure



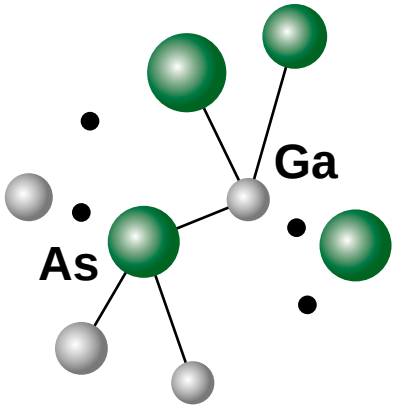
dielectric function



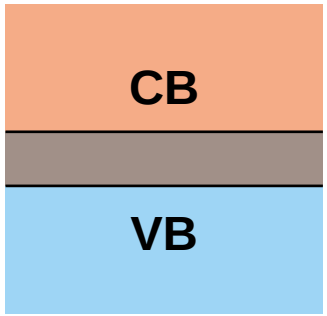
Discussion

long time scale, high fluence

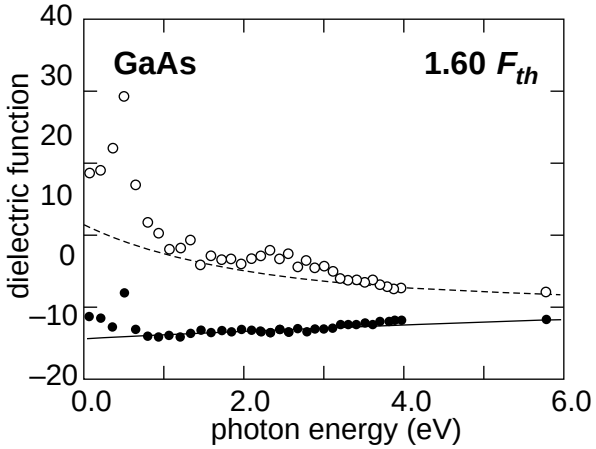
structure



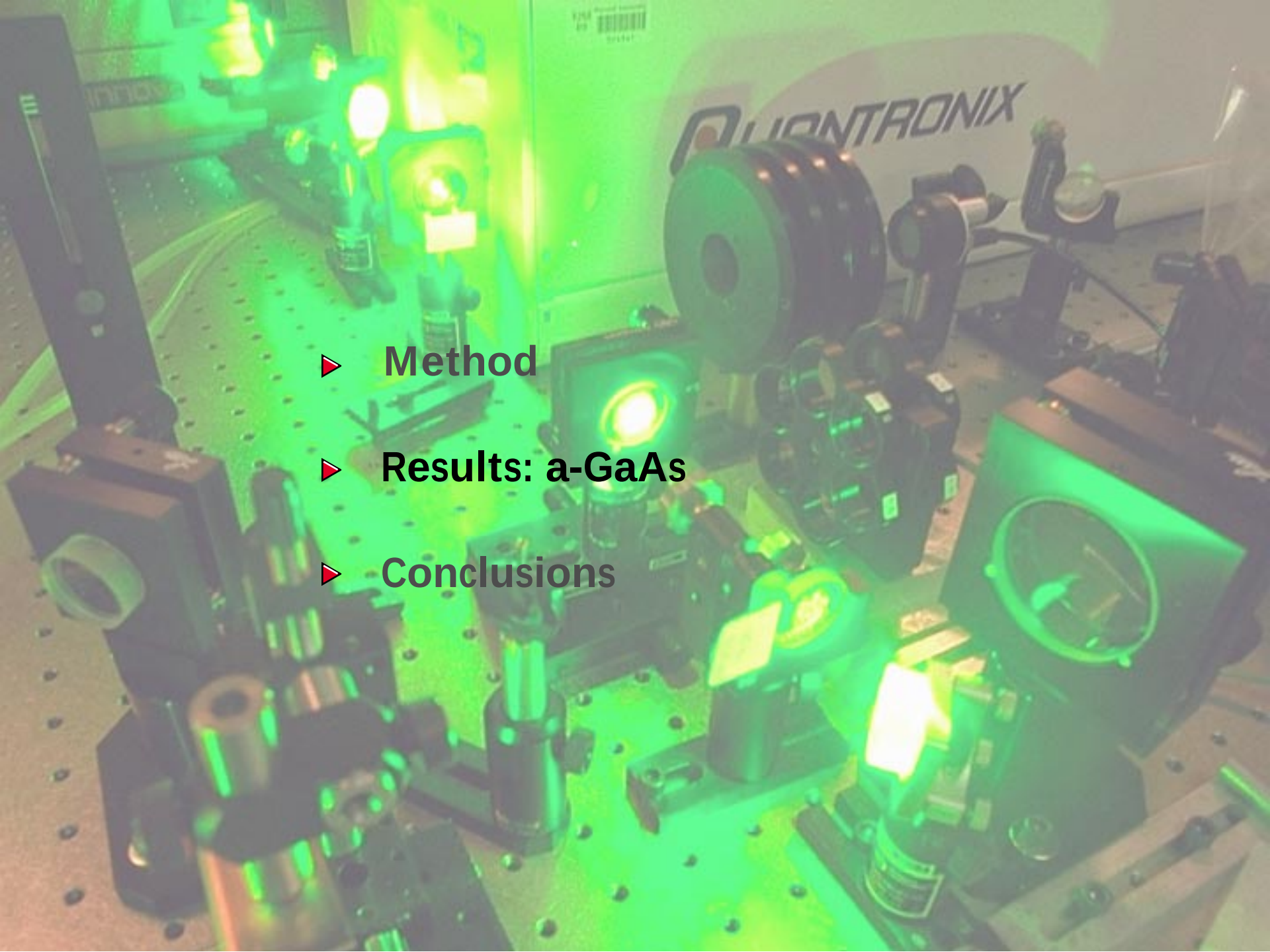
bandstructure



dielectric function

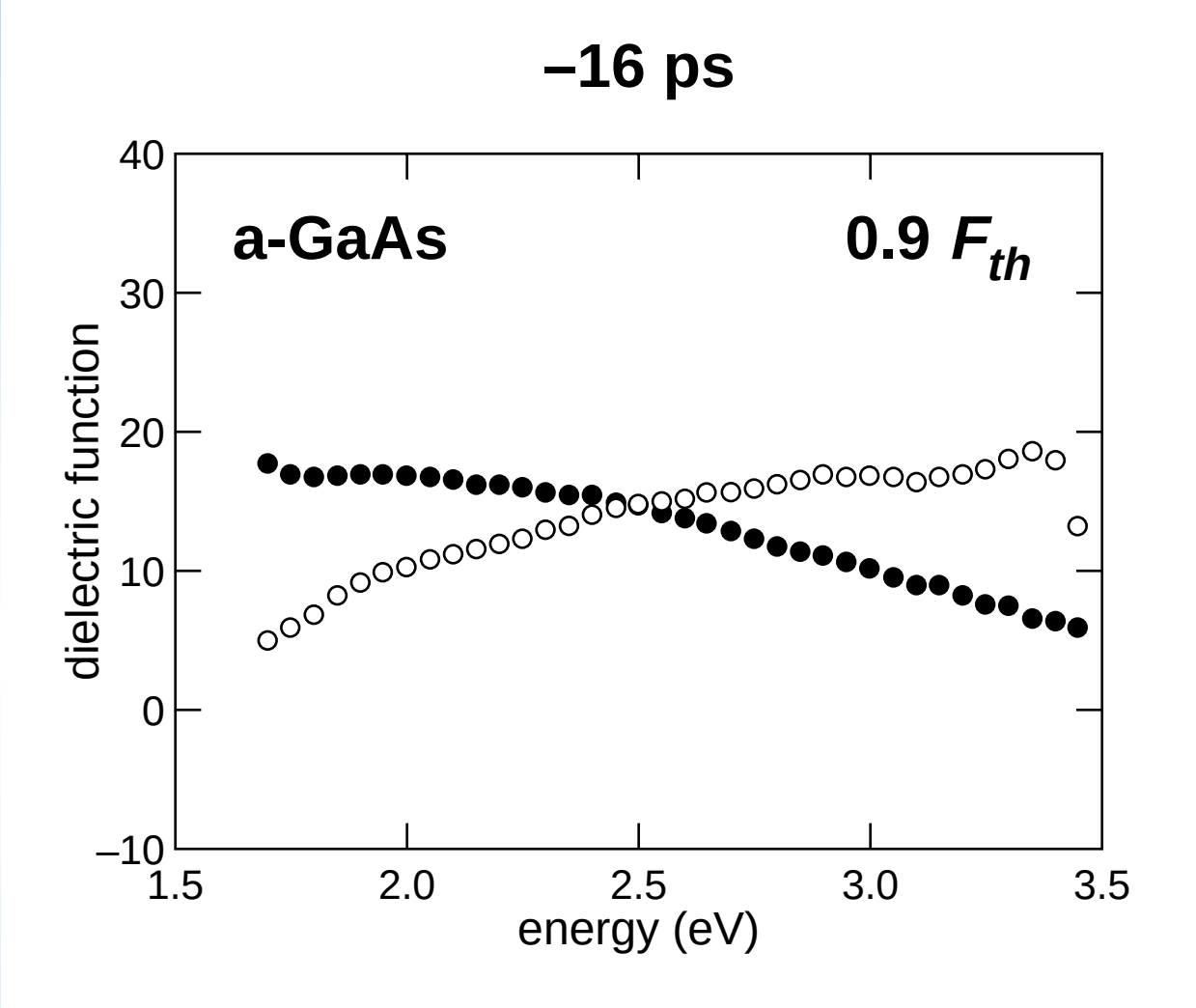


gradual drop in gap → not electronic effect

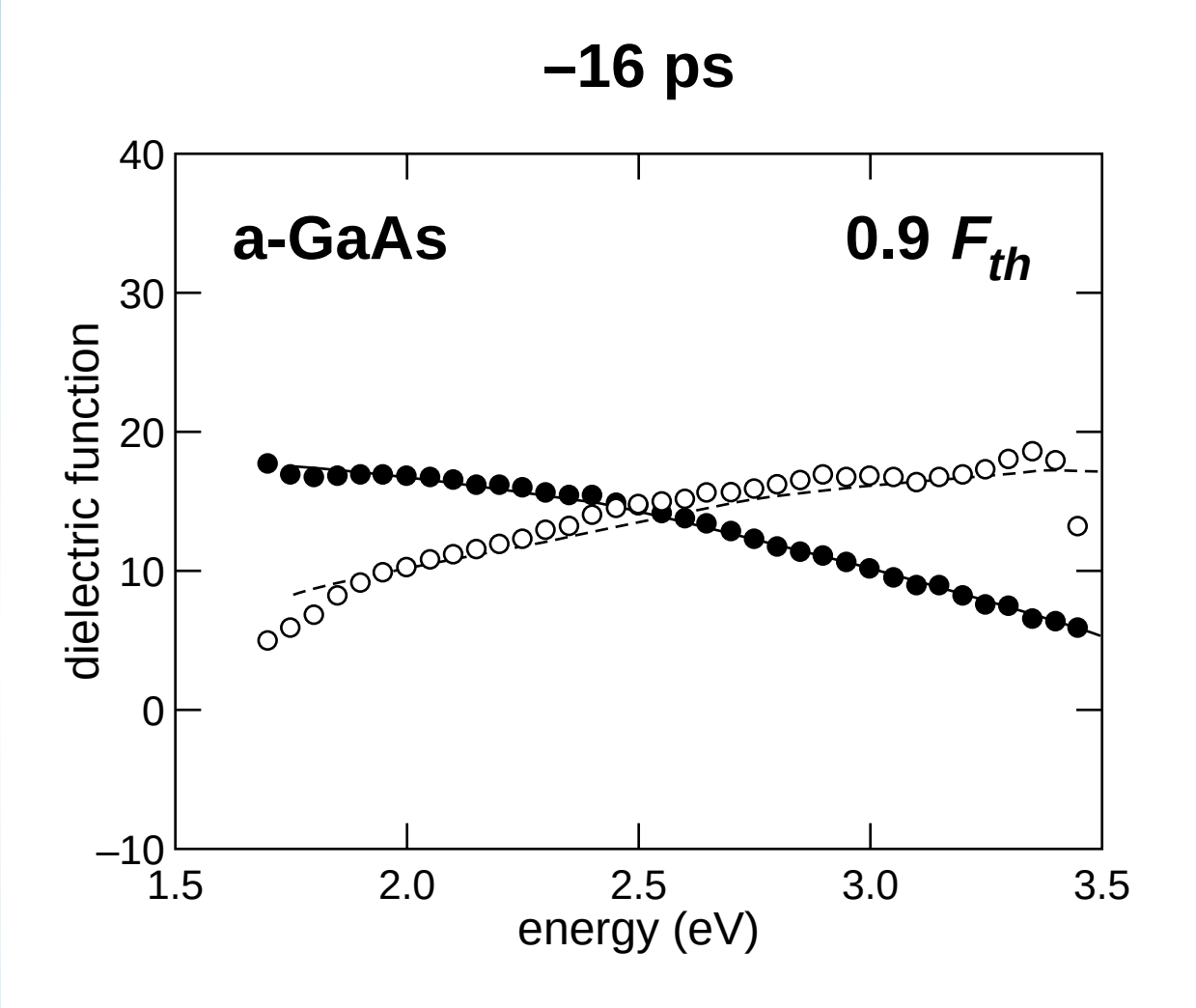


- Method
- **Results: a-GaAs**
- Conclusions

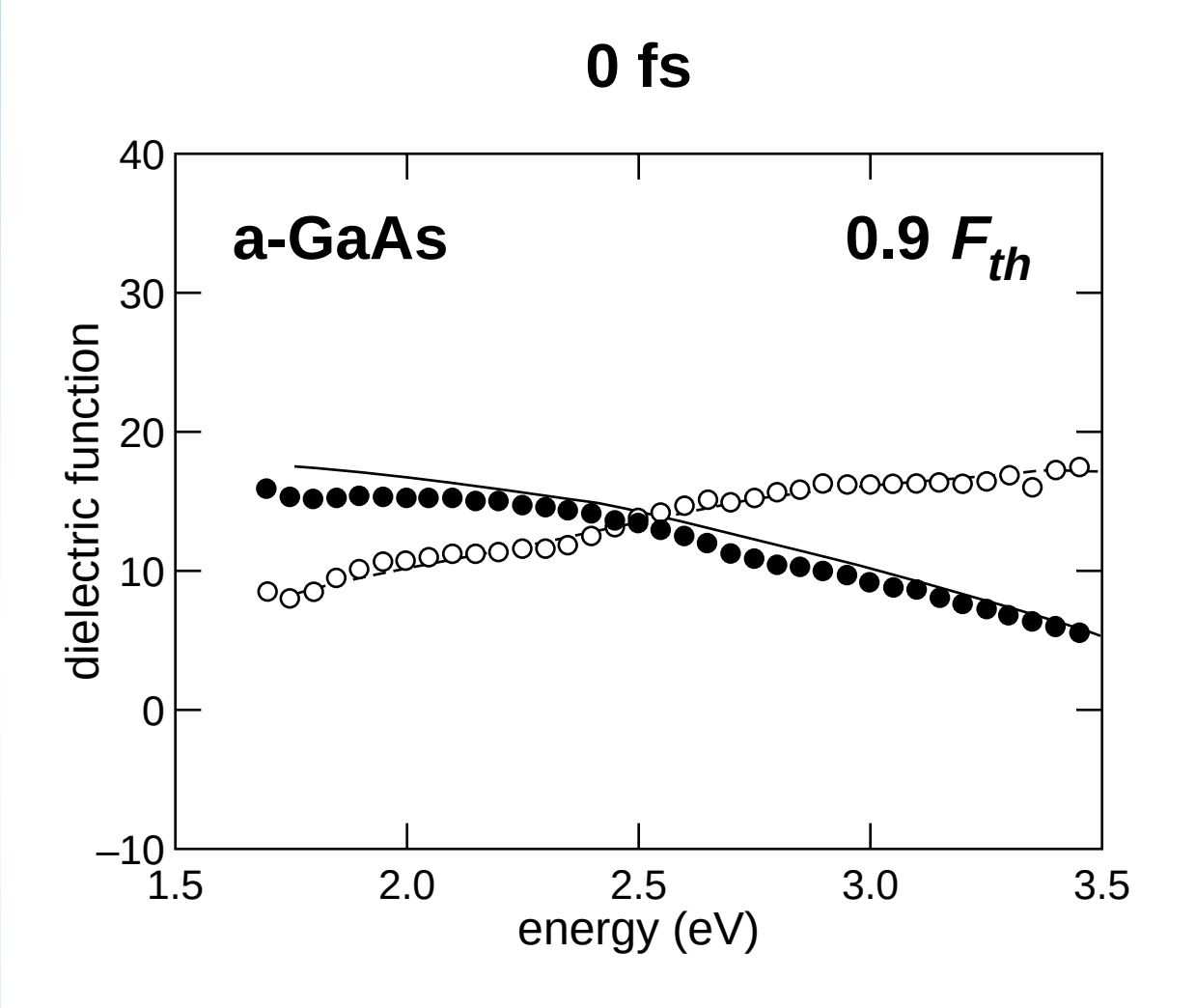
Results


–16 ps
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

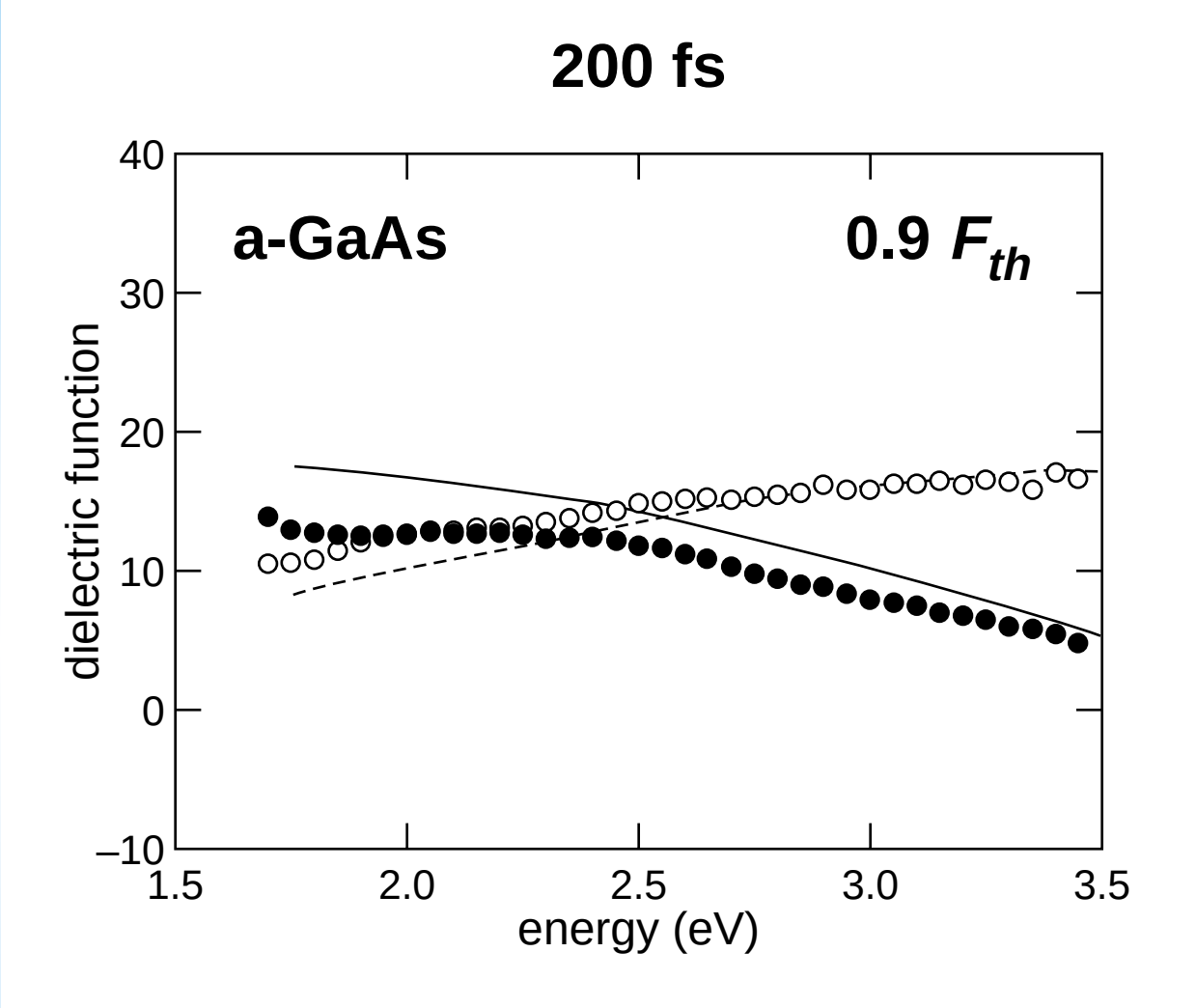
Results


–16 ps
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

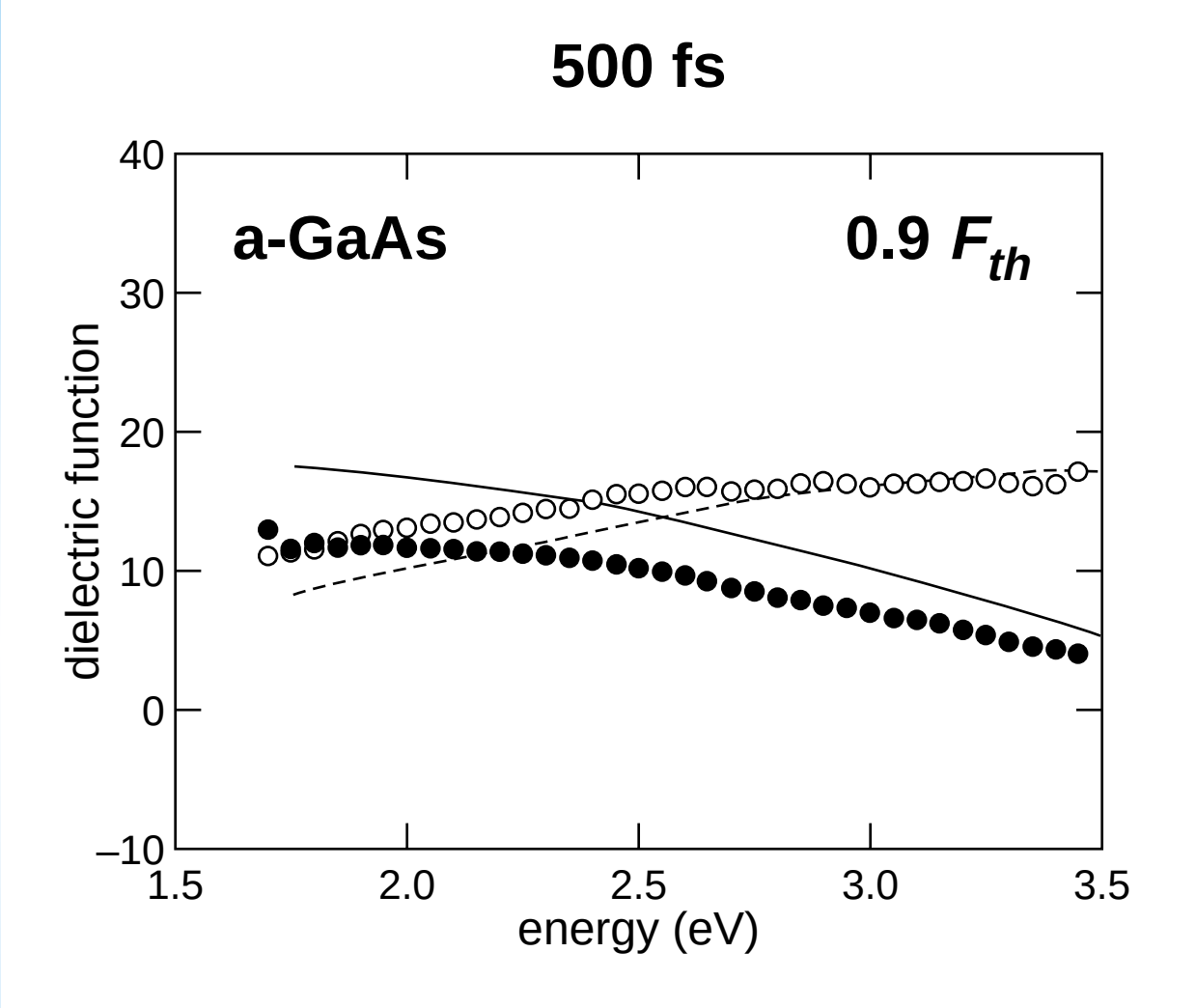
Results


0 fs
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

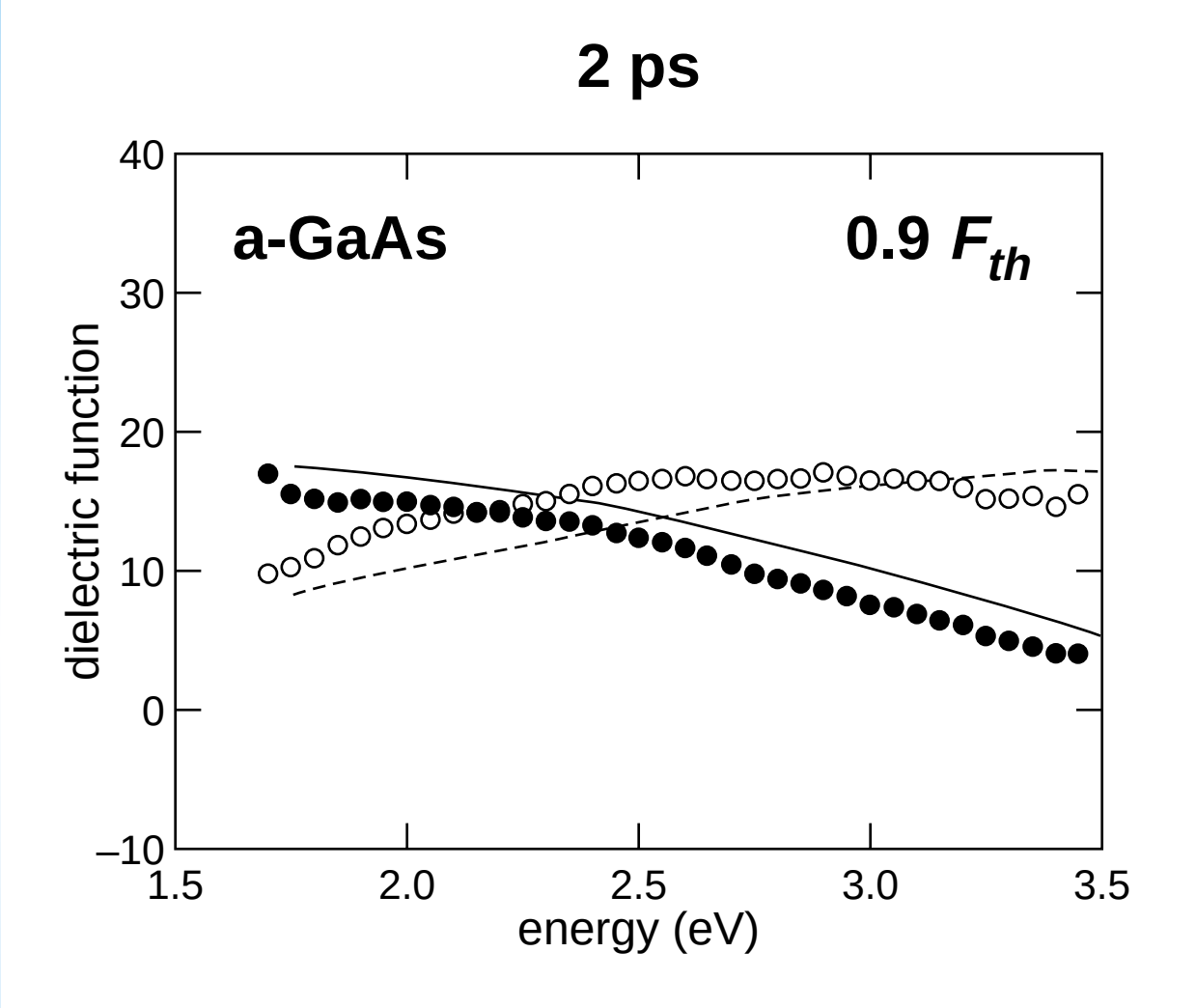
Results


200 fs
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

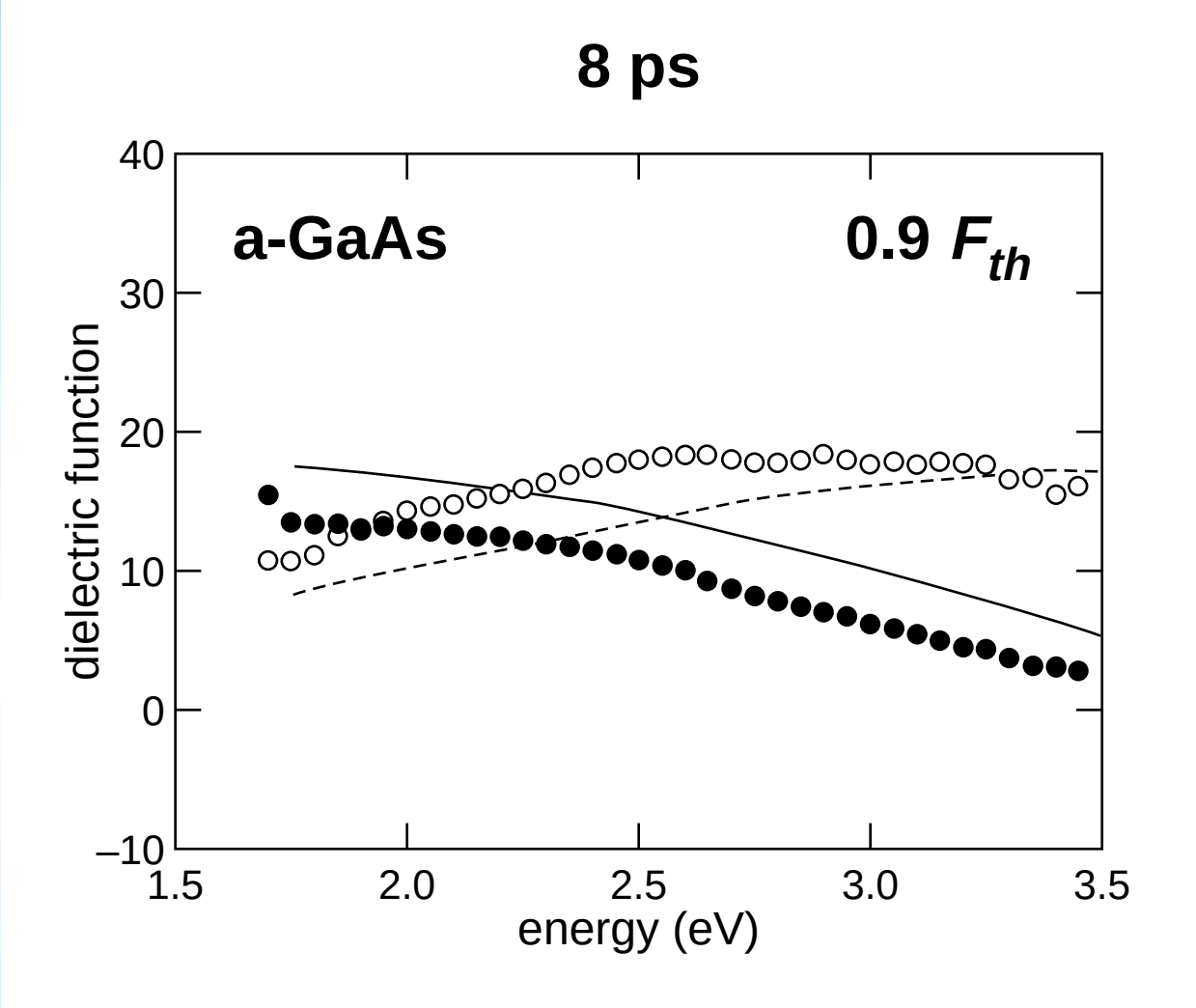
Results


500 fs
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

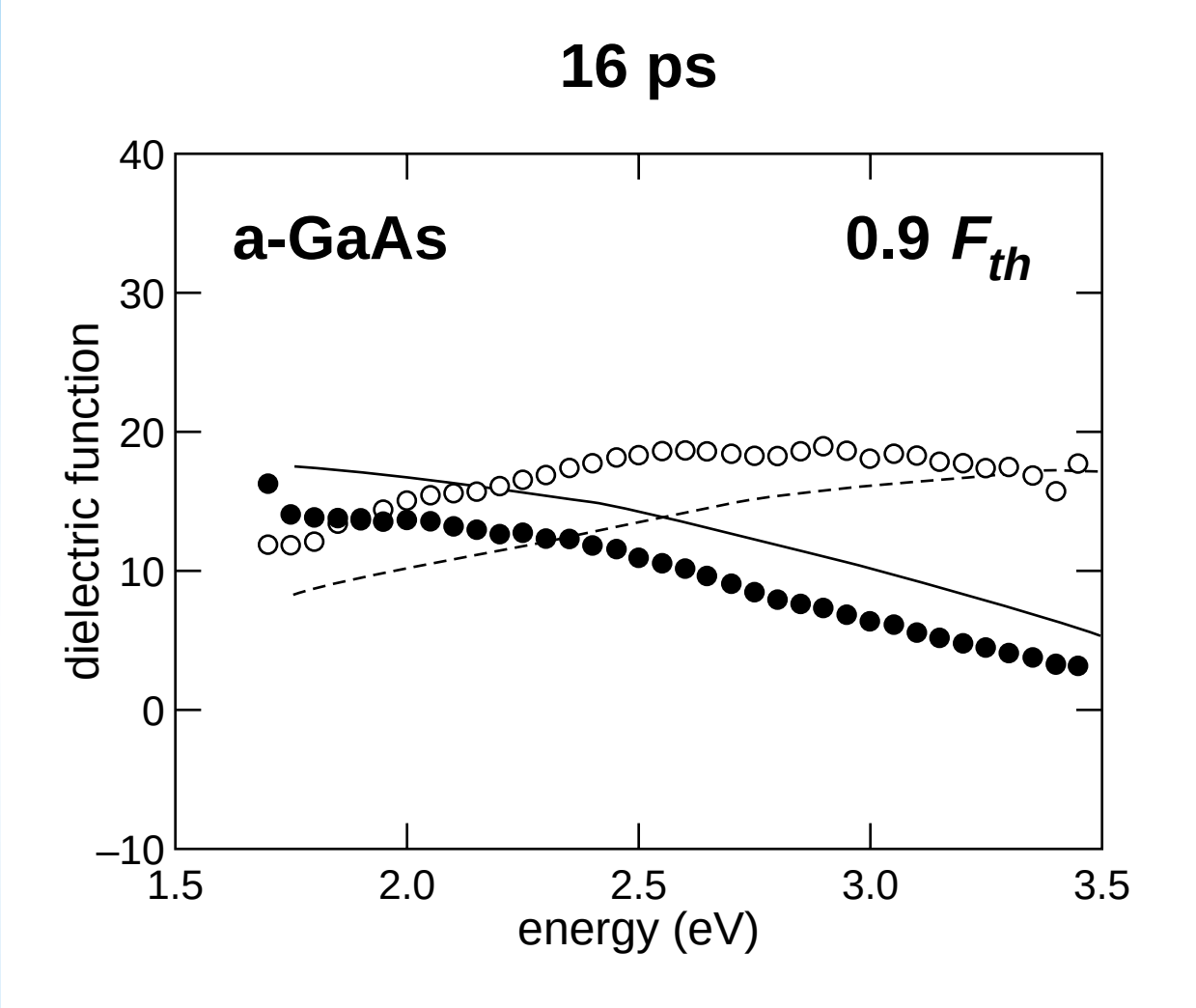
Results


2 ps
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

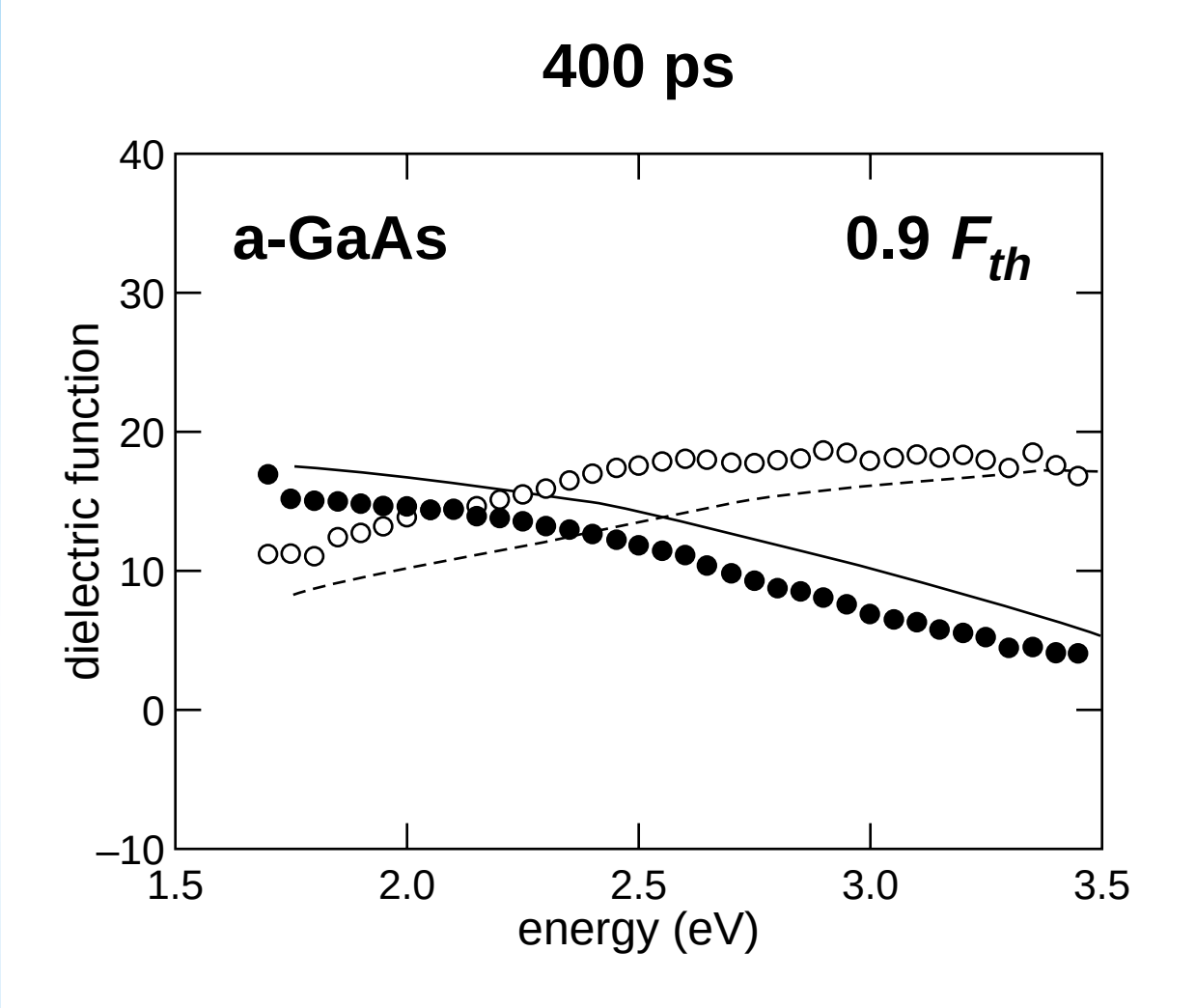
Results


8 ps
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

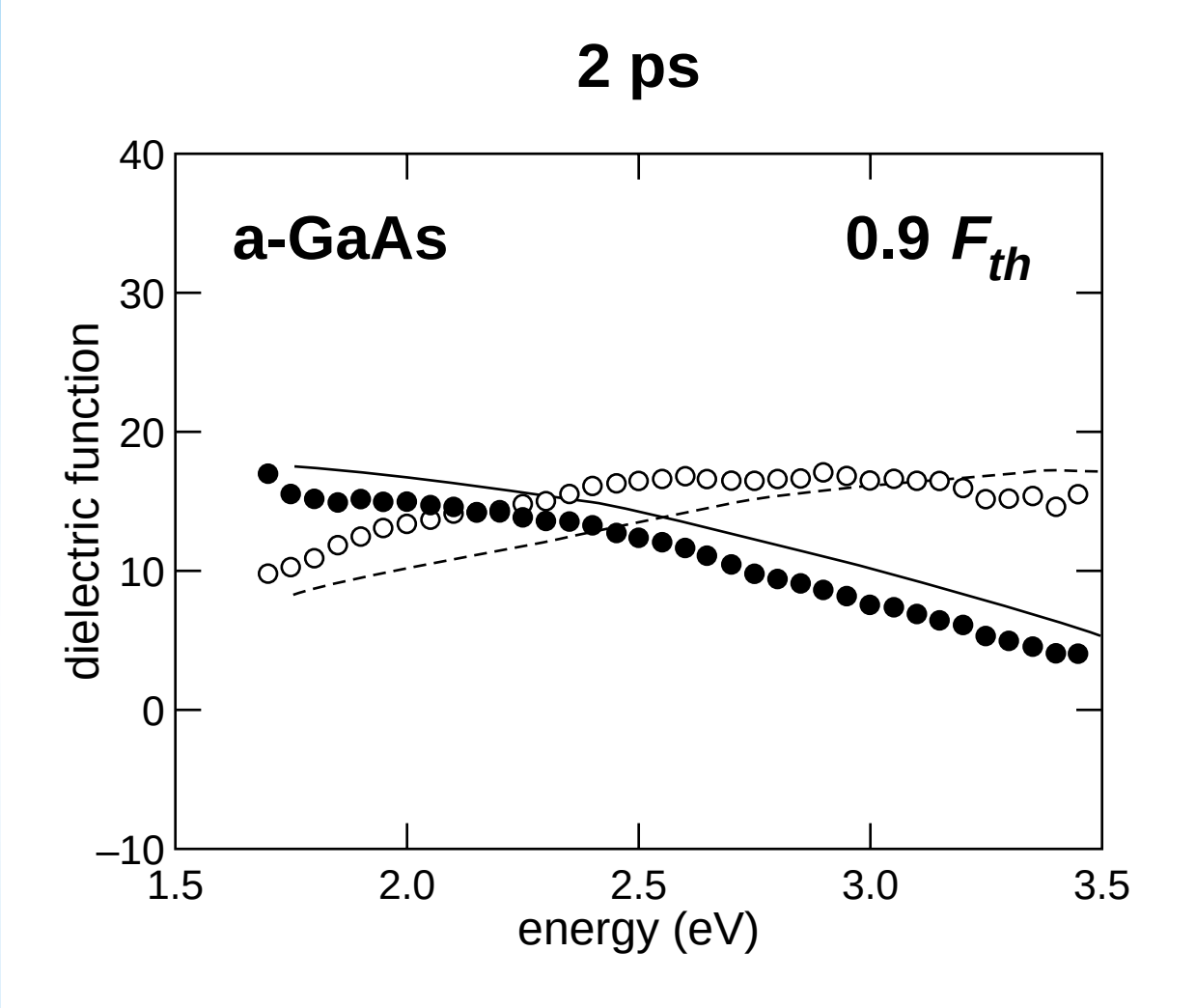
Results


16 ps
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

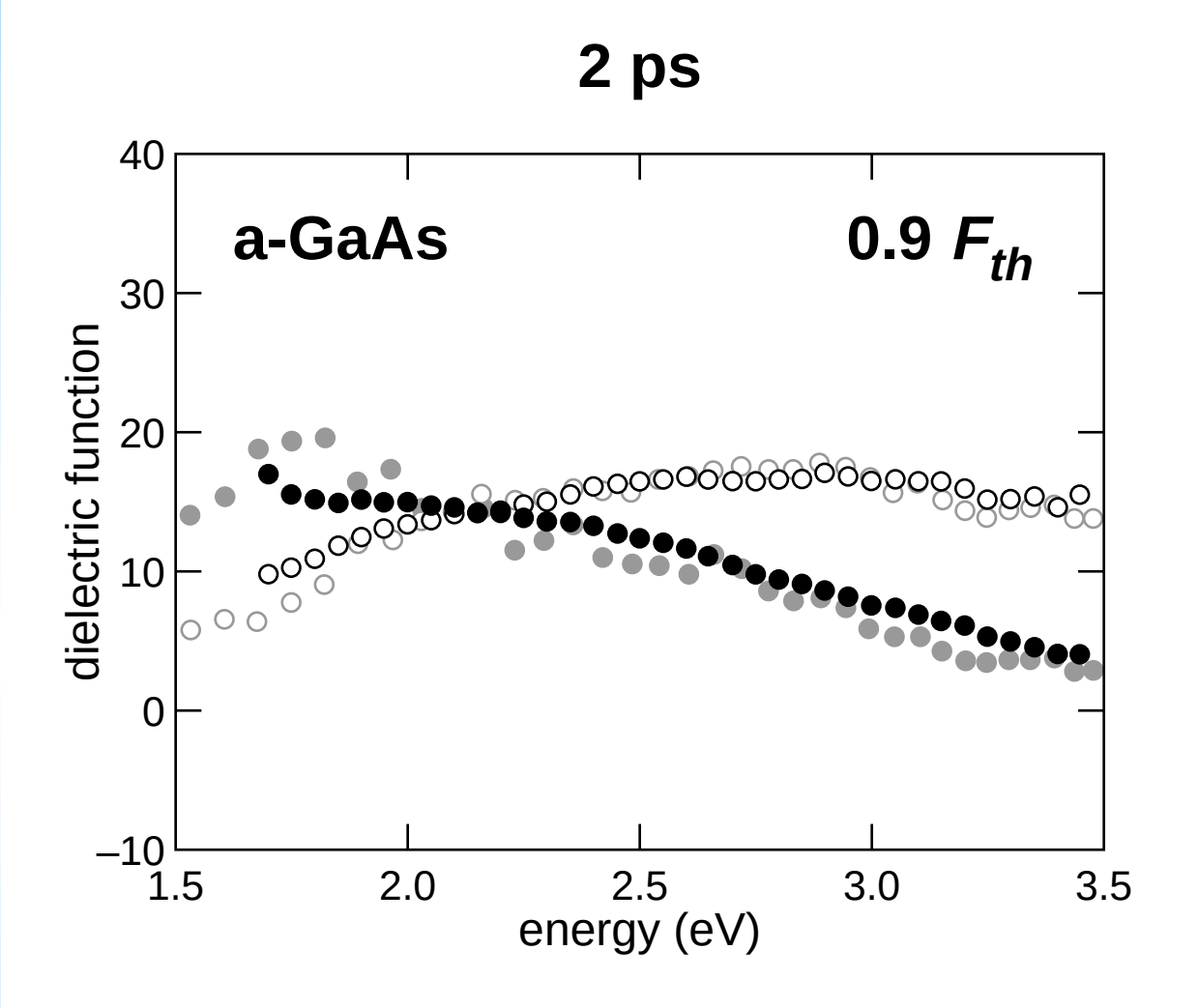
Results


400 ps
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

Results

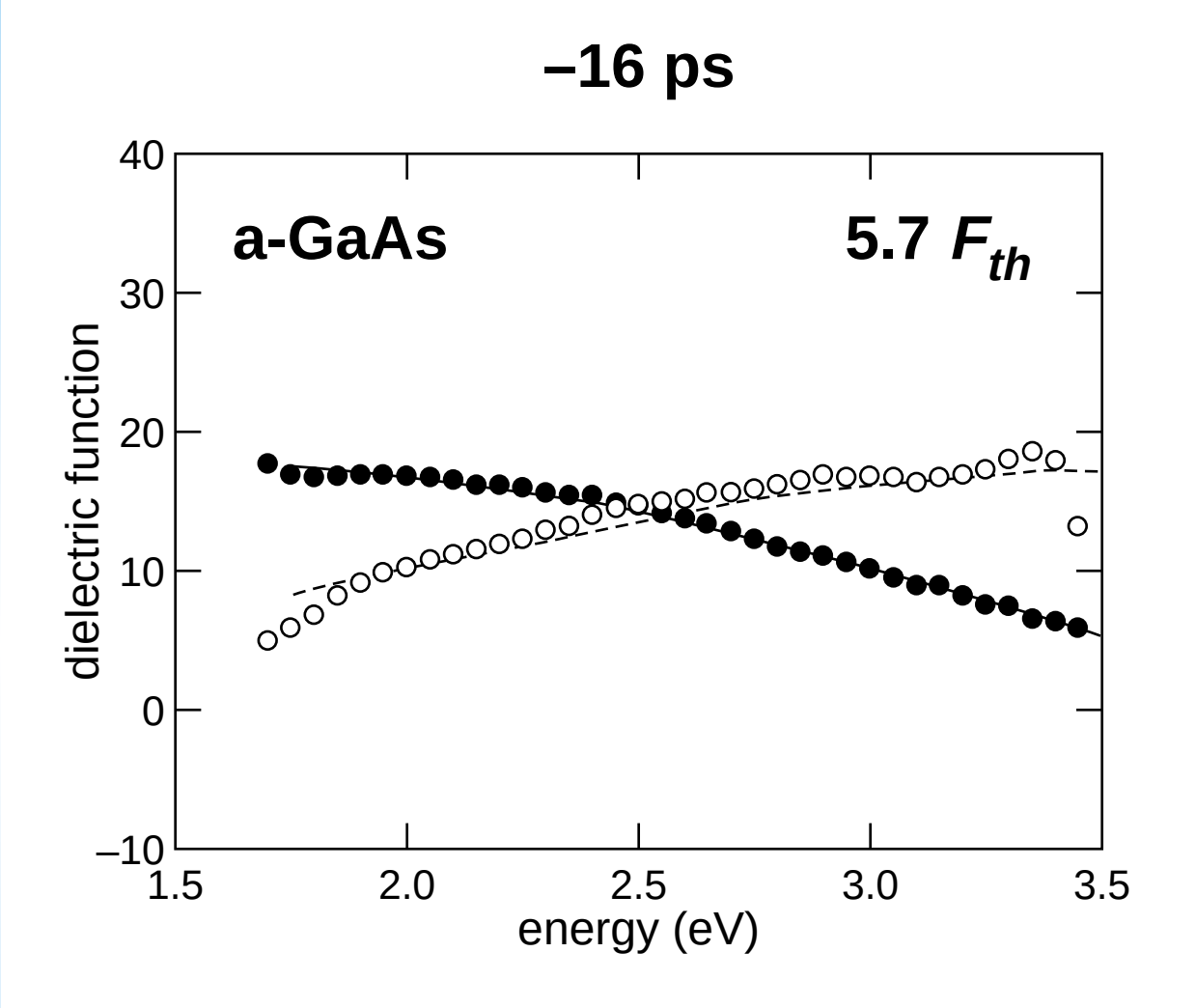

2 ps
a-GaAs
0.9 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

Results

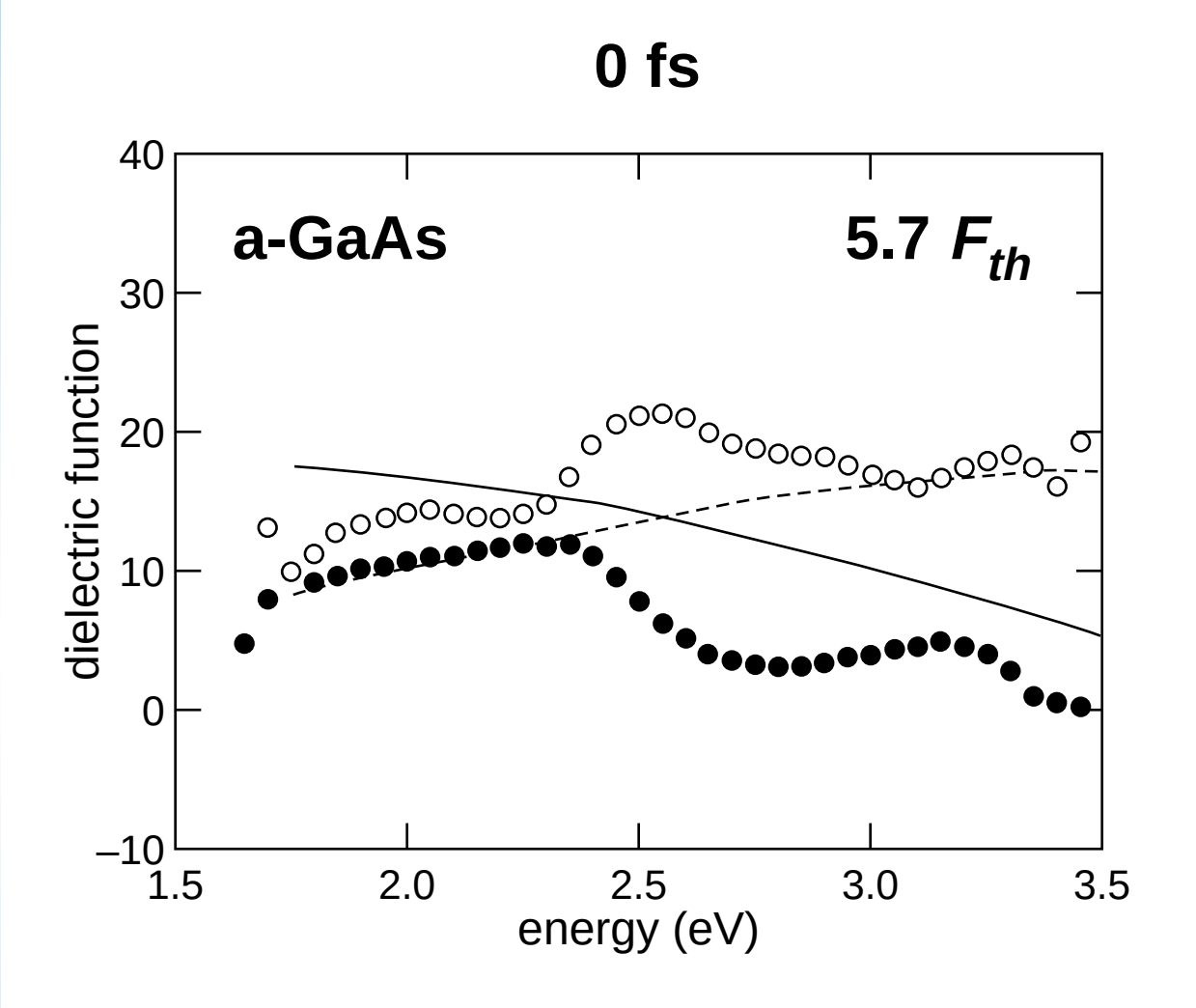


matches disordered c-GaAs

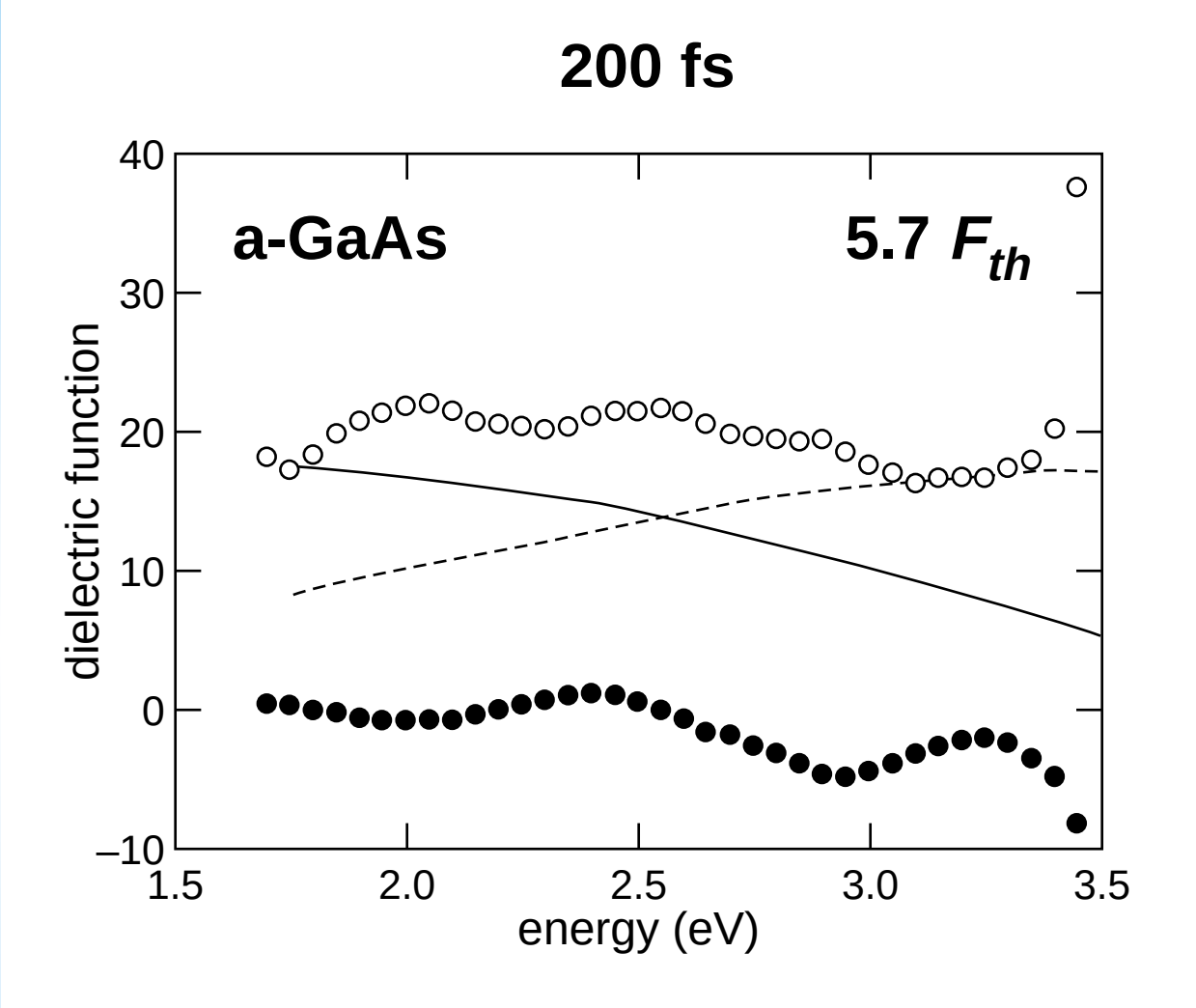
Results


–16 ps
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

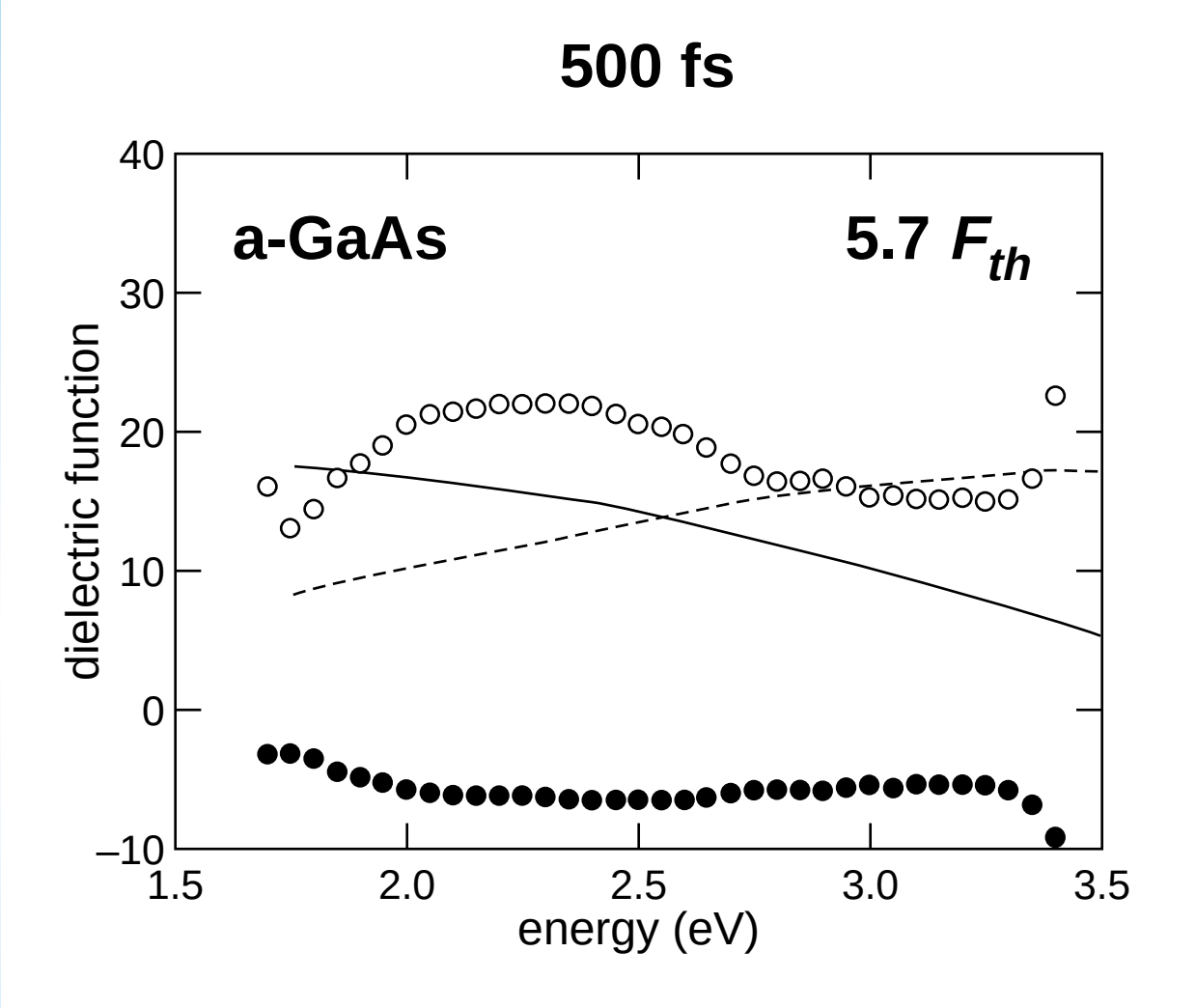
Results


0 fs
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

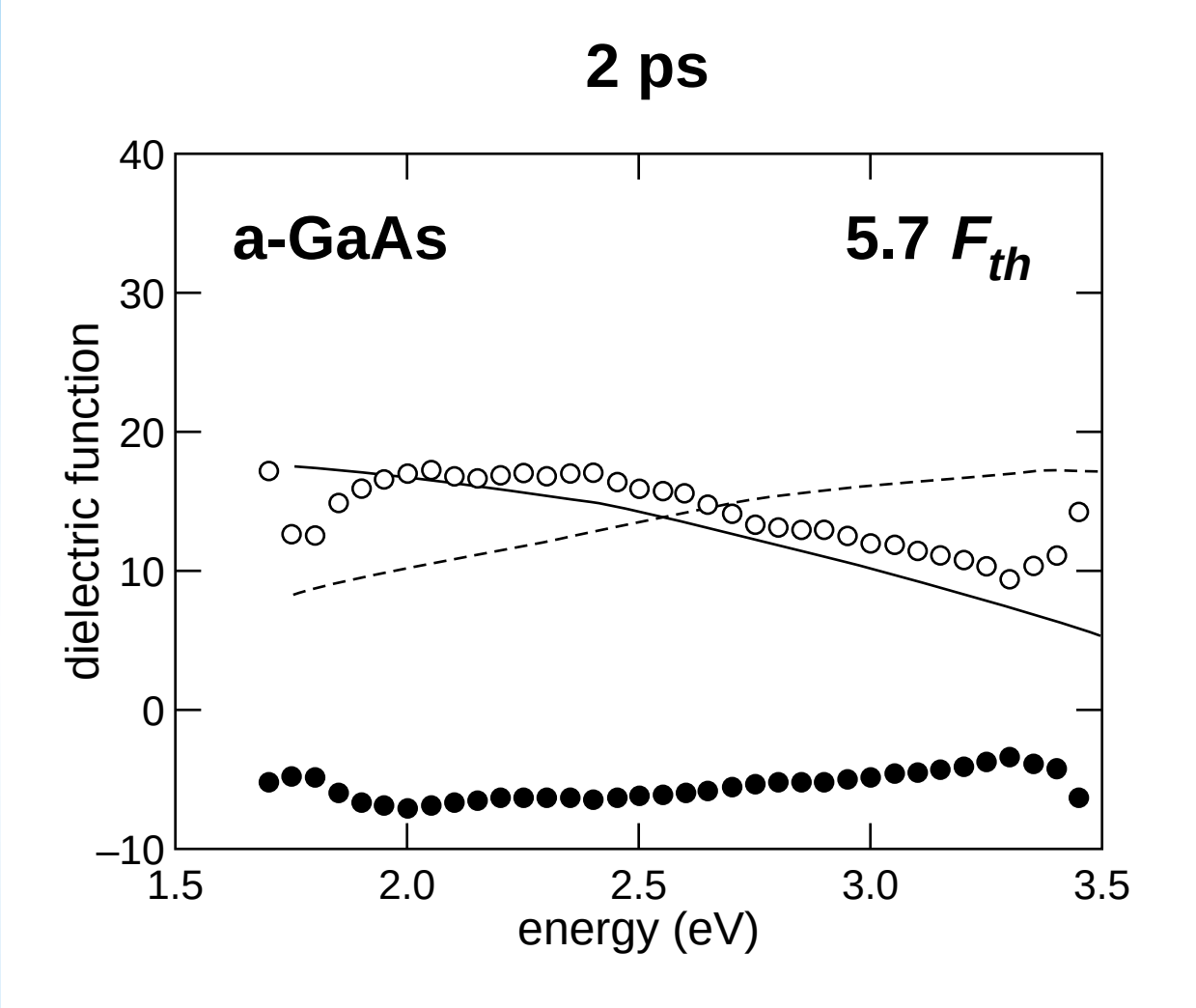
Results


200 fs
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

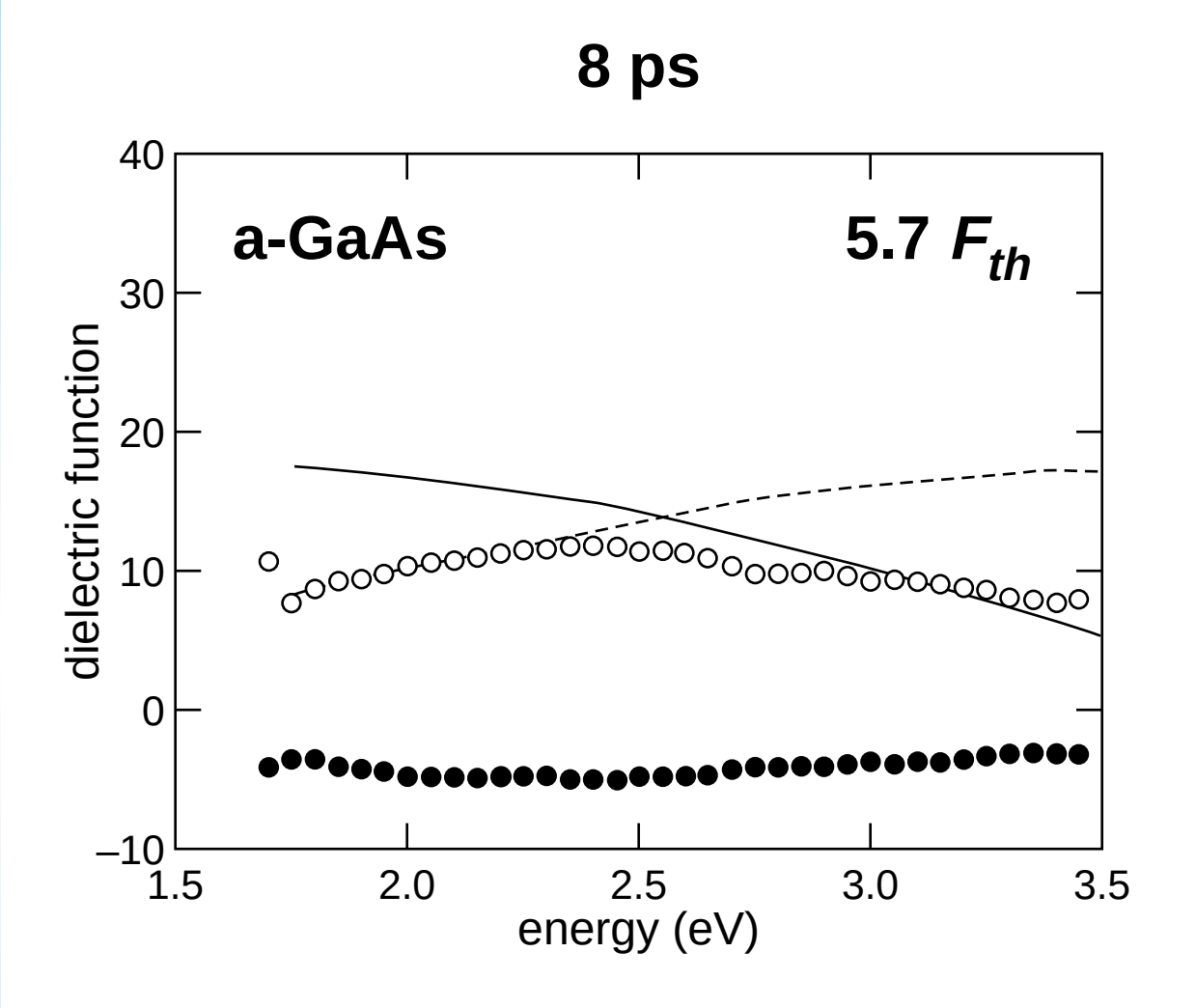
Results


500 fs
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

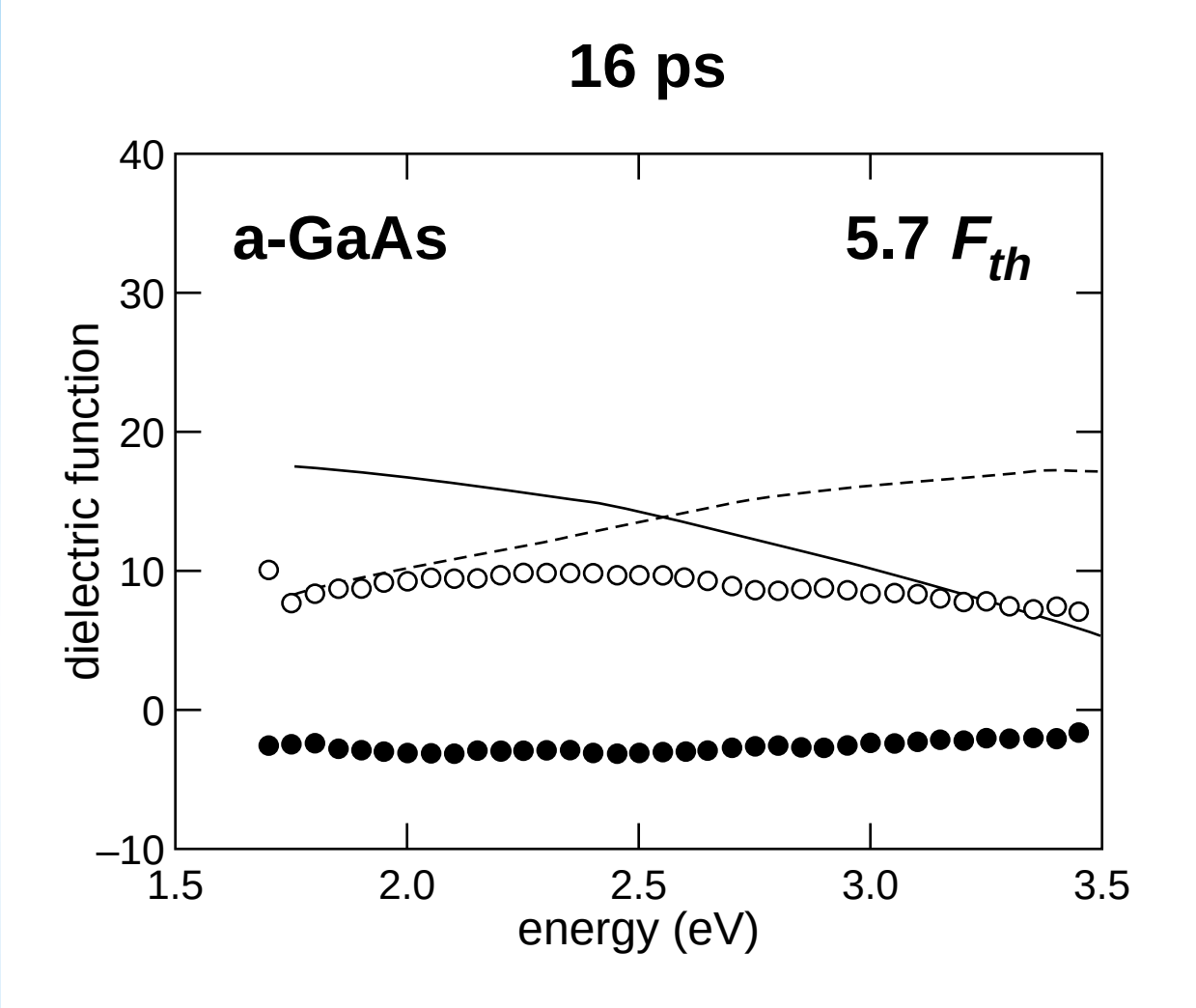
Results


2 ps
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

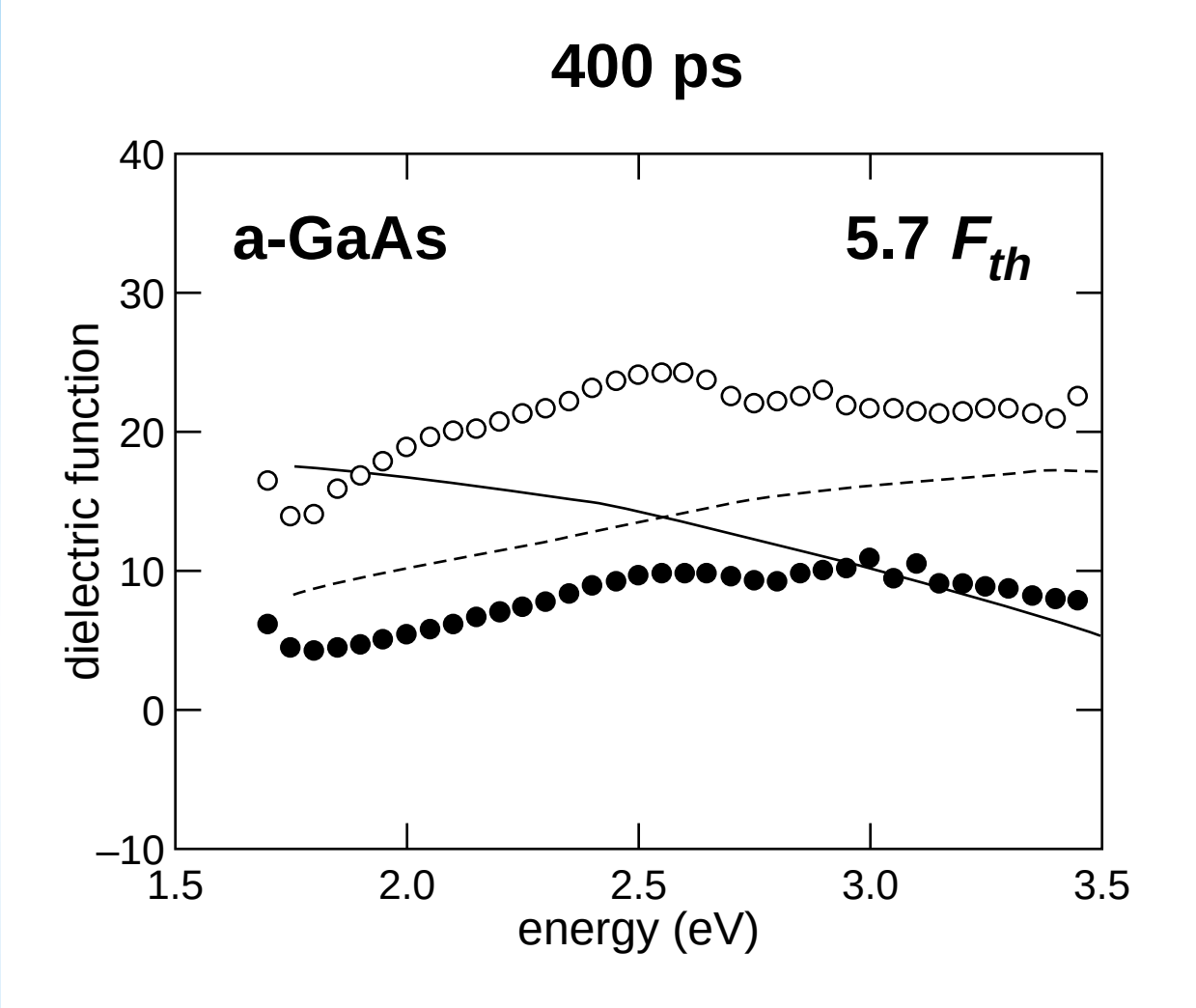
Results


8 ps
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

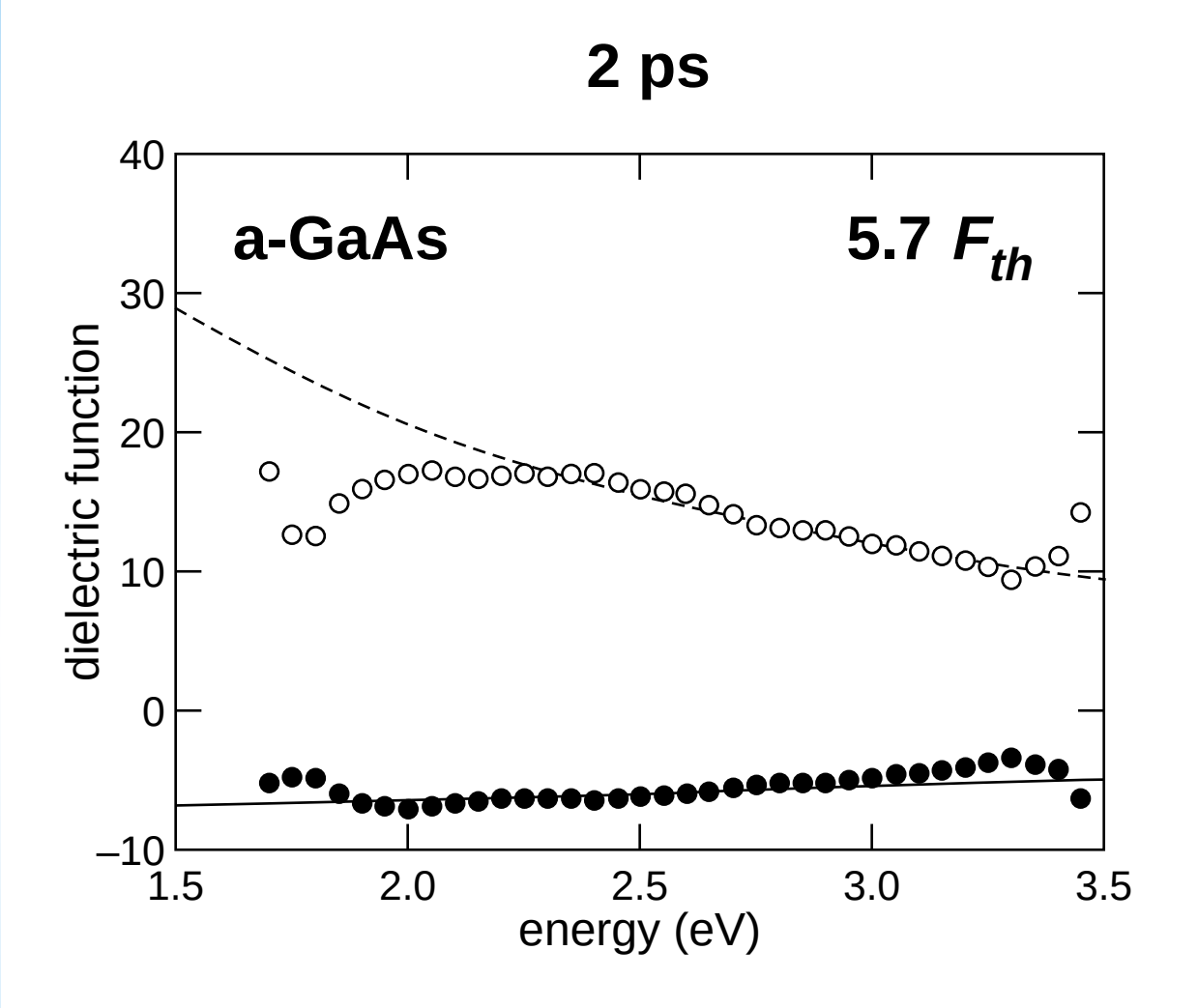
Results


16 ps
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

Results

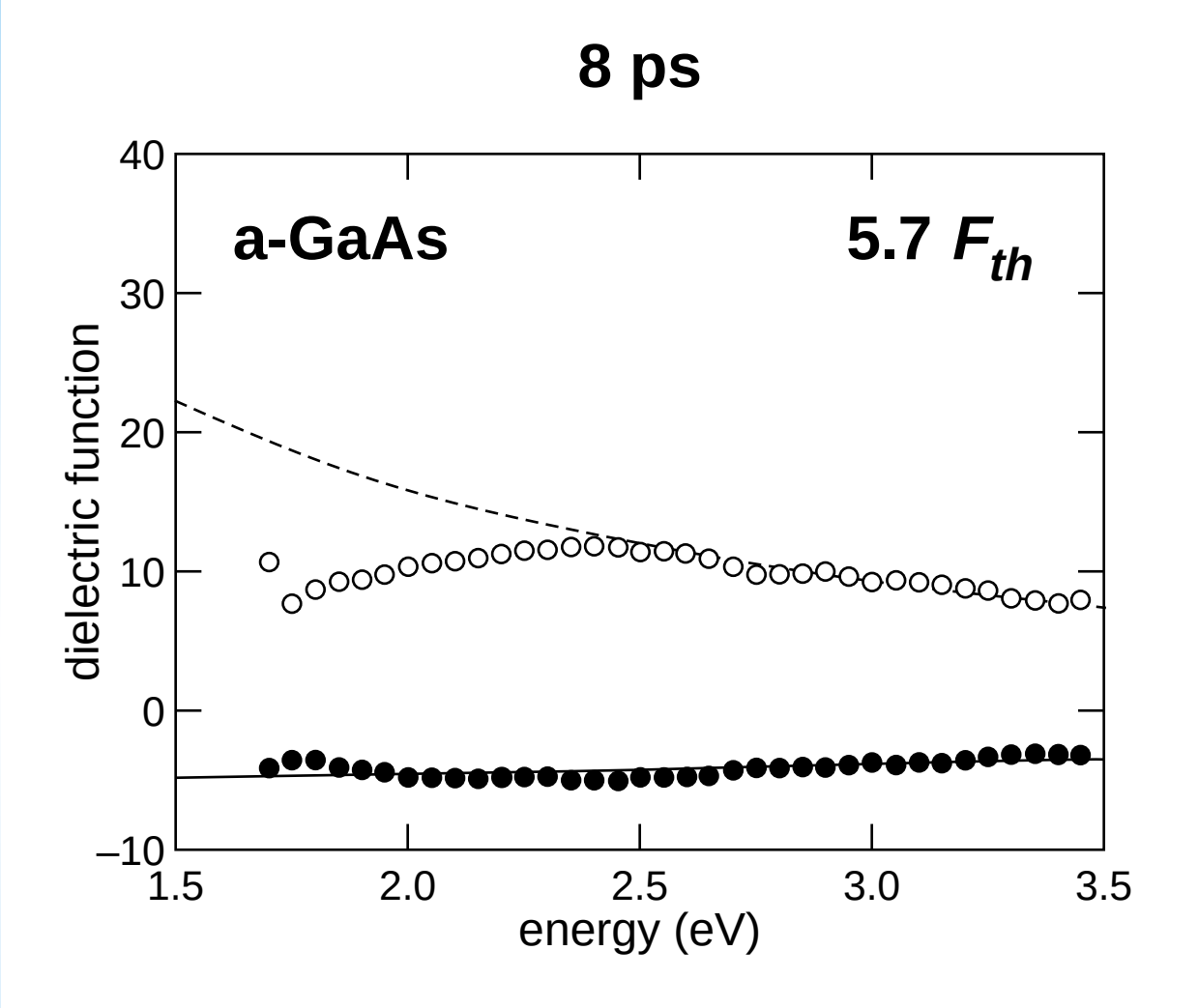

400 ps
a-GaAs
5.7 F_{th}
dielectric function
energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5

Results



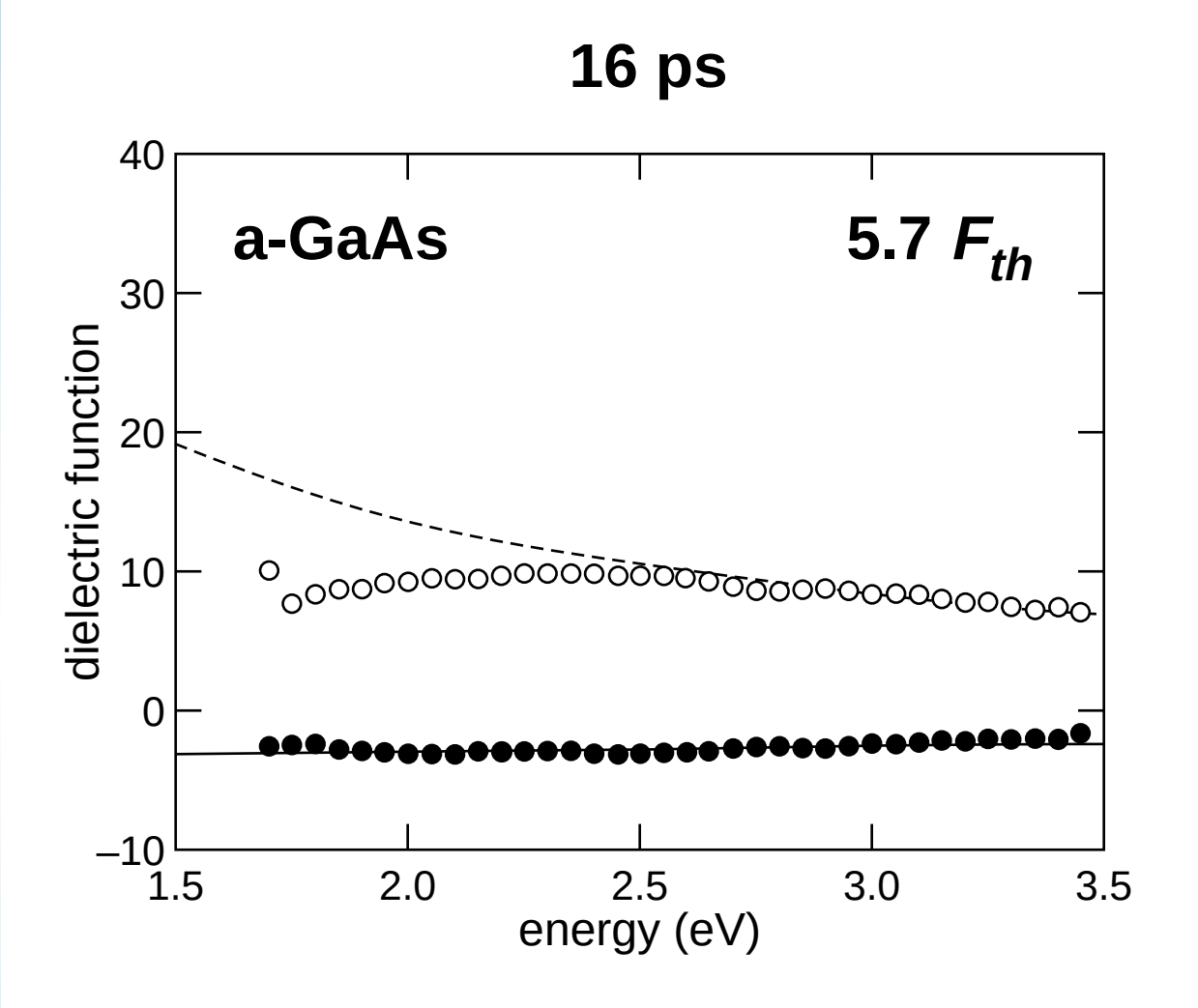
Drude-like after 2 ps

Results



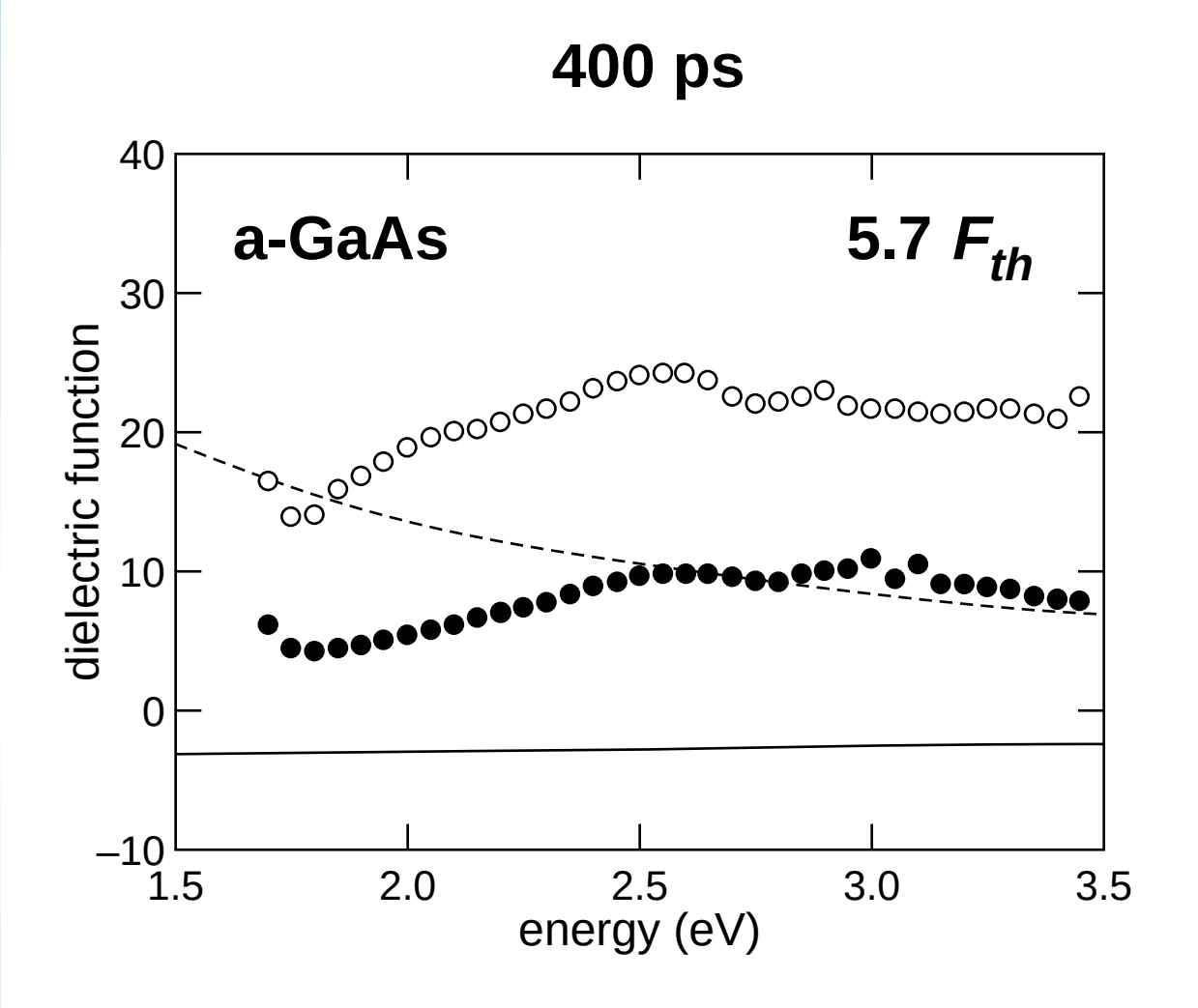
plasma frequency decreases

Results

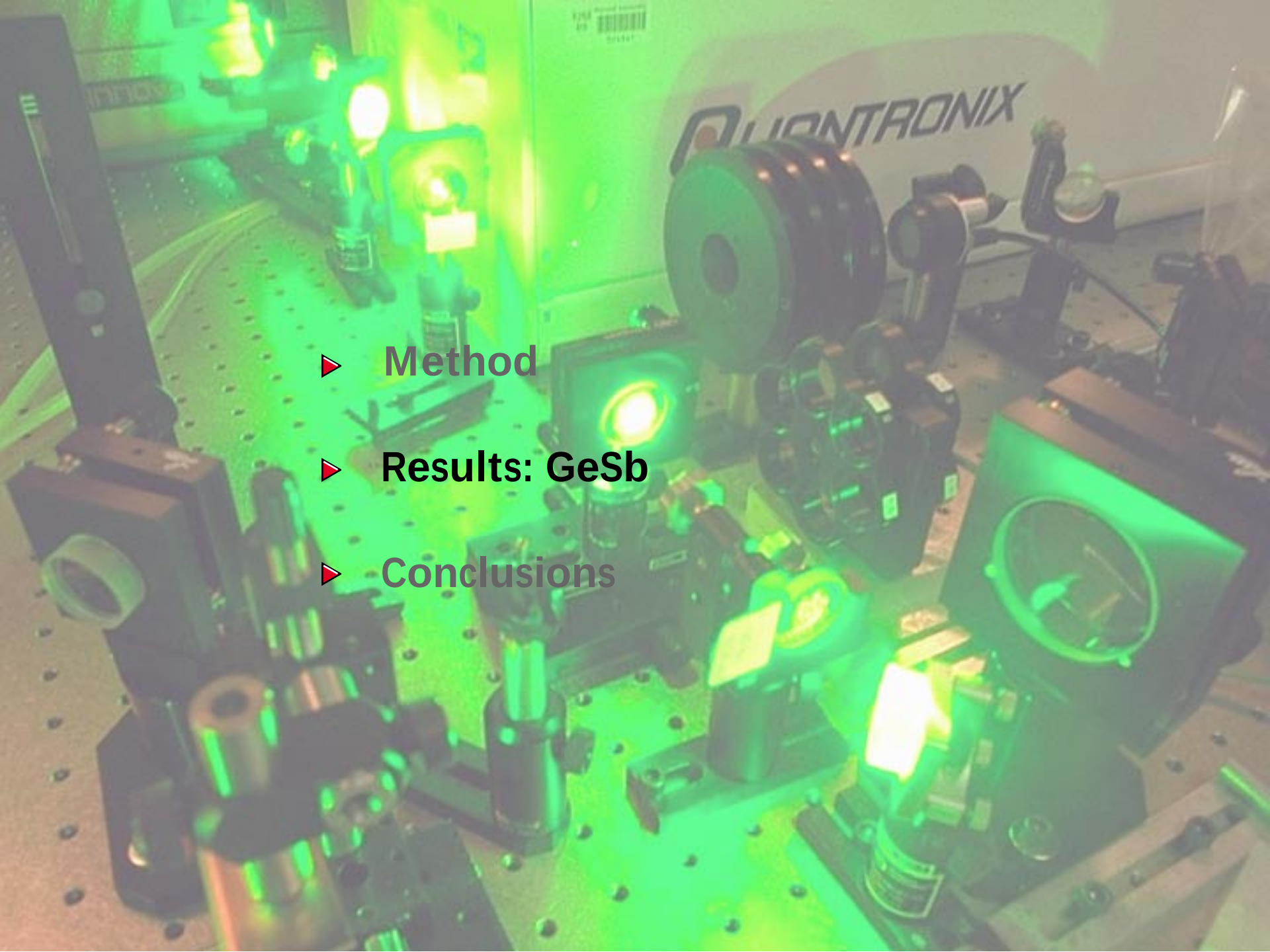


plasma frequency decreases

Results

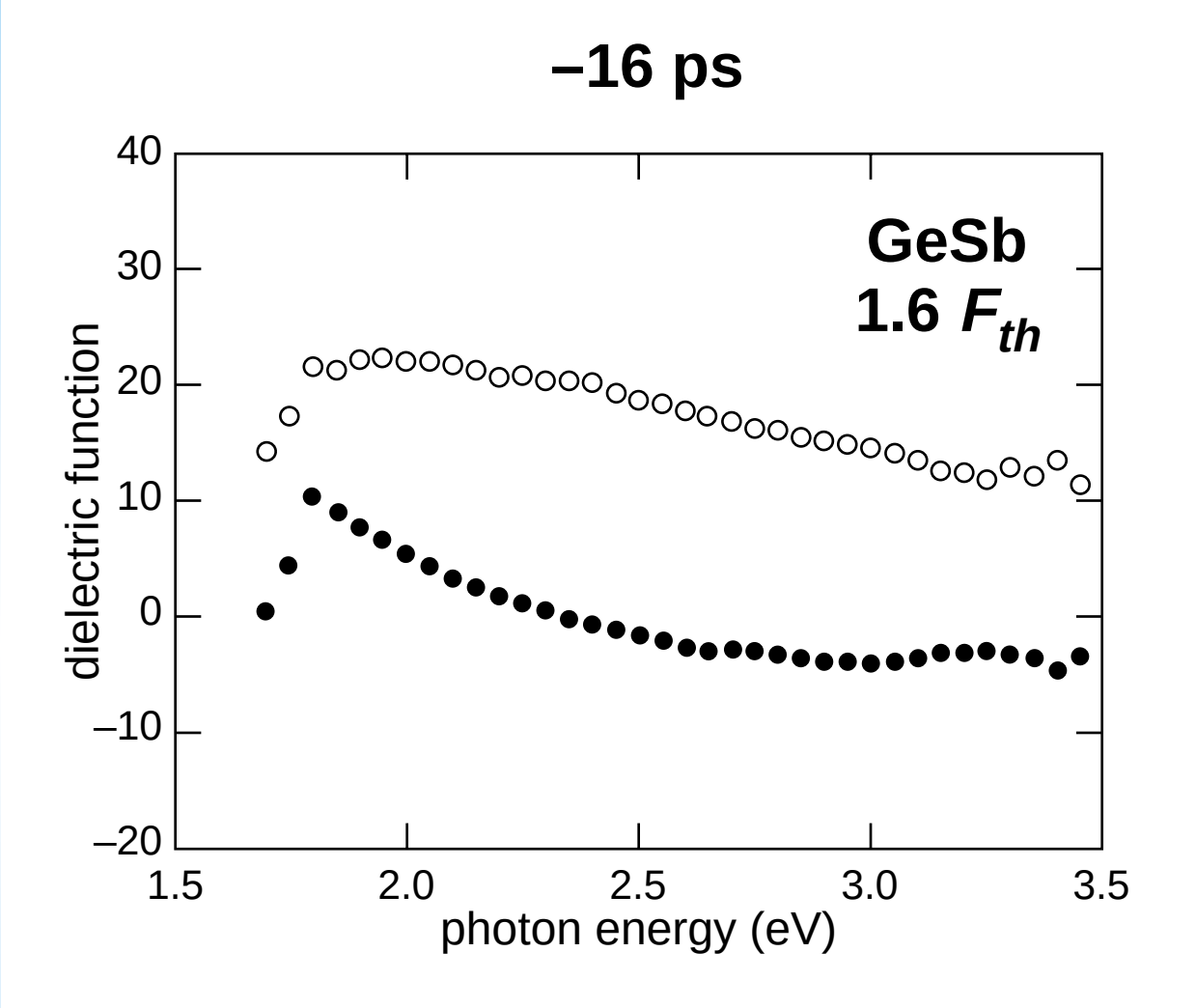


resolidification

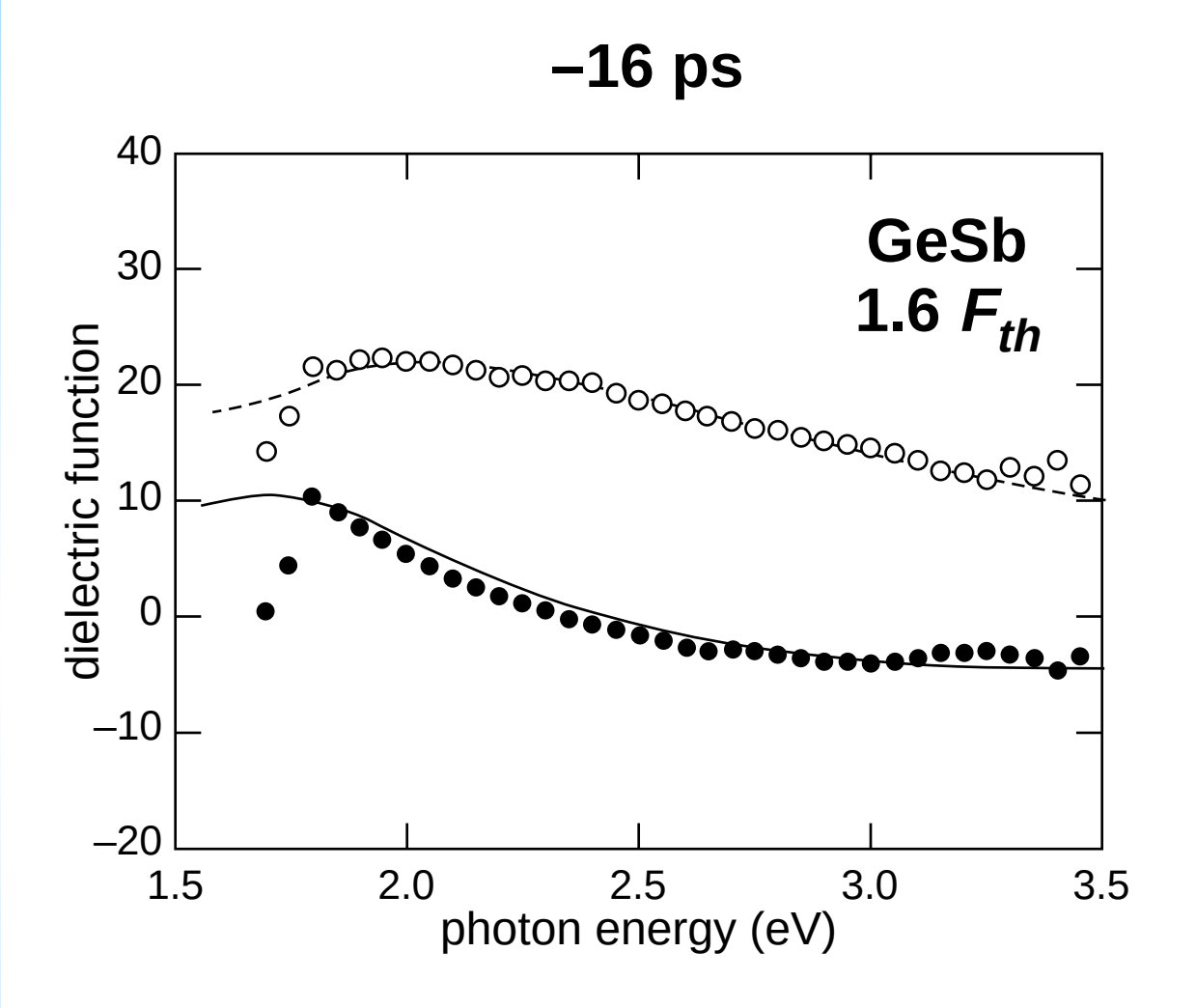


- Method
- **Results: GeSb**
- Conclusions

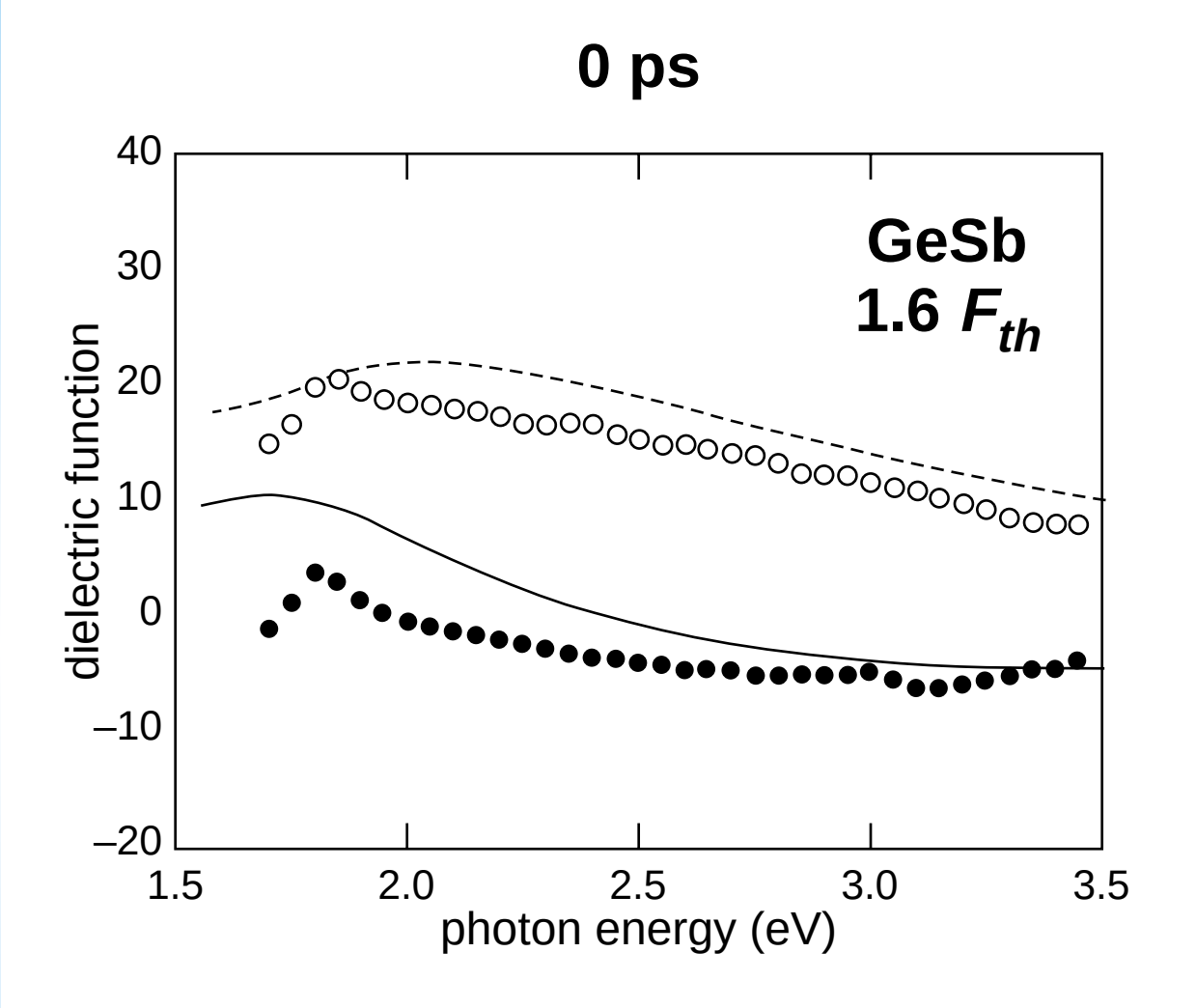
Results


–16 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

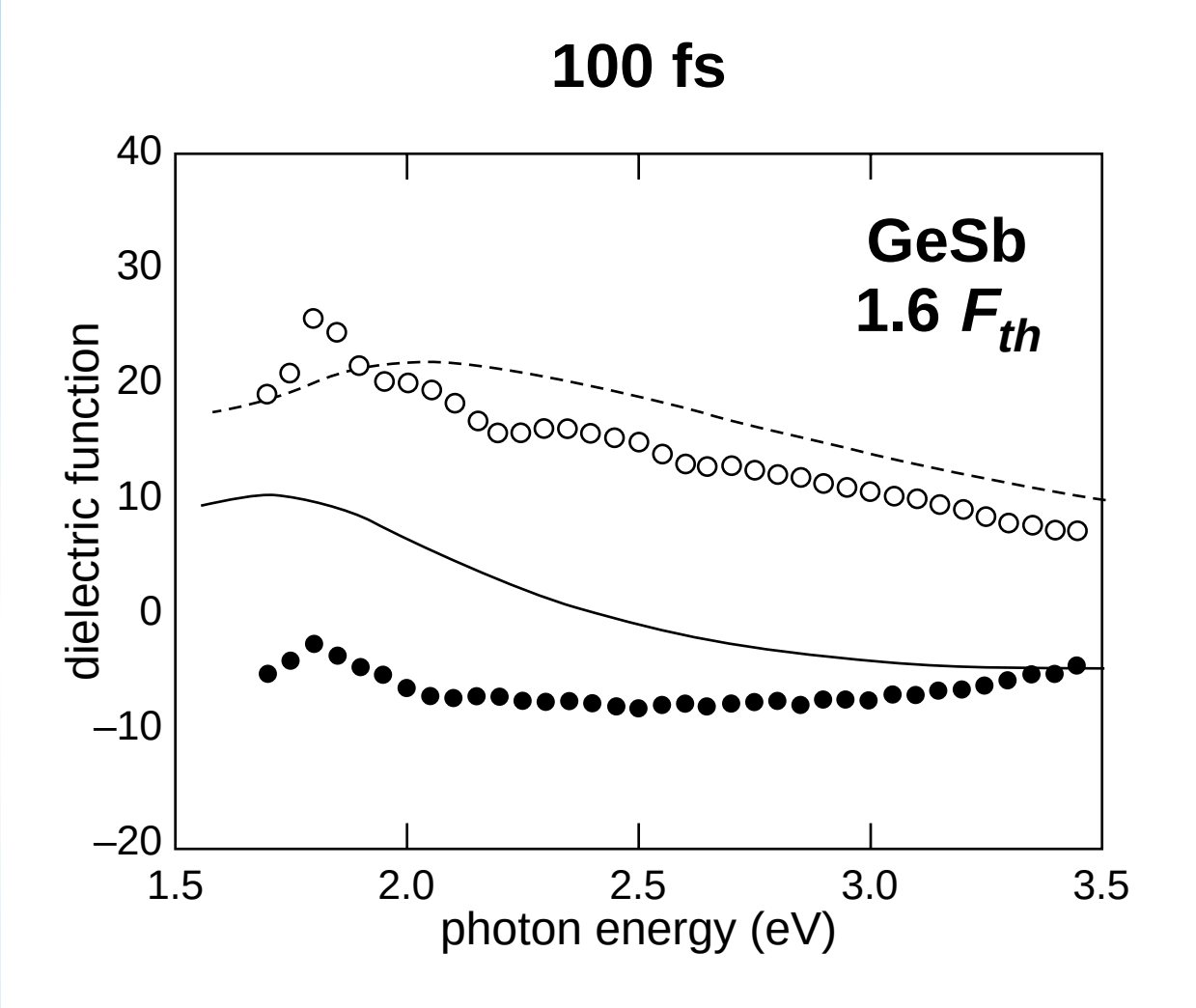
Results


–16 ps
GeSb
1.6 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5

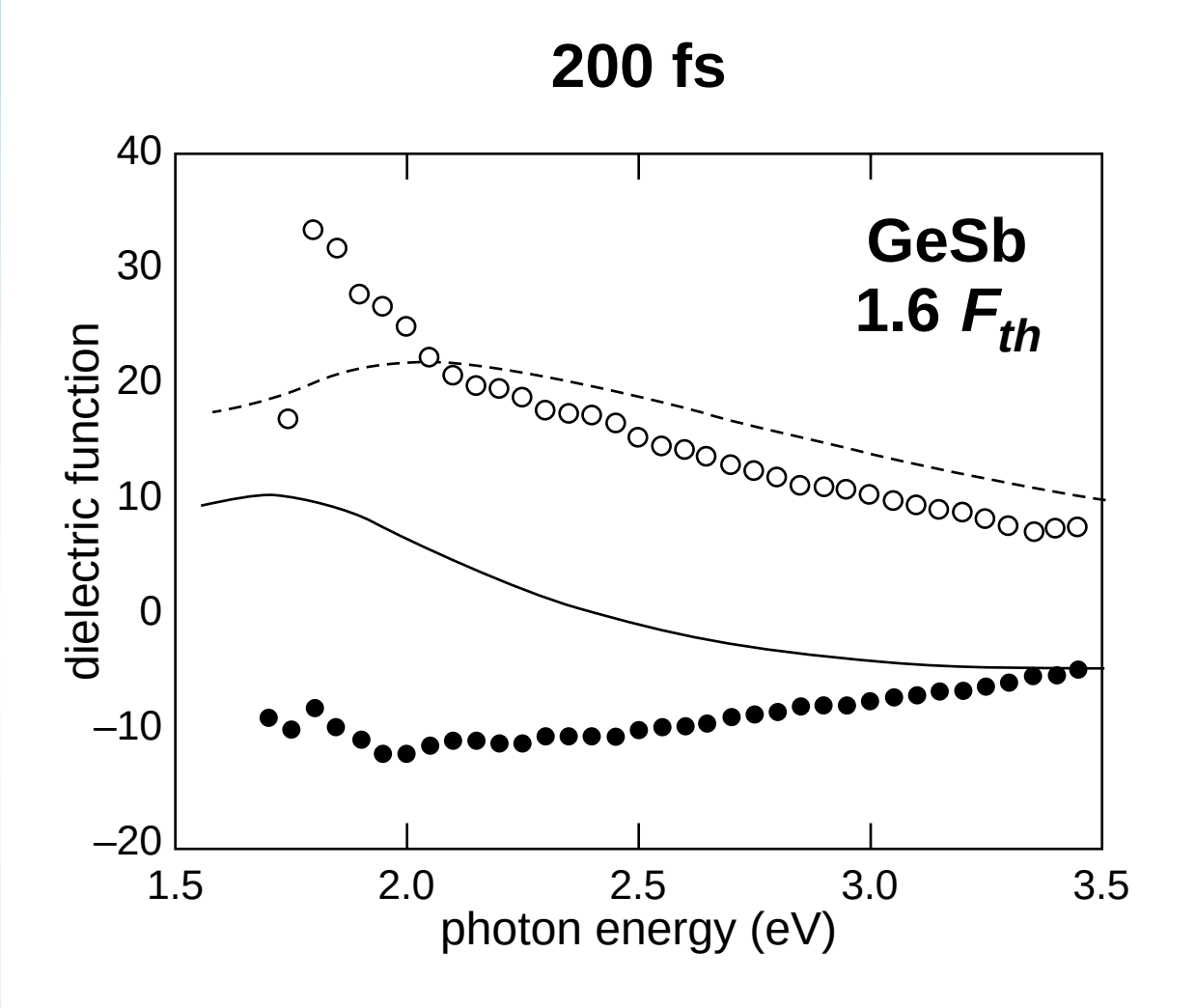
Results


0 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

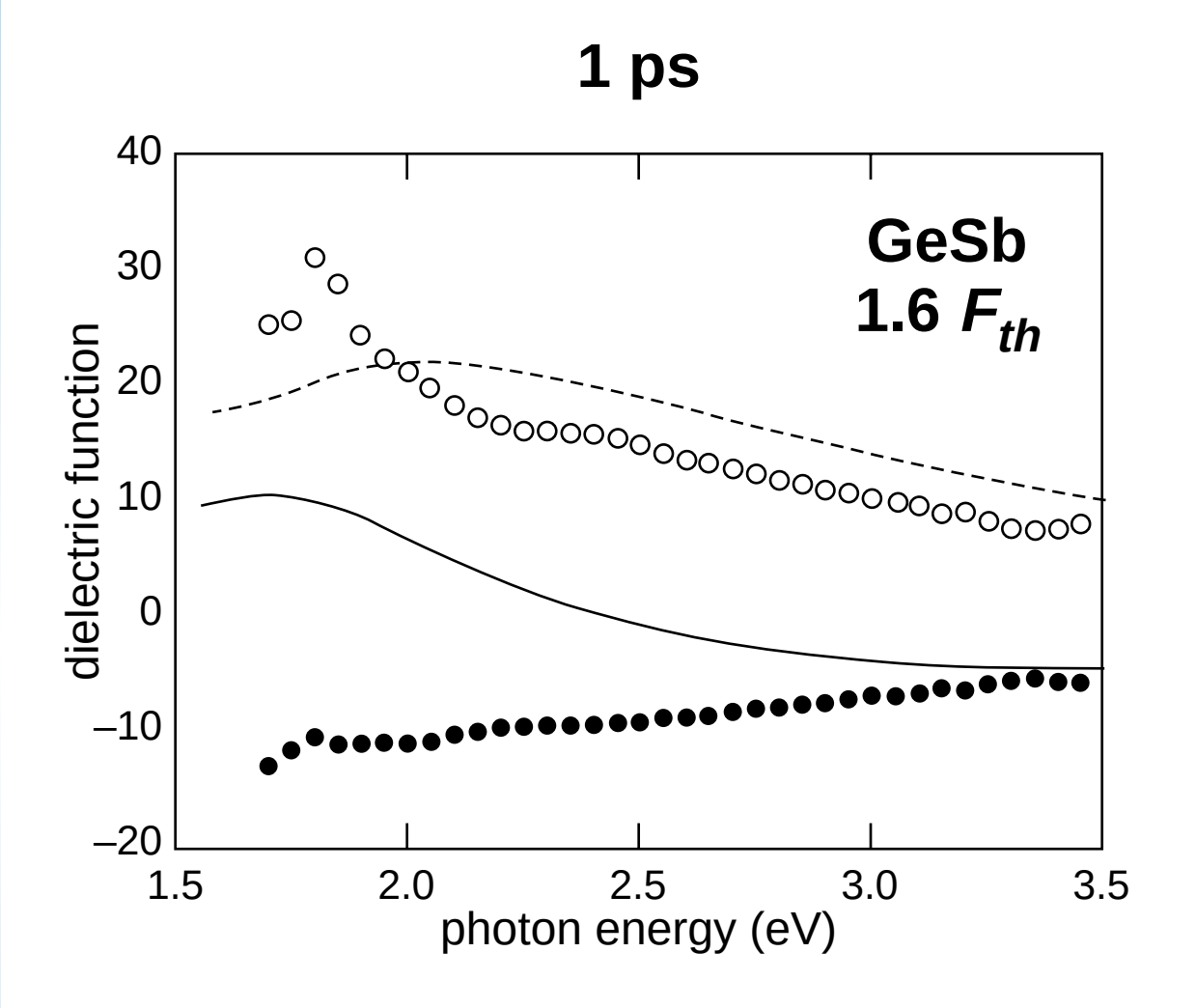
Results


100 fs
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

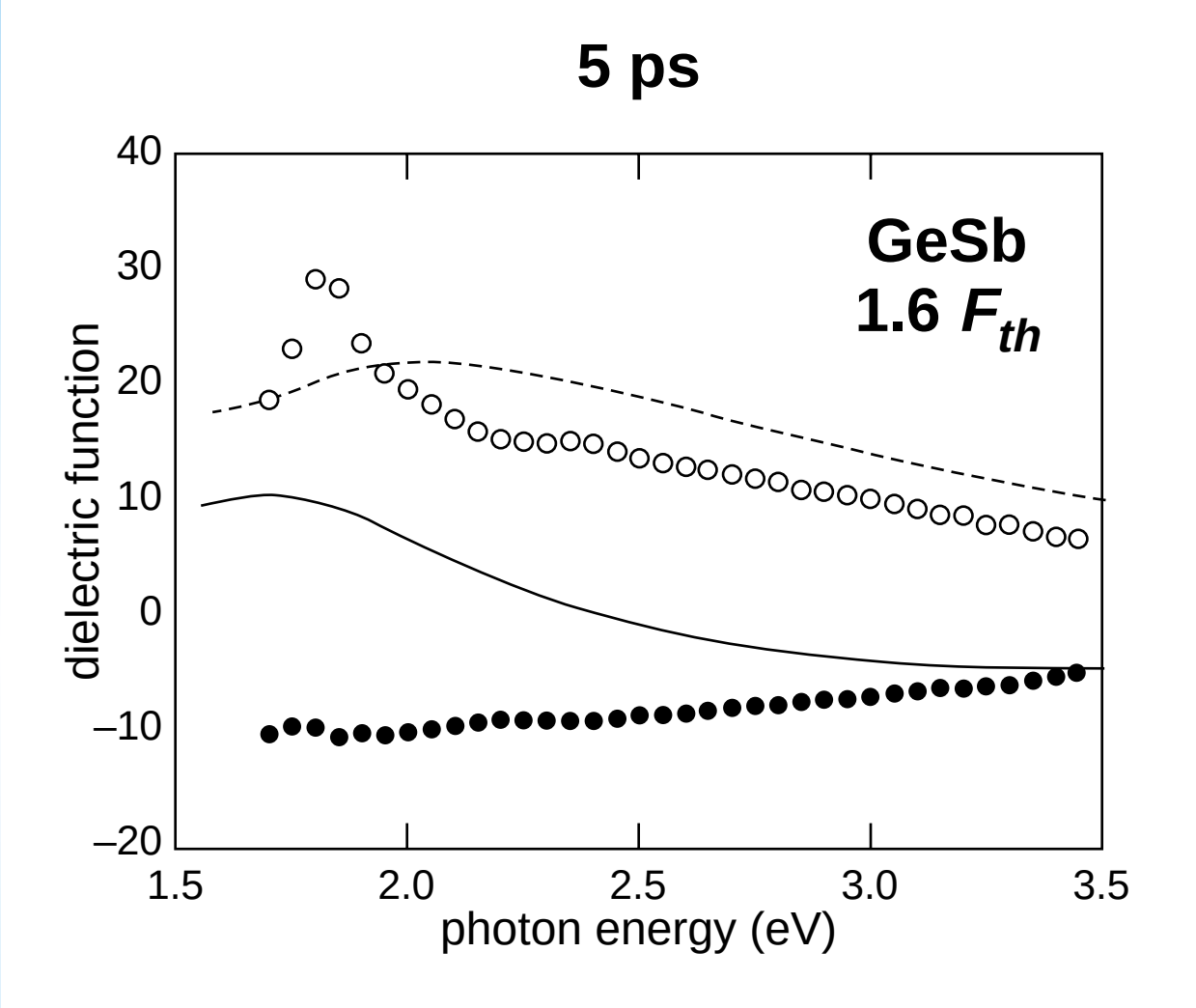
Results


200 fs
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

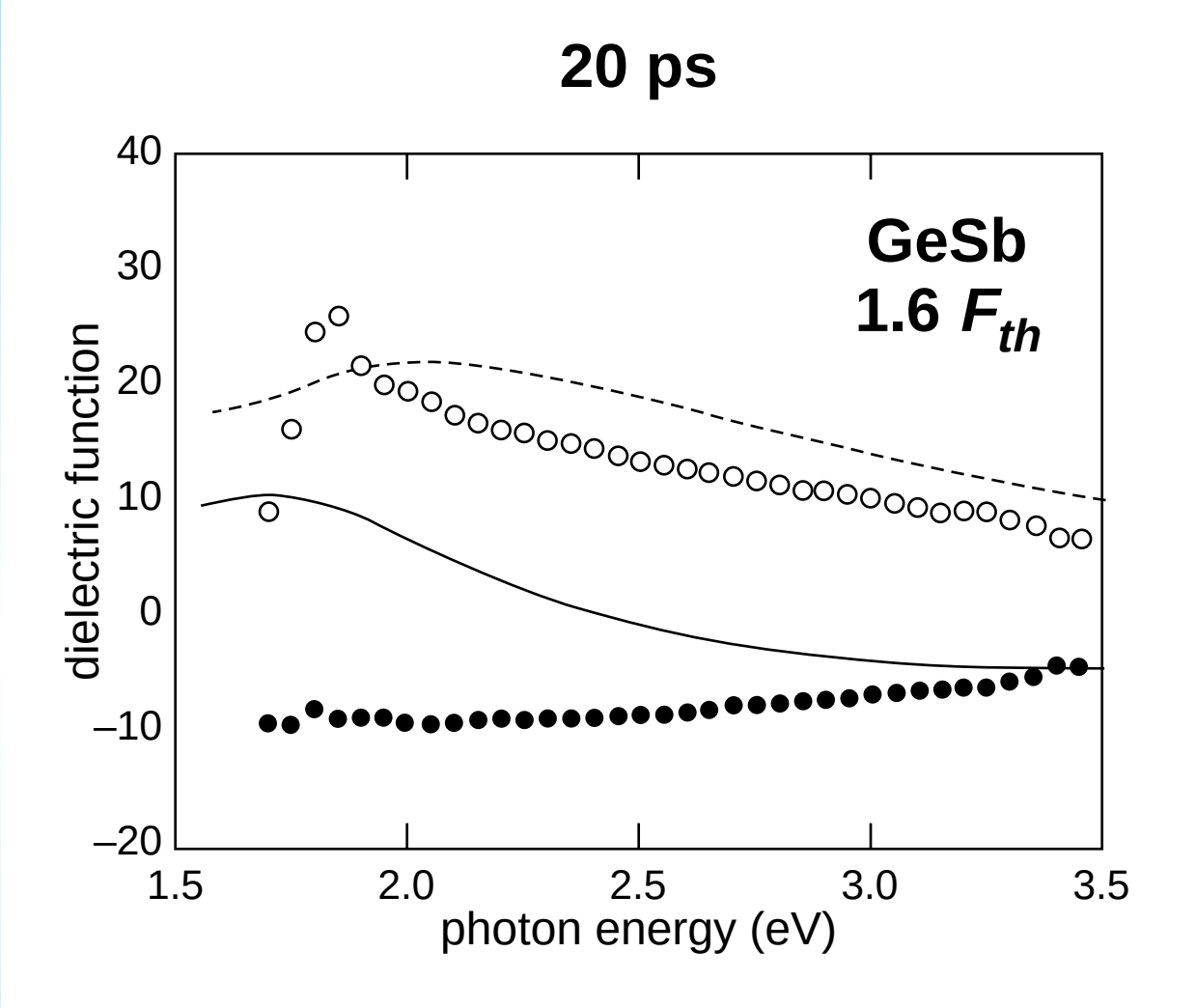
Results


1 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

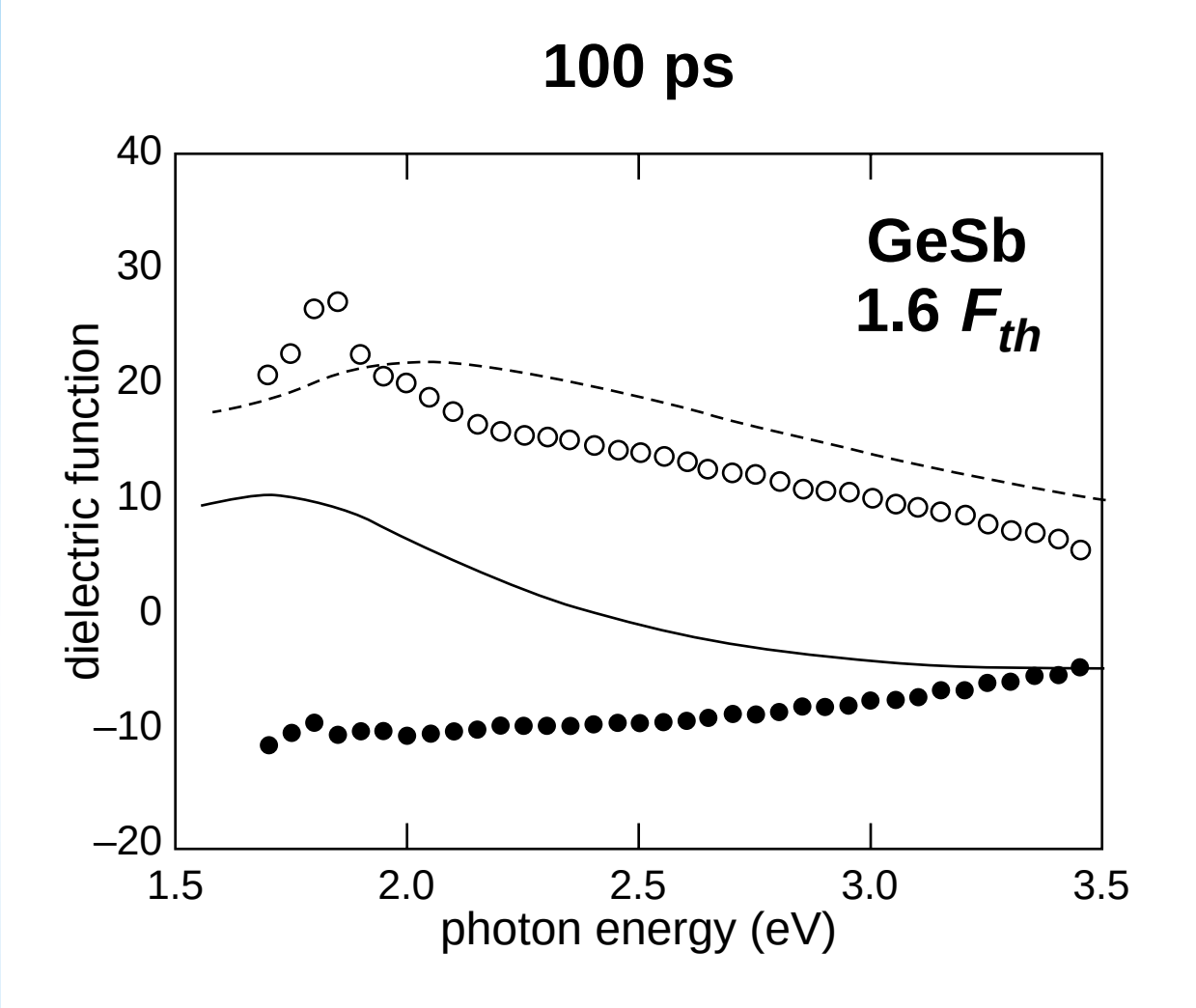
Results


5 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

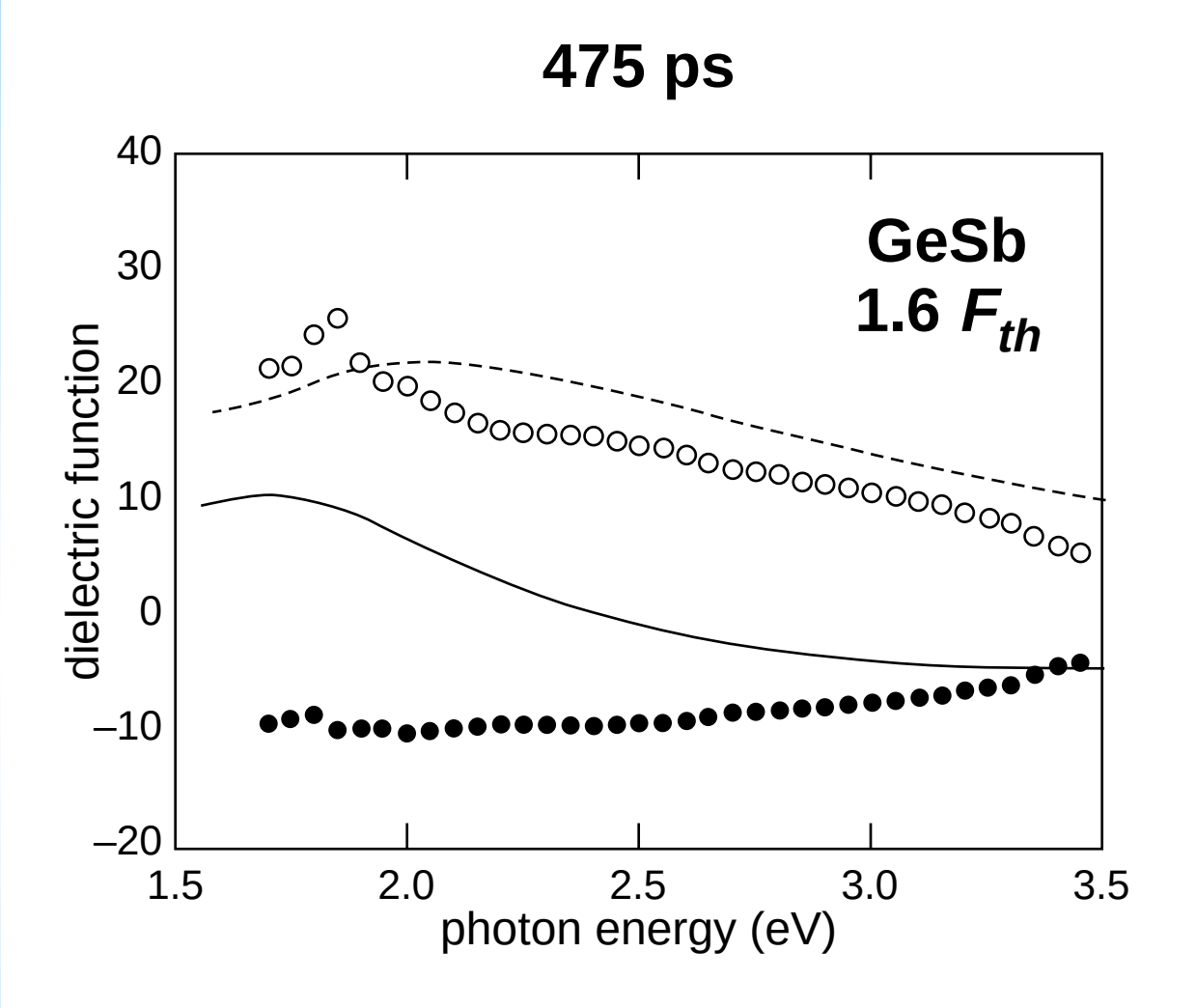
Results


20 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

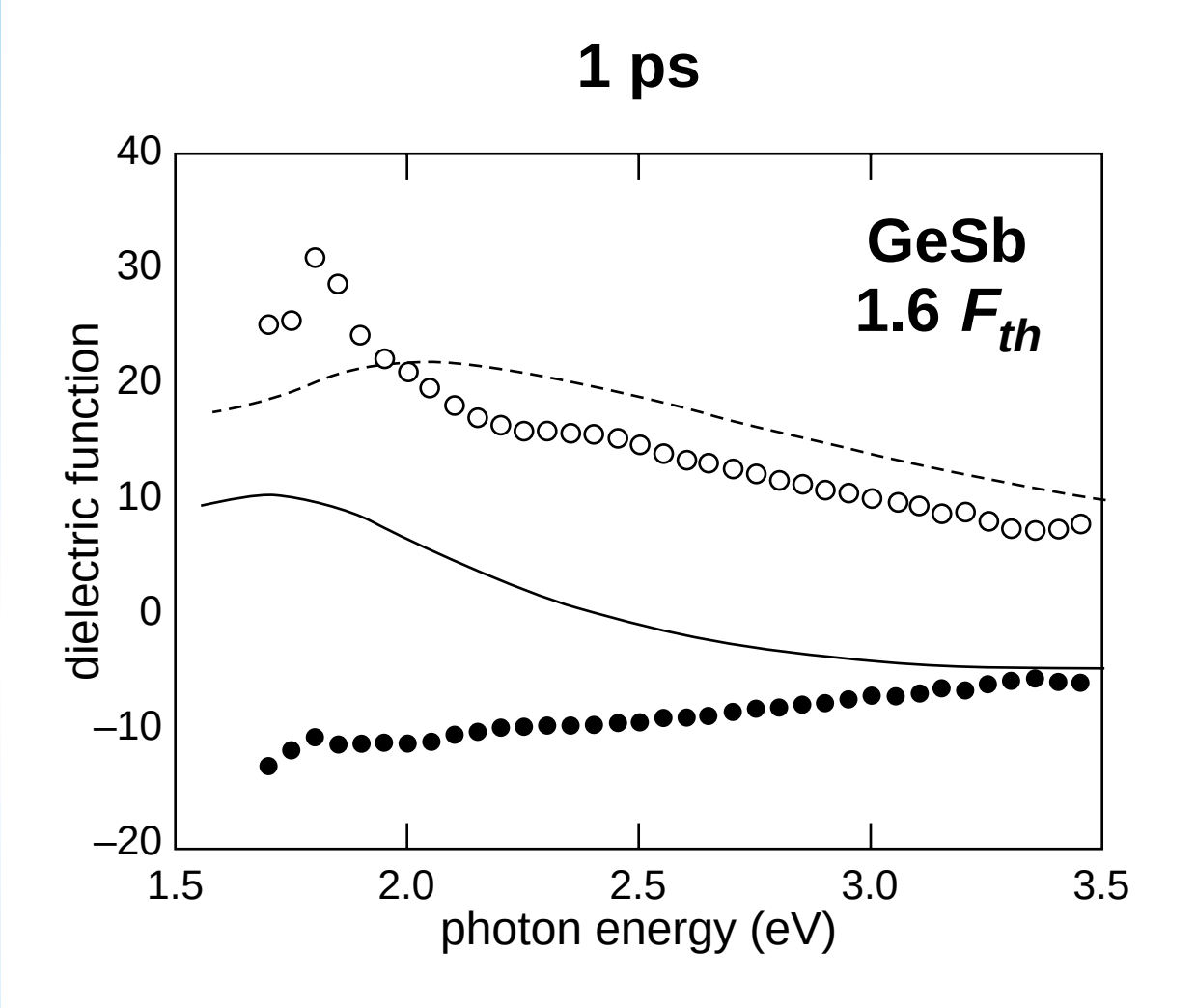
Results


100 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

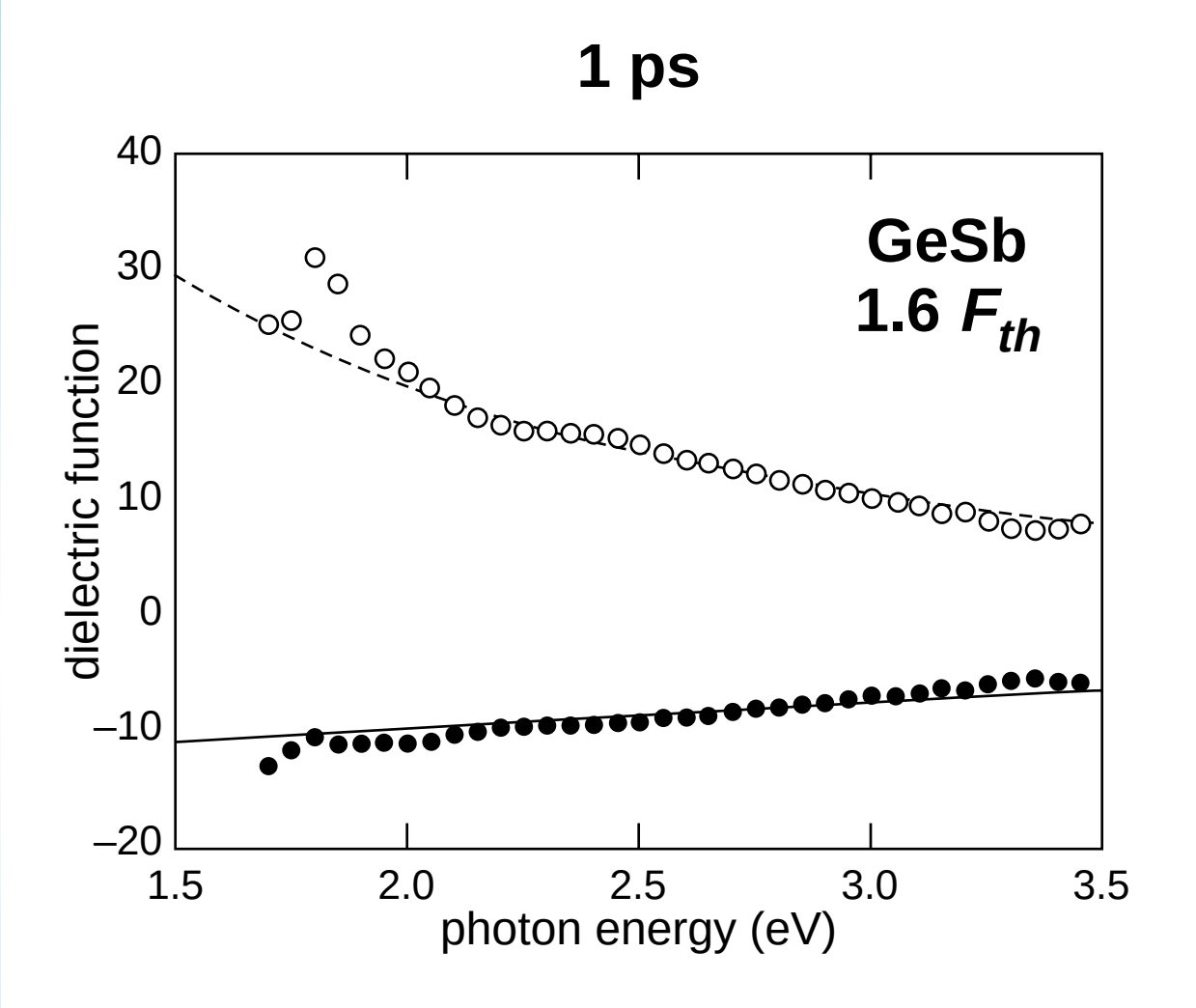
Results


475 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

Results

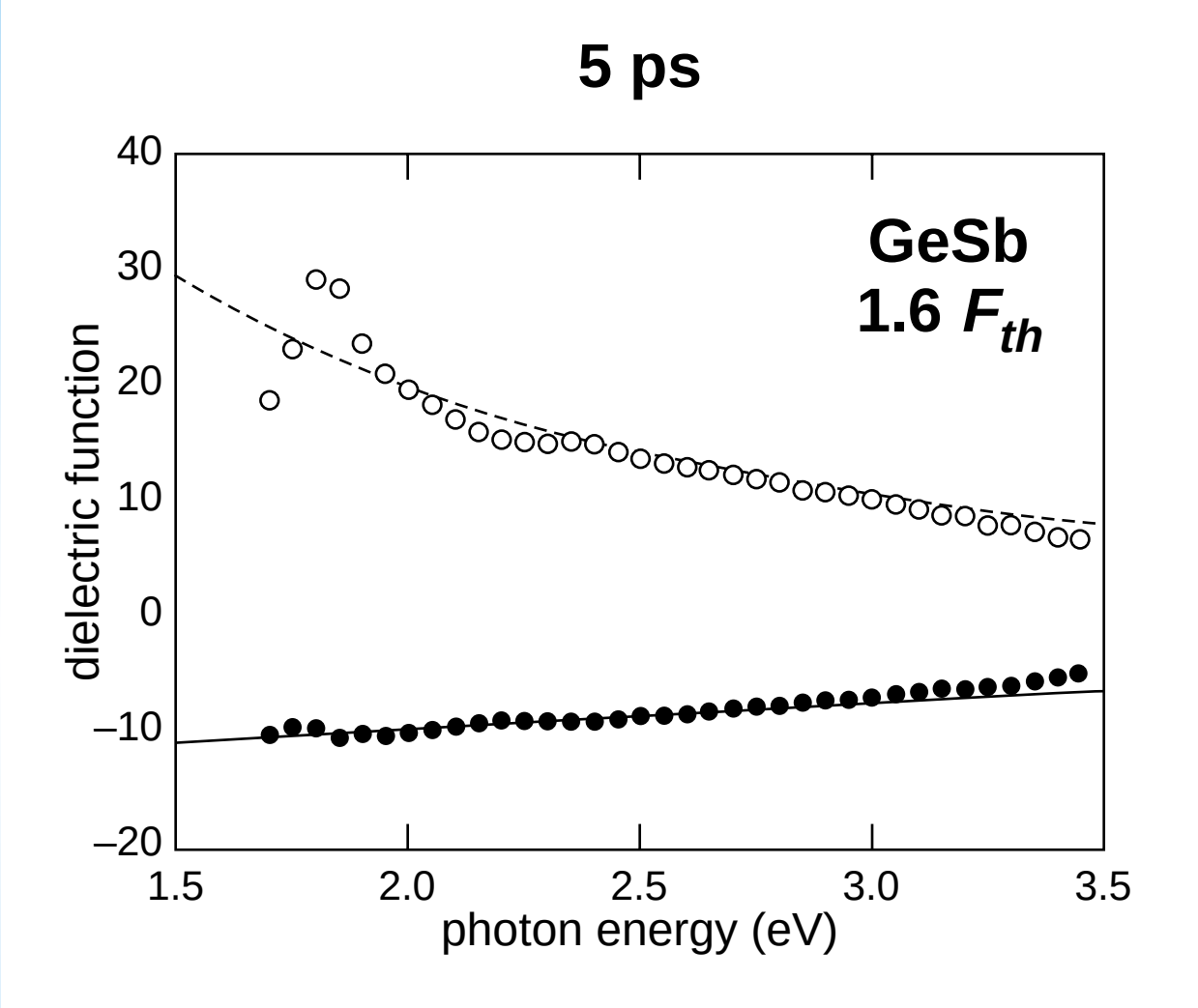

1 ps
GeSb
1.6 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

Results



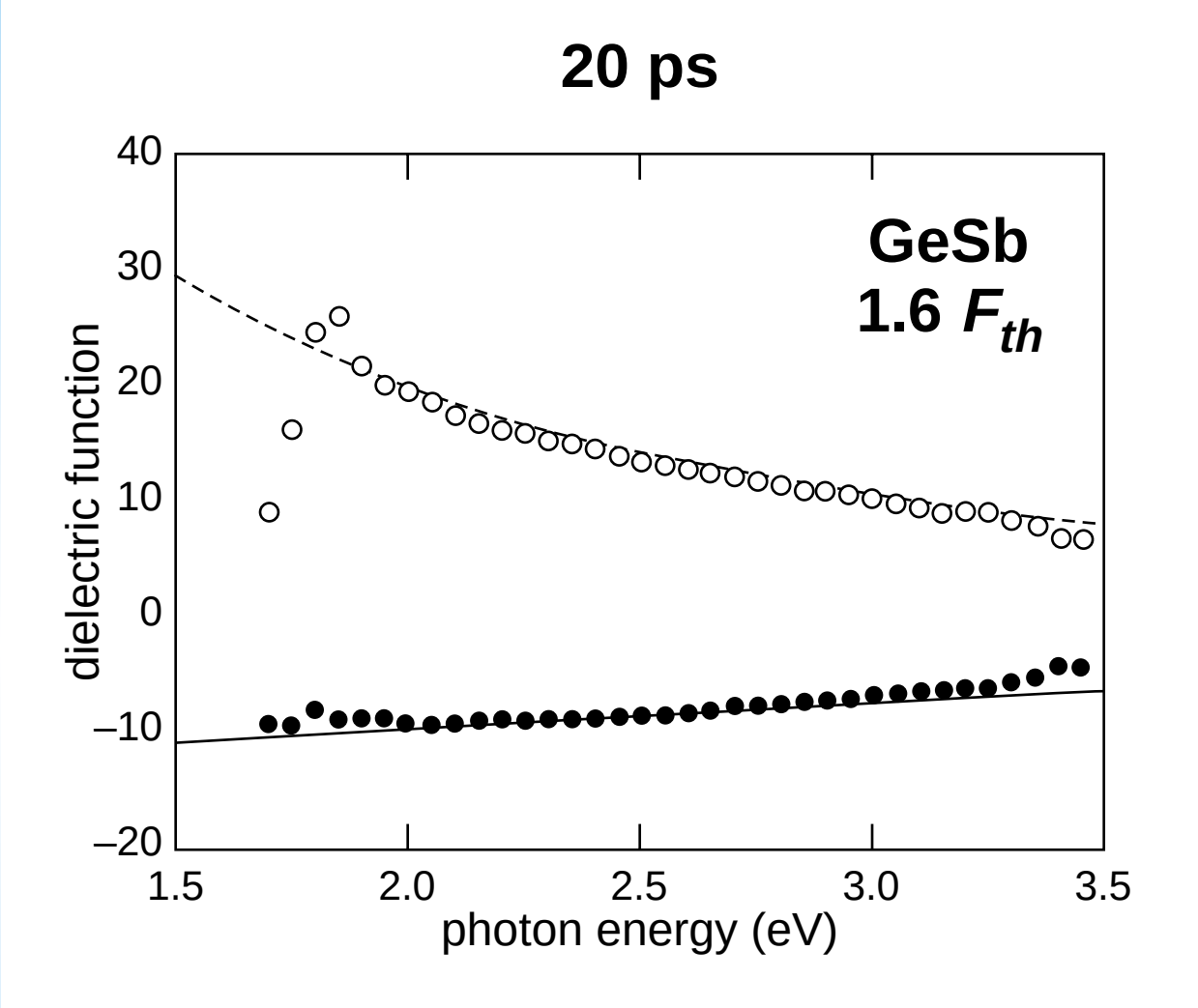
Drude-like after 1 ps

Results



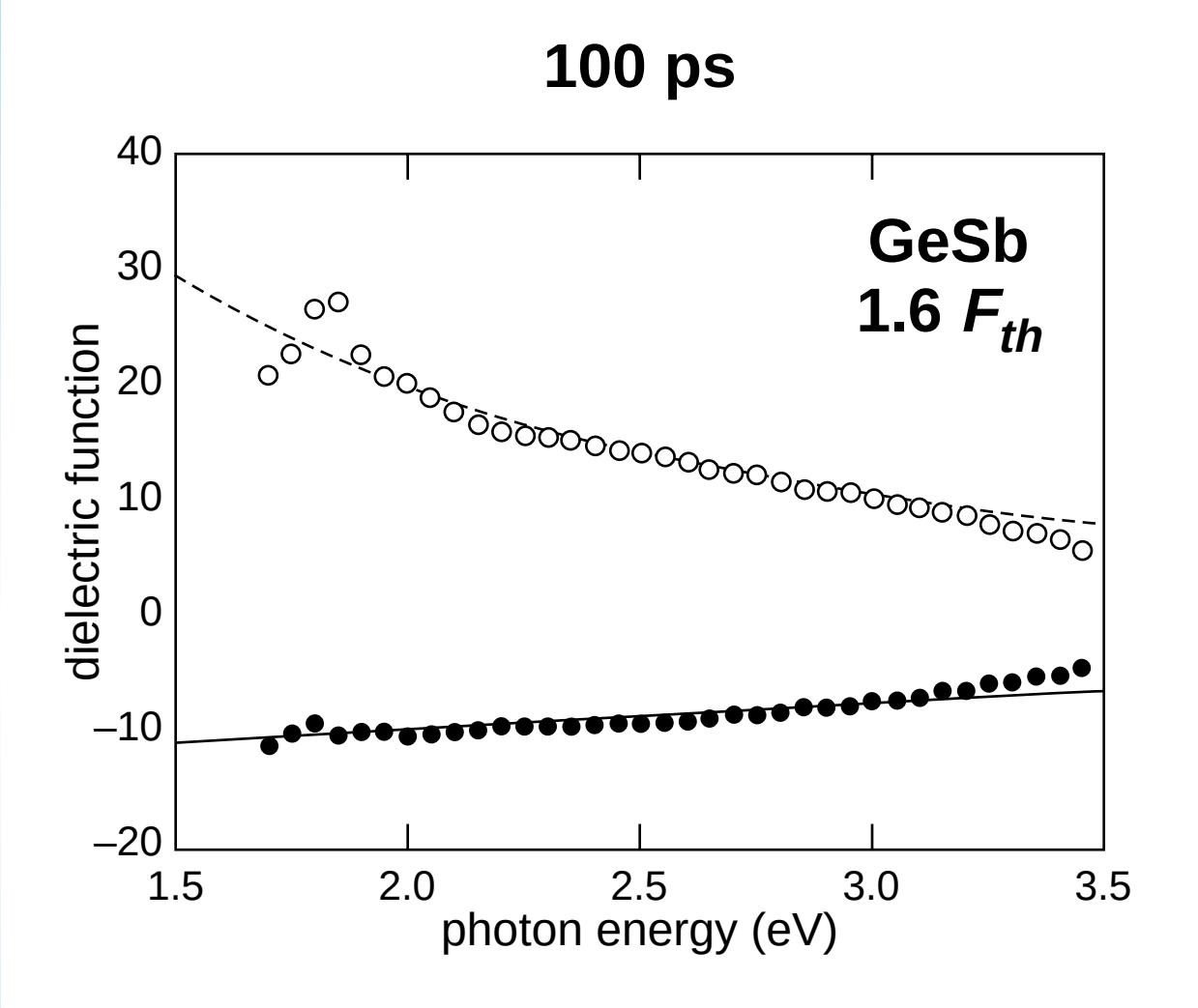
Drude-like after 1 ps

Results



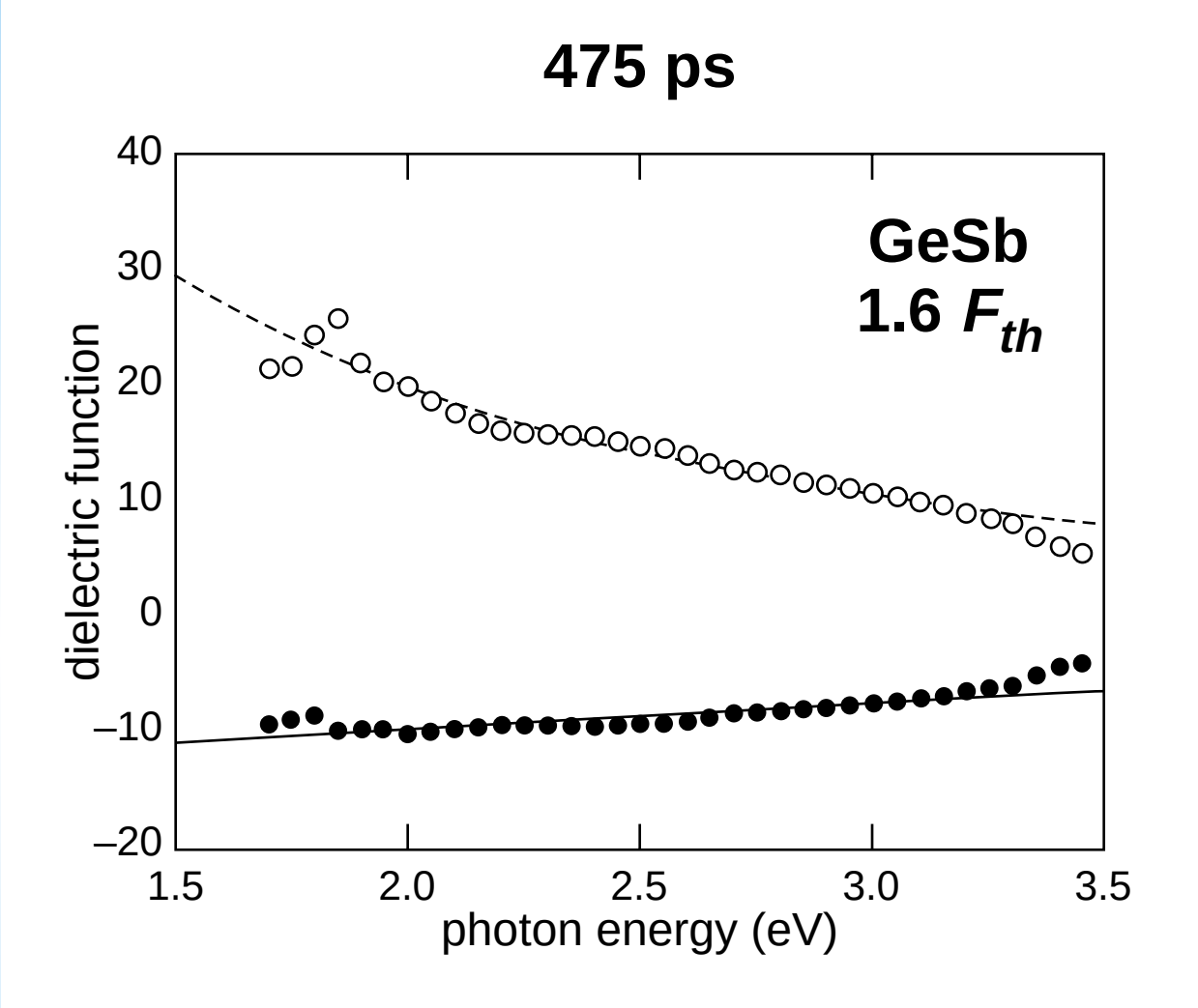
Drude-like after 1 ps

Results



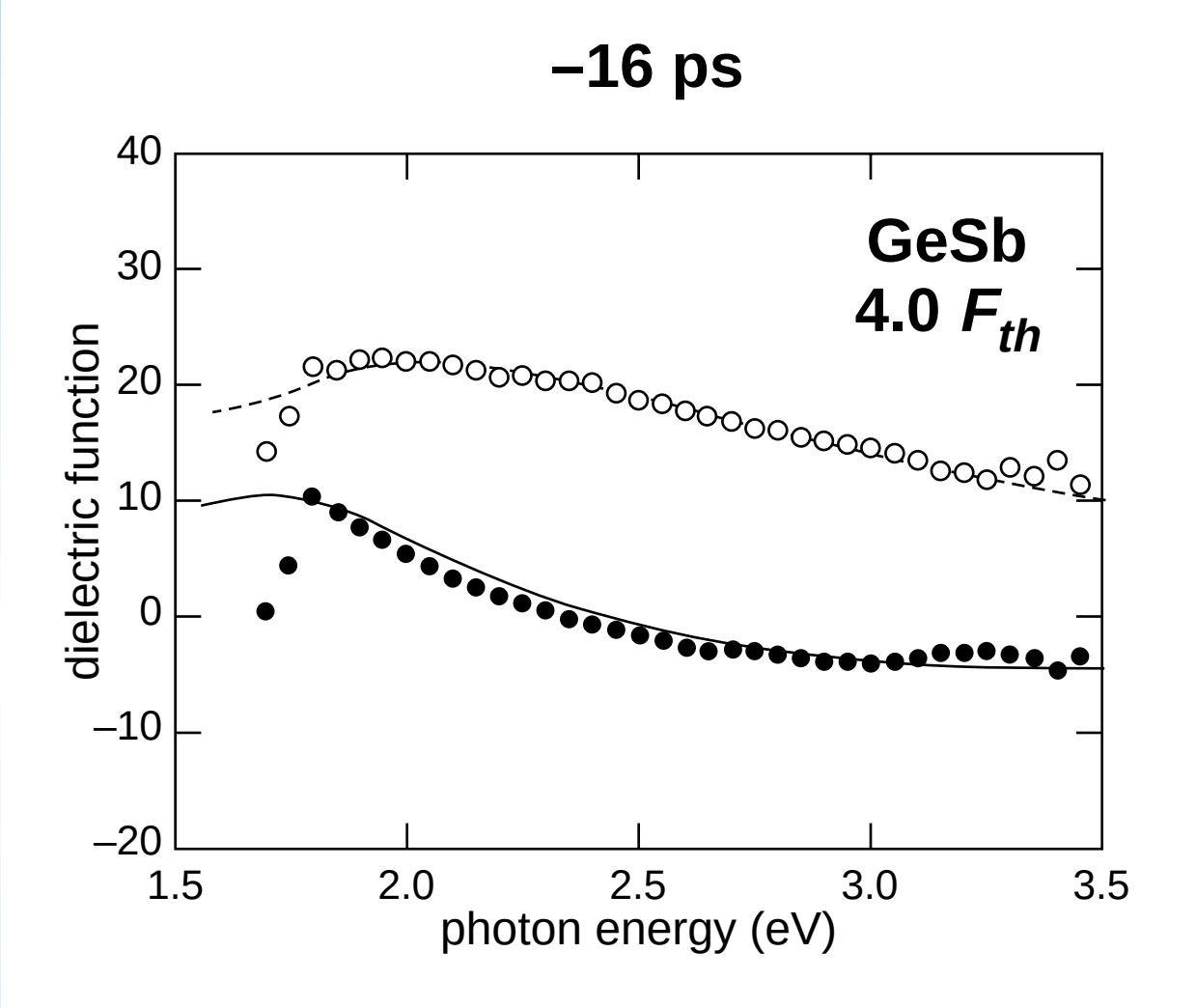
Drude-like after 1 ps

Results

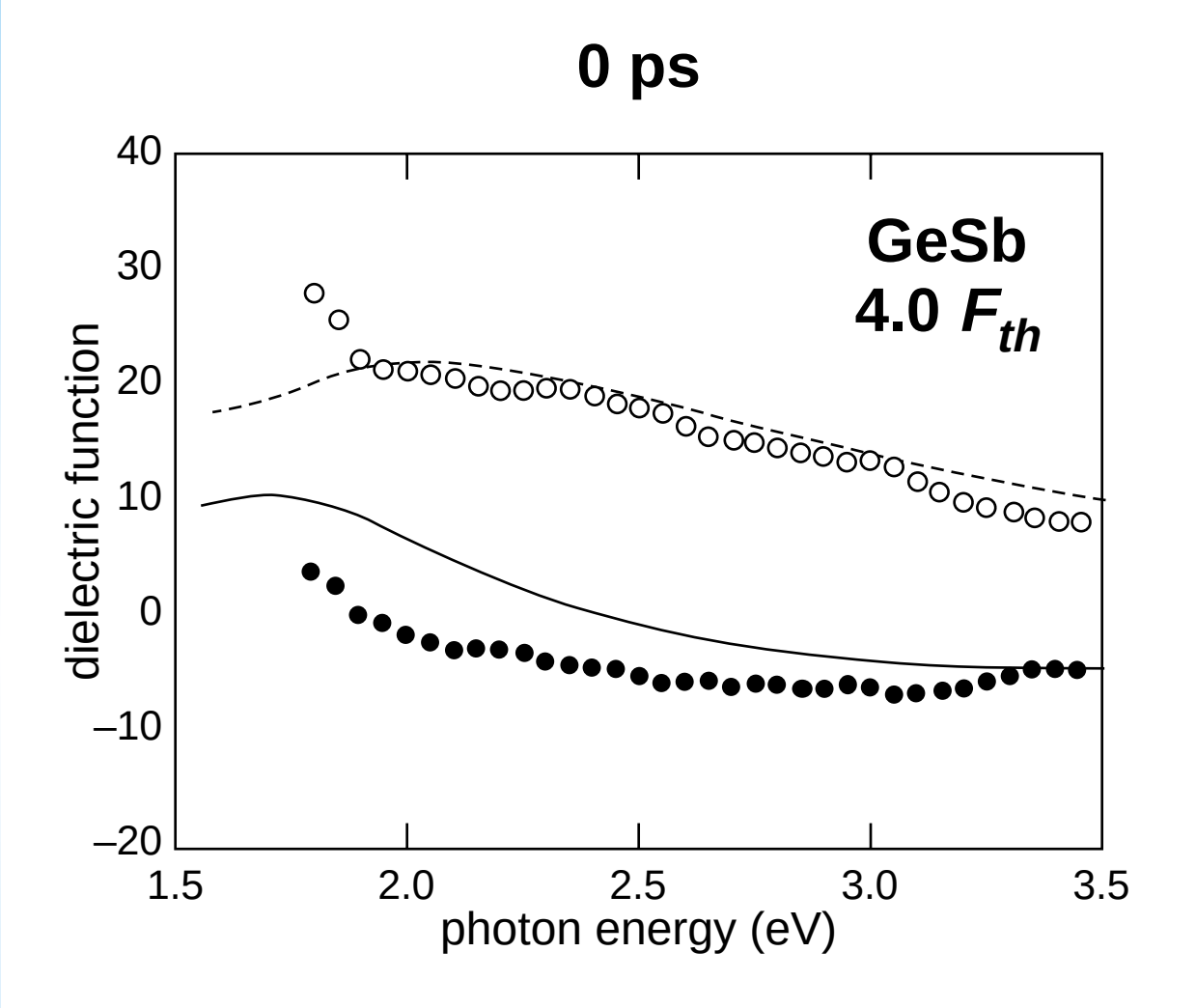


Drude-like after 1 ps

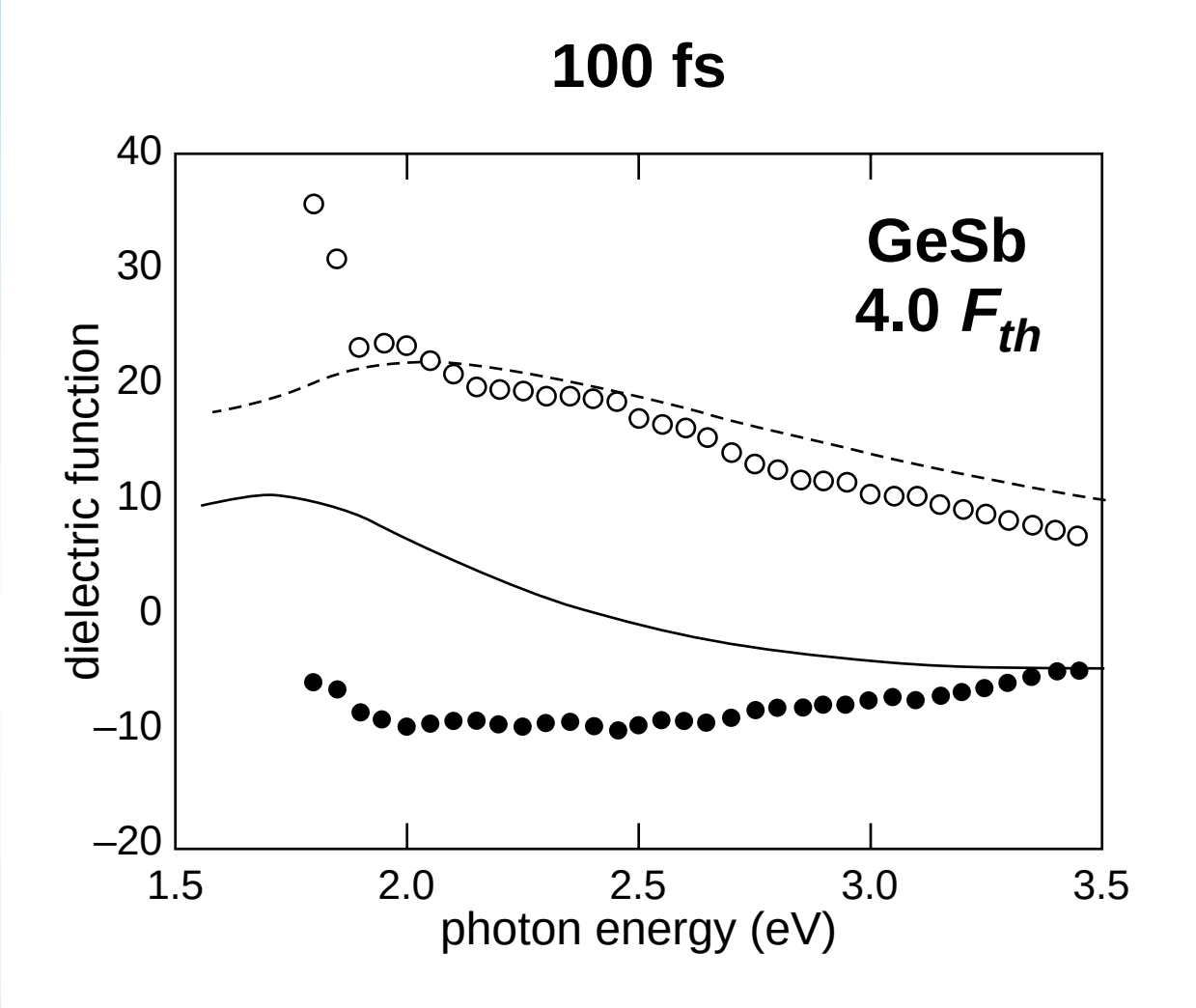
Results


–16 ps
GeSb
4.0 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5

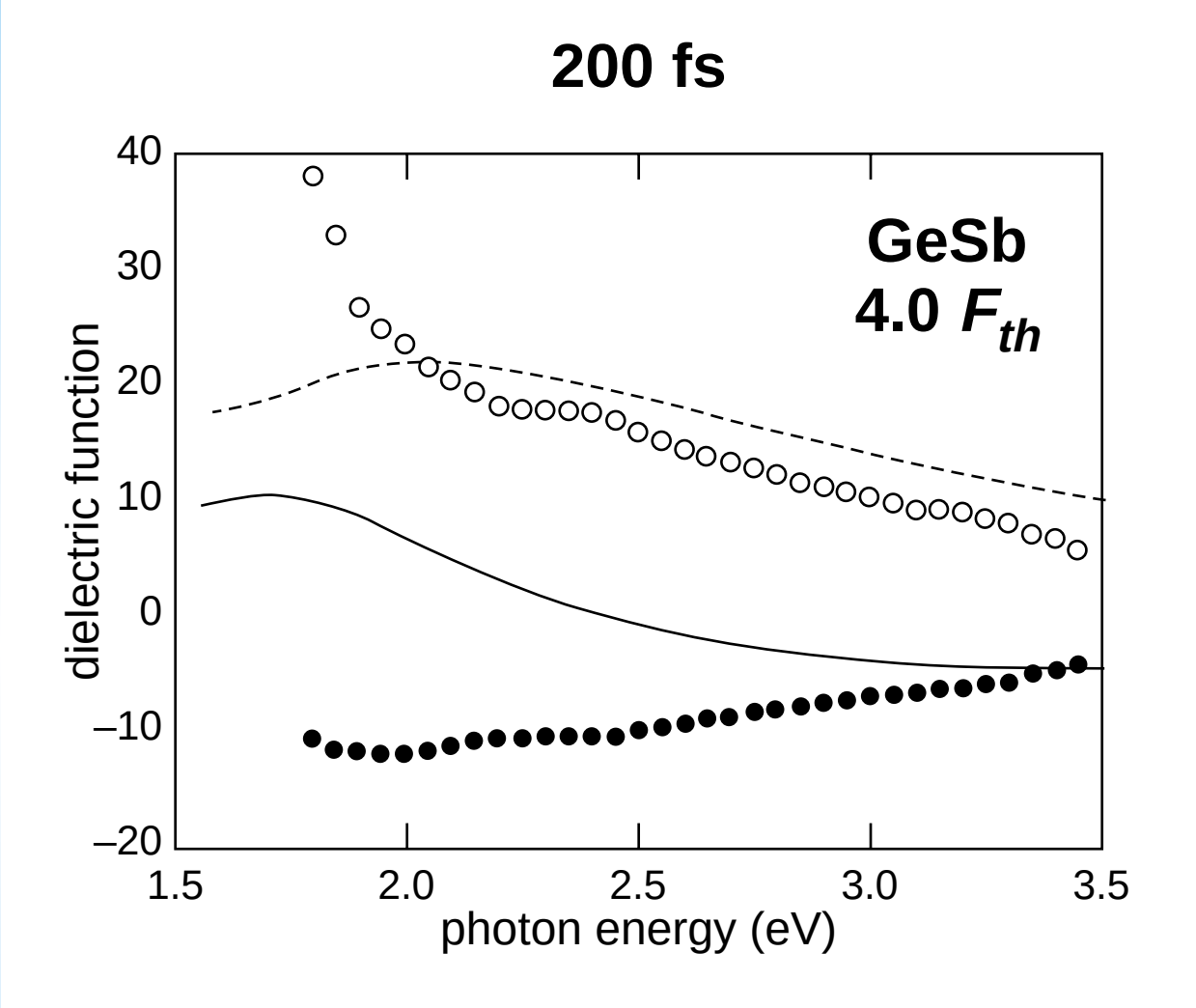
Results


0 ps
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

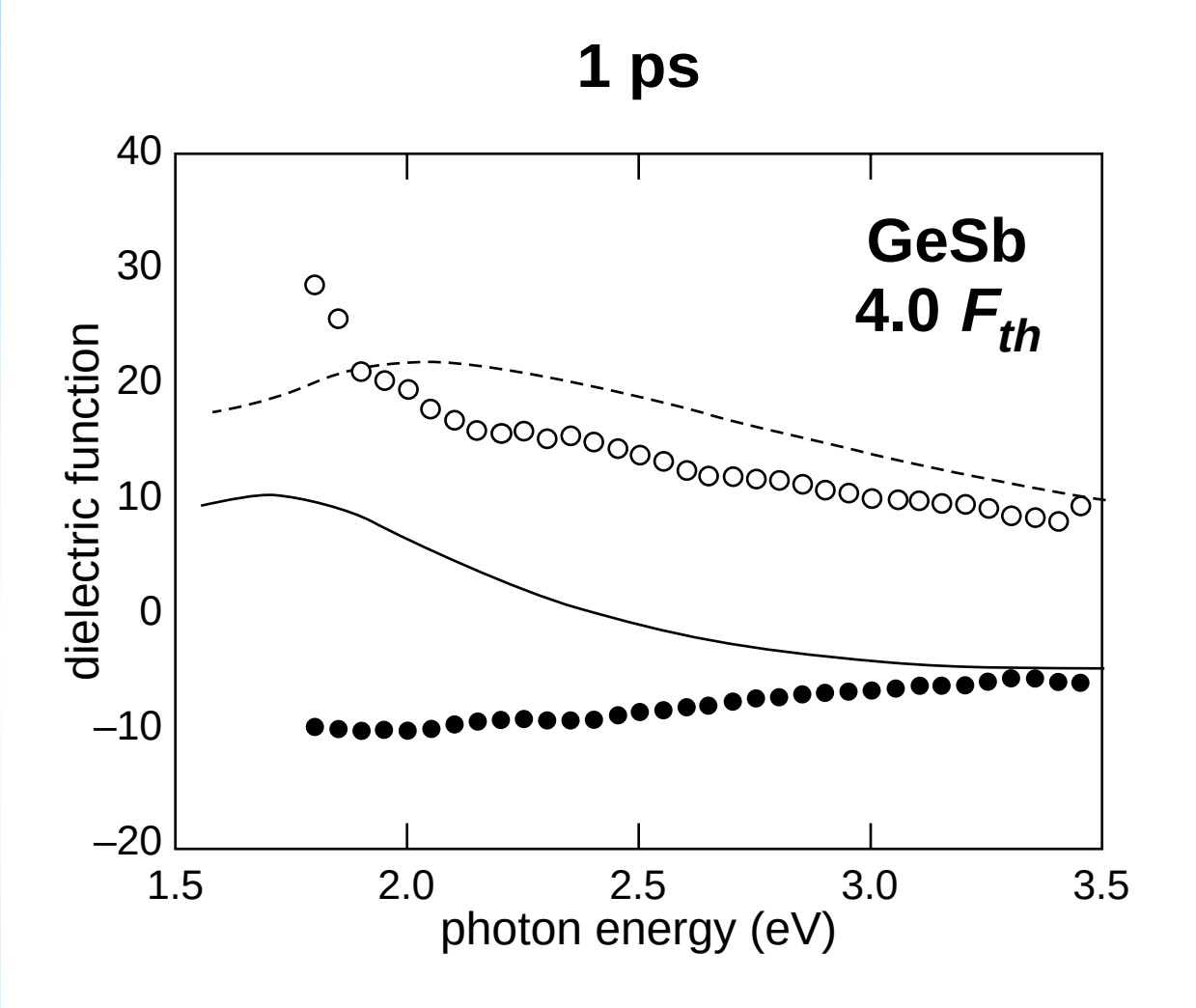
Results


100 fs
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

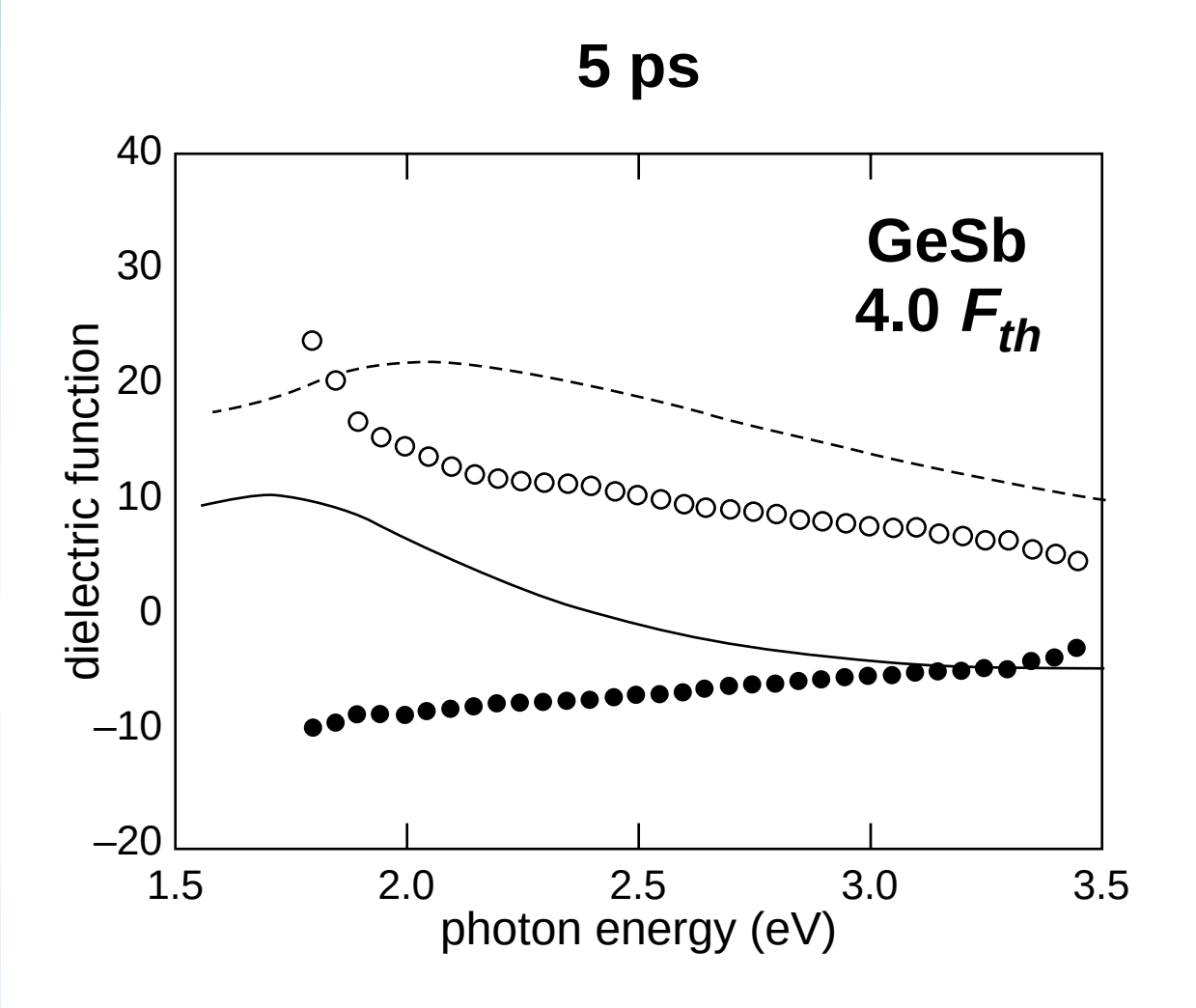
Results


200 fs
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

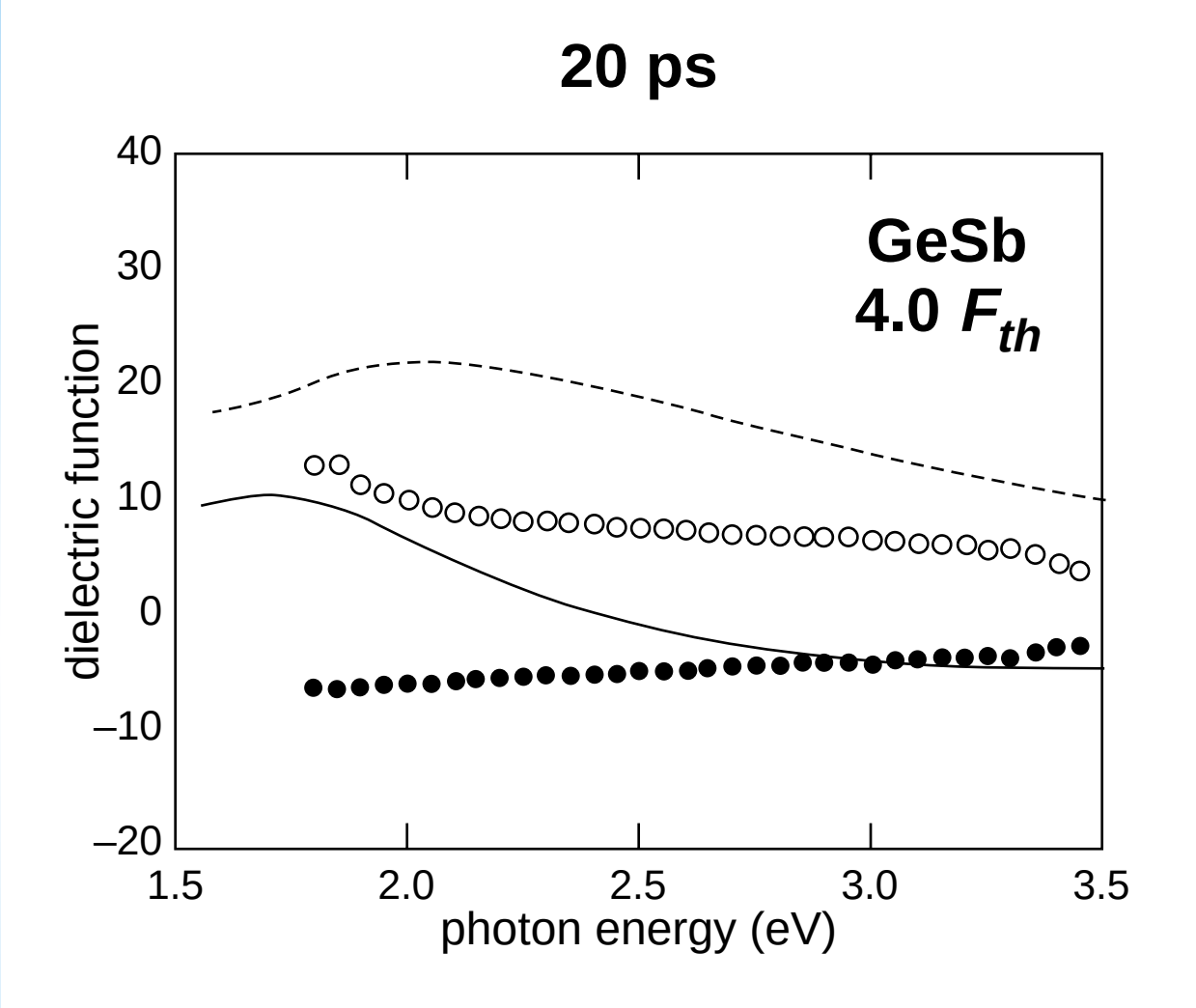
Results


1 ps
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

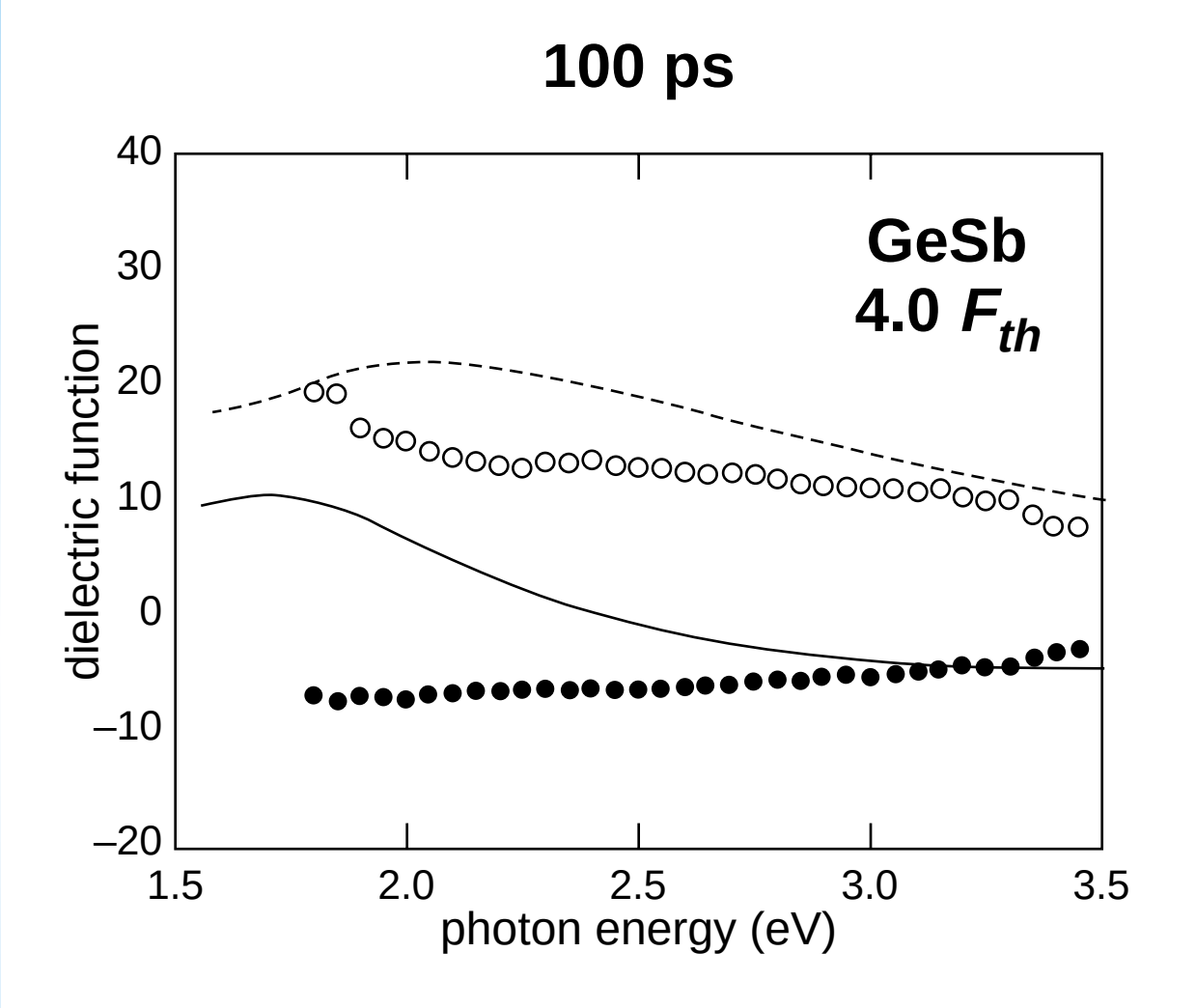
Results


5 ps
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

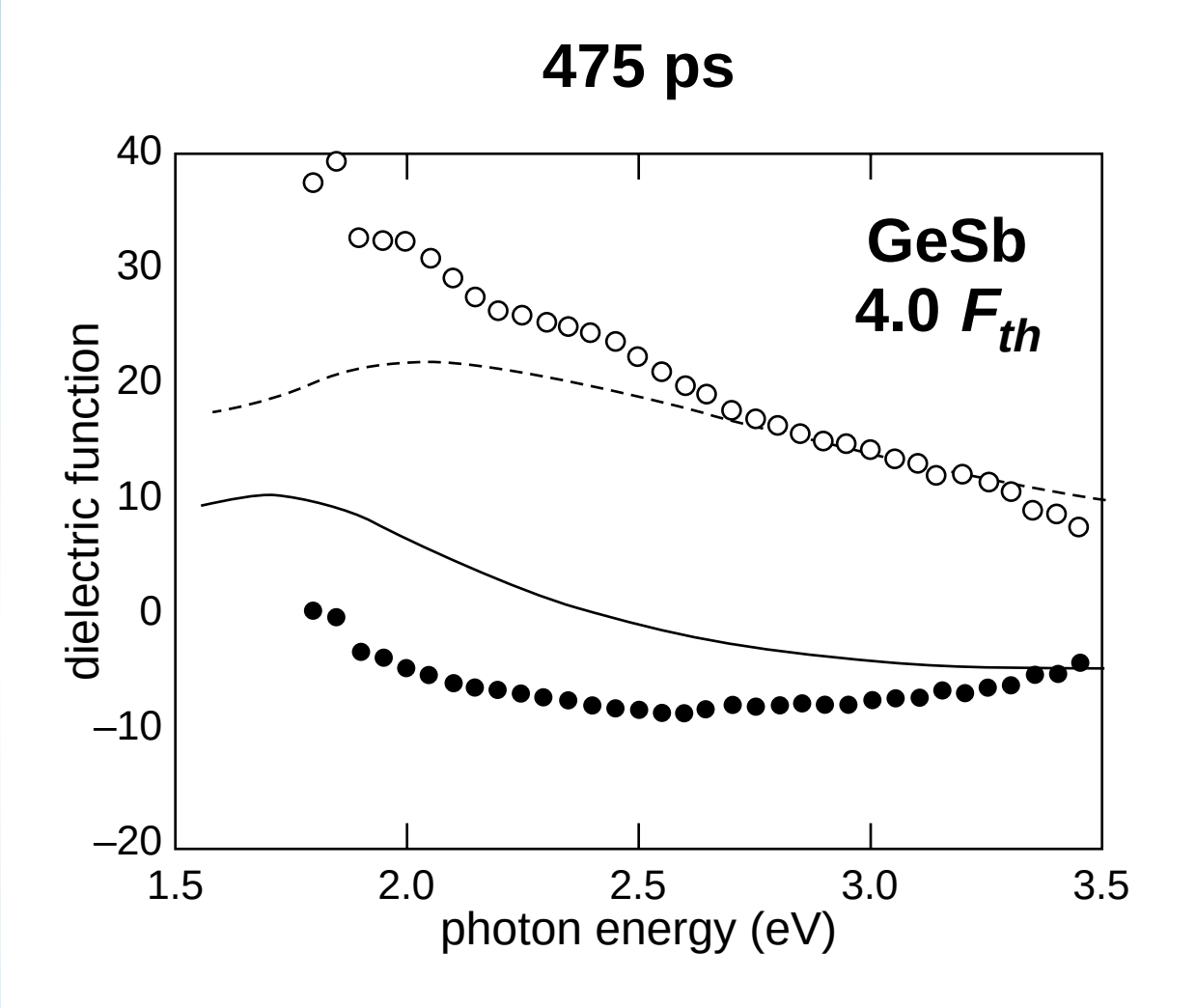
Results


20 ps
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

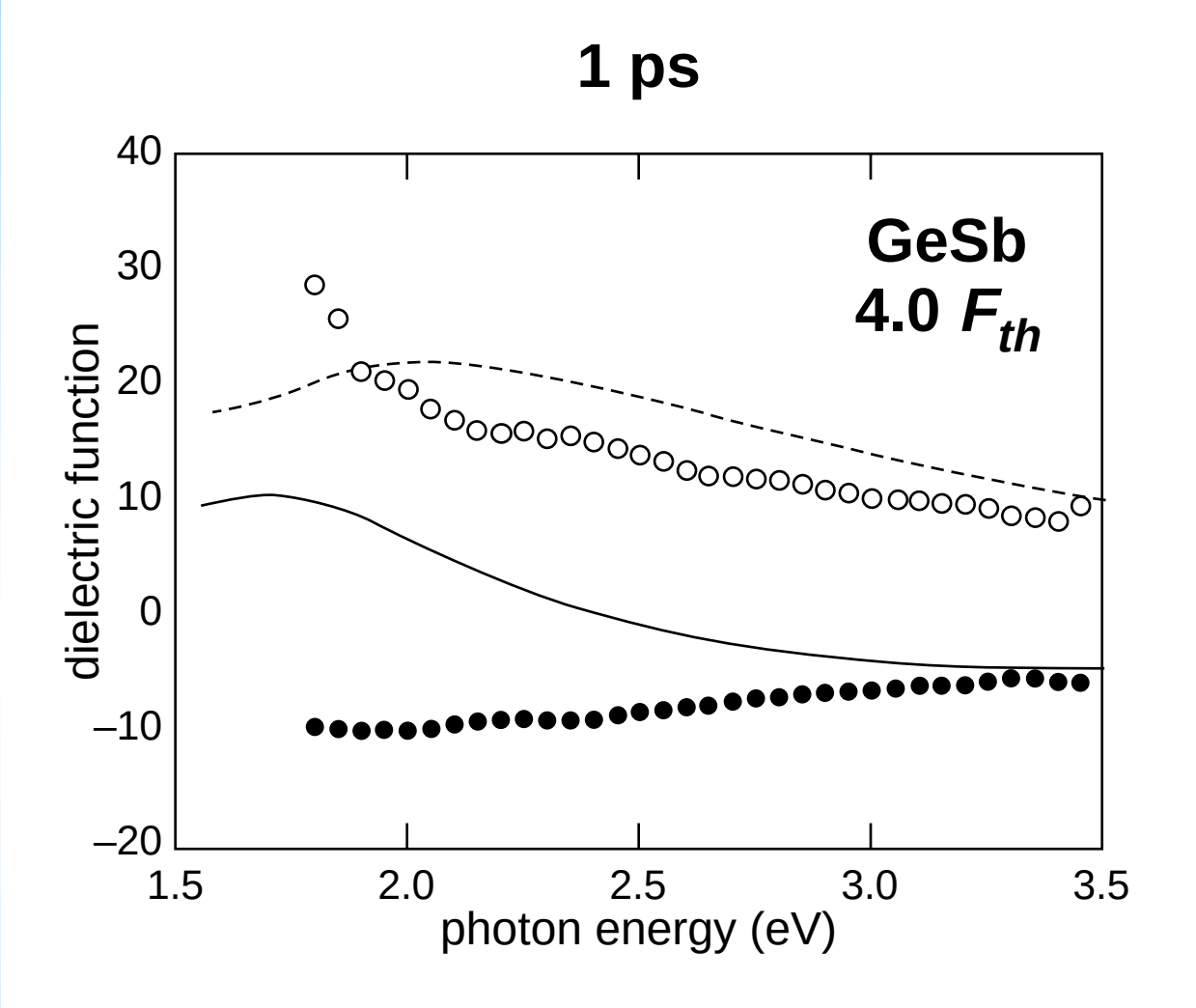
Results


100 ps
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

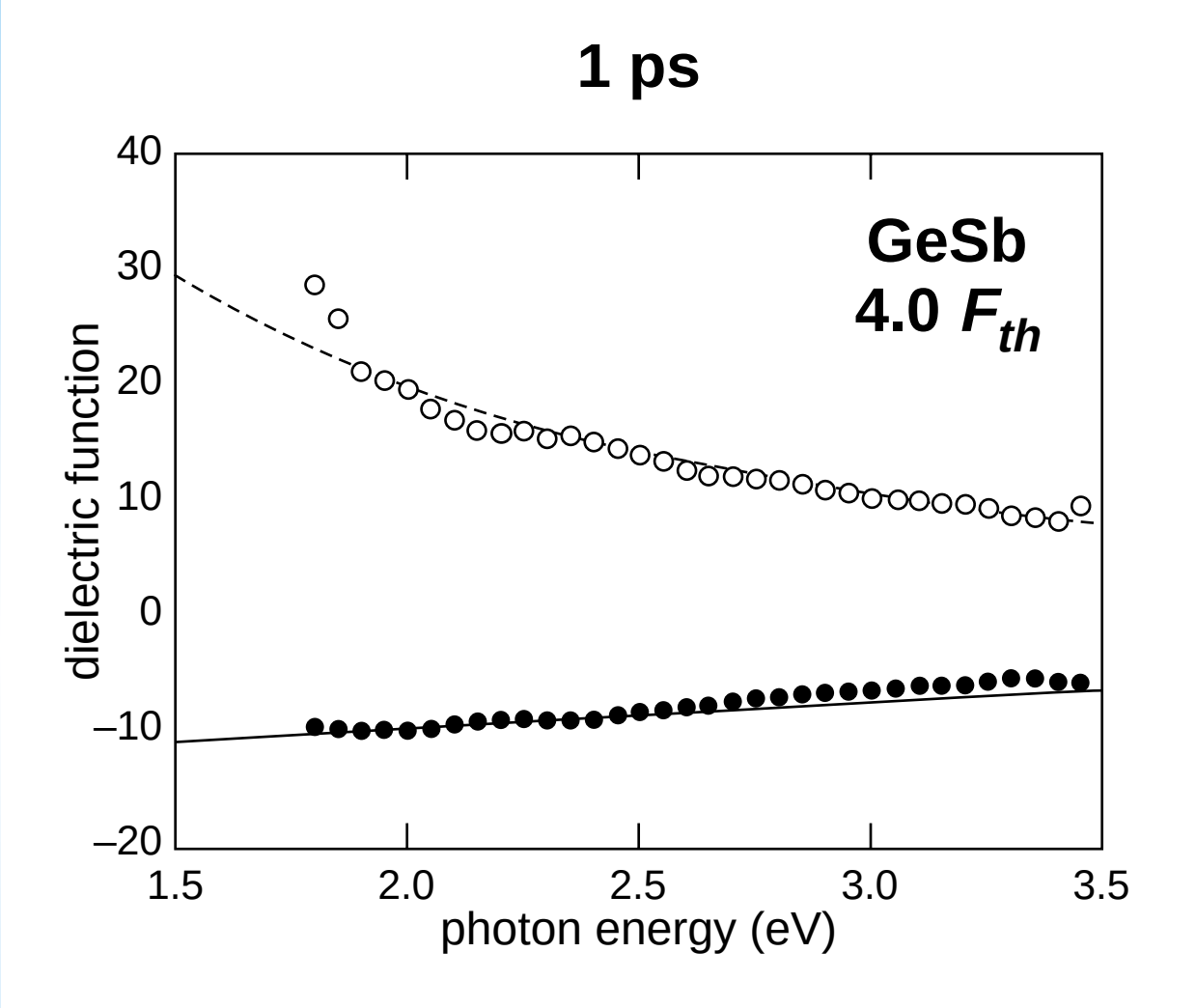
Results


475 ps
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

Results

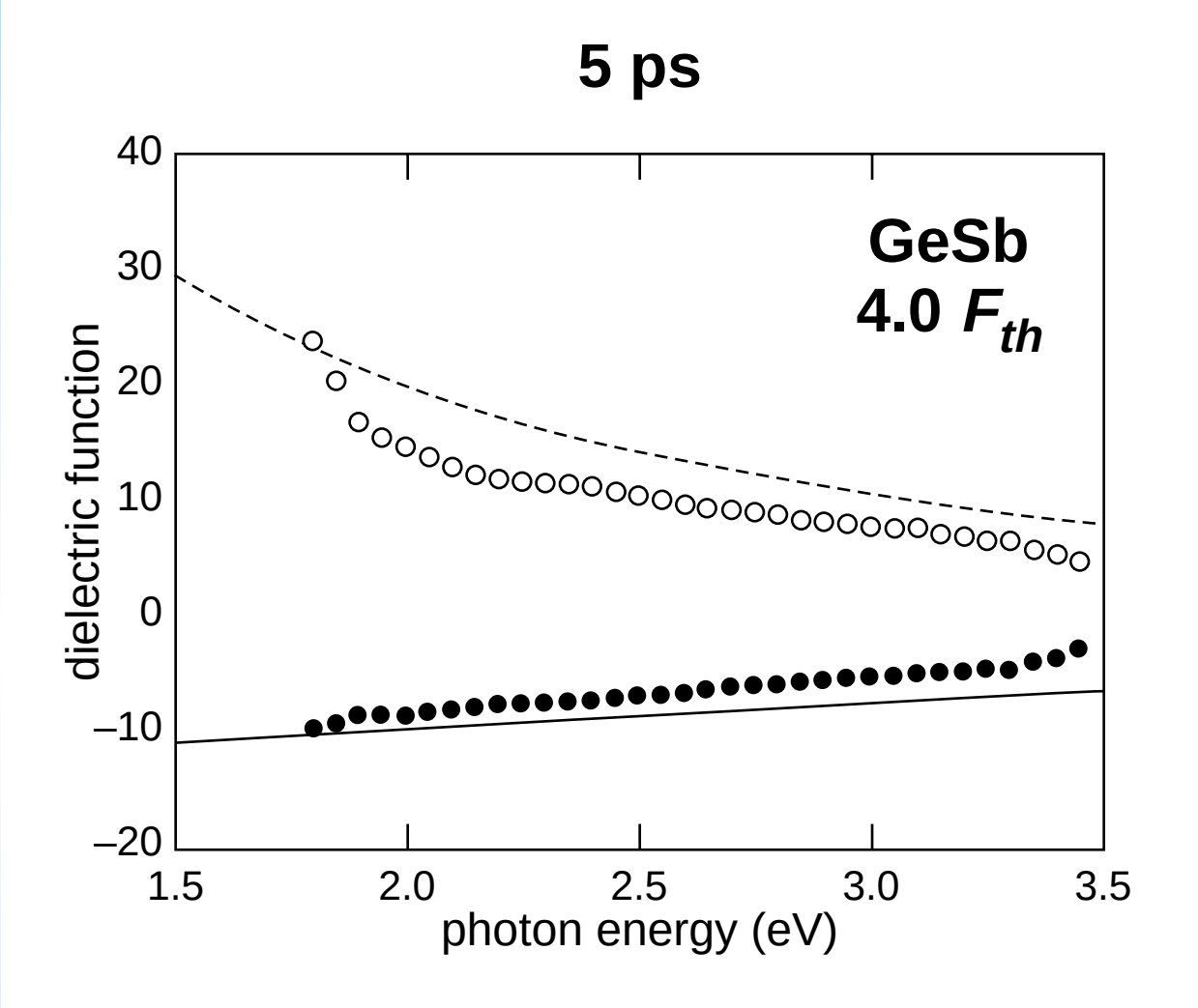

1 ps
GeSb
4.0 F_{th}
dielectric function
40
30
20
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

Results



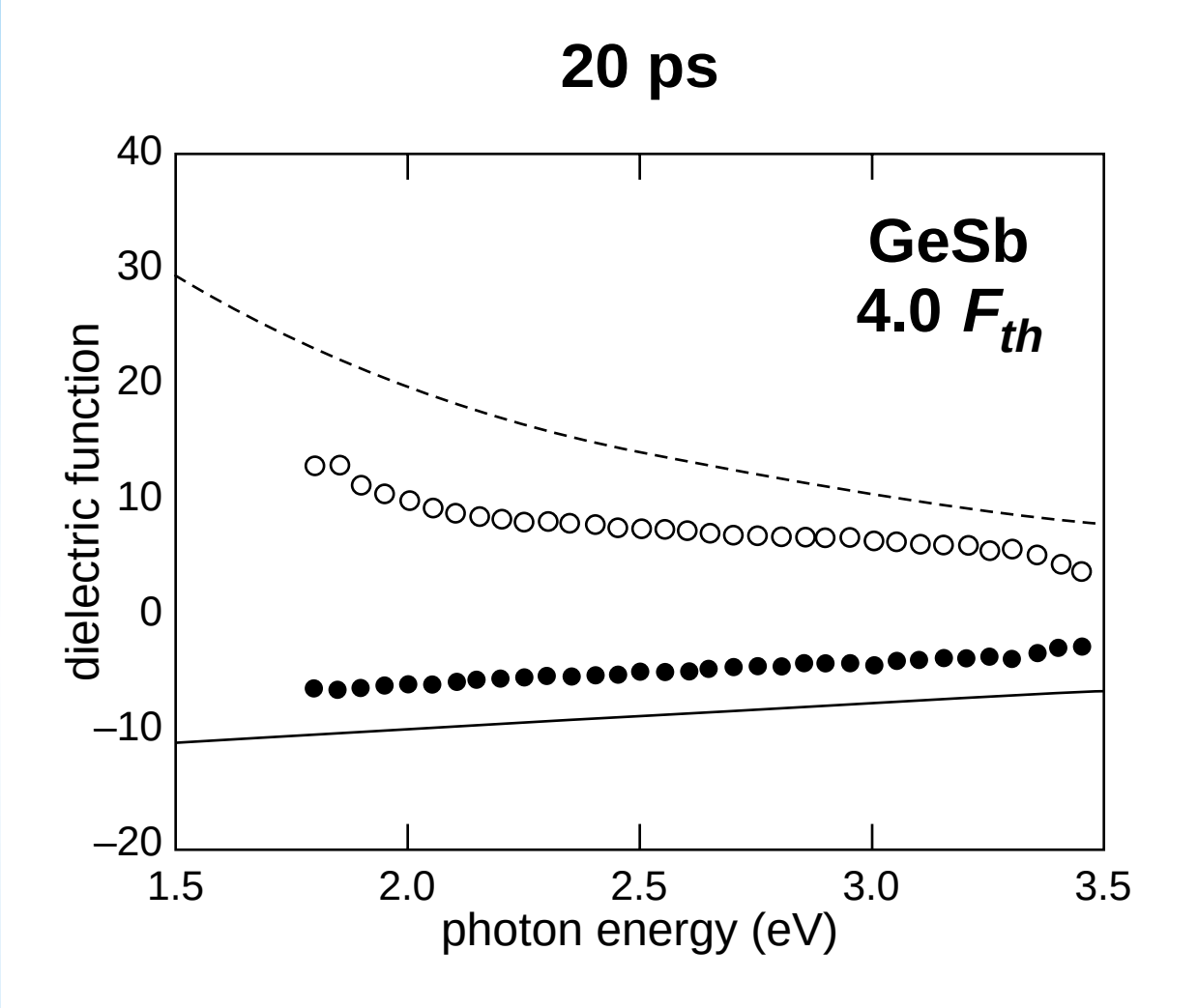
Drude-like after 1 ps

Results



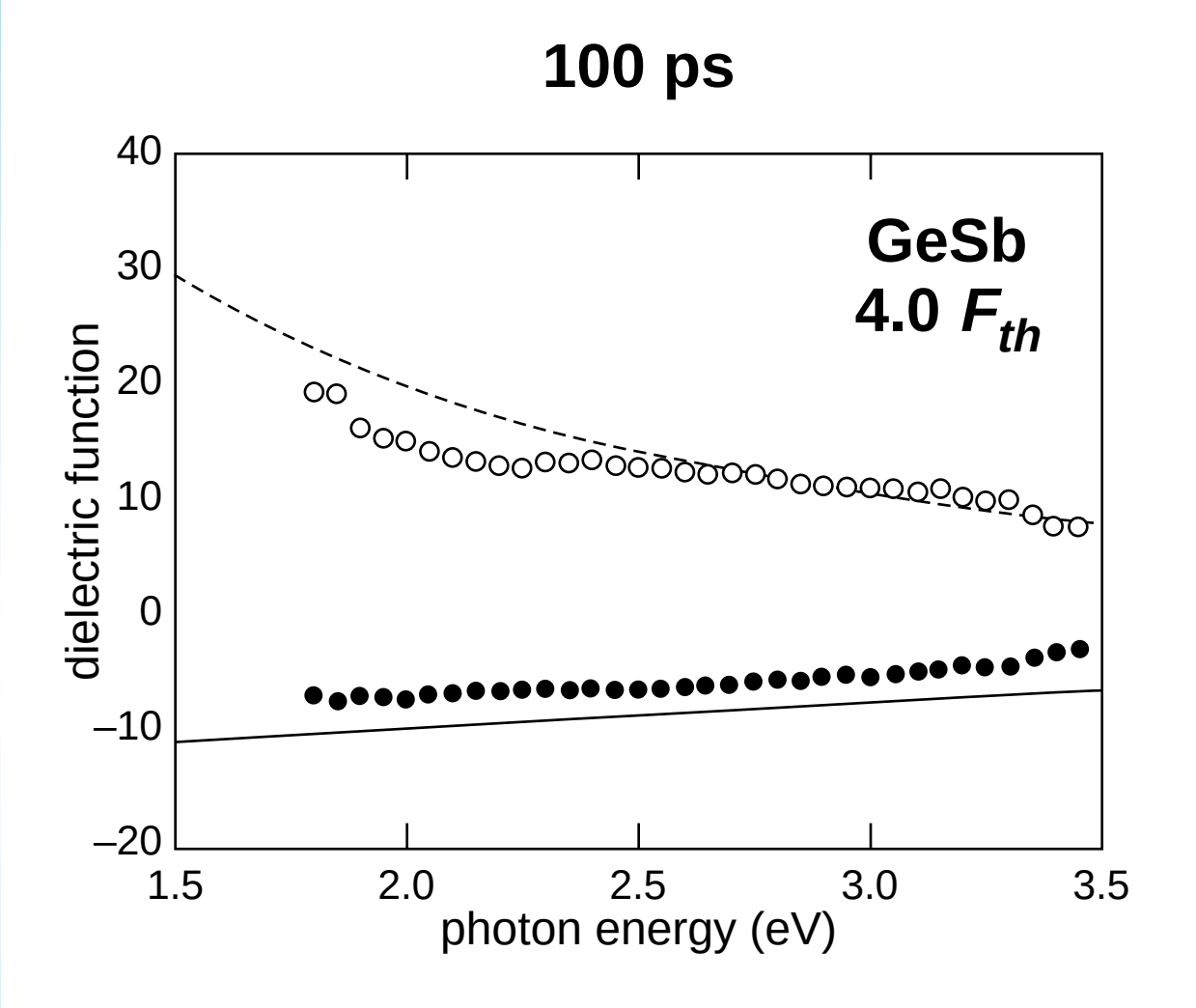
plasma frequency decreases

Results



plasma frequency decreases

Results


100 ps
GeSb
4.0 F_{th}
dielectric function
40
30
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10
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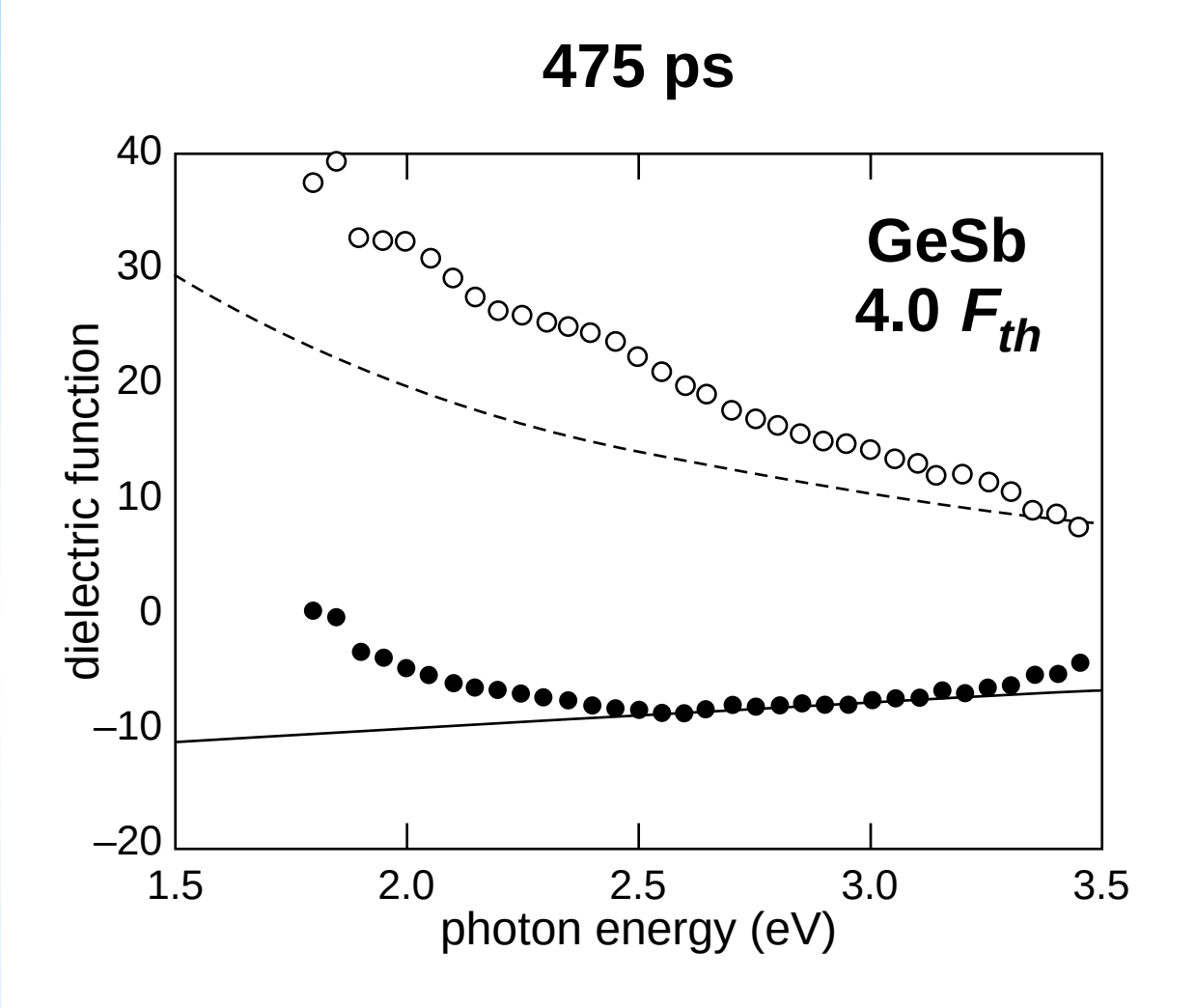
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

Results


475 ps
GeSb
4.0 F_{th}
dielectric function
40
30
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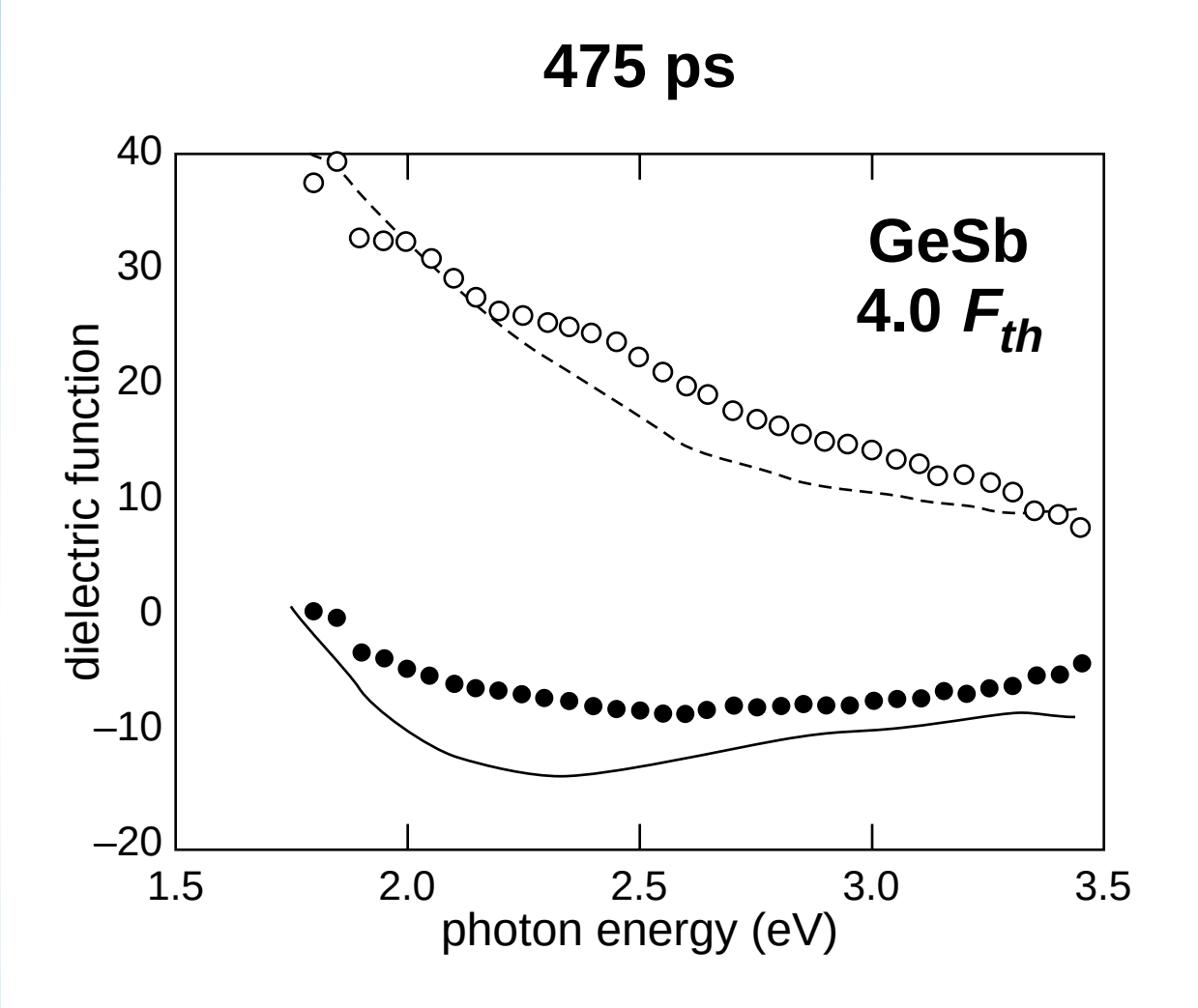
10
0
–10
–20
1.5
2.0
2.5
3.0
3.5
photon energy (eV)

Results



laser-induced recrystallization

Summary

Universal features:

Summary

Universal features:

- **semiconductor-to-metal transition**

Summary

Universal features:

- **semiconductor-to-metal transition**
- **decrease in bonding-antibonding splitting takes picoseconds**

Summary

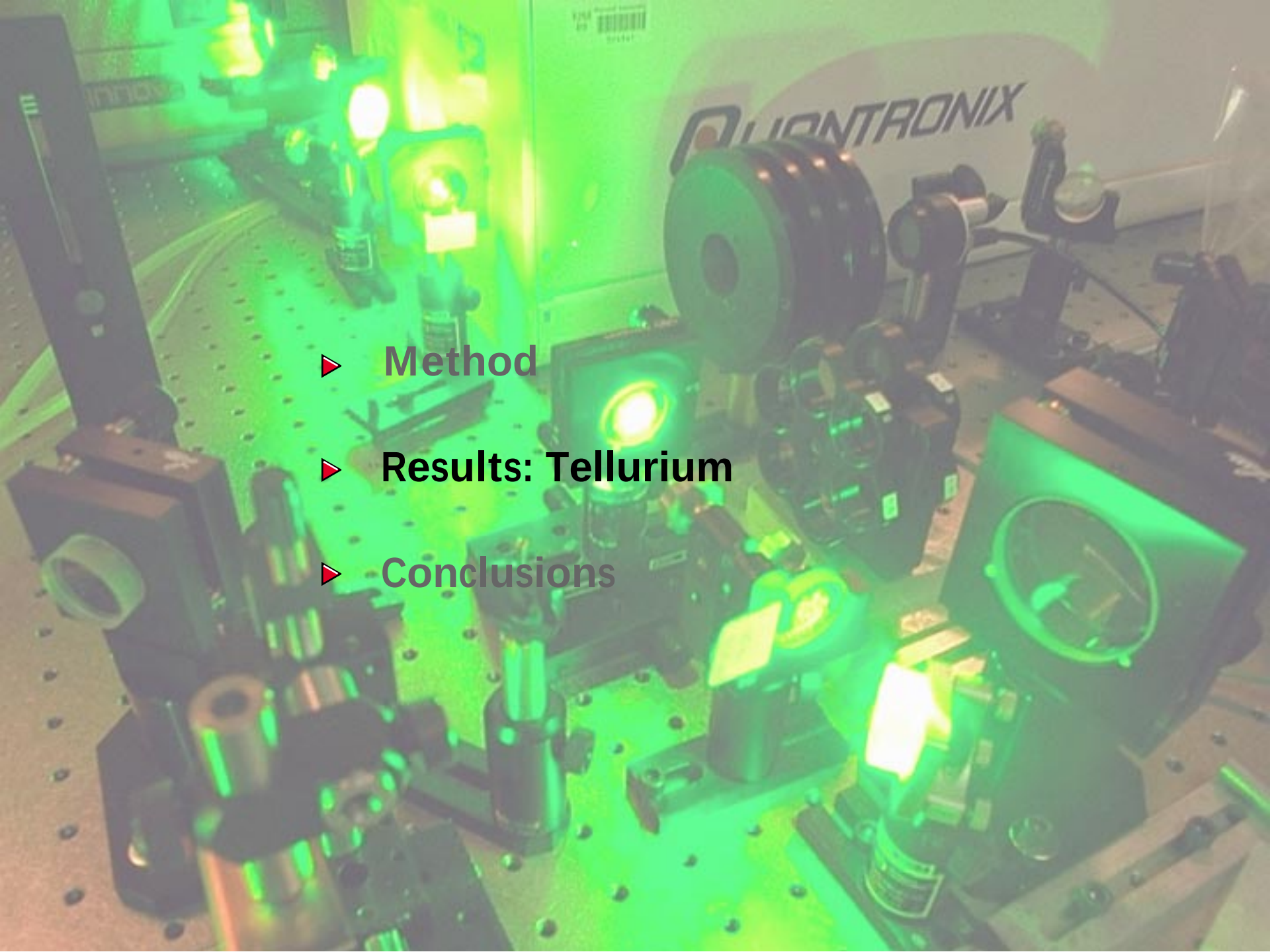
Universal features:

- **semiconductor-to-metal transition**
- **decrease in bonding-antibonding splitting takes picoseconds**
- **plasma frequency decreases with time**

Summary

Universal features:

- **semiconductor-to-metal transition**
- **decrease in bonding-antibonding splitting takes picoseconds**
- **plasma frequency decreases with time**
- **plasma frequency decreases with increasing fluence**



- Method
- **Results: Tellurium**
- Conclusions

Why Tellurium?

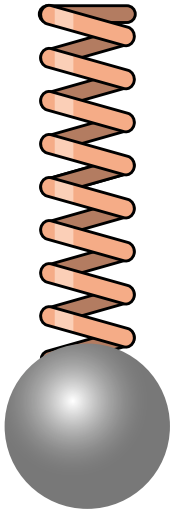
- **semimetal (0.3 eV bandgap)**
- **large ΔR due to A_1 phonons**
- **A_1 phonons at 3.6 THz**

Why Tellurium?

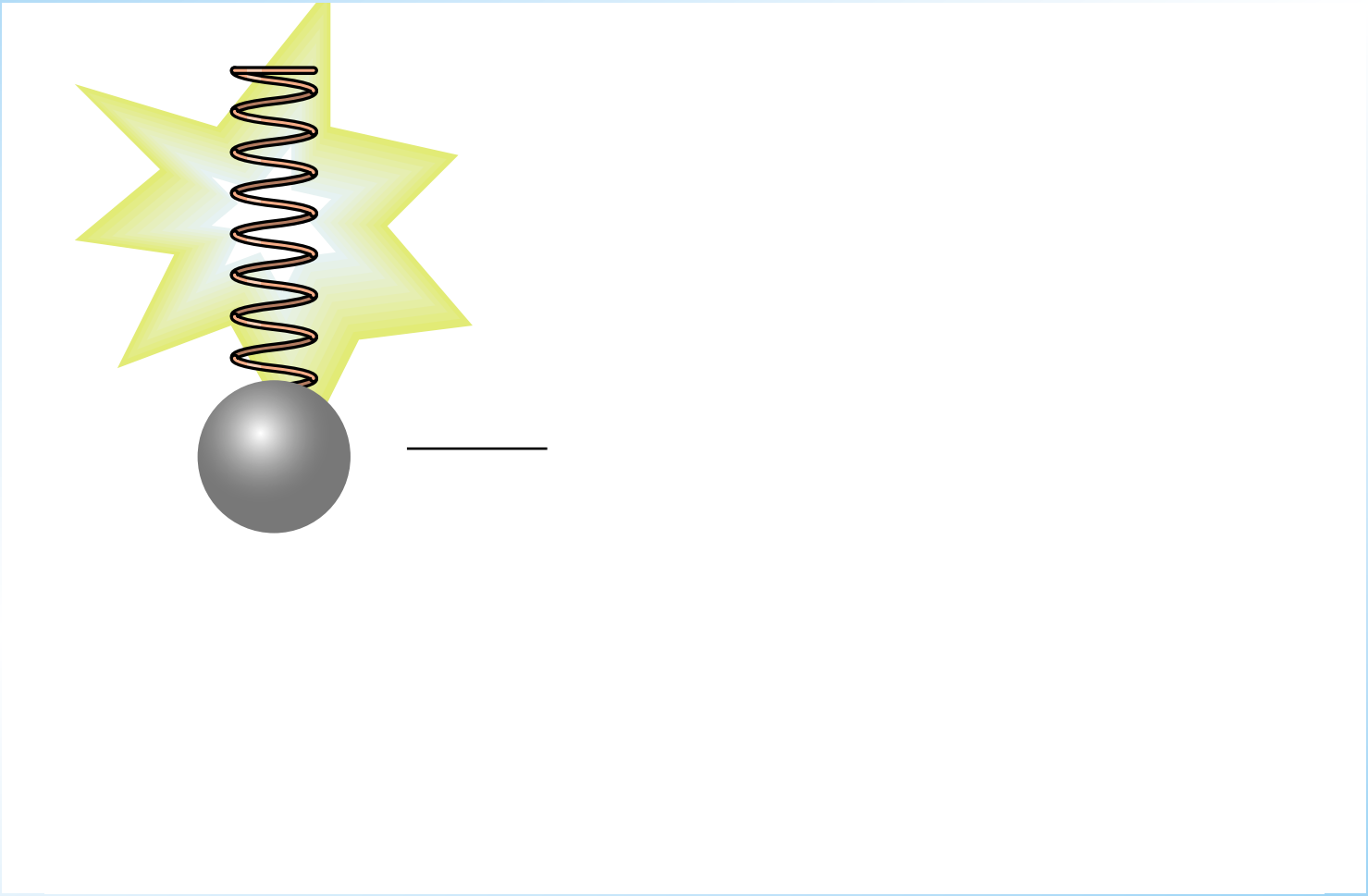
- **semimetal (0.3 eV bandgap)**
- **large ΔR due to A_1 phonons**
- **A_1 phonons at 3.6 THz**

… but A_1-mode is not IR-active

Displacive excitation

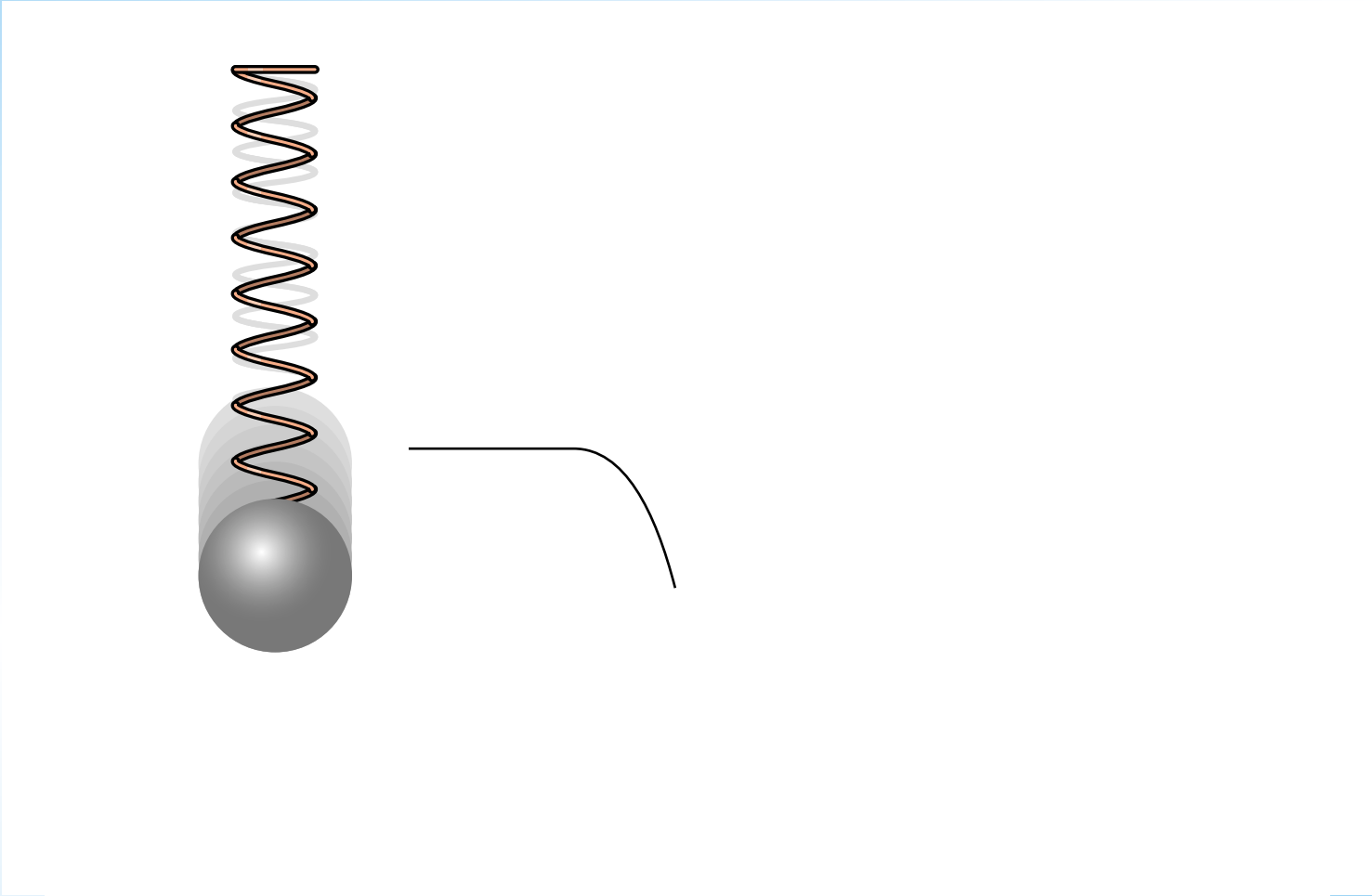


Displacive excitation



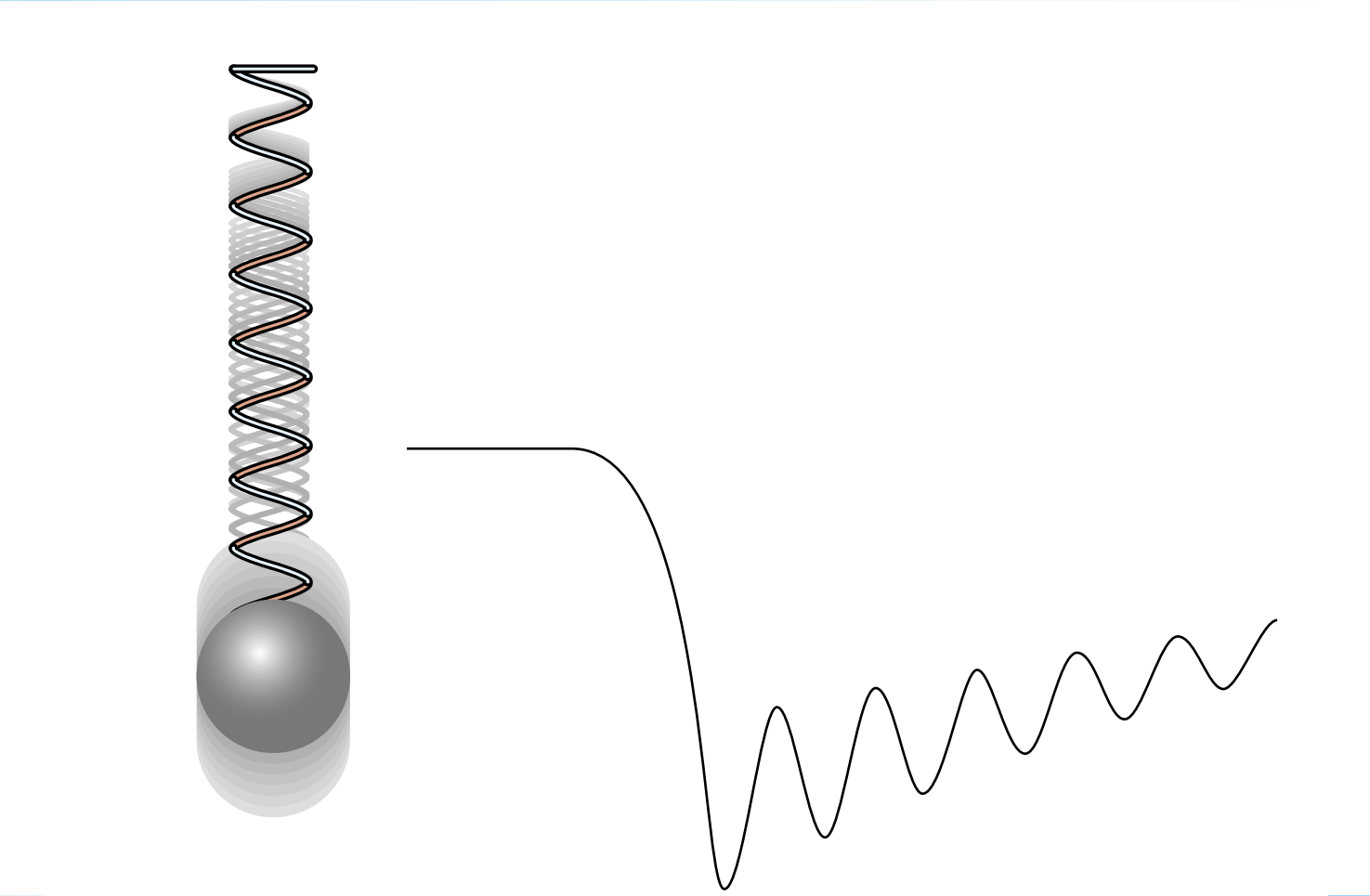
excitation weakens binding force

Displacive excitation



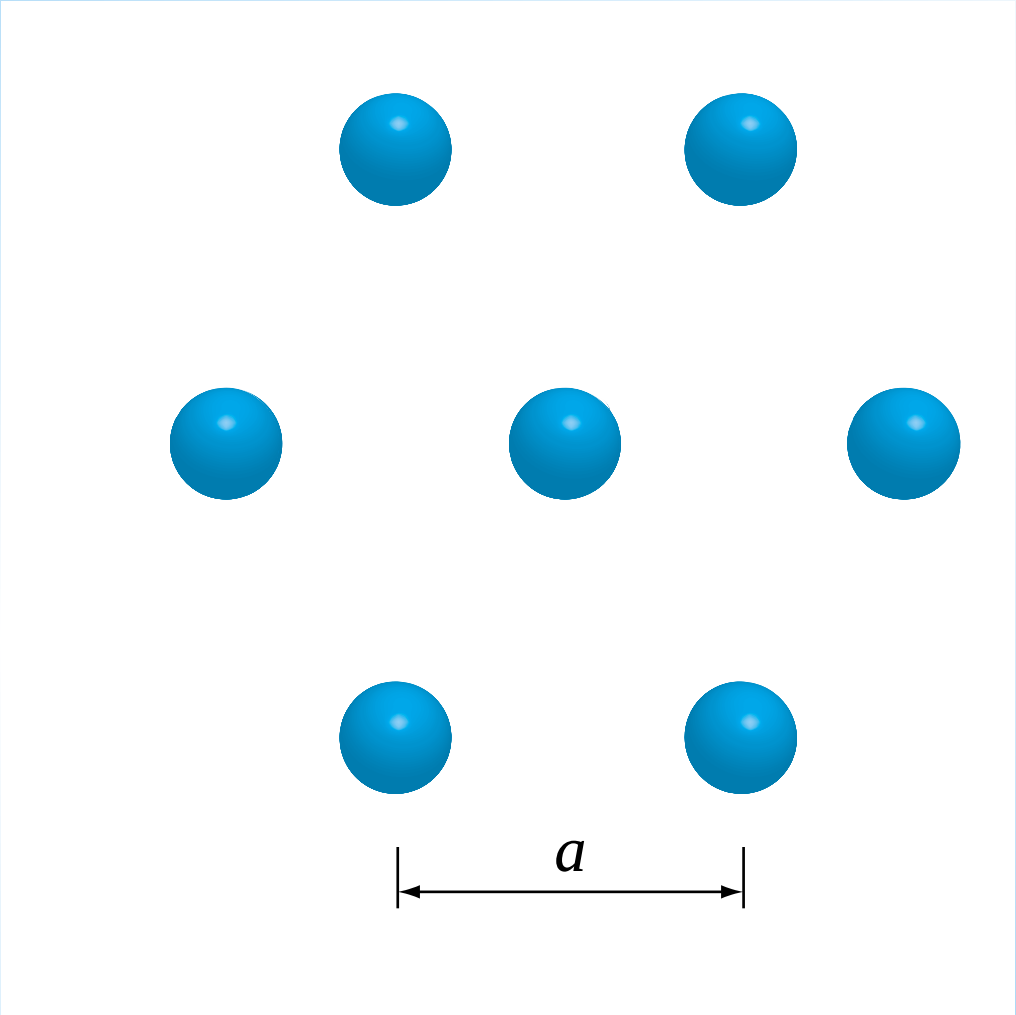
ion moves to new equilibrium position

Displacive excitation

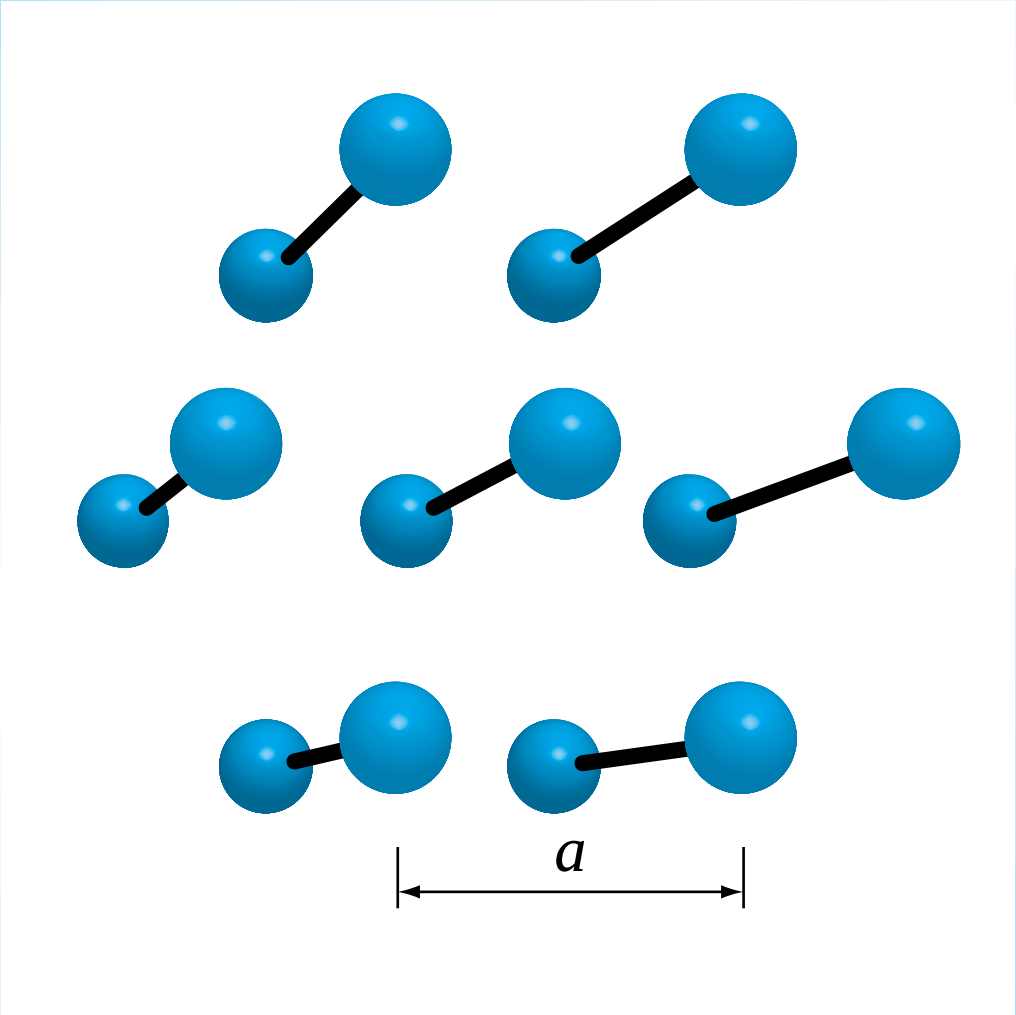


oscillation damps out as force is restored

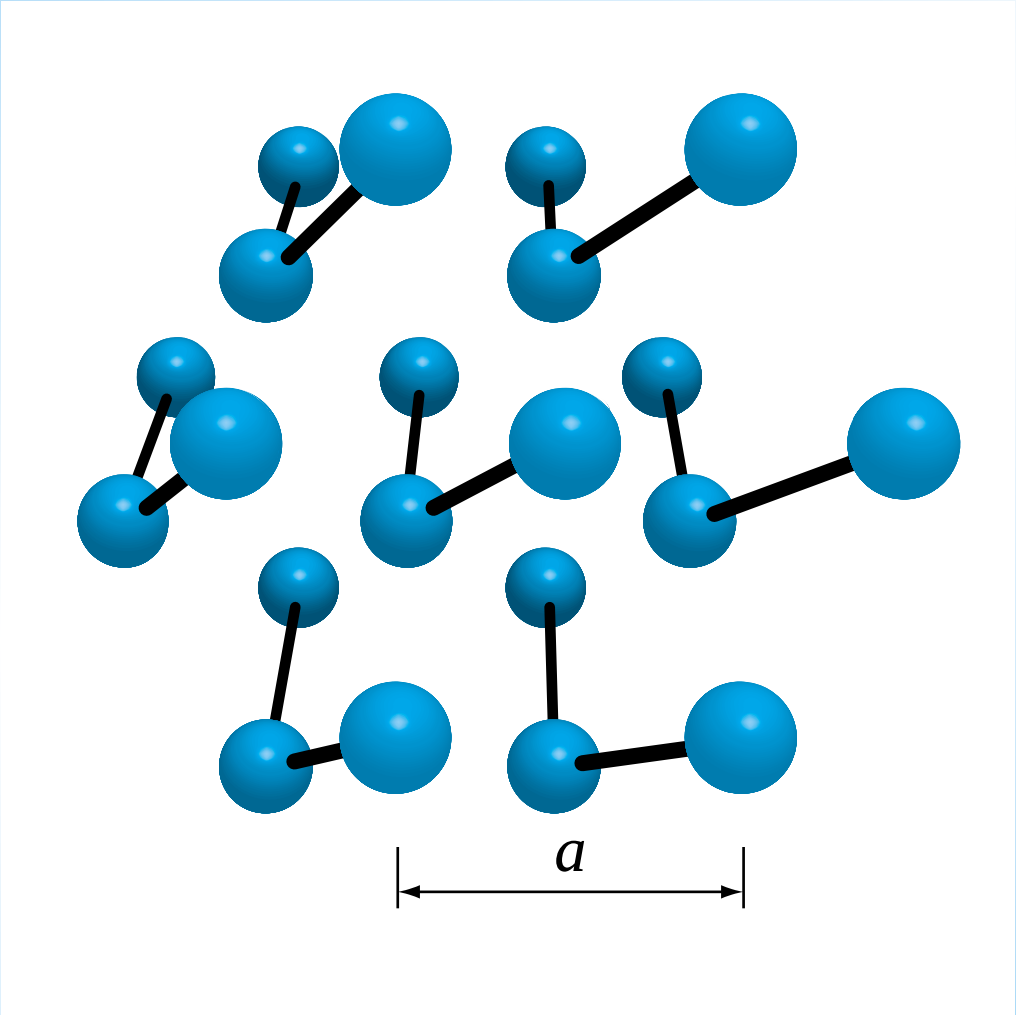
Tellurium structure



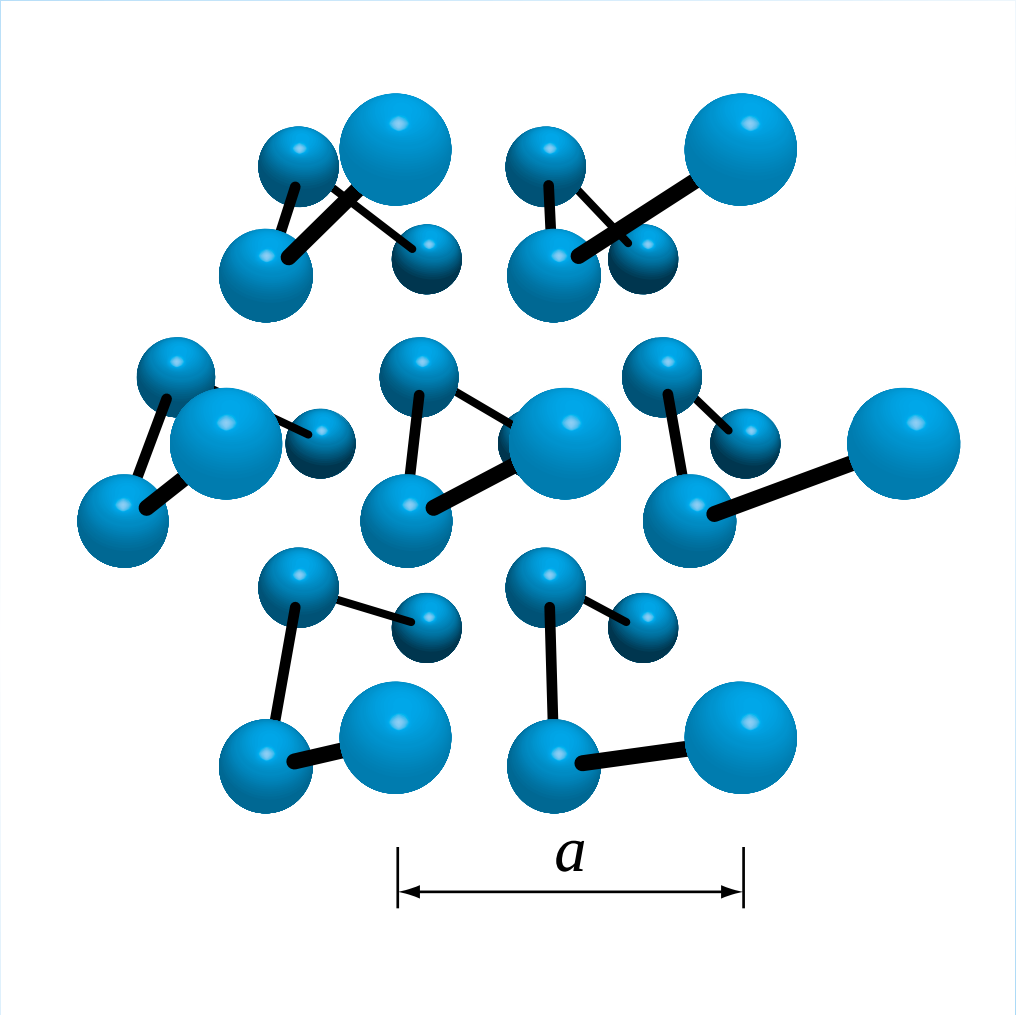
Tellurium structure



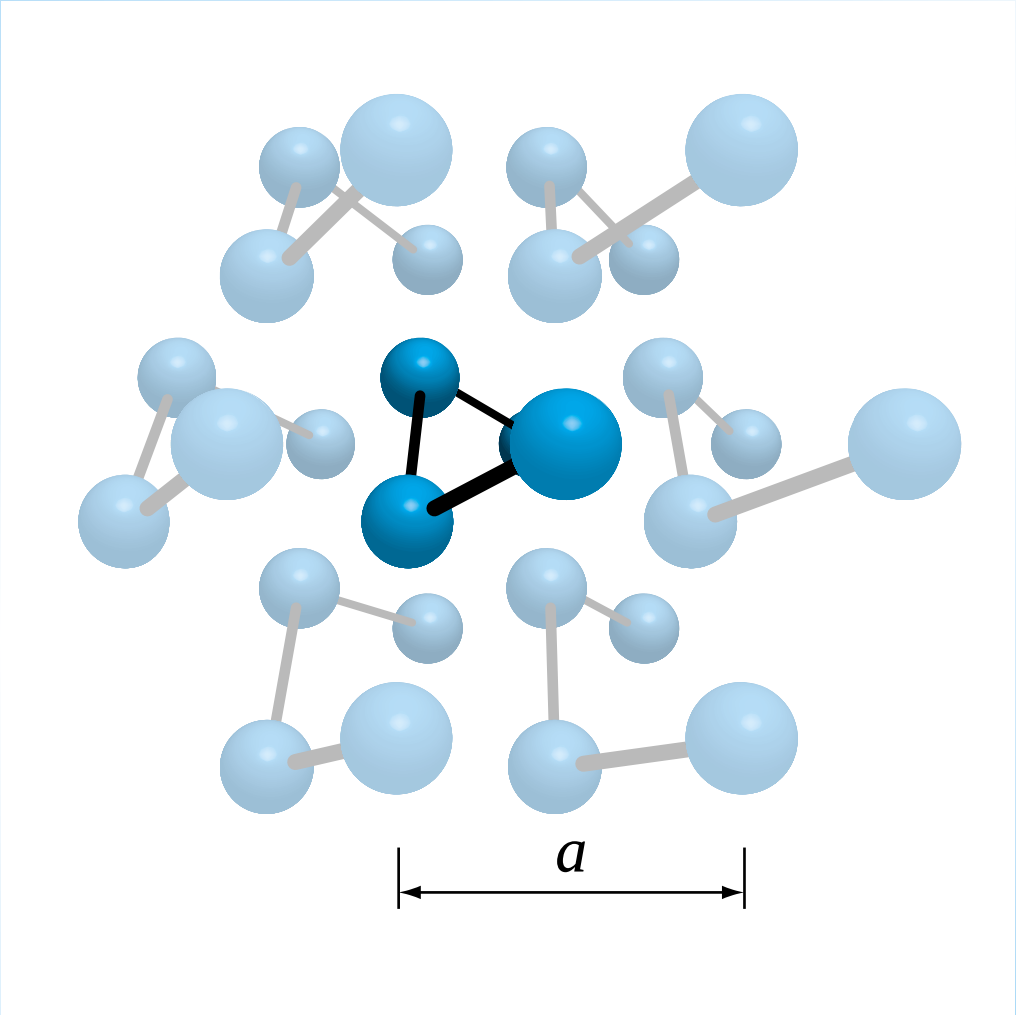
Tellurium structure



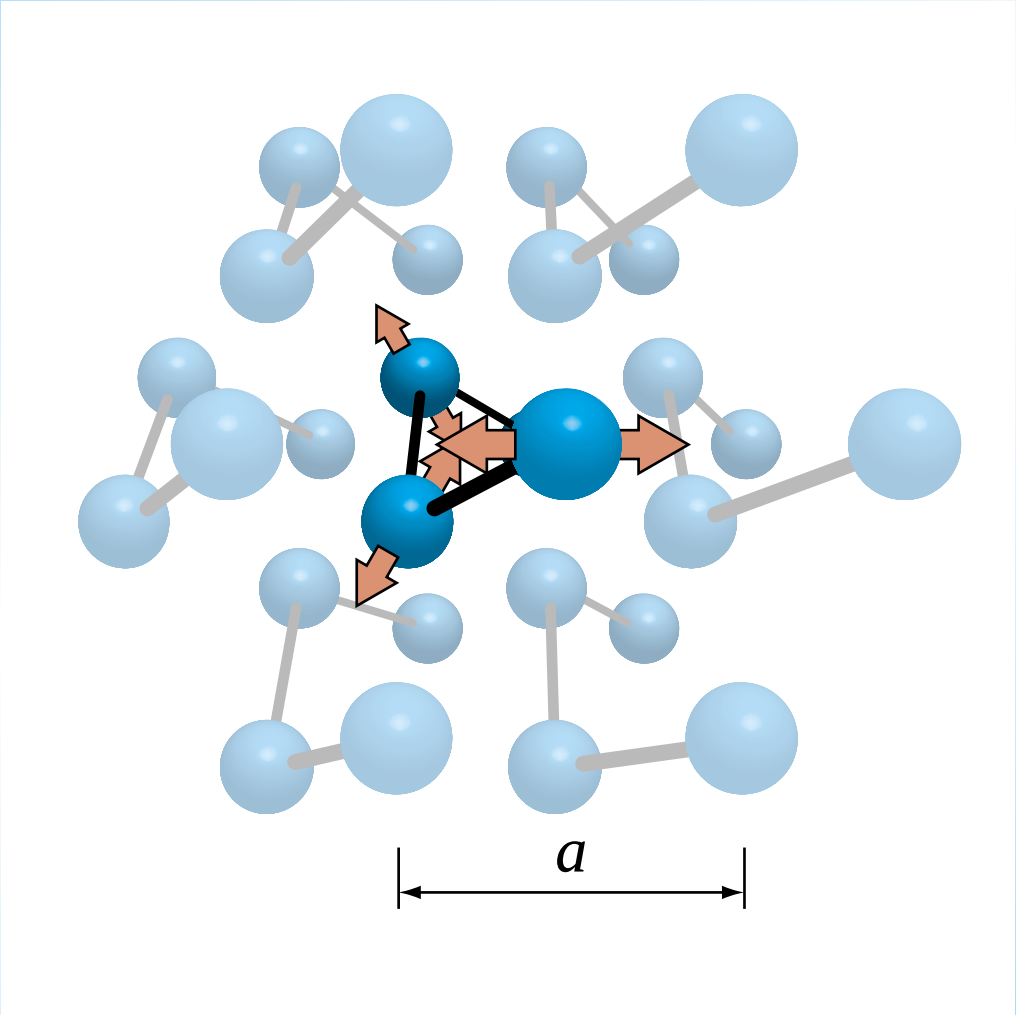
Tellurium structure



Tellurium structure

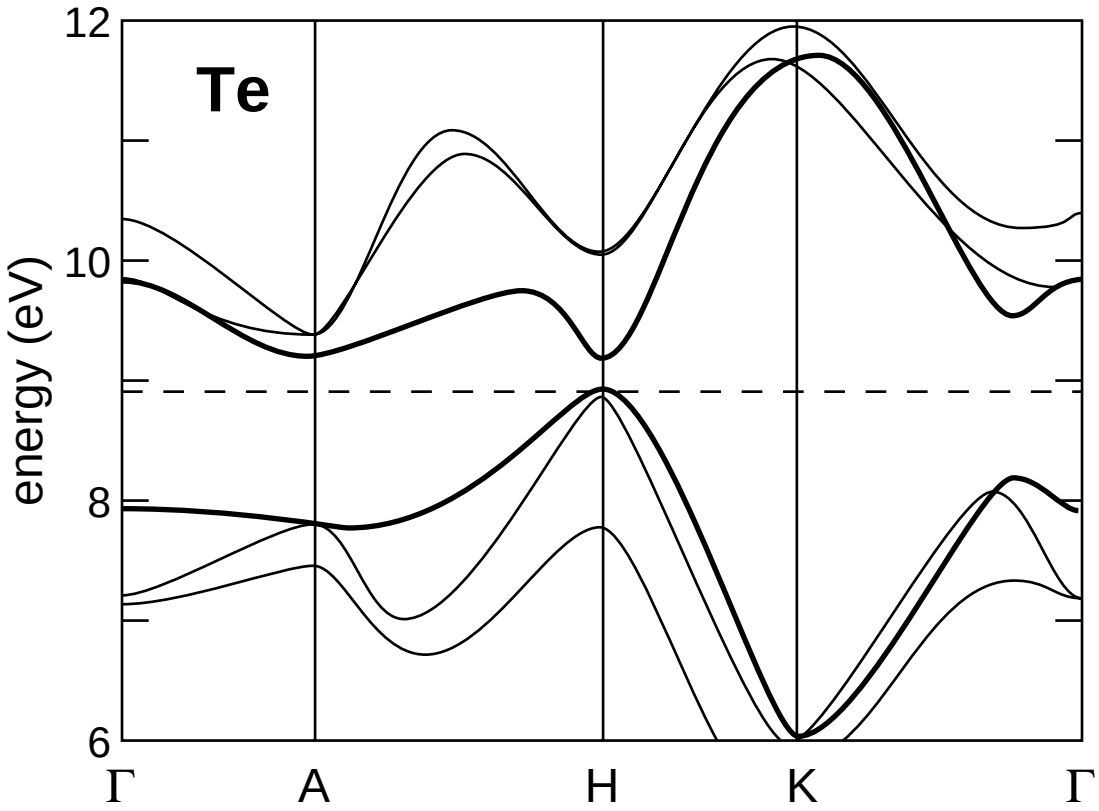


Tellurium structure



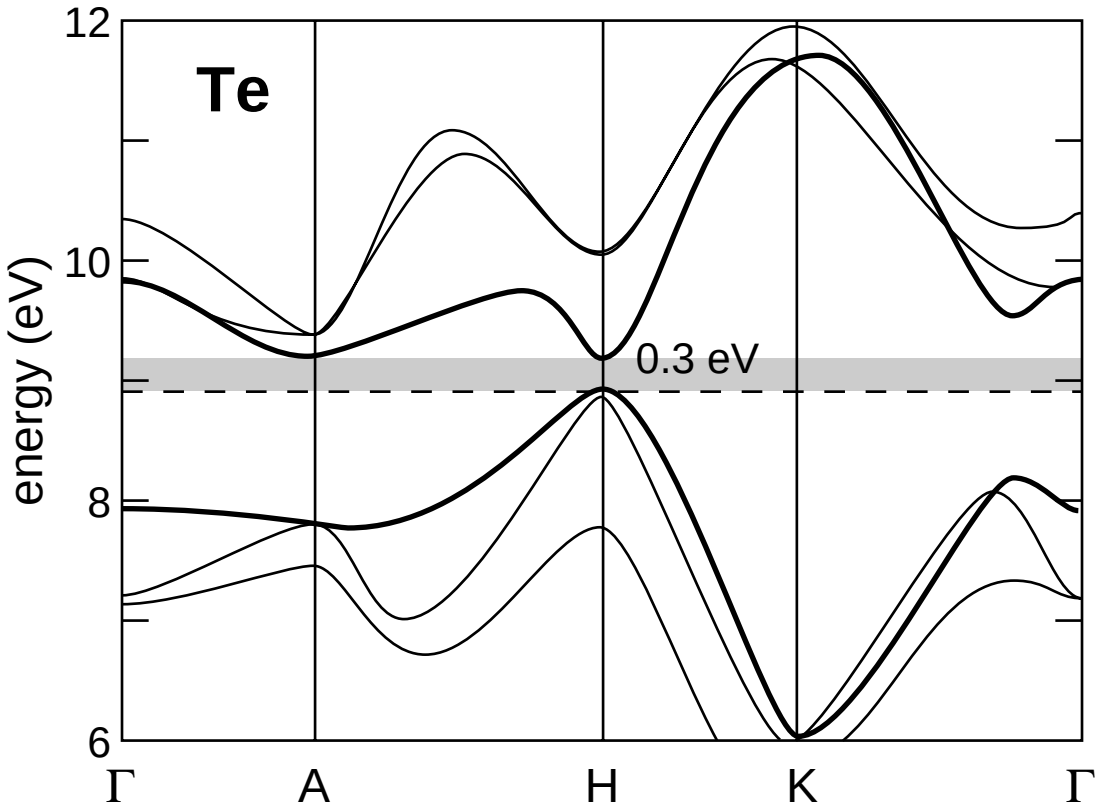
A_1 mode

Tellurium bandstructure



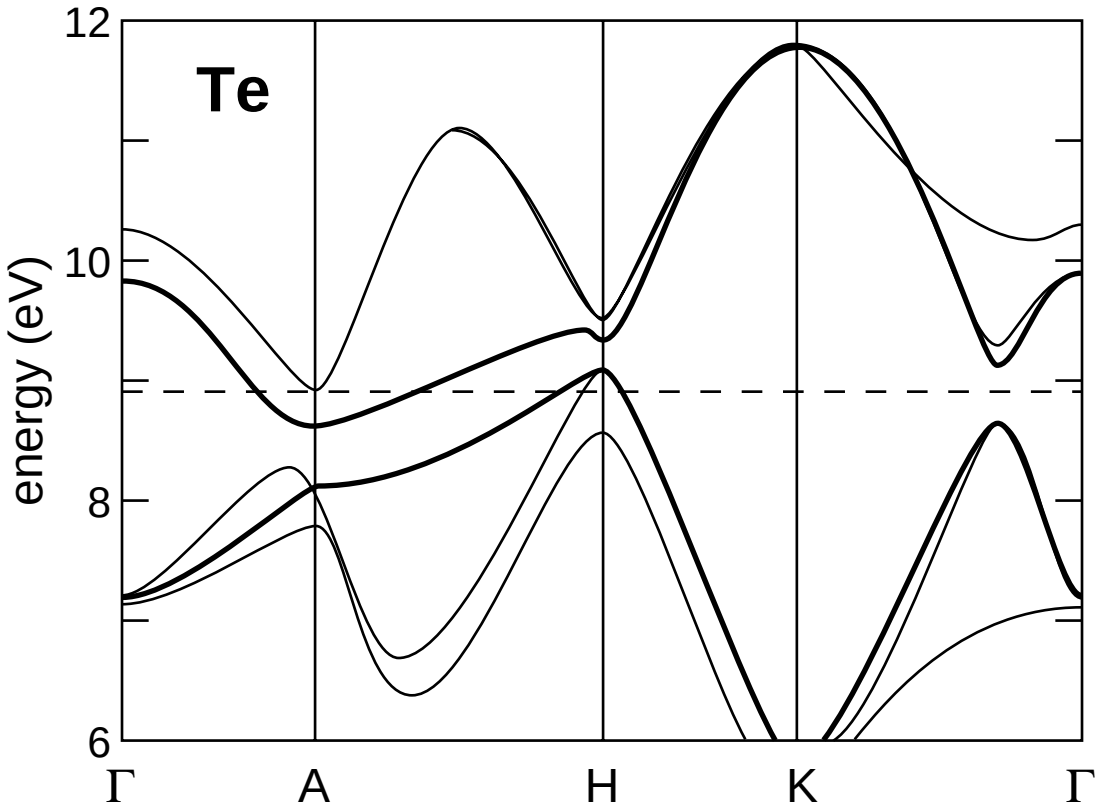
P. Tangney (Princeton) and S. Fahey (Cork), *private communication*

Tellurium bandstructure



P. Tangney (Princeton) and S. Fahey (Cork), *private communication*

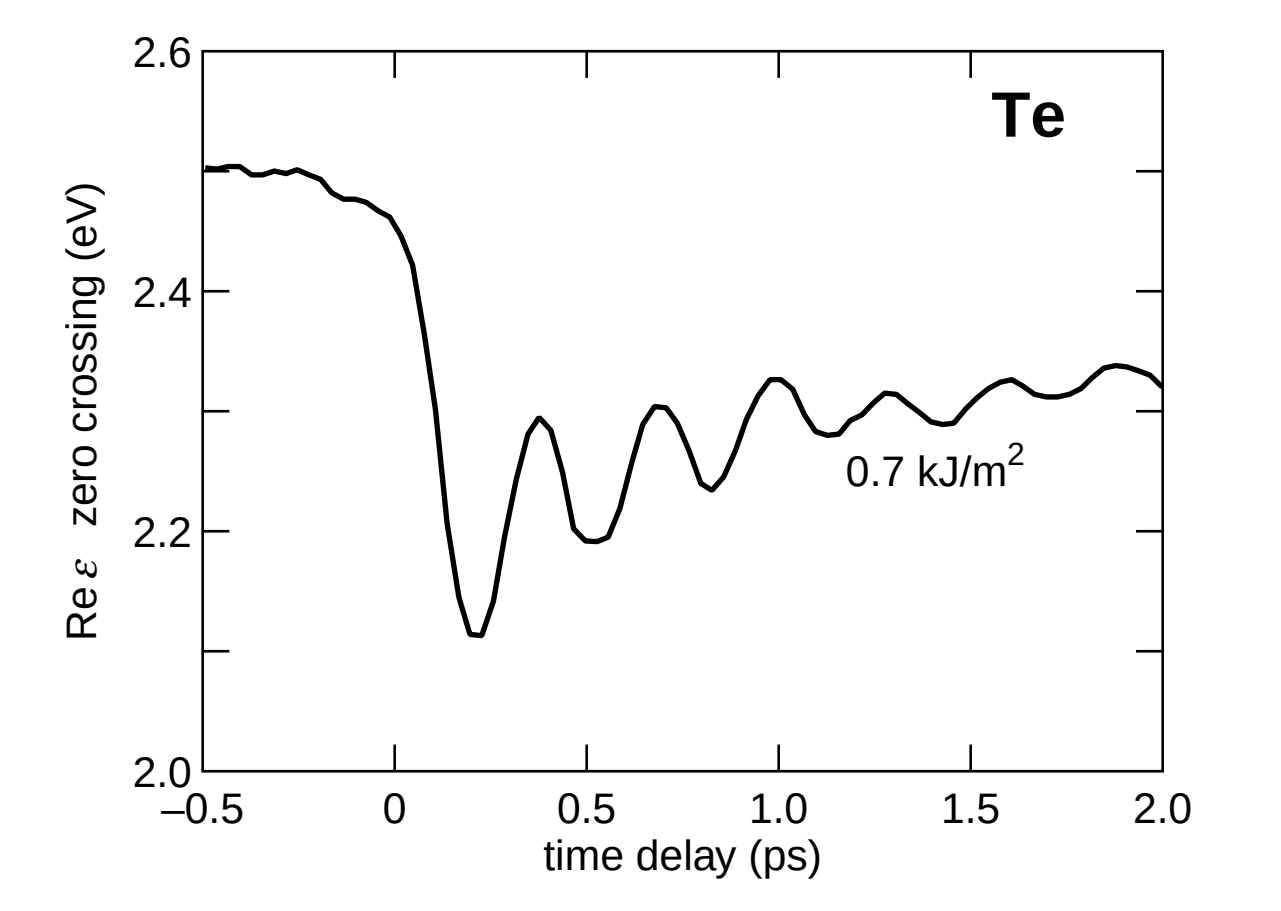
Tellurium bandstructure



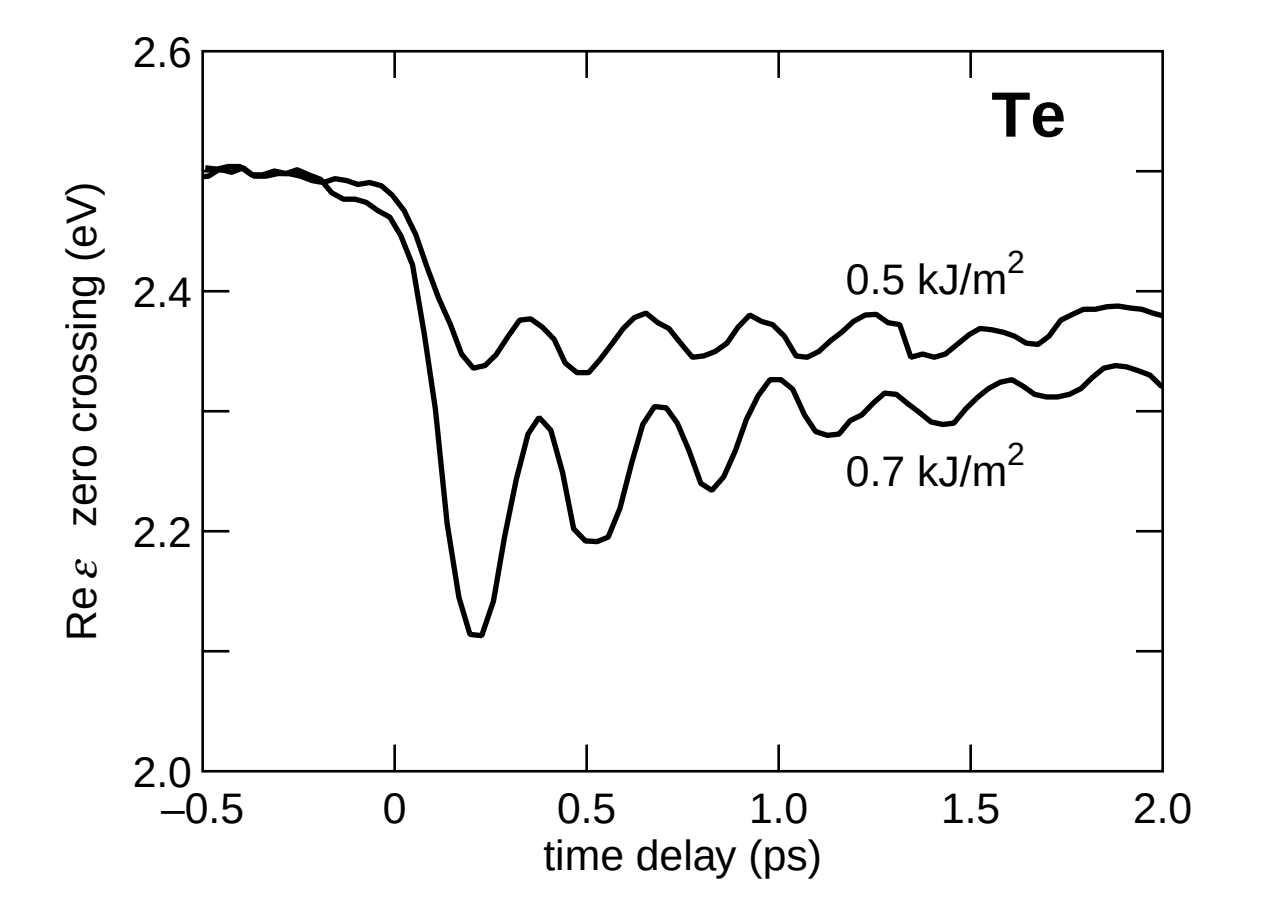
P. Tangney (Princeton) and S. Fahey (Cork), *private communication*

Results

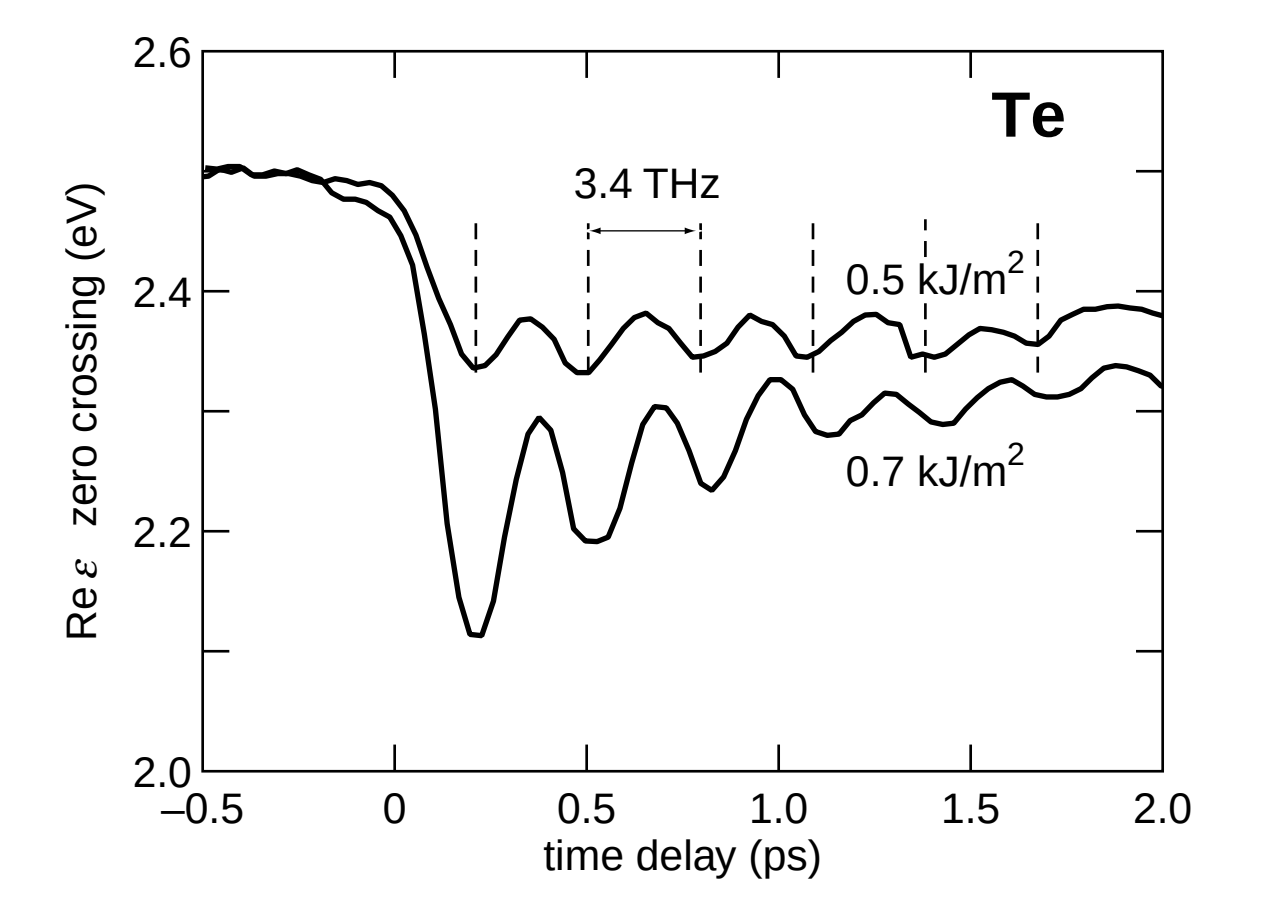
Results


Te
0.7 kJ/m2
Re ε zero crossing (eV)
2.6
2.4
2.2
2.0
–0.5
0
0.5
1.0
1.5
2.0
time delay (ps)

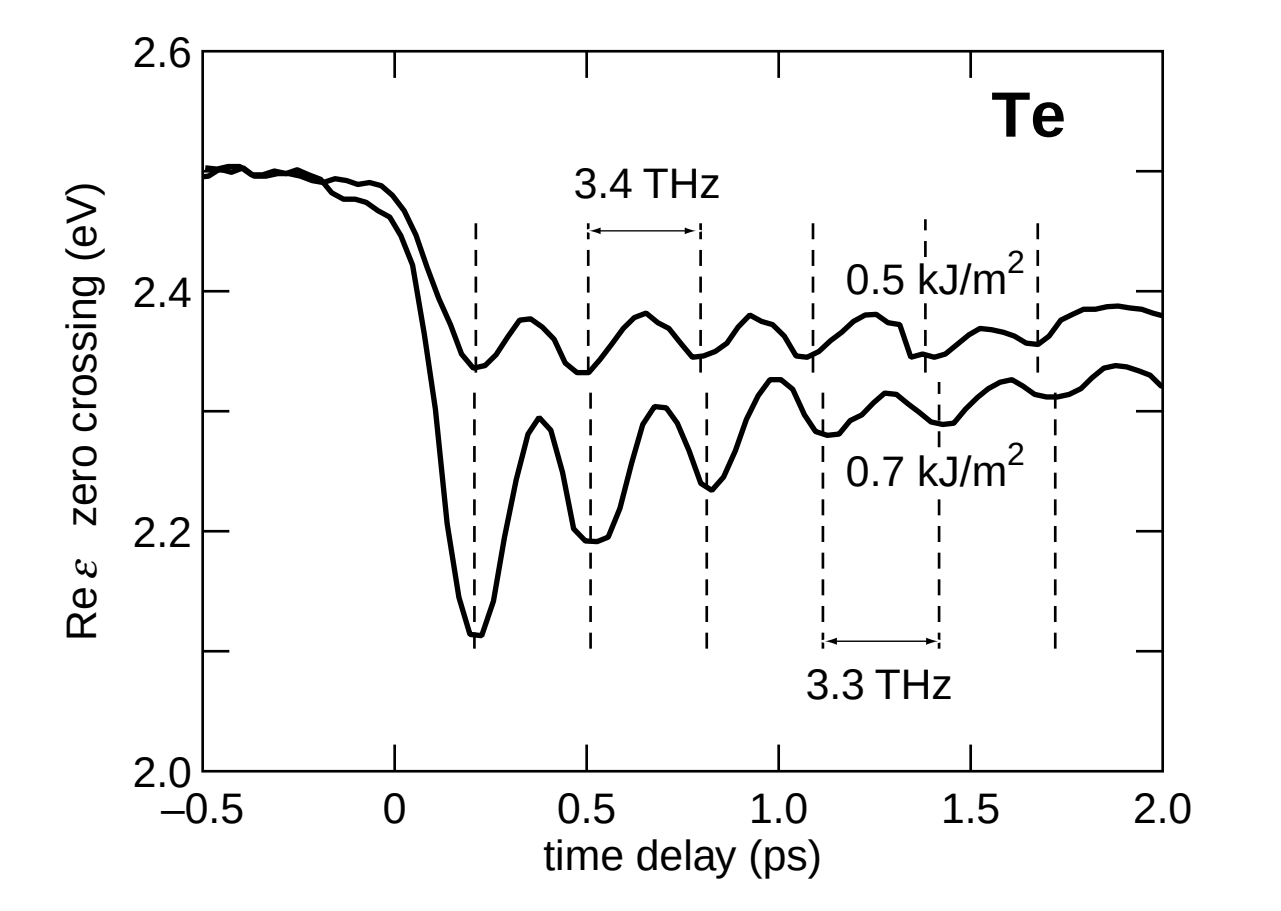
Results


Te
0.5 kJ/m2
0.7 kJ/m2
Re ε zero crossing (eV)
2.6
2.4
2.2
2.0
–0.5
0
0.5
1.0
1.5
2.0
time delay (ps)

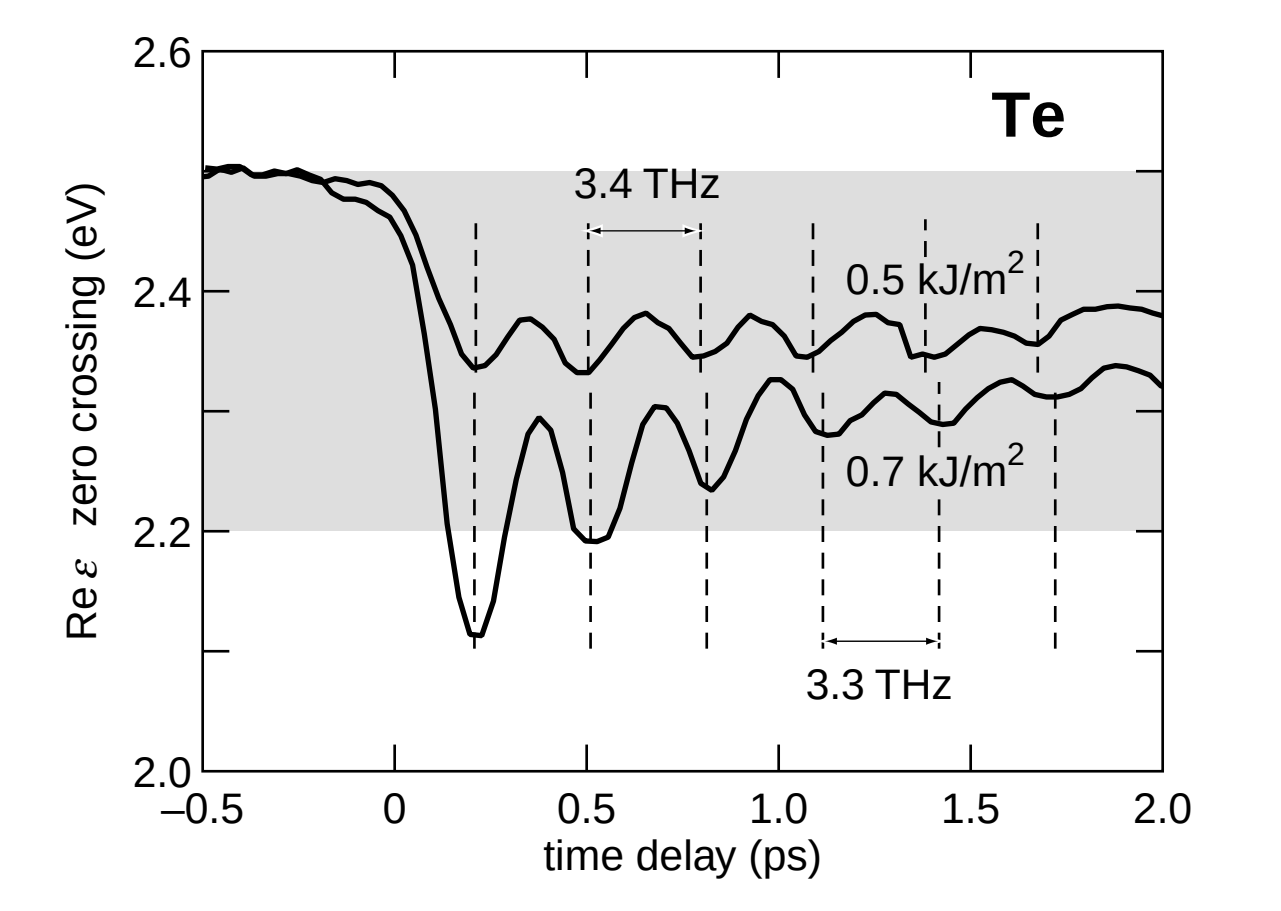
Results


Te
3.4 THz
0.5 kJ/m2
0.7 kJ/m2
Re ε zero crossing (eV)
2.6
2.4
2.2
2.0
–0.5
0
0.5
1.0
1.5
2.0
time delay (ps)

Results


Te
3.4 THz
0.5 kJ/m2
0.7 kJ/m2
3.3 THz
Re ε zero crossing (eV)
2.6
2.4
2.2
2.0
–0.5
0
0.5
1.0
1.5
2.0
time delay (ps)

Results


Te
3.4 THz
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3.3 THz
Re ε zero crossing (eV)
2.6
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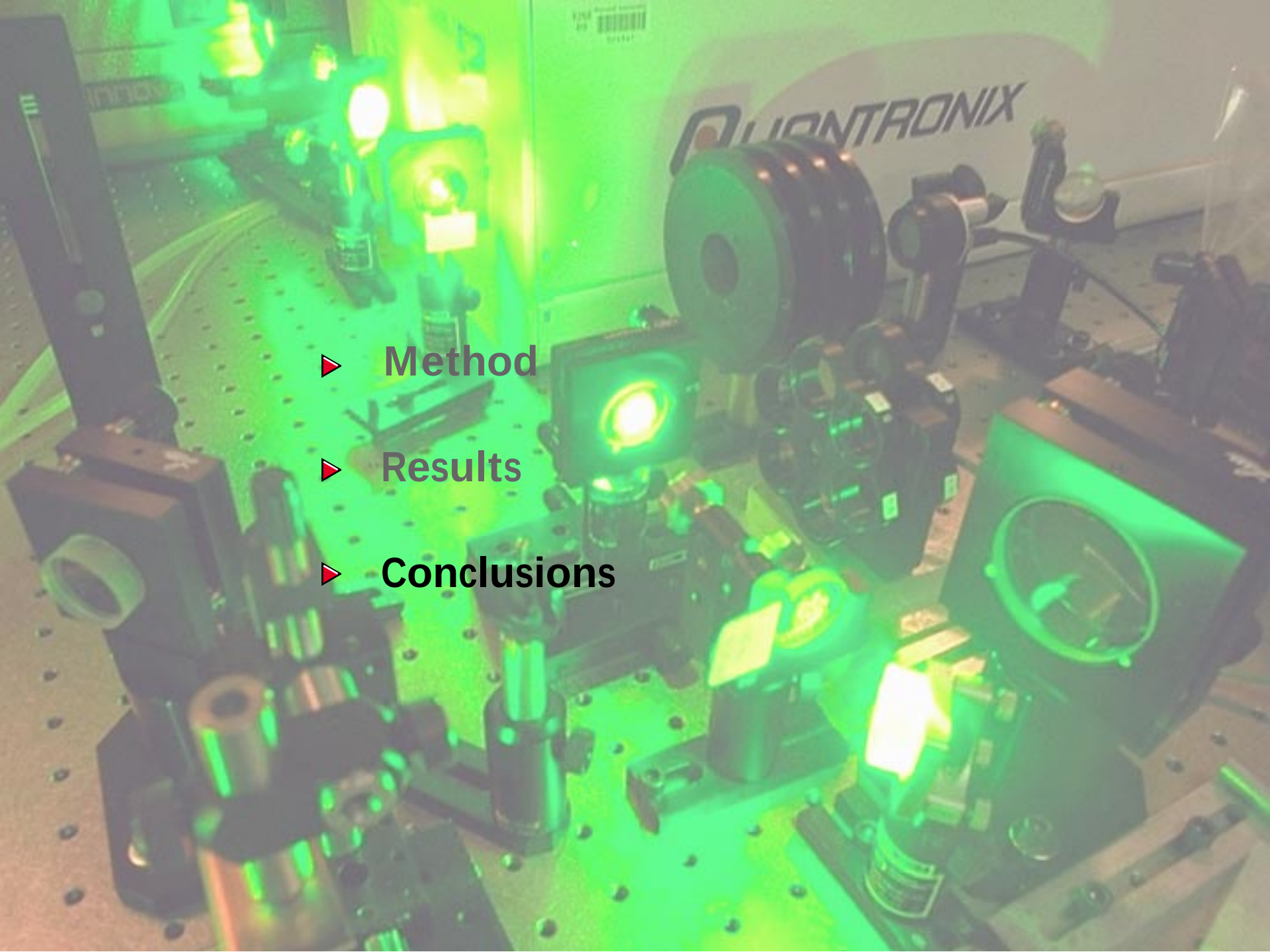
2.2
2.0
–0.5
0
0.5
1.0
1.5
2.0
time delay (ps)



- Method
- Results
- Conclusions

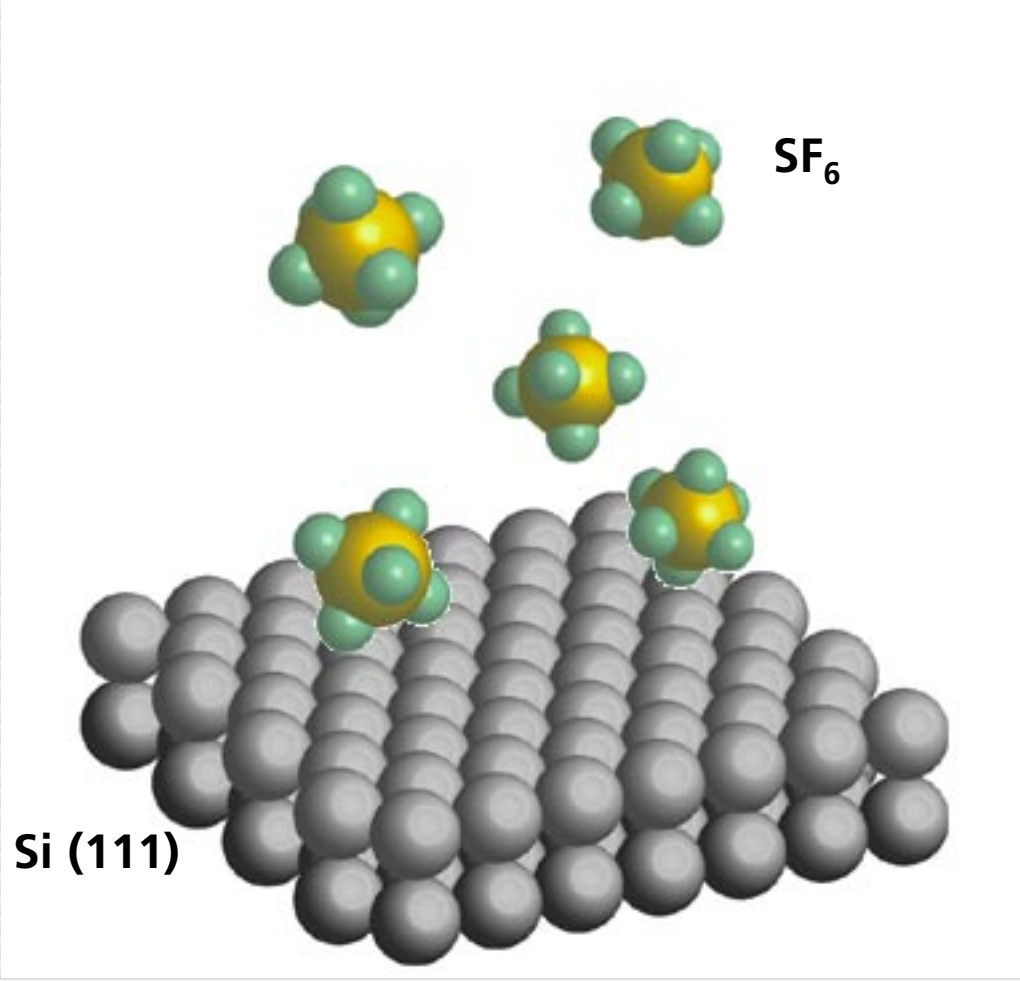
Conclusions

- **Femtosecond time-resolved ellipsometry powerful tool for tracking ultrafast electron and lattice dynamics in highly excited solids**
- **Strong electronic excitation can drive a structural transition**
- **Direct observation of coherent phonons**

Outline

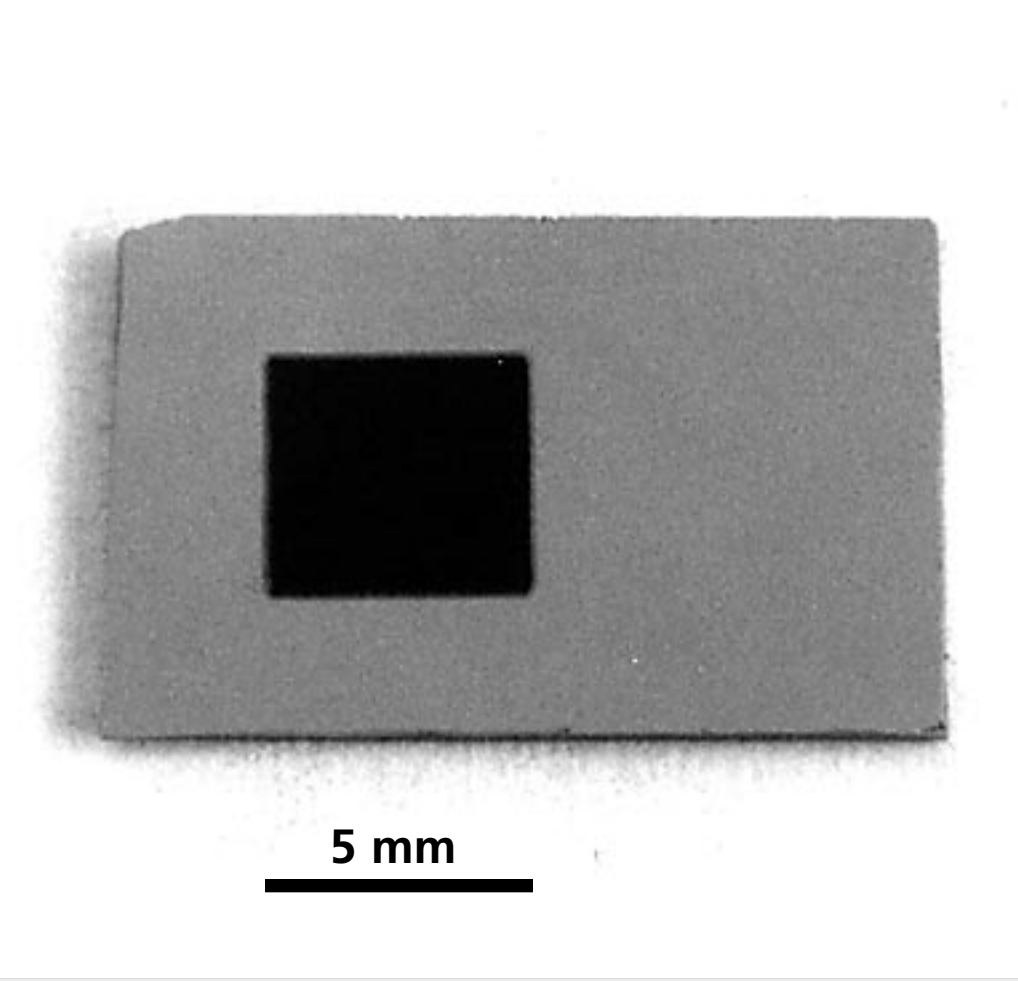
- linear and nonlinear propagation
- femtosecond measurements
- dynamics of phase transitions
- **laser-assisted surface etching**

Introduction



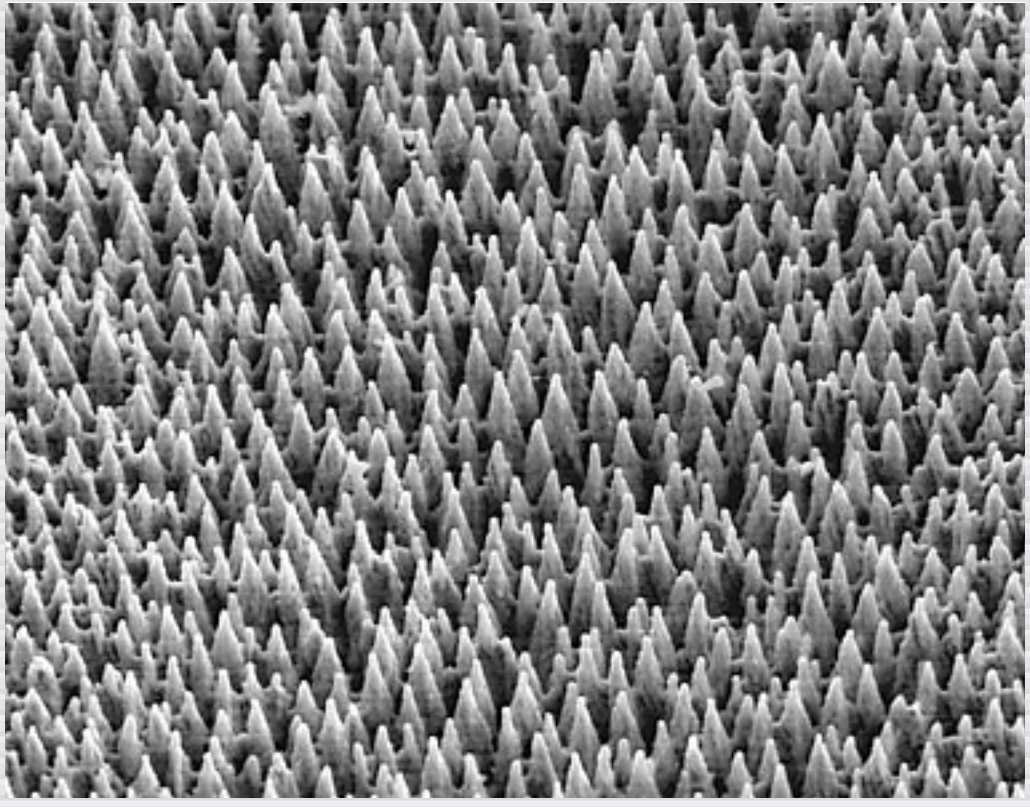
irradiate with 100-fs 10 kJ/m^2 pulses

Introduction



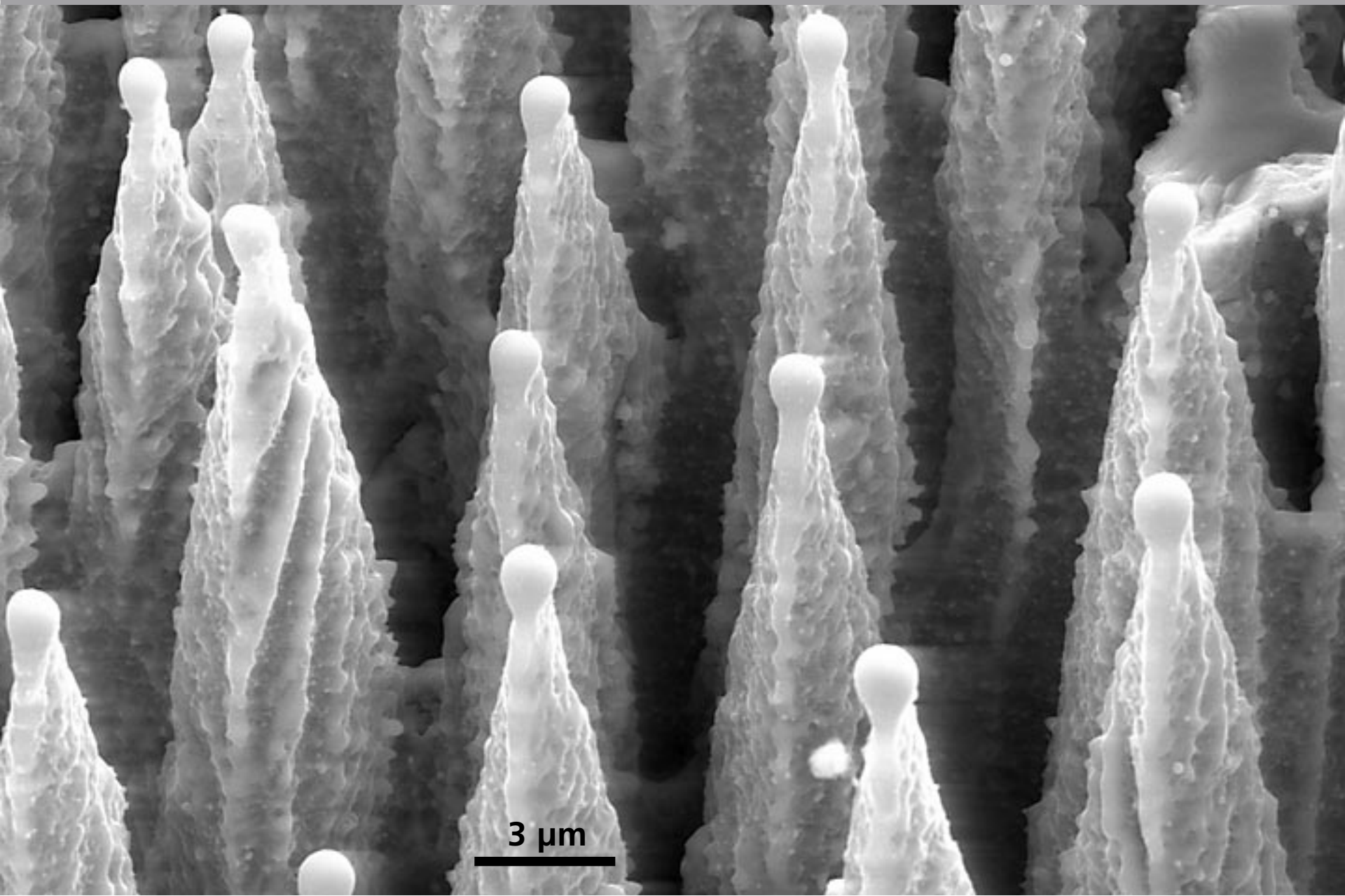
“black silicon”

Introduction

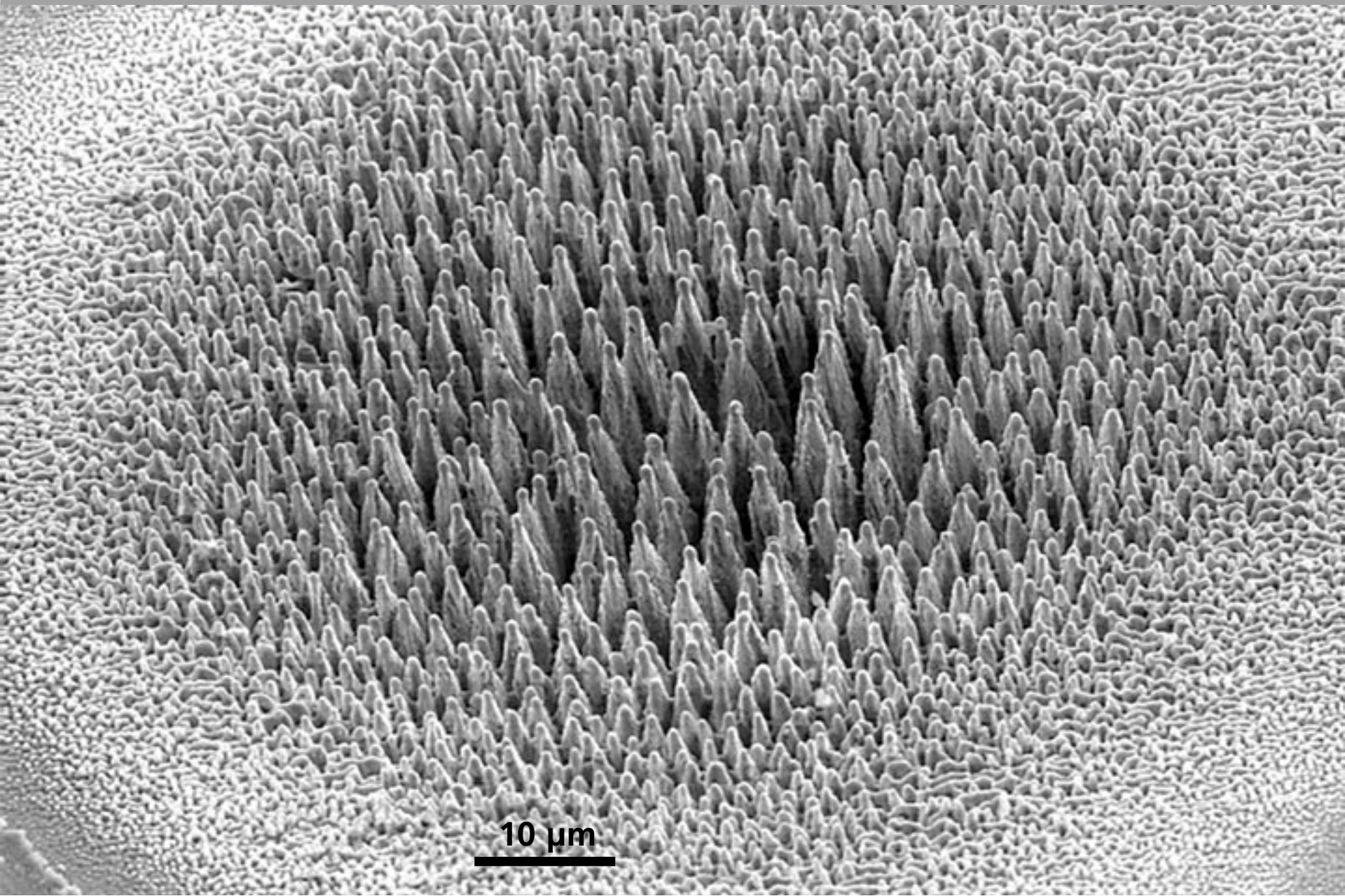


20 μm

Introduction

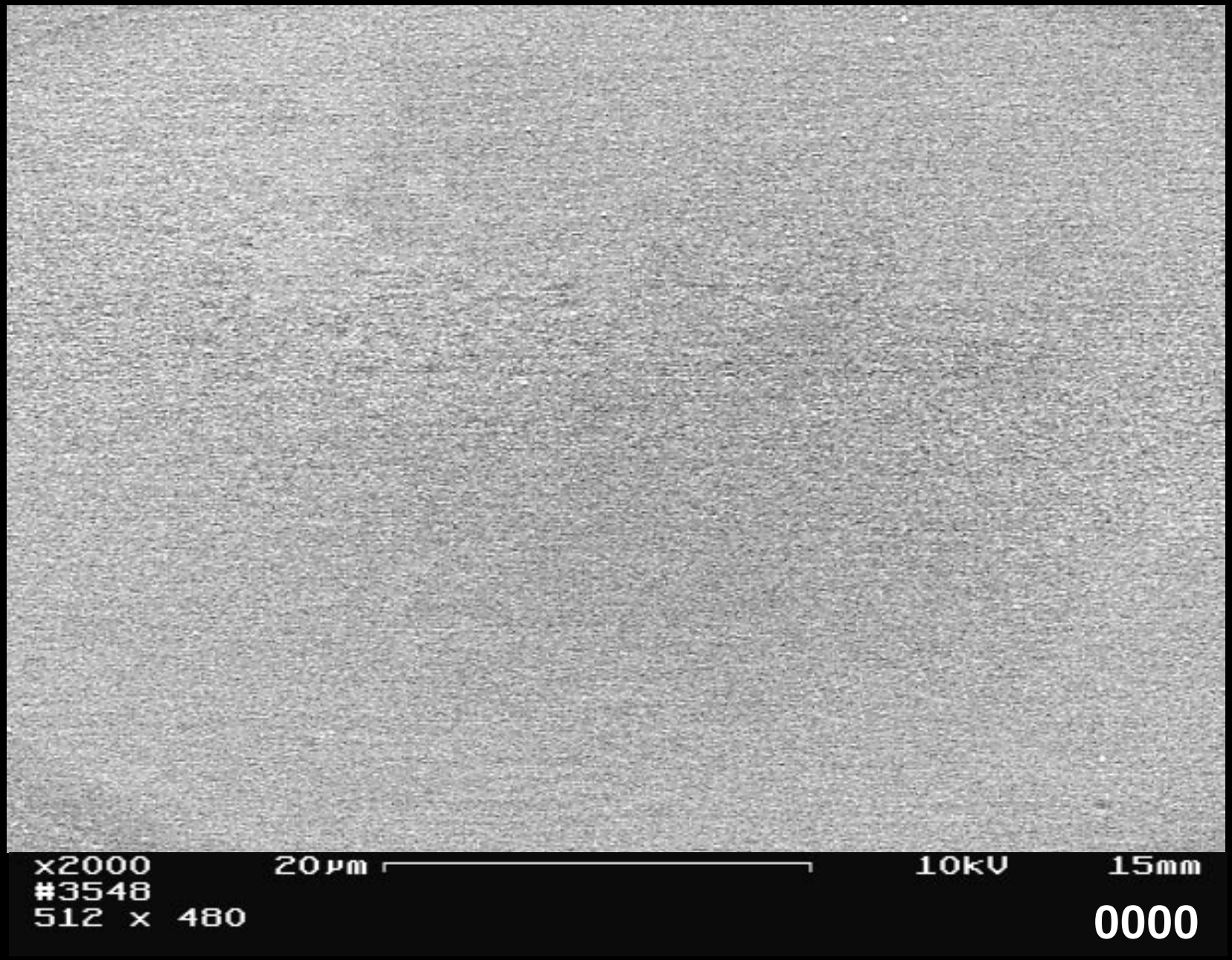


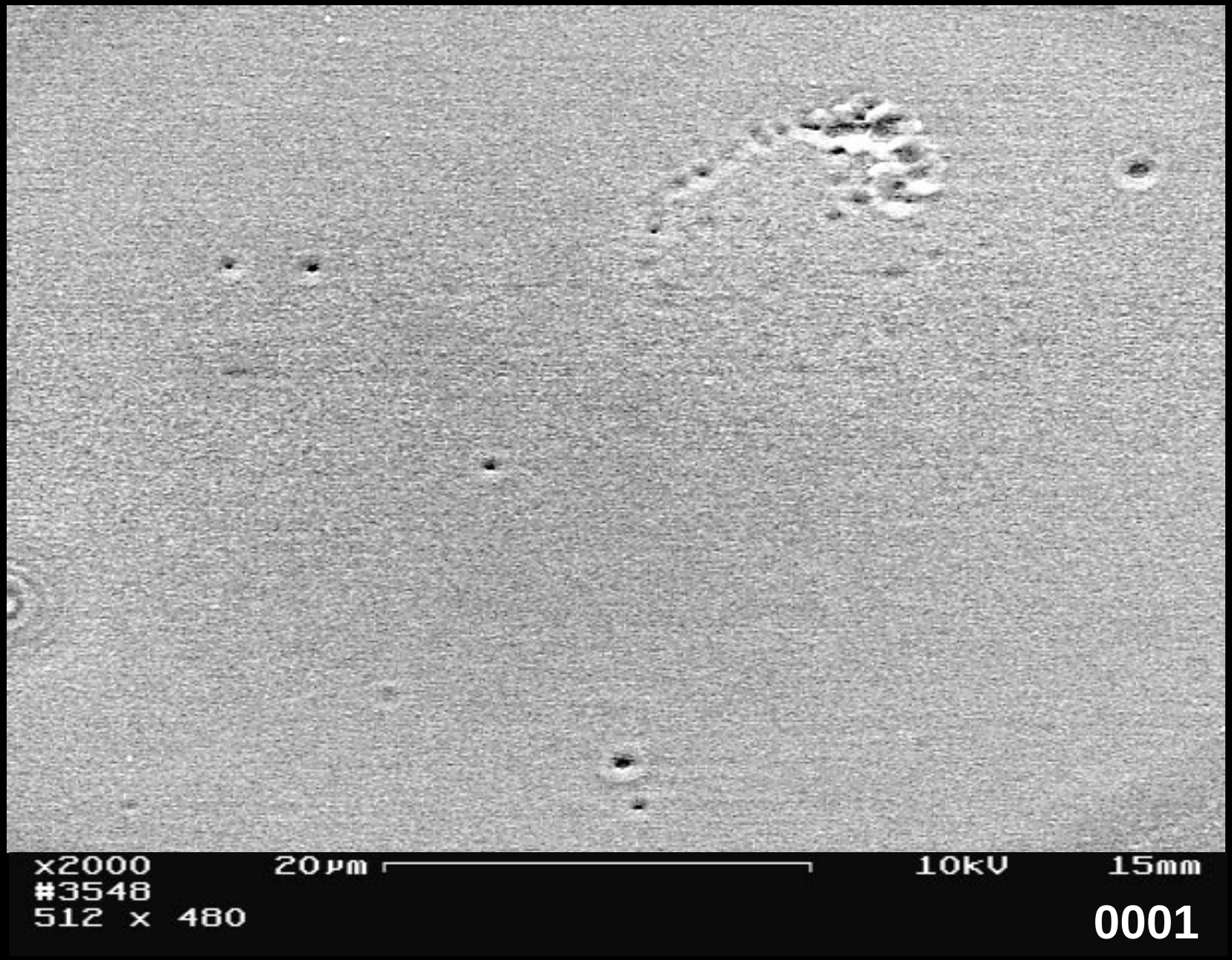
Introduction

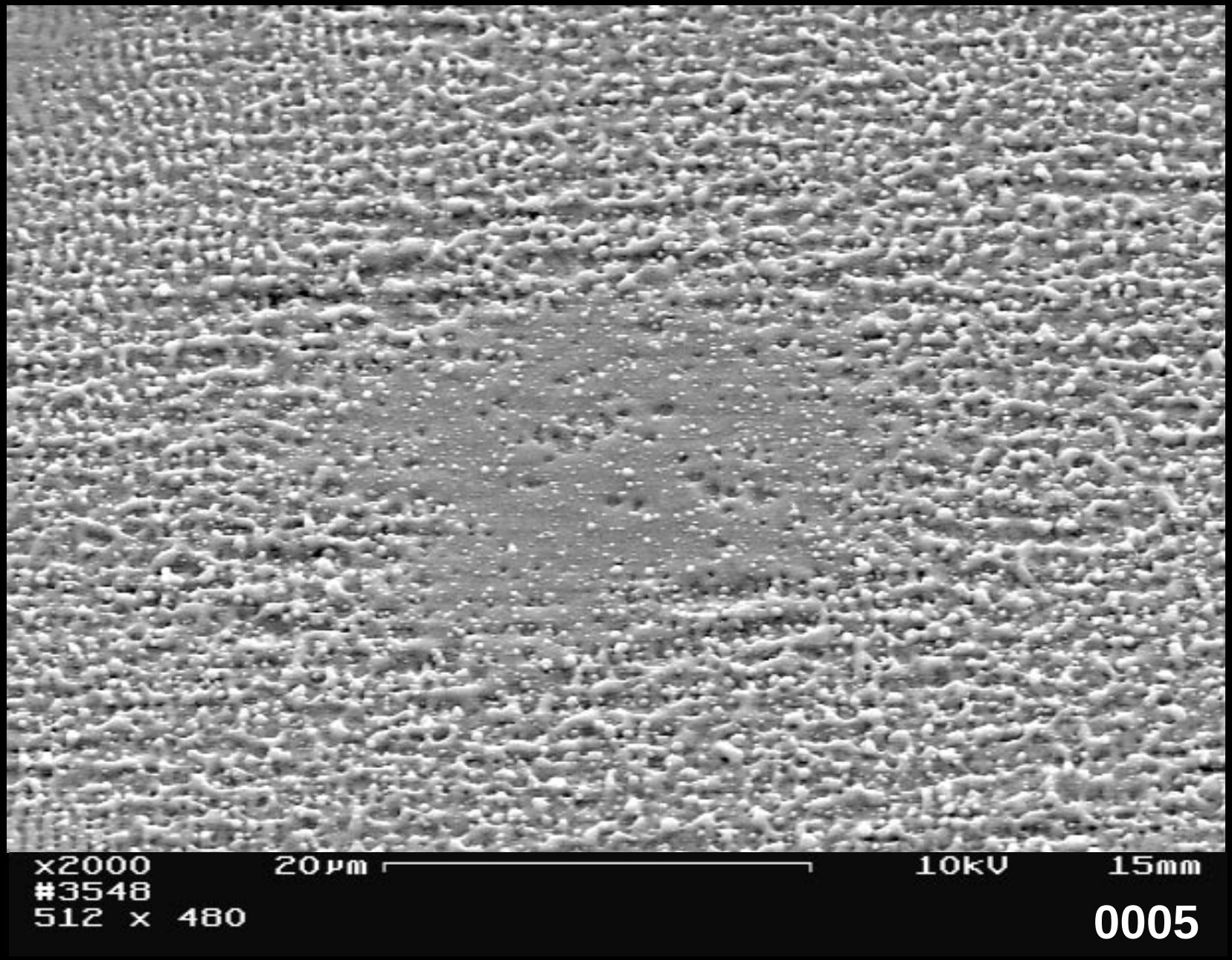


Outline

- Background
- Results
- Discussion


x2000
20µm
10kV
15mm
#3548
512 x 480
0000


x2000
20µm
10kV
15mm
#3548
512 x 480
0001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20µm
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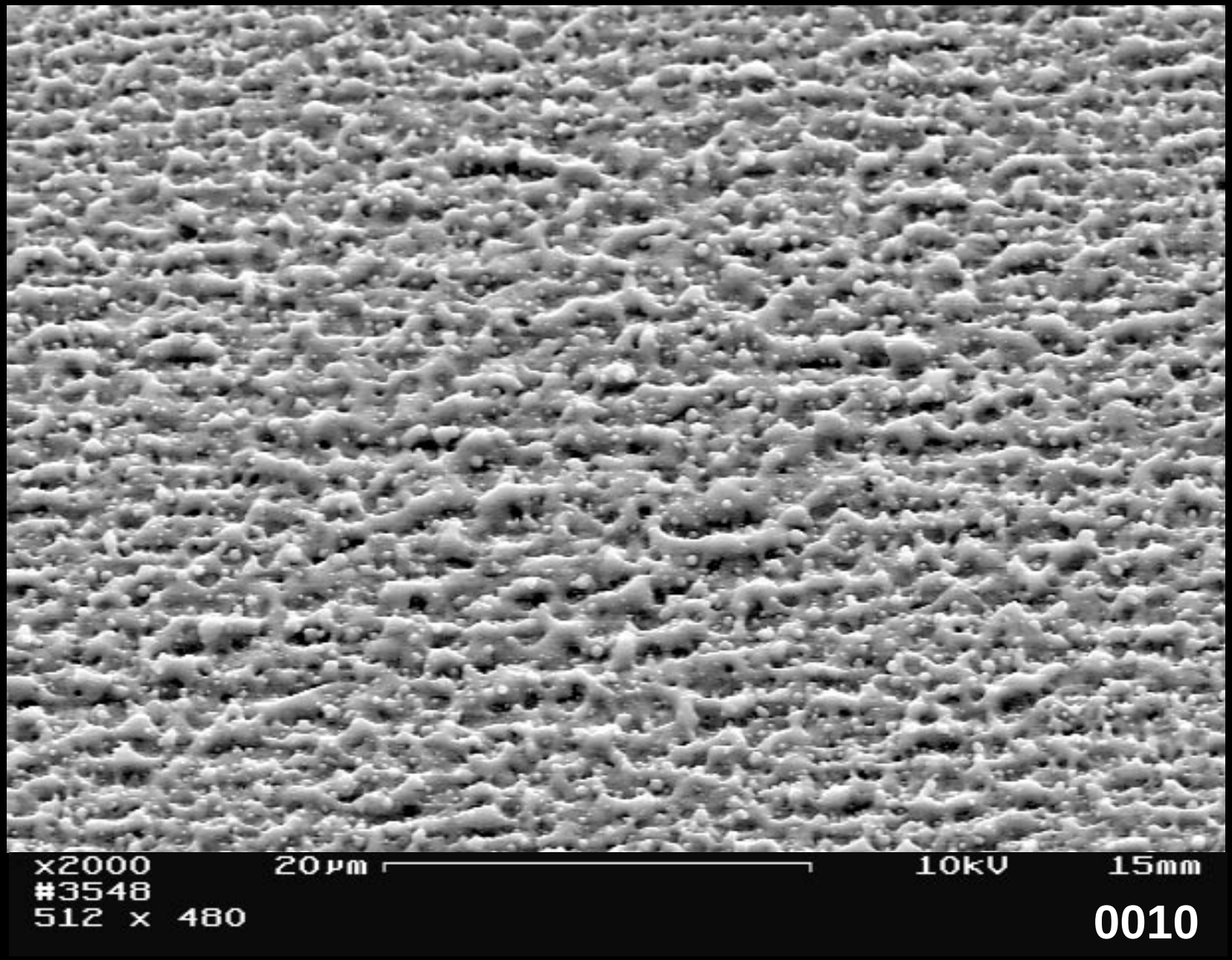
0005


x2000
20µm
10kV
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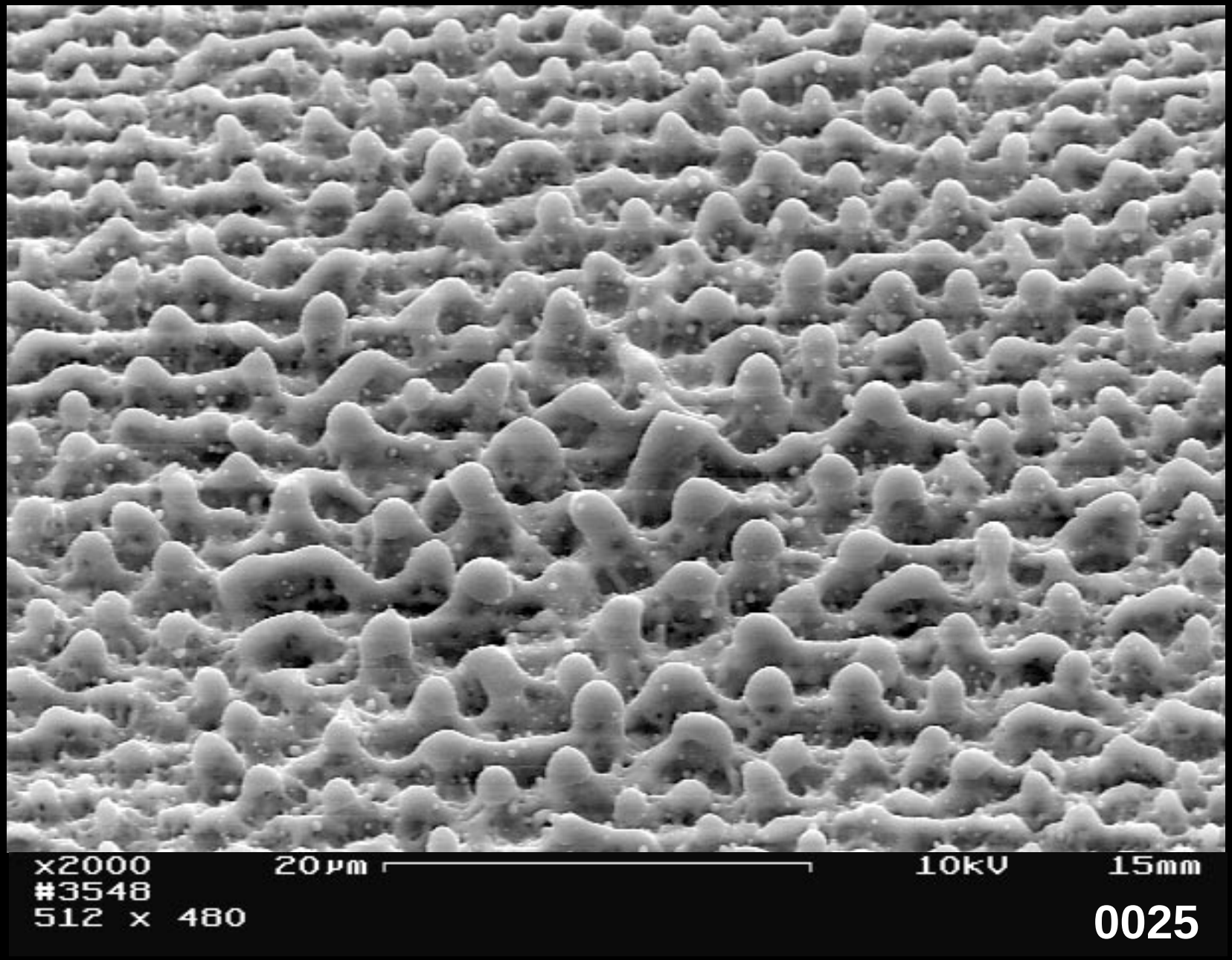
512 x 480
0010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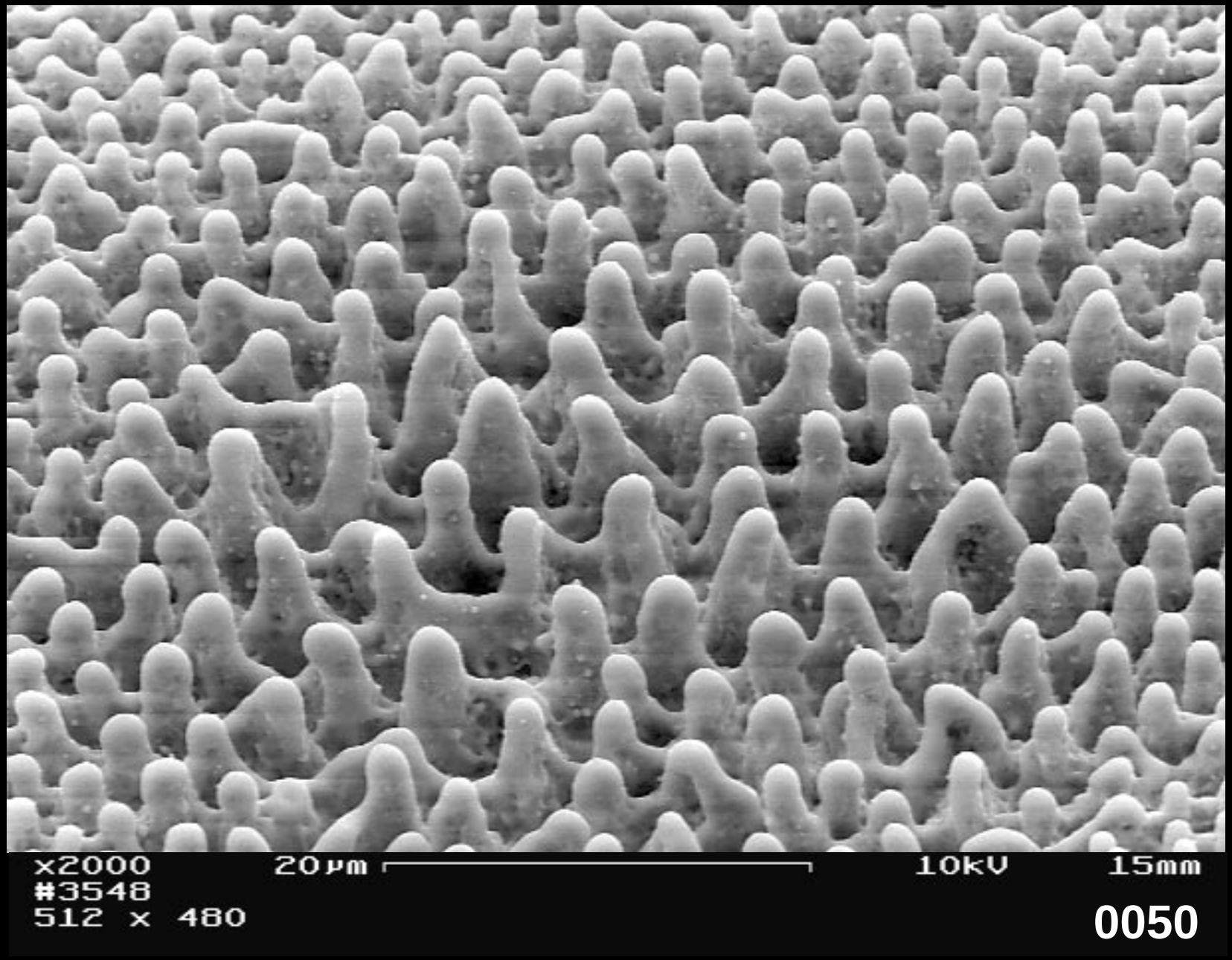
0025


x2000
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512 x 480
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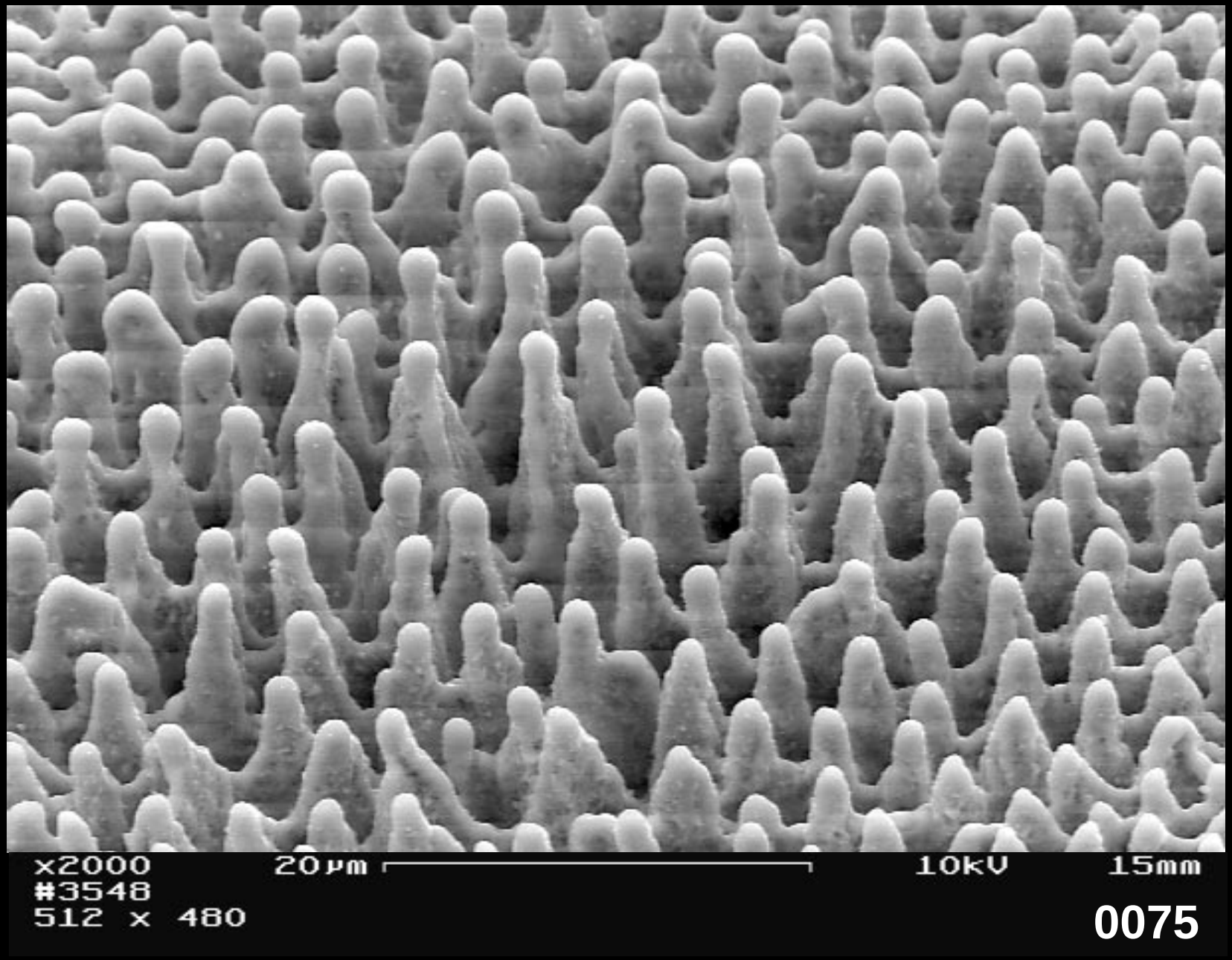
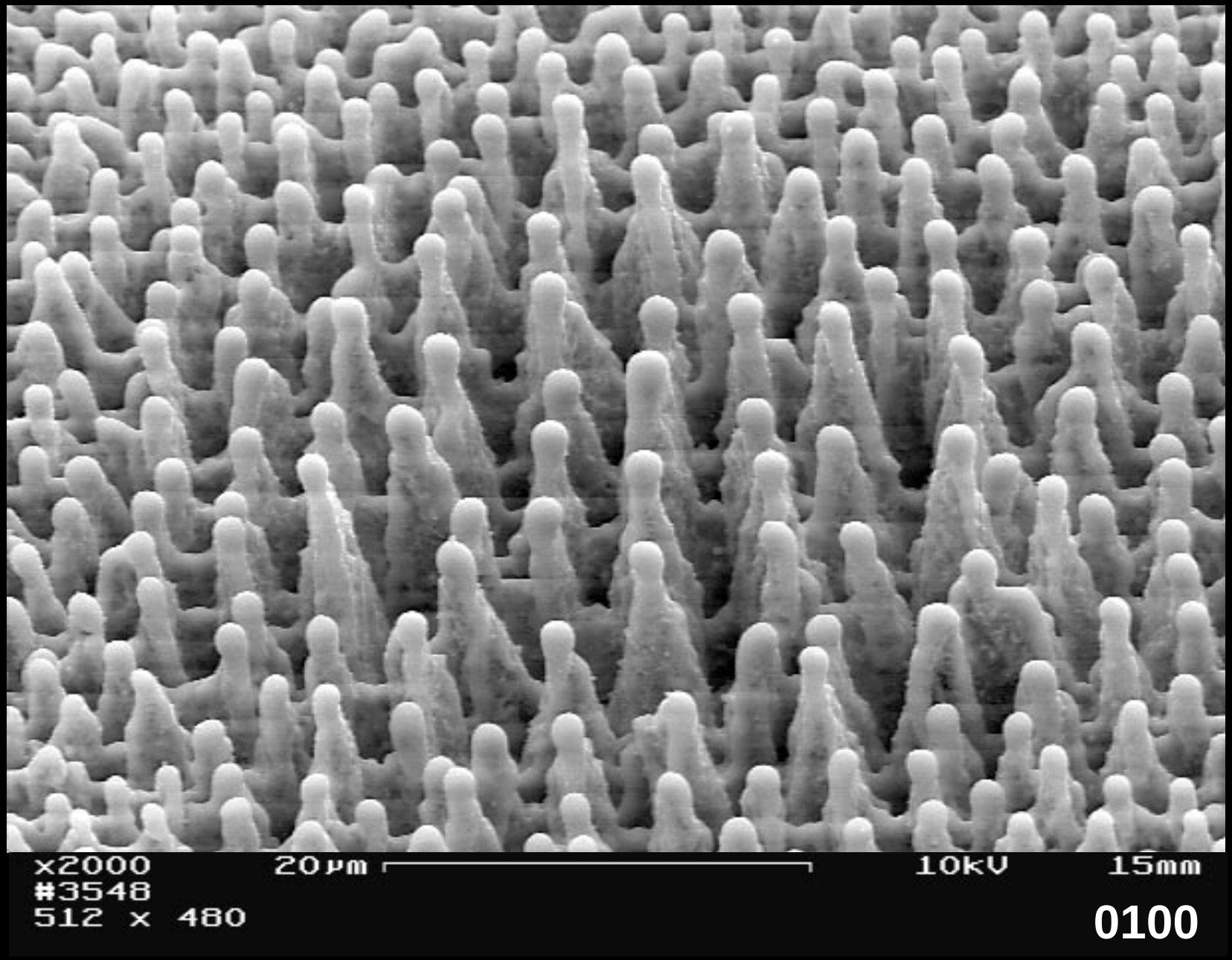
10kV
15mm
0050


x2000
#3548
512 x 480
20µm
10kV
15mm
0075


x2000
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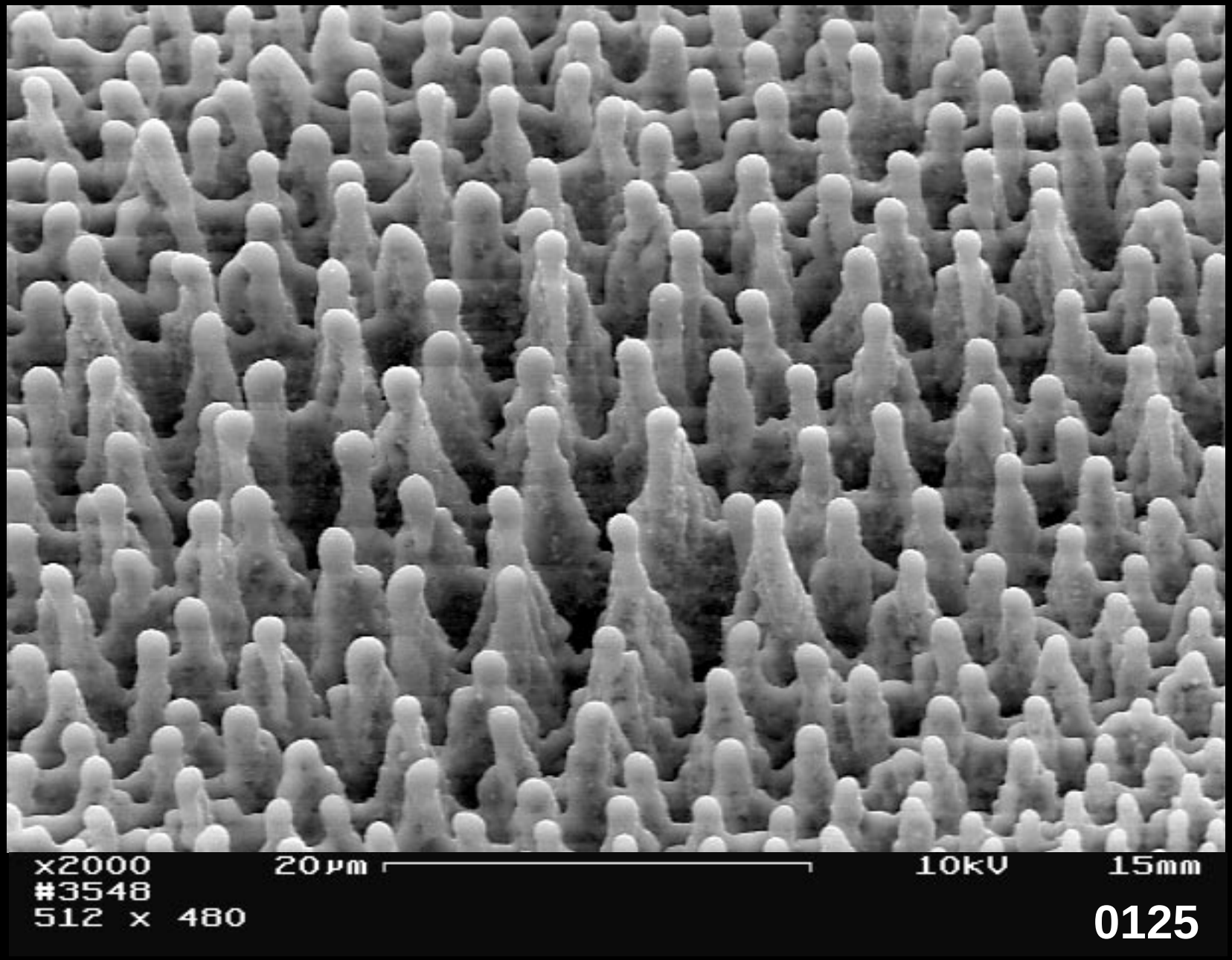
512 x 480
20µm
10kV
15mm
0100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20µm
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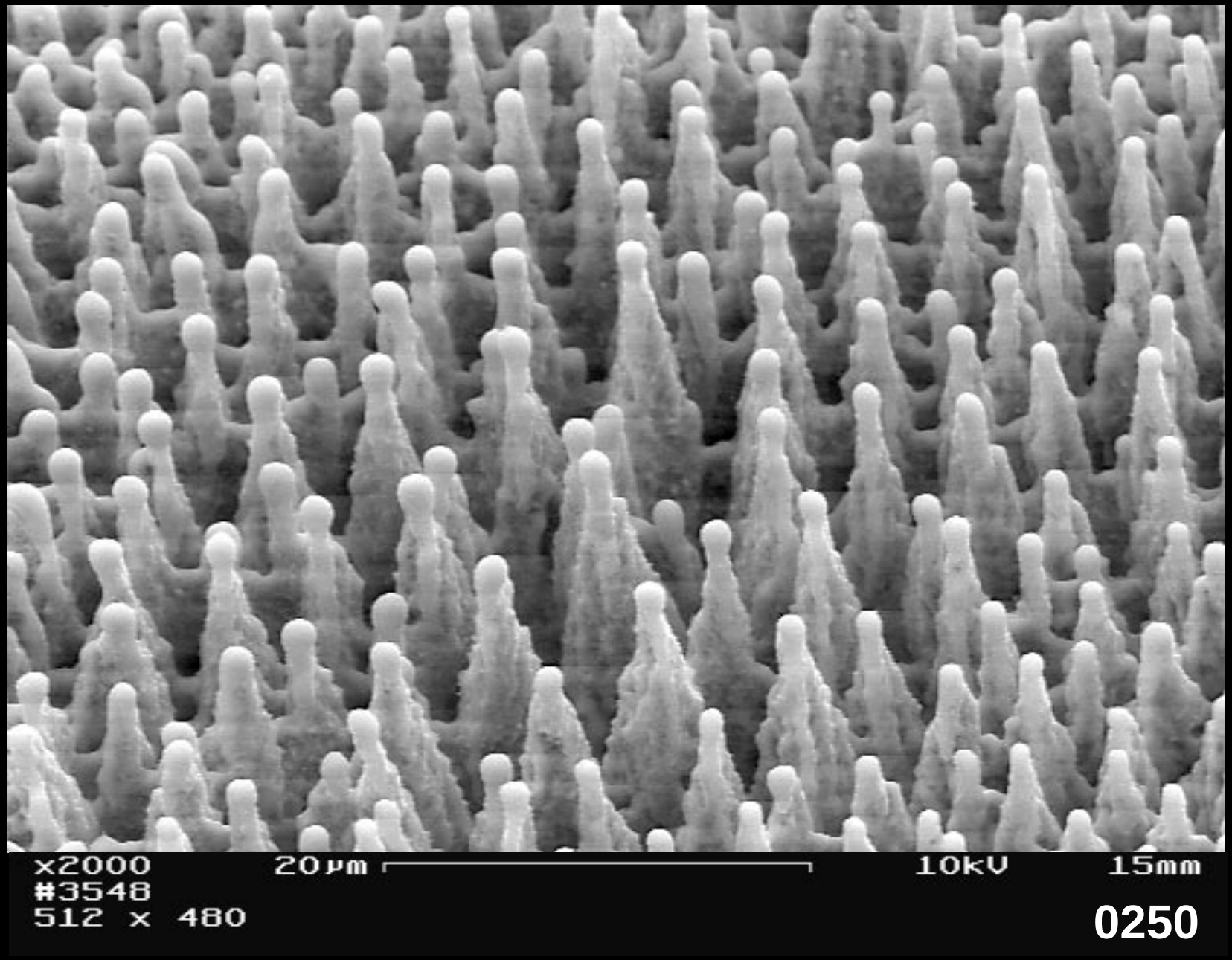
15mm
0125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20µm
10kV
15mm
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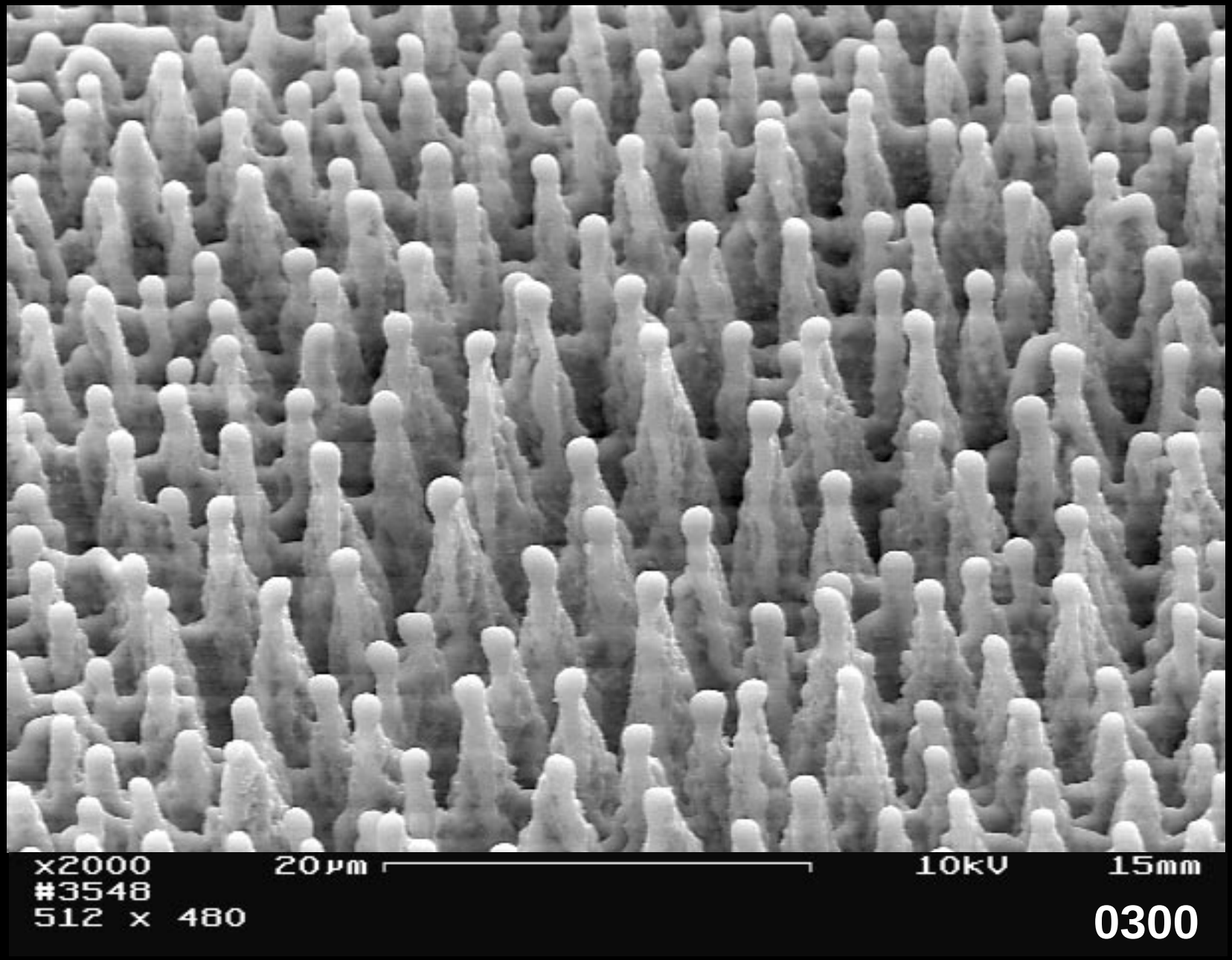
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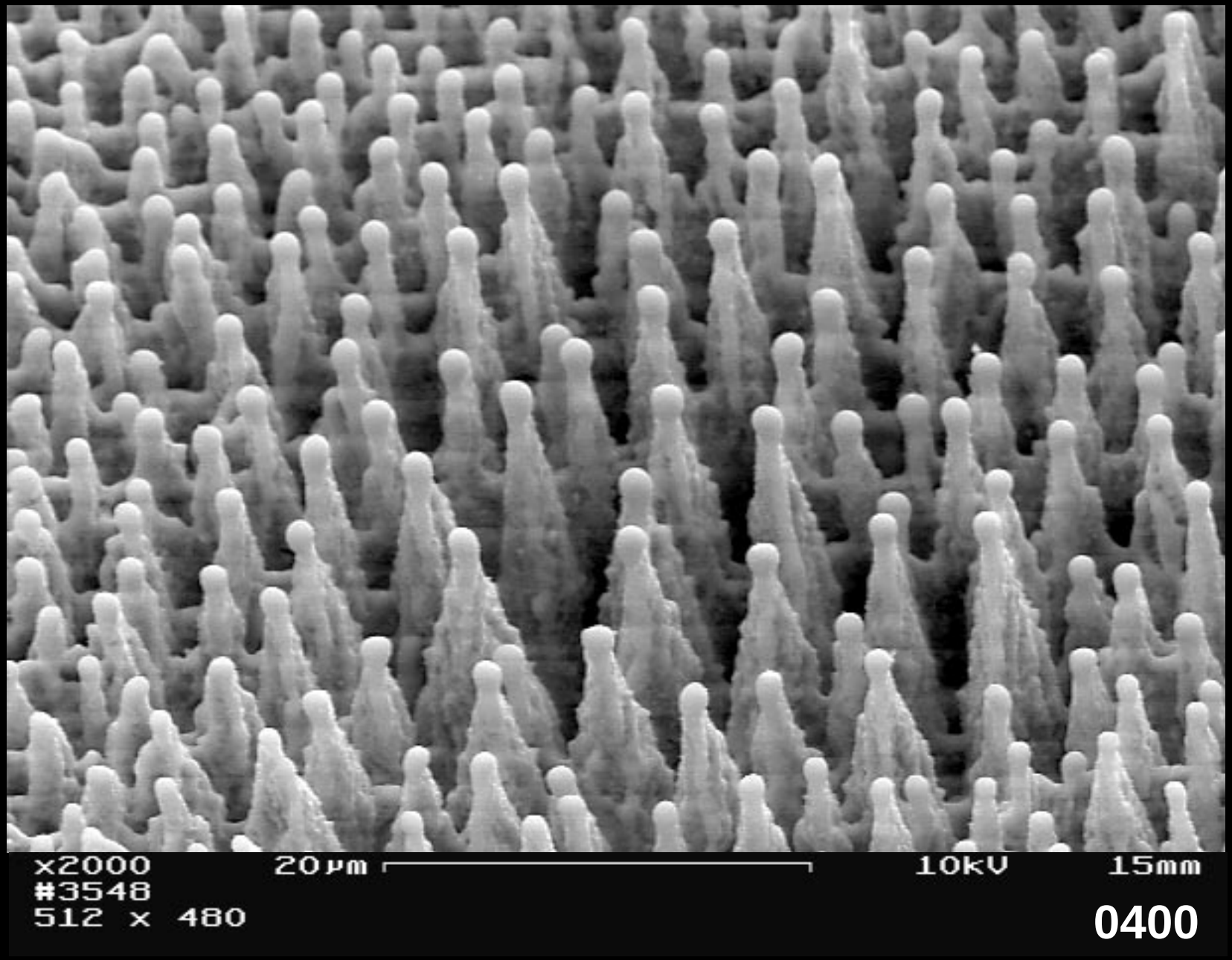
10kV
15mm
0300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10kV
15mm
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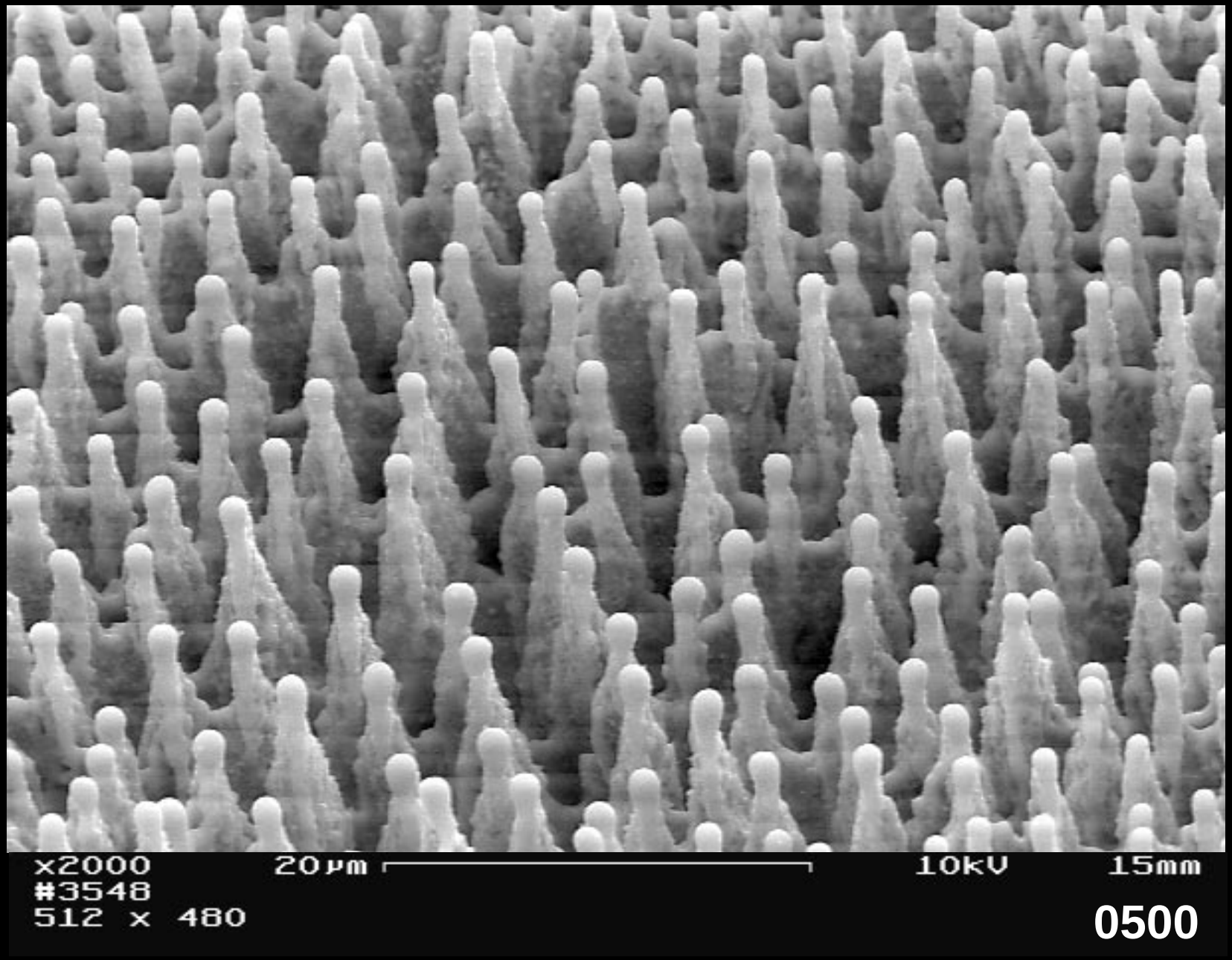
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20µm
10kV
15mm
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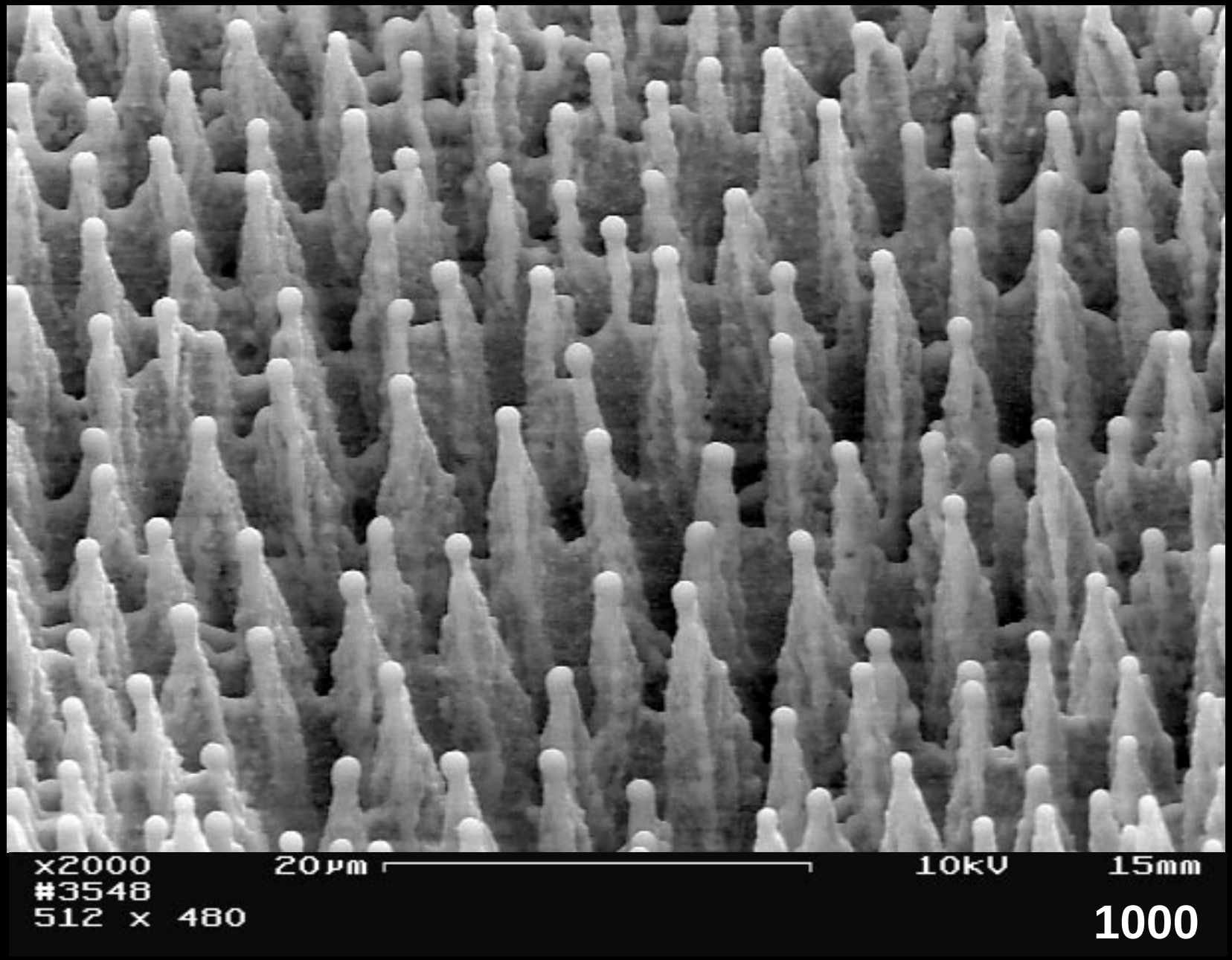
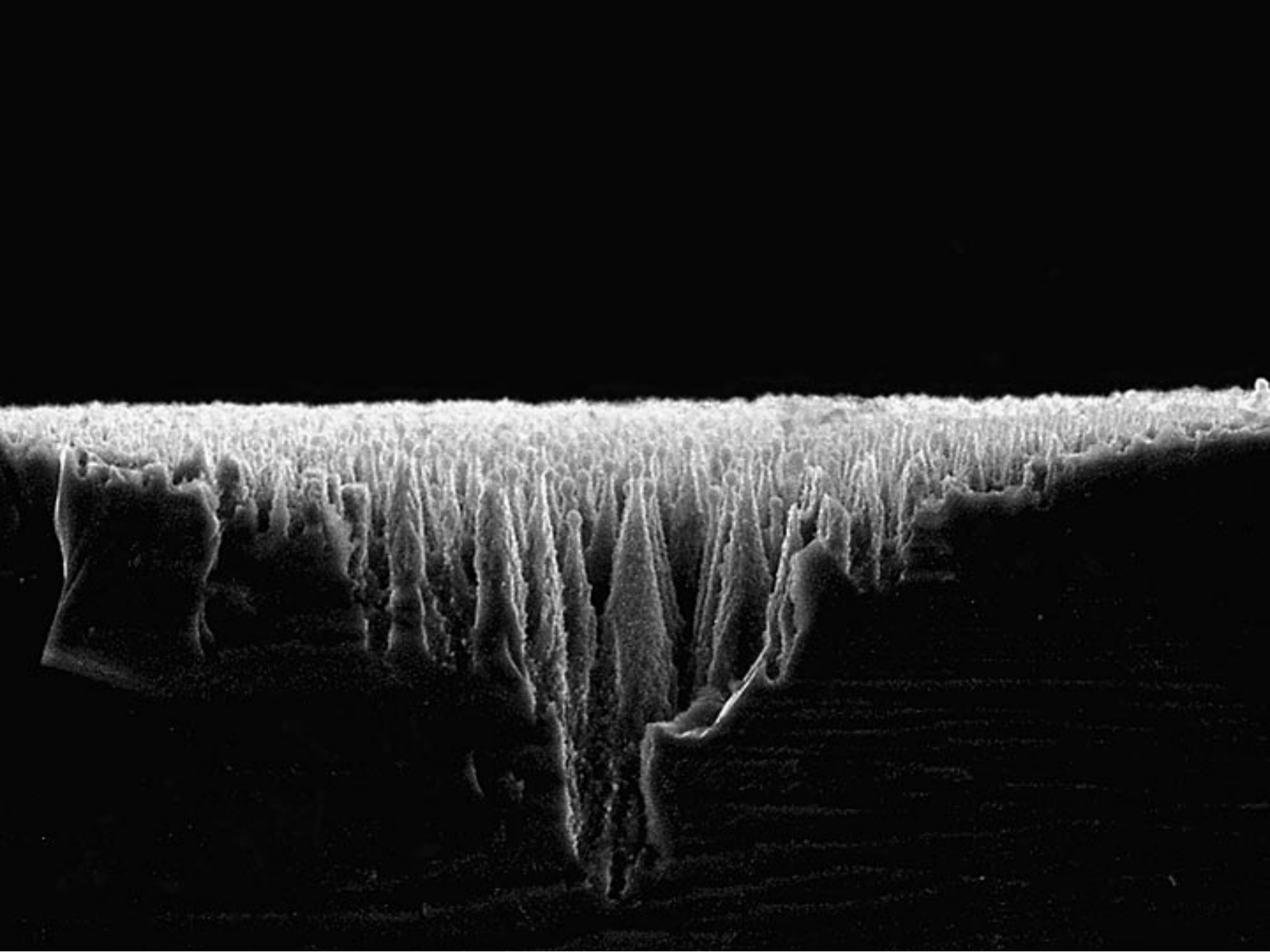
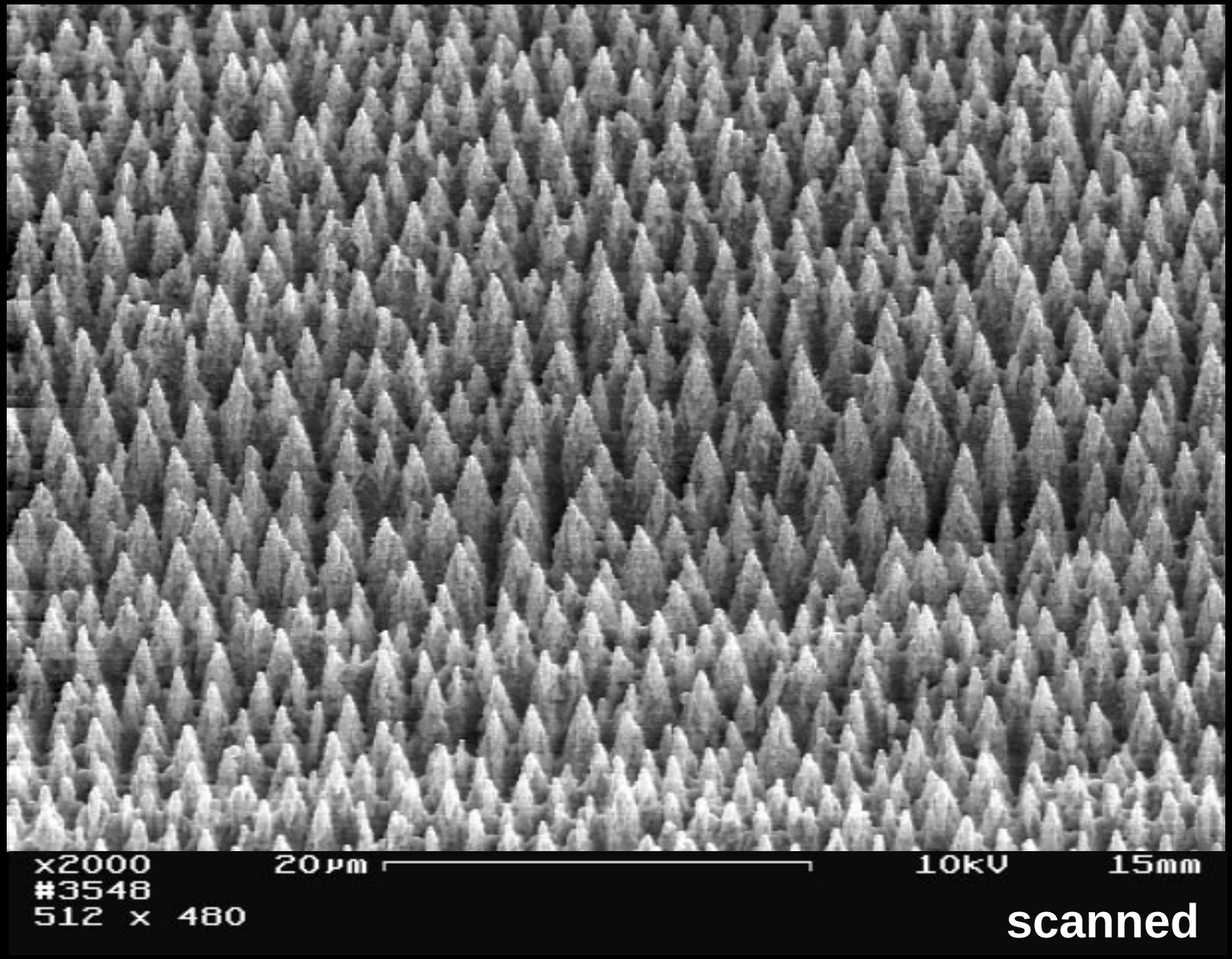
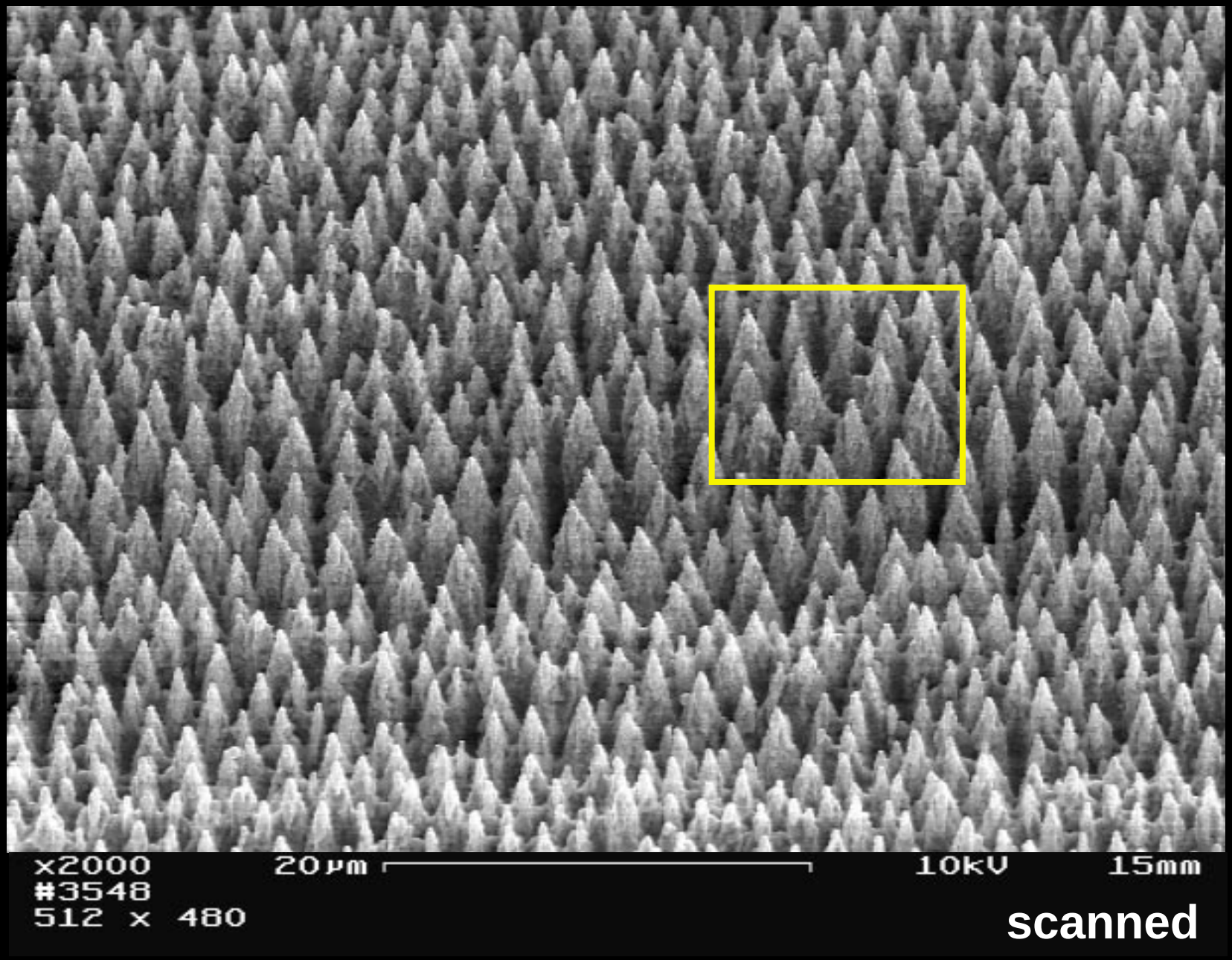
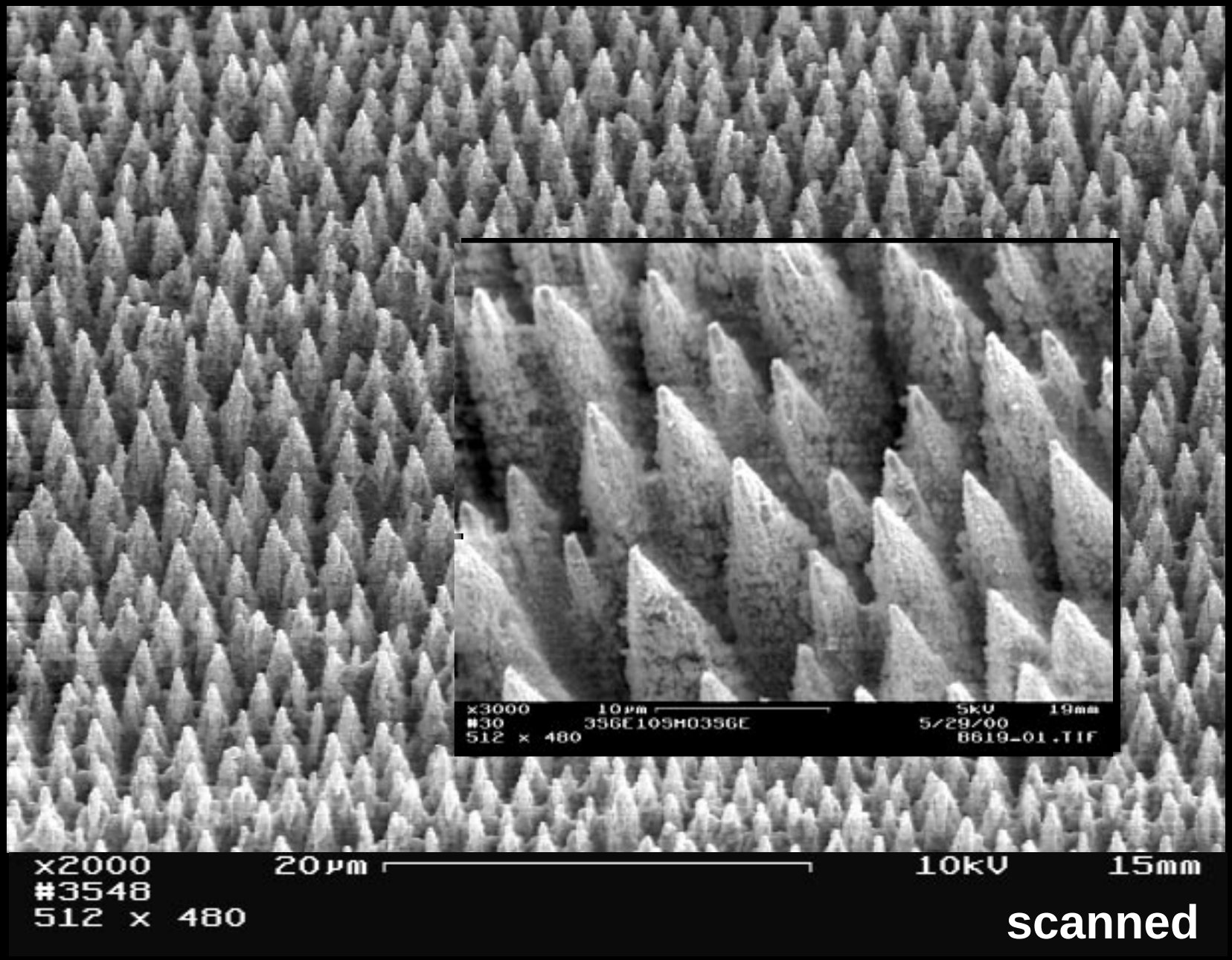

x2000
#3548
512 x 480
20µm
10kV
15mm
1000




x2000
#3548
512 x 480
20µm
10kV
15mm
scanned


x2000
#3548
512 x 480
20µm
10kV
15mm
scanned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512 x 480 8819-01.TIF
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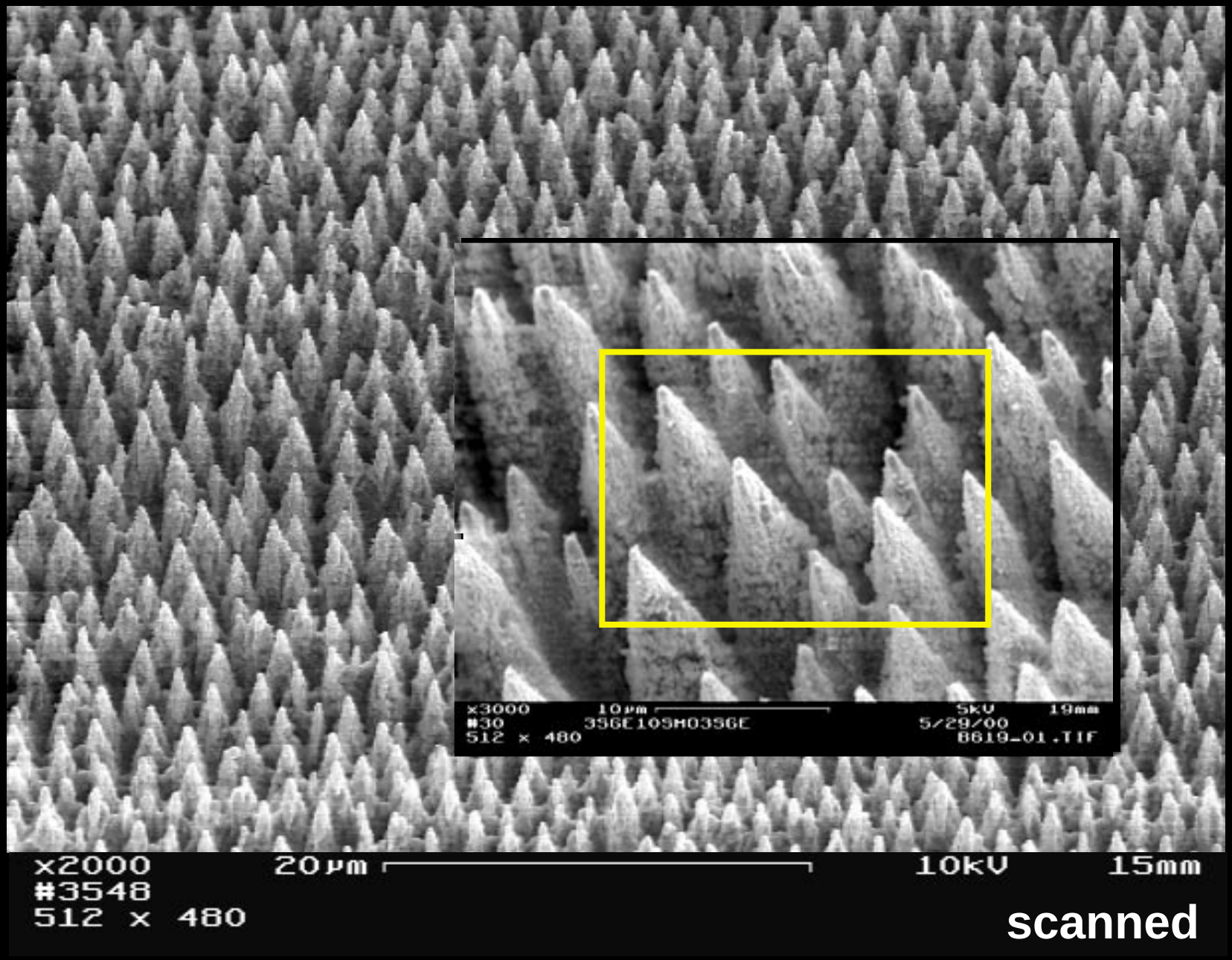
#3548
512 x 480

scanned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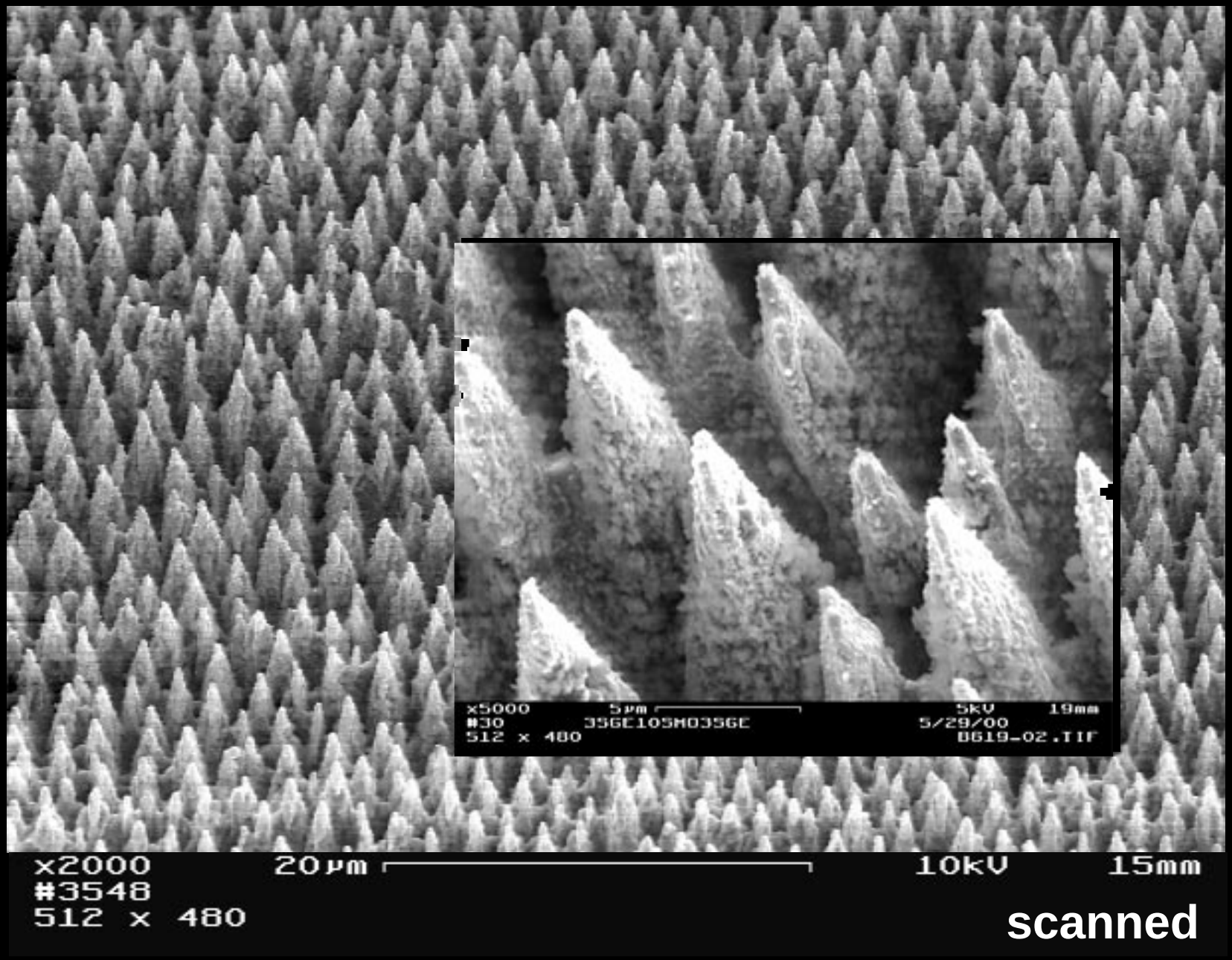
512 x 480

scanned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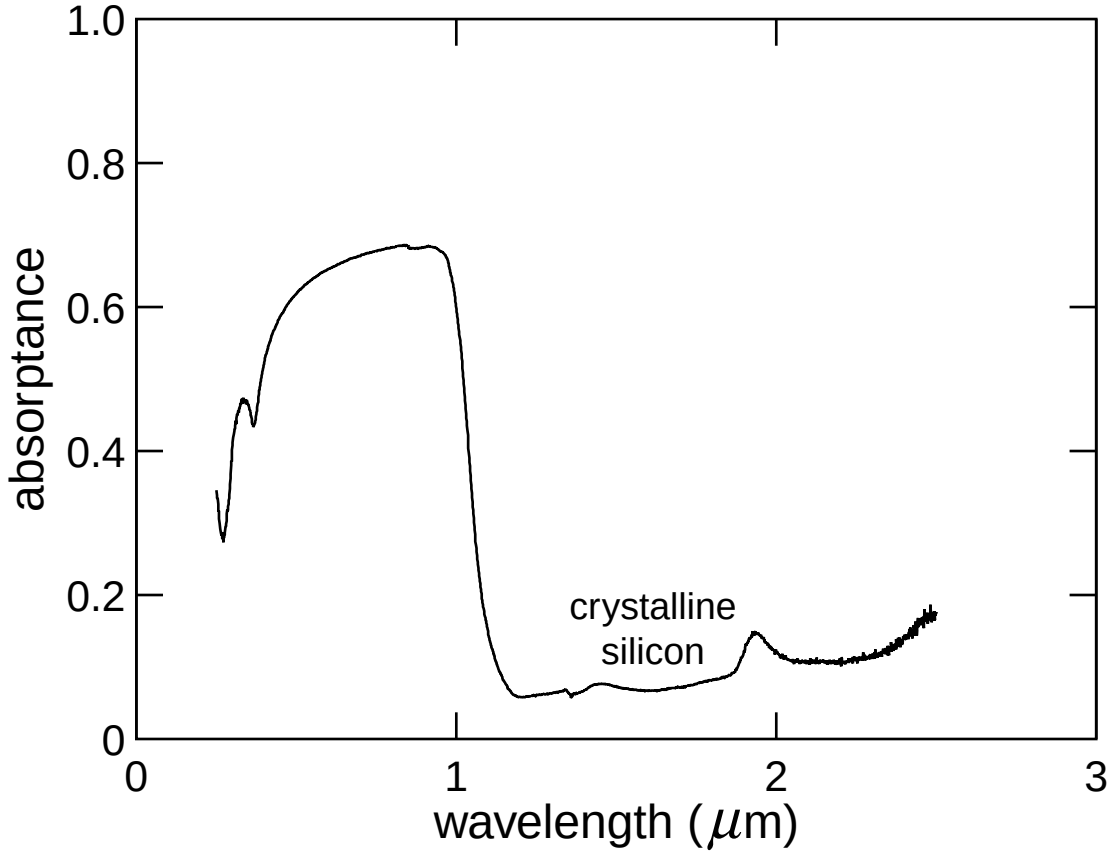
#30 35GE105M035GE 5/29/00
512 x 480 B619-02.TIF
x2000 20µm 10kV 15mm
#3548
512 x 480

scanned

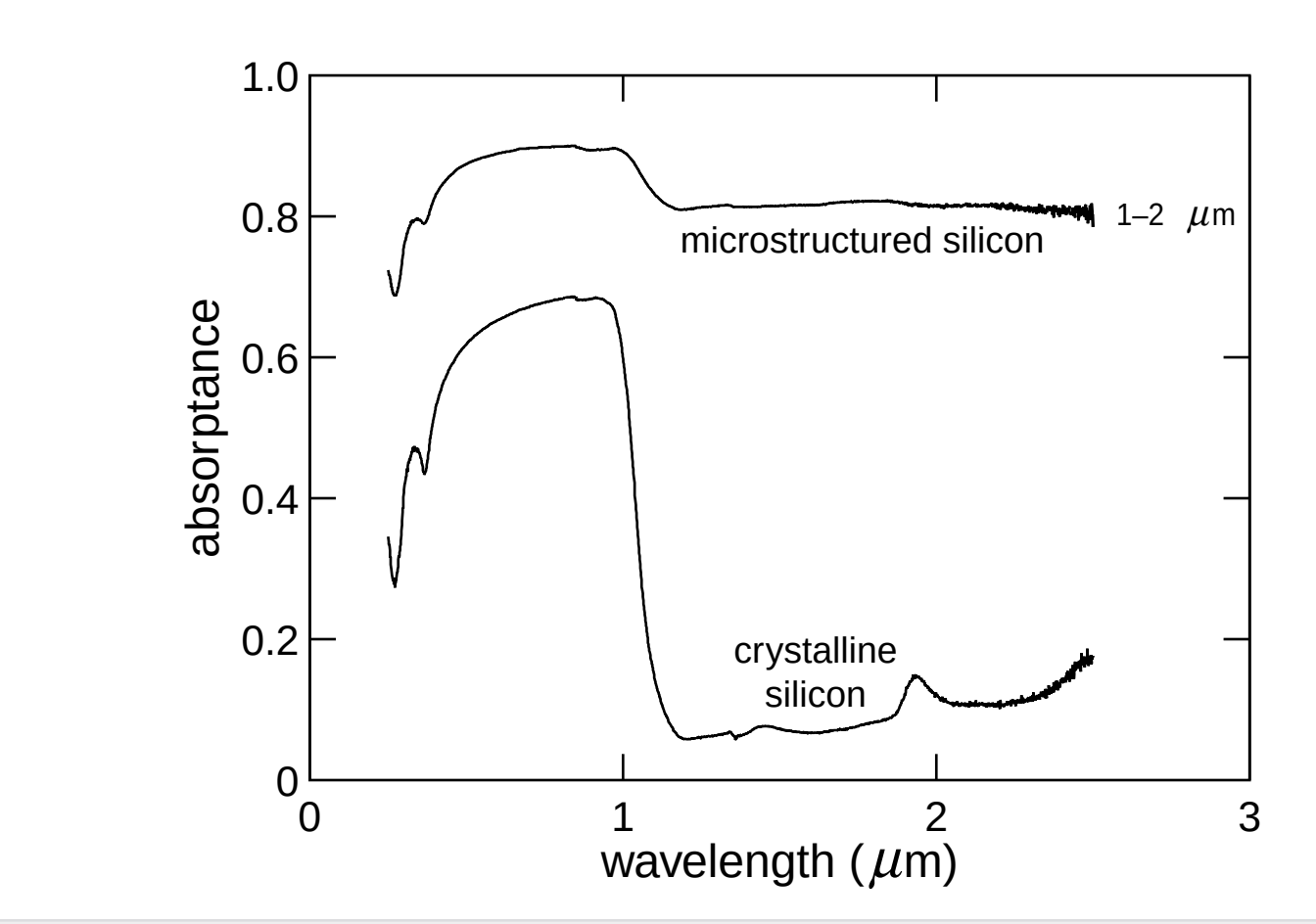
Background

absorptance



Background

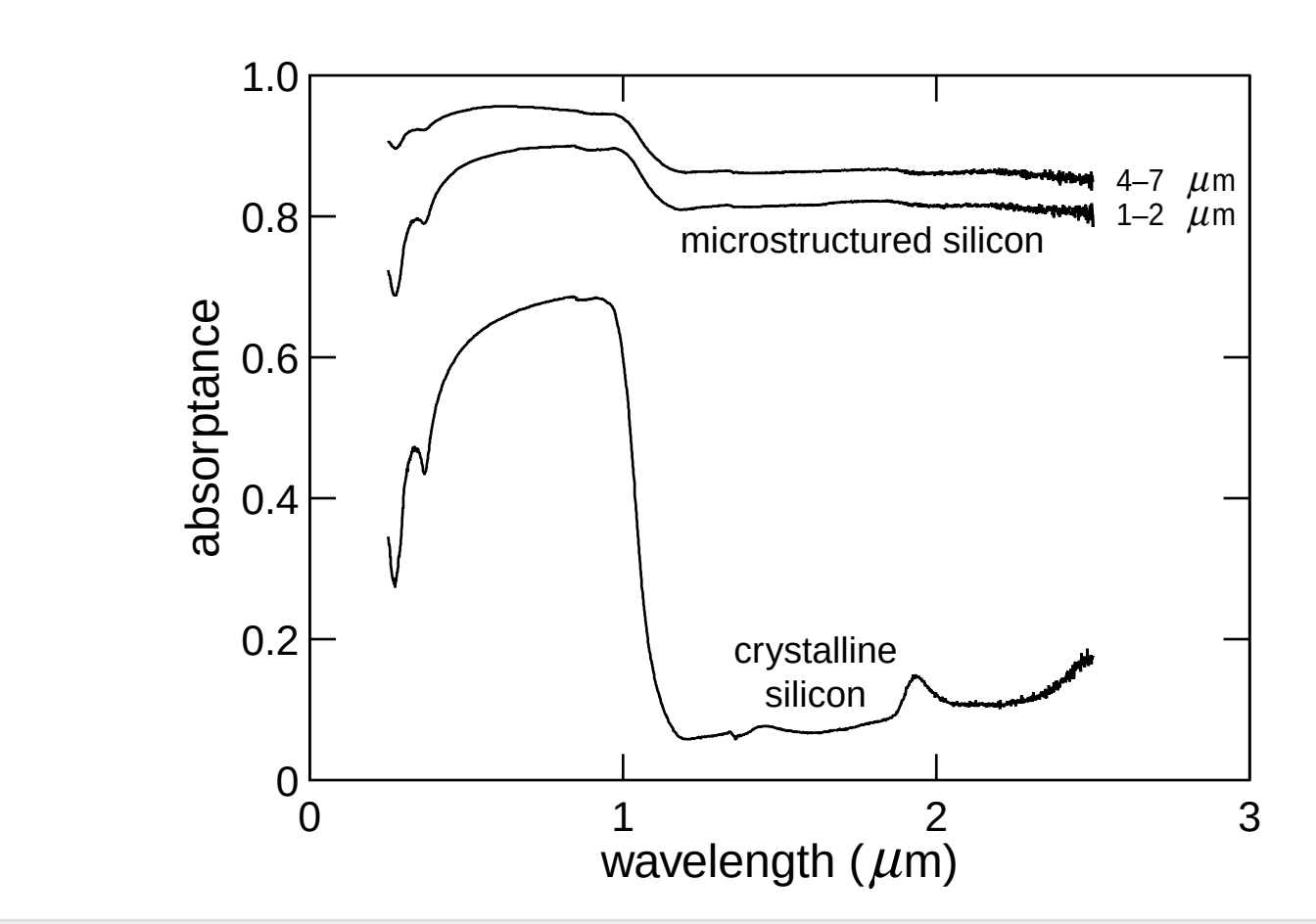
absorptance



C. Wu, *et al.*, *Appl. Phys. Lett*. 78, 1850 (2001)

Background

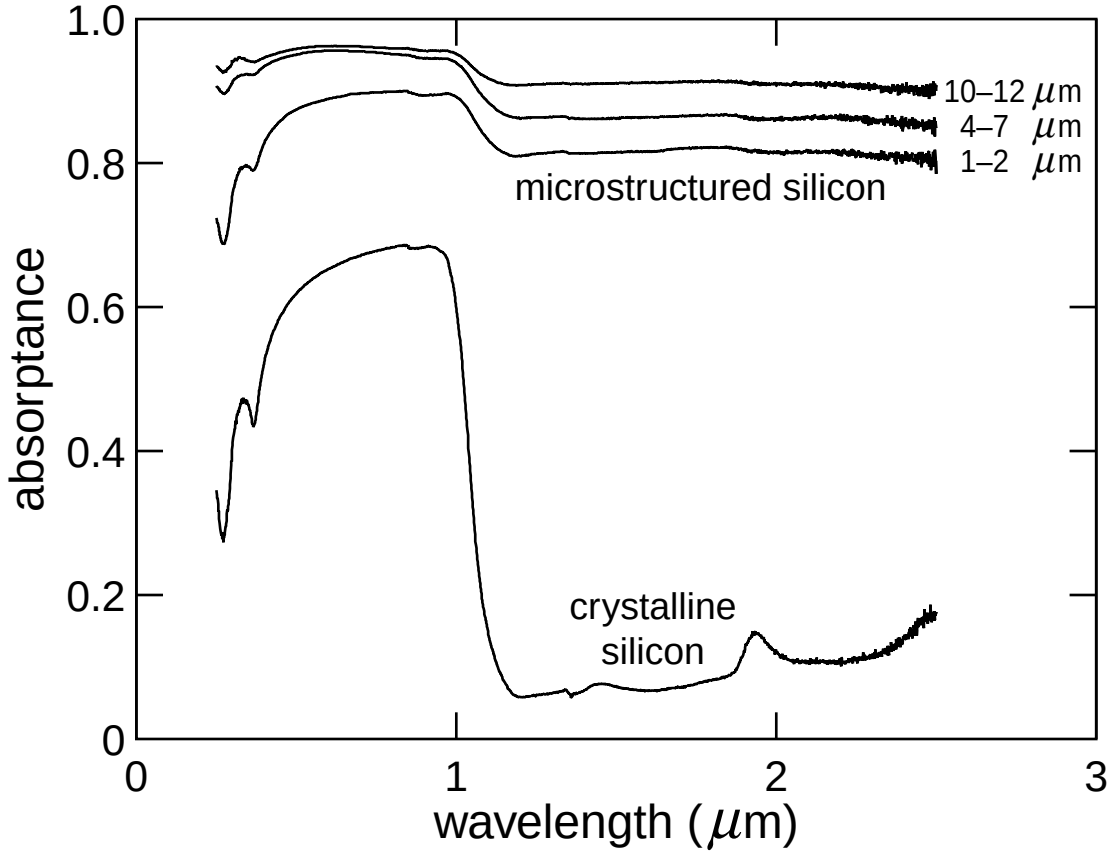
absorptance



C. Wu, *et al.*, *Appl. Phys. Lett.* **78**, 1850 (2001)

Background

absorptance



C. Wu, *et al.*, *Appl. Phys. Lett*. 78, 1850 (2001)

Background

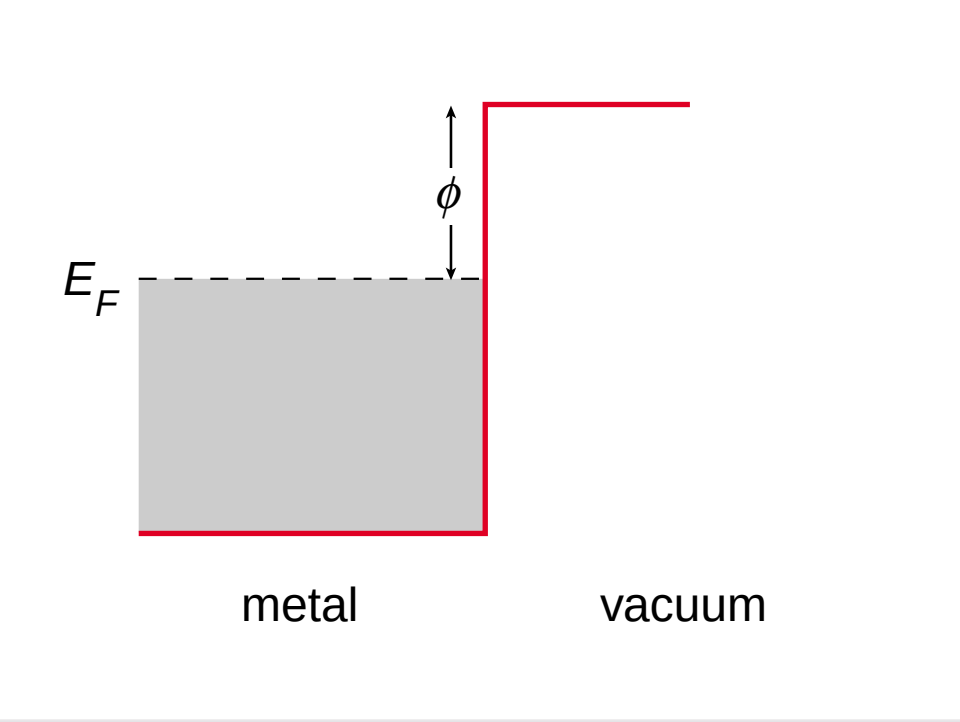
Points to keep in mind:

- **one-step, maskless process**
- **large area with uniform high density of spikes**
- **band structure change**

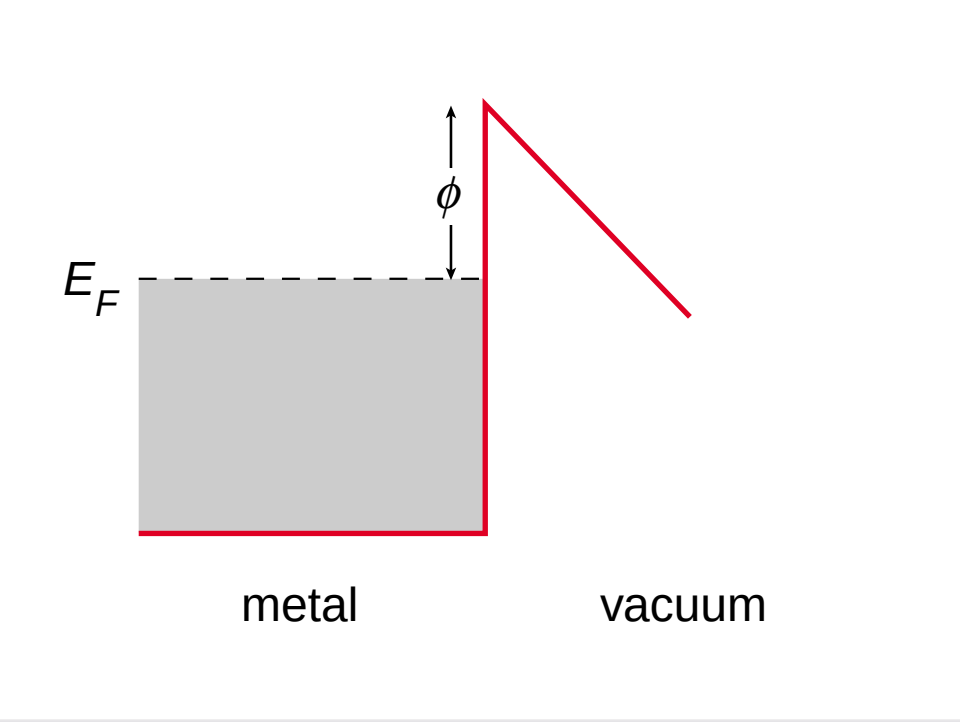
Outline

- Background
- **Results**
- Discussion

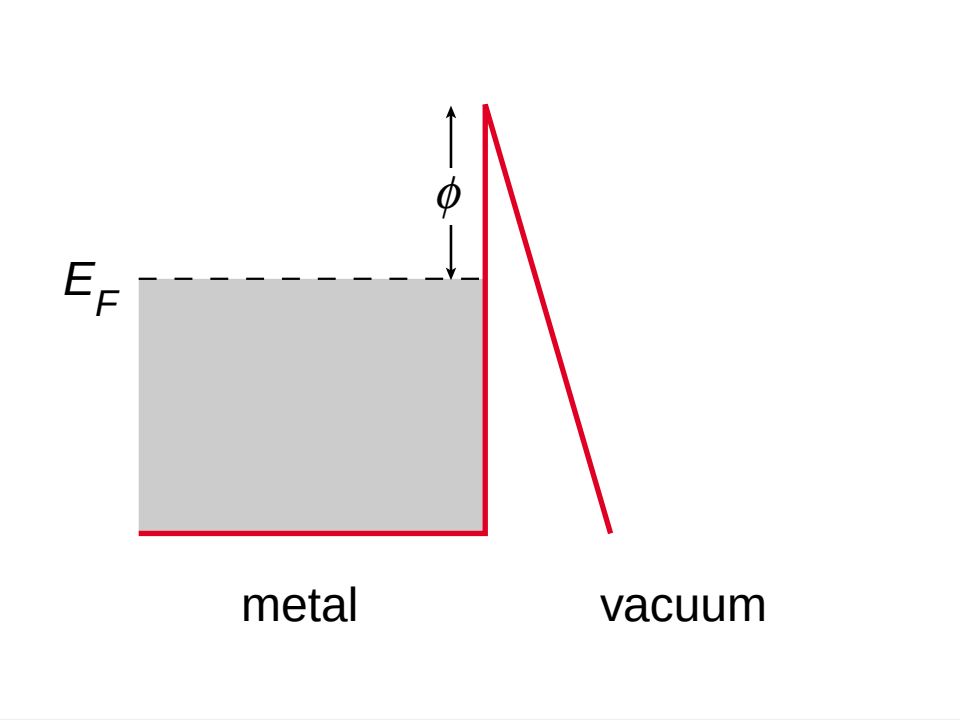
Field emission



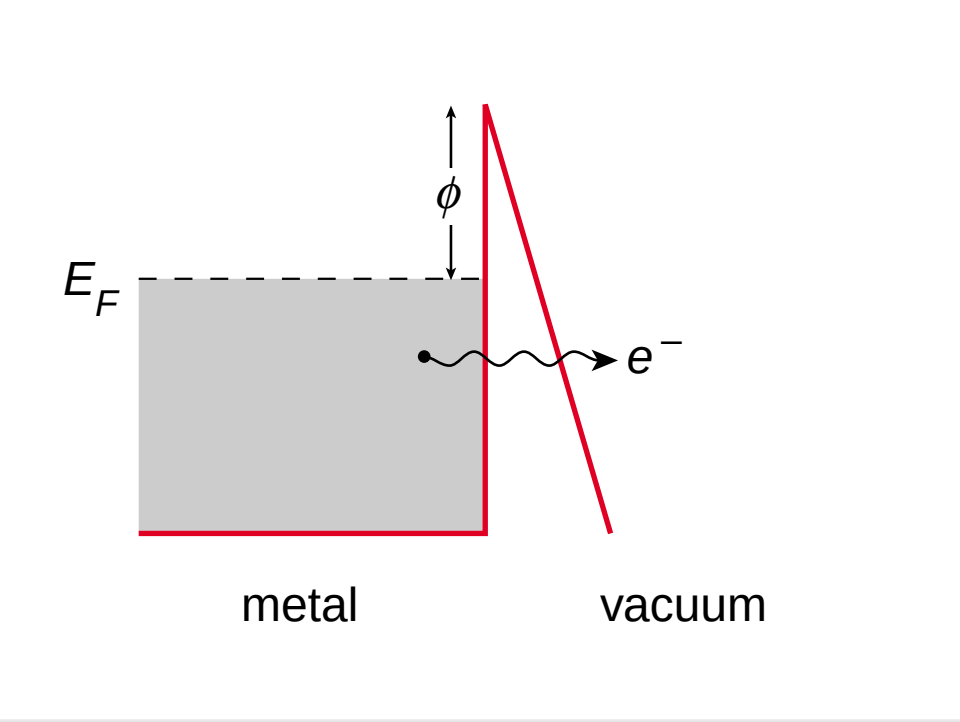
Field emission



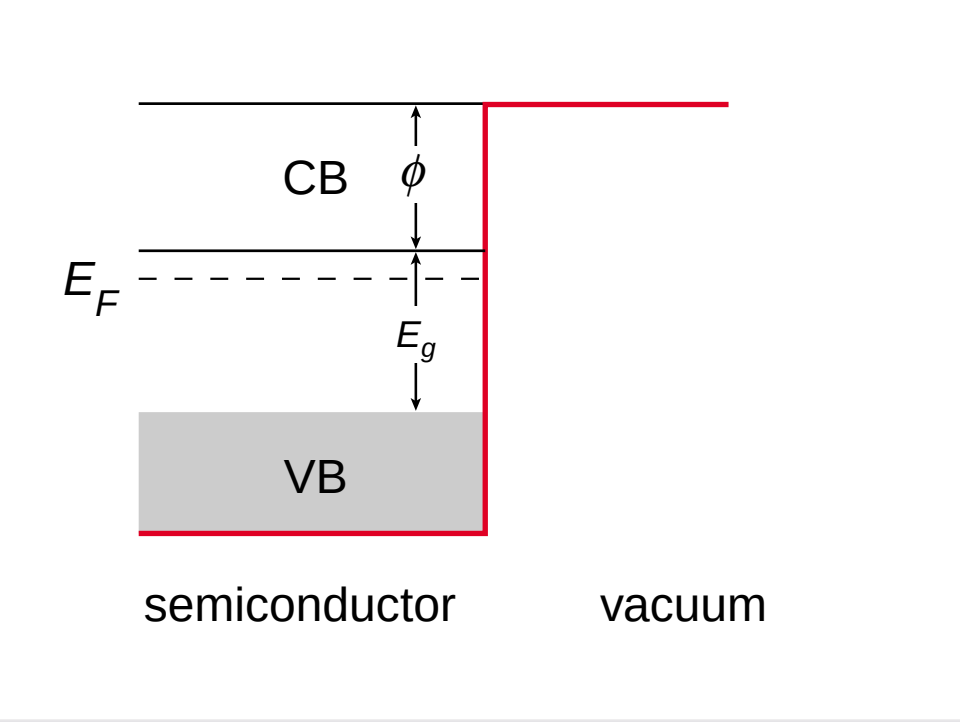
Field emission



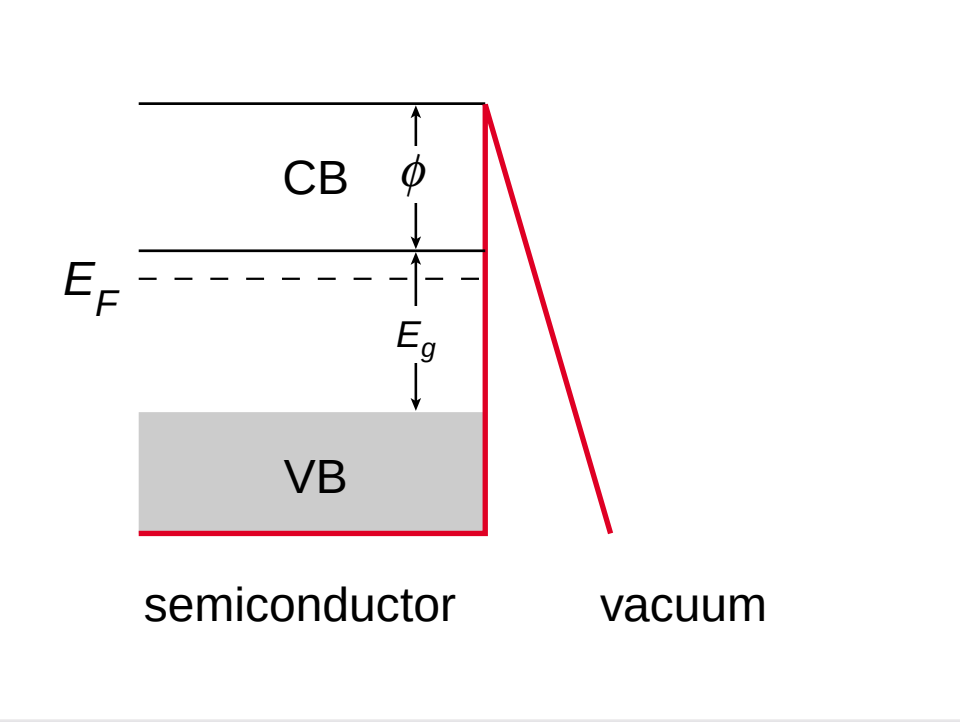
Field emission



Field emission



Field emission



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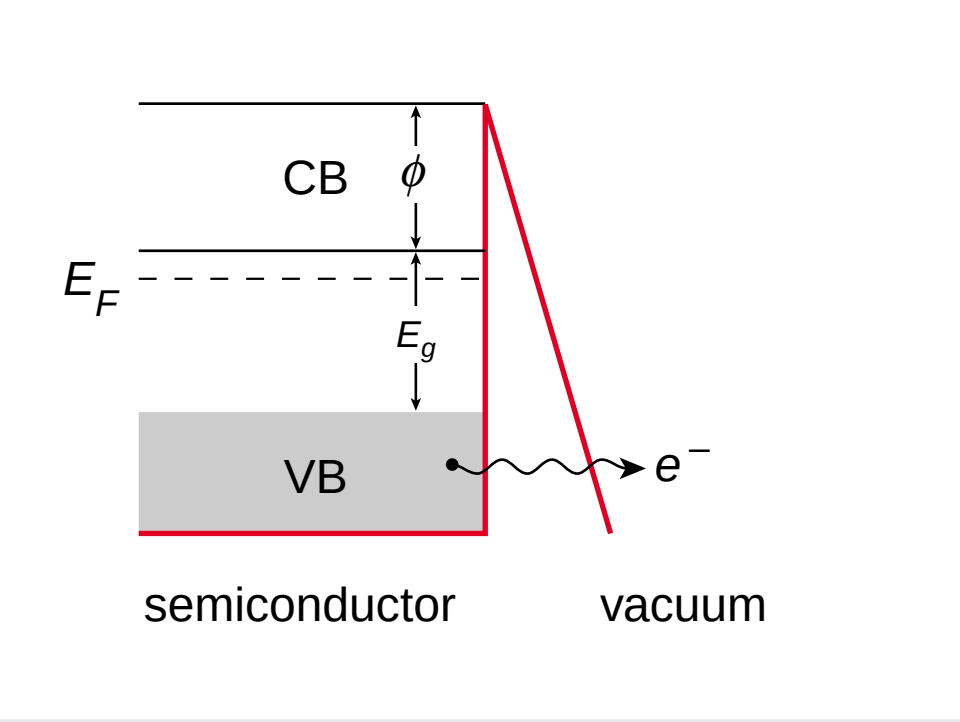
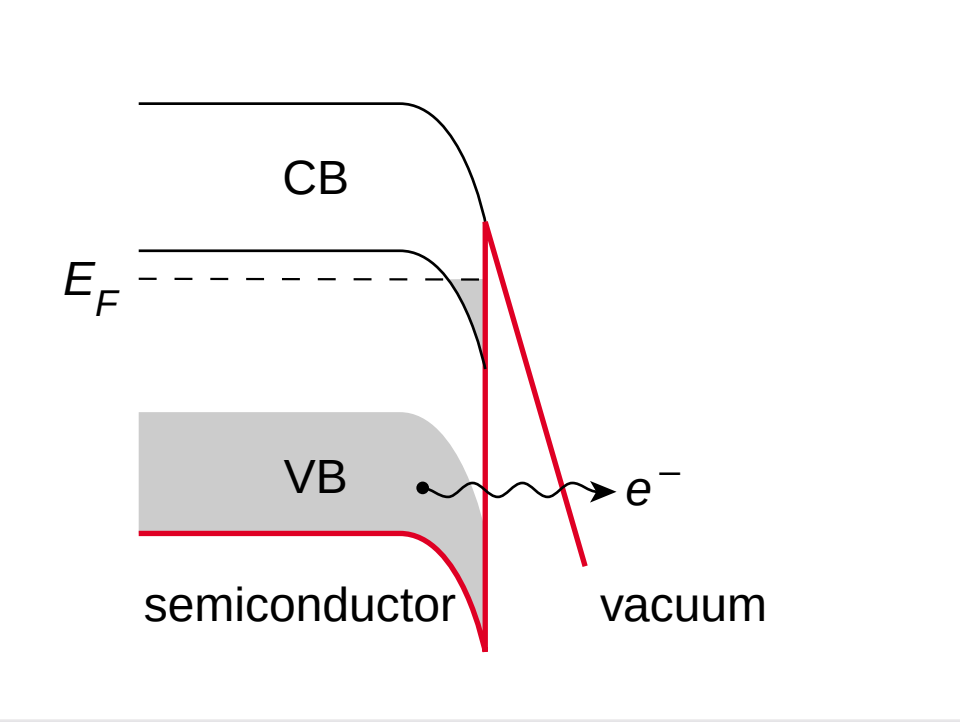
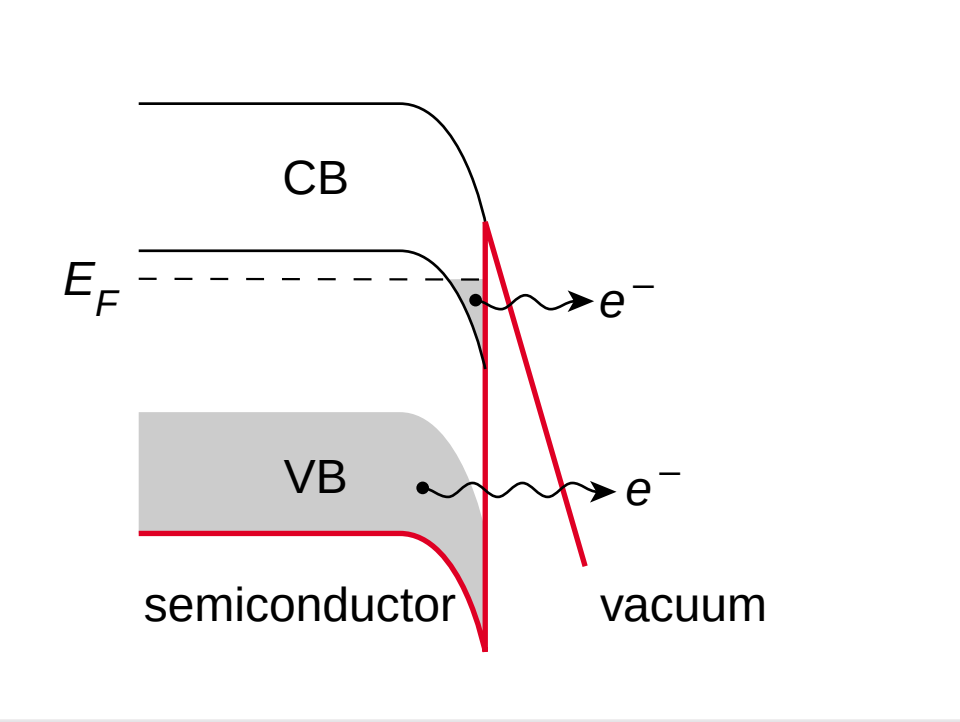


Field emission

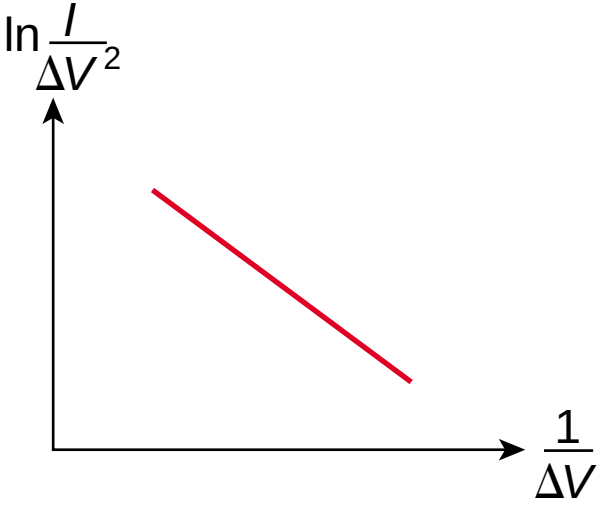


Field emission



Field emission

$$\ln \frac{I}{\Delta V^2} = \ln a - b\frac{1}{\Delta V}$$



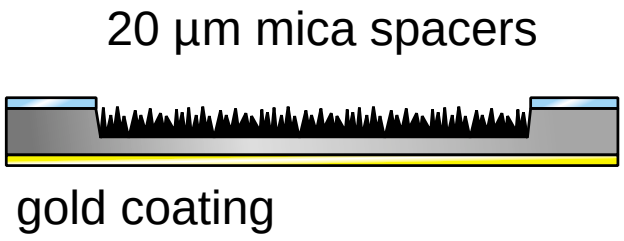
R.H. Fowler and L. Nordheim, *Proc. R. Soc. Lond. A* (1928)

Setup

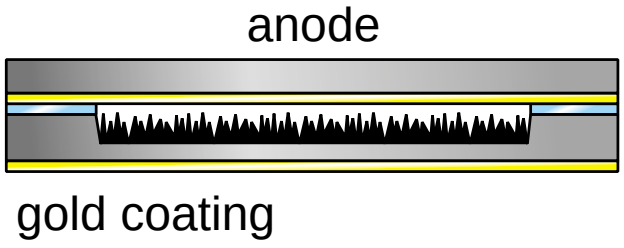
Setup

gold coating

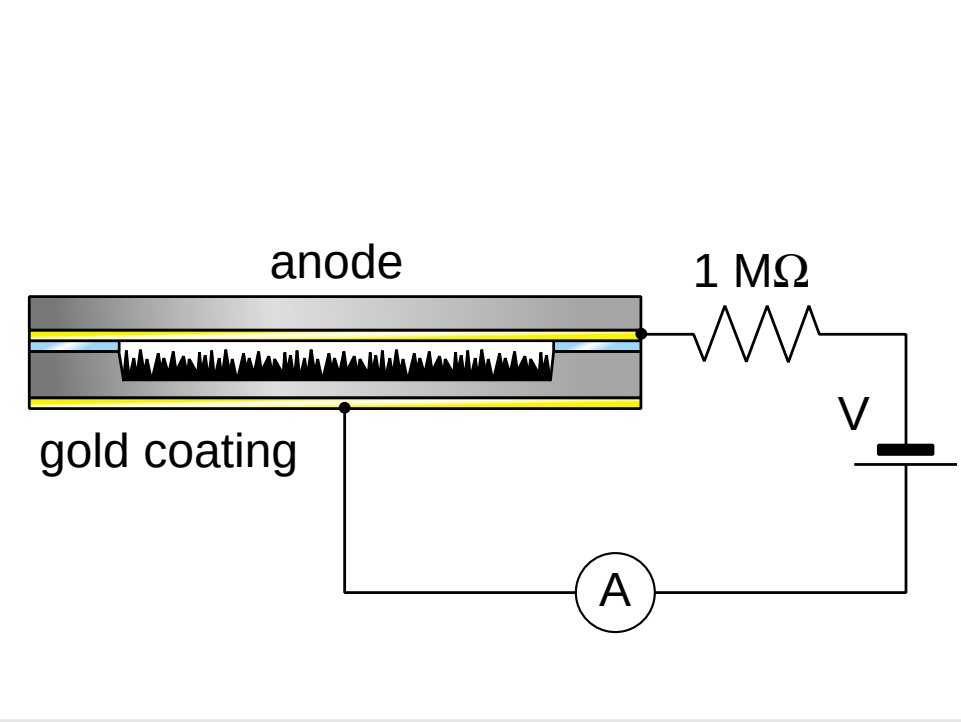
Setup



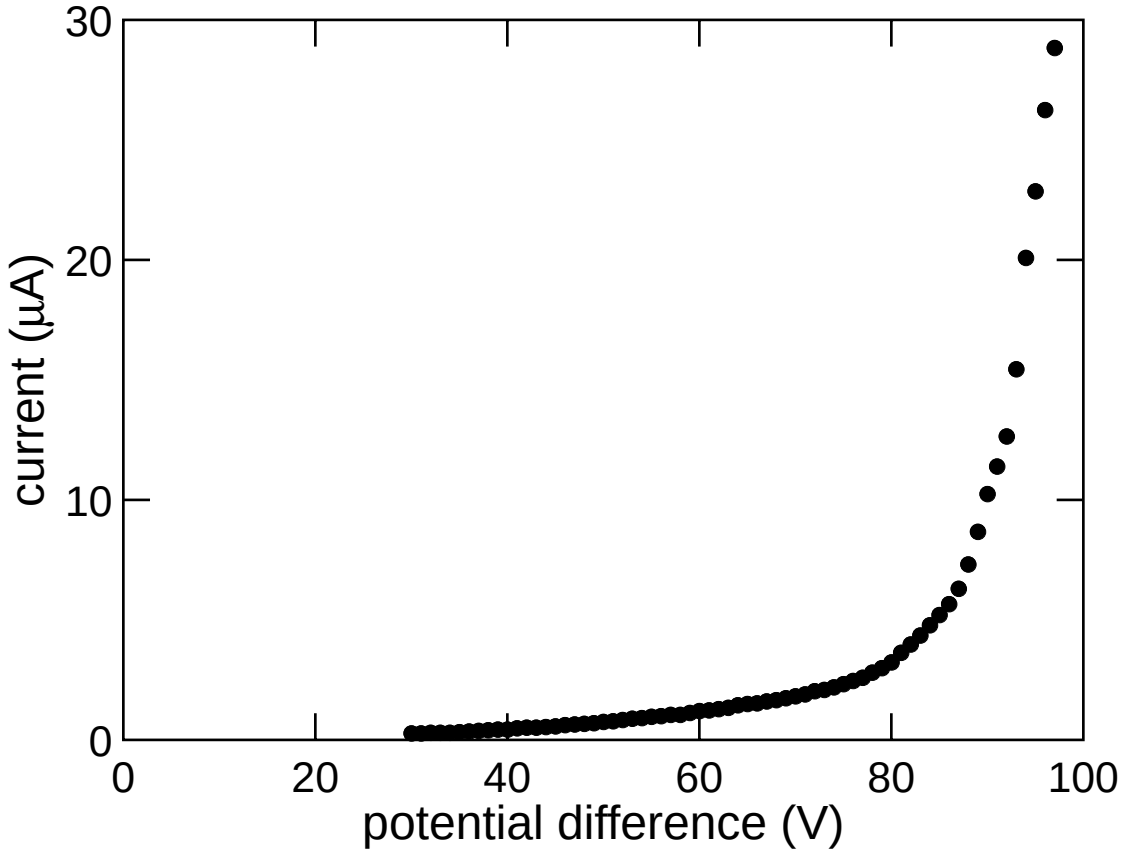
Setup



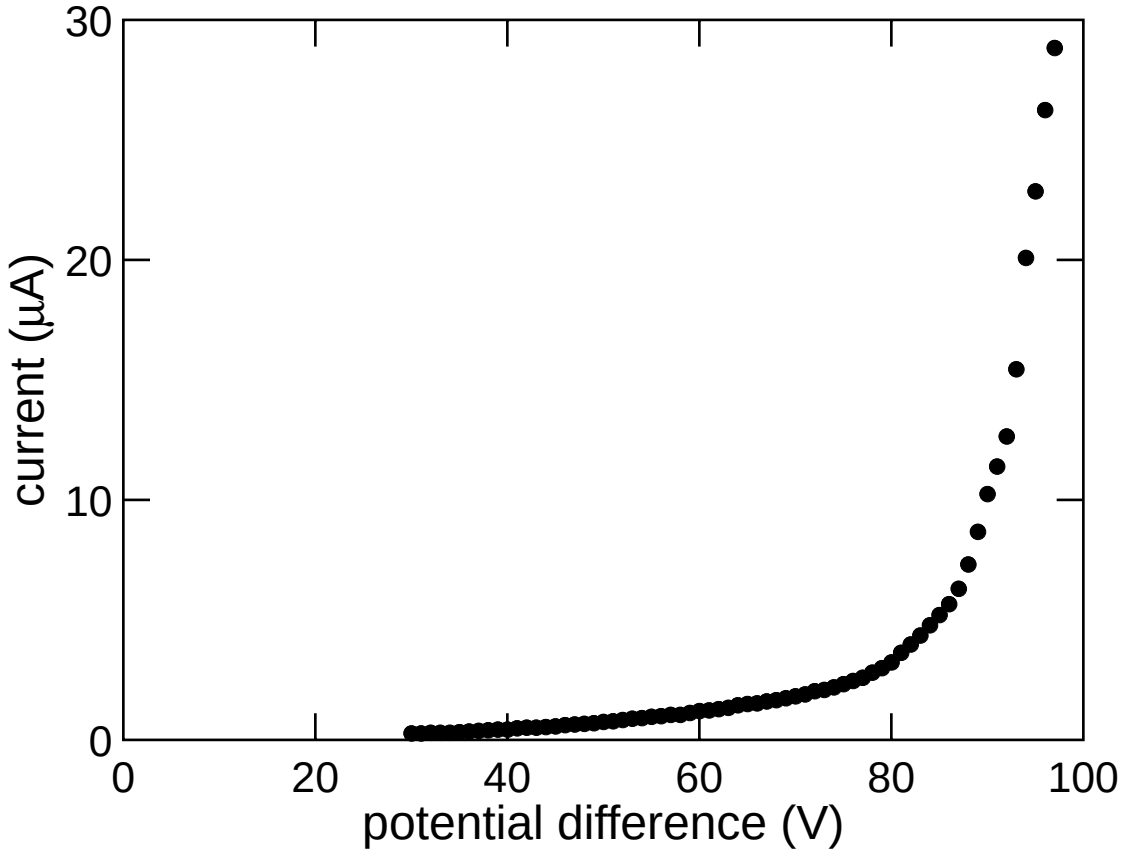
Setup



Results

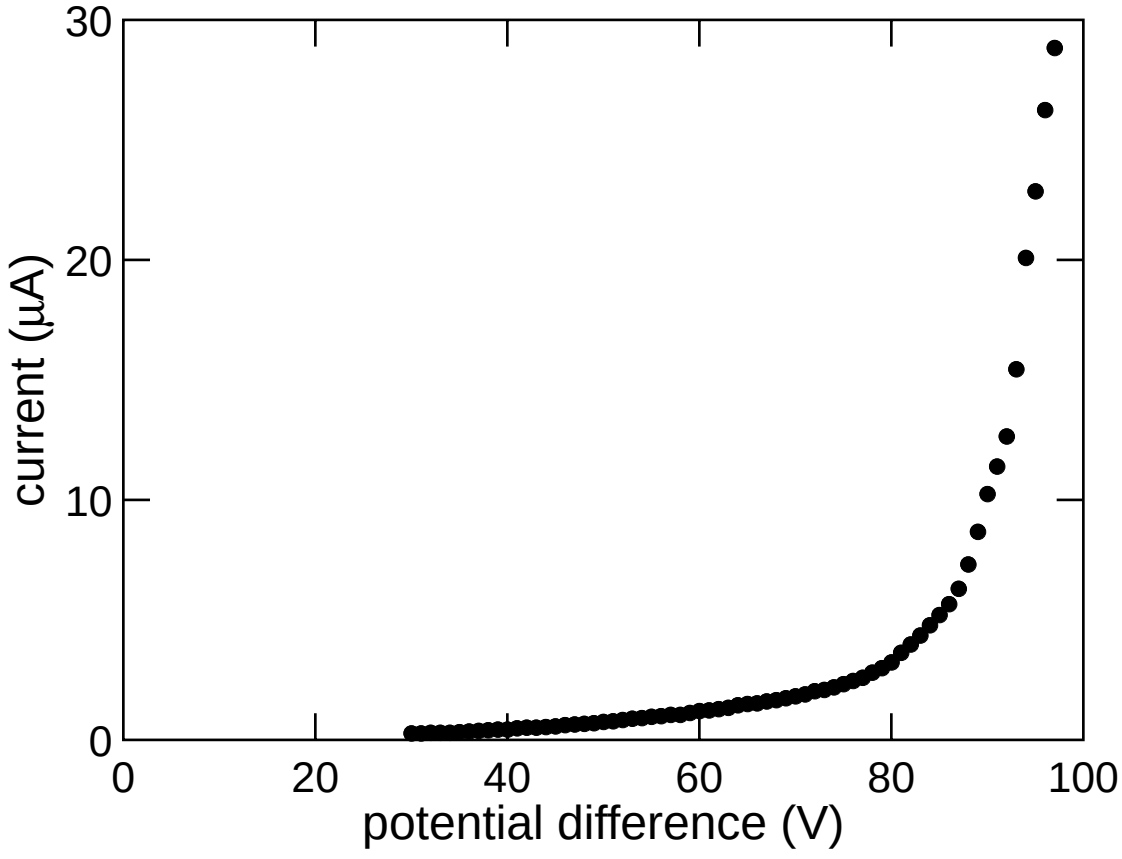


Results



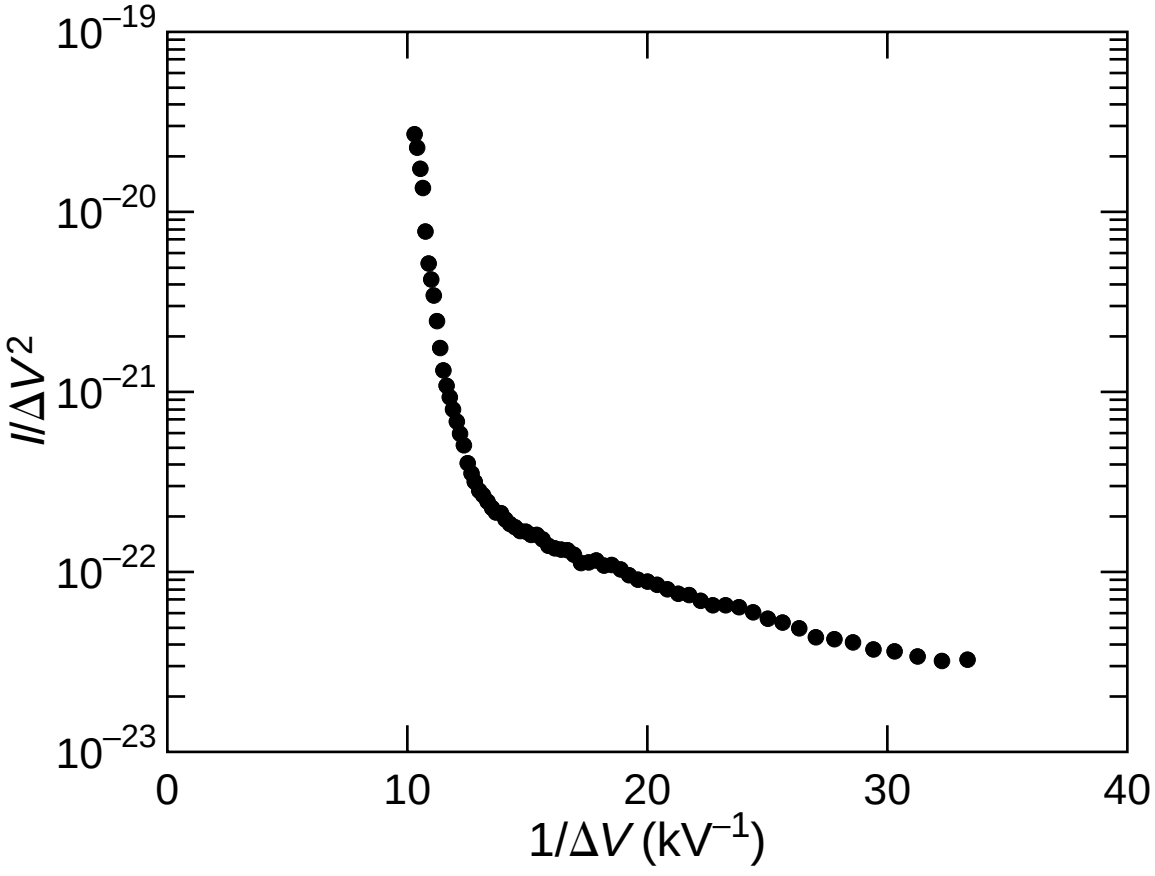
turn-on field (1 μA/cm^2): 1.3 V/μm

Results

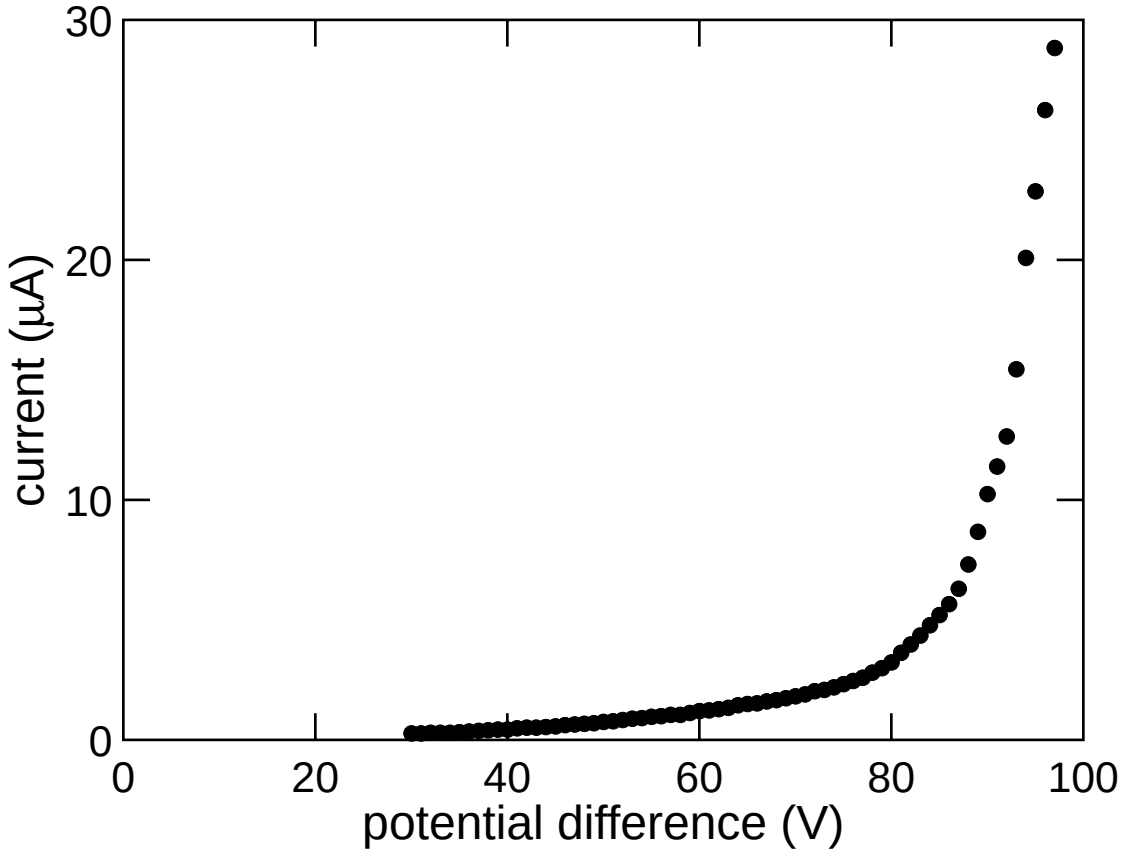


threshold field (10 μA/cm^2): 2.15 V/μm

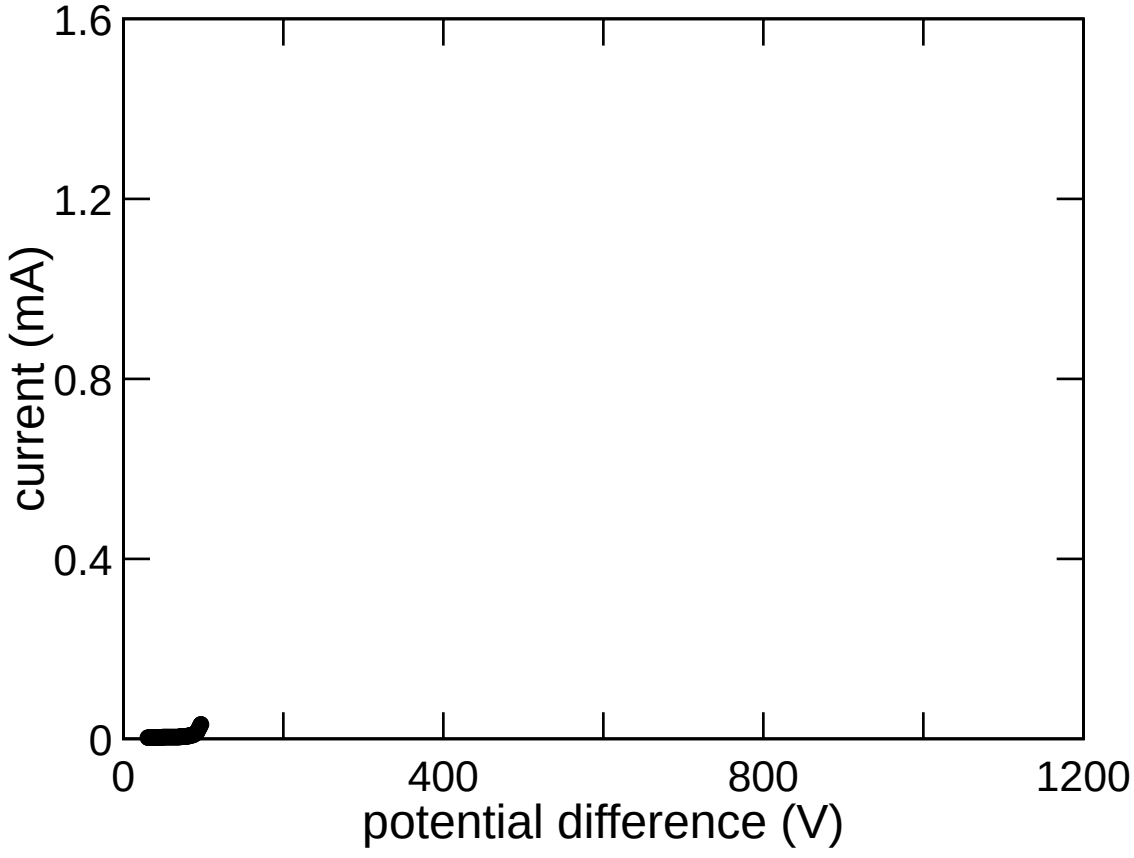
Results



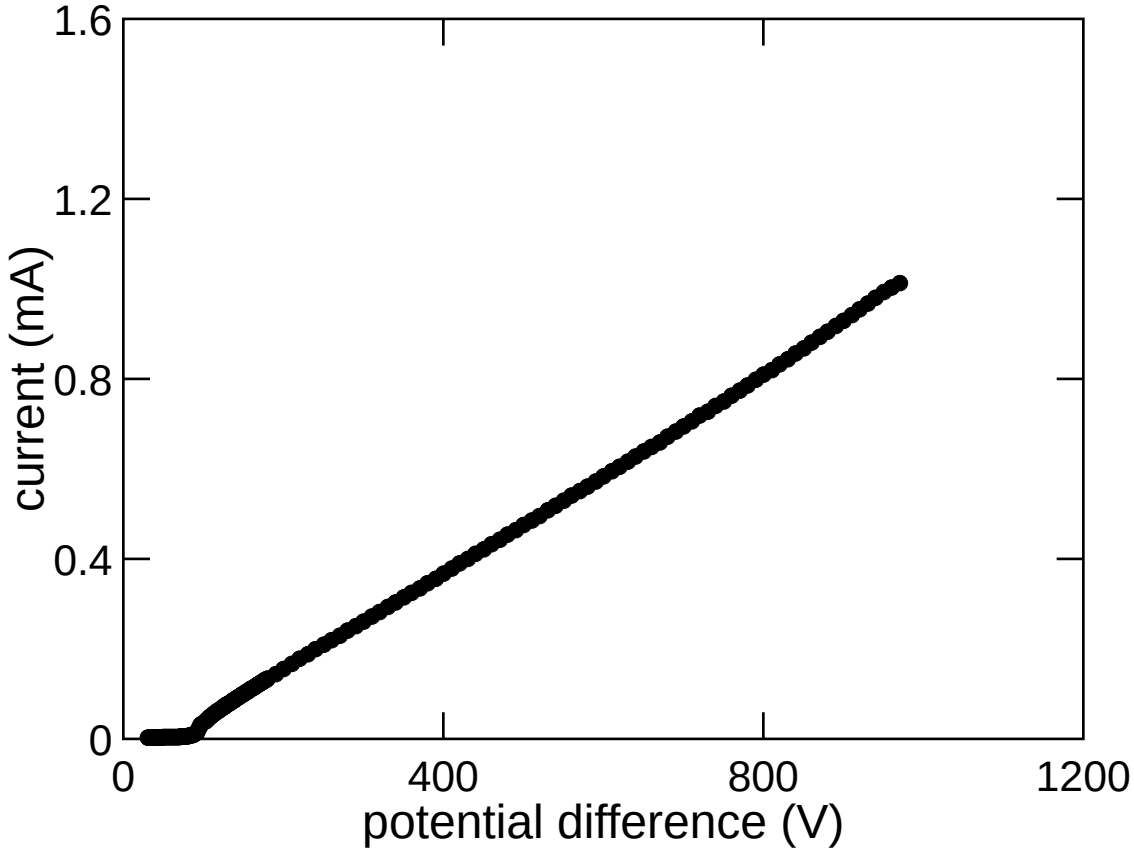
Results



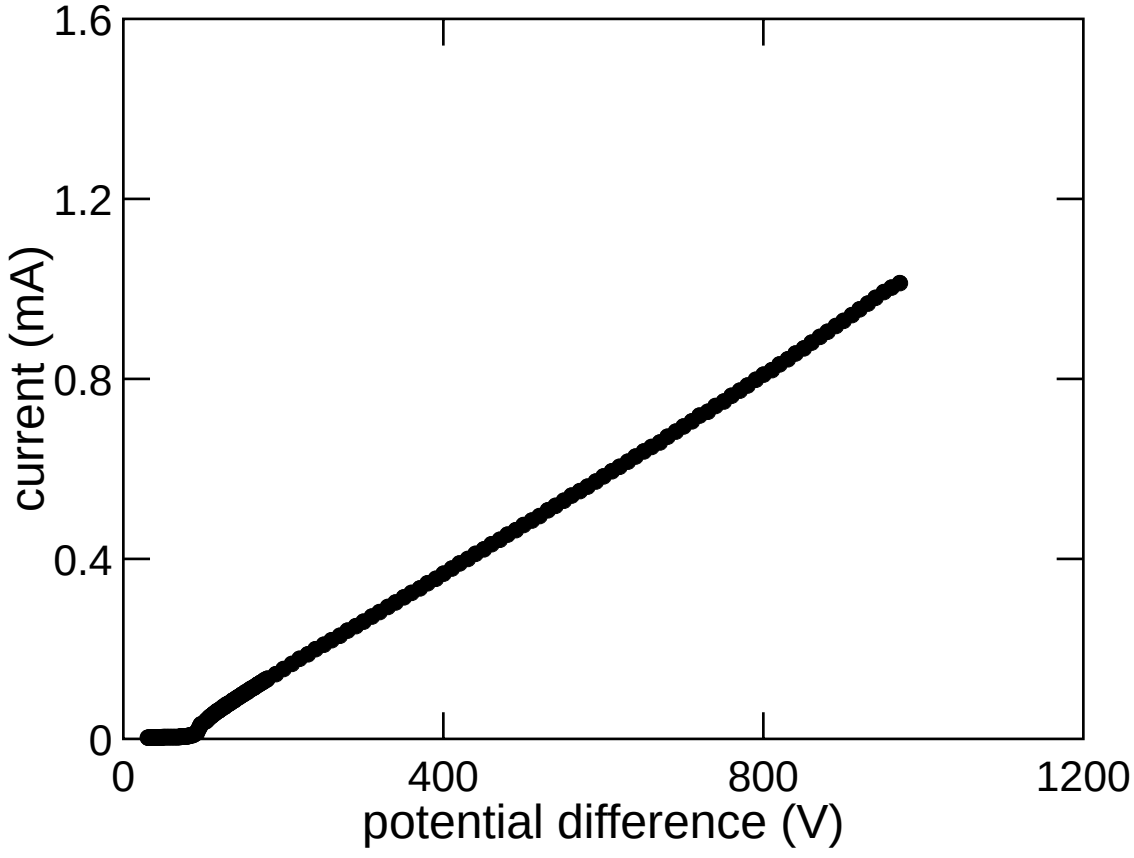
Results



Results

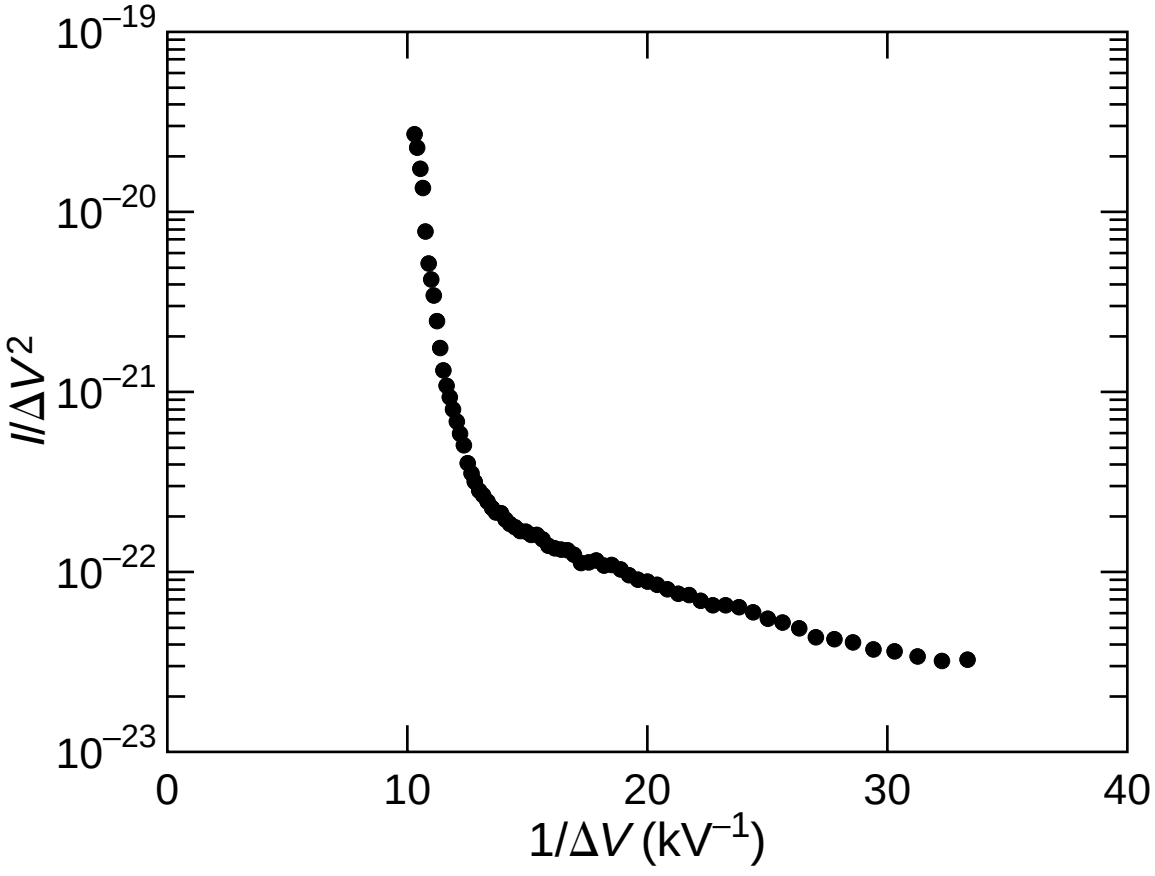


Results

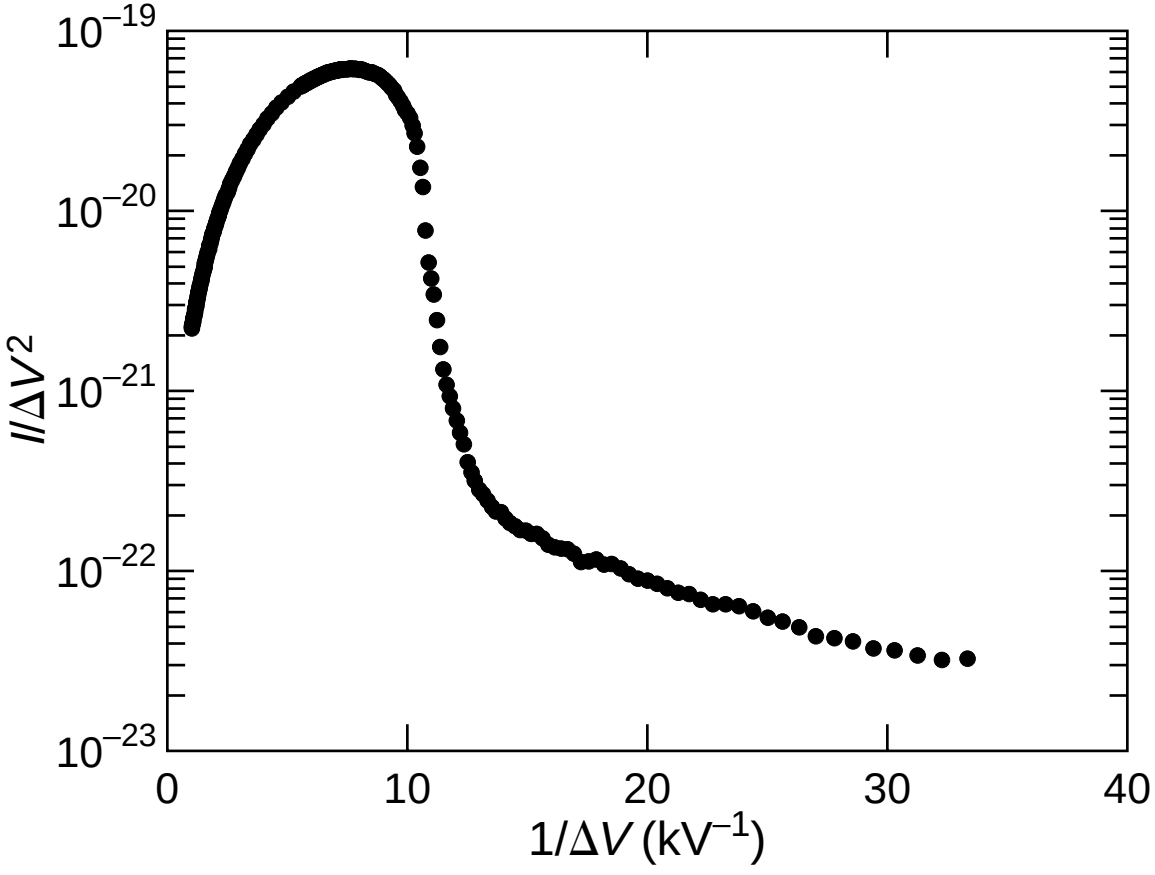


maximum current: 2 mA (4 mm^2 sample)

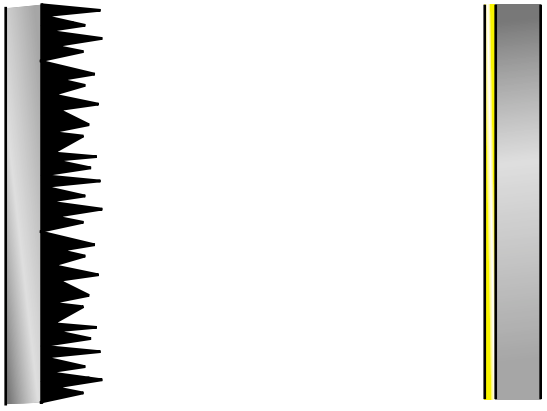
Results



Results

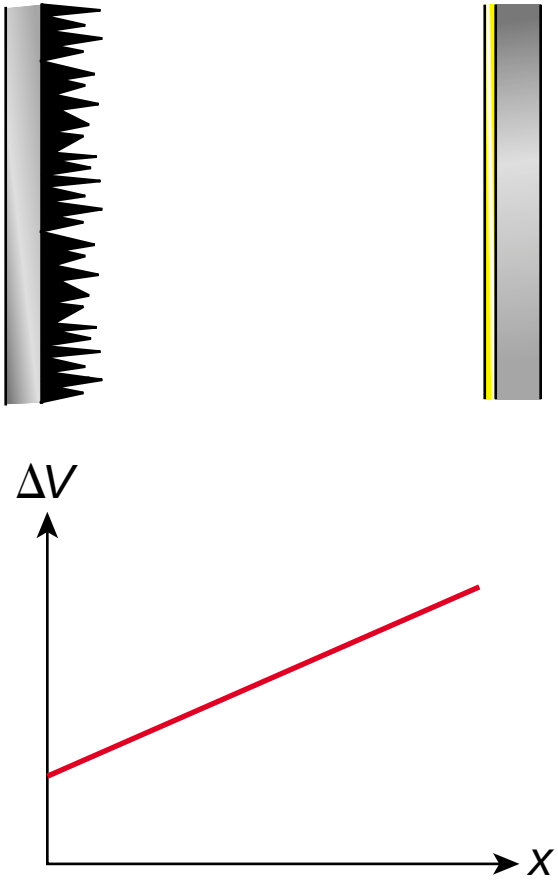


Space charge effect



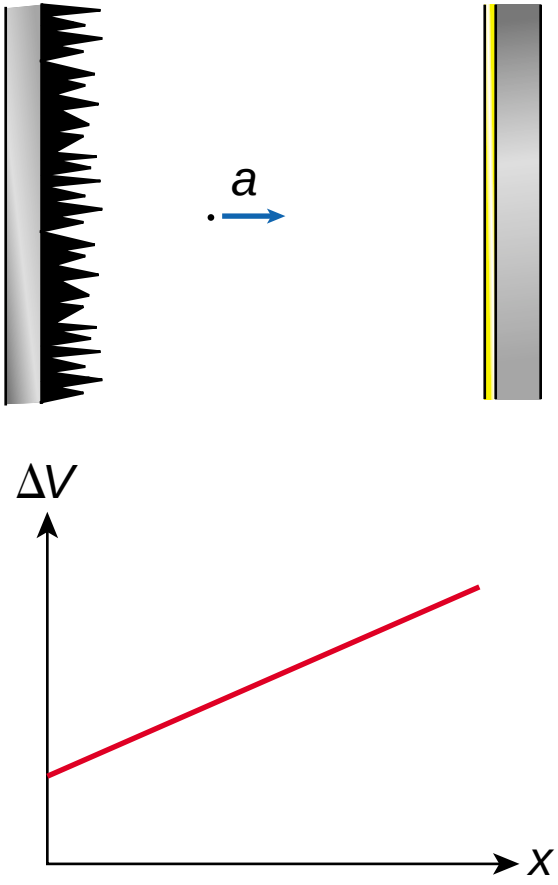
Y.Y. Lau *et al*., *Phys. Plasmas* 1, 2082 (1994)

Space charge effect



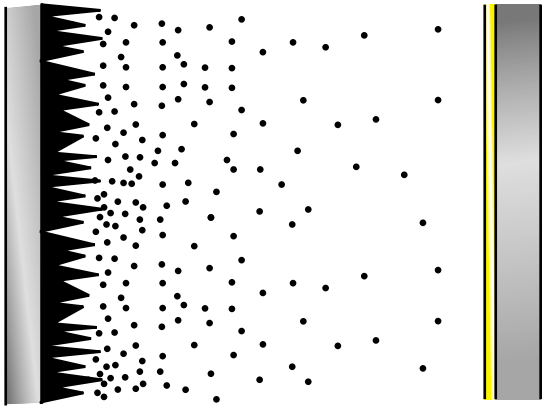
Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

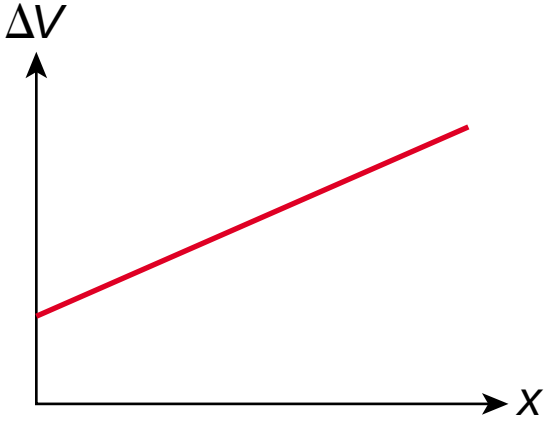
Space charge effect



Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

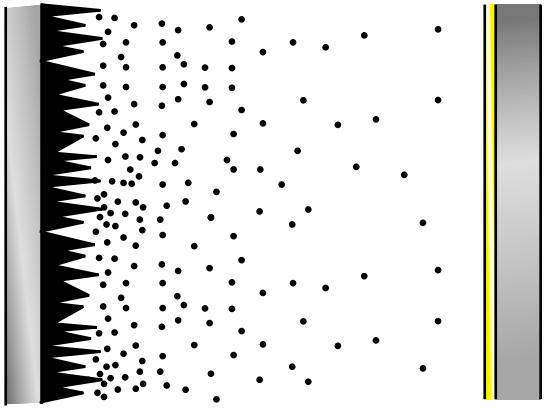
Space charge effect

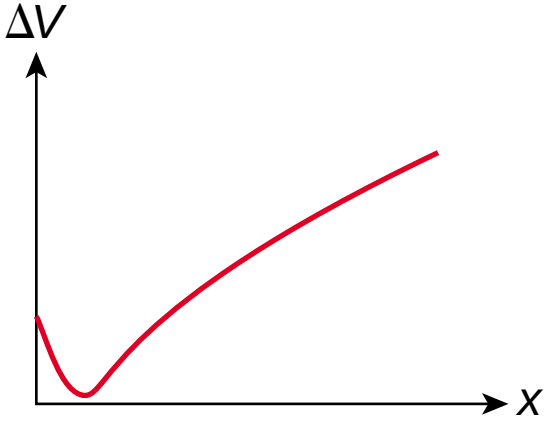




Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

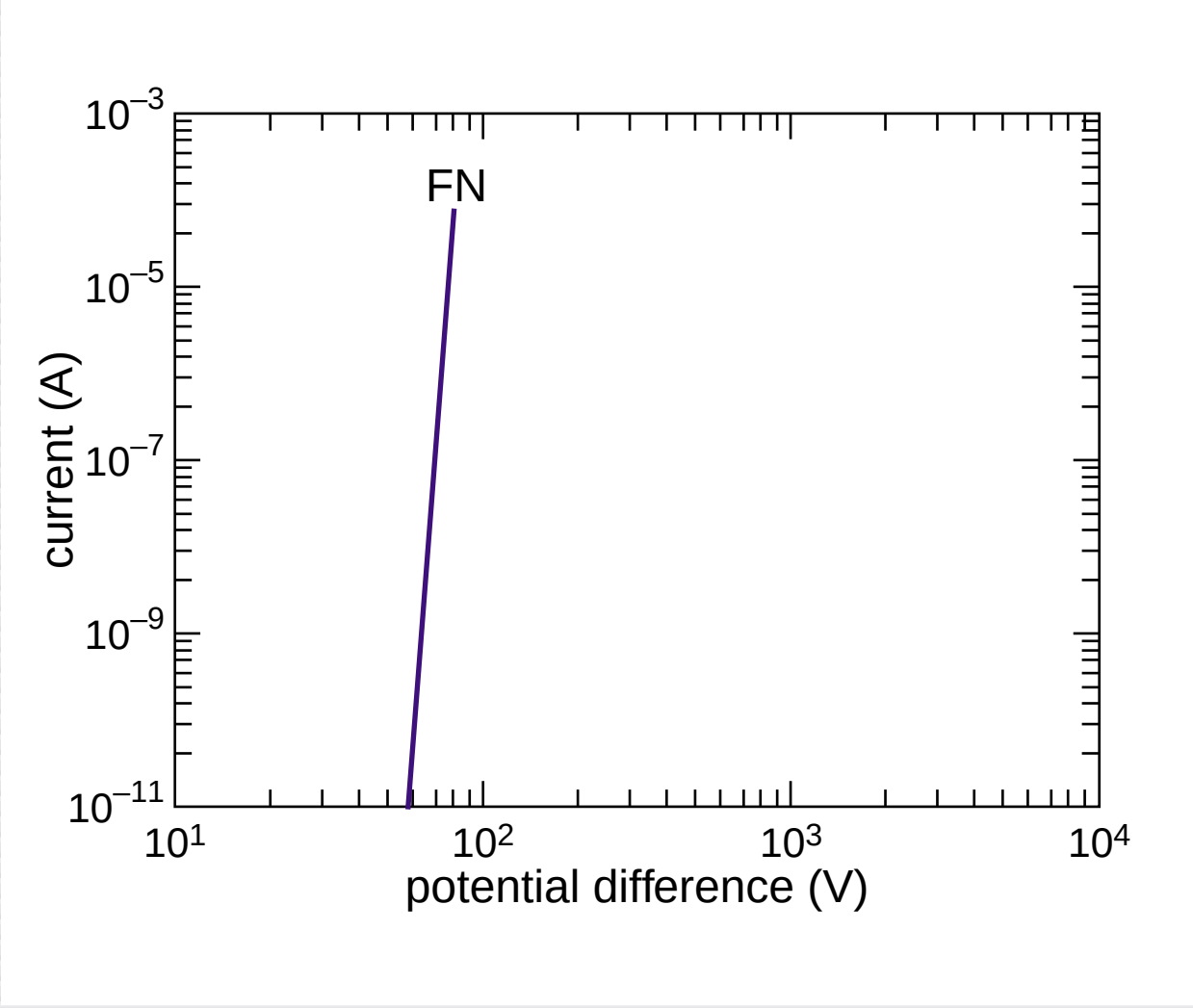
Space charge effect





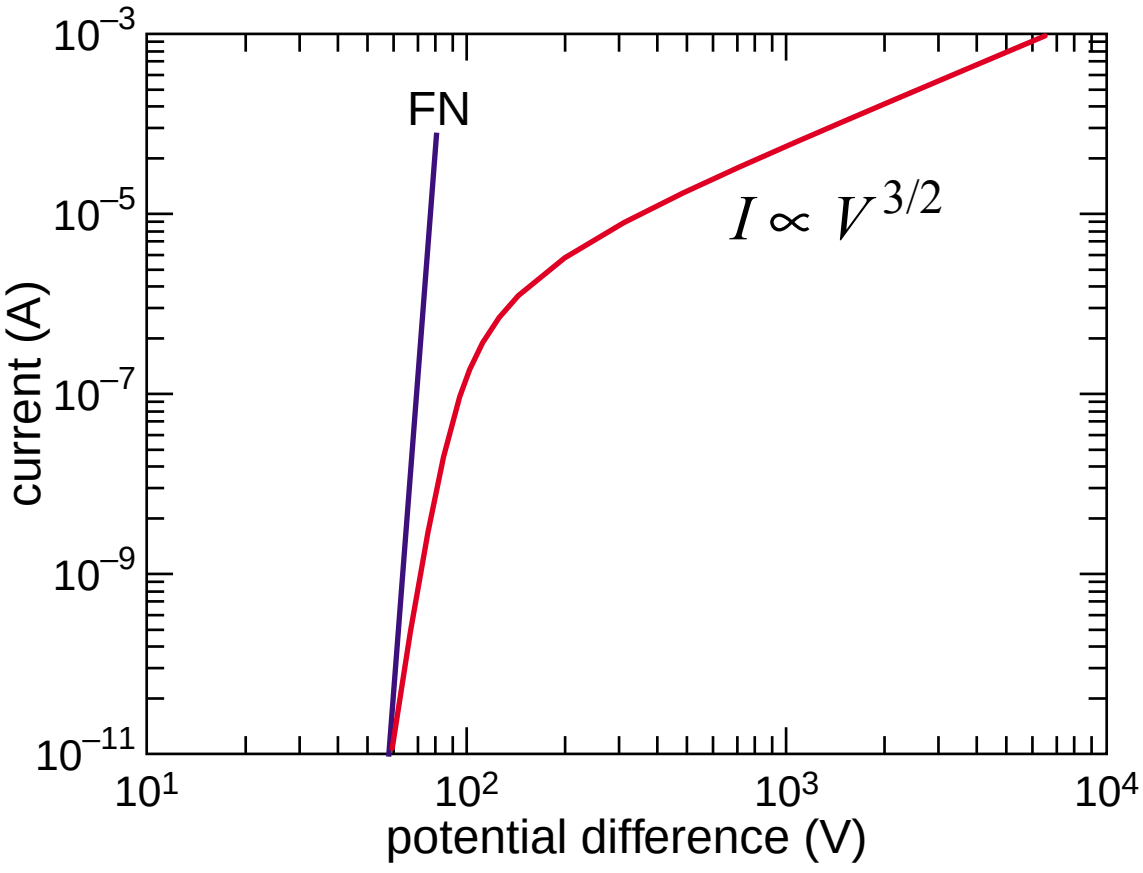
Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

Space charge effect



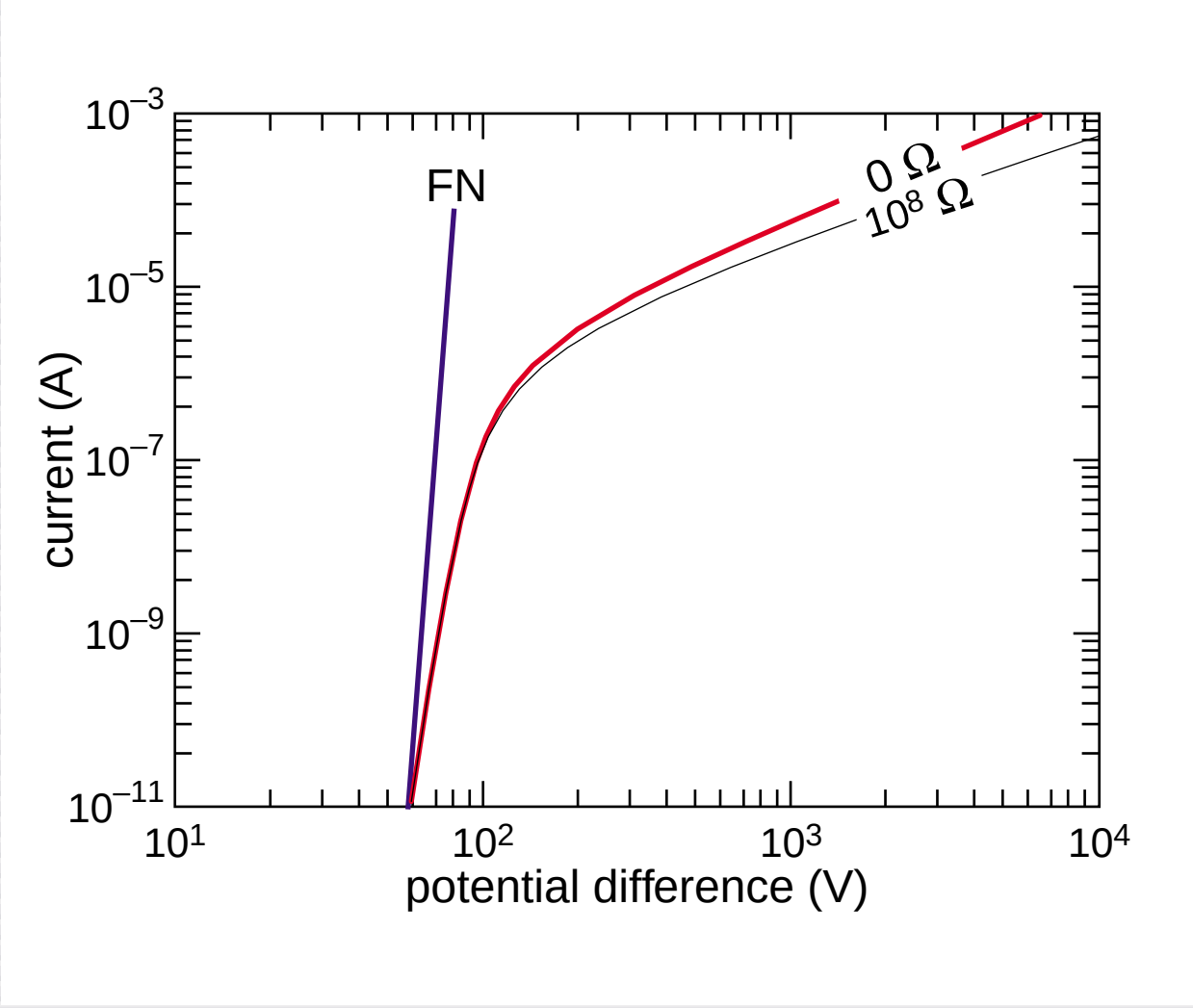
Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

Space charge effect



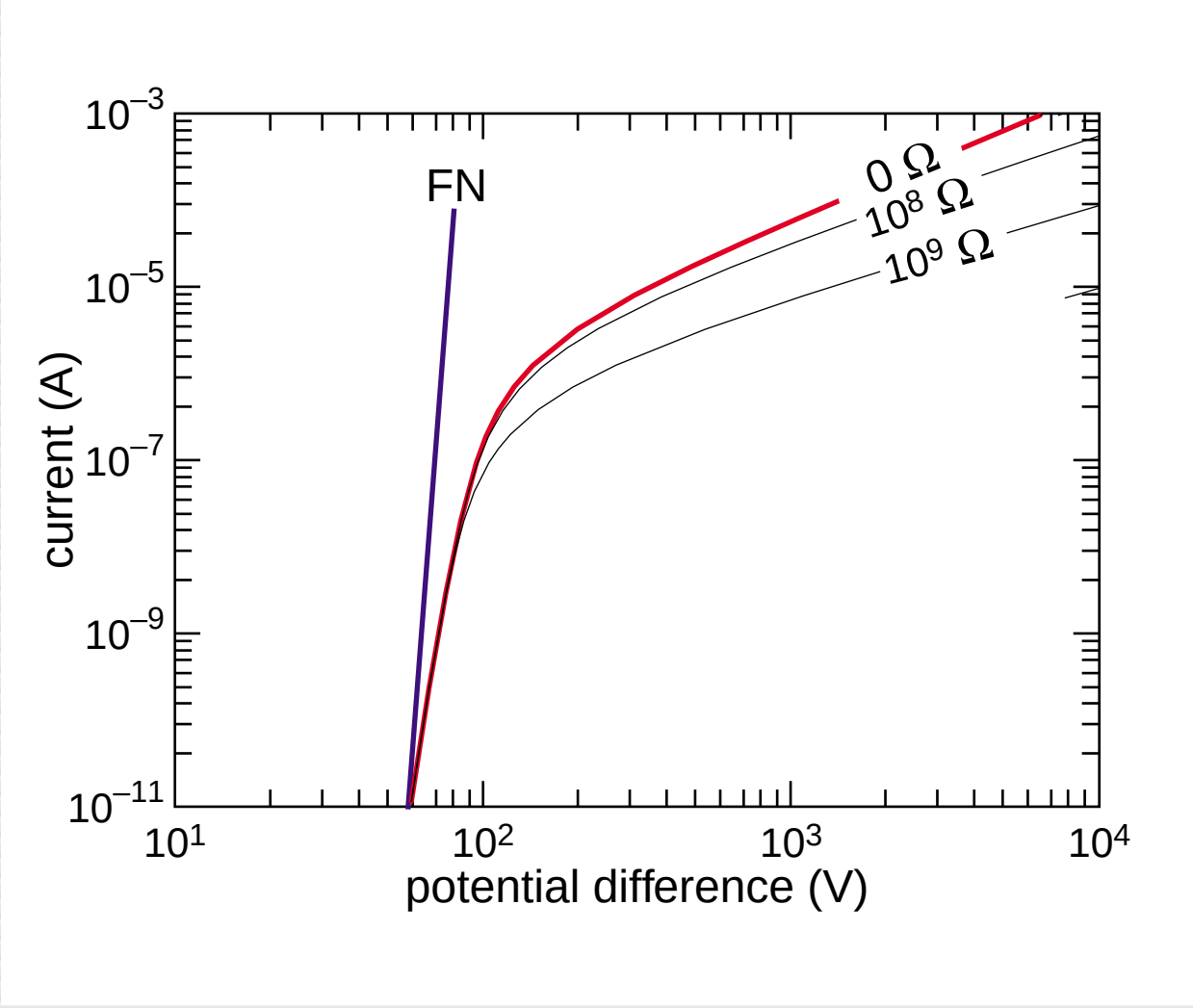
Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

Space charge effect



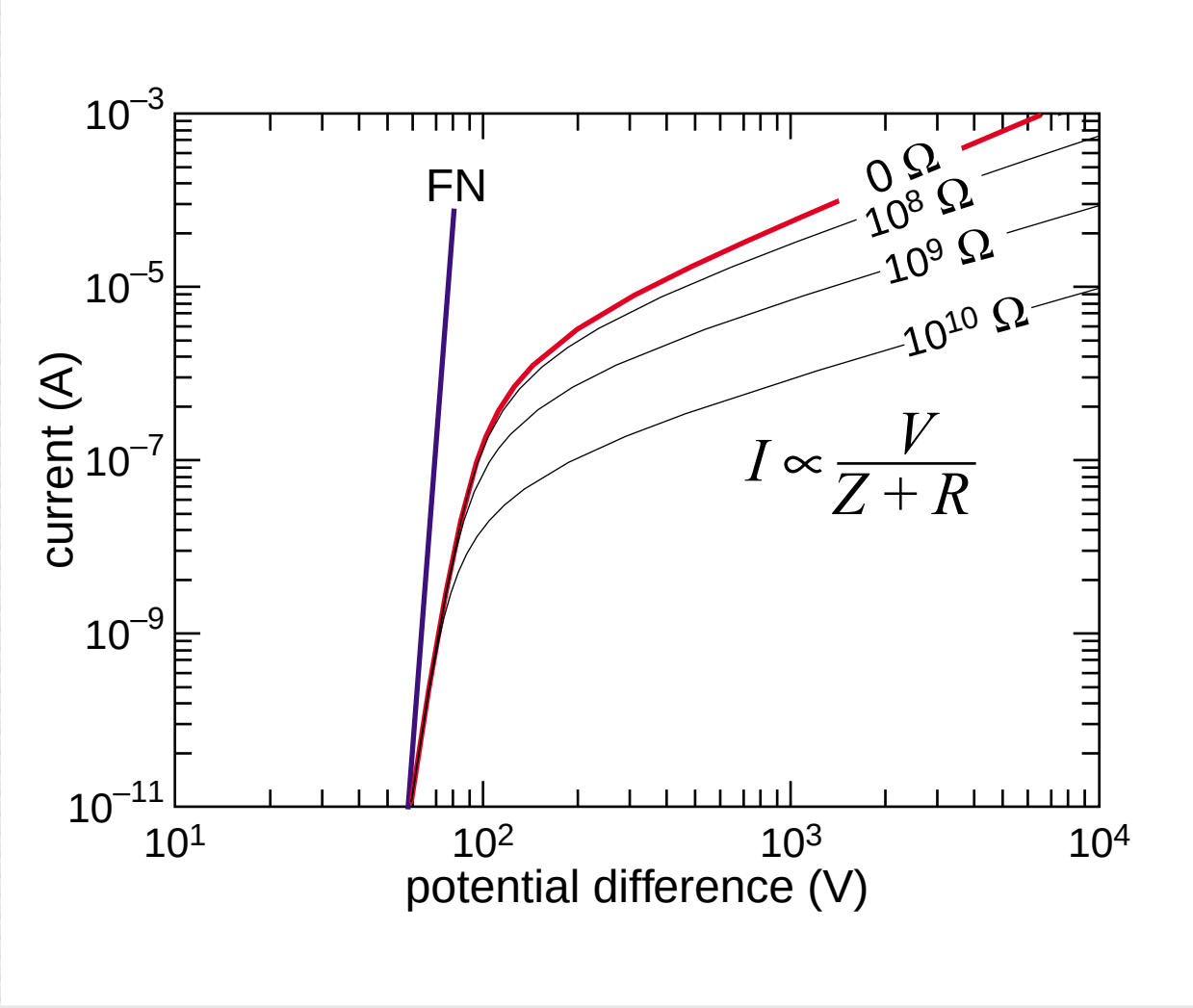
Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

Space charge effect



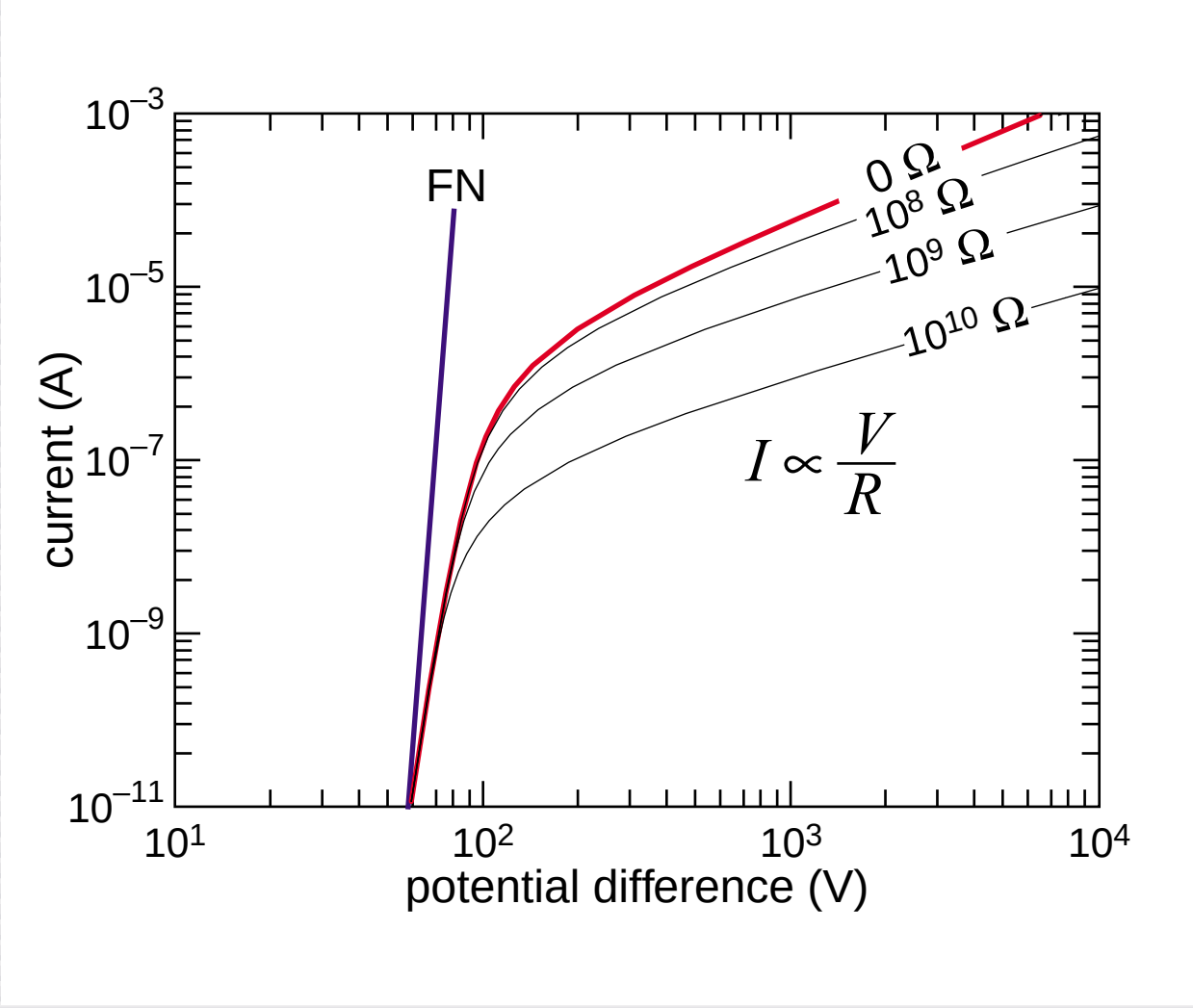
Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

Space charge effect



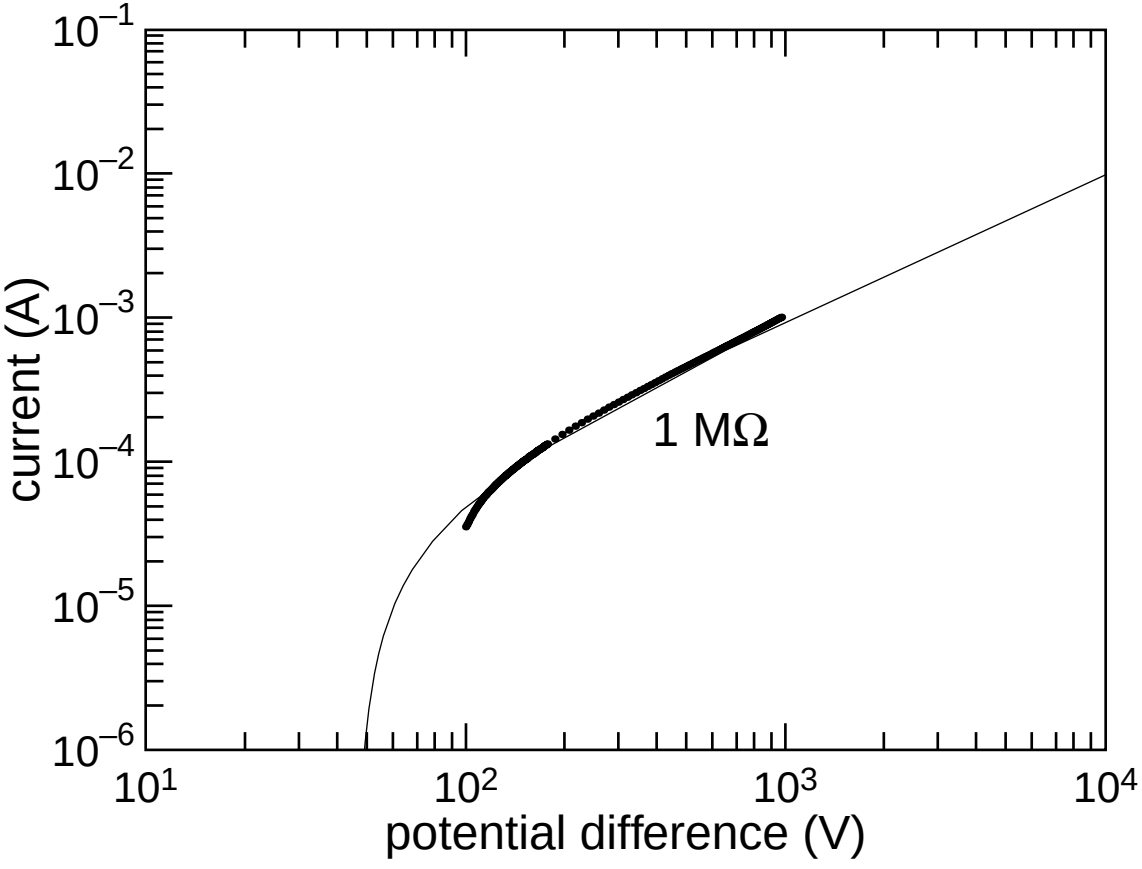
Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

Space charge effect

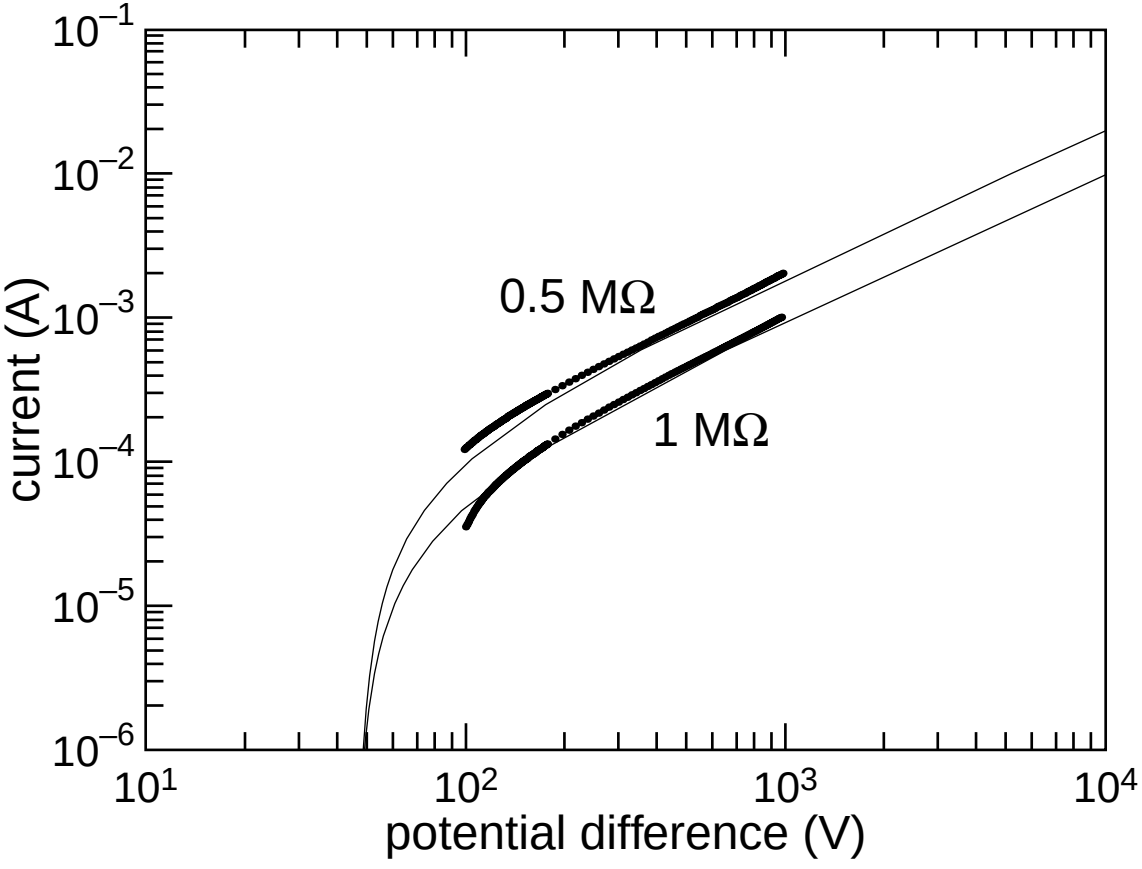


Y.Y. Lau *et al.*, *Phys. Plasmas* 1, 2082 (1994)

Space charge effect



Space charge effect



Outline

- Background
- Results
- **Discussion**

Discussion

Ion channeling and electron backscattering

- **spikes retain crystalline order**
- **high density of defects**

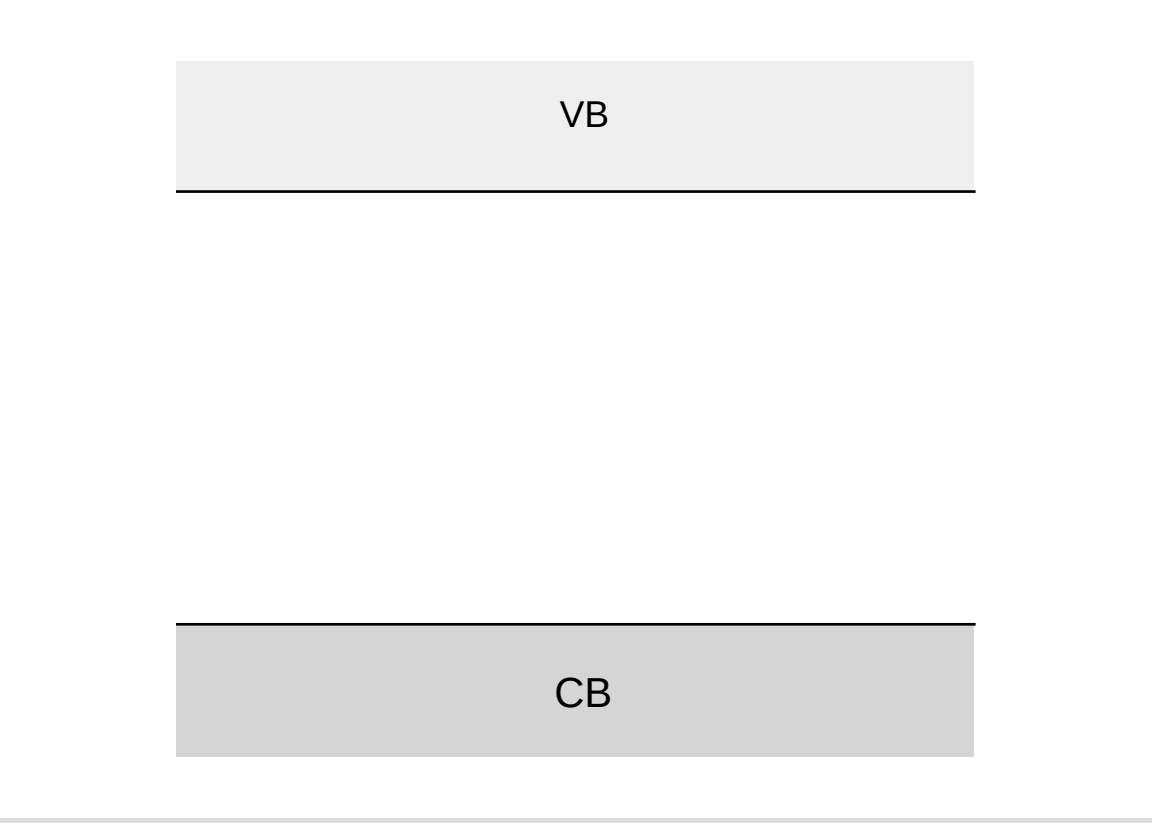
Discussion

Secondary ion mass spectrometry:

- 10^{20} cm^{-3} sulfur
- 10^{17} cm^{-3} fluorine

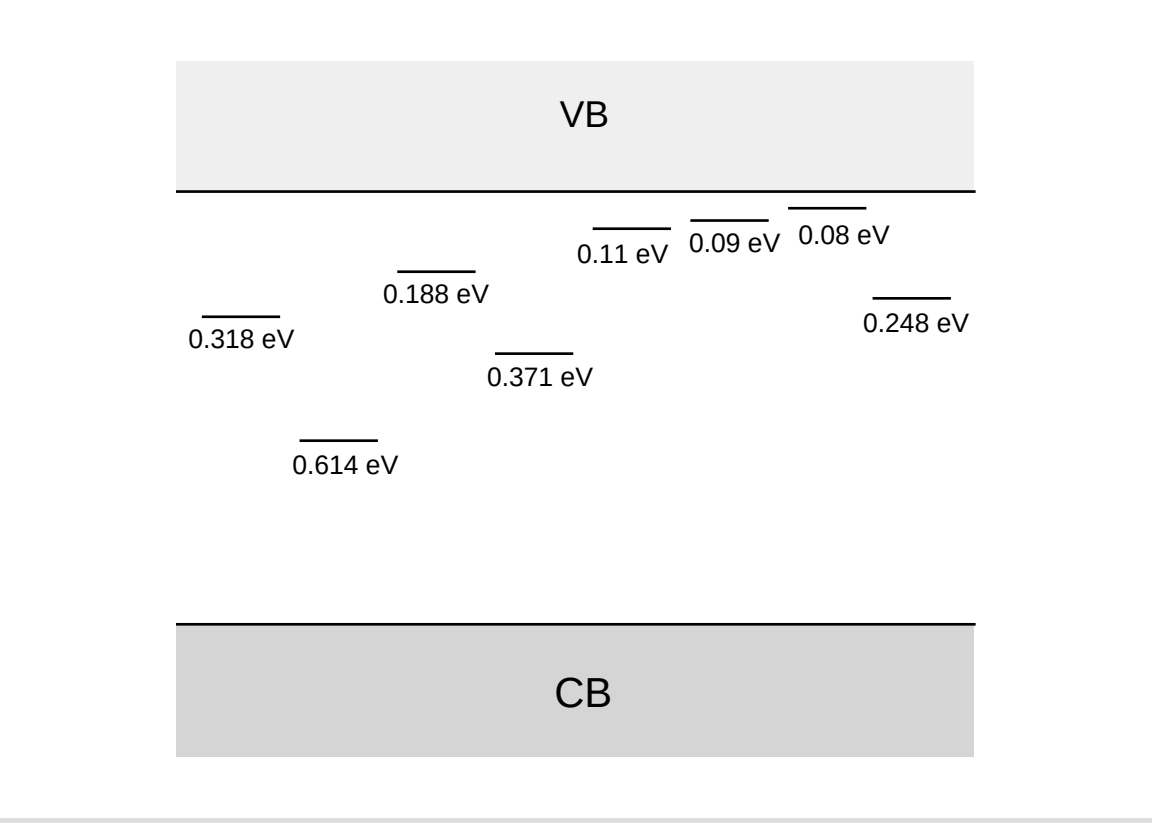
Discussion

sulfur introduces states in the gap



Discussion

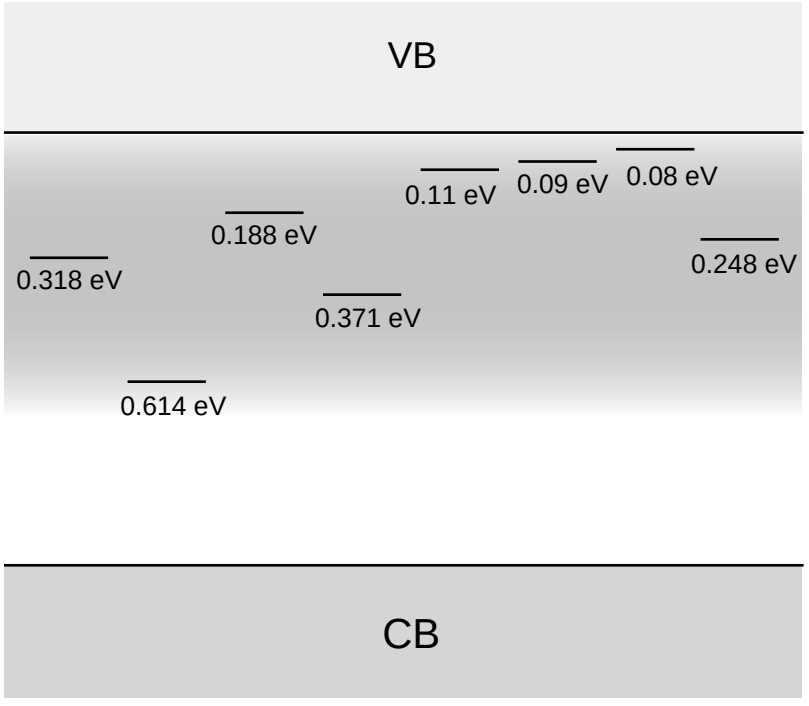
sulfur introduces states in the gap



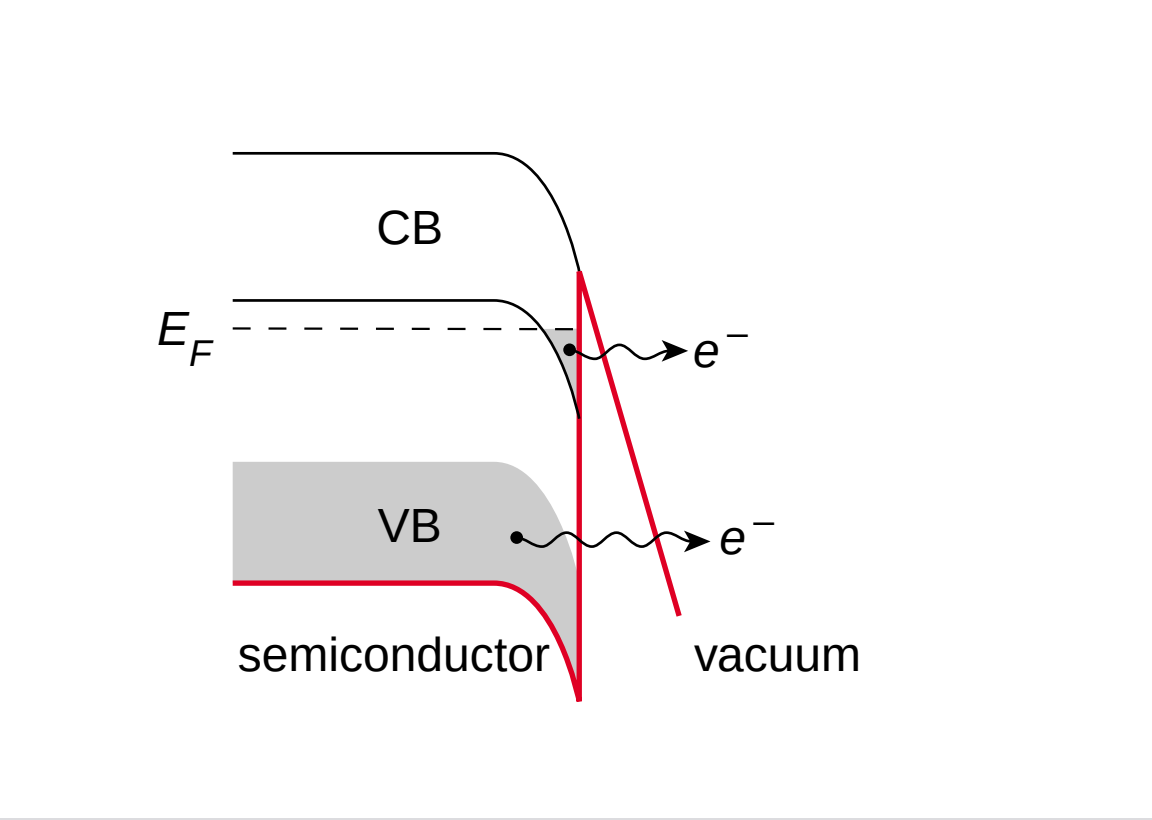
Janzén, *et al.*, *Phys. Rev. B* **29**,1907 (1984)

Discussion

states broaden into a band

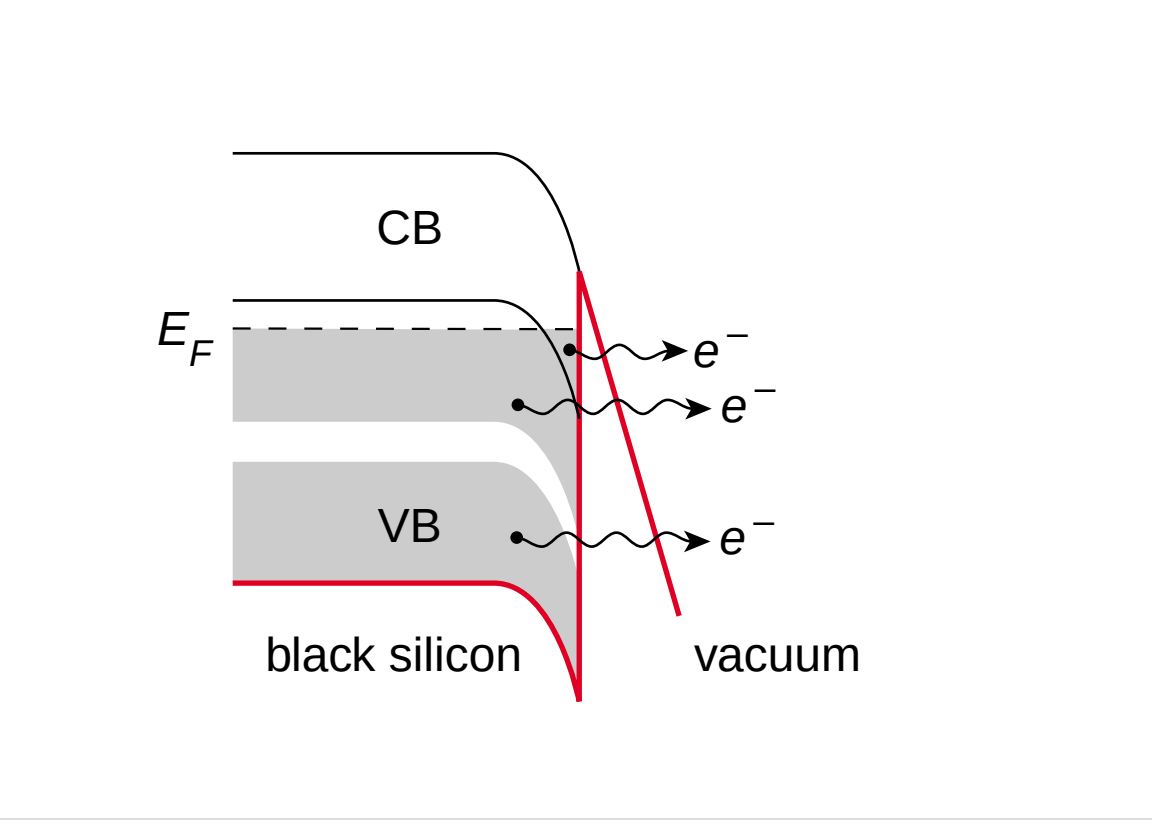


Discussion



Discussion

sulfur band provides additional electrons



Summary

Microstructured silicon

- **fabricated by simple, maskless process**

Summary

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- **fabricated by simple, maskless process**
- **can be integrated with microelectronics**

Summary

Microstructured silicon

- **fabricated by simple, maskless process**
- **can be integrated with microelectronics**
- **provides stable, high field-emission current**

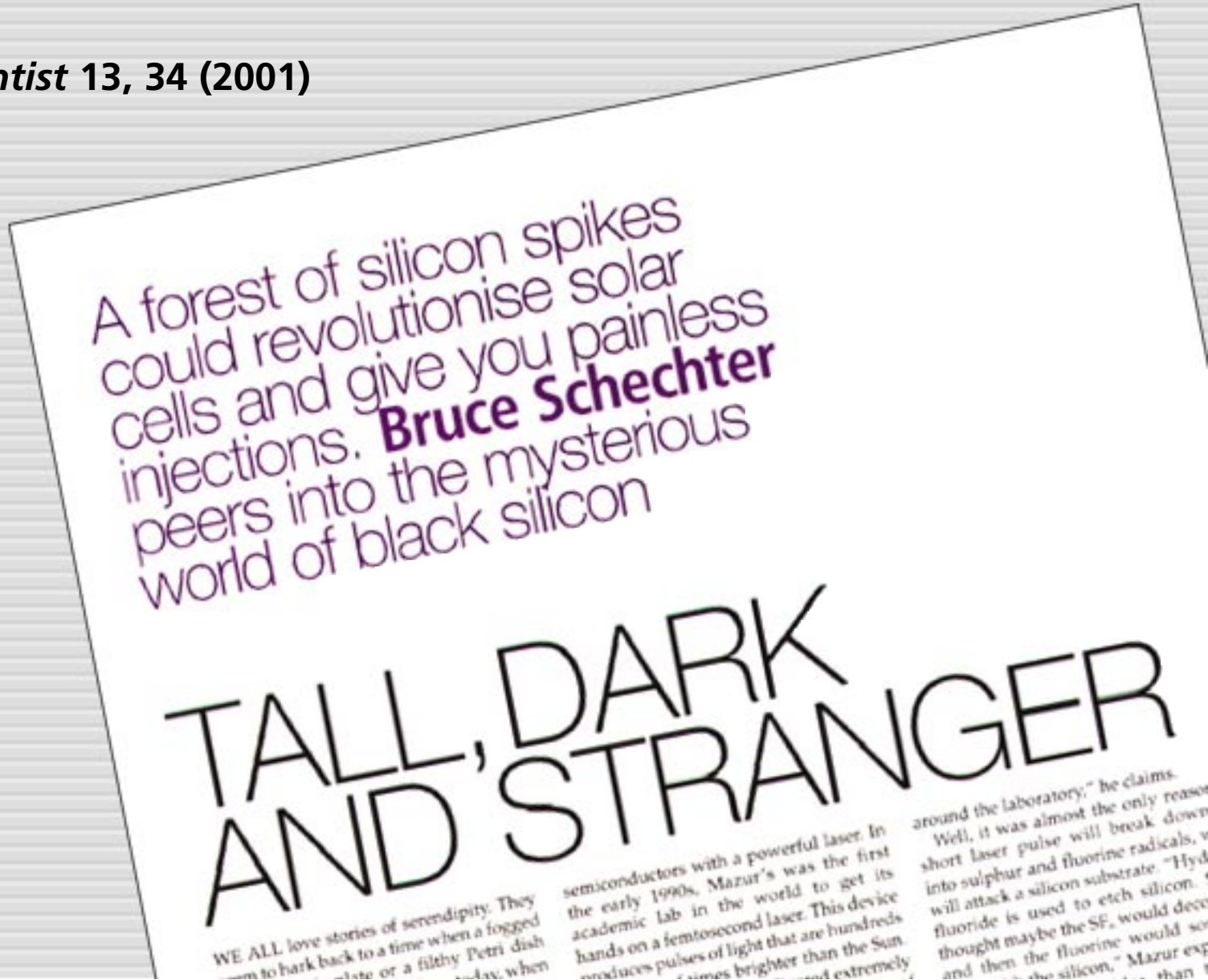
Summary

Microstructured silicon

- **fabricated by simple, maskless process**
- **can be integrated with microelectronics**
- **provides stable, high field-emission current**
- **is durable**

Summary

***New Scientist* 13, 34 (2001)**



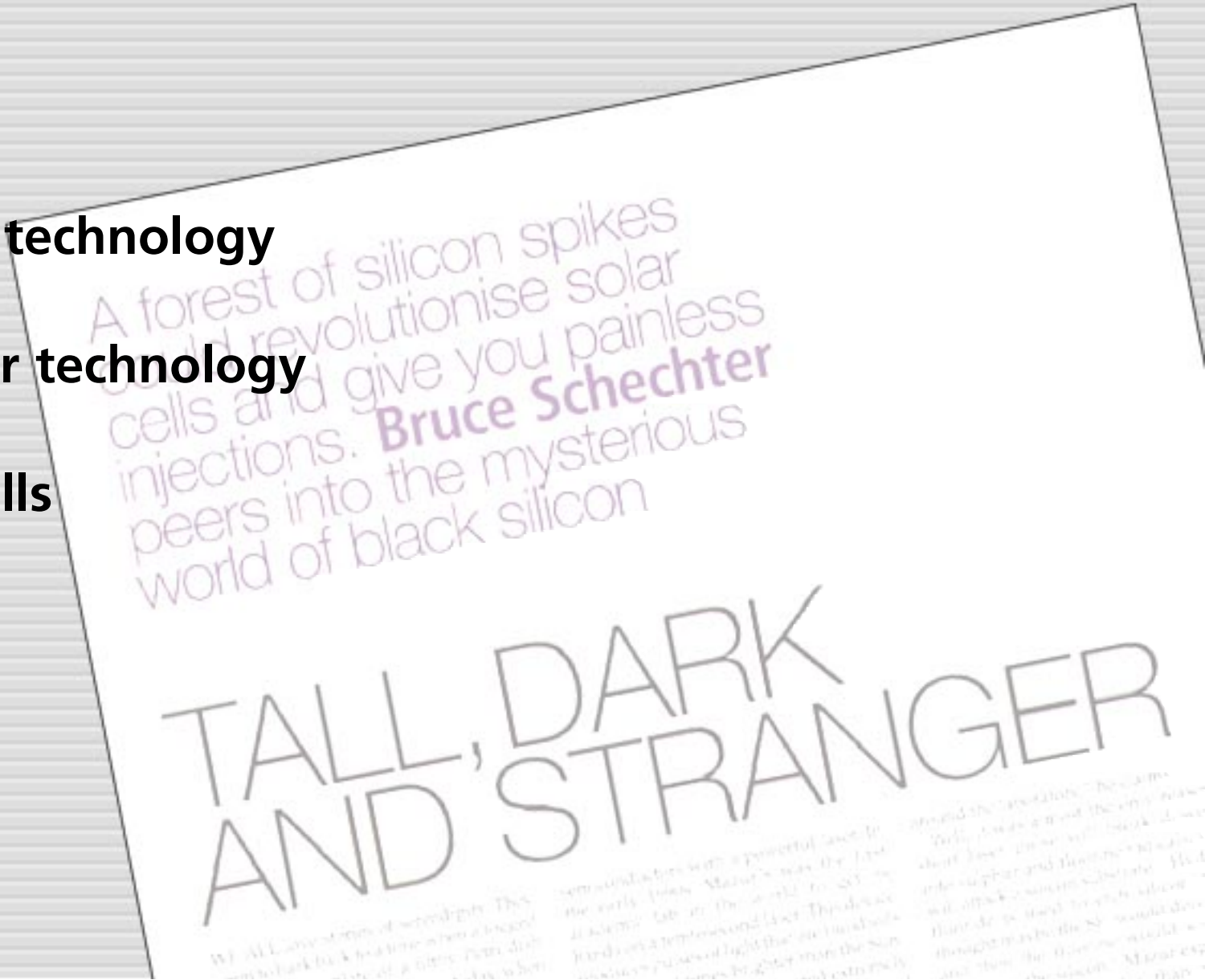
A forest of silicon spikes could revolutionise solar cells and give you painless injections. **Bruce Schechter** peers into the mysterious world of black silicon

TALL, DARK AND STRANGER

WE ALL love stories of serendipity. They
semiconductors with a powerful laser. In
the early 1990s, Mazur's was the first
academic lab in the world to get its
hands on a femtosecond laser. This device
produces pulses of light that are hundreds
around the laboratory," he claims.
Well, it was almost the only reason
short laser pulse will break down
into sulphur and fluorine radicals,
will attack a silicon substrate. "Hyd
fluoride is used to etch silicon.
thought maybe the SF_6 would deco
and then the fluorine would so
the silicon," Mazur exp

Applications

- display technology
- detector technology
- solar cells



Applications

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