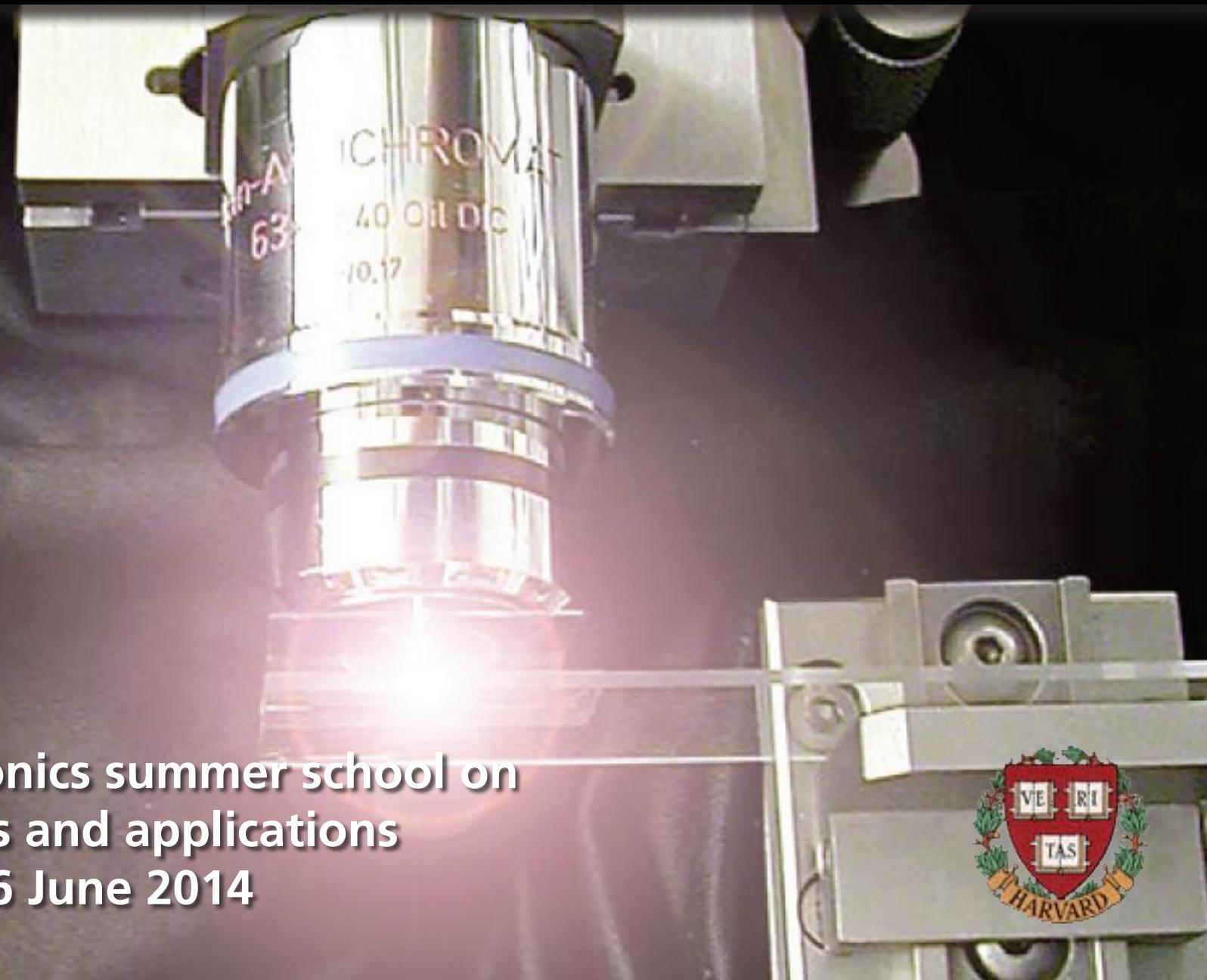


Fundamentals of intense laser interactions with solids



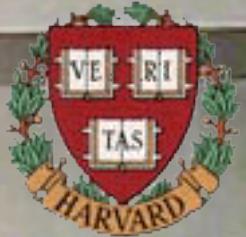
Extreme Photonics summer school on
Ultrafast lasers and applications
Ottawa, CA, 26 June 2014

Fundamentals of intense laser interactions with solids



@eric_mazur

Extreme Photonics summer school on
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Outline

- propagation of pulses
- nonlinear optics
- femtosecond micromachining

Propagation of EM wave 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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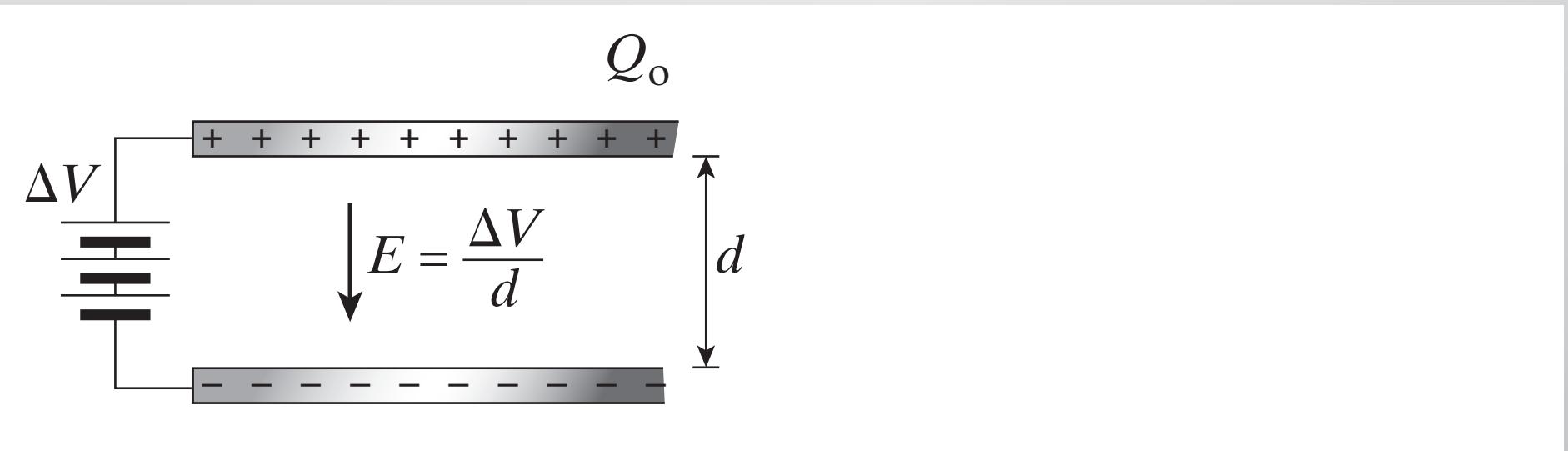
In nonferromagnetic media $\mu \approx 1$, and so $n \approx \sqrt{\epsilon}$.

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

Propagation of EM wave through medium

Dielectric constant measures increase in capacitance

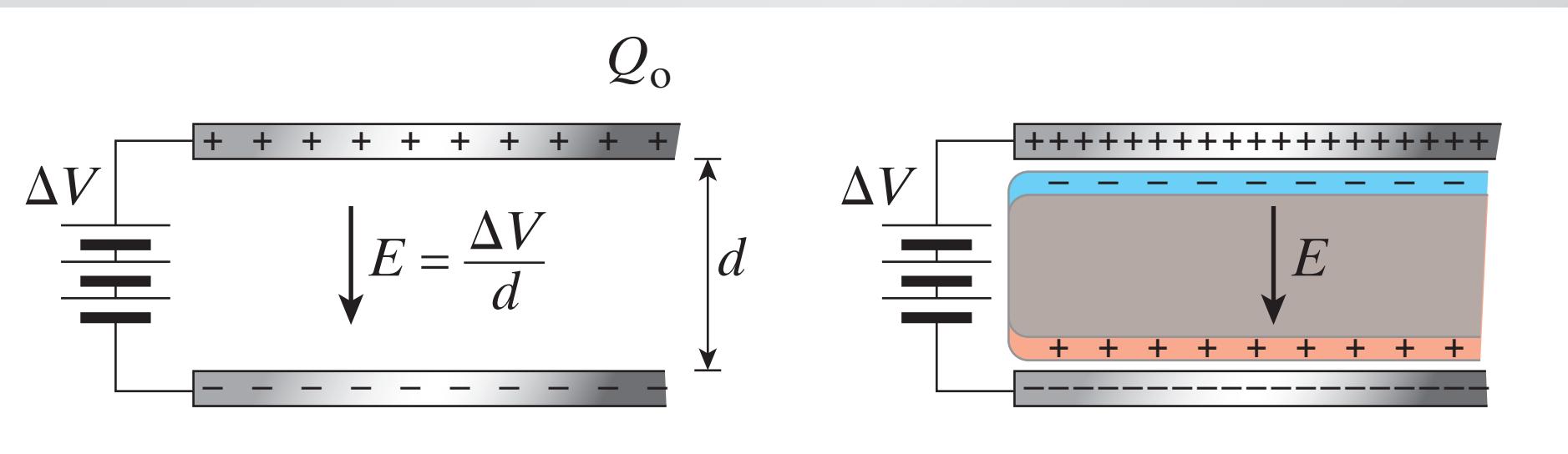
$$\epsilon = \frac{C_d}{C_o}$$



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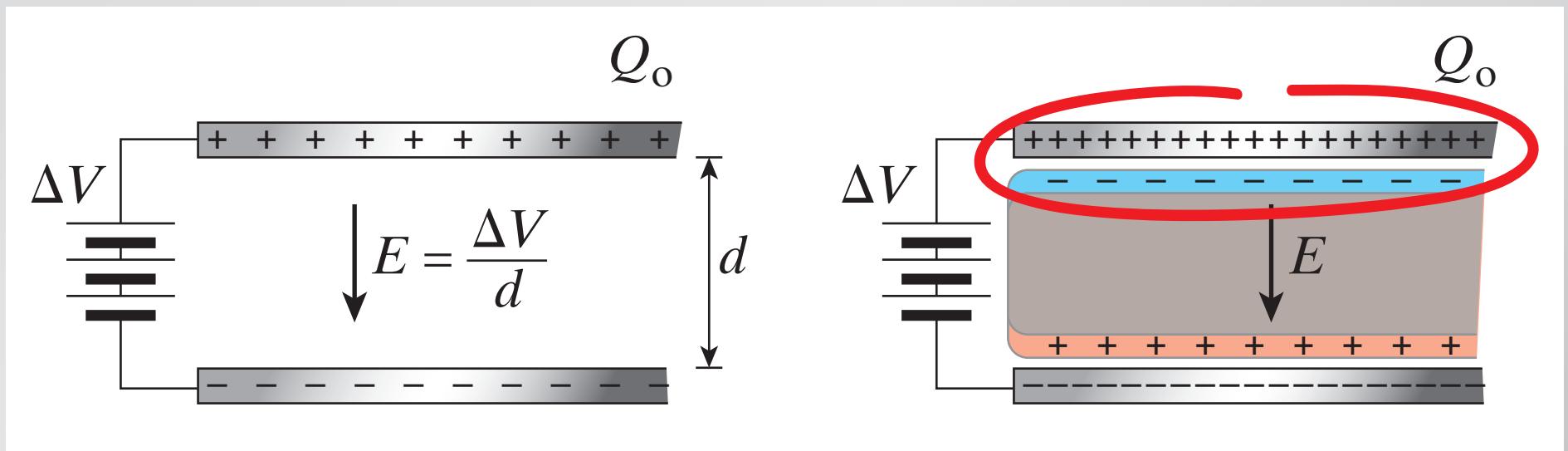
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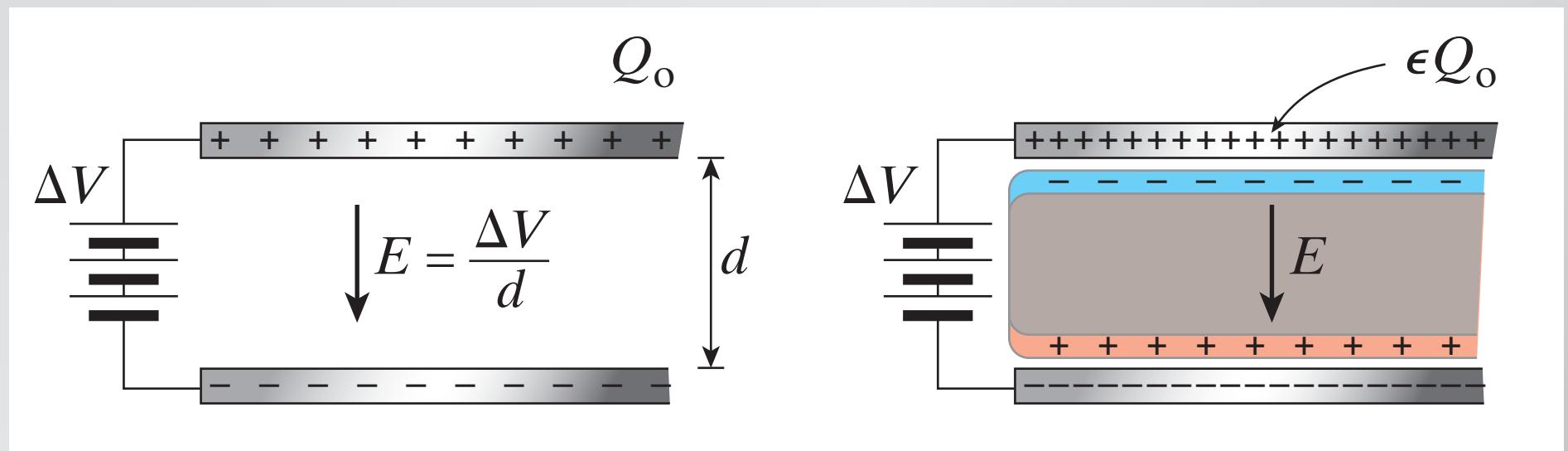
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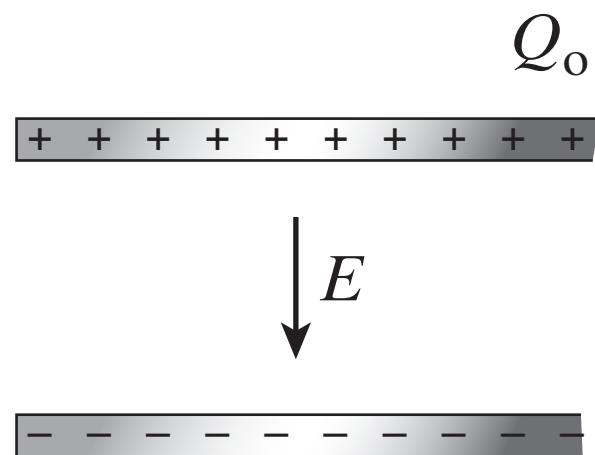
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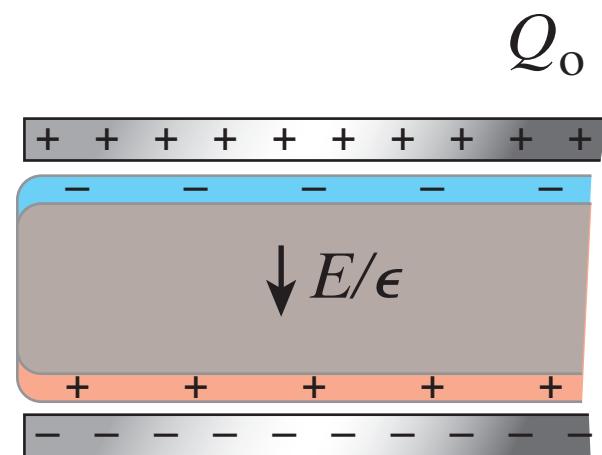
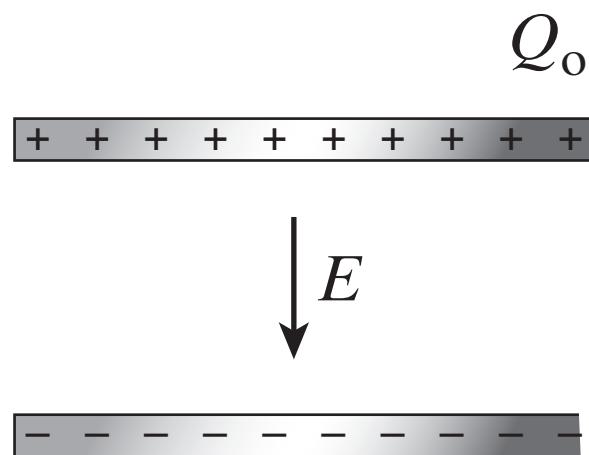
Propagation of EM wave through medium

Alternatively ϵ is measure of attenuation of electric field



Propagation of EM wave through medium

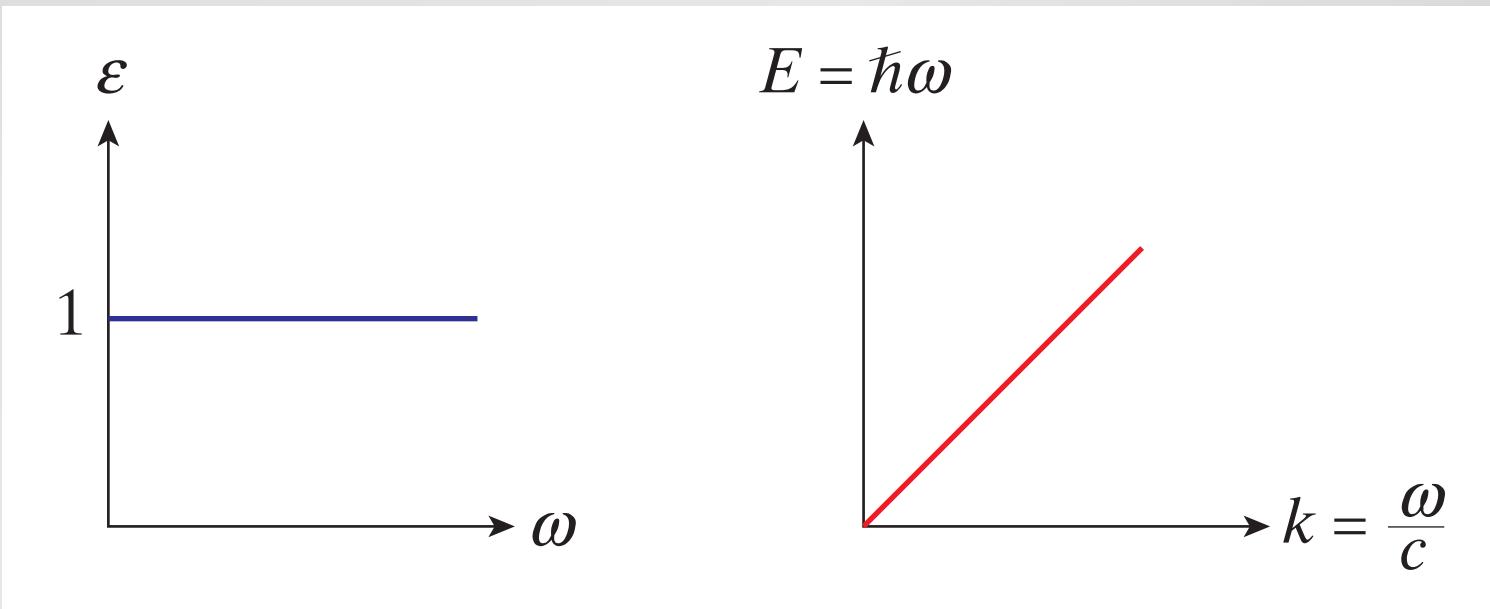
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Propagation of EM wave through medium

In vacuum:

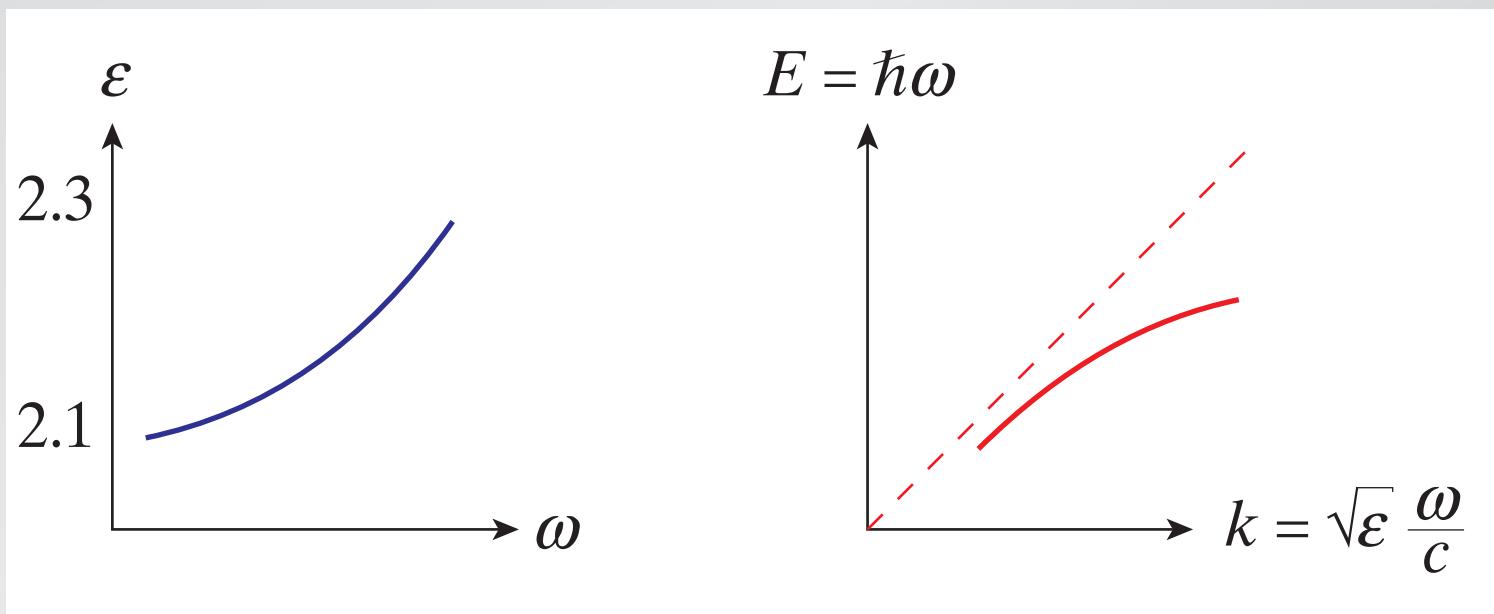
$$f\lambda = \frac{\omega}{k} = c \quad \Rightarrow \quad \omega = c k$$



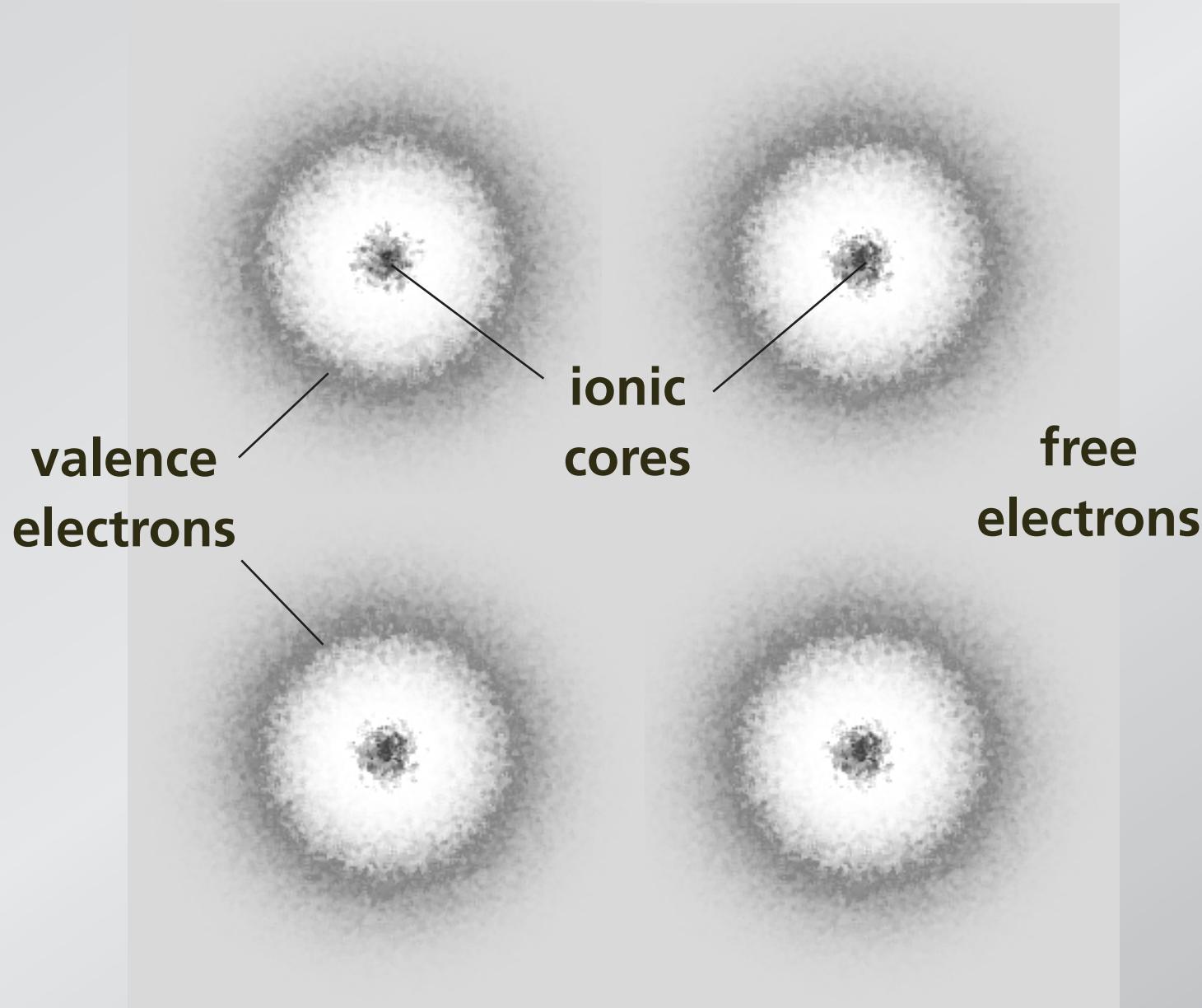
Propagation of EM wave through medium

In medium:

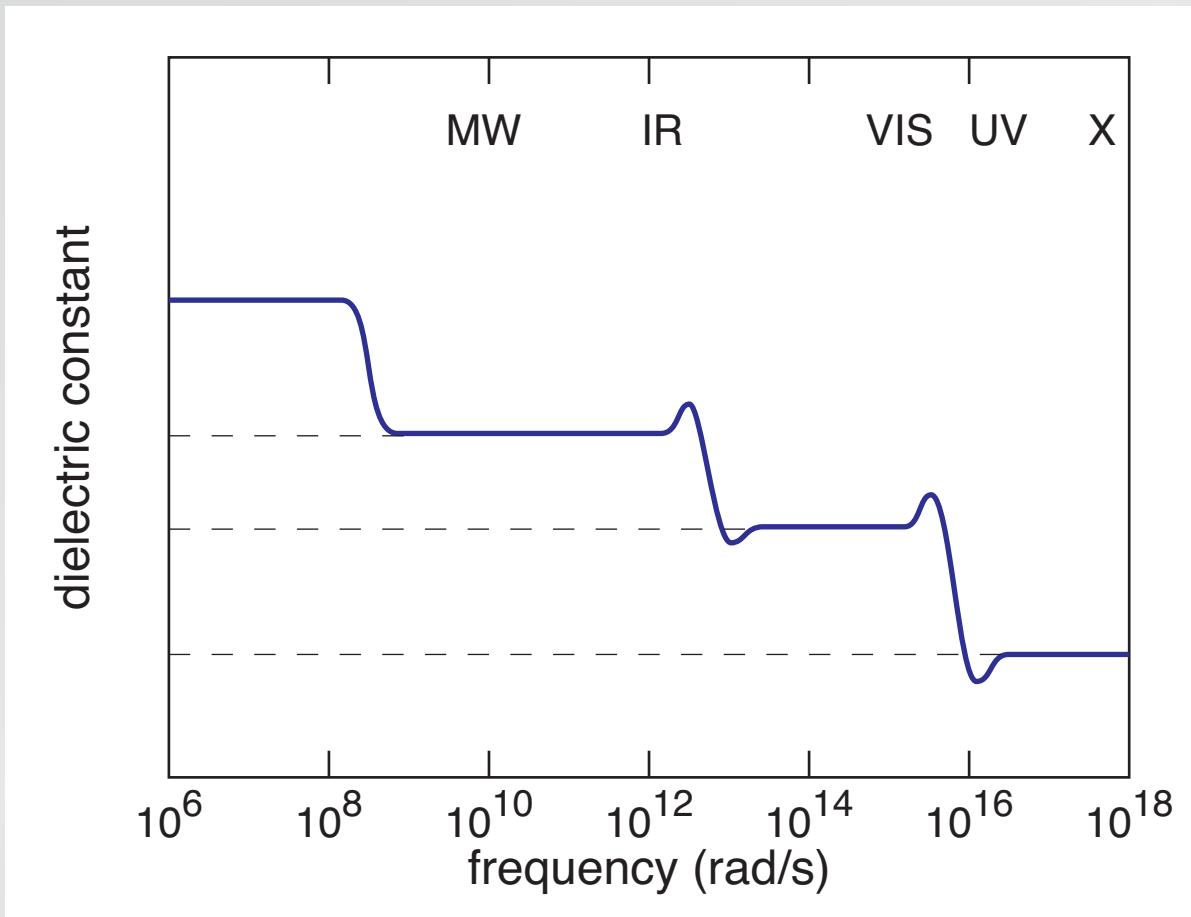
$$v = \frac{c}{\sqrt{\epsilon(\omega)}} = \frac{c}{n} \quad \Rightarrow \quad \omega = \frac{c}{\sqrt{\epsilon}} k$$



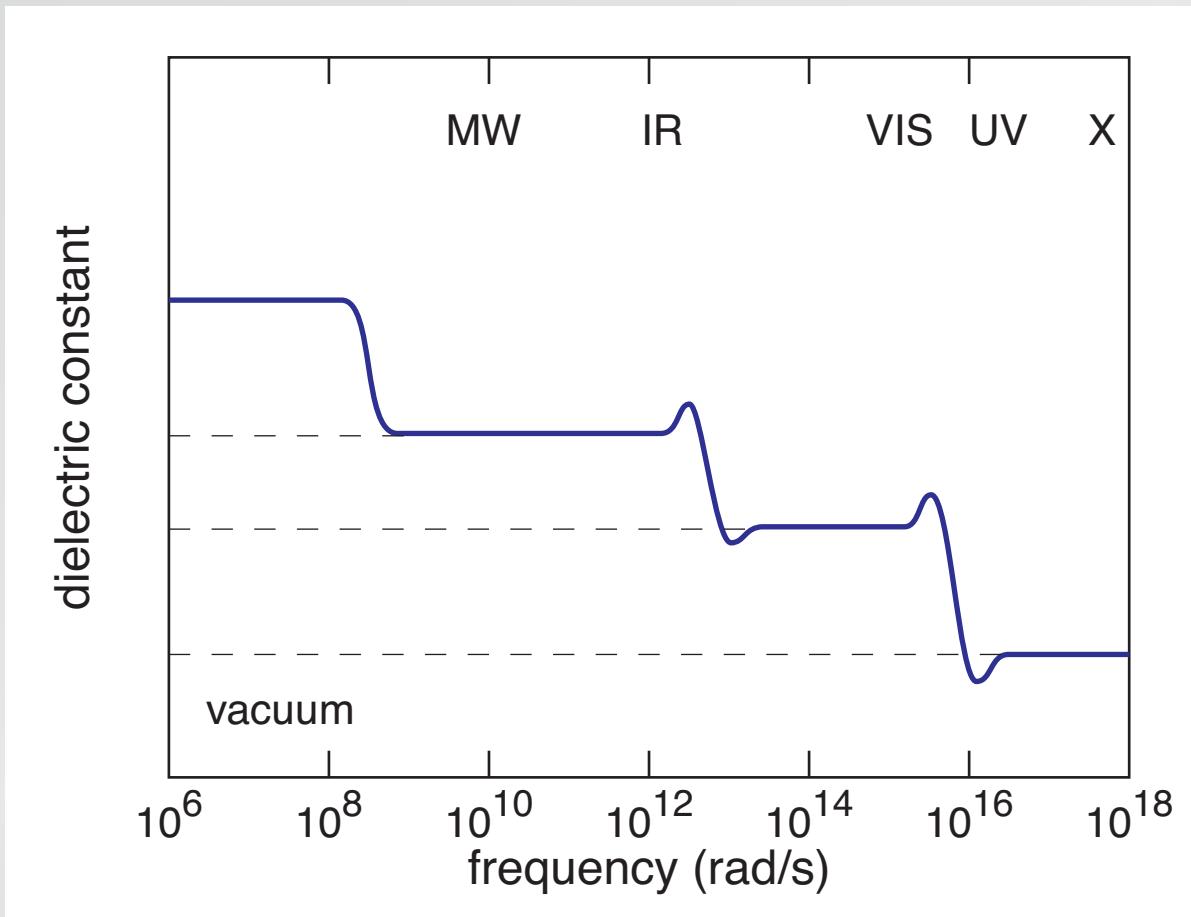
Which charges participate?



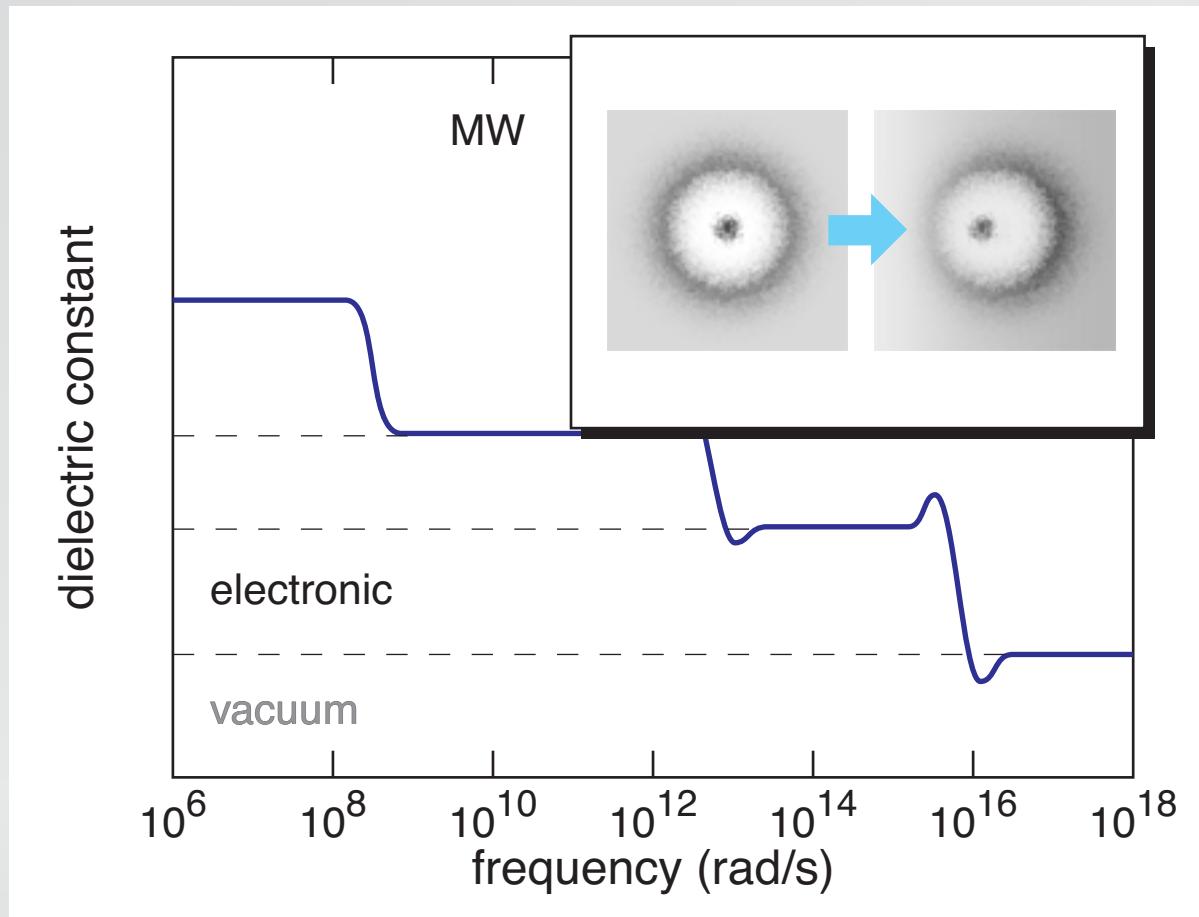
Dielectric function



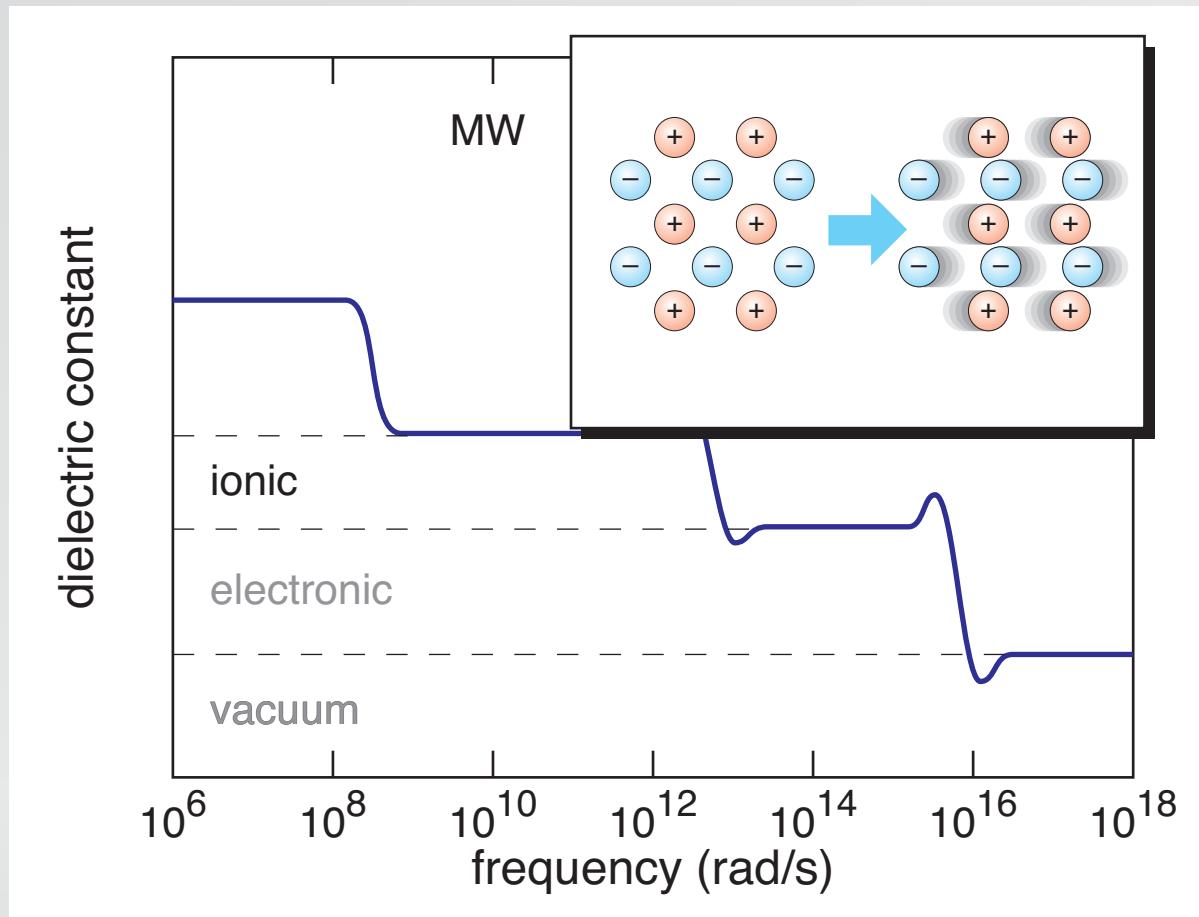
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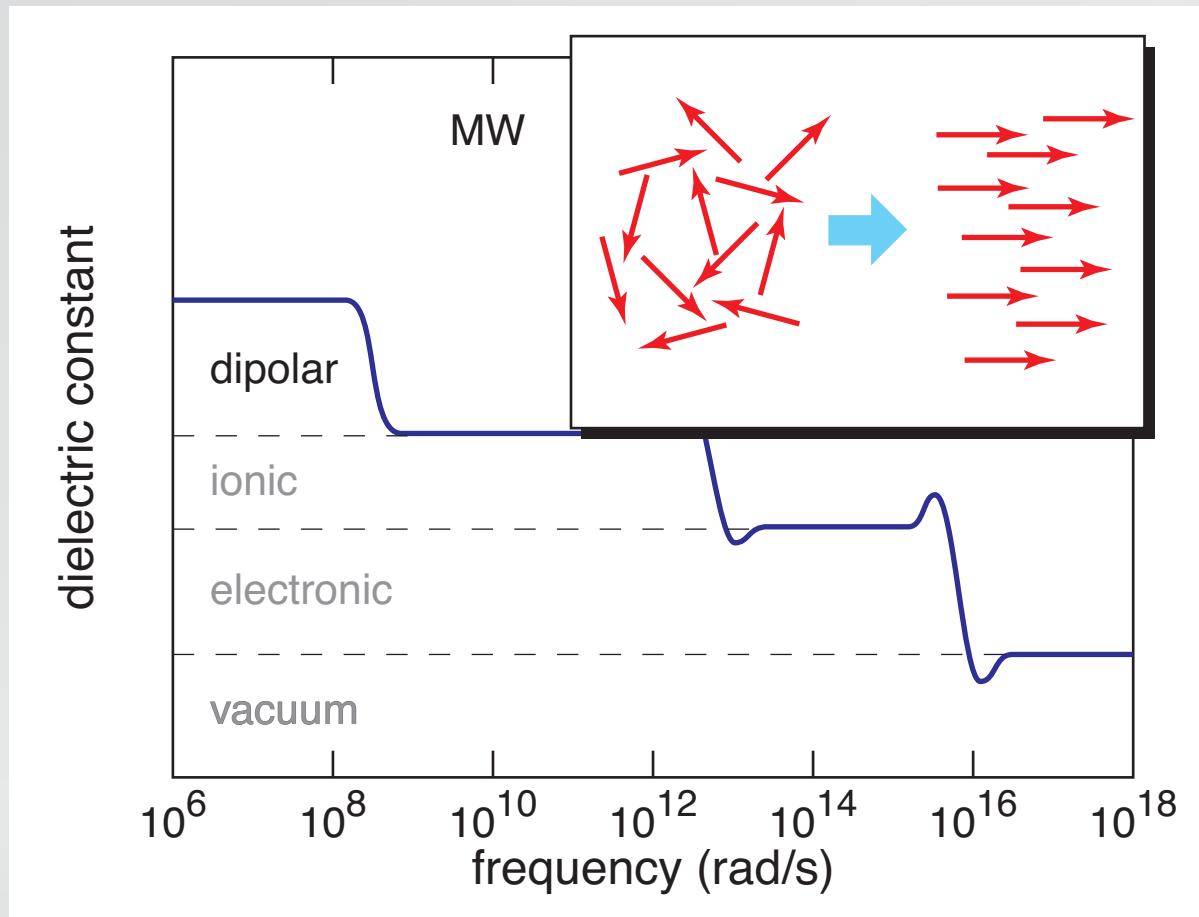
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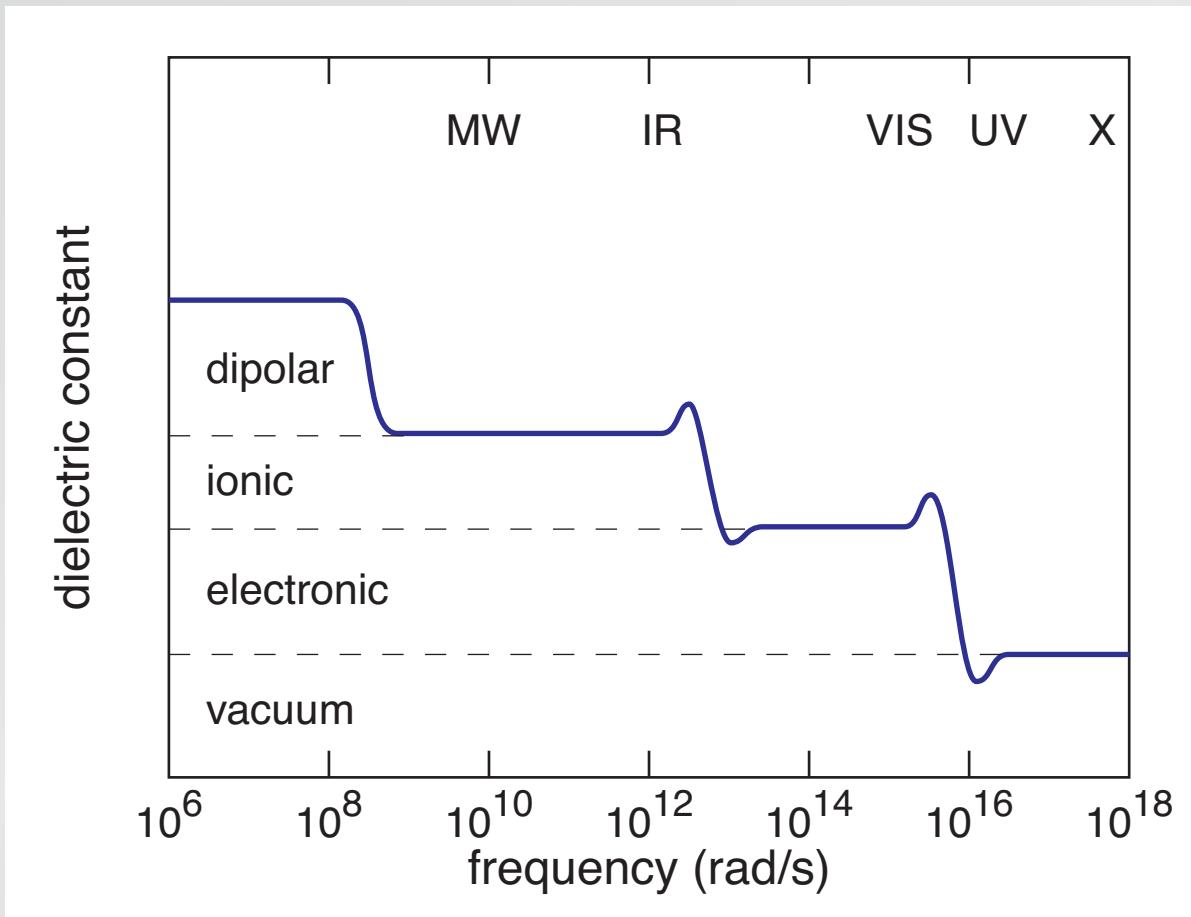
Dielectric function



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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

$$x(t) = x_o e^{-i\omega t}$$

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Polarization

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

Bound electrons

Dielectric function

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

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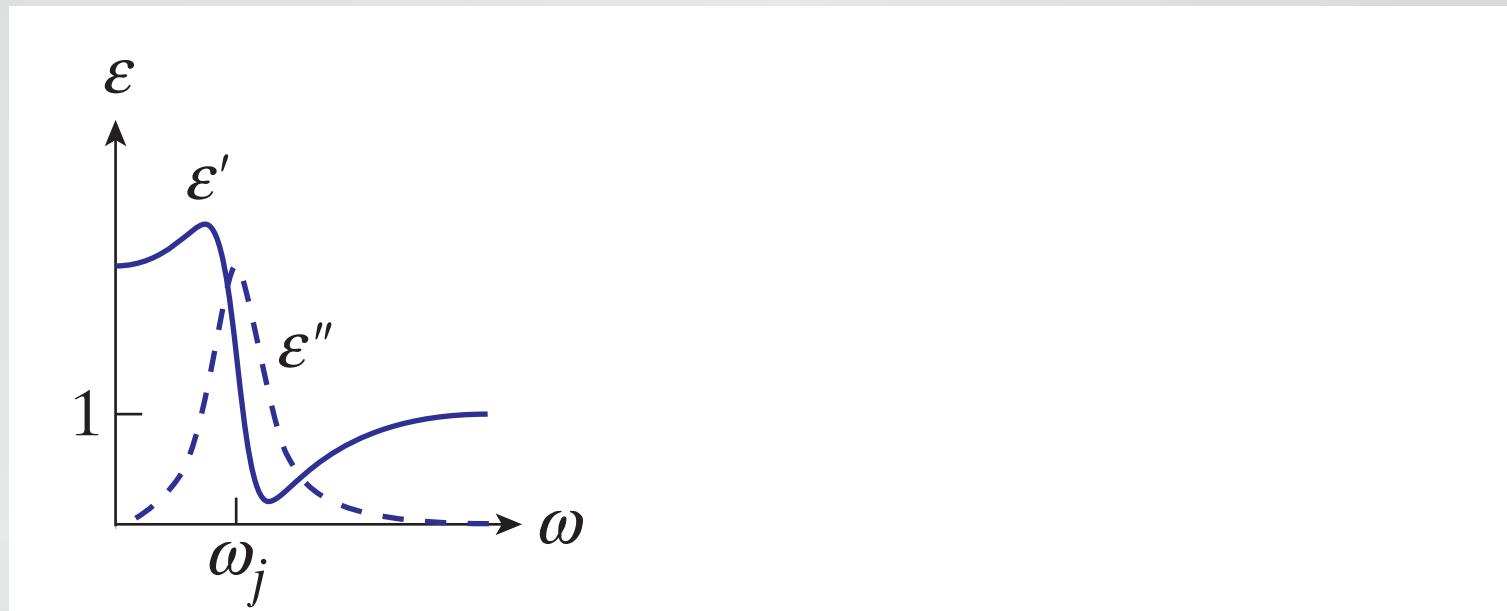
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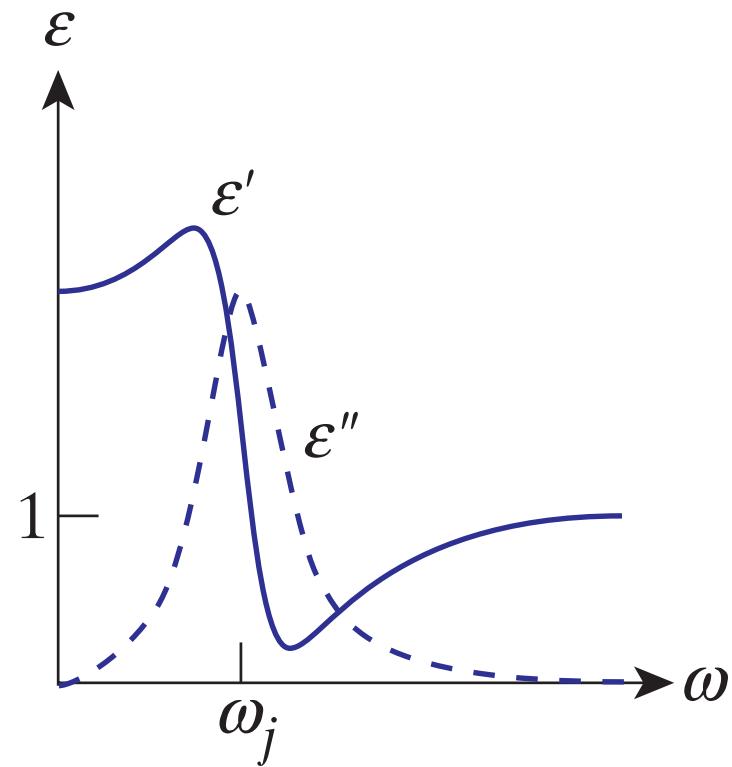
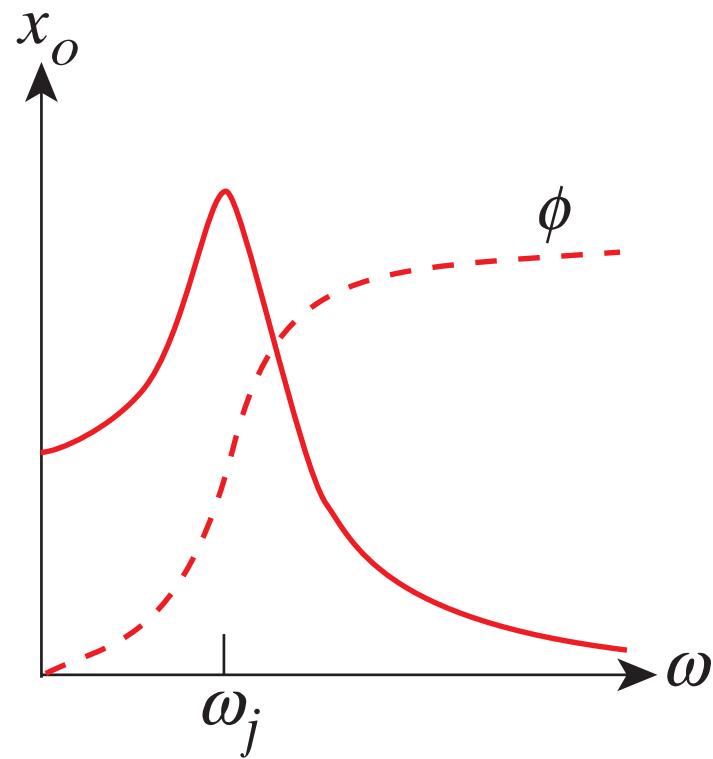
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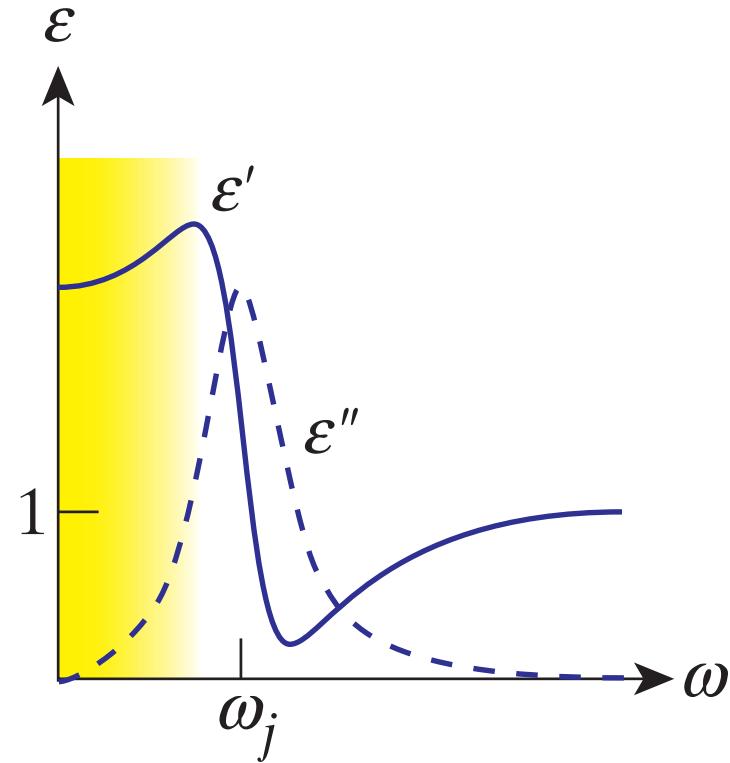
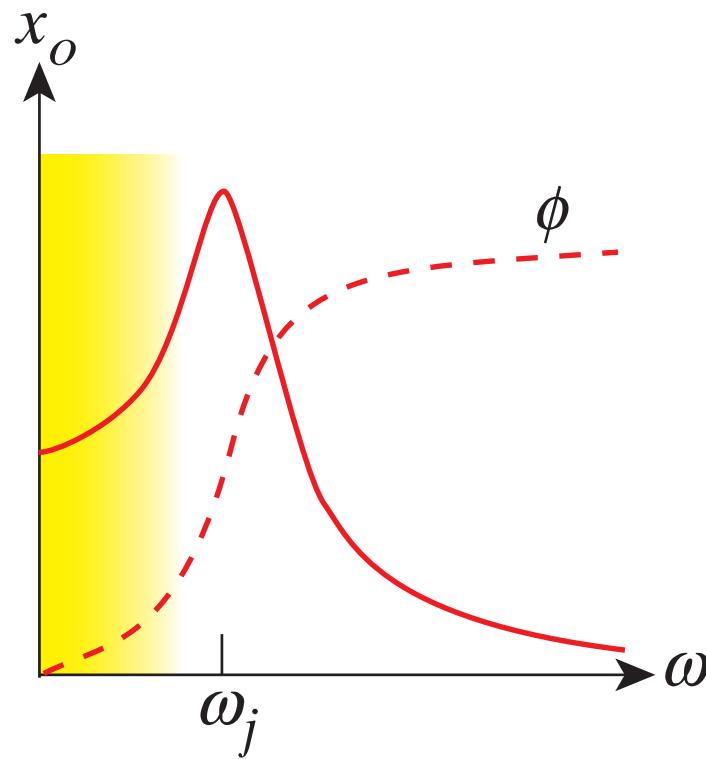
Bound electrons

Amplitude of bound charge oscillation



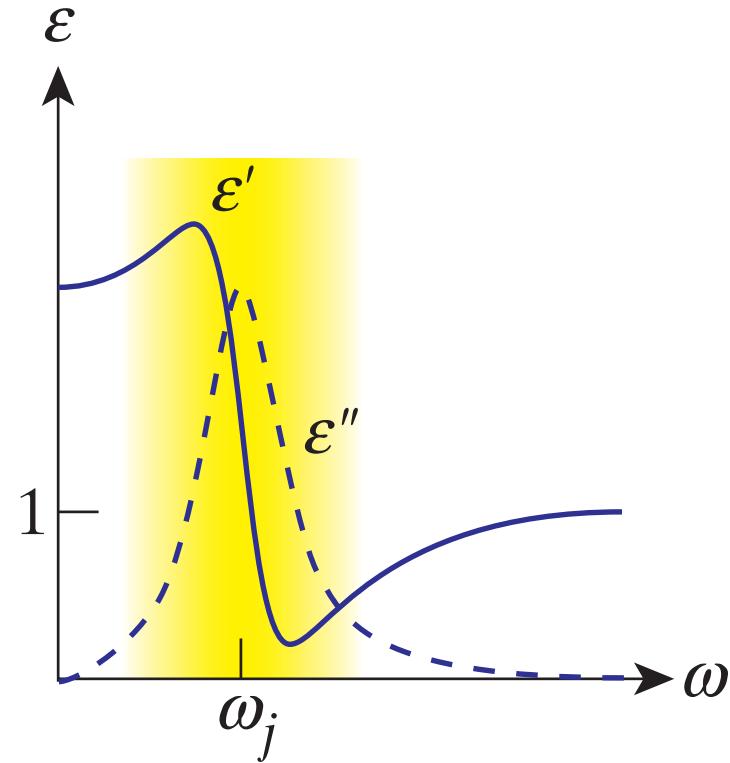
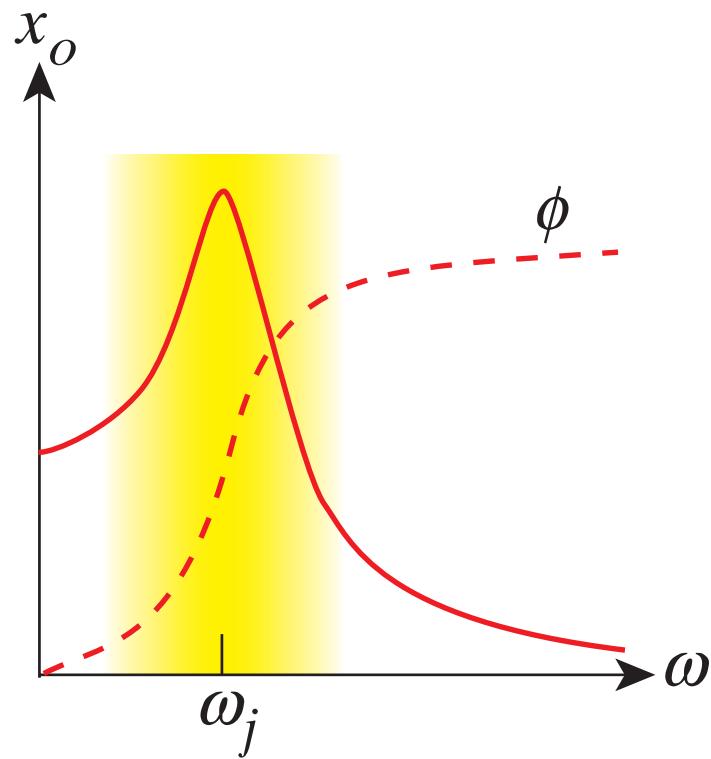
Bound electrons

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



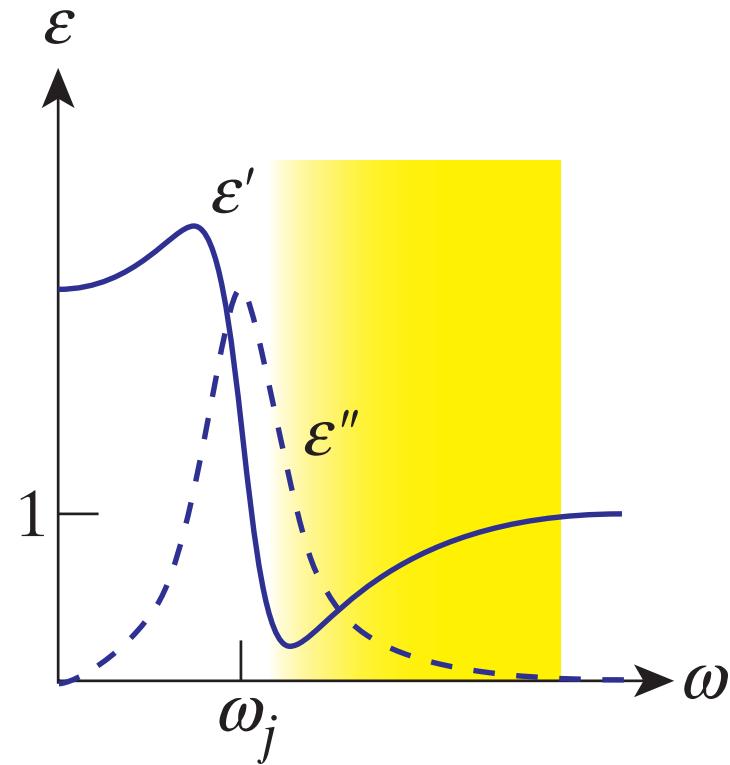
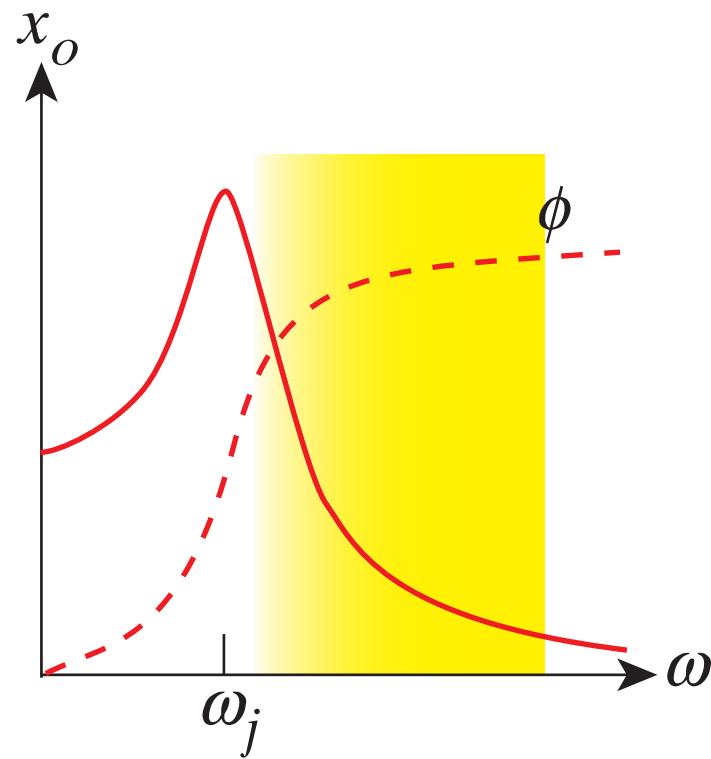
Bound electrons

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



Bound electrons

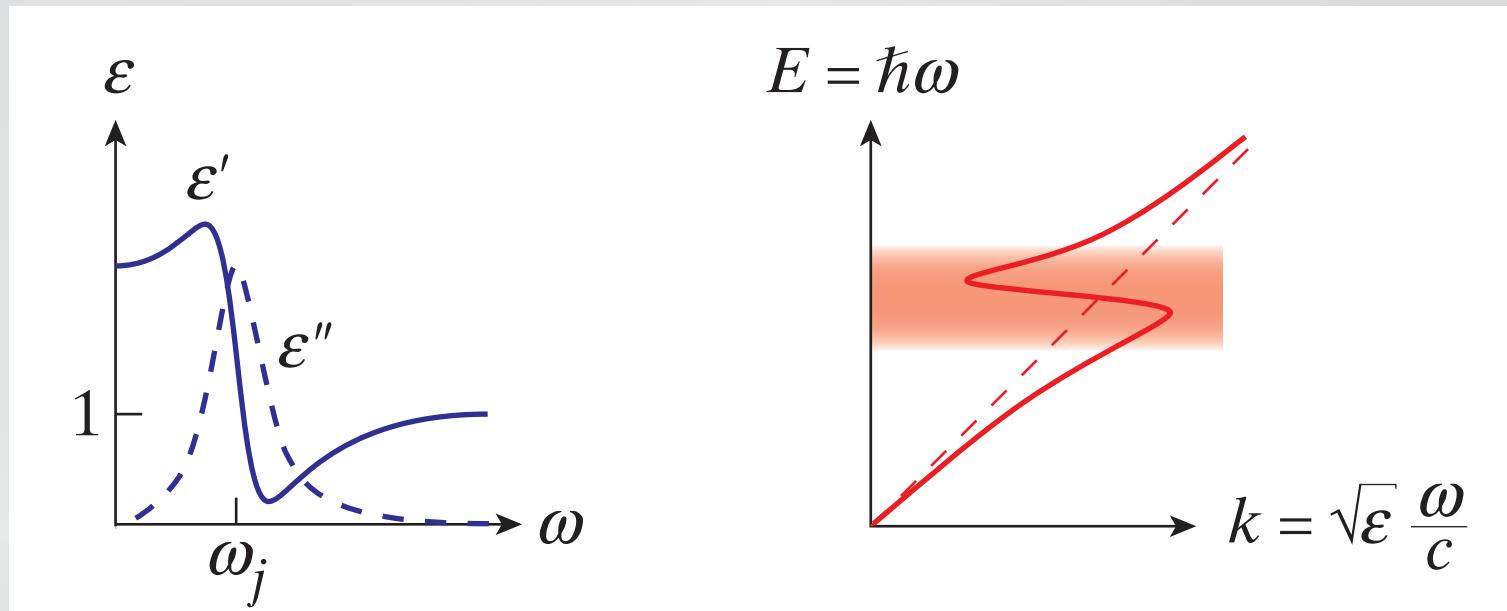
Above resonance: bound charges cannot keep up
with driving field \Rightarrow dielectric like a vacuum



Bound electrons

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



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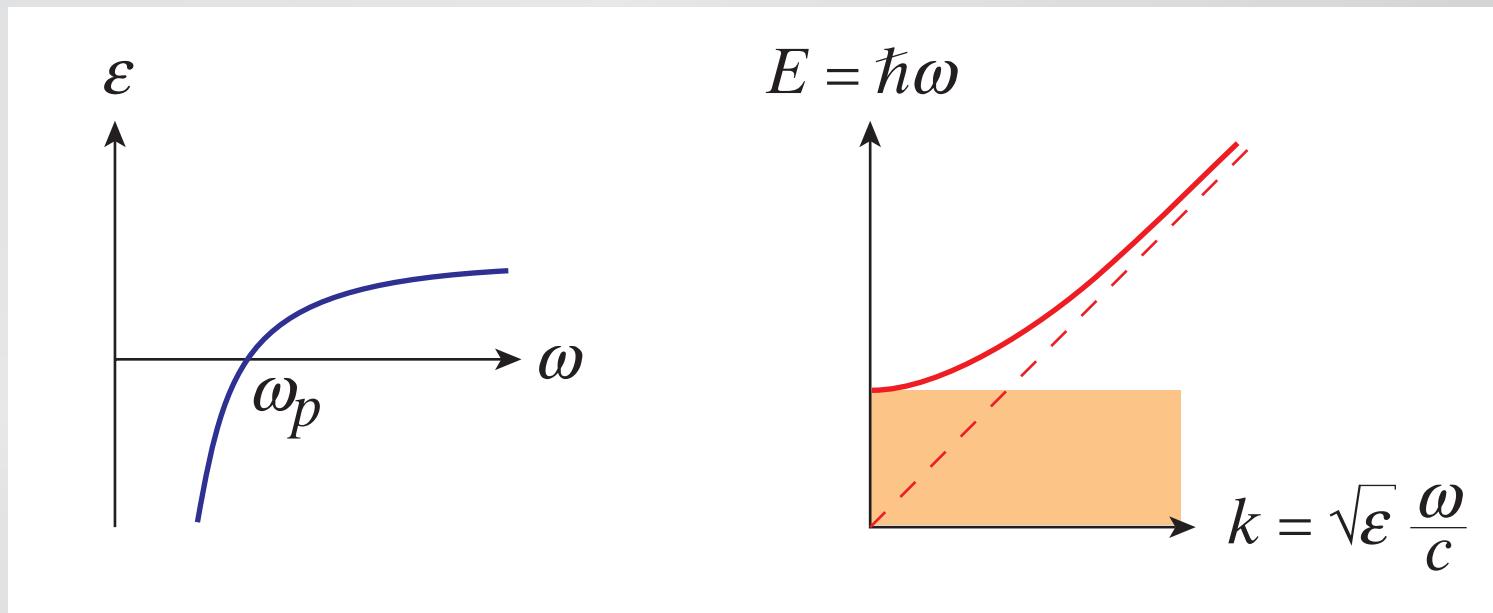
$$\epsilon'(\omega) = 1 - \frac{Ne^2}{m\epsilon_0} \frac{1}{\omega^2} \equiv 1 - \frac{\omega_p^2}{\omega^2}$$

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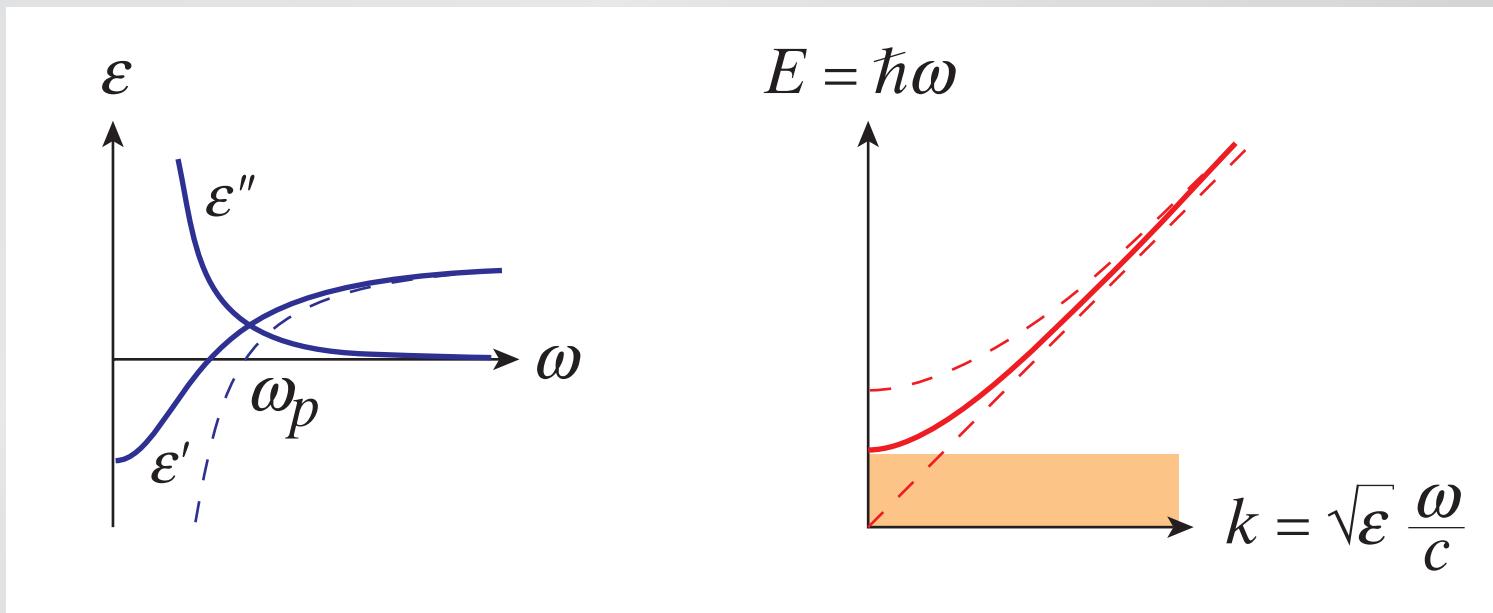


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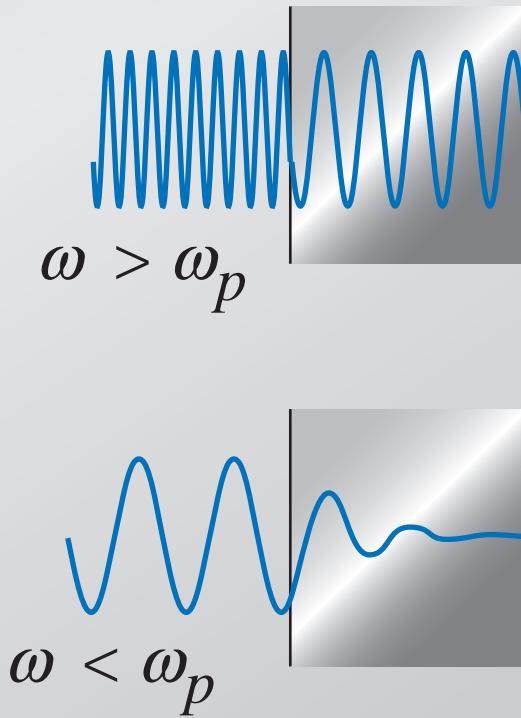
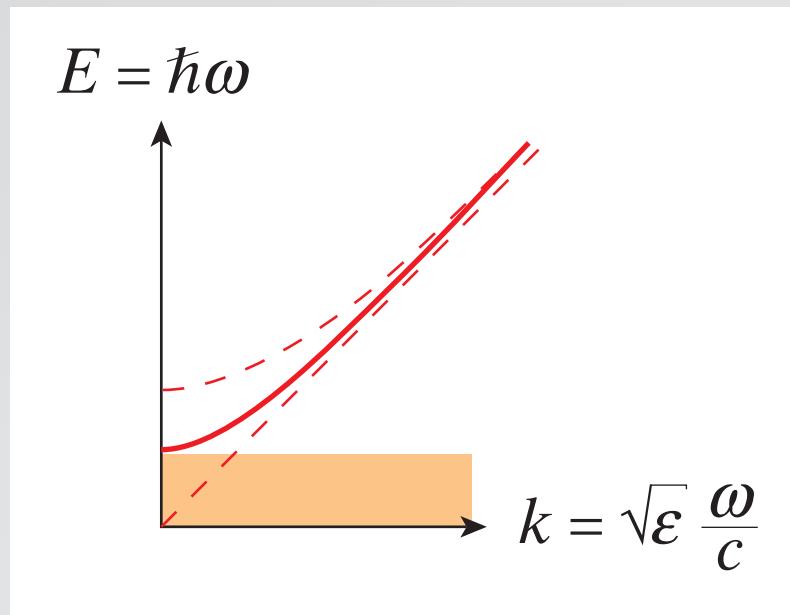
Add damping:

$$\gamma \lesssim \omega_p$$



Free electrons

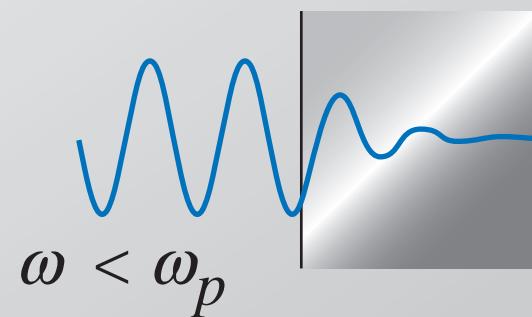
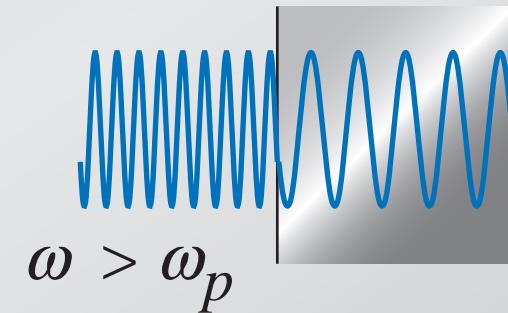
Plasma acts like a high-pass filter



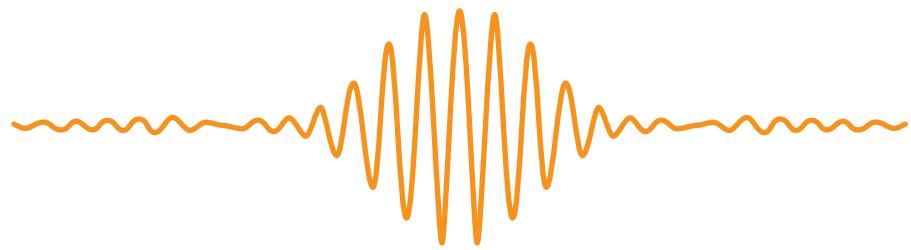
Free electrons

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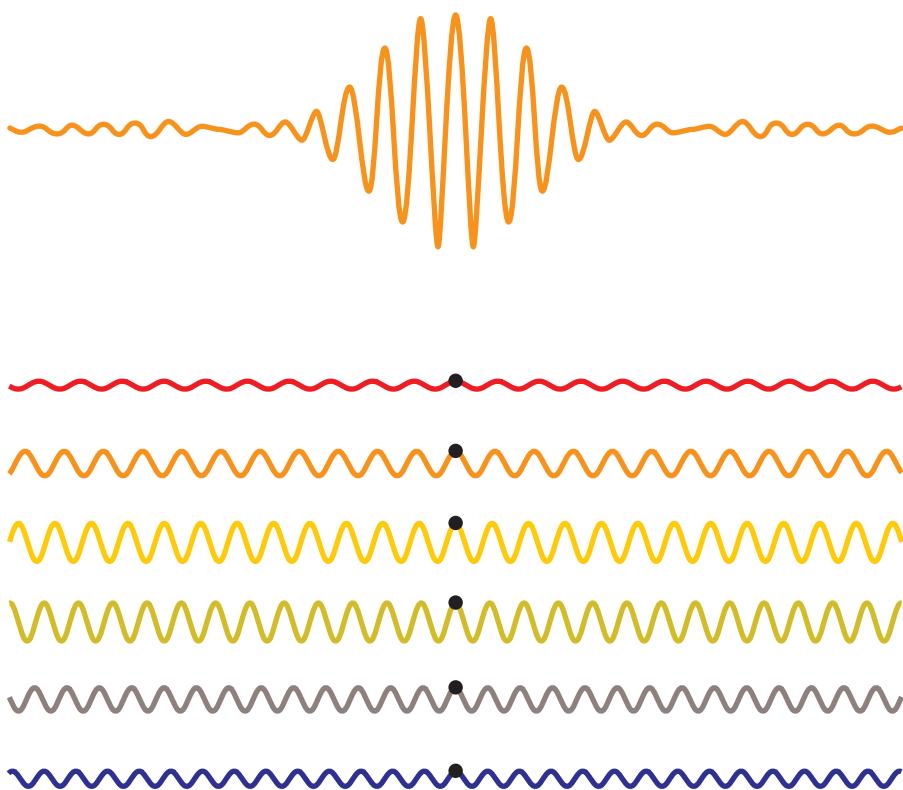
$\log N$ (cm $^{-3}$)	ω_p (rad s $^{-1}$)	λ_p
22	6×10^{15}	330 nm
18	6×10^{13}	33 μm
14	6×10^{11}	3.3 mm
10	6×10^9	0.33 m



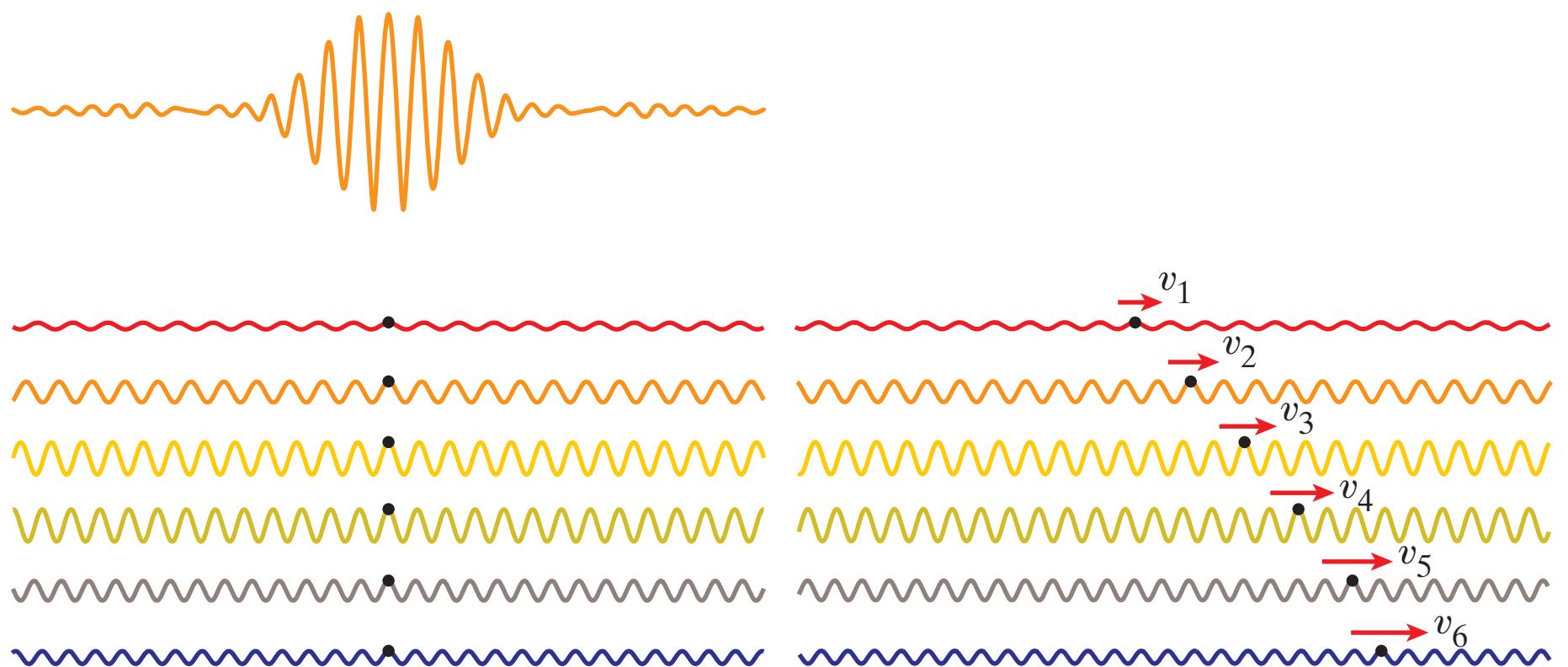
Pulse dispersion



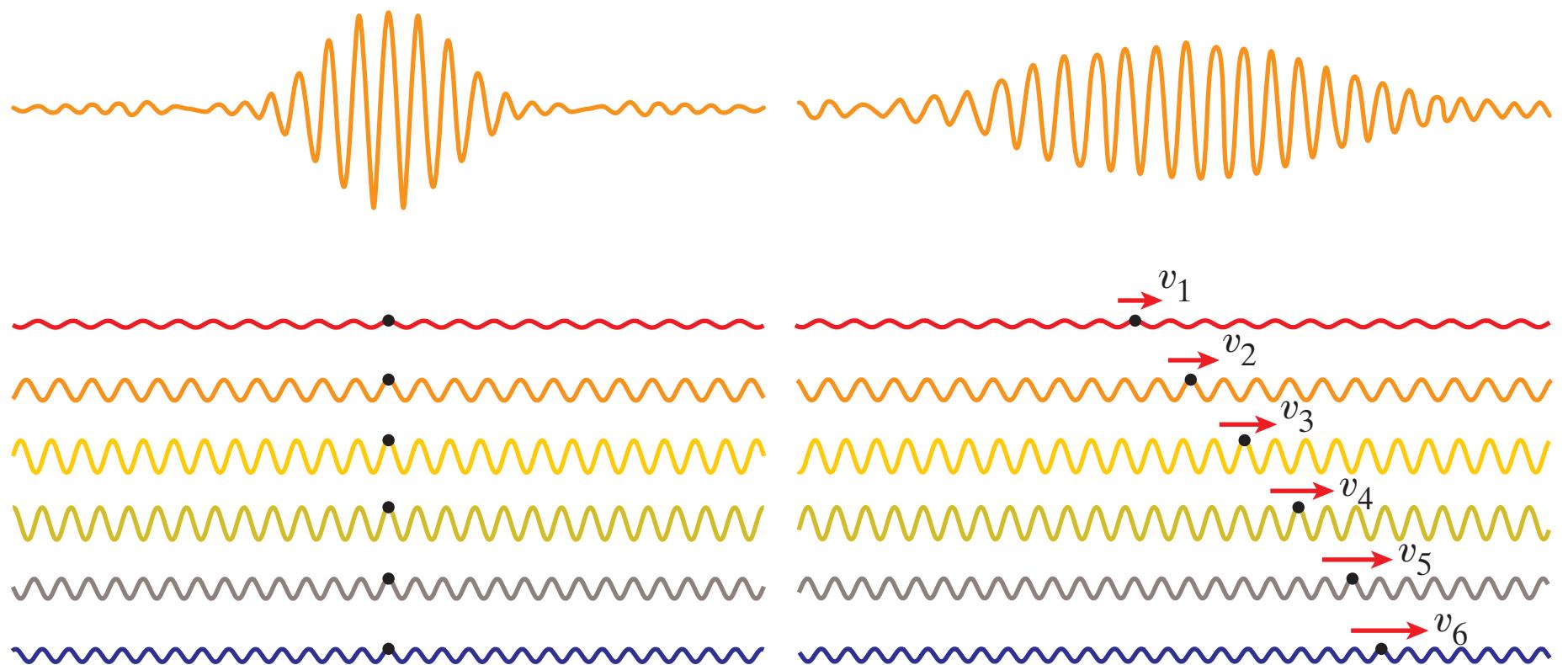
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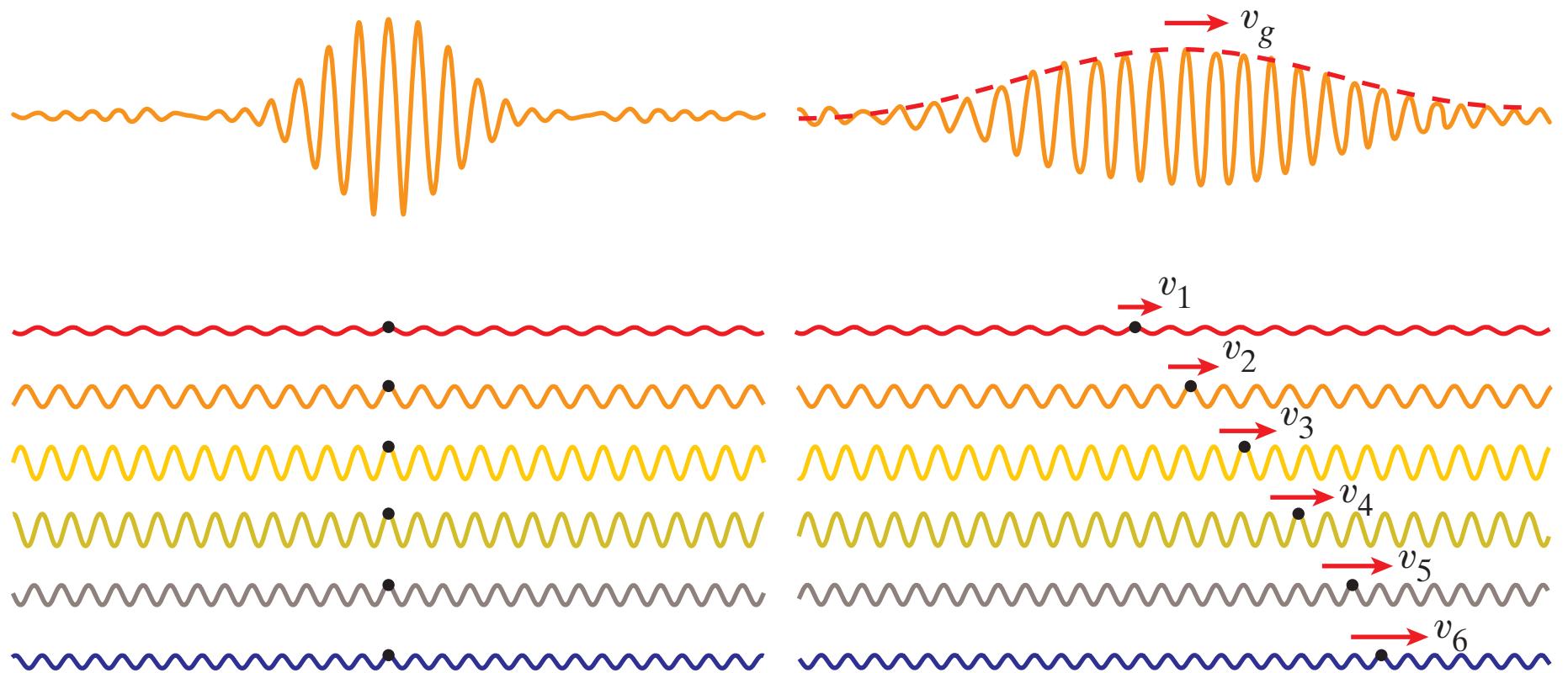
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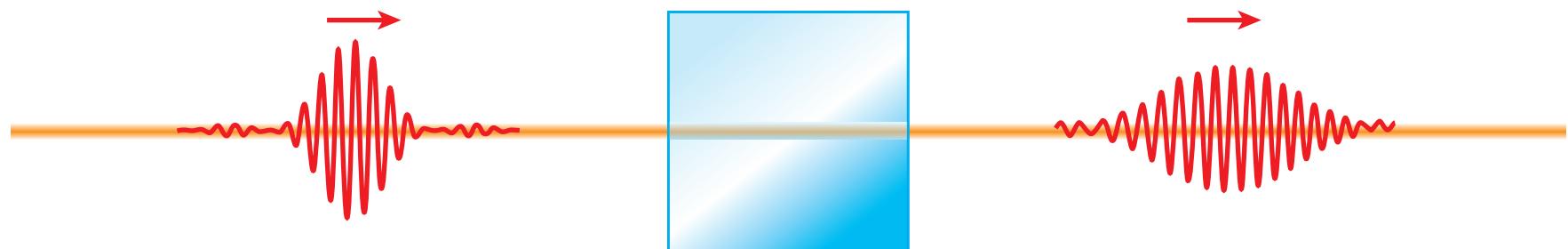


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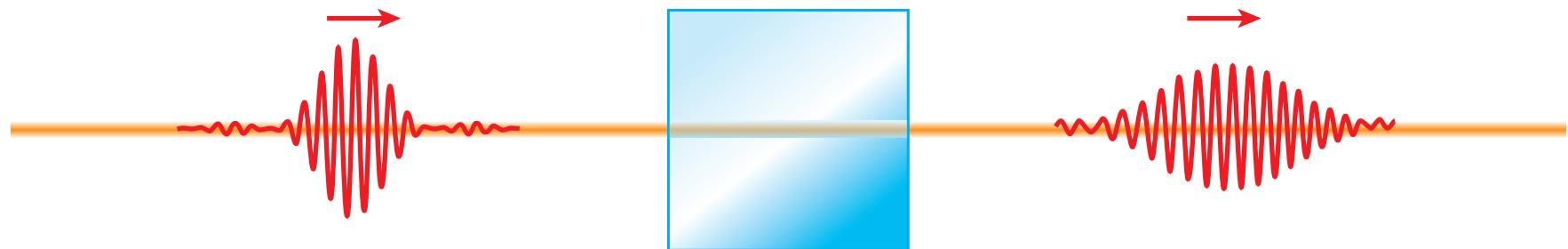
Pulse dispersion

medium causes pulse to stretch



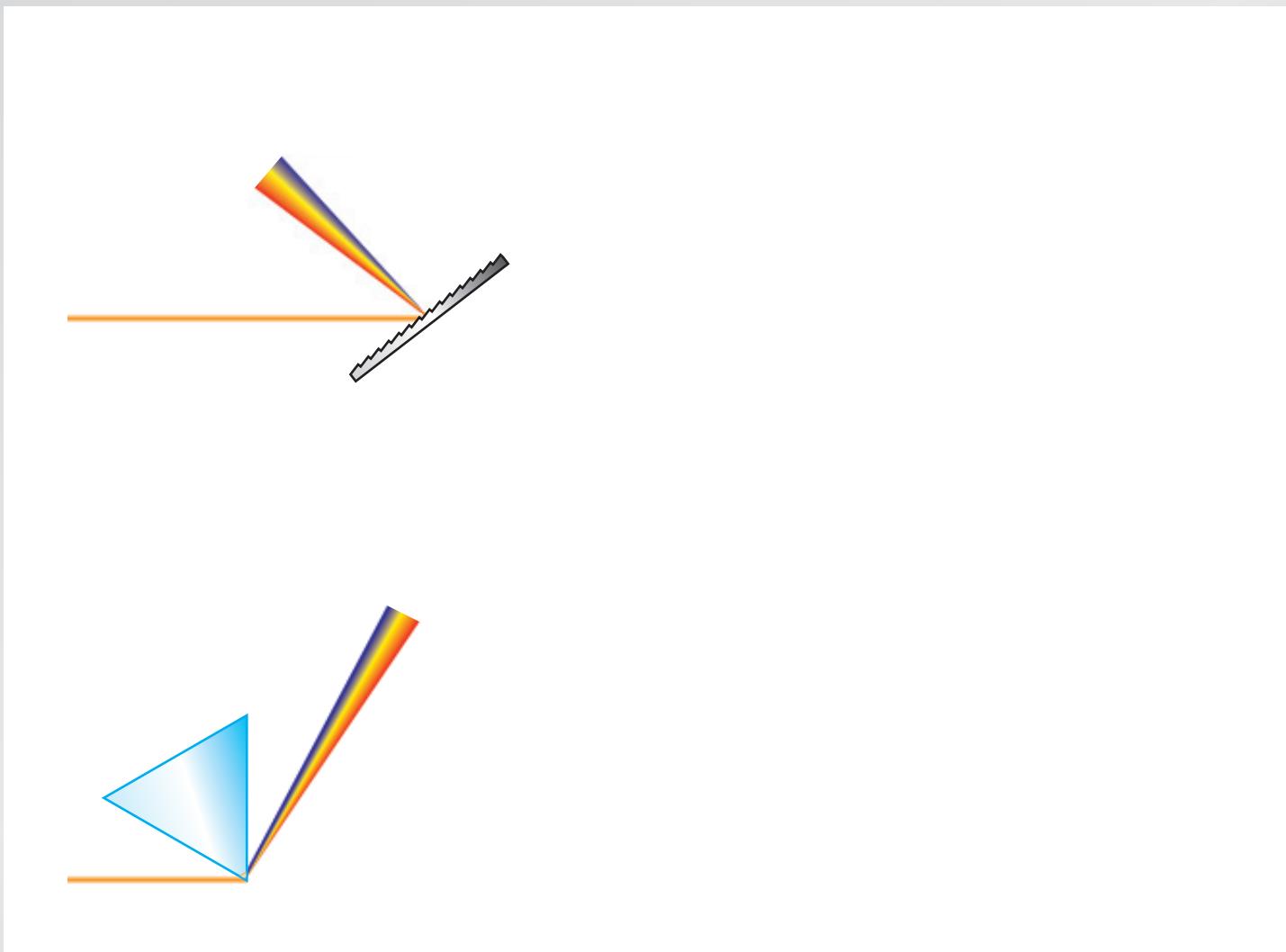
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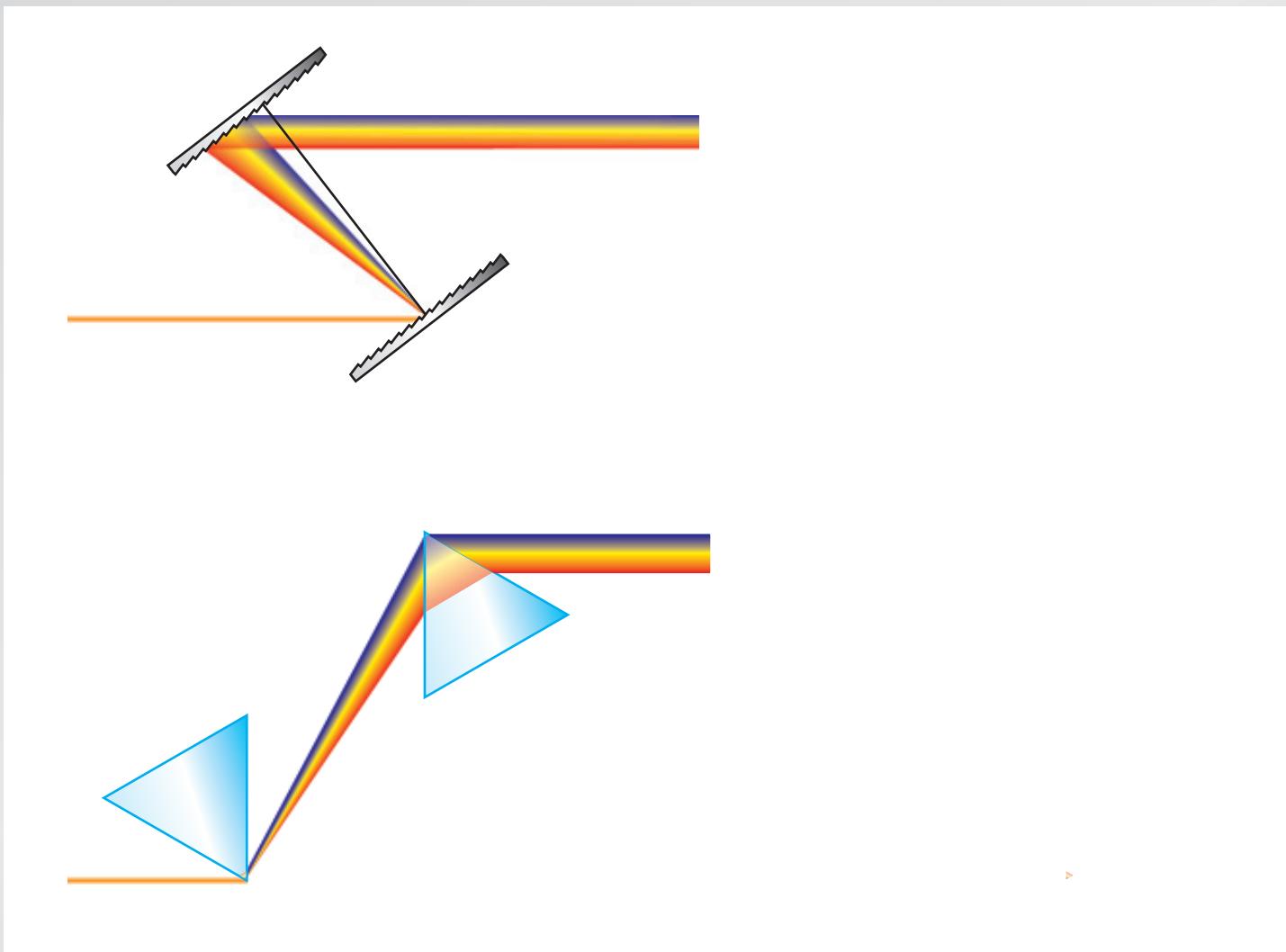


compensate by rearranging spectral components!

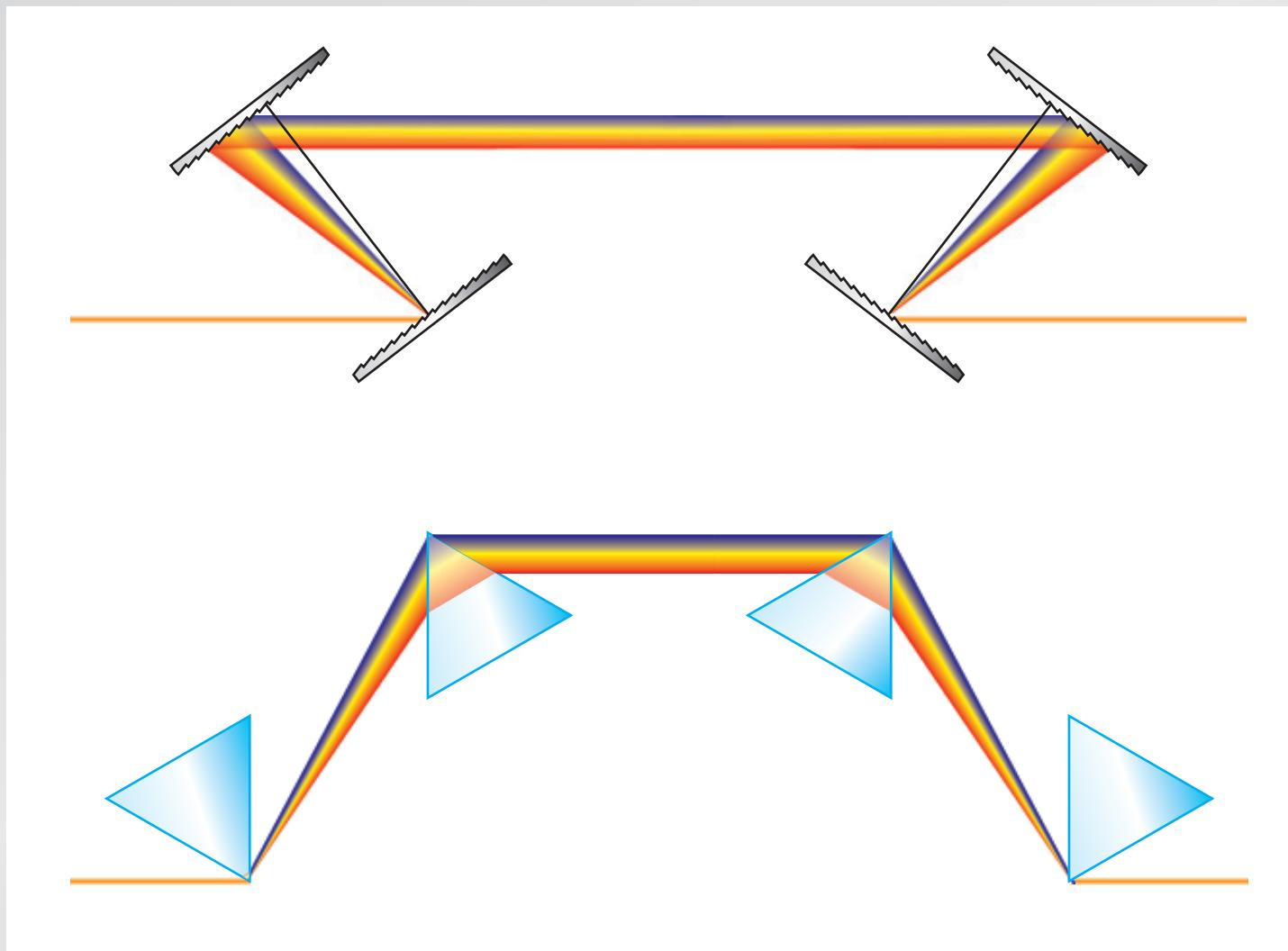
Pulse dispersion compensation



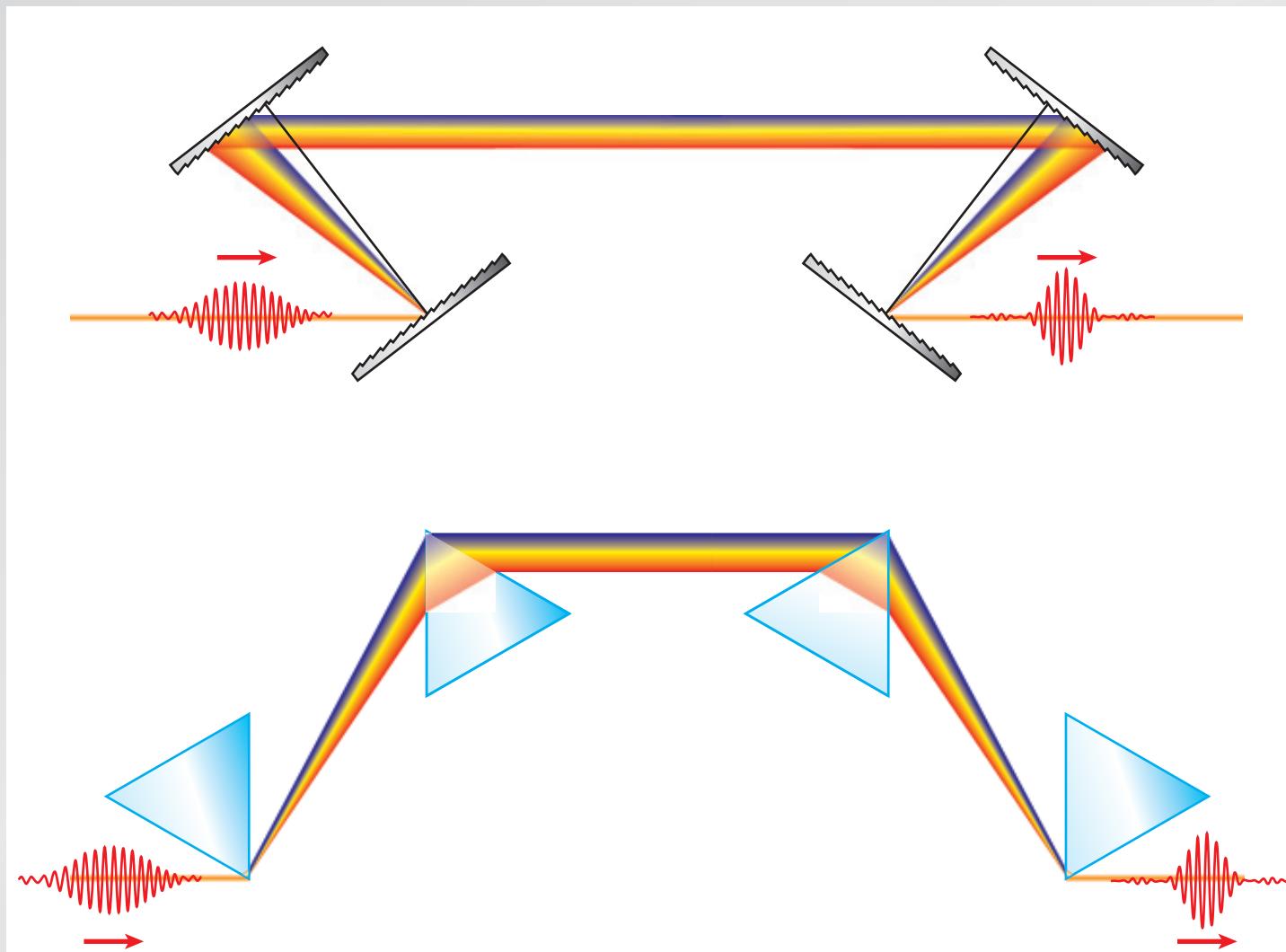
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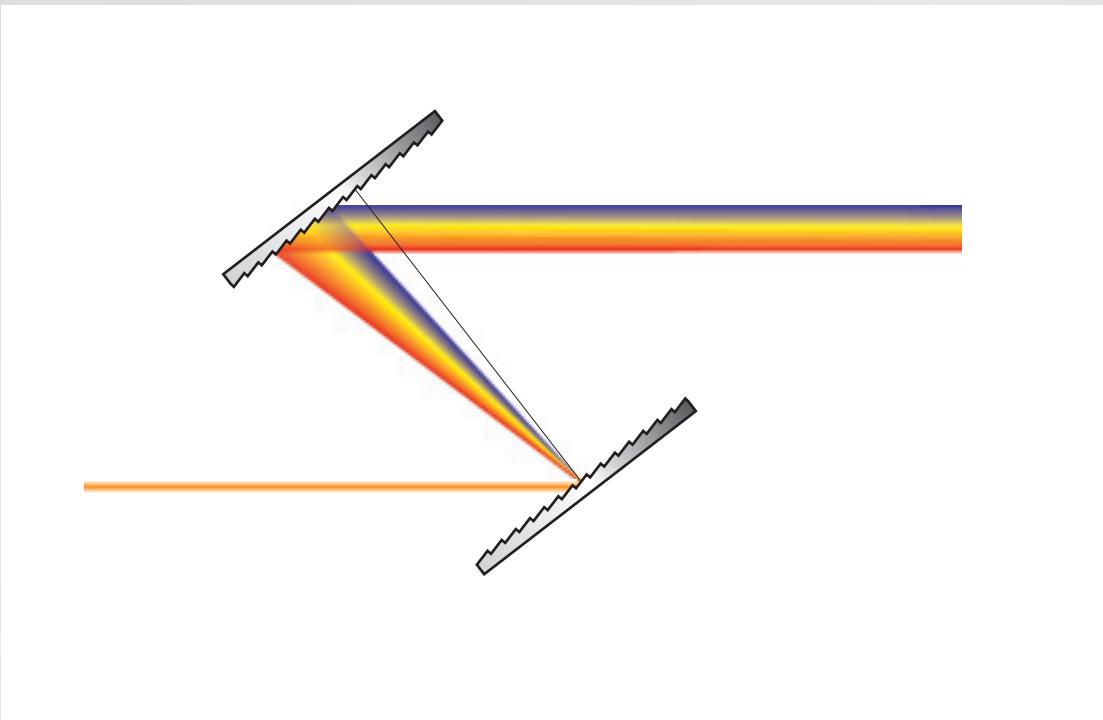
How do these arrangements work?

Pulse dispersion compensation

Does path length difference compensate?

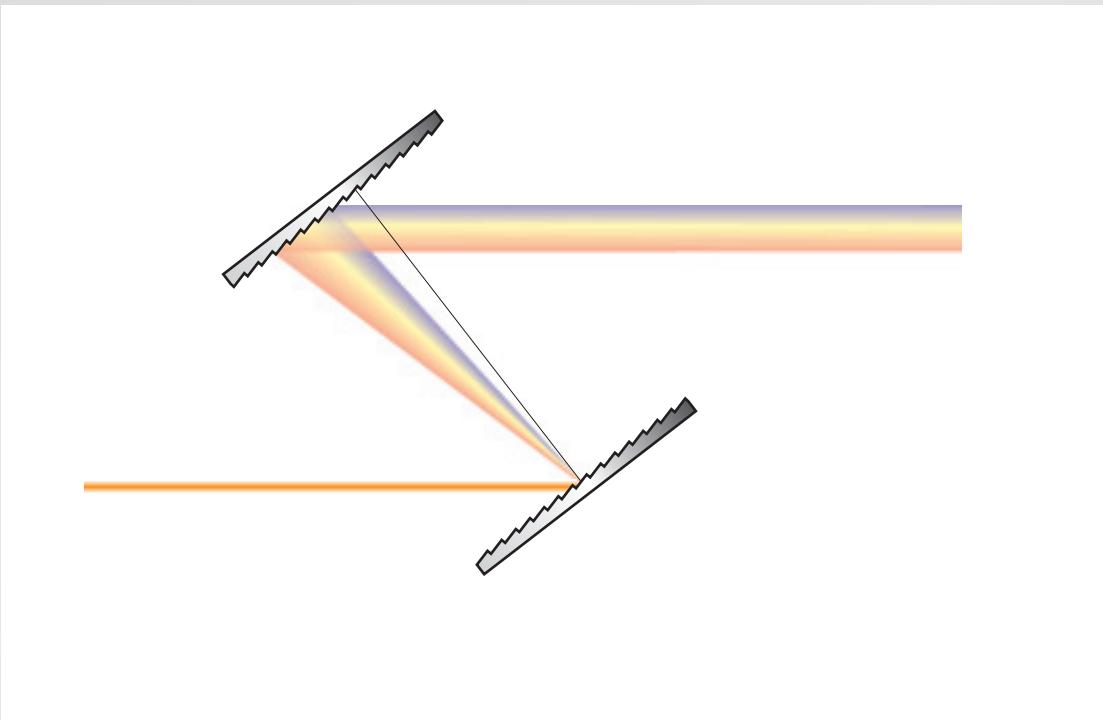
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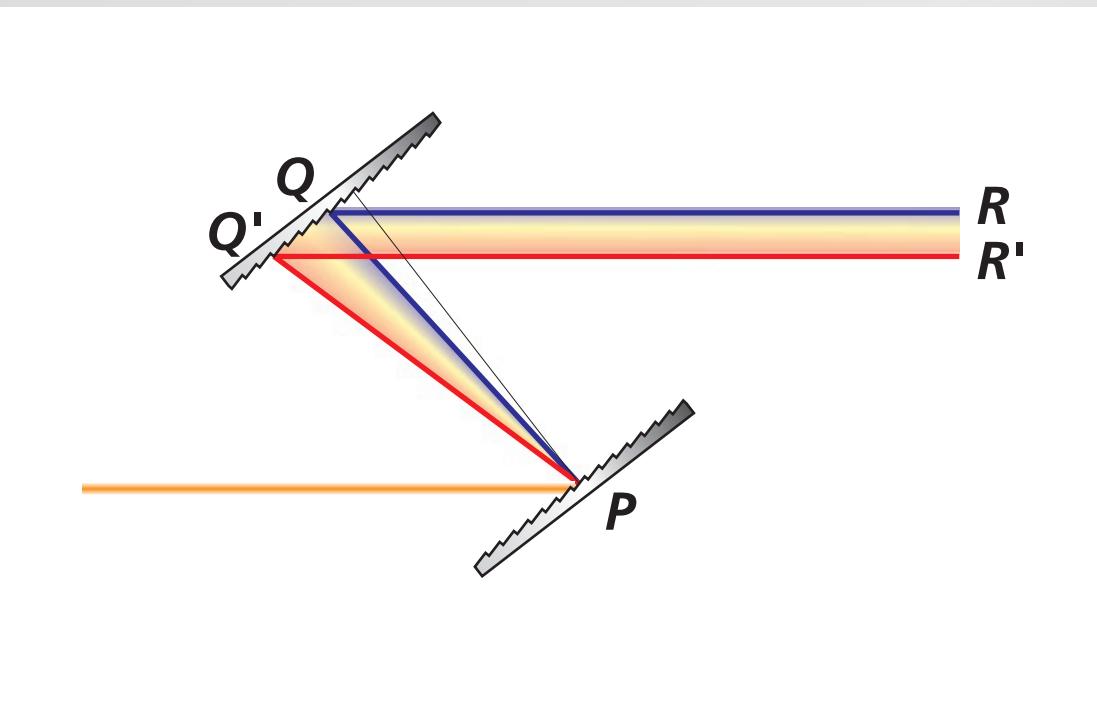
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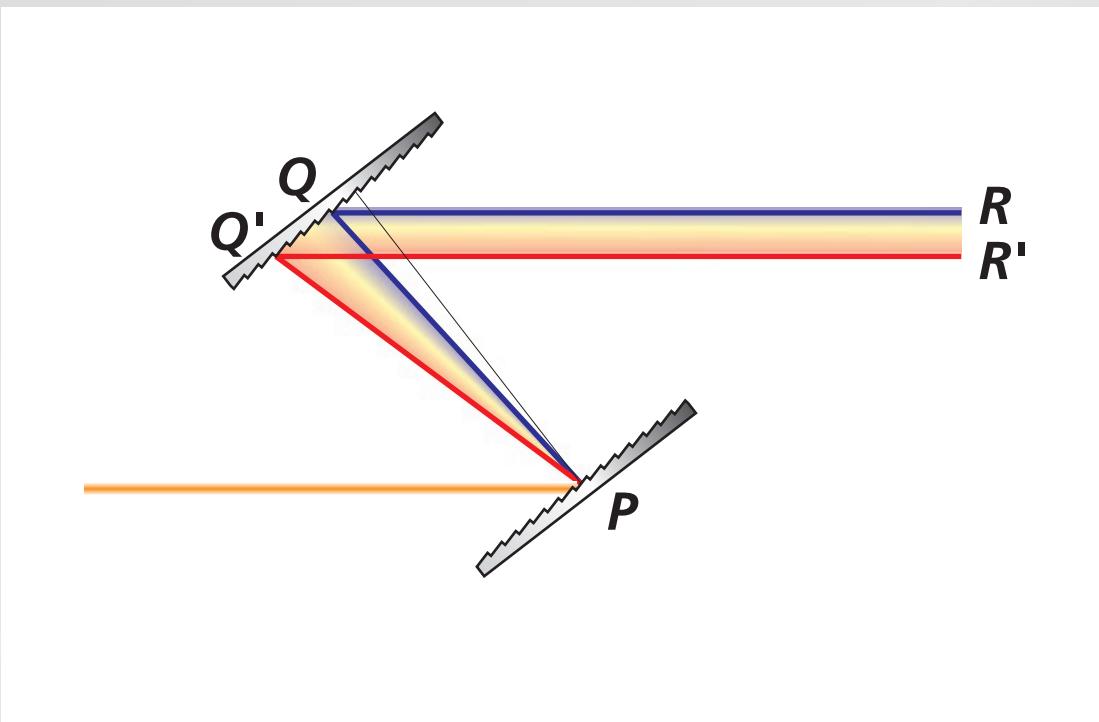
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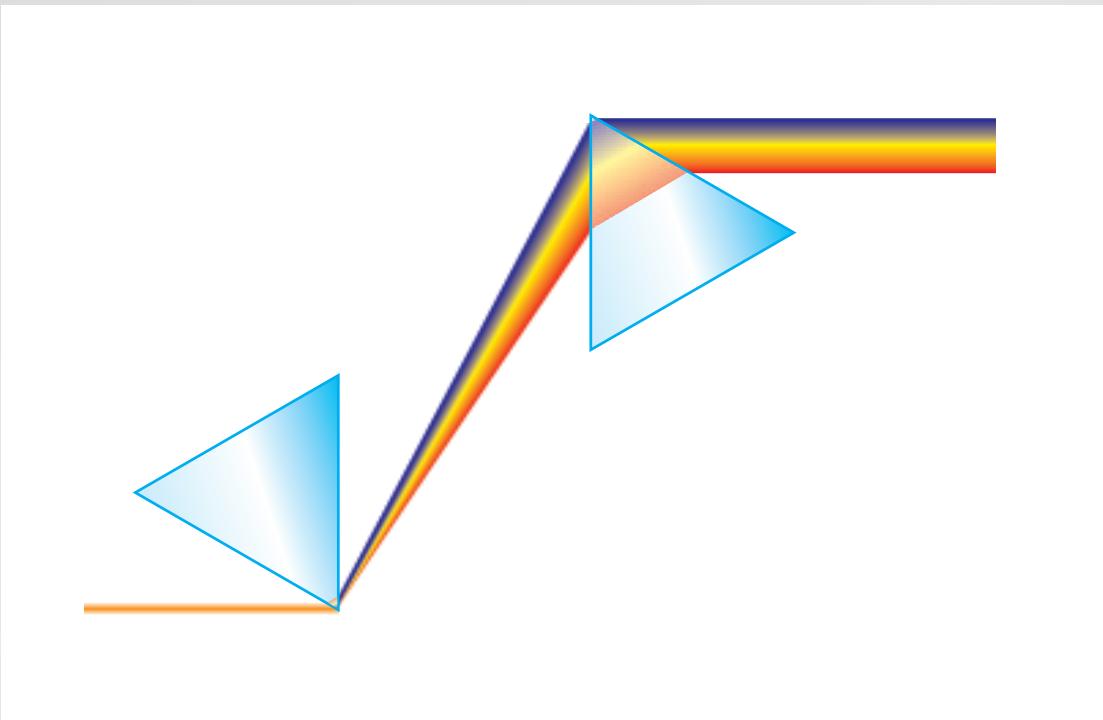
Does path length difference compensate?



grating gives low frequency longer path length!

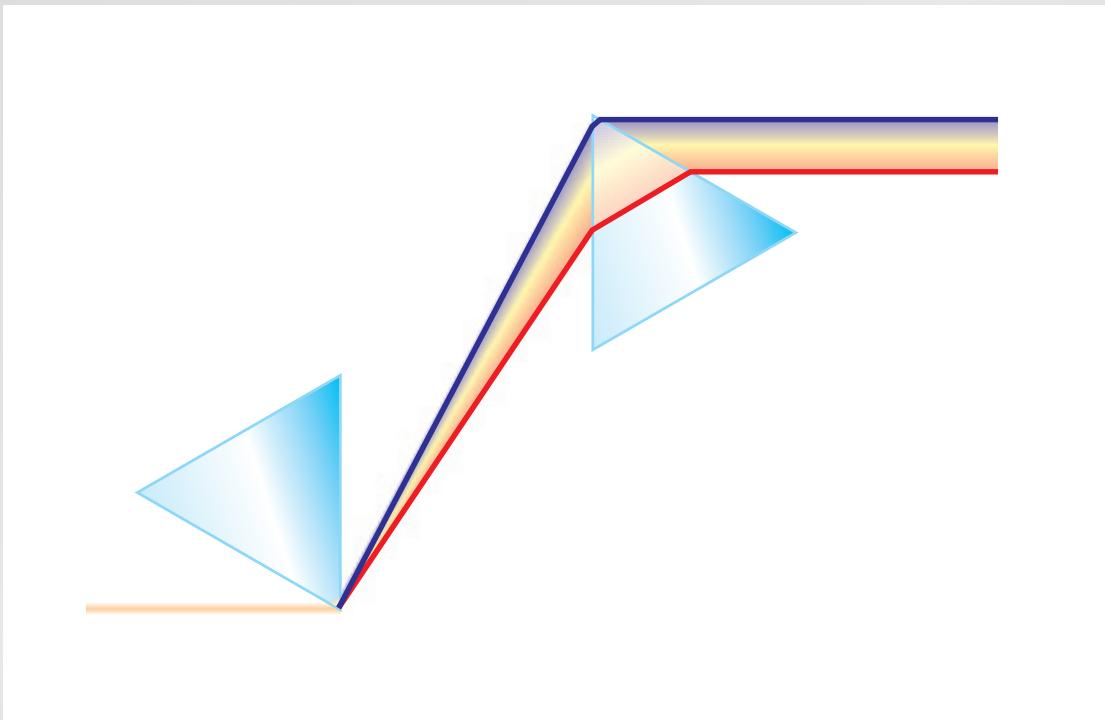
Pulse dispersion compensation

Does path length difference compensate?



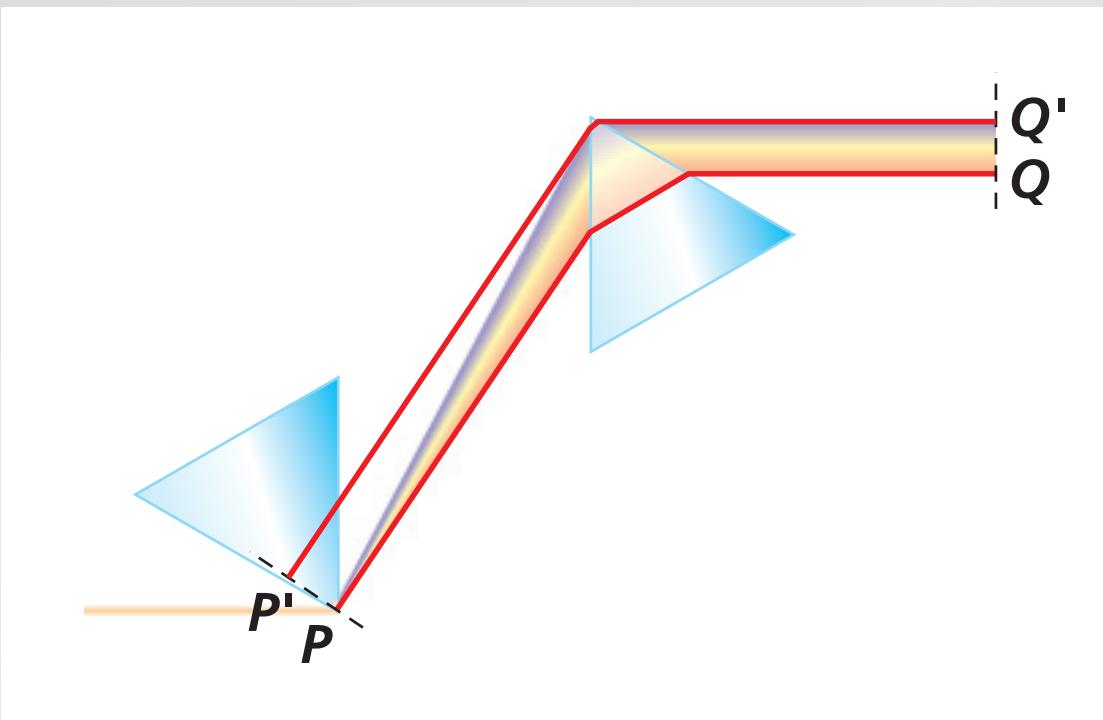
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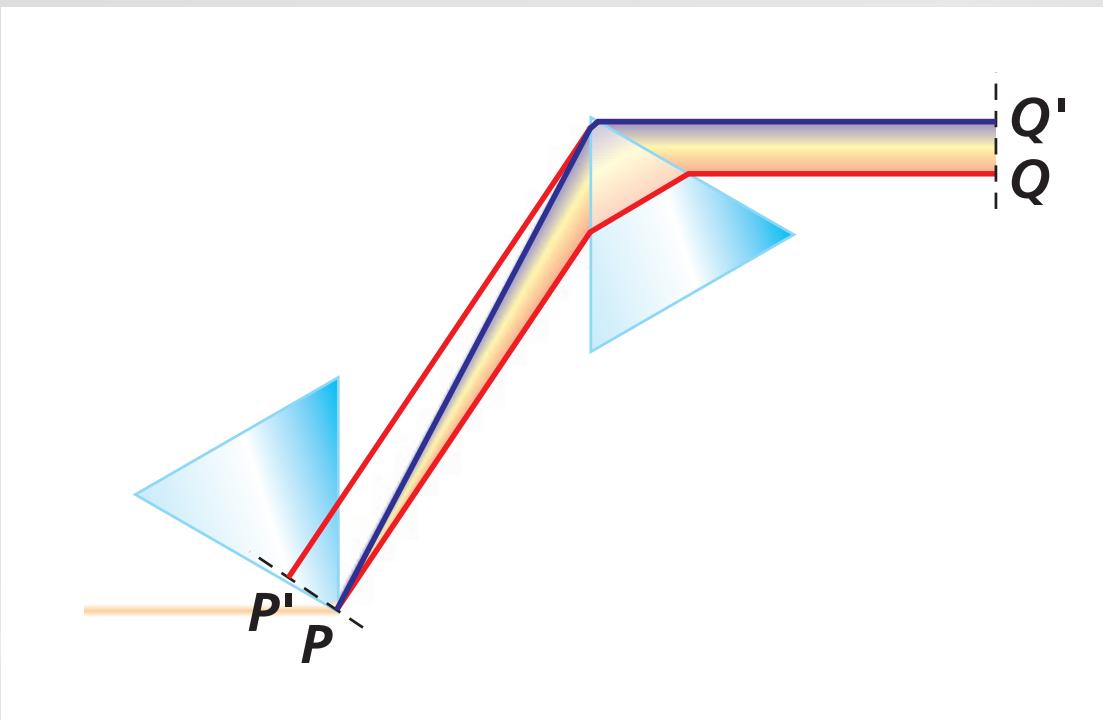
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Does path length difference compensate?



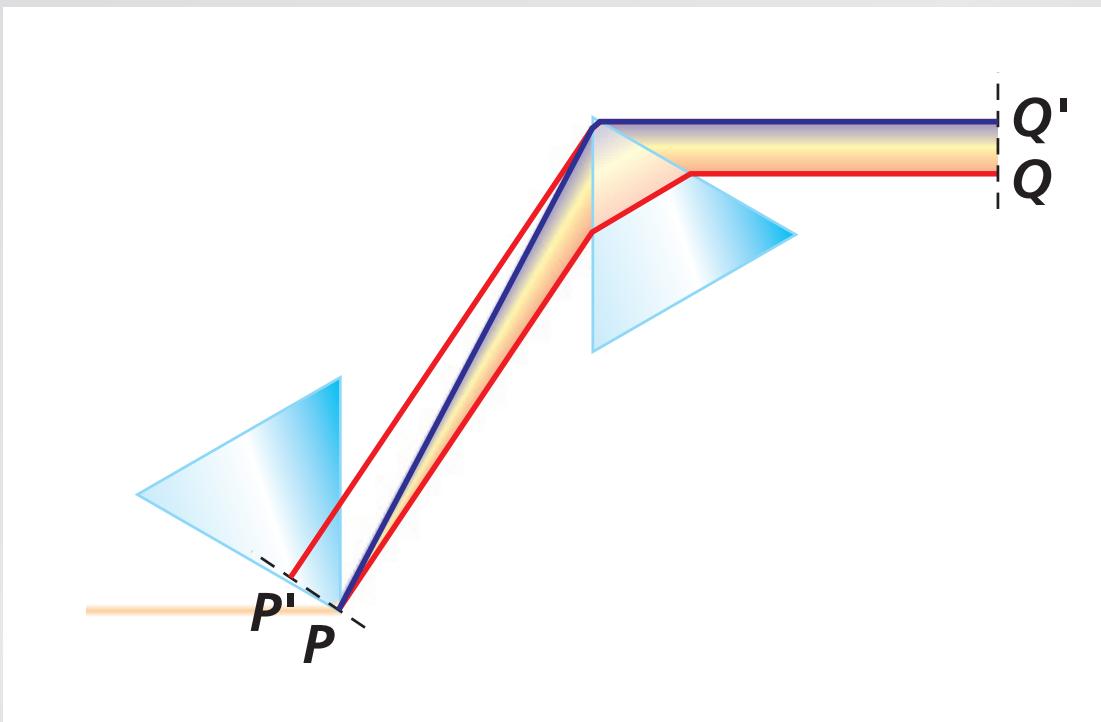
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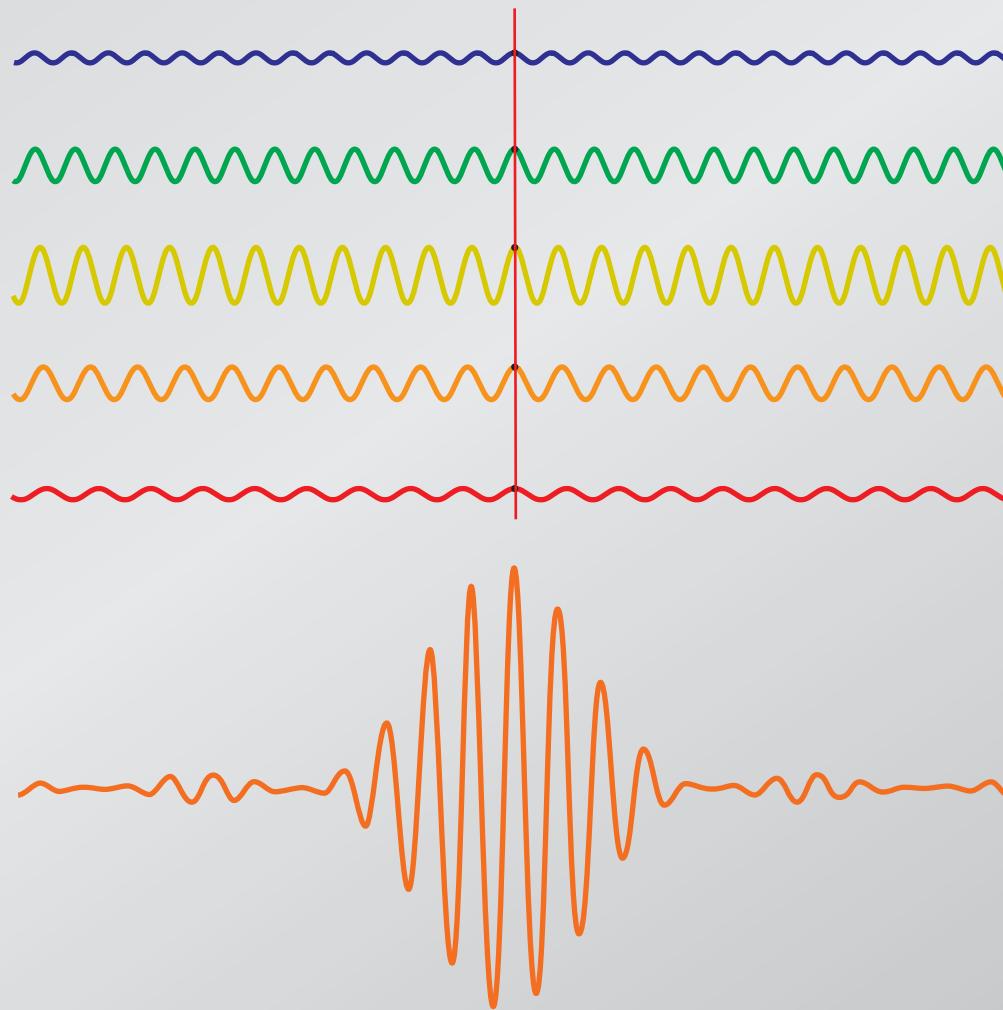
Pulse dispersion compensation

Does path length difference compensate?



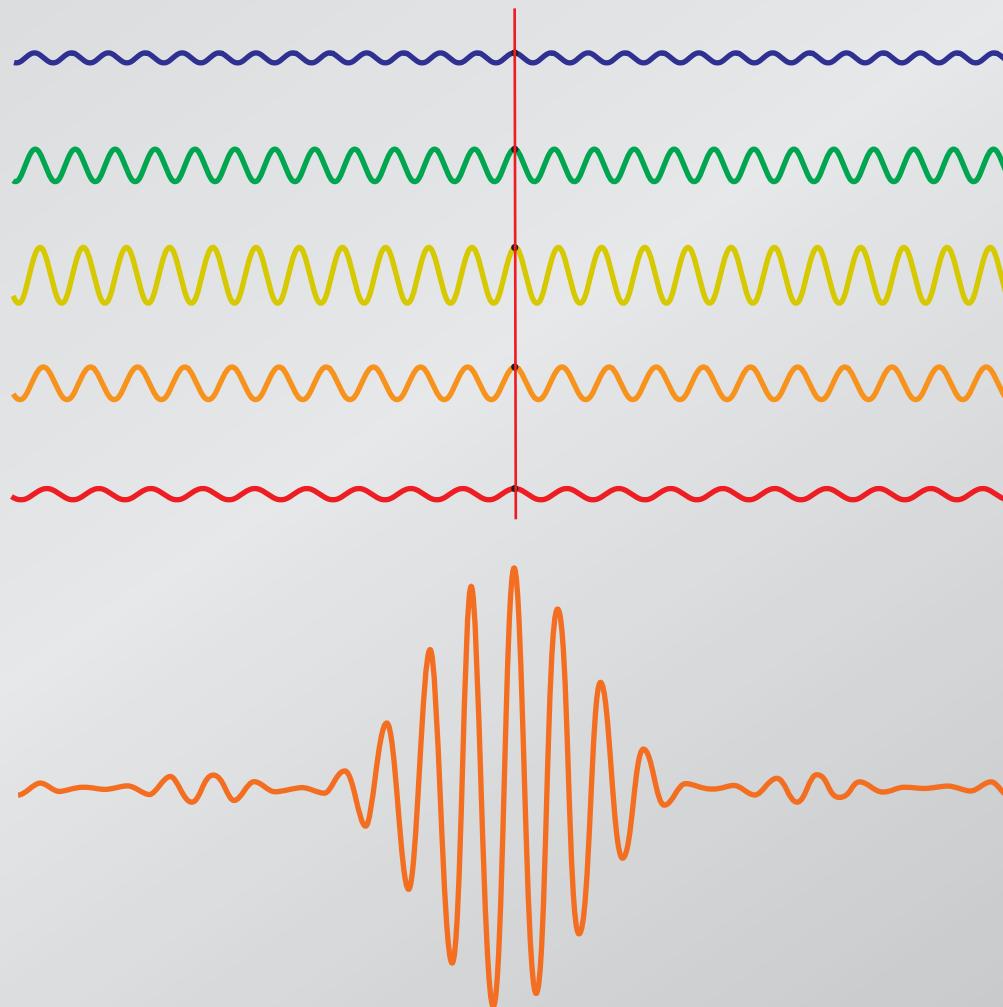
...so prism gives low frequency *shorter* path length!

Pulse dispersion compensation



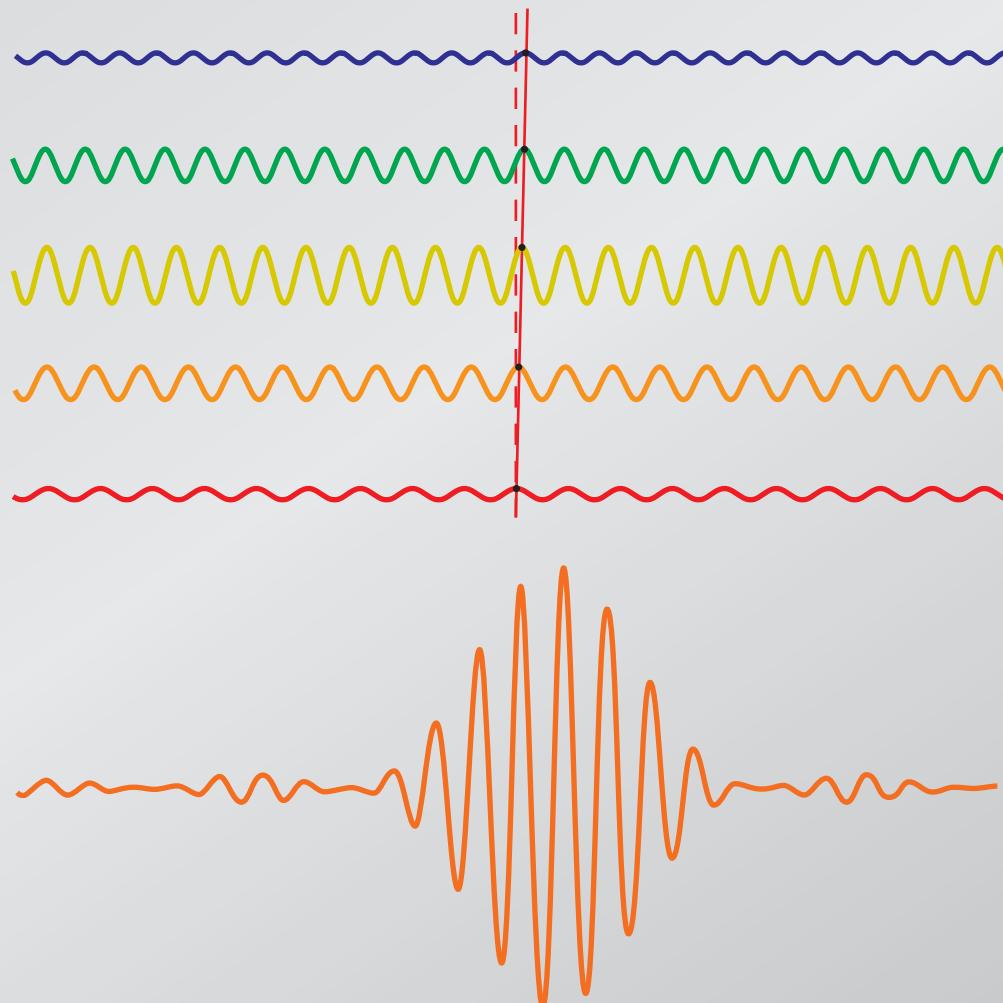
Pulse dispersion compensation

linear dispersion



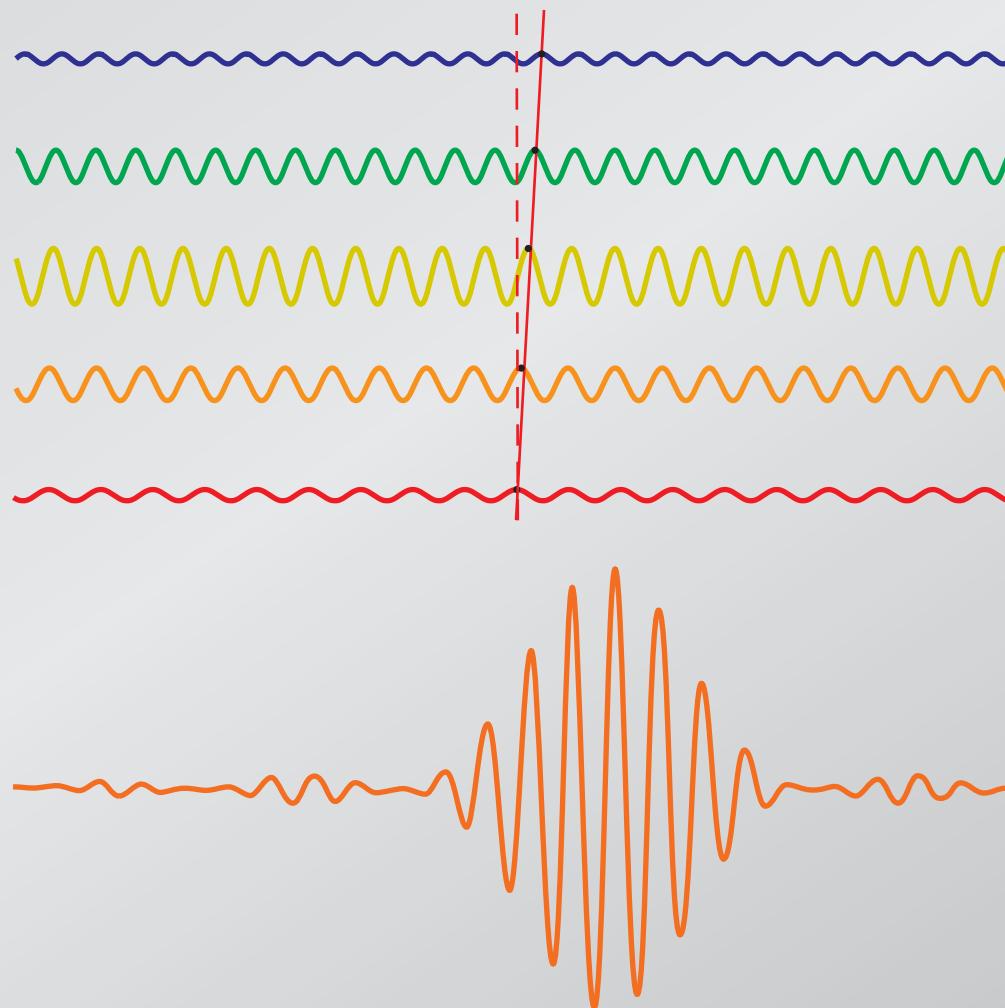
Pulse dispersion compensation

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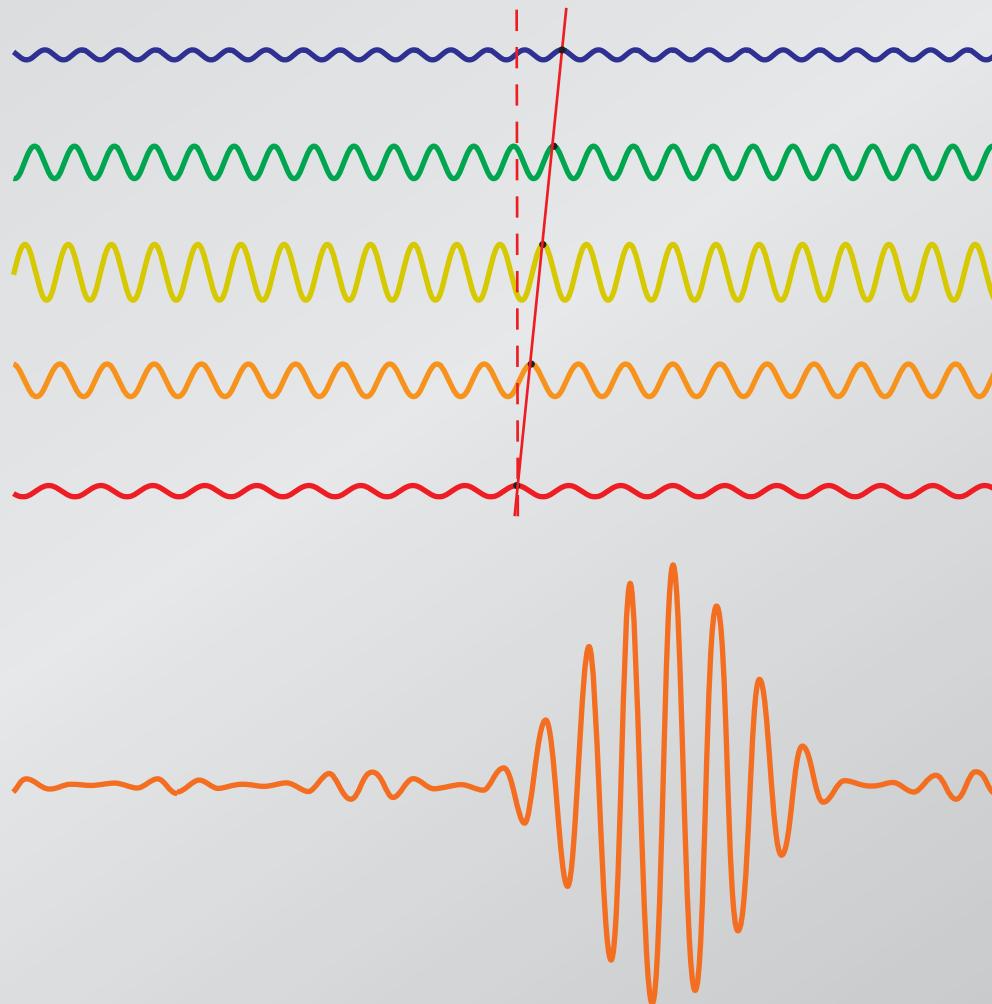
Pulse dispersion compensation

linear dispersion



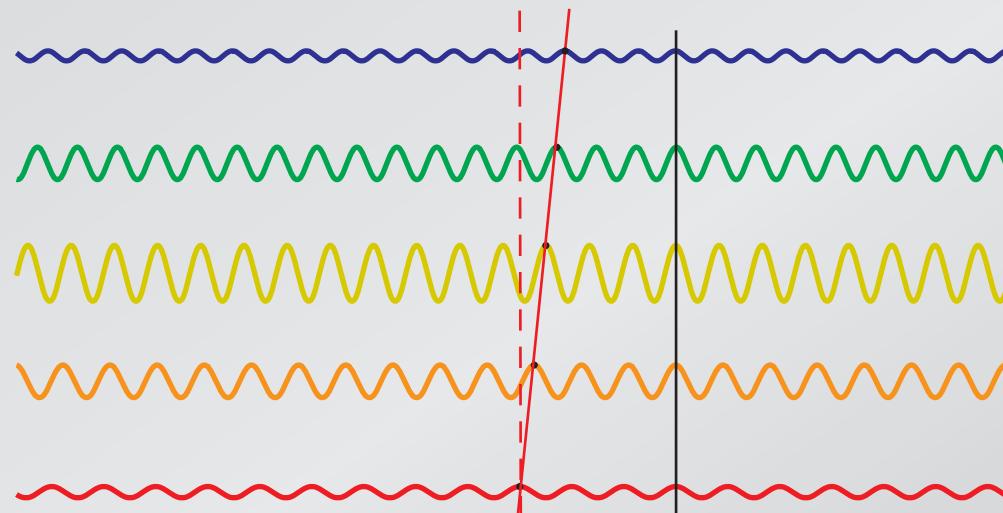
Pulse dispersion compensation

linear dispersion



Pulse dispersion compensation

linear dispersion

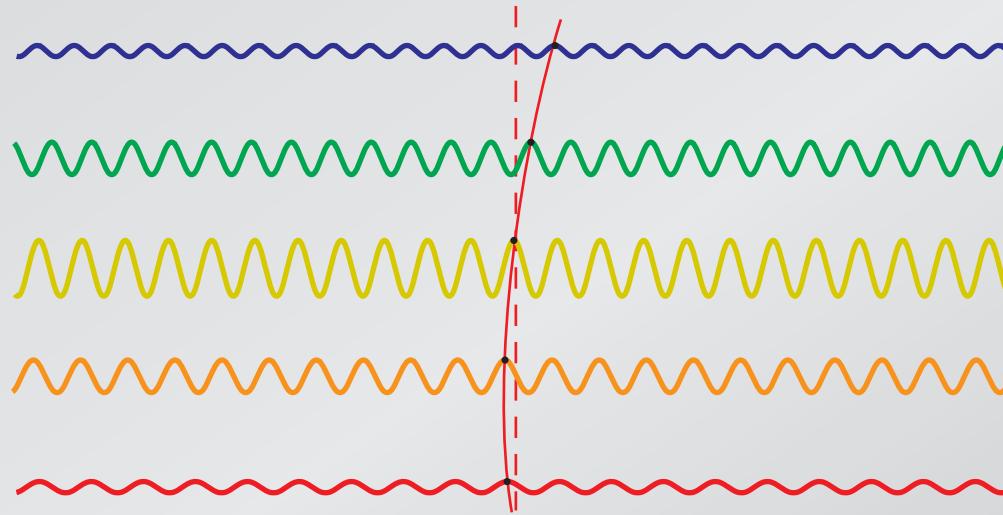


$$\frac{d\phi}{d\omega} = \text{constant}$$

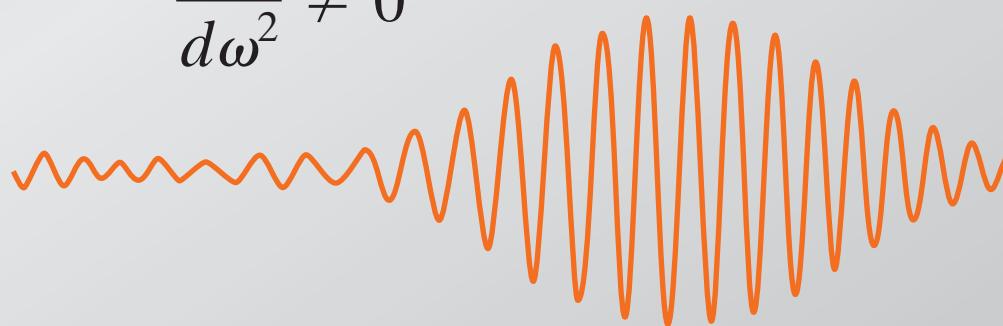


Pulse dispersion compensation

only *nonlinear* dispersion changes pulse shape!



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



Outline

- propagation of pulses
- nonlinear optics
- femtosecond micromachining

Nonlinear optics

Linear optics:

$$\vec{P} = \chi \vec{E}$$

Nonlinear optics

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Nonlinear polarization:

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

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$P^{(2)} \approx P^{(1)}$ **when** $E = E_{at} \approx \frac{e}{a}$, **and so** $\chi^{(n)} \approx \frac{\chi^{(1)}}{E_{at}^{n-1}}$.

Nonlinear optics

Nonlinear polarization can drive new field:

$$\nabla^2 \vec{E} + \frac{n^2}{c^2} \frac{\partial^2 \vec{E}}{\partial t^2} = \frac{4\pi}{c^2} \frac{\partial^2 \vec{P}}{\partial t^2}$$

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But even terms disappear in media with inversion symmetry!

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

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

Nonlinear optics

Consider oscillating electric field:

$$E(t) = E e^{i\omega t} + \text{c.c.}$$

Nonlinear optics

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Nonlinear optics

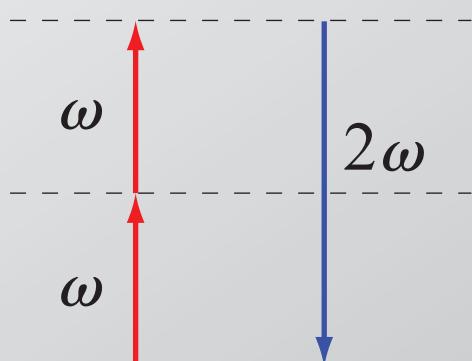
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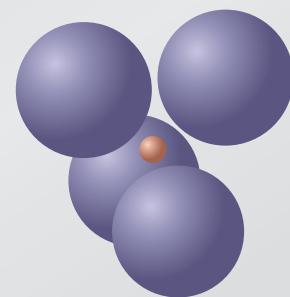
Physical interpretation:



Nonlinear optics

Linear response:

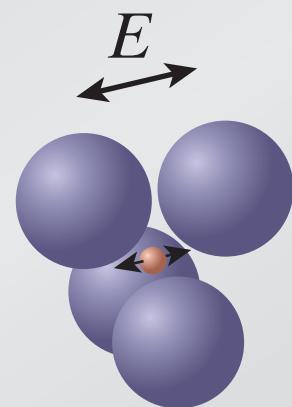
$$\vec{P} = \chi \vec{E}$$



Nonlinear optics

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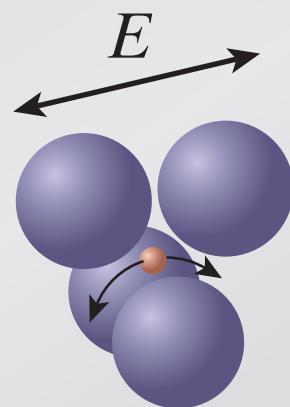
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Nonlinear optics

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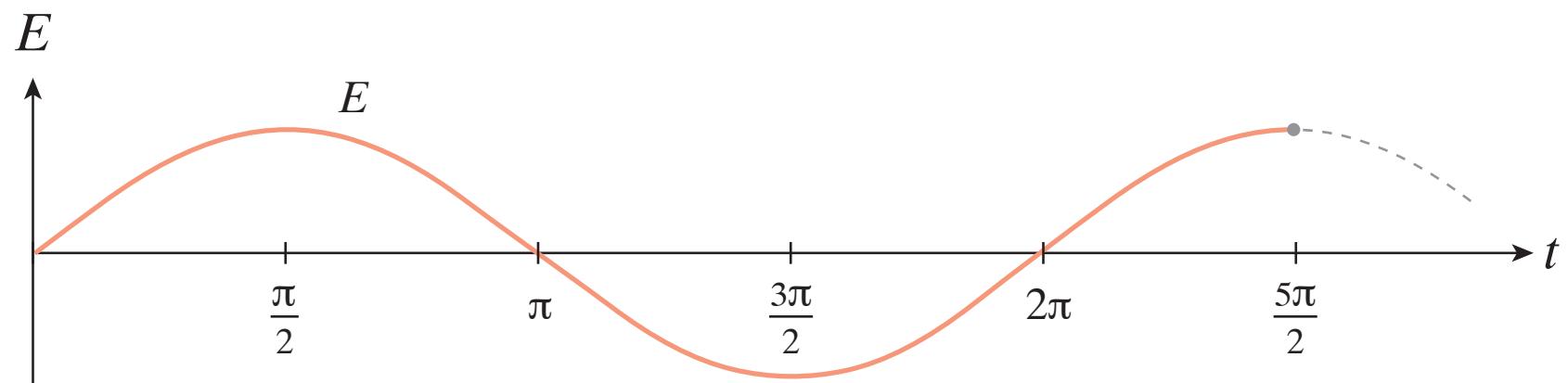
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Nonlinear optics

Nonlinear response:

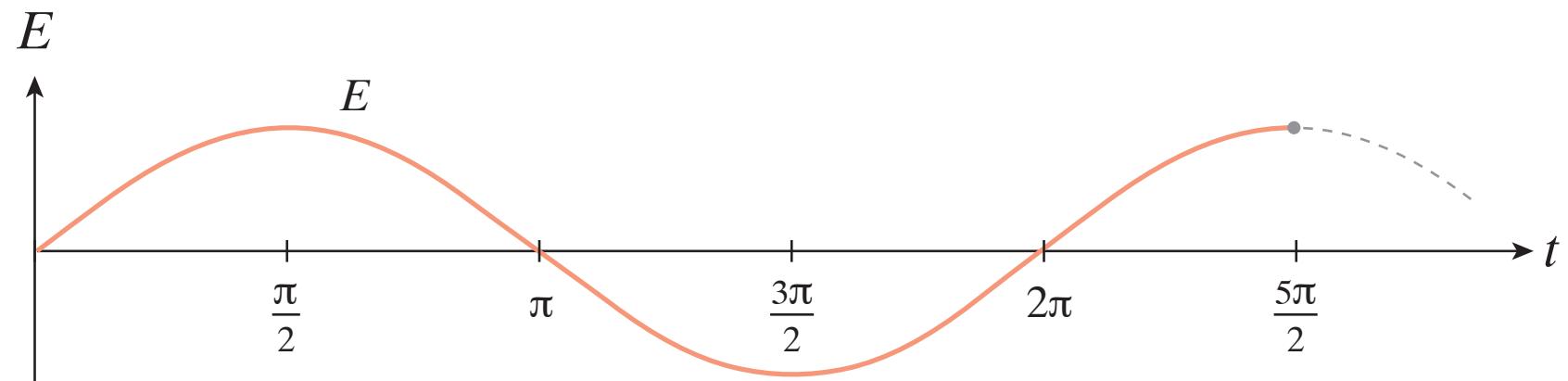
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Nonlinear optics

Nonlinear response:

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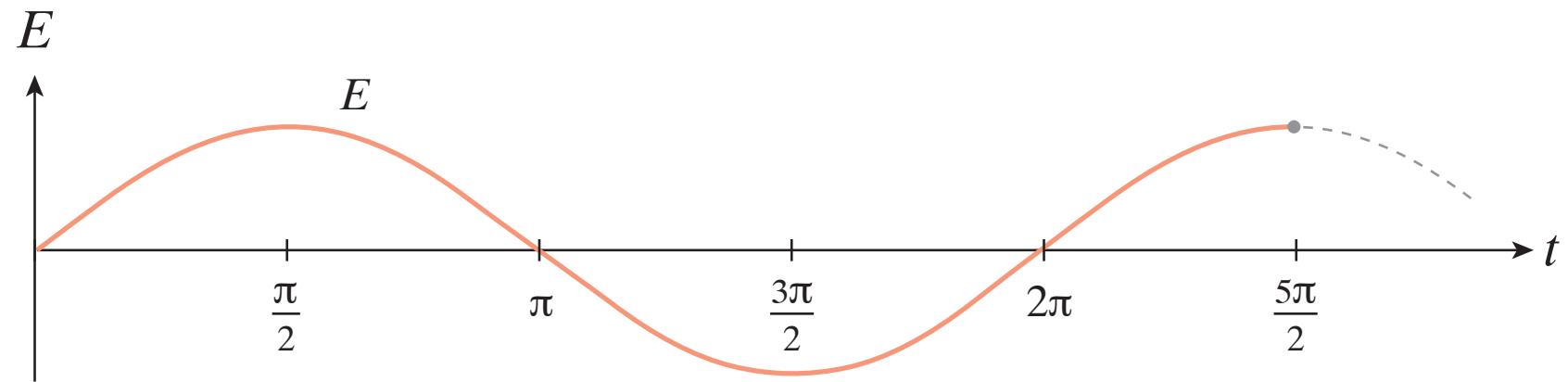
$$E = 0$$



Nonlinear optics

Nonlinear response:

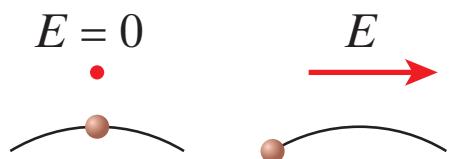
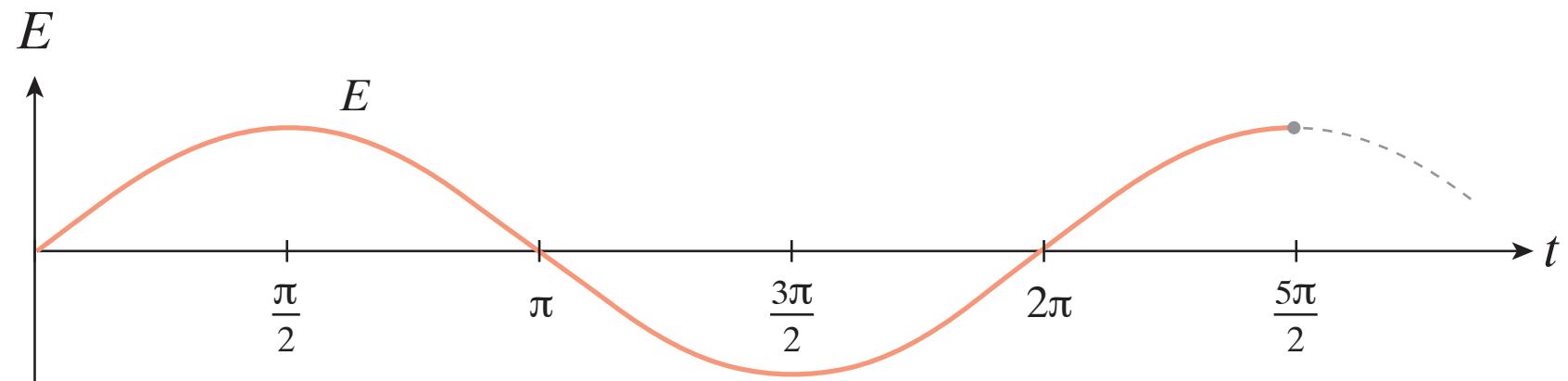
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Nonlinear optics

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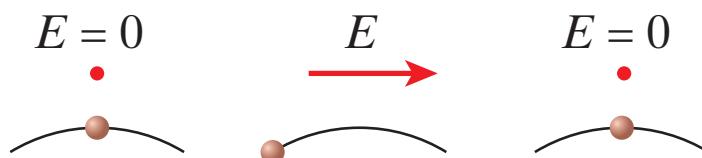
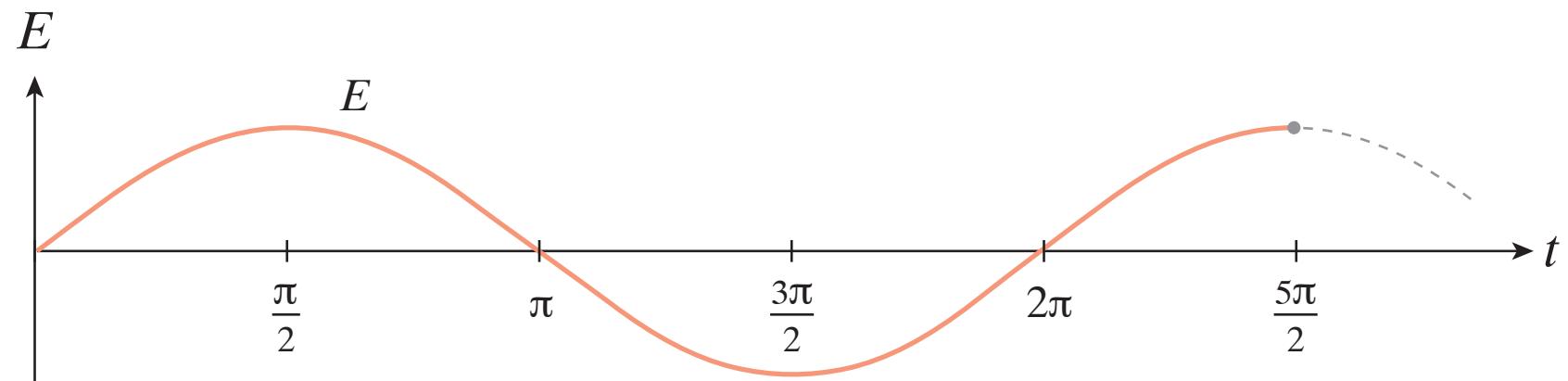
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Nonlinear optics

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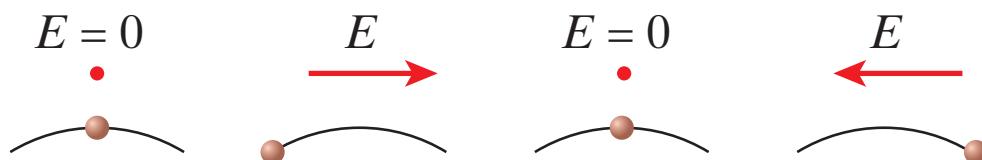
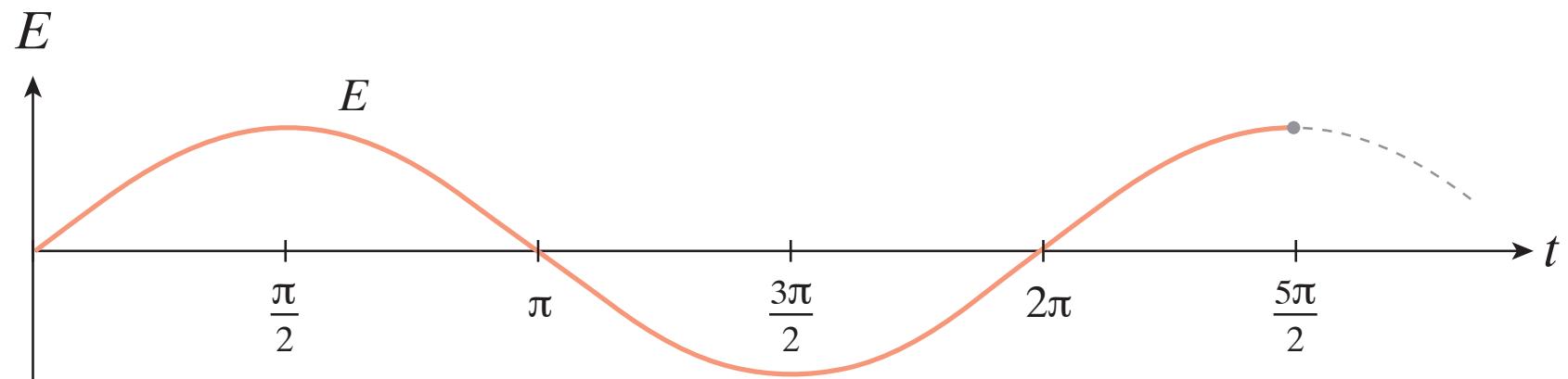
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Nonlinear optics

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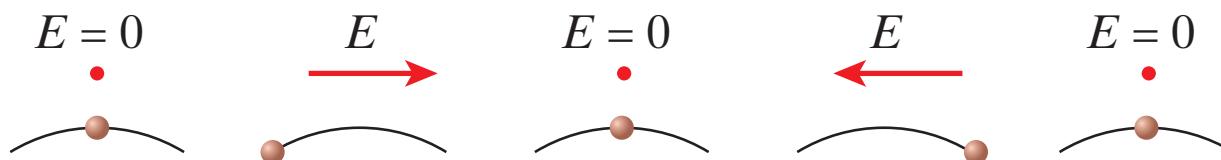
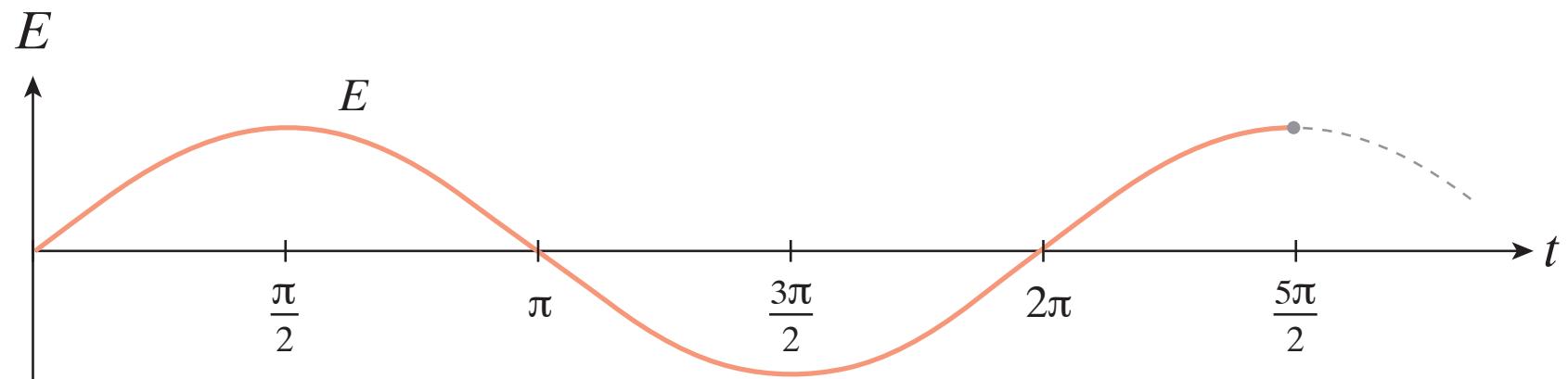
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Nonlinear optics

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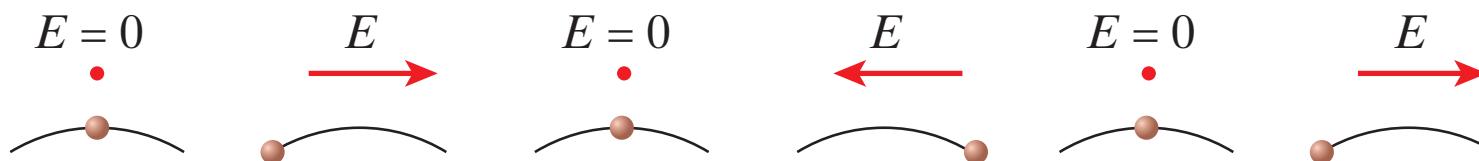
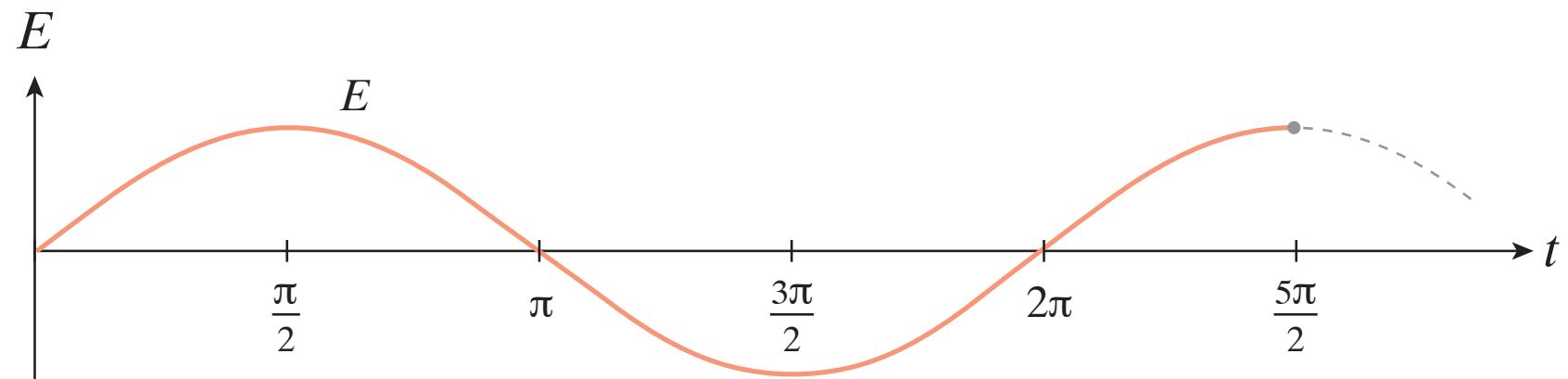
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Nonlinear optics

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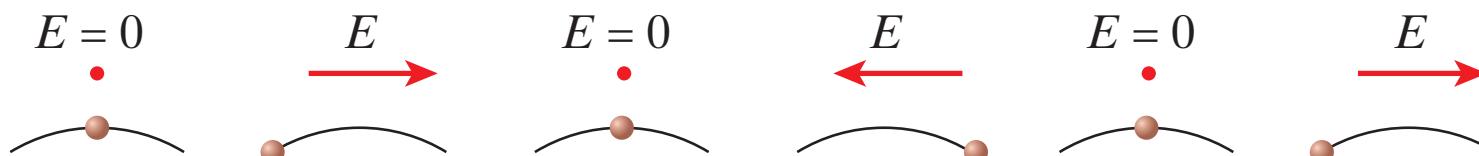
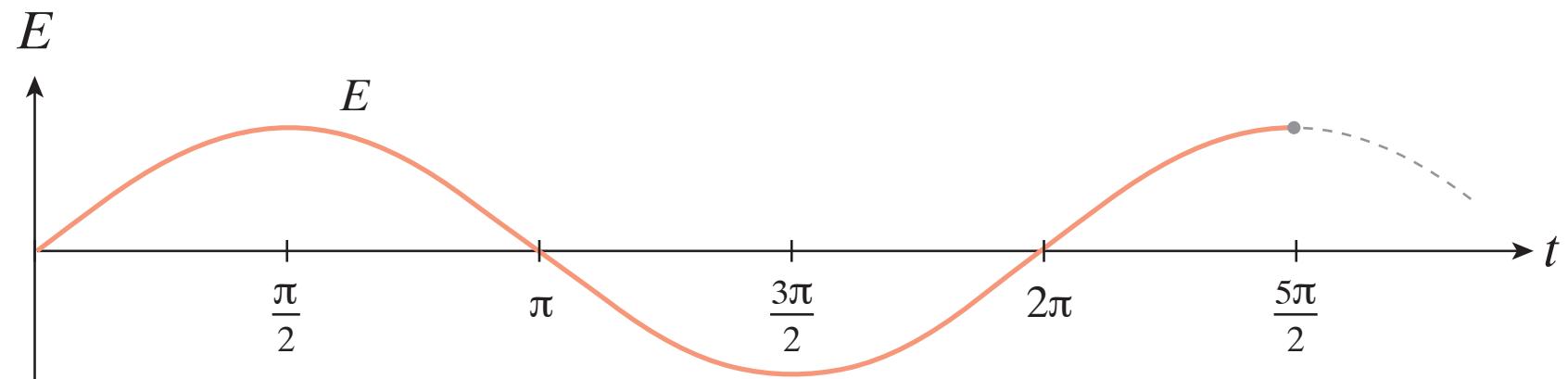
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Nonlinear optics

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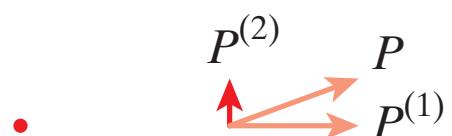
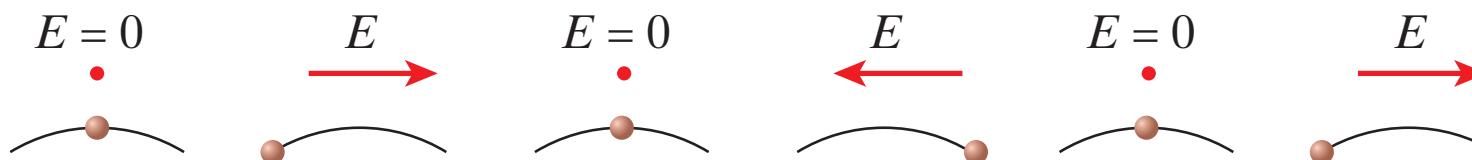
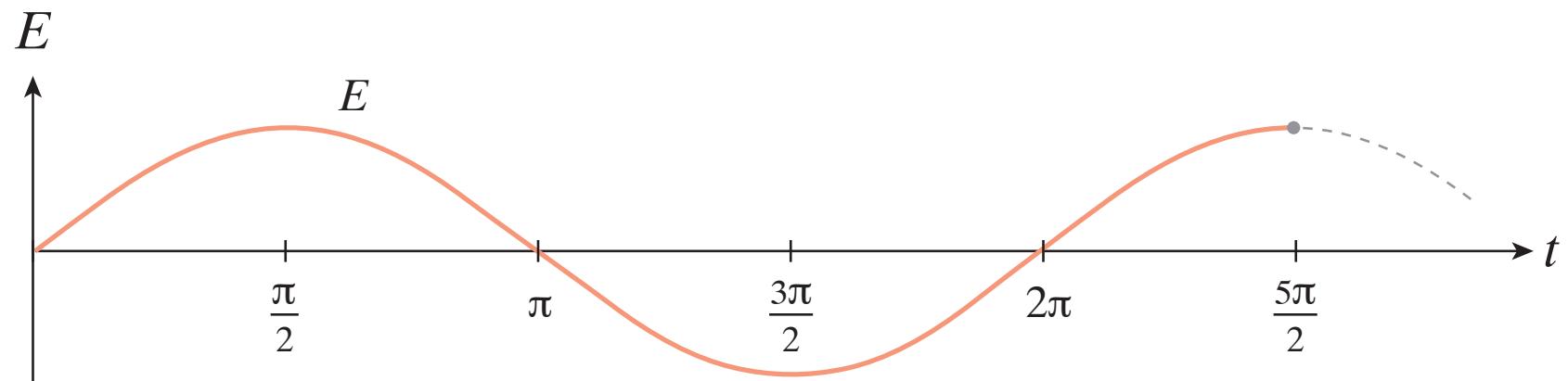


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Nonlinear optics

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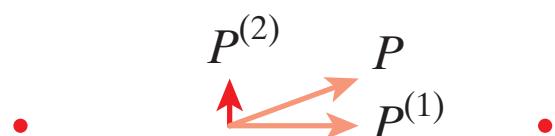
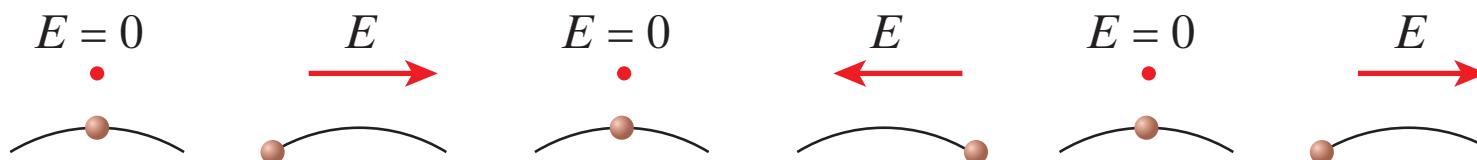
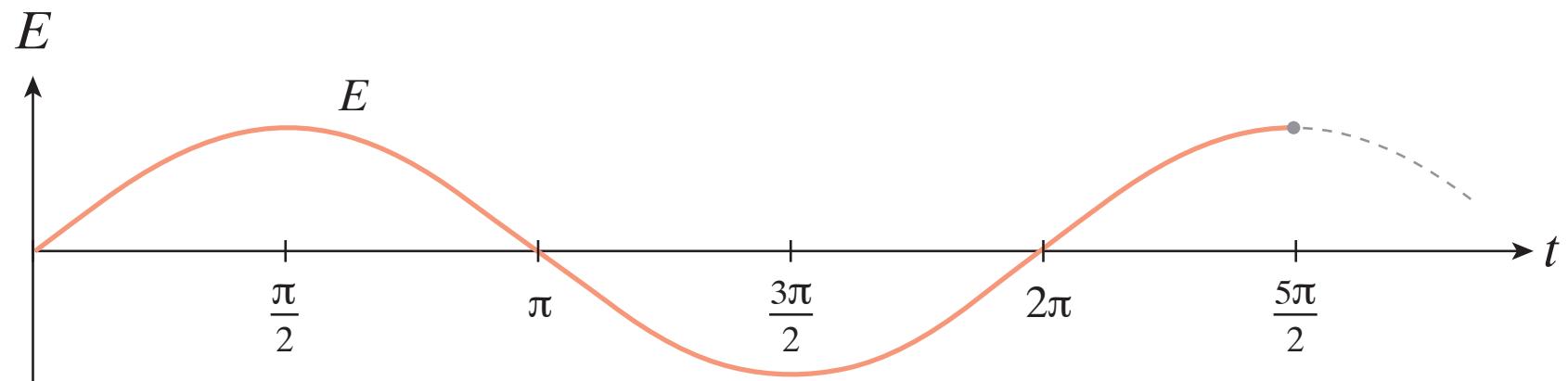
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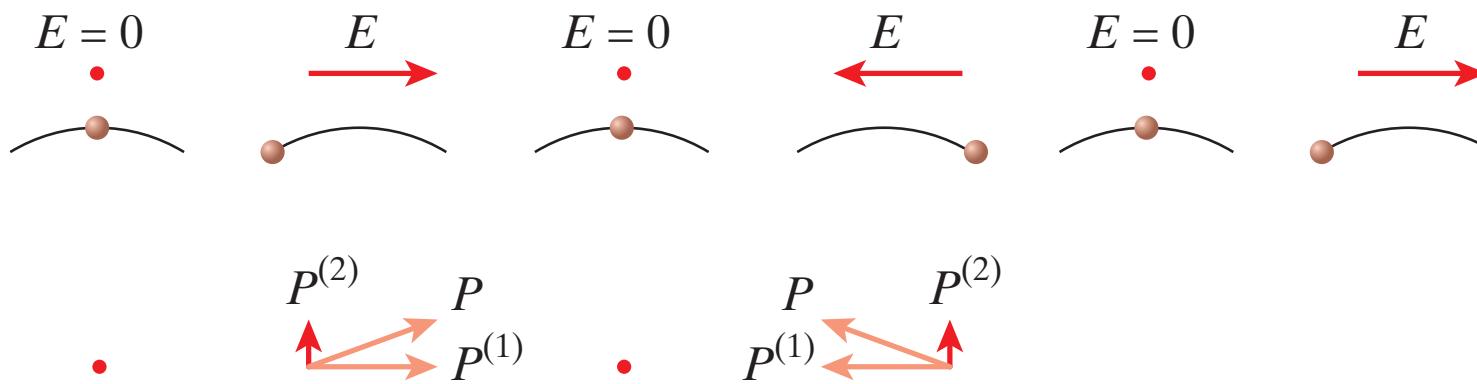
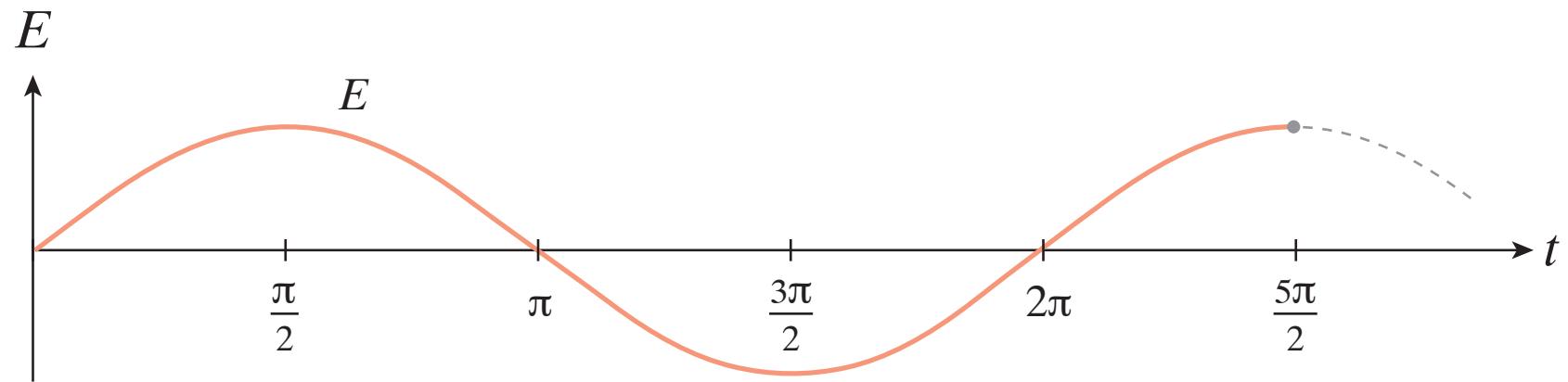
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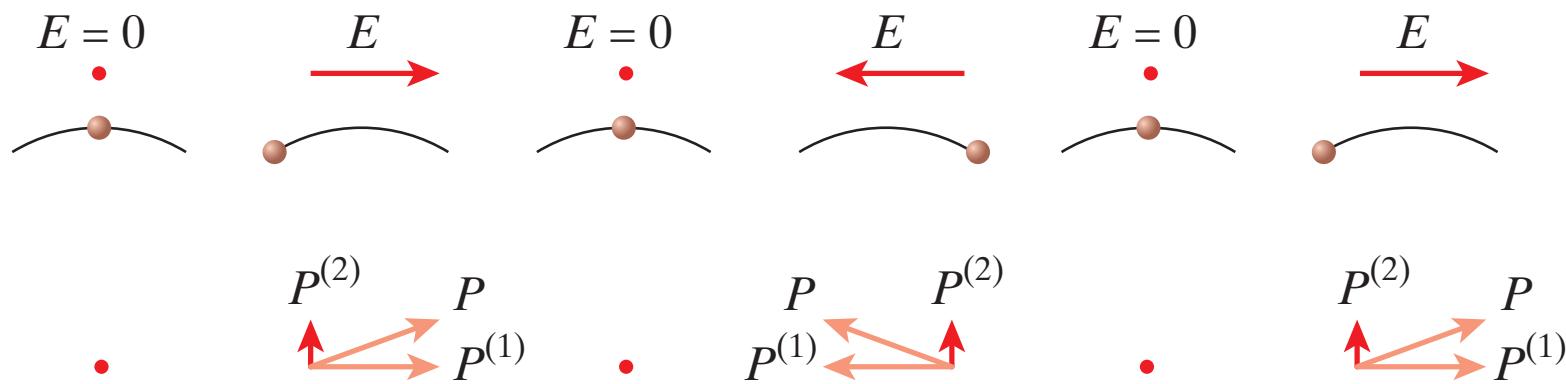
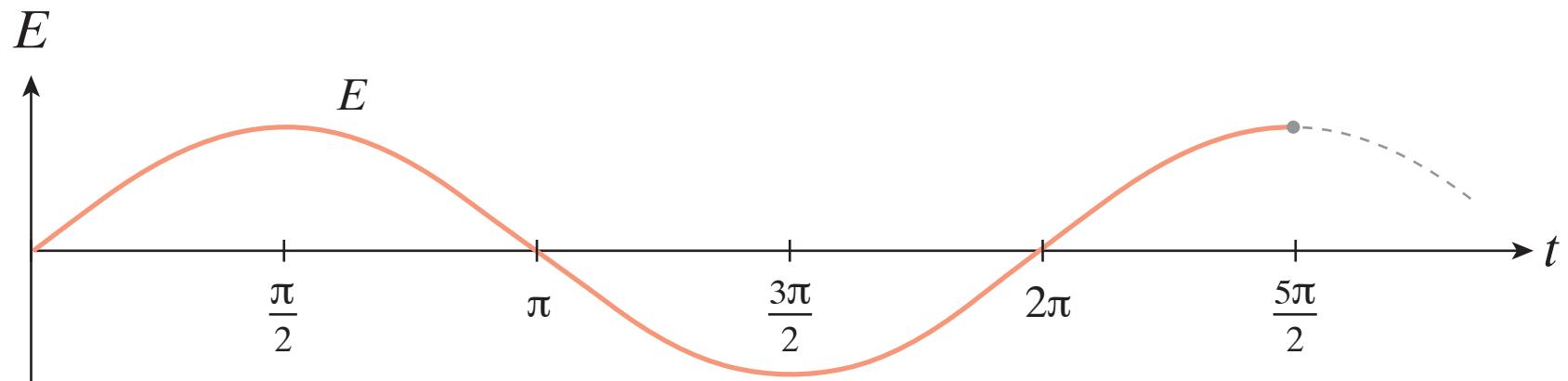
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Nonlinear optics

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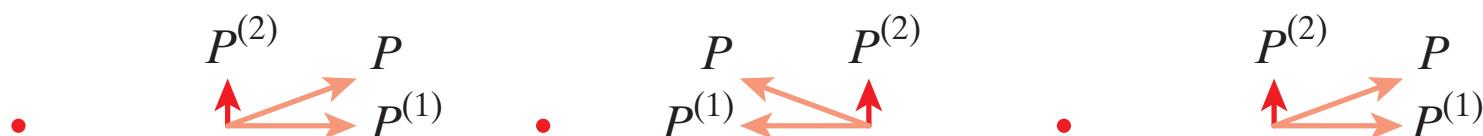
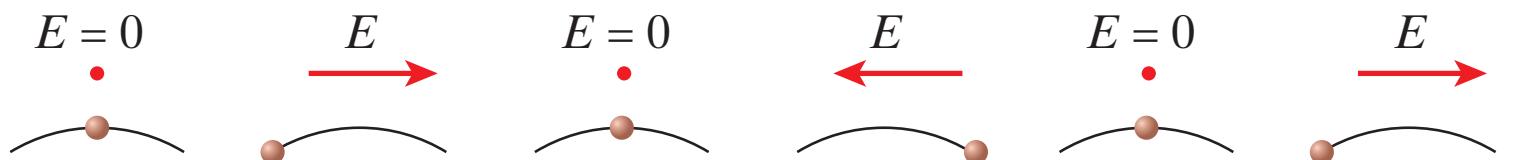
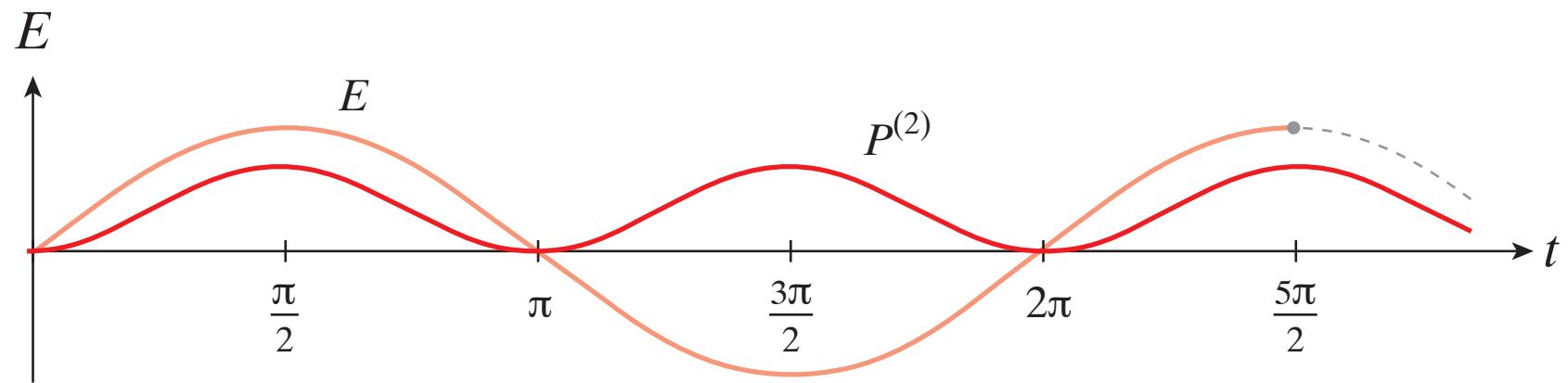
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Nonlinear optics

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Nonlinear optics

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Q: Silicon atoms are arranged in this way. Does bulk silicon generate second harmonic?



1. Yes, silicon is not centrosymmetric (as the unit cell shows)
2. No, the crystal as a whole is centrosymmetric
3. No, any radiation at the second harmonic is absorbed
4. Other

Nonlinear optics

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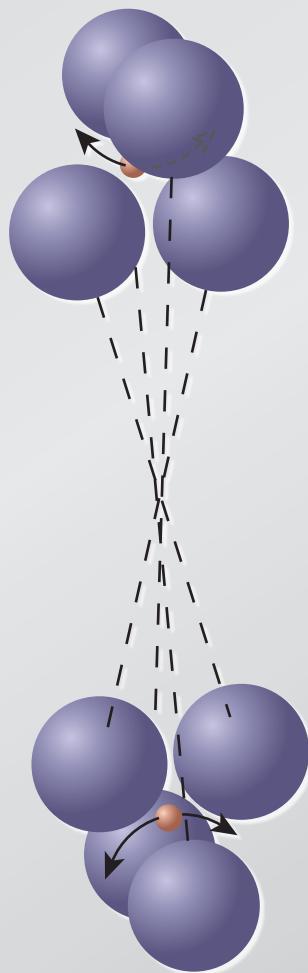
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Nonlinear optics

Third-order polarization: $P^{(3)}(t) = \chi^{(3)} E^3(t)$

Nonlinear optics

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3 frequencies, 3 terms + c.c.: complicated! In general

$$\cos^3 \omega t = \frac{1}{4} \cos 3\omega t + \frac{3}{4} \cos \omega t$$

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Intensity dependent term at fundamental frequency:

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

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Nonlinear optics

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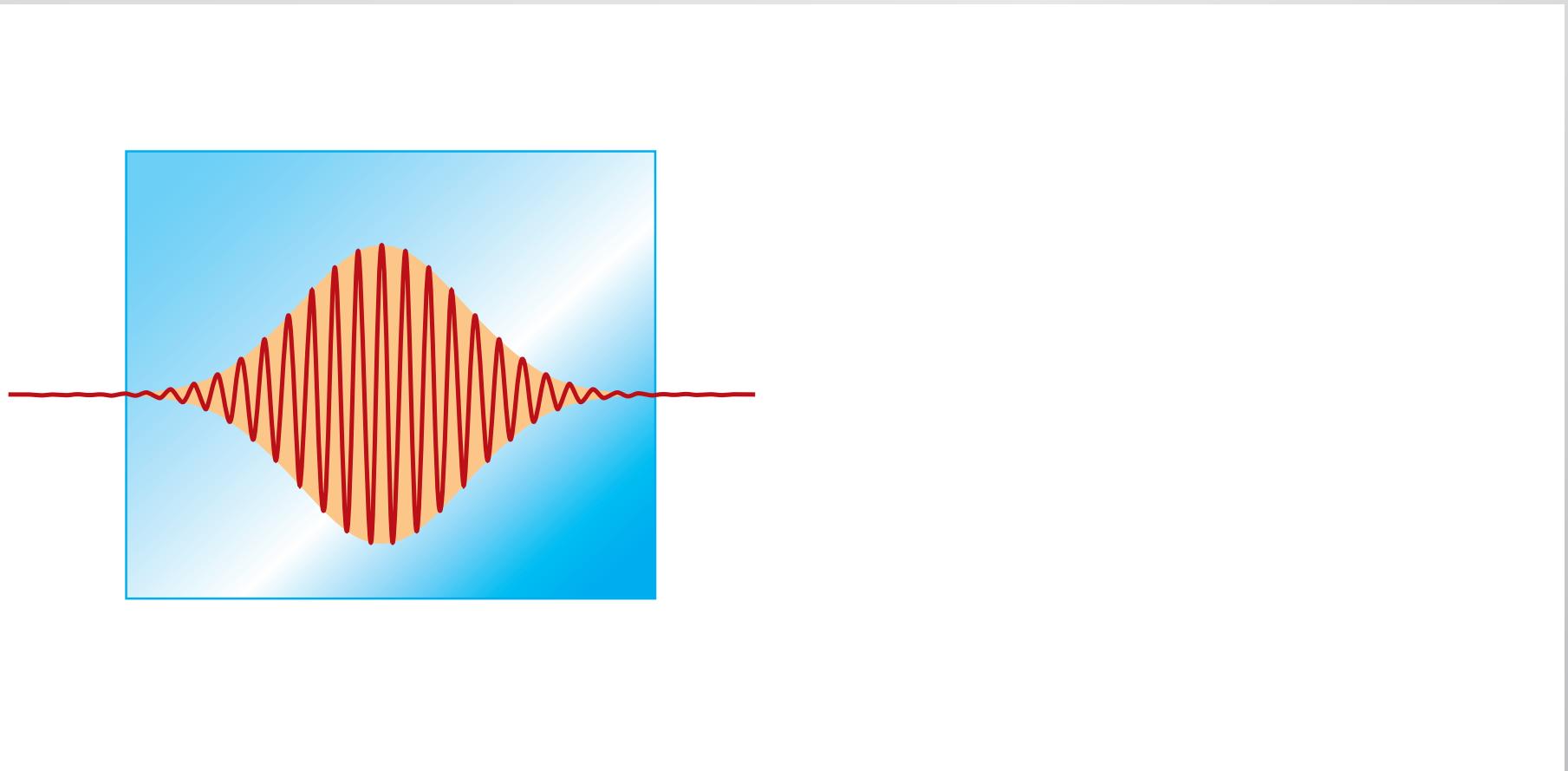
and so $P = P^{(1)} + P^{(3)} = (\chi^{(1)} + \chi^{(3)} I) E \equiv \chi_{eff} E$

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

Nonlinear optics

Intensity-dependent index of refraction:

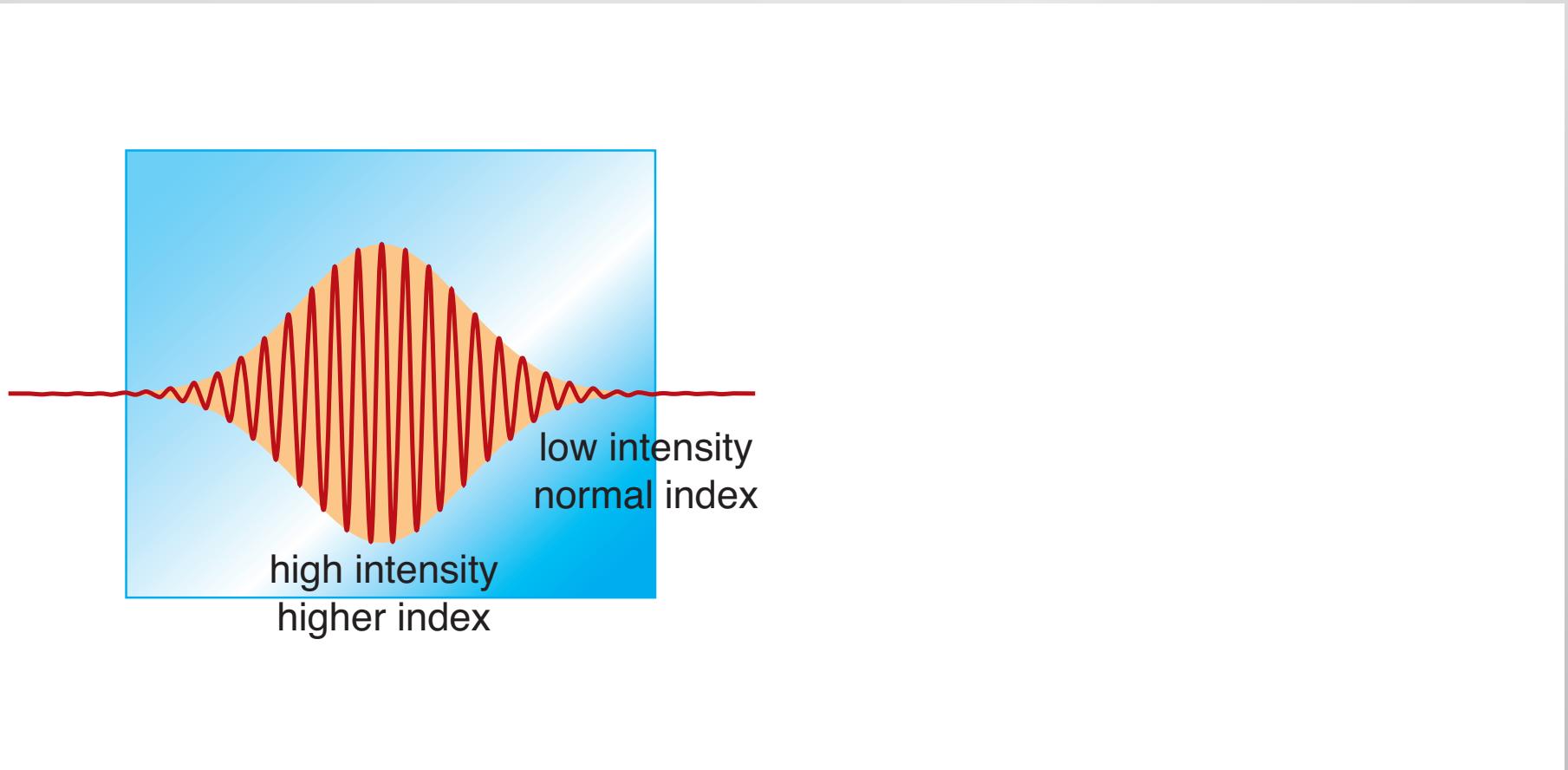
$$n = n_o + n_2 I$$



Nonlinear optics

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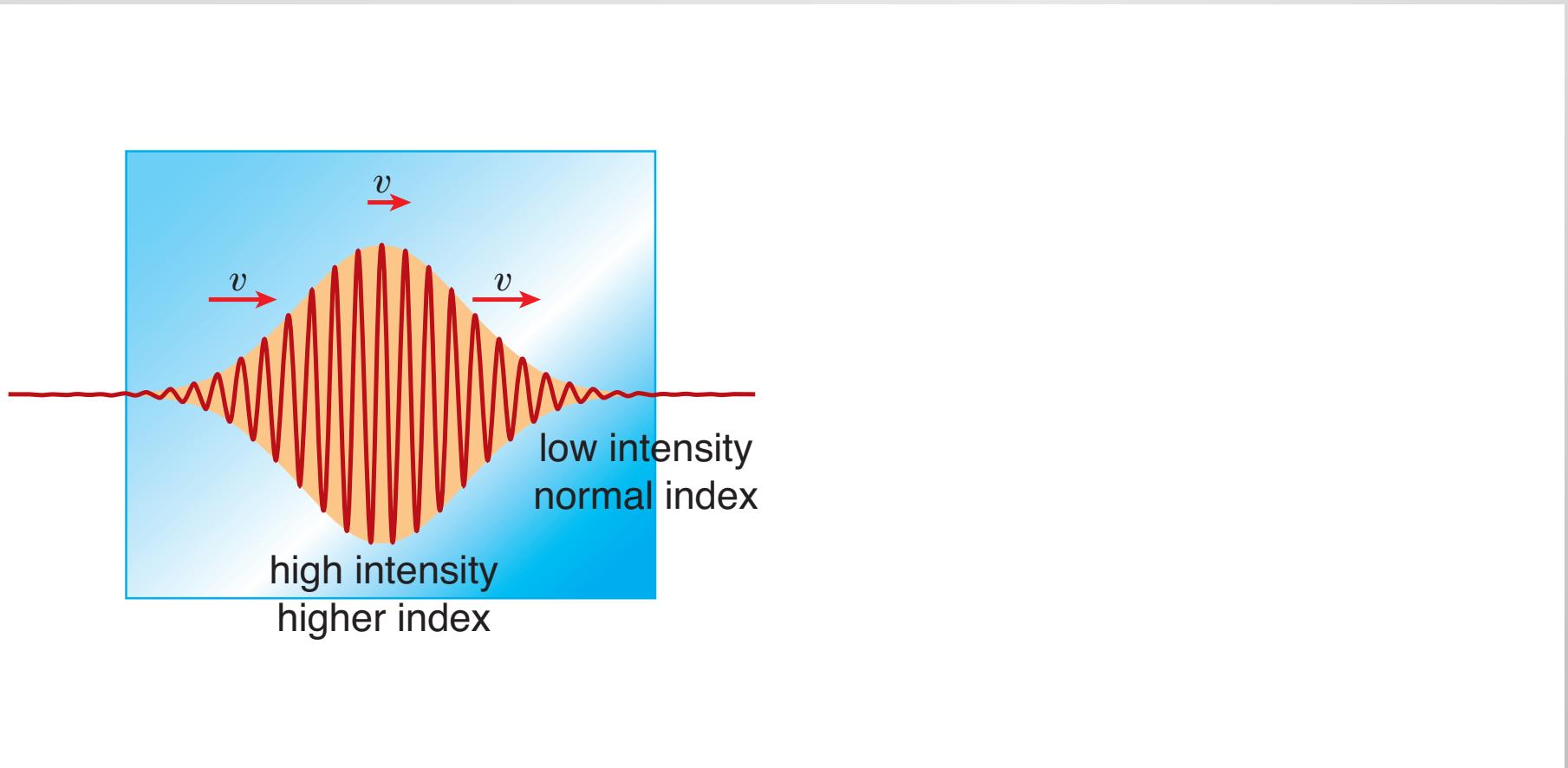
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Nonlinear optics

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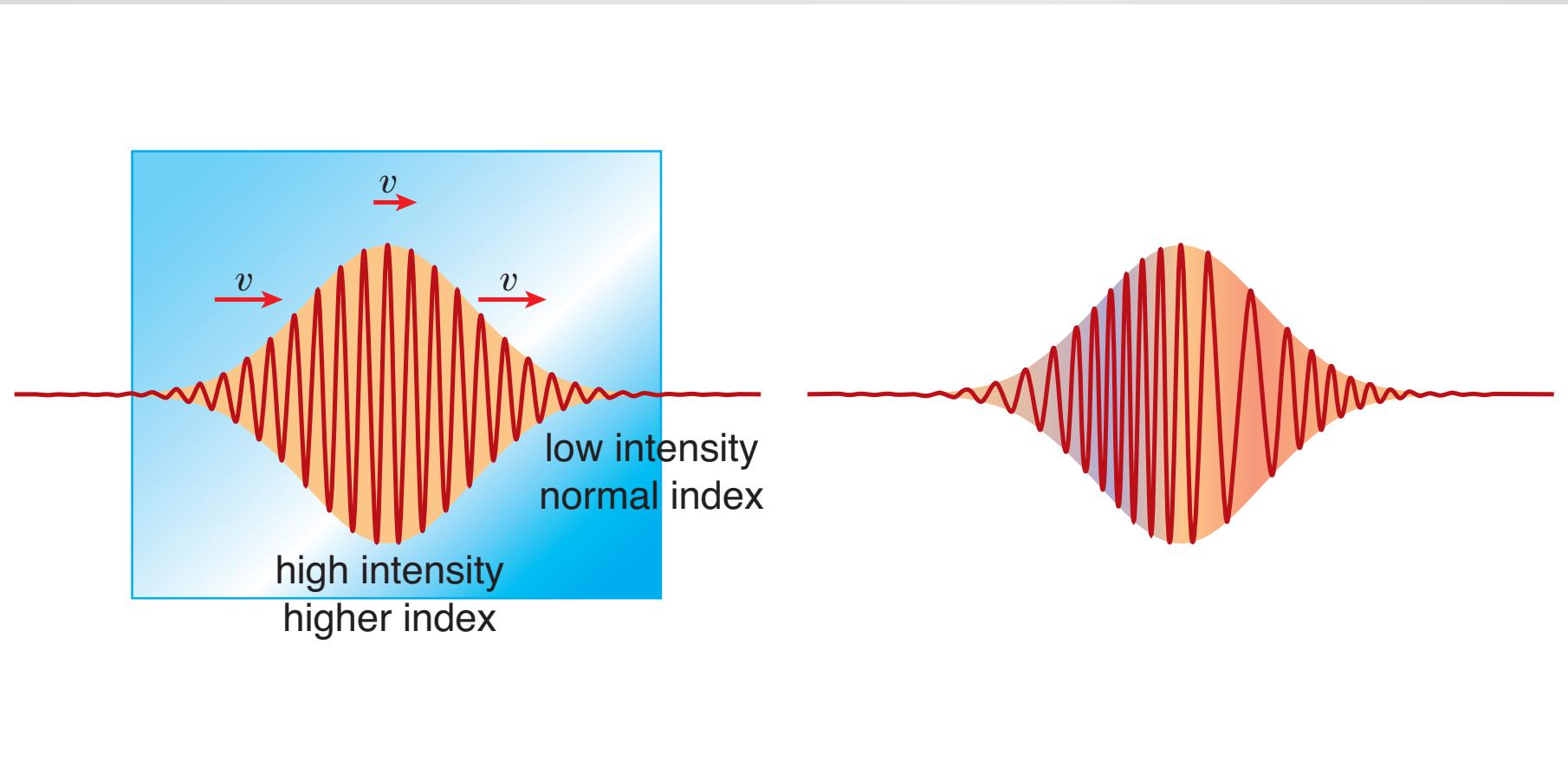
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Nonlinear optics

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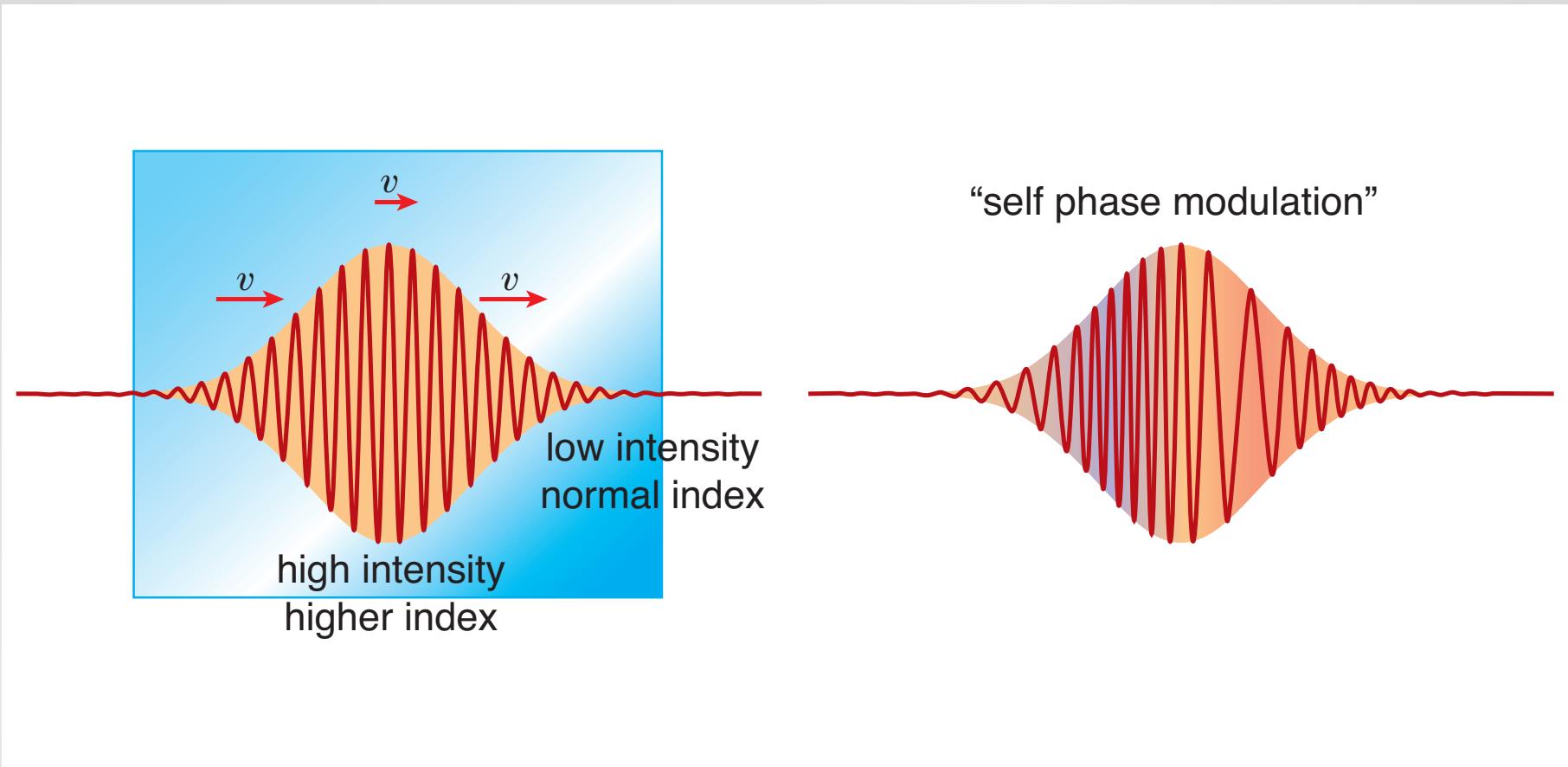
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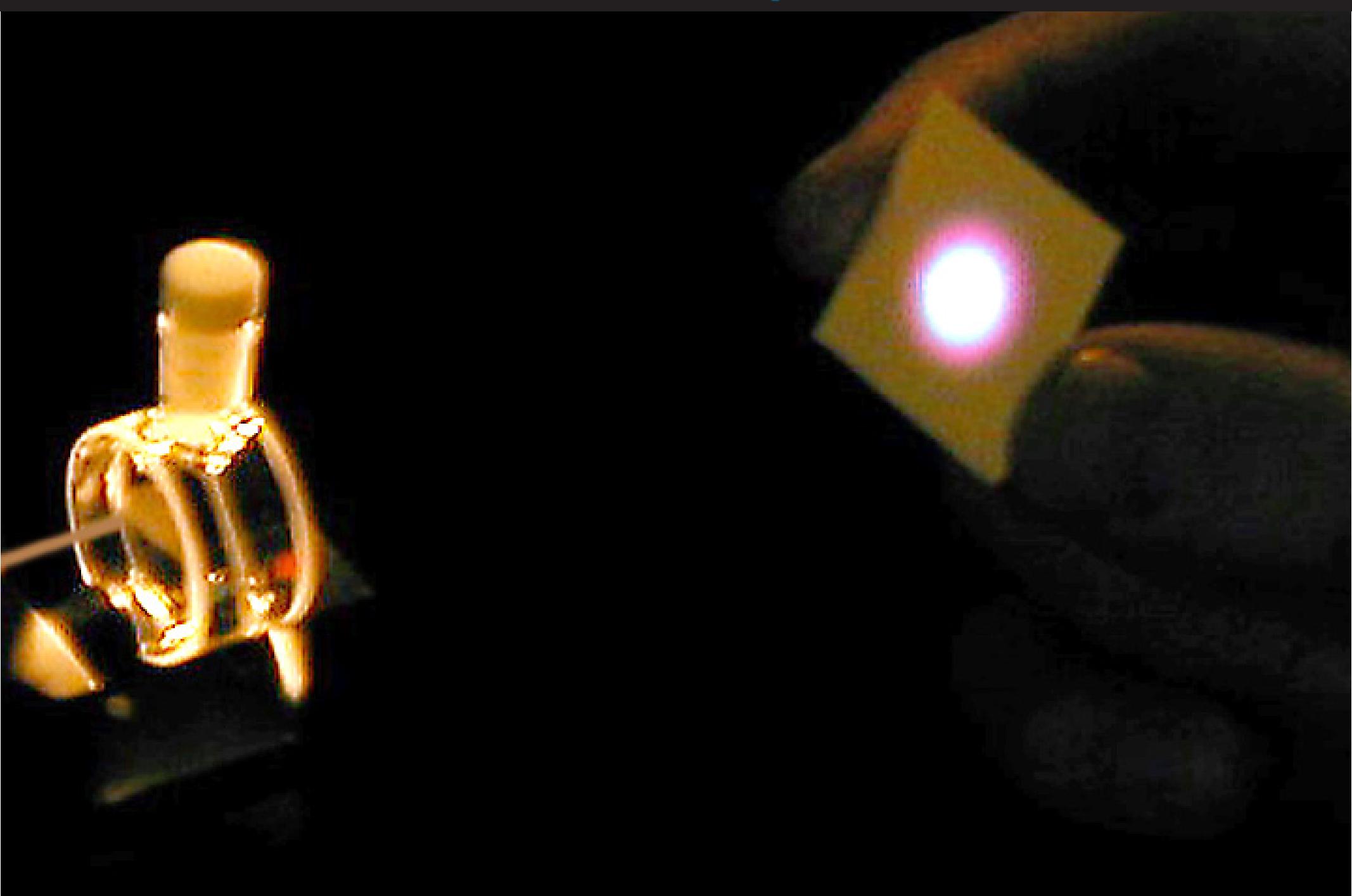
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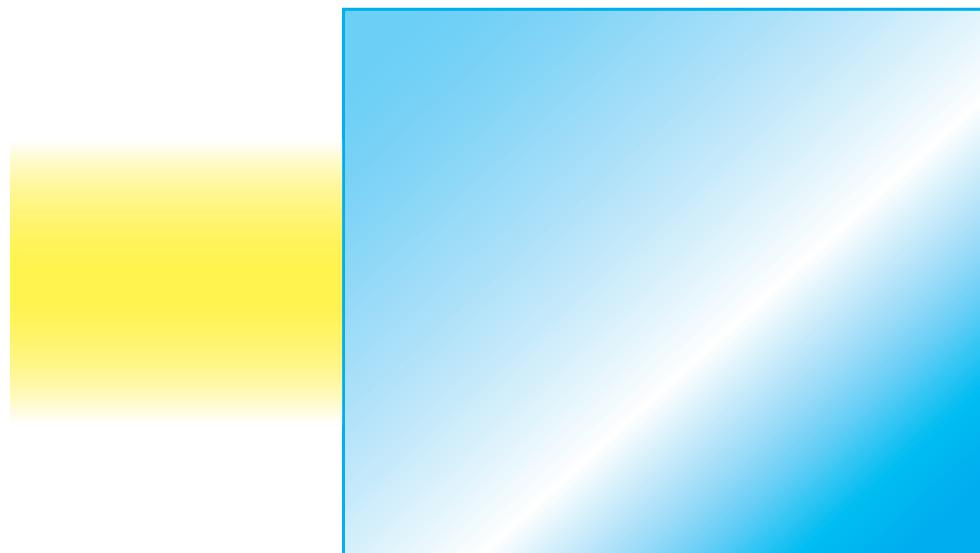
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Nonlinear optics

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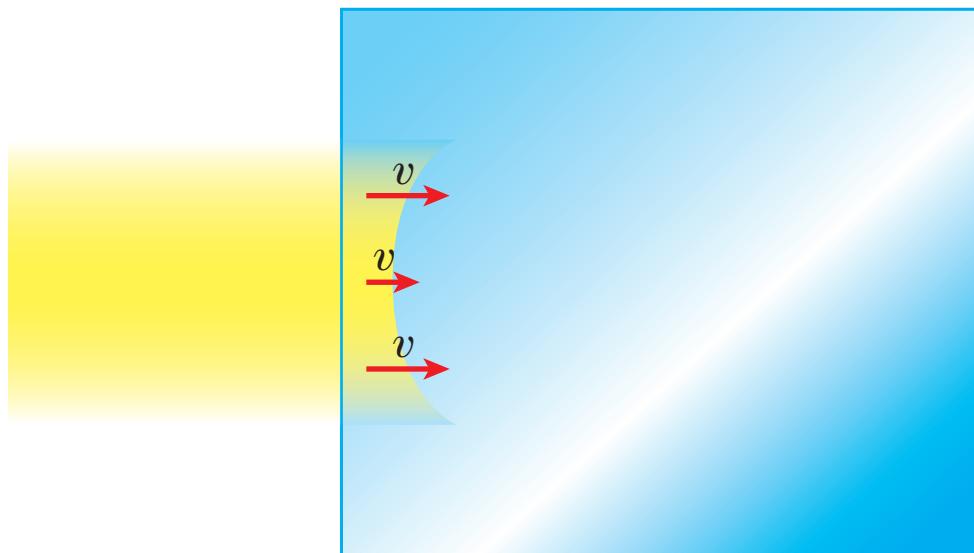
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Nonlinear optics

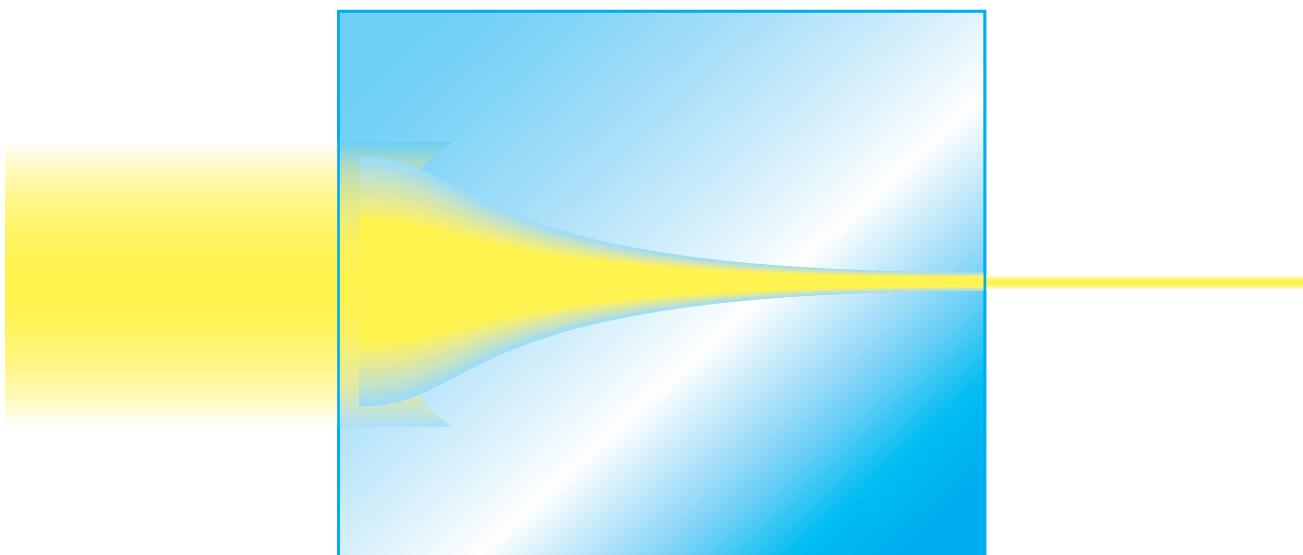
Intensity-dependent index of refraction:

$$n = n_o + n_2 I$$



Nonlinear optics

self-focusing



Nonlinear optics

but susceptibility is complex!

susceptibility

real part

imaginary part

linear

refraction

absorption

nonlinear

SHG, SFG, DFG, THG,...

multiphoton absorption

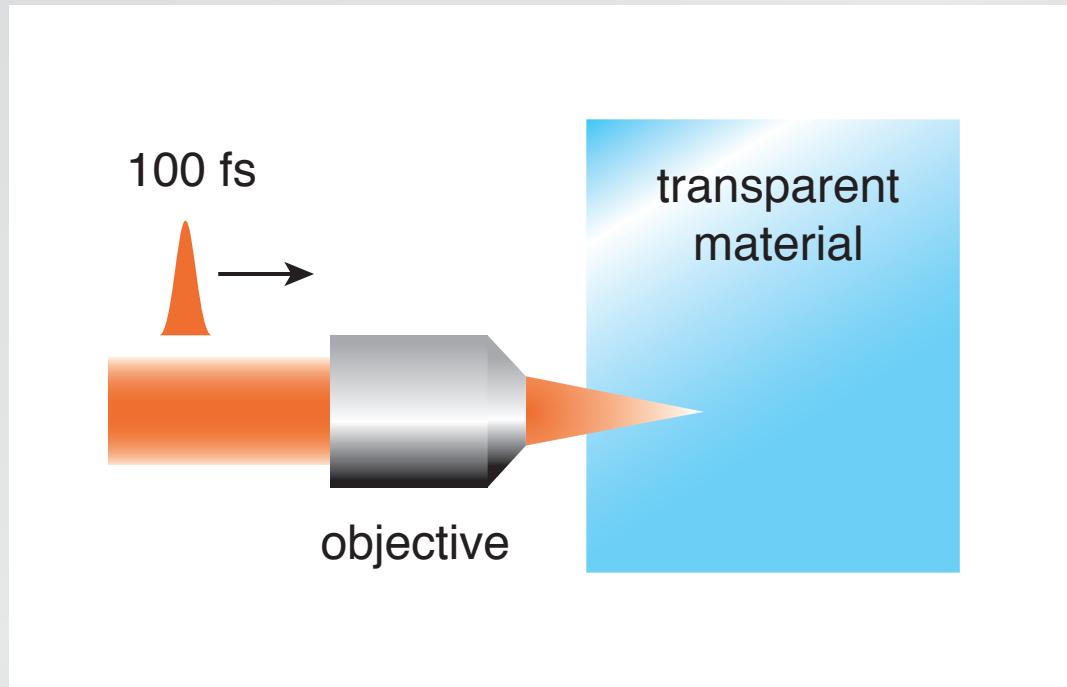
$$\alpha = \alpha_o + \beta I + \gamma I^2 + \dots$$

Outline

- propagation of pulses
- nonlinear optics
- femtosecond micromachining

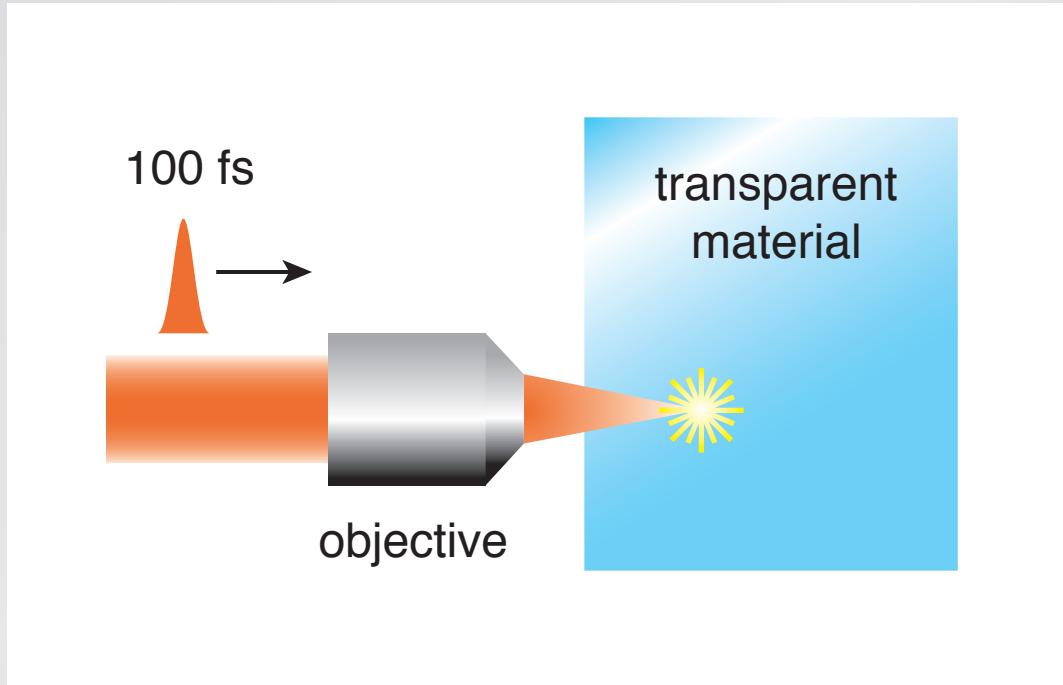
Femtosecond micromachining

high intensity at focus...



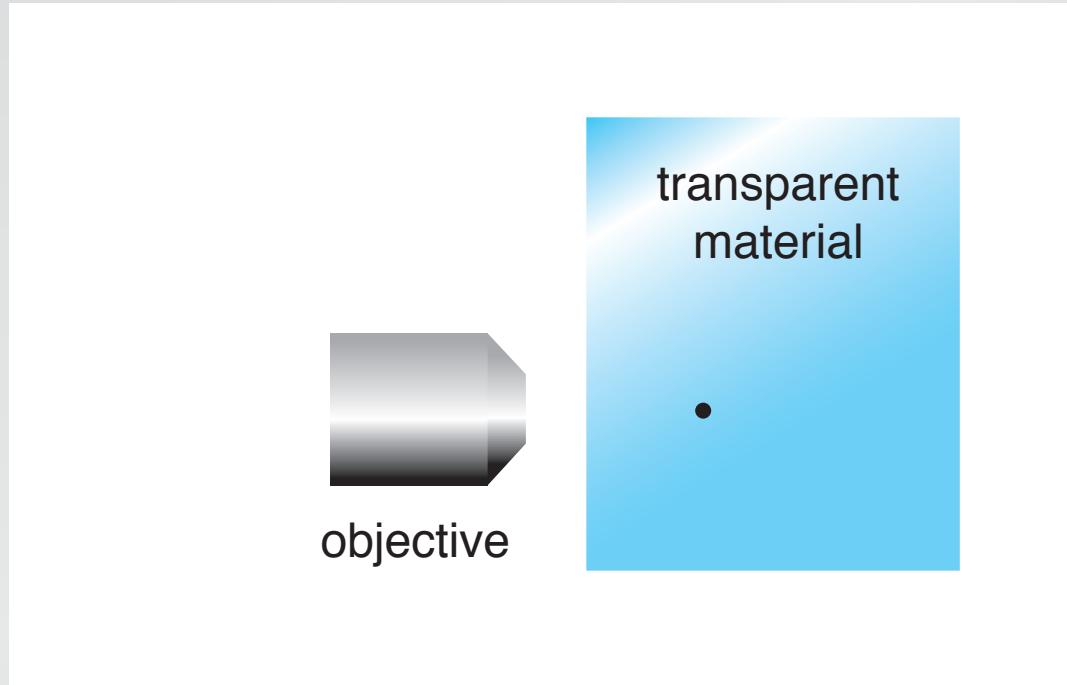
Femtosecond micromachining

...causes nonlinear ionization...



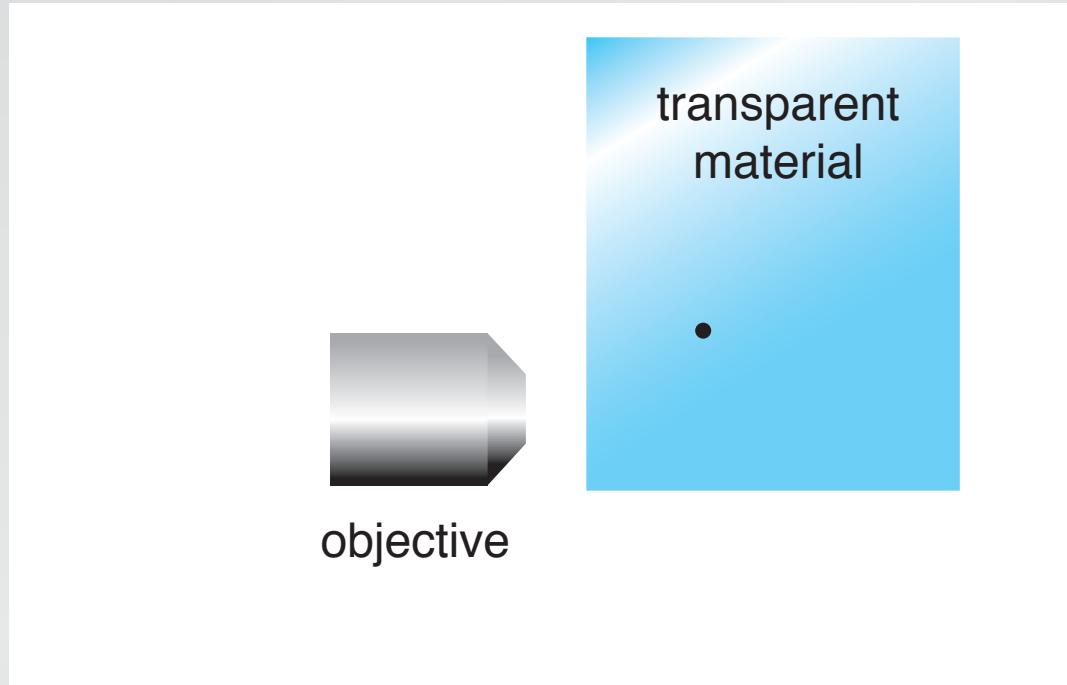
Femtosecond micromachining

and 'microexplosion' causes microscopic damage...



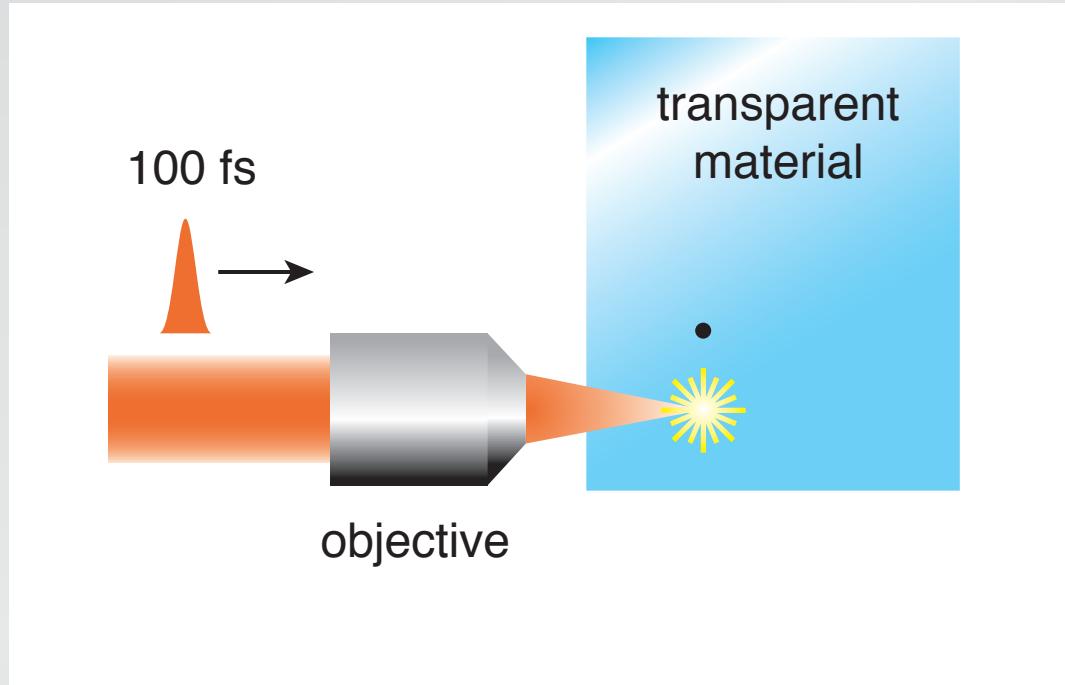
Femtosecond micromachining

translate sample



Femtosecond micromachining

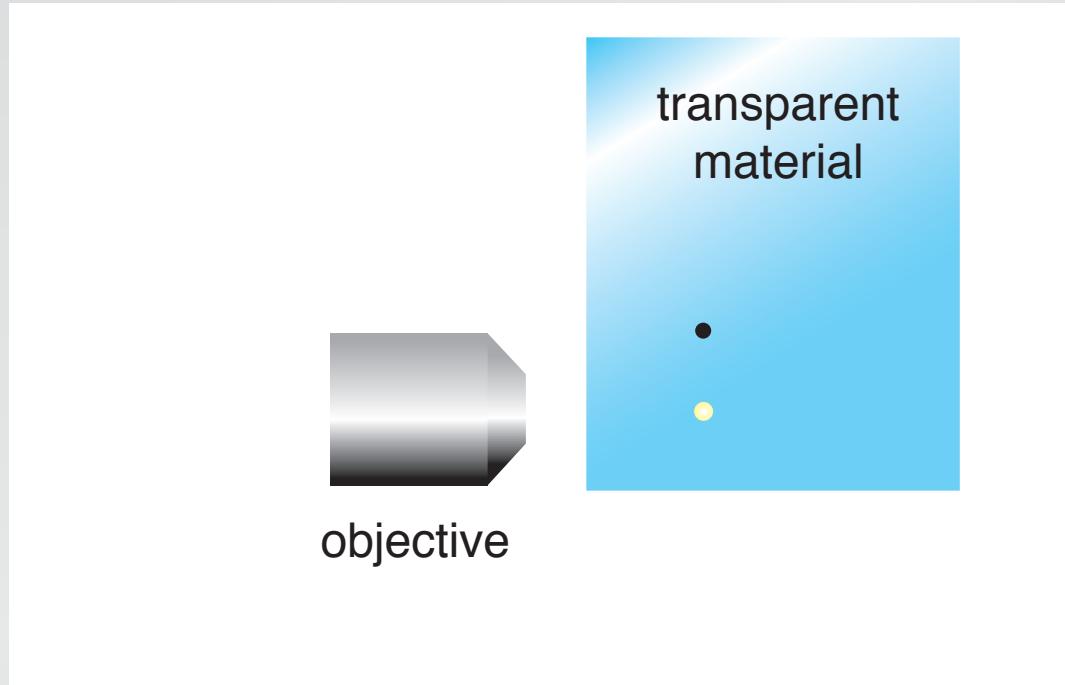
time scales



100 fs: laser energy transferred to electrons

Femtosecond micromachining

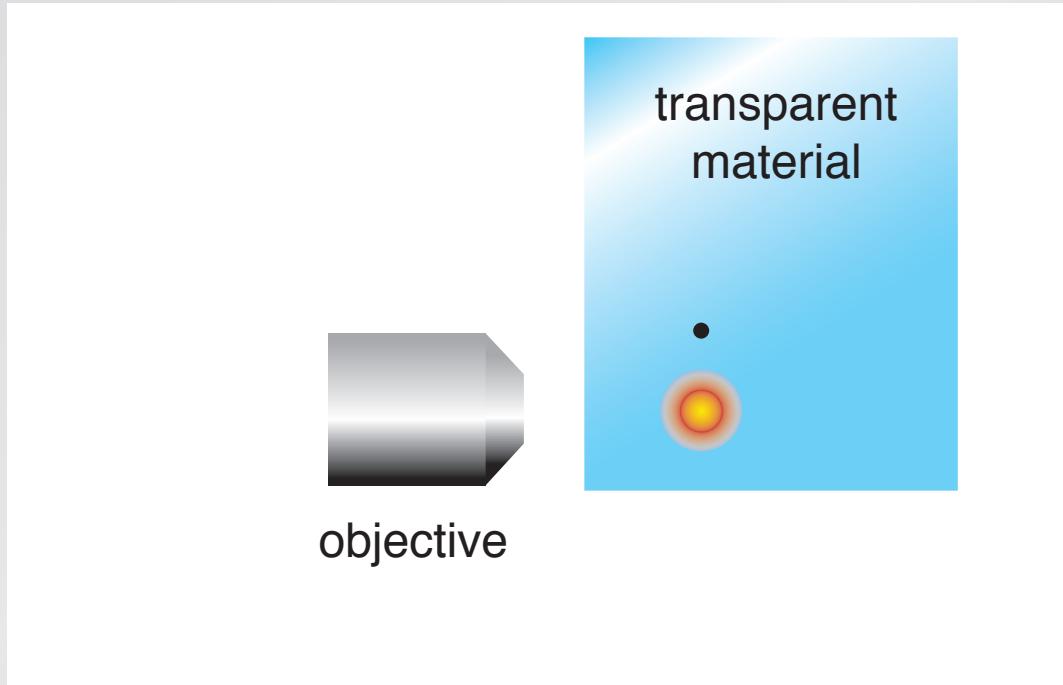
time scales



10 ps: energy transfer to ions

Femtosecond micromachining

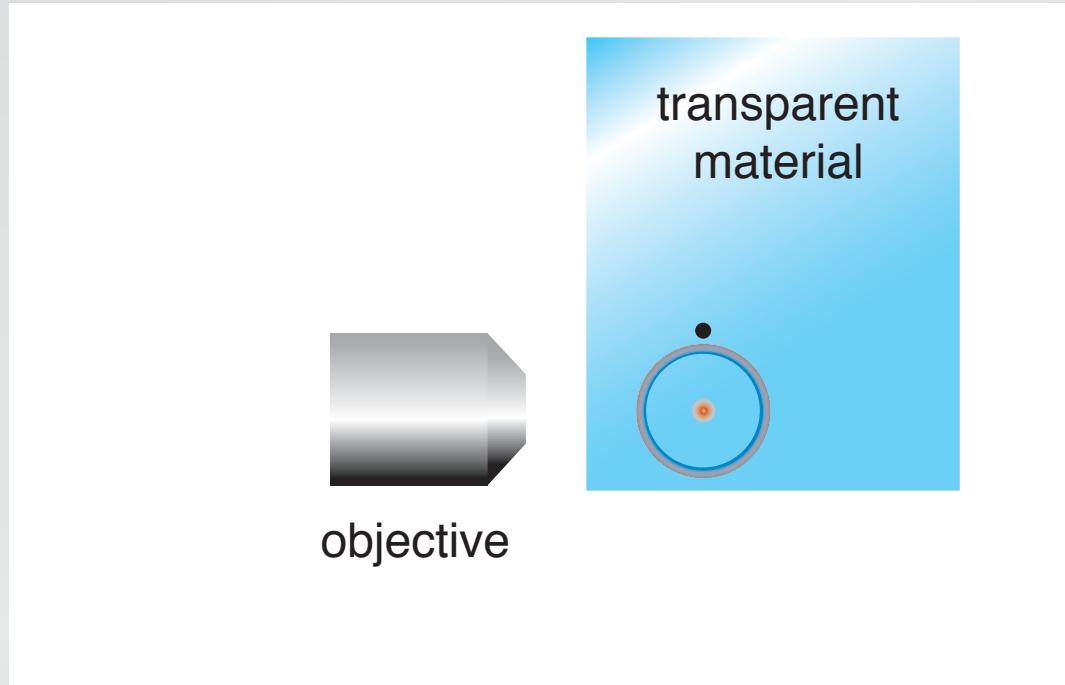
time scales



100 ps: plasma expansion

Femtosecond micromachining

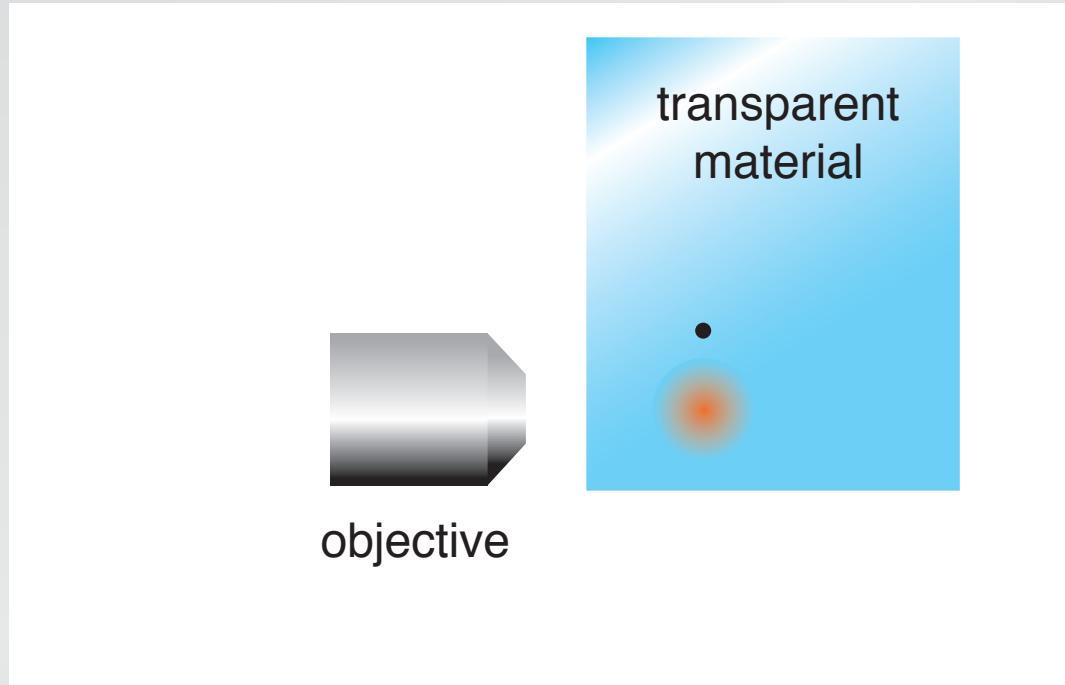
time scales



10–100 ns: shock propagation

Femtosecond micromachining

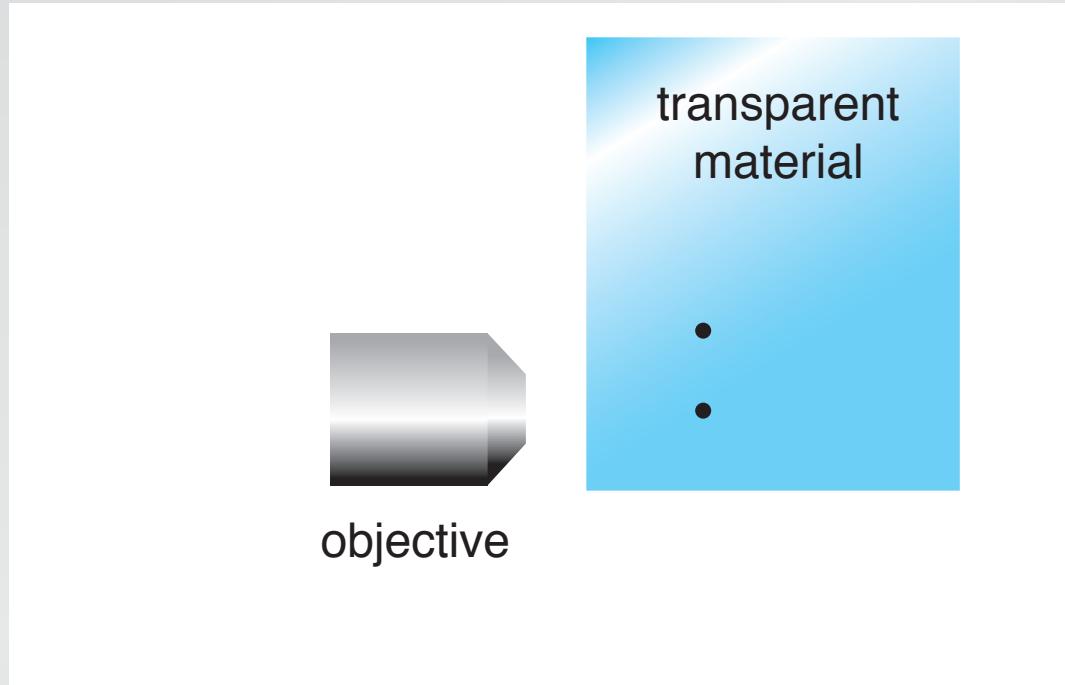
time scales



1 μ s: thermal expansion

Femtosecond micromachining

time scales

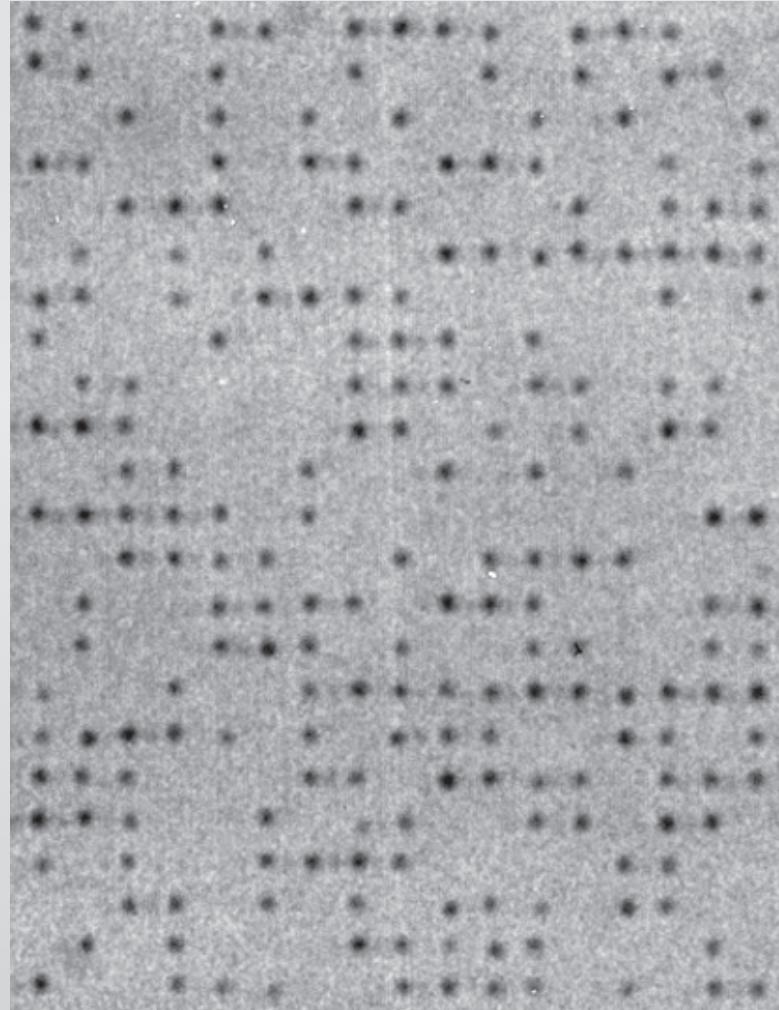


1 ms: permanent structural damage

Femtosecond micromachining

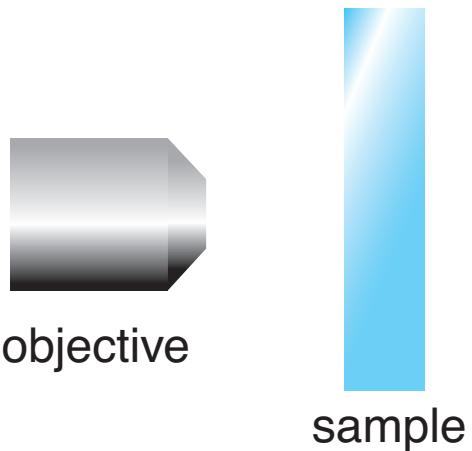
Some applications:

- data storage
- waveguides
- microfluidics



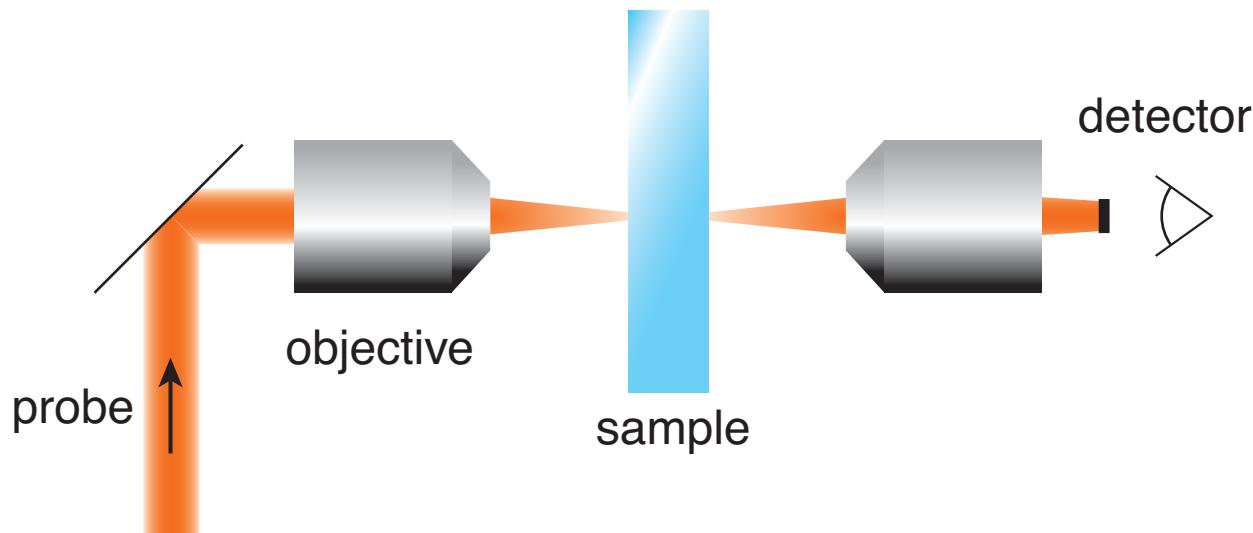
Femtosecond micromachining

Dark-field scattering



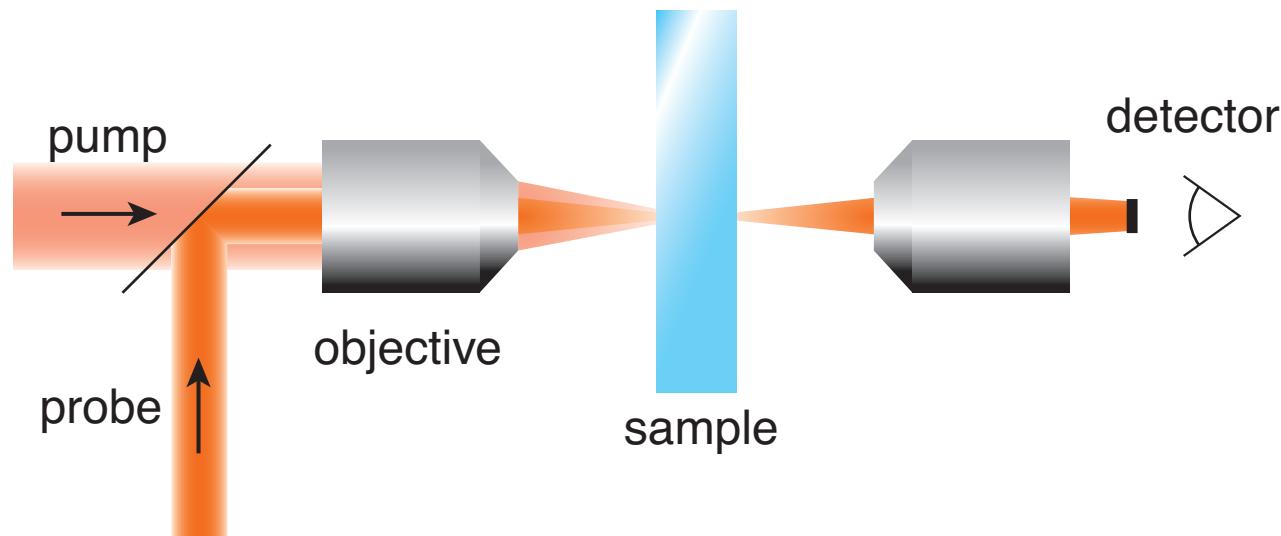
Femtosecond micromachining

block probe beam...



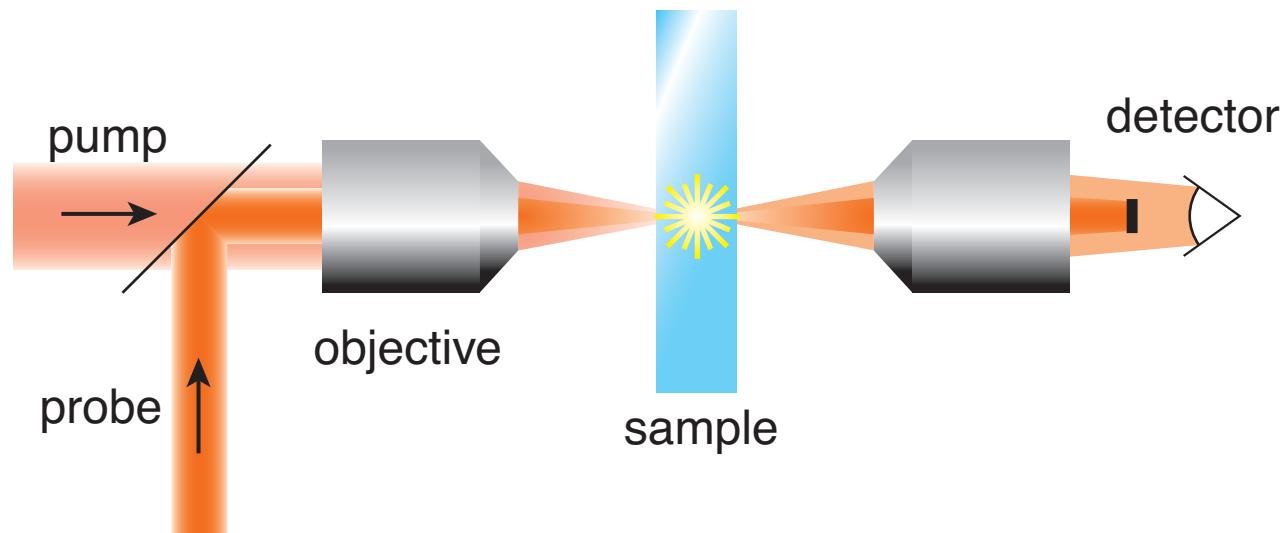
Femtosecond micromachining

... bring in pump beam...



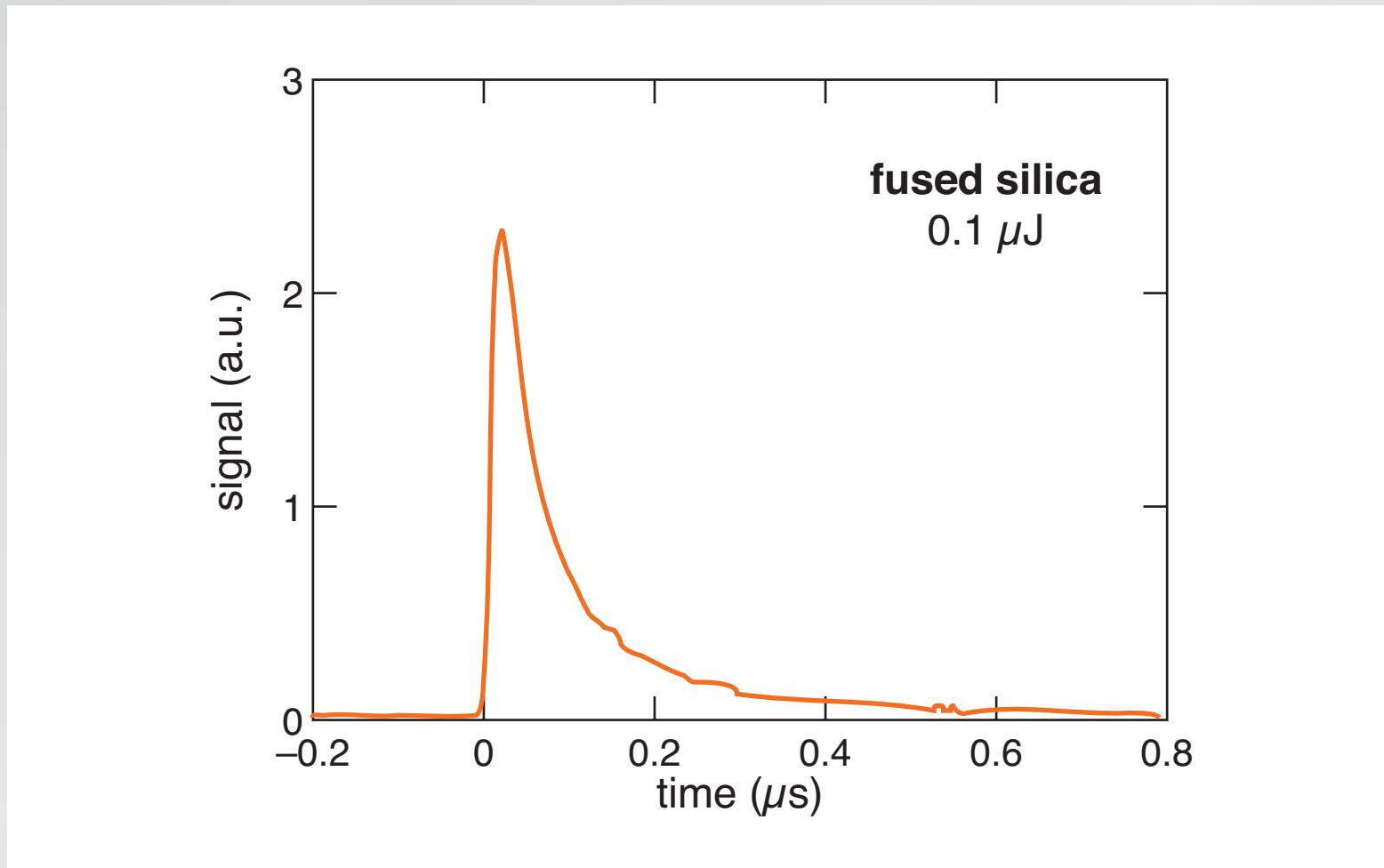
Femtosecond micromachining

... damage scatters probe beam



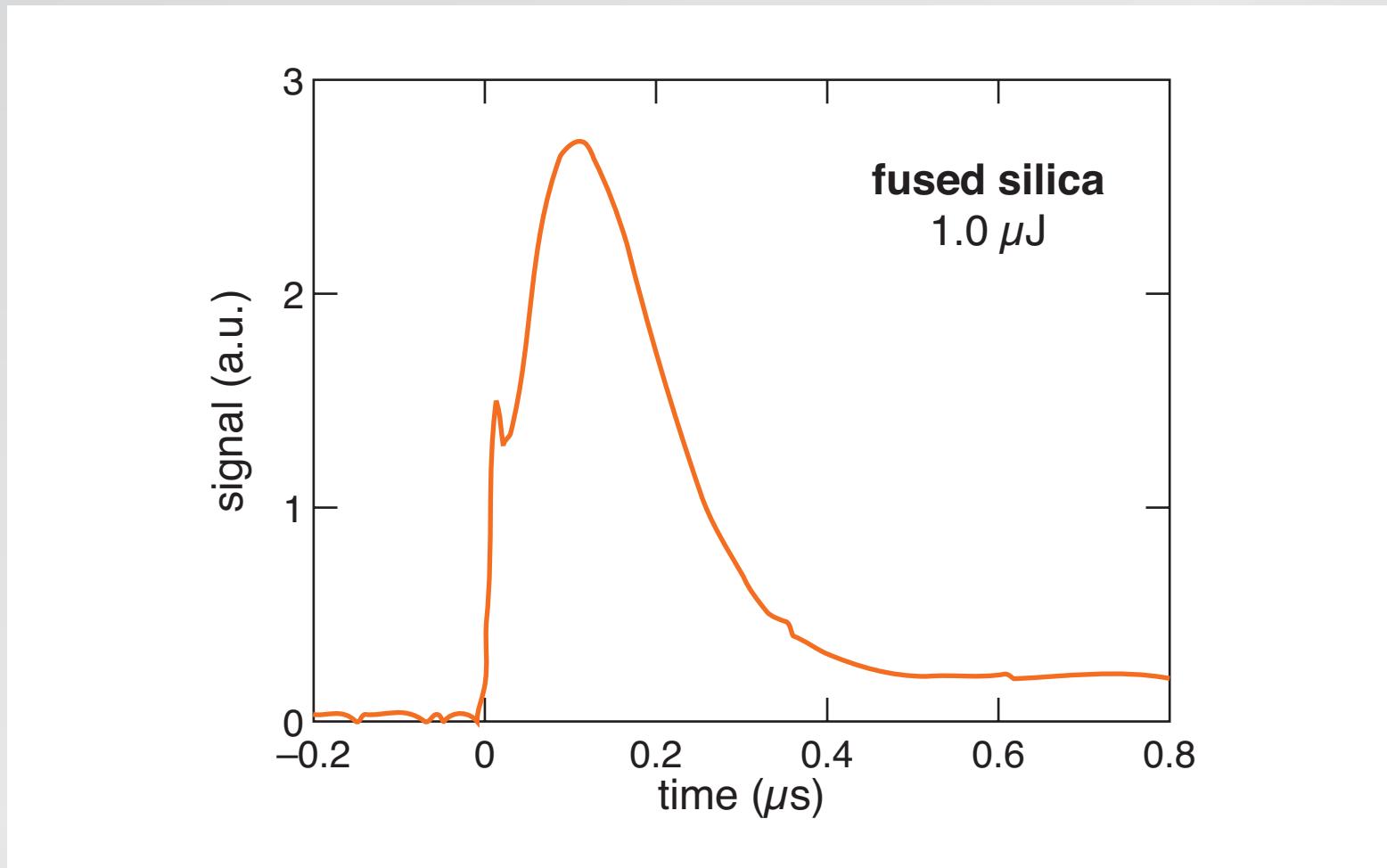
Femtosecond micromachining

scattered signal



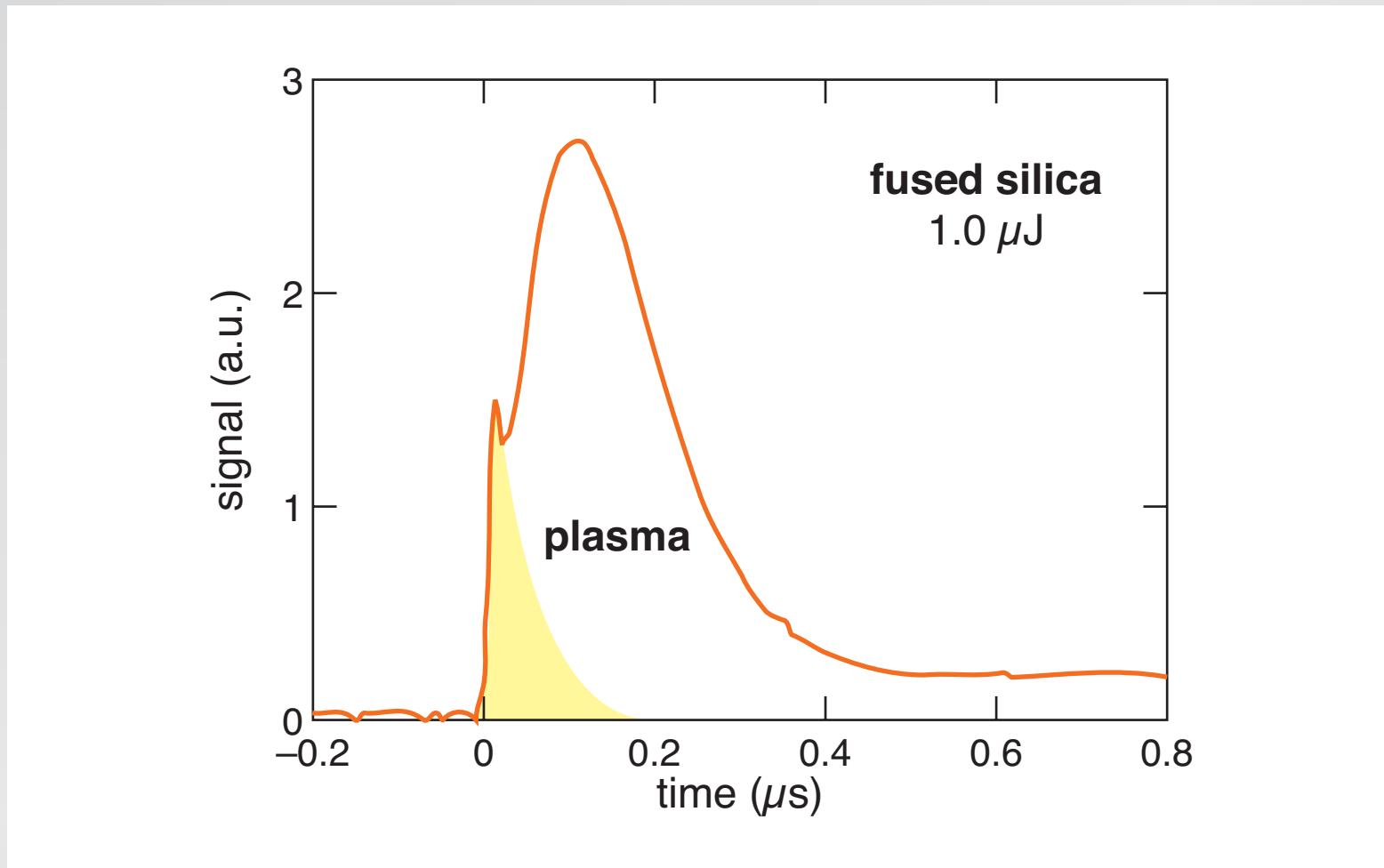
Femtosecond micromachining

scattered signal



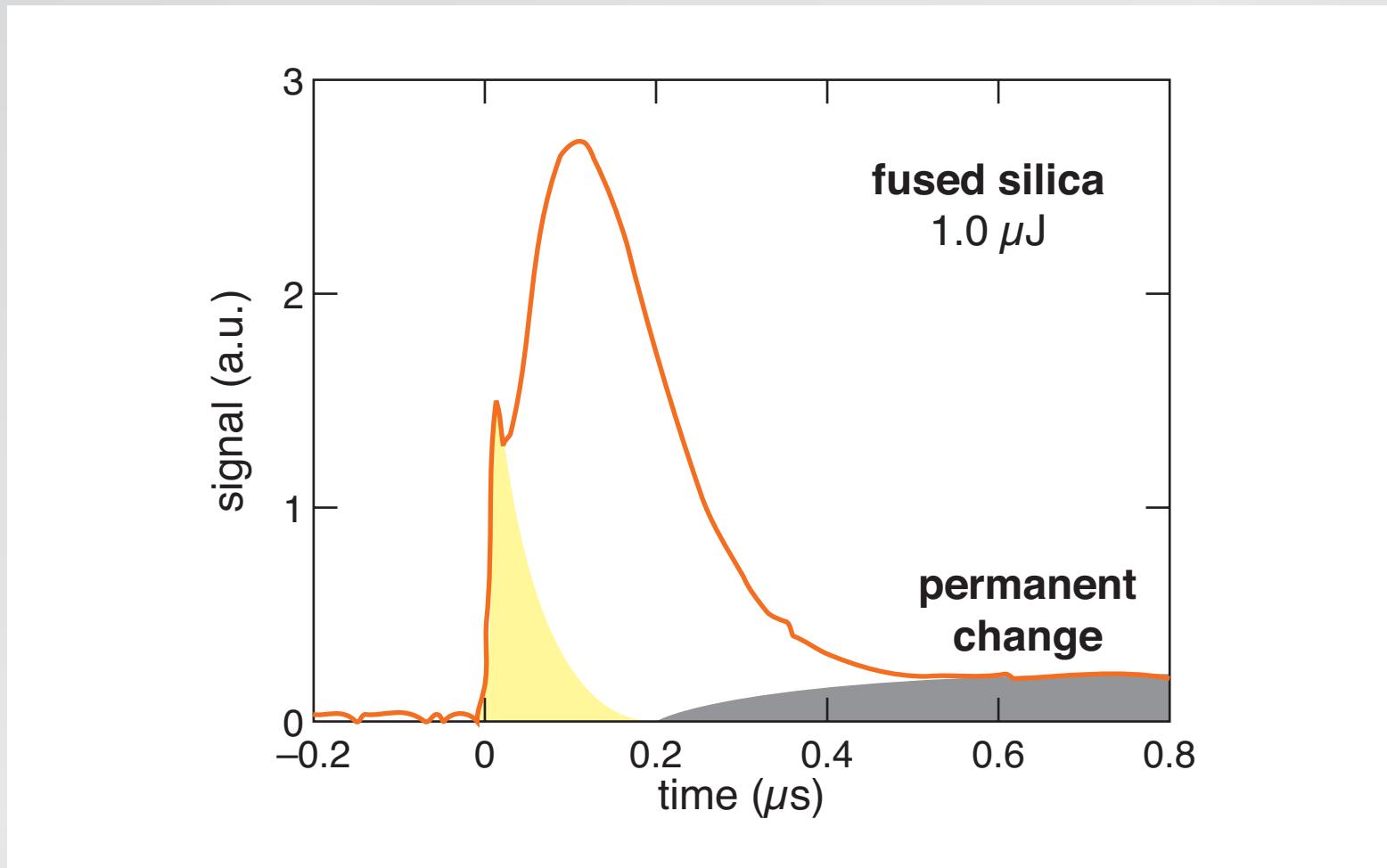
Femtosecond micromachining

scattered signal



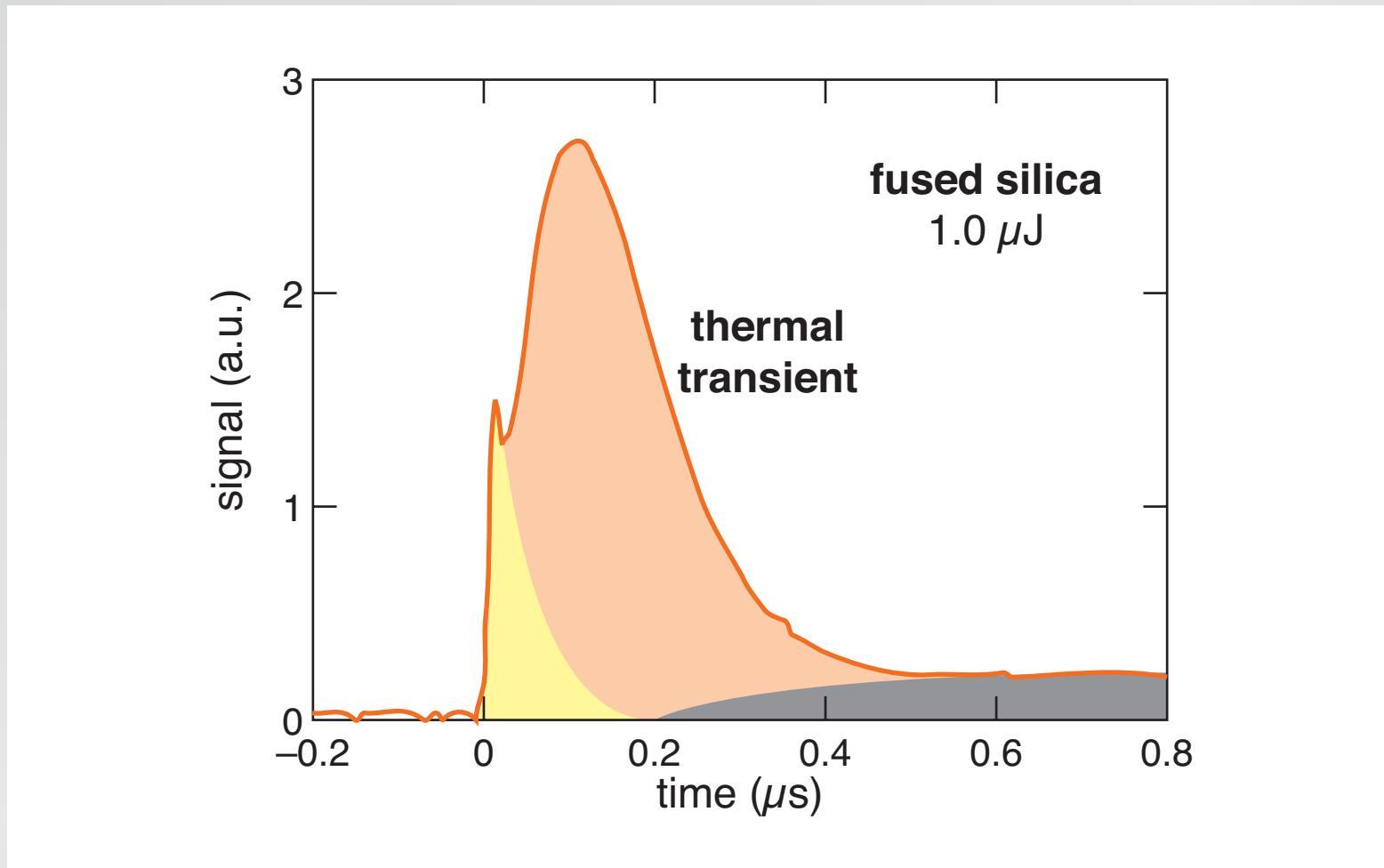
Femtosecond micromachining

scattered signal



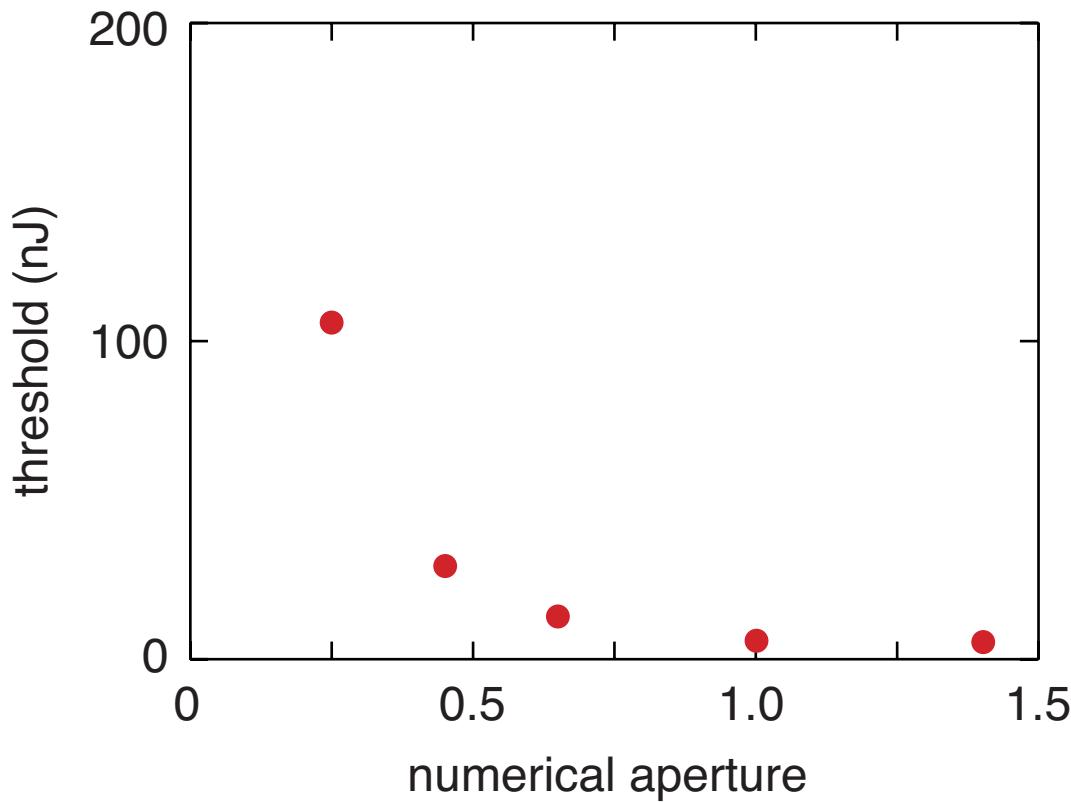
Femtosecond micromachining

scattered signal



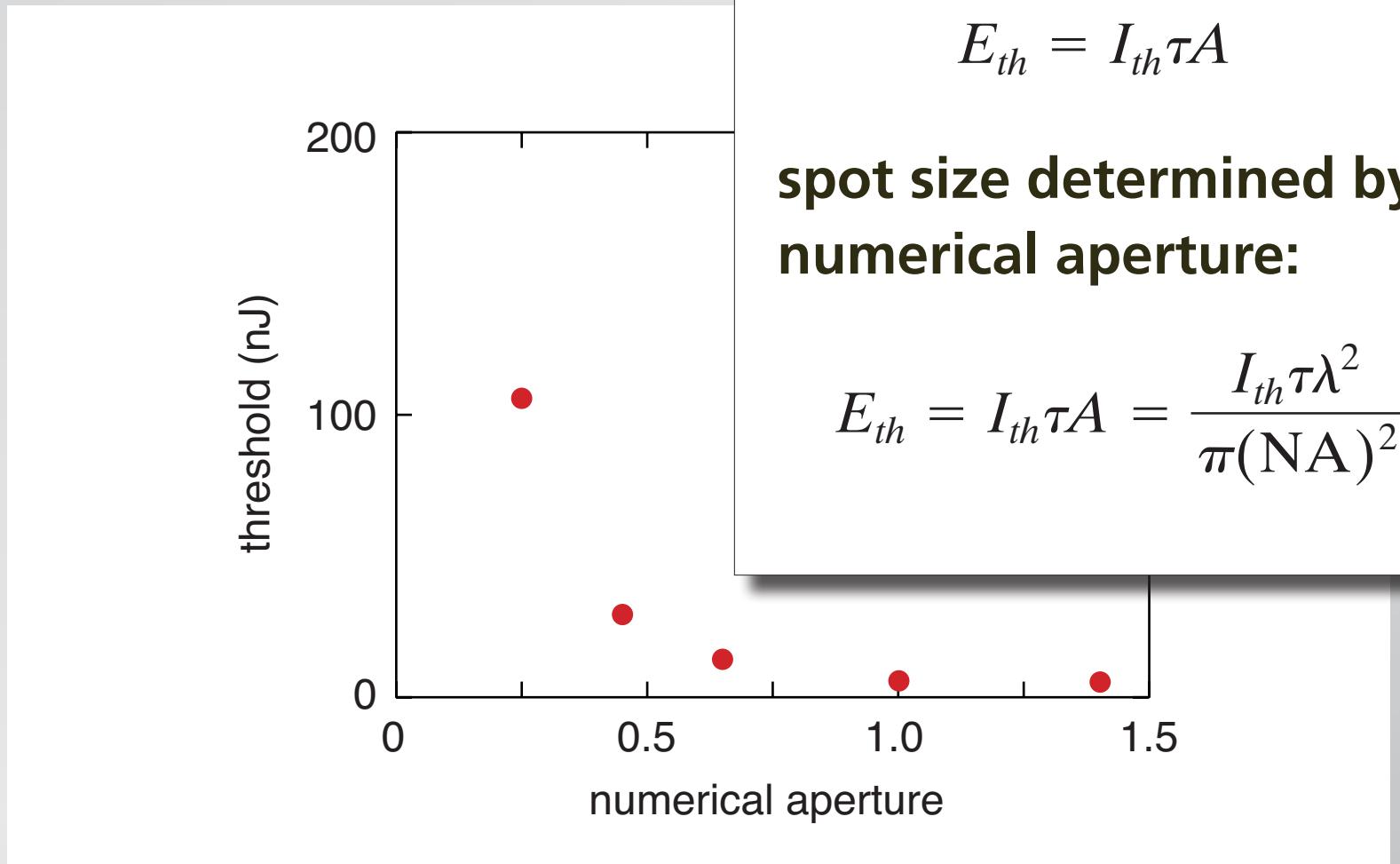
Femtosecond micromachining

vary numerical aperture



Femtosecond micromachining

vary numerical aperture:



intensity threshold:

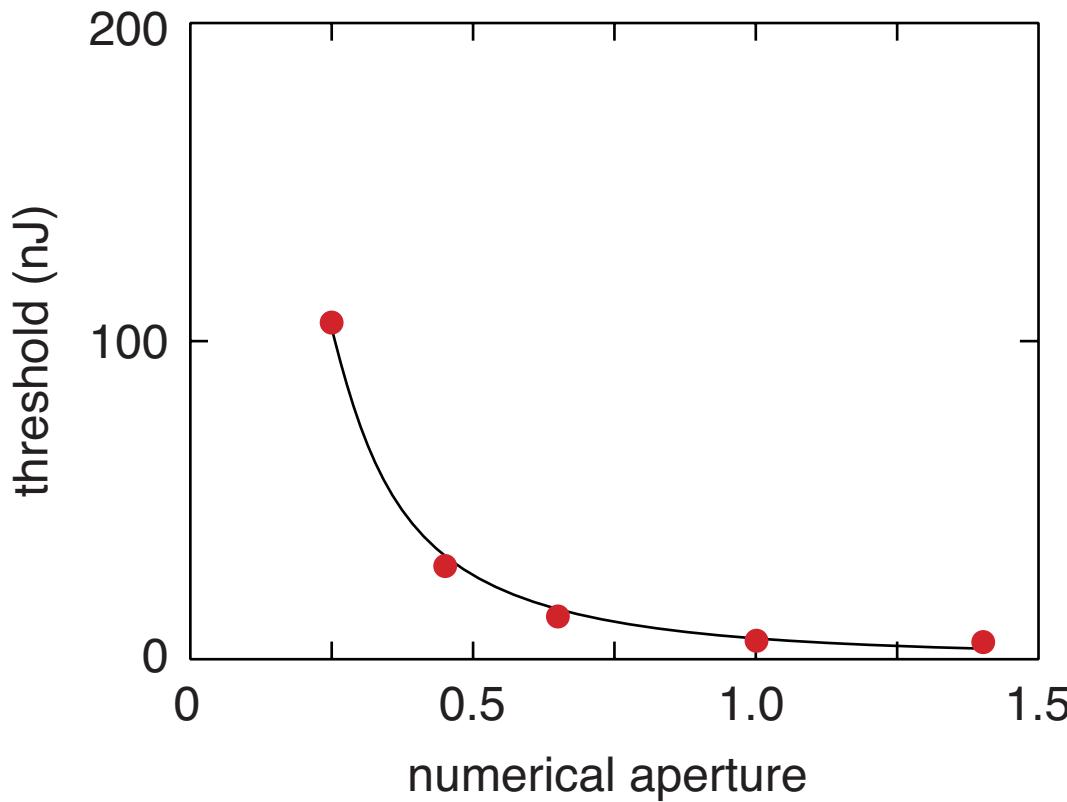
$$E_{th} = I_{th}\tau A$$

spot size determined by numerical aperture:

$$E_{th} = I_{th}\tau A = \frac{I_{th}\tau\lambda^2}{\pi(\text{NA})^2}$$

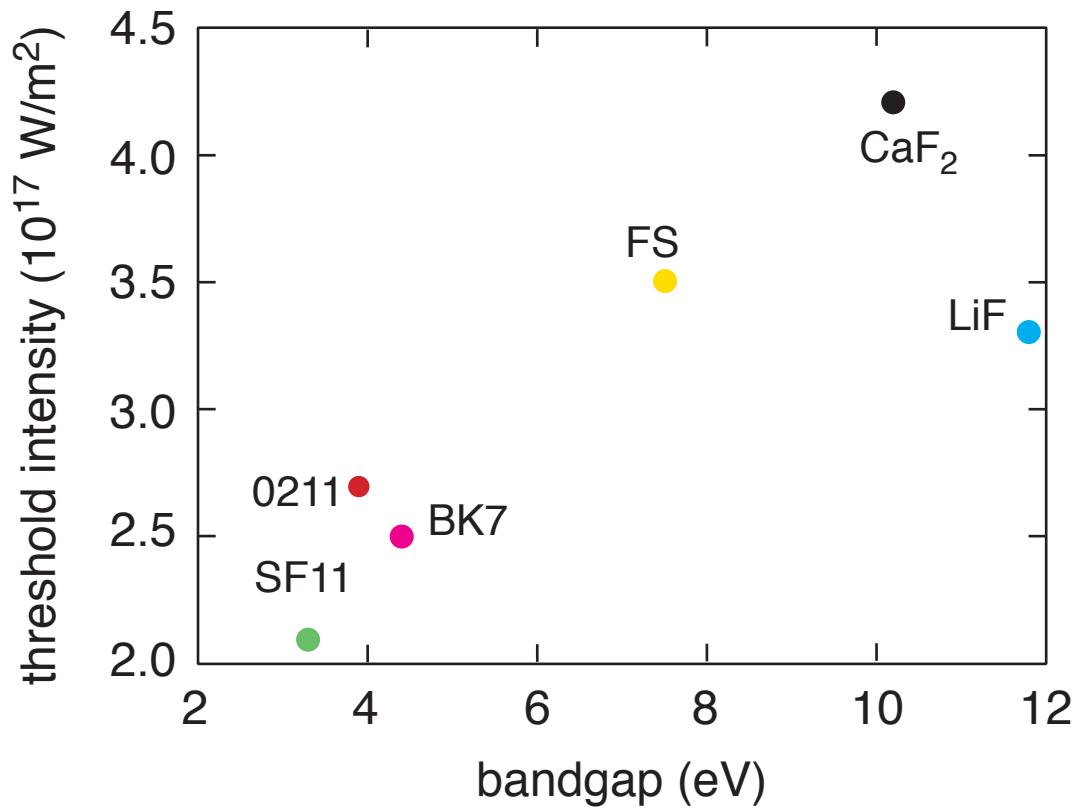
Femtosecond micromachining

fit gives threshold intensity: $I_{th} = 2.5 \times 10^{17} \text{ W/m}^2$



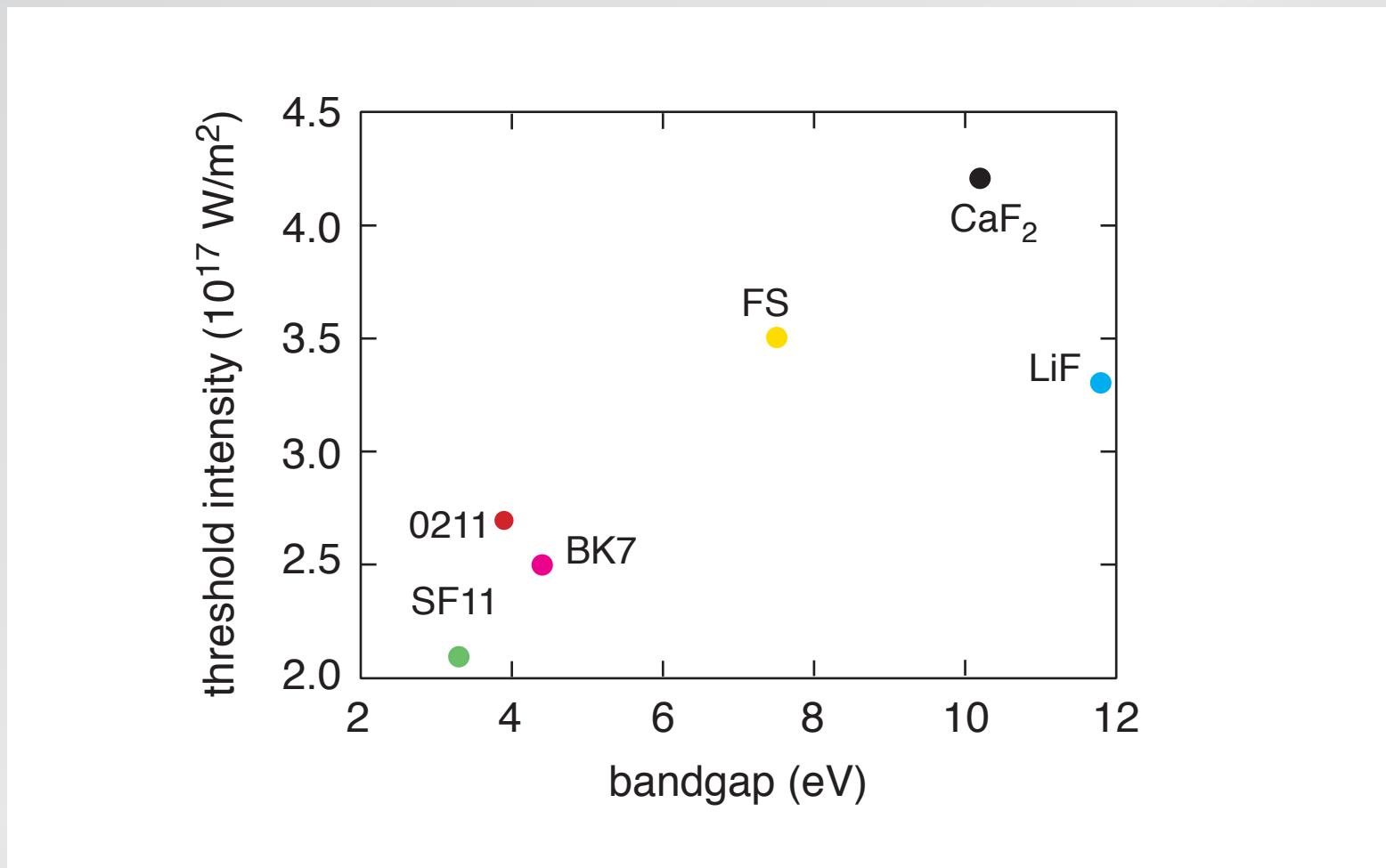
Femtosecond micromachining

vary material...



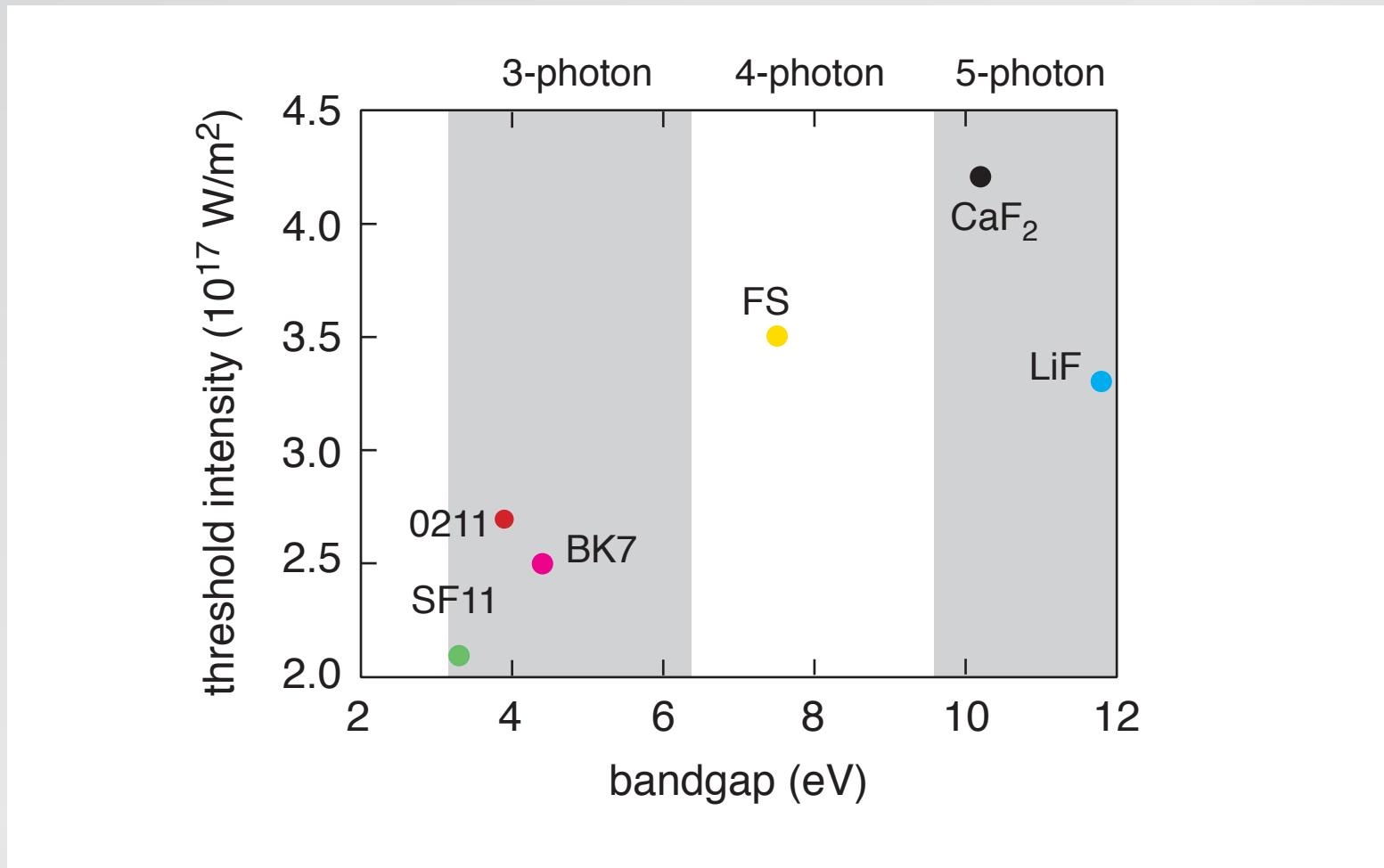
Femtosecond micromachining

...threshold varies with band gap (but not much!)



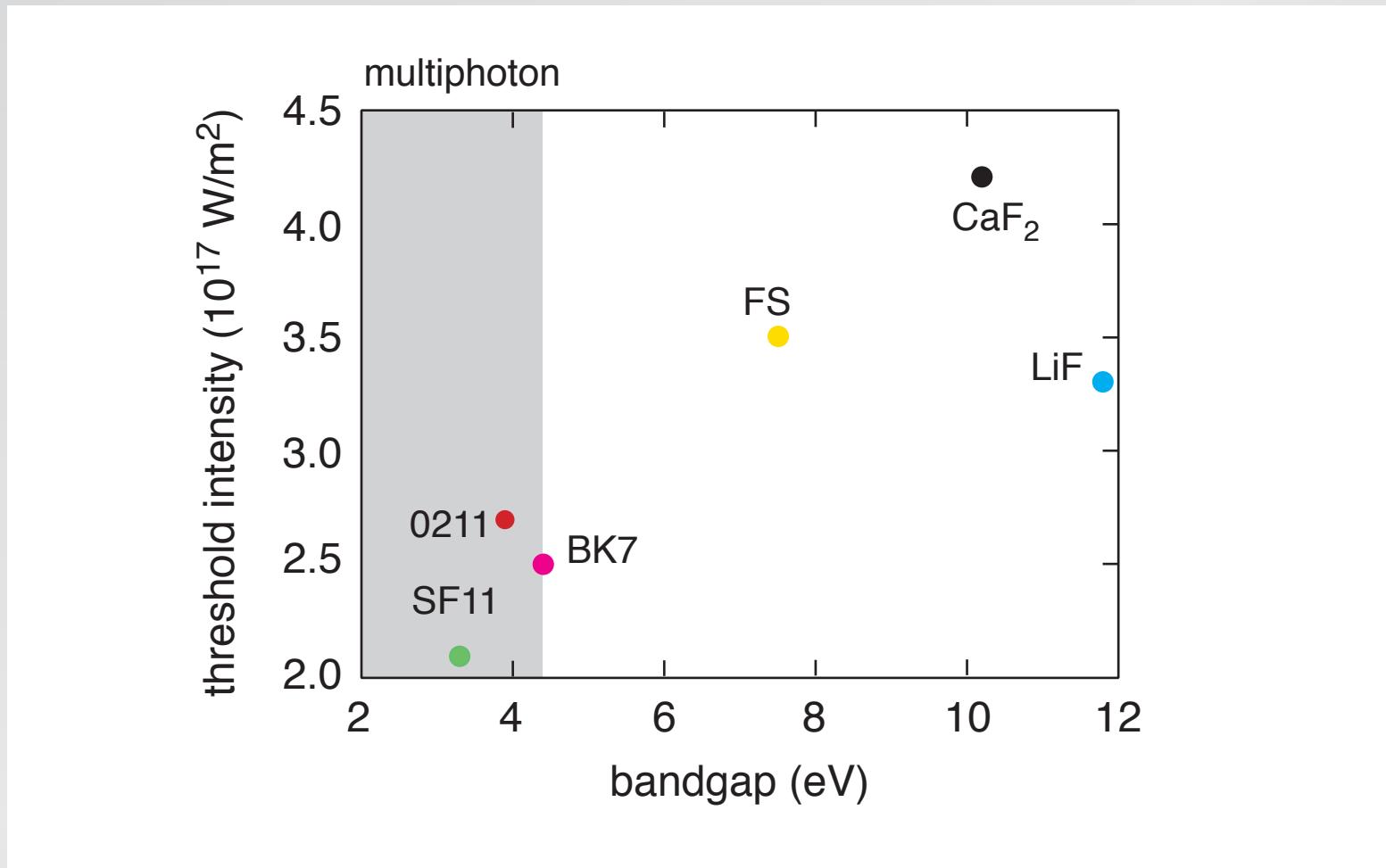
Femtosecond micromachining

would expect much more than a factor of 2



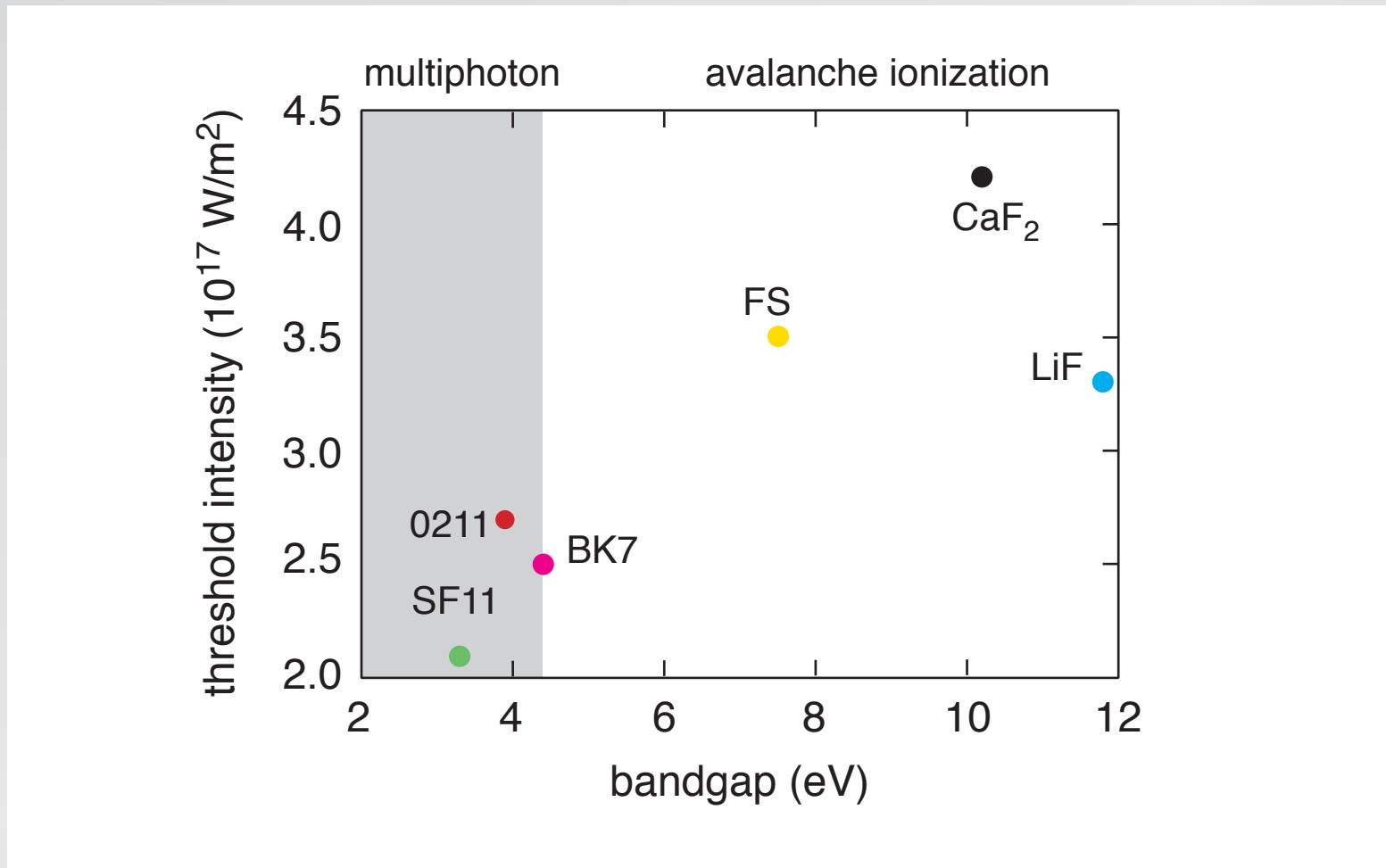
Femtosecond micromachining

critical density reached by multiphoton for low gap only



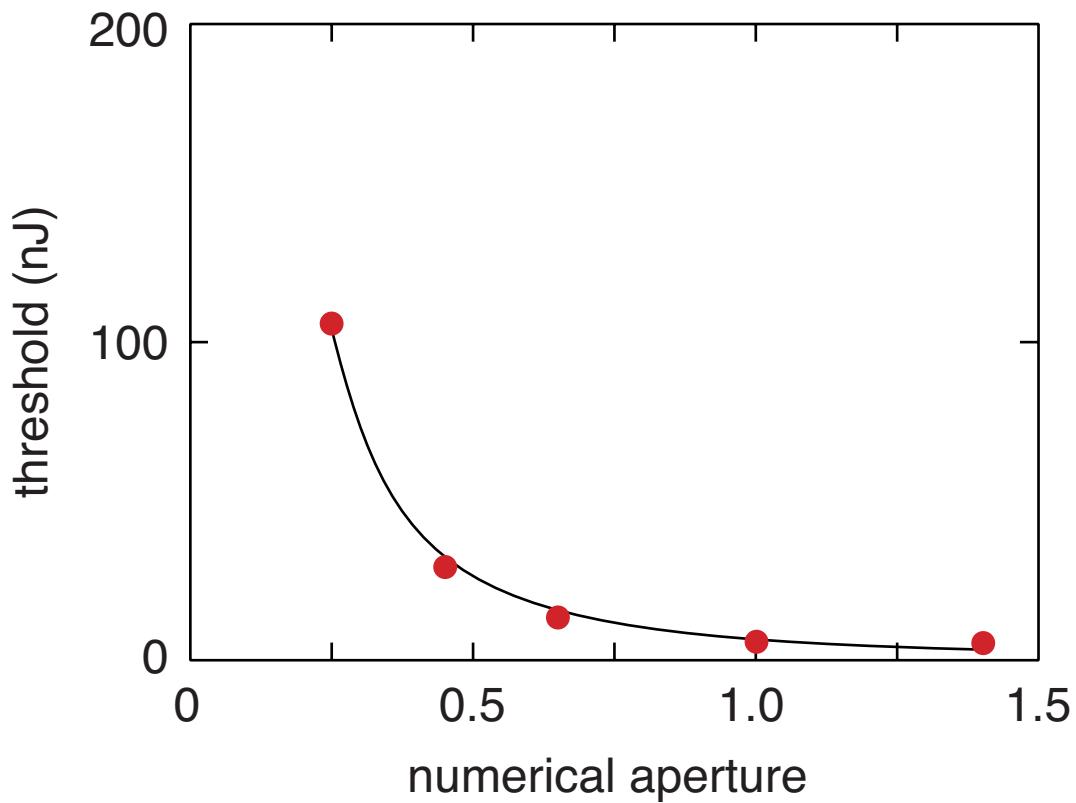
Femtosecond micromachining

avalanche ionization important at high gap



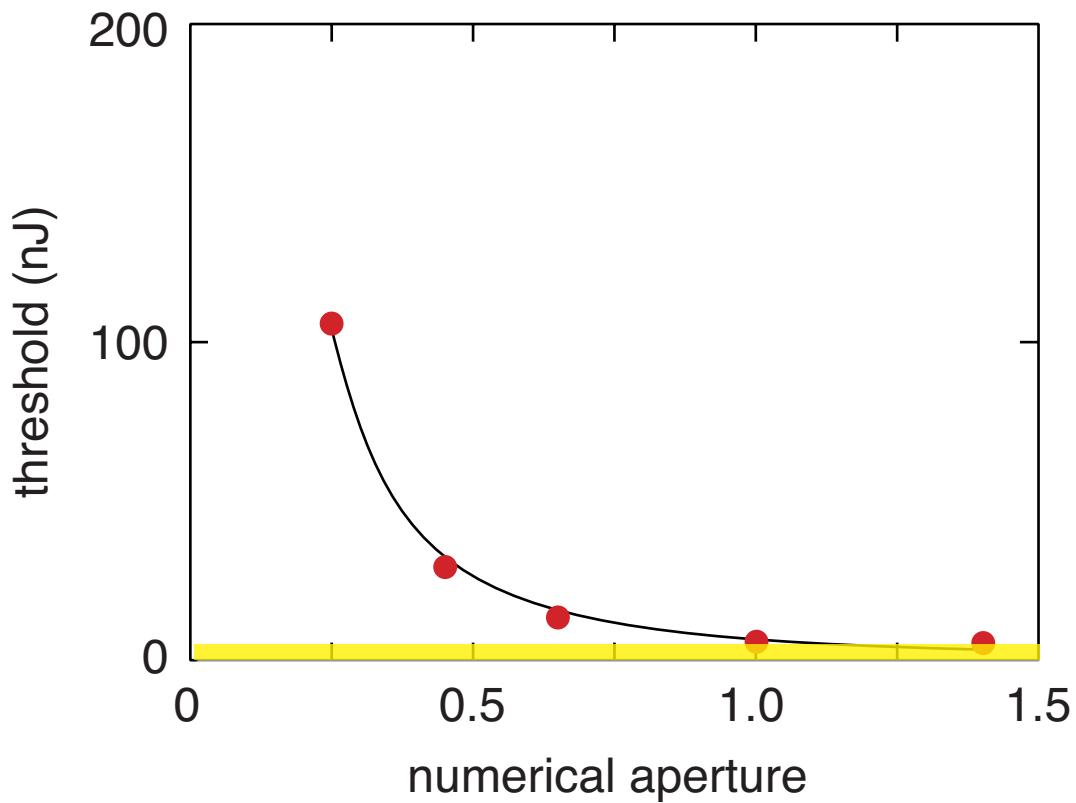
Low-energy machining

threshold decreases with increasing numerical aperture



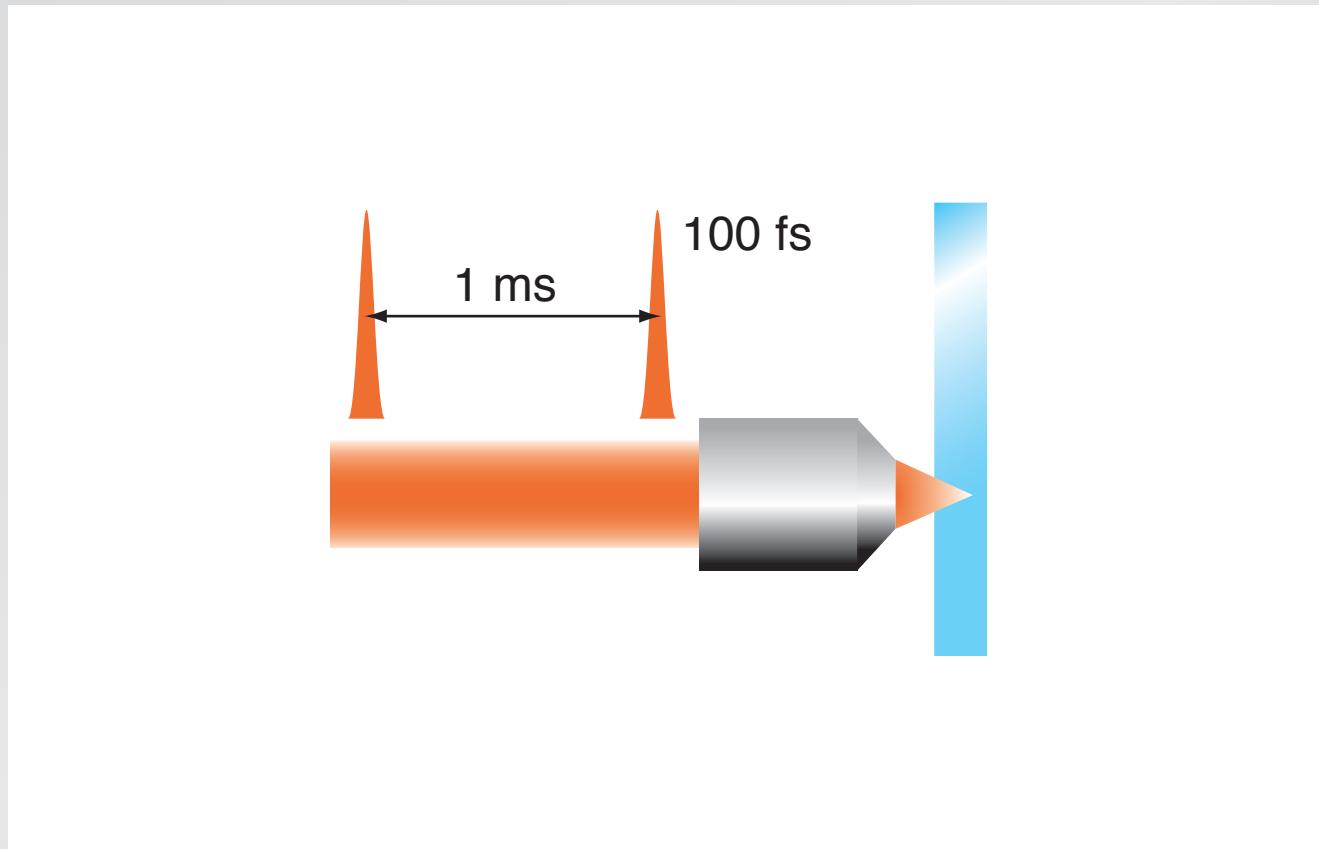
Low-energy machining

less than 10 nJ at high numerical aperture!



Low-energy machining

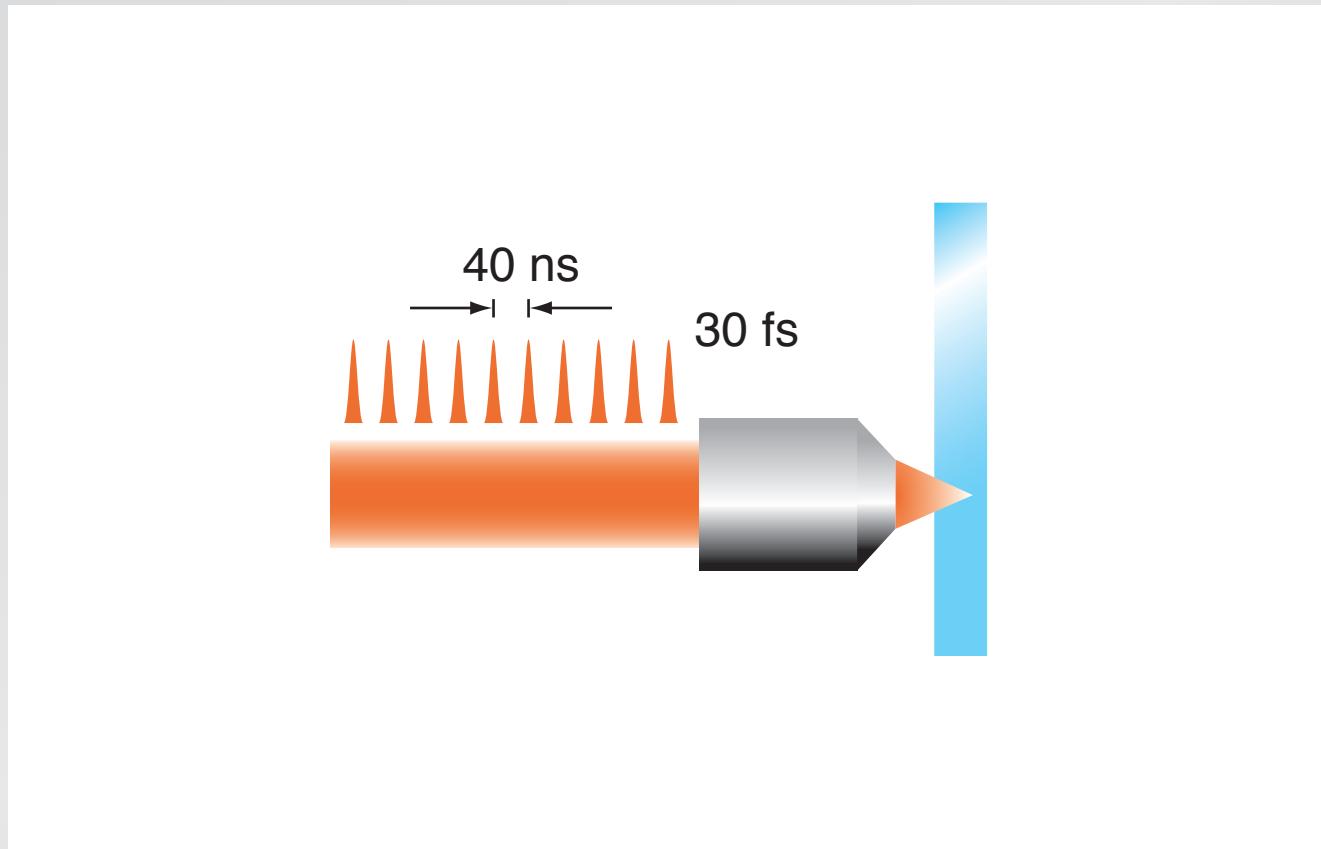
amplified laser: 1 kHz, 1 mJ



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

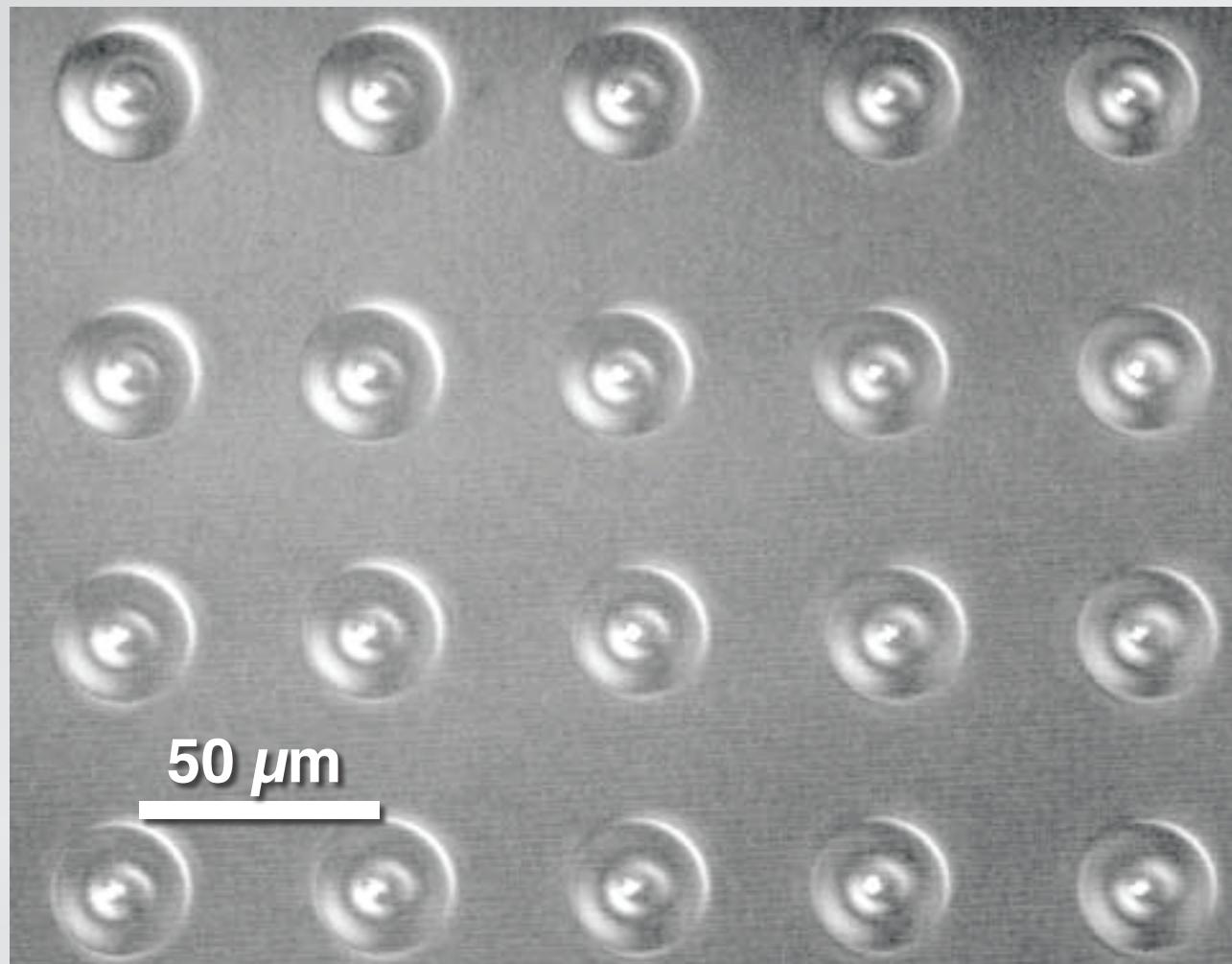
Low-energy machining

long cavity oscillator: 25 MHz, 25 nJ



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

Low-energy machining



Low-energy machining

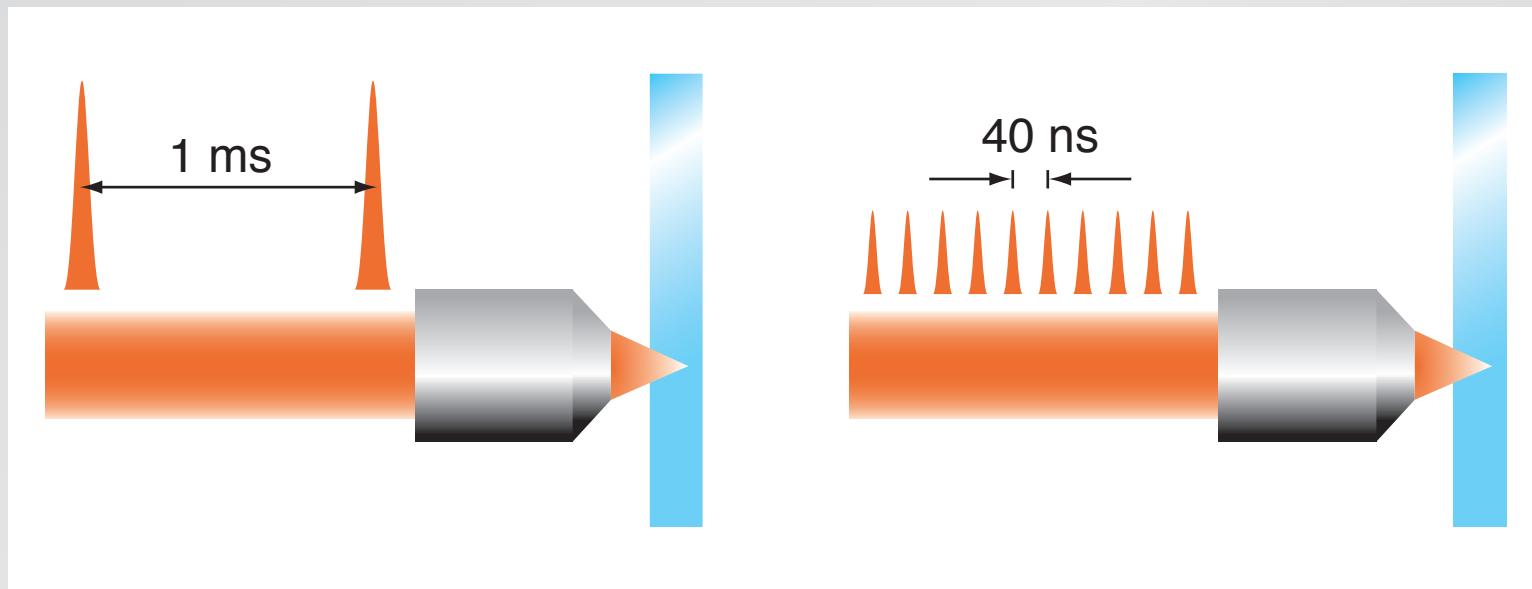
High repetition-rate micromachining:

- structural changes exceed focal volume
- spherical structures
- density change caused by melting

Low-energy machining

amplified laser

oscillator



repetitive

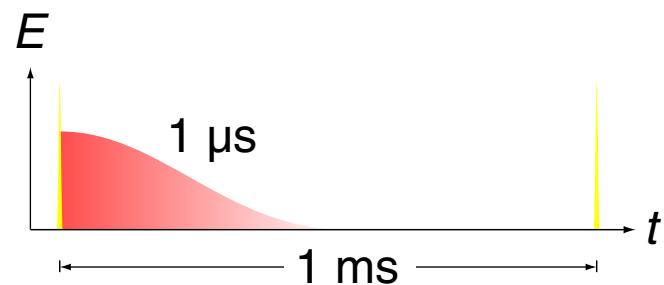
cumulative

Low-energy machining

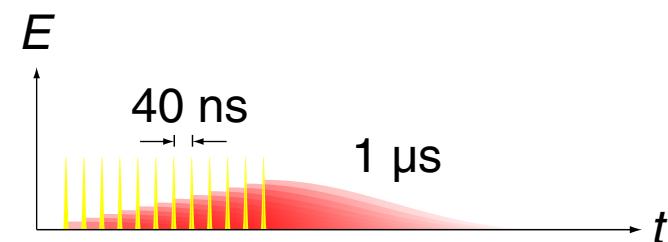
amplified laser

oscillator

low repetition rate



high repetition rate

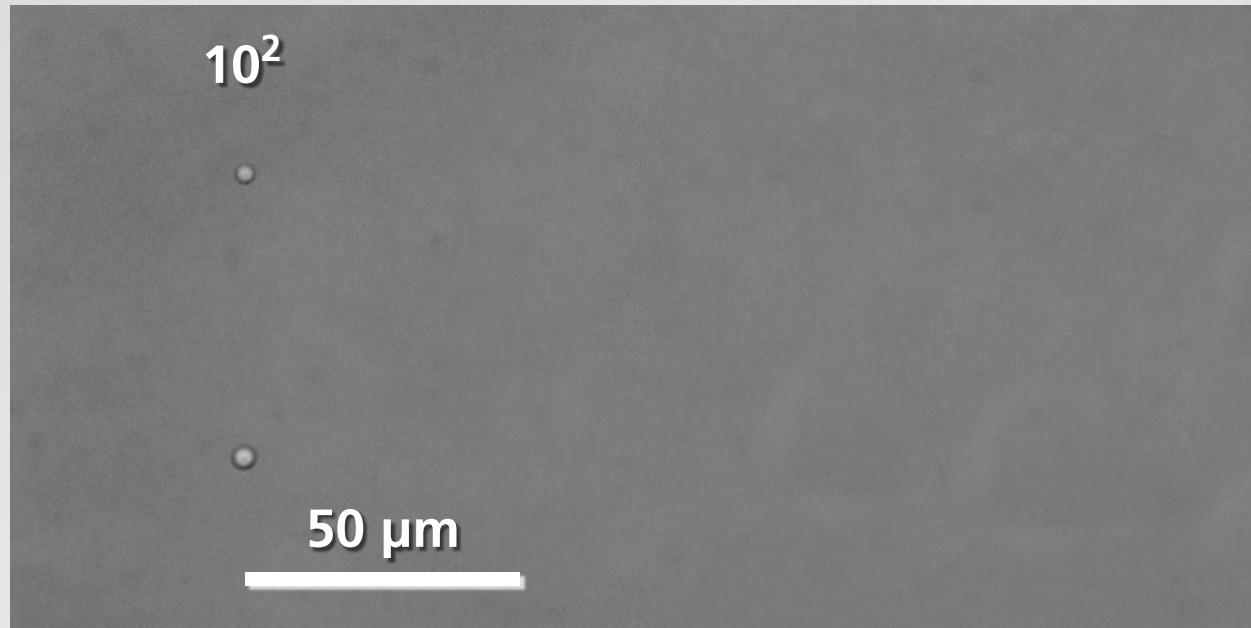


repetitive

cumulative

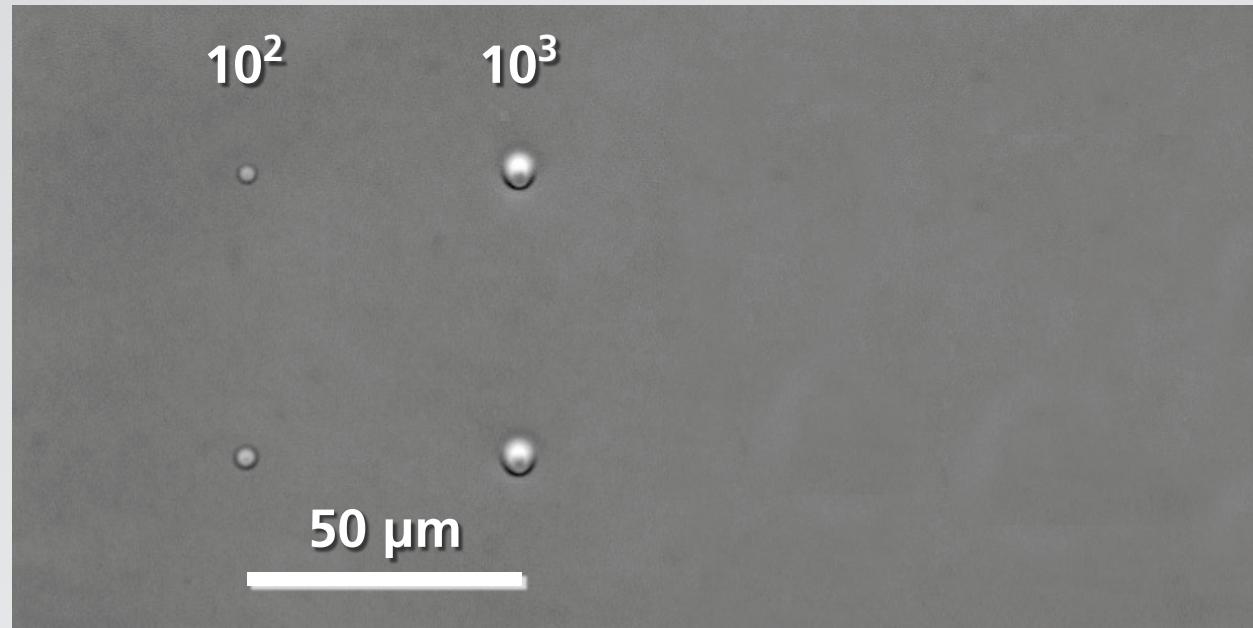
Low-energy machining

the longer the irradiation...



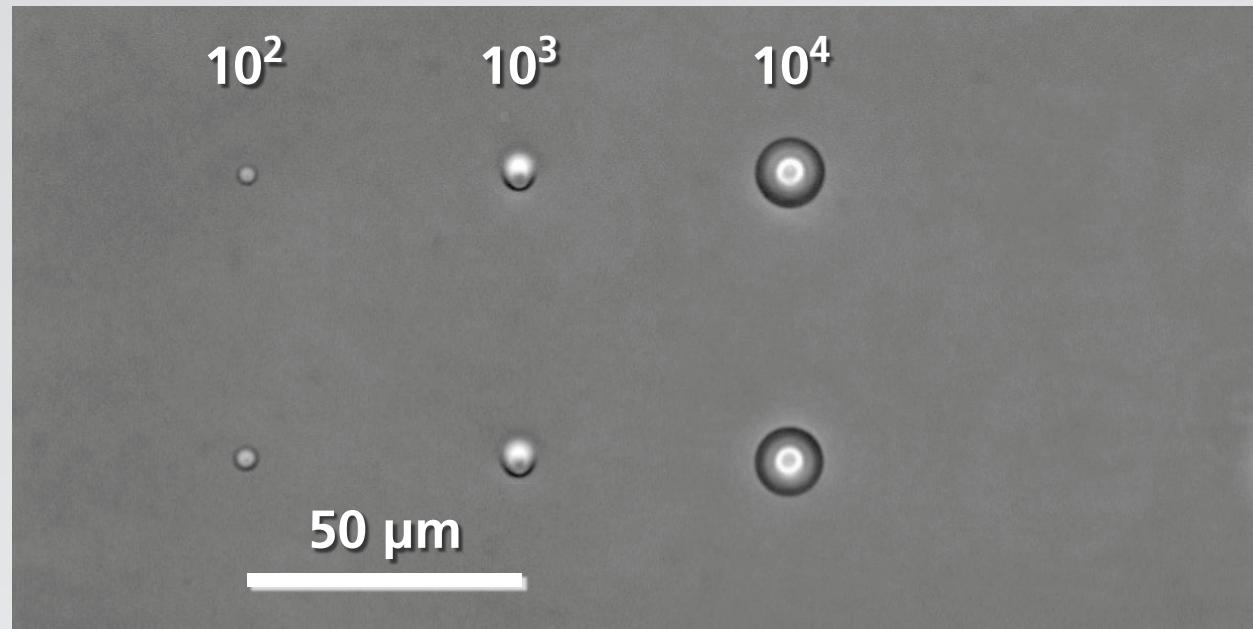
Low-energy machining

the longer the irradiation...



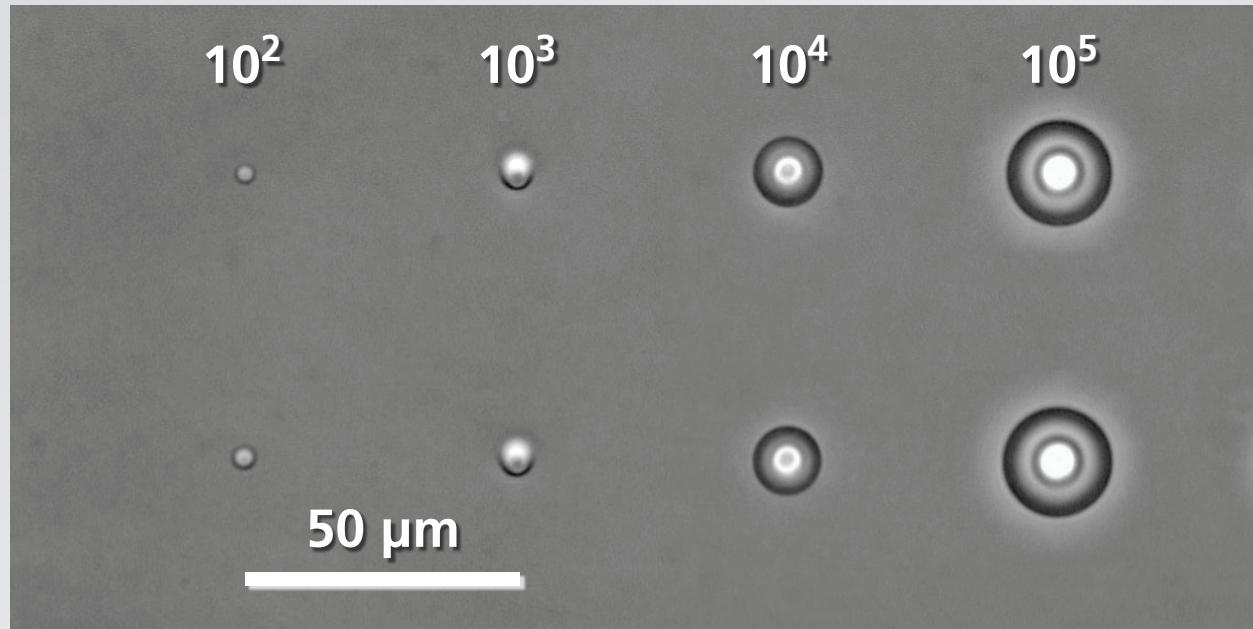
Low-energy machining

the longer the irradiation...



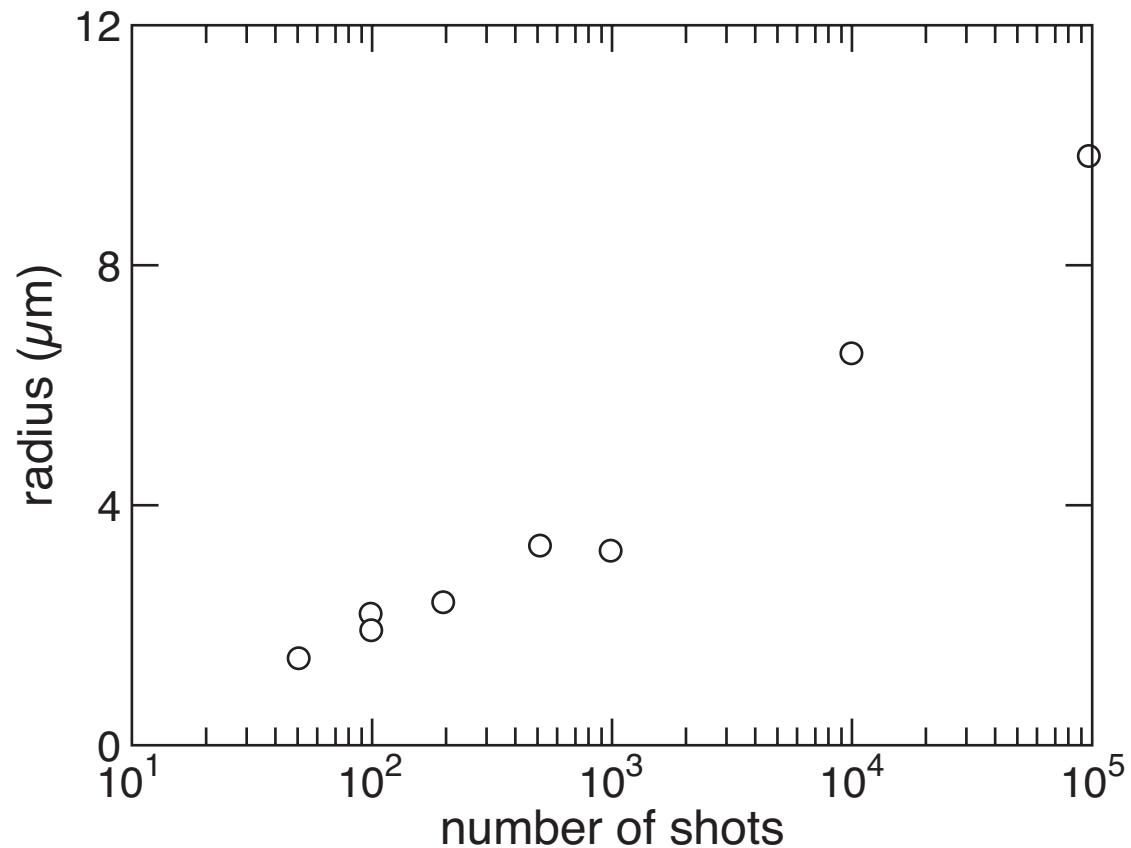
Low-energy machining

the longer the irradiation...



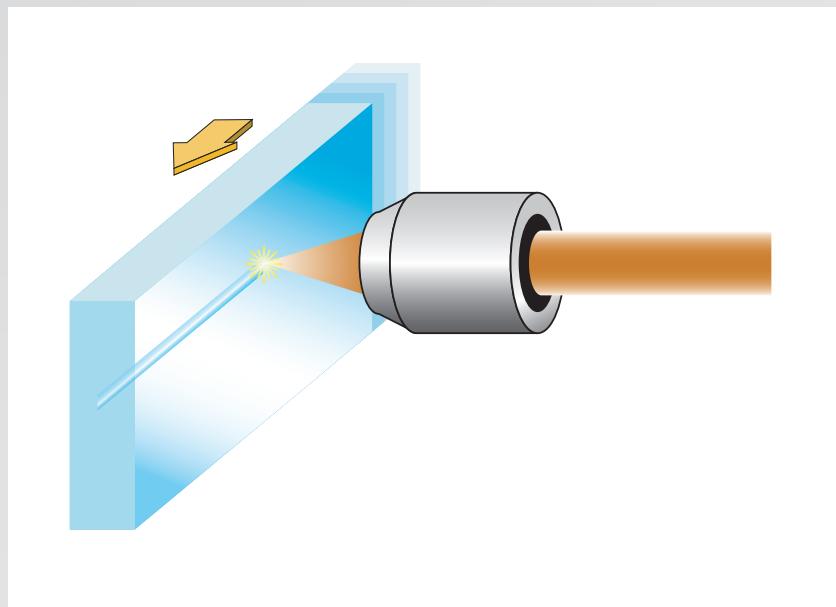
... the larger the radius

Low-energy machining



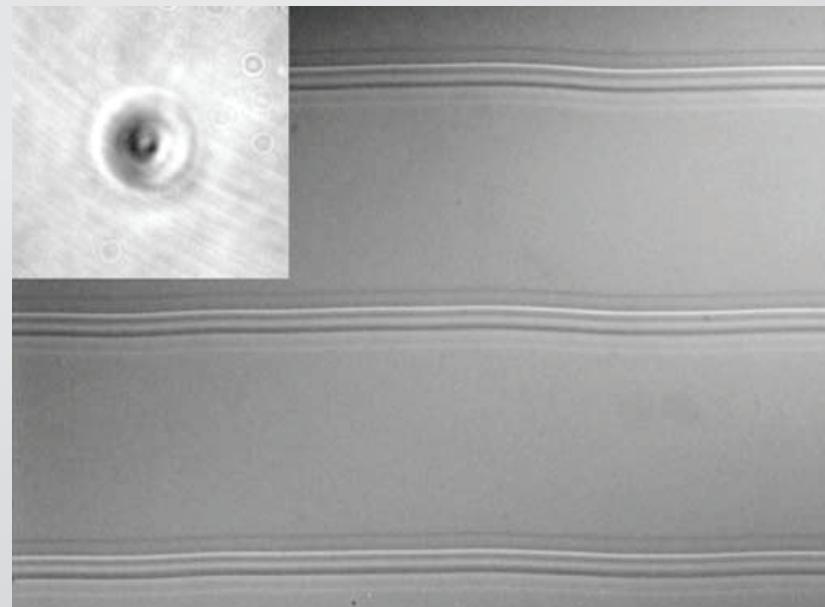
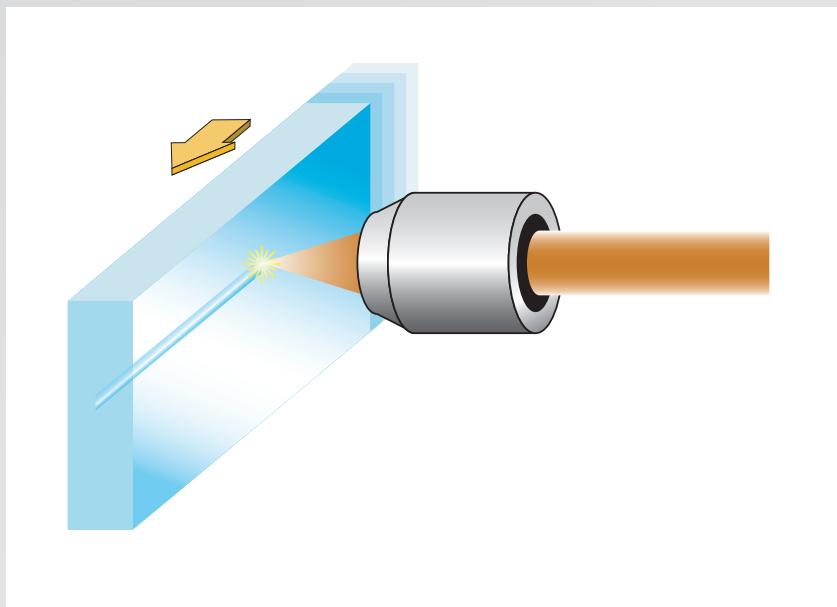
Low-energy machining

waveguide micromachining



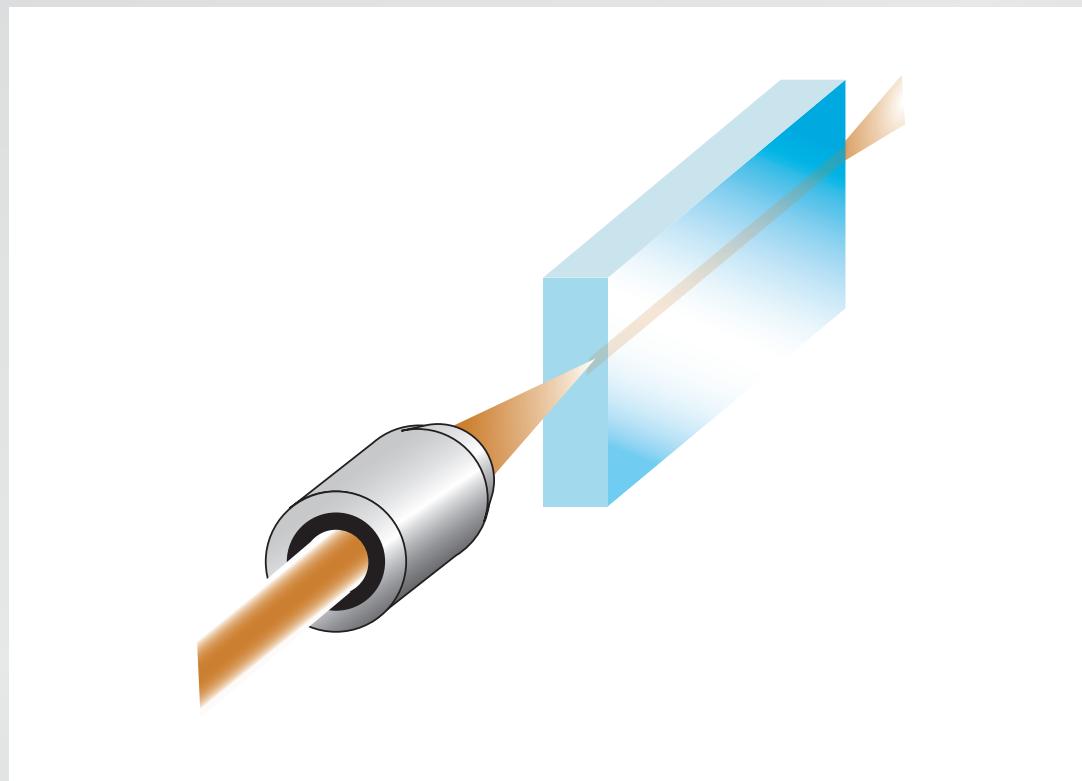
Low-energy machining

waveguide micromachining



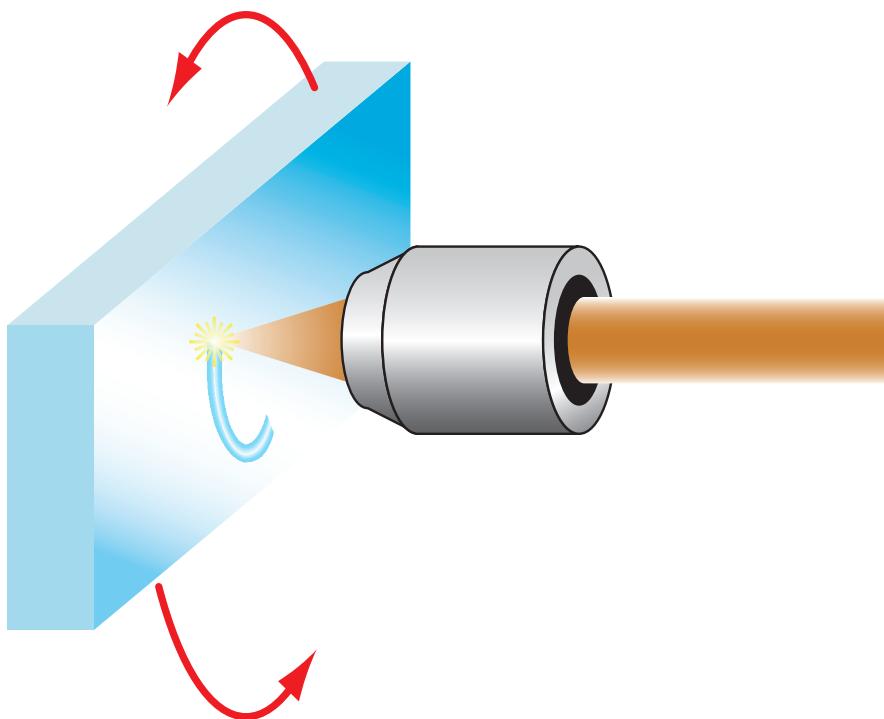
Low-energy machining

structures guide light



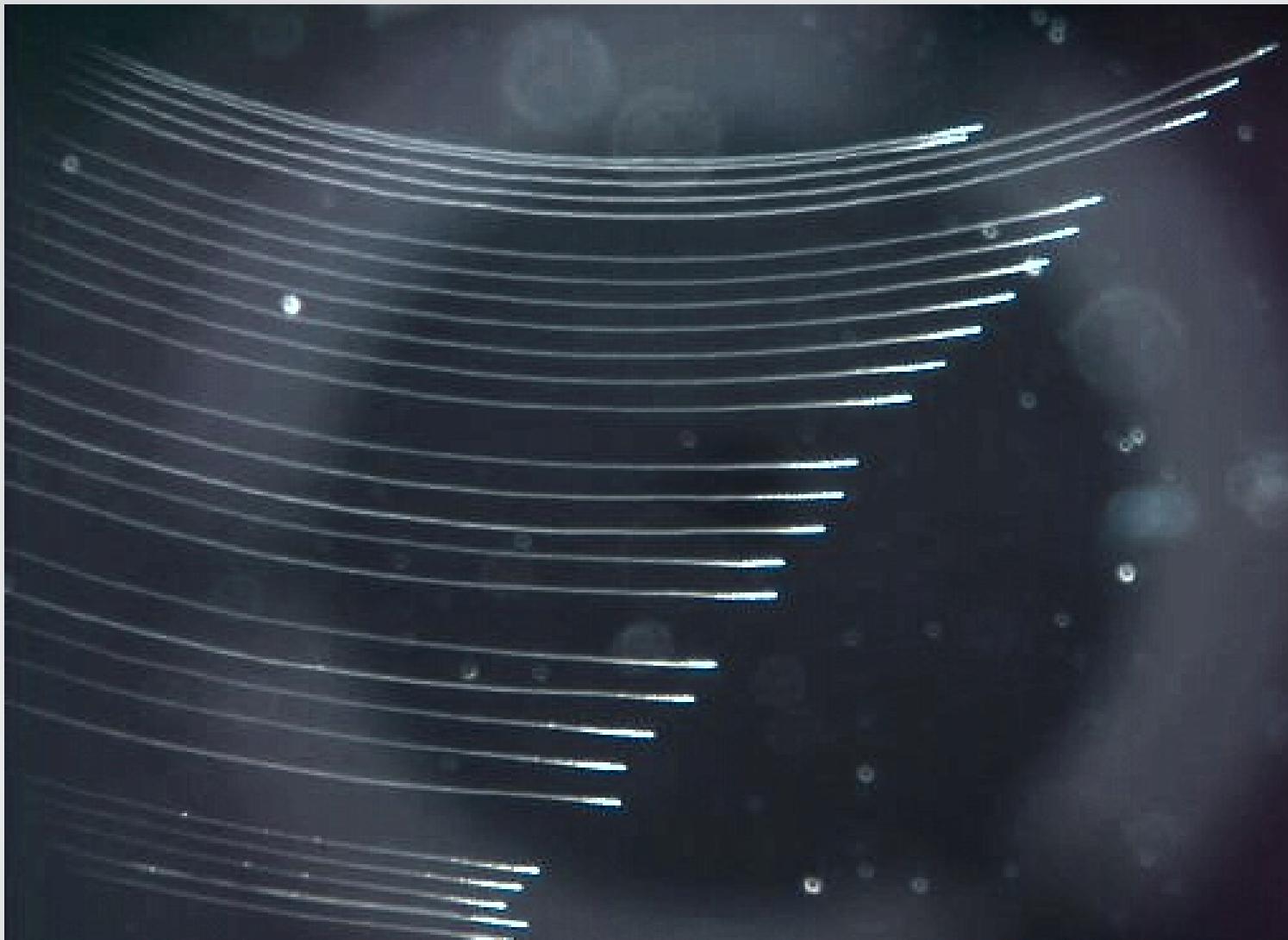
Applications

curved waveguides



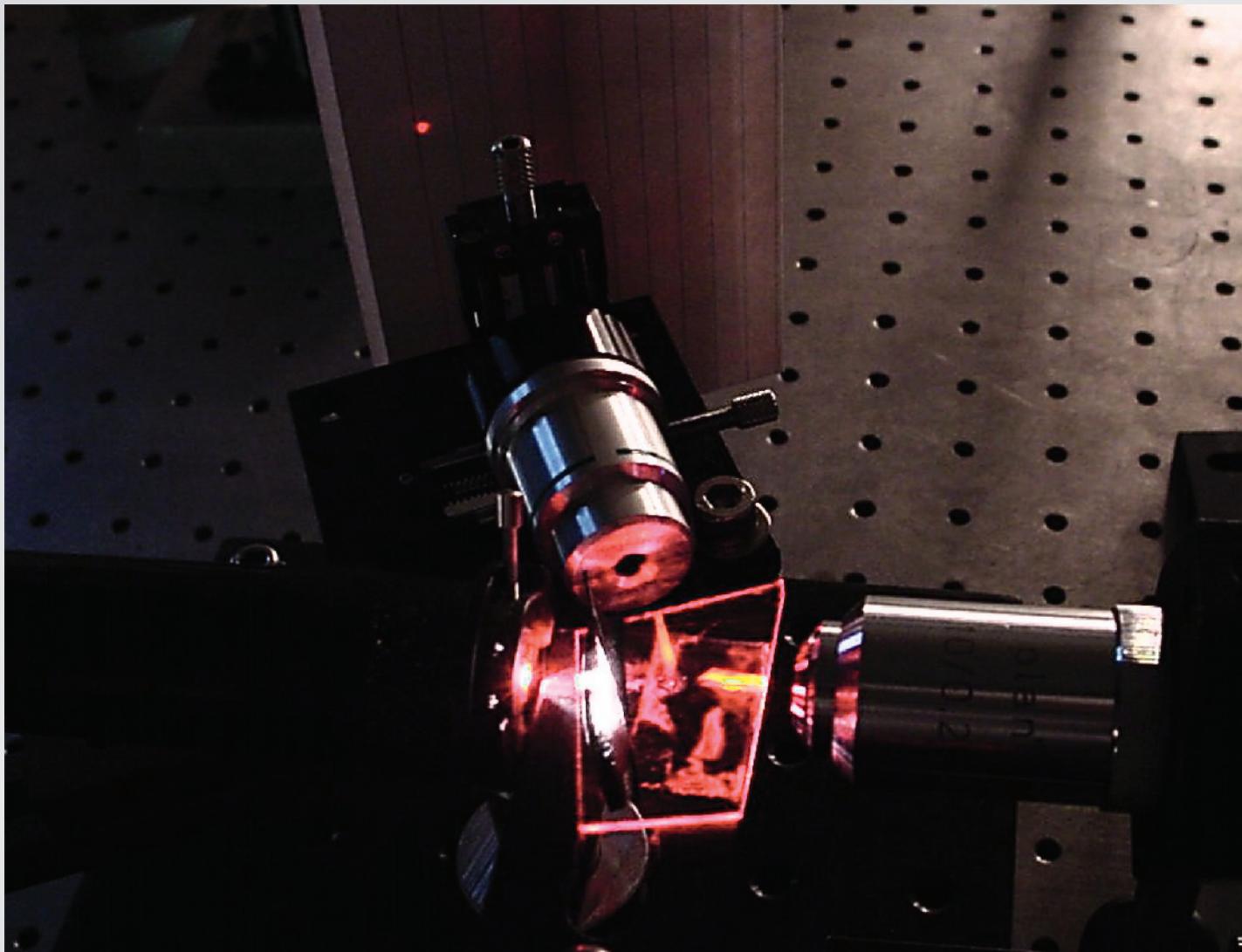
Applications

curved waveguides



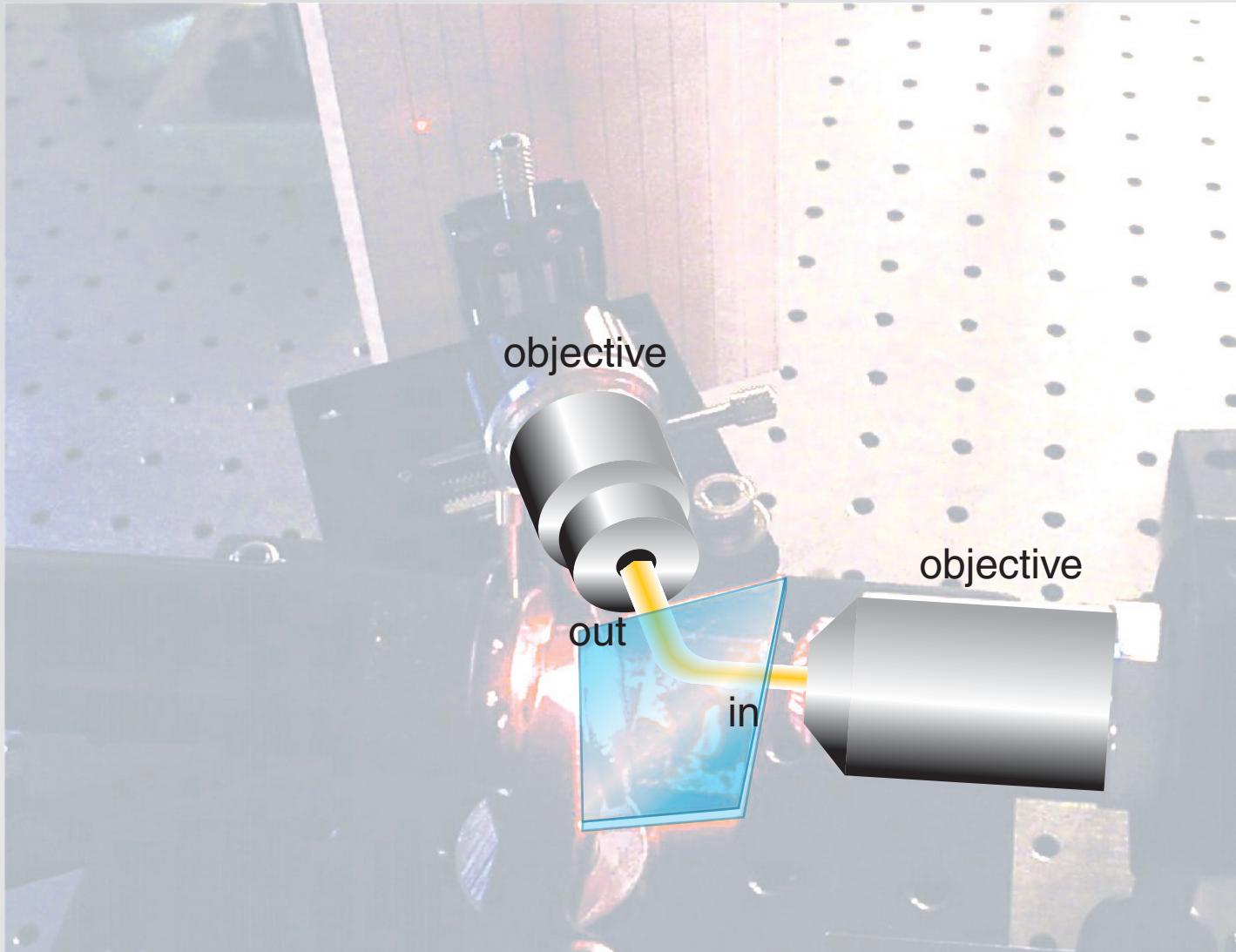
Applications

curved waveguides



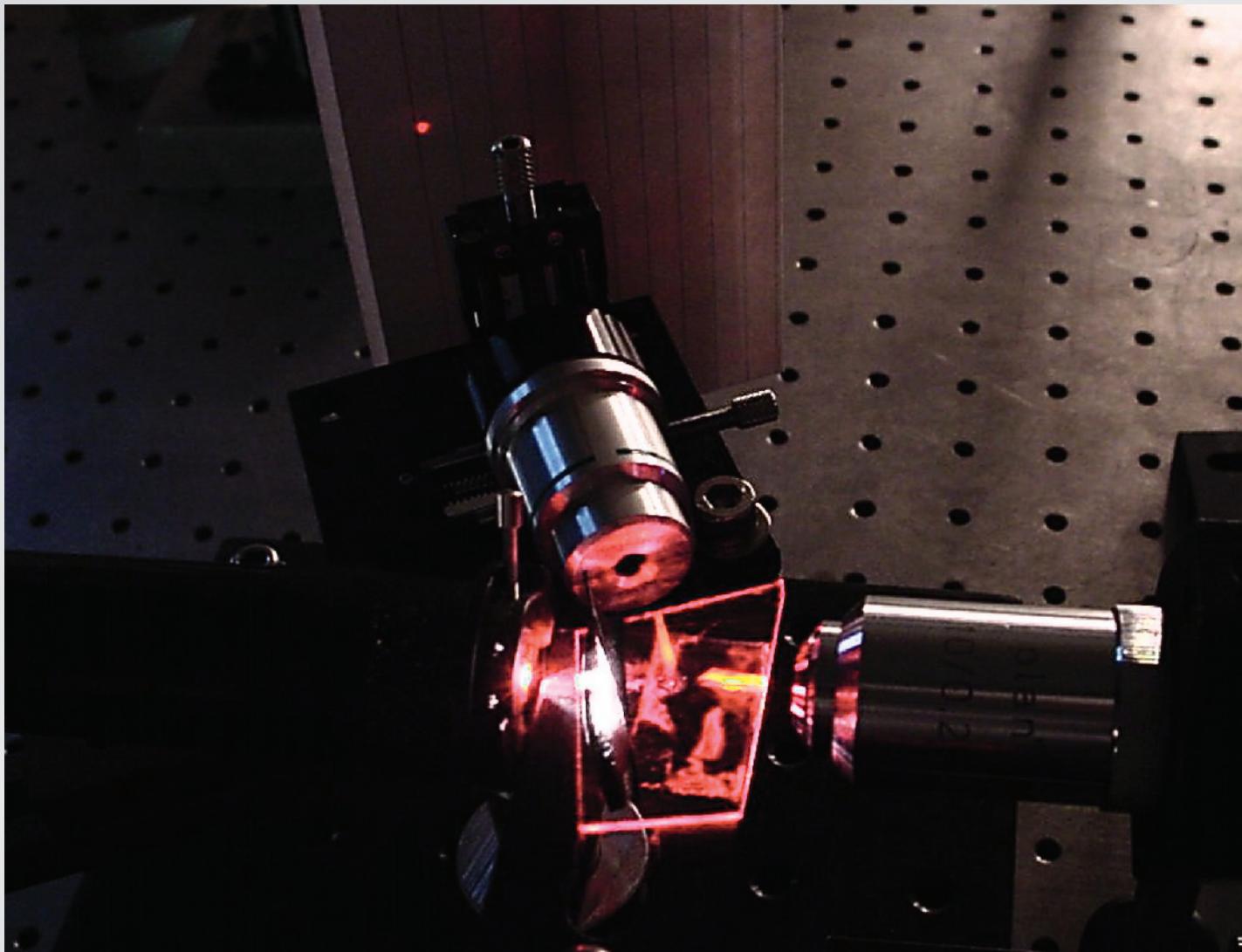
Applications

curved waveguides



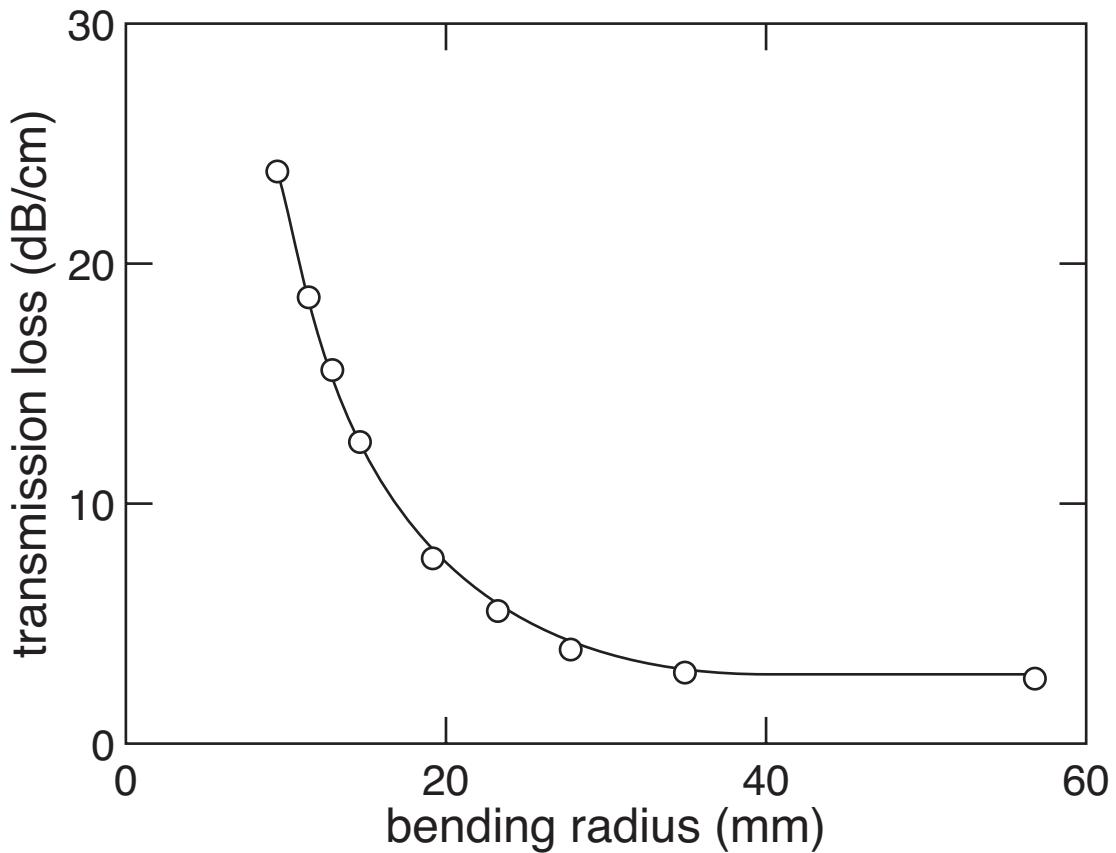
Applications

curved waveguides



Applications

curved waveguides



Applications

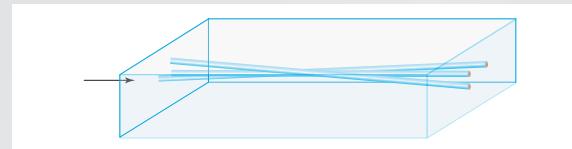
photonic fabrication techniques

	fs micromachining	other
loss (dB/cm)	< 3	0.1–3
bending radius	36 mm	30–40 mm
Δn	2×10^{-3}	$10^{-4} – 0.5$
3D integration	Y	N

Applications

photonic devices

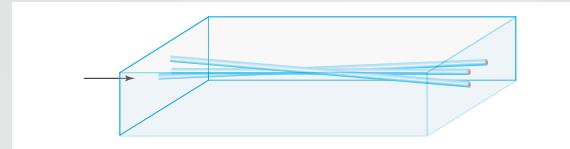
3D splitter



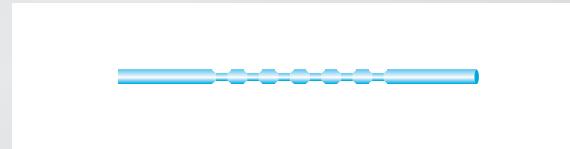
Applications

photonic devices

3D splitter

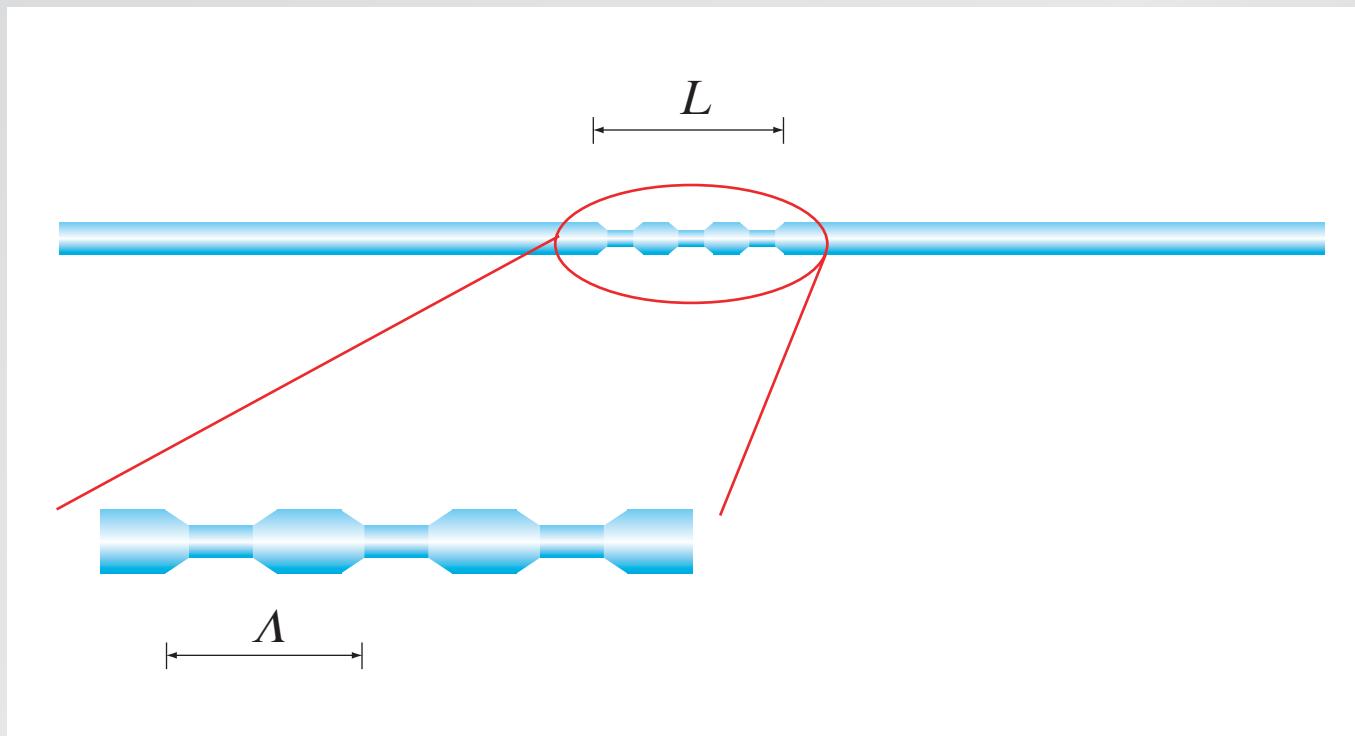


Bragg grating



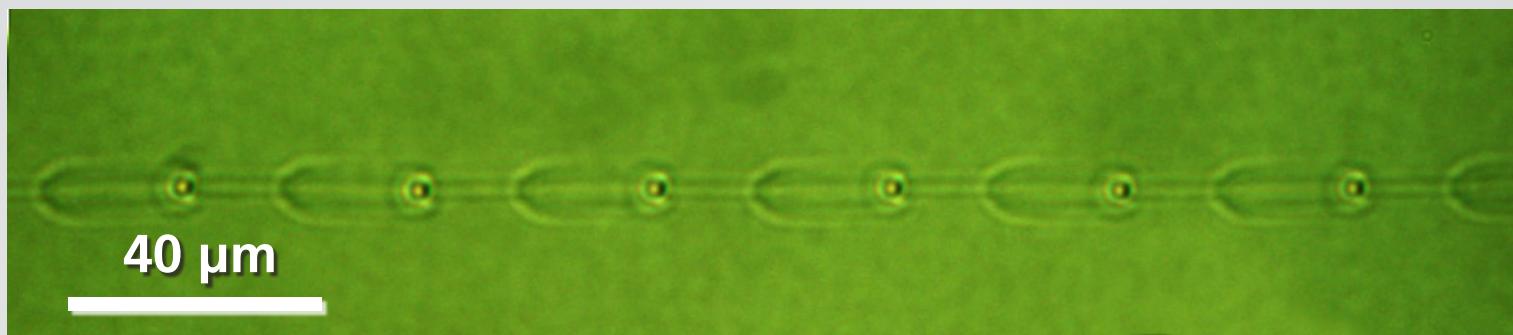
Applications

Bragg grating



Applications

Bragg grating



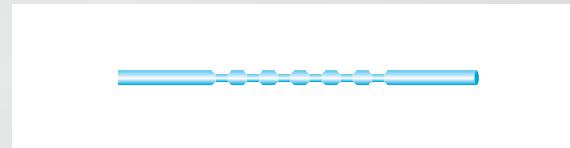
Applications

photonic devices

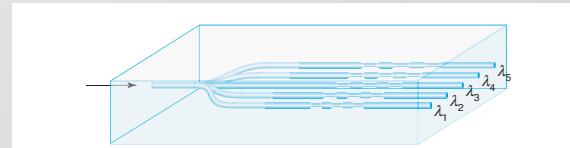
3D splitter



Bragg grating



demultiplexer



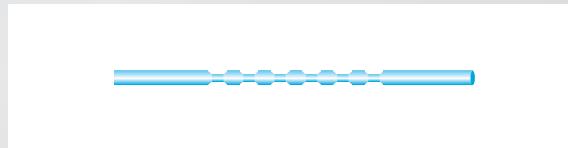
Applications

photonic devices

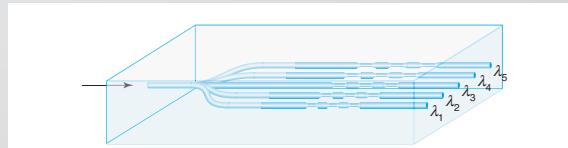
3D splitter



Bragg grating



demultiplexer



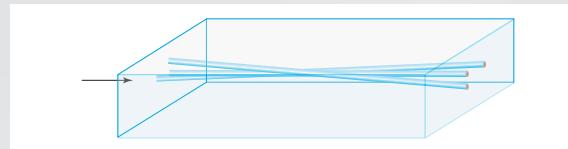
amplifier



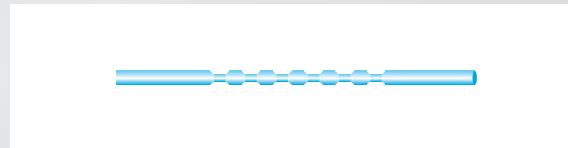
Applications

photonic devices

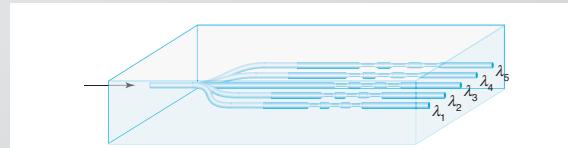
3D splitter



Bragg grating



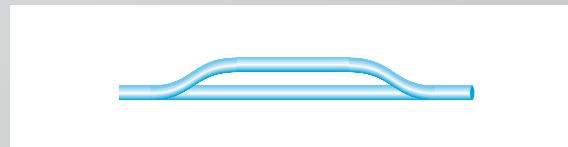
demultiplexer



amplifier

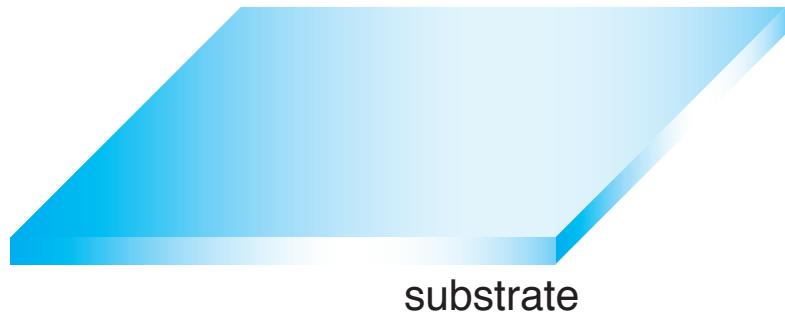


interferometer



Applications

all-optical sensor



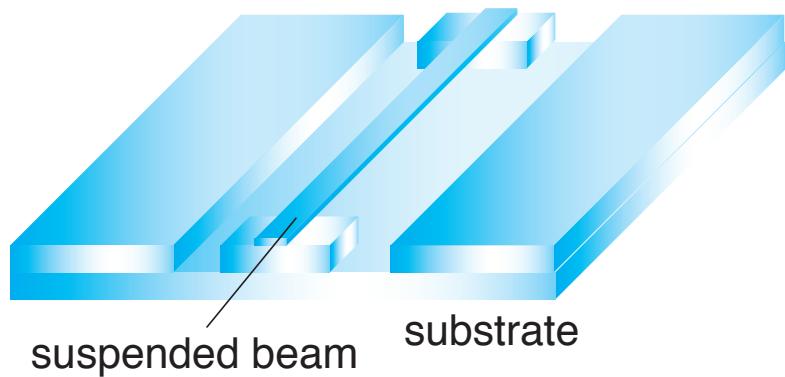
Applications

all-optical sensor



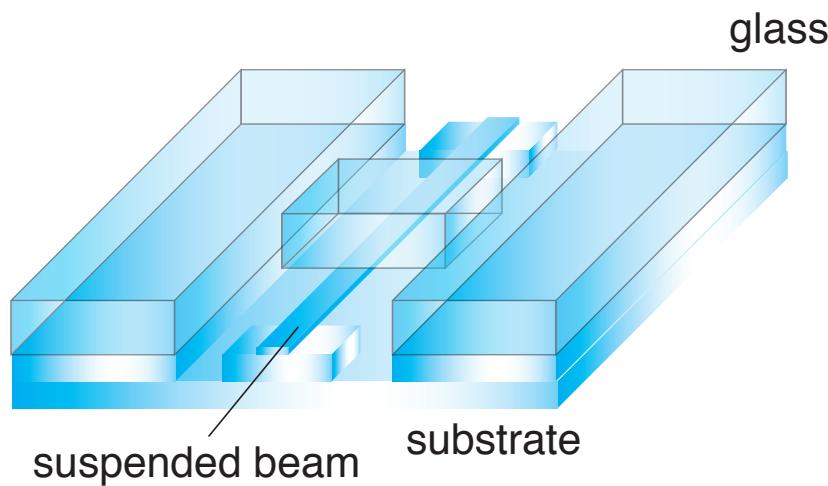
Applications

all-optical sensor



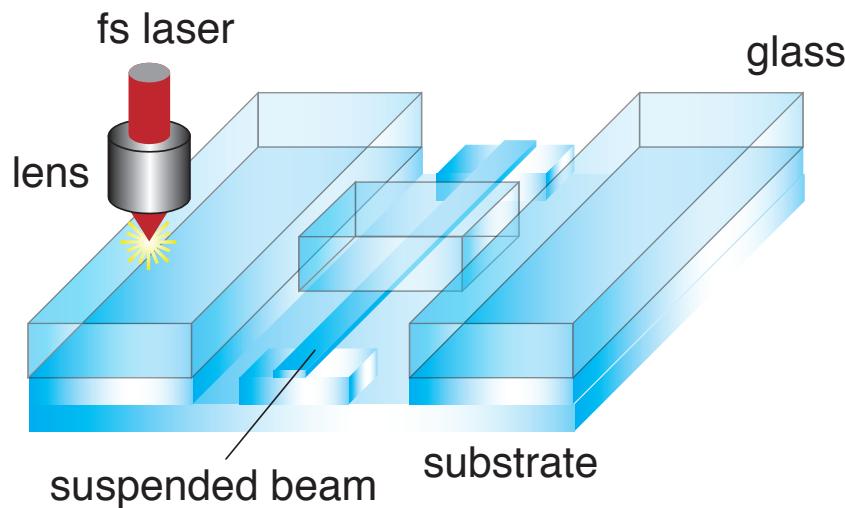
Applications

all-optical sensor



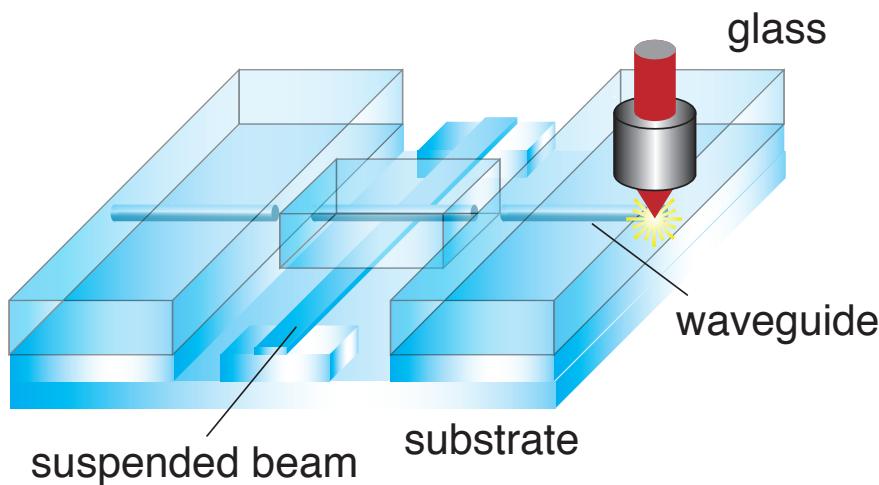
Applications

all-optical sensor



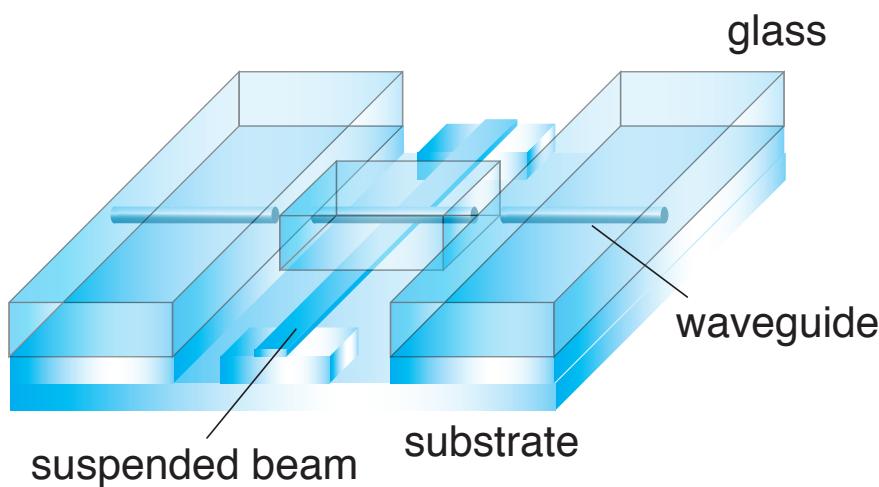
Applications

all-optical sensor



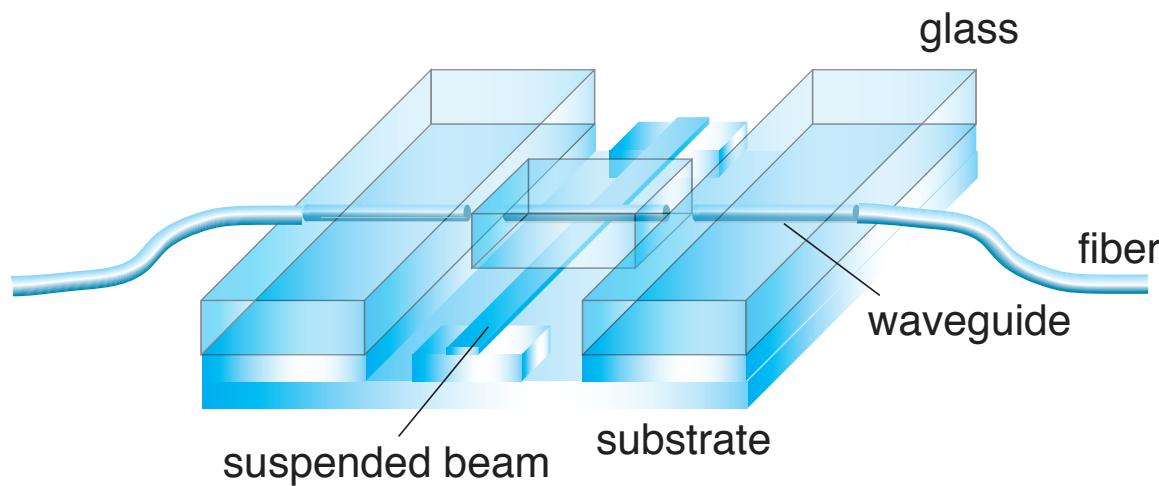
Applications

all-optical sensor

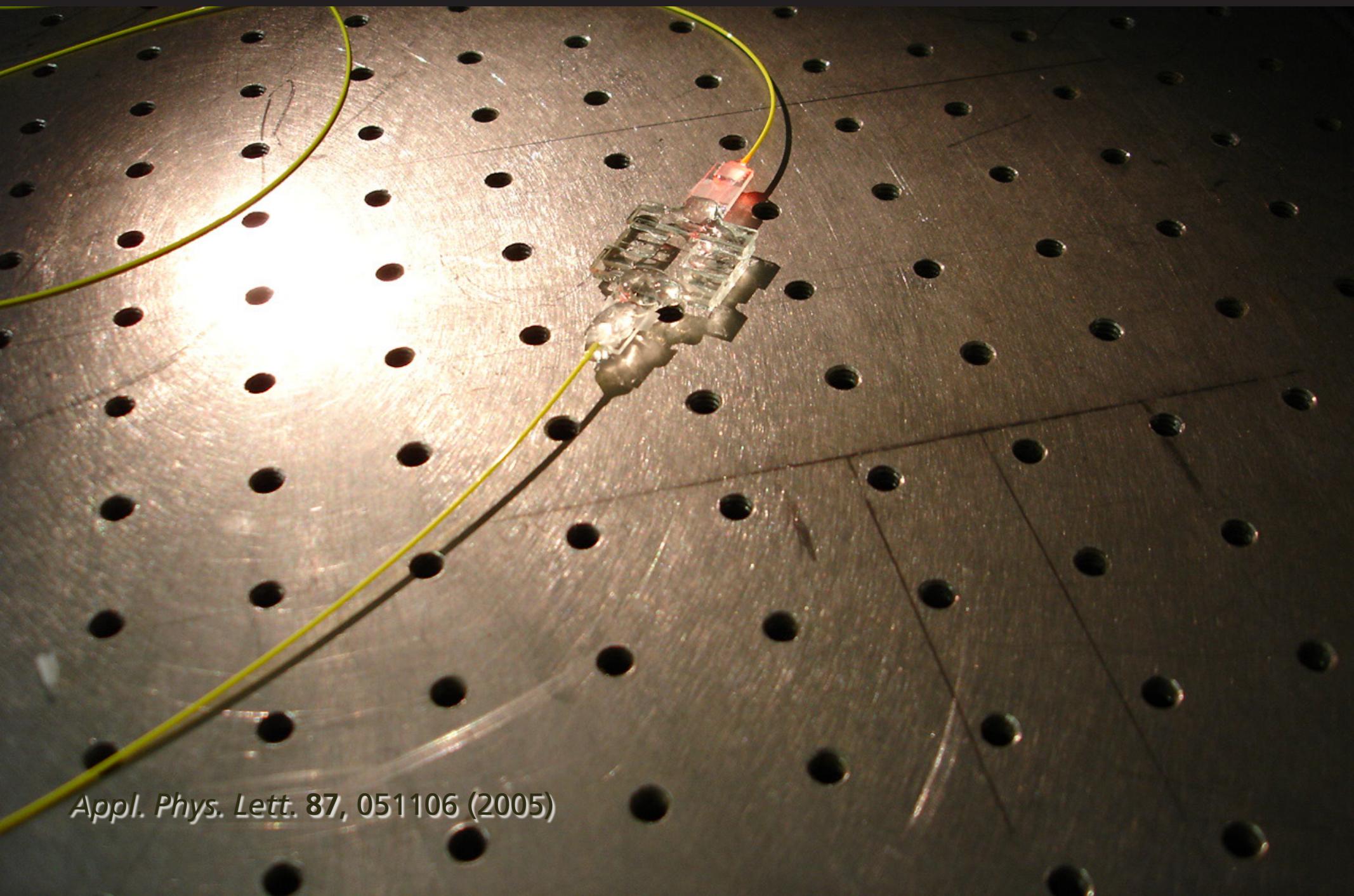


Applications

all-optical sensor



Applications

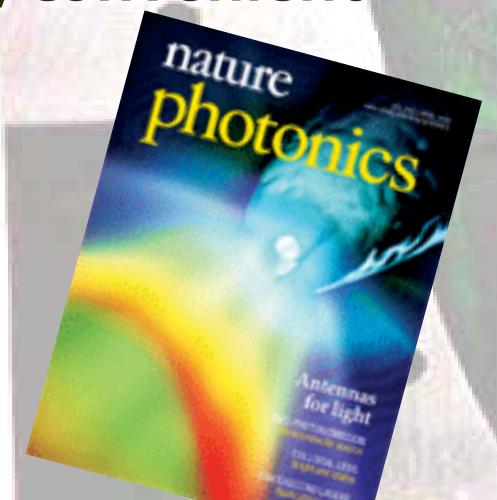


Appl. Phys. Lett. **87**, 051106 (2005)

Summary

- important parameters: focusing, energy, repetition rate
- nearly material independent
- two regimes: low and high repetition rate
- high-repetition rate (thermal) machining fast, convenient

Nature Photonics 2, 219 (2008)







Funding:

Army Research Office

DARPA

Department of Energy

NDSEG

National Science Foundation

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