

OPTICS IN THE (ULTRA)FAST LANE: SCULPTING MATTER WITH LIGHT

Mazur Group

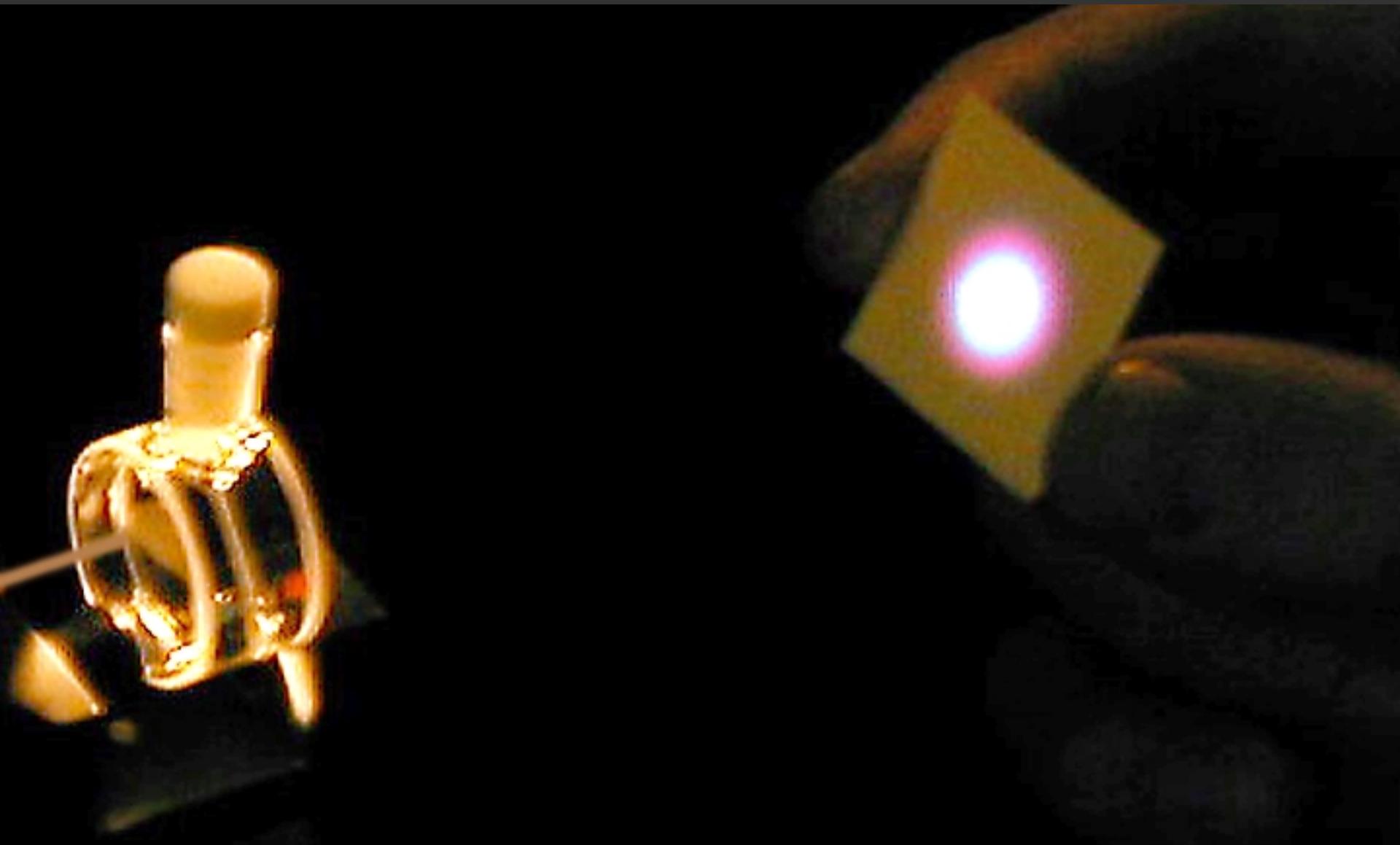
**Narayananamurti/Mazur
Joint Lunch Meeting
3 May 1999**



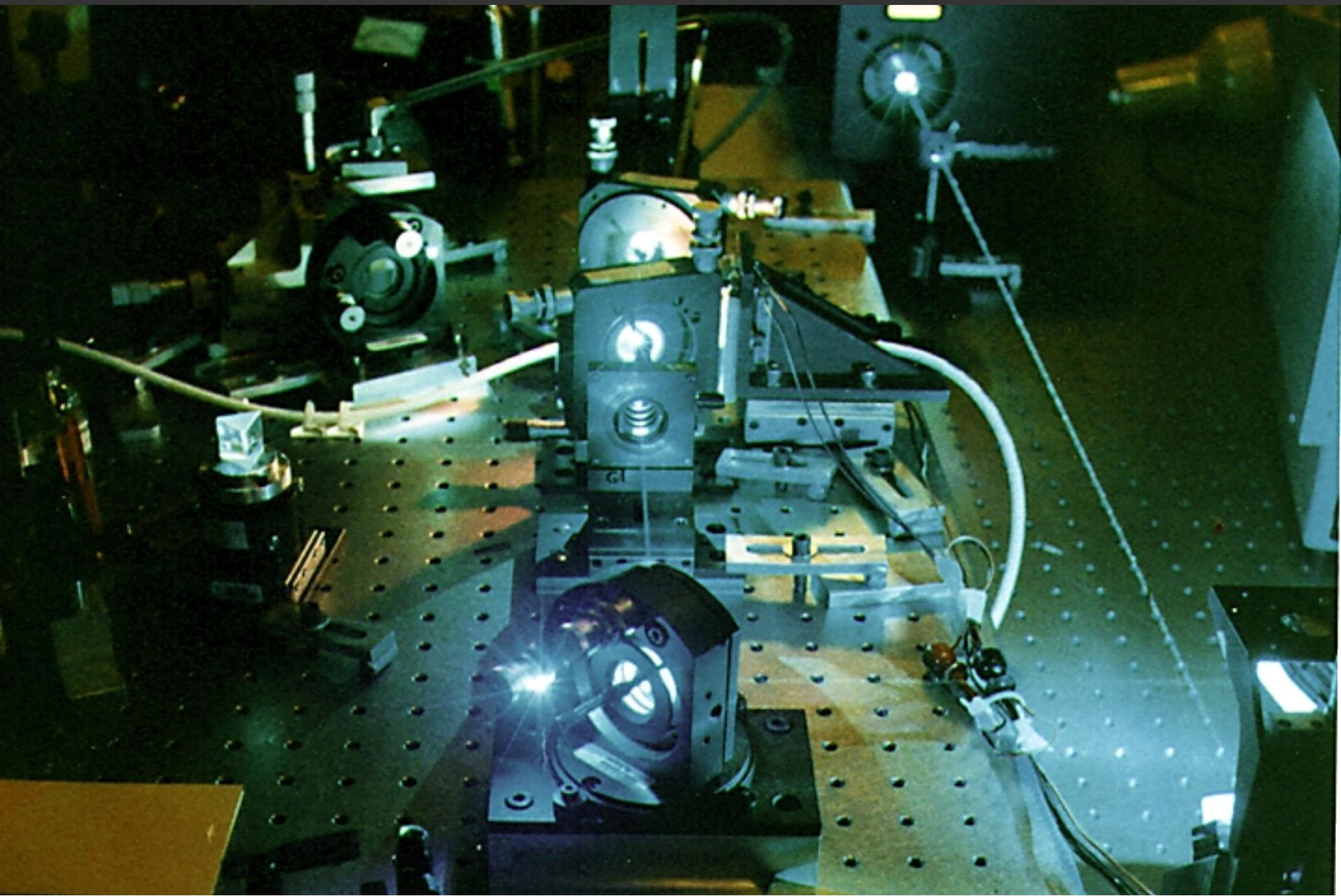
Introduction

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

Introduction

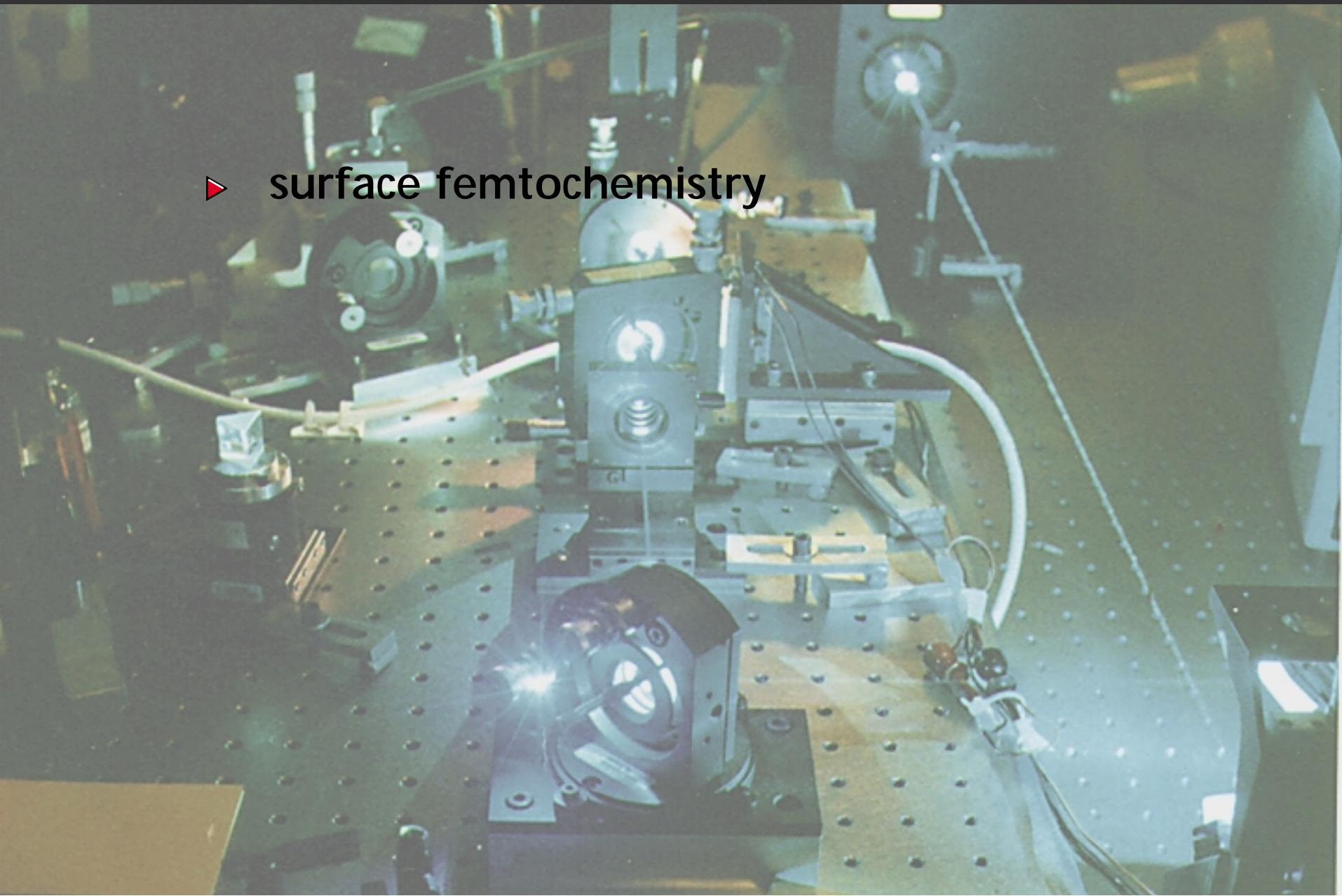


Overview



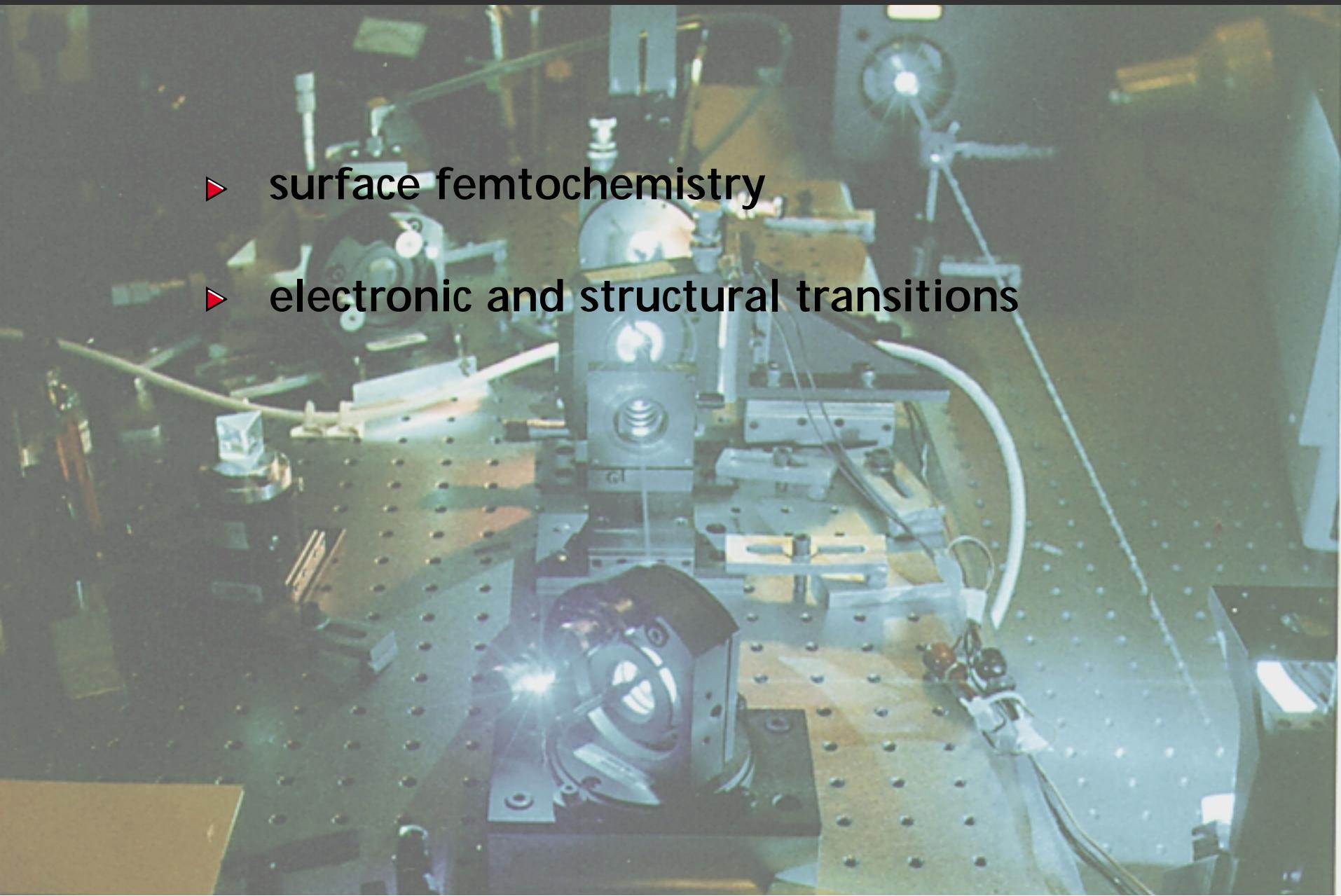
Overview

- ▶ surface femtochemistry



Overview

- ▶ surface femtochemistry
- ▶ electronic and structural transitions



Overview

- ▶ surface femtochemistry
- ▶ electronic and structural transitions
- ▶ microstructuring of materials



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- ▶ electronic and structural transitions
- ▶ microstructuring of materials
- ▶ nonlinear optics and propagation

Overview

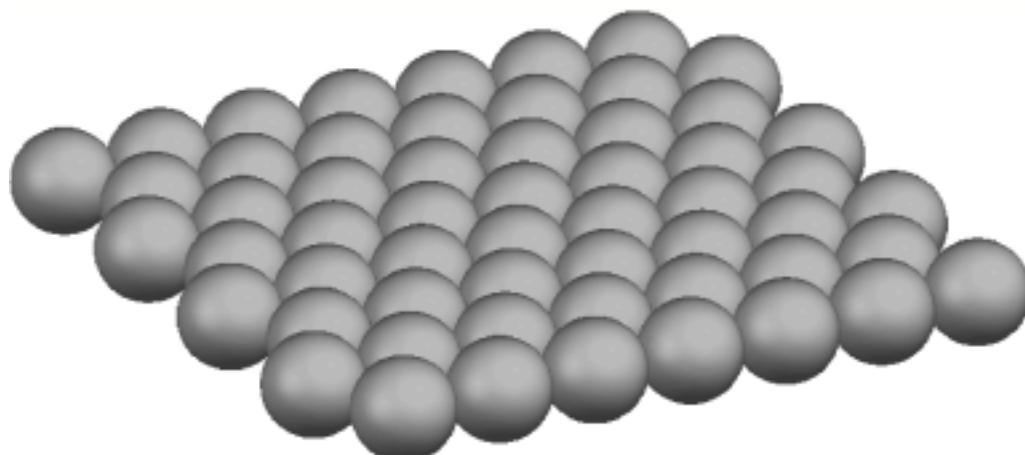
- ▶ surface femtochemistry
- ▶ electronic and structural transitions
- ▶ microstructuring of materials
- ▶ nonlinear optics and propagation
- ▶ laser surgery

Overview

- ▶ surface femtochemistry
- ▶ electronic and structural transitions
- ▶ microstructuring of materials
- ▶ nonlinear optics and propagation
- ▶ laser surgery

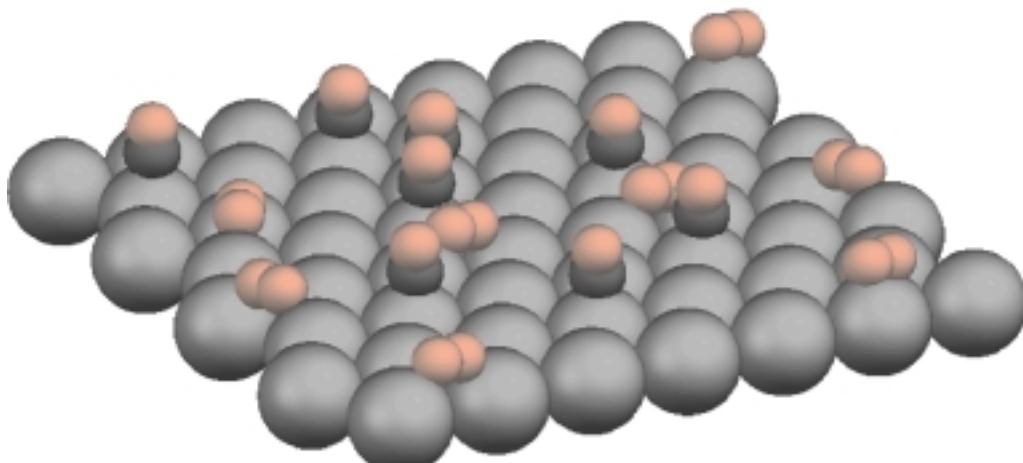
Surface femtochemistry

clean Pt(111) surface at 90 K...



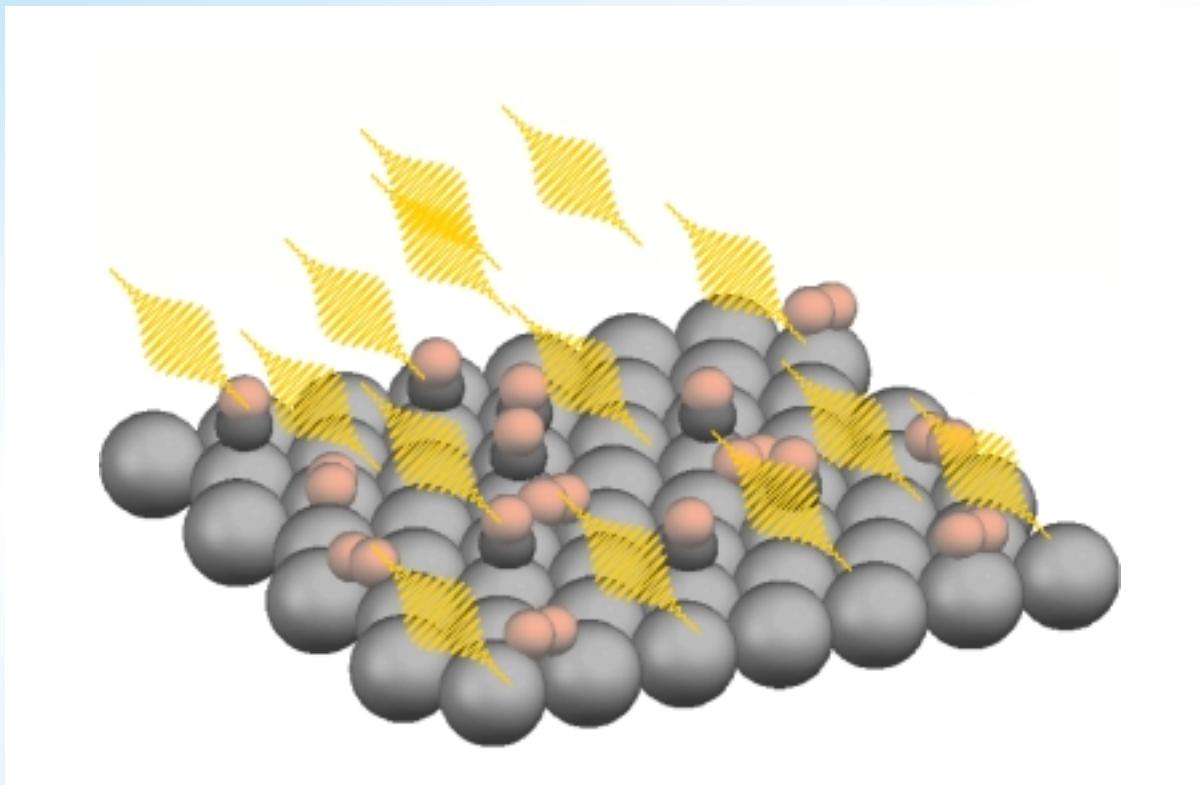
Surface femtochemistry

dose with O₂ and CO...



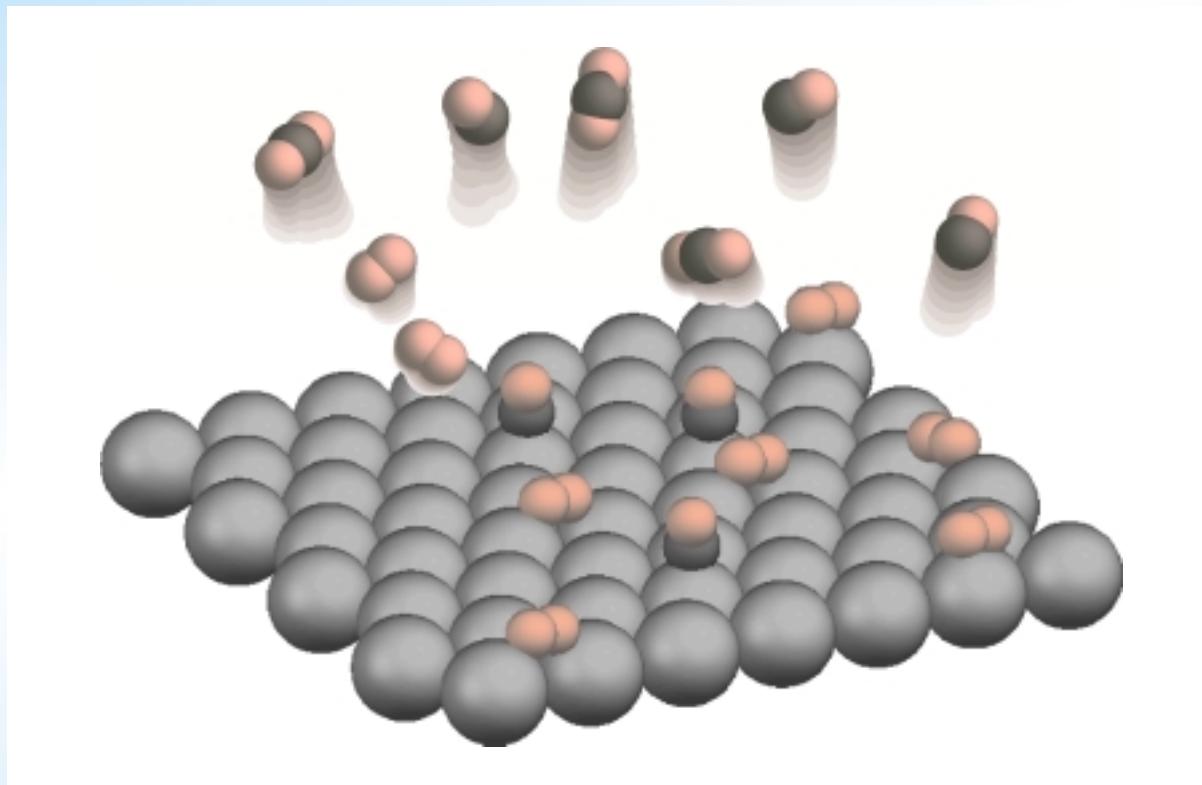
Surface femtochemistry

photons excite sample...



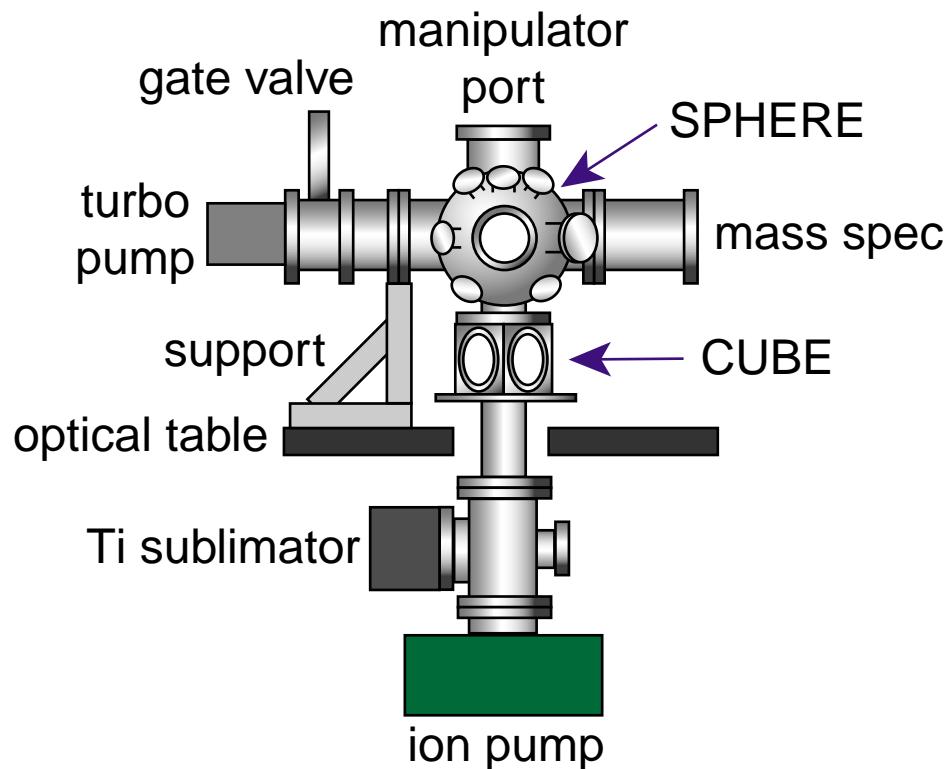
Surface femtochemistry

breaking and rearranging bonds



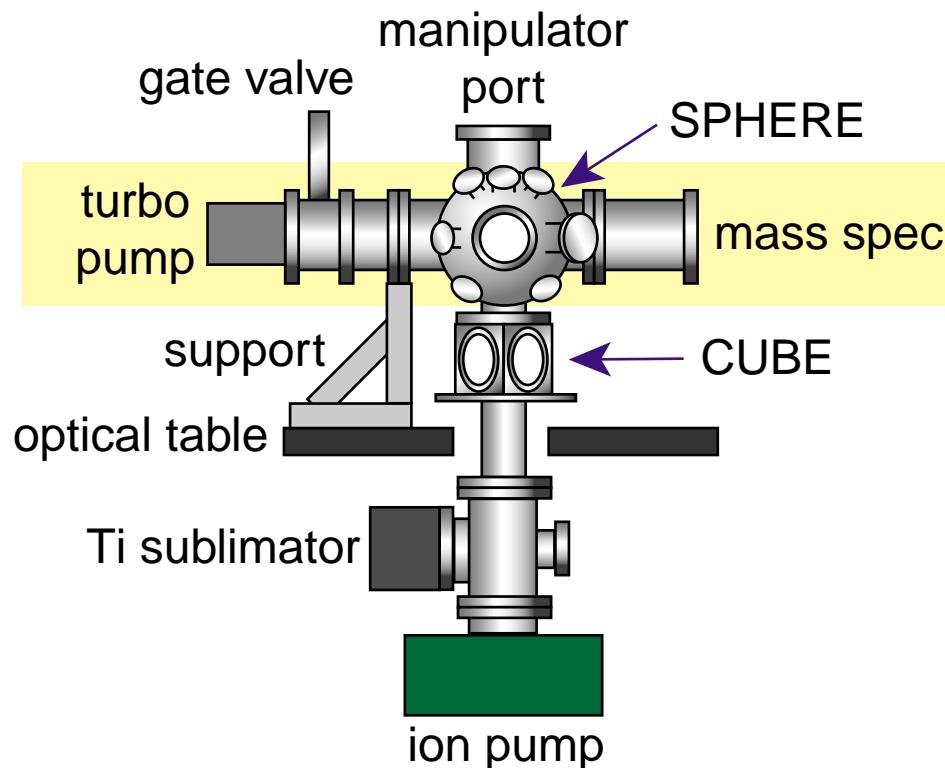
Surface femtochemistry

UHV chamber side view



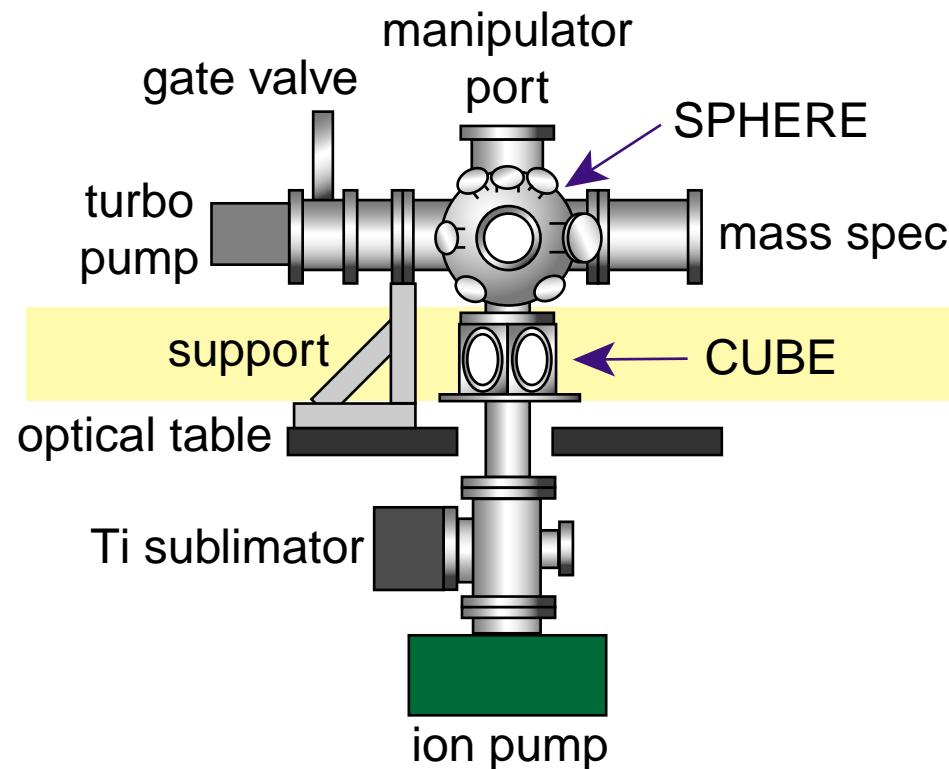
Surface femtochemistry

UHV chamber side view

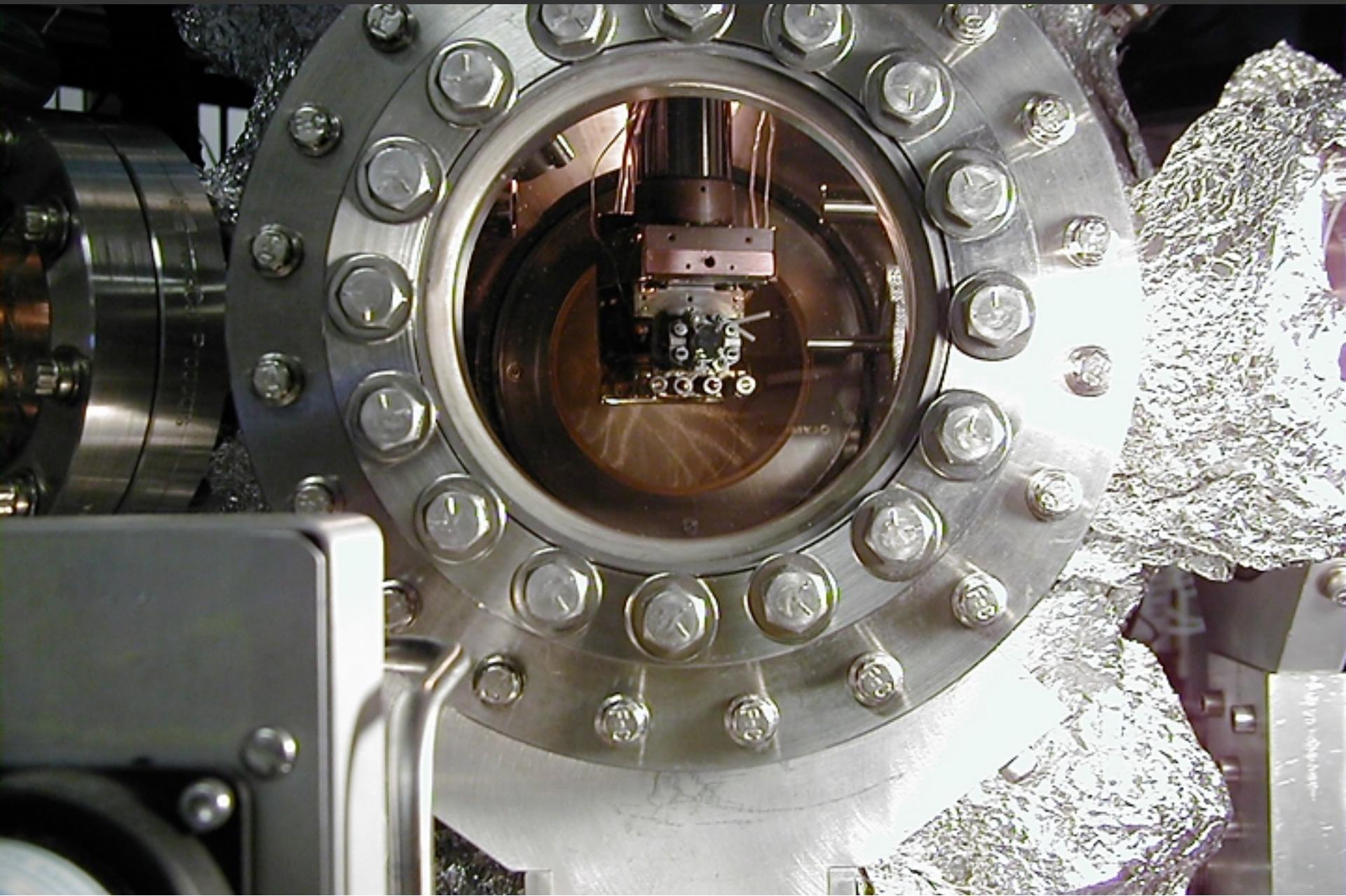


Surface femtochemistry

UHV chamber side view

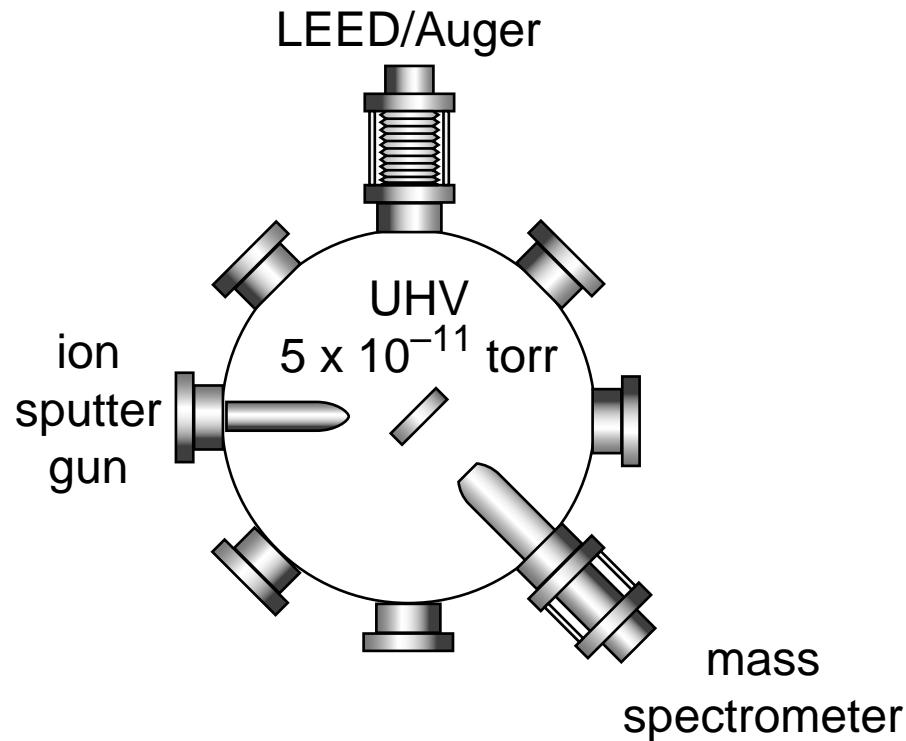


Surface femtochemistry

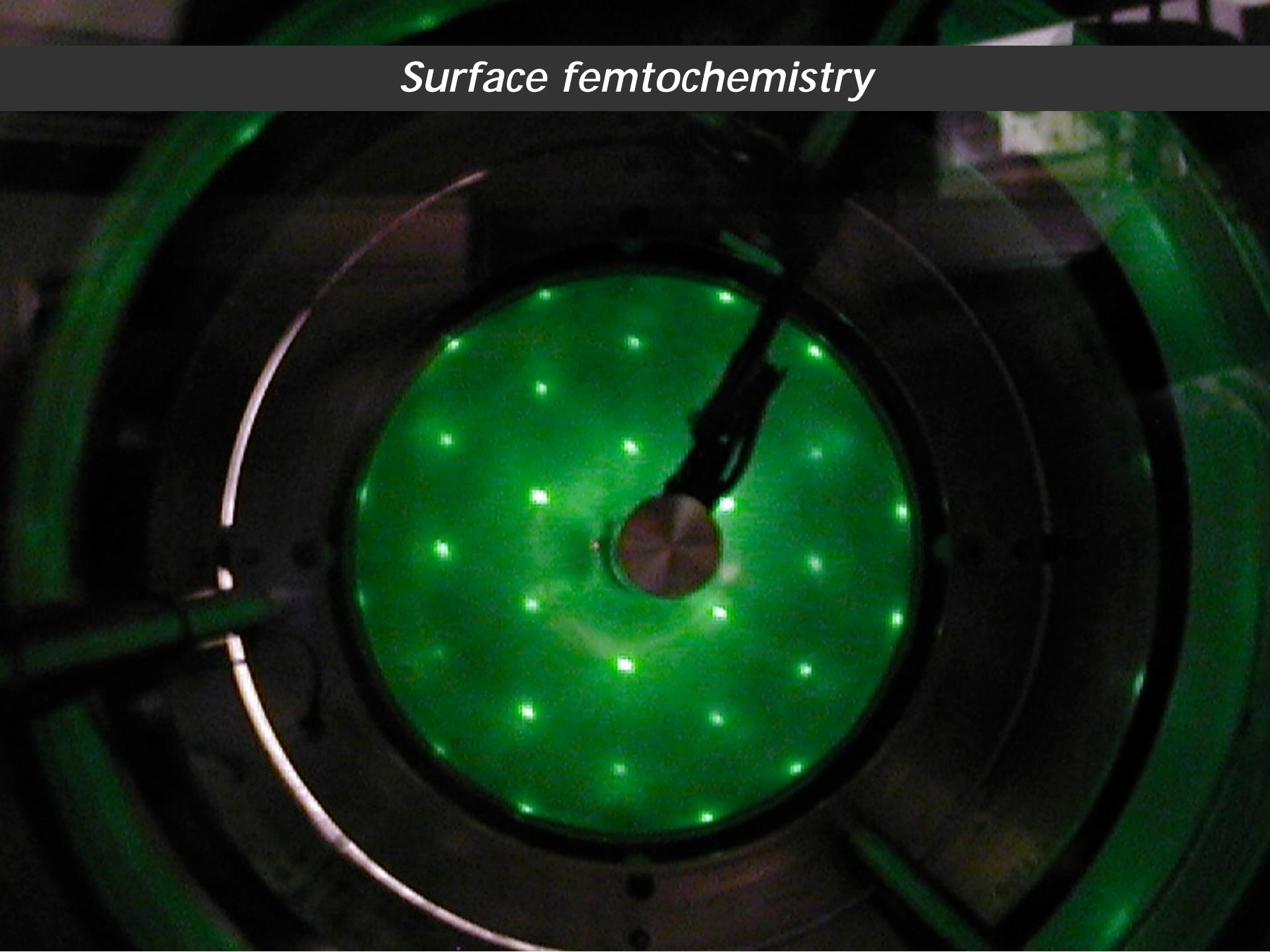


Surface femtochemistry

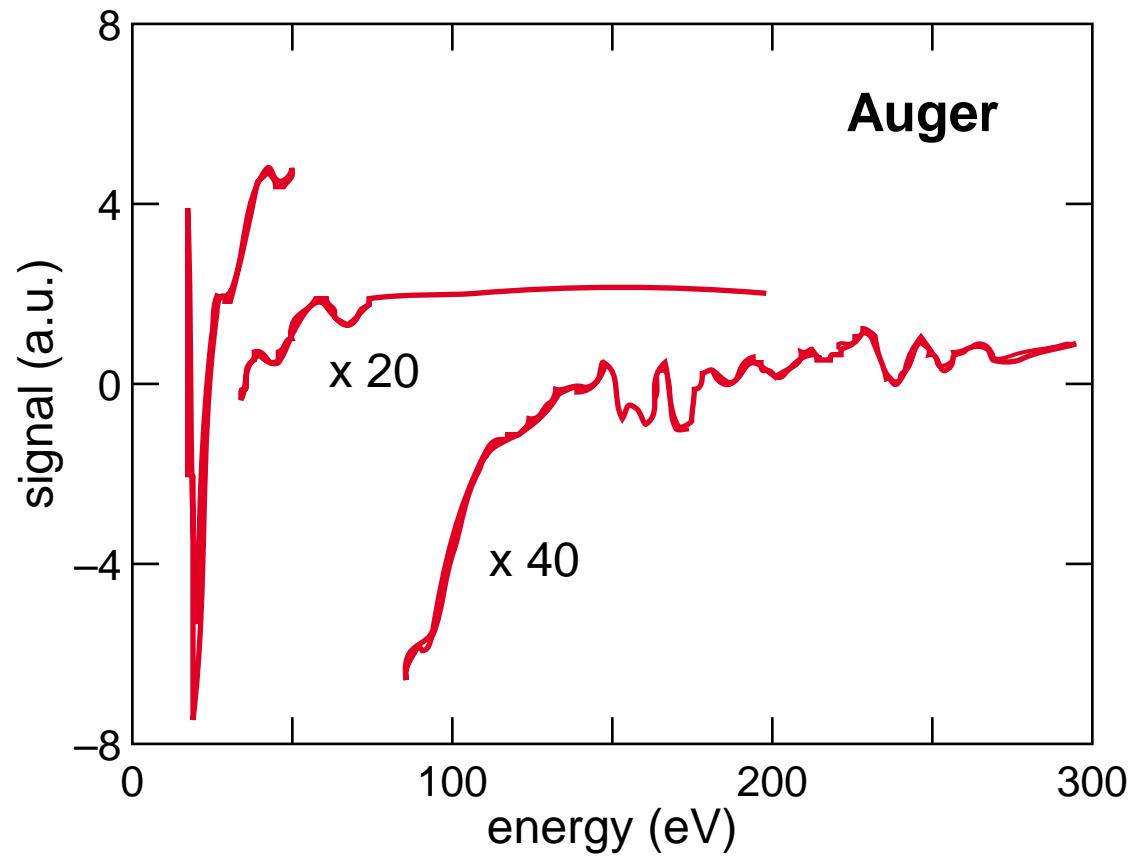
UHV chamber top view



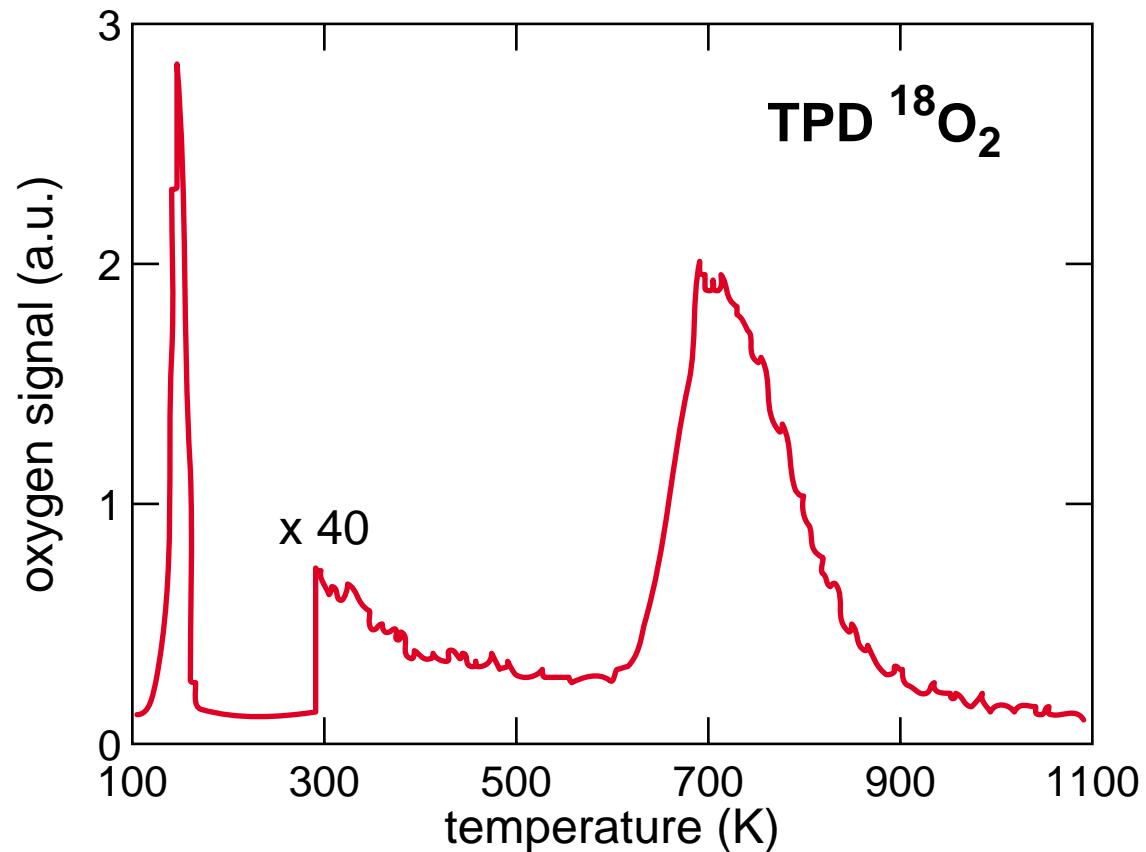
Surface femtochemistry



Surface femtochemistry

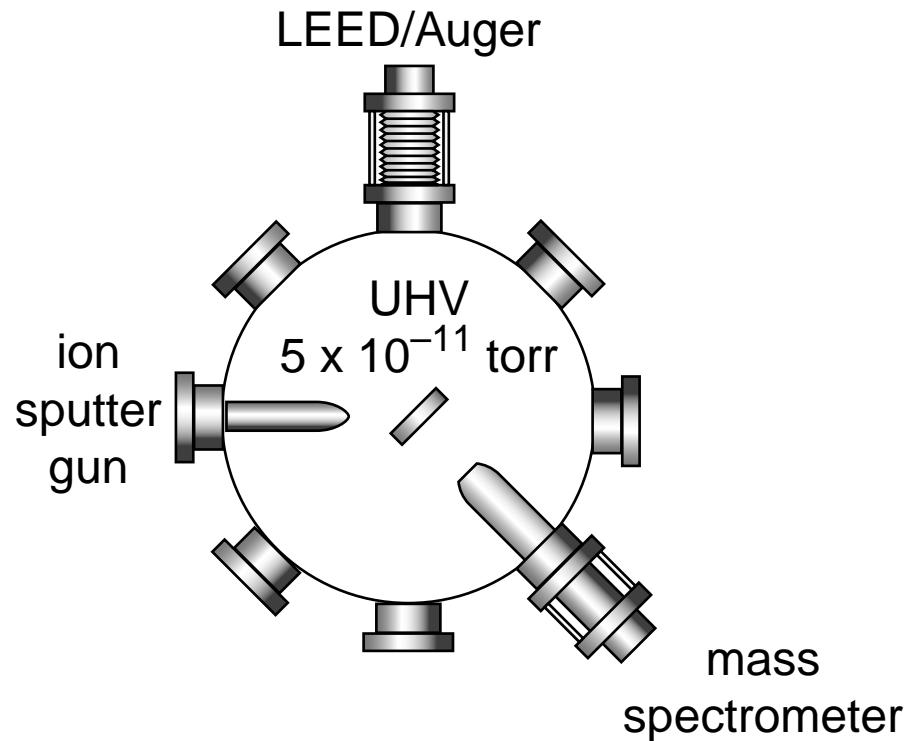


Surface femtochemistry



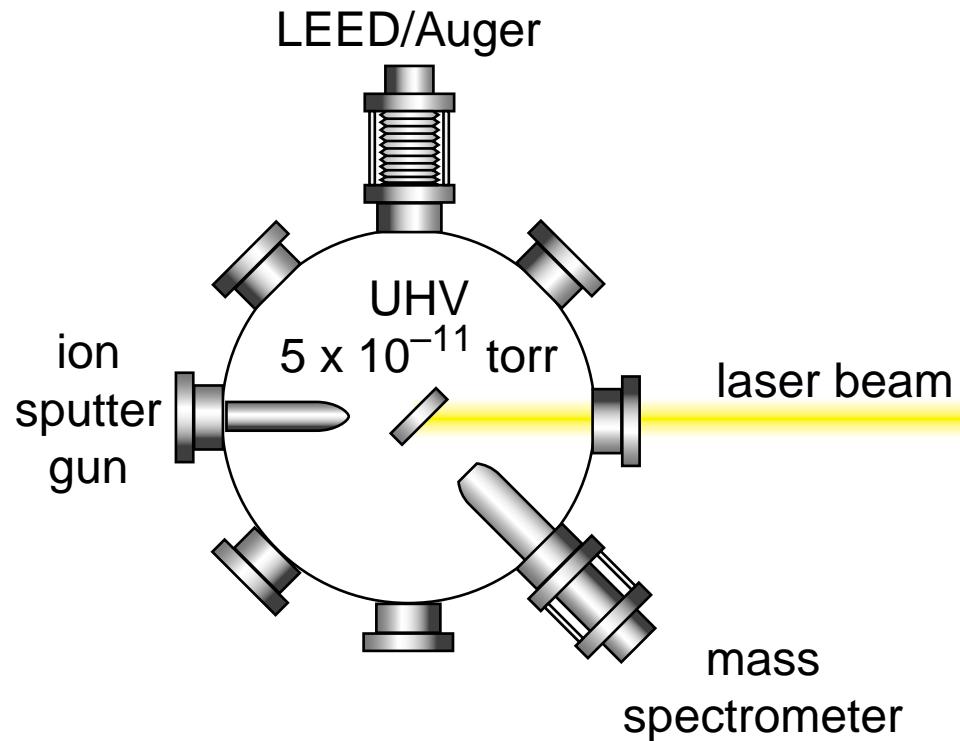
Surface femtochemistry

UHV chamber top view



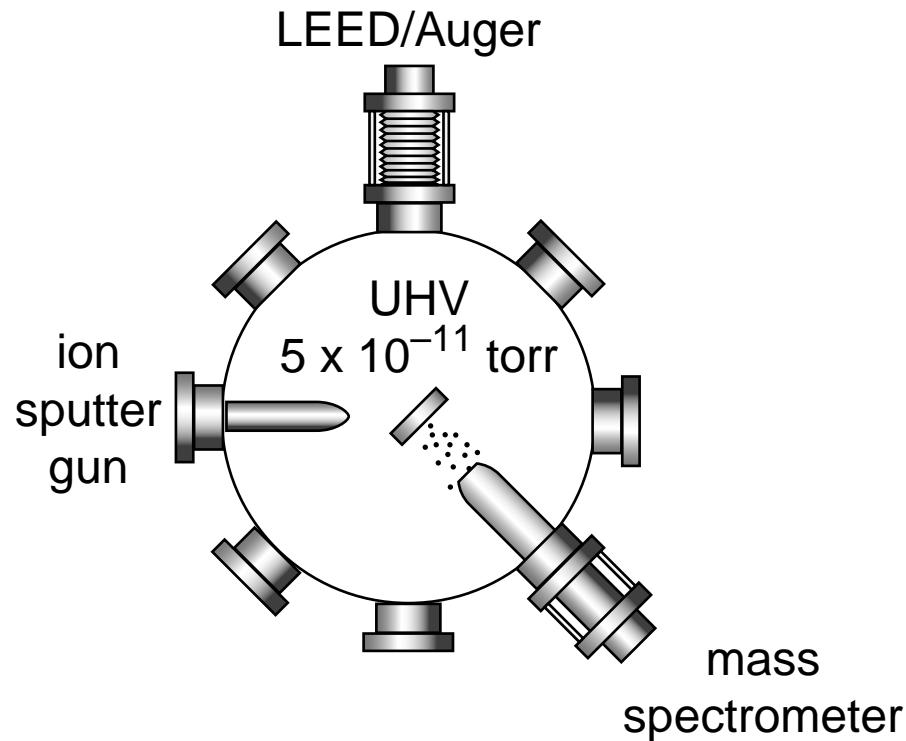
Surface femtochemistry

UHV chamber top view



Surface femtochemistry

UHV chamber top view

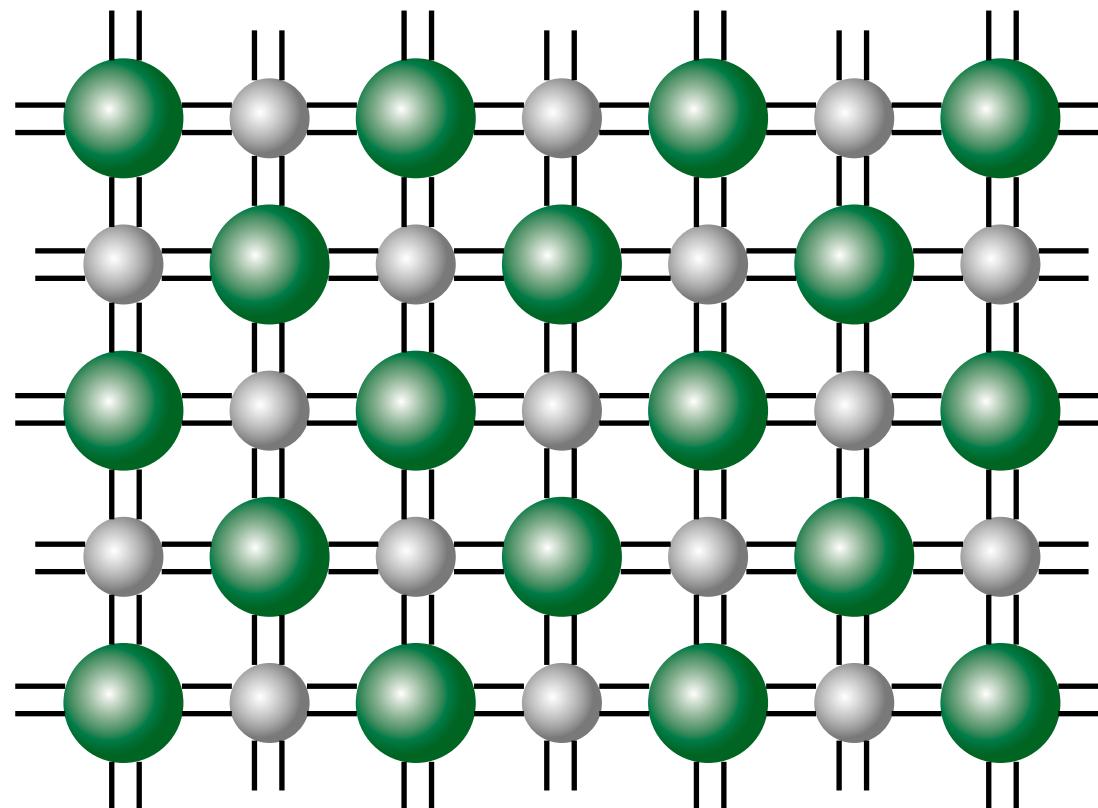


Overview

- ▶ surface femtochemistry
- ▶ electronic and structural transitions
- ▶ microstructuring of materials
- ▶ nonlinear optics and propagation
- ▶ laser surgery

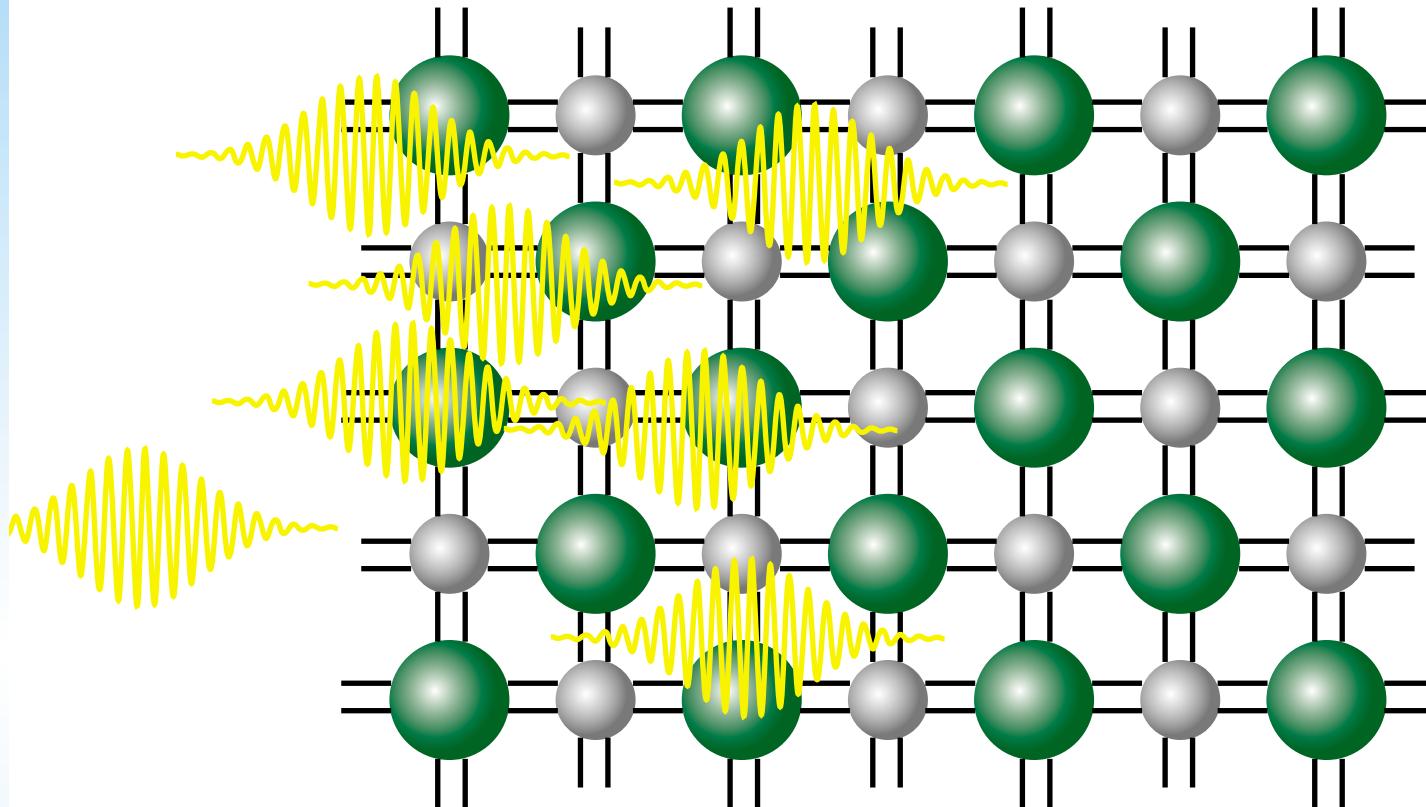
Electronic and structural transitions

how do femtosecond laser pulses alter a solid?



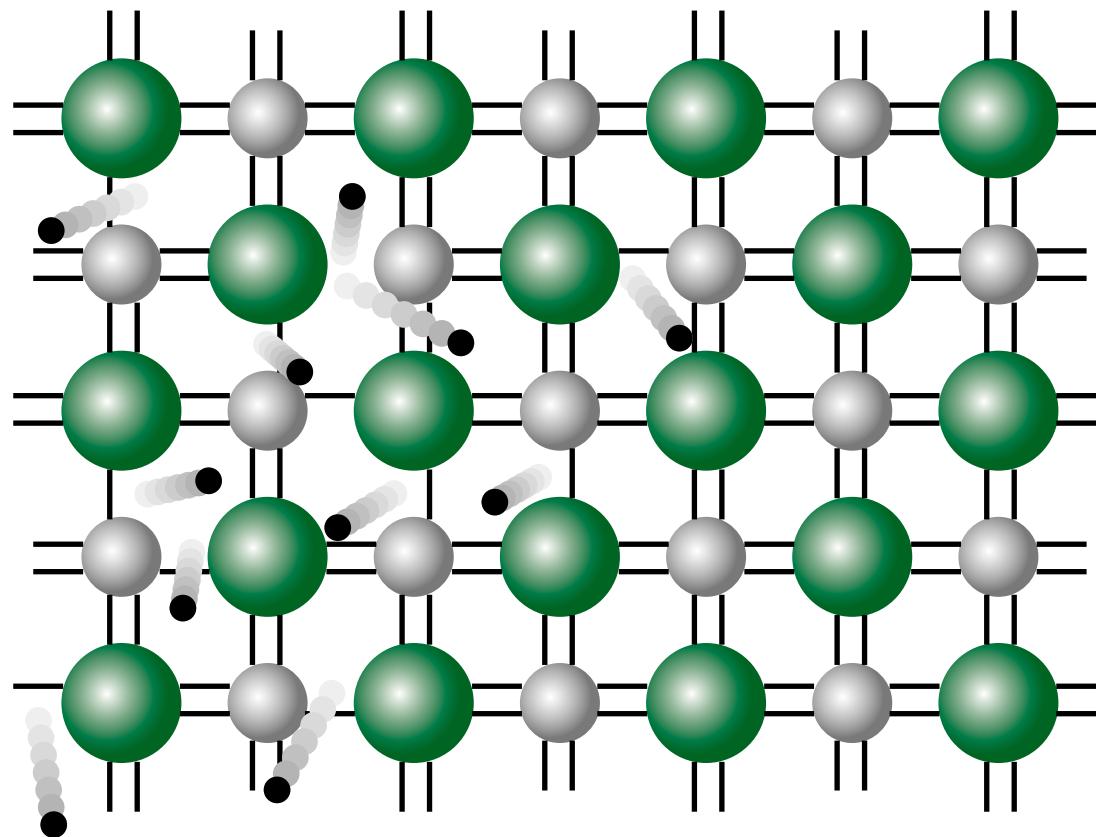
Electronic and structural transitions

photons excite valence electrons...



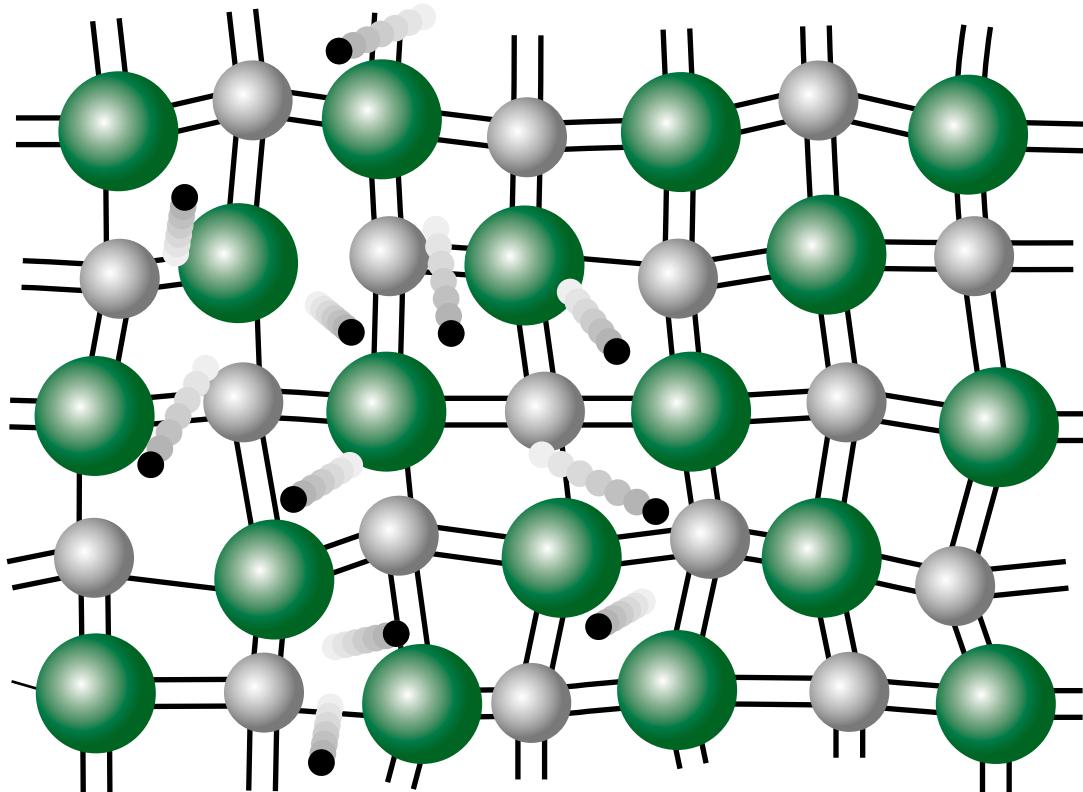
Electronic and structural transitions

...and create free electrons...



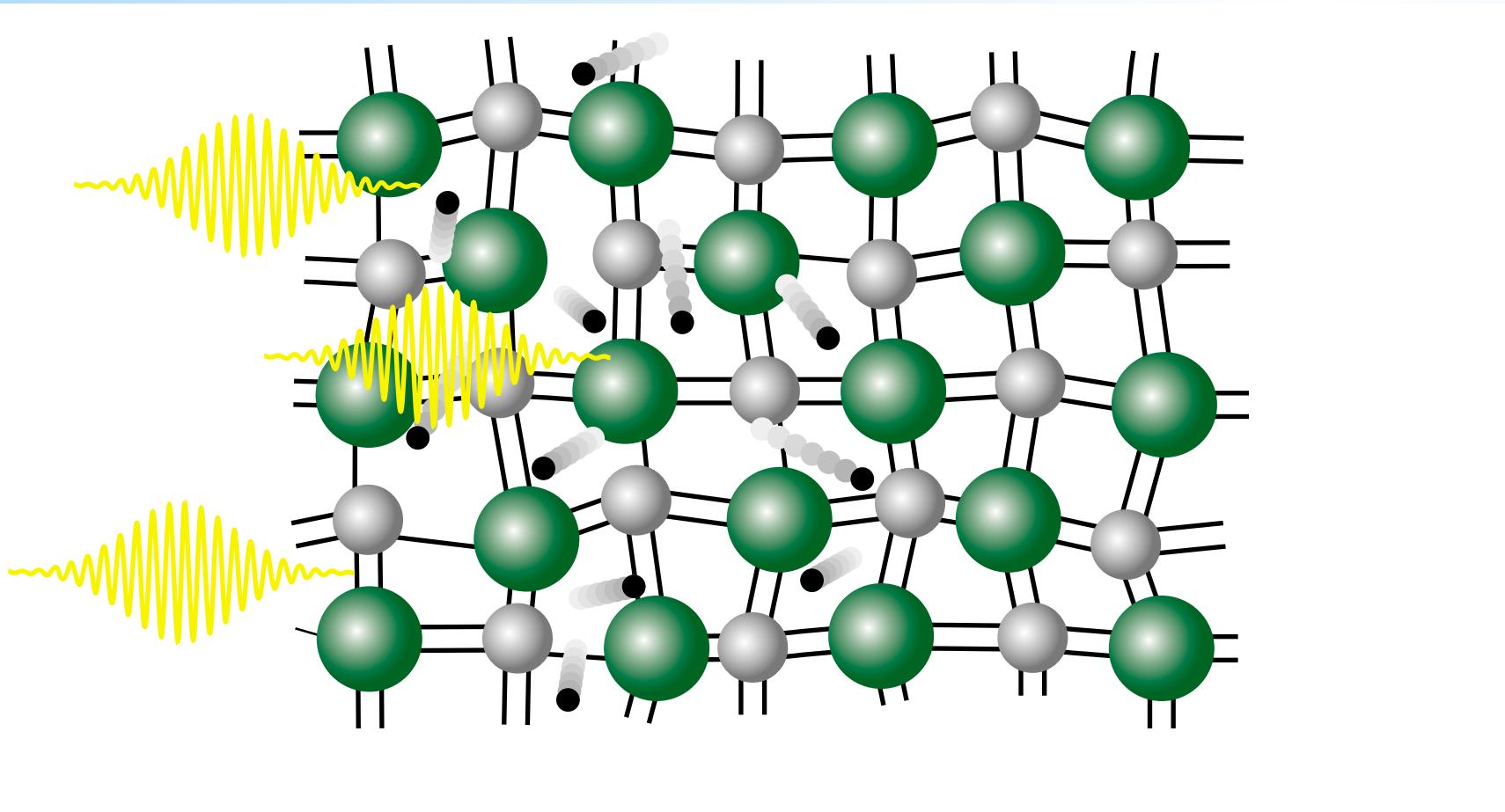
Electronic and structural transitions

...causing electronic and structural changes...



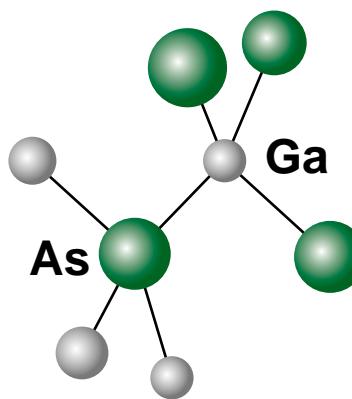
Electronic and structural transitions

...which we measure with another pulse

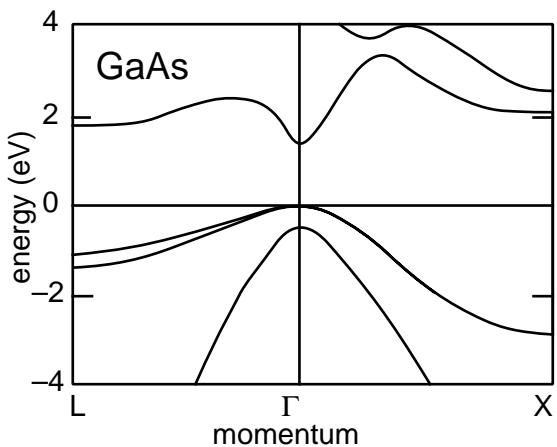


Electronic and structural transitions

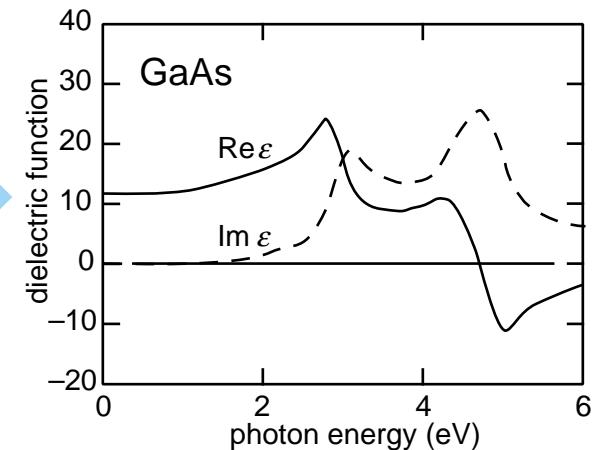
structure



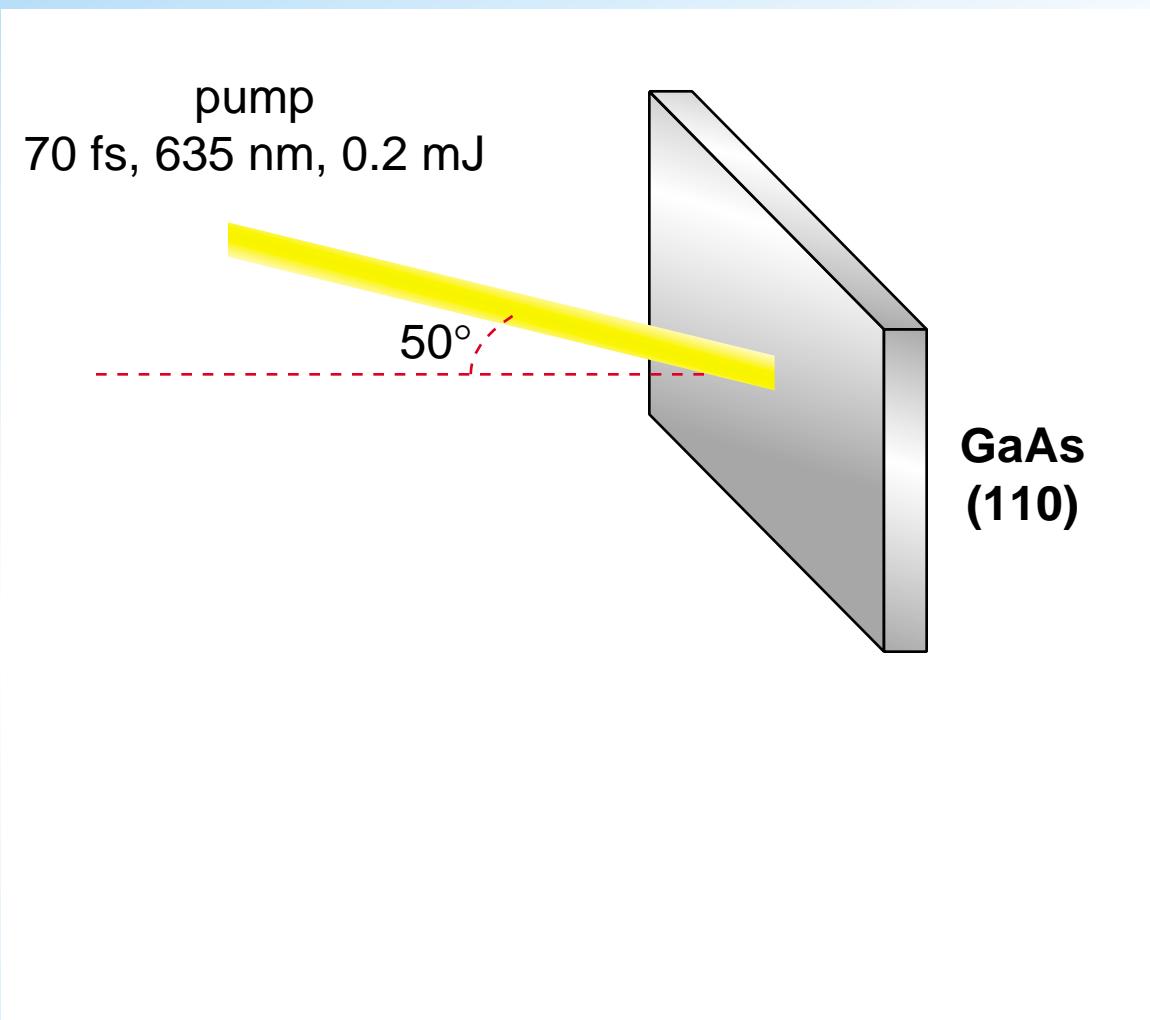
bandstructure



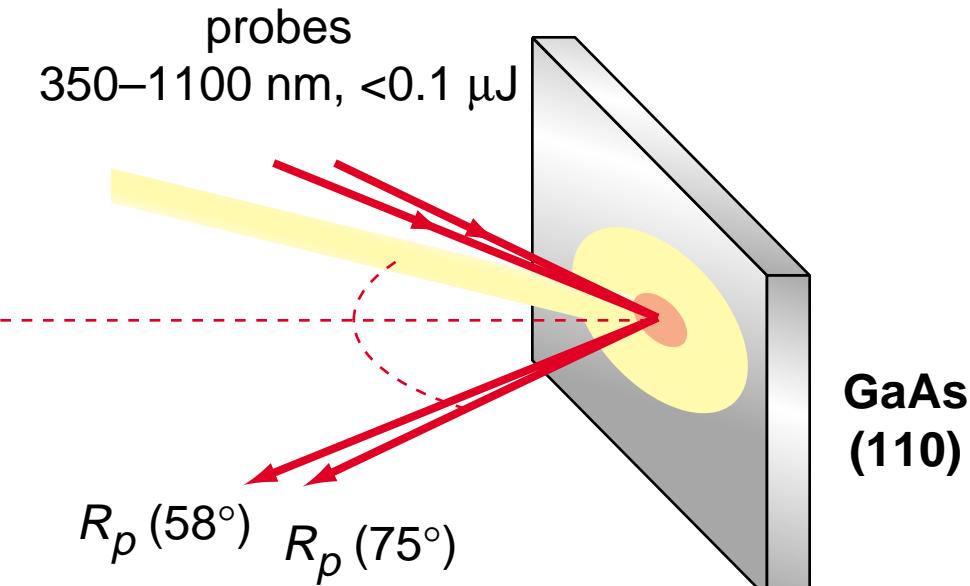
dielectric function



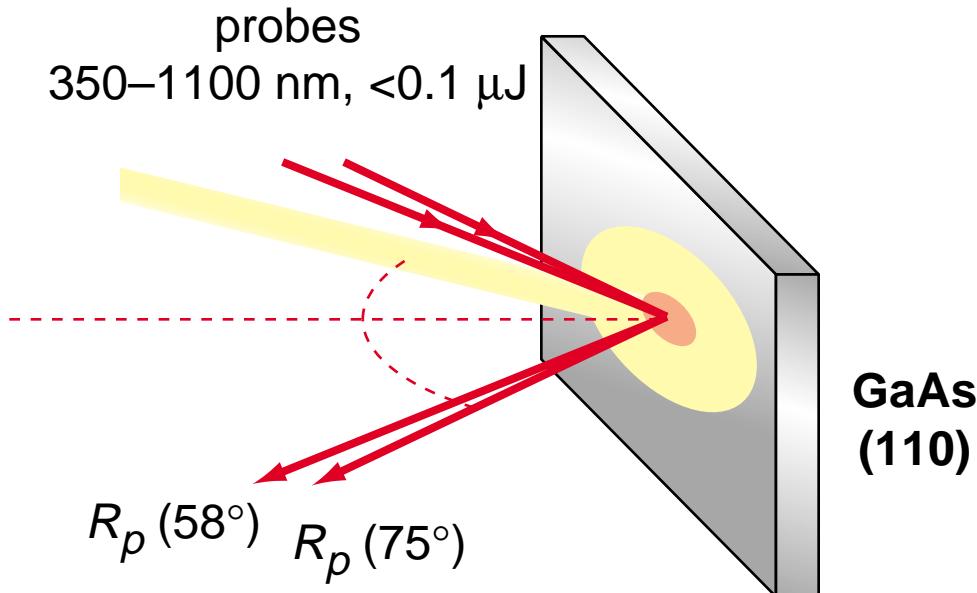
Electronic and structural transitions



Electronic and structural transitions



Electronic and structural transitions

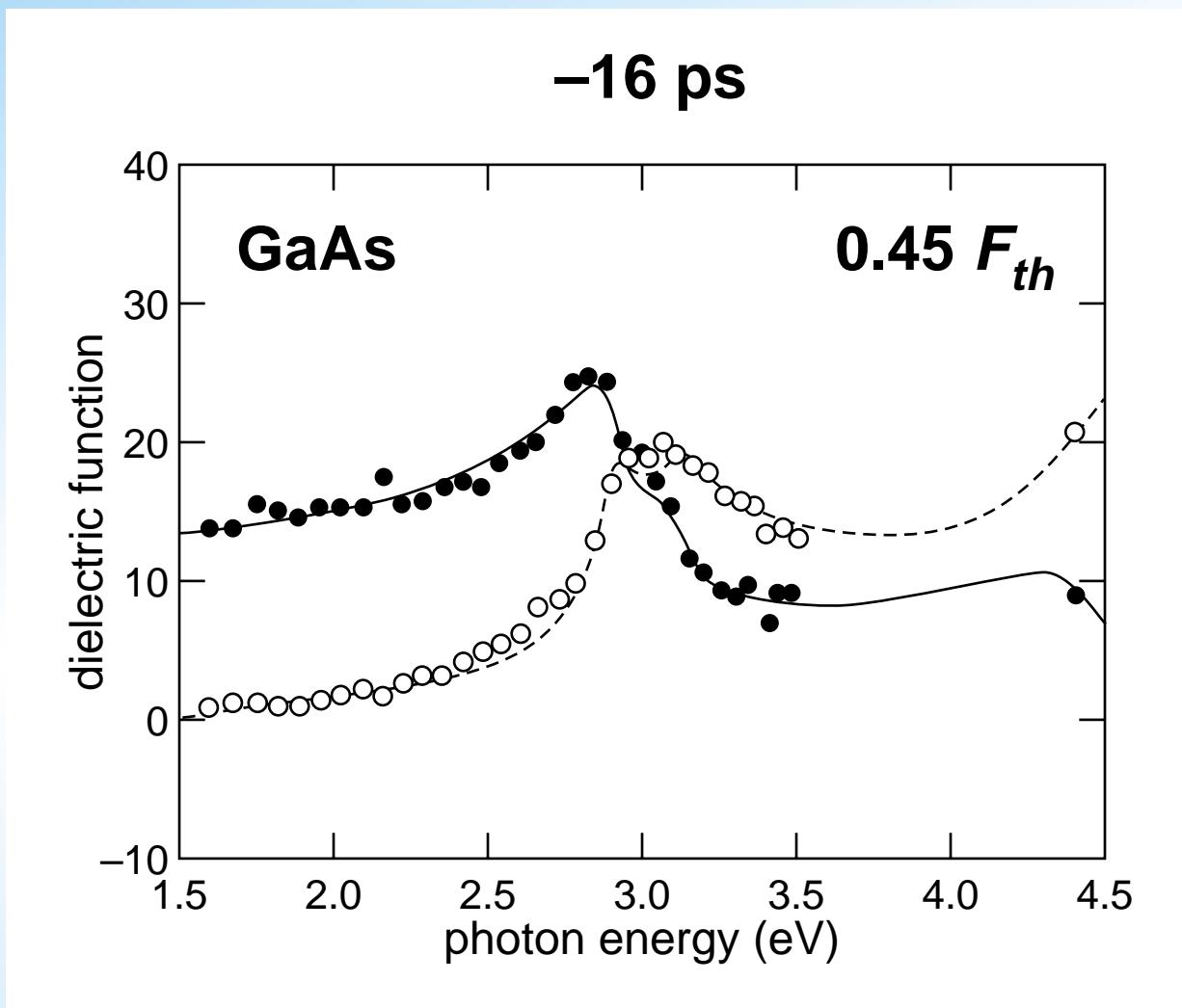


Fresnel
equations

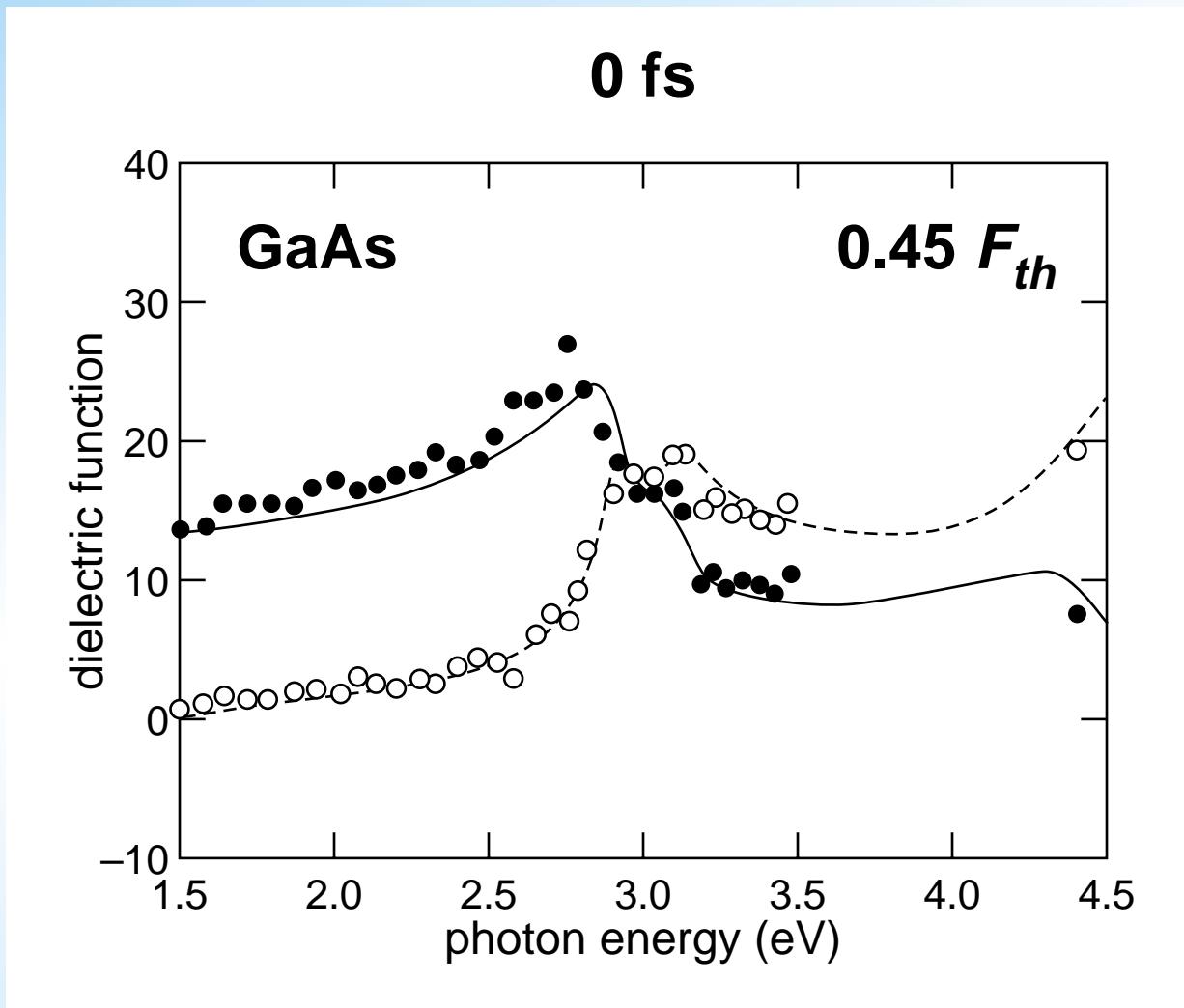


$\text{Re } \varepsilon(\omega)$
 $\text{Im } \varepsilon(\omega)$

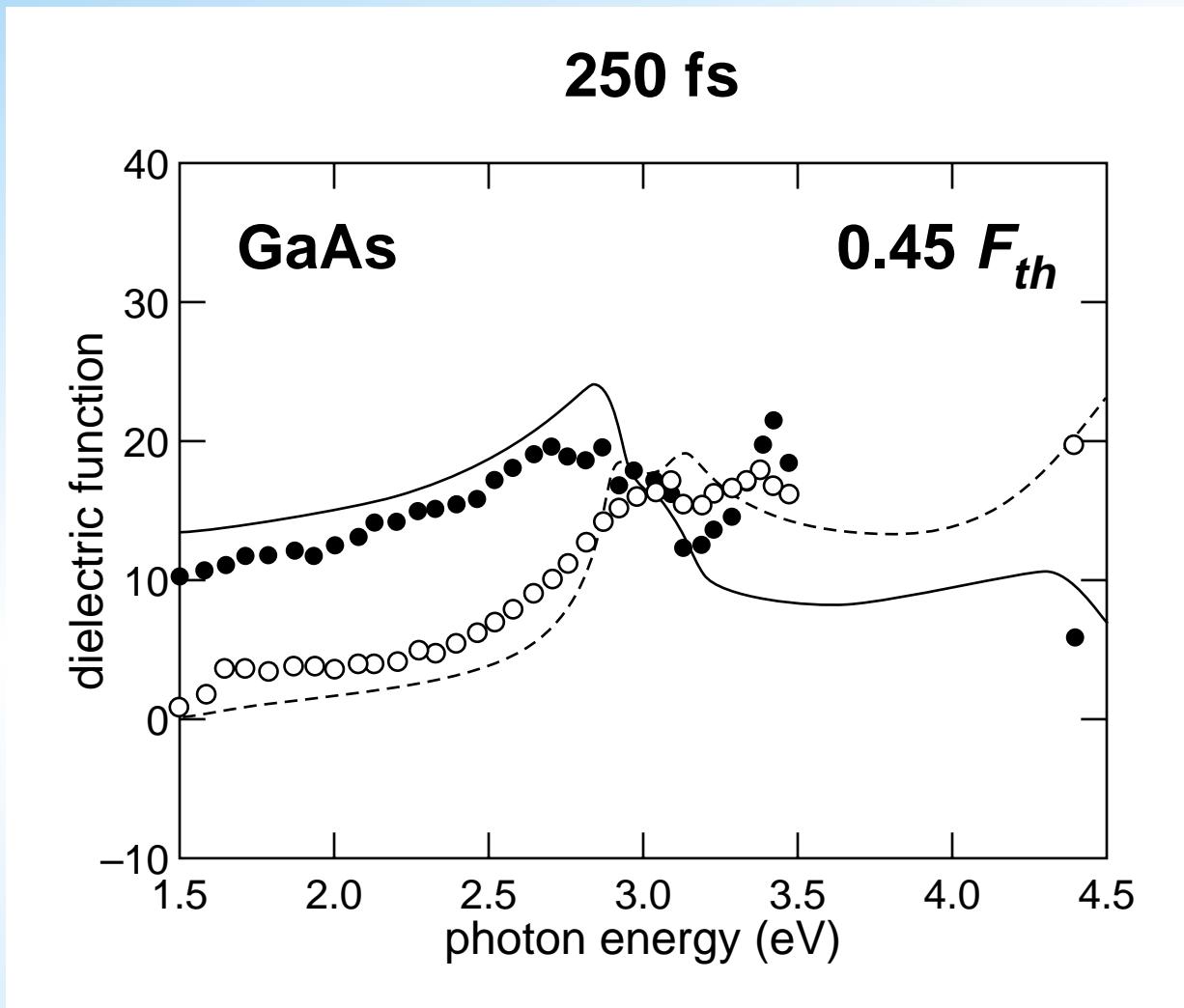
Electronic and structural transitions



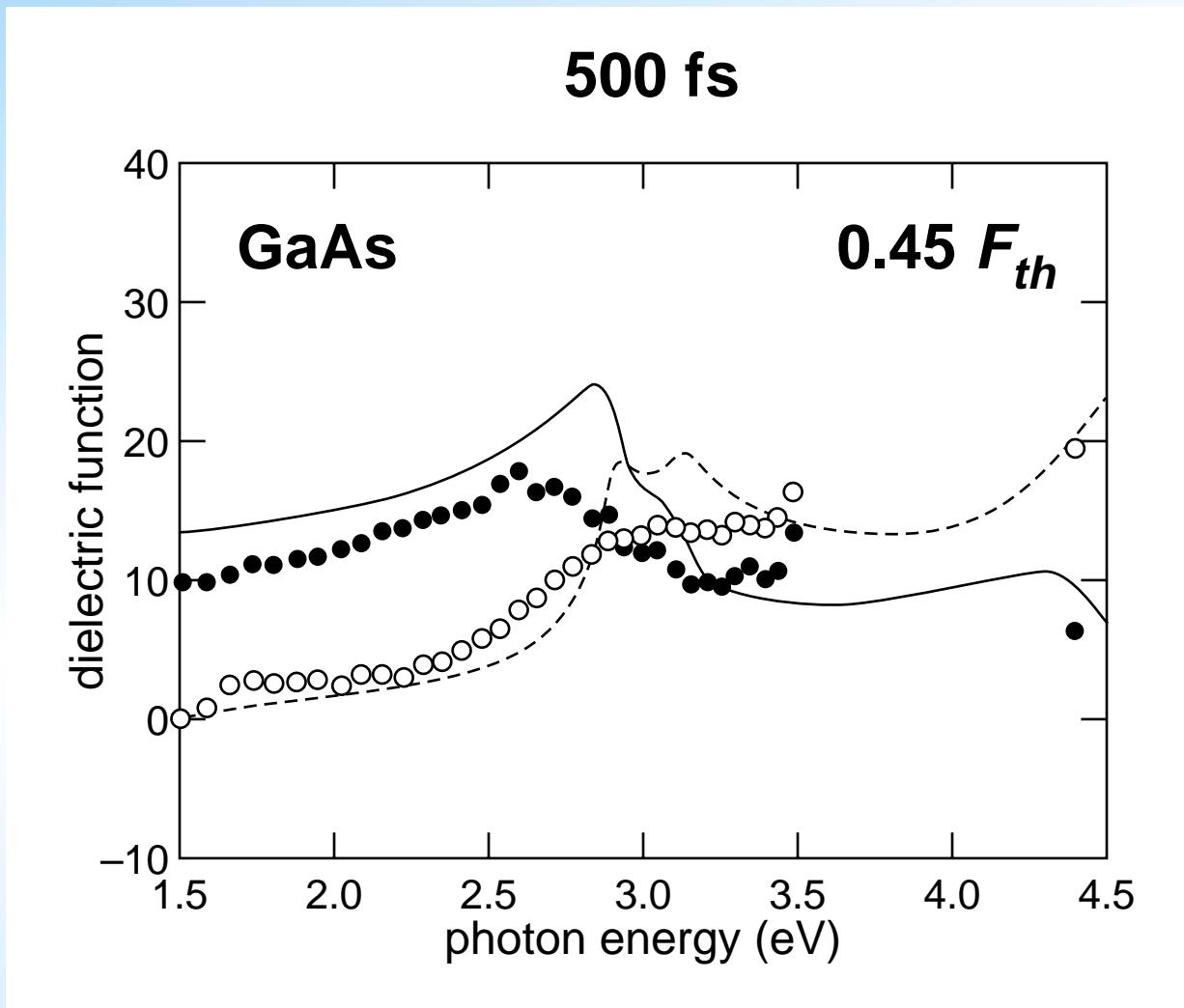
Electronic and structural transitions



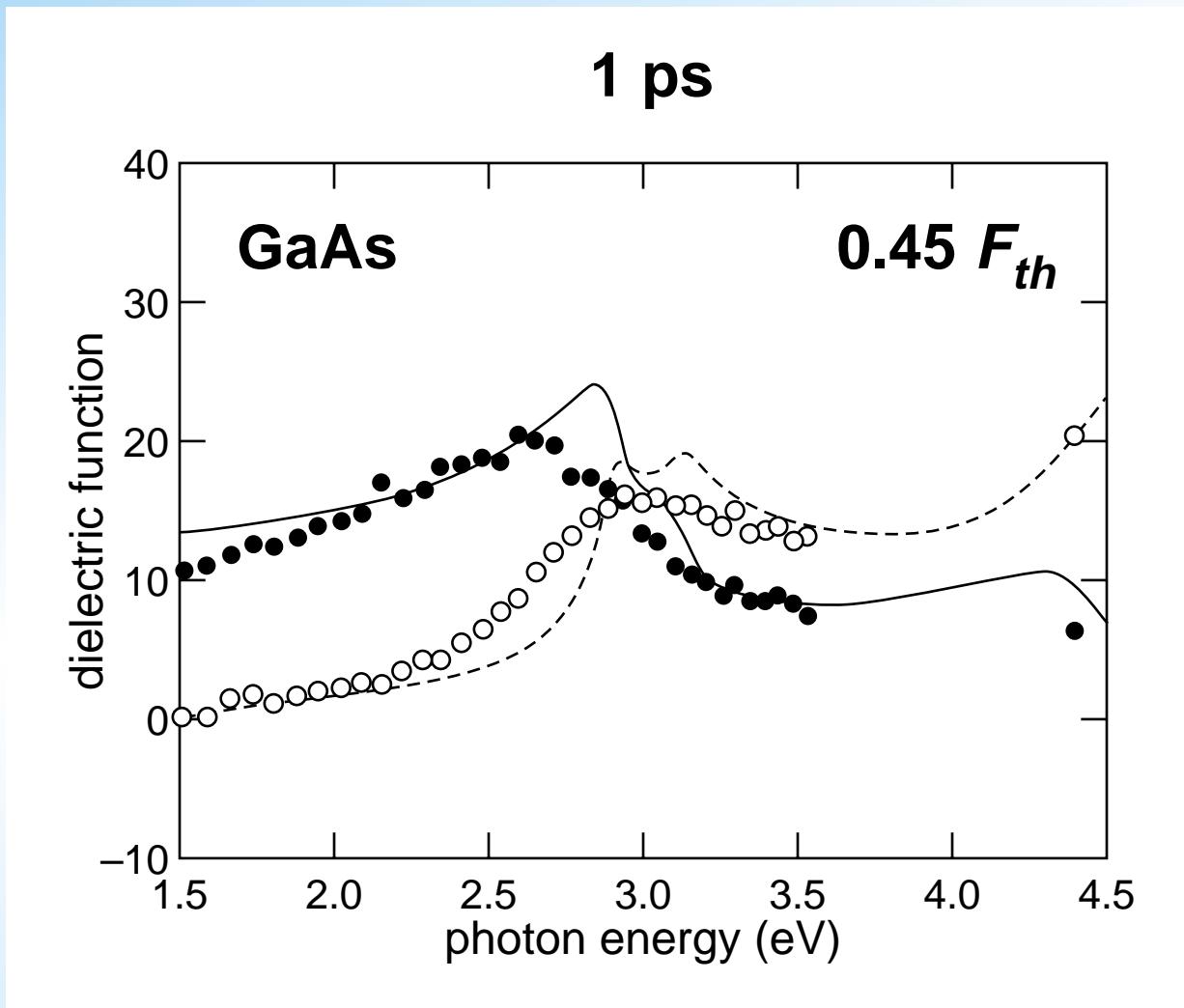
Electronic and structural transitions



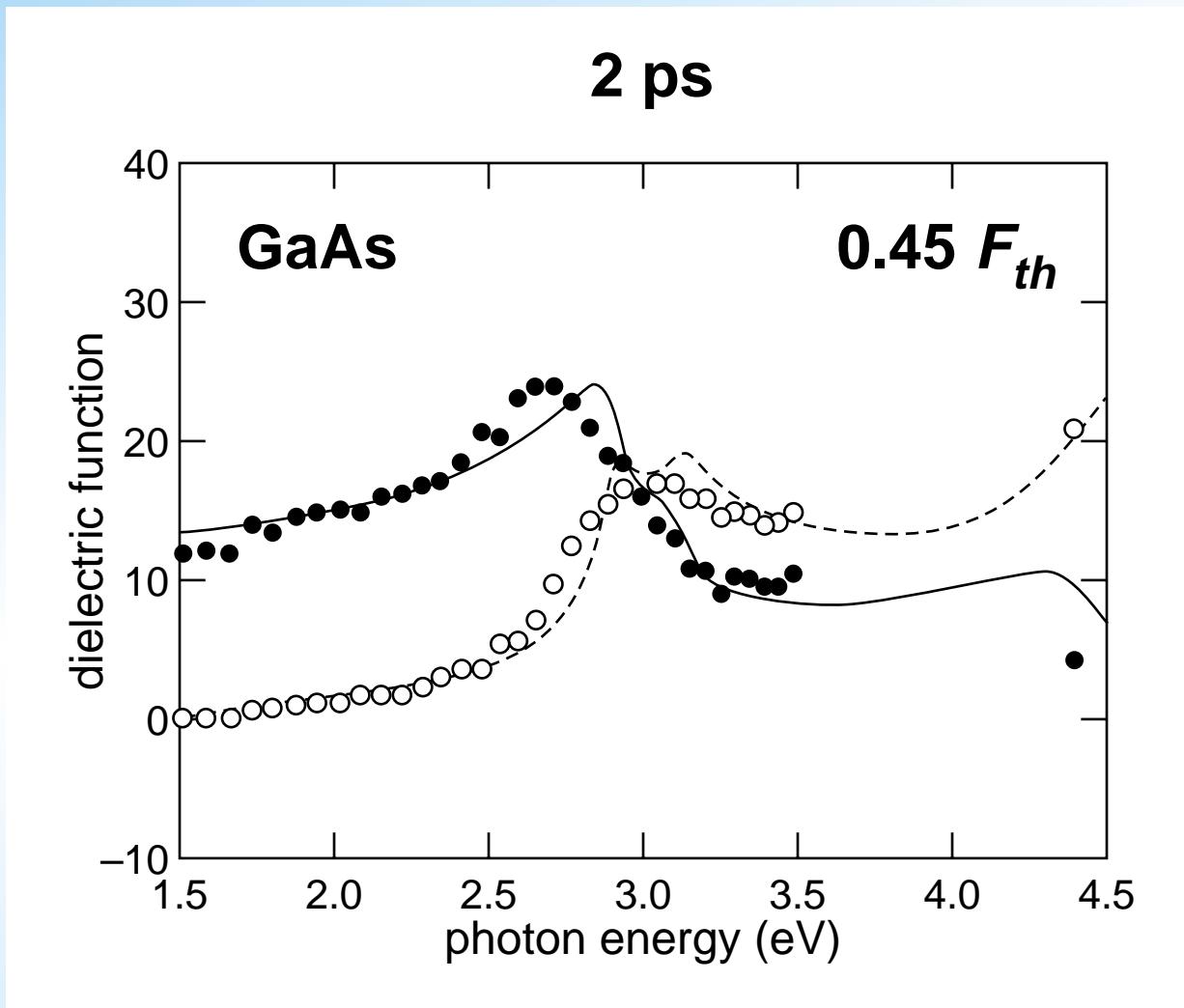
Electronic and structural transitions



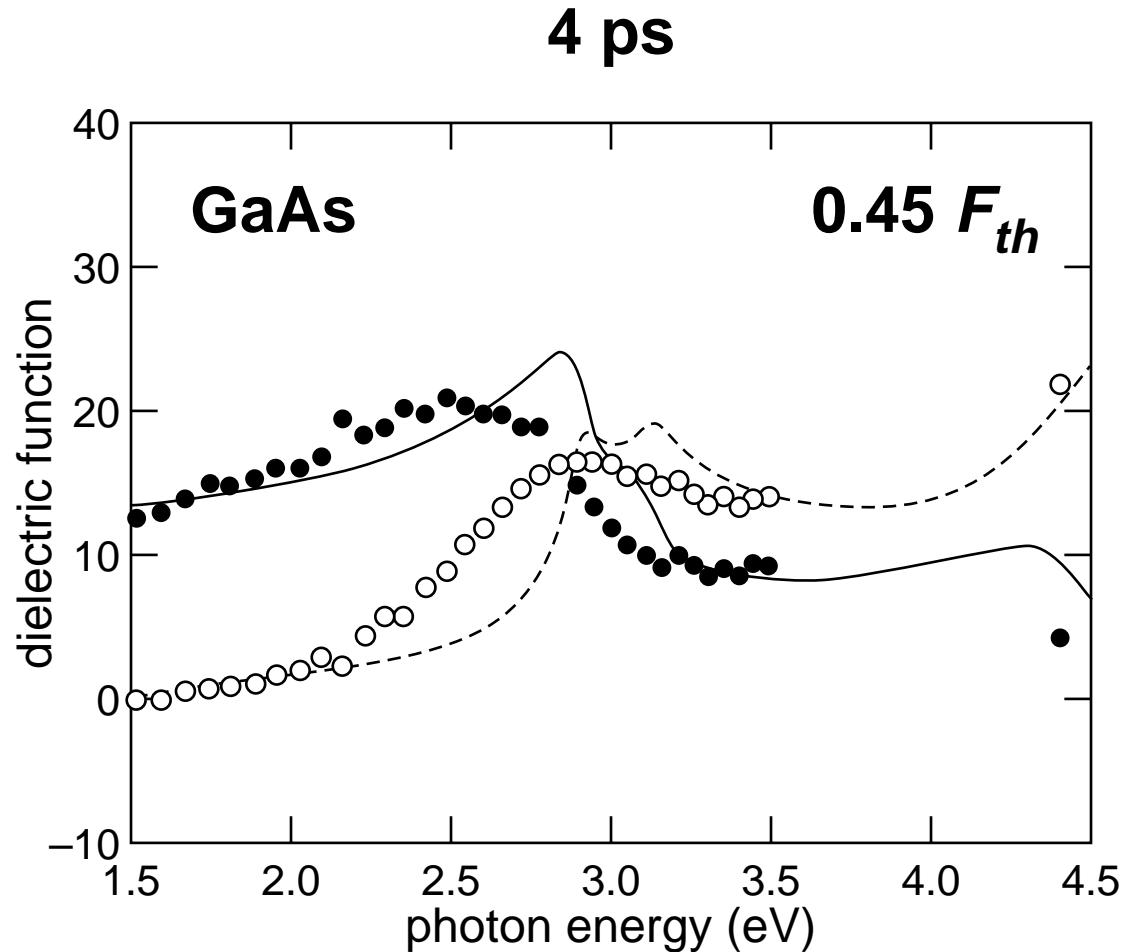
Electronic and structural transitions



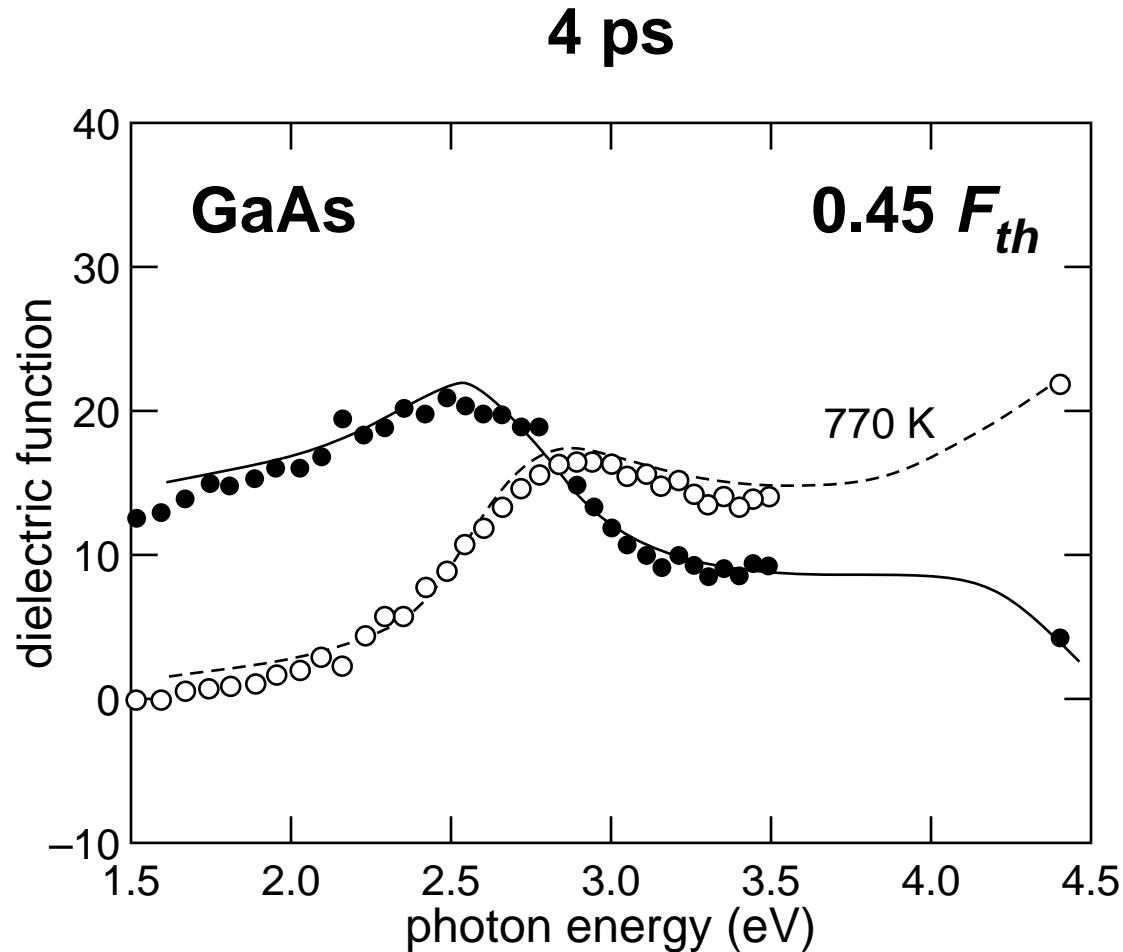
Electronic and structural transitions



Electronic and structural transitions

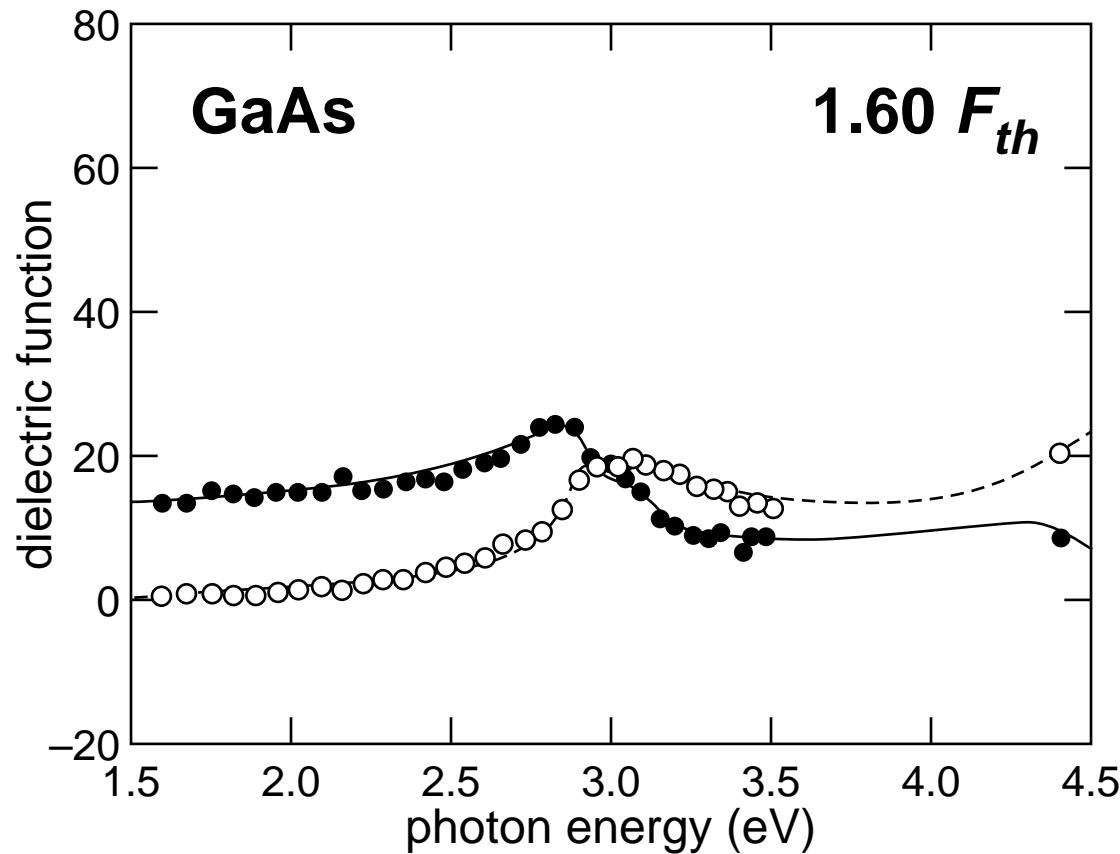


Electronic and structural transitions

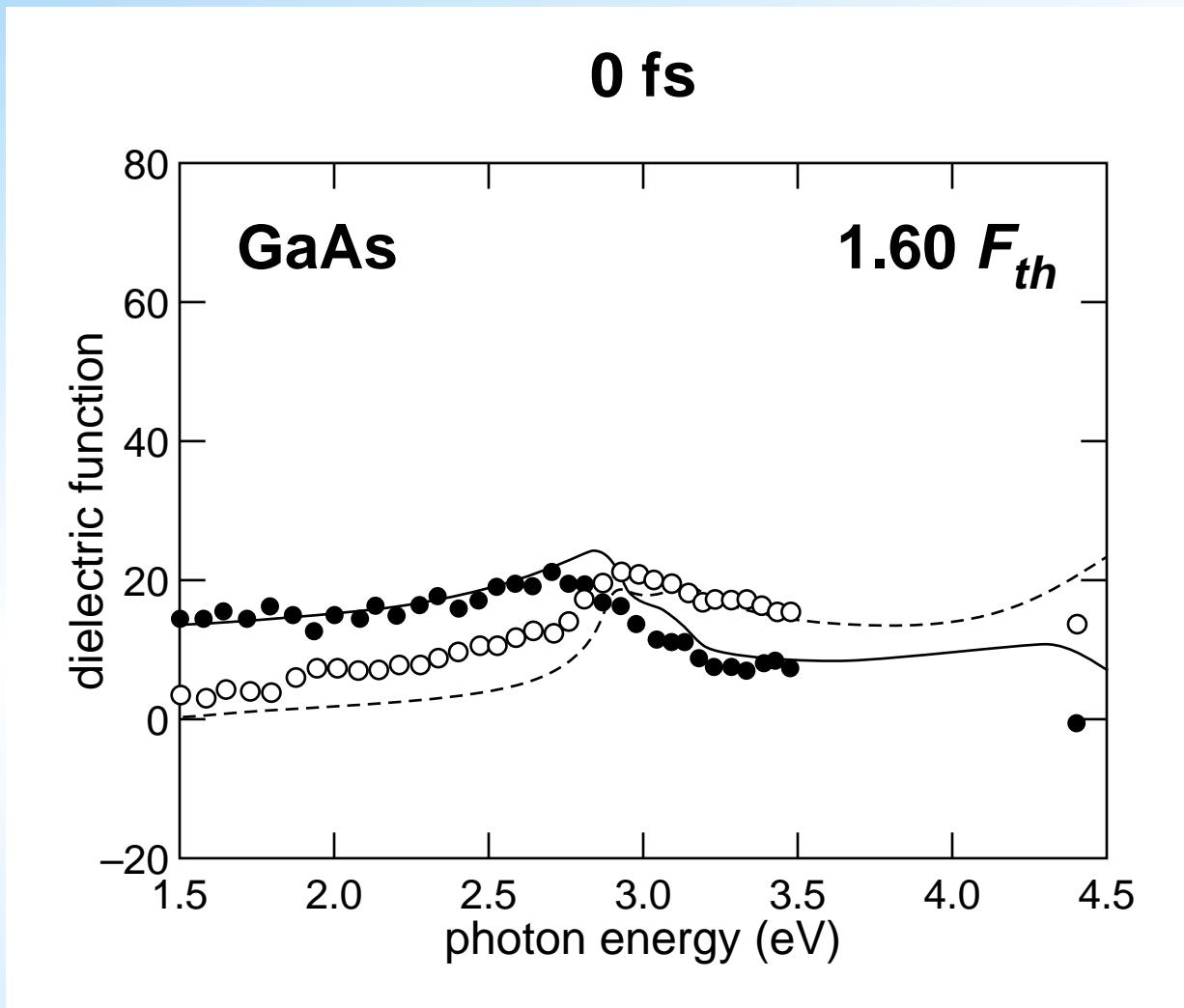


Electronic and structural transitions

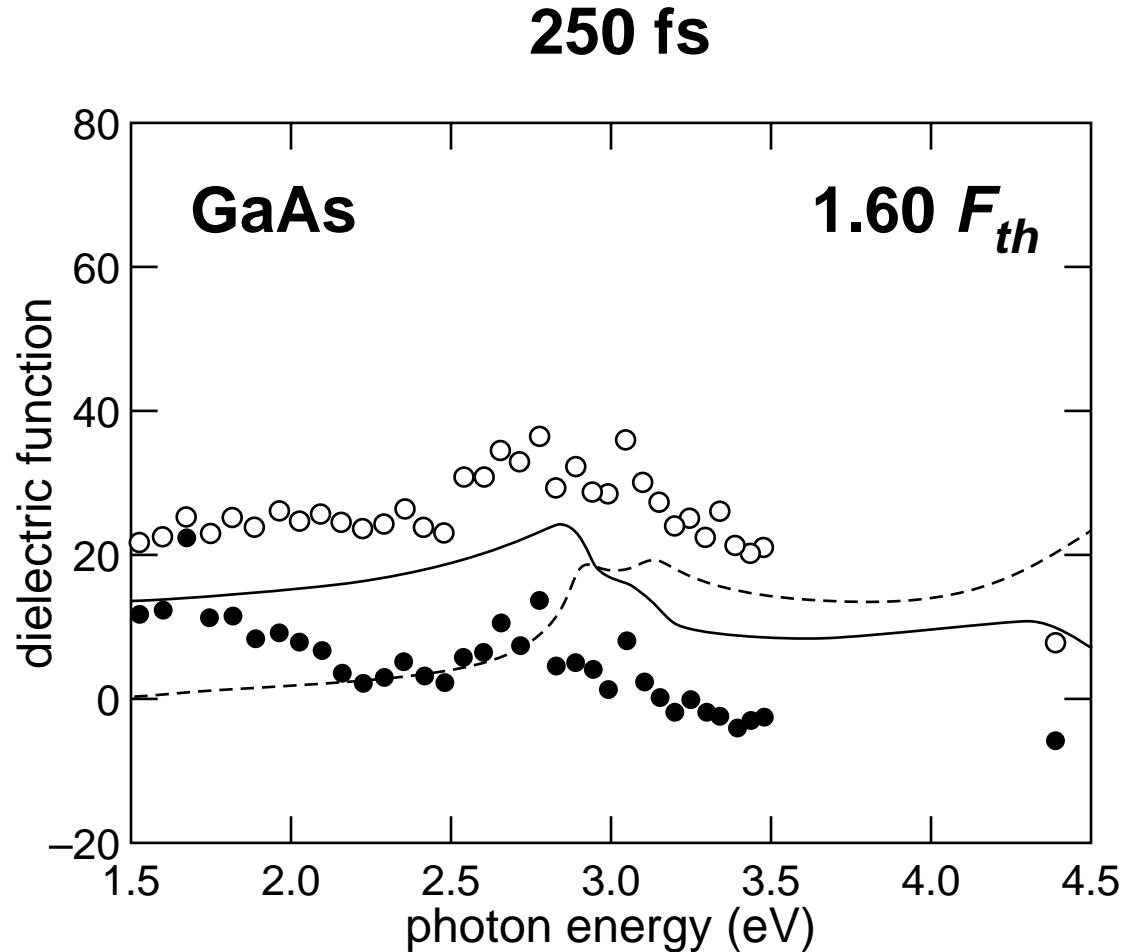
-16 ps



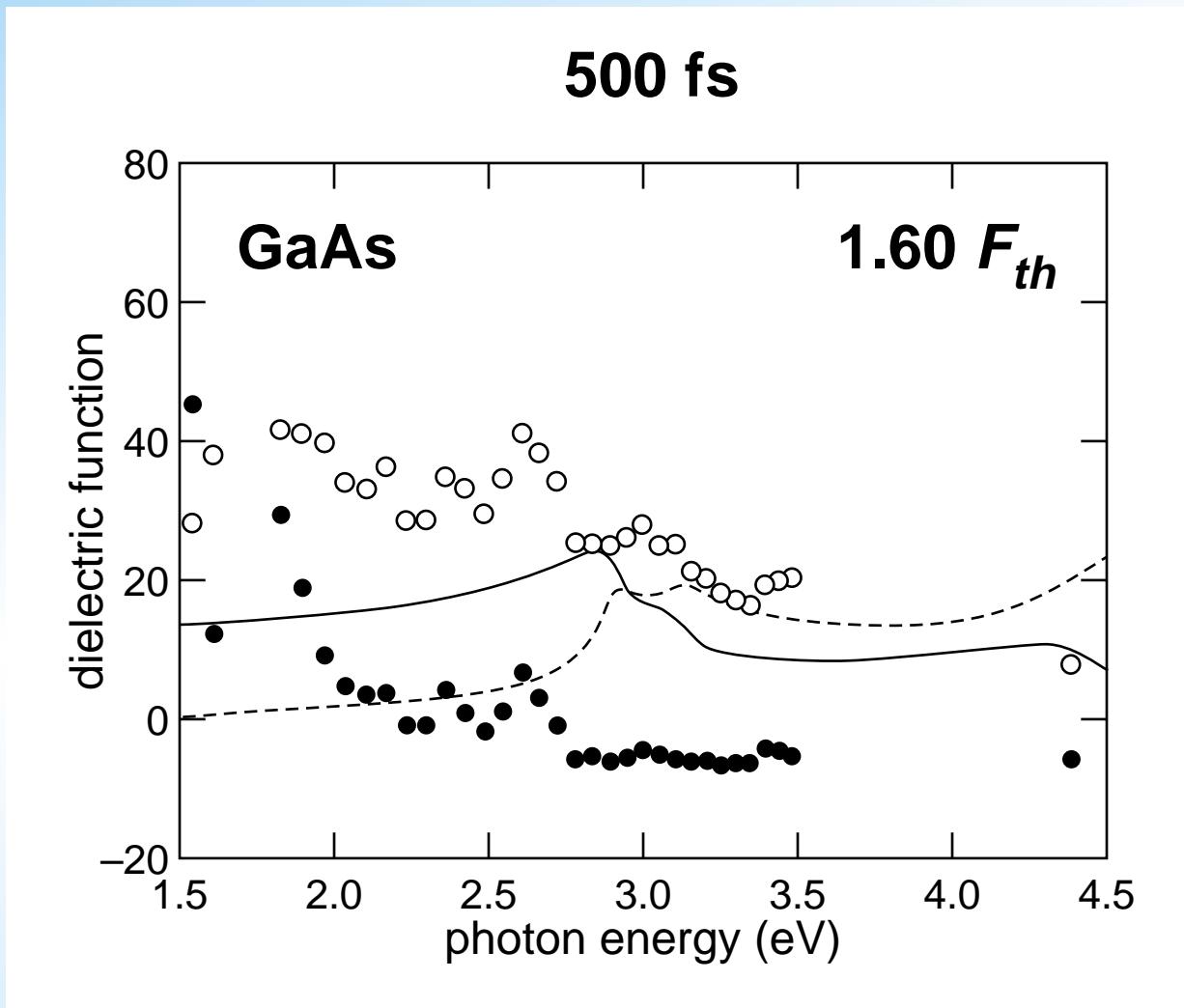
Electronic and structural transitions



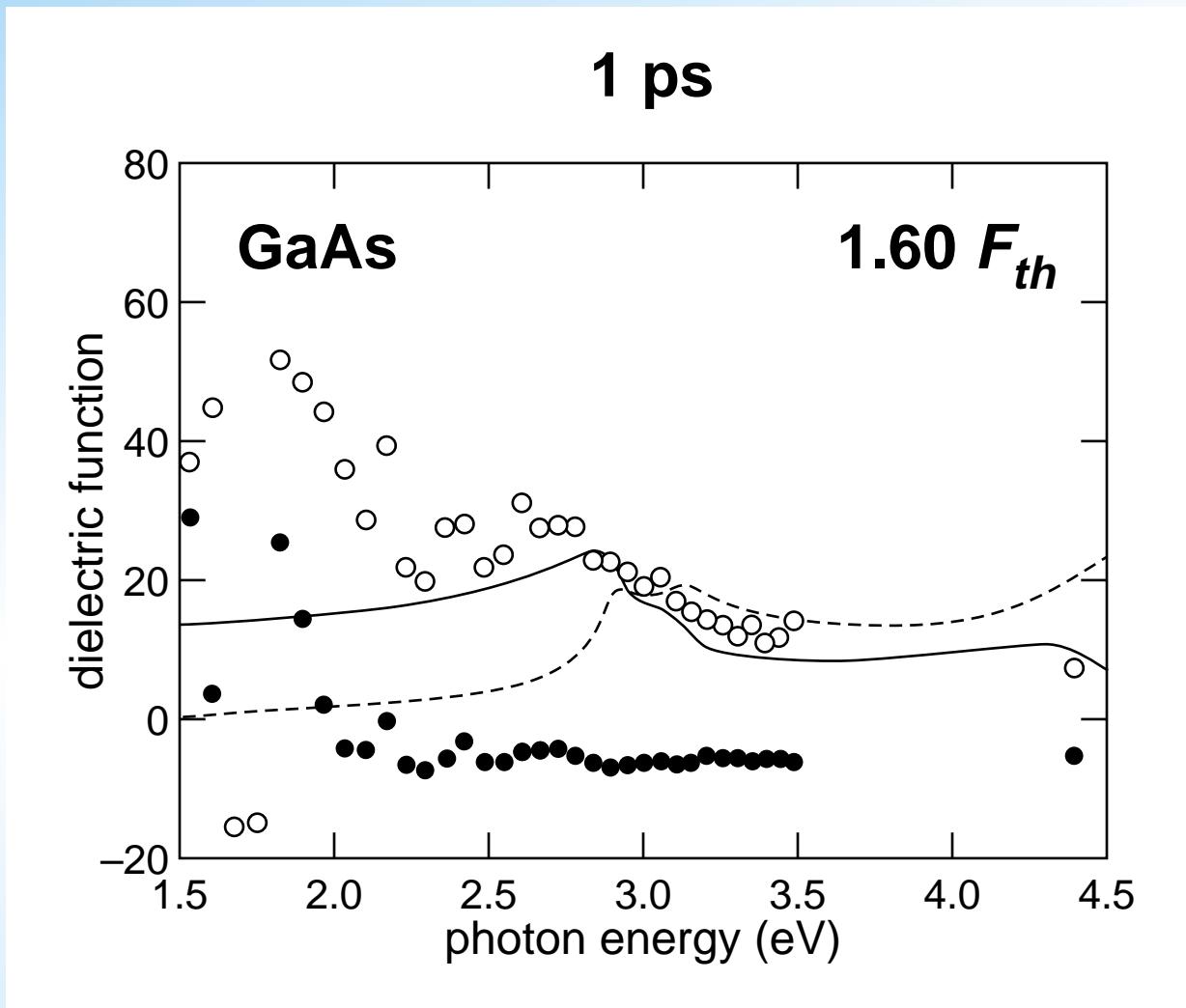
Electronic and structural transitions



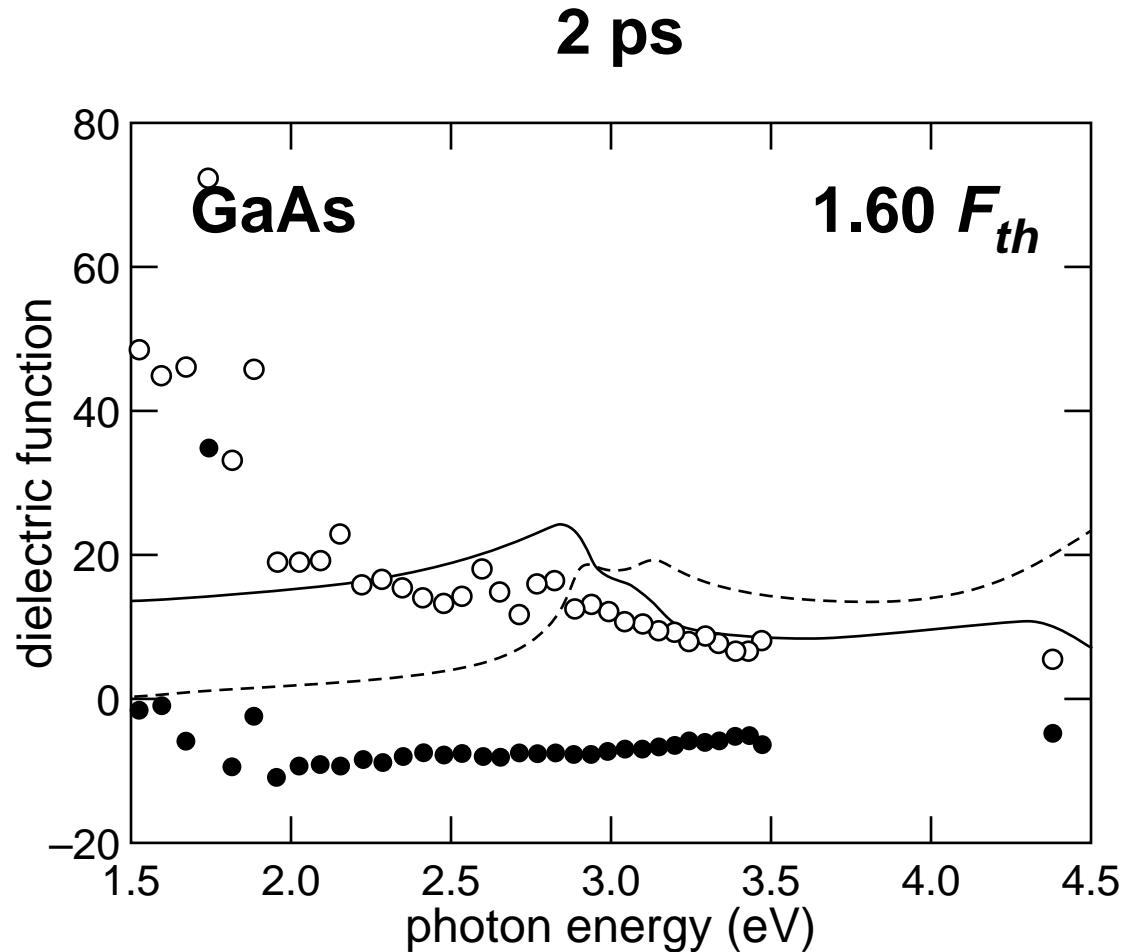
Electronic and structural transitions



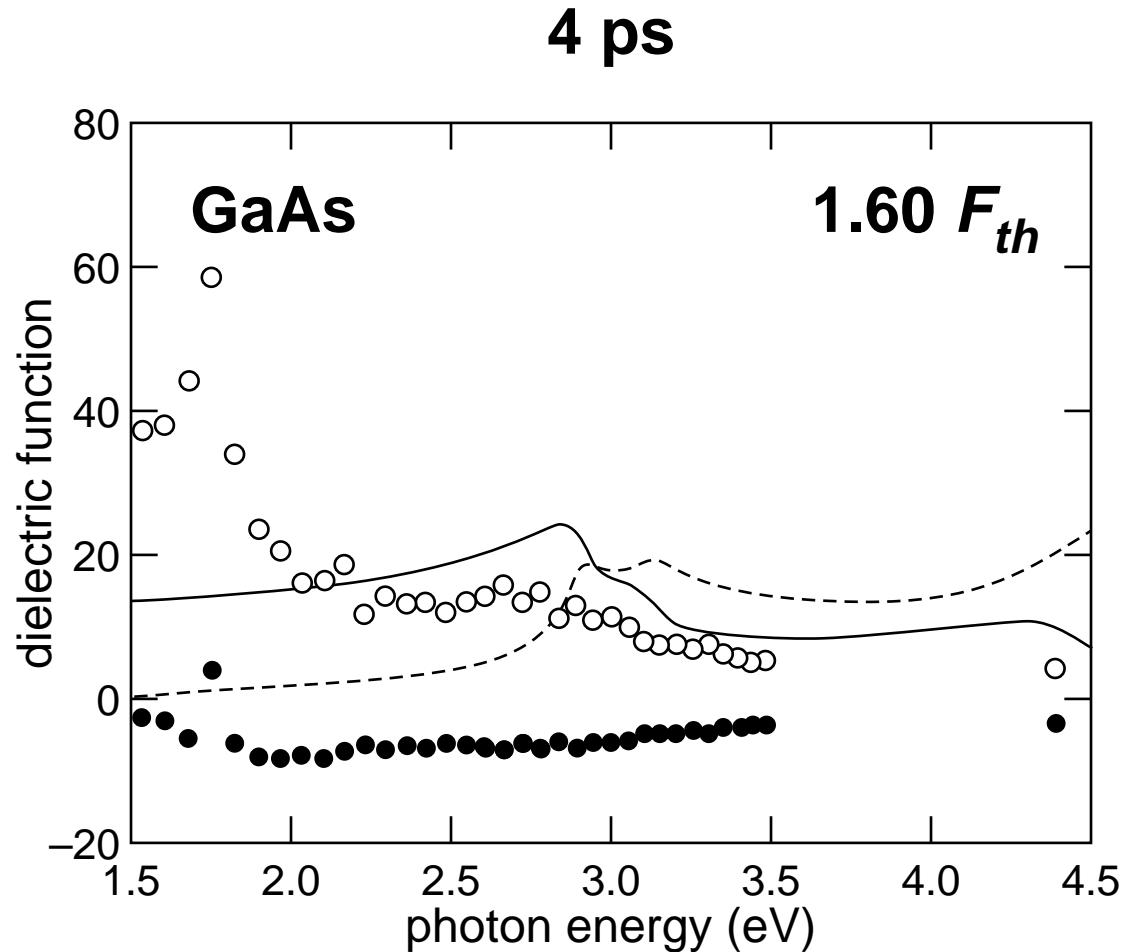
Electronic and structural transitions



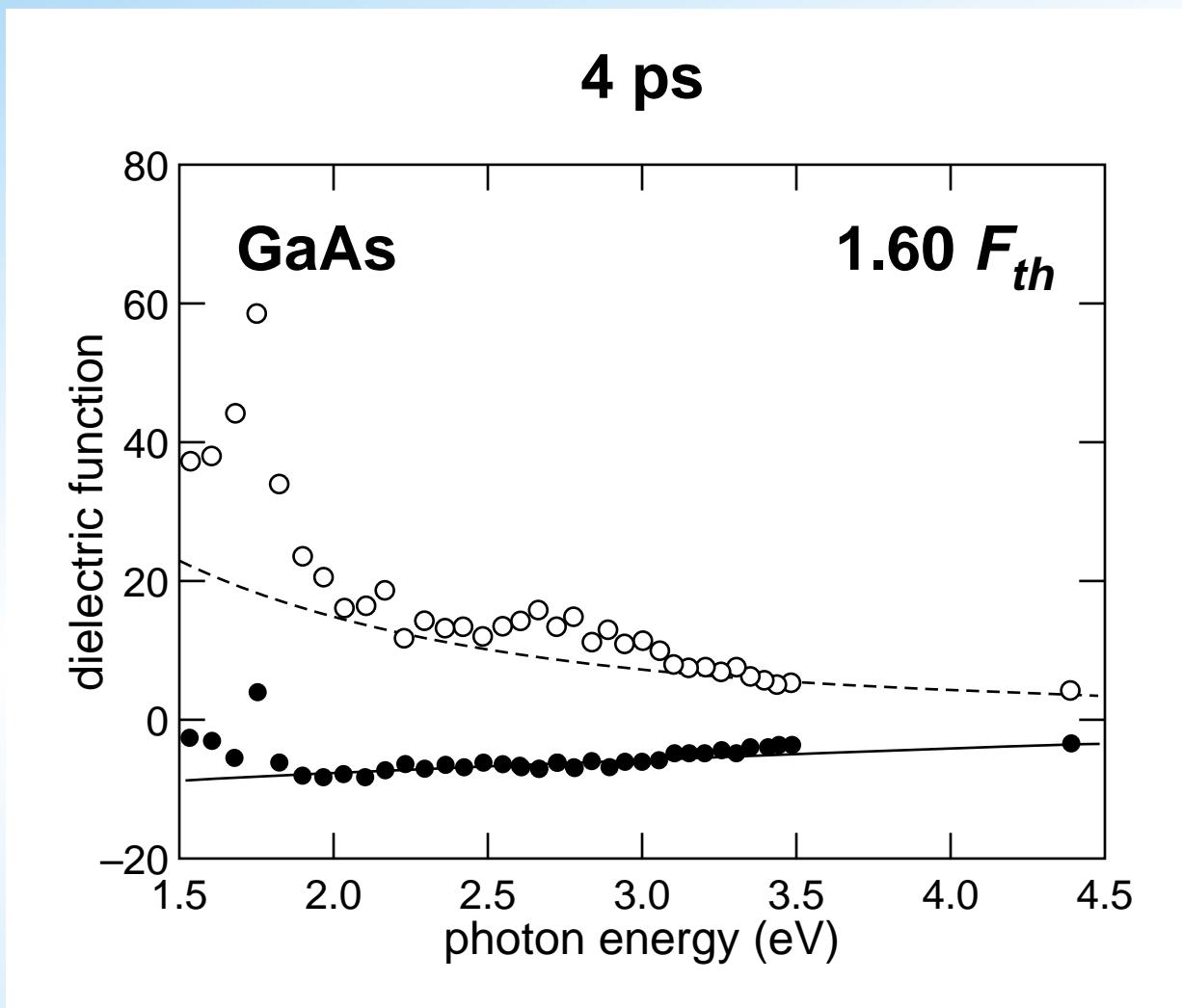
Electronic and structural transitions



Electronic and structural transitions



Electronic and structural transitions

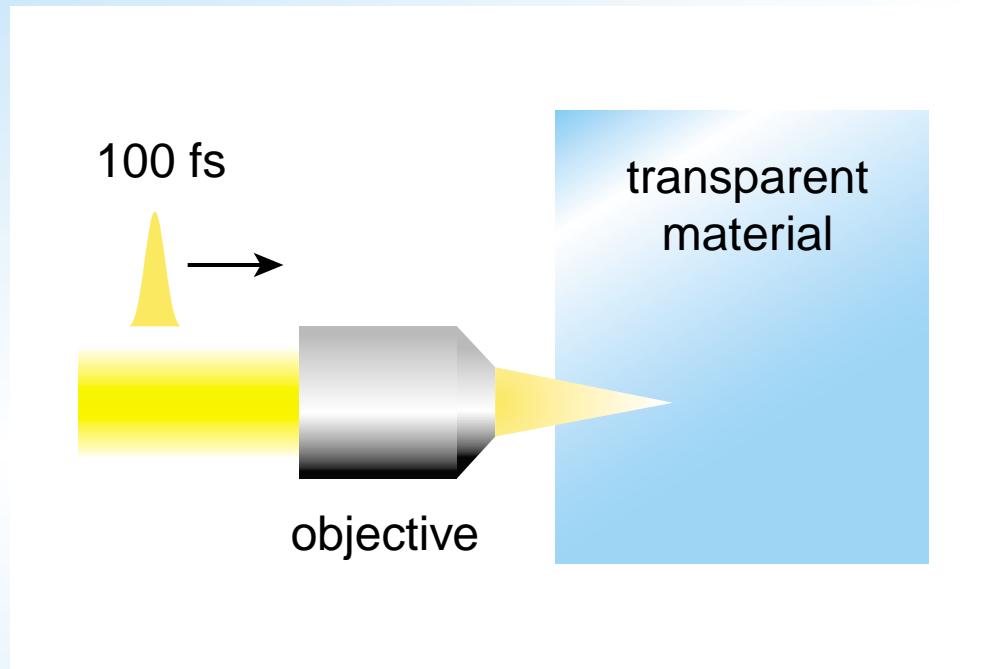


Overview

- ▶ surface femtochemistry
- ▶ electronic and structural transitions
- ▶ microstructuring of materials
- ▶ nonlinear optics and propagation
- ▶ laser surgery

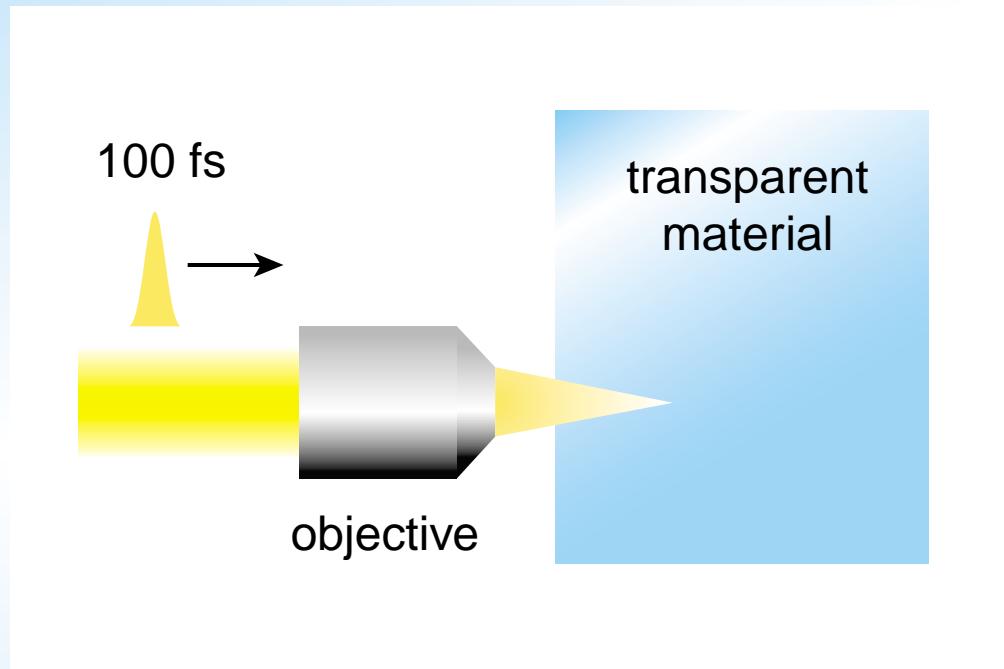
Microstructuring

focus laser beam inside material...



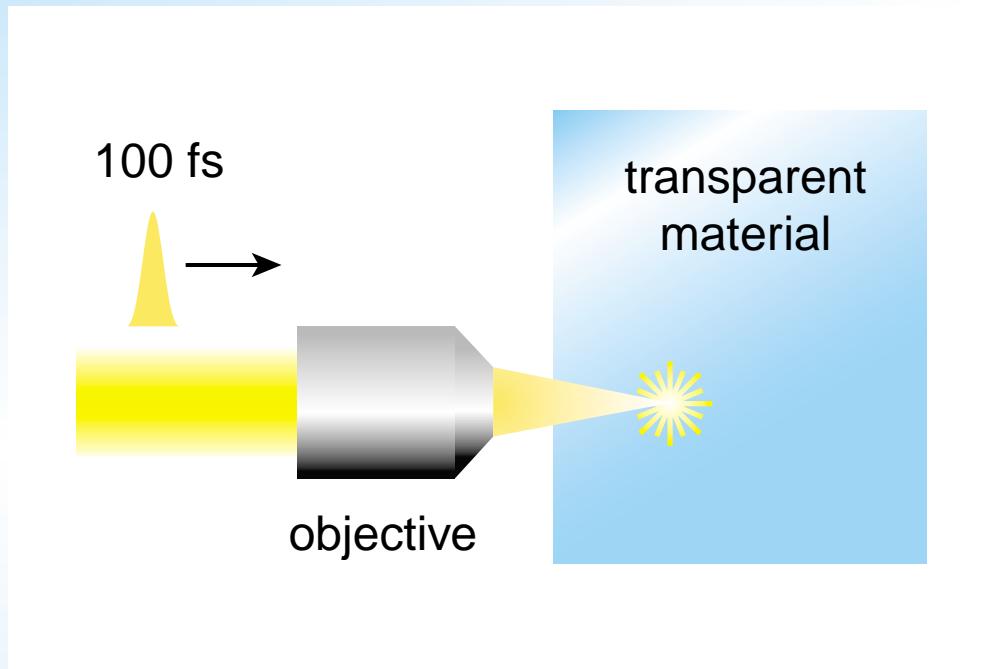
Microstructuring

high intensity at focus...



Microstructuring

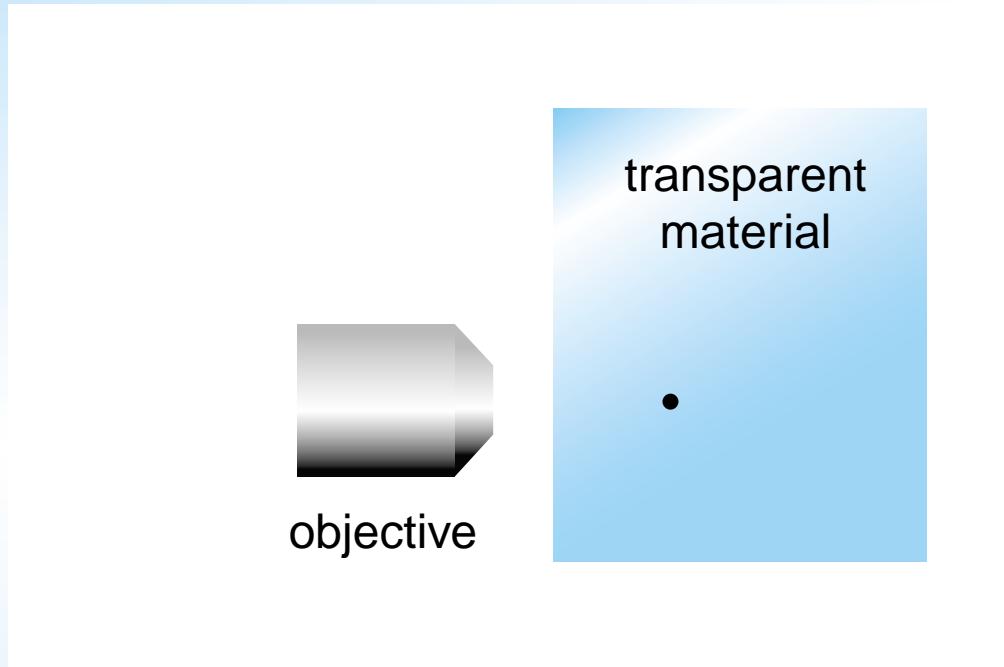
... causes nonlinear ionization...



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

Microstructuring

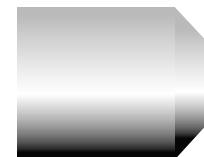
and microscopic bulk damage



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

Microstructuring

What are the conditions at focus?



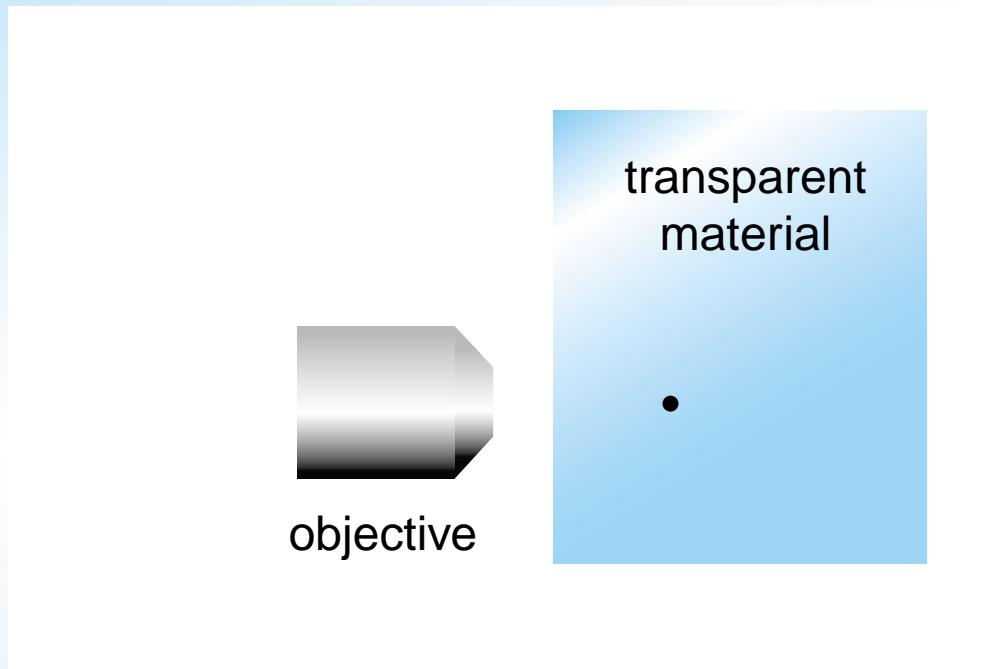
objective

transparent
material

•

Microstructuring

What are the conditions at focus?



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

Microstructuring

What temperature?

Microstructuring

What temperature?

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

Microstructuring

What temperature?

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

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

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

Microstructuring

What temperature?

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

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

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

So, 1 μJ in 1 μm^3 gives

~1,000,000 K!

Microstructuring

What pressure?

Microstructuring

What pressure?

Treat ionized material as an ideal gas:

$$pV = nRT$$

Microstructuring

What pressure?

Treat ionized material as an ideal gas:

$$pV = nRT$$

Gives

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

Microstructuring

So:

microexplosion

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

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

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

Microstructuring

So:

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

Microstructuring

So:

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

creating stellar conditions on an optical bench!

Microstructuring

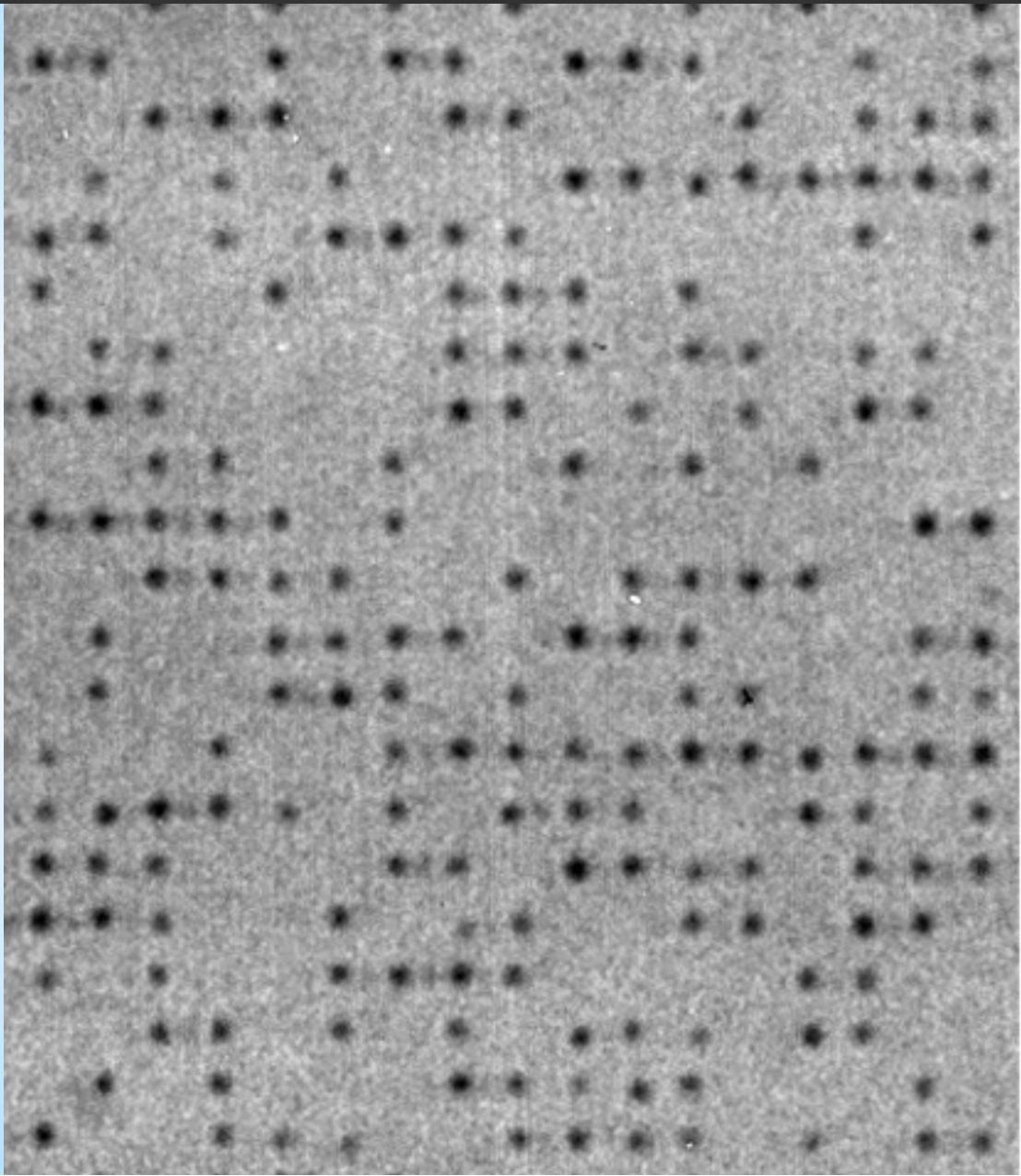
optical microscopy

$2 \times 2 \mu\text{m}$ array

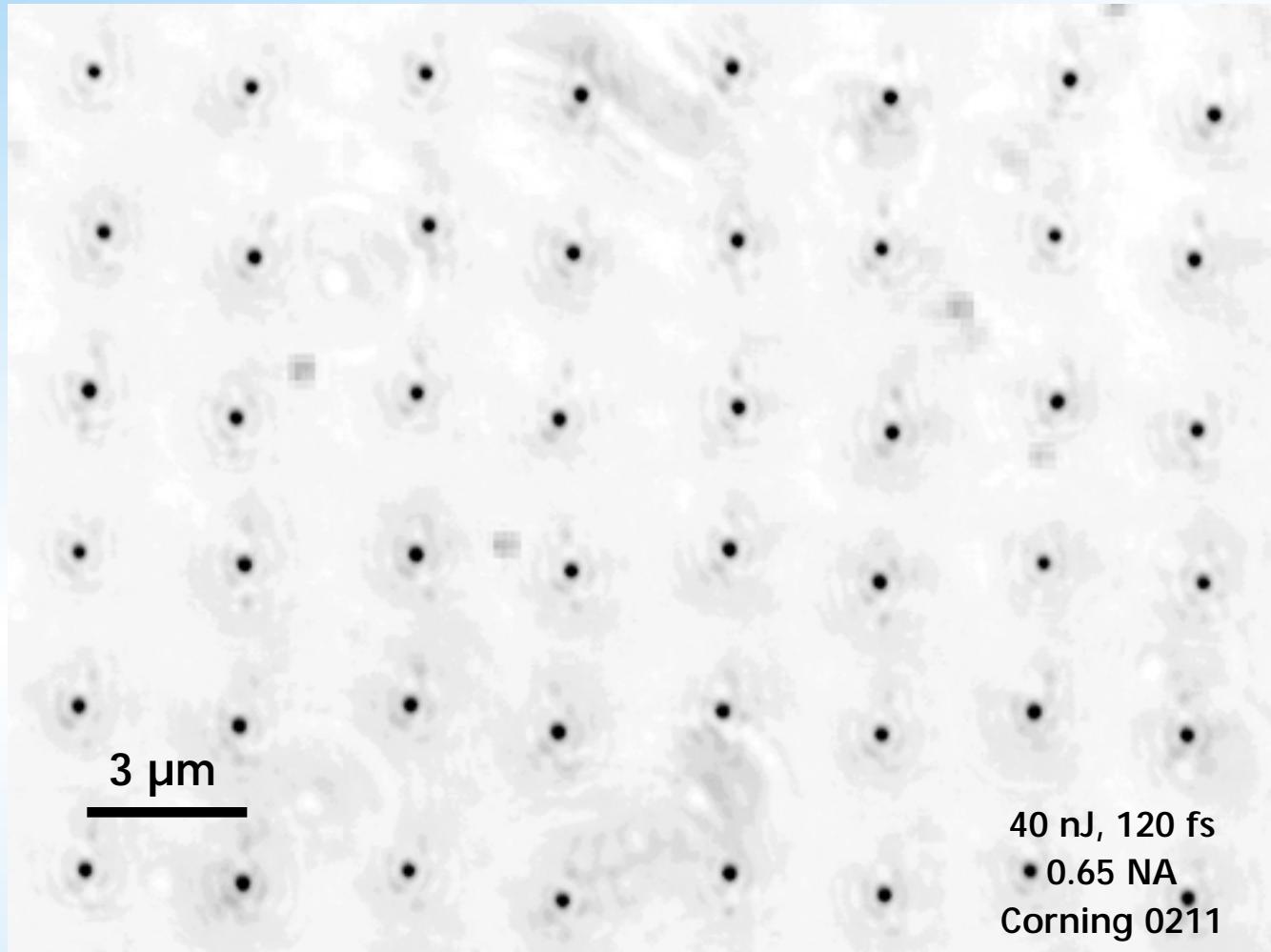
fused silica

$0.5 \mu\text{J}$, 100 fs, 800 nm

Opt. Lett. 21, 2023 (1996)



Microstructuring



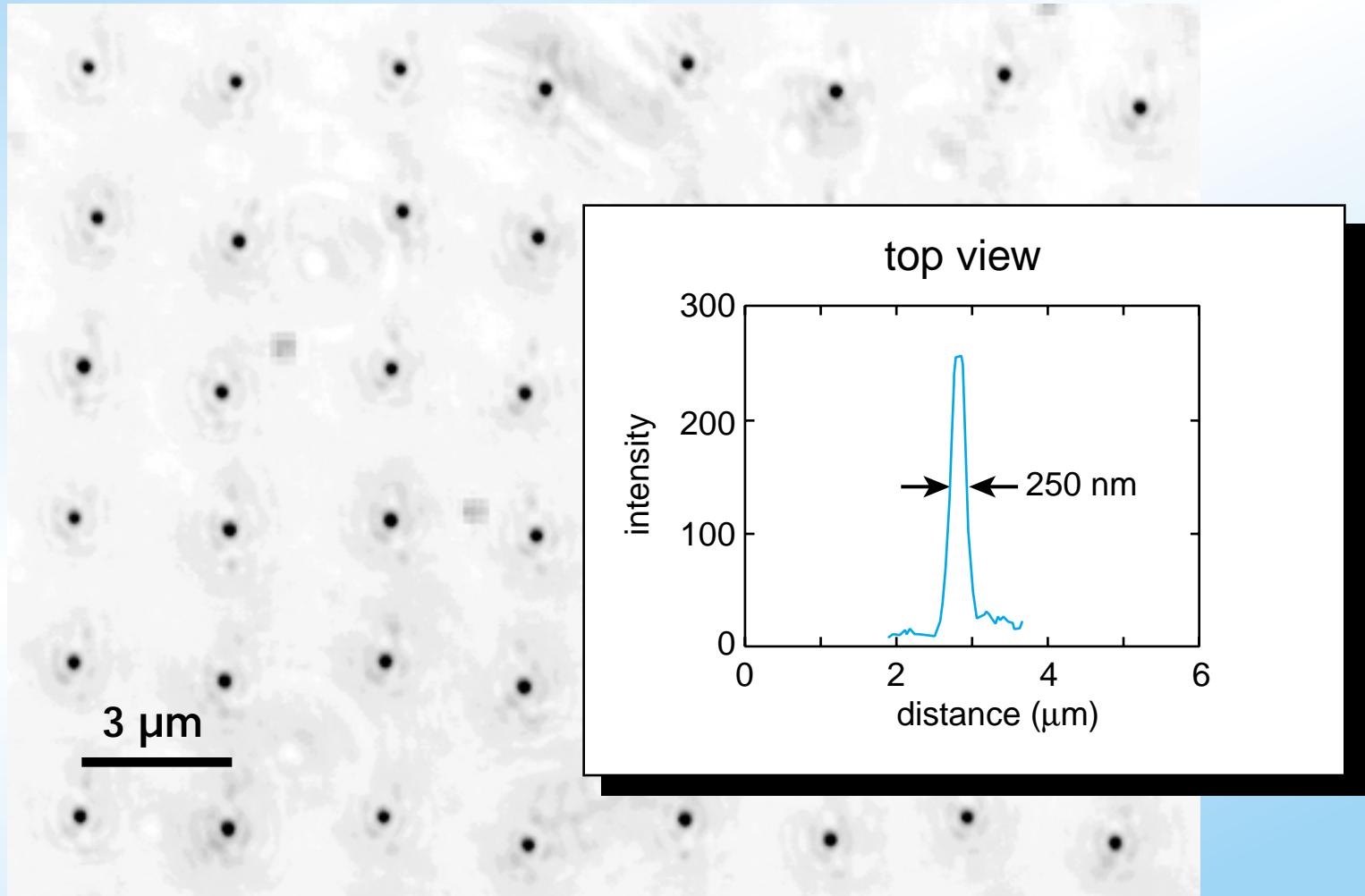
3 μm

40 nJ, 120 fs

0.65 NA

Corning 0211

Microstructuring



Microstructuring



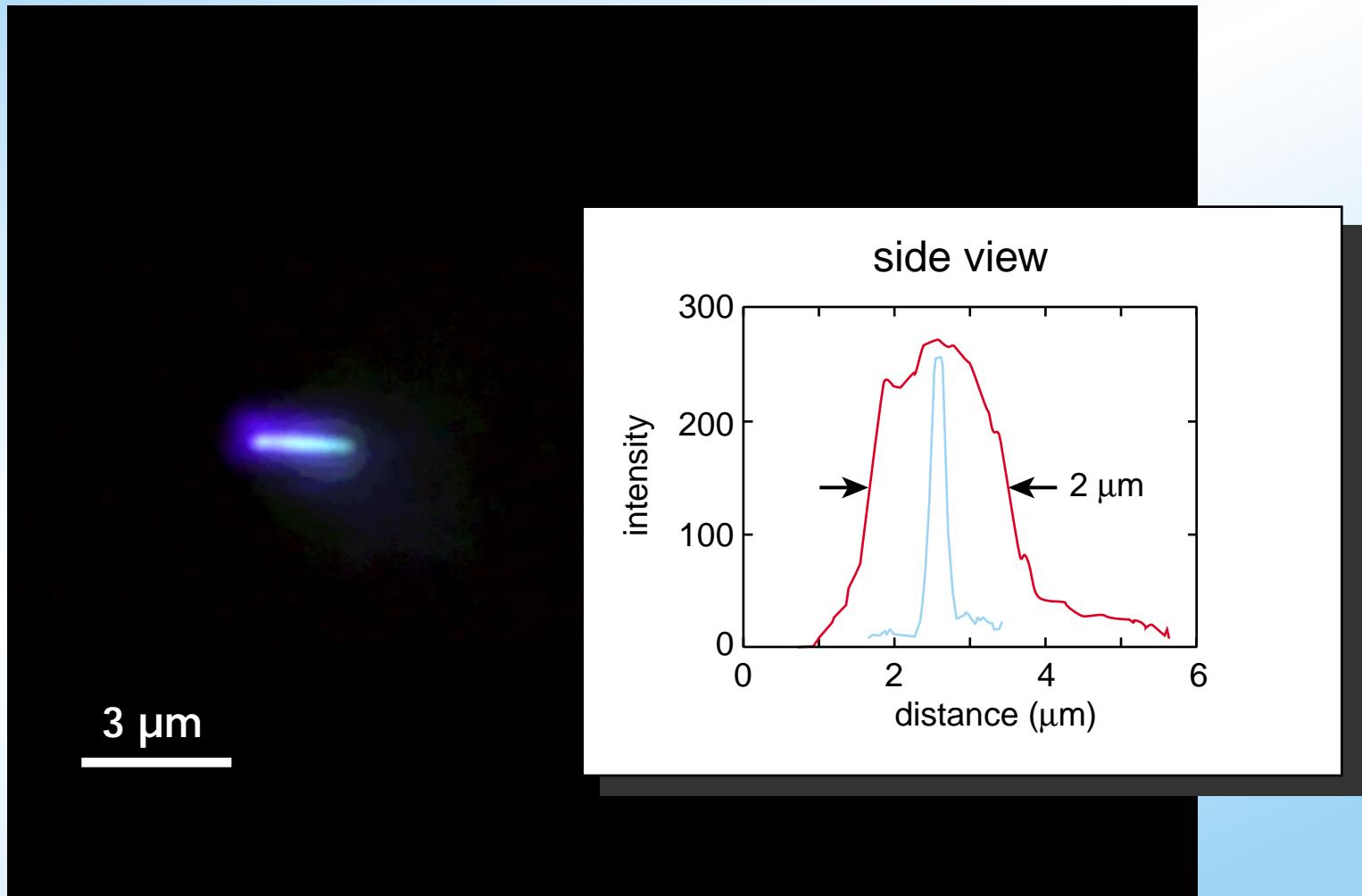
3 μm

40 nJ, 120 fs

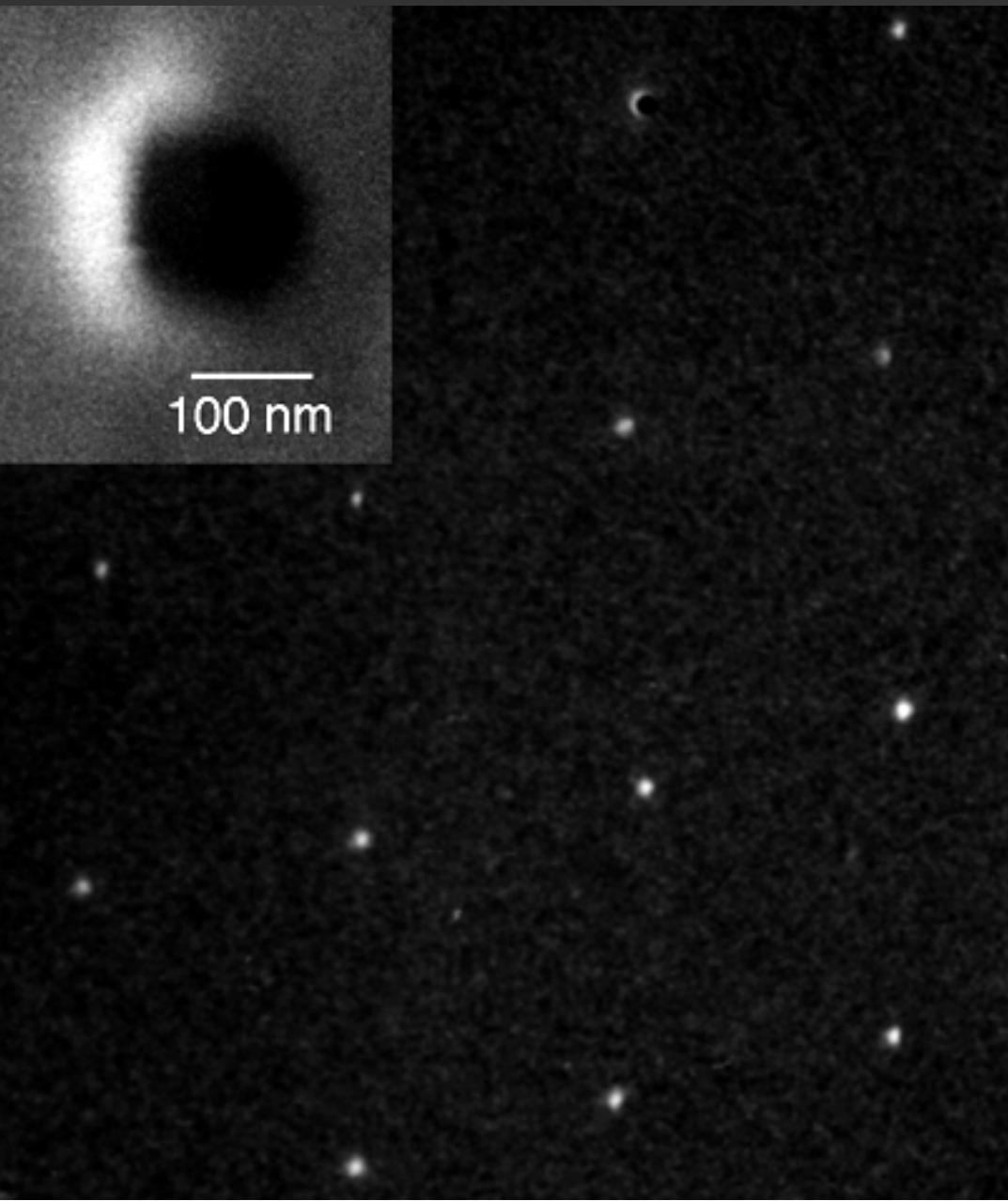
0.65 NA

Corning 0211

Microstructuring



Microstructuring



SEM:

bumps & pits!

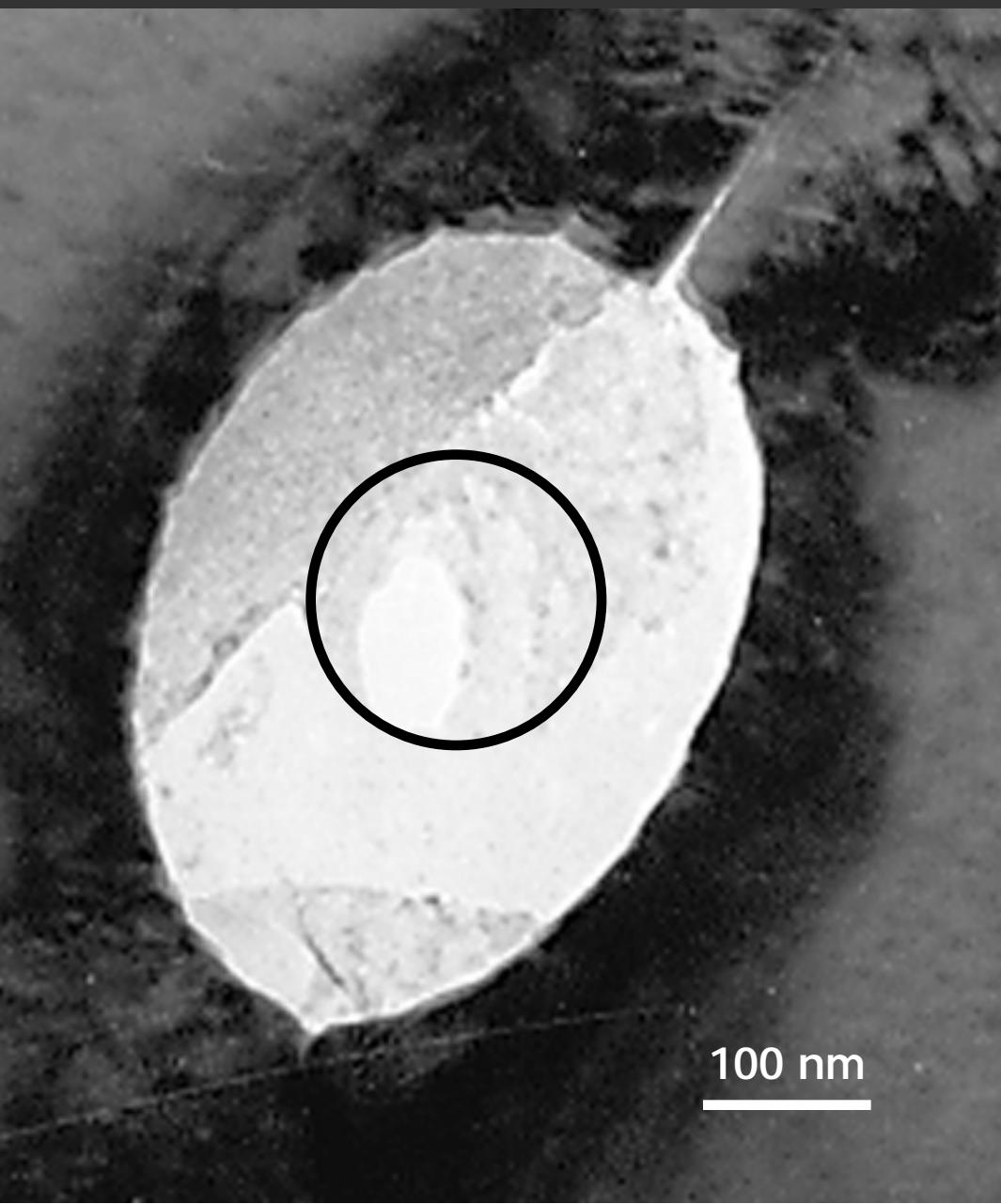
Microstructuring



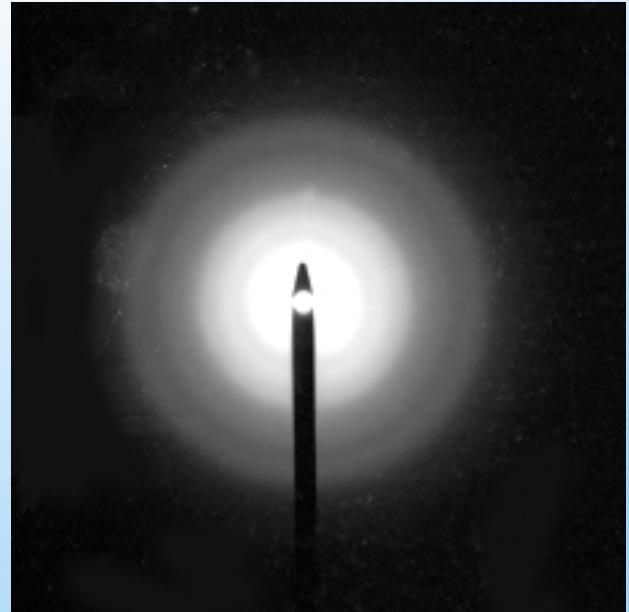
TEM picture

sapphire

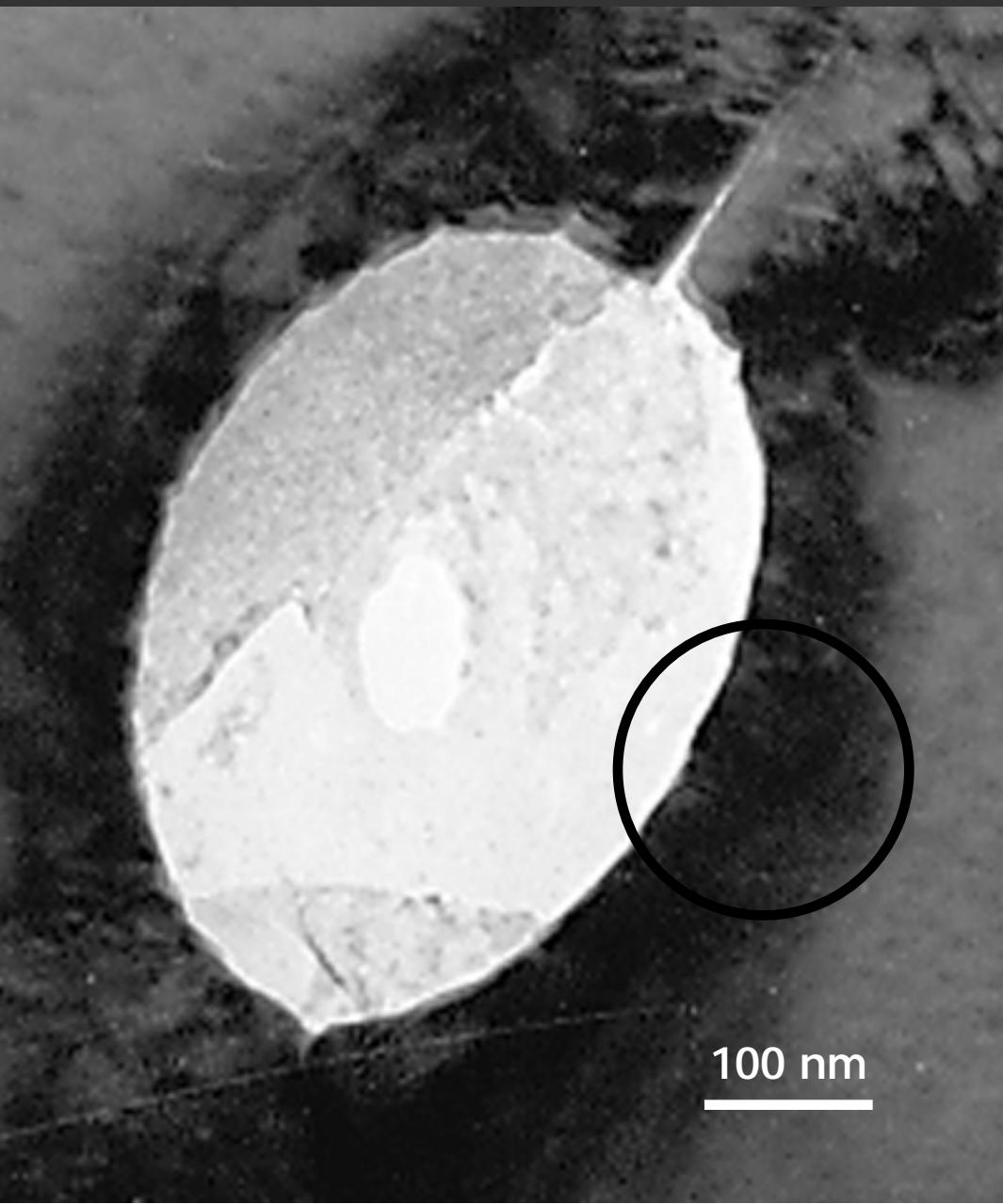
Microstructuring



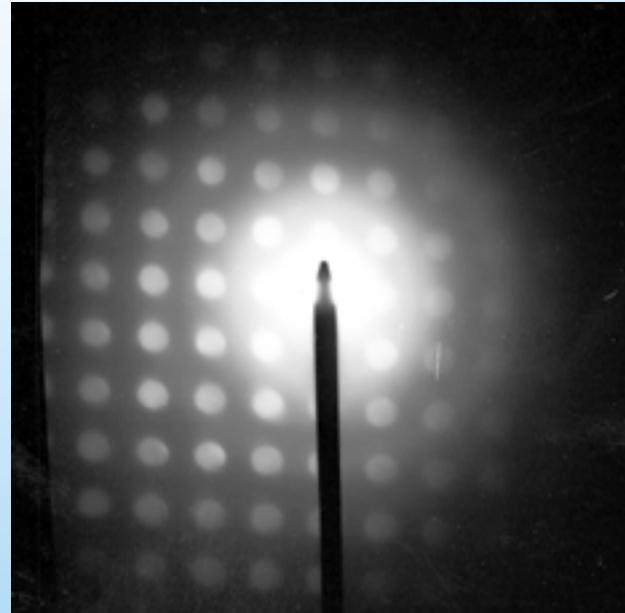
electron diffraction:
amorphous?



Microstructuring

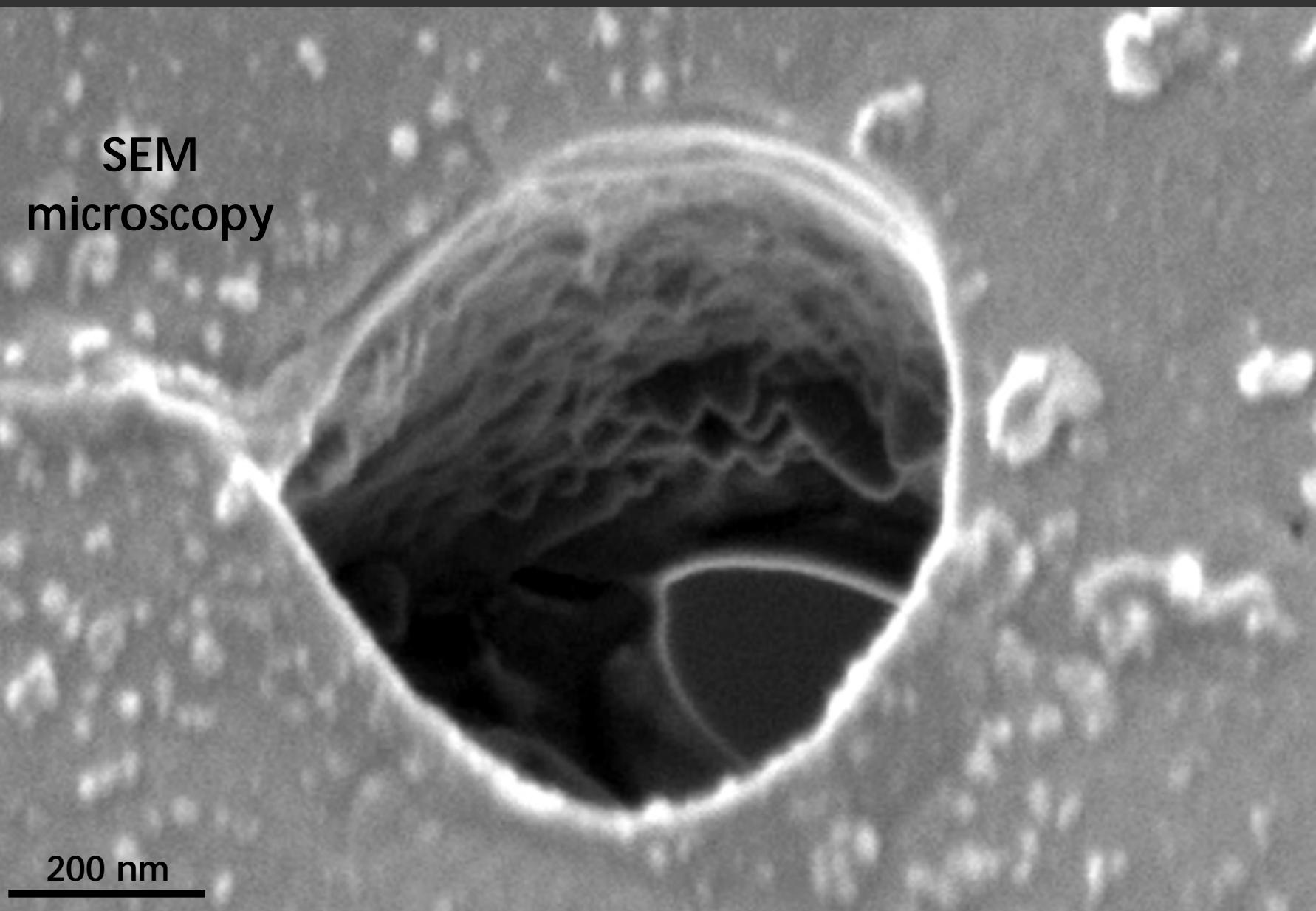


**electron diffraction:
crystalline**



Microstructuring

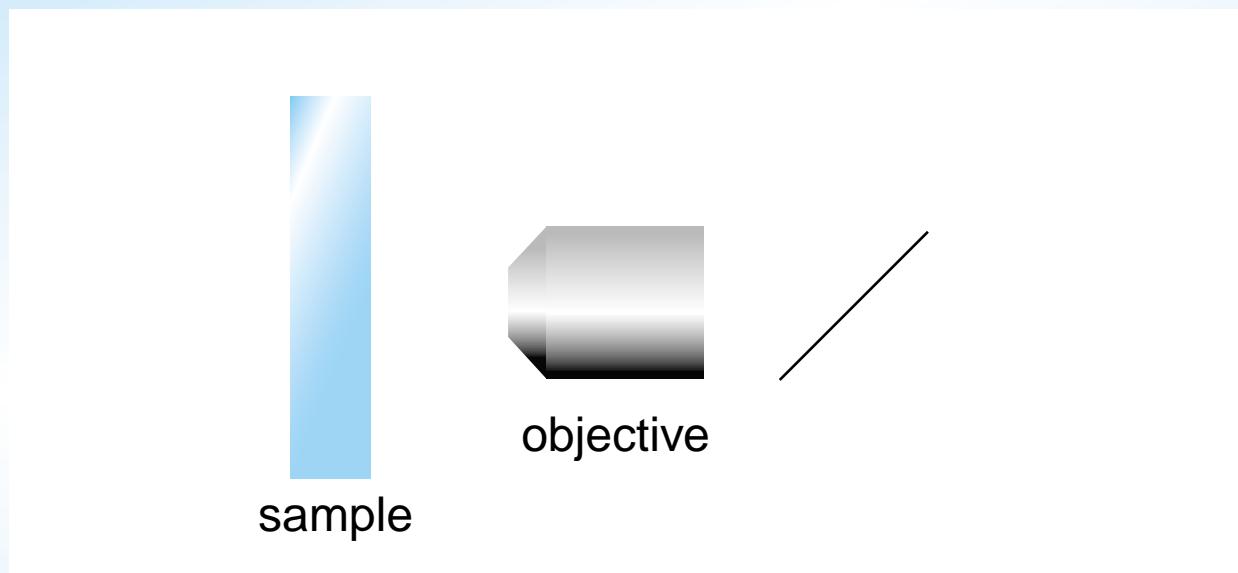
SEM
microscopy



200 nm

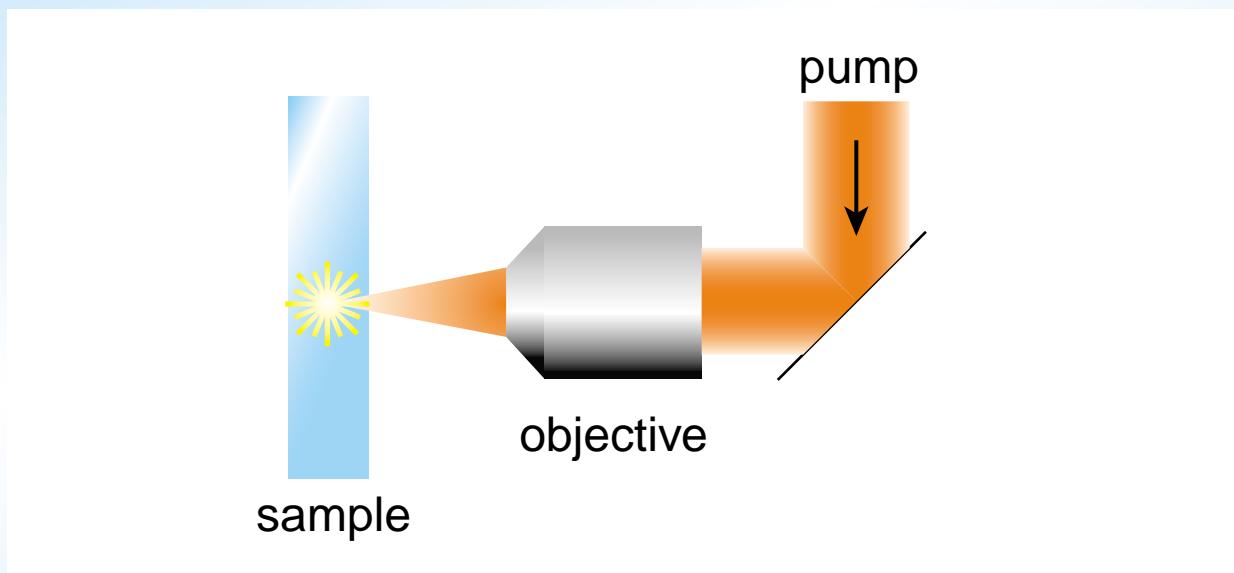
Microstructuring

imaging setup



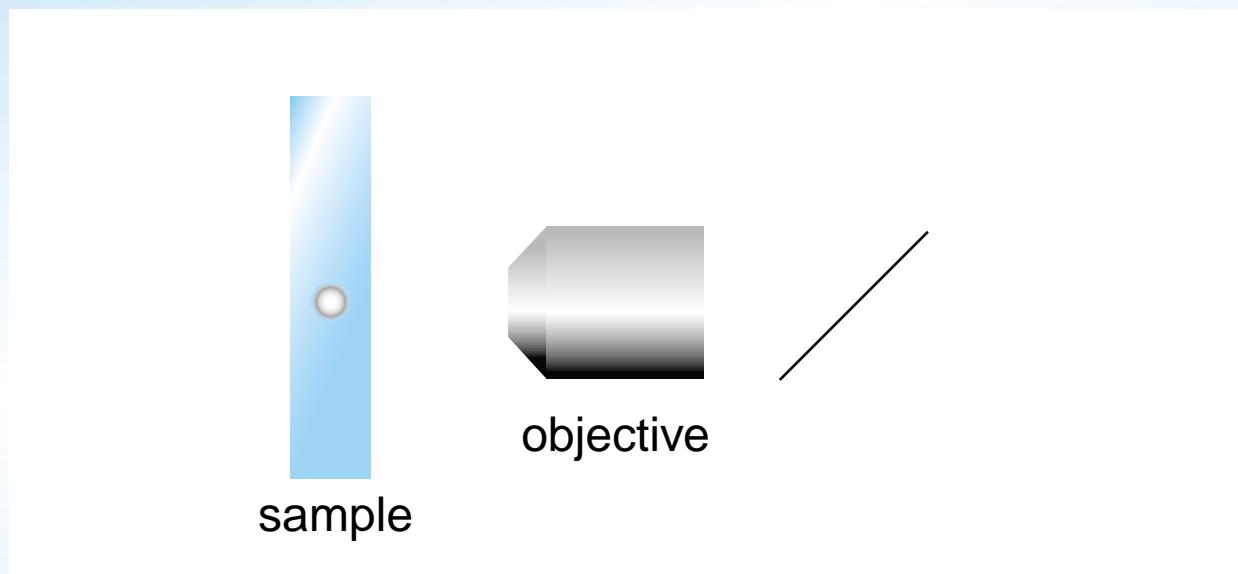
Microstructuring

imaging setup



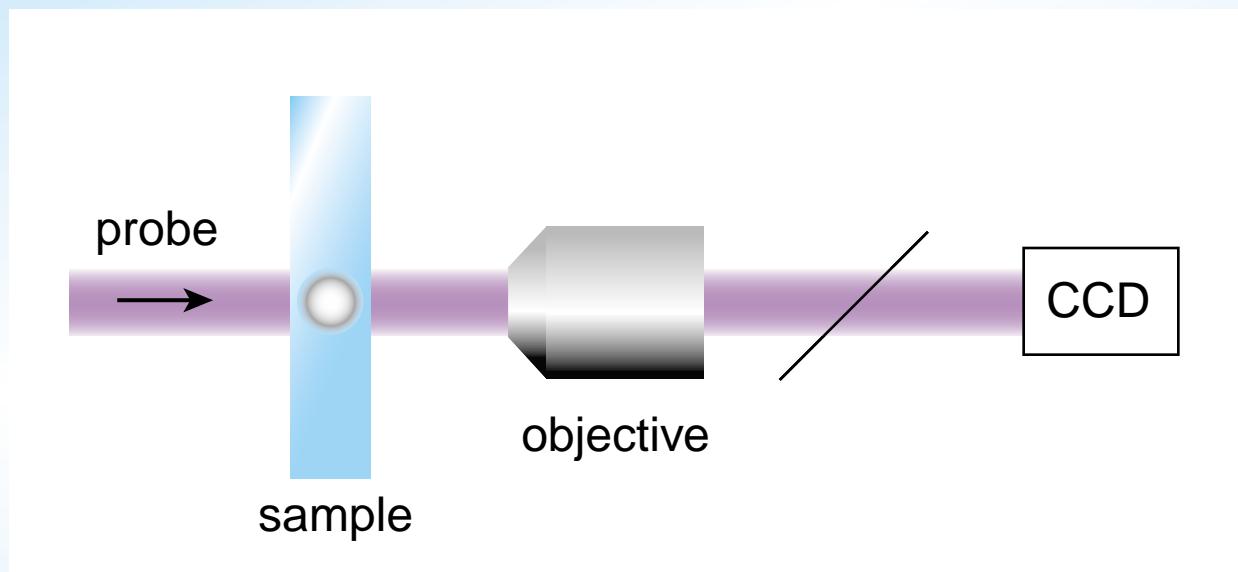
Microstructuring

imaging setup



Microstructuring

imaging setup



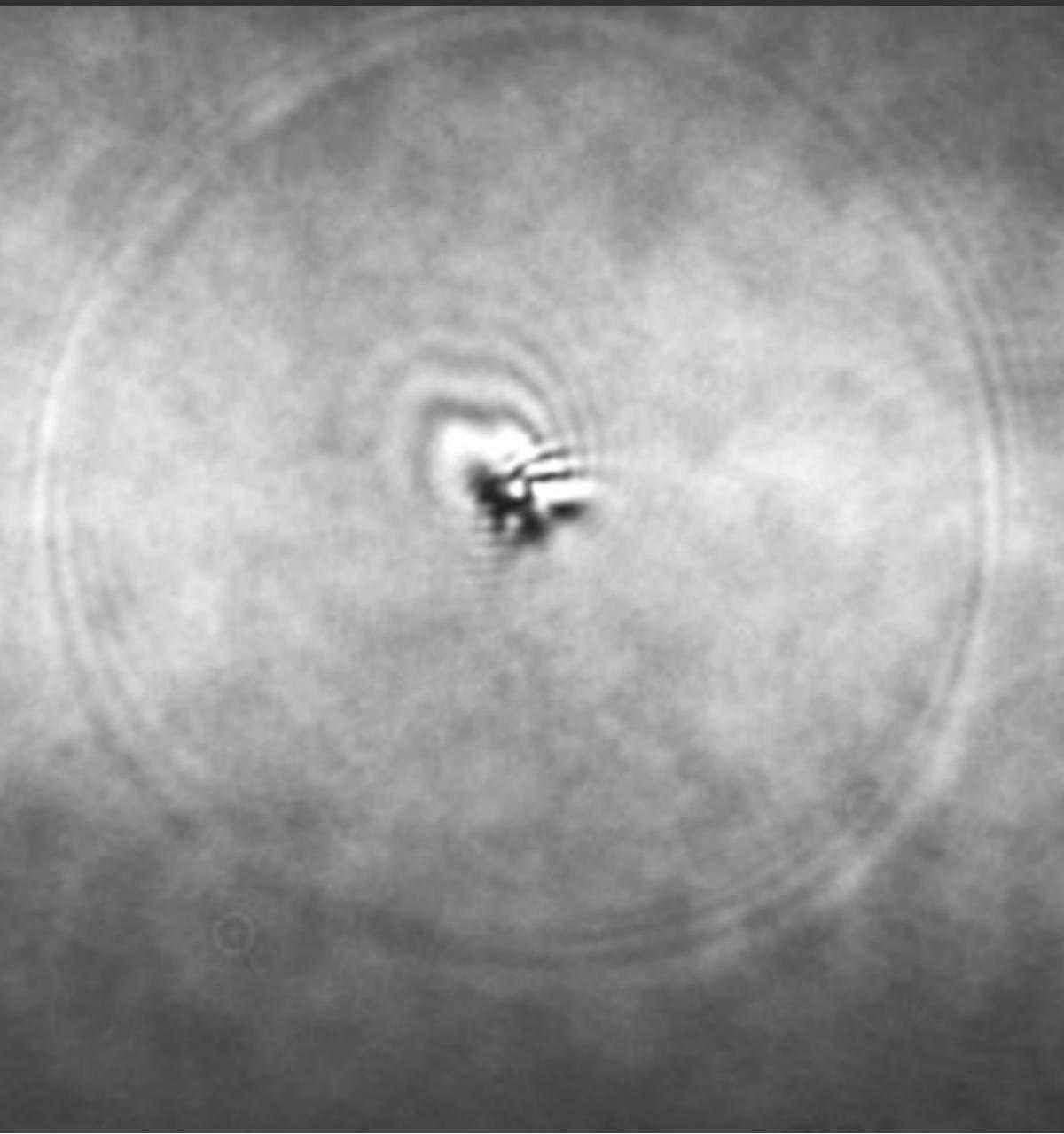
Microstructuring

sapphire

3 μJ pulse

3.8 ns delay

40 μm radius



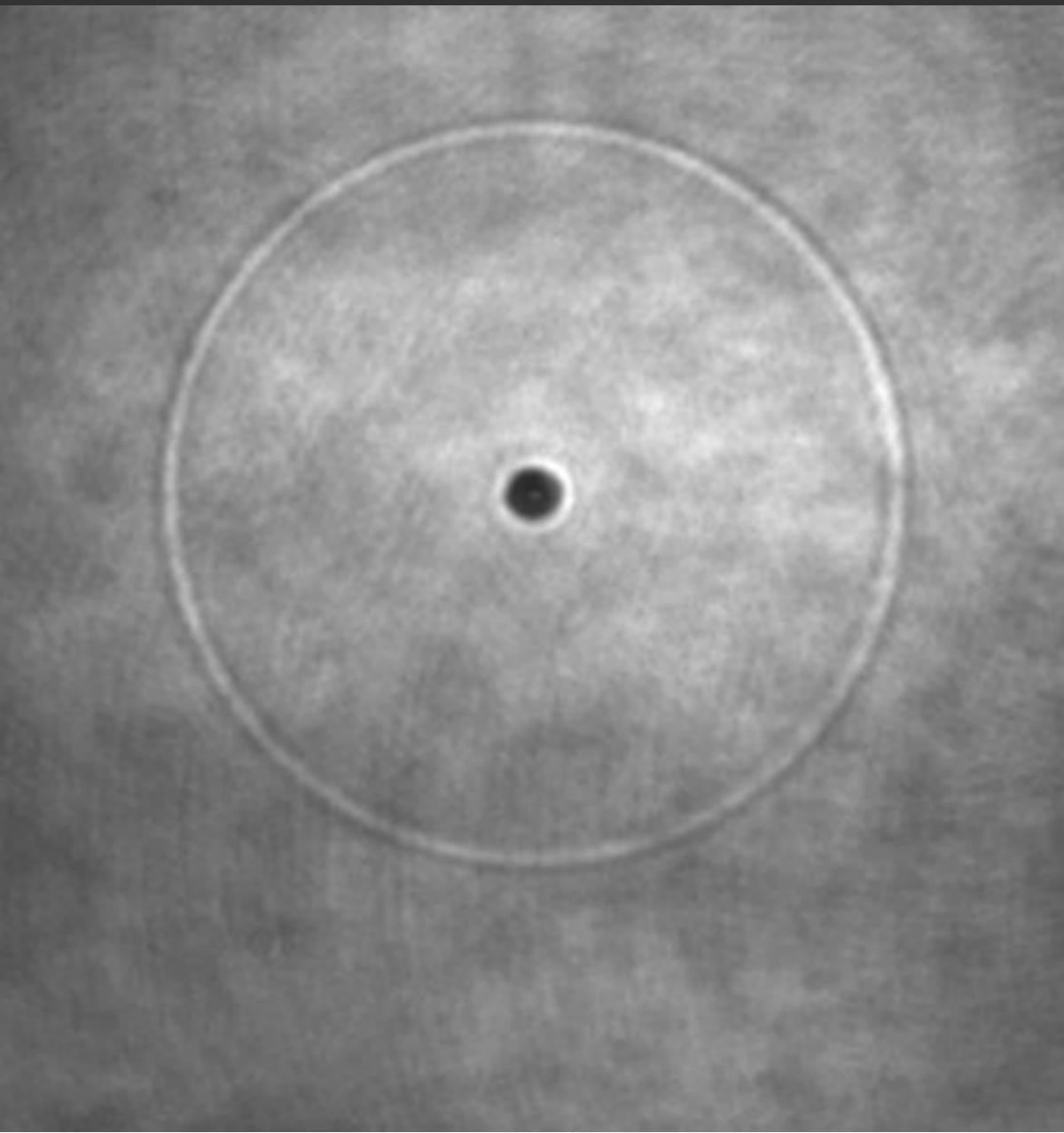
Microstructuring

water

1.0 μJ pulse

35 ns delay

58 μm radius



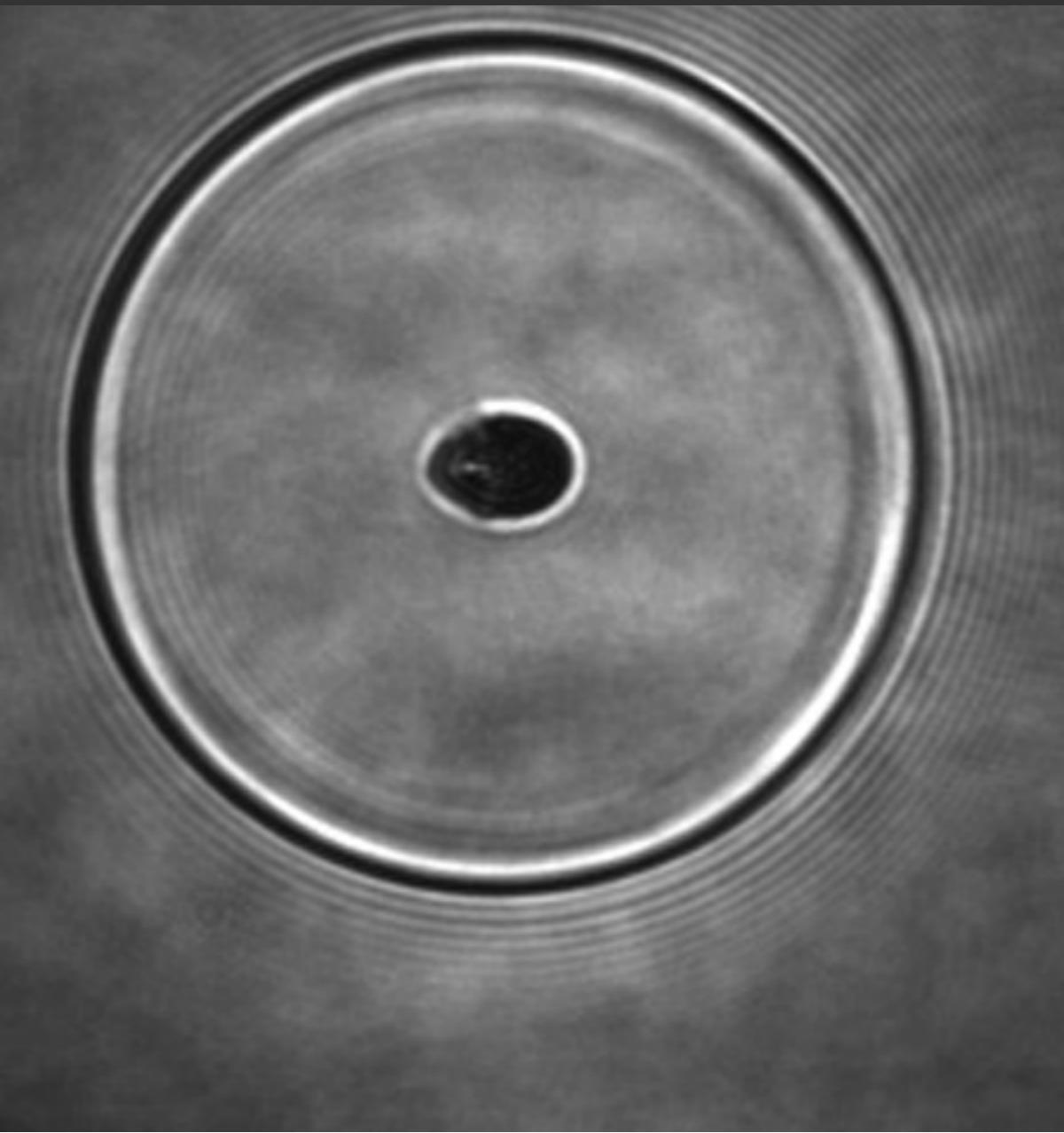
Microstructuring

water

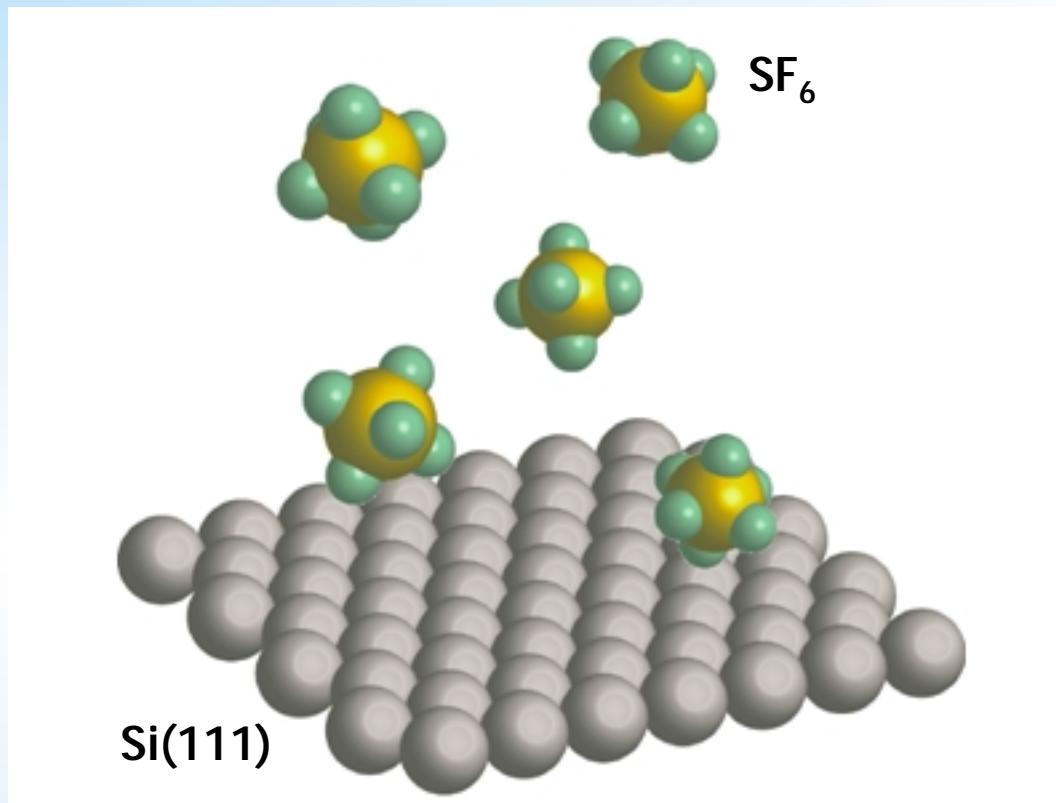
14 μJ pulse

35 ns delay

64 μm radius

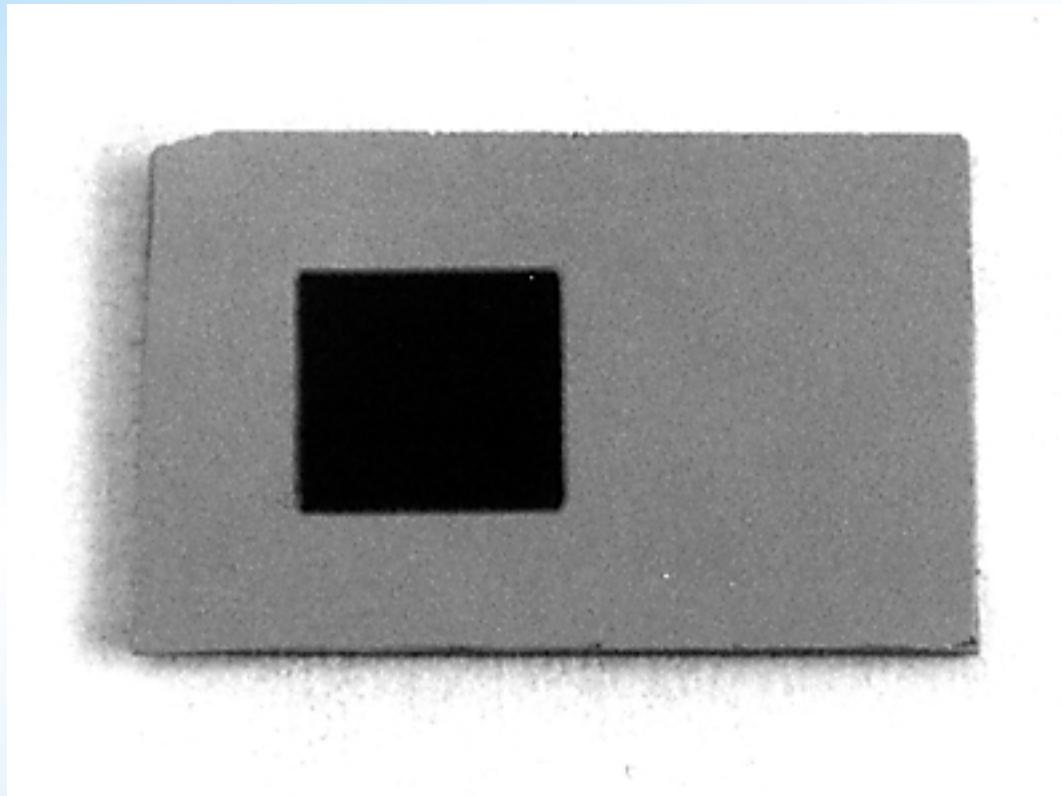


Microstructuring



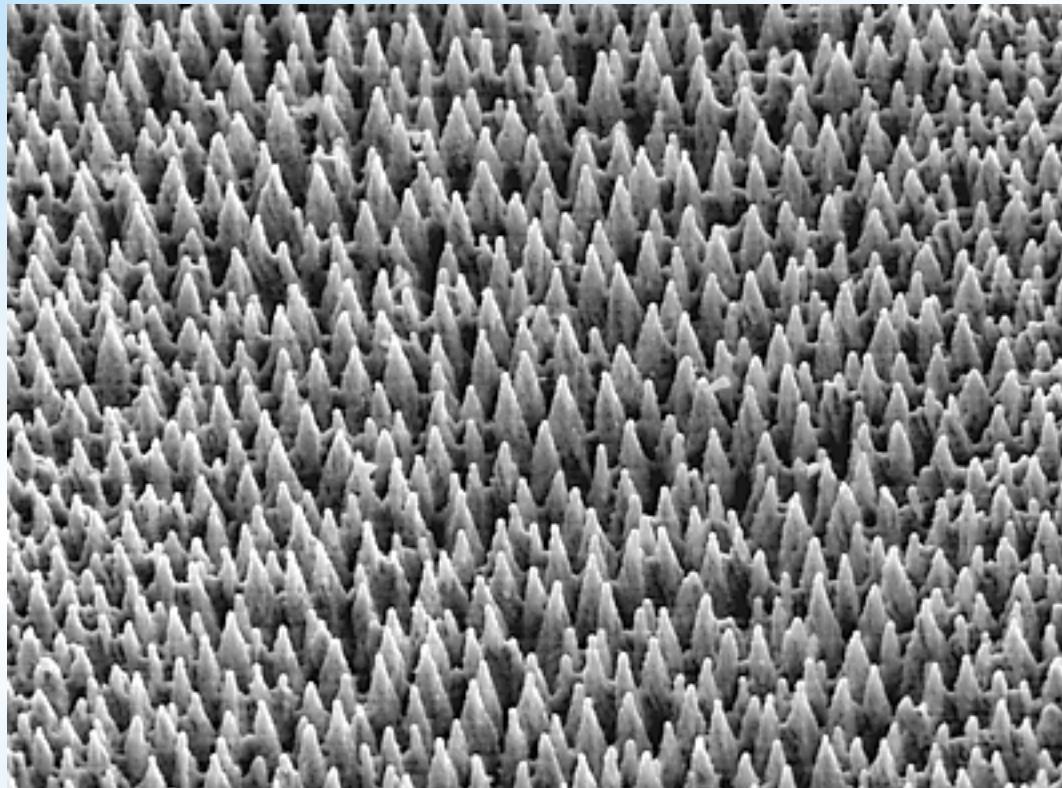
irradiate with 100 fs 10 kJ/m^2 laser pulses

Microstructuring



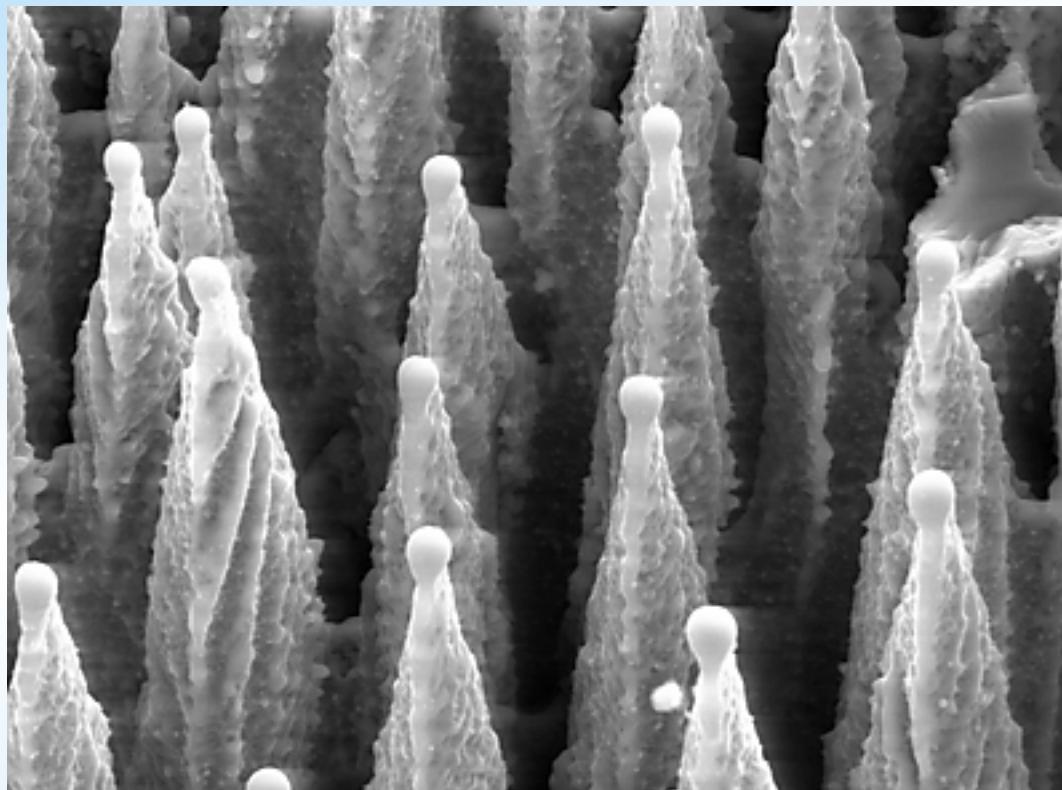
5 mm

Microstructuring



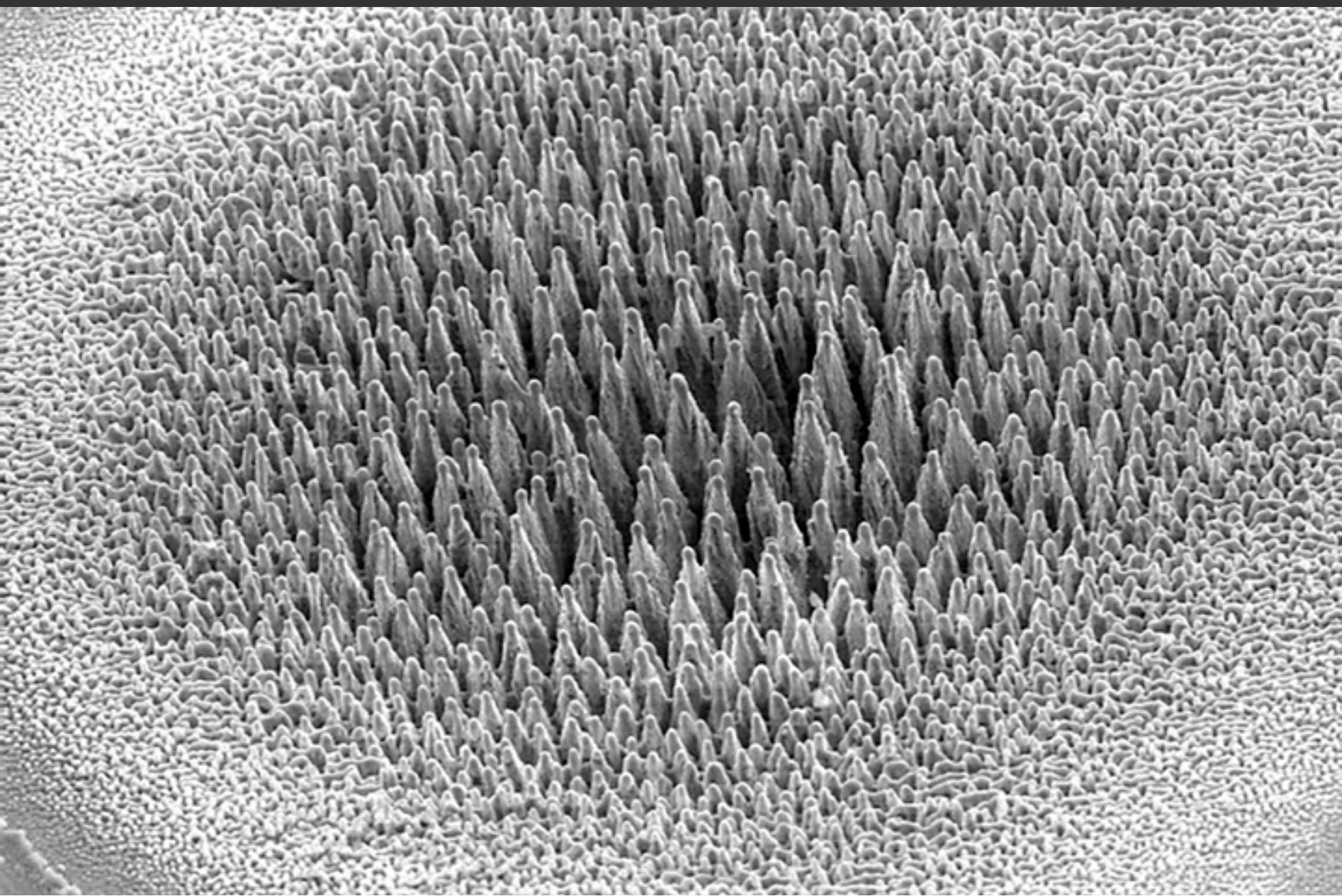
20 μm

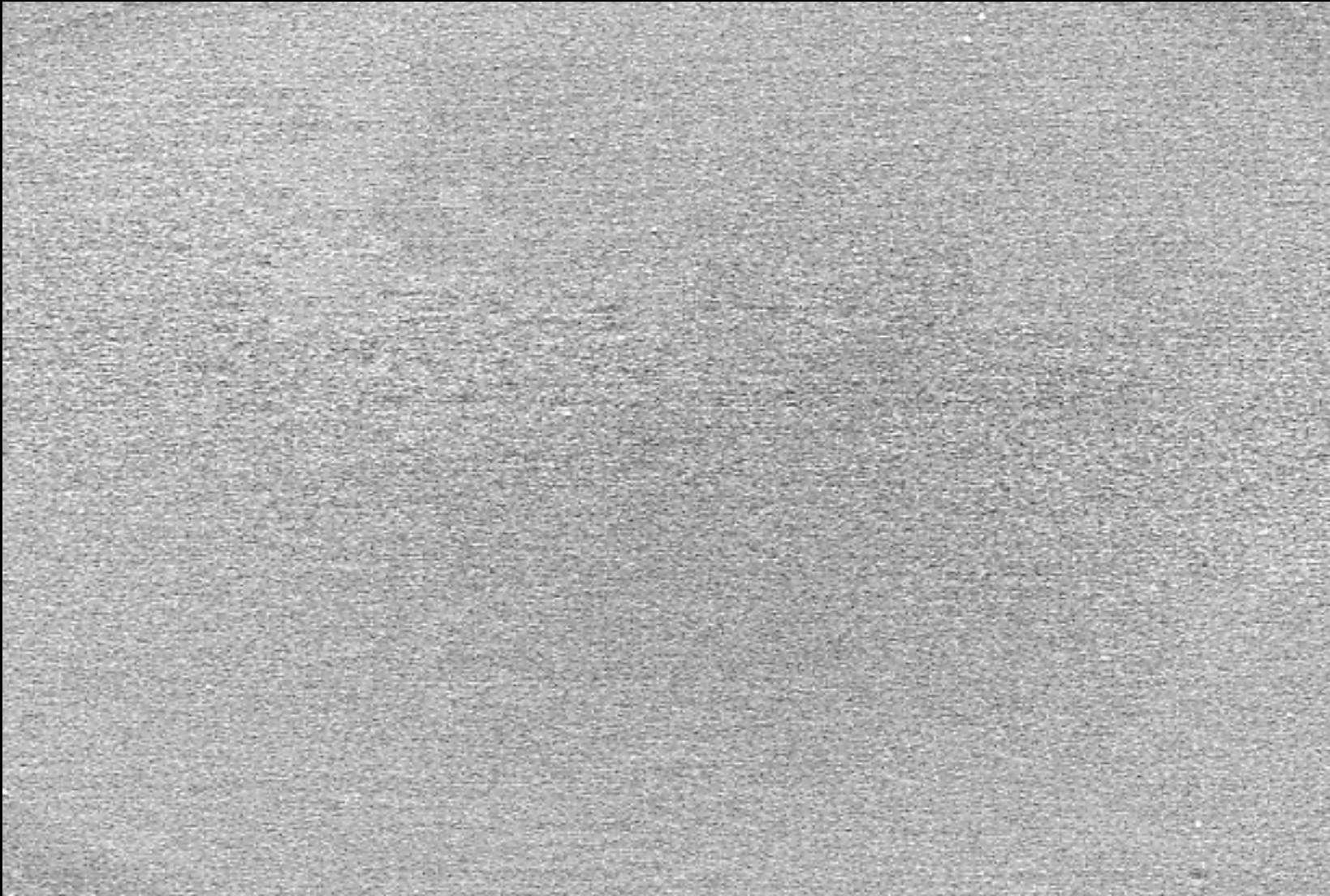
Microstructuring



4 μm

Microstructuring





x2000

#3548

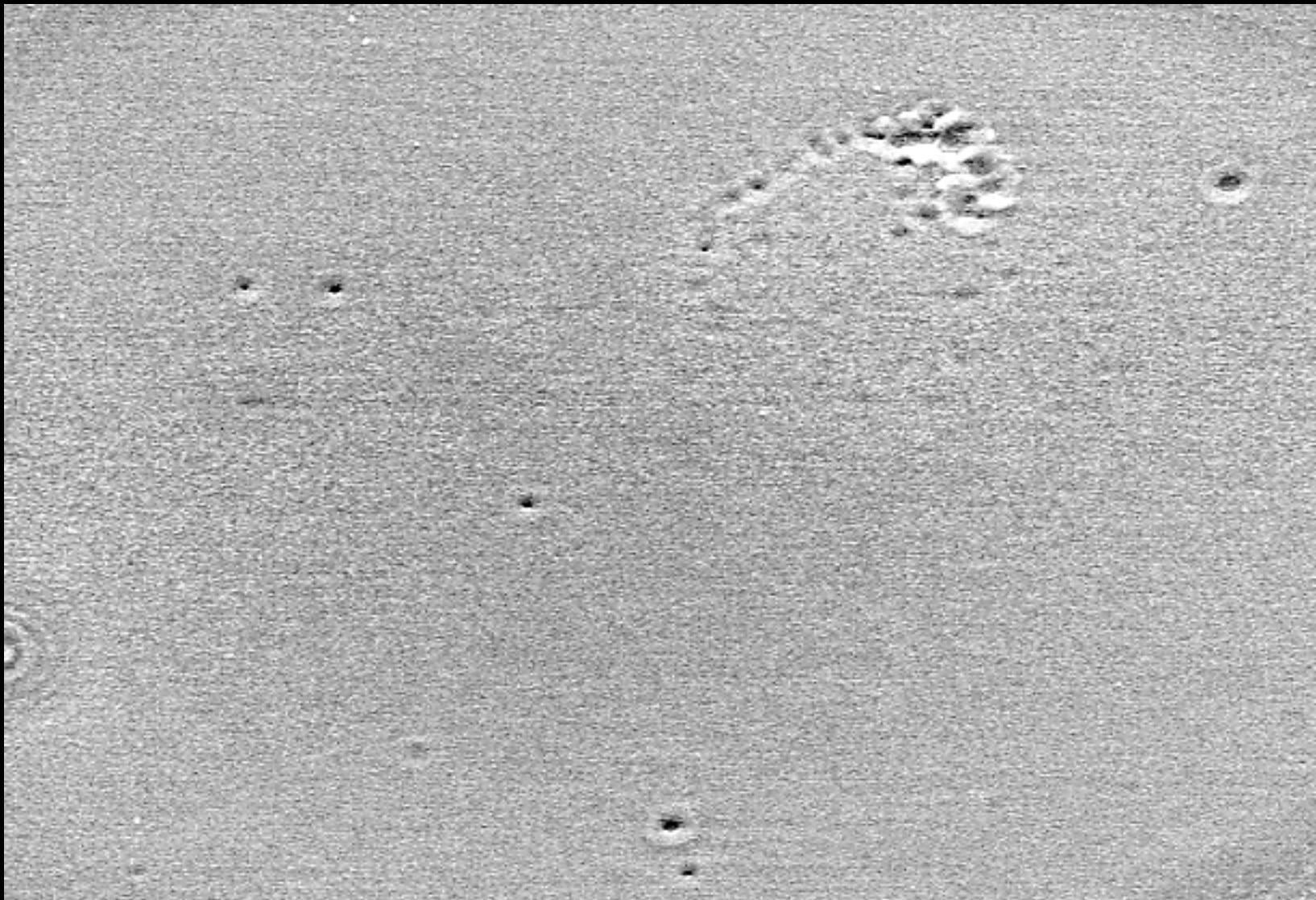
512 x 480

20 μm

10kV

15mm

0000



x2000

#3548

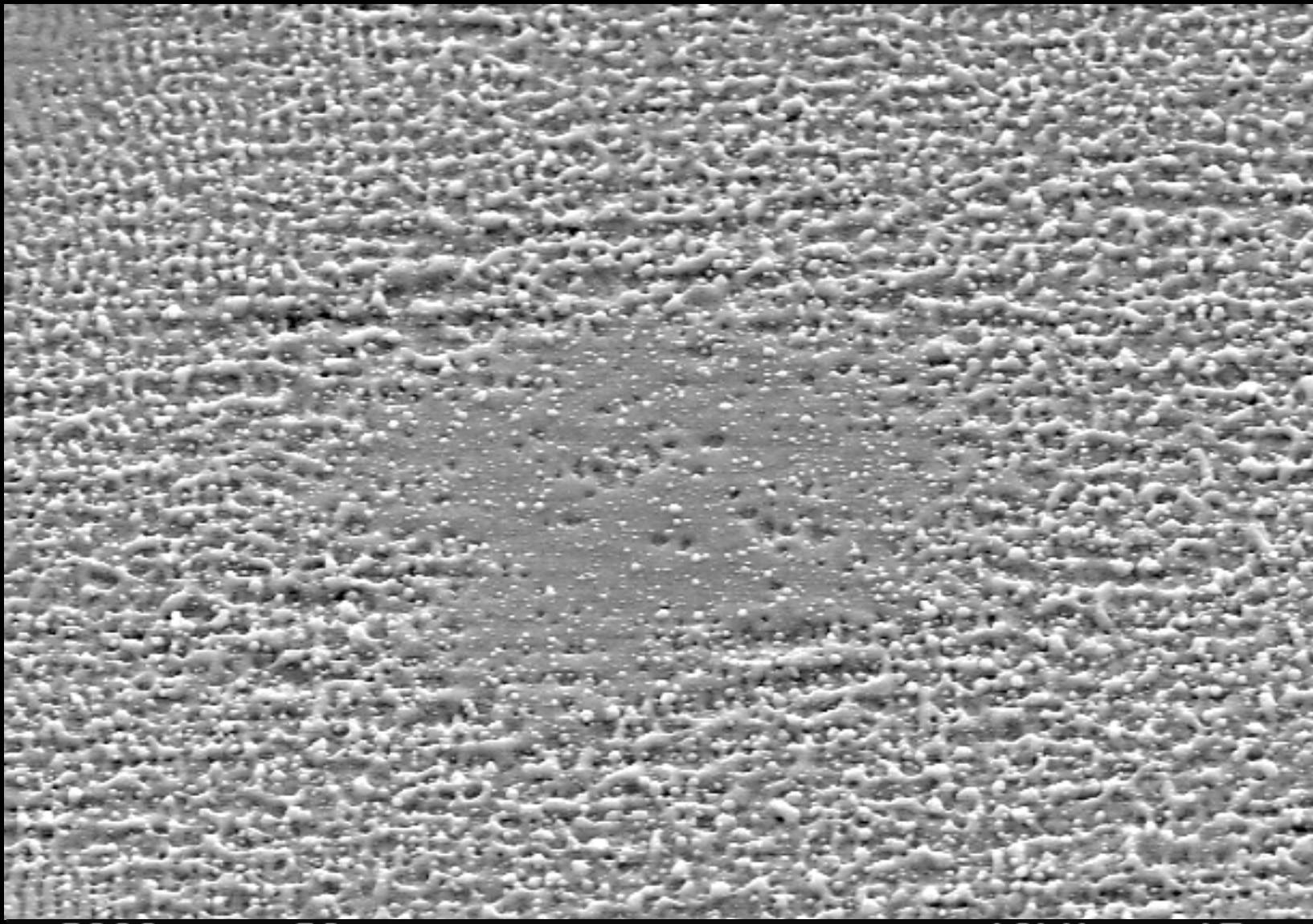
512 x 480

20 μm

10kV

15mm

0001



x2000

20 μm

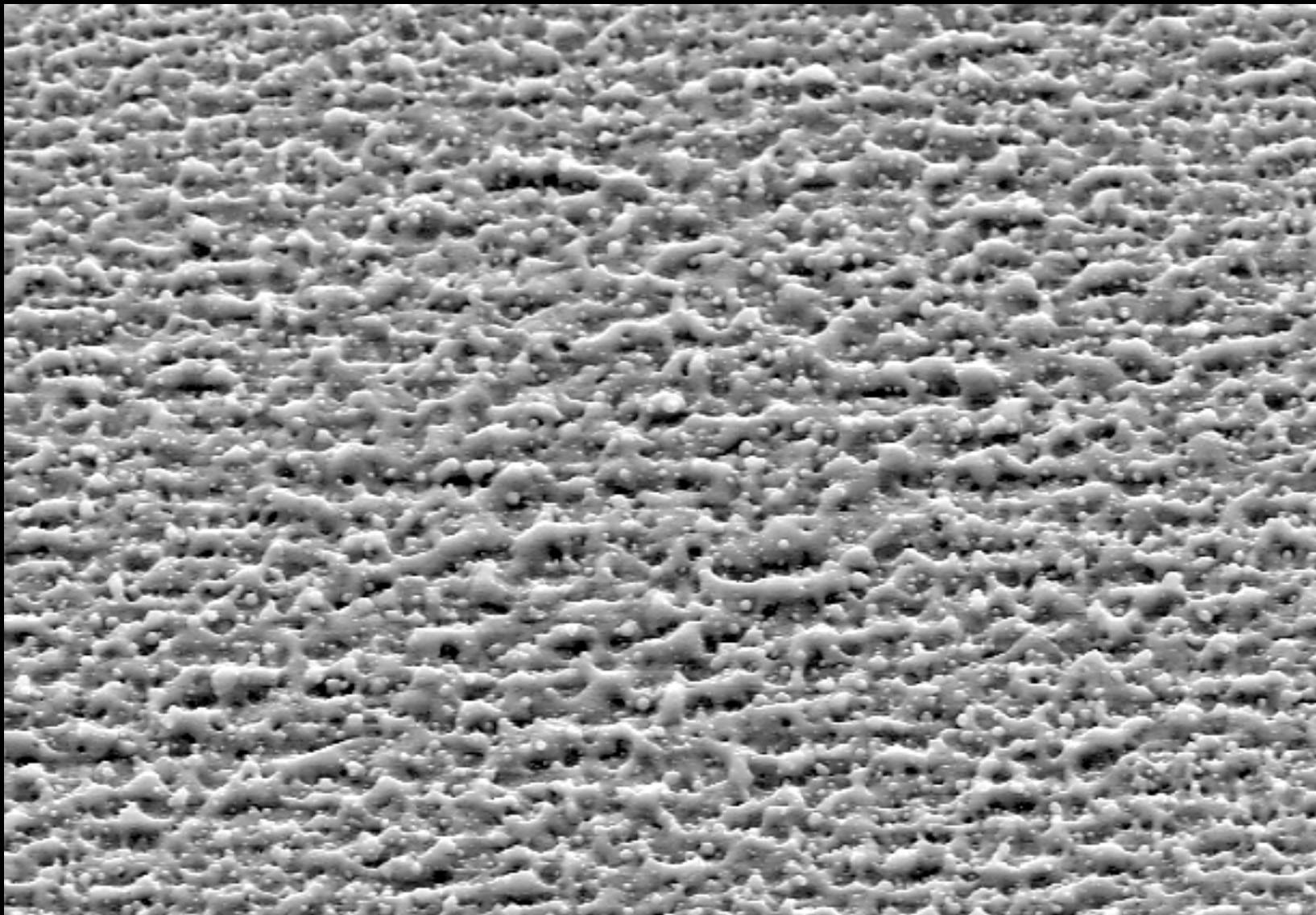
#3548

512 x 480

10kV

15mm

0005



x2000

20 μm

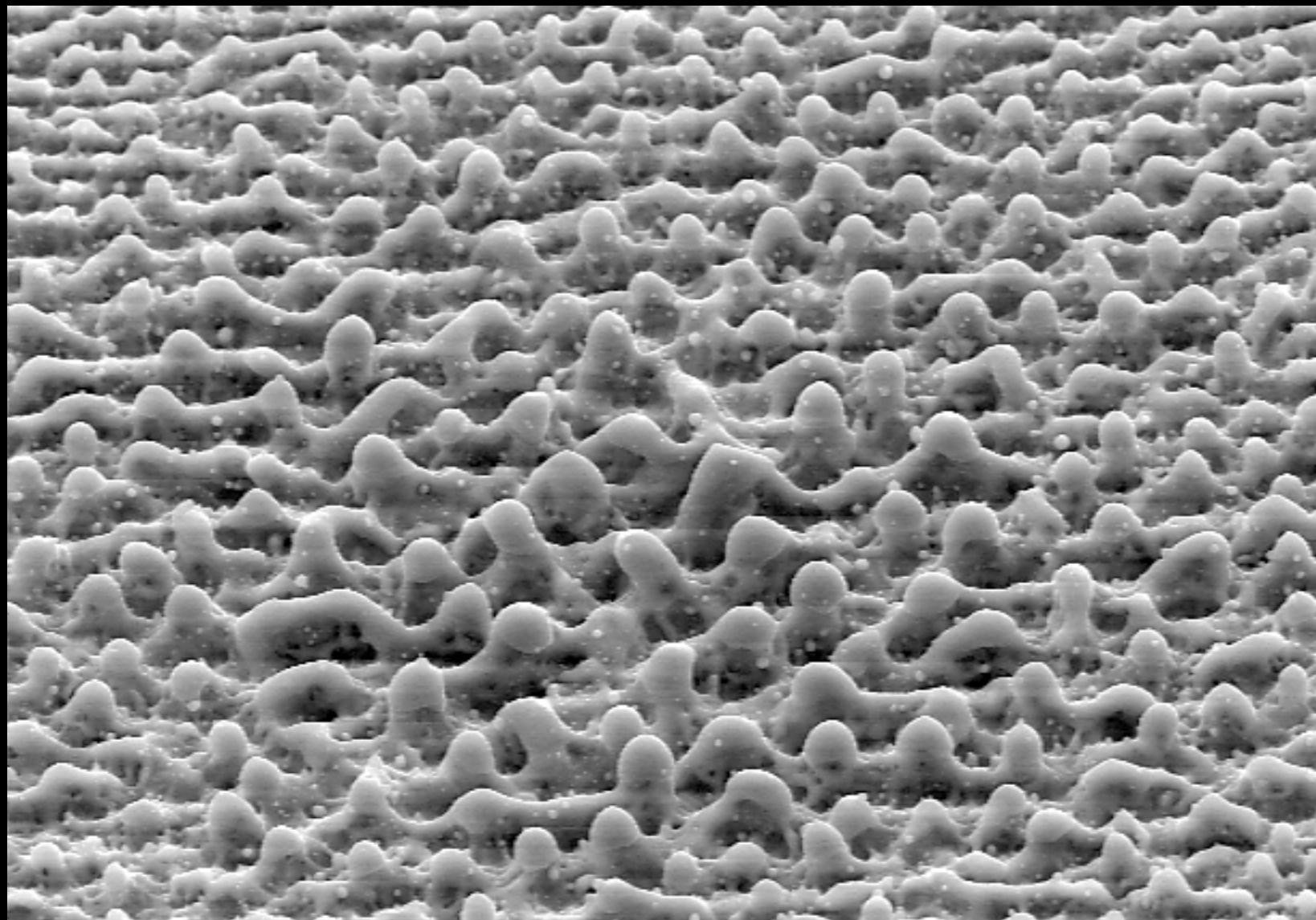
#3548

512 x 480

10kV

15mm

0010



x2000

20 μm

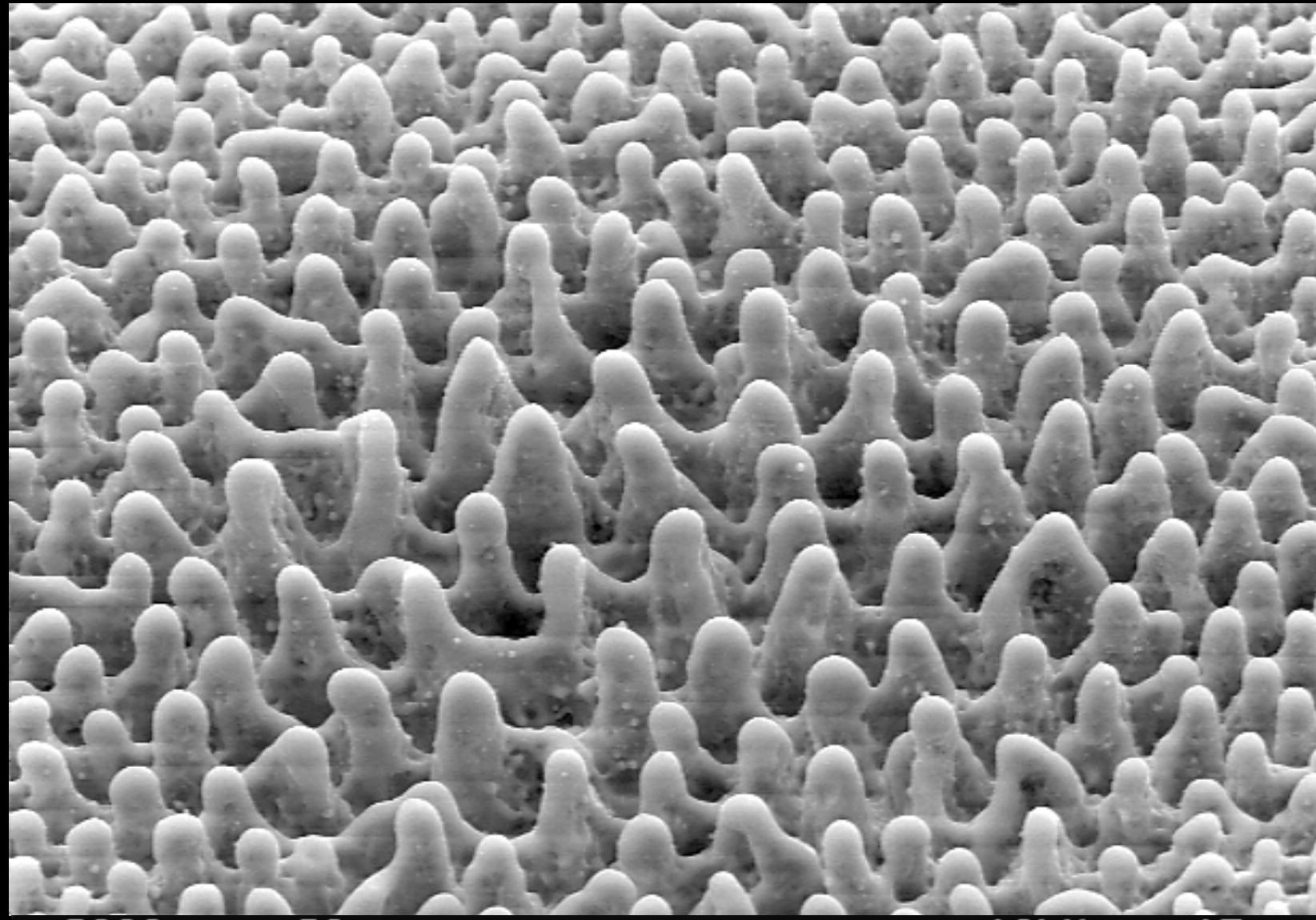
10kV

15mm

#3548

512 x 480

0025



x2000

20 μm

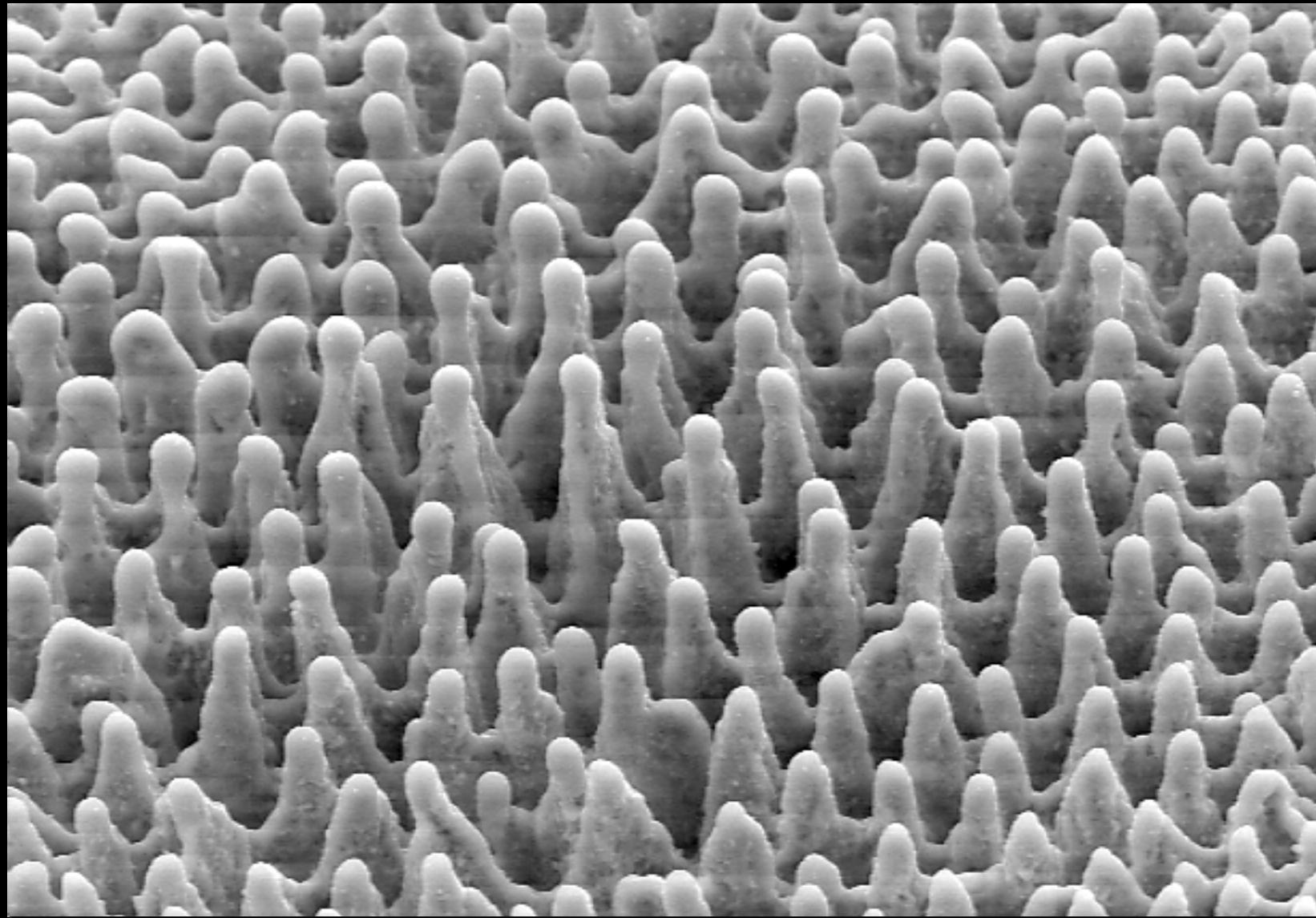
10kV

15mm

#3548

512 x 480

0050



x2000

20 μm

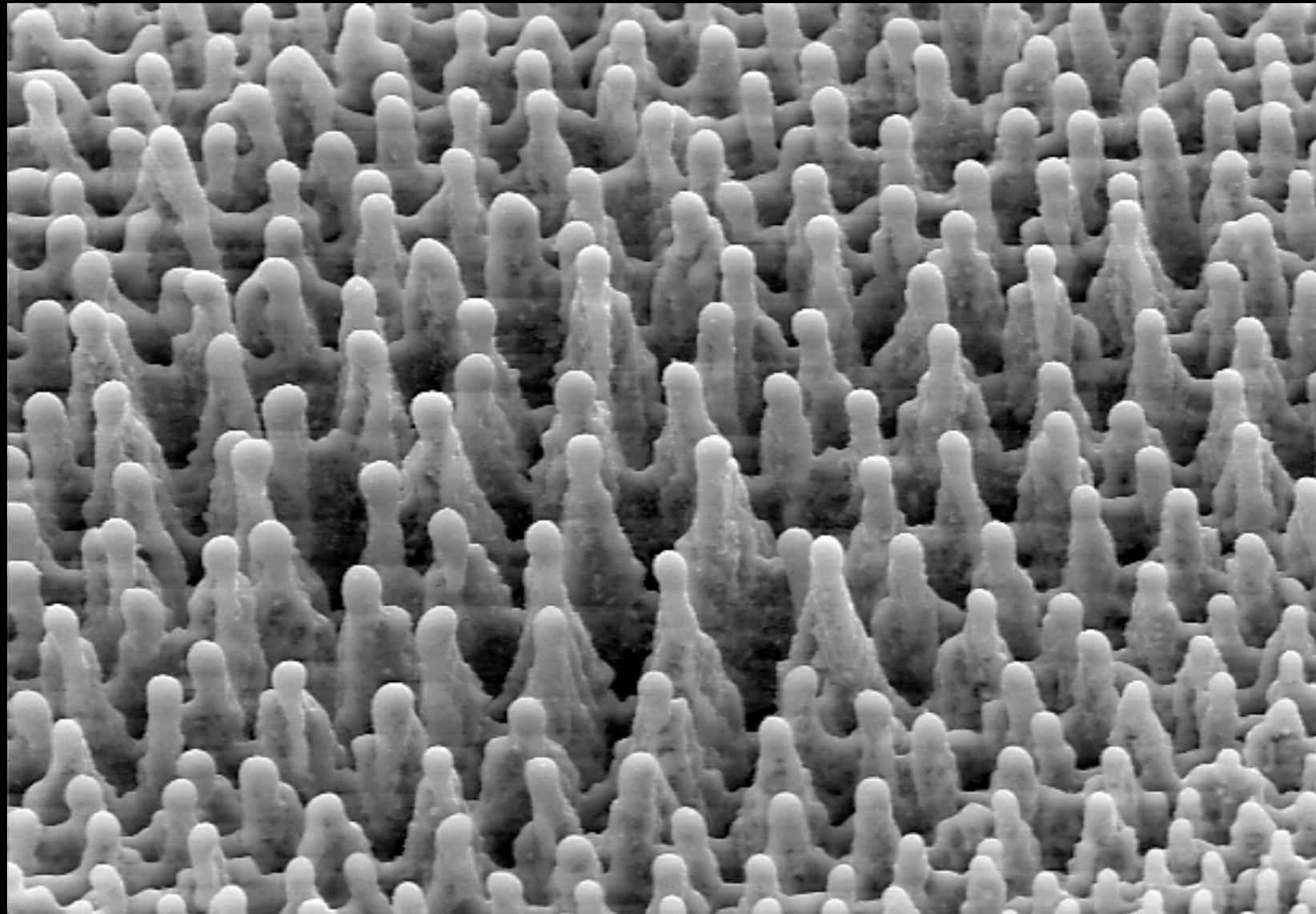
10kV

15mm

#3548

512 x 480

0075



x2000

20 μm

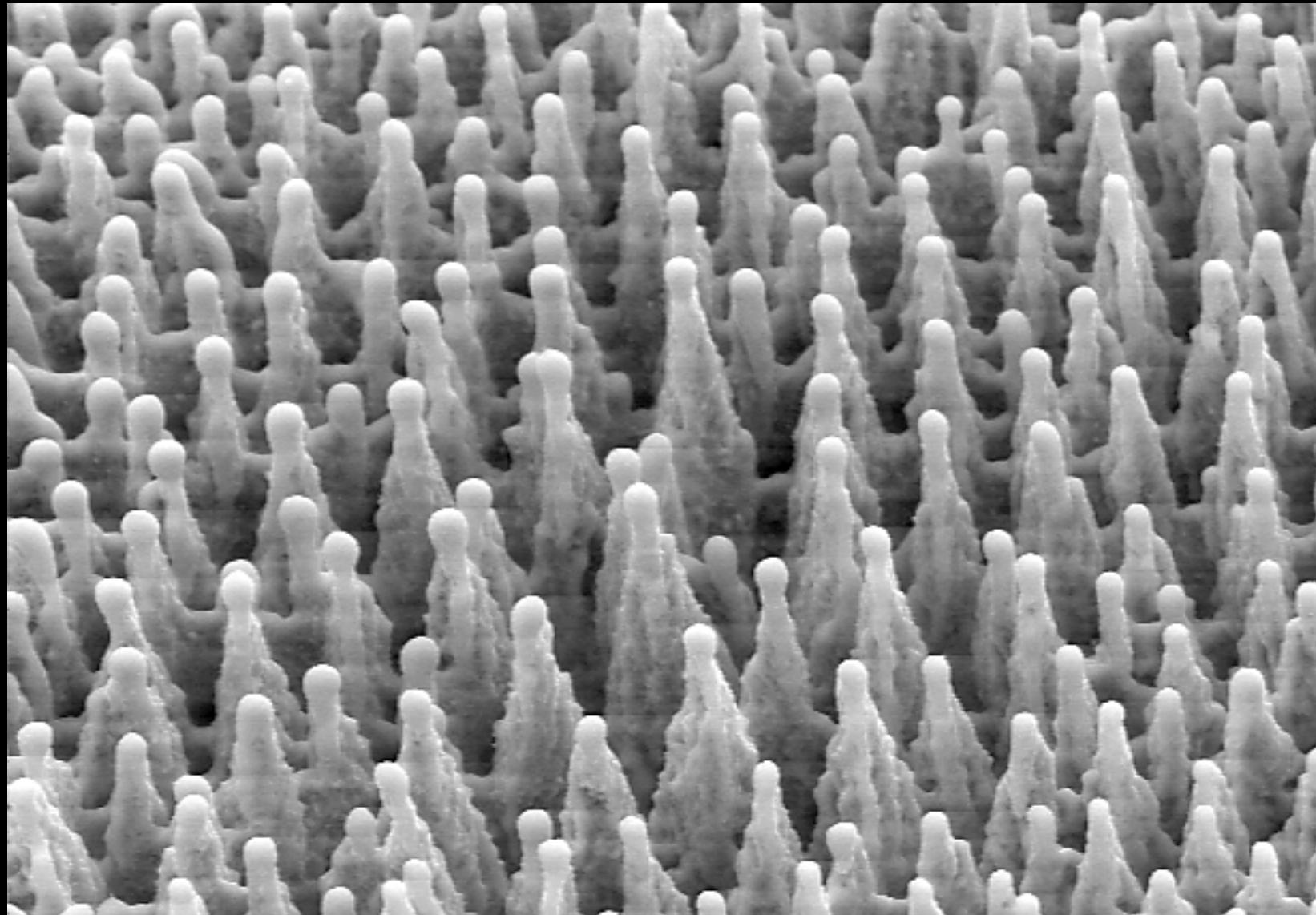
10kV

15mm

#3548

512 x 480

0125



x2000

20 μm

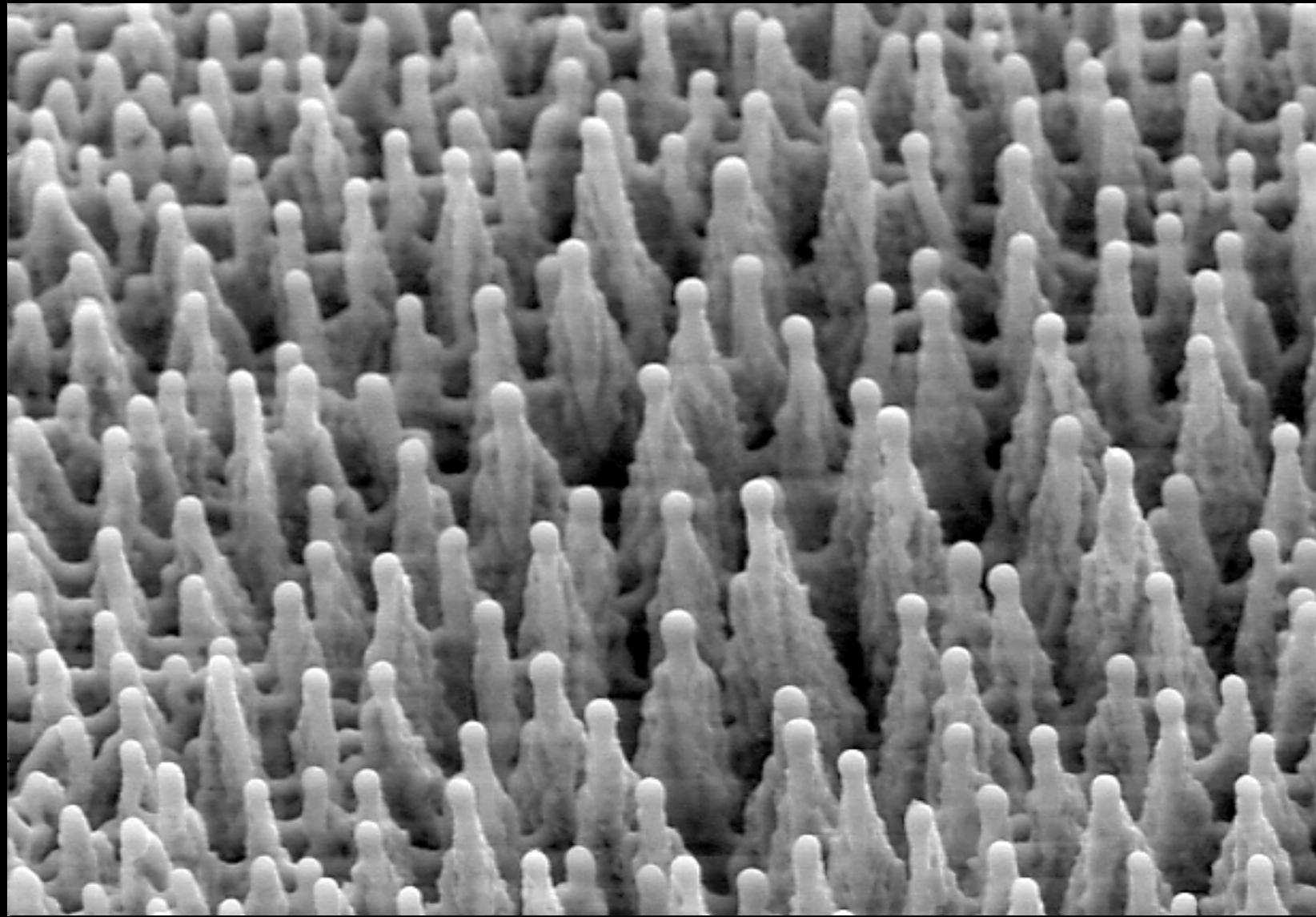
10kV

15mm

#3548

512 x 480

0250



x2000

20 μm

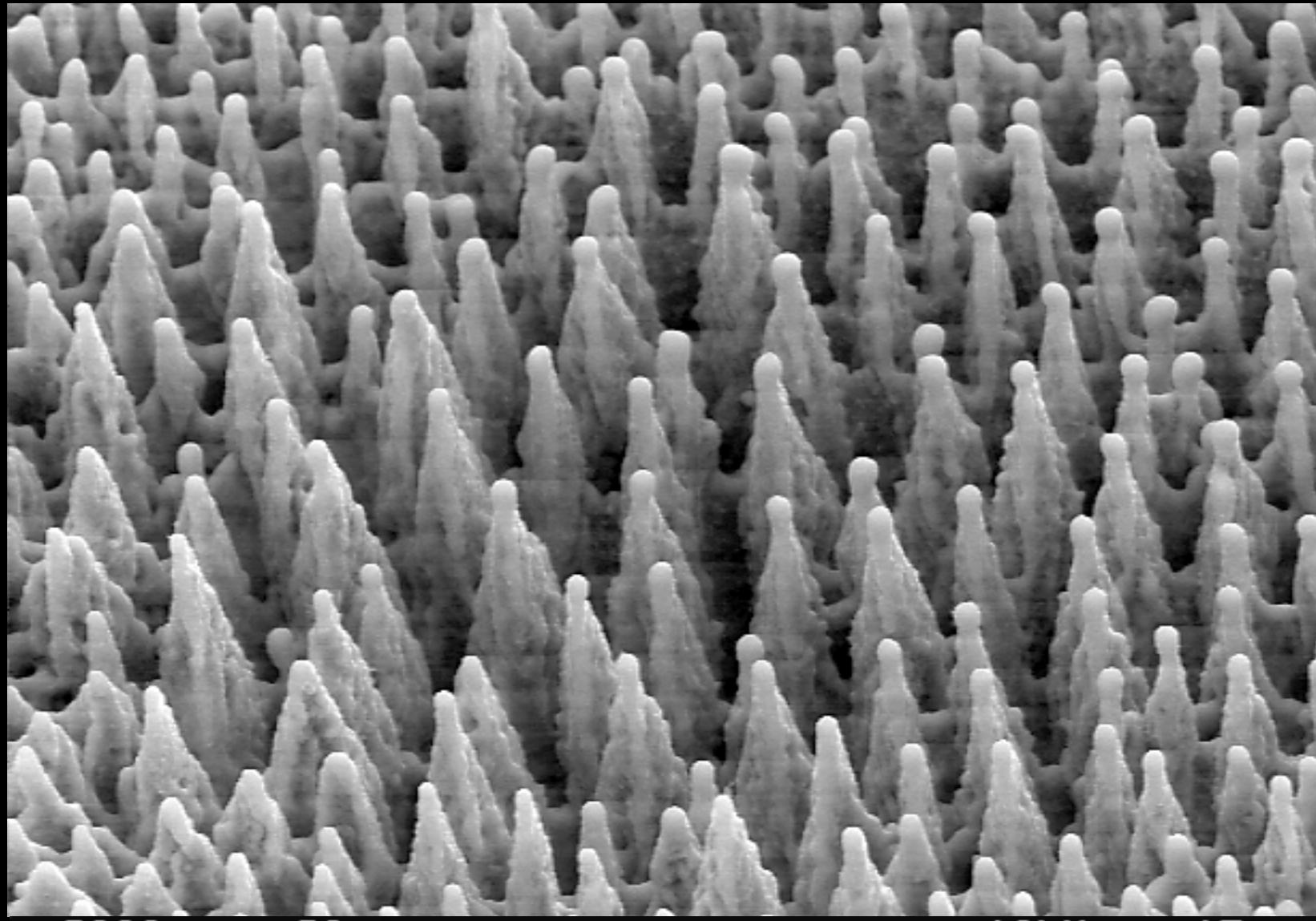
10kV

15mm

#3548

512 x 480

0350



x2000

20 μm

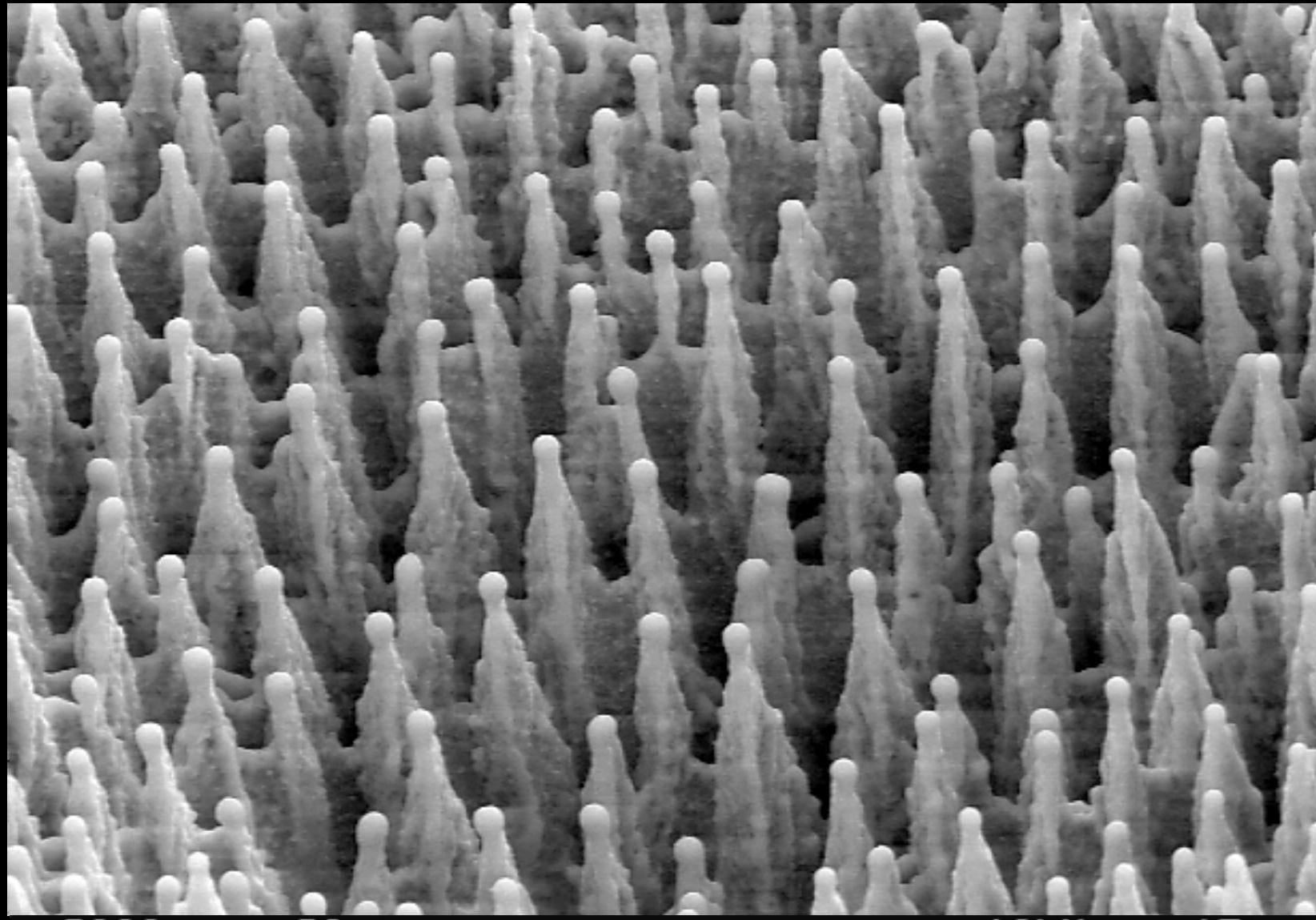
10kV

15mm

#3548

512 x 480

0450



x2000

20 μm

10kV

15mm

#3548

512 x 480

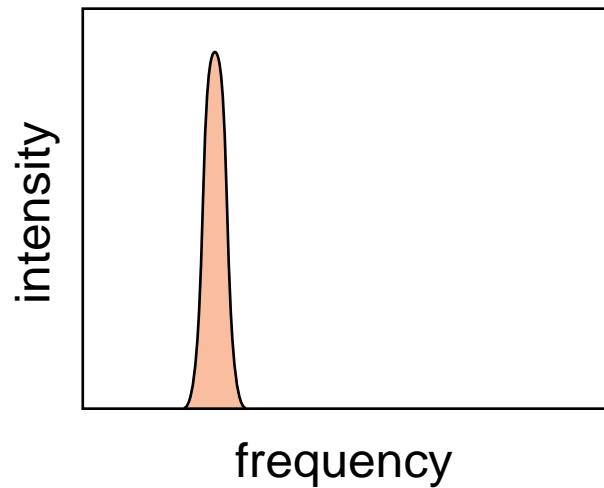
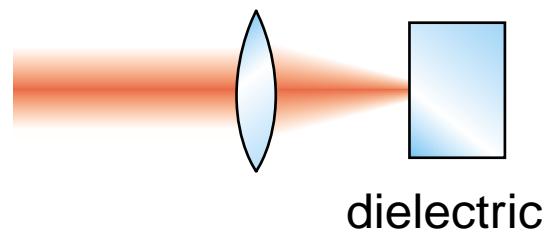
1000

Overview

- ▶ surface femtochemistry
- ▶ electronic and structural transitions
- ▶ microstructuring of materials
- ▶ nonlinear optics and propagation
- ▶ laser surgery

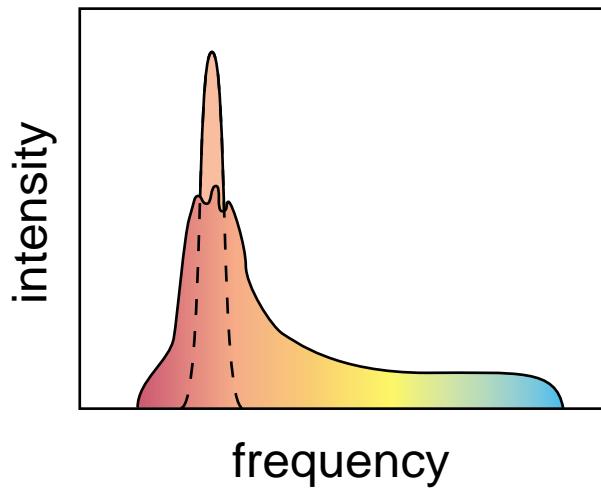
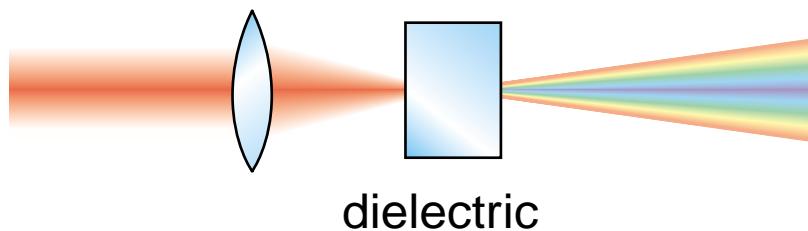
Nonlinear optics and propagation

high-power femtosecond laser pulse...



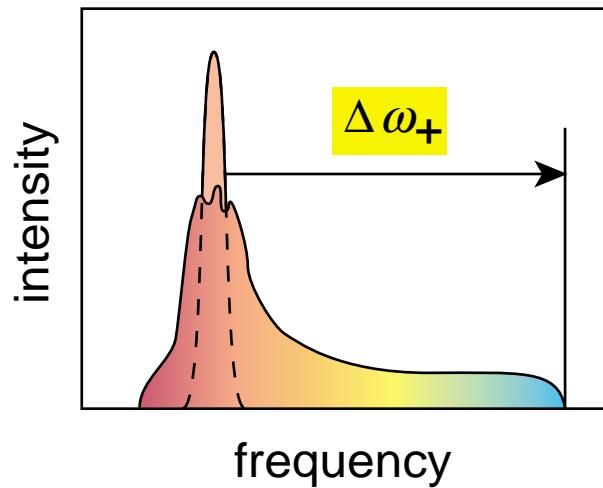
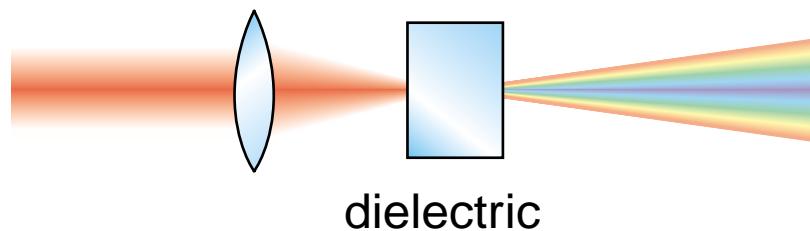
Nonlinear optics and propagation

...produces broad continuum



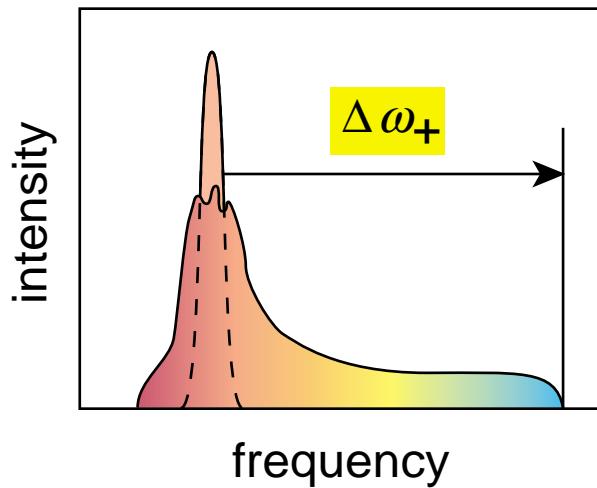
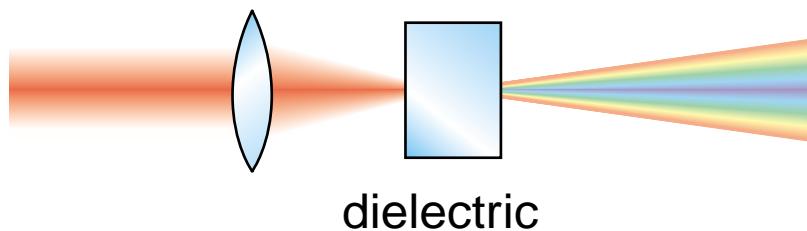
Nonlinear optics and propagation

...produces broad continuum



Nonlinear optics and propagation

How does $\Delta\omega_+$ vary with laser frequency and material?



Nonlinear optics and propagation

self-phase modulation: $n = n_0 + n_2 I$



dielectric

Nonlinear optics and propagation

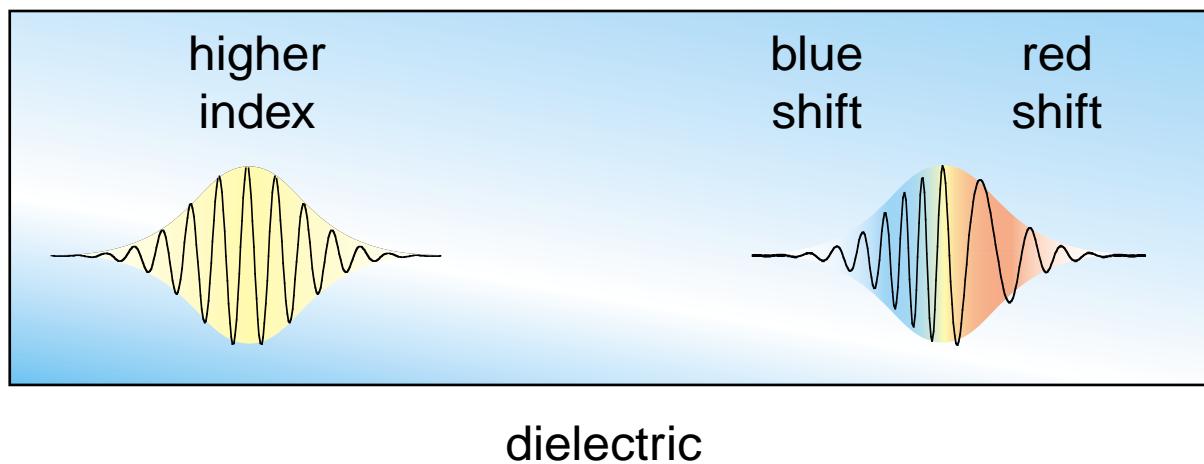
self-phase modulation: $n = n_0 + n_2 I$



dielectric

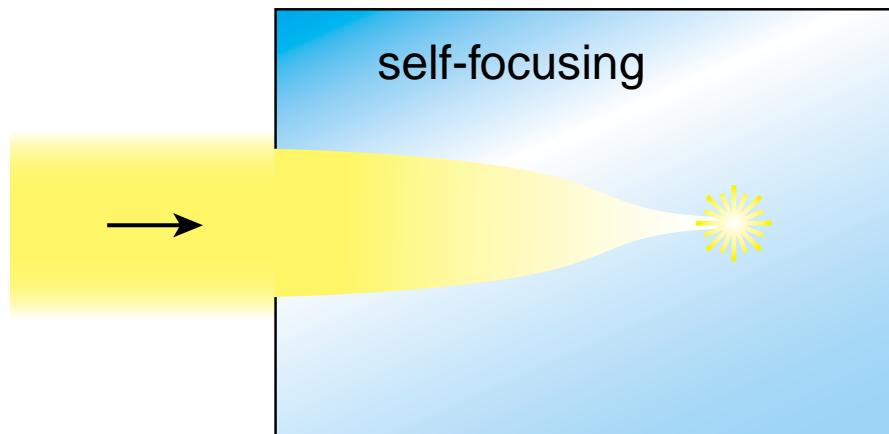
Nonlinear optics and propagation

self-phase modulation: $n = n_0 + n_2 I$



Nonlinear optics and propagation

... but what is the intensity at the focus?



Nonlinear optics and propagation

Measure $\Delta\omega_+$

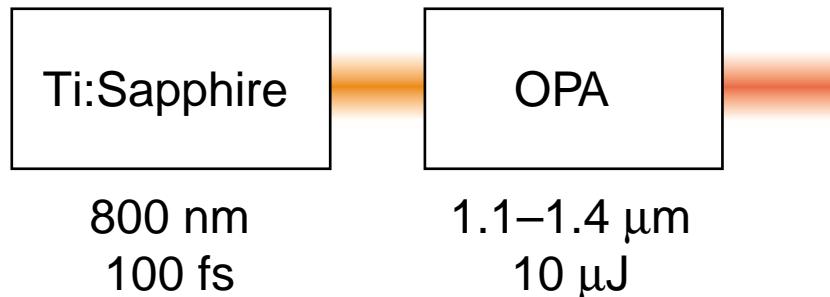
Ti:Sapphire

800 nm

100 fs

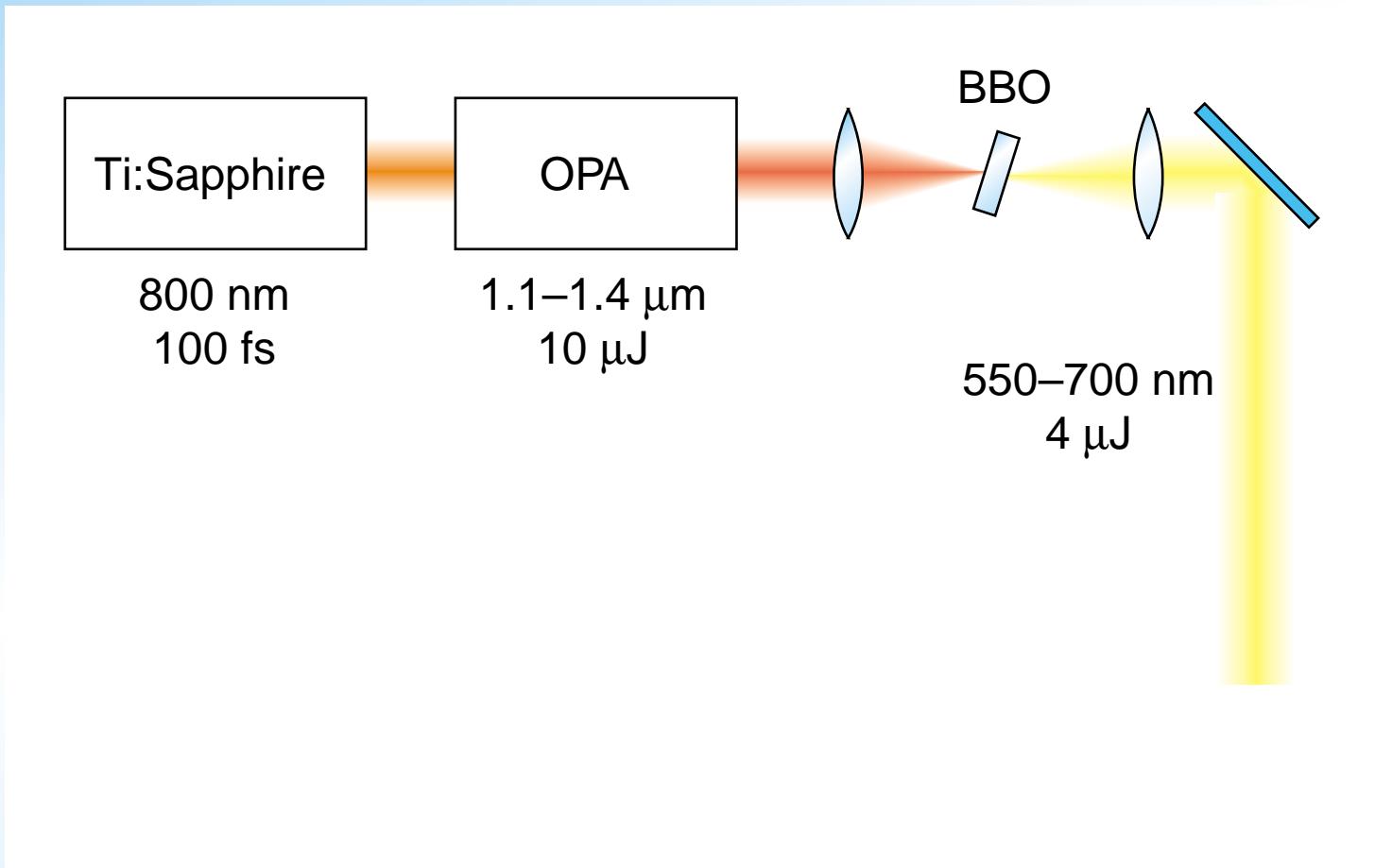
Nonlinear optics and propagation

Measure $\Delta\omega_+$



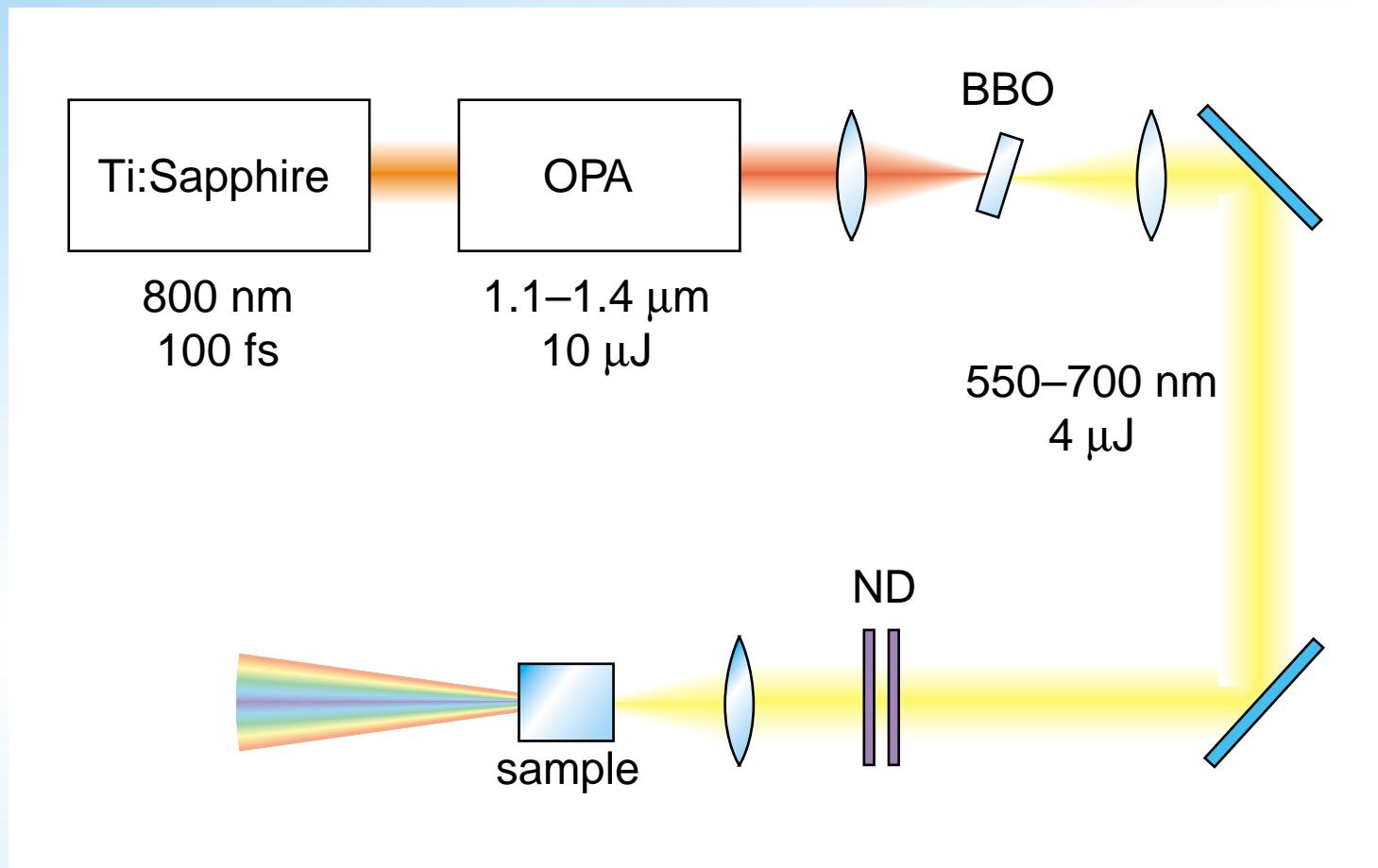
Nonlinear optics and propagation

Measure $\Delta\omega_+$



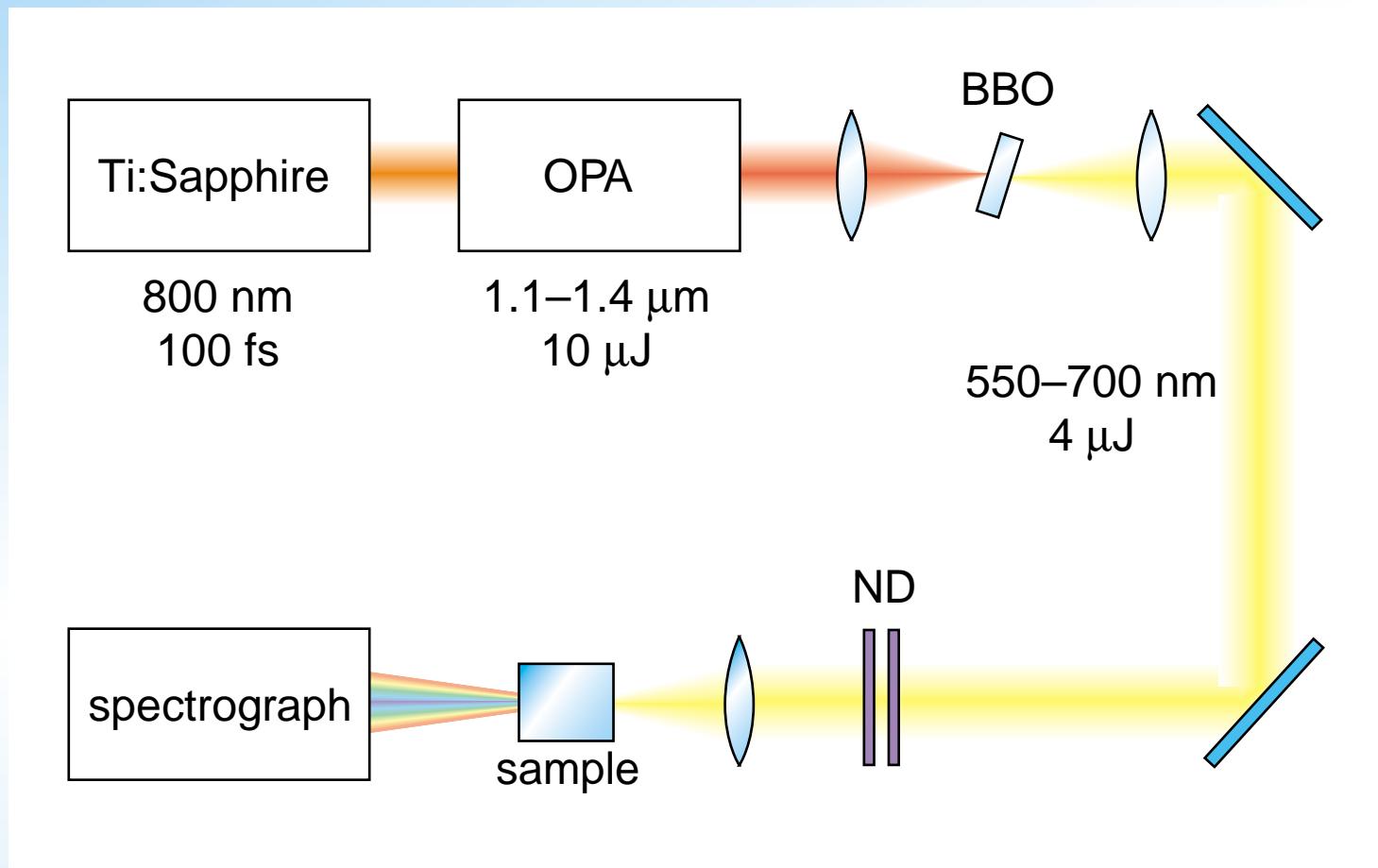
Nonlinear optics and propagation

Measure $\Delta\omega_+$

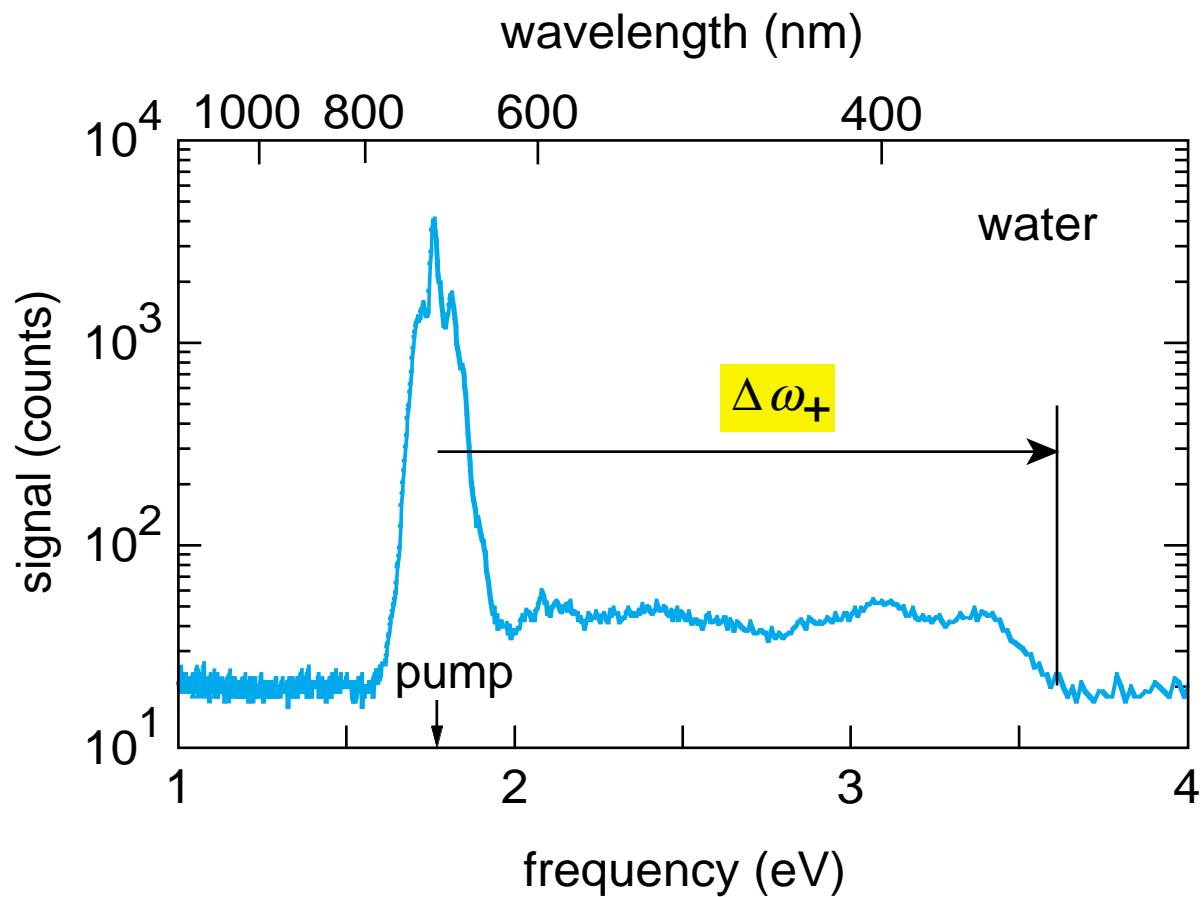


Nonlinear optics and propagation

Measure $\Delta\omega_+$



Nonlinear optics and propagation



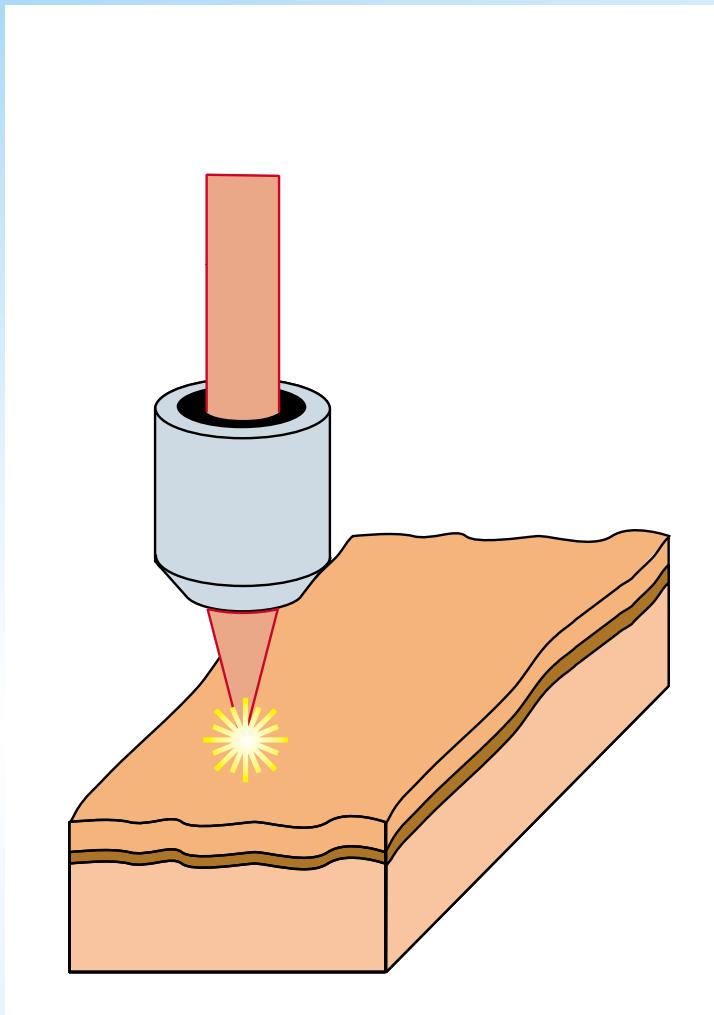
Nonlinear optics and propagation

- ▶ $\Delta\omega_+$ increases with bandgap
- ▶ $\Delta\omega_+$ decreases with pump frequency

Overview

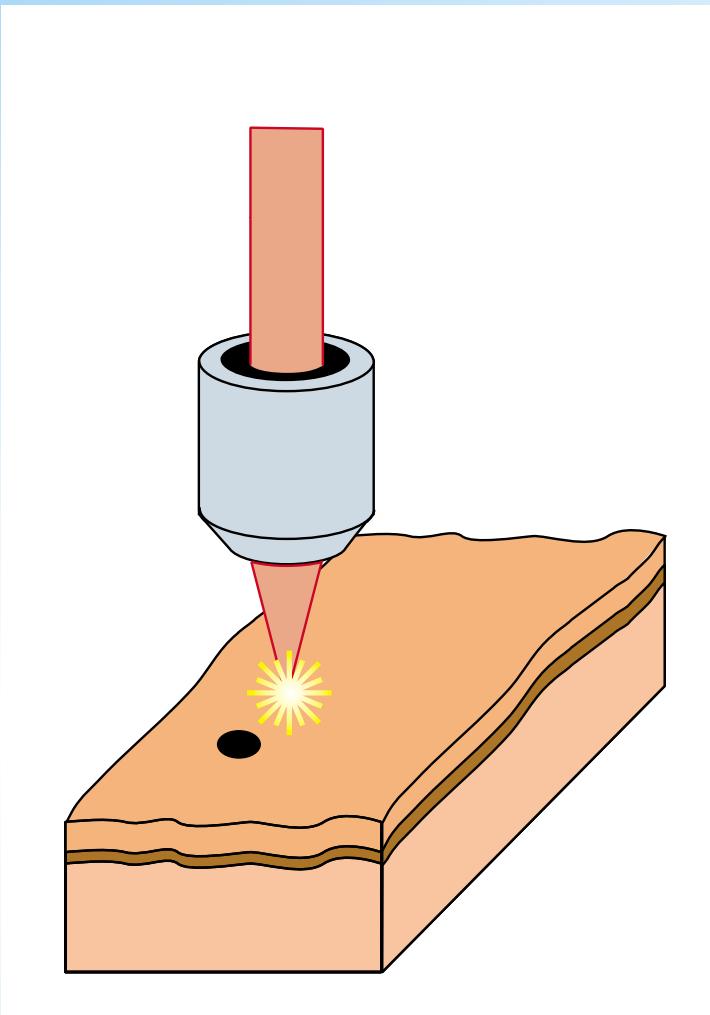
- ▶ surface femtochemistry
- ▶ electronic and structural transitions
- ▶ microstructuring of materials
- ▶ nonlinear optics and propagation
- ▶ laser surgery

Laser surgery

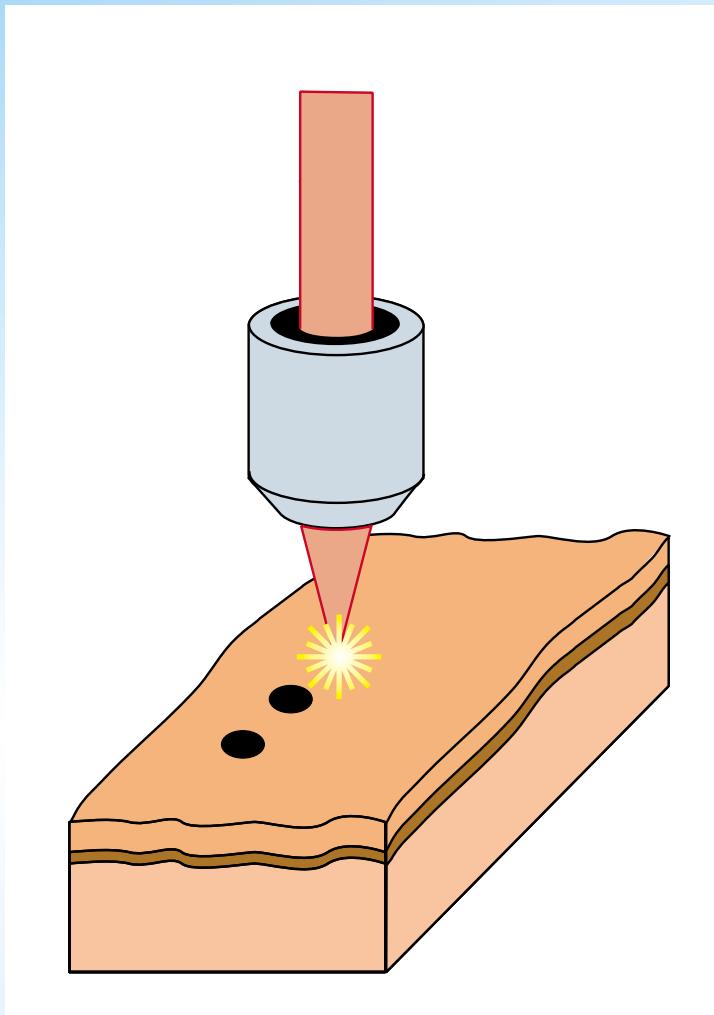


EpiDerm from MatTek Corp.
stratified skin model

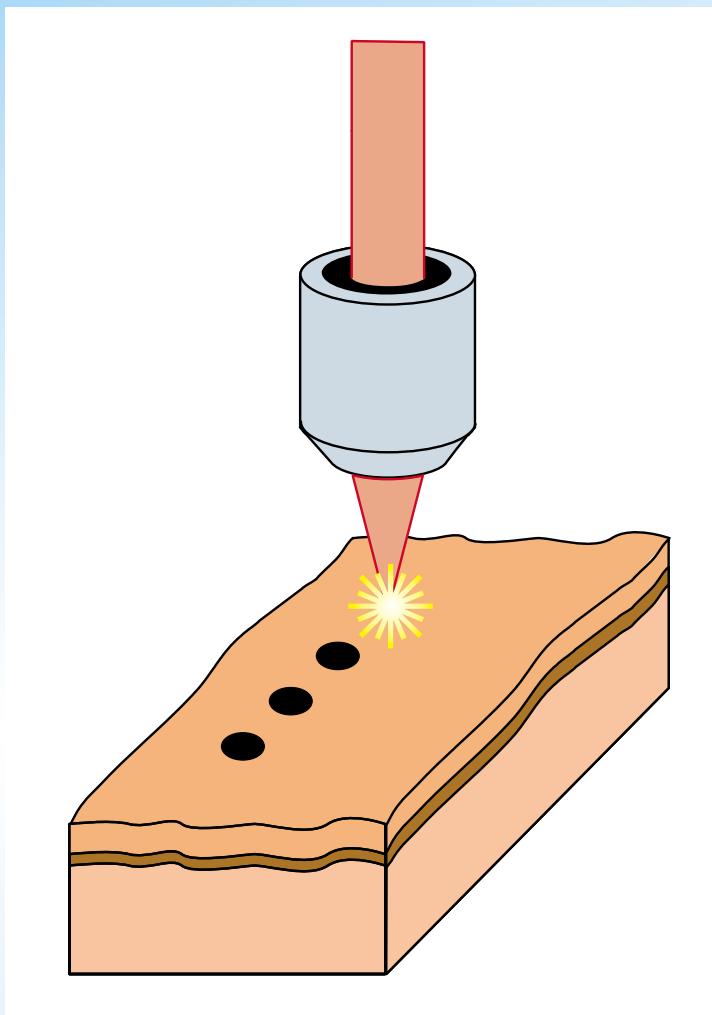
Photodisruption in epidermis



Laser surgery



Laser surgery

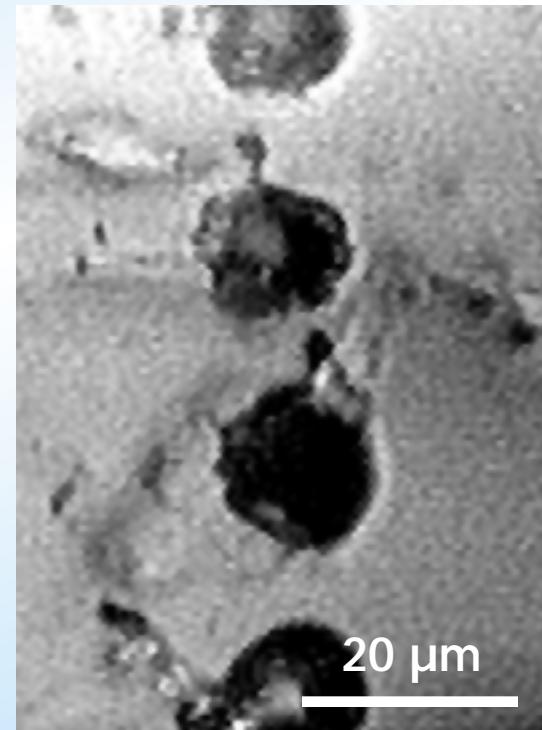


Laser surgery

200 ps, 40 μJ

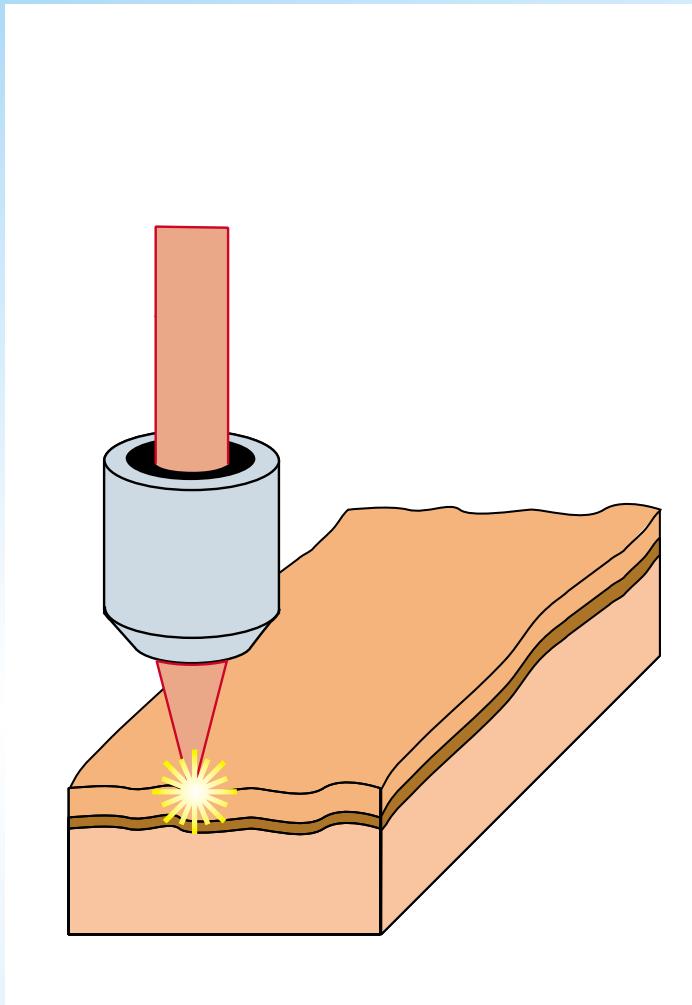


100 fs, 40 μJ

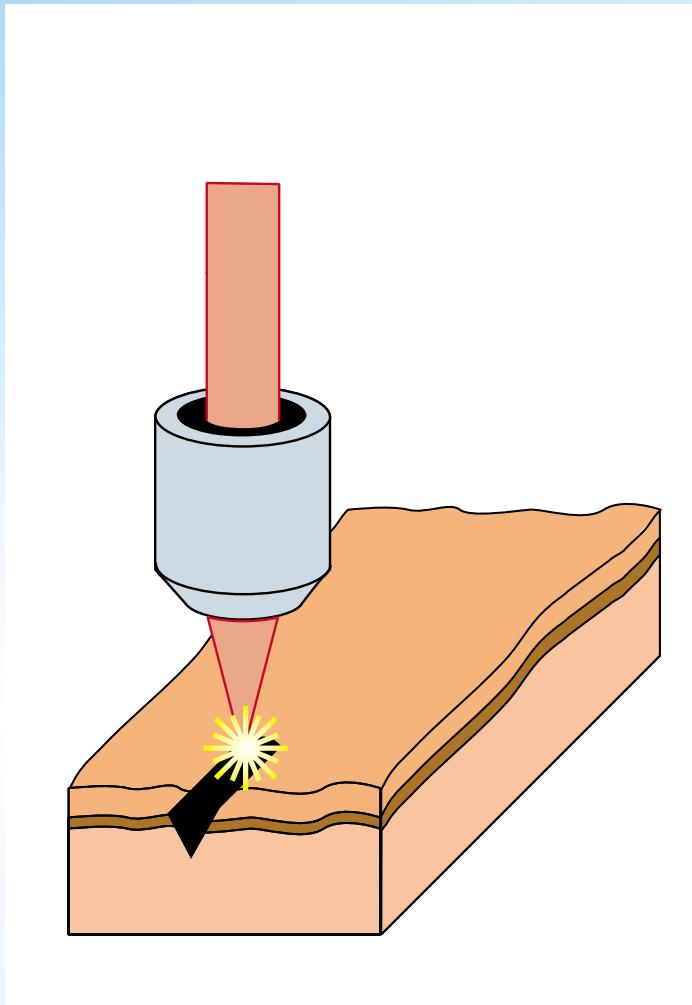


fs pulses reduce collateral damage

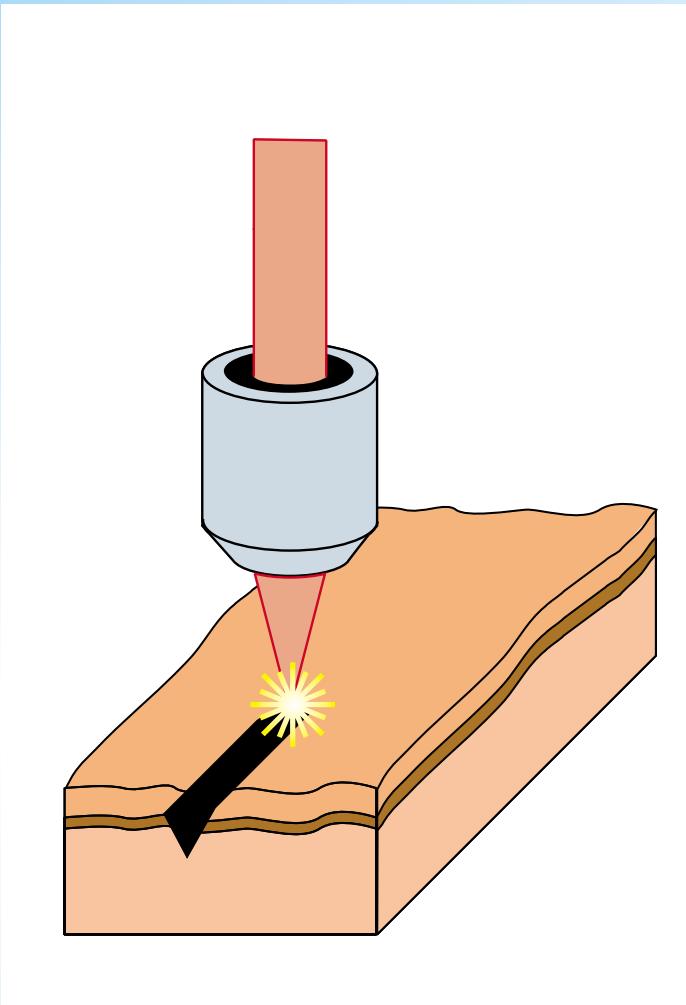
Laser surgery



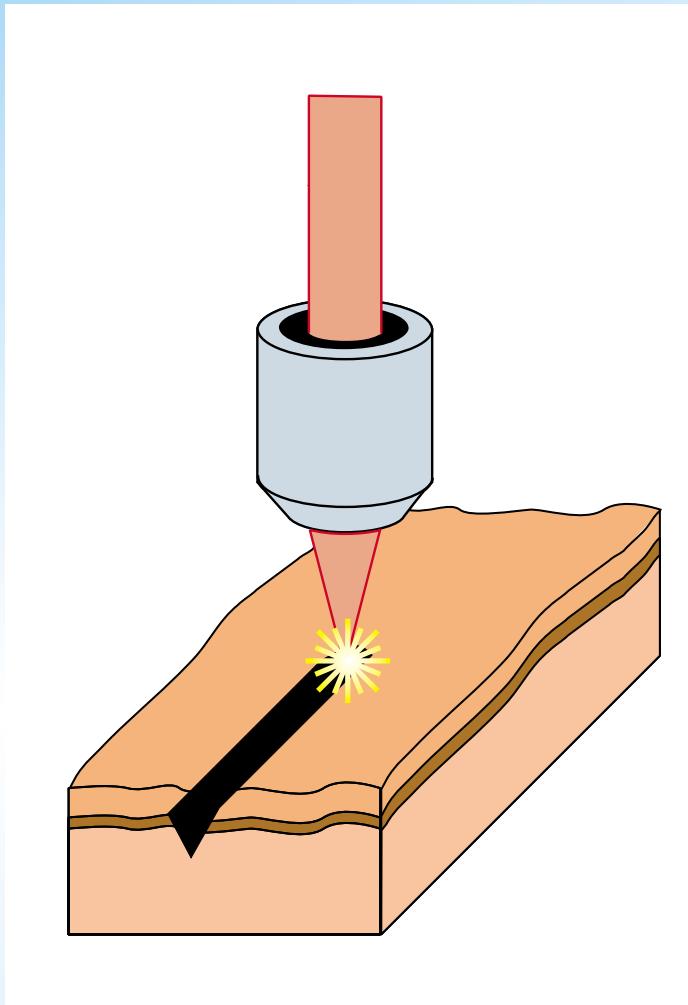
Laser surgery



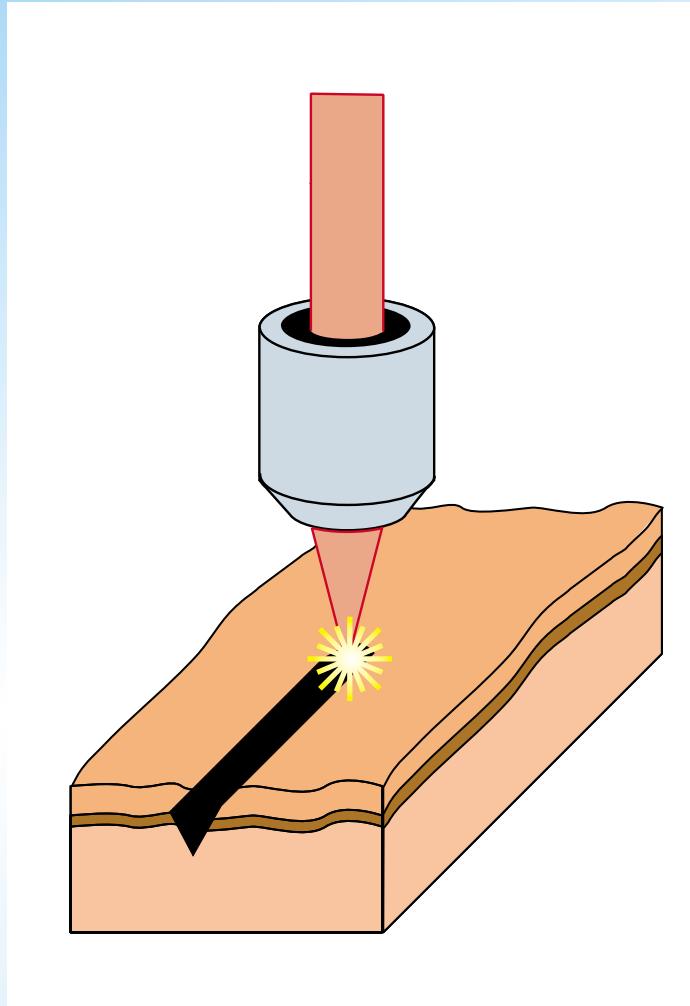
Laser surgery



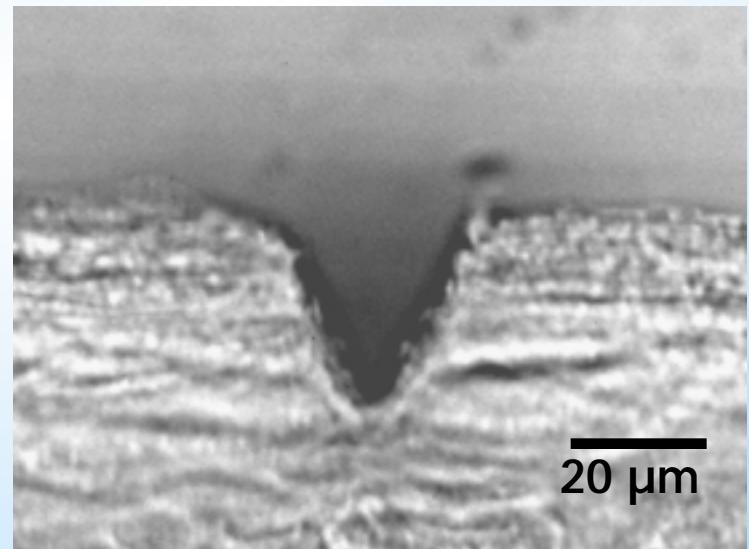
Laser surgery



Laser surgery

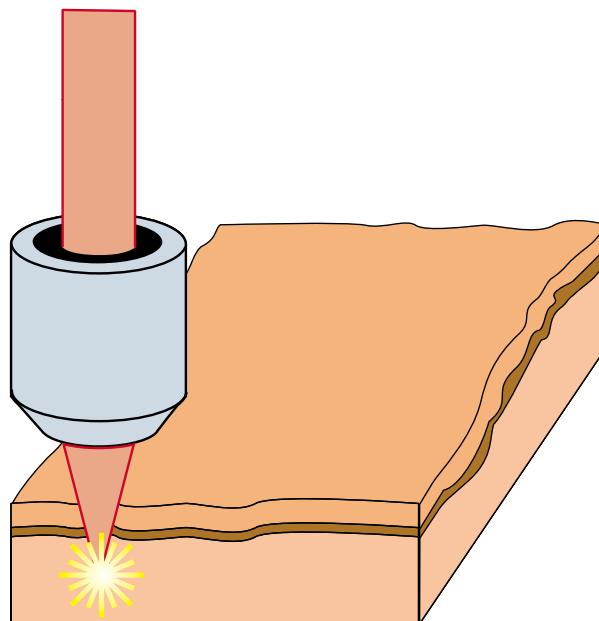


100 fs, 4 μ J



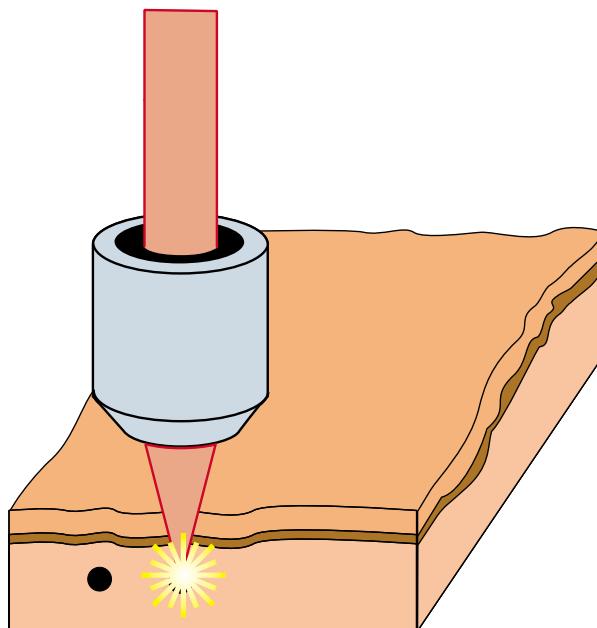
Laser surgery

focus below surface...



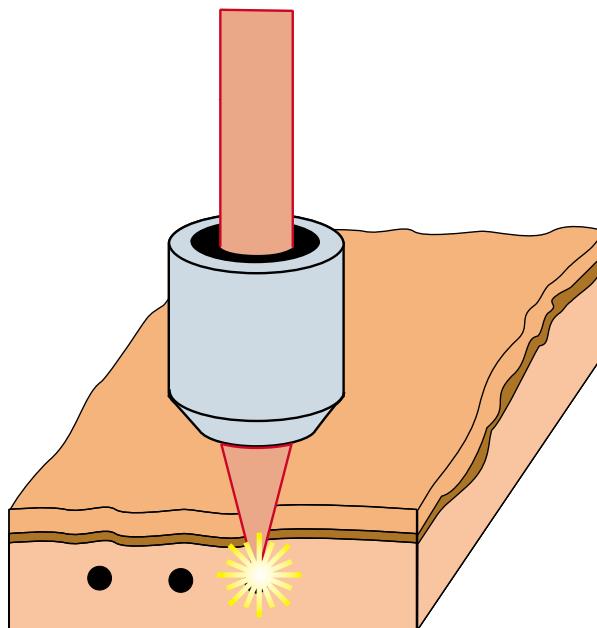
Laser surgery

... and translate beam



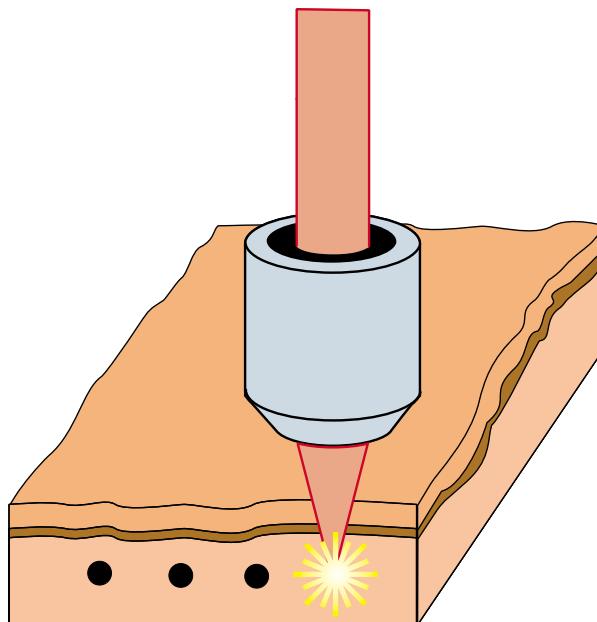
Laser surgery

... and translate beam



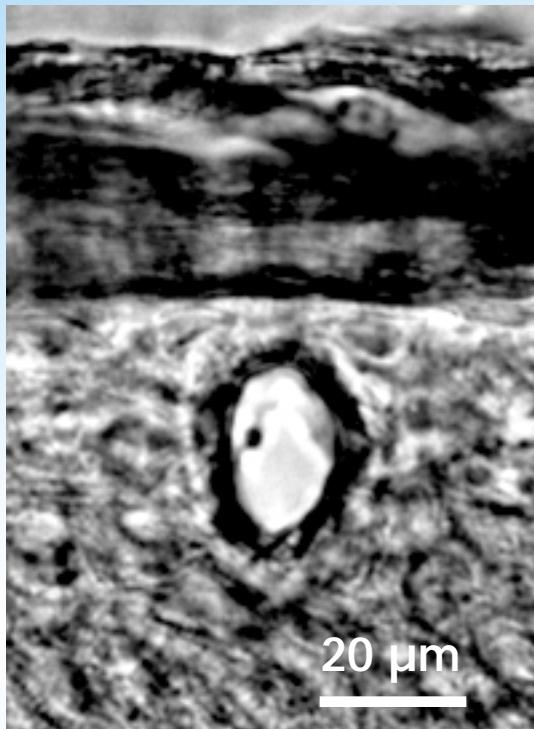
Laser surgery

... and translate beam

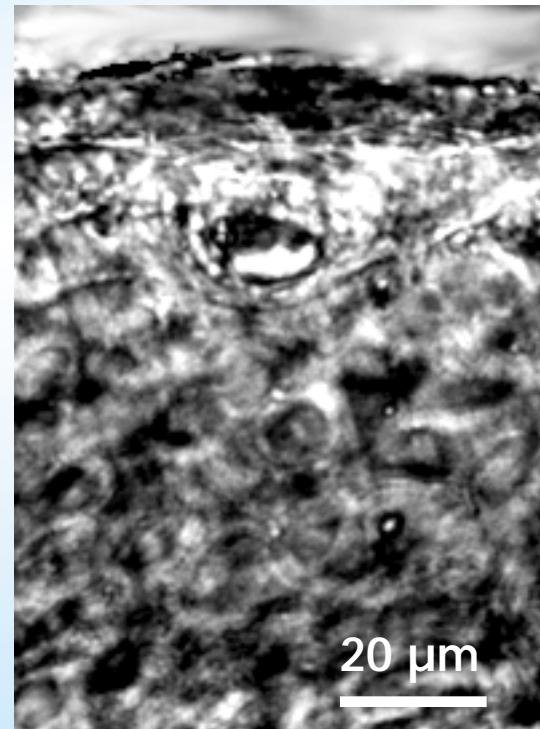


Laser surgery

200 ps, 20 μJ



100 fs, 20 μJ



undamaged surface

Summary

- ▶ diversified use of femtosecond lasers

Summary

- ▶ diversified use of femtosecond lasers
- ▶ unprecedented view into dynamics

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For a copy of this presentation and
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<http://mazur-www.harvard.edu>