

Femtosecond pulses in transparent materials 1: Nonlinear ionization and white-light generation

**Chris B. Schaffer
Jonathan B. Ashcom
André Brodeur
Eric Mazur**

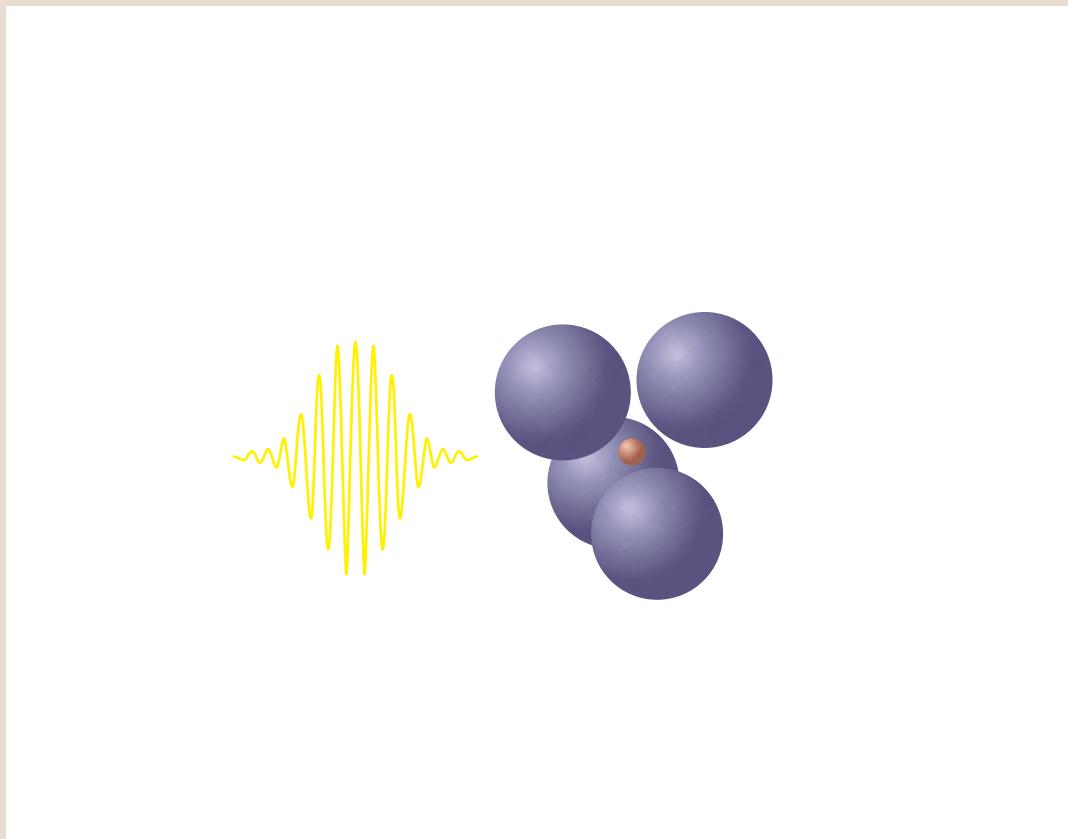


MRS Spring Meeting
April, 2001

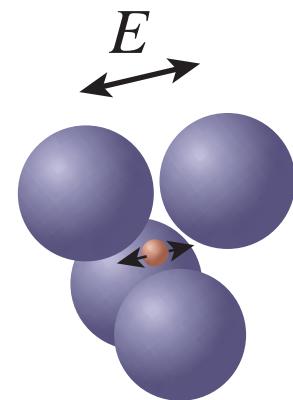
Harvard University
Department of Physics

INTRODUCTION

light-matter interactions

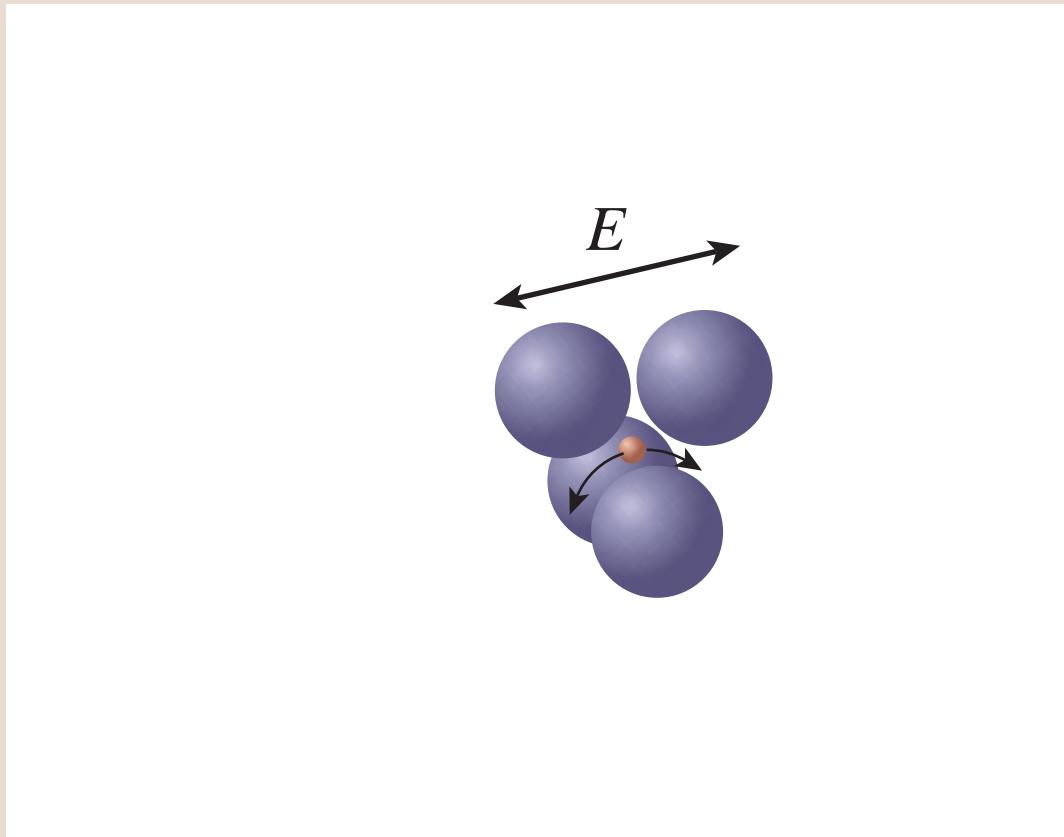


linear response



"stiffness" determines index of refraction

nonlinear response



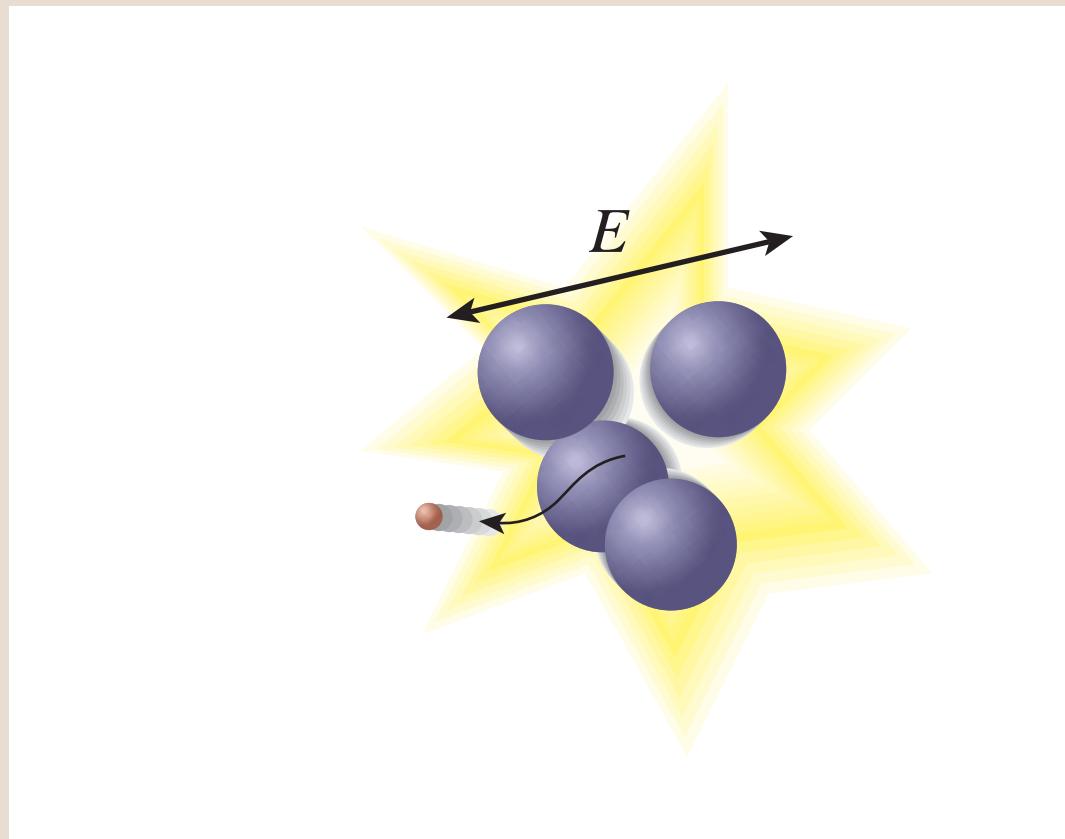
second harmonic generation

INTRODUCTION

"extremely" nonlinear response



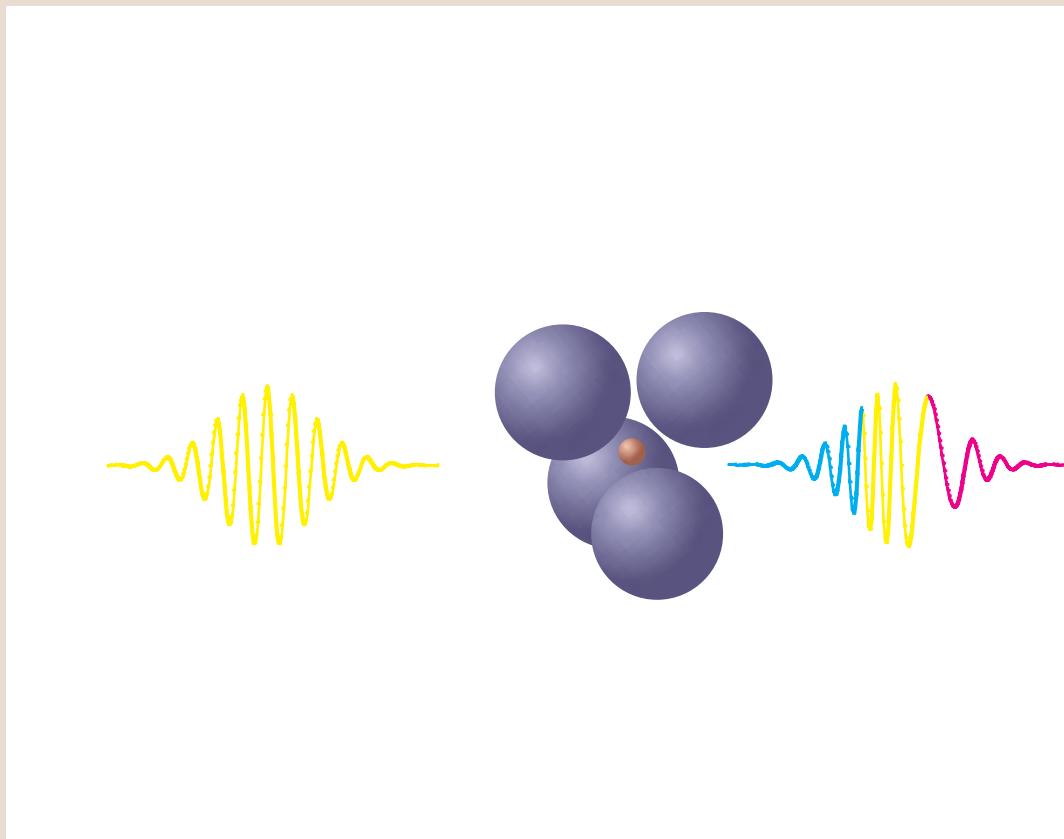
"extremely" nonlinear response



laser pulse alters the material

INTRODUCTION

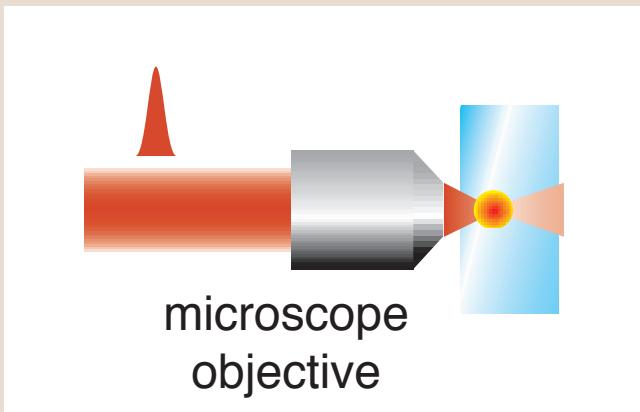
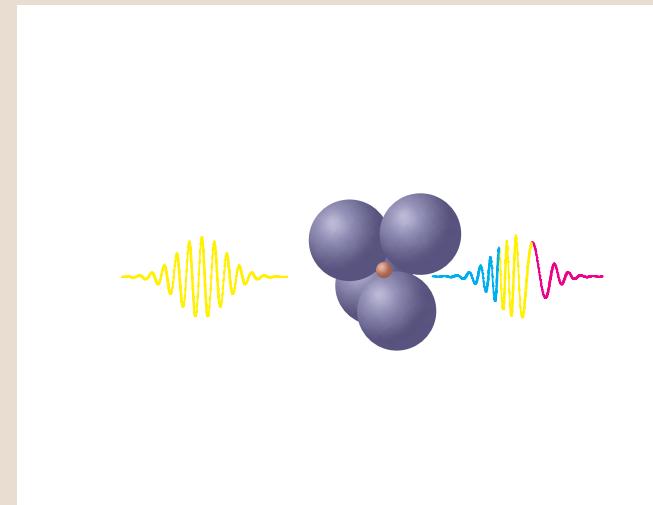
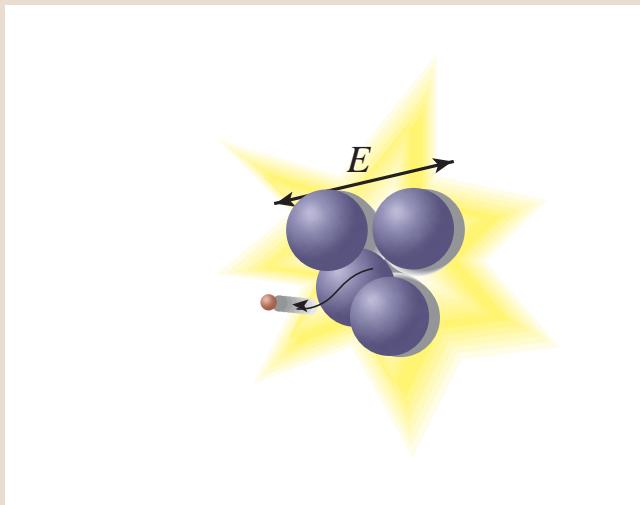
"extremely" nonlinear response



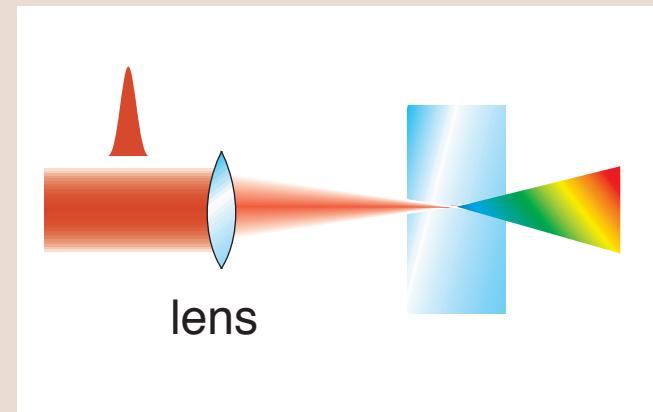
material changes the laser light

INTRODUCTION

"extremely" nonlinear response



tight focusing

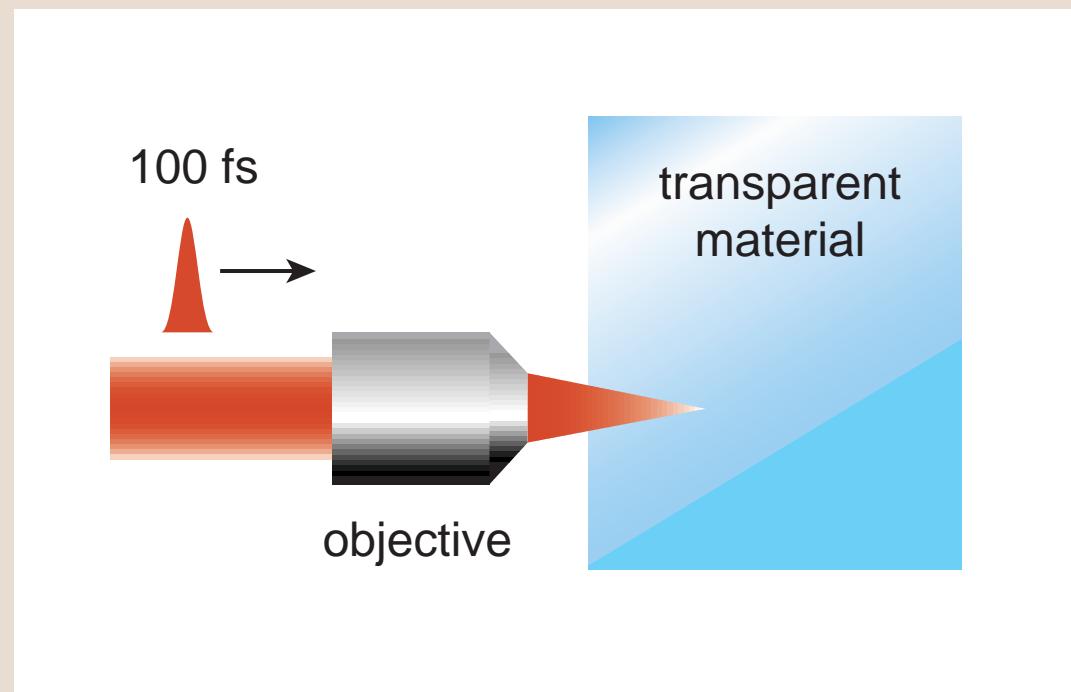


loose focusing

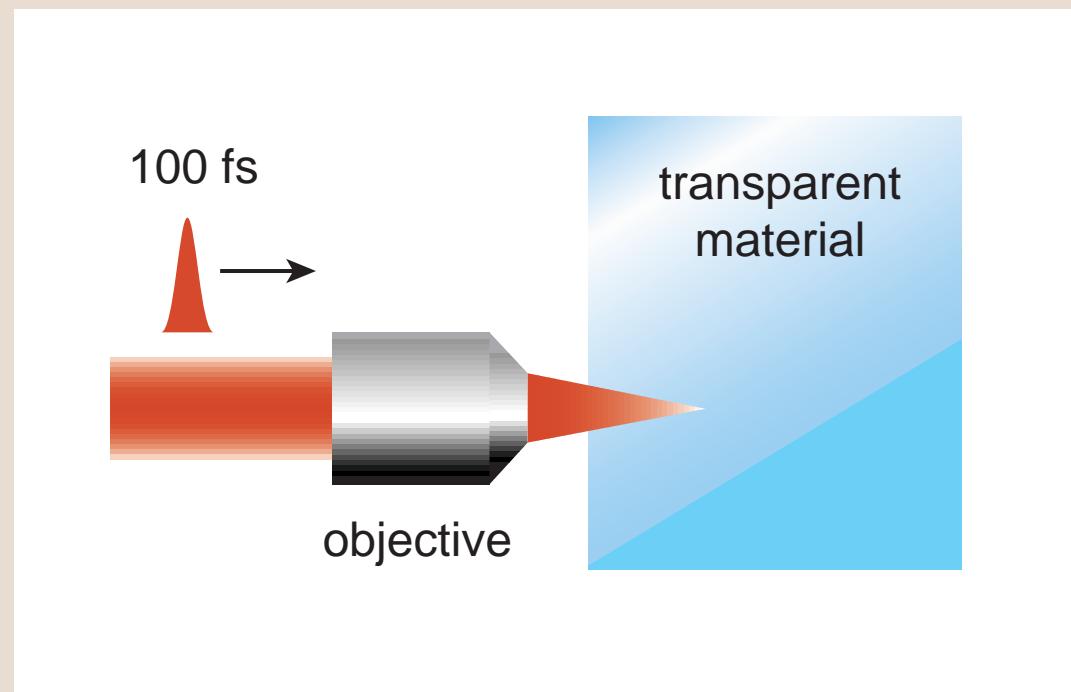
INTRODUCTION

tight focusing

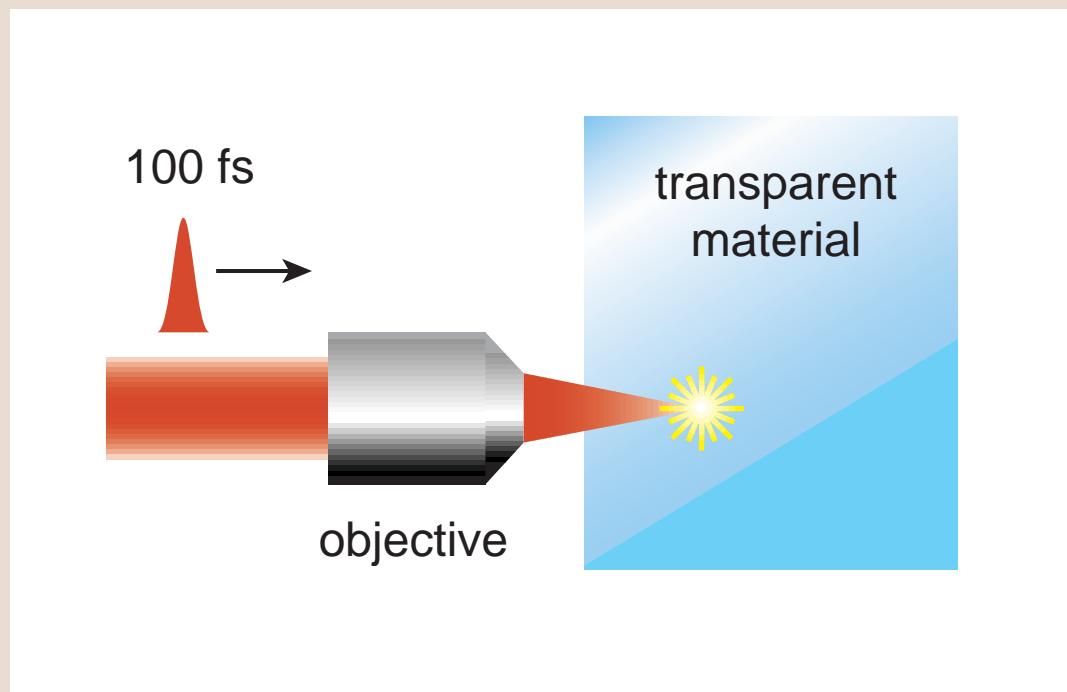
focus laser beam inside material



high intensity at focus

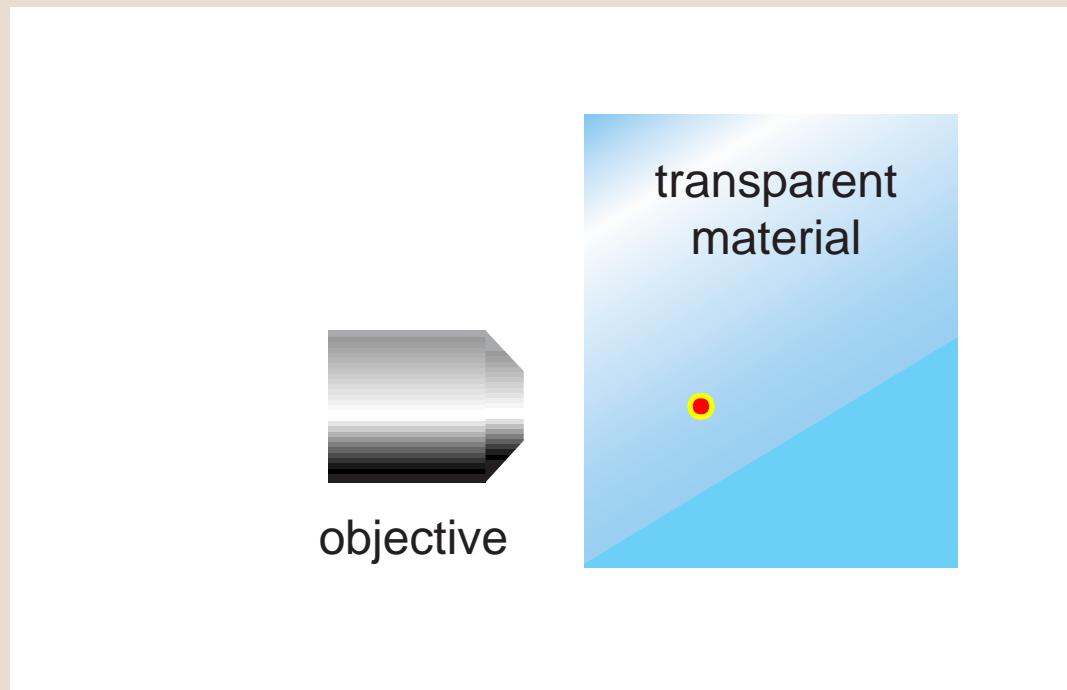


causes nonlinear ionization



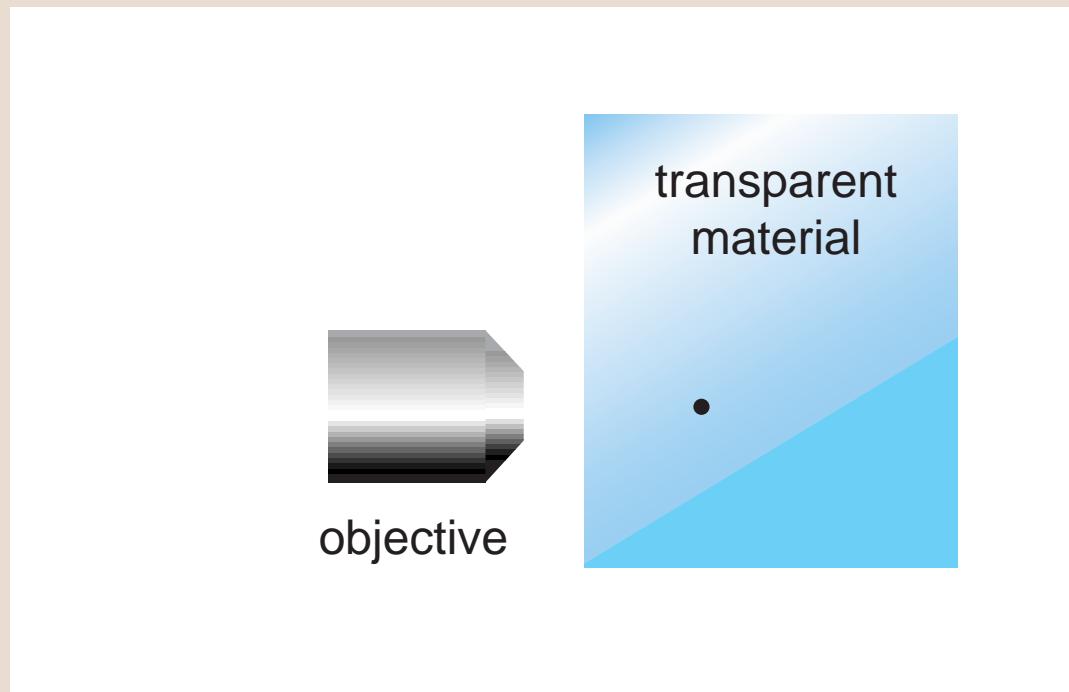
INTRODUCTION

energy is deposited in the focal volume



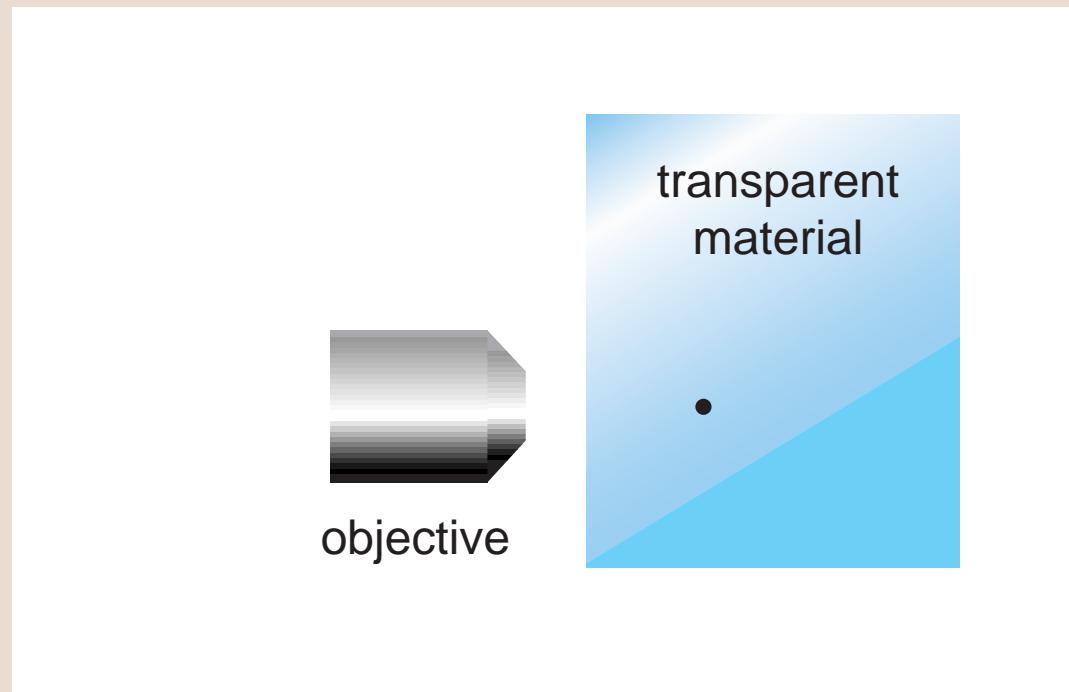
INTRODUCTION

producing microscopic bulk damage



INTRODUCTION

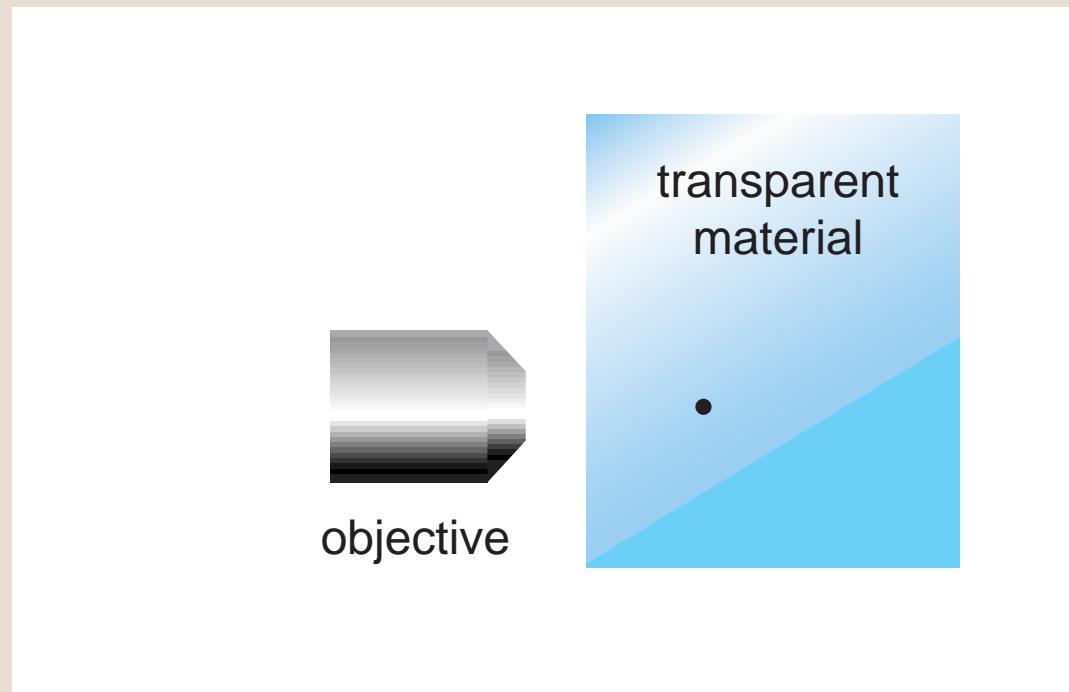
producing microscopic bulk damage



with only tens of nanojoules!

INTRODUCTION

producing microscopic **bulk** damage

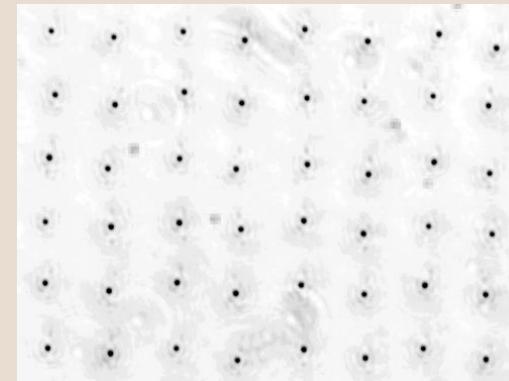


with only tens of **nanojoules**!

why bulk?

why bulk?

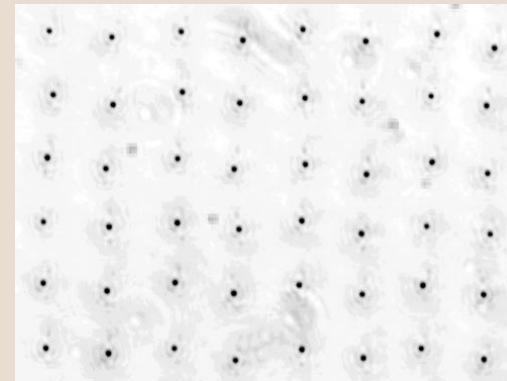
three-dimensional micromachining



INTRODUCTION

why bulk?

three-dimensional micromachining

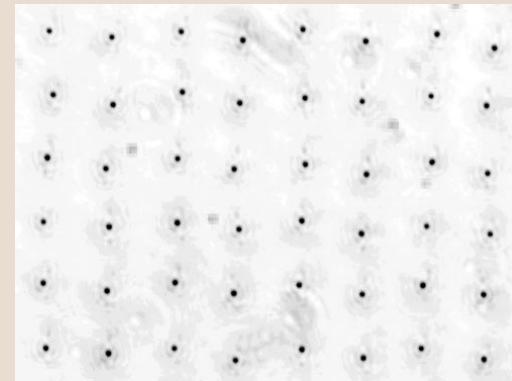


why nanojoules?

INTRODUCTION

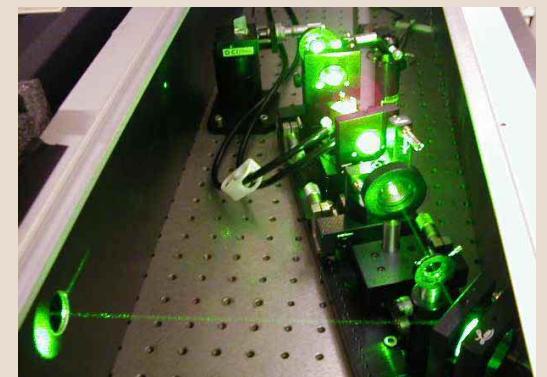
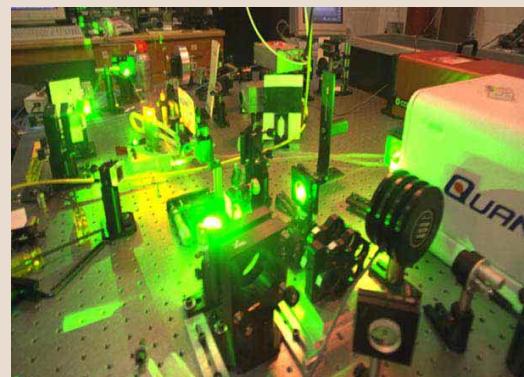
why **bulk?**

three-dimensional micromachining

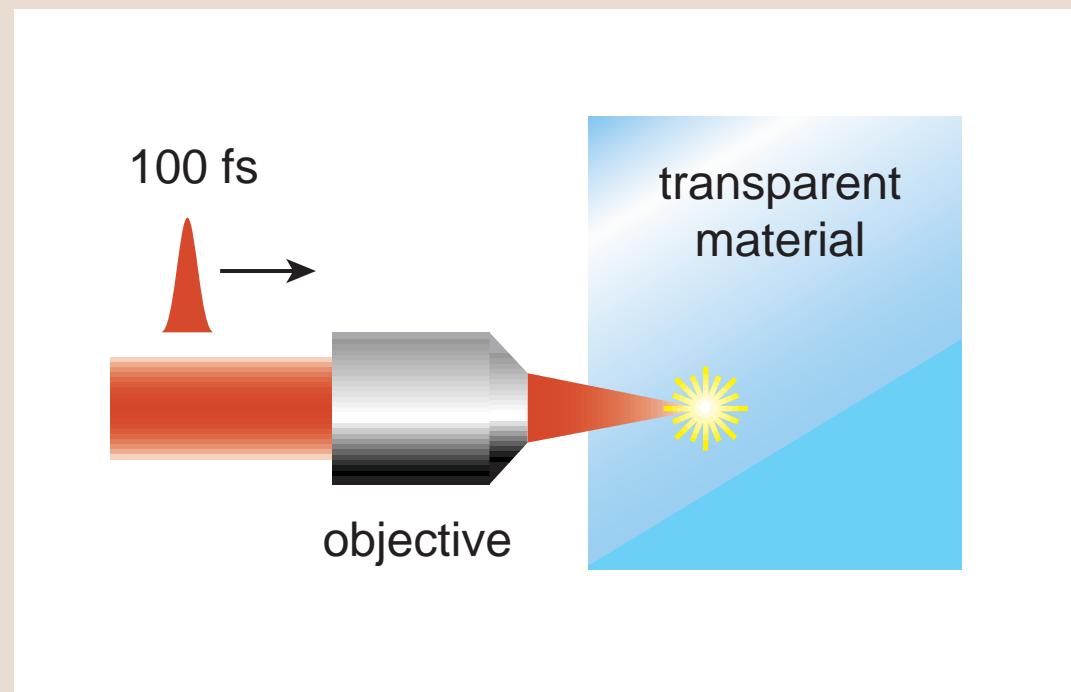


why **nanojoules?**

non-amplified micromachining



what is the nature of the nonlinear ionization?

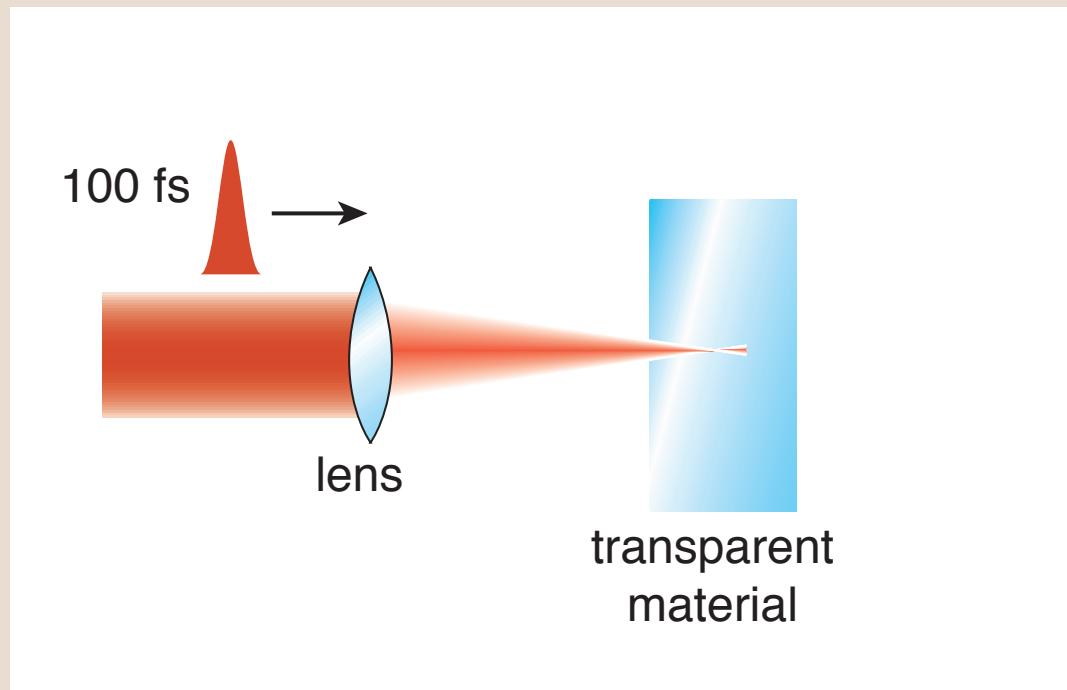


INTRODUCTION

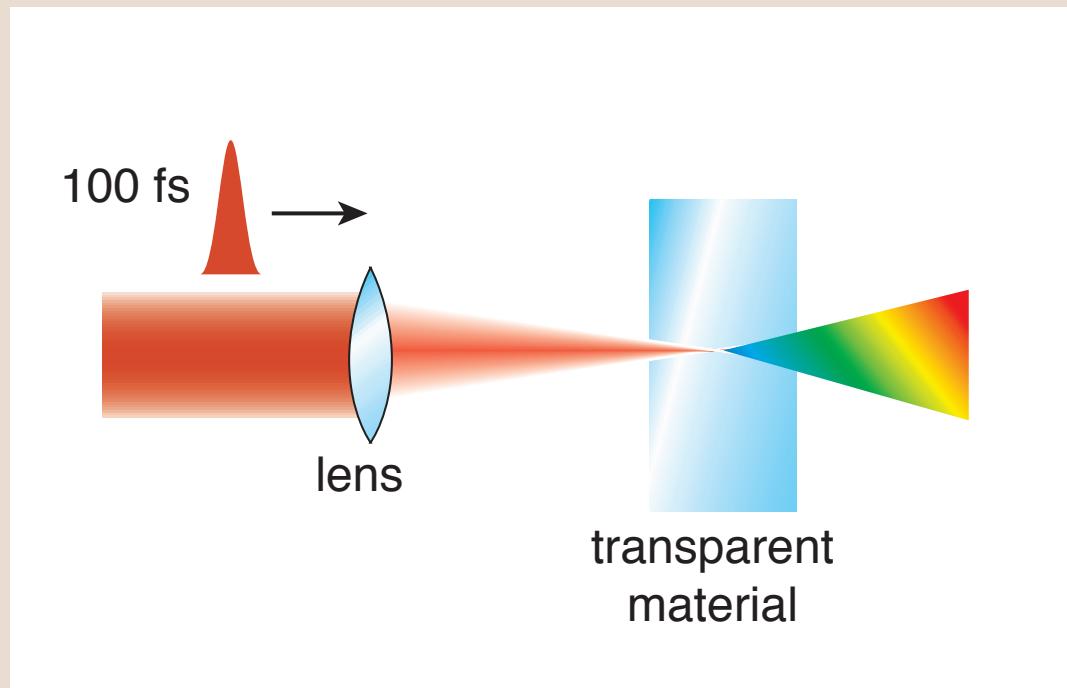
slow focusing

INTRODUCTION

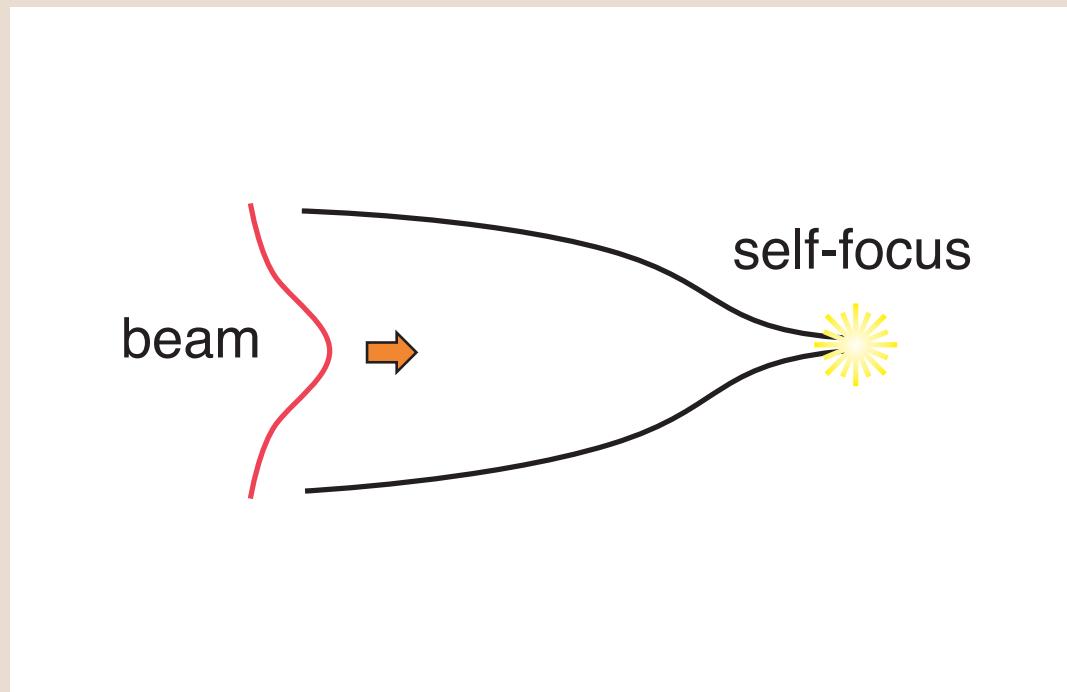
loosely focus femtosecond pulse into a transparent material



pulse is transformed into a white-light continuum

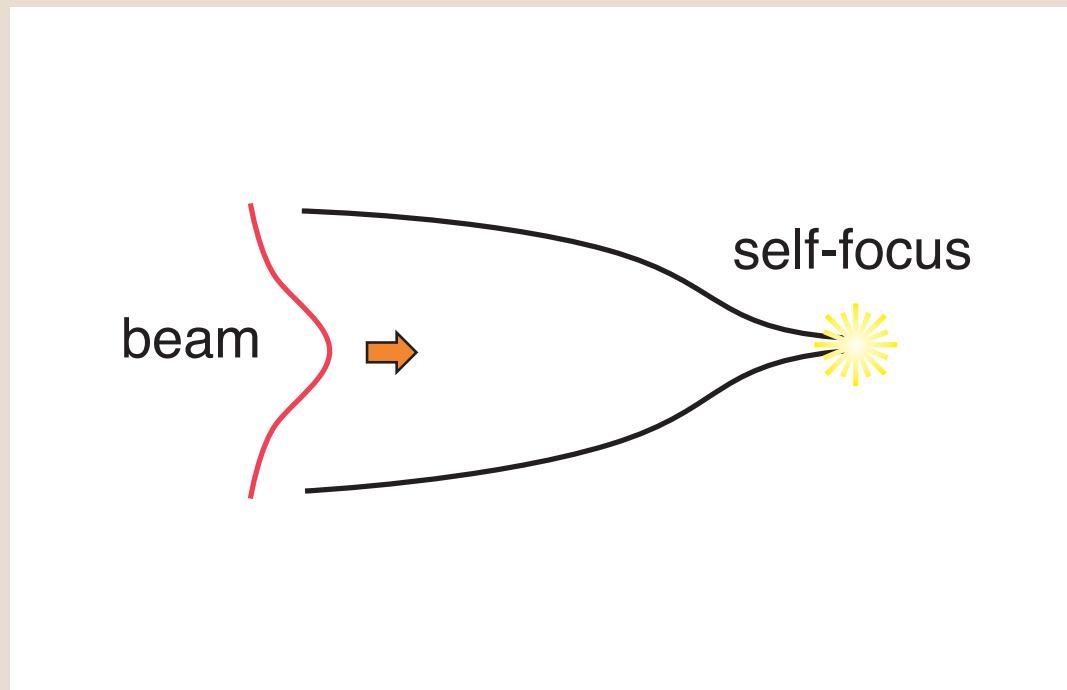


mechanisms



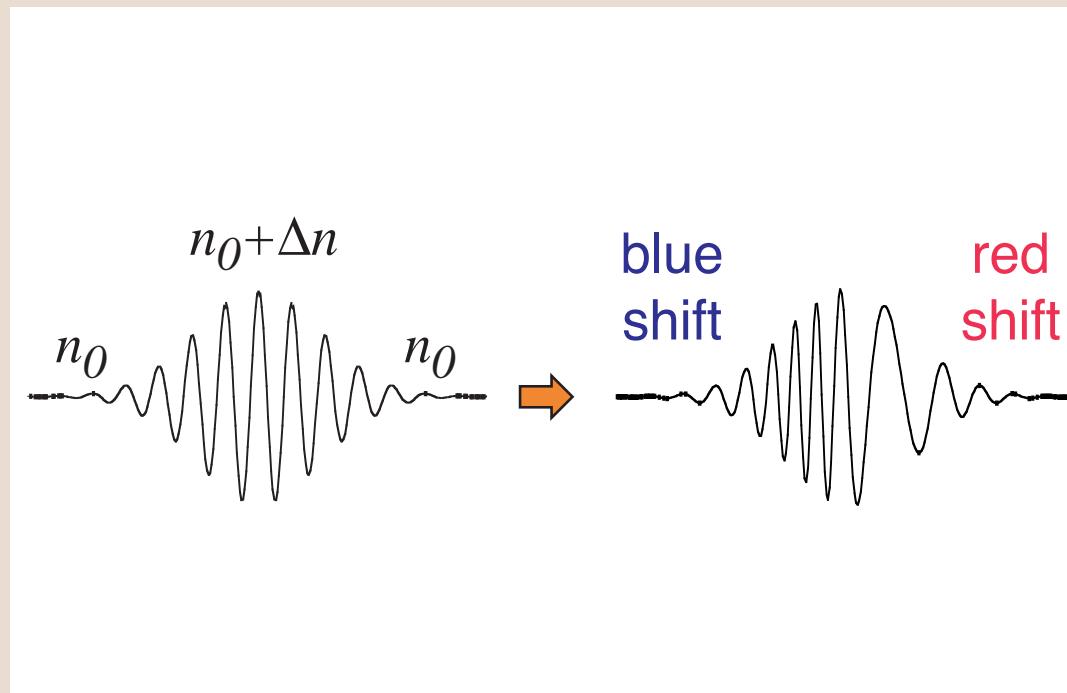
self-focusing: $n(r) = n_0 + n_2 I(r)$

mechanisms



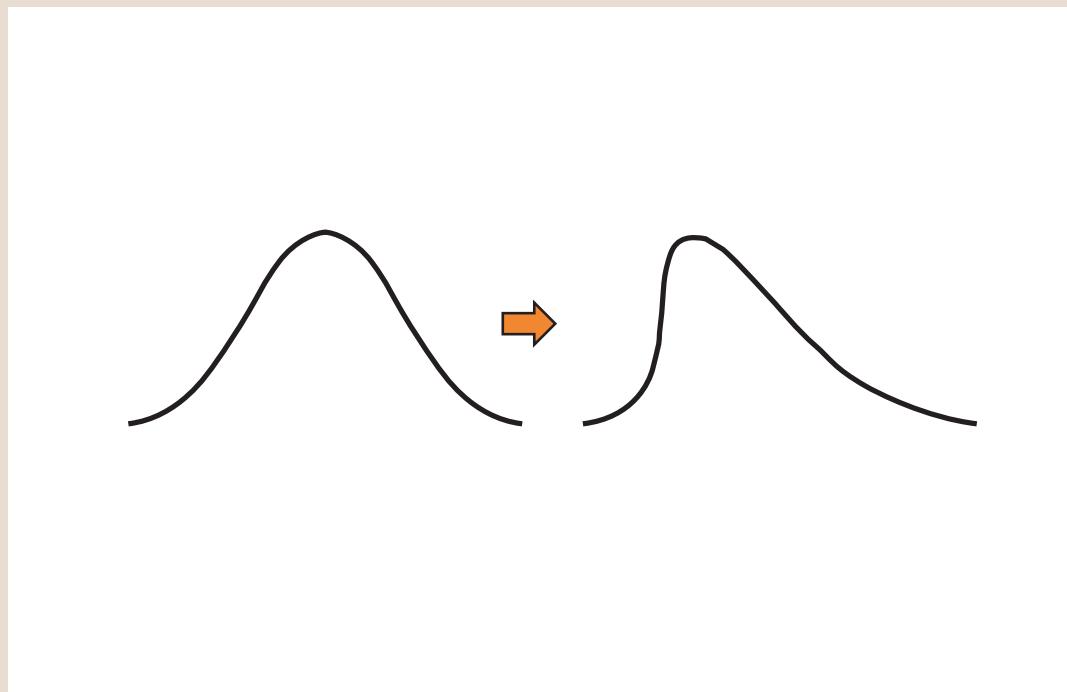
self-focusing: POWER dependent

mechanisms



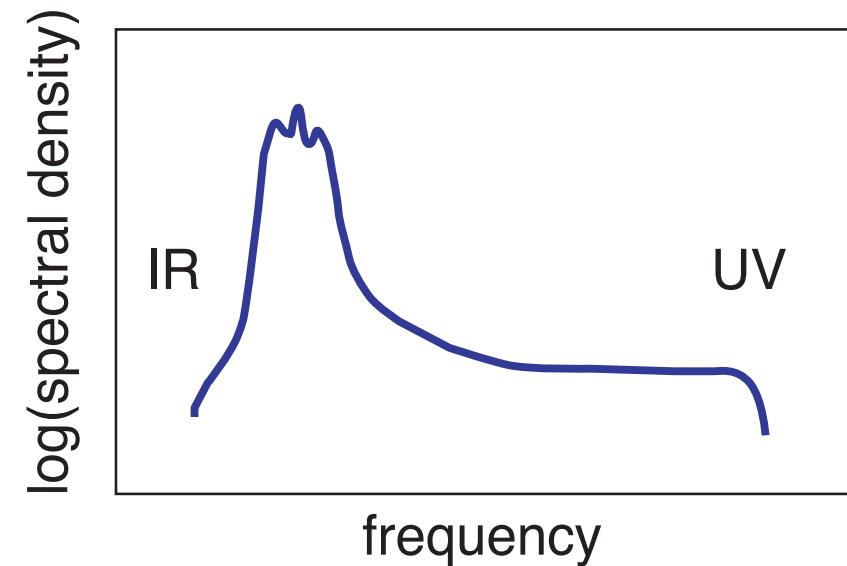
self phase modulation: $n(t) = n_0 + n_2 I(t)$

mechanisms



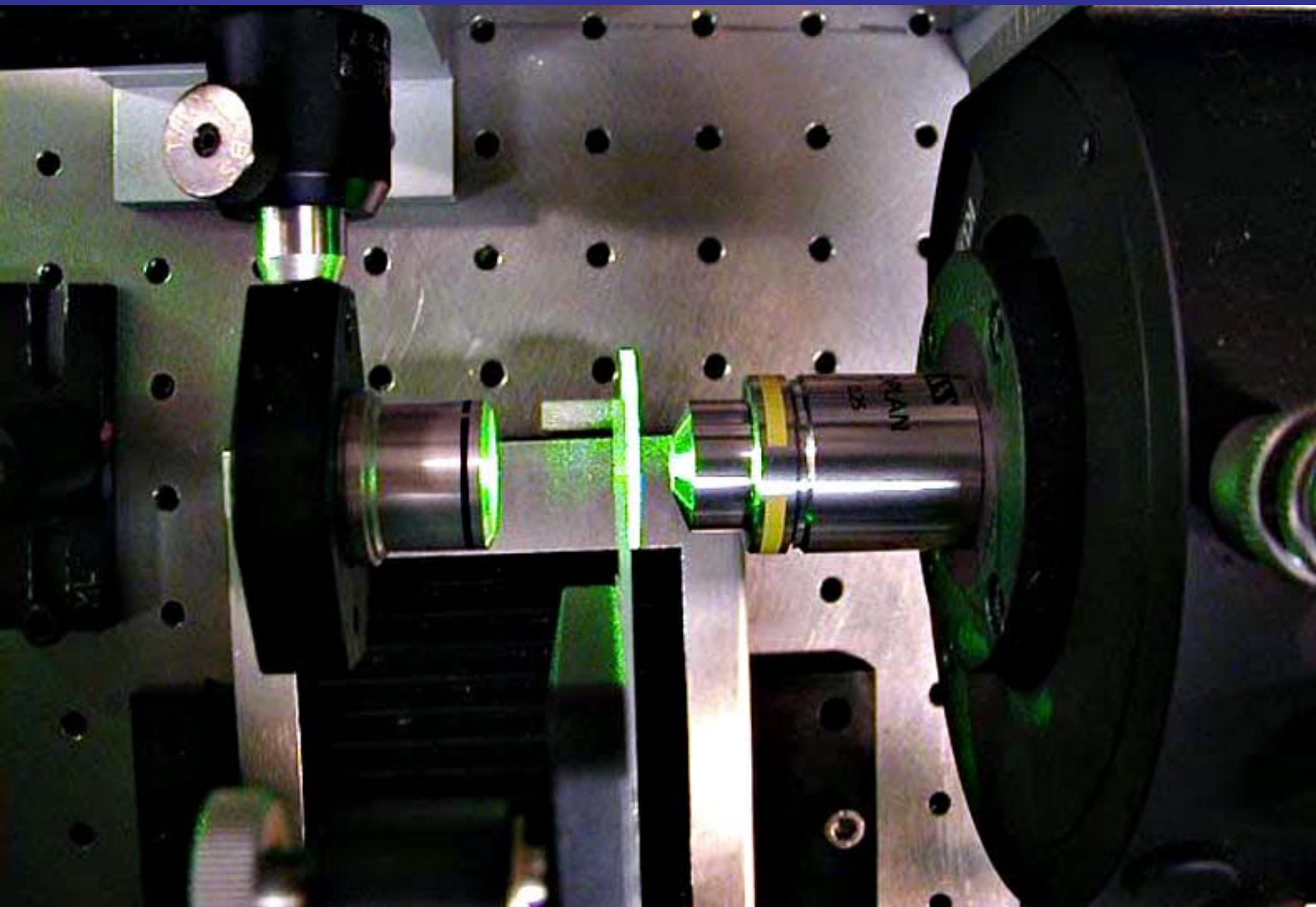
group velocity depends on intensity

continuum generation



why the asymmetry?

OUTLINE



Bulk damage thresholds

Ionization mechanisms

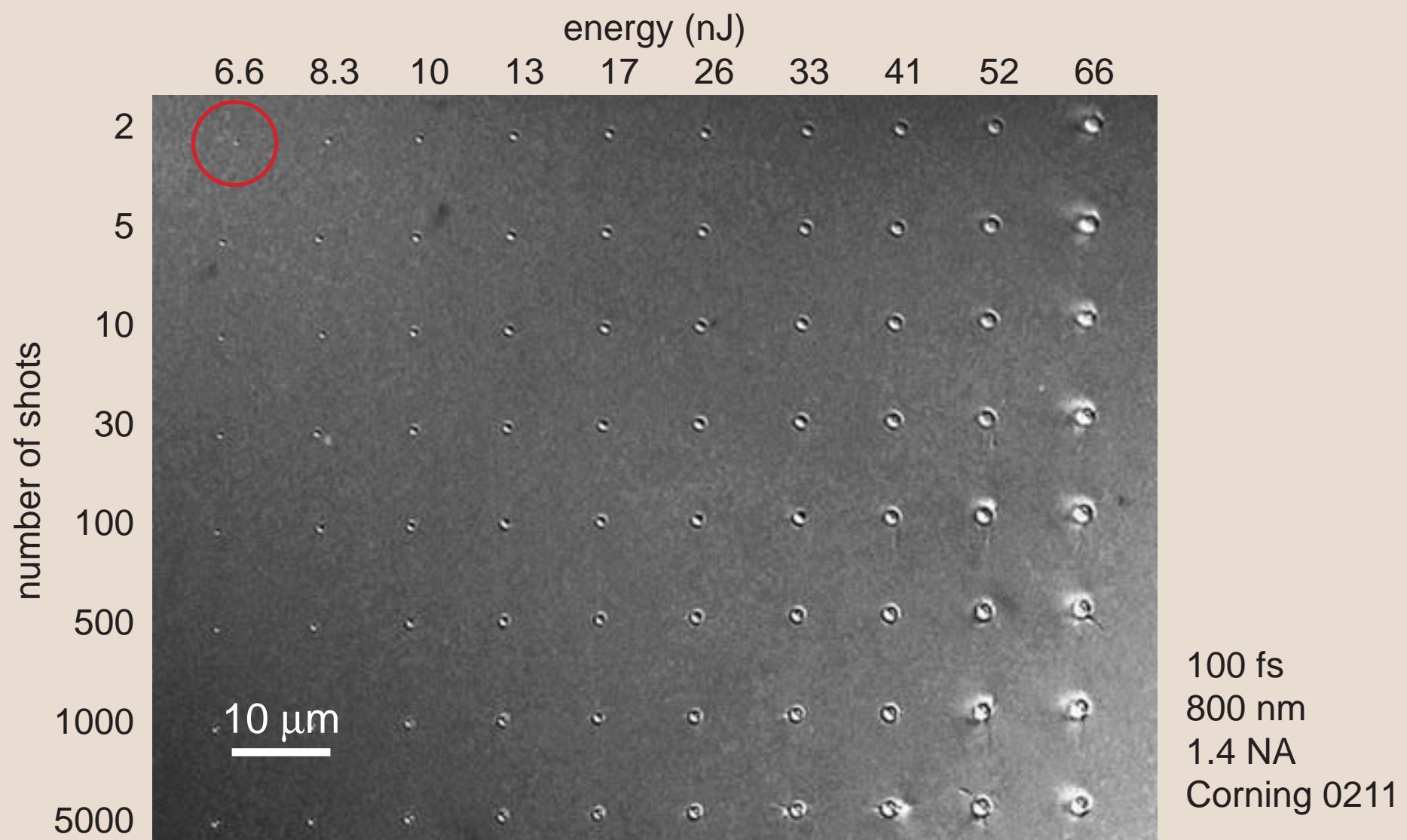
Continuum generation

Connections?

THRESHOLDS

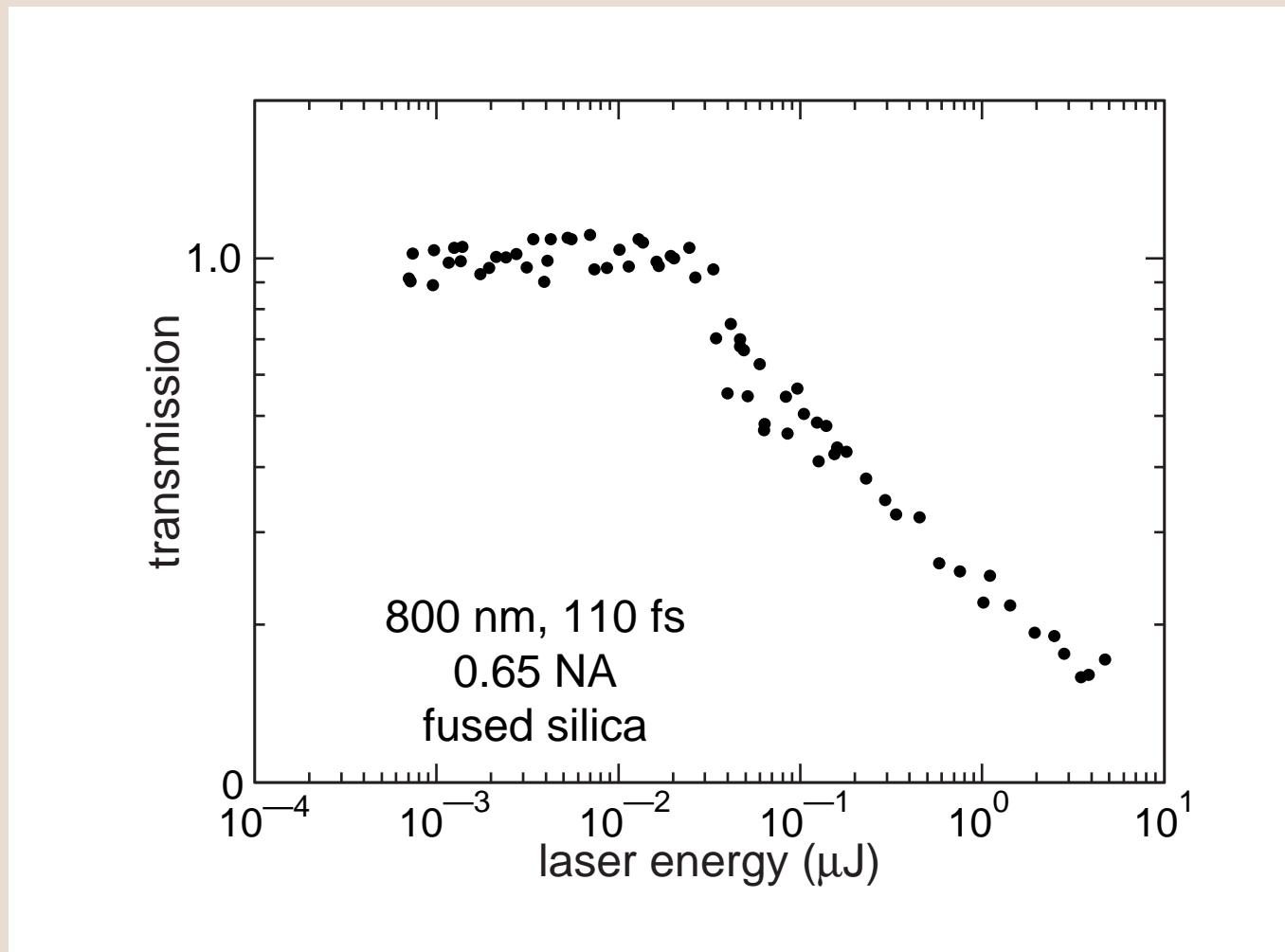
**how little energy produces
permanent changes?**

THRESHOLDS

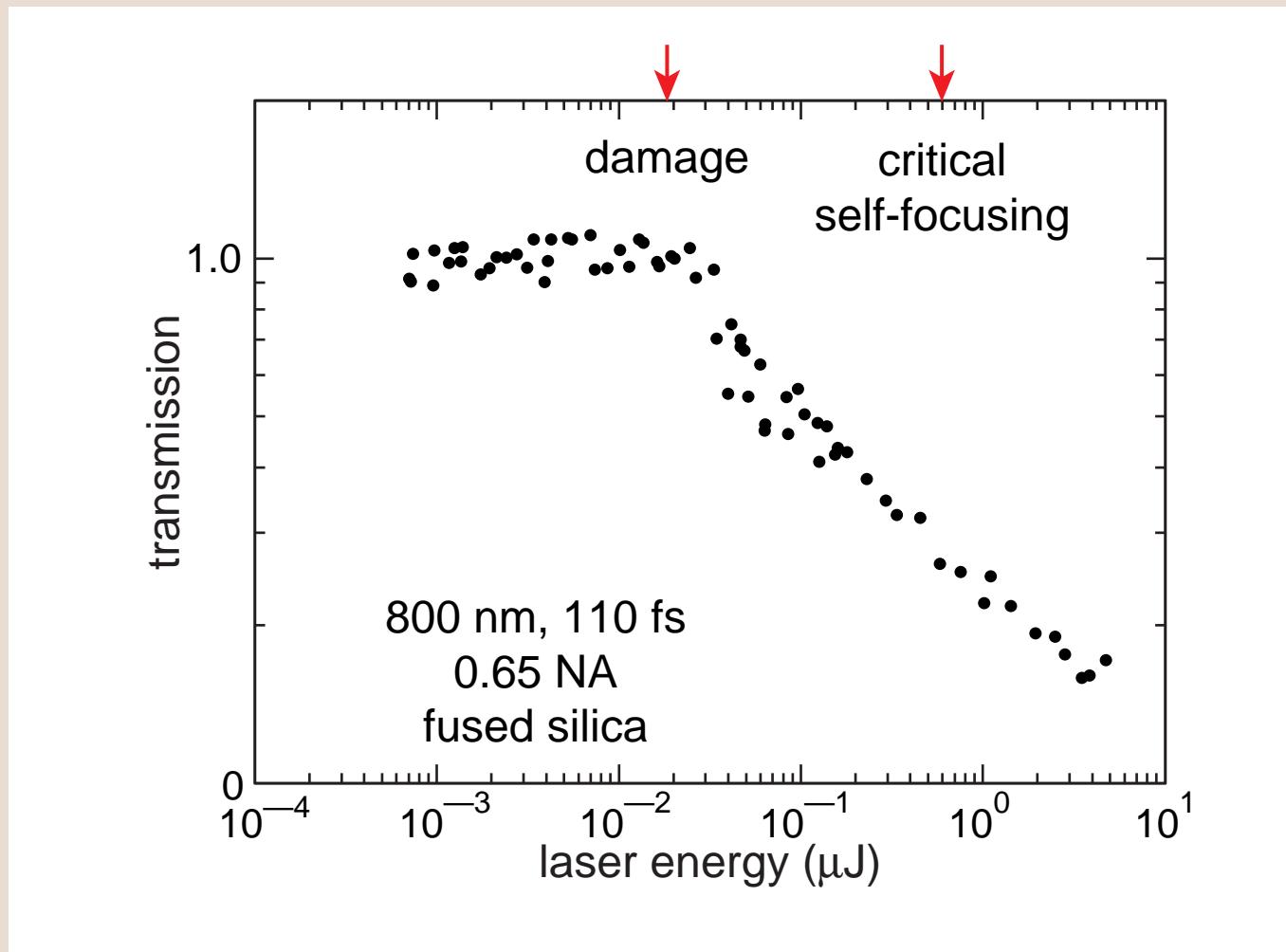


post-mortem microscopy: threshold < 6.6 nJ

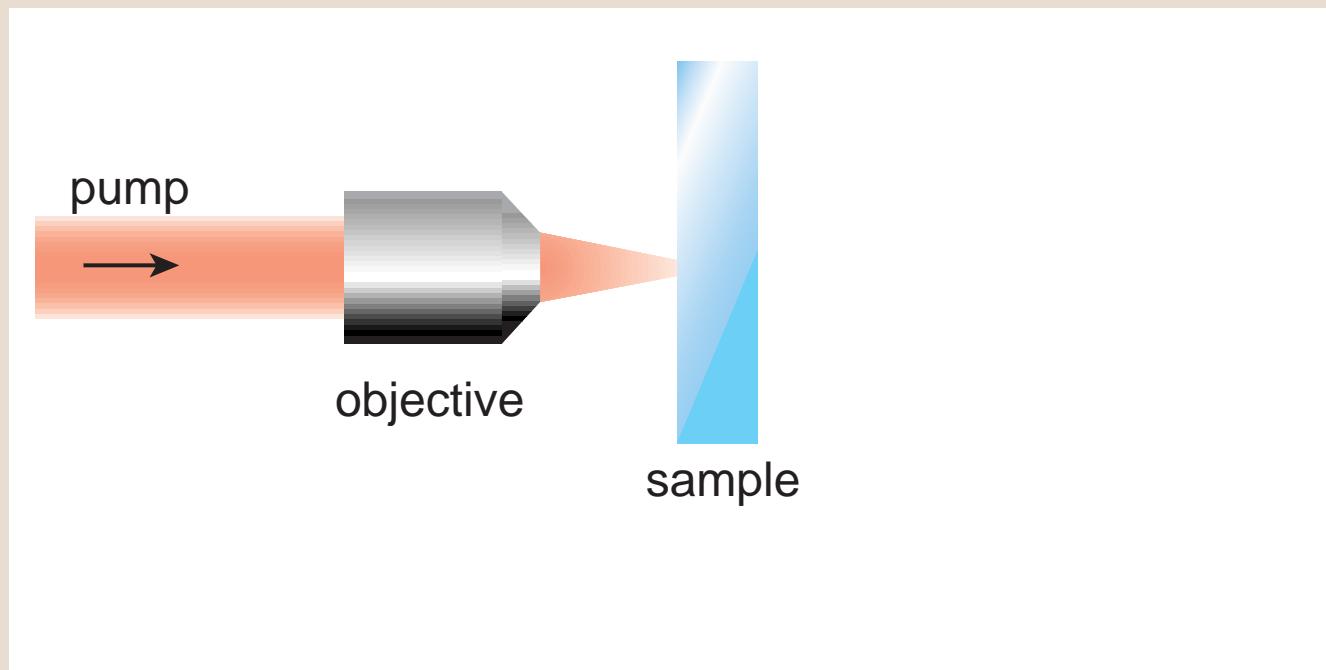
transmission of laser pulse as a function of energy



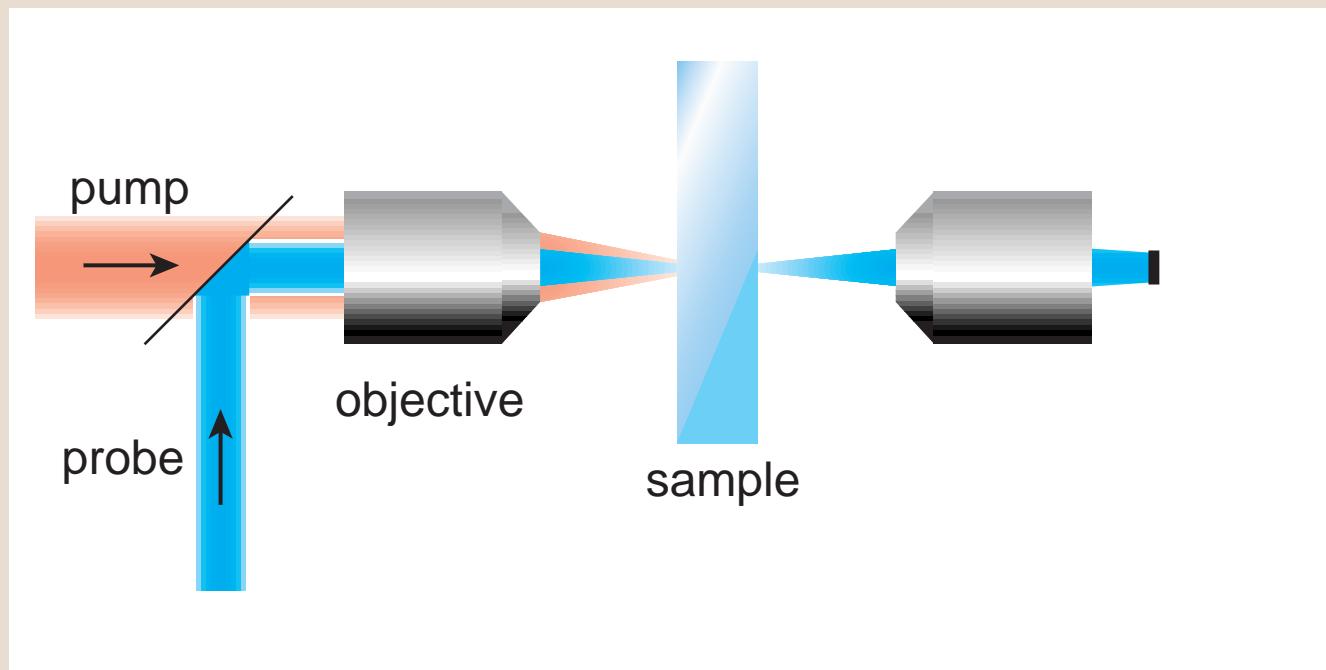
self-focusing threshold much higher



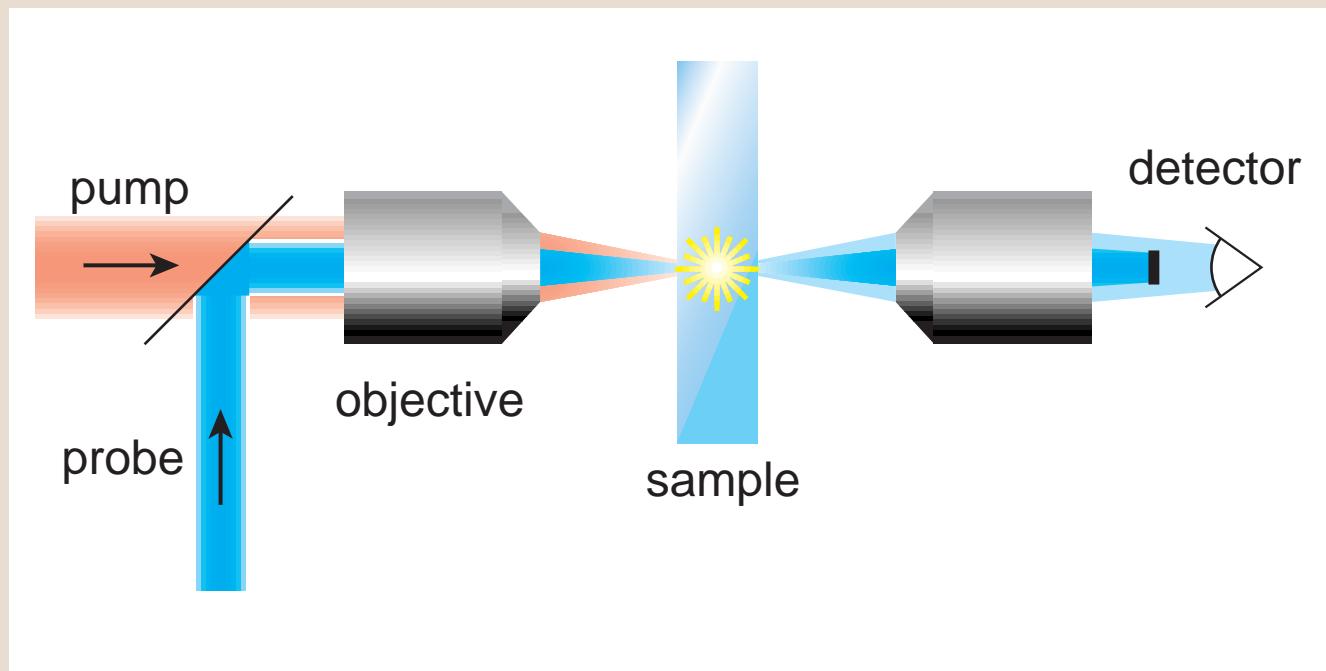
pump sample with femtosecond pulse



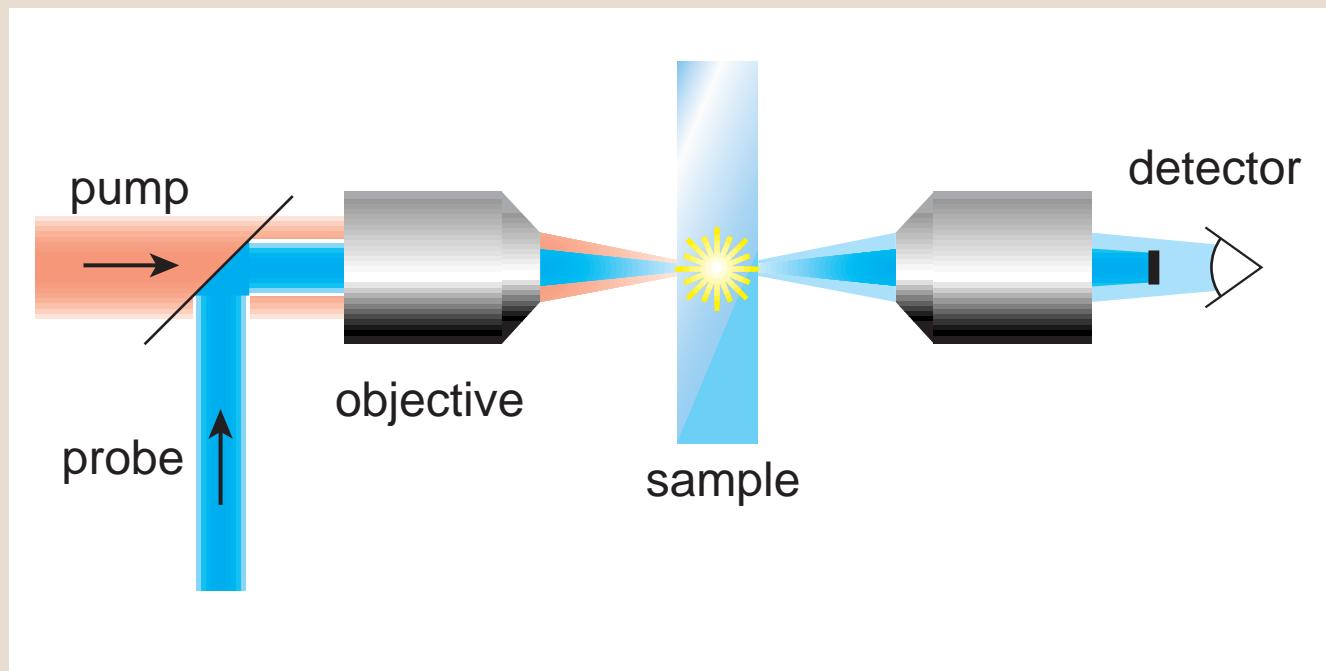
block probe beam



detect light scattered by damage

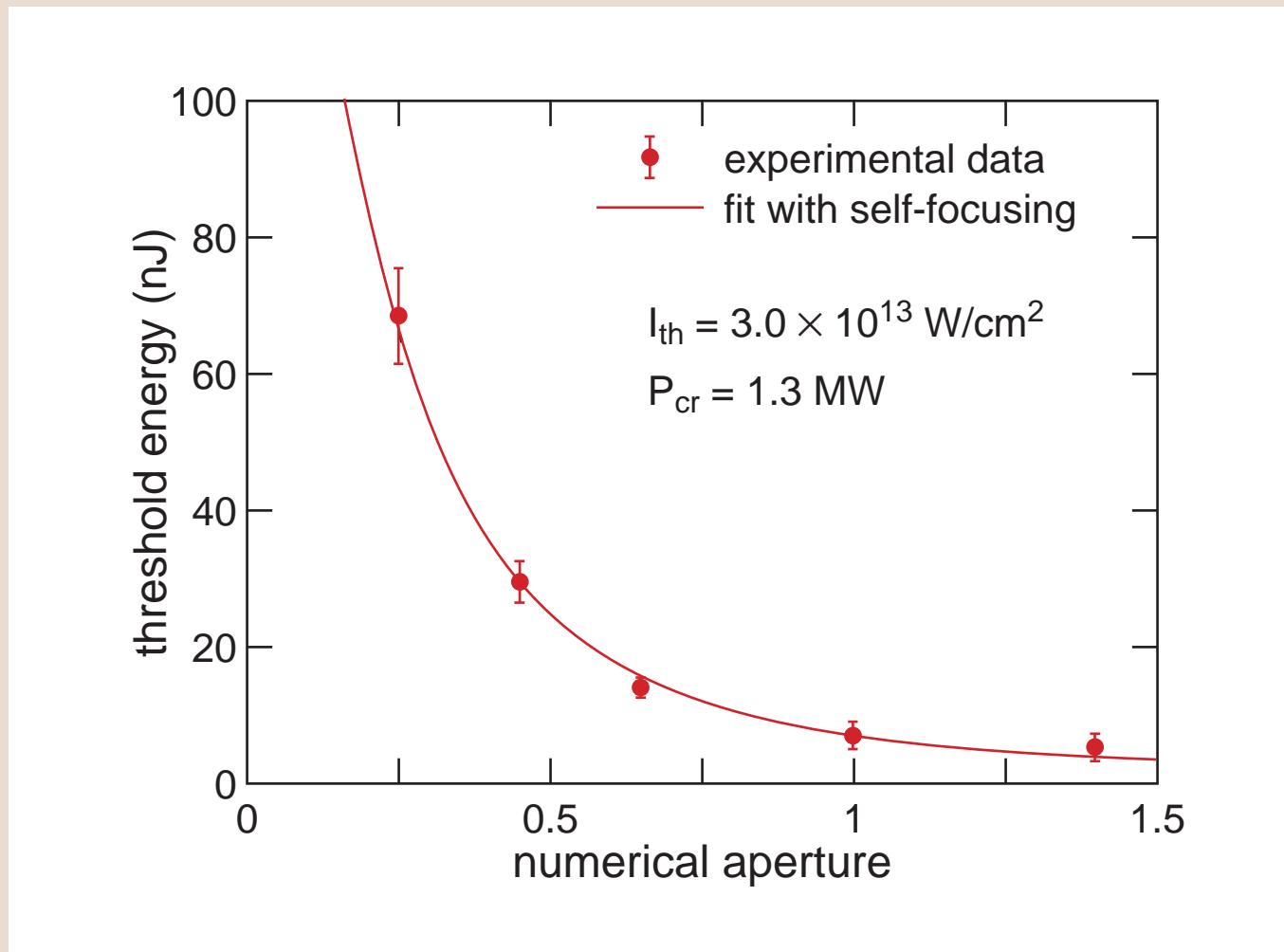


THRESHOLDS

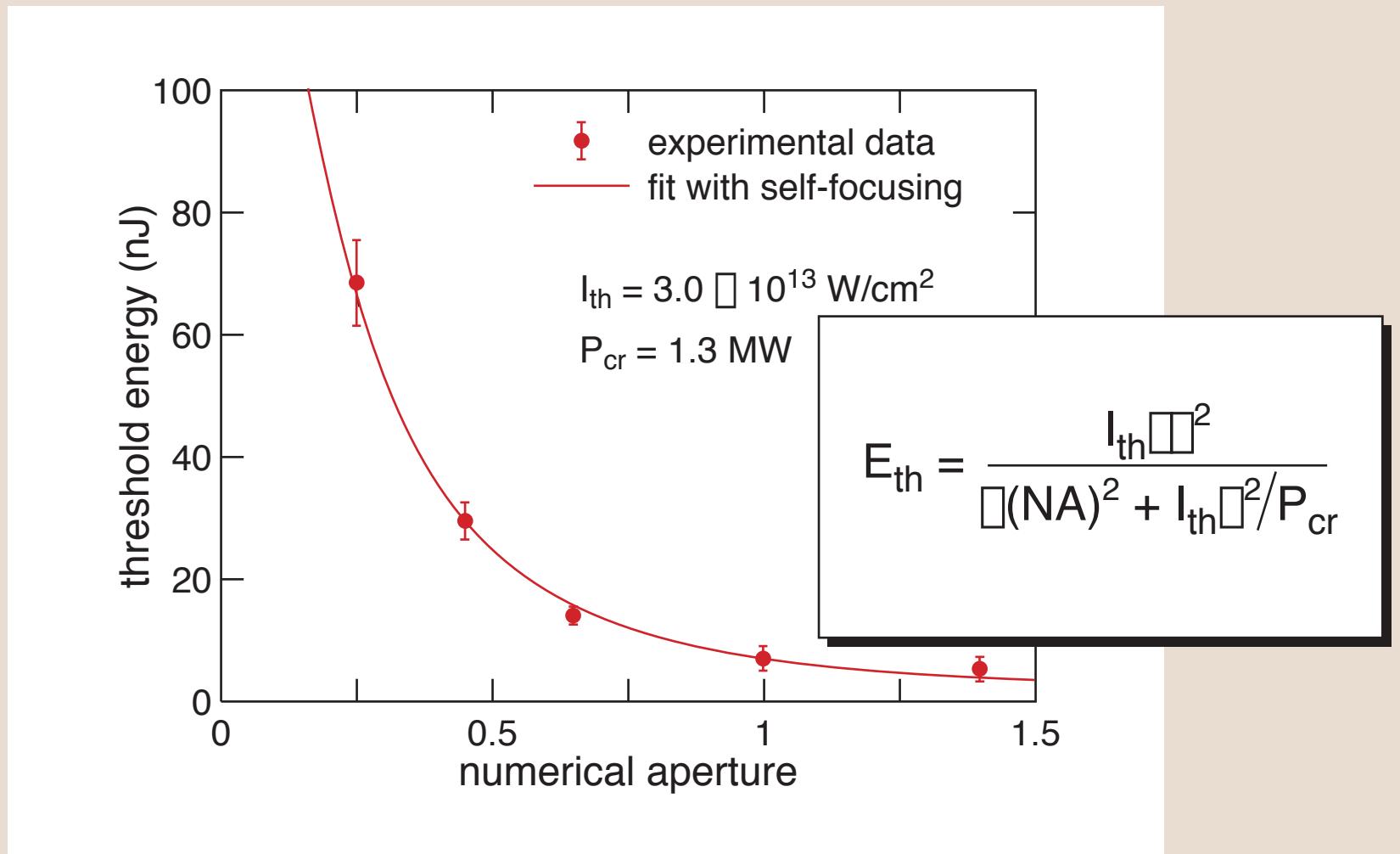


vary NA, material, pump wavelength

THRESHOLDS

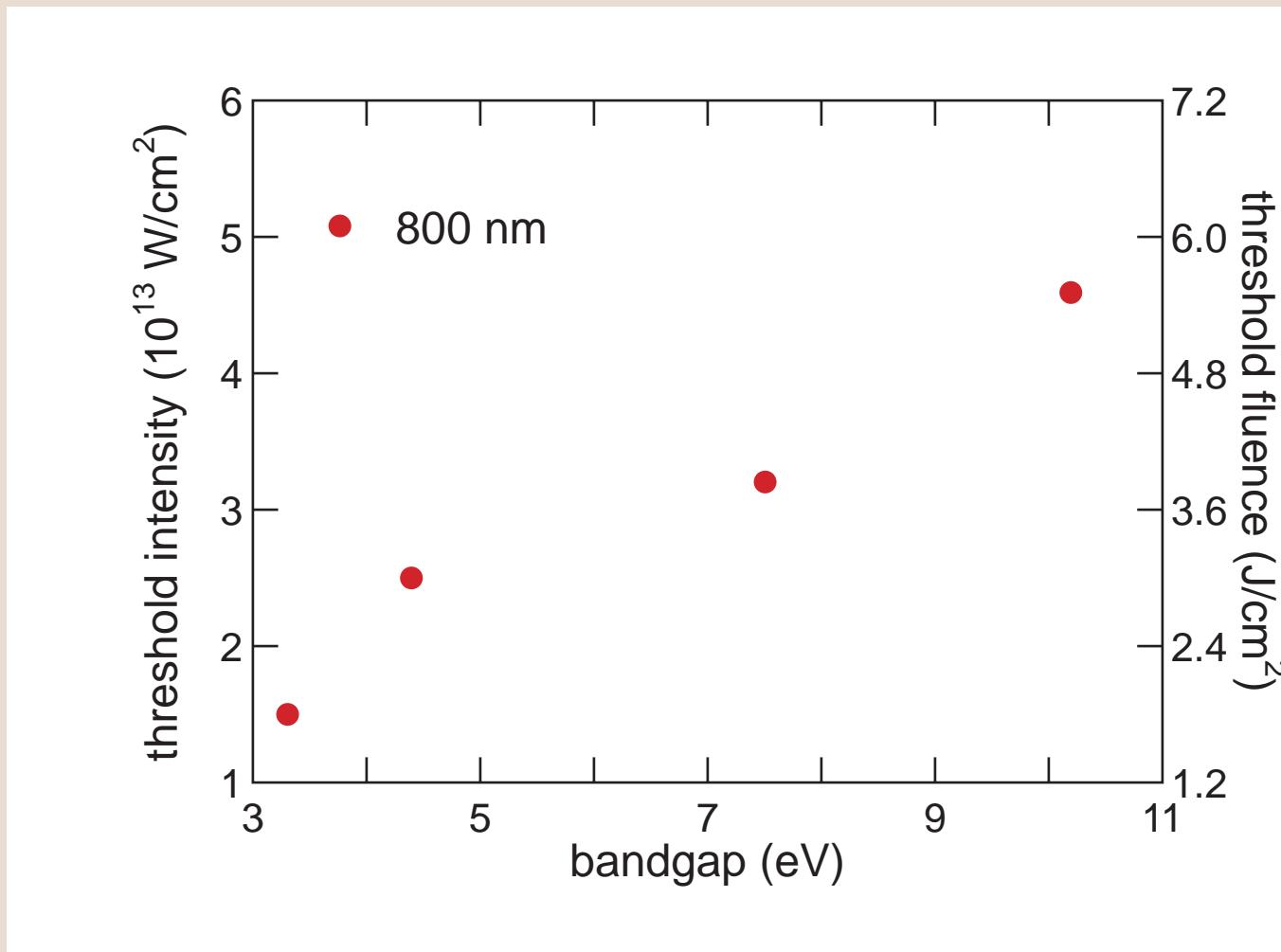


energy threshold vs. NA for 100-fs pulses in Corning 0211

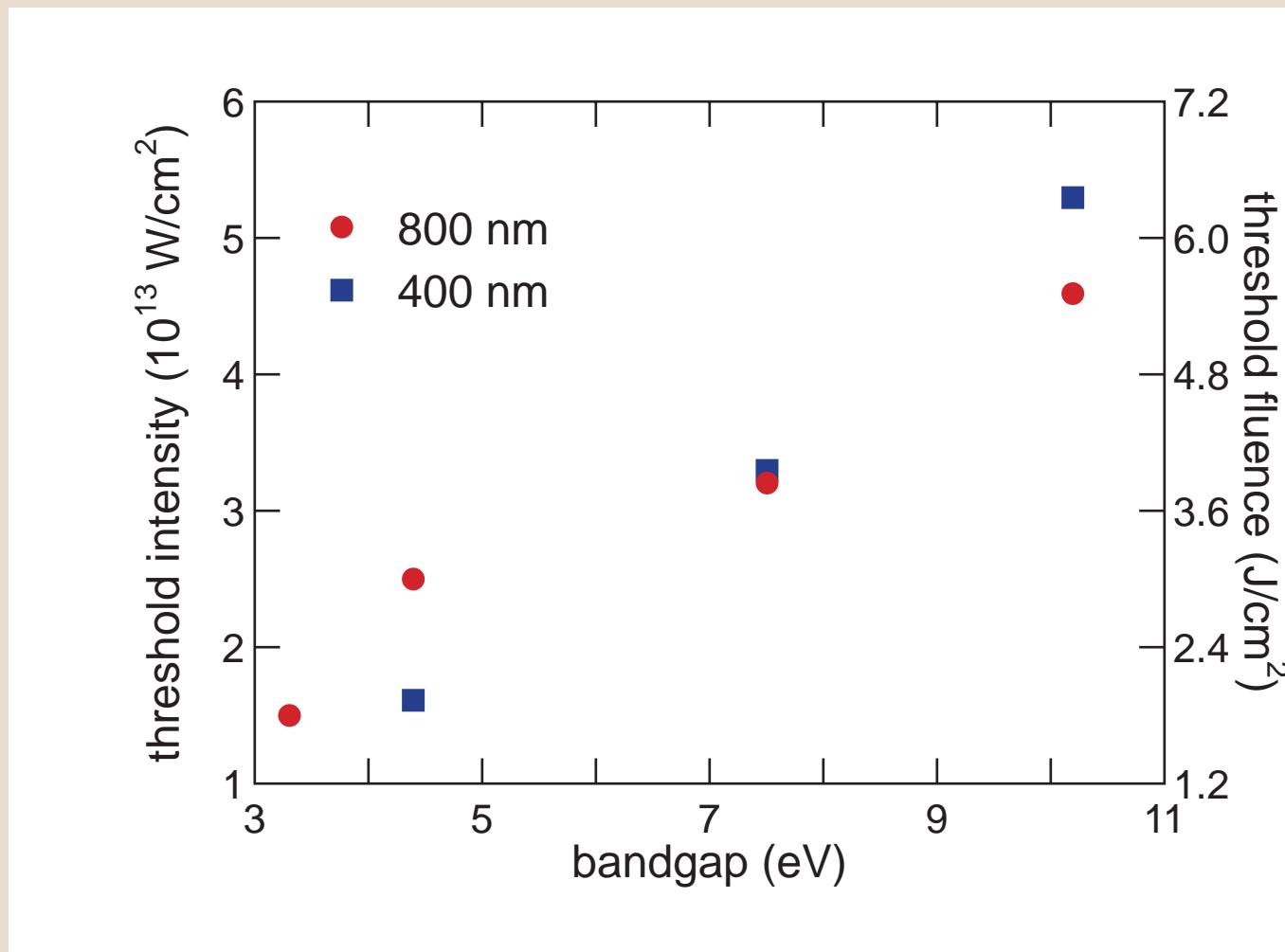


energy threshold vs. NA for 100-fs pulses in Corning 0211

bandgap dependence of threshold intensity



repeat experiment for frequency-doubled pulses

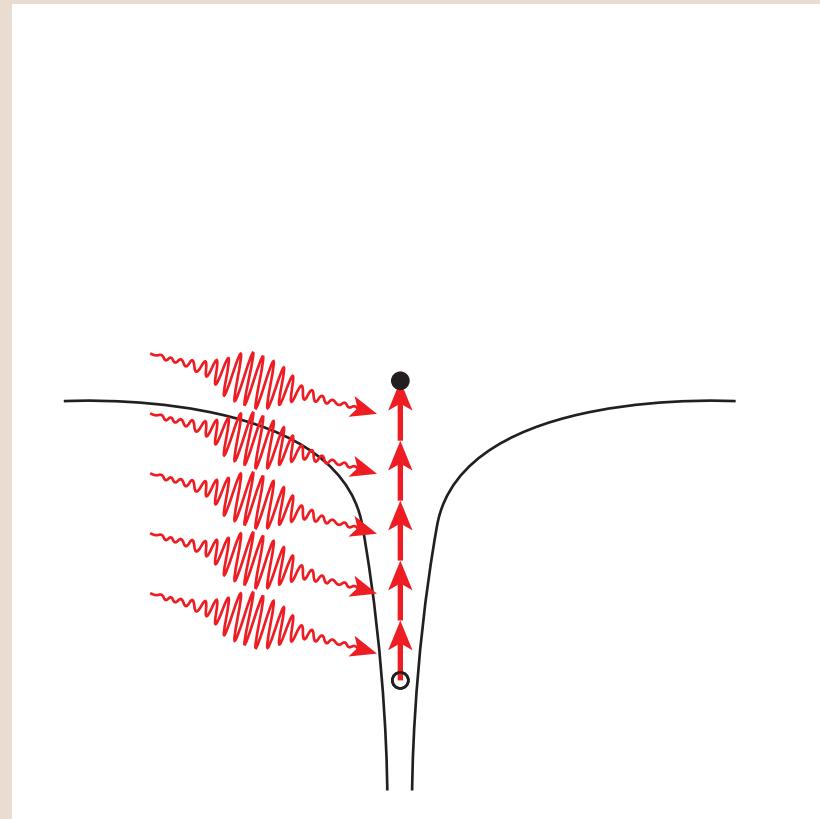


IONIZATION MECHANISMS

what do these thresholds tell us
about fundamental processes?

IONIZATION MECHANISMS

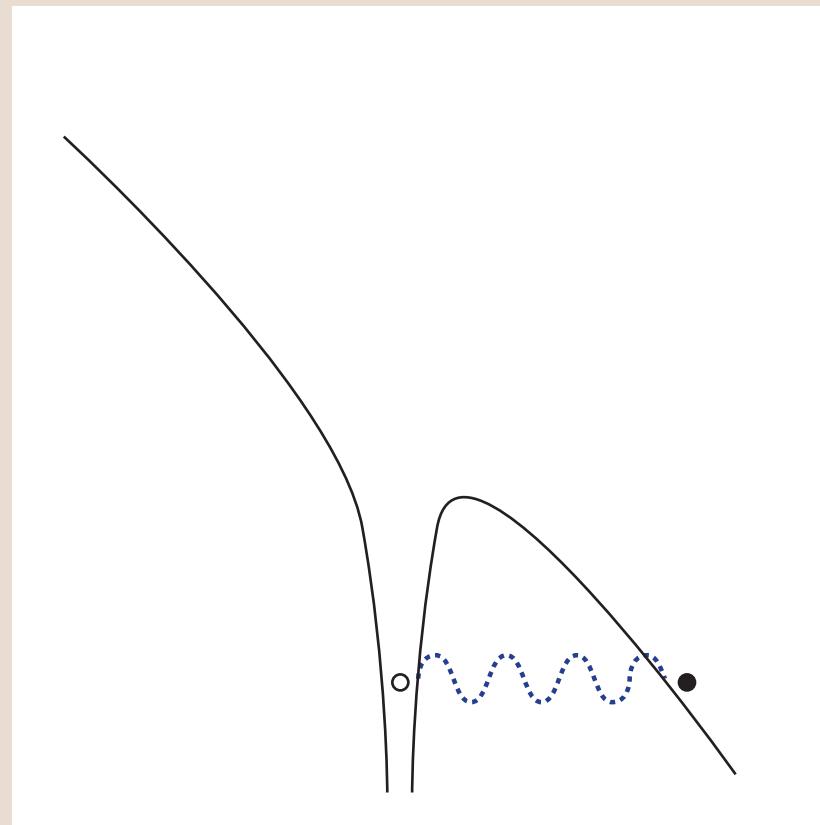
photoionization: multiphoton



nonlinear: I^n

IONIZATION MECHANISMS

photoionization: tunneling



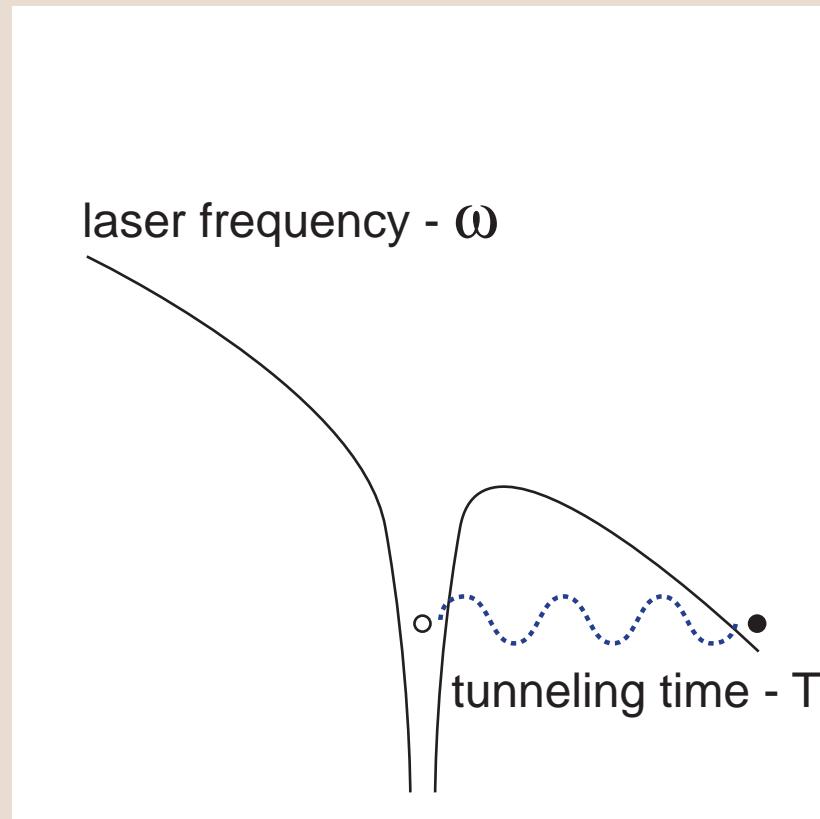
nonlinear: $I^{-1/2} \exp(I^{-1/2})$

IONIZATION MECHANISMS

Keldysh parameter

$$\gamma = (\omega T) / 2^{1/2}$$

$\gamma > 1.5$ MPI
 $\gamma < 1.5$ tunneling

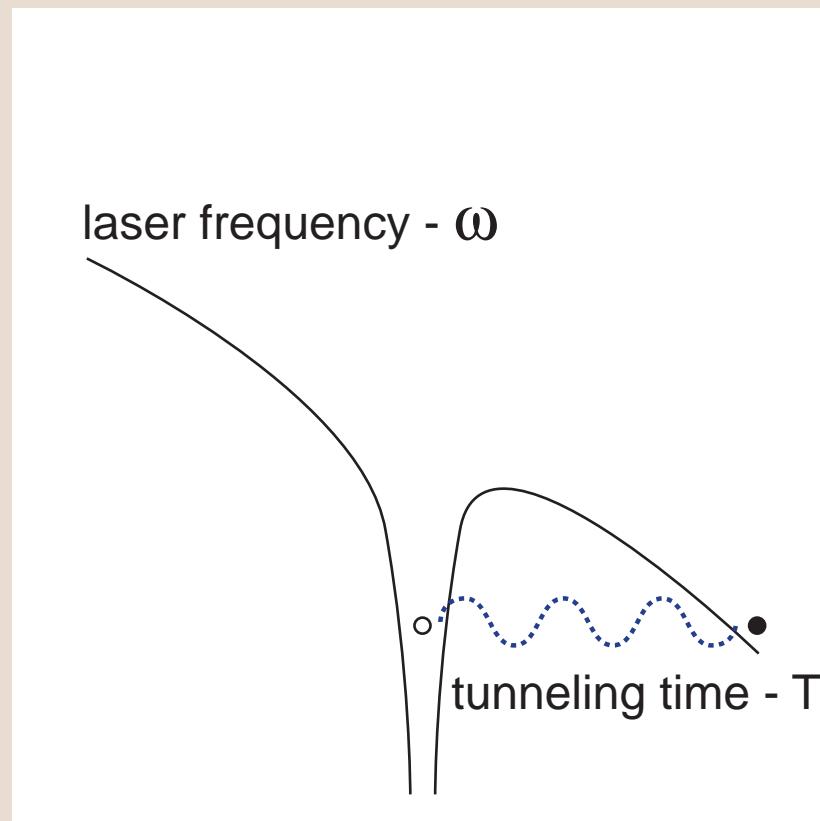


Keldysh parameter

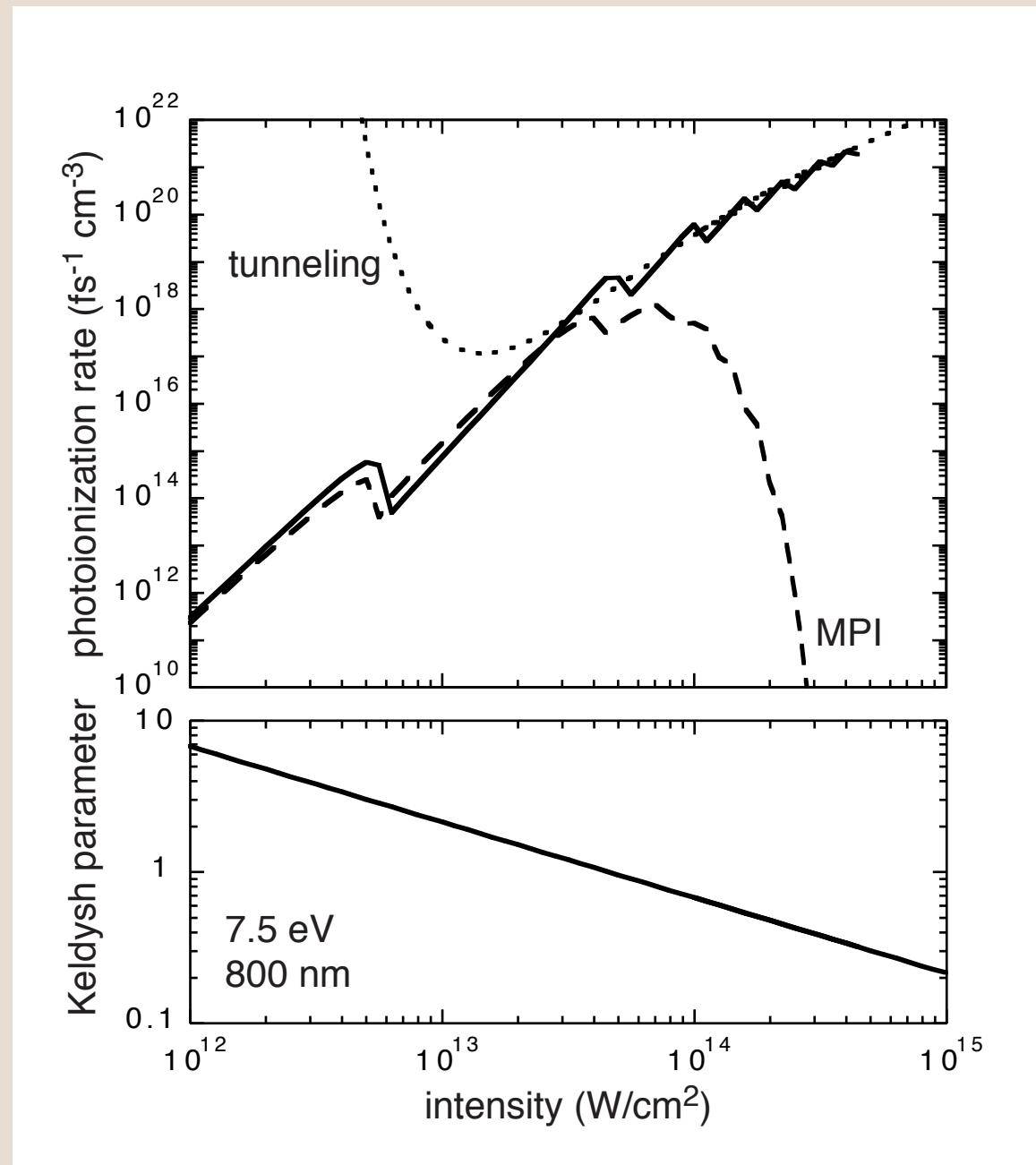
$$\gamma = (\omega^2 m c n \epsilon_0 E_g / e^2 I)^{1/2}$$

$\gamma > 1.5$ MPI

$\gamma < 1.5$ tunneling

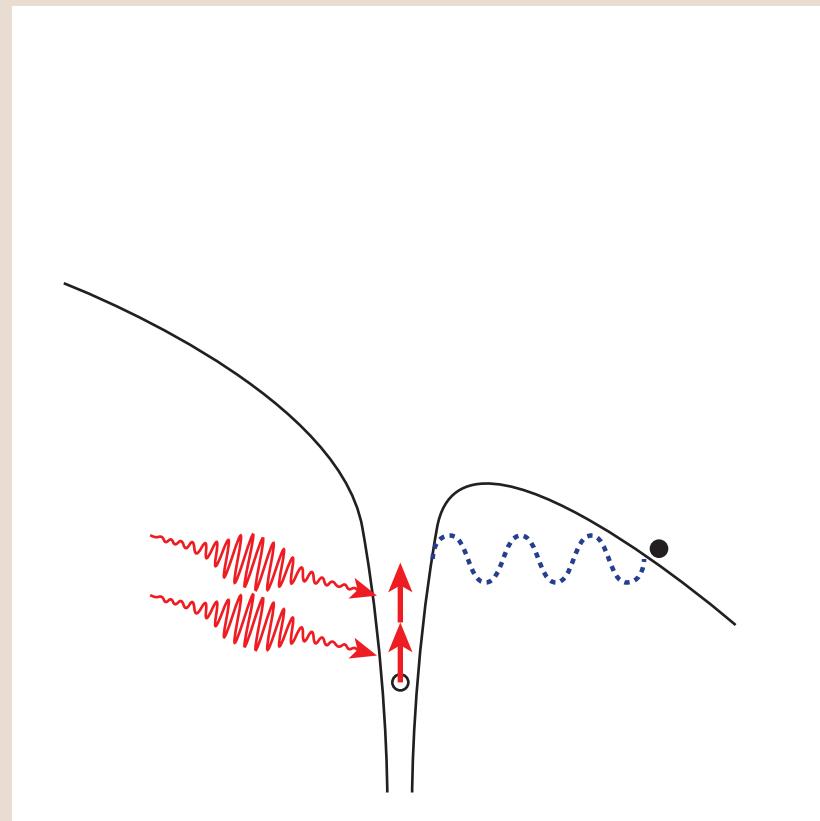


photoionization rates and Keldysh parameter vs. laser intensity



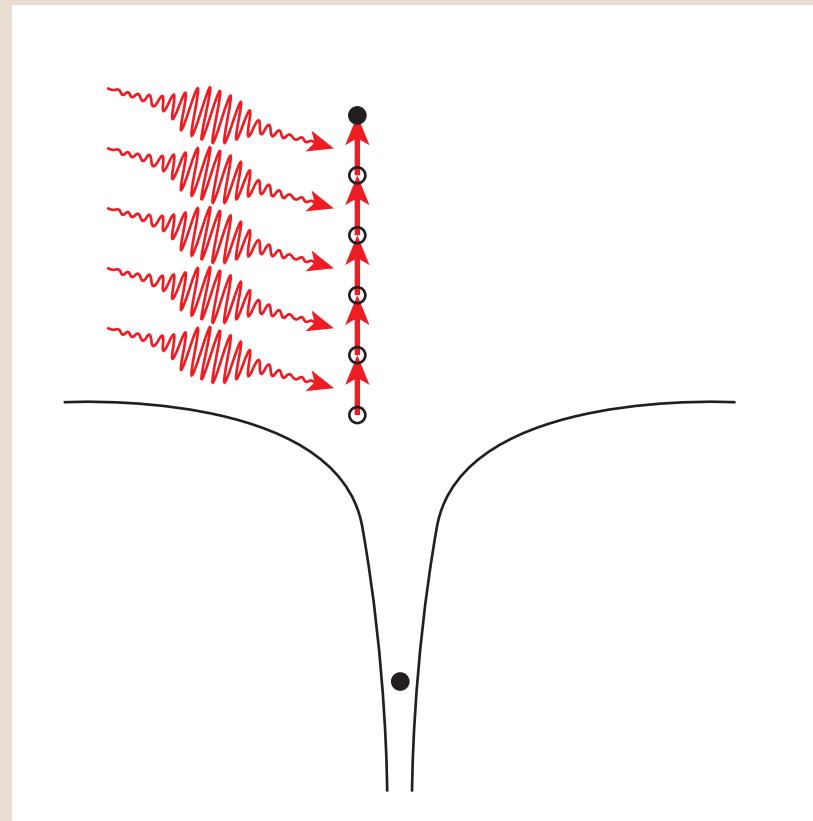
IONIZATION MECHANISMS

photoionization: in between



IONIZATION MECHANISMS

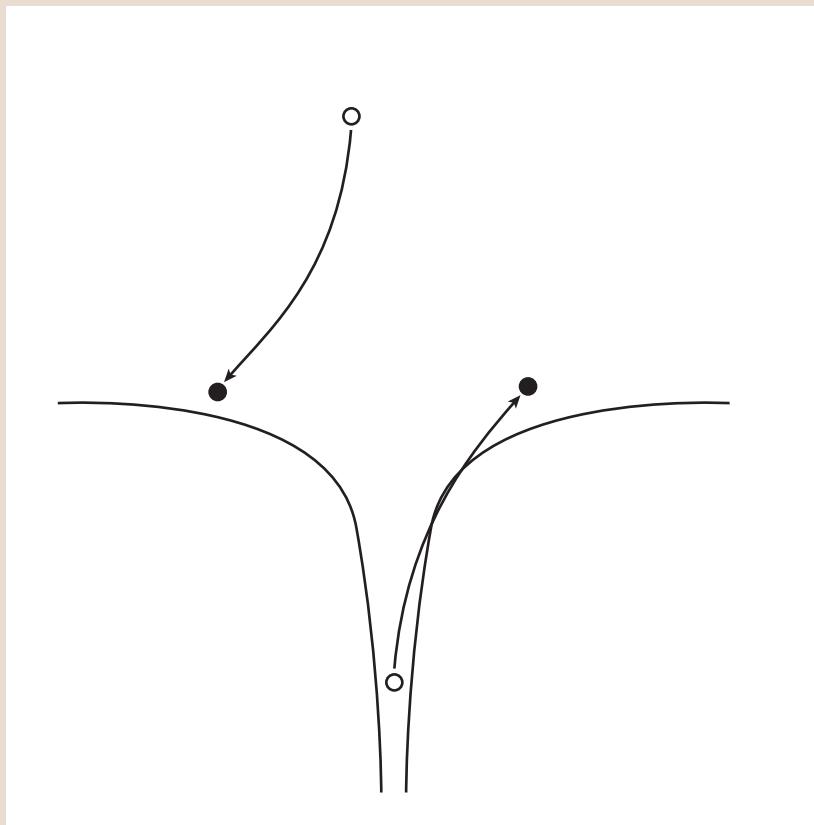
avalanche ionization: free carrier absorption



linear: nI

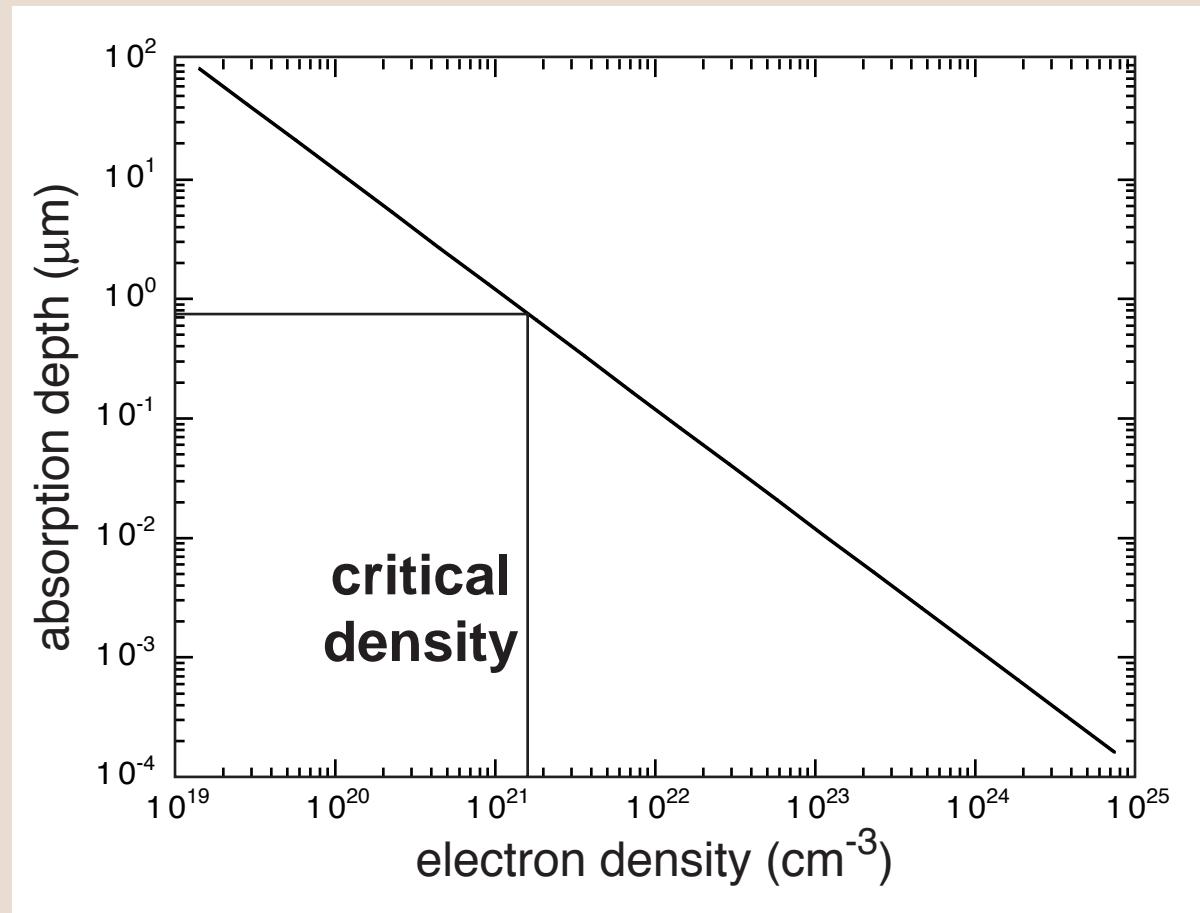
IONIZATION MECHANISMS

avalanche ionization: impact ionization



linear: nI

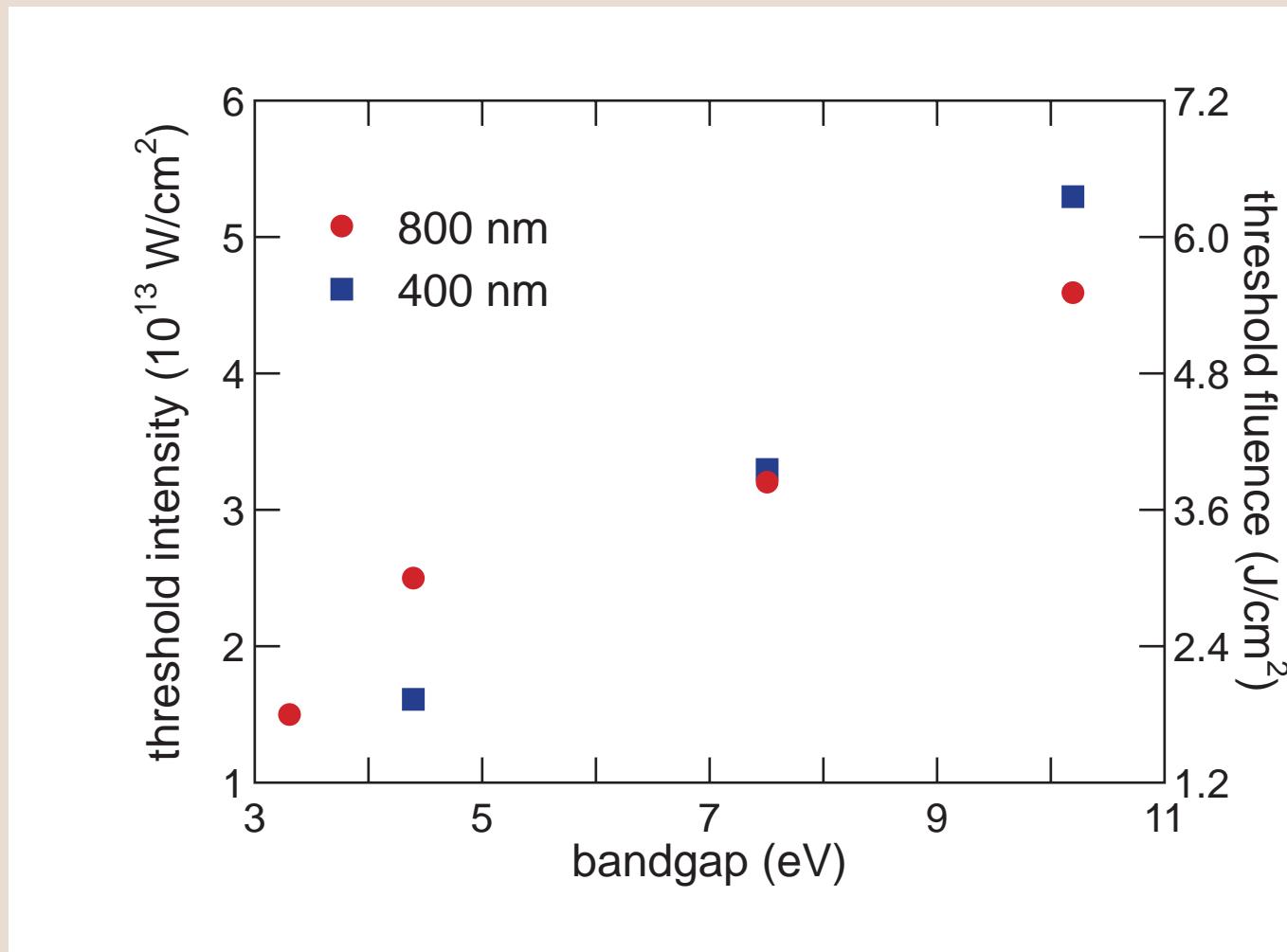
IONIZATION MECHANISMS



plasma absorption depth at 800 nm

IONIZATION MECHANISMS

use our measured threshold intensities



IONIZATION MECHANISMS

Keldysh parameter

$$\gamma = (\omega^2 m c n \epsilon_0 E_g / e^2 I)^{1/2}$$

$\gamma > 1.5$ MPI
 $\gamma < 1.5$ tunneling

material	γ (800 nm)
CaF ₂	1.2
FS	1.2
O2I1	1.1
SF11	1.3

IONIZATION MECHANISMS

Keldysh parameter

$$\gamma = (\omega^2 m c n \epsilon_0 E_g / e^2 I)^{1/2}$$

$\gamma > 1.5$ MPI
 $\gamma < 1.5$ tunneling

material	γ (800 nm)	γ (400 nm)
CaF ₂	1.2	2.1
FS	1.2	2.4
O2I1	1.1	2.6
SF11	1.3	

IONIZATION MECHANISMS

Keldysh parameter

$$\gamma = (\omega^2 m c n \epsilon_0 E_g / e^2 I)^{1/2}$$

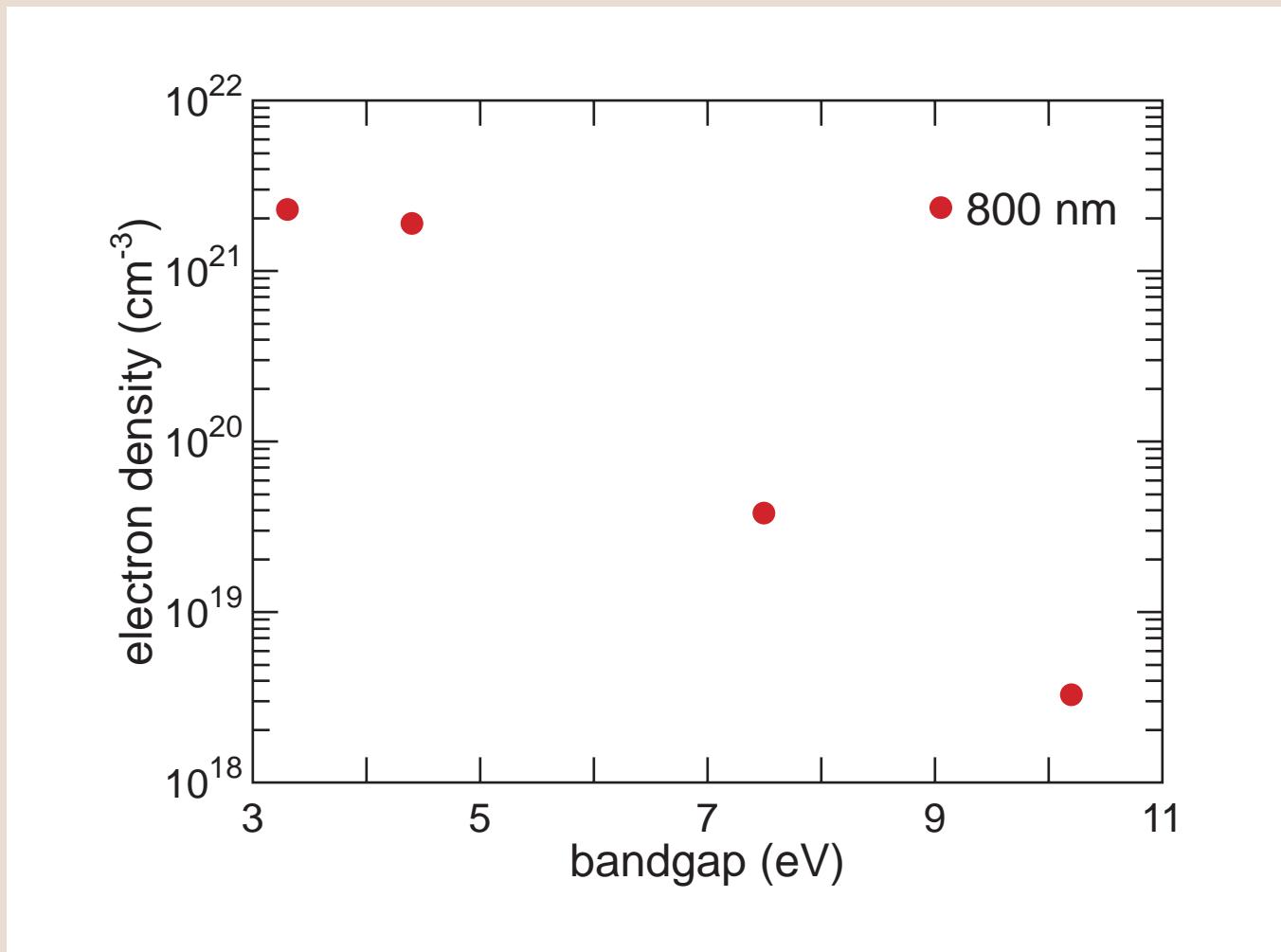
$\gamma > 1.5$ MPI
 $\gamma < 1.5$ tunneling

material	γ (800 nm)	γ (400 nm)
CaF ₂	1.2	2.1
FS	1.2	2.4
O211	1.1	2.6
SF11	1.3	

tunneling at 800 nm, MPI at 400 nm

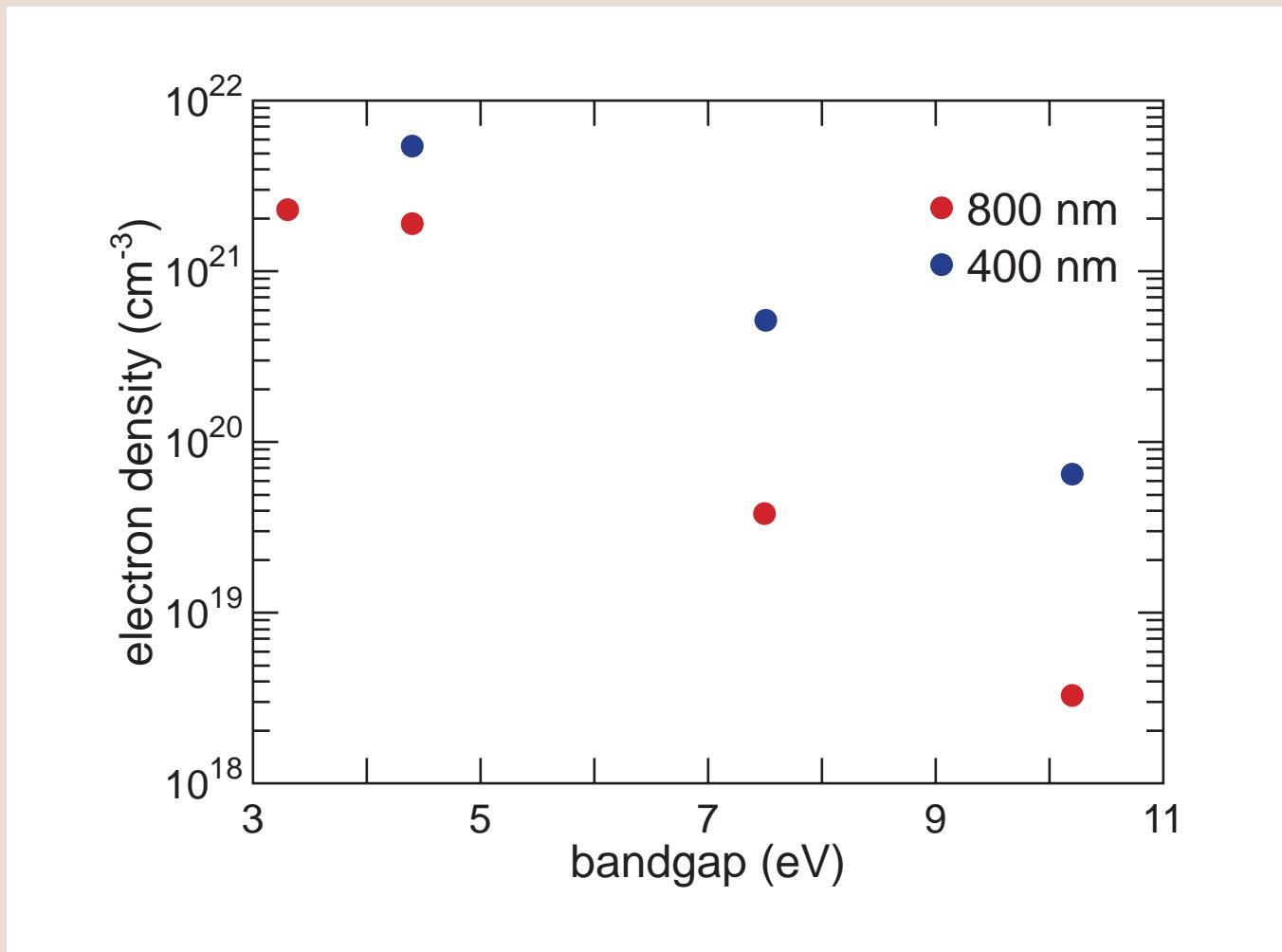
IONIZATION MECHANISMS

calculate electron density produced by MPI and tunneling



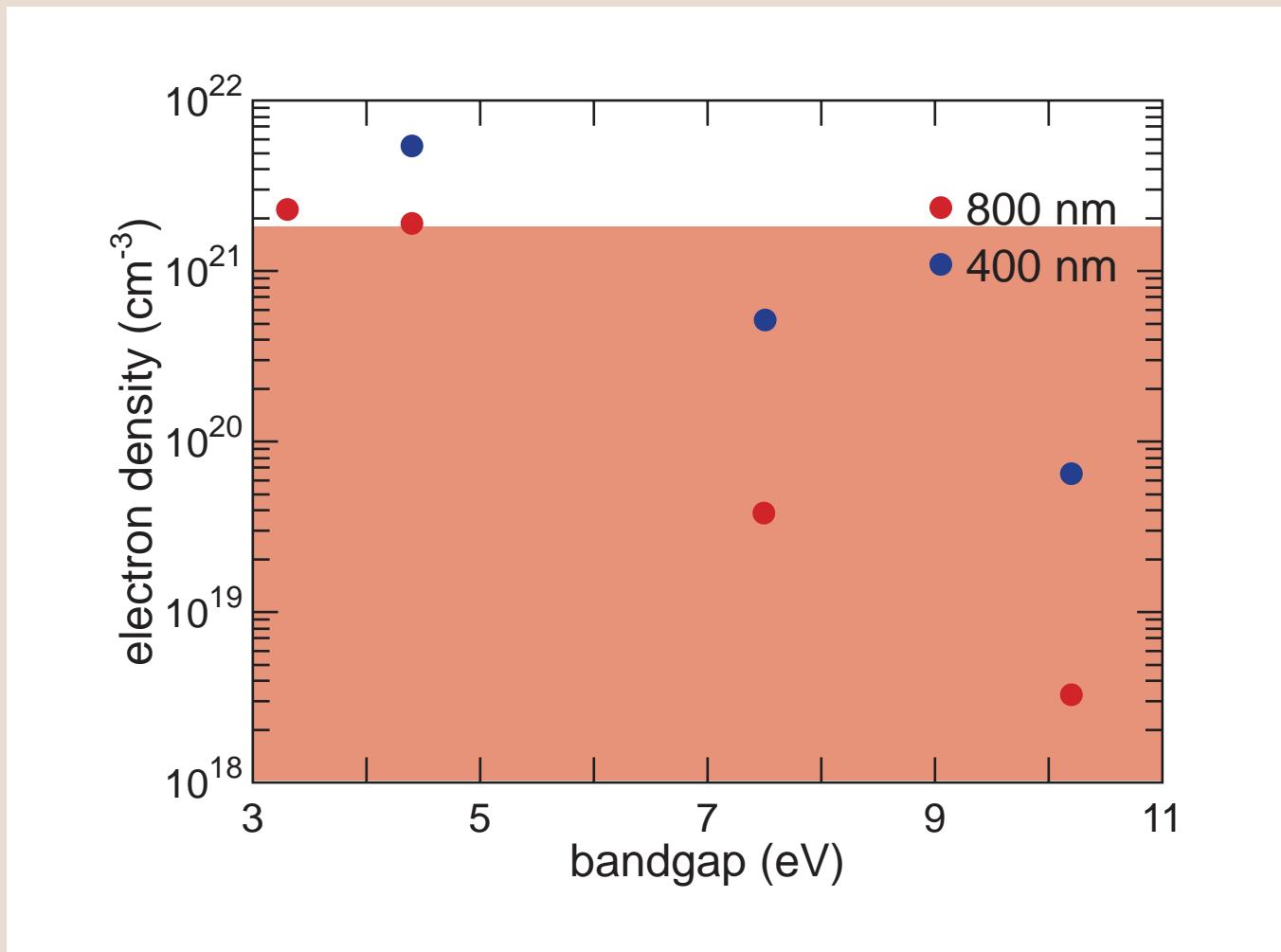
IONIZATION MECHANISMS

calculate electron density produced by MPI and tunneling

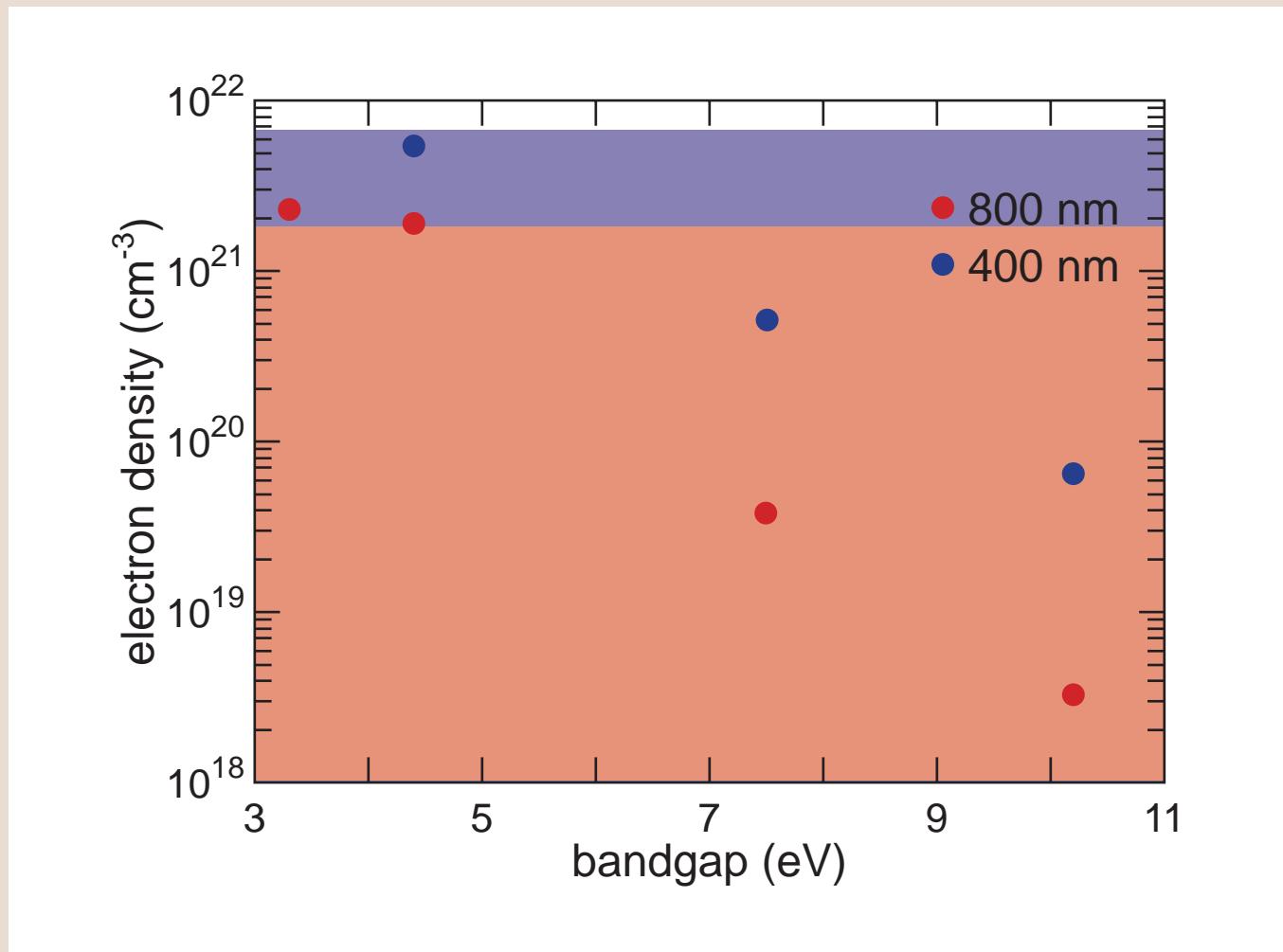


IONIZATION MECHANISMS

800 nm critical density

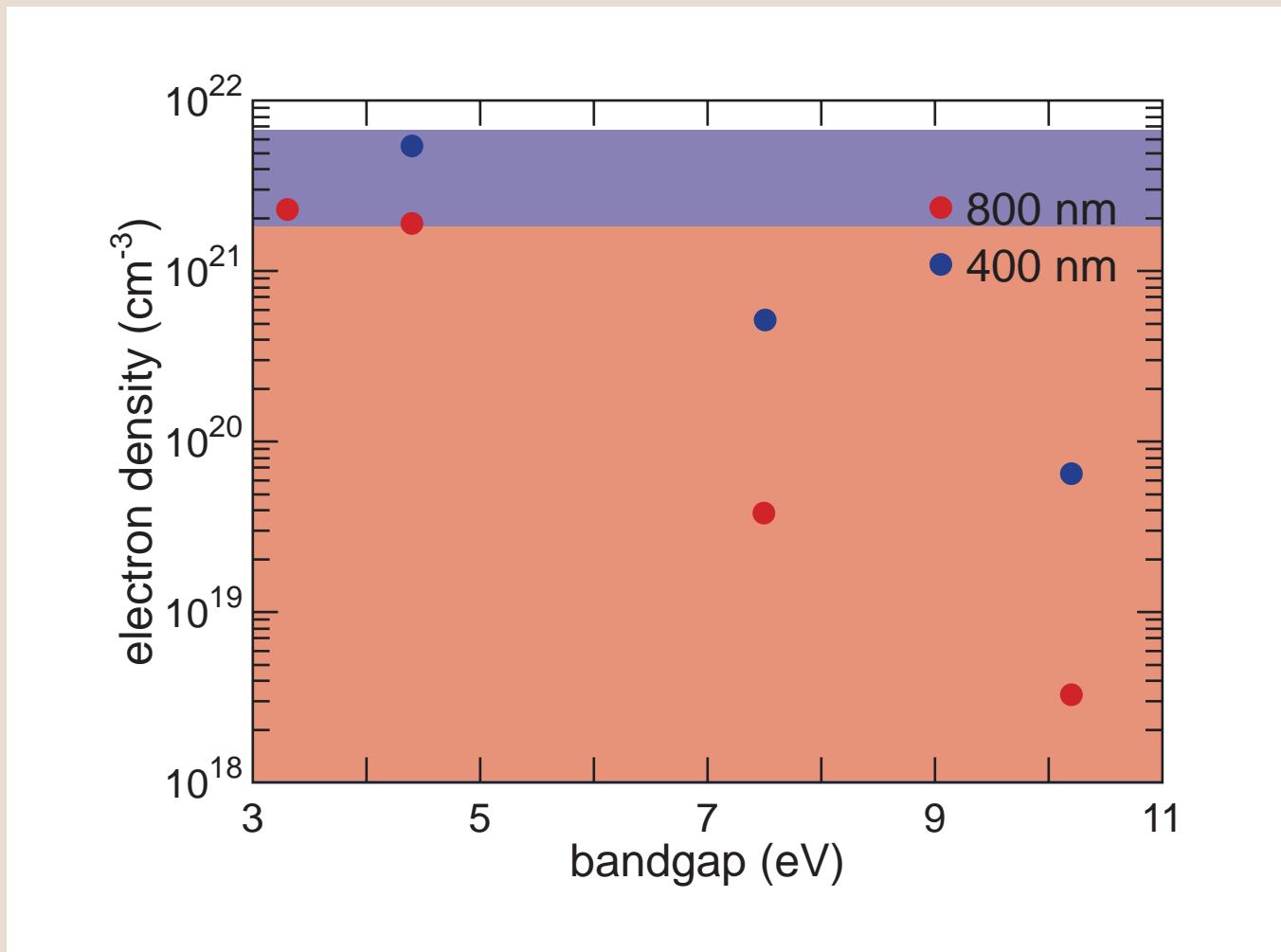


400 nm critical density



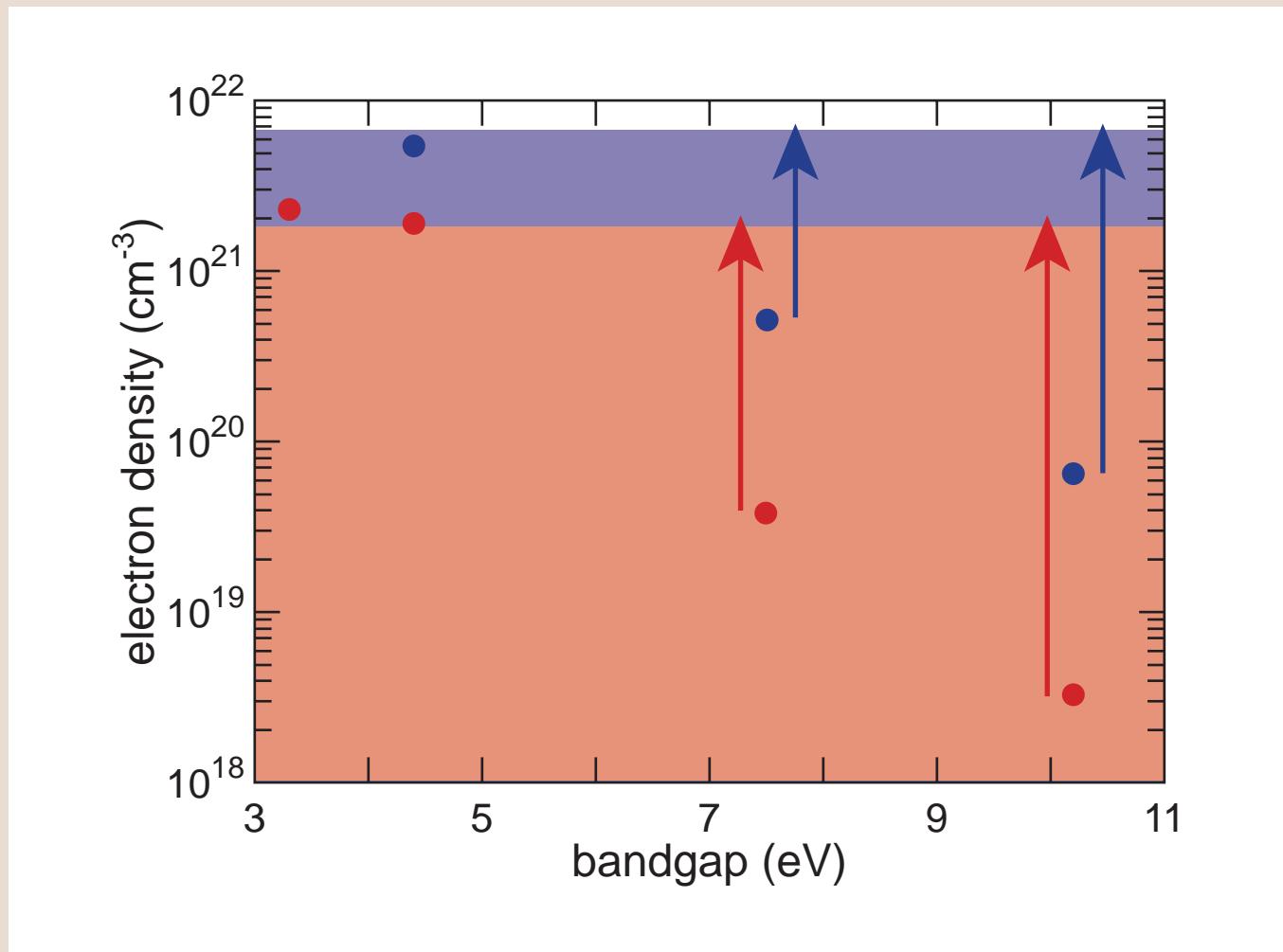
IONIZATION MECHANISMS

tunneling or MPI sufficient at low gap

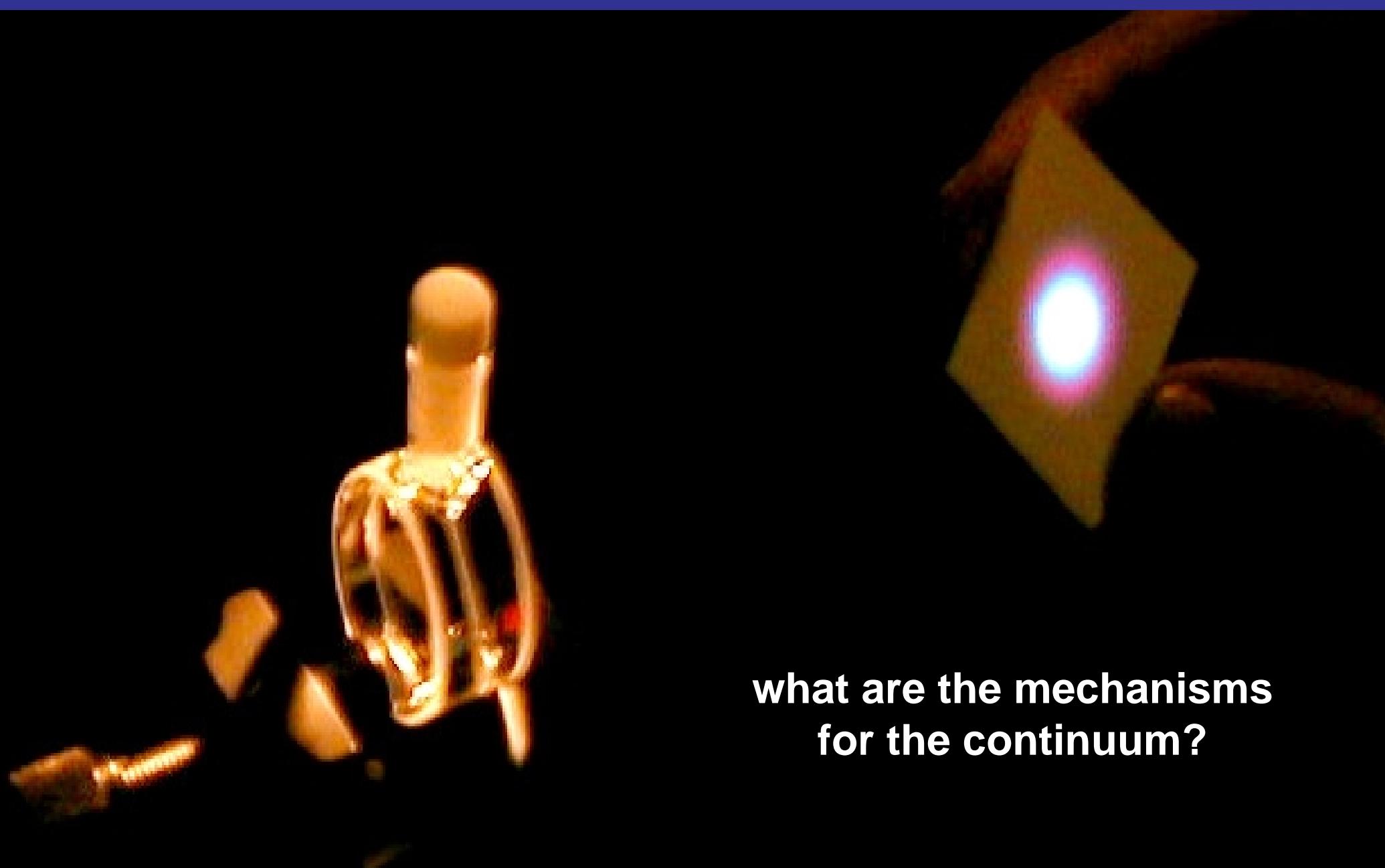


IONIZATION MECHANISMS

avalanche required at large gap



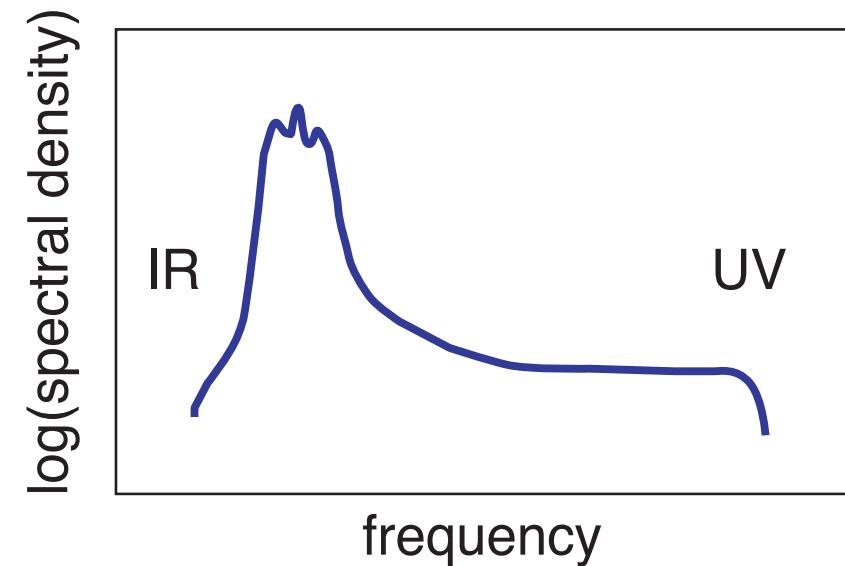
WHITE LIGHT GENERATION



**what are the mechanisms
for the continuum?**

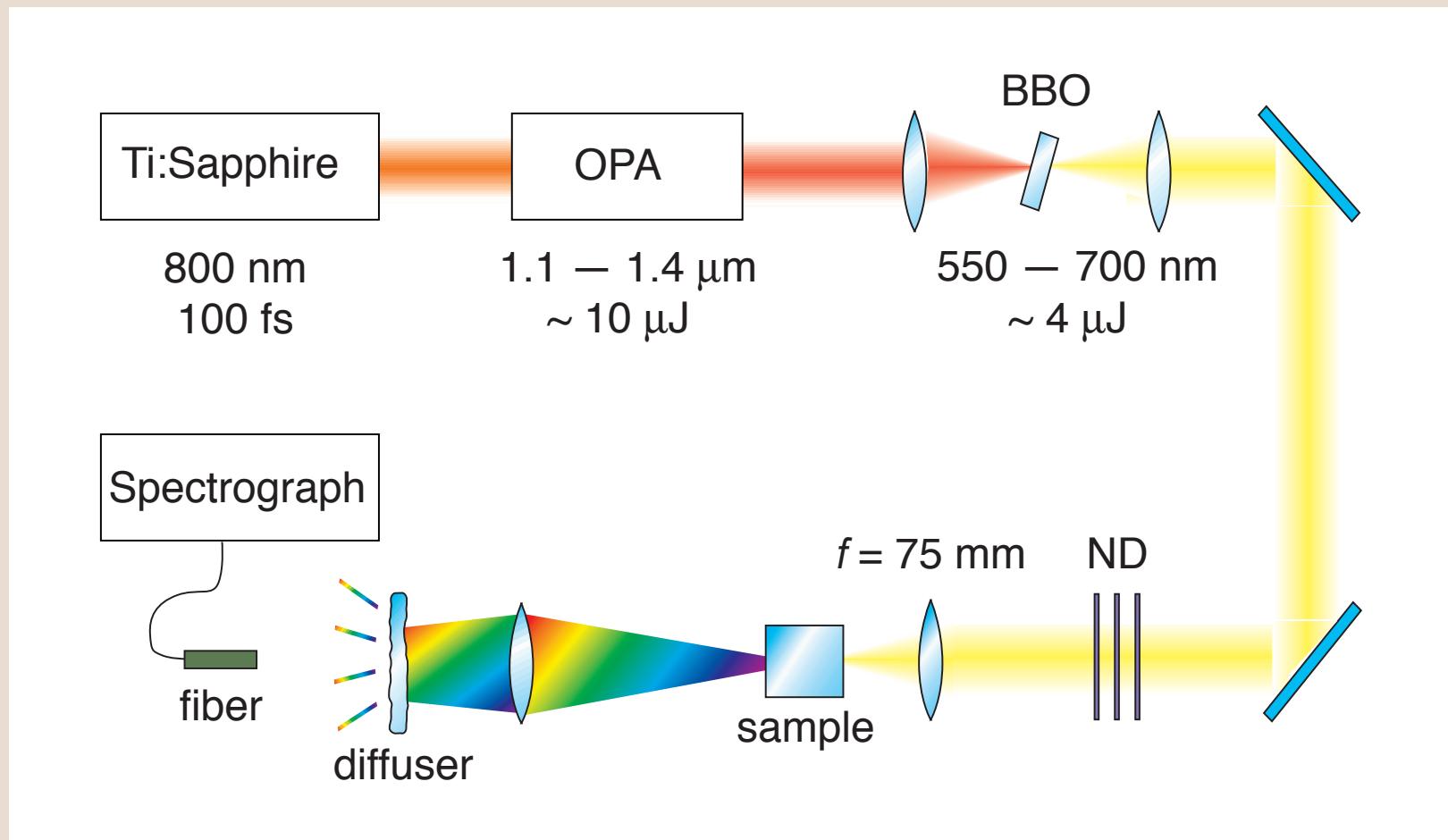
WHITE LIGHT GENERATION

dependence on material and laser parameters?



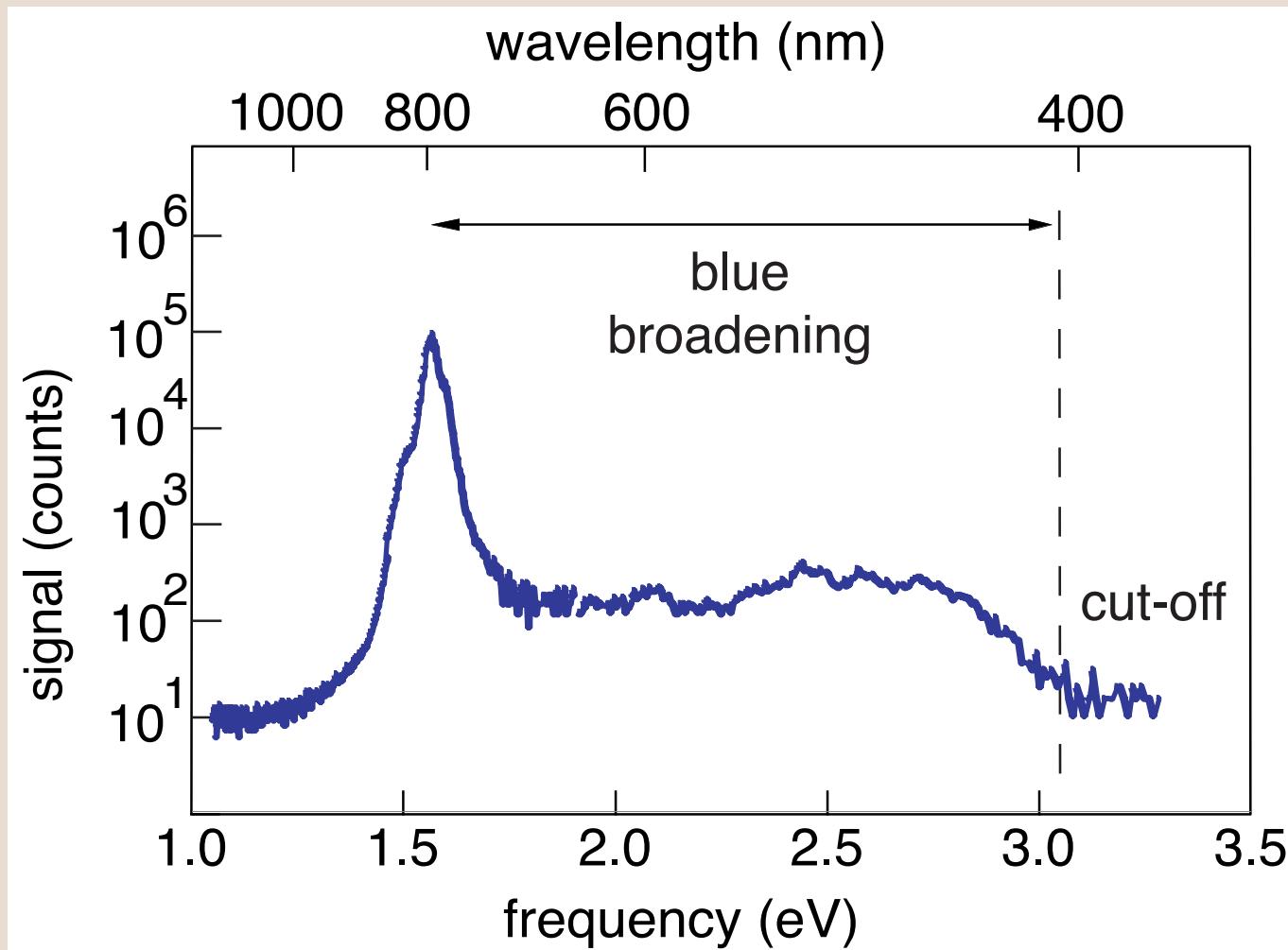
why the asymmetry?

WHITE LIGHT GENERATION



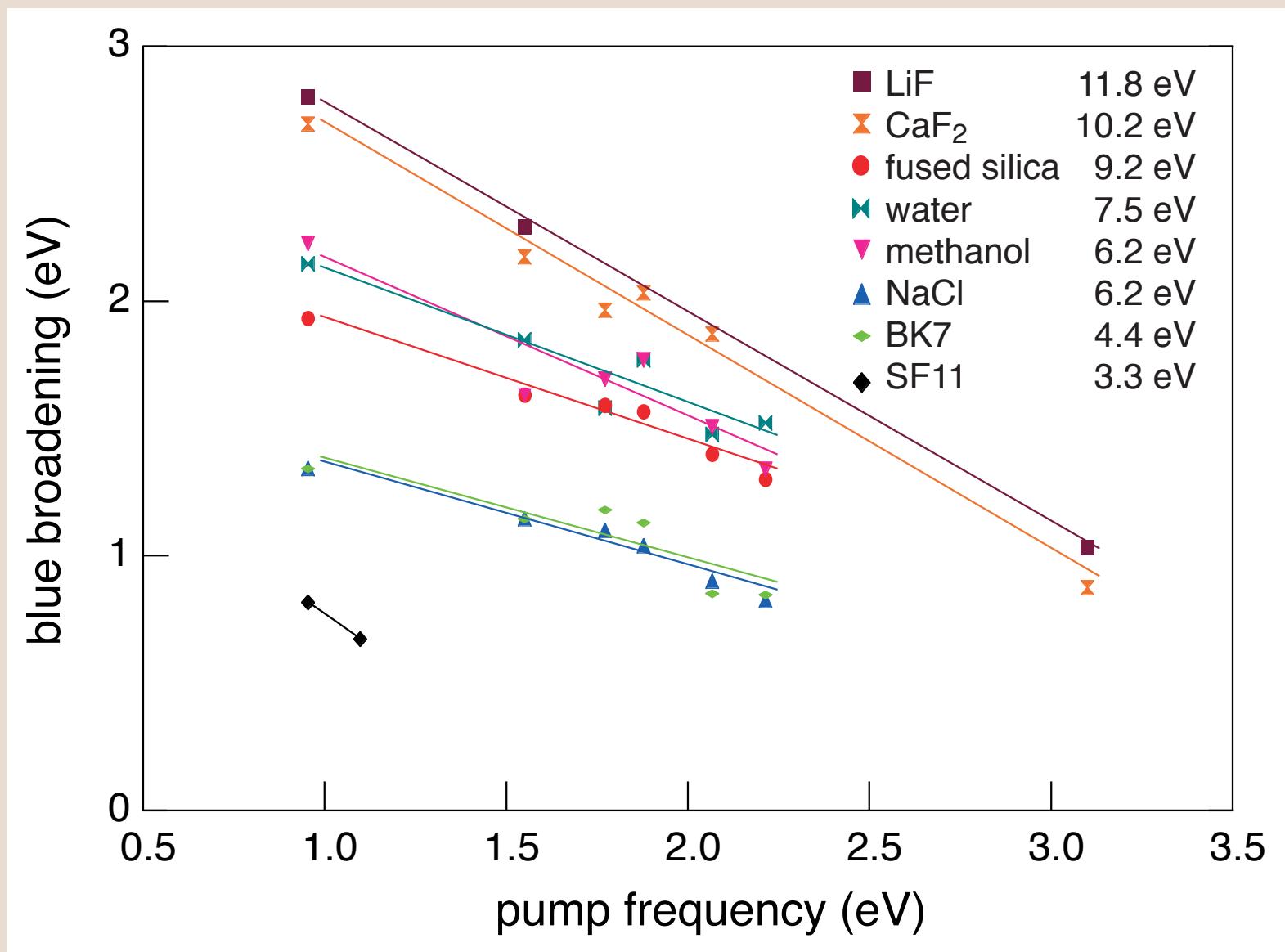
experimental setup

WHITE LIGHT GENERATION



continuum in fused silica produced by 800-nm, 100-fs pulses

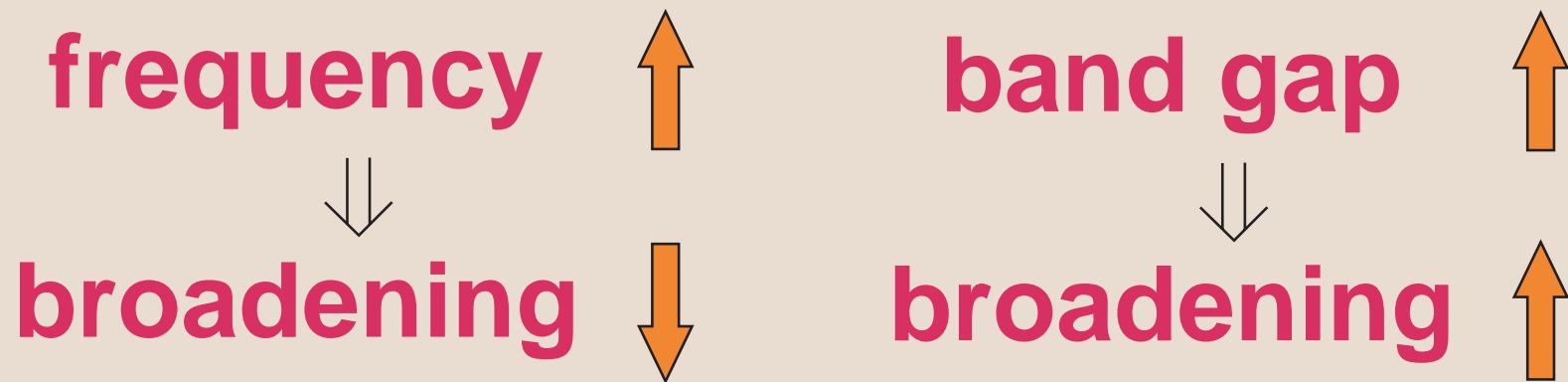
WHITE LIGHT GENERATION



vary pump wavelength and material bandgap

WHITE LIGHT GENERATION

two clear trends in blue broadening



also have to explain asymmetric spectrum

WHITE LIGHT GENERATION

proposed mechanism

**self focusing
increases laser intensity**

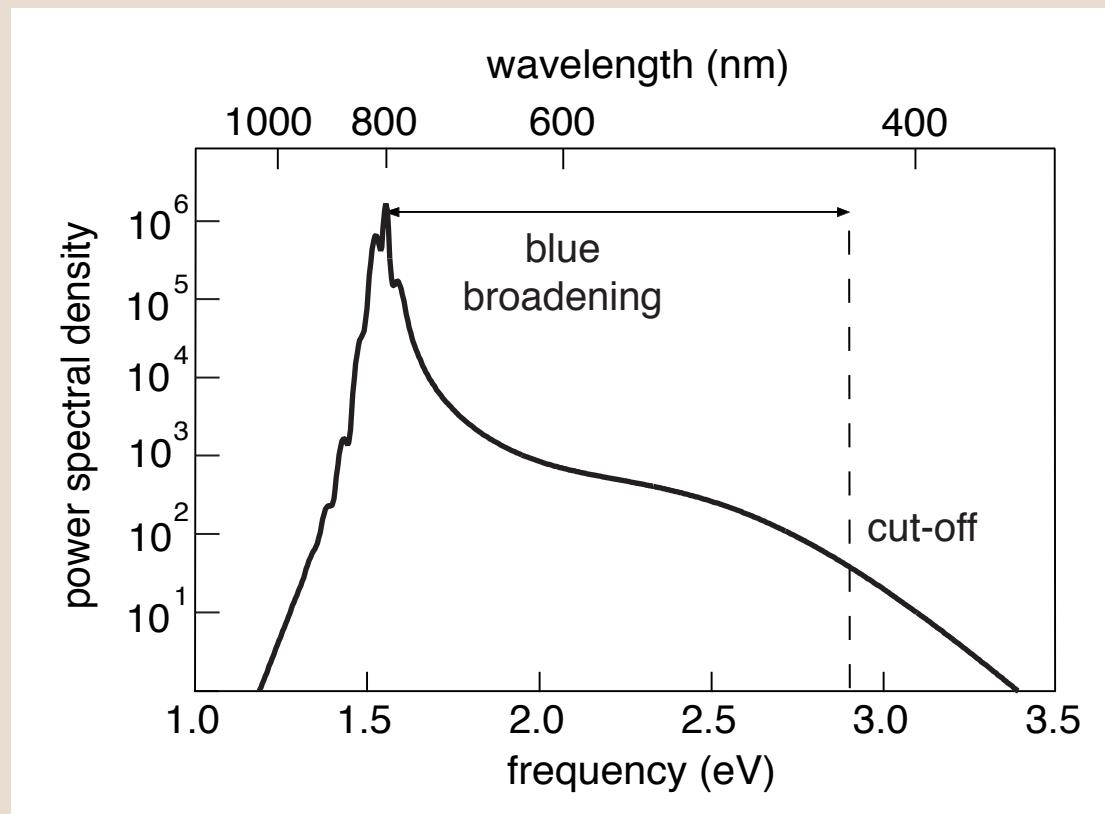
**SPM broadens spectrum
Self steepening occurs**

**asymmetric pulse produces
asymmetric SPM (blue shift)**

**extent of blue broadening
limited by ionization (plasma defocusing)**

WHITE LIGHT GENERATION

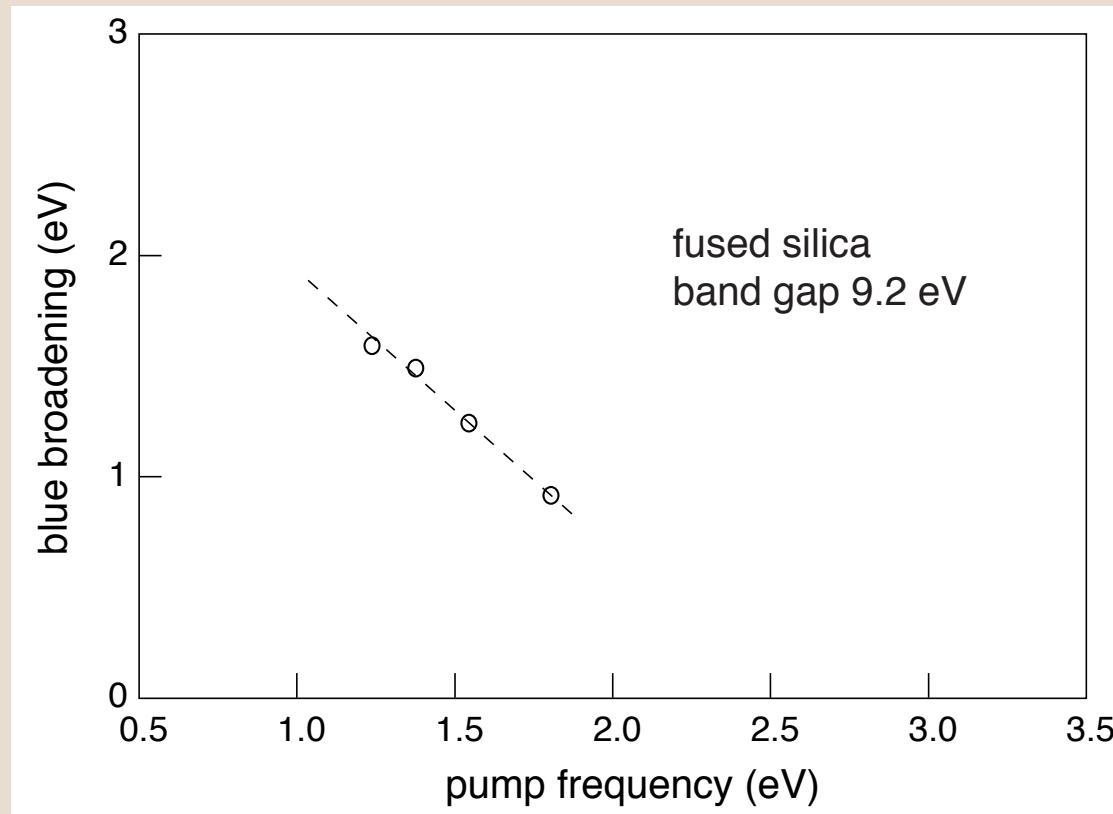
simulations (A. Gaeta, Cornell)



fused silica, 800 nm pump

WHITE LIGHT GENERATION

simulations (A. Gaeta, Cornell)



fused silica, pump frequency dependence

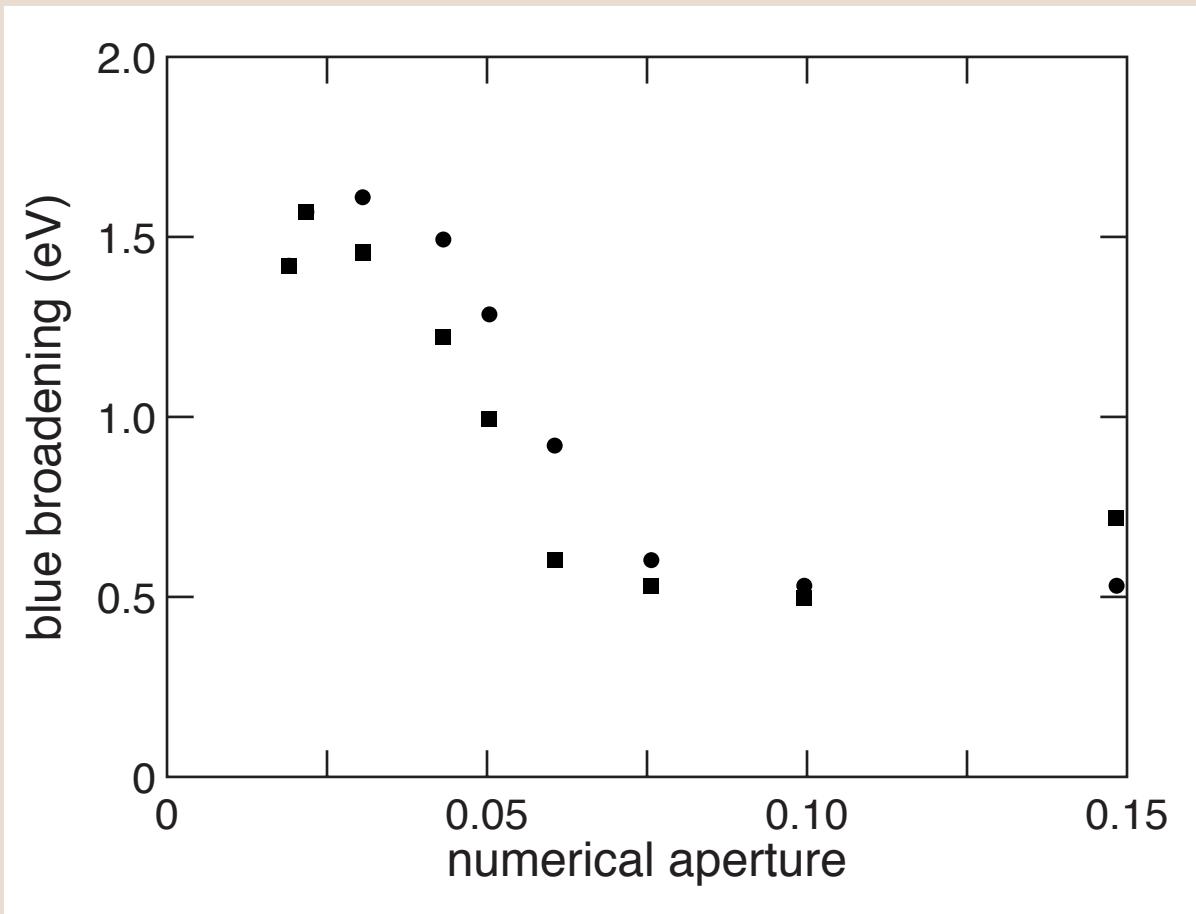
White light mechanisms

- Why no bulk damage?
- Focusing angle dependence of white light and damage threshold



FUTURE DIRECTIONS

**fused silica
60 fs
800 nm**



drop in blue broadening around 0.06 NA

White light mechanisms

- Why no bulk damage?
- Focusing angle dependence of white light and damage threshold

Ionization mechanisms

- Measure pulsedwidth dependence of threshold
- Model-independent determination of ionization rates

ACKNOWLEDGEMENTS

**A. Gaeta
Prof. N. Bloembergen**

National Science Foundation

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

<http://mazur-www.harvard.edu/>

focusing conditions

tight focusing

nonlinear
ionization
Intensity

structural
modification
Energy

slow focusing

self-focusing
Power

continuum
generation
Intensity

Femtosecond laser pulses in transparent materials 2: damage morphology and micromachining

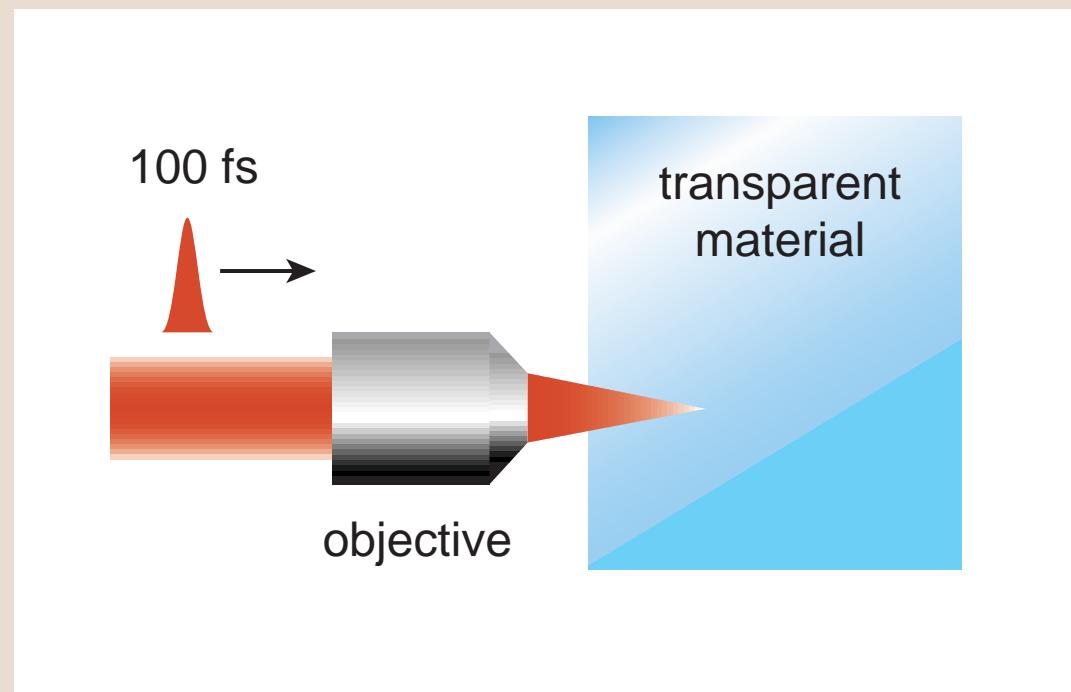
**Chris B. Schaffer
José F. Garcia
Alan O. Jamison
Nan Shen
Eric Mazur**



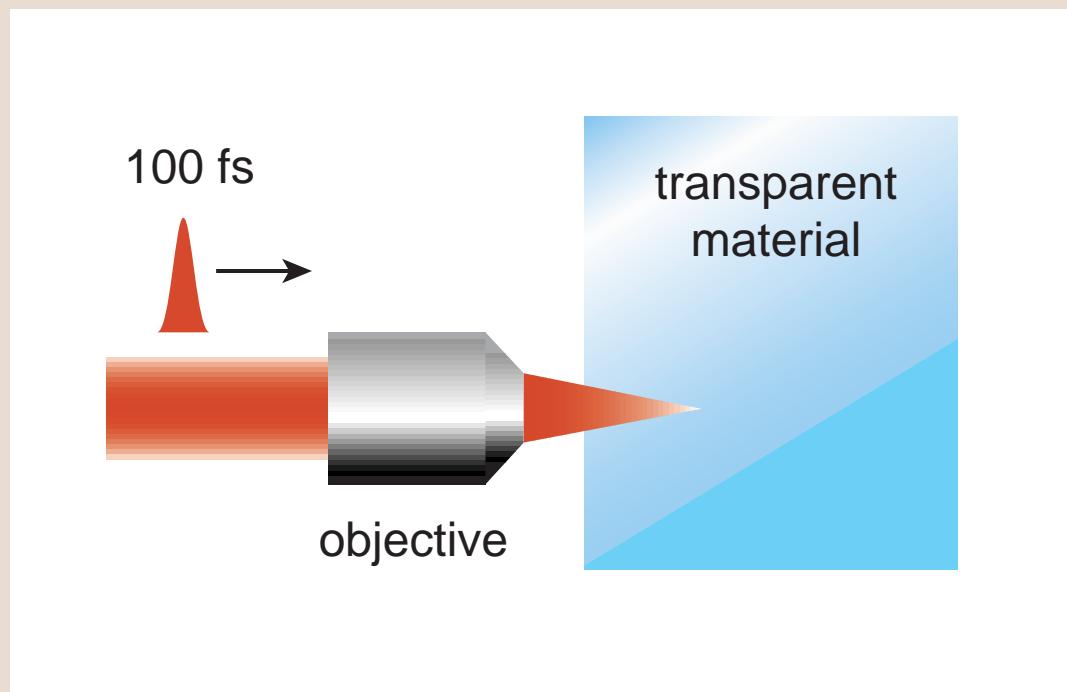
MRS Spring Meeting
April, 2001

Harvard University
Department of Physics

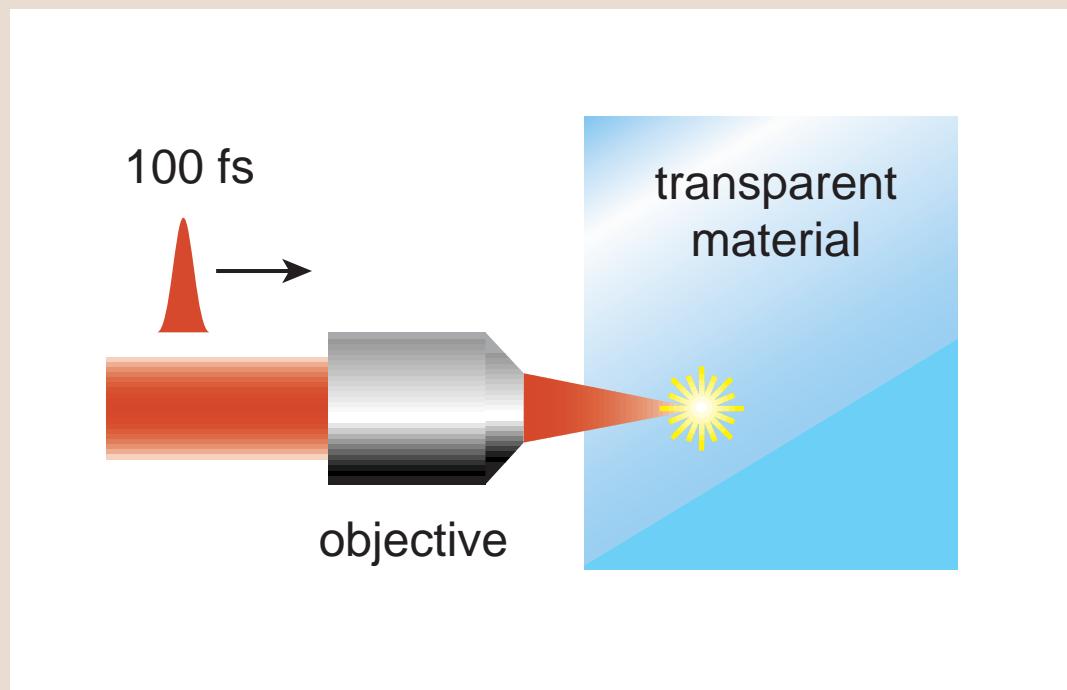
focus laser beam inside material



high intensity at focus

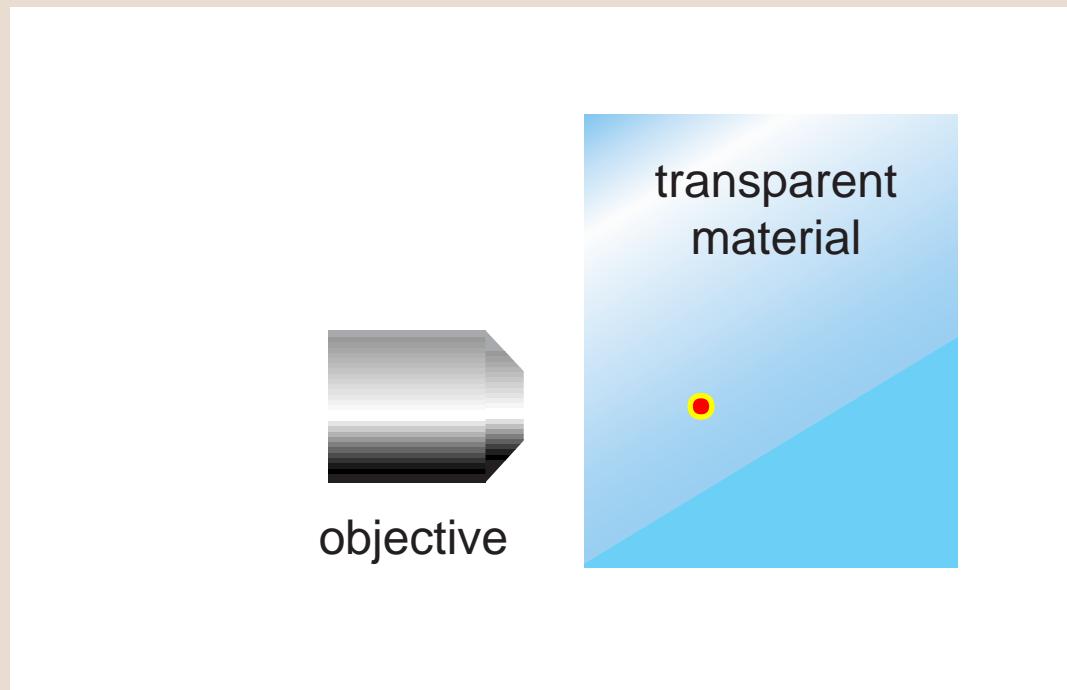


causes nonlinear ionization



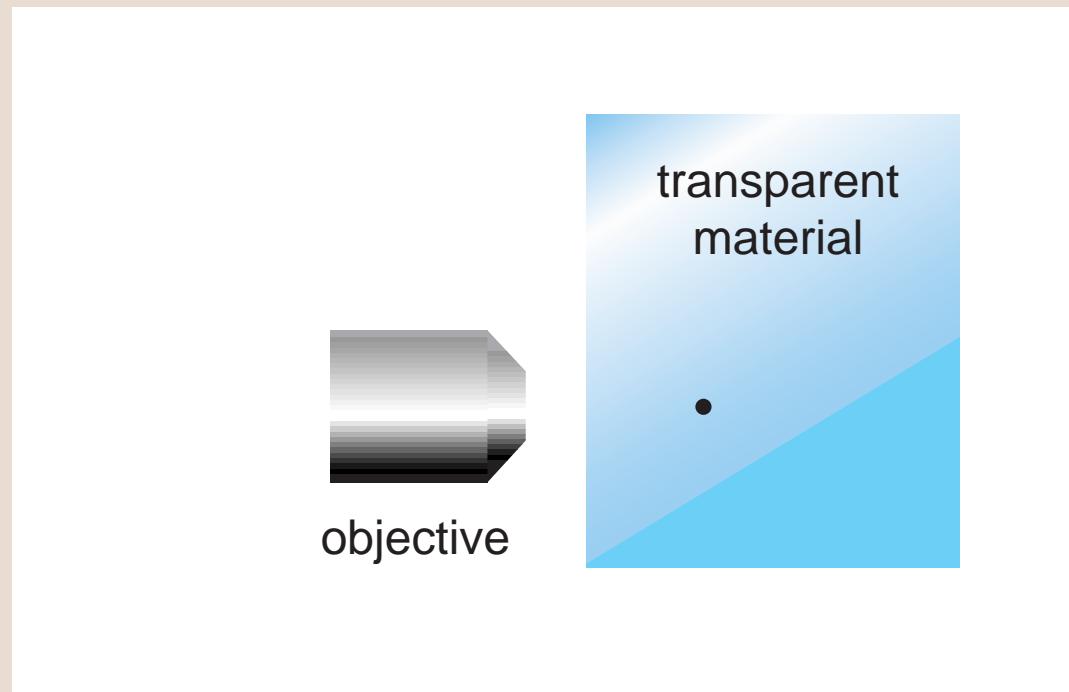
INTRODUCTION

energy is deposited in the focal volume



INTRODUCTION

producing microscopic **bulk** damage

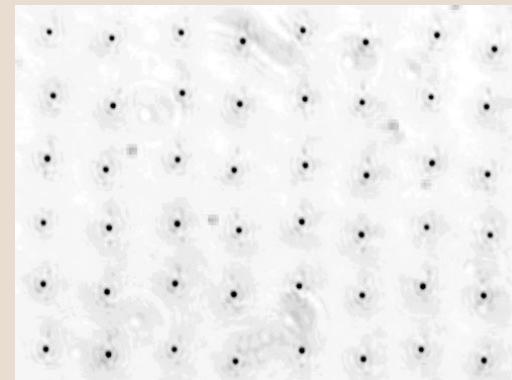


with only tens of **nanojoules**!

INTRODUCTION

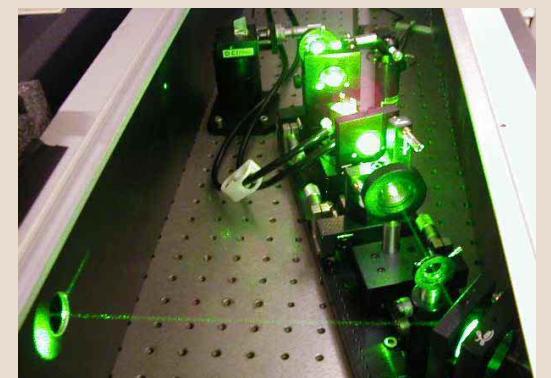
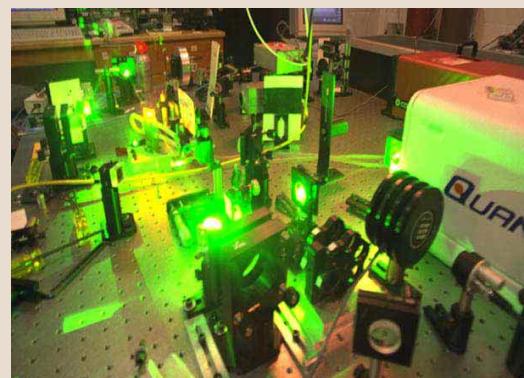
why **bulk?**

three-dimensional micromachining

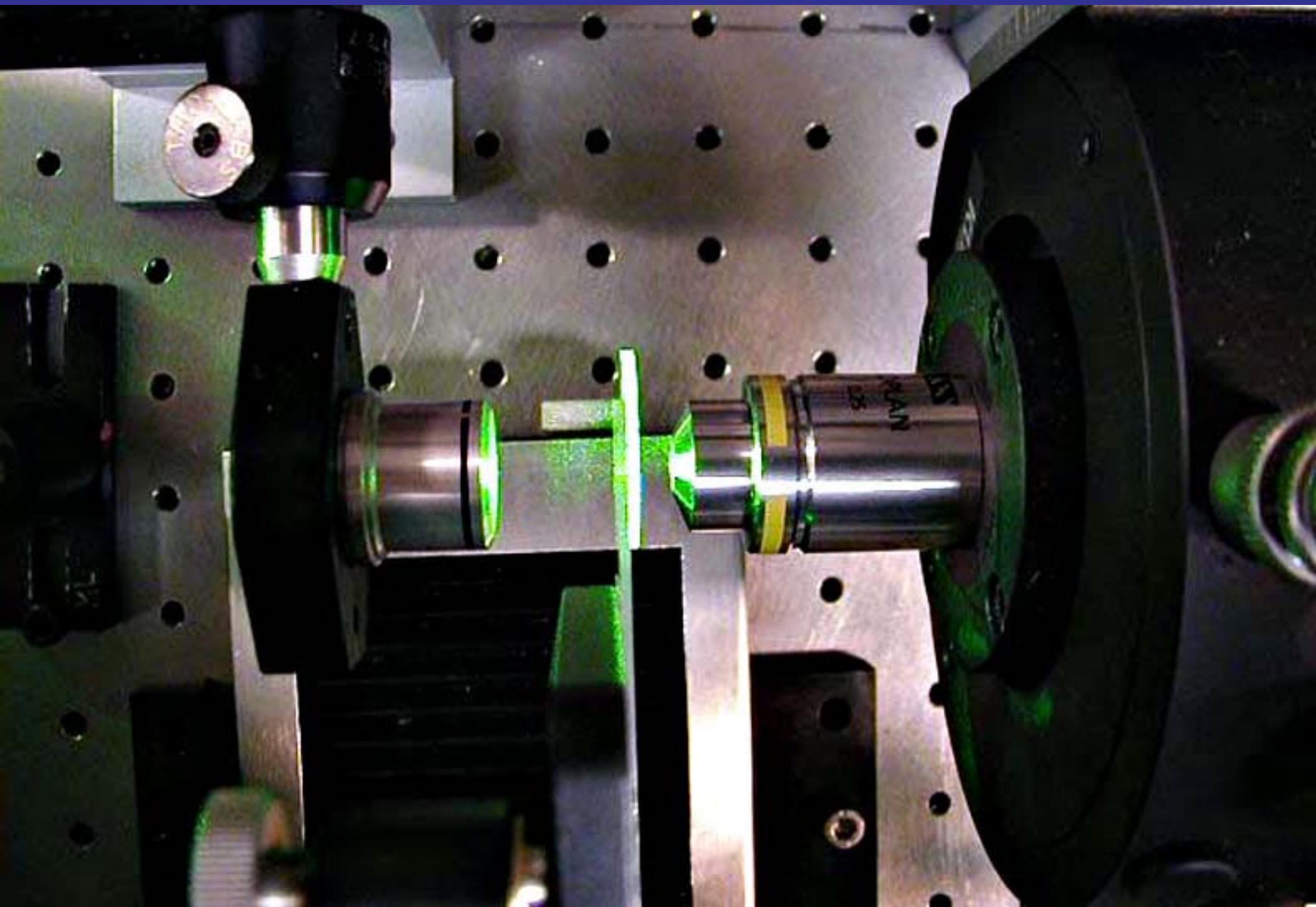


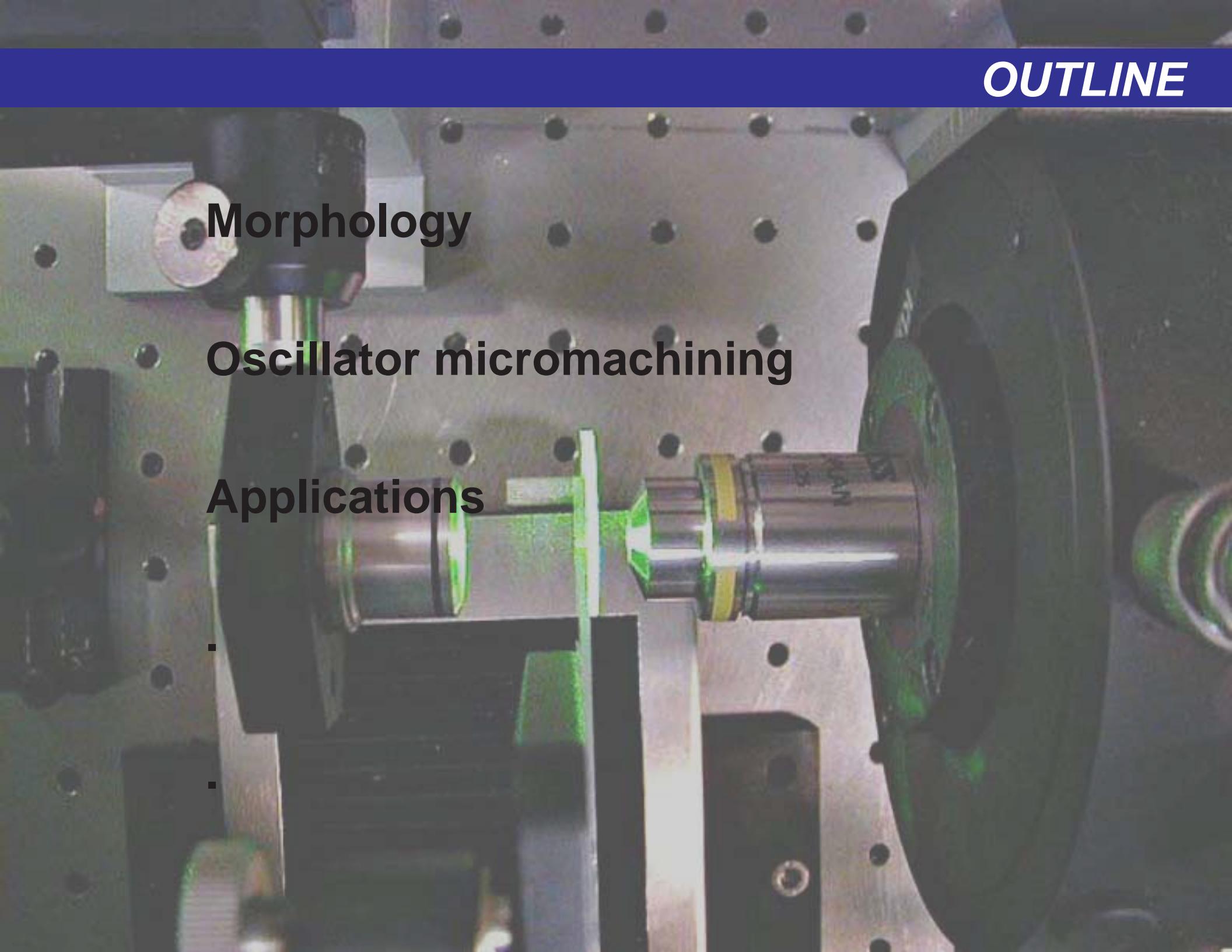
why **nanojoules?**

non-amplified micromachining



OUTLINE



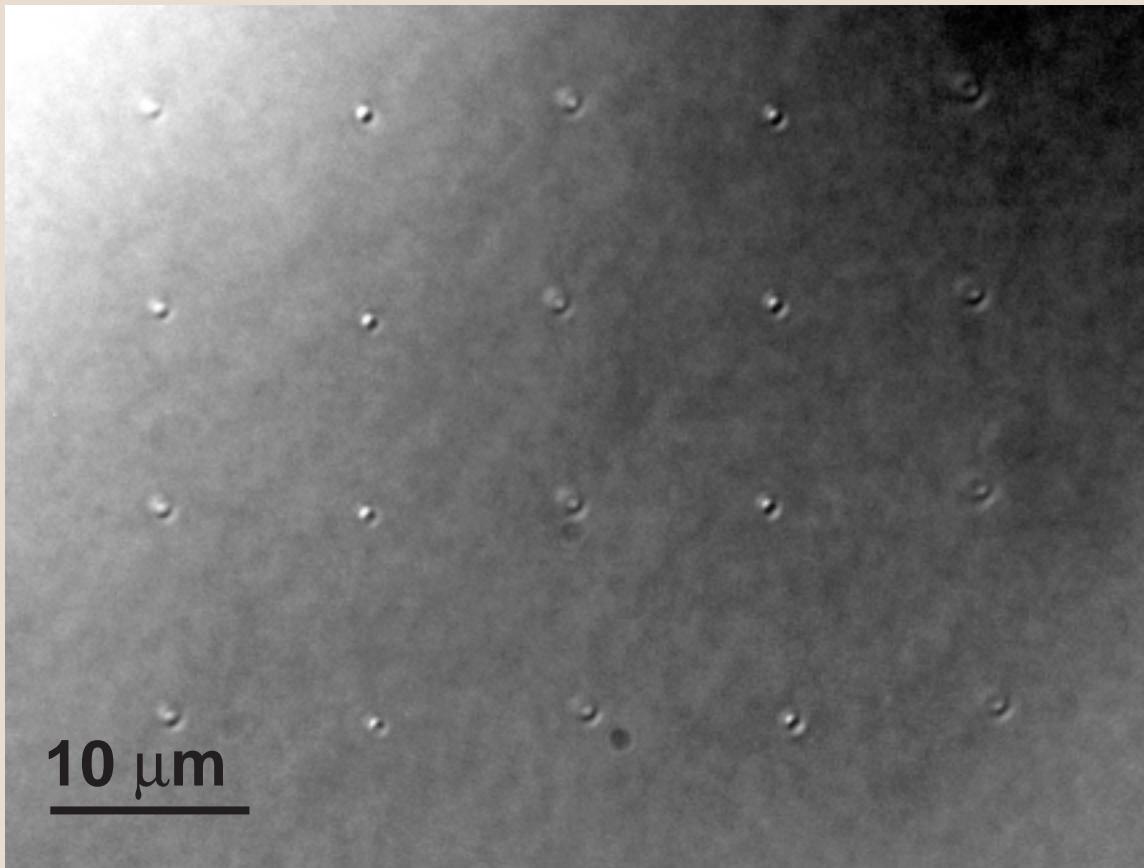


Morphology

Oscillator micromachining

Applications

MORPHOLOGY

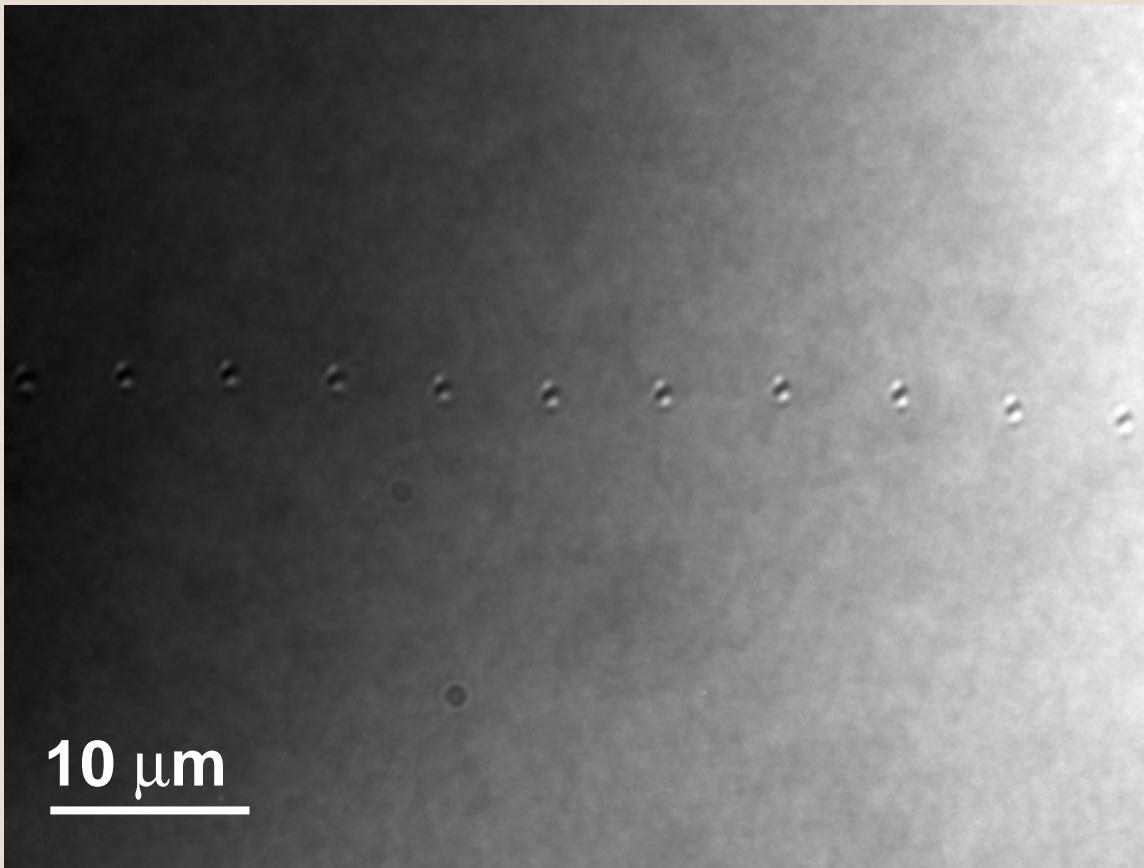


10 nJ
100 fs
1.4 NA
Corning 0211

\otimes^k

top view

MORPHOLOGY



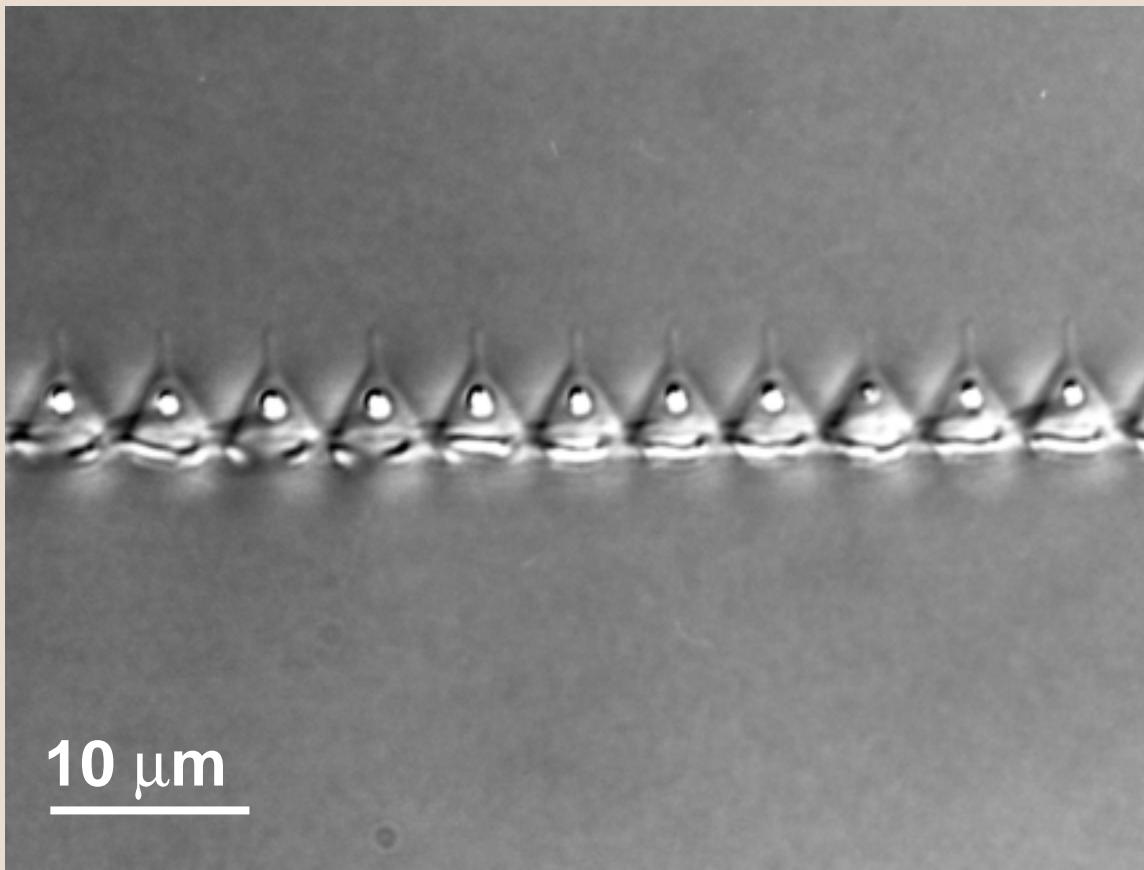
15 nJ
100 fs
1.4 NA
Corning 0211

10 μm

↑*k*

side view

MORPHOLOGY



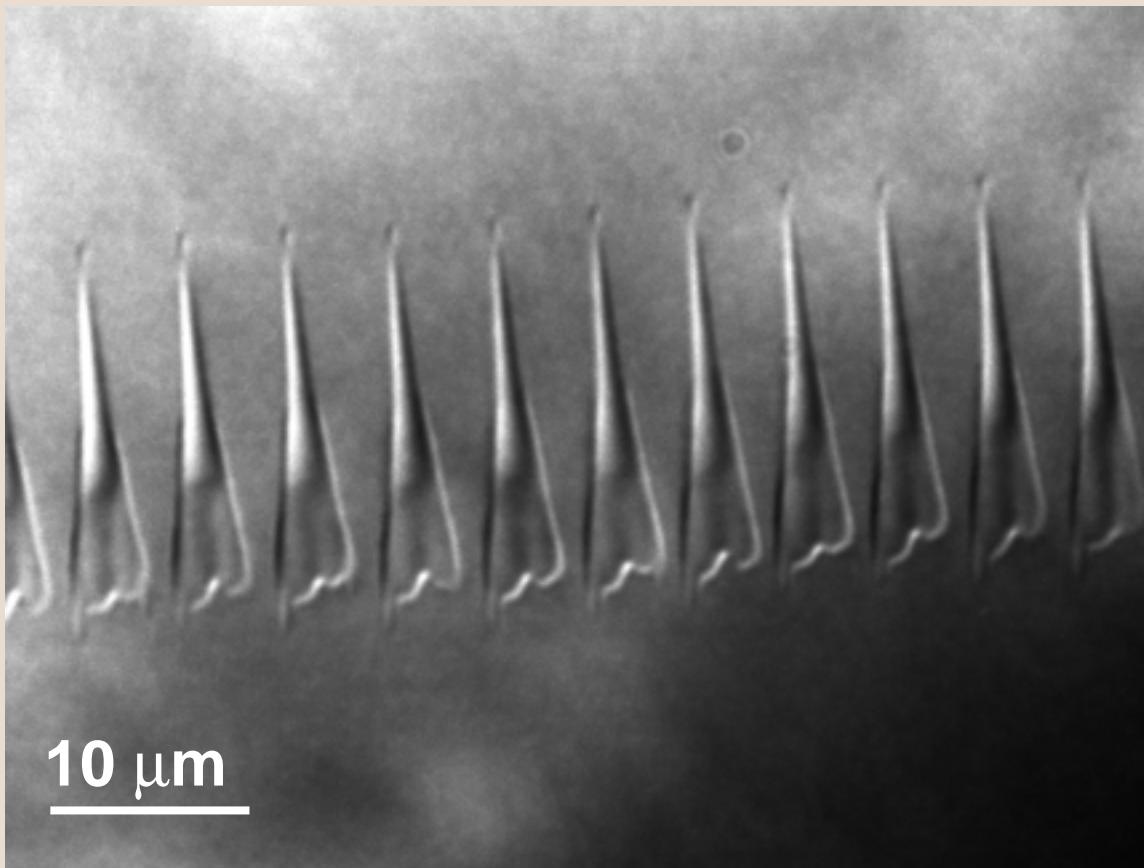
500 nJ
100 fs
1.4 NA
Corning 0211

10 μm

↑
k

higher pulse energy

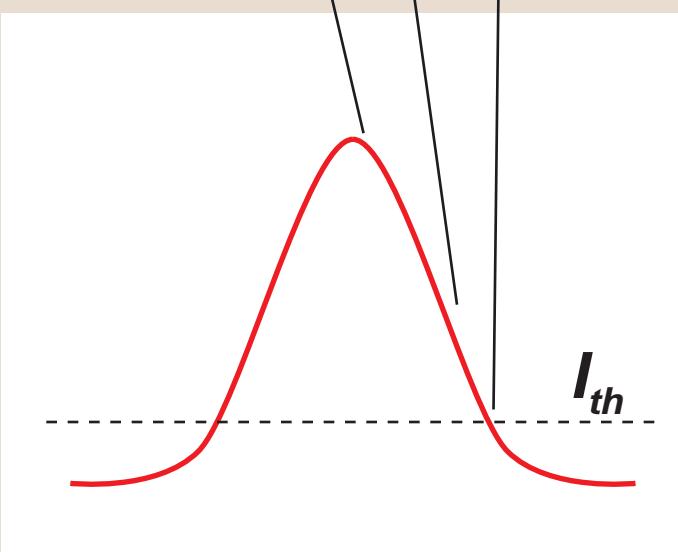
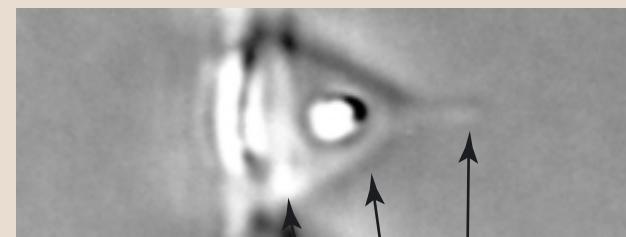
MORPHOLOGY



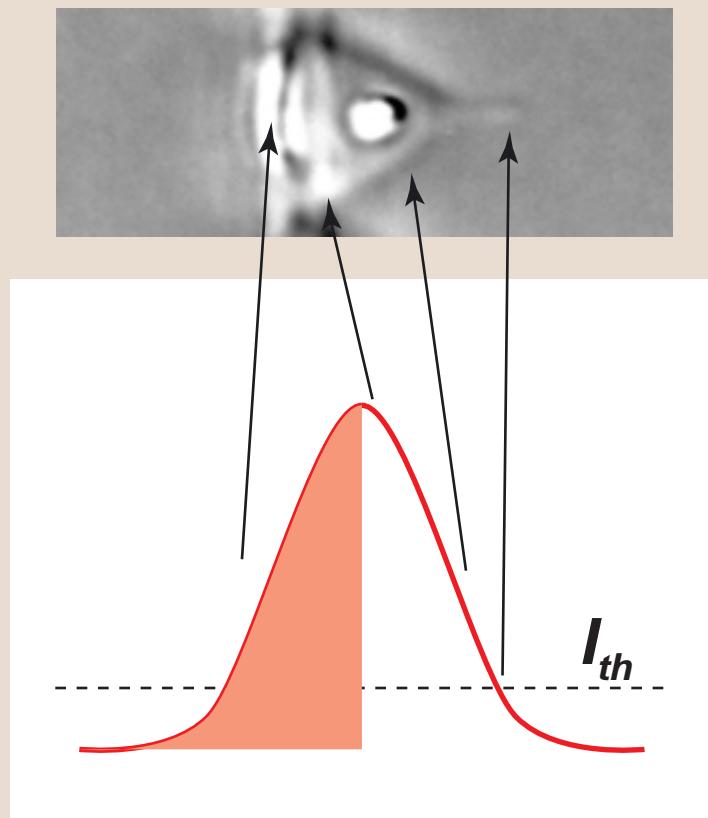
500 nJ
100 fs
0.45 NA
Corning 0211

slower focusing

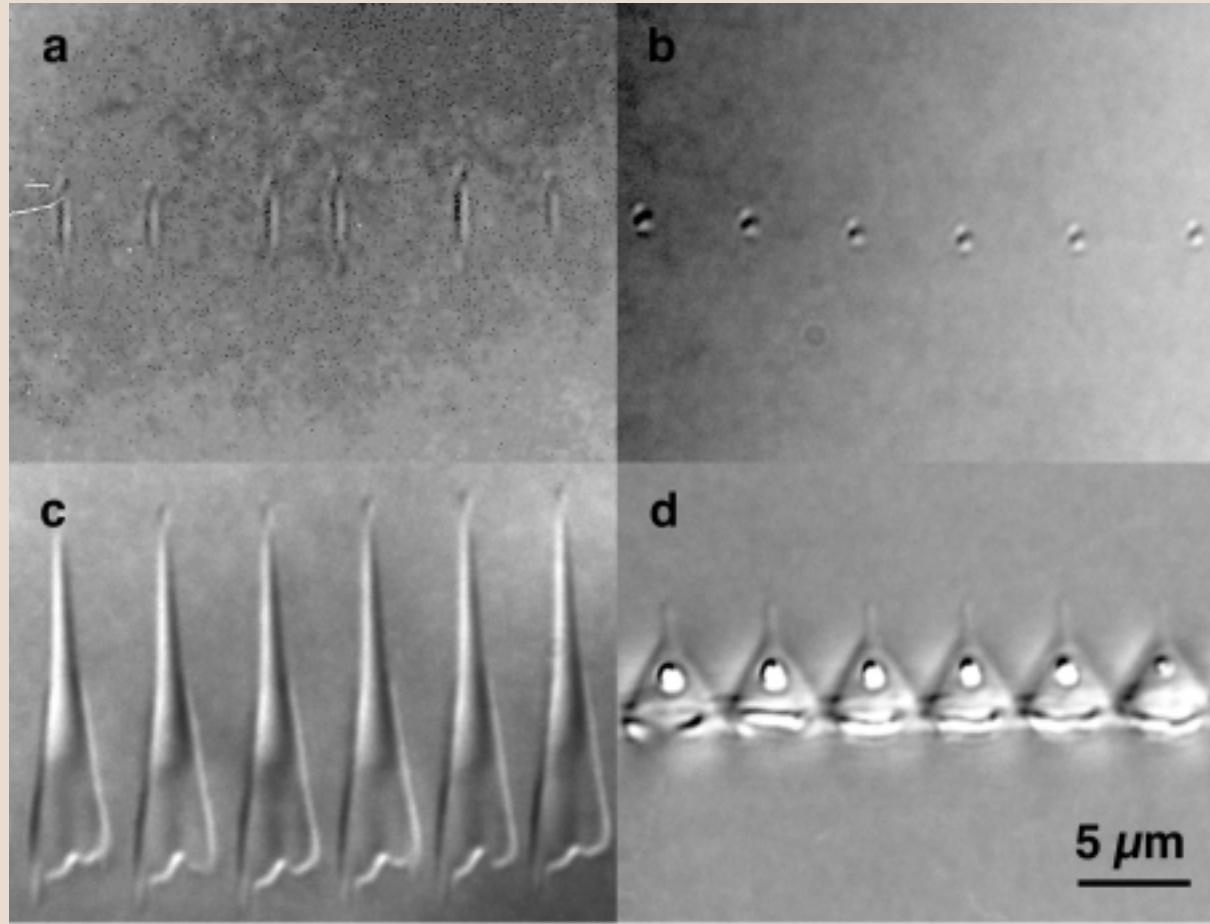
cone formation mechanism



cone formation mechanism



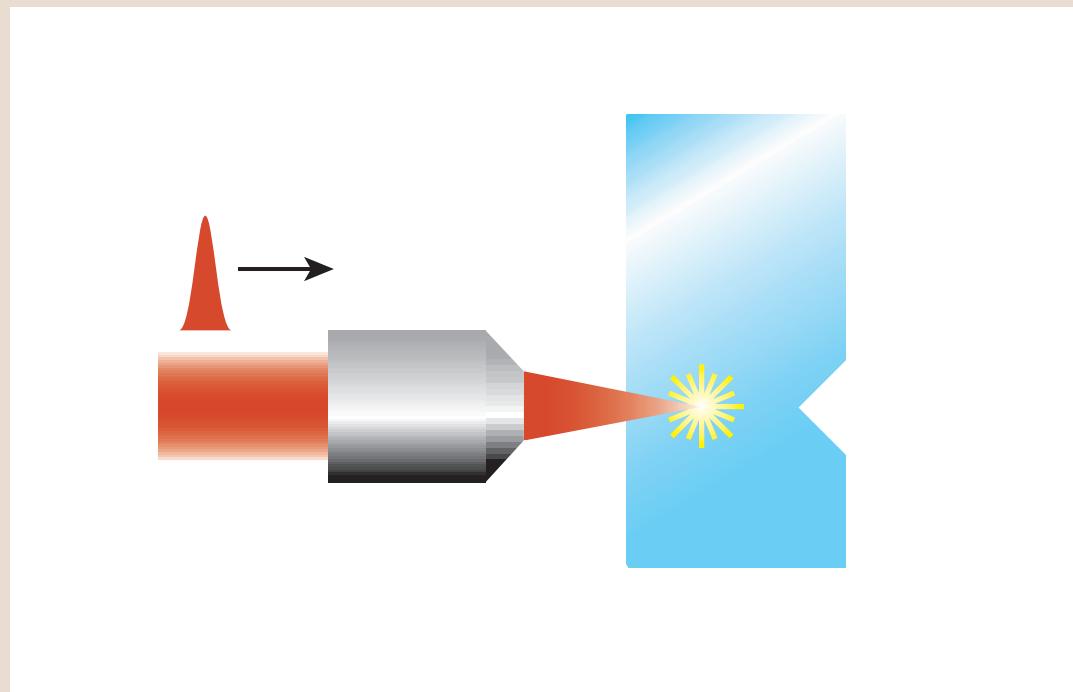
MORPHOLOGY



a: 0.45 NA, 50 nJ
b: 1.4 NA, 15 nJ
c: 0.45 NA, 500 nJ
d: 1.4 NA, 500 nJ

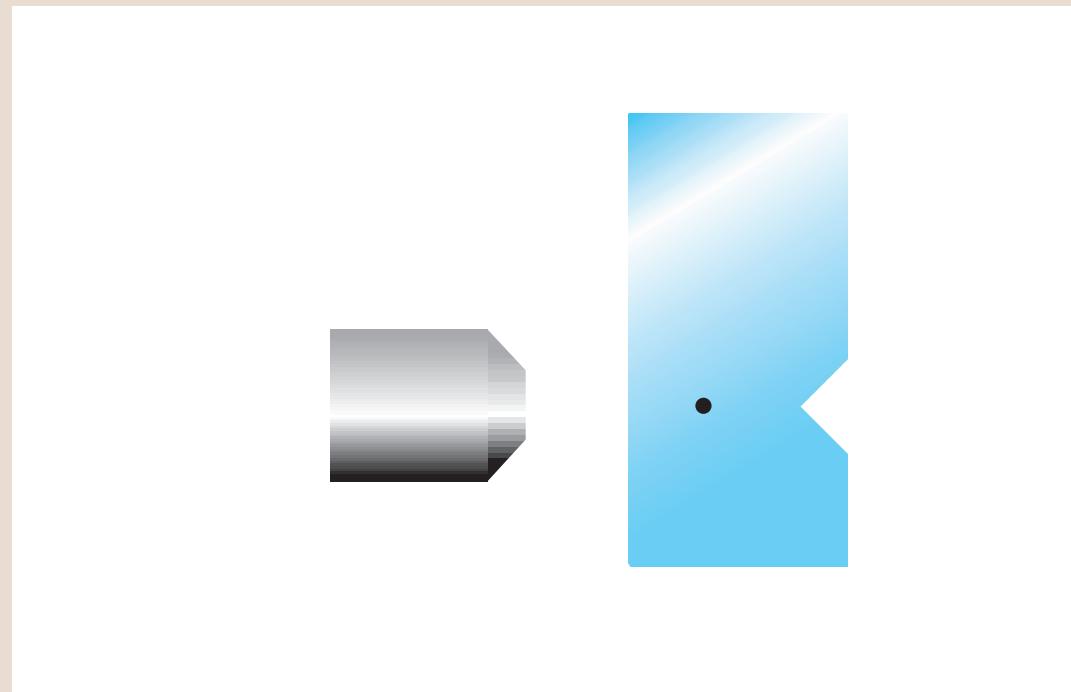
best micromachining precision near threshold

SEM sample preparation



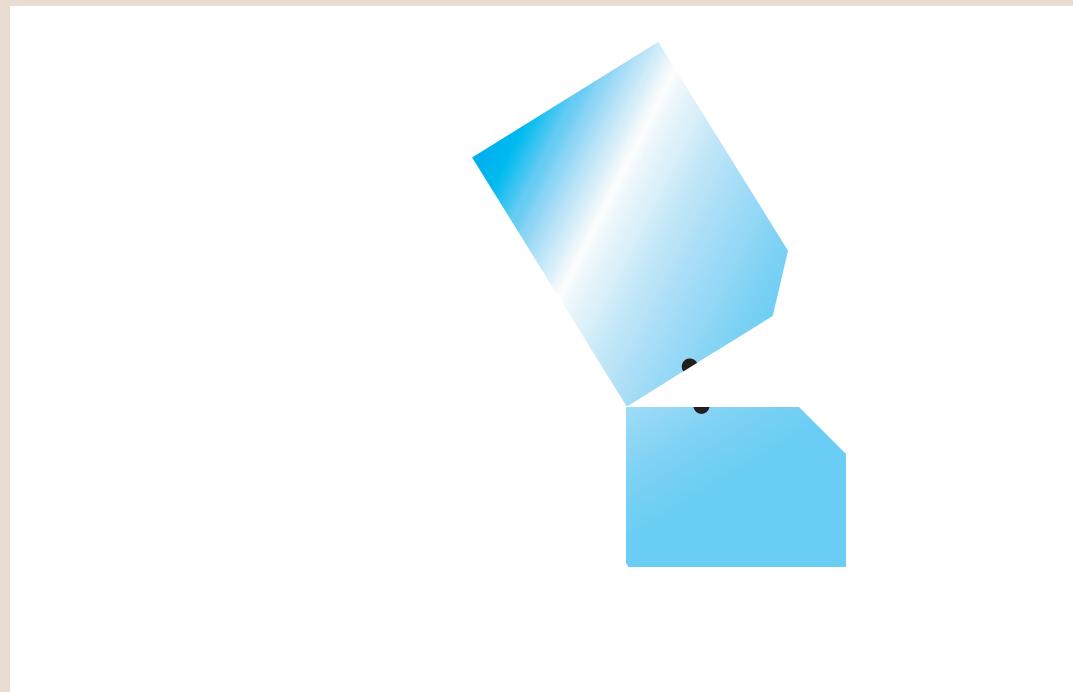
irradiate scribed sample

SEM sample preparation



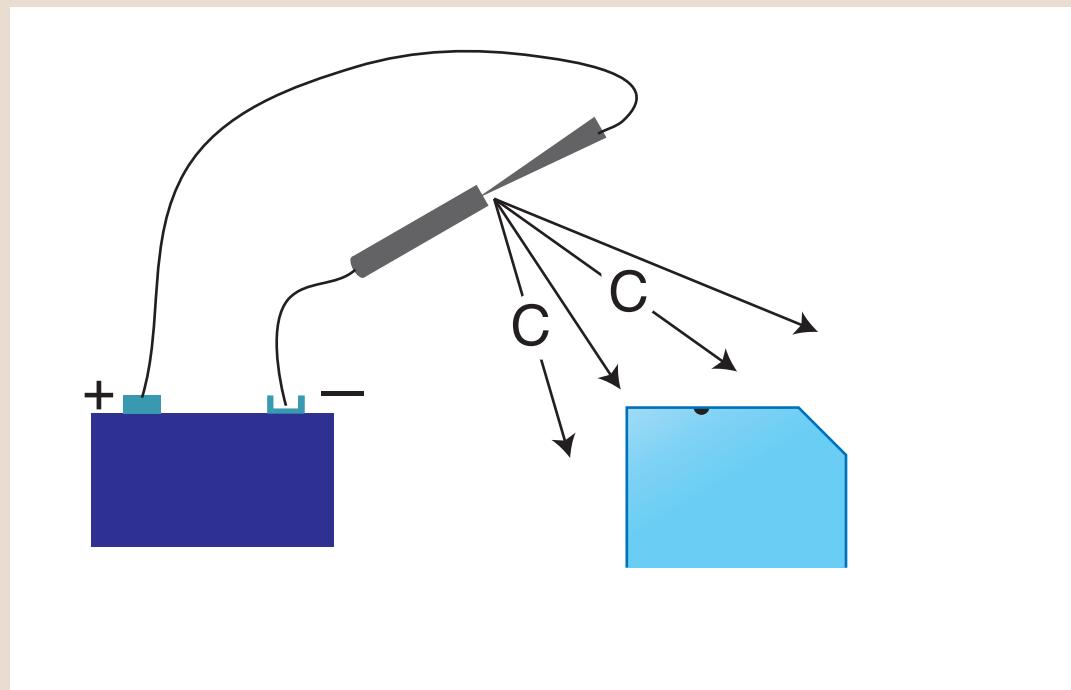
dense pattern of structures above scribe line

SEM sample preparation



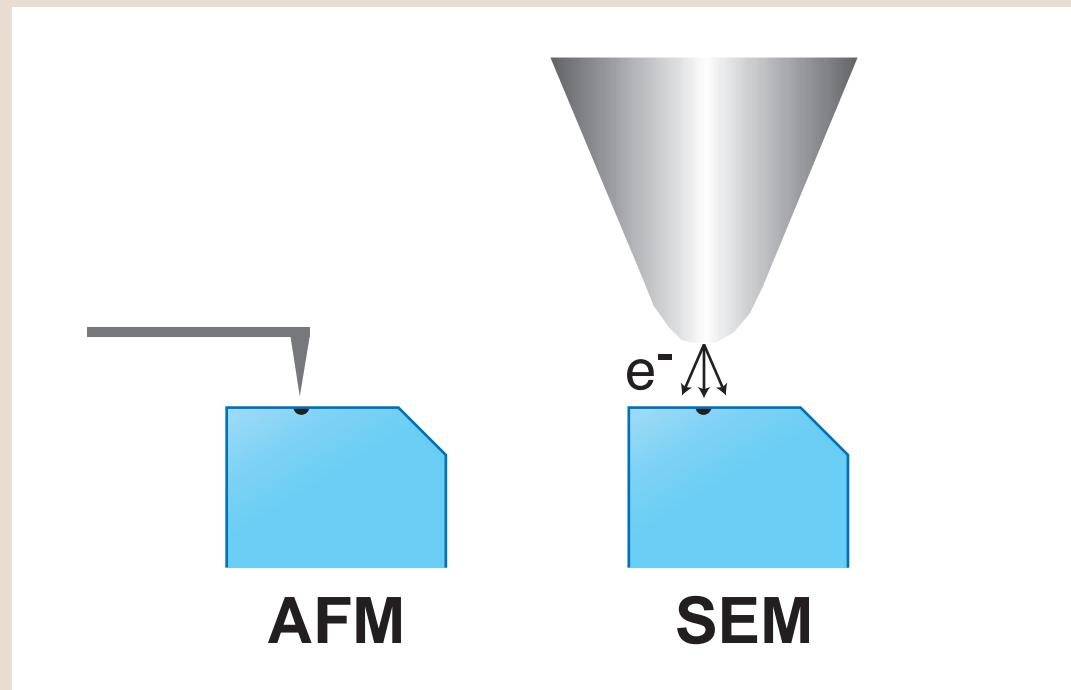
fracture samples to bring some structures to surface

SEM sample preparation



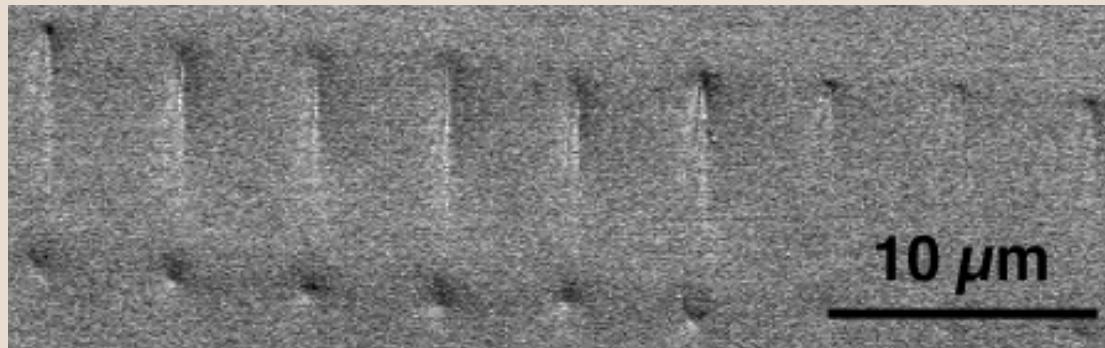
coat with graphite

SEM sample preparation

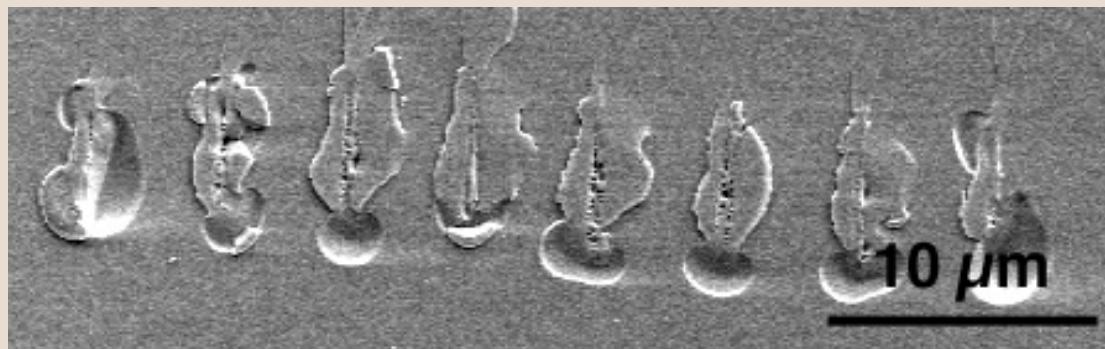


image

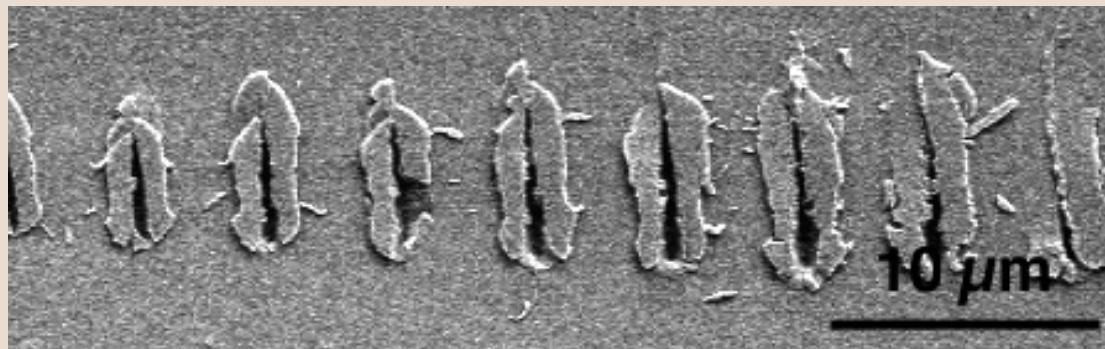
MORPHOLOGY



140 nJ



250 nJ

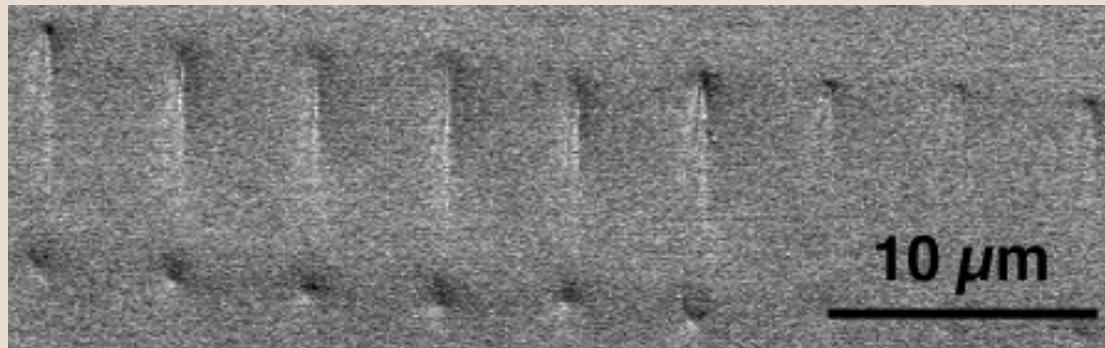


540 nJ
↑
k

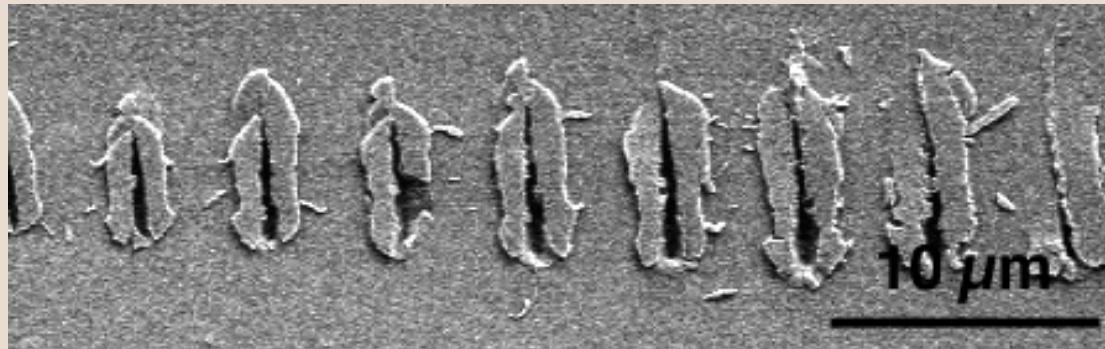
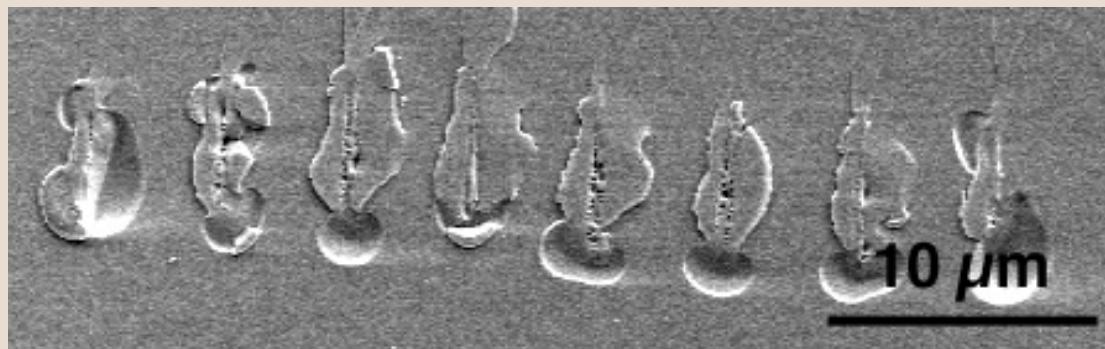
100 fs
800 nm
0.45 NA
Corning 0211

side view SEM

MORPHOLOGY



thermal ?
mechanism

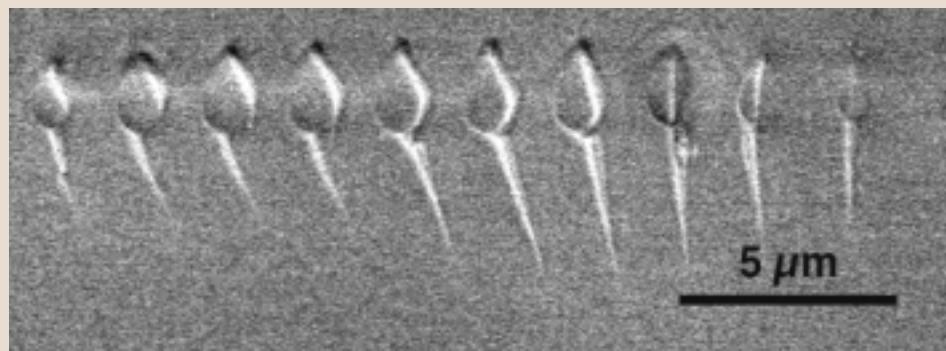


explosive
mechanism

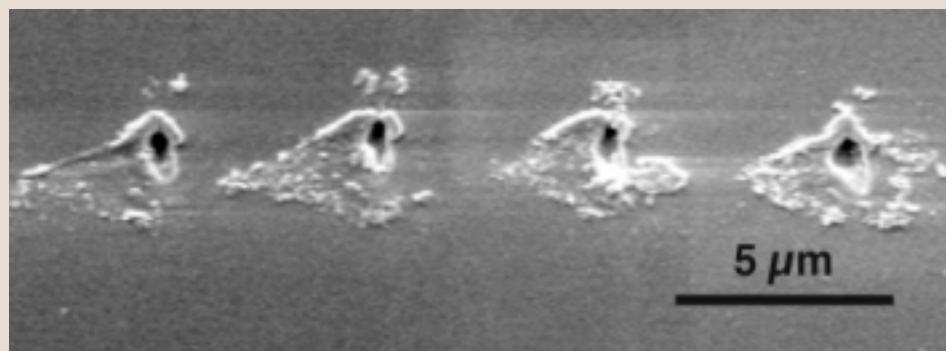
100 fs
800 nm
0.45 NA
Corning 0211

side view SEM

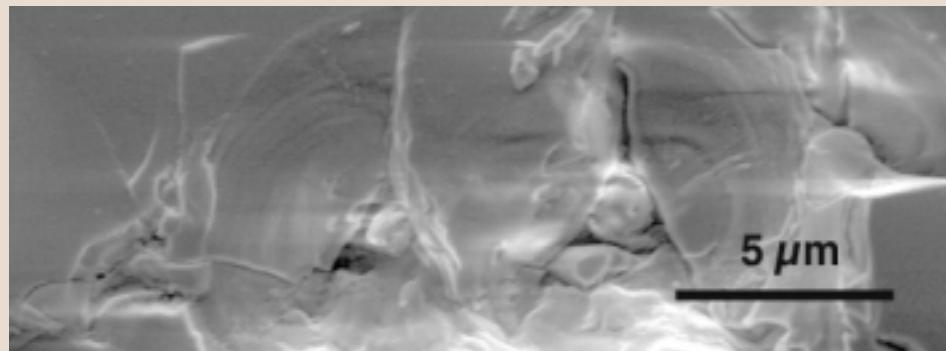
MORPHOLOGY



36 nJ



140 nJ



↑ k
500 nJ

100 fs
800 nm
1.4 NA
Corning 0211

tighter focusing

MORPHOLOGY

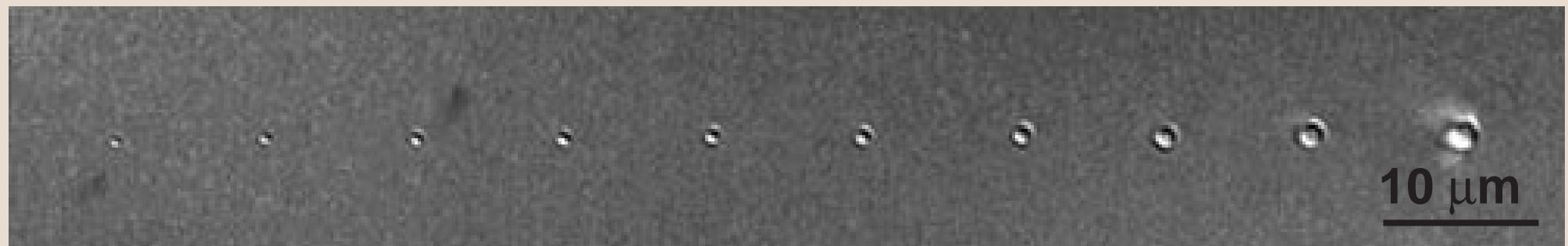
vary laser energy

6.6 nJ

13 nJ

33 nJ

66 nJ



100 fs
1.4 NA
Corning 0211

\otimes^k

MORPHOLOGY

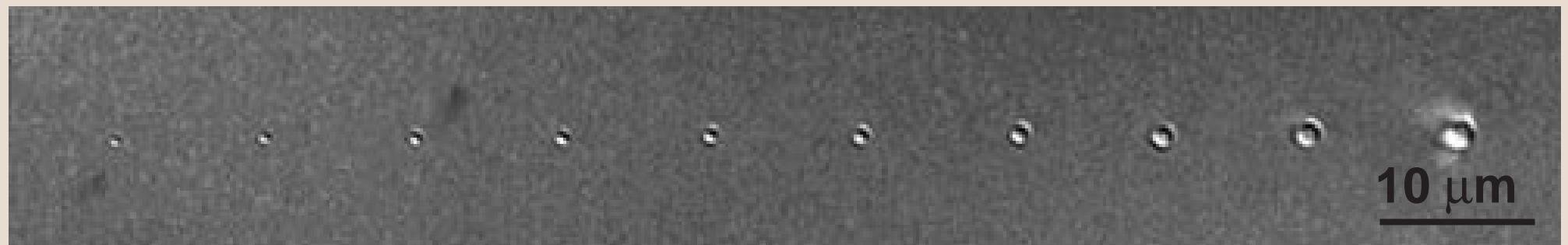
vary laser energy

6.6 nJ

13 nJ

33 nJ

66 nJ

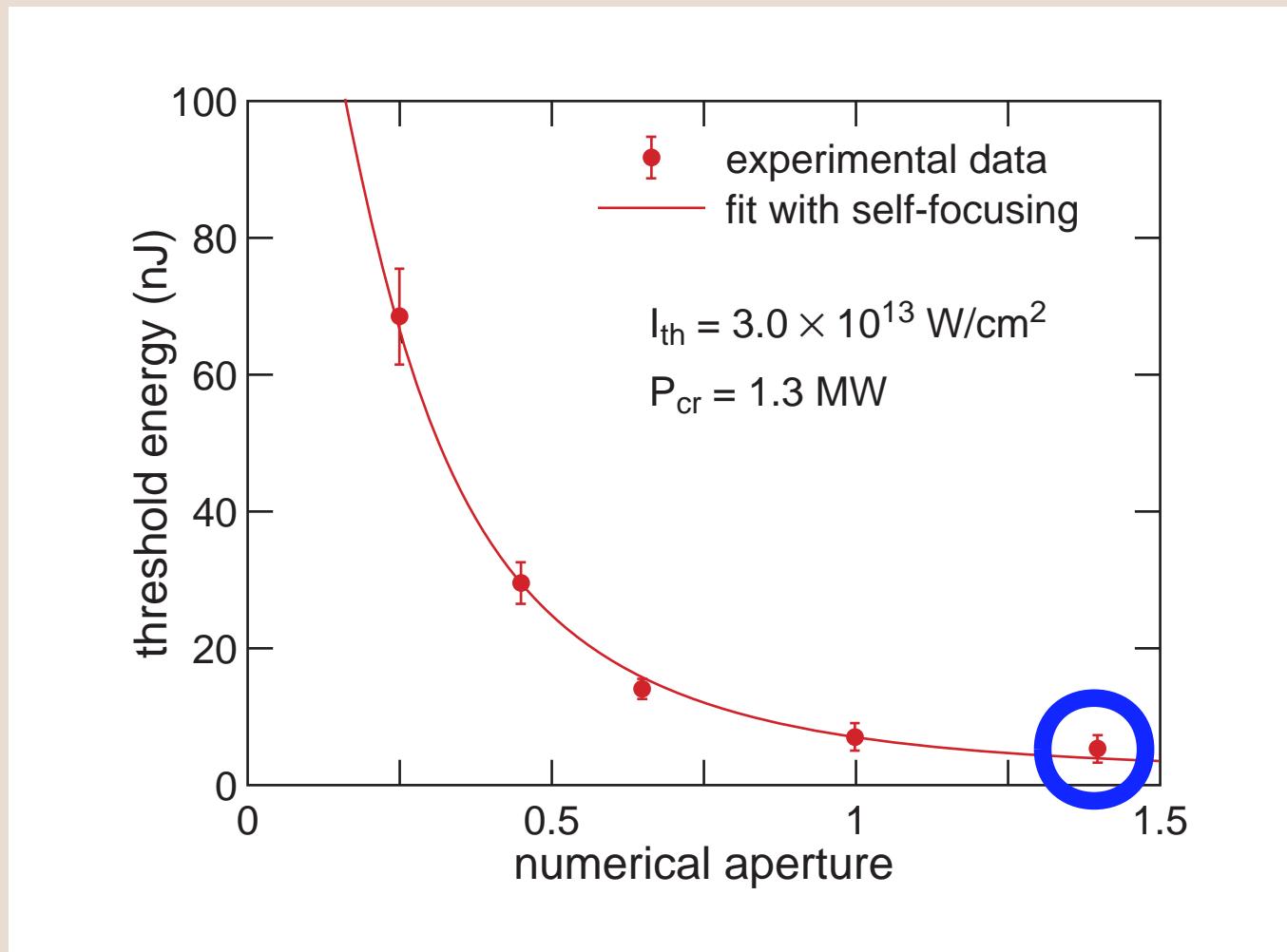


100 fs
1.4 NA
Corning 0211

\otimes^k

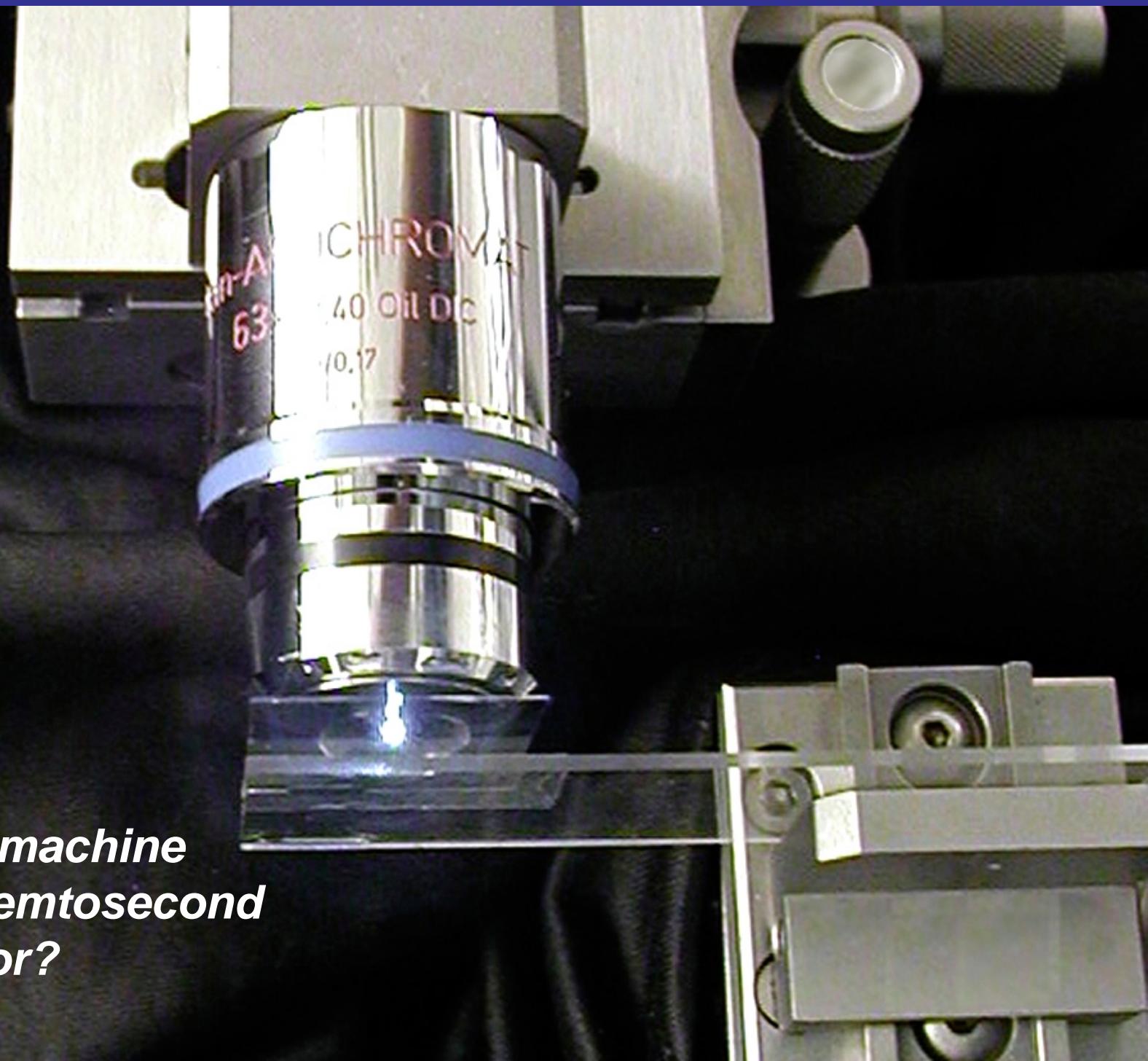
less than 10 nJ necessary for micromachining

MORPHOLOGY



5-nJ threshold

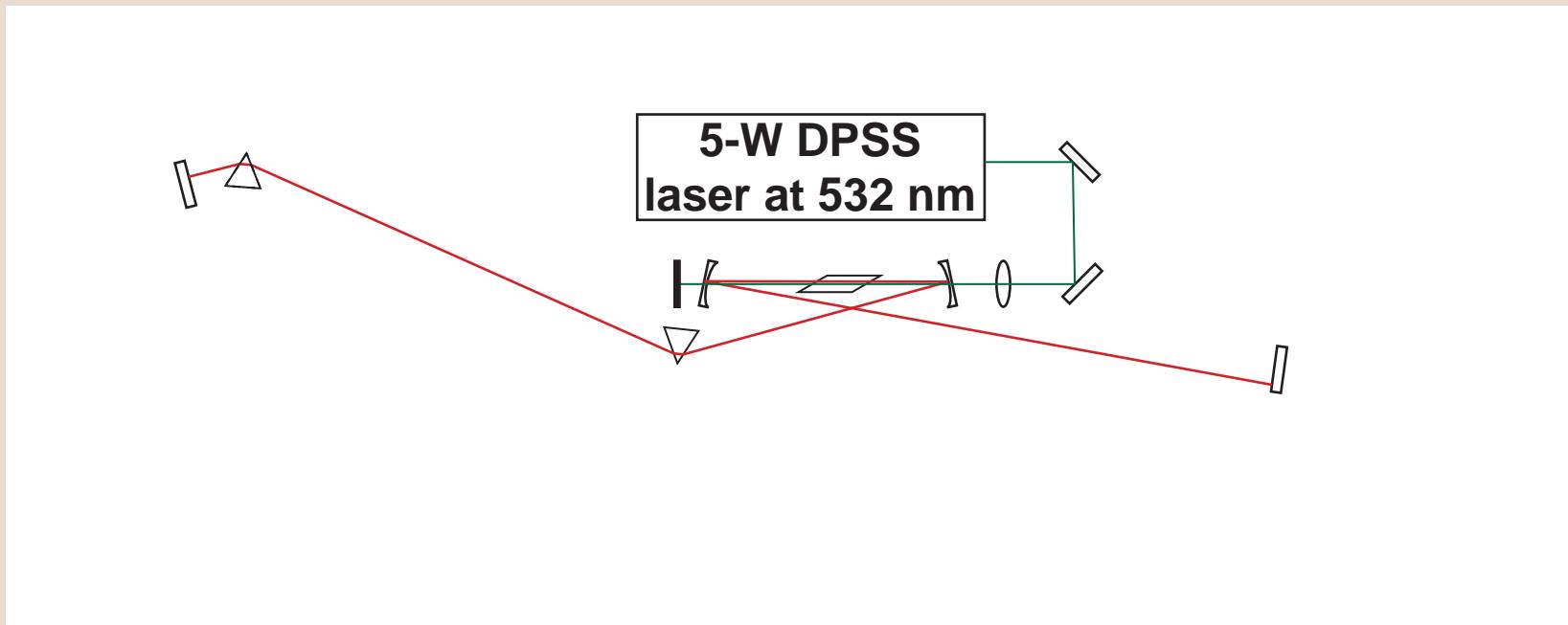
OSCILLATOR MICROMACHINING



*can we micromachine
using just a femtosecond
laser oscillator?*

OSCILLATOR MICROMACHINING

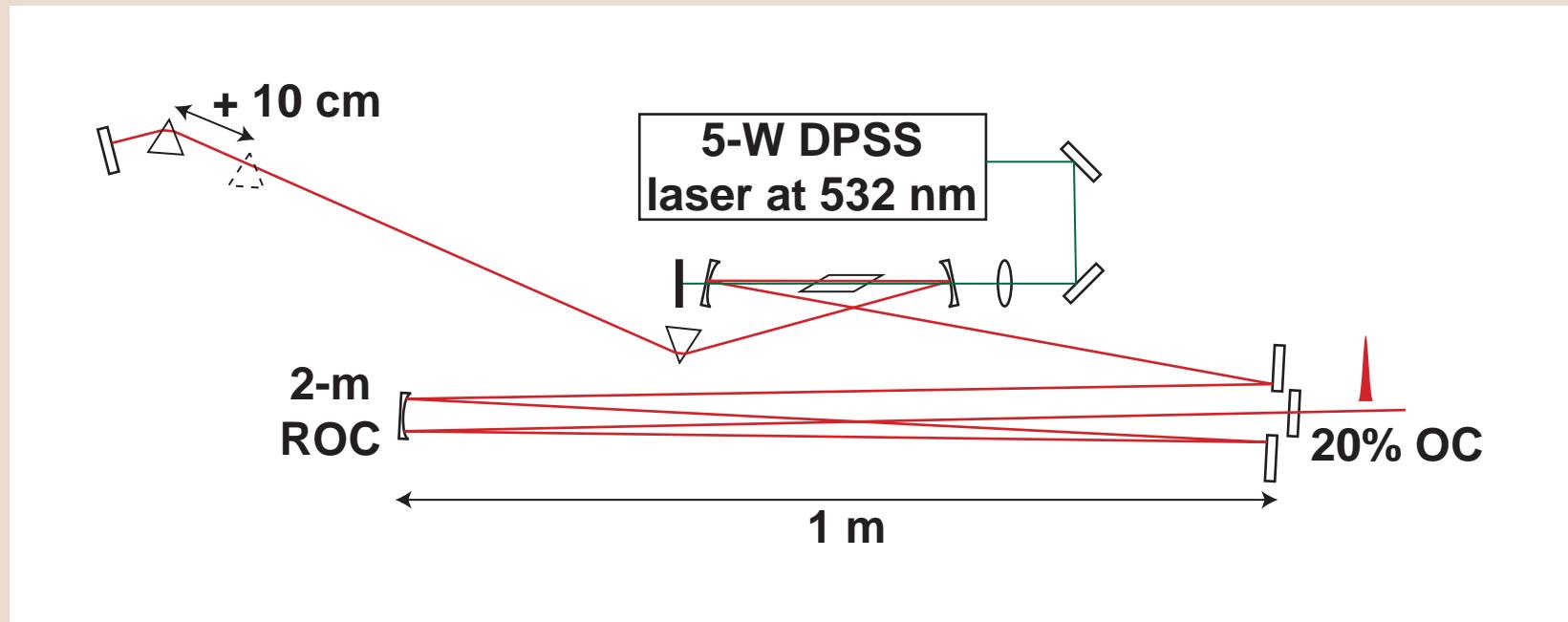
to deliver 5 nJ to the sample



after losses in the objective and prism compressor...

OSCILLATOR MICROMACHINING

... extend cavity of standard Ti:Sapph oscillator

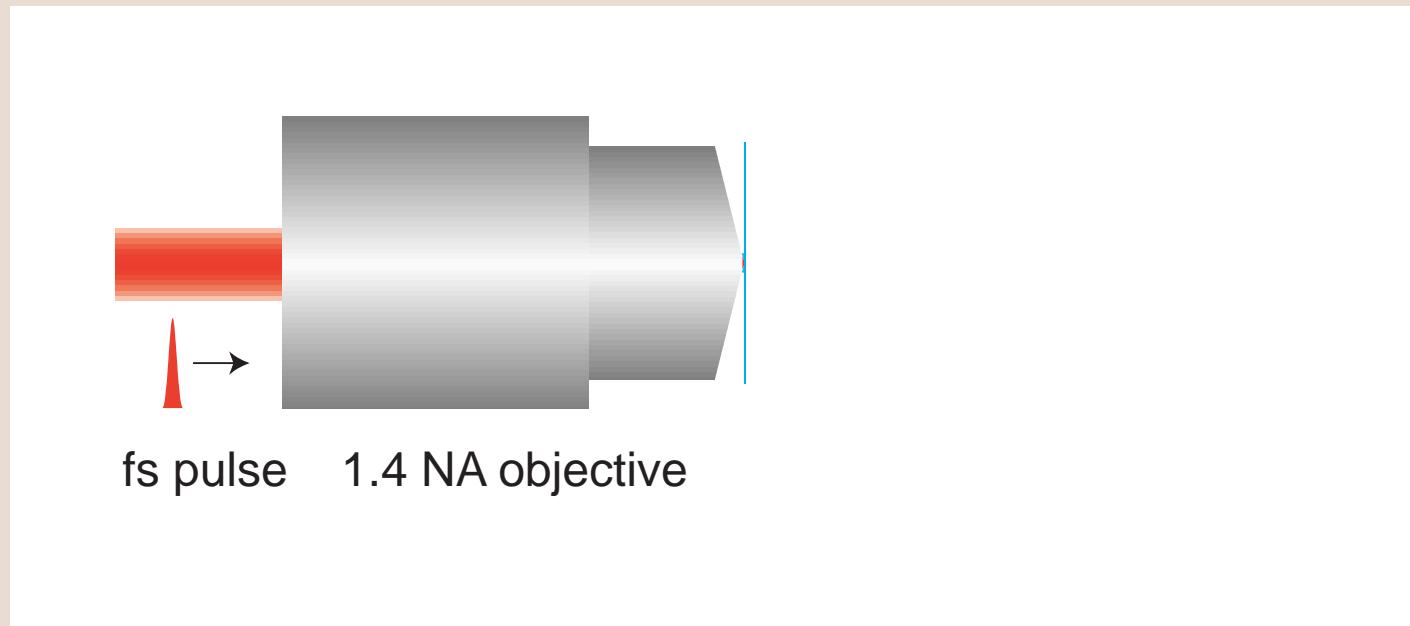


laser specs: 20 nJ, 25 MHz, 20 fs

Ref: A.R. Libertun, et.al., CLEO 1999; S.H. Cho, et. al., CLEO 1999.

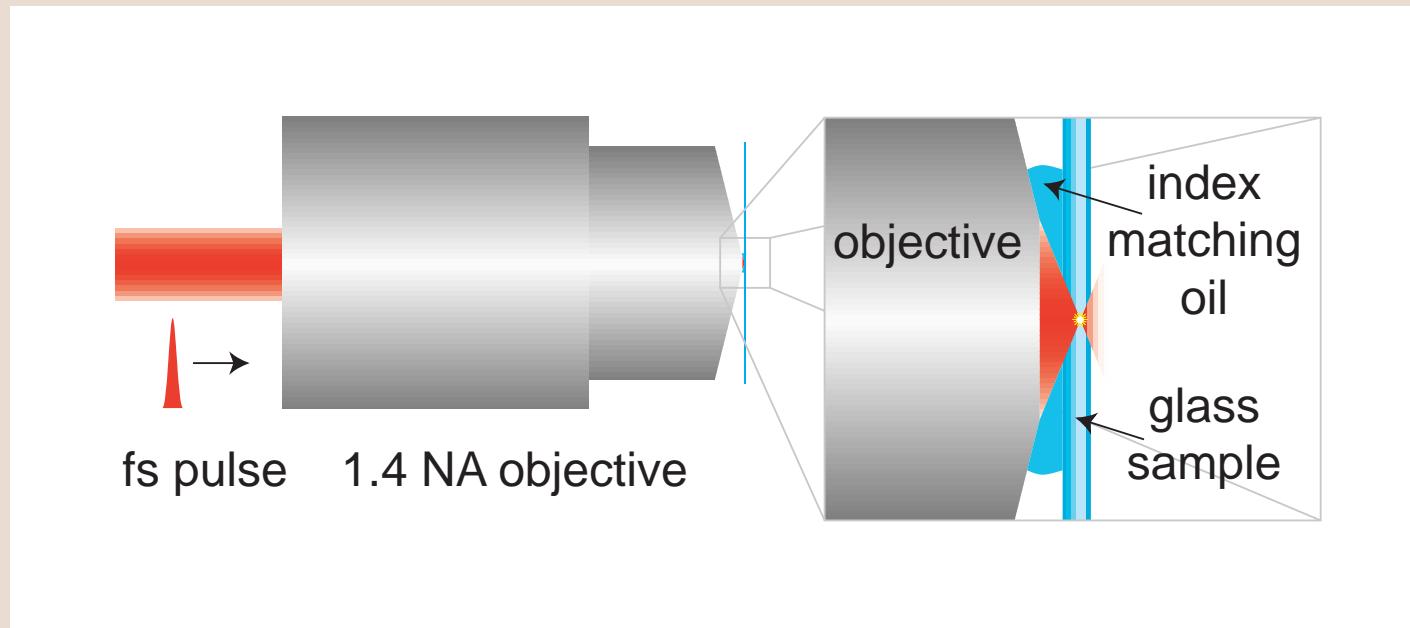
OSCILLATOR MICROMACHINING

scale model of 1.4 NA focusing geometry



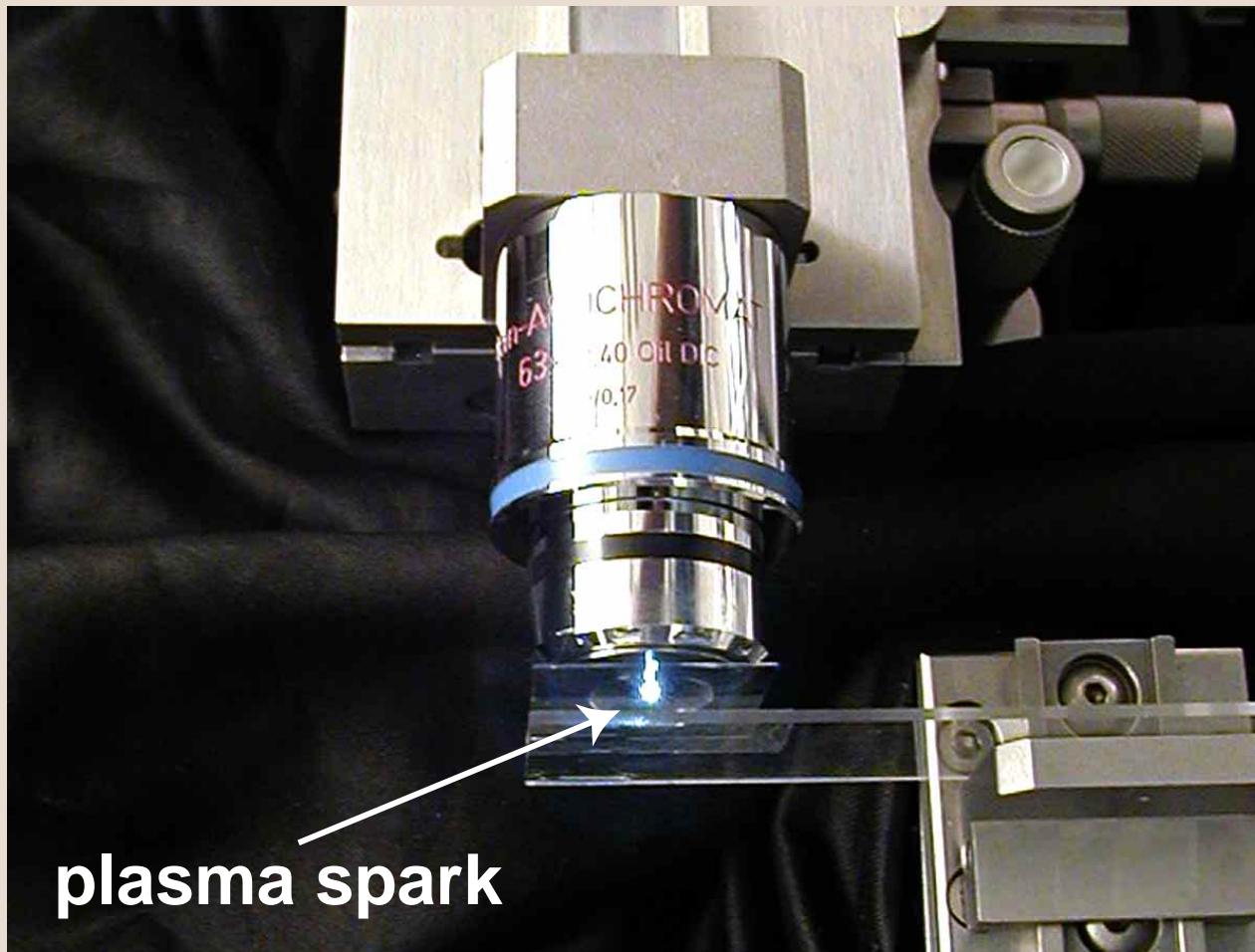
OSCILLATOR MICROMACHINING

scale model of 1.4 NA focusing geometry



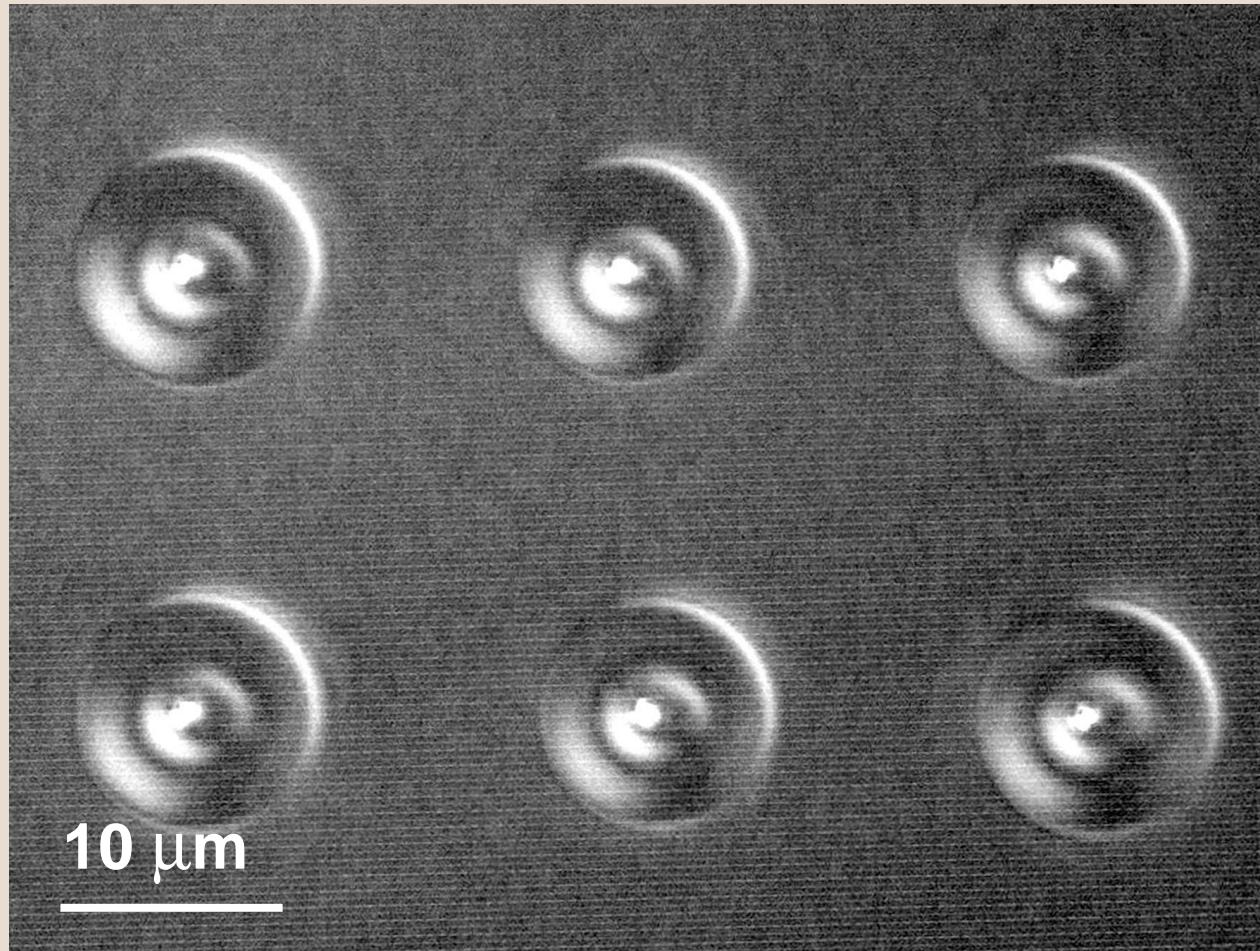
OSCILLATOR MICROMACHINING

actual 1.4 NA focusing geometry



OSCILLATOR MICROMACHINING

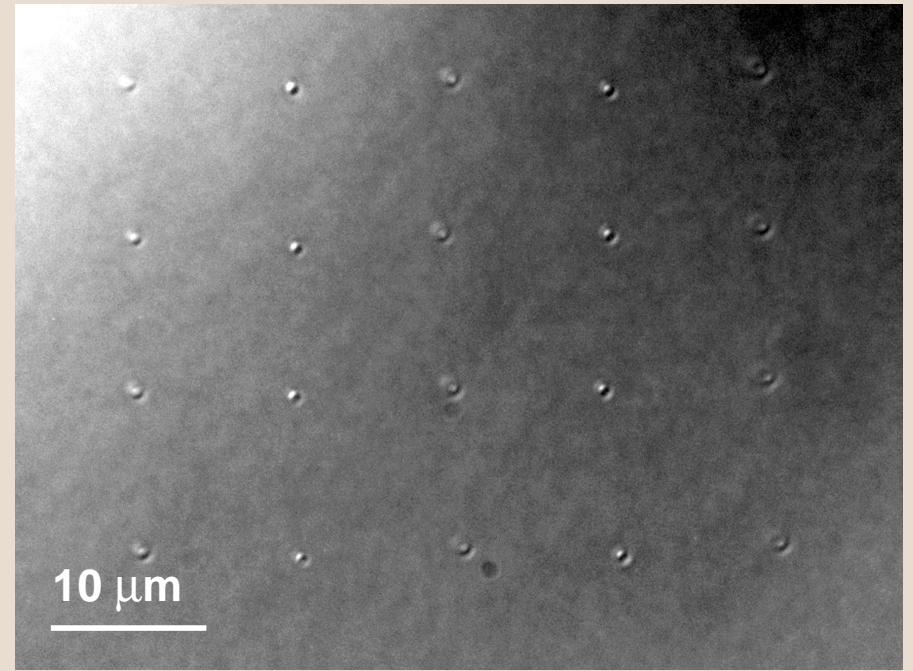
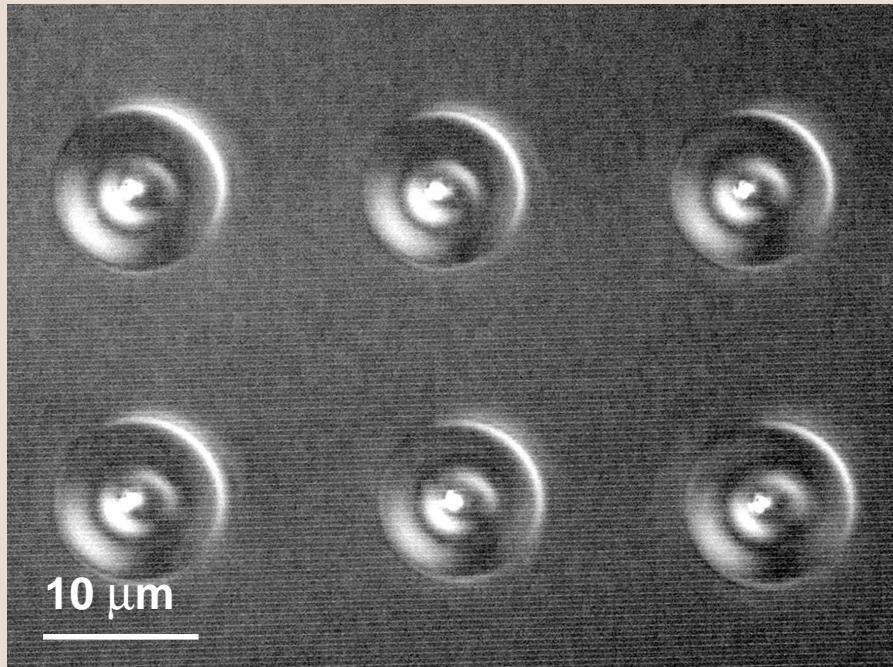
damage made with 25-MHz oscillator: 5 nJ; 25,000 shots



< 100 fs
800 nm
1.4 NA
Corning 0211

OSCILLATOR MICROMACHINING

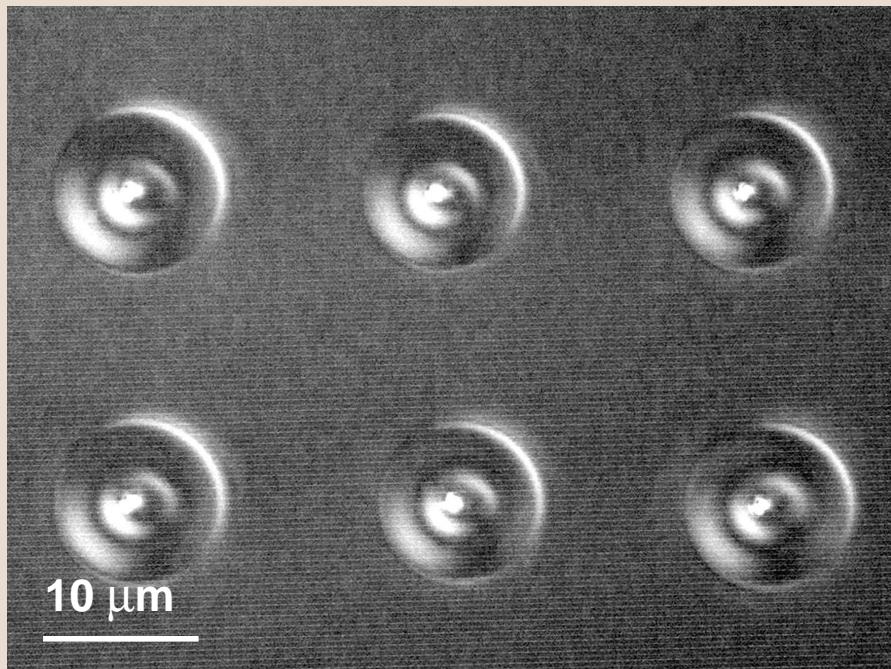
5 nJ; 25,000 shots



10 nJ; single shot

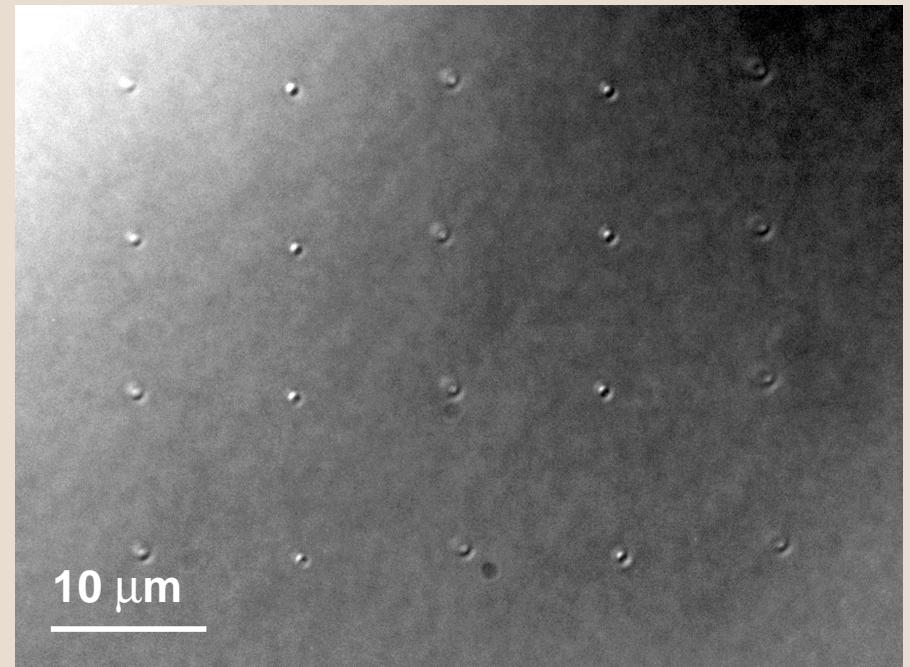
OSCILLATOR MICROMACHINING

5 nJ; 25,000 shots



**cumulative heating by
successive pulses melts
the glass**

**explosive or small-scale
melting mechanism**



10 nJ; single shot

OSCILLATOR MICROMACHINING

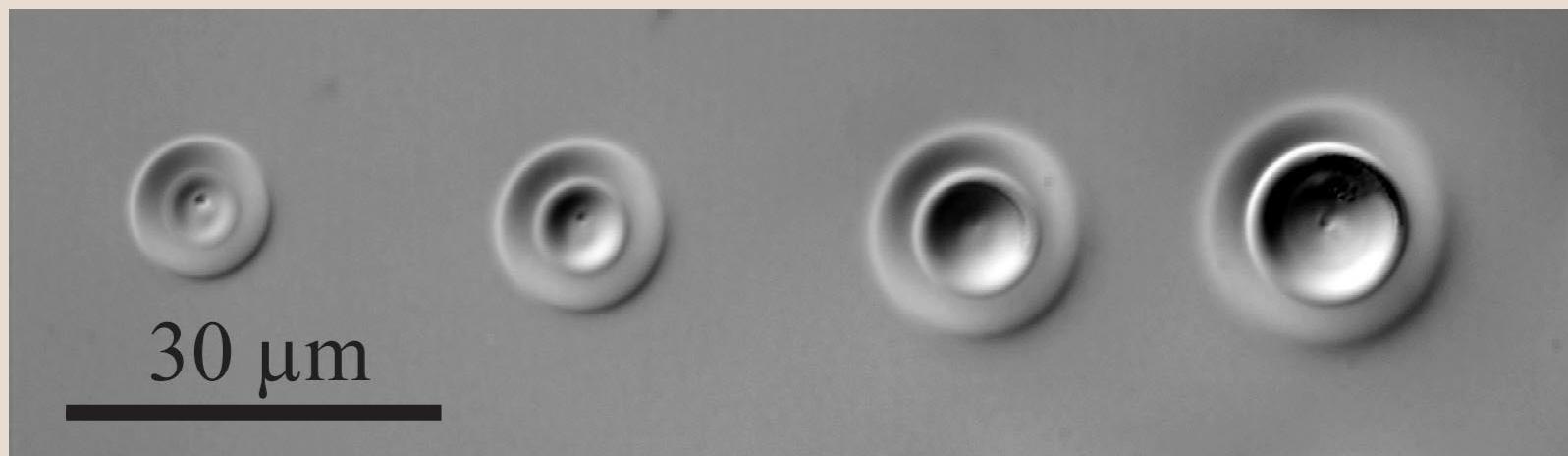
number of laser shots

2.5×10^4

2.5×10^5

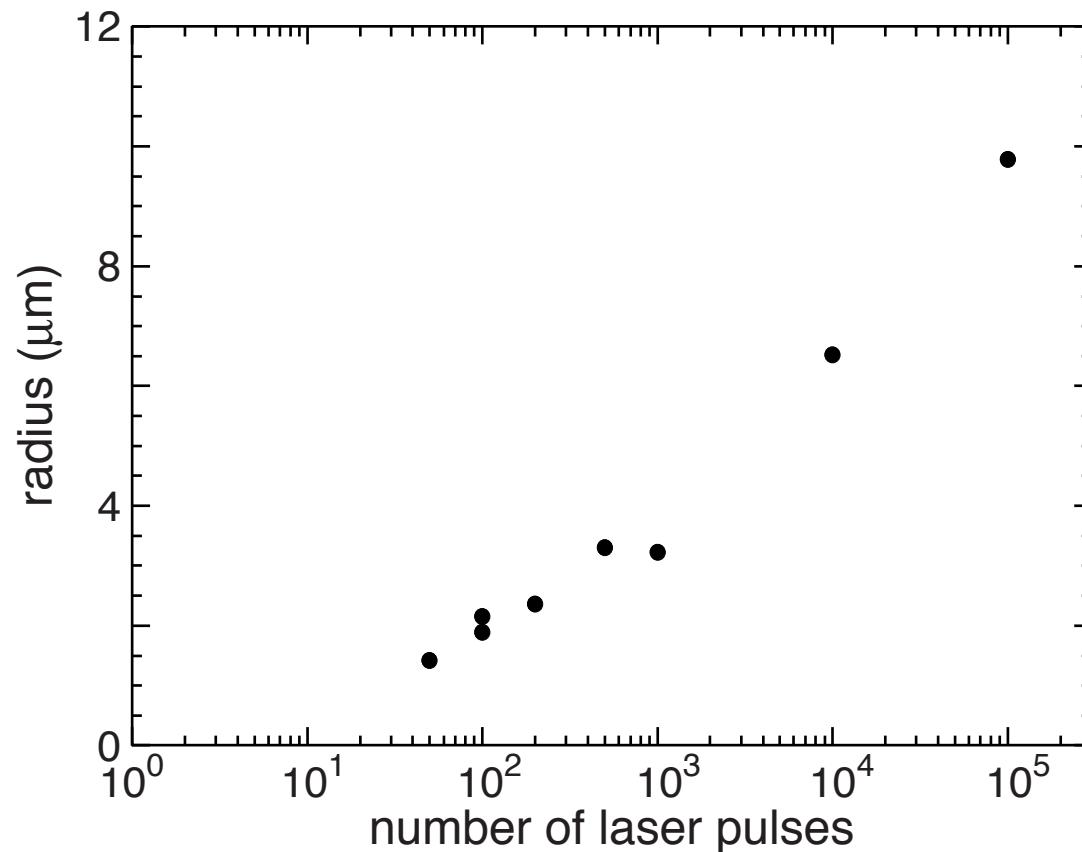
2.5×10^6

2.5×10^7



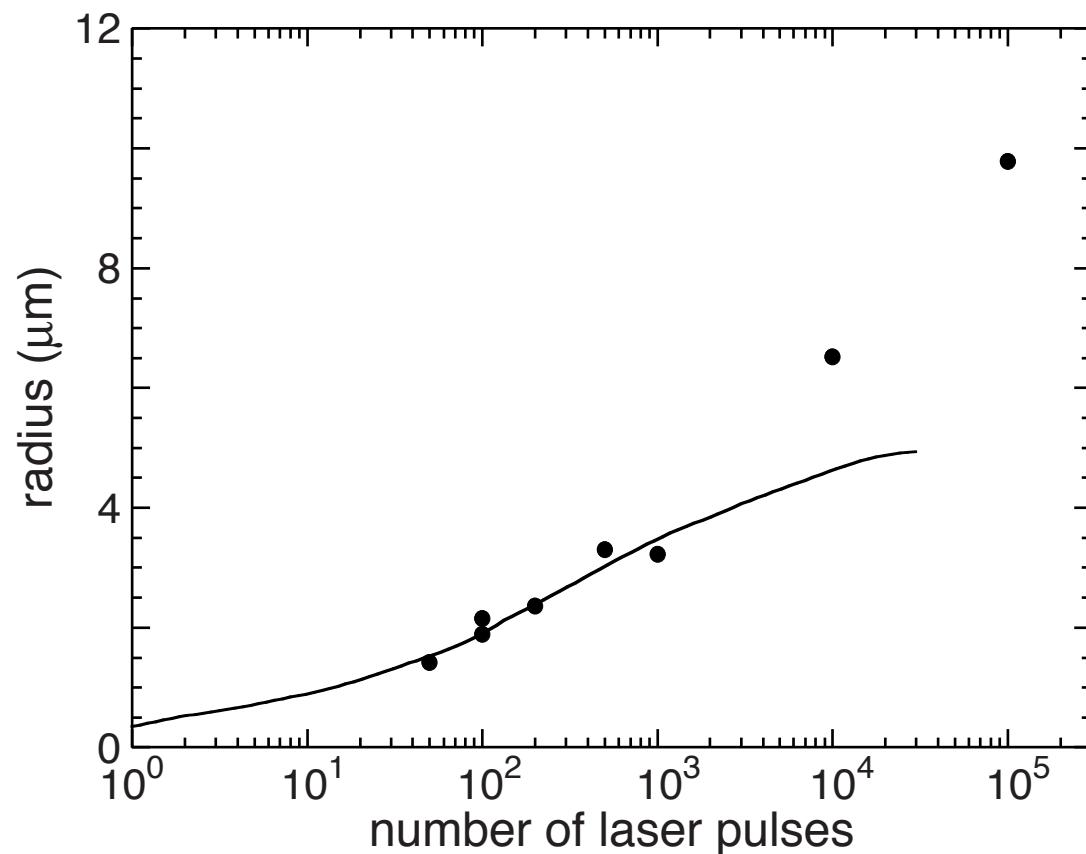
more shots melt larger volumes

OSCILLATOR MICROMACHINING



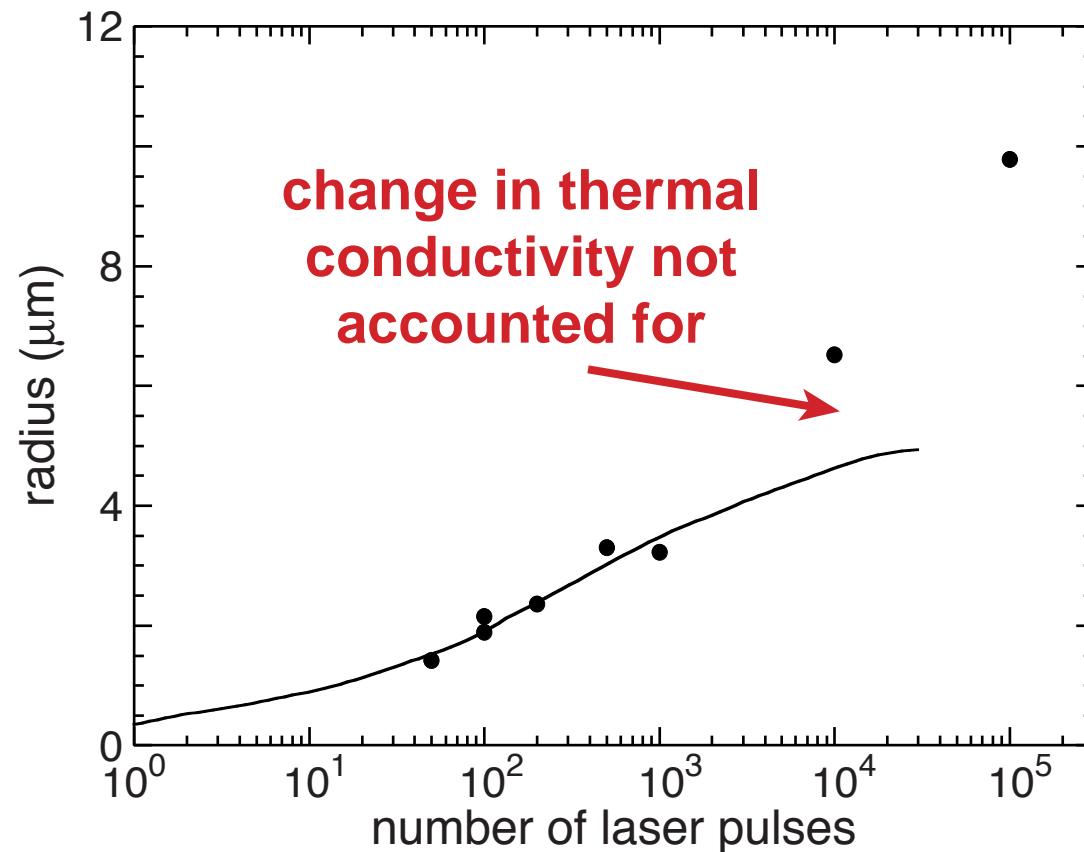
radius of melted structure

OSCILLATOR MICROMACHINING



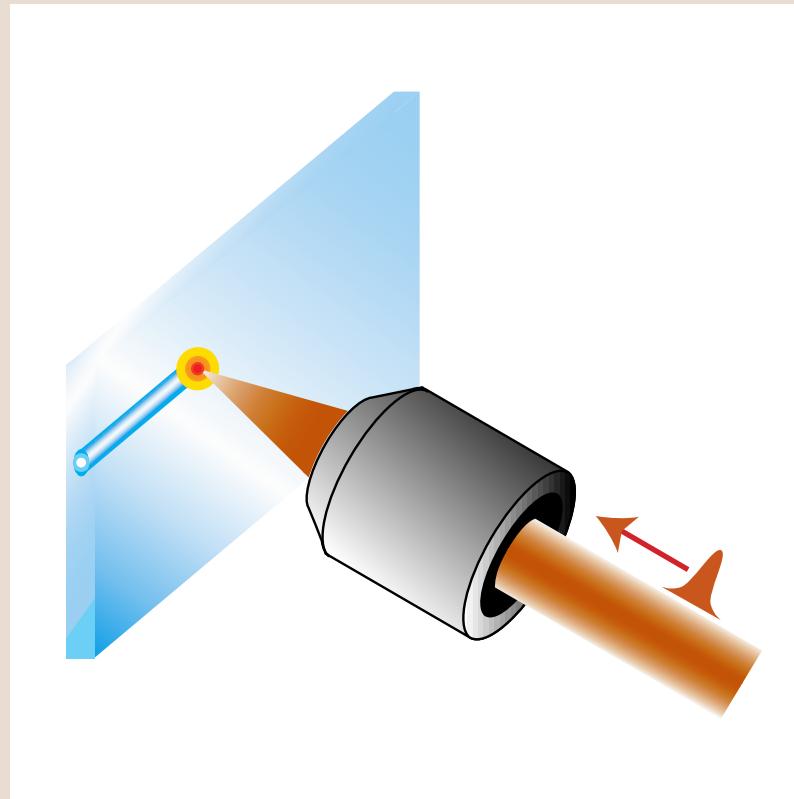
model with diffusion equation

OSCILLATOR MICROMACHINING



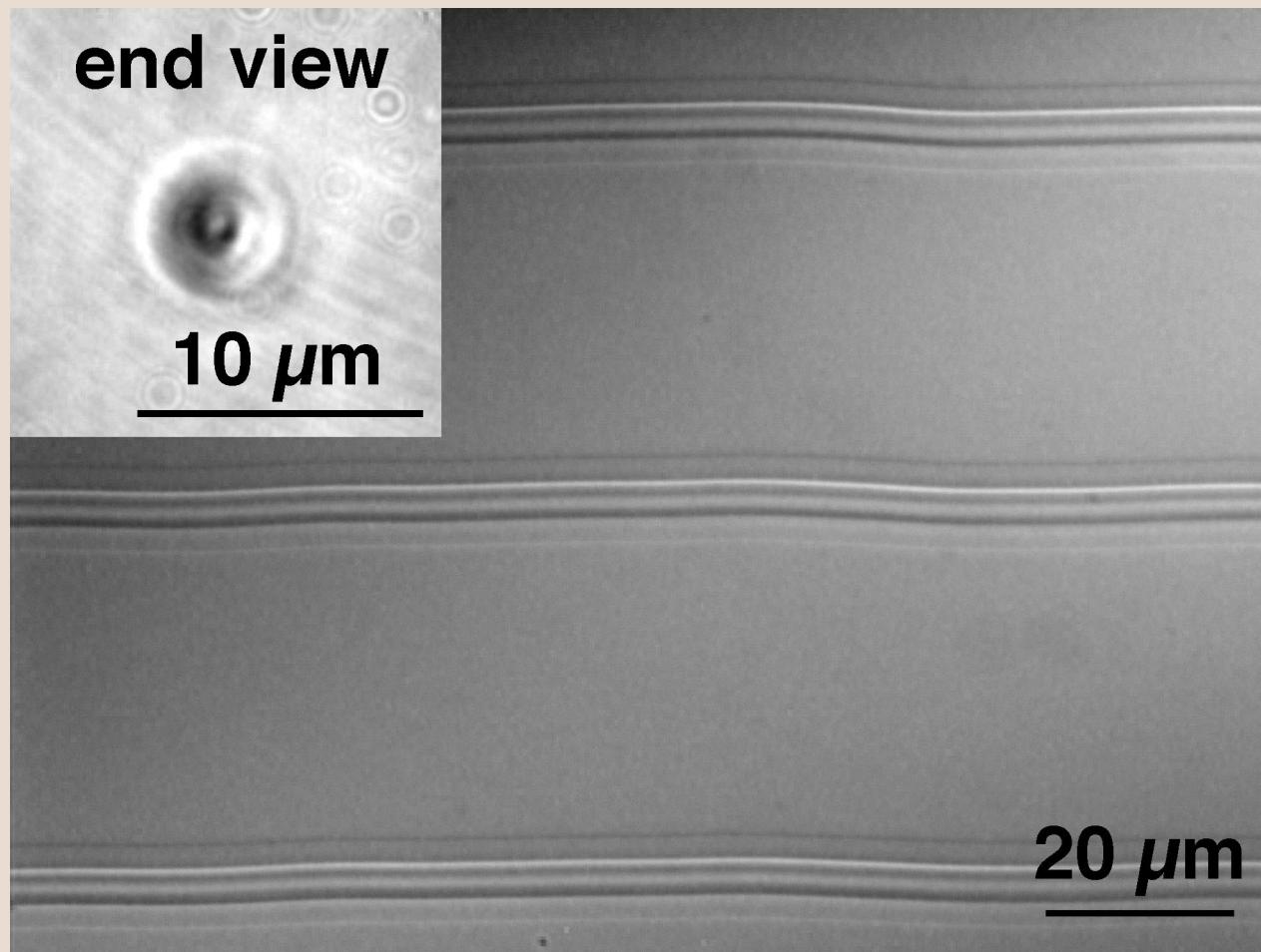
model with diffusion equation

how can we use femtosecond thermal micromachining?



write waveguides!

waveguide morphology: 20 mm/s machining speed



waveguide mode analysis

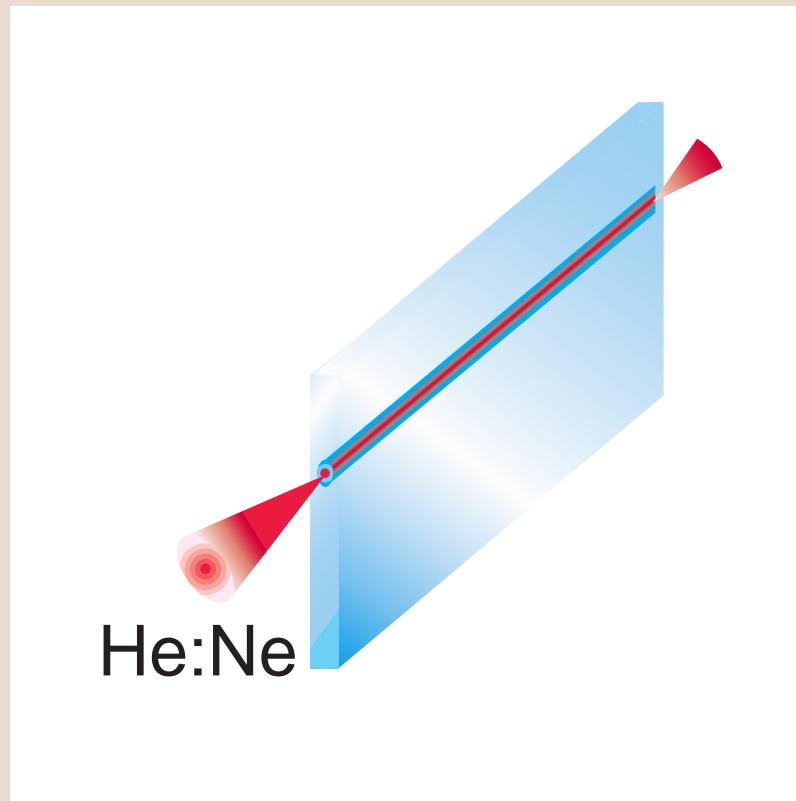
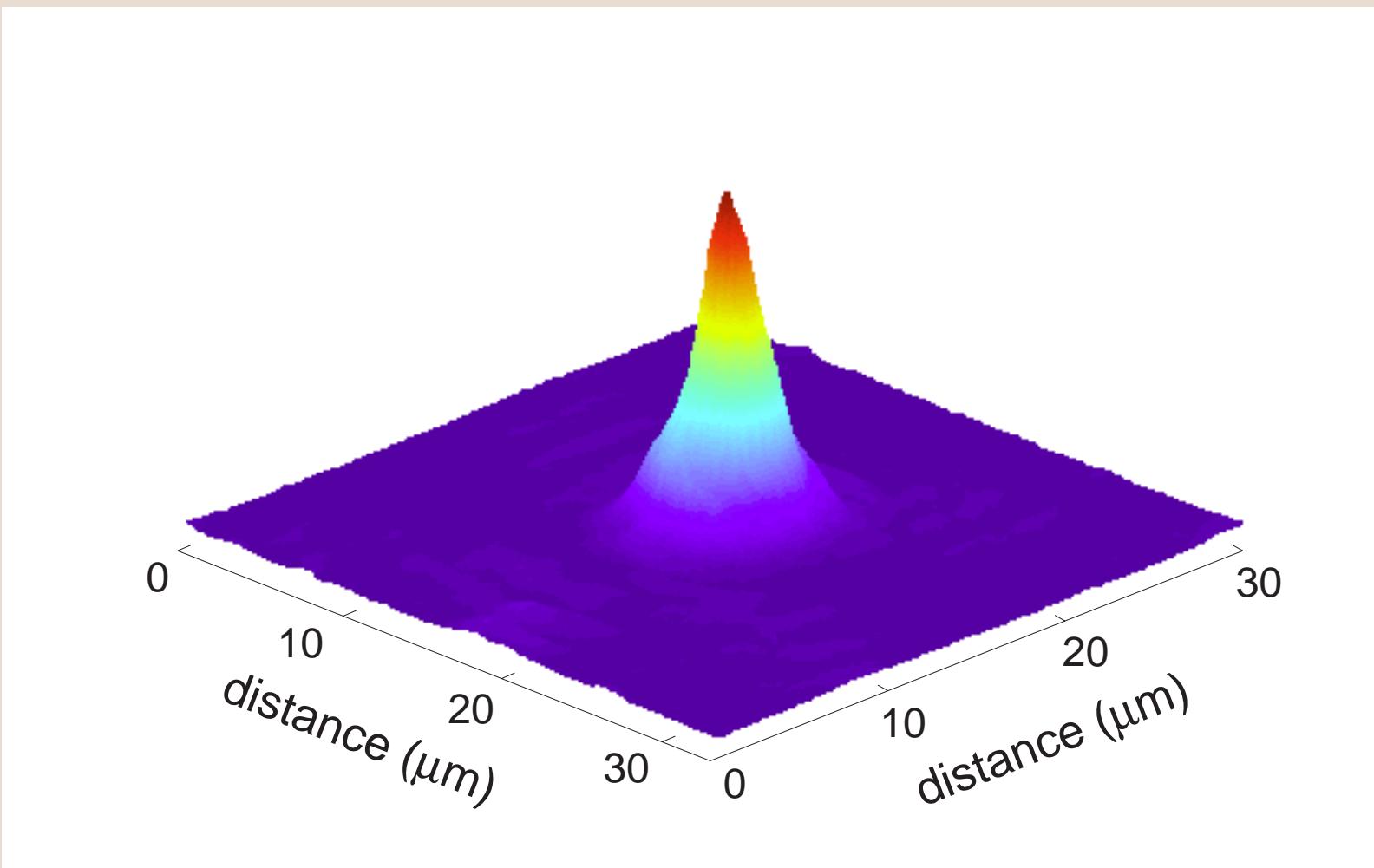
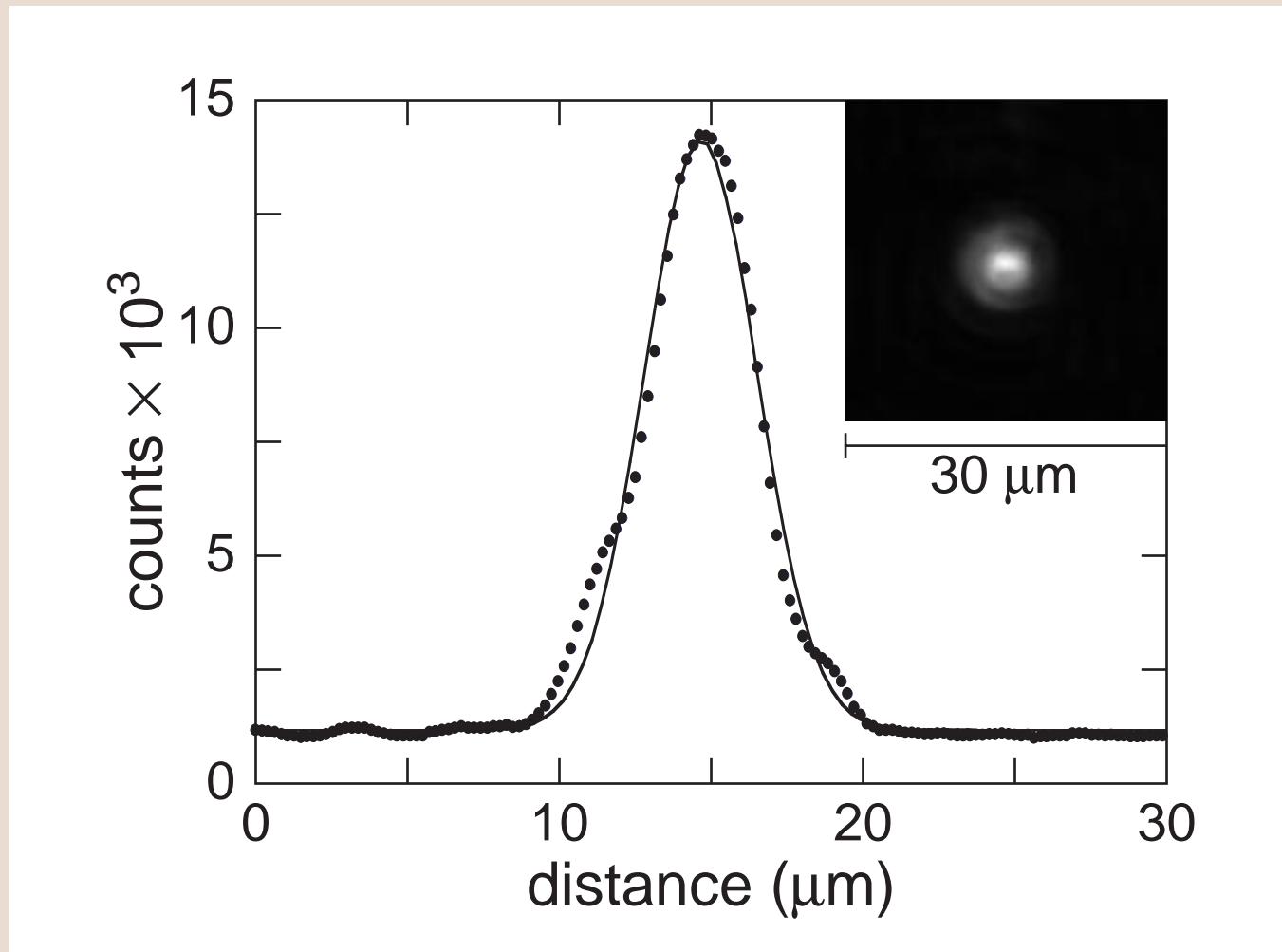


image of near field mode



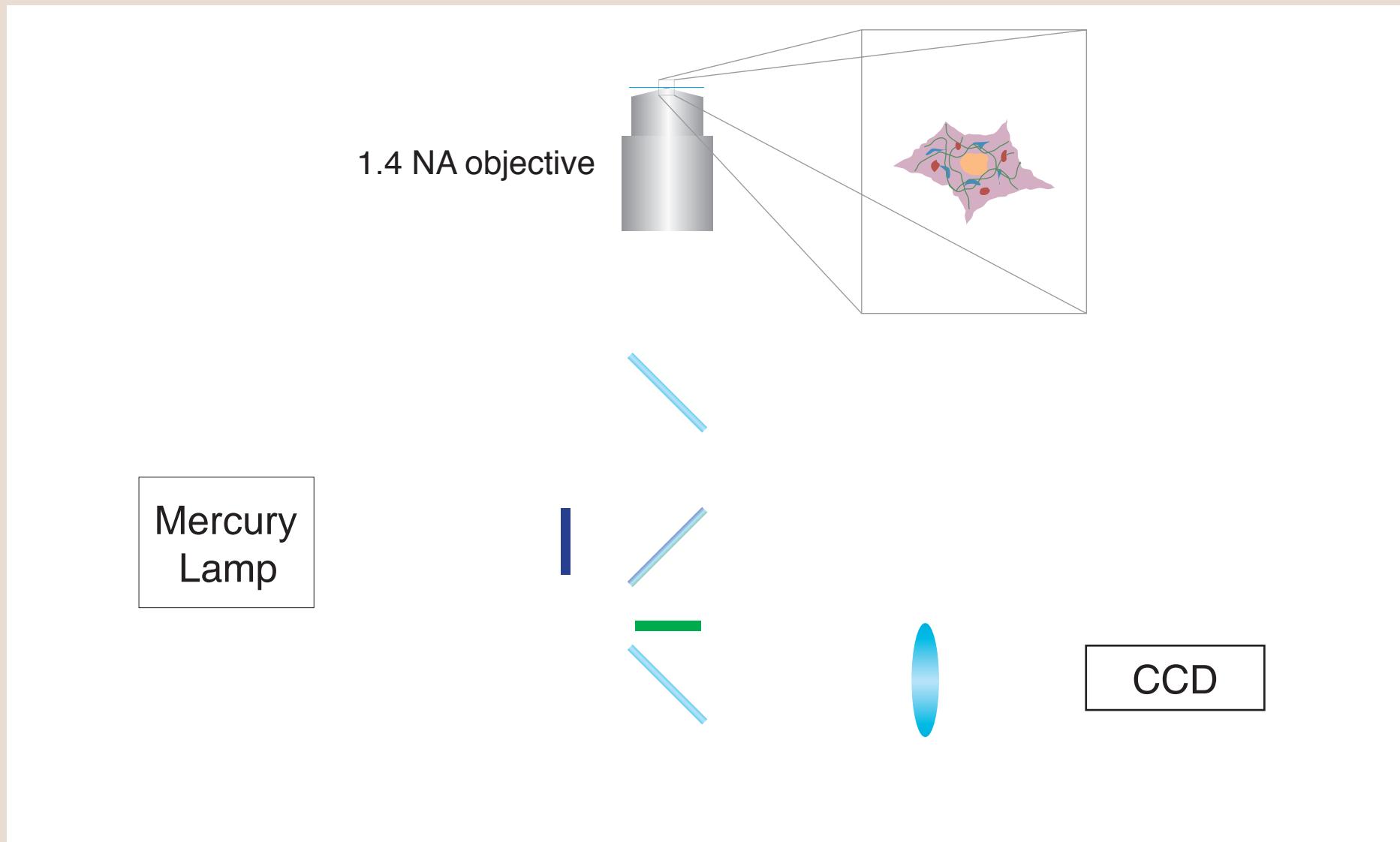


line-out of near field waveguide mode for 633 nm

A fluorescence microscopy image showing a single cell with a complex network of green fluorescent filaments, likely actin or microtubules, distributed throughout its cytoplasm and extending towards the periphery. The cell has a rounded nucleus and some darker, unlabeled regions.

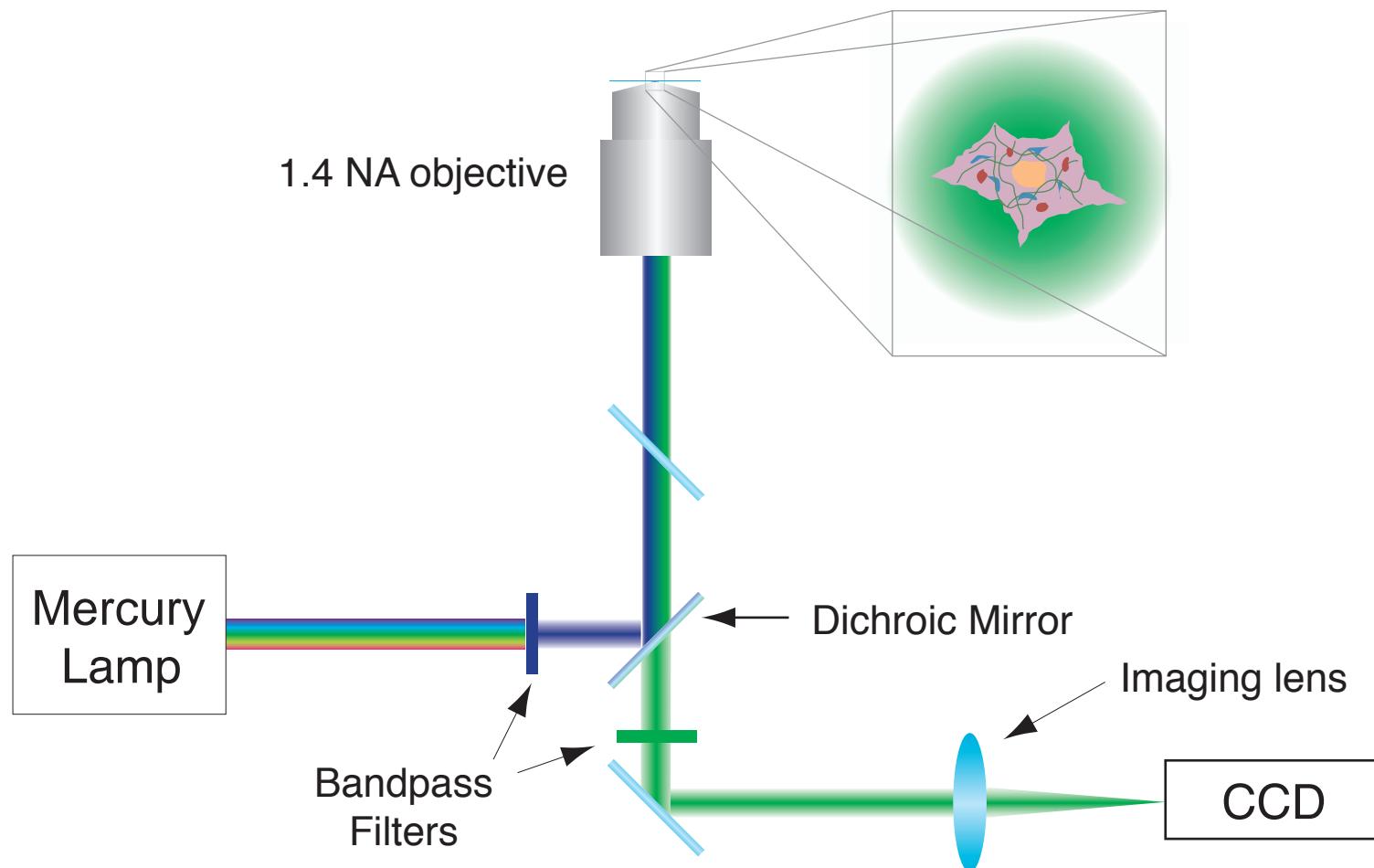
**can we do
sub-cellular
photodisruption?**

microscope

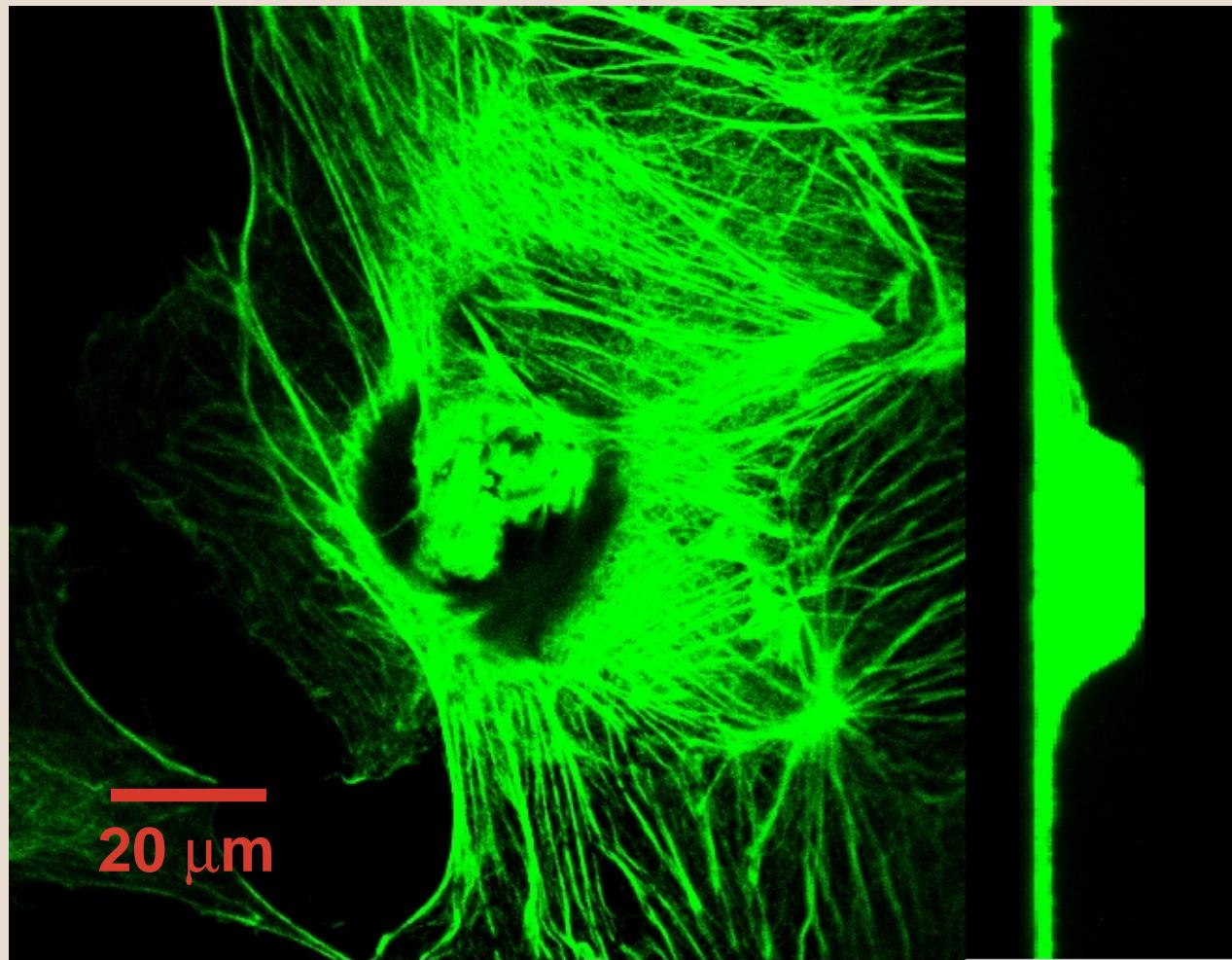


APPLICATIONS

image

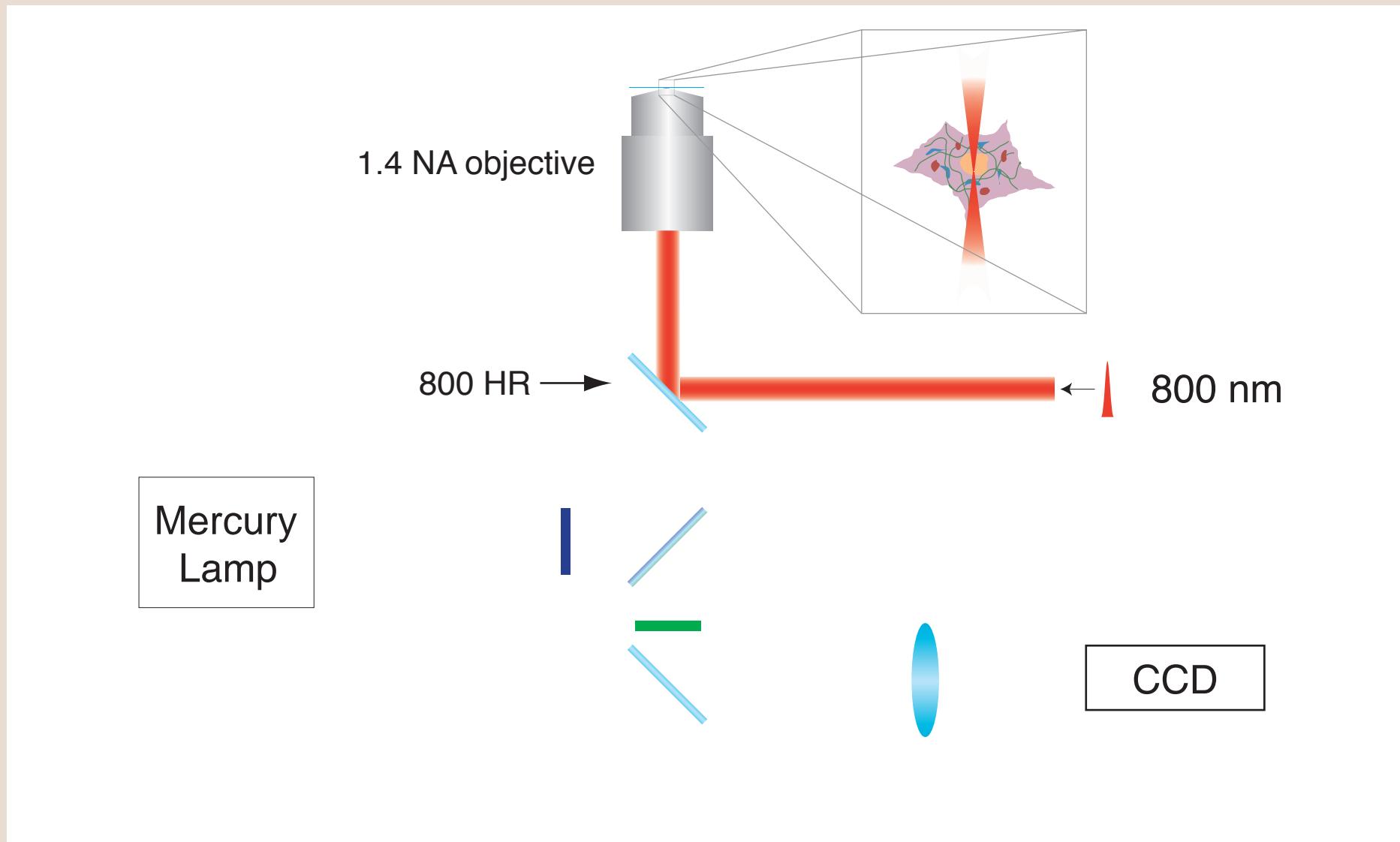


APPLICATIONS



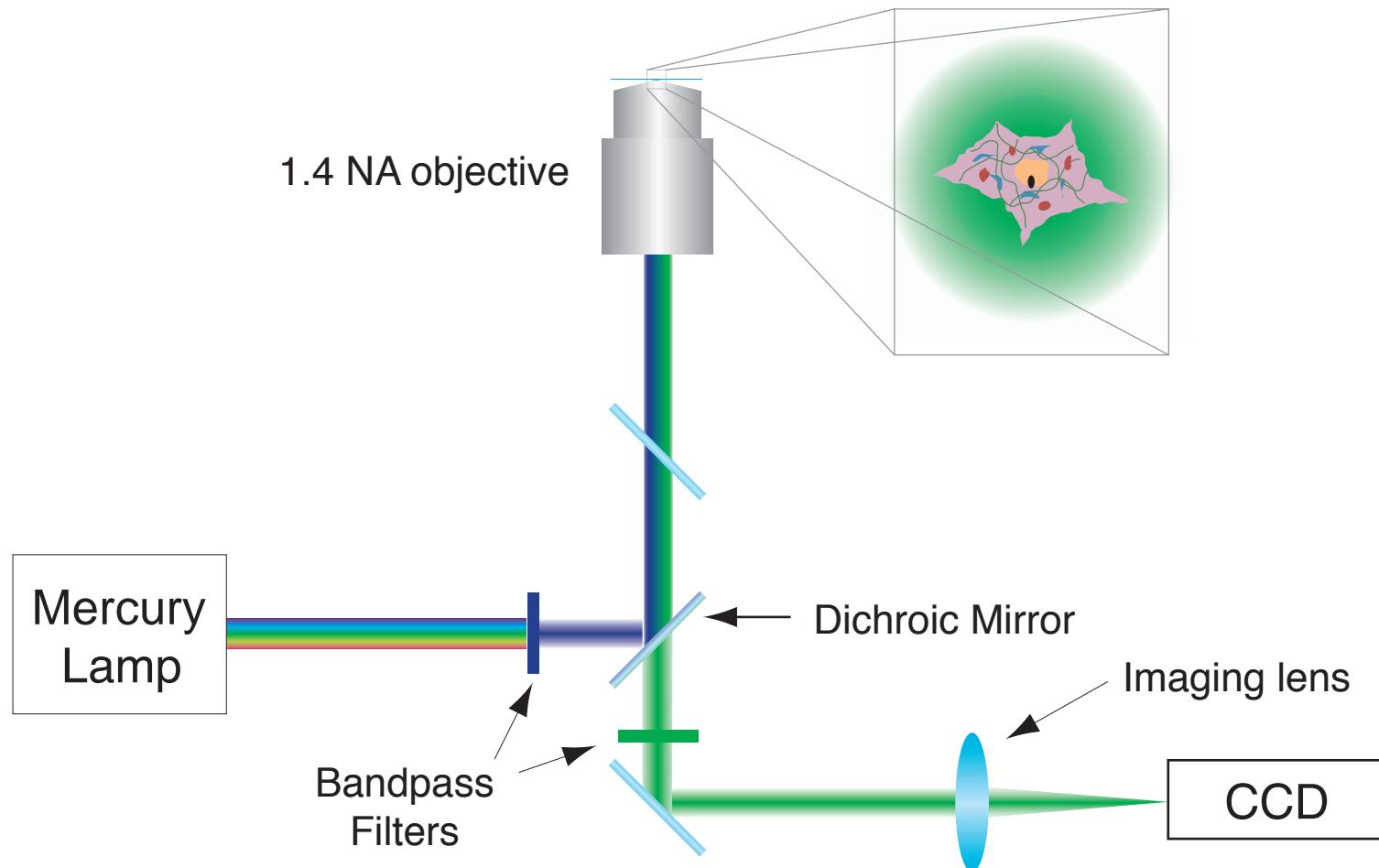
APPLICATIONS

irradiate

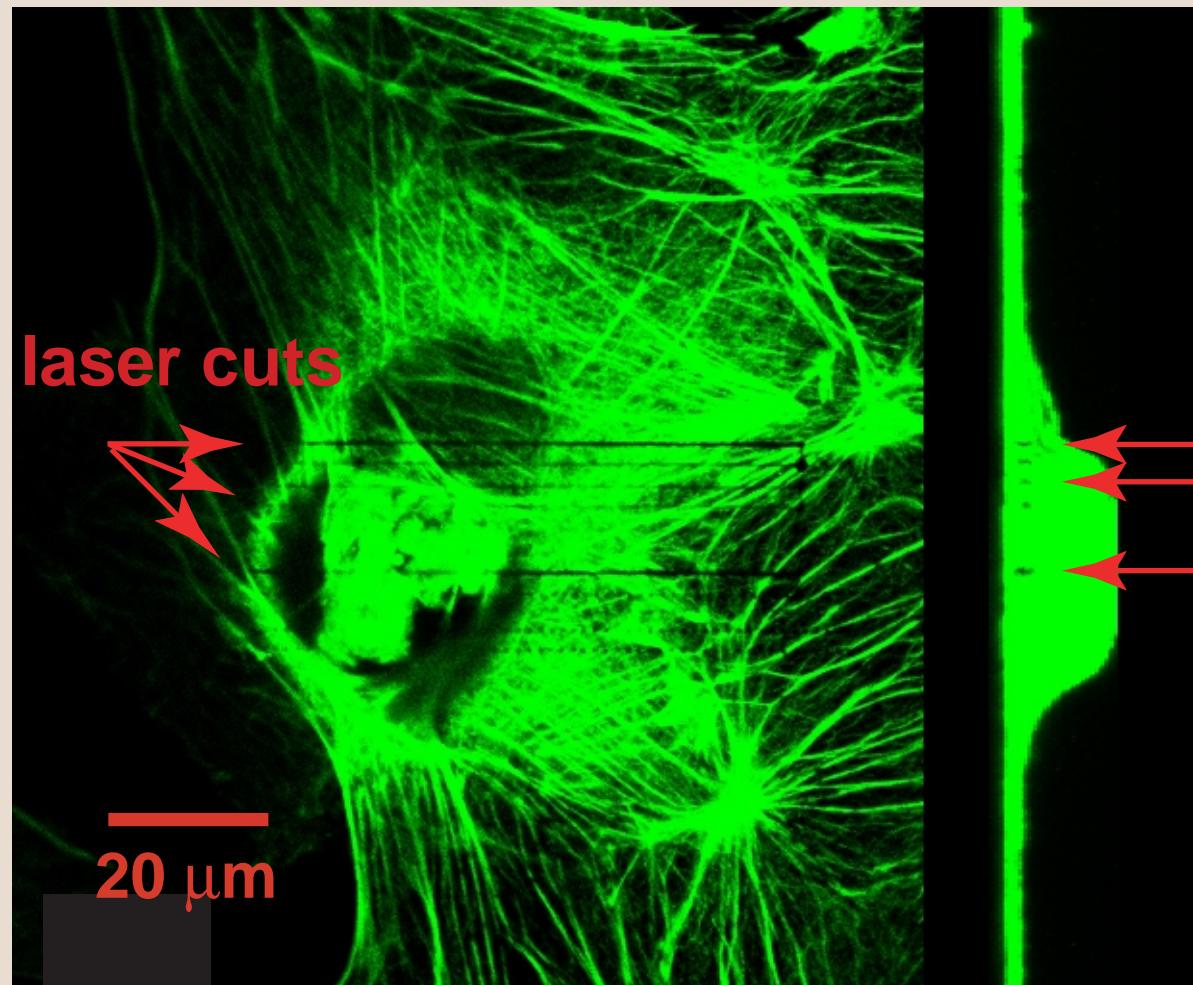


APPLICATIONS

re-image



APPLICATIONS



1 - 3nJ, 100 fs, 1.4 NA

Structural change morphology

- Connect morphology to mechanisms

morphologies and mechanisms

single shot

multiple shot
high rep. rate

low energy

sub- μm Δn
small melt ?

μm -sized Δn
cumulative melting

high energy

sub- μm voids
explosive

CAN'T AFFORD!
\$\$\$\$

Structural change morphology

- Connect morphology to mechanisms
- Optical, SEM, AFM studies

Structural change morphology

- Connect morphology to mechanisms
- Optical, SEM, AFM studies

Applications

- Oscillator-only waveguide micromachining
- Sub-cellular photodisruption
 - functional studies of sub-cellular structures in live cells

ACKNOWLEDGEMENTS

**W. Leight and D. Datta
Prof. N. Bloembergen**

National Science Foundation

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

<http://mazur-www.harvard.edu/>

3-D micromachining of photonics devices

structural change morphology

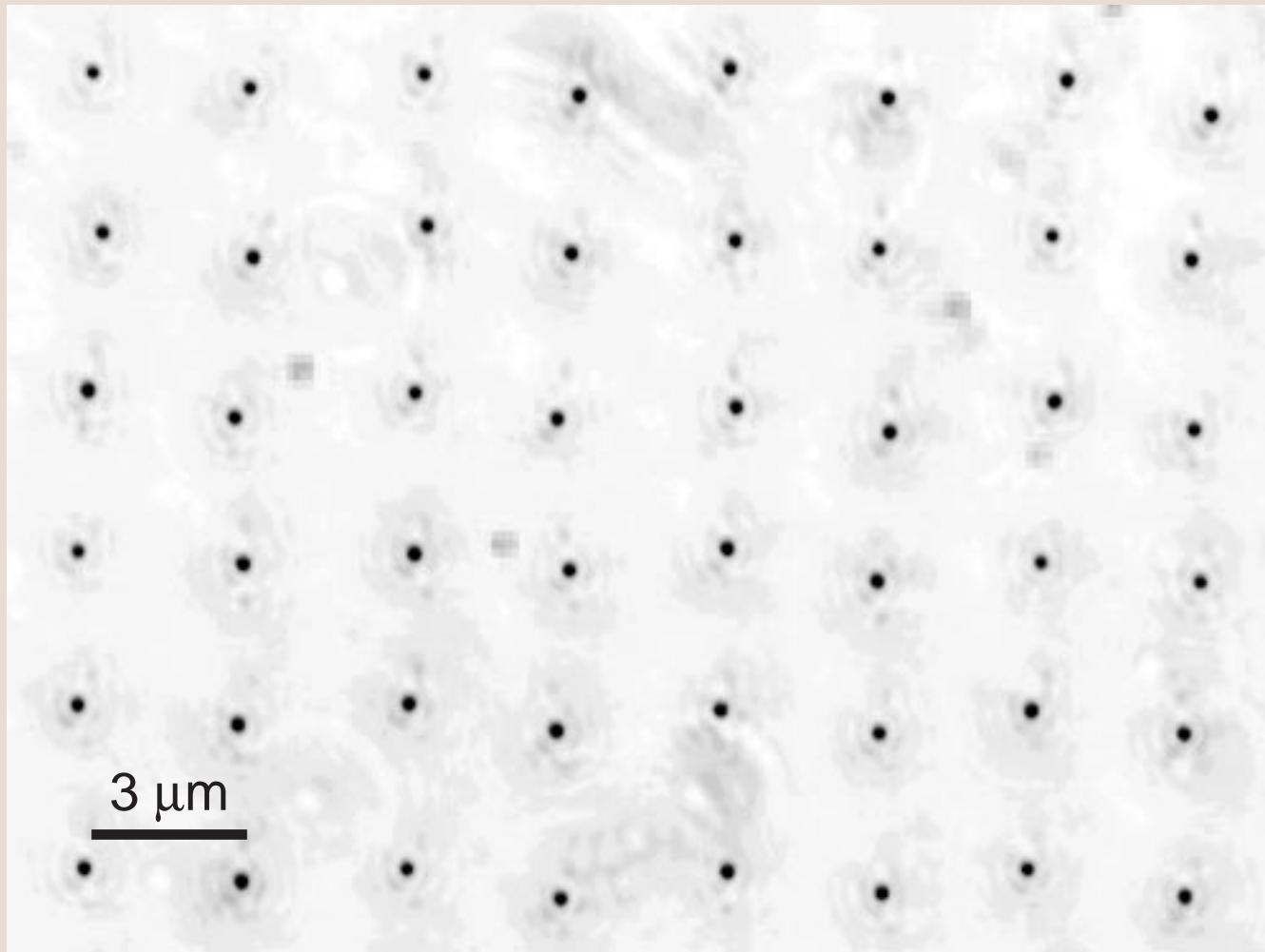
nonlinear ionization

sub-cellular laser surgery

laser induced chemical changes

white light generation

MORPHOLOGY

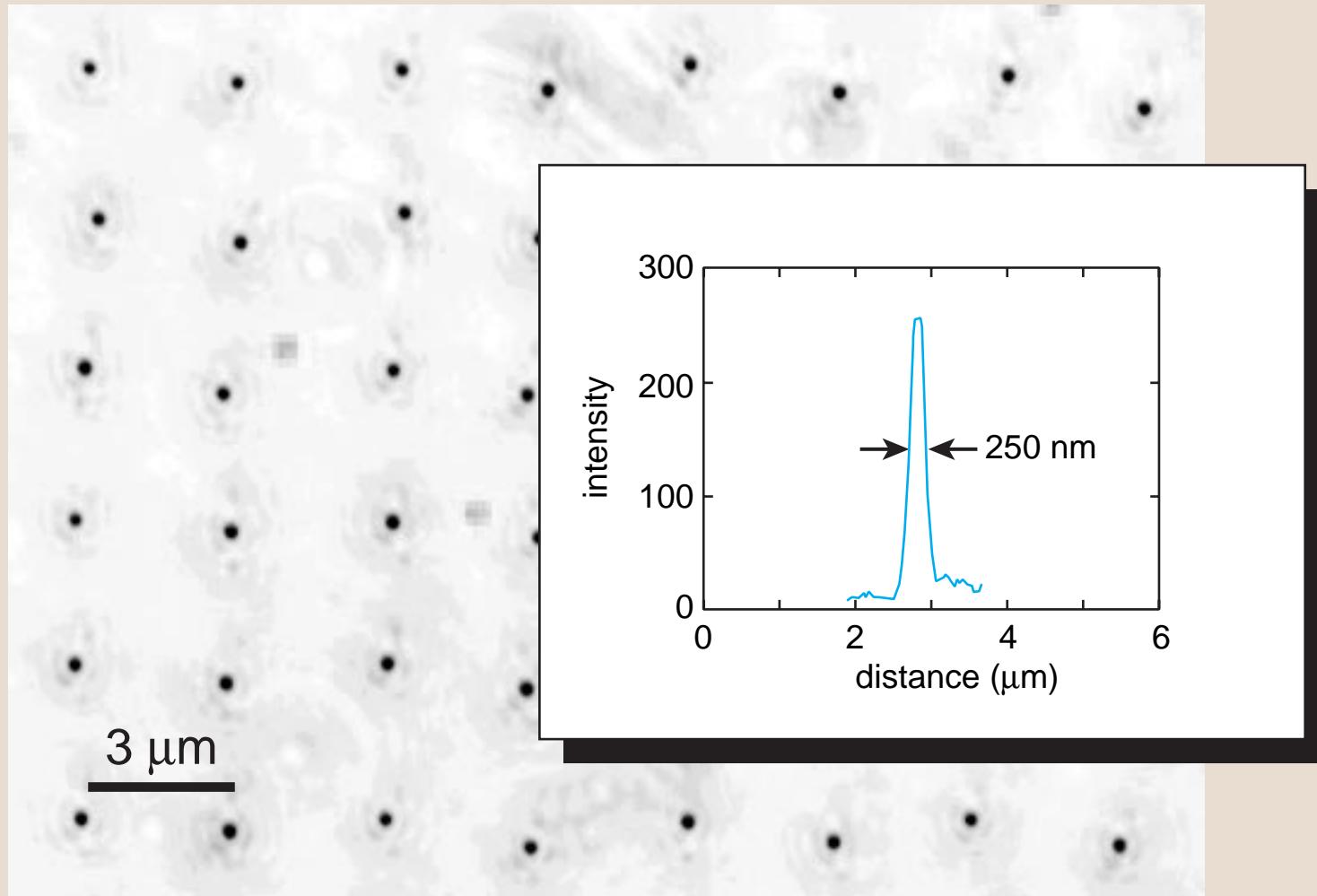


40 nJ
100 fs
800 nm
0.65 NA
Corning 0211

top view

MORPHOLOGY

40 nJ
100 fs
800 nm
0.65 NA
Corning 0211



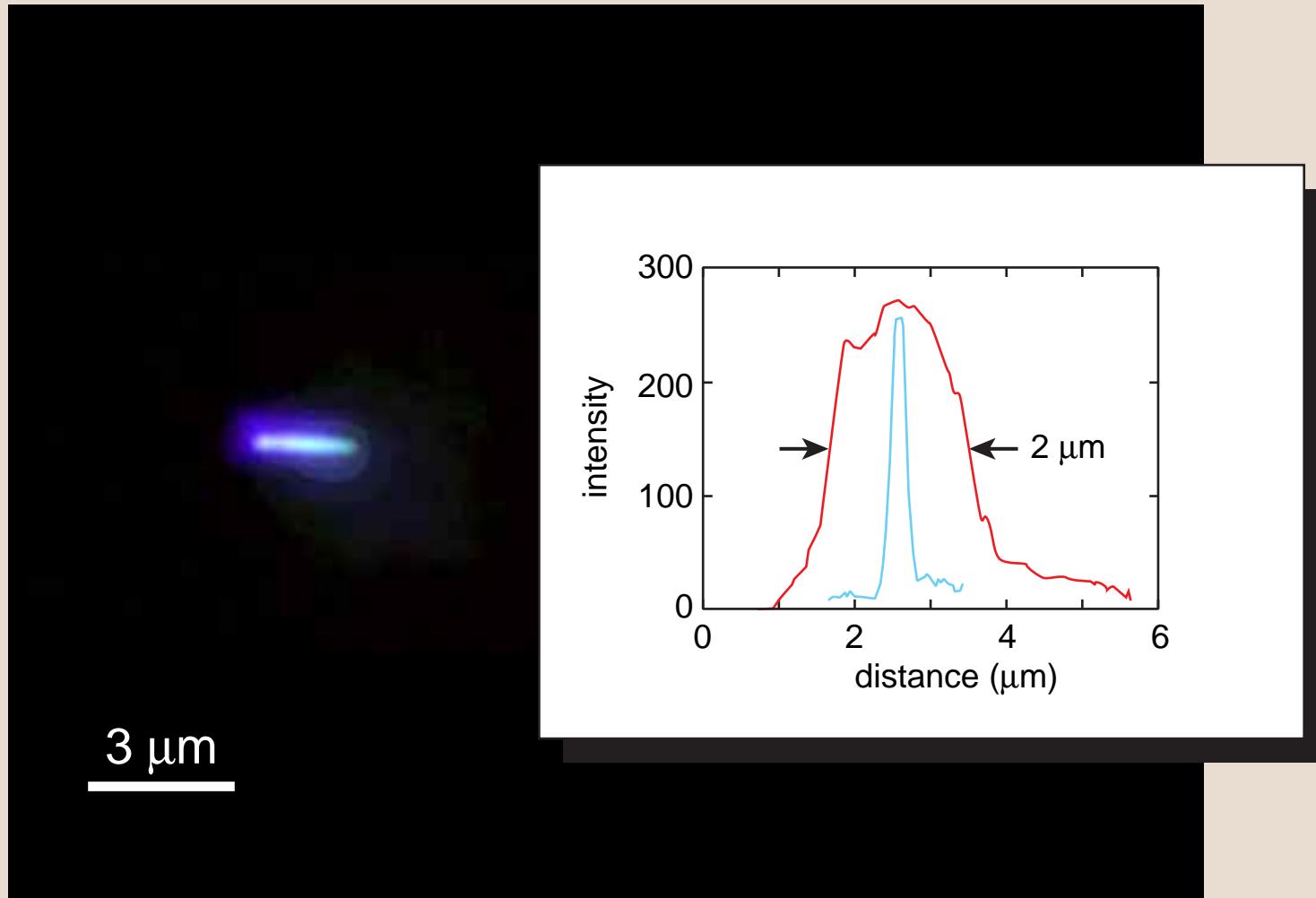
MORPHOLOGY



side view

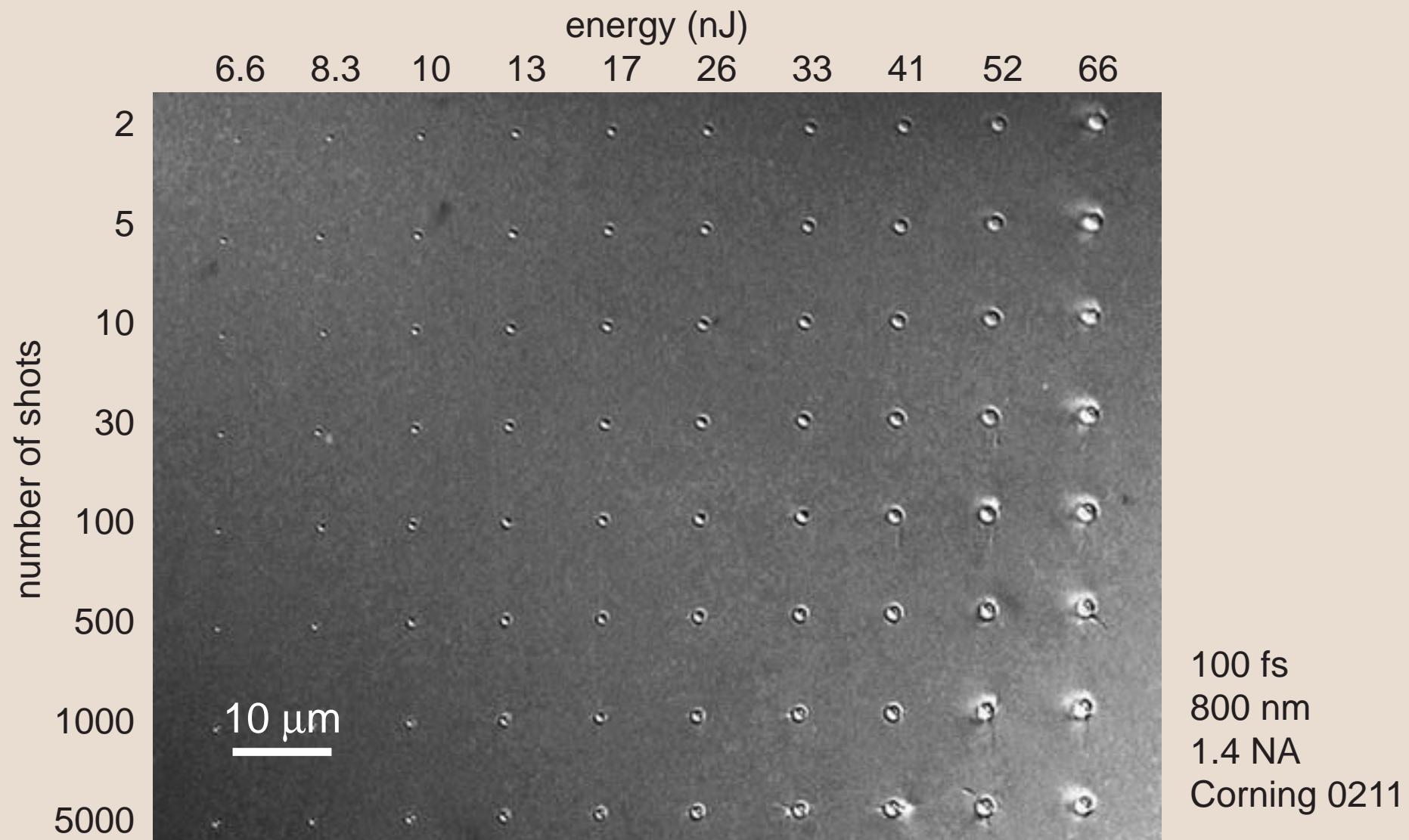
MORPHOLOGY

40 nJ
100 fs
800 nm
0.65 NA
Corning 0211



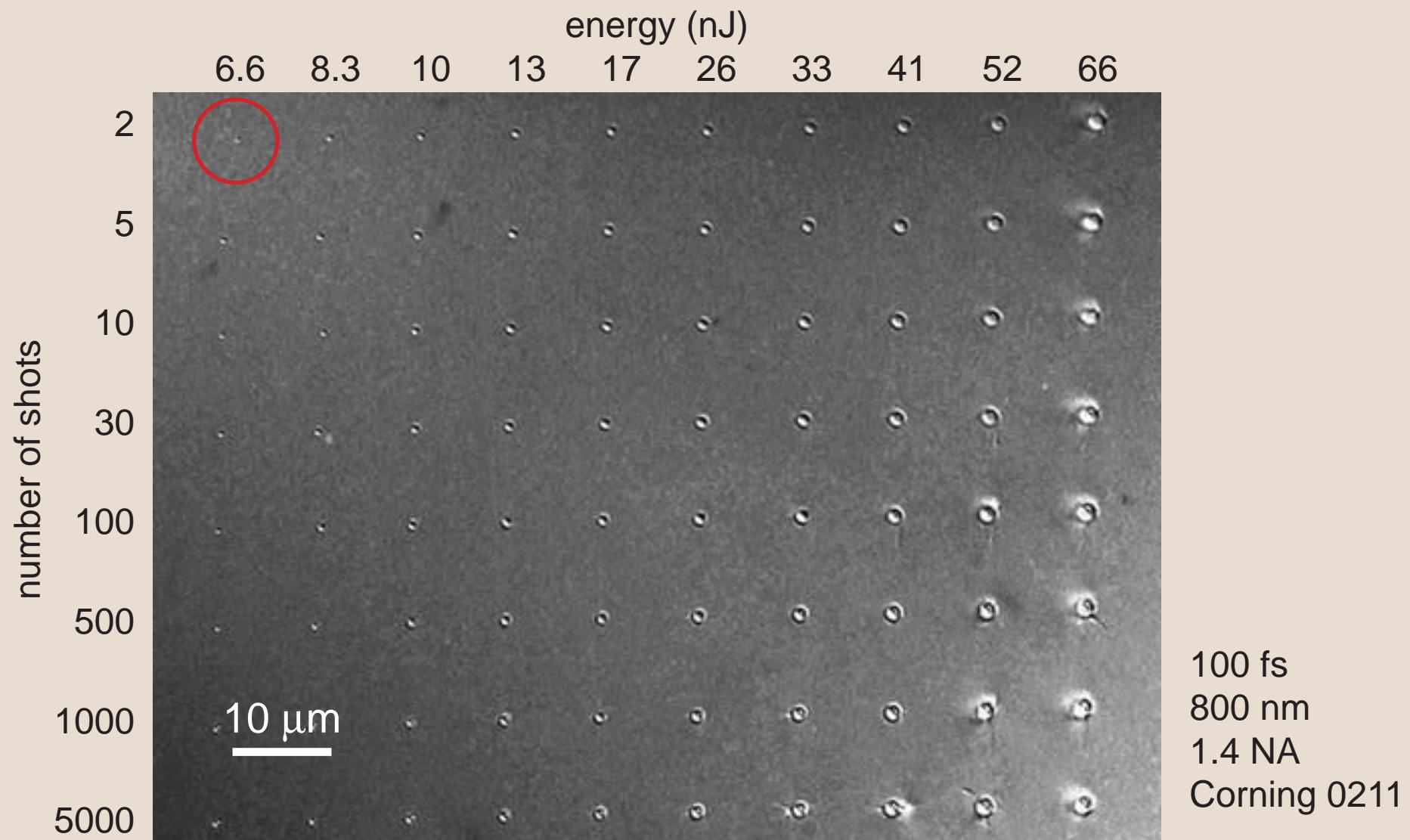
side view

AMPLIFIER MICROMACHINING



shot number and energy dependence at 1 kHz

AMPLIFIER MICROMACHINING



only 6.6 nJ; can we use only an oscillator?