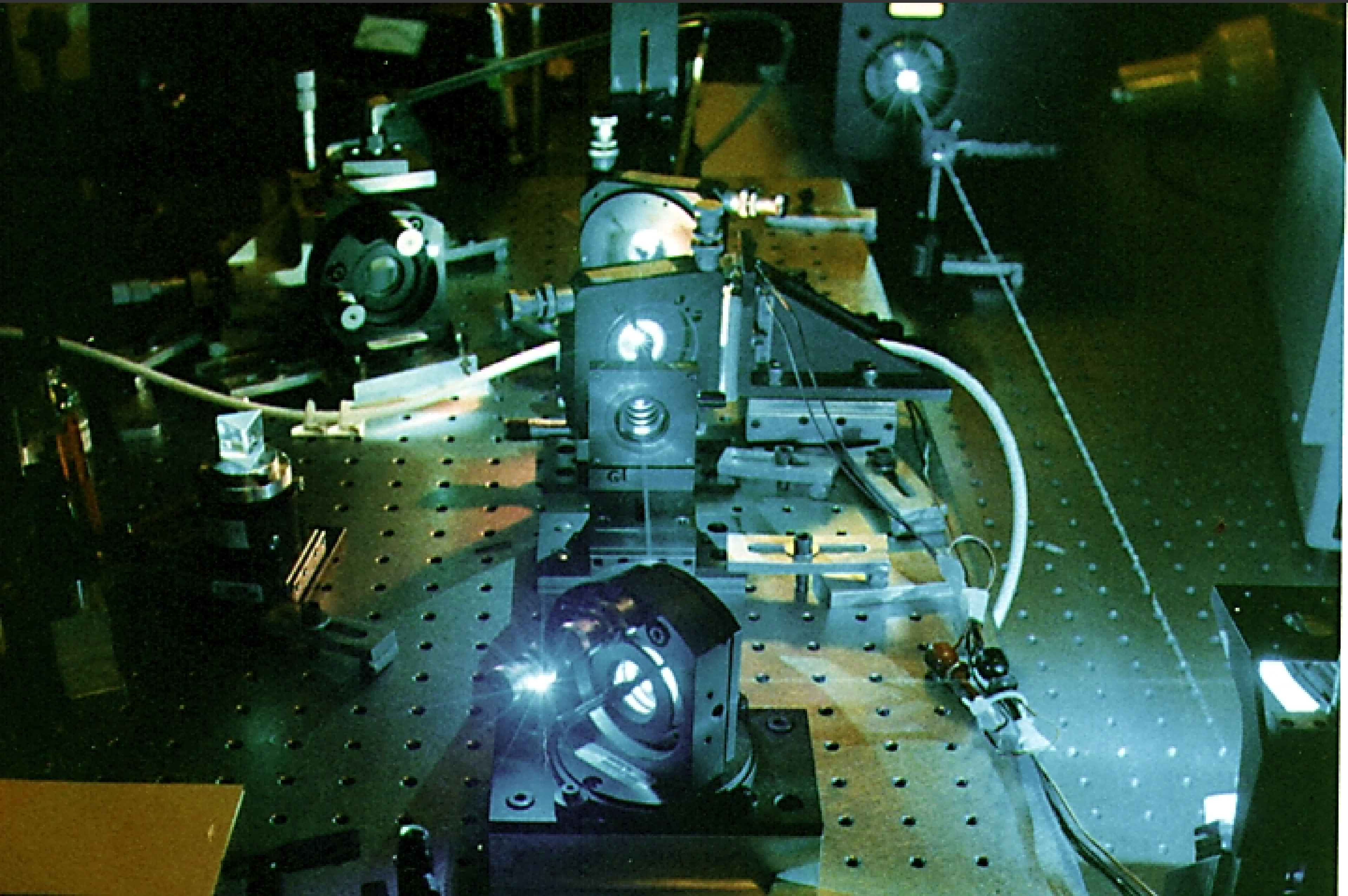
Phase Changes and Microexplosions Caused by Femtosecond Laser Pulses

Paul Callan
Albert Kim
Chris B. Schaffer
André Brodeur
José Garcia
Eric Mazur

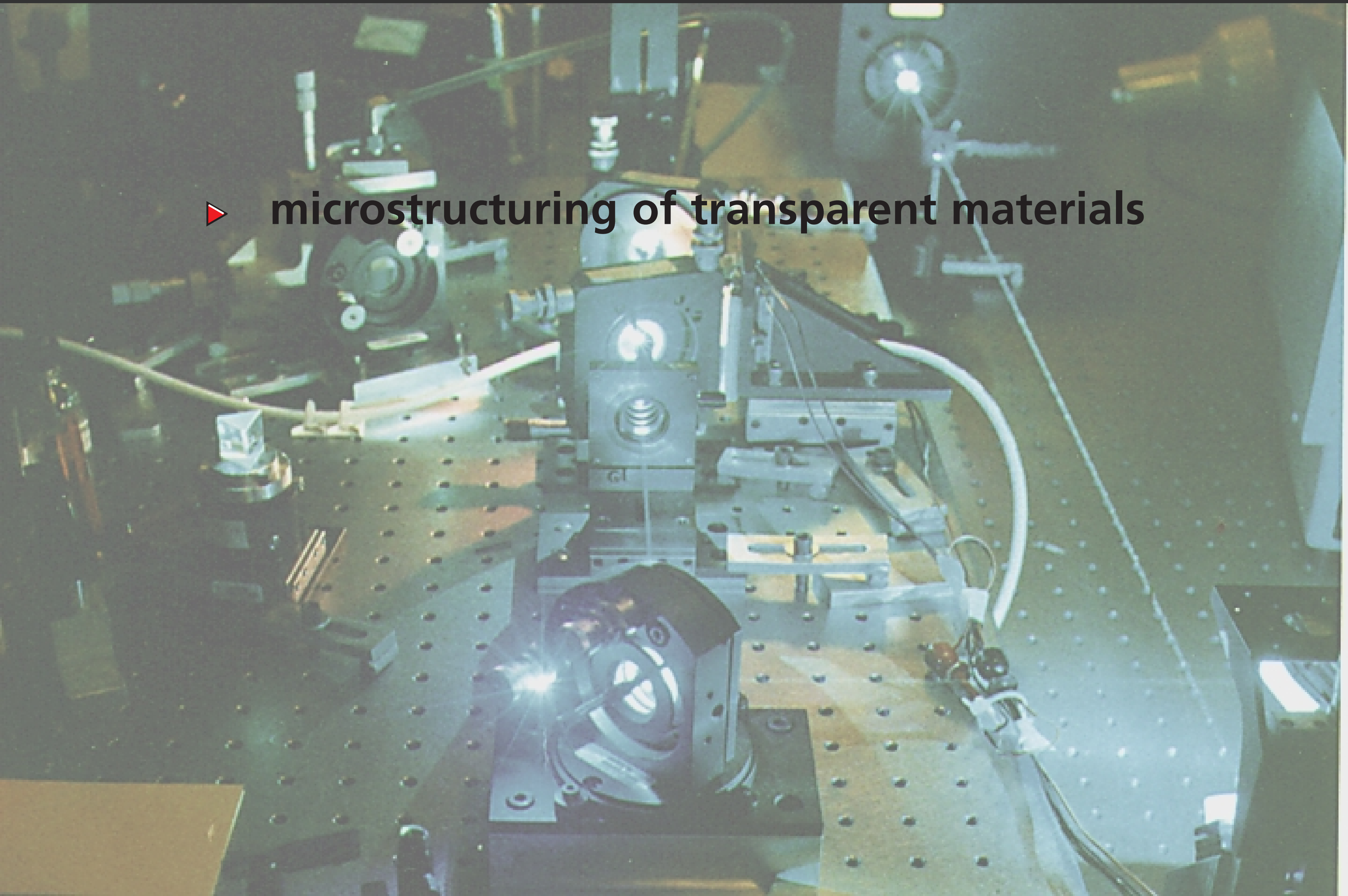
University of Illinois
21 January 2000

Introduction

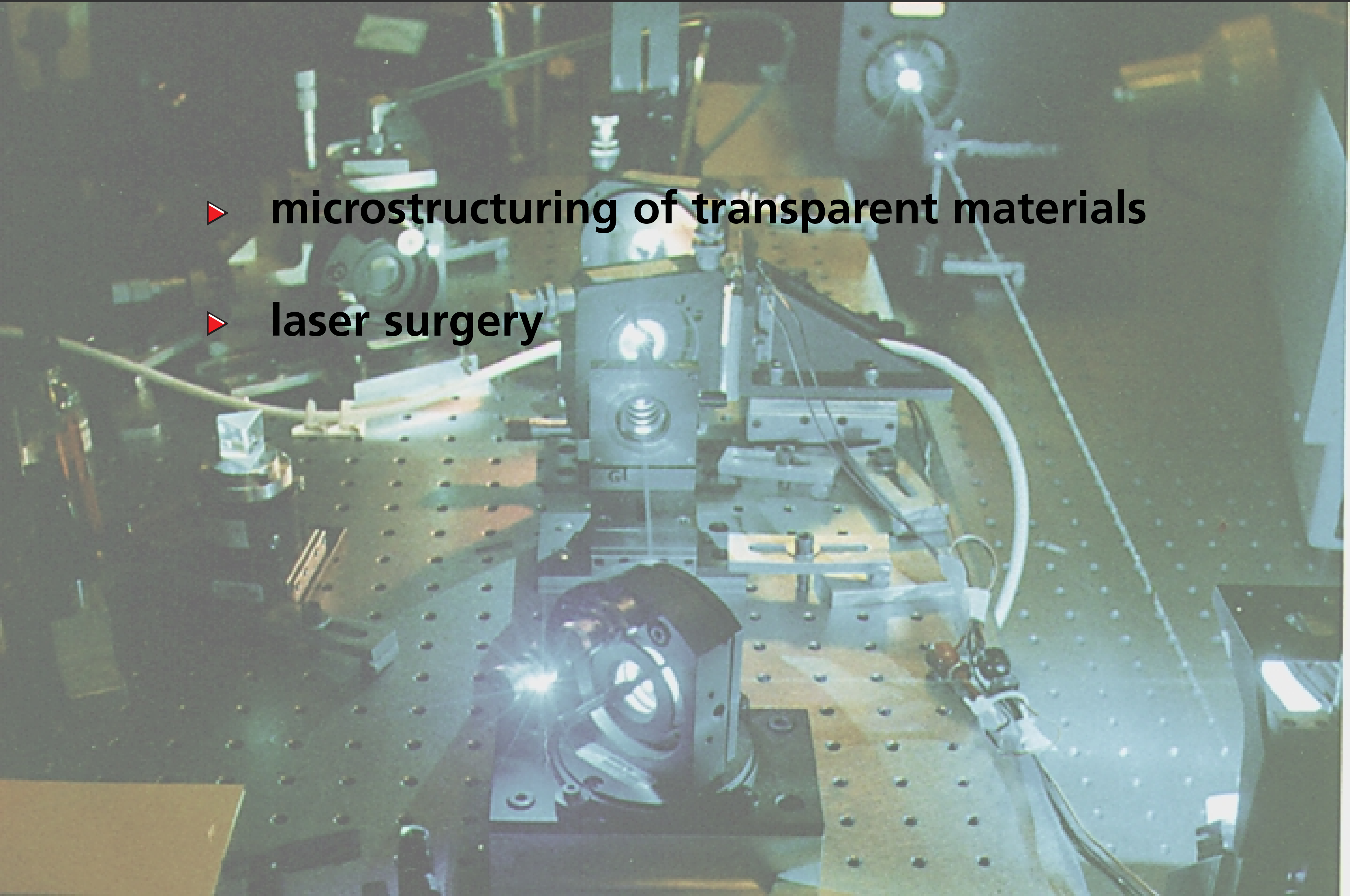


Introduction

- microstructuring of transparent materials



Introduction

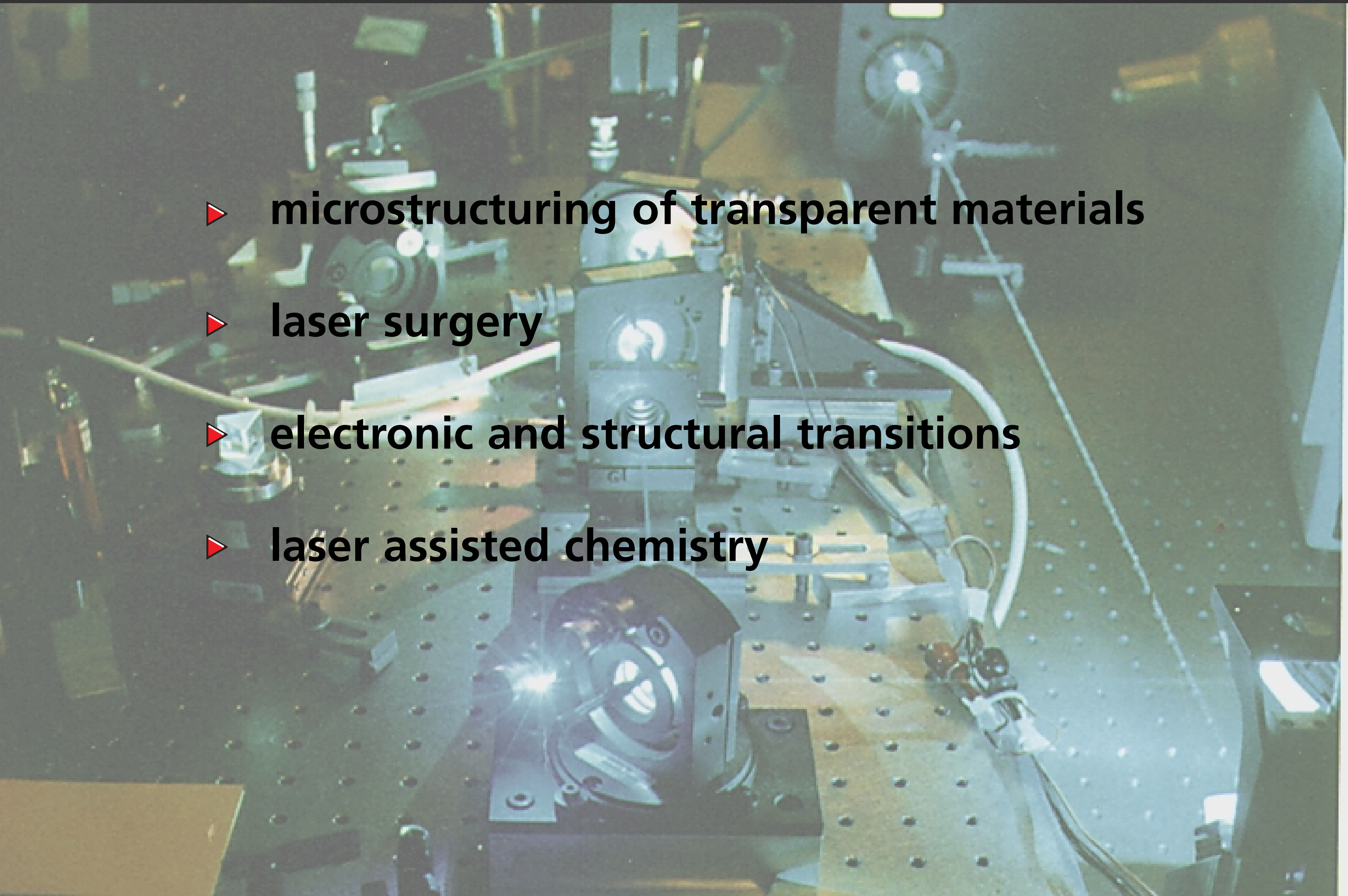


- microstructuring of transparent materials
- laser surgery

Introduction

- microstructuring of transparent materials
- laser surgery
- electronic and structural transitions

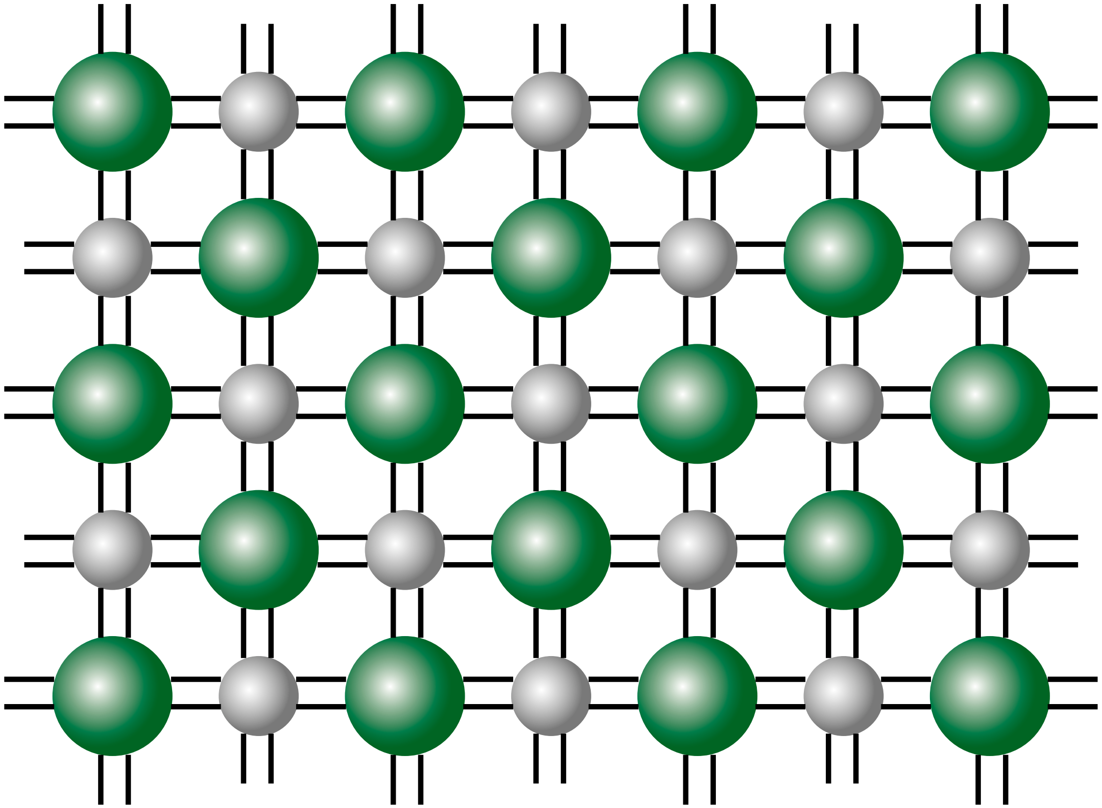
Introduction



- **microstructuring of transparent materials**
- **laser surgery**
- **electronic and structural transitions**
- **laser assisted chemistry**

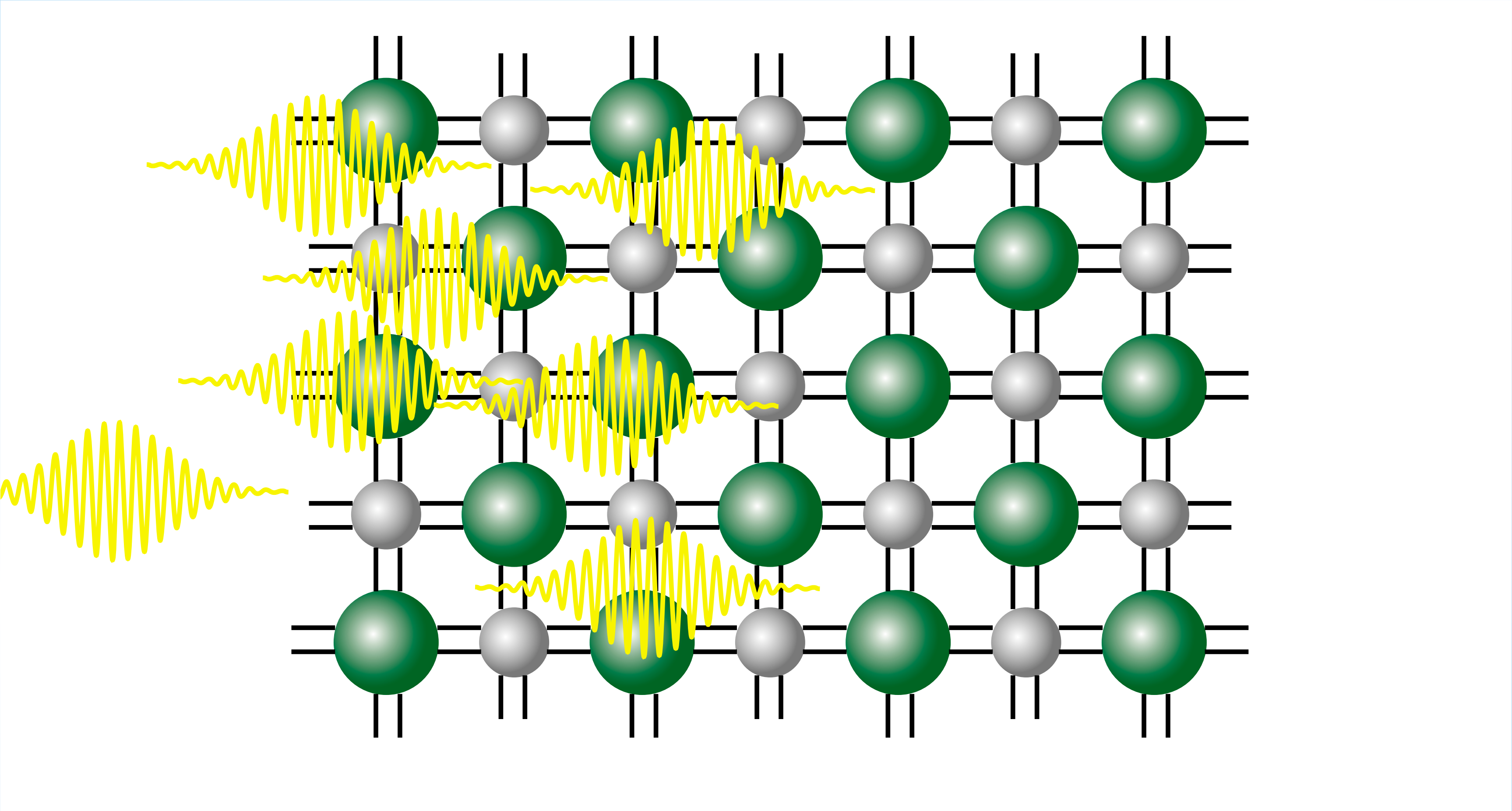
Introduction

how do femtosecond laser pulses alter a solid?



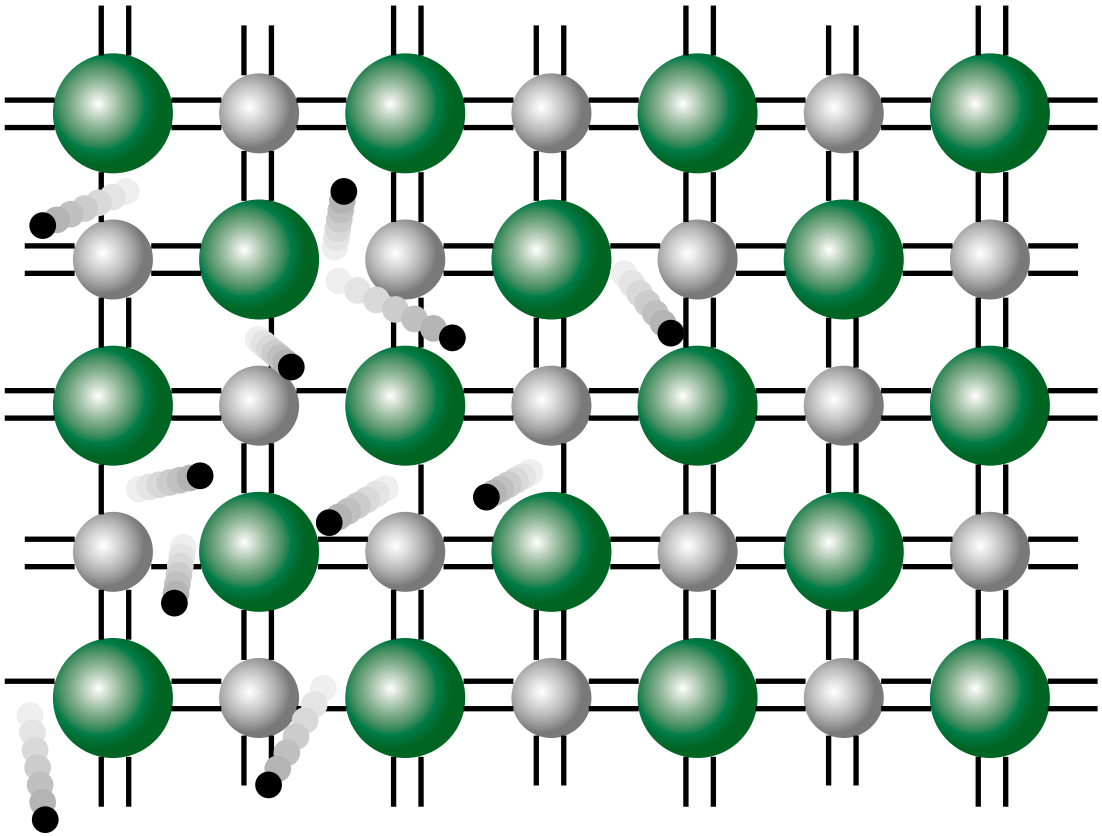
Introduction

photons excite valence electrons...



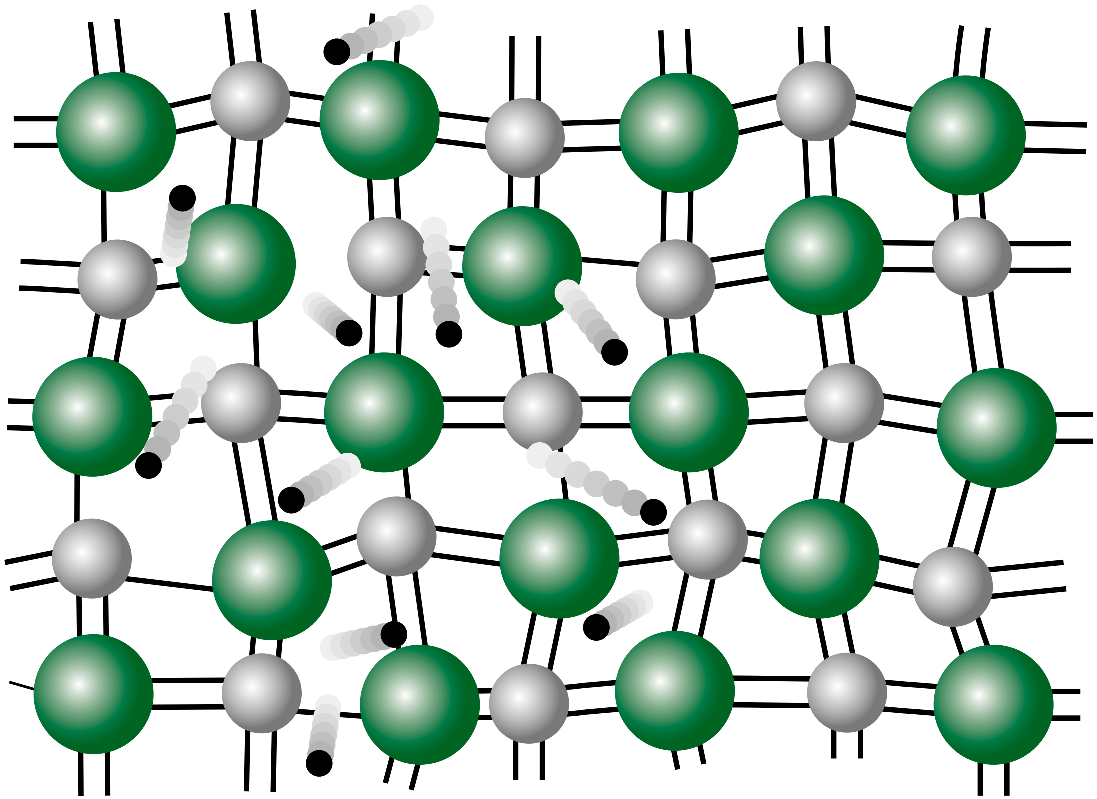
Introduction

…and create free electrons…

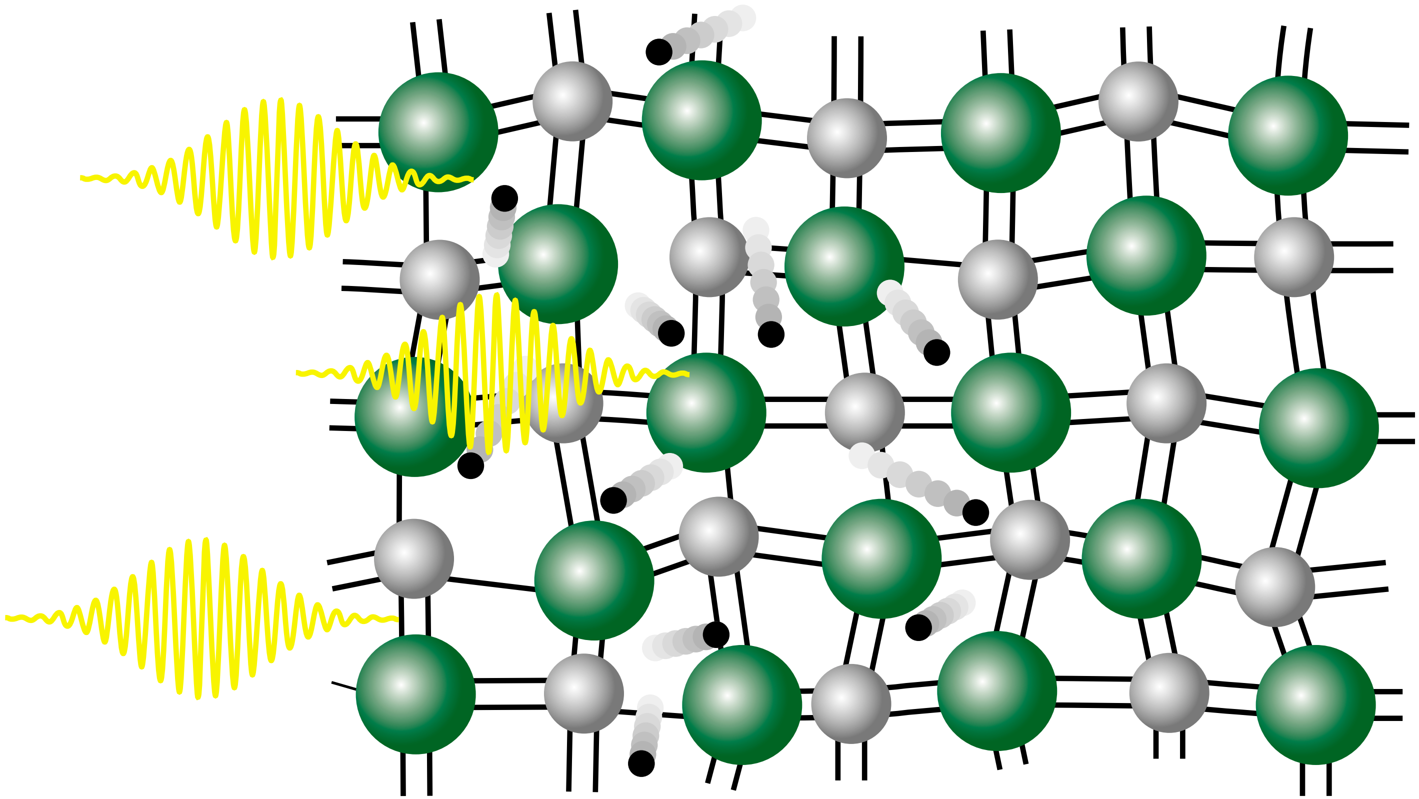


Introduction

…causing electronic and structural changes…

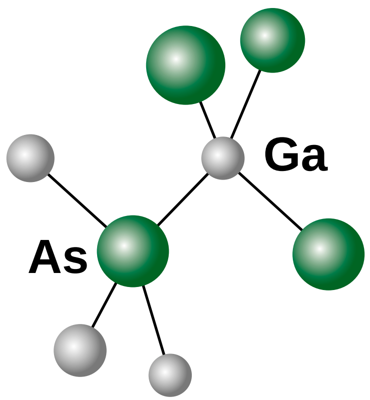


…which we measure with another pulse



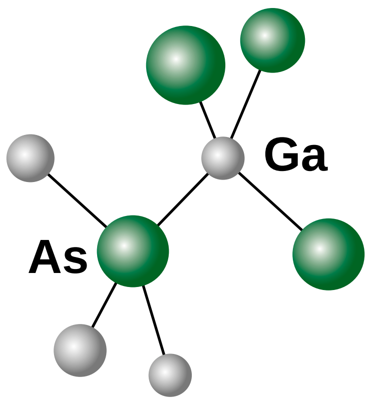
Introduction

structure

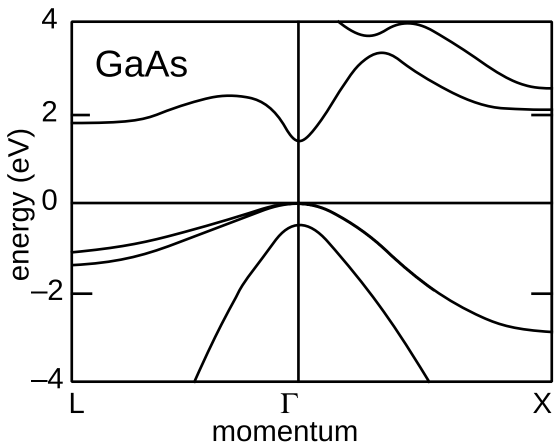


Introduction

structure

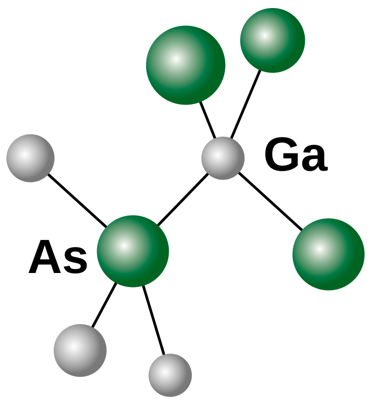


bandstructure

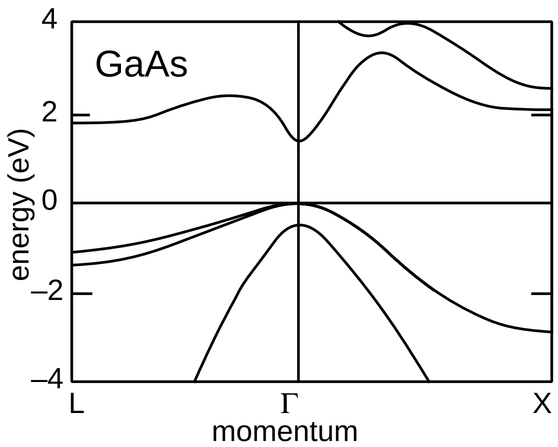


Introduction

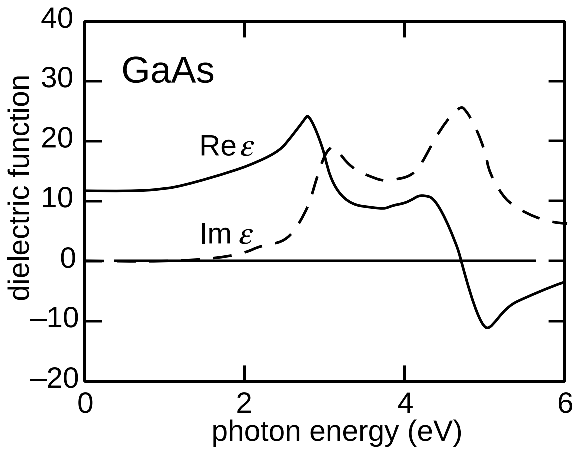
structure



bandstructure

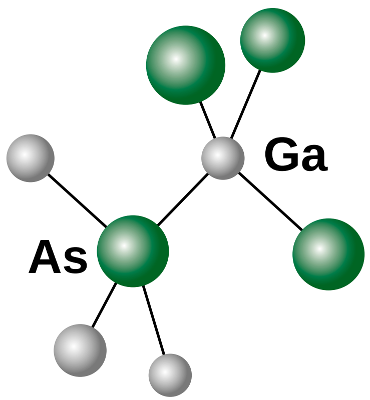


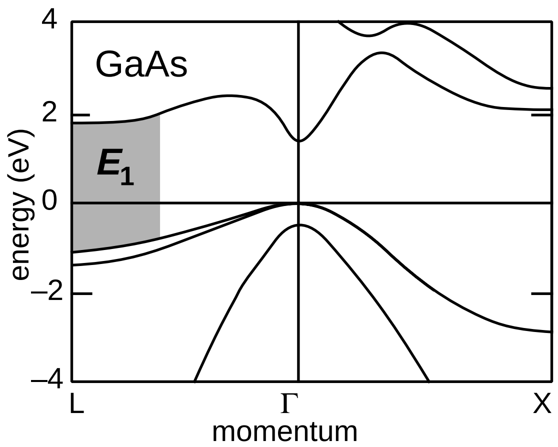
dielectric function

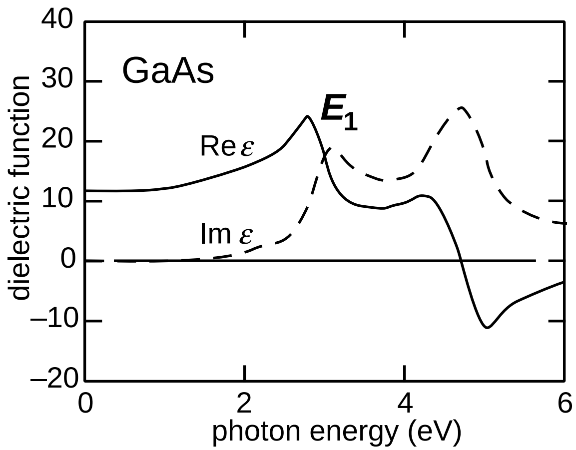


Introduction

structure | **bandstructure** | **dielectric function**

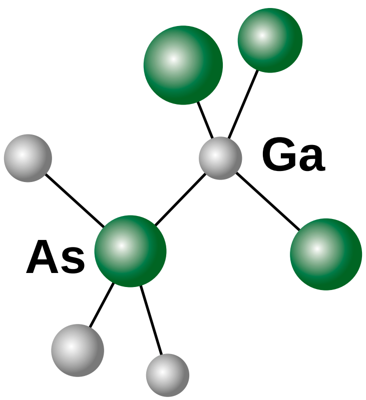




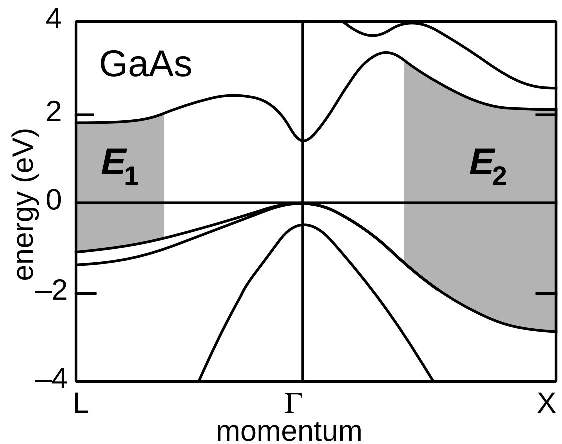


Introduction

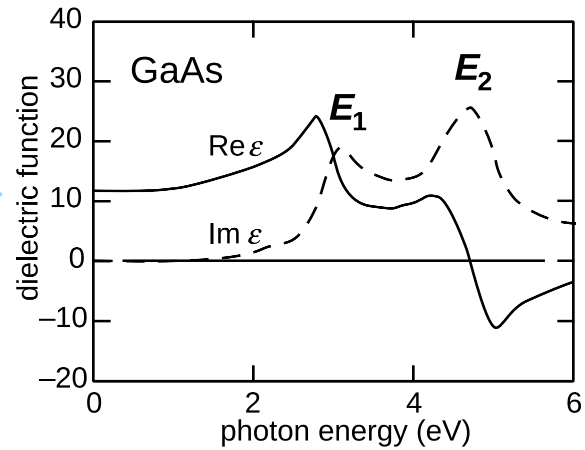
structure



bandstructure

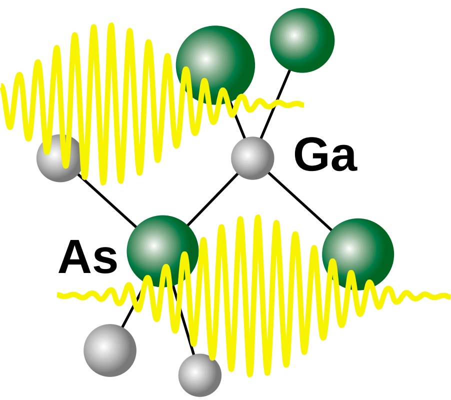


dielectric function



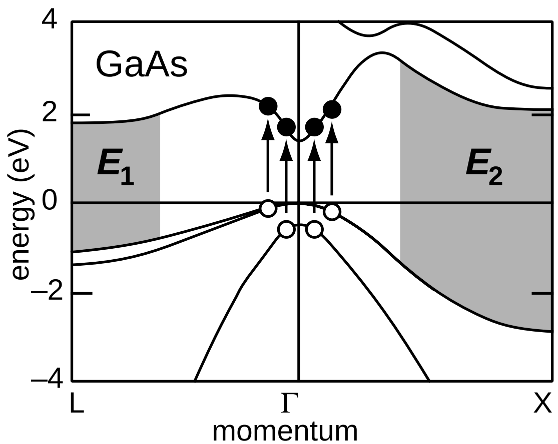
Introduction

structure



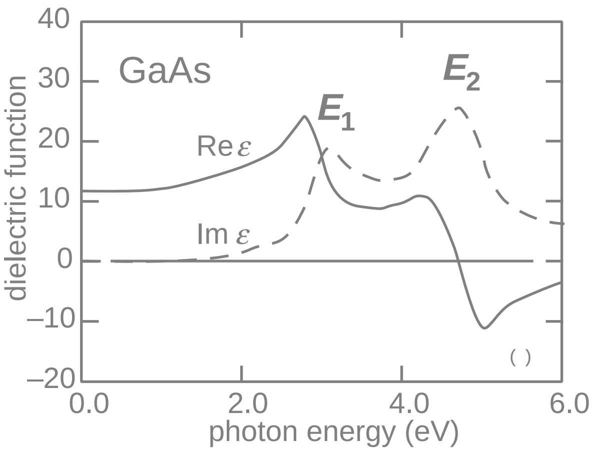
70 fs, 10^{16} W/m^2

bandstructure



10^{20} – 10^{22} cm^{-3}

dielectric function

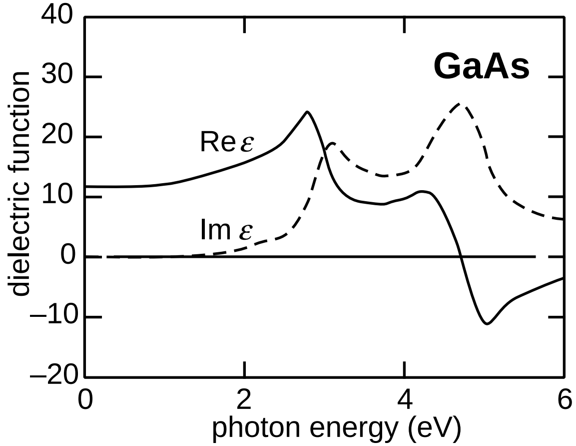


Introduction

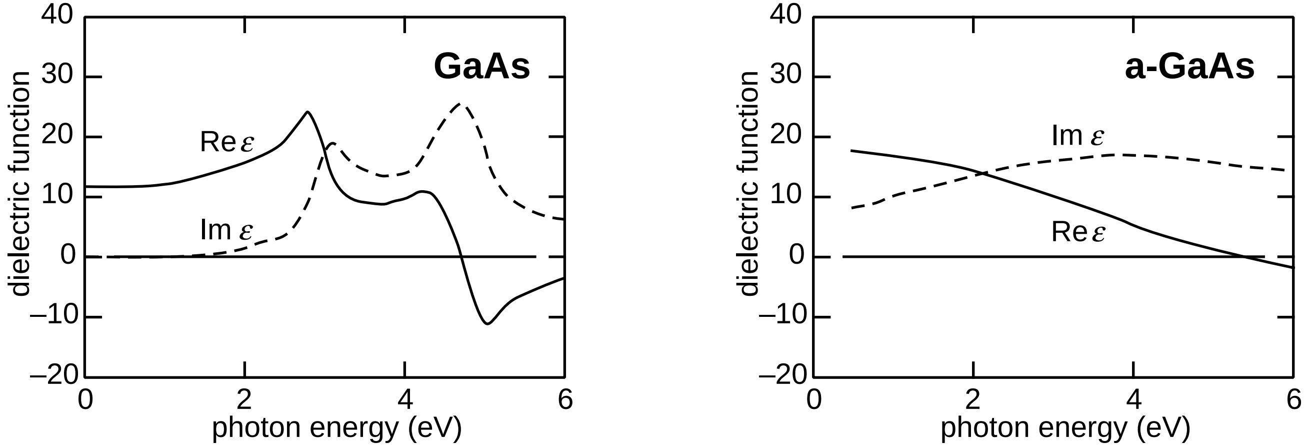
structure **bandstructure** **dielectric function**

? **?** **?**

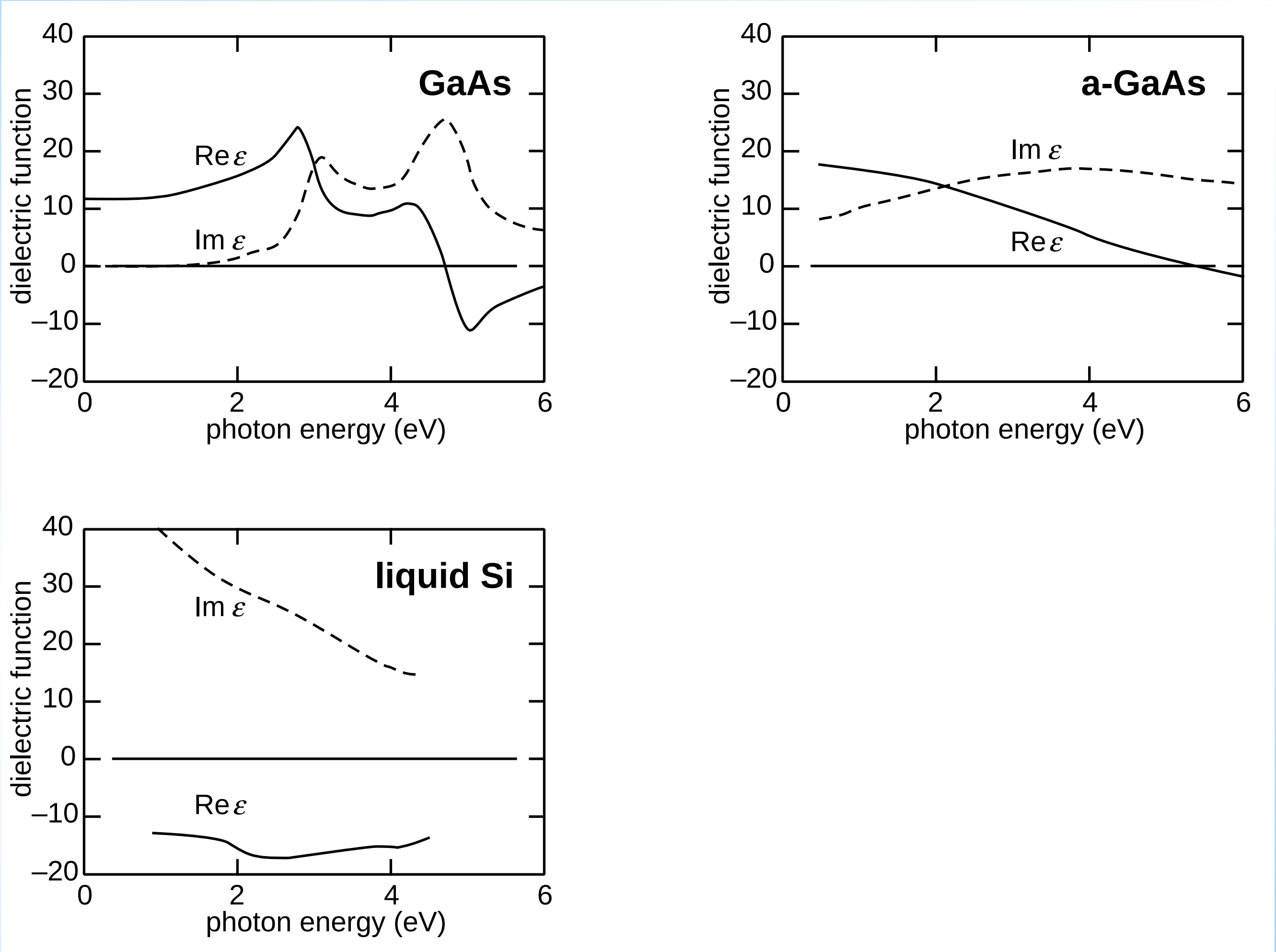
Introduction



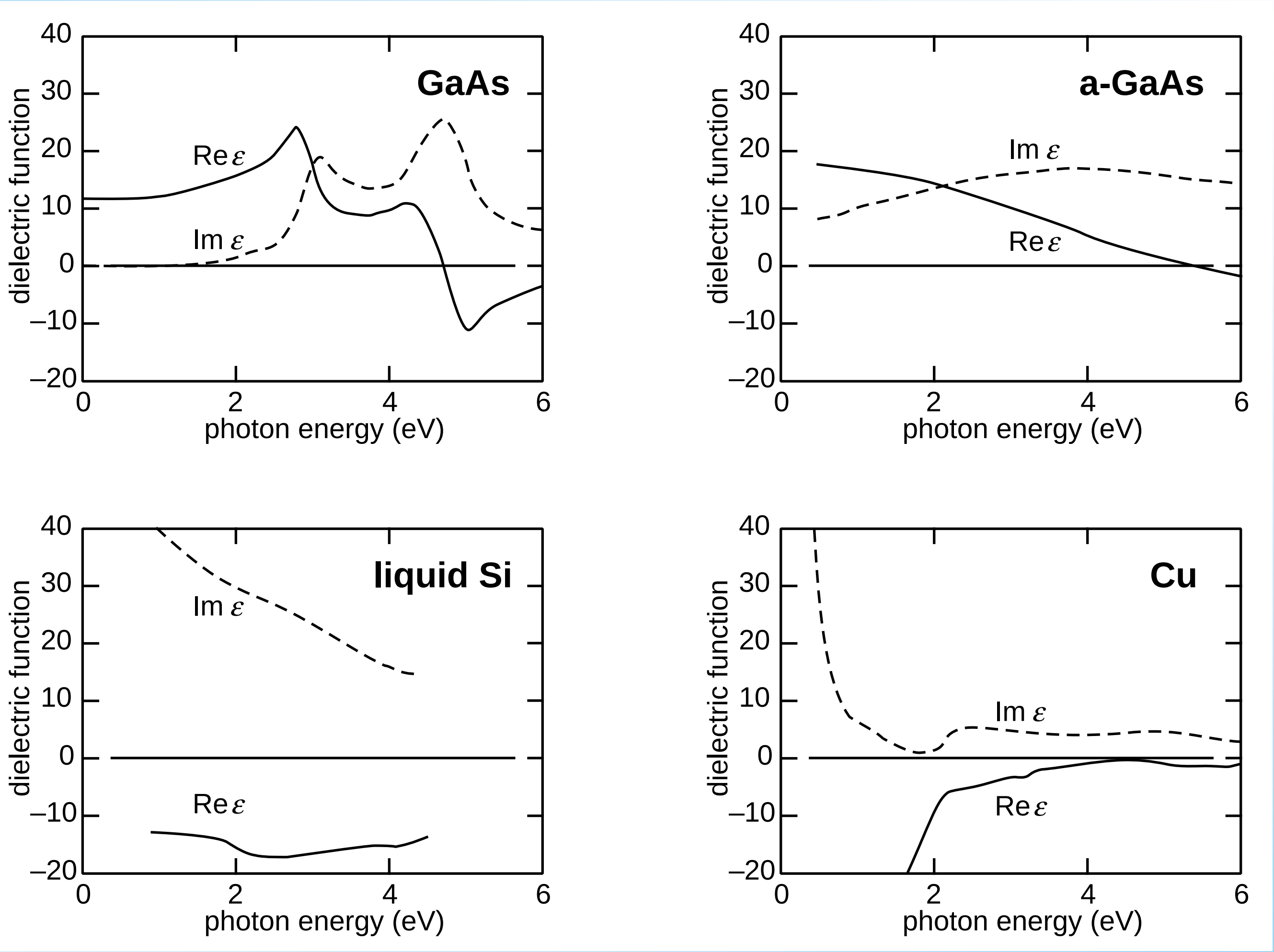
Introduction



Introduction



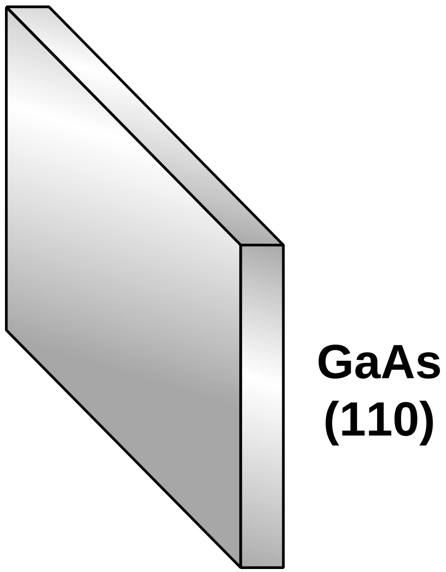
Introduction



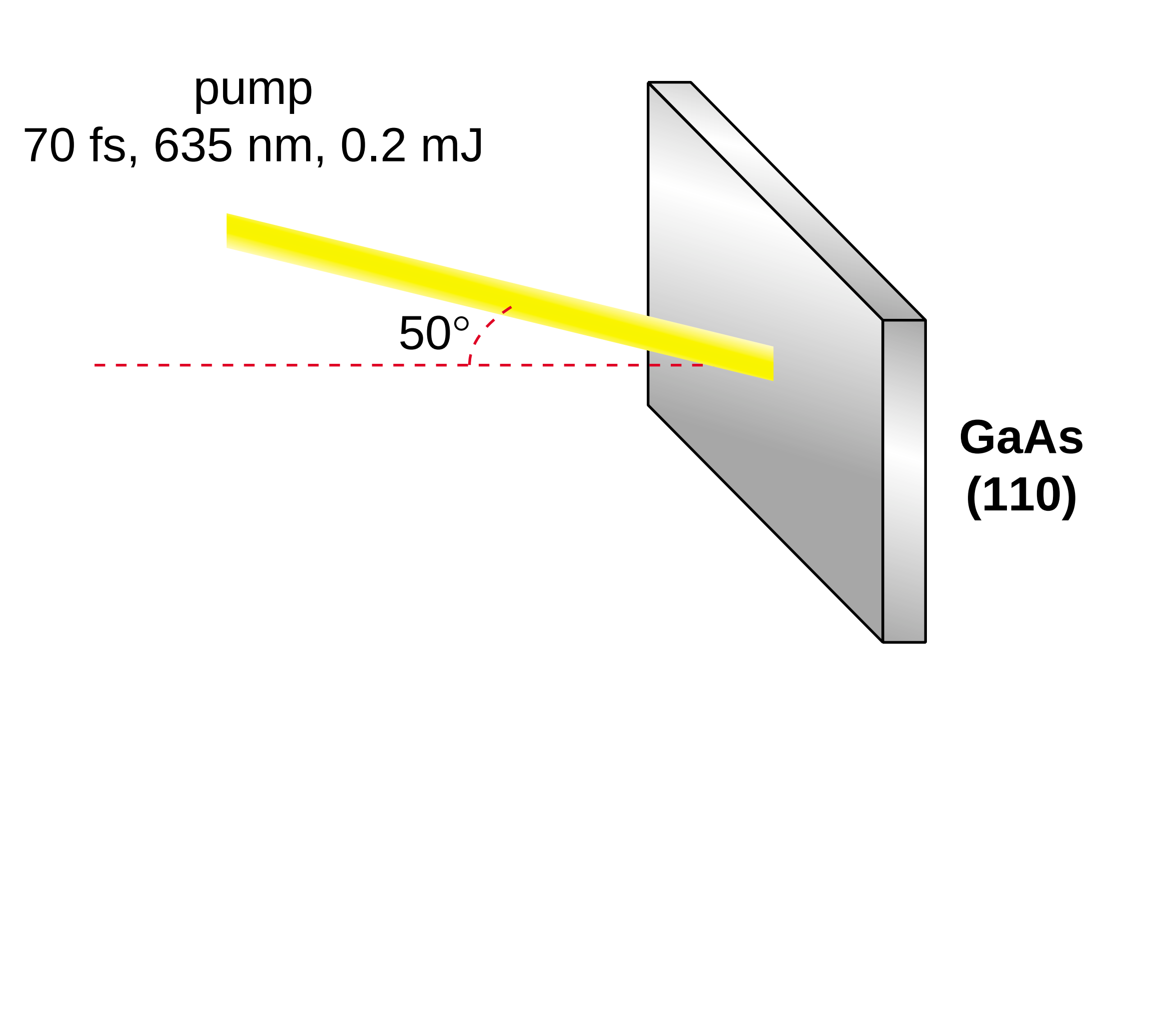
Outline

- **Method**
- **Results**
- **Discussion**

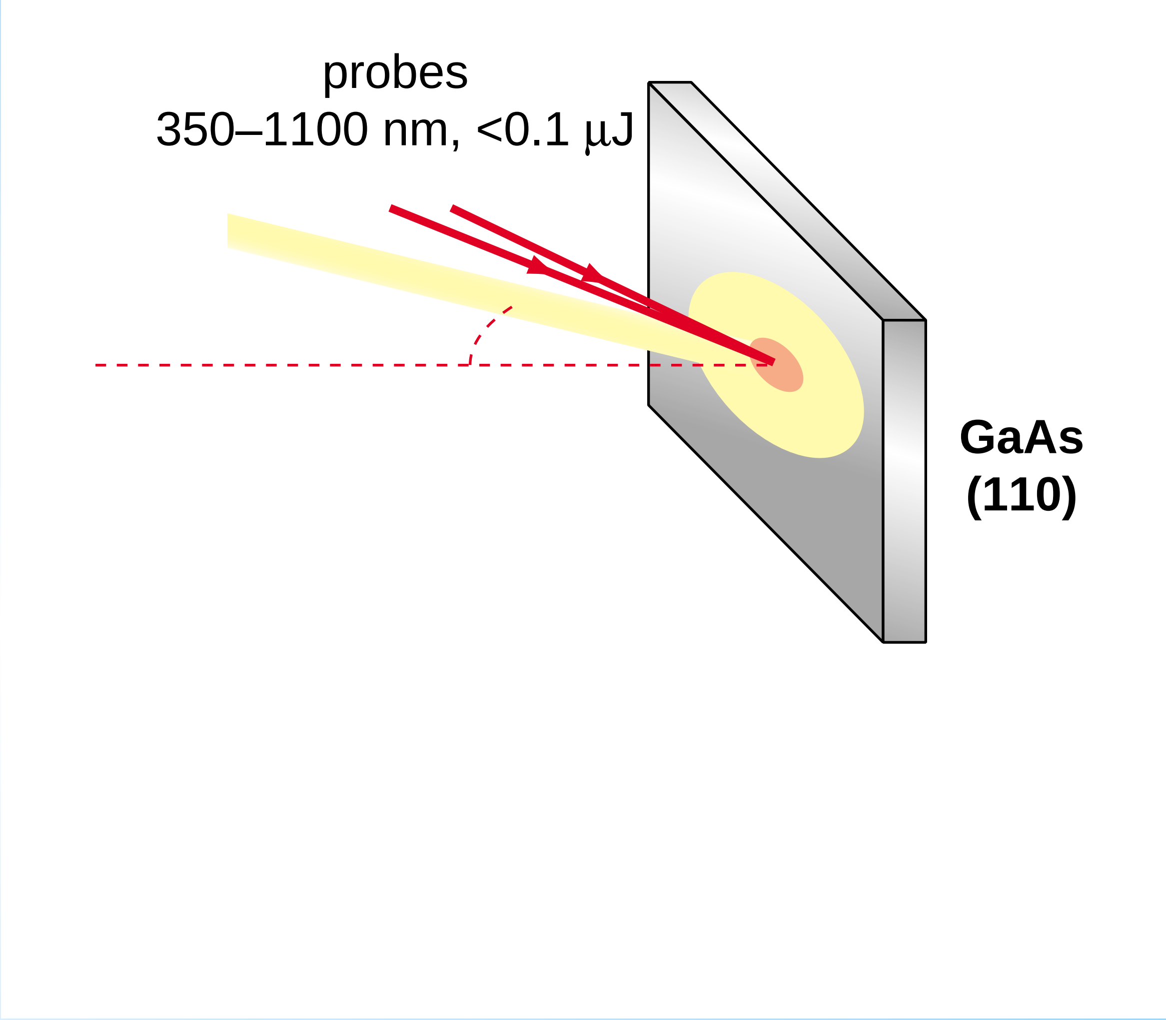
Method



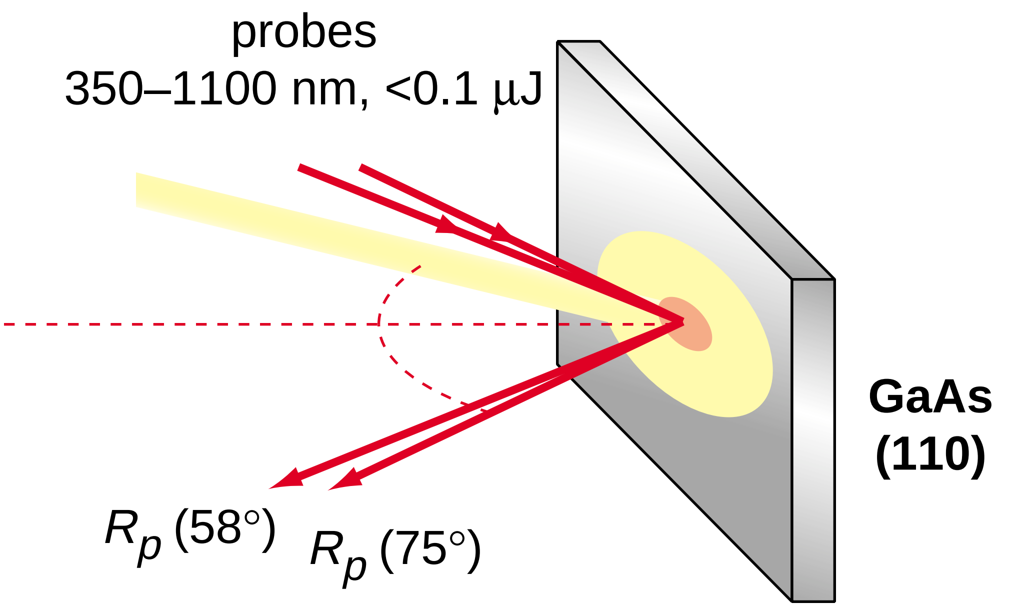
Method



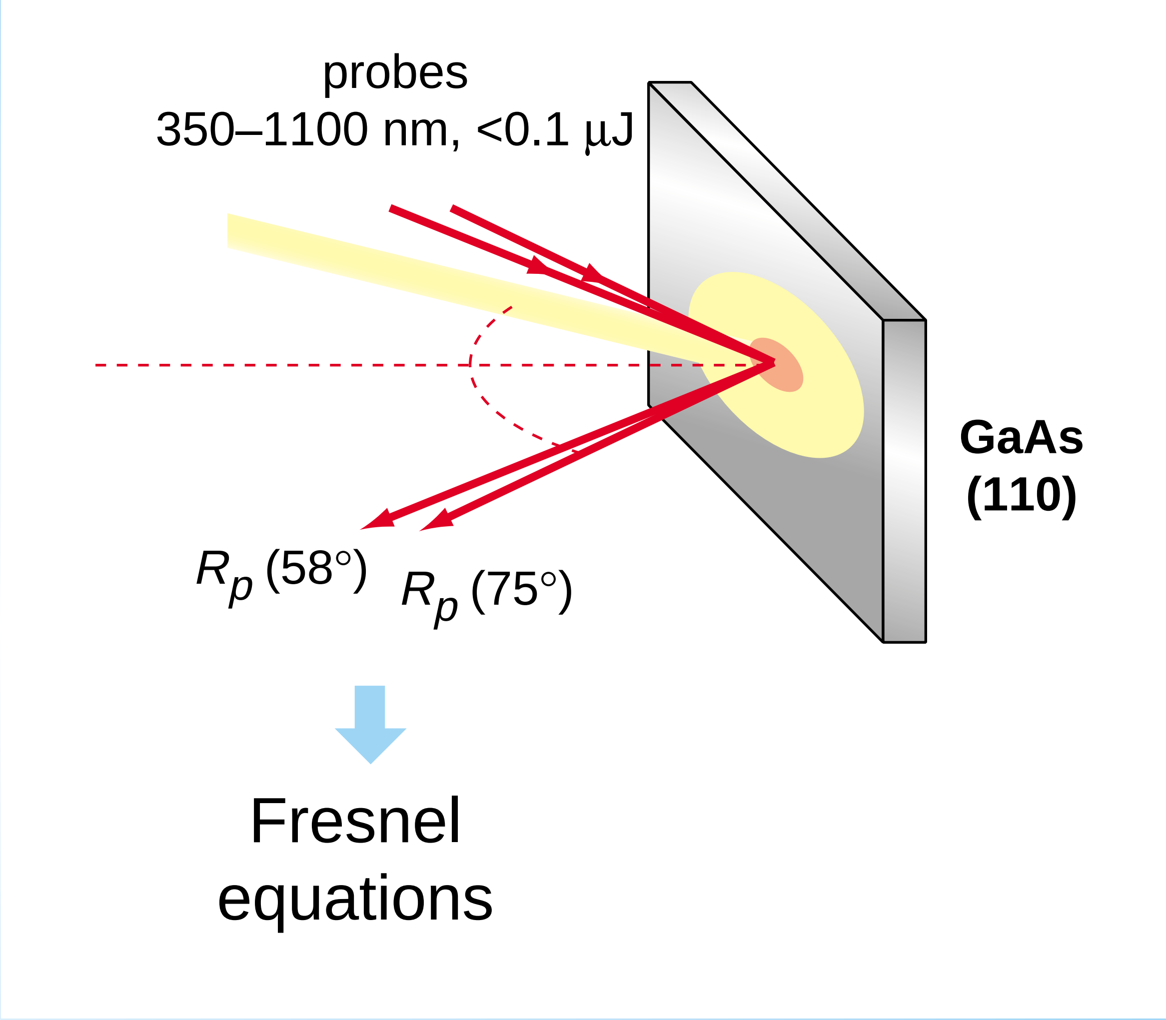
Method



Method

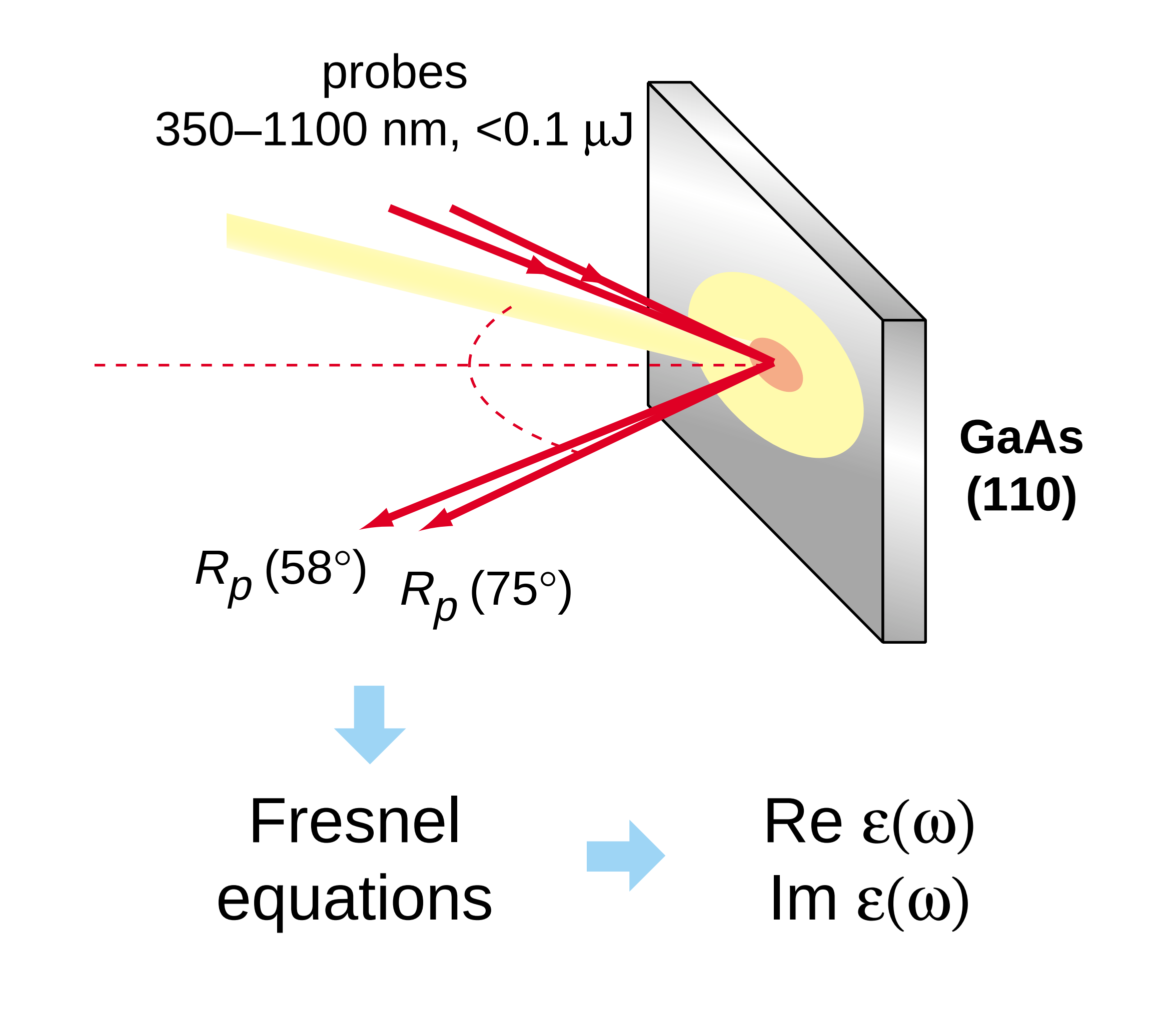


Method



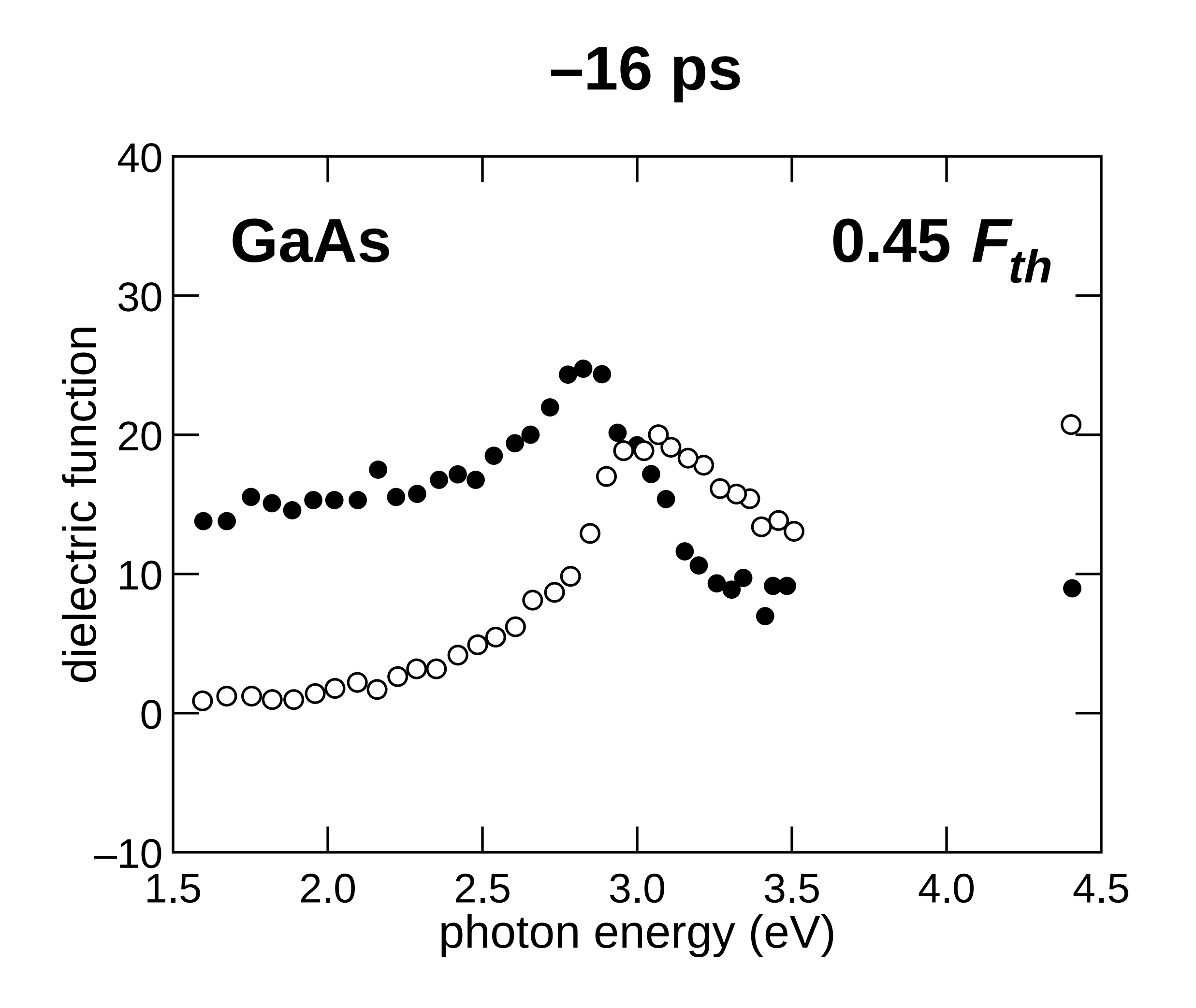
probes
350–1100 nm, <0.1 μJ
GaAs
(110)
R_p (58°)
R_p (75°)
Fresnel
equations

Method


probes
350–1100 nm, <0.1 μJ
GaAs
(110)
Rp (58°)
Rp (75°)
Fresnel
equations
Re ε(ω)
Im ε(ω)

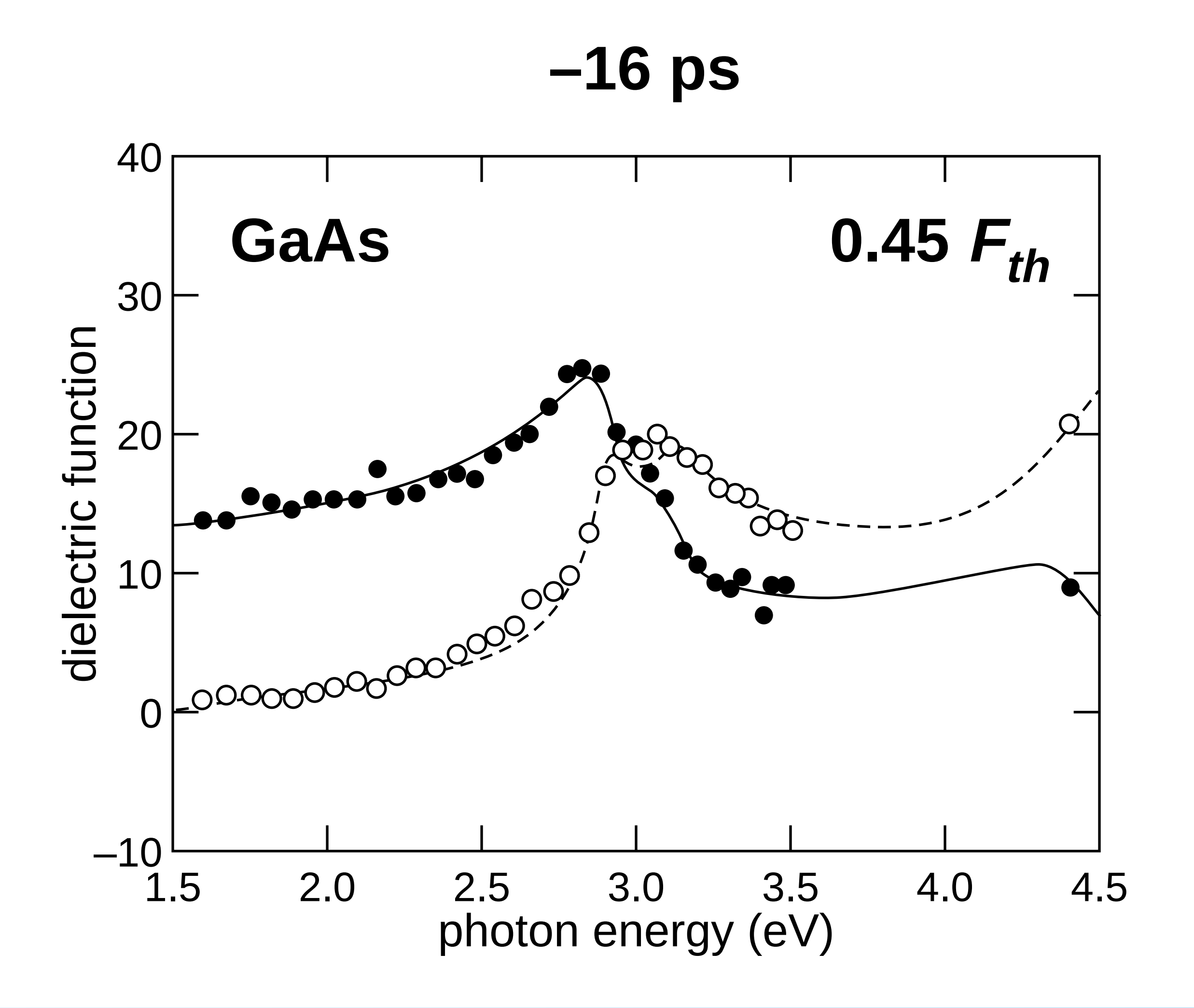
Method

Results



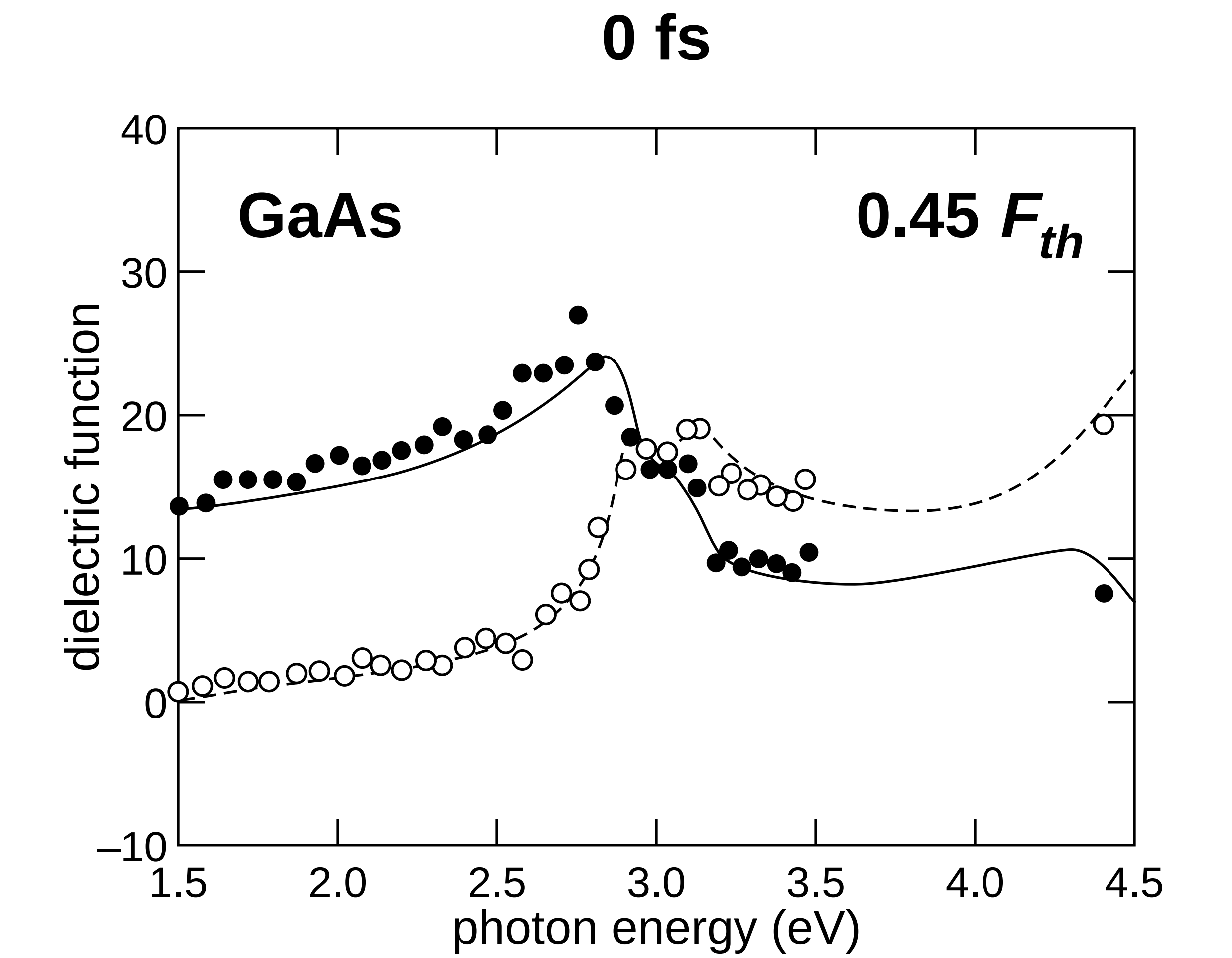
–16 ps
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



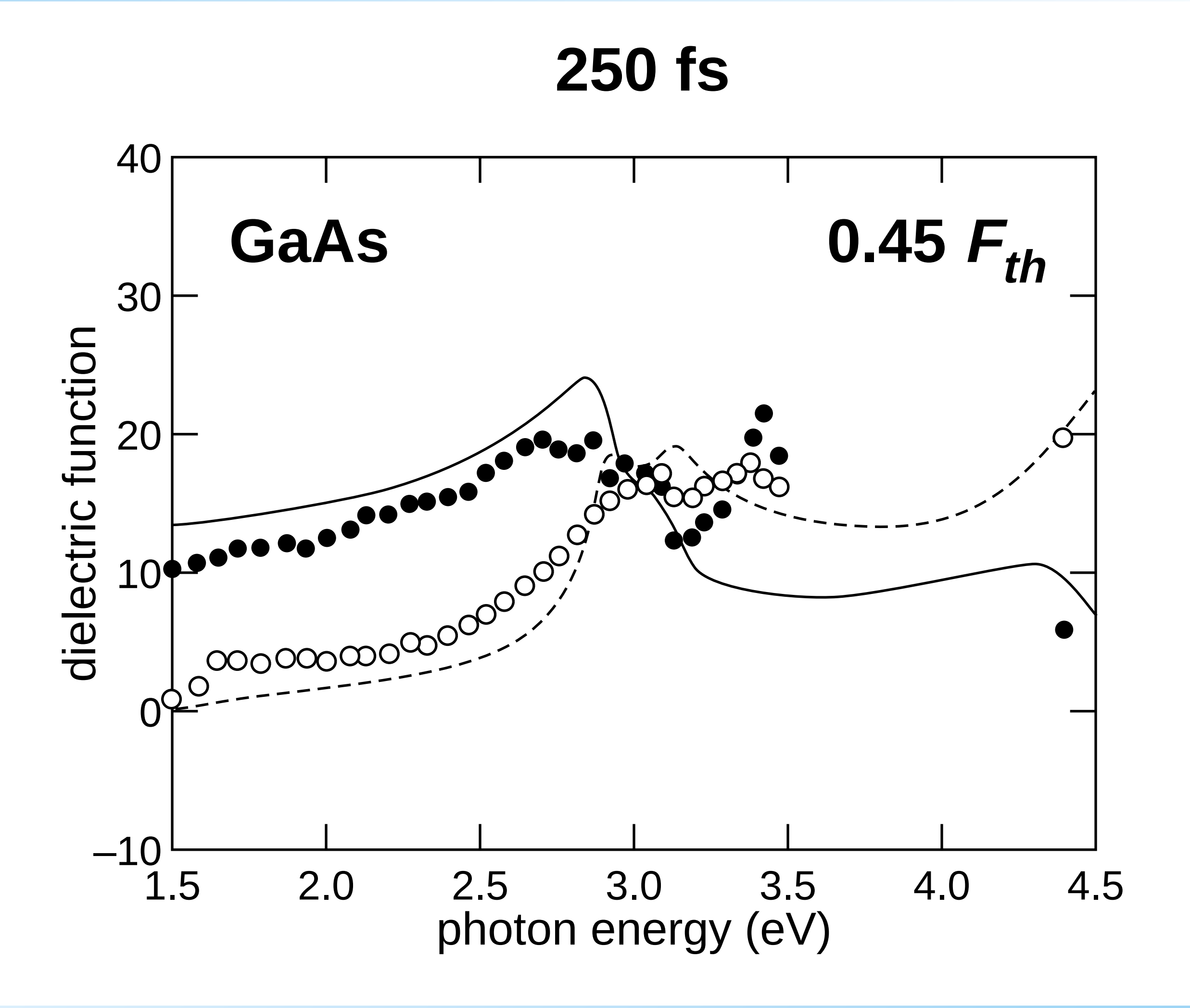
–16 ps
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results

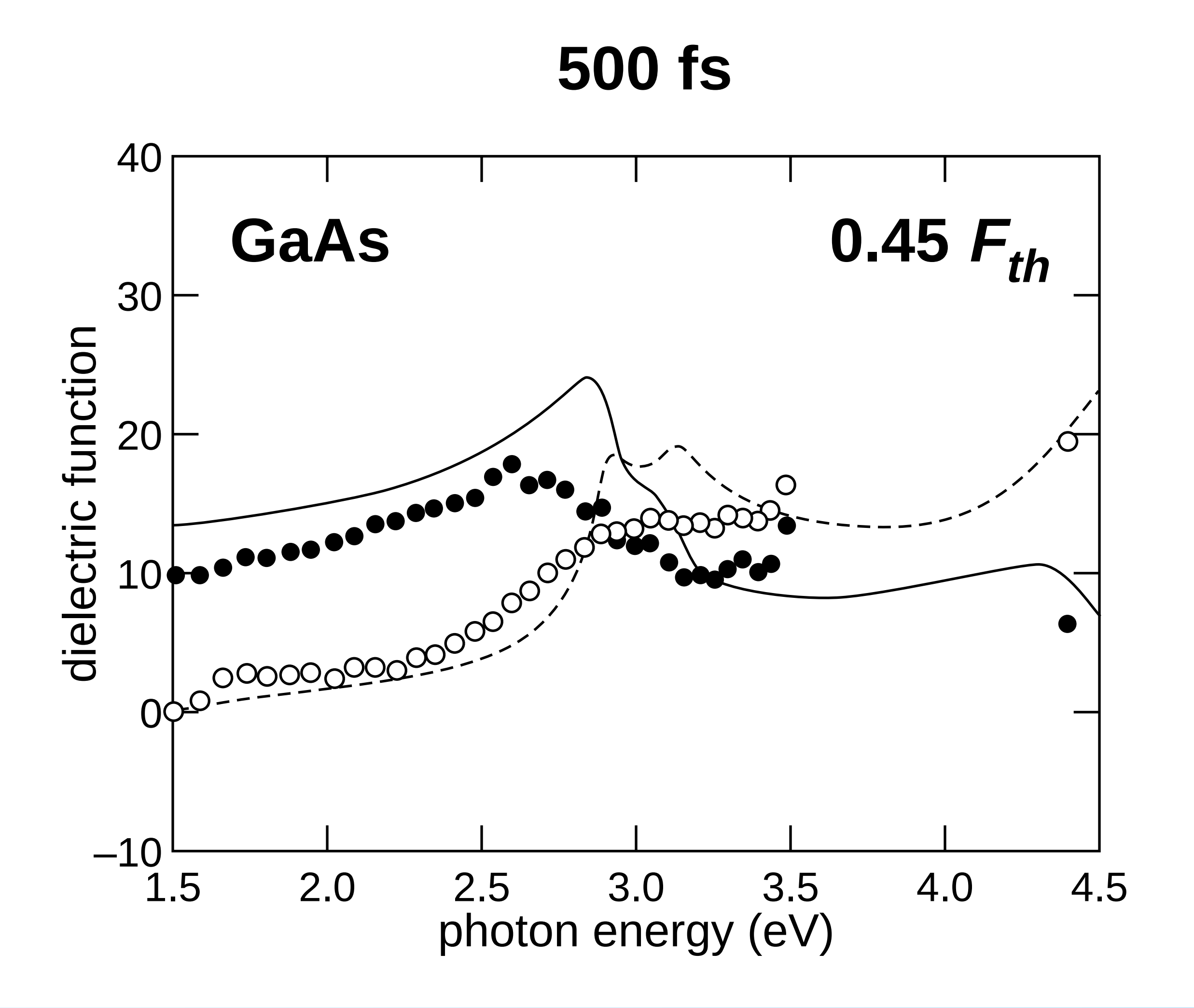


0 fs
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results

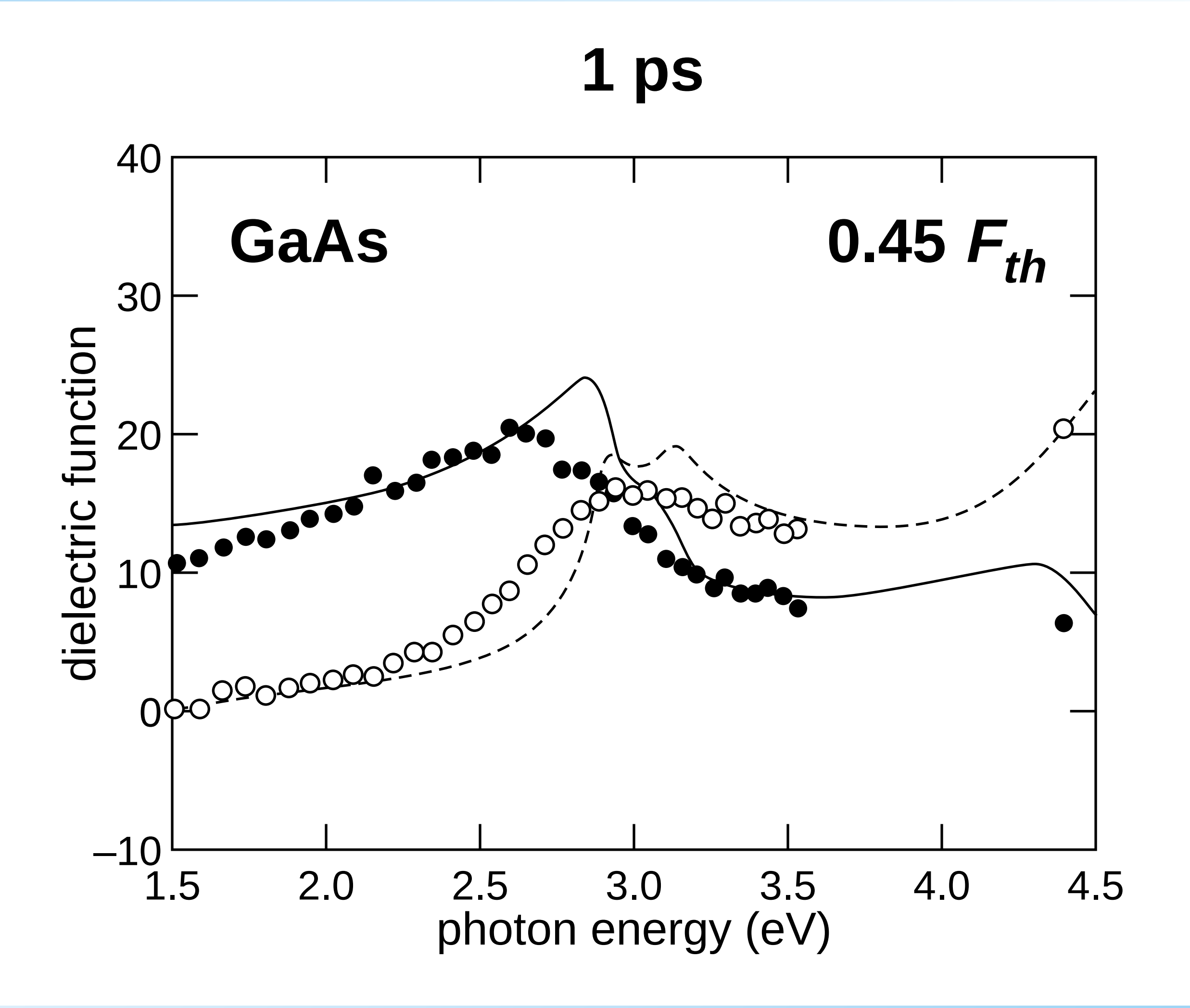

250 fs
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



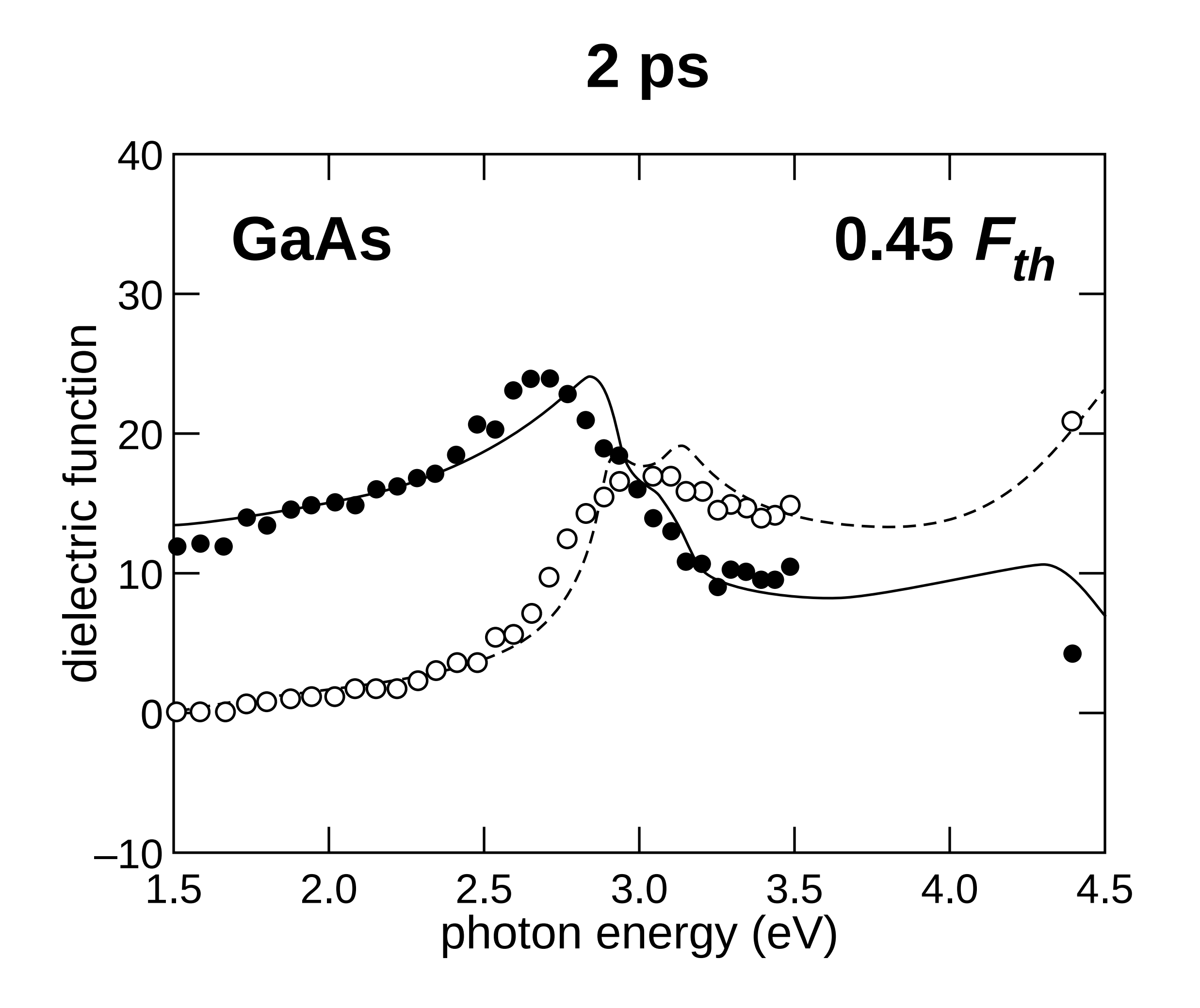
500 fs
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



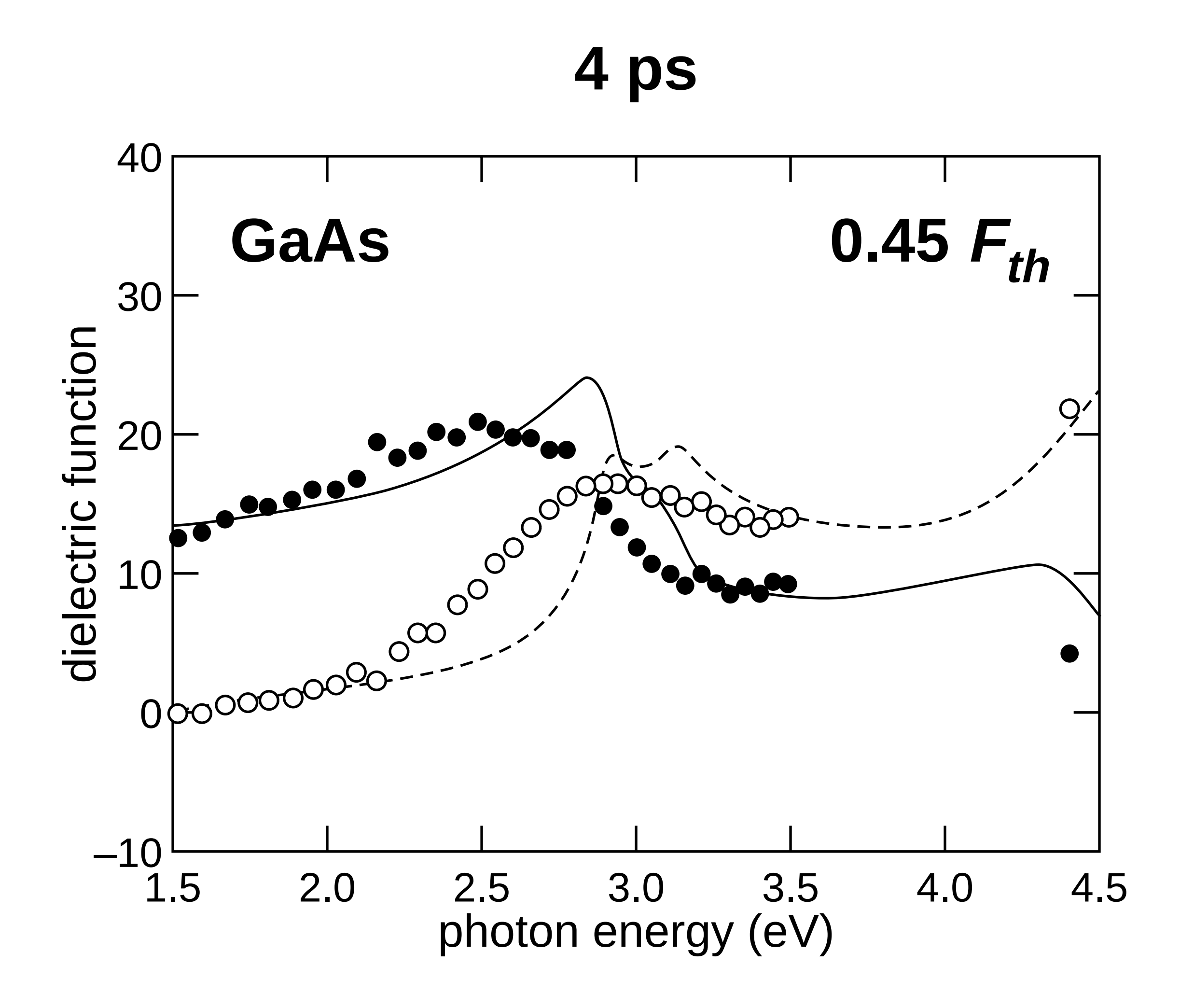
1 ps
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



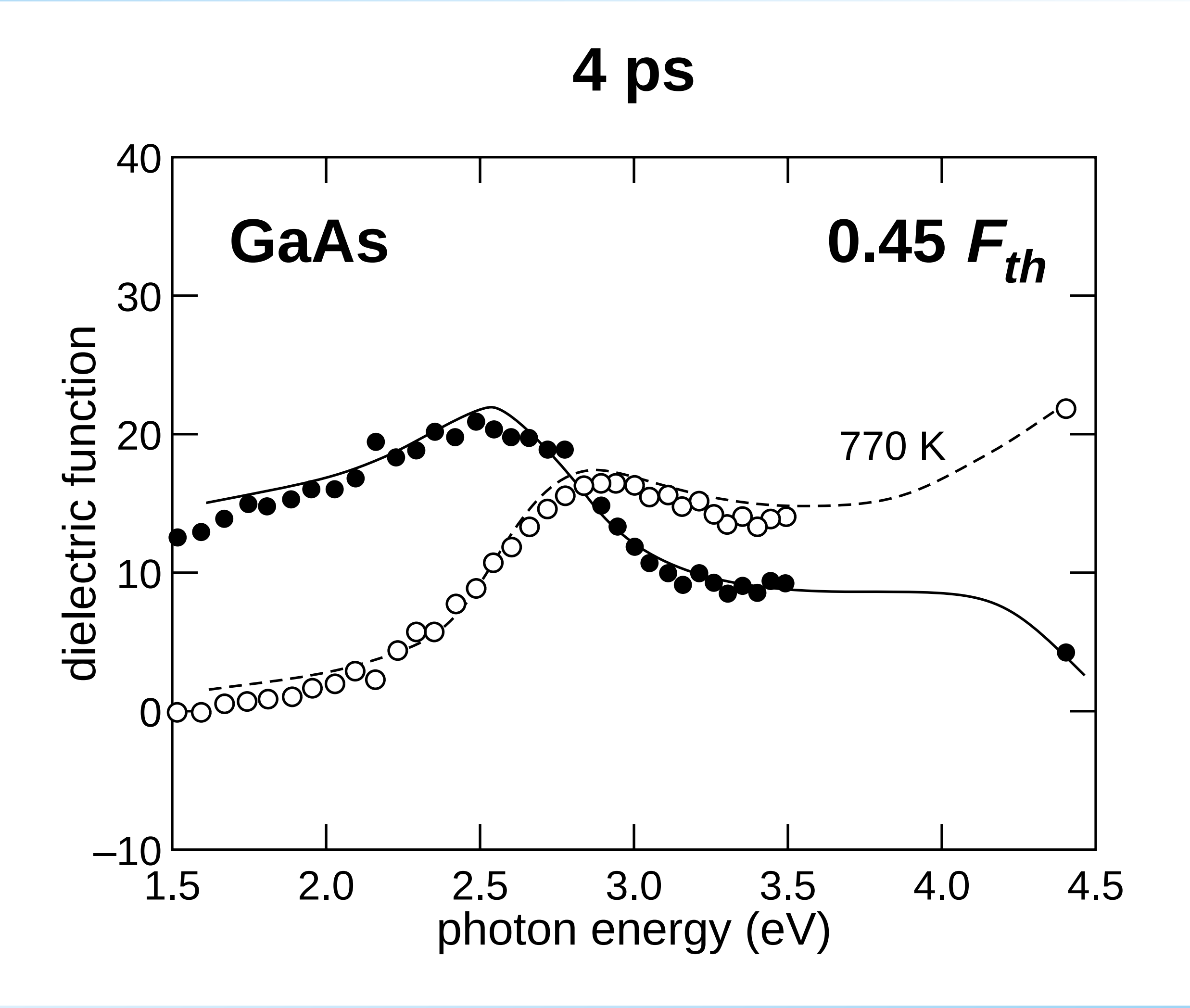
2 ps
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



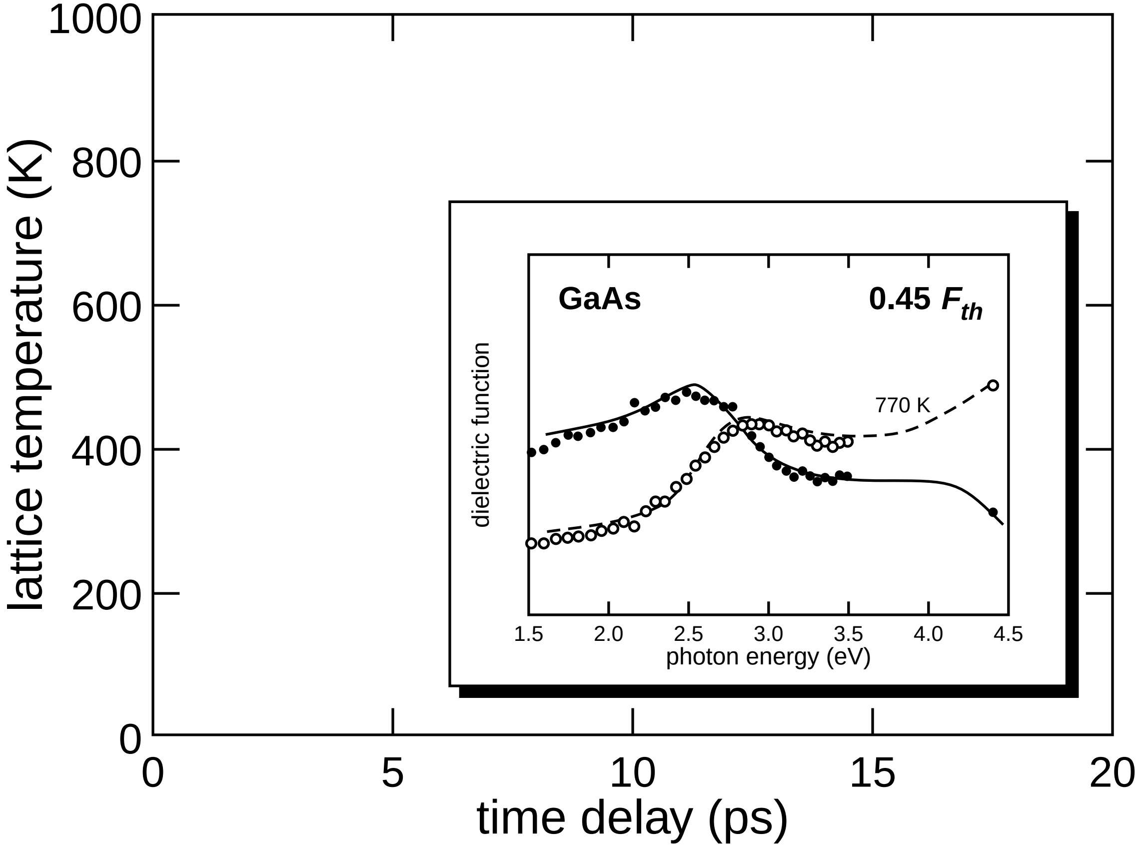
4 ps
GaAs
0.45 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



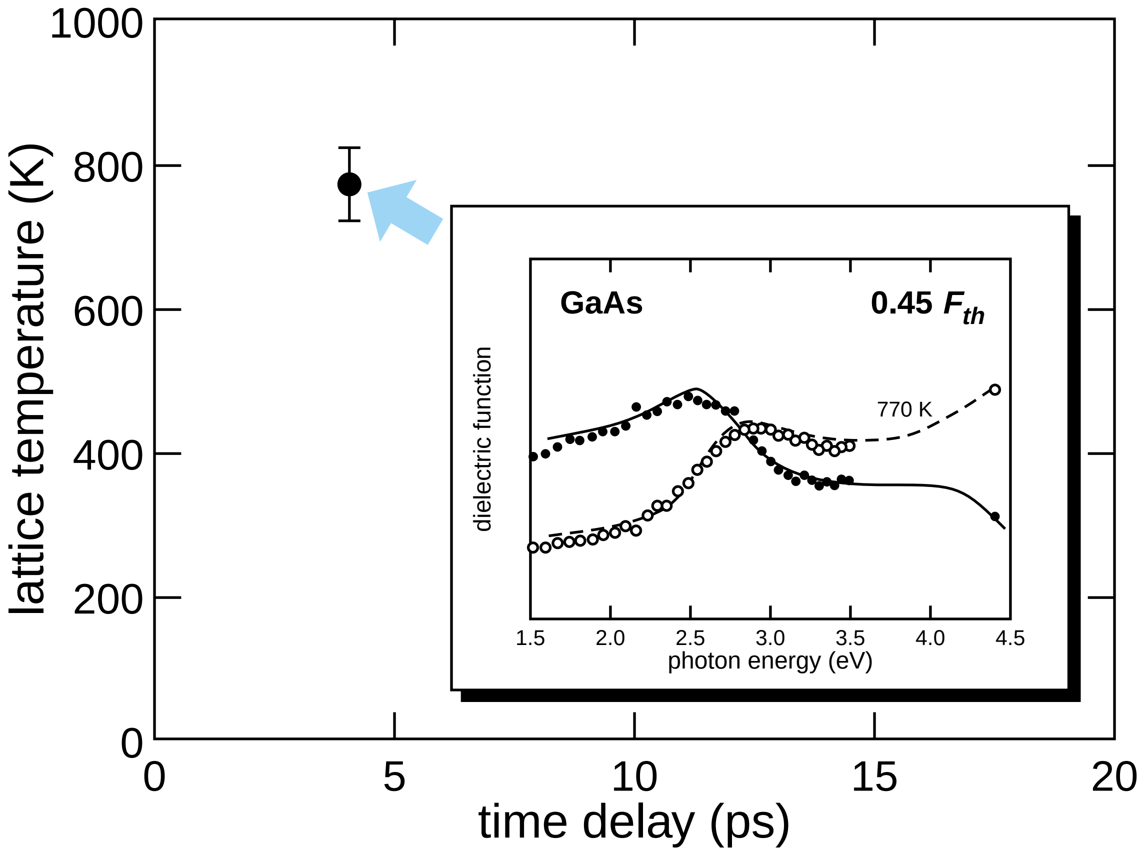
4 ps
GaAs
0.45 F_{th}
770 K
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



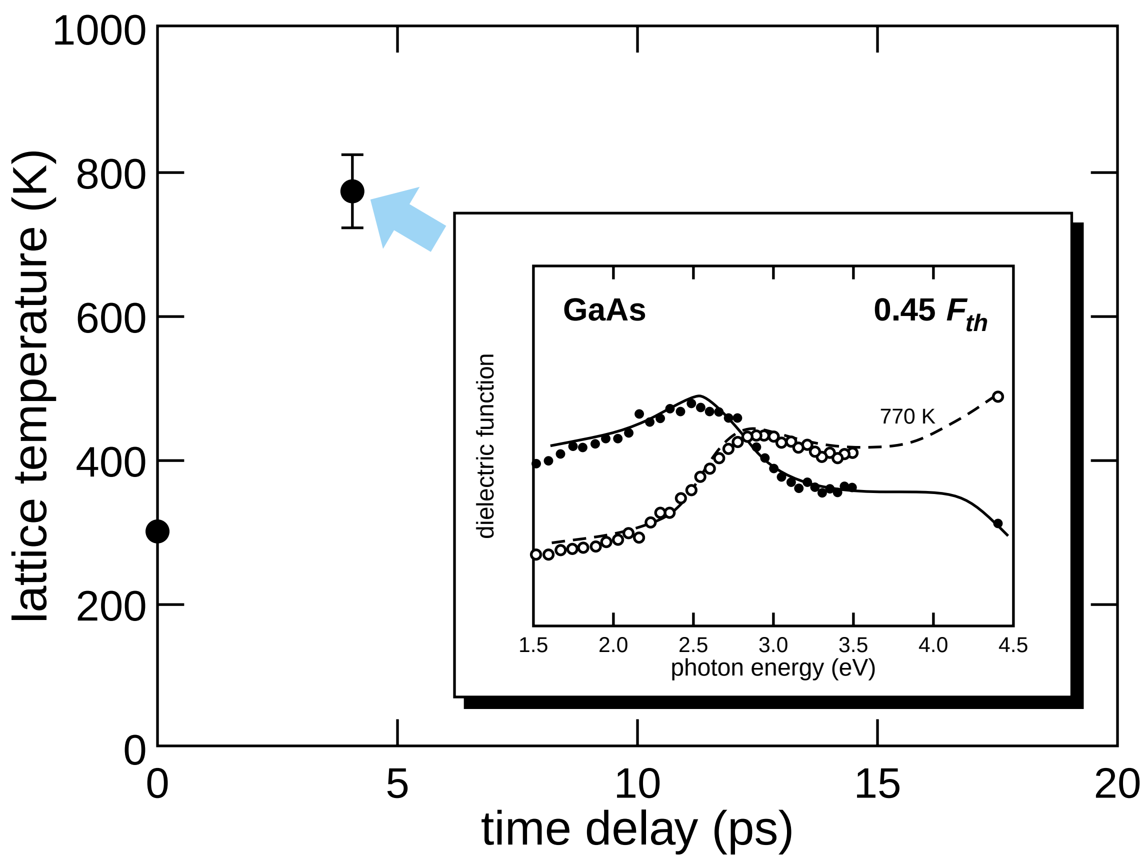
1000
800
600
400
200
0
lattice temperature (K)
0
5
10
15
20
time delay (ps)
GaAs
0.45 F_{th}
770 K
dielectric function
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

Results



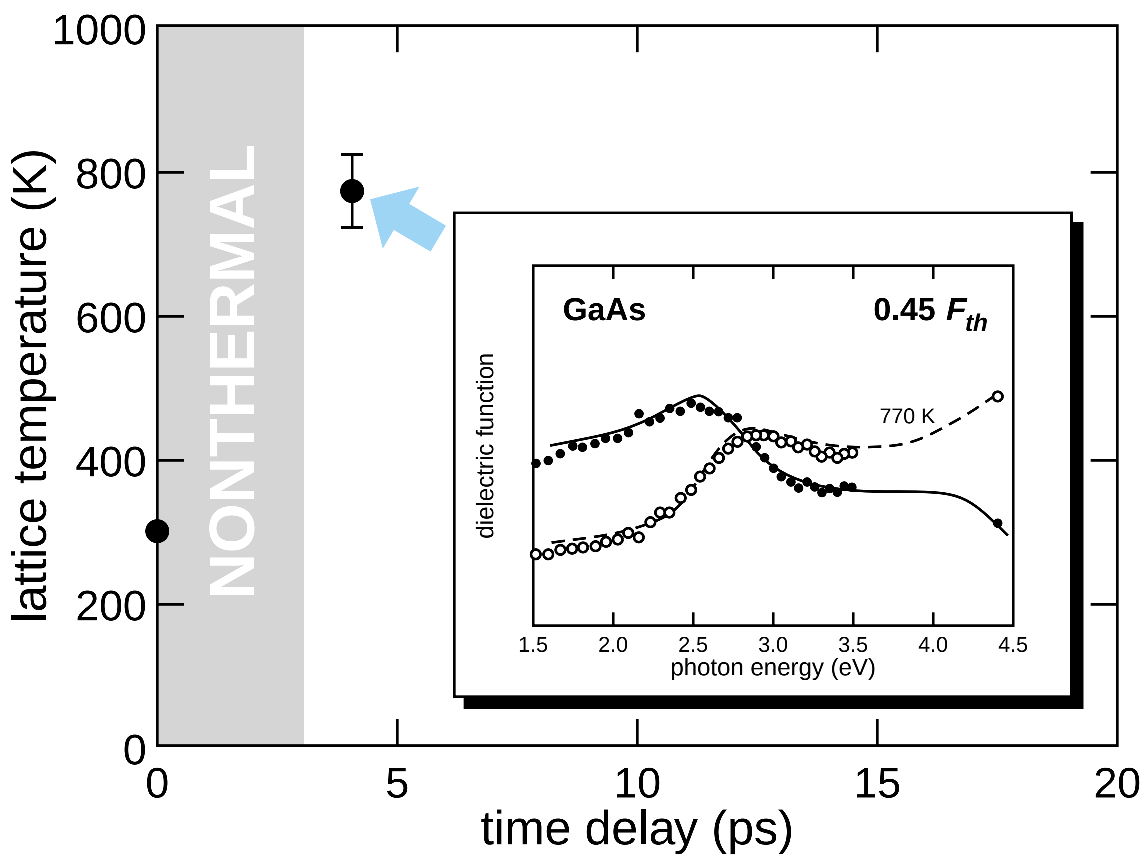
lattice temperature (K)
1000
800
600
400
200
0
0
5
10
15
20
time delay (ps)
GaAs
0.45 F_{th}
770 K
dielectric function
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

Results



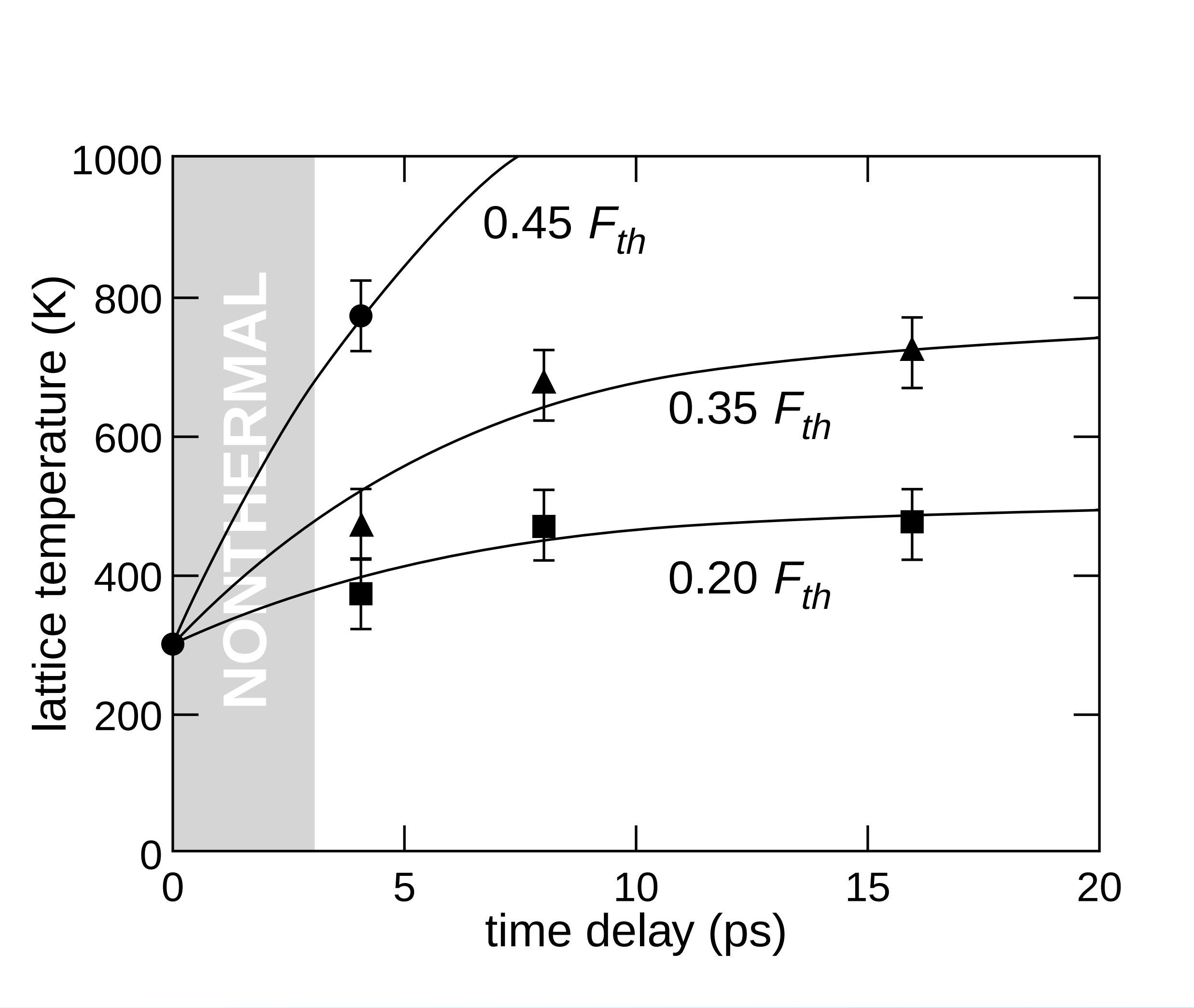
1000
800
600
400
200
0
lattice temperature (K)
0
5
10
15
20
time delay (ps)
GaAs
0.45 F_{th}
770 K
dielectric function
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

Results



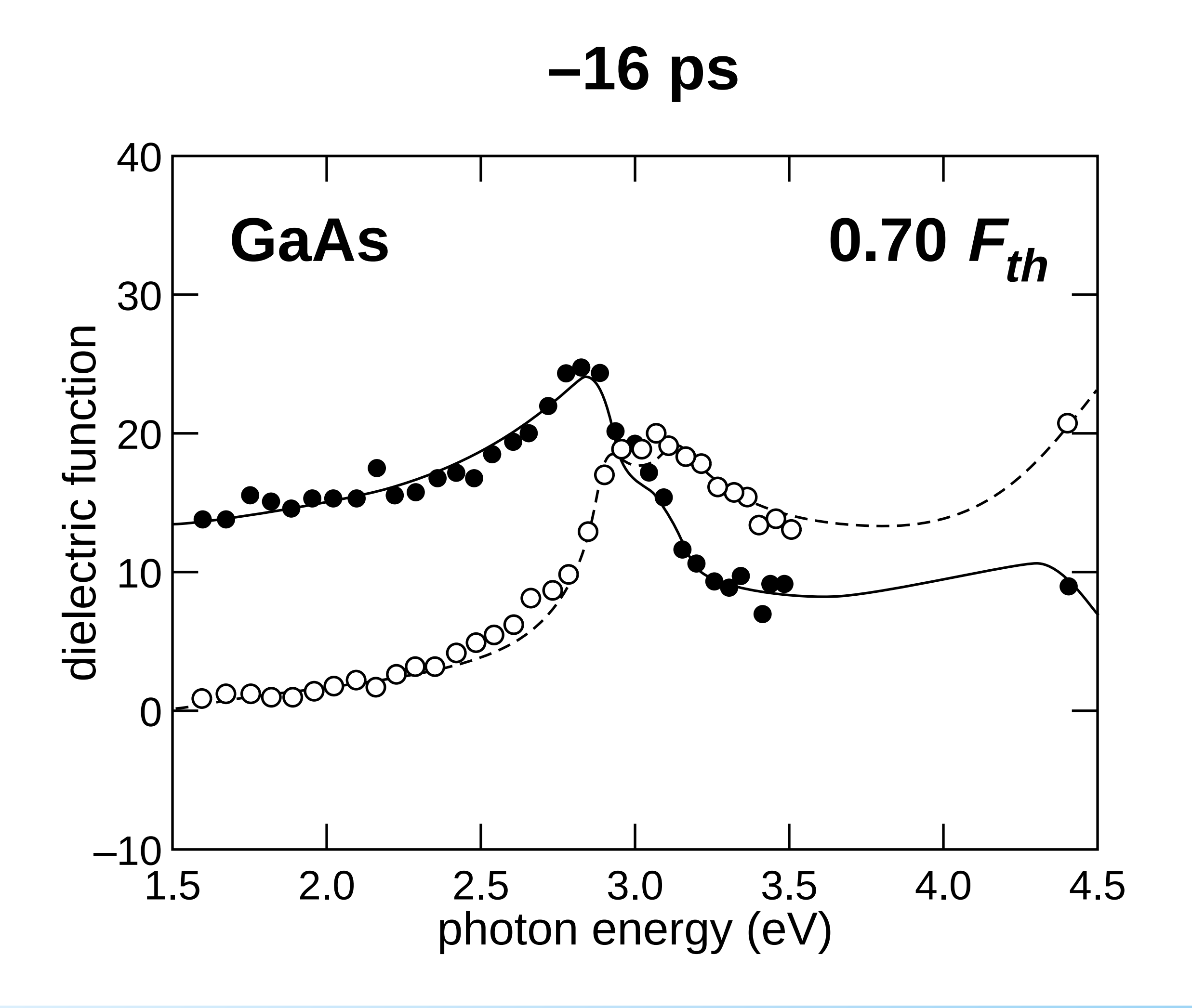
1000
800
600
400
200
0
lattice temperature (K)
NONTHERMAL
0
5
10
15
20
time delay (ps)
GaAs
0.45 F_{th}
770 K
dielectric function
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

Results



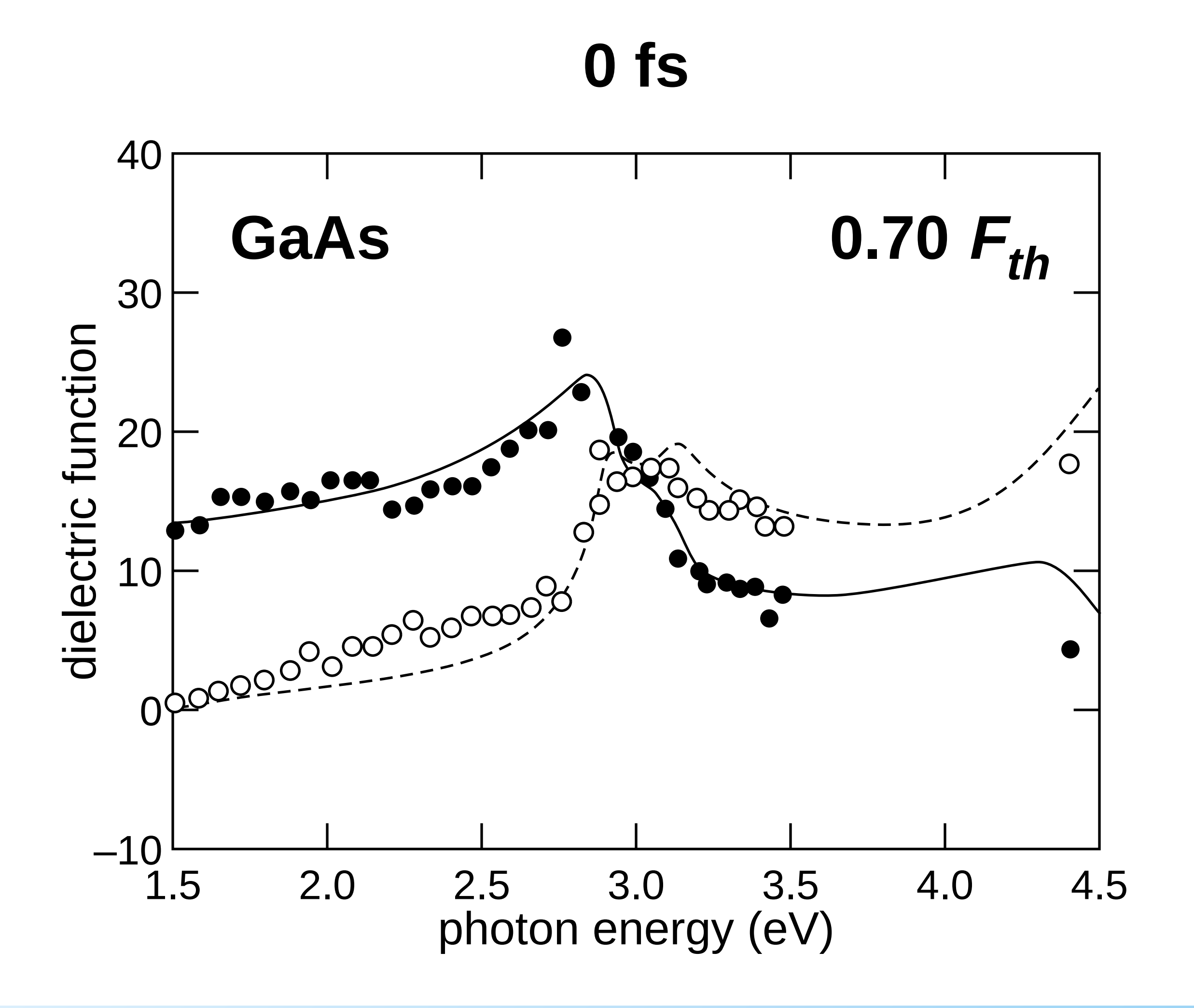
1000
800
600
400
200
0
lattice temperature (K)
NONTHERMAL
0.45 F_{th}
0.35 F_{th}
0.20 F_{th}
0
5
10
15
20
time delay (ps)

Results



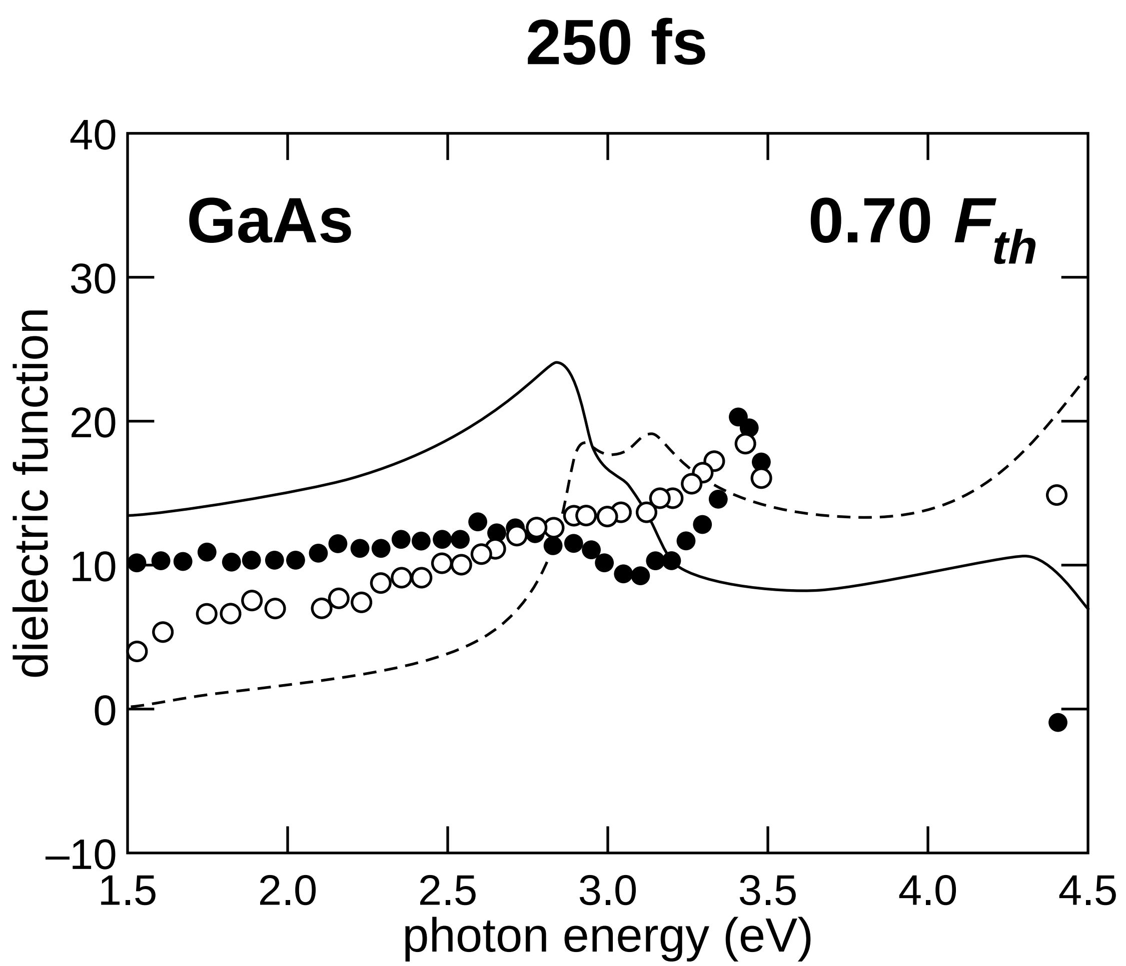
–16 ps
GaAs
0.70 F_{th}
dielectric function
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

Results

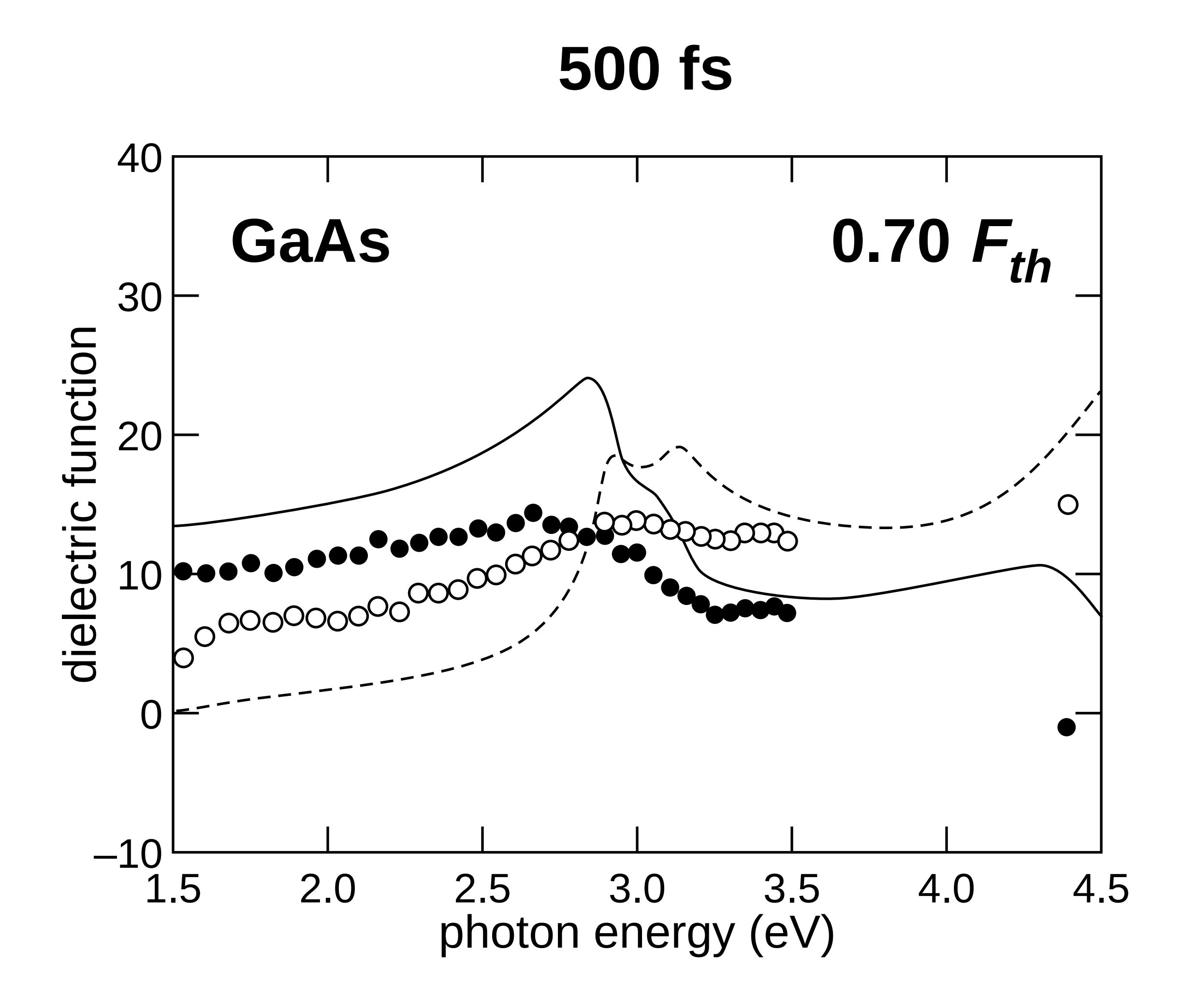


0 fs
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results

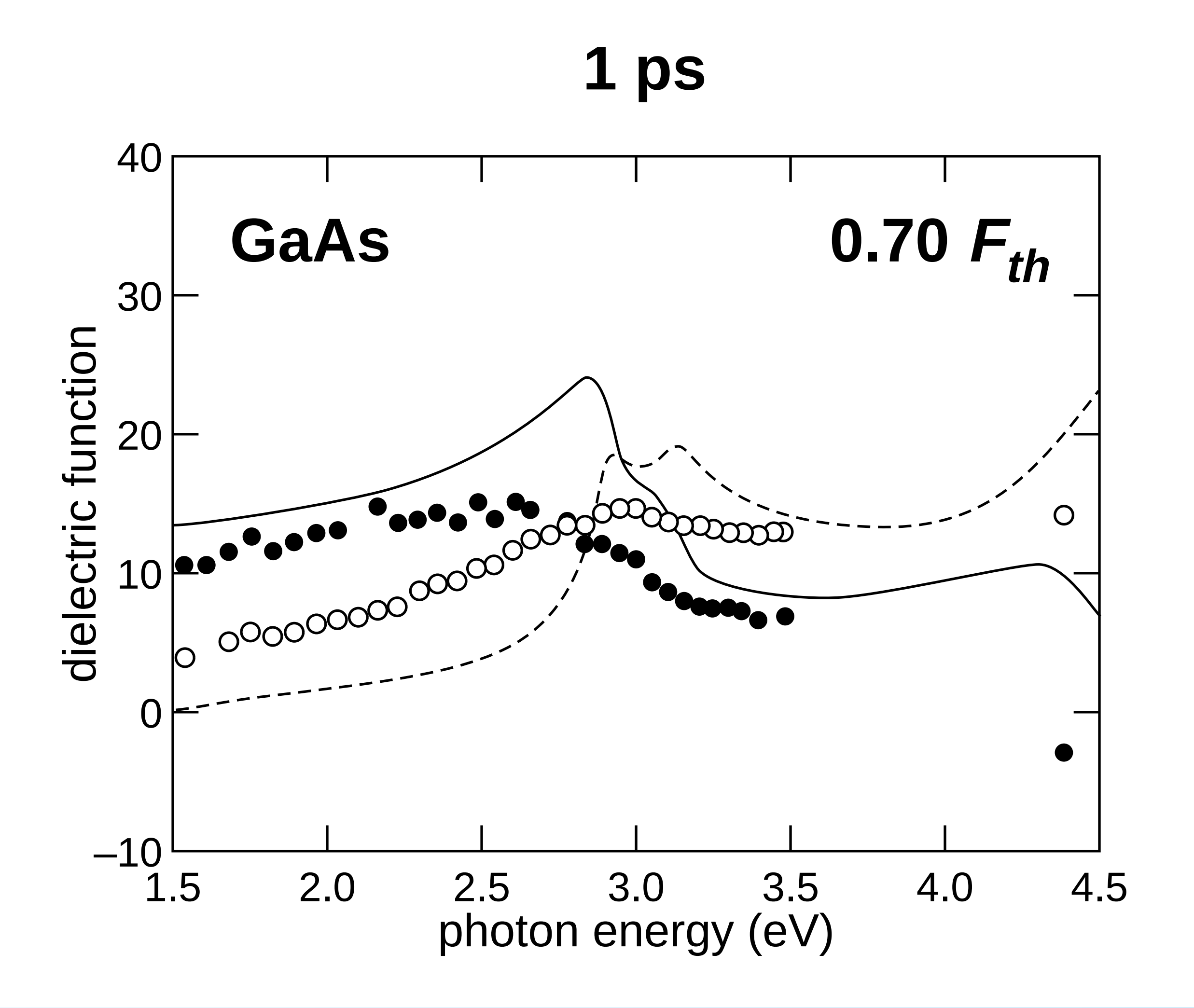

250 fs
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



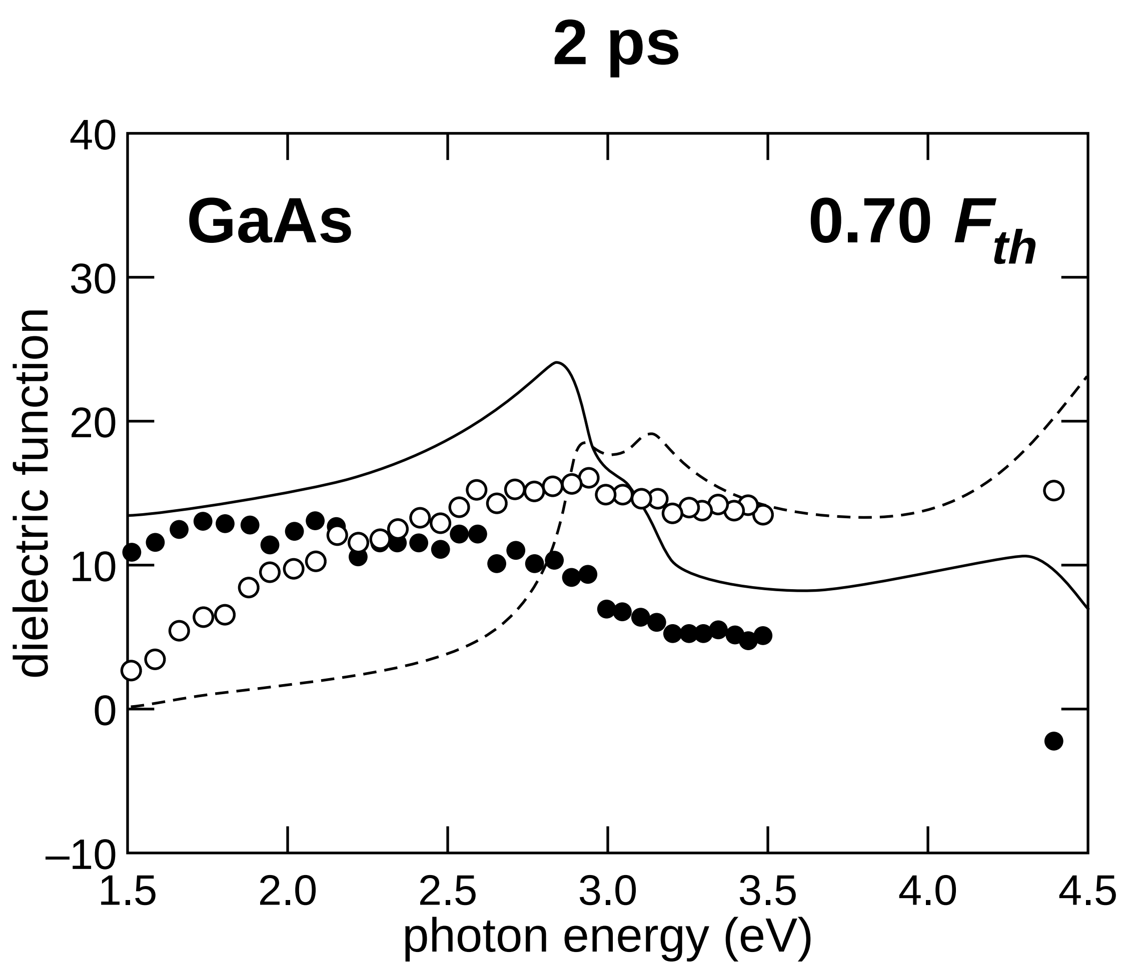
500 fs
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results

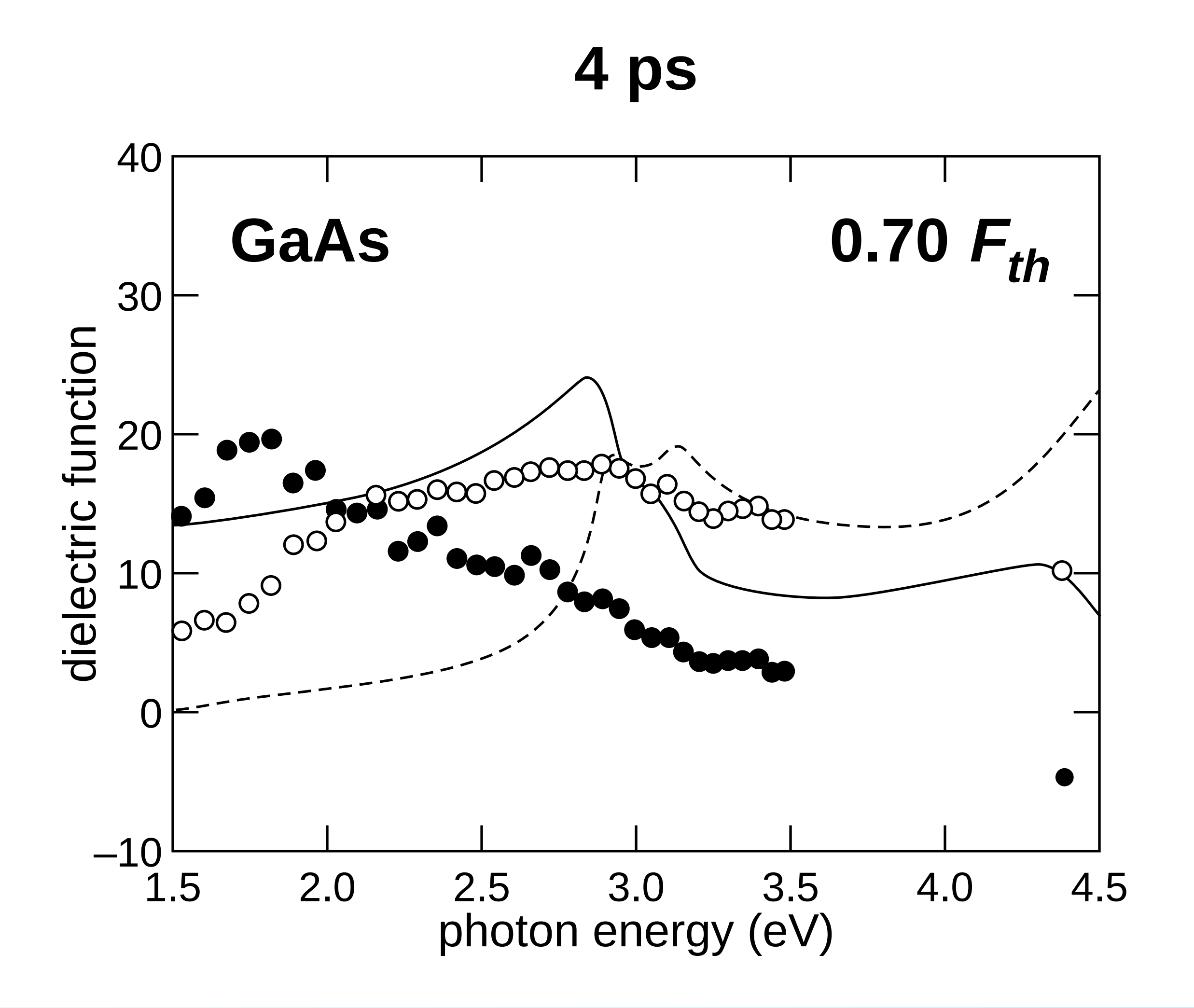


1 ps
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results

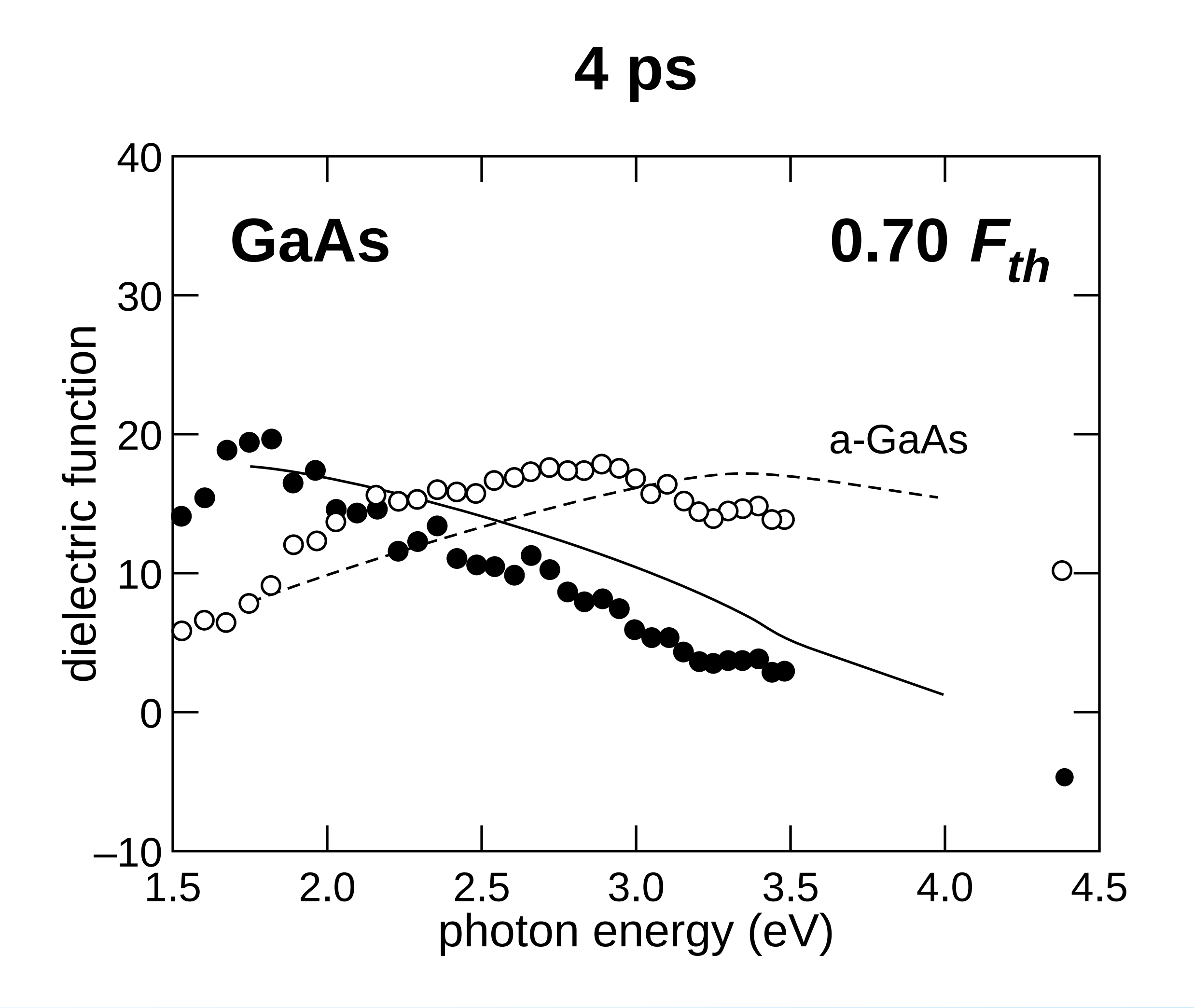

2 ps
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



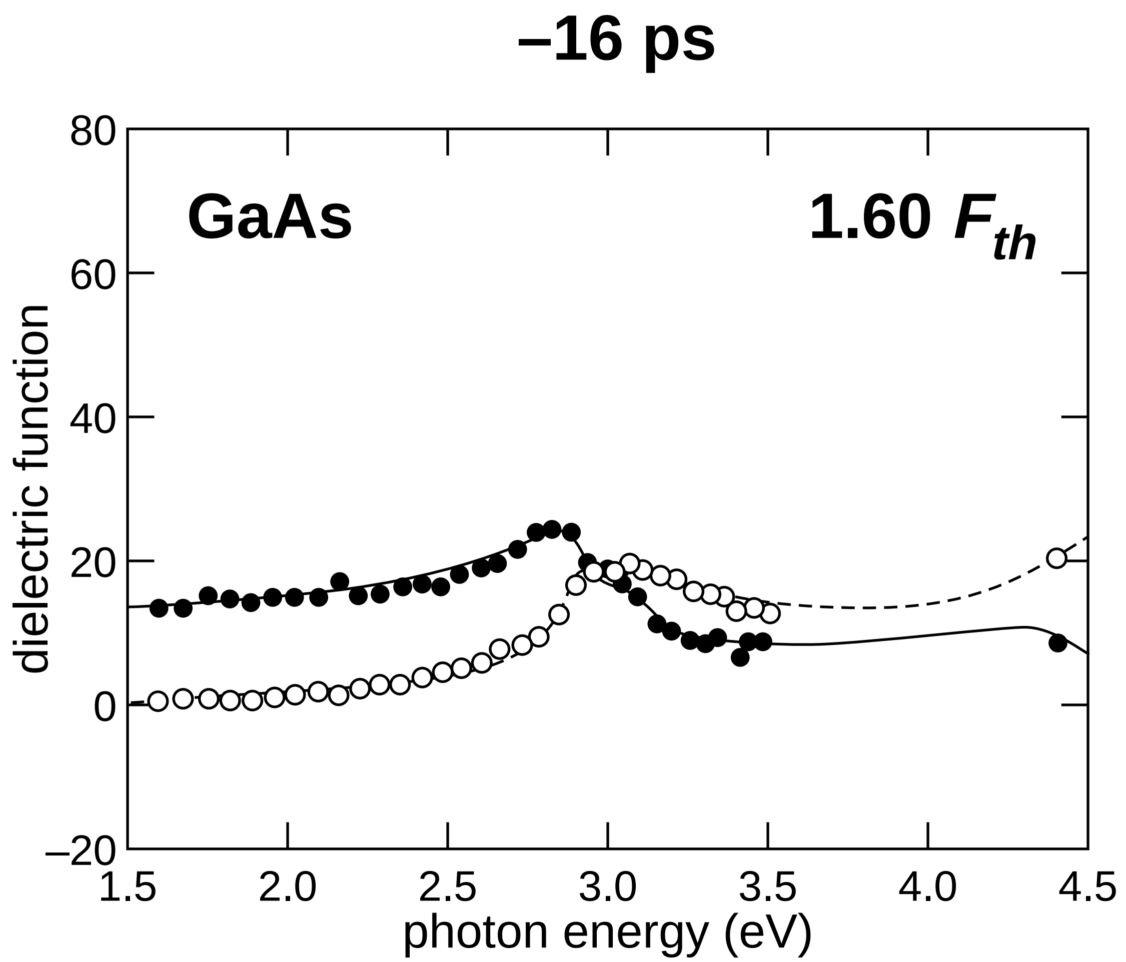
4 ps
GaAs
0.70 F_{th}
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



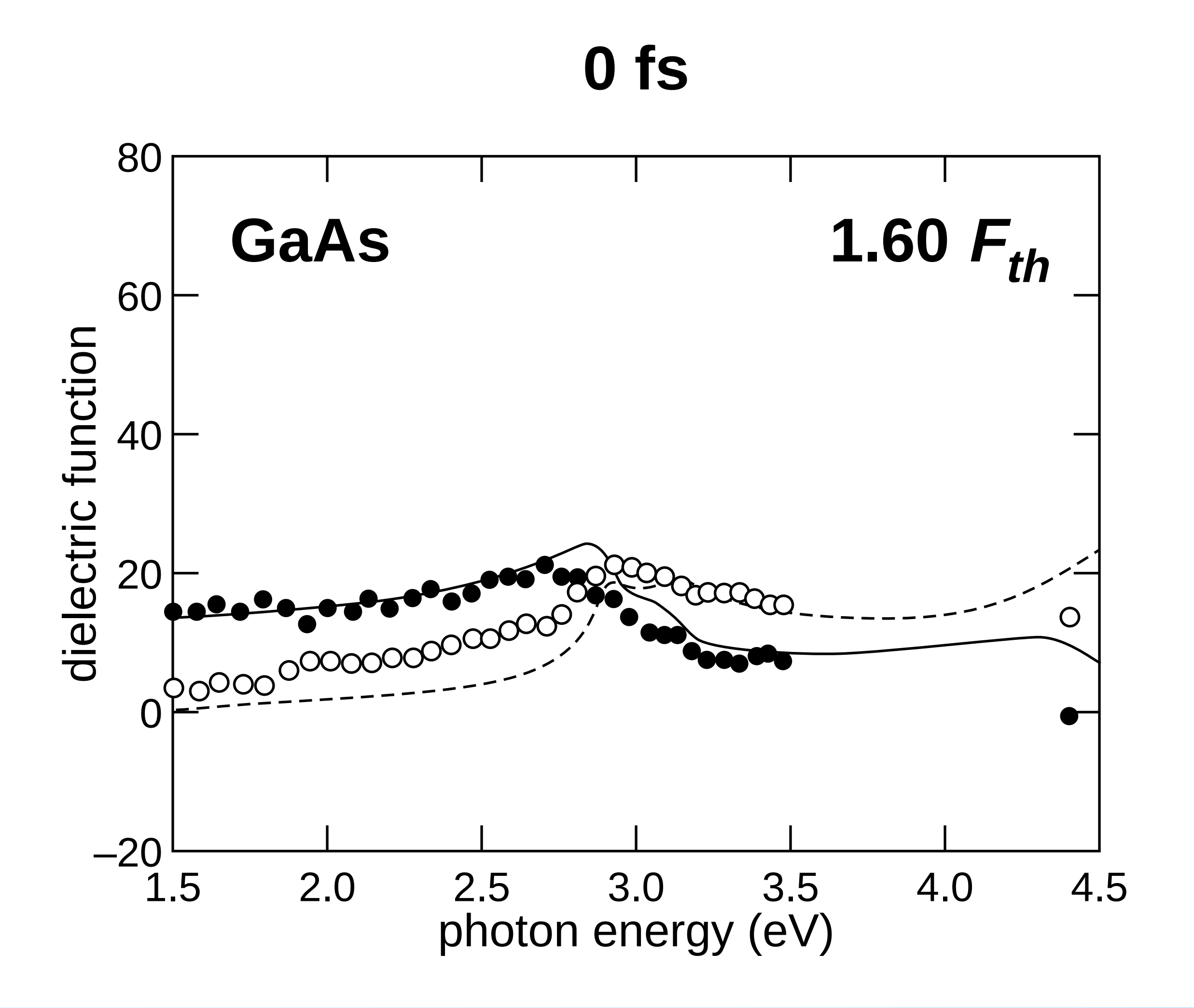
4 ps
GaAs
0.70 F_{th}
a-GaAs
dielectric function
photon energy (eV)
40
30
20
10
0
–10
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



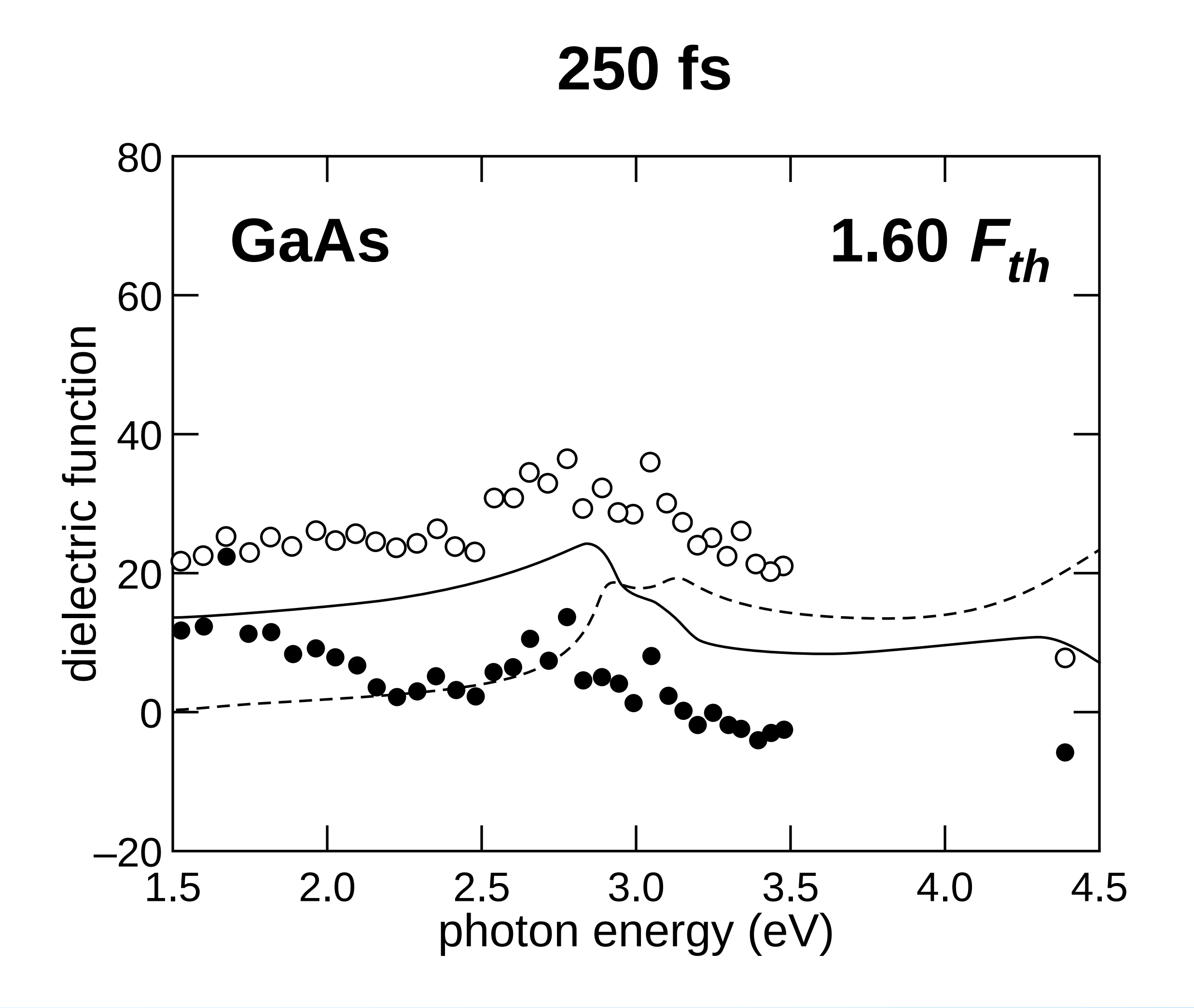
–16 ps
GaAs
1.60 F_{th}
dielectric function
photon energy (eV)
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



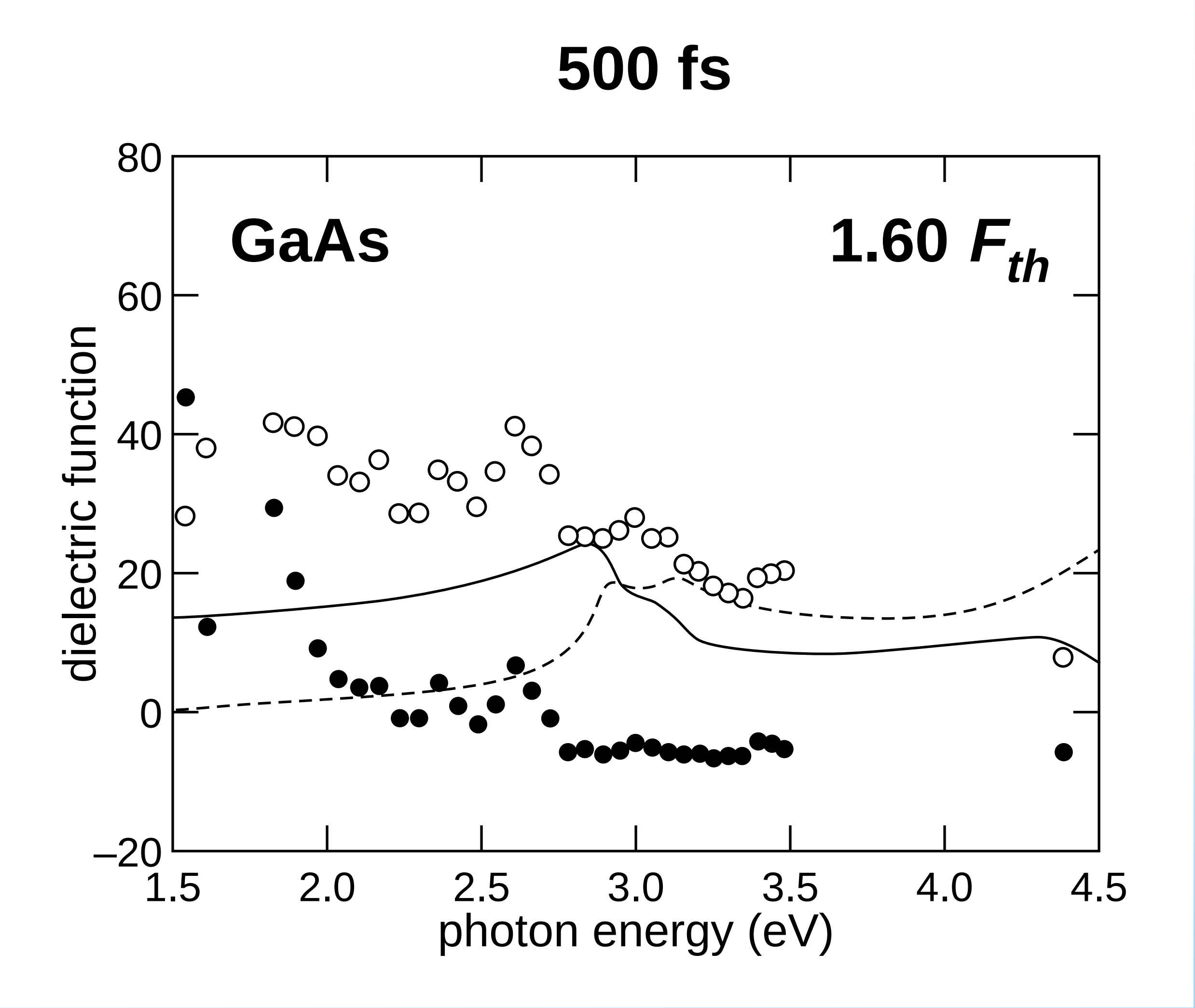
0 fs
GaAs
1.60 F_{th}
80
60
40
20
0
–20
dielectric function
1.5
2.0
2.5
3.0
3.5
4.0
4.5
photon energy (eV)

Results



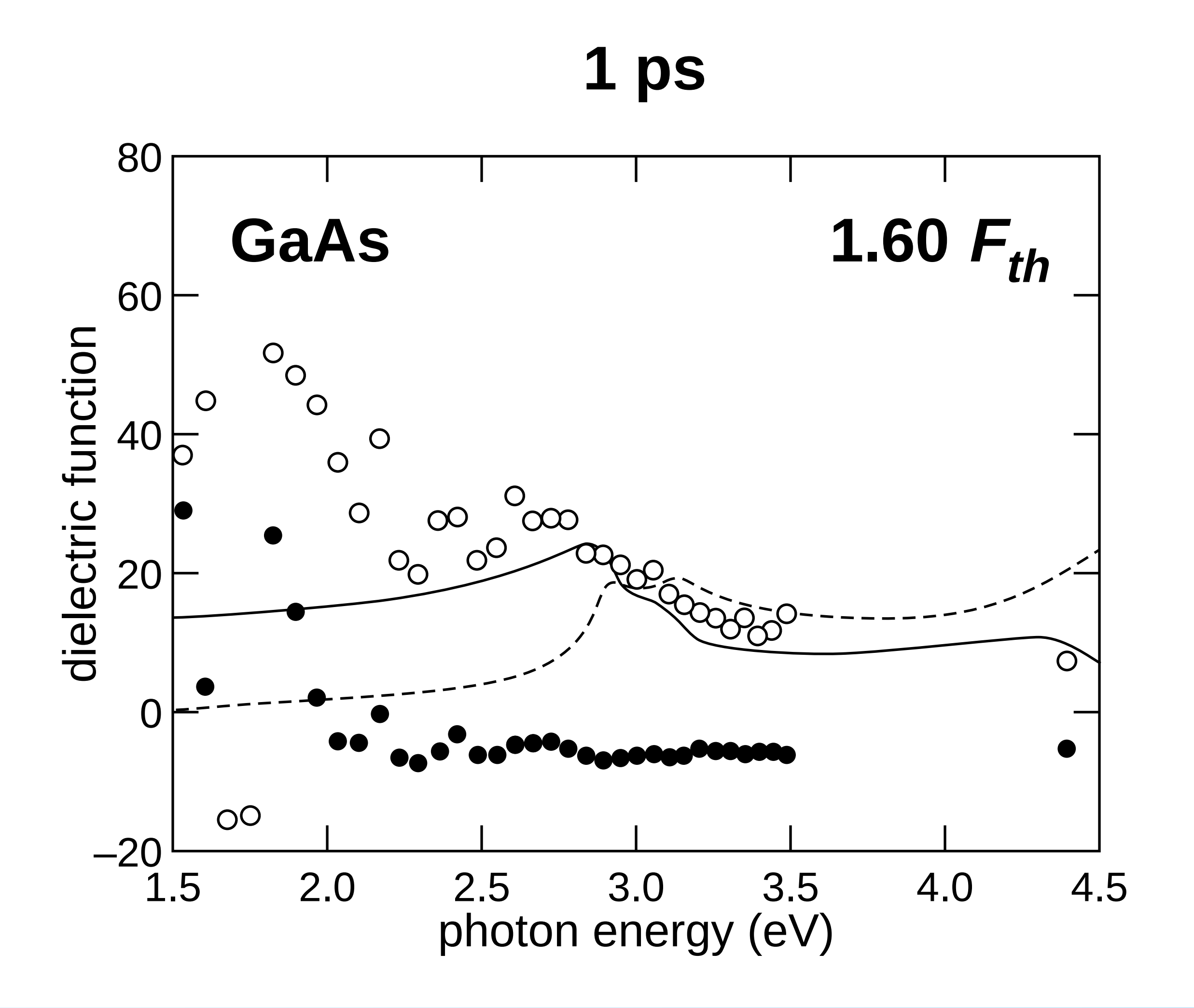
250 fs
GaAs
1.60 F_{th}
dielectric function
photon energy (eV)
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



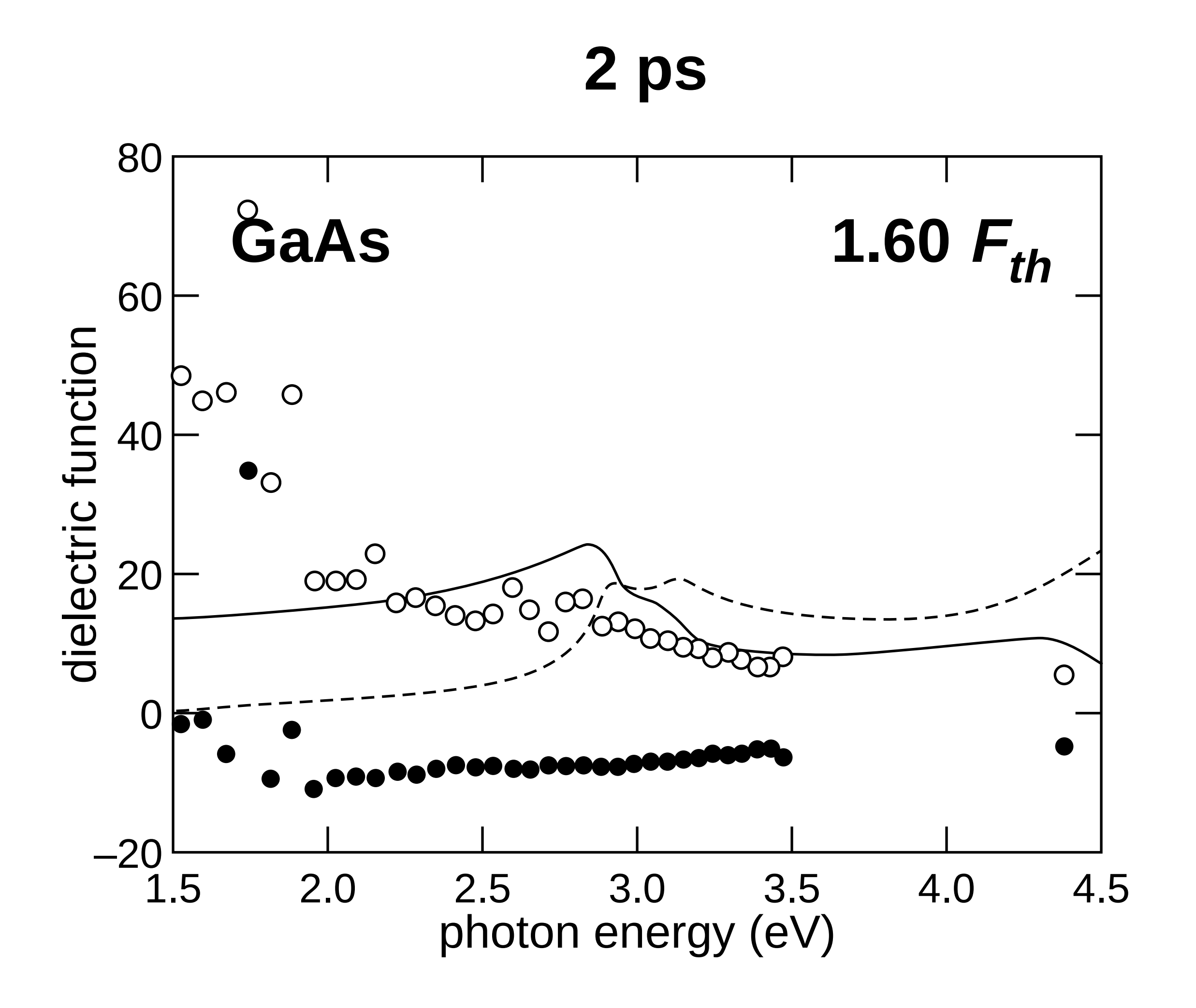
500 fs
GaAs
1.60 F_{th}
dielectric function
photon energy (eV)
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



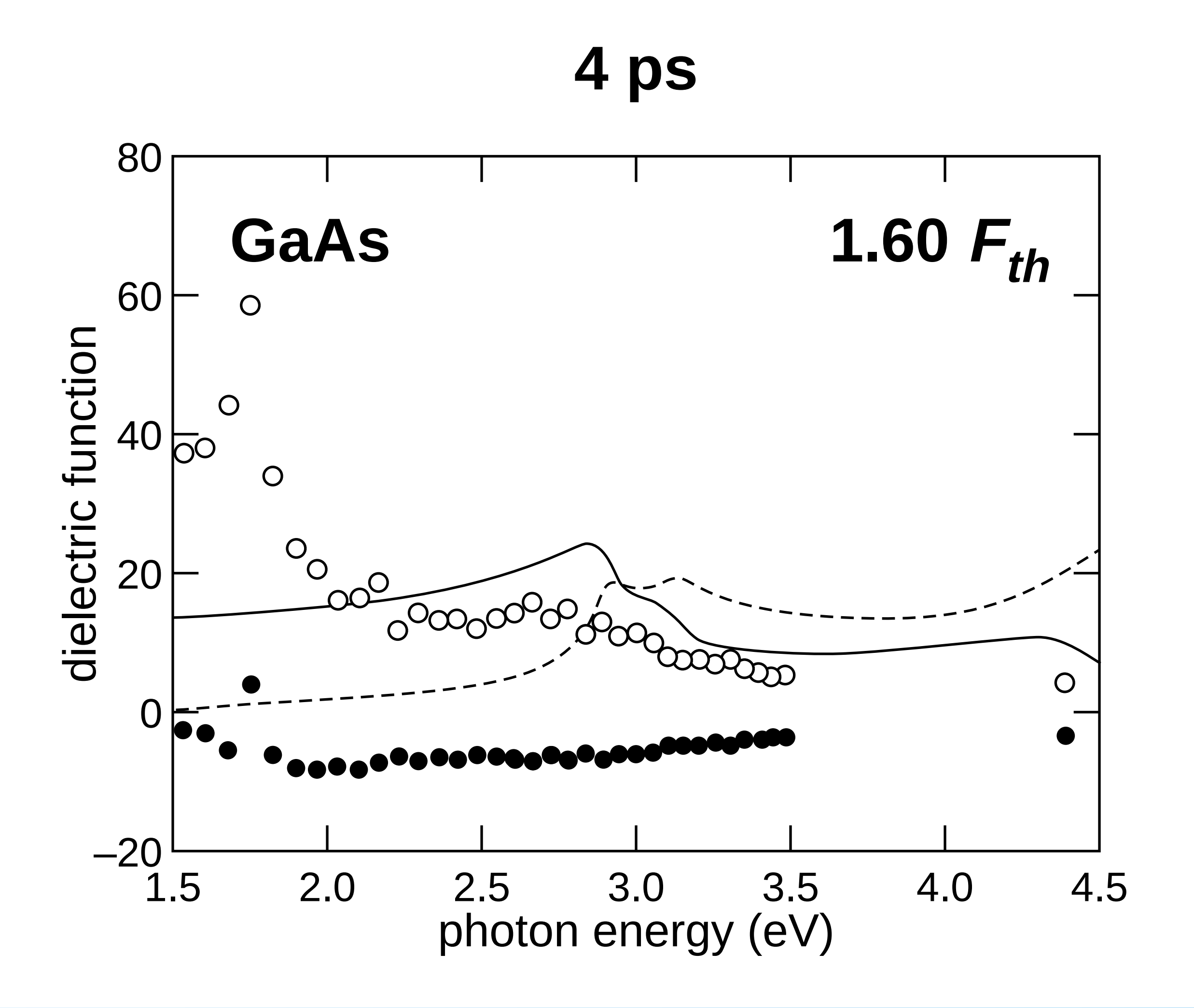
1 ps
GaAs
1.60 F_th
dielectric function
photon energy (eV)
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



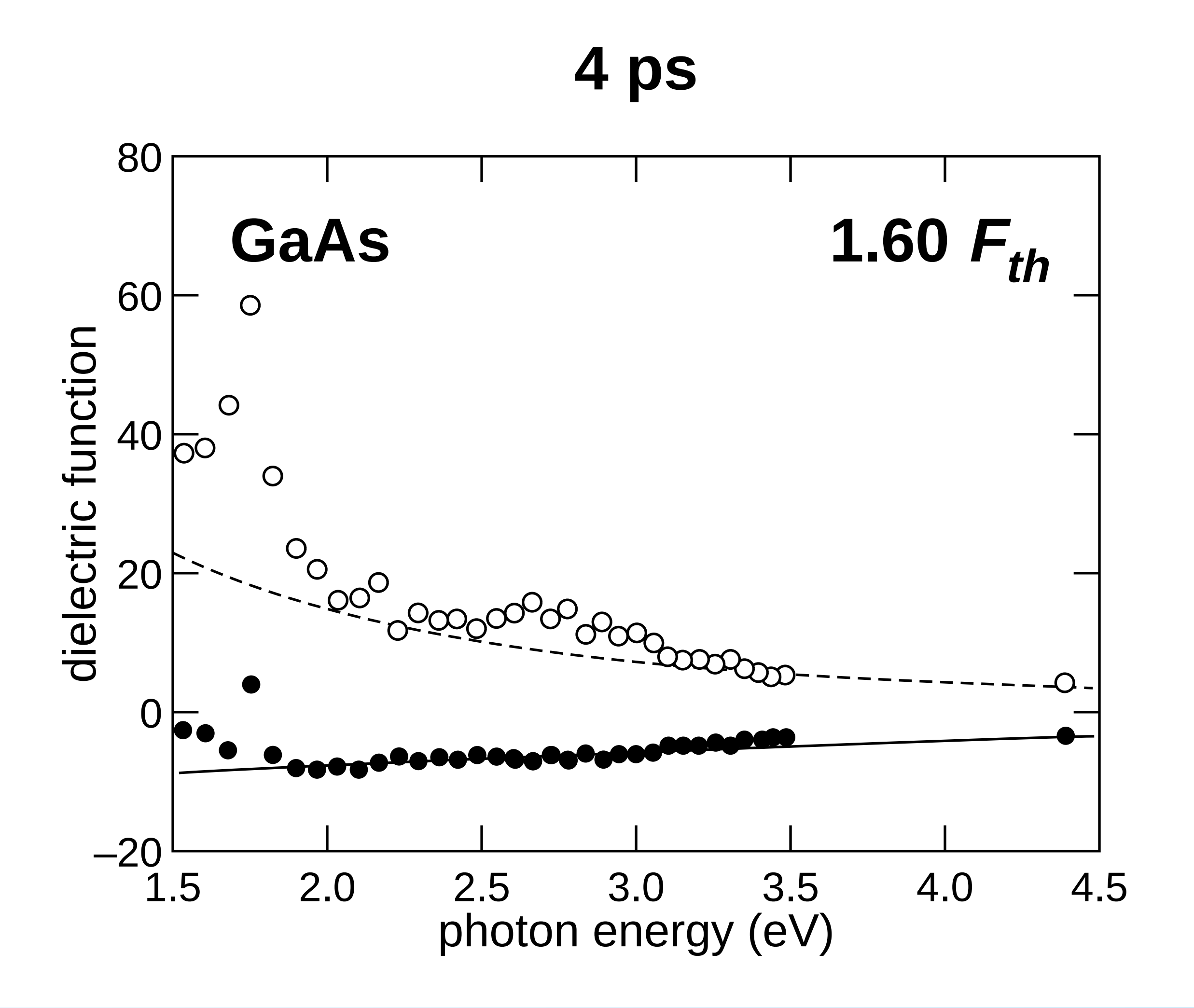
2 ps
GaAs
1.60 F th
dielectric function
photon energy (eV)
80
60
40
20
0
–20
1.5
2.0
2.5
3.0
3.5
4.0
4.5

Results



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

Results



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

Summary

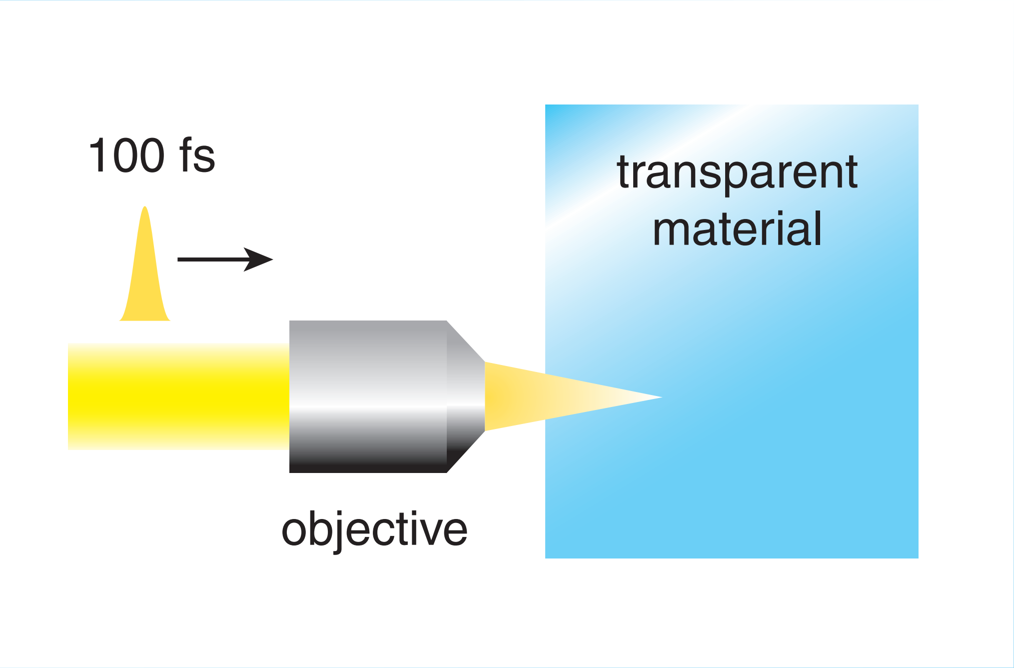
- **measurement of $\varepsilon(\omega)$ identifies ultrafast phase changes**
- **initial response is electronic, via band structure and electron occupation changes**
- **structural effects dominate after a few ps**
- **interesting reversible regime**

Conclusions

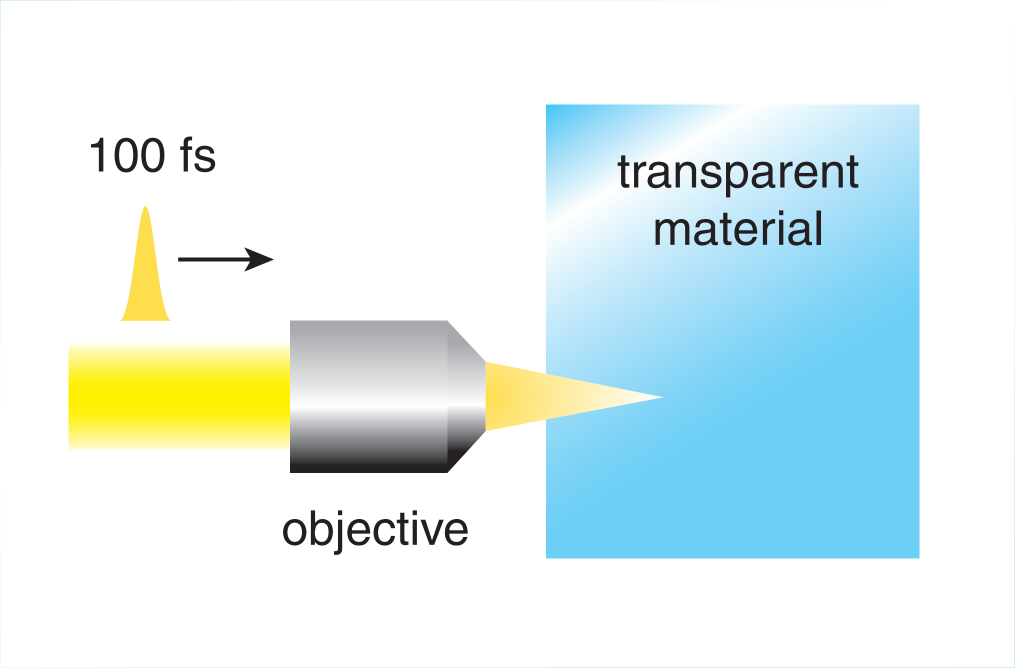
- **strong electronic excitation can drive a structural transition**

- **femtosecond lasers allow us to see the dynamics of the transition**

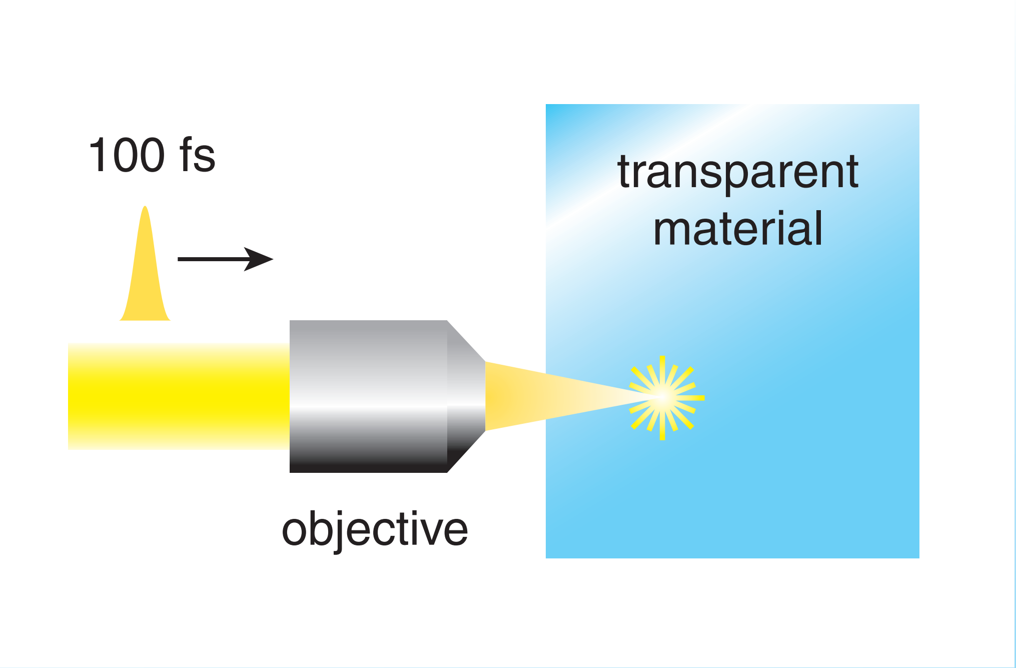
focus laser beam inside material…



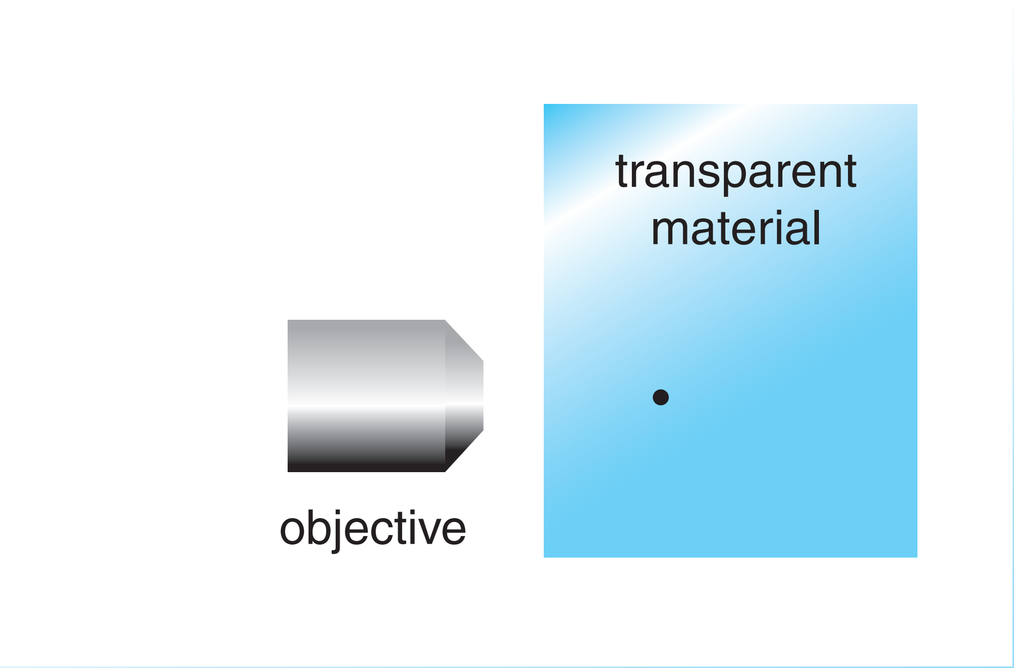
high intensity at focus...



… causes nonlinear ionization…



and microscopic bulk damage



Introduction

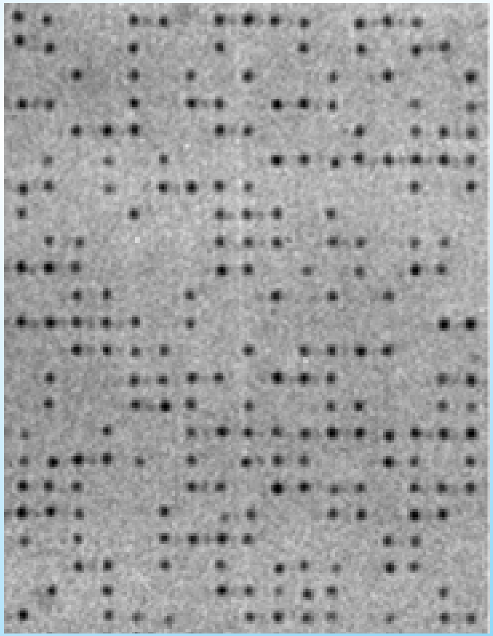
Applications:

- **data storage**

Introduction

Applications:

- data storage



Introduction

Applications:

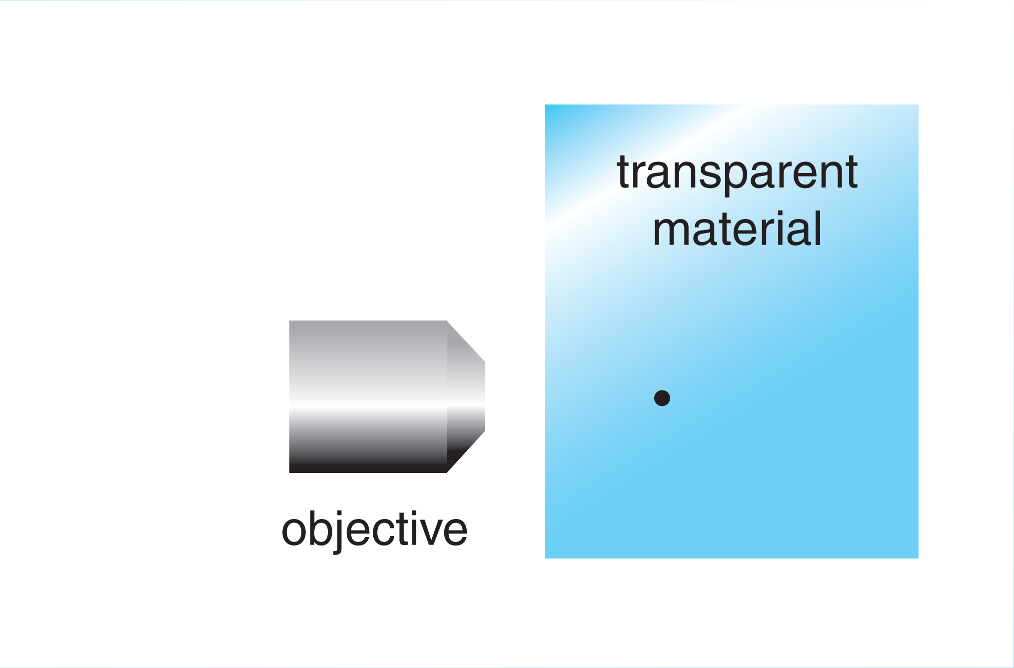
- **data storage**
- **photonic devices**

Introduction

Applications:

- **data storage**
- **photonic devices**
- **internal micromachining**

What are the conditions at focus?



laser deposits energy in ~1 μm^3

Introduction

What temperature?

Introduction

What temperature?

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

What temperature?

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

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

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

Introduction

What temperature?

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

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

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

So, 1 μJ in 1 μm^3 gives

~1,000,000 K!

Introduction

What pressure?

Introduction

What pressure?

Treat ionized material as an ideal gas:

$$pV = nRT$$

Introduction

What pressure?

Treat ionized material as an ideal gas:

$$pV = nRT$$

Gives

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

So:

	microexplosion
T	$\approx$1 MK
p	$\approx$10 MBar
ρ	2.2×10^3 kg/m^3

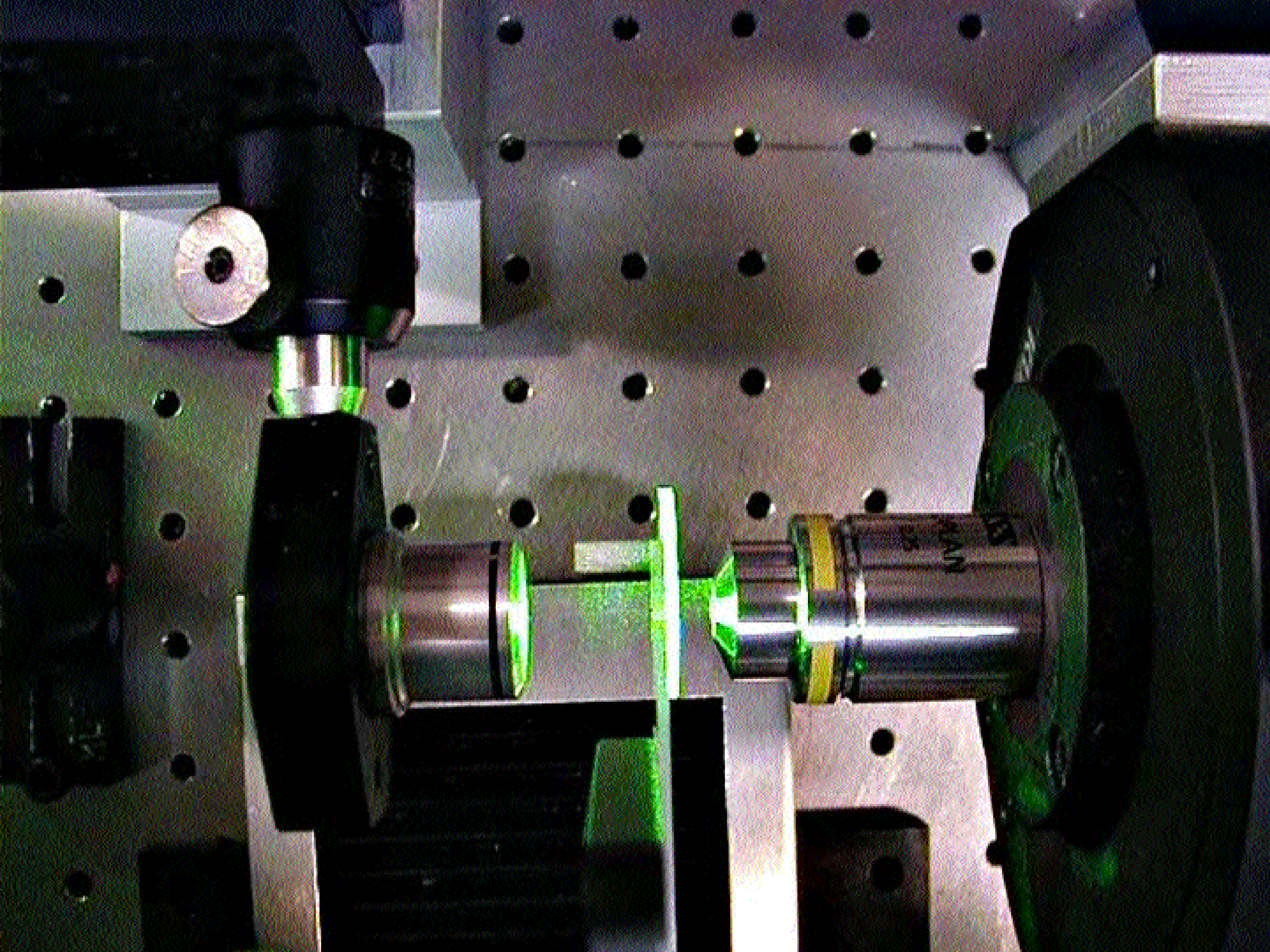
So:

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

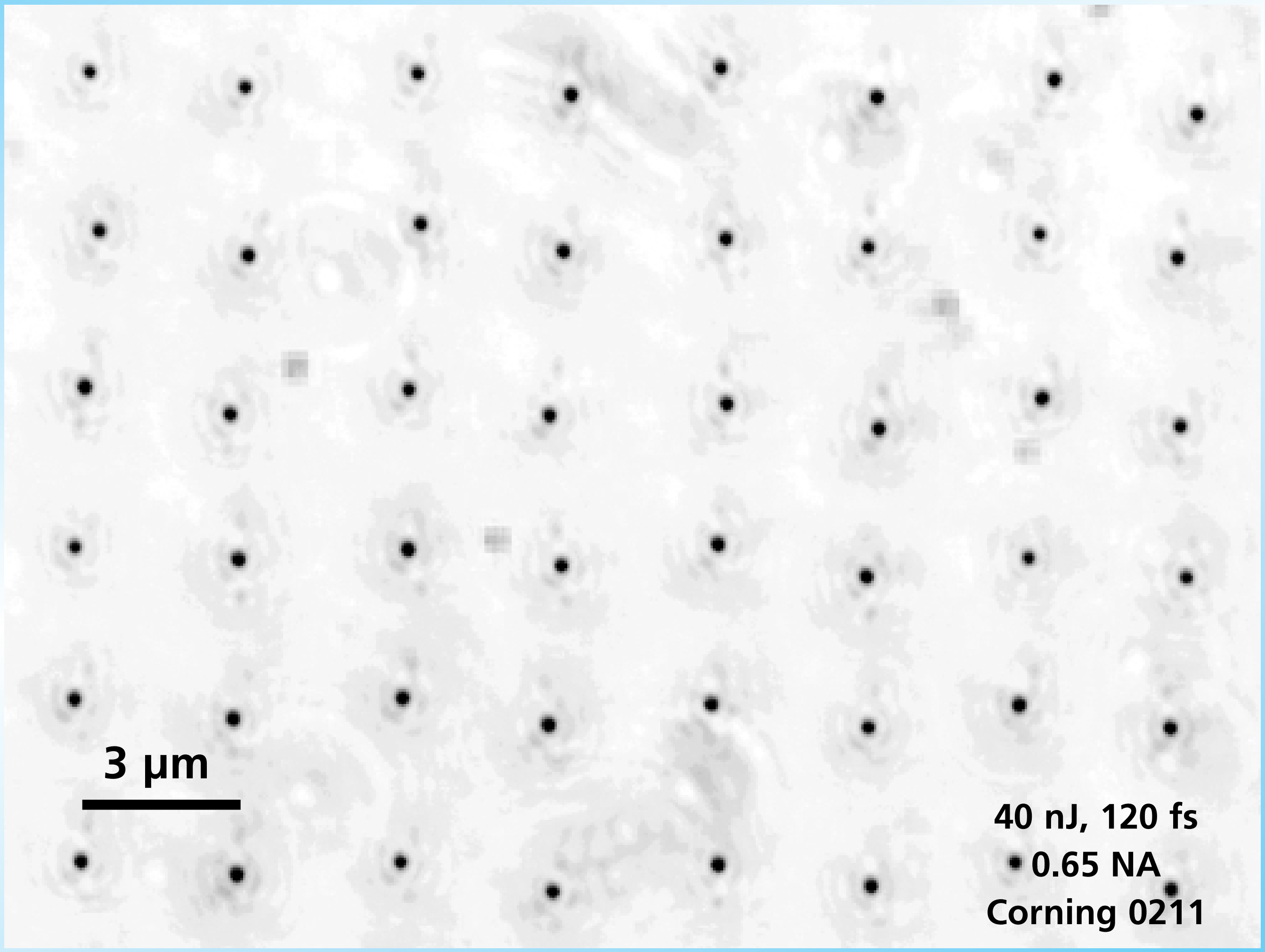
So:

	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

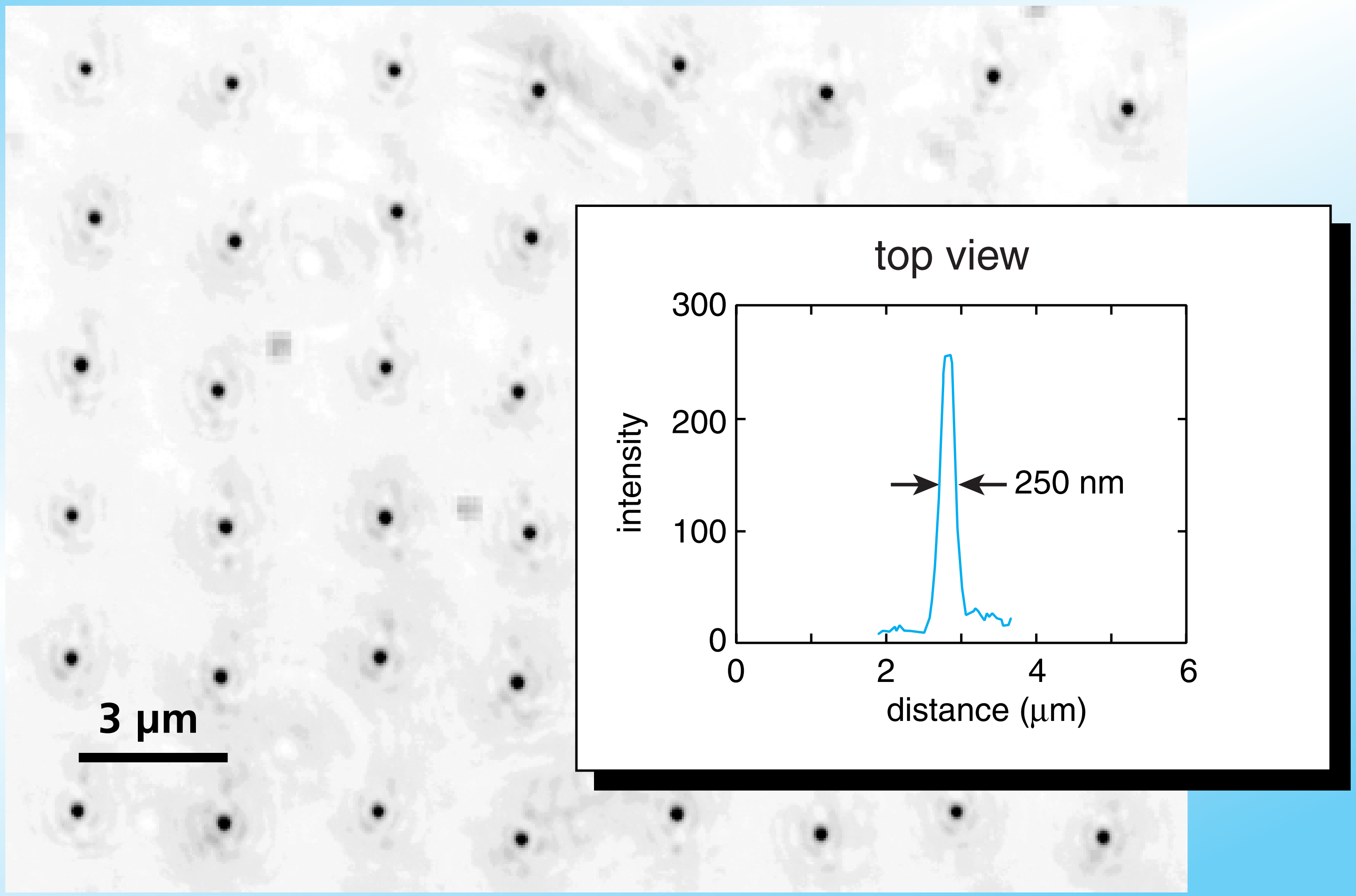
creating stellar conditions in lab!



Damage morphology

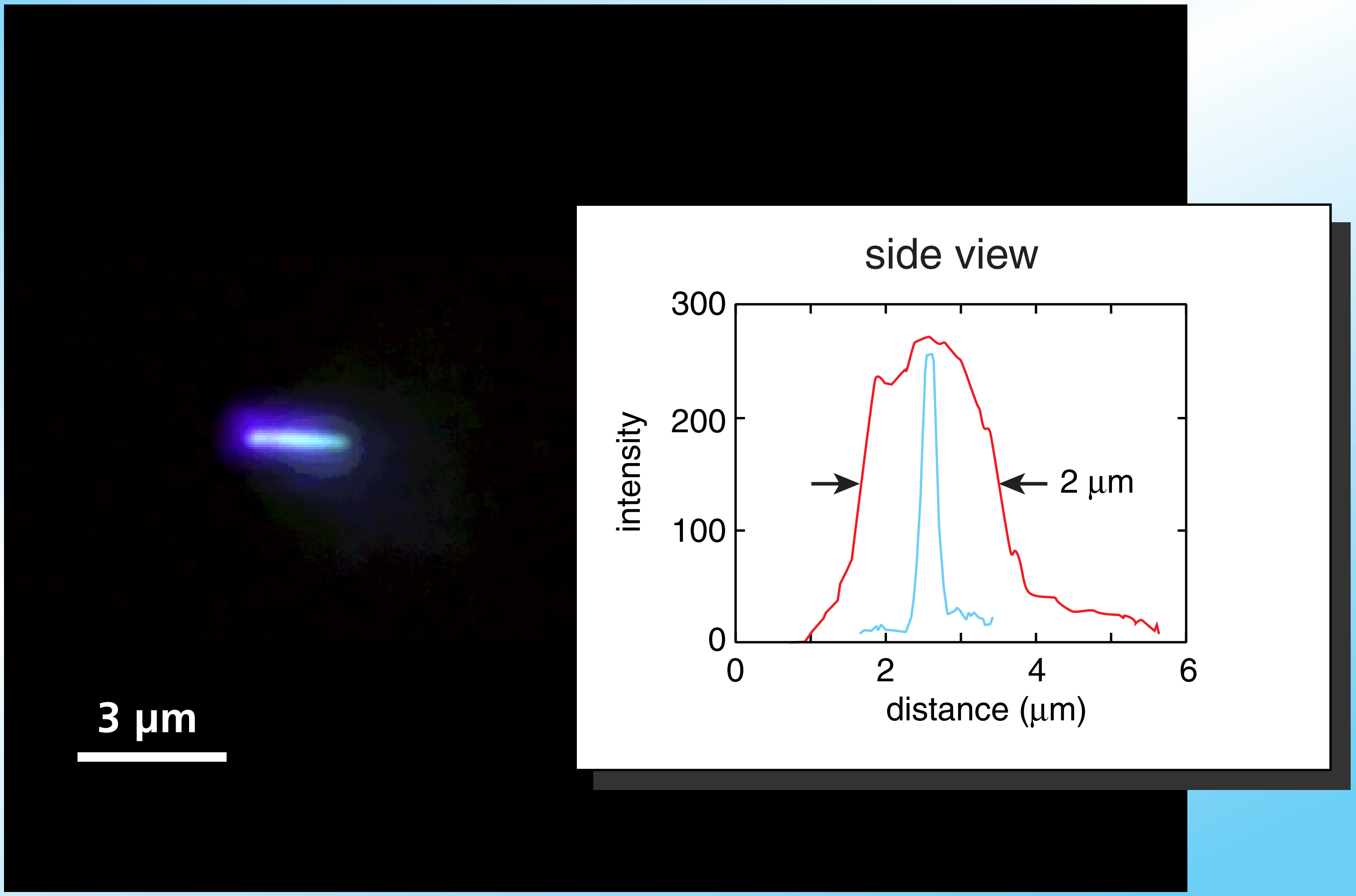


Damage morphology

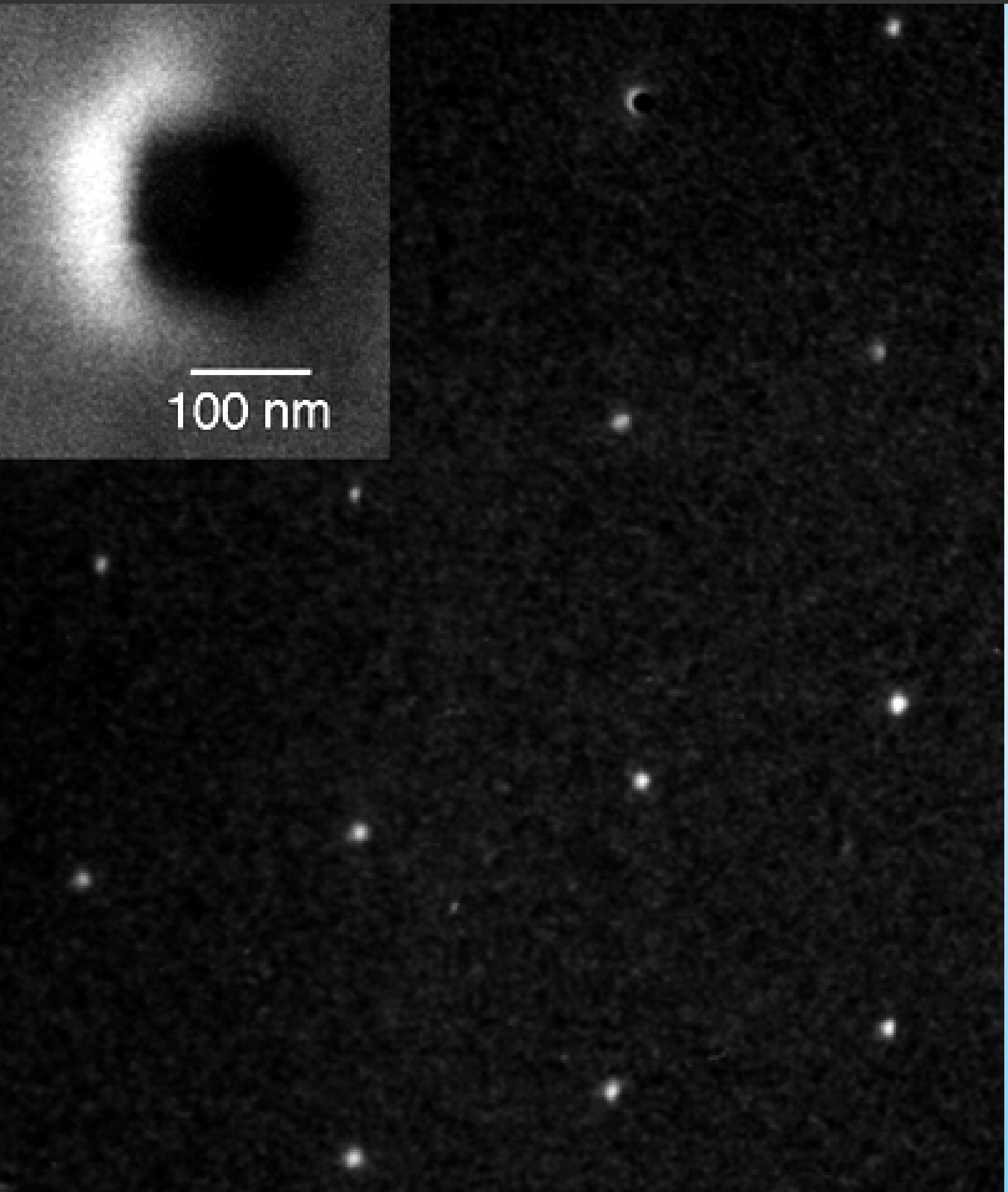


Damage morphology

Damage morphology



Damage morphology

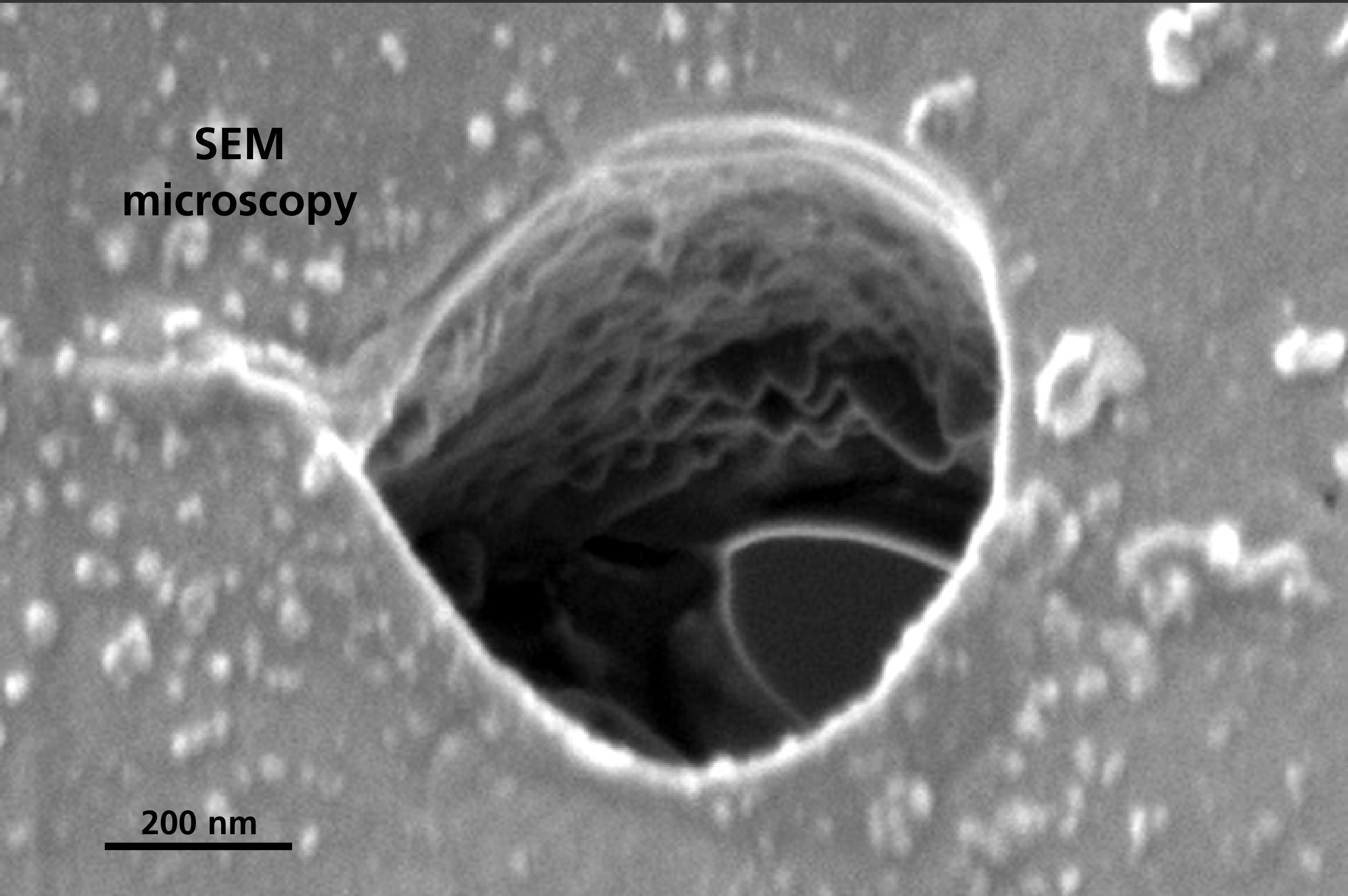


Electron Microscopy:

explosive damage forms voids

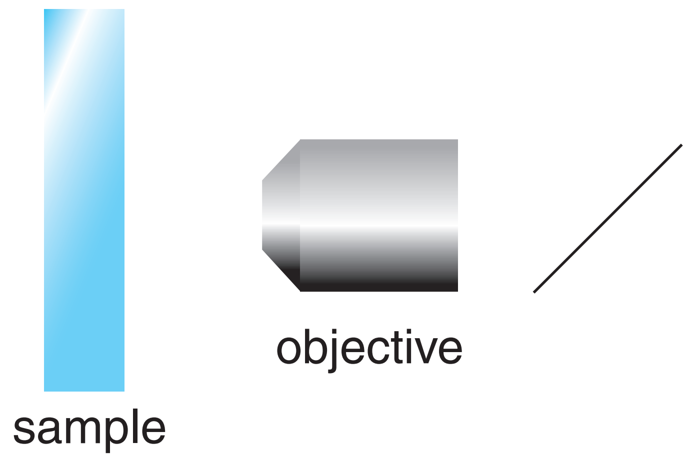
100 fs, 500 nJ
0.65 NA
fused silica

Damage morphology

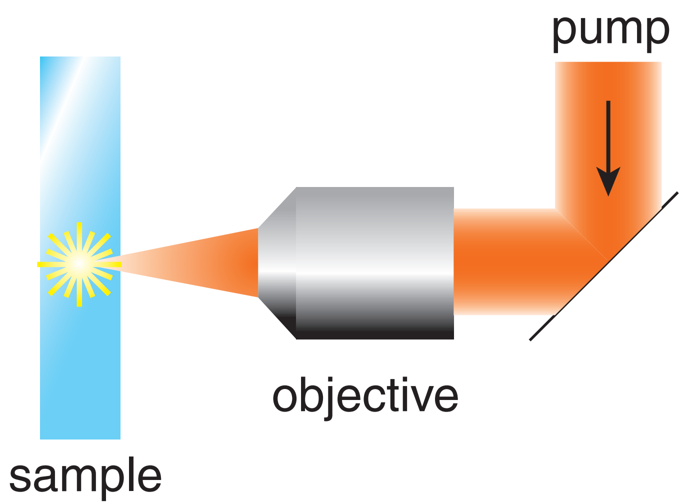


Dynamics

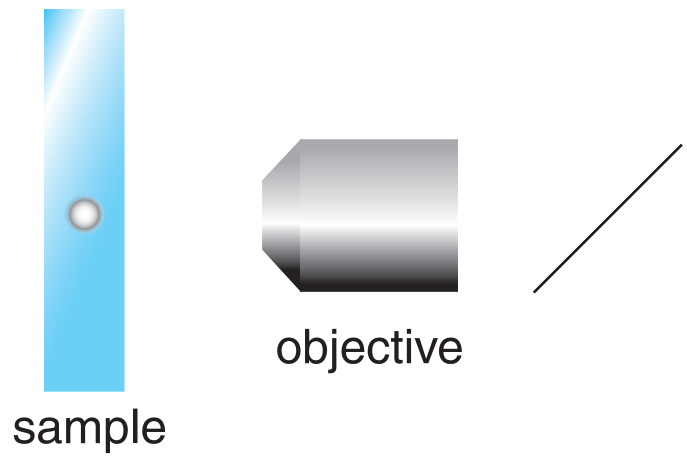
imaging setup



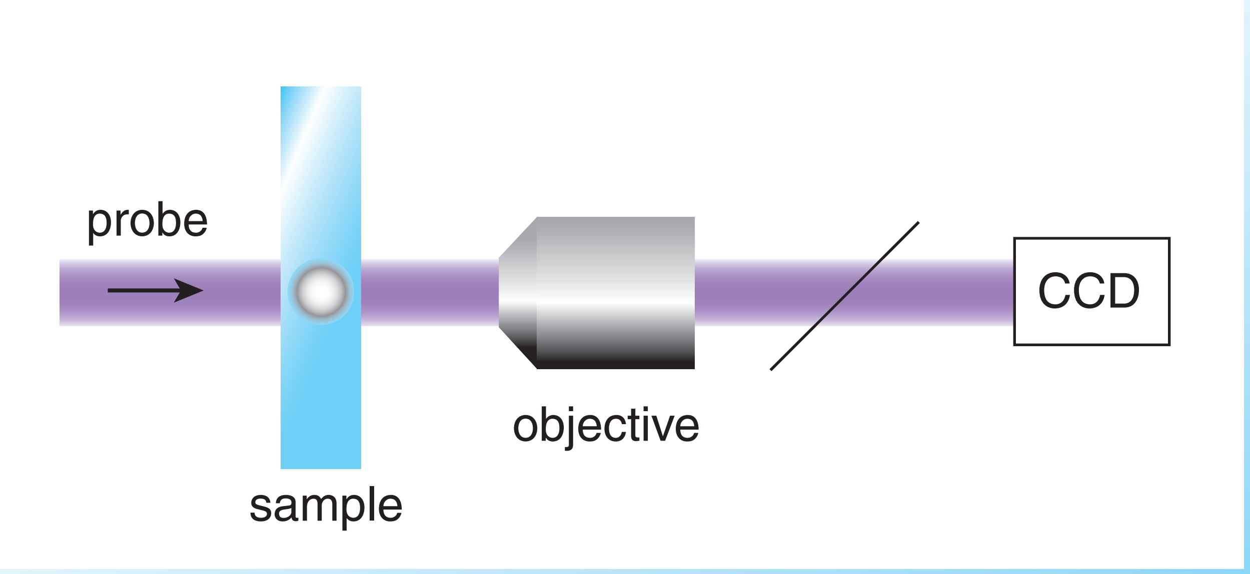
imaging setup



imaging setup



imaging setup



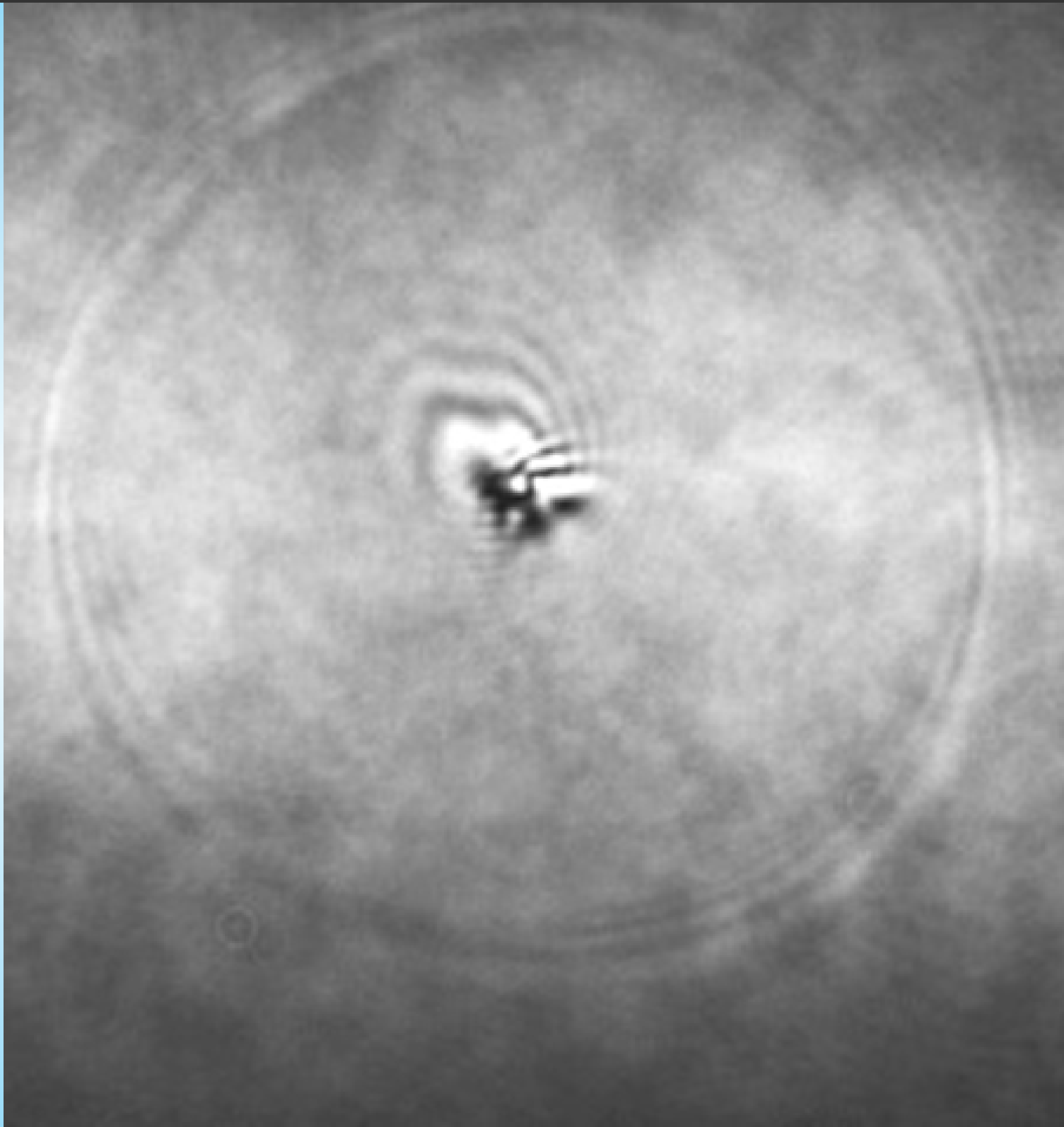
Dynamics

sapphire

3 μJ pulse

3.8 ns delay

40 μm radius



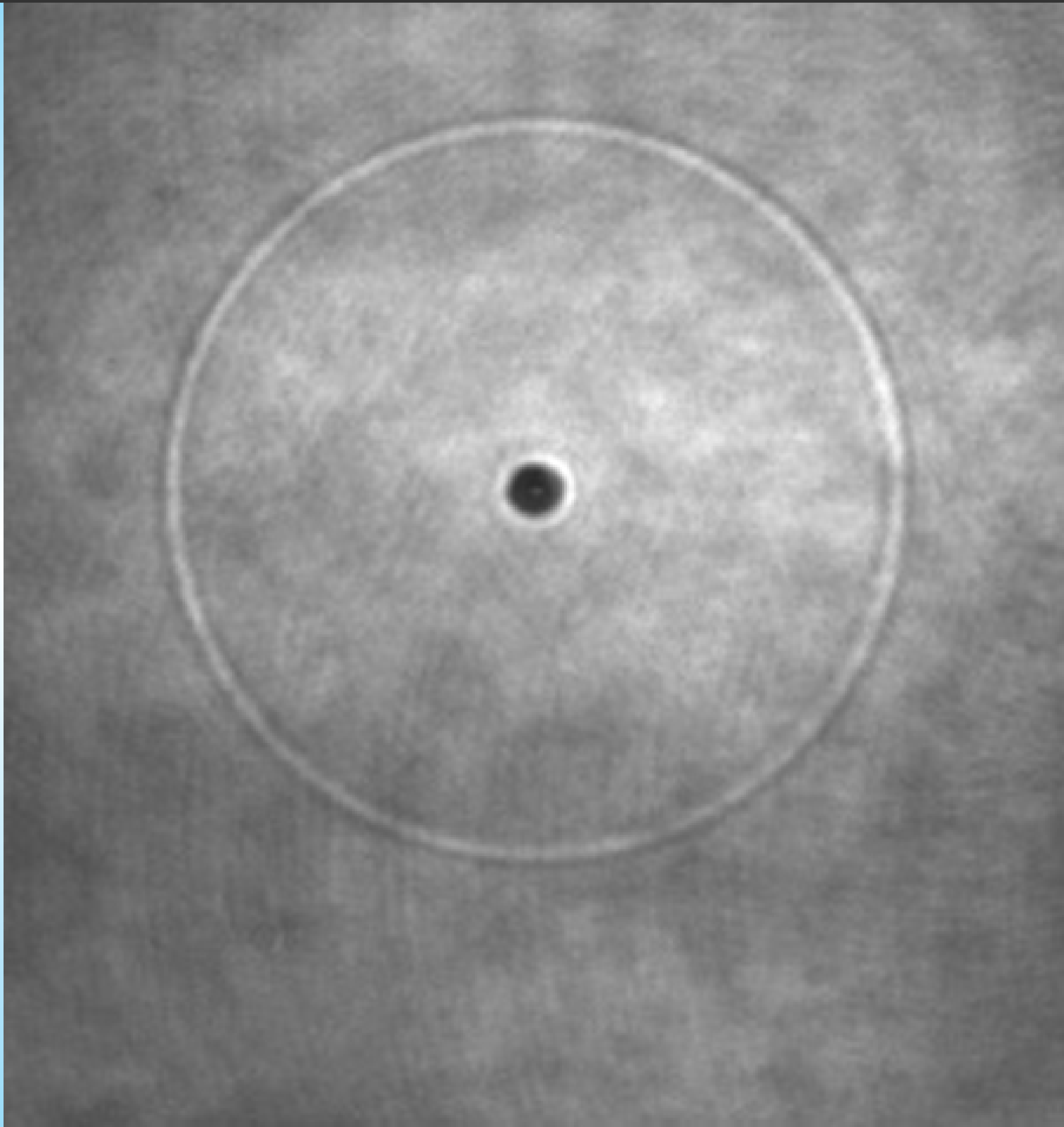
Dynamics

water ("self-healing")

1.0 μJ pulse

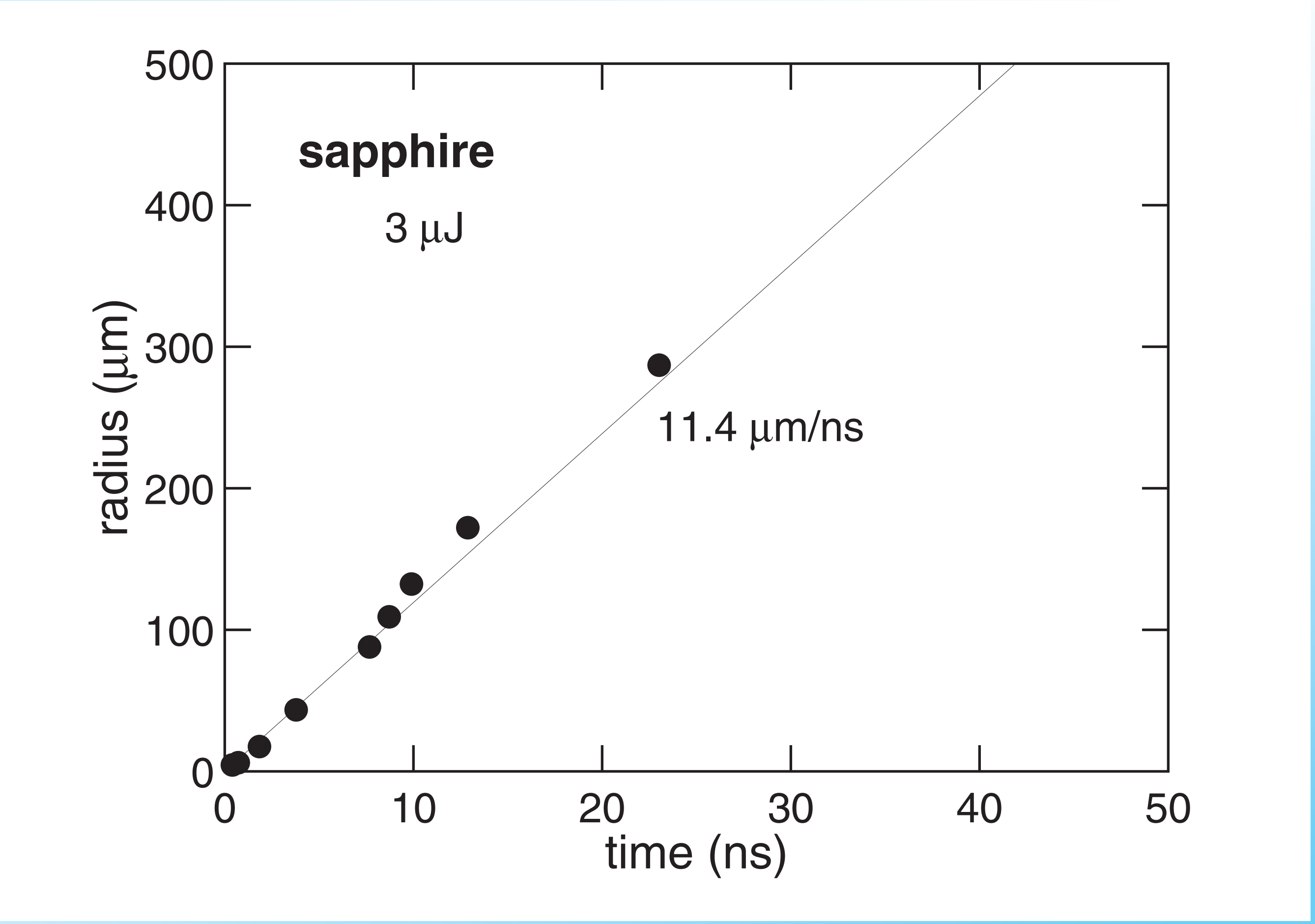
35 ns delay

58 μm radius



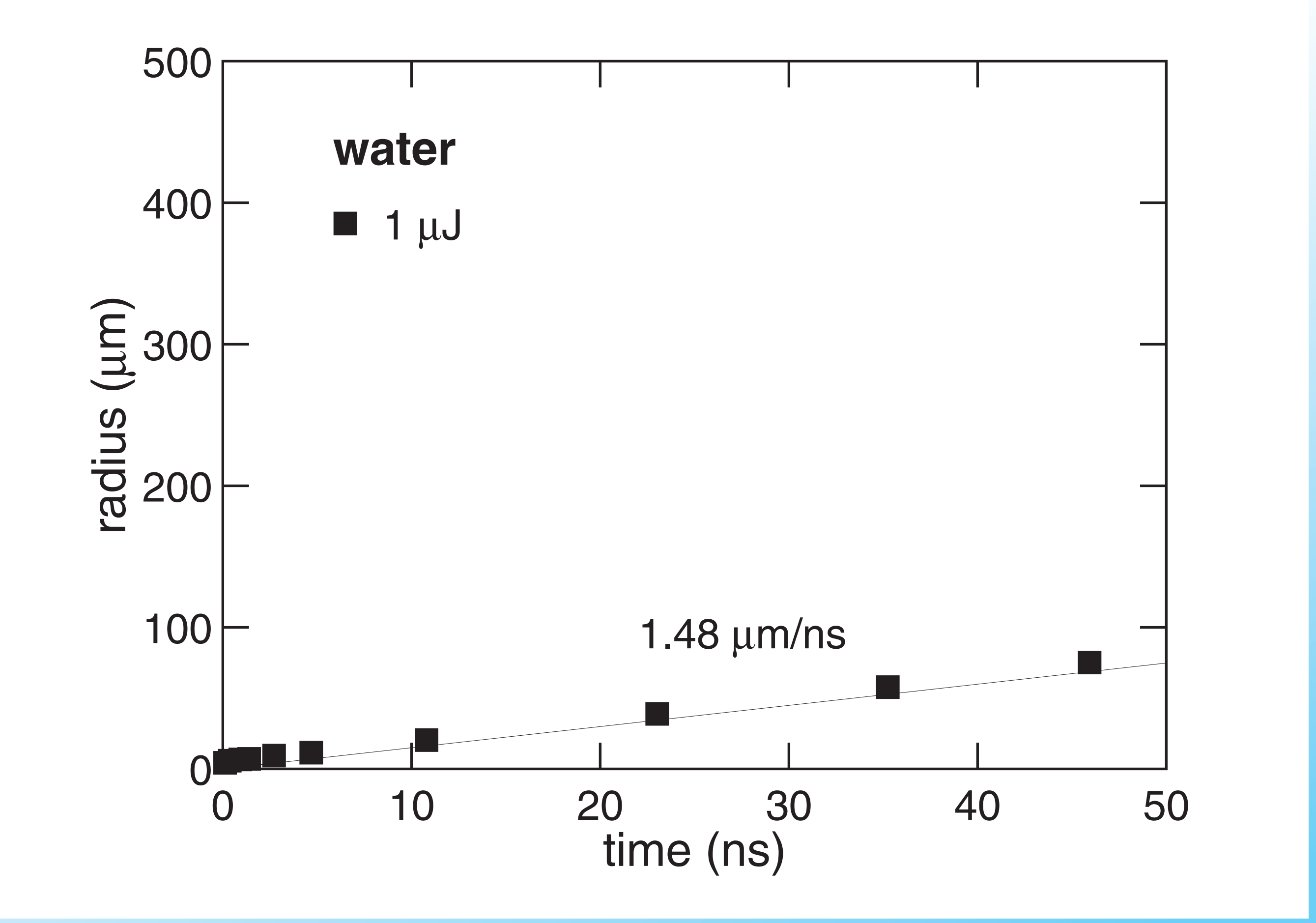
Dynamics

Dynamics



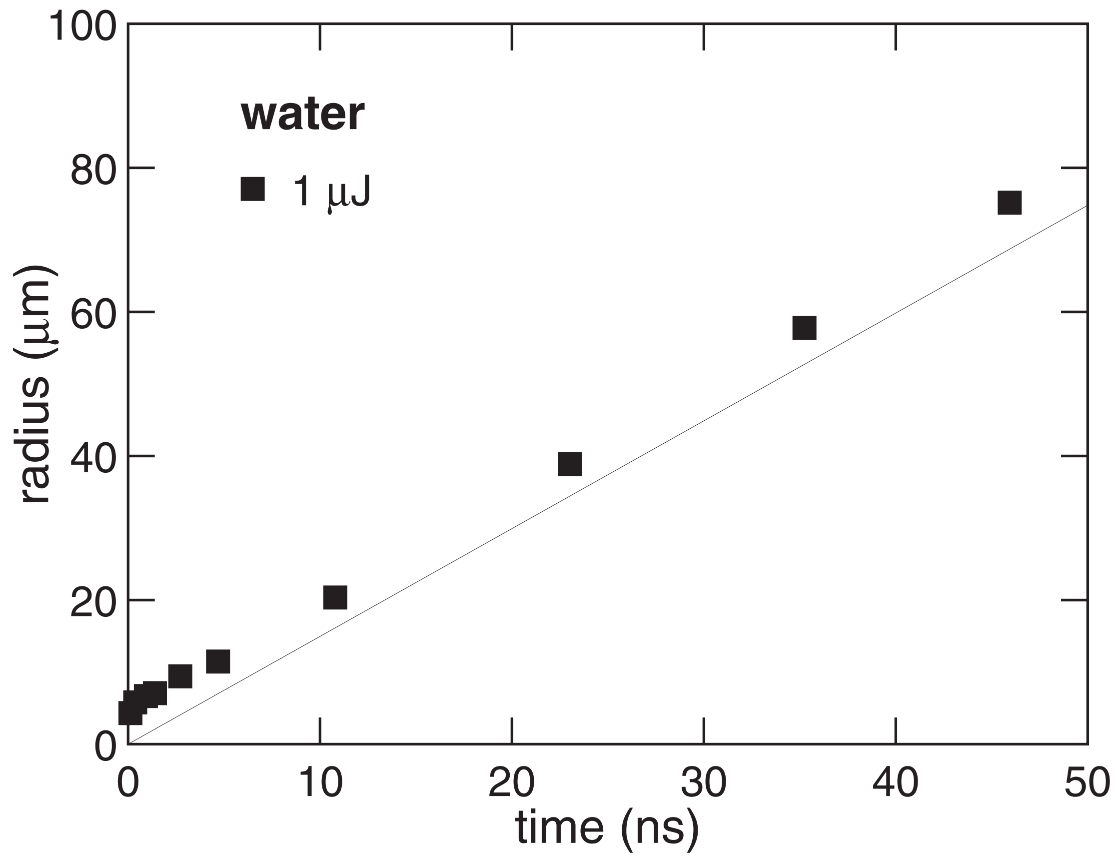
sapphire
3 μJ
11.4 μm/ns
radius (μm)
time (ns)
0
100
200
300
400
500
0
10
20
30
40
50

Dynamics

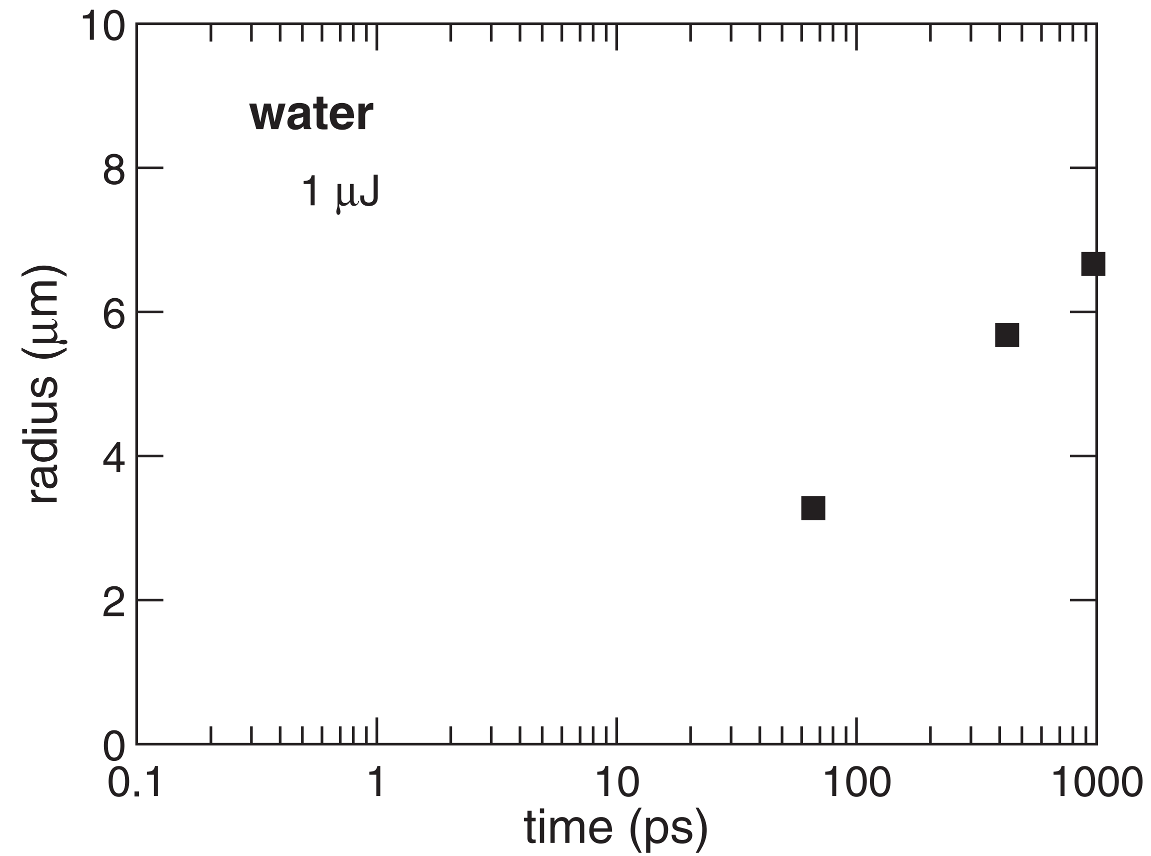


water
1 μJ
1.48 μm/ns
radius (μm)
time (ns)
500
400
300
200
100
0
0
10
20
30
40
50

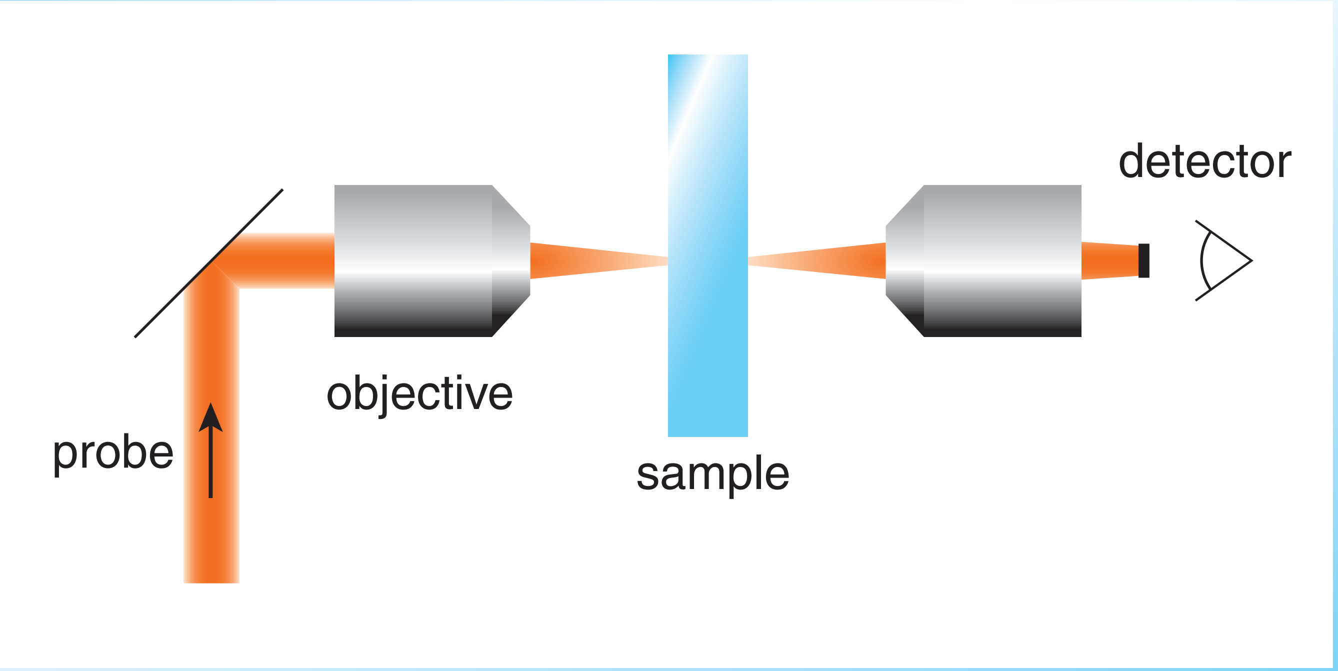
Dynamics



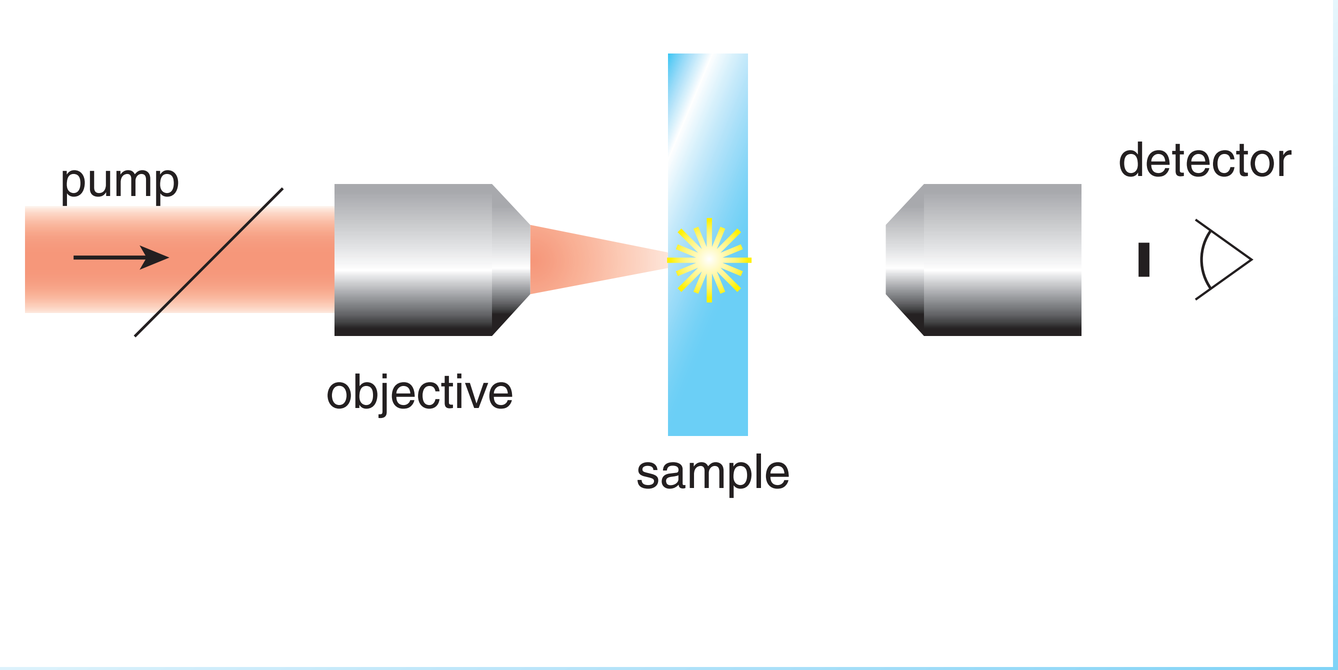
Dynamics



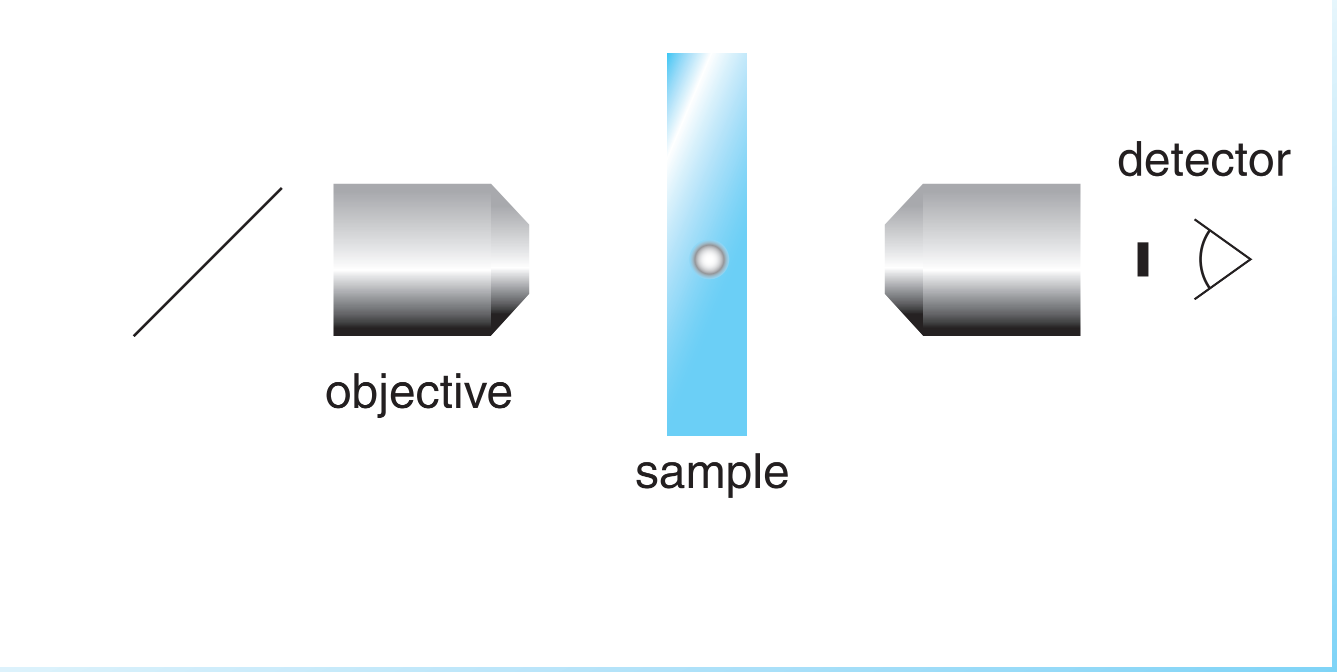
time-resolved scattering setup



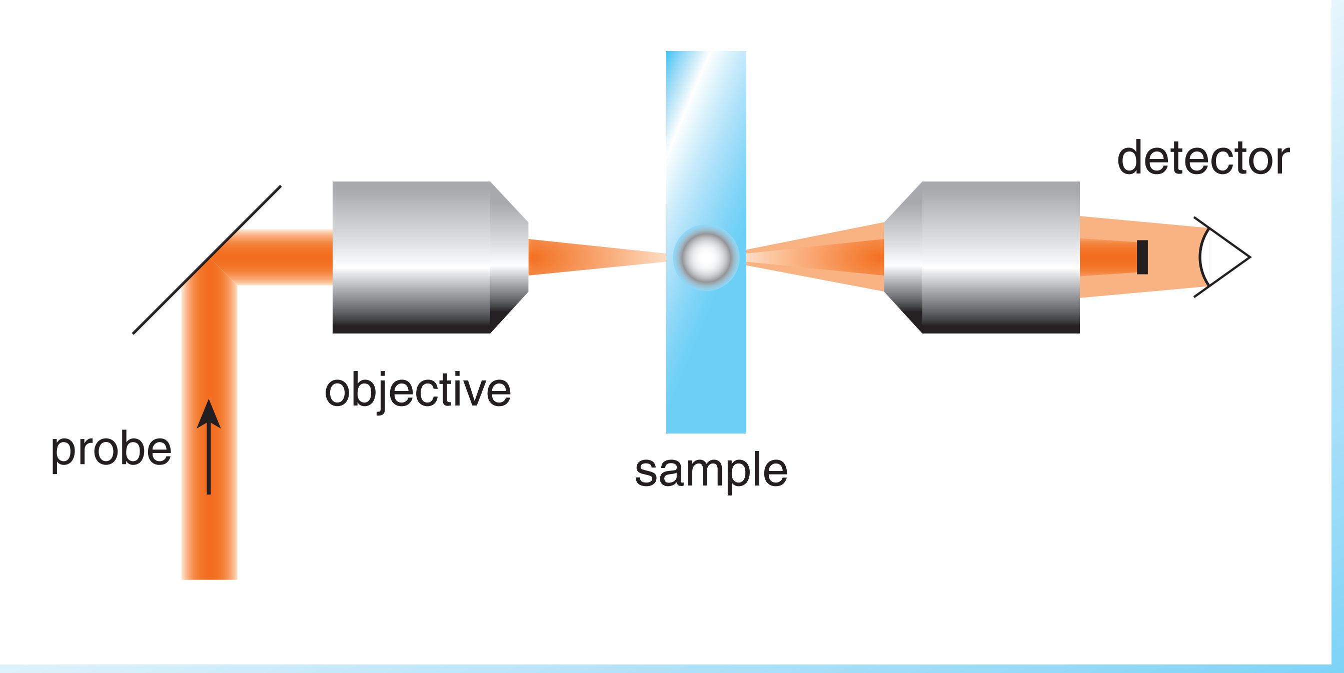
time-resolved scattering setup



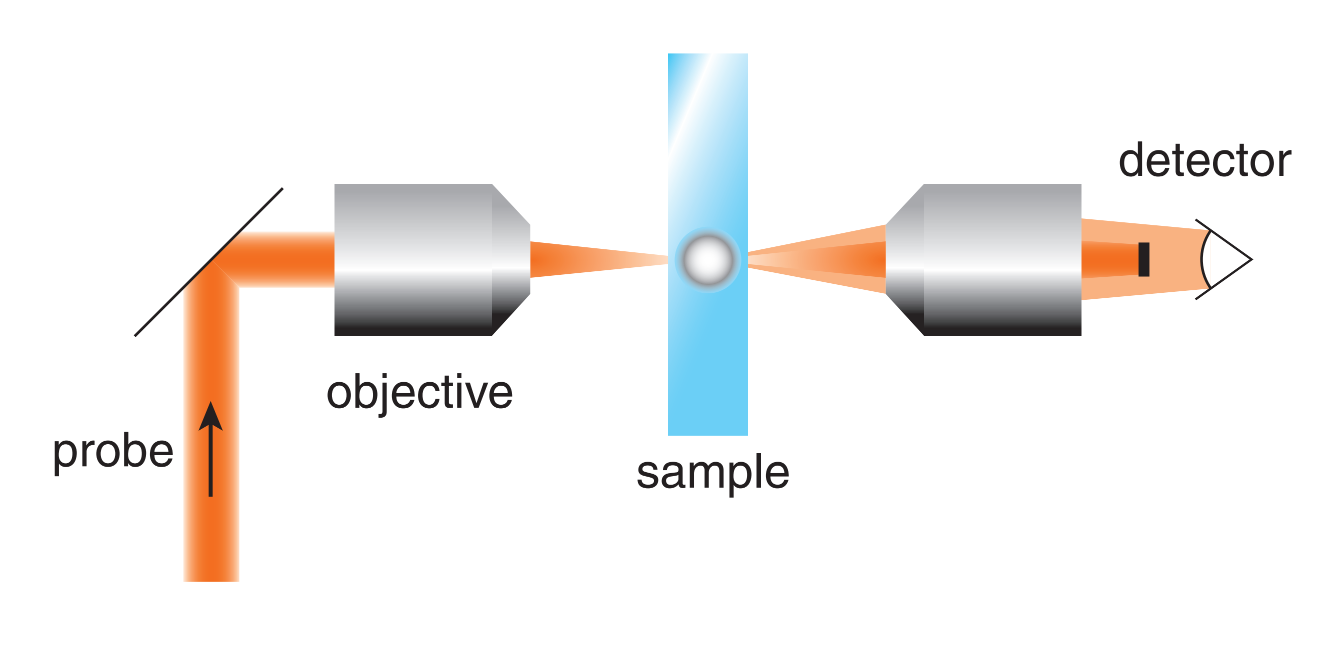
time-resolved scattering setup



time-resolved scattering setup

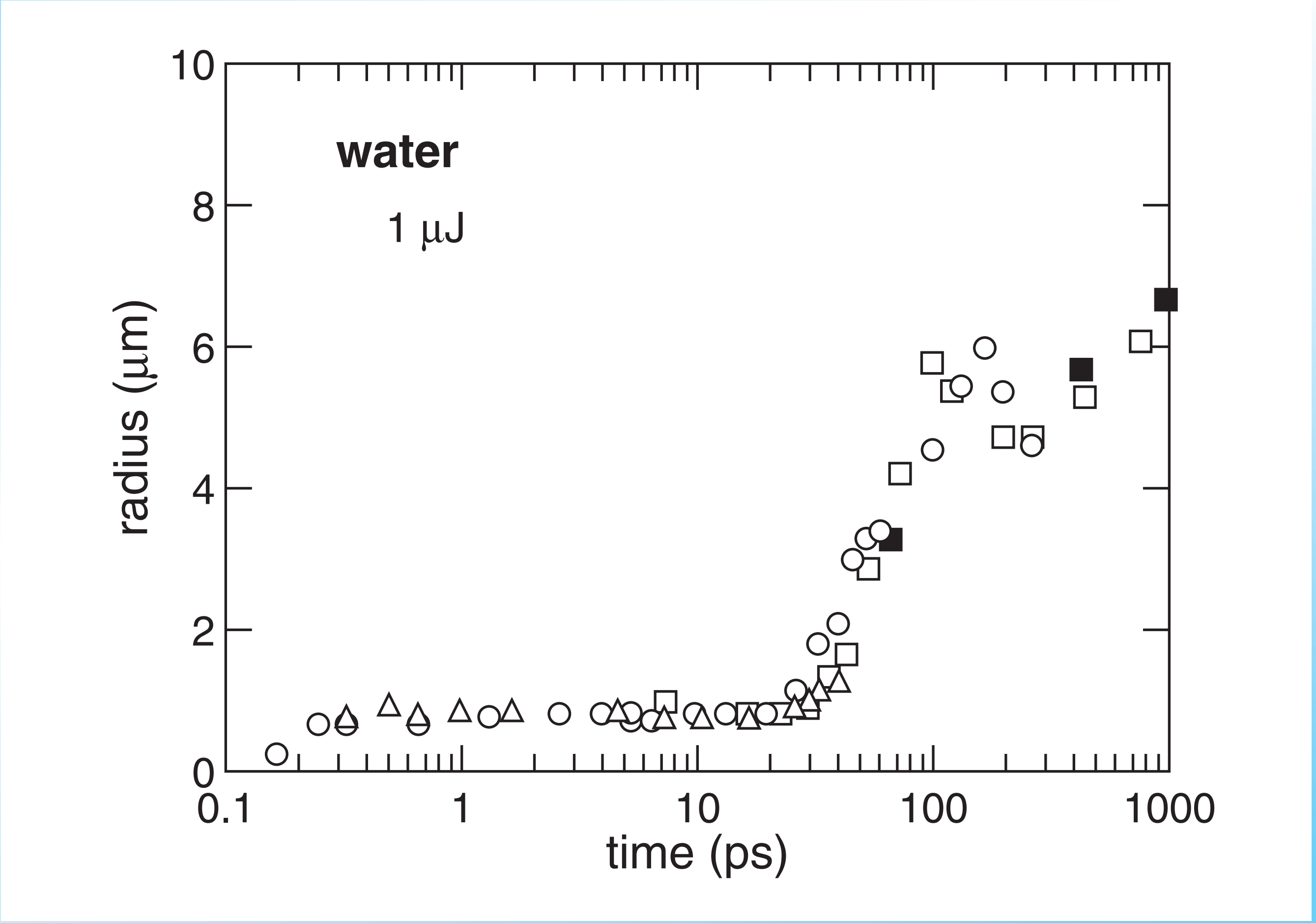


time-resolved scattering setup

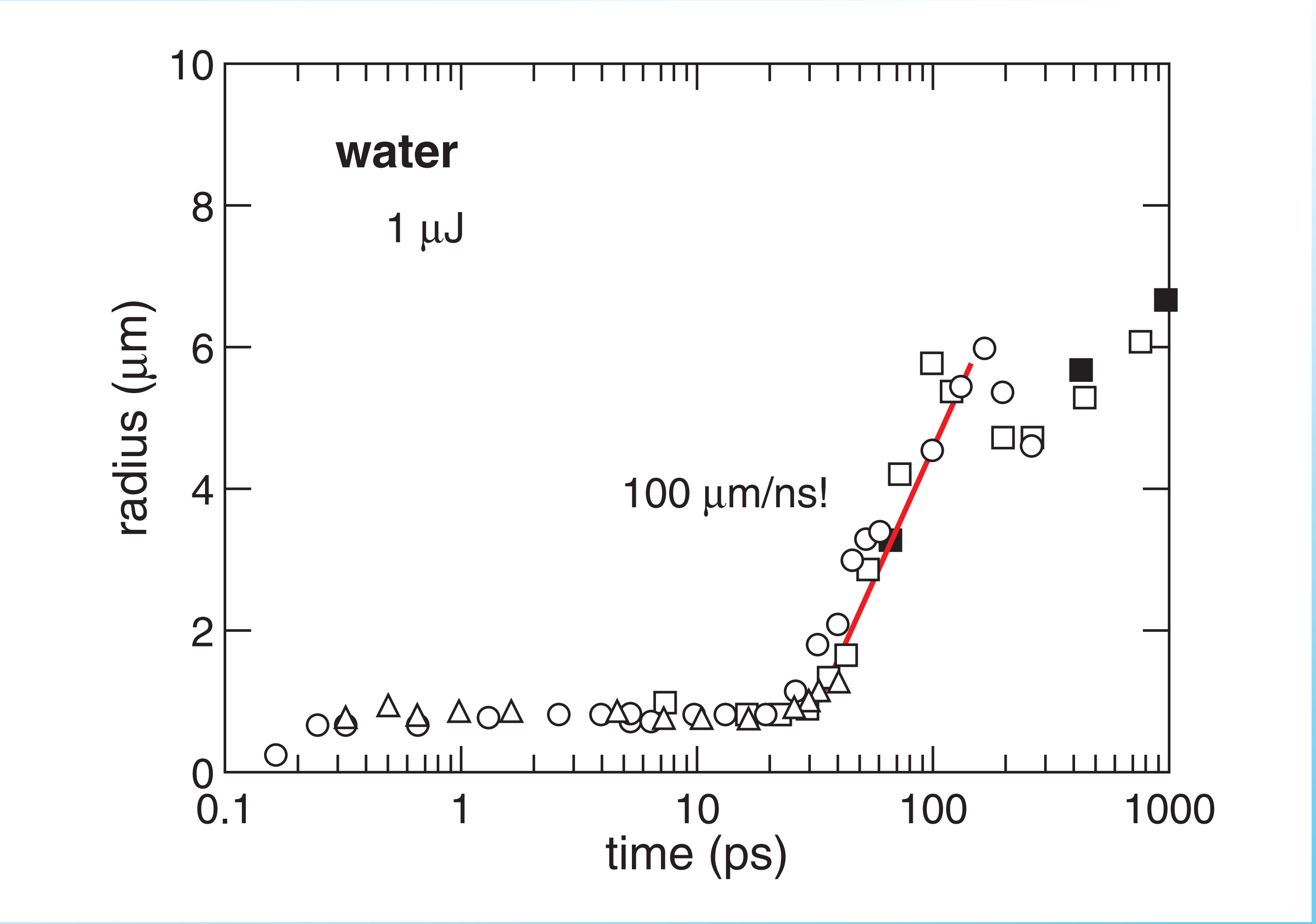


signal proportional to area of scatterer

Dynamics



Dynamics



Conclusions

- **submicron-scale bulk micromachining**
- **weak bandgap and wavelength dependence**
- **only a few nanojoules required**


GORDON McKAY
LABORATORY OF
APPLIED SCIENCE
HARVA

Funding: National Science Foundation

Acknowledgments:
Prof. N. Bloembergen
Prof. H. Ehrenreich
Prof. T. Kaxiras
Prof. C. Klingshirn

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

http://mazur-www.harvard.edu