Quick as a flash: Observing Ultrafast Laser-Induced Dynamics in Semiconductors

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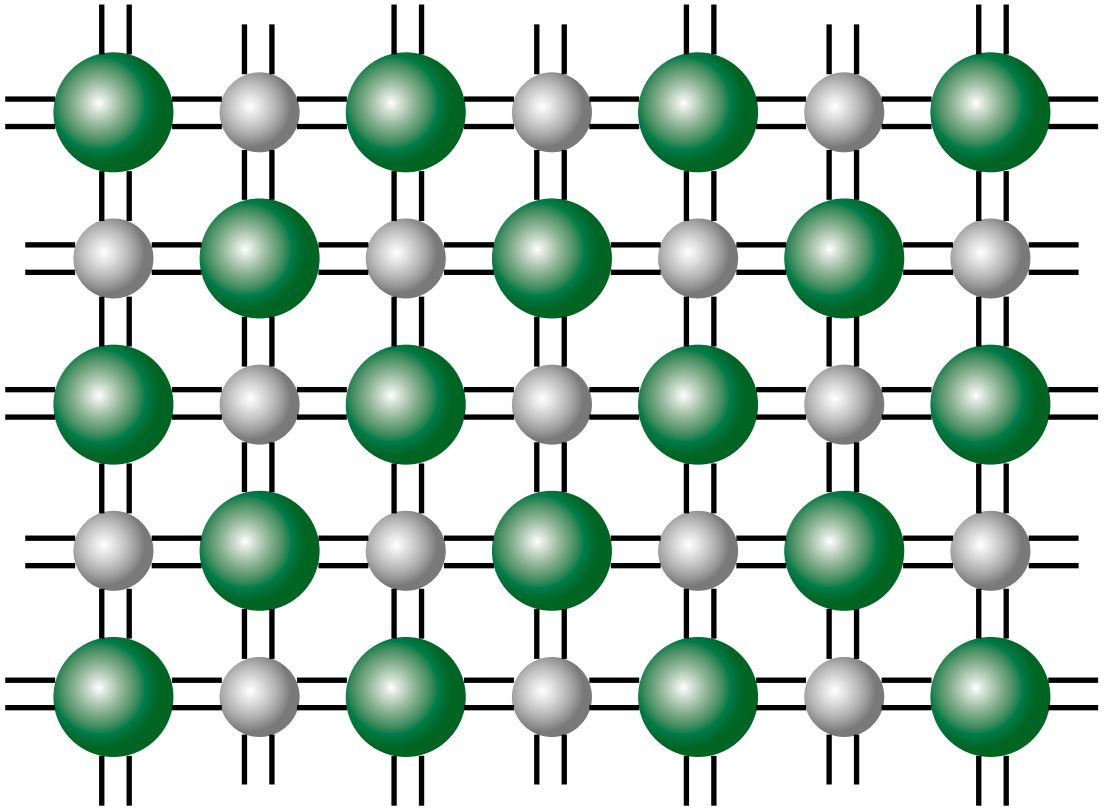
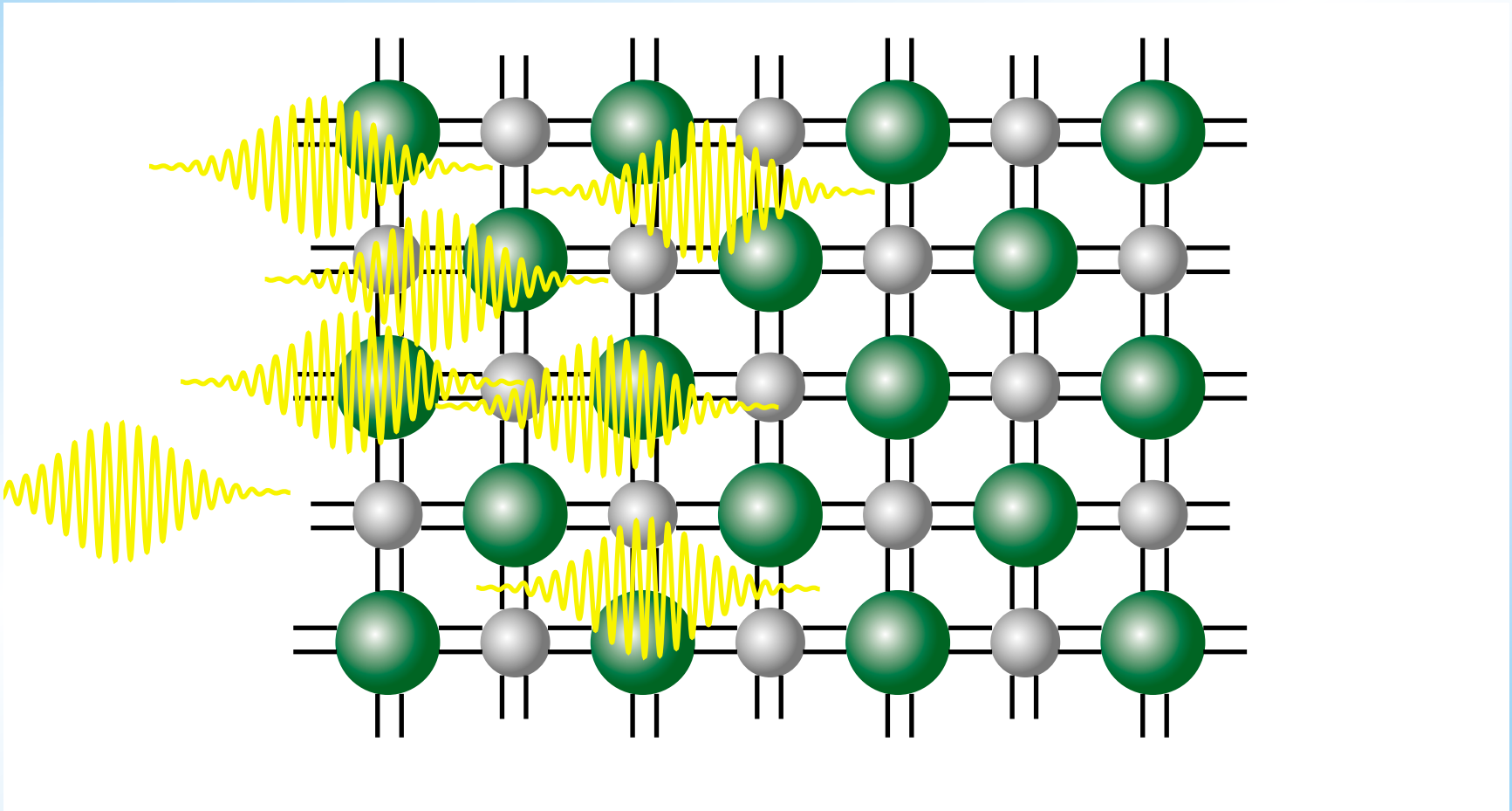
Introduction

how do femtosecond laser pulses alter a solid?



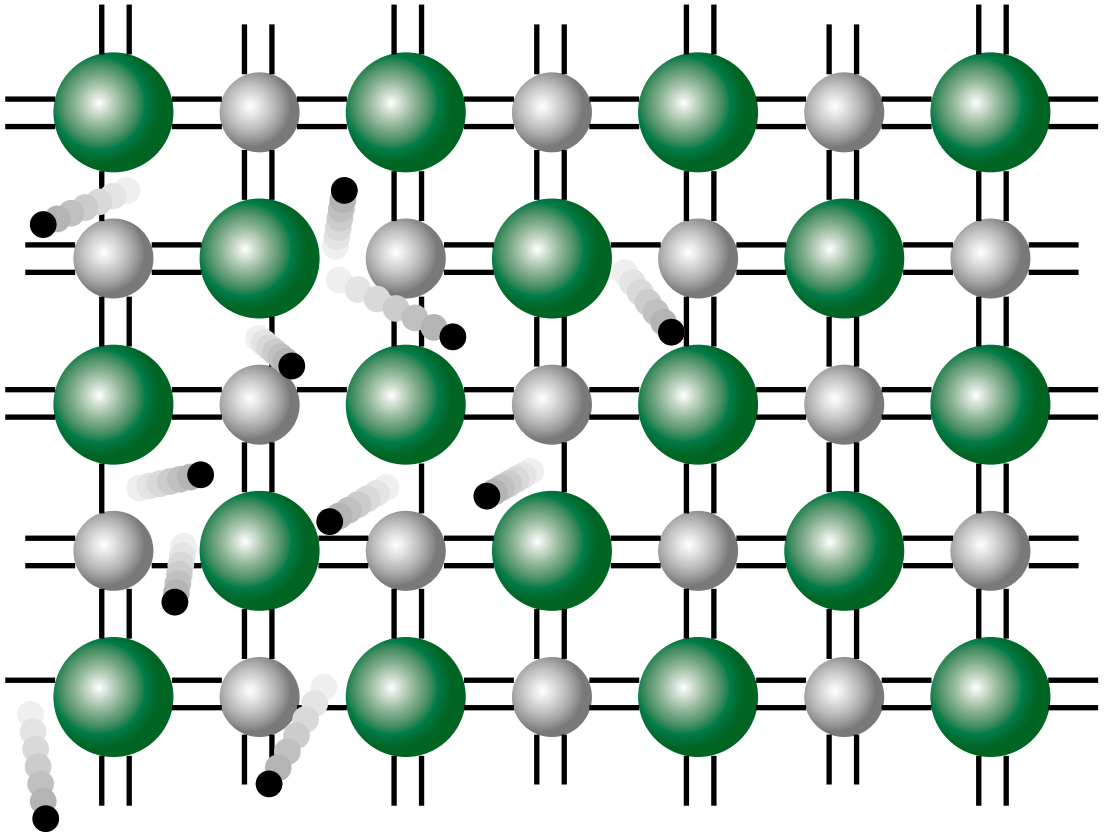
Introduction

photons excite valence electrons…



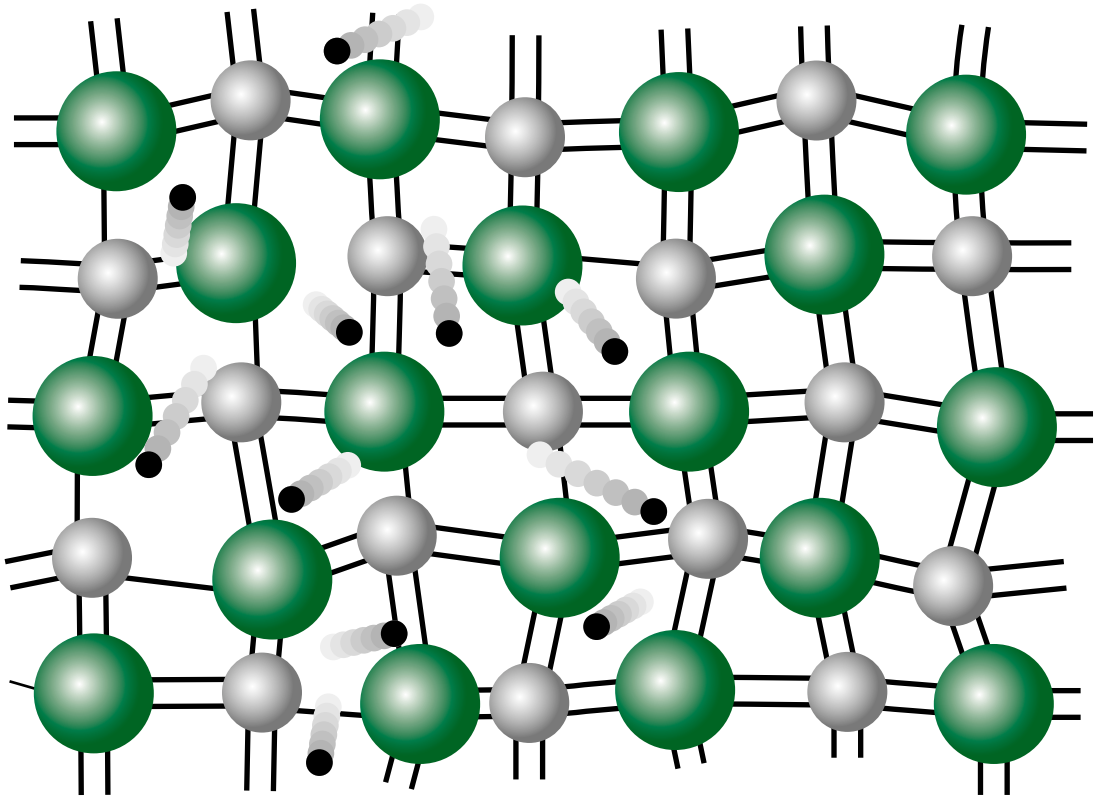
Introduction

…and create free electrons…



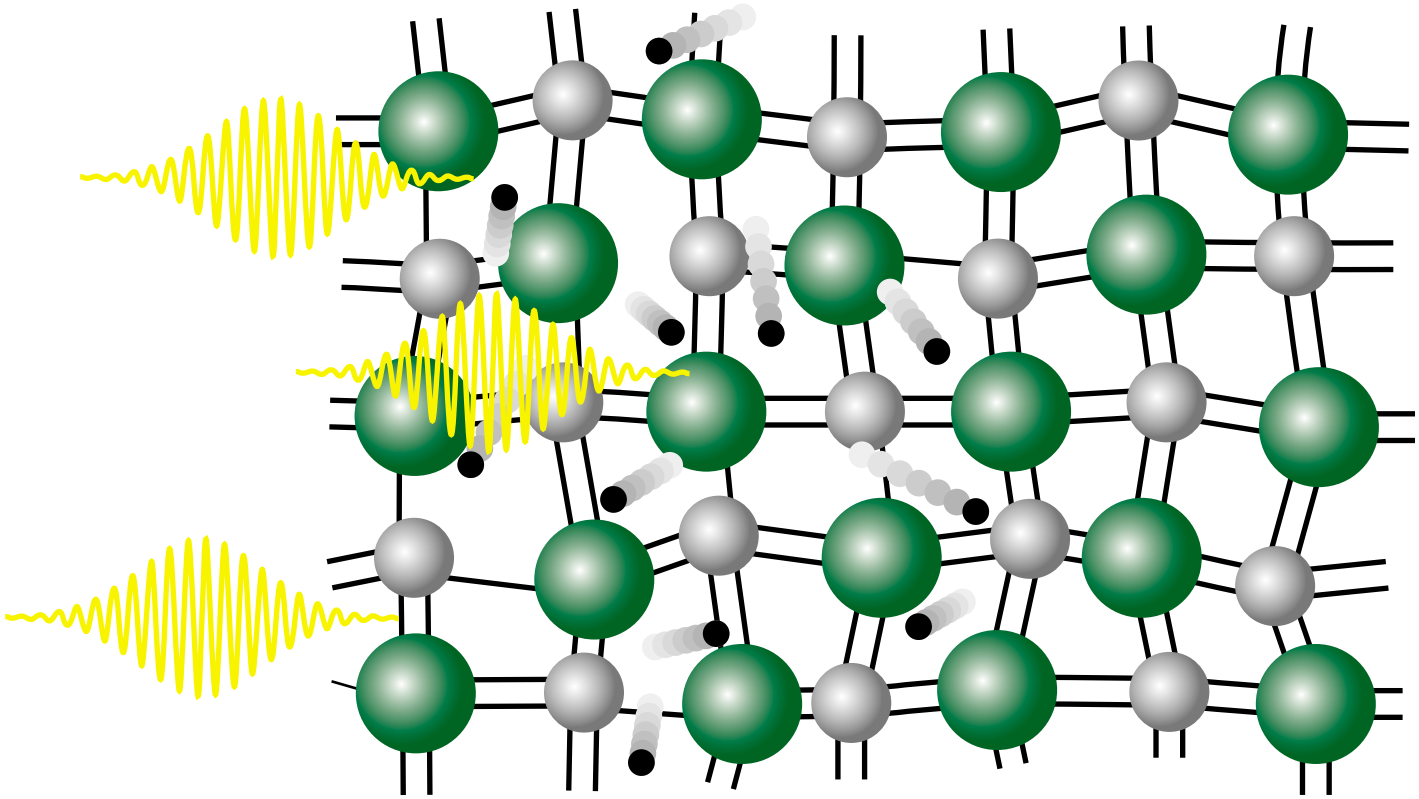
Introduction

…causing electronic and structural changes…



Introduction

…which we measure with another pulse



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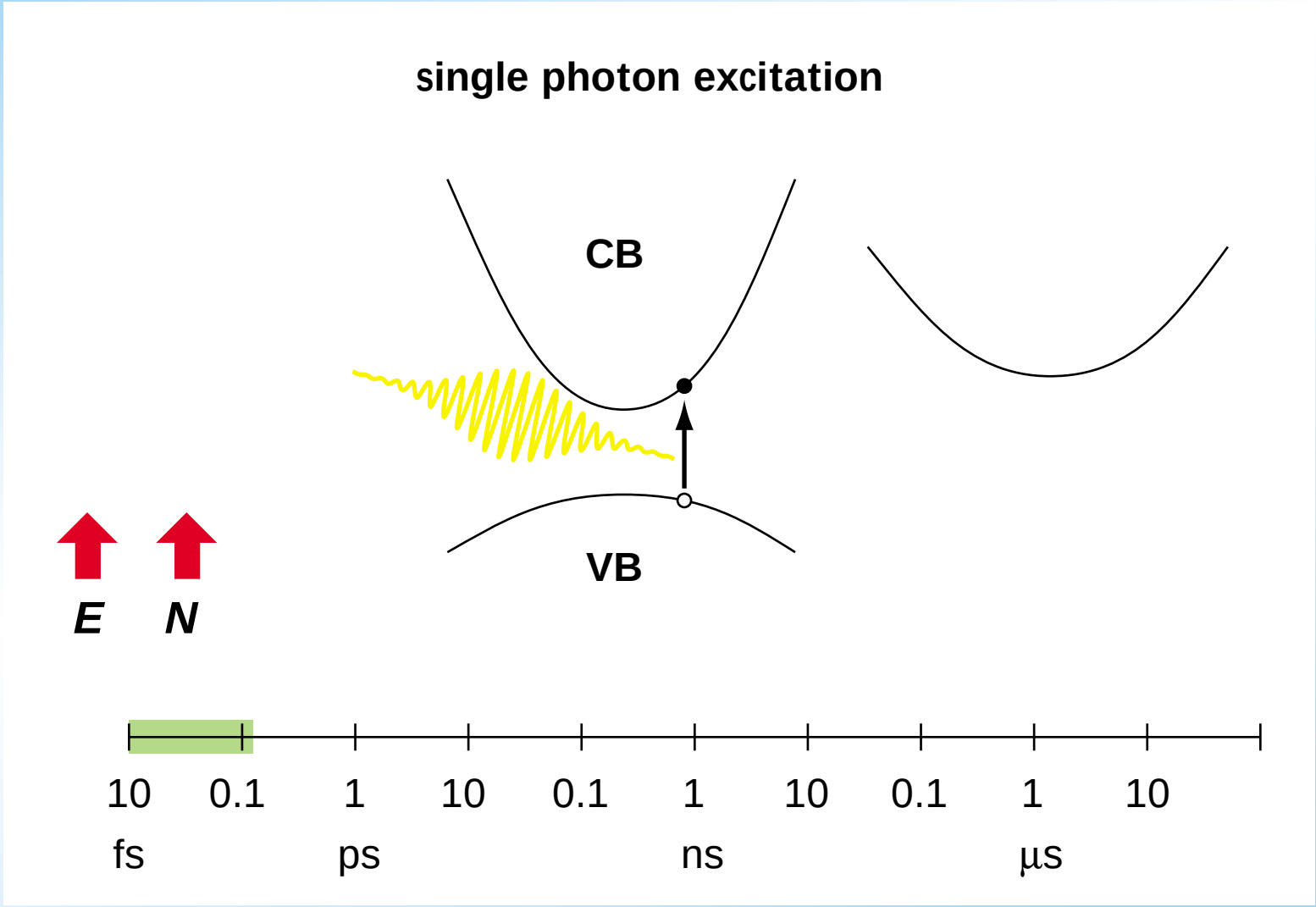

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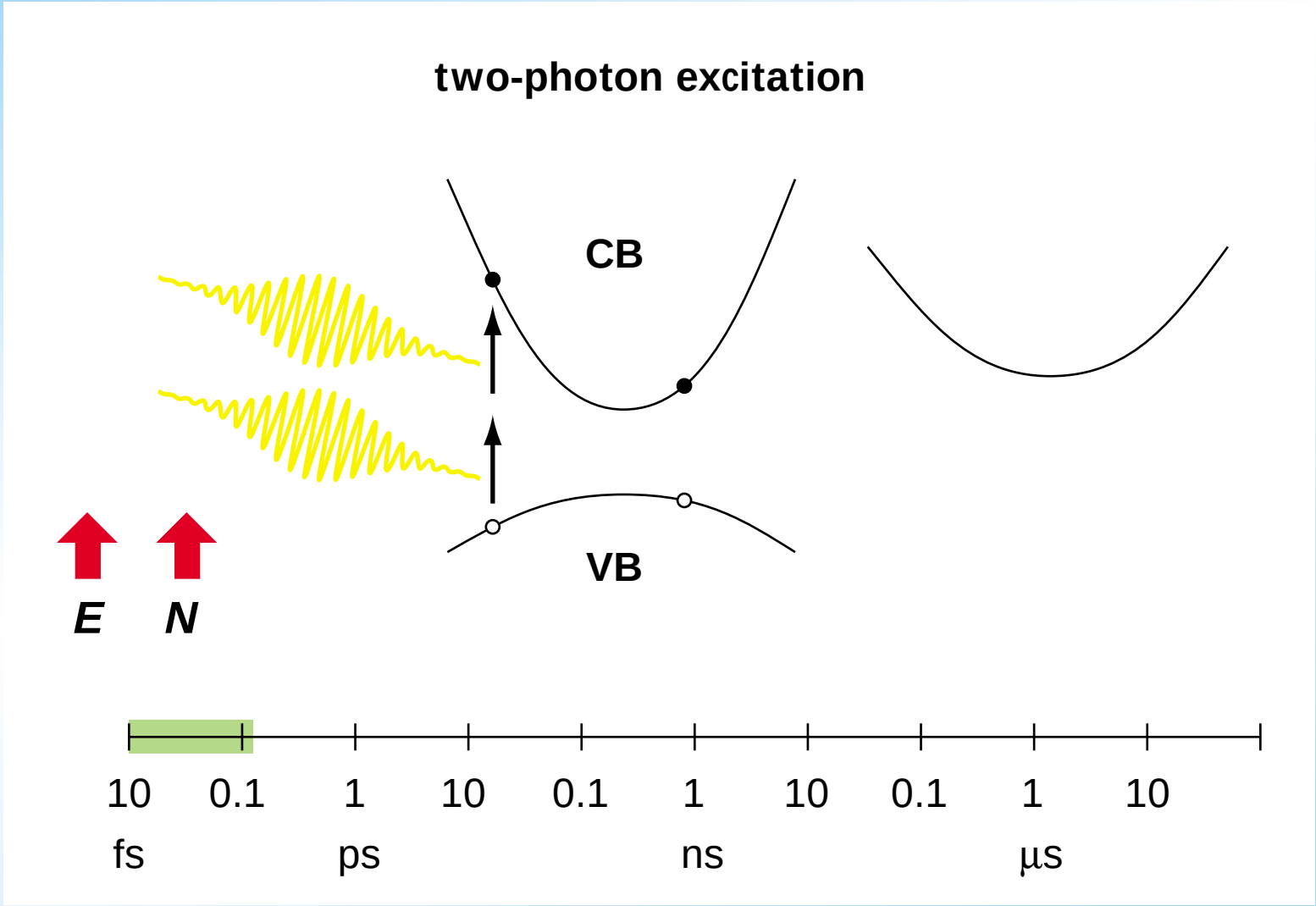

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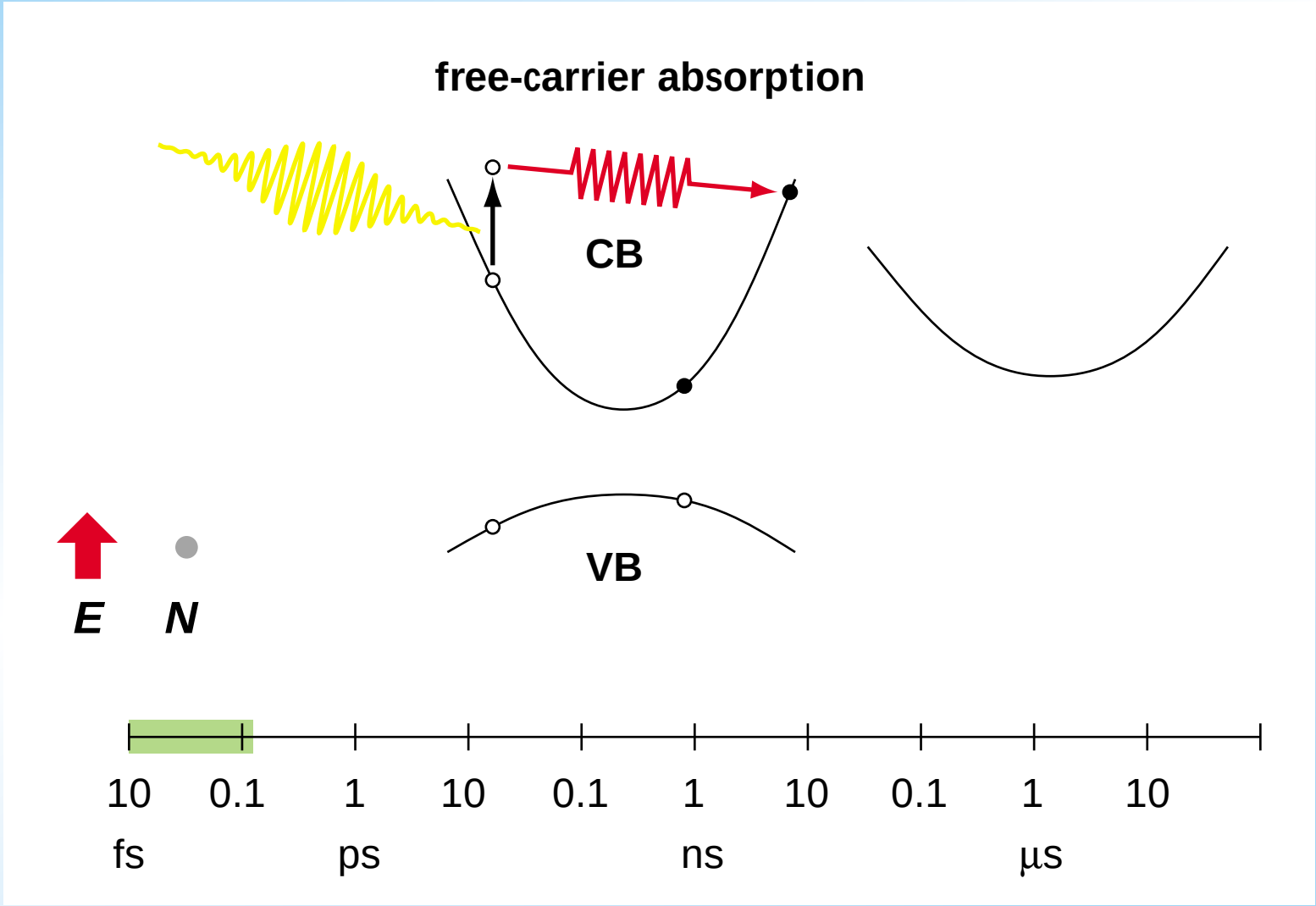

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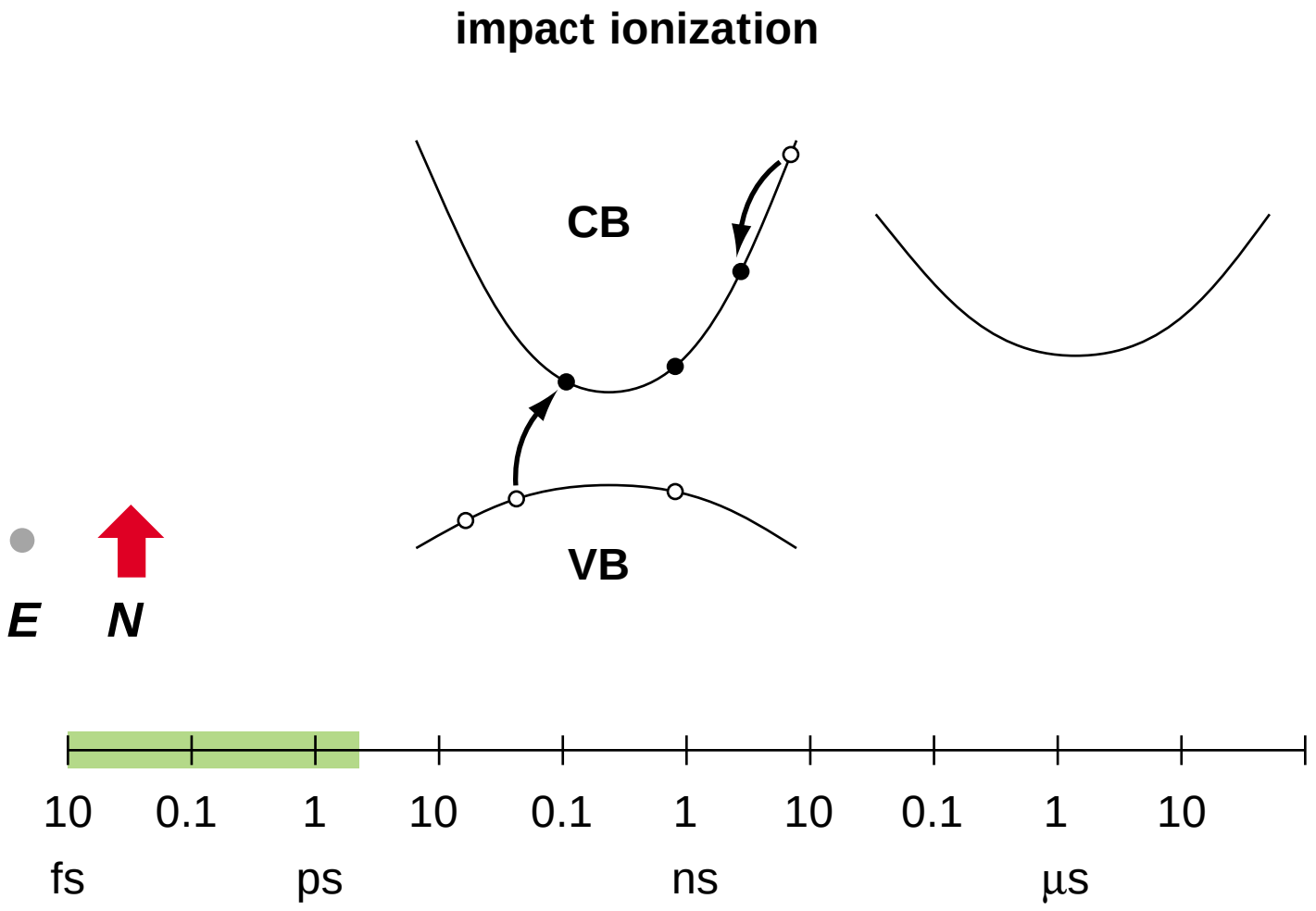

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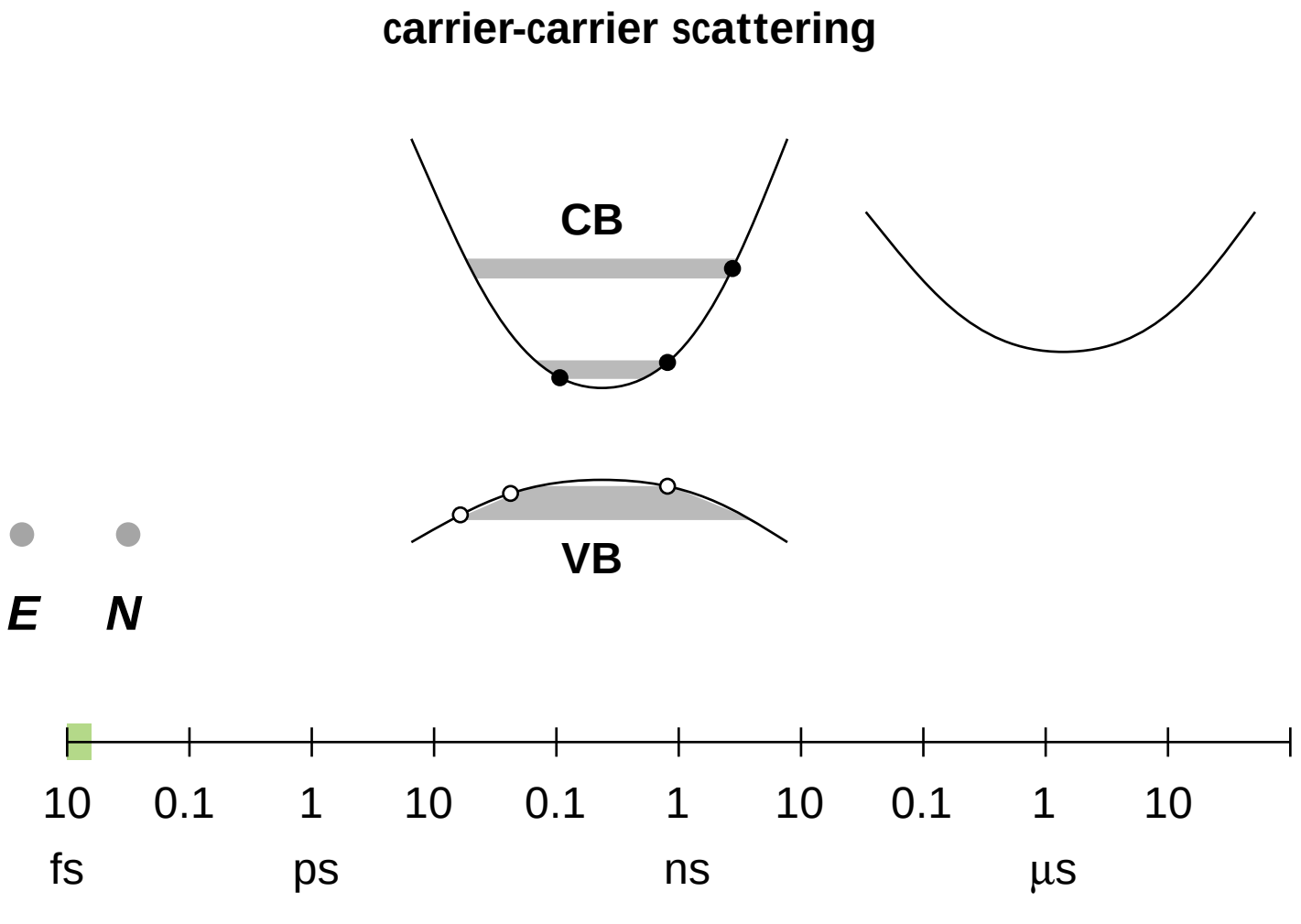

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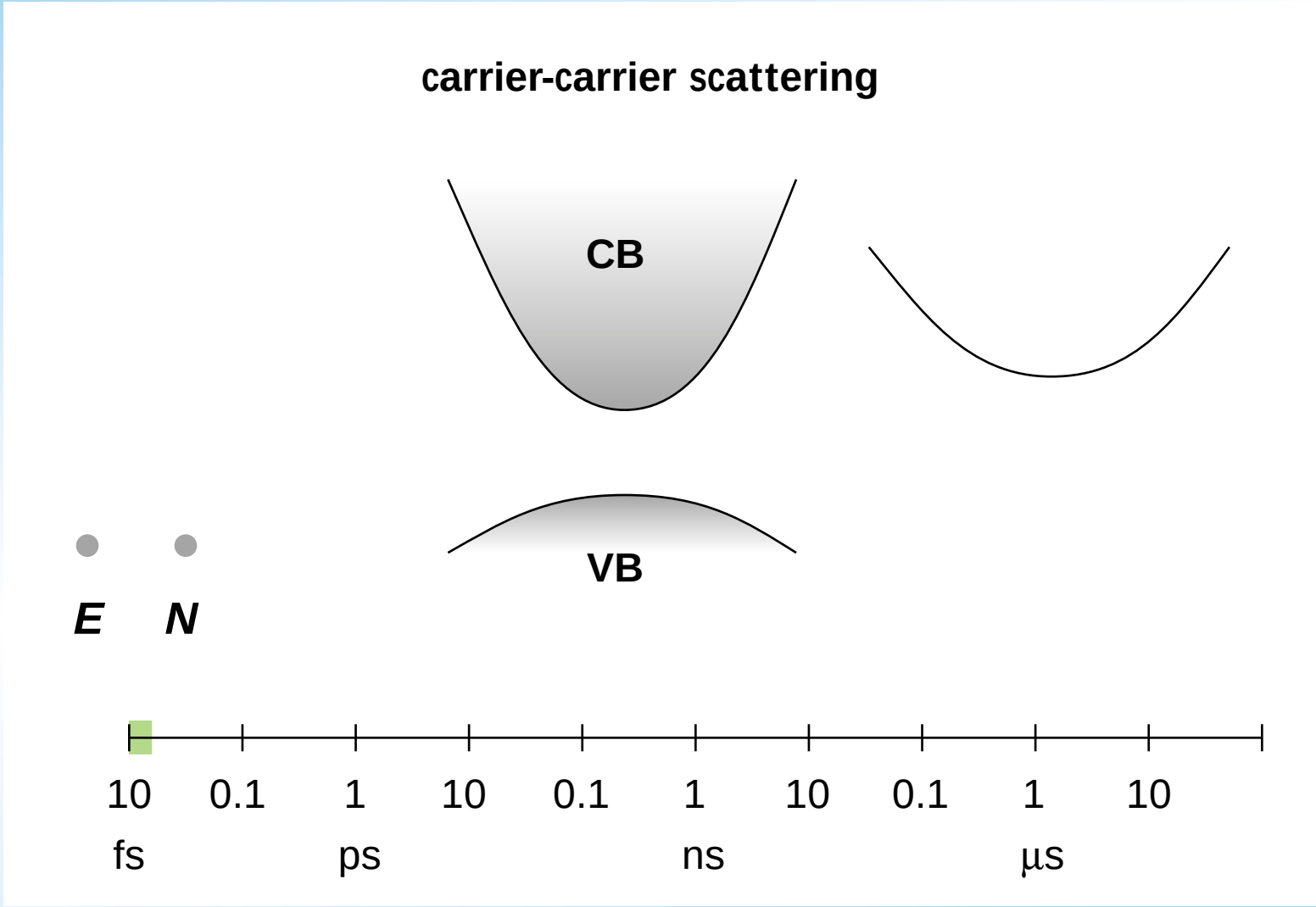

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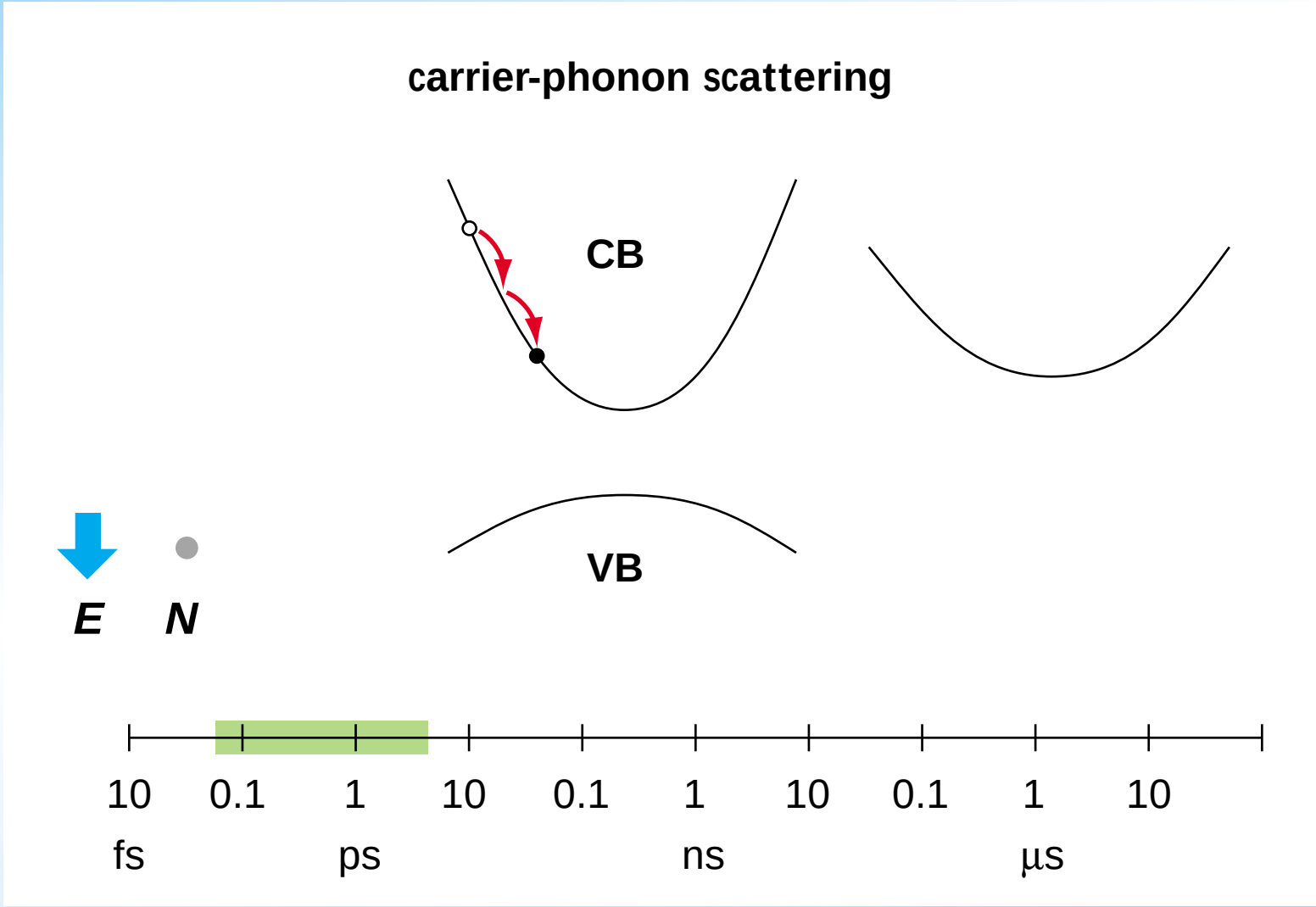

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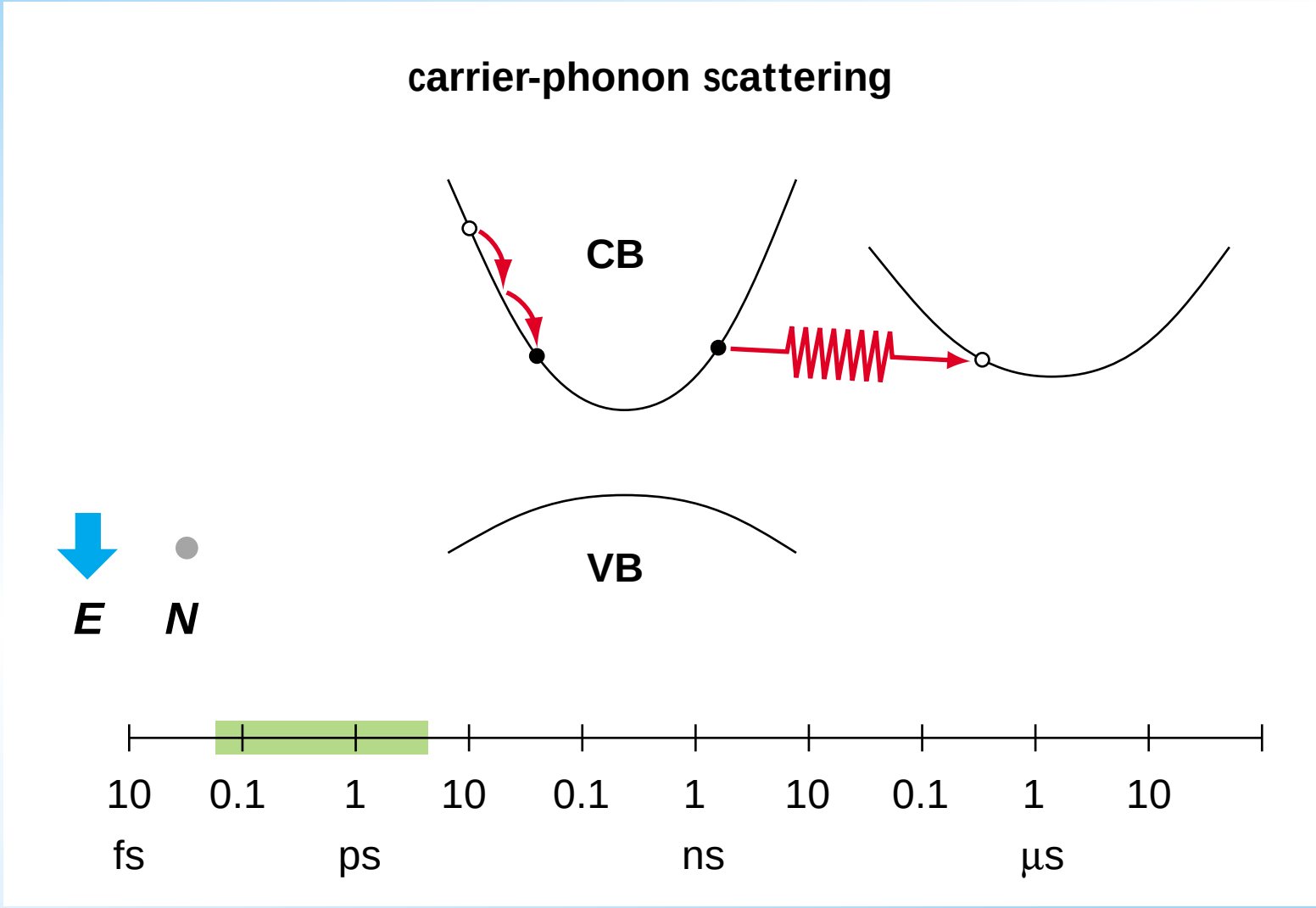

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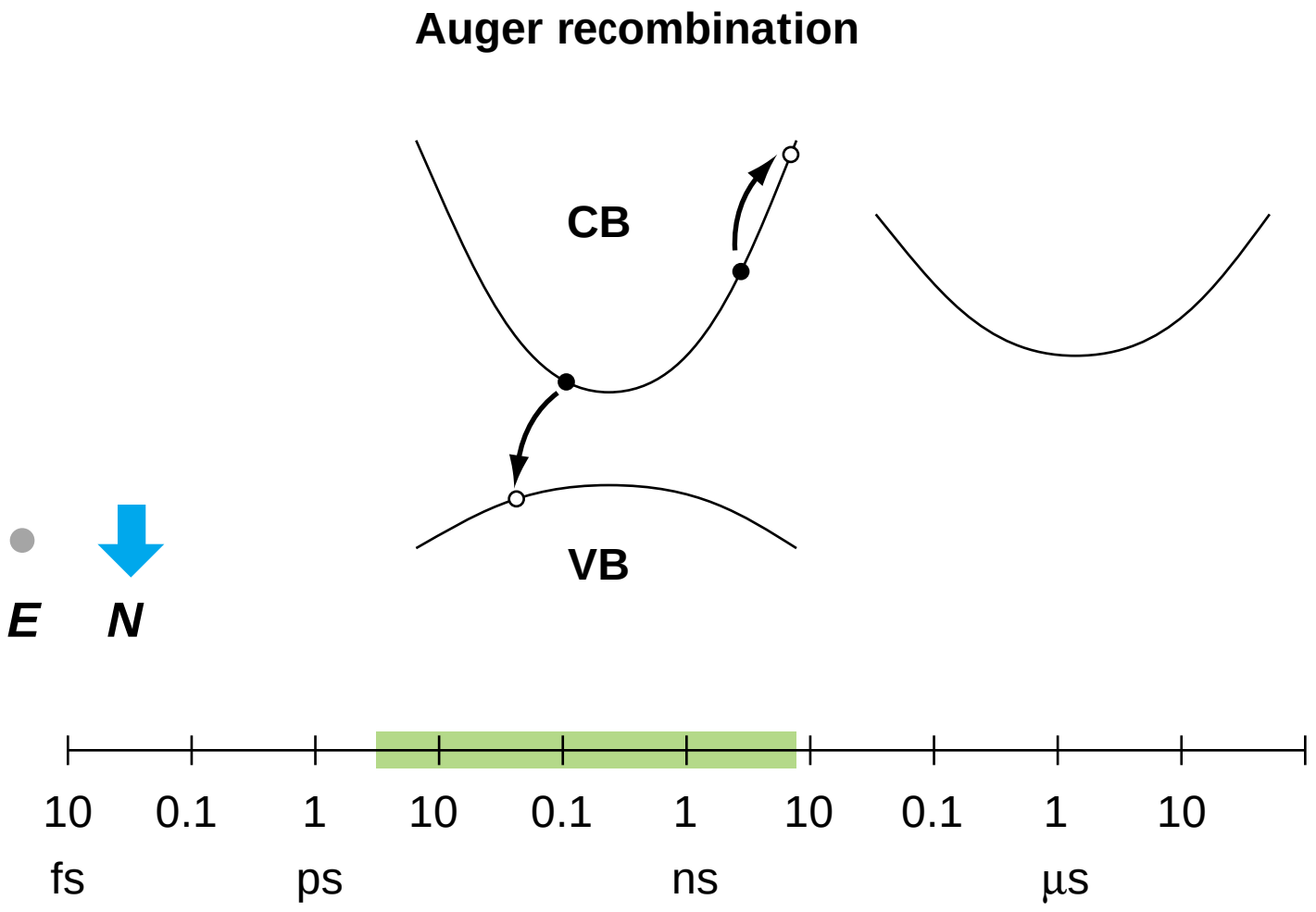

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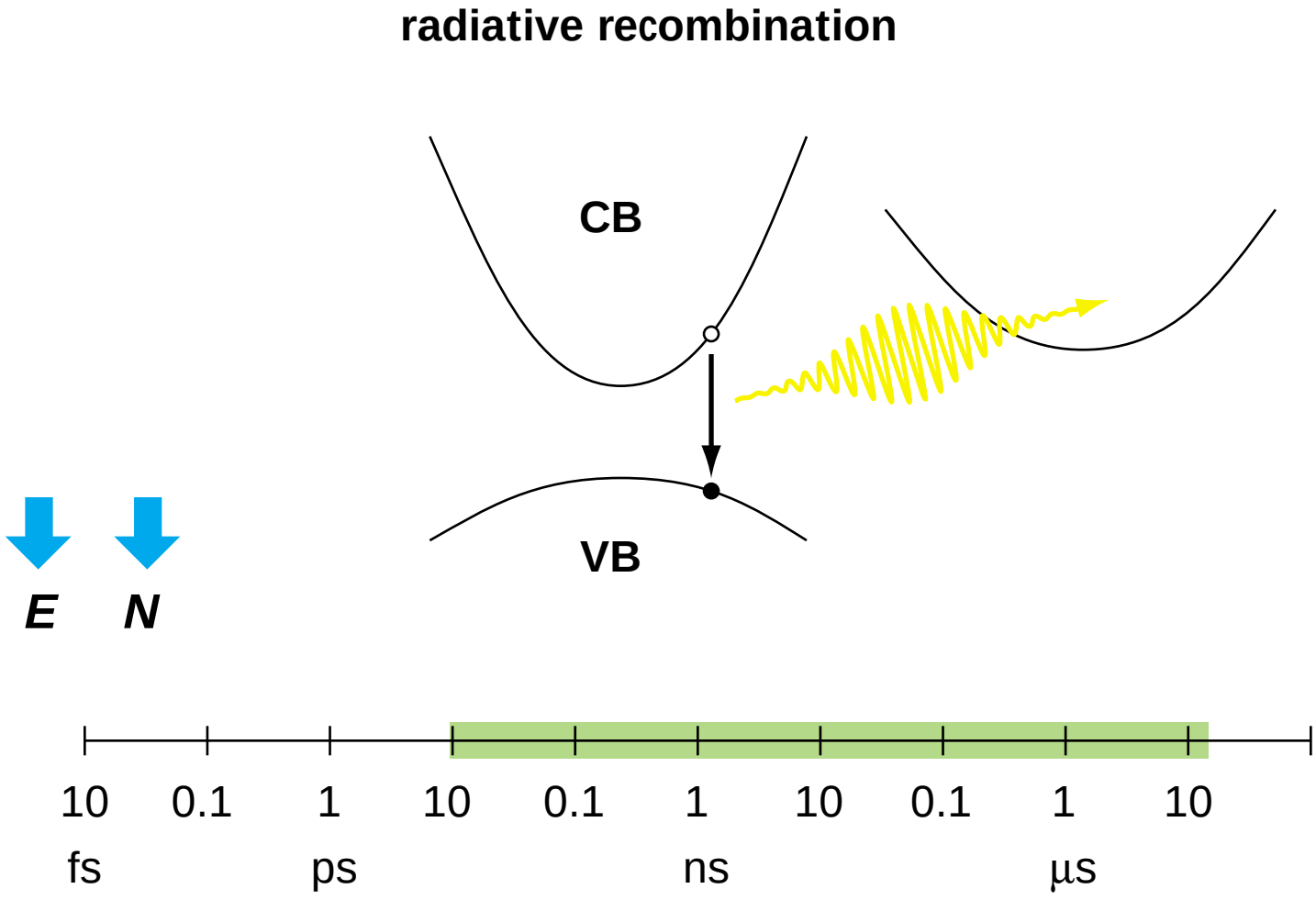

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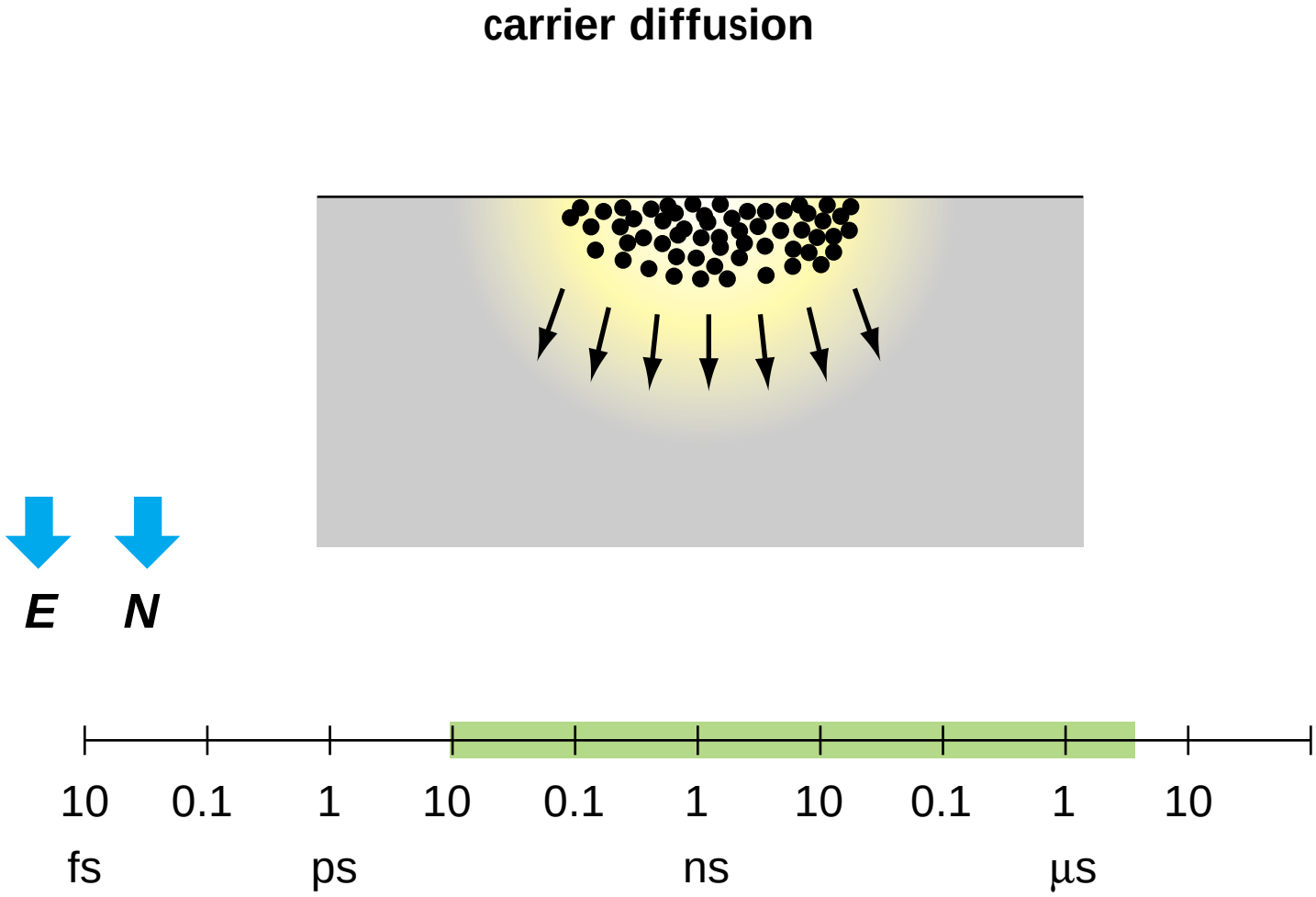

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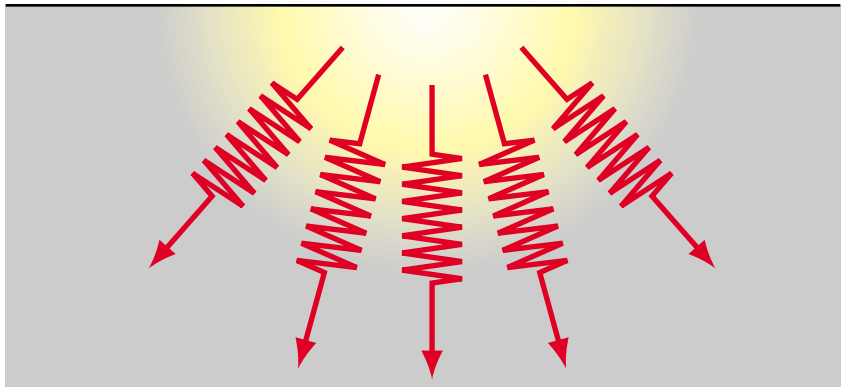
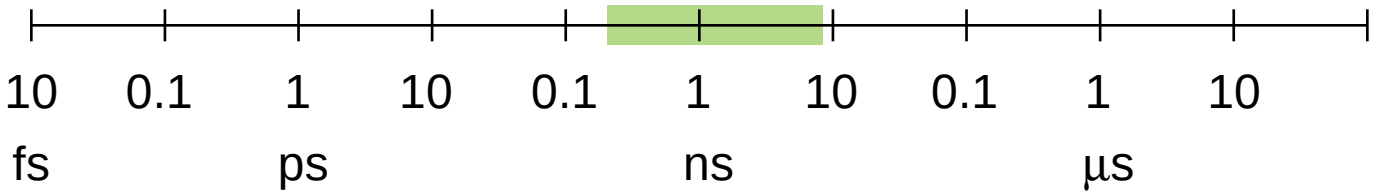
thermal diffusion





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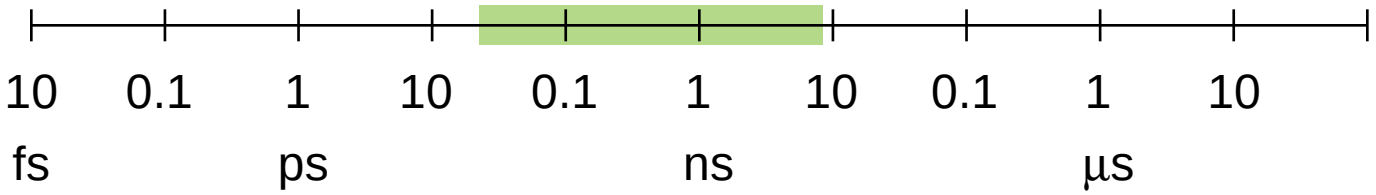
ablation and evaporation



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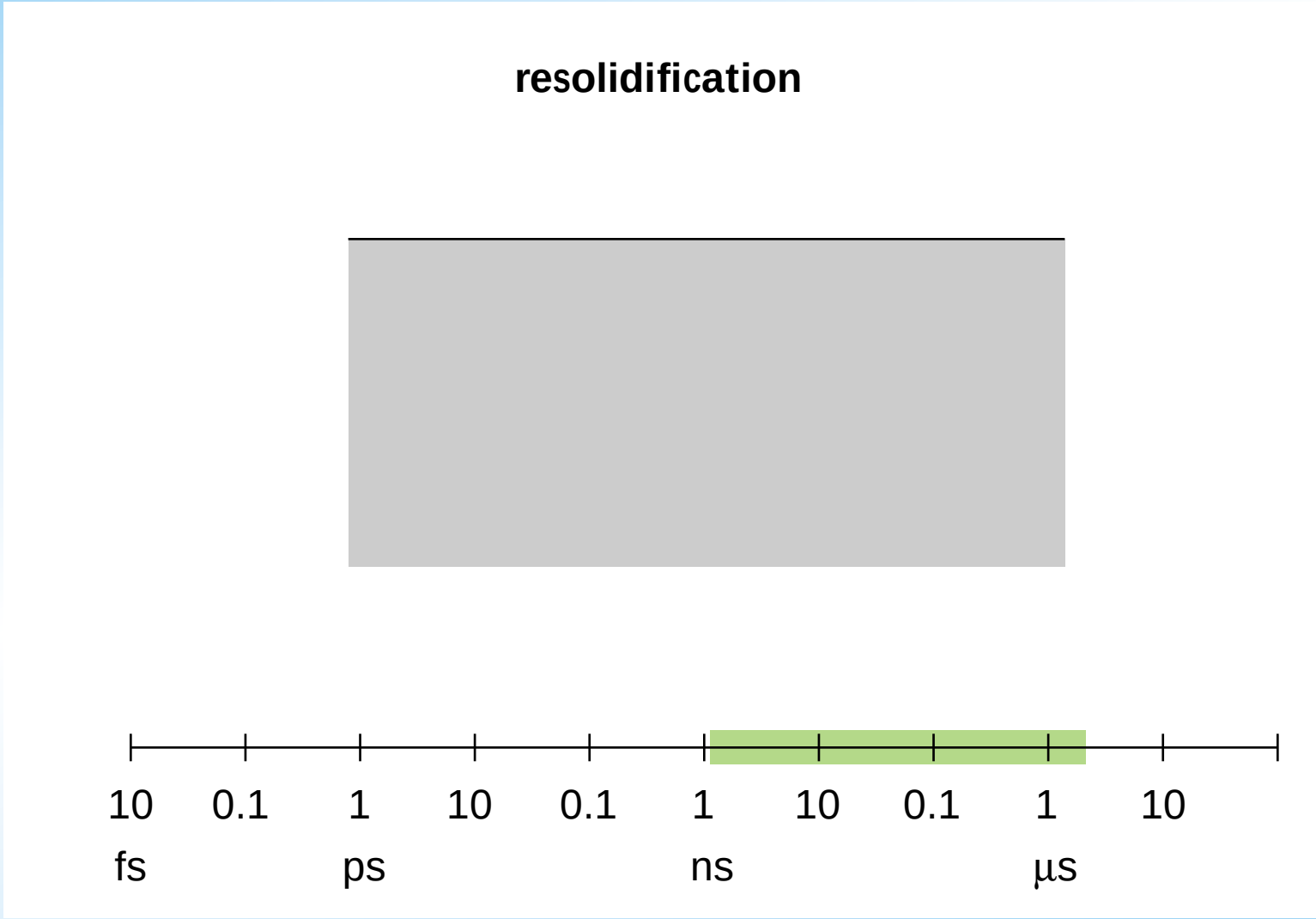

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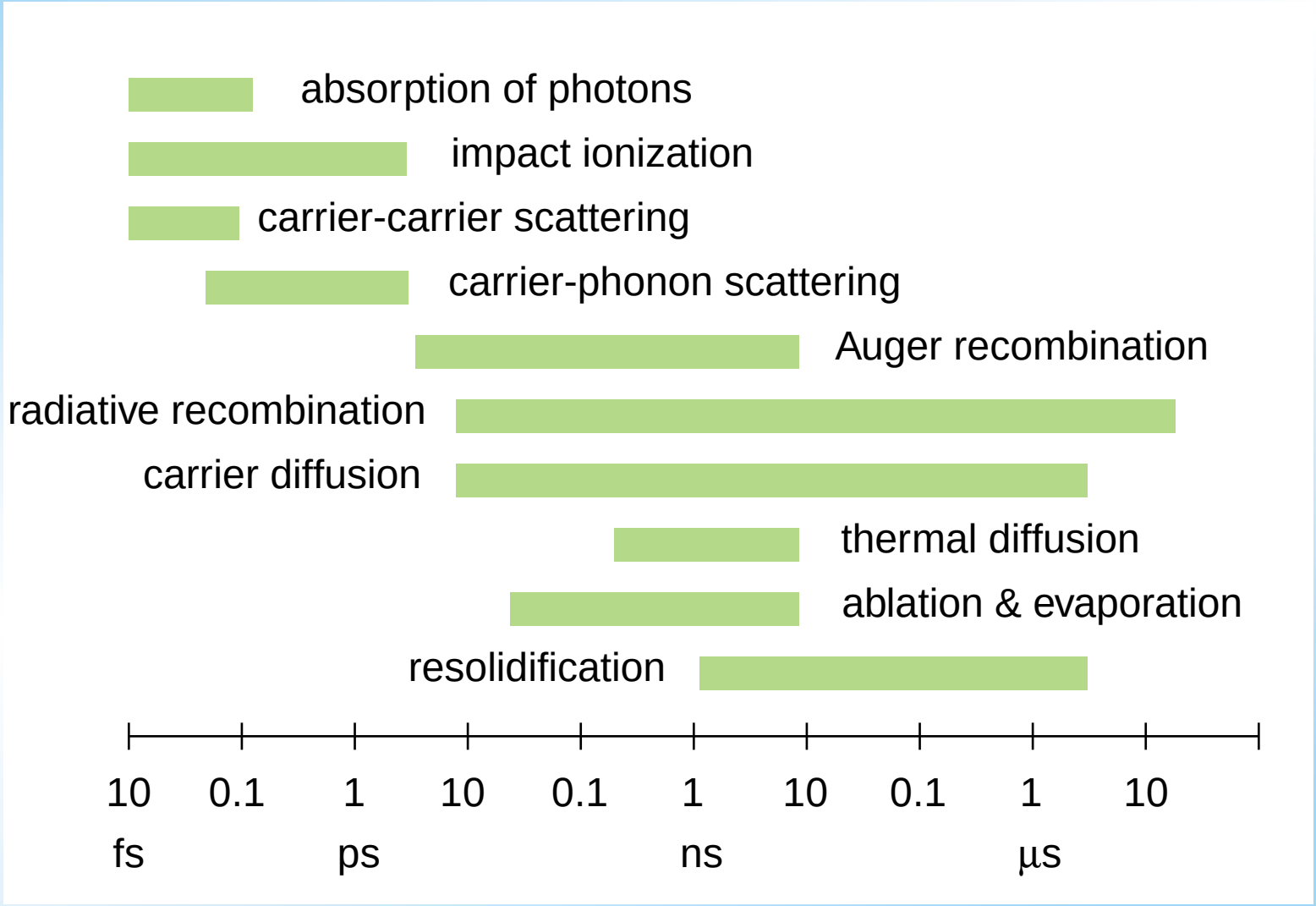

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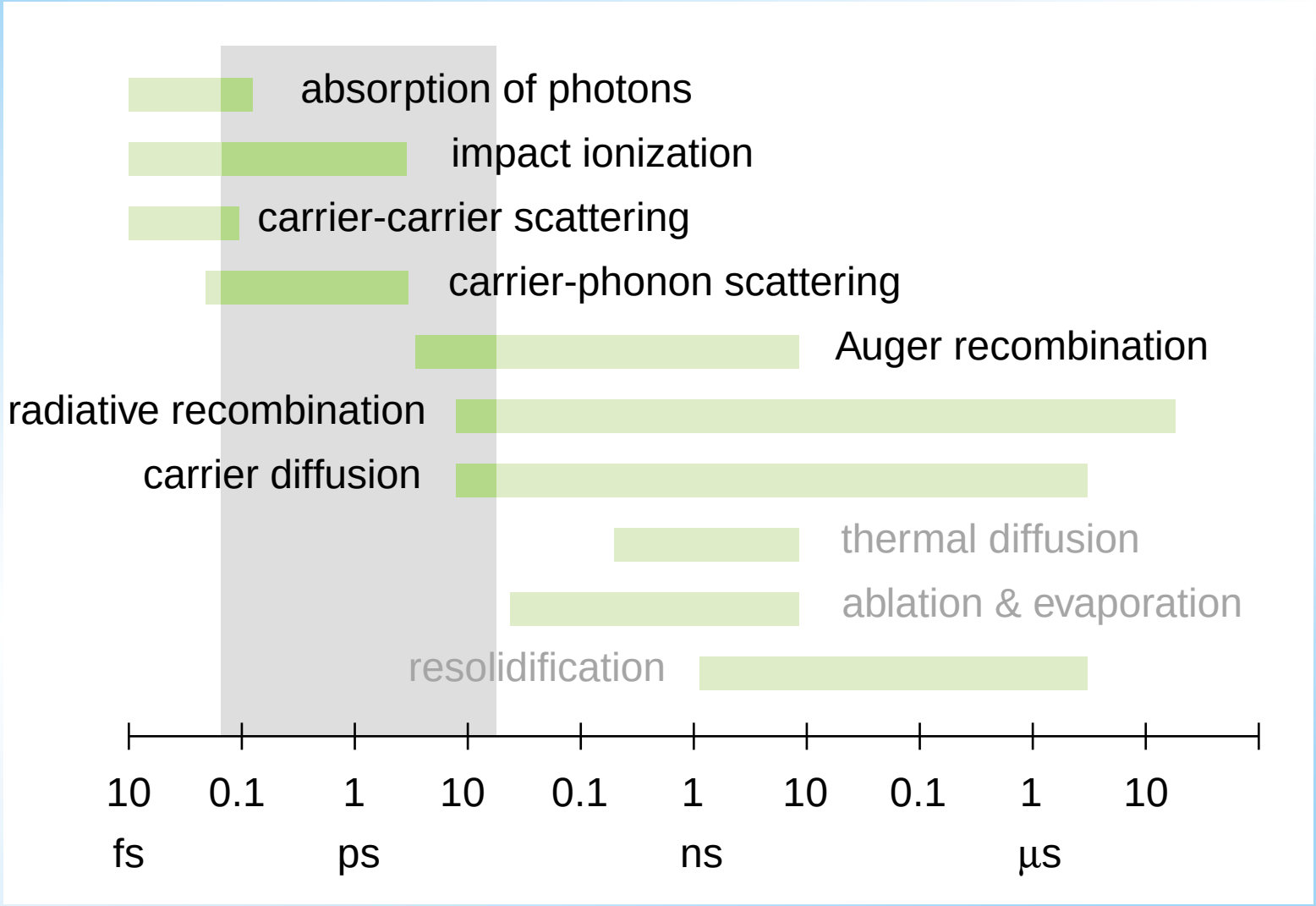

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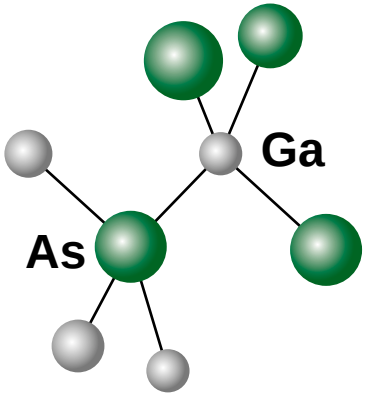
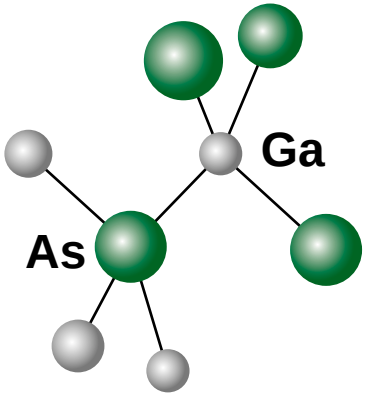
structure

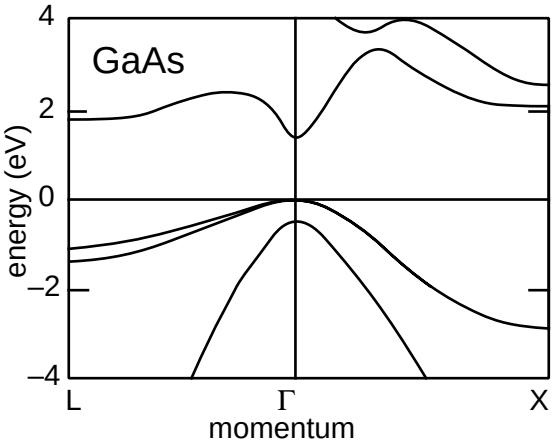


Introduction

structure



bandstructure



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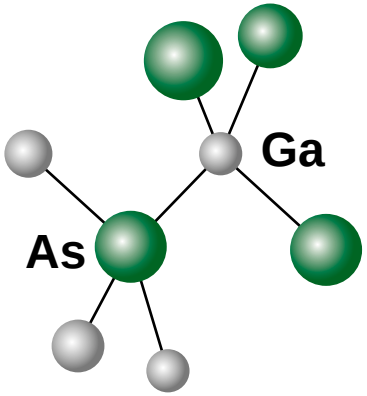
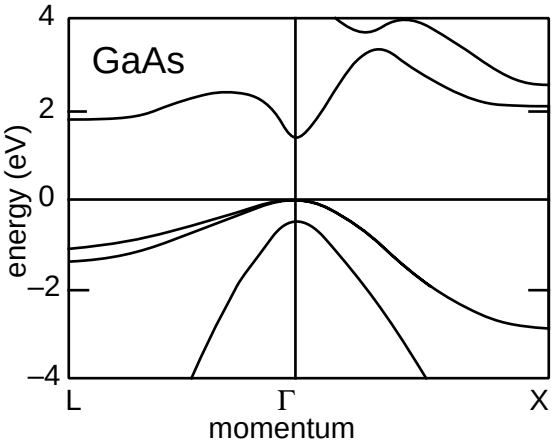
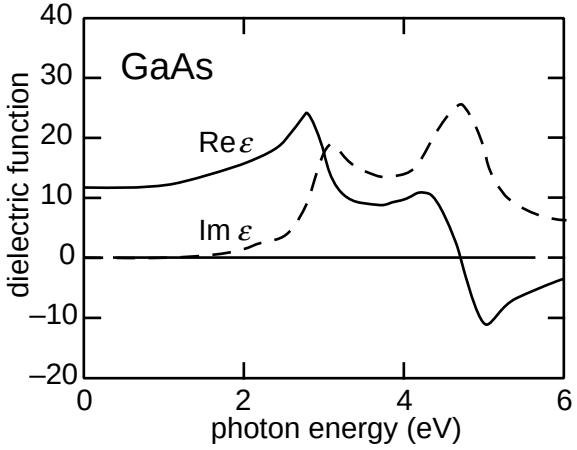
structure



bandstructure



dielectric function



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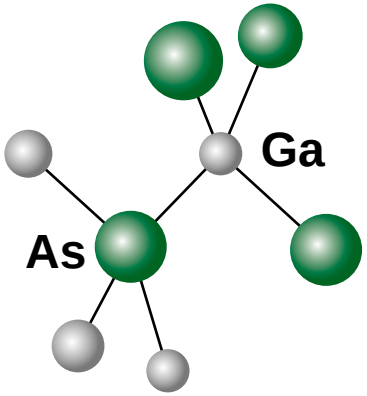
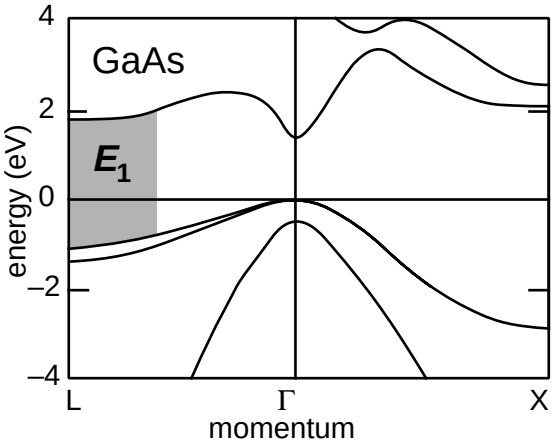
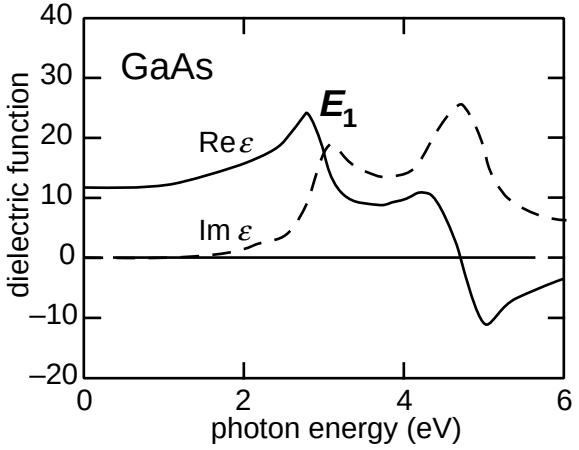
structure



bandstructure



dielectric function



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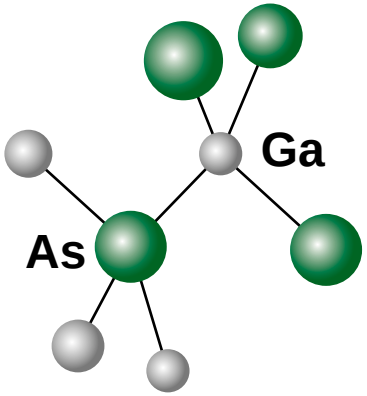
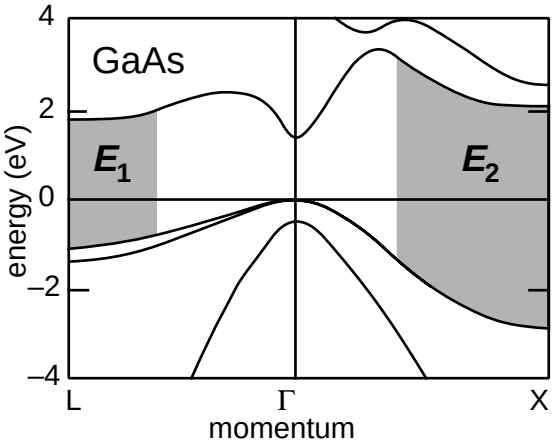
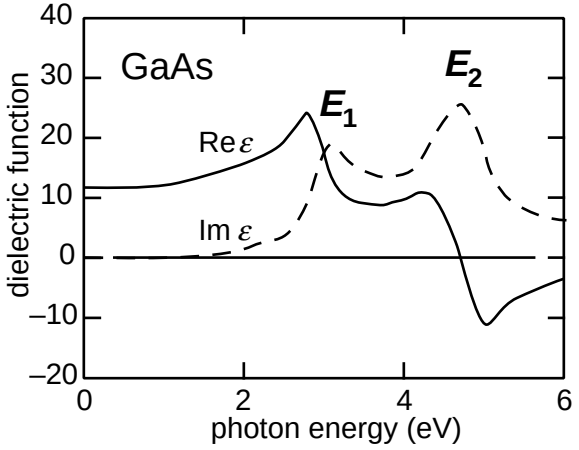
structure



bandstructure

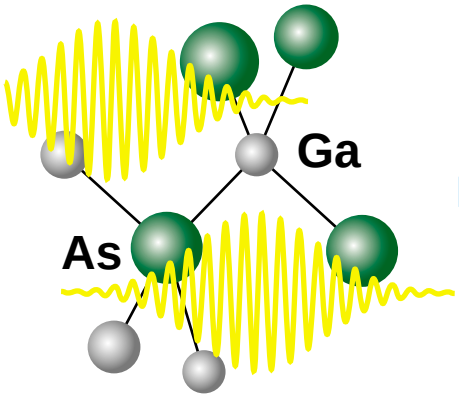


dielectric function



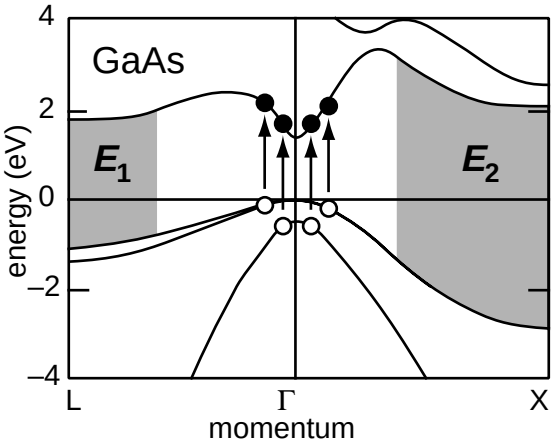
Introduction

structure



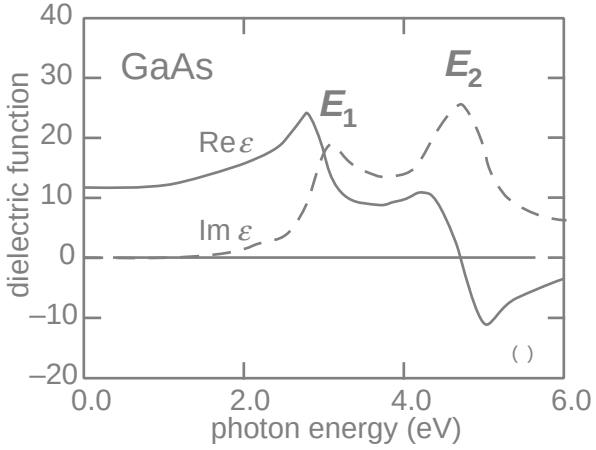
70 fs, 10^{16} W/m^2

bandstructure



10^{20} – 10^{22} cm^{-3}

dielectric function



Introduction

structure

?

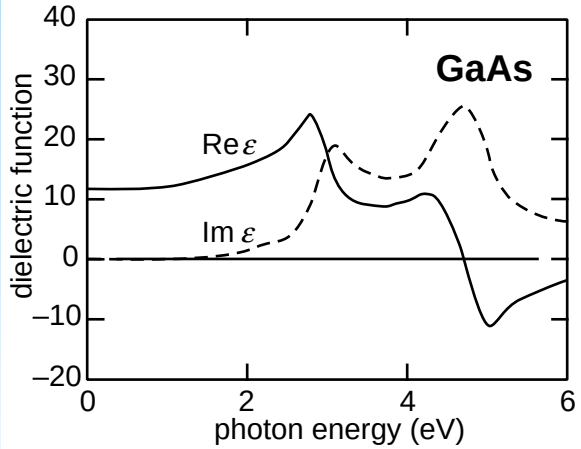
bandstructure

?

dielectric function

?

Introduction



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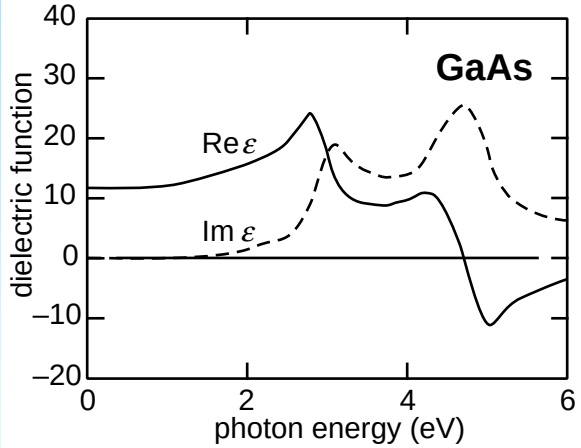

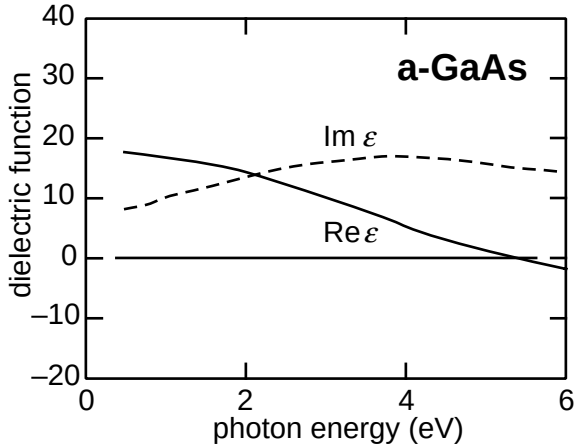

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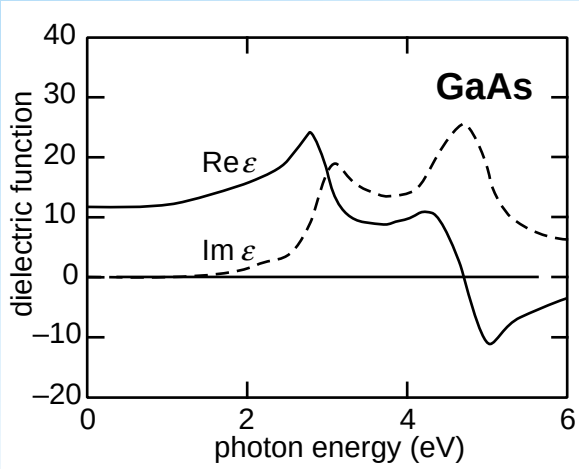

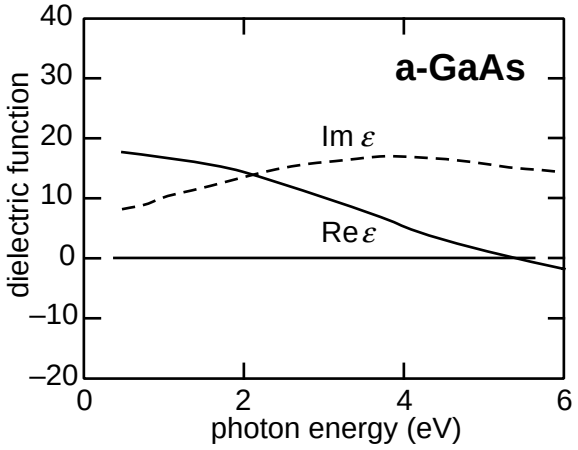

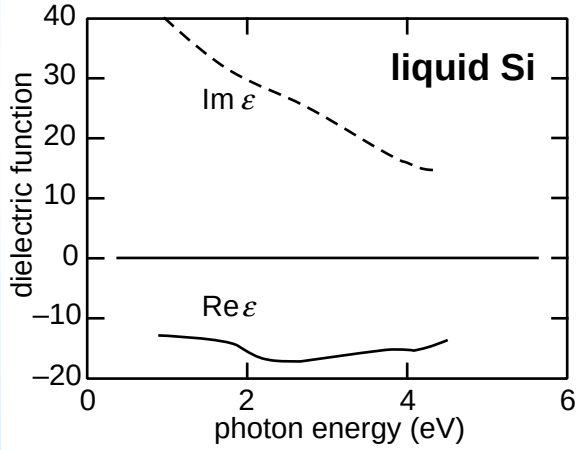

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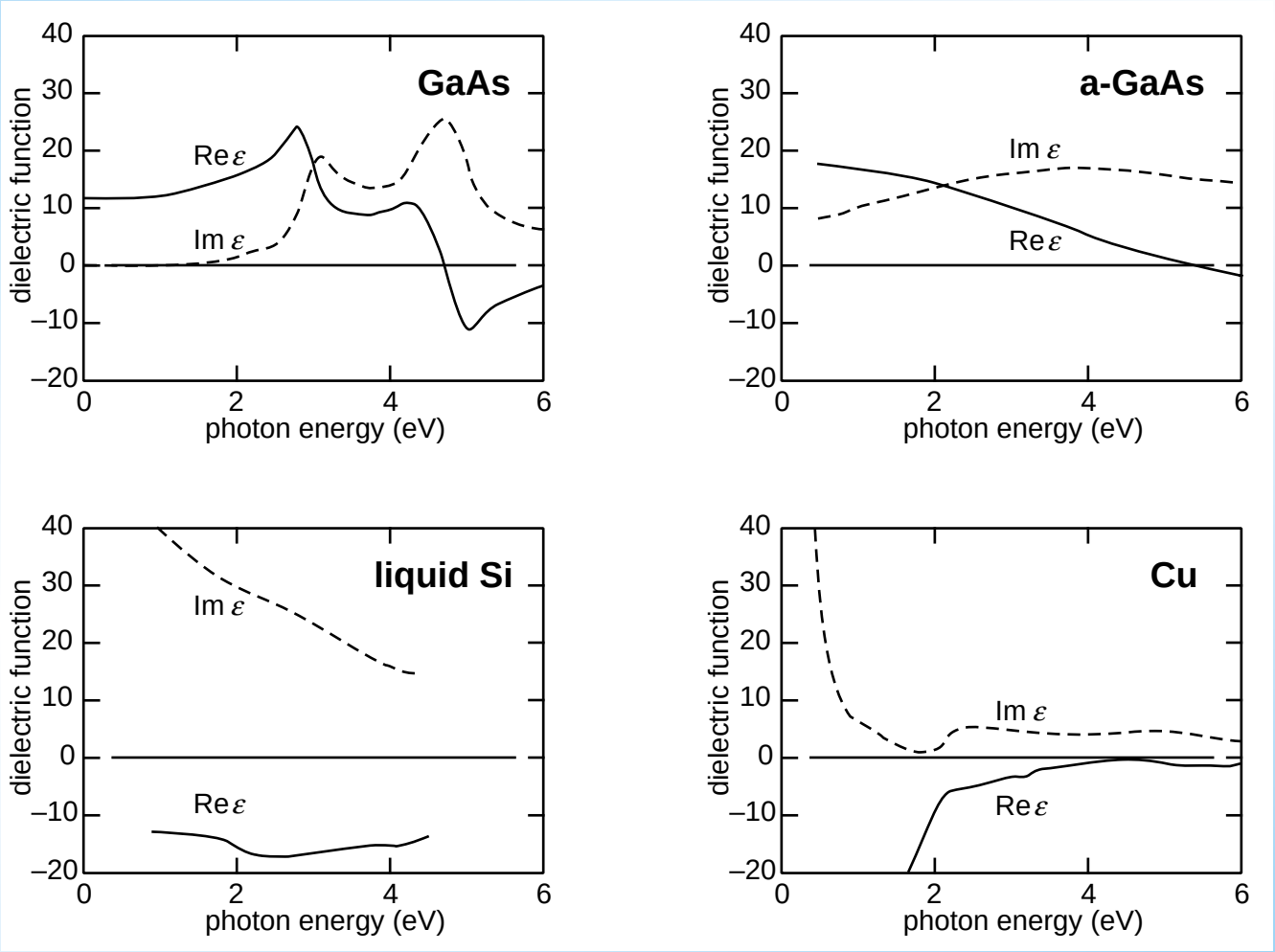
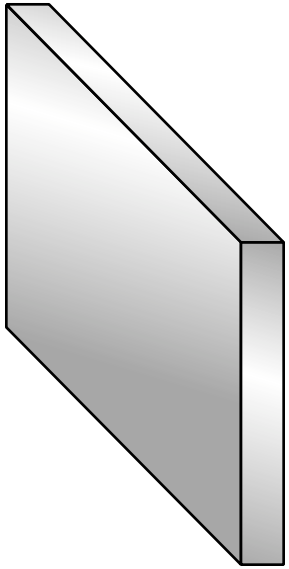


Outline

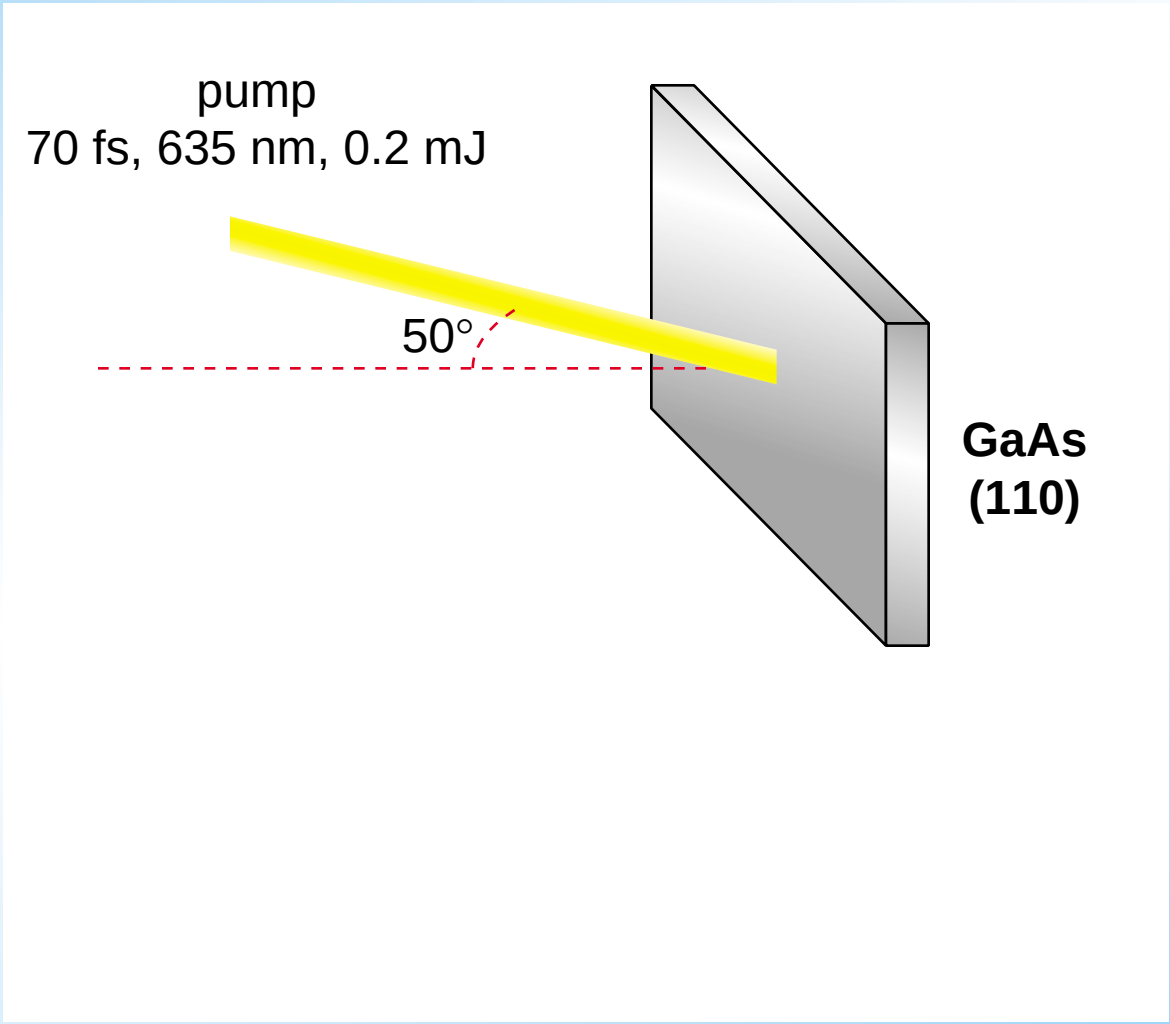
- **Method**
- **Results**
- **Discussion**

Method

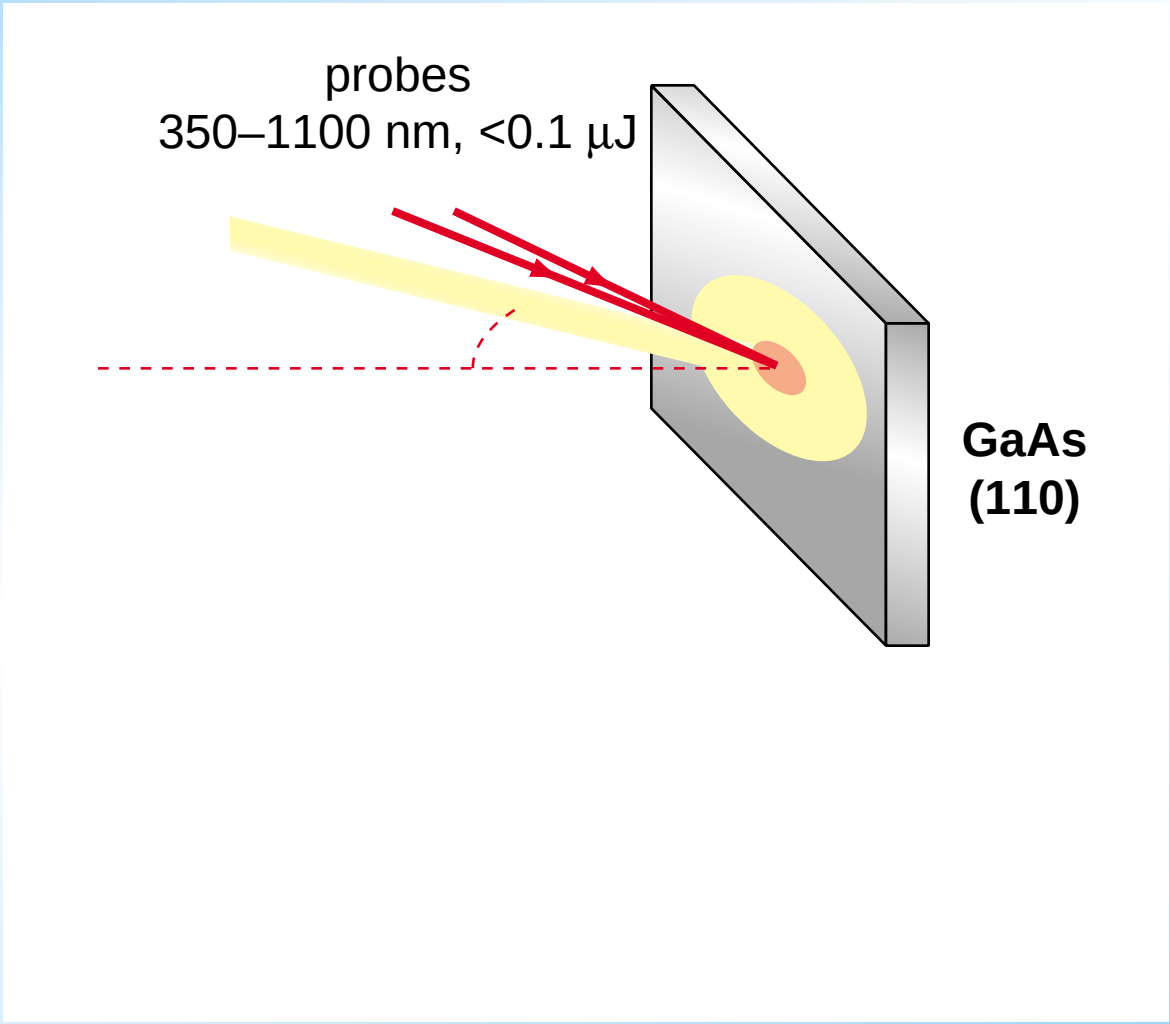


GaAs
(110)

Method



Method



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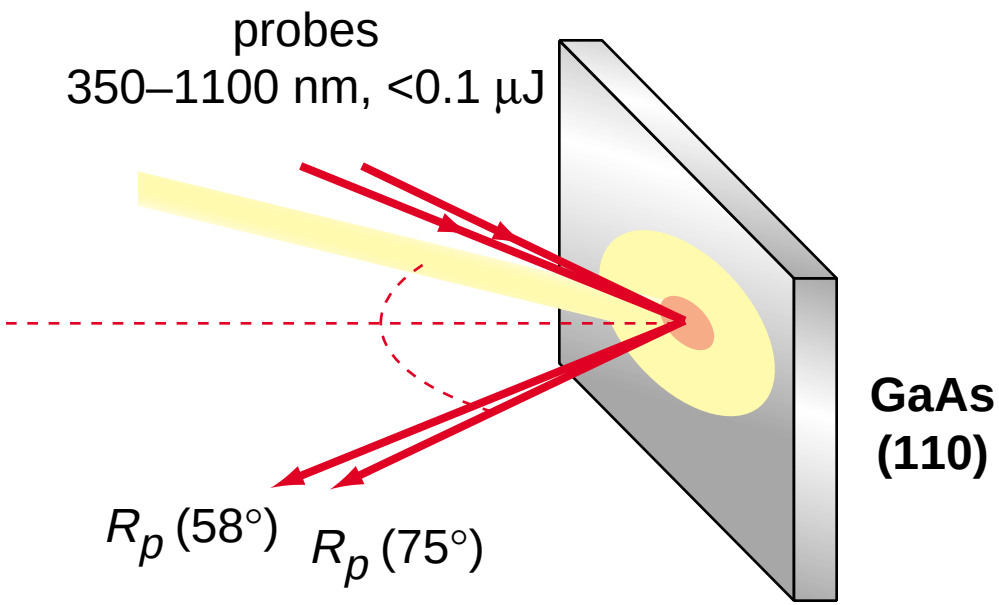

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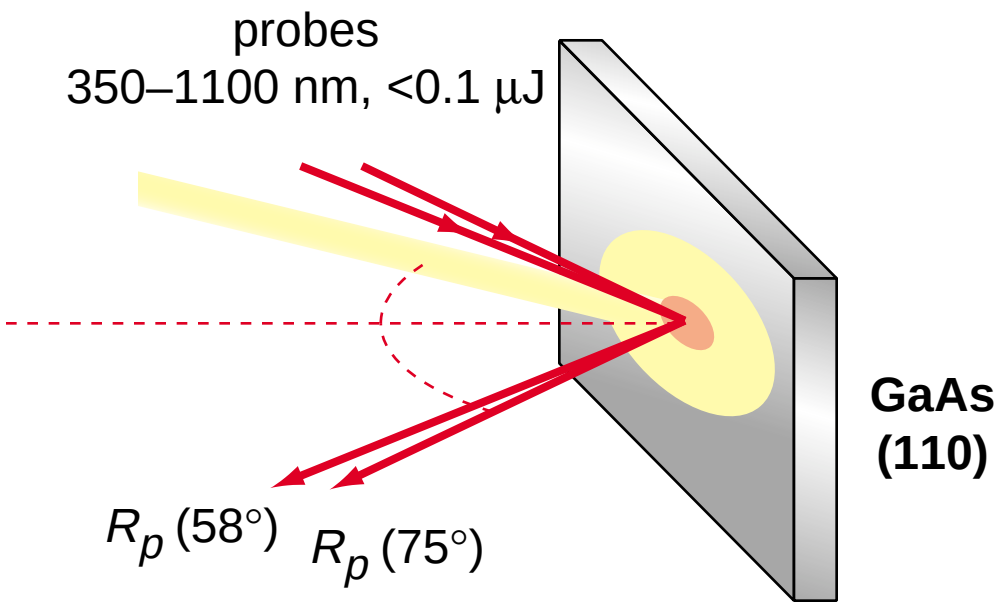
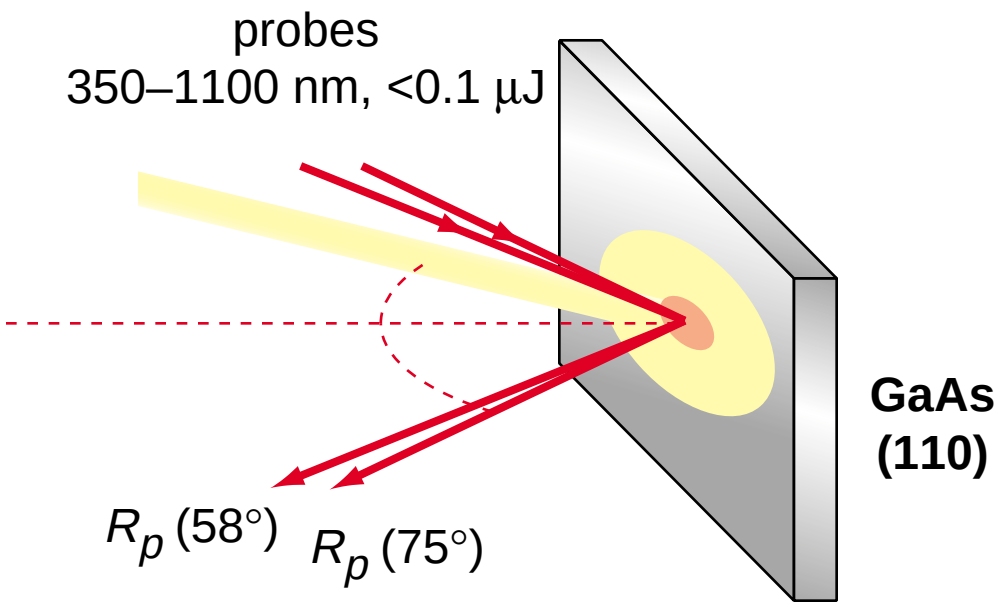


Fresnel equations

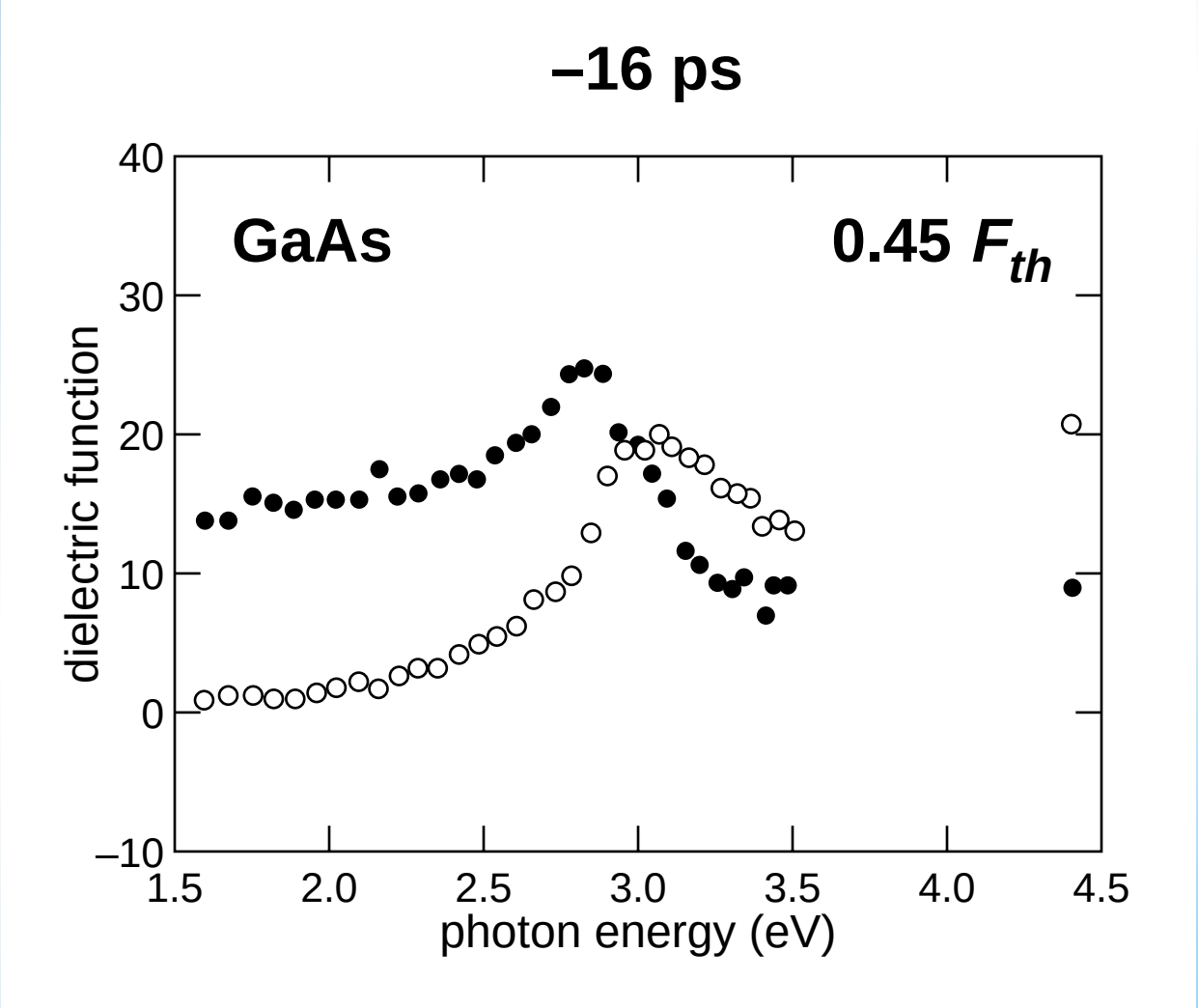
Method



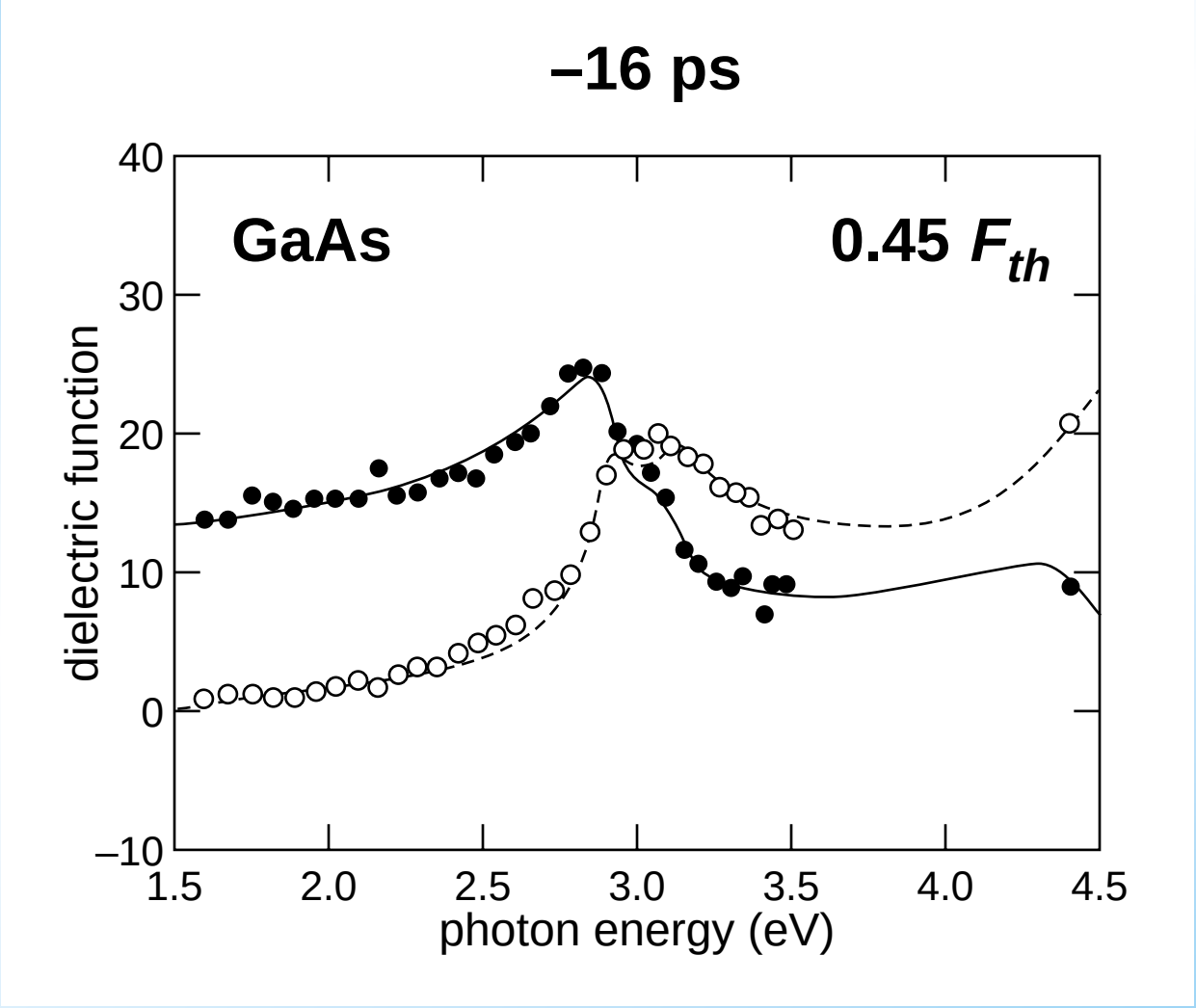
Fresnel equations → Re ε(ω), Im ε(ω)

Method

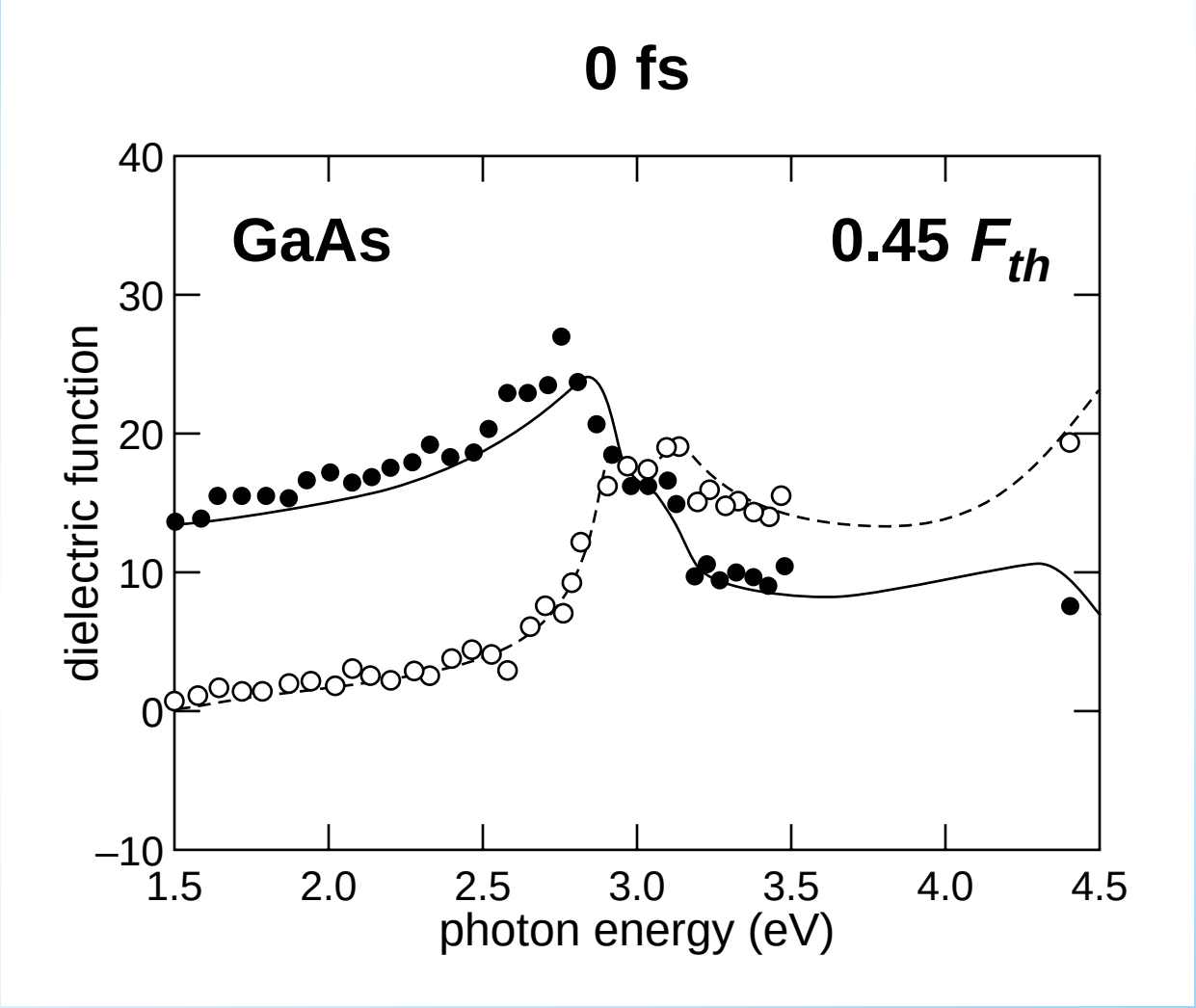
Results


–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

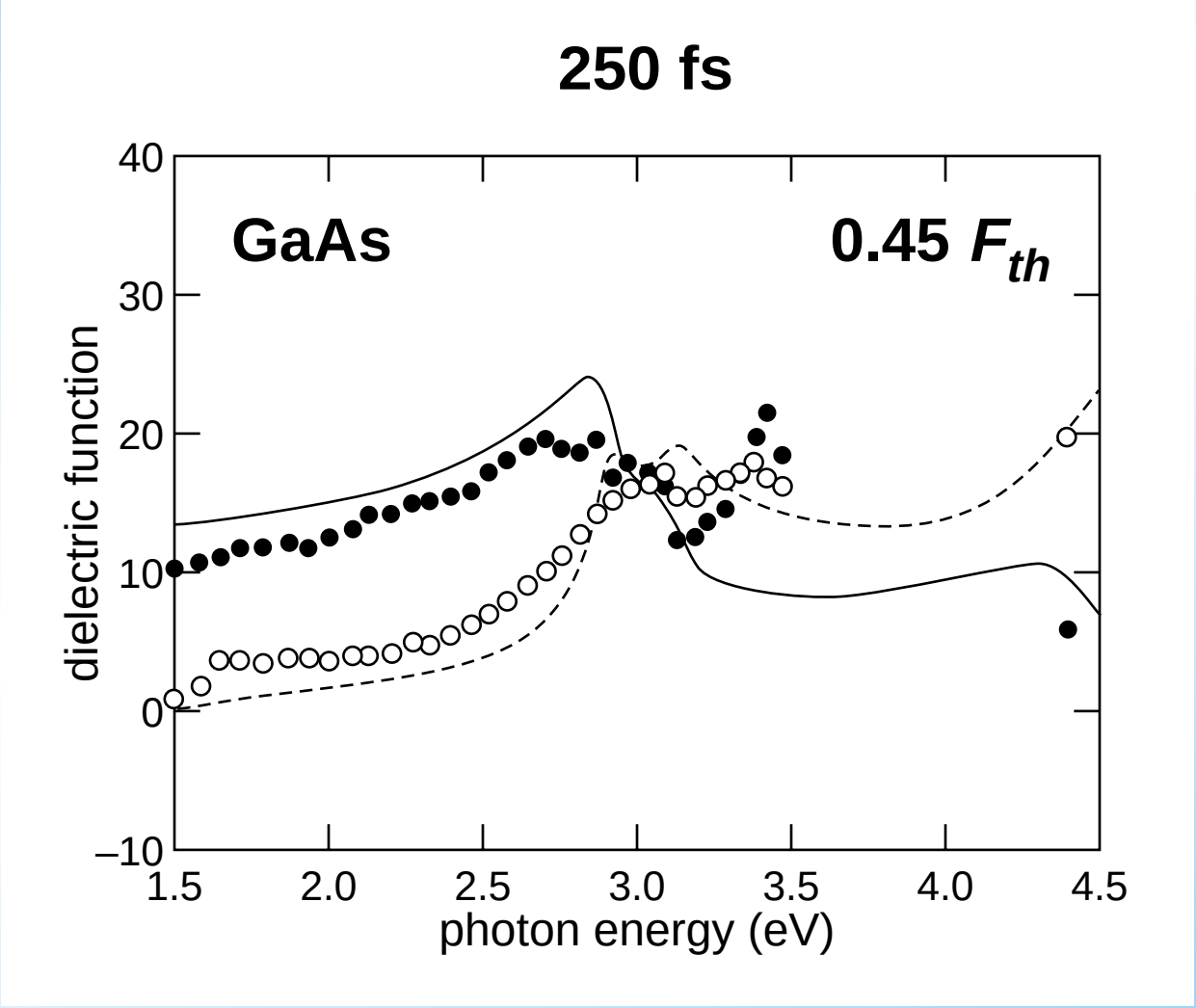
Results


–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

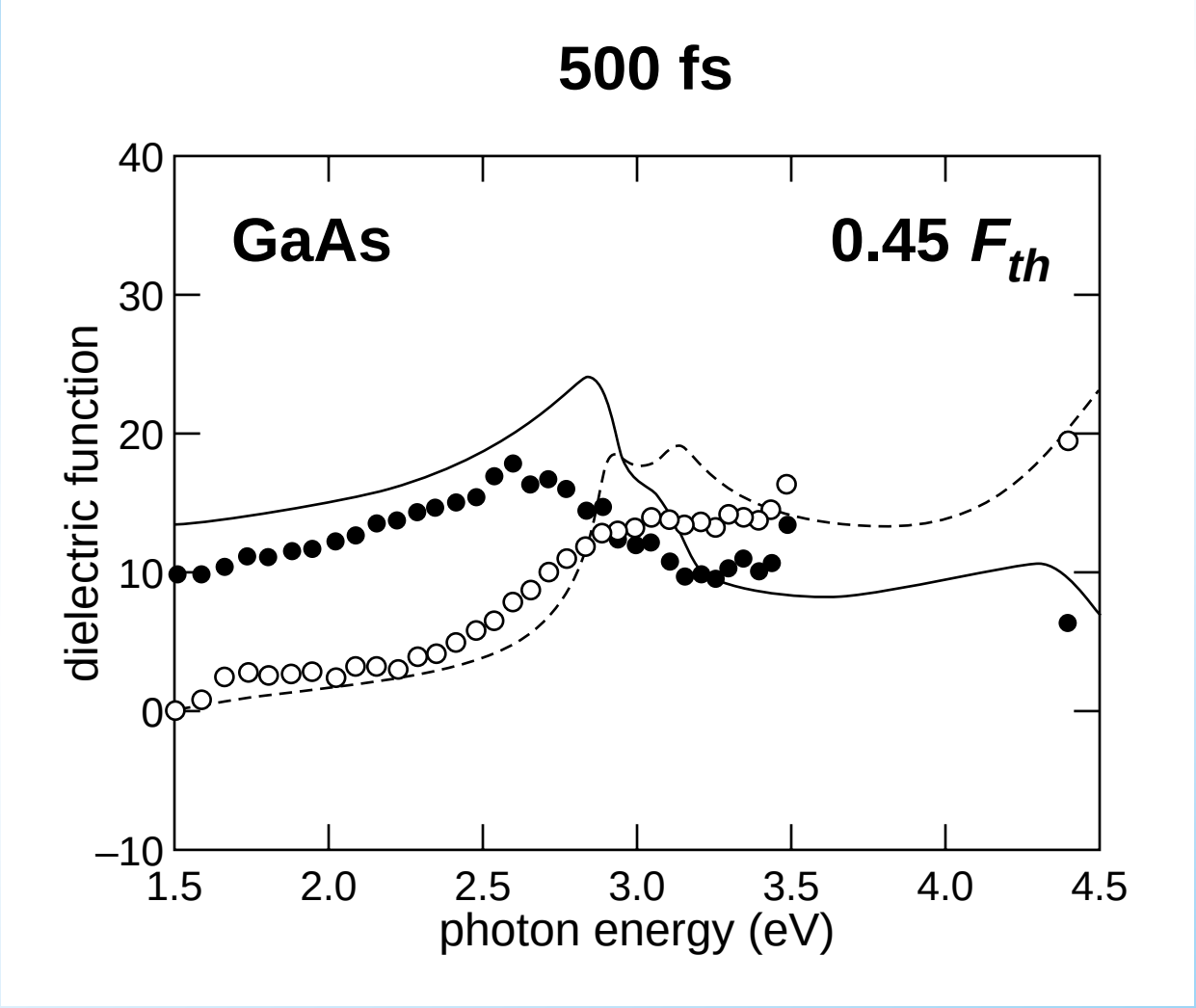
Results


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

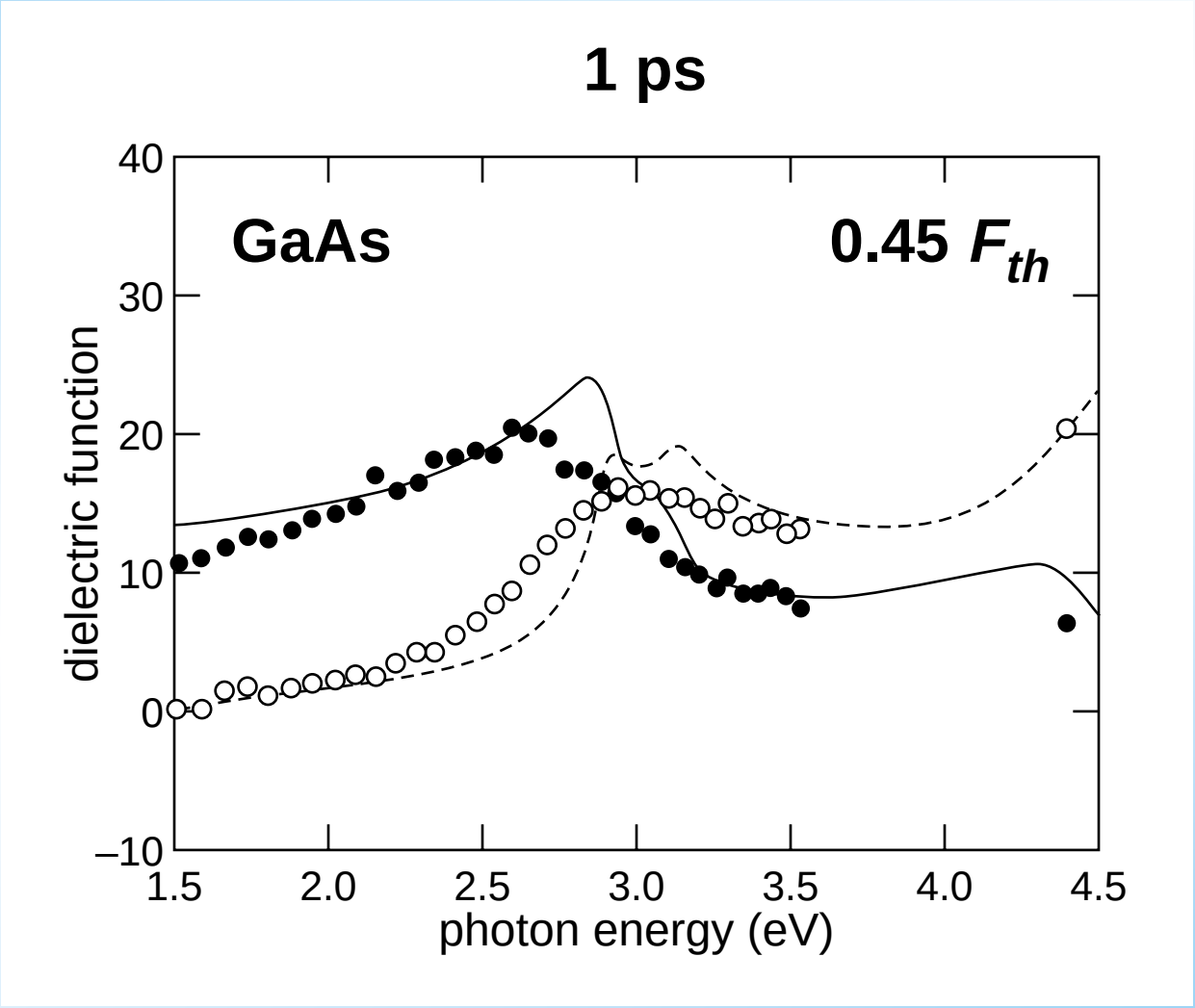
Results


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

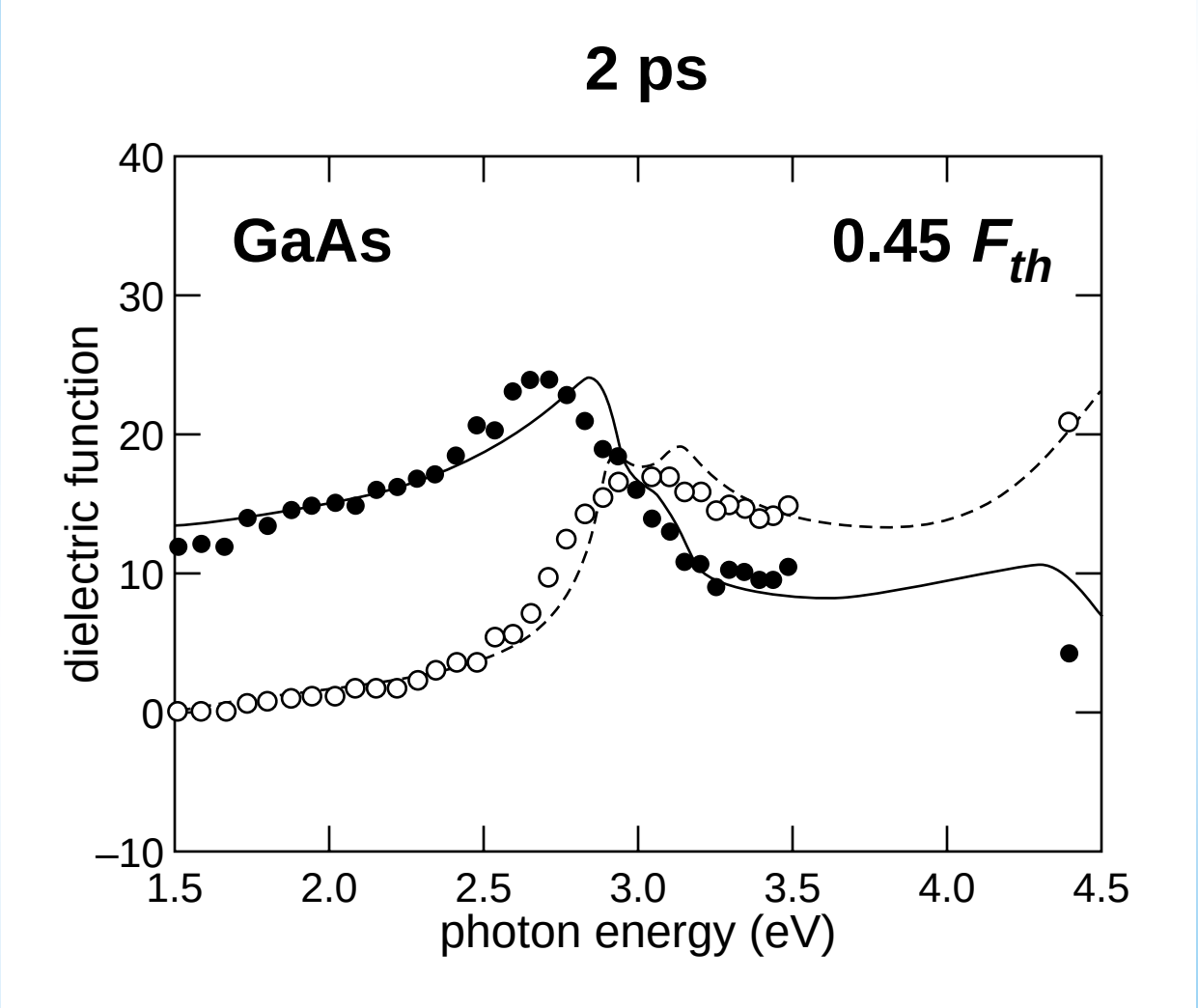
Results


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

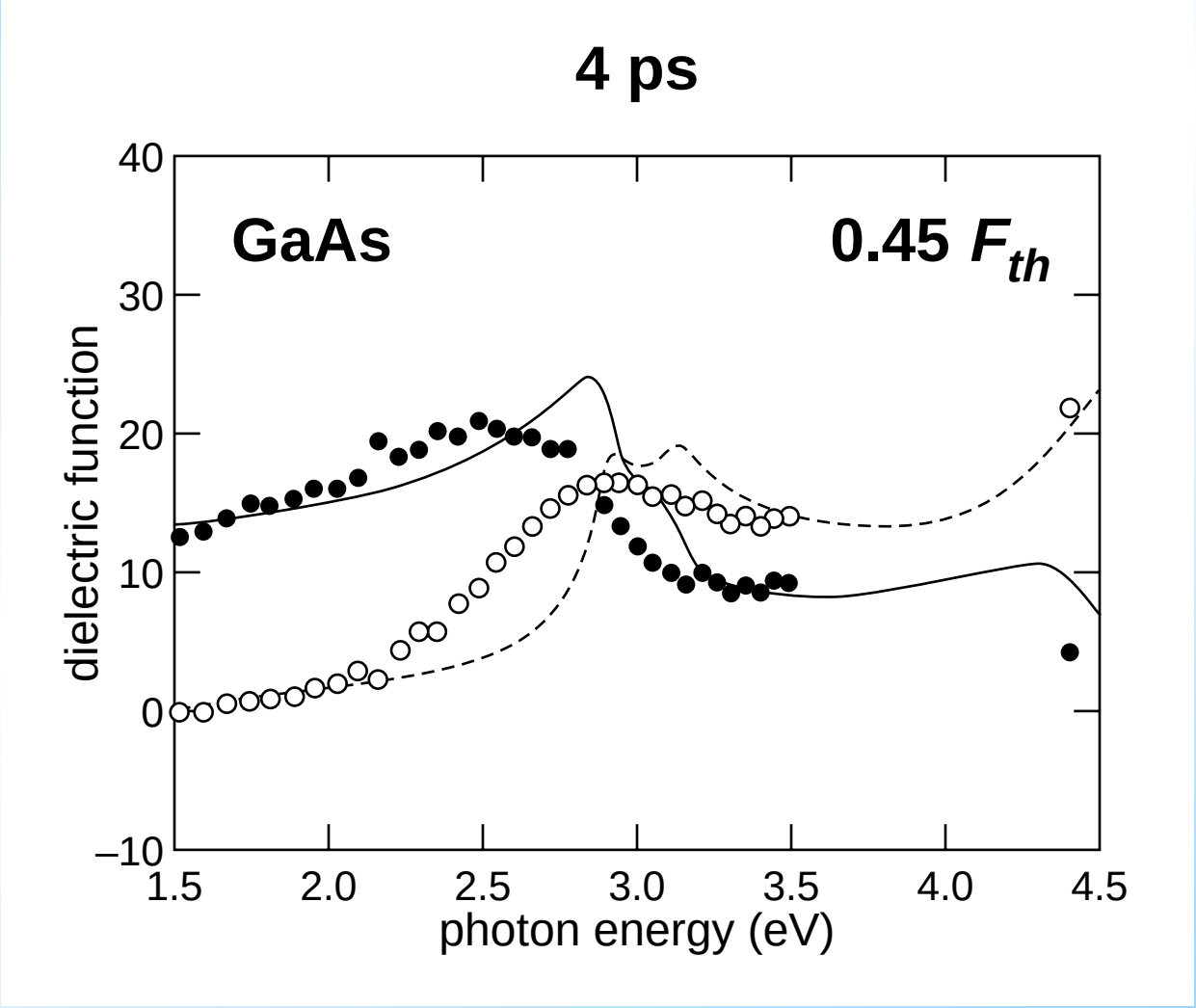
Results


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

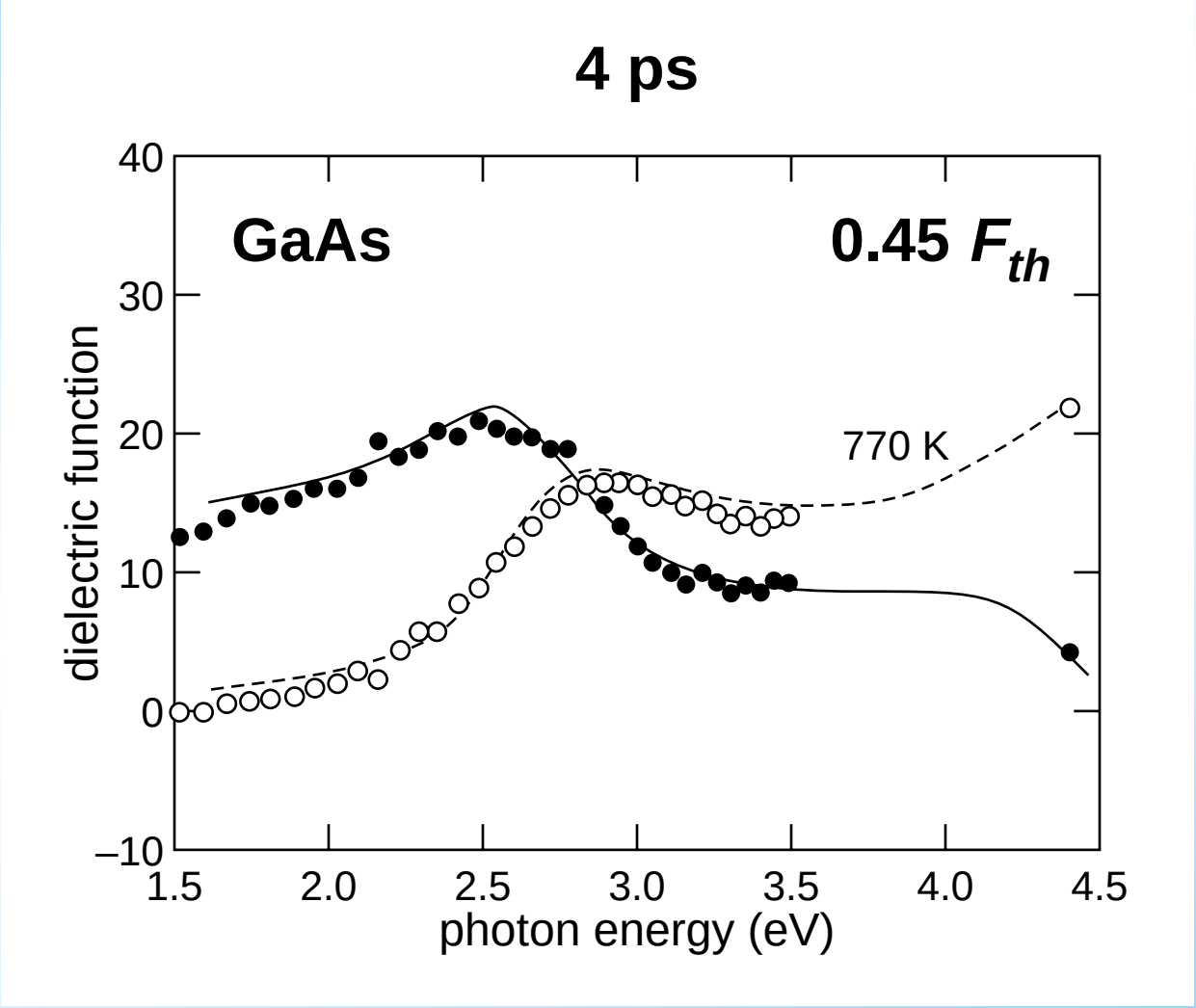
Results


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

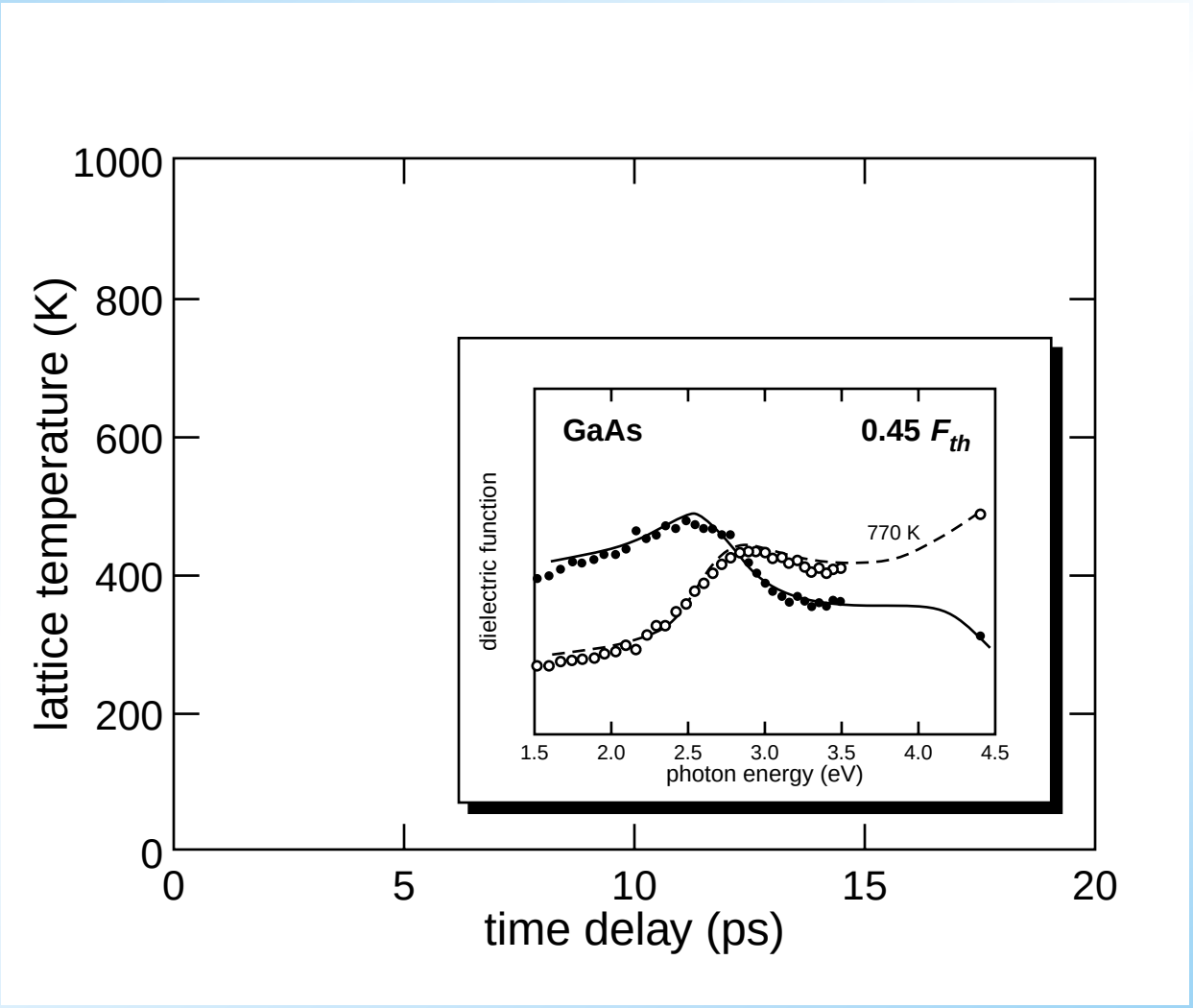
Results


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

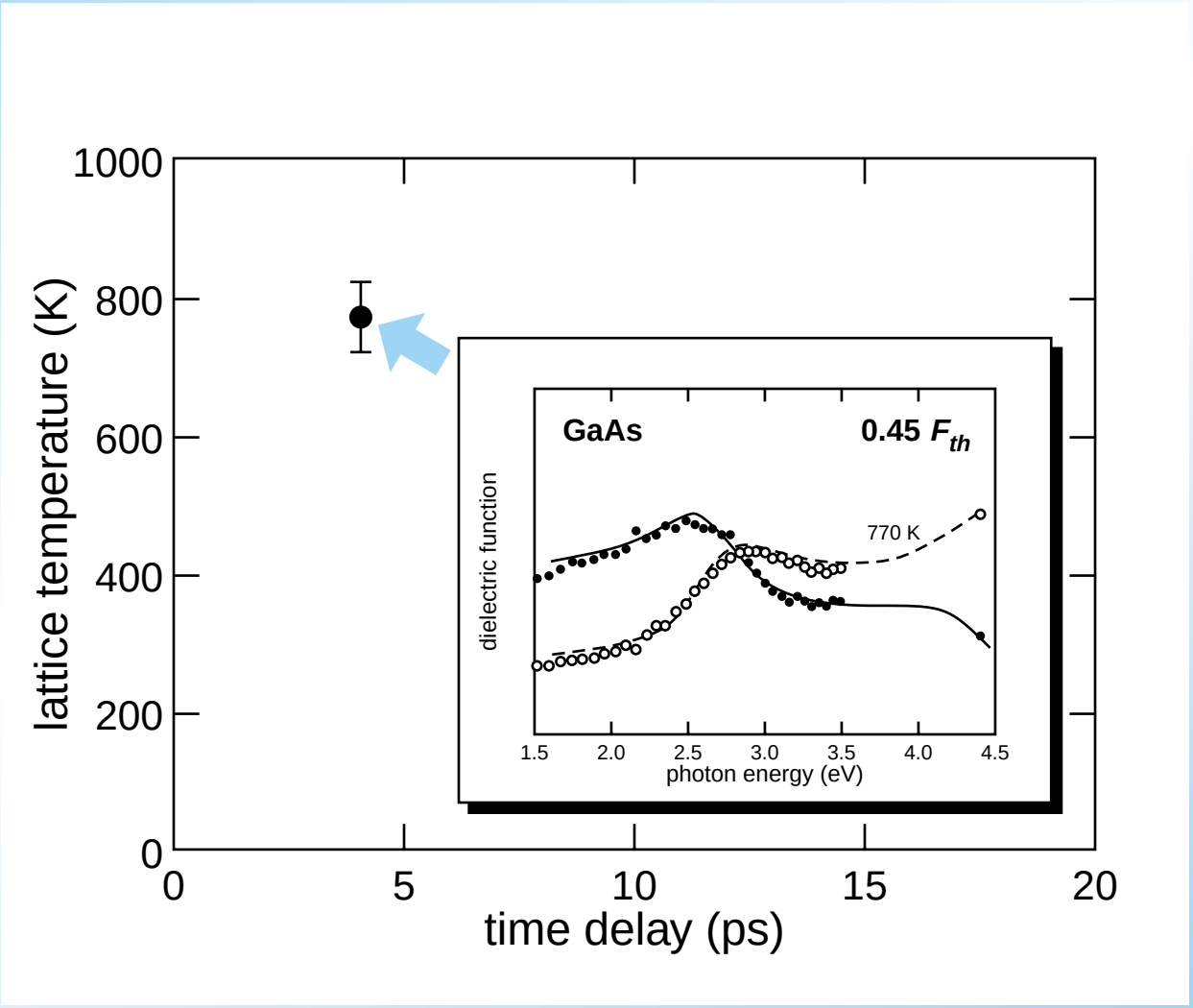
Results


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

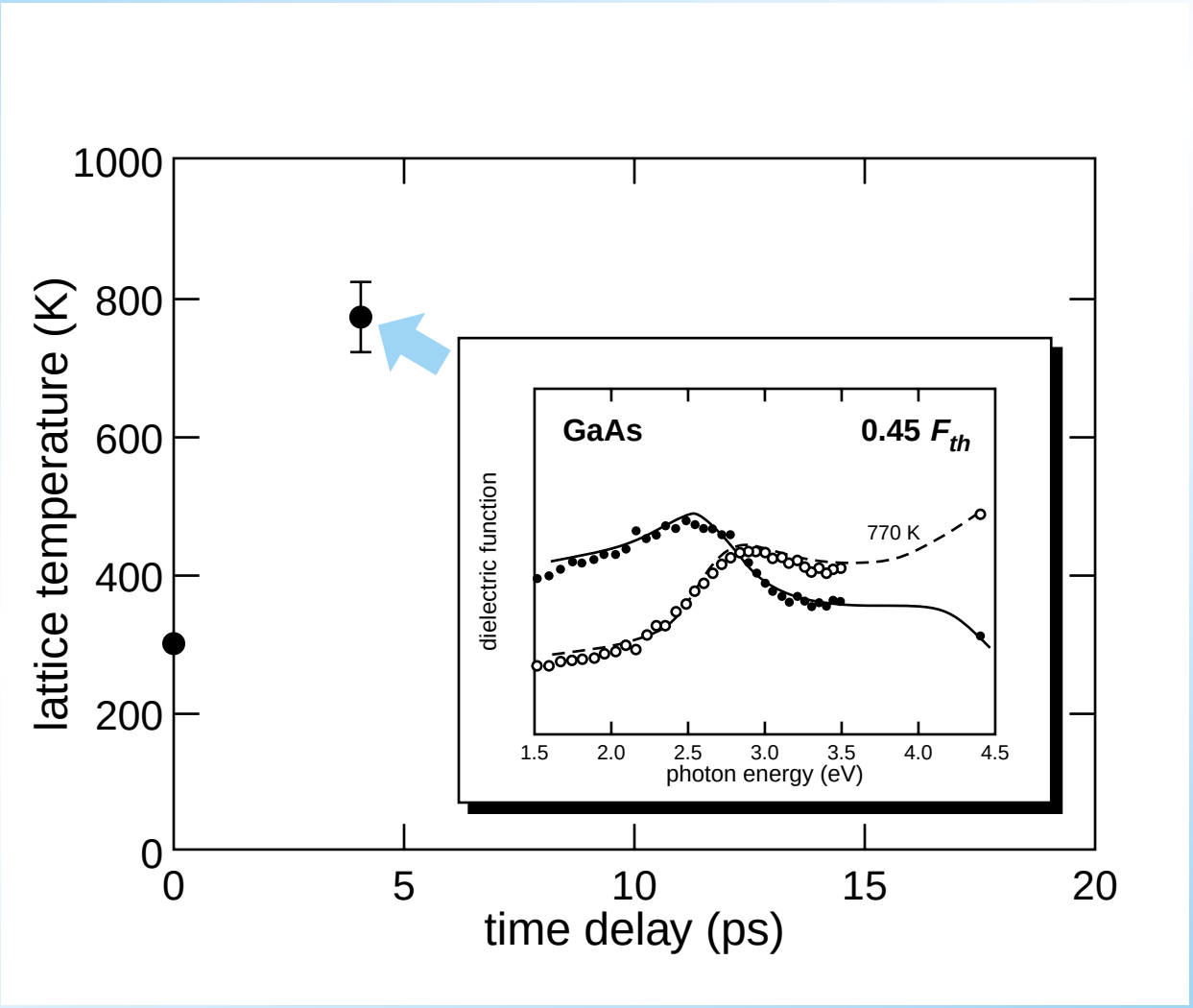
Results


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)

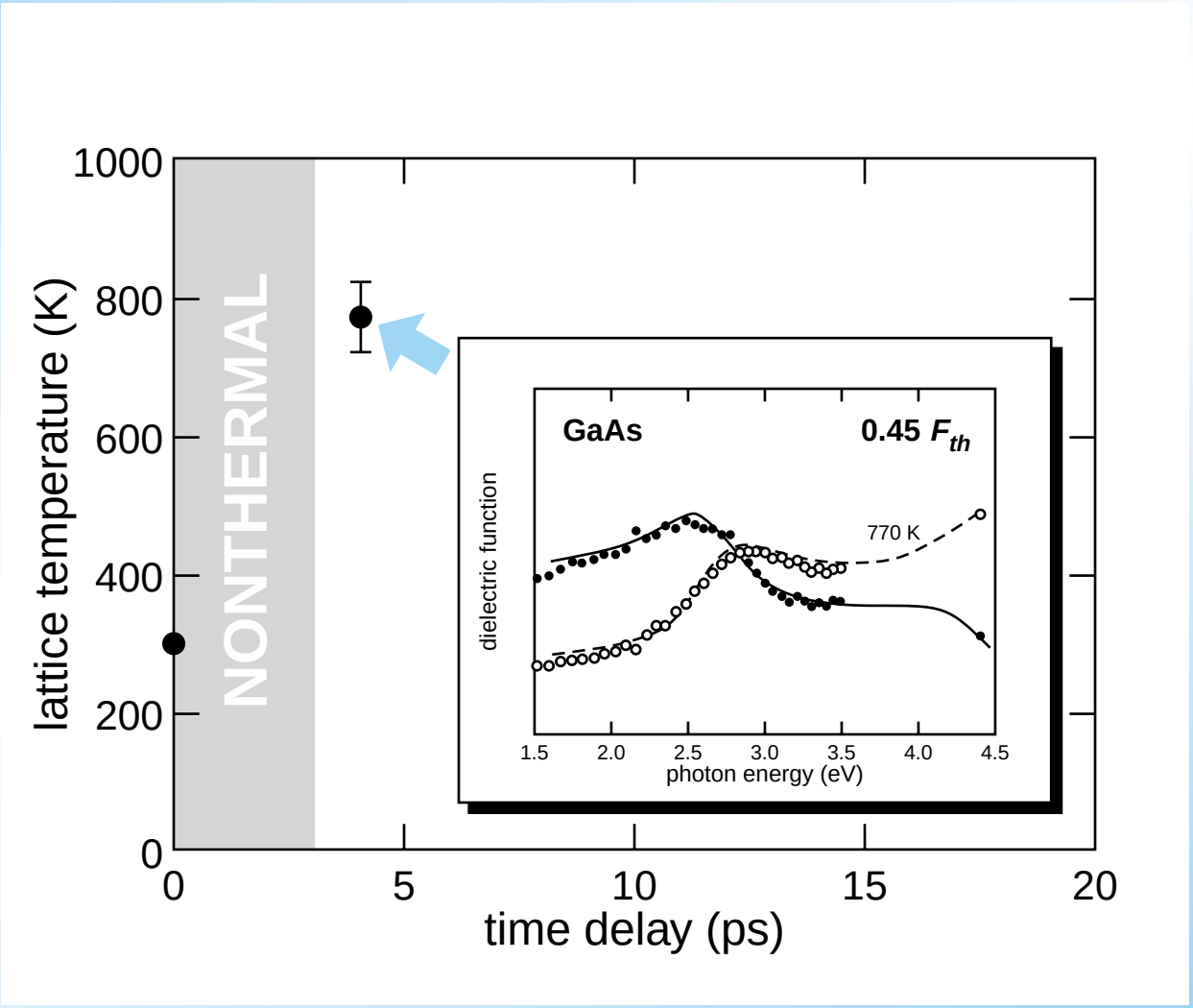
Results


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)

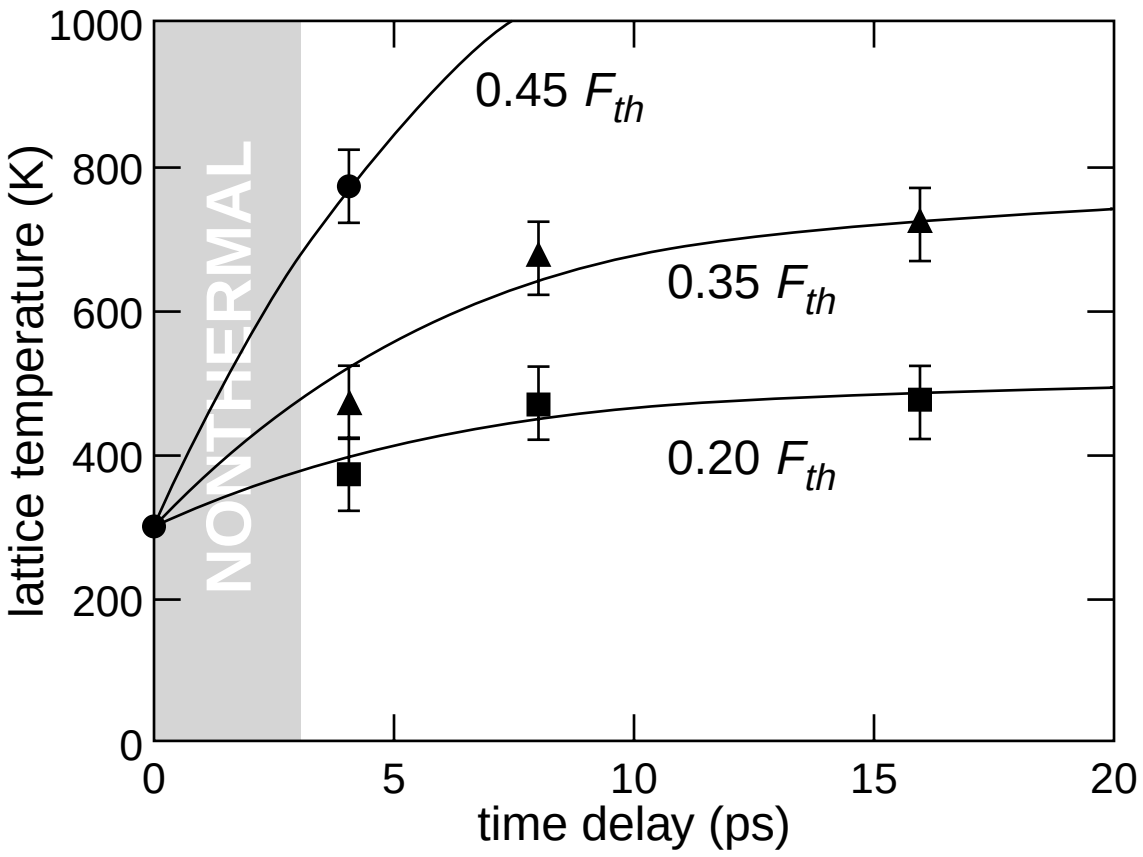
Results


1000
800
600
400
200
0
lattice temperature (K)
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)

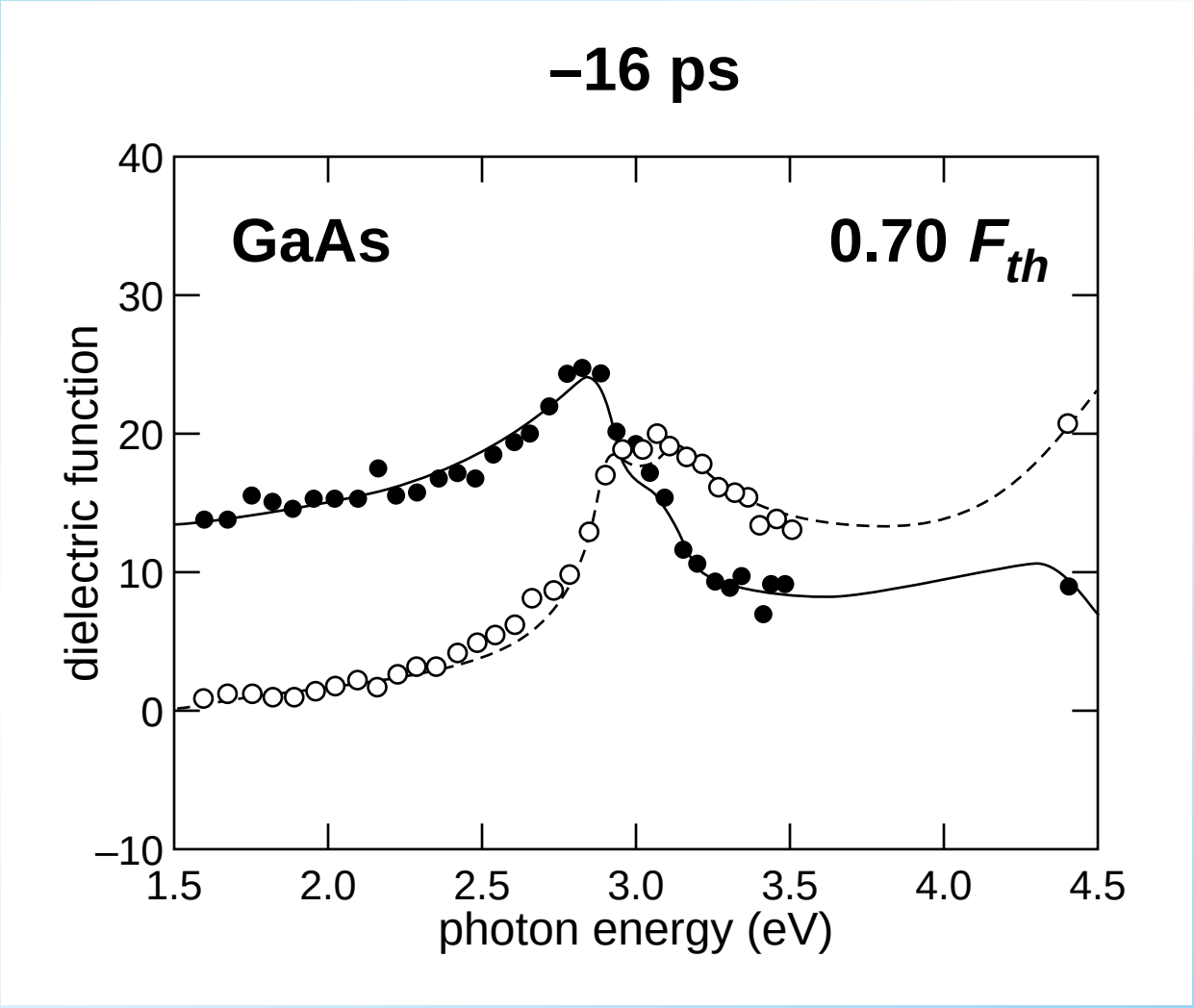
Results


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)

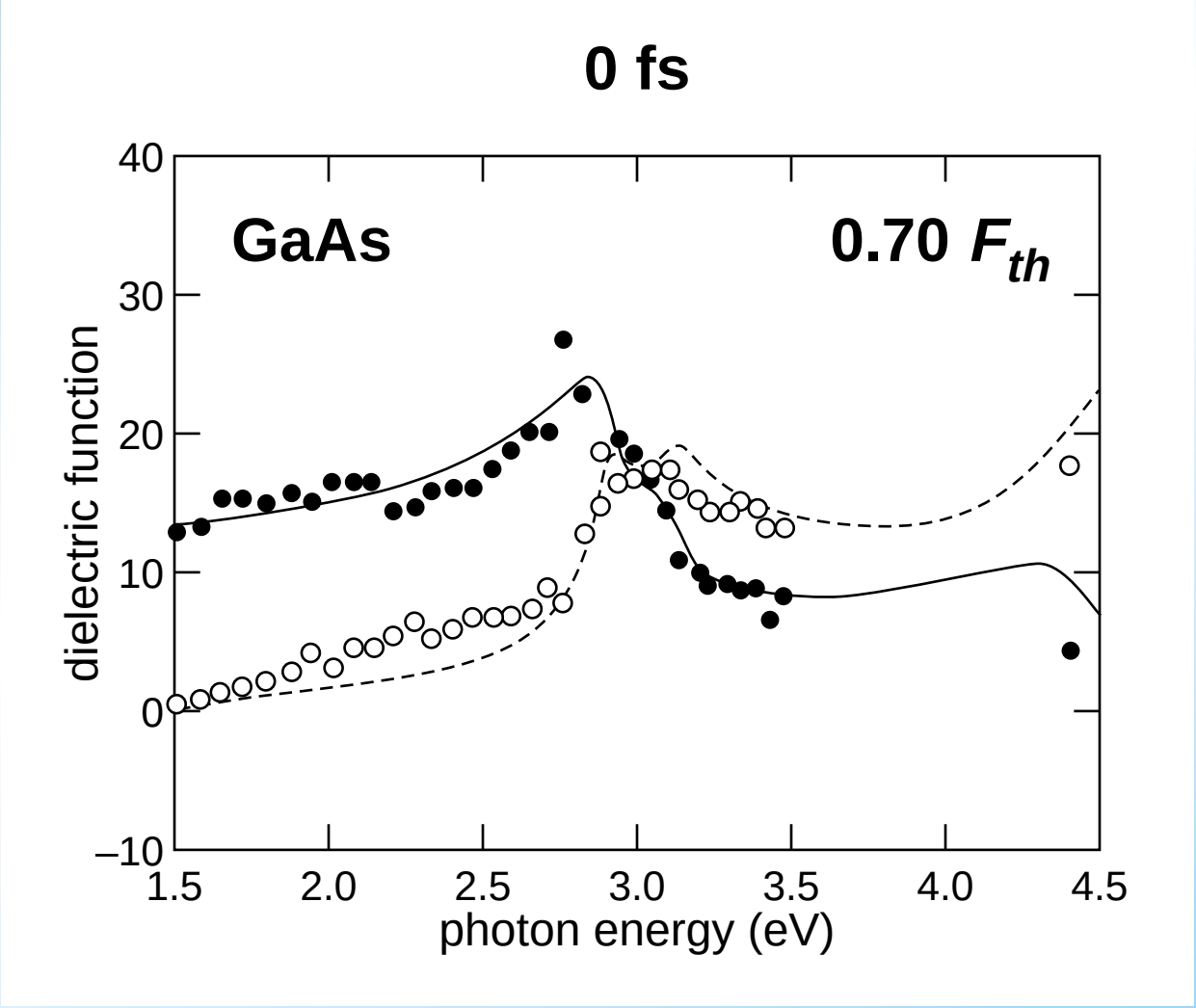
Results


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

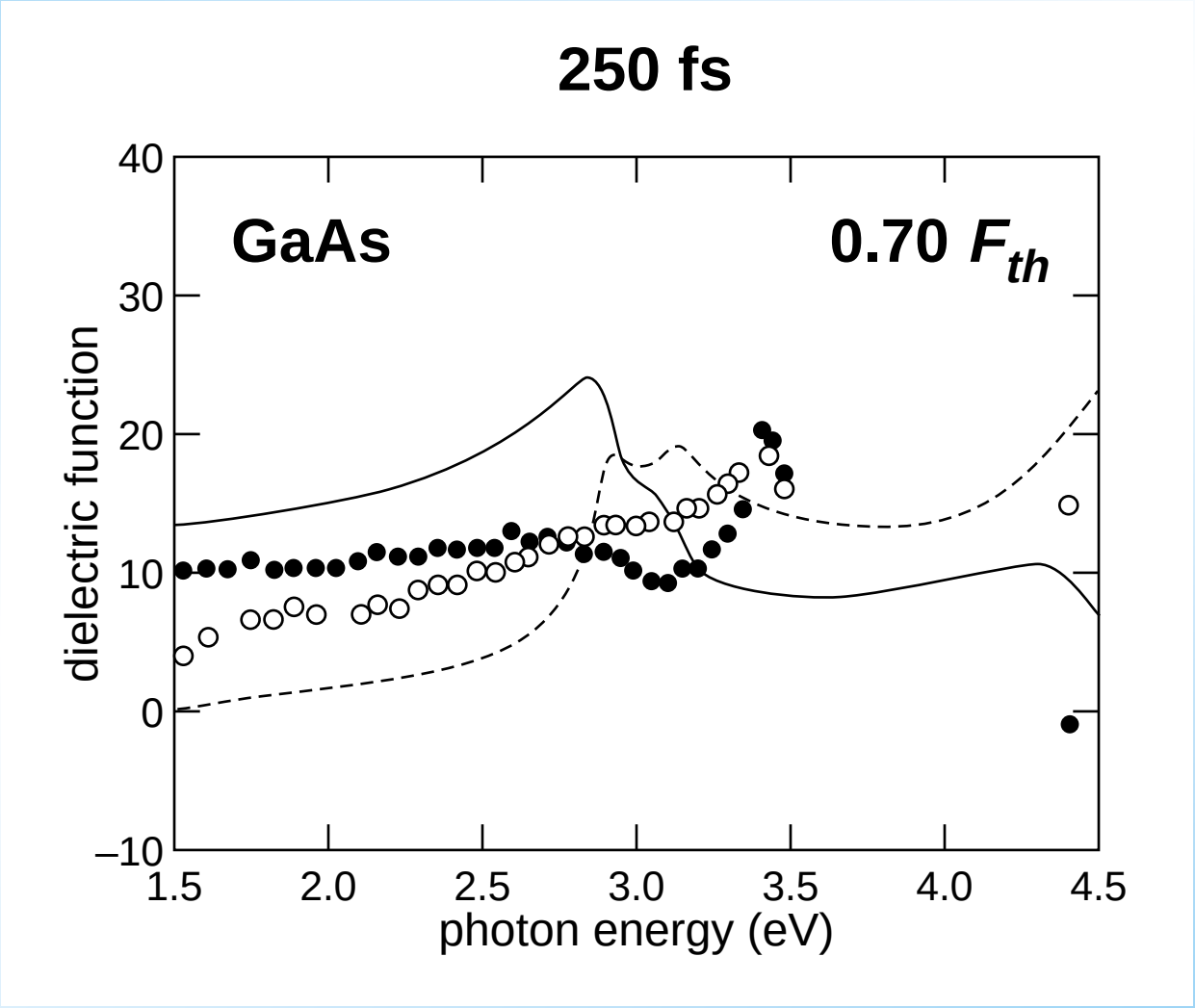
Results


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

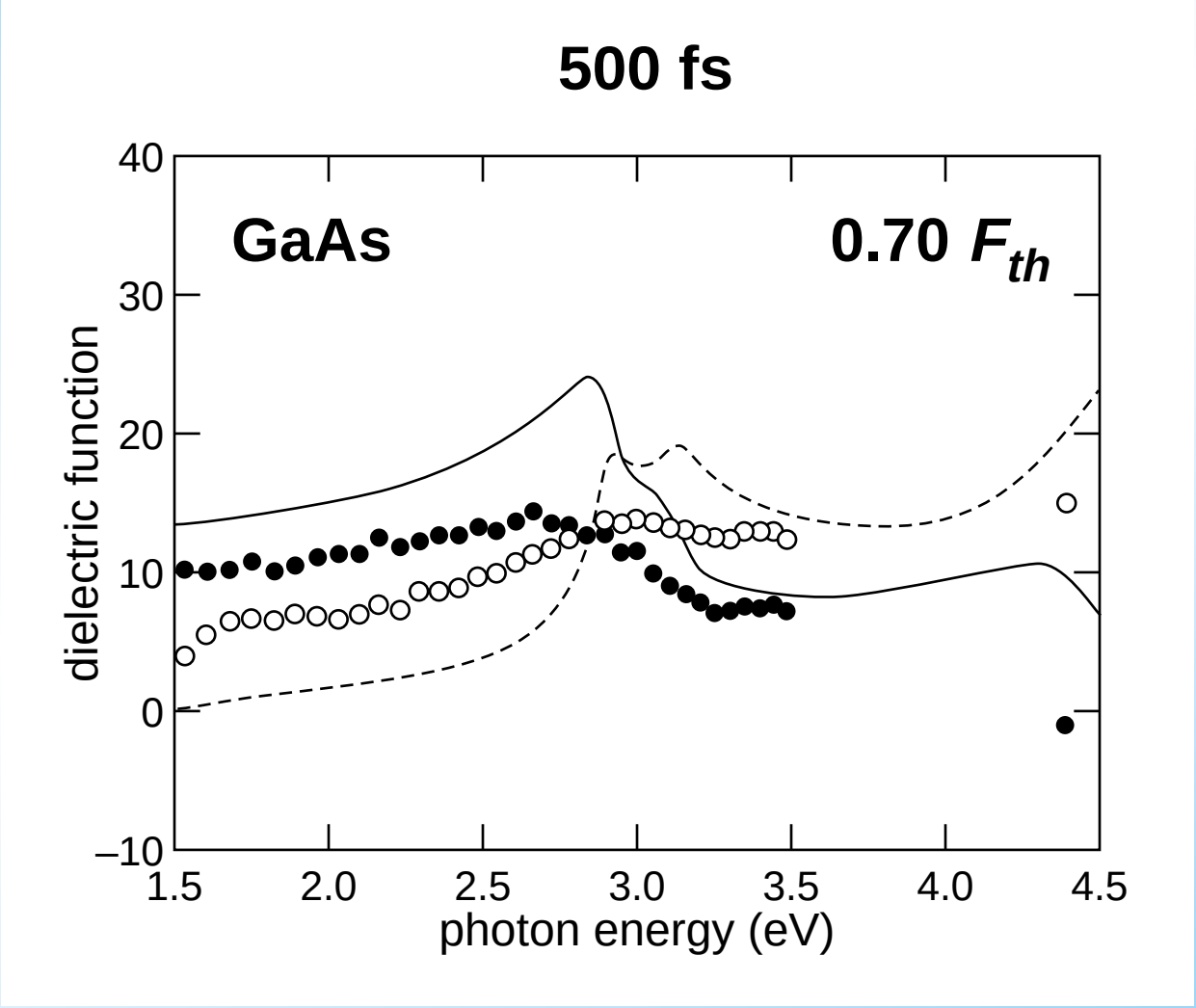
Results


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

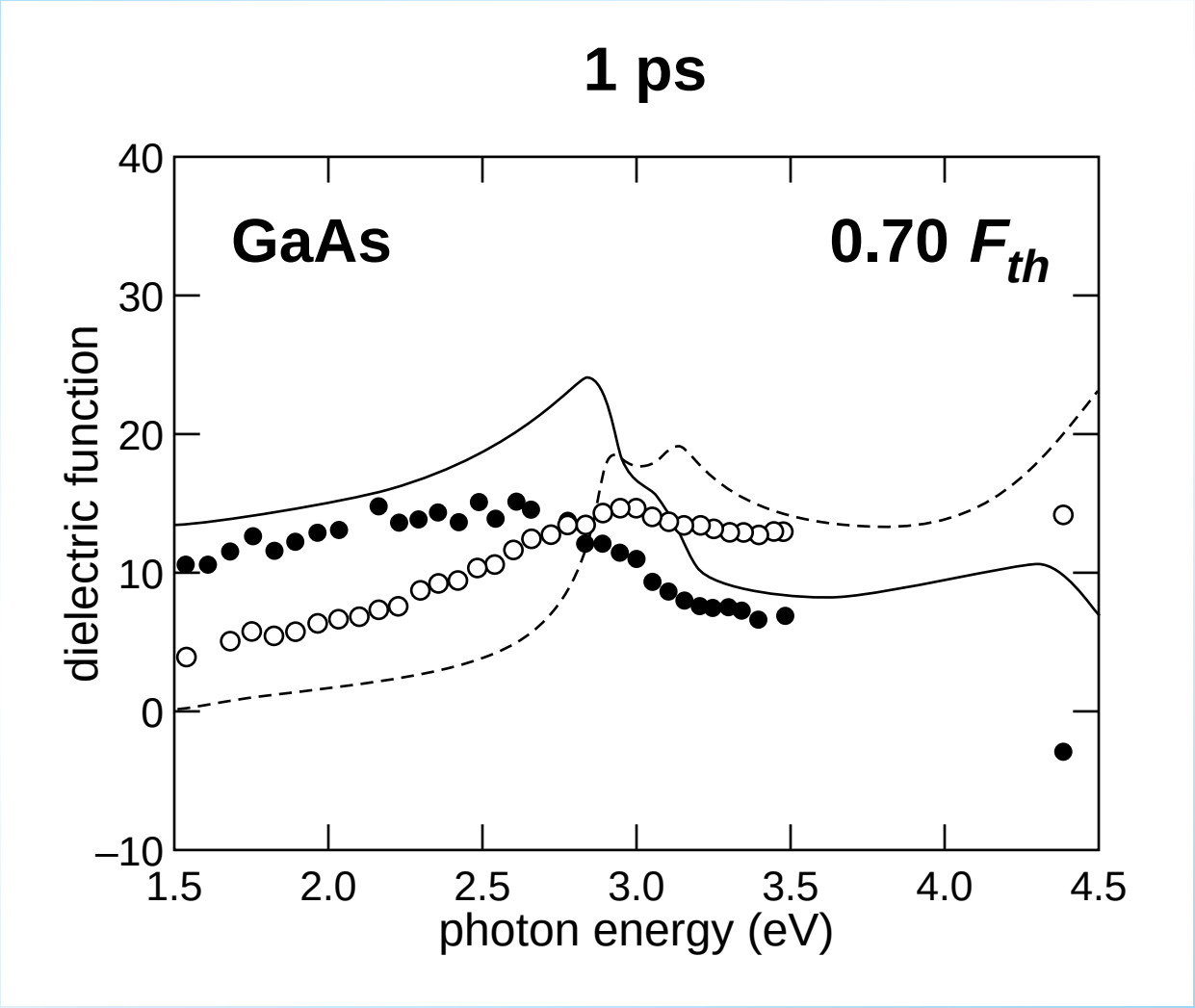
Results


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

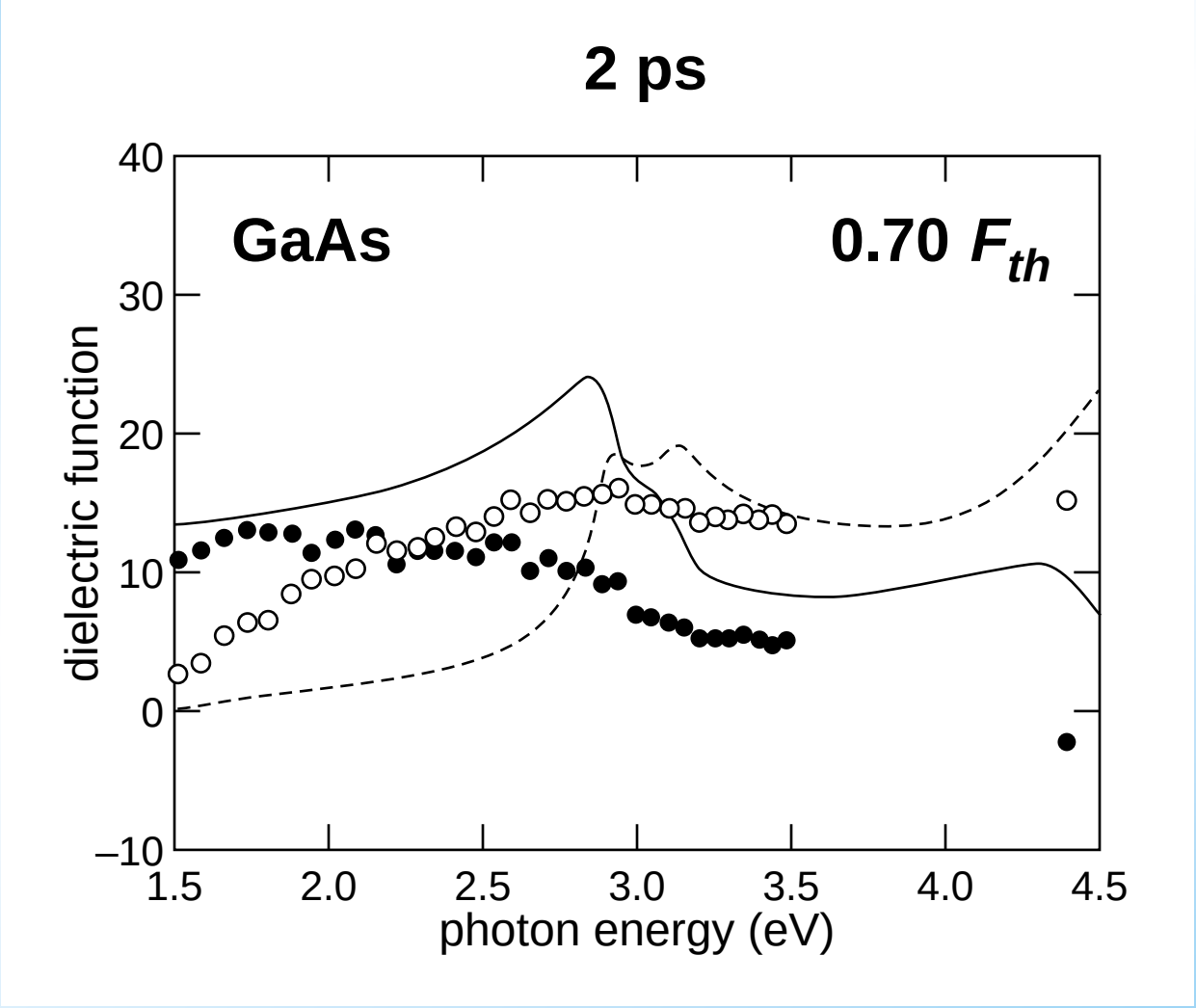
Results


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

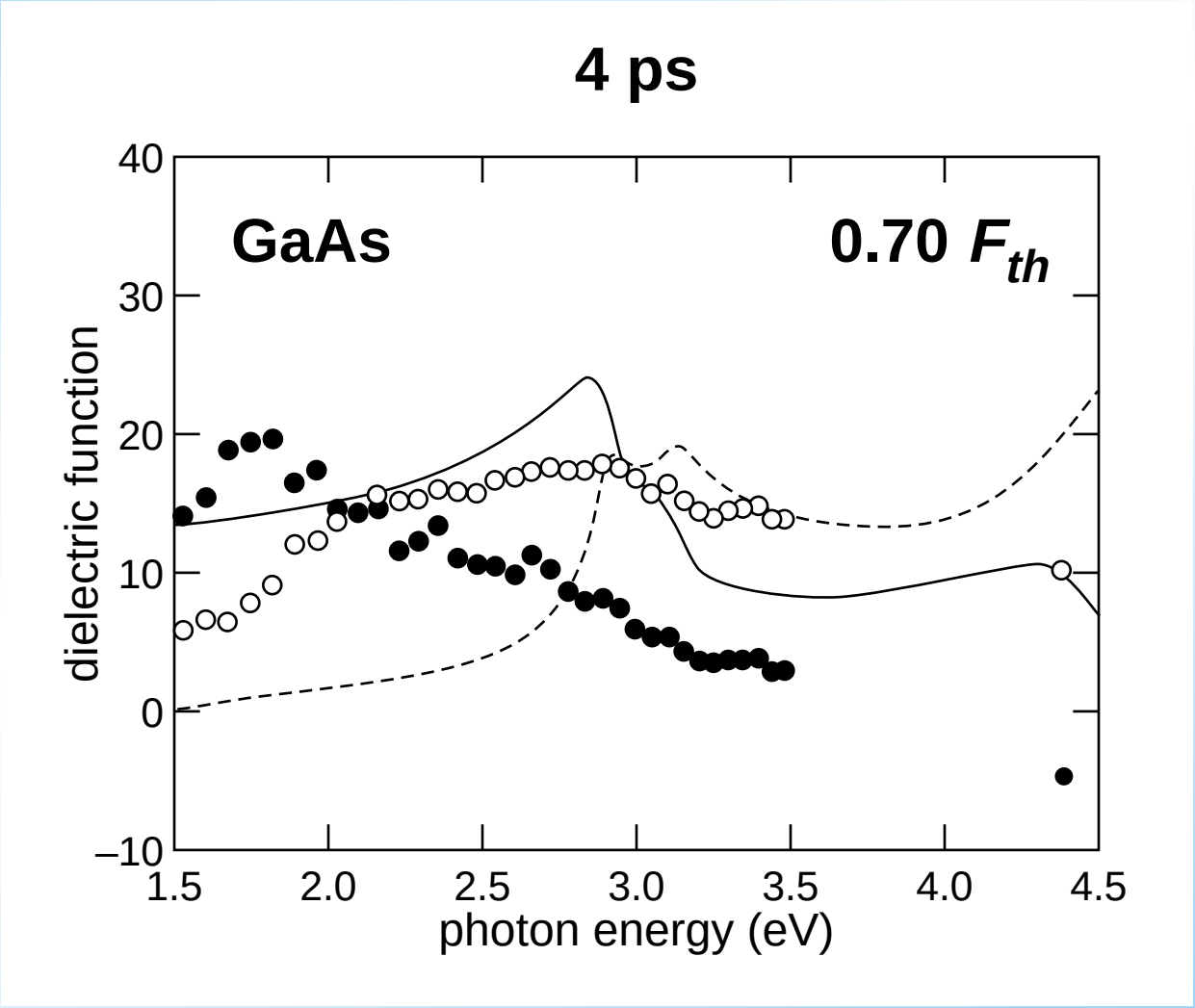
Results


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

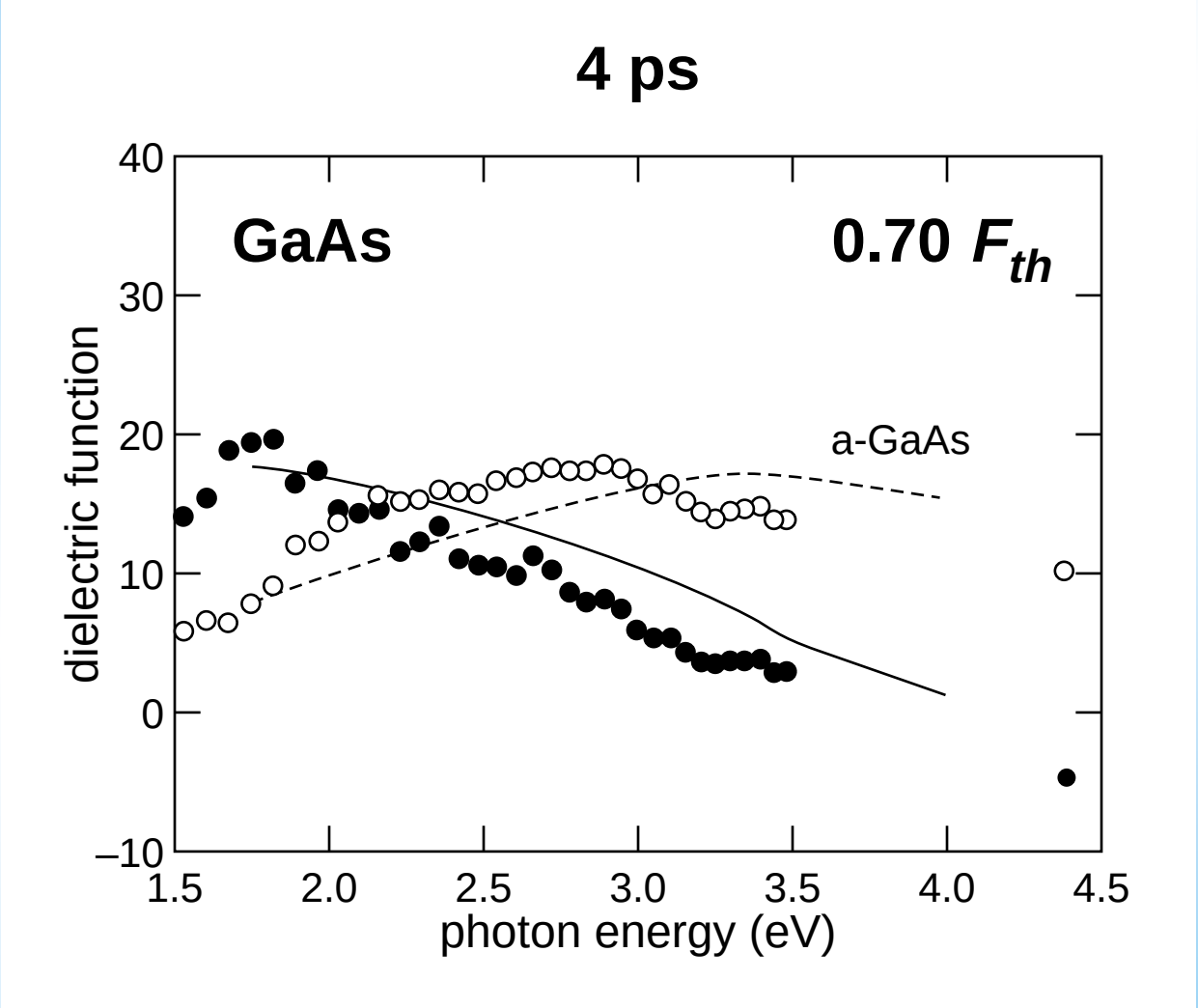
Results


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

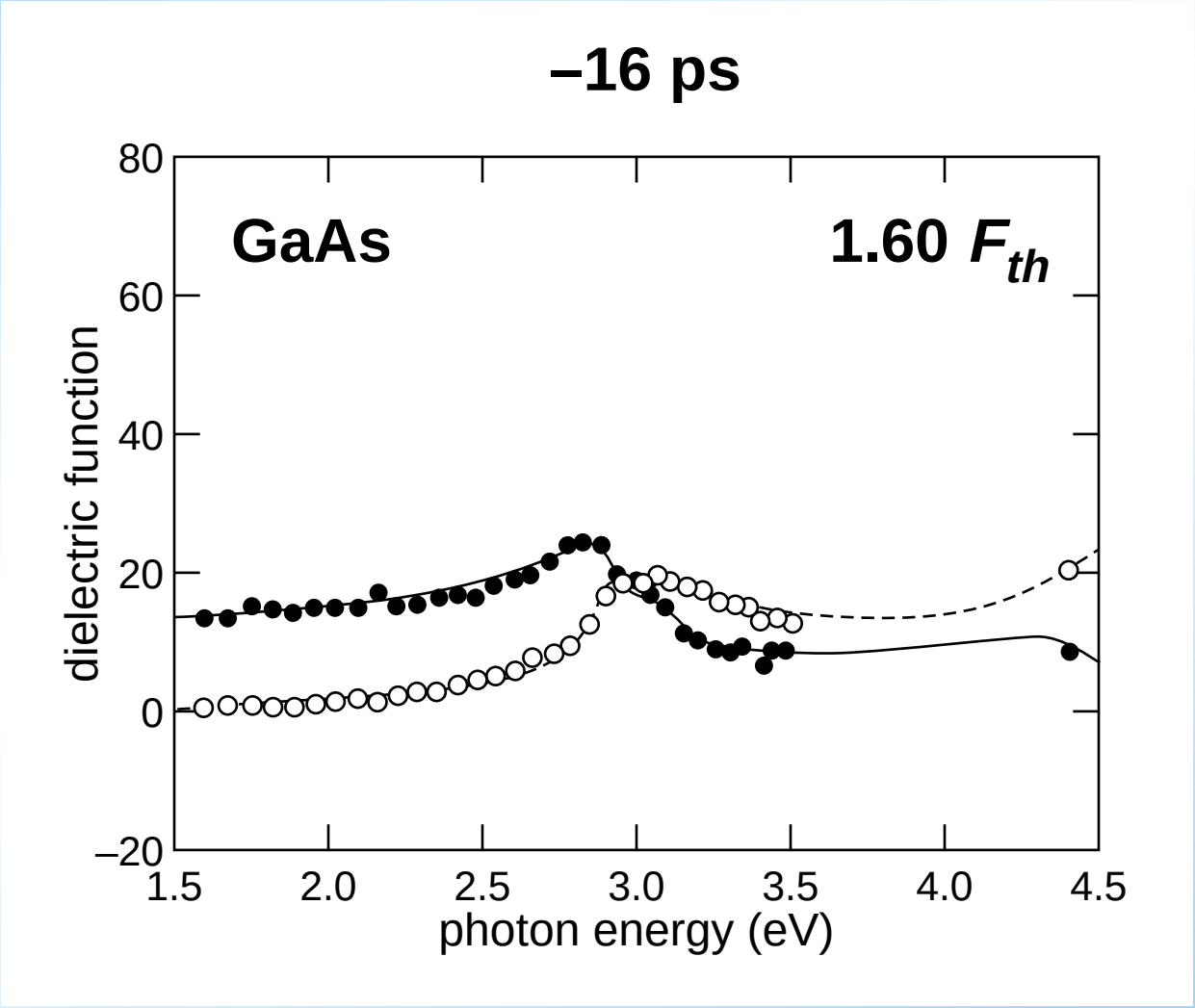
Results


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

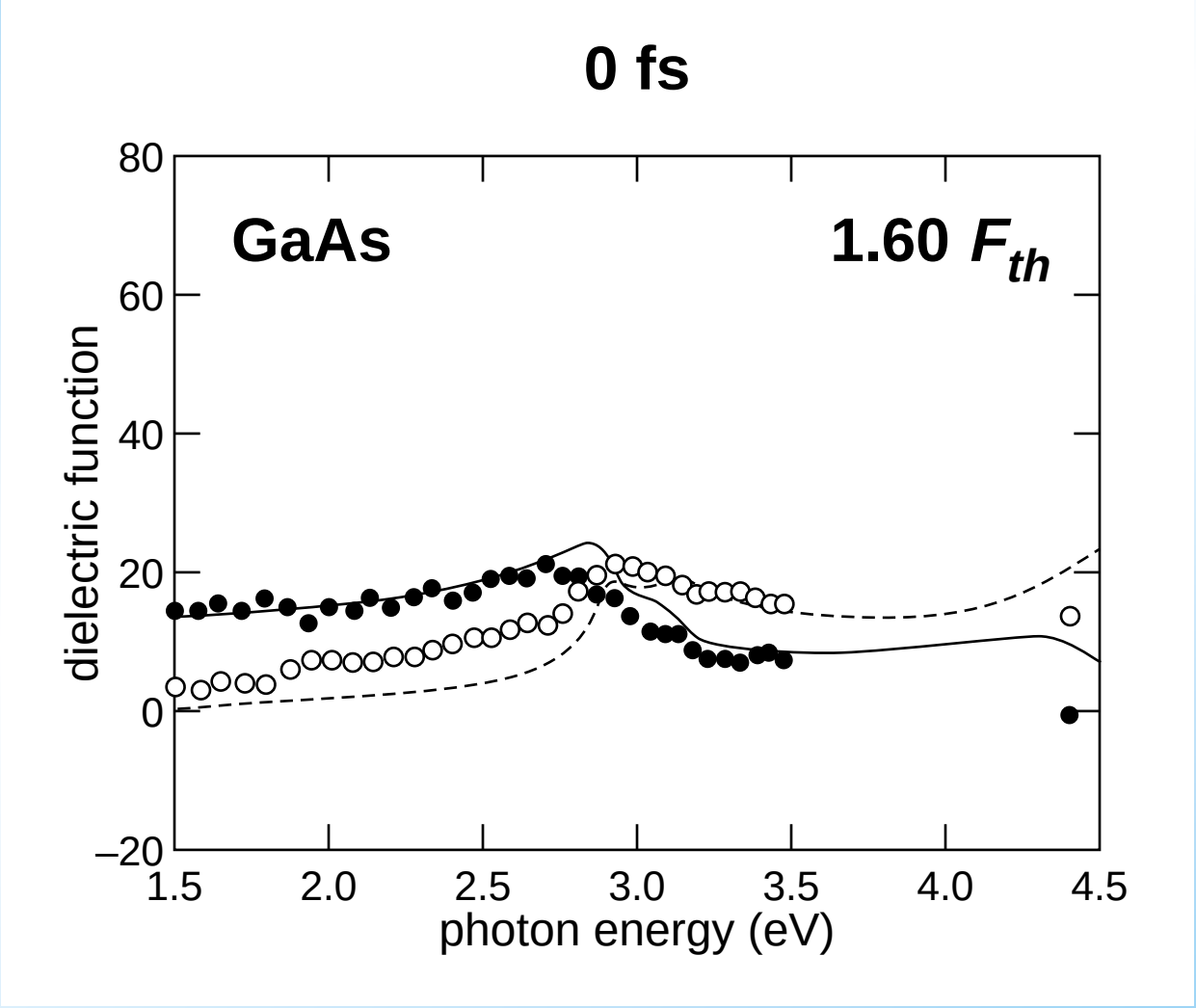
Results


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

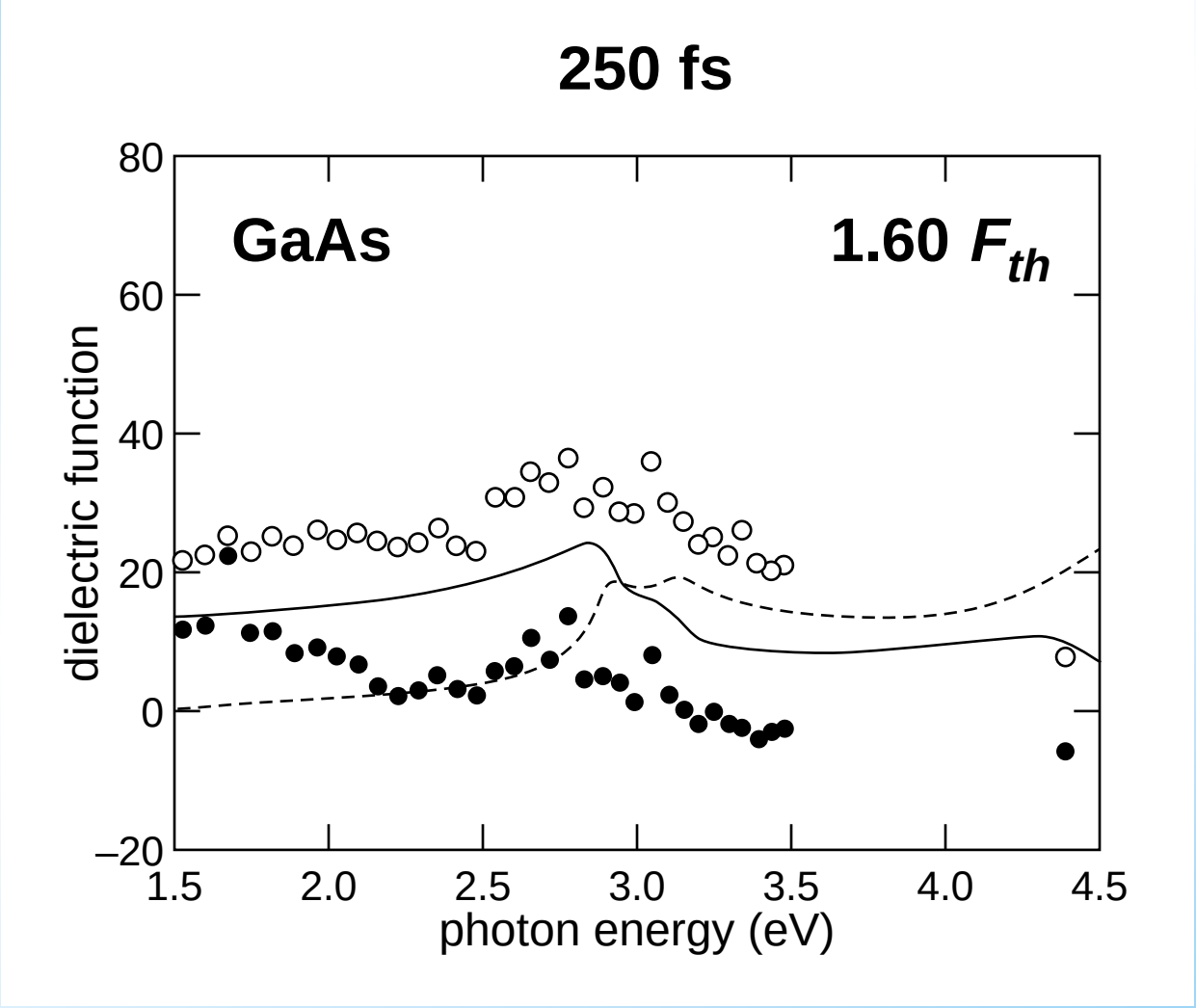
Results


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

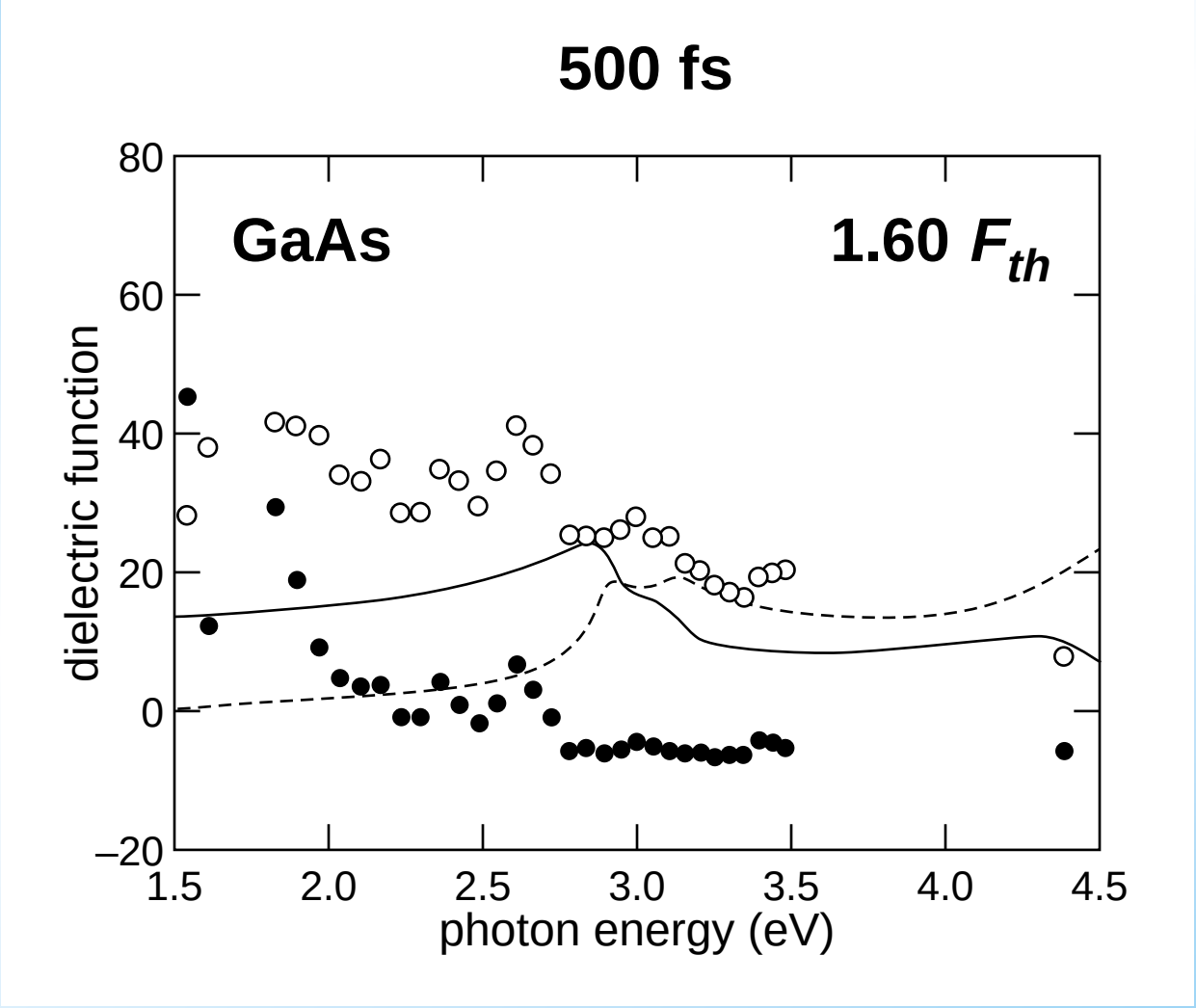
Results


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

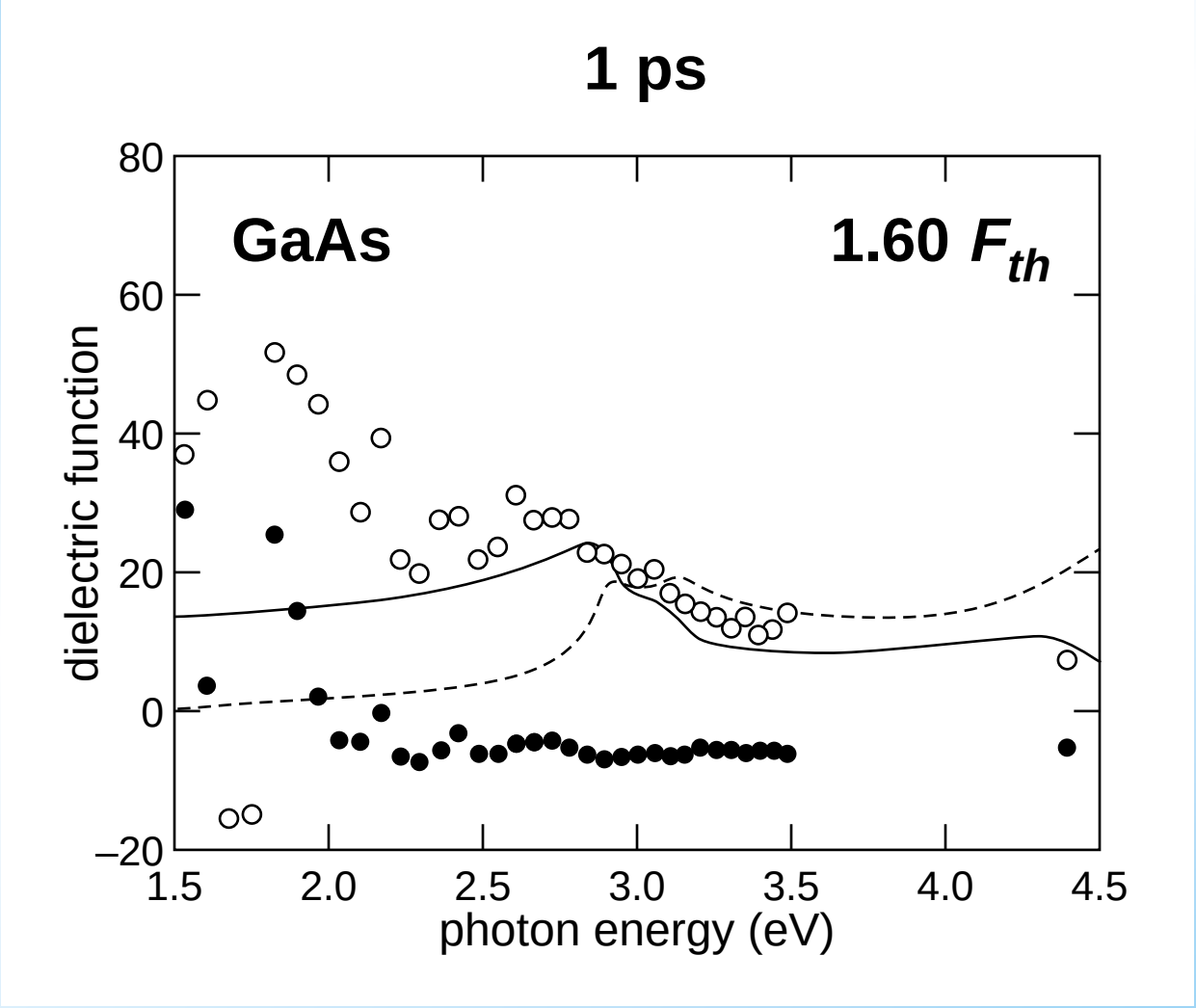
Results


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

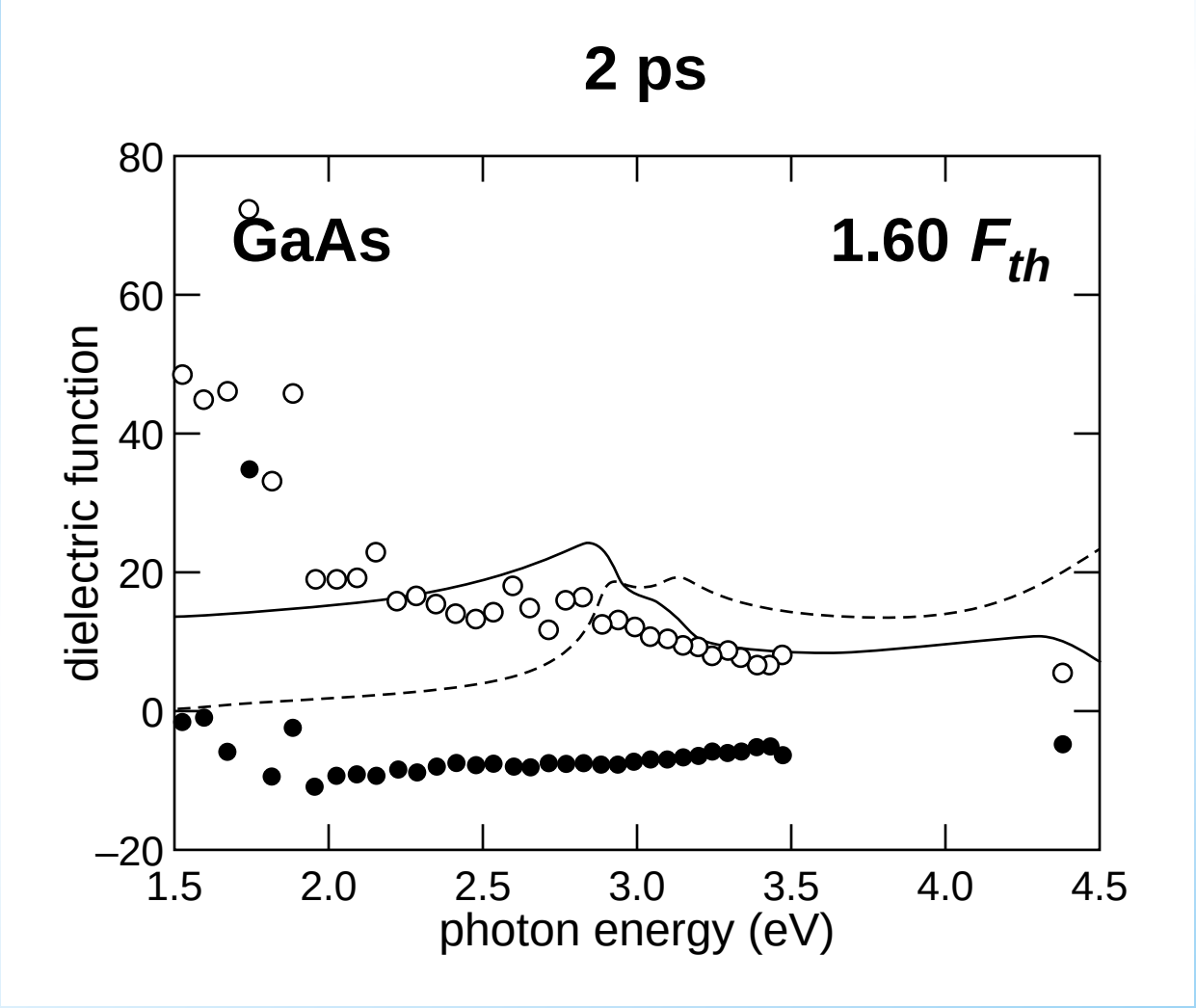
Results


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

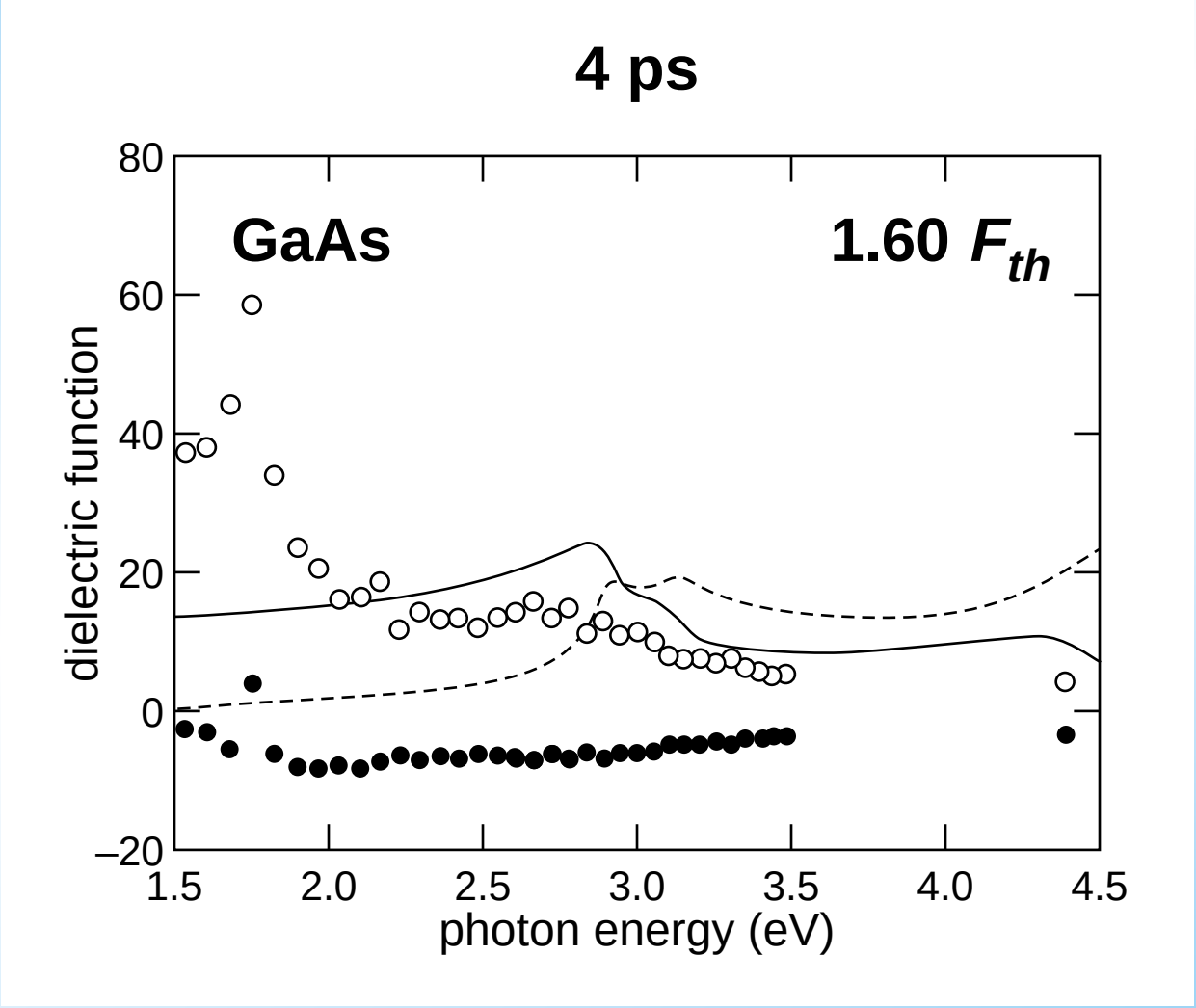
Results


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

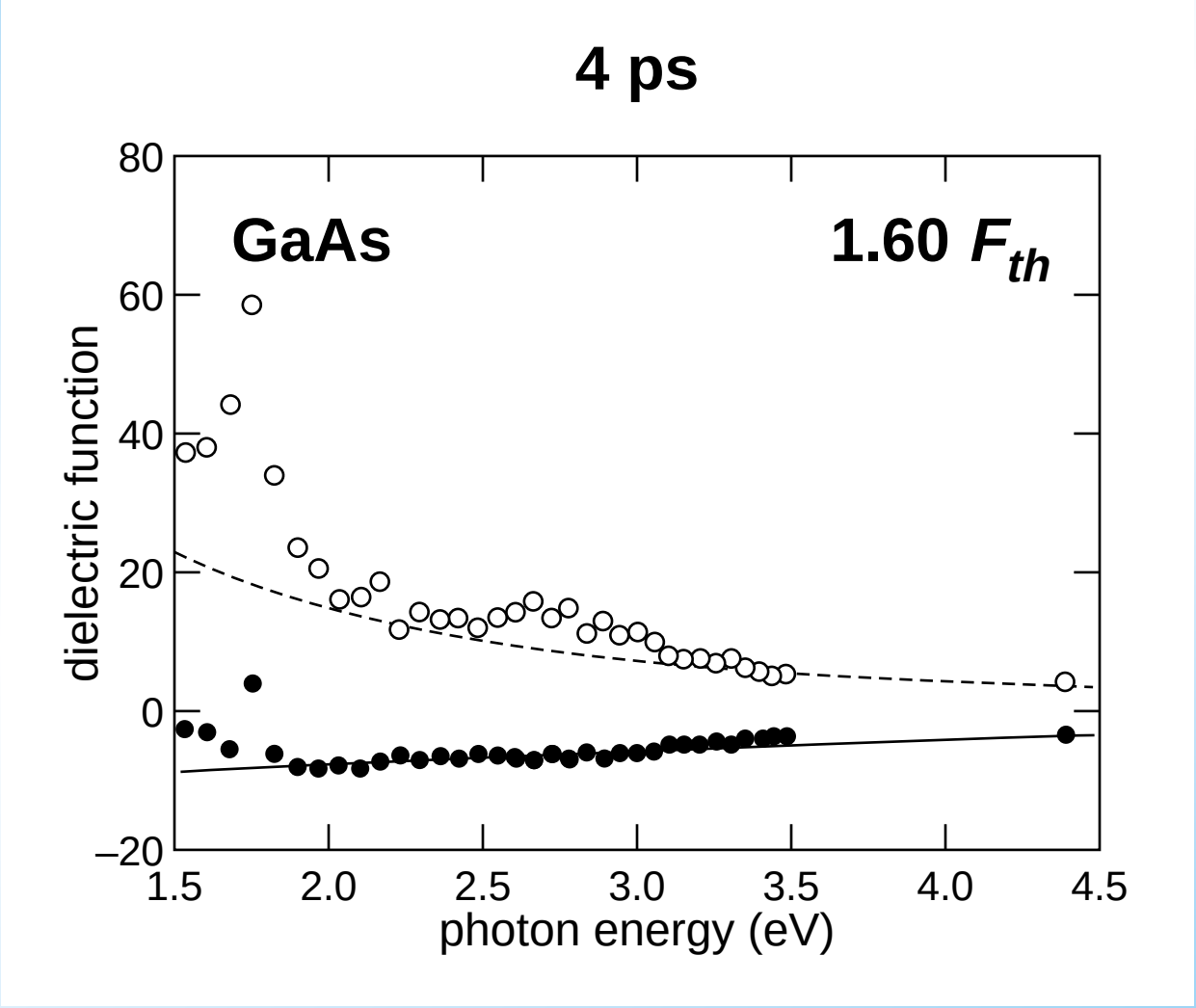
Results


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

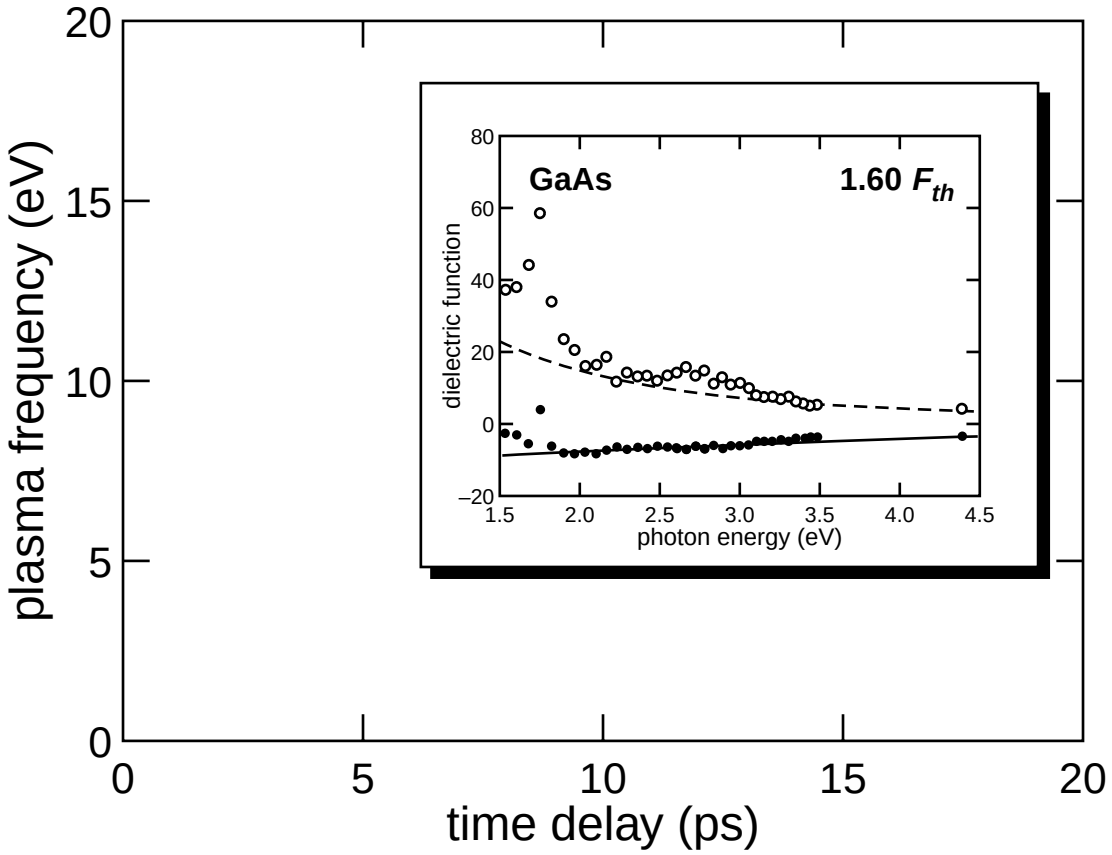
Results


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

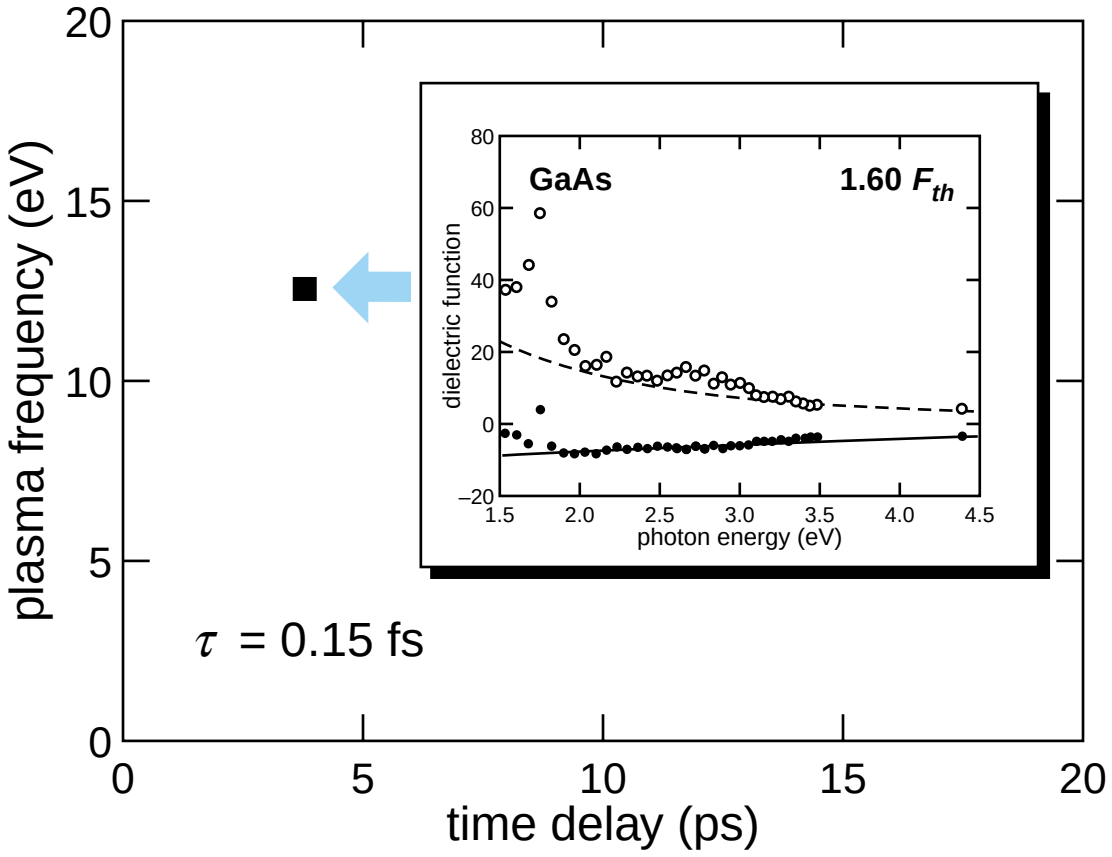
Results


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

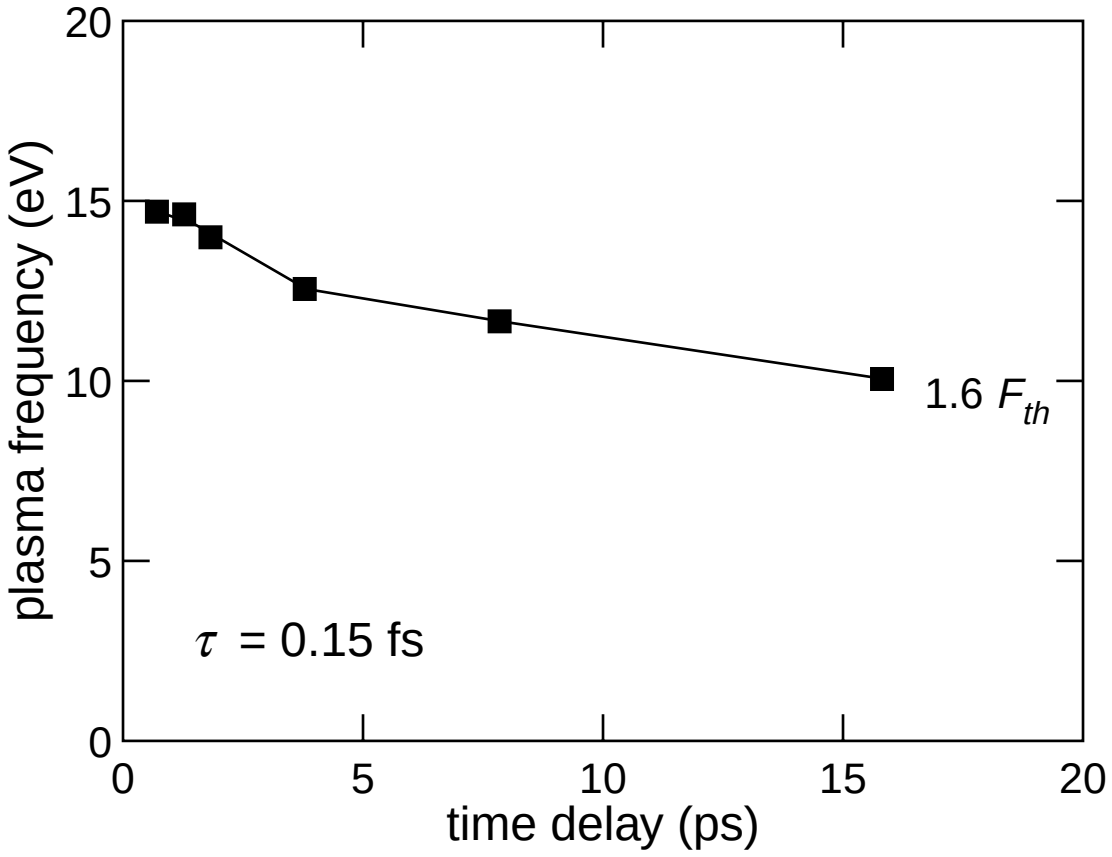
Results


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

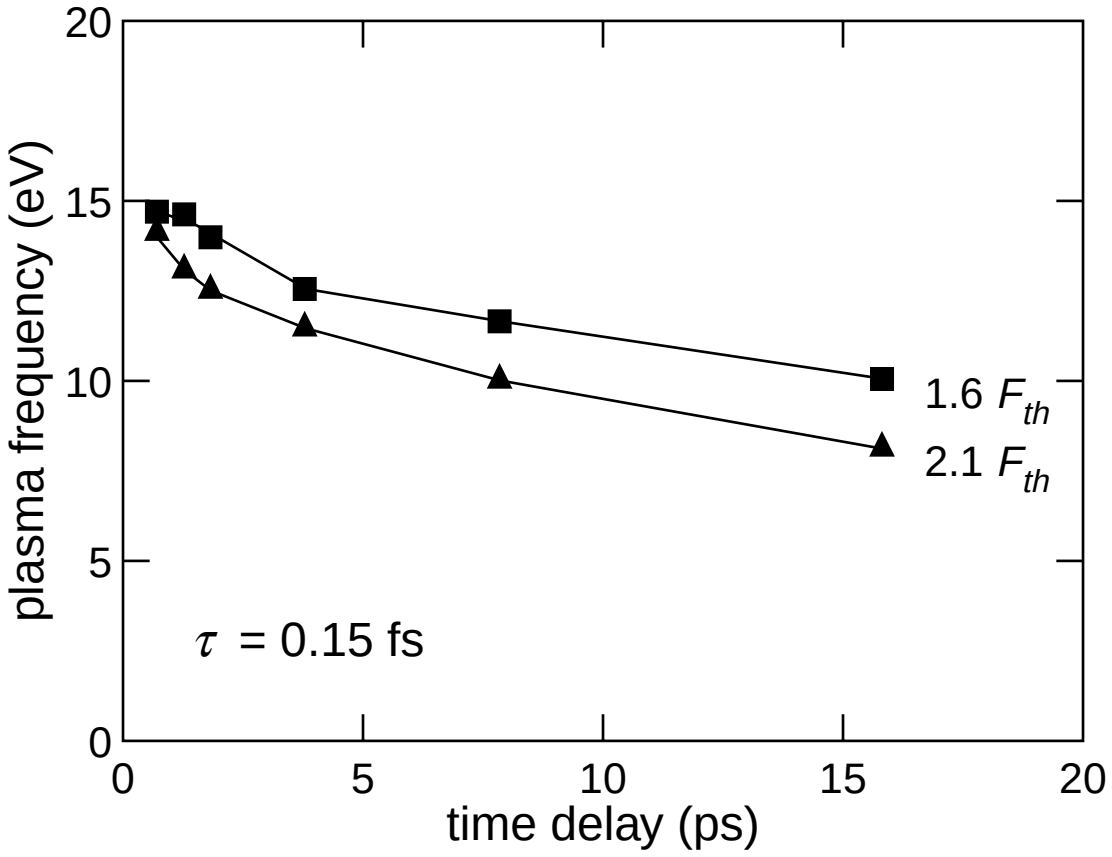
Results


plasma frequency (eV)
time delay (ps)
τ = 0.15 fs
GaAs
1.60 F_th
dielectric function
photon energy (eV)

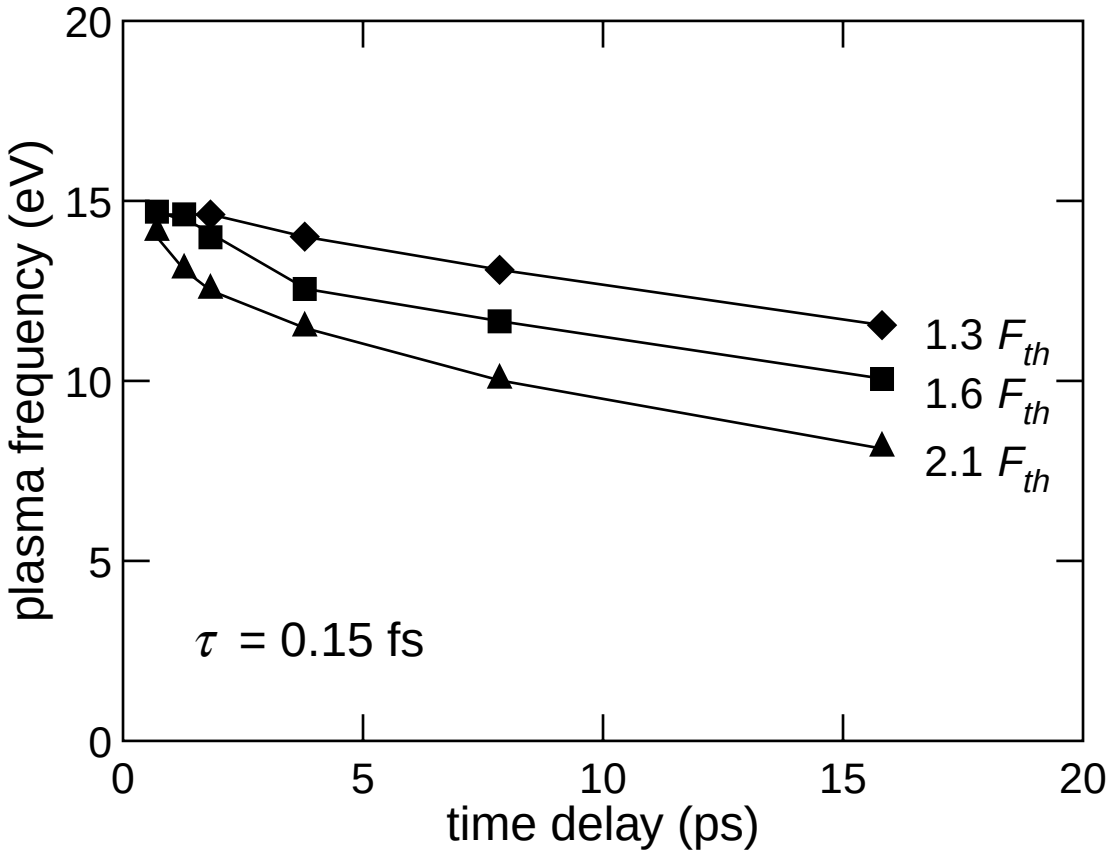
Results



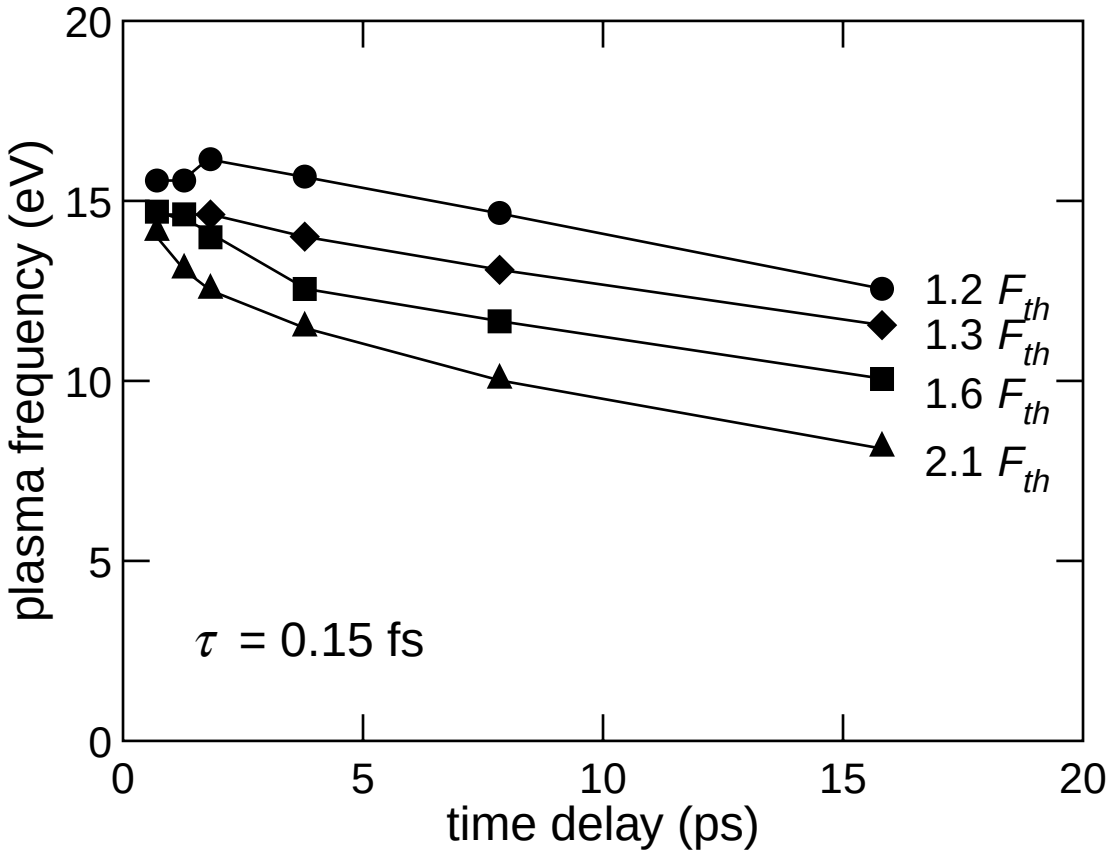
Results


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

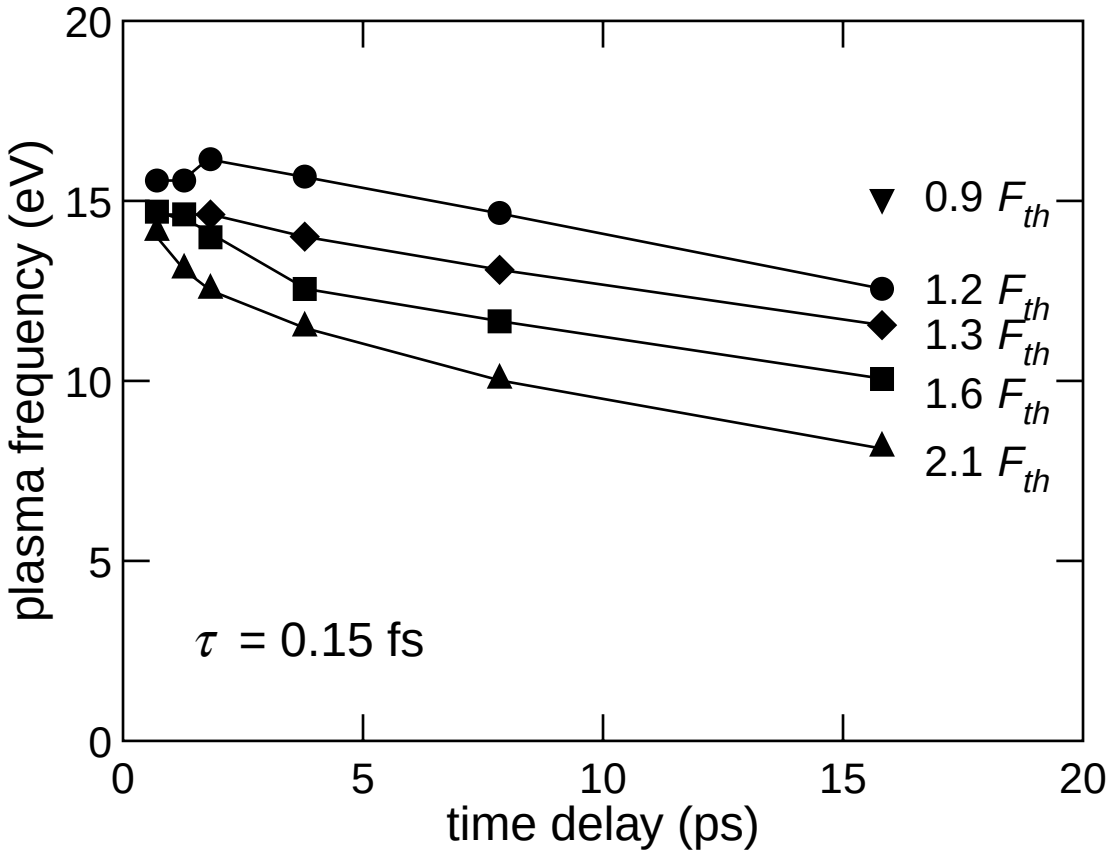
Results


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

Results


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

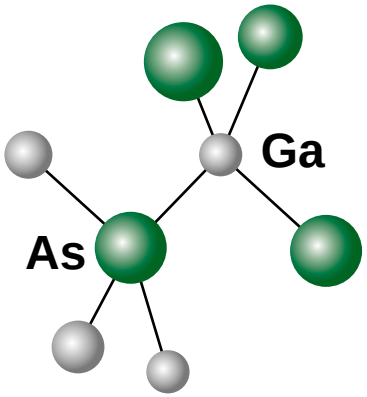
Results


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

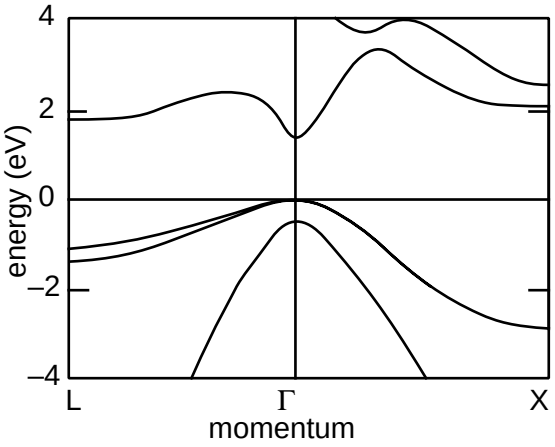
Discussion

short time scale

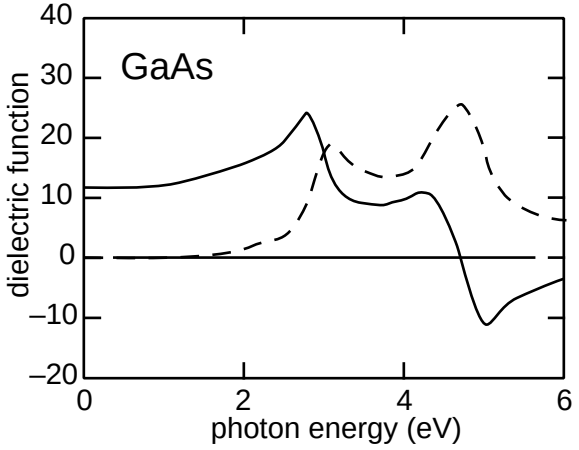
structure



bandstructure



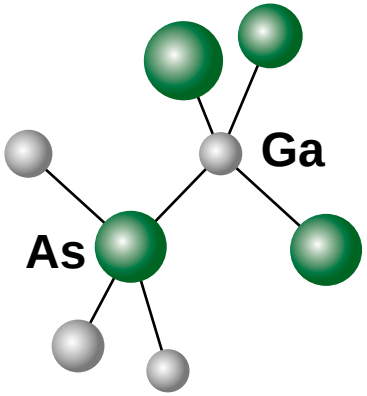
dielectric function



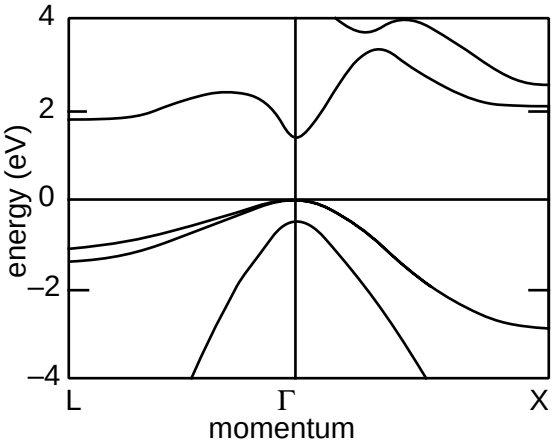
Discussion

short time scale

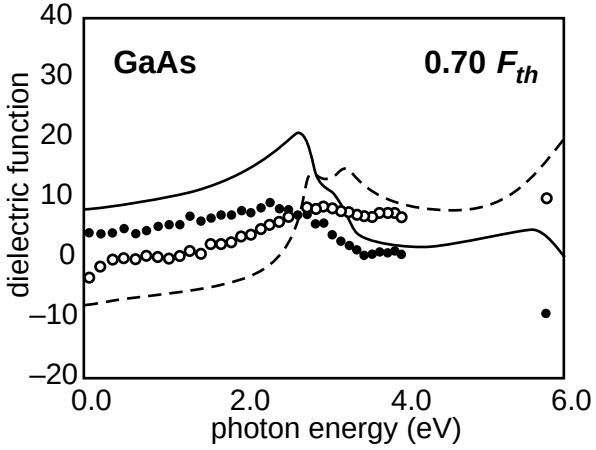
structure



bandstructure



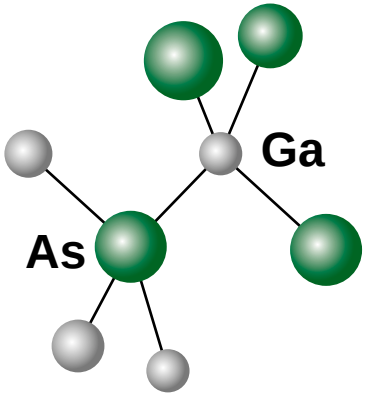
dielectric function



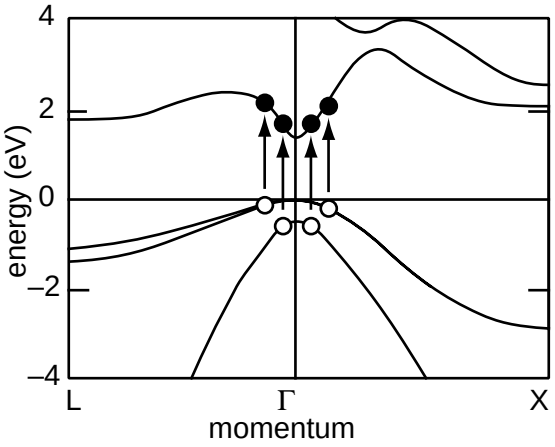
Discussion

short time scale

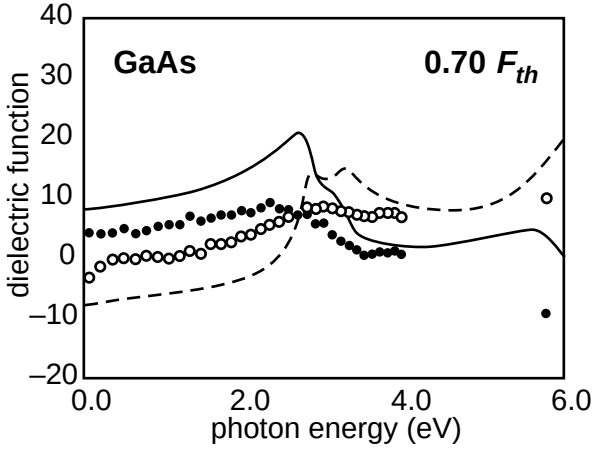
structure



bandstructure



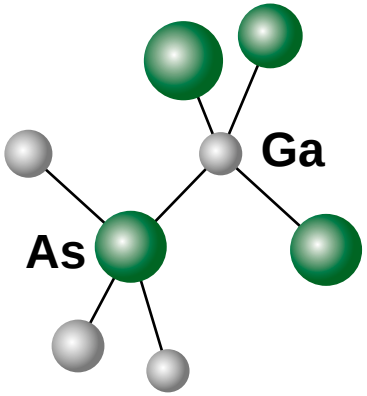
dielectric function



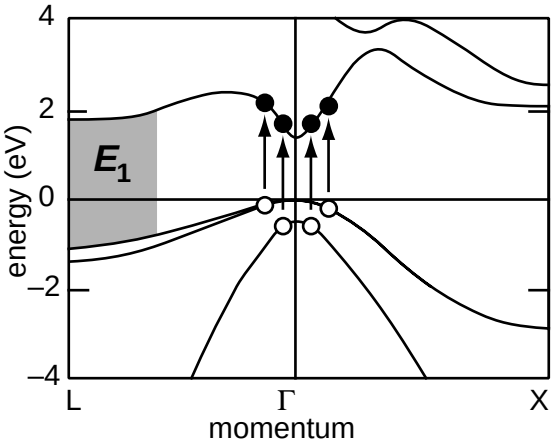
Discussion

short time scale

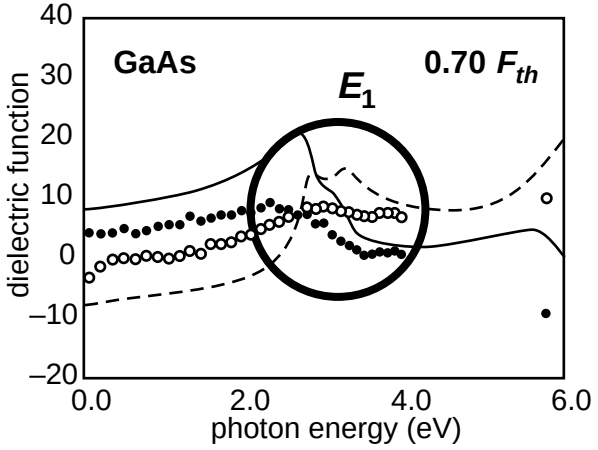
structure



bandstructure



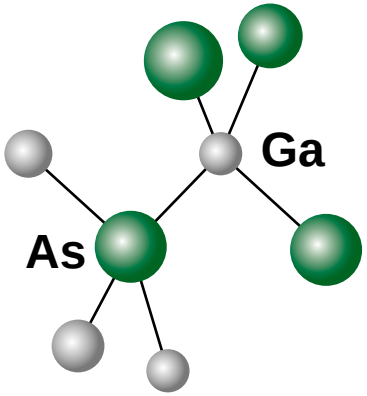
dielectric function



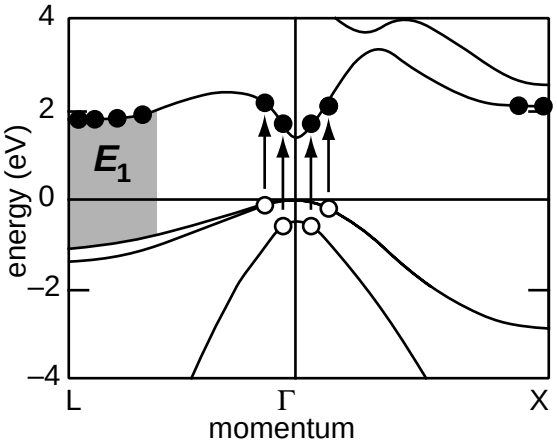
Discussion

short time scale

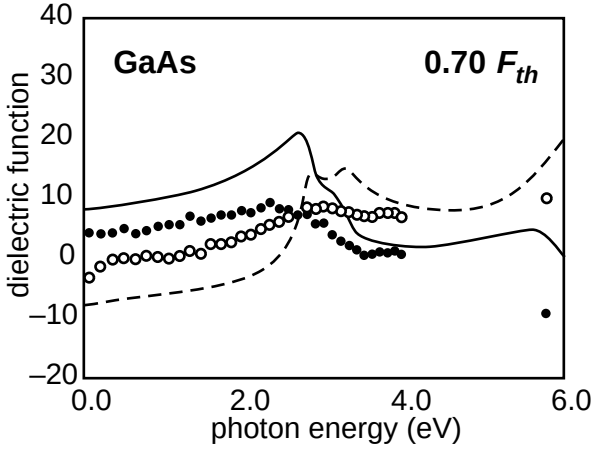
structure



bandstructure



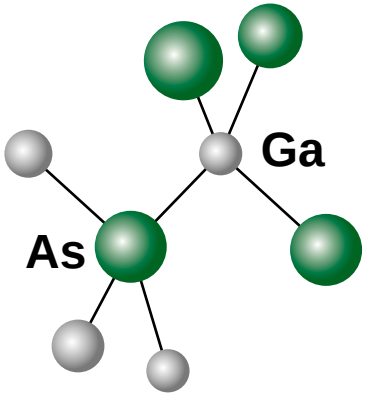
dielectric function



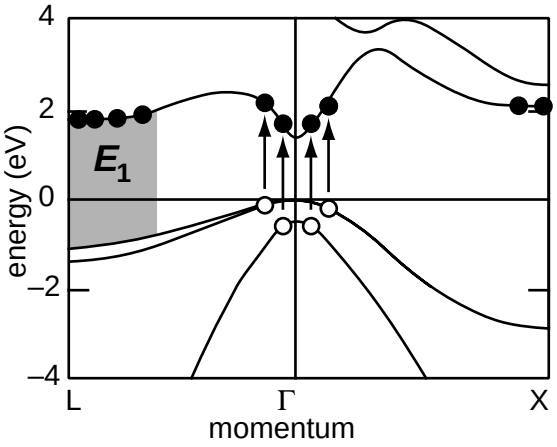
Discussion

short time scale

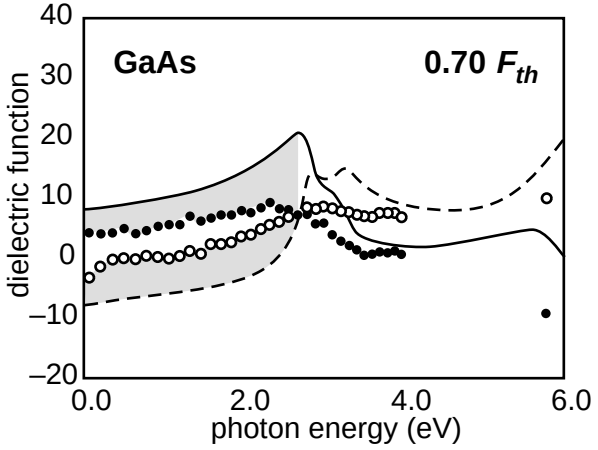
structure



bandstructure



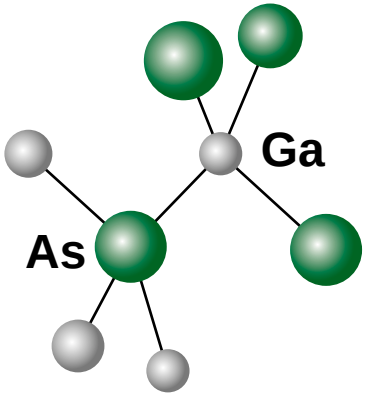
dielectric function



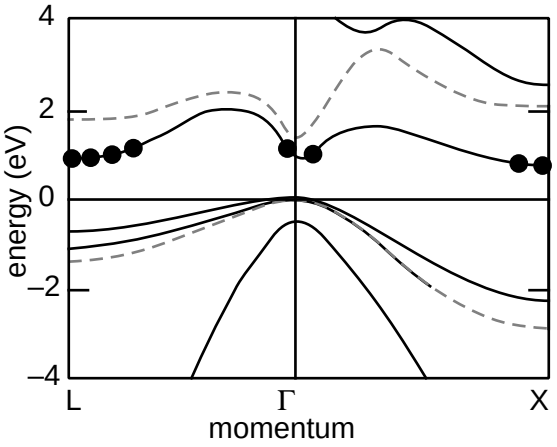
Discussion

short time scale

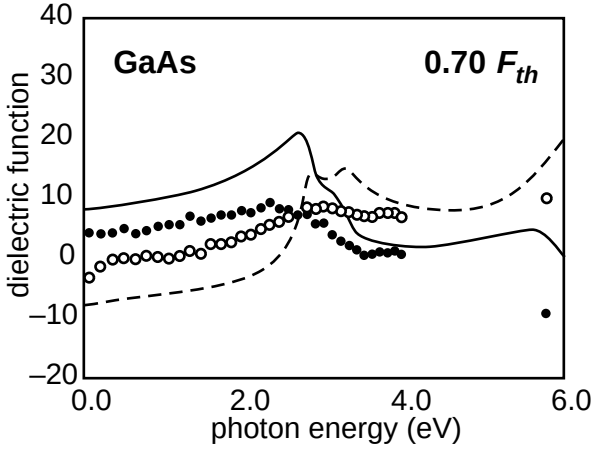
structure



bandstructure



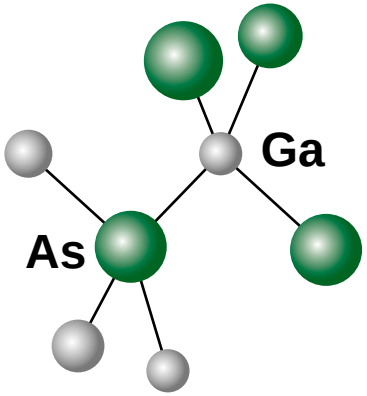
dielectric function



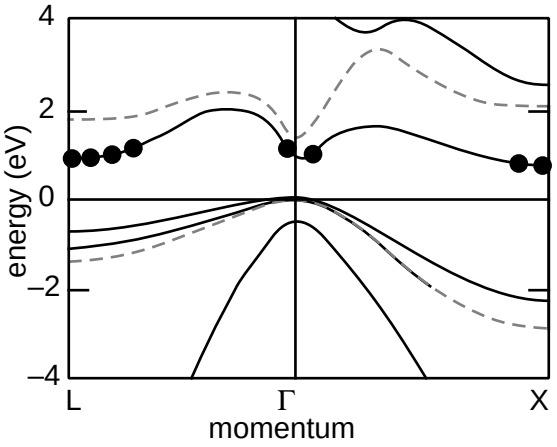
Discussion

short time scale

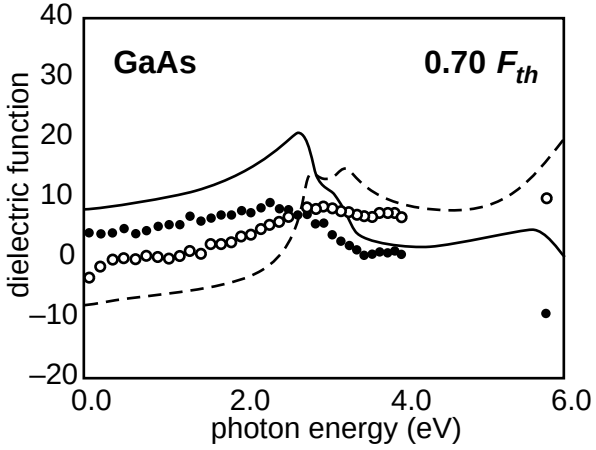
structure



bandstructure



dielectric function

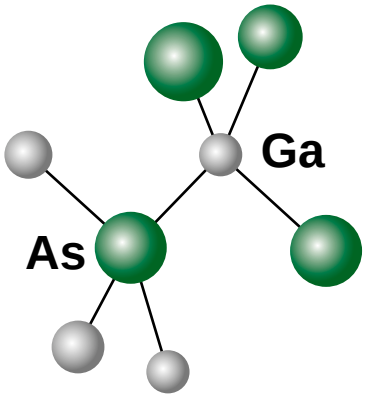


D.H. Kim, *et al.*, *Sol. State Comm.* 89, 119 (1994)

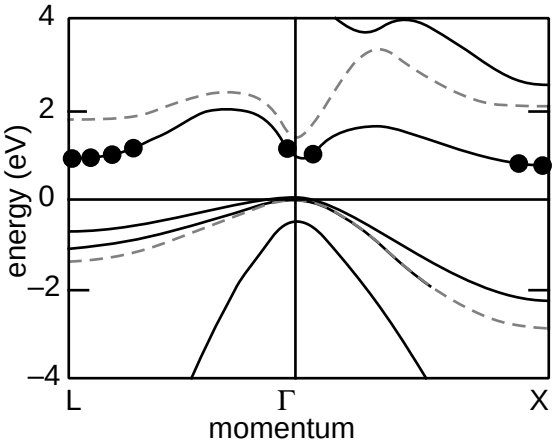
Discussion

short time scale

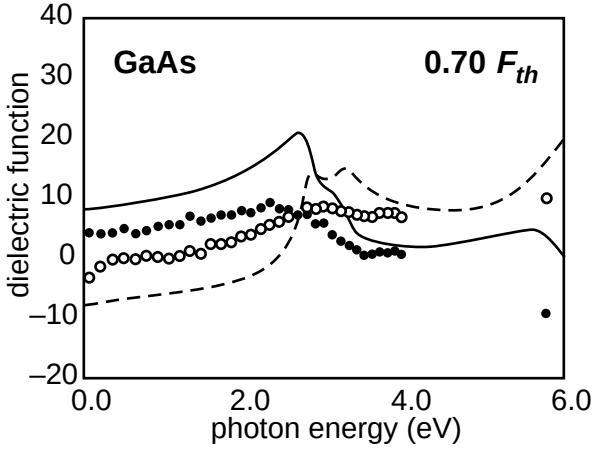
structure



bandstructure



dielectric function

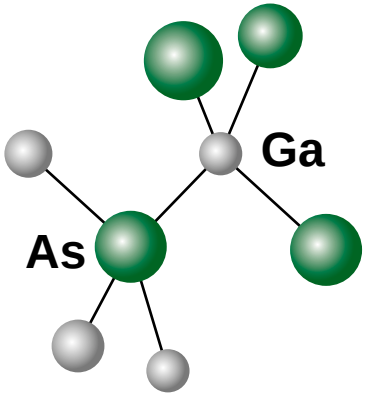


electronic effects dominate at short time scales…

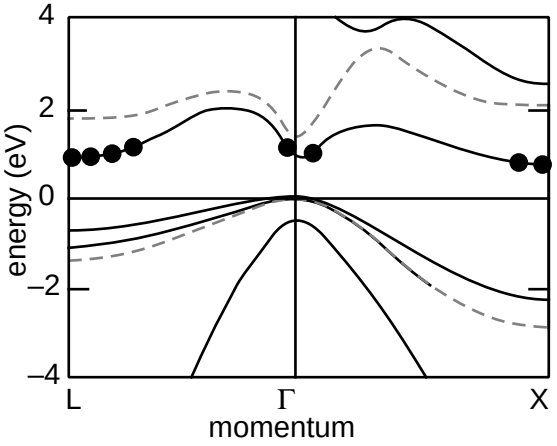
Discussion

short time scale

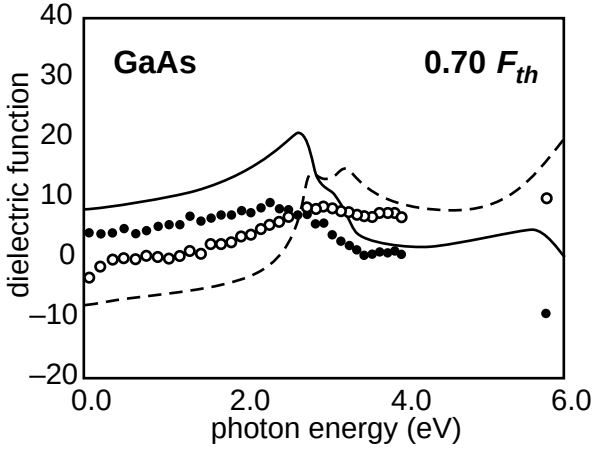
structure



bandstructure



dielectric function

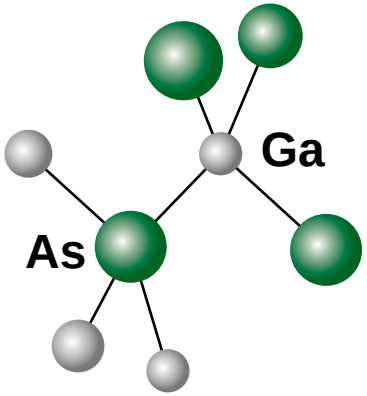


…but they are not as simple as we used to think

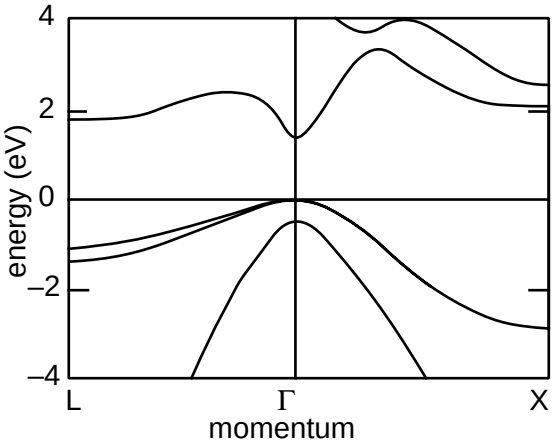
Discussion

long time scale, low fluence

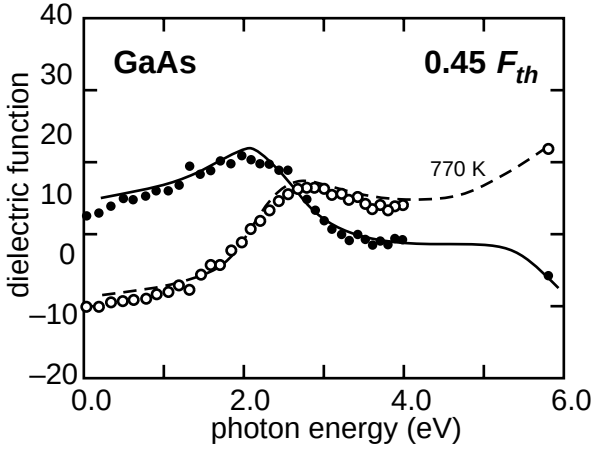
structure



bandstructure



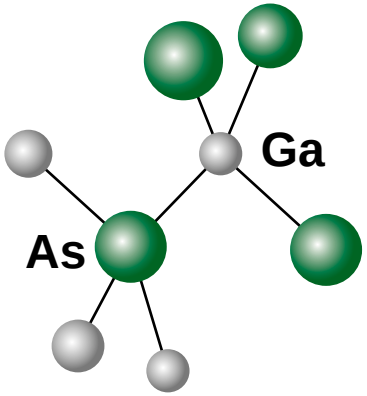
dielectric function



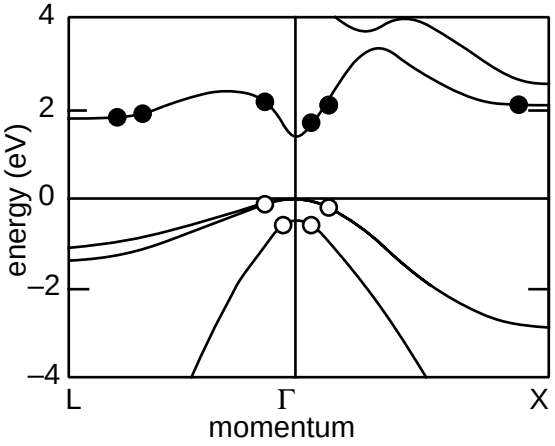
Discussion

long time scale, low fluence

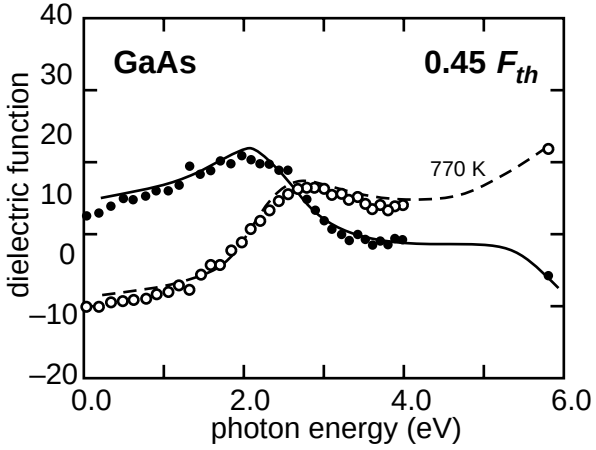
structure



bandstructure



dielectric function

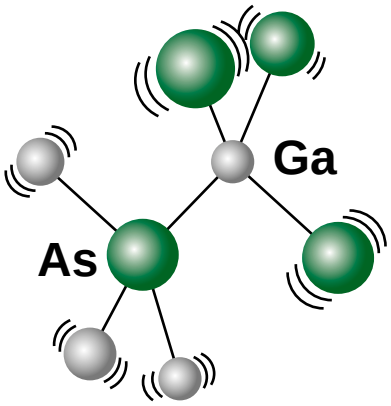


carrier density down, electronic effects subsided…

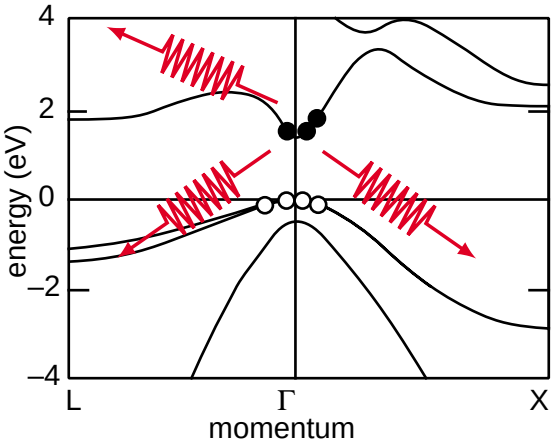
Discussion

long time scale, low fluence

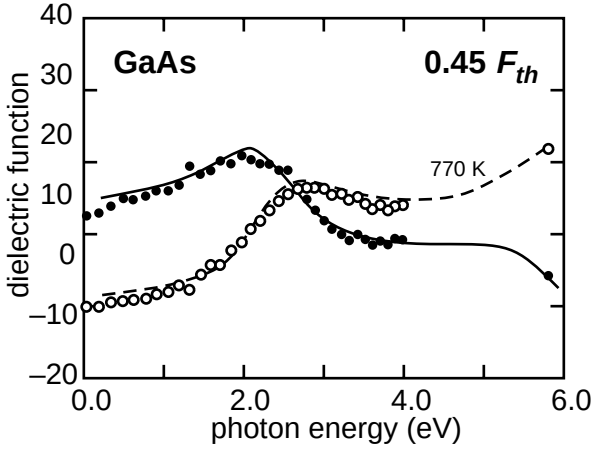
structure



bandstructure



dielectric function

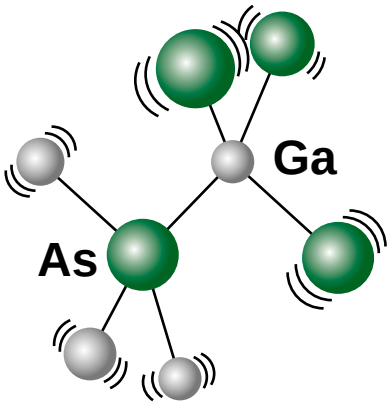


…and lattice heats due to carrier relaxation

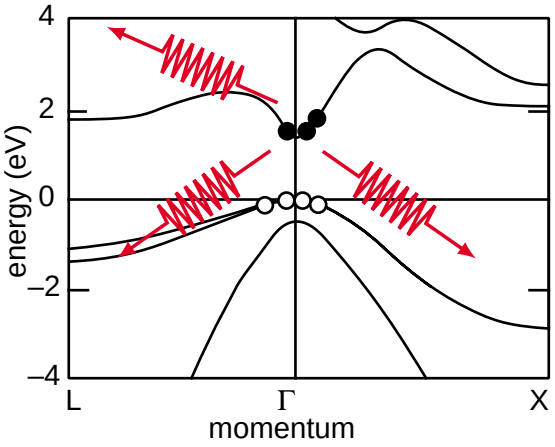
Discussion

long time scale, low fluence

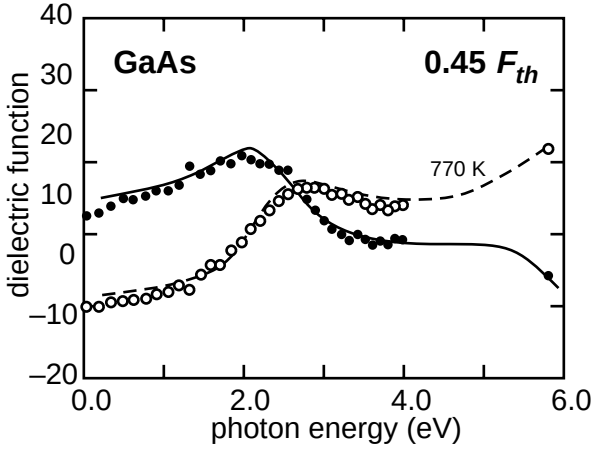
structure



bandstructure

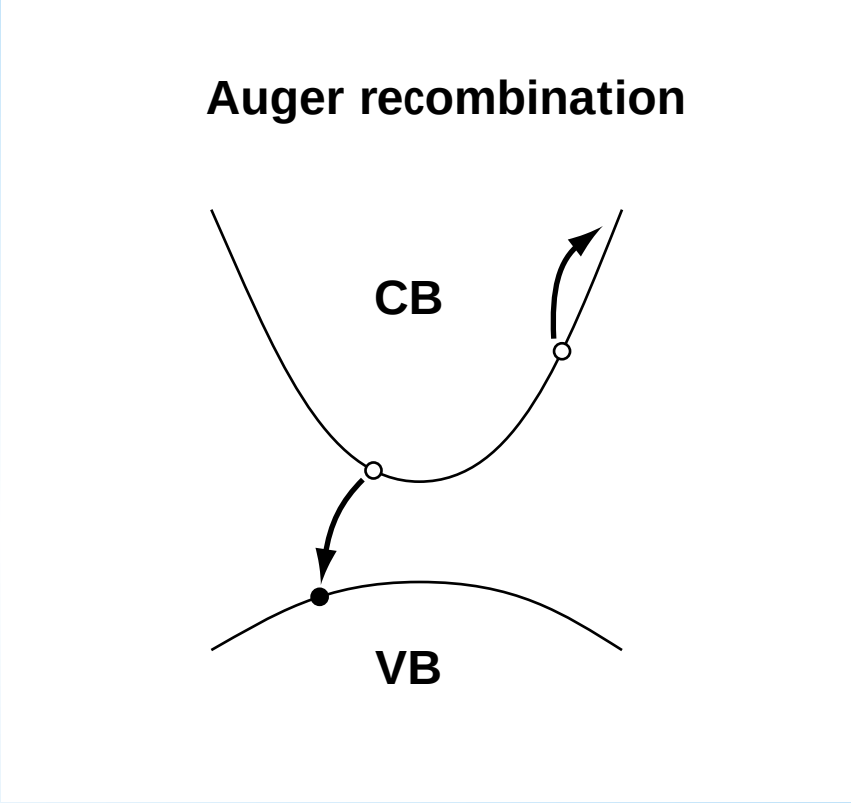


dielectric function



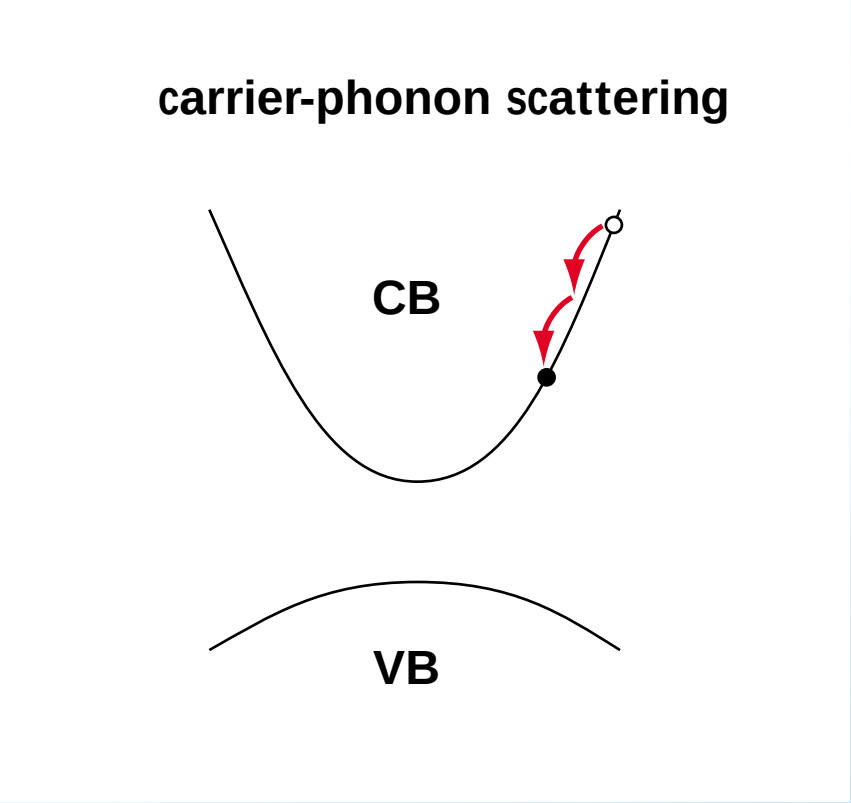
But… why do electronic effects disappear after 2 ps while lattice heats in 7 ps?

Discussion



Auger lowers *N* without changing *E* in 2–4 ps…

Discussion

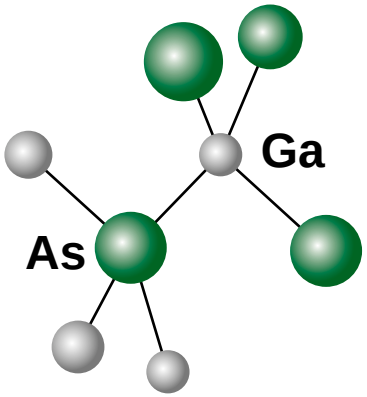


…and highly excited electrons cool to lattice in 7 ps

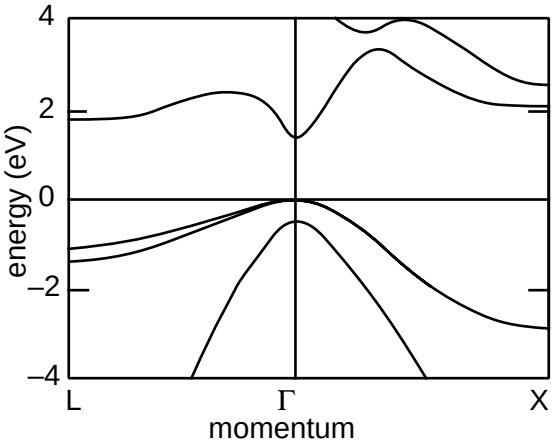
Discussion

long time scale, high fluence

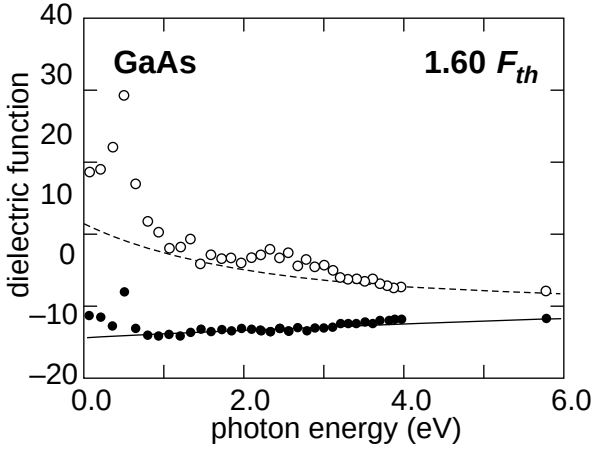
structure



bandstructure



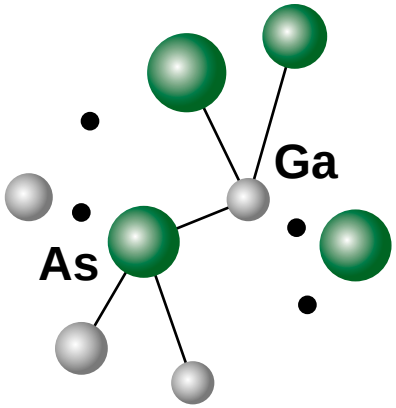
dielectric function



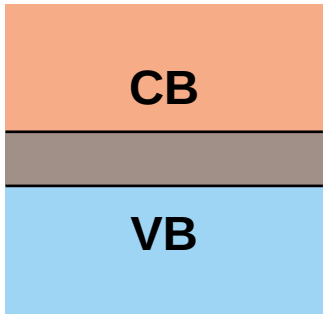
Discussion

long time scale, high fluence

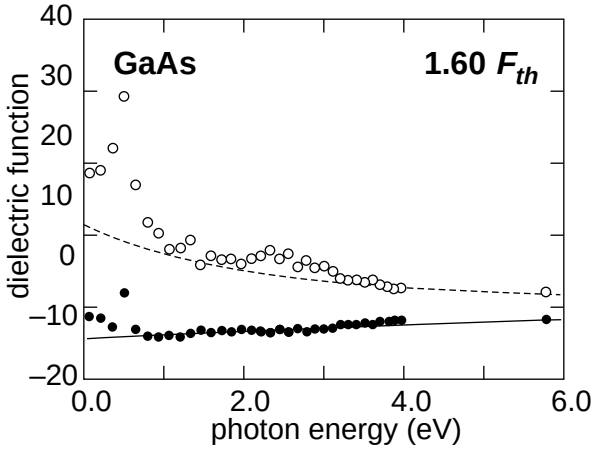
structure



bandstructure

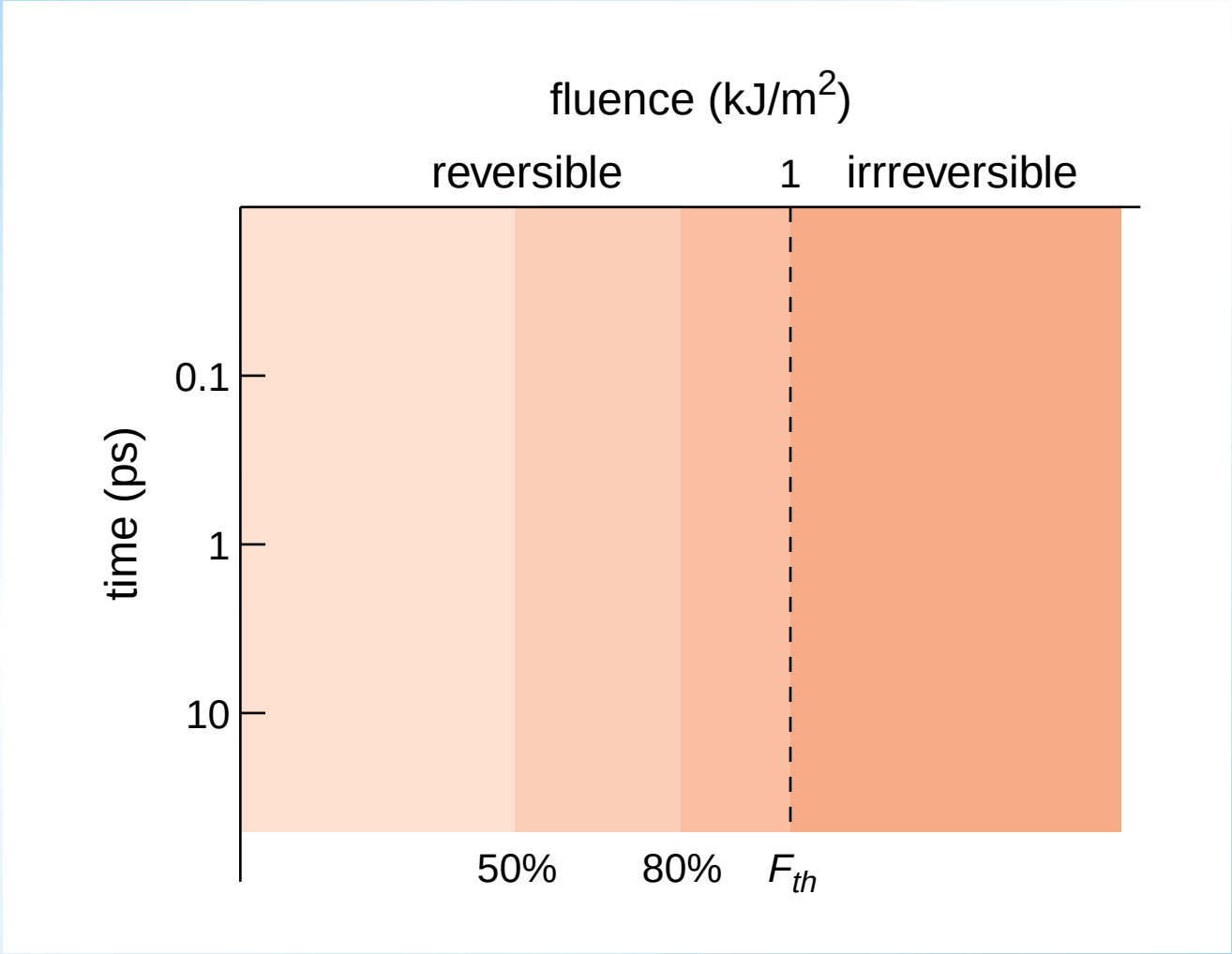


dielectric function

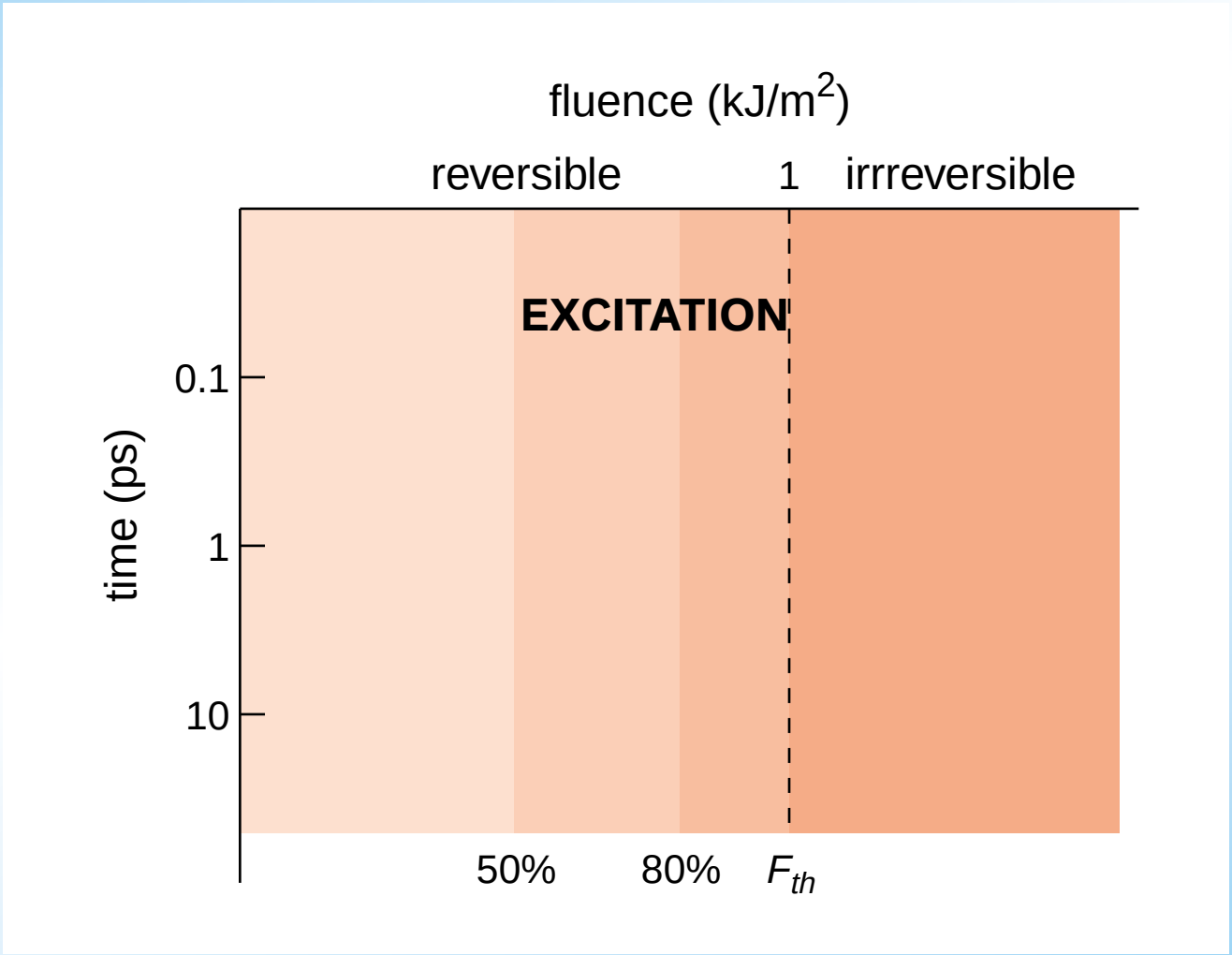


gradual drop in gap → not electronic effect

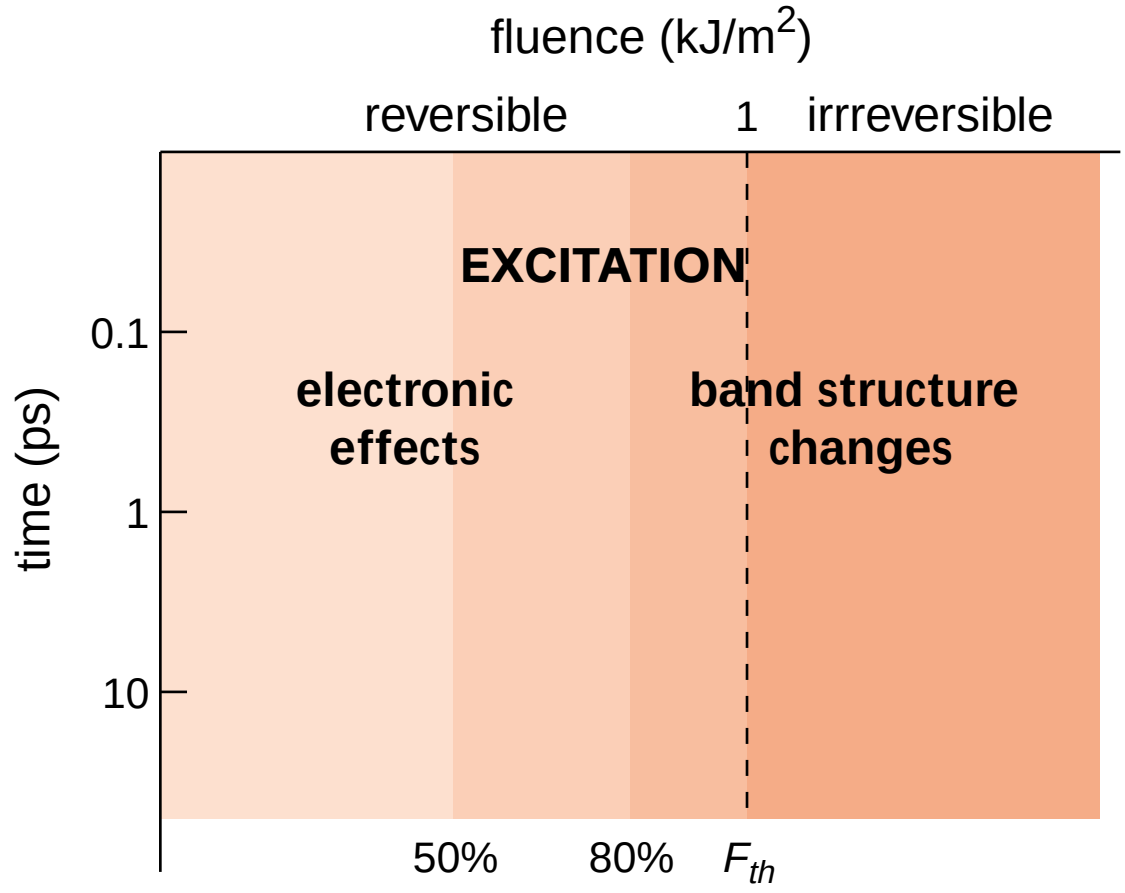
Discussion


fluence (kJ/m^2)
reversible
1
irrreversible
time (ps)
0.1
1
10
50%
80%
F_{th}

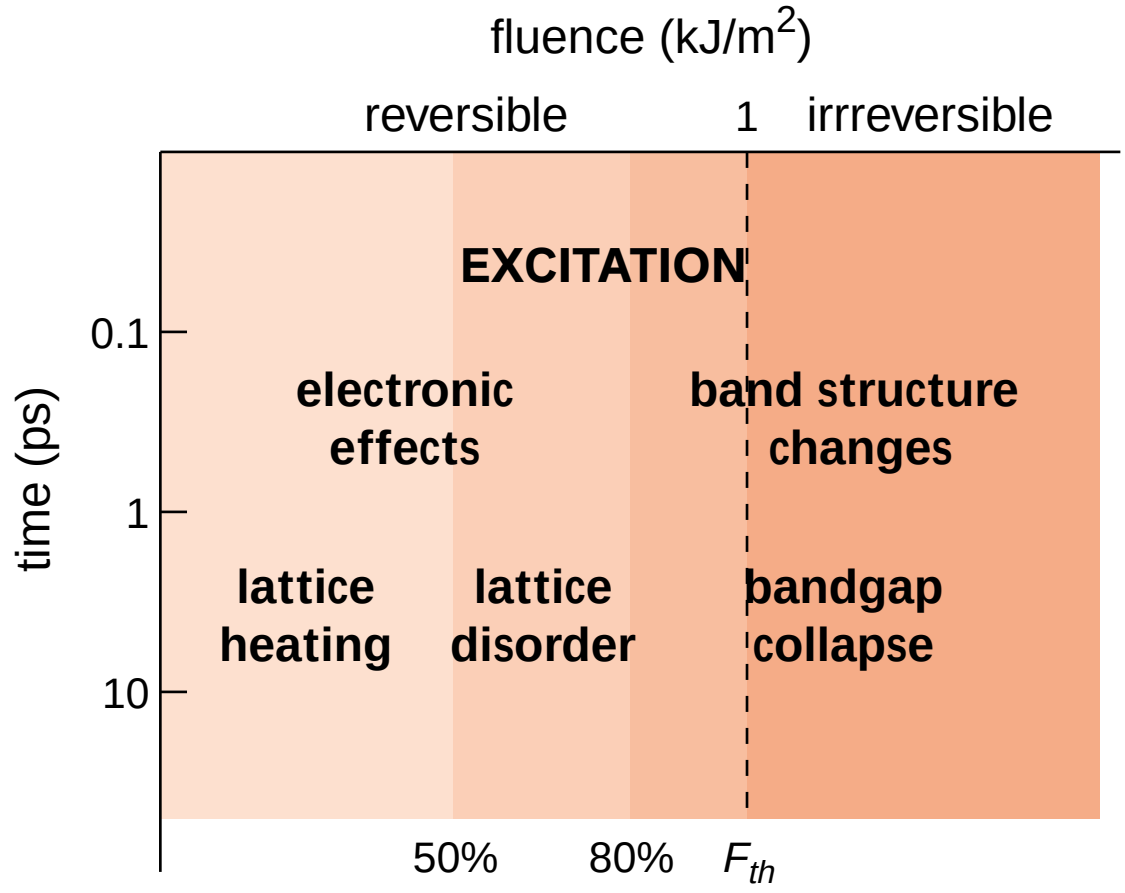
Discussion


fluence (kJ/m^2)
reversible
1
irrreversible
EXCITATION
0.1
1
10
time (ps)
50%
80%
F_{th}

Discussion


fluence (kJ/m^2)
reversible
1
irrreversible
EXCITATION
electronic effects
band structure changes
time (ps)
0.1
1
10
50%
80%
F_{th}

Discussion


fluence (kJ/m2)
reversible
1
irrreversible
EXCITATION
0.1
1
10
time (ps)
electronic effects
band structure changes
lattice heating
lattice disorder
bandgap collapse
50%
80%
Fth

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**

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Prof. C. Klingshirn

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

http://mazur-www.harvard.edu