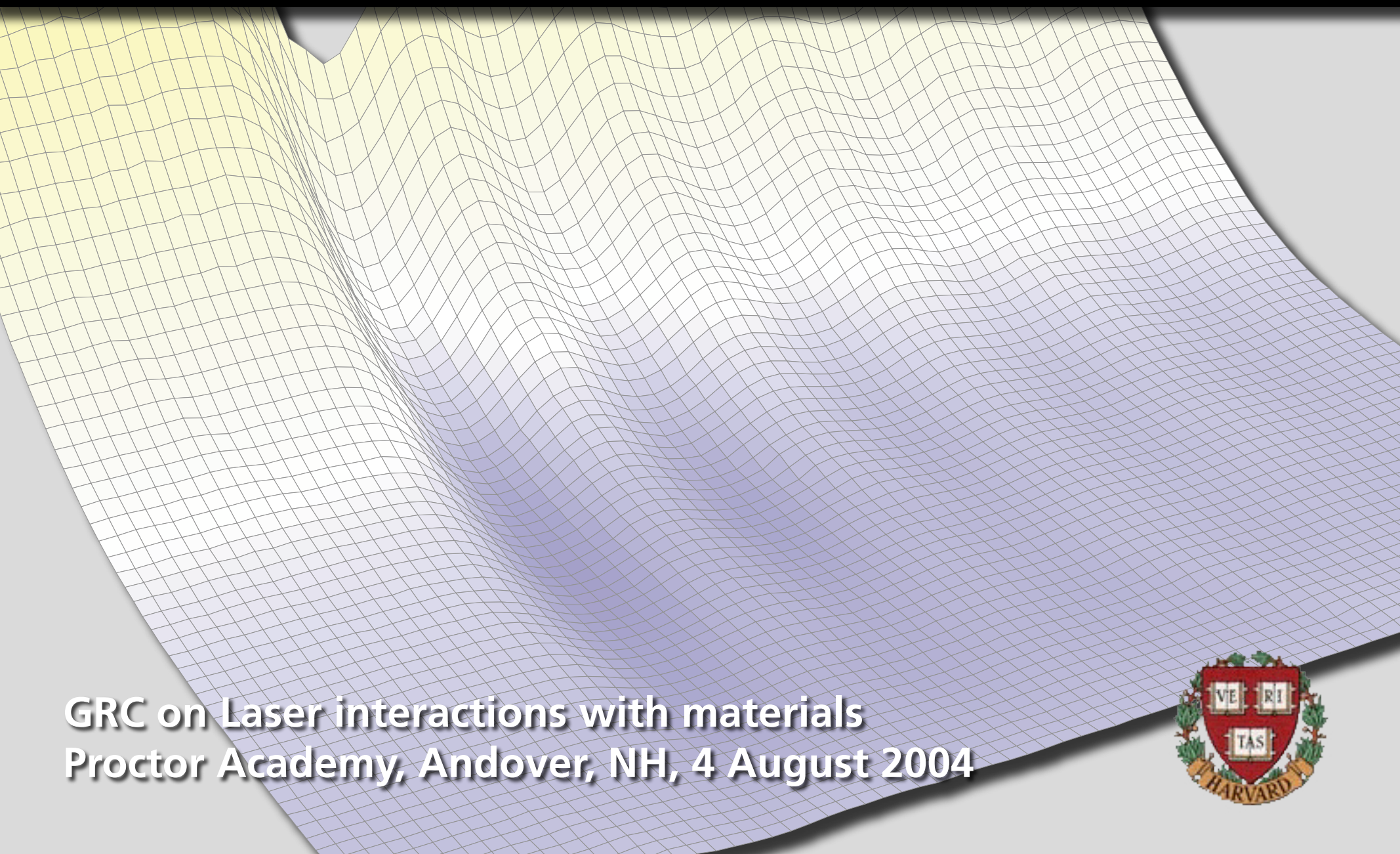
Materials phase transformations induced by ultrafast laser pulses

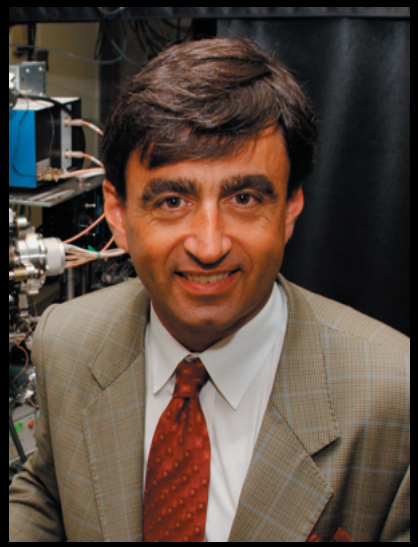


GRC on Laser interactions with materials
Proctor Academy, Andover, NH, 4 August 2004

Chris Roeser

Maria Kandyla

Arantza Mendioroz



Eric Mazur

and also....

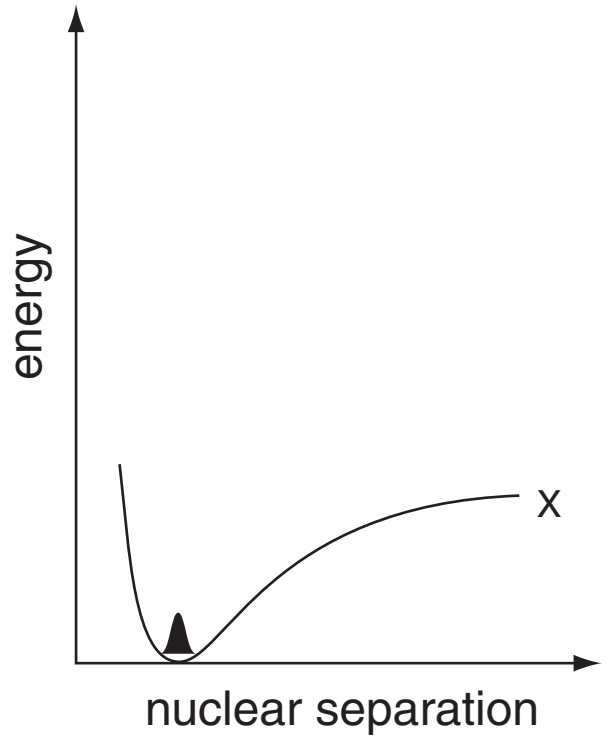
Albert Kim
Paul Callan
Eli Glezer
Li Huang
Yakir Siegal
Prakriti Tayalia
Jason Orcutt

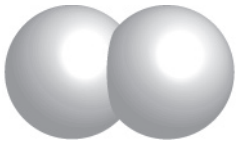
Dr. Peter Grosse (Aachen)
Dr. Paul Tangney (Princeton)
Prof. Steven Fahy (Cork)
Nick Choly (Harvard University)
Prof. Tim Kaxiras (Harvard University)

P.T.B.

Introduction

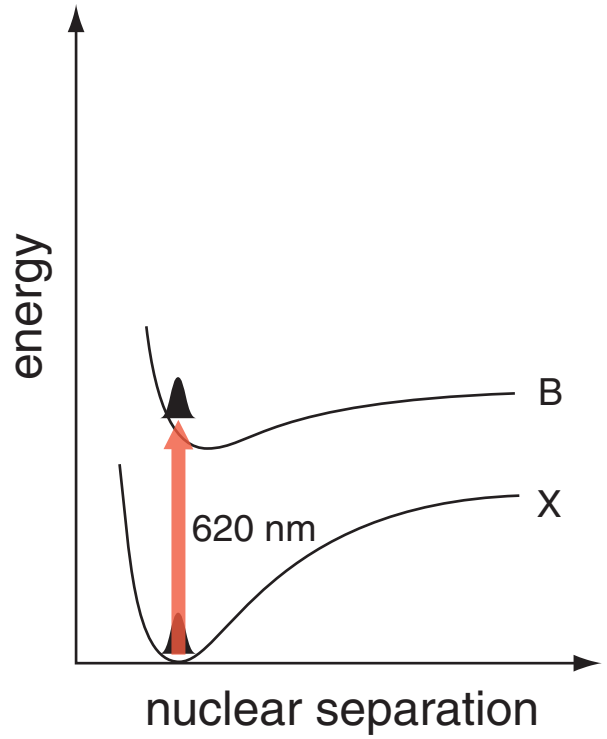
iodine

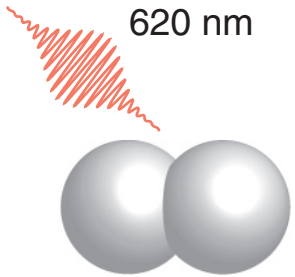




Introduction

iodine





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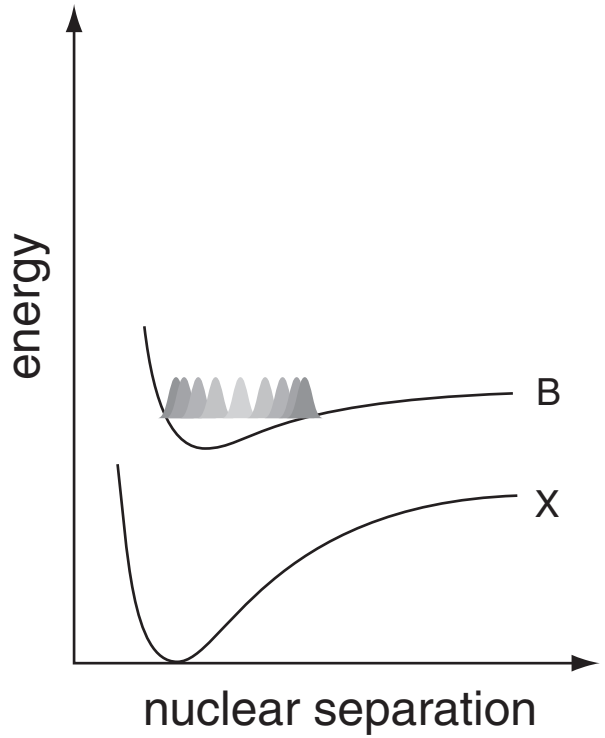
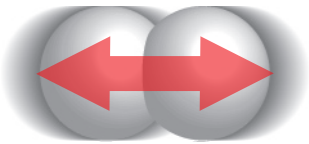
iodine





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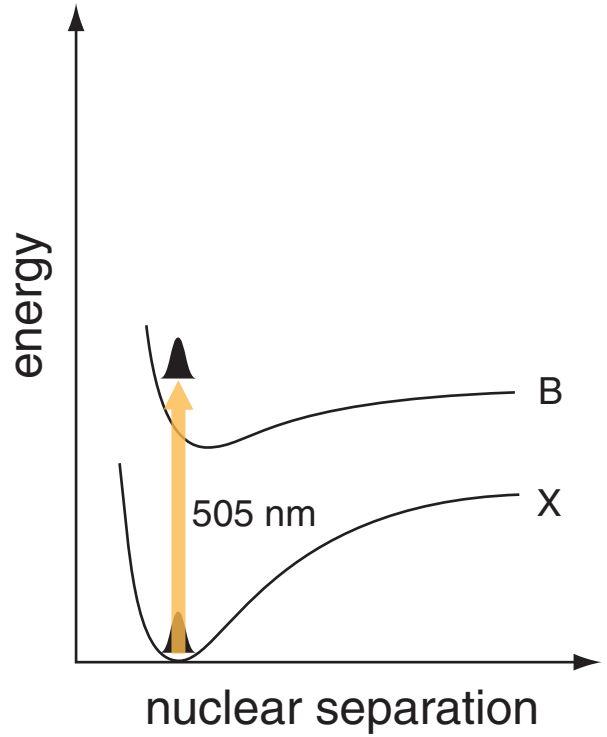
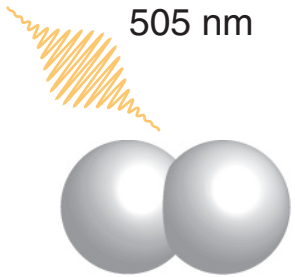
iodine





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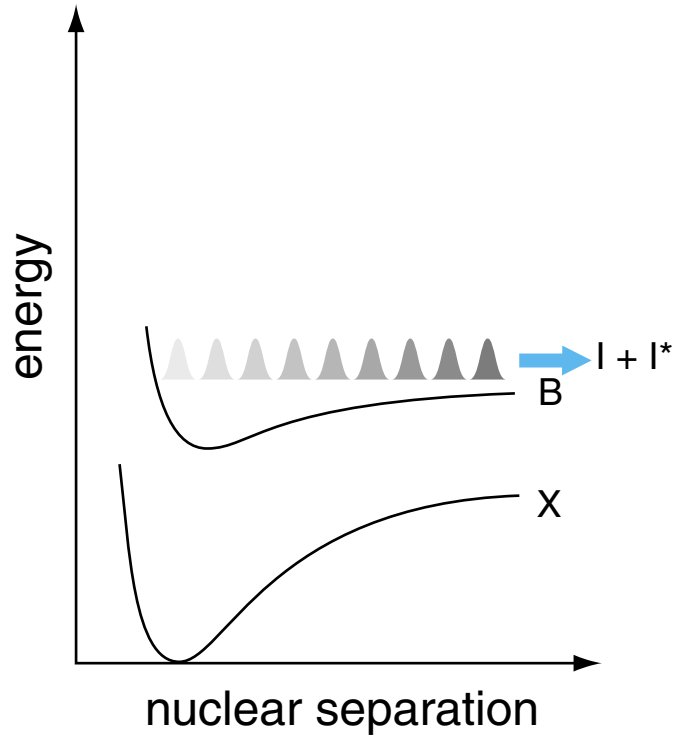
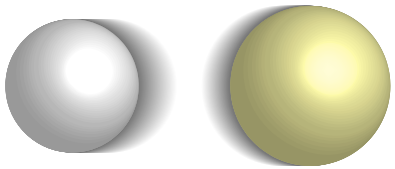
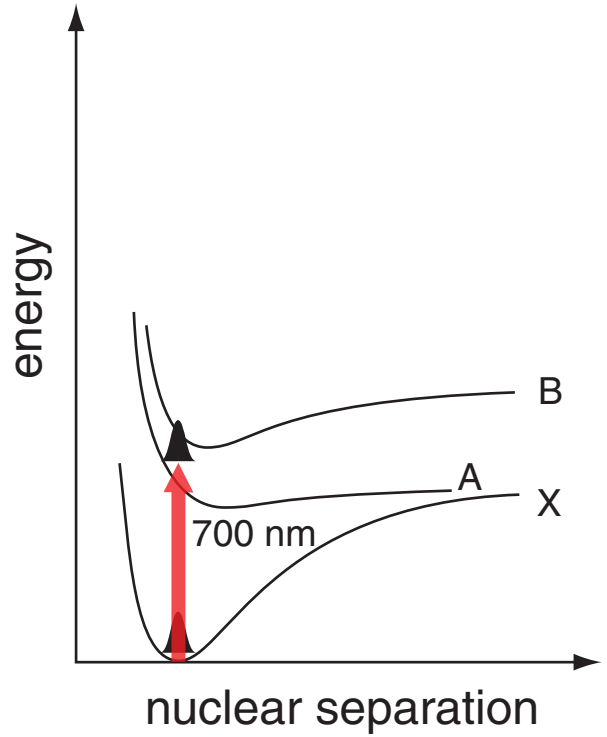
iodine

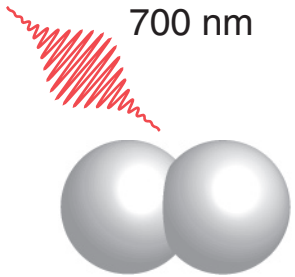




Introduction

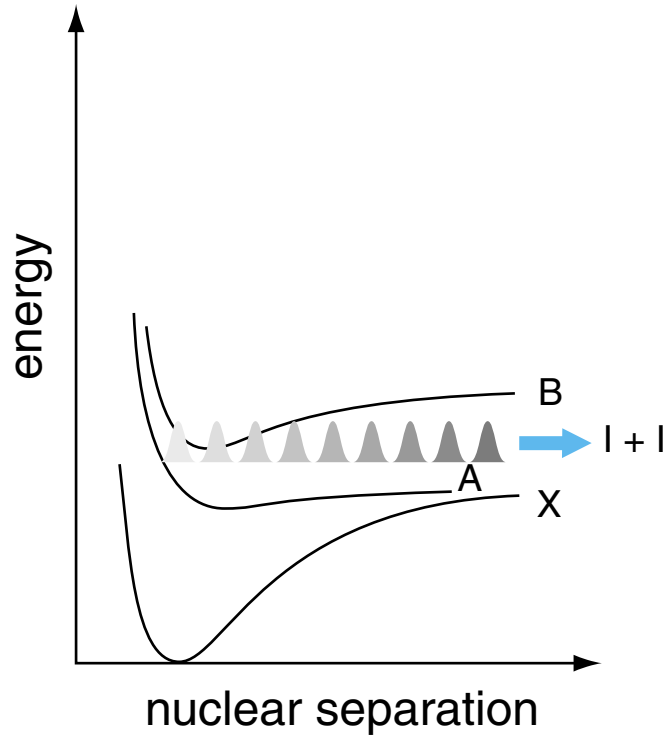
iodine

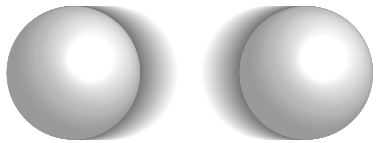




Introduction

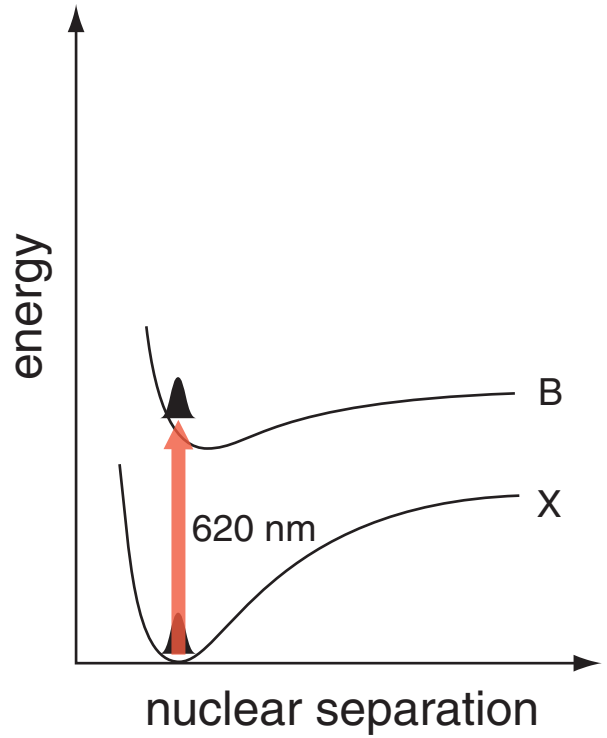
iodine





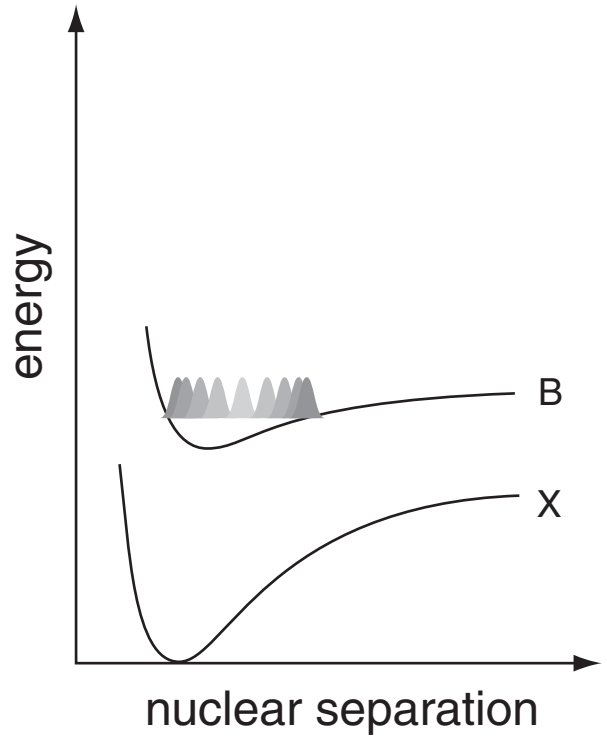
Introduction

probe wave packet dynamics



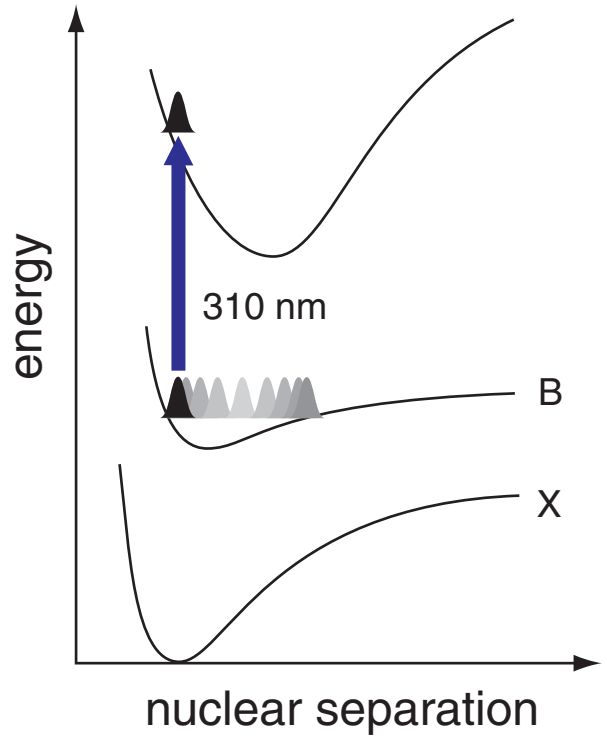
Introduction

probe wave packet dynamics



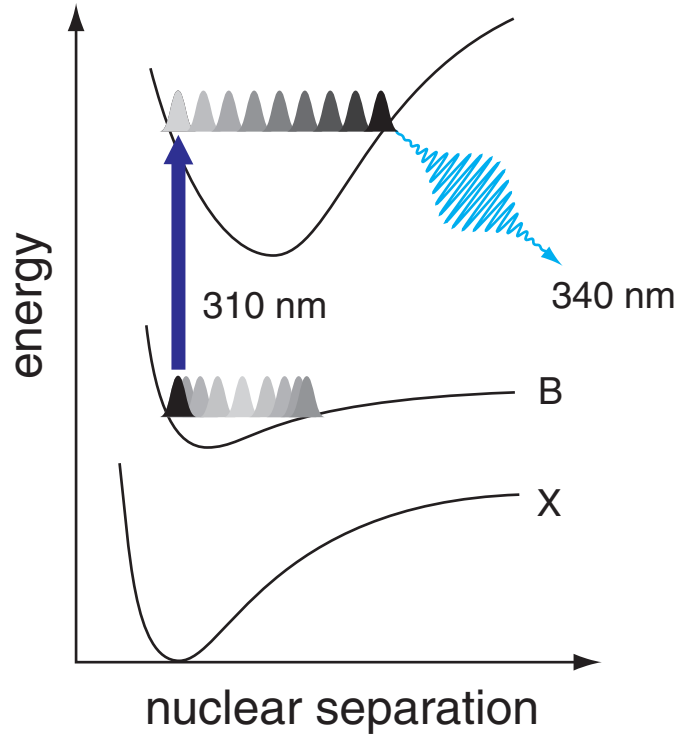
Introduction

probe wave packet dynamics



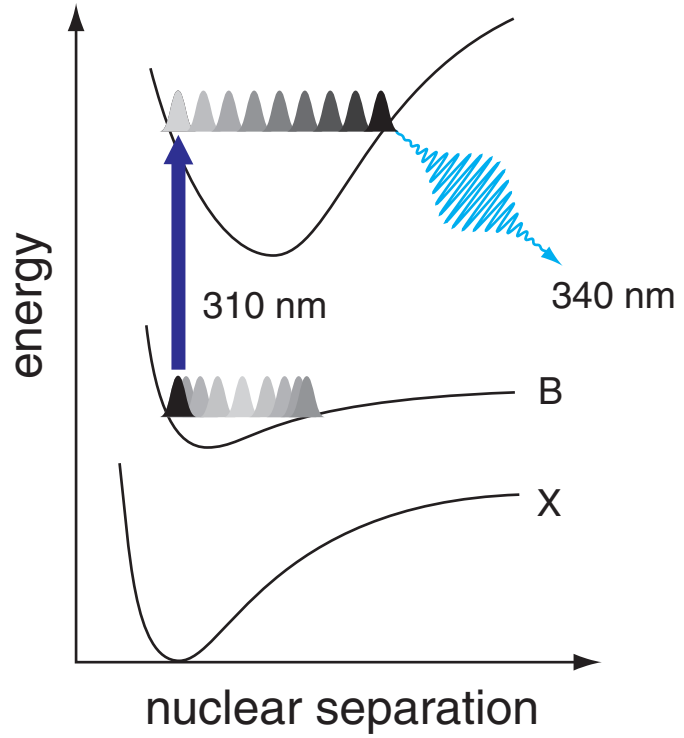
Introduction

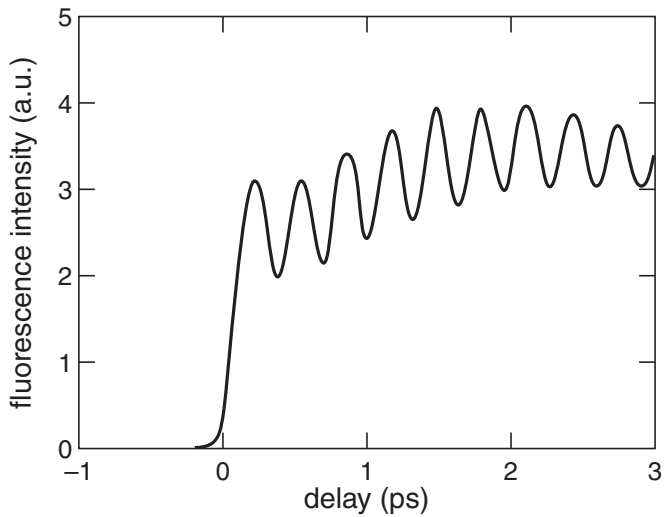
probe wave packet dynamics



Introduction

probe wave packet dynamics





Bowman *et al.*, *Chem. Phys. Lett.* 161, 297 (1989)

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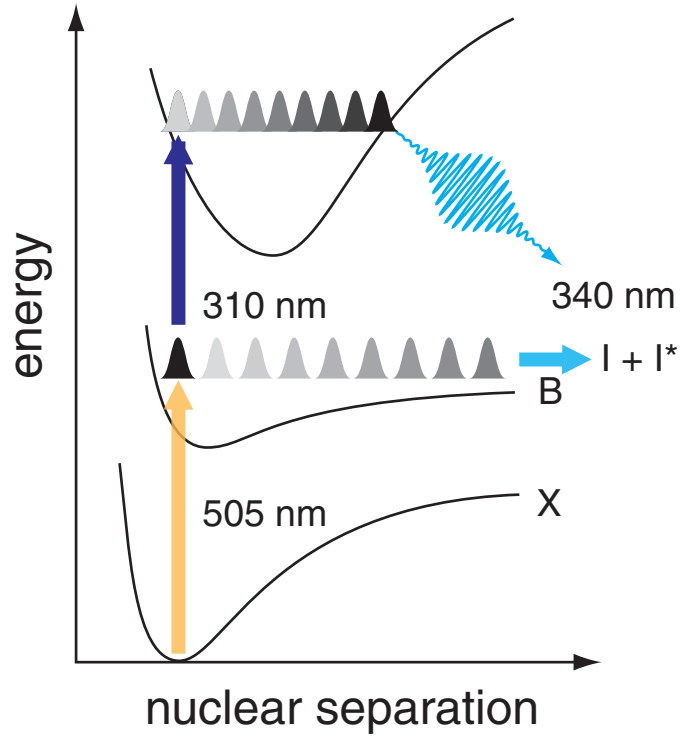
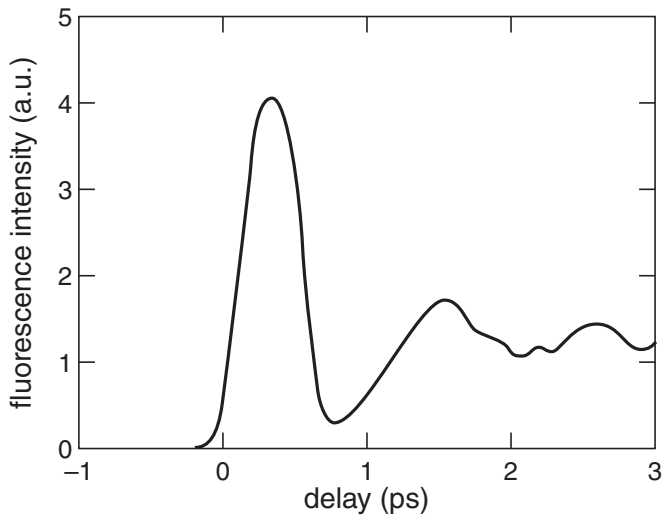
probe wave packet dynamics





Bowman *et al.*, *Chem. Phys. Lett.* 161, 297 (1989)

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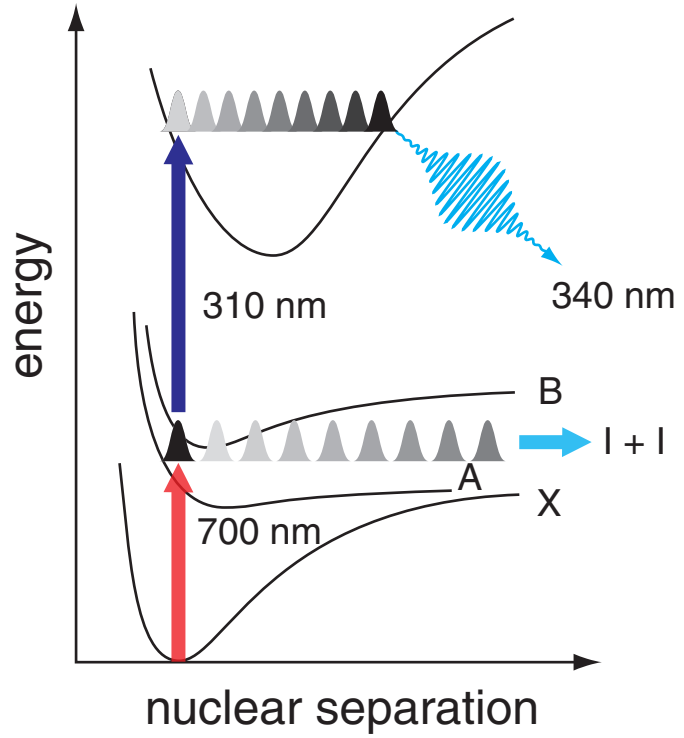
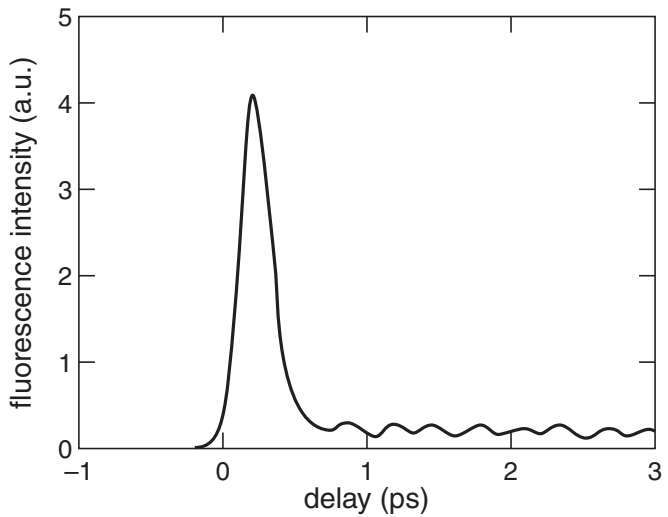
probe wave packet dynamics





Bowman *et al.*, *Chem. Phys. Lett.* 161, 297 (1989)

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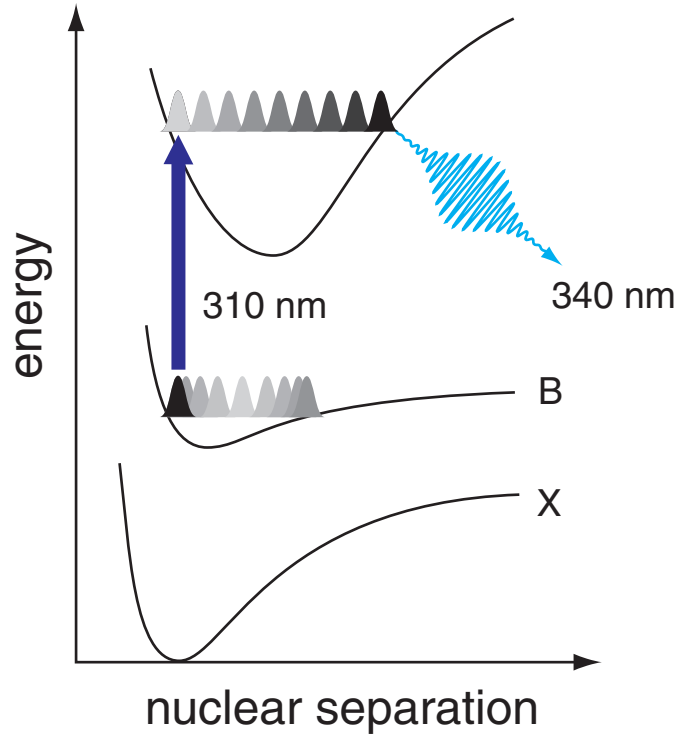
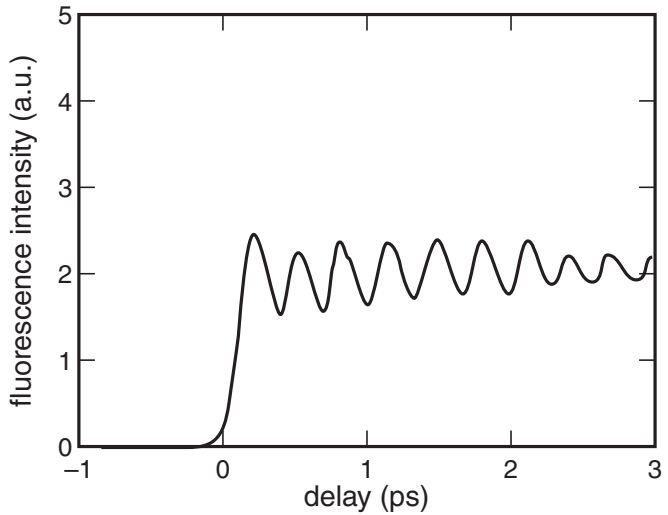
wave packet control

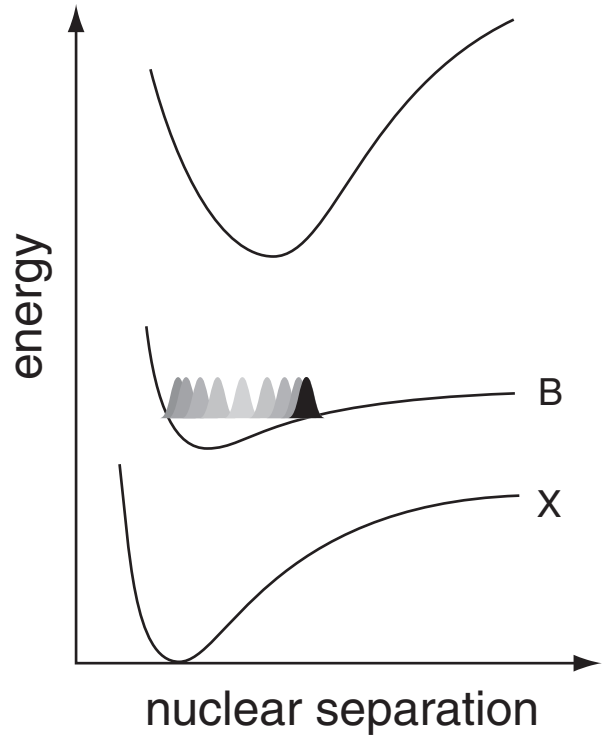


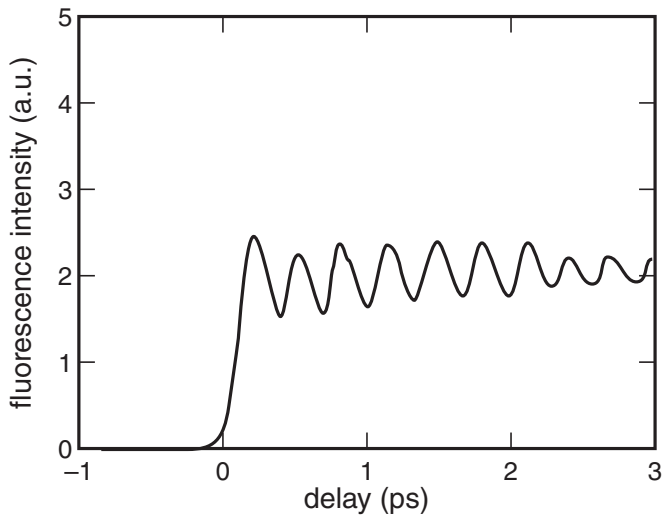


Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

Introduction

wave packet control





Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

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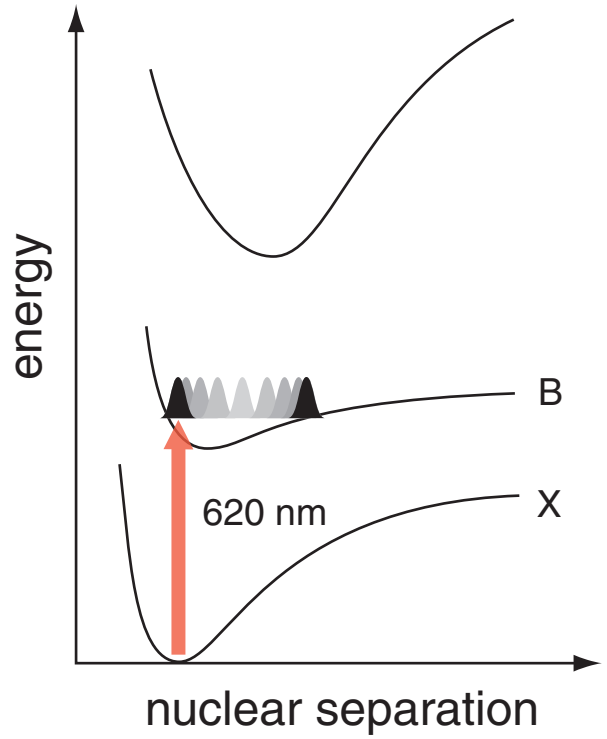
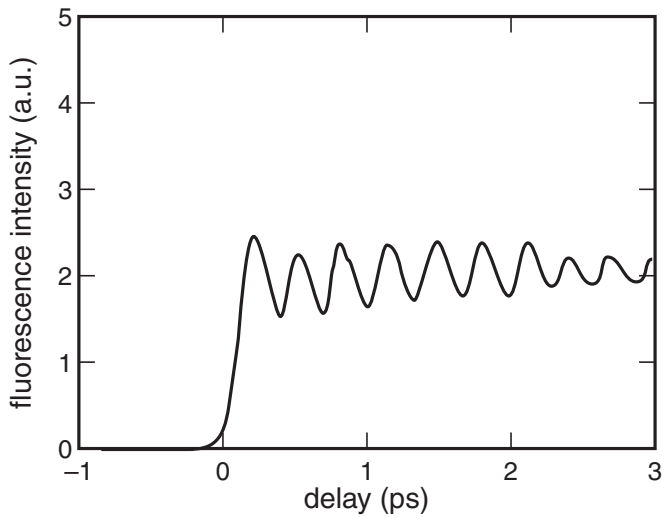
phase delay: π

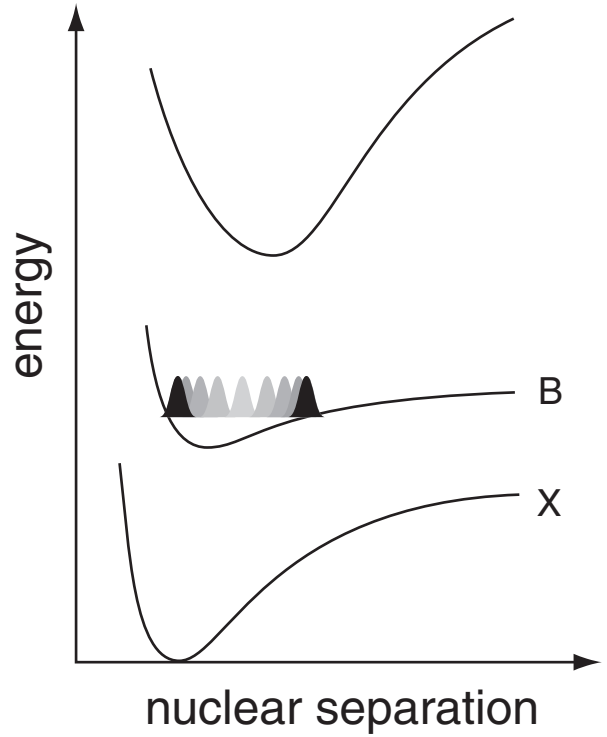


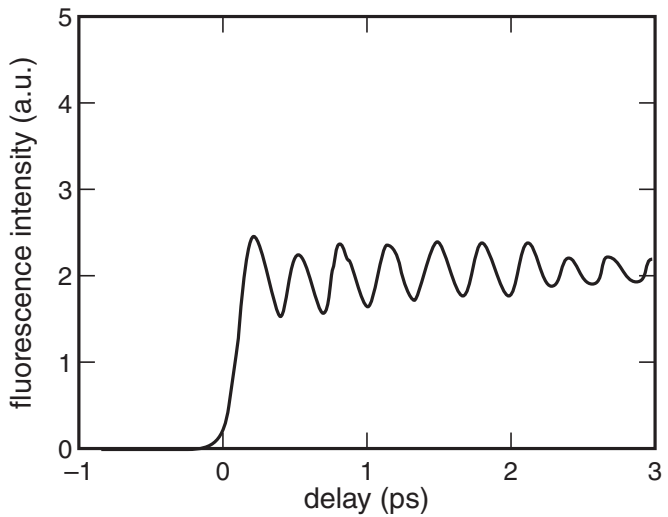


Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

Introduction

phase delay: π

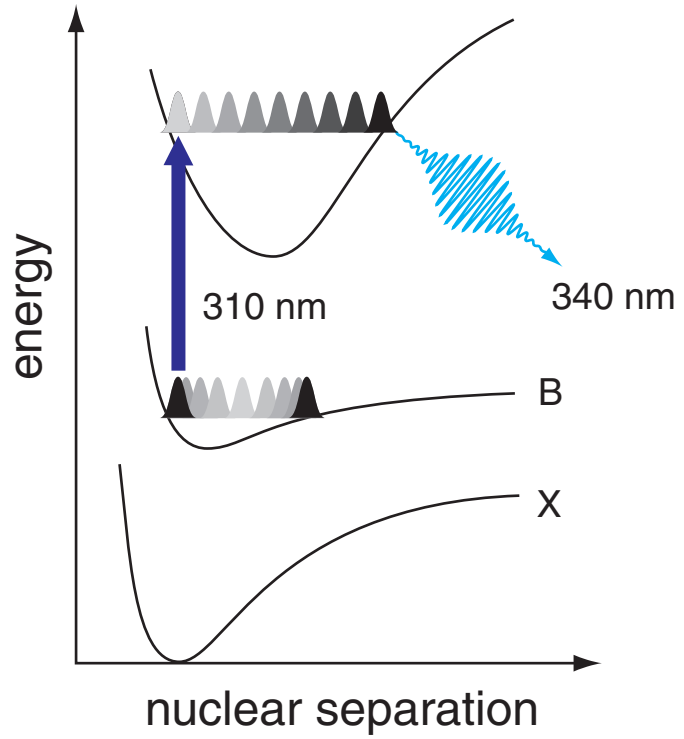


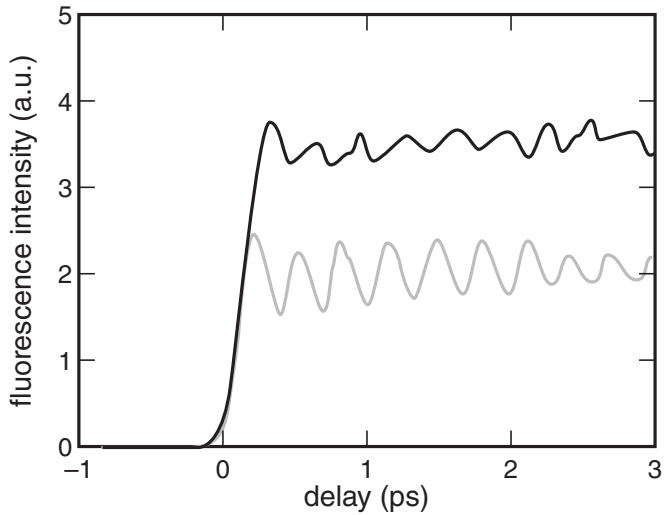


Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

Introduction

phase delay: π





Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

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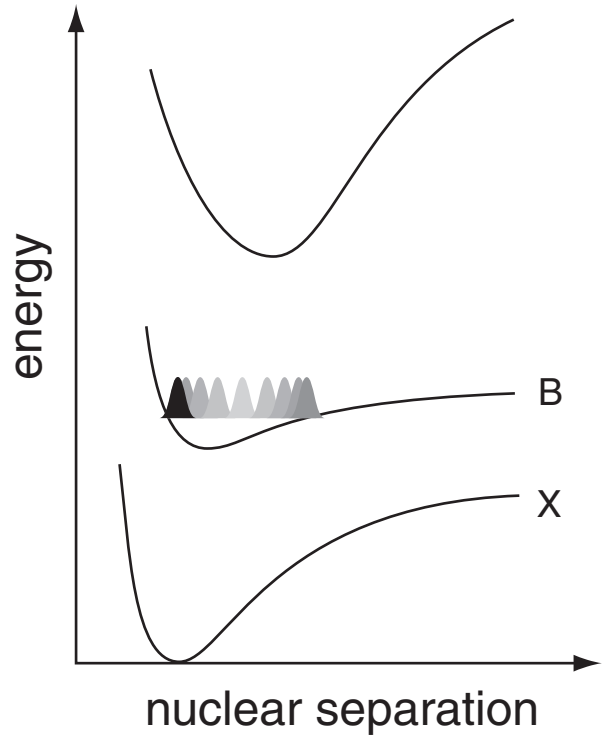
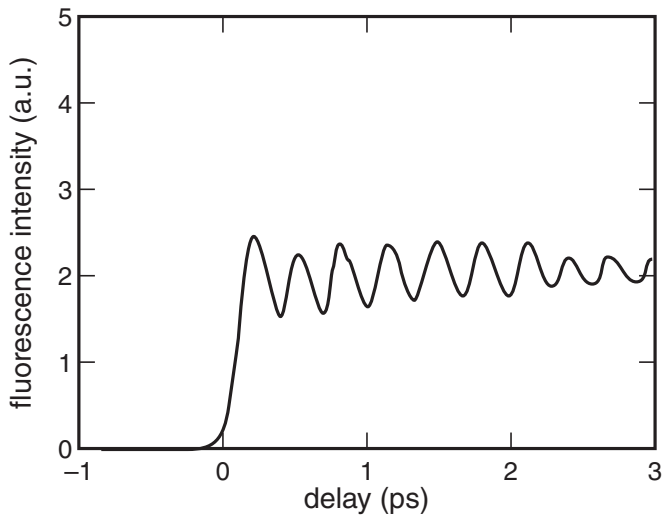
phase delay: 2π

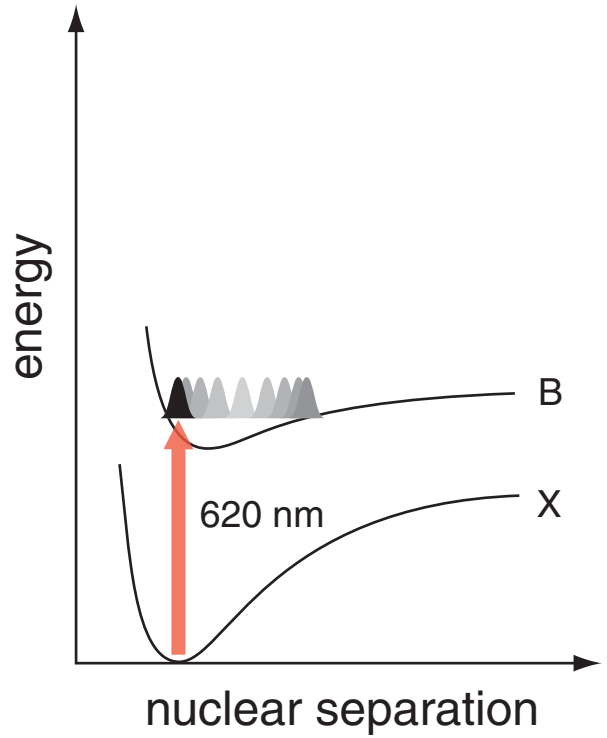


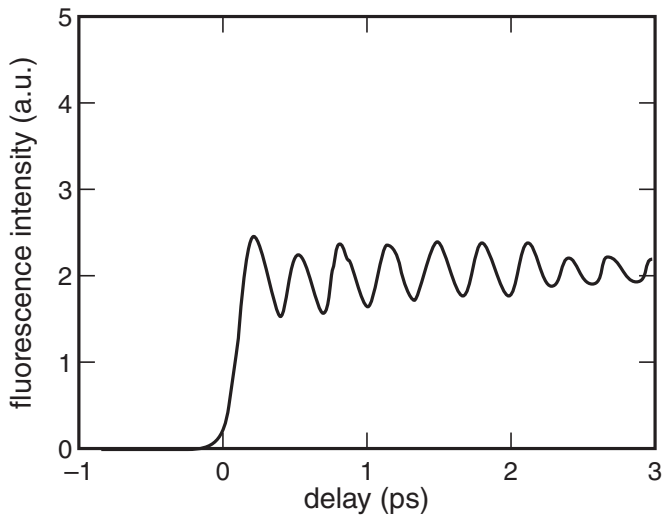


Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

Introduction

phase delay: 2π

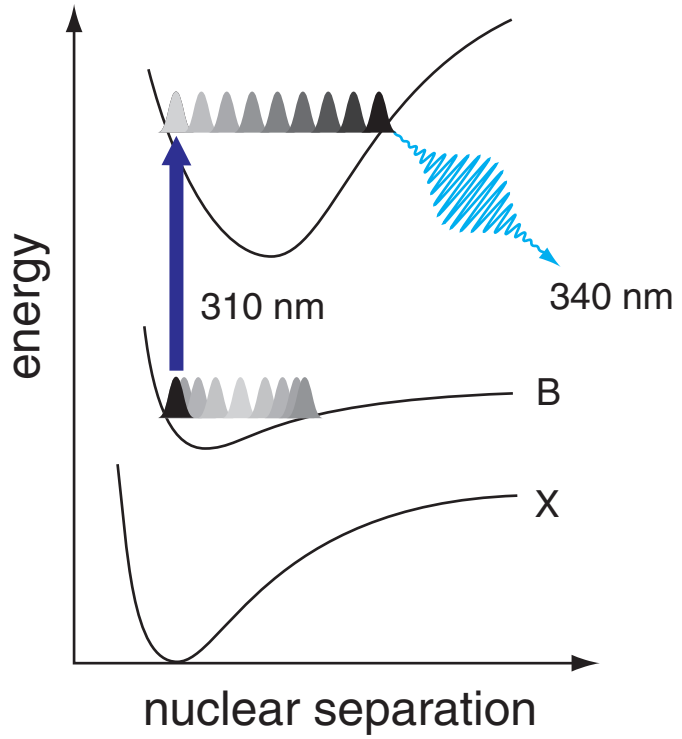


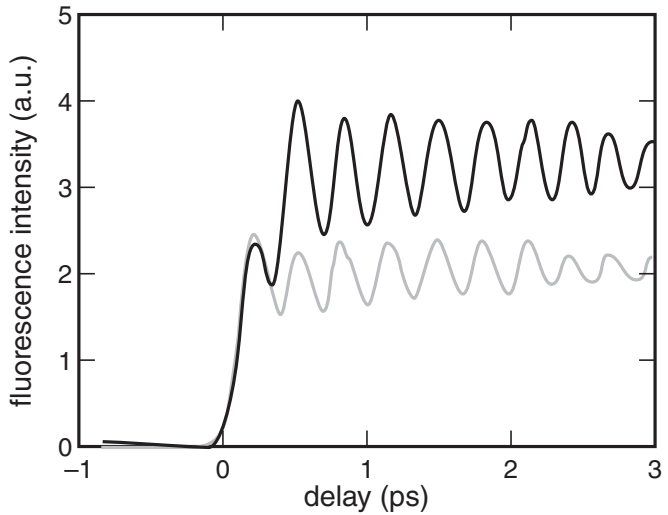


Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

Introduction

phase delay: 2π





Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

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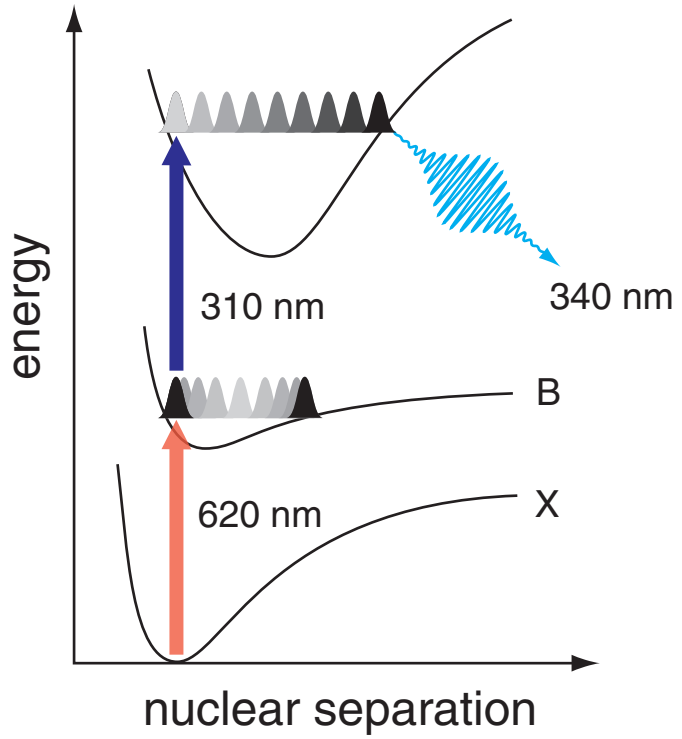
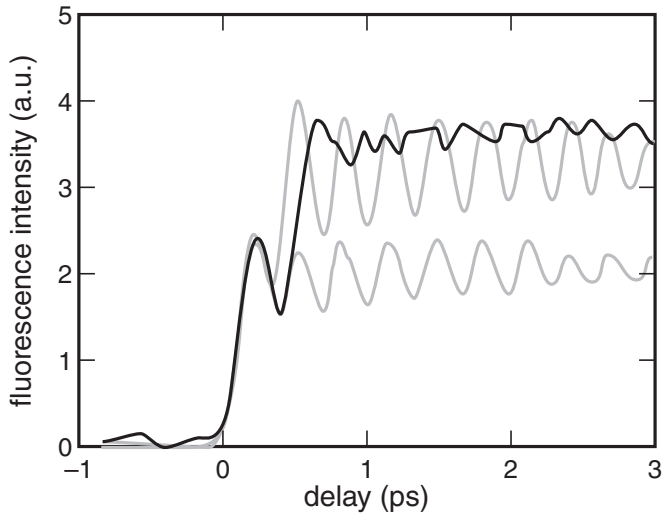
phase delay: 3π





Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

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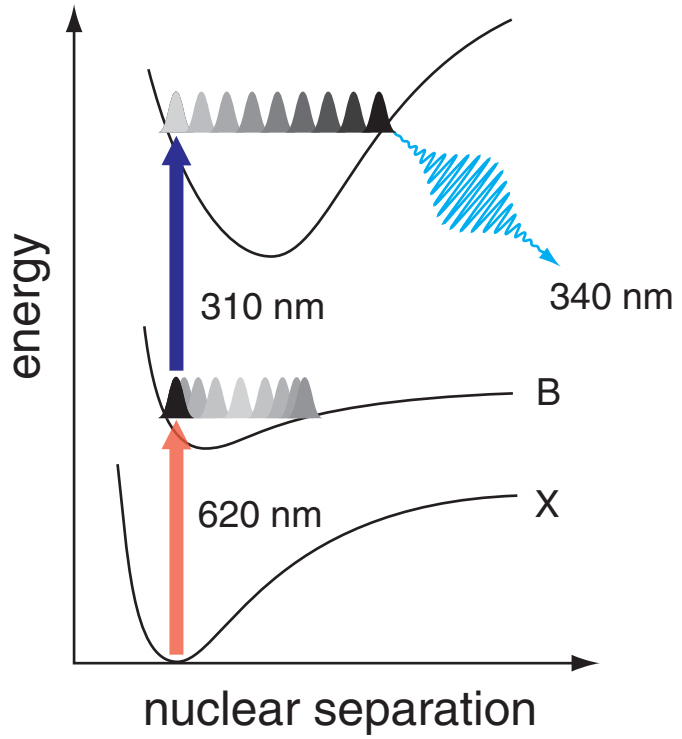
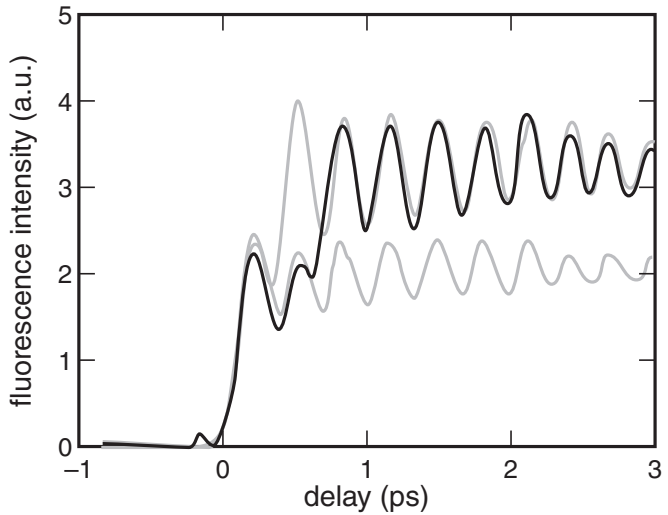
phase delay: 4π





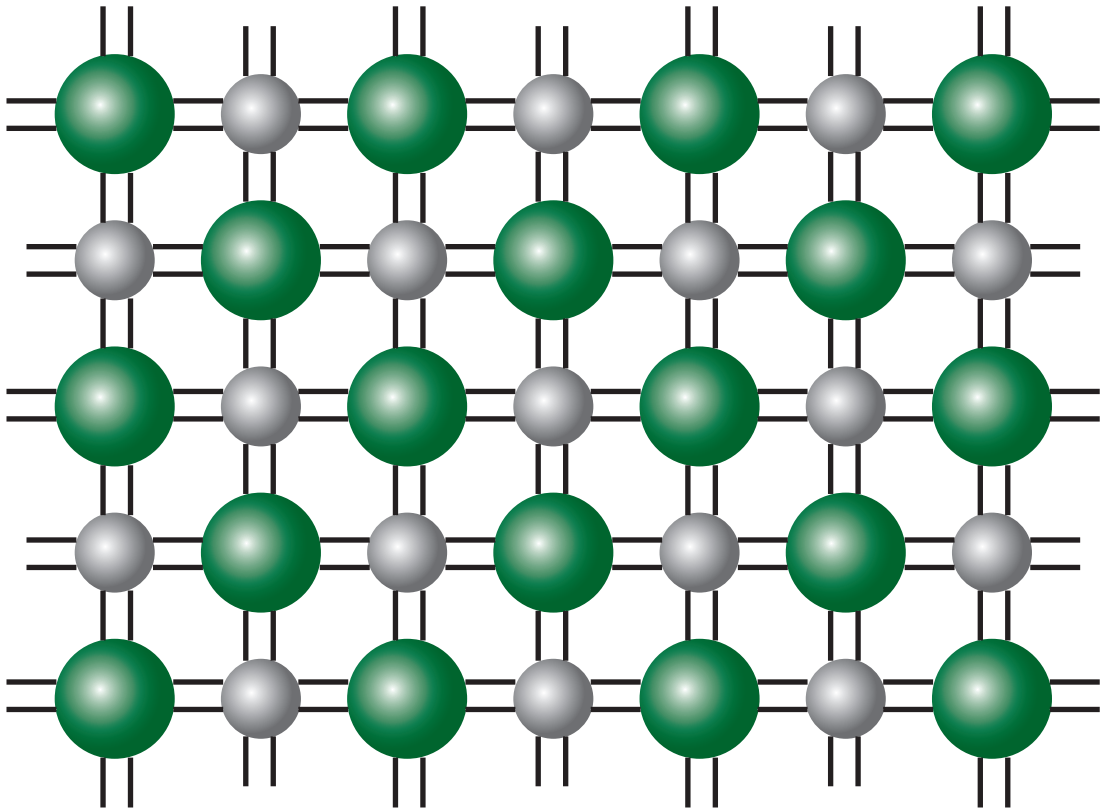
Gerdy *et al.*, *Chem. Phys. Lett.* 171, 1 (1990)

Introduction

limited control of chemical reactions demonstrated

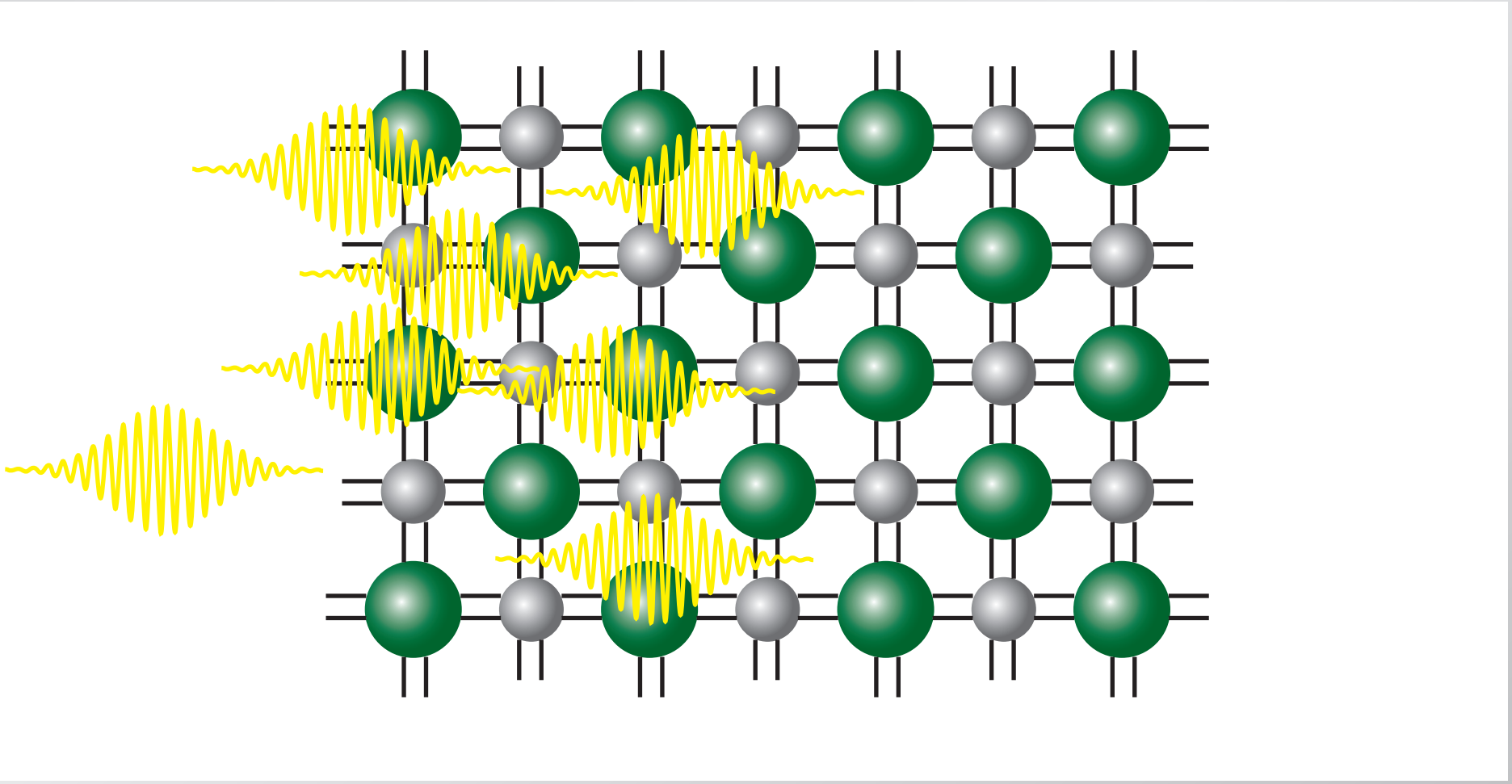
Introduction

can we optically control the state of a solid?



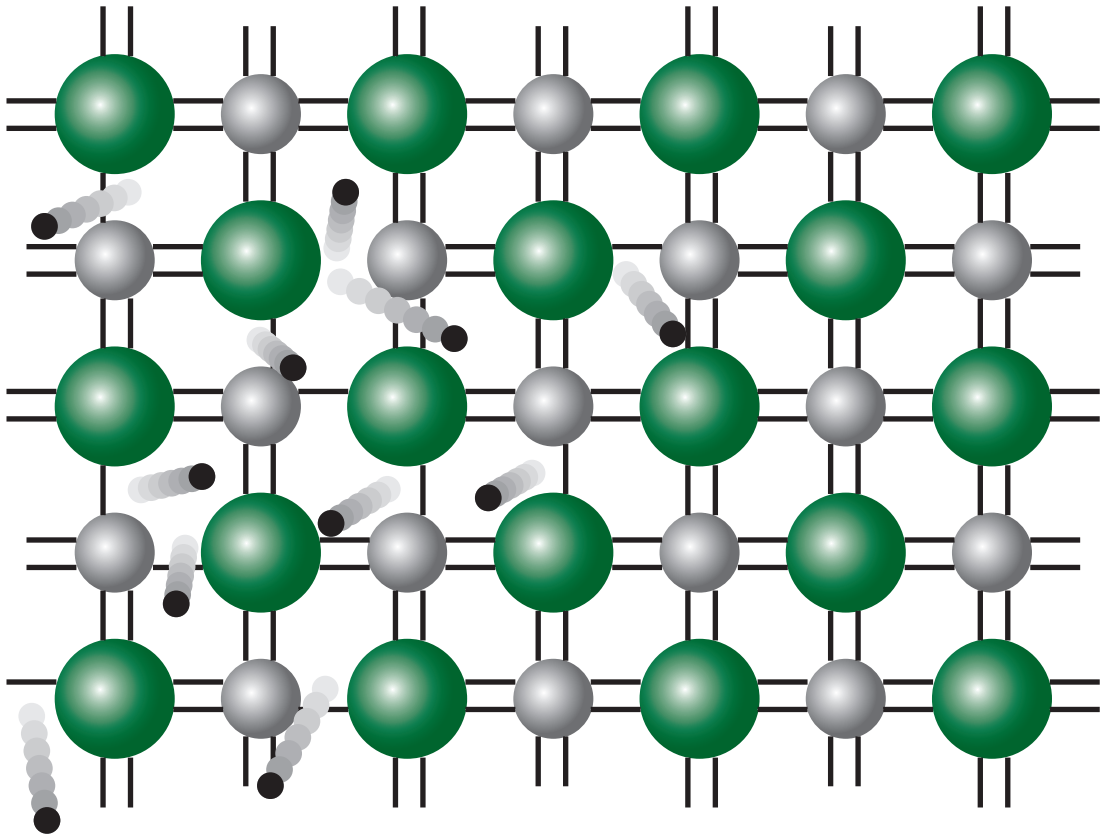
Introduction

photons excite valence electrons…



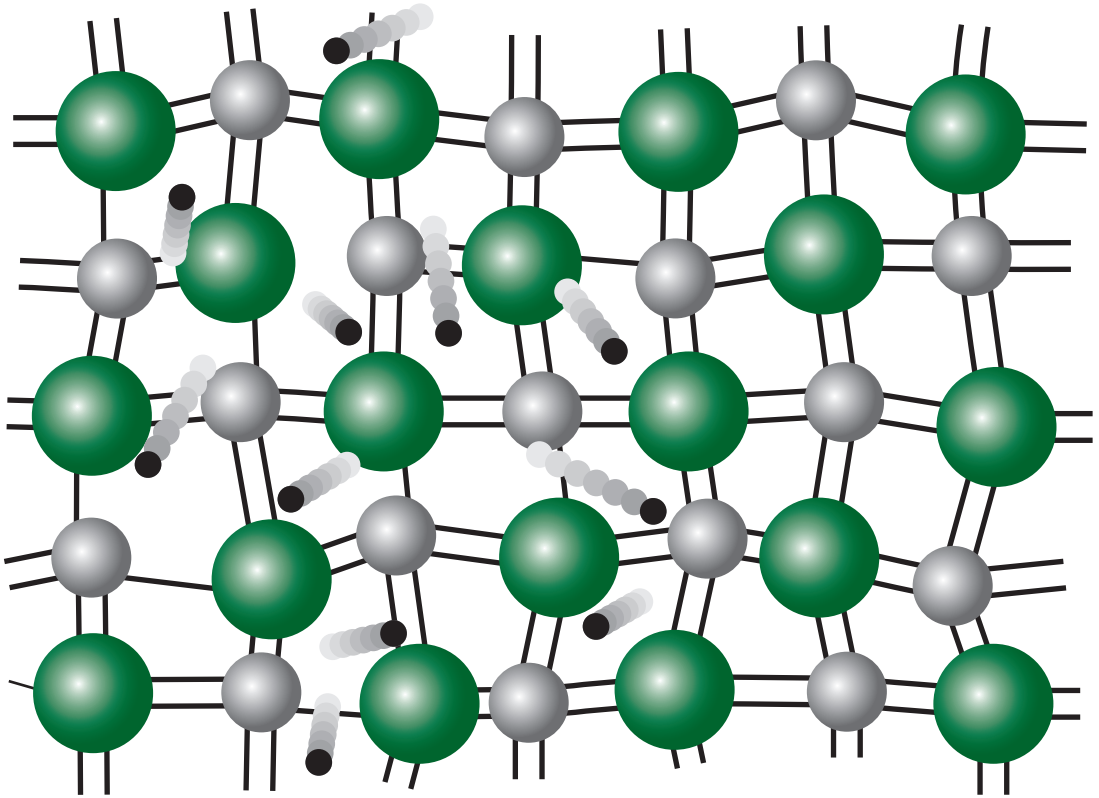
Introduction

…and create free carriers…



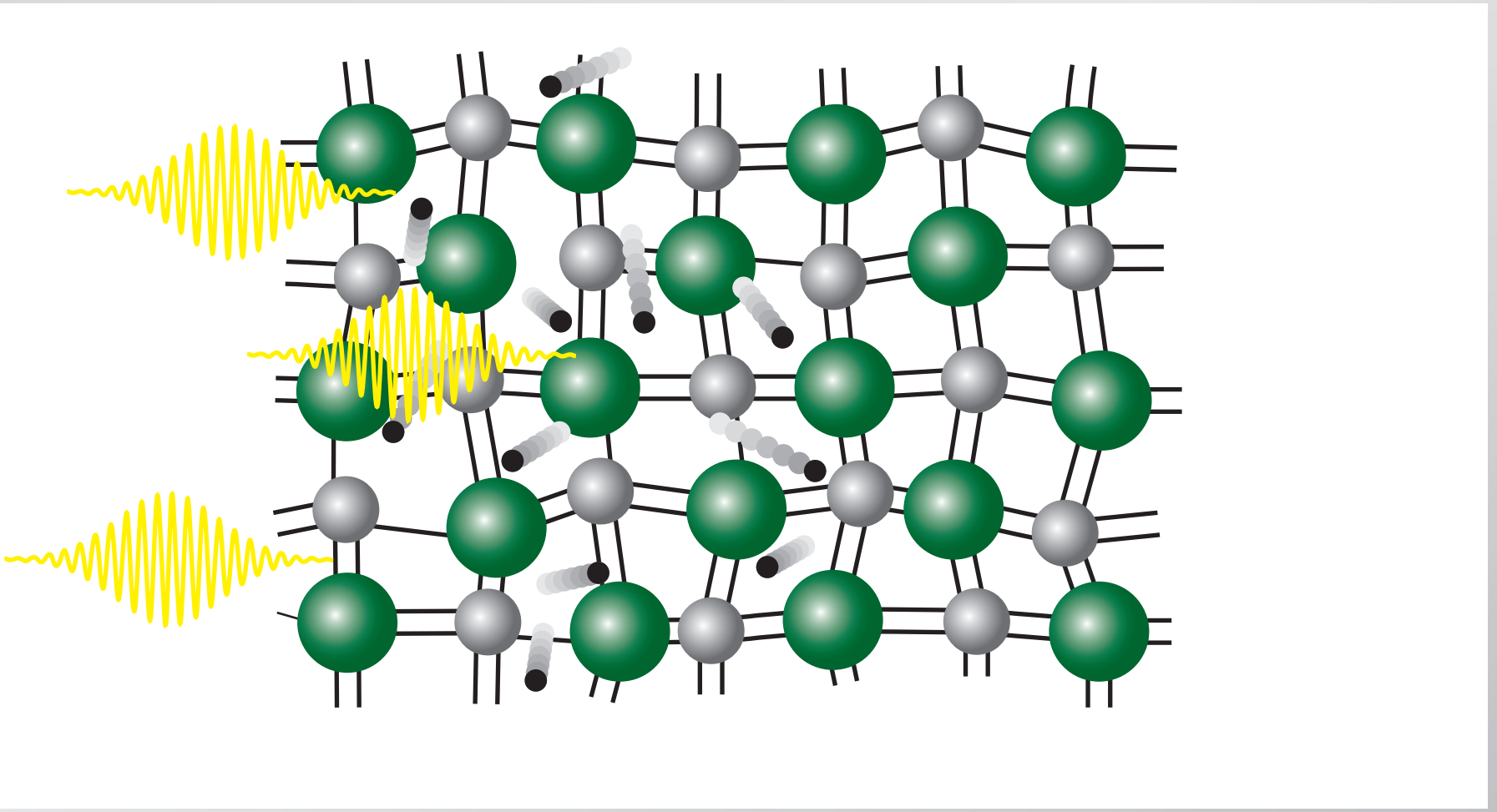
Introduction

…causing electronic and structural changes…



Introduction

…which we detect with a second laser pulse.



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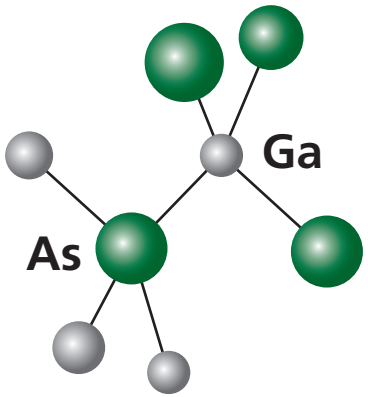
structure



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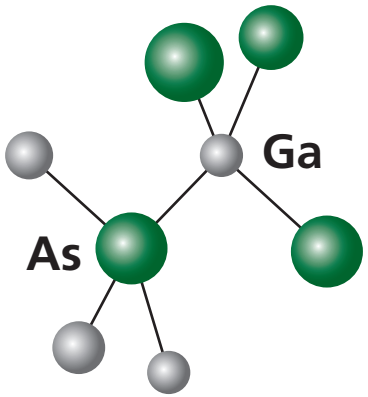
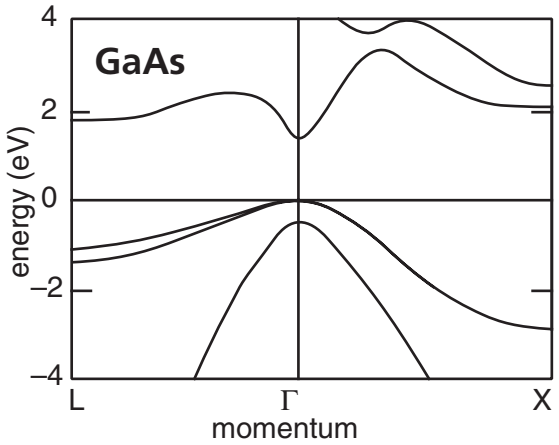
structure

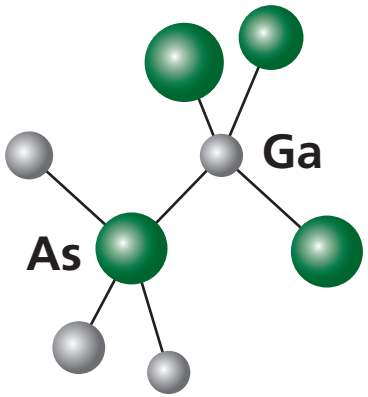


band structure



Introduction

structure



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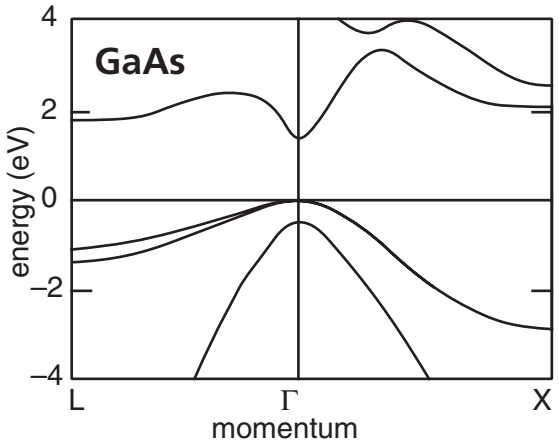

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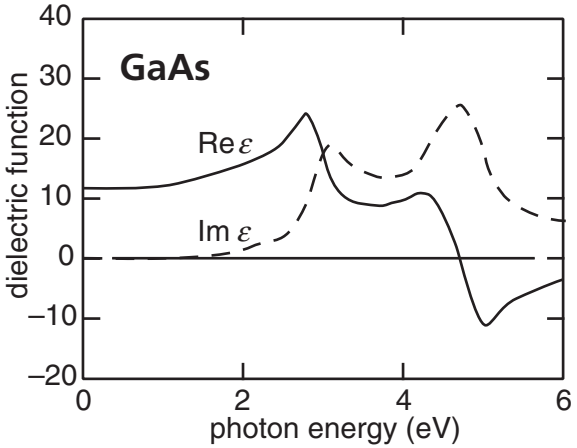

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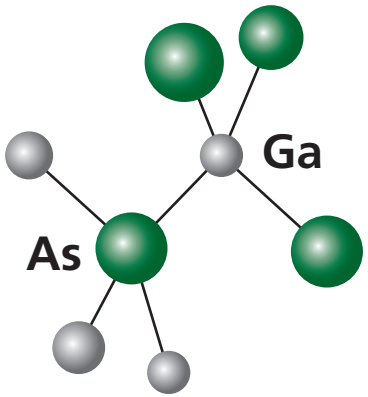

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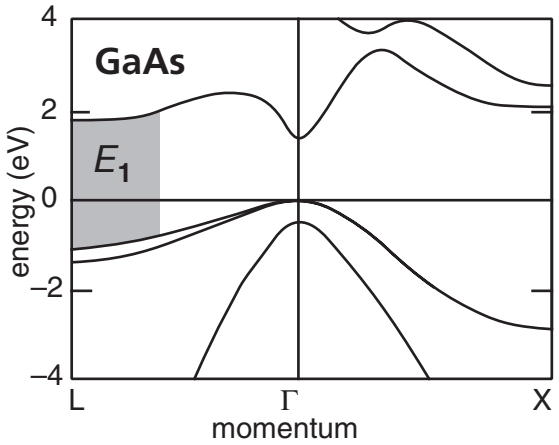

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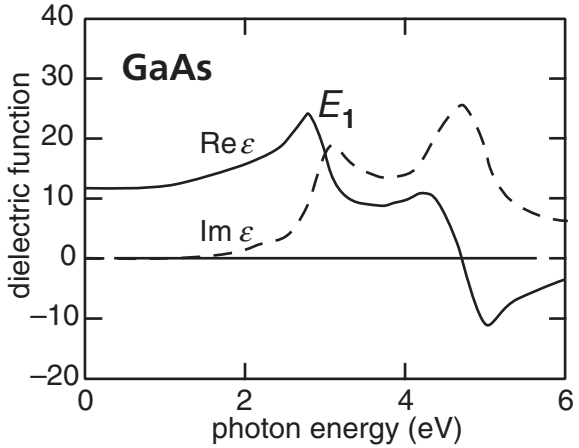

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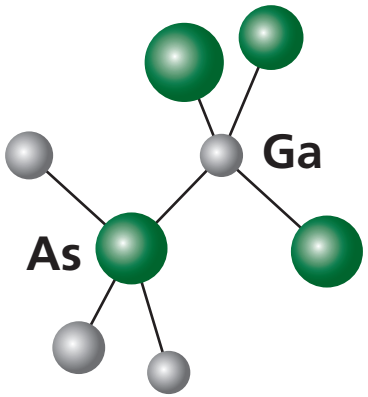

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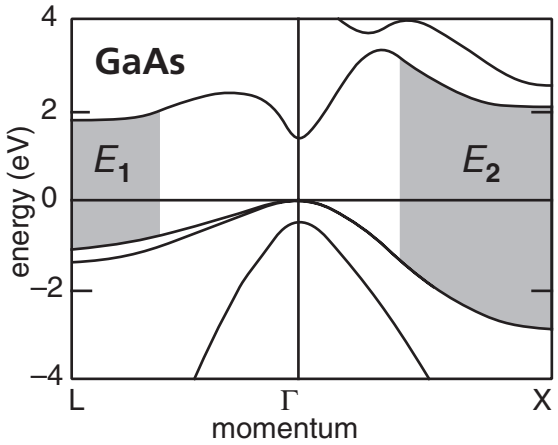

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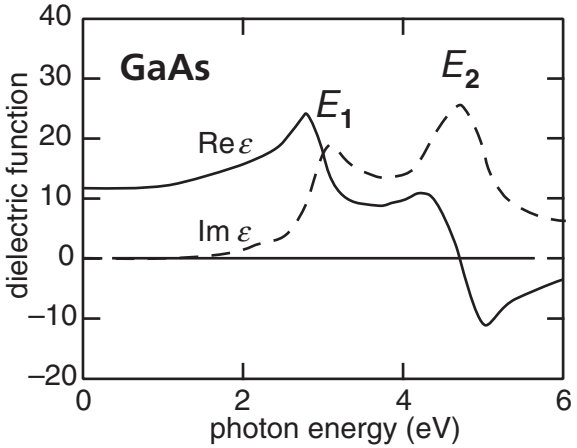

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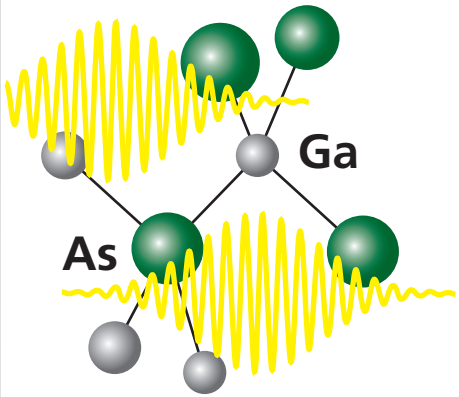

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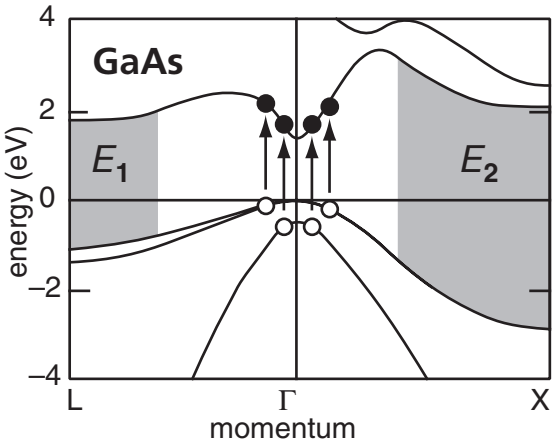

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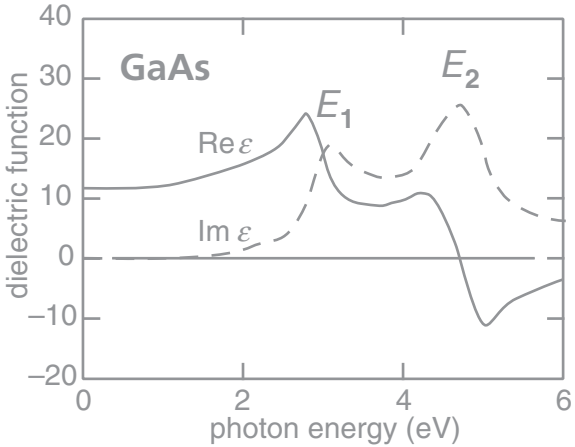

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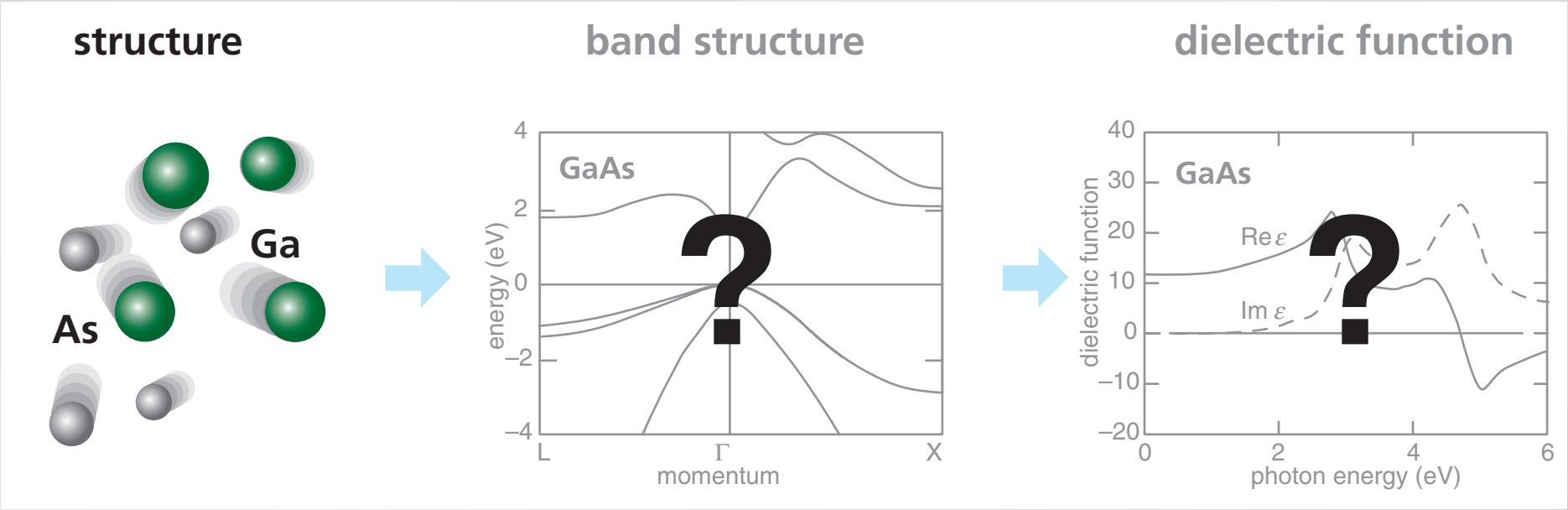

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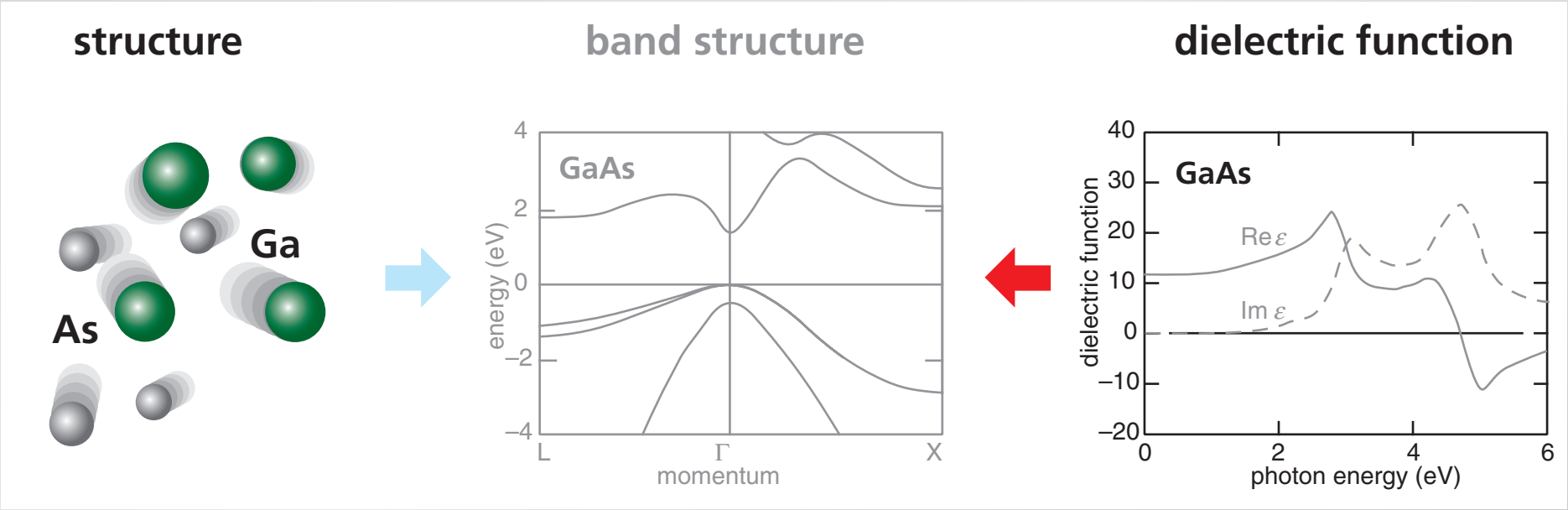

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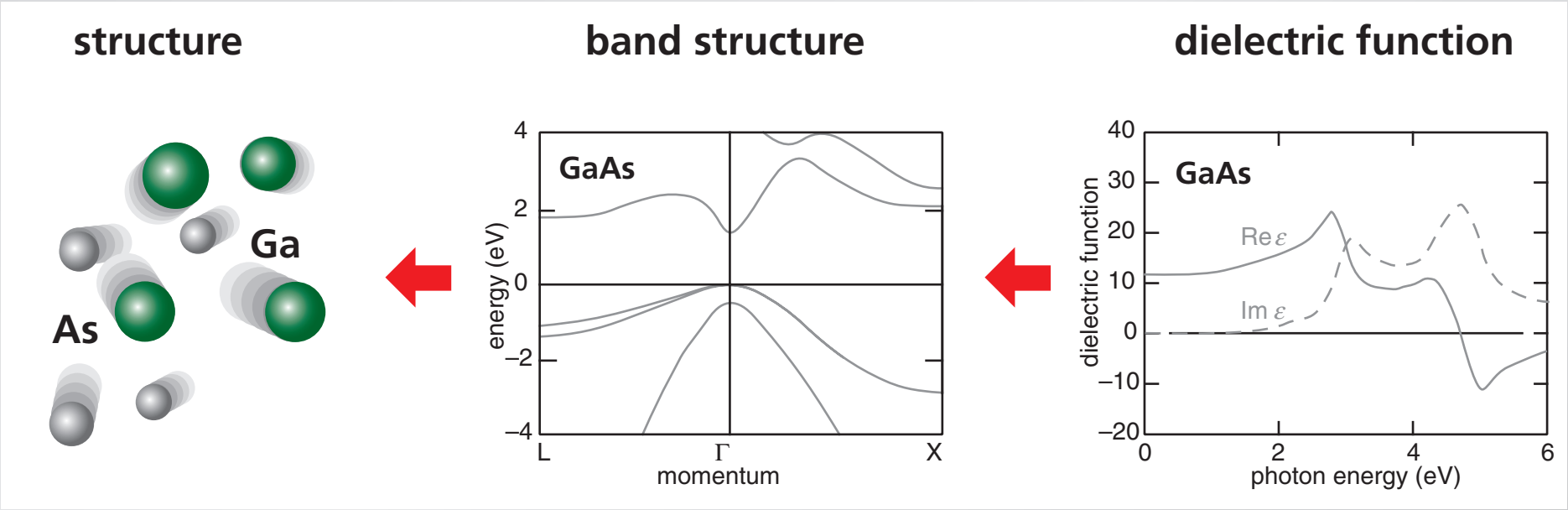

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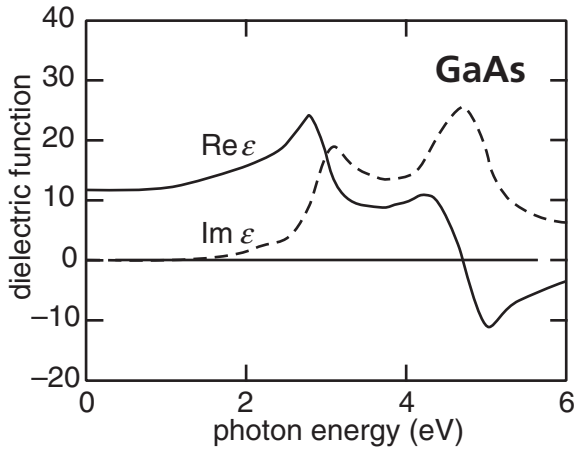

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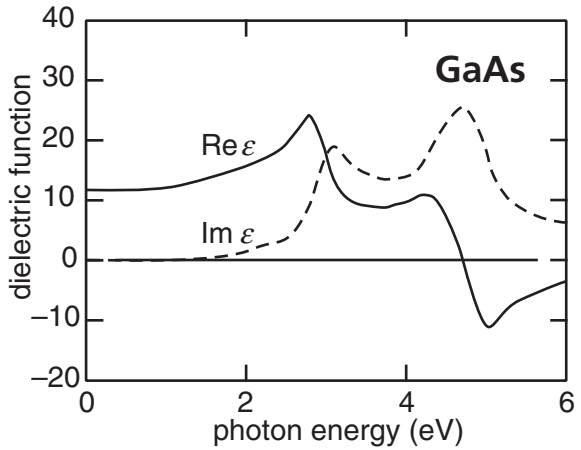

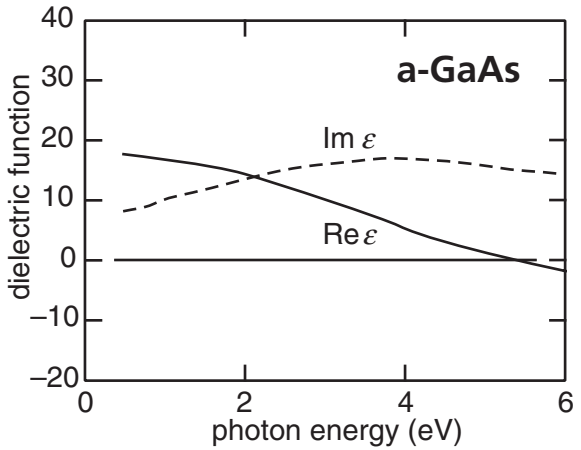

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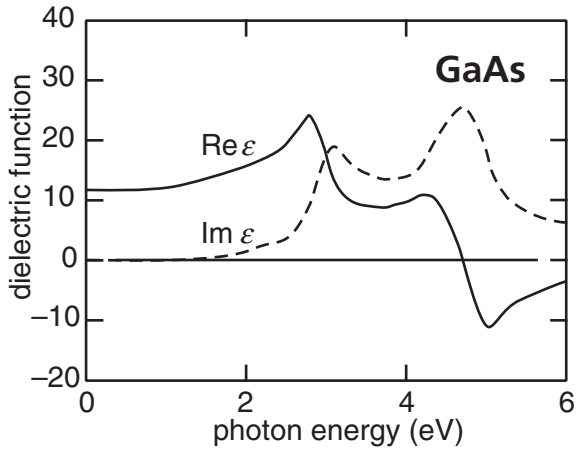

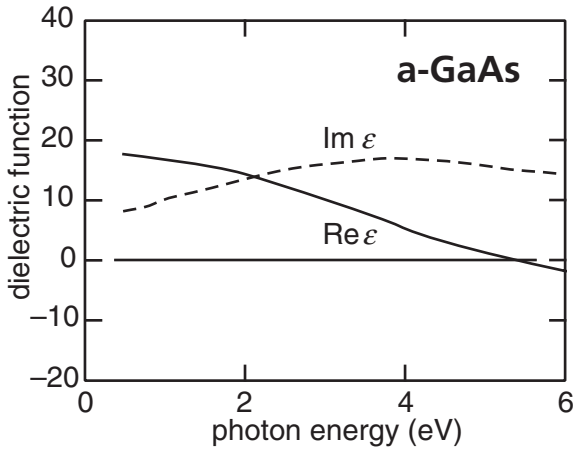

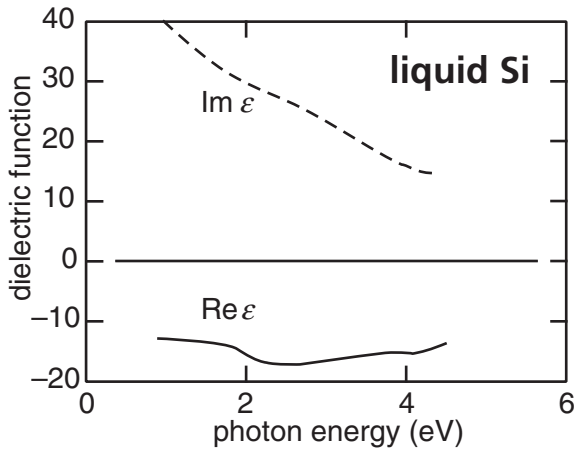

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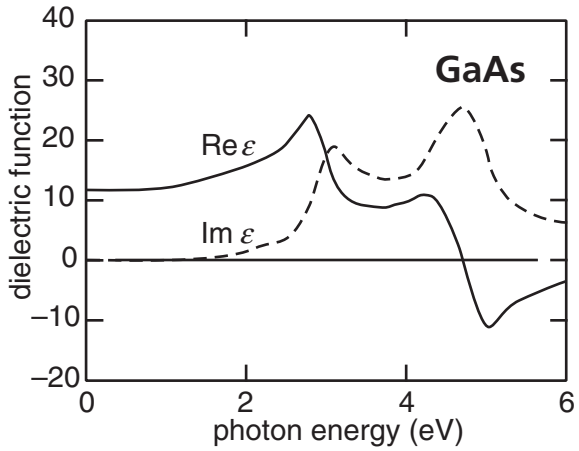
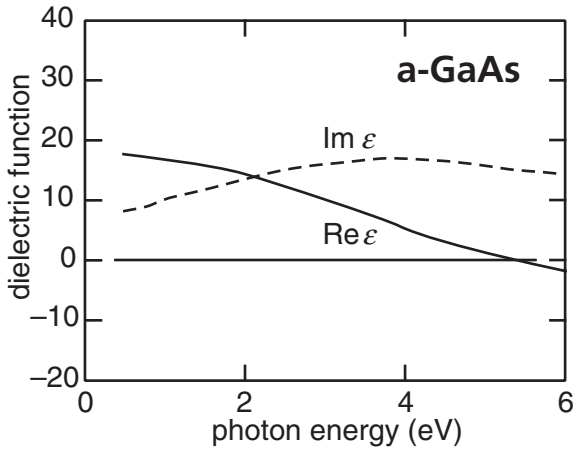
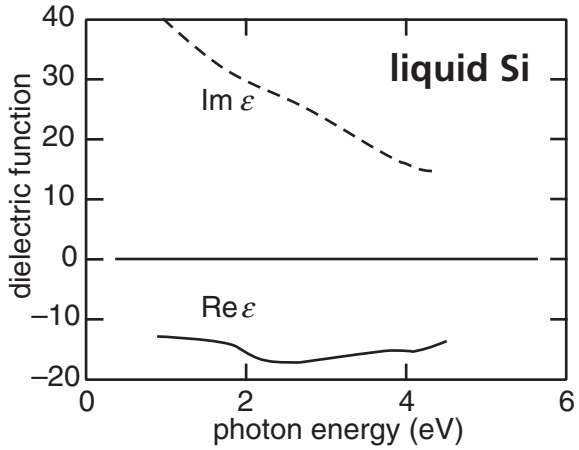
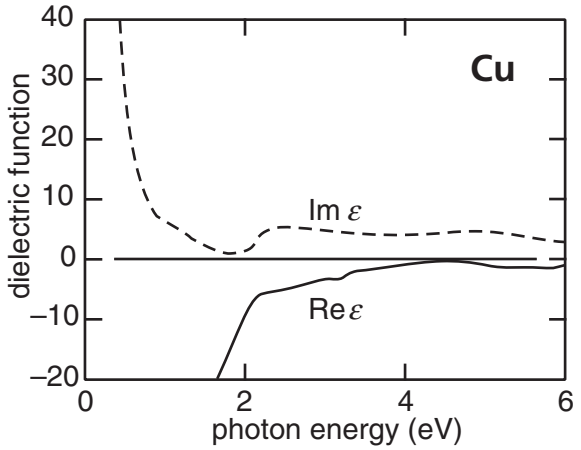






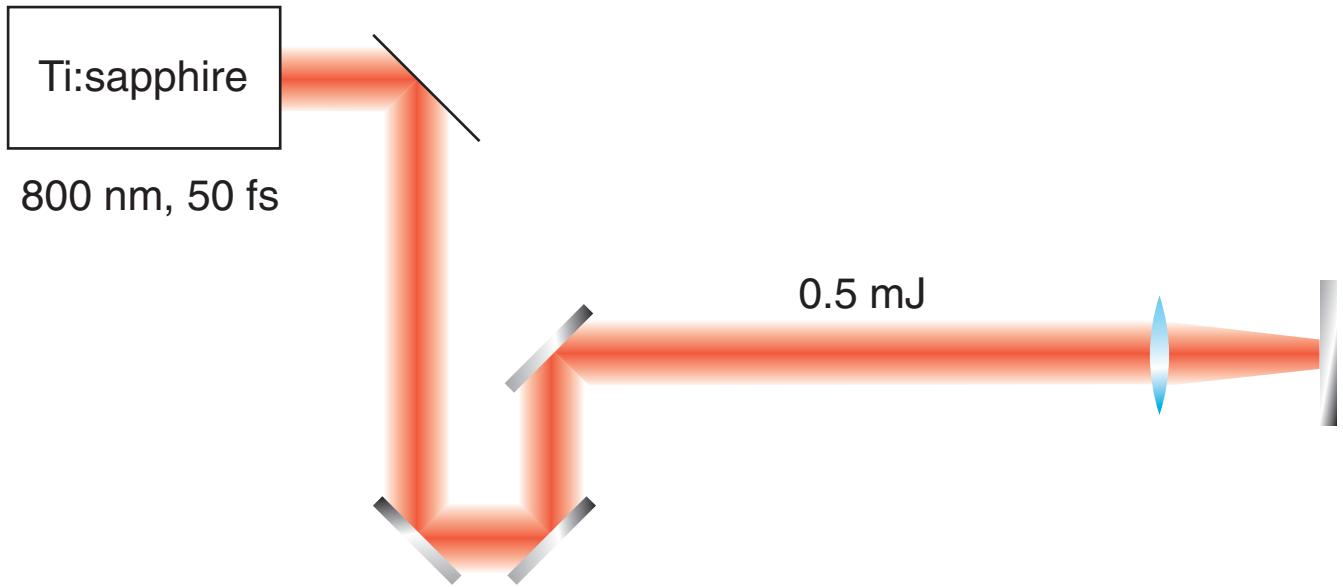


Outline

- experimental
- coherent phonons
- optical control

Experimental

time-resolved dual-angle reflectometry



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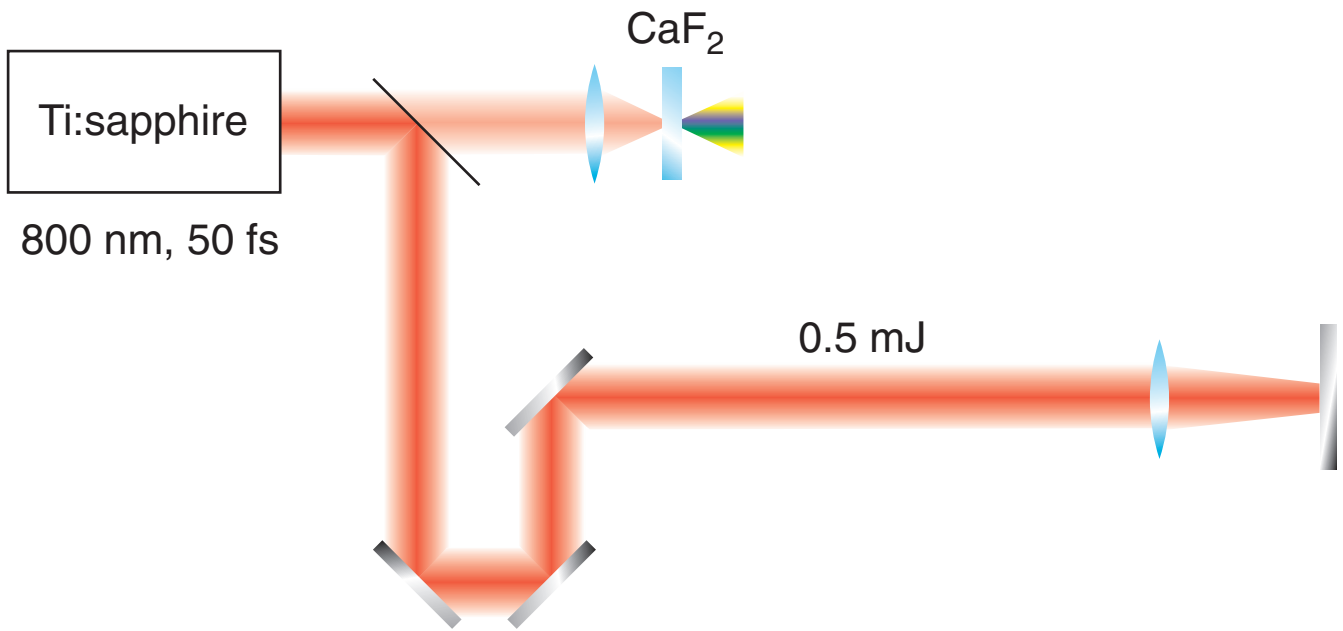
time-resolved dual-angle reflectometry



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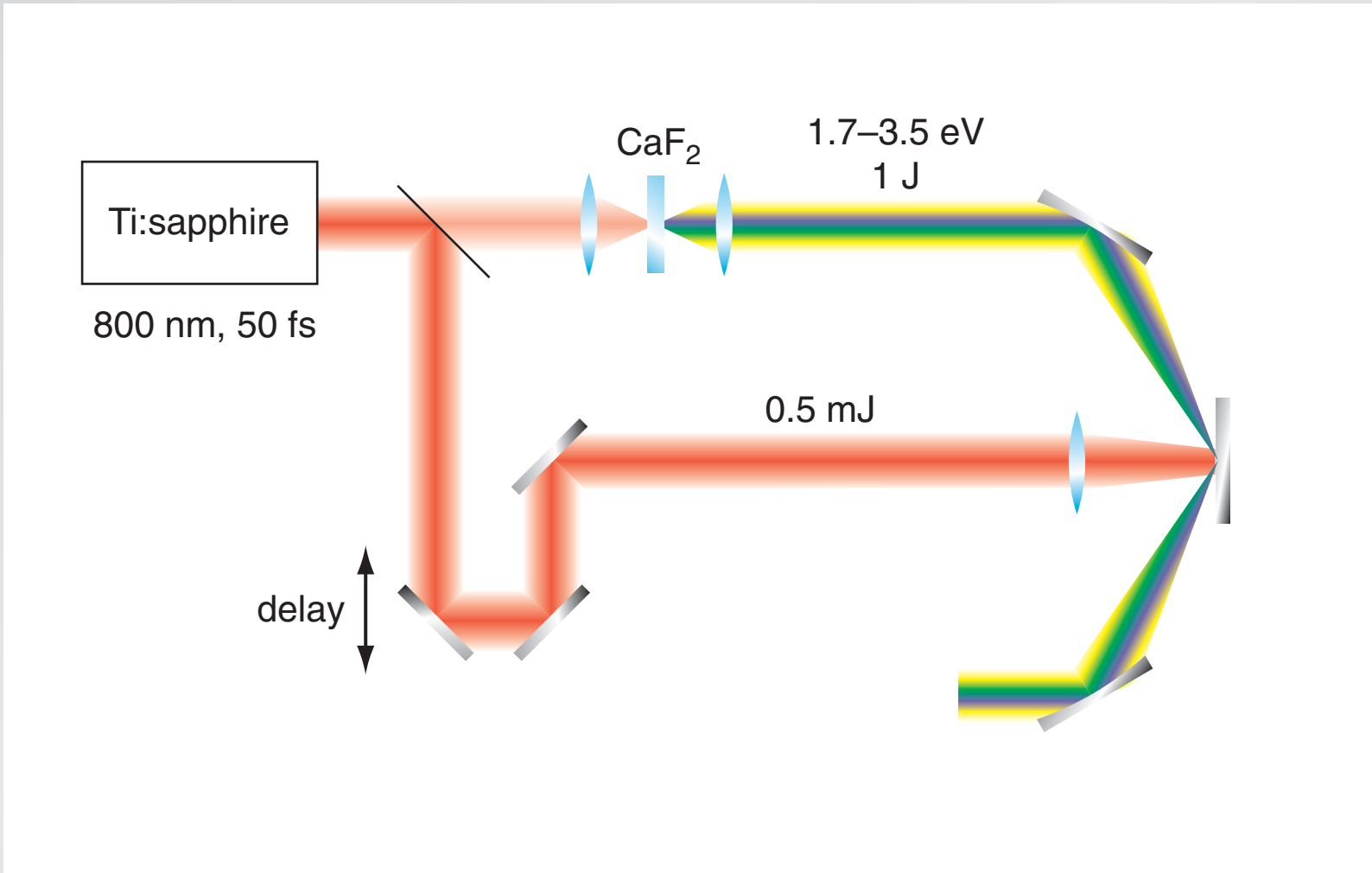
time-resolved dual-angle reflectometry



Experimental

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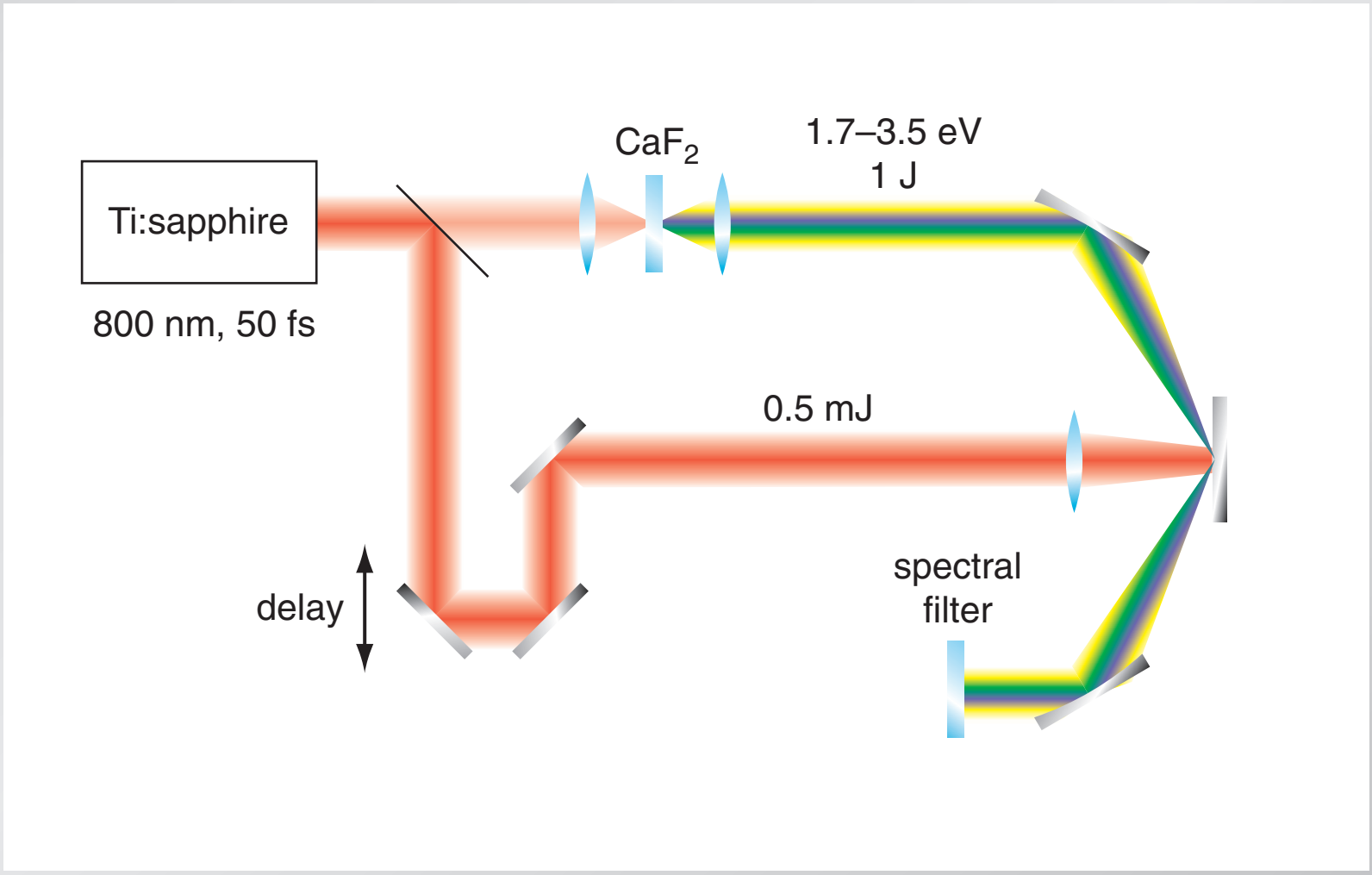

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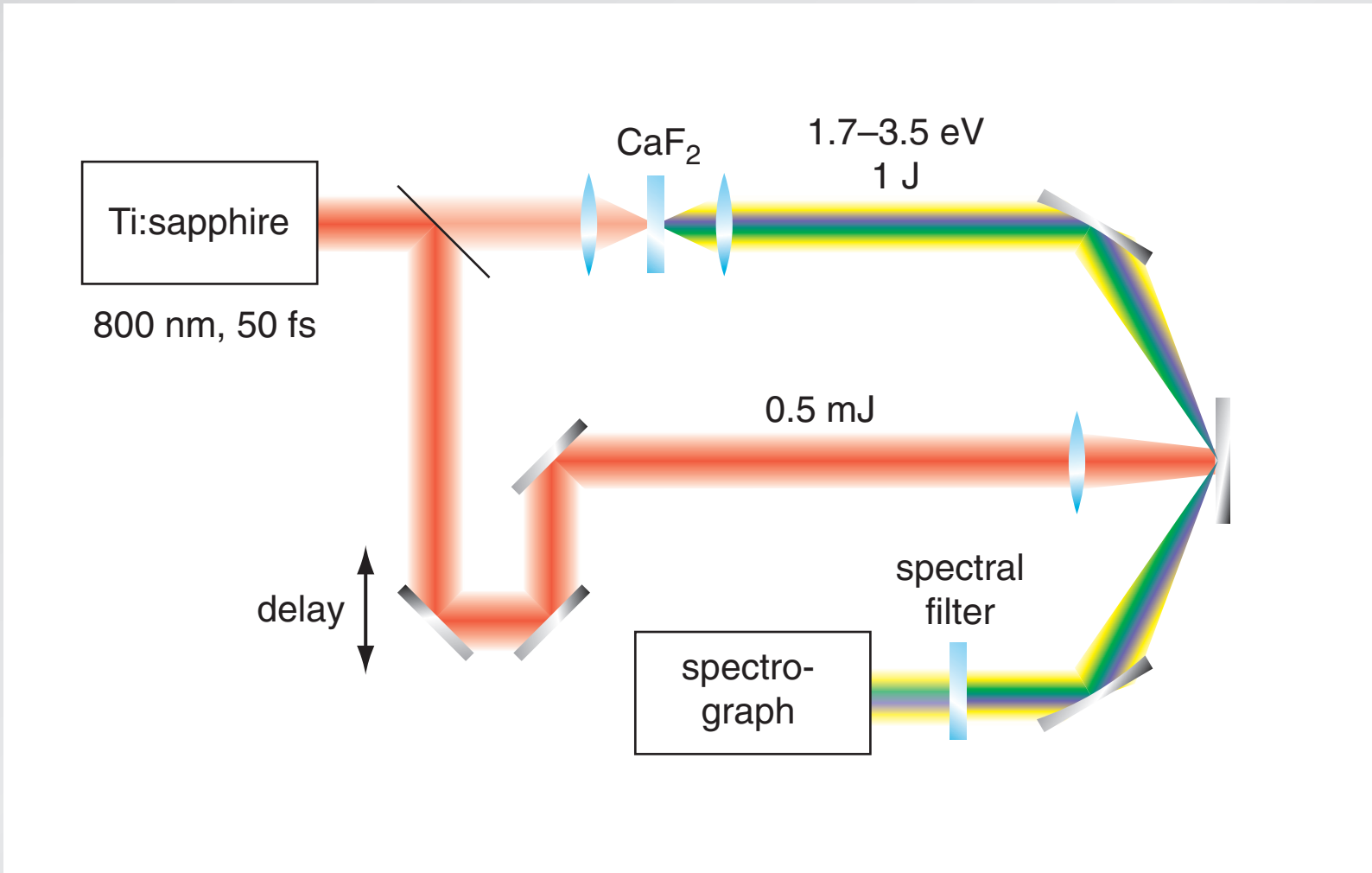

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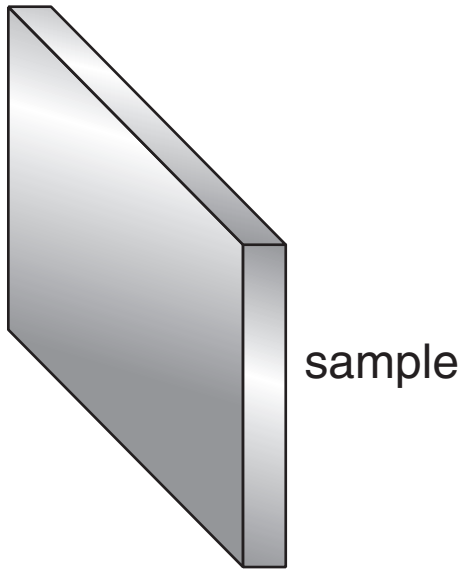

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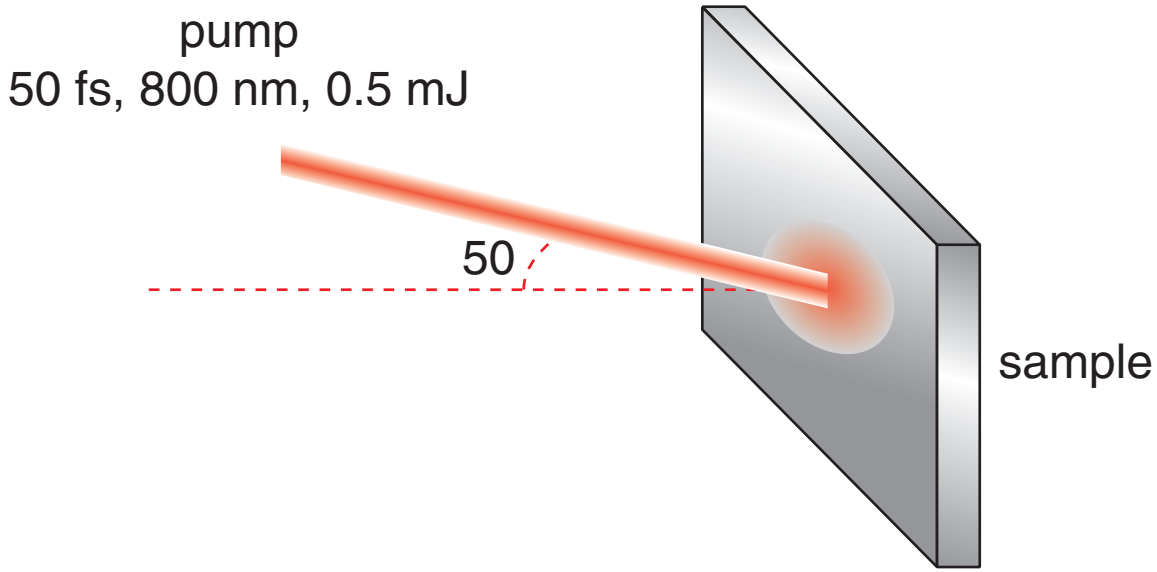
time-resolved dual-angle reflectometry



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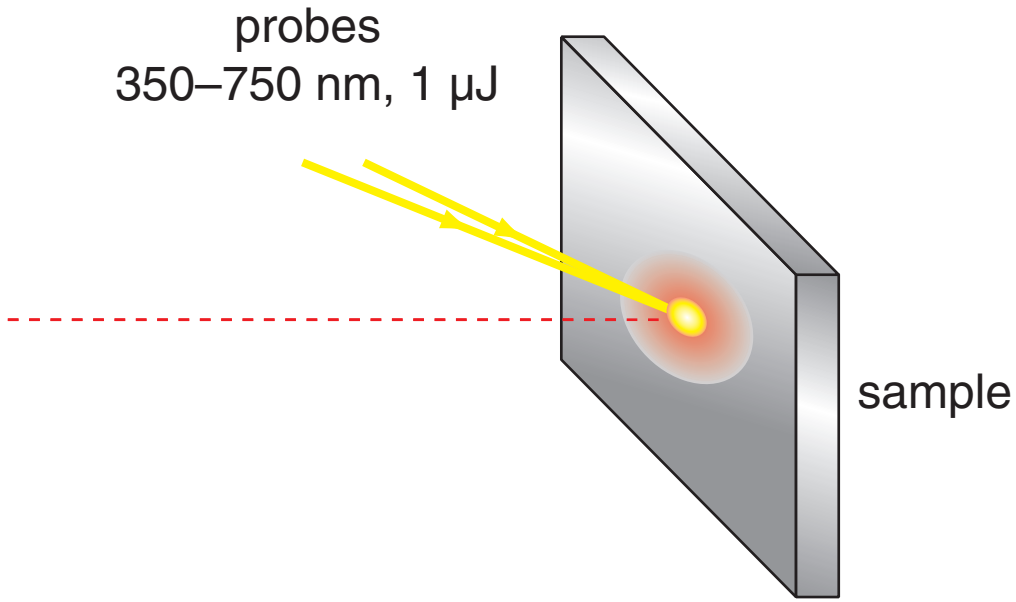
time-resolved dual-angle reflectometry



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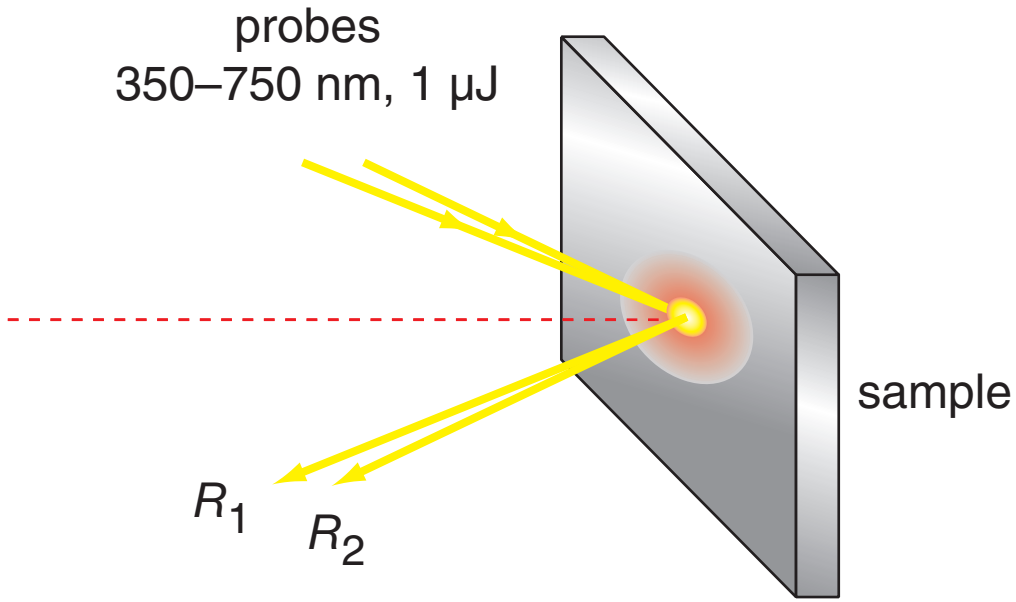

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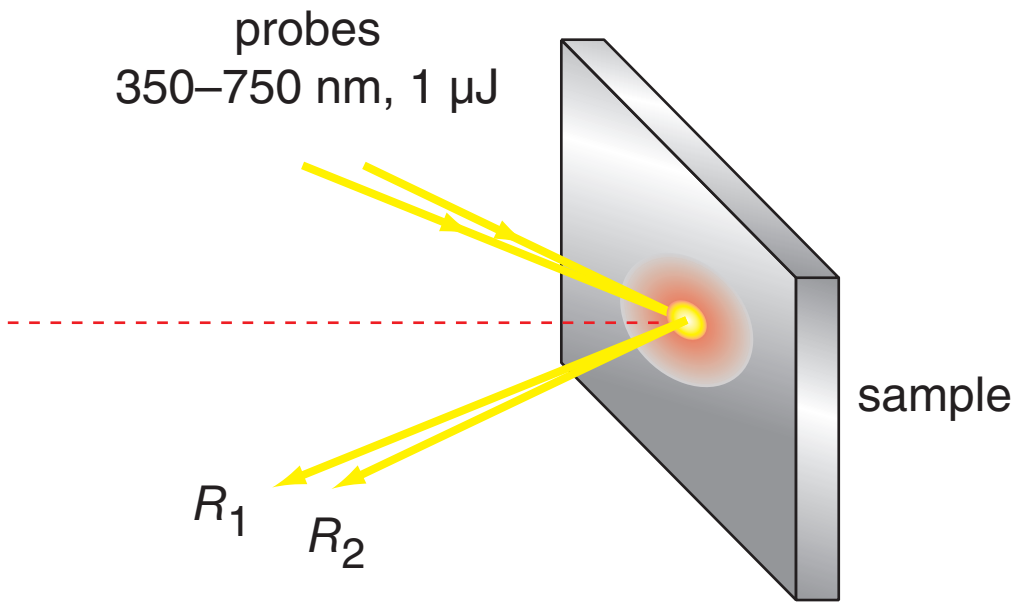
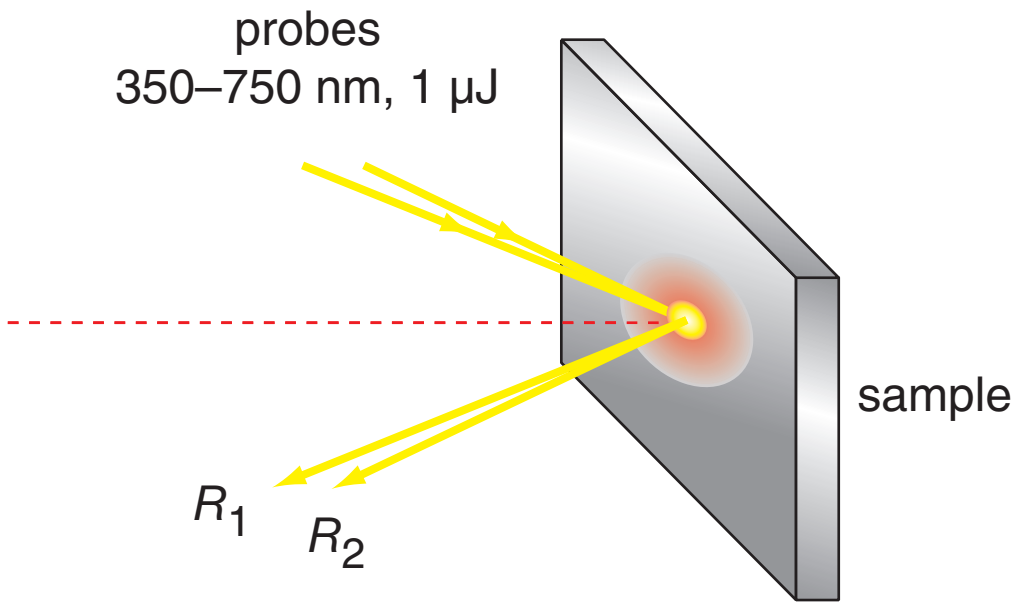


Fresnel equations

Experimental

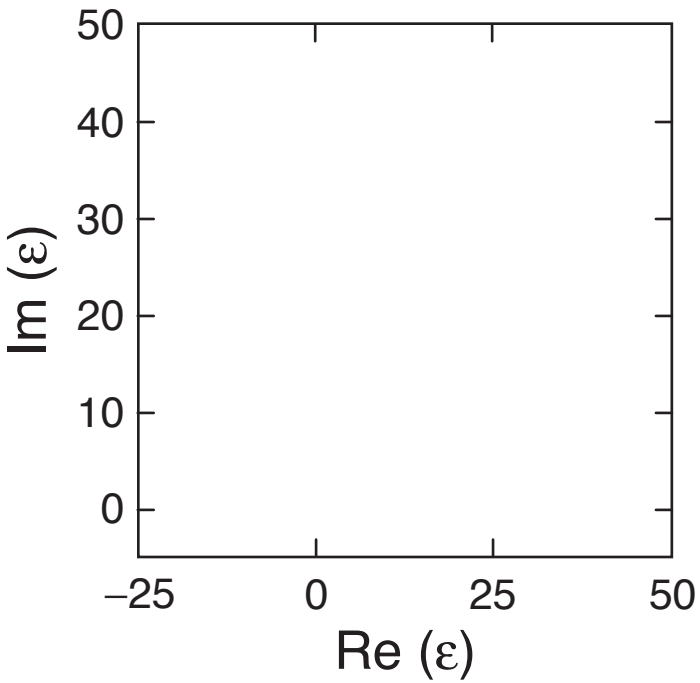
time-resolved dual-angle reflectometry

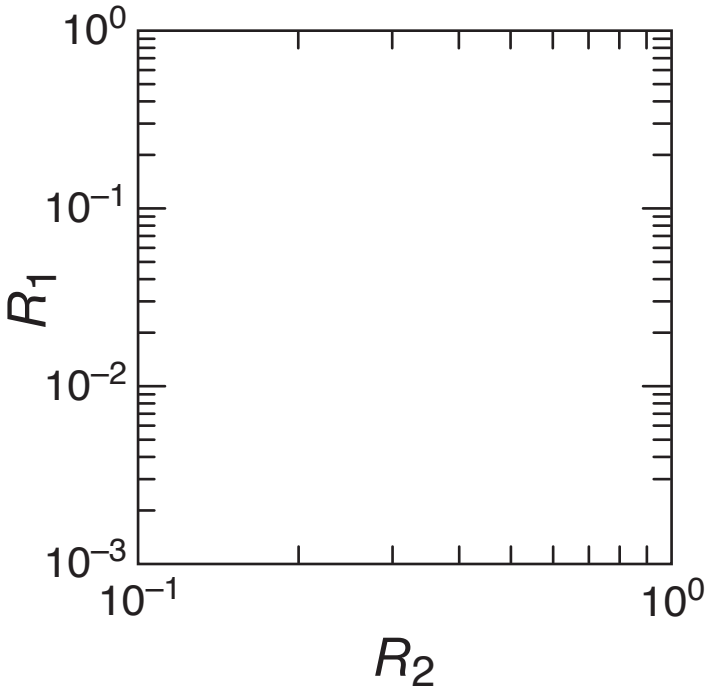


Fresnel equations → Re $\varepsilon(\omega)$, Im $\varepsilon(\omega)$

Experimental

choice of angles

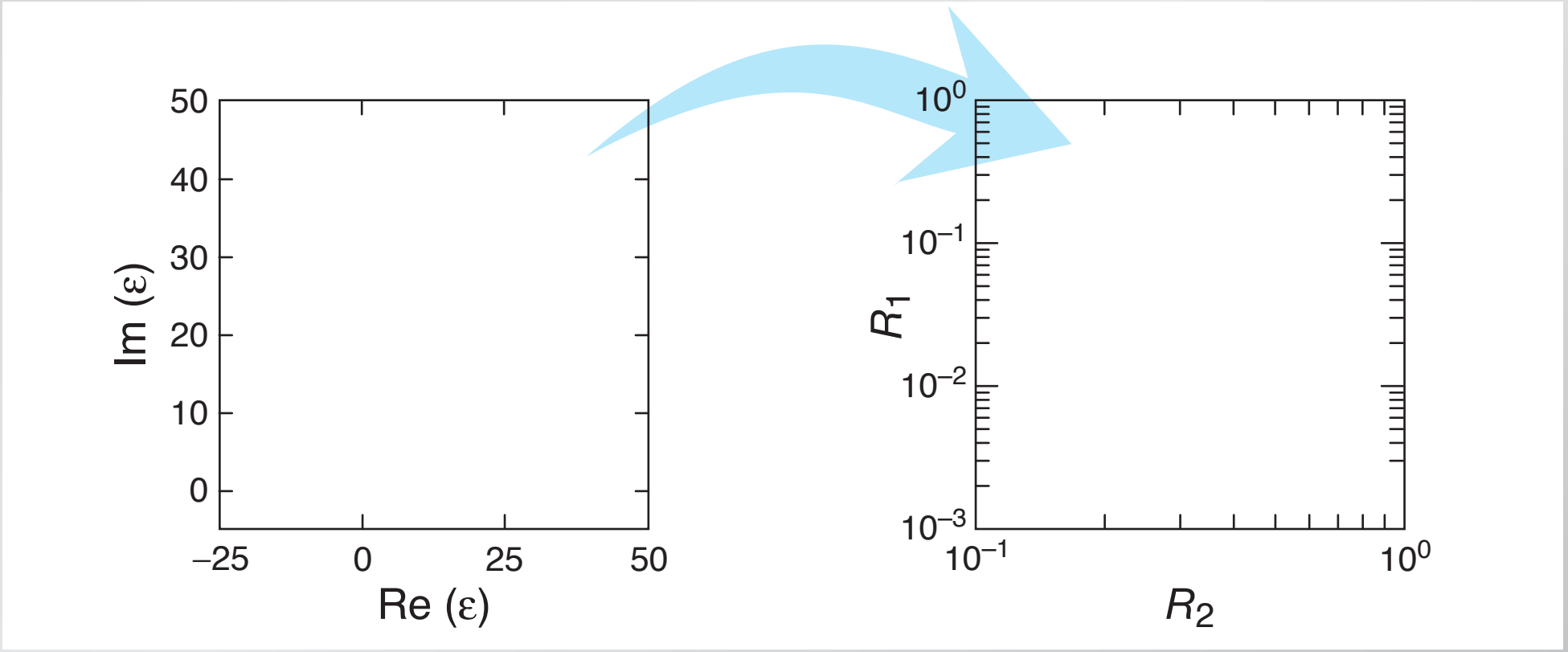




Fresnel equations cannot be inverted analytically

Experimental

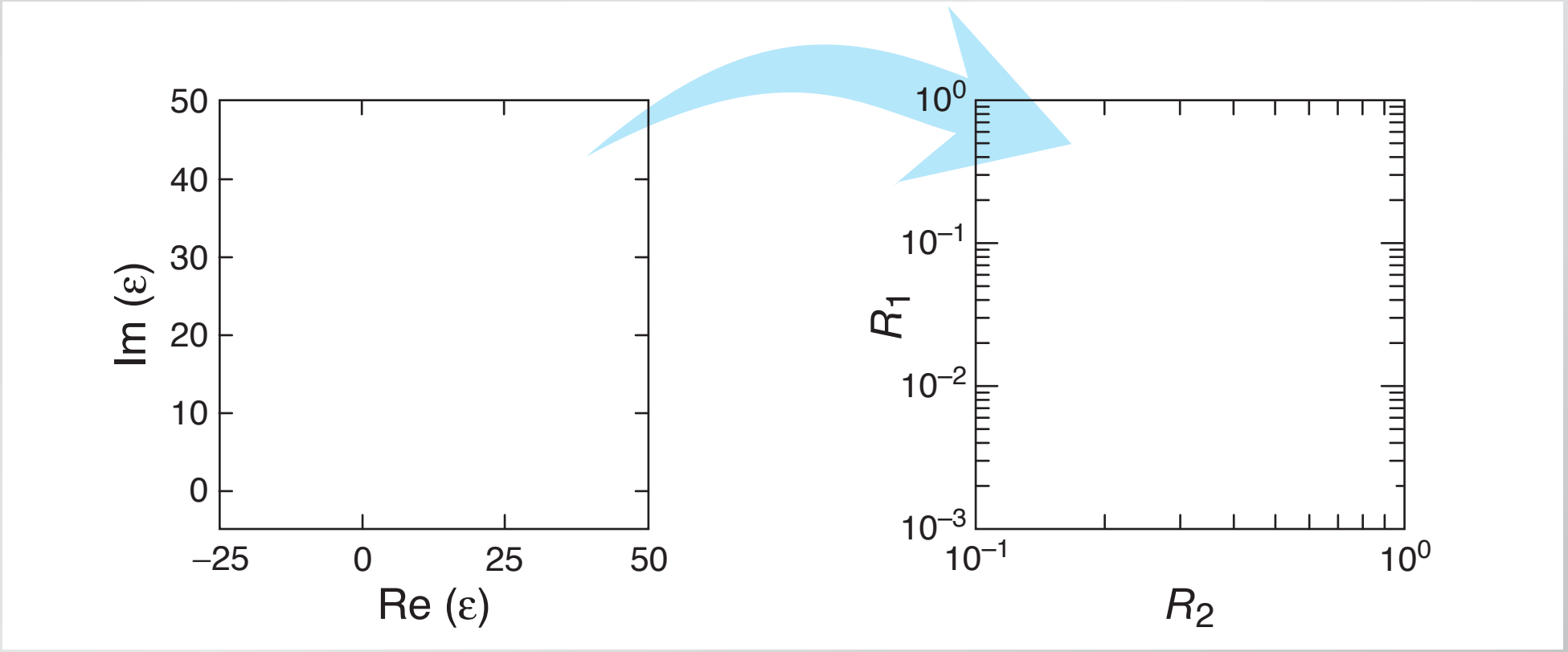
choice of angles



need numerical inversion

Experimental

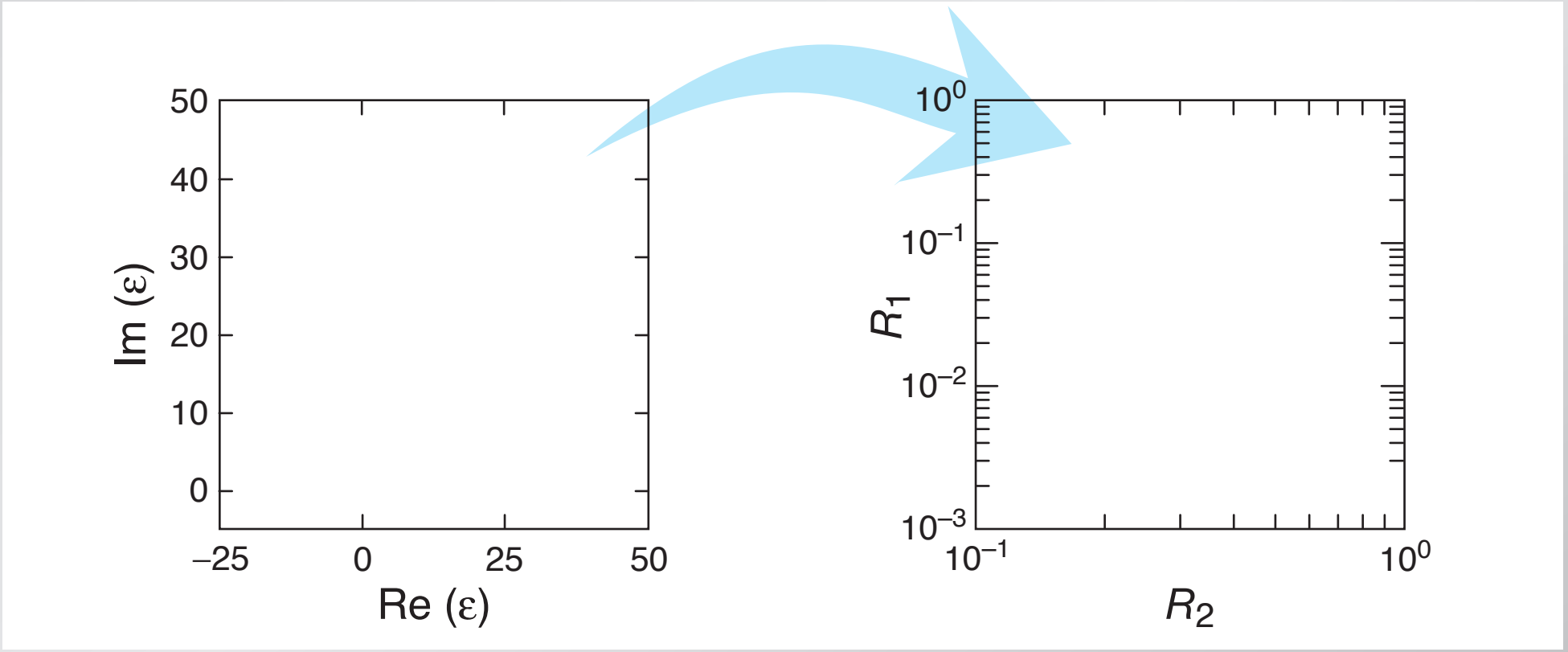
choice of angles



R_1 = 45° *p*-pol, R_2 = 45° *s*-pol

Experimental

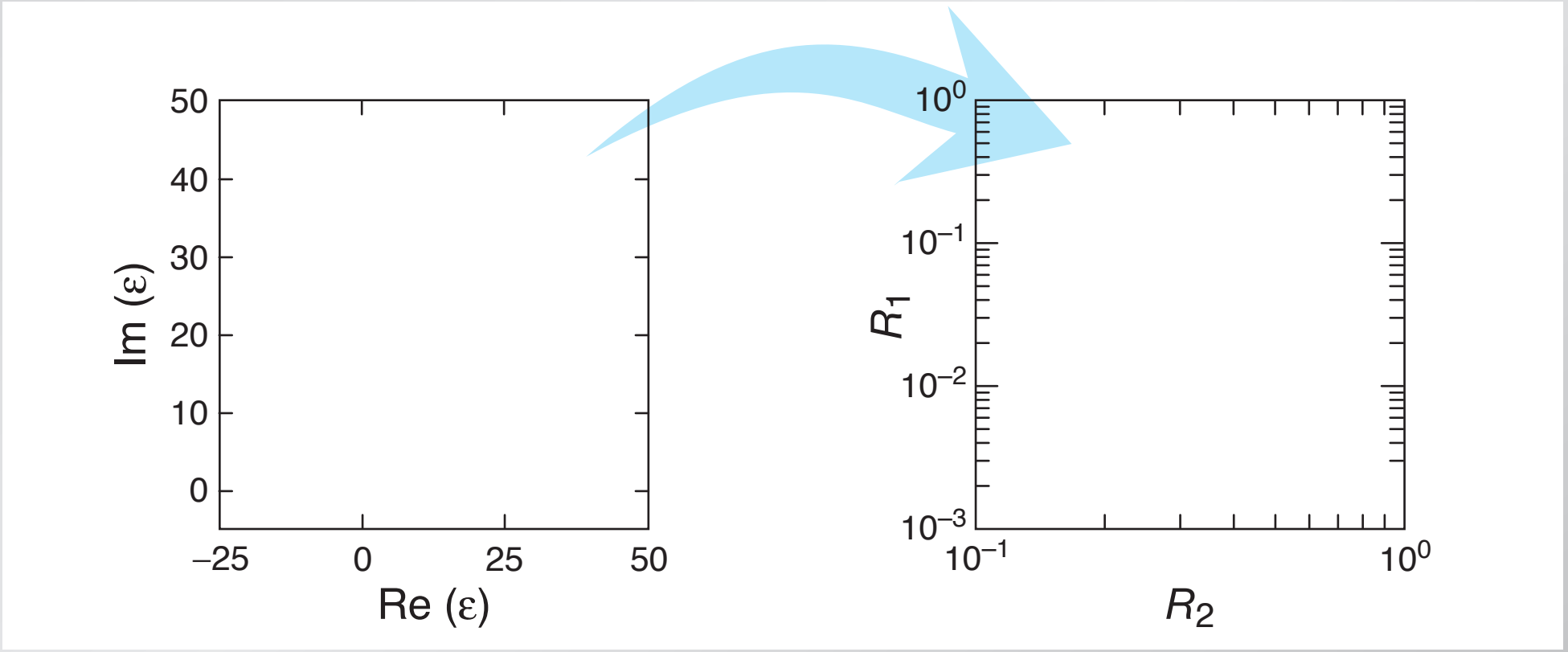
choice of angles



R_1 = 60° *p*-pol, R_2 = 45° *p*-pol

Experimental

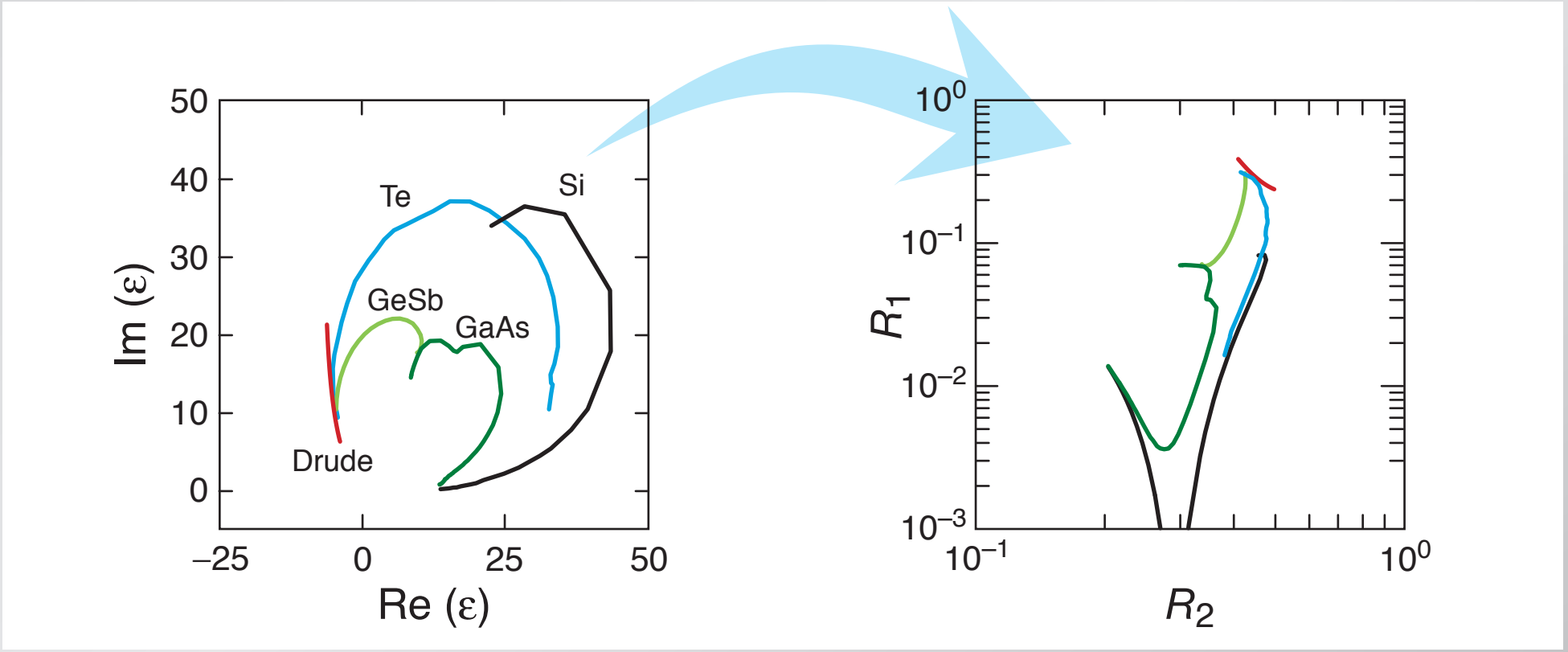
choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

Experimental

choice of angles



R_1 = 78° *p*-pol, R_2 = 45° *p*-pol

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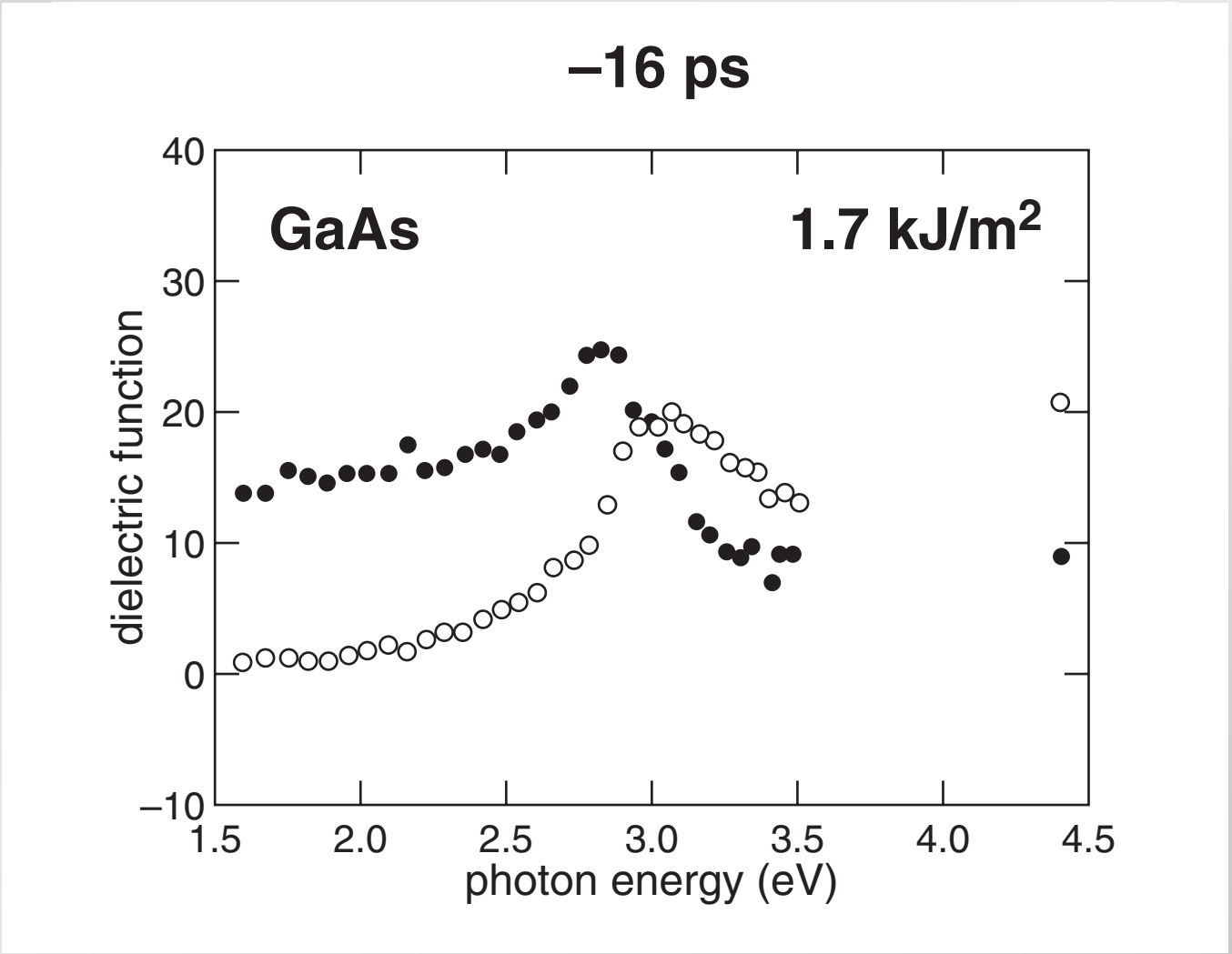
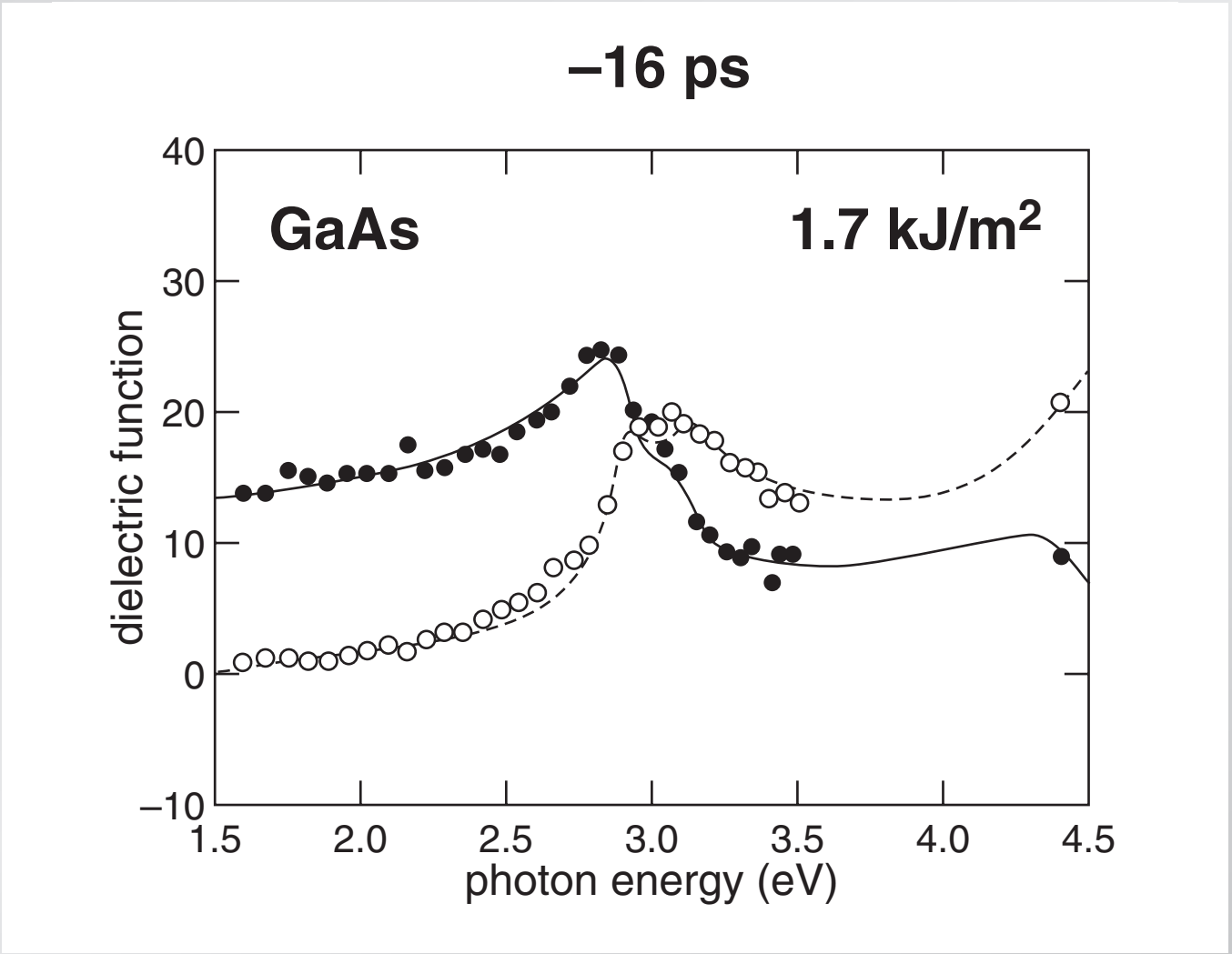


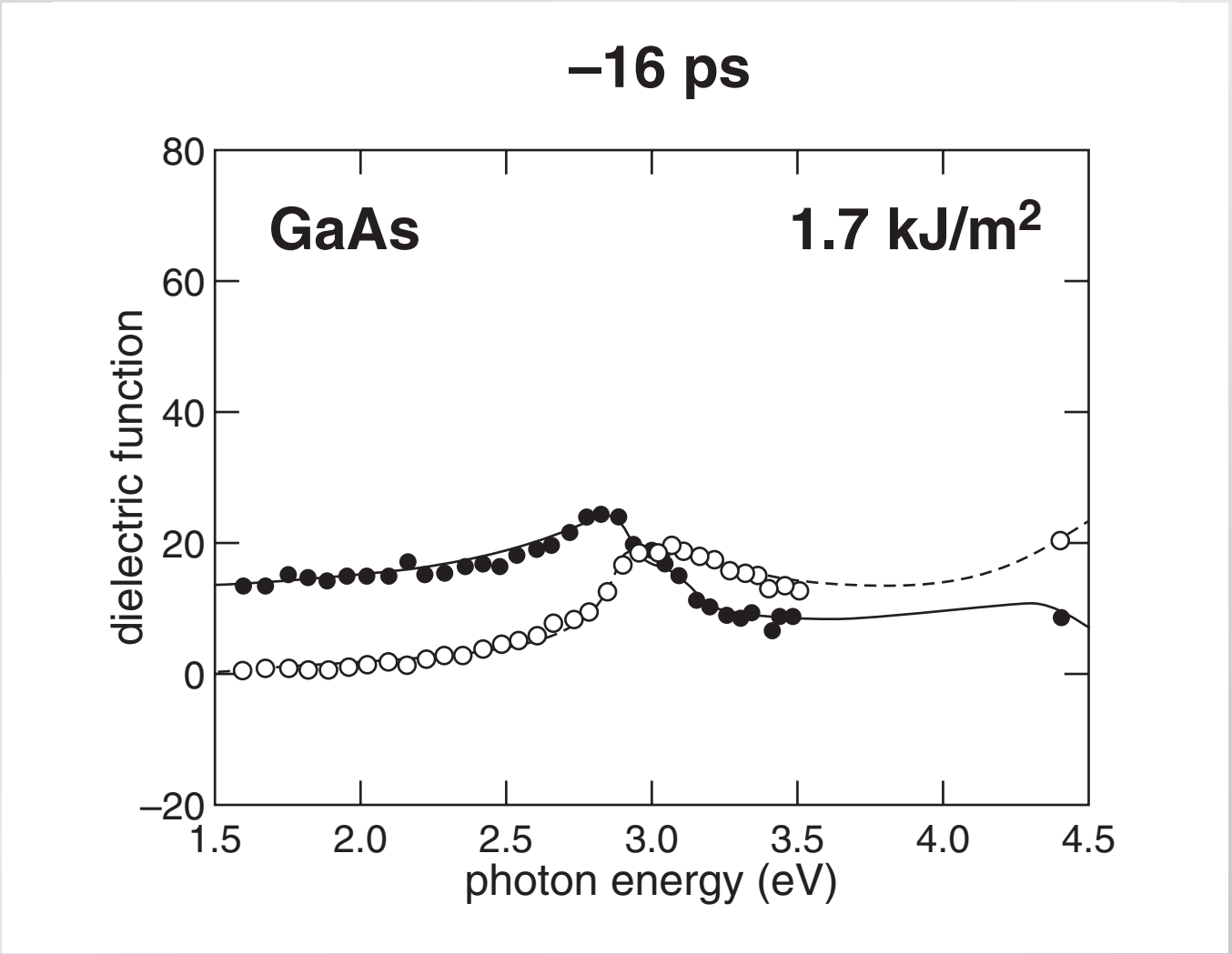
Phys. Rev. Lett. 80, 185 (1998)

Experimental



E.D. Palik, *Handbook of Optical Constants of Solids* (Academic Press, 1985)

Experimental



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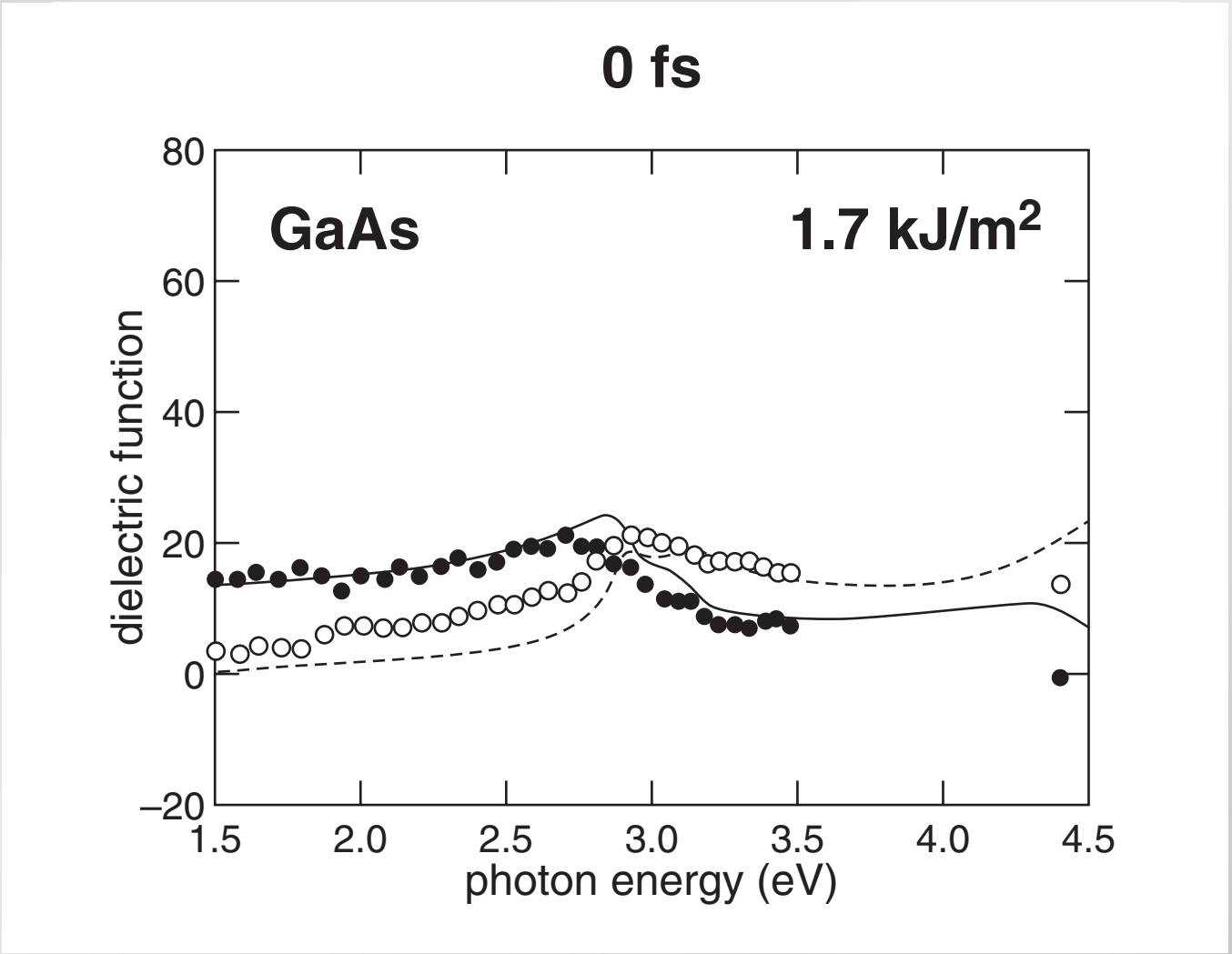

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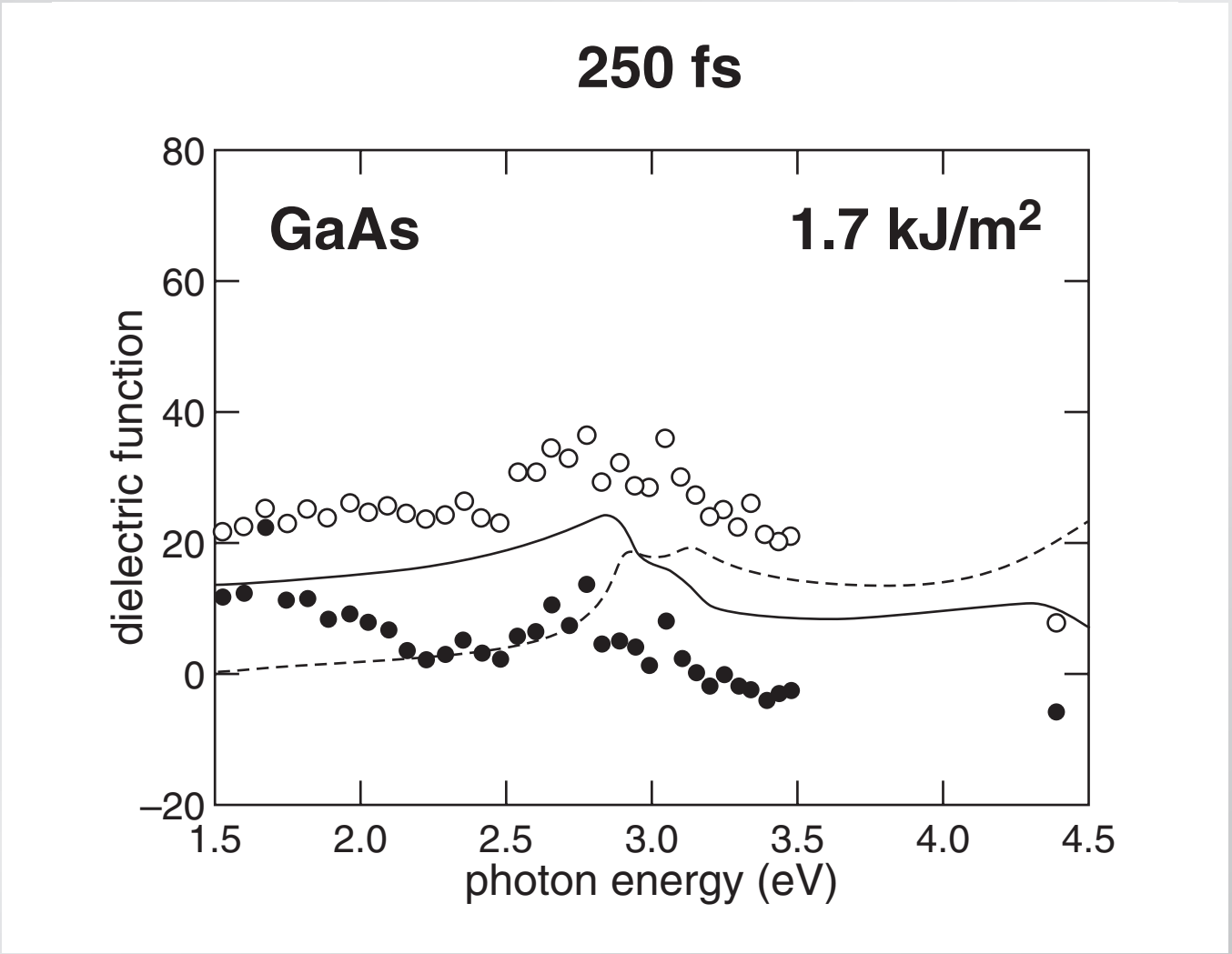

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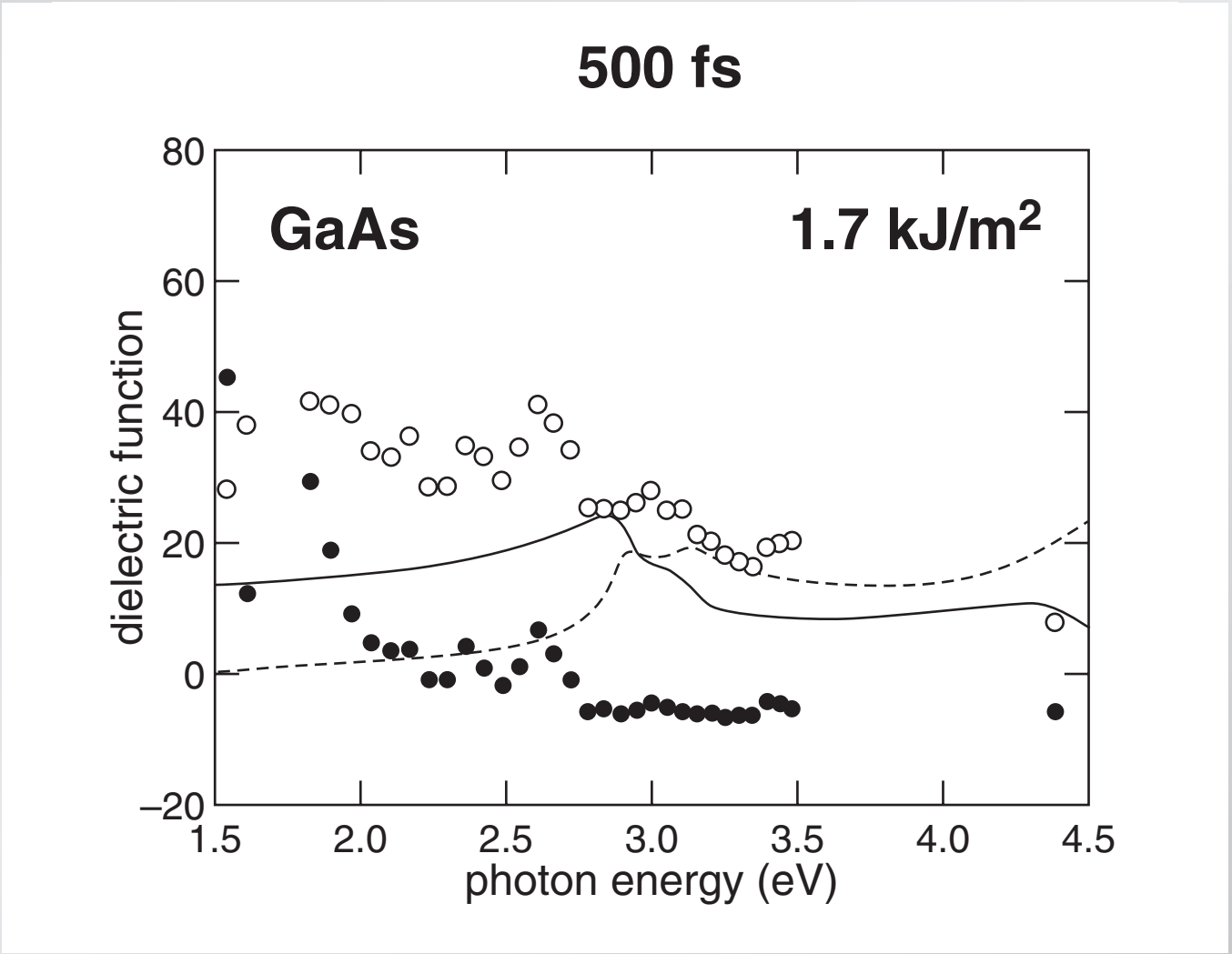

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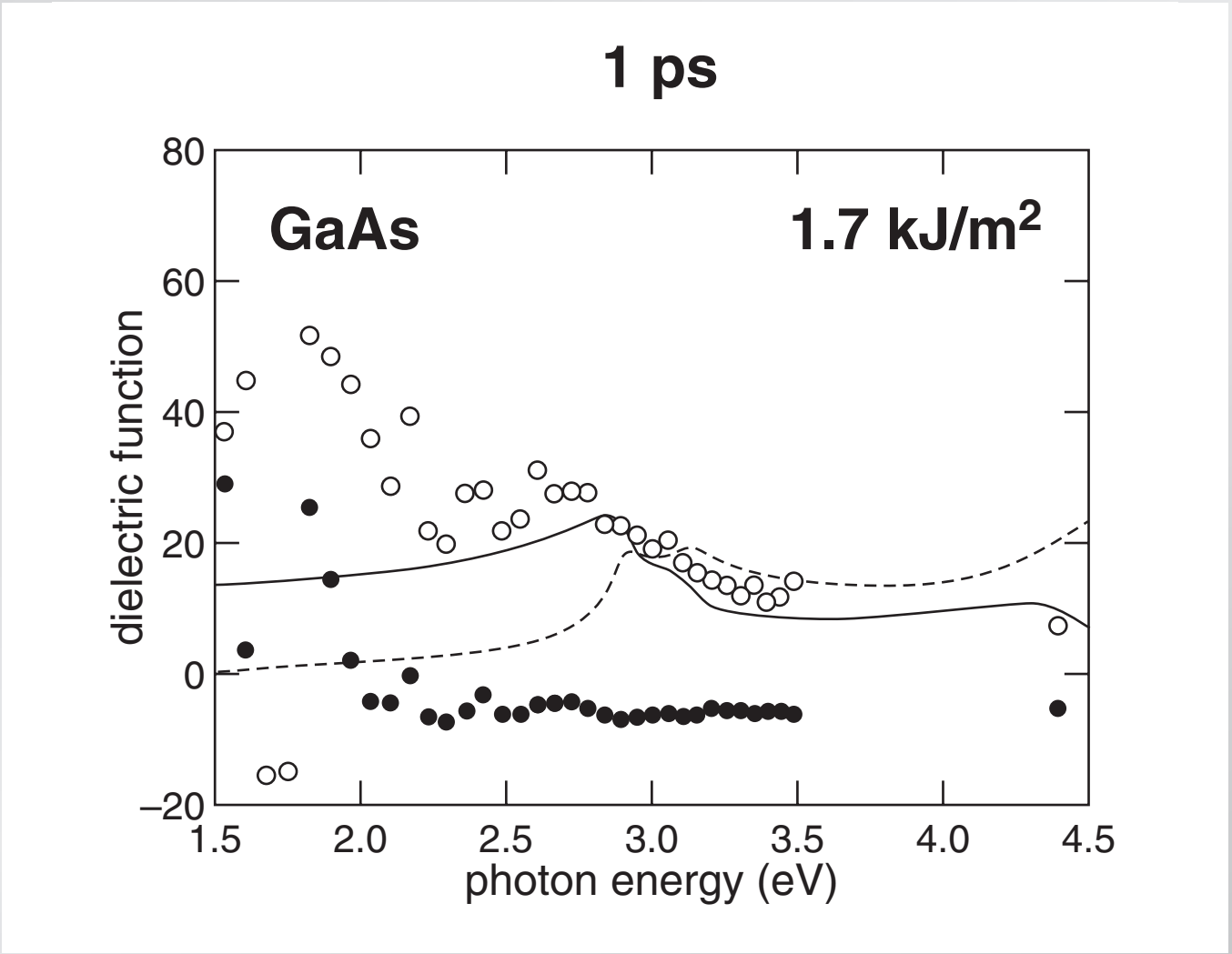

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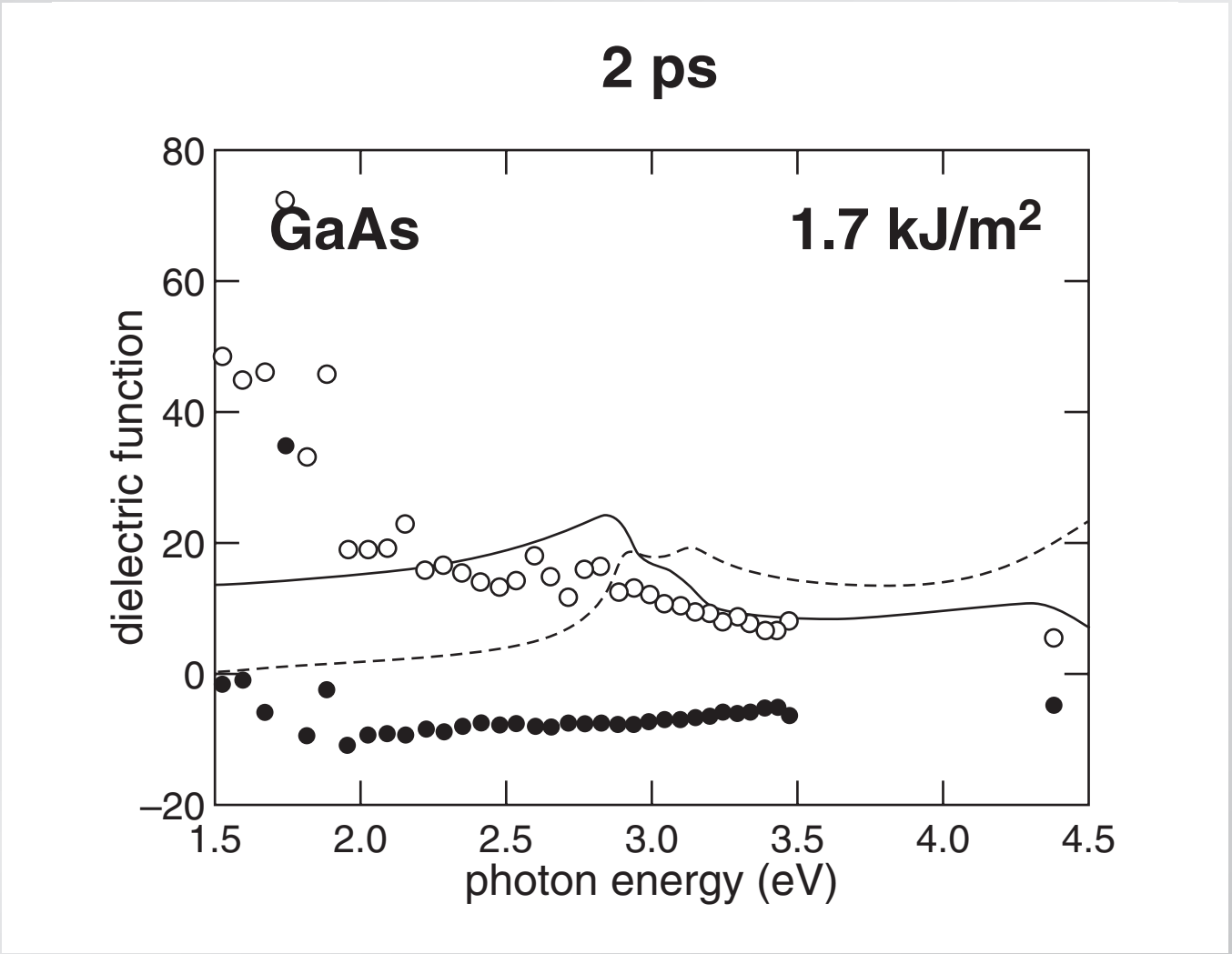

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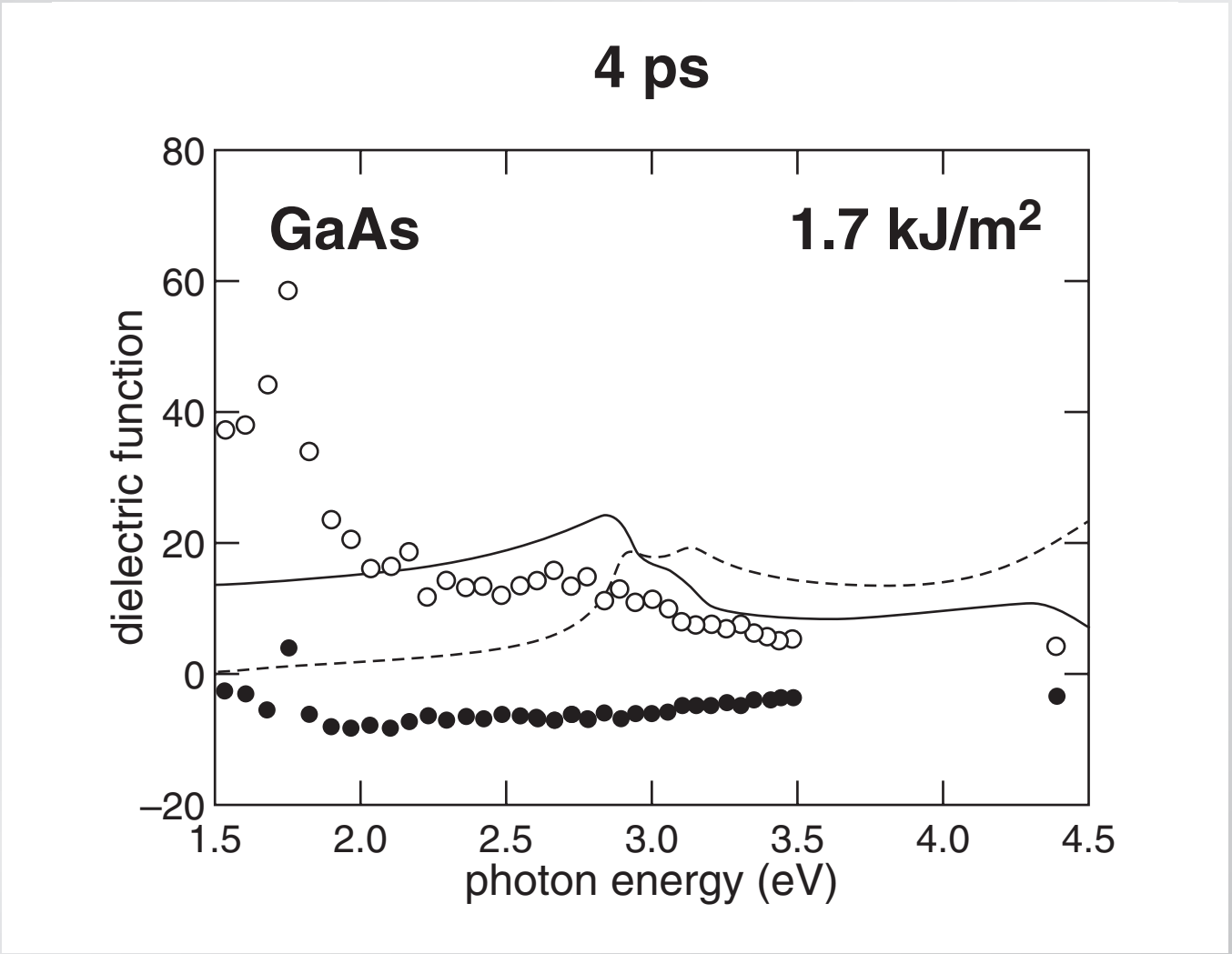
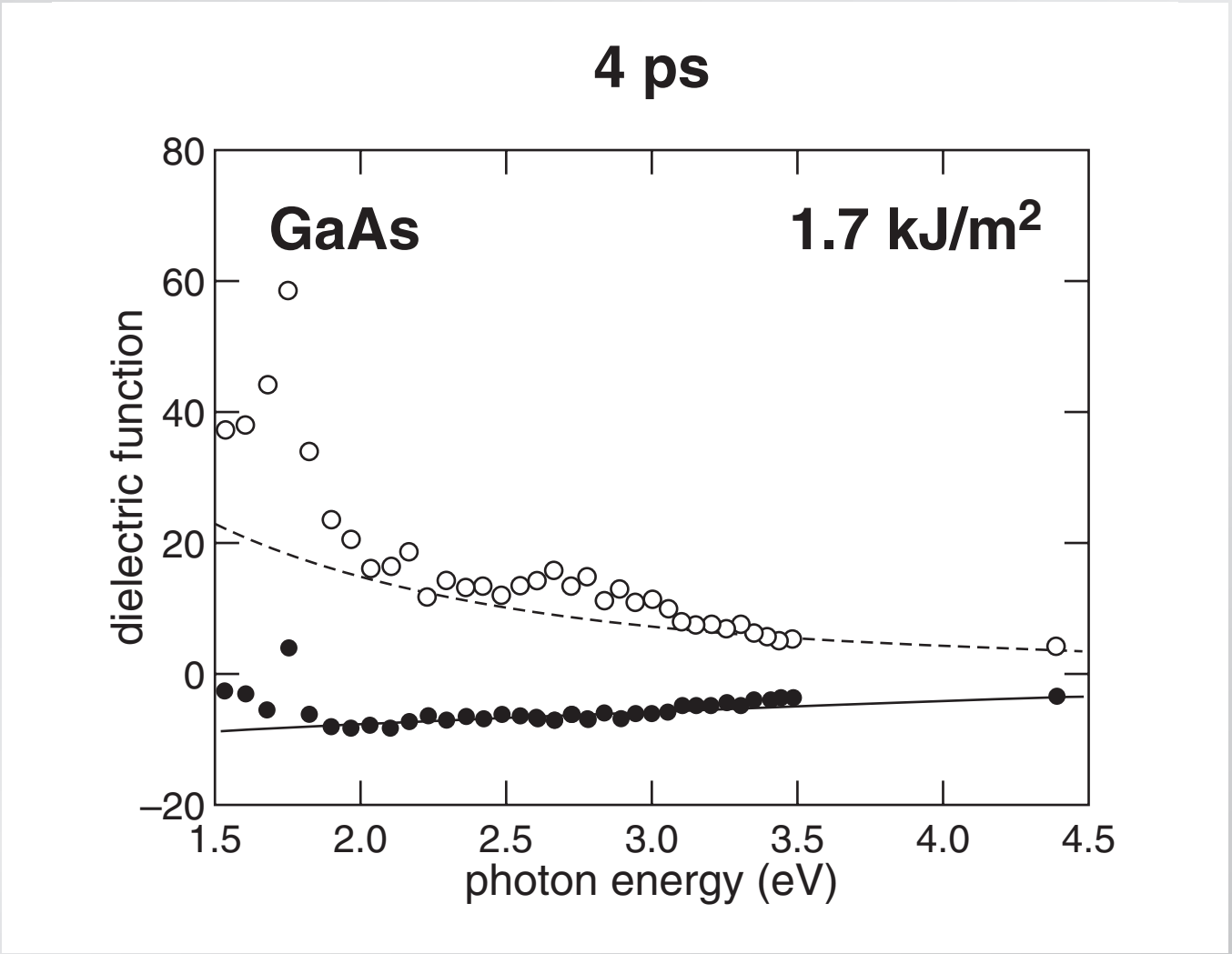


Phys. Rev. Lett. 80, 185 (1998)

Experimental



Phys. Rev. Lett. 80, 185 (1998)

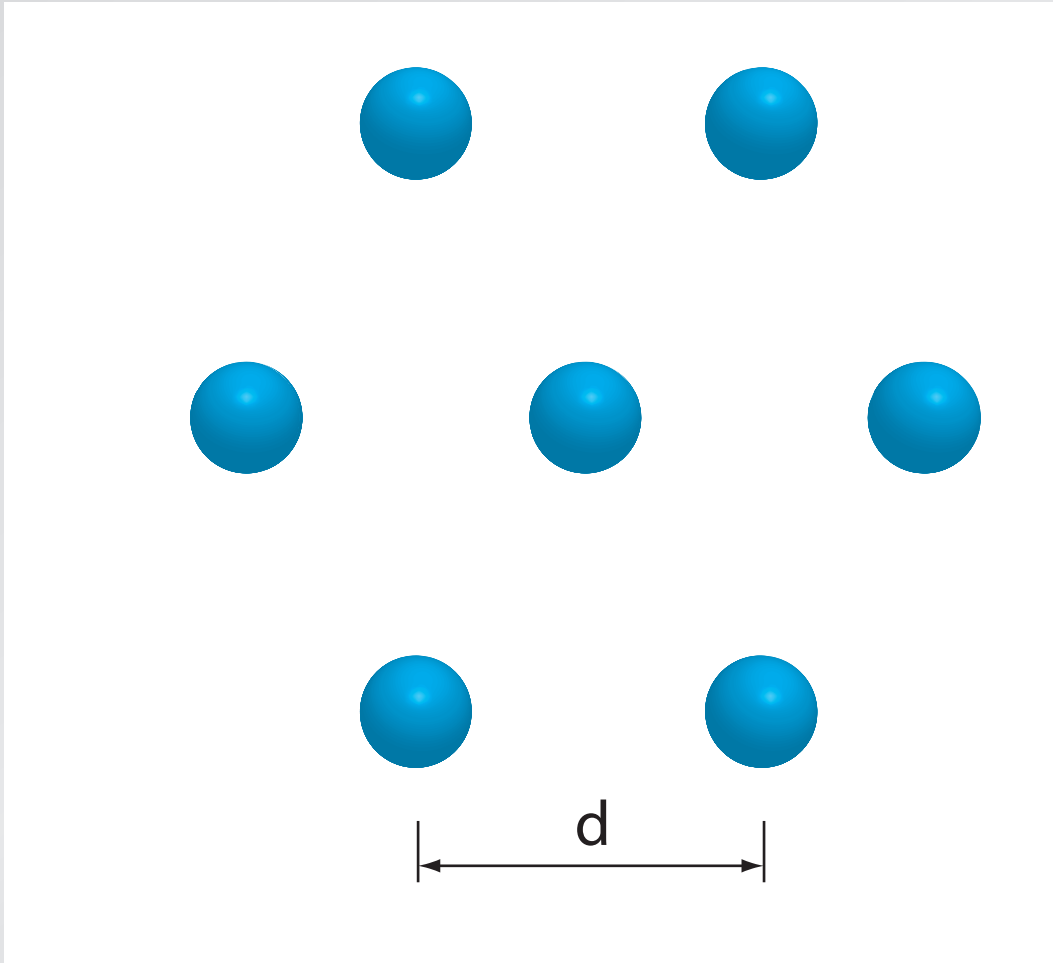
Experimental

can observe dielectric to metallic transition

Outline

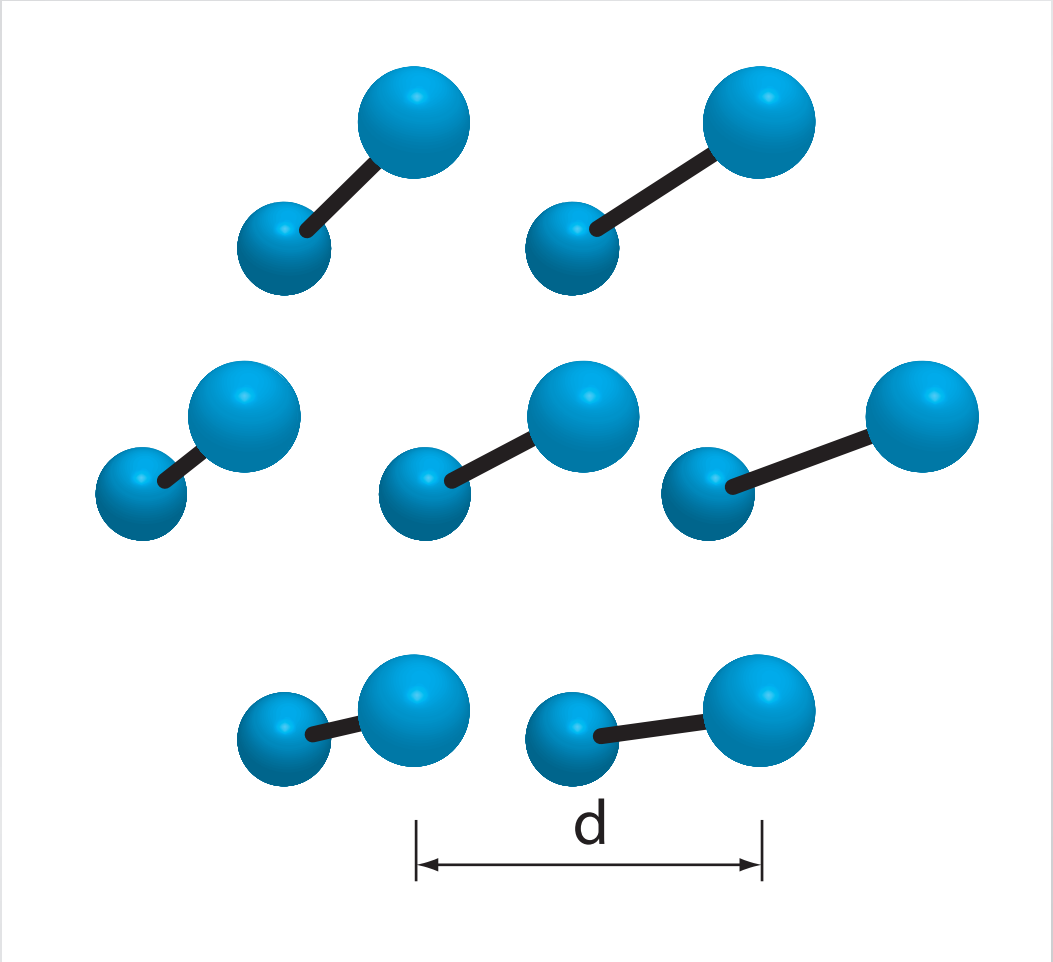
- experimental
- coherent phonons
- optical control

Coherent phonons



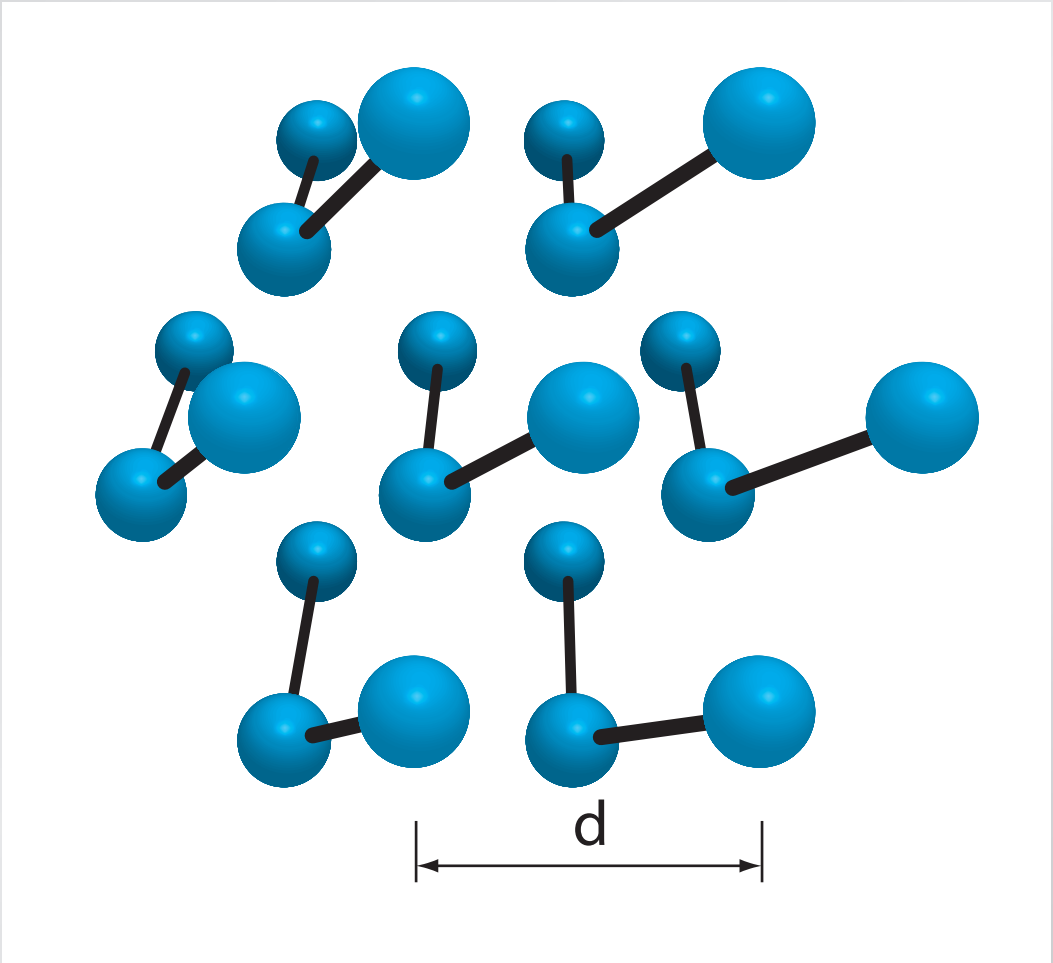
tellurium has hexagonal arrangement

Coherent phonons



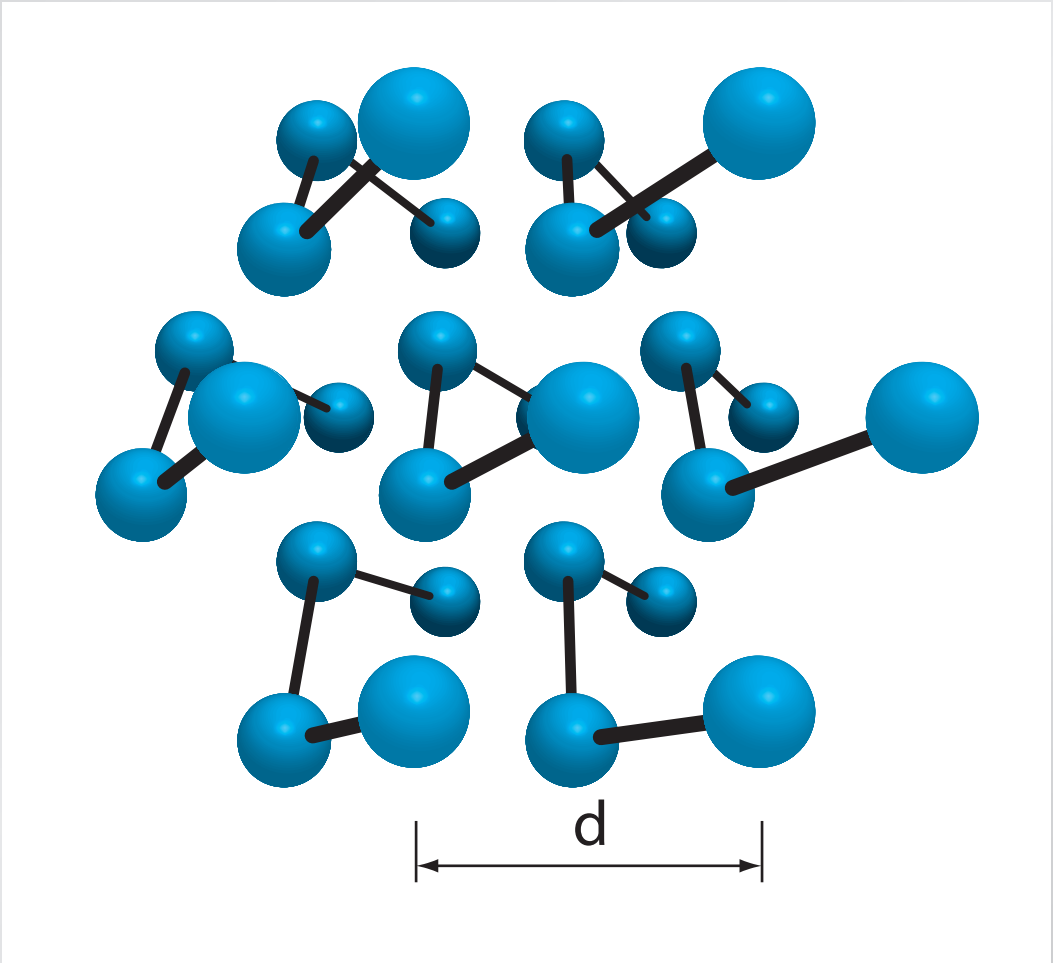
with lattice planes offset in spiral fashion

Coherent phonons



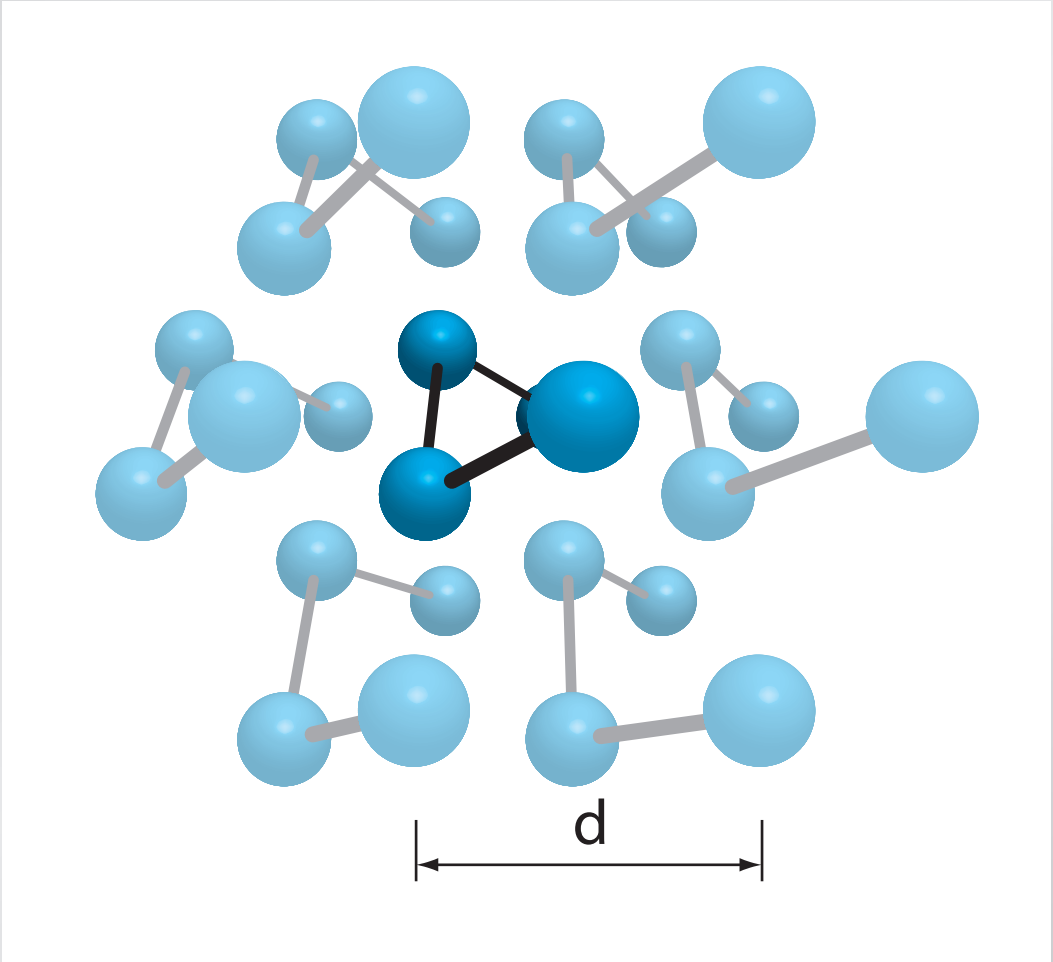
with lattice planes offset in spiral fashion

Coherent phonons



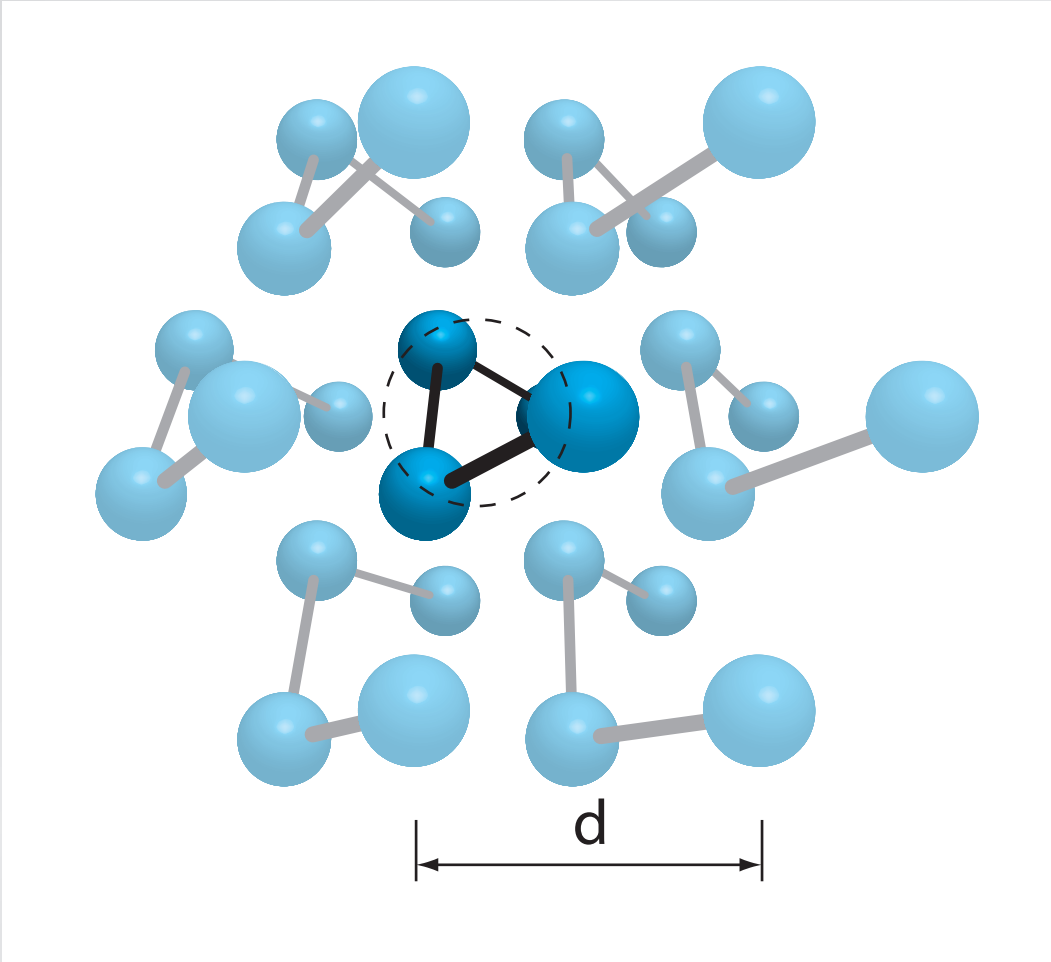
with lattice planes offset in spiral fashion

Coherent phonons



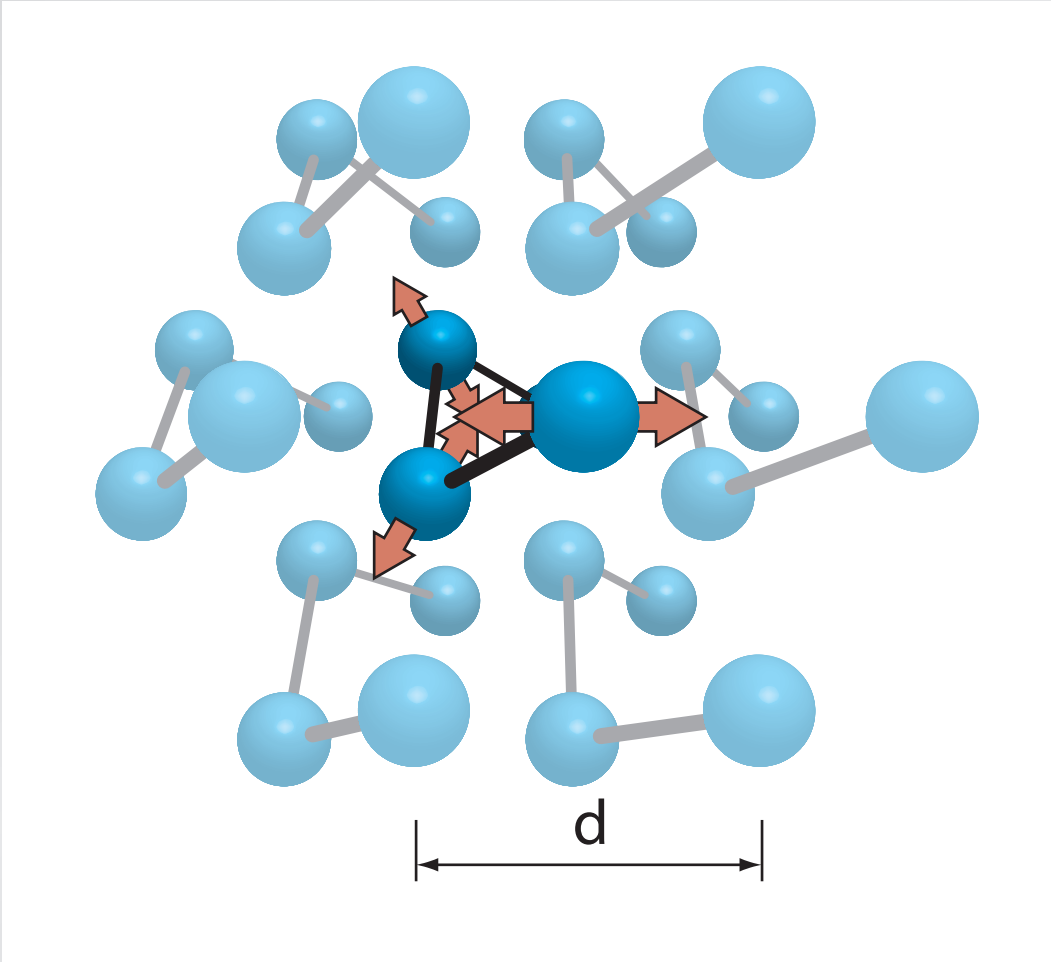
with lattice planes offset in spiral fashion

Coherent phonons



helical radius $x = 0.27d$

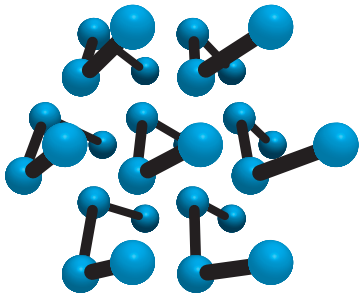
Coherent phonons



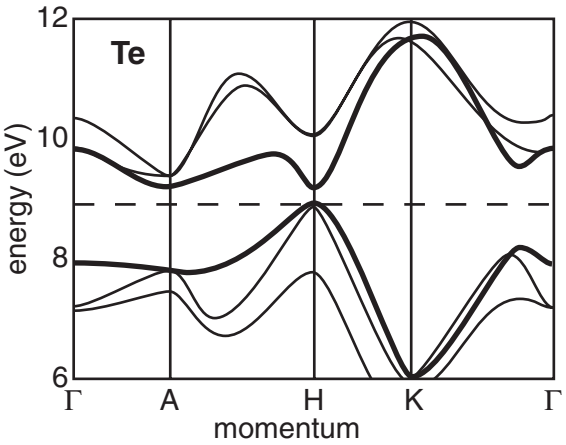
A_1 mode modulates x

Coherent phonons

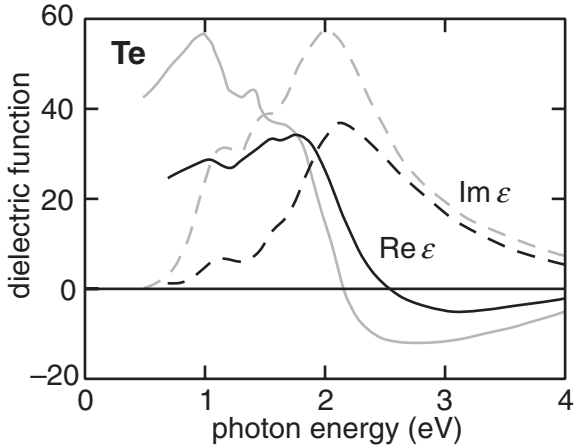
structure



band structure

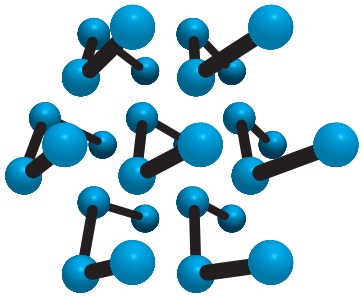


dielectric function

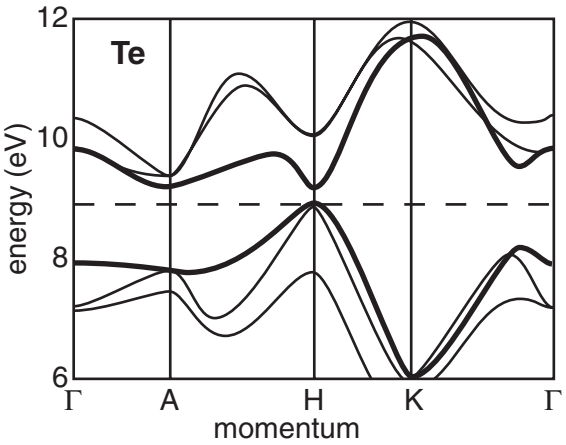


Coherent phonons

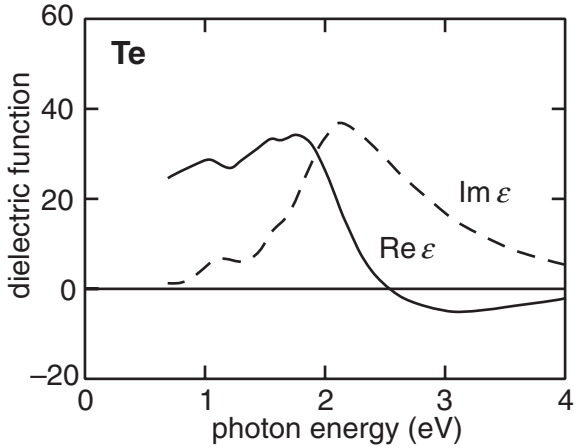
structure



band structure

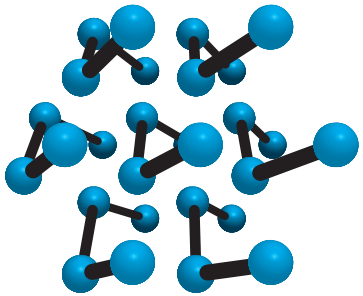


dielectric function

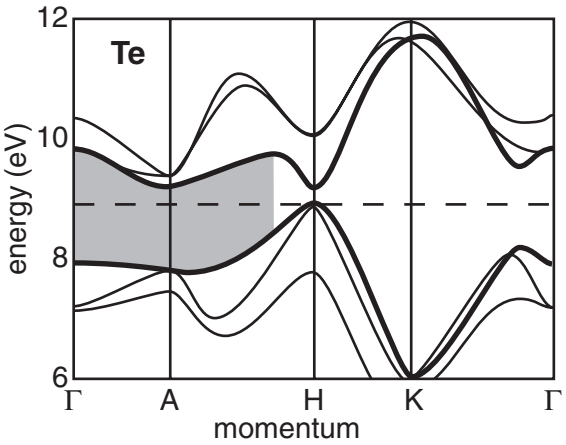


Coherent phonons

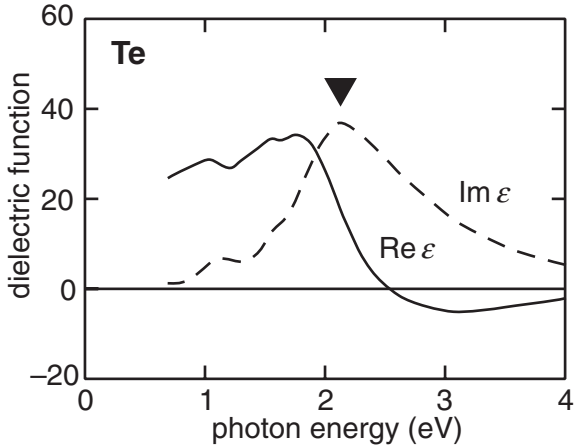
structure



band structure



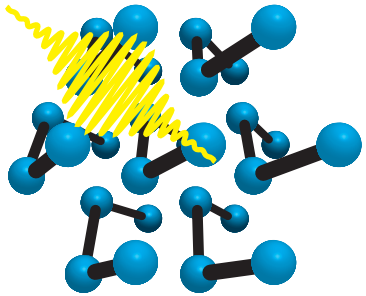
dielectric function



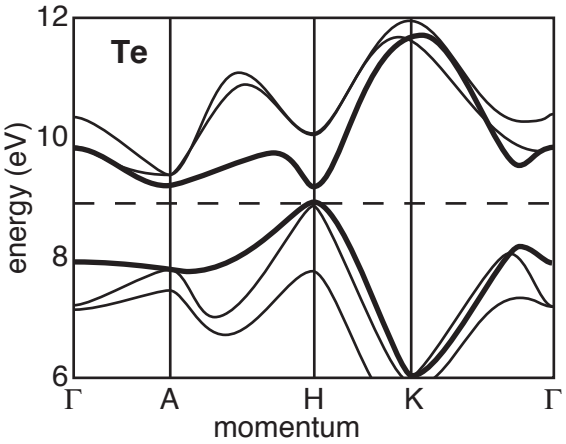
Coherent phonons

photoexcitation causes modulation of helical radius

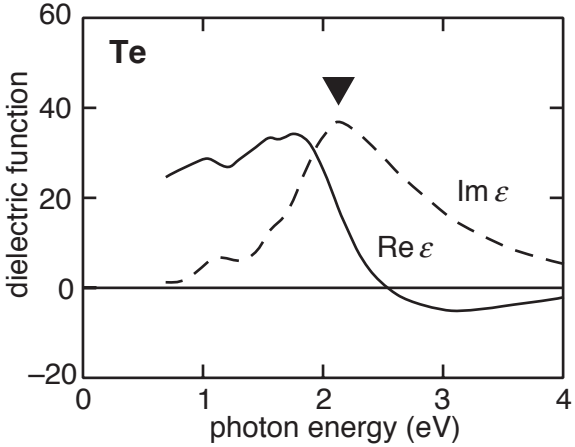
structure



band structure



dielectric function

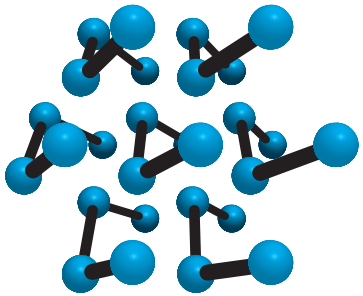


band structure very sensitive to helical radius

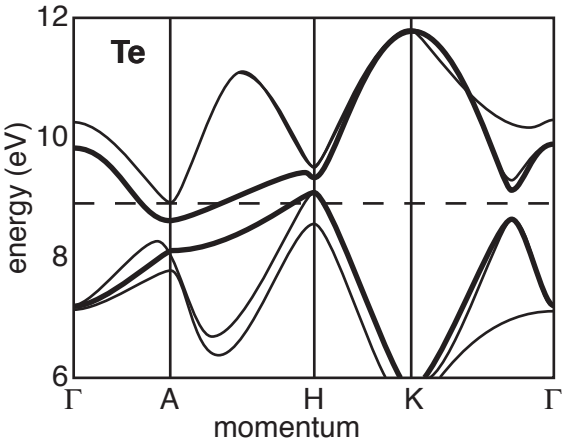
Coherent phonons

15% change drastically alters band structure

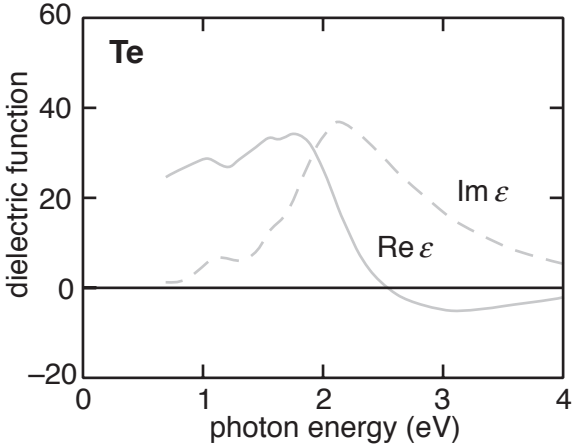
structure



band structure



dielectric function

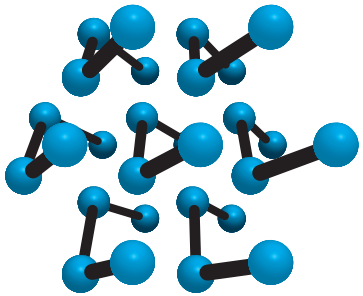


Tangney and Fahy, *Phys. Rev. B 65*, 054302 (2002)

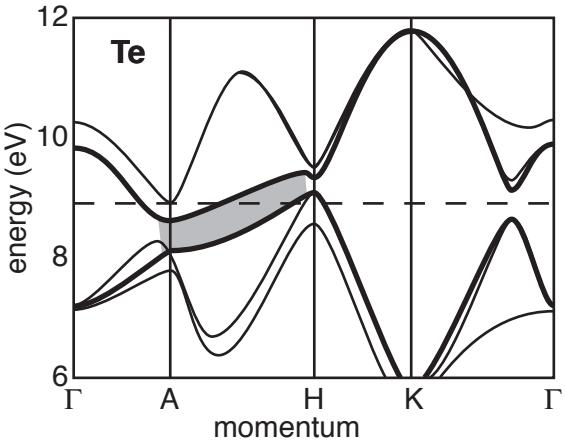
Coherent phonons

should cause a red-shift of dielectric function

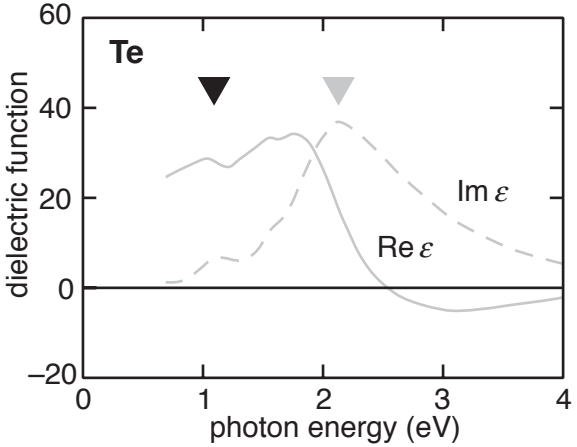
structure



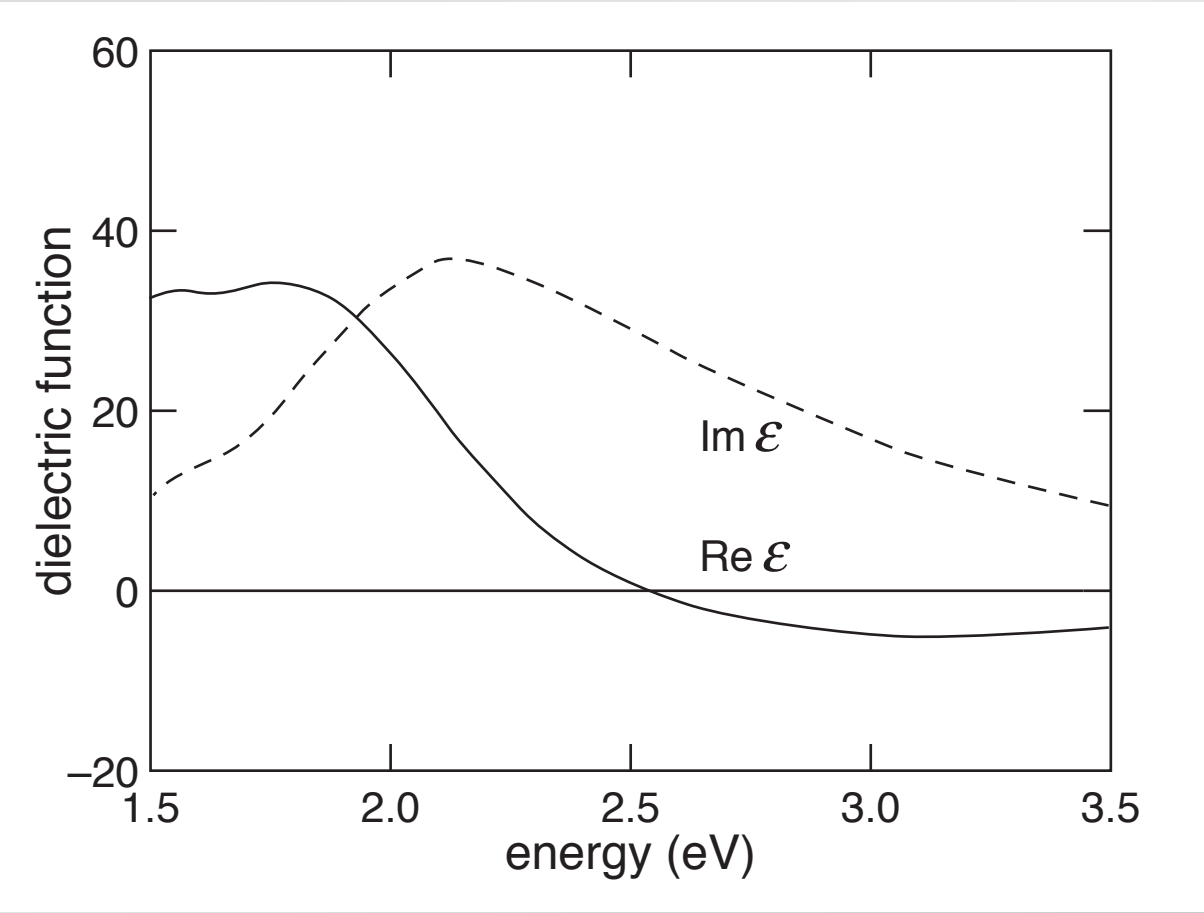
band structure



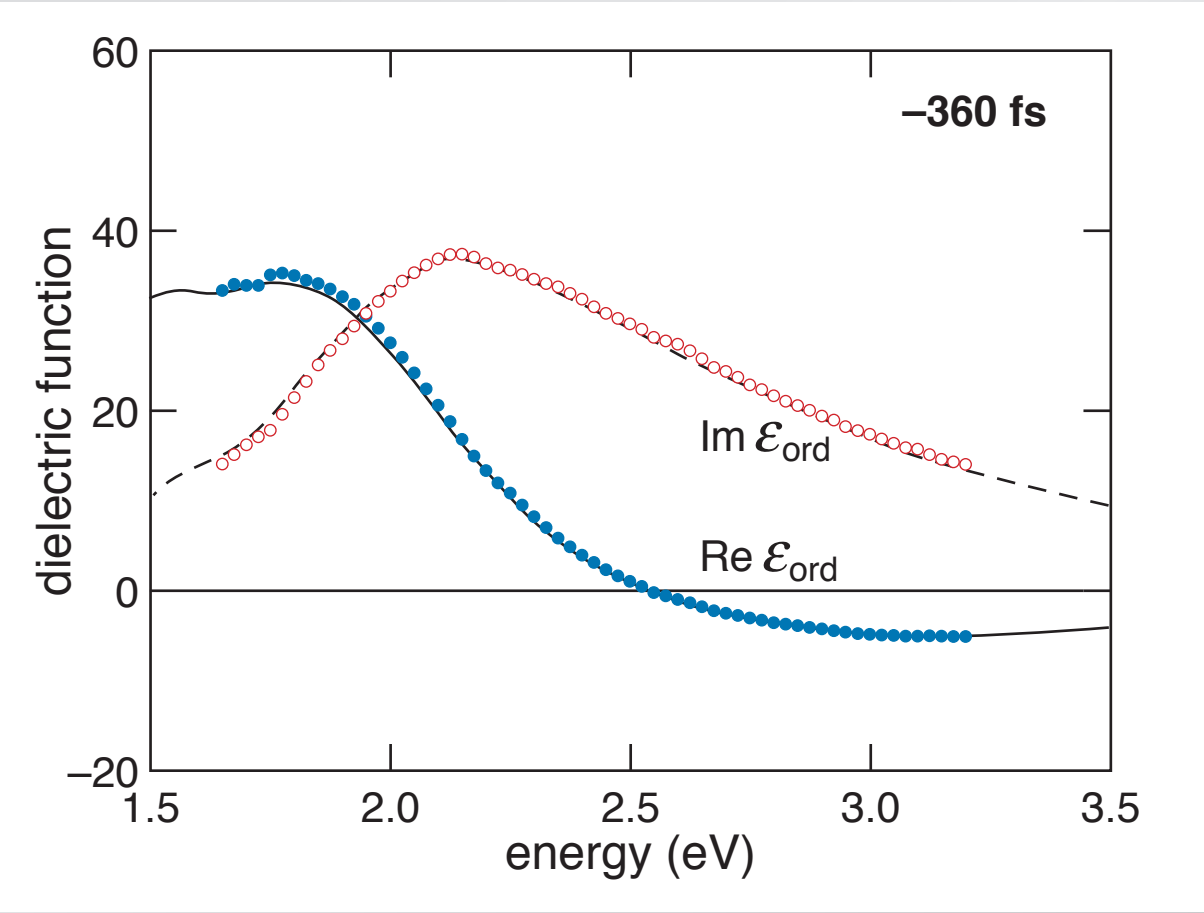
dielectric function



Coherent phonons

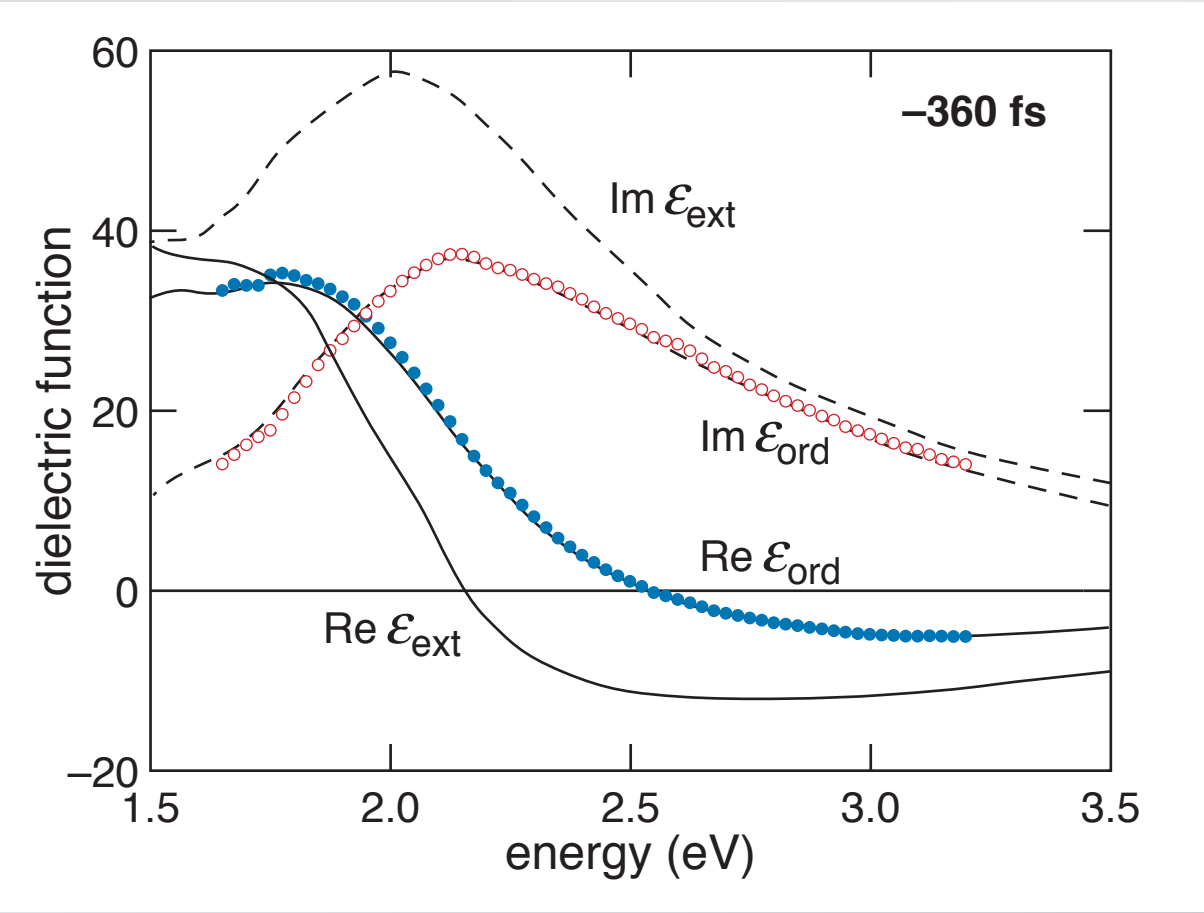


Coherent phonons



Phys. Rev. B 68, 012301 (2003)

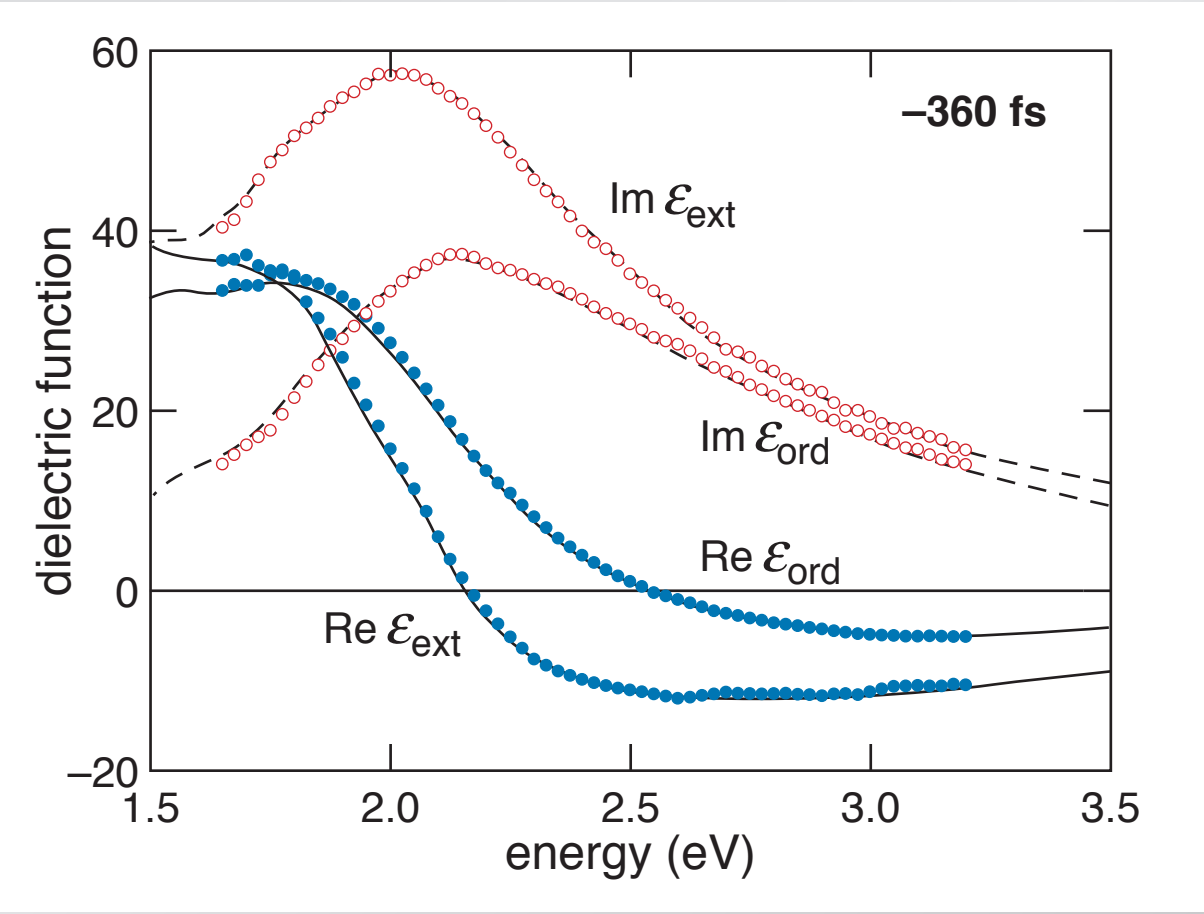
Coherent phonons



Phys. Rev. B 68, 012301 (2003)

Coherent phonons

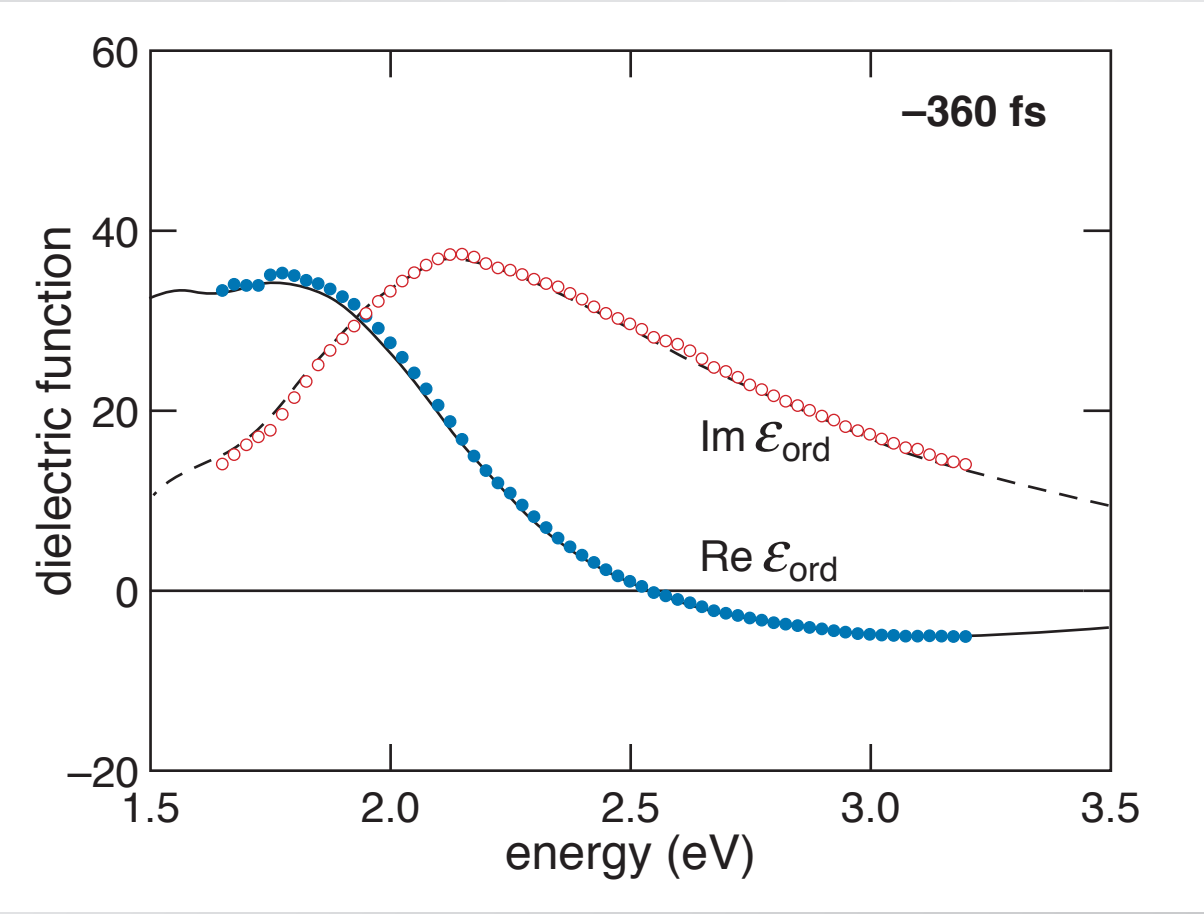
data agree well with literature values



Phys. Rev. B 68, 012301 (2003)

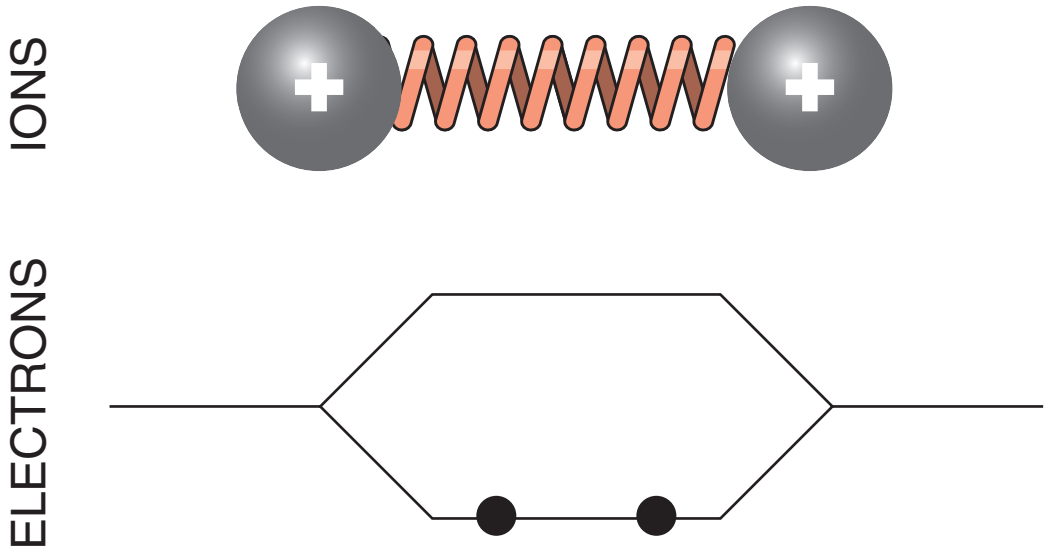
Coherent phonons

now vary pump probe delay



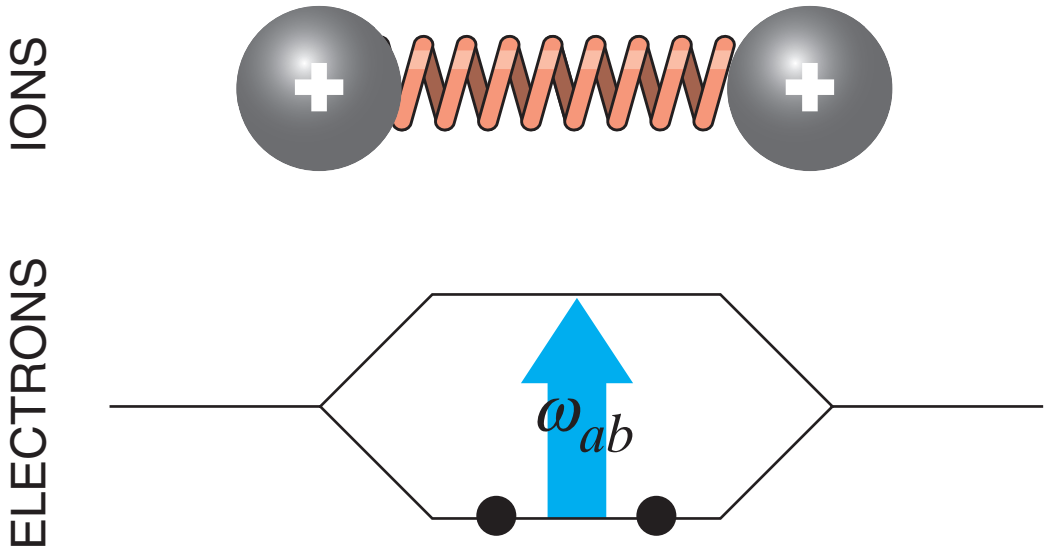
Phys. Rev. B 68, 012301 (2003)

Coherent phonons



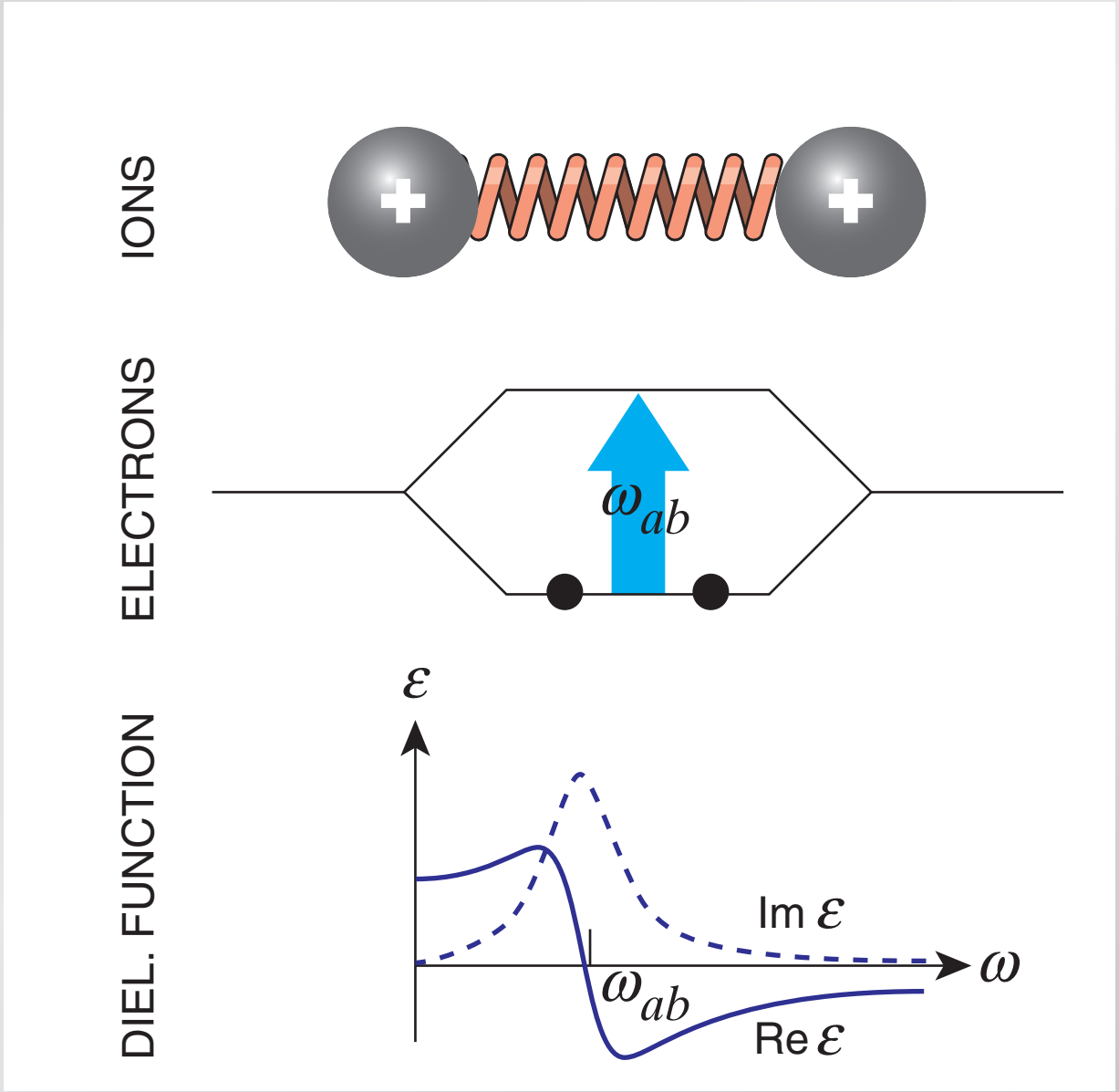
"two-atom model"

Coherent phonons



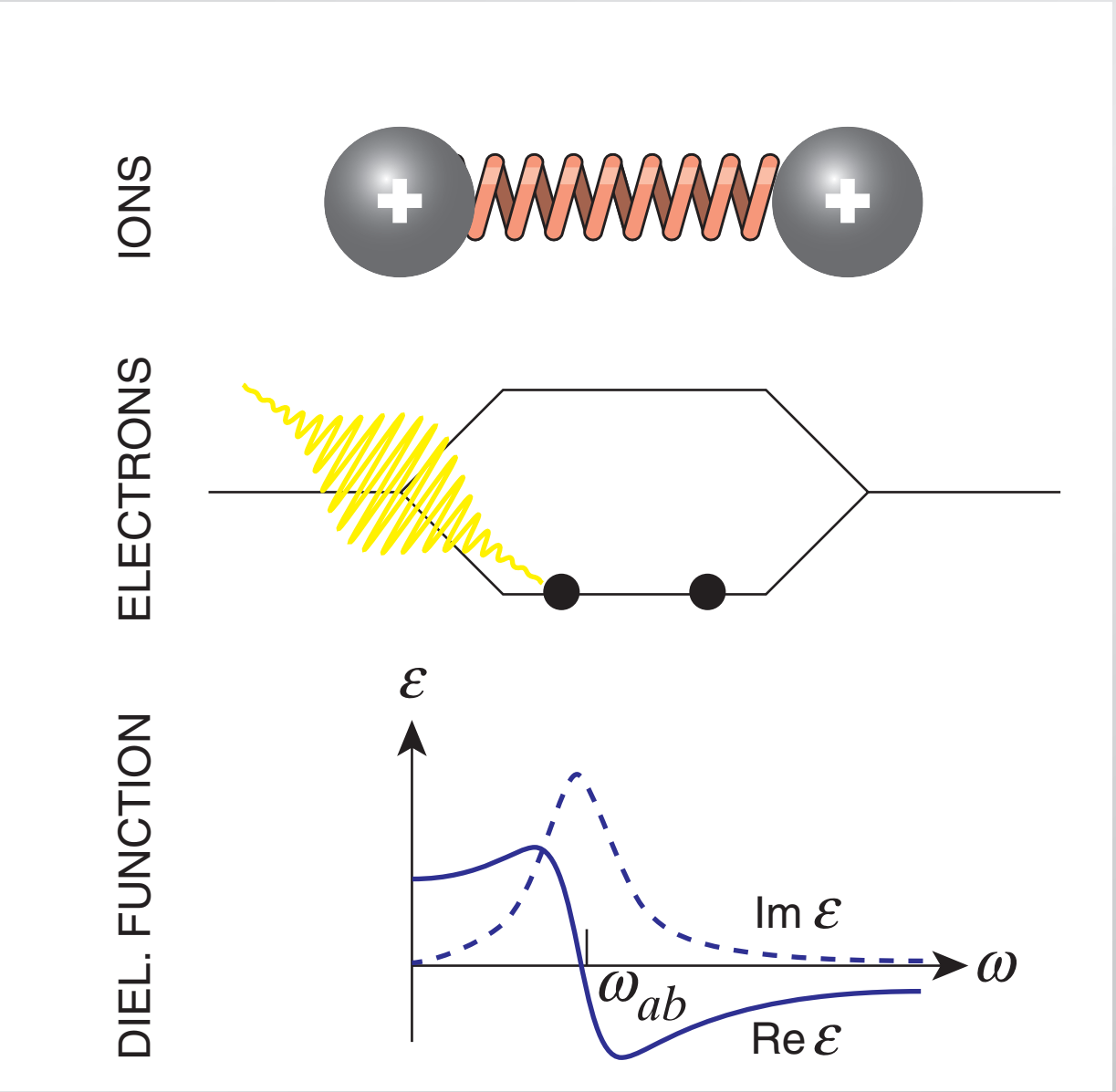
bonding-antibonding splitting

Coherent phonons



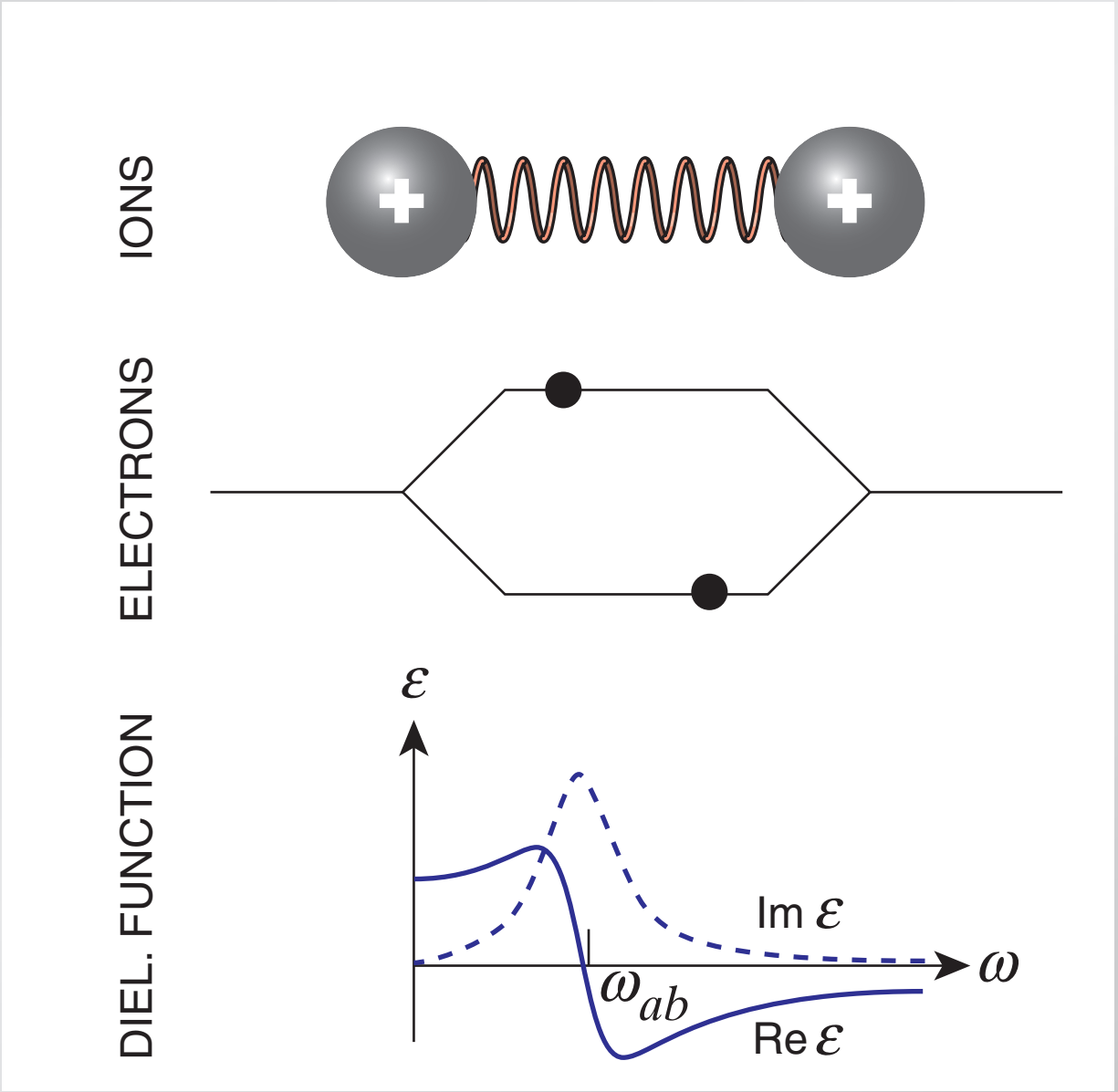
Lorentz model

Coherent phonons



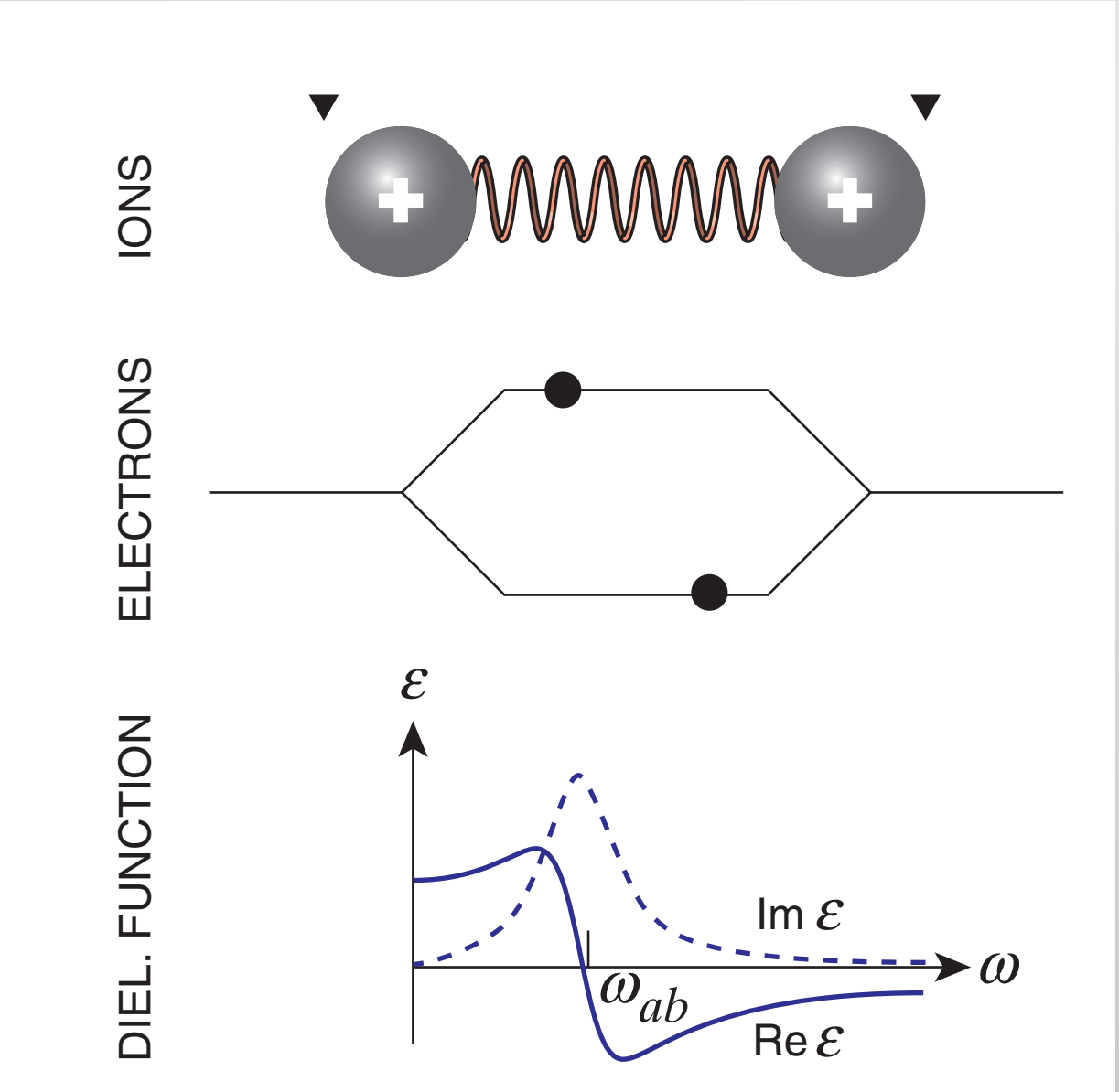
photon promotes electron…

Coherent phonons



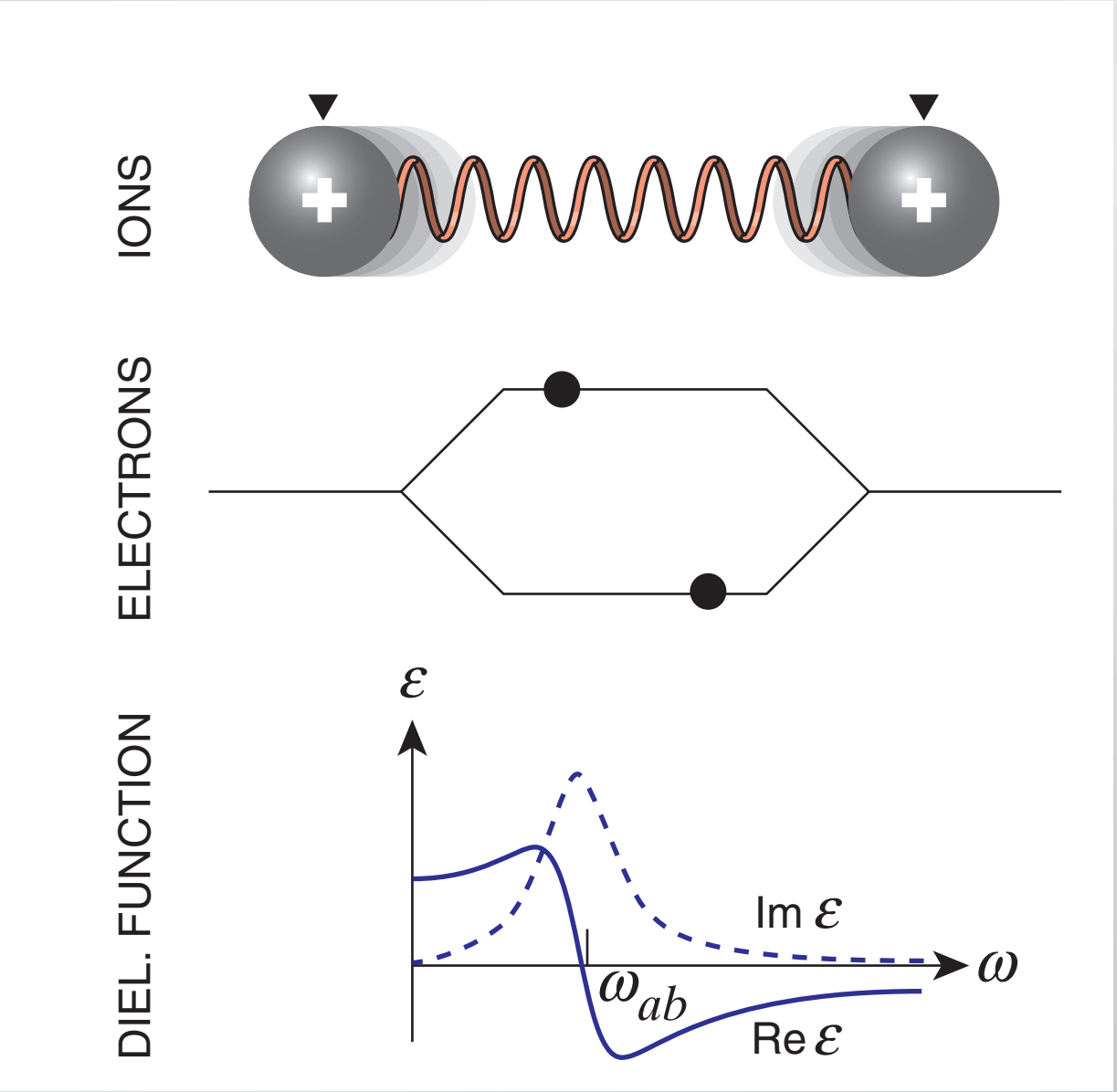
…weakening binding force…

Coherent phonons



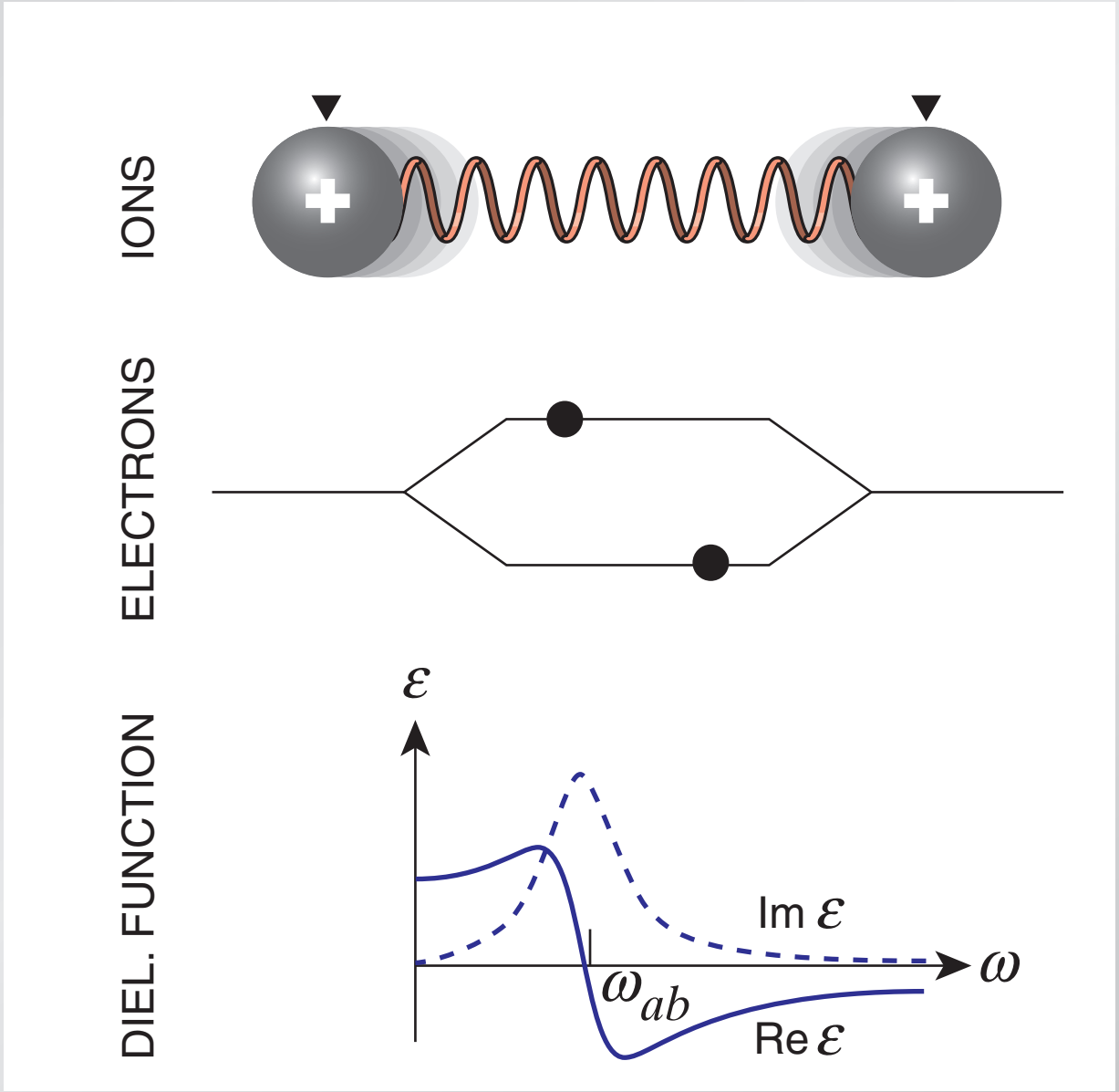
…establishing new equilibrium positions

Coherent phonons



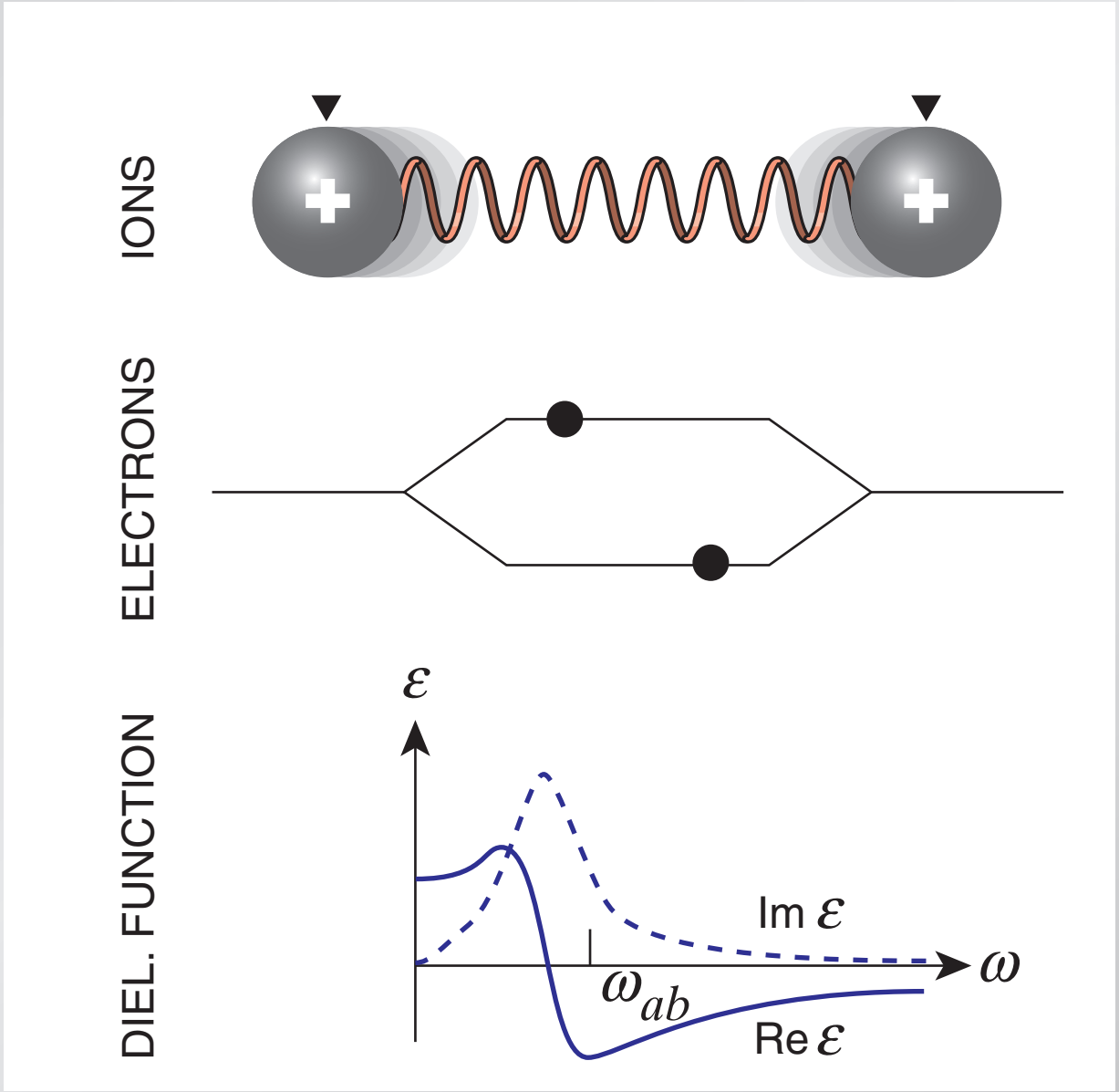
ions move to new equilibrium positions…

Coherent phonons



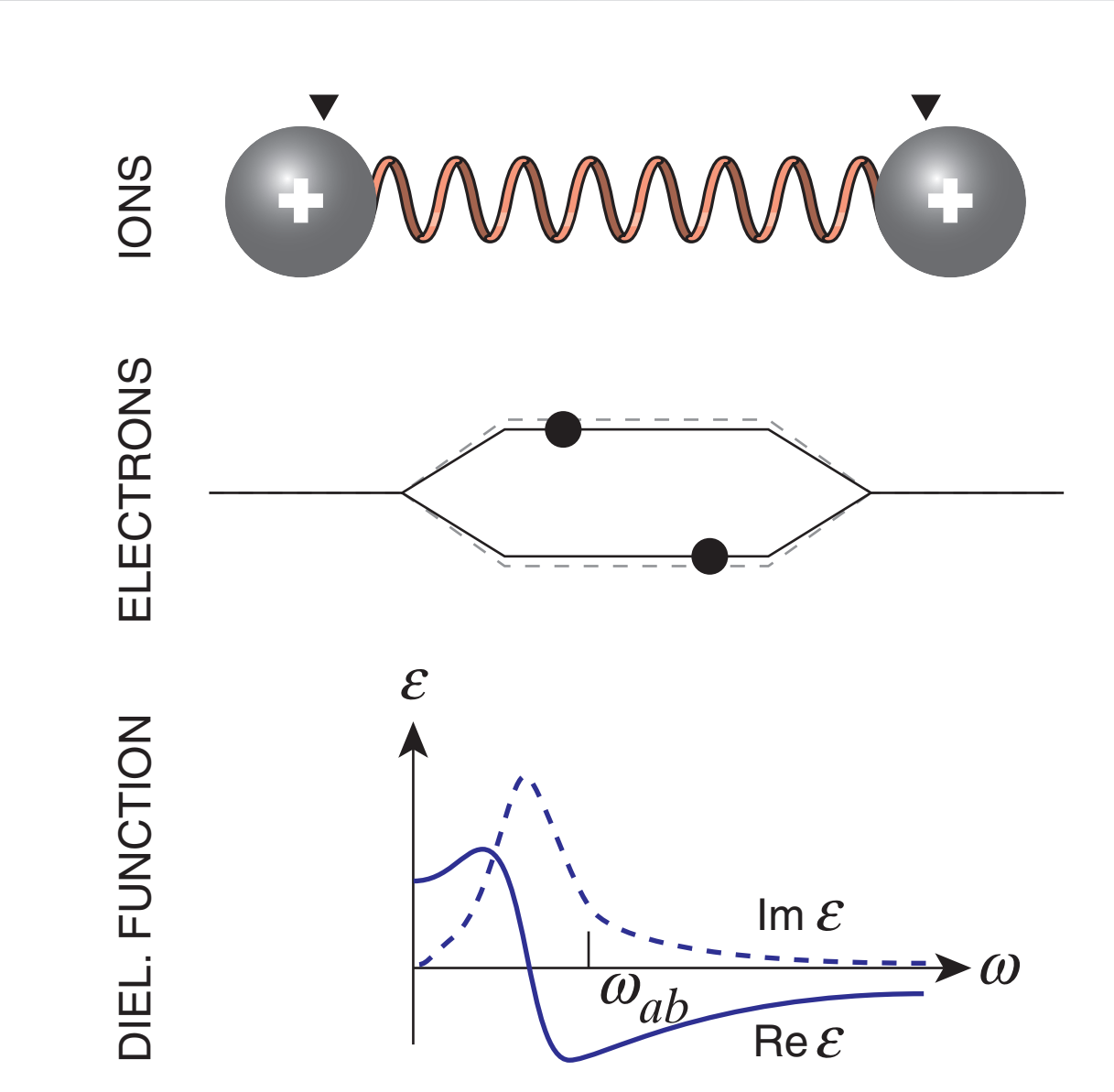
…diminishing splitting…

Coherent phonons



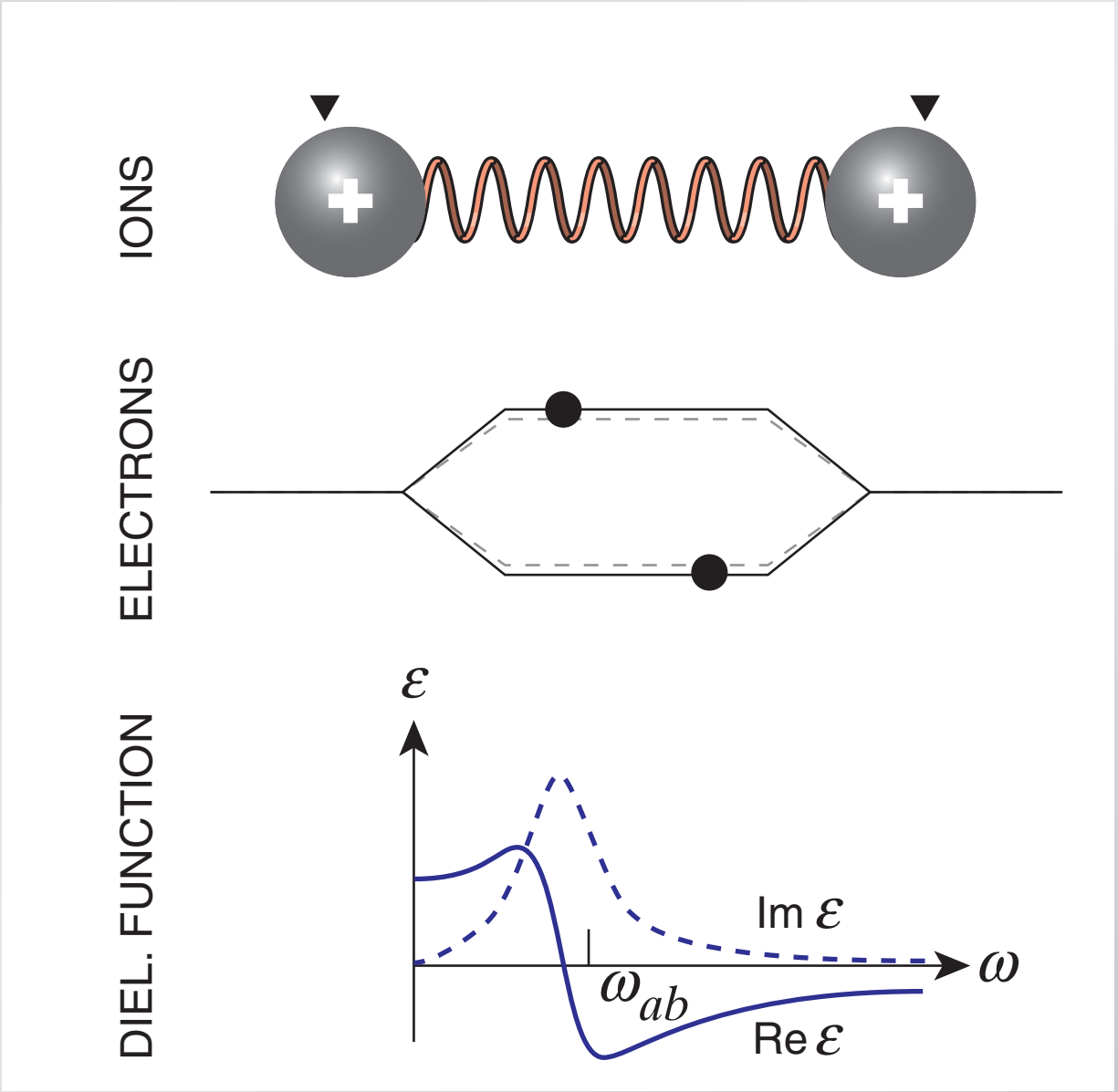
…and red-shifting the dielectric function

Coherent phonons



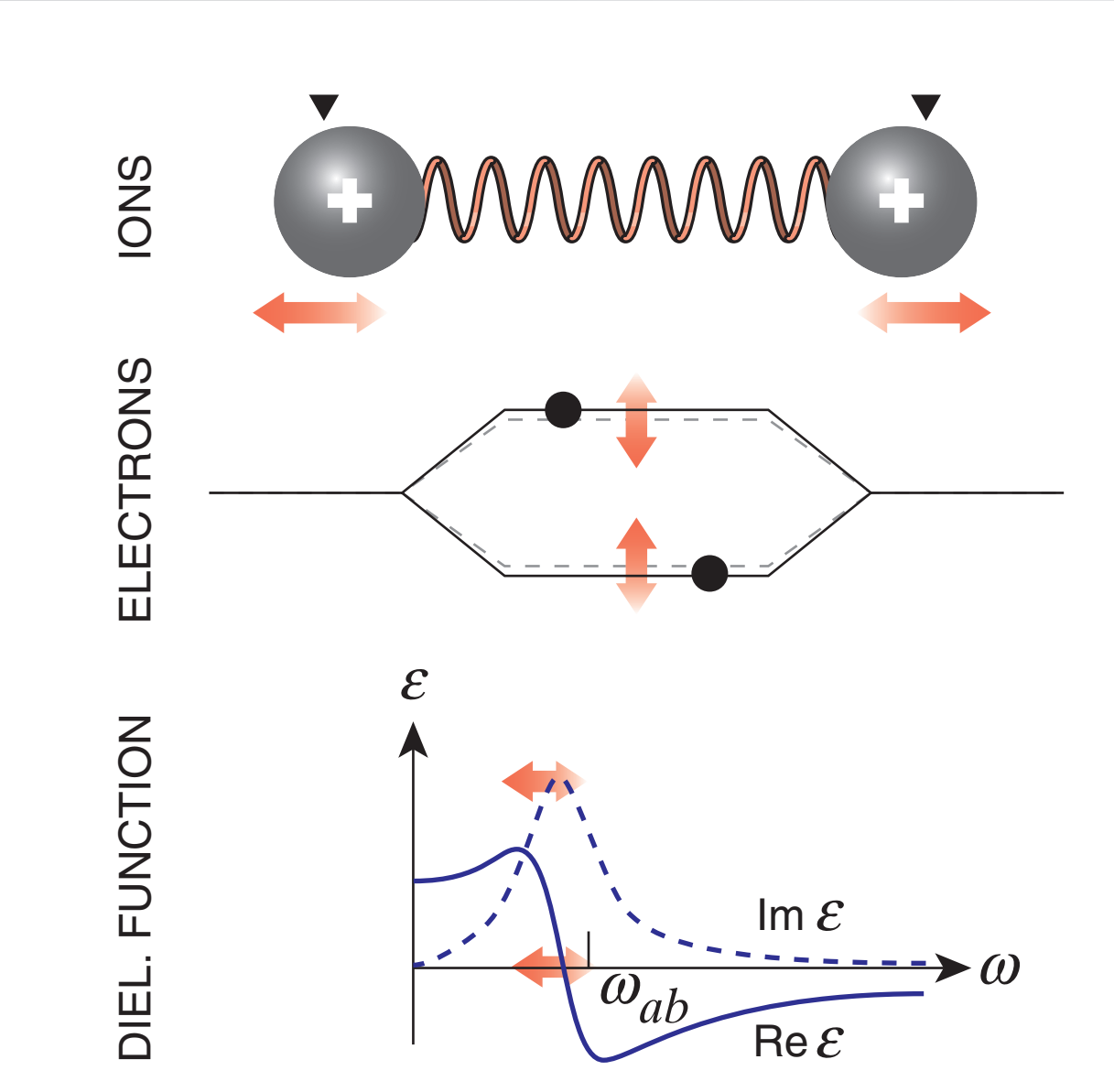
ions overshoot equilibrium position…

Coherent phonons



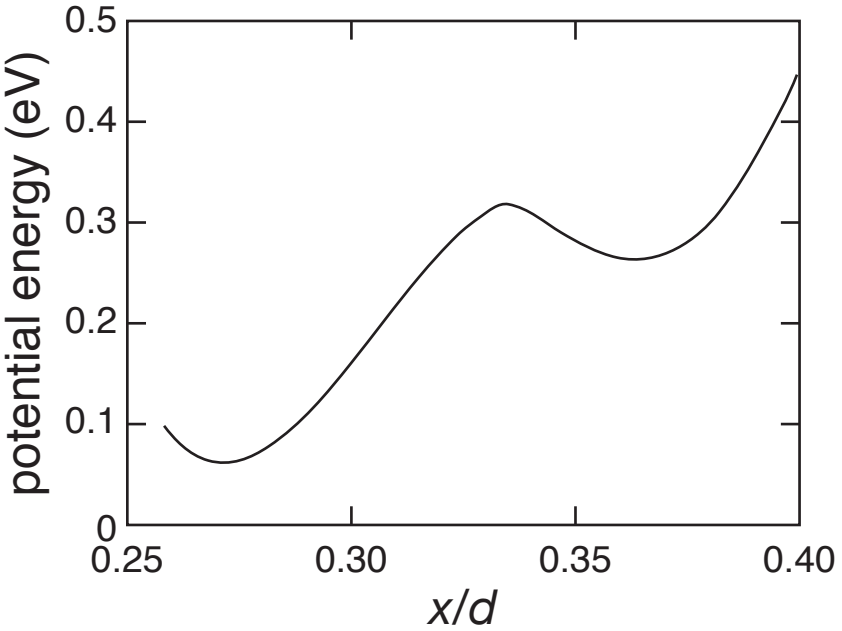
…reversing travel and overshooting again

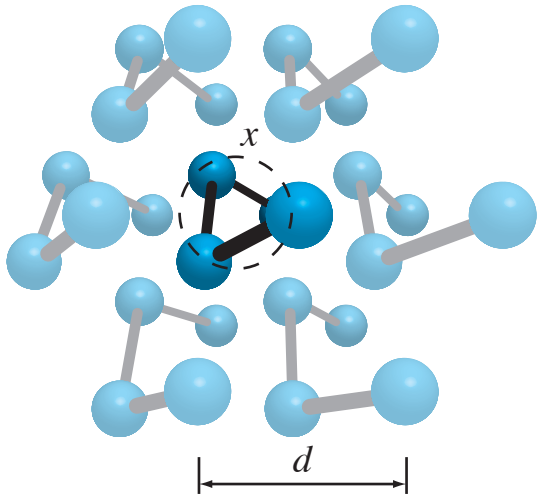
Coherent phonons



oscillation around “displaced” equilibrium

Coherent phonons

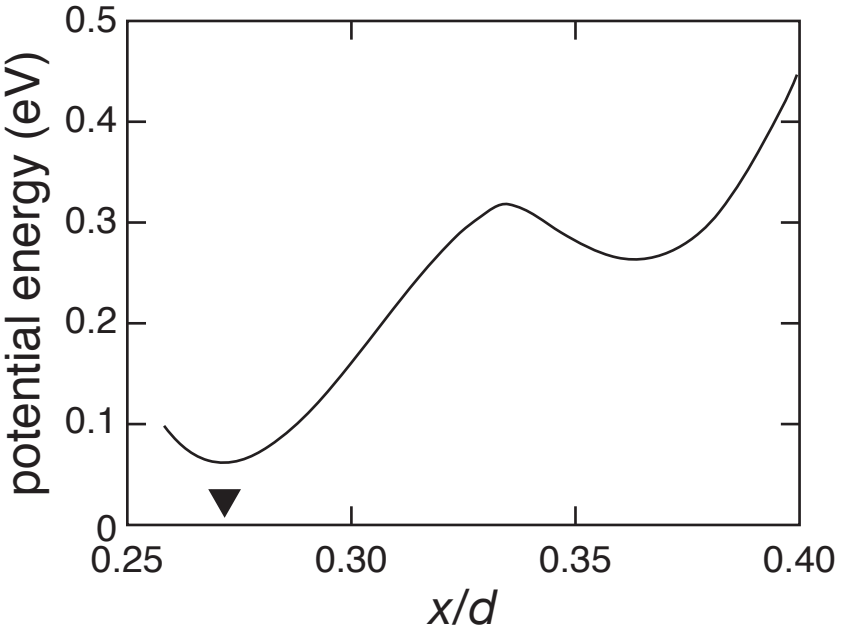


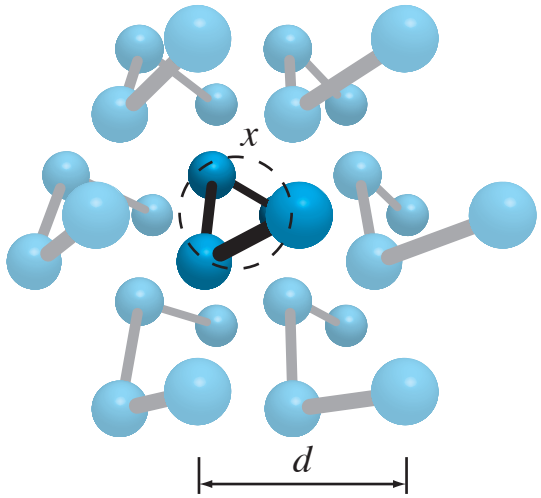


Tangney and Fahy, *Phys. Rev. B 65*, 054302 (2002)

Coherent phonons

ground state equilibrium at $x/d = 0.27$

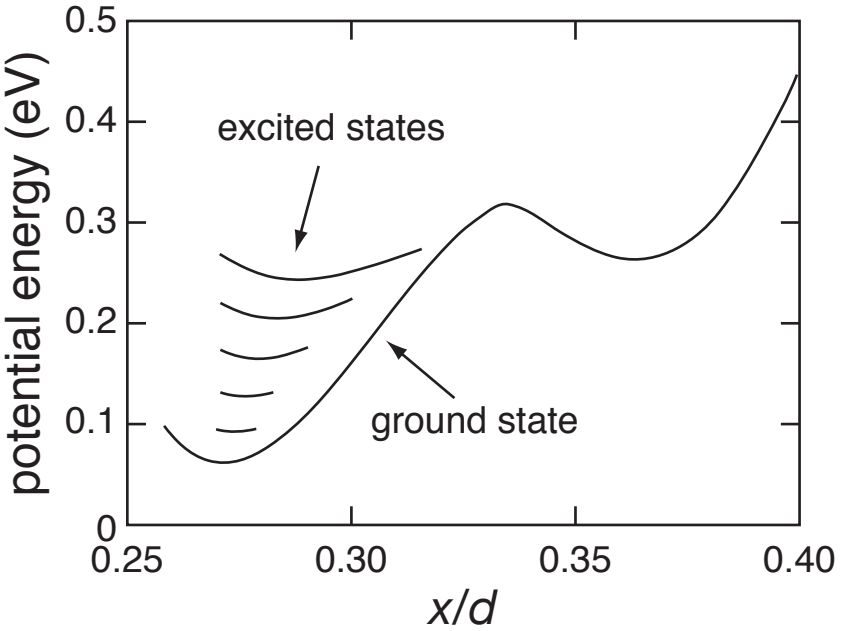


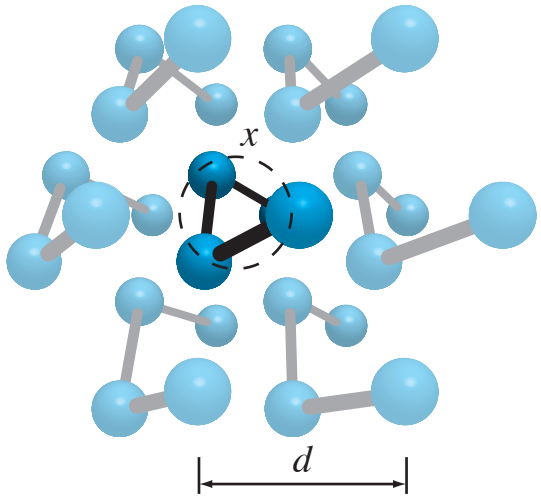


Tangney and Fahy, *Phys. Rev. B 65*, 054302 (2002)

Coherent phonons

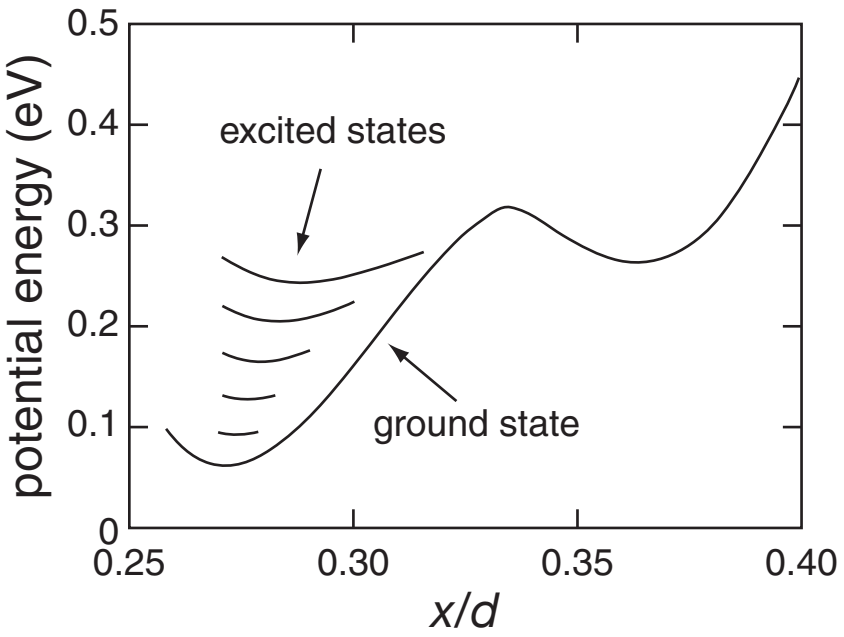
equilibrium position shifts upon excitation

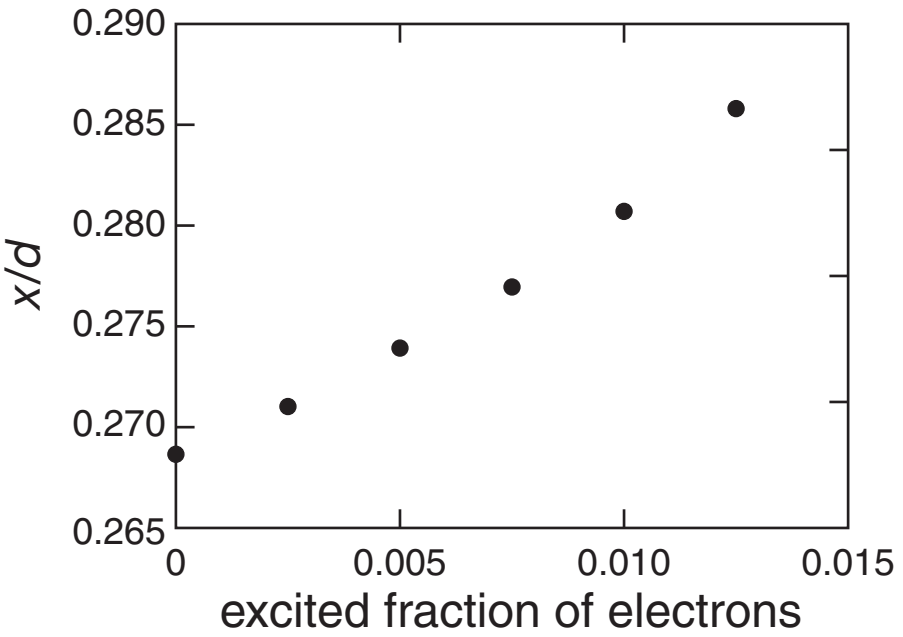




Tangney and Fahy, *Phys. Rev. B 65*, 054302 (2002)

Coherent phonons

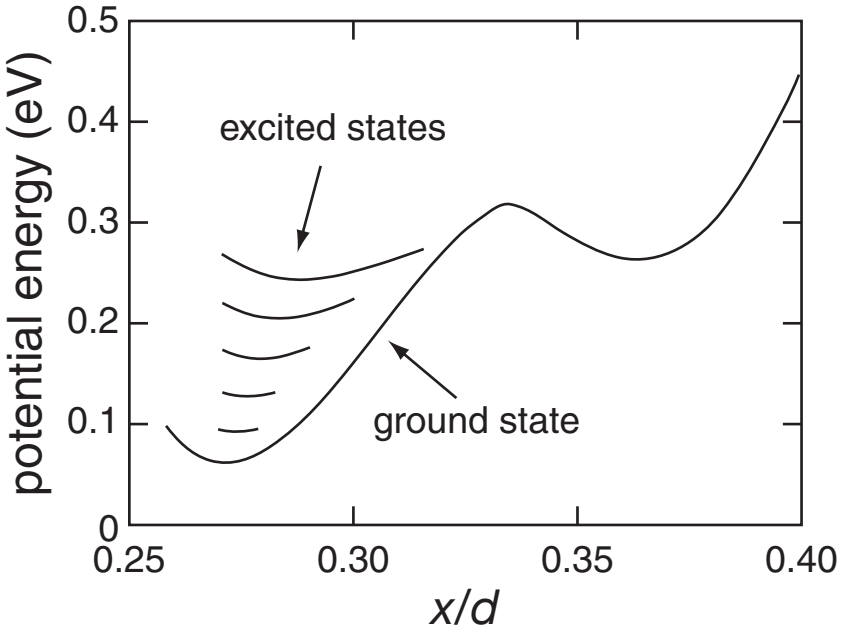


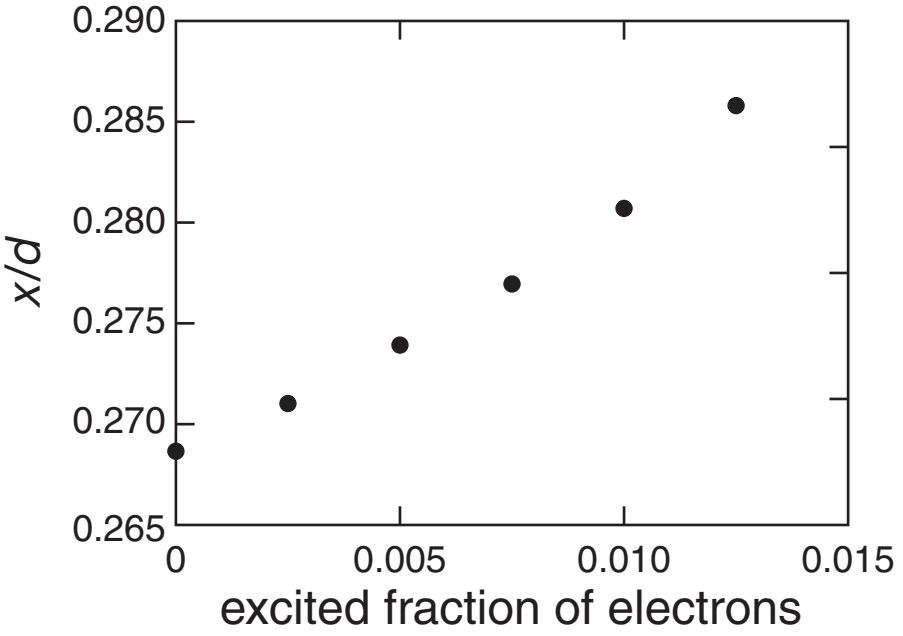


Tangney and Fahy, *Phys. Rev. B 65*, 054302 (2002)

Coherent phonons

band structure depends on lattice configuration

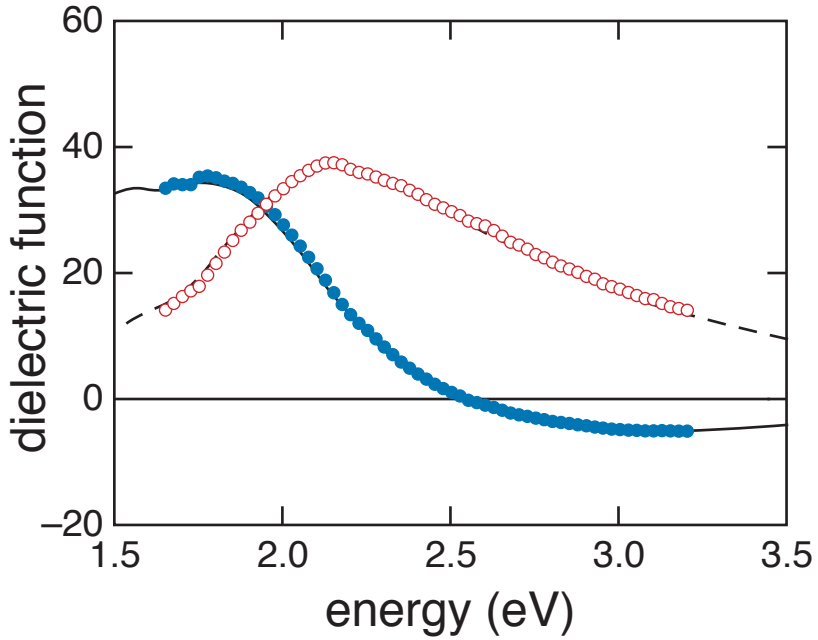




Tangney and Fahy, *Phys. Rev. B 65*, 054302 (2002)

Coherent phonons

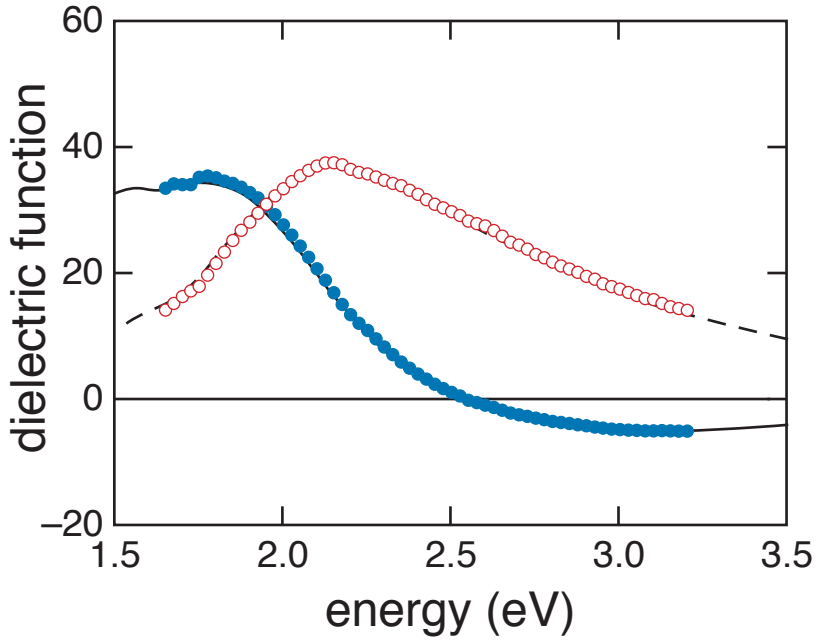
dielectric function reveals band structure changes

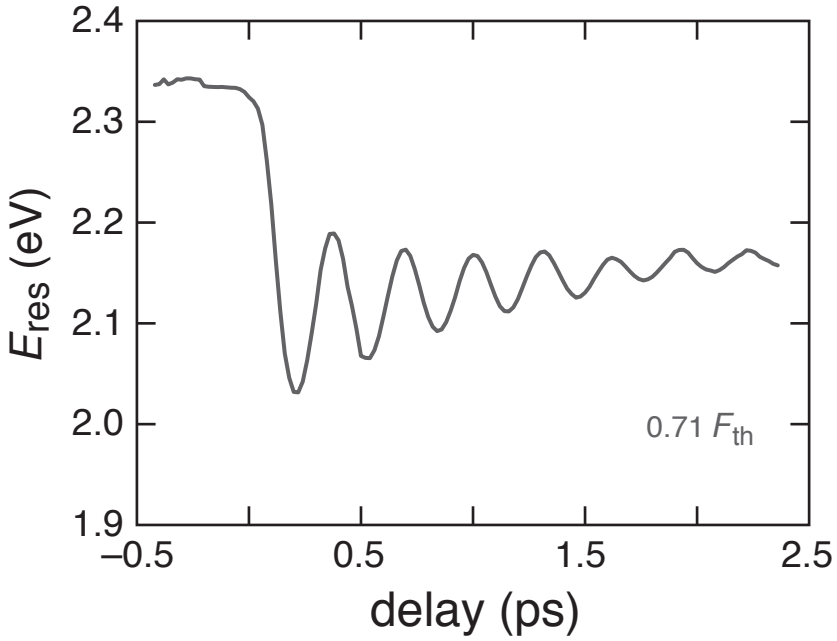


Phys. Rev. B 68, 012301 (2003)

Coherent phonons

track resonance energy





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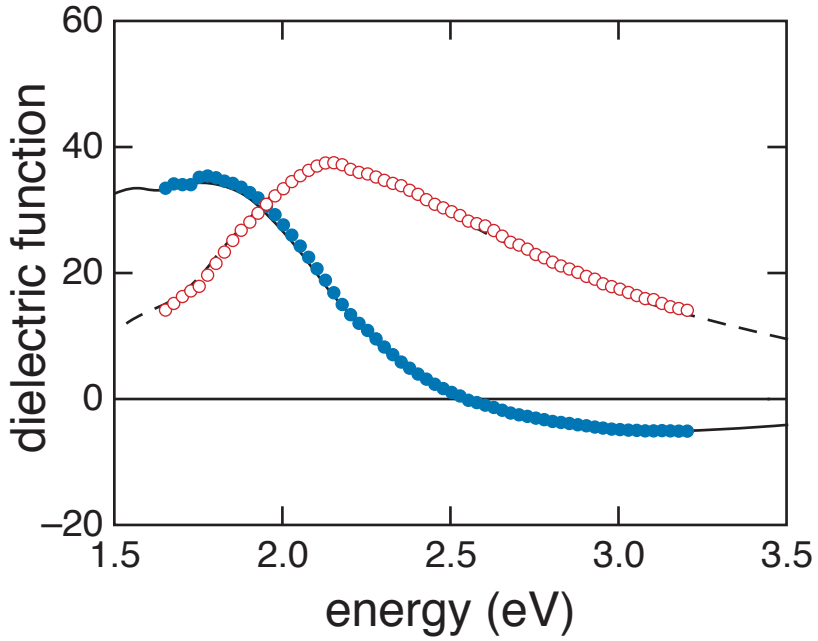
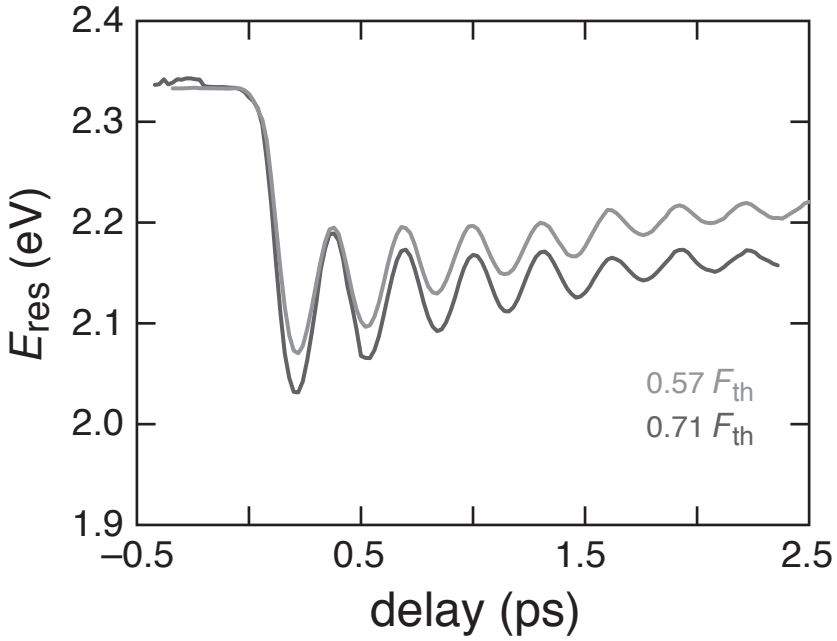
Coherent phonons

track resonance energy





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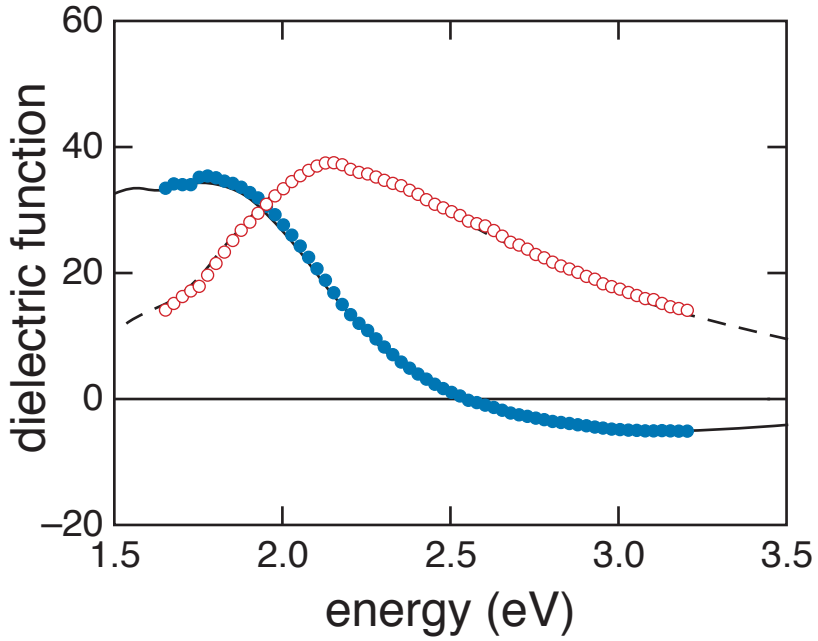
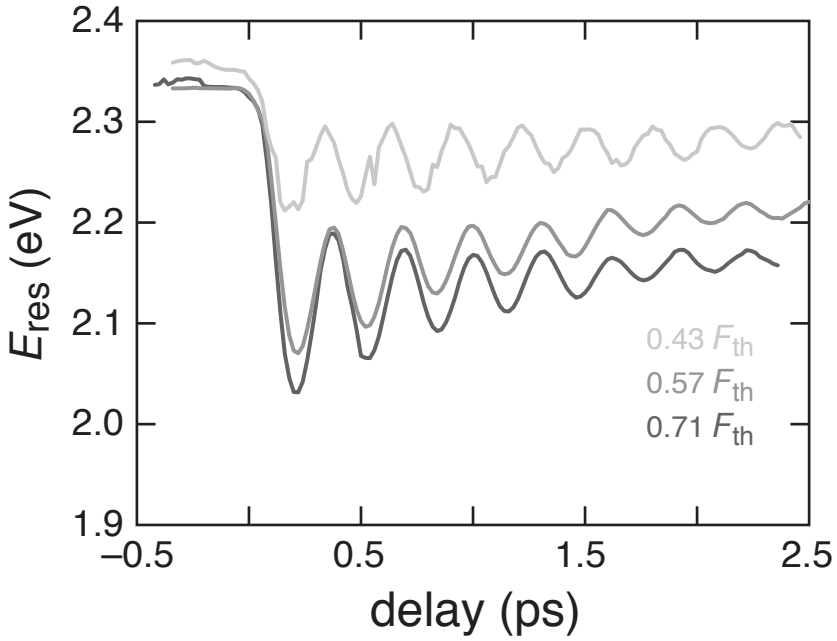
Coherent phonons

track resonance energy

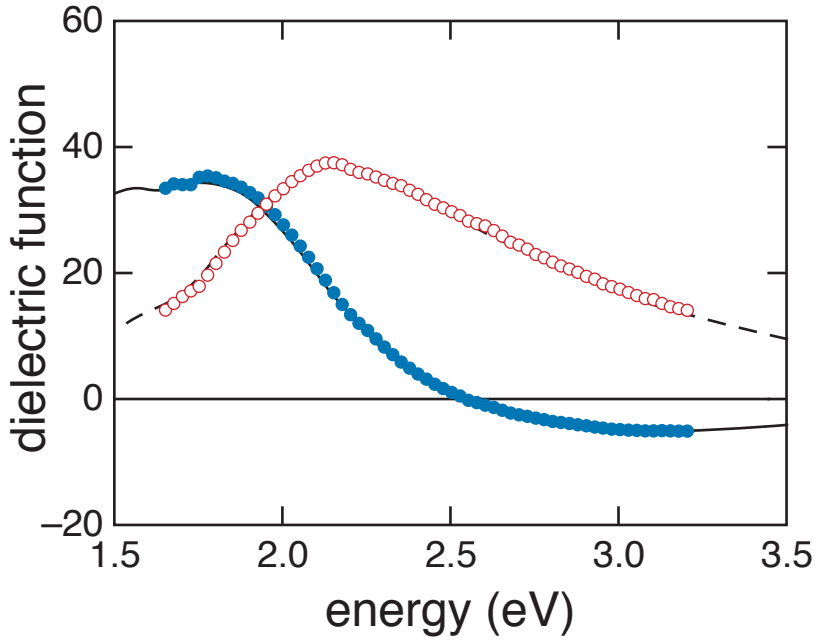


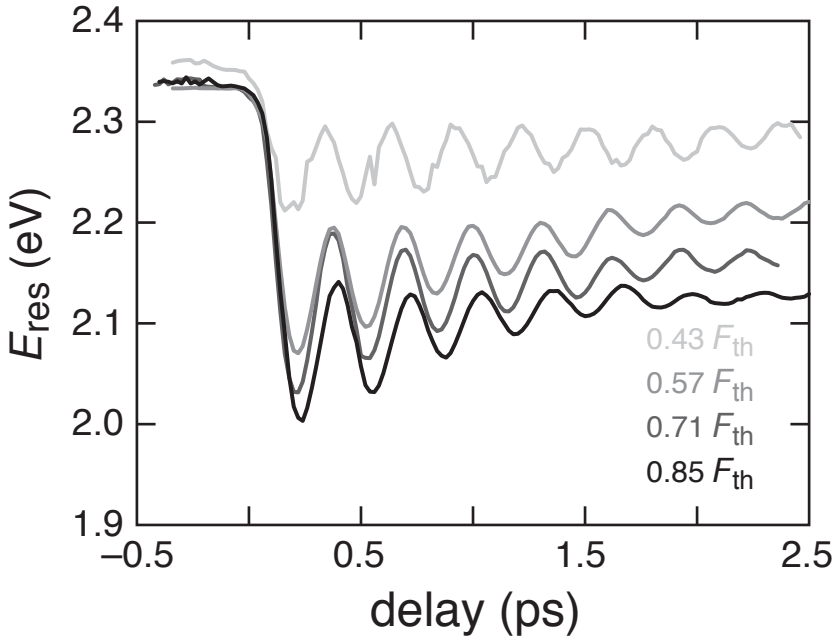


Phys. Rev. B 68, 012301 (2003)

Coherent phonons

track resonance energy





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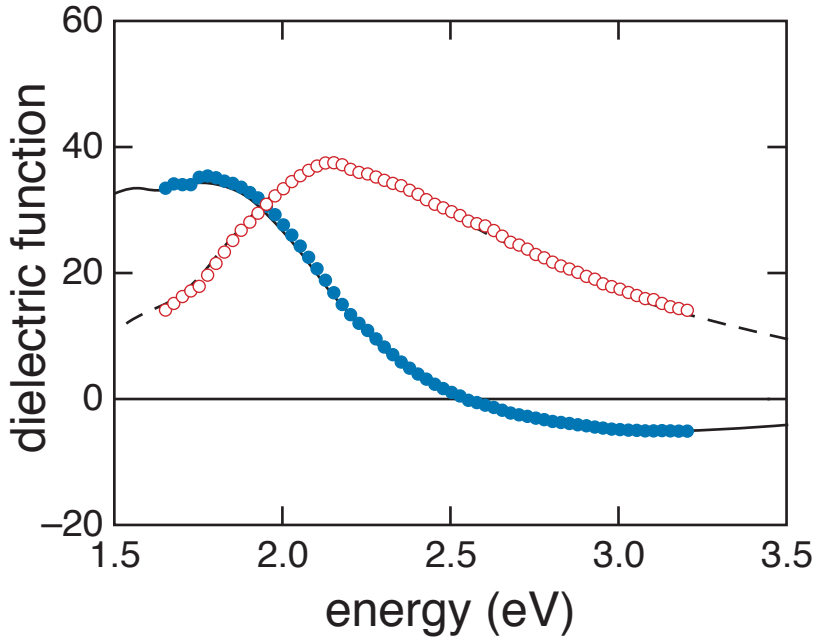
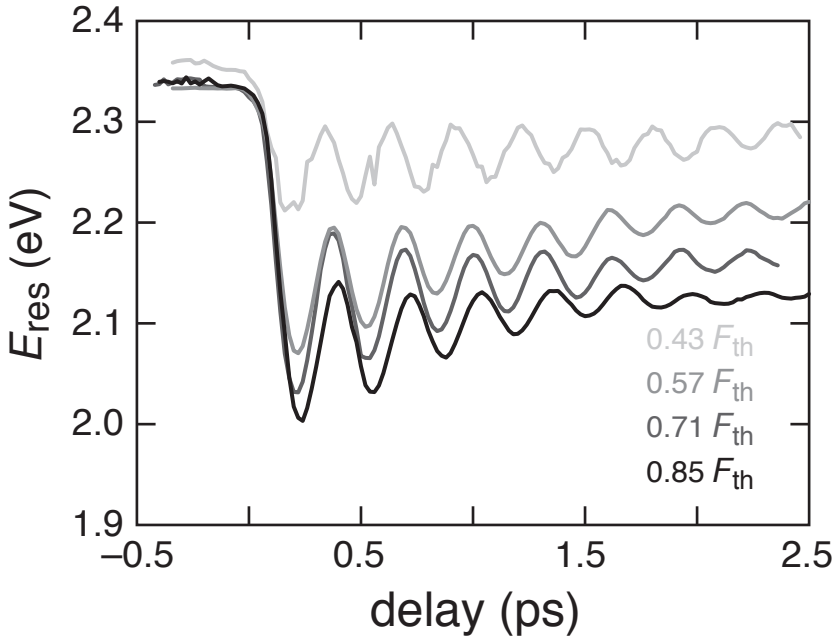
Coherent phonons

$\Delta E_{max} \approx 0.3$ eV and so $\Delta x/x \approx 0.05$





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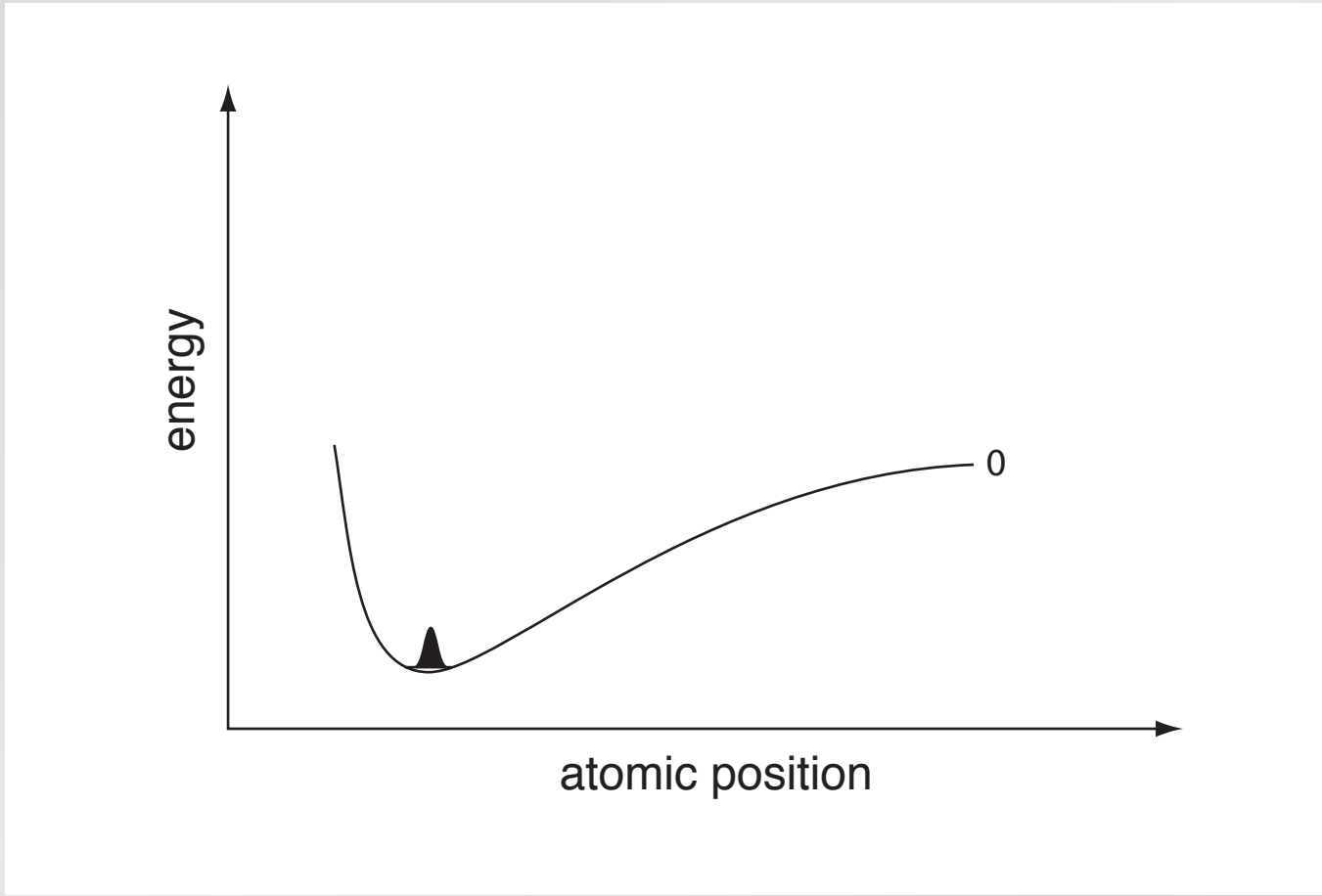
Outline

- experimental
- coherent phonons
- optical control

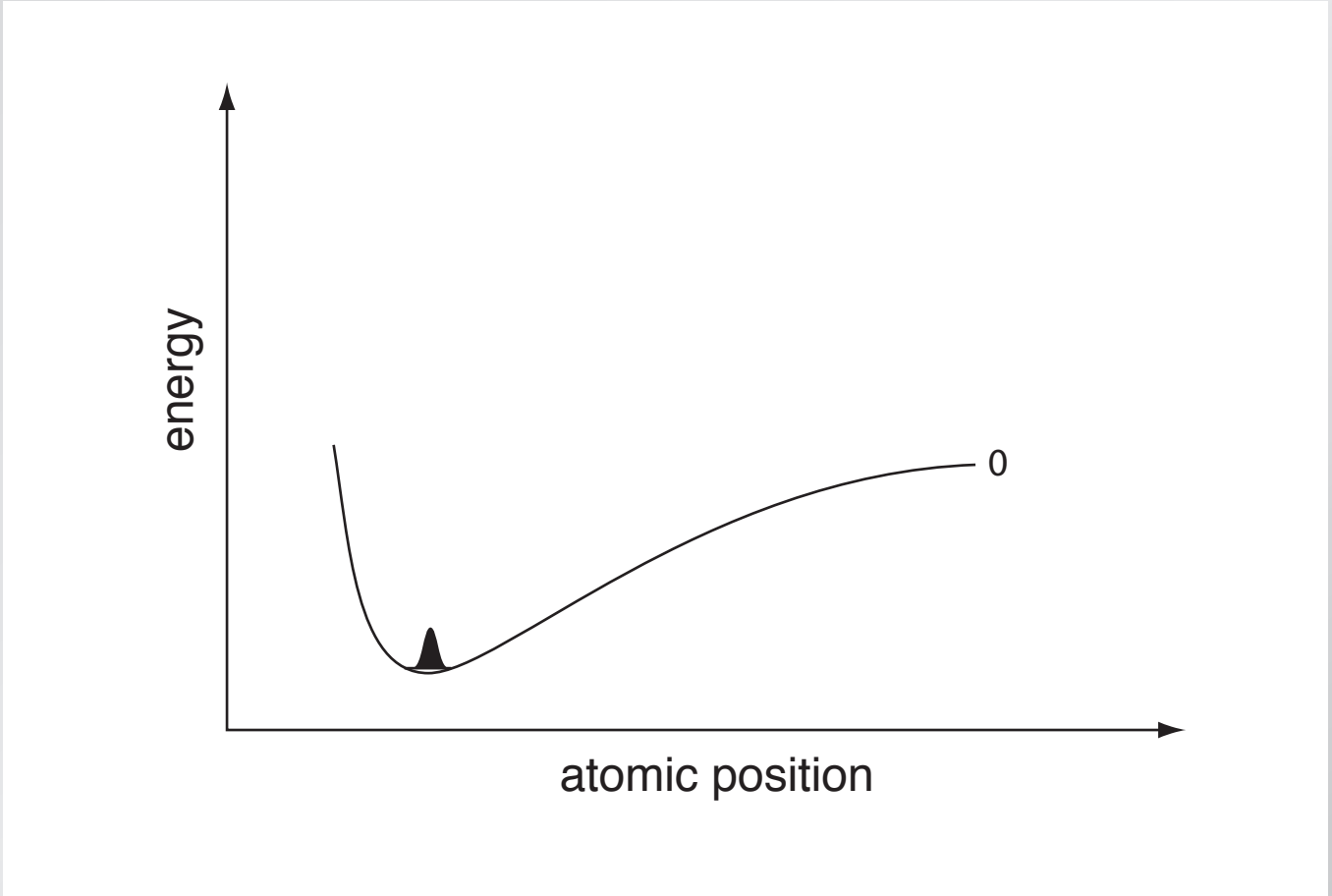
Optical control

semiclassical model of nuclear motion



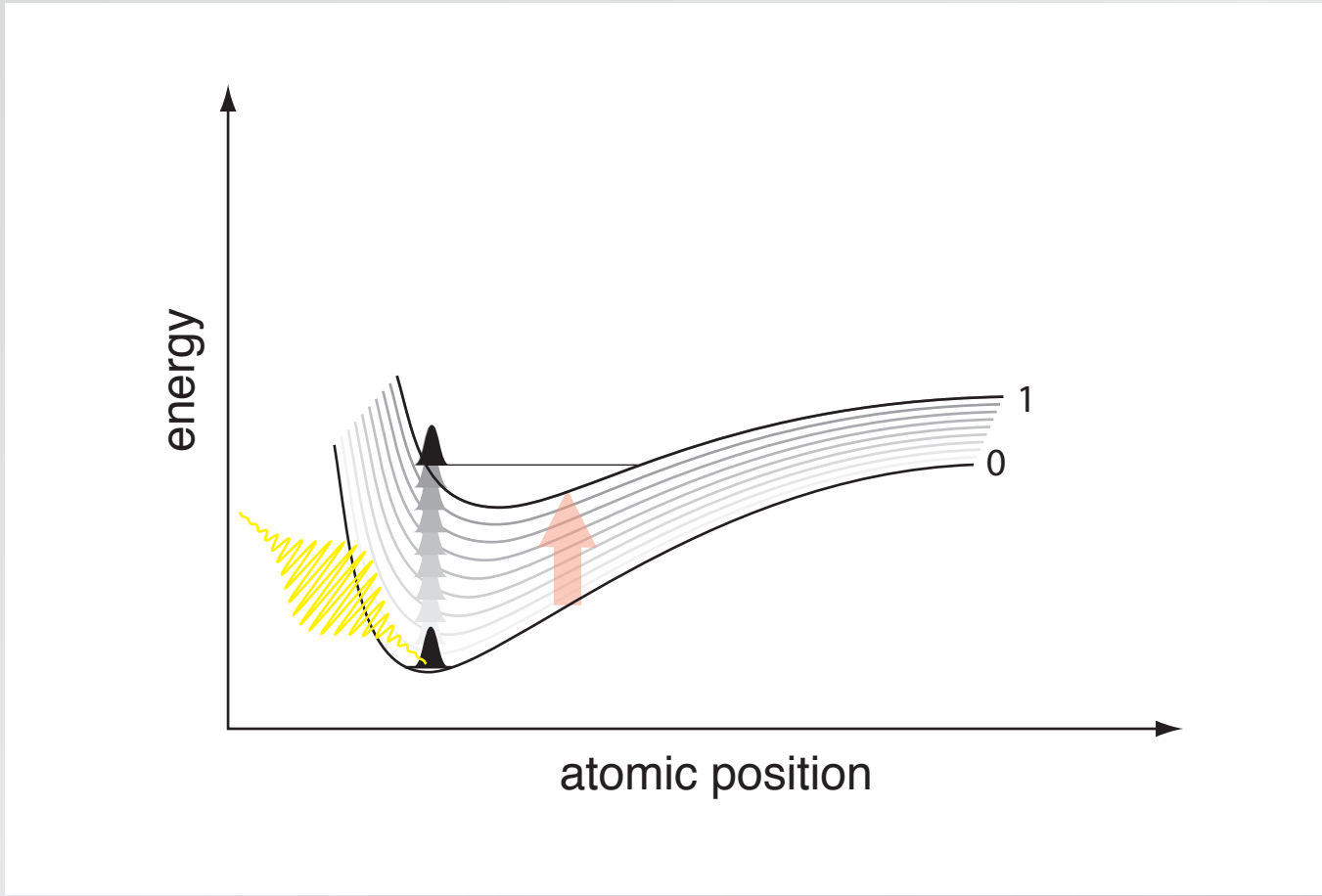
Optical control

nuclear wave packet sits at minimum



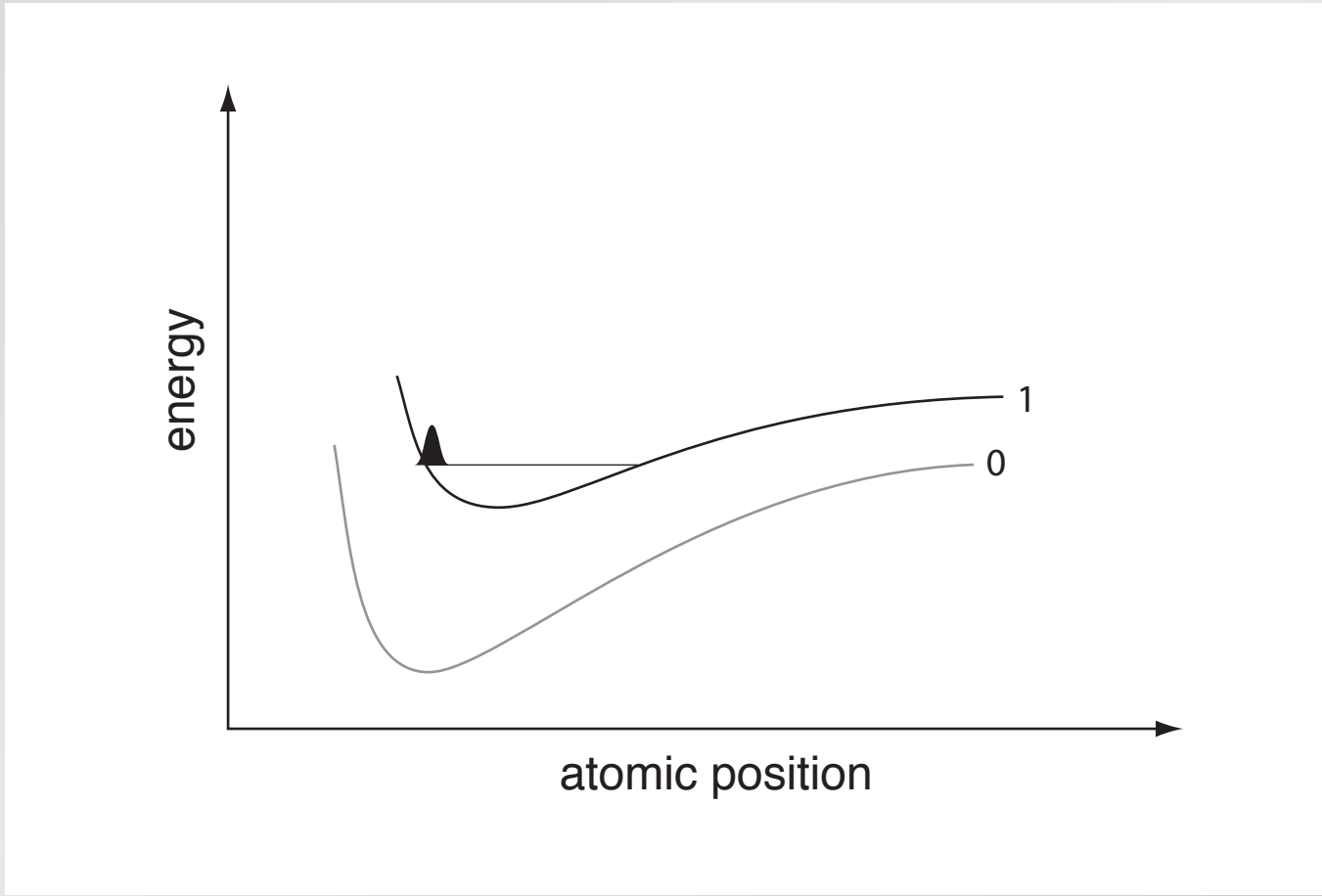
Optical control

laser pulse excites electrons, alters potential



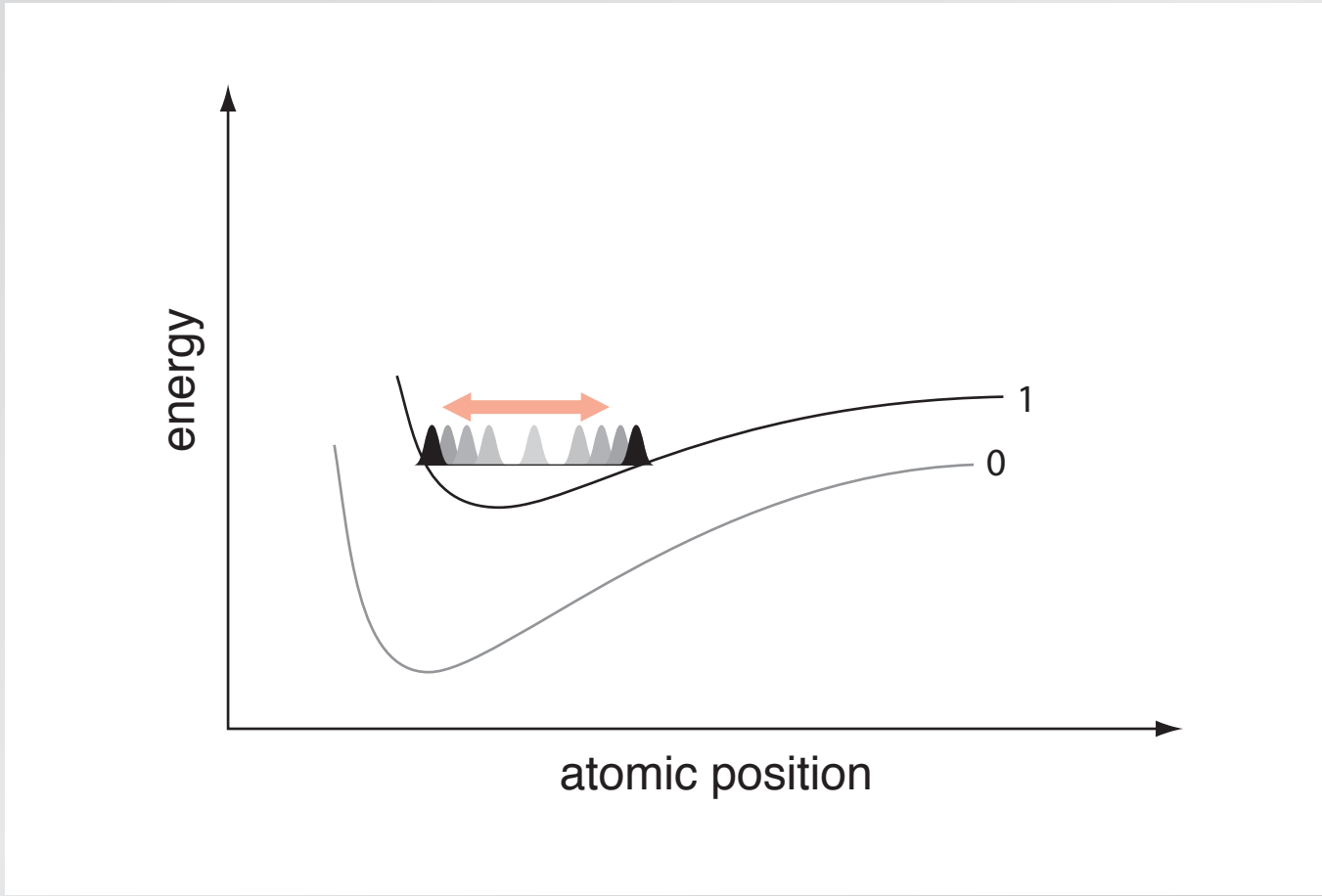
Optical control

nuclear wave packets on new potential



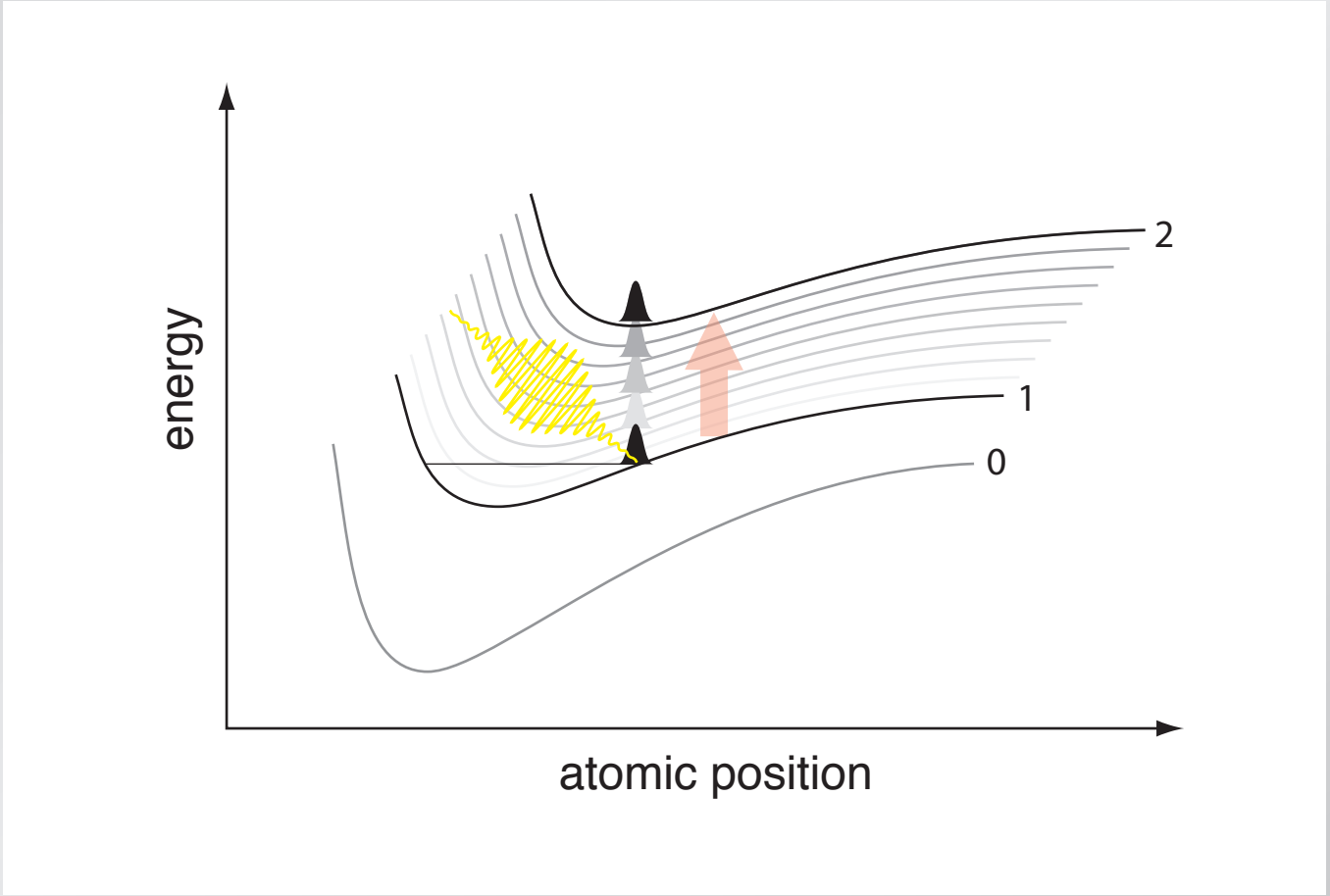
Optical control

wave packet oscillates on new potential



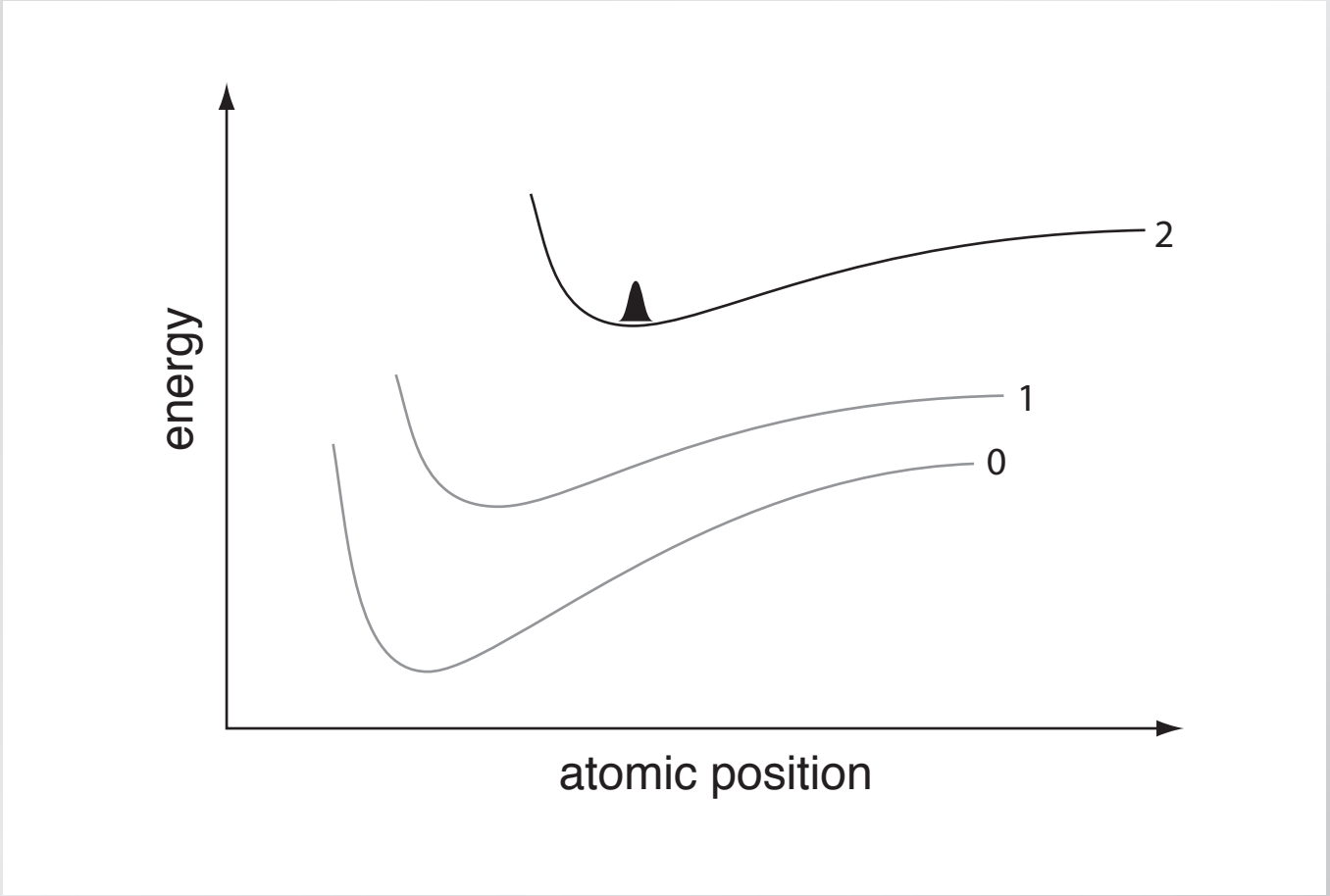
Optical control

excite again at turning point…



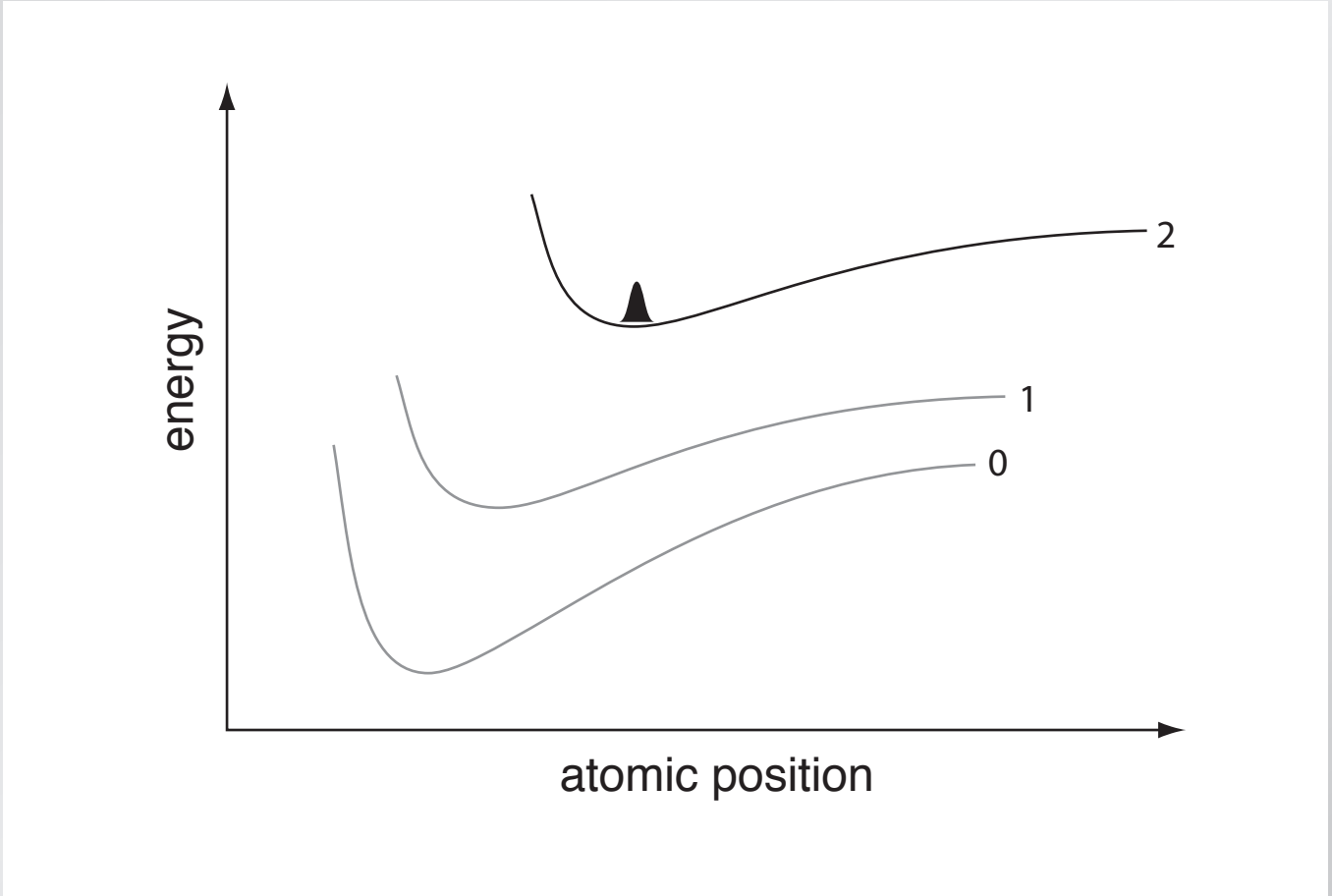
Optical control

…so wave packet lands at minimum in new potential



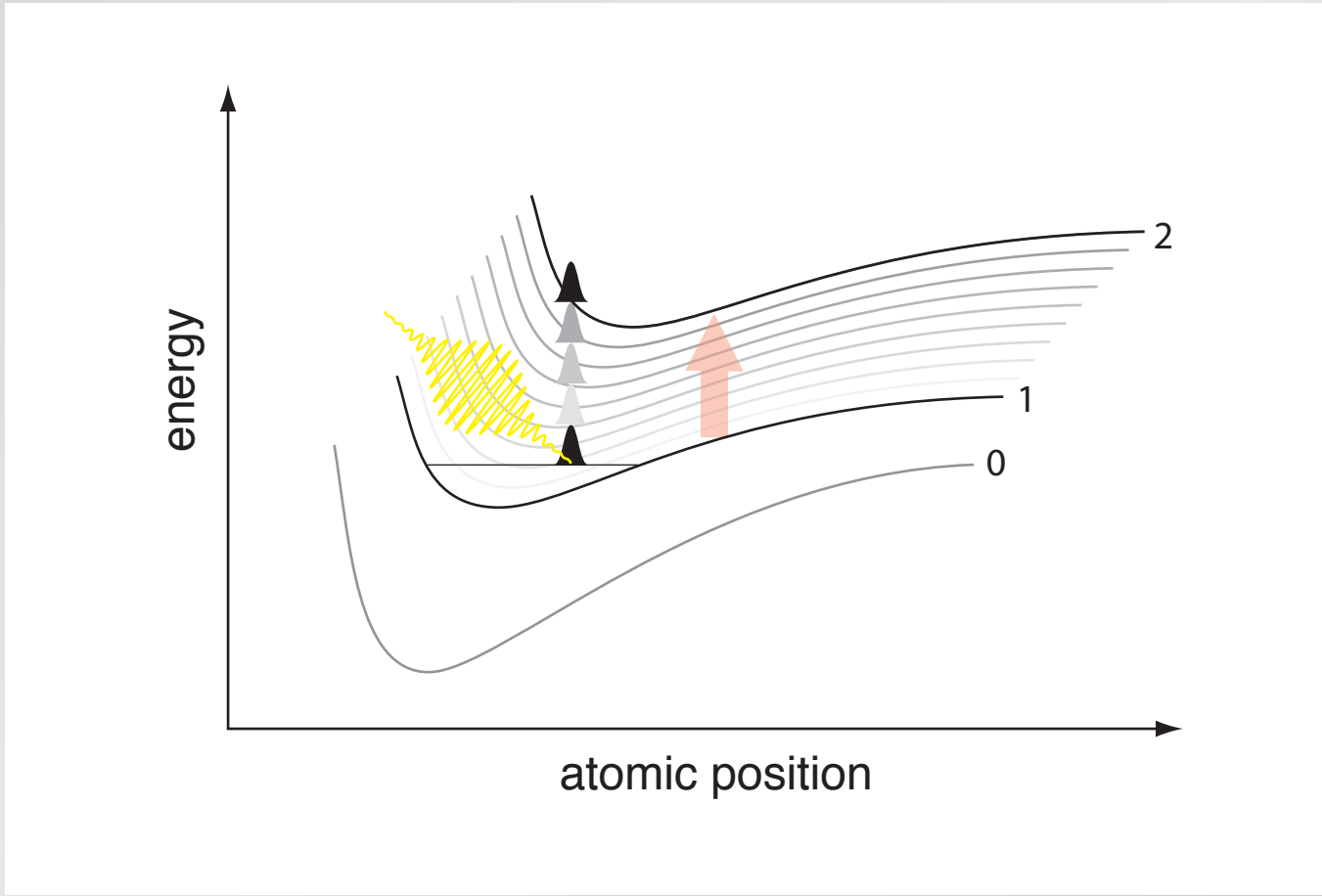
Optical control

leaving lattice displaced (without oscillations)



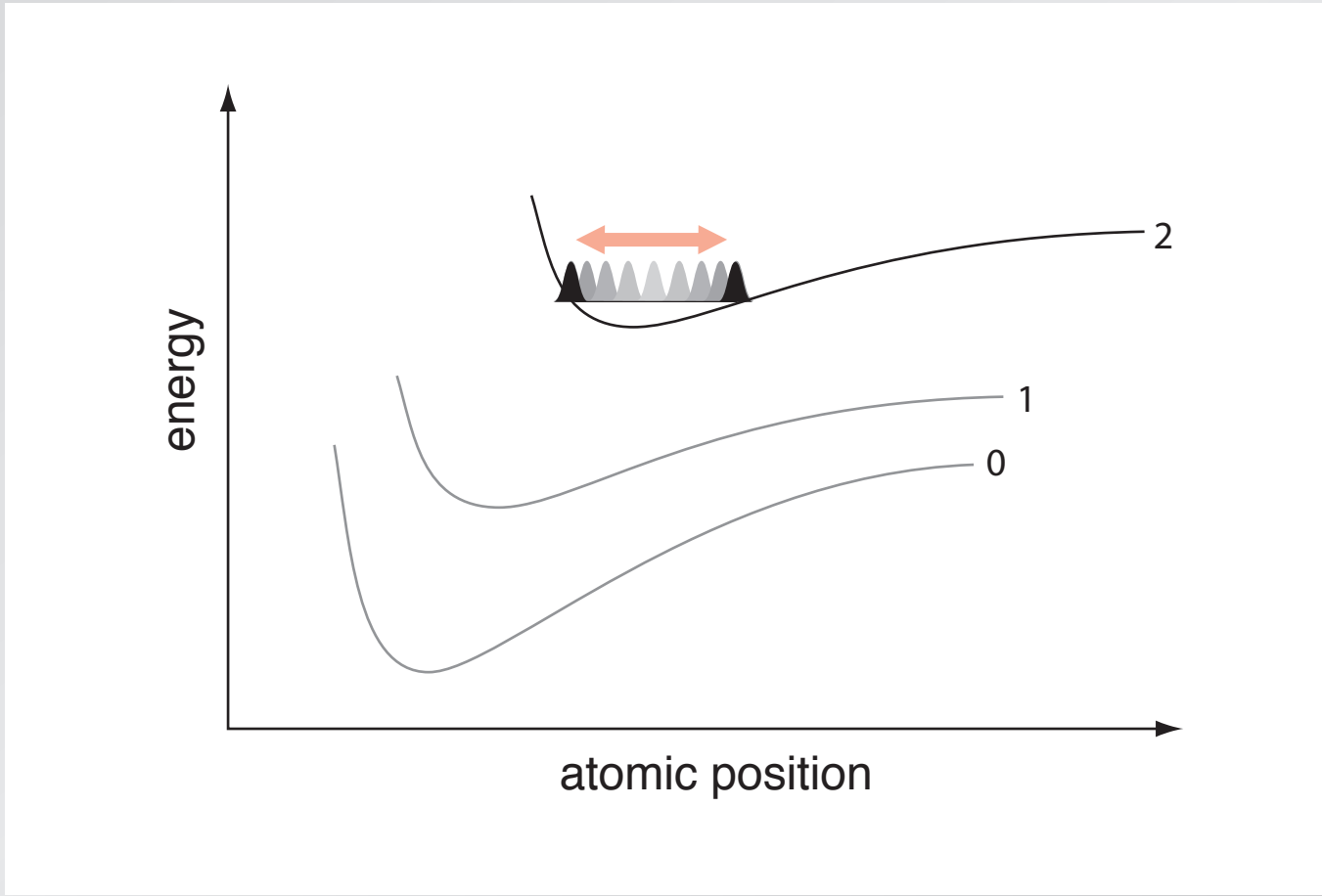
Optical control

if timing wrong…



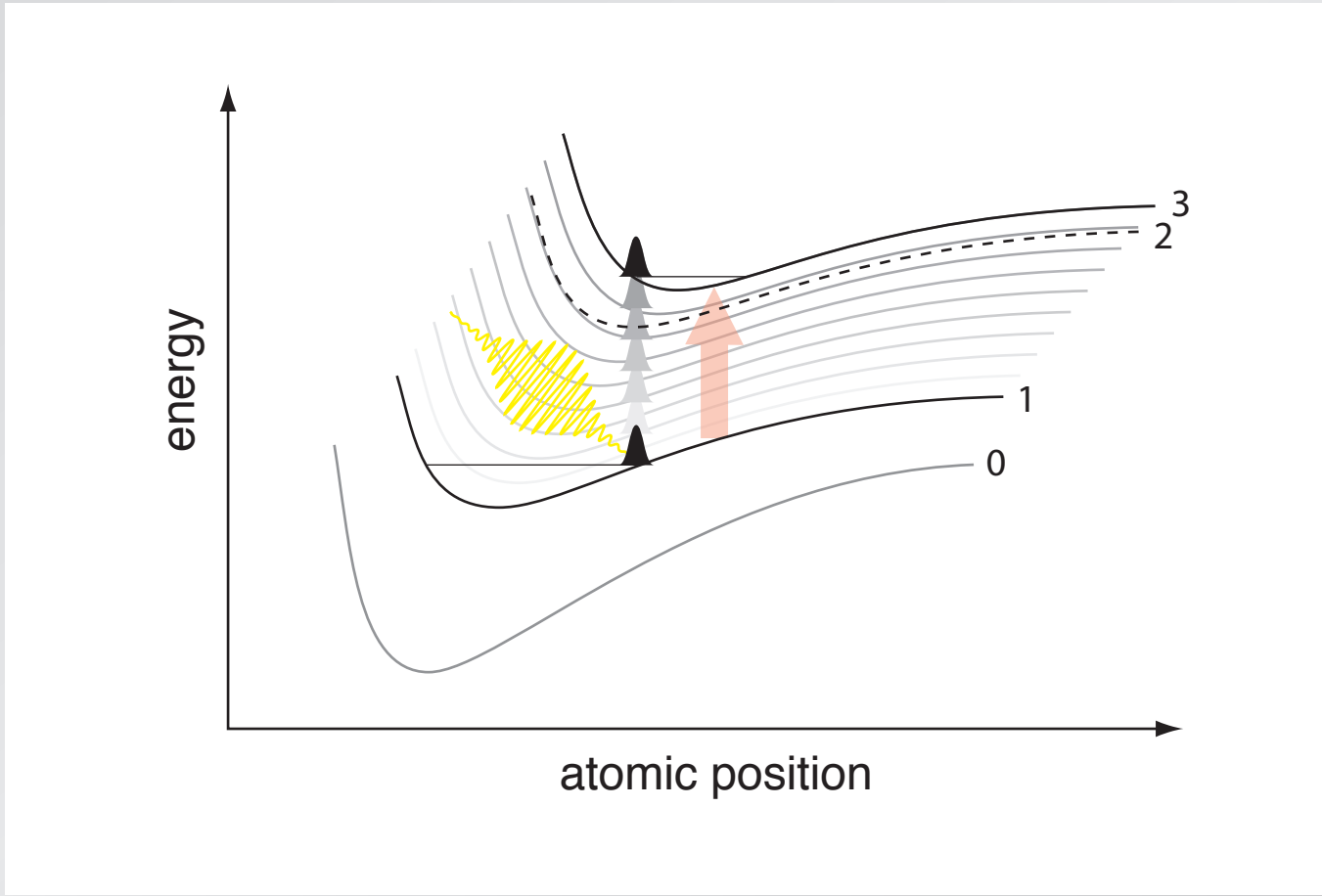
Optical control

…we get oscillations on the new potential



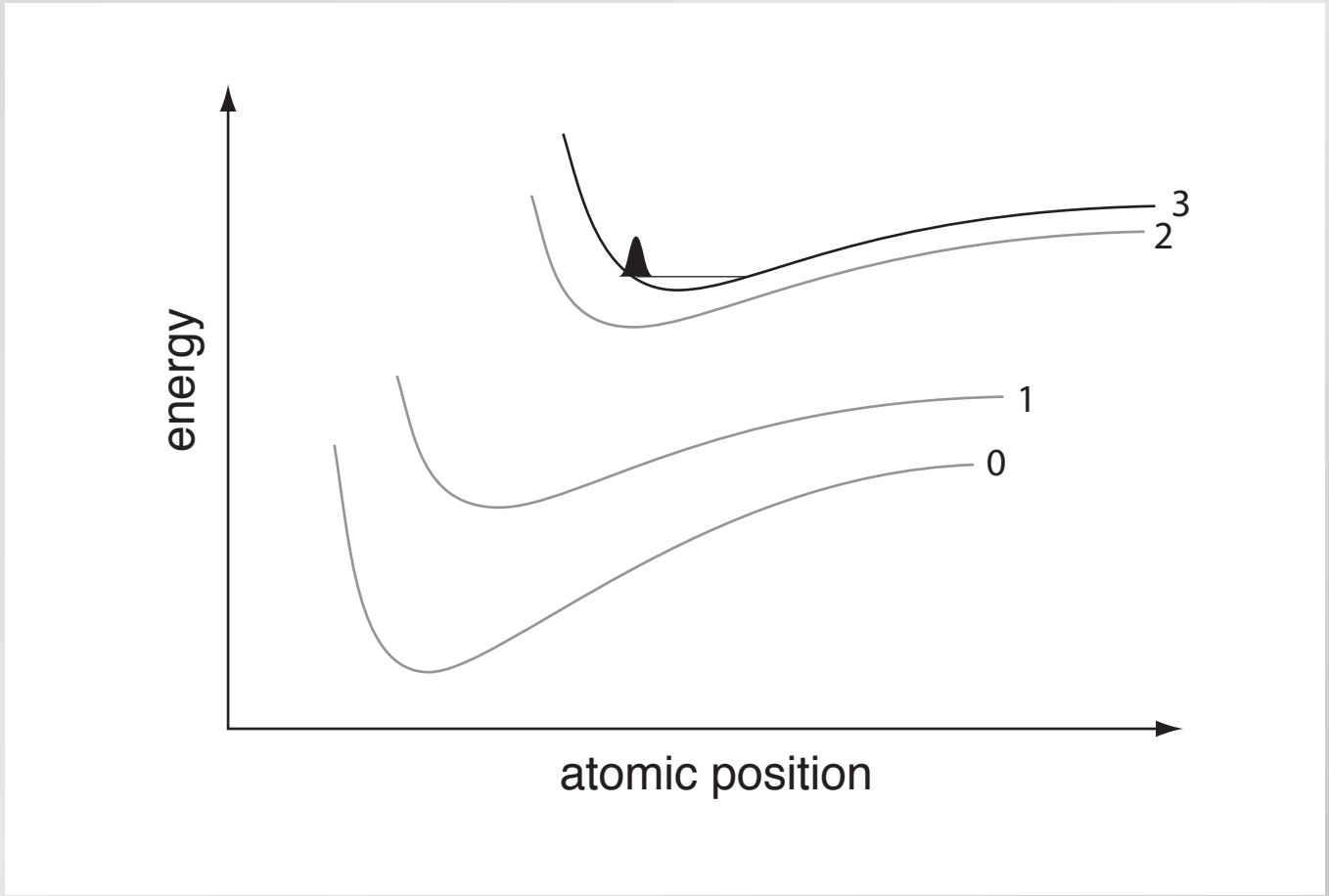
Optical control

if fluence wrong…



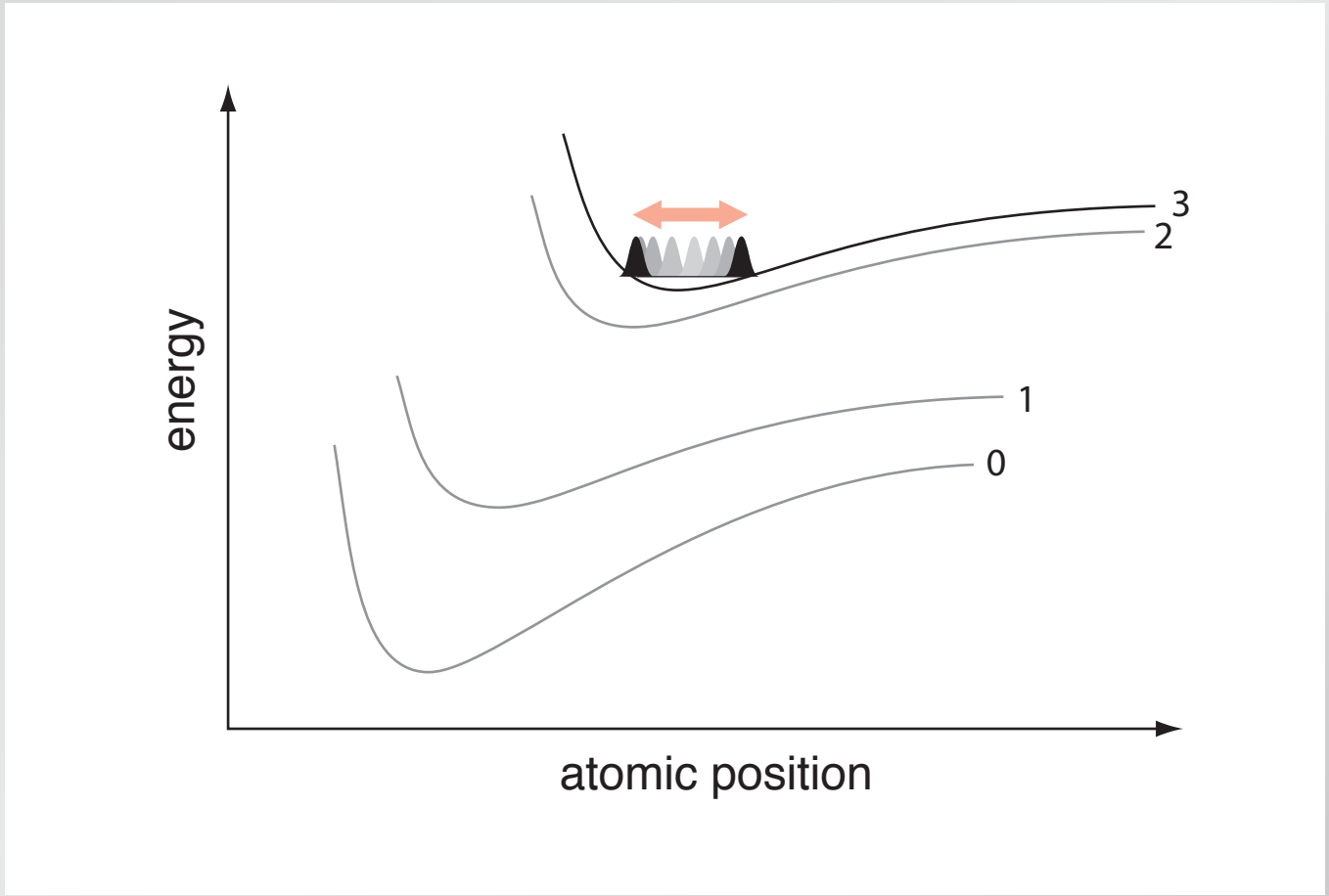
Optical control

excite to other potential surface…



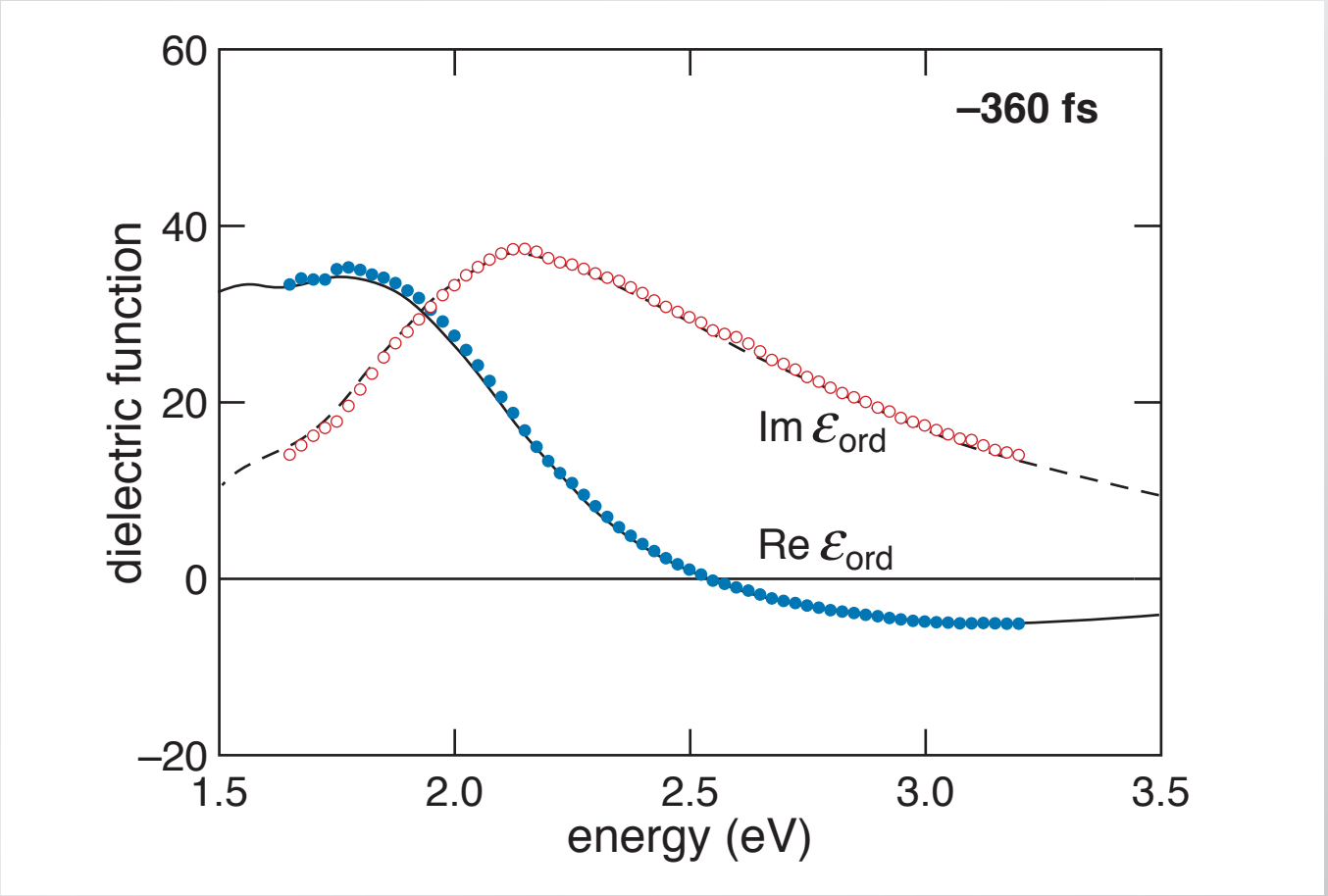
Optical control

…and wave packet oscillates



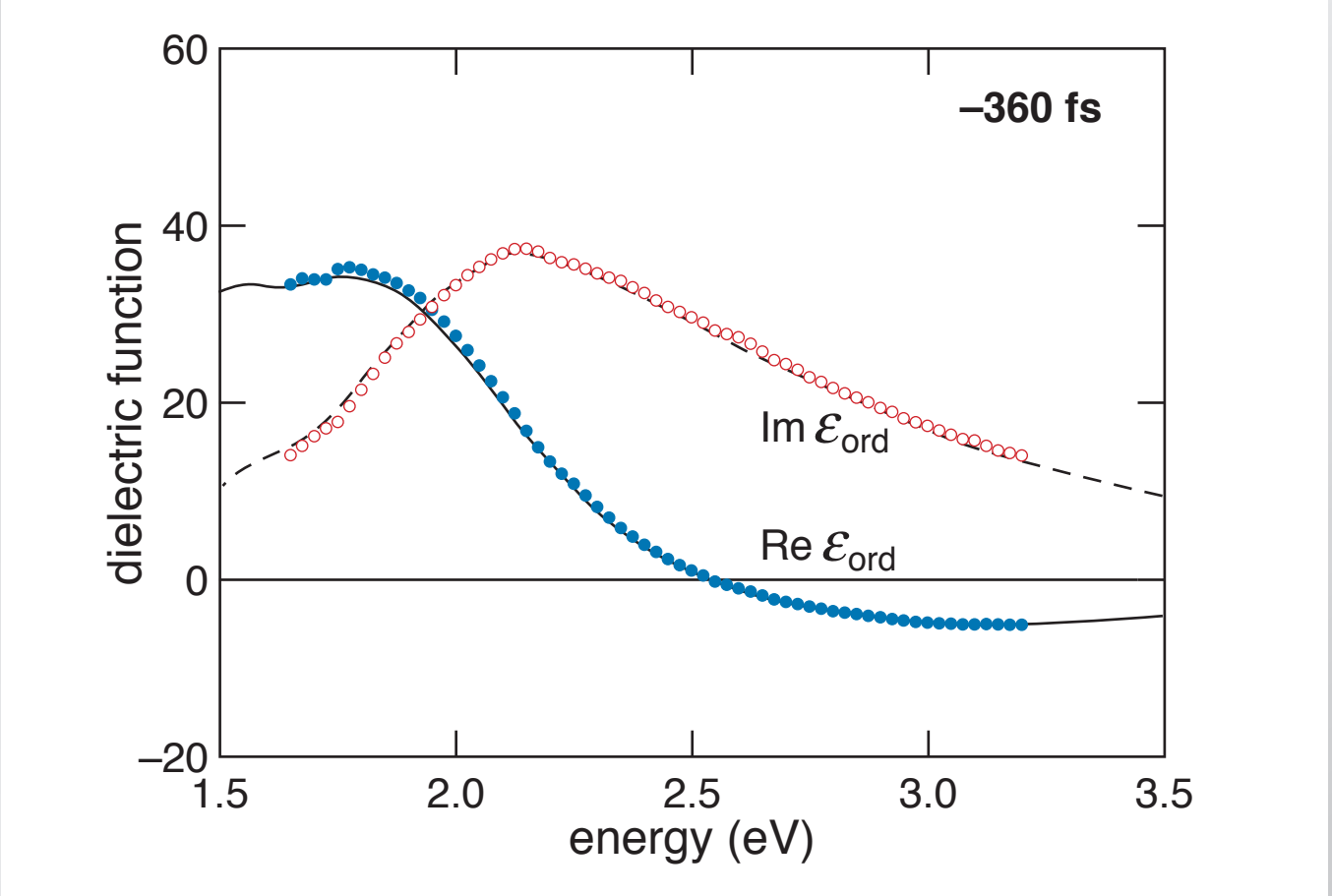
Optical control

$F_1 = 0.71\ F_{th}$ $F_2 = 0.43\ F_{th}$ $\tau = 467$ fs



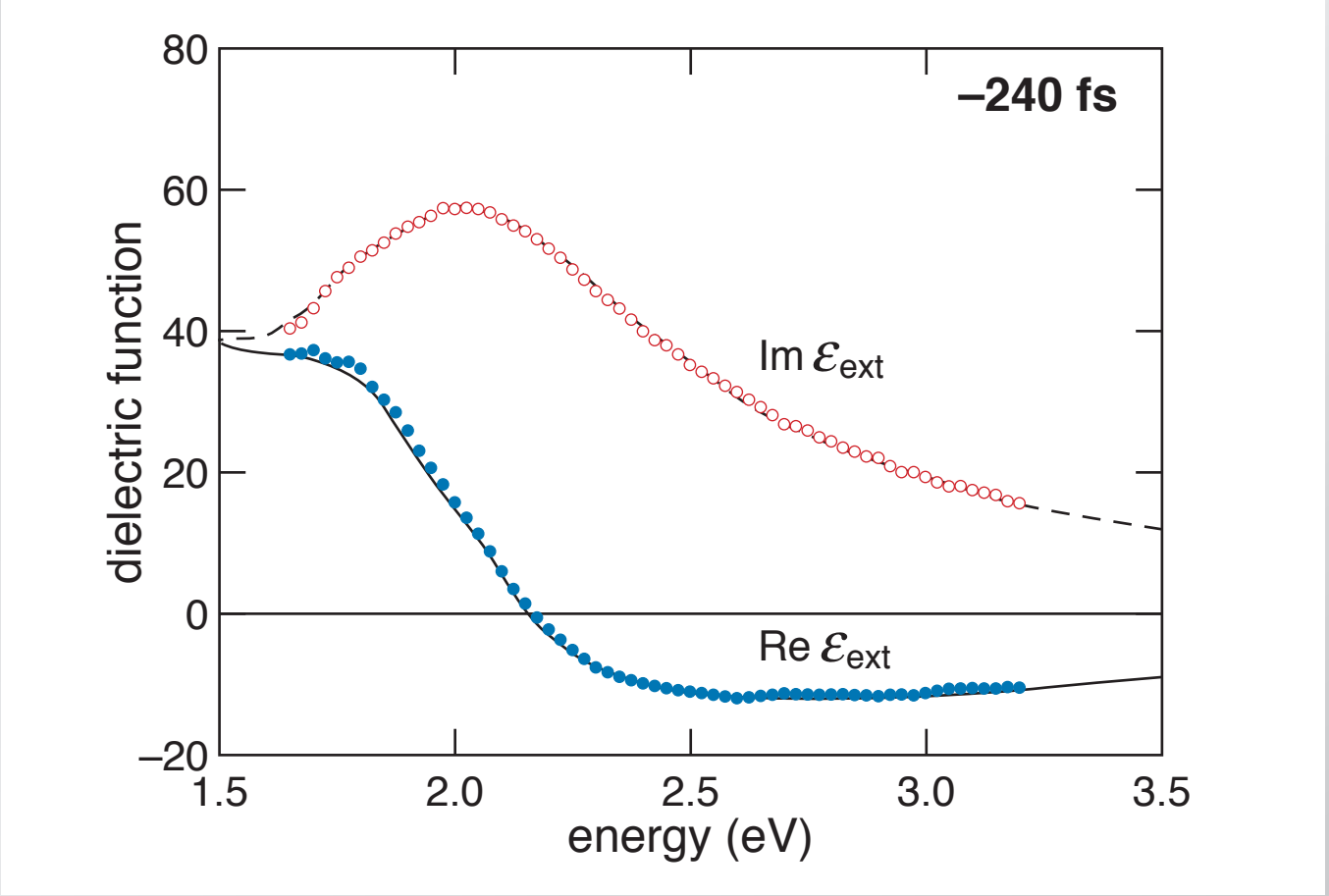
Optical control

$F_1 = 0.43\ F_{th}$ $F_2 = 0.35\ F_{th}$ $\tau = 127$ fs



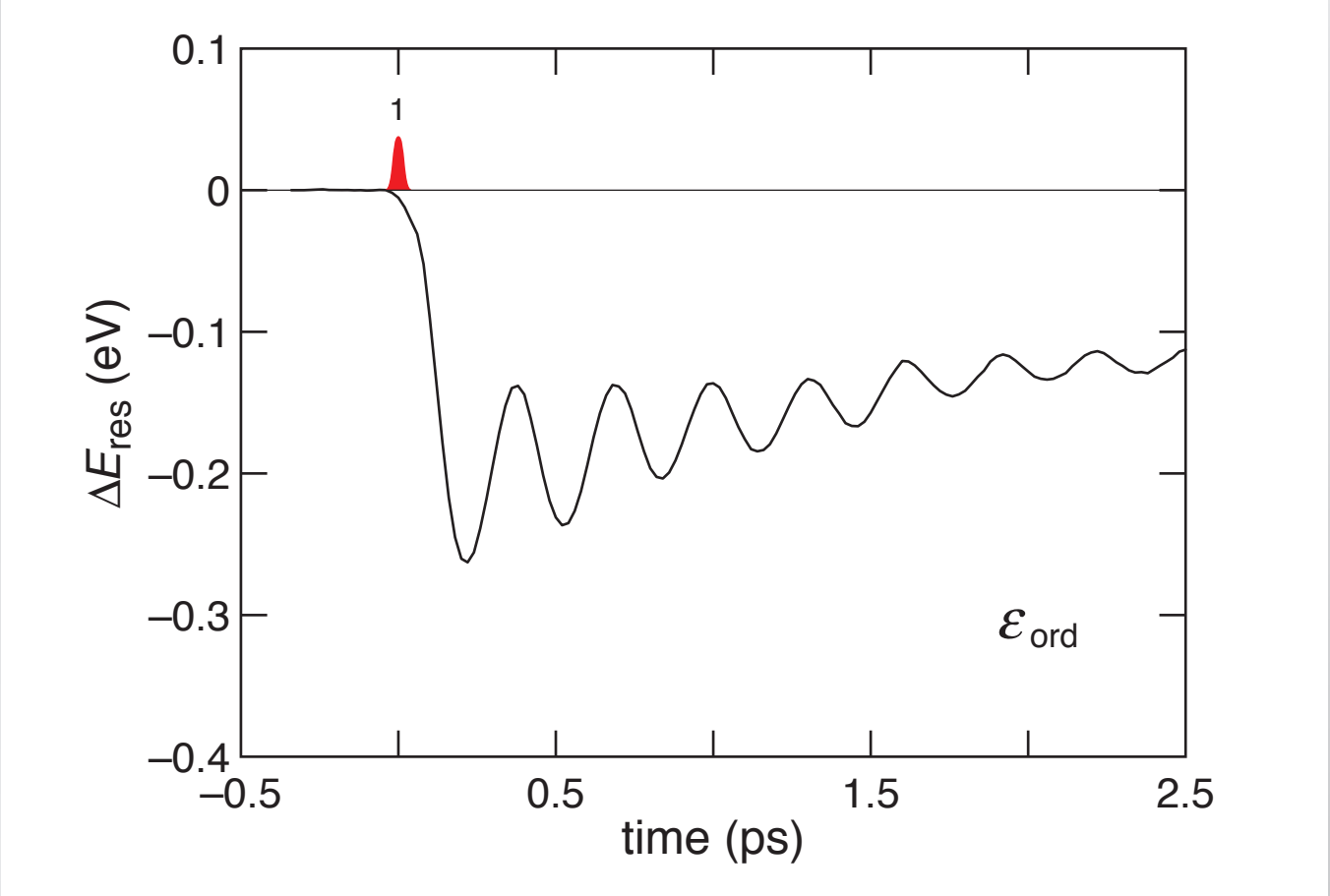
Optical control

$F_1 = 0.43\ F_{th}$ $F_2 = 0.33\ F_{th}$ $\tau = 127$ fs



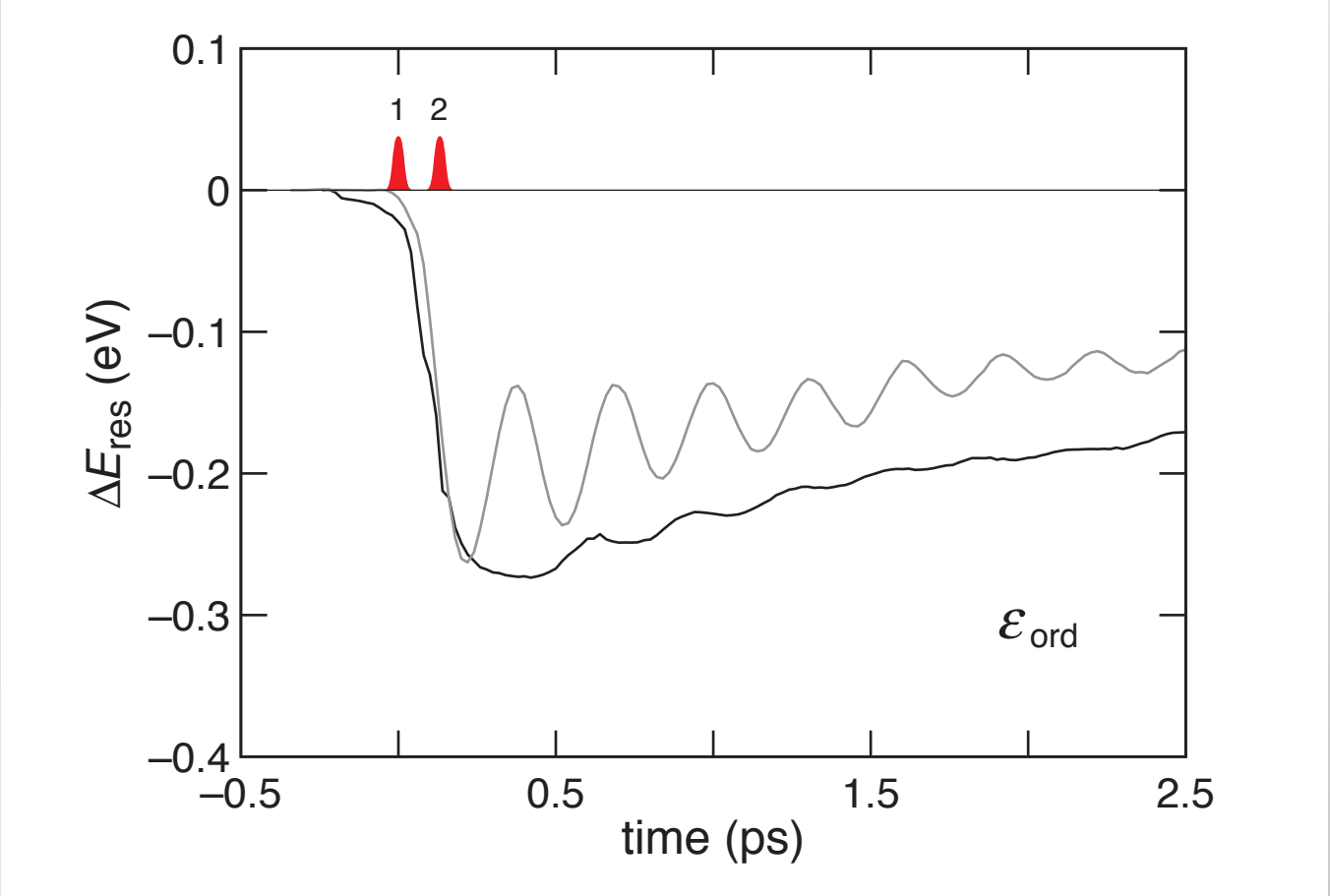
Optical control

$F_1 = 0.57\ F_{th}$



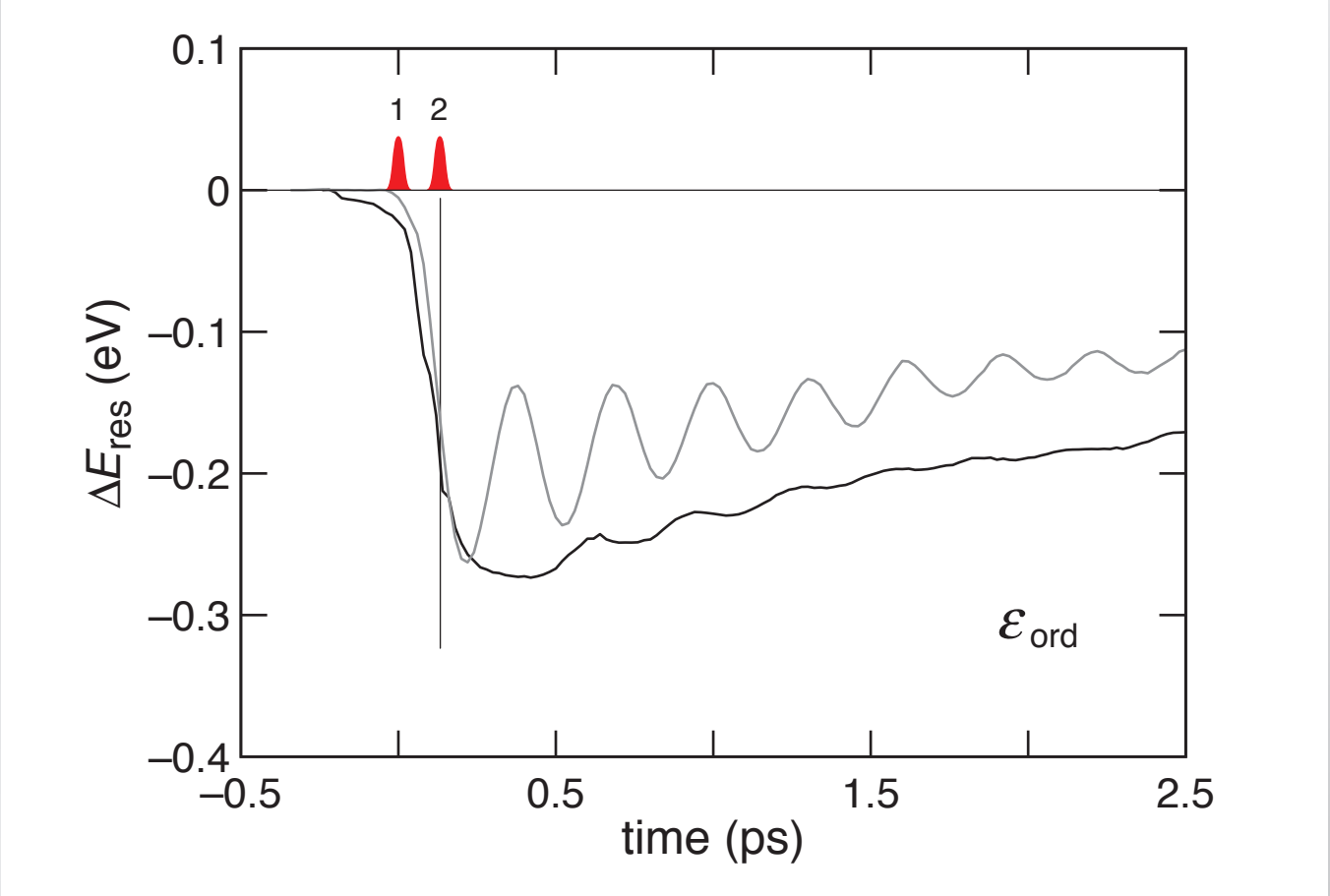
Optical control

$F_1 = 0.57\ F_{th}$ $F_2 = 0.46\ F_{th}$ $\tau = 133$ fs



Optical control

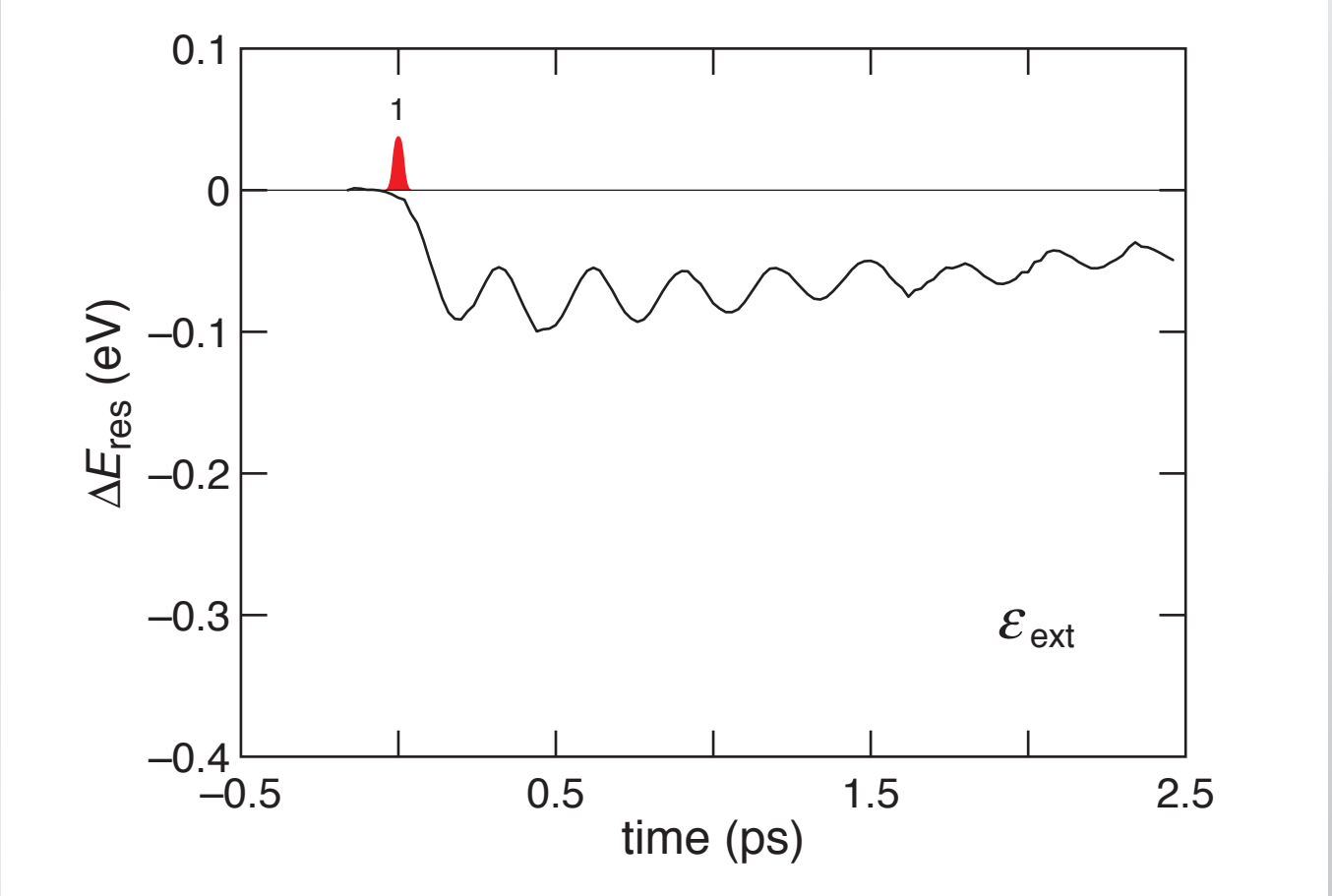
$F_1 = 0.57\ F_{th}$ $F_2 = 0.46\ F_{th}$ $\tau = 133$ fs



…but delay a bit less than half a period

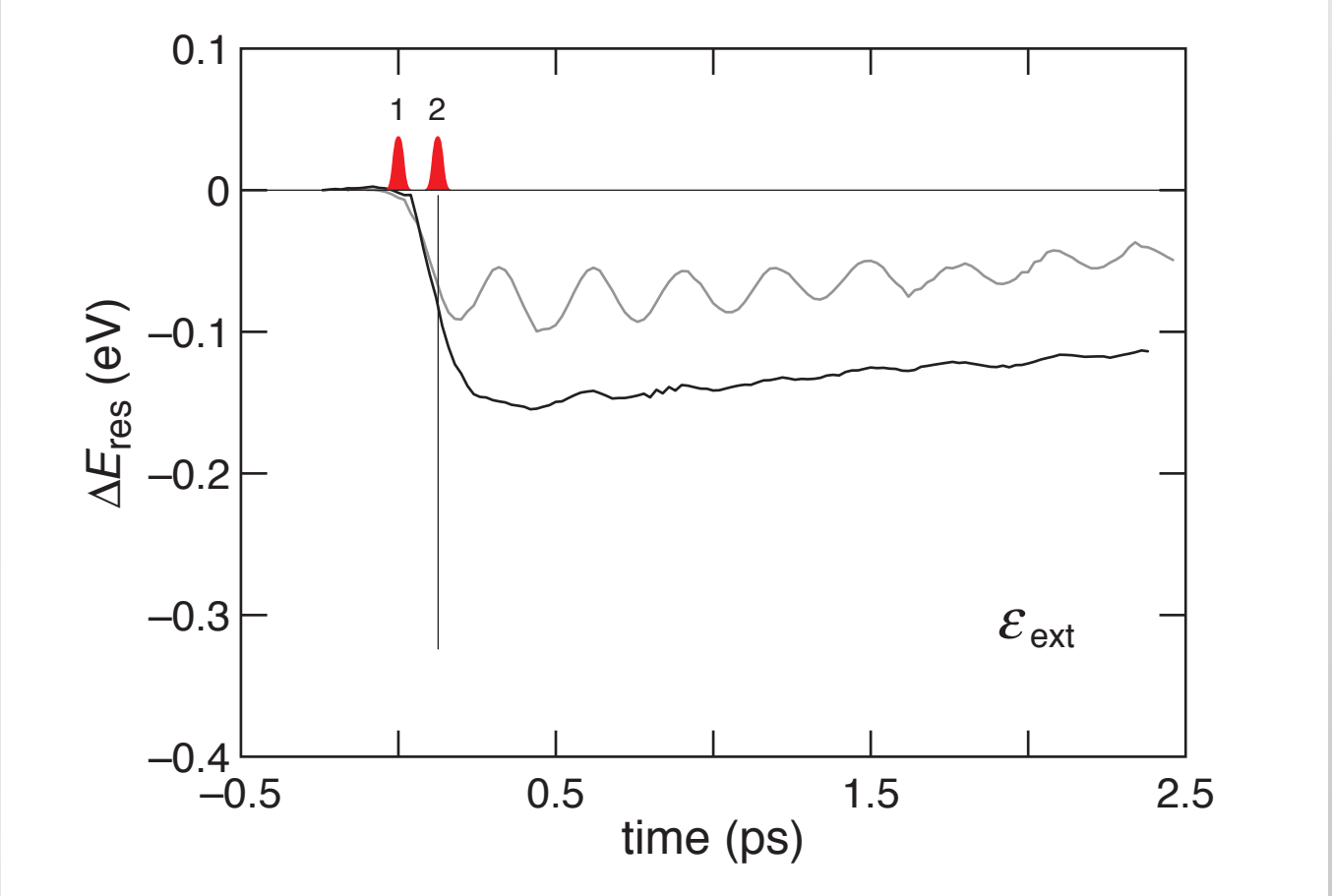
Optical control

$F_1 = 0.43\ F_{th}$



Optical control

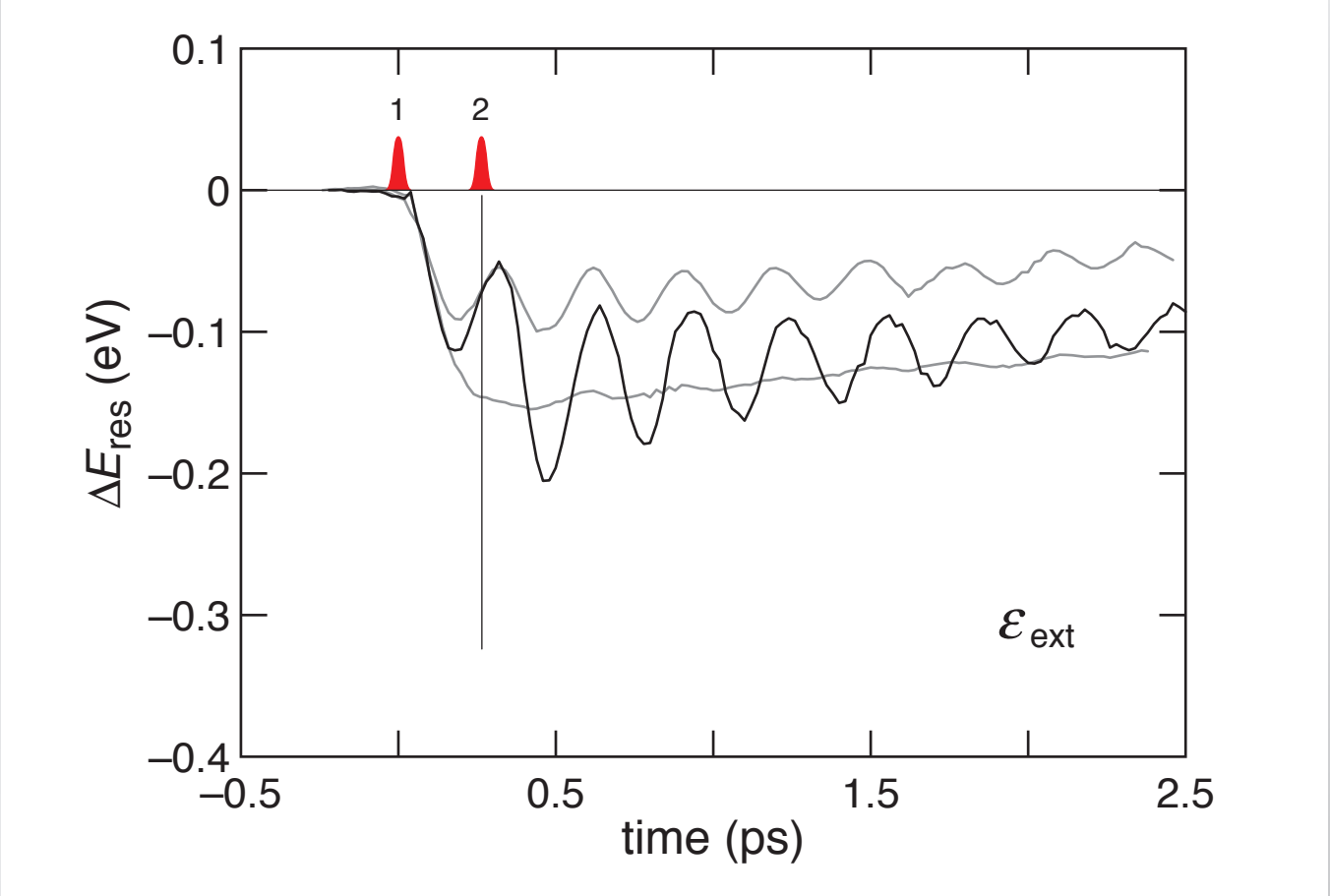
$F_1 = 0.43\ F_{th}$ $F_2 = 0.33\ F_{th}$ $\tau = 127$ fs



delay again less than half a period

Optical control

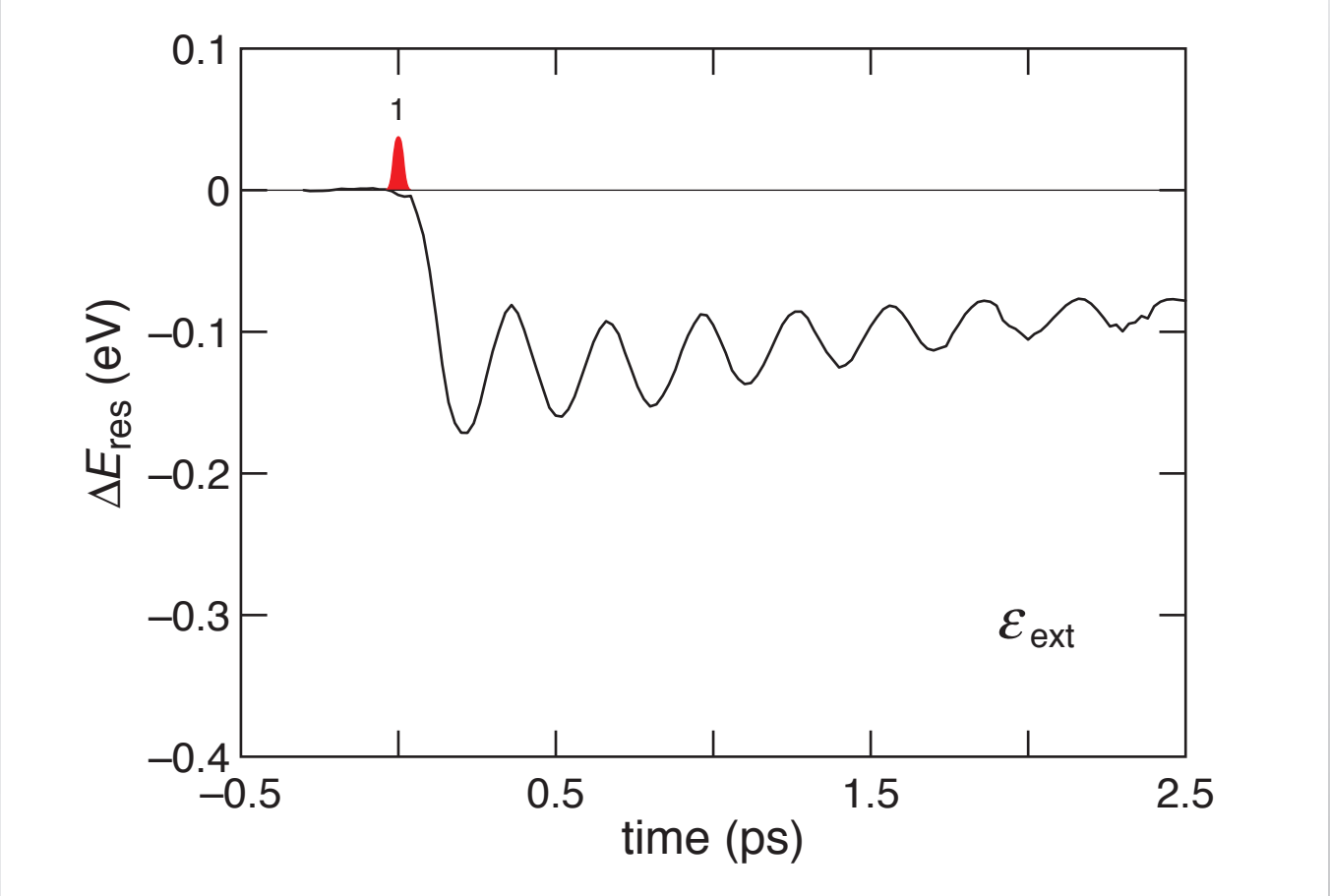
$F_1 = 0.43\ F_{th}$ $F_2 = 0.33\ F_{th}$ $\tau = 267$ fs



delay a bit less than a period

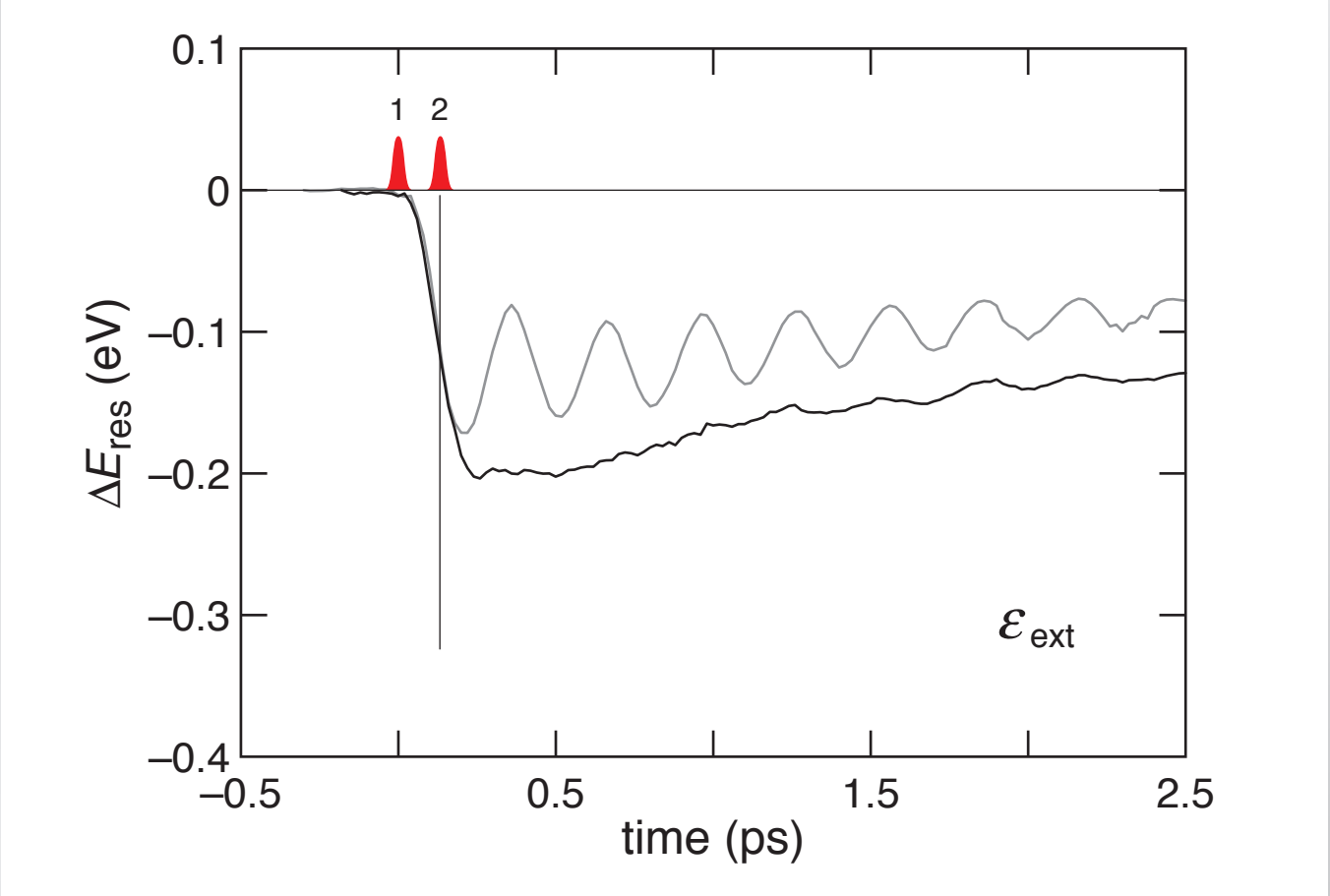
Optical control

$F_1 = 0.57\ F_{th}$



Optical control

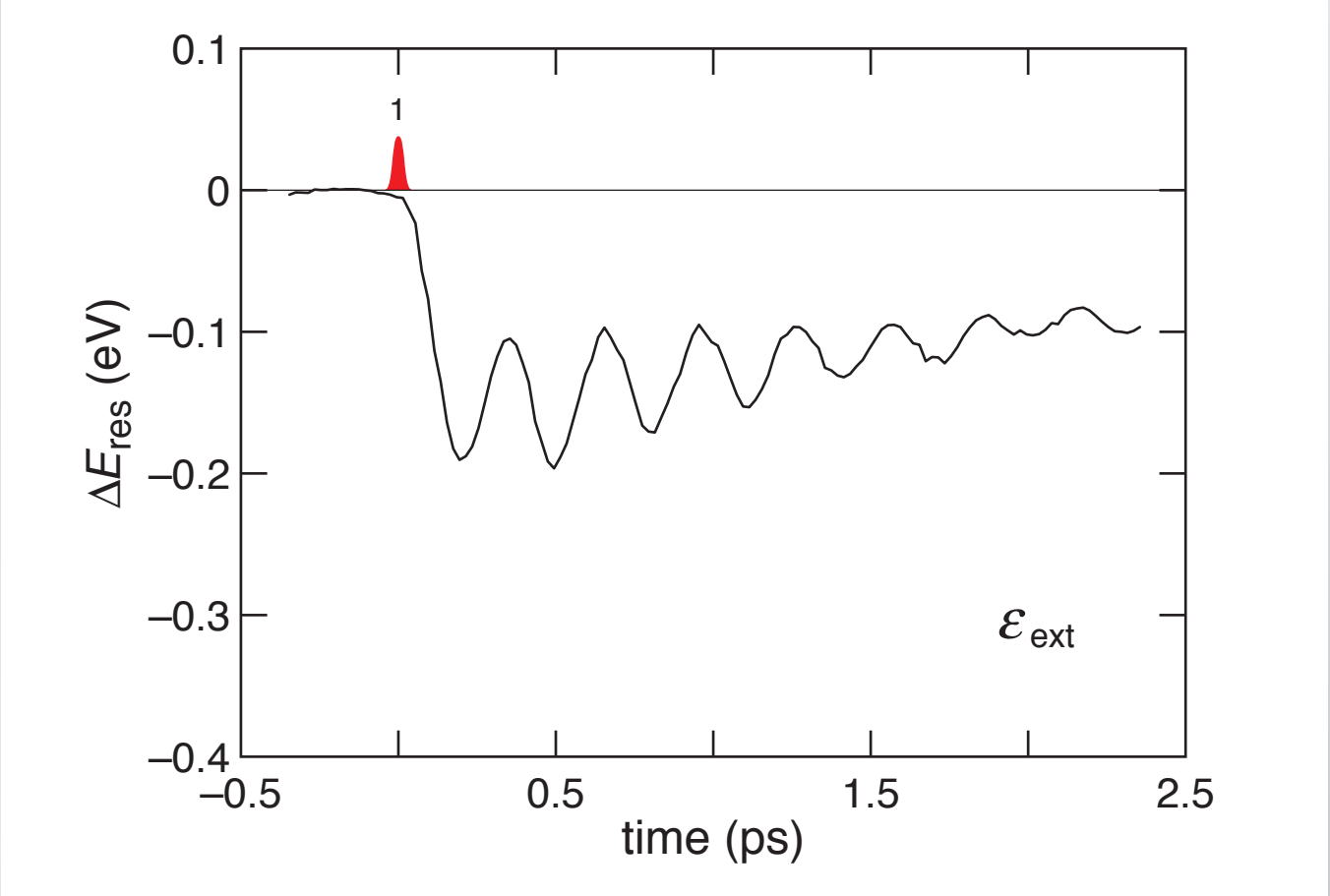
$F_1 = 0.57\ F_{th}$ $F_2 = 0.45\ F_{th}$ $\tau = 133$ fs



cancellation on first swing

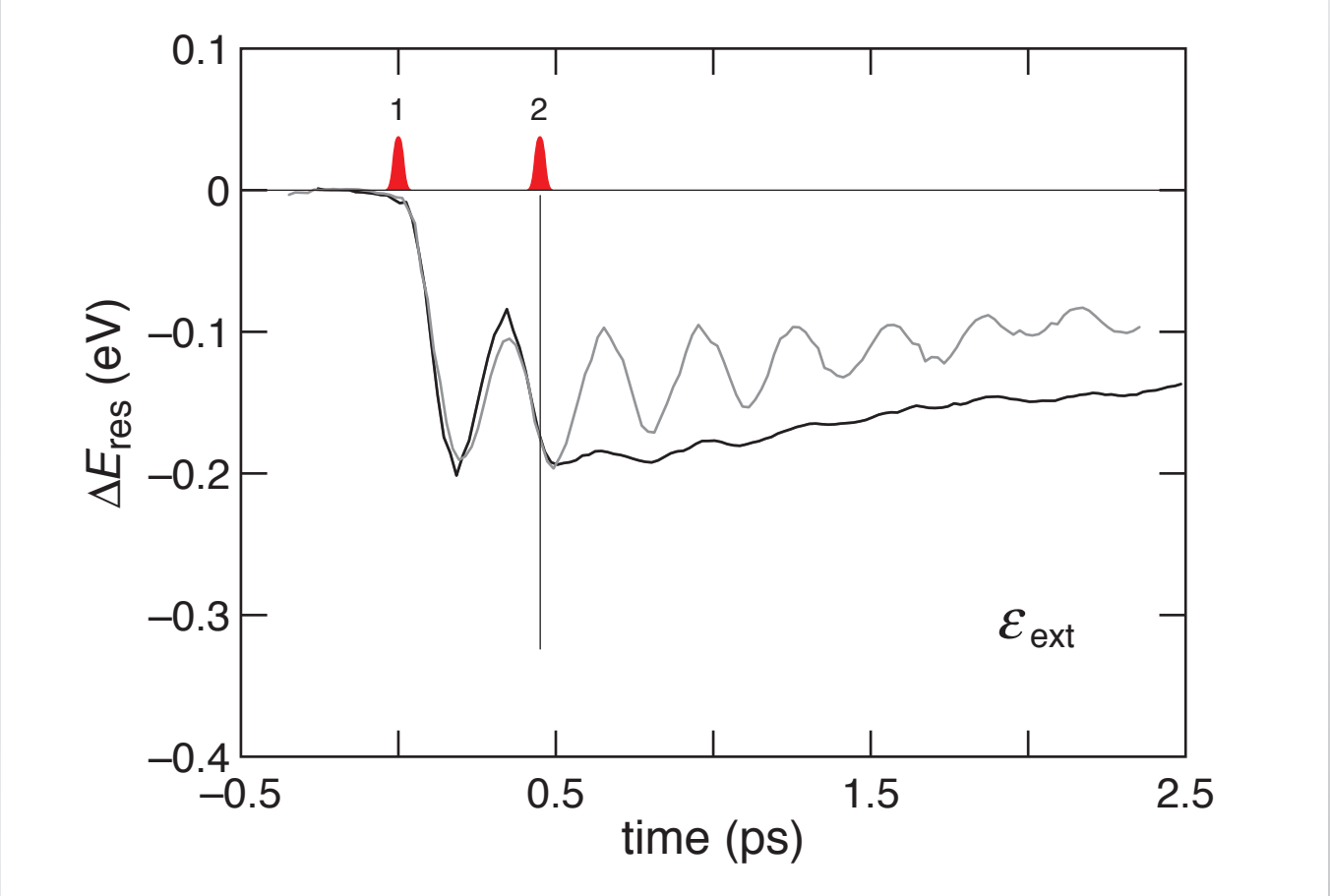
Optical control

$F_1 = 0.71\ F_{th}$



Optical control

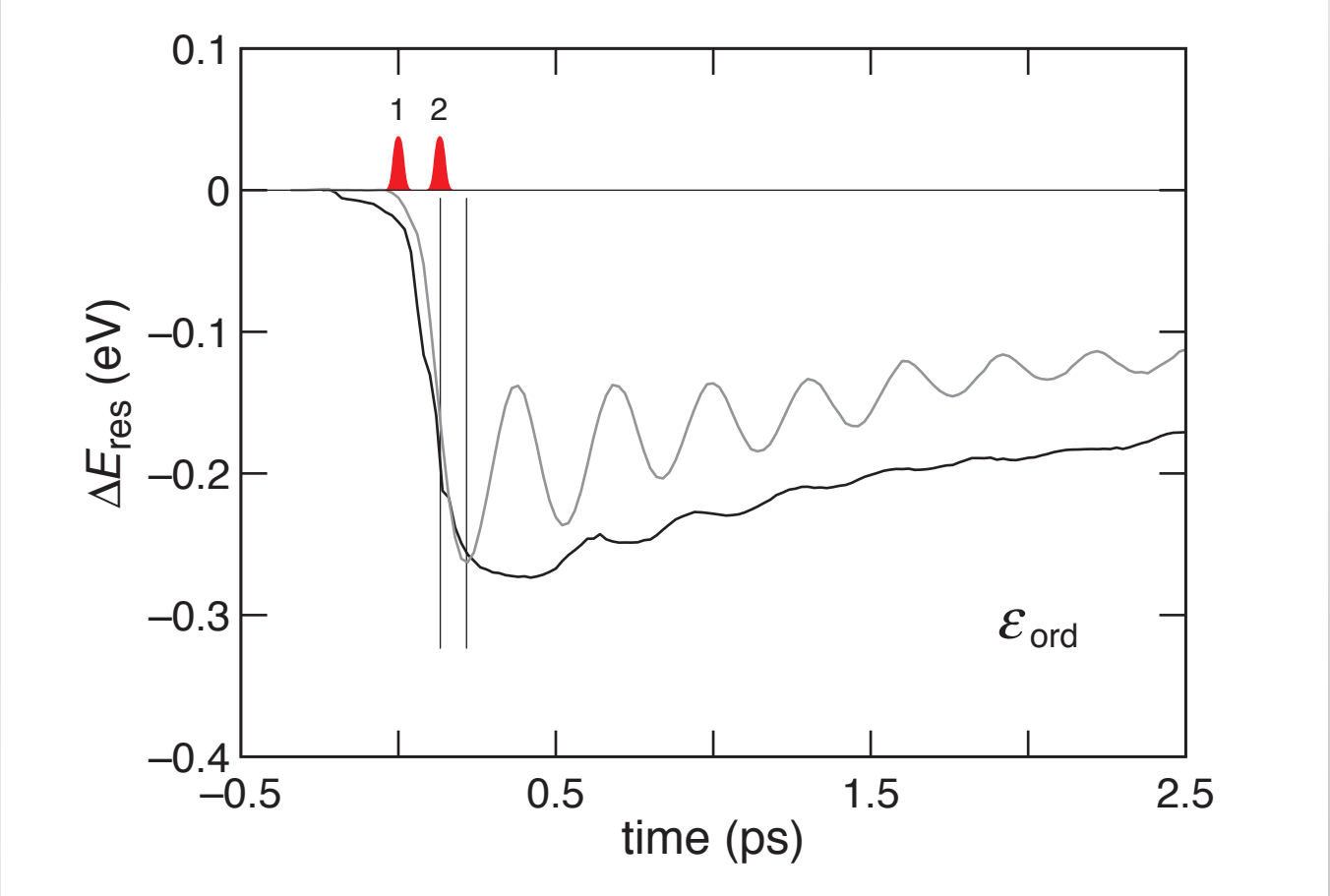
$F_1 = 0.71\ F_{th}$ $F_2 = 0.34\ F_{th}$ $\tau = 467$ fs



cancellation on second swing

Optical control

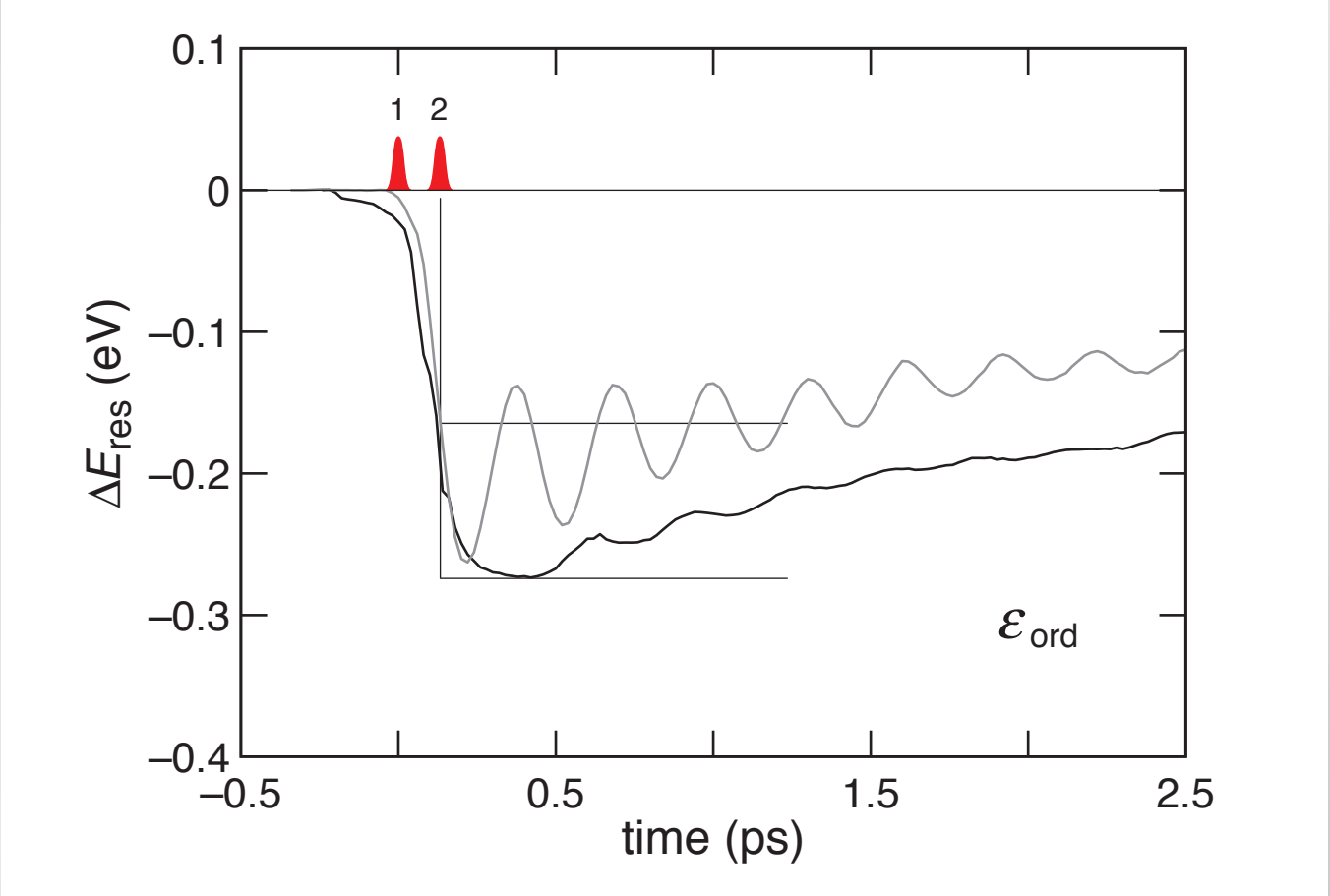
two things to note:



second pulse always earlier than expected, and…

Optical control

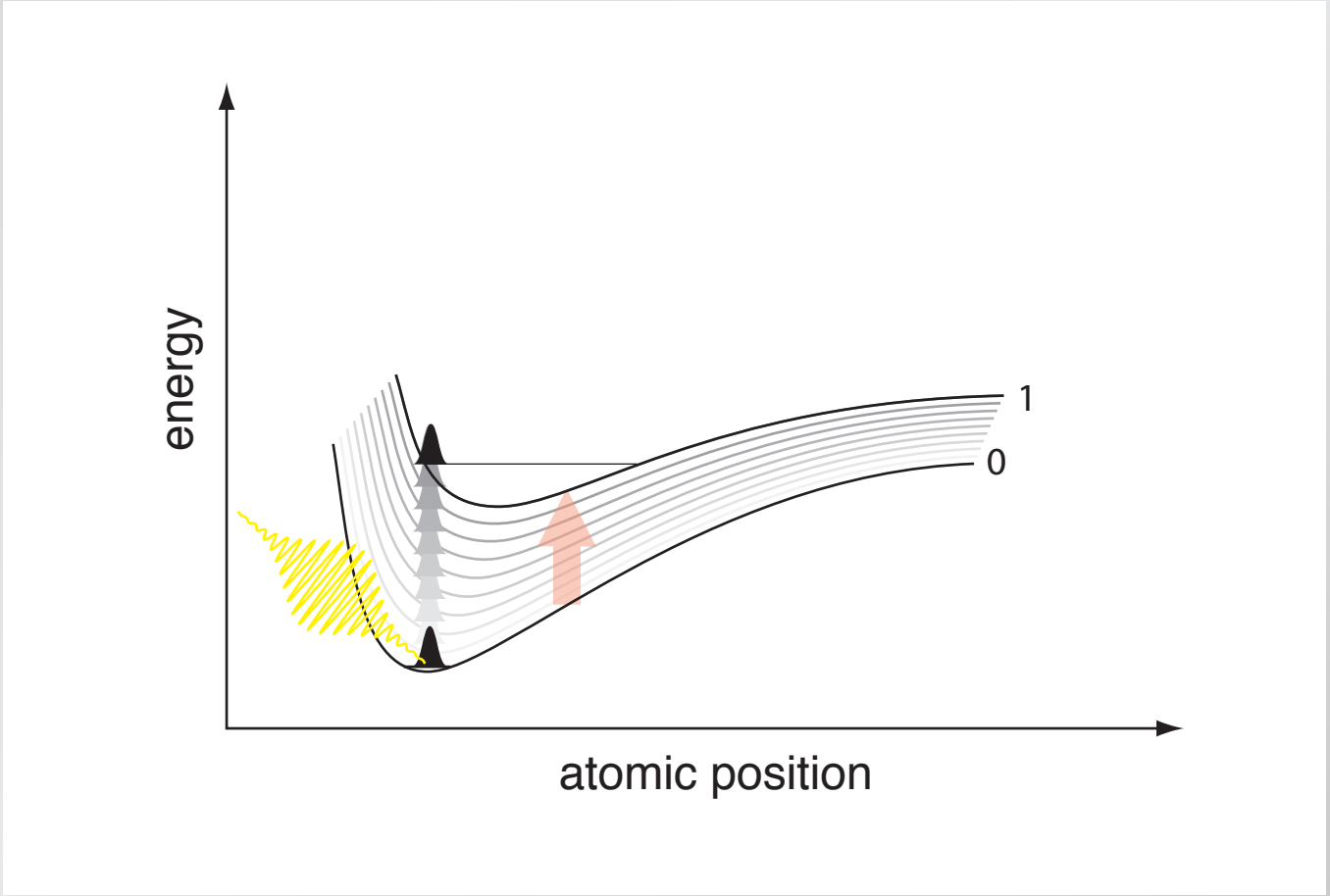
two things to note:



… resonance continues to shift after second pulse

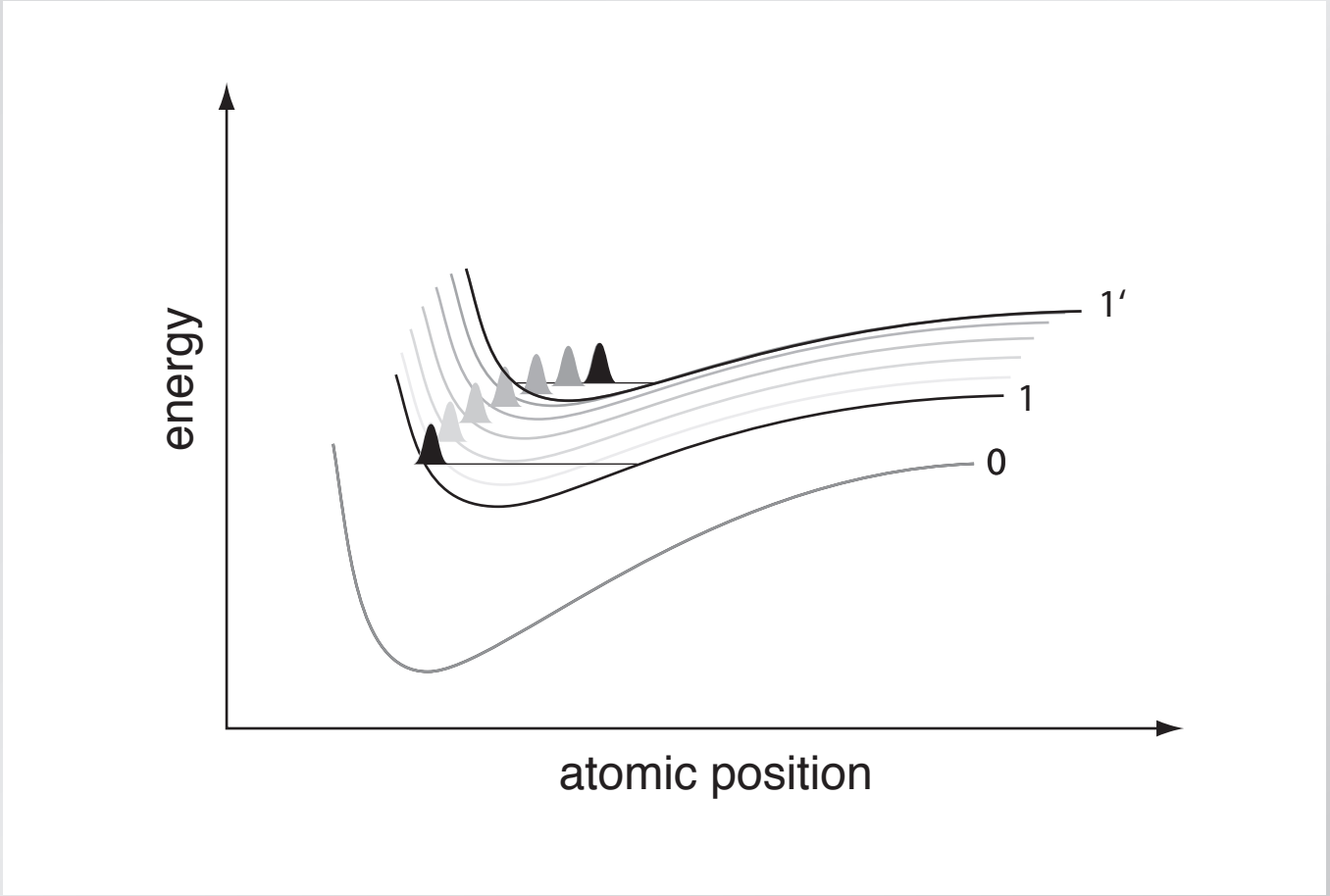
Optical control

excited electrons ‘instantaneously’ alter potential



Optical control

but nuclear rearrangement also alters potential



Summary

- can observe dynamics of ultrafast phase transition
- excitation of large-amplitude coherent phonons
- phonons can be controlled optically
- electronic and nuclear configurations affect dynamics

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Funding:

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